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

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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
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/mmb, $Z = 2$), 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

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

U\(_2\)Co\(_2\)In displays the lowest magnetization. If we neglect a possible Co contribution to the magnetization, a value of 0.14\( \mu_\text{B} \) 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\( \mu_\text{B} \)/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 \( M_{\text{fix}}/M_{\text{free}} \) with values that can be calculated. If \( M_{\text{fix}}/M_{\text{free}} \) is close to 0.50, this indicates uniaxial type of anisotropy, whereas a \( M_{\text{fix}}/M_{\text{free}} \) value close to 0.79 stands for (multi-axial) easy-plane anisotropy. In the majority of the \( \text{UT}_2X_2 \) compounds, \( d(1) \) is the shortest uranium distance. Only in \( U_2Ni_2Sn \) and \( U_2Pd_2Sn \), \( d(2) \) 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 U$_2$Pd$_2$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 U$_2$T$_2$X compounds (T = Co, Ni, Rh, Pd, Ir, Pt and X = In, Sn), which crystallize in the tetragonal U$_3$Si$_2$ structure. Most of the compounds investigated possess the antiferromagnetic ground state, except for U$_2$Co$_2$In, U$_2$Rh$_2$In, U$_2$Pt$_2$In, U$_2$Co$_2$Sn and U$_2$Ir$_2$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.

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