XMCD studies of UH_3-based ferromagnets: $(UH_3)_{1-x}Mo_x$ and $(UH_3)_{1-x}Zr_x$.

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Understanding the interaction of uranium and hydrogen is of great importance, e.g. in the field of nuclear energy safety – the formation of uranium hydrides affects the mechanical integrity of uranium, forming a very fine pyrophoric powder. On the other hand, hydrides, in general, provide a tool for investigating the effects of lattice expansion (it can be viewed as application of negative pressure), which, in the case of uranium, might lead to formation of *5f* moments and magnetic ordering [1].

UH_3 is the first reported *5f* ferromagnetic system; synthesized more than half a century ago [2]. UH_3 exists in two structural modifications: the ferromagnetic β-UH_3 ($T_C \approx 165$ K) and the thermodynamically unstable α-UH_3. Attempts to stabilize the α-UH_3 phase upon alloying successfully yielded two stable compounds [1, 3]: $(UH_3)_{1-x}Zr_x$ and $(UH_3)_{1-x}Mo_x$. The Zr based one keeps the α-UH_3 structure whereas the Mo hydride is found to be a nanocrystalline material featuring β-UH_3 crystal structure (see fig.1). Despite their different crystal structures, both hydrides have quite similar basic bulk magnetic characteristics. Both compounds are ferromagnetically ordered below T_C slightly higher compared to the stable β-UH_3 and their macroscopic physical properties have been thoroughly investigated [4].

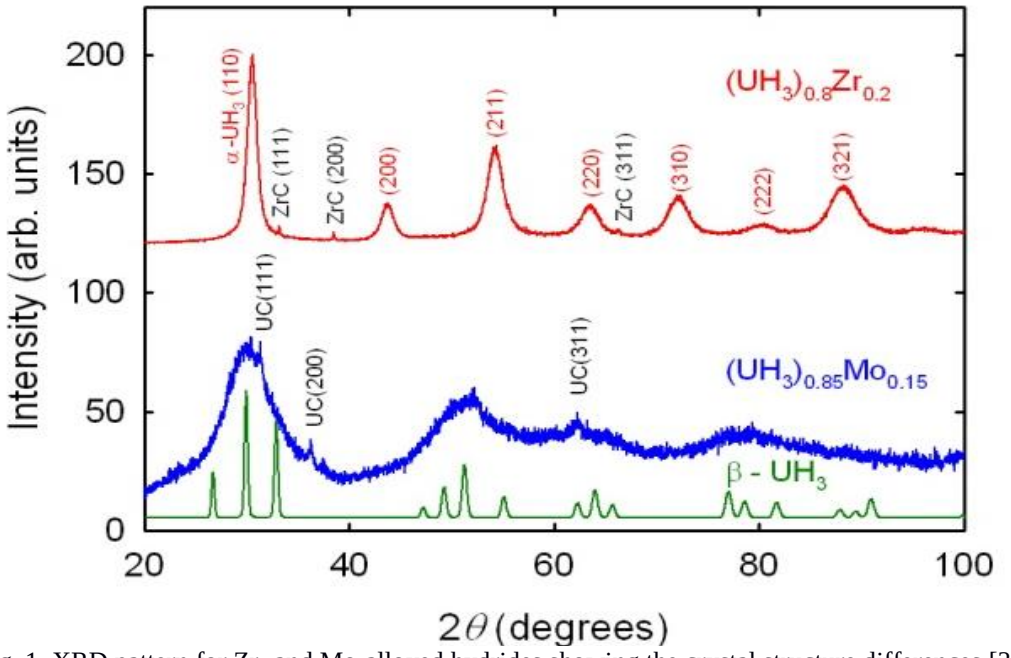

Fig. 1. XRD pattern for Zr- and Mo-alloyed hydrides showing the crystal structure differences [3].

Here we present element selective magnetic measurements of $(UH_3)_{1-x}Mo_x$ and $(UH_3)_{1-x}Zr_x$ powders using X-ray Magnetic Circular Dichroism (XMCD) at the $M_{4,5}$-edges of Uranium and at the $L_{2,3}$-edeges of Zirconium and Molybdenum. Normalized Uranium XMCD spectra together with the corresponding XANES spectra are reproduced in Fig. 2. Using the so called magneto-optical sum rules we were able to determine the spin (~-0.76 μ_B, ~-0.57 μ_B) and orbital (~1.72 μ_B , ~1.4 μ_B) moments of Uranium in the Zirconium and Molybdenum alloyed samples, respectively. These results are in a rather good agreement with bulk measurements. On the other hand, theoretical calculations also predict large spin and orbital *5f* moments of Uranium which are antiparallel, and an induced sizeable moments on the alloying atoms (Mo, Zr) [5].

XMCD measurements revealed several differences in the magnetic properties between these two hydrides. Whereas an ordered magnetic moment was indeed found on Mo atoms, Zr atoms seem to do not acquire a moment (the XMCD signal is below the present detection limit). The measured magnetic moments on Uranium atoms in the samples also differ by about ~0.2 μ_B and the unoccupied part of the *5f* band appears to be narrower for the Mo alloyed sample.

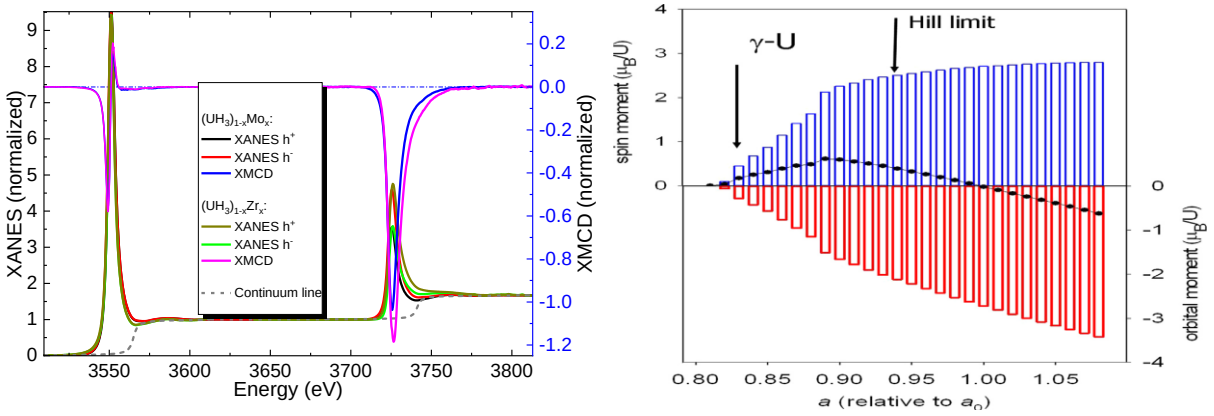

Fig. 2. Left – XANES and XMCD measurements of $(UH_3)_{1-x}Mo_x$ and $(UH_3)_{1-x}Zr_x$ measured at uranium's $M_{4,5}$ absorption edges. Right – Variations of U spin and orbital moments in α-UH_3, calculated by means of the FPLO method, as a function of the lattice parameter (a) normalized to the experimental value a_0. The black dots indicate the total U moments [taken from 5].

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