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UTX COMPOUNDS IN HIGH MAGNETIC FIELDS

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High-field magnetic properties are presented of ternary compounds of the type UTX, where T is a transition metal and X is Si or Ge. The comparison of results obtained on powder free to be oriented by the field and on randomly oriented powder fixed in frozen alcohol reveals strong anisotropy effects. Complicated magnetic structures are indicated by one or more transitions seen in the magnetization curves of the antiferromagnetic compounds.

1. Introduction

The magnetic behaviour of U intermetallic compounds is intimately connected with the 5f electrons of U. The electronic structure of these materials is dominated by a more or less narrow 5f band located at E_F . The 5f-band formation is mainly determined by inter-site 5f-wave function overlap and the hybridization of 5f states with the ligand states. The first one dominates in compounds in which the distance between nearest-neighbour U atoms is smaller than the Hill limit (340–360 pm). The second delocalizing mechanism becomes important especially in the case of a reduced or absent overlap of 5f-wave functions.

Within the framework of an extensive study of the electronic properties of UTX compounds, with T a transition metal and X a p-block metal, we have investigated the systematics in the development of the 5f-ligand hybridization in the compounds with X = Si and Ge as it is reflected in the magnetization in high magnetic fields.

2. Experimental

The various UTX compounds with X = Si or Ge were prepared by arc melting in an argon atmosphere. The majority of the samples were found to have crystallized in the orthorhombic CeCu₂-type structure. The corresponding lattice constants are in a good agreement with the results of Troc and Tran [1] and Chevalier et al. [2]. The X-ray diagrams of UNiSi and UPdSi could not be originally indexed on the basis of the CeCu₂ type. After annealing the sample at 650°C for 4 weeks the X-ray pattern of UNiSi changed drastically and showed the structure of the AlB₂ type.

The same heat treatment of UPdSi resulted in the CeCu₂ type of structure.

The magnetization measurements were performed in the high magnetic field installation at the University of Amsterdam [3] on samples prepared in two forms: a) powder being free in the sample holder so that it can be oriented by the applied magnetic field, b) powder mixed with alcohol and cooled down to 4.2 K in zero magnetic field. In this way the frozen alcohol fixes the powder in a state of randomly-oriented grains.

3. Results and discussion

The magnetization curves taken on the UTSi and UTGe compounds at 4.2 K in fields up to 38 T are shown in fig. 1. The following features can be read out.

a) The highest magnetic moments are found for the compounds containing T metals with nearly-filled d states (Ni, Pd, Pt). As the d occupation is reduced, the magnetism weakens; the magnetic moments are reduced and finally the magnetic ordering disappears. b) The magnetic properties become more pronounced when moving from the silicides to the isoelectronic germanides.

An analogous development of ground-state properties has also been found for UTX compounds with the Fe₂P (ZrNiAl)-type of structure where X = Al, Ga, Sn and In [3].

This systematics can be reconciled with the expected tendencies in the 5f-ligand (5f–d and 5f–p) hybridization delocalizing 5f electrons and, consequently, destabilizing the 5f-moments.

The huge anisotropy, which can be concluded from

the comparison of the results on free and fixed powders, is found already in the paramagnetic state. Note that the almost identical magnetization curves measured on free and oriented powder of the very weak paramagnets UCoSi, URuSi and URuGe do not necessarily indicate absence of magnetic anisotropy. Very weak torque forces acting on the powder grains, not sufficiently strong to orient them, may account for this result. Only measurements on single crystals can answer this question.

The estimated anisotropy fields in the magnetically-ordered compounds exceed 100 T. This observation

can be associated with the anisotropy of the 5f-ligand hybridization (besides the 5f-5f overlap) which yields strong anisotropy of $N_{sf}(E_F)$ and also mediates the anisotropic exchange interaction, responsible for magnetic ordering of the 5f moments.

The orthorhombic symmetry of the CeCu₂ compounds, in which the nearest-neighbour U atoms form one-dimensional zig-zag chains, leads to complicated magnetic structures documented by one or more transitions seen in the magnetization curves of the compounds which have been reported as antiferromagnetic [1,2]. In order to resolve the magnetic arrangements

Fig. 1. Magnetization curves of U3dSi and U3dGe (a), U4dSi and U4dGe (b) and U5dSi and U5dGe (c) compounds at 4.2 K measured on free (Δ) and randomly-oriented powder ($\circ$). The solid lines correspond to data taken with pulses in which the field increases (lower curves) and subsequently decreases (upper curves), both linearly in time (~ 40 T/s). On the solid curves with maximum field of 38 T, the data with decreasing field have been taken with B decreasing linearly in time (~ 55 T/s). The data with increasing field were collected during much faster, uncontrolled increase of the field and therefore are less accurate. Therefore, the difference below 15 T between the fixed-powder magnetization curves for UPtSi must be considered as an artifact of the measurement. Note that the magnetic moments of UPtSi and UPtGe should be considerably larger than these found in the highest available field of 38 T, because the metamagnetic transitions on the magnetization curves are not yet completed in this field (see fig. 1(c)). The extension of the magnetization measurements on these compounds to fields in the 60 T region is currently in progress at the High Magnetic Field Facility at Osaka University.

Fig. 1. (cont.).

the necessary neutron-diffraction studies are currently performed.

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