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Synthesis of UH_7 and UH_8 superhydrides: Additive-volume alloys of uranium and atomic metal hydrogen down to 35 GPa

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UH_7 and UH_8 have been synthesized in a pure form by direct reaction of uranium with excess hydrogen in a laser-heating diamond anvil cell up to 96 GPa. The sequence UH_3, UH_7, and UH_8 is observed under increasing pressure using synchrotron x-ray diffraction. UH_7 is stable above 35 GPa and UH_8 above 87 GPa. The crystal structures of these two superhydrides correspond to the *ab initio* predicted structures with their cagelike sublattice of atomic hydrogen. Their volume expansion per H atom falls on the hypothetical atomic metal hydrogen compression curve. A critical survey of the literature data for other recently observed superhydrides enables one to generalize such a trend in the partial volume occupied by the hydrogen atom. Hence, superhydrides made only of atomic hydrogen can be considered additive-volume alloys of atomic metal hydrogen and the metal. With its lowest equilibrium pressure, UH_7 is the superhydride for which the concept of chemical precompression to stabilize an atomic metal hydrogen analog works the most effectively so far.

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I. INTRODUCTION

High pressure promotes the formation of polyhydrides with unusually high hydrogen-to-metal ratios (H:M). This novel class of compounds, called superhydrides and typically with H:M $\geqslant 5$, has been discovered by searching how atomic metal hydrogen could be doped with an element to lower its synthesis pressure [1]. Superhydrides are currently the subject of great interest because some possess superconducting properties analogous to the ones of atomic metal hydrogen, thus forming a novel class of high critical temperature (T_C) superconductors [2]. A record T_C has been measured recently in LaH_{10} [3,4], about 250 K. The most remarkable properties are expected when the hydrogen component forms a dense sublattice of H units only, somehow approaching atomic solid hydrogen doped with a metal. Indeed, the LaH_{10} compound has a very interesting cagelike network of hydrogen atoms with the La atom embedded in the cages [5]. Very similar hydrogen cagelike structures have been predicted ubiquitous for binary alkaline-earth or rare-earth metal superhydrides [6–9], with H:M ratios in the 6 to 10 range. Some have recently been synthesized, such as YH_6 and YH_9 [10,11], CeH_9 [12,13], and PrH_9 [14]. Actinides have also been observed to form superhydrides with cagelike atomic H structures, such as ThH_{10} [15] and UH_7 or UH_8 [16]. Interestingly, the first observed superhydride composed of H atoms has been FeH_5, which exhibits a layered structure made of atomic hydrogen slabs [17].

So far, most of the superhydrides have been synthesized at pressures exceeding 100 GPa. On the other hand, UH_7 and UH_8 have been predicted to be stable below 100 GPa, and even as low as 22 GPa for UH_7 [16]. Recovering UH_7 metastably at ambient pressure might thus be possible. The first experiment aimed at synthesizing superhydrides in the U-H system has observed UH_7 down to about 30 GPa but the observed possible UH_8 compound seemed to have a too large H:M ratio [16]. Besides, the x-ray diffraction (XRD) patterns reported were those of multiphases, indicating that the excess hydrogen conditions might not have been well controlled during the synthesis. Here we take a different look at the synthesis of superhydrides in the U-H system. The first aim of the present study was to obtain uranium superhydrides in pure form and to unambiguously determine the pressure domains over which each single-phase polyhydride can be synthesized in excess hydrogen in a laser-heated diamond anvil cell. The second aim was to accurately measure their associated volume per H atom to determine whether or not these uranium superhydrides could be considered as additive-volume alloys between atomic metal hydrogen and uranium, as observed for FeH_5 [17]. To find a general trend, this question is then further investigated by analyzing the expansion volume per H atom for all the superhydrides synthesized so far which have a dense atomic hydrogen sublattice.

II. EXPERIMENTAL METHODS

The sample configuration and the synchrotron XRD measurements were similar to those used in our previous studies of compounds formed in the (Fe, S, Cr, Pd)-H systems at high pressures [18–21]. Uranium samples were prepared (the surface layer of a bulk uranium piece was first scrubbed using a diamond file to remove the UO_2 layer) and loaded in the diamond anvil cell (DAC) inside a glovebox under argon atmosphere. A sample of about 10–15 μm was then selected and placed in the experimental cavity. The cavity was then

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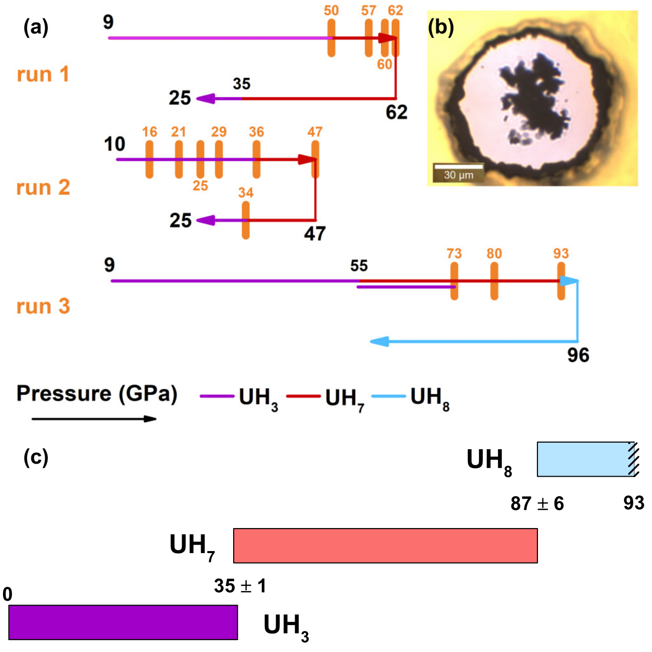

FIG. 1. (a) Pressure-temperature synthesis history for runs 1, 2, and 3. The orange bars mark the laser-heating pressures (with T about 1300 K). The pressure arrow has a color scale to indicate the compound observed. (b) Picture of the sample. A piece of pure uranium is surrounded by dense H_2 in excess. The rhenium gasket has a gold coating of 700 Å to prevent hydrogen diffusion. A gold piece is used as a pressure gauge. (c) Pressure domains of the synthesis of the three observed polyhydrides β-UH_3, hcp-UH_7, and fcc-UH_8, from laser-heating U in excess hydrogen in a DAC.

sealed, and re-opened only when under H_2 atmosphere in the high pressure hydrogen loading apparatus. Hydrogen loading was made at 1400 bars. The uranium sample reacted with hydrogen during the loading cycle. The volume increased dramatically and one could see the U sample fragmentation resulting from hydrogen absorption, as shown in Fig. 1. Three runs were carried out, and YAG laser-heating was performed at several pressures to kinetically favor the transition to the stable compound. The temperature reached about 1300 K (which was the minimal detectable temperature). Diamond anvil culets were 300 μm in diameter for runs 1 and 2 and 100 μm for run 3. A typical sample configuration is shown in Fig. 1, with a large excess of hydrogen surrounding the uranium sample. Doing so, we are sure that the hydride with the highest H:U ratio stable is synthesized in pure phase. The XRD data were collected at the ID27 beamline of the ESRF using 0.3738 Å wavelength. At a given pressure we took several XRD patterns, with pressure measured in between using the gold piece. This procedure allowed us to ensure that the pressure was stable and to measure very accurate $V(P)$. XRD patterns were then analyzed using DIOPTAS [22], the FULLPROF software suite [23], and the XRDA suite [24]. Uncertainty in volume is about ± 0.07 Å^3. The pressure was measured using the equation of state of a small gold piece [25]. The uncertainty in pressure is about 2%.

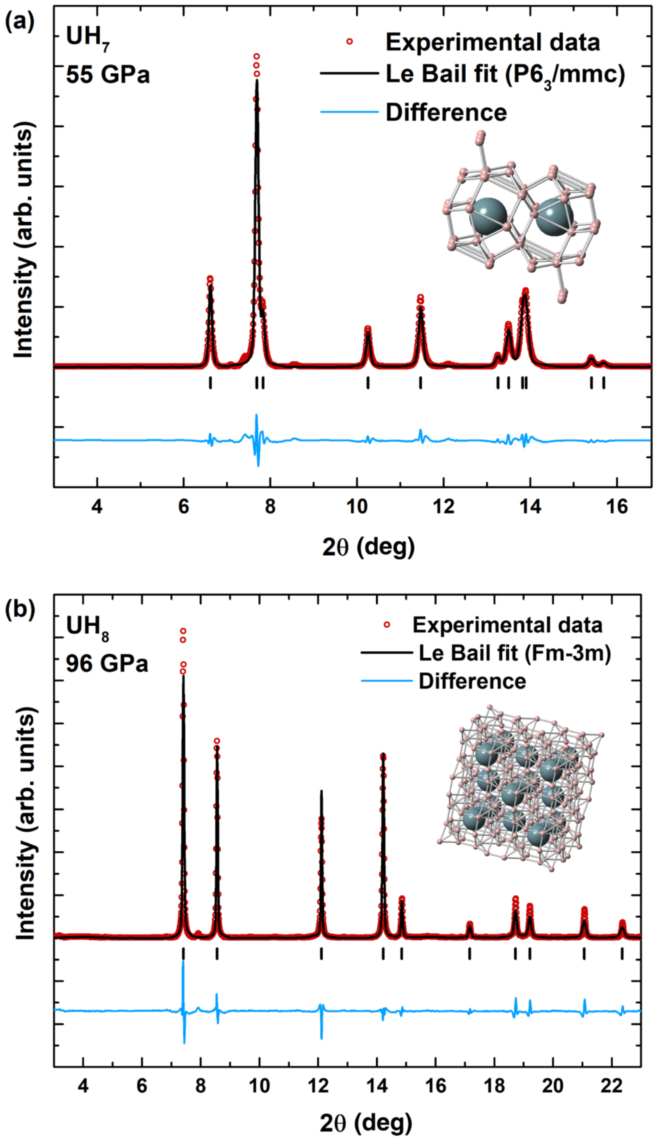

FIG. 2. (a) XRD pattern obtained at 55 GPa, fitted with the $P6_3/mmc$ space group with $a = 3.749(6)$ and $c = 5.473(2)$ ($\chi^2 = 0.8814$). Unfitted weak reflexions come from hydrided rhenium and gold. (b) XRD pattern obtained at 96 GPa, fitted with the $Fm\bar{3}m$ space group with $a = 5.008(9)$ ($\chi^2 = 1.748$). The unfitted weak reflexions come from hydrided rhenium. The cagelike sublattice of the hydrogen atoms is shown in the unit cell of both structures.

III. THE SEQUENCE OF URANIUM POLYHYDRIDES UNDER PRESSURE

Uranium is known to react spontaneously with hydrogen at ambient conditions to form UH_3 [26]. Two phases have been discovered (α and β), although the bcc α phase remains elusive [27]. In the present study, only β-UH_3 was observed. The β phase has an U sublattice of the A15-type (also known as β-W) with the H atoms in the distorted tetrahedral sites. In run 1, β-UH_3 remained the only phase up to 50 GPa. After laser heating the sample at 50 GPa, a complete transformation to the hcp-UH_7 compound is observed, as shown in Fig. 2(a). The hcp crystal structure of the metal sublattice of UH_7, predicted in Ref. [16], enables a very good fit of the diffraction

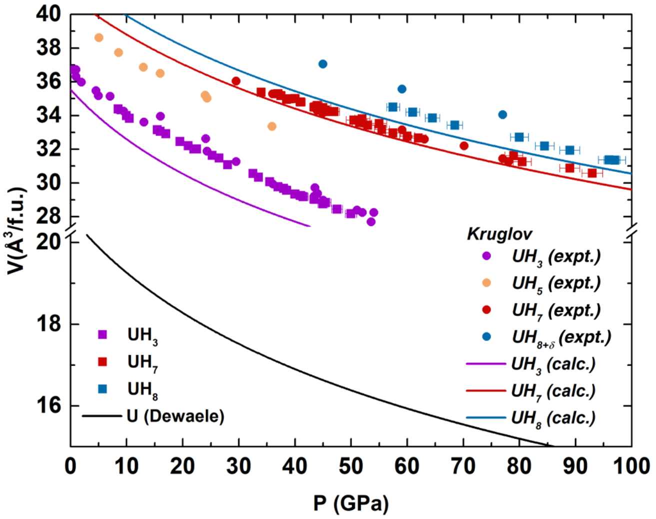

FIG. 3. Experimental volume per formula unit as a function of pressure for UH_3 (purple squares), UH_7 (red squares), and UH_8 (blue squares). Our data are plotted together with the data from Ref. [16] for UH_x hydrides: UH_3 (purple circles), UH_5 (yellow circles), UH_7 (red circles), and $UH_{8+\delta}$ (blue circles). If not plotted, error bars are smaller than the symbols size. The solid color lines are the DFT calculations of Ref. [16], while the solid black line is the EOS of uranium from Ref. [30].

pattern, and the associated unit cell volume is very close to the one calculated for UH_7. When the pressure was further increased and the sample was laser heated at 57, 60, and 62 GPa, only hcp-UH_7 was observed. Upon pressure release, the UH_7 sample transformed spontaneously back into the β-UH_3 phase around 34 GPa.

In run 2, laser heating at different pressures allowed us to check the β-UH_3 stability up to 36 GPa. The sample was heated at 16, 21.5, 25.5, and 29 GPa without any change in the crystal structure. At 36 GPa, after the sample was laser heated, the whole β-UH_3 phase transformed to hcp-UH_7. No further structural change of the sample was observed under laser heating at 47 GPa. Pressure was then released and at 34 GPa, the β-UH_3 Bragg reflections reappeared under laser heating. At 25 GPa, a pure β-UH_3 XRD pattern was measured.

In run 3, hcp-UH_7 mixed with UH_3 spontaneously appeared under compression at 55 GPa. After laser heating the sample, a pure hcp-UH_7 phase was obtained. A second laser heating at 80.5 GPa confirmed the stability of hcp-UH_7. Yet, at 93 GPa, laser heating produced a structural transformation into a new phase, identified, by looking at the predicted uranium polyhydrides [16], as pure fcc-UH_8. As shown in Fig. 2(b), a Le Bail fit with the $Fm\bar{3}m$ space group fits our data nicely. Moreover, the volume expansion compared to pure U matches a H:U ratio of 8, as can be seen in Fig. 3. After reaching 96 GPa we started decompression, and the sample remained single-phase fcc-UH_8 down to 58 GPa. The next decompression step resulted in a sudden pressure drop down to 5 GPa, after which only β-UH_3 could be observed.

Uranium transforms into the sequence of compounds β-UH_3, hcp-UH_7, and fcc-UH_8, when compressed in excess hydrogen. Although in calculations [16] various other polyhydrides have been predicted stable on the convex hull, only the three hydrides UH_3, UH_7, and UH_8, which in turn correspond to a clear marked minimum enthalpy upon increasing pressure, were observed. The structures observed are those predicted by *ab initio* calculations [16]. That is further evidence of the reliability of *ab initio* structure/compound search of superhydrides under pressure. The unit cell of UH_7 and UH_8, hcp $P6_3/mmc$ and fcc $Fm\bar{3}m$, respectively, are represented in Fig. 2, following the Wyckoff positions for the hydrogen atoms, as given in Kruglov's work [16]. Both structures are made of a dense atomic hydrogen cagelike sublattice.

The predicted UH_5 compound was not observed in the present experiment. Essentially, two reasons can be invoked to explain the discrepancies between two similar laser-heating DAC experiments. First, a possible contamination of the sample, since in Kruglov's experiment naturally oxidized uranium samples were loaded in H_2 medium, whereas great care was paid here to load unoxidized U samples. Second, an insufficient reserve of hydrogen, since the uranium sample was here always compressed in a large excess of hydrogen, whereas that condition was not proven in Ref. [16]. That could explain the synthesis of various metastable phases at the same time. Indeed, the evidence of $UH_{8+\delta}$ and UH_5 was obtained from weak peaks in a complex multiphases diffraction pattern in Ref. [16]. Also, looking at Fig. 3, it is easily seen that there is a large systematic uncertainty in the P-V data points of UH_3 measured in Ref. [16], with a pressure uncertainty larger than 3 GPa that can explain the difference of the stability pressure of UH_7 between Ref. [16] and the present work.

The stability domains in excess hydrogen and under laser-heating conditions for UH_3, UH_7, and UH_8, shown in Fig. 1, could be deduced from the synthesis paths followed. UH_3 remains the most stable phase up to 35 GPa. The stability equilibrium pressures of both UH_7 and UH_8 are observed higher than estimated by *ab initio* calculations [16]: For UH_7, 35 GPa instead of 22 GPa and for UH_8, 87 GPa instead of 52 GPa. Such pressure difference between *ab initio* predictions and experimental observation of transition pressures is not unusual and it is generally ascribed to approximations in the exchange-correlation functional, especially here due to the U atoms' $5f$ electrons. Also here, thermal contributions could explain the stability pressure difference between the 0 K-*ab initio* calculations and the ~300 K synthesis. In lower hydrides, a dT/dP slope of the order of 100 K/GPa was reported [28]. In our three runs, samples spontaneously transformed back to UH_3 upon pressure release. UH_7 and UH_8 could be decompressed as single phases down to 34 and 57 GPa, respectively. None of these two uranium superhydrides could be recovered at ambient conditions.

IV. COMPRESSION CURVES AND VOLUMES PER H ATOM

The compression data $V(P)$ for UH_3, UH_7, and UH_8 are plotted in Fig. 3. Compared to Kruglov's measurement [16], the present volume of UH_3 is slightly smaller, the one of UH_7 is in good agreement with their data, and the one of UH_8 is significantly smaller. The present volume for UH_8 is now coherent with a H:U ratio of 8. Besides, neither the so-called $UH_{8+\delta}$ nor the UH_5 compounds were observed in

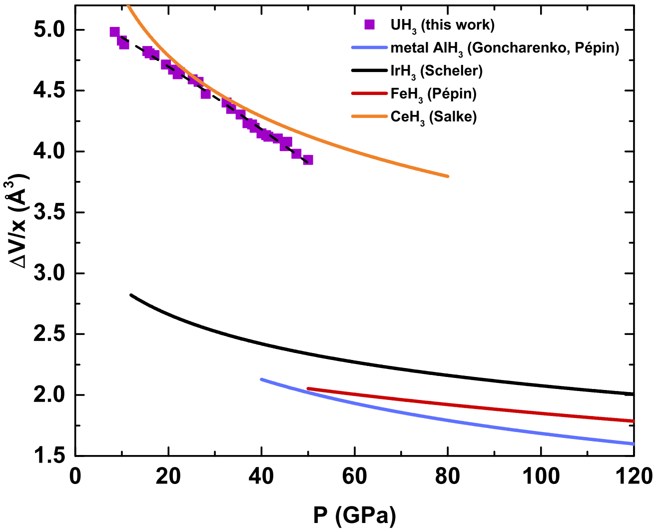

FIG. 4. Volume per H atom vs pressure for UH_3 (purple squares), together with data from Refs. [12,20,33–35]: FeH_3 (red line), metal AlH_3 (blue line), A15-CeH_3 (orange line), and IrH_3 (black line). The dashed line is the average evolution of $\Delta V/x$ in UH_3.

our experiments. The compression data are also compared to *ab initio* calculations: Experimental volumes are 6% greater, 1% greater, and 2% greater than the calculated ones for UH_3, UH_7, and UH_8, respectively. To describe precisely the properties of uranium based compounds, the correlation of the U atoms' $5f$ electrons sometimes needs to be explicitly treated. Yet, it is not taken into account in Kruglov's calculations [16], which could explain the larger volume deviation for UH_3. For uranium superhydrides, it was recently shown that the influence of $5f$ electronic correlation effects should be small [29].

The volume per H atom in UH_x, $\Delta V/x$, is calculated at a given pressure, by subtracting pure uranium volume of Ref. [30] from the measured volume of UH_x and dividing by x the number of H atoms. $\Delta V/x$ vs pressure for UH_3 is plotted in Fig. 4. The volume expansion of the U lattice is due to the uptake of H atoms in the tetrahedral interstitial sites of the cubic β-W-like uranium sublattice. As seen in Fig. 4, $\Delta V/x$ in UH_3 is much larger than the typical volume expansion of 2.2–3.2 Å^3 for interstitial hydrides [31]. Such a large value in UH_3 has been explained by a significant electron density localized near the proton, due to the overlapping reduction in the $5f$ electrons between nearest U atoms from the hydrogen uptake [32]. $\Delta V/x$ decreases linearly with pressure, as observed previously in interstitial hydrides such as CrH [19], FeH [20], or PdH [21], though with a steeper slope. $\Delta V/x$ in UH_3 is also larger than in other metallic MH_3 hydrides (i.e., made of H entities only) already measured such as FeH_3 [20], metal AlH_3 [33,34] (measured on decompression after the transition to the cubic metallic phase at 107 GPa), or IrH_3 [35], but is similar to the high pressure A15-CeH_3 phase [12]. β-UH_3 and A15-CeH_3 share the same A15-type structure and $5f$-electrons correlation influence, while metal AlH_3 has the same structure as α-UH_3. It is interesting to note the very large dispersion of the value of $\Delta V/x$ in these various metallic MH_3 hydrides. The volume per H atom in UH_7 and UH_8 is smaller than for UH_3 and larger than for interstitial hydrides. In the next section it is shown to fall close to the atomic metal hydrogen compression curve, which is seemingly a universal trend for the superhydrides synthesized so far.

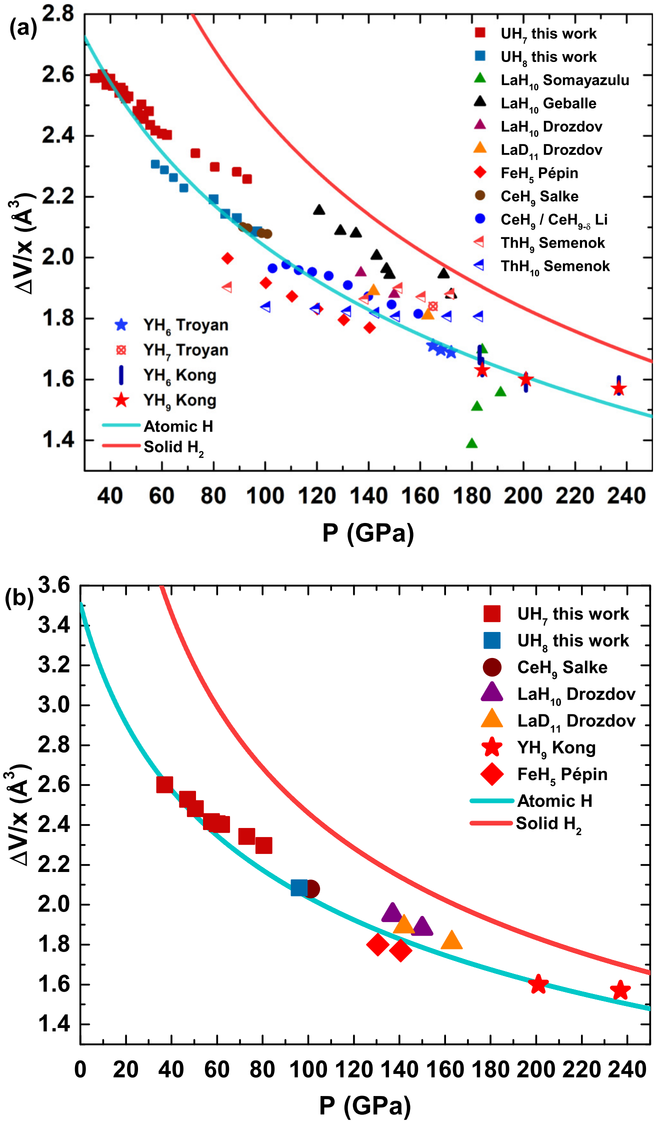

FIG. 5. (a) Volume per H atom for the current experimental corpus of superhydrides having a dense hydridic sublattice. Symbols labeled in the figure correspond to: Present data for UH_7 and UH_8; Refs. [3–5] for LaH(D)$_x$; Ref. [17] for FeH_5; Refs. [12,13] for CeH_x; Ref. [15] for ThH_x; and Refs. [10,11] for YH_x. The solid cyan curve is the compression curve calculated for hypothetical atomic metal hydrogen (with Vinet parameters $V_0 = 3.50(7)$ Å^3, $K_0 = 79.3(1)$ GPa, and $K_0' = 3.28(8)$ [17]) and the red plain line is the extended compression curve of solid H_2 (with Vinet parameters $V_0 = 13.85$ Å^3, $K_0 = 0.191$ GPa, and $K_0' = 7.039$ [36]). (b) Same plot as in (a) after selecting the data, as explained in the paper.

V. ADDITIVE-VOLUME ALLOYS OF ATOMIC METAL HYDROGEN AND METALS

In the study of FeH_5 [17], it was shown that the volume per H atom was very close to the volume per atom in atomic metal hydrogen. The volume of FeH_5 could then be seen as the ideal-mixing-volume of Fe and atomic metal hydrogen at a given pressure. We can wonder whether this observation is fortuitous or if superhydrides in general can be considered as additive-volume alloys of the metal and atomic metal hydrogen. The present data obtained on UH_7 and UH_8 are especially valuable since they are in the 35–96 GPa pressure range, which has not been covered previously in other studies of superhydrides. To answer this question, the $\Delta V/x$ data points for UH_7, UH_8, and all the superhydrides synthesized so far with a H:M $\geqslant 5$, in the La-H system [3–5], Ce-H system [12,13], Y-H system [10,11], Th-H system [15], and Fe-H system [17] are plotted in Fig. 5(a). At first glance, a significant scatter around the atomic metal hydrogen compression curve does exist. But part of it should be ascribed to a poor knowledge of the exact H:M ratio in these synthesized superhydrides. That is clear in the case of LaH_{10} which has been synthesized independently by three groups and for which $\Delta V/x$ is seen to deviate positively and negatively from the atomic metal hydrogen volume. In the case of ThH_{10} [15], $\Delta V/x$ seems almost independent of pressure, which is rather surprising. Two reasons could be invoked. First, the H:M ratio can vary when the superhydride is compressed in its metastable domain. Second, if the sample is not laser heated in a real excess of hydrogen, a mixture of phases and intermediate stoichiometries could be obtained. This phenomenon seems to be favored by the use of NH_3BH_3 pressure medium as the hydrogen reserve. Most of the $\Delta V/x$ data points deviating from the atomic metal hydrogen compression curve have been measured for superhydrides synthesized using the NH_3BH_3 pressure medium, such as LaH_{10} in Refs. [4,5] and ThH_{10} in Ref. [15]. A poorer diffusion and also lower hydrostatic conditions than in pure hydrogen are indeed expected when using NH_3BH_3 as a pressure medium. Besides, when no laser heating is performed, it is difficult to ensure that the most stable compound has been obtained, as for CeH_9 in Ref. [13] and YH_6 in Ref. [10].

In Fig. 5(b) we have consequently selected only the $\Delta V/x$ data points which have been measured just after laser heating the metal in excess hydrogen. In all cases, volume measurements were obtained from XRD patterns of a single phase (sometimes with minor impurities). Thus, one can be confident that the superhydride, with the highest H:M ratio and stable at the synthesis pressure, has been made (namely LaH_{10} and LaD_{11} [3], FeH_5 [17], CeH_9 [12], and YH_9 [10]). In Fig. 5(b), all these $\Delta V/x$ data points are now in good agreement with atomic metal hydrogen volume. In the 150 GPa pressure range and above, part of the small scatter could come from the uncertainty in the pressure determination.

VI. CONCLUSION

Figure 5(b) offers a compelling heuristic evidence that superhydrides based on atomic hydrogen only, and with a H:M $\geqslant 5$, can thus be considered as additive-volume alloys between a metal and atomic metal hydrogen. Those superhydrides are good candidates to observe some remarkable properties predicted for atomic metal hydrogen [37]. The measurements of very high critical temperatures of superconductivity in LaH_{10} at 250 K [3,4], in YH_6 at 220 K [10,11], and in ThH_{10} at 160 K [15] are already the twins of the predicted ambient temperature superconductivity of atomic metal hydrogen. The feasibility of synthesizing UH_7 and UH_8 in pure phases, and their low pressure stability, offer a unique opportunity to investigate other effects, such as a high protonic mobility [38]. That should now motivate future studies.

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