C1Po1E-04

Development status of the in-situ measurement system for ortho-to-parahydrogen fractions for the ESS Cryogenic Moderator System (CMS)

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Abstract

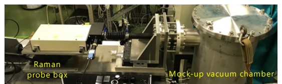
The Cryogenic Moderator System (CMS) is equipped with a catalyst to convert hydrogen from the ortho state to the para state in order to keep desirably high parahydrogen fractions of 99.5%. An in-situ measurement system for the ortho and para fractions of liquid hydrogen (OPMS) by means of a Raman spectroscopy with a precision of 0.1% is being developed to detect an undesirable shift towards a high orthohydrogen fractions caused by neutron scattering driven para-to-ortho back conversion. We have demonstrated that our developed sapphire window, through which the laser and backscattered photon travel, has endured the required pressure cycle thermal cycle in liquid hydrogen environment. Meanwhile, a Raman optics system has been developed using normal hydrogen at 0.3 MPa and 300 K. Eventually, we have succeeded in detecting the parahydrogen peak at $J = 4$ (1245 cm^{-1}), which is our preliminary goal, because the peak ratio of parahydrogen at 1245 cm^{-1} to ortho hydrogen at 587 cm^{-1} corresponds to 0.5%. It can be expected that it would achieve our requirement for liquid hydrogen because it has300 times higher higher density.

Introduction

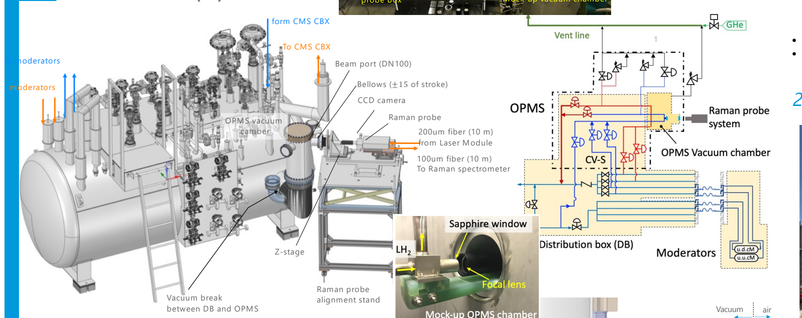
At the ESS target, high energy spallation neutrons are produced by impinging 5 MW proton beam on the high-Z material, tungsten. The proton beam is pulsed with a repetition of 14 Hz and a pulse length of 2.86 ms. The moderator system consists of a water pre-moderator and two liquid hydrogen cold moderators, which are optimized to achieve a high cold neutron brightness. The neutronic performance of the cold moderators degrades rapidly with the decreasing parahydrogen fraction below 99.5%. The neutron collisions in the cold moderators increases the orthohydrogen fraction if it is not compensated by the ortho- to parahydrogen conversion driven by the catalyst system. Therefore, the Cryogenic Moderator System (CMS) is equipped with an ortho-parahydrogen (OP) catalyst to keep desirably high parahydrogen fraction and the ortho-to-parahydrogen ratio will be measured by ***the in-situ ortho-parahydrogen measurement system (OPMS)*** by means of a Raman spectroscopy. The CMS provides subcooled liquid hydrogen with a temperature of 17 K and at a pressure of 1 MPa to the two moderators at a flow rate of 0.5 kg/s to remove nuclear heating at the moderators.

The purpose of this study is
(1) Pressure and thermal cycle test results of the developed sapphire window in liquid hydrogen
(2) R&D status for the high-precision Raman optics system.

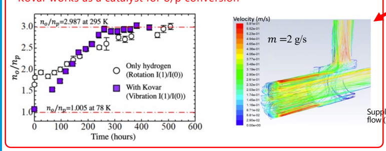
ESS OPMS Design

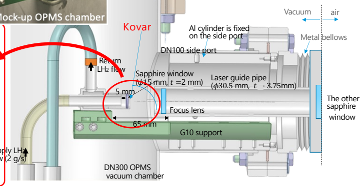


CMS Distribution box (DB) –installed in 2022



Kovar works as a catalyst for o/p conversion





- Two sampling pipes with one sapphire window (15 mm dia.) section.
  - Design pressure is the same pressure of 1.7 MPa as that of the CMS process line.
  - The sapphire window section is exchangeable by a VCR fitting before its lifetime, ***which has been determined by the pressure and thermal cycle tests.***
- Laser guide with a focal lens is supported by the beam port flange, which hasthe other sapphire window with the diameter of 33 mm to form a boundary between the atmosphere and vacuum.
- The laser beam and the collected light are transmitted via optical fibers to the laser module and a Raman spectrometer placed in an electrical cabinet, which is 5 meters away.

<u>OPMS data aquision system</u>

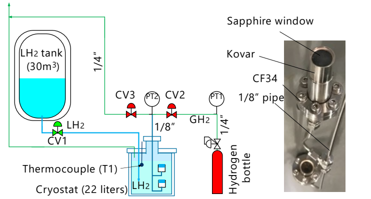
- The spectrometer will be connected through USB to an Industrial Personal Computer (IPC) that will be also installed in the electrical cabinet and run an EPICS input and output controller (IOC) locally and interfaces with the open-source library driver "Ocean Optics SeaBreeze API".
- All the signal processing will be performed by the IOC. The IOC will be connected to the ESS Technical Network and interface with all the ESS Applications.

Sapphire window developwmt

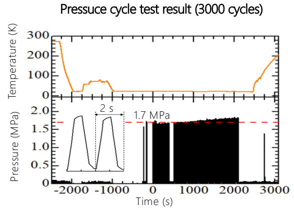
- It is necessary that a high reliable sapphire window is developed to deliver the incident laser beam and the Raman scattered light. The ESS is proceeding the R&D in collaboration with the Japan Aerospace Exploration Agency (JAXA), which had developed a 60 mm-diameter observation sapphire window for the internal visualization of the liquid hydrogen rocket tank at 0.6 MPa.
- The ESS sapphire window diameter is reduced to 15 mm because the design pressure is increased up to 1.7 MPa. The 2.0 mm-thickness sapphire window was brazed to a pipe made of Kovar (Fe-Ni-Co alloy) in a vacuum environment, because its thermal expansion ratio is almost the same as that of the sapphire.
- Required lifetime of the sapphire window: 5 years.
  -Two kinds of the endurance tests:
  (1) more than 100 thermal cycles between 20 K and 300 K
  (2) more than 100 pressure cycles between 0.1 MPa and the design pressure of 1.7 MPa.

1. Pressure cycle test

- Liquid hydrogen is continuously supplied from the liquid hydrogen tank with the volume of 30 m³
- Liquid level was always kept above the sapphire window, being monitored by a thermocouple (T1).
- One side of the sapphire window is connected to a 1/8" pipe to be pressurized to 1.7 MPa or depressurized to atmospheric pressure quickly by hydrogen gas.

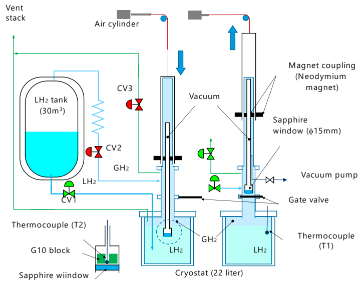


Pressuce cycle test result (3000 cycles)



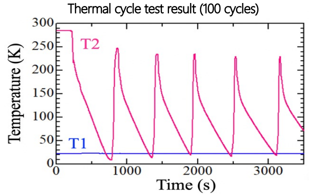
- 3,000 pressure cycles were applied to three sapphire window test samples at 2 s per cycle.
- After warming it up, we have confirmed that ***<u>no harmful leak</u>*** was detected (below the criteria of 1 x 10^{-10} Pa m³/s.)

2. Thermal cycle test



- Sapphire window was directily submerged in LH2 to cool it down rapidly and lifted and sprayed warm GH2.
- There is a gate valve on the cryostat in order to prevent the evaporated cold hydrogen gas from coming to the warm section.
- For the hydrogen safety, hydrogen was confined in the test equipment without leakage to its surrounding.
- A shaft connecting to the sapphire window and the outside guide pipe were coupled by neodymium magnets in order to move the sapphire window up and down in a non-contact matter.
- The sapphire window can be indirectly moved by means of a pneumatic cylinder.

Thermal cycle test result (100 cycles)



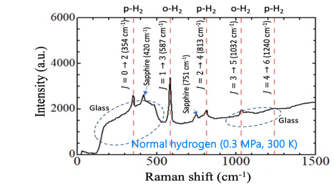
- The required submerging times were estimated to 7.5 minutes
- The warm up time was 80 seconds.
- The cooling speed from 285 to 155 K was 1.7 K/s and that from 155 to 20 K was 0.35 K/s.
- ***No harmful leak by the helium leak test after both of 3,000 pressure-cycle and 100 thermal-cycle tests.***

Development of a Raman optics system (0.1% accuracy of o/pH2 fraction)

- First generation EES Raman system -

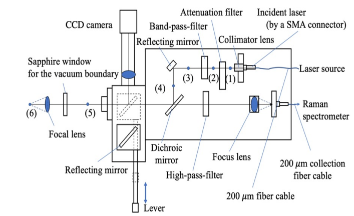
- 532 nm multi-mode laser with an output of 50 mW
- Fiber optic sampling probe for backscattering geometry with the diameter of 12.7 mm and the length of 108 mm by InPhotonics
- Raman spectrometer (model QE-Pro high sensitivity fiber optic spectrometer by Ocean Optics).

J	Rotational transiton ($J \to J+2$)	$E(J+2,0) - E(J,0)$: Raman shift (cm^{-1})	$I(J)/(0)$	Para /ortho
0	0 → 2	354	0.327	Para
1	1 → 3	587	1.00	Ortho
2	2 → 4	813	0.154	Para
3	3 → 5	1032	0.111	Ortho
4	4 → 6	1204	0.00494	Para

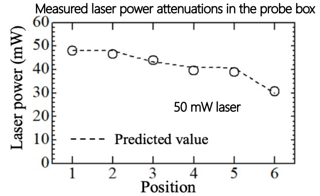


- Spectra of glass from a focal lens appeared over a wide wavelength range
- Peaks of sapphire were close to those of p-H2 ($J = 0$) and o-H2 ($J = 1$)
- Background level was too high to detect the peak at 1204 cm^{-1} → **No accuracy of 0.5% fraction.**

- Developed EES Raman system -

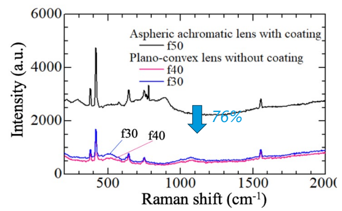


Measured laser power attenuations in the probe box

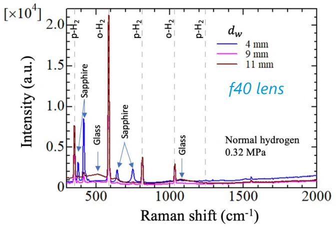


- Incident laser: a 532nm wavelength 100 mW diode laser.
- Collimator lens (f30), which is connected to the optical fiber directly by means of a SMA connector.
- It is focused through a focal lens with the diameter of 25.4 mm into the hydrogen environment through the sapphire window.
  - Spread of the laser beam below 12 mm at the location of the focal lens, which is 1.1 m away from it.
- Laser power attenuations in the probe box are as designed.
- Raman spectrometer, whose resolution has been changed from 5.5 cm^{-1} to 3.5 cm^{-1} with a slit size of 50 μm.

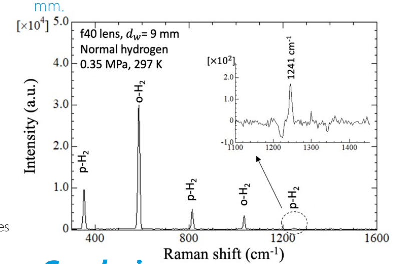
Effect of focal lens



Effect of focal point



Raman spectra of nH$_2$ at 0.35 MPa and 295 K with the f40 plano-convex lens at d_w = 9 mm.



- Development has successfully achieved our first goal of clearly detecting the p-H2 peak at 1240 cm^{-1}.
- The peak intensity is proportional to the hydrogen density.
- It is expected that the Raman intensities would be able to increase 265-fold at the nominal condition (17 K) because the density of the liquid hydrogen 265 times higher than gaseous hydrogen.

Conclusion

- In-situ measurement system by Raman spectroscopy has been developed.
- It was verified that the developed sapphire windows meets our requirement.
- The developed Raman optical probe system successfully achieved our preliminary goal of 0.5% accuracy.