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# Magnetic anisotropy in UTX compounds

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## Abstract

We compare results of measurements of magnetization, specific heat, electrical resistivity and neutron diffraction on single crystals and oriented polycrystalline powders of various UTX compounds. Very strong anisotropy, persisting also in the paramagnetic state, is a general feature of these materials. Compounds with the hexagonal ZrNiAl-type structure exhibit, as a rule, a uniaxial anisotropy with an easy-magnetization direction along the *c* axis. Energies of the magnetic anisotropy (of the order of hundreds of kelvins) correlate with the expected development of hybridization strength within planes of the highest coordination of uranium atoms. As a result of this evidence the hybridization-mediated anisotropic exchange is tentatively assumed to be a source of the huge anisotropy. Particular symmetry of anisotropy reflects the type of arrangement of uranium atoms.

## 1. Introduction

Nearly 50 ternary intermetallic compounds of the formula UTX (T≡transition metal; X≡p electron metal) constitute one of the largest isostoichiometric groups of uranium intermetallics. The UTX compounds crystallize in six different crystal structure types [1]. The hexagonal ZrNiAl-type structure (Fig. 1) is observed in 21 cases, whereas 15 UTX compounds adopt the orthorhombic TiNiSi ($CeCu_2$) structure type [1–4].

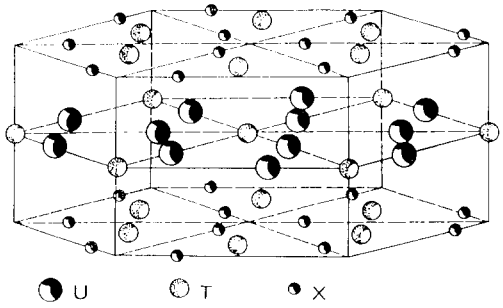


Fig. 1. Schematic drawing of the ZrNiAl-type structure adopted by a large group of UTX compounds.

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Such large isostructural groups of compounds provide convenient means for systematic investigation of various aspects of the 5f electron behaviour due to the different chemical surroundings realized by the various T and X components, whereas the geometry of the atom arrangements in the crystal lattice is conserved. UTX compounds exhibit a large variety of magnetic phenomena, ranging from weak paramagnetism to various types of magnetic ordering. The observed systematics of the ground-state properties [5] can be correlated with the development of the electronic structure due to the varying 5f ligand hybridization, which causes delocalization of the 5f electrons. As a consequence, Pauli paramagnets UFeAl and UFeGa are found in the strong-hybridization limit, whereas a gradual reduction in the 5f–d hybridization is expected for more populated d states of a transition metal. Indeed, we meet ordered magnetic moments in compounds with the last two transition metals of each transition metal series (cobalt and nickel; rhodium and palladium; irridium and platinum). The effect of the 5f hybridization with the p states of the non-transition element X on the magnetic behaviour of UTX compounds is also found. Thus a tendency towards the appearance of magnetic moments and magnetic cooperative phenomena is observed along series of compounds with p metals changing in group III or IV of the periodic table. These tendencies have been established in the course of experimental studies of magnetic, transport and thermal properties of UTX compounds of the two largest isostructural series [1, 5] and also emerge from band structure calculations on UTAl compounds [6–8].

Uranium compounds often exhibit strong magnetocrystalline anisotropy, indicating that the 5f electrons possess considerable orbital moments. The huge anisotropy fields observed in light actinide compounds are difficult to interpret in terms of single-ion anisotropy due to crystal field effects only. Cooper *et al.* [9, 10] have successfully explained the magnetic anisotropy and related effects in a number of cerium and light-actinide compounds by considering a two-ion exchange interaction mediated by anisotropic hybridization. The aim of the present paper is to demonstrate the presence of strong magnetic anisotropy in UTX compounds and to discuss the connection with the geometry of specific crystal structures and the strength of the 5f ligand hybridization.

## 2. Results and discussion

We have prepared polycrystalline samples of all compounds of the ZrNiAl type. In the cases of selected compounds (UCoAl, UNiAl, UNiGa, URuAl, URhAl and URuGa), single crystals were grown. Magnetization curves at 4.2 K were measured in the High-Field Installation of the University of Amsterdam, where semicontinuous magnetic fields up to 40 T can be generated [11]. Results obtained on the UNiGa single crystal in magnetic fields $B \| c$ and $B \perp c$ are displayed in Fig. 2. Like all magnetically ordered compounds of this group, the magnetic moment $M^c$ lies along the $c$ axis, whereas a very weak

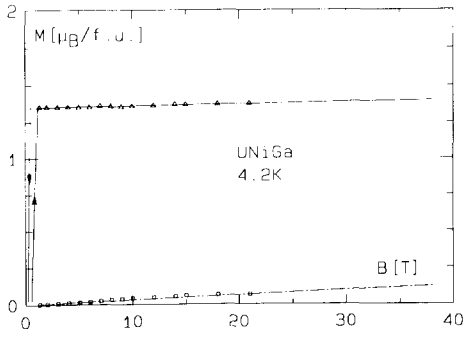


Fig. 2. Magnetization of UNiGa at 4.2 K measured on a single crystal in a magnetic field applied parallel (△) and perpendicular (□) to the $c$ axis. f.u., formula unit. The lines represent the results obtained within continuous field sweeps.

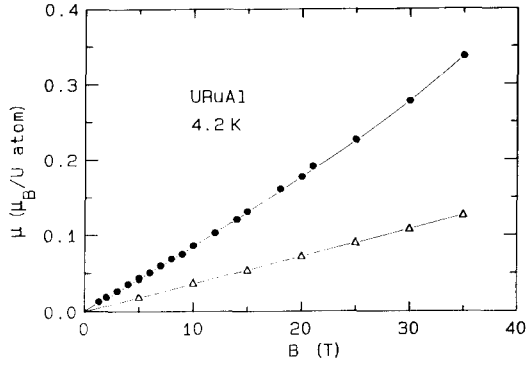


Fig. 3. Magnetization of URuAl at 4.2 K measured on a single crystal in a magnetic field applied parallel (●) and perpendicular (△) to the $c$ axis. The lines are guides for the eye.

linear magnetic response is measured with the field applied in the basal plane ($M^a$). The magnetization of URuAl (Fig. 3), which does not order magnetically, also exhibits strong uniaxial anisotropy with the hexagonal $c$ axis as the easy-magnetization direction. In order to assess the anisotropy by measurements on polycrystals we have measured magnetization of two types of powder sample: loose powder particles free to be oriented by the applied magnetic field and randomly oriented powder particles fixed in frozen alcohol. If there is sufficient orientation in the first case, the recorded magnetization $M_{or}$ should represent the behaviour of a single crystal with the field applied along the direction of the easy-magnetization axis, whereas in the latter case ($M_{rnd}$) an ideal polycrystal is simulated. For the free powder, almost full orientation was proved by observing $M_{or} \approx M^c$ for the compounds for which single crystals are available. A general relation $M_{or} \approx 2M_{rnd}$ was found for the UTX compounds with the ZrNiAl-type structure, implying uniaxial anisotropy to be a general feature of these materials. Exceptions are the Pauli paramagnets UFeAl and UFeGa for which no anisotropy of the magnetization could be detected.

Uniaxial anisotropy is also observed in the paramagnetic range as seen in Fig. 4 for UNiGa. The temperature dependence of the magnetic susceptibility for both directions of magnetic field, $\chi^c(T)$ for $B \| c$ and $\chi^a(T)$ for $B \perp c$, can be fitted with a modified Curie–Weiss law

$$\chi = \frac{C}{(T - \Theta_p^{c,\,a})} + \chi_0 \tag{1}$$

The values of the Weiss constants $\Theta_p^c$ and $\Theta_p^a$ are observed to differ by several hundreds of kelvins (Table 1), whereas the Curie constant $C$ and the small temperature-independent susceptibility $\chi_0$ do not depend on the magnetic field direction. The difference $\Delta\Theta_p = \Theta_p^c - \Theta_p^a$ can be taken as a measure of the magnetic anisotropy energy (in kelvins) in the paramagnetic range. Values for the anisotropy field can be estimated from the magnetization curves recorded at low temperatures. An estimate of $H_A$ can be tentatively obtained as the a field at which the high field extrapolations of $M^c$ and $M^a$ intersect. It is found that the values for $\mu_0 H_A$ (T) and $\Delta\Theta_p$ (K) (see Table 1) correlate quite well.

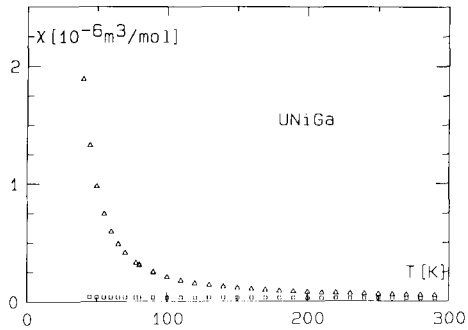


Fig. 4. Temperature dependence of the magnetic susceptibility of UNiGa measured on a single crystal in the magnetic field applied parallel (△) and perpendicular (□) to the $c$ axis.

TABLE 1

Basic magnetic characteristics of several single crystals of UTX compounds (for definitions see text)

| Compound | $M^c$ (35 T) ($\mu_B$) | $M^a$ (35 T) ($\mu_B$) | $\mu_0 H_A$ (T) | $\Theta_p^c$ (K) | $\Theta_p^a$ (K) | $\Delta\Theta_p$ (K) |
|---|---|---|---|---|---|---|
| URuAl | 0.3 | 0.12 | [a] | −53 | −410 | 357 |
| UCoAl | 0.6 | 0.10 | >320 | 33 | −400 | 433 |
| URhAl | 1.1 | 0.13 | >400 | 33 | −390 | 423 |
| UNiAl | 1.2 | 0.14 | 320 | −15 | −430 | 415 |
| UNiGa | 1.4 | 0.11 | 330 | 48 | −300 | 348 |
| UPdIn [12] | 1.5 | 0.5 | 110 | 34 | −74 | 108 |

[a]Available data prevent estimation of a reliable value.

Besides the strong magnetic anisotropy a pronounced anisotropy of electrical resistance (Fig. 5) is also observed, which can be attributed to a strong coupling of conduction electrons with the 5f electron system. Note that the two pronounced anomalies on $\rho(T)$ curves at around 40 K can be attributed to magnetic phase transitions in UNiGa [13].

Recent results of neutron diffraction studies on UCoAl [14], UNiGa [15], UNiAl [16] and URhAl [17] present more details of the arrangements of the uranium magnetic moments. The magnetic structure of most of these compounds is essentially composed of ferromagnetic (in the case of UNiAl almost ferromagnetic [16]) basal plane sheets with the direction of magnetic moments parallel to the $c$ axis, which is closely connected with the fact that the ZrNiAl-type structure is built up of two types of atom layer, namely U–T and T–X layers. The ordering of the uranium moments within the basal plane sheets should be a consequence of strong ferromagnetic interaction within these U–T layers where the 5f states of uranium and the d states of transition metal atoms are strongly hybridized. The confinement of the 5f charge density within the U–T layers leads to orientation of the magnetic moments perpendicular to these layers, *i.e.* along the $c$ direction. The 5f–d hybridization within the basal plane layers is much stronger than the interplanar hybridization, which is strongly supported by the observed difference between magnetic moments of rhodium found in U–Rh and Rh–Al planes [17]. The large reduction in the anisotropy energy is observed in the case of UPdIn (see Table 1), for which a substantial reduction of 5f–d hybridization can be expected.

The question remains about what would happen with the UTX compound if magnetic fields larger than the estimated anisotropy fields were applied in the basal plane. The strong connection of the geometry of the crystal lattice and the magnetic behaviour suggests that the direction of the magnetic moments is intimately connected with the bonding symmetry. Within the models, in which the hybridization-mediated two-ion exchange anisotropy is taken as the origin of the coupling of the magnetic system with the crystal

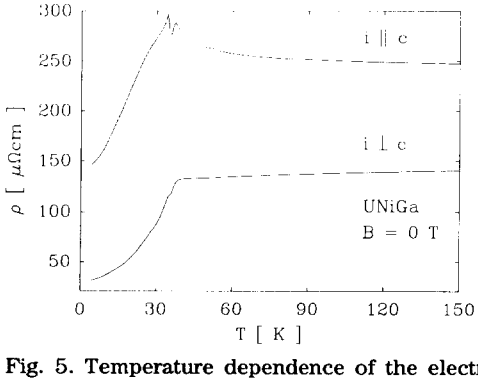


Fig. 5. Temperature dependence of the electrical resistivity of UNiGa measured by the d.c. four-probe method on single crystals (0.5 mm×0.5 mm×4 mm) for the electrical current parallel and perpendicular to the $c$ axis.

lattice, the effort to reach states with another orientation of magnetic moments would lead to violation of bonding. Also an estimate of magnetic anisotropy energy is of similar magnitude to the difference of the cohesion energies of different structures (about 100 meV).

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