ESRF	**Experiment title:** Stability and structures of lead hydrides under pressure	**Experiment number**: HC-3680
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Report:

It is now a proven fact that hydrogen solubility in metals is drastically increased under pressure. Hydrides with nontraditional stoichiometry are predicted for most metals and few have now been observed, such as LiH_6 [1], H_3S [2] or FeH_5 [3]. Many of these superhydrides are predicted to be high temperature superconductors and the observation of a record critical temperature (T_C) of 200 K in compressed H_2S has attracted a lot of interest [4]. These systems could also possess other intriguing properties associated to their dense hydrogenic sublattices. PbH_4 and PbH_8 have been predicted stable above 130 GPa [5,6]. The lead-hydrogen system is special because it combines one of the heaviest elements (Pb) to the lightest (H) with the implication of a clear time scale dynamic separation between the two constituents. As a consequence, diffusive or liquid-like ground states of the H sublattice could exist [5]. Pb hydrides are also candidates as high-T_C superconductors, with the electron-phonon coupling due essentially to the H atoms in a molecular H_2 form.
The aim of this proposal was to investigate the stability and structures of lead hydrides under pressure. We therefore performed a detailed structural study of the lead hydrides synthesized directly under pressure by laser heating mixtures of $Pb+H_2$ up to 145 GPa. We also studied a lead sample embedded in Helium as reference. A major goal was to investigate the possible existence of lead polyhydrides. We have carried out four 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 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	Pb+He	300	18 - 55	300	
Run 2	$Pb+H_2$	100	47 - 77	300	47, 77
Run 3	$Pb+H_2$	100	90 - 145	300	126, 129, 140
Run 4	$Pb+H_2$	70	0.3 - 50	300	25, 44

Table 1: Conditions of the four experimental runs.

The equation of state derived form these experiments are plotted on Fig. 1. We performed a Vinet fit of our data with V_0 = 20.2(4) A^3/atom, K_0 = 44(4) GPa and K_0' = 5.55(20). The deviation observed coul be well explained by the texturation of the crystal phases after laser-heating, as it tends to form monocrystals (or at least heavily textured phases). Laser-heating in the 25 – 140 GPa range did not lead to any change in volume, and thus in stoichiometry. Nonetheless, laser-heating was sufficient to initiate the *hcp*-to-*bcc* phase transition of lead at 135 GPa, indicating that we brought enough energy to the system to help it transit to a more stable state.

This experiment shows that there is no stable hydride in the Pb-H system up to 140 GPa, contrarily to what was predicted. Interestingly, lead seems to be inert regarding a reaction with hydrogen, in the same way that gold is. Moreover, *ab initio* calculations seemingly underestimated the equilibrium pressure of the PbH_4 hydride, and this study should contribute to help DFT predictions being more accurate.

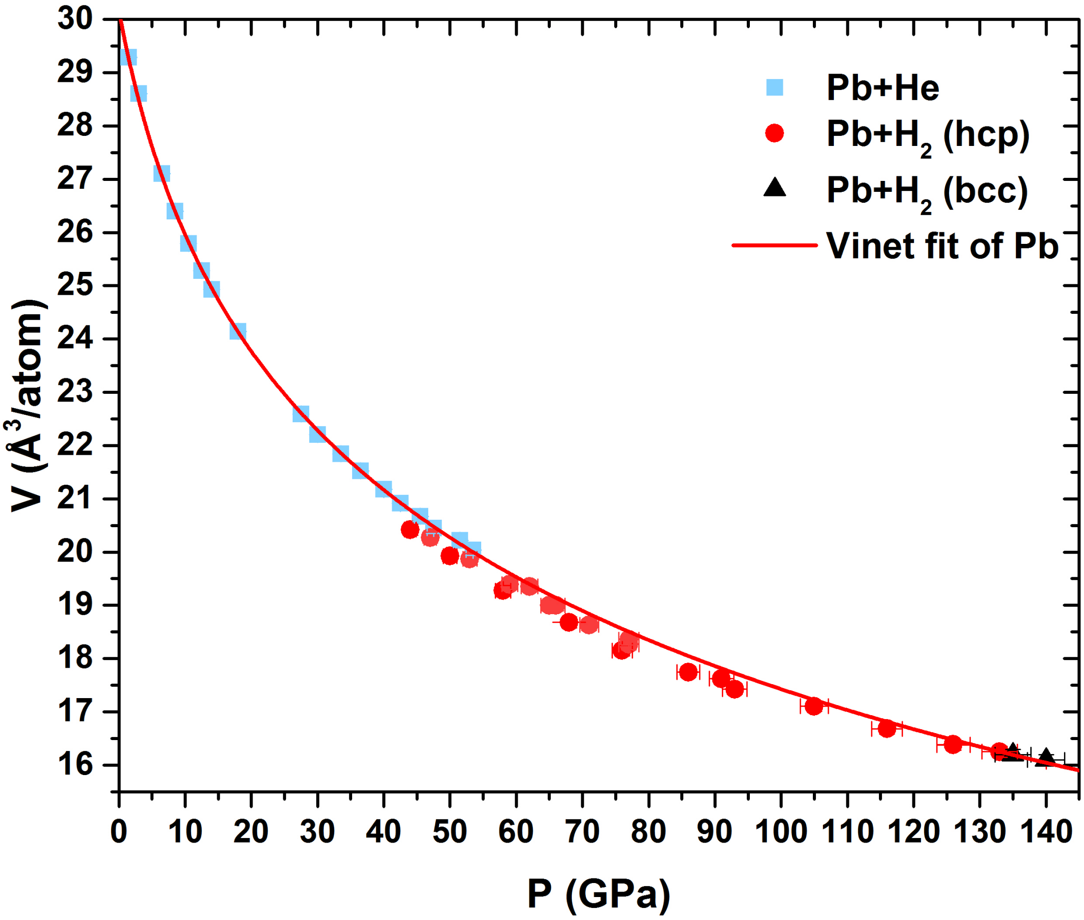

Figure 1: Evolution of the volume as a function of pressure for Pb embedded in H_2 and He, together with the Vinet fits of the data.

References:

[1] C. Pépin, PNAS 112, 7673 (2015)

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