Magnetic properties of RTX intermetallic compounds

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Recently the properties of the ternary RTAl, where R = Gd, Tb, Ho and T = Rh, Pd, Ni were examined [1-5]. The investigations of the monocrystalline GdPdAl, GdNiAl and polycrystalline TbPdAl samples showed that these compounds belong to the ternary intermetallics with unstable crystal structure. Their crystal structure was sensitive to the history of the sample and a treatment. The ternary lanthanide aluminides RPdAl were reported for the first time by Dwight [6]. The annealed samples obtained by Dwight crystallized in the orthorhombic TiNiSi – type structure, while the rapidly cooled samples turned out to be hexagonal ZrNiAl – type as reported Hulliger [7, 8]. The samples GdPdAl and GdNiAl grown by the Czochralski method crystallized in the hexagonal ZrNiAl – type structure. However, after powdering some parts of single crystal the extra lines connected with the TiNiSi type structure in the powder diffraction spectra were observed. This suggests a change of the crystal structure under pressure during powdering process. The lattice parameters, electrical resistivity and magnetic susceptibility measurements gave evidence of an isostructural phase transition, from a high temperature modification HTM I phase to a low temperature modification HTM II phase, at about 200 K for GdPdAl and GdNiAl and at about 100 K for TbPdAl. The jump related to this transition causes a decrease of the lattice parameter a and an increase of the lattice parameter c during the cooling process (Fig.1). This caused a rapid contraction of the unit cell volume. Besides the isostructural phase transition, a magnetic ordering of the Gd sublattice are observed below 48 K and 60 K for GdPdAl and GdNiAl respectively. TbPdAl was ordered below 43 K. Additionally a spin reorientation process at about 20 K occured. The XPS measurements of GdPdAl revealed a dominating contribution of the d electrons at the Fermi level. A formation of a narrow Pd 4d band was found [8]. The effective magnetic moment was enhanced in the HTM II phase for all compounds and was equal 8.65 μ_B for GdNiAl, 8.35 for GdPdAl and 10.5 for TbPdAl. In the HTM I phase it was exactly equal to the free ion value of Gd^{3+} or Tb^{3+}. The GdRhAl single crystal also grown by the Czochralski method crystallized in the TiNiSi type structure [5]. This compound exhibited no isostructural phase transition as was found from the lattice parameters, electrical resistivity and magnetic susceptibility measurements. The ordering temperature was 30 K. The effective magnetic moment was 8.06 μ_B. Recently examined HoPdAl, which crystallized in the ZrNiAl type structure, exhibited no isostructural phase transition [4]. This compound was ordered at 16 K and the effective magnetic moment was in agreement with the free ion value.

The electronic structure measurements reveal that GdPdAl, TbPdAl, DyPdAl and HoPdAl belong to the intermetallic compounds with a narrow Pd 4d band. The narrow band is formed due to the separation of the palladium atoms in ZrNiAl – type structure leading to a reduction of the overlapping of the 4d orbitals of the neighbouring Pd atoms. The localization of the Pd 4d and the hybridization process are responsible for the coupling in these compounds and influence the magnetic properties. Palladium does not contribute to the density of states at the

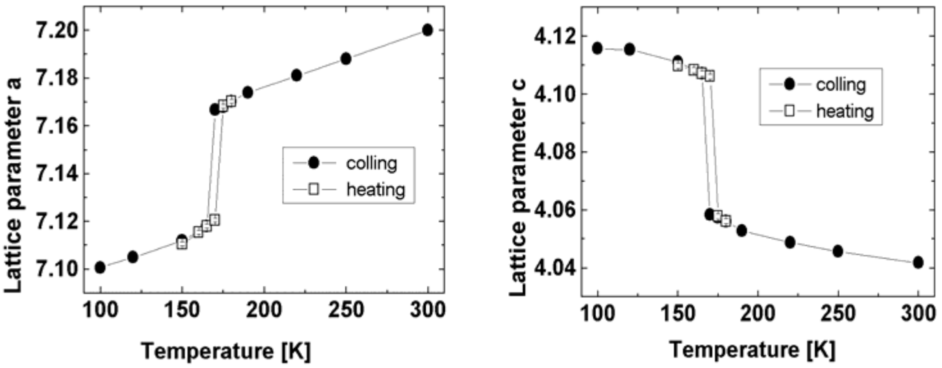

Fig. 1. The variation of the lattice parameters a and c of the GdPdAl single crystal on heating and cooling.

Fermi level for DyPdAl, HoPdAl and also for GdPdAl and TbPdAl within HTM I phase. Growing interaction in Gd sublattice with decreasing temperature caused a movement of rare earth atoms towards palladium and a rapid shortening Gd-Pd1 bond length in the isostructural transition [9]. The energy of the system enhanced by a geometrical frustration, present in these compounds, may be reduced by such isostructural transition. The increase of the lattice parameter *c* for GdPdAl may allow alignment of the Gd magnetic moments along this direction. This transition leads to the electronic structure changes what was confirmed by the temperature changes of the XPS valence band for GdPdAl. A variation of the valence band, from a flat plateau within HTM I to a narrow peak within HTM II phase, accompanied by depletion of the Pd 4d band was found [9]. This indicates that the palladium 4d electrons participate in the coupling of the system in the HTM II phase. The compounds DyPdAl, HoPdAl and ErPdAl exhibit no isostructural transition.

Such isostructural transition were also not observed for GdPdX (X = Al, Si, Ga, Ge, Sn) and GdRhAl, which grew in the orthorhombic structures. The ordering temperatures GdRhAl and GdPdX are similar and they are close to 30 K (Fig.2) [5].

This suggests that not only the contribution of d electrons but also the number of sp electrons and their spatial distribution, are very important for hybridisation process, influence the crystal structure and magnetic properties of the examined compounds.

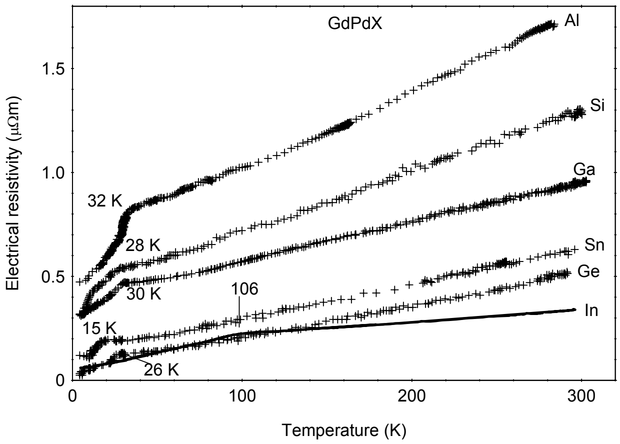

Fig. 2. Thermal variation of the electrical resistivity of the GdPdX grown in the orthorhombic crystal structure.

In conclusion, the electronic structure measurements reveal that GdPdAl, TbPdAl, DyPdAl and HoPdAl belong to the intermetallic compounds with a narrow Pd 4d band. Not only a contribution of d electrons but also a number of sp electrons and their spatial distribution, are very important for hybridisation process, influence the crystal structure and magnetic properties of the examined compounds.

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