IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

*Faire avancer la sûreté nucléaire*

# Thermal Scattering Law for Light Water

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# Agenda

- **Motivation**
- **Experimental approach**
- **Theoretical approach**
- **Conclusions and perspectives**


**"Neutron Man"**

* Neutron Scattering: A Primer by Roger Pynn

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## Our goal can be expressed as:

- Improve the thermal scattering cross-section data of light water at operating conditions of nuclear power reactors (High temperature and pressure).

- Study the uncertainties associated with it, namely the systematic and statistical uncertainties.

**How to achieve these goals?**

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

- On the experimental side: **Perform new time-of-flight (TOF) experiments**.

- On the theoretical side: Review the existing models and perform **Molecular Dynamics (MD) simulations**.

# *Thermal Scattering law:* S(α,β)

- The molecular binding effects are described by a S(α,β) function which is often termed as thermal scattering law (TSL).
- The probability of interaction of thermal neutrons are described by the double differential scattering cross-section $\frac{\partial^2\sigma}{\partial\Omega\partial E}$
- The double differential cross-section (DDXS) for neutrons with incident energy E, secondary energy E′ and scattering angle Ω is related to S(α,β) by :

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'}(E \rightarrow E', \Omega \rightarrow \Omega') = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}\, e^{-\frac{\beta}{2}}\, S(\alpha, \beta)$$

*Momentum transfer* is represented by: $\alpha = \frac{E' + E - 2\sqrt{EE'}\,\mathrm{Cos}\theta}{AkT}$

*Energy transfer* is represented by: $\beta = \frac{E' - E}{kT}$

**S(α, β)**

# Dynamic Structure factor: $S(\vec{q}, \omega)$

- In neutron scattering experiment the measured intensity is proportional to the double differential cross section:

$$\frac{\partial^2 \sigma_T}{\partial \Omega \partial \omega} = \frac{\sigma_b}{4\pi} \frac{K_i}{K_f} S(\vec{q}, \omega)$$

*Momentum transfer* is represented by:

*Energy transfer* is represented by:

$$\vec{q} = \vec{k_i} - \vec{k_f}$$

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

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

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

**TOF Experiments:**

- We make use of neutrons to measure $S(\vec{q}, \omega)$
- **Quantum thermal scattering law (Asymmetric)**
- **Contains the detail balance**

**MD Simulations:**

- We directly obtain the $S(\vec{q}, \omega)$
- **Classical thermal scattering law (Symmetric)**
- **We have to apply some quantum correction**

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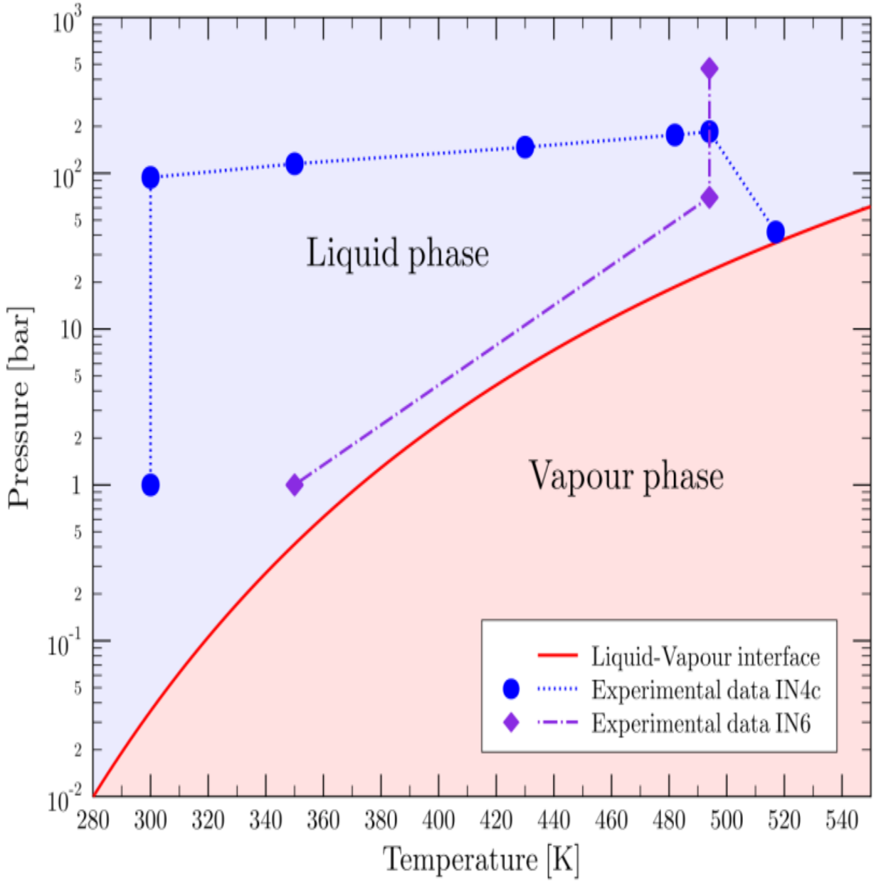
## Time-of-flight experiment at ILL, Grenoble:

### IN4c

### IN6

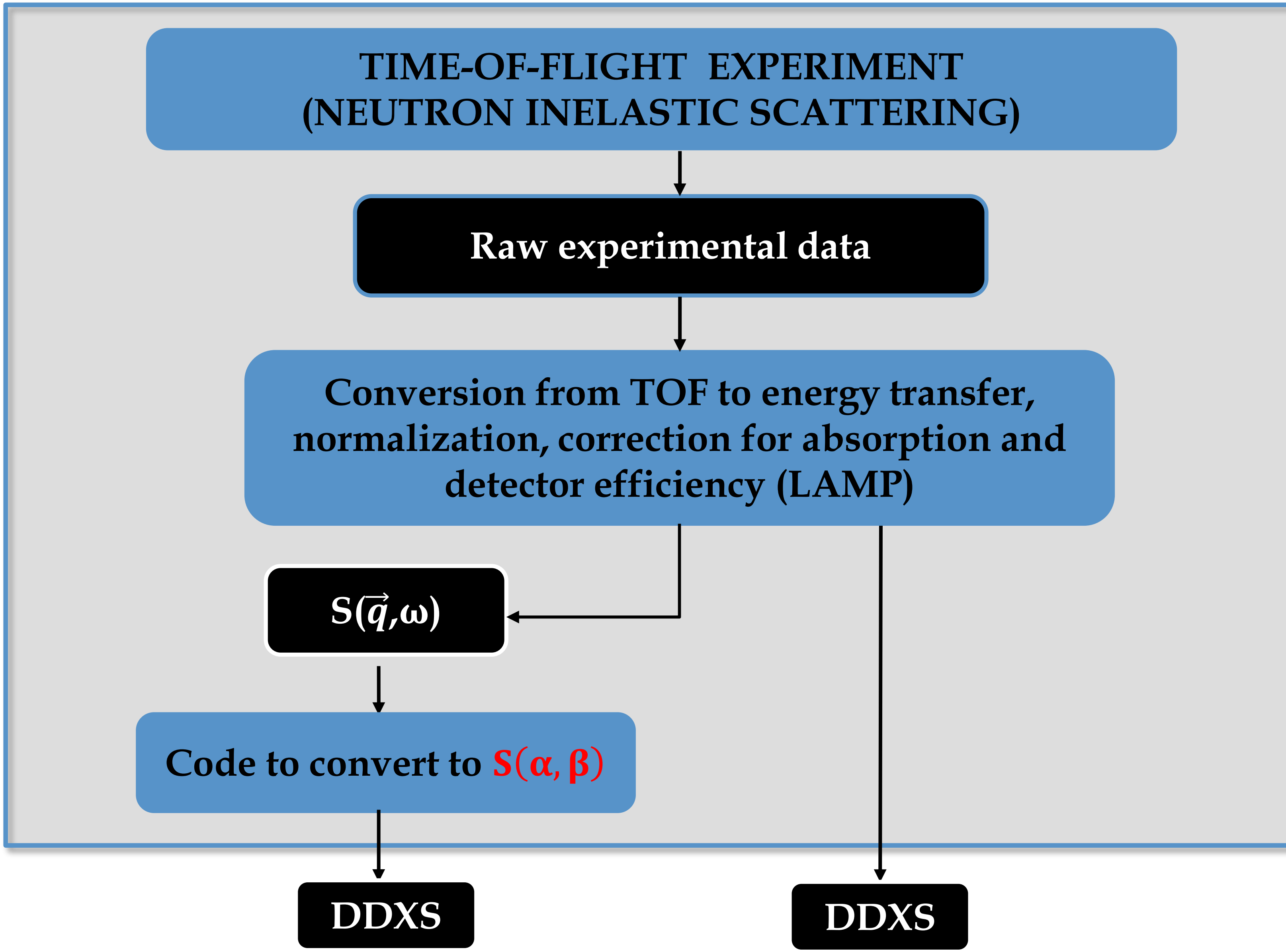
## Time-of-flight experiment at ILL, Grenoble:



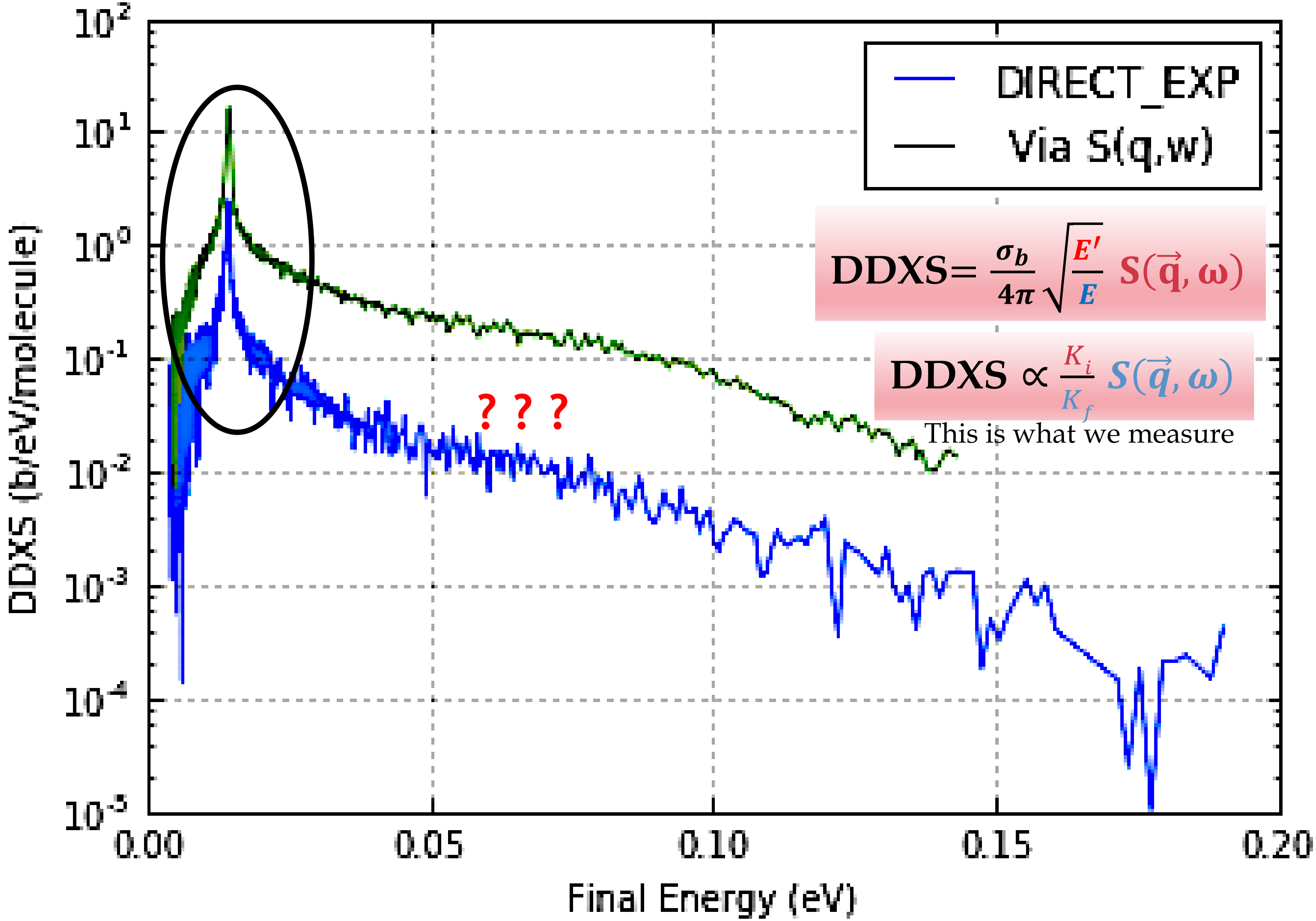

| TOF spectrometer | Temperature (K) | Pressure (bar) |
|---|---|---|
| IN4c<br>λ =2.4 Å;<br>E= 14.2 meV | 300 | 1 |
| | 300 | 88-100<br>(94) |
| | 350 | 115 |
| | 392-466<br>(430) | 128-165<br>(147) |
| | 479-485<br>(482) | 172-180<br>(176) |
| | 490-497<br>(494) | 185 |
| | 517 | 42 |
| IN6<br>λ =5.1 Å;<br>E= 3.15 meV | 350 | 1 |
| | 494 | 70 |
| | 494 | 340-600<br>(470) |

**Thermodynamic states (experimental conditions) of light water for the data measurement.**

# Experimental approach



# *Result (Double differential cross-section)*

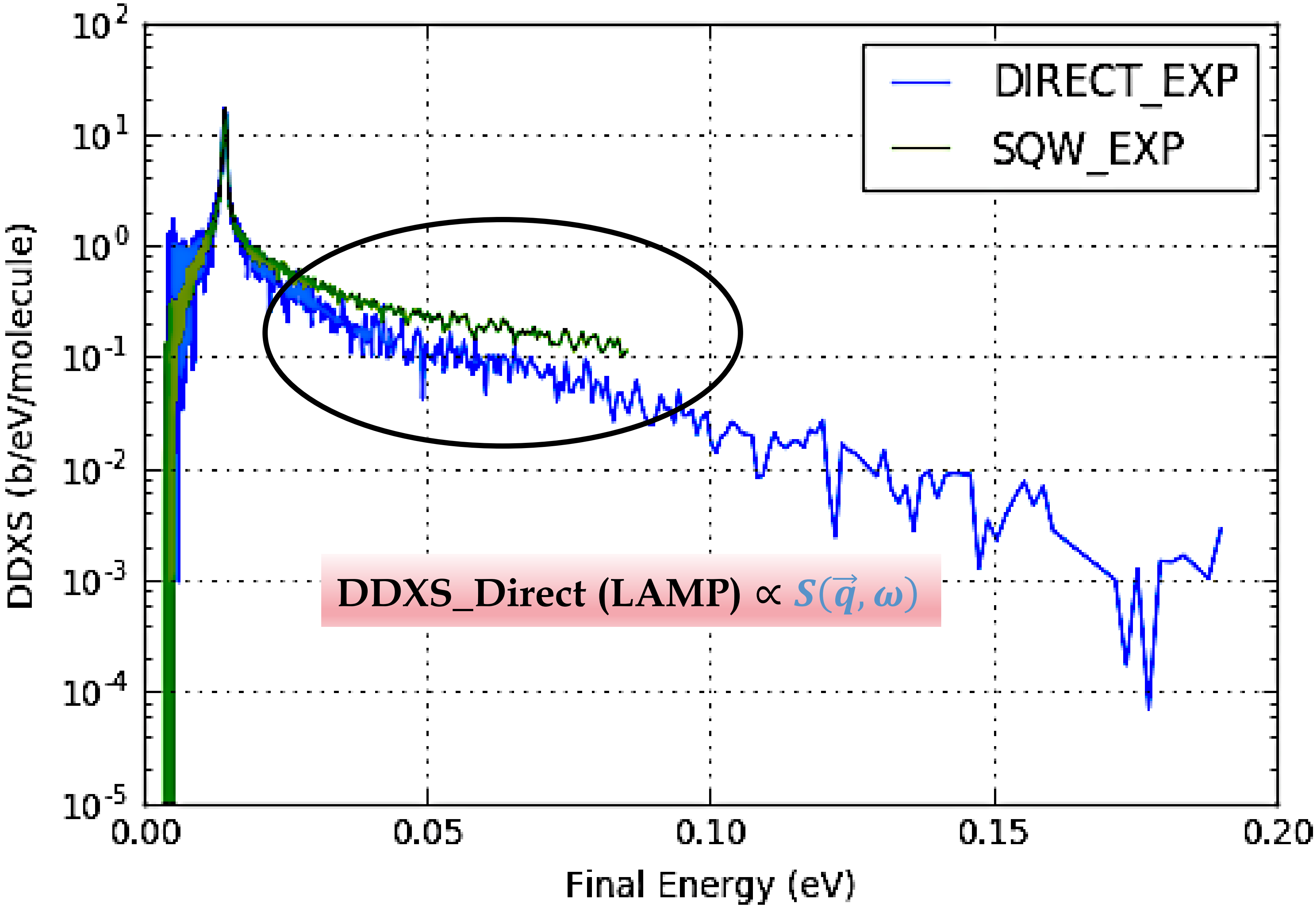



$$\mathrm{DDXS} = \frac{\sigma_b}{4\pi}\sqrt{\frac{E'}{E}}\, S(\vec{q},\omega)$$

$$\mathrm{DDXS} \propto \frac{K_i}{K_f}\, S(\vec{q},\omega)$$

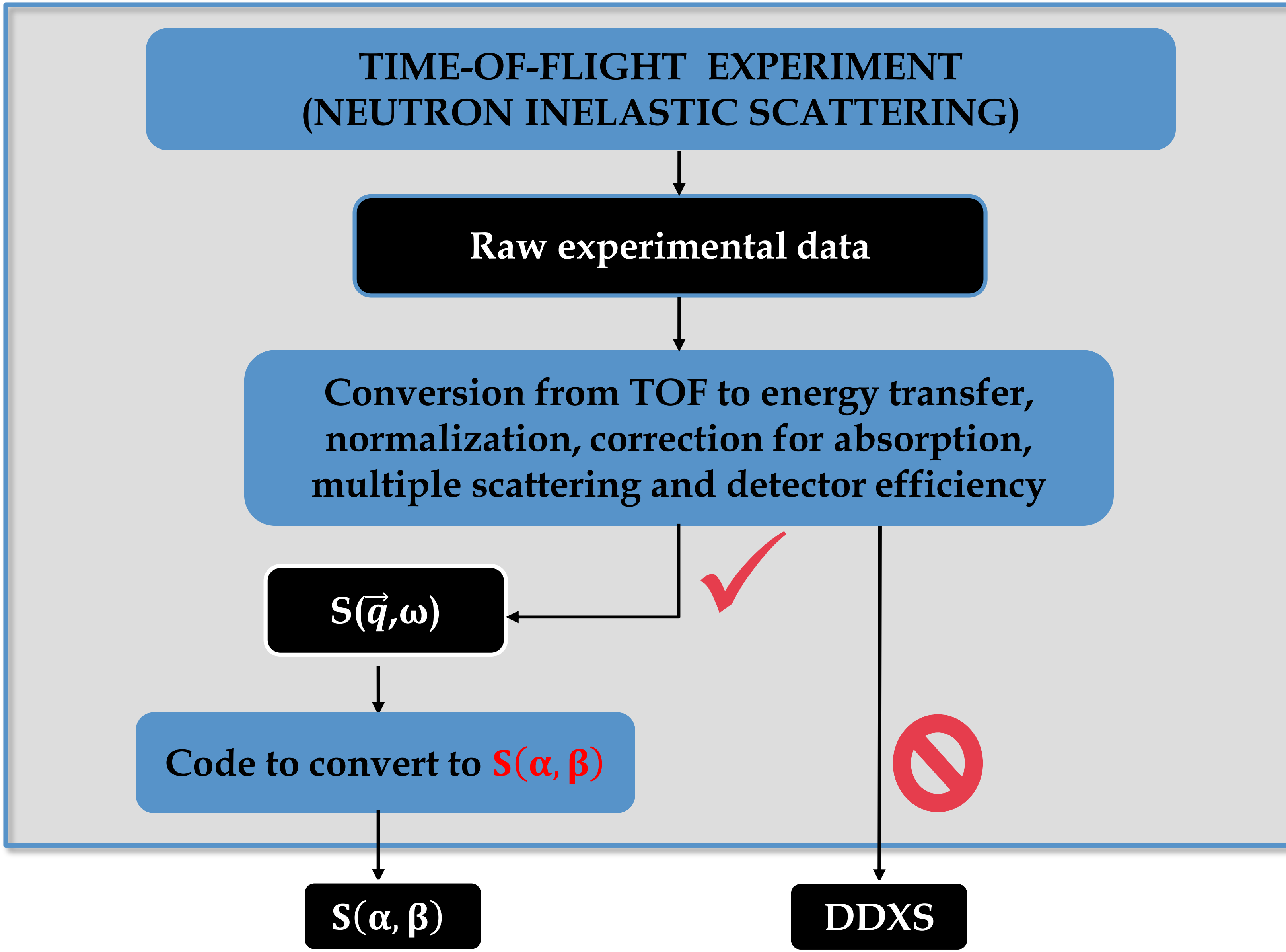
This is what we measure

# *Result (Double differential cross-section)*

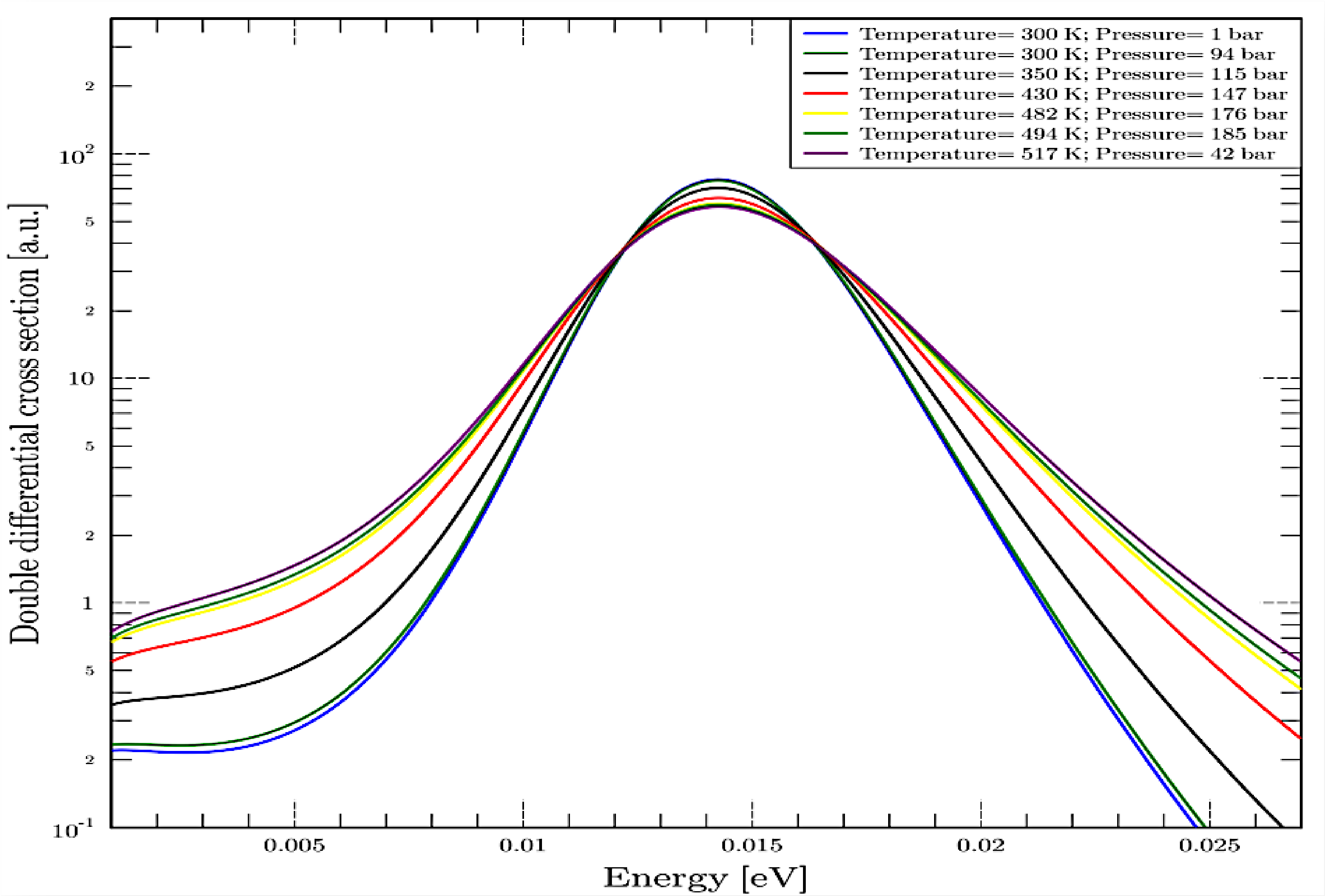



**DDXS_Direct (LAMP) ∝** $S(\vec{q}, \omega)$

# Experimental approach

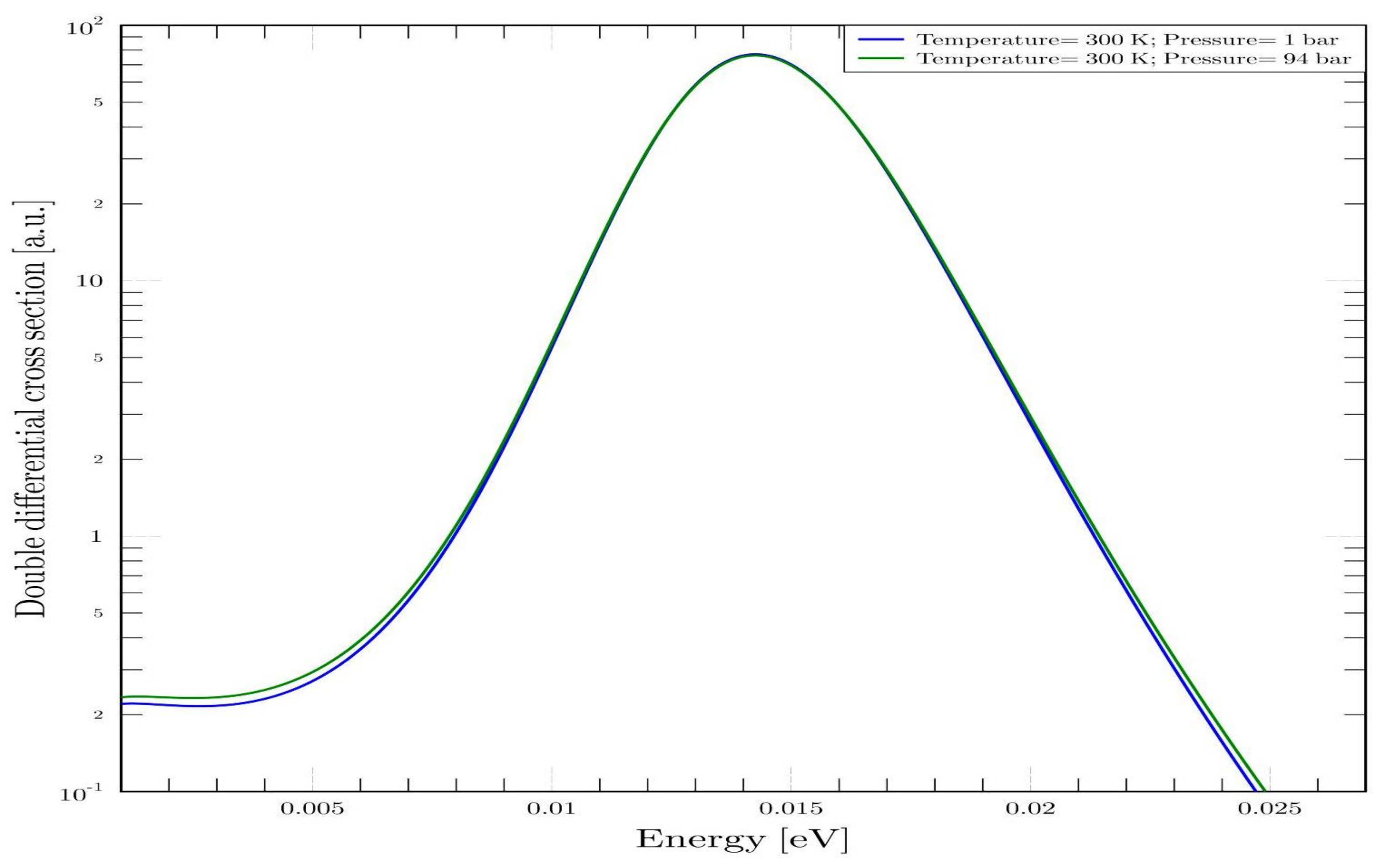


# *Result (Double differential cross-section)*




**Double differential scattering cross-section of light water measured at the IN4c ($E_i$=14.2 meV; θ=15°)**

# *Result (Double differential cross-section)*




**Double differential scattering cross-section of light water measured at the IN4c ($E_i$=14.2 meV; θ=15°)**

- At the same temperature there is negligible impact of the change in pressure on the double differential scattering cross-section.

- Presented this work at ND2016 (International conference on Nuclear Data for Science and Technology)
- Accepted for publication in the European Physics Journal (EPJ conference proceedings)

# Measurement of double differential cross-section of light water at high temperature and pressure to generate S(α,β)

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**Abstract.** A series of double differential inelastic scattering cross-section measurements were performed on light water at several temperatures and pressures using high resolution time-of-flight inelastic spectrometers, namely the IN4c and the IN6 at the Institut Laue-Langevin (ILL), Grenoble, France to investigate the impact of temperature and pressure on S(**q**,ω) and thus on the S(α,β) thermal scattering kernel. The present work aims at extending previous measurements with light water at room temperature and pressure to more realistic operating conditions in connection with nuclear power reactors.

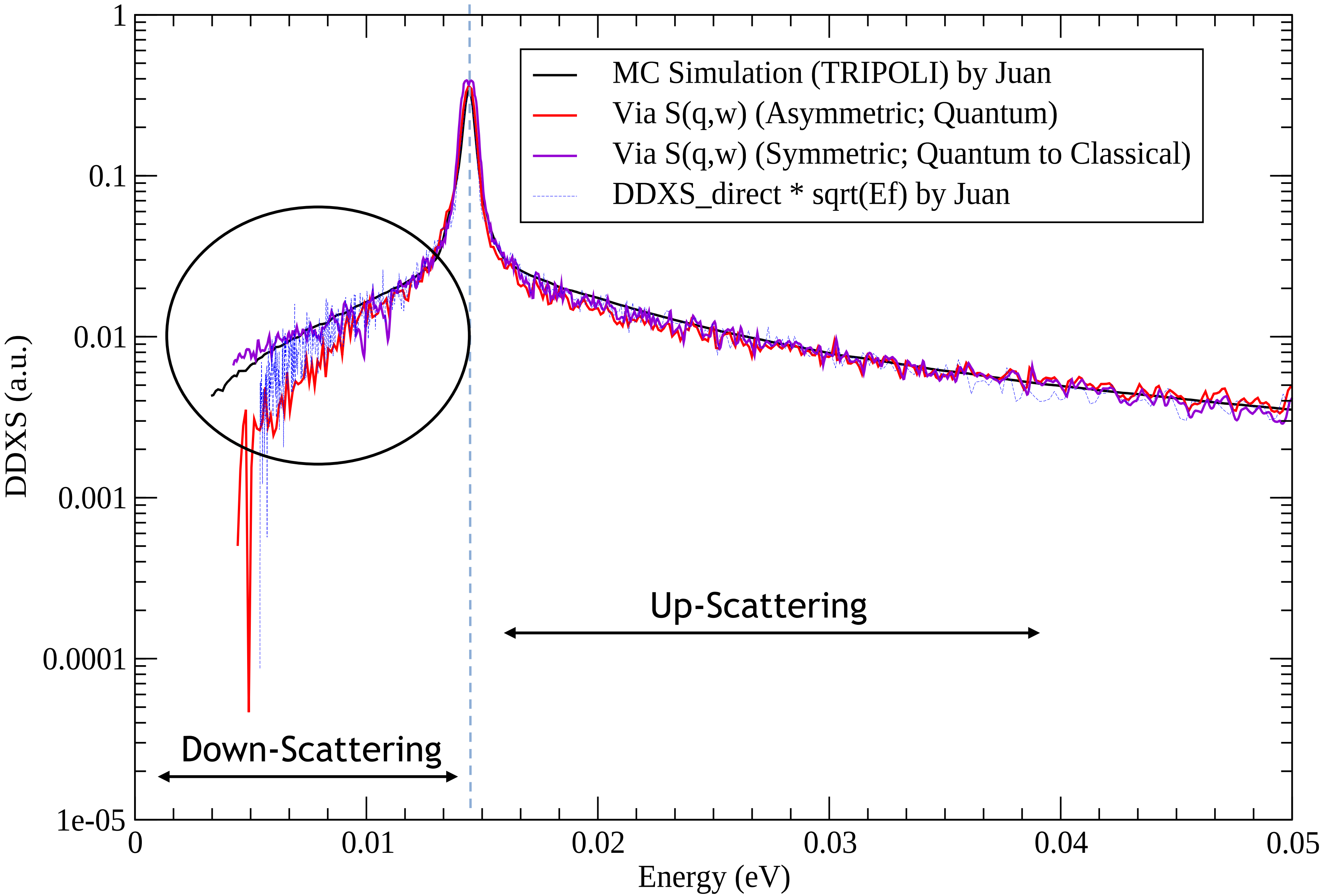
## Validation of experimental data:

- Experimental data is effected by multiple scattering due to thick sample.
- Experimental data inherently depends on the resolution of the spectrometer.
- Multiple scattering correction is necessary to compare experimental and theoretical data.

**As a preliminary solution?**

- Perform a monte-carlo simulation of the actual experiment with the multiple scattering effect to obtain the double differential cross-section.
- Compare it with experimental double differential cross-section.

# *Result (Double differential cross-section)*



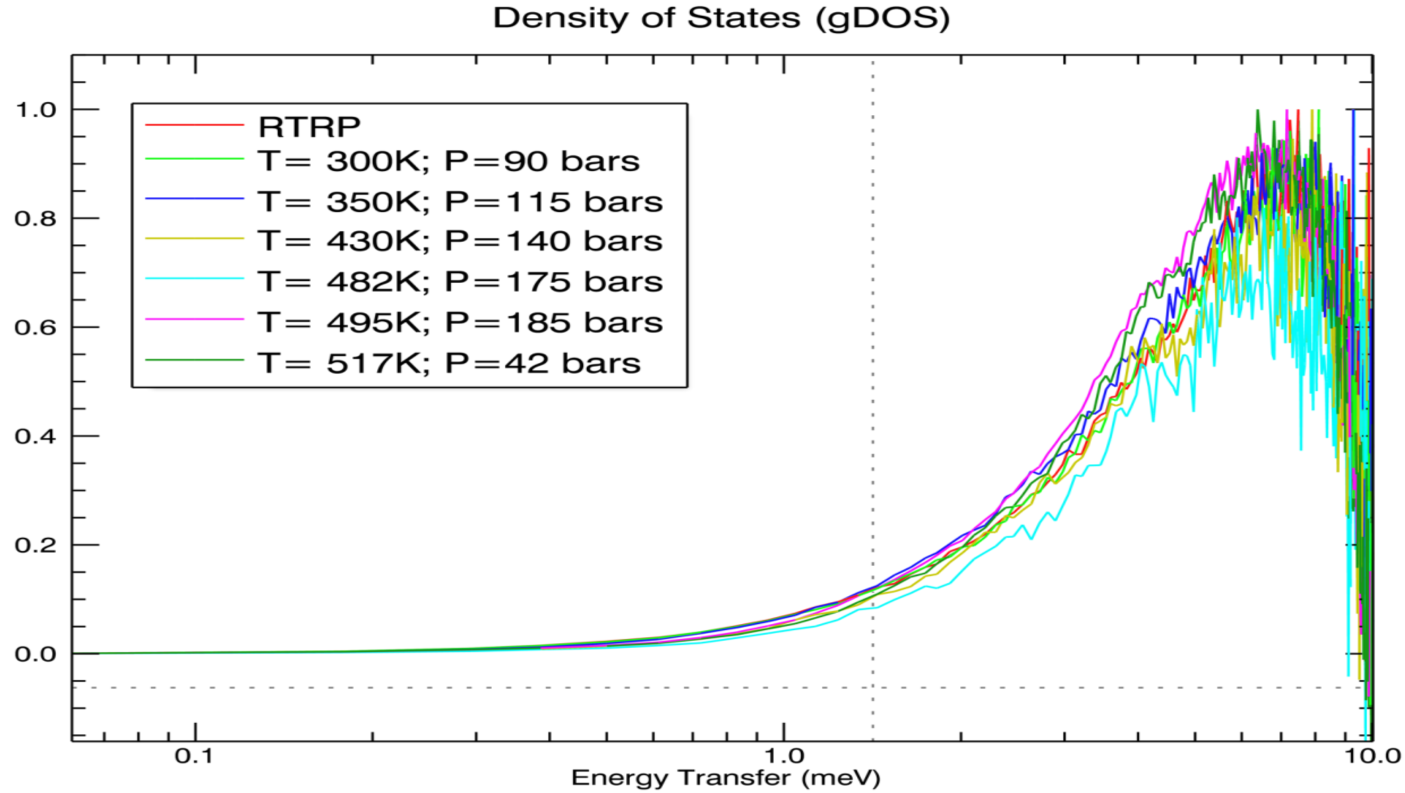

MC simulations were carried out by Juan Pablo Scotta, CEA, Cadarache.

## gDOS: Generalized density of states

with IN4c…

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

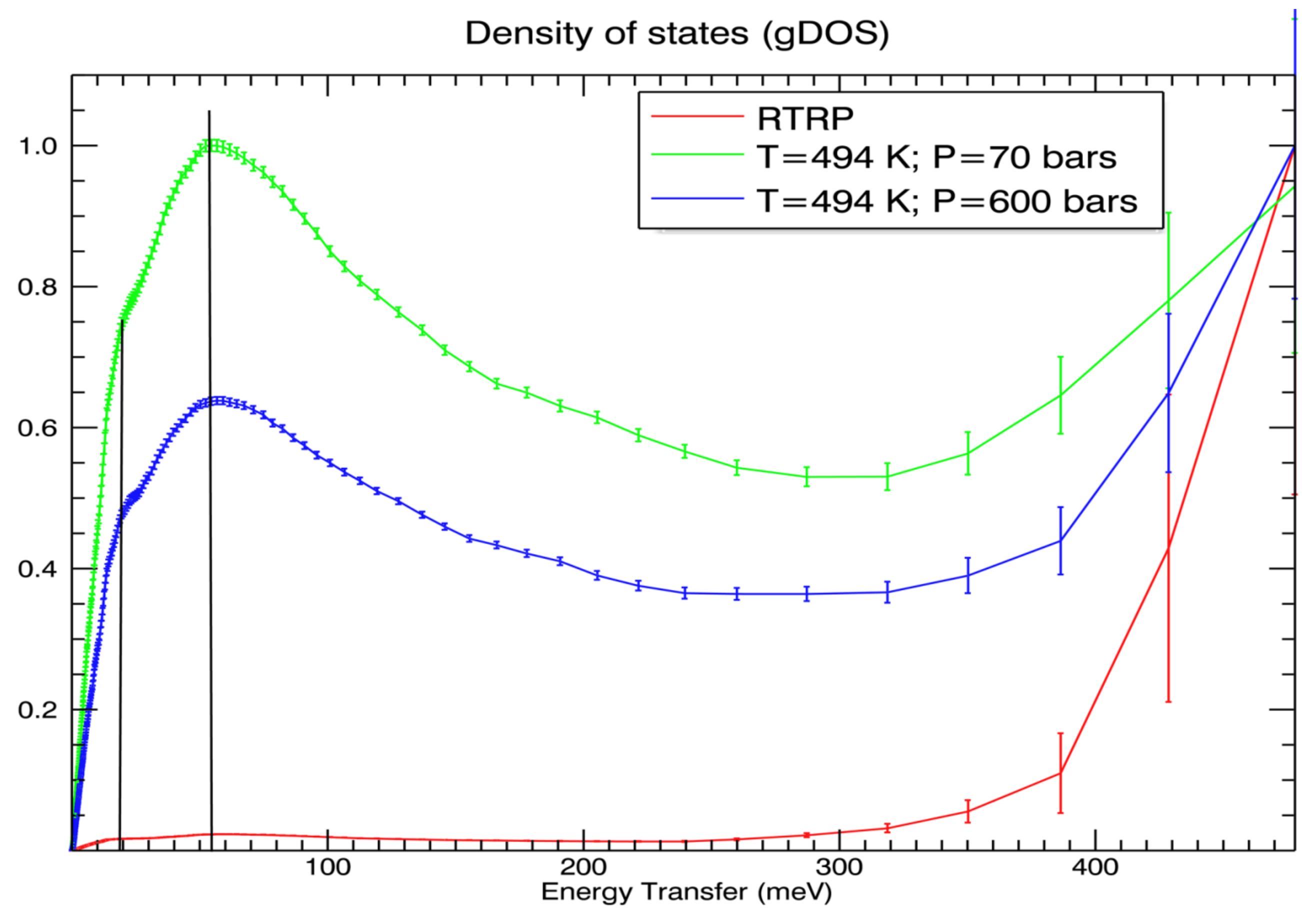
$$\lim_{q \to 0} \frac{\omega^2}{q^2} S_{sym}(\vec{q}, \omega) \simeq g(\omega)$$




The translation motion peak can be seen around 7 meV

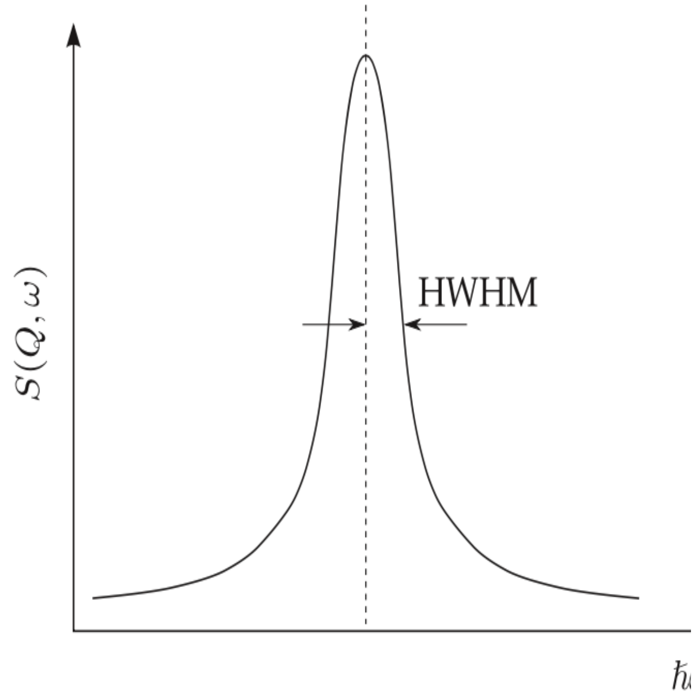
## gDOS: Generalized density of states

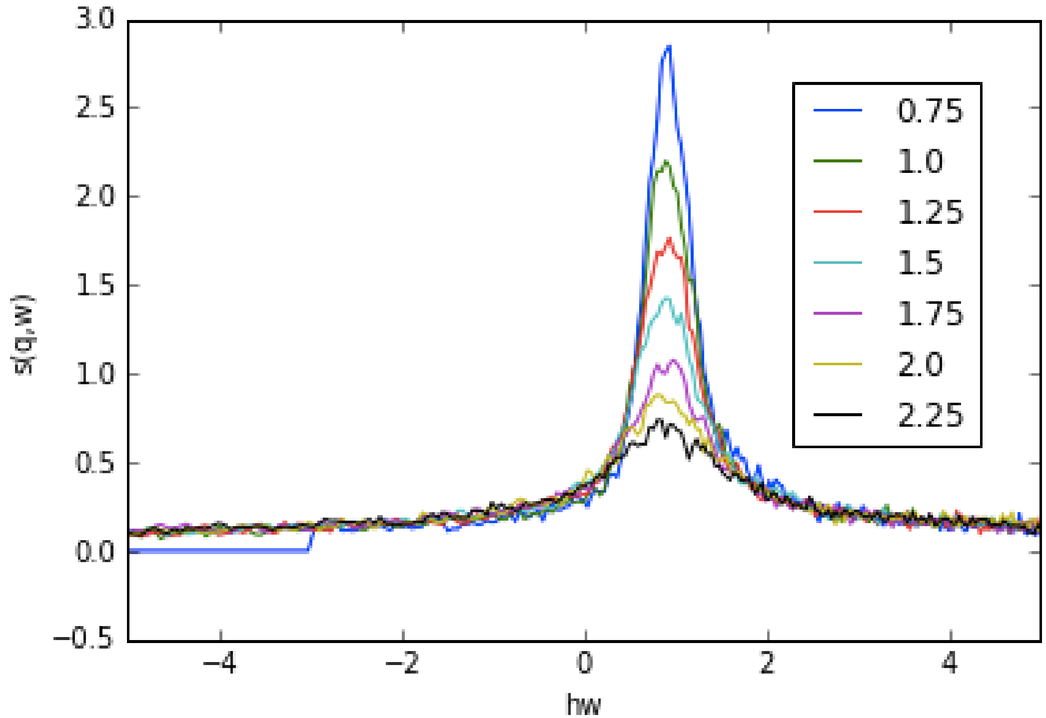
**with IN6…**

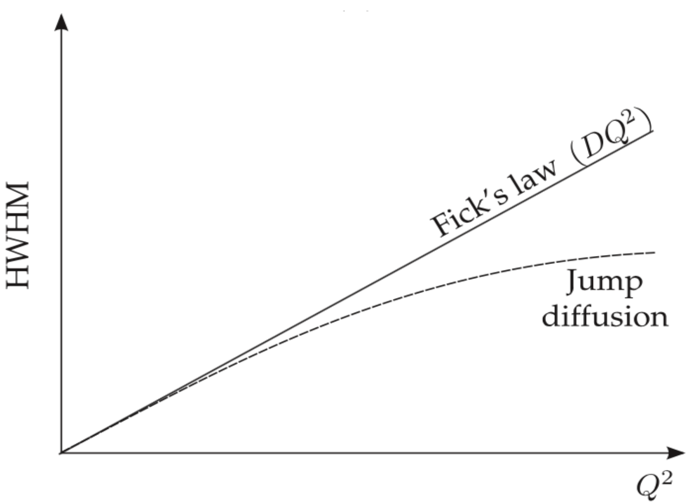


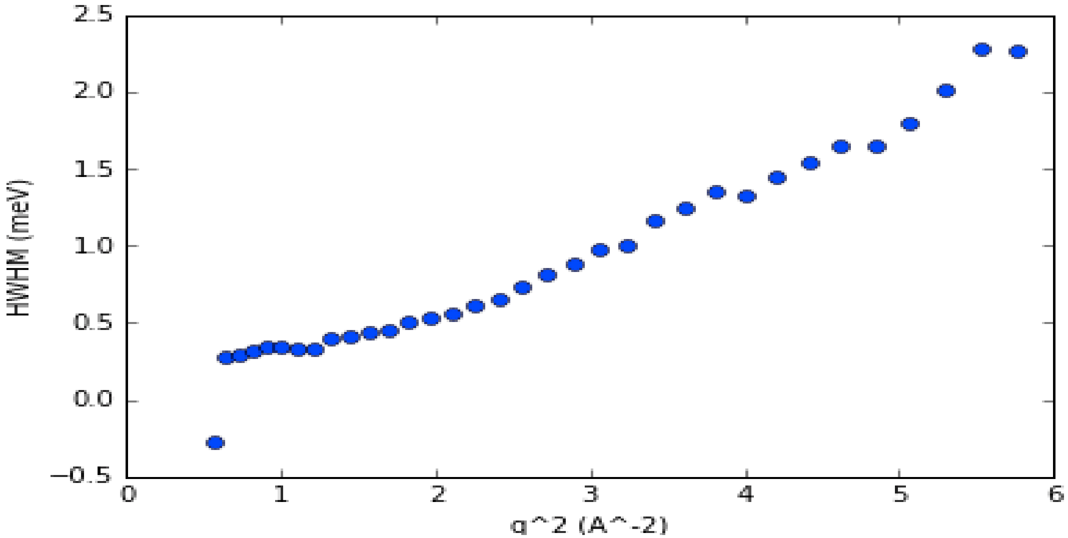

Liberation bands of light water is seen around 55 meV

# *Diffusion models (Preliminary result)*



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# *Theoretical approach*

**MOLECULAR DYNAMICS SIMULATIONS**

↓

Treat simulations like an experiment

| [system setup] | → | [equilibration] | → | [simulation run] | → | [output] |
|---|---|---|---|---|---|---|
| sample selection | | sample preparation | | property average | | data analysis |

↓

How MD does this.. ?

- MD is the solution of the classical equations of motion for atoms and molecules to obtain the time evolution of the system.

Integrate these equations of motion to obtain continuous trajectories.

↓

**TRAJECTORIES**

**MOLECULAR DYNAMICS SIMULATIONS**

↓

**TRAJECTORIES**

↓

**VACF**
**Velocity Auto Correlation Function**

↓ $VACF(\tau) = < v(t).v(t+\tau) >$

**g(ω):**
**vDOS Density of states**

↓

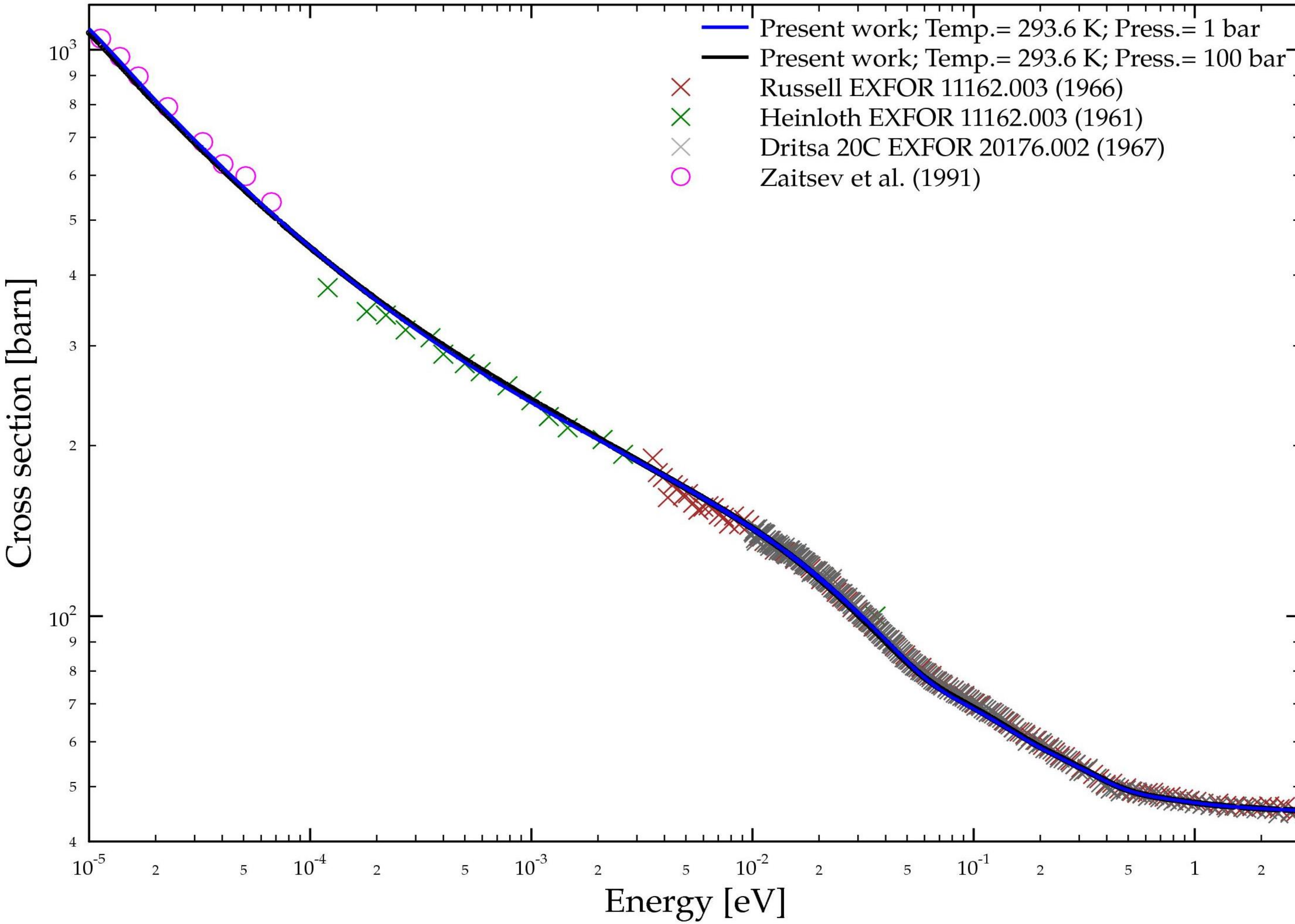
**LEAPR (NJOY): Uses g(ω) as an input to generate S(α, β)**

↓

**S(α, β)**

\# CAB approach

# *Result (Total cross-section)*



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# *Conclusions and perspectives*

- DDXS of light water depends only on the temperature and is independent of the pressure (Inference from the experiments).

- To verify the above claim, a similar calculation was performed using MD simulations and the results are same (pressure independence).

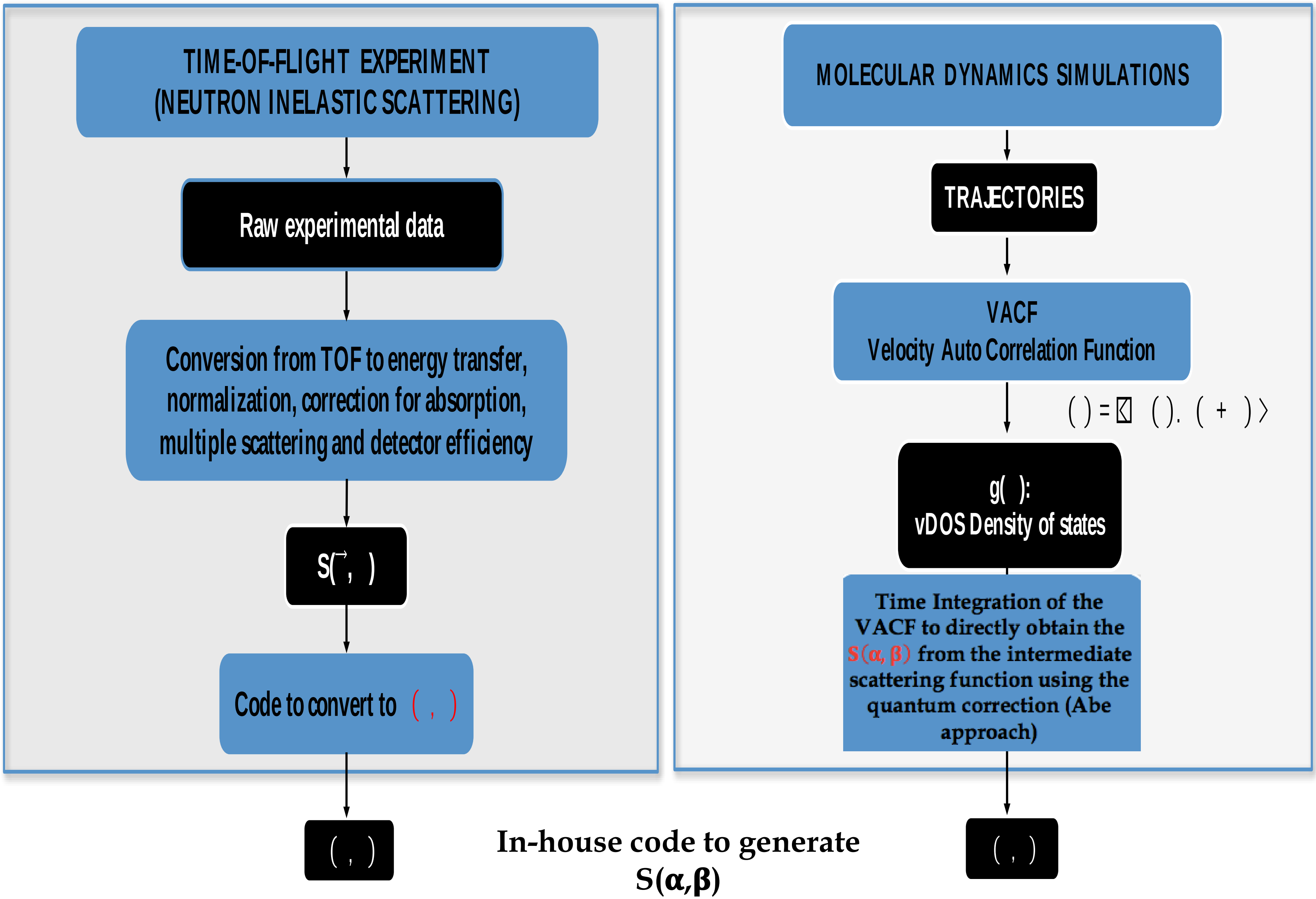
- Comparison of the DDXS of the experimental and MD data.

- Transform the evaluated $S(\alpha, \beta)$ in a form suitable for reactor and criticality safety applications (MORET, TRIPOLI ..)

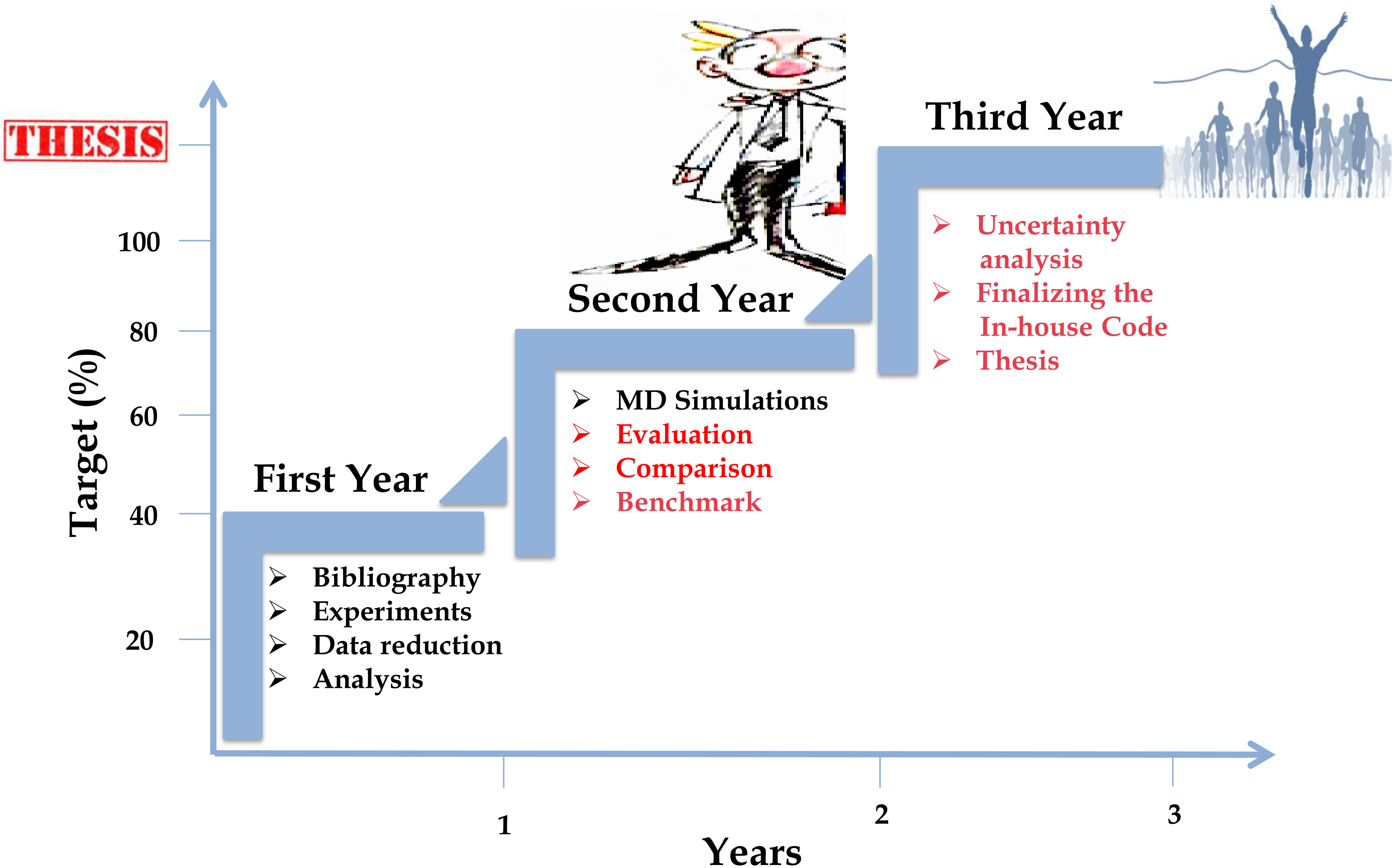
- Calculation of the uncertainties from the experimental data, especially study of the systematic and the statistical uncertainty.

- A parallel methodology is under development which can directly calculate the $S(\vec{q},\omega)$ from the VACF, thus avoiding the use of LEAPR module of NJOY.

# *Conclusions and perspectives*



# Timeline




**Thank you for your attention!**