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Ac-susceptibility and neutron diffraction studies of the $URu_{1-x}Pd_xGa$ system

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Abstract

Ac-magnetic susceptibility and neutron diffraction studies are reported for the solid solutions $URu_{1-x}Pd_xGa$. Results of both experiments confirm the nonmagnetic-magnetic crossover in this investigated system for $x \approx 0.1$, detected previously by magnetization and electrical resistivity measurements. With increasing Pd-composition three different magnetic states are observed; the ferromagnetic state is stable in a wide concentration range ($0.1 \leq x < 0.8$), the ferrimagnetic ordering is found for $x \approx 0.8$ and finally the antiferromagnetic spin-arrangement of sine-modulated type for $x = 0.9$ and 1.0. As a result of these measurements, the magnetic phase diagram, x-T, in zero-field has been constructed, which is compared with that determined previously at B = 0.5 T.

Keywords: $URu_{1-x}Pd_xGa$; Magnetic phase diagram; Ac-susceptibility; Neutron powder diffraction

1. Introduction

It is generally accepted that the magnetic behaviour of the equiatomic ternary uranium compounds (U)-transition metal (T)-metalloid element (M) correlates strongly with the degree of the hybridization between the U5f- and Td-electrons [1]. In order to obtain a better understanding of the influence of this effect on the physical properties of the UTM series, one of the experimental methods, for example, is a systematic study of the solid solutions of the type $UT_{1-x}T'_xM$, where T and T′ differs with the number of nd-electrons [2–4]. Among others, for such a study we have chosen the $URu_{1-x}Pd_xGa$ system, because in this system, one proceeds through a transition from the non-magnetic phase (URuGa) and finally to an antiferromagnetic phase (UPdGa) [5,6]. Moreover, a continuous change in the conduction electron concentration in this system and a change in the atomic volume may help one to understand the role of the 5f-nd hybridization and its systematic decay with increasing Pd-content.

Previously, we have studied the crystallographic, magnetic [7] and transport properties [8] of these solid solutions $URu_{1-x}Pd_xGa$ with $0 \leq x \leq 1.0$. On the basis of magnetization measurement results, the magnetic phase diagram of the system has been constructed [7], where an evolution of the magnetic ground state, from an itinerant 5f-electron paramagnetic (URuGa), through a ferromagnetic ($x = 0.2$–0.7) or ferrimagnetic ($x \approx 0.8$), and finally to antiferromagnetic one ($x = 0.9$ and UPdGa) takes place. For the ferromagnetic alloys, the Curie temperature, T_C, increases with increasing Pd-content and goes through a maximum near $x = 0.5$. We have explained the results obtained in terms of a Kondo-lattice model.

In order to examine the magnetic behaviour of the above system in zero-field we have extended our studies to neutron powder diffraction and ac-magnetic susceptibility measurements. In this contribution we have focused mainly on the ac-susceptibility investigations, because more detailed neutron diffraction data will be published elsewhere [9]. Finally, we present a magnetic phase diagram deduced from the results of ac-magnetic susceptibility and neutron diffraction measurements.

2. Experimental details

The preparation of the $URu_{1-x}Pd_xGa$ samples was described previously [7]. The quality of the samples was assessed by means of powder X-ray diffraction at room temperature, and for several samples by neutron powder diffraction investigation. It has been reported that all the

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investigated compositions crystallize in the crystal structure of the hexagonal HoNiAl-type with space group P62m (i.e. the ordered version of Fe_2P-type). There is no evidence for the presence of any impurity in the samples with compositions $x<0.8$, but a trace of a second phase of an orthorhombic structure was detected in samples with $x\geq 0.8$.

Ac-susceptibility measurements were carried out with a LAKE SHORE susceptometer (series 7000) between 4.5 and 100 K. The applied ac-field was of the typical value of 10 Oe and frequency of 110 Hz. Neutron diffraction measurements were done with the G4.1 and 3T1 diffractometers installed at Léon Brillouin Laboratory, CEN-Saclay (France). The experimental neutron diffraction data were analyzed by Rietveld profile fitting method using the FULLPROF program [10]. Neutron scattering lengths were taken from [11] and U^{3+} magnetic form factor from [12].

3. Results

Fig. 1a displays the temperature dependence of the ac-susceptibility (χ_{ac}) for the $x=0$ and $x=0.1$ samples. For URuGa ($x=0$), the susceptibility increases with decreasing temperature and levels off at about 6 K. Such behaviour is in agreement with that of previous dc-magnetization measurements [13], where an upturn in the $\sigma(T)$-dependence was observed below 7 K. This phenomenon is due to a very weak ferromagnetism of the sample, evidenced by a small value of the spontaneous magnetization [7,13]. It should be noted that the magnetic behaviour of URuGa alters with a small deviation from stoichiometry of the compound. For example, the Ru-deficient sample, $URu_{0.9}Ga$, shows identical susceptibility behaviour to that of stoichiometric, URuGa, but the Ga-deficient sample, $URuGa_{0.9}$, exhibits a smooth $\chi_{ac}(T)$-dependence down to 4.5 K, typical of a paramagnet. Thus, the susceptibility behaviour of our $URuGa_{0.9}$ sample is rather similar to that reported by Sechovsky et al. [5], on URuGa. These authors reported that their URuGa showed no anomaly in ac-susceptibility down to 1.2 K and the magnetization of this material at 4.2 K was linear in magnetic fields up to 40 T.

Furthermore, upon the substitution of 10% of the ruthenium atoms with the palladium atoms in URuGa, a distinct maximum in $\chi_{ac}(T)$ at 11 K appears. This is indicative of a development of some ferromagnetic correlation between uranium atoms in this composition. With further increasing Pd-content the temperature of χ_{ac}-maximum increases and for the $x=0.2$ sample it reaches 43 K (Fig. 1b). Simultaneously, the measured ac-susceptibility reaches as high a value of 12×10^{-3} emu/g, indicating the presence of some ferromagnetic order in the sample. The neutron diffraction patterns (NDP) of this alloy taken below 40 K show only additional contributions to all the nuclear reflections, except for (00l) ones. Therefore, the neutron diffraction study determines unambiguously the ferromagnetic ordering of the uranium sublattice for this composition. The refinement of magnetic peaks indicates that the magnetic structure of the $x=0.2$ composition consists of the ferromagnetic (001)-planes with the uranium magnetic moments parallel to the c-axis. The ordered moment value in this composition is rather small and at 1.4 K it amounts to about 0.40(5) μ_B/U.

Previously, we have shown that the magnetization [7] and electrical resistivity [8] measurements taken for the $x=0.3$ sample reveals only a single anomaly at $T_C=65$ K. However, the ac-susceptibility behaviour of this sample seems to be more complicated, owing to two observed maxima in both the real and imaginary part of $\chi_{ac}(T)$ (Fig. 1b). The appearance of more than one maximum in the $\chi_{ac}(T)$-curve may indicate two transitions of the magnetic origin in this sample. We label them T_C and T_{C1}. These maxima in the $\chi'_{ac}(T)$-curves, are marked in Fig. 1b by the arrows. For the $x=0.4$ sample the real ac-susceptibility peak becomes quite broad but a double structure can be resolved in the imaginary part of the ac-susceptibility, at 72.2 and 77.8 K, as shown in Fig. 1b. In the case of $x=0.5$ (Fig. 1c) the real ac-susceptibility peak at $T_C=79$ K is also very broad and the two maxima seen in the imaginary part of the susceptibility reach the temperatures of 73.8 and 77.9 K. Any further increasing in Pd-content causes the peaks to become more and more broadened. Simultaneously, their positions are shifted to lower temperatures, and their heights are slightly reduced (except for the $x=0.8$ sample). For the $x=0.6$ sample we have observed a broad χ'_{ac}-peak at $T_C=74$ K and a shoulder at about 55 K. However, the neutron diffraction data of this composition exhibit no change in either the crystal or ferromagnetic structures between 50 and 70 K. A supplementary contribution to the Bragg peaks (100), (110), (011) and (200) appears in NDP collected below 70 K. The intensity of the magnetic Bragg peaks increases with decreasing temperature and below about 20 K, remains essentially constant. The refinement of the observed magnetic peaks reveals the magnetic structure of the same type as that found for $x=0.2$. The ordered magnetic U-moment in the $x=0.6$ sample is considerably higher, comparing to that in the $x=0.2$ sample and at 1.4 K it amounts to 1.13(2) μ_B/U.

For the $x=0.8$ sample, despite the broad phase transitions centred at about 55 and 61 K in the $\chi'_{ac}(T)$-curve, the neutron diffraction studies on this composition show no additional magnetic transitions between these temperatures. Just below 52 K, the NPD patterns do exhibit a number of additional magnetic peaks, which can be assigned to some antiferromagnetic (AF) component, as well as to some contribution to nuclear peaks corresponding to a ferromagnetic component. These two magnetic components belong to the hexagonal phase. No evidence of another phase has been detected. Thus, the magnetic structure of the $x=0.8$ sample is of ferrimagnetic type, characterized by magnetic propagation vectors $\boldsymbol{k}_1=(0, 0, 0)$ and $\boldsymbol{k}_2=(0, 0, 0.2)$. The absence of (00$l$)

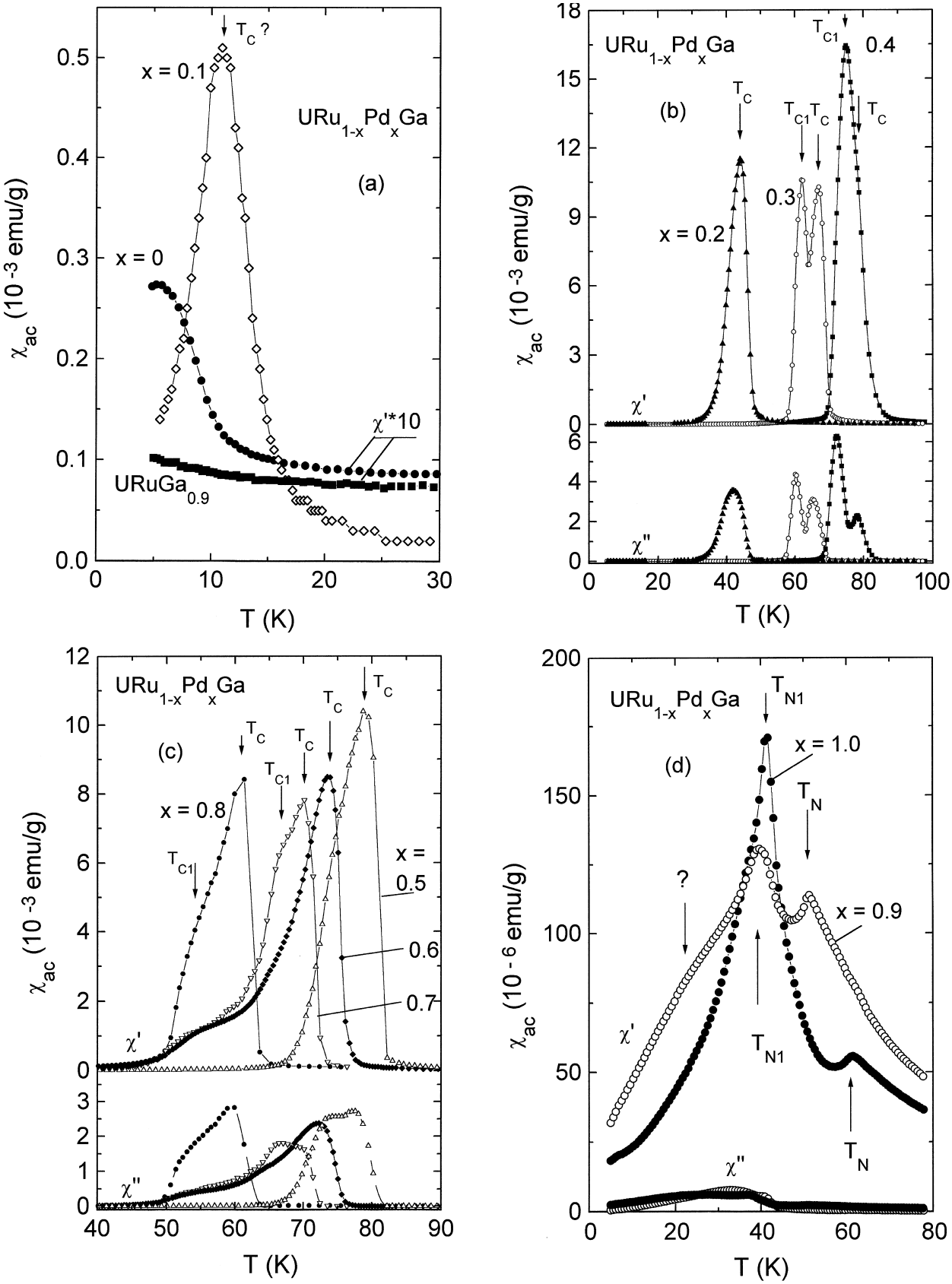

Fig. 1. Temperature dependence of the ac-magnetic susceptibility measured in an ac-magnetic field of 10 Oe with a frequency of 110 Hz for $URu_{1-x}Pd_xGa$ for the following concentrations: (a) $x=0$ and 0.1, (b) $0.2\leq x\leq 0.4$, (c) $0.5\leq x\leq 0.8$, and (d) 0.9 and 1.0.

reflections indicates that the alignment of magnetic moments is along the *c*-axis. Least-squares fitting of magnetic peak intensities reveals that the contribution of the antiferromagnetic component increases considerably at low temperatures. For example, at 1.4 K the antiferro- and ferromagnetic components amount to $\mu_{AF}=1.25(5)\ \mu_B$ and $\mu_F=0.85(5)\ \mu_B$, respectively. Near 30 K these two components are almost equal to each other and vanish at the same temperature of about 52 K.

Clear evidence of the existence of two phase transitions at T_N and T_{N1} is shown in Fig. 1d for $URu_{0.1}Pd_{0.9}Ga$ and UPdGa. In this figure, one can see that the position of the peak at T_N for each of the samples is different but that at T_{N1} remains approximately at the same temperature. This suggests that the observed phase transition at T_{N1} may arise from the same origin in these samples. Moreover, in the previous work [7], we have shown that the former transition was shifted down to lower temperature when the magnetic field was applied on the sample.

In the $x=0.9$ sample we also observe a shoulder in the $\chi'_{ac}(T)$-curve near $T=30$ K (see Fig. 1d). The analysis of the NPD patterns of this sample at 1.4 K has revealed extra magnetic peaks, which can be ascribed to two magnetic phases. One group of magnetic reflections corresponds to a sine-modulated structure of the hexagonal phase with $\boldsymbol{k}=(0, 0, 0.2)$, while the other group of magnetic peaks corresponds to the antiferromagnetic structure of the orthorhombic phase. The magnetic peaks belonging to the hexagonal phase appear below 50 K and those of the orthorhombic one are found below about 30 K. Therefore, it is quite probable that this orthorhombic phase is responsible for the low temperature χ'_{ac}-shoulder at 30 K, as shown in Fig. 1d. The magnetic structure of the orthorhombic phase is collinear with magnetic moments parallel to the *c*-axis. The details about the magnetic structure of this new orthorhombic phase will be given elsewhere [9]. We have estimated the ordered magnetic moment of the hexagonal phase at 1.4 K which is about 0.5 μ_B/U.

For UPdGa, the magnetic peaks appearing in the neutron diagram at 7 K correspond to an antiferromagnetic structure with incommensurate longitudinal modulation, characterized by a propagation vector $\boldsymbol{k}=(0, 0, 0.3)$ and $\mu_{ord} \| c$-axis. A least-squares fitting of magnetic peak intensities gives $\mu_{ord}=1.37(5)\ \mu_B$/U at this temperature. At 40 K the positions of the magnetic peaks are of the same type as those observed at 7 K, so that the magnetic structure of this compound does not change with temperature in this temperature region. Therefore, the magnetic behaviour of the $x \geq 0.9$ alloys between T_{N1} and T_N is still unclear and need further studies, which we plan to make in the near future.

4. Magnetic phase diagram and discussion

The experimental results presented here and those derived from the previous bulk measurements (dc-magnetic susceptibility, electrical resistivity) have allowed us to determine some details of the magnetic phase diagram for the solid solutions of the $URu_{1-x}Pd_xGa$ system (Fig. 2). In accordance with the previous studies it appears that the nonmagnetic-magnetic crossover occurs near the $x=0.1$ composition and the ferrimagnetic transition takes place in the $x=0.8$ sample. However, the part of the present magnetic phase diagram near the characteristic transition temperatures T_C and T_N is still in doubt, indicating that the origin of the magnetic behaviour in this region is not yet quite understood.

A convenient model to explain these transitions mentioned above may be some balance between the ferro- and antiferromagnetic components. This model seems to be plausible in the case of $x=0.8$, where these two components have been detected by neutron diffraction experiments. However, we have no such evidence for the remaining solid solutions studied. Another possible explanation could be that there is some magnetic ordering in the transition metal sublattice. Further neutron diffraction studies on a single crystal are needed to establish the magnetic nature of the involved transition metal atoms. In the magnetic structure refinements of NPD patterns obtained in this work we have assumed the non-magnetic properties of the Ru and Pd atoms.

Recently, Andreev et al. have investigated a similar

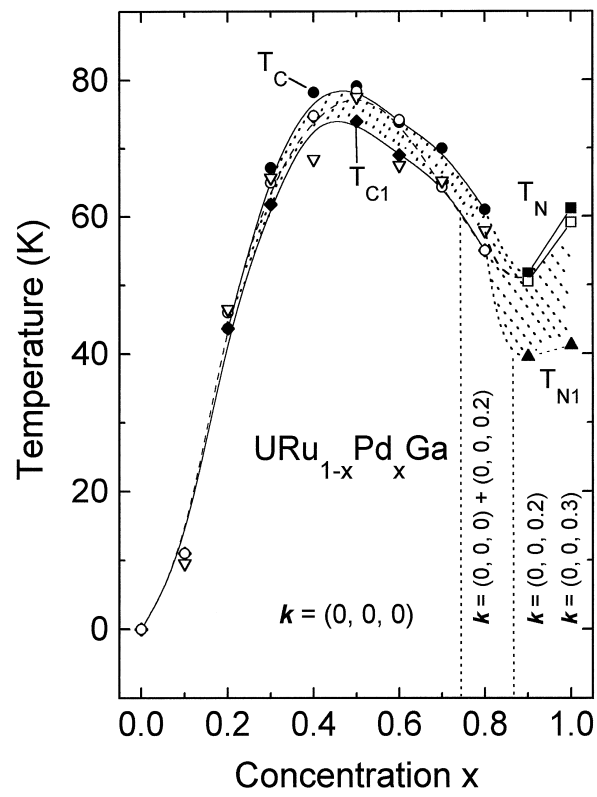

Fig. 2. Magnetic phase diagram of the $URu_{1-x}Pd_xGa$ system. The zero field characteristic temperatures T_C (●), T_{C1} (♦), T_N (■), T_{N1} (▲) are obtained from the ac-susceptibility and T_C (▽) from the electrical resistivity measurements [8], while T_C (○) or T_N (□) from magnetization measurements in a magnetic field of 0.5 T [7].

solid solution, i.e. $UCo_{1-x}Ru_xAl$ [4]. The authors have also observed the secondary phase transition in the ferromagnetic state of their system for $0.5<x<0.7$. This behaviour has been attributed by these authors either to an additional magnetic phase transition or to a sudden change in the magnetization process at a critical magnetic field, B_{cr} [4]. Moreover, they have excluded any important inhomogeneity in their samples.

We have determined magnetic structures for several compositions of the $URu_{1-x}Pd_xGa$ system. The magnetic structure of the ferromagnetic samples is collinear, where the uranium magnetic moments are ferromagnetically coupled within the (001)-plane and are aligned parallel to the hexagonal *c*-axis. For the antiferromagnetic compositions ($x\geq0.9$), the magnetic structure consists of the ferromagnetic (001)-planes, but with the magnetic moments being sine-modulated from plane to plane. The magnetic propagation vector changes from composition to composition, e.g. $\boldsymbol{k}=(0, 0, 0.2)$ for $x=0.9$ and $\boldsymbol{k}=(0, 0, 0.3)$ for UPdGa. It is worth noting that the $x=0.8$ composition is a borderline between the ferro- and antiferromagnetic states. Thus, the magnetic structure of this composition is intermediate and of a ferrimagnetic type. For all the magnetically ordered alloys, the value of the magnetic moment depends on the Pd-content in the sample. A maximum value of μ_{ord} is among studied ferromagnets for the $x=0.6$ composition, while a minimum is found near $x=0.9$. This dependence corresponds well to that of $T_{C/N}$ behaviour (Fig. 2) and the magnetisation study [7]. A reduced uranium magnetic moment observed for the $x=0.9$ alloy suggests the existence of the Kondo effect in this sample. Thus, the competition of this effect with the magnetic interactions of RKKY-type plays an important role in the magnetic behaviour of these solid solutions, as has been discussed previously [7].

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