ESRF	**Experiment title: Structural data of phase IV of solid H2.**	**Experiment number**: Hc-1910
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Shifts: 12	**Local contact(s)**: Mohamed Mezouar and Gaston Garbarino.	

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Report:

The aim of this proposal was to extend the determination of the structural data of solid H2 at 300 K up in the domain of phase IV, i.e above 220 GPa. Phase IV was discovered by Raman measurements [1]. Many ab-initio calculations have converged to a good candidate structure for phase IV [2]: It is a mixed phase with alternate layers of free rotating molecules on a hexagonal lattice and quasi-graphene H planes. A direct structural determination is now needed to confirm this intriguing structure.

X-ray diffraction measurements of solid H2 at very high pressure are very challenging because of the weak scattering power of the sample: at very high pressures, the solid H2 x-ray spectra is usually swamped by the Compton signal of the diamond anvils. The maximum pressures for the structural determinations of solid hydrogen achieved so far are 120 GPa at 300K [3] and 180 GPa at 77K [4]. Using a new development on ID27, i.e a multichannel collimator (MCC) that could reduce drastically the diamond anvils Compton scattering signal, we could in a first run (HC-1375) perform X-ray diffraction measurements on phase I up to 200 GPa. Due to the orientation of the crystal only the {100} peaks could be followed. Also, one anvil prematurely broke at 200 GPa due to H diffusion.

In this second run (HC-1910), a second attempt was made to measure structural data of solid hydrogen in the stability field of phase IV. Three DACs were loaded, two equipped with anvils of 40 µm culets and another one with 30 µm culets. The three single crystals were characterized and oriented about a pressure of 7 GPa. The best crystal was selected for the high pressure run. Two peaks were followed (100) and (101). The Multichannel Collimator was used to reduce the Compton background signal. A gain of a factor five in the signal to backgrtound ratio was hence obtained. XRD was performed with the 33KeV monochromatic beam. Pressure was measured with the volume of a small ball of gold (2µm diameter) placed in the sample chamber (no formation of gold-hydride was observed). A Raman setup operating in the red was mounted on a sliding stage on the X-ray table to measure the Raman signature of the phase transition to phase IV. The vibron frequency shift of diamond was measured and also the shift of the Raman phonon of diamond at sample-culet

interface. The diamond phonon Raman frequency shift is used as a secondary pressure scale for spectroscopic measurements in the multi-megabar pressure range [5].

The evolutions of the 100 d-spacing and 101 d-spacing are shown in the figure below.
Also, the pressure as calculated from the shift of the diamond phonon edge and from the volume of Au is compared. There seems to be an intrinsic uncertainty of ~15 GPa in the diamond edge estimation of pressure but the calibration of Akahama is correct at least up to 200 GPa.

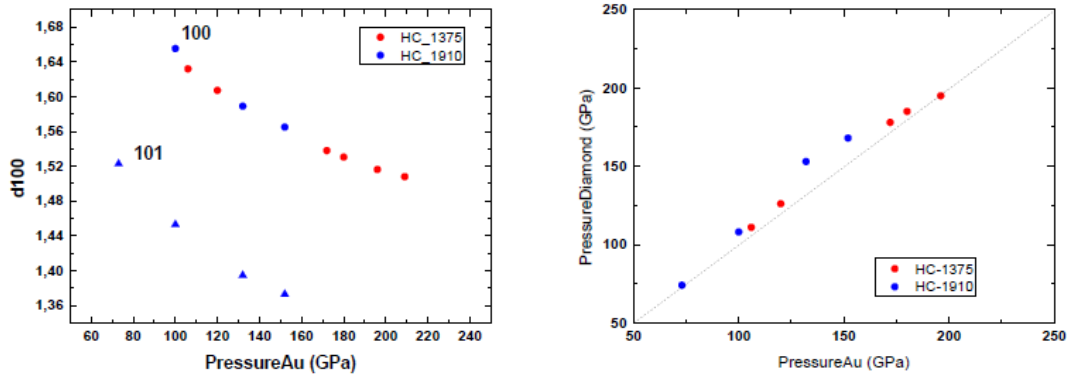

Figure 1 : Left: Evolution of d_{100} and d_{101} of solid H_2 at 300 K in phase. The data of runs HC-1375 and HC-1910 are represented in red and blue respectively.
Right: pressure estimated from the diamond anvil phonon versus the one obtaonedfrom the volume of a ball of gold embedded in hydrogen.

References:

[1] R. Howie et al. PRL 108, 125501 (2012).
[2] C. Pickard and R. Needs. PRB 85, 214114 (2012).
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