ESRF	**Experiment title:** Structural properties of H_2 and He up to 200 GPa at very low temperature by angle-dispersive single crystal X-ray diffraction.	**Experiment number**: HS-1208
Beamline: **ID30**	**Date of experiment**: from: June, 14 2000 to: June, 20 2000	**Date of report**: 29/08/00
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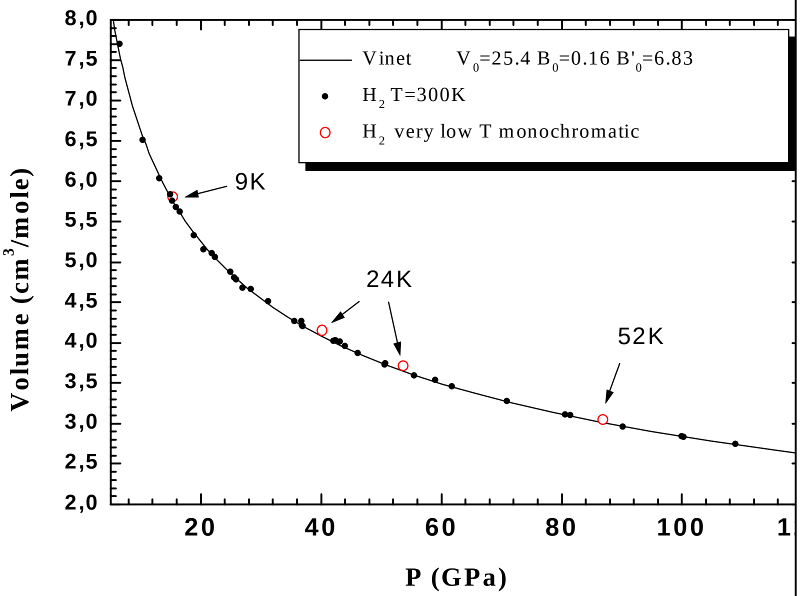
Report:

We have performed single-crystal x-ray diffraction with a monochromatic beam on solid H_2 and solid ^{3}He up to almost the Mbar range.

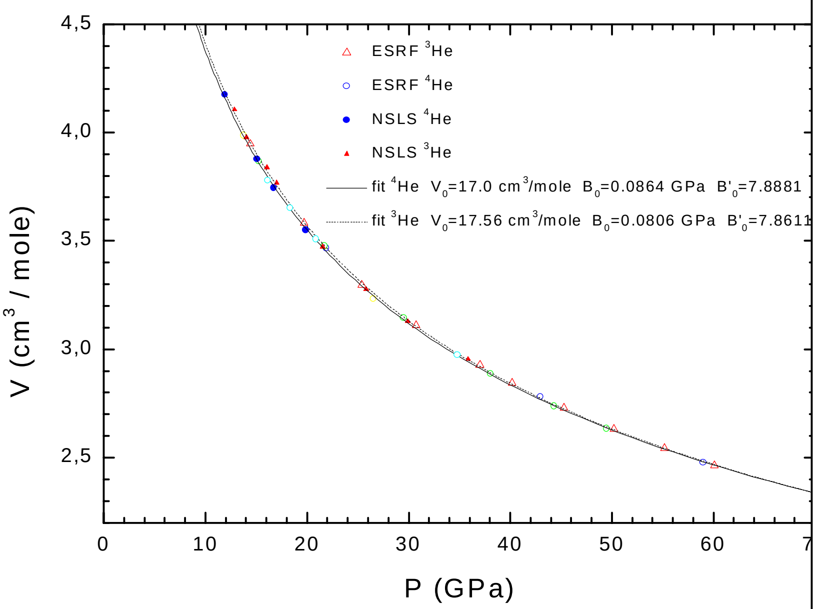
The combined use of a focussed monochromatic beam with a fast readout on-line image plate (IP) detector to perform single crystal diffraction in a diamond anvil cell has been installed and commissioned on ID30 by L.Finger and M.Kunz (report HS349). In particular, they have shown that they could correlate the reflections of an hcp H_2 single-crystal and follow the three more intense reflections (100, 101 and 002) to 25 GPa. The aims of the present experiment was:

- First, to extend such measurements on hydrogen in the Mbar range; To show that the monochromatic technique enables the measurement of reflections in a broken crystal whereas they would be hidden by the Compton scattering in the white beam technique; to show that these high pressure single-crystal measurements can be also performed at very low temperature in a cryostat (down to 10K).
- Second, to determine the structure of phase II and phase III, a long-sought issue in the problem of dense solid hydrogen; to measure the isotopic shift on the equation of state between ^{3}He and ^{4}He.

The measurements were performed on ID30. The undulator emission was monochromatized at 0.3738 Å and focussed in a spot of 20µm (even for 40 µm diameter sample chamber, the tail of the spot produced diffraction from the rhenium gasket that could saturate the IP for long accumulations and hence perturbed the measurements). The fastscan IP detector was used. Five diamond anvil cells were prepared to achieve high pressure and low temperature, 3 loaded with hydrogen, one with ^4He and one with ^3He. In all of them, single-crystals had been grown just above the solidification pressure at 300K (12 GPa for He and 5.4 GPa for H_2). The H_2 crystals were embedded either in a ring of gold or in helium to prevent their fragmentation under pressure. In about 3 shifts, reflections, their rocking curves and the orientation matrix of a single crystal could be obtained. Unfortunately, we had problems in trying to achieve very low temperature at pressure above 40 GPa. Our cryostat is based on a flow of liquid helium inside the DAC. We discovered that the seals in the DAC to confine liquid helium operate well when moderate forces is applied on the piston but leak when more important forces are applied (due to a small deformation of the DAC) therefore leading to a vacuum problem and a rapid warming of the DAC. Three samples were lost because of such a problem. Fortunately, two samples
gave very encouraging results:



H_2: Four volumes were measured (as circles) at various temperatures (due to the above-discussed problem of the cryostat, the minimum temperature was increasing with pressure). The sample chamber was 40µm in diameter with a ring of gold (100 µm flat bevelled anvils were used). Three reflections (101, 011 1-10) could be followed within the optical aperture of the DAC. They became too weak to be measured above 85 GPa. A similar experiment performed with the white beam technique would have stopped at 40 GPa. A negative thermal expansion is clearly measured. This can be understood if we take into account the ortho-para conversion at low temperature and consequently the quadrupole pressure. It is shown here that this spin effect has macroscopic consequences and therefore nuclear spins should now be taken into account to accurately describe dense hydrogen. This could explain the very small isotopic shift between the EOS of H_2 and D_2.



^3He: The crystal was 25 µm in diameter (300 µm flat anvils were used but the loading of the DAC was done at a pressure 6 times smaller than what is currently used for ^4He). Measurements were performed at 300K. The data, as shown with open triangles on the figure, extend a previous determination performed at NSLS. Compared with the ^4He data collected at the ESRF, a crossing between the EOS of ^3He and ^4He is now unambiguously shown at around 60 GPa. The same measurements should now be performed at 9K to understand this anomalous quantum effect.