The CAB Models for Water: Thermal scattering libraries for H_2O and D_2O in the 283-600 K range

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WPEC SG-42 kick off meeting
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NEA Headquarters, Issy-les-Moulineaux, France

CONICET

Instituto Balseiro

CNEA

Neutron Physics Department at Centro Atomico Bariloche

- Centro Atomico Bariloche is one of the research centers of the Atomic Energy Commission of Argentina, and it is located in the south of the country.
- The neutron physics department was founded in 1969 by Hector Antunez, one of the alumni of the legendary neutron physics group at General Atomics in San Diego.
- The group was formed around a small pulsed neutron source, a 25 MeV electron Linac, similar to the accelerator at RPI.
- Now we are 23 people (counting researchers, students and technical staff) working on neutron physics and applications to condensed matter research, materials science and nuclear engineering.
- Our main current activity is the development of neutron scattering instruments for the forthcoming RA-10 reactor, which will be similar to the OPAL reactor that the Argentine company INVAP built in Australia.

Who we are: Thermal scattering nuclear data group at Centro Atomico Bariloche

Rolando Granada

Scattering theory and advanced neutron sources

Florencia Cantargi

Cold moderator materials and neutron filters

Ignacio Marquez

Nuclear reactor applications and benchmarking

Past members: Monica Sbaffoni (currently at IAEA), Victor Gillette (currently at University of Sharjah, U.A.E).

What we do: Scattering kernels for thermal moderators

Material	SSF	LEAPR	Users and collaborators
H_2O	•	•	Slovak University of Technology CEA Cadarache
D_2O	•	•	Canadian Nuclear Laboratories
CH_2	•		Chalmers University of Technology
C_6H_6	•	•	
Plexiglass	•		Institute of Nuclear Physics, Poland
Ethanol	•		
Dowtherm	•		
Metal Hydrides	•		
Dodecane	•		Century Research Center Corp., Japan
Tributylphosphate	•		Century Research Center Corp., Japan
Mesitylene		•	Joint Institute of Nuclear Physics, Russia
Graphite	•	•	

What we do: Scattering kernels for cold moderators

Material	SSF	LEAPR	Users and collaborators
Liquid H_2		•	INVAP S.E., Argentina
Liquid D_2		•	INVAP S.E., Argentina
Solid D_2		•	Paul Scherrer Institut
Liquid and Solid CH_4		•	Indiana University, U.S.A.
Solid H_2O		•	Bhabha Research Center, India
			Hokkaido University, Japan
Clathrate		•	JESSICA collaboration
Solid Mesitylene		•	JESSICA collaboration
			Joint Institute of Nuclear Physics, Russia
			Hokkaido University, Japan
			TRIUMF, Canada
			LANL, USA
Mesitylene/Toluene		•	TRIUMF, Canada
Liquid Ethane		•	
Triphenylmethane		•	

(See talk SG42-10 by Florencia Cantargi, tomorrow afternoon at 16:00)

What we do: Scattering kernels for neutron filters

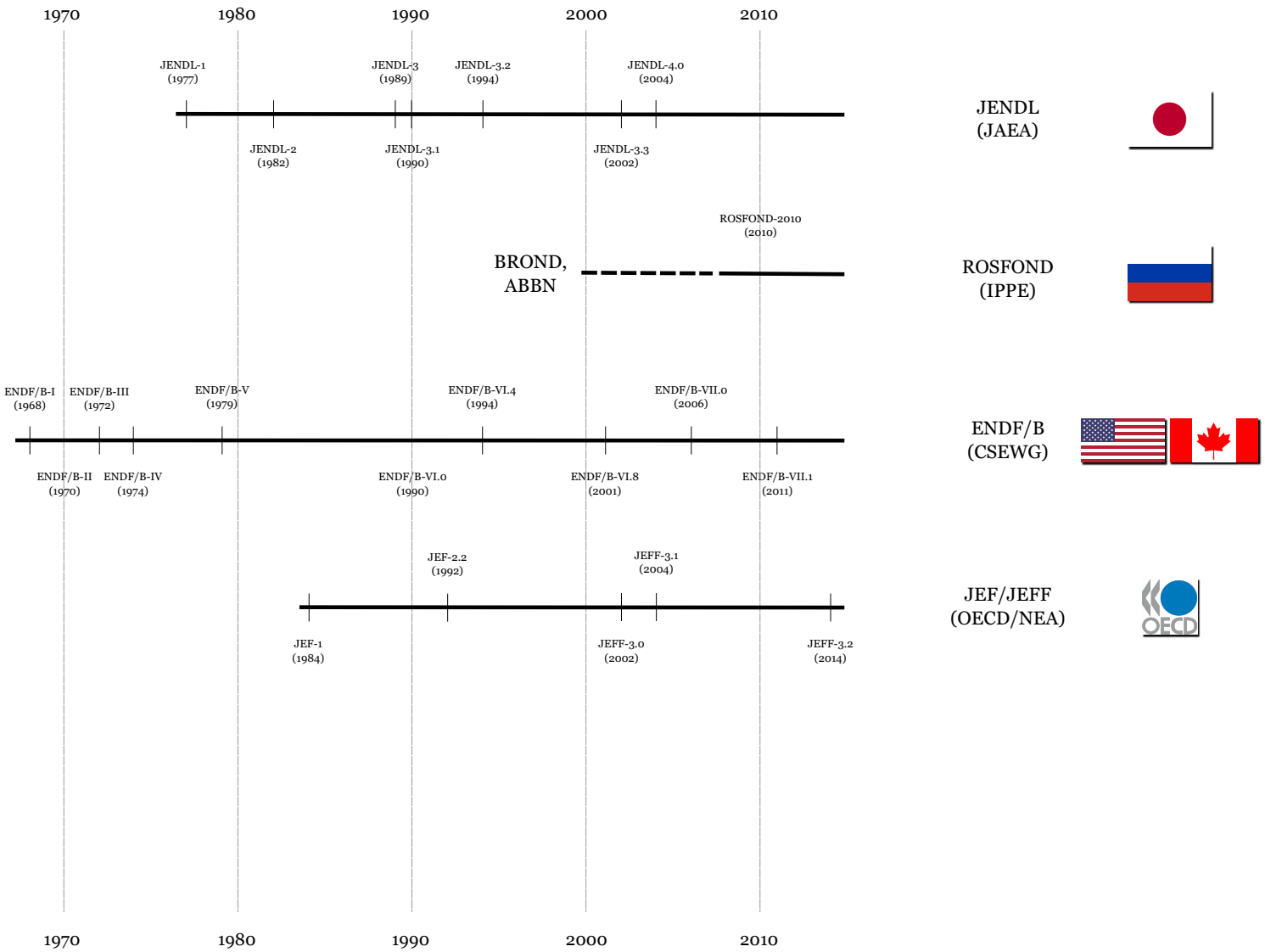
Material	SSF	LEAPR	Users and collaborators
Silicon		•	RA-10 reactor project, Argentina
			Paul Scherrer Institut, Switzerland
Sapphire		•	RA-6 reactor, Argentina
		•	RA-3 reactor, Argentina
			Paul Scherrer Institut, Switzerland
Bismuth		•	RA-6 reactor, Argentina
Silica		•	ORNL, U.S.A

(See talk SG42-10 by Florencia Cantargi, tomorrow afternoon at 16:00)

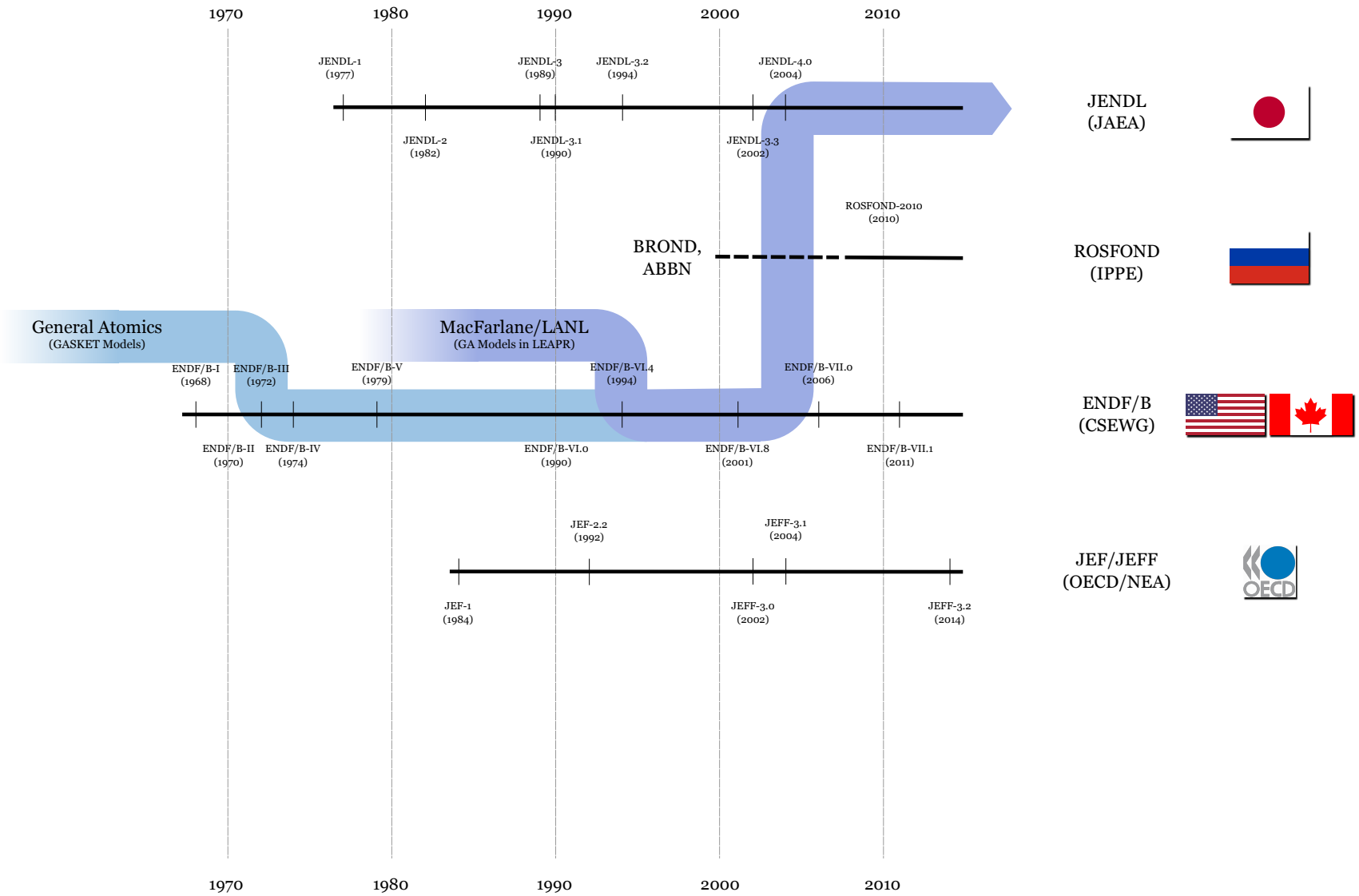
The CAB Evaluation for water: some context

- In 2008 Rolando proposed to revisit the scattering of neutrons in water, applying the recent experience of the group in advanced moderators.
- Red Cullen had recently published the reports *How Accurately Can We Calculate Neutrons Slowing Down In Water?* and *How Accurately can we Calculate Thermal Systems?*, which caught our attention.
- We decided it would be a good PhD topic for me, who at that time was participating in the ICSBEP project.

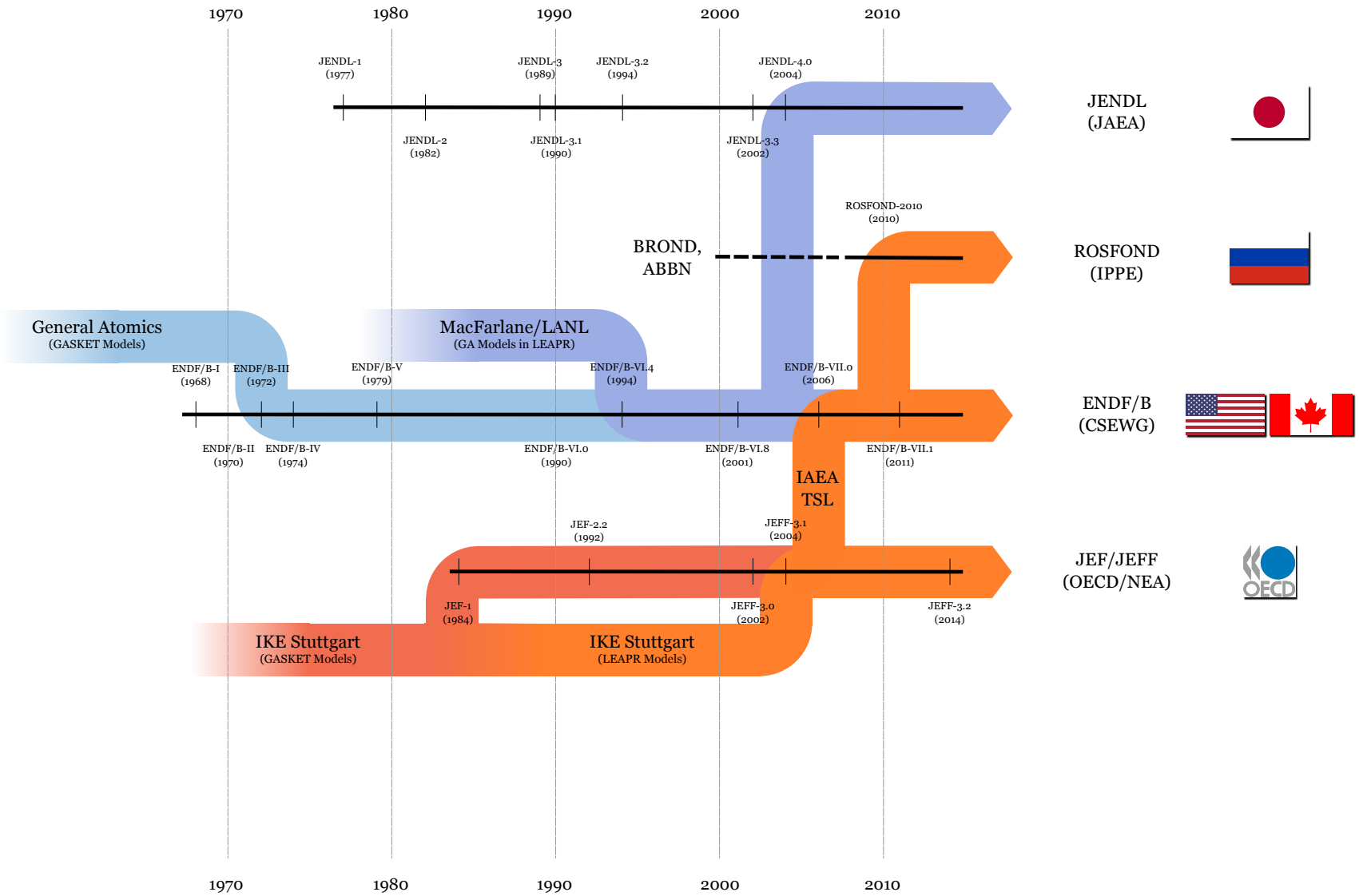
THERMAL SCATTERING MODELS IN THE EVALUATED NUCLEAR LIBRARIES



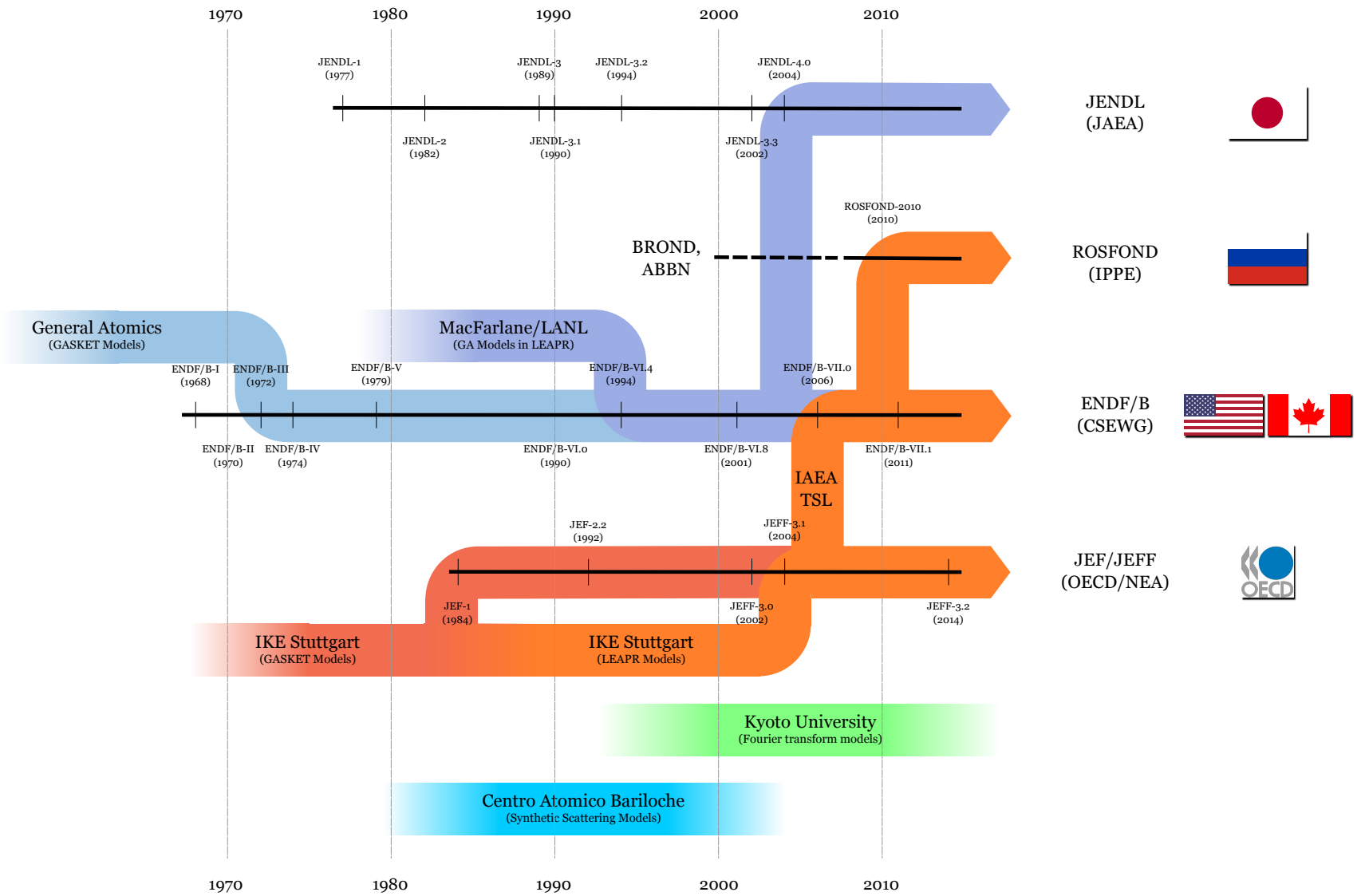
Thermal scattering models in the evaluated nuclear libraries: General Atomics Model



Thermal scattering models in the evaluated nuclear libraries: IKE Stuttgart Model



Other water models (not included in the evaluated nuclear libraries)



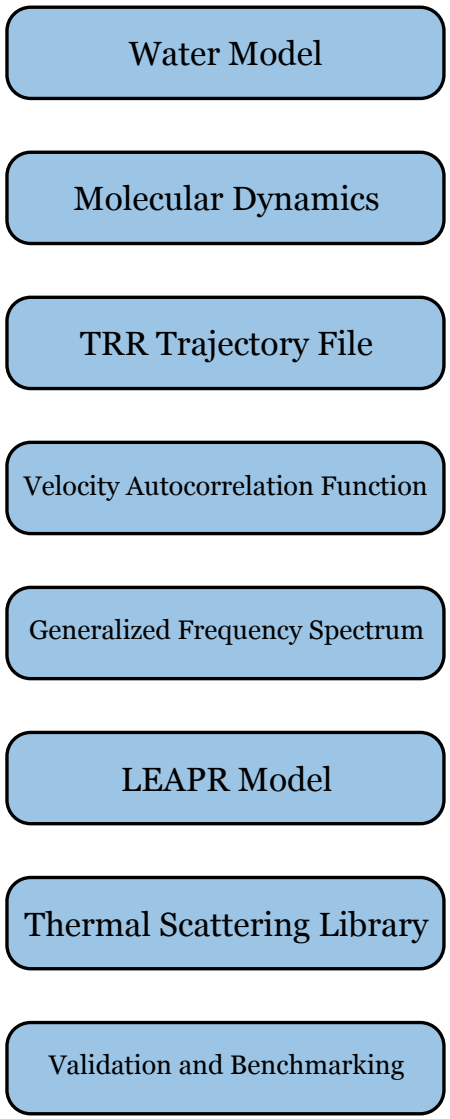
Evaluation methodology: principles

1. Use **standard** tools to ensure the scattering kernel could be processed in NJOY ⇒ LEAPR.
2. Use the best available experimental information on water, and complement it with molecular dynamics. In particular we knew from the work by Marti, Padro and Guardia at UPC that it was possible to compute the frequency spectrum of water correctly. As newcomers to molecular dynamics, we started with help from David Malaspina from Northwestern University.
3. Include molecular diffusion to improve the quasielastic region. It is not perfect, but it is better than the current models that use free gas, and works out of the box in LEAPR (see 1).
4. Add structural corrections when necessary (deuterium and oxygen) using the Sköld approximation and structure data from Alan Soper and molecular dynamics.

Evaluation methodology: why LEAPR?

- Aside of being the standard, there is a big problem when looking at alternatives to LEAPR: the wide dynamic range involved:
  - An ENDF-6 standard thermal scattering evaluation should work in a wide incident energy range: from $E_{in} = 10^{-5}$ eV to 5-10 eV.
  - Representing the angular distribution from $E_{in} = 10^{-5}$ eV requires a scattering vectors from ~ 0.150 Å^{-1} to ~ 150 Å.
  - The quasielastic peak width at $Q = 0.150$ Å^{-1} is $\hbar D Q^2 \simeq 30 \times 10^{-9}$ eV. If we discretize that peak using ten points, the required $\epsilon = \hbar\omega$ range goes from 3×10^{-9} eV to 10 eV.
- As an alternative, we did compute $S(Q, \omega)$ as a Fourier transform in time and space of $G(r, t)$ for limited ranges of Q and $\hbar\omega$. But a full (ENDF-6 compatible) evaluation would require to compute $G(r, t)$ in a timescale from 0.1 fs to 10 μs (10^{10} timesteps!).
- We decided to stick with LEAPR until we find (or develop) something better.

Evaluation workflow

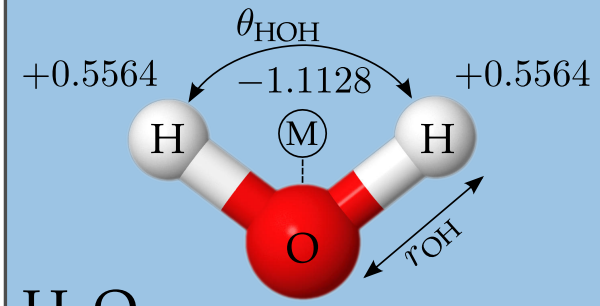


Evaluation workflow

- Water Model
- Molecular Dynamics
- TRR Trajectory File
- Velocity Autocorrelation Function
- Generalized Frequency Spectrum
- LEAPR Model
- Thermal Scattering Library
- Validation and Benchmarking

$$V = \sum_{i \neq j} 4\epsilon_O \left[\left(\frac{\sigma_O}{|r_{O_i} - r_{O_j}|} \right)^{12} - \left(\frac{\sigma_O}{|r_{O_i} - r_{O_j}|} \right)^{6} \right] + \frac{q_i q_j}{r_{ij}}$$

$$+ D \left[1 - e^{\beta (r - r_{\mathrm{OH}})^2} \right] + \frac{1}{2} k^{\theta}_{HOH} \left[\theta_{HOH} - \theta^{0}_{HOH} \right]^2$$



H_2O

$\sigma = 3.1644 \times 10^{-1}$ nm
$\epsilon = 7.7490 \times 10^{-1}$ kJ mol^{-1}
$k^{\theta}_{HOH} = 367$ kJ mol^{-1} rad^{-2}
$\theta_{\mathrm{HOH}} = 104.79$ °
$D = 432.581 \times 10^{5}$ kJ mol^{-1}
$\beta = 22.87$ nm^{-1}

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A flexible model for water based on TIP4P/2005

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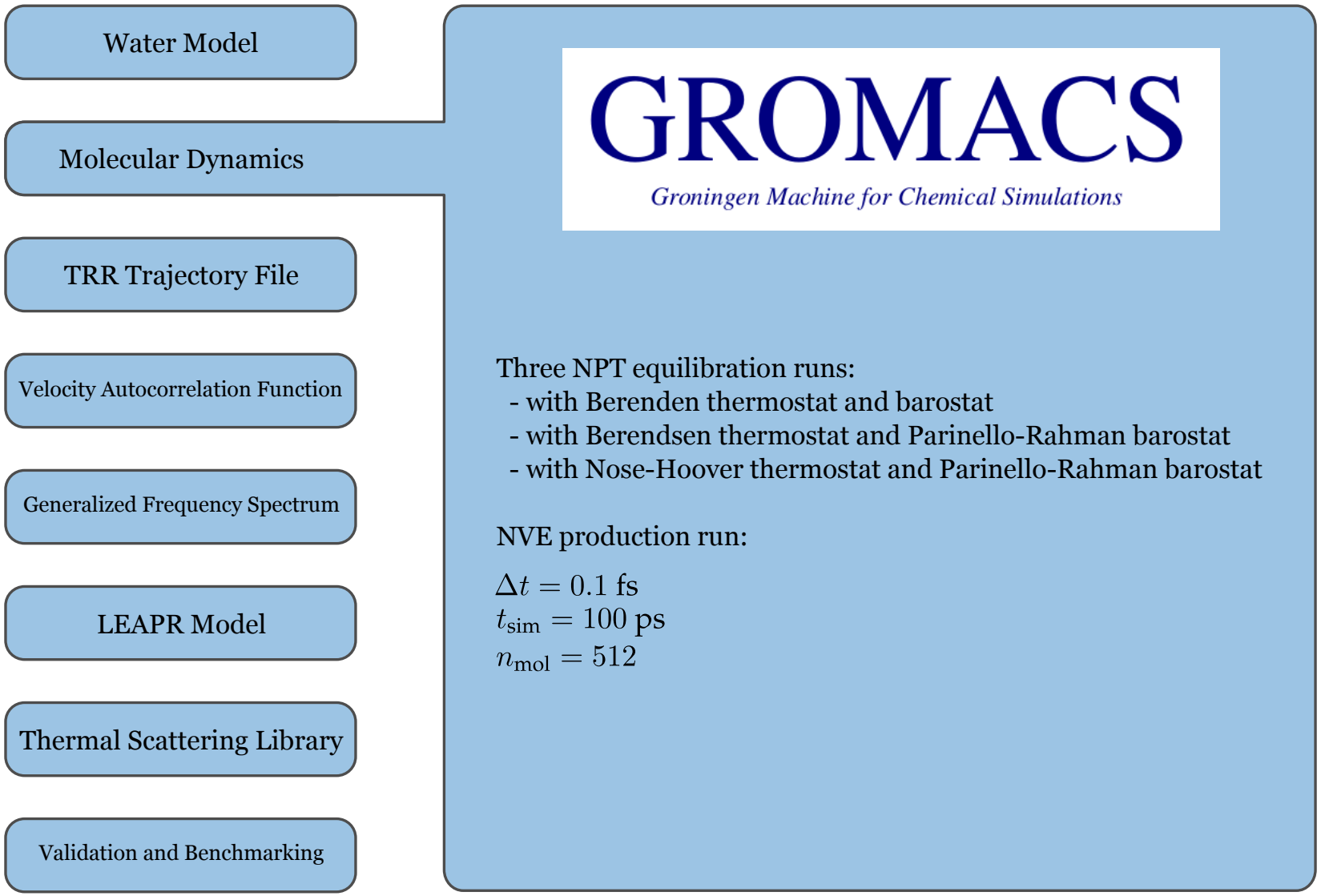
(Received 29 July 2011; accepted 2 November 2011; published online 14 December 2011)

Evaluation workflow



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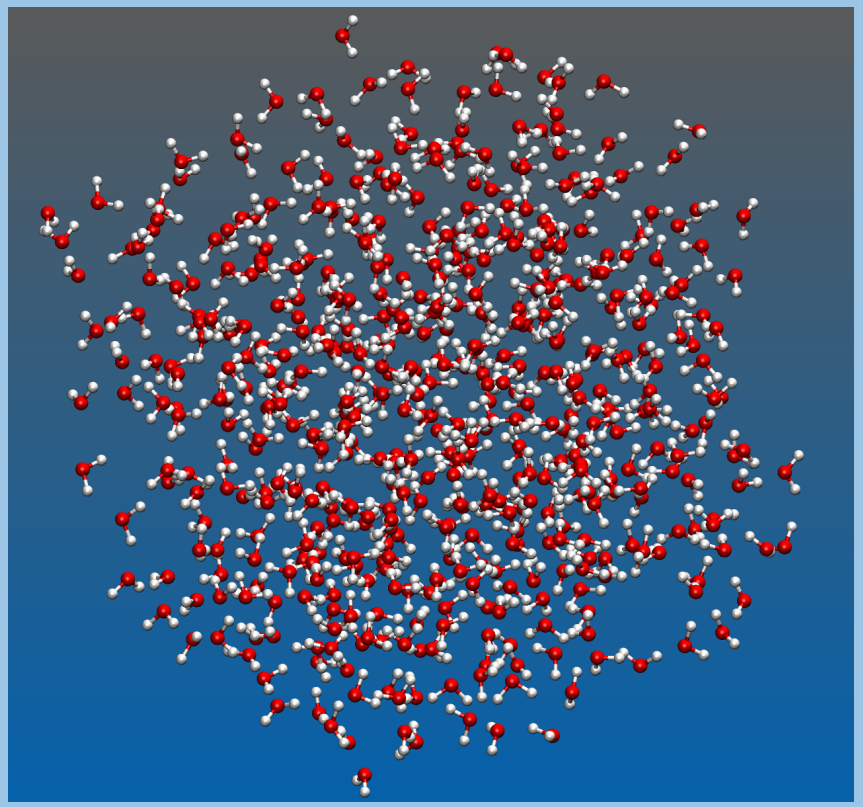

100 ps with velocity vector saved every 6 frames: ~ 8 Gb

Evaluation workflow

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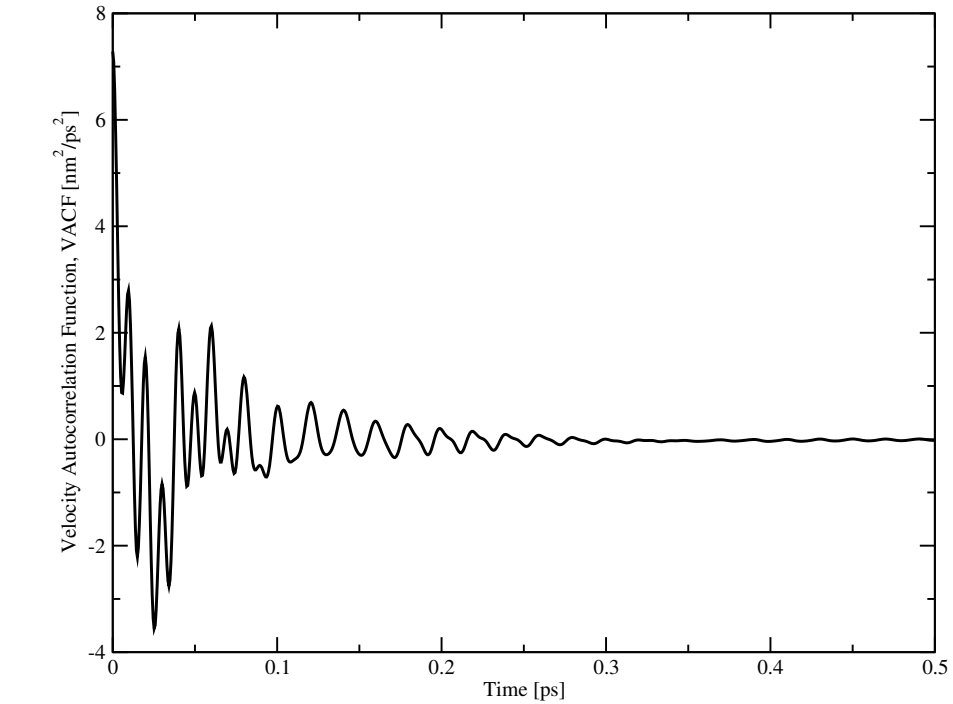

Velocity autocorrelation function computed with g_velacc_d

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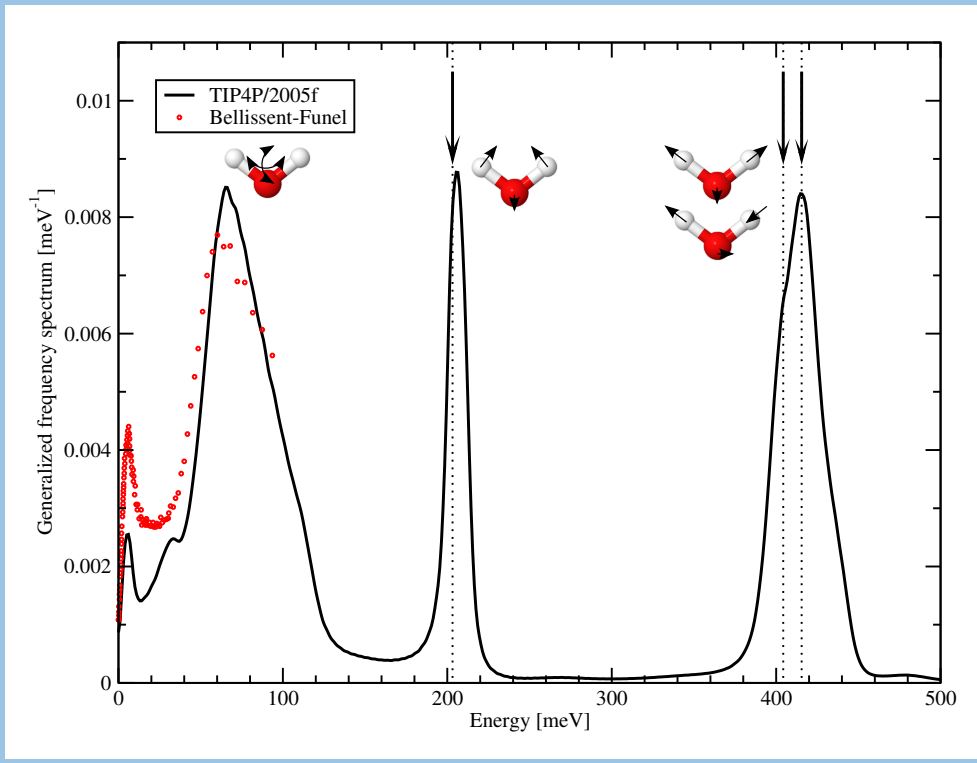

Generalized frequency spectrum for H bound in H2O at room temperature, computed from molecular dynamics. Arrows mark the internal vibrational modes as measured by Lappi.

Evaluation workflow

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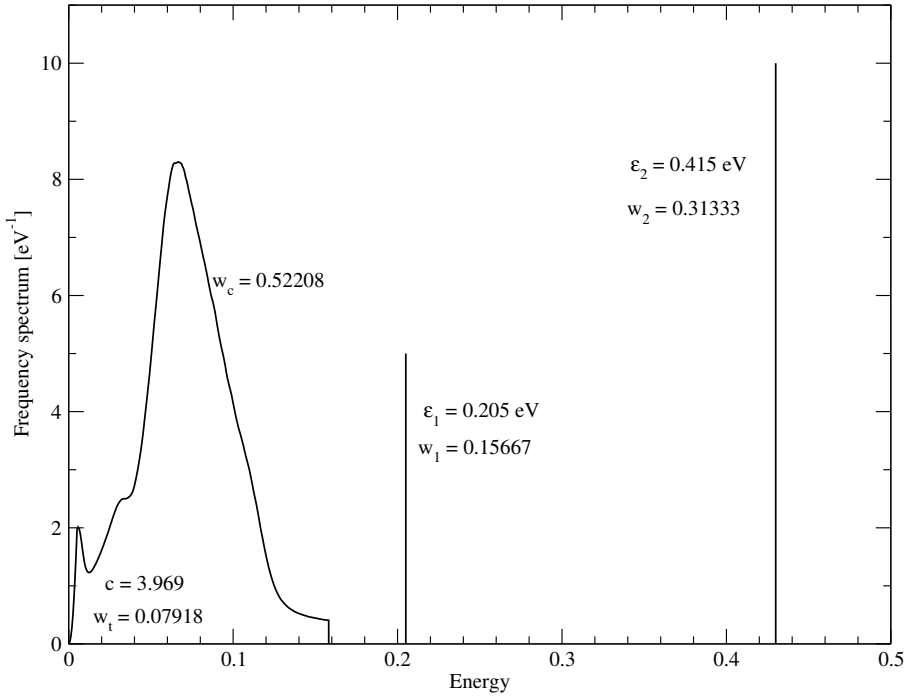

Parameters for the LEAPR model for H bound in H2O at 293 K

Evaluation workflow

- Water Model
- Molecular Dynamics
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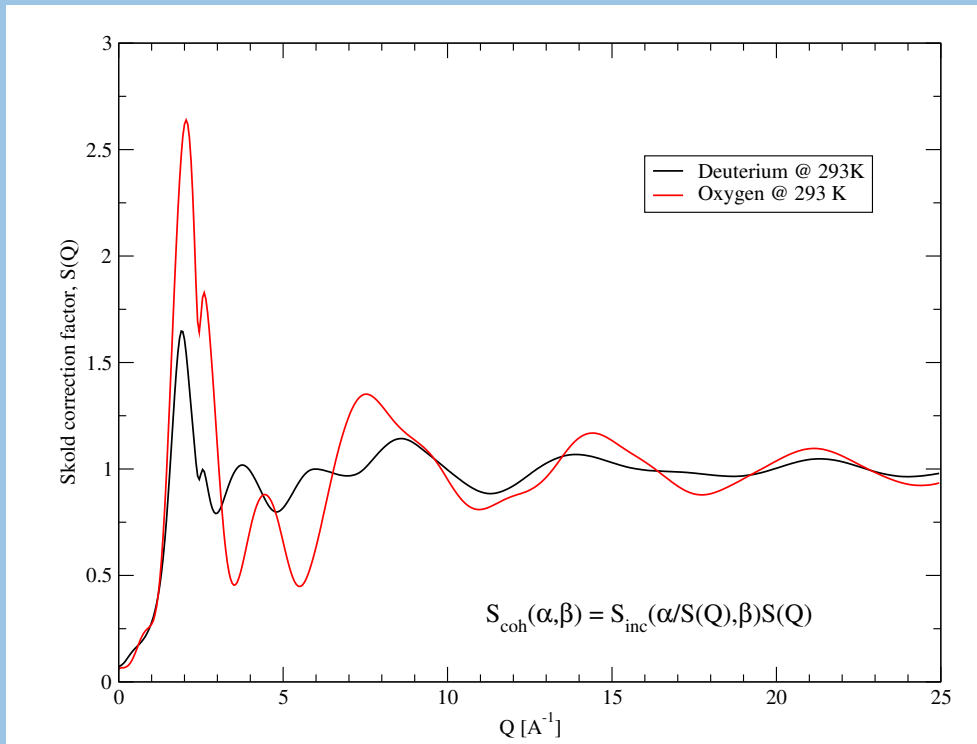

Sköld correction factors for deuterium and oxygen bound in D2O

Evaluation workflow

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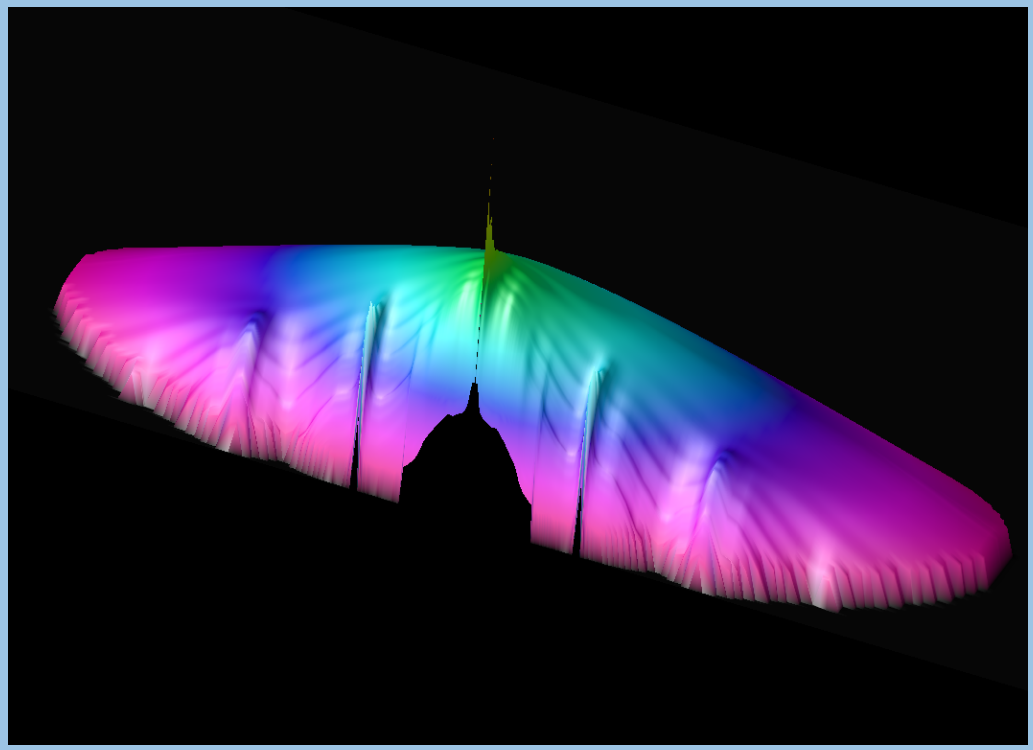

Log plot of the symmetric $S(\alpha, \beta)$ for deuterium bound in heavy water

Evaluation workflow

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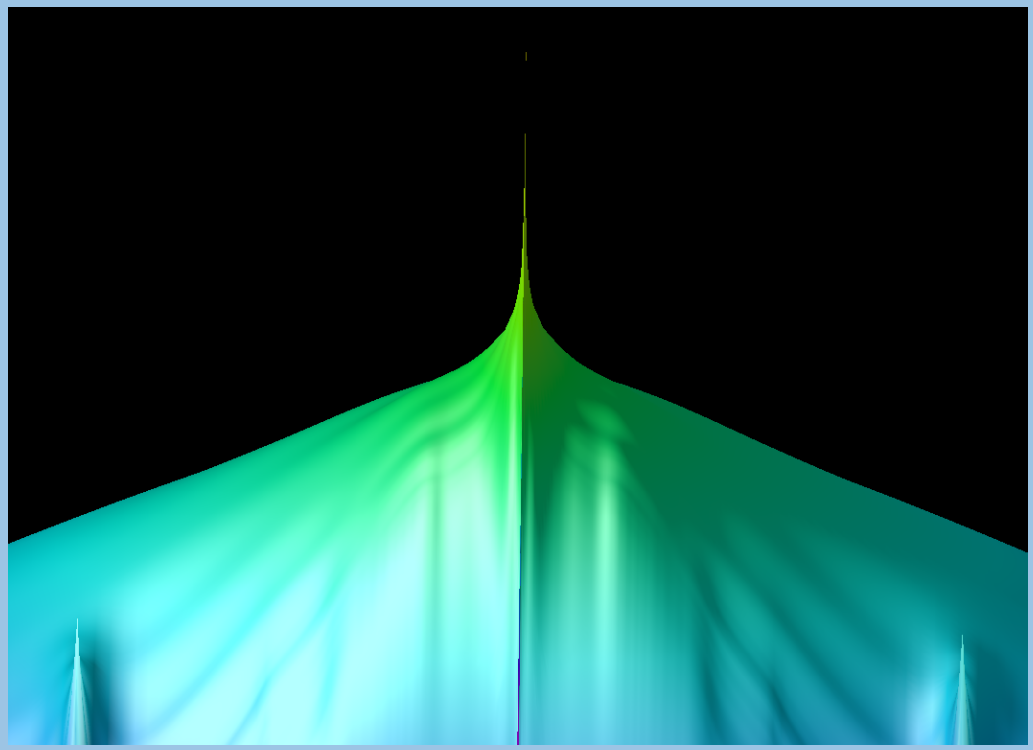
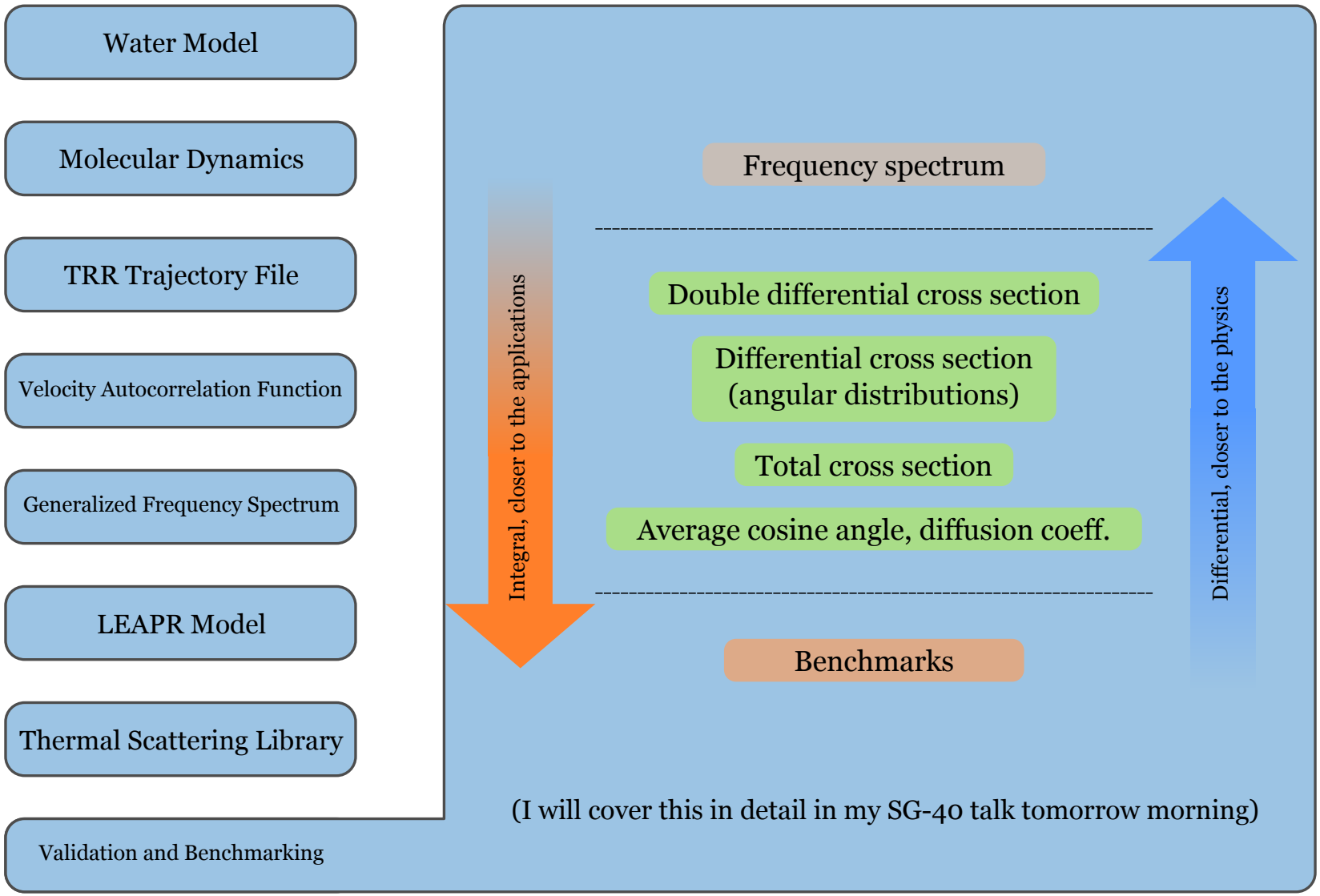


Detail of the quasielastic peak.

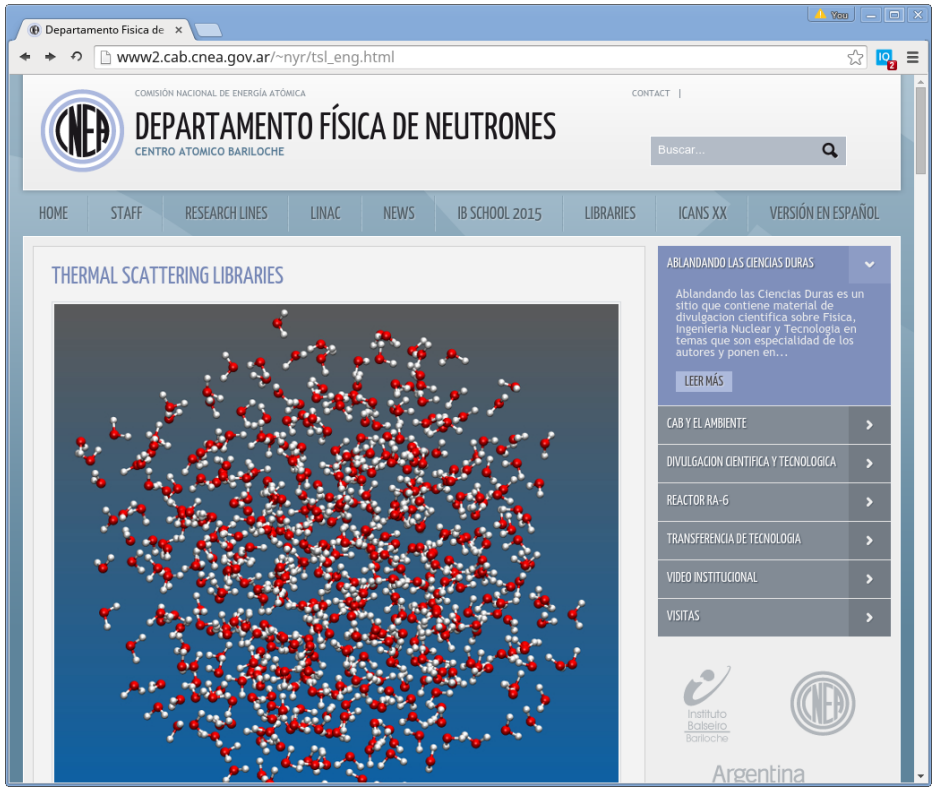
Evaluation workflow



EVALUATION: LIBRARY FILES

Using this methodology we evaluated the thermal scattering law for hydrogen bound in light water and for deuterium and oxygen in heavy water from 283 to 600 K.
The libraries are available in ENDF-6 and ACE format in the webpage of the Neutron Physics Department:

`http://www2.cab.cnea.gov.ar/~nyr/tsl_eng.html`



Active collaborations related to this project

Around this project we developed an collaboration network with international partners:

- Danila Roubtsov, from Canadian Nuclear Laboratories: library processing and refinement; validation with criticality safety and reactor physics benchmarks (see talk SG42-11 tomorrow morning at 9:30)
- Gilles Noguere and Juan Pablo Scotta from CEA/Cadarache: application to light water reactors and covariance matrix calculations (see talk SG40-12 tomorrow morning at 11:00).
- David Baxter from Indiana University: total cross section measurements for light and heavy water at LENS (see talk SG42-6 tomorrow afternoon at 15:30).

Summary and conclusions

- The Neutron Physics Department at Centro Atomico Bariloche has more than 40 years of continuous experience working in neutron scattering applied to nuclear data and nuclear engineering.
- During the past 6 years our group has been working on a new evaluation of the thermal scattering law for light and heavy water. As I will show tomorrow morning, these evaluations are an improvement over existing evaluations.
- The libraries are available for downloading at our website, and we encourage you to use them and give us feedback. We look forward to do the required work to make these evaluations available in the evaluated nuclear data libraries.
- Surrounding this work we developed a collaboration network with diverse expertise and common interest in neutron scattering libraries.


Thanks for your time.
Bariloche, Argentina