| ESRF | **Experiment title:** Synthesis of novel uranium hydrides up to 100 GPa: Structural refinement and magnetic signature of superconductivity | **Experiment number**: HC-3947 |
|---|---|---|
| **Beamline**: ID27 | **Date of experiment**: from: 16/11/2018 to: 20/11/2018 | **Date of report**: 05/03/2019 |
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## Report:

Novel polyhydrides of metals are predicted ubiquitous under high pressure and hold great promise as a class of high critical temperature ($T_c$) superconductors. Few experiments only have tested the large corpus of predictions and already striking results have been observed, such as 200 K-superconductivity in compressed $H_2S$ at 150 GPa [1] or the stability of slabs of atomic hydrogen in $FeH_5$ at 130 GPa, with a calculated $T_c$ of 50 K [2,3]. However, up to now, there is a lack of simultaneous structural refinement with the associated high $T_c$ superconductivity measurements on a pristine superhydride sample, since such study has proven to be very challenging above 100 GPa. Interestingly, a recent work proposed that uranium superhydrides could be high $T_c$ superconductors below the megabar [4], allowing both structural study and magnetometry. The aim of this proposal was to synthesize uranium superhydrides ($UH_7$ and $UH_8$, stable below 80 GPa) by direct reaction of uranium embedded in hydrogen. Such process allows one to obtain a fine quality powder, which is mandatory to refine the structure of these uranium polyhydrides and to characterize their superconducting properties by SQUID magnetic measurements of the Meissner effect, using our recently developed miniature DAC (mDAC) [5]. Being able to synthesize and fully study U-H systems from both the structural and the magnetic point of views is a unique opportunity to fully test the theoretical prediction of high $T_c$ superconductivity in superhydrides.

We therefore performed a detailed structural study of the uranium hydrides synthesized directly under pressure by laser heating mixtures of $U+H_2$ up to 100 GPa. A major goal was to investigate the U-H system to verify *ab initio* structural and stability predictions, and thus pave the way for future magnetic measurements in a SQUID magnetometer. We have carried out three experiments at 300 K, in different pressure ranges. The sample was annealed using a YAG laser, at various pressures (see Table 1). The temperature reached was about 1300 K. The pressure was measured using either a gold volumic gauge. The volume was measured using angular-dispersive x-ray diffraction. The conditions of the experiments are summarized in **Table 1**.

| Name | Sample | Culet diameter (μm) | Pressure range (GPa) | T (K) | P laser annealing (GPa) |
|---|---|---|---|---|---|
| Run 1 | $U+H_2$ | 300 | 9 – 62 – 25 | 300 | 50, 57, 60, 62 |
| Run 2 | $U+H_2$ | 300 | 10 – 47 – 25 | 300 | 16, 21, 25, 29, 34, 36, 47 |
| Run 3 | $U+H_2$ | 150 | 9 – 96 – 0 | 300 | 73, 80, 93 |

**Table 1:** Conditions of the three experimental runs.

The measurements of *V* vs. *P* for uranium hydrides at 300 K are plotted in **figure 1**. Uranium spontaneously forms the $UH_3$ hydride in the gas loading apparatus, at 1400 bars. We successfully synthesized the $UH_7$ and $UH_8$ hydrides, predicted to be superconducting above 50 K. We did not observe neither $UH_5$ nor $UH_6$, though predicted stable by *ab initio* calculations. The synthesized hydrides were obtained in pure phases, and with a complete stoichiometry, unlike the work by Kruglov et al. [4]. Our rigorous protocol allowed us to accurately determined the stability and metastbility domains of $UH_7$ and $UH_8$, with $UH_7$ being stable around 40 GPa and thus being a promising candidate for further SQUID magnetometry experiments. $UH_8$ appears to be metastable down to 55 GPa, far from the metastability at ambient pressure predicted by calculations. Nonetheless, it is the highest stoichiometry ever synthesized at such low pressure.

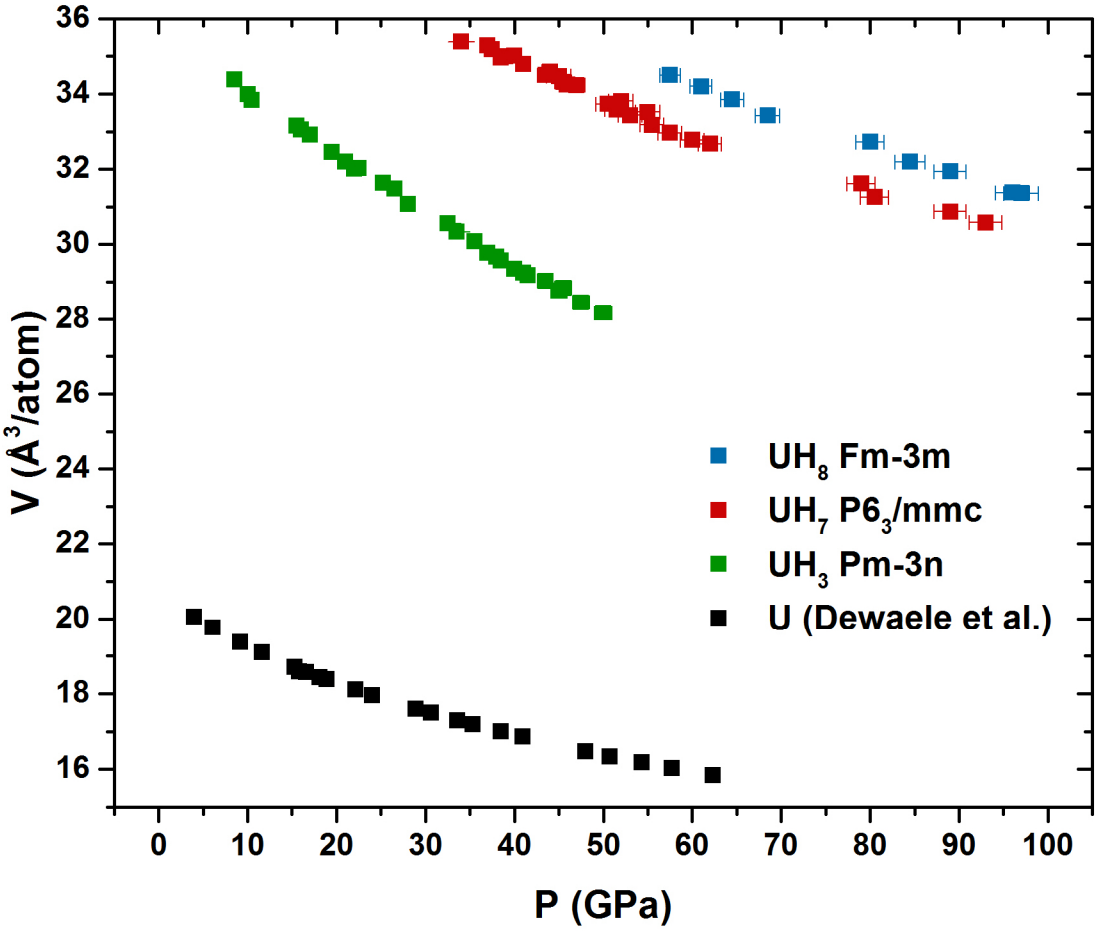


**Figure 1**: Equations of State of $UH_3$, $UH_7$ and $UH_8$, together with data from ref 6.

**References:**

[1] A. P. Drozdov *et al.*, Nature 525, 73-76 (2015)
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