

Version Date: July 14, 1999

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OCT 12 1999

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Structural and Magnetic Properties of  $UCo_{1/3}T_{2/3}Al$  Solid Solutions (T = Ru, Pt, Rh)

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## Abstract

We report on neutron diffraction studies of  $UCo_{1/3}T_{2/3}Al$  (T = Ru, Pt, Rh). All three solid solutions form in the hexagonal  $ZrNiAl$  structure. The Ru-containing compound is found to be chemically ordered, while the Pt-containing compound is nearly disordered and the Rh-containing compound is purely disordered. All three compounds exhibit long-range magnetic order with rather small U moments.

Key Words: Uranium Intermetallics, Crystal Structure, Site Occupation, Magnetic

## Properties

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Among UTX (T = late transition metal and X = p-electron element) compounds that form in the hexagonal ZrNiAl structure, UCoAl is of particular interest due to its position on the verge of magnetic order. It has a paramagnetic ground state, but below 16 K even a small magnetic field (~1 T) applied along the c-axis can induce a metamagnetic transition [1, 2]. The magnetic moments are associated almost exclusively with the uranium atoms but the magnetic behavior is, nonetheless, very sensitive to substitutions of other transition metals for Co. For example, a ferromagnetic ground state develops with as low as 1% substitution of Ru for Co [3]. This points to the importance of the hybridization of 5f electrons with valence electrons of ligands, especially with d electrons of the T metals. Solid solutions,  $UCo_{1-x}T_xAl$  may provide a way to systematically study the role of hybridization in the development of magnetism in these UTX compounds. In the present work we have studied the properties of solid solutions in which 2/3 of the Co atoms were replaced with Ru, Pt and Rh.

Bulk studies indicate ferromagnetic ordering at about 40 K, 41K and 26 K for  $UCo_{1/3}Ru_{2/3}Al$ ,  $UCo_{1/3}Pt_{2/3}Al$  and  $UCo_{1/3}Rh_{2/3}Al$ , respectively [3-5]. We performed Neutron diffraction above and below the ordering temperature for each of the three compounds using the High Intensity Powder Diffractometer at the Intense Pulsed Neutron Source (Argonne National Laboratory). Low temperature data were taken at 20 K for  $UCo_{1/3}Ru_{2/3}Al$ , 14.5 K for  $UCo_{1/3}Pt_{2/3}Al$  and 16 K for  $UCo_{1/3}Rh_{2/3}Al$ . The data were

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analyzed with the Rietveld refinement package, GSAS, which allows for parallel refinements of crystal and magnetic structure [6]. At present we have assumed localized 5-f moments only.

The hexagonal ZrNiAl structure (see fig. 1) was confirmed for all three solid solutions. In this structure, there are two crystallographically distinct transition metal sites,  $T_1$  and  $T_2$ , with twice as many  $T_1$  sites as there are  $T_2$  sites. In order to properly interpret the development of magnetism with varying composition, it is necessary to know the distribution of transition metals over the two positions. Table 1 shows the structural parameters for each compound, including the occupation at each site, and the main features of the crystal and magnetic structures are presented below.

$UCo_{1/3}Ru_{2/3}Al$  forms in a chemically ordered structure. All Ru atoms reside on the  $T_2$  sites resulting in staggered layers of U-Co and Al-Ru planes. The presence of the (100) nuclear peak in the diffraction pattern, which is absent in the other two compounds, gives evidence of crystallographic ordering. While the (100) peak is not forbidden for this structure, only the ordered structure gives significant intensity. Our refinement of the magnetic structure suggests a ferromagnetic ground state for  $UCo_{1/3}Ru_{2/3}Al$  with very small U moments ( $\mu \sim 0.1\mu_B/U$ ) along the c-axis.

The distribution of Pt and Co atoms in  $UCo_{1/3}Pt_{2/3}Al$  is nearly statistical, with the Pt atoms exhibiting a slight preference for the  $T_1$  sites, i.e. about 60% of the Pt atoms occupy the  $T_1$  sites. As in  $UCo_{1/3}Ru_{2/3}Al$ , our magnetic refinement indicates that  $UCo_{1/3}Pt_{2/3}Al$  is a c-axis ferromagnetic but with slightly larger U moments ( $\mu \sim 0.4\mu_B/U$ ).

$UCo_{1/3}Rh_{2/3}Al$  has a purely statistical distribution of Rh and Co atoms on the two transition metal sites. Unlike  $UCo_{1/3}Ru_{2/3}Al$  and  $UCo_{1/3}Pt_{2/3}Al$ , for which neutron data are

in excellent agreement with bulk magnetization studies [3, 4], our best magnetic refinement for  $\text{UCo}_{1/3}\text{Rh}_{2/3}\text{Al}$  gives a moment of  $\sim 0.7 \mu_B/\text{U}$ : almost double the value observed in bulk studies [4]. Furthermore, our data seem to indicate anti-ferromagnetic, in-plane components to the magnetic moment.

The disparate magnetic ground states found in the three solid solutions may be explained in terms of the anisotropic nature of the 5f-ligand hybridization. We argue that preferential occupation of the transition metal sites by the Co and T atoms changes the ratio of in plane and out of plane contributions to the hybridization, which in turn affects both the formation of local U moments and the moment configuration.

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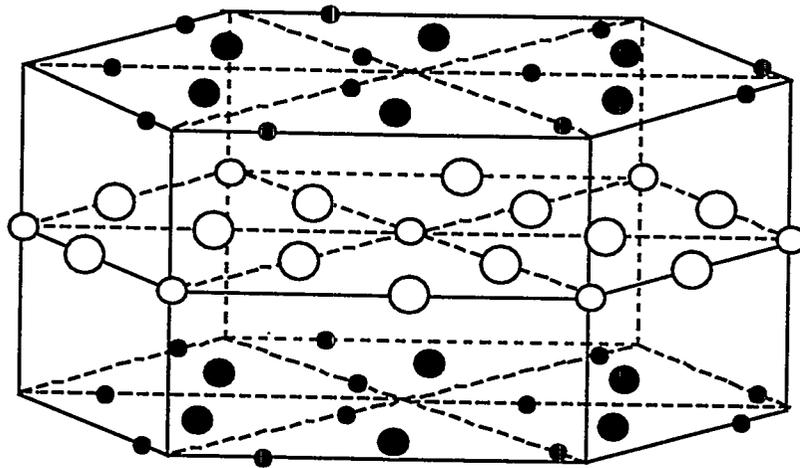
Table 1. Refined structural parameters for  $\text{UCo}_{1/3}\text{Ru}_{2/3}\text{Al}$ ,  $\text{UCo}_{1/3}\text{Pt}_{2/3}\text{Al}$  and  $\text{UCo}_{1/3}\text{Rh}_{2/3}\text{Al}$

at 300 K. Errors appear in the last significant digit for each parameter.

	Lattice Parameters (Å)	Atom	Position	x	y	z	Occupation
$\text{UCo}_{1/3}\text{Ru}_{2/3}\text{Al}$ $\chi^2 = 7.1$ $r\text{Wp} = 8.0\%$	$a = 6.847$	U	3g	0.58079	0	1/2	1
	$c = 3.981$	Co	1b	0	0	1/2	1
		Ru	2c	1/3	2/3	0	1
		Al	3f	0.23356	0	0	1
$\text{UCo}_{1/3}\text{Rh}_{2/3}\text{Al}$ $\chi^2 = 1.4$ $r\text{Wp} = 7.3\%$	$a = 6.861$	U	3g	0.57881	0	1/2	1
	$c = 4.002$	Co	1b	0	0	1/2	1/3
		Co	2c	1/3	2/3	0	1/3
		Rh <sub>1</sub>	1b	0	0	1/2	2/3
		Rh <sub>2</sub>	2c	1/3	2/3	0	2/3
		Al	3f	0.23725	0	0	1
$\text{UCo}_{1/3}\text{Pt}_{2/3}\text{Al}$ $\chi^2 = 4.1$ $r\text{Wp} = 5.9\%$	$a = 6.925$	U	3g	0.57640	0	1/2	1
	$c = 4.066$	Co	1b	0	0	1/2	0.128
		Co	2c	1/3	2/3	0	0.436
		Pt <sub>1</sub>	1b	0	0	1/2	0.872
		Pt <sub>2</sub>	2c	1/3	2/3	0	0.564
		Al	3f	0.23830	0	0	1

## Figure Captions

Fig. 1. Schematic representation of UTX compounds with the hexagonal ZrNiAl structure. The figure shows three unit cells.



Plane 1

○ U (3g)

○ T<sub>1</sub> (1b)

Plane 2

● T<sub>2</sub> (2c)

● Al (3f)