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Impact of Thermal Scattering Law (TSL) for Light Water on the French Plutonium Temperature Effect Experimental Program

WONDER 2018: 5th International Workshop On Nuclear Data Evaluation for Reactor Applications

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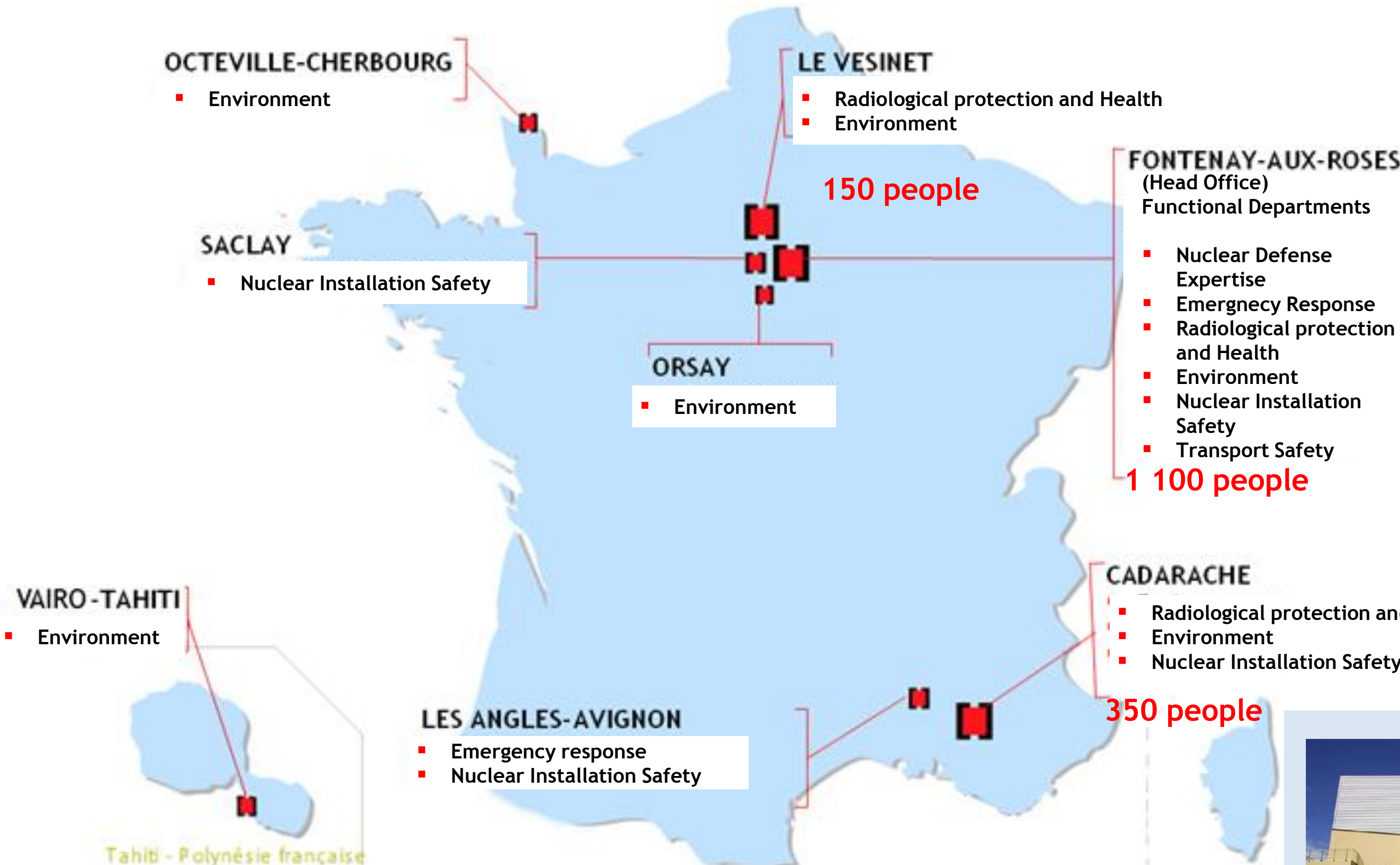
1800 personnel at 8 sites



Agenda

- **Motivation**
- **Presently available TSL for light water**
- **French Plutonium Temperature Effect Experimental Program**
- **GAIA: SAB module**
- **Results**
- **Conclusions and Perspectives**

"Neutron Man"

* Neutron Scattering: A Primer by Roger Pynn

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Motivation

- Light water is the most common moderator in thermal nuclear reactors like Pressurized Water Reactors (PWRs) and nuclear critical systems.

- Reliable thermal scattering cross section data, often termed as Thermal Scattering Law (TSL) or S(α,β) for light water is important for reactor physics and criticality safety applications.

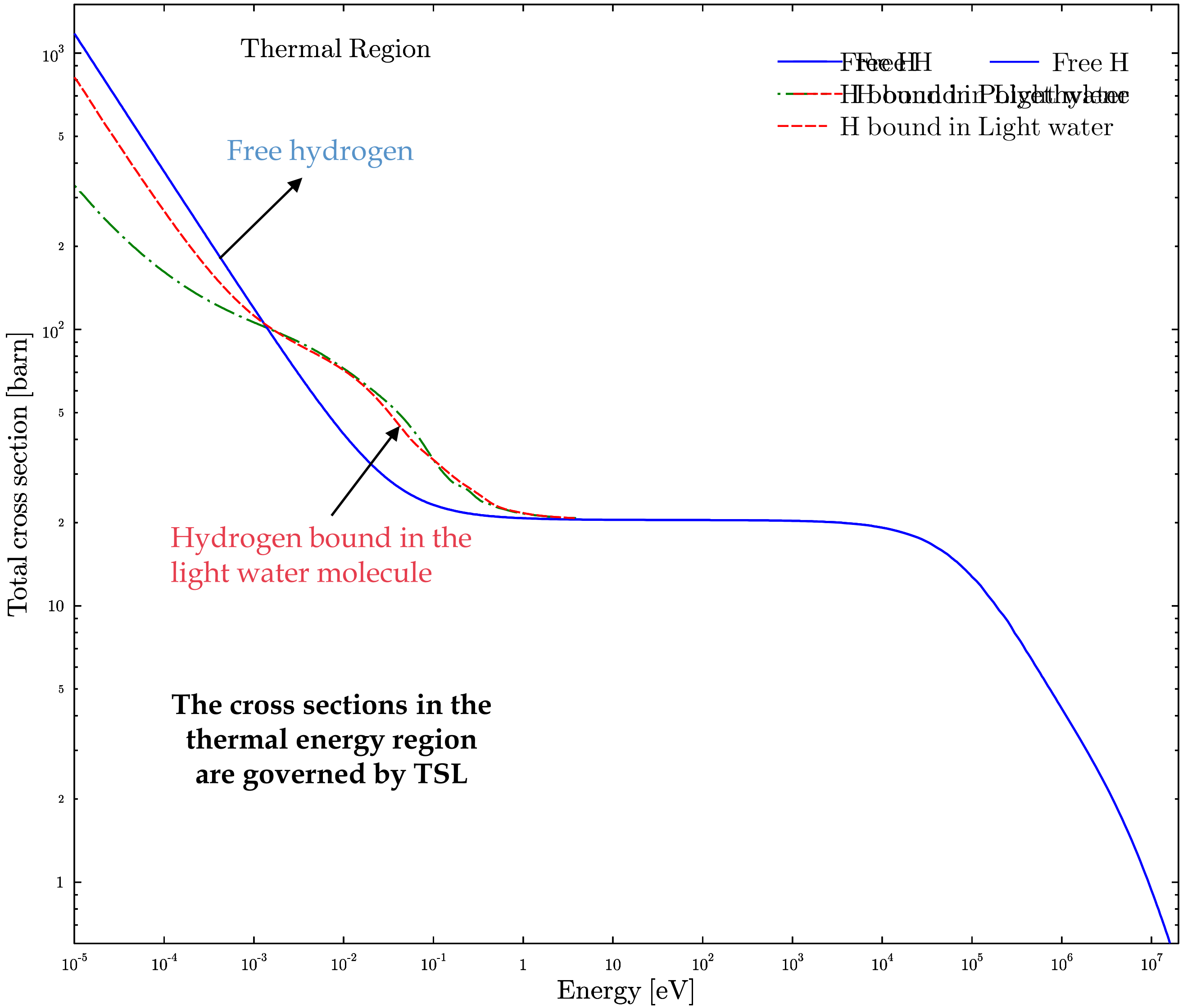
- All operating nuclear thermal power reactors in France are PWRs and they operate with light water at high temperature and pressure, i.e. 600 K and 150 bar.

- Presently, TSLs are considered only **temperature dependent**.

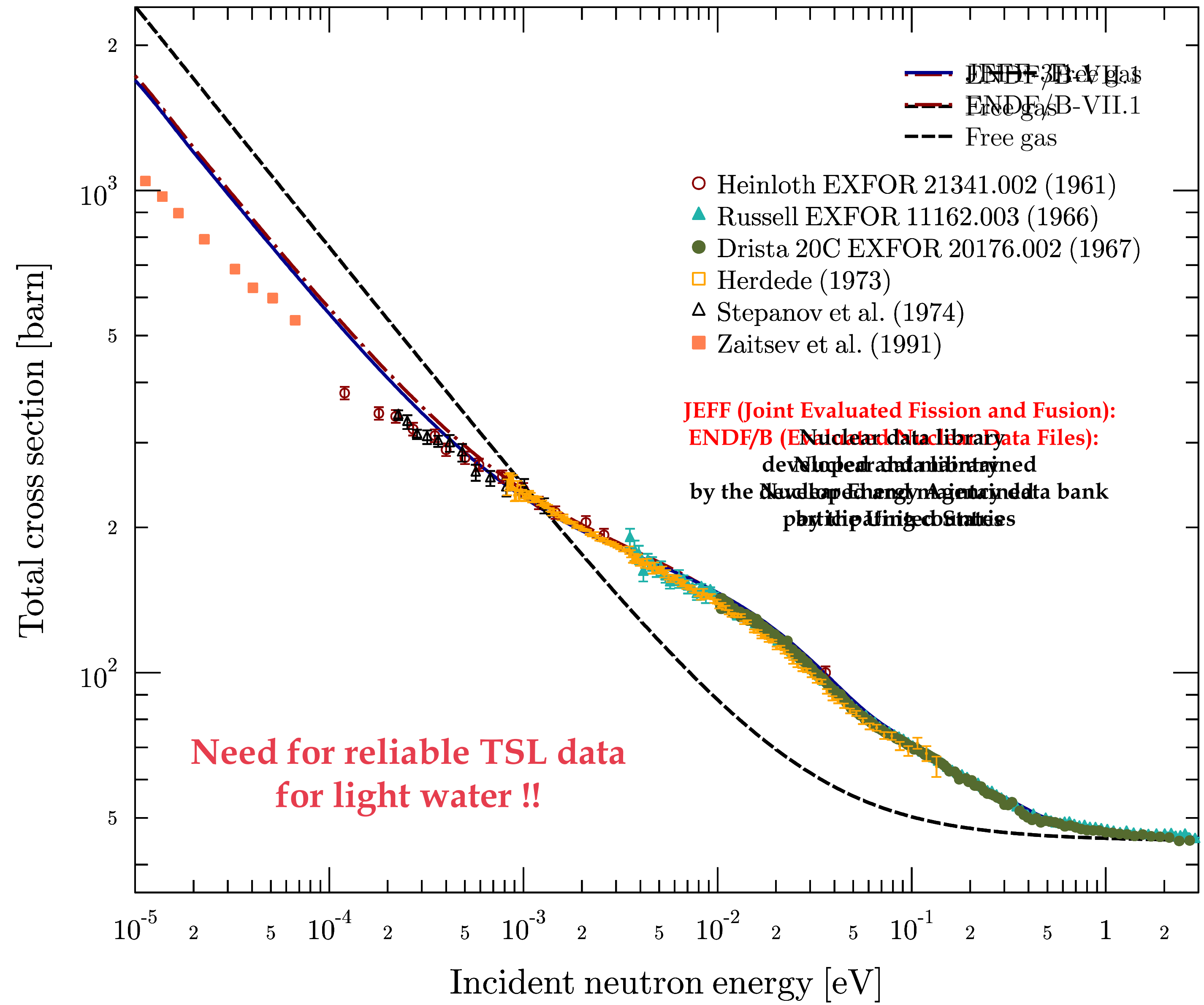
- Pressure is not taken into account when generating TSLs.

- TSL for light water at appropriate temperature is necessary to observe various physical phenomenon (For instance, **French Plutonium Temperature Effect Experimental Program**).

Motivation



Total cross section of hydrogen at 293.6 K.


Total cross section [barn]
Incident neutron energy [eV]
ENDF/B-VII.1
Free gas
Heinloth EXFOR 21341.002 (1961)
Russell EXFOR 11162.003 (1966)
Drista 20C EXFOR 20176.002 (1967)
Herdede (1973)
Stepanov et al. (1974)
Zaitsev et al. (1991)
JEFF (Joint Evaluated Fission and Fusion):
Need for reliable TSL data
for light water !!

Motivation

Goal : Improve the thermal scattering cross section for light water at both normal and operating conditions of nuclear power reactors.

How??

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

TOF Experiment

Perform Molecular Dynamics (MD) simulations

Codes: GROMACS/PolarisMD

Generation of new thermal scattering law for light water

Perform Time-of-Flight (TOF) experiments

Facility: ILL (France), SNS (US)

MD Simulations

*Neutron Man (Neutron Scattering: A Primer by Roger Pynn)

Theoretical formalism

- The double differential cross-section for neutrons with incident energy **E**, secondary energy **E′**, μ is the neutron scattering cosine and scattering angle Ω is related to S(α,β) by :

$$\frac{d^2\sigma}{d\Omega dE'}(E \to E', \Omega) = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}S(\alpha,\beta)$$

S(α, β)

Thermal Scattering Law

$$\alpha = \frac{E + E' - 2\mu\sqrt{EE'}}{Ak_BT} \qquad \beta = \frac{E' - E}{k_BT}$$

Momentum transfer *Energy transfer*

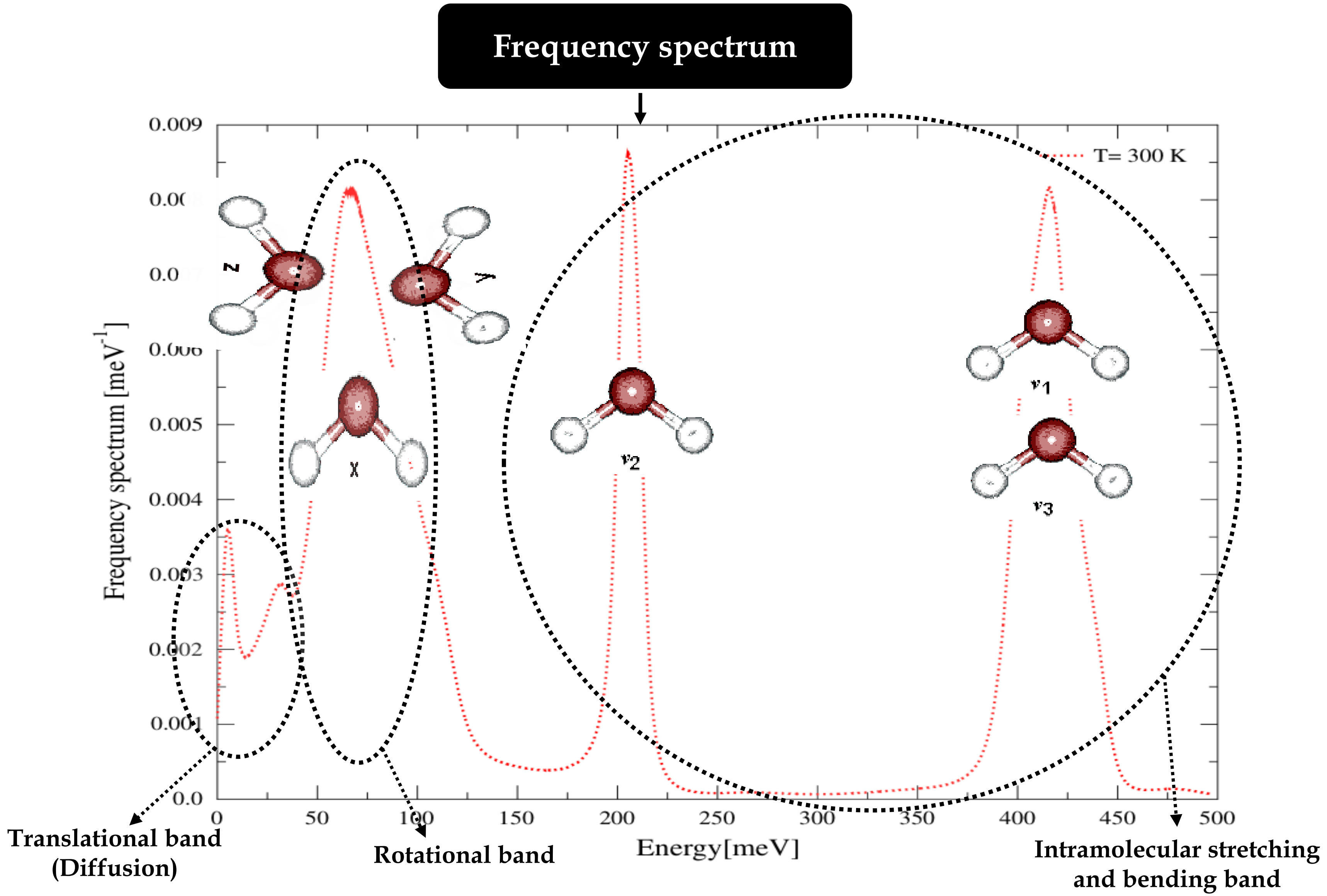
$$S(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{\iota\beta\hat{t}}\, e^{-\gamma(\hat{t})} d\hat{t}$$

$$\gamma(\hat{t}) = \alpha\int_{-\infty}^{+\infty} P(\beta)(1 - e^{-\iota\beta\hat{t}})\, e^{-\beta/2} d\beta$$

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)}$$

← ***Frequency spectrum / Density of states***

Frequency spectrum of light water



[J.I. Marquez Damian et al., J. Chem. Phys. 139, 024504 (2013)]
https://www-nds.iaea.org/index-meeting-crp/CM-THSC-2015/

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Presently available TSL for light water

Based on TOF Experiments

Using neutrons to measure the structural properties of light water.

Based on MD simulations

Using classical water models to indirectly obtain the structural properties of light water.

- LEAPR module of the NJOY code which uses numerous approximations (Incoherent and Gaussian to name a few).
- **$S(\alpha, \beta)$** is deduced from an **analytical function $\rho(\omega)$, which is the frequency spectrum** (most common approach used presently)

$\rho(\omega)$ → **LEAPR (NJOY)** → **$S(\alpha, \beta)$**

TSL libraries for light water

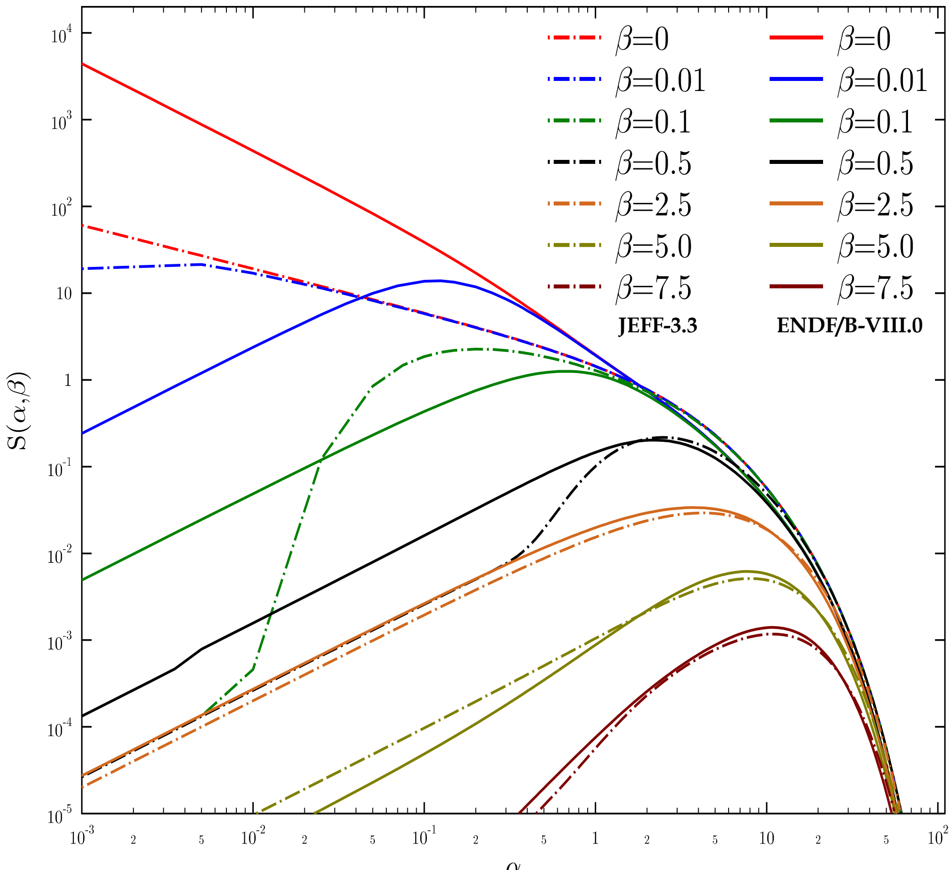
ENDF/B-VII.1, JEFF-3.1, JEFF-3.2, **JEFF-3.3**

Based on TOF experiments performed by Haywood and Page in the 60s

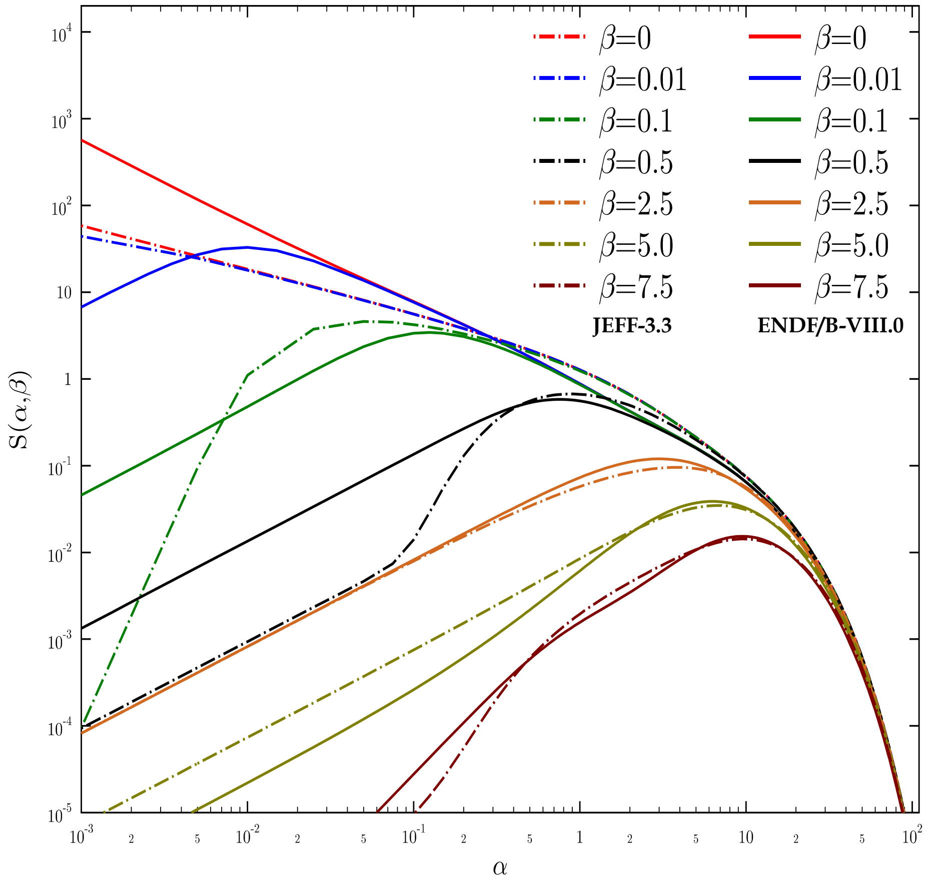
ENDF/B-VIII.0

Based on MD simulations using the GROMACS code And TIP4P/2005f water model

S(a,β) for light water: JEFF-3.3 & ENDF/B-VIII.0

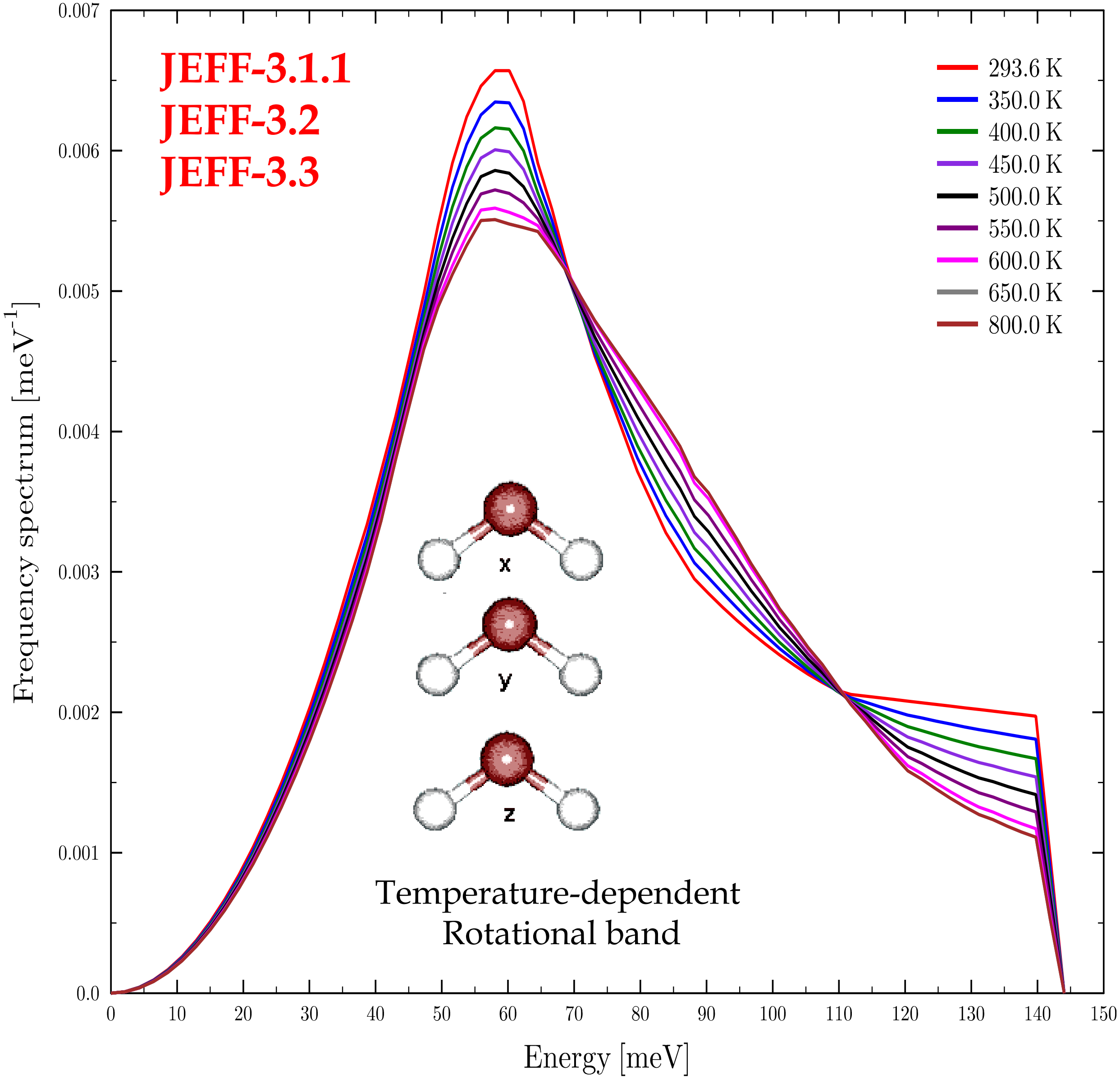


TSL at 293.6 K as a function of α for different β values



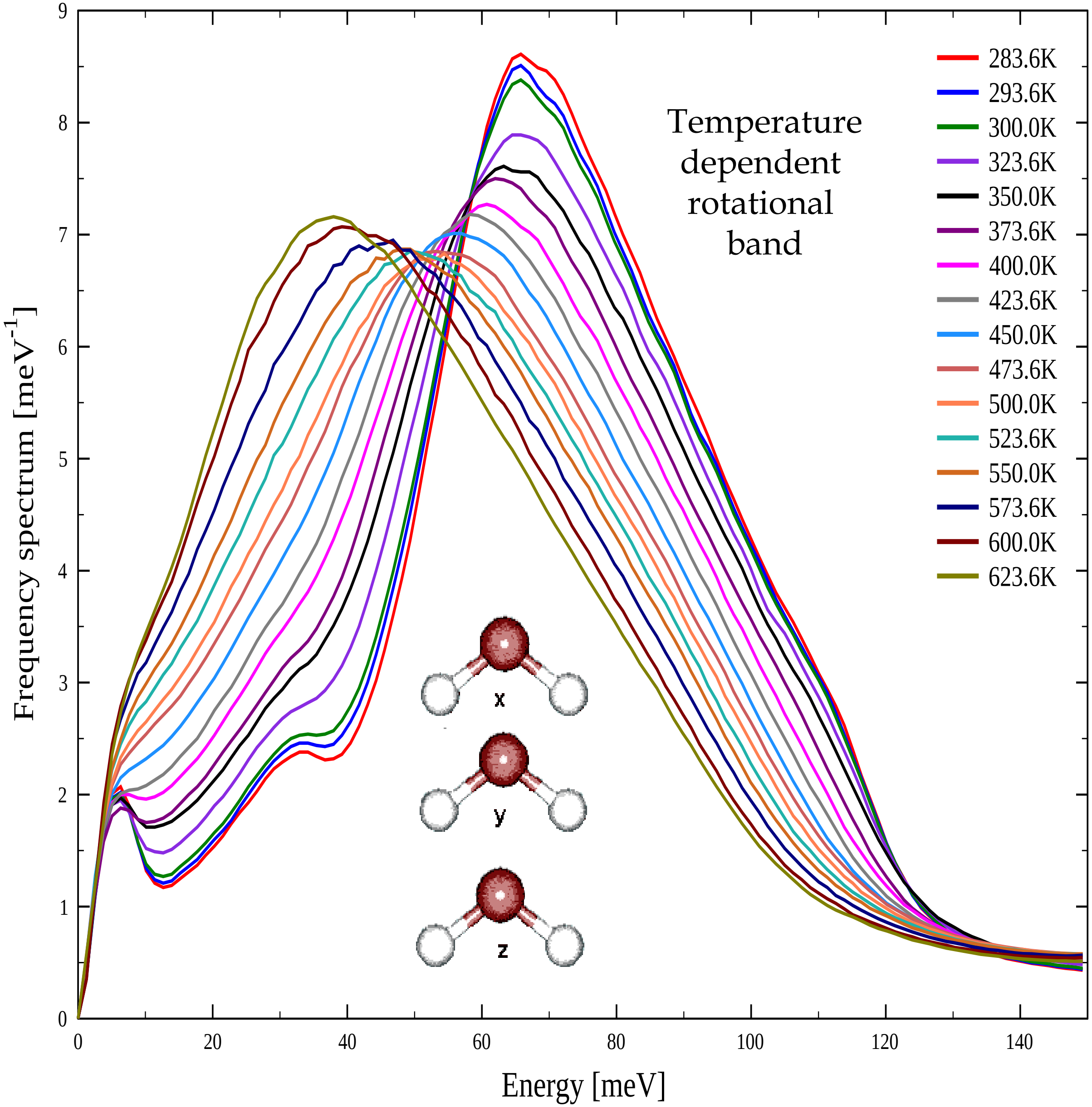
TSL at 573.6 K as a function of α for different β values

TSL for light water in JEFF-3.3 library

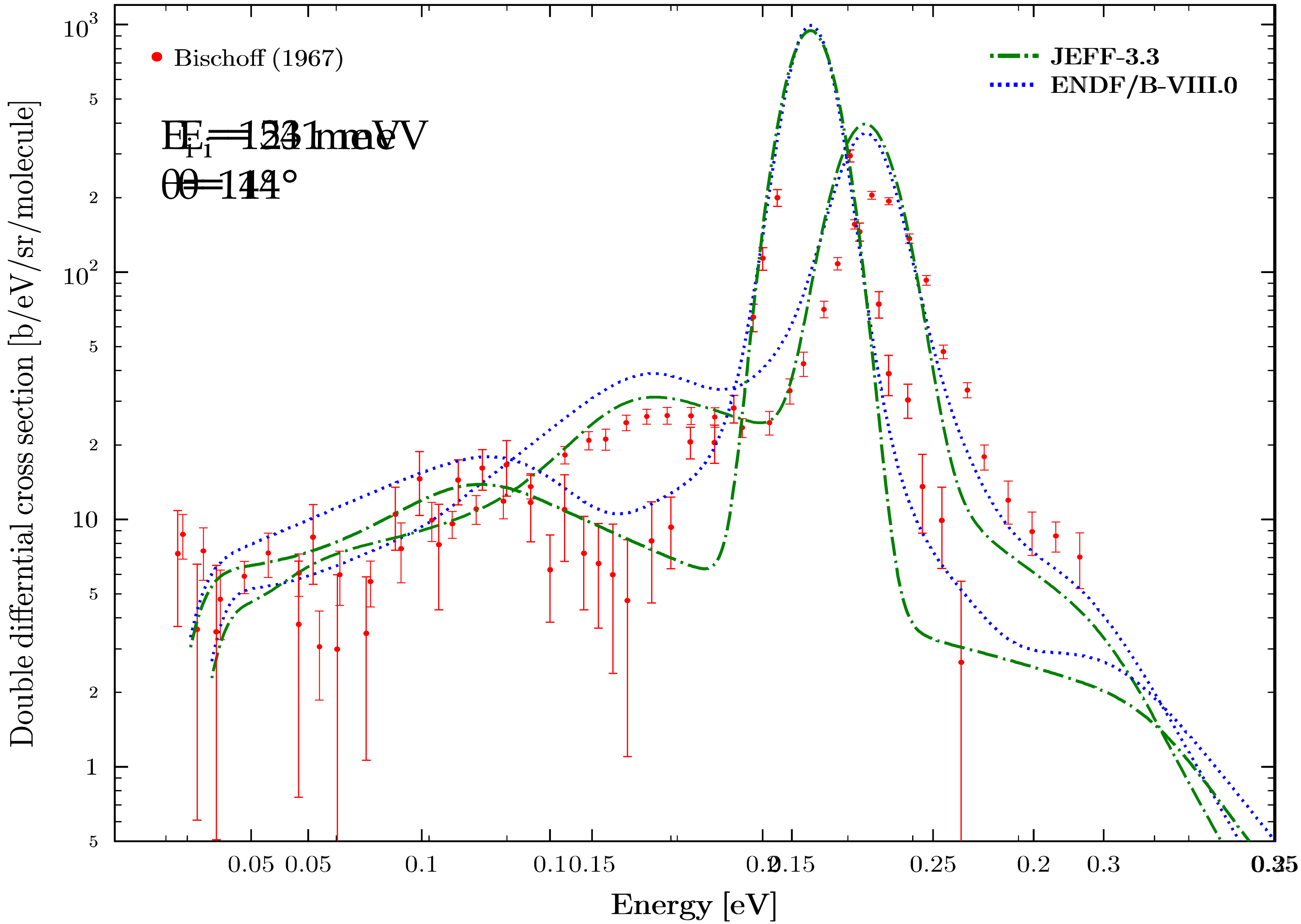


- **Frequency spectrum derived from TOF experiments performed by Haywood and Page.**
- **Frequency spectrum limited upto the rotation band.**
- **Discrete oscillator to model bending (205 meV) and stretching modes (436 meV).**
- **Free gas model to describe diffusion.**

TSL for light water in ENDF/B-VIII.0 library

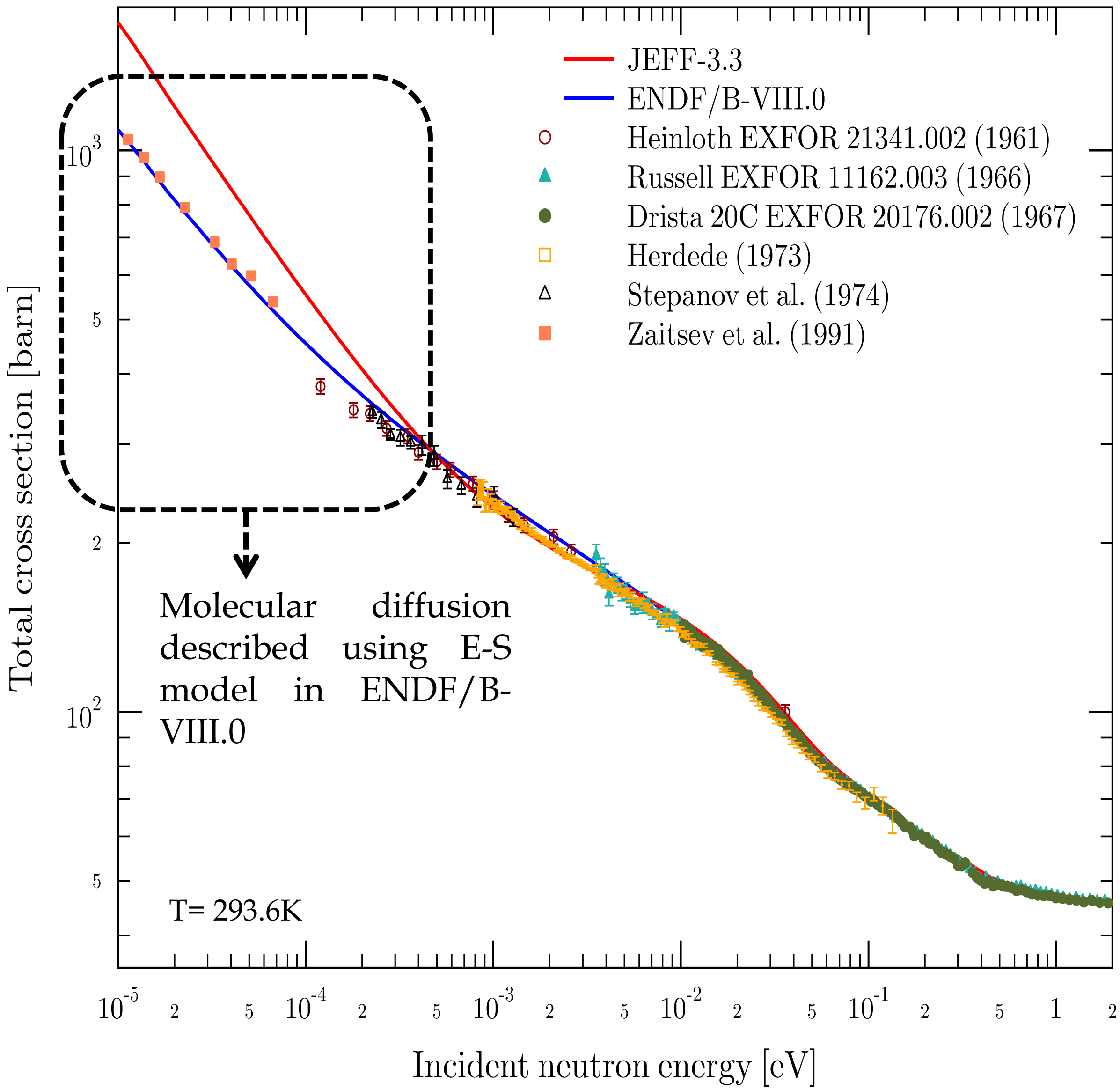


- **Frequency spectrum derived from MD simulations using GROMACS and TIP4P/2005f water potential**
- **Frequency spectrum limited up to the rotation band.**
- **Discrete oscillator to model bending (205 meV) and stretching modes (436 meV).**
- **Egelstaff- Schofield (E-S) model to describe diffusion.**



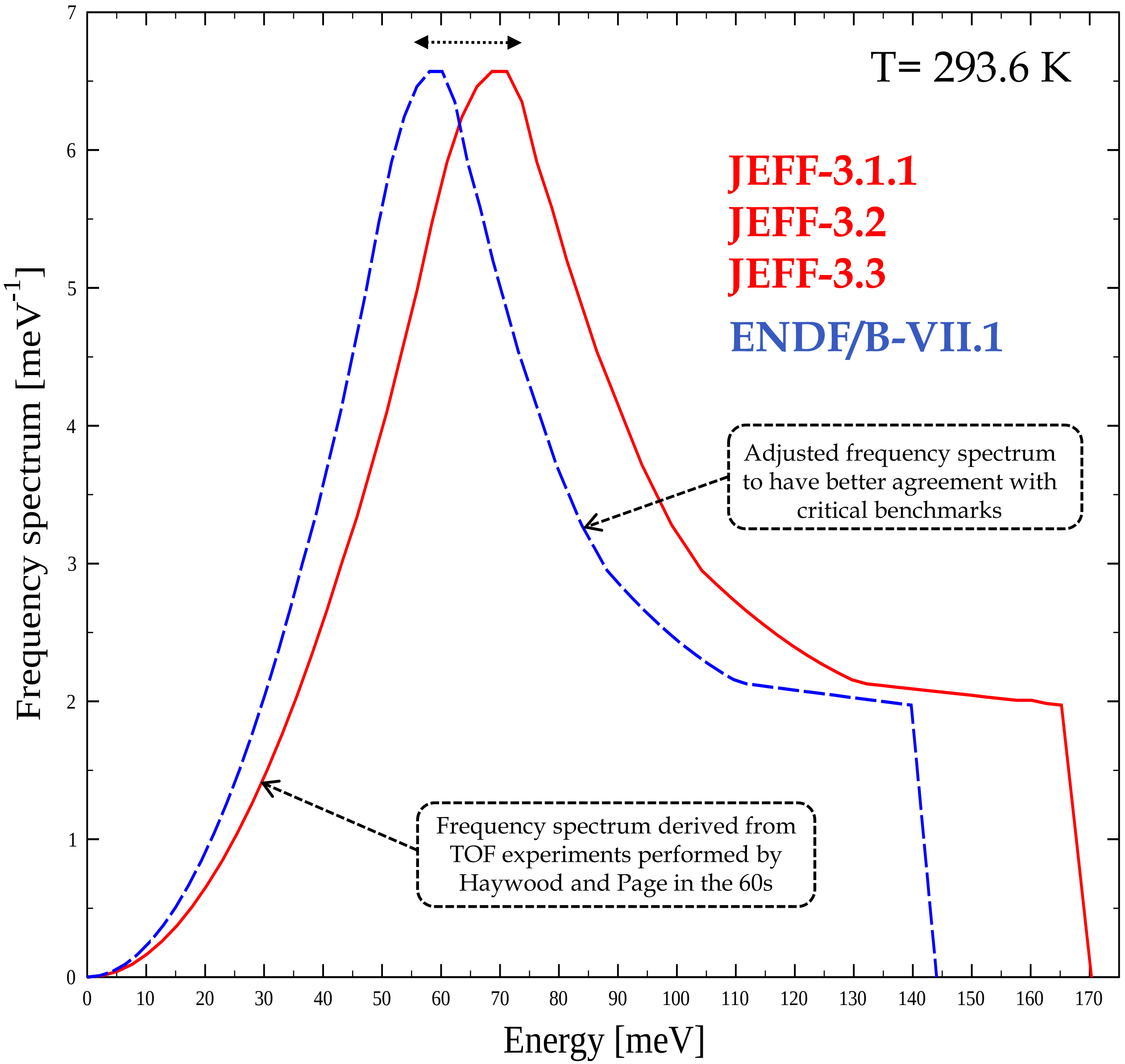
Both JEFF-3.3 and ENDF/B-VIII.0 TSL evaluation has its own features
ENDF/B-VIII.0 reproduces the quasi elastic peak well

Total cross section: JEFF-3.3 & ENDF/B-VIII.0



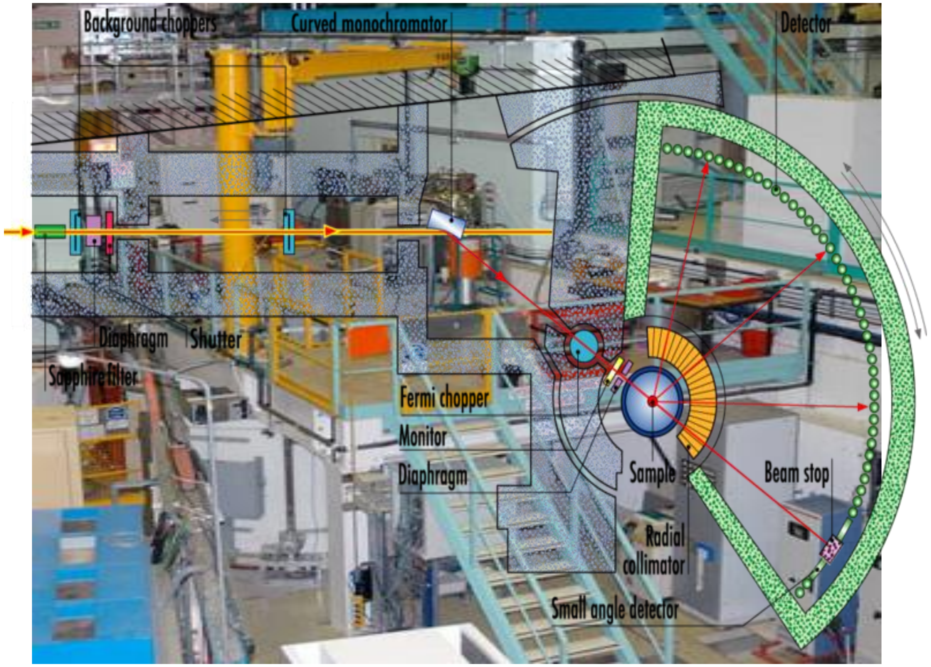
- **JEFF-3.3:** **Agreement with the experimental total cross section in the thermal energy region.**
- **ENDF-B/VIII.0:** **Agreement with the experimental total cross section in the thermal energy region as well as low energy region.**
- **Discrete oscillator to model bending and stretching modes in both the evaluations.**
- **E-S diffusion model improves the agreement in the low energy region**

Frequency spectrum of light water: JEFF-3.1.1 & ENDF/B-VII.1

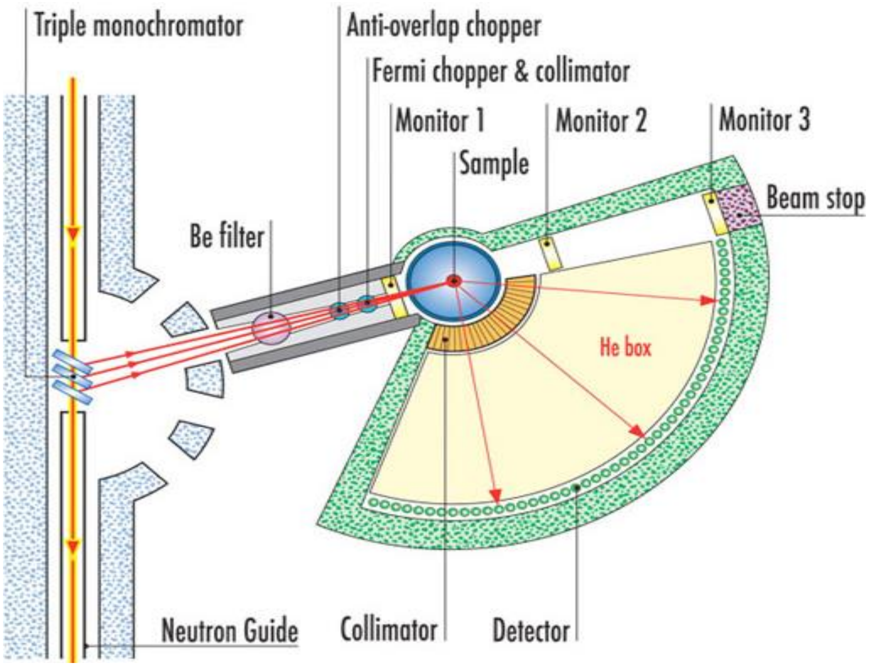


- **Frequency spectrum in ENDF/B-VII.1 was adjusted to have better agreement with thermal critical benchmarks.**
- **The position of the rotation band was shifted from 70 to 60 meV in order to achieve this goal.**
- **Implies nuclear critical systems as well as nuclear reactors can be sensitive to the position of the rotation band.**

Experimental approach: TOF Experiment



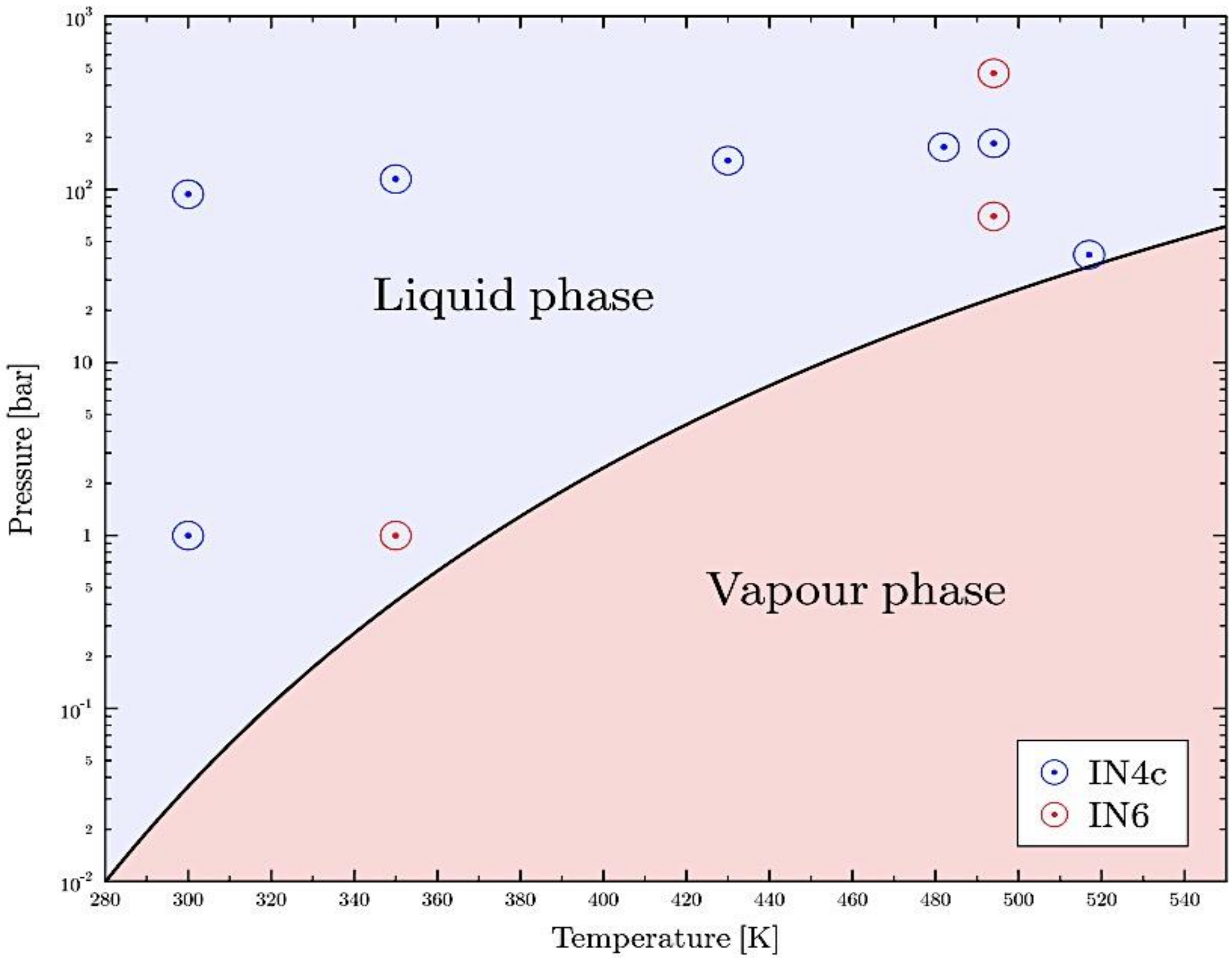
IN4c spectrometer



IN6 spectrometer

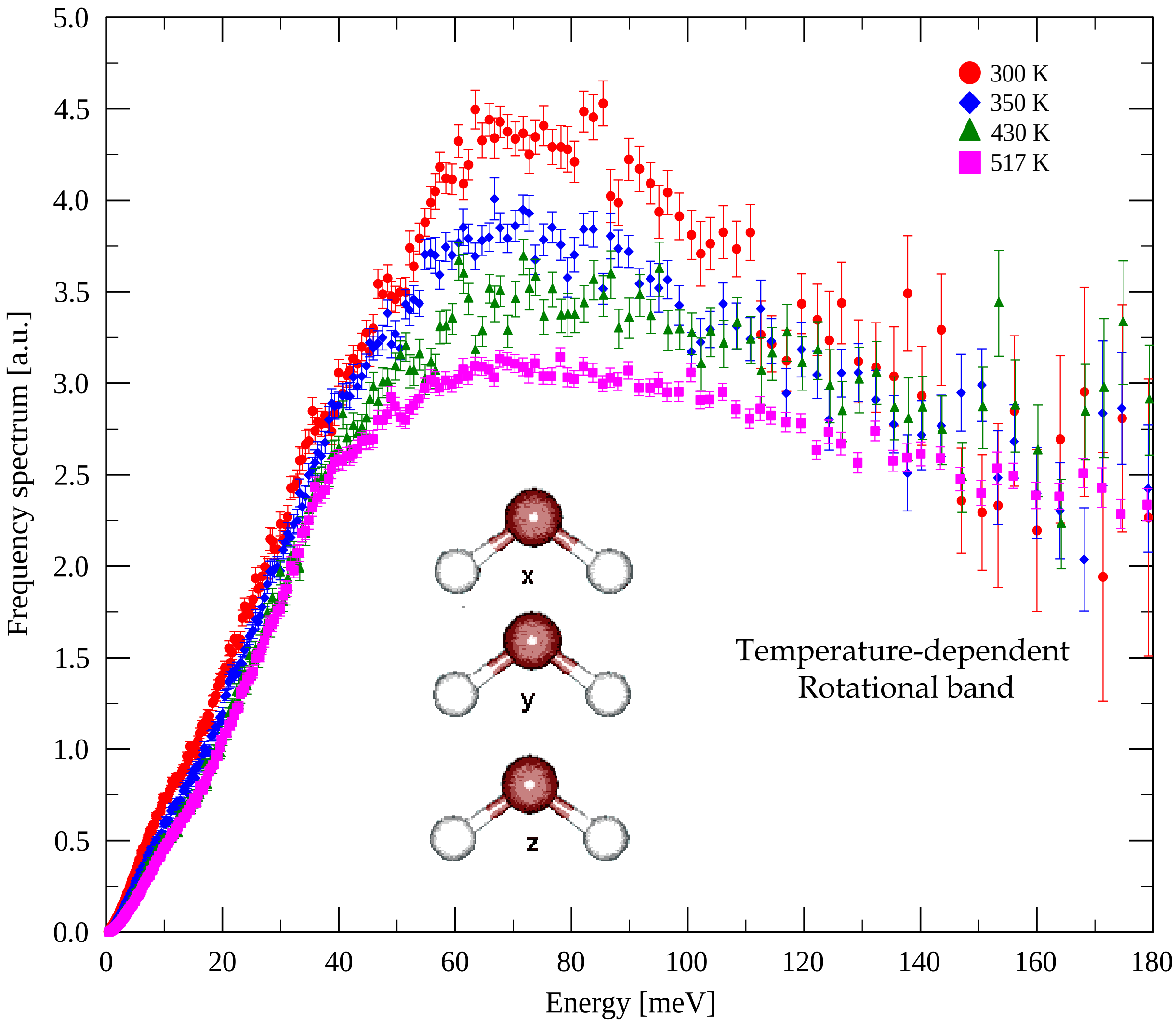
- Incident neutron energy= 14.2 meV
- Scattering angular range θ = 15°-120°
- Resolution = 4%

- Incident neutron energy= 3.15 meV
- Scattering angular range θ = 15°-110°
- Resolution = 2%



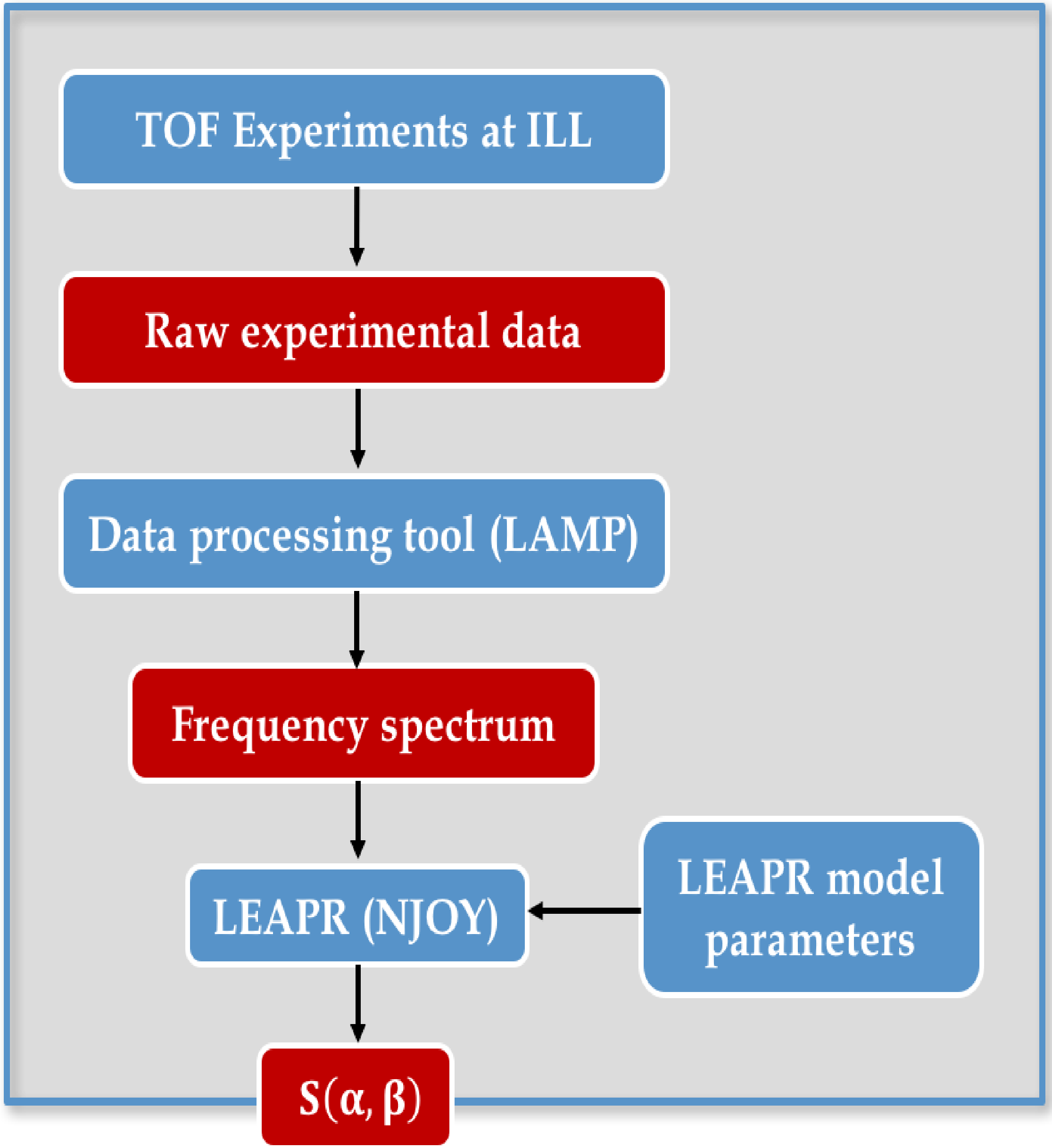
Thermodynamic states (experimental conditions) of light water for the data measurement at the ILL, Grenoble.

Temperature dependence on the frequency spectrum



- Frequency spectrum is limited up to the rotation band.
- The energy of the rotation band is peaked around 75 meV.
- Frequency spectrum is dependent on the temperature of light water.
- The energy of the rotation band does not significantly shifts with increasing temperature.

Generation of new TSL evaluation: $S(\alpha, \beta)_{ILL}$



- Frequency spectrum obtained from IN4c TOF spectrometers used to generate TSL evaluation.
- Frequency spectrum limited up to the rotation band.
- Two discrete oscillators to model the bending and the stretching modes.
- Egelstaff - Schofield model to describe molecular diffusion.

MOLECULAR DYNAMICS SIMULATIONS

↓

- Choice among available molecular dynamics codes?

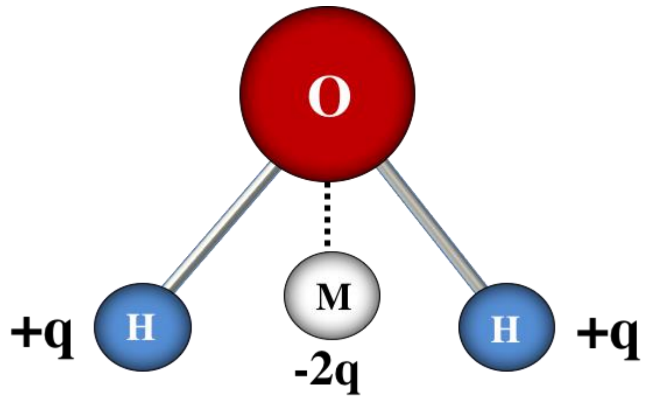
GROMACS, DL_POLY, LAMMPS, PolarisMD

- Choice among different water models suitable at high temperature and pressure?

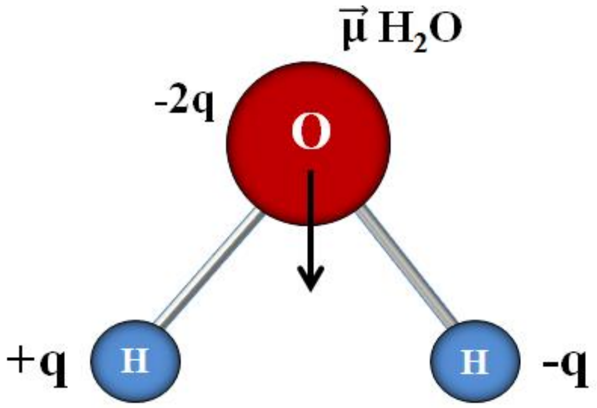
SPC, TIP4P, TIP4P/2005f, TCPE

A variety of classical water models exists based on:

- Treatment of covalent bonds: Flexible or rigid.
- Water models might or not might not take into account the polarizable effect.
- Hydrogen bonding taken into account or not.



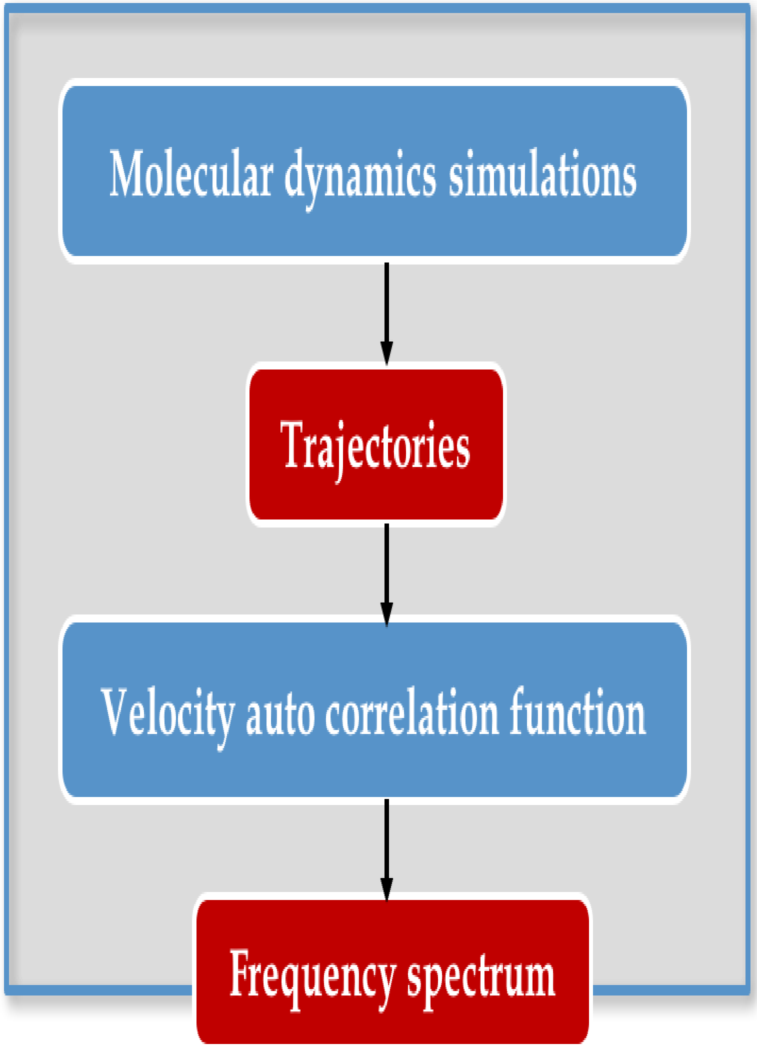
Flexible non-polarizable TIP4P/2005f water model



Rigid polarizable TCPE water model with explicit hydrogen bonding

- Adding flexibility to a water model might not necessarily improve its overall performance.
- Polarizable models respond to fluctuations in the electric field due to molecular motion.
- Polarizable model is a promising solution for reactor physics applications where the reactors operate at high temperatures and pressure.
- TCPE water model though being a rigid model accurately reproduces the experimental self-diffusion coefficient of water for a series of high temperatures.

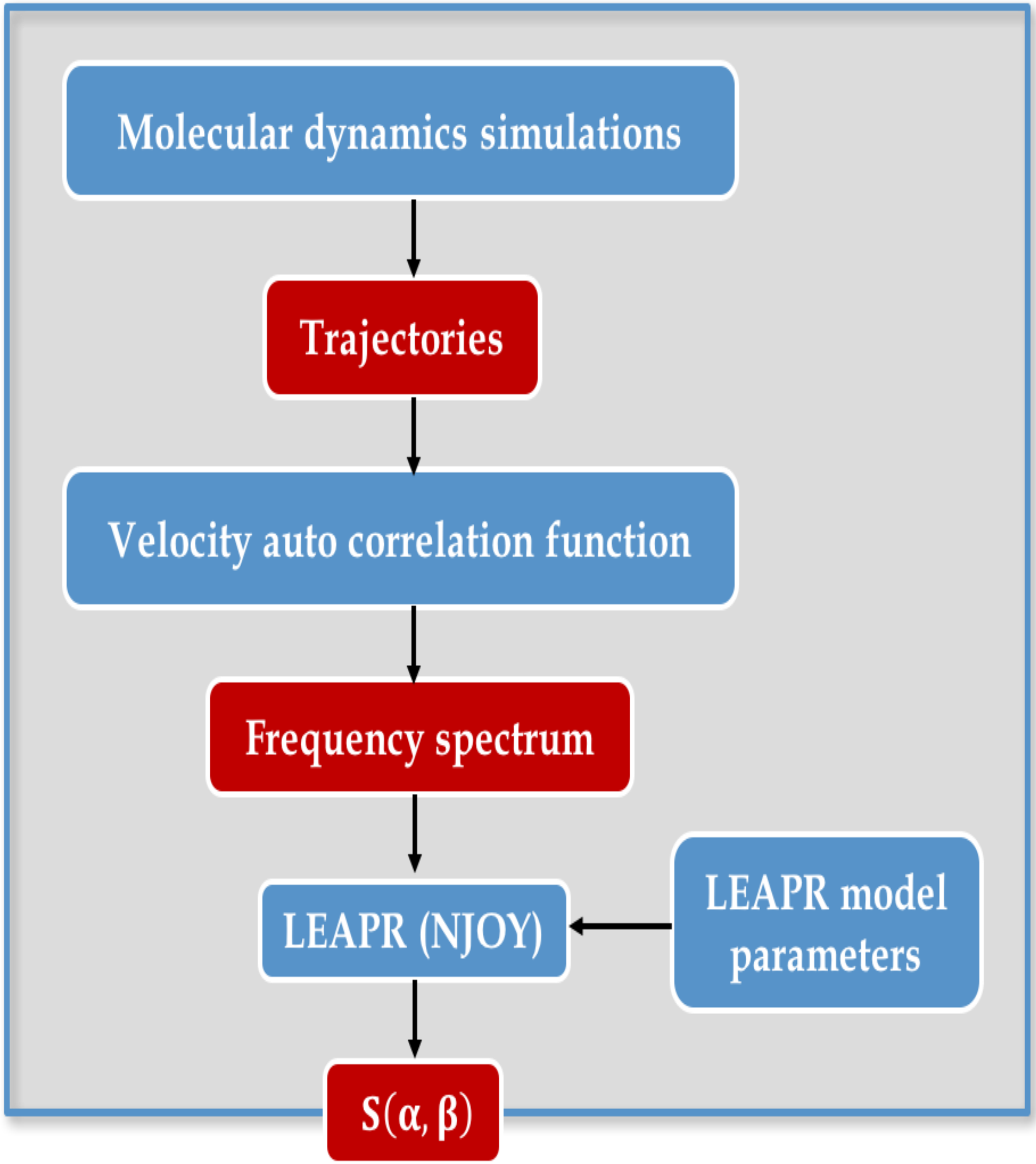
MD simulations using TCPE water model



Temperature	Pressure
(K)	(bar)
283.6	1
293.6	1
295.0	1
323.6	1
350.0	1
373.6	3
400.0	8
423.6	10
450.0	20
473.6	20
500.0	30
523.6	50
550.0	75
573.6	100
600.0	150

Simulation with TCPE potential at different temperatures and pressures

Generation of new TSL evaluation: $S(\alpha, \beta)_{TCPE}$



- Frequency spectrum obtained from TCPE water models.
- Frequency spectrum limited up to the rotation band (Rigid model)
- Two discrete oscillators to model the bending and the stretching modes.
- Egelstaff - Schofield model to describe molecular diffusion.

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Plutonium nitrate temperature effect experimental program

- Series of experiments were conducted by the IRSN at the 'Apparatus B' facility of CEA VALDUC (May-July 2007).

- Experiments at different temperatures to study the positive temperature effect for diluted plutonium solutions.

22 °C **(295.15 K)** : PU-SOL-THERM-038 & 28 °C **(301.15 K)**: PU-SOL-THERM-039

Experimental Setup

- Positive temperature Effect was observed experimentally between this temperature range.

- Monte Carlo transport simulations on these benchmarks showed negative temperature effect between these two temperatures.

??

- A totally different behaviour is observed when using a slightly different temperature for the TSL data.

Wim Haeck et al., "The Plutonium Temperature Effect Experimental Program", International Conference on the Physics of Reactors "Nuclear Power: A Sustainable Resource" Casino-Kursaal Conference Center, Interlaken, Switzerland, September 14- 19, 2008.

Wim Haeck et al., "Thermal scattering data and criticality safety", International Conference on the Physics of Reactors "Nuclear Power: A Sustainable Resource" Casino-Kursaal Conference Center, Interlaken, Switzerland, September 14-19, 2008.

Plutonium nitrate temperature effect experimental program

- TSL for light water at appropriate temperature is necessary to observe such a positive temperature effect.

- The closest temperature to this particular experiment in the libraries are:

JEFF-3.3: **293.6 K** and **350 K**

ENDF/B-VIII.0: **293.6 K** and **300 K**.

Need to generate TSL at appropriate temperatures to see the observed phenomenon !!

- Development of a module names as SAB in the *GAIA* ***nuclear data processing code***.

- Capability to generate TSL at any desired temperature.

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GAIA: An IRSN Nuclear data processing code

- GAIA: An IRSN in-house nuclear data processing code under development as a part of the INSIDER project (**I**nvestigations in **N**eutronics for **S**afety margin assessment based on data assimilation from **I**ntegral and **D**ifferential **E**xperimental **R**esearches) to process nuclear cross section data libraries with innovative features and processing capabilities.

- The project is taking shape in the form of two different codes:

  - GAIA 1: An NJOY wrapper to minimize the effort for the generation of nuclear data libraries in the ACE format to be used by the MCNP and MORET codes.

  - GAIA 2:

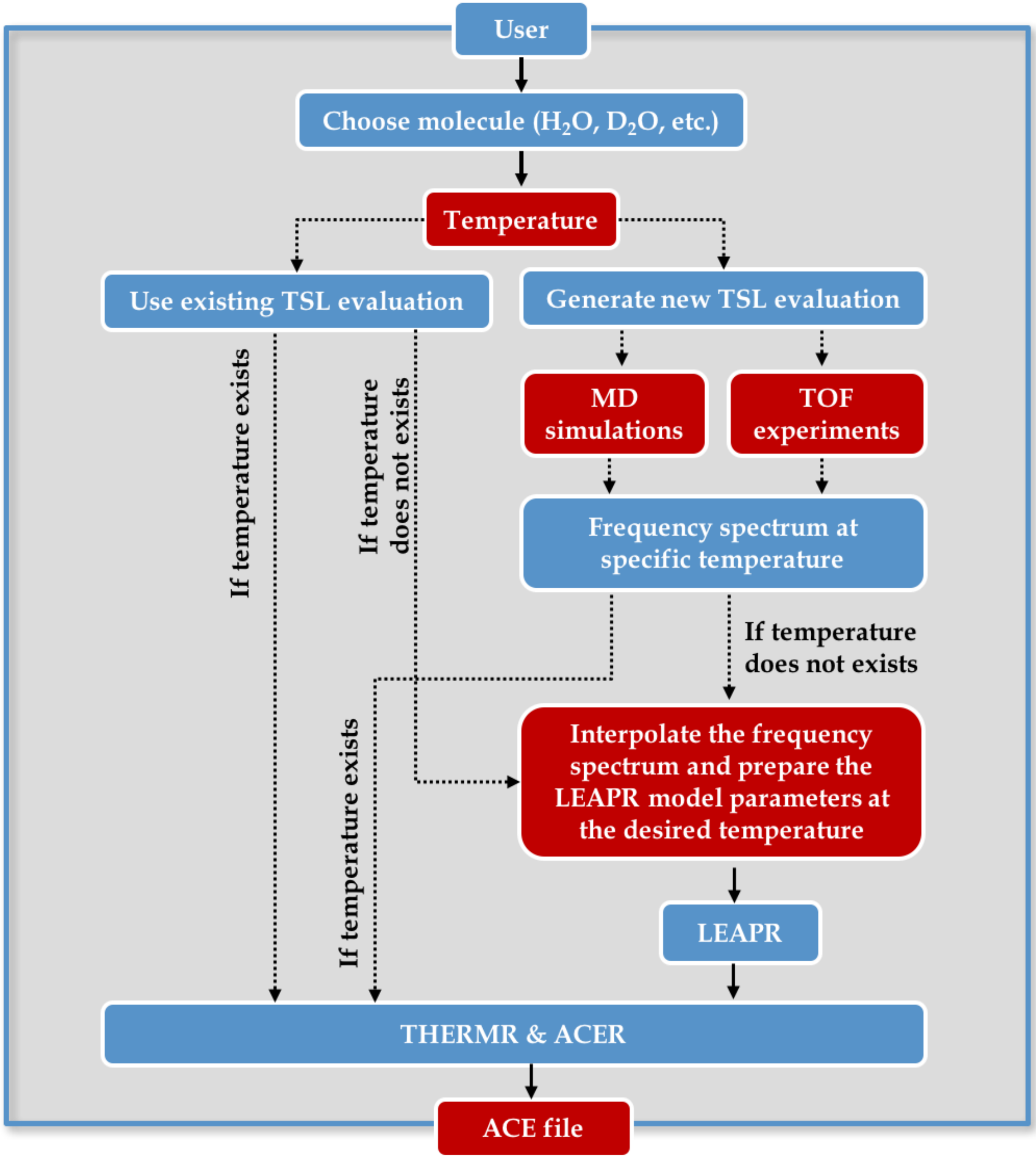
    - Implement the knowledge acquired on processing methodologies at IRSN to a nuclear data processing code.

    - Generate nuclear cross section data libraries in specific formats **for present and future neutronics tools**.

    - Generalized R-matrix formalism for resonance reconstruction.

    - Improved formalism in the **unresolved resonance region** (Ongoing PhD subject)

    - Implementation of the developments carried out during this PhD thesis, to generate TSL evaluations as a new module named as "**SAB module**".



- User has to specify the temperature of the TSL data.
- Can generate new TSL using existing TSL evaluations (JEFF-3.3 and ENDF/B-VIII.0)
- Can generate TSL using TOF experimental data or based on MD simulations.
- If the requested temperature exist, it directly runs THERMR & ACER to generate TSL in the ace format.
- If the requested temperature does not exist, it interpolates the frequency spectrum and other LEAPR model parameters and then runs LEAPR, THERMR & ACER to generate TSL in the ace format.

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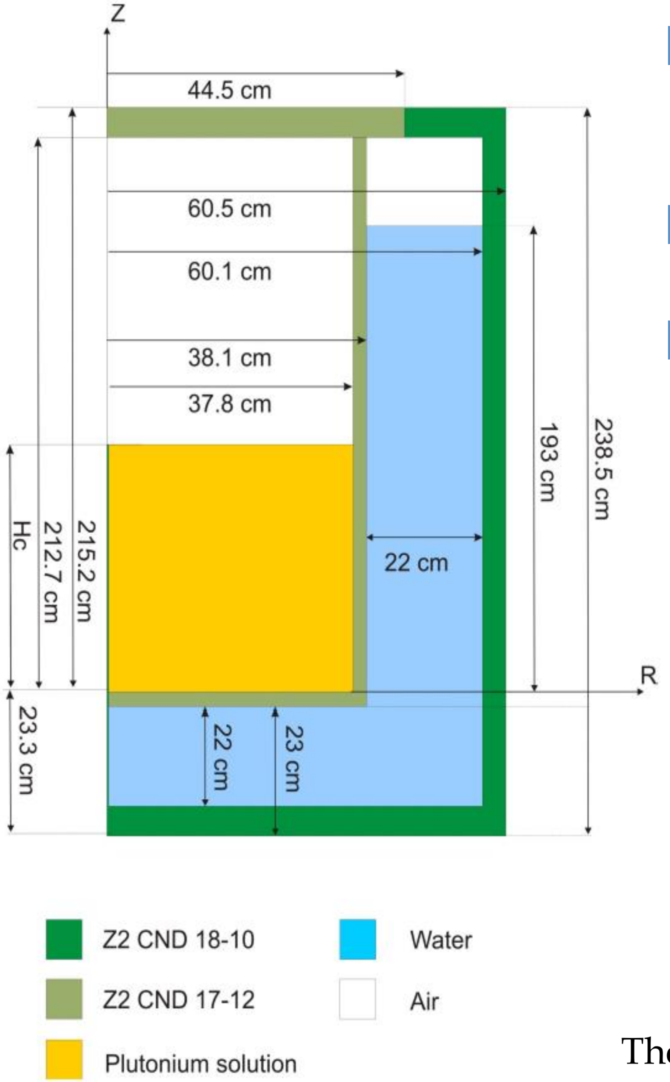
Plutonium nitrate temperature effect experimental program



MORET Benchmark model

- Monte Carlo continuous energy transport code, MORET 5.D.1 was utilized in this study.
- JEFF-3.3 library used as base for the cross-section data.
- The TSL evaluations (existing/newly generated) were replaced in the JEFF-3.3 evaluation for each specific calculation.

TSL evaluation for light water	Effective multiplication factor k_{eff} at T		Reactivity difference
	22 °C	28 °C	Δk_{eff} (pcm)
JEFF-3.3	0.99730 (293.6 K)	0.99696 (293.6 K)	-34
ENDF/B-VIII.0	0.99553 (293.6 K)	0.99586 (300.0 K)	+33
Interpolating ENDF/B-VIII.0 LEAPR model parameters using the SAB module	0.99570 (295.15 K)	0.99600 (301.15 K)	+30

The temperature of the TSL evaluation is given in parentheses. Monte Carlo standard deviation in all the calculations was set to 5 pcm.

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Conclusions and perspectives

- TSL behavior both at room and high temperatures and have been reviewed.

- Some discrepancies were observed between the frequency spectra utilized in the JEFF-3.3 (based on TOF experimental data) and ENDF/B-VIII.0 (based on classical MD simulations with TIP4P/2005f flexible non-polarizable water model) TSL evaluation.

- Accurate TSL data at required temperature is important to reproduce physical properties.

- The SAB module of the GAIA code was developed with capabilities to generate TSL libraries for light water at the desired temperature requested by the user.

- Positive temperature effect was observed when using correct temperature of light water TSL on the French Plutonium Temperature Effect Experimental Program.

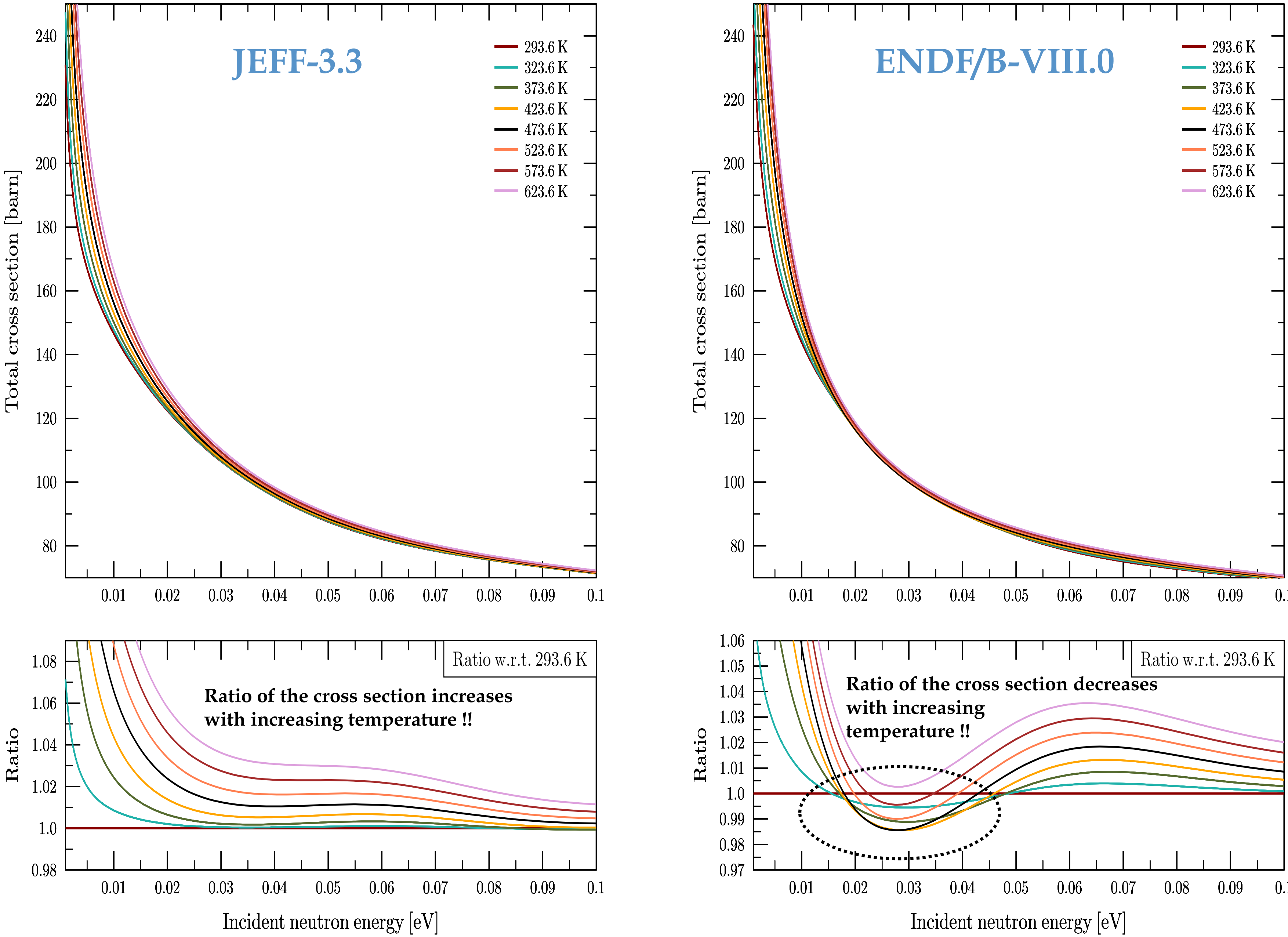
- Further tests on the French Plutonium Temperature Effect Experimental Program are required with TSL obtained using new ILL experiments and MD simulations.

Hey, I have a question.. !

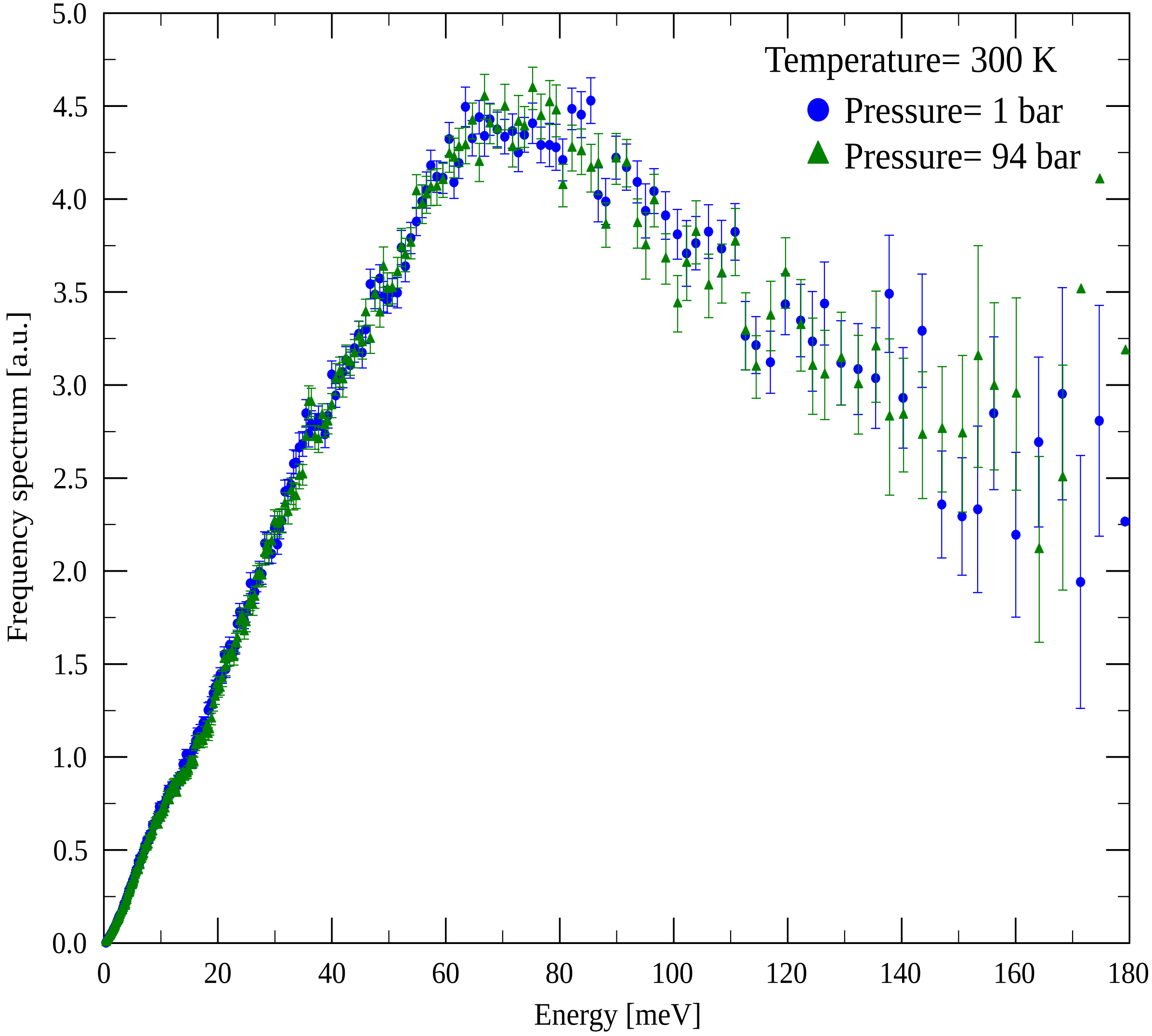
Yes ! Go ahead !!
Will be happy to answer!!

Hey, I have a backup.. !

Total cross section of light water: JEFF-3.3 & ENDF/B-VIII.0

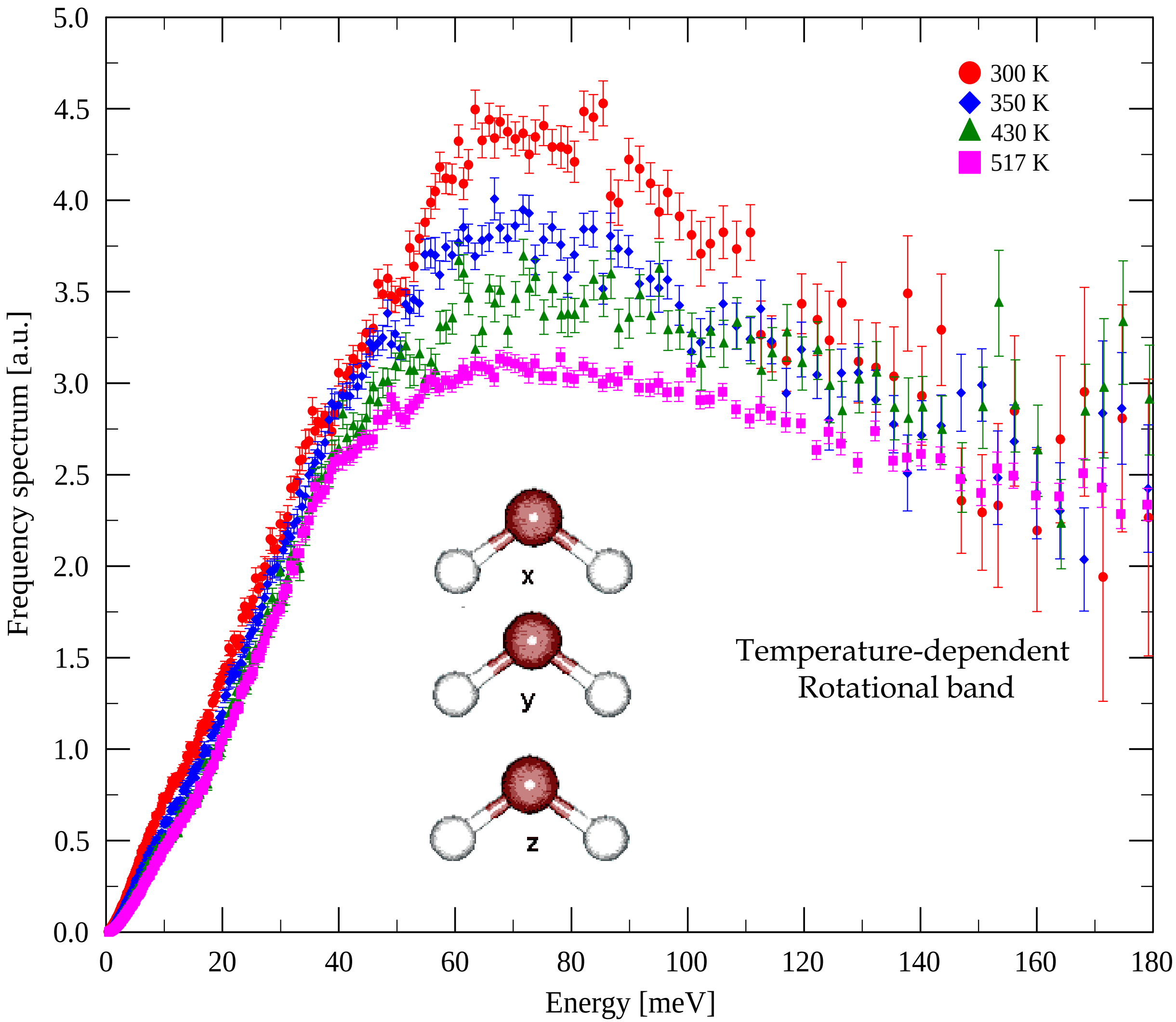


Pressure dependence on the frequency spectrum



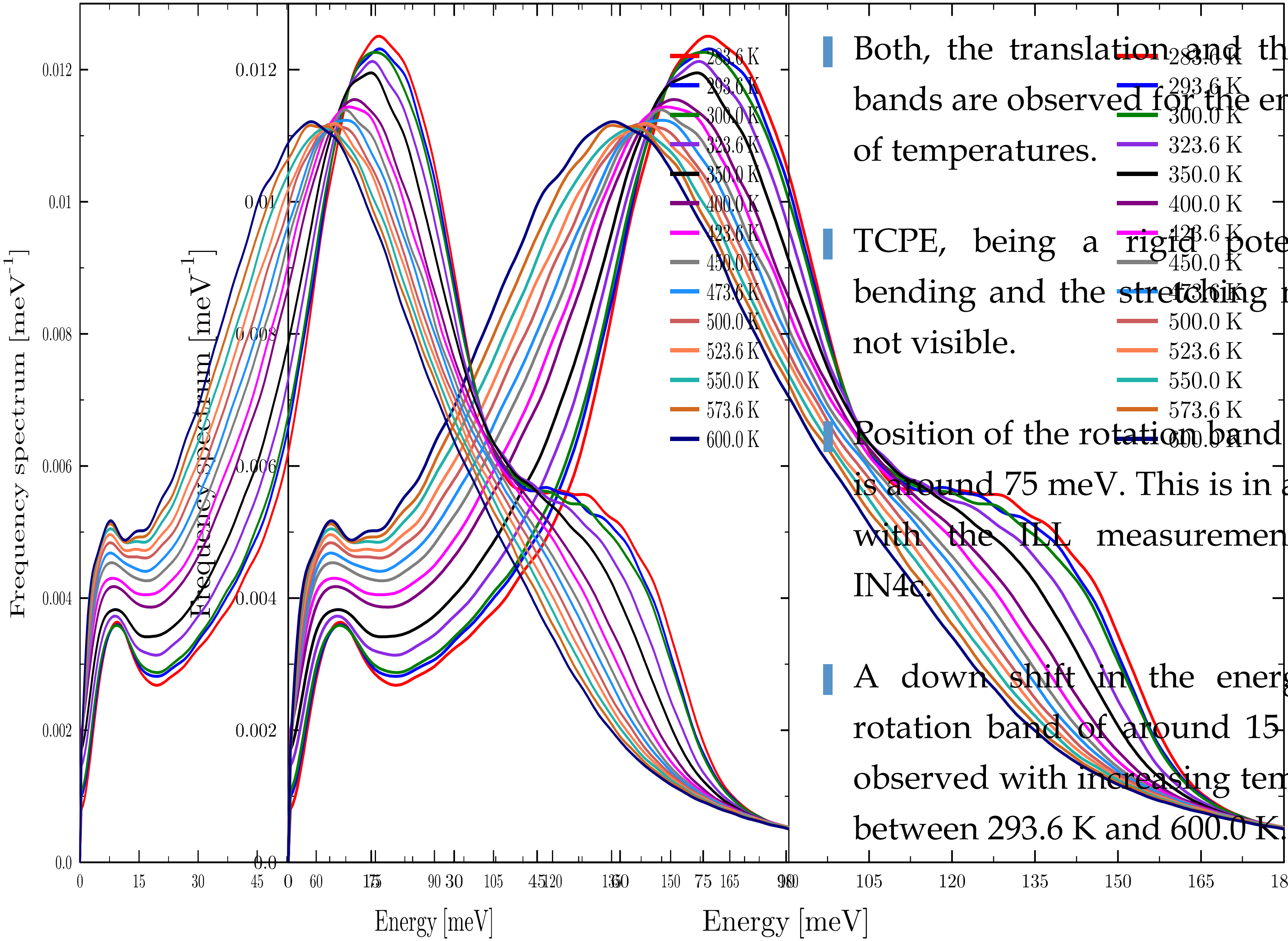
- Frequency spectrum is limited up to the rotation band.
- The energy of the rotation band is peaked around 75 meV.
- Frequency spectrum is weakly dependent on the pressure of light water.
- Implies that total cross section is independent of the pressure !!

Temperature dependence on the frequency spectrum



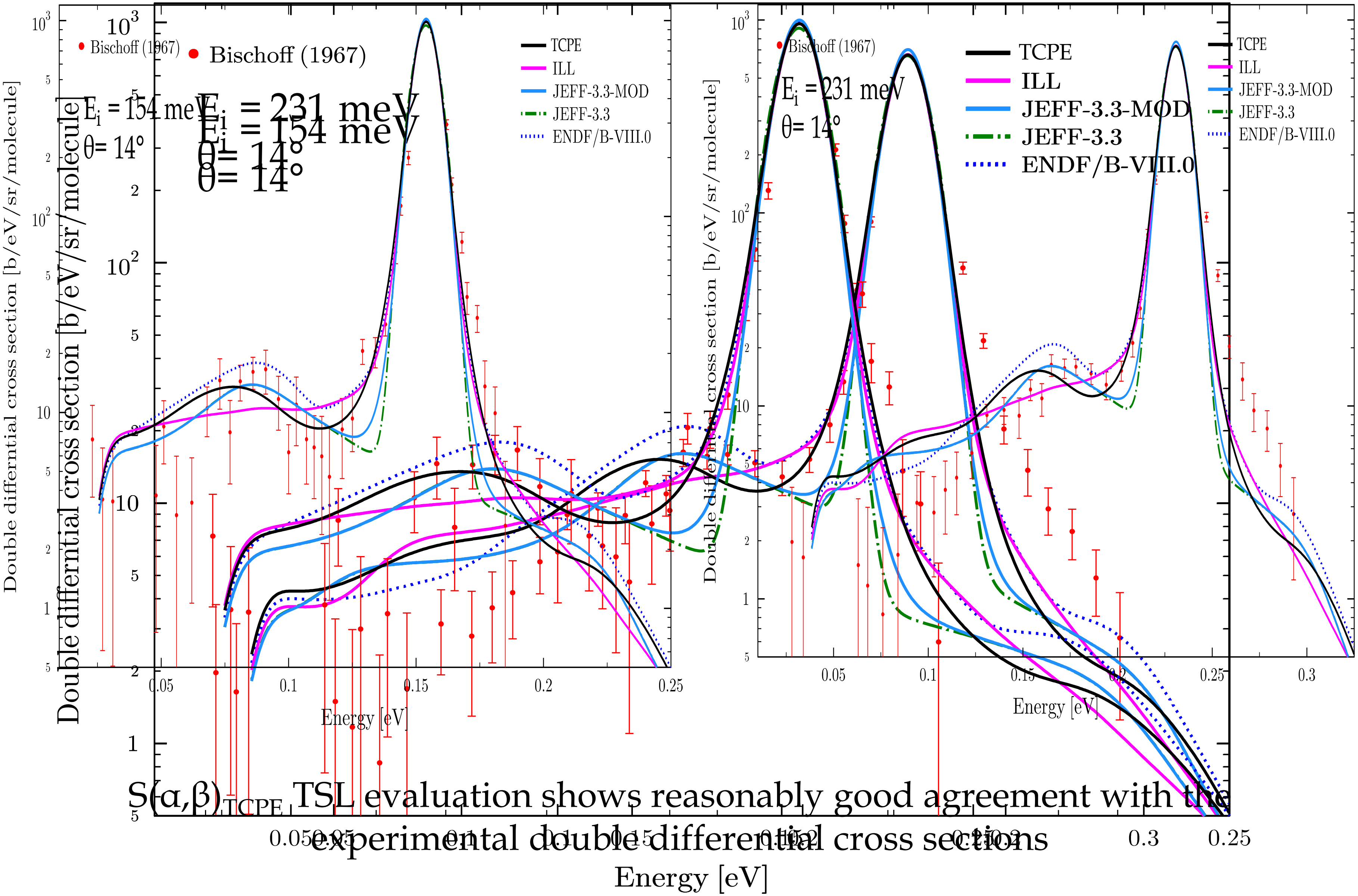
- Frequency spectrum is limited up to the rotation band.
- The energy of the rotation band is peaked around 75 meV.
- Frequency spectrum is dependent on the temperature of light water.
- The energy of the rotation band does not significantly shifts with increasing temperature.

Frequency spectrum of light water based on TCPE potential



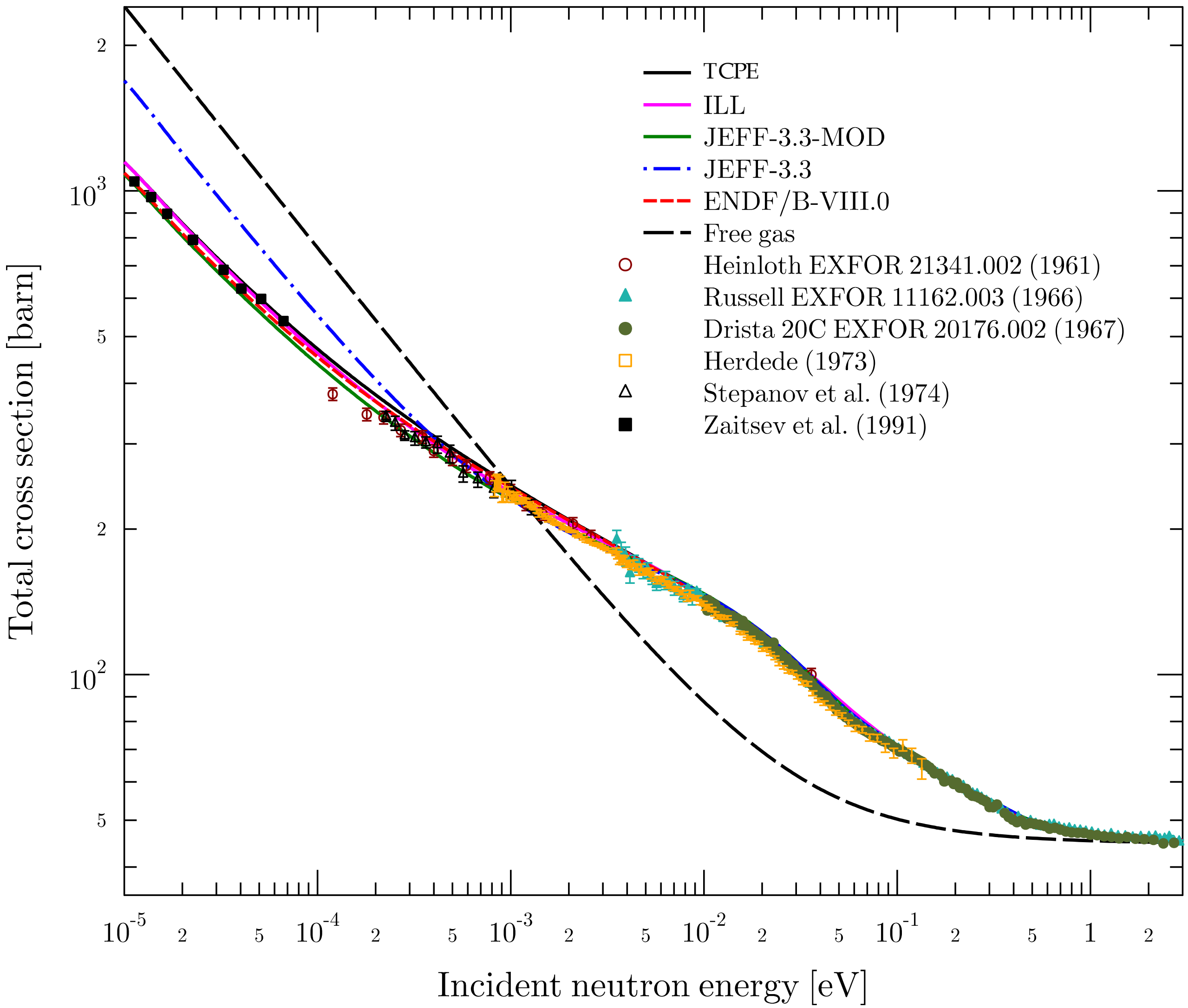
- Both, the translation and the rotation bands are observed for the entire range of temperatures.
- TCPE, being a rigid potential, the bending and the stretching modes are not visible.
- Position of the rotation band at 293.6 K is around 75 meV. This is in agreement with the ILL measurements at the IN4c.
- A down shift in the energy of the rotation band of around 15 meV was observed with increasing temperatures between 293.6 K and 600.0 K.

Double differential cross section based on new TSL evaluations



$S(\alpha,\beta)_{TCPE}$ TSL evaluation shows reasonably good agreement with the experimental double differential cross sections

Total cross section based on new TSL evaluations



$S(\alpha,\beta)_{TCPE}$ TSL evaluation (POLARIS) shows reasonably good agreement with the experimental total cross sections