ESRF	Experiment title: Structural properties of H_2 and He up to 200 GPa	**Experiment number:** **HS499**
Beamline: **ID9**	**Date of experiment:** from: July 1 1998 to: July 10 1998	**Date of report:** **2/09/98**
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Report: The technique to achieve structural diffraction on low-Z solids, such as H_2 and He at very high pressure in a Diamond Anvil Cell (DAC) is based on a combination of single-crystal and energy-dispersive technique with a ploychromatic radiation (1). This method was shown to give very accurate structural data allowing for example the determination of the isotopic shift on the equation of state of LiH (2). Also, fine effects could be observed such as the observation of superlattice reflections in ice VII revealing the stability of modulated phases (3). We started to develop this technique at the ESRF in 1994 for the determination of the EOS of H_2. In 1996, the reflections of hydrogen could be measured up to 120 GPa by embedding the single-crystal in helium so as to limit its fragmentation by pressure (4). Similarly, the EOS of He was measured to 130 GPa. But extending such measurements in the 200 GPa range remains a real challenge.

In a previous report on the runs HS 140, HS 141 and HS 191 also dedicated to this project, we have explained the reasons of our many failures. In fact, new problems occur upon decreasing the sample chamber to reach very high pressures. It took few runs to disclose the problems as: -a) increased fragmentation of the single-crystal. To reach higher pressures the central flat has to be reduced in the 50 pm - 100 pm diameter and we observed that the thickness of the sample chamber is not stabilized before 80 GPa. - b) Diffusion of hydrogen in the rhenium gasket. Helium worked perfectly to stop this diffusion in the previous experiments because the sample chamber was larger and thus the thickness of the ring of helium in the samz proportion. -c) Breakage of the diamond anvils due to the diffusion of helium in the small defects of the stone that generally opens up when the diamond start to deform that is around 50 GPa. We found out that one

anvil out of four will break, that means one experiment out of two. We have tried to find criteria to select the good stones by topography at ESRF beforehand but without success.
The history of the samples studied during the HS499 beamtime is summarized in the table below.

Samples	**Flat of the anvils**	**Maximum pressure**	**Reason of failure**
H_z in a ring of gold	**75μm** ⊗ 100 pm	7 GPa	Poor growth of the cryatl at high temperature (500K).
He	50 **μm ⊗ 50μm**	40 GPa	Fragmentation of crystal
H_z in a ring of gold	**75μm** ⊗ 100 **μm**	30 GPa	Good crystal but instability of the ring of gold with P.
H_2 in He	**85 μm ⊗ 85 μm**	21 GPa	Growth at 500 K. No peak found at 21 GPa.
H_2 in He	**100μm ⊗ 100μm**	60 GPa	Breakage of anvil by He
H_2 in He with gold ring	**100μm ⊗ 100μm**	15 GPa	Bad orientation of the crystal. Too few reflections
He	**75μm ⊗ 100μm**	40 GPa	Breakage of anvil by He

In summary:

We have observed that using culets smaller than 100 **μm** leads to a too small thickness in the 100 GPa range and thus to a dramatic fragmentation of the single crystal. On the other hand, we are now confident that a good quality single crystal could be kept up to the maximum pressure of the 100 **μm** culet anvil, that is around 170 GPa. This is sufficient to address the problem of the structure the phase III of hydrogen.

We believe that we now master the proper parameters for the stability of the ring of gold. In particular, it has to be very thin **(~5μm)** before the loading of the cell but that is a very difficult operation to achieve. This also allows the growth of the crystal at high temperature, thus starting at higher pressure with a good quality single-crystal therefore reducing the fragmentation in the 100 GPa range.

The breakage of diamond by the diffusion of helium in defects is a serious problem because it occurs in the going up in pressure **(~50GPa)** on good samples. This is quite beamtime consumming but it cannot be anticipated. It is just a question of statistics.

Finally, the x-ray viewing angle of both sides of the sample is **±40°** in our DAC and for some orientations of the hcp structure gives only few peaks accessible that would not give enough information at the phase transition in dense hydrogen.

We are slowly progressing on this difficult problem. We now know that we can gain the 50 GPa needed to probe the structure of phase III at 77K. More beamtime and a little chance (quality of diamonds + orientation of the crystal) are needed.

References:

I) R. LeToullec et al, High Pressure Research 8, 691 (I 992).
2) P.Loubeyre et al, Phys. Rev. B 57, 10403 (1998).
3) P.Loubeyre et al, to appear in Nature.
4) P.Loubeyre et al, Nature 383, 702 (1996).