ESRF	**Experiment title: Structural data of phase IV of solid H2.**	**Experiment number**: Hc-1375
Beamline: ID27	**Date of experiment**: May,2 – May, 6 2014	**Date of report**: 15/09/06
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. Over the past three years, many studies have been devoted to the determination of phase IV of solid H2 based on Raman and IR measurements and on ab-initio calculations. A very intriguing structure seems a good candidate [1]: 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.

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 [2] and 180 GPa at 77K [3]. A new development on ID27 appeared promising to go to higher pressure, i.e a multichannel collimator (MCC) that could reduce drastically the diamond anvils Compton scattering signal.

A specific experimental configuration has been adapted for this study. The DAC has to be mounted on a rotator to bring the diffraction peaks in the area of the image plate detector accessible through the MCC. A Raman setup operating in the red has to be mounted on a sliding stage on the X-ray table to measure the Raman signature of the phase transition to phase IV. Above 200 GPa, there is some dispersion of the pressure determination among the various spectroscopic studies, which were based on the shift of the Raman phonon of diamond at sample-culet interface. This diamond Raman based measurement of pressure was also used in the present study but the volume of a little Au ball embedded in the sample was also used for the pressure determination. A good agreement between this two determinations was obtained to the maximum pressure.

Three samples were prepared for the allocated beamtime. Their characteristics are summarized in the table below. The first DAC was equipped with 50 µm culet anvils. We tried to optimize the dimension of the H2 sample so as to secure a measurable signal above 120 GPa. Unfortunately, there has been some instability in the sample chamber and the crystal broke at 80 GPa. In the second sample, there was gold impurities in the H2 solid due to the Au coating of the gasket . Fortunately, the third samples worked very well up to 215 GPa. The DAC broke before reaching the expected limit of 250 GPa. In all three cases, about 4 peaks among the ones of the (100] and (101) classes were accessible within the x-ray aperture of the DAC. That confirms previous observations that the H2 crystals tend to grow with a c-axis almost aligned perpendicular to the diamond culet. Consequently, many different samples will probably have to be studied to be able to obtain a good crystal orientation that will enable to follow the pressure evolution of the [002] peak.

Sample	**Anvils**	**Measurements**	**Maximum pressure of measurements**
H2	50 µm culets	[100], [10-1],[-1,0] peaks	80 GPa. Two thick sample, breakage of crystal.
H2	40 µm culets	[100], [1-10], [-100], [-110]	Au dirt in H2 sample. Problem of gasket coating.
H2	40 µm culets	[100], [1-10], [-110], [-100], [010]	215 GPa

The evolution of the 100 d-spacing measured of the last sample is shown as red dots in the figure below. These data extends previous measurements at 300K, as black dots, from 120 GPa up to 215 GPa. Raman measurements of the H2 vibron enable to check that solid H2 remained in phase I. On the right part of the figure below, the raw x-ray and Raman data at 215 GPa are presented. Measurements up to the 260 GPa range should certainly be feasible. It should be noted that the signal to noise ratio has been increased by a factor 10 by using the MCC device. A factor 20 was expected but the alignment of the MCC device is very sharp.

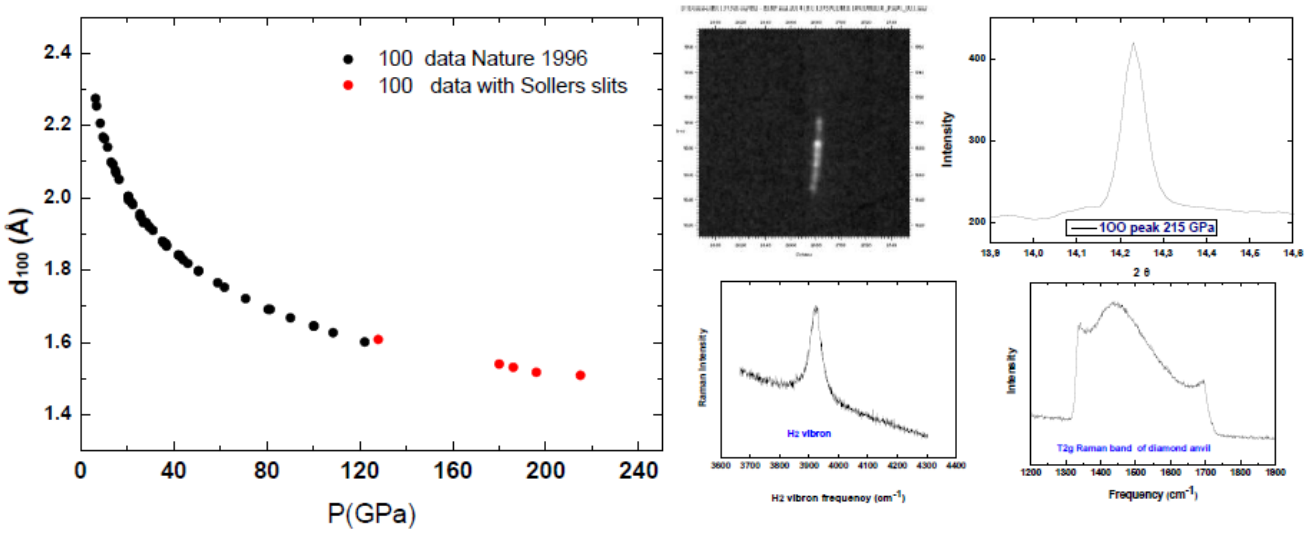

Figure 1 : Left: Evolution of d_{100} of solid H_2 at 300 K in phase I. Black dots previous measurements (ref.1), red dots new measurements obtained with Sollers_Slits configuration.
Right: Data recorded at 215 GPa. The 100 peak and its ciorresponding IP signal ; the H2 vibron and the diamond anvil phonon.

References:
[1] R. Howie et al. PRL 108, 125501 (2012).
[2] C. Pickard and R. Needs. PRB 85, 214114 (2012).
[3] I. Naumov, R. Cohen and R. Hemley. PRB 88, 045125 (2013).
[4] P.Loubeyre et al, Nature 383, 702 (1996).