UvA-DARE (Digital Academic Repository)

High-field magnetization of U₂T₂X compounds (T=Co, Ni, Rh, Pd, Ir, Pt and X=In, Sn)

Nakotte, H.; Prokes, K.; Brück, E.H.; Tang, N.; de Boer, F.R.; Svoboda, P.; Sechovsky, V.; Havela, L.; Winand, J.M.; Seret, A.; Rebizant, J.; Spirlet, J.C.

DOI

[10.1016/0921-4526\(94\)91093-6](https://doi.org/10.1016/0921-4526(94)91093-6)

Publication date

1994

Published in

Physica B-Condensed Matter

[Link to publication](#)

Citation for published version (APA):

Nakotte, H., Prokes, K., Brück, E. H., Tang, N., de Boer, F. R., Svoboda, P., Sechovsky, V., Havela, L., Winand, J. M., Seret, A., Rebizant, J., & Spirlet, J. C. (1994). High-field magnetization of U₂T₂X compounds (T=Co, Ni, Rh, Pd, Ir, Pt and X=In, Sn). *Physica B-Condensed Matter*, 201, 247-250. [https://doi.org/10.1016/0921-4526\(94\)91093-6](https://doi.org/10.1016/0921-4526(94)91093-6)

General rights

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <https://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

UvA-DARE is a service provided by the library of the University of Amsterdam (<https://dare.uva.nl>)

High-field magnetization of U_2T_2X compounds ($T = Co, Ni, Rh, Pd, Ir, Pt$ and $X = In, Sn$)

H. Nakotte^a, K. Prokeš^a, E. Brück^a, N. Tang^a, F.R. de Boer^{a,*}, P. Svoboda^b,
V. Sechovsky^b, L. Havela^b, J.M. Winand^c, A. Seret^c, J. Rebizant^c, J.C. Spirlet^c

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

^b Department of Metal Physics, Charles University, Ke Karlovu 5, 12116 Prague 2, Czech Republic

^c Commission of the European Communities, Joint Research Center, Institute for Transuranium Elements, Postfach 2340,
7500 Karlsruhe 1, Germany

Abstract

Structure parameters and magnetization in quasistatic fields up to 38 T at 4.2 K are reported for U_2T_2X compounds with $T = Co, Rh, Ni, Pd, Ir, Pt$ and $X = In, Sn$, which crystallize in the tetragonal U_3Si_2 structure. Even the highest applied fields are not sufficient to achieve saturation of the magnetization in any of these compounds of which the majority is antiferromagnetic and some are Pauli paramagnets. The importance of 5f-ligand hybridization for the magnetism in these compounds is discussed.

1. Introduction

Recently, a large new group of isostructural compounds with composition U_2T_2X ($T =$ transition metal and $X =$ p-electron element), crystallizing in the tetragonal U_3Si_2 structure, has been reported independently by Mirambet et al. [1] and Peron et al. [2]. From the U_2T_2In compounds, all compounds except U_2Co_2In are reported to possess the antiferromagnetic ground state as derived from DC-susceptibility [2,3] and AC-susceptibility [3]. Our preliminary specific-heat and electrical-transport measurements indicate absence of magnetic order also for U_2Rh_2In , U_2Pt_2In , U_2Co_2Sn and U_2Ir_2Sn .

In the case of equiatomic UTX compounds, a clear trend from itinerant 5f-electron behaviour to well defined local 5f magnetic moments has been found upon reducing the 5f-ligand hybridization [4]. Furthermore, the

strong magnetocrystalline anisotropy was found to orient the U-moments as a rule perpendicular to the strong U–U bonding distance (i.e. the shortest inter-uranium distance) [5].

The new family of U_2T_2X compounds offers a possibility to study further the role of the 5f-ligand hybridization and the connection between the shortest inter-uranium distance and the type of magnetocrystalline anisotropy. It is particularly interesting in this family of compounds that, depending on the constituting elements T and X , the shortest inter-uranium distance is found either within the basal plane or along the c axis.

In this paper, we present the structure and low-temperature high-field magnetization of a number of U_2T_2X compounds.

2. Sample preparation and characterization

U_2T_2X compounds with $T = Co, Ni, Rh, Pd, Ir, Pt$ and $X = In, Sn$ were synthesized by arc-melting

* Corresponding author.

Fig. 1. Double unit cell of U_2T_2X . The two shortest U distances, $d_{U-U}^{(1)}$ and $d_{U-U}^{(2)}$, are indicated.

appropriate amounts of the constituting elements. No further heat treatment was given to the ingots. By X-ray powder diffraction, all compounds were determined to form as ternary derivative of the U_3Si_2 structure (space group $P4/mbm$, $Z = 2$), reported earlier also for U_2Co_2Al [6]. The diffraction patterns of most of the compounds did not show additional reflections, indicating the amount of possible impurities to be less than 5%. Only in U_2Ir_2In , an appreciable amount of UIr is present.

The structures and atomic positions were determined on small single crystals extracted from the bulk material by means of an Enraf–Nonius four-circle diffractometer. No single crystal of U_2Ir_2In could be obtained. The U atoms occupy the 4h positions ($x, x + 0.5, 0.5$), while the T atoms are found on the 4g positions ($y, y + 0.5, 0$)

and the X atoms on the 2a positions (0, 0, 0). Hence, the structure consists of two alternating basal-plane layers, one containing only U and the other T and X atoms (Fig. 1). There are two nearest neighbours along the c axis at a distance $d_{U-U}^{(1)} = c$ and a third nearest neighbour in the basal plane at distance $d_{U-U}^{(2)}$, which is not very different from $d_{U-U}^{(1)}$. Depending on the choice of T and X, the distance $d_{U-U}^{(2)}$ can be larger or smaller than $d_{U-U}^{(1)}$. In Table I, the values for the two inter-uranium distances can be found together with the lattice parameters and the position parameters x and y .

3. Experimental results and discussion

Magnetization measurements at 4.2 K were performed in the Amsterdam High-Field Installation in magnetic fields up to 38 T on two kinds of samples consisting of powder particles of a size smaller than 100 μm , which are assumed to be single crystalline. Firstly, the powder particles were measured free to be oriented by the applied field, and secondly, they were measured fixed in a random orientation by frozen alcohol. The free-powder experiment is thought to represent a measurement on a single crystal along the easy magnetization direction, whereas the fixed powder represents an 'ideal' polycrystal. Consequently, the value of the free-powder magnetization is usually larger than that of the fixed powder. By comparing both results, information about the type of magnetocrystalline anisotropy may be gained. Furthermore, the free-powder measurement yields the value of the ordered moment in the case of complete ferromagnetic alignment.

Table 1
Structure parameters of U_2T_2X compounds

Compound	Lattice parameters		Position parameters		Inter-uranium distances	
	a (pm)	c (pm)	x	y	$d_{U-U}^{(1)}$ (pm)	$d_{U-U}^{(2)}$ (pm)
U_2Co_2In	736.1	343.1	0.1696	0.3697	343.1	353.1
U_2Ni_2In	737.5	357.2	0.1727	0.3761	357.2	360.2
U_2Rh_2In	755.3	360.5	0.1707	0.3667	360.5	364.7
U_2Pd_2In	763.7	375.2	0.1744	0.3716	375.2	376.6
U_2Ir_2In	759.6	358.2				
U_2Pt_2In	765.4	372.5	0.1727	0.3701	372.5	373.8
U_2Co_2Sn	728.9	350.5	0.1713	0.3706	350.5	353.1
U_2Ni_2Sn	726.3	369.1	0.1743	0.3756	369.1	358.0
U_2Rh_2Sn	752.4	363.0	0.1740	0.3702	363.0	370.3
U_2Pd_2Sn	760.3	378.5	0.1757	0.3726	378.5	377.8
U_2Ir_2Sn	756.6	360.1	0.1732	0.3692	360.1	370.7
U_2Pt_2Sn	767.0	369.8	0.1728	0.3703	369.8	374.8

Fig. 2. Field dependence of the magnetization of U_2T_2In compounds at 4.2 K. The symbols correspond with results obtained in step-wise field pulses on free (Δ) and fixed ($\circ$) powders. The solid lines represent measurements with pulses in which the field increases in an uncontrolled way and decreases linearly with time at a rate of 50 T/s.

Fig. 3. Field dependence of the magnetization of U_2T_2Sn compounds at 4.2 K. The symbols correspond with results obtained in step-wise field pulses on free (Δ) and fixed ($\circ$) powders. The solid lines represent measurements with pulses in which the field increases in an uncontrolled way and decreases linearly with time at a rate of 50 T/s.

In Figs 2 and 3, the magnetization is presented for the U_2T_2In and the U_2T_2Sn compounds, respectively, as the magnetic moment per formula unit. In all cases, the maximum applied field is not sufficient to achieve complete ferromagnetic alignment. For some compounds, like U_2Pd_2In and U_2Rh_2Sn , the antiferromagnetic ground state is very clearly corroborated by pronounced metamagnetic transitions. For others, like U_2Ni_2In , U_2Ni_2Sn , U_2Pd_2Sn and U_2Pt_2Sn , much less pronounced metamagnetic transitions or merely onsets are found. The magnetization of the compounds U_2Co_2In , U_2Rh_2In , U_2Pt_2In , U_2Co_2Sn and U_2Ir_2Sn is approximately linear in higher fields, indicating a Pauli-paramagnetic ground state in these compounds. The curvature of the magnetization below 10 T seen in some compounds is likely to be attributed to the magnetic saturation of impurity phase(s).

U_2Co_2In displays the lowest magnetization. If we neglect a possible Co contribution to the magnetization, a value of $0.14\mu_B$ is found for the U moment in 35 T. In 35 T, the strongest response for a free powder is found for U_2Pd_2In with a value of $0.60\mu_B/U$ -atom. The obtained values are much lower than in analogous UTX compounds [4,7], which may reflect stronger hybridization effects in these compounds, but the general trends of the

5f-ligand hybridization are found to be the same as in UTX compounds.

It has been found as an empirical rule [5] that the direction of the magnetic moments in uranium intermetallics with strong 5f-ligand hybridization is perpendicular to the shortest inter-uranium distance. In the case of magnetic saturation (complete ferromagnetic alignment), conclusions can be drawn regarding the type of magnetic anisotropy by comparing the observed ratio M_{fix}/M_{free} with values that can be calculated. If M_{fix}/M_{free} is close to 0.50, this indicates uniaxial type of anisotropy, whereas a M_{fix}/M_{free} value close to 0.79 stands for (multi-axial) easy-plane anisotropy. In the majority of the UT_2X_2 compounds, $d_{U-U}^{(1)}$ is the shortest uranium distance. Only in U_2Ni_2Sn and U_2Pd_2Sn , $d_{U-U}^{(2)}$ is the shortest uranium distance. On the basis of the empirical rule, only in the latter two compounds uniaxial anisotropy may be expected.

All U_2T_2X compounds investigated exhibit magnetic anisotropy, which is reflected by the different results for the free- and fixed-powder measurements. Unfortunately, even in the highest fields applied in the present study, magnetic saturation is not achieved in the U_2T_2X compounds. Therefore, we cannot be decisive about the type of anisotropy in the compounds studied. For this,

measurements in still higher fields are needed. The example of $\text{U}_2\text{Pd}_2\text{In}$, for which in higher fields $M_{\text{fix}}/M_{\text{free}}$ is larger than 1, illustrates that in these compounds rather complex phenomena may occur and that, in the case of absence of magnetic saturation, one should be extremely careful in using the $M_{\text{fix}}/M_{\text{free}}$ values to distinguish between easy-axis and easy-plane anisotropy.

4. Conclusions

Structure and magnetization data have been presented for the new family of $\text{U}_2\text{T}_2\text{X}$ compounds ($\text{T} = \text{Co}, \text{Ni}, \text{Rh}, \text{Pd}, \text{Ir}, \text{Pt}$ and $\text{X} = \text{In}, \text{Sn}$), which crystallize in the tetragonal U_3Si_2 structure. Most of the compounds investigated possess the antiferromagnetic ground state, except for $\text{U}_2\text{Co}_2\text{In}$, $\text{U}_2\text{Rh}_2\text{In}$, $\text{U}_2\text{Pt}_2\text{In}$, $\text{U}_2\text{Co}_2\text{Sn}$ and $\text{U}_2\text{Ir}_2\text{Sn}$ which, very likely, are all Pauli-paramagnets. The available fields up to 38 T are not sufficient to achieve ferromagnetic alignment in any compound investigated. Therefore, regarding the types of magnetic anisotropy no conclusions can be drawn from the $M_{\text{fix}}/M_{\text{free}}$ values. The importance of hybridization effects in these compounds is reflected by a relatively low magnetic response.

In order to be more conclusive on the magnetism of these compounds, magnetization measurements in higher fields, measurements on single crystals and neutron-diffraction experiments are necessary.

Acknowledgements

The present investigation was supported by the 'Stichting voor Fundamenteel Onderzoek der Materie' (FOM), which is financially supported by the 'Nederlandse organisatie voor Wetenschappelijk Onderzoek' (NWO). The work of P.S., V.S. and L.H. was supported by the Grant Agency of the Czech Republic under grant no. 202/93/0184.

References

- [1] F. Mirambet, P. Gravereau, B. Chevalier, L. Trut and J. Etourneau, *J. Alloys Comp.* 191 (1993) L1.
- [2] M.N. Peron, Y. Kergadallan, J. Rebizant, D. Meyer, S. Zwirner, L. Havela, H. Nakotte, J.C. Spirlet, G.M. Kalvius, E. Colineau, J.L. Oddou, C. Jeandey, J.P. Sanchez and J.M. Winand, *J. Alloys Comp.*, in press.
- [3] Z. Zolnieriek and A. Zaleski, 23ièmes Journées des Actinides (Schwarzwald, Germany, 1993).
- [4] V. Sechovsky and L. Havela, in: *Ferromagnetic Materials*, Vol. 4, eds. E.P. Wohlfarth and K.H.J. Buschow (North-Holland, Amsterdam, 1988) p. 309.
- [5] L. Havela, V. Sechovsky, F.R. de Boer, E. Brück and H. Nakotte, *Physica B* 177 (1992) 159.
- [6] A.O. Sampaio, E. Santa Marta, H.L. Lukas and G. Petzow, *J. Less-Common Met.* 14 (1968) 472.
- [7] V. Sechovsky, L. Havela, P. Nozar, E. Brück, F.R. de Boer, A.A. Menovsky, K.H.J. Buschow and A.V. Andreev, *Physica B* 163 (1990) 103.