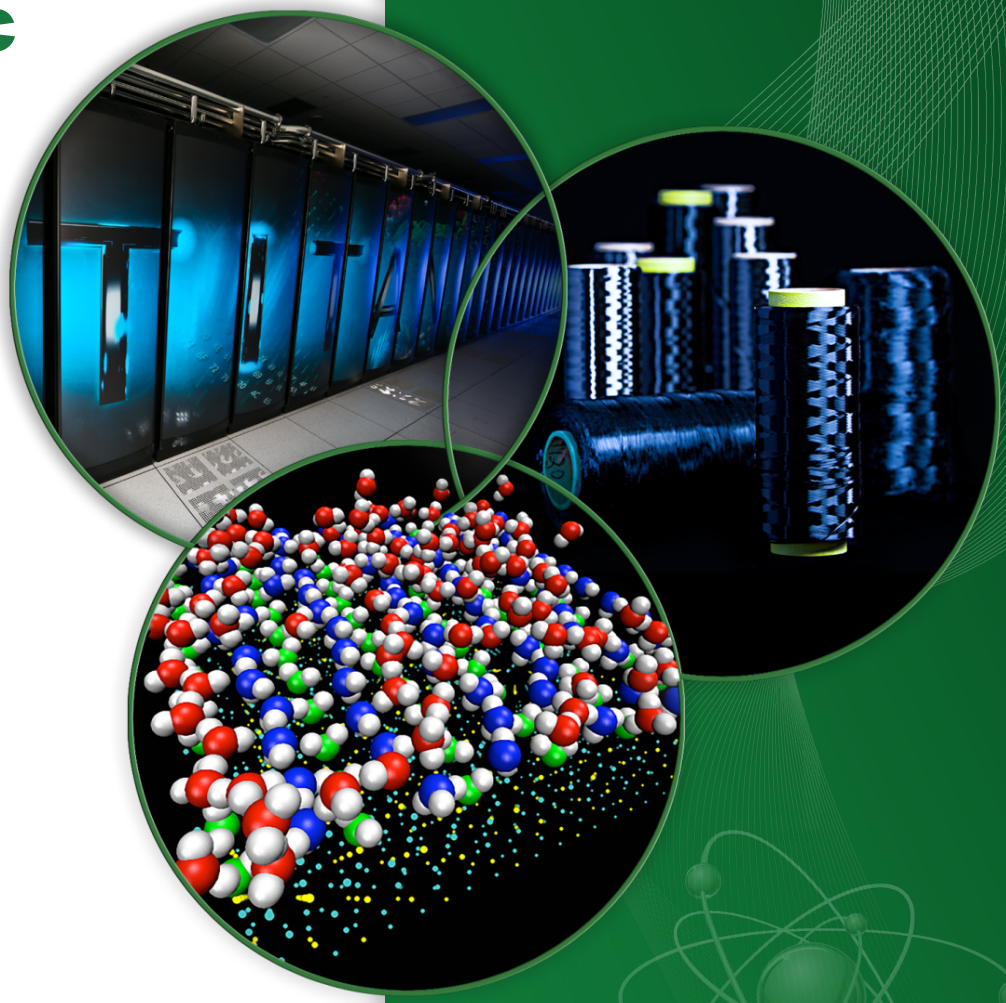
High Throughput Data Analysis for Inelastic Neutron Scattering Instruments:

Virtual Experiments in Spectroscopy with neutrons (VirtuES)

LDRD 7739



Timmy Ramirez-Cuesta,

Luke Daemen,

YQ Cheng (CEMD)

Stuart Campbell (NDAV)

June 4, 2015

ORNL is managed by UT-Battelle
for the US Department of Energy

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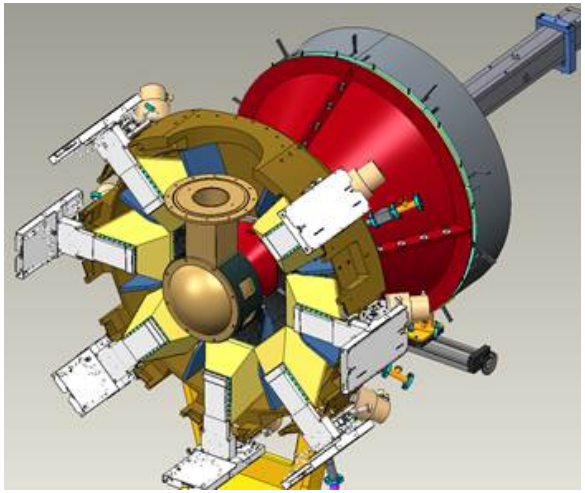
Chemical Spectroscopy VISION



4000 x TOSCA at ISIS, 1 TB of data/day

World's only high throughput INS spectrometer

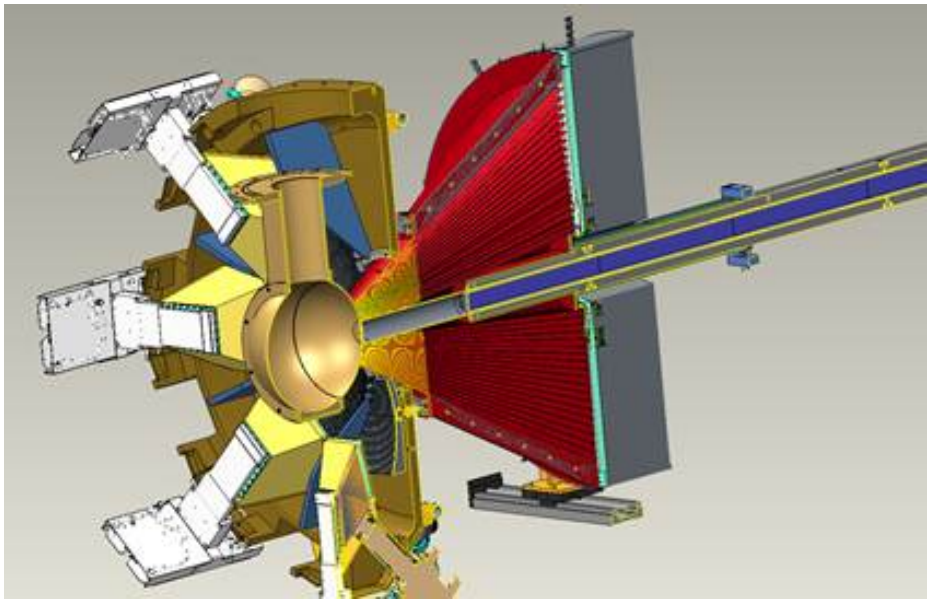
DFT modeling is of INS spectra is rigorous

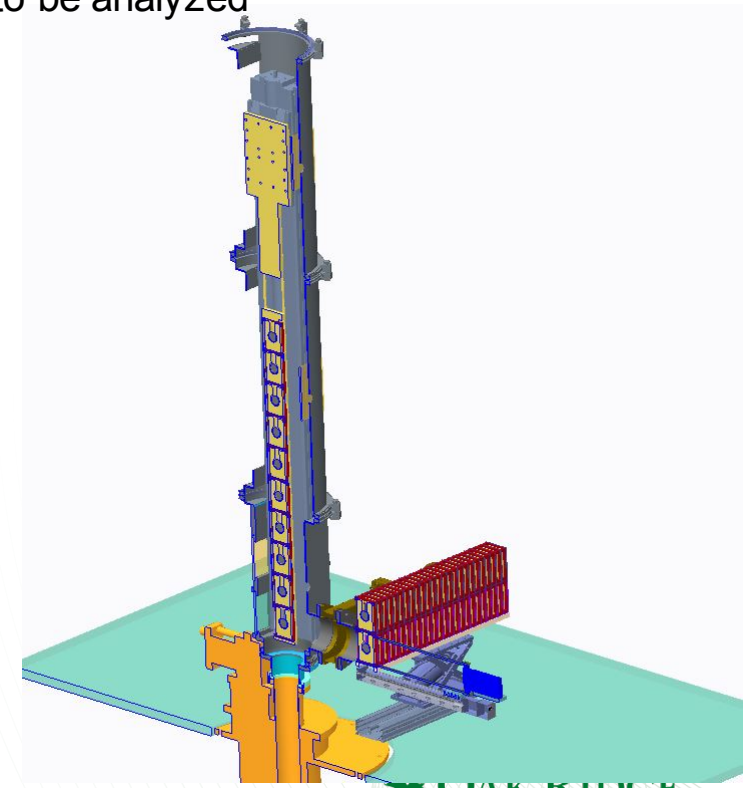
Gas handling is trivial

LDRD funding to build a computer cluster to support experimental program

Sample changer is being designed and will be ready by 2016

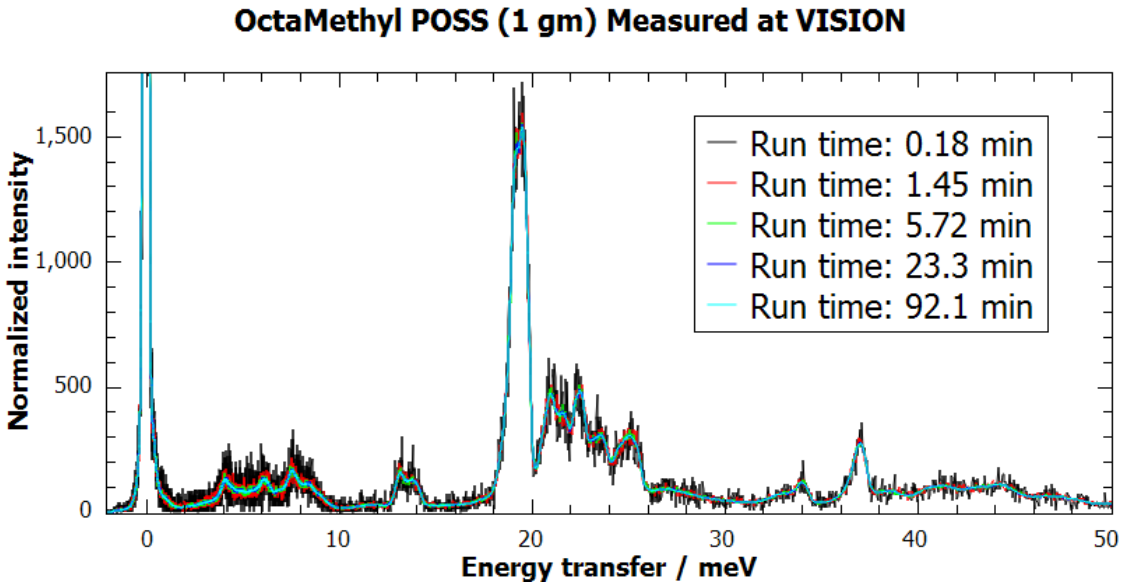
Expecting 10's of samples a day to be analyzed

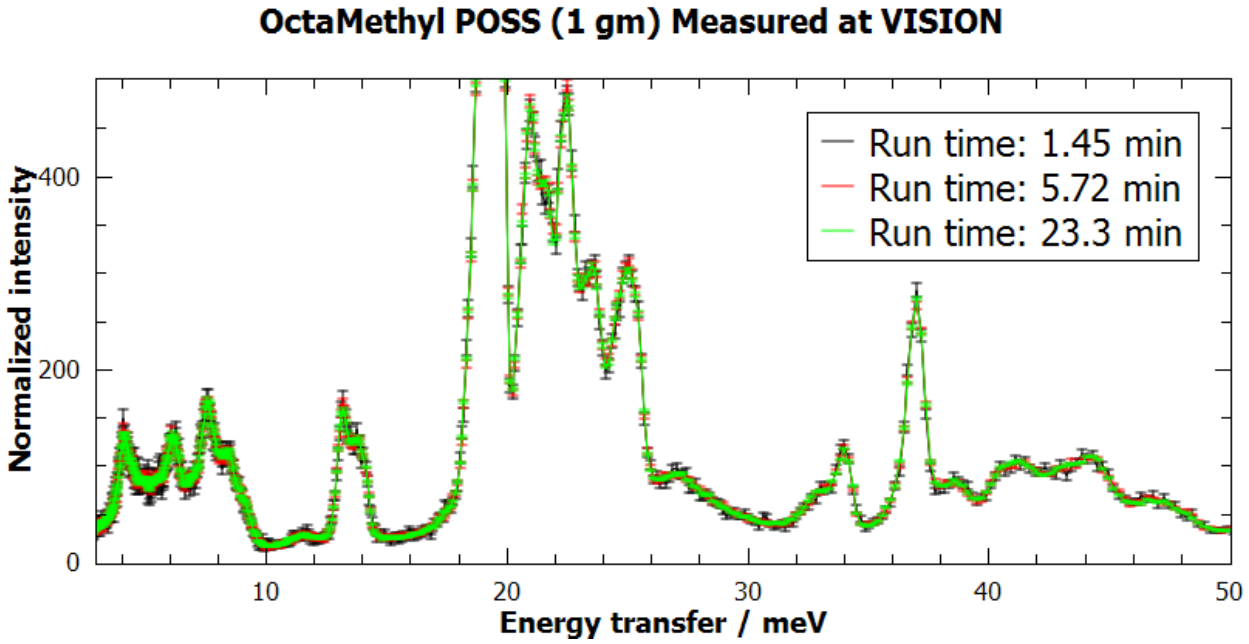




VIRTUES

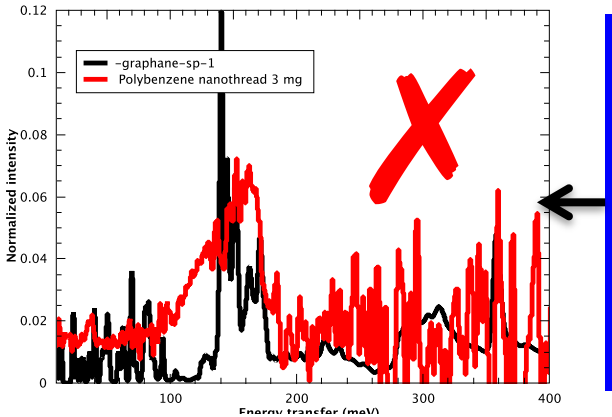
Examples from VISION

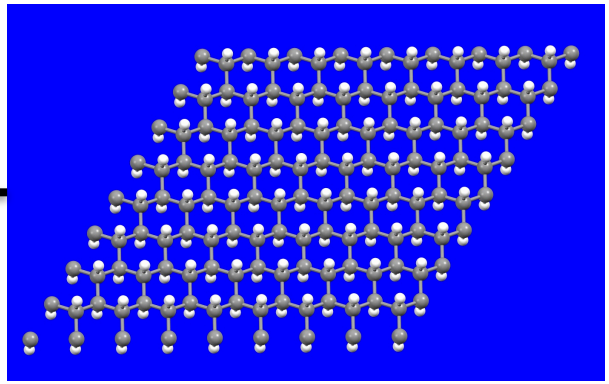


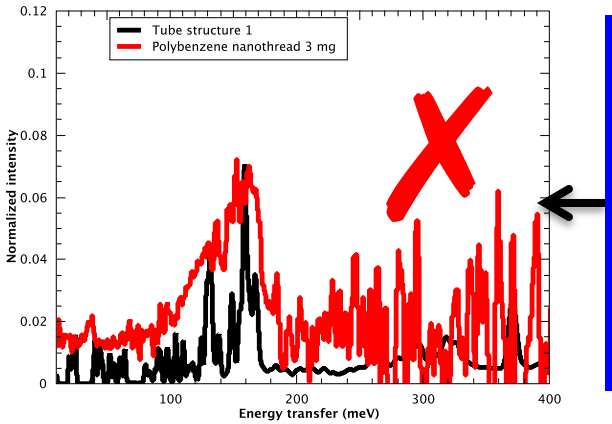


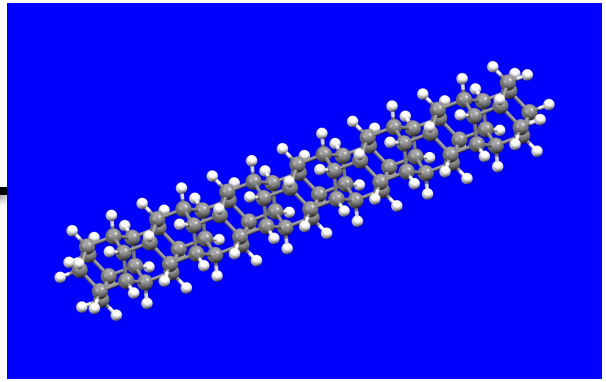
Polybenzene (nanothreads 3mg) formed at high pressure. Structural inference through modeling

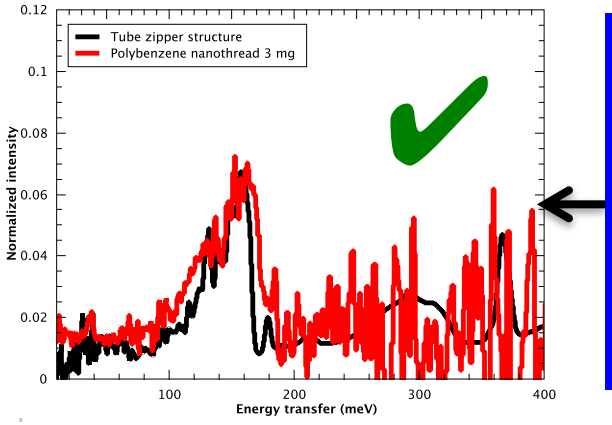
Malcolm Guthrie, John Badding, Vin Crespi,

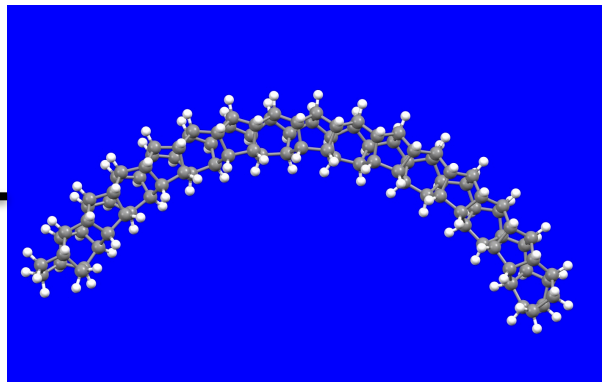










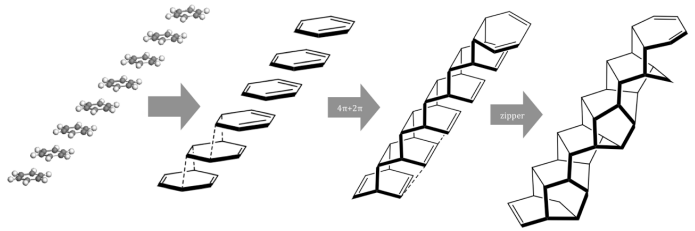


Compression-induced polymerized benzene. 3 mg sample synthesized on SNAP and measured in VISION.

Comparing the experimental data from VISION and a series of DFT calculations of hypothetical structures that contain sp3 carbon and the correct stoichiometry (H:C ratio 1:1) allows us to determine which structure correspond to the measured spectra.

Figures on the far left show the superposition of the measured spectra of polybenzene with calculated INS spectra of the structures (containing sp3 carbon) shown in the middle column.

Top) graphane, middle) tubular structure (highly symmetric) and bottom) the zipper structure. The zipper structure provides the better agreement between calculation and experiment.



Proposed mechanism of polymerization (zipper structure)

Computer modeling is vital to understand the spectra. The calculations shown here took up to 36 hours using 1024 cores.

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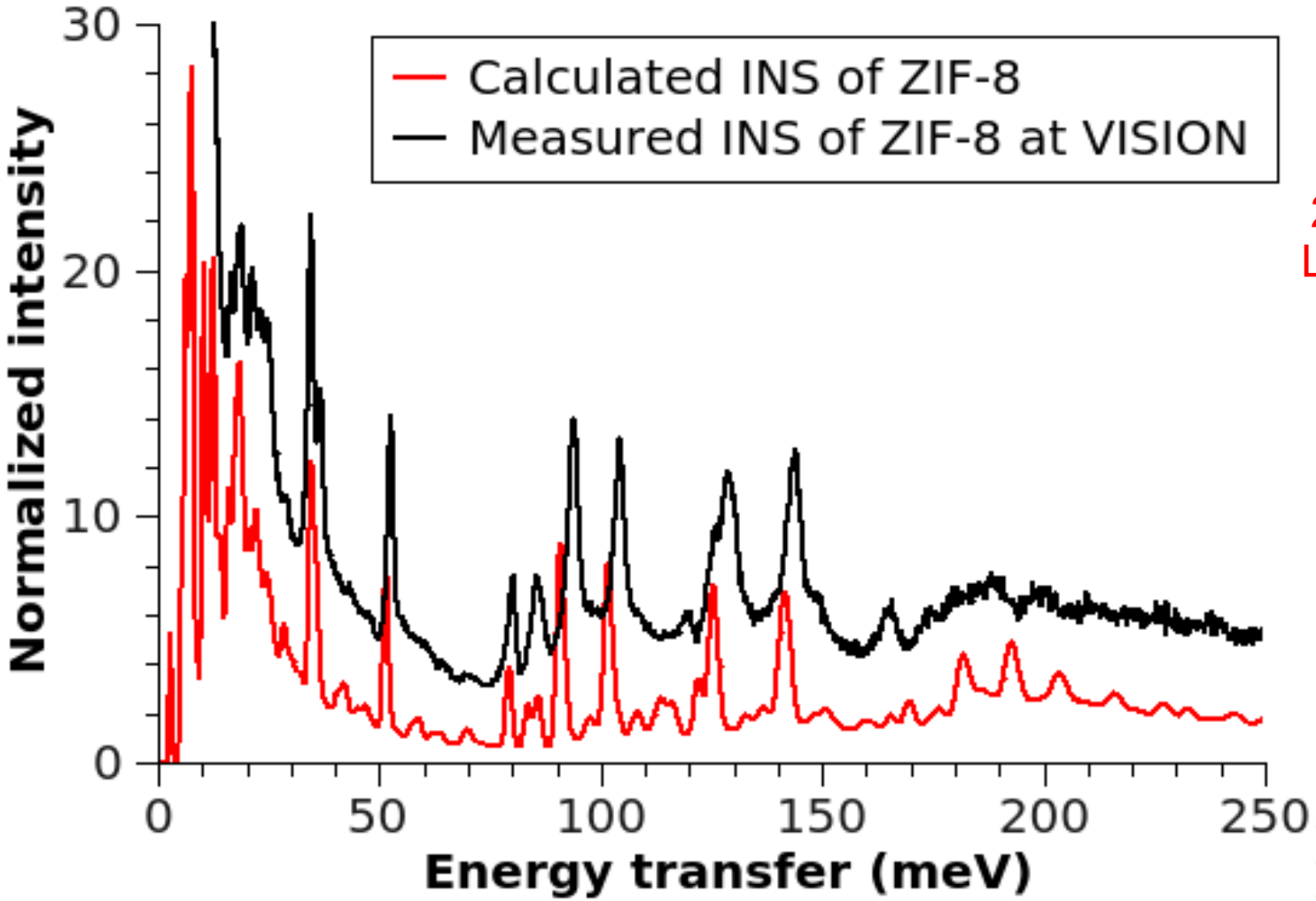
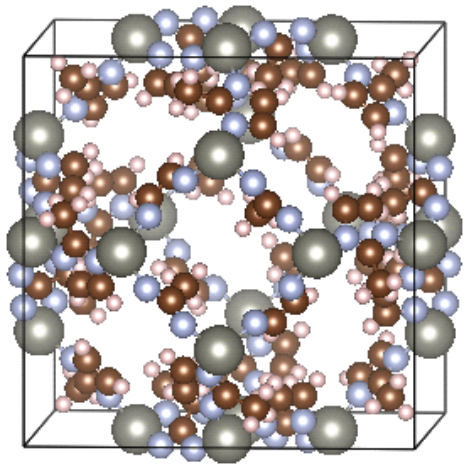
Vibrational spectrum simulation on materials with large unit cell or complicated structure



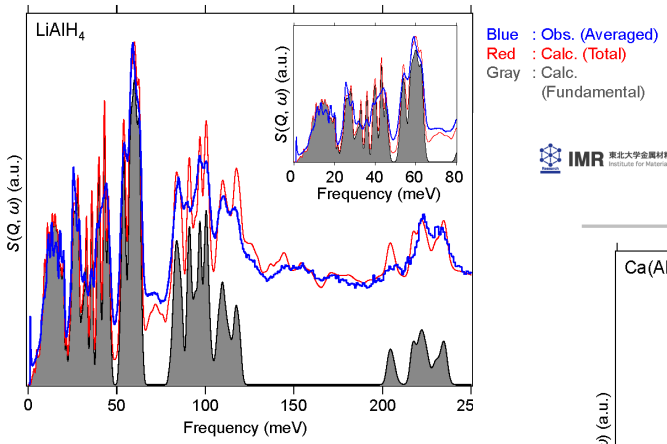
278 atoms in unit cell
Lattice parameter 17Å



On-the-fly modeling/analysis of VISION data

IMR 東北大学金属材料研究所 Institute for Materials Research, Tohoku University

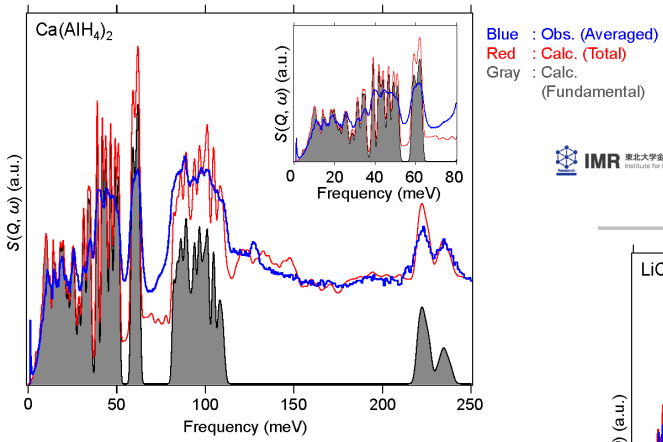
aClimax ($LiAlH_4$)



Modeling completed upon or even before the experimental data is available

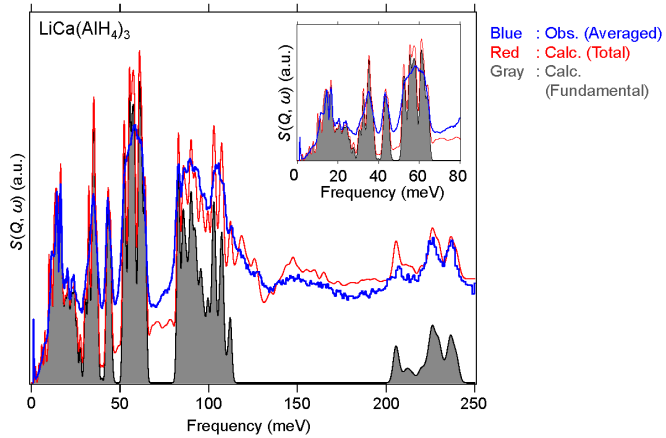
IMR 東北大学金属材料研究所 Institute for Materials Research, Tohoku University

aClimax ($Ca(AlH_4)_2$)



IMR 東北大学金属材料研究所 Institute for Materials Research, Tohoku University

aClimax ($LiCa(AlH_4)_3$)



Dear all,

This is Toyoto.

I really appreciate our experiments at VISION.
Thanks to you, I could get extremely good results in this time.
I believe that VISION is the one of the best neutron vibrational spectroscopy in the world.
I am so happy to have the experiments by VISION.
.....

Thank you very much for everything on the INS experiments and DFT calculations in ORNL in the last week.
I am very happy to have the experiments and get the excellent results.

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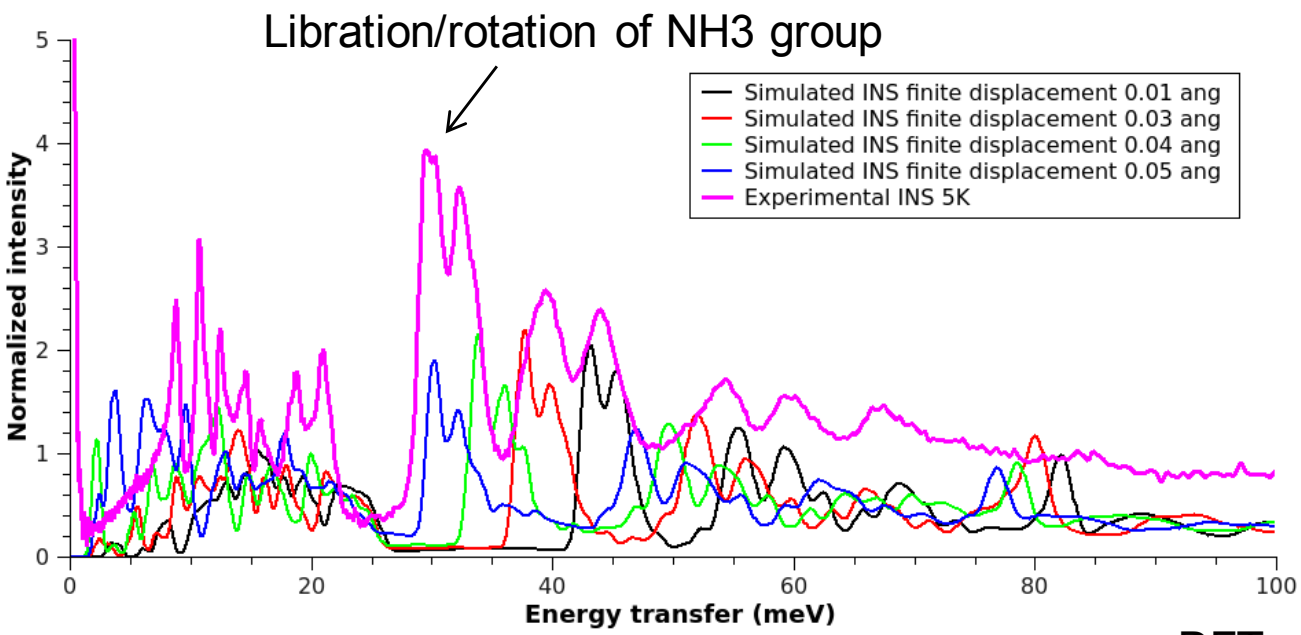
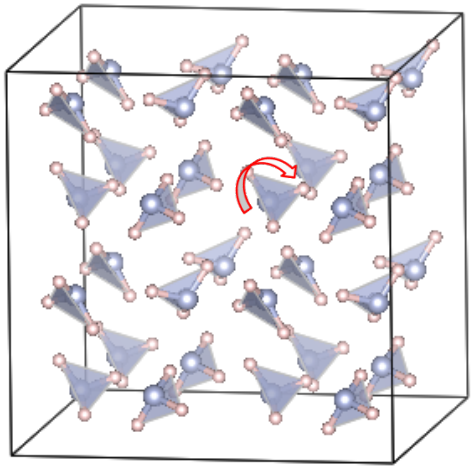
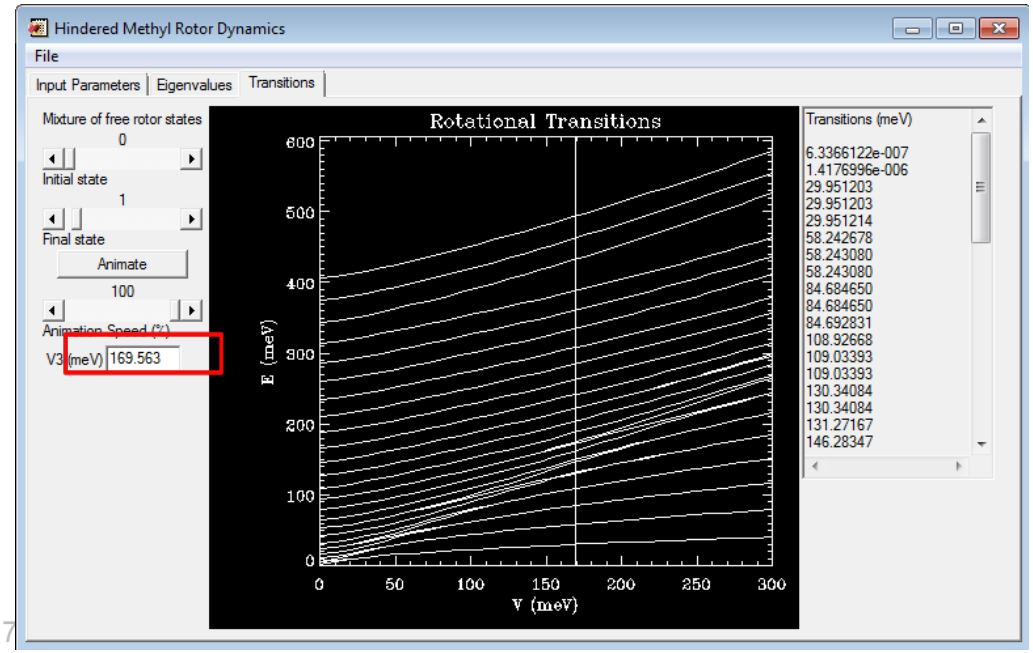
Beyond harmonic approximation or DFT

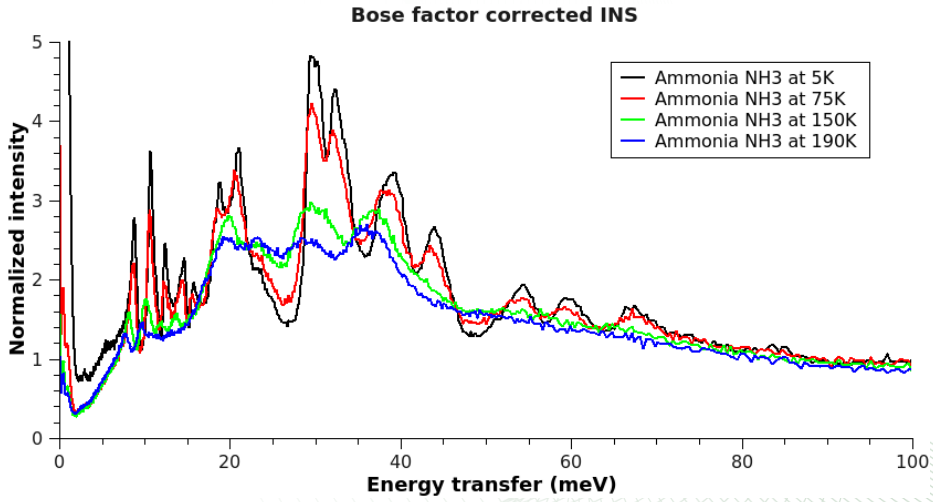




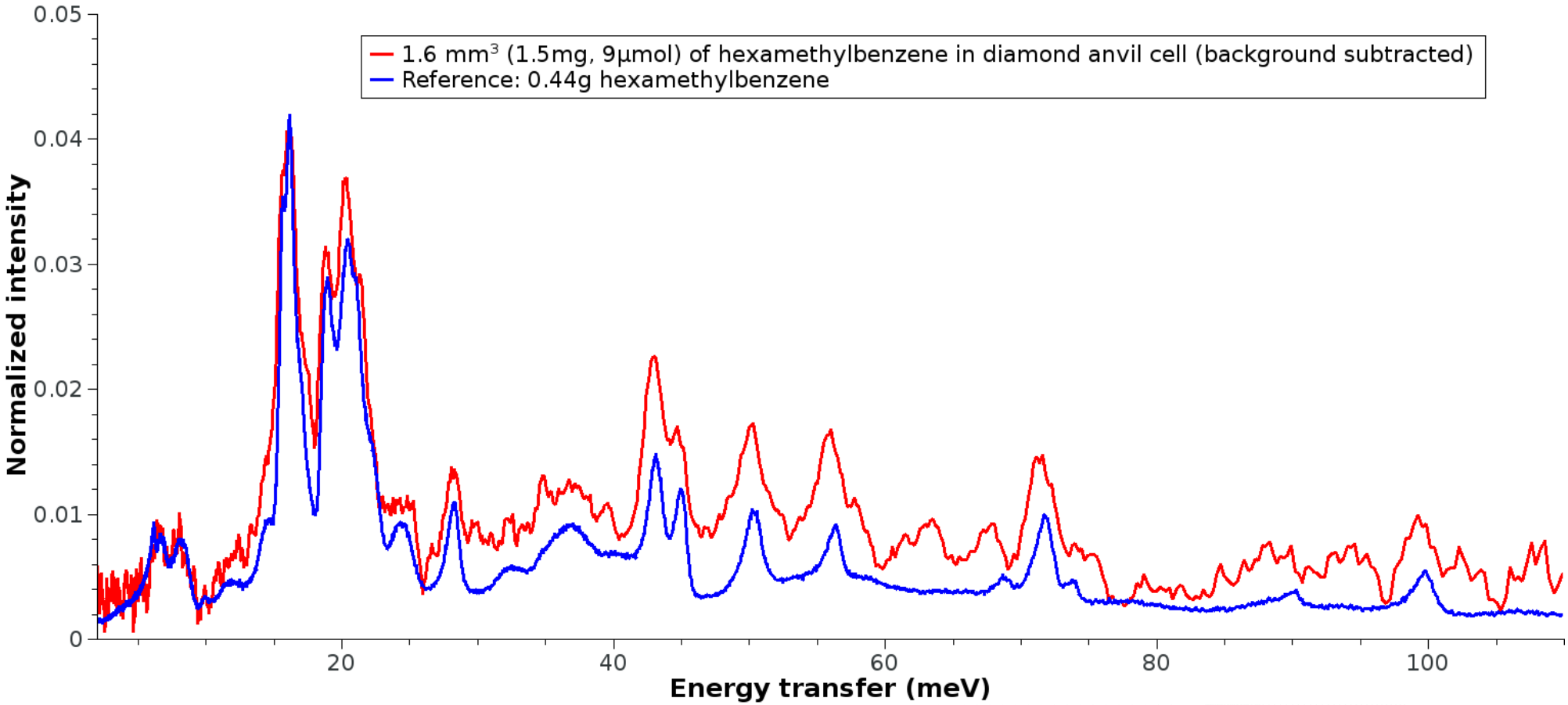
A 2x2x2 supercell of the ammonia (NH3)



- DFT calculated energy barrier for rigid rotation of NH3: 180 meV
- Energy barrier solved from the rotor model : 170 meV

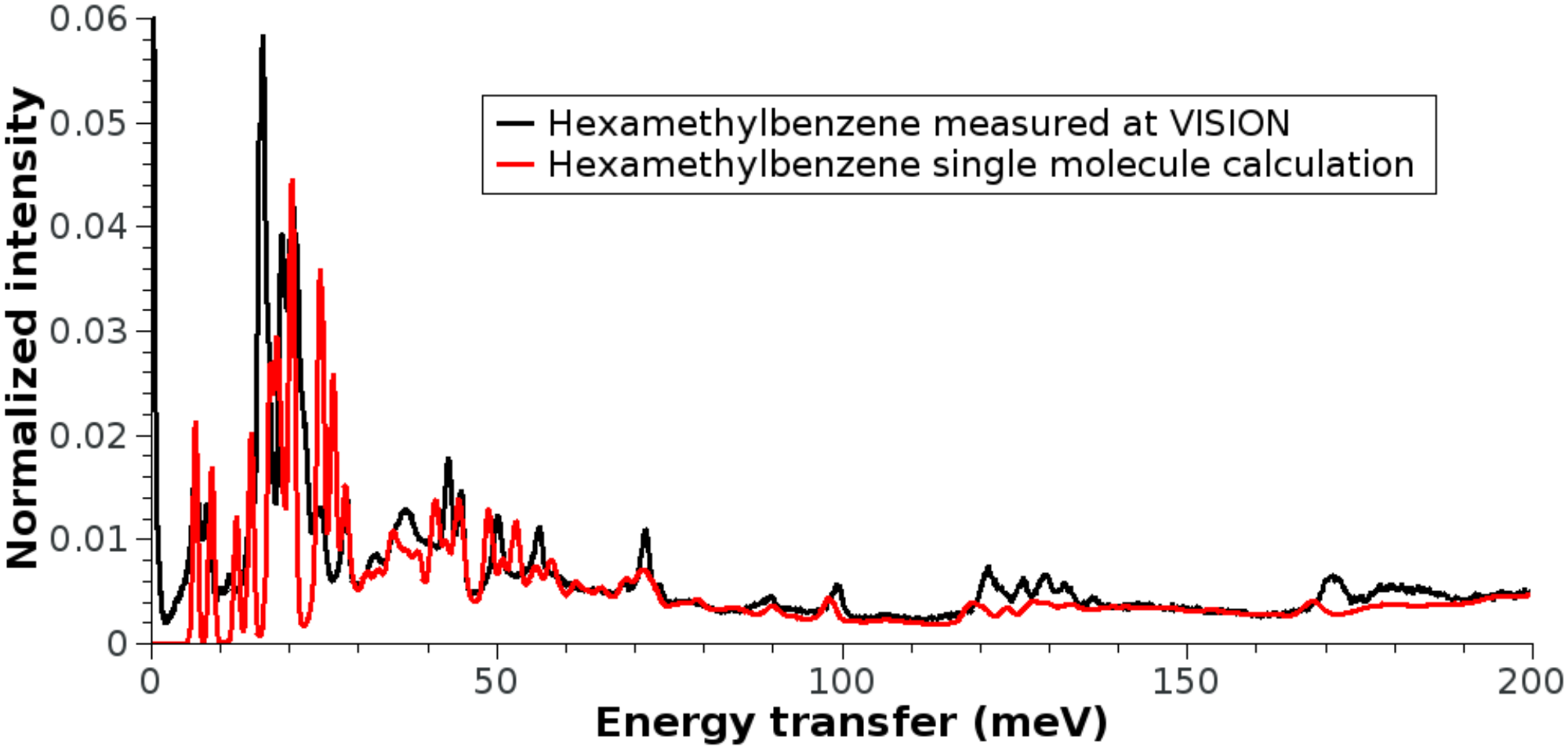


Test of DAC at VISION for high pressure INS experiments

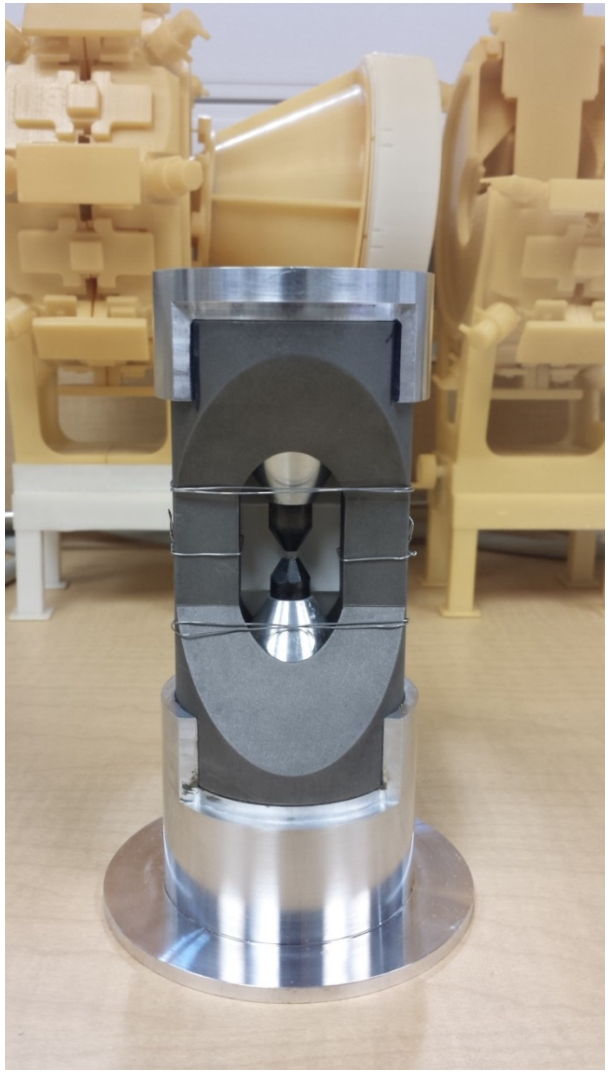


This is the actual sample size for a high pressure experiment with DAC!
1.6 mm^3 Hexamethylbenzene

Calculation (this morning)

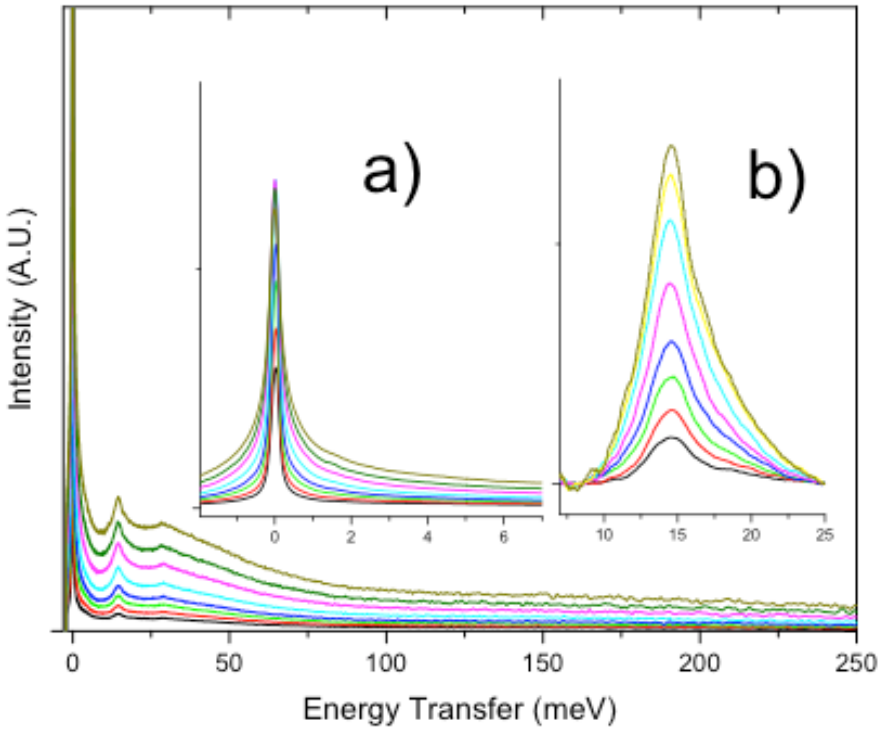


Test of DAC at VISION for high pressure INS experiments

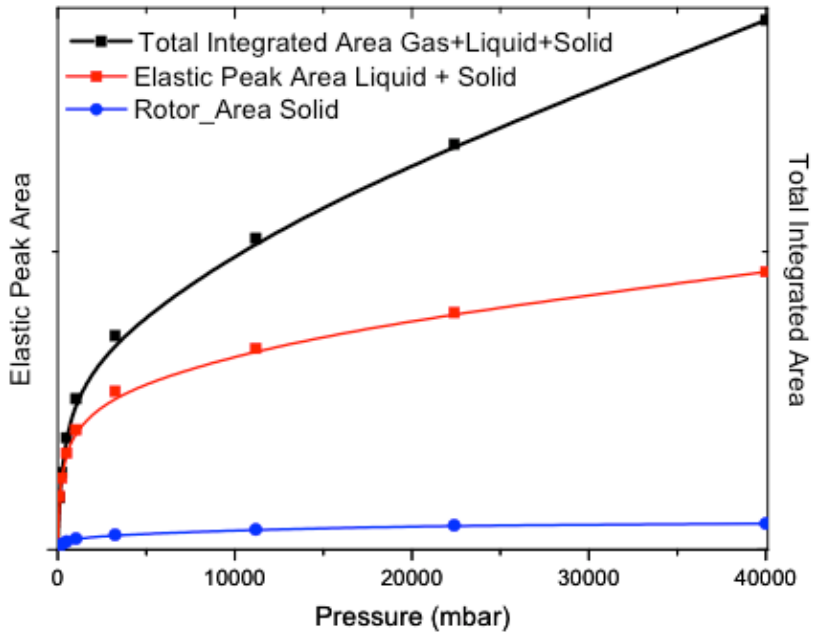


Largest single crystal diamond for DAC!
Cd sheets and B4C funnel added to minimize the background.

Molecular hydrogen in porous carbon



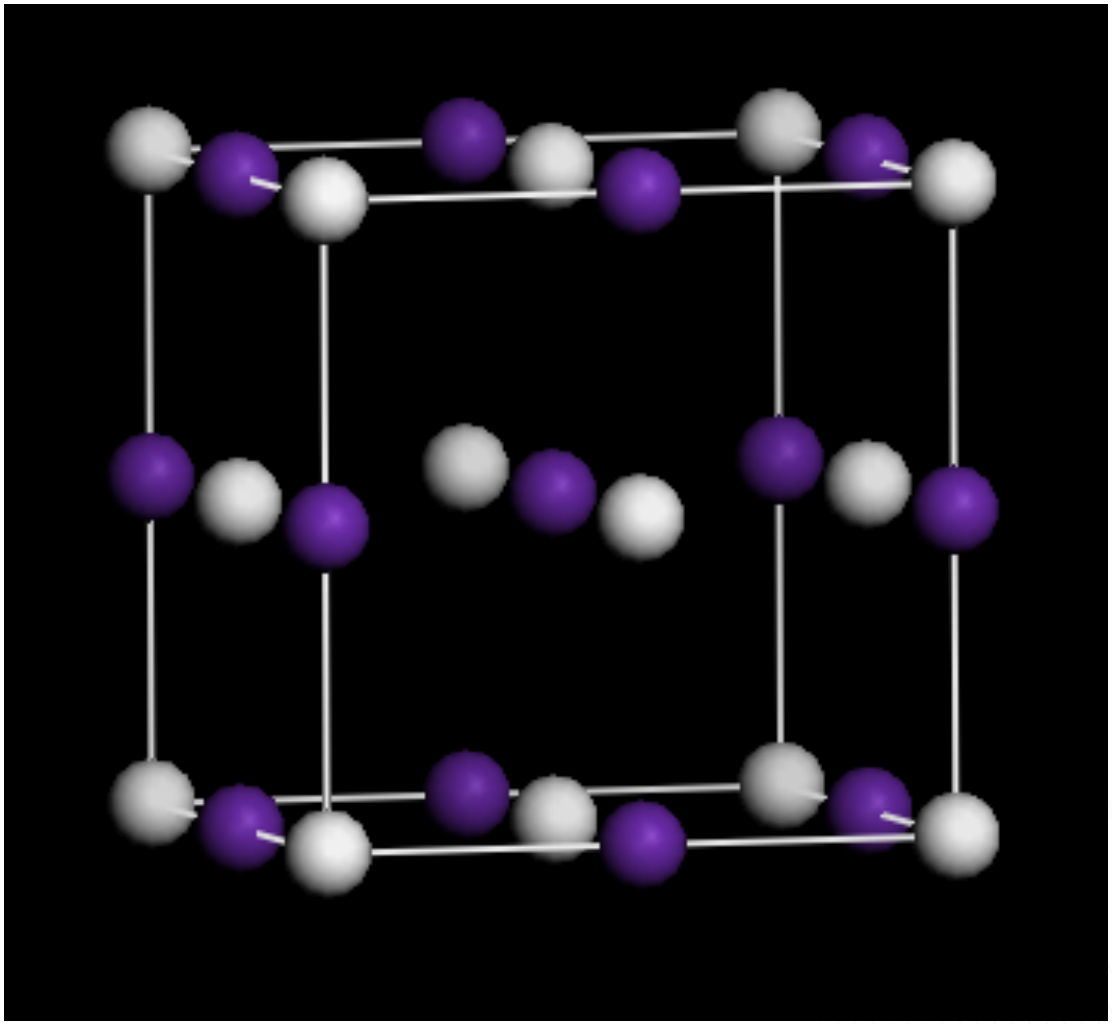
Presence of elastic line at 77K is indication of highly dense molecular hydrogen in the pores.
The broadening of the elastic line is a consequence of the enhanced mobility of the molecules as the amount of hydrogen increases in the system. Larger pores, where hydrogen is less constrained have more mobility. In the gas the signal is extremely broad.

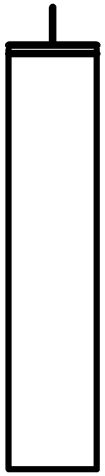


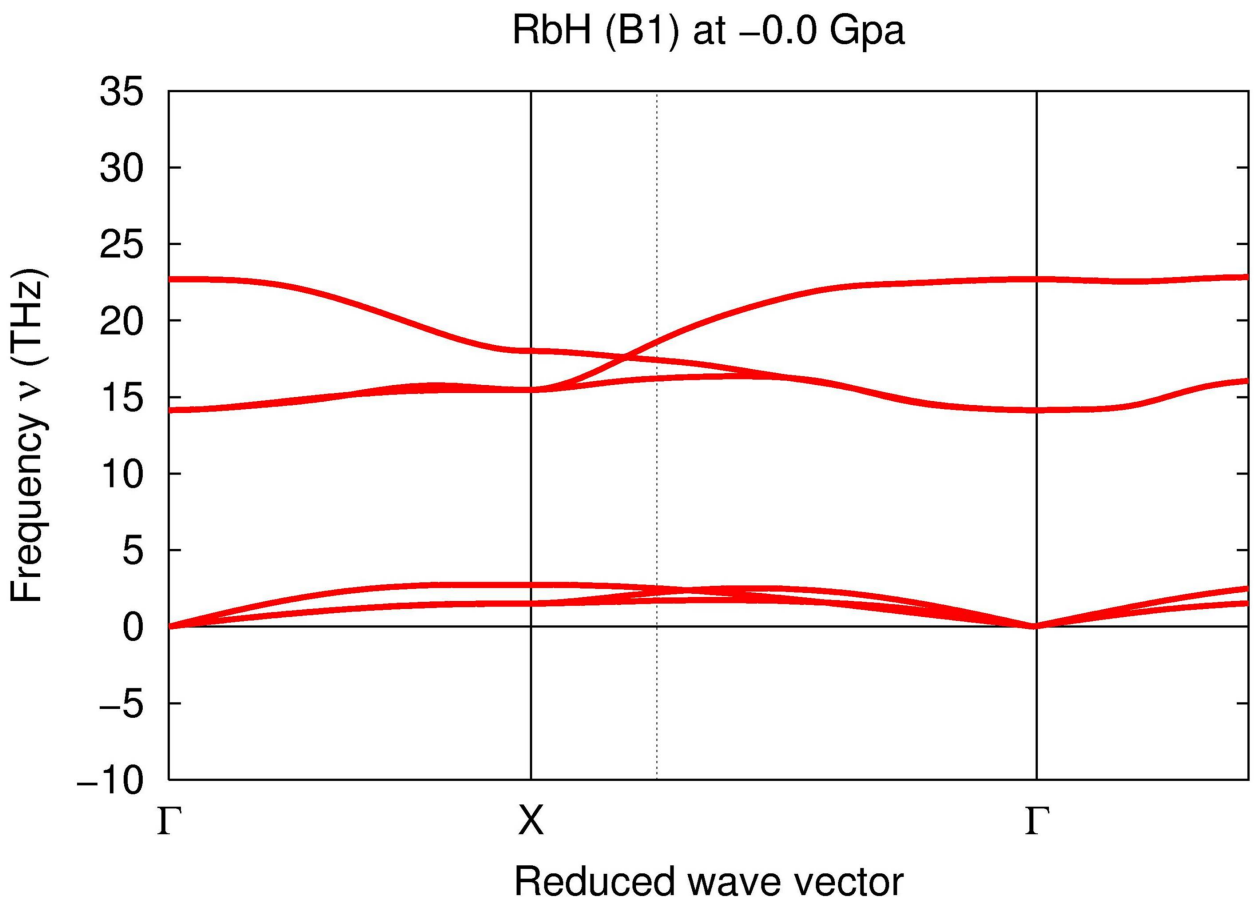
Presence of the rotor line at 77K is indication of completely immobile molecular hydrogen in the pores. In the case of pure para-hydrogen (previous figure) the line disappears when the hydrogen melts. The load keeps increasing even at 40 bar.

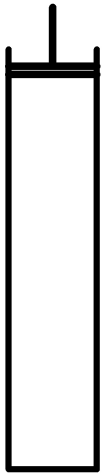
1. The total integral of the spectral intensity is proportional to the amount of hydrogen in the system
2. The integrated area under the elastic peak is proportional to the amount of hydrogen that is in a liquid and solid like phase
3. The integrated area under the rotor line is proportional to the amount of hydrogen in solid like phase (right panel)

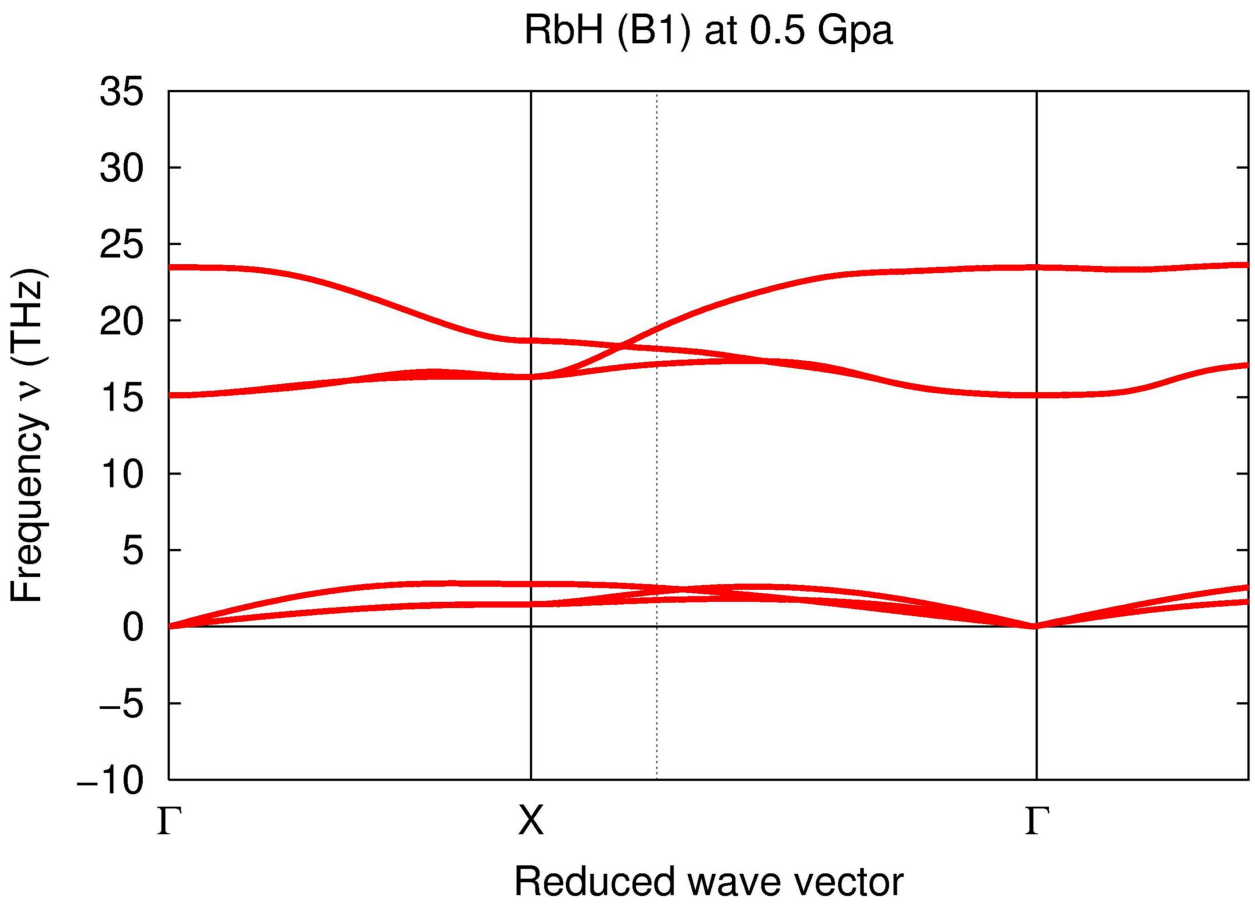
Dispersion curves of RbH vs P NaCl (B1) structure

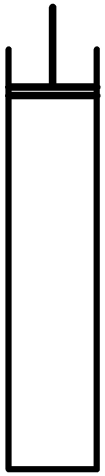


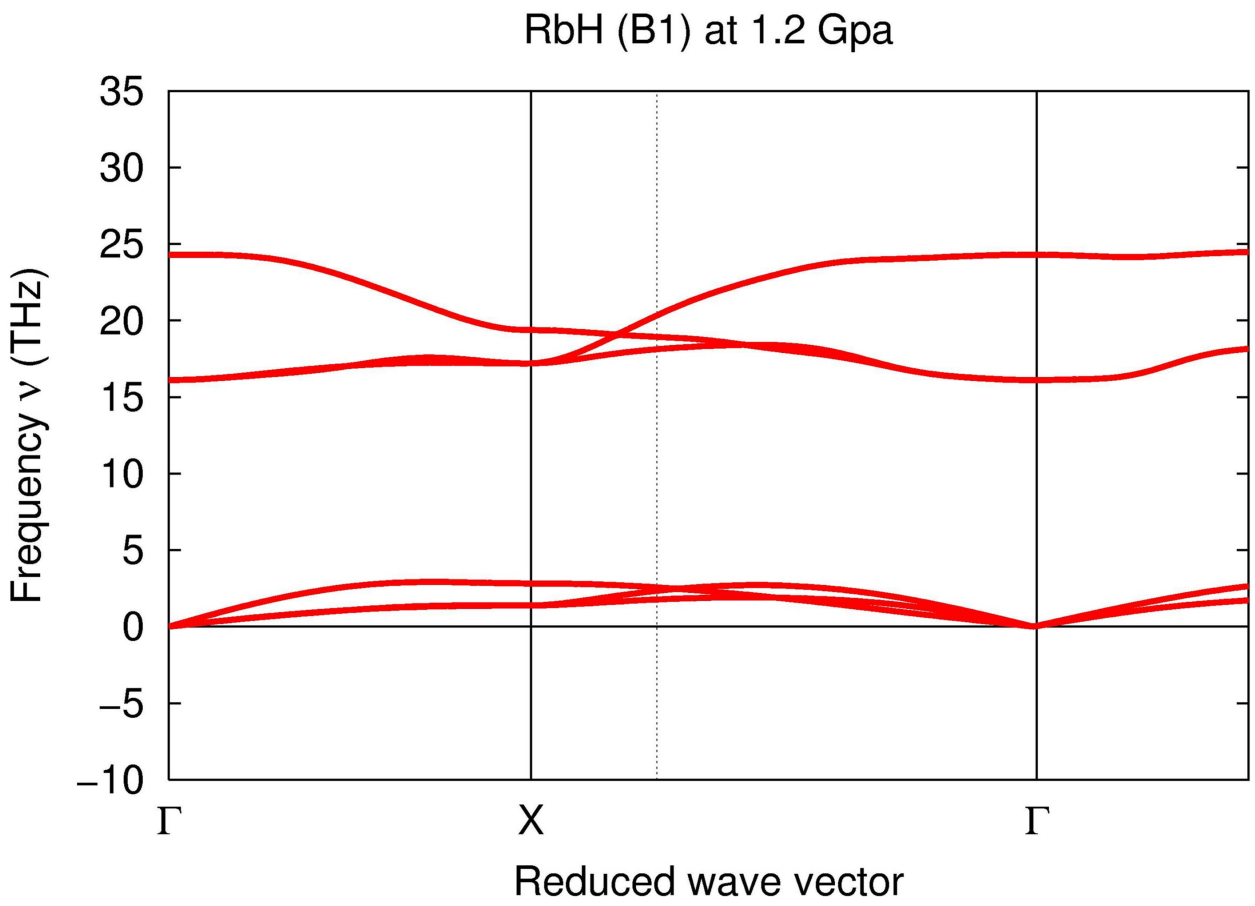


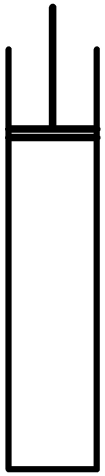

RbH (B1) at –0.0 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
–5
–10
Γ
X
Γ
Reduced wave vector

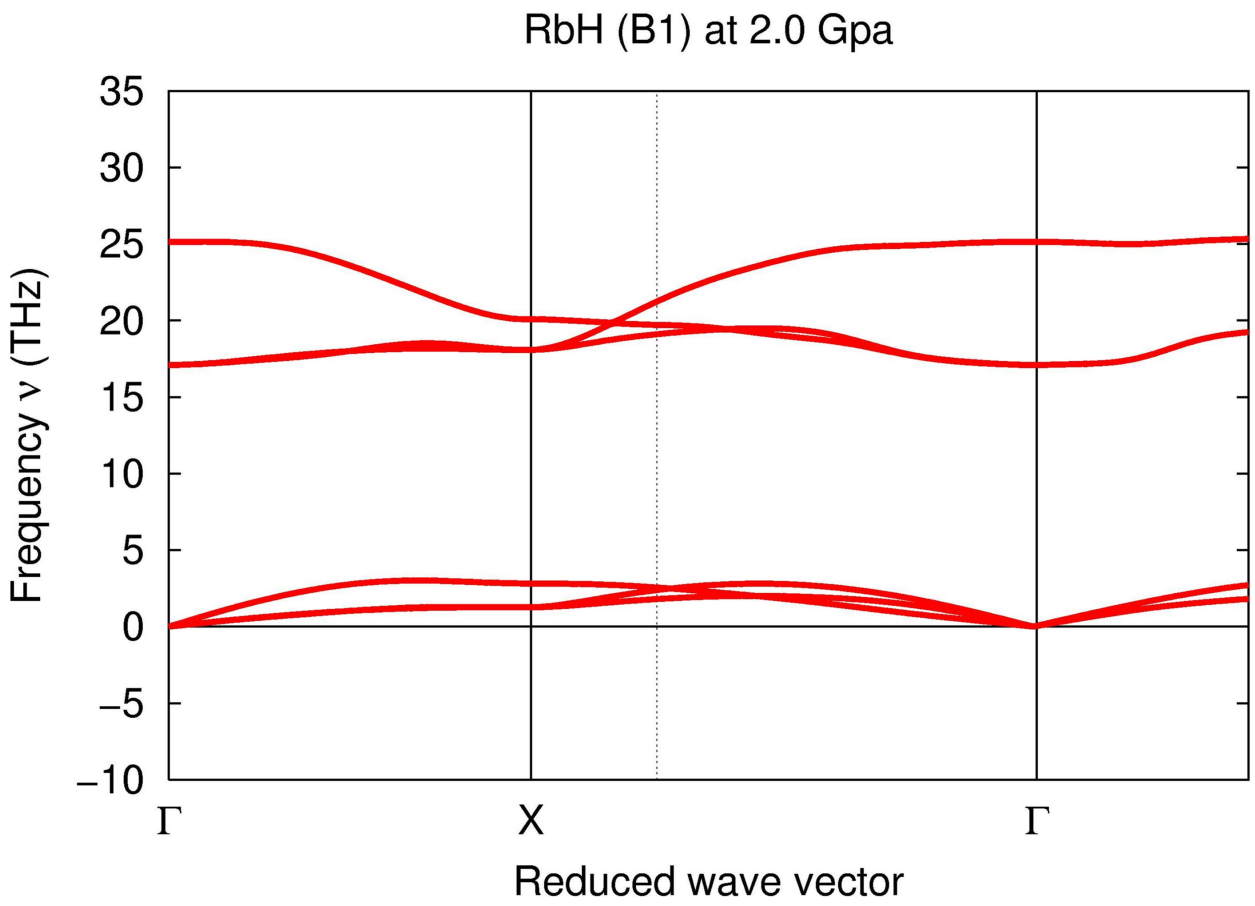


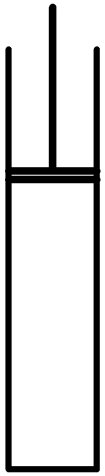

RbH (B1) at 0.5 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

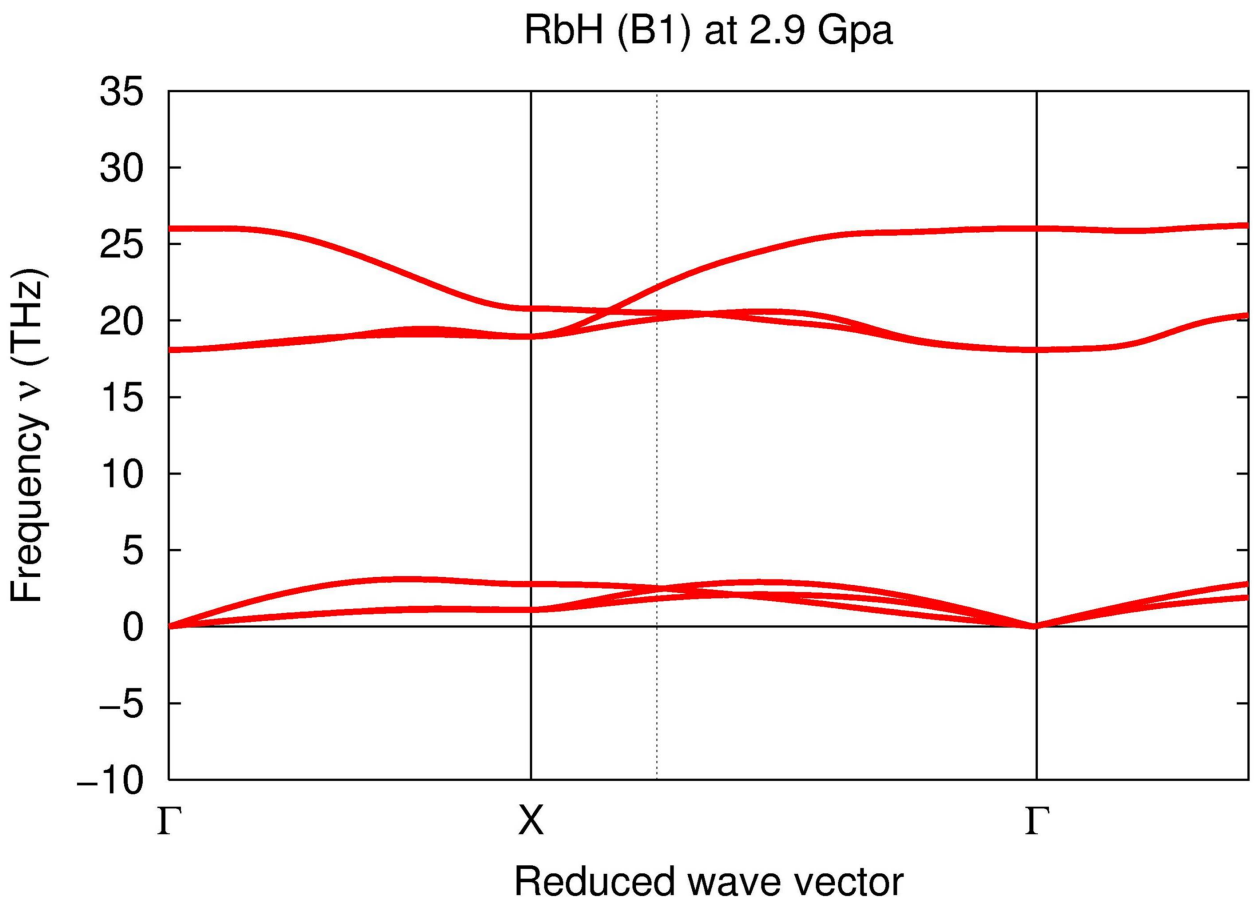


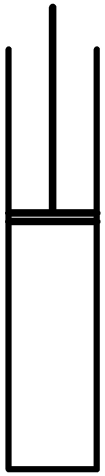

RbH (B1) at 1.2 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

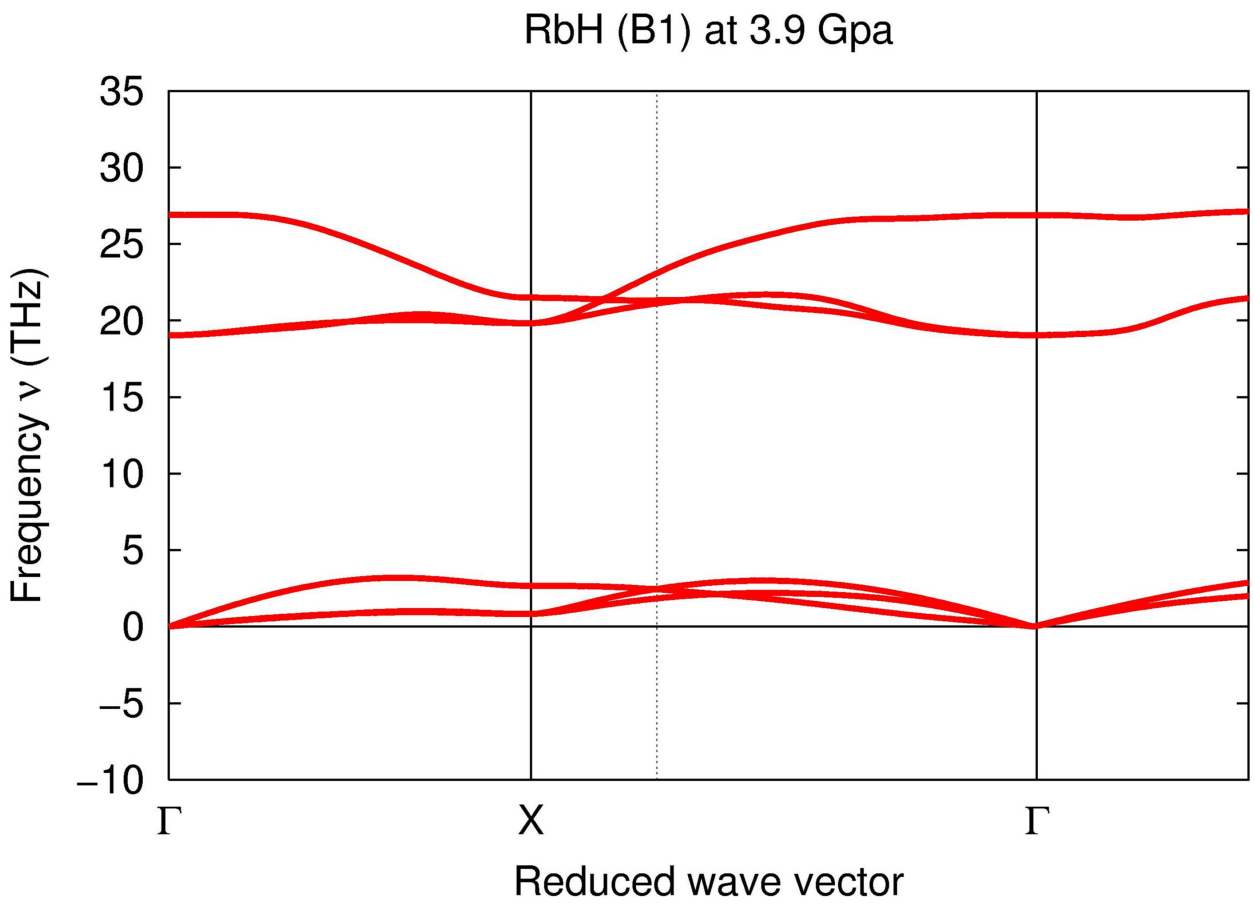


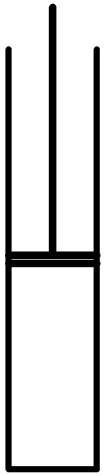

RbH (B1) at 2.0 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

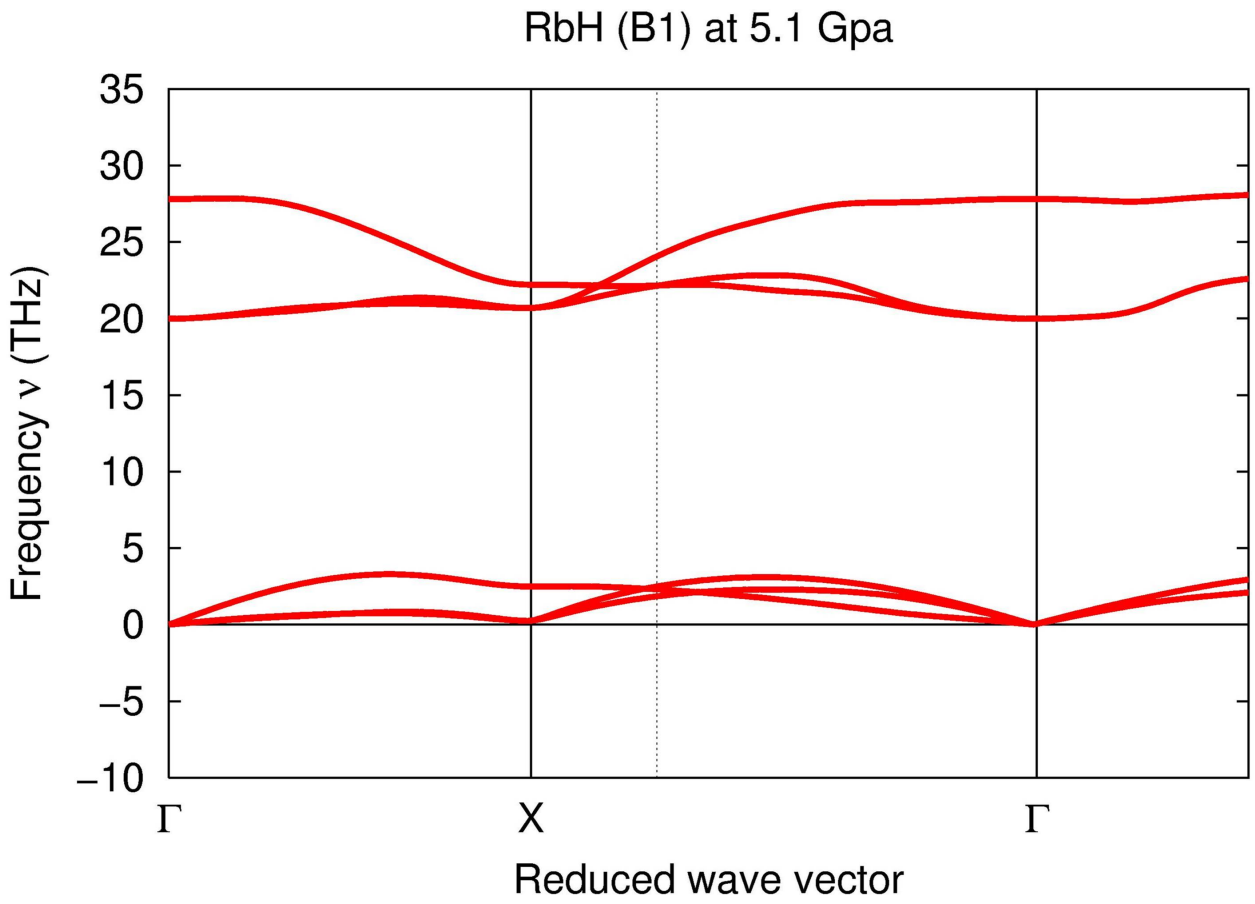


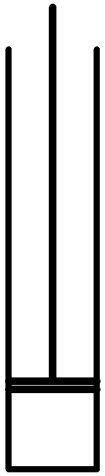

RbH (B1) at 2.9 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

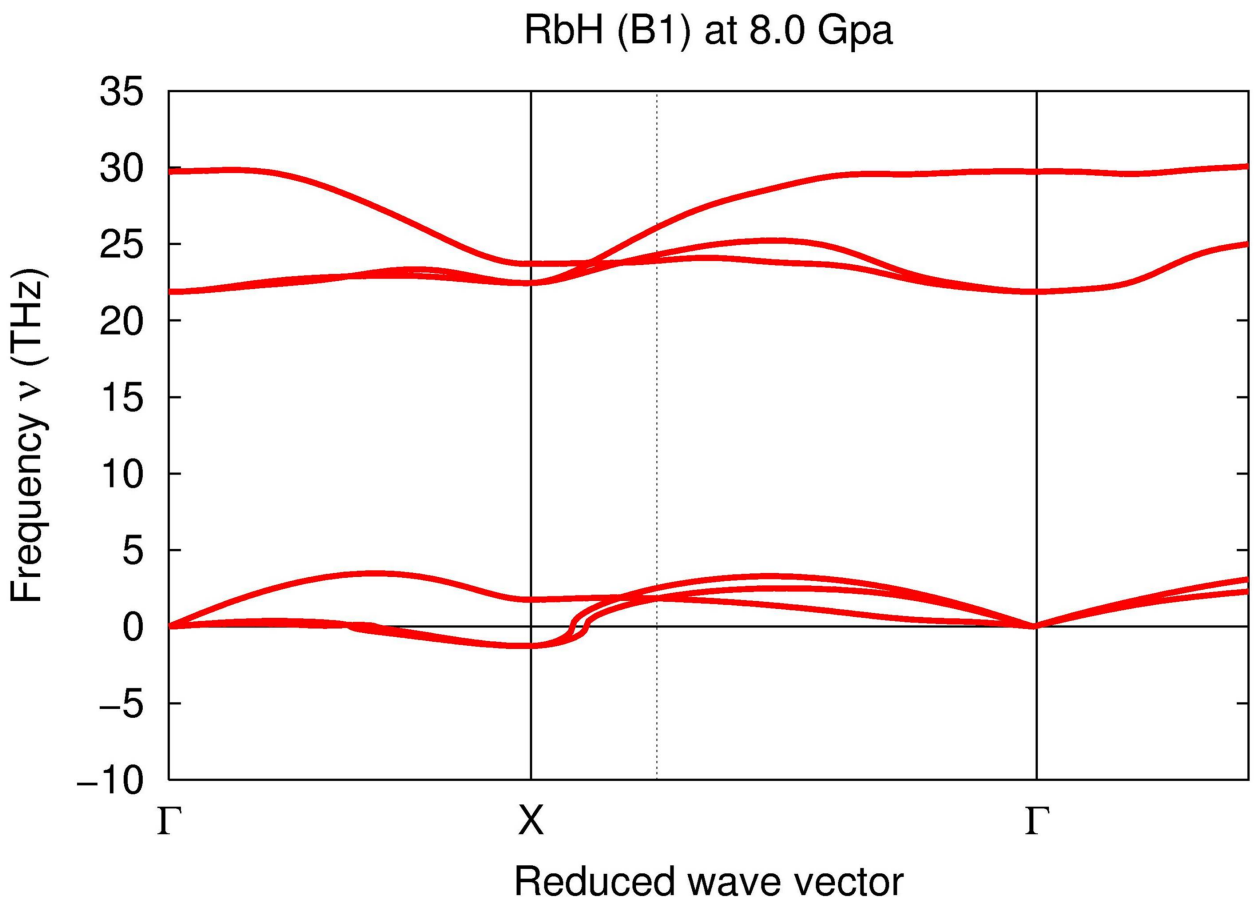



RbH (B1) at 3.9 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

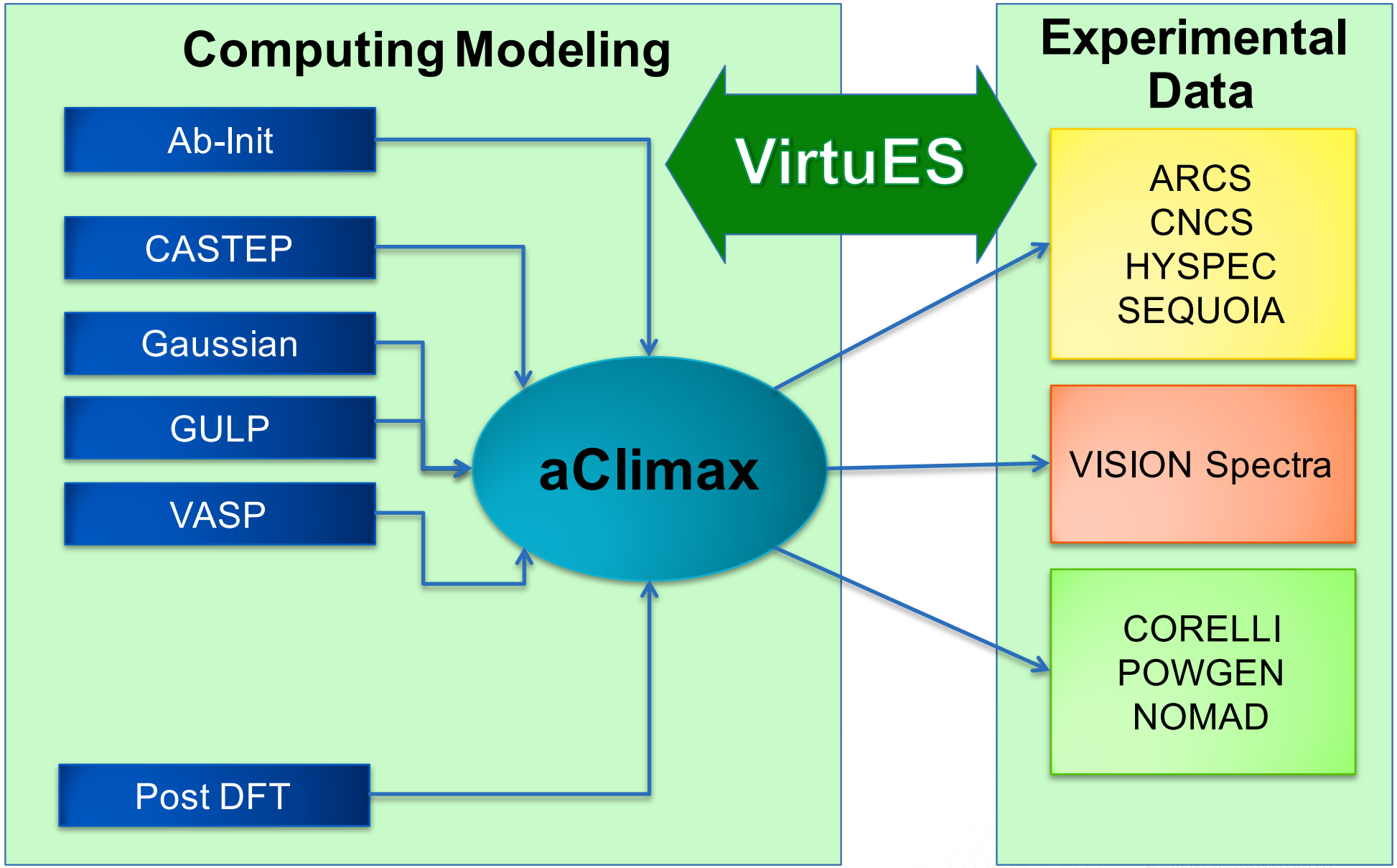



RbH (B1) at 5.1 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector




RbH (B1) at 8.0 Gpa
Frequency ν (THz)
35
30
25
20
15
10
5
0
−5
−10
Γ
X
Γ
Reduced wave vector

The FUTURE



Low energy structural dynamics and constrained libration of Li(NH3)4, the lowest melting point metal

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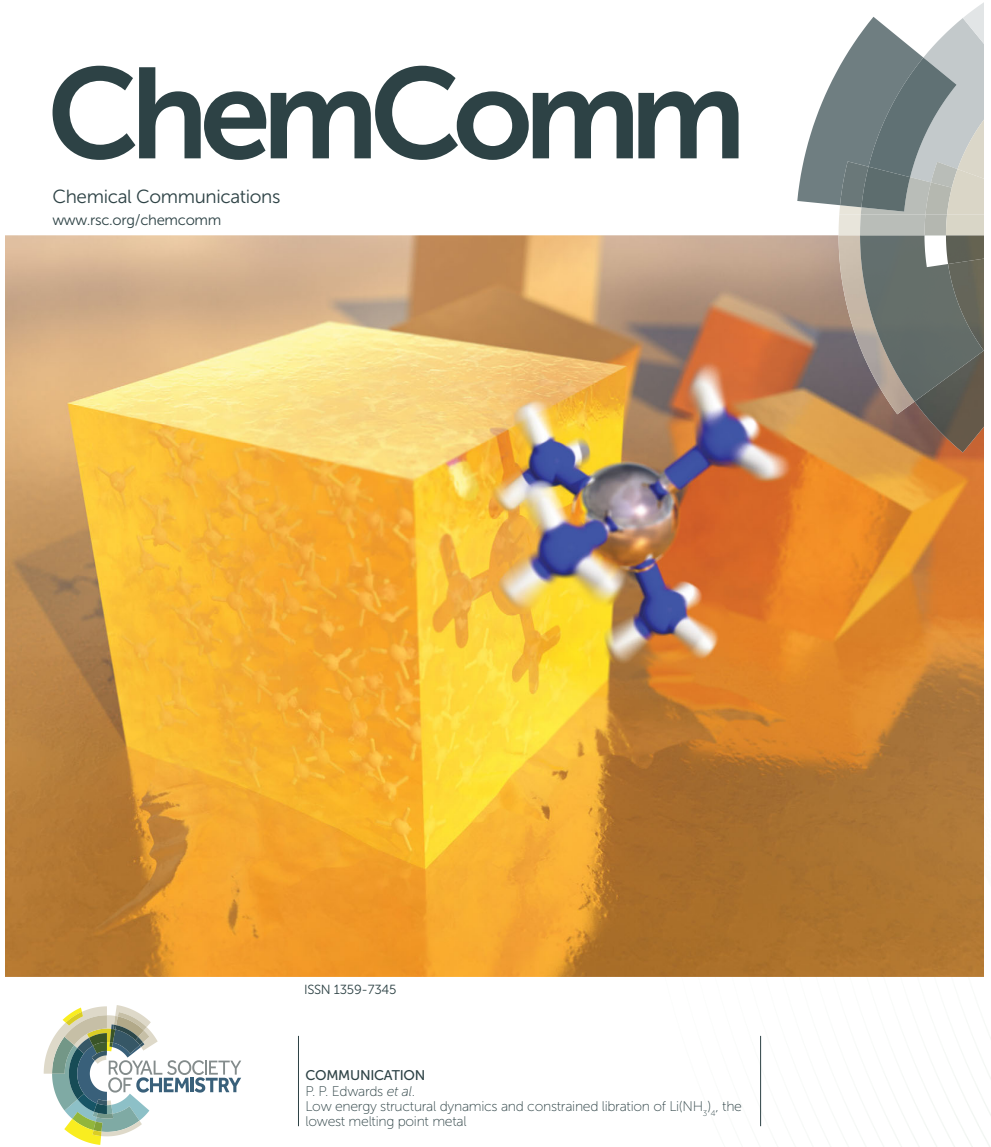

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INS requires DFT modeling to interpretation

Virtues (Virtual Experiments in Spectroscopy)
Computer modeling is crucial to understand and interpret INS data. The VirtuES cluster provides 2500+ cores and a number of DFT codes for VISION data analysis and interpretation. VISION is the first SNS instrument that has computer modeling as integral part of the data analysis and interpretation of the spectra.

neutrons.ornl.gov/vision