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Published in:
Physica B-Condensed Matter

DOI:
[10.1016/0921-4526\(89\)90500-0](https://doi.org/10.1016/0921-4526(89)90500-0)

[Link to publication](#)

Citation for published version (APA):

Havela, L., Bruck, E. H., Veenhuizen, P. A., de Boer, F. R., Sechovsky, V., Buschow, K. H. J., & Menovsky, A. A. (1989). High-Field Magnetization of Ternary Uranium 1-1-1 Intermetallics. *Physica B-Condensed Matter*, 155(1-3), 221-224. DOI: 10.1016/0921-4526(89)90500-0

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HIGH-FIELD MAGNETIZATION OF TERNARY URANIUM 1:1:1 INTERMETALLICS

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Examples are presented to illustrate that high-field magnetization measurements on the class of ternary compounds UTX, in which T is a transition metal and X a non-transition metal, provide crucial information regarding the electronic structure of these compounds.

1. Introduction

The class of ternary intermetallic U compounds UTX, where T is a transition or a noble metal and X a non-transition metal, comprises more than forty compounds. The two largest isostructural groups are connected with the hexagonal ZrNiAl (Fe_2P) and the orthorhombic CeCu₂ structure types. In both groups the compounds are formed by U with late transition metals and with X = Al, Ga, In, Sn, Sb in the former group and X = Si and Ge in the latter one. General trends in ground-state properties can be visualized in schematic diagrams, as shown in fig. 1 for the compounds with the ZrNiAl structure, in which the development of the magnetic behaviour is shown together with the electronic-specific-heat coefficient γ .

The magnetic behaviour of U intermetallic compounds is intimately connected with the 5f electrons, which are on the border between itinerant and localized. The electronic structure is dominated by a more or less narrow 5f band located near E_F . The 5f bandwidth, and consequently the degree of 5f-electron delocalization, is basically determined by the overlap of 5f wave functions which is substantial in compounds where the nearest interuranium distance

$d_{\text{U-U}}$ is lower than the Hill limit (340–350 pm) [1]. The values of $d_{\text{U-U}}$ in the Fe_2P -type compounds range from 350 pm (UFeAl) to 394 pm (UPdIn). Therefore in these compounds we expect the 5f–5f overlap to be small and the hybridization of 5f states with s, p, d states of the T and X component becomes a dominating mechanism delocalizing the 5f electrons (effectively broadening the 5f band). In the UTX compounds the ground state develops (see fig. 1) from Pauli paramagnetic (UFeAl) through paramagnetic with spin-fluctuation features (UCoAl, URuAl) towards stable magnetic moments originating from 5f electrons (as indicated by a huge anisotropy of the overall magnetic behaviour) with either antiferromagnetic ordering connected with heavy-fermion features (UNiAl and UPdIn, which is not displayed in fig. 1) or ferromagnetic ordering. This development can be considered as a consequence of a gradual reduction of the 5f – ligand hybridization when either filling the d states of the transition metals and/or increasing the radii of the ligands similar to results of the analysis of UX_3 compounds made by Koelling et al. [2]. The delicate situation of the 5f electrons at the border line between being itinerant and localized makes them sensitive to various external parameters like pressure or magnetic field.

We have performed magnetization measure-

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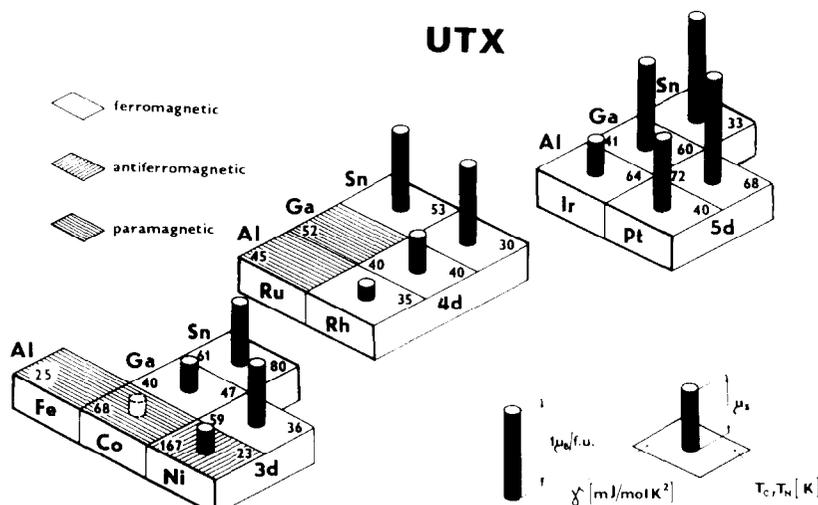


Fig. 1. Development of the ground-state properties in UTX compounds with the ZrNiAl (Fe_3P) structure.

ments at liquid-helium temperatures in fields up to 40 T in the High Magnetic Field Installation at the University of Amsterdam. In this paper we present some examples which demonstrate that to understand the strongly anisotropic properties of these compounds, measurements in high fields are indispensable.

2. Experimental

The various UTX compounds were prepared by arc melting in argon atmosphere and in some cases subsequent annealing between 600 and 900°C for periods up to several weeks. The quality was checked by X-ray diffraction. Then the samples were crushed into powder and measured in the following two forms:

(a) Powder being free in the sample holder so that it can be oriented by the high magnetic fields applied.

(b) Powder mixed with alcohol and cooled down to the temperature of the measurement in zero magnetic field. Thus the frozen alcohol fixes the powder in a state of randomly-oriented grains.

The highly anisotropic magnetic properties observed in the UTX compounds have led us to the preparation of single crystals. Until now, single

crystals of URuAl and URhAl have been grown [3].

3. Results and discussion

The low-field magnetic behaviour of URuAl is very similar to what is found for exchange-enhanced Pauli paramagnets like Pd or TiBe_2 : the susceptibility displays a broad maximum, which for URuAl is located around 50 K [4]. Therefore it is of interest to investigate whether also in this compound the observed increase of the low-temperature susceptibility with increasing temperature is accompanied by an increase of the differential susceptibility with increasing field. The magnetization curves measured at 4.2 K on a URuAl single crystal are shown in fig. 2. The magnetization is highly anisotropic and, if measured with the field in the direction of the c -axis, shows a pronounced positive deviation from linearity in fields above 20 T, whereas the magnetization measured within the basal plane is much smaller and remains linear. The magnetization of a randomly-oriented sample also shows the high-field upturn.

Since URhAl is ferromagnetic one may expect that substitution of Rh for Ru in URuAl will enhance the susceptibility and eventually lead to

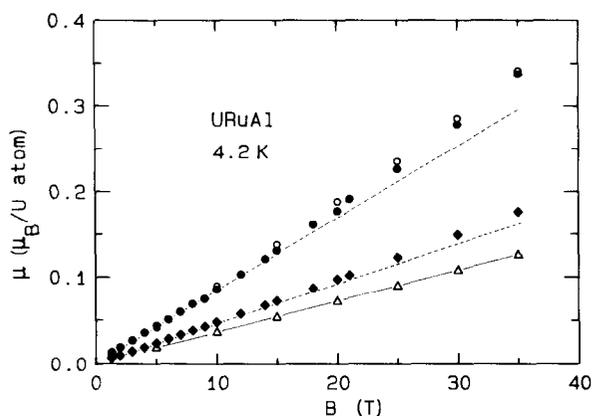


Fig. 2. Magnetization of URuAl at 4.2 K measured on a single crystal with the magnetic field applied parallel (○) and perpendicular (△) to the c -axis and on free (●) and randomly-oriented powder (◆). The solid line is a guide to the eye. The dashed lines represent extrapolations of the linear parts of the magnetization curves.

ferromagnetism. Indeed, a transition to the ferromagnetic state has been established in $URu_{1-x}Rh_xAl$ at about $x=0.2$ [4]. It can be seen in fig. 3 that upon Rh substitution the onset of the high-field upturn is first shifted to lower fields and that it becomes more pronounced. The upturn is strongly suppressed with further in-

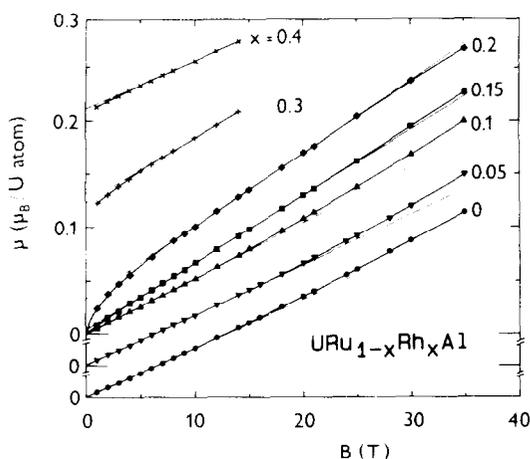


Fig. 3. Magnetization of $URu_{1-x}Rh_xAl$ compounds at 4.2 K measured on randomly-oriented powders. The solid lines are guides to the eye. The dashed lines represent extrapolations of the linear parts of the magnetization curves.

creasing Rh concentration when the system approaches the ferromagnetic instability. These observations obey the general correlation reported earlier [5] concerning the sign of the temperature and field dependence of the low-temperature susceptibility.

The magnetic isotherms measured at 4.2 K on the URhAl single crystal (fig. 4) demonstrate the general features of all ferromagnets in this group of compounds. The magnetization in the c -axis direction is almost twice that measured on a polycrystal, which suggests the c -axis to be an easy magnetization axis. The hard-axis magnetization is very low, showing no remanence, and the anisotropy field is clearly extremely high. The result obtained on a free powder is in reasonable agreement with the easy-axis magnetization.

UPdIn is an interesting representant of the antiferromagnetic compounds. The magnetic isotherms at 4.2 K are shown in fig. 5. In this compound the largest magnetic moment in the group of ZrNiAl-type compounds is found which is consistent with the expectation that the 5f-ligand hybridization should be minimal. The magnetic data combined with the specific-heat information suggest [6] that UPdIn becomes antiferromagnetic below 20.4 K and that the antiferromagnetic structure becomes canted

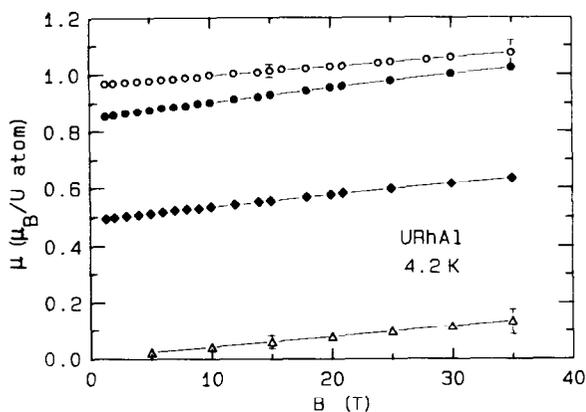


Fig. 4. Magnetization of URhAl at 4.2 K measured on a single crystal with the magnetic field applied parallel (○) and perpendicular (△) to the c -axis and on free (●) and randomly-oriented (◆) powder.

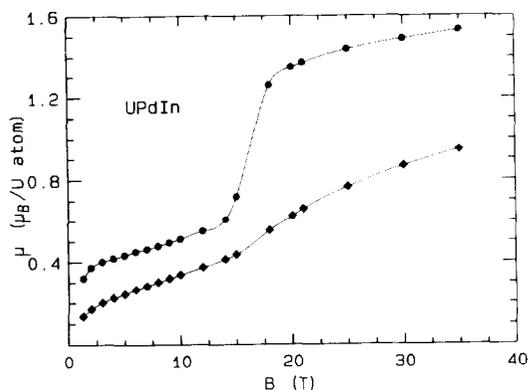


Fig. 5. Magnetization curves of UPdIn at 4.2 K measured on free (●) and randomly-oriented powder (◆). The solid lines are guides to the eye.

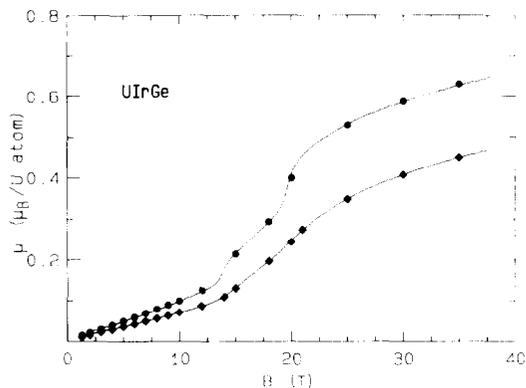


Fig. 6. Magnetization curves of UIrGe at 4.2 K measured on free (●) and randomly-oriented powder (◆). The solid lines correspond to data taken with a pulse in which the field decreases linearly in time.

below approximately 6 K which leads to a ferromagnetic component in the magnetization. The antiferromagnetic arrangement is destroyed above about 14 T, where $\mu_U B$ roughly equals $k_B T_N$.

Another example of a complex antiferromagnetic structure is UIrGe, which belongs to the group of compounds with the CeCu₂ type of structure. This compound has been suggested [7] to become antiferromagnetic below 16.1 K. The two steps in the magnetization (fig. 6) indicate a complex magnetic structure. The ratio of the magnetization values obtained on the two types of samples suggests that in this case the magnetic anisotropy is not uniaxial. Further experiments on this compound are in progress in order to determine the magnetic structure.

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