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Magnetic anisotropy in UTX compounds

L. Havela^a, V. Sechovsky^a, F.R. de Boer^b, E. Brück^b and H. Nakotte^b

^a*Department of Metal Physics, Charles University, Ke Karlovu 5, CS-12116 Prague 2, Czechoslovakia*

^b*Van der Waals–Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands*

The huge magnetic anisotropy in UTX compounds is found to be closely connected with the coordination of U atoms in the crystal lattice. Generally, the easy magnetization direction is oriented perpendicular to the highest coordination plane or direction. The magnetization behaviour in ordered state yields effective anisotropy fields in the range 100–400 T. The anisotropy persists in the paramagnetic state where the values of the paramagnetic Curie temperature for the easy and hard magnetization directions differ by 100–400 K. The results are discussed in the framework of a model considering the anisotropic two-ion exchange interaction mediated by the 5f-ligand hybridization.

1. Introduction

An enormous magnetic anisotropy of light-actinide intermetallic compounds has been more emphasized with experimental exploration extending from simpler cubic materials to compounds with a lower symmetry of crystal lattice. In particular, the uniaxial anisotropy found frequently for hexagonal or tetragonal symmetry makes the anisotropy effects very apparent. The origin of the anisotropy is generally related to strong spin–orbit interactions and the presence of large orbital moments, which apparently are preserved even in compounds, for which the band description seems to be appropriate [1]. Here we concentrate on general features of the magnetic anisotropy in ternary UTX compounds crystallizing in different structure types. We show that the anisotropy is preserved even in compounds without magnetic ordering, where neutron diffraction evidence for orbital moments is still missing. The symmetry of the magnetic anisotropy is conspicuously related to the coordination of U atoms, which supports the idea of hybridization-mediated exchange anisotropy as primary at least in this class of U compounds.

For this purpose, we compare compounds of three different structure types, namely UNiAl

(hexagonal ZrNiAl-type structure), UNiGe (orthorhombic TiNiSi type), and UPdSn (hexagonal GaGeLi type). All three compounds are anti-ferromagnetic at low temperatures.

2. Experimental

Magnetic measurements reported here were carried out on single crystals grown by means of a tri-arc Czochralski technique. The magnetic susceptibility in the temperature range 4.2–300 K in magnetic fields up to 1.3 T was measured by means of a pendulum magnetometer. Magnetization measurements at 4.2 K were performed in semicontinuous fields up to 38 T in the High Field Installation at the University of Amsterdam.

3. Results and discussion

UNiAl crystallizes in the hexagonal ZrNiAl-type of structure consisting of a sequence of U–Ni and Ni–Al basal-plane layers. Each U atom has four U nearest neighbours within the basal U–Ni plane ($d_{U-U} = 349$ pm), two neigh-

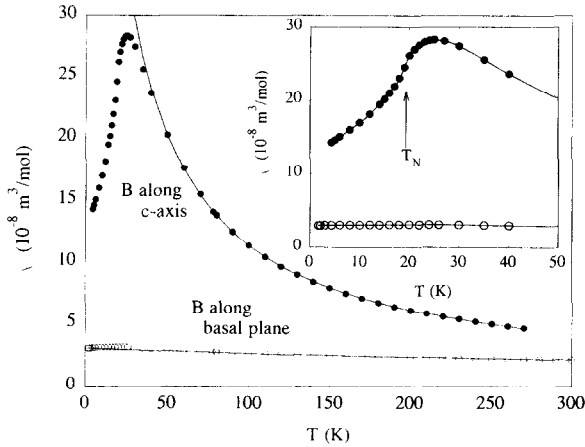


Fig. 1. Temperature dependence of the magnetic susceptibility of a UNiAl single crystal in the c -axis and in the basal plane. Full lines represent the MCW fit with parameters mentioned in text.

bours along the c -direction are separated by $d_{U-U} = 403$ pm.

The temperature dependence of the magnetic susceptibility for fields applied along the c -axis ($B \parallel c$) displays a pronounced broad maximum at 26 K (fig. 1). The magnetic ordering temperature $T_N = 19$ K derived from specific heat measurements [2] is considerably lower than the maximum in $\chi(T)$ but it coincides with the temperature of the maximum in $\partial(T\chi)/\partial T$ versus T . The susceptibility above 40 K obeys the Modified Curie–Weiss (MCW) law:

$$\chi = C/(T - \Theta_p) + \chi_0, \quad (1)$$

with $\mu_{\text{eff}} = 2.85\mu_B/\text{U}$ atom, $\Theta_p = -13$ K and a small χ_0 term of $1 \times 10^{-9} \text{ m}^3/\text{mol f.u.}$ The susceptibility in the basal plane ($B \perp c$) is much smaller and only weakly temperature dependent, but at high temperatures ($T > 120$ K), it can also be approximated by the MCW law. Naturally, there remains some uncertainty due to the contribution of a possible projection of the c -axis susceptibility (crystal mosaicity, error in crystal mounting), but the approximate values $\mu_{\text{eff}} = 2.25\mu_B/\text{U}$ atom, $\Theta_p = -380$ K and $\chi_0 = 1 \times 10^{-8} \text{ m}^3/\text{mol f.u.}$ can be extracted. The magnetization curves at 4.2 K (fig. 2) also demonstrate that the main magnetic response in

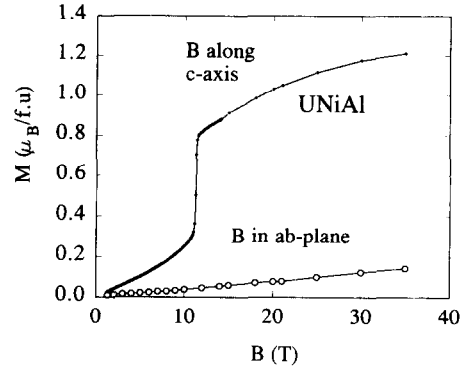


Fig. 2. Magnetization curves of a UNiAl single crystal at 4.2 K for $B \parallel c$ and $B \perp c$ -axis. Lines are guides to the eye.

UNiAl is confined to the c -axis. When the magnetic field is applied in this direction, a metamagnetic transition is induced at $B \approx 11$ T. In fields above this transition, the magnetization gradually saturates to a value of about $1.3\mu_B$. The magnetization in the ab -plane is very small and linear in the magnetic field ($M_{35T}^{ab} = 0.14\mu_B/\text{f.u.}$). Therefore, it can be concluded that the c -axis is equivalent with an easy magnetization direction, which is a general feature of all documented UTX compounds with this type of structure. Another remarkable example is the ferromagnet URhAl where the c -axis is the only direction in which a magnetic remanence can be found [3]. In the basal plane it exhibits a paramagnetic response.

UNiGe crystallizes in the TiNiSi structure type, which is an ordered ternary variant of the orthorhombic CeCu_2 type of structure [4]. U atoms form zig-zag chains along the a -axis. The intra-chain U–U spacing is around 350 pm, whereas the separation of U atoms belonging to different chains is about 360 pm.

As inferred from figs. 3 and 4, the magnetic response depends on the orientation with respect to the crystallographic axes. Although the high temperature ($T > 80$ K) susceptibility along all three axes obeys a Curie–Weiss law with $\mu_{\text{eff}} \approx 2.7\mu_B/\text{f.u.}$, the paramagnetic Curie temperature Θ_p differs significantly, $\Theta_p^a = -98$ K, $\Theta_p^b = 3$ K and $\Theta_p^c = 41$ K. In the low temperature region, χ^a shows a broad maximum around 60 K whereas a sharp maximum at the ordering tem-

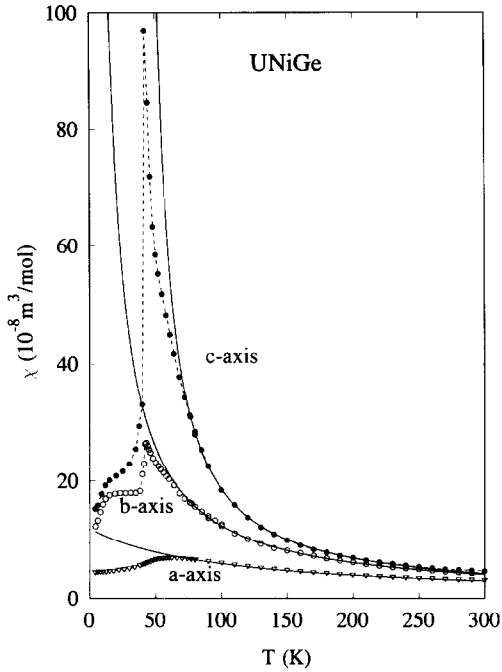


Fig. 3. Temperature dependence of the magnetic susceptibility of a UNiGe single crystal with field along the a -, b - and c -axis. The dashed lines are guides to the eye, the full lines represent the Curie-Weiss fits.

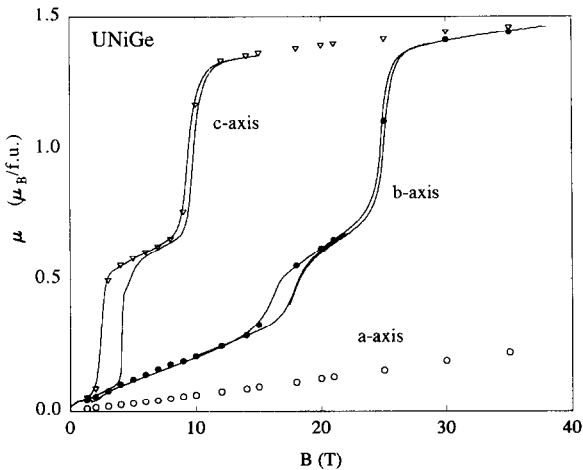


Fig. 4. Magnetization curves of the UNiGe single crystal at 4.2 K for $B\parallel a$, $B\parallel b$ and $B\parallel c$ -axis. Full lines represent measurements with field swept continuously up and down.

perature $T_N = 42$ K is found in both the $\chi^b(T)$ and the $\chi^c(T)$ curve. The magnetization along the a -axis is linear up to 35 T and a rather small value of $0.23\mu_B$ is found at the highest field. The magnetization curve for $B\parallel b$ displays two metamagnetic transitions at about 18 and 25 T. Two metamagnetic transitions are found also on the magnetization curve for $B\parallel c$, but the onset fields are much smaller, about 4 and 10 T, respectively. In both directions the first transition has a large hysteresis of about 1.6 T, while the hysteresis of the second transition is only 0.3–0.4 T. The first transition lifts the magnetization in both directions to about $0.55\mu_B/\text{f.u.}$, while the values of $1.37\mu_B/\text{f.u.}$ (b -axis) and $1.32\mu_B/\text{f.u.}$ (c -axis) are found just above the second transition. In both directions, a non-negligible high-field susceptibility remains at 35 T.

UPdSn crystallizes in the hexagonal GaGeLi-structure type, which is the ordered ternary variant of the CaIn_2 type of structure. U atoms form linear chains along the c -axis with an inter-uranium separation $d_{\text{U-U}} = 365$ pm. The separation of the chains is 460 pm [5].

From fig. 5, it is seen that, also for this compound, the susceptibility is anisotropic. Measuring with a magnetic field along the basal plane (the a and b directions are nearly equivalent), we observe two distinct maxima at 28 K and 38 K, respectively, which indicate two magnetic phase transitions ($\text{AF2} \rightarrow \text{AF1}$ and $\text{AF1} \rightarrow \text{P}$). For B parallel to the hexagonal c -axis, the susceptibility is much smaller and the lower maximum appears only in the form of a shoulder. In the temperature range 50–300 K, the susceptibilities for $B\parallel a$ and b are the same. They are well described by the Curie-Weiss law with $\theta_p = -2.5$ K and $\mu_{\text{eff}} = 3.3\mu_B/\text{f.u.}$ The $\chi^c(T)$ data yield the value $\theta_p \approx -60$ K [5] and a somewhat smaller effective moment than for the other two directions ($\approx 2.9\mu_B/\text{f.u.}$), if a small temperature-independent term $\chi_0 \approx 3 \times 10^{-9} \text{ m}^3/\text{mol f.u.}$ is included. However, these values can also be affected by a projection of the much higher susceptibilities for other axes. Magnetization curves at 4.2 K (fig. 6) for $B\parallel a$ and $B\parallel b$ suggest that these axes are magnetically nearly equivalent. Below 3 T, the magnetization is low

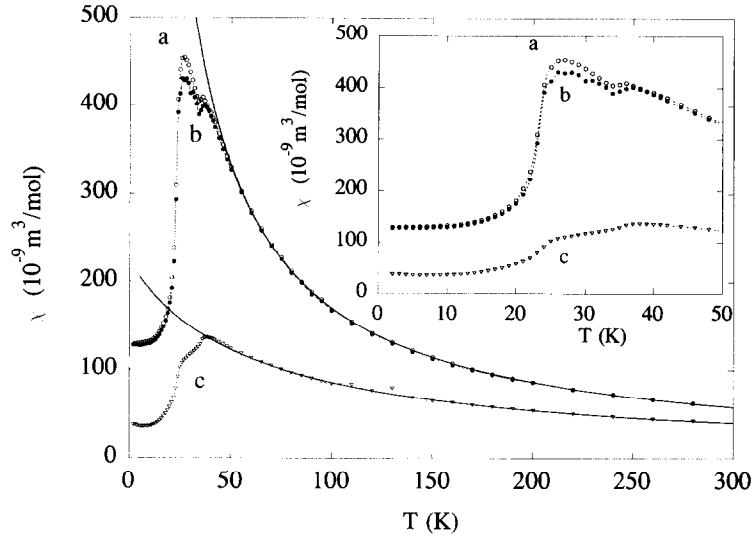


Fig. 5. Temperature dependence of the magnetic susceptibility ($\chi = M/B$) of a UPdSn single crystal in the a -, b - and c -axis (orthorhombic representation) in magnetic field of 1 T. Full lines represent the MCW fit with parameters mentioned in text.

and proportional to the magnetic field. The field of 3 T, where $\partial M/\partial B$ increases abruptly, can be taken as the field where a moment-reorientation transition commences. We cannot, however, distinguish where the transition terminates, because a smooth course of $M(B)$ gradually tending towards saturation can be followed up to 35 T, where $M^a = 1.7\mu_B/\text{f.u.}$ and $M^b = 1.6\mu_B/\text{f.u.}$ The c -axis is undoubtedly the hard magnetization

direction. The magnetization curve for $B\parallel c$ consists of three linear parts separated by anomalies at 3 T (an increase of the $M(B)$ slope) and 14 T (a small metamagnetic like anomaly). A maximum moment of only $0.6\mu_B/\text{f.u.}$ is reached in $B = 35$ T.

The enormous values of the effective anisotropy fields are definitely much larger than the maximum field attainable in our experiments. A rough estimate can be obtained from the intersection field of the high-field extrapolation of the $M(B)$ curves for B along the easy and hard magnetization axes. For the three compounds UNiAl, UNiGe and UPdSn, this estimate yields $B_a \approx 320, 230$ and 110 T, respectively. It is interesting to note that the difference between the values of the paramagnetic Curie temperature obtained from the susceptibility behaviour along the easy and hard magnetization axes, which can be taken as a measure of anisotropy in the paramagnetic state, is in these three cases $\Delta\Theta_p \approx 370, 140$ and 60 K. These values would yield anisotropy energies quite comparable to those derived from anisotropy fields above.

The uniaxial anisotropy found in UNiAl and other compounds with the ZrNiAl-structure type can be due to a strongly hybridized $5f-3d$ subsystem expected in the U-T layers due to a d_{U-U}

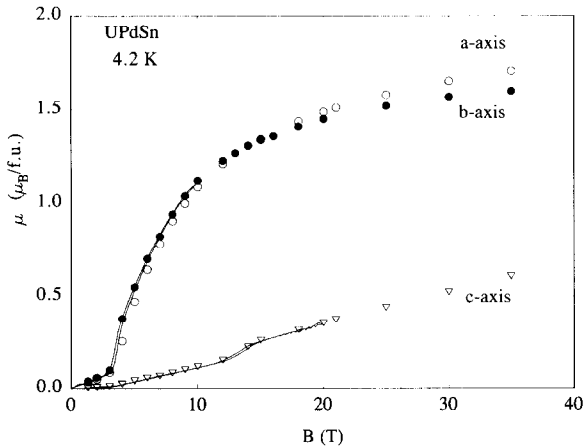


Fig. 6. Magnetization curves of a UPdSn single crystal at 4.2 K for $B\parallel a$, $B\parallel b$ and $B\parallel c$ -axis (orthorhombic representation). Full lines represent measurements with field swept continuously up and down.

value of the order of the critical Hill spacing and/or the presence of Ni atoms. This situation should lead to a compression of the 5f charge density into the basal plane and locking of the angular momenta in the *c*-direction. Such a model has been envisaged theoretically by Cooper et al. [6, 7], who introduced the notion of anisotropic two-ion exchange interaction mediated by the f-ligand hybridization, which leads to a hybridization-mediated exchange anisotropy. The easy-axis anisotropy appears here because the energy gain due to hybridization is maximal if the f orbitals pile up the charge along the interionic axis and the angular momentum of such an orbital is perpendicular to this direction. This also explains the large anisotropy energies (>10 meV), which can reach values characterizing differences of cohesion energies in different structure types. Such a model should be justified unless all atomic character of the 5f-wave functions is destroyed by a too strong 5f delocalization. We can speculate that the conclusions of this model as to the anisotropy symmetry are similar in the case of dominating homogeneous 5f–5f hybridization, i.e. for a non-negligible overlap of the 5f-wave functions.

Consistently, the *c*-direction is the hard magnetization axis is UPdSn with a linear coordination of U atoms along this direction. The less pronounced anisotropy of the U coordination in UNiGe makes this case less transparent, but also here the magnetically hard direction coincides with the axis of U chains.

4. Conclusions

The directions of the high coordination of the U atoms in UTX compounds are associated with

the hard magnetization directions. This symmetry and the strength of the anisotropy can be related to the hybridization-induced two-ion interactions as origin of the magnetic anisotropy.

Acknowledgements

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