ON IMPROVEMENT OF THERMAL NEUTRON SCATTERING LIBRARIES ($S(\alpha, \beta)$ data) FOR H_2O and D_2O

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This work is International Collaboration project

in the area of *Nuclear Data* R&D

for Nuclear Science and Technology applications

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Motivation

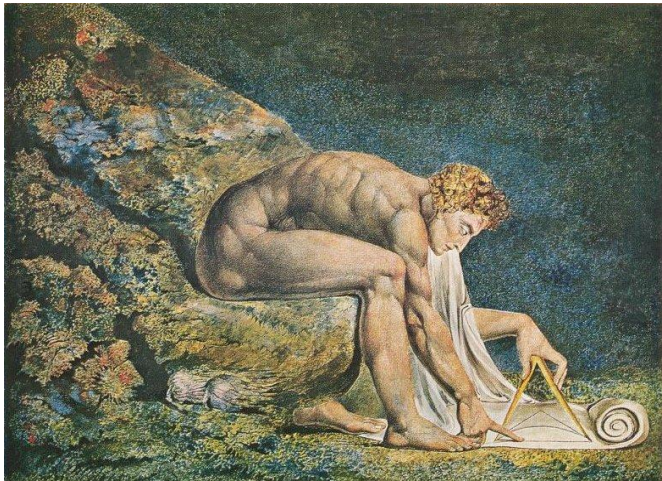
Thermal Scattering Laws (**TSL** or S(α,β) data) for liquids, **H_2O** and **D_2O**, are available in the **evaluated nuclear data libraries**, such as,

ENDF/B-VII.0 (2006) $\rightarrow$ **ENDF/B-VII.1** (2011) / USA /

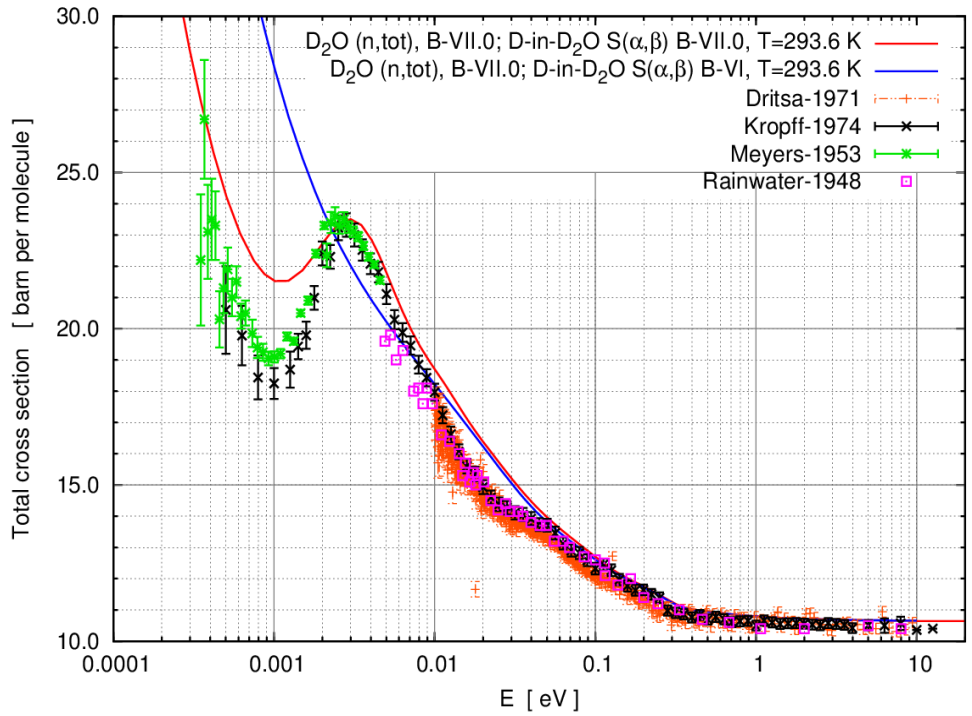
JEFF 3.1 (2005), **JEFF 3.2** (2014) / EU /
also in JENDL /Japan/, BROND (ROSFOND).

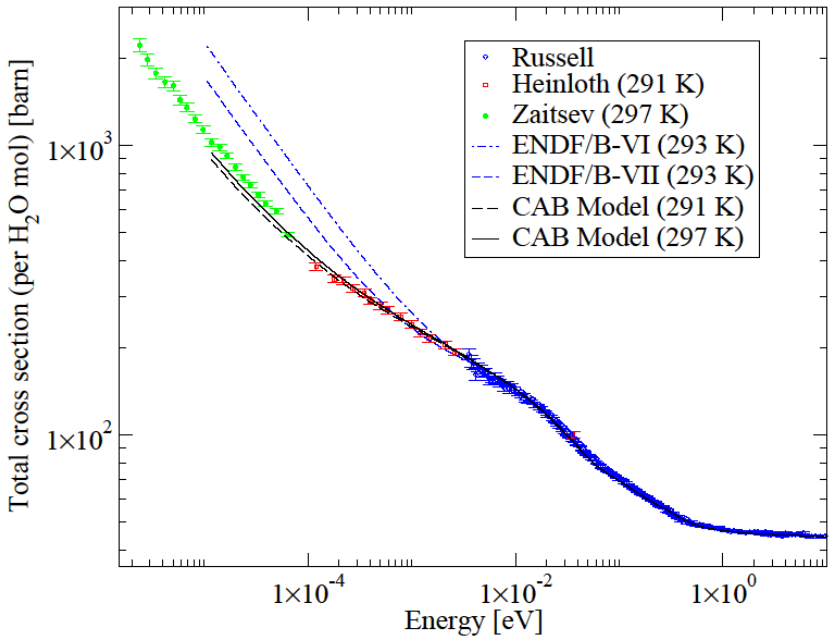
There are also **multi-group** TSL libraries (*CLES*) developed by Kyoto University group (Morishima, Edura *et al*.), for cold moderators.

What can be improved in modern evaluated S(α,β) data from the standpoint of (**Reactor Physics**) **Applications** (?) from the standpoint of advances in computational **CMP** ?



TSL: liquid D_2O, and D_2O *vs.* H_2O





<u>Total cross sections</u>: n + D_2O *vs.* n + H_2O at *E* < 10 eV

$\sigma_{tot}(E)$ in *barn per molecule* for D_2O ($\approx {}^2H_2{}^{16}O$) in heavy water at Room Temp (RT)

Look at *E* near E_{th} = 0.0253 eV : for D_2O at RT , we have "heavy water challenge":
(*Exp.* – *Calc.*) / *Calc.* ≈ –8.4% (using B-VII.0 **S(α,β)** for D-in-D_2O and **FG model** for ^{16}O)

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S(α,β) data for water: outline of our approach

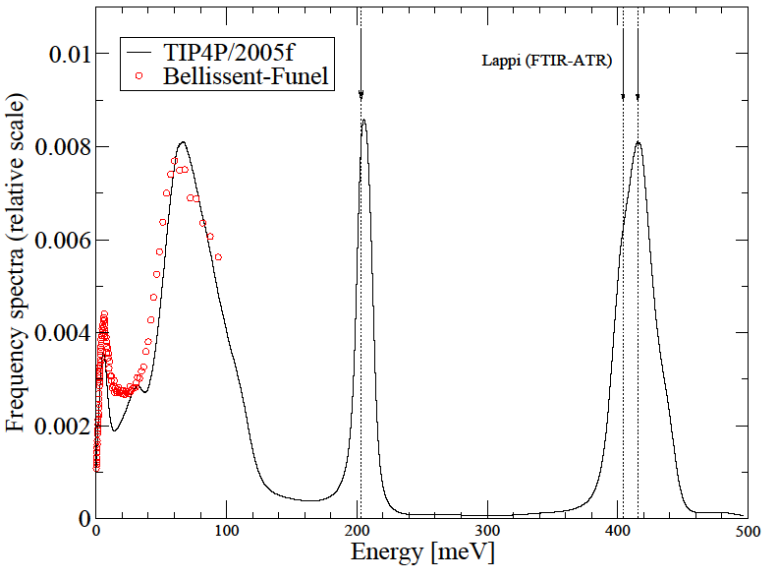
- New **evaluation** (in ENDF format) is based on combining **molecular dynamics** (MD) simulations and *experimental data*, and the resulting **models** are implemented / compatible in / with **LEAPR** module of **NJOY** (nuclear data post-processing code, LANL)

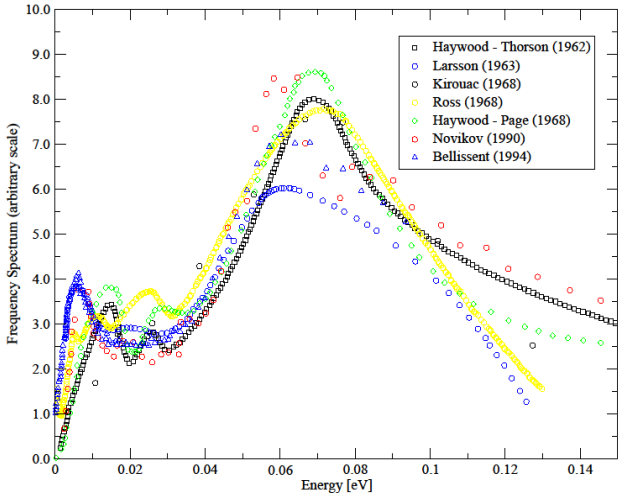
- The **key points** for building **CAB models** are:

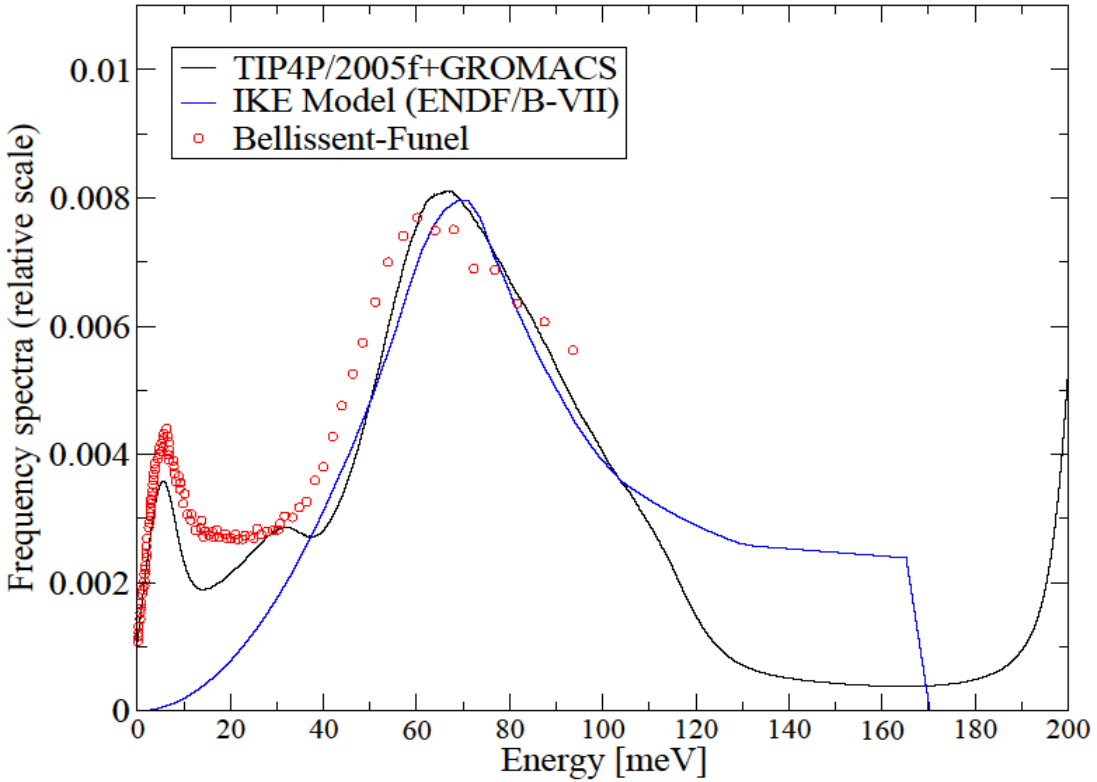
1. use of molecular **(self)diffusion** for translational motion of H_2O / D_2O (instead of free gas approximation (FG) used in **all** evaluated ND libraries);
2. continuous **vibrational spectra** computed from molecular dynamics (**MD**) simulation at a given thermodynamic state of the liquid (*p, T*) and $\rho(p, T)$ (instead of derived / adjusted from neutron scattering experiments);
3. a more precise description of **the structure of liquid:** models for D **and** O in D_2O are based on **experimental results** (instead of using the incoherent approximation in ENDF/B-VI or the Lennard-Jones **model** for D-D structure in JEFF 3.1 and ENDF/B-VII.0 $\rightarrow$ VII.1)
4. better **numerics** (*e.g.*, extended grid(s), and NJOY data processing options revisited, also with NJOY (NJOY99up396) patches in `leapr`, `thermr`)

- The resulting scattering kernels / cross sections are an improvement over existing evaluations: **they are compared with measurements** of double differential scattering cross sections, quasi-elastic neutron scattering measurements, angular distributions of out-scattered neutrons, average cosine of the scattering angle, and **total cross sections**.

H_2O: from gen. vibrational spectrum $\rho(E)$ to cross sections (for H-1)

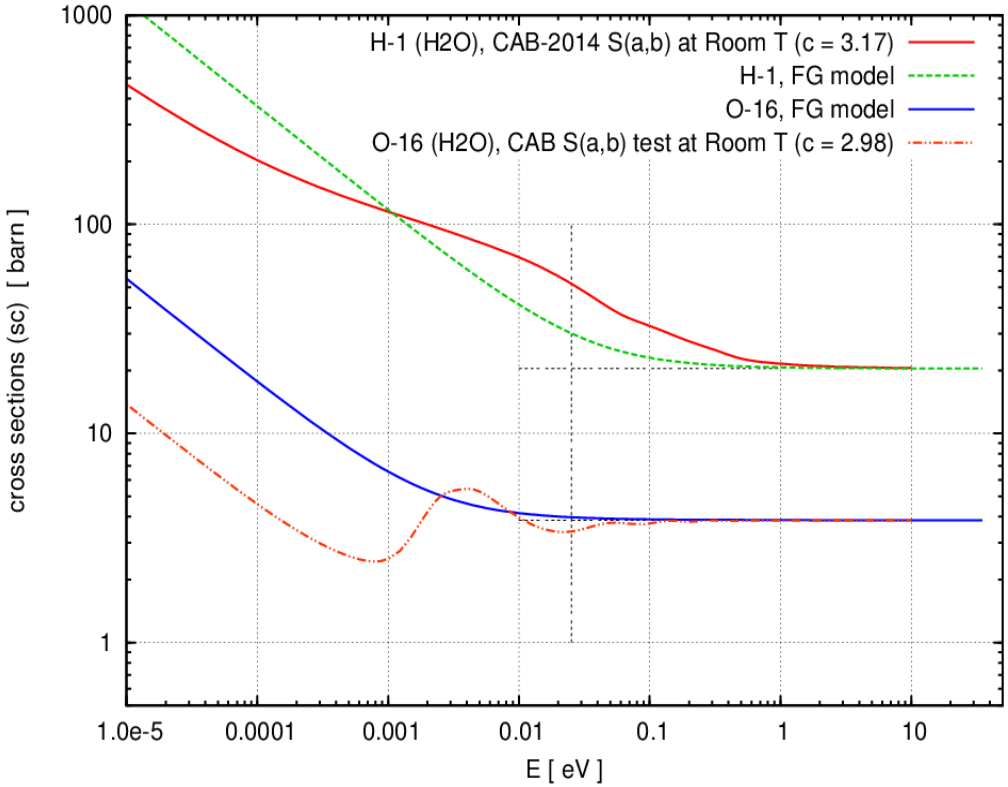


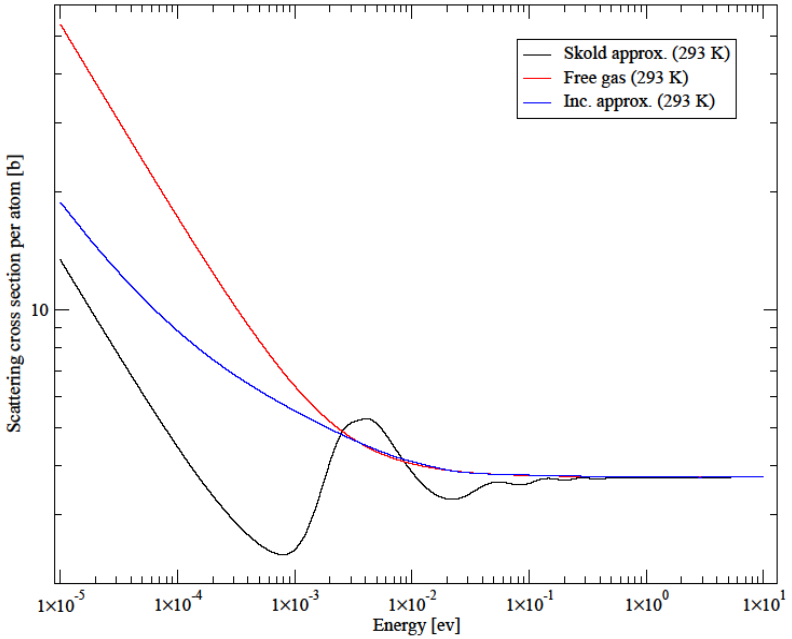




- low-energy modes (E < 40 meV): based on modeling (use MD code GROMACS)

O-16 in H_2O, Room T (1)





- At E ~ 10 eV, O-16 contributes ~ 8.5 % to the scattering x-sections of H_2O (**barn per mol.)**
- At E = 0.0253 eV, O-16 (FG) contributes ~ 3.6 % to the scattering x-sections of H_2O (RT) and 3.6% → 3.1% if we use CAB model for S(α, β) with $\rho_{O\text{-in-}H2O}(E)$ and structure factor $S_O(q)$

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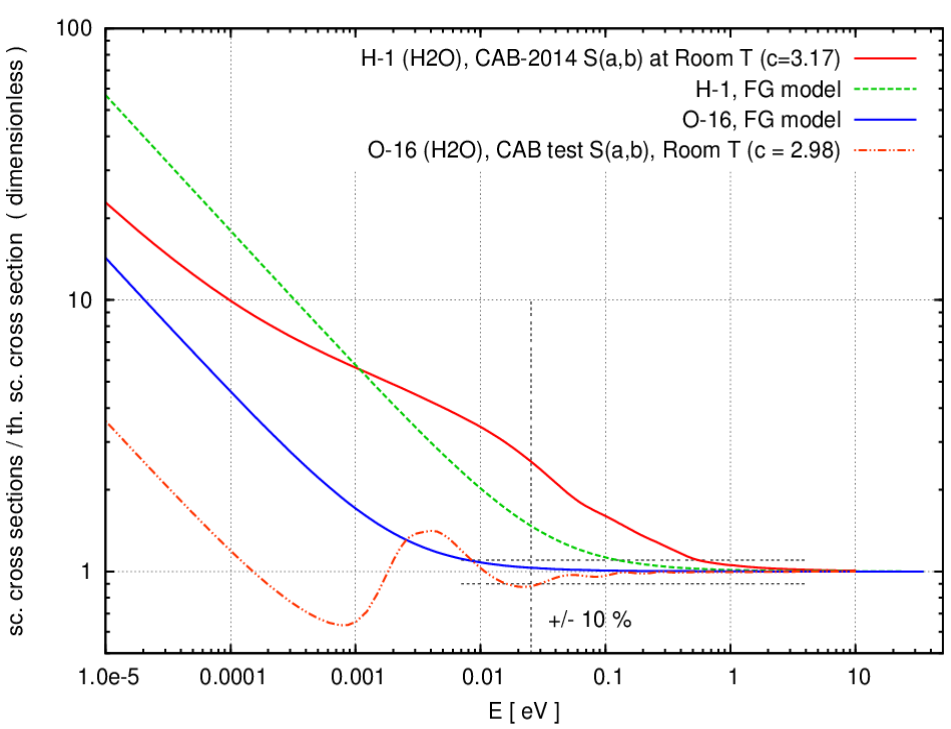
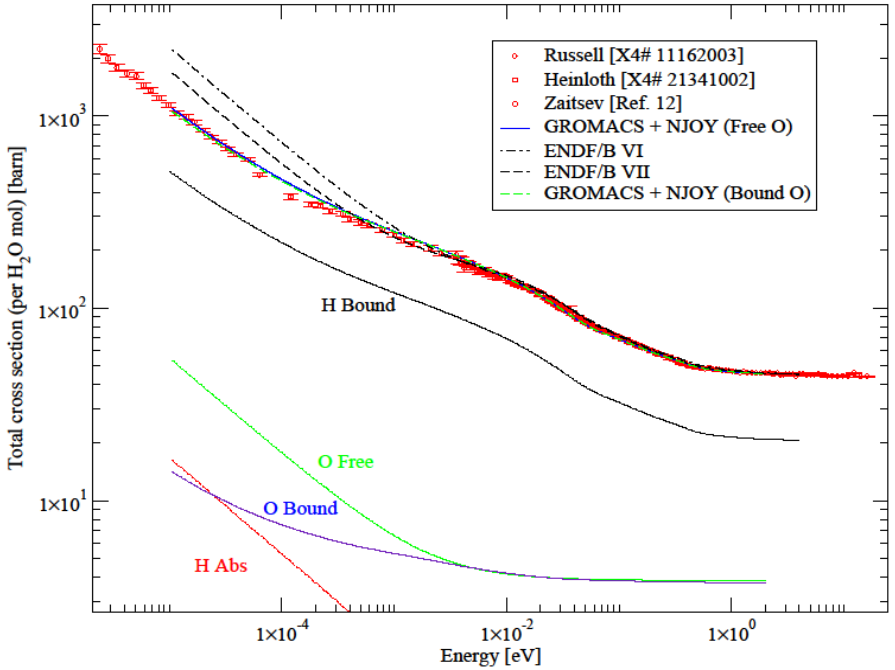
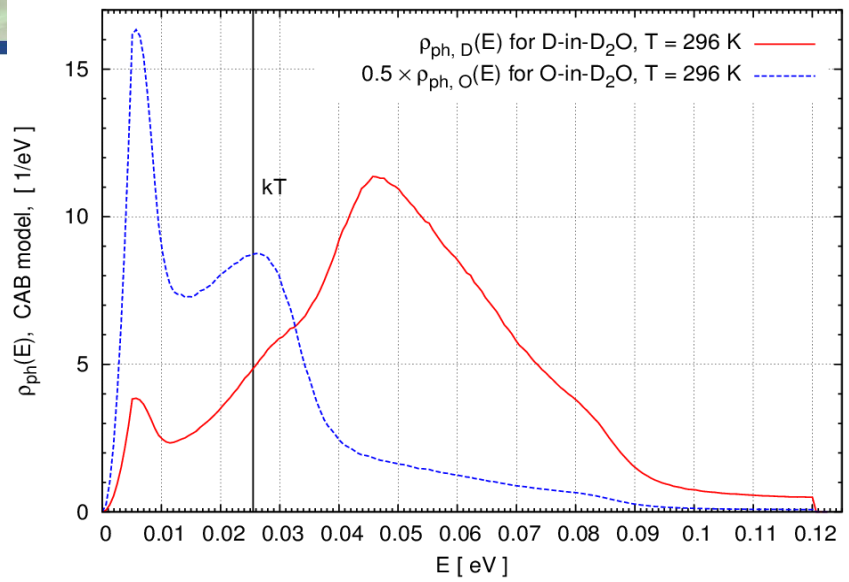
O-16 in H_2O, Room T (2)

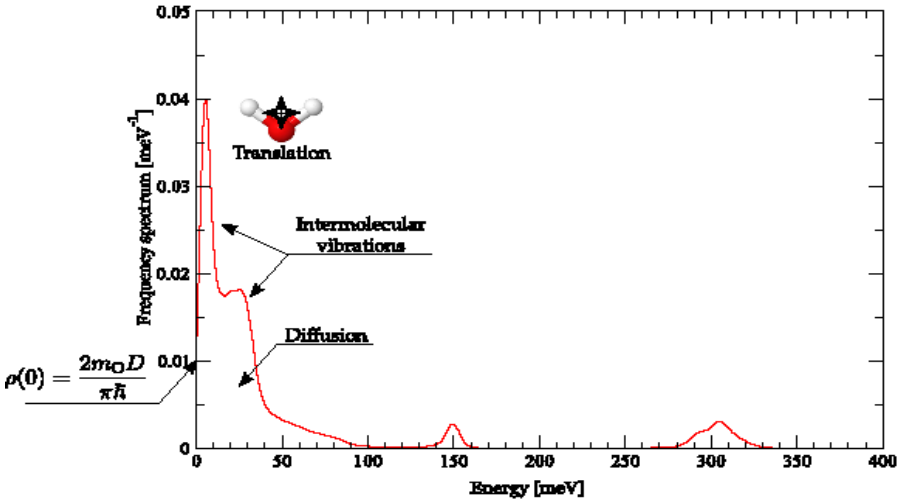


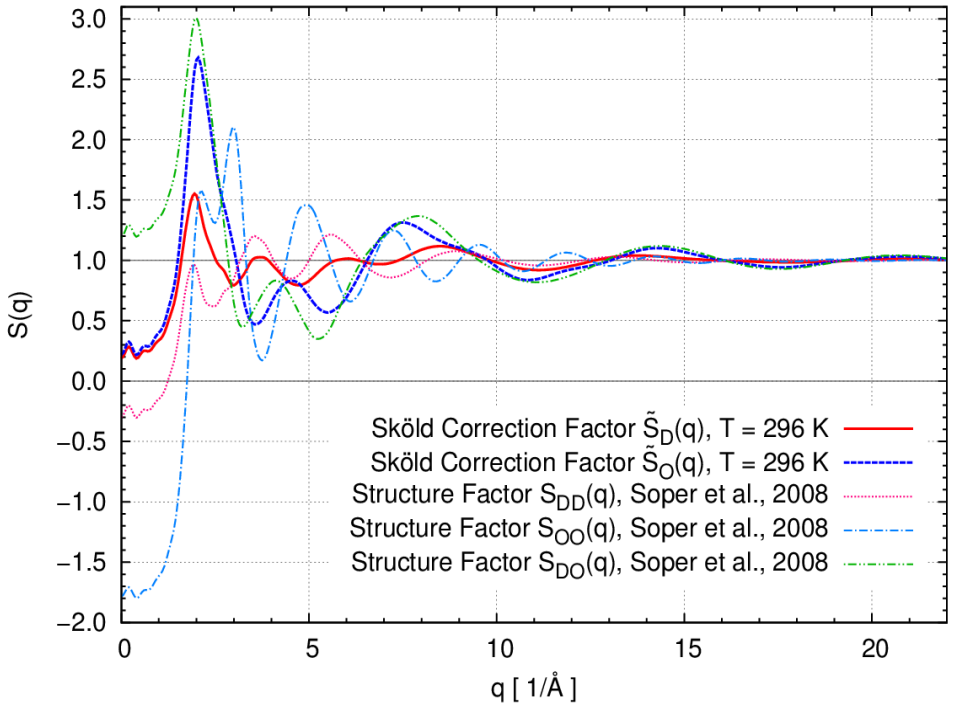


- Plot the ratio, $\sigma_s(E) / \sigma_{s,\,th}$, for H-1 in H_2O and O-16, T = 293.6 K.
- For example, at E = 0.0253 eV, $\sigma_s(E;\,\text{O-16}) / \sigma_{s,\,th} \approx 1.032$ (FG) and ≈ 0.88 (O in H_2O)

S(α, β) for D_2O: need $\rho_{ph,i}(E)$ and $S_i(q)$, and c_i

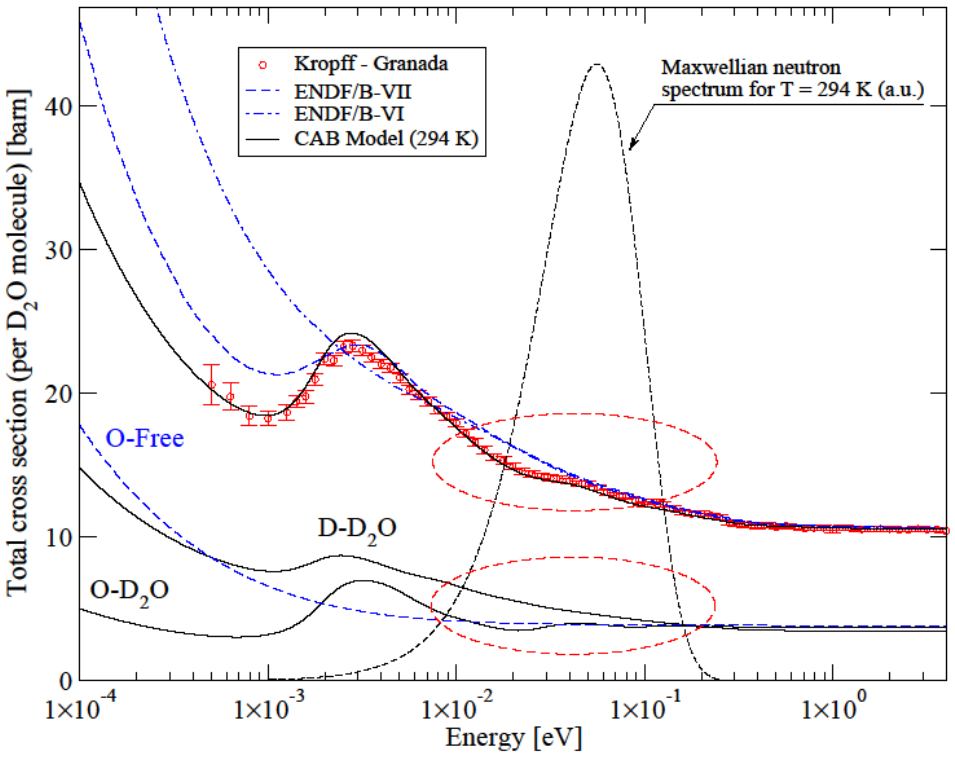


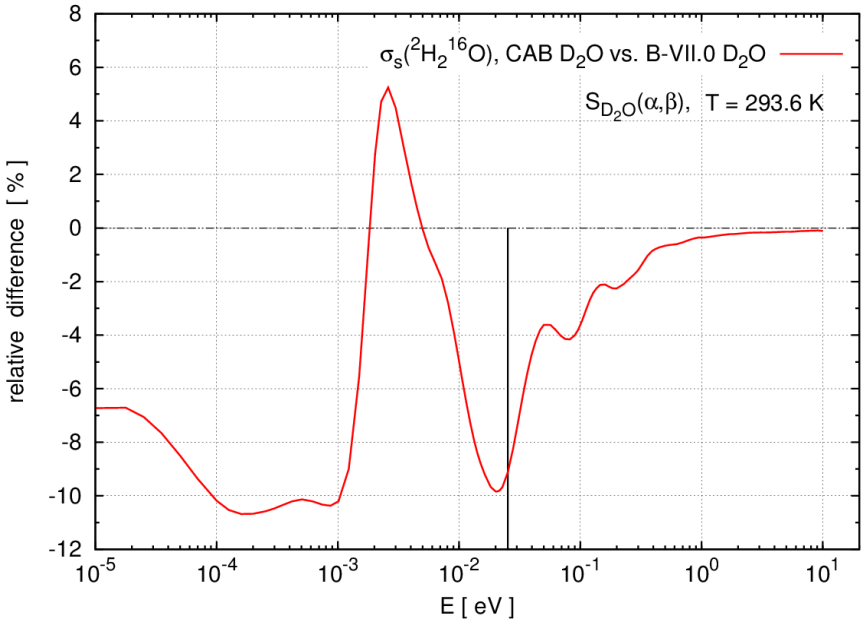




- $\rho_{ph,i}(E) \propto E^2$ at $E < 5$ meV and $\rho_{ph,i}(E) = 0$ at $E > E_{cut} \approx 0.12$ eV and $S_i(q) > 0$

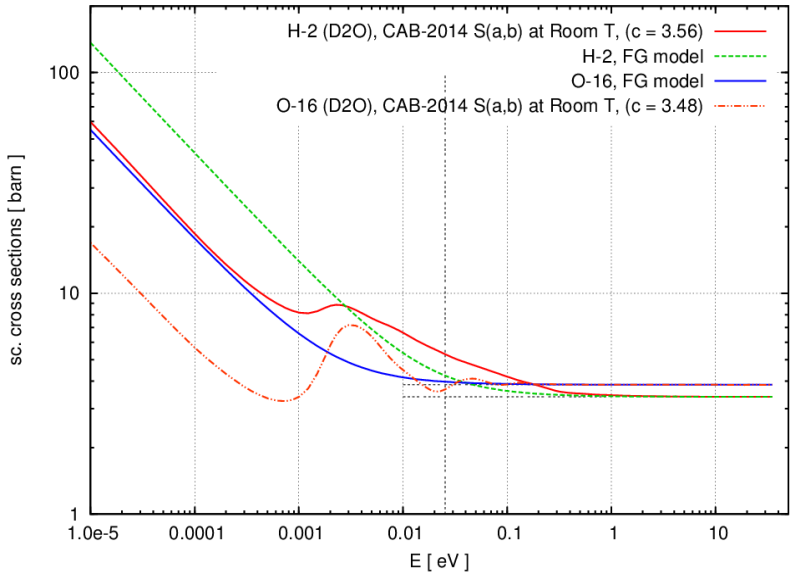
Result (D_2O): total cross sections in barn (per mol.)

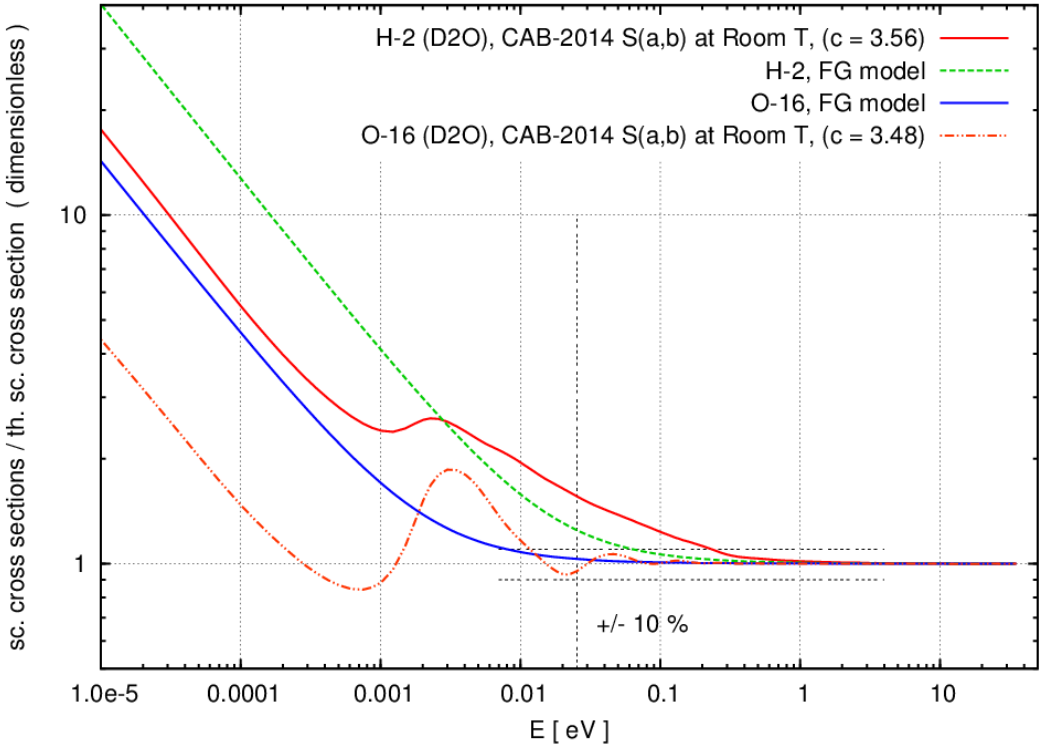




Total cross sections for heavy water (D_2O) at T = 294 K (exp. data: **Kropff-1984**) are compared with calculations based on the **CAB model**, and ENDF/**B-VII.0** (~ IKE model) and ENDF/**B-VI** (~ GA model).
Ellipses mark the differences in the total cross sections at the energies which are very important for accurate modeling of the critical systems with the thermal neutron spectrum.
We plot **Maxwellian spectrum** at RT (T_{eff} = 294 K, in a.u.), which would be expected for fully thermalized neutrons (shown for reference).

O-16 in D_2O (RT)



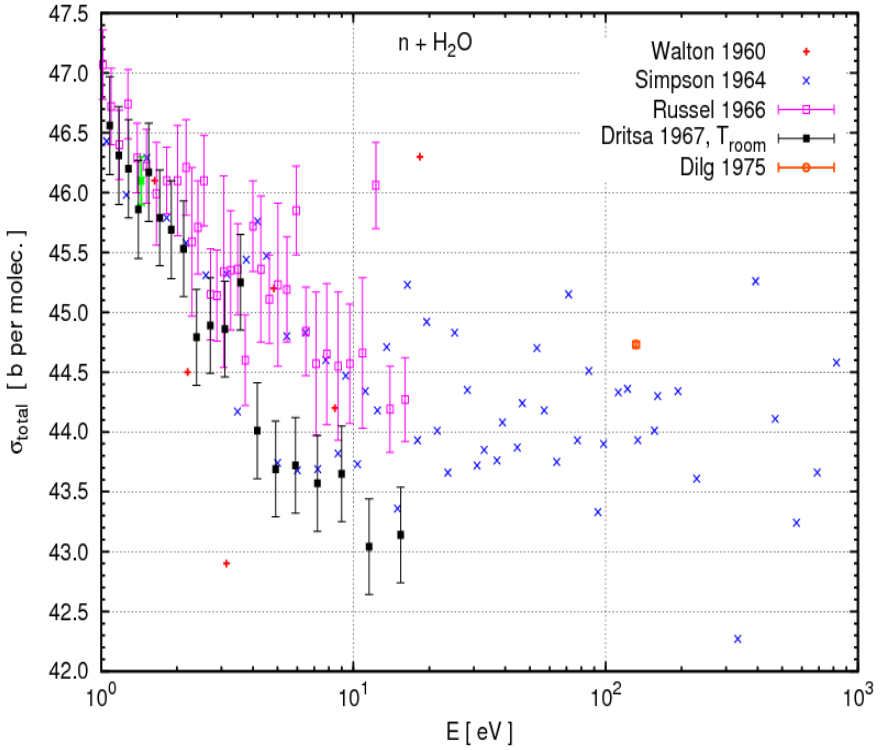


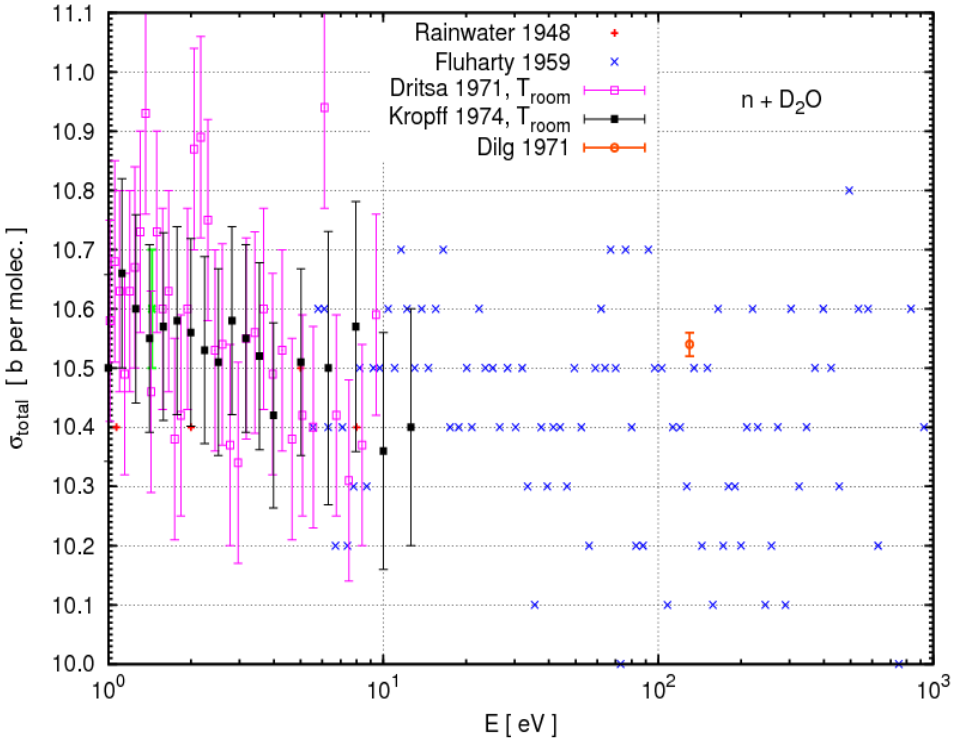
- At E ~ 10 eV, O-16 contributes ~ 36 % to the scattering cross sections of D_2O (barn per mol.)
- At E = 0.0253 eV,

  O-16 contributes ~ 26 % to the scattering cross sections of D_2O (using CAB model = S(α, β) with $\rho_{O\text{-in-}D2O}(E)$ and structure factor $S_O(q)$, **at RT**), $\sigma_s(E; O\text{-}16) / \sigma_{s,\,th} \approx 0.95$, and

  O-16 (FG) contributes ~ 25 % to the scattering cross sections of D_2O in ENDF/B-VII model, RT

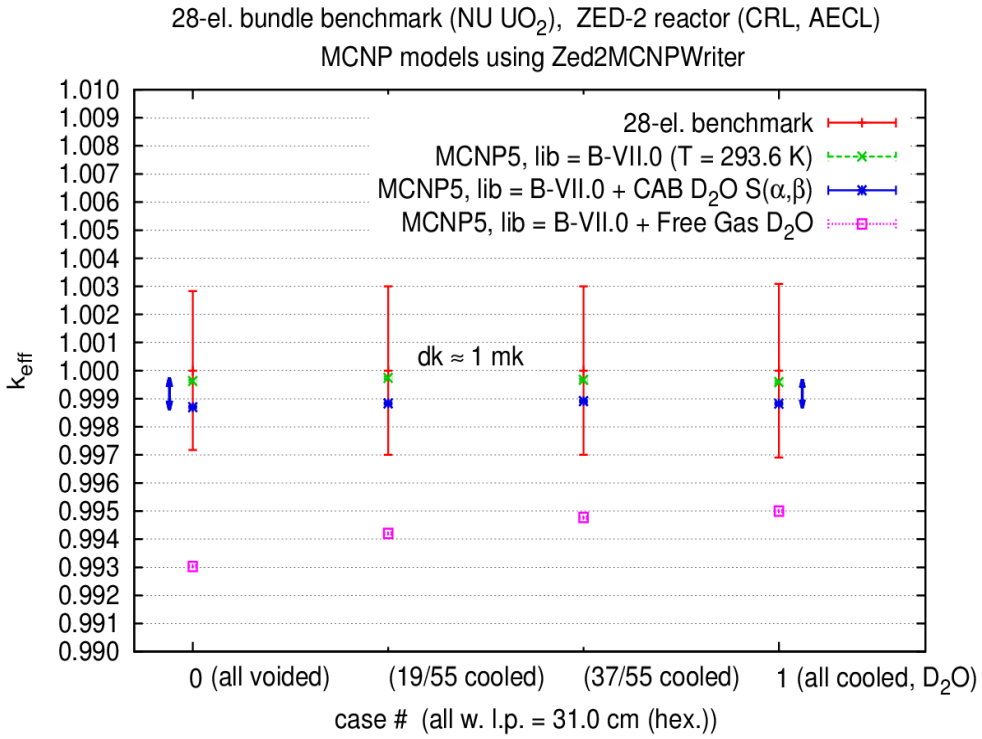
Again, E ~ 1 (4) – 10 (100) eV asymptotic of σ_s (σ_{tot}) vs. *E*: σ_s (E) ~ const, and where are we?

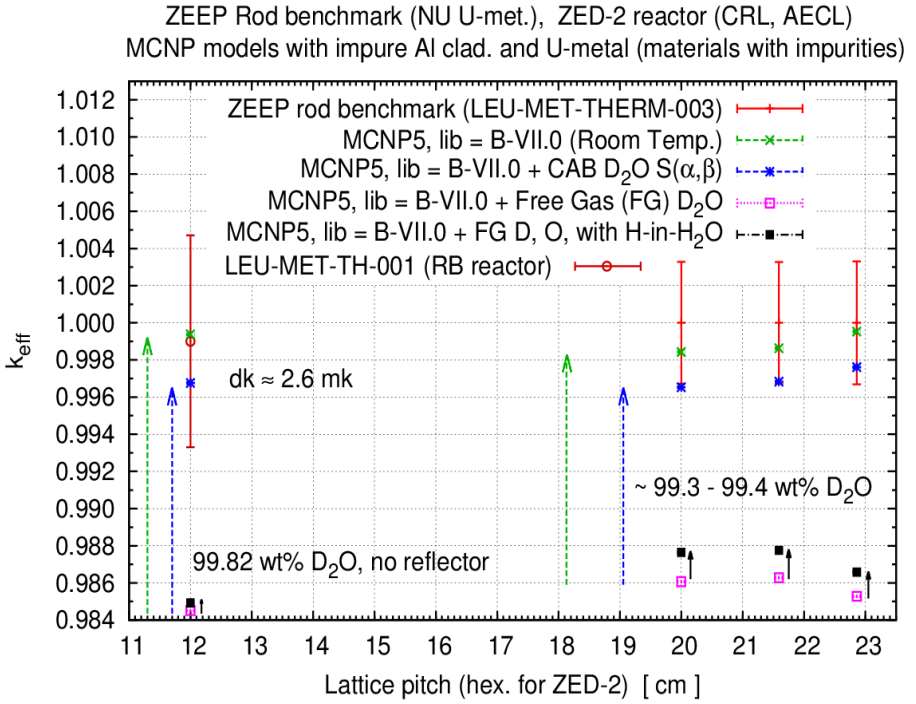




- **H_2O**: Dilg-1975 original result for H_2O,
  $\sigma_{tot}(H_2O, E = 132\ eV) = 44.731 \pm 0.027$ barn per molecule (± **0.06**% !), in comparison with other (similar) results found in EXFOR
- **D_2O**: Dilg-1971 original result for D_2O,
  $\sigma_{tot}(D_2O, E = 130\ eV) = 10.54 \pm 0.02$ barn per molecule (± **0.19**% !), in comparison with other (similar) results found in EXFOR.

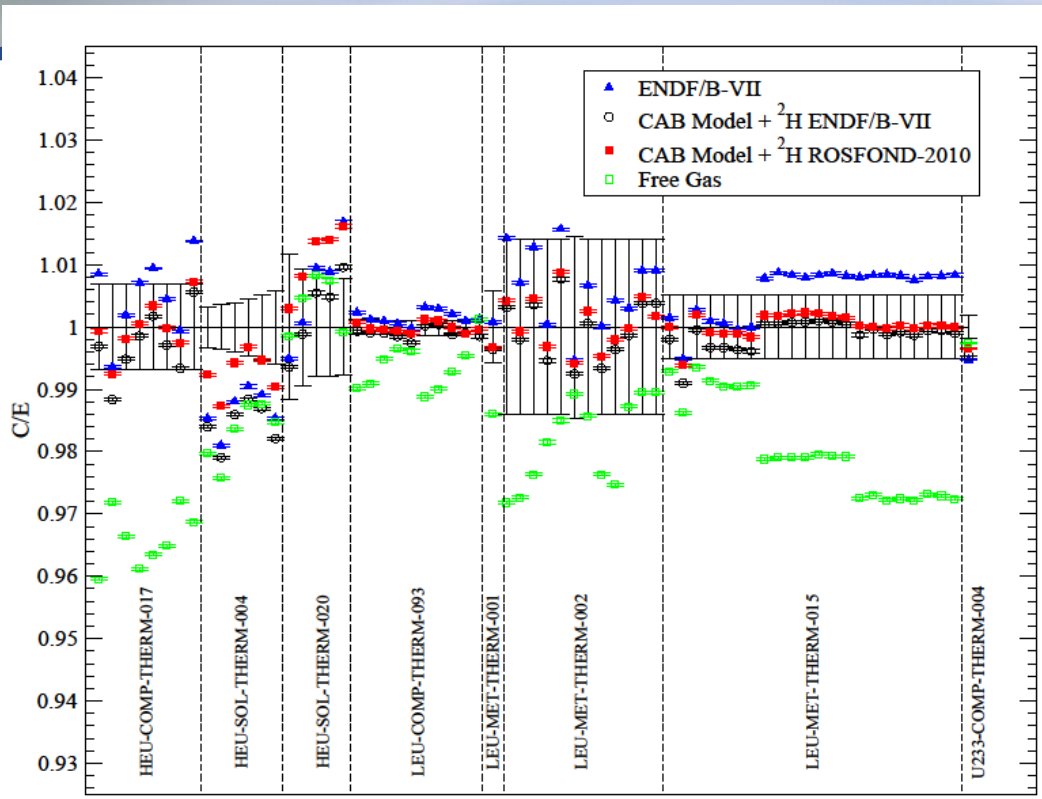
Testing S(α, β): often need to run with FG model





- We expect that k_{eff} (**CAB** D_2O) **<** k_{eff} (**B-VII.0** D_2O),
  but what is $dk = k_{eff}$ (B-VII.0 D_2O) – k_{eff} (CAB D_2O) = **?**
  answer: dk ≈ 100 pcm for ZED2-HWR-EXP-001 (ZED-2 reactor in CRL-AECL)
  $|\, dk_{CAB\text{-}VII.0} \,| < \Delta k_{Bench}$: **not conclusive...**

Testing: (almost) all thermal critical cases, D_2O moderated / reflected, near RT, from ICSBEP handbook



- Calculate **C/E ratio**, with **C** = k_{eff} calculated using MCNP5, lib. = ENDF/B-VII.0;
  E = benchmark $k_{eff} \pm \Delta k_{bench}$
- overall, it is an improvement if we change
  B-VII.0 D_2O S(α,β) → **CAB** D_2O S(α,β)

(the average change in the multiplication factor is $dk_{CAB - VII.0} \approx 5.43$ mk = 543 pcm)

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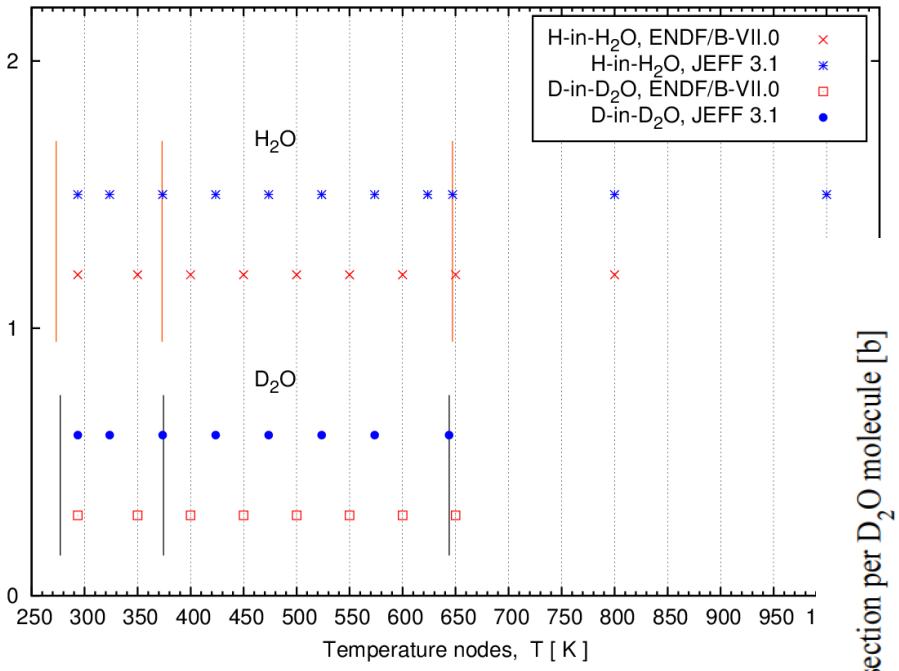
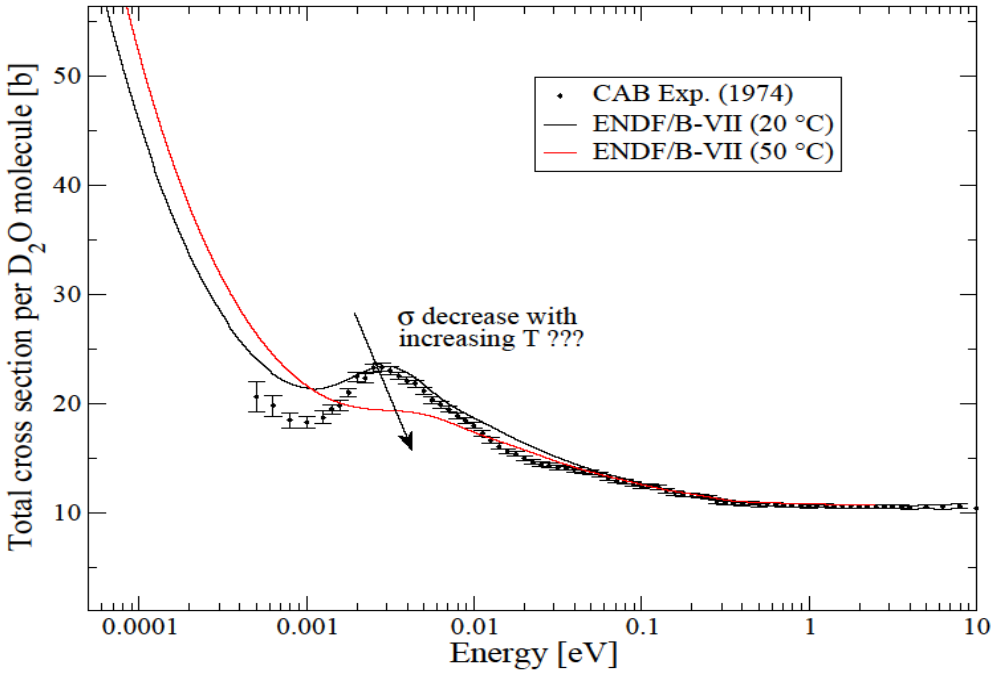
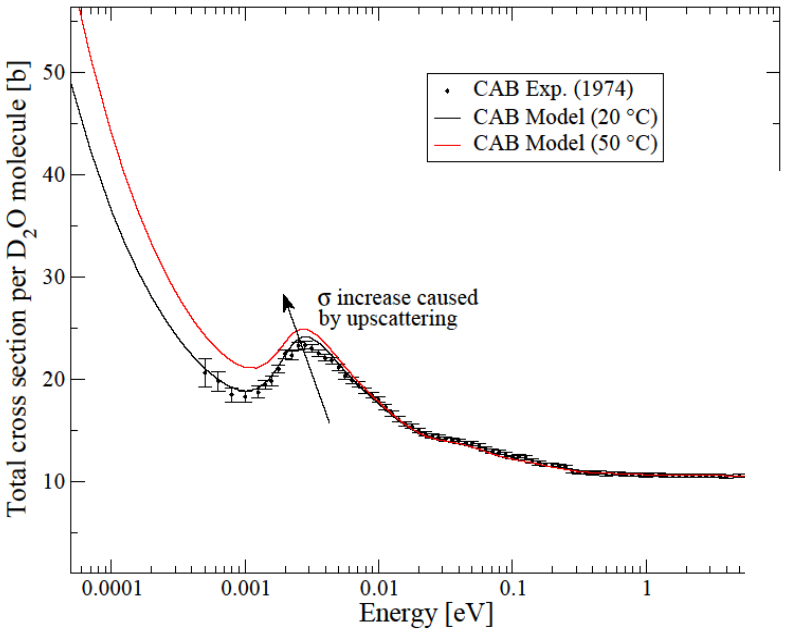
Toward new evaluation of heavy water D_2O S(α,β): from RT to 50-70 deg C

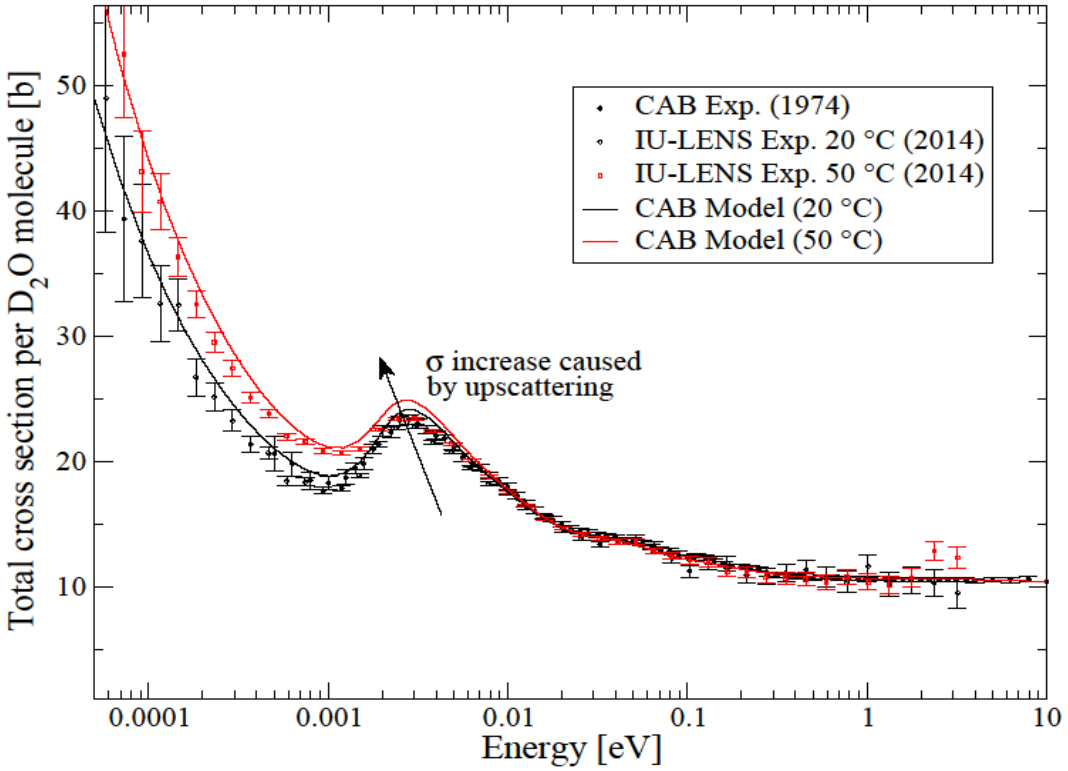




- Temp. nodes (ENDF/B-VII *vs.* JEFF-3): e.g., T = 323.15 K

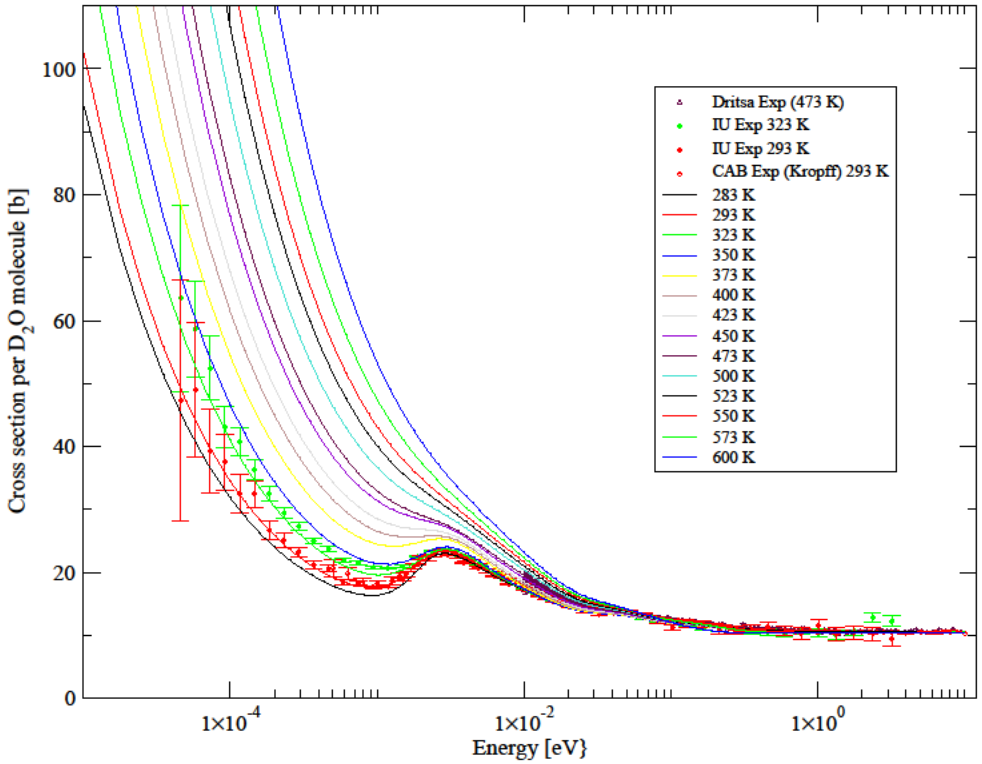
Toward new evaluation of heavy water D_2O S(α,β): from RT to 50 deg. C



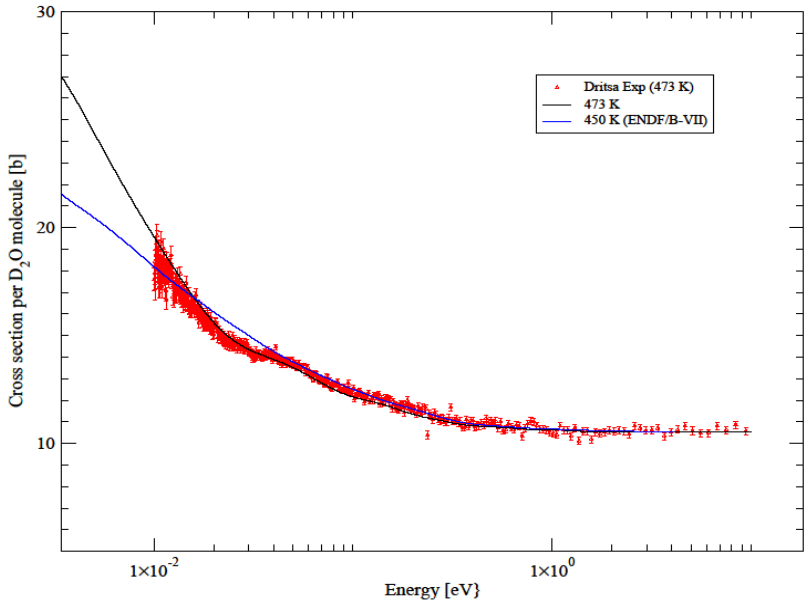


- New experiments (July 2014) at the LENS facility in Indiana University (IU, USA)
- Prof. David V. Baxter, http://www.indiana.edu/~iubphys/faculty/baxterd.shtml

Toward a new evaluation of heavy water D_2O S(α,β): we extend # of T nodes

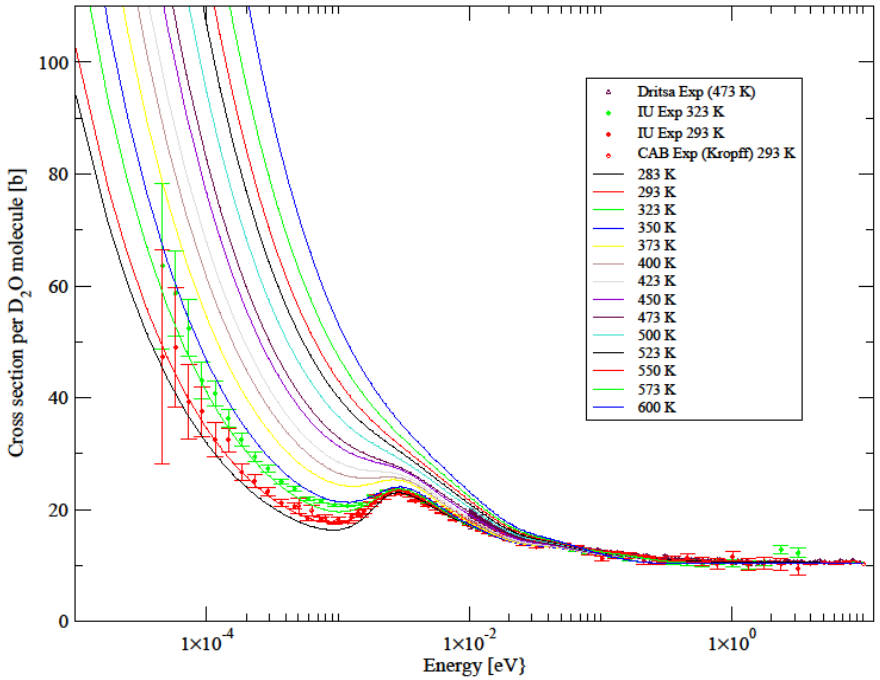


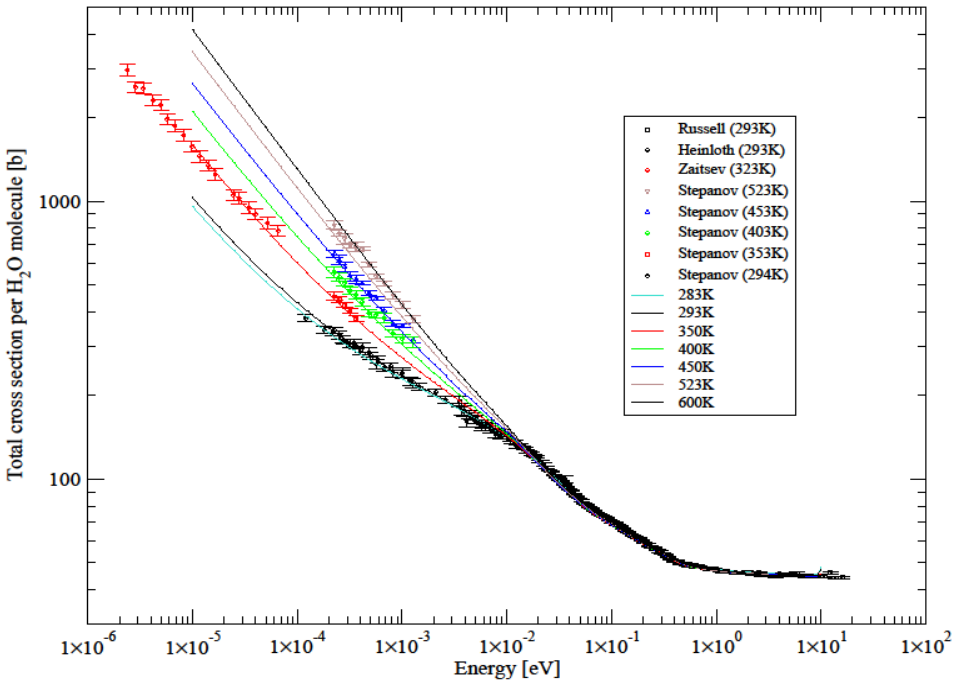
Dritsa: ~ 200 deg. C;
More data at high T ?



- Add one low temperature node (~ 10 deg. C) and combine ENDF/B-VII and JEFF-3 temperature nodes (up to 600.0 K at the moment)
- **Liquid** D_2O: at T > 3.8 deg. C = 276.95 K (at p = 1 atm.)
- At *T* > ~ 371 deg. C ~ 643.9 K ($p > p_c$): supercritical fluid (heavy water)

Toward a new evaluation of water H_2O / D_2O S(α,β) : we extend # of T nodes





- H_2O: can go beyond incoherent approximation to get scattering cross sections for cold neutrons better (E < 1 meV)

Conclusion

- The **new evaluation** (in endf) for thermal scattering law files for **heavy** / light **water** is under development and testing.

  It represents an improvement over existing scattering law files available in the modern evaluated nuclear data libraries (ENDF/B, JEFF, JENDL)

- When the new thermal scattering libraries are applied to the calculation of **international neutron criticality benchmarks** (ICSBEP Handbook), we find a significant (up to ~ 11 mk = 1100 pcm) difference in the results of multiplication factors, and improving the calculation in ≈ 60% of the critical cases.

- New measurements started: at **IU-LENS** (with D. Baxter, 2014) and also at CRL (**NRU reactor n-beam** and using a triple-axis spectrometer)

J.I. Márquez Dámian, J.R. Granada, and D.C. Malaspina, "*CAB models for water: A new evaluation of the thermal neutron scattering laws for light and heavy water in ENDF-6 format*," Annals of Nuclear Energy, Vol. **65**, pp. 280-289, **2014** (March).

J.I. Márquez Dámian, J.R. Granada, and D. Roubtsov, "*Improvement on the calculation of D_2O moderated critical systems with new thermal neutron scattering libraries*," Annals of Nuclear Energy, Vol. **71**, pp. 206-210, **2014** (September).

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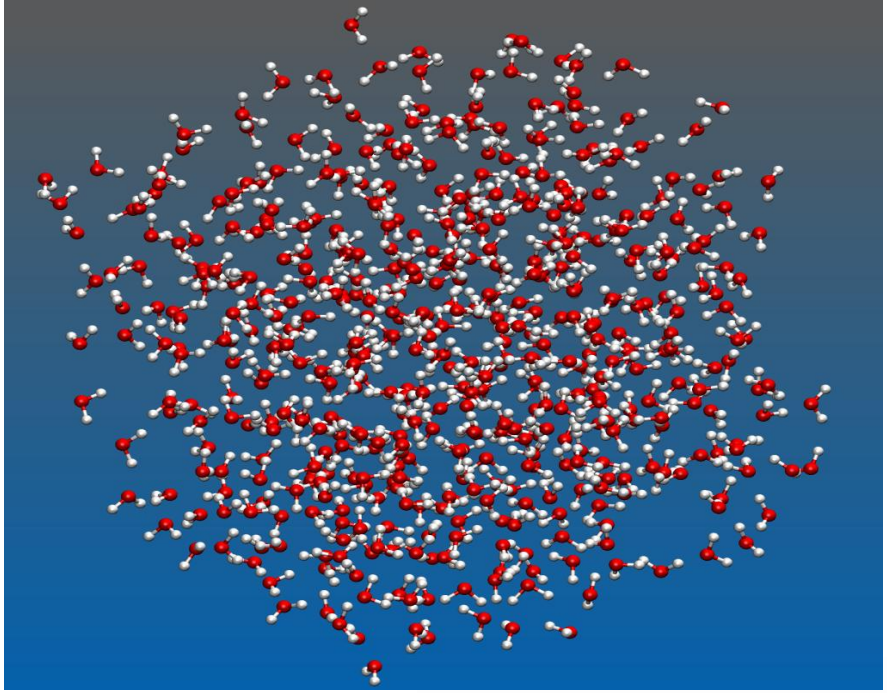

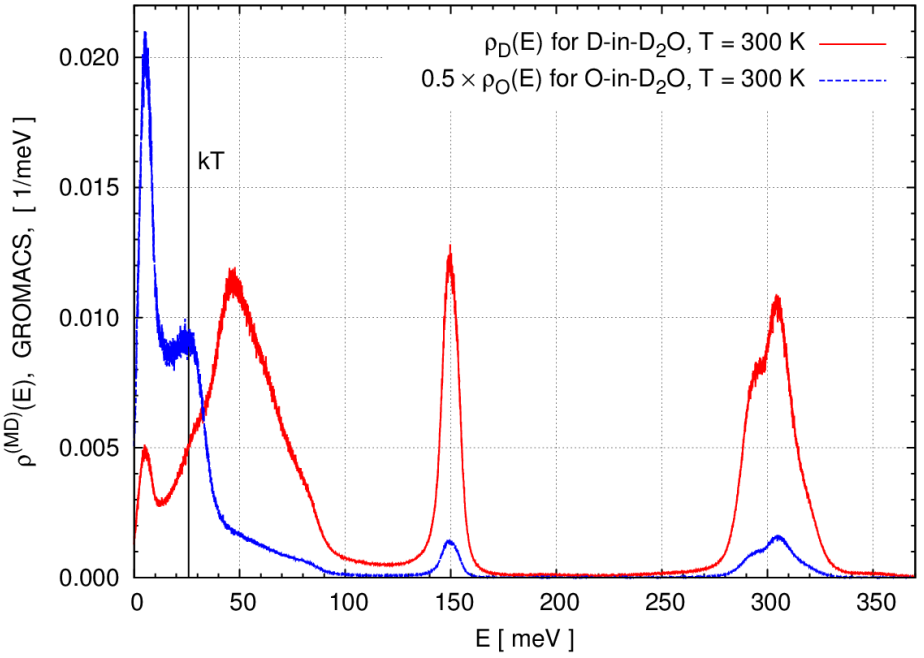
ρ_D(E) for D-in-D_2O, T = 300 K
0.5 × ρ_O(E) for O-in-D_2O, T = 300 K
kT
$\rho^{(MD)}$(E), GROMACS, [1/meV]
0.020
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0.010
0.005
0.000
0
50
100
150
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250
300
350
E [meV]

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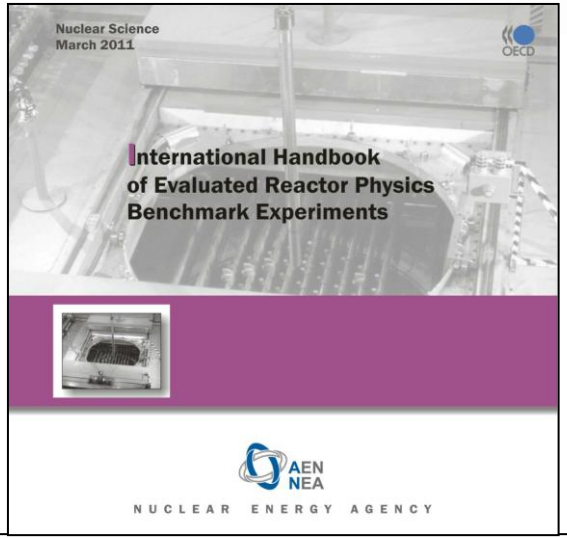
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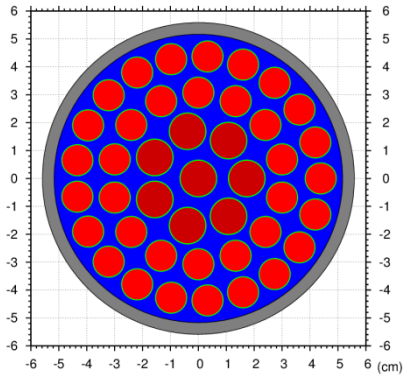
6
5
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-6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6 (cm)