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## MAGNETIC PROPERTIES AND HYBRIDIZATION EFFECTS IN UTX COMPOUNDS

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**Abstract.** – Systematic trends in the results of magnetic and specific-heat studies of UTX compounds (T = transition metal, X = p element) are discussed in terms of the degree of the U-5f electron delocalization which is controlled by the strength of 5f-ligand hybridization. Continuous changes of this parameter and its influence on ground-state properties were investigated in solid solutions of selected UTX compounds.

The main feature in the electronic structure of most of uranium compounds is a more or less narrow 5f band in the vicinity of  $E_F$ . Therefore the magnetic properties (formation of a 5f magnetic moment, magnetic ordering), as well as other electronic properties (transport properties, specific heat) are intimately connected with the degree of 5f-electron localization. The direct overlap of 5f-wave functions of neighbouring atoms in case of compounds with sufficiently short inter-uranium distances is one of main delocalizing mechanisms.

The further mechanism delocalizing the 5f electrons in compounds is the ubiquitous hybridization of 5f states with the s, p or d states brought to the valence band by other-component atoms. This so called 5f-ligand hybridization [1] is the principal delocalizing process in compounds where a larger separation of U atoms prevents the direct 5f-5f overlap. The considered trends in hybridization can be tested by studying magnetic and other electronic properties within extended groups of isostructural compounds with different ligand elements.

In this contribution we discuss the development of the 5f-ligand hybridization and its influence on electronic properties of UTX compounds (T = transition metal, X = element from the group III. – V. of the Periodic Table). These compounds crystallize in different structure types. The two most extended isostructural groups are characterized by the structure types  $ZrNiAl$  (hexagonal) or  $CeCu_2$  (orthorhombic). In the case of a relatively weak 5f-ligand hybridization, i.e. for late d metals and for larger X and T constituents, magnetic ordering of the ferromagnetic type is found in the hexagonal compounds. One of the prominent features of compounds with this structure is a strong anisotropy of the magnetic properties (uniaxial with the easy-magnetization axis being along the c axis in all compounds except in  $UPdIn$  where it is probably located in the basal plane), which is one of arguments for the 5f origin of magnetism. In ferromagnetic compounds anisotropy fields as high as 200 T were esti-

mated from magnetization measurements on  $UCoGa$  and  $UNiGa$  [2, 3]. Our high-field experiments on most of hexagonal compounds proved the presence of such a strong anisotropy to be a general feature irrespective to the type of ground state.

The compounds with stronger hybridization ( $URuAl$ ,  $URuGa$ ) show spin-fluctuation (SF) character of the magnetic susceptibility and no magnetic ordering is found down to lowest temperatures. As the Rh isotopes are ferromagnetic, the details of the SF  $\rightarrow$  F transition can be studied on the  $U(Ru,Rh)Al$  and  $U(Ru,Rh)Ga$  pseudoternary systems.

The development of the temperature dependences of magnetic susceptibility in  $URu_{1-x}Rh_xAl$  compounds is seen in figure 1. Although the character of the low-temperature susceptibility is different for  $URuGa$  and  $URuAl$  [3], the substitution of Rh smears out the characteristic low-temperature deviations from the CW law in both systems already for  $x > 0.1$ . The onset of ferromagnetism is observed around  $x = 0.2$ . The susceptibility at higher temperatures is well described by a modified Curie-Weiss law over the whole concentration region. The concentration dependences of  $T_C$  and  $\theta_p$ ,

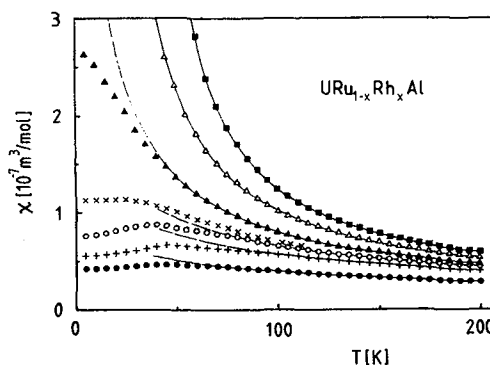


Fig. 1. – Temperature dependence of the susceptibility in  $URu_{1-x}Rh_xAl$  for  $x = 0.0$  (●),  $0.05$  (+),  $0.10$  (○),  $0.15$  (×),  $0.2$  (▲),  $0.3$  (△),  $0.4$  (■). The curves are Curie-Weiss fits.

the absolute value of which can be roughly associated with  $T_{SF}$  in the SF region, are displayed in figure 2. It is seen that  $\theta_p$  changes the sign in the region of the onset of magnetic ordering. Nevertheless, it approaches  $T_C$  only for more than 60 % Rh. It should be pointed out that no enhancement of the specific-heat-coefficient  $\gamma$  was found near the onset of magnetic ordering and the value of  $\gamma \simeq 80 \text{ mJ/K}^2\text{mol}$  is the upper limit in the mentioned systems. In contrast with ferromagnetic compounds, substantially enhanced  $\gamma$ -values were found in the materials with the antiferromagnetic ground state, namely in UNiAl ( $167 \text{ mJ/K}^2\text{mol}$ ) [5], and in UPdIn ( $280 \text{ mJ/K}^2\text{mol}$ ), where a more complex magnetic structure is supposed [6].

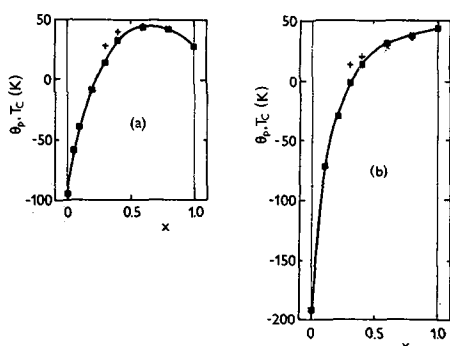


Fig. 2. - Concentration dependence of  $\theta_p$  ( $\blacksquare$ ) and  $T_C$  ( $\circ$ ) of  $\text{URu}_{1-x}\text{Rh}_x\text{Al}$  (a) and of  $\text{URu}_{1-x}\text{Rh}_x\text{Ga}$  (b). The curves are guides to the eye.

General rules for variation of hybridization seem to be valid also for the UTe and UTSi compounds crystallizing in the  $\text{CeCu}_2$  structure. The compounds with  $T = \text{Co}$  and  $\text{Ru}$  are non-magnetic while for transition metals with higher occupation of the d states ( $\text{Ni}$ ,  $\text{Rh}$ ,  $\text{Pd}$  and  $\text{Pt}$ ) magnetic ordering appears [7]. The magnetic properties of these compounds are again strongly anisotropic in consistence with the dominant role of U-5f electrons. We have performed the high-field measurements on UPdGe, which is ferromagnetic below 30 K, showing the anisotropy field to be of the order of magnitude of 200 T. The comparison of results obtained on field-oriented powders and randomly-oriented fixed powders suggests that the magnetic anisotropy is not uniaxial. The magnetic moment extrapolated to 0 T is  $1.5 \mu_B / \text{f.u.}$  similar to UPdIn.

We have also performed specific-heat experiments on UTe compounds with 3d and 4d transition metals which yield, except for URhGe, only moderate  $\gamma$ -values. The values of 48 and  $34 \text{ mJ/K}^2\text{mol}$  were found for the itinerant paramagnets UCoGe and URuGe,

respectively, and 25 and 22 for UNiGe and UPdGe, respectively, which show already magnetic ordering. In URhGe (ferromagnetic below 9 K) the value extrapolated from the paramagnetic region is as high as  $100 \text{ mJ/K}^2\text{mol}$ . A similar value of  $\gamma$  ( $145 \text{ mJ/K}^2\text{mol}$ ) was reported by Ramirez *et al.* [8] for the iso-electric compound UIrGe. This compound is, however, antiferromagnetic below 17 K, in contrast with URhGe, which was claimed to be a weak itinerant ferromagnet [7]. It is useful to note that these compounds are located (within this group of isostructural compounds) at the crossover from itinerant paramagnetism to stable-magnetic-moment behaviour which is connected with gradually reduced 5f-d hybridization when proceeding to compounds with transition metals with nearly filled d states.

These tendencies were found to be common for both groups of isostructural UTX compounds ( $\text{ZrNiAl}$ - and  $\text{CeCu}_2$ -type of structure) and are in fact demonstrating the correlation between the hybridization effects on electronic structure and the magnetic properties, respectively.

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