Inelastic Neutron Scattering Indirect Geometry Instrumentation

Erice School
“Neutron Science and Instrumentation
Neutrons for Chemistry And Materials
Science Applications”

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Neutron Scattering Division
Oak Ridge National Laboratory

July 9th, 2018

OAK RIDGE
National Laboratory

Myself


Start new
Distance
22,566.65 km (14,022.27 mi)
3D
Google
IBCAO Landsat / Copernicus PGC/NASA U.S. Geological Survey Data SIO, NOAA, U.S. Navy, NGA, GEBCO Camera : 16,198 km 16°06'53"N 80°47'42"W 100%

Myself


LEAGUE
IONSHIPS
USTA
LEAGUE


6.0 Mixed National In
USTA
LEAGUE


UNI
QLO
Wilson

Myself

- MSc Mathematics
- BSc Physics
- PhD Physical Chemistry
- Postdoc at University of Milwaukee
- Assistant professor Argentina
- Fellow CONICET, Argentina
- Postdoc Reading University
- TOSCA Instrument Scientist at ISIS

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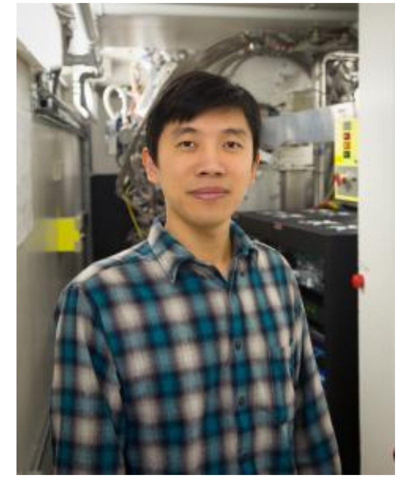
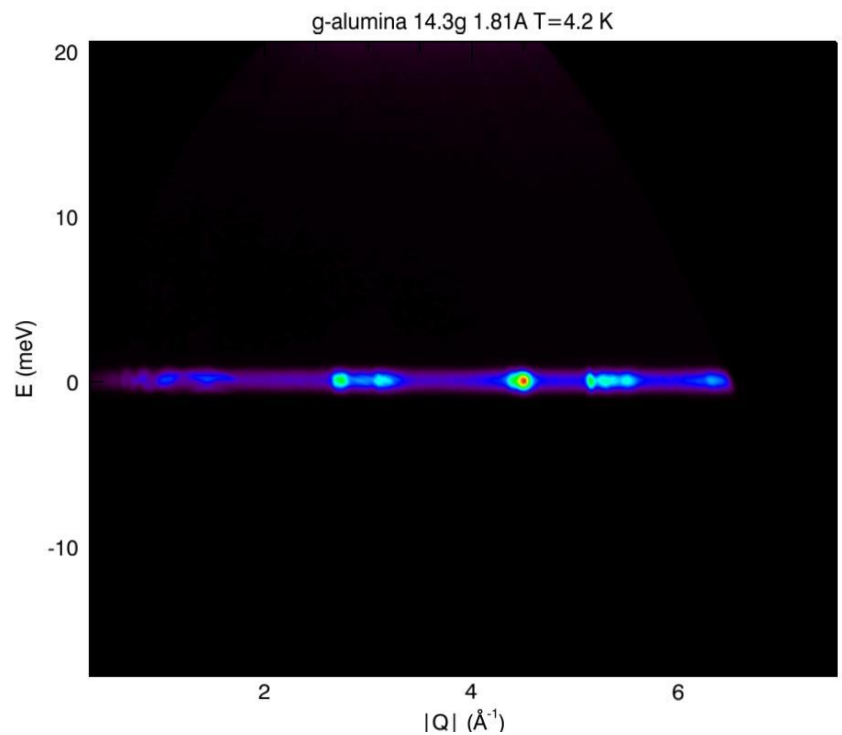
- NScD, ORNL

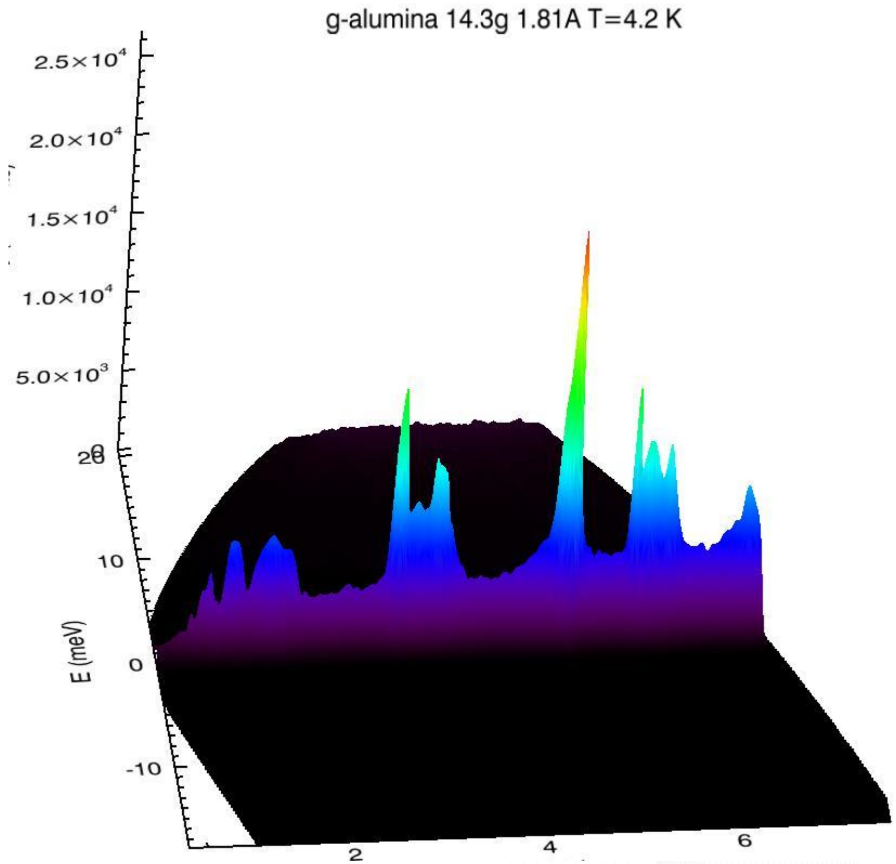







The S(Q,ω) Map

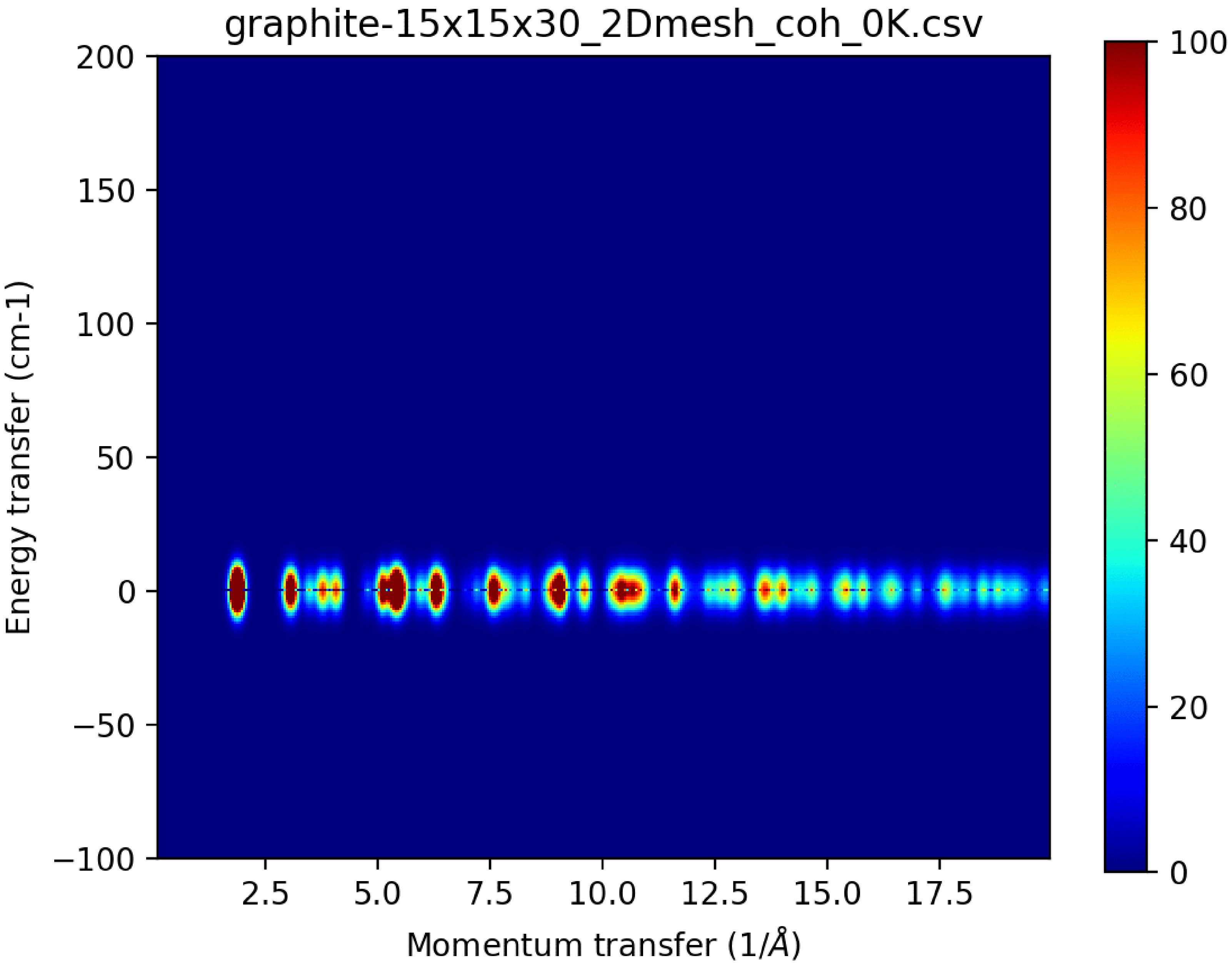


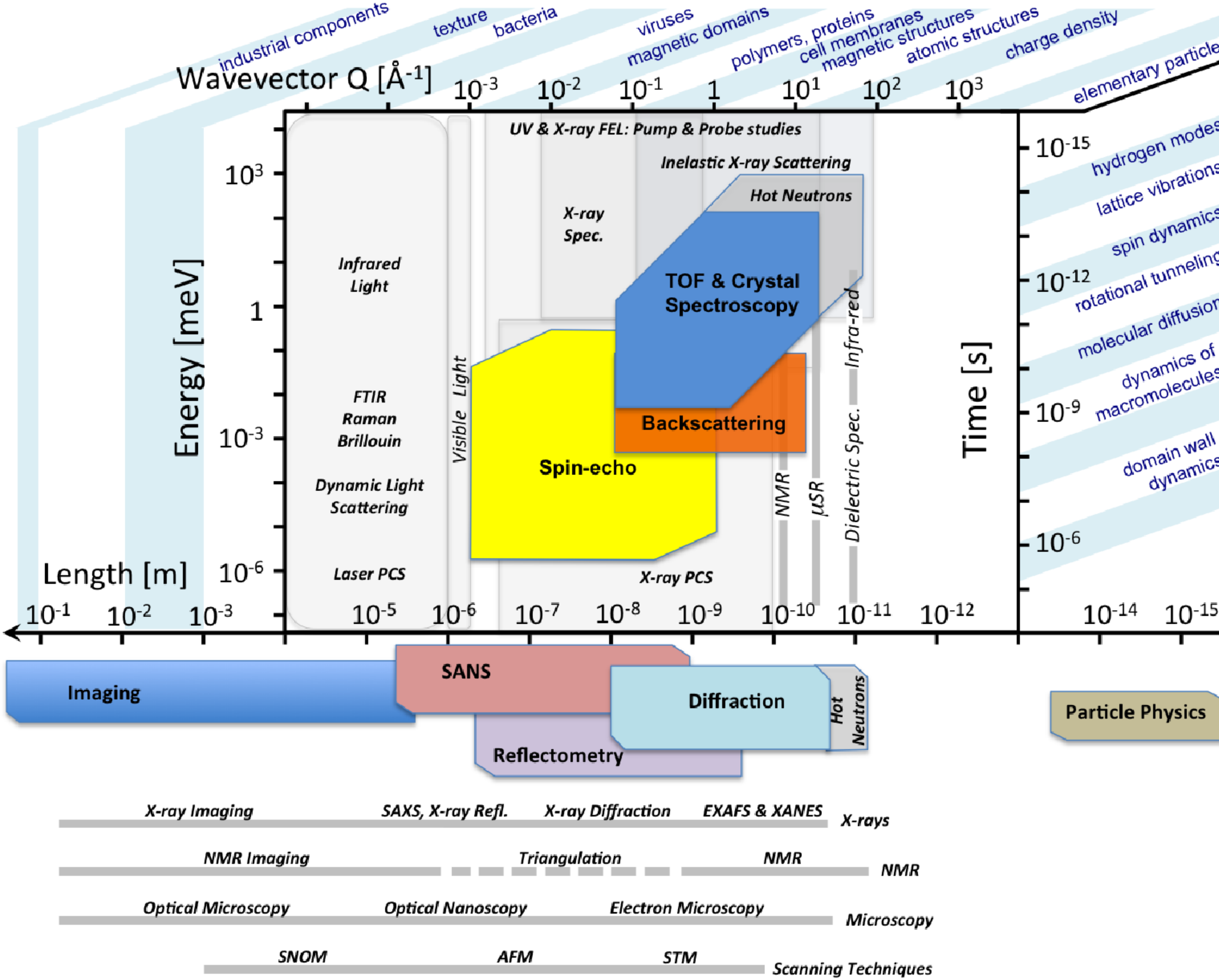


ω=0 Elastic Scattering Diffraction

Structural Information

"Let there be light"

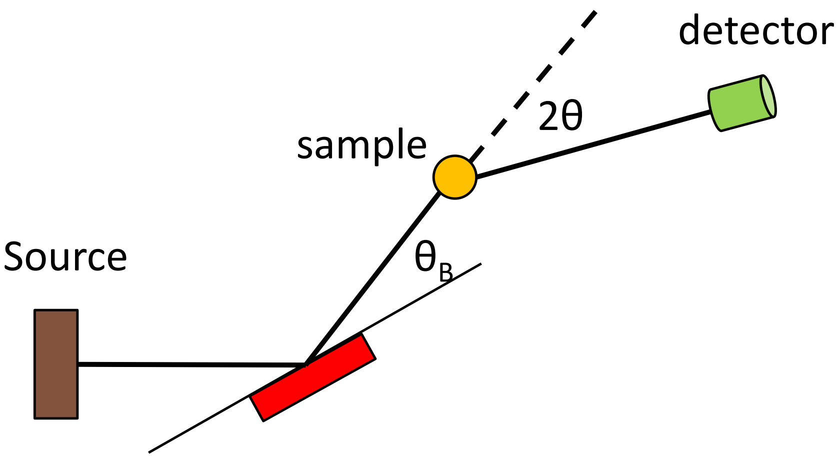



industrial components
texture
bacteria
viruses
magnetic domains
polymers, proteins
cell membranes
magnetic structures
atomic structures
charge density
elementary particles
Wavevector Q [Å-1]
10-3
10-2
10-1
1
101
102
103
UV & X-ray FEL: Pump & Probe studies
Inelastic X-ray Scattering
Hot Neutrons
X-ray Spec.
Infrared Light
TOF & Crystal Spectroscopy
Visible Light
Infra-red
FTIR
Raman
Brillouin
Backscattering
Spin-echo
Dynamic Light Scattering
NMR
μSR
Dielectric Spec.
Laser PCS
X-ray PCS
Energy [meV]
103
1
10-3
10-6
Time [s]
10-15
10-12
10-9
10-6
hydrogen modes
lattice vibrations
spin dynamics
rotational tunneling
molecular diffusion
dynamics of macromolecules
domain wall dynamics
Length [m]
10-1
10-2
10-3
10-5
10-6
10-7
10-8
10-9
10-10
10-11
10-12
10-14
10-15
Imaging
SANS
Diffraction
Hot Neutrons
Reflectometry
Particle Physics
X-ray Imaging
SAXS, X-ray Refl.
X-ray Diffraction
EXAFS & XANES
X-rays
NMR Imaging
Triangulation
NMR
NMR
Optical Microscopy
Optical Nanoscopy
Electron Microscopy
Microscopy
SNOM
AFM
STM
Scanning Techniques

The easy part: Neutron Diffraction

Instrument layouts

CW

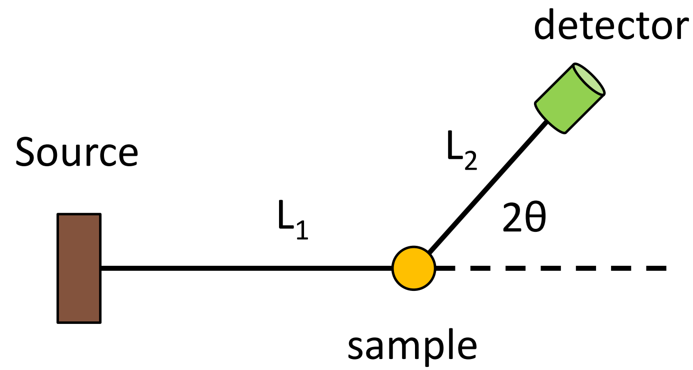


$$l = \frac{2 d_c \sin \theta_B}{n}$$

$$\frac{\Delta\lambda}{\lambda} = \frac{\delta d}{d} + \cot\theta\delta\theta$$

Correlation between l and θ_B

TOF



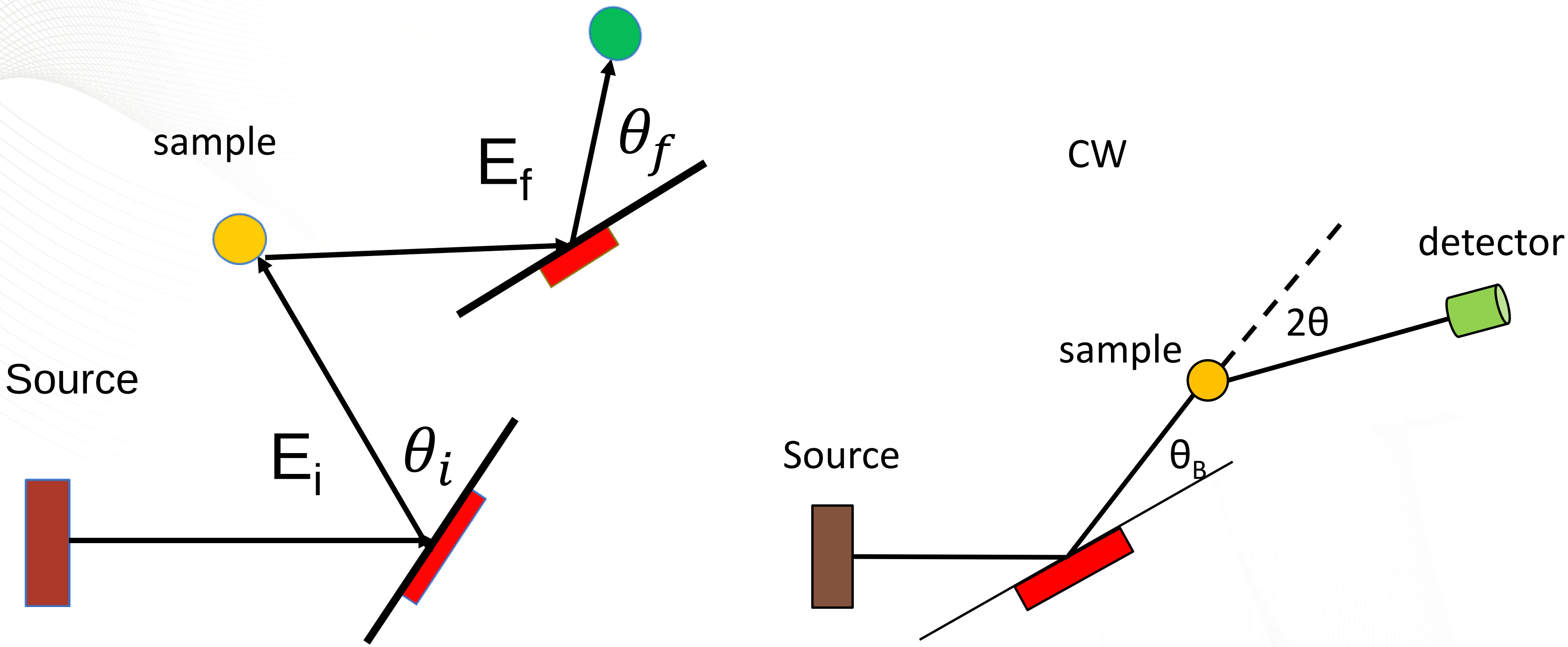
$$l = \frac{3956}{v} = \frac{3956\,(t - t_0)}{L_1 + L_2}$$

$$\Delta\lambda \sim \delta t_0, \delta t, \delta L$$

No correlation between l and θ

Science & Technology Facilities Council ISIS

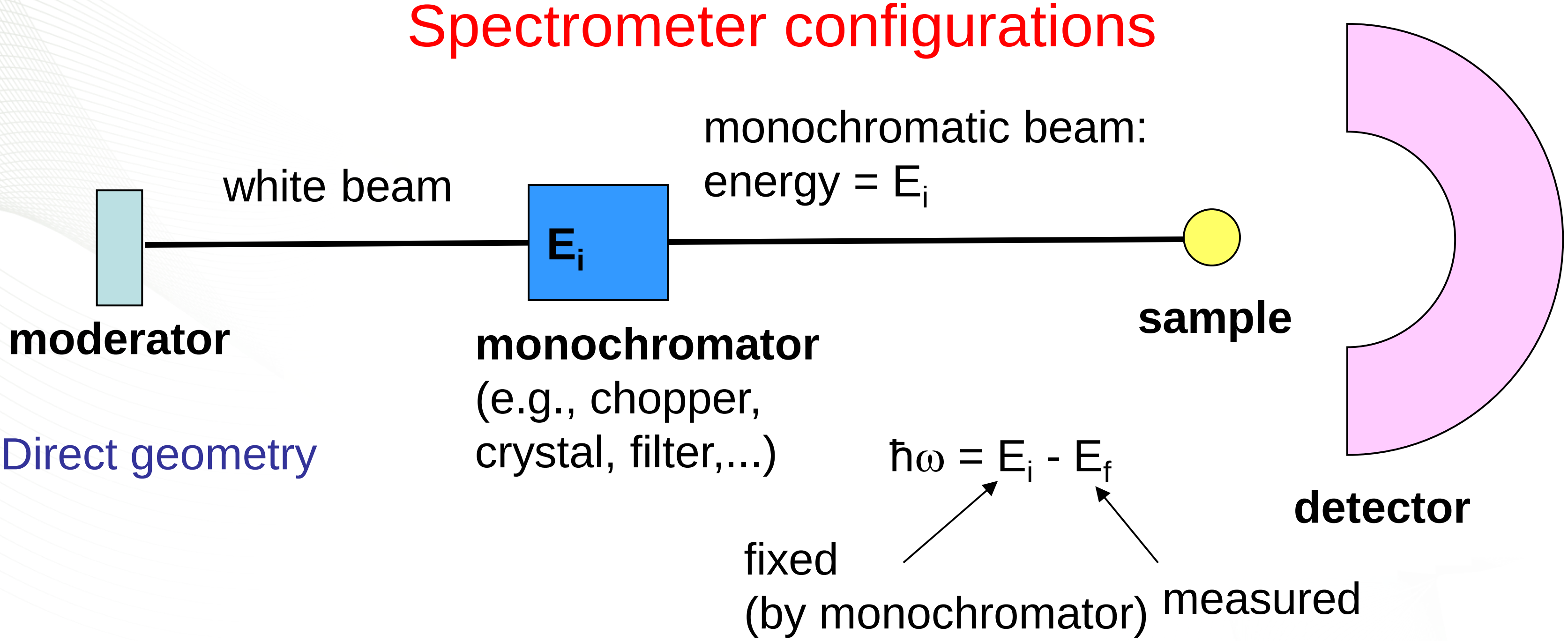
The hard part: Neutron Spectroscopy

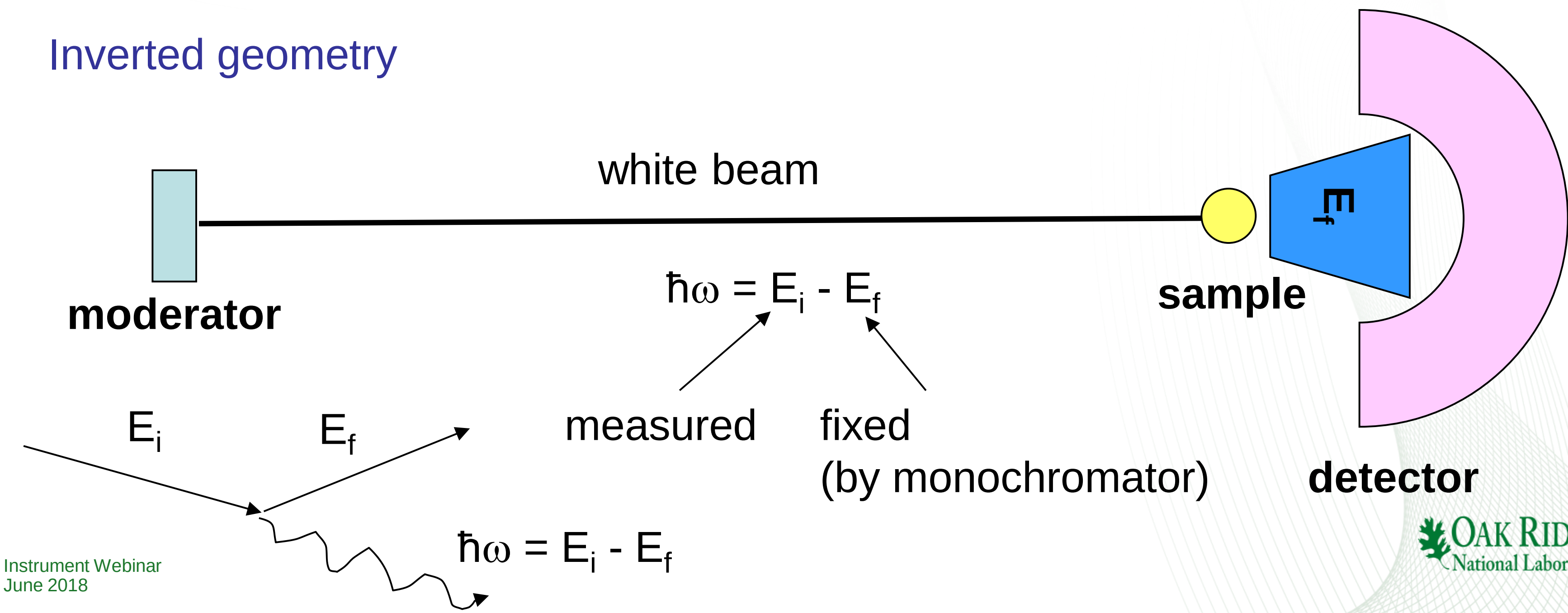


In order to discriminate the final energy, we need 2 crystals instead of one!
This is a triple axis instrument (in a continuous source)
We need to work twice as hard!

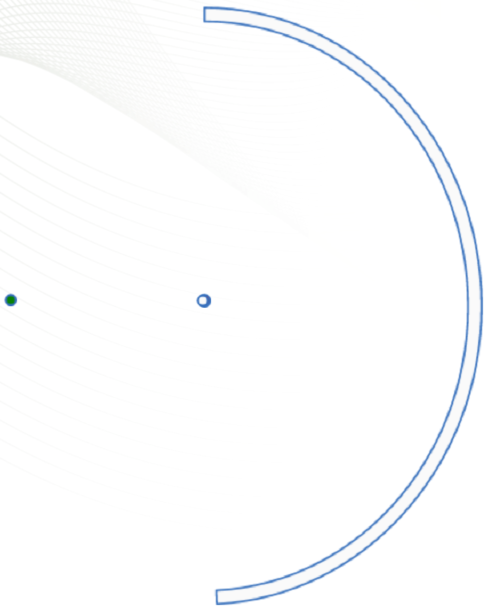
Instruments

Spectrometer configurations

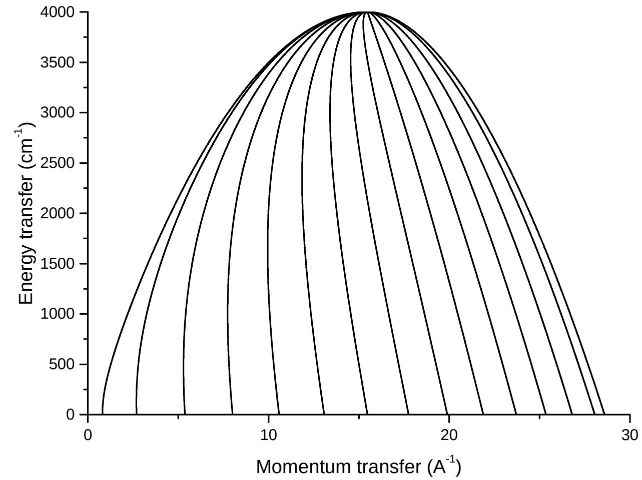




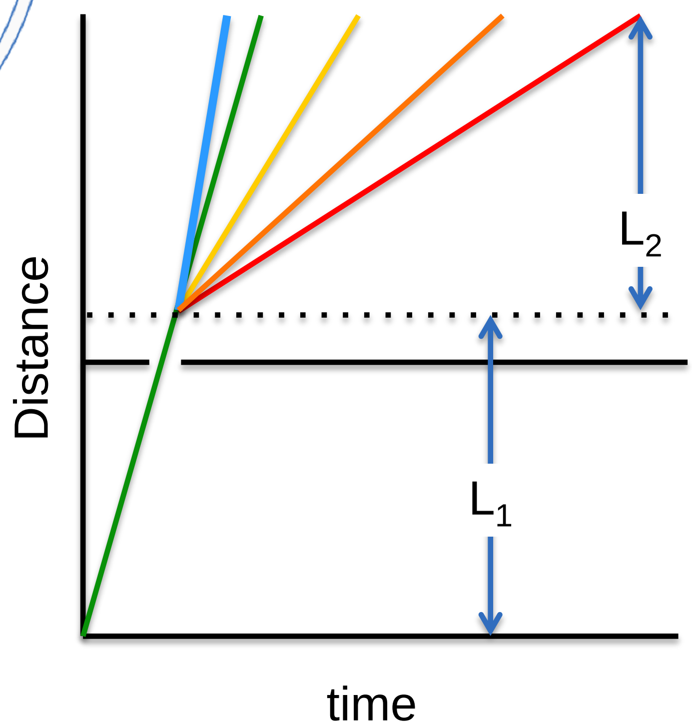
How to measure INS (1)
Direct Geometry Instrumentation

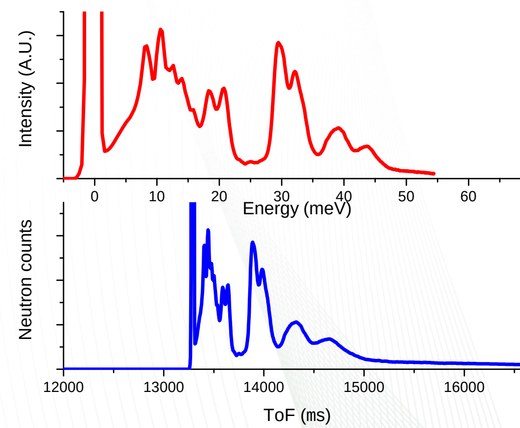


Direct geometry instruments measure Q trajectory is determined by the angle and energy transfer.
Examples: ARCS, CNCS, HYSPEC, SEQUIOA



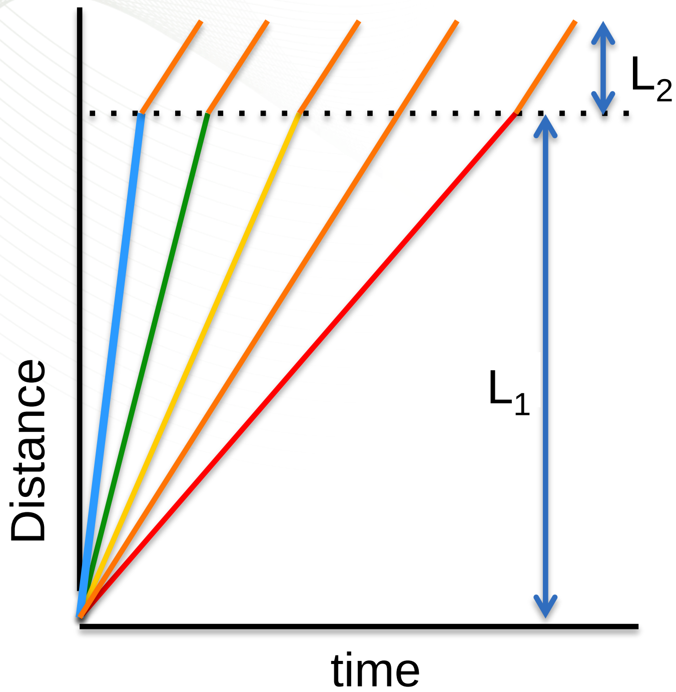
Incident neutron beam is monochromatic determining the incident energy E_1.
That determines T_1. We measure the ToF and we can work out T_2.



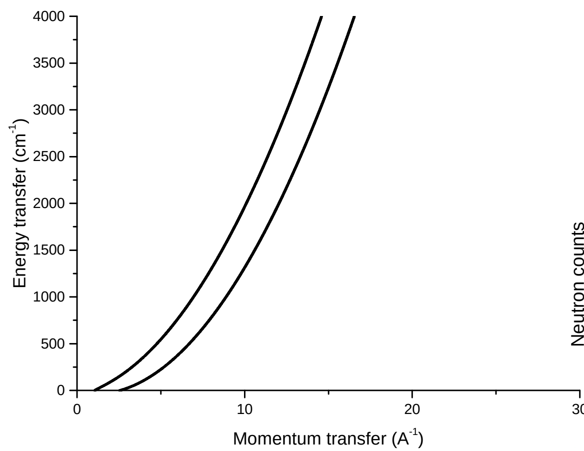


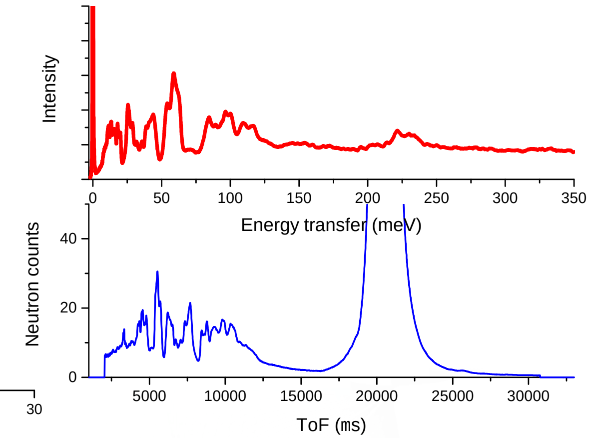
Resolution is almost constant in units of E_i

How to measure INS (2)
Indirect Geometry Instrumentation

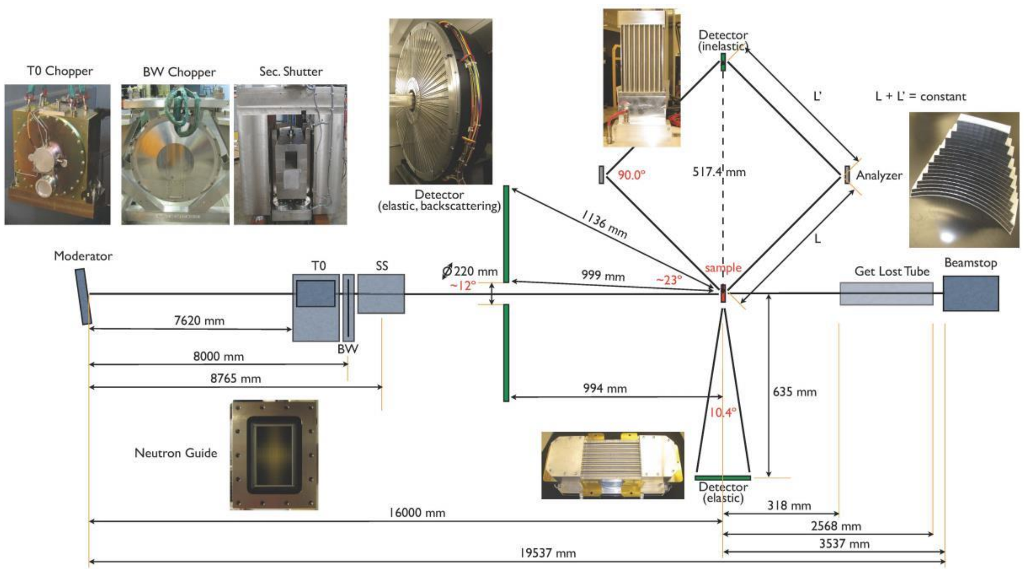


Incident neutron beam is white. We fix the energy of the scattered neutrons using a analyzer and filter device.
That fixes T_2. We measure the ToF and we can work out T_1.



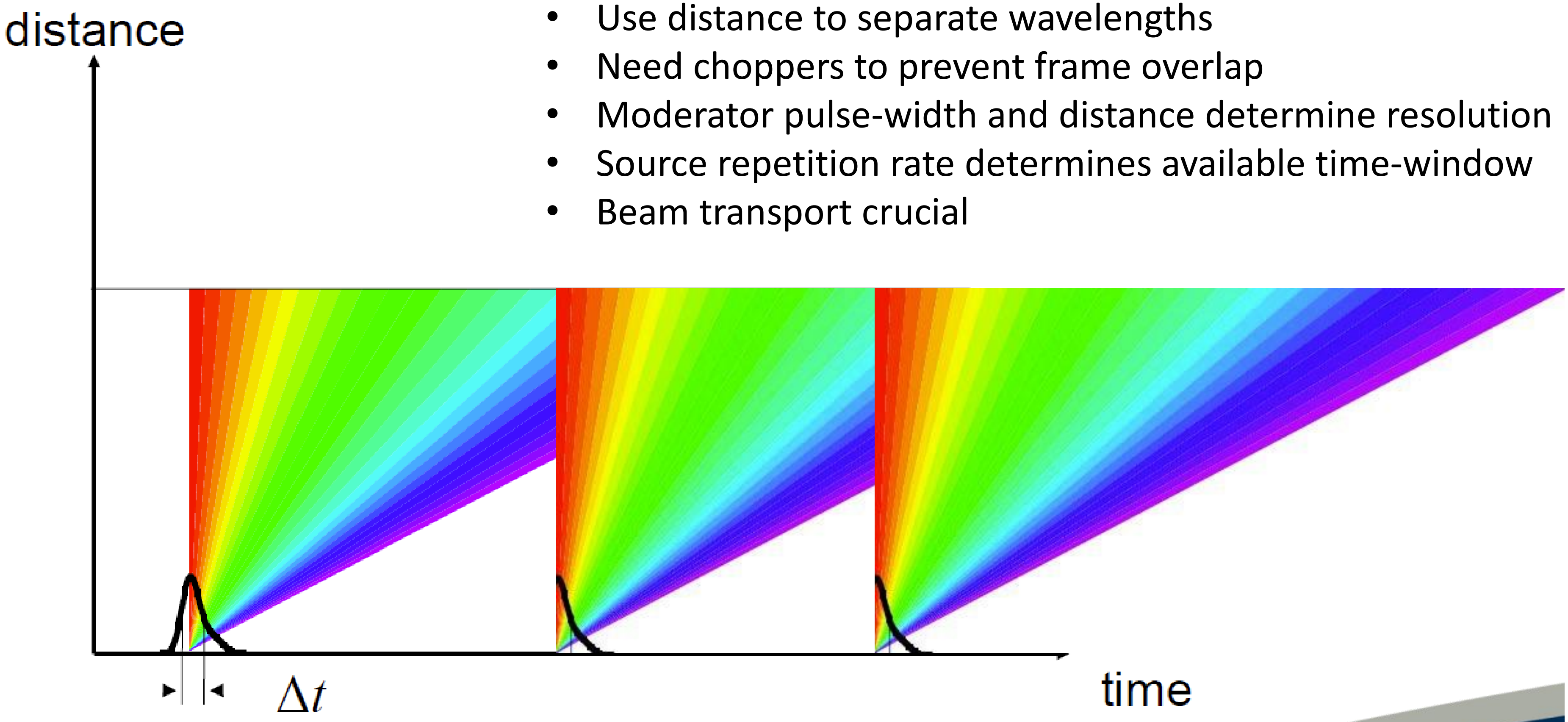


Resolution is almost constant in units of $\Delta\omega/\omega$~1.5%

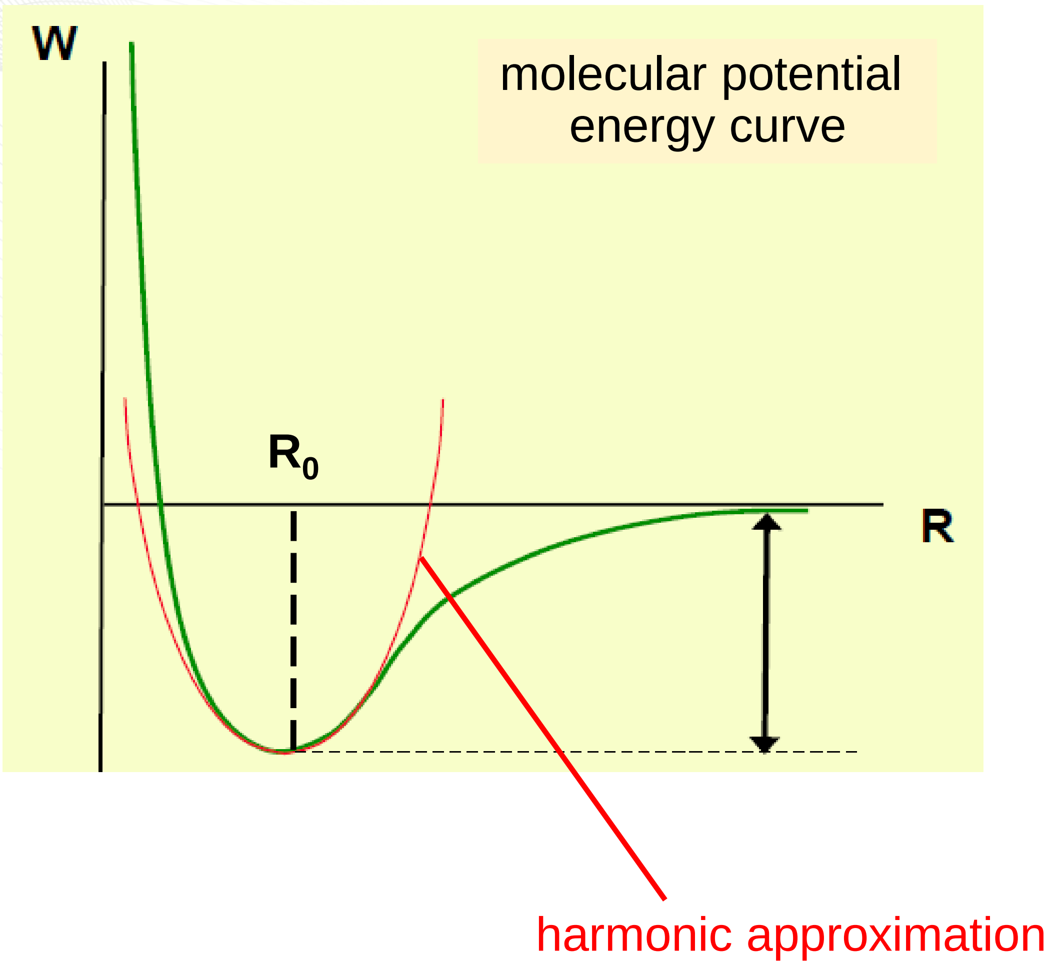


TOF method

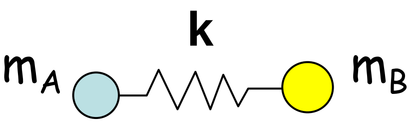
- Use distance to separate wavelengths
- Need choppers to prevent frame overlap
- Moderator pulse-width and distance determine resolution
- Source repetition rate determines available time-window
- Beam transport crucial



Vibrational spectroscopy



As a first approximation, a chemical bond between two atoms can be thought of as a spring connecting two masses:



Classical mechanics shows that this system vibrates with a characteristic frequency:

$$\nu = \left(\frac{1}{2\pi}\right)\sqrt{k/\mu}$$

where μ is the reduced mass:

$$\frac{1}{\mu} = \frac{1}{m_A} + \frac{1}{m_B}$$

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Vibrational spectroscopy

Dynamics at the atomic level is determined by quantum mechanics rather than by classical mechanics. The relevant problem here is the quantum harmonic oscillator.

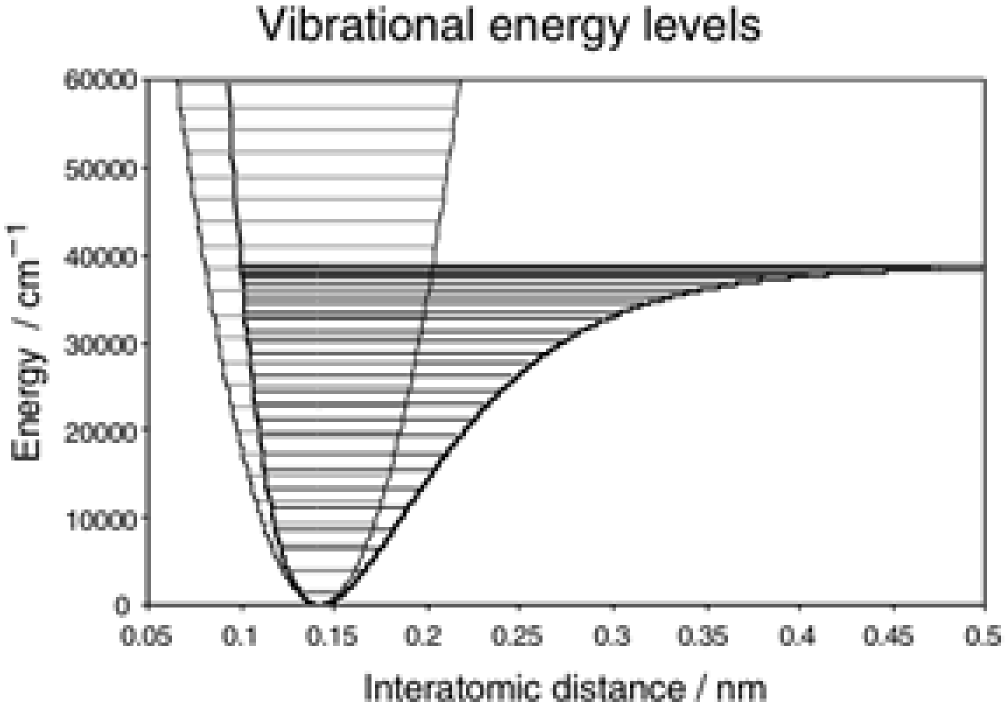
This is still an elementary problem of quantum mechanics. The energy levels of the oscillator are quantized and given by:

$$E_n = h\nu\left(n + \frac{1}{2}\right)$$

(n = 0, 1,2, 3, ...)

but the characteristic frequency, ν, is still given by the classical value:

$$\nu = \left(\frac{1}{2\pi}\right)\sqrt{k/\mu}$$



Vibrational spectroscopy

- A molecule with N atoms is a collection of N masses connected with harmonic springs.

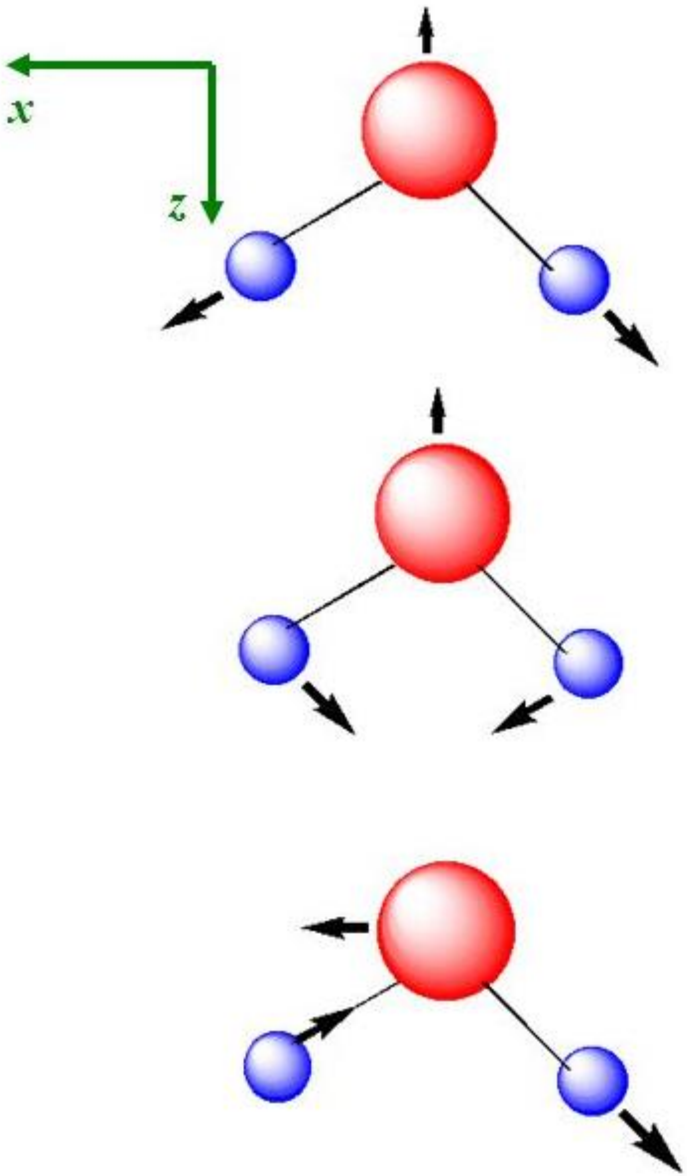
- Classical mechanics tells us that such a systems has 3N degrees of freedom.

- Three of these degrees of freedom correspond to translation of the molecule (position of its center of gravity in space), and three correspond to the orientation of the molecule in space (rotation about the center of gravity). This leaves

$$3.N - 3 - 3 = 3.N - 6$$

vibrational modes.

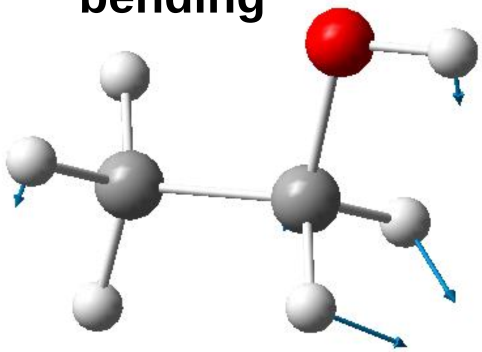
- For example H_2O (N=3) has 3.3 - 6 = 3 modes of vibration.



Vibrational spectroscopy

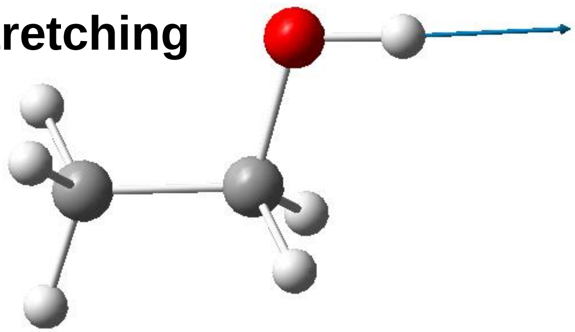
Several types of vibrational modes in molecules

bending



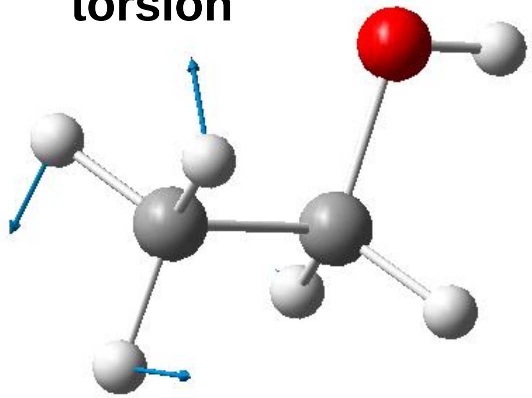
(bond angle changes)

stretching



(bond distance changes)

torsion



(rotation about bond axis; dihedral angle changes)

Two quantities define a vibrational mode:

- frequency
- set of atomic displacements

Notice that in a normal mode of vibration all atoms move in phase.

Vibrational spectroscopy

Molecular vibrations are useful to chemists because:

- they depend on molecular structure and interatomic or intermolecular forces (chemical bonding)

- specific bonds and functional groups are easily identified (analytical tool)

Table 1 Absorption frequencies of some common bonds (shown in bold type)

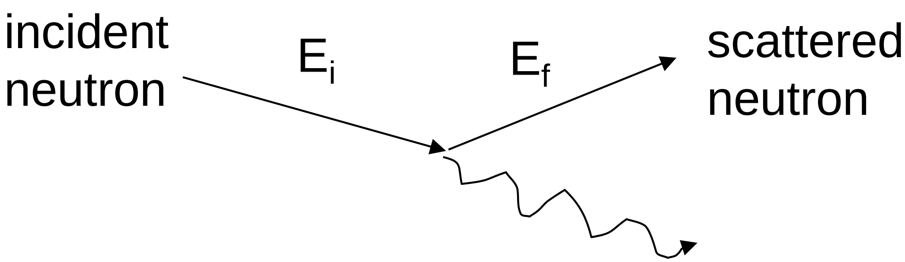
bond		*type of compound*	*frequency*
–C–H	(stretch)	alkanes	2800–3000
=C–H	(stretch)	alkenes, aromatics	3000–3100
≡C–H	(stretch)	alkynes	3300
–O–H	(stretch)	alcohols, phenols	3600–3650 (free) 3200–3500 (H-bonded) (broad)
–O–H	(stretch)	carboxylic acids	2500–3300
–N–H	(stretch)	amines	3300–3500 (doublet for NH_2)
–C(=O)–H	(stretch)	aldehydes	2720 and 2820
–C=C–	(stretch)	alkenes	1600–1680
–C=C–	(stretch)	aromatics	1500–1600
–C≡C–H	(stretch)	alkynes	2100–2270
–C(=O)–	(stretch)	aldehyde, ketones, carboxylic acids	1680–1740
–C≡N	(stretch)	nitriles	2220–2260
C–N	(stretch)	amines	1180–1360
–C–H	(bending)	alkanes	1375 (methyl)
–C–H	(bending)	alkanes	1460 (methyl and methylene)
–C–H	(bending)	alkanes	1370 and 1385 (isopropyl split)

Vibrational spectroscopy

How do we observe vibrational modes experimentally ?

<u>Crystallographers</u> use diffraction of some form of radiation (light, electron, x-ray, neutron,...) to obtain information on the periodic arrangement of atoms in space. The wavelength of the radiation is comparable to interatomic distances.

<u>Spectroscopists</u> use (inelastic) scattering of radiation (light, x-ray, neutron,...) to excite vibrational modes. The energy of the radiation is comparable to the energy associated with the vibrational excitations.



$$\hbar\omega = E_i - E_f$$

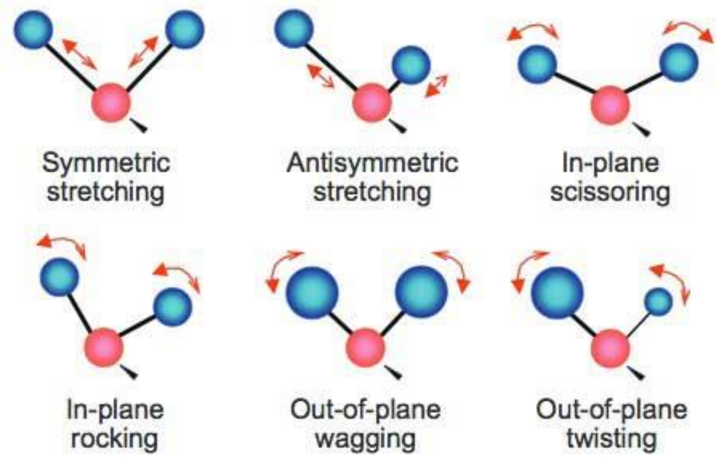
(conservation of energy)

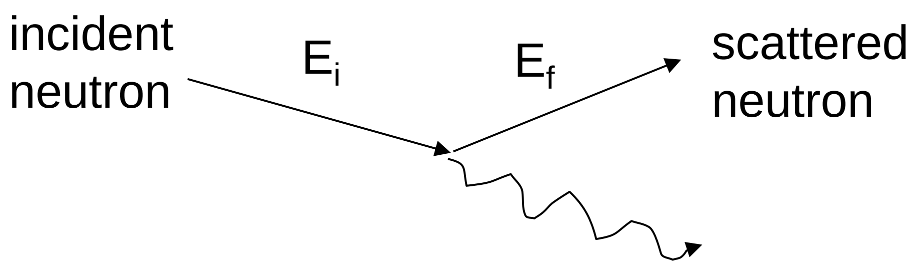
Upon interacting with a vibrational mode, the incident neutron loses energy (from E_i to E_f). The difference in kinetic energy is used to create a vibrational quantum.

Momentum is also exchanged !

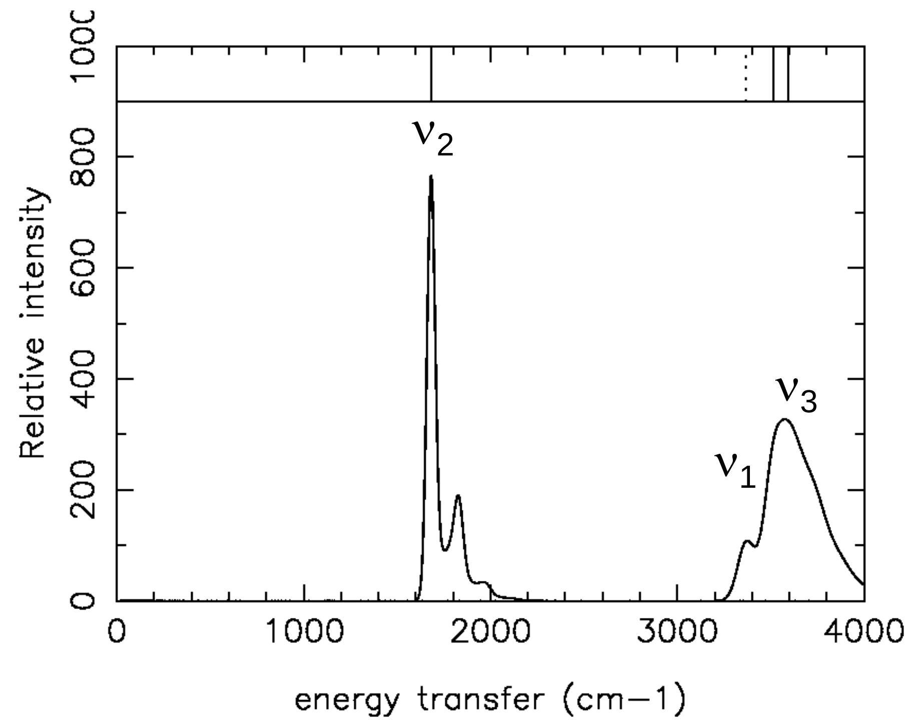
Vibrational spectroscopy

As long as we have a way to determine E_i and E_f and the number of particles with energy E_i and E_f, we can determine the number of excitations (vibrational modes) created with an energy of $\hbar\omega = E_i - E_f$. The result is the vibrational spectrum:





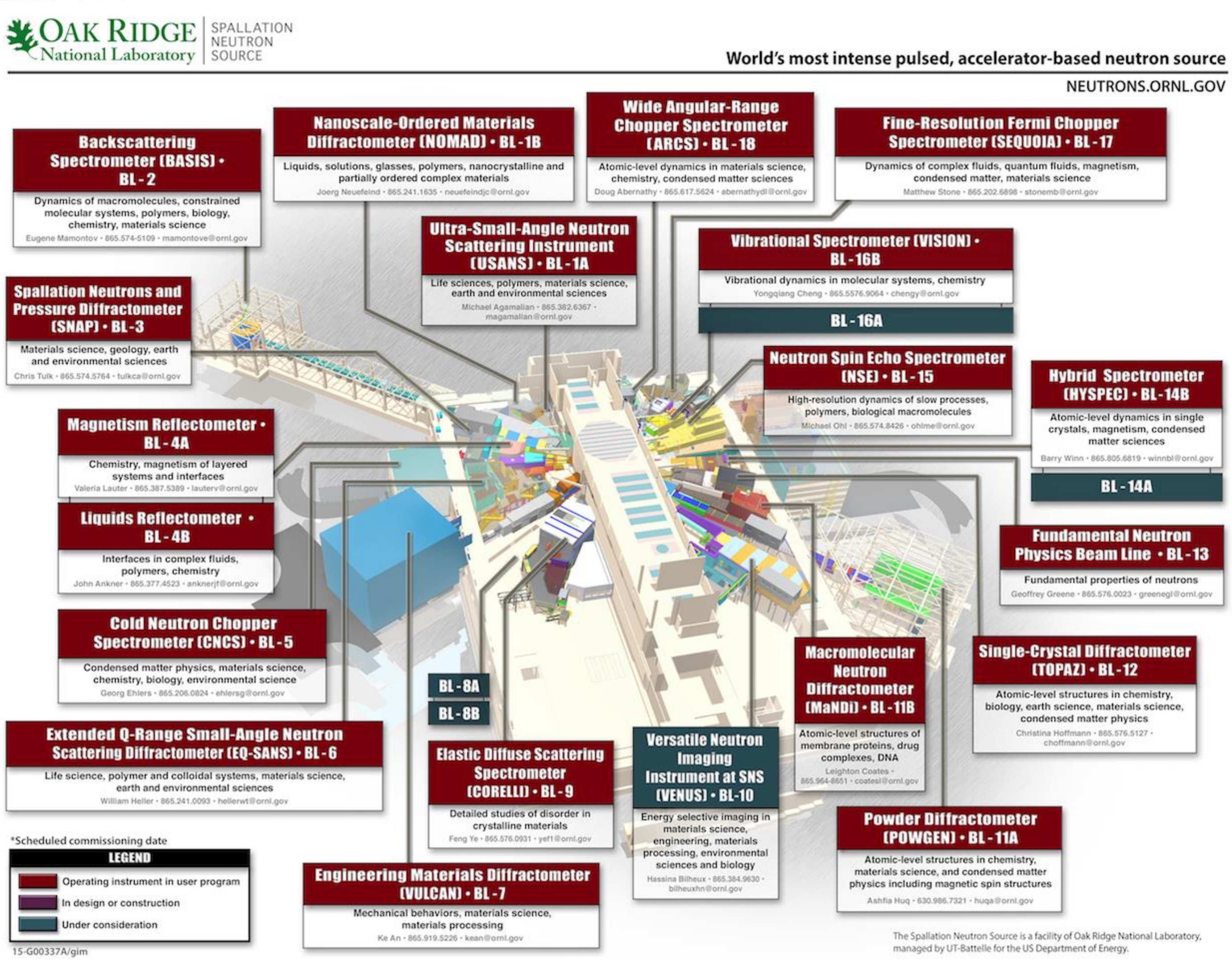
$\hbar\omega = E_i - E_f$



Vibrational spectroscopy

VISION (INS)	Raman/Infrared
Measures dynamics of nuclei (direct)	Measures response of electrons (indirect)
No selection rules	Selection rules apply
Great sensitivity to H	Cannot always see H
High penetration (bulk probe)	Low penetration (surface probe)
Easy access to low energy range (librational and translational modes)	Low energy cutoff applies (on the order of 100 cm^{-1})
Q trajectories in the (ω,Q) map; averaging over the Brillouin zone	Gamma point only
Weighted by neutron scattering cross section	Weighted by change in polarizability or dipole moment
Easy to simulate/calculate	Difficult to simulate/calculate
No energy deposition in sample	Heating, photochemistry, ...

SNS Instrument Suite



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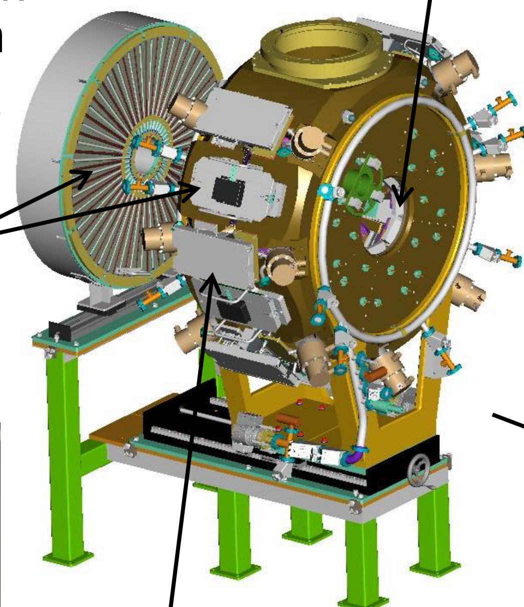
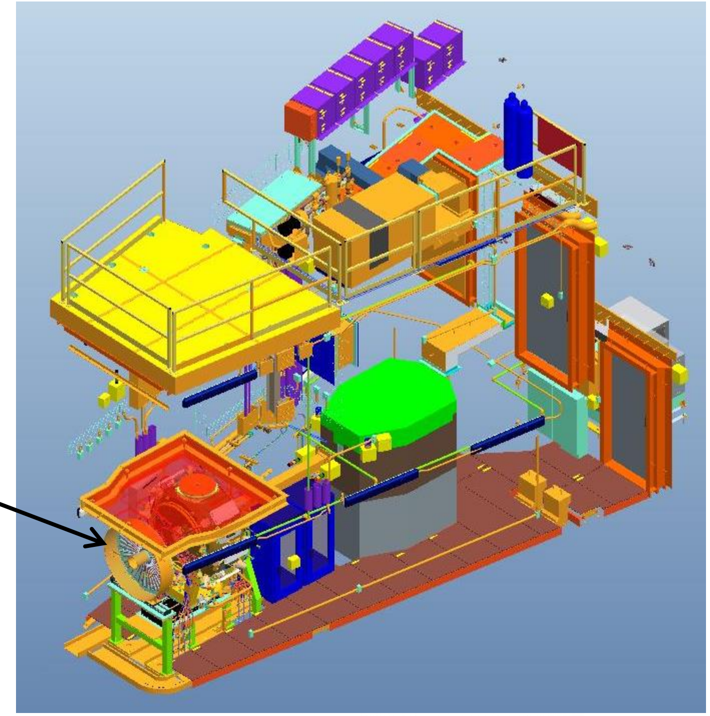
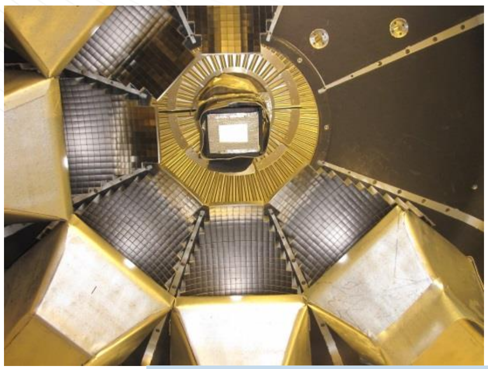
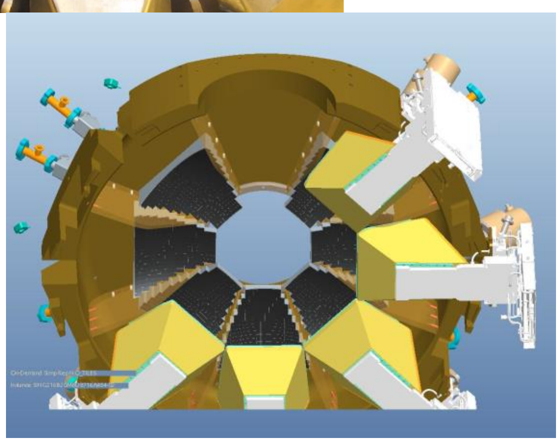
VISION



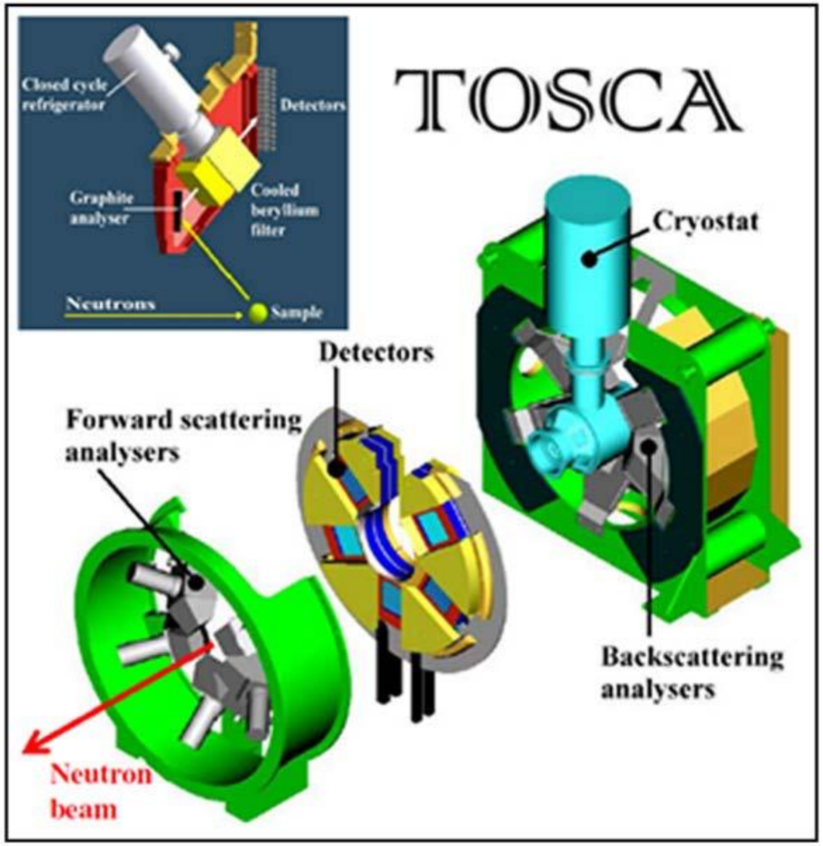






- Vibrational spectroscopy with neutrons
- Beam line started commissioning 3 years ago
- Multifunctional beam line: simultaneous spectroscopy and diffraction
- Dynamic range: 0-1000 meV; resolution: < 1.5%
- Diffraction: 1.5 - 30 A^{-1}
- Temperature range: 5-700K
- Sample environment: high pressure, electric field, gas loading, ...
- Great sensitivity to hydrogen, no selection rules, penetration through matter, ...

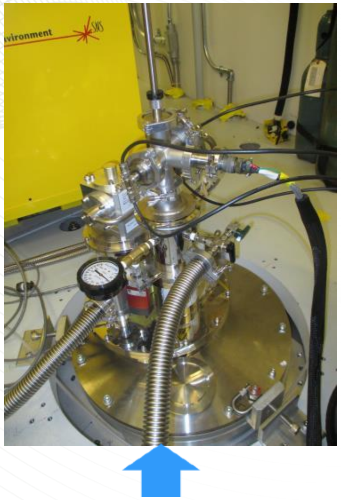
TOSCA



Closed cycle refrigerator
Detectors
Graphite analyser
Cooled beryllium filter
Neutrons
Sample
TOSCA
Cryostat
Detectors
Forward scattering analysers
Backscattering analysers
Neutron beam

25/10/2005

Sample environment

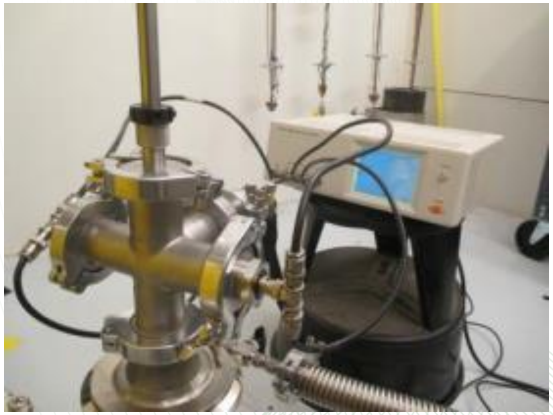


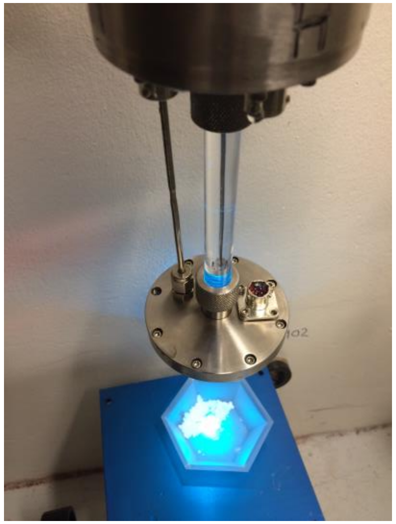
JANIS closed-cycle refrigerator (5-600K)

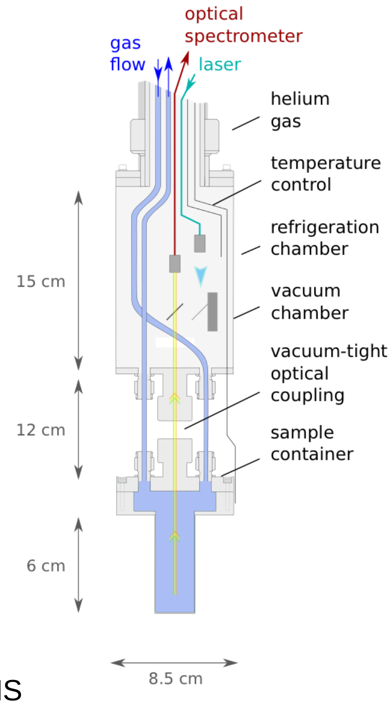
ortho/para H_2 converter

Largest single crystal diamond for DAC!

in situ electrochemical impedance spectroscopy (EIS)

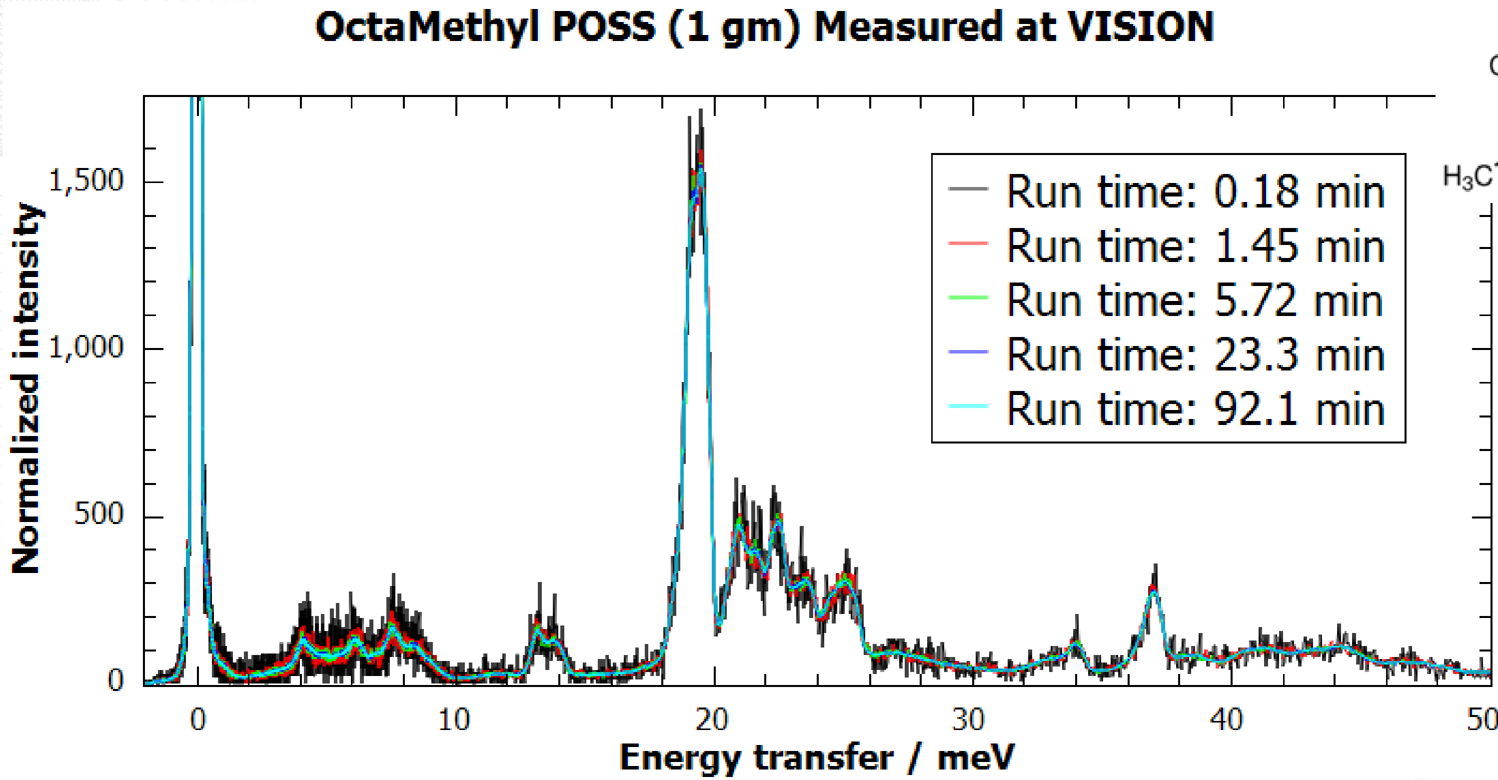


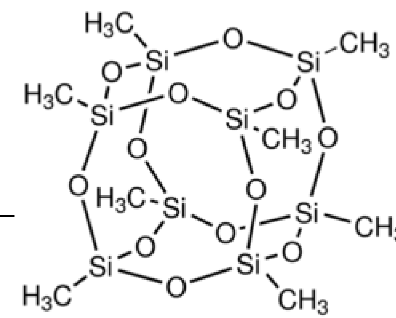


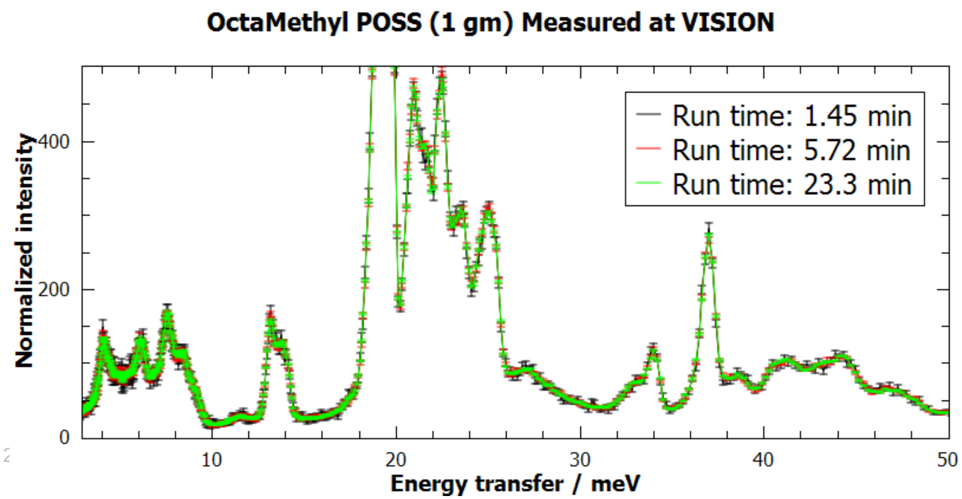


in situ simultaneous Raman and INS

High throughput: INS in minutes





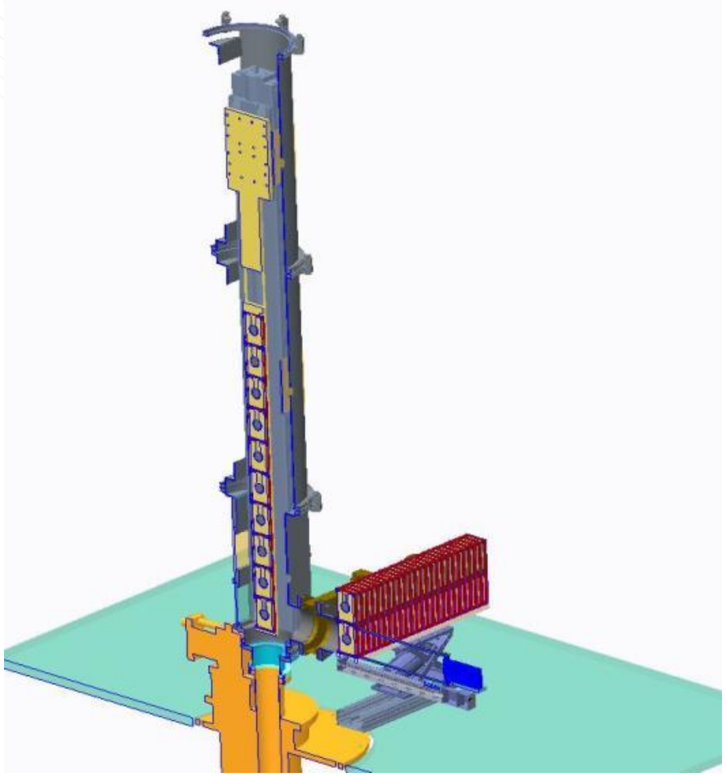


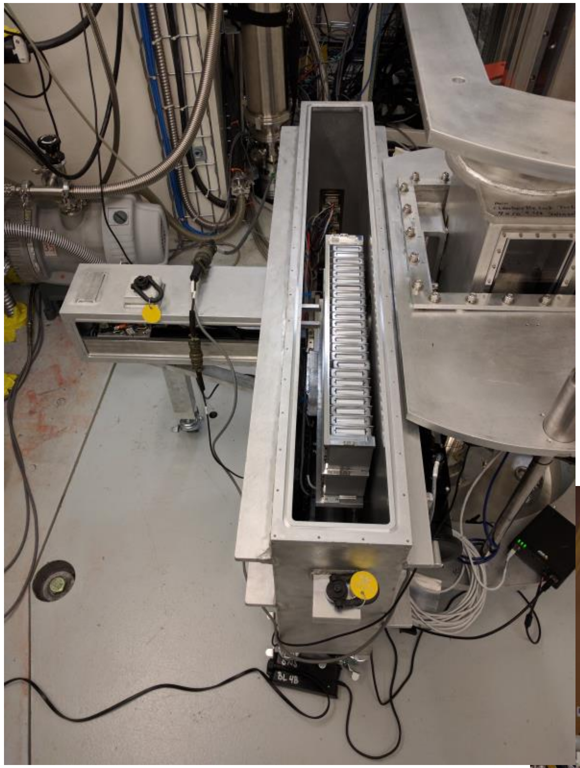
With 14 inelastic banks and 16 diffraction banks, VISION has the highest data rate (up to millions of events per second) among all neutron beamlines in the world.

- INS database
- Parametric studies
- INS study of kinetics

VISION Sample changer

The high throughput rate of VISION requires very rapid sample changes to make the best use of neutron beamtime and run mail-in program. A sample changer has been tested in January 2017 and will be commissioned this cycle.





High Throughput

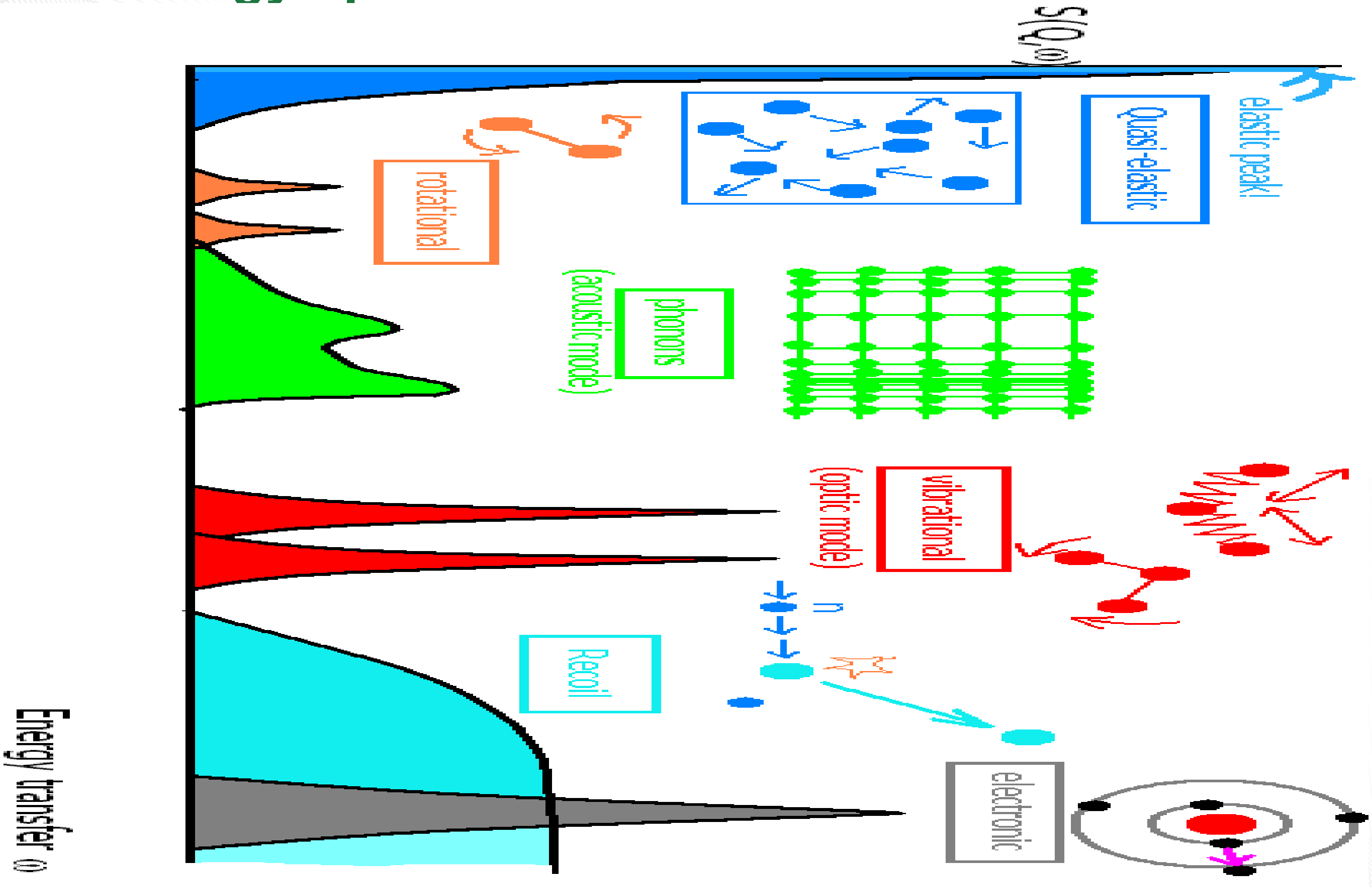
Challenges

- Large volume of Data
- How to handle large number of samples
  - Sample changers
  - Sample environment
  - Gas handling
- How to model and interpret the results

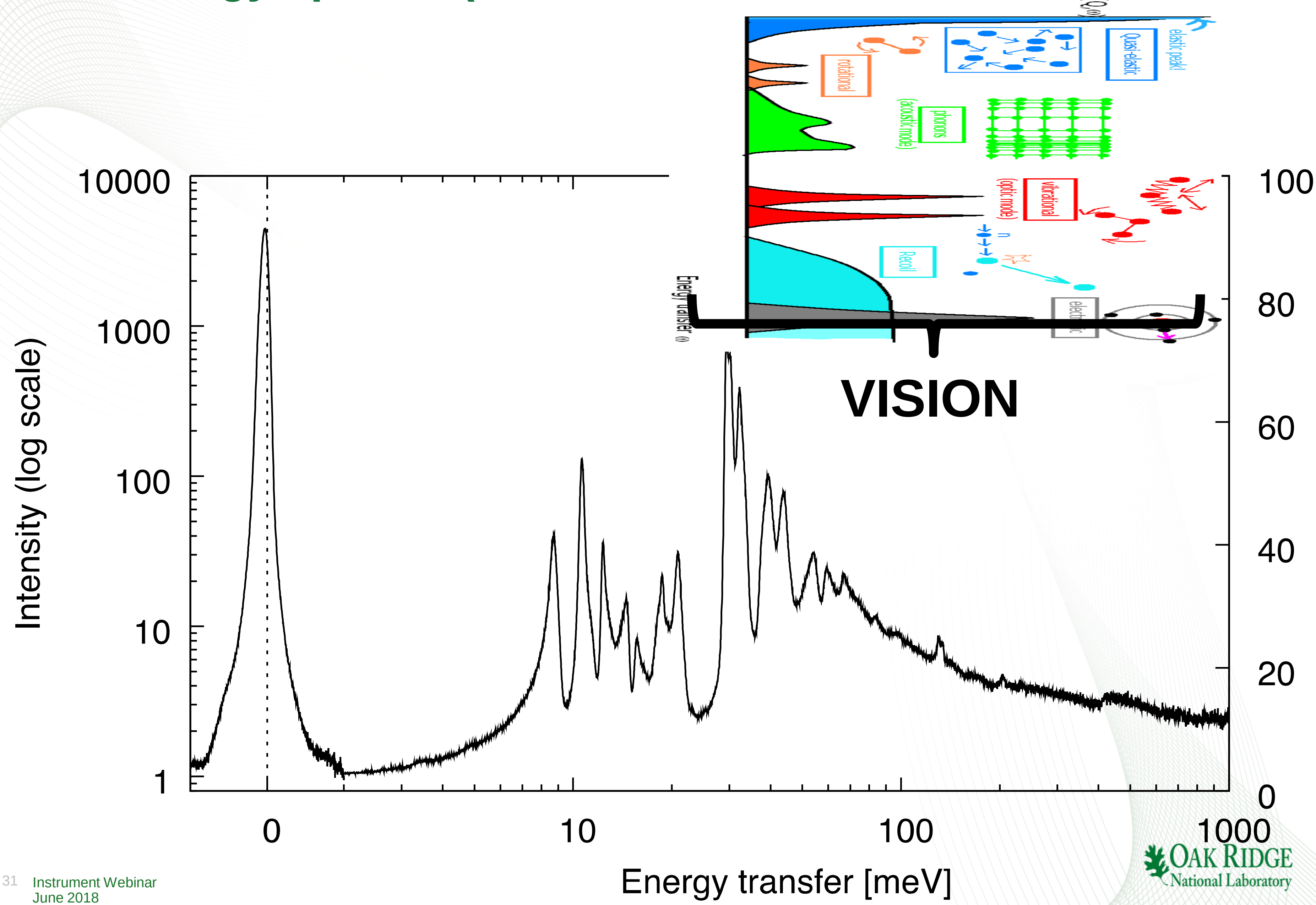
Opportunities

- Databases and Libraries
- Parametric studies
- Kinetic studies
- In-situ studies
- Small signals in large backgrounds
- Modeling

The energy spectra

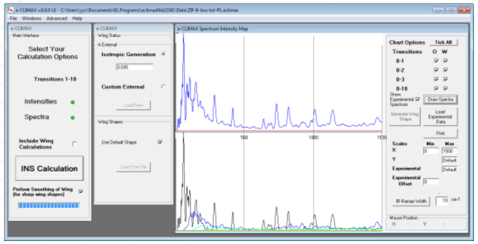


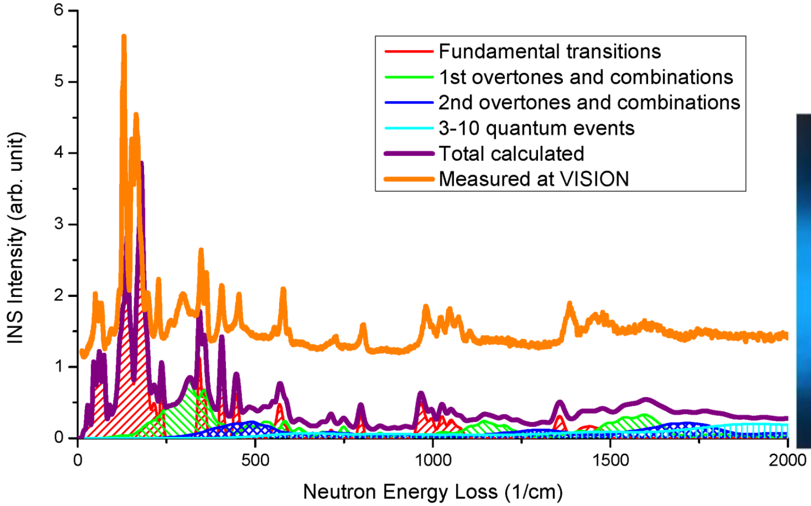
The energy spectra (according to VISION)



Integrated modeling for data interpretation

Today this is what we do at **VISION** using **VirtuES** (Virtual Experiments in Spectroscopy)
1600 cores cluster dedicated to VISION





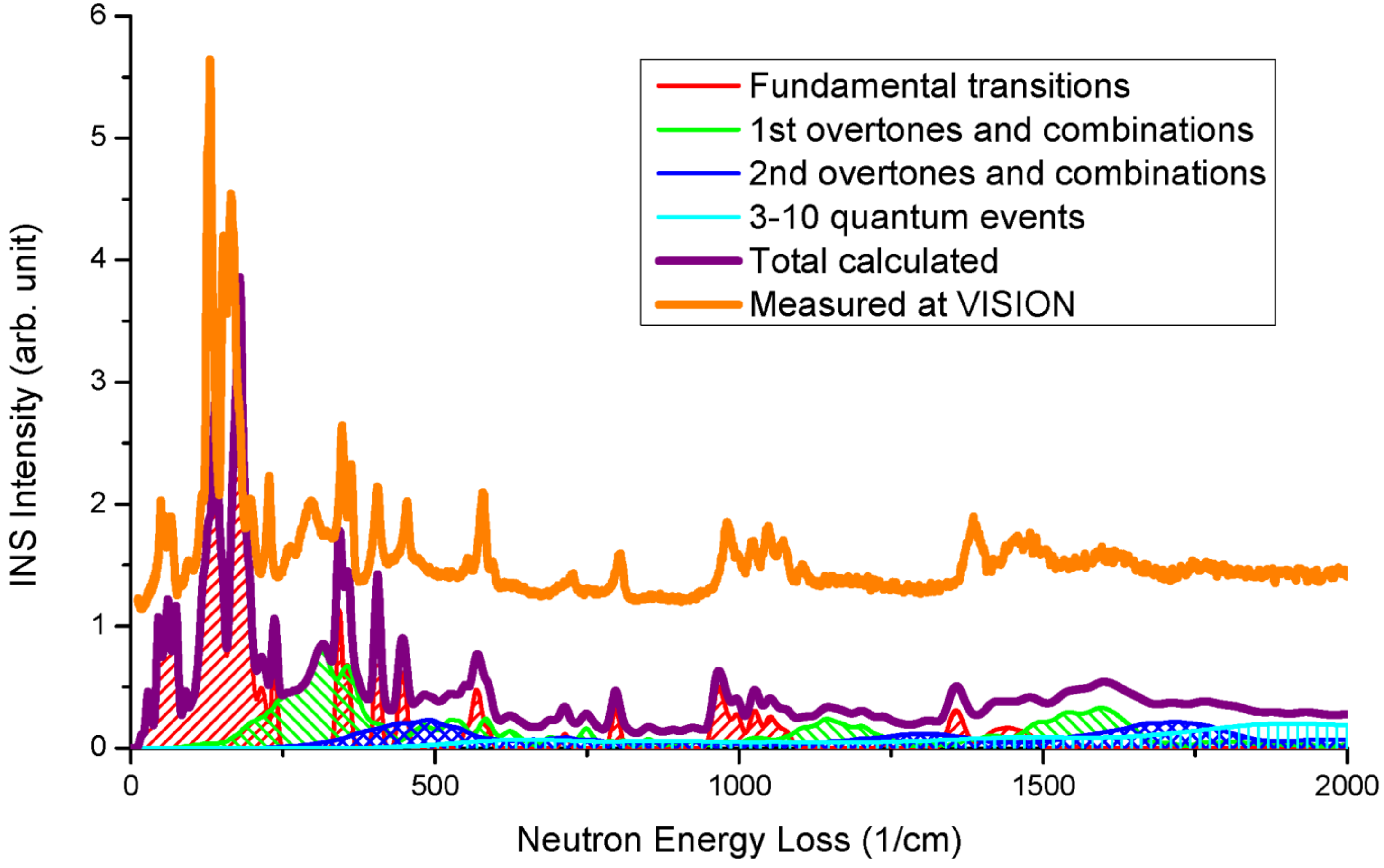
CASTEP
VASP
Quantum Espresso
Gaussian
→ Vibrational modes and frequencies → INS simulation (OClimax) → Simulated INS spectra

Sample → VISION → Data reduction and analysis (Mantid) → Measured INS spectra; → Measured diffraction

Simulated INS spectra, Measured INS spectra, Measured diffraction → Peak assignment

Peak assignment:
- Structure-dynamics correlation → Understanding mechanisms and properties at atomic level
- (An)harmonicity → Mapping out the local potential energy profile using finite displacement, frozen phonon, molecular dynamics
- Phase transition → Revealing the kinetics and the transition pathway

Comparison



sample is hexamethybenzene

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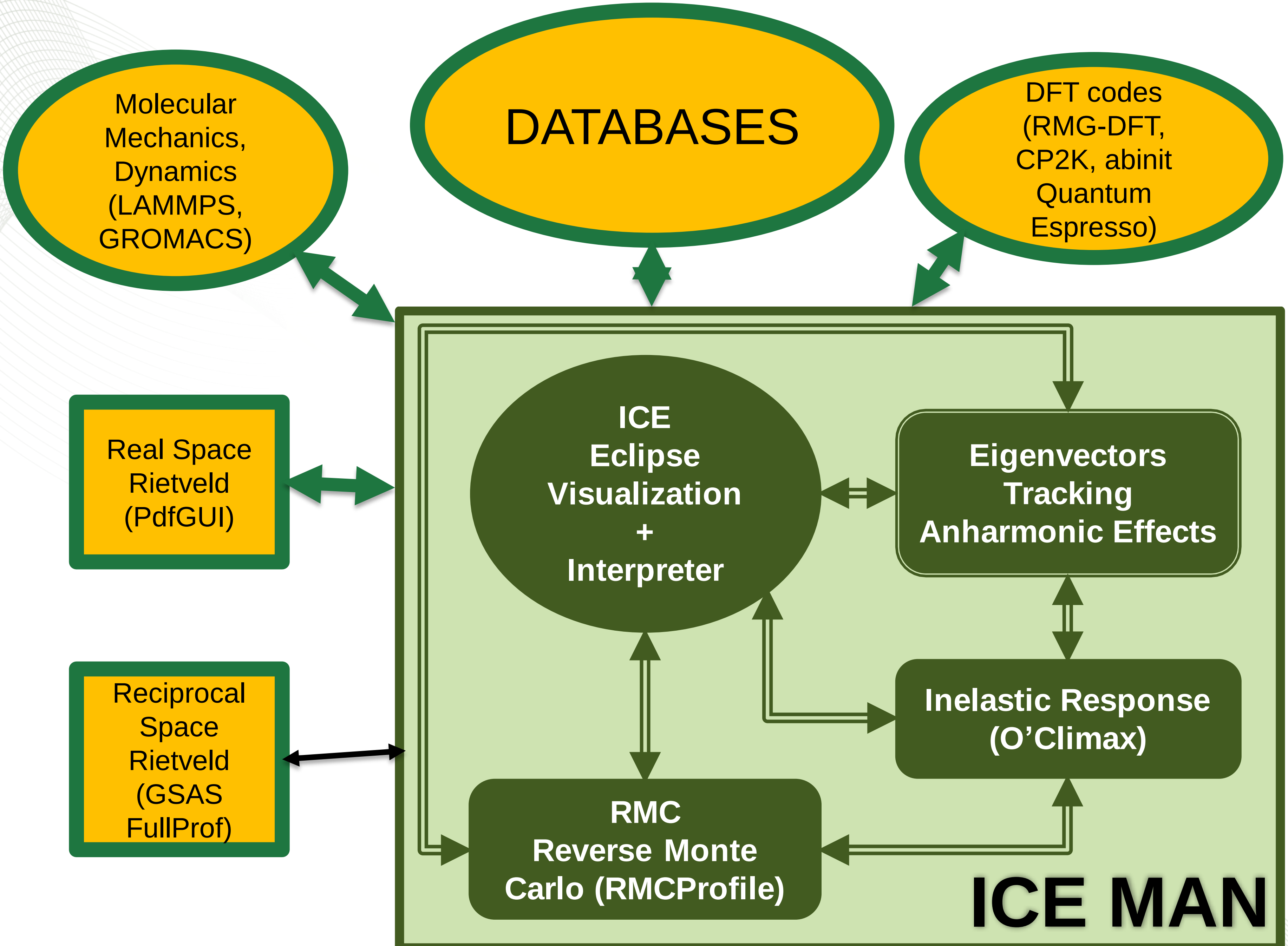
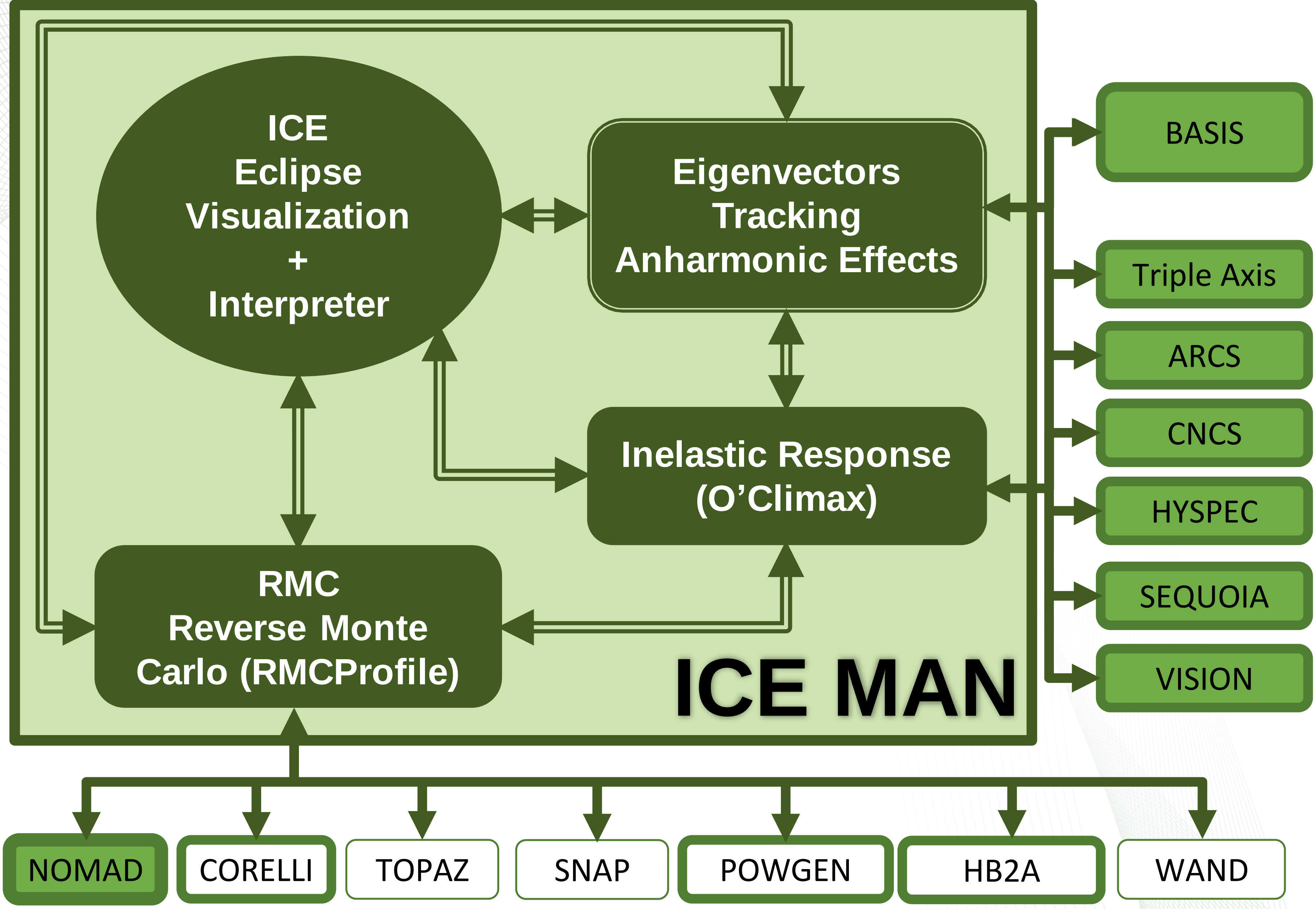

Molecular Mechanics, Dynamics (LAMMPS, GROMACS)
DATABASES
DFT codes (RMG-DFT, CP2K, abinit Quantum Espresso)
Real Space Rietveld (PdfGUI)
Reciprocal Space Rietveld (GSAS FullProf)
ICE Eclipse Visualization + Interpreter
Eigenvectors Tracking Anharmonic Effects
Inelastic Response (O'Climax)
RMC Reverse Monte Carlo (RMCProfile)
ICE MAN


ICE
Eclipse
Visualization
+
Interpreter
Eigenvectors
Tracking
Anharmonic Effects
Inelastic Response
(O'Climax)
RMC
Reverse Monte
Carlo (RMCProfile)
ICE MAN
BASIS
Triple Axis
ARCS
CNCS
HYSPEC
SEQUOIA
VISION
NOMAD
CORELLI
TOPAZ
SNAP
POWGEN
HB2A
WAND

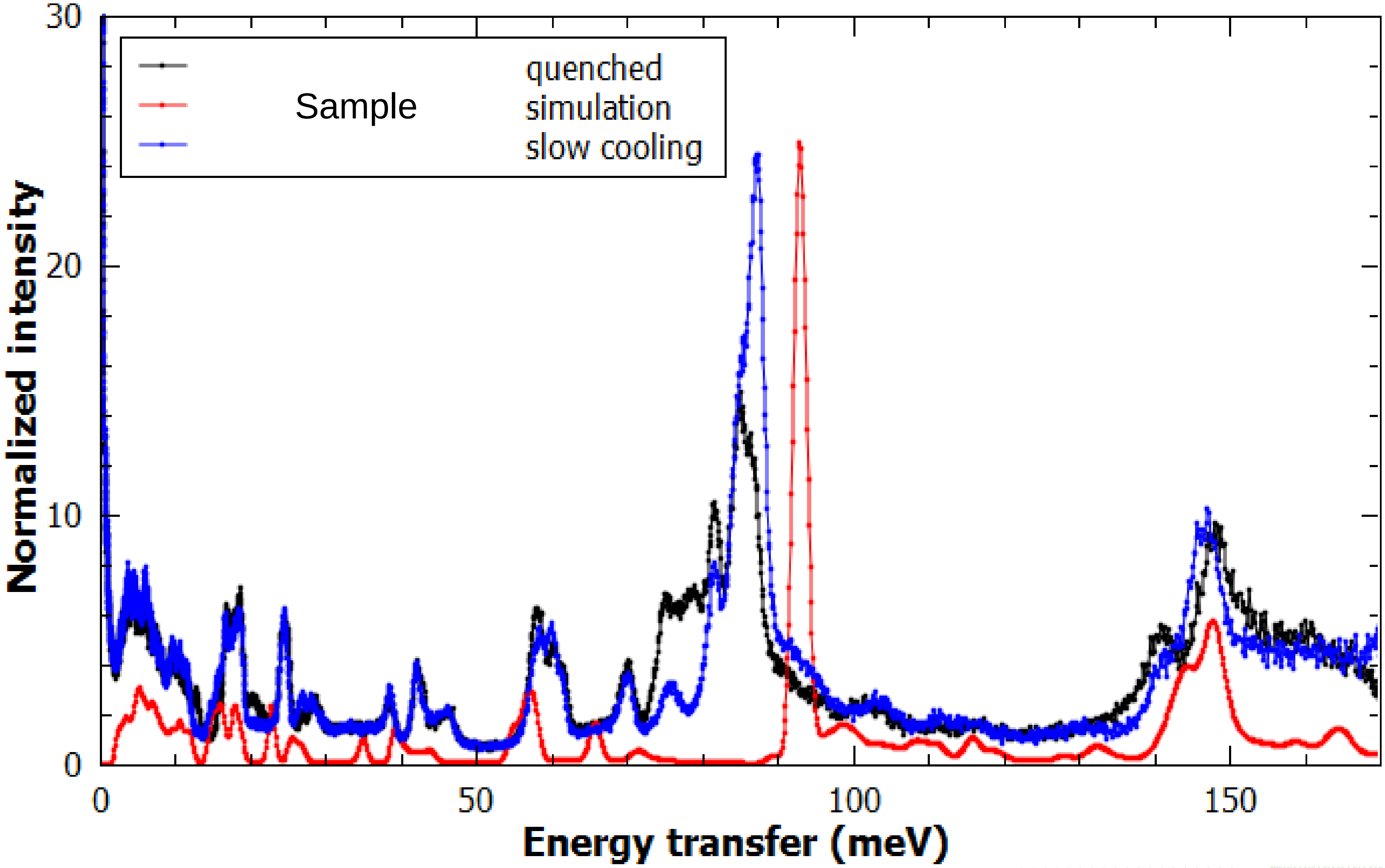
VirtuES helped users to make decisions on-the-fly

```
[yyc@or-condo-login02 CF3SO2OH]$ ls -lhtr
-rw-r--r-- 1 yyc users 3.6K Nov  4 15:50 cell
-rw-r--r-- 1 yyc users 1.1K Nov  4 15:50 param
-rw-r--r-- 1 yyc users 3.9K Nov  4 15:51 _PhonDOS.cell
-rw-r--r-- 1 yyc users  735 Nov  4 15:52 _PhonDOS.param
-rw-r----- 1 yyc users 1.1M Nov  4 16:46 castep
-rw-r----- 1 yyc users 7.3M Nov  5 06:15 _PhonDOS.phonon
-rw-r----- 1 yyc users 232K Nov  5 06:15 _PhonDOS.castep
-rw-r--r-- 1 yyc users 3.3M Nov  5 08:56 aclimax

[yyc@analysis-node02 manualreduce]$ ls -lhtr
-rw-rwx---+ 1 yyc users 2.2M Nov  5 12:34 VIS_20557_5K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 13:28 VIS_20559_50K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 14:23 VIS_20561_75K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 15:56 VIS_20563_100K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 17:21 VIS_20565_125K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 18:44 VIS_20567_150K_for_0.9hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 20:23 VIS_20570_175K_for_1.2hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 21:58 VIS_20572_200K_for_1.2hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  5 23:29 VIS_20574_225K_for_1.2hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  6 01:00 VIS_20576_250K_for_1.2hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  6 02:28 VIS_20578_275K_for_1.2hr.nxs
-rw-rwx---+ 1 yyc users 2.2M Nov  6 03:57 VIS_20580_300K_for_1.2hr.nxs
```

Simulation was started at the beginning of the experiment. By the time when experimental data were collected, the calculation was already finished with theoretical predication available to be compared with experiment. This eventually led to a critical decision made by the user (see next slide).

VirtuES helped users to make decisions on-the-fly



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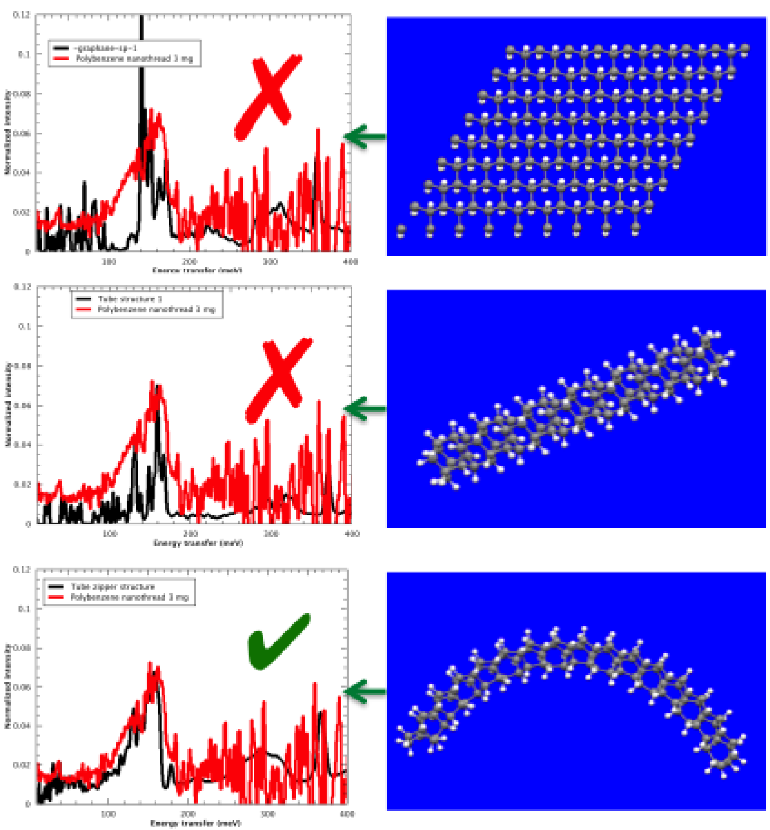
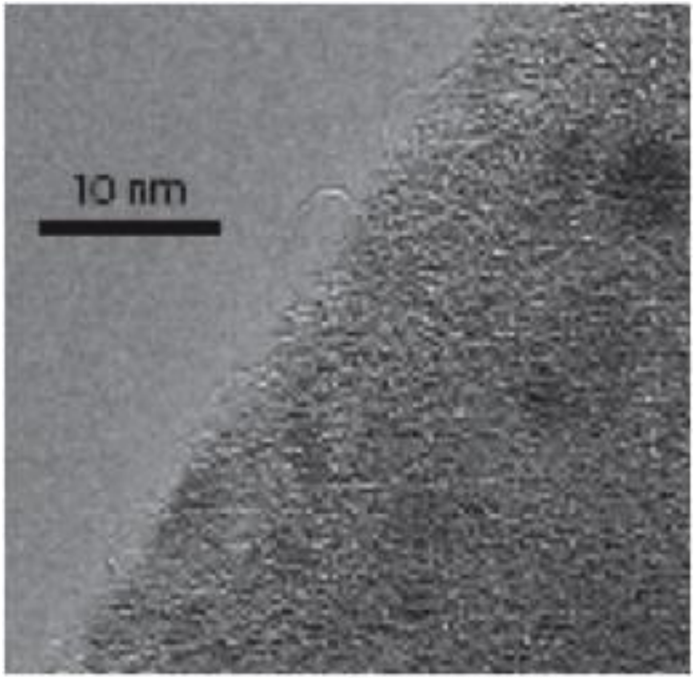
VISION: Unprecedented capabilities and opportunities

3 mg of nanothread sample



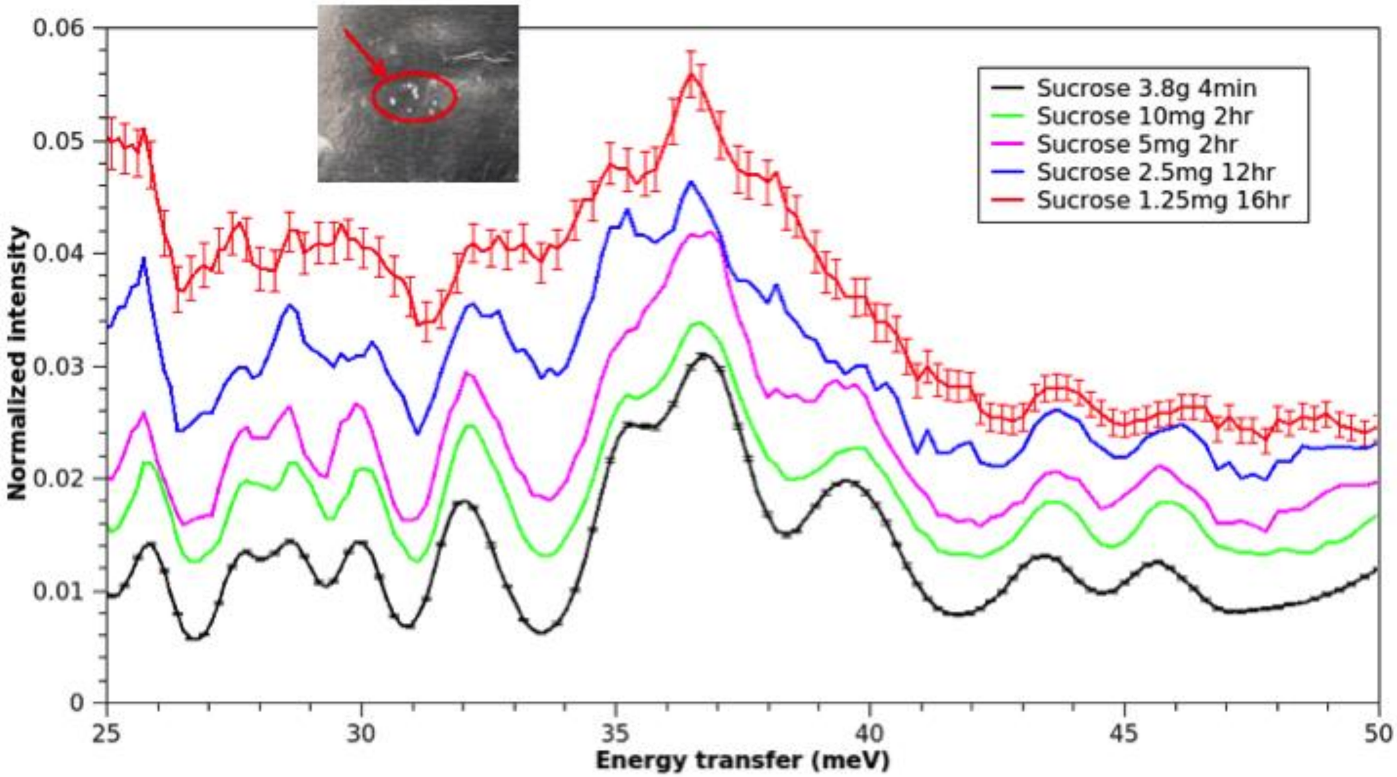
Comparison of the experimental data from VISION and a series of DFT calculations of hypothetical structures that contain sp^3 carbon and the correct stoichiometry (C:H ratio 1:1) allows us to determine which structure corresponds to the measured spectra.



Collaboration with Malcolm Guthrie, John Badding, Vin Crespi. Original publication on carbon nanothreads: Nature Materials, **14**, 43 (2014)

High sensitivity: milligrams of samples

1.25 mg of table sugar



Extraordinary sensitivity, this is the smallest amount of sample ever measured using INS. Diamond anvil cells will be used in VISION

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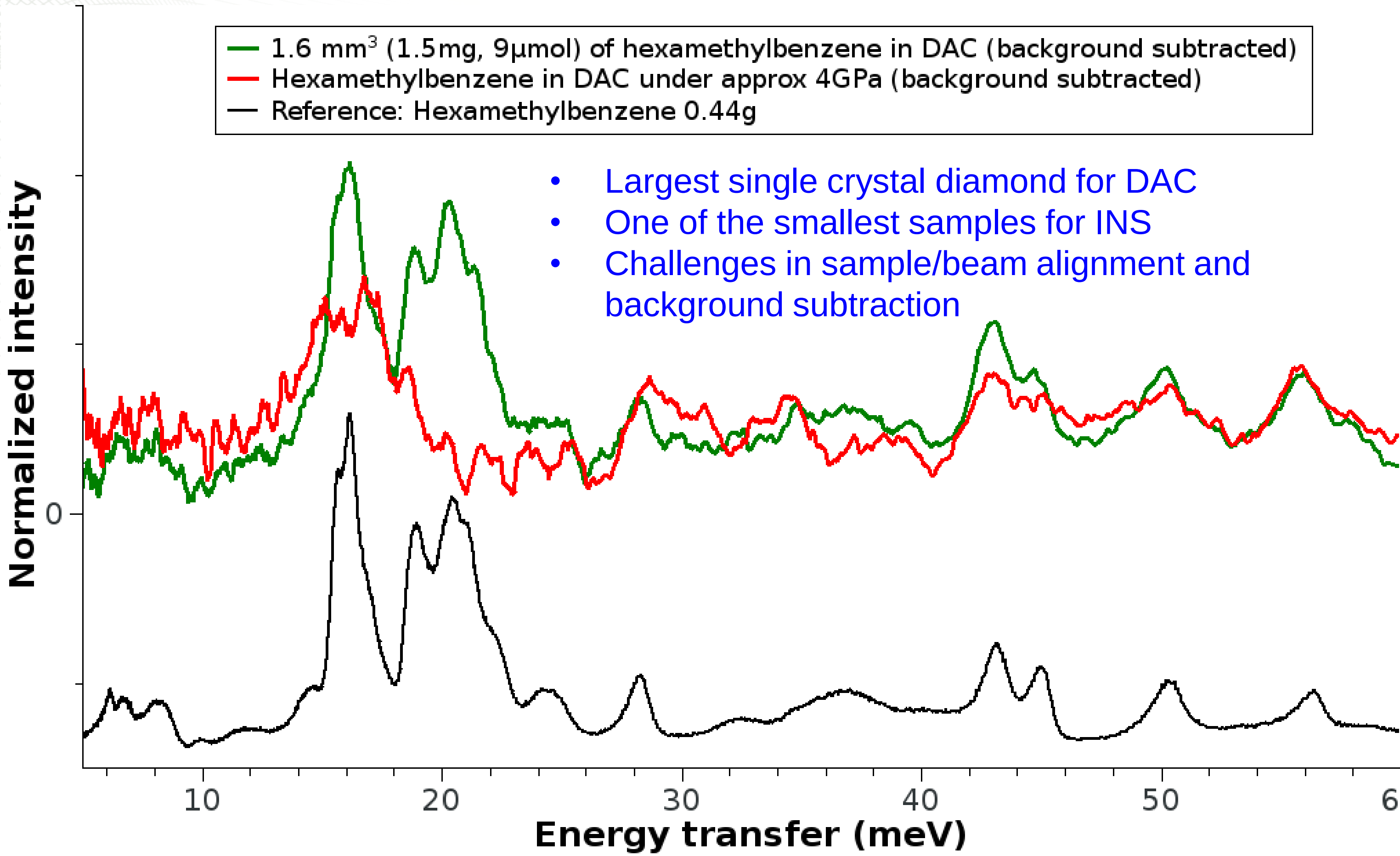
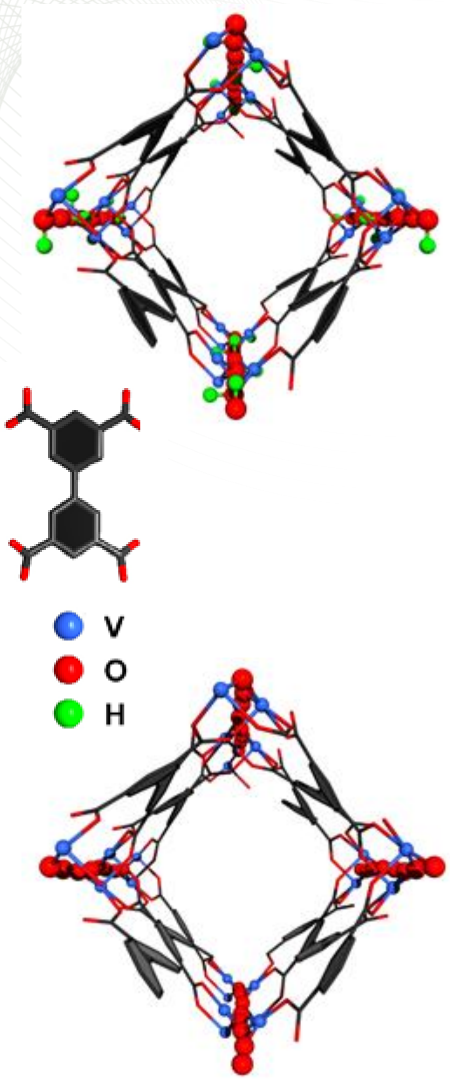
A successful proof-of-principle test at VISION: using diamond anvil cell (DAC) for high pressure INS experiments

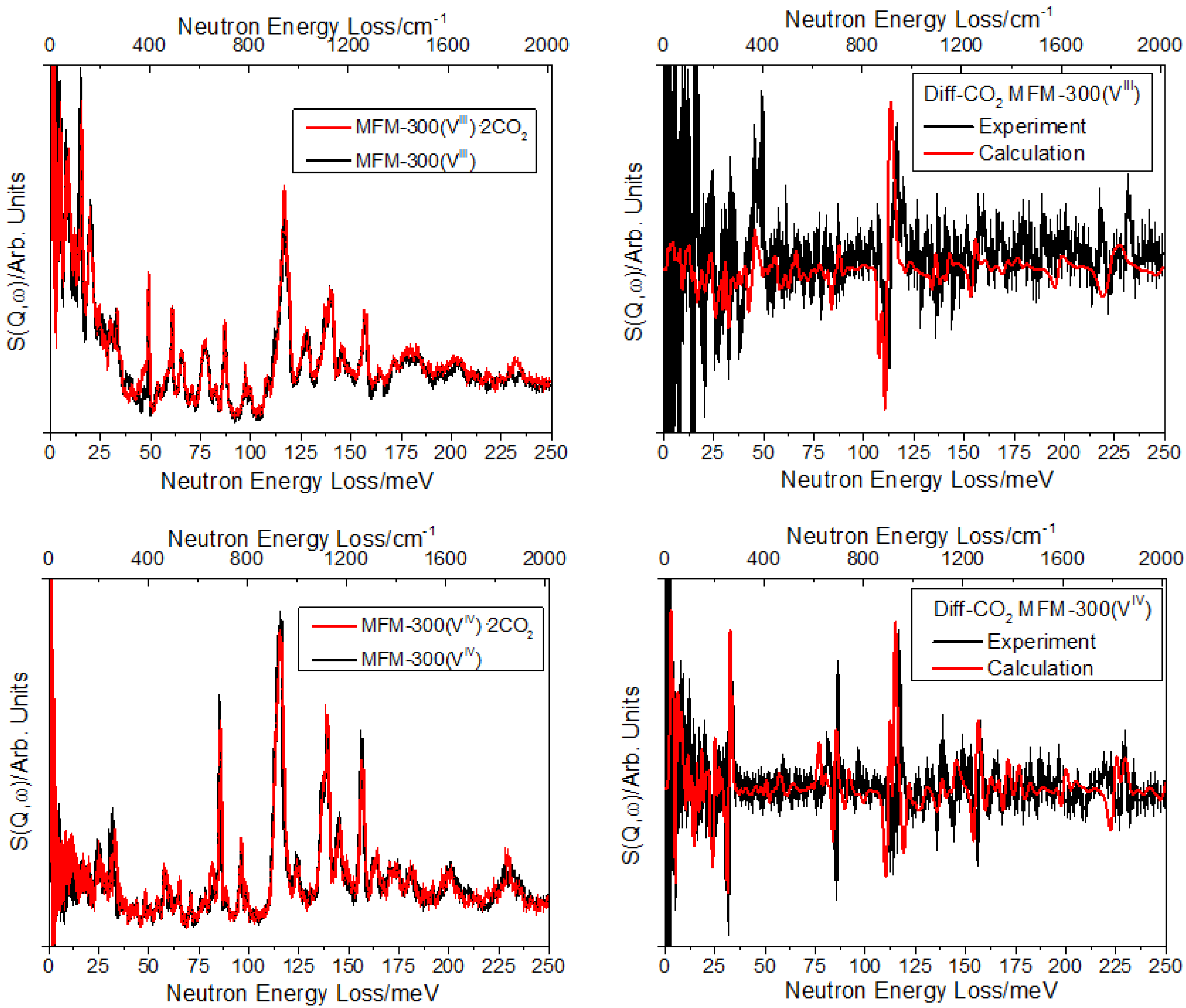


- Largest single crystal diamond for DAC
- One of the smallest samples for INS
- Challenges in sample/beam alignment and background subtraction

- INS spectrum from 1.6mm^3 (1.5mg, 9μmol) sample loaded in the DAC was successfully extracted, with significant details retained.
- Approx. 4GPa pressure was applied, leading to major changes in the spectrum.
- The unprecedented capability will open the door to many new areas using INS to study materials dynamical behavior under high pressure.

NOTT-V MOF and CO2 adsorption



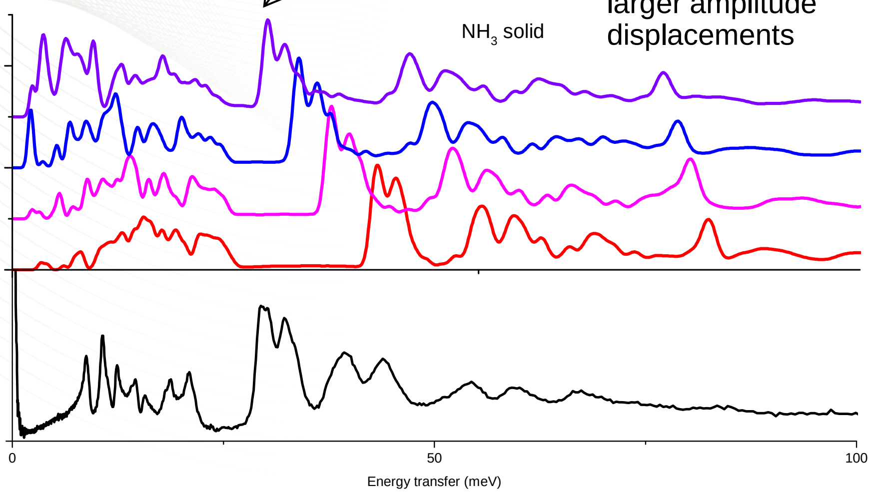


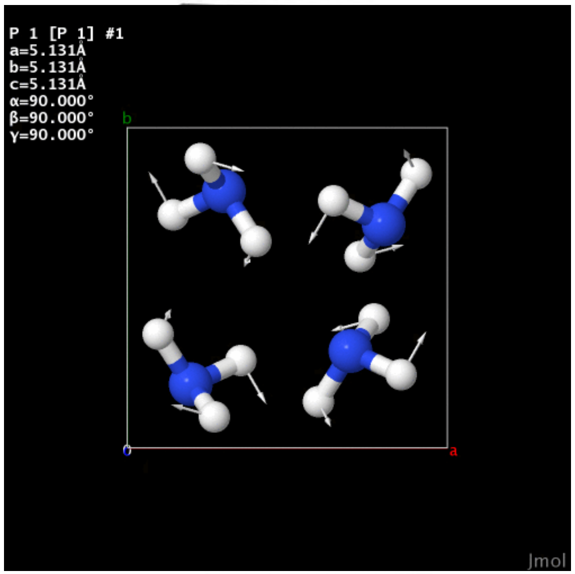
Collaboration with Sihai Yang and Martin Schroder at University of Manchester.

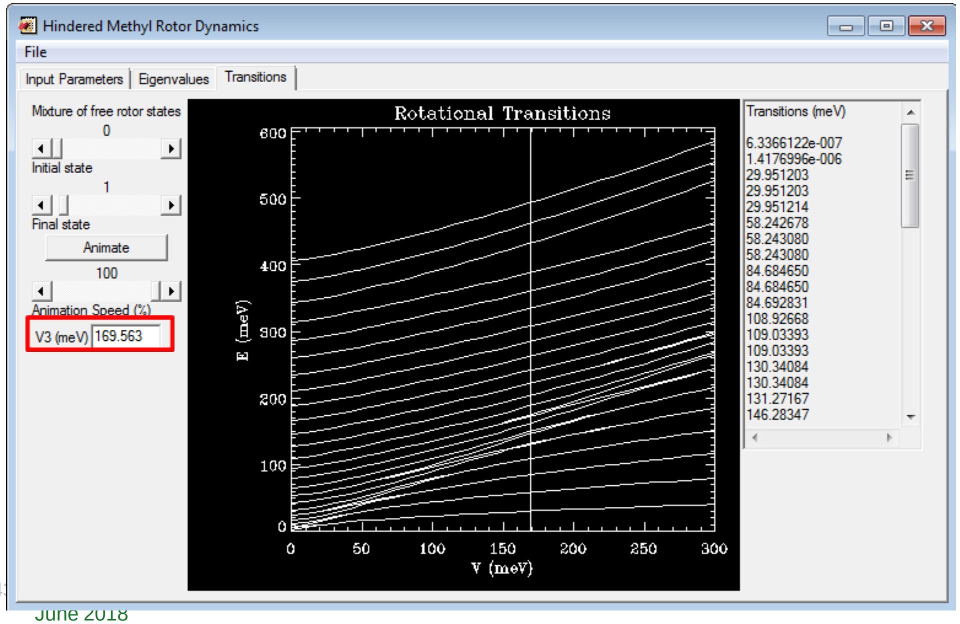
Hydrogen in a “simple” molecular solid: Beyond DFT and harmonic approximation

Libration/rotation of NH3 group

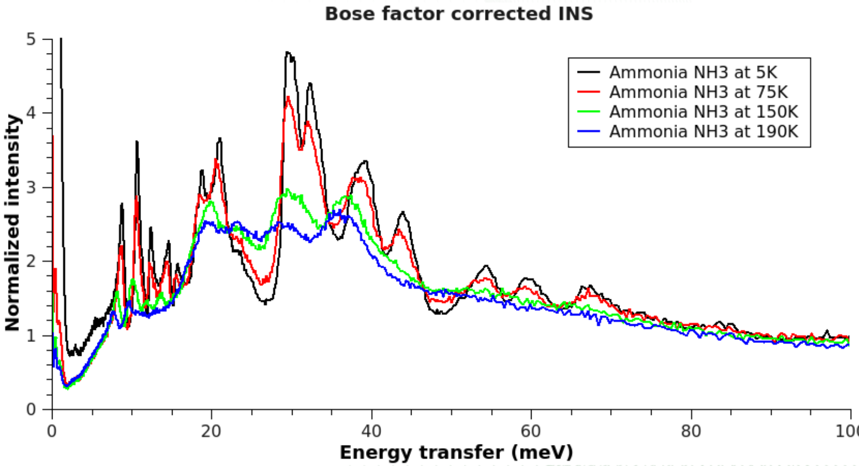
Calculated INS using larger amplitude displacements



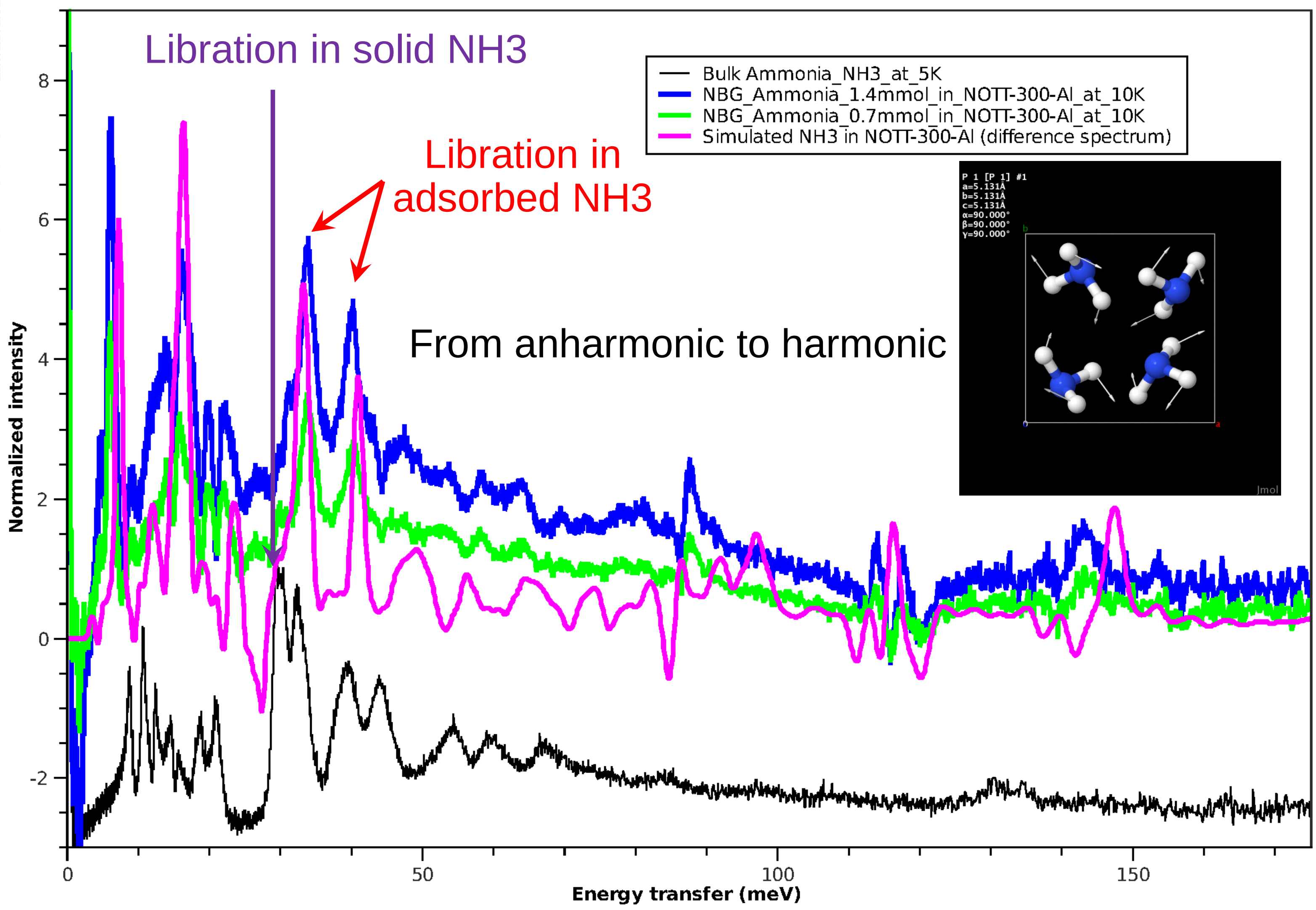




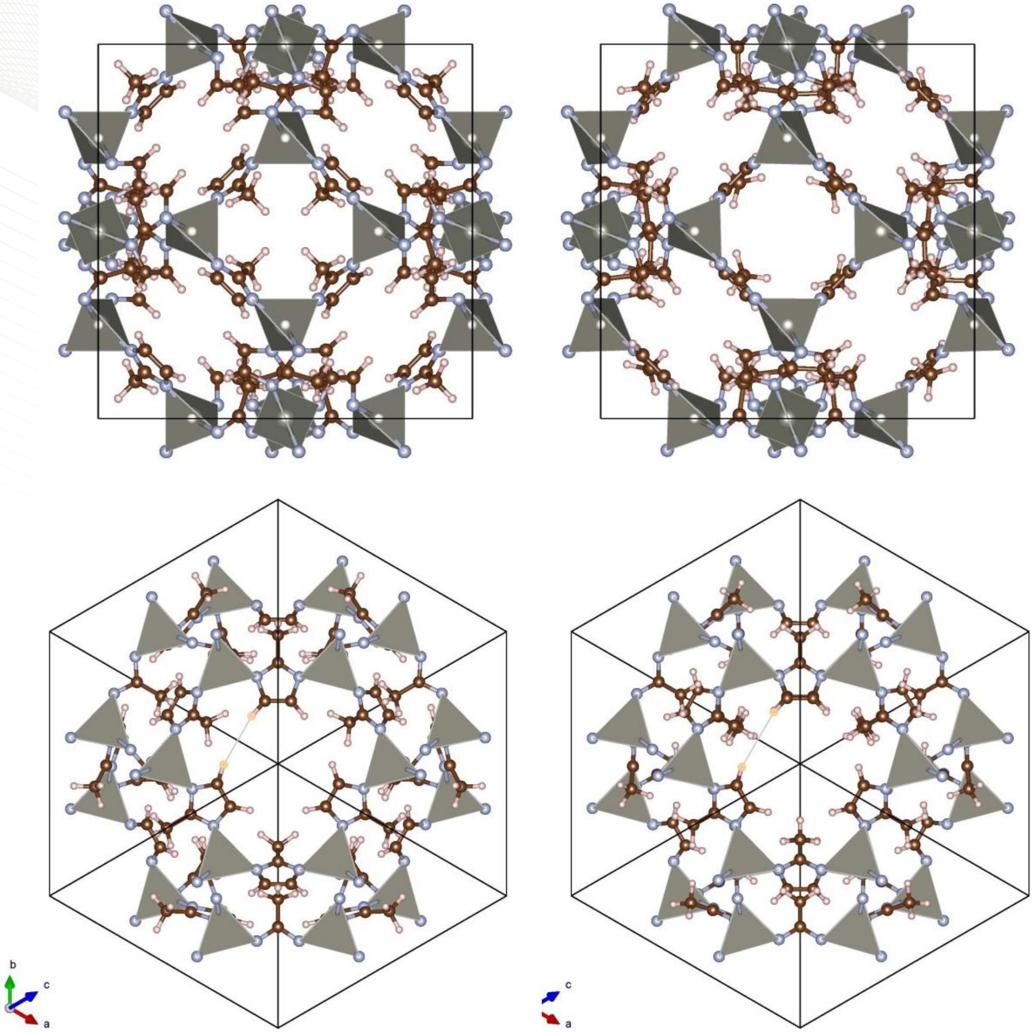
- DFT calculated energy barrier for rigid rotation of NH3: 180 meV
- Energy barrier solved from the rotor model : 170 meV



Solid NH3 vs NH3 in MOF



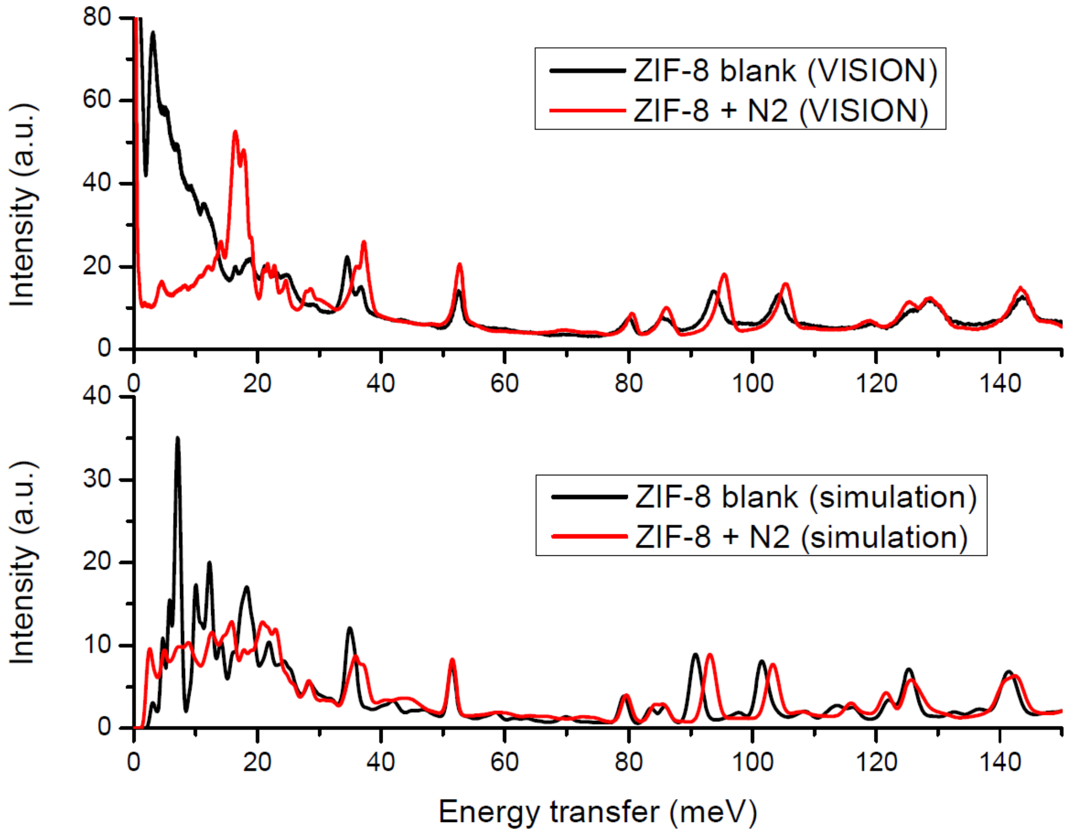
Gate-opening in a metal-organic framework



Structure of blank ZIF-8 and ZIF-8 loaded with N_2. The rotation of the methyl groups and the swinging of the imidazolate rings associated with the gate opening can be seen by comparing the marked areas

For clarity the N_2 molecules are not shown.

Gate-opening in a metal-organic framework



Measured (upper panel) and simulated (lower panel) INS spectra of blank ZIF-8 and ZIF-8+N_2.

The strong peaks are mainly due to vibrational modes involving large displacement of hydrogen (in the methyl groups and the imidazolate rings)

Casco, M. E. et al. Gate-opening effect in ZIF-8: the first experimental proof using inelastic neutron scattering. Chemical Communications, v. 52, n. 18, p. 3639-3642, 2016

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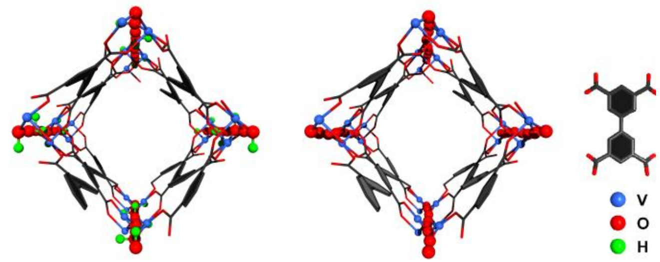
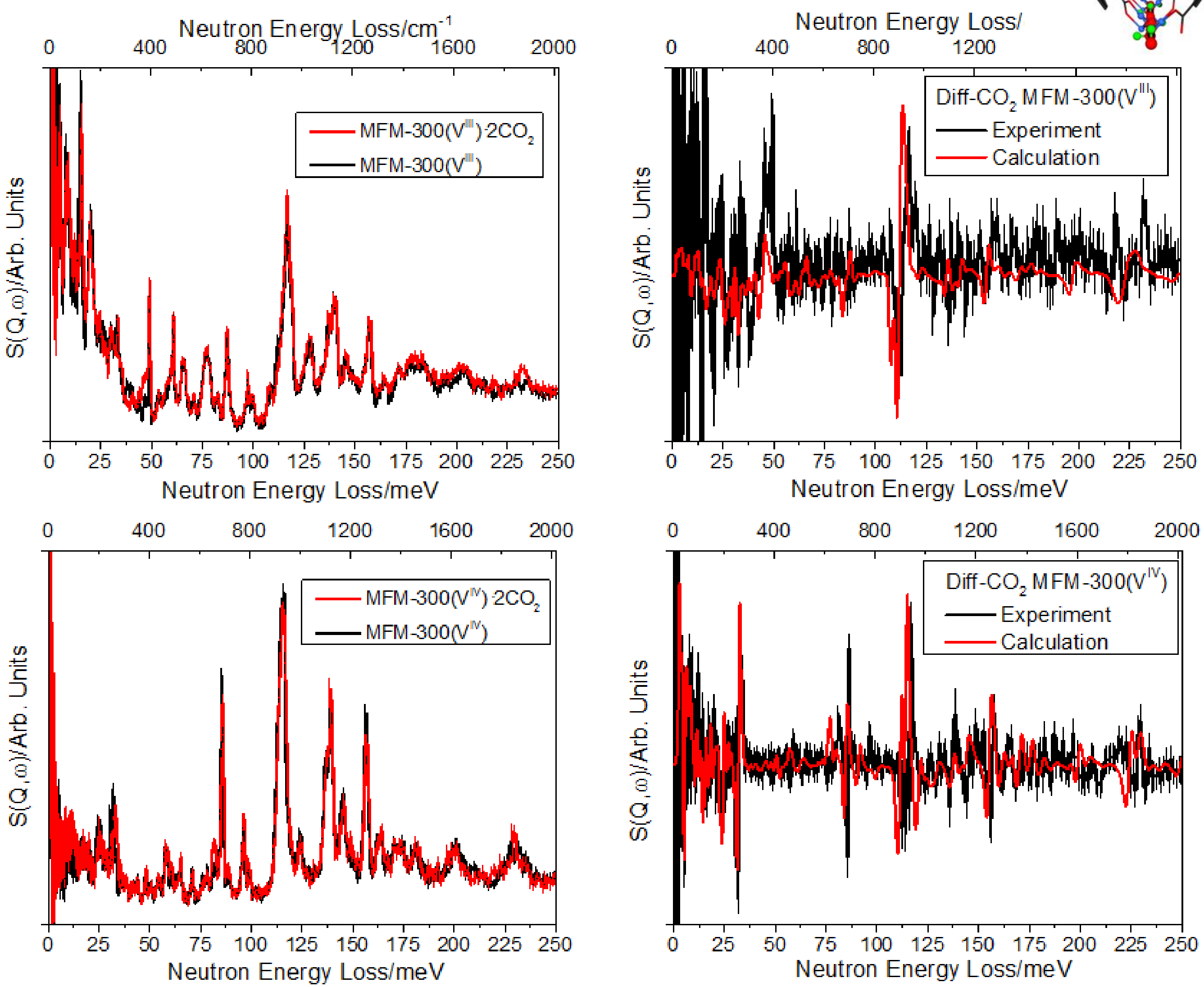
INS signature of CO_2 capture

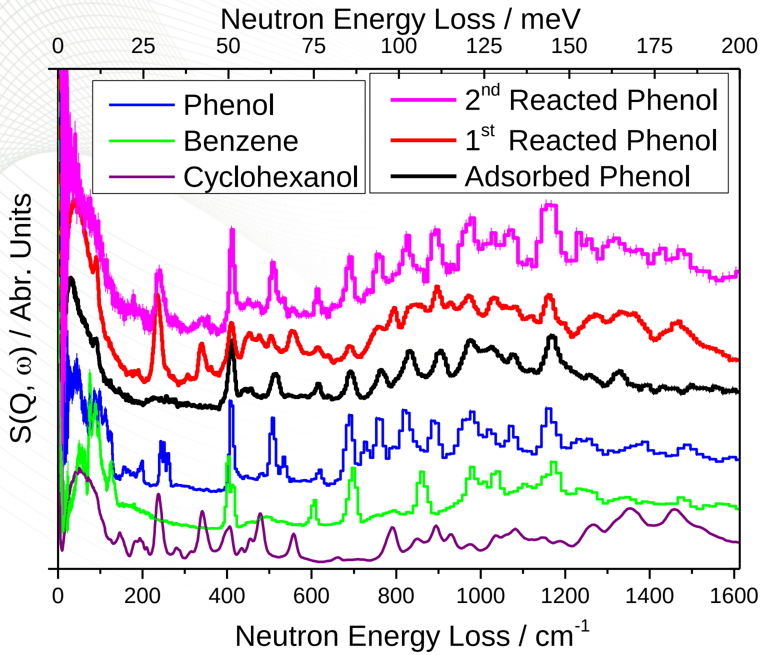


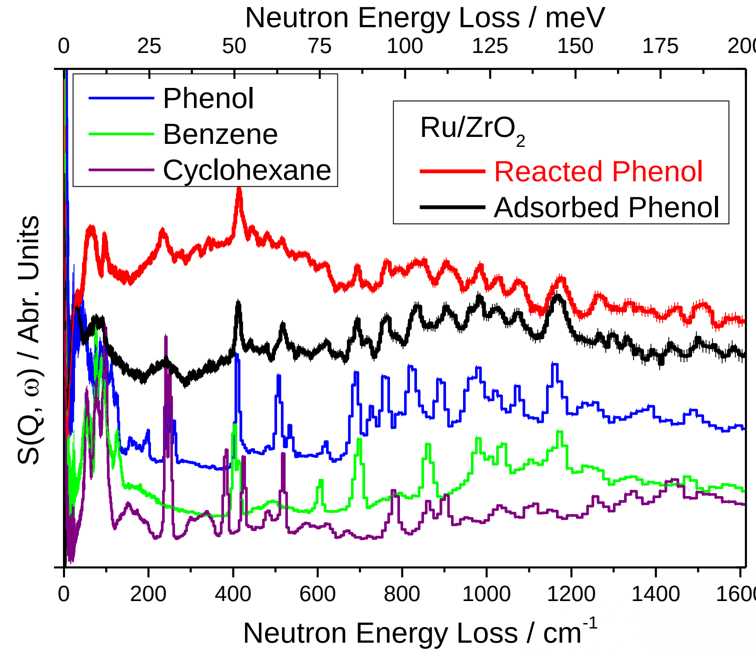


This study combining NPD, INS and modelling has unambiguously determined the CO_2 binding sites and structural dynamics for MFM-300(VIII) and MFM-300(VI) . It is confirmed that the proton on the hydroxy group can not only attract and localise adsorbed CO_2 molecules via direct formation of hydrogen bonds, but also affects the macroscopic packing and arrangement of CO_2 molecules in the extended channel.

Z. Lu et al. "Modulating Supramolecular Binding of Carbon Dioxide in a Redox-Active Porous Host“ under review at Nature Communications

Catalytic hydrodeoxygenation of phenol

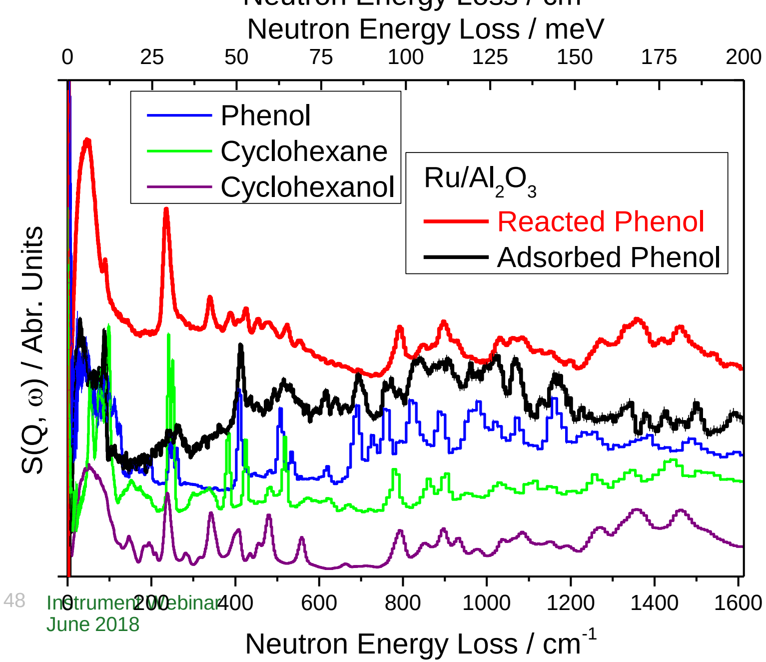


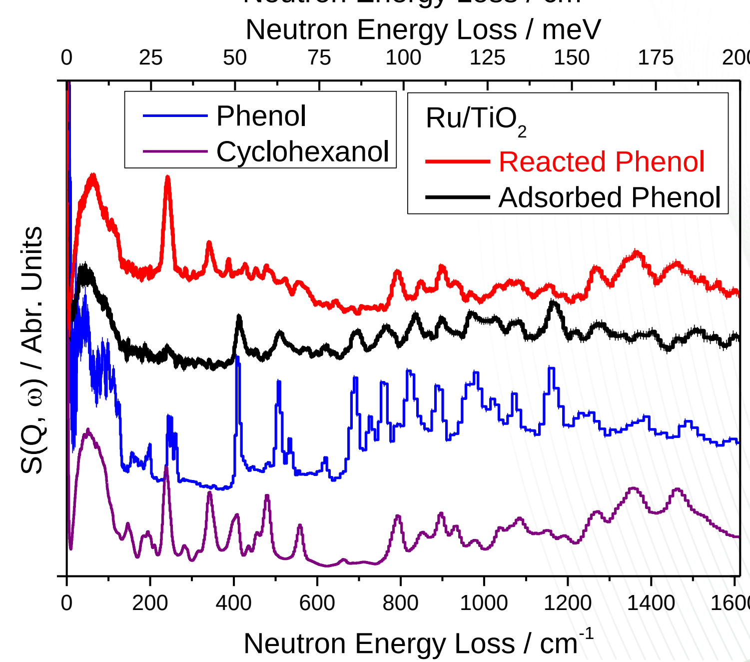


Main products:

Ru/Nb_2O_5:
Benzene
Cyclohexanol

Ru/ZrO_2:
Benzene
Cyclohexane

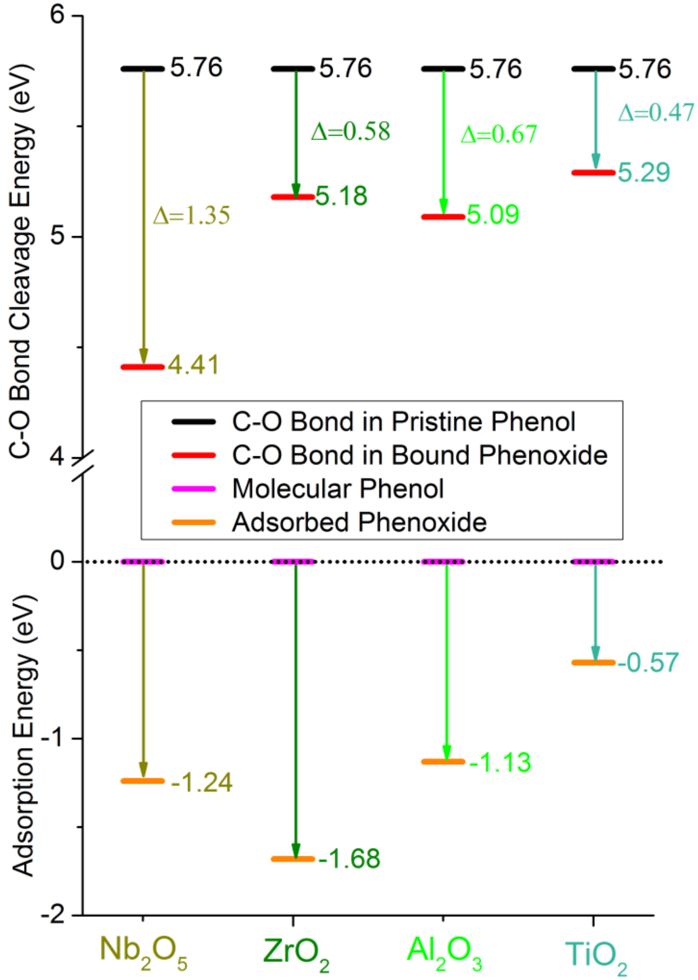


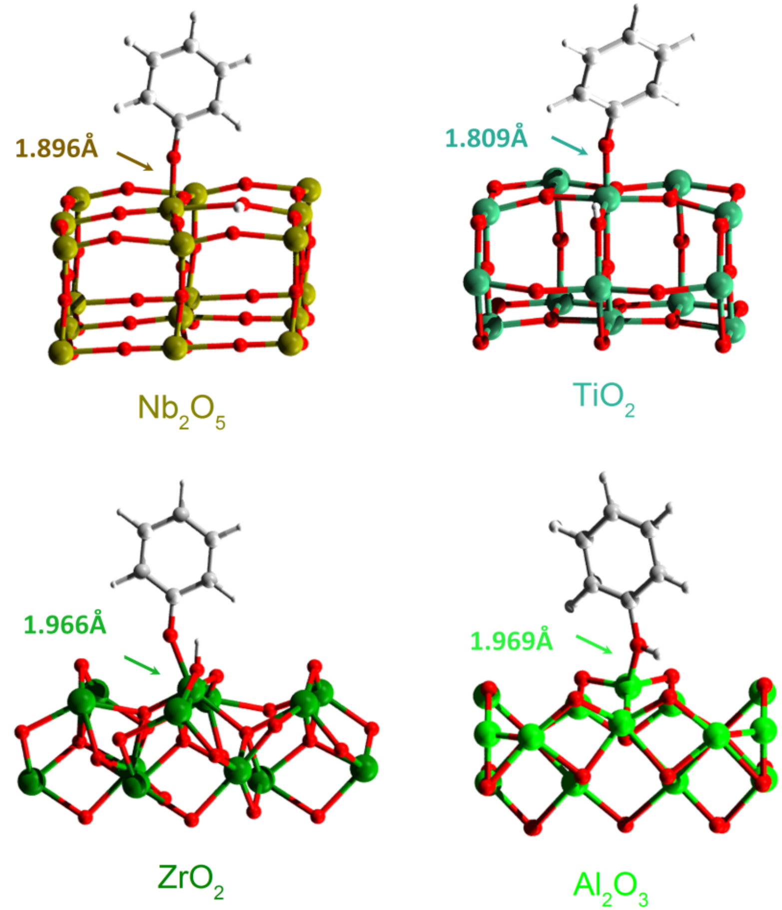


Ru/Al_2O_3:
Cyclohexane
Cyclohexanol

Ru/TiO_2:
Cyclohexanol

Catalytic hydrodeoxygenation of phenol





Shao, Y. et al. Selective production of arenes via direct lignin upgrading over a niobium-based catalyst. Nat. Commun. 8, 16104 (2017).

OH removal: strength of phenol chemisorption on the surface

Selectivity between the competitive processes of $C_{aromatic}$–O bond cleavage and C6-ring hydrogenation: reduction in C-O bond cleavage energy

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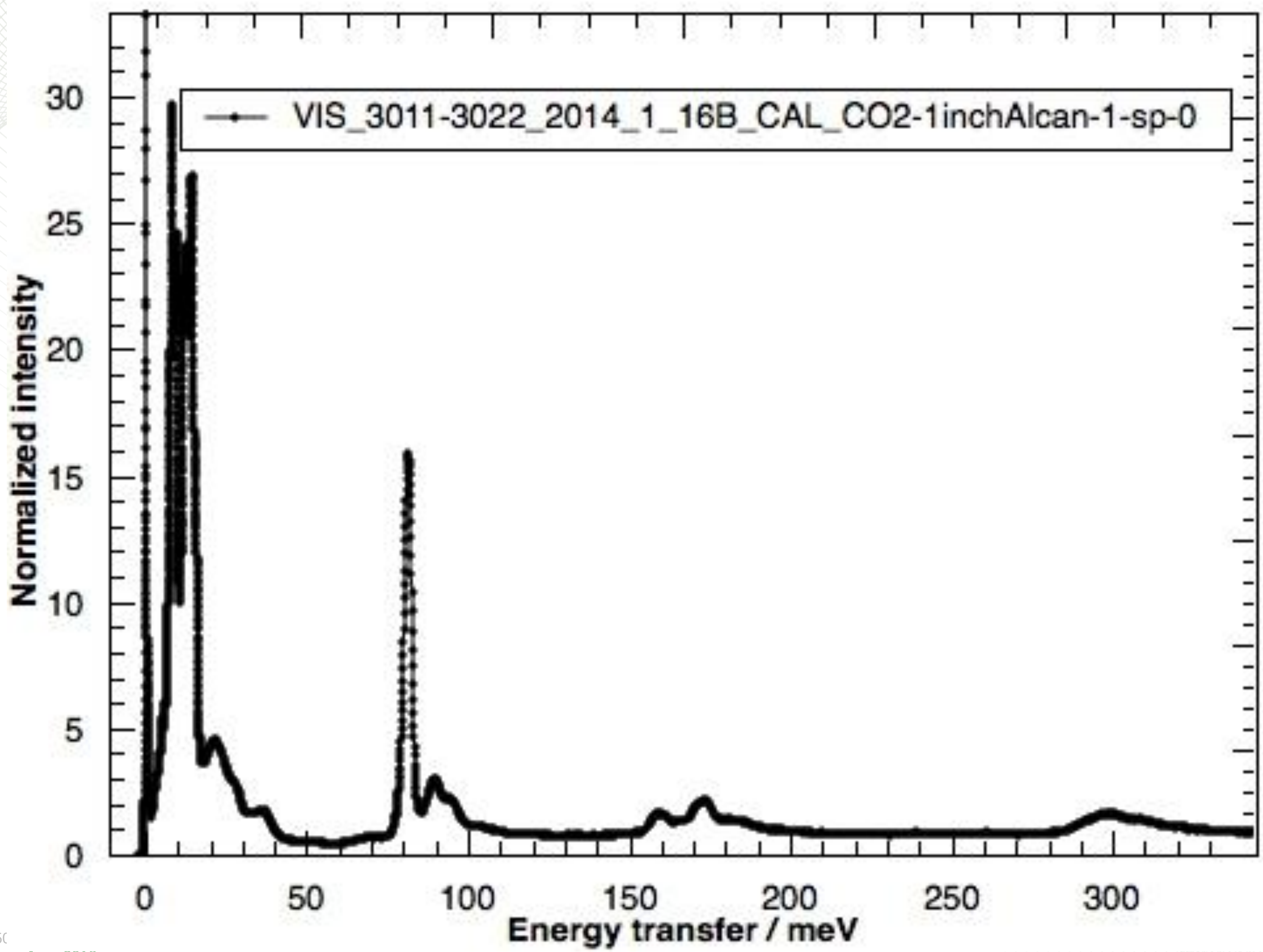
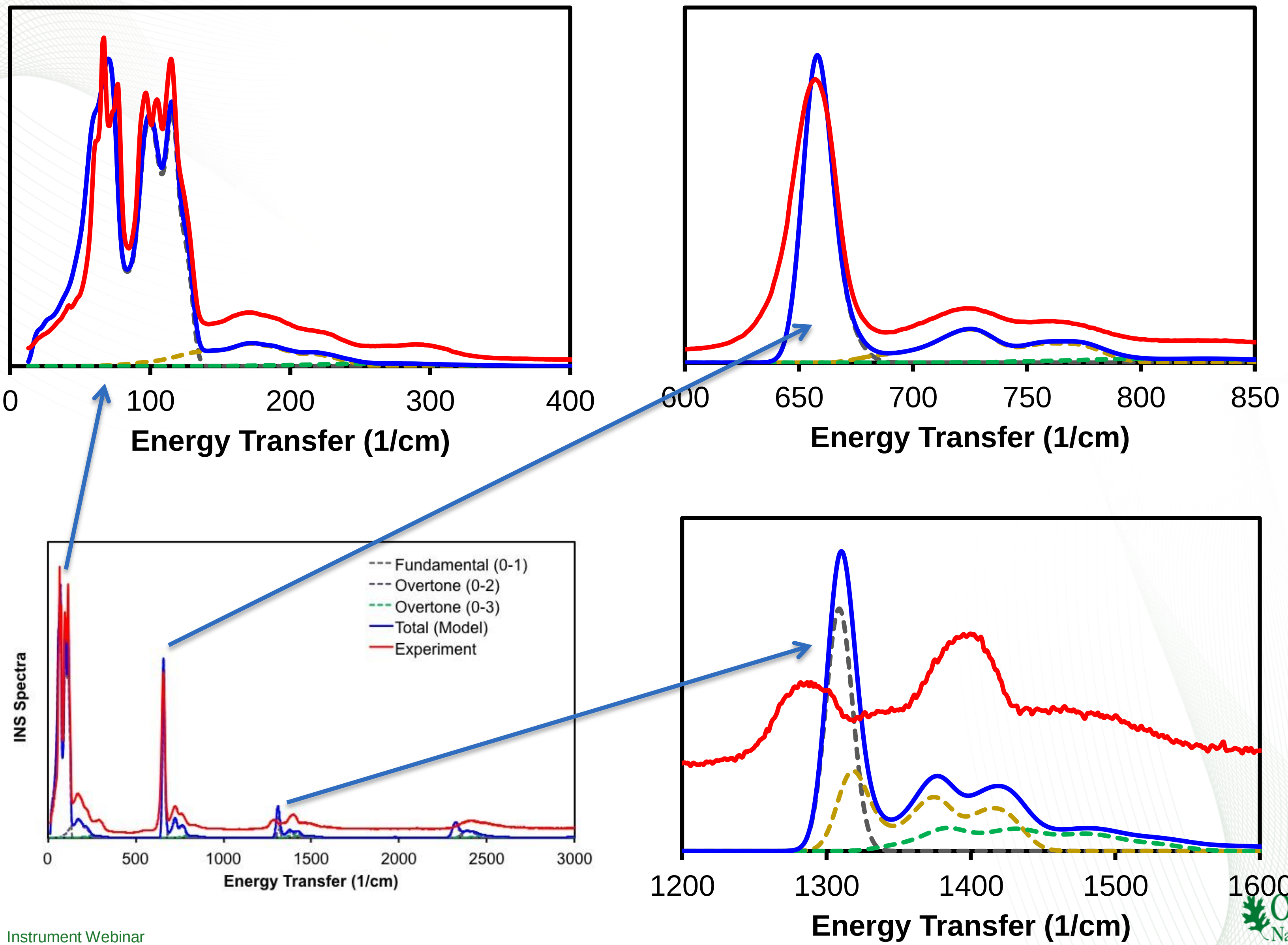
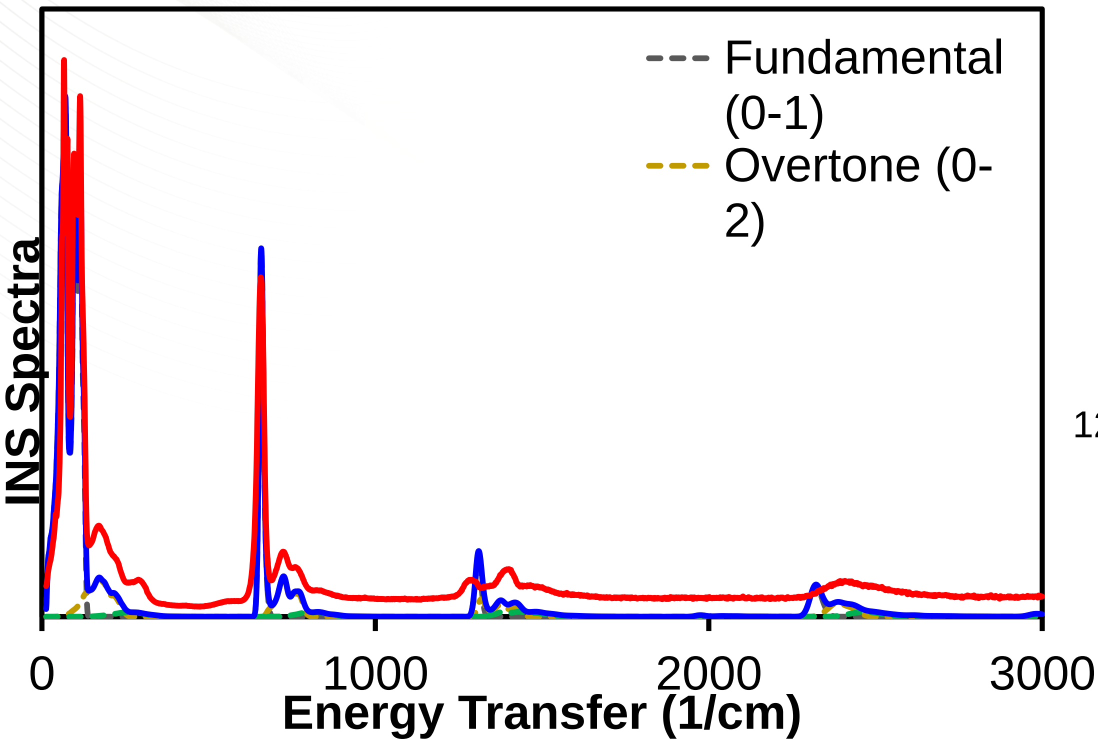
CO_2

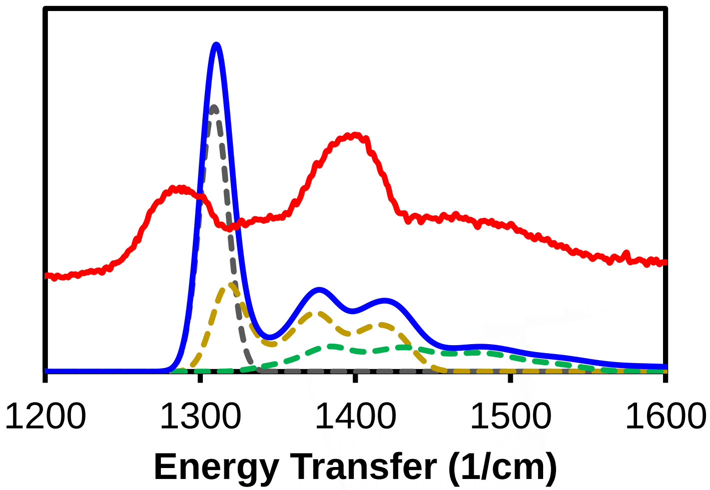

VIS_3011-3022_2014_1_16B_CAL_CO2-1inchAlcan-1-sp-0
Normalized intensity
Energy transfer / meV
0
5
10
15
20
25
30
0
50
100
150
200
250
300

CO_2 in the solid phase



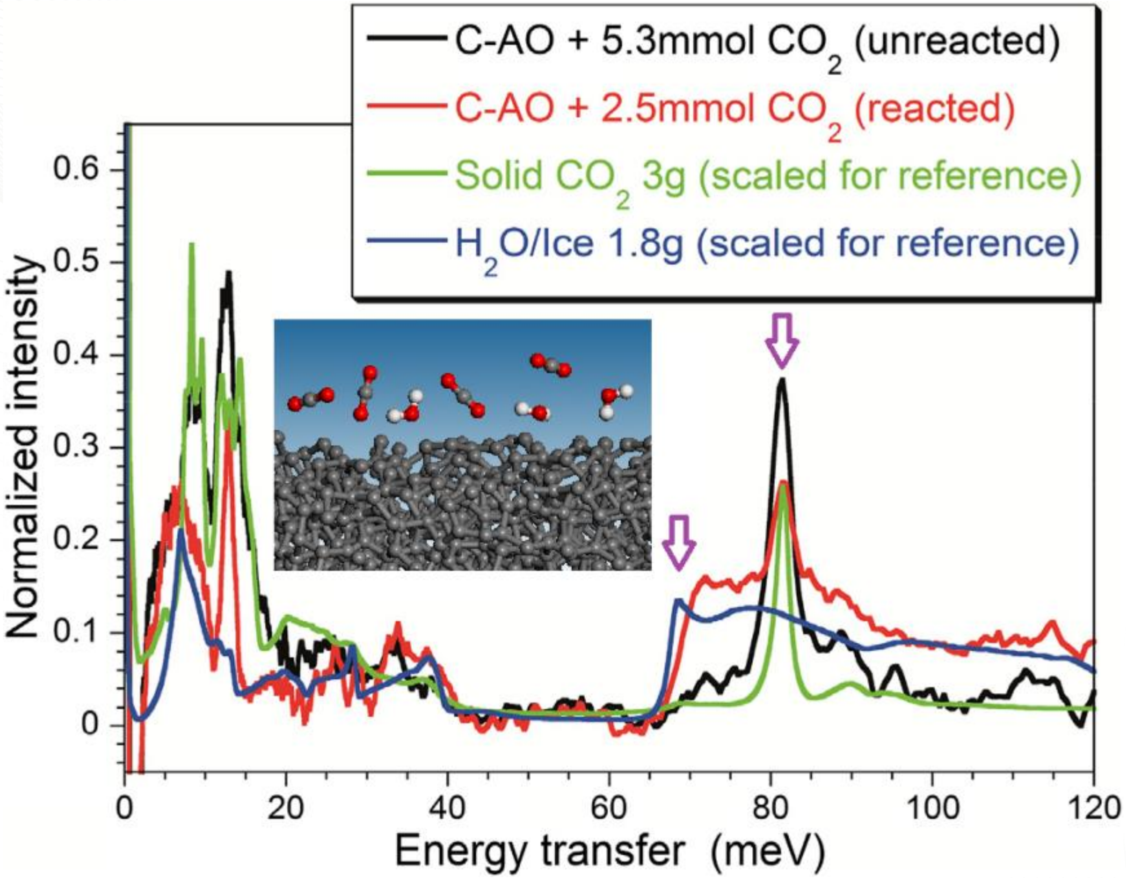
Examples from VISION





- Overlap of the an overtone (of the translational modes) with a fundamental (the C-O symmetric stretching): the *Fermi resonance*
- First observation of CO_2 Fermi resonance *using INS*

Small amount of non-hydrogenous samples

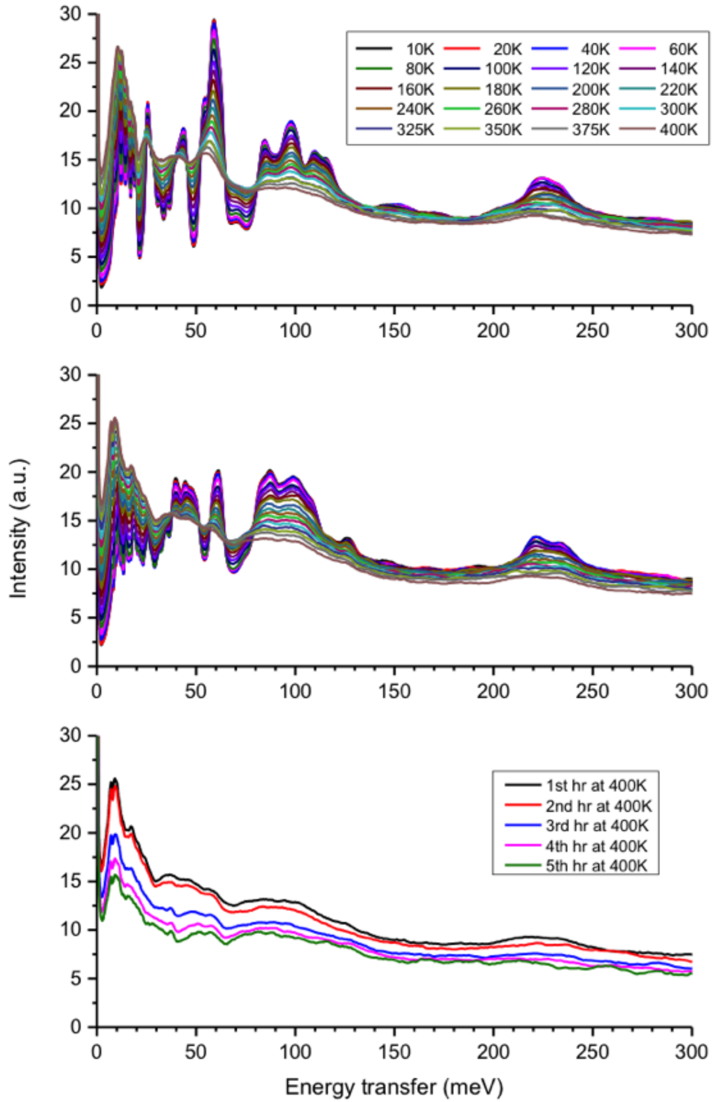


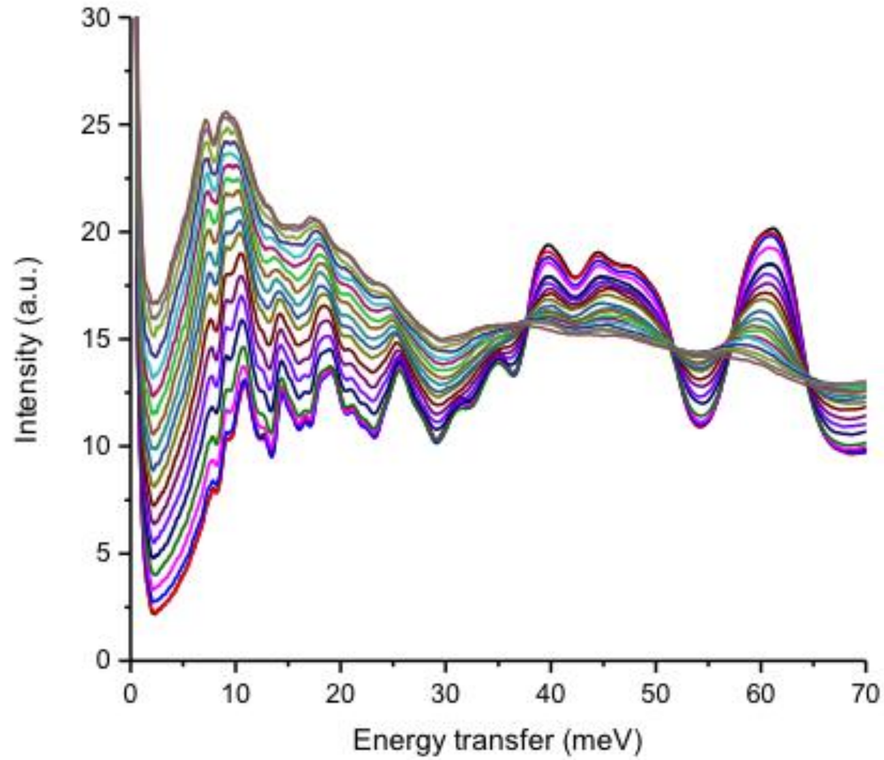
The difference INS spectra before and after CO_2 dosing in C-AO (a nanoporous carbon sample), in comparison with the reference spectra for bulk solid CO_2 and H_2O. Signal from the background and the blank C-AO has been subtracted.

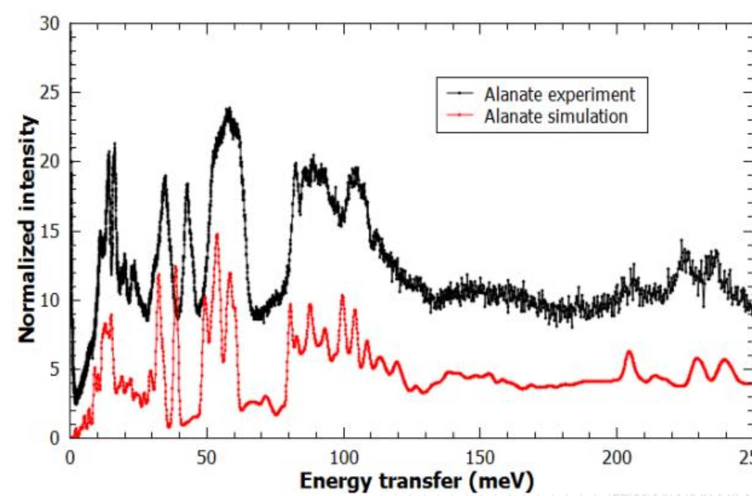
Very small amount of non-hydrogenous gas. In situ observation of surface reactions. Surface science, catalysis, gas capture and storage.

T. J. Bandosz, M. Seredych, E. Rodríguez-Castellón, Y. Q. Cheng, L. L. Daemen, and A. J. Ramirez-Cuesta. *Carbon*. 96 (2016): 856–863.

Hydrogen in metal alanates

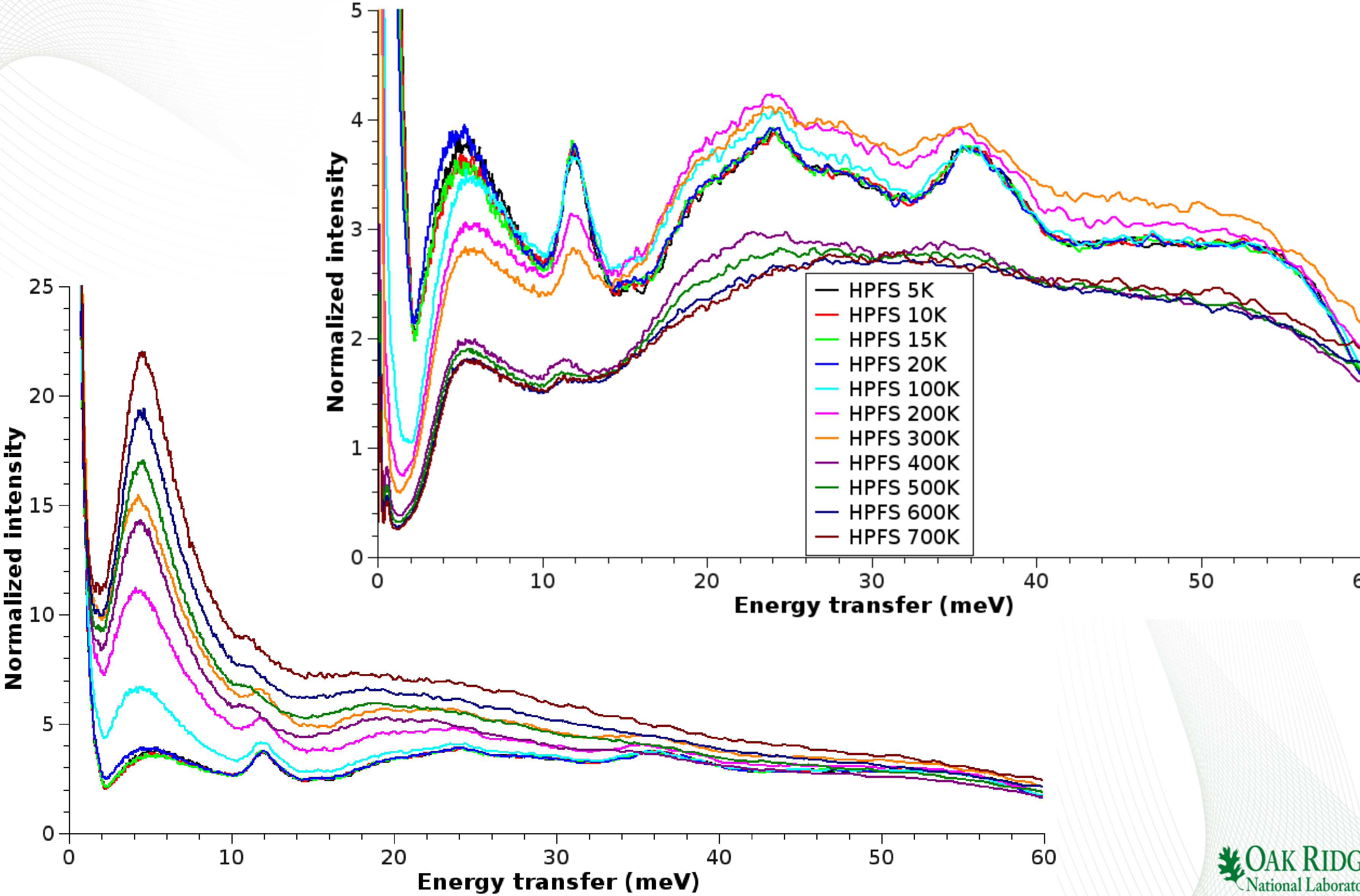




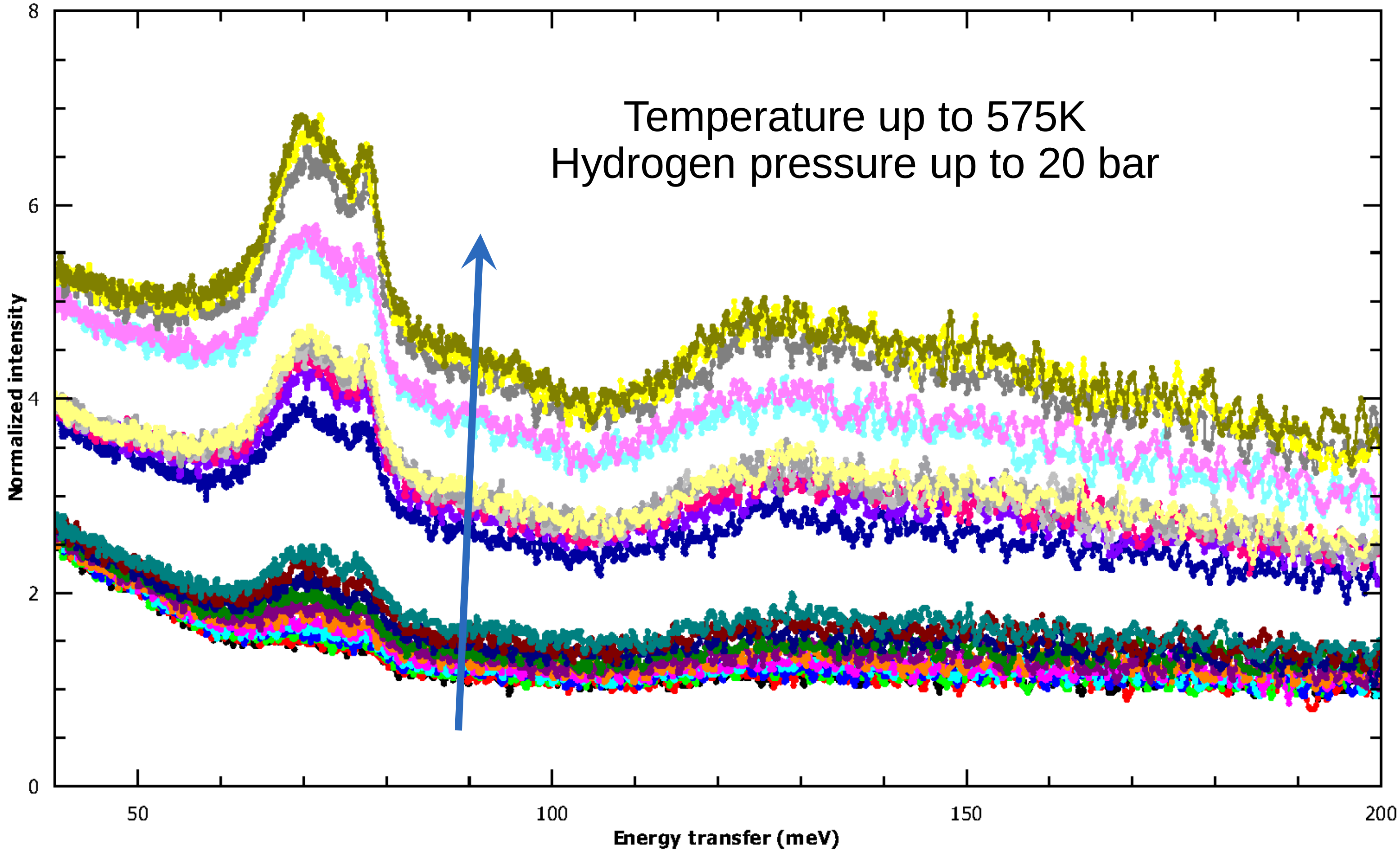


Thanks to Shin-ichi Orimo, Toyoto Sato, Shigeyuki Takagi and Keisuke Tomiyasu, Tohoku University, Japan.

High temperature measurement up to 700K



In situ observation of metal hydride formation



Collaboration with Jacques Huot at UQTR Canada.

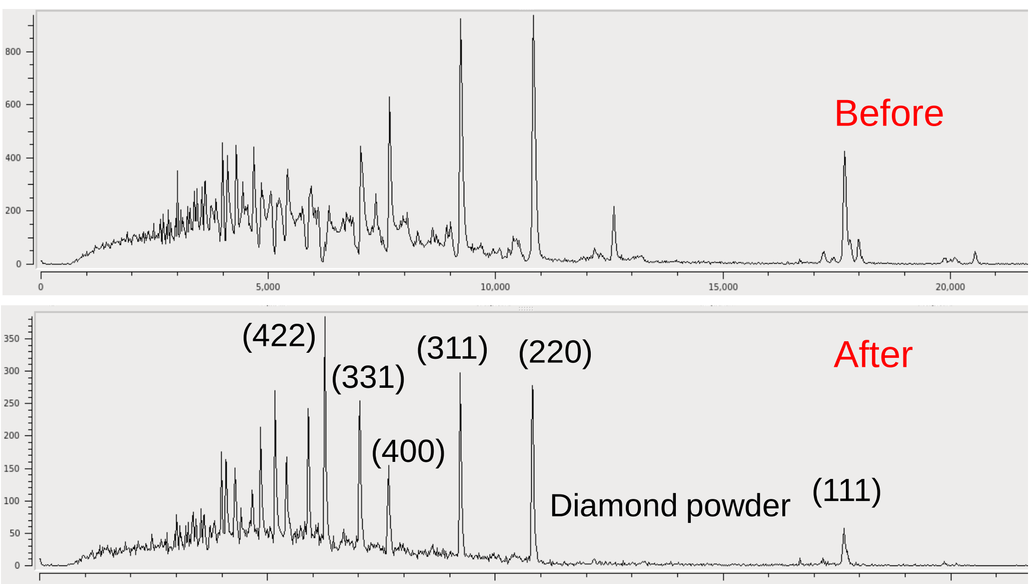
Diffraction at VISION

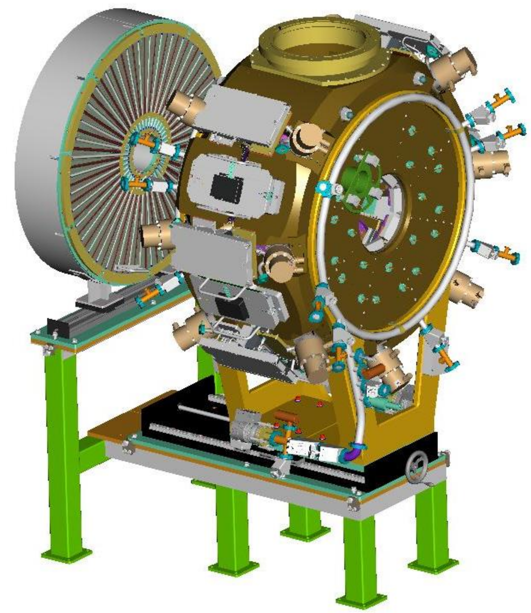
Simultaneous diffraction and inelastic neutron scattering

3D printed collimators have been tested for VISION to be used in the backscattering diffraction bank.

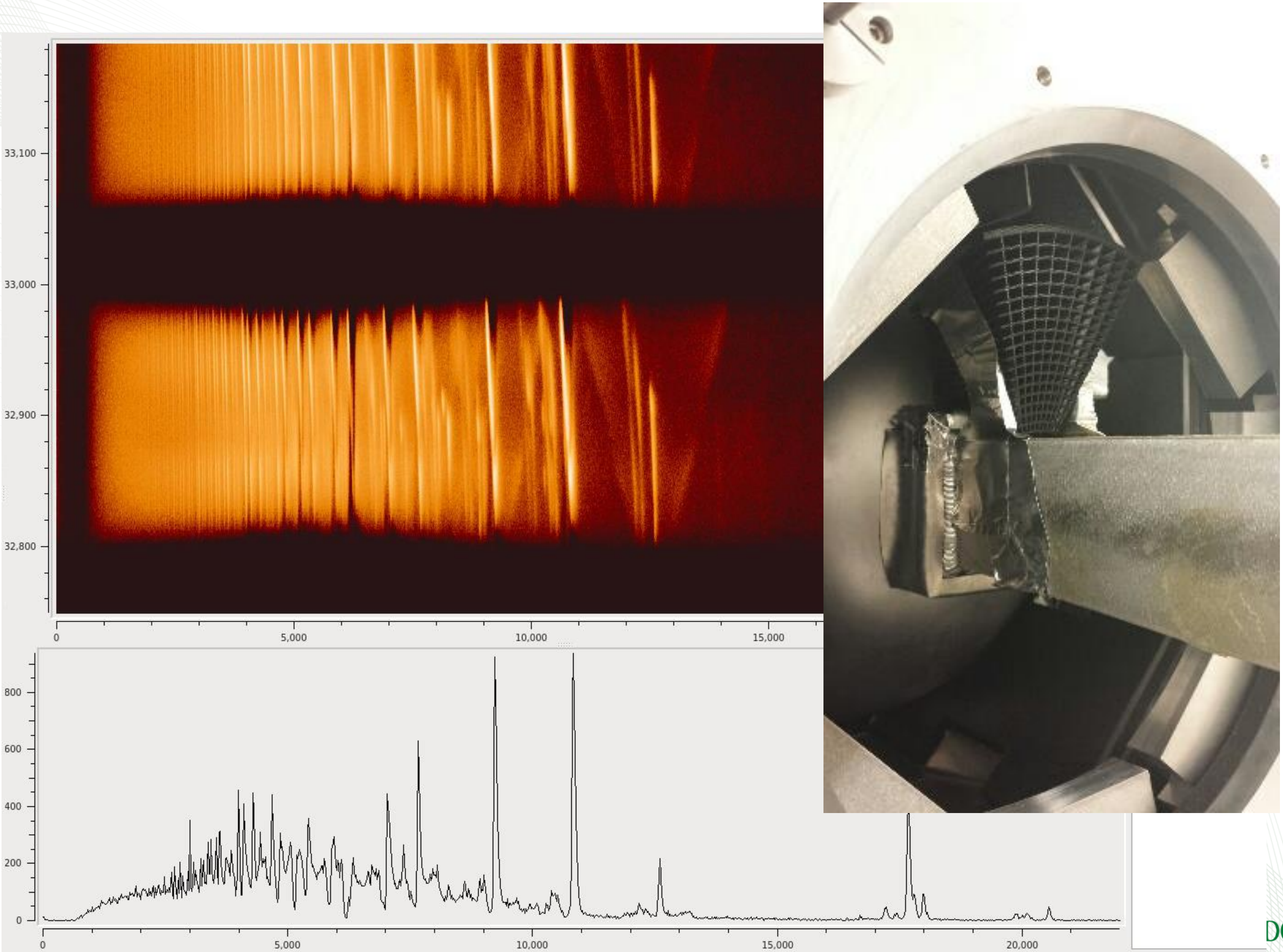
The reduction of the spurious peaks from the sample is very much noticeable.

Data collected in histogram or event mode

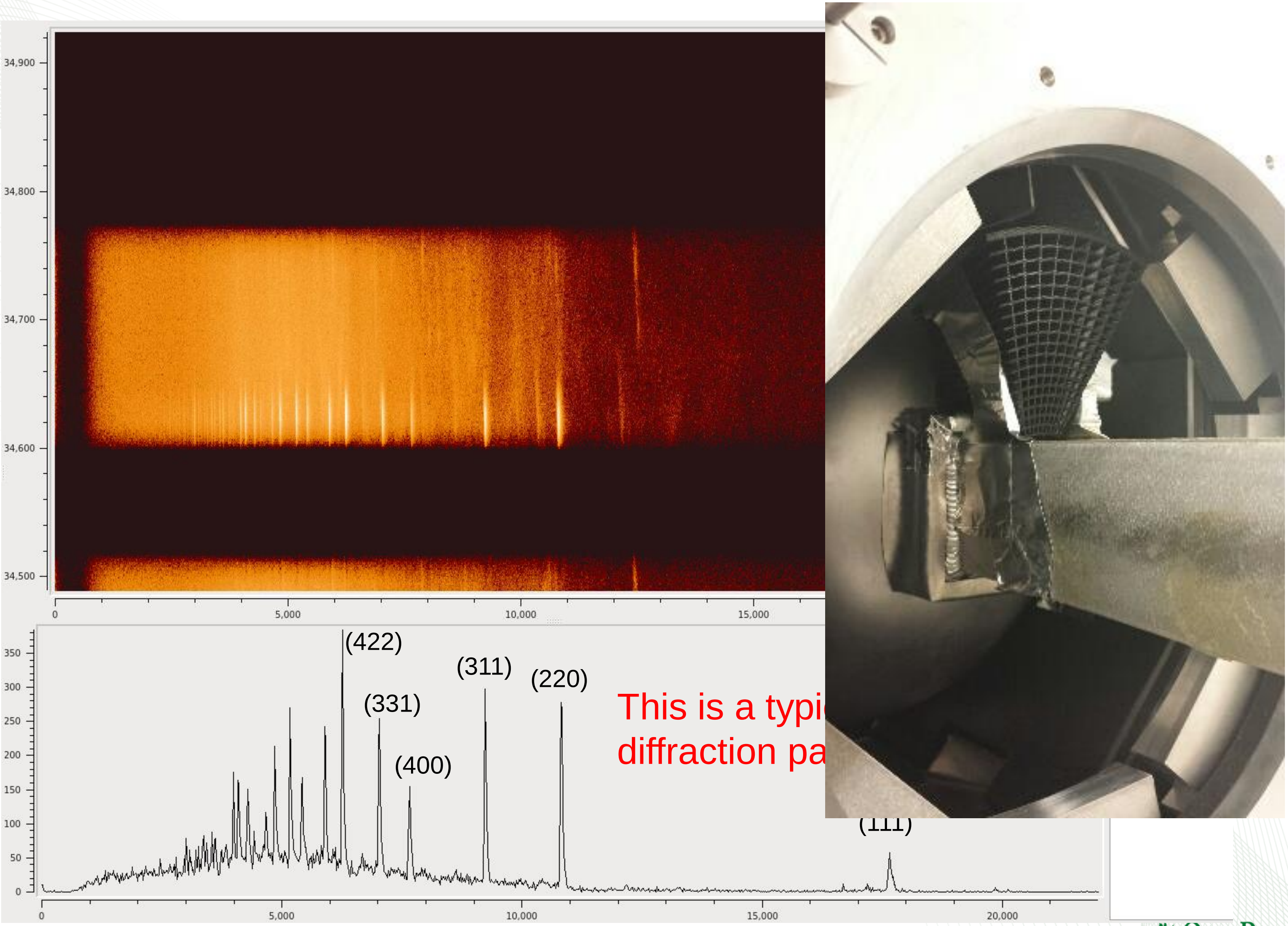




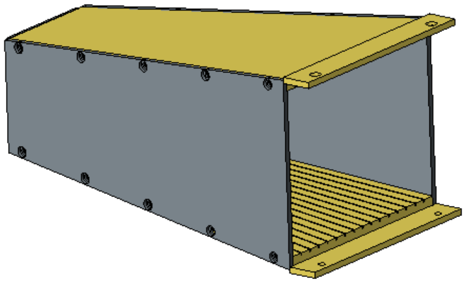
Bank 24, Tube 1 (NOT covered by collimator)

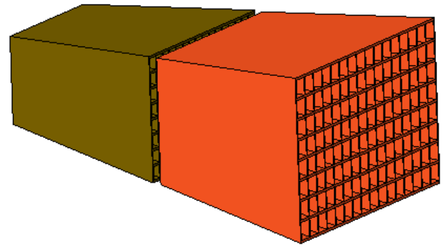


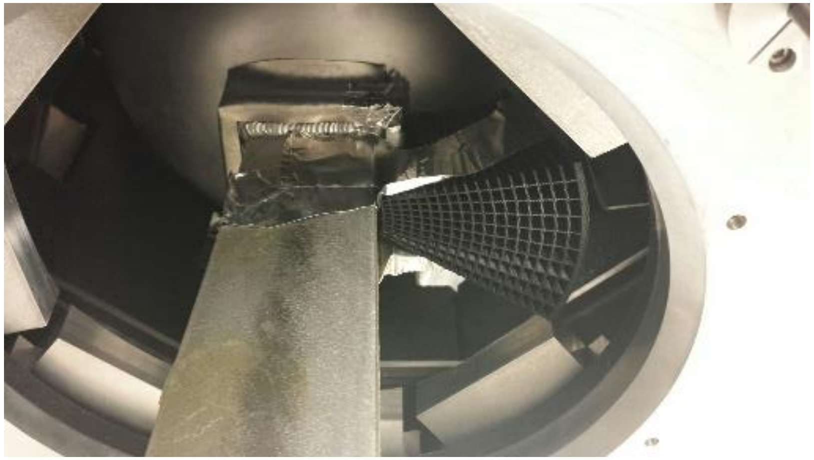
Bank 24, Tube 8 (covered by collimator)

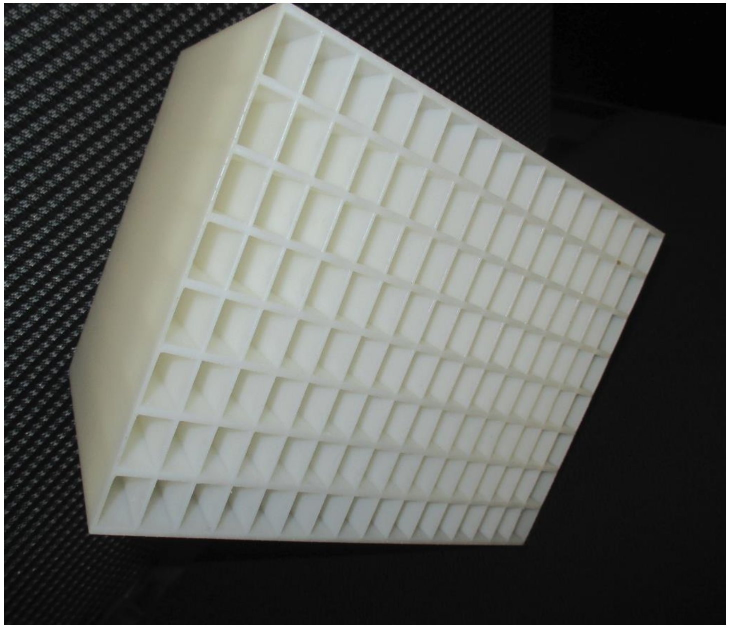


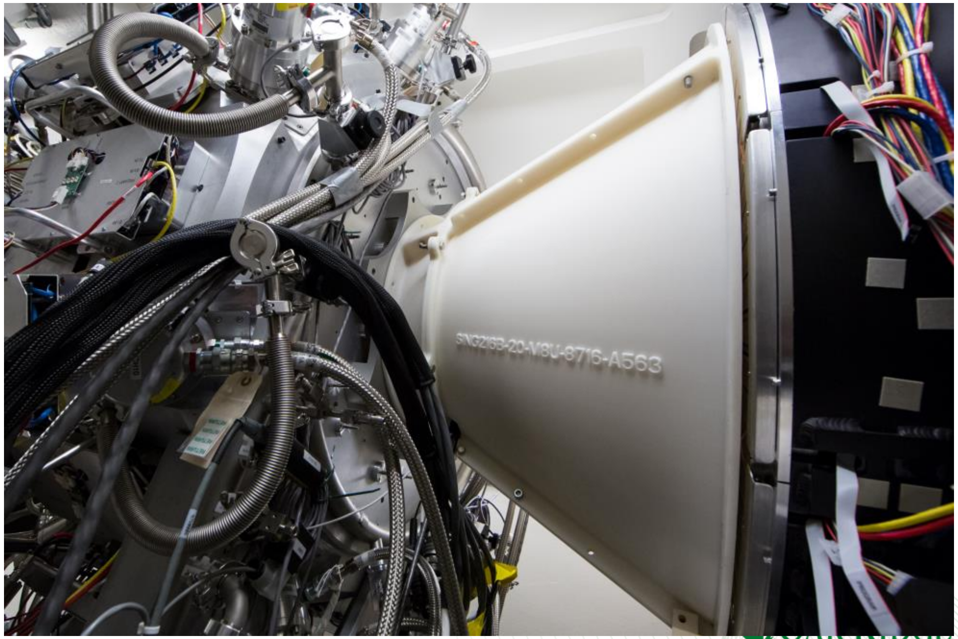
VISION pioneered the use of 3D printed collimators



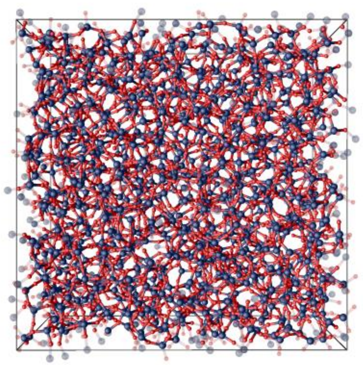


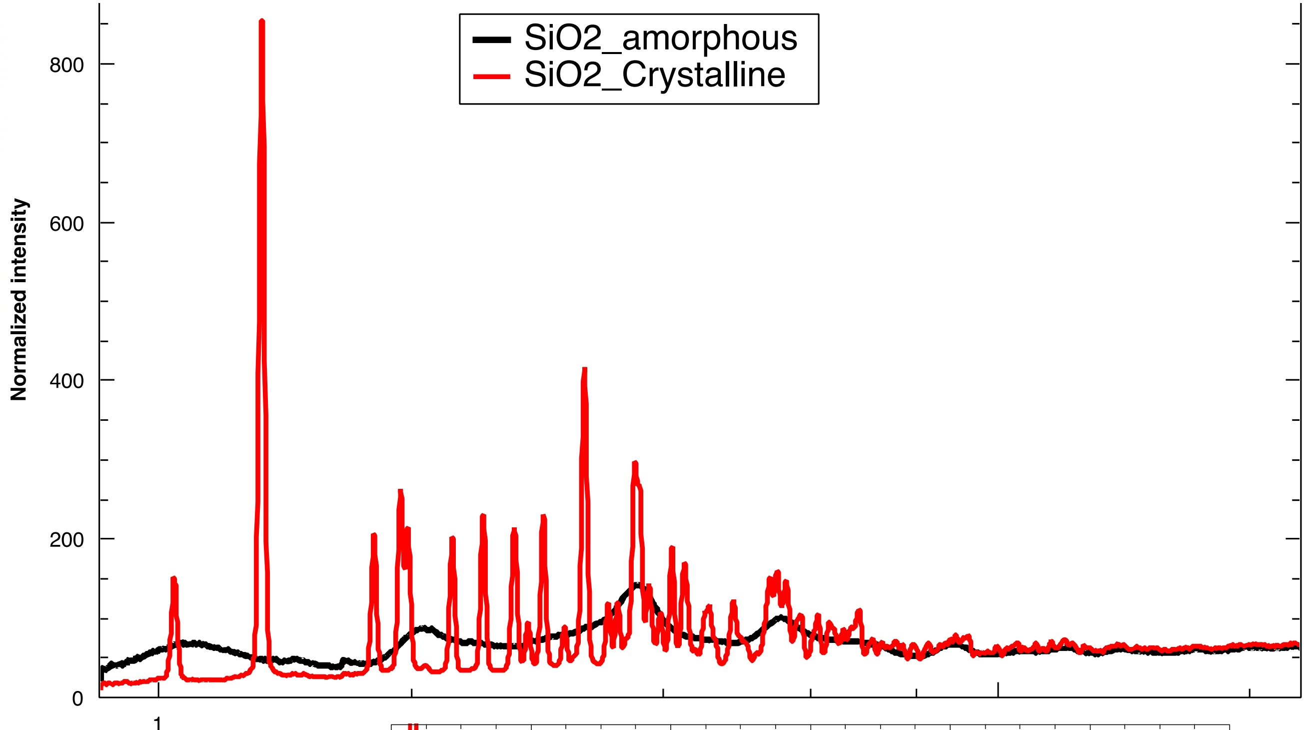


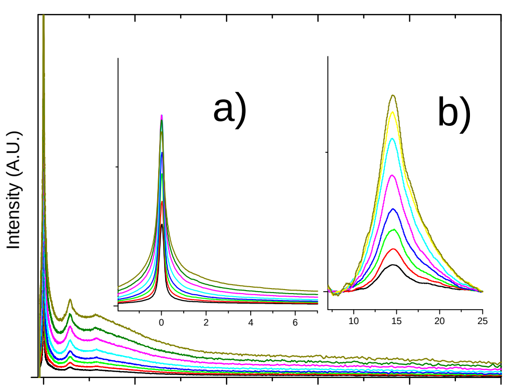


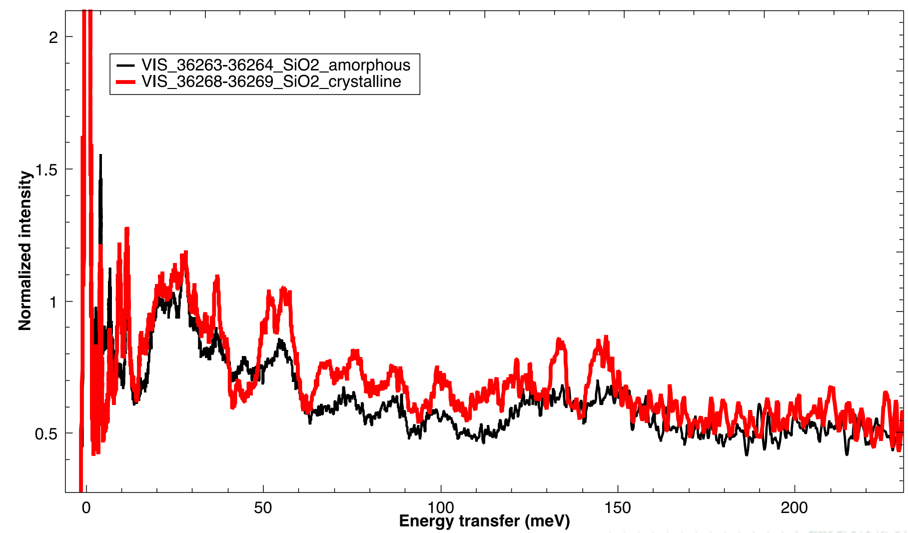


VISION: Inelastic, Diffraction and QENS

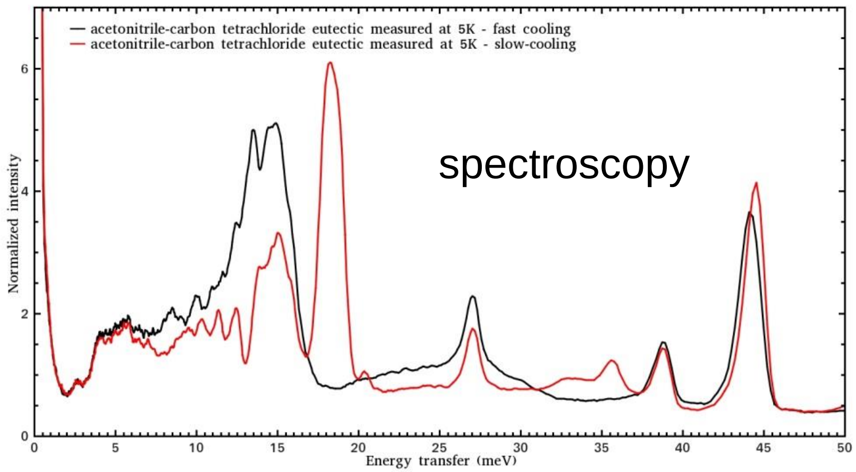


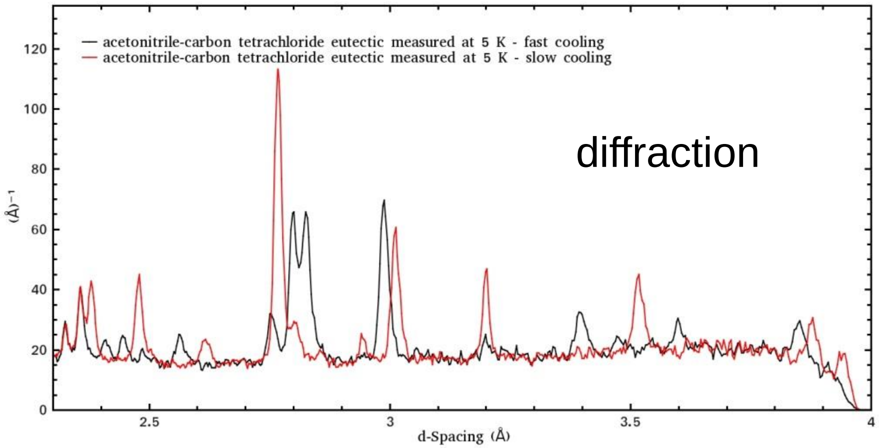






Structure and dynamics of liquids and solutions





- CD_3CN-CCl_4 (deep) eutectic system

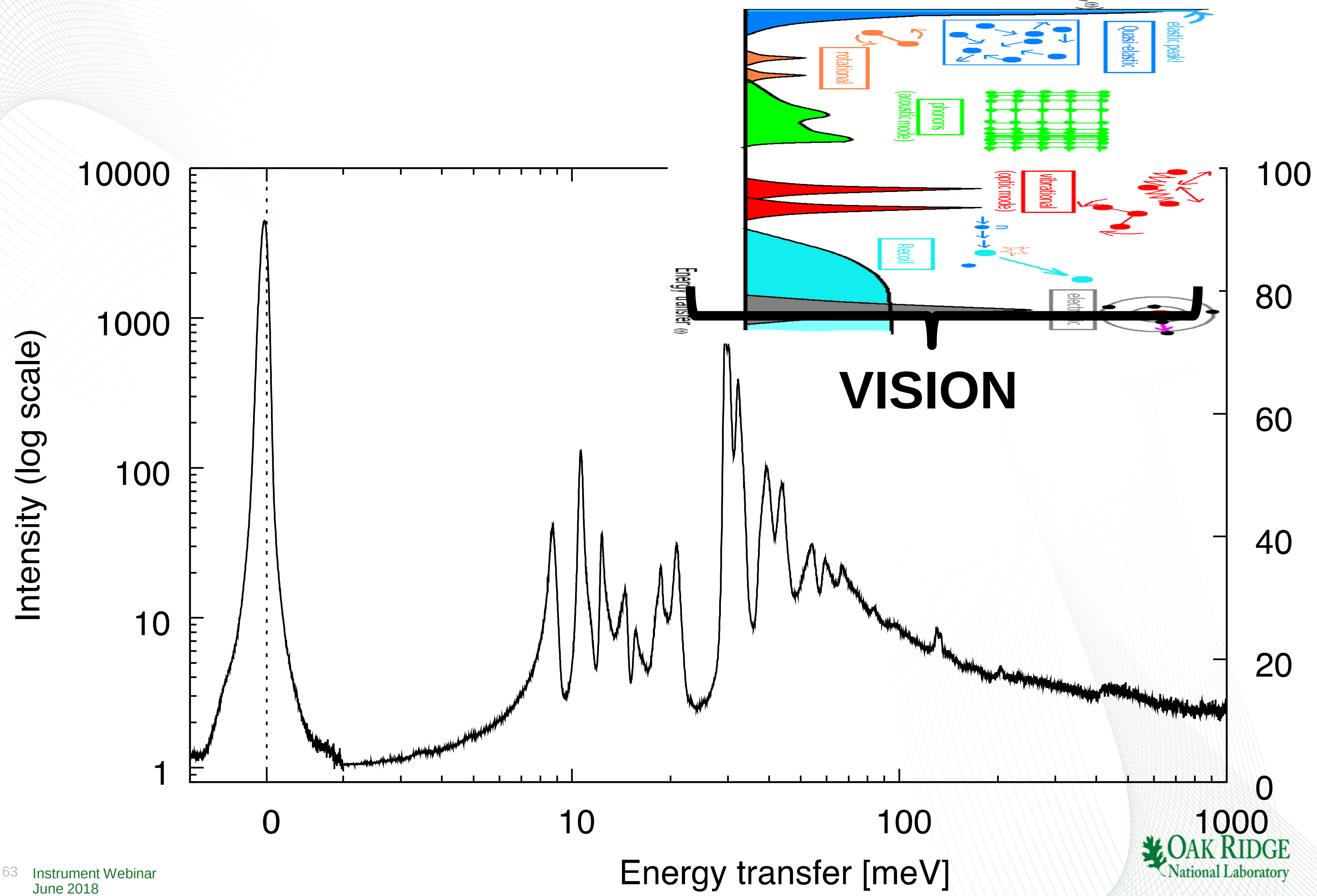
- T_E = 210 K; x(CD_3CN) at eutectic composition is 0.75

- no hydrogen bonding, but highly non-ideal system with ΔH_{excess} = - 800 J/mol)

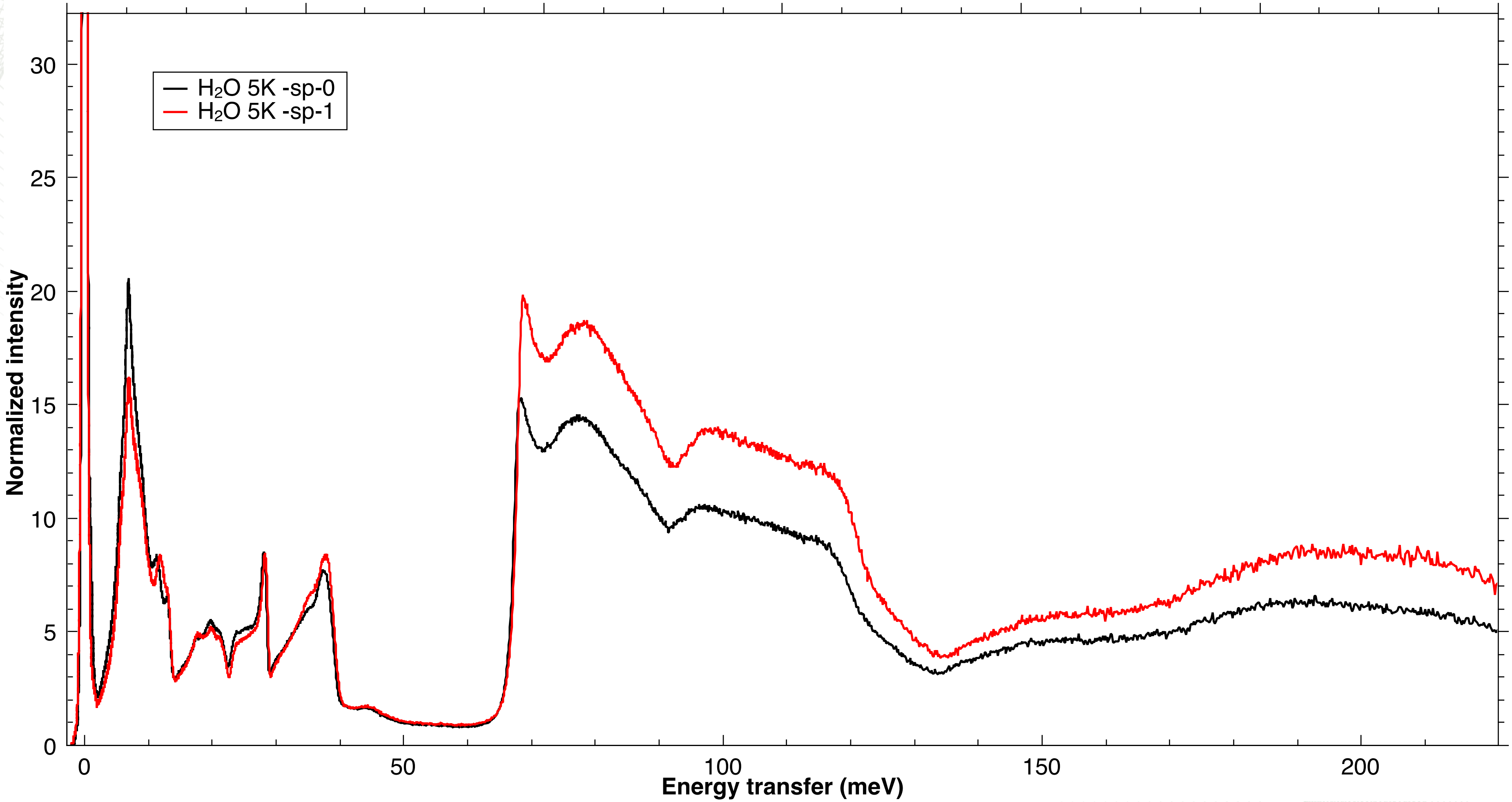
- eutectic structure differs when liquid is cooled quickly or slowly from room temperature

- simultaneous diffraction/spectroscopy is invaluable

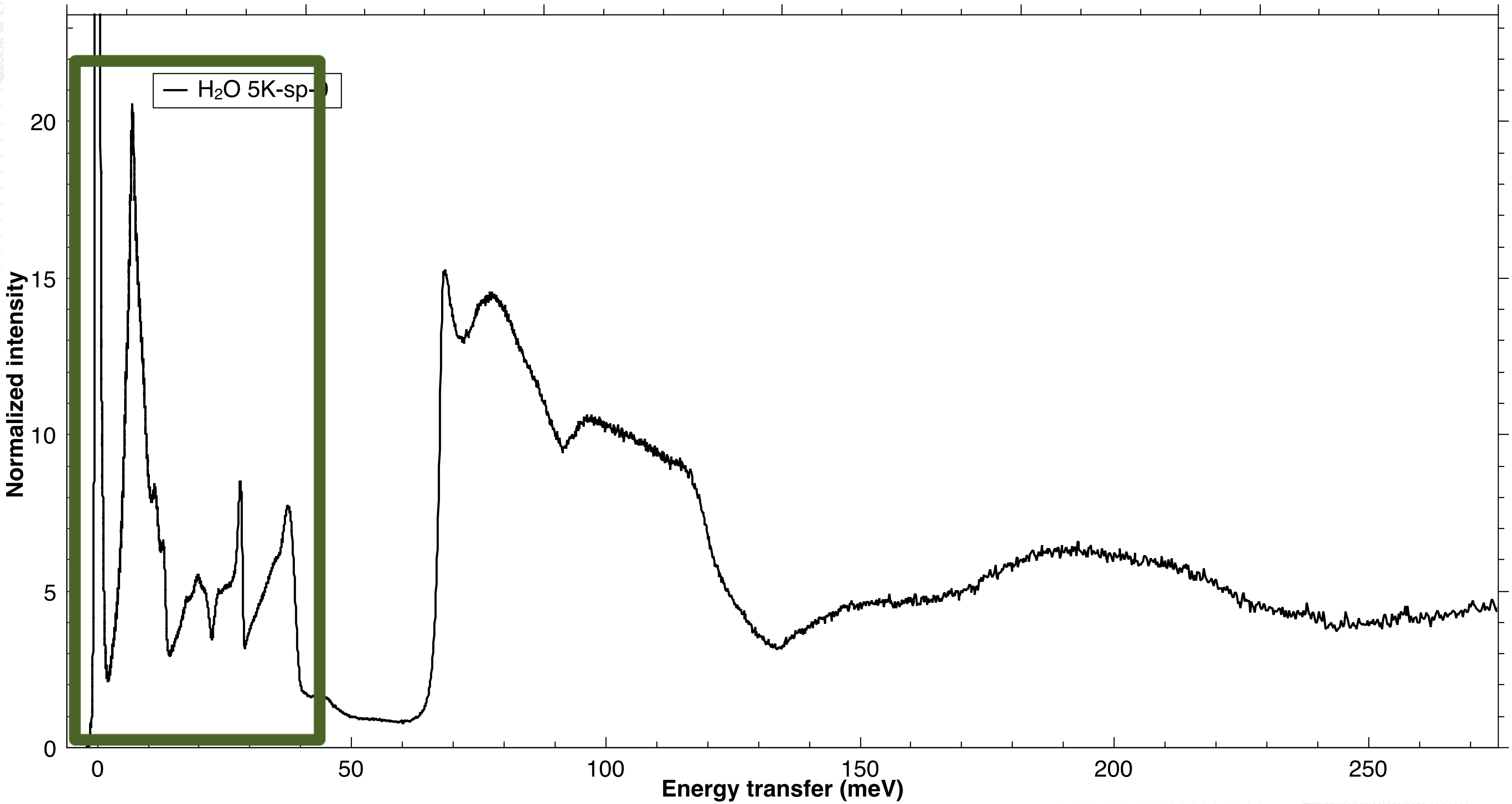
The energy spectra (according to VISION)



Water (ice) in VISION



Water (ice) in VISION



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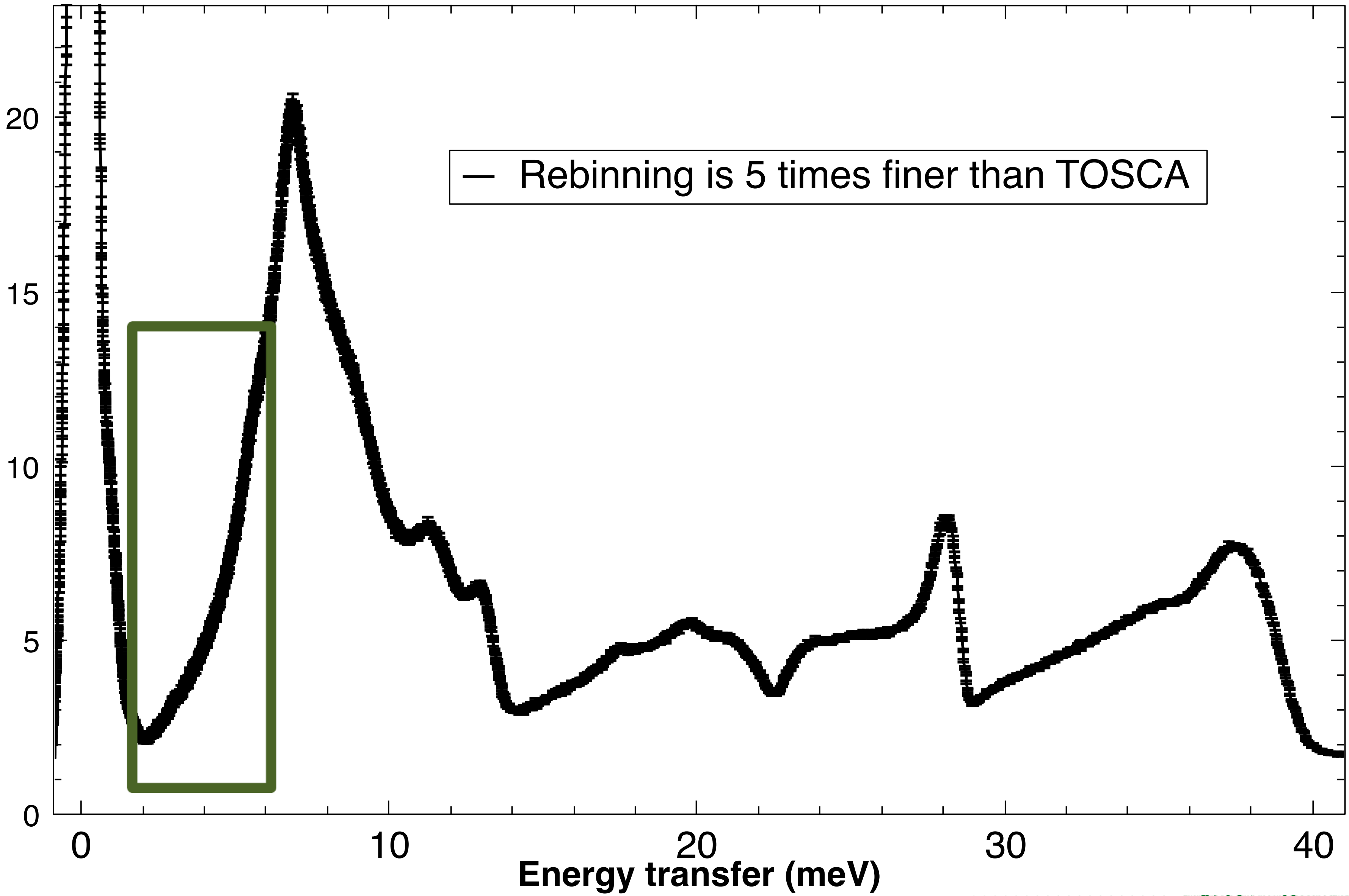
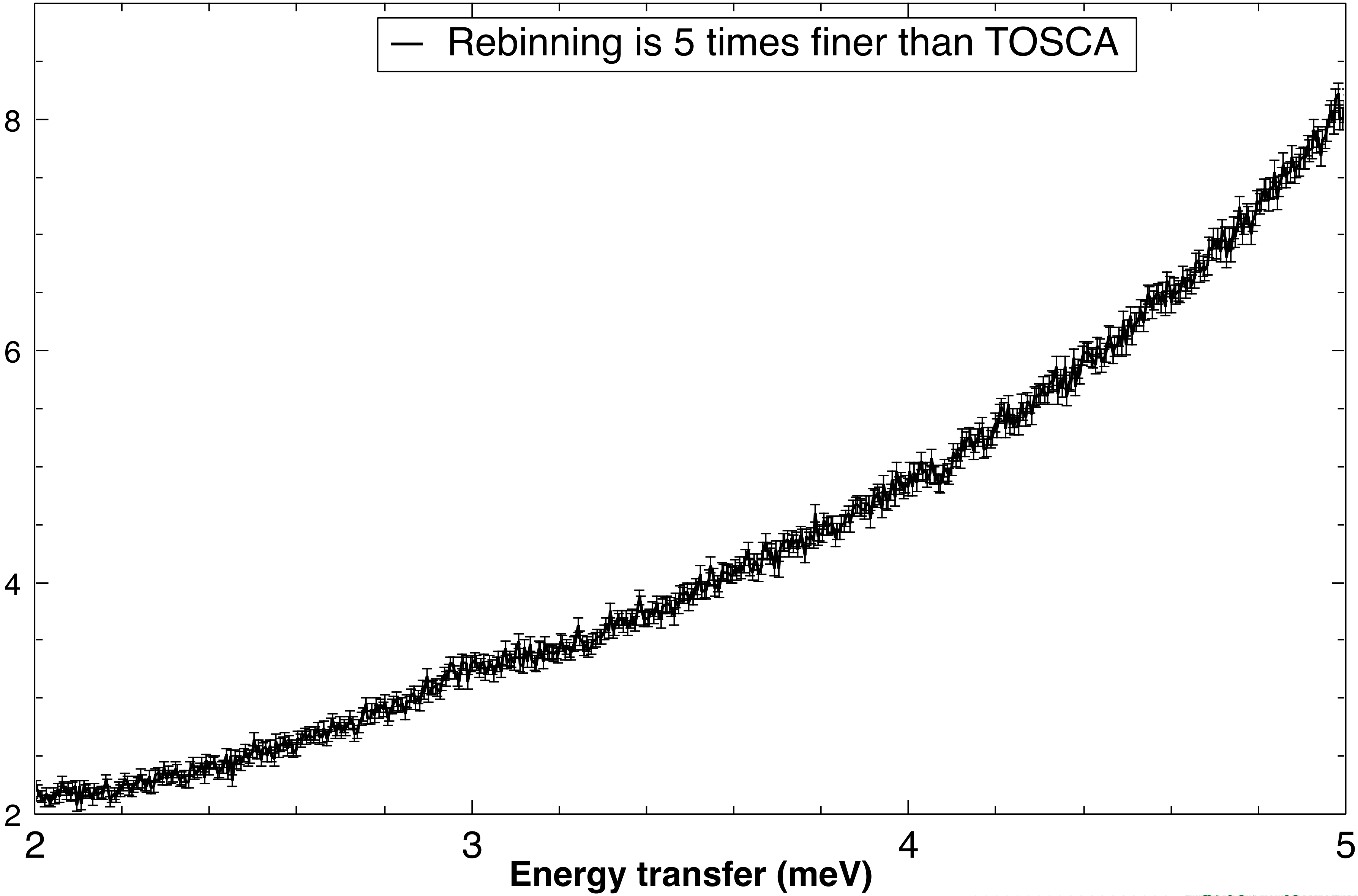
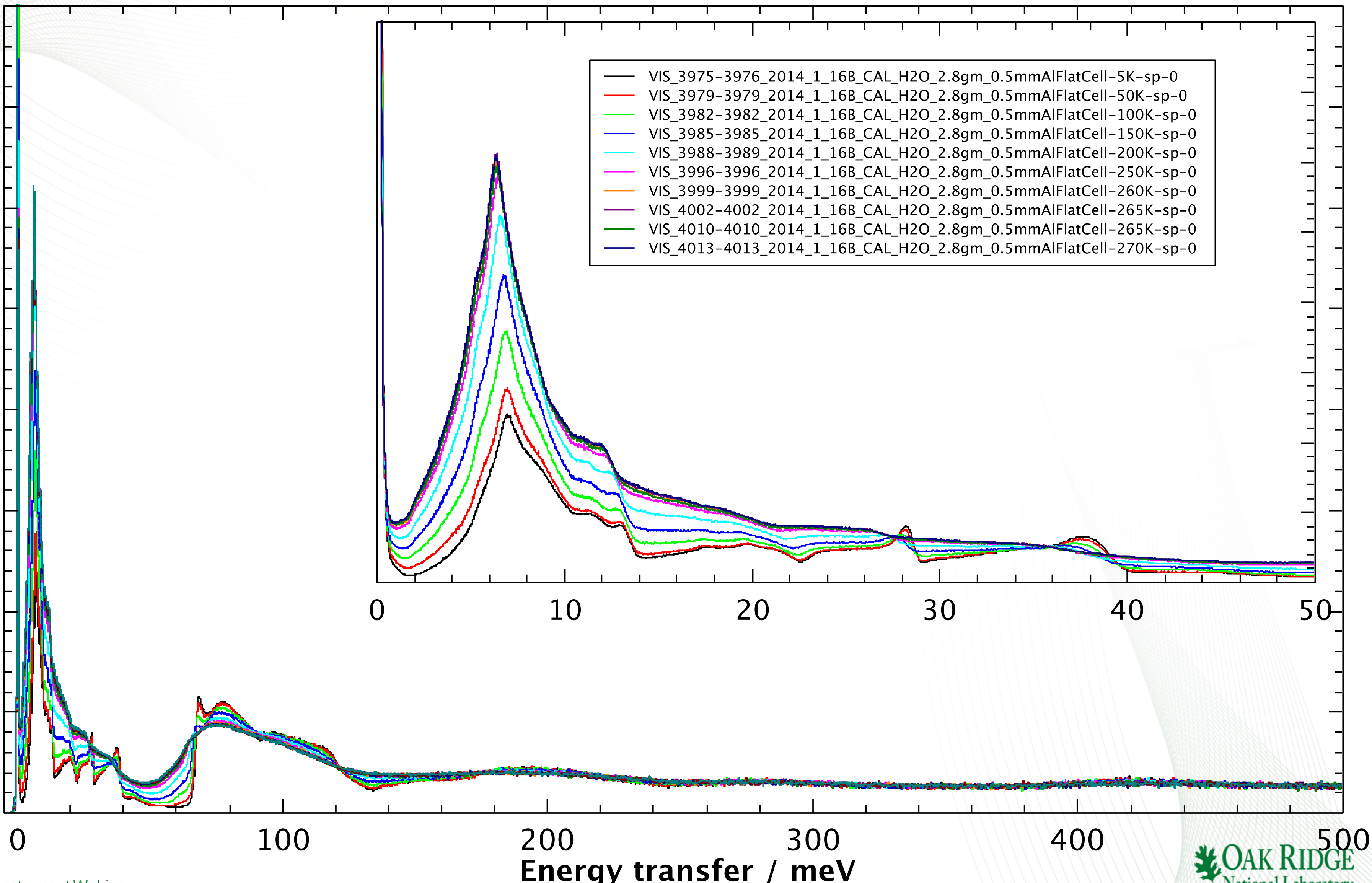
Water (ice) in VISION Zoomed in



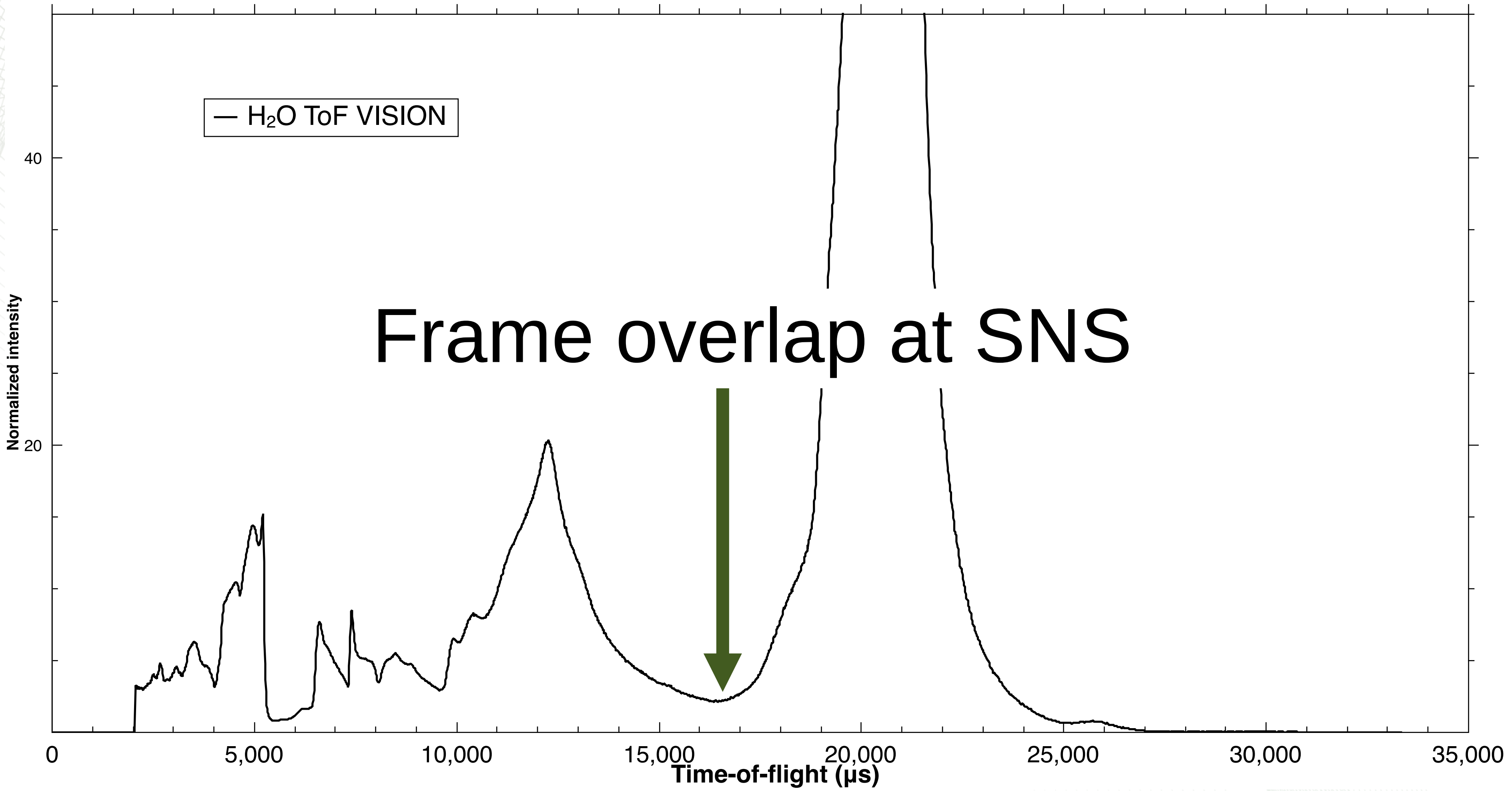
Water (ice) in VISION Zoomed in



Water in VISION
(as function of temperature)



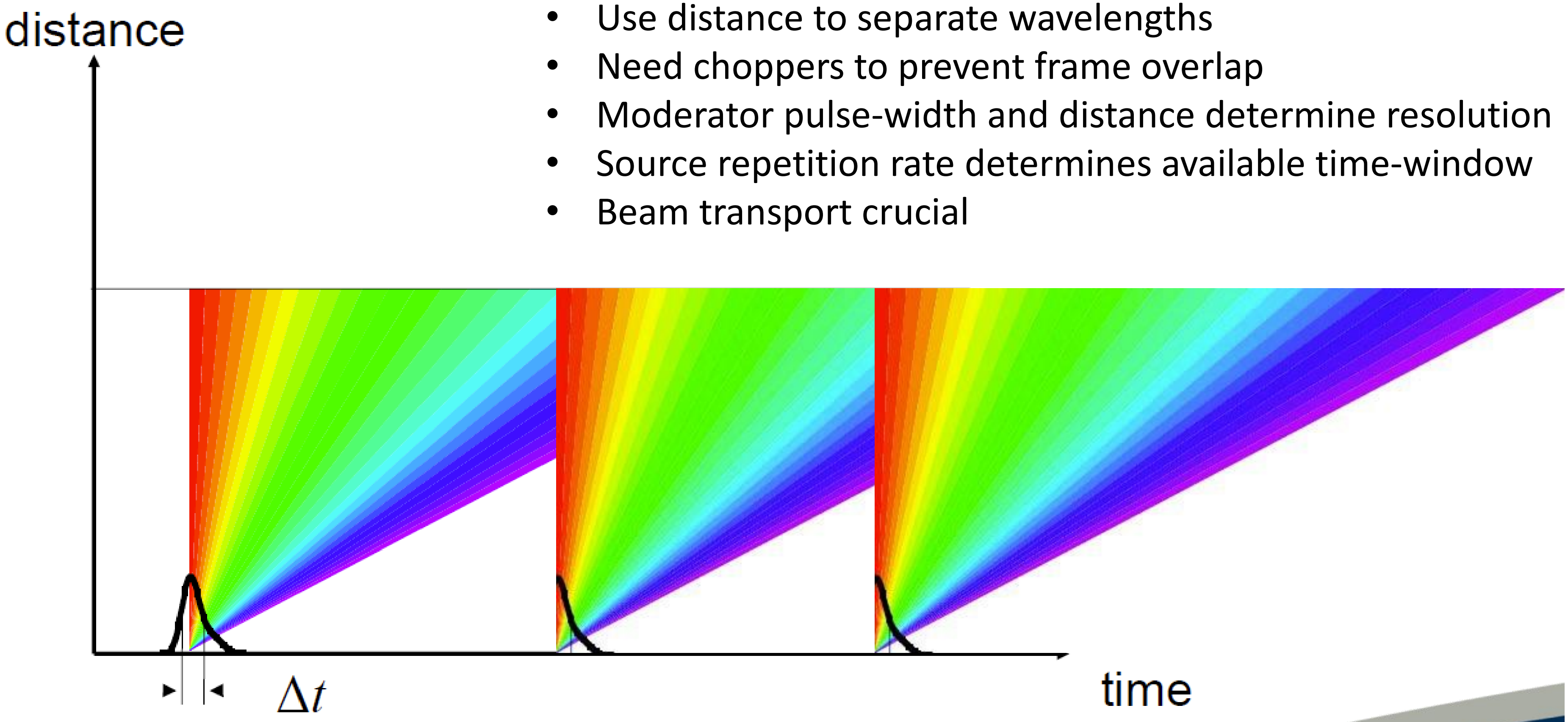
Water (ice) in VISION converted in ToF



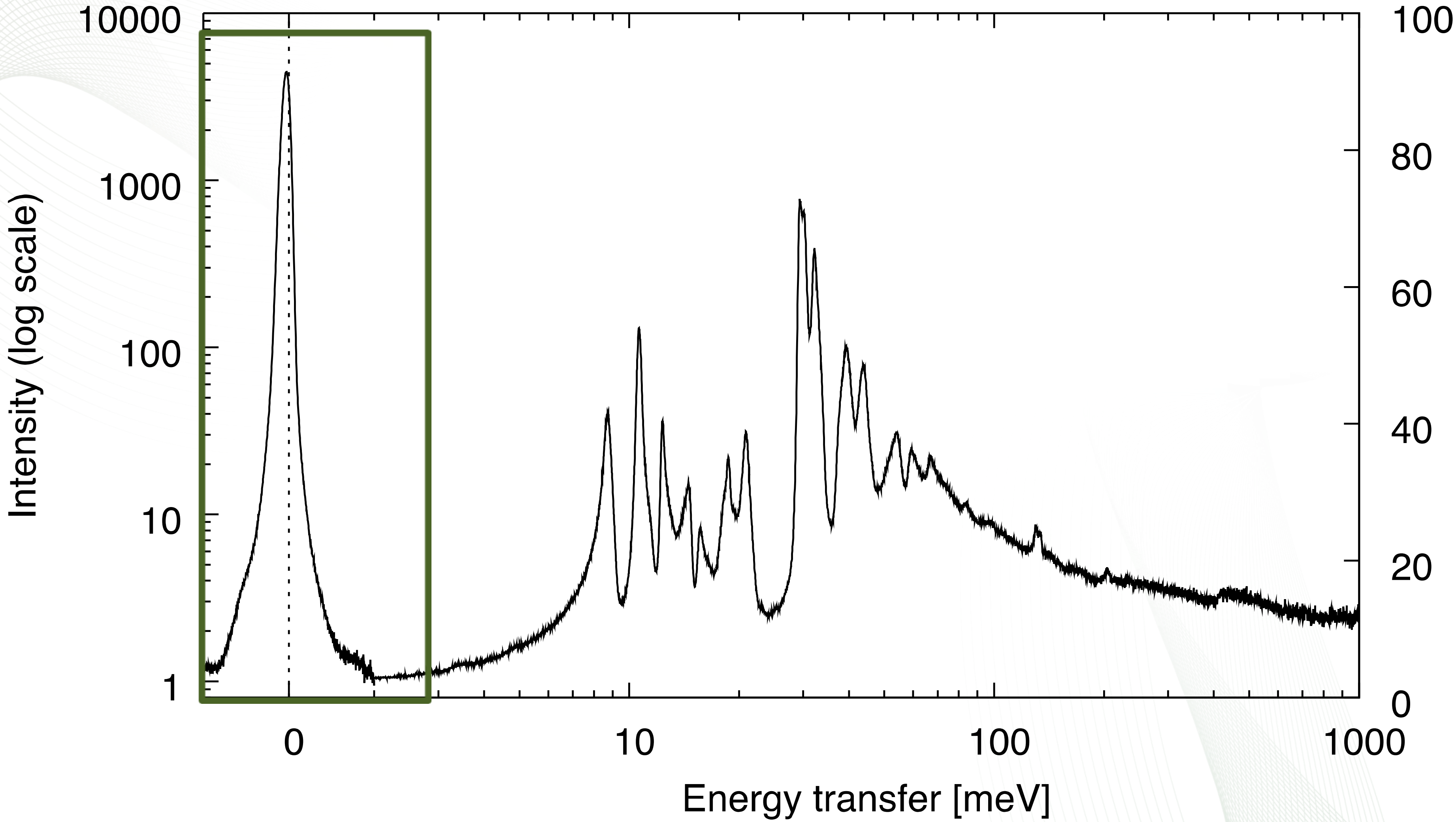
The T_0 and frame overlap chopper are setup to run the instrument at 30Hz. This decision means that the intensity of the spectrum above 3.5 meV is reduced by 50%. But the benefits completely outweigh the reduction in flux.

TOF method

- Use distance to separate wavelengths
- Need choppers to prevent frame overlap
- Moderator pulse-width and distance determine resolution
- Source repetition rate determines available time-window
- Beam transport crucial



What do I get around the elastic line?



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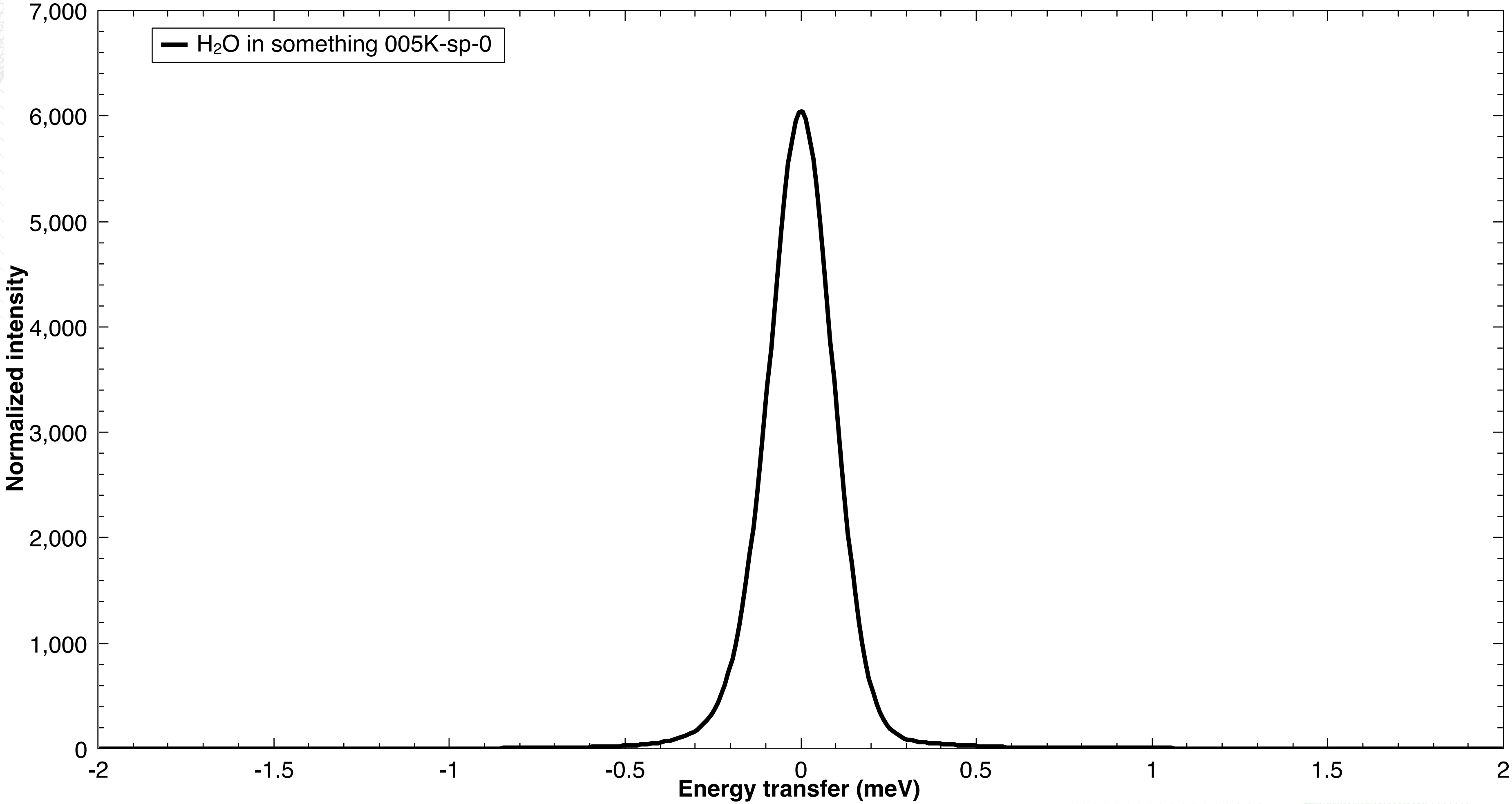
What do I get around the elastic line?



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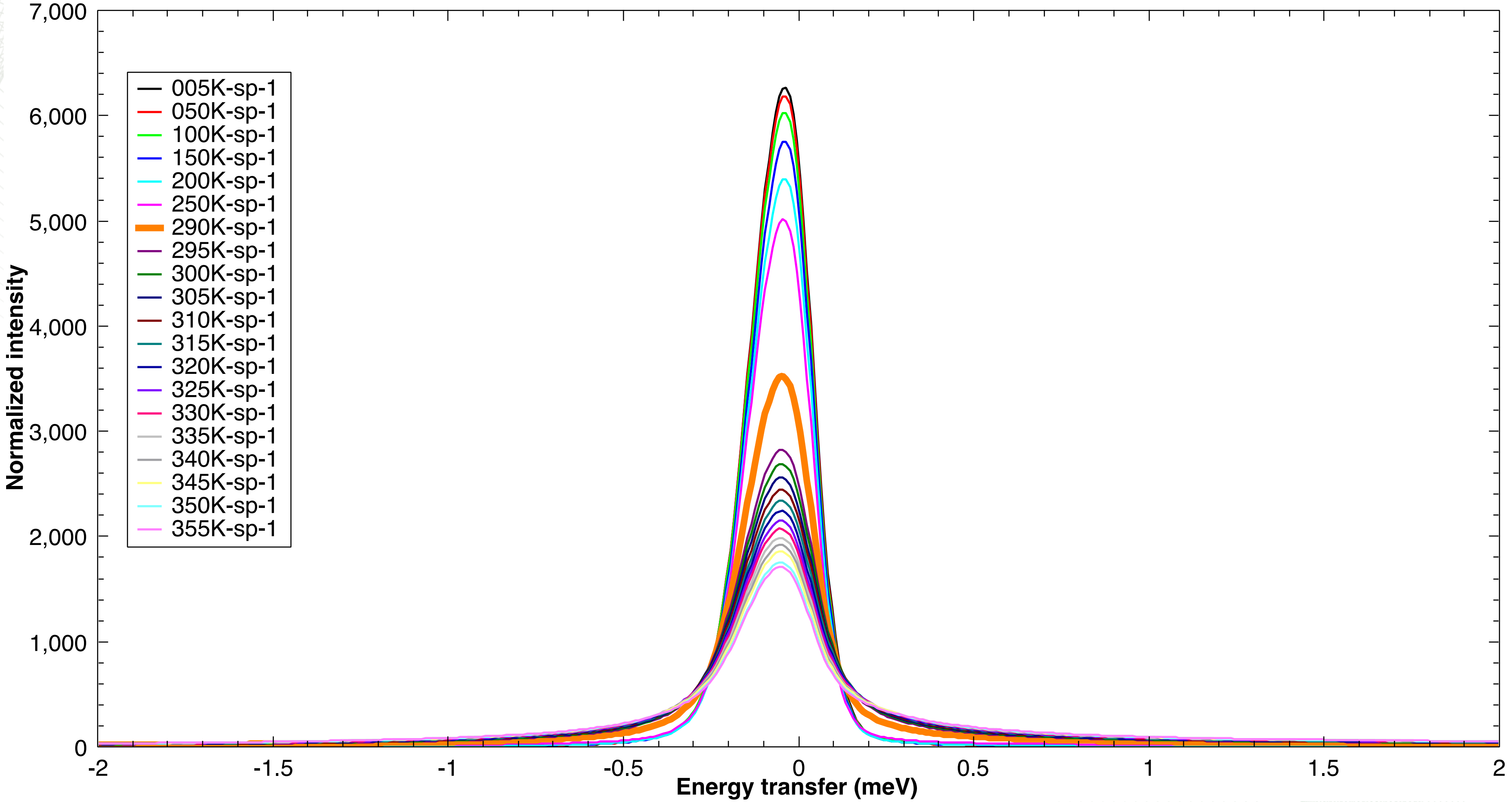
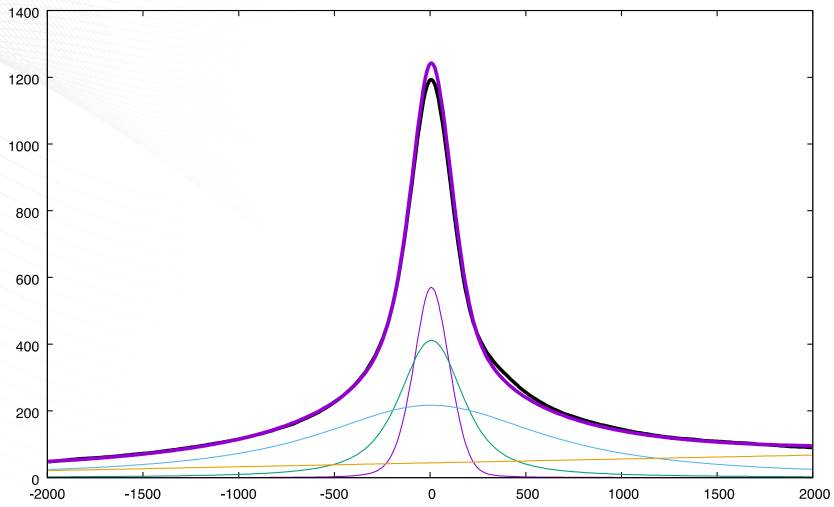
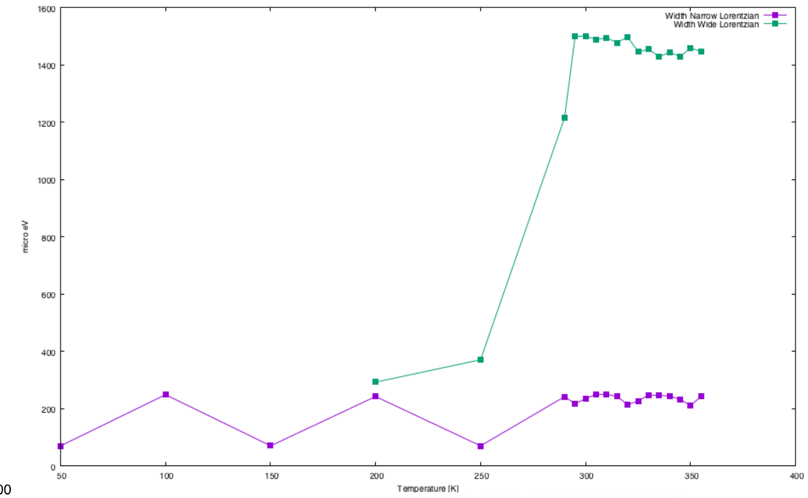
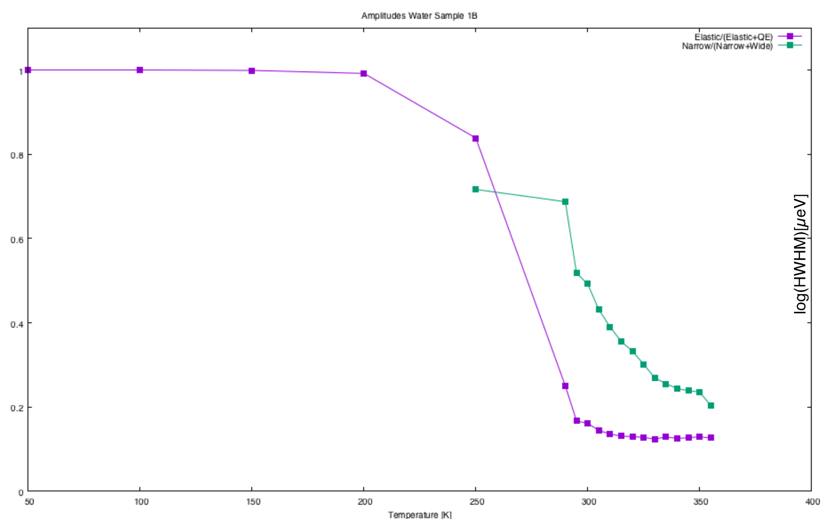
What do I get around the elastic line?

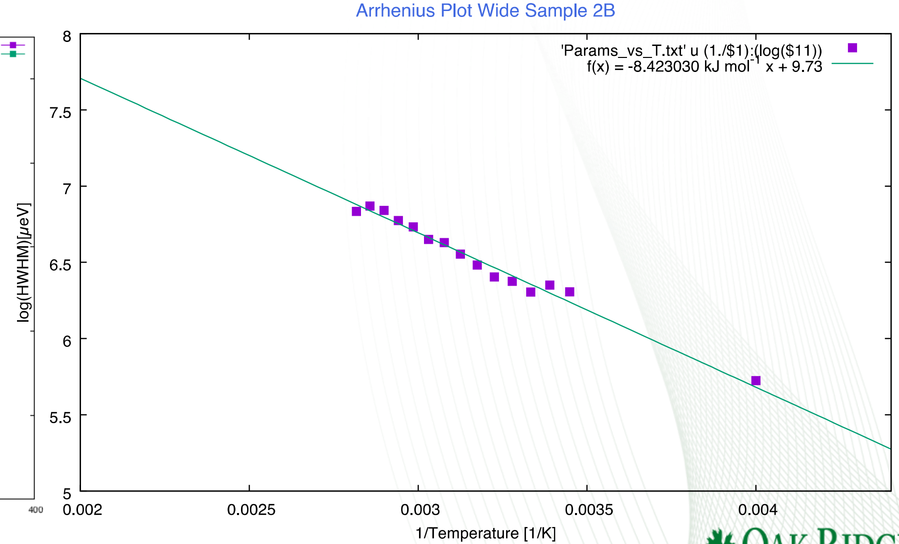


What do I get around the elastic line?

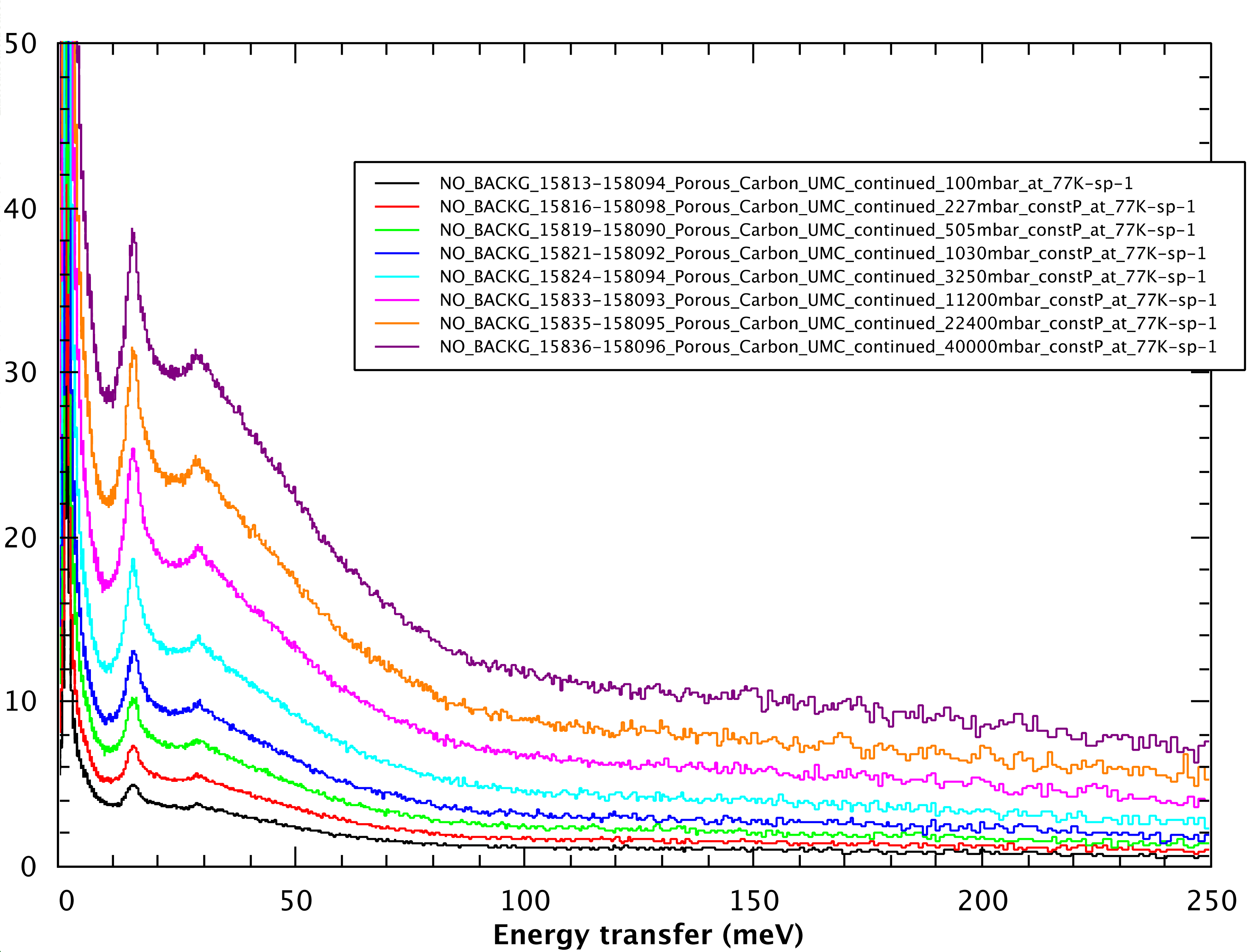




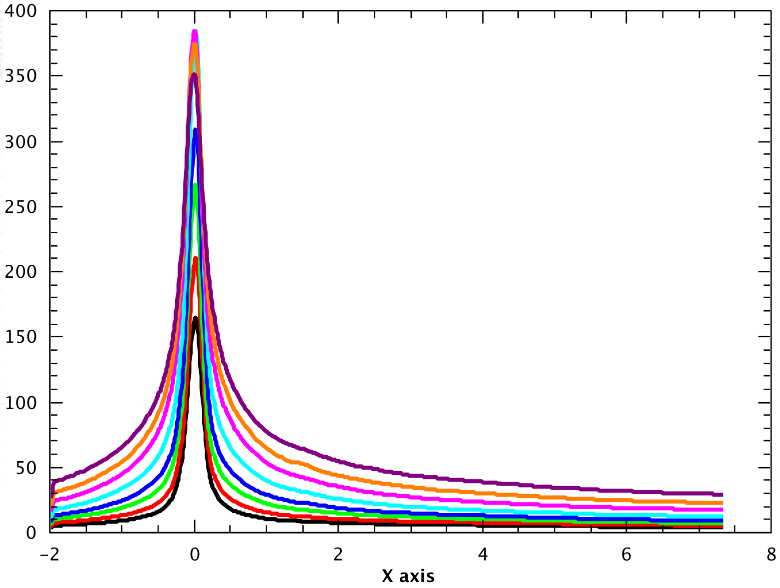




Molecular hydrogen in porous carbon

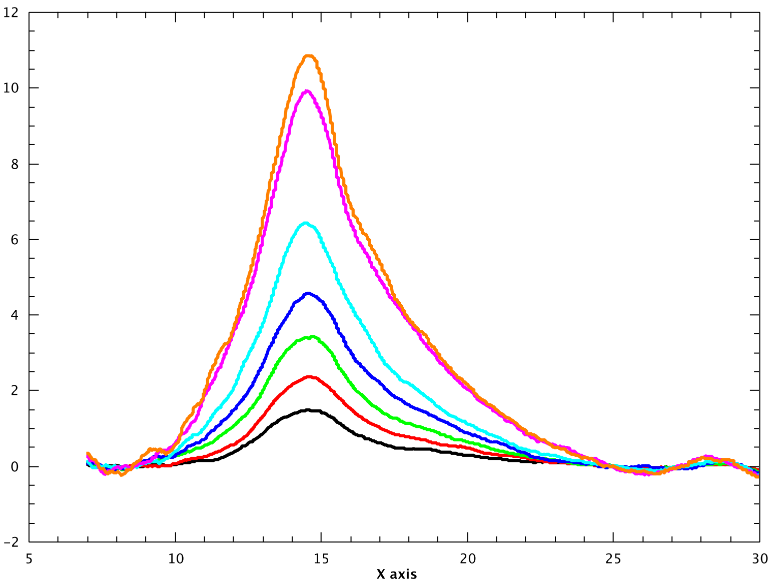


Molecular hydrogen in porous carbon



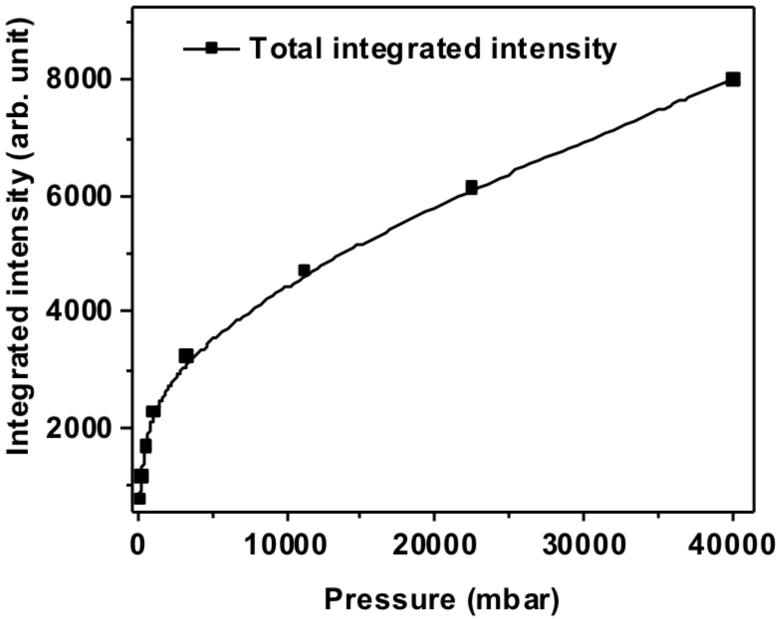
Presence of elastic line at 77K is indication of highly dense molecular hydrogen in the pores.
The broadening of the elastic line is a consequence of the enhanced mobility of the molecules as the amount of hydrogen increases in the system. Larger pores, where hydrogen is less constrained have more mobility.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

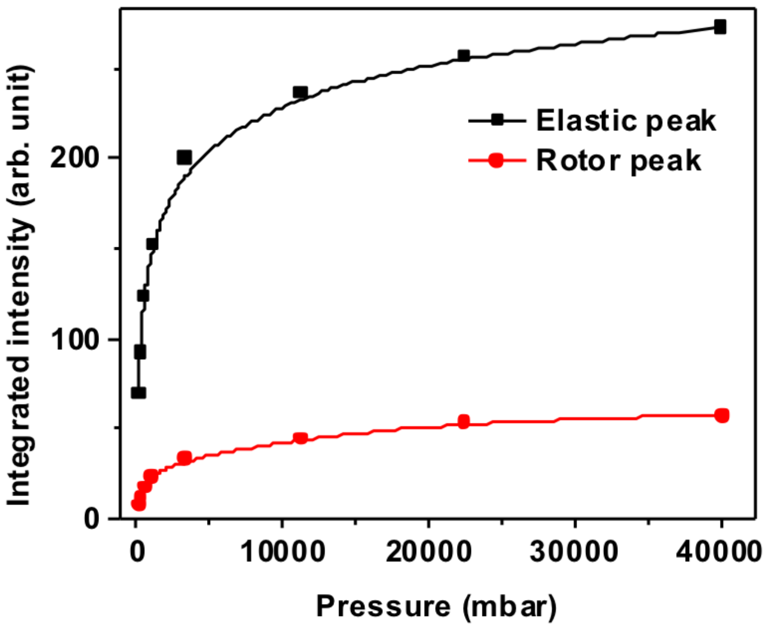
Presence of the rotor line at 77K is indication of completely immobile molecular hydrogen in the pores. In the case of pure para-hydrogen (previous figure) the line disappears when the hydrogen melts.
There is very little broadening of the rotor line, since the momentum transfer is larger that the corresponding one at the elastic line (dynamical trajectory of indirect geometry).
The load keeps increasing even at 40 bar.



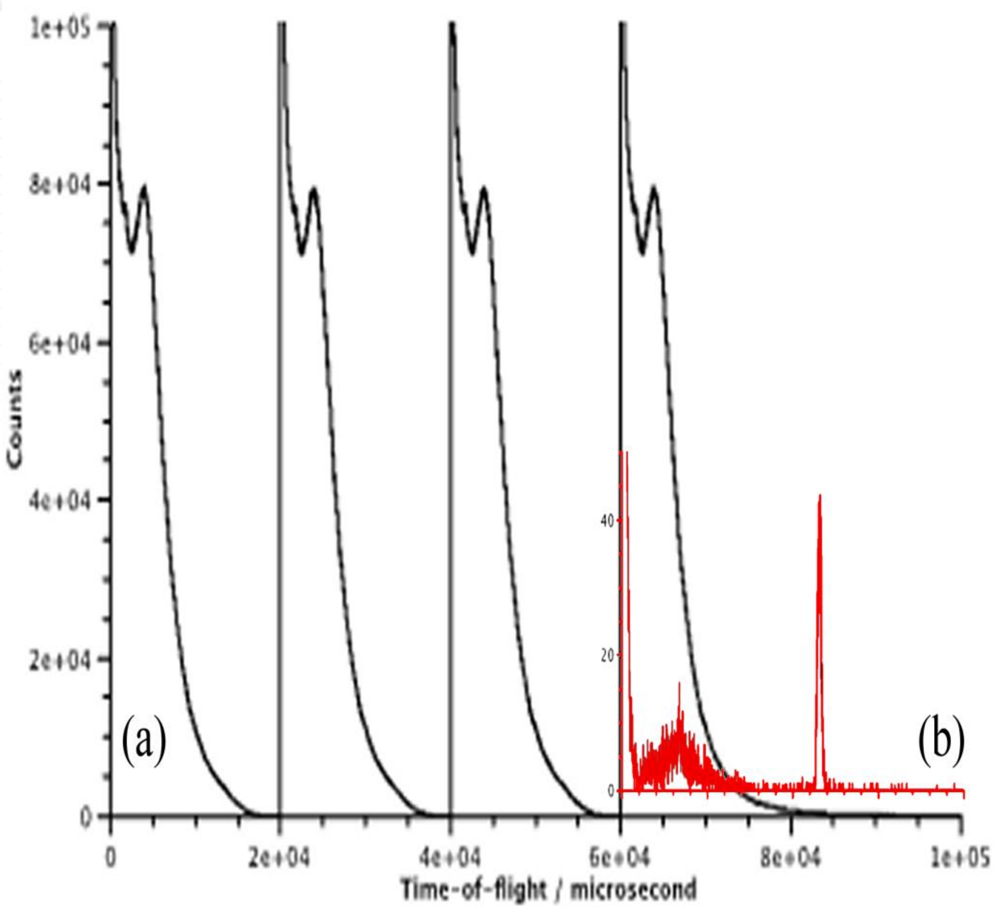
Molecular hydrogen in porous carbon

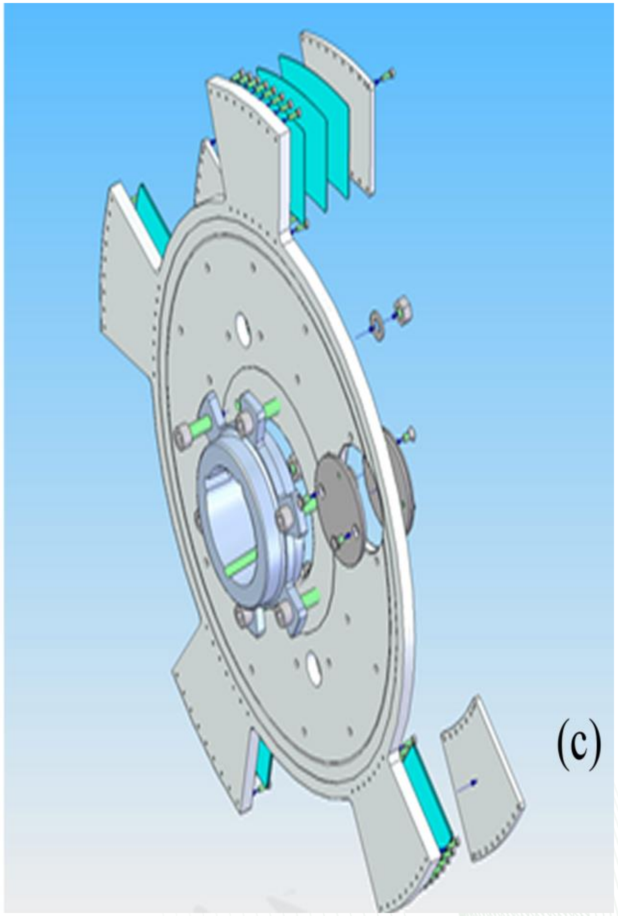
1. The total integral of the spectral intensity is proportional to the amount of hydrogen in the system (left plot)
2. The integrated area under the elastic peak is proportional to the amount of hydrogen that is in a liquid like and solid like phase (right panel)
3. The integrated area under the rotor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





TOSCA Elastic line



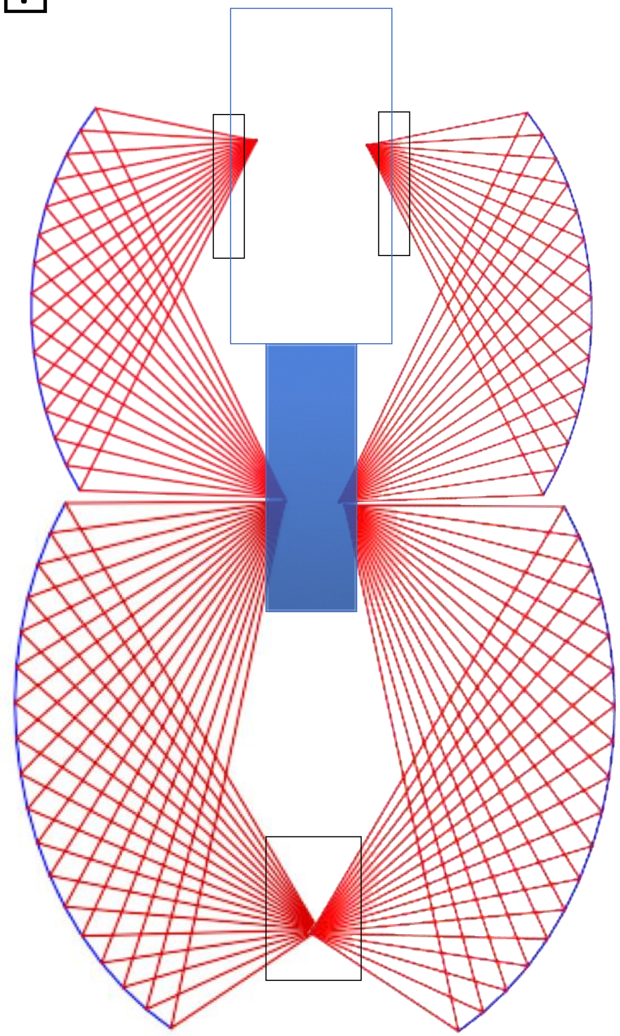


SPHIINXS

SPHerical Indirect INelastic Xtal Spectrometer (SPHIINXS)

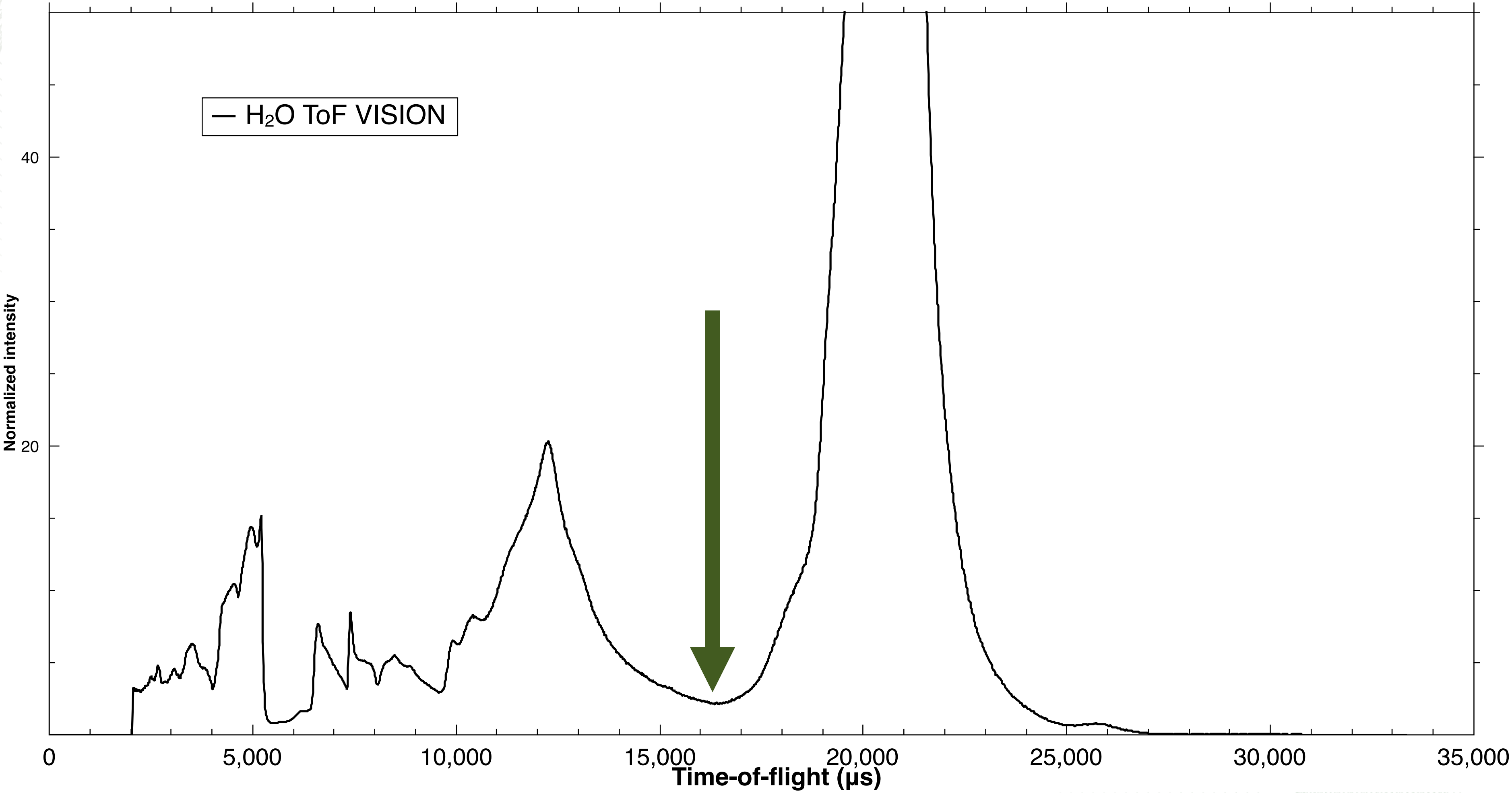
Expanding the VISION concept

- Graphite analysers on a parametric surface
- Range 3 meV to 1000 meV
- Large solid angle coverage ~10 sr
- Focussing neutron guide (1 cm^2)
- Powders and single crystal samples
- Energy resolution 1% Δω/ω and ~70 µeV at the elastic line
- Positioned at 35-40 m from moderator



Water (ice) in VISION converted in ToF

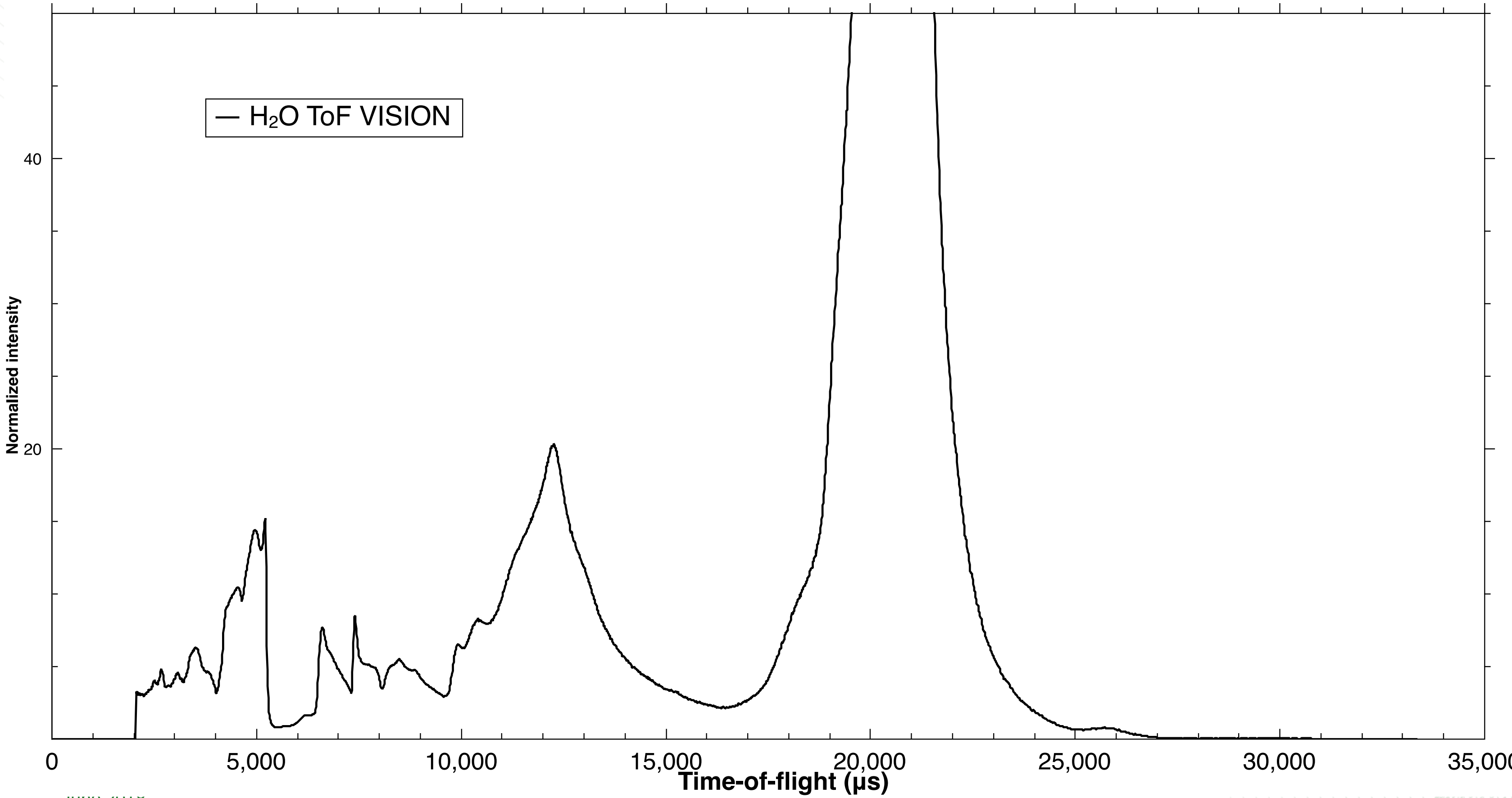
Very simple "back of an envelope" calculation



Water (ice) in VESPA converted in ToF

Very simple “back of an envelope” calculation

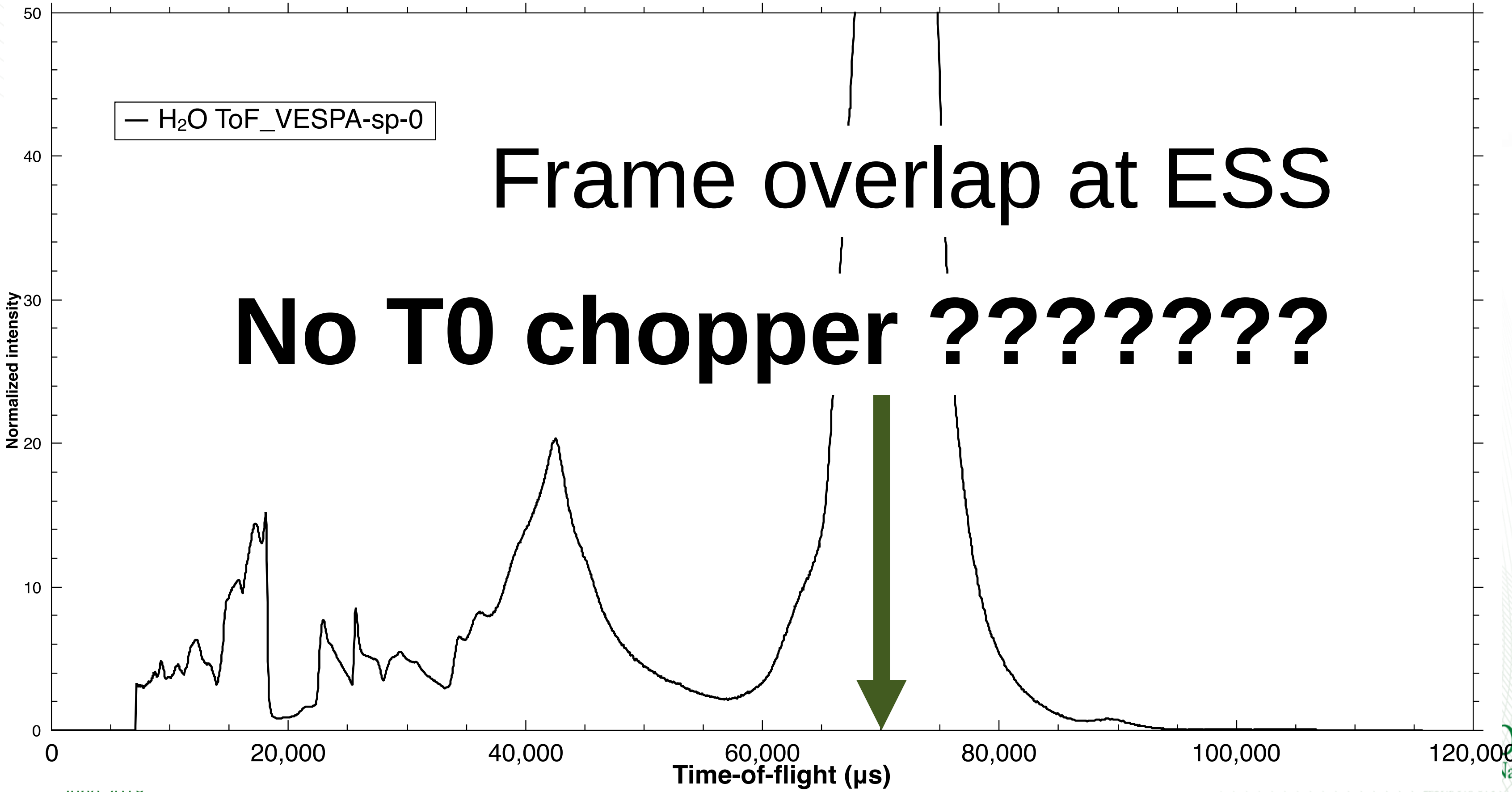
$$ToF_{VESPA} = \frac{L_{i_{VESPA}}}{L_{i_{VISION}}} ToF_{VISION}$$



Water (ice) in VESPA converted in ToF

Very simple "back of an envelope" calculation

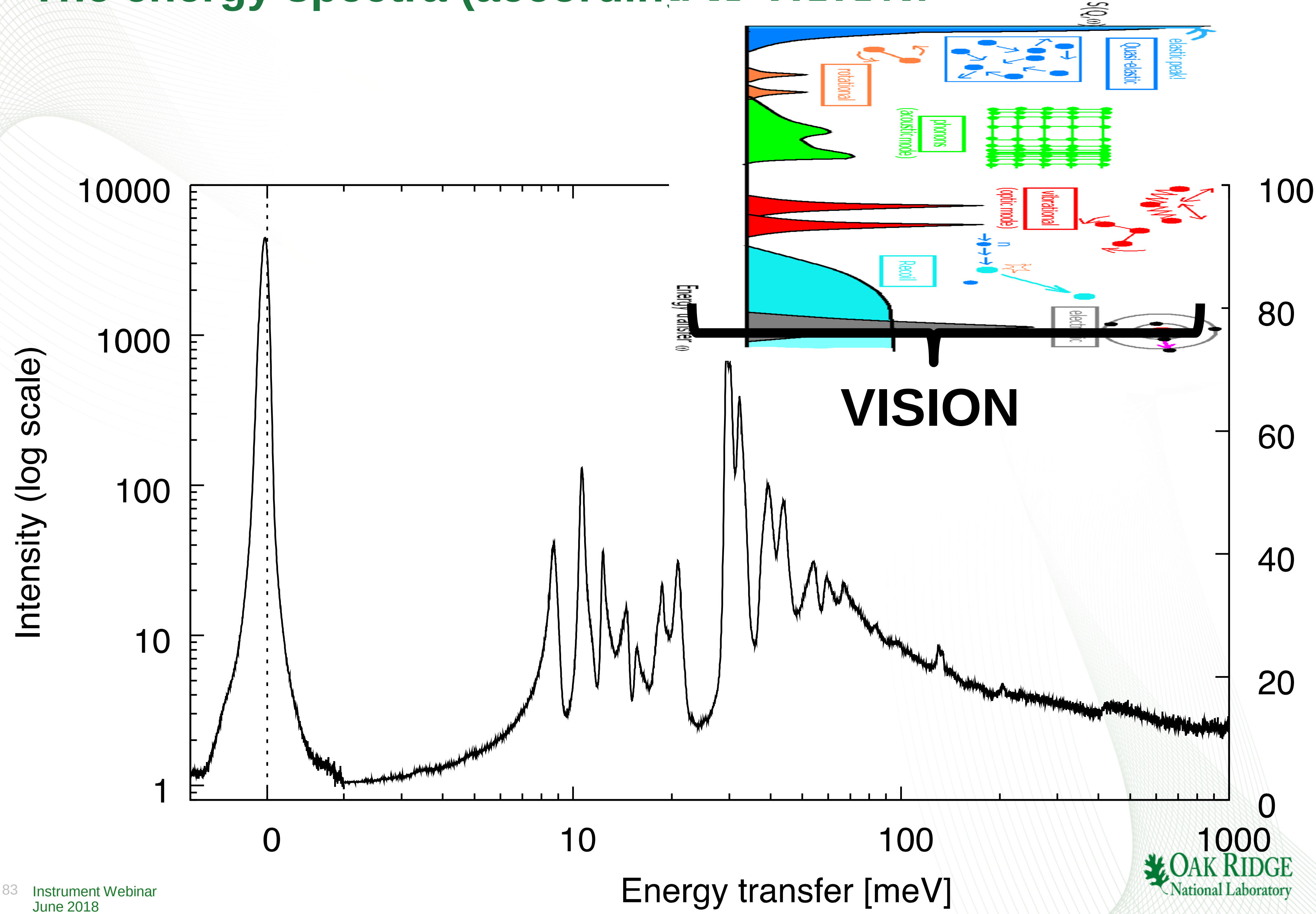
$$ToF_{VESPA} = \frac{L_{i_{VESPA}}}{L_{i_{VISION}}} ToF_{VISION}$$



Frame overlap at ESS

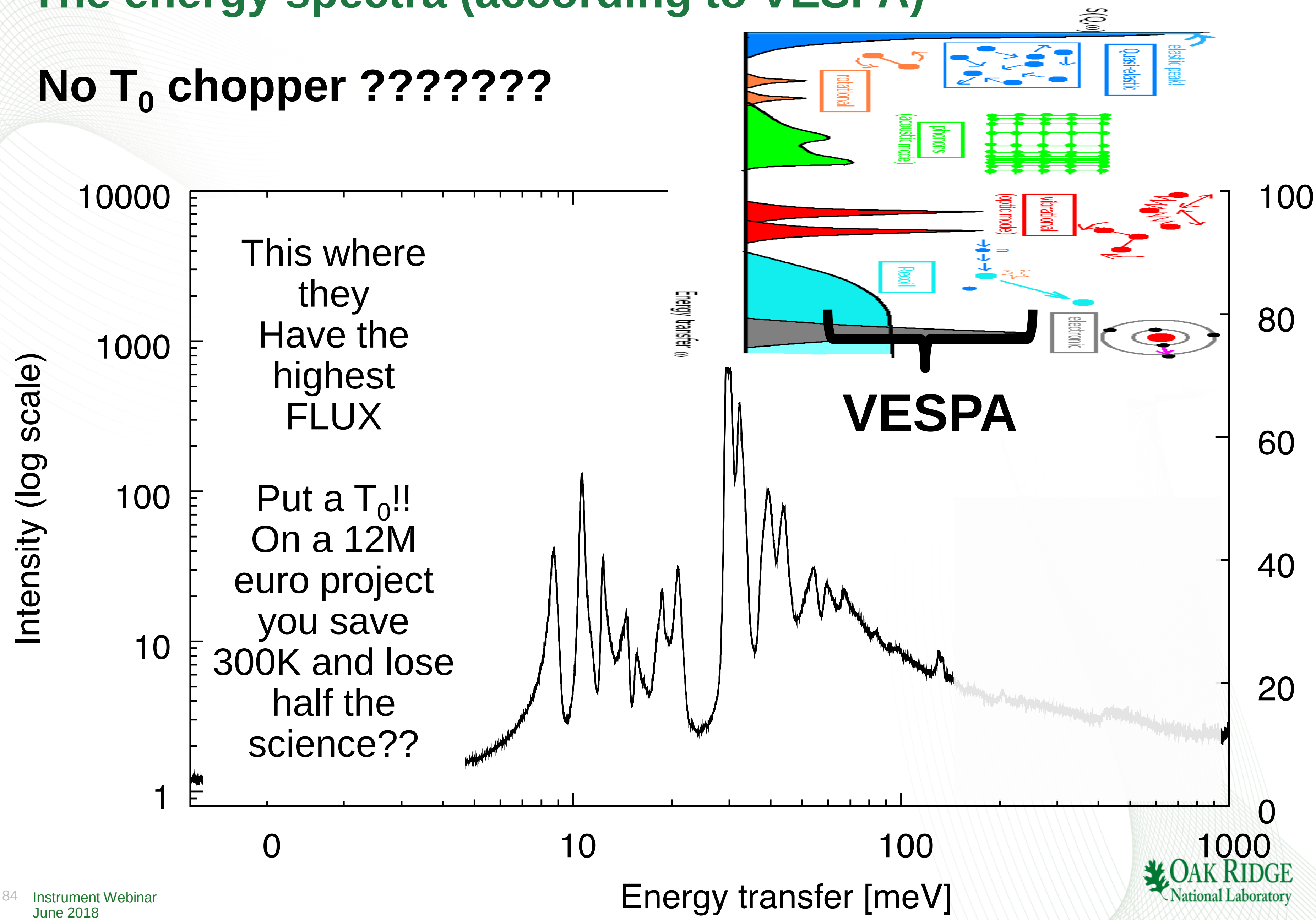
No T0 chopper ????????

The energy spectra (according to VISION)



The energy spectra (according to VESPA)

No T_0 chopper ???????



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VESPA vs VISION

NO T_0 Chopper

				...ar ...ution
VI...				✓
VE...	✗	✓	✗	✓

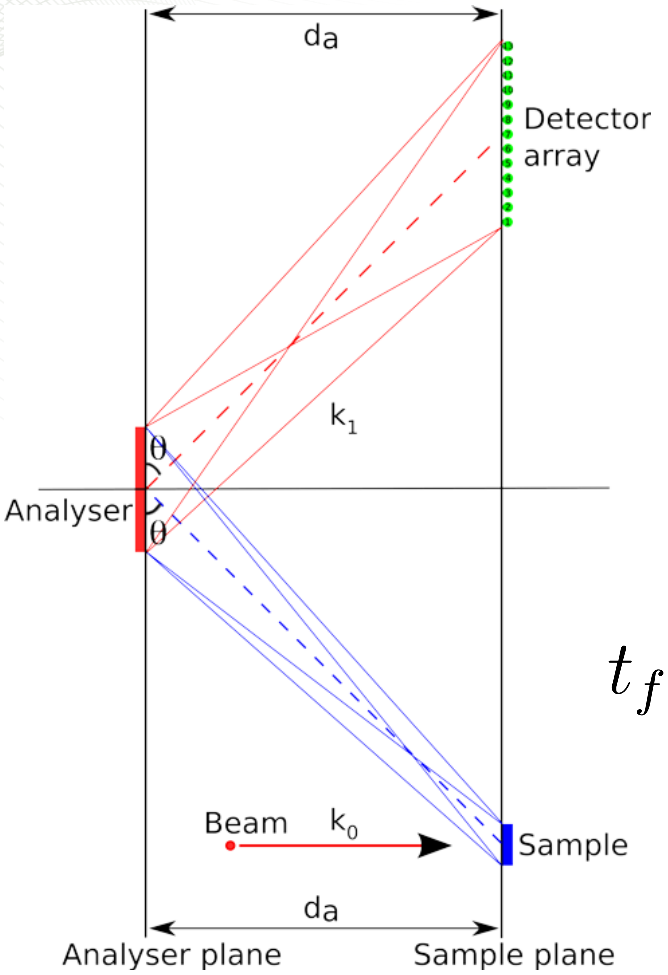
VISION Kills VESPA
Does it makes any sense at all to build VESPA?

WITH T_0 Chopper

				✓
	✗	✓	✓	✓

VESPA beats VISION in the QENS Regime
VESPA has world class capability

Secondary analyzer

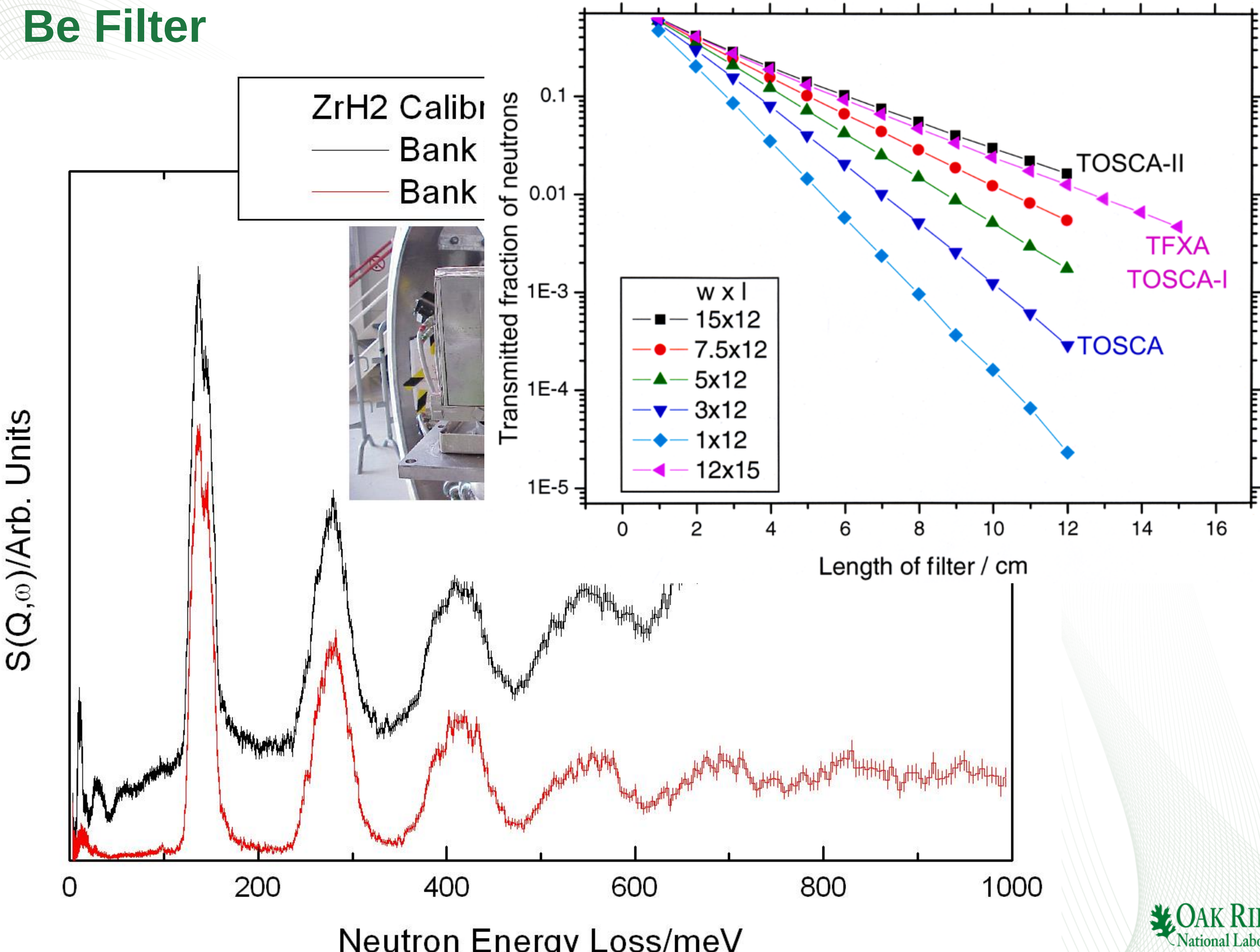


$$\lambda_f = 2d_{PG} sin\theta$$

$$\frac{h}{m_n}\frac{t_f}{d_f} = 2d_{PG}\frac{2d_a}{d_f}$$

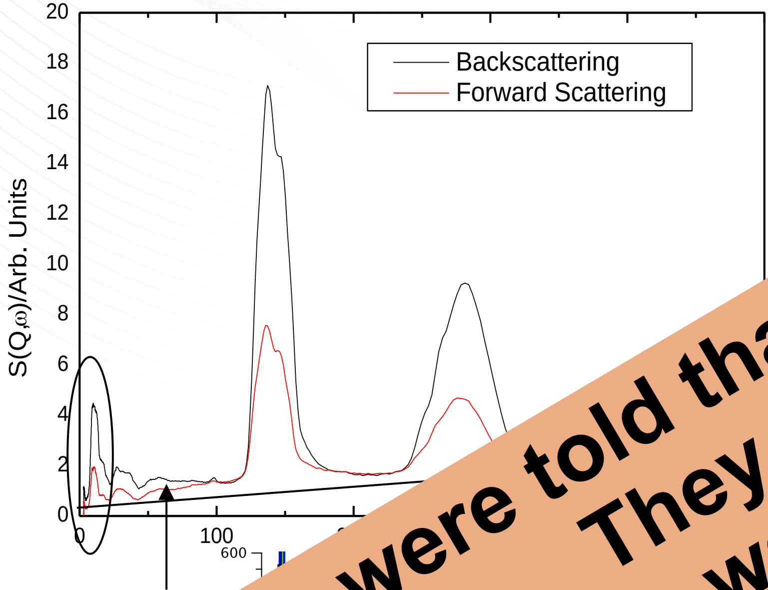
$$t_f = 4d_{PG}d_a\left(\frac{m_n}{h}\right) = constant$$

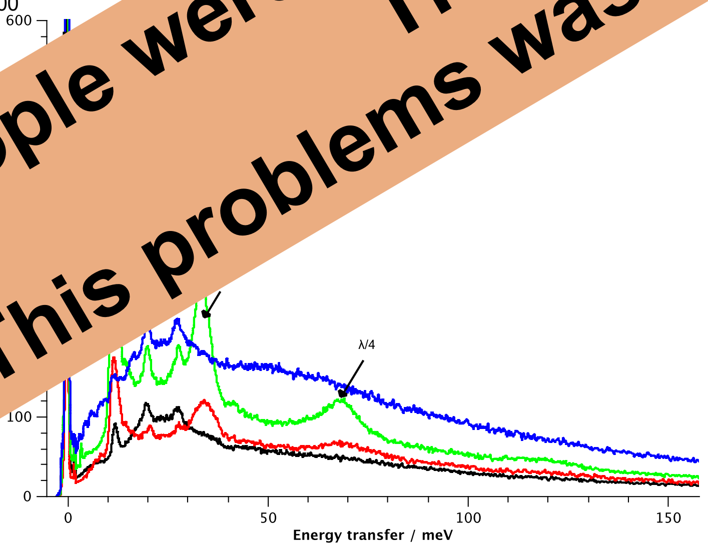
Be Filter

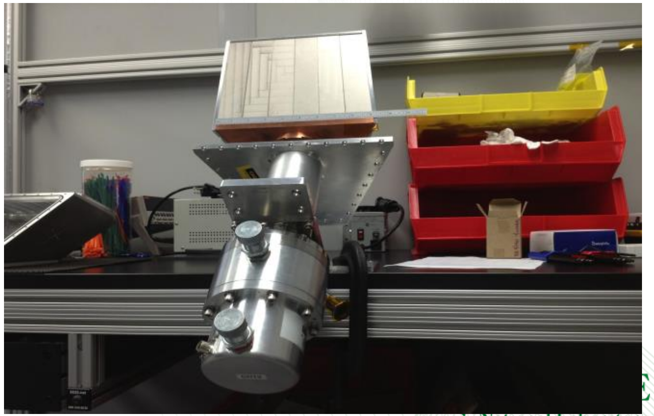

ZrH2 Calibr
Bank
Bank
S(Q,ω)/Arb. Units
0
200
400
600
800
1000
Neutron Energy Loss/meV
Transmitted fraction of neutrons
0.1
0.01
1E-3
1E-4
1E-5
0
2
4
6
8
10
12
14
16
Length of filter / cm
w x l
15x12
7.5x12
5x12
3x12
1x12
12x15
TOSCA-II
TFXA
TOSCA-I
TOSCA

REPEATING MISTAKES! TOSCA & VISION

- We have an non existing feature at low energies and a big increase of the signal at high energy transfers.
- The effect is most noticeable in forward scattering







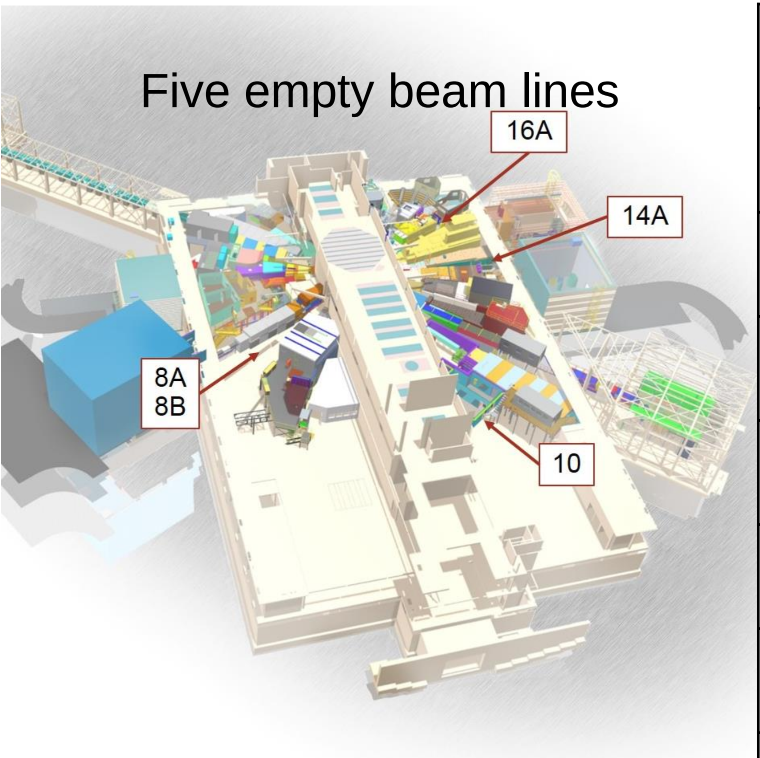
People were told that something was wrong

They didn't listen

This problems was relatively easy to solve!


STEVEN SPIELBERG PRESENTS
BACK TO THE FUTURE
PG
A ROBERT ZEMECKIS FILM
OUTATIME

High priority instruments at the SNS First Target Station will leverage its high wavelength resolution strength

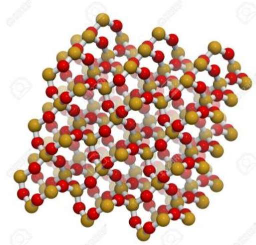


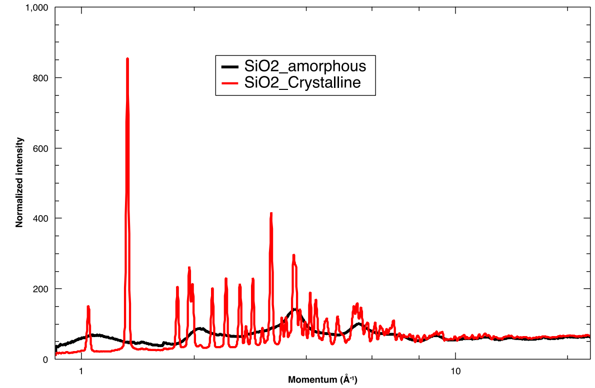
HIGGS	Inverse geometry spectrometer	BL-8A dc-p H_2O
MICRON	Compact, texture, special purpose diffractometer	
DISCOVER	Medium resolution/flux diffractometer	BL-8B dc-p H_2O
VENUS	Time-of-flight neutron imaging station	BL-10 dc-p para-H_2
INVENT	Concept development station	BL-14A c para-H_2
SANS/GI-SANS	SANS and/or GI-SANS	
BeFAST	Beryllium filter spectrometer	BL-16A dc-p H_2O
HiResPD	High Resolution Powder Diffractometer	Needs dc-p para-H_2 100 m flight path

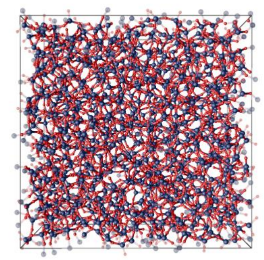
dc-p: decoupled, poisoned
c: coupled
H_2O: thermal neutrons
para-H_2: cold neutrons

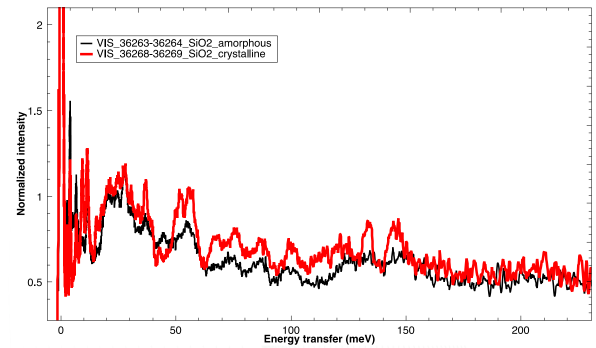
VISION: Inelastic, Diffraction and QENS

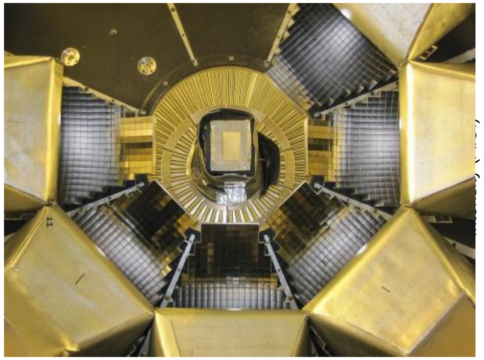
- Chemistry Oriented INS spectrometer
- White incident beam, fixed final energy (indirect geometry)
- High flux (~5x10^7 neutrons/cm^2/s) and double-focusing
- Broadband (-2 to 1000 meV at 30Hz, 5 to 500 meV at 60 Hz)
- Constant ∆E/E throughout the spectrum (~1.5%)
- Elastic line HMFW ~120 µeV
- Backward and 90° diffraction banks
- 4000 x its predecessor

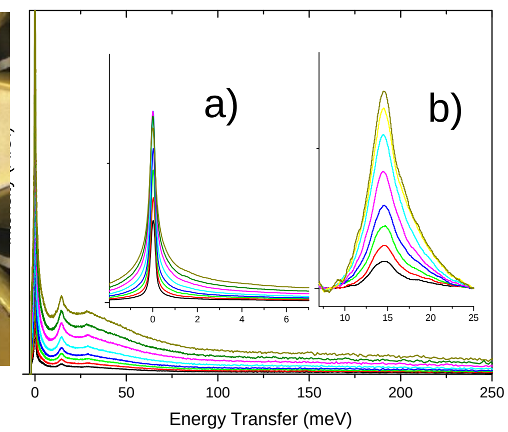






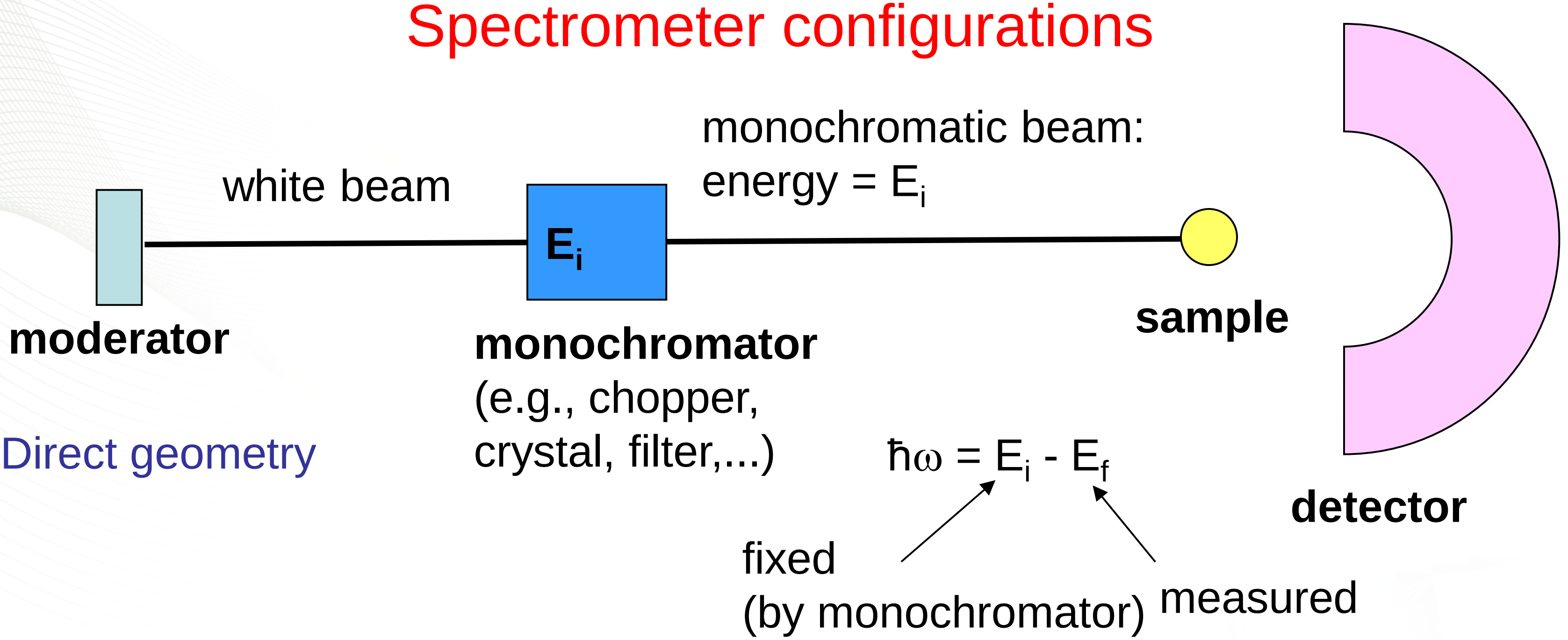




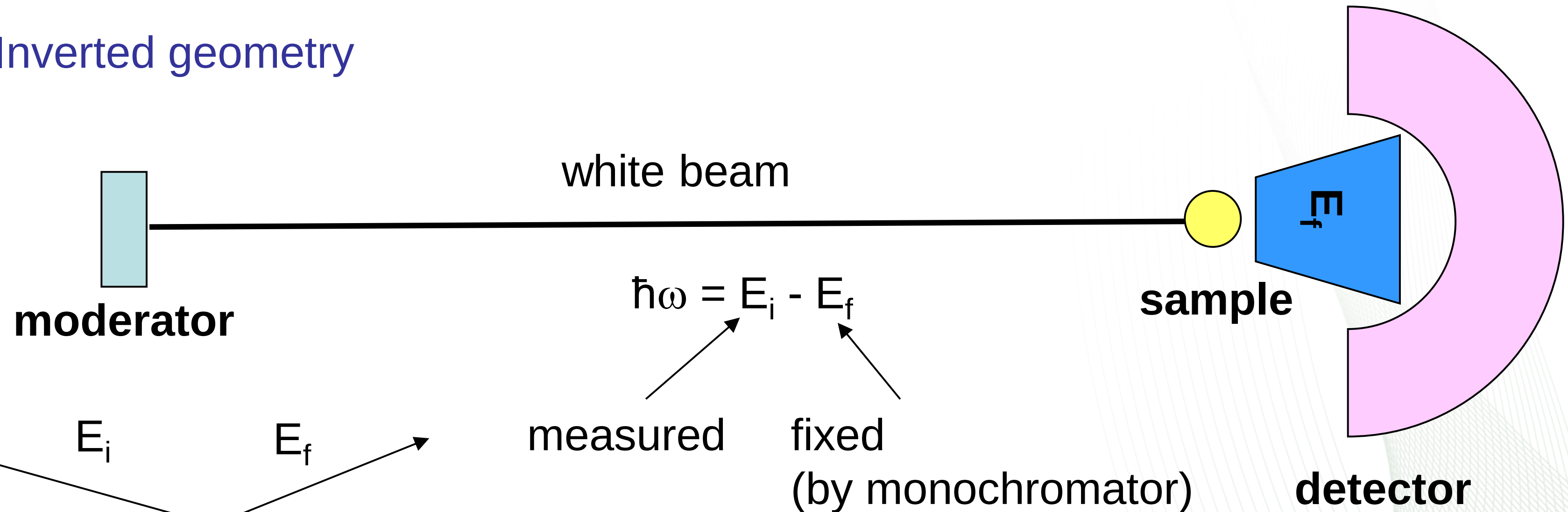


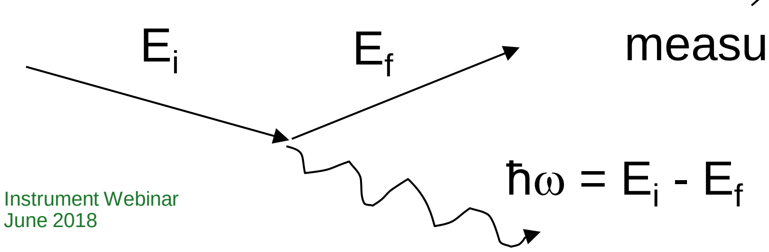
Spectrometer configurations

Direct geometry

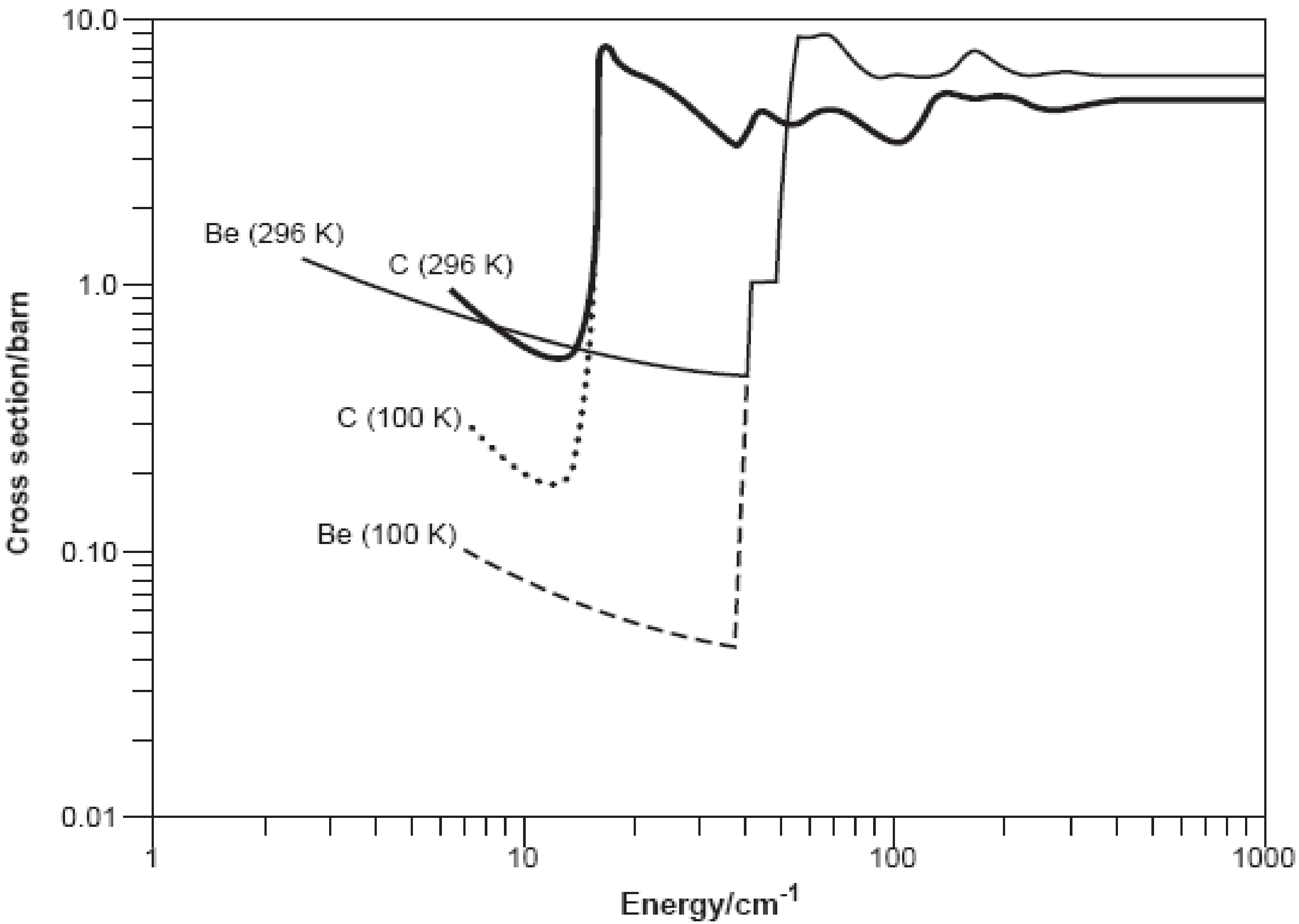


Inverted geometry





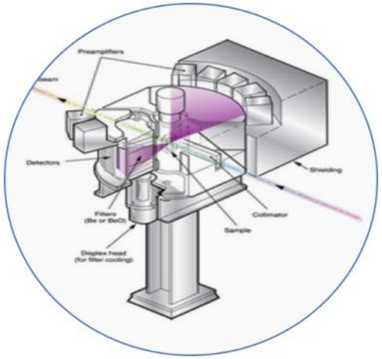
Be filter

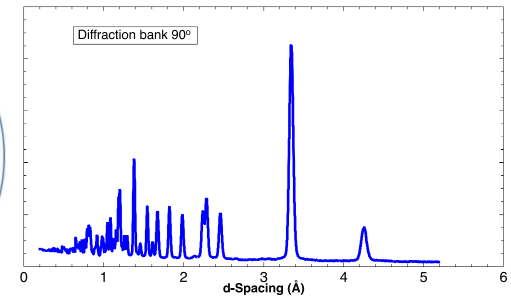


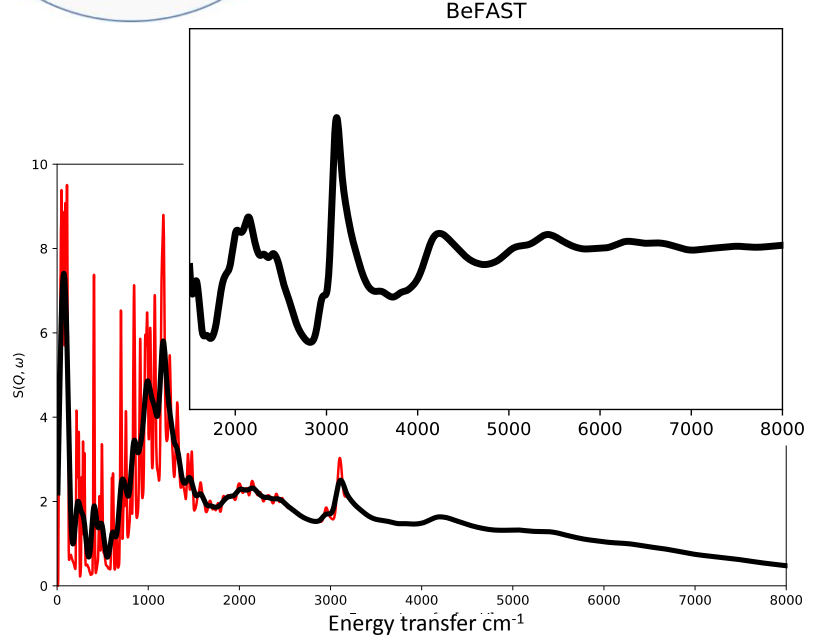
10.0
1.0
0.10
0.01
Cross section/barn
Be (296 K)
C (296 K)
C (100 K)
Be (100 K)
1
10
100
1000
Energy/cm-1

BeFAST – Beryllium Filter Analyzer Spectrometer

- NScD Point-of-Contact: Timmy Ramirez-Cuesta
- Community Point-of-Contact: TBD
- Science Themes: catalysis, “real-world” systems
- Day 1 capabilities: BeFAST will measure in situ reactions by tracking CH or OH stretching and bond breaking (data collection as fast as 10 min)
  - Measure inelastic neutron scattering to 8000 cm^{-1}, overlapping Raman and IR spectroscopy
  - Complements the VISION spectrometer with over 3-order of magnitude signal gain in the range 3000 to 8000 cm^{-1} with slightly lower resolution
  - Will have a 90º diffraction bank
  - Large solid angle coverage
  - Very compact, probably only instrument that will fit in BL-16A
  - Inexpensive and simple, can be built in a short time
  - Low m value guide (for half of the length), and needs one frame-overlap chopper







Neutron inelastic spectra from triphenylmethane. Red is calculated for VISION, black for BeFAST BeFAST increased flux above 1800cm^{-1}

Thank you!