

IRSN

INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

Measurement of double differential cross section of light water at high temperature and pressure

Supervisors

Luiz LEAL
Wim HAECK

Vaibhav JAISWAL
vaibhav.jaiswal@irsn.fr
PSN-EXP/SNC/LNR,
Fontenay-aux-Roses, France

- **Objective of my PhD Thesis**
- **Experiment: ILL**
- **Data Analysis & Preliminary Results**
- **Future work & Remarks**

Motivation?

- IRSN is working on the improvement of $S(\alpha, \beta)$ thermal scattering kernel of light water at high temperature and pressure (at normal operating conditions of nuclear power reactors) for criticality safety studies and reactor physics applications.

What is $S(\alpha, \beta)$?

Nuclear cross section data libraries use derived thermal neutron scattering, represented by $S(\alpha, \beta)$ scattering kernel where α and β stand for the unit-less momentum and energy transfers respectively.

- The double differential thermal scattering cross section for neutrons with incident energy E , secondary energy E' , Scattering angle Ω , bound scattering cross section σ_b and at temperature T can be written as:

$$\frac{\partial^2 \sigma_T}{\partial \Omega \partial E'} (E \rightarrow E', \Omega) = \frac{\sigma_b}{4\pi kT} \sqrt{\frac{E'}{E}} e^{-\frac{\beta}{2}} S(\alpha, \beta)$$

Scattering Kernel

What do we have..?

- Thermal scattering data available to users from the evaluated data libraries come from essentially two different models, prepared by:
 - General Atomics group (GA model) in the 1960s
 - Mattes and Keinert at IKE Stuttgart (IKE model) in the 1980s and updated in the mid 2000s

Both the above models are based on...

- **Experimental data measured in the 1960s** with a low accuracy due to the limitations of the experimental and computational capabilities at that time.
- Numerous approximations used for the evaluation of the thermal scattering cross section.

New experimental data for the verification of generated thermal neutron scattering data are very scarce.

What we want...?

- **Our goal:** Improve the thermal scattering cross section data of light water at **operating conditions of nuclear power reactors**.
- Study the uncertainties associated with it, namely the systematic and statistical uncertainties.

HOW? ↓

Study the structure and dynamics of water incorporating both the **theory and experiments**.

- On the theoretical side: Review the existing models and use **Molecular dynamics simulations**.
- On the experimental side: **Perform new experiments** to generate thermal scattering kernel for light water.

We are here !!!

Objective of my PhD thesis

$S(\alpha, \beta)$ formalism

- In the thermal scattering kernel $S(\alpha, \beta)$

Momentum transfer is represented by:

$$\alpha = \frac{E' + E - 2\sqrt{EE'} \cos\theta}{AkT} = \frac{q^2 \hbar^2}{2MkT}$$

Energy transfer is represented by:

$$\beta = \frac{E' - E}{kT} = \frac{-\hbar\omega}{kT}$$

$S(\alpha, \beta)$

$S(\vec{q}, \omega)$

- In the thermal scattering function $S(\vec{q}, \omega)$

Wave Vector transfer is represented by:

$$\vec{q} = \vec{k}_f - \vec{k}_i$$

Energy transfer is represented by:

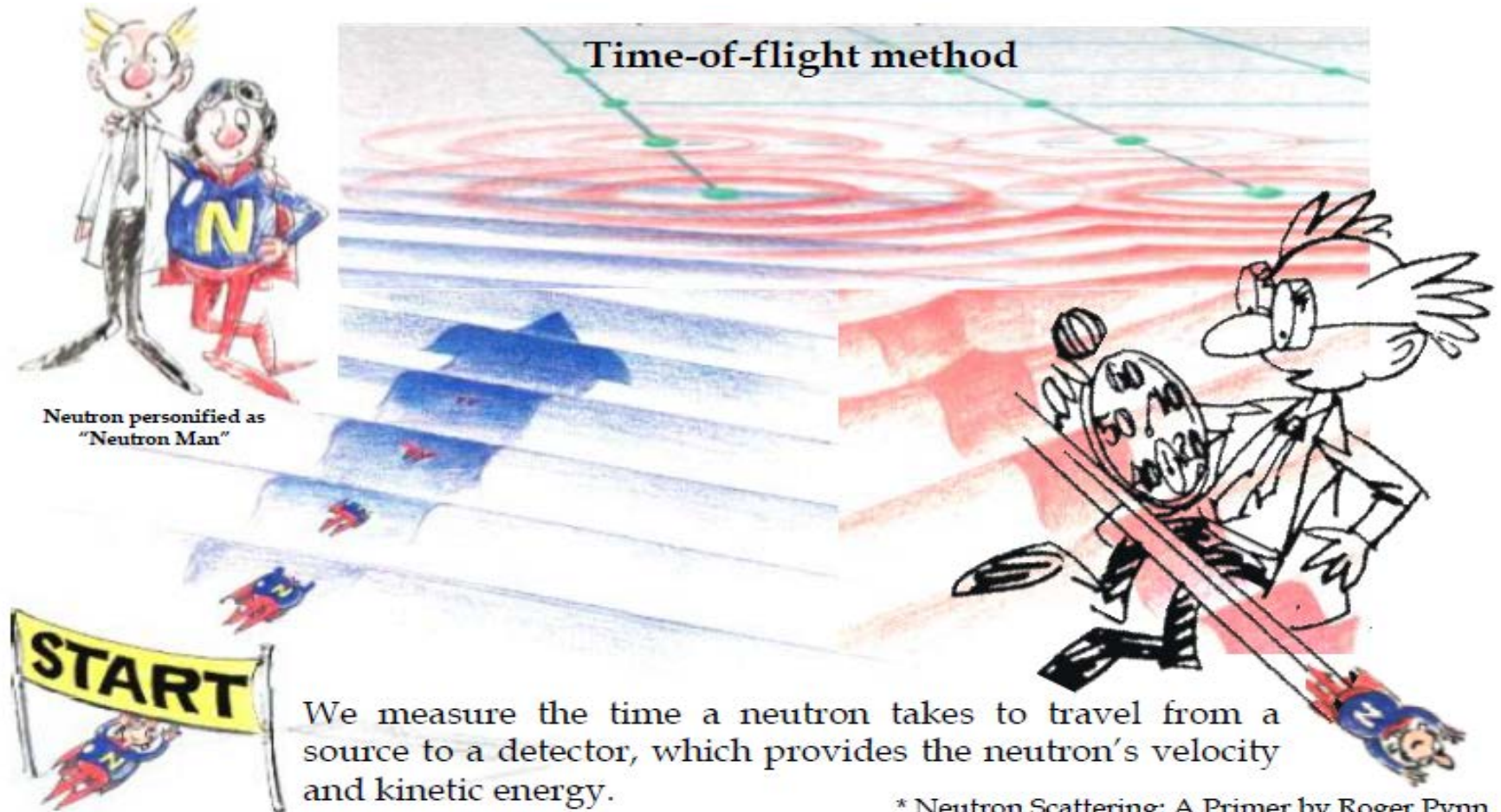
$$\omega = \frac{E_f - E_i}{\hbar}$$

← *Experiments*

$S(\alpha, \beta)$ is analogous to the scattering function $S(\vec{q}, \omega)$

Objective of my PhD thesis

$S(\vec{q}, \omega)$ which measures the dynamic structure factor, can be directly measured by performing **time-of-flight inelastic thermal neutron scattering experiments** with light water at desired temperature and pressure, in order to generate $S(\alpha, \beta)$.



* Neutron Scattering: A Primer by Roger Pynn

- Two consecutive inelastic thermal neutron scattering experiments with light water have been carried out at the Institut Laue-Langevin (ILL, Grenoble) in Dec. 2015, using the IN4c and IN6 high resolution **time-of-flight** spectrometers.
- Improved $S(\vec{q}, \omega)$ and neutron scattering cross section of light water **at ambient temperature and pressure** has been successfully carried out at ILL in 2010.
- Extended the previous measurements beyond room temperature and pressure to more realistic operating conditions in connection to nuclear power reactors.
- Thus the accuracy of the thermal scattering cross section data could be enhanced by direct introduction of the measured dynamic structure factors $S(\vec{q}, \omega)$.

IN4c:

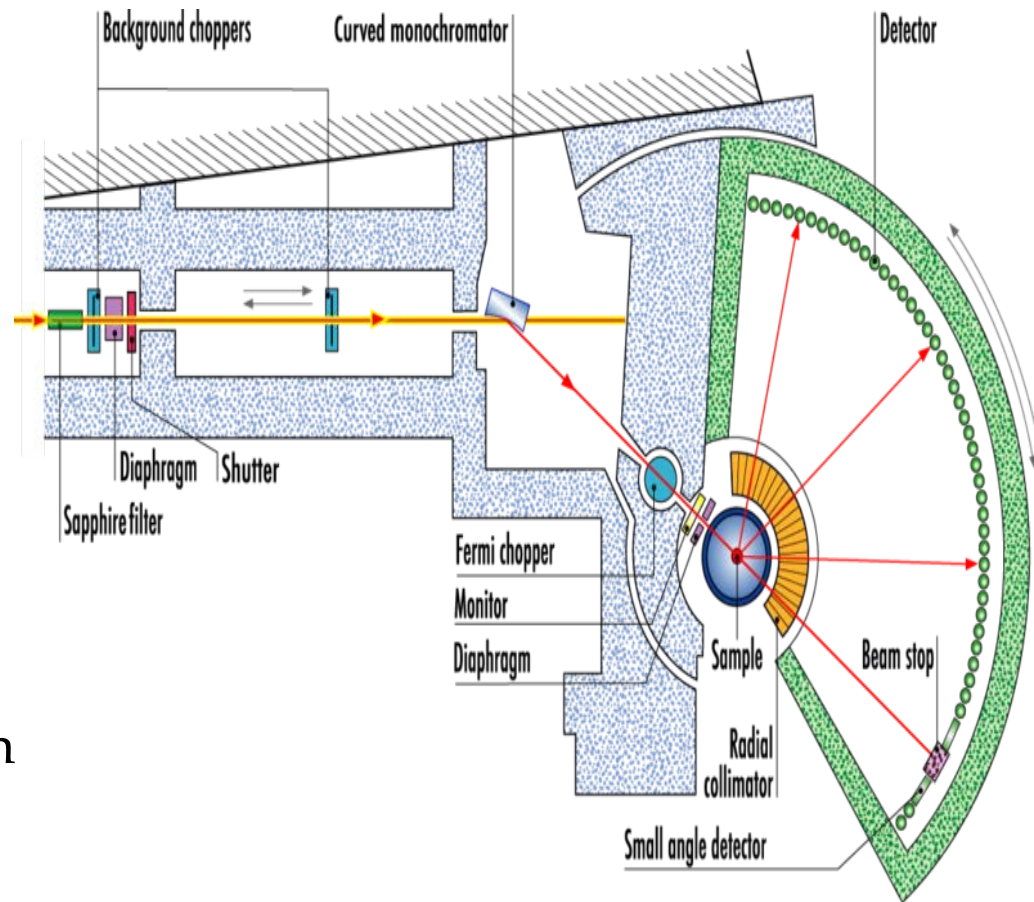


IN4c Experimental facility

■ Incident Neutron Wavelength

$\lambda : 2.4\text{\AA}$

■ Corresponding incident neutron energy: 14.20 meV



Schematic representation of the IN4c

IN stands for **In**elastic neutron scattering.

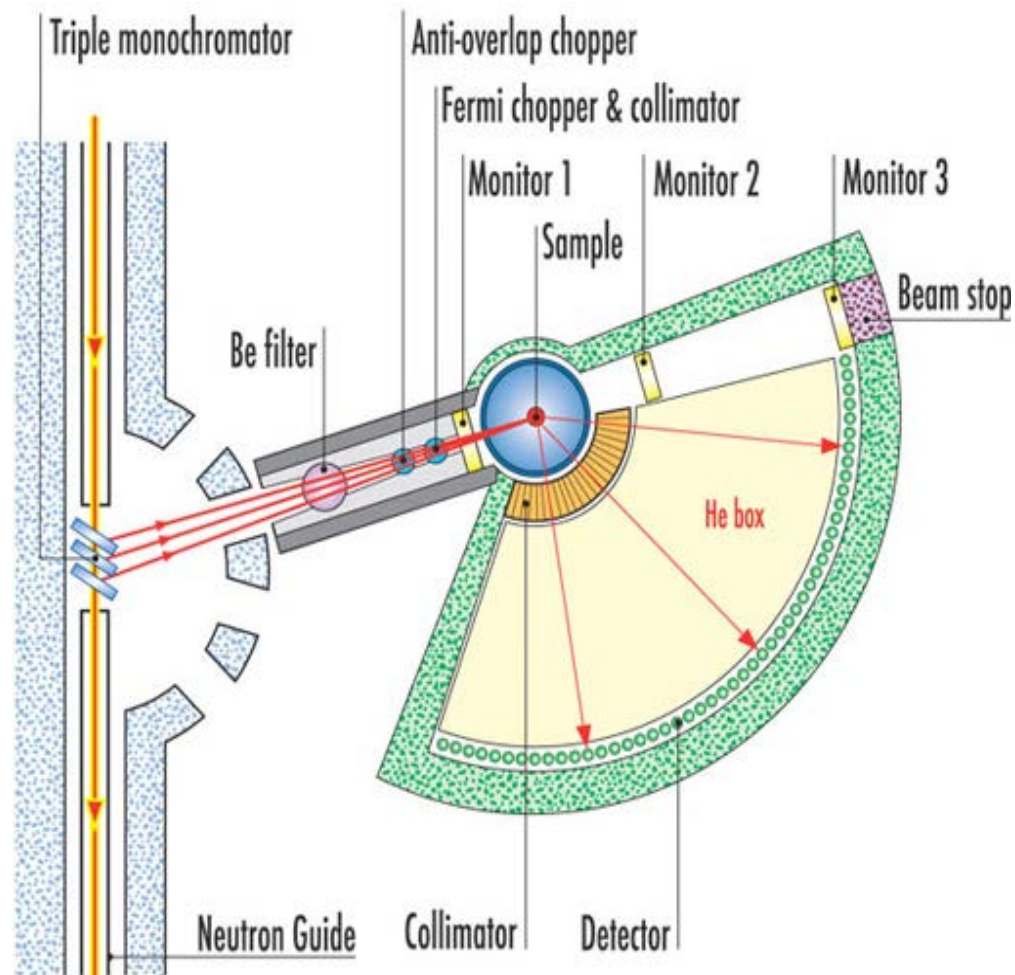
(Instruments are identified by the instrument's symbols which correspond to experimental techniques)

IN6:



IN6 Experimental facility

- Incident neutron wavelength λ : 5.1Å
- Corresponding incident neutron energy: 3.145 meV



Schematic representation of the IN6

For light water: Measurements at several temperature and pressure combinations have been carried out.

Experimental conditions with IN4c
Wavelength= 2.4 Å; $E_i= 14.20$ meV

Temperature (Kelvin)	Pressure (bar)
300	1
300	88-100
350	115
392-466	128-165
479-485	172-180
490-497	185
517	42



Cryofurnace

Experimental conditions with IN6
Wavelength= 5.1 Å; $E_i= 3.14$ meV

Temperature (Kelvin)	Pressure (bar)
350	1
494	70
494	340-600

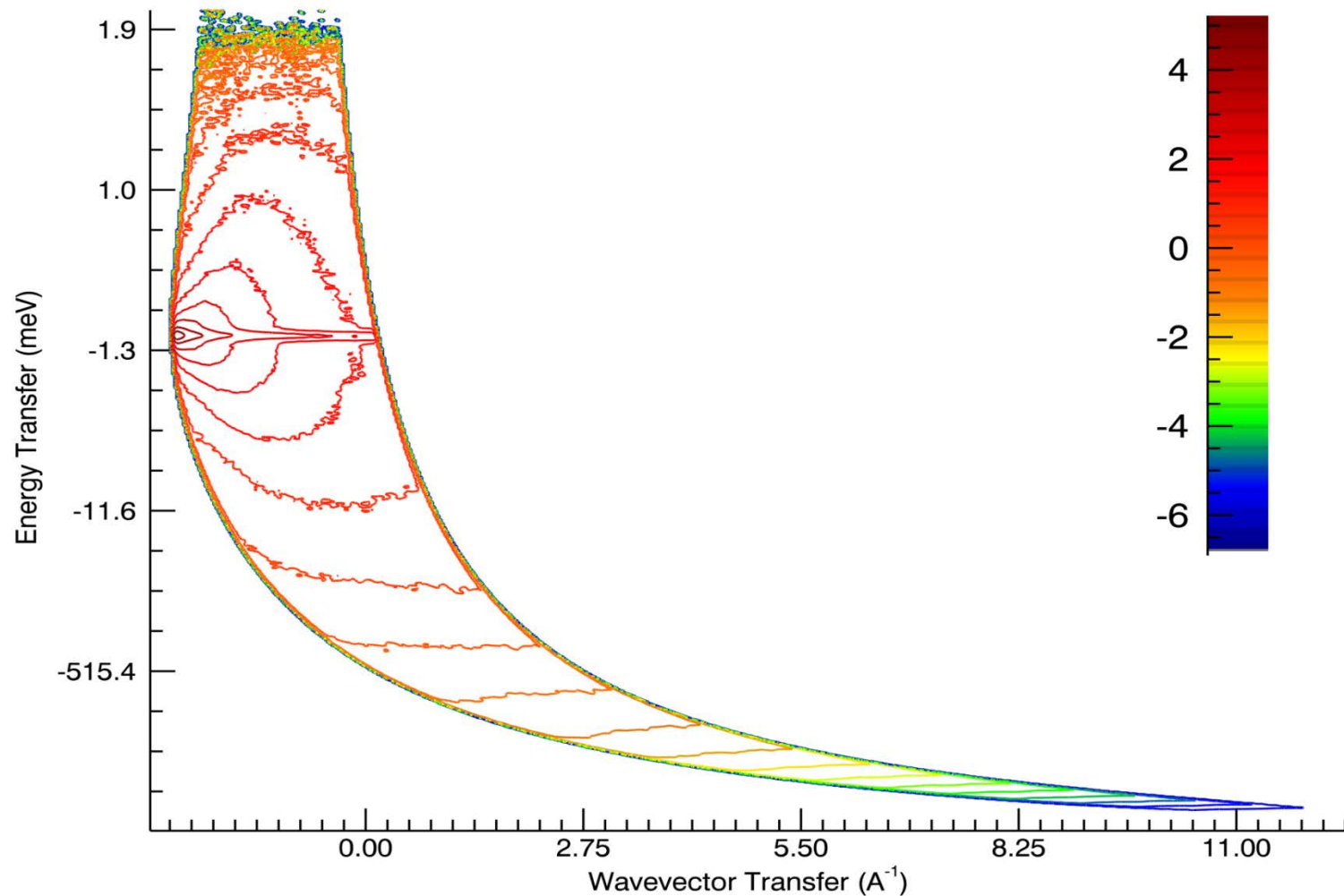
High intensity neutron beam from ILL Réacteur à Haut Flux (RHF)

A detailed data reduction of the raw time-of-flight data is done using data analysis tools (LAMP, iFit) in order to:

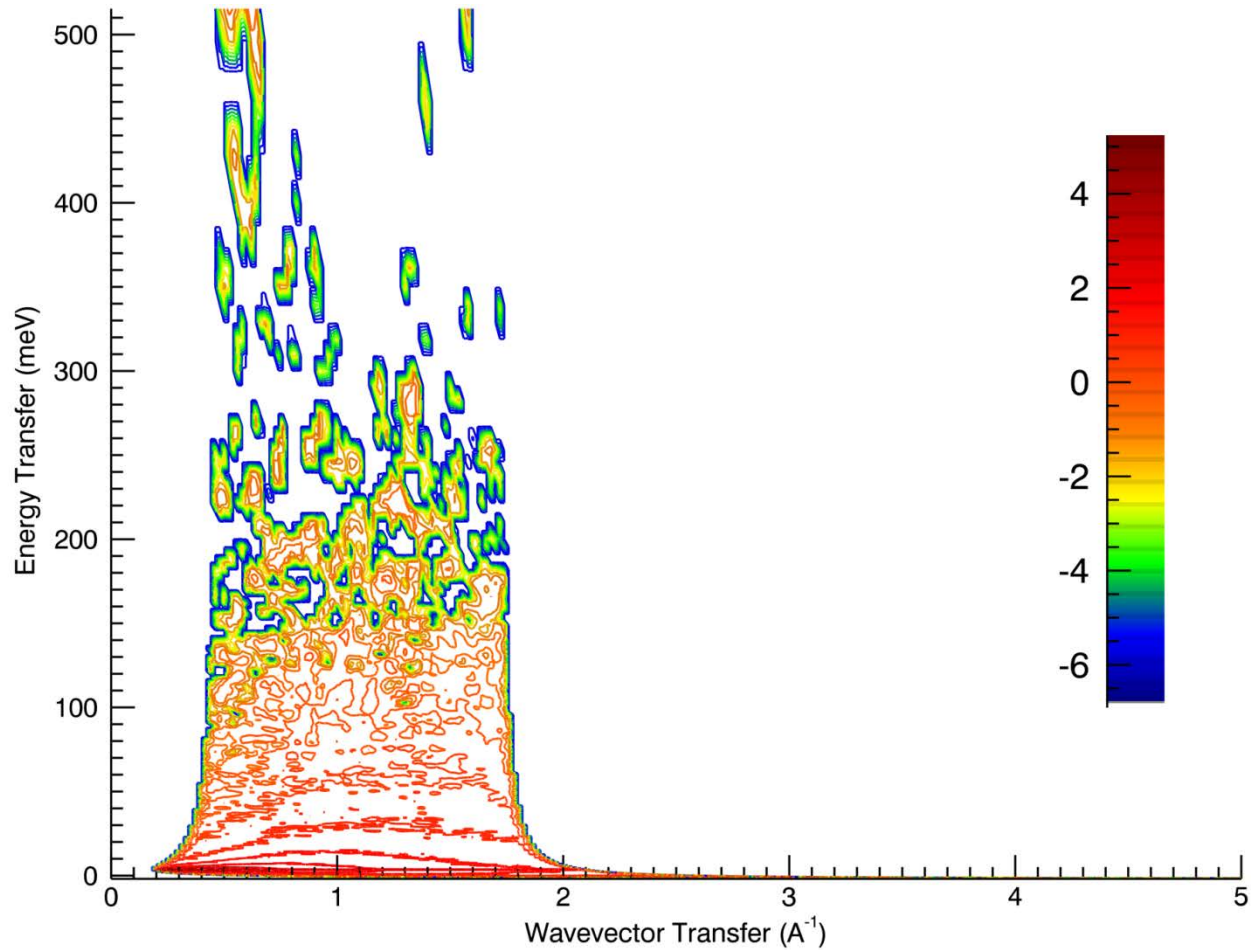
- Normalize the data with the monitor
- Subtract the empty cell
- Correct for self-shielding/self-absorption
- Calibrate with the vanadium reference
- Correct for the detector efficiency
- Account for the multiple scattering contribution (to be done)

Finally we obtain $S(\vec{q}, \omega)$ for each experimental conditions (Temperature and pressure)

$S(\vec{q}, \omega)$ of light water measured at 494 K & 70 bar (IN6)

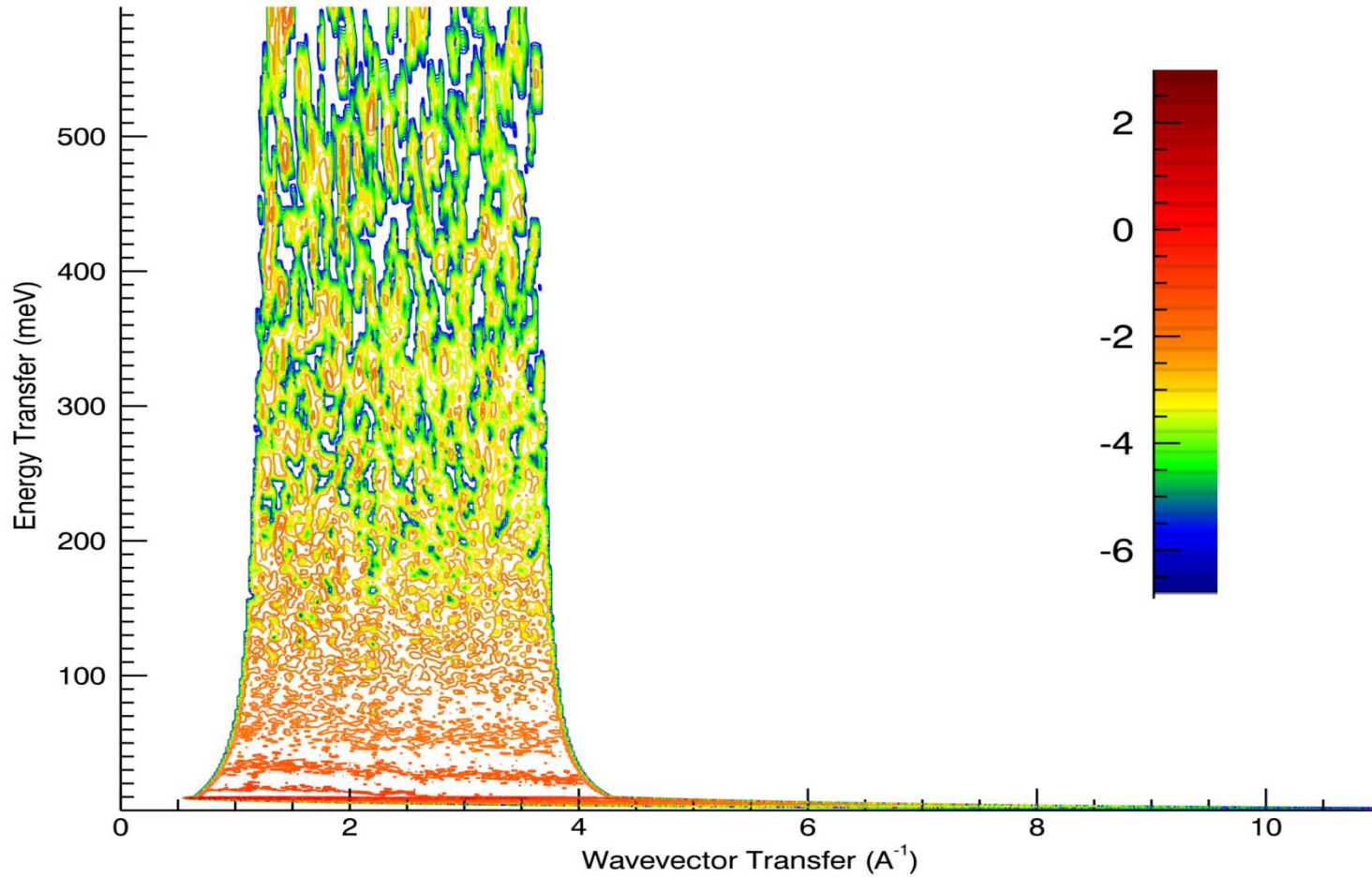


$S_{\text{sym}}(\vec{q}, \omega)$ of light water measured at 494 K & 70 bar (IN6)

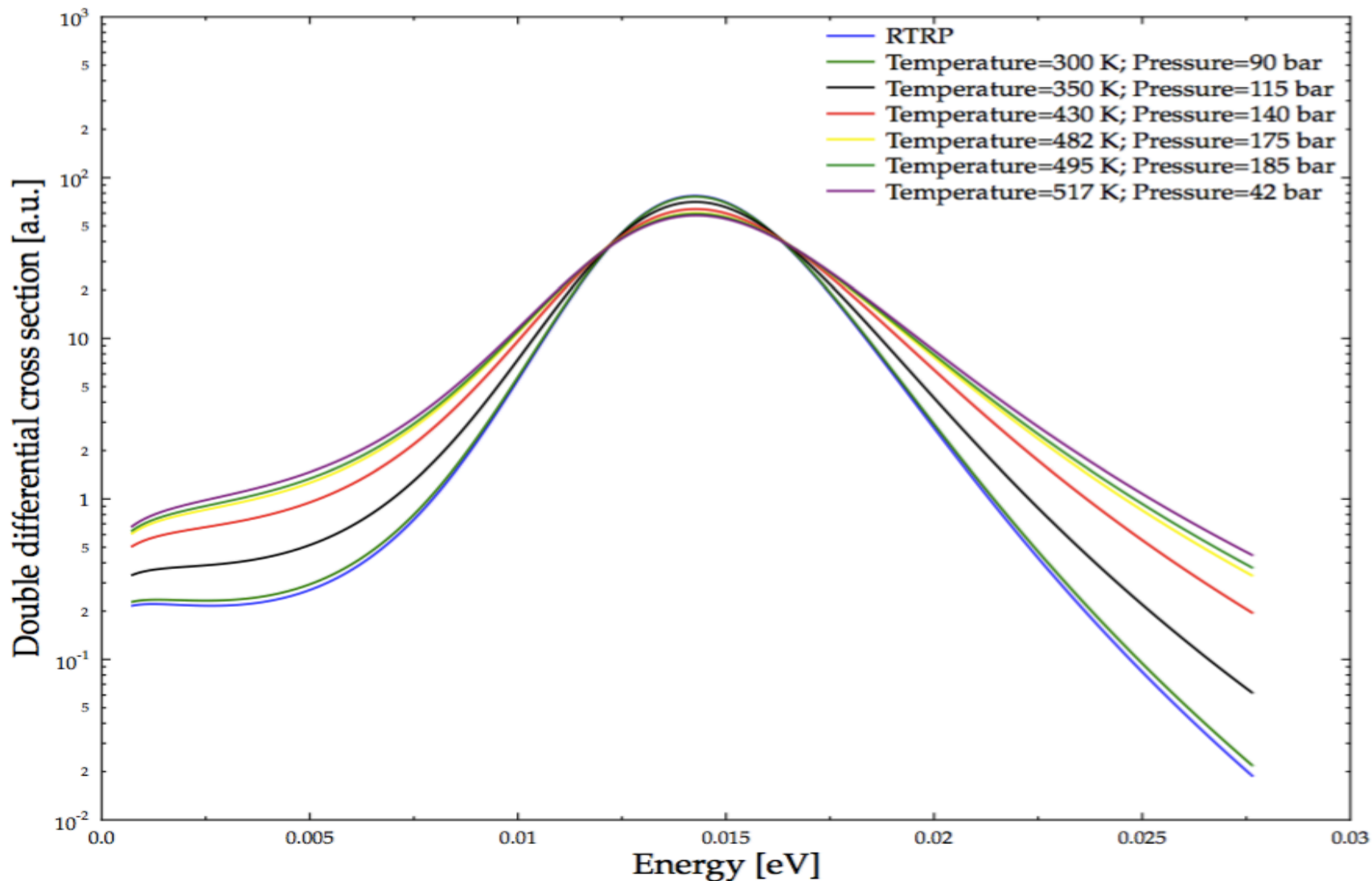


Preliminary Results

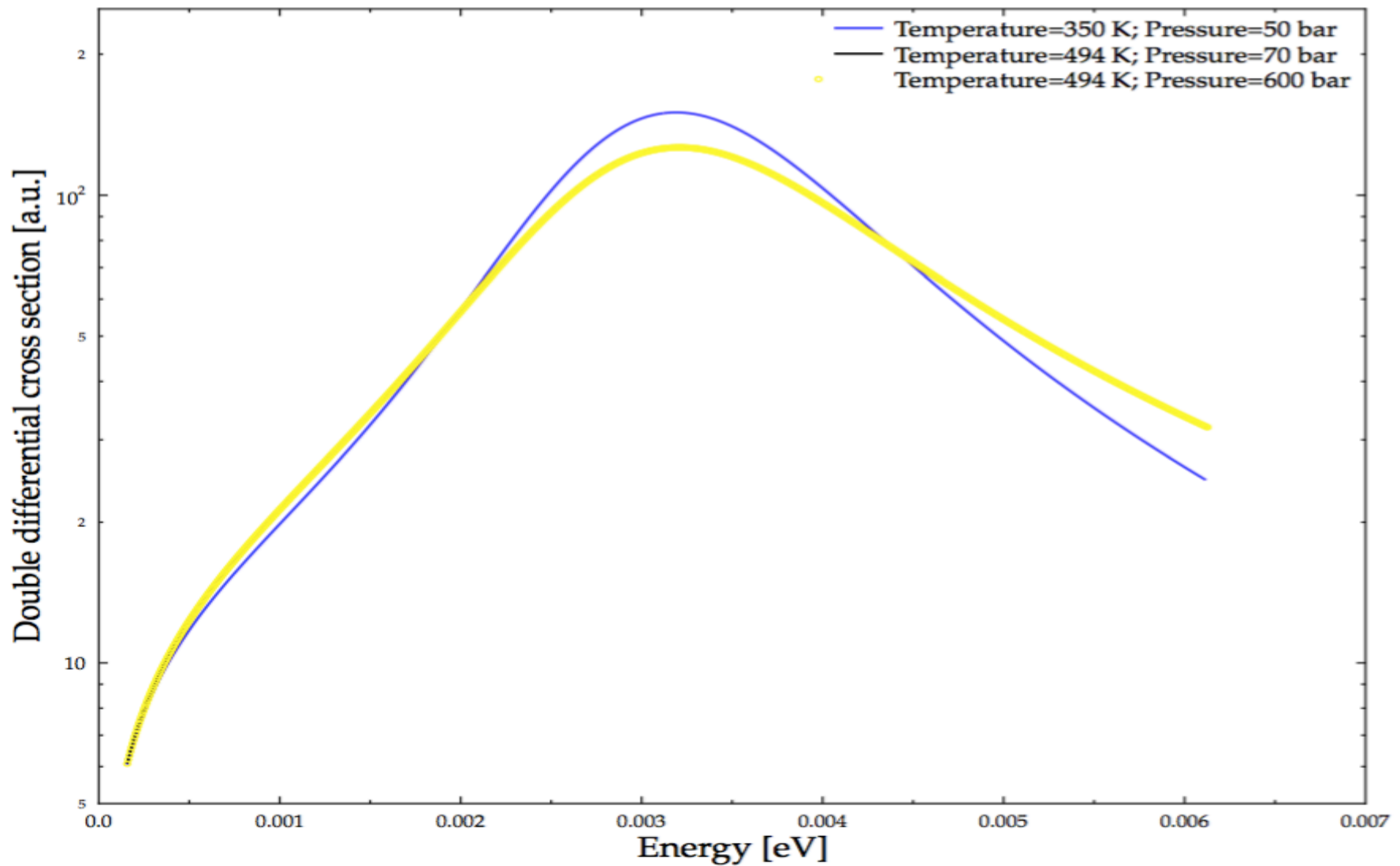
$S_{\text{sym}}(\vec{q}, \omega)$ of light water measured at 495 K & 185 bar (IN4c)



Double differential cross section ($E_i = 14.2$ meV; $\theta = 15^\circ$)



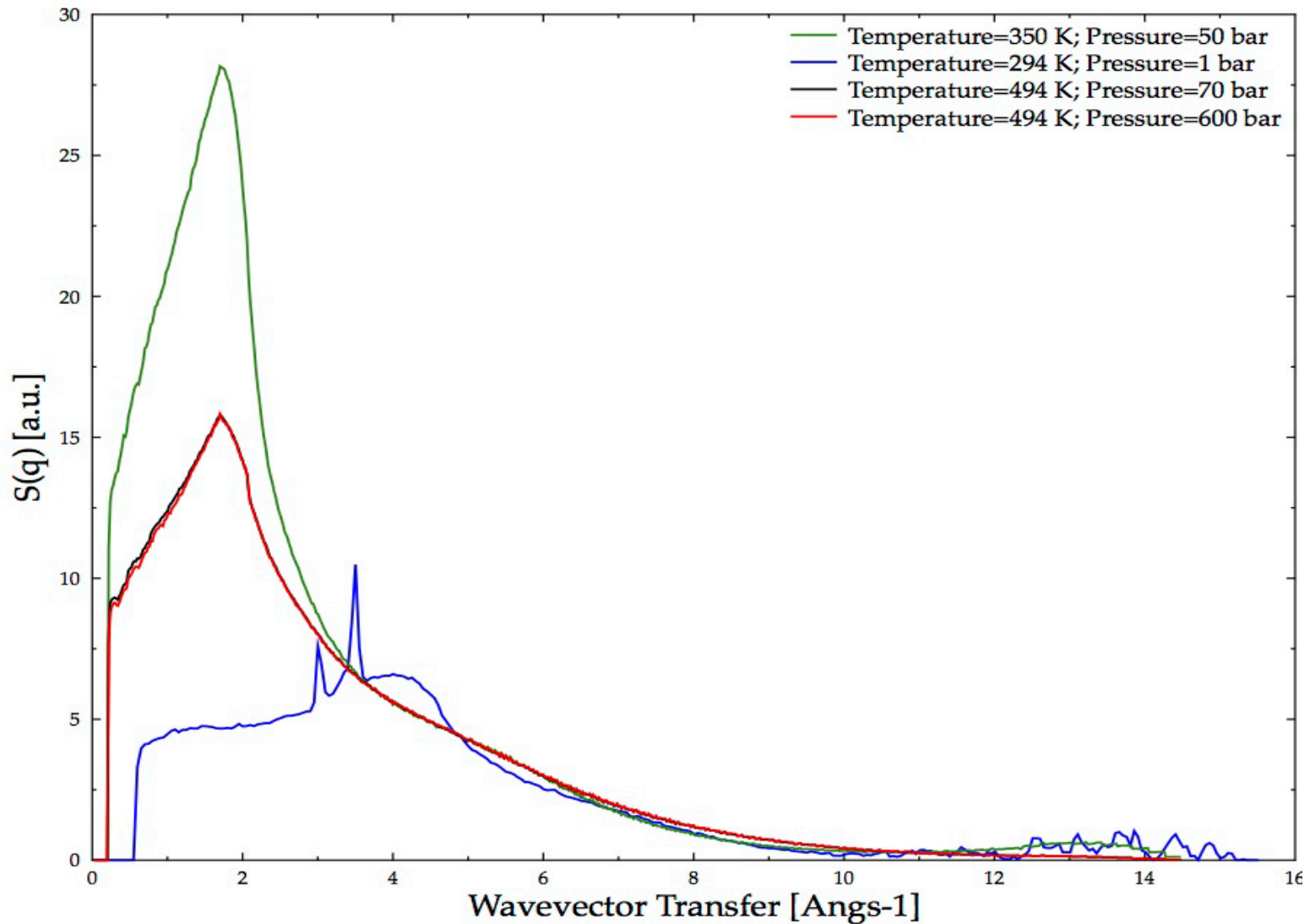
Double differential cross section ($E_i = 3.14$ meV; $\theta = 15^\circ$)



Preliminary Results

$S(q)$: Experimental structure factor

$$S(q) = \int_{-\infty}^{\infty} S(\vec{q}, \omega) d\omega$$

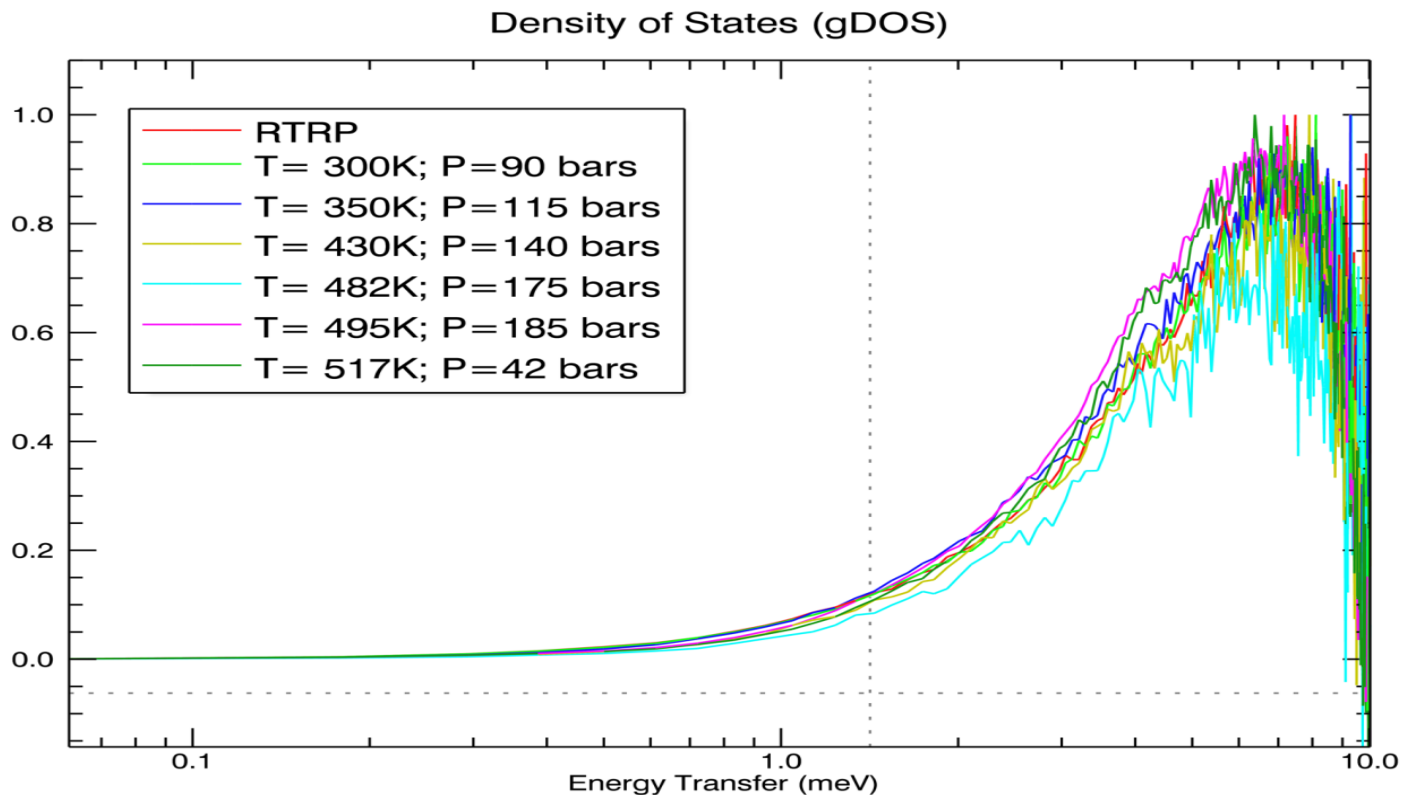


gDOS: Generalized density of states

with IN4c...

Measured generalized density of states (gDOS) is related to the classical dynamic structure factor by

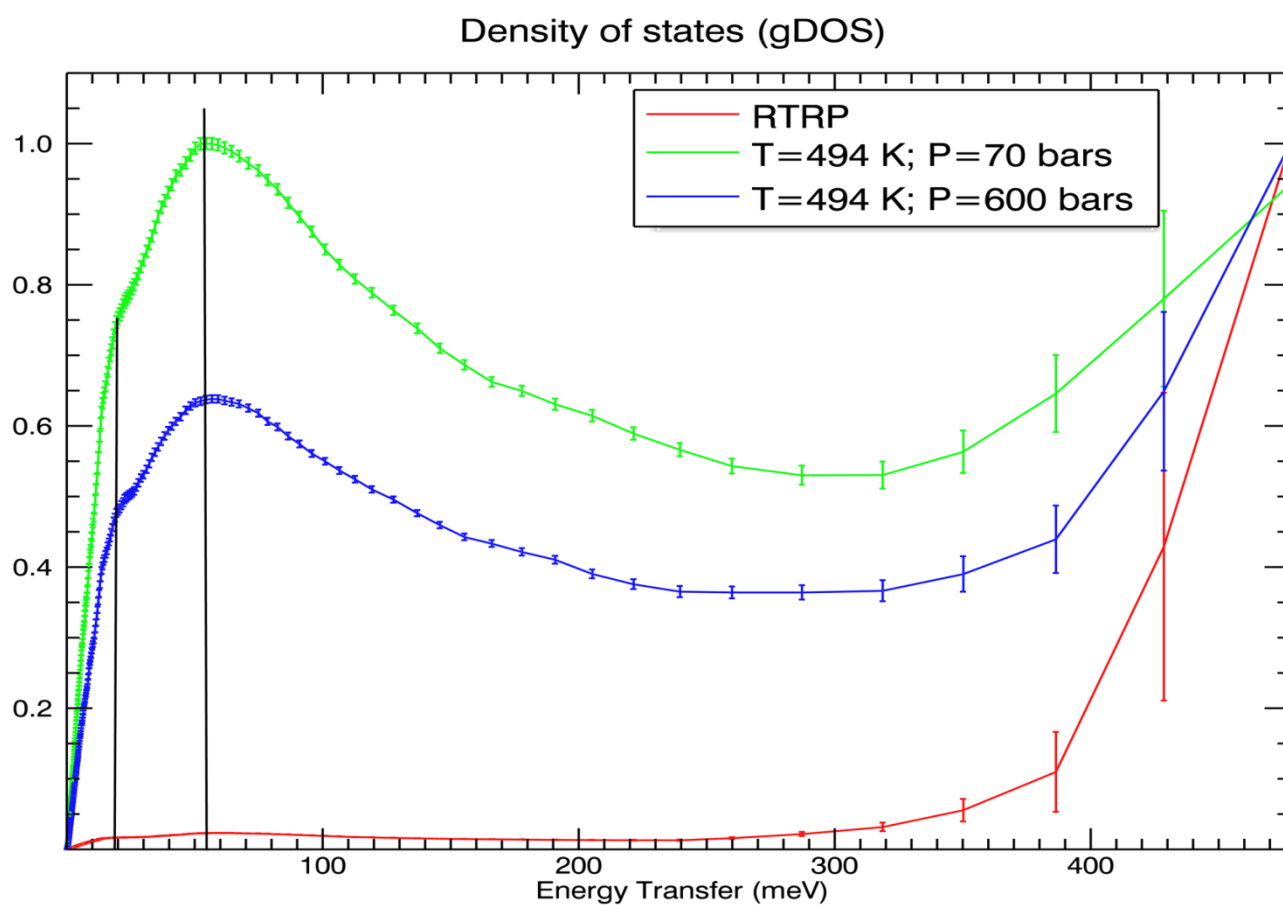
$$\lim_{q \rightarrow 0} \frac{\omega^2}{q^2} S_{\text{sym}}(\vec{q}, \omega) \simeq g(\omega)$$



The translation motion peak can be seen around 7 meV

gDOS: Generalized density of states

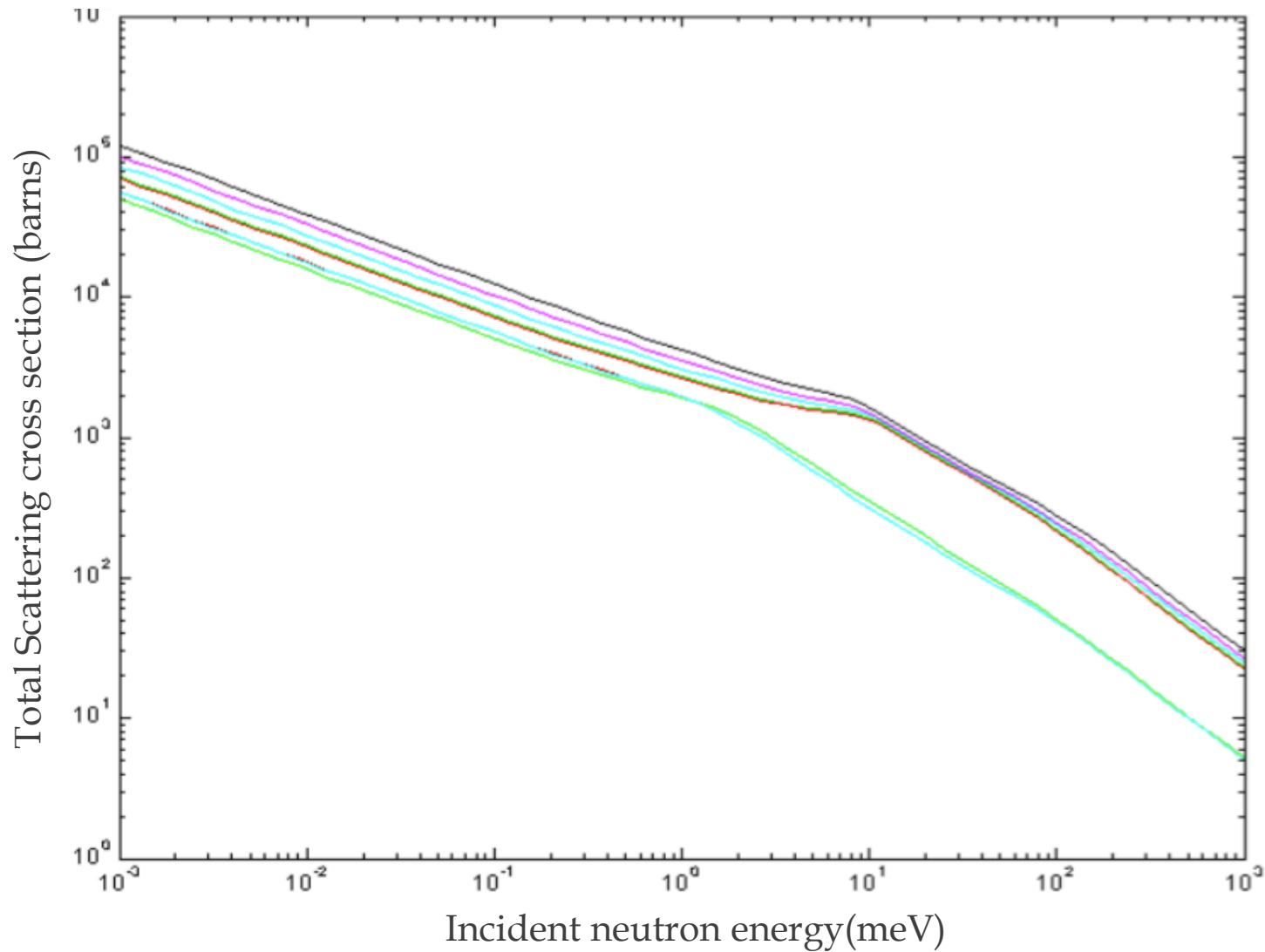
with IN6...



Liberation bands of light water is seen around 55 meV

Total cross section:

(Results using iFit 1.8)



Where are we right now?

EXPERIMENT



$$S(\vec{q}, \omega) = \exp\left(\frac{\hbar\omega}{2k_B T}\right) S_{\text{sym}}(\vec{q}, \omega)$$

$$S(\alpha, \beta) = k_B T S_{\text{sym}}(\vec{q}, \omega)$$

+

MOL. DYNAMICS



$$S(\alpha, \beta)$$



$$g(\omega) \approx \lim_{q \rightarrow 0} \frac{\omega^2}{q^2} S_{\text{sym}}(\vec{q}, \omega)$$



LEAPR (NJOY)



$$S(\alpha, \beta)$$



COMPARE

Done!

To be done!

Future work & Remarks :

- Correct the experimental data for the multiple scattering contribution.
- Combine data from both the IN4c, IN5 and IN6 spectrometers to increase the statistical accuracy.
- Due to kinematic restrictions and limited angular range of detectors, $S(\vec{q}, \omega)$ is measured only in a limited region of the (q, ω) space. Hence investigate the possible use of molecular dynamics codes to access $S(\vec{q}, \omega)$ over a wider range.
- Prepar new experimental $S(\alpha, \beta)$ data in the ENDF format.
- Analyze the uncertainties associated with the experimental conditions in connection with the measurements, namely the systematic and the statistical uncertainties.

Emmanuel FARHI (ILL)
Yoann CALZAVARA (ILL)

Gilles NOGUERE (CEA)
Juan Pablo SCOTTA (CEA)

Experimental support:

Stephane ROLS (IN4c scientist)
Jacques OLLIVIER (IN6 scientist)

Olivier MEULIEN (IN4 technician)
Jerome HALBWACHS (IN6 technician)

Claude PAYRE (Pressure technician)

Thank you !