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Properties of UNiAlD_{2,1} and UNiAlH_{2,3}

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Neutron diffraction and bulk magnetic studies have been performed in UNiAlD_{2,1} and UNiAlH_{2,3}. Due to the lattice expansion and weakening of the 5*f*-ligand hybridization, UNiAlD_{2,1} orders magnetically at a much higher temperature than the isostructural UNiAl.

Keywords: Crystal structures, Heavy fermions, Magnetic order, Powder diffraction

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UNiAl crystallizes in the hexagonal ZrNiAl-type crystal structure consisting of U-Ni and U-Al basal plane layers. The inter-uranium spacing $d_{U-U} = 3.49 \text{ \AA}$ is proportional to the lattice parameter a (6.733 \AA), while the distance between the U-containing layers is given by c (4.035 \AA) [1]. UNiAl orders antiferromagnetically (AF) at 19.3K, and exhibits heavy fermion features [2]. Neutron-diffraction studies in single crystals revealed a complex arrangement of magnetic moments with $\mathbf{q} = (0.1, 0.1, 0.5)$ [3]. Hybridization of the uranium 5f states with the electron states of the ligands plays a crucial role in the magnetic properties of UNiAl. Hydrogenation/deuteration of the material changes its inter-atomic distances, thus affecting the degree of hybridization, and its magnetic properties. In this work we address the effect of hydrogenation/deuteration on the magnetic and structural properties of UNiAl.

UNiAlD_{2.1} and UNiAlH_{2.3} have been synthesized using a two-stage process. First the parent intermetallic UNiAl was arc-melted from the constituent elements under Ar atmosphere, and the phase purity was verified by means of X-rays diffraction. Secondly the material was crushed and hydrogenated, by means of activating the specimen in a vacuum of 10^{-6} Torr at 350°C for 1 hour, followed by exposure to D(H) at 55atm pressure. The amount of absorbed D(H) was determined by monitoring the decrease of pressure in a calibrated volume. The magnetic susceptibility measurements were performed with a Faraday balance.

The temperature dependence of the magnetic susceptibility of UNiAlD_{2.1} and UNiAlH_{2.3} are displayed in Figure 1, which shows that both samples order AF with $T_N = 94\text{K}$ for UNiAlD_{2.1} and $T_N = 99\text{K}$ for UNiAlH_{2.3}. These T_N values are much higher than the Néel temperature (19.3K) of the parent compound UNiAl. We have not determined if the difference in ordering temperature is due to slightly different H(D)-content. Above 100 K, both the deuteride and the hydride compounds show modified

Curie-Weiss behavior with a temperature independent term χ_0 , $\mu_{\text{eff}} = 2.42 \mu_{\text{B}}/\text{f.u.}$ and $2.43 \mu_{\text{B}}/\text{f.u.}$, $\Theta_{\text{P}} = -42 \text{ K}$ and -50 K , $\chi_0 = 7 \cdot 10^{-9} \text{ m}^3/\text{mol}$ and $8 \cdot 10^{-9} \text{ m}^3/\text{mol}$, respectively.

Because of the higher coherent cross section of D compared to H, neutron diffraction data was obtained only for the UNiAlD_{2.1} specimen. The data was collected on the powder diffractometers HIPD and SEPD at IPNS. The crystal structure at room temperature was determined from SEPD data, and analyzed with GSAS [4]. The best structure refinement indicates that UNiAlD_{2.1} preserves the ZrNiAl structure and that D occupies 3 interstitial positions as shown in Table I. Compared to UNiAl before hydrogenation, the lattice is 6.65% elongated along the *a* axis and 1.2% contracted along the *c* axis. Our findings are similar, but not equal, to those reported on ref. 5.

Additional magnetic intensities at 3.29 Å and 4.01 Å, which cannot be indexed in the ZrNiAl structure, are observed below 100K (see inset of Figure1). Because the latter reflection corresponds to d_{hkl} slightly higher than the lattice parameter *c*, an AF structure involving a simple cell doubling can be excluded. Also the magnetic reflections could not be indexed assuming the **q**-vector reported for the UNiAl [3].

While we were not able to unambiguously determine the propagation vector for the magnetic ordering on the basis of these two peaks only, some conclusions can be drawn. We may argue that hydrogenation/deuteration of UNiAl leads to a reduction of hybridization effects, and consequently higher T_{N} of UNiAlD_{2.1} and UNiAlH_{2.3}.

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Tables and Captions:

Table I – Structural parameters of UNiAlD_{2,1}, with $a=b=7.18069(8)\text{\AA}$, $c=3.98849(7)\text{\AA}$;

$R_{wp} = 9\%$, $R_p = 6.5\%$; $\chi^2 = 4$.

	Site	x	y	z	n
U	3g	0.666(2)	0	½	1
Ni ₁	1b	0	0	½	1
Ni ₂	2c	1/3	2/3	0	1
Al	3f	0.333(2)	0	0	1
D ₁	3g	0.332(3)	0	½	0.95(2)
D ₂	2e	0	0	0.033(1)	0.80(1)
D ₃	4h	1/3	2/3	0.406(1)	0.337(5)

List of Figure Captions:

Figure1: Temperature dependence of the magnetic susceptibility of UNiAlD_{2.1} (▲) and UNiAlH_{2.3} (■) taken at $\mu_0 H = 5$ T. For clarity some experimental points were omitted above 100K. Inset: Difference spectrum of neutron-diffraction data for UNiAlD_{2.1}, with magnetic peaks marked by *.

Figure 1:

