DE LA RECHERCHE À L'INDUSTRIE

Covariance matrices of the neutron thermal scattering law of light water for reactor applications

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SG-42: Thermal Scattering Kernel S(α,β)

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PRESENTATION PLAN

- Motivations and objectives
- Brief recall of neutron thermal scattering
- TSL models for ^1H in H_2O
- Uncertainty quantification of the models
- Uncertainty propagation to microscopic data
- Conclusions

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MOTIVATIONS

1. New and modern techniques for producing thermal scattering data (TSL) are available nowadays (lattice dynamics/molecular dynamic simulation codes).

2. No covariance matrices for any TSL were reported.

Material number	JEFF-3.1.1	ENDF/B-VII	JENDL-4.0
1	H(H20)	H(H2O)	H(H2O)
2		Para Hydrogen	Para Hydrogen
3		Ortho Hydrogen	Ortho Hydrogen
7	H(ZrH)	H(ZrH)	H(ZrH)
8	H(CaH2)		
11	D(D2O)	D(D20)	D(D2O)
12		Para Deuterium	Para Deuterium
13		Ortho Deuterium	Ortho Deuterium
26	Be metal	Be metal	Be metal
27		Be(BeO)	Be(BeO)
28		O(BeO)	
31	Graphite	Graphite	Graphite
33		Liquid Methane	Liquid Methane
34		Solid Methane	Solid Methane
37	H(CH2)	H(CH2)	
40		Benzine	Benzine
45		Al metal	
52	Mg metal		
56		Fe metal	
58		Zr(ZrH)	Zr(ZrH)
59	Ca(CaH2)		
75		O(UO2)	
76		U(UO2)	
Number of materials	**9**	**20**	**14**

OBJECTIVES

- Analyze microscopically the differences between the TSL of ^{1}H in H_2O of IKE (JEFF-3.1.1 and ENDF/B-VII.1) and CAB model (molecular dynamic simulations).

- Produce covariance matrices for both models and propagate uncertainties to microscopic data.

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THERMAL NEUTRON INELASTIC SCATTERING

Total cross section $\sigma_{tot}(E) = \sigma_\gamma(E) + \sigma_n(E)$

Capture cross section $\sigma_\gamma(E) = \sigma_{\gamma 0}\sqrt{\frac{E_0}{E}}$ ($\sigma_{\gamma 0}$ = 0.332 b ^1H)

Inelastic scattering cross section $\sigma_n(E) = \iint \frac{d^2\sigma}{d\Omega dE} dE' d\Omega = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}} S(\alpha,\beta)$

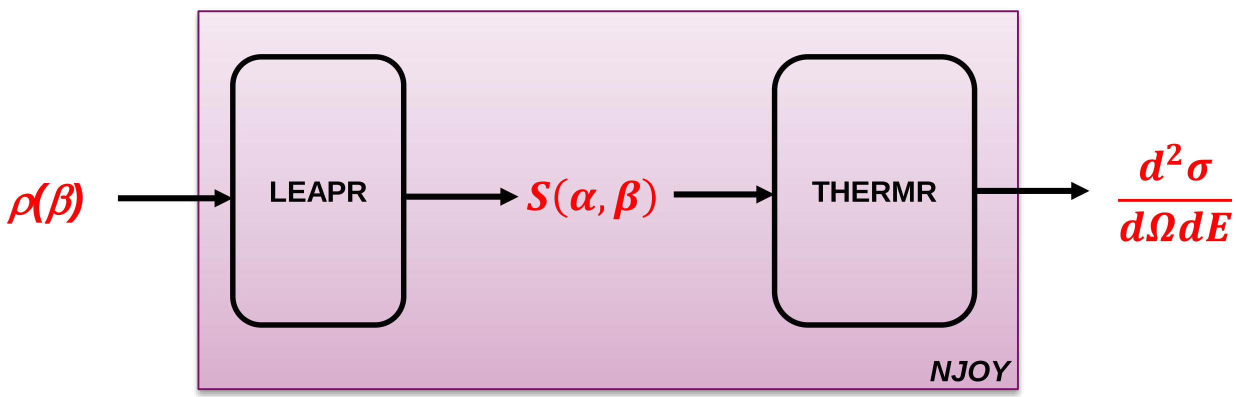
Double differential inelastic scattering cross section

Thermal scattering function

$S(\alpha,\beta) = f[\boldsymbol{\rho(\beta)}]$

The dynamics of the scattering target (H_2O) are defined by the frequency spectrum $\boldsymbol{\rho(\beta)}$

The evolution of S(α,β) is done with LEAPR module in the *incoherent* and *Gaussian* approximation



The main effort of the research groups resides in providing a frequency spectrum ρ(β)

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TSL MODELS FOR ^{1}H IN H_2O

IKE MODEL (Mattes and Keiniert – Stuttgart)

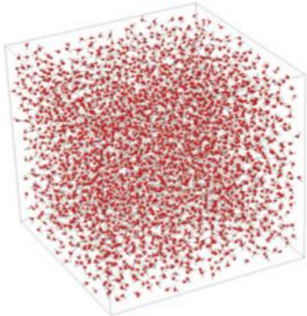
- The intermolecular vibration modes are based on experimental measures.
- The molecular translation, is represented with the Free Gas Law.
- The intramolecular vibration modes are described by 2 discrete oscillators.
- ^{16}O is treated as free gas.
- JEFF-3.1.1 and ENDF/B-VII.1 nuclear data libraries

M. Mattes and J. Keinert, Thermal Neutron Scattering Data for the moderator Materials H2O, D2O and ZrHx in ENDF-6 Format and as ACE Library for MCNP(x) Codes. International Nuclear Data Committee - 0470 (2005).

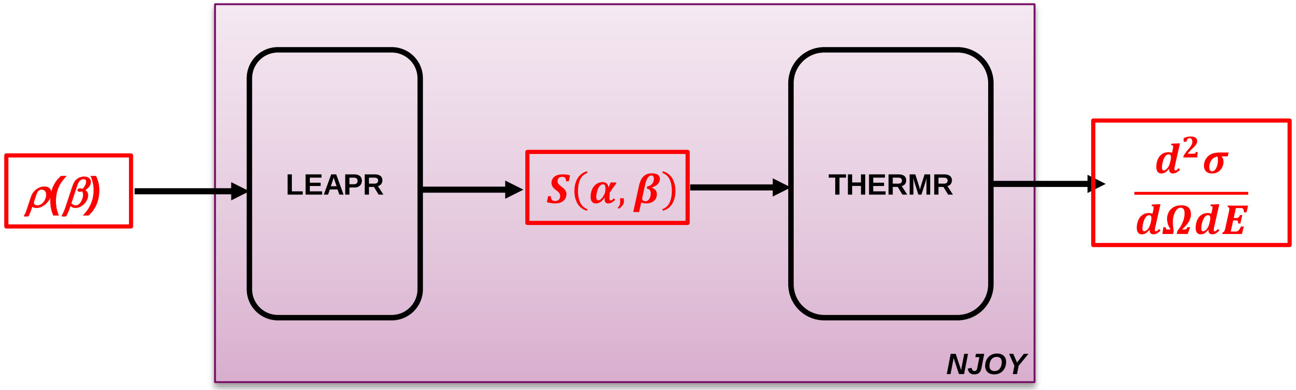
CAB MODEL (J. I. Marquez - Argentina)

- The intermolecular vibration modes are based on molecular dynamic simulations.
- The molecular translation is represented by a Diffusion model established by Egelstaff and Schofield.
- The intramolecular vibration modes are described by 2 discrete oscillators.
- ^{16}O is treated as free gas.

J.I. Marquez Damian et al., Ann. Nucl. Energy 65, 280 (2014)



COMPARISON OF TSL MODELS FOR ^1H IN H_2O

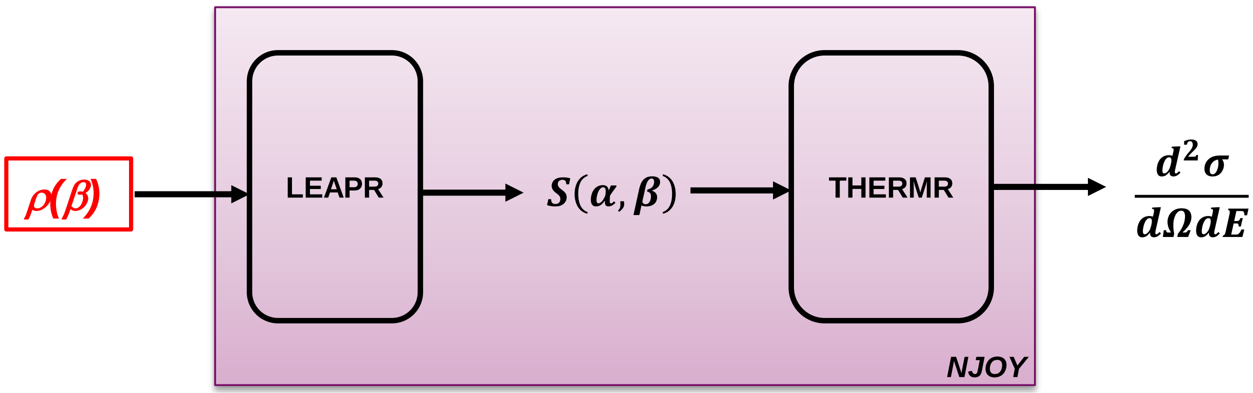


Frequency spectrum

Thermal scattering function

Double differential inelastic scattering cross section

COMPARISON OF TSL MODELS FOR ^{1}H IN H_2O



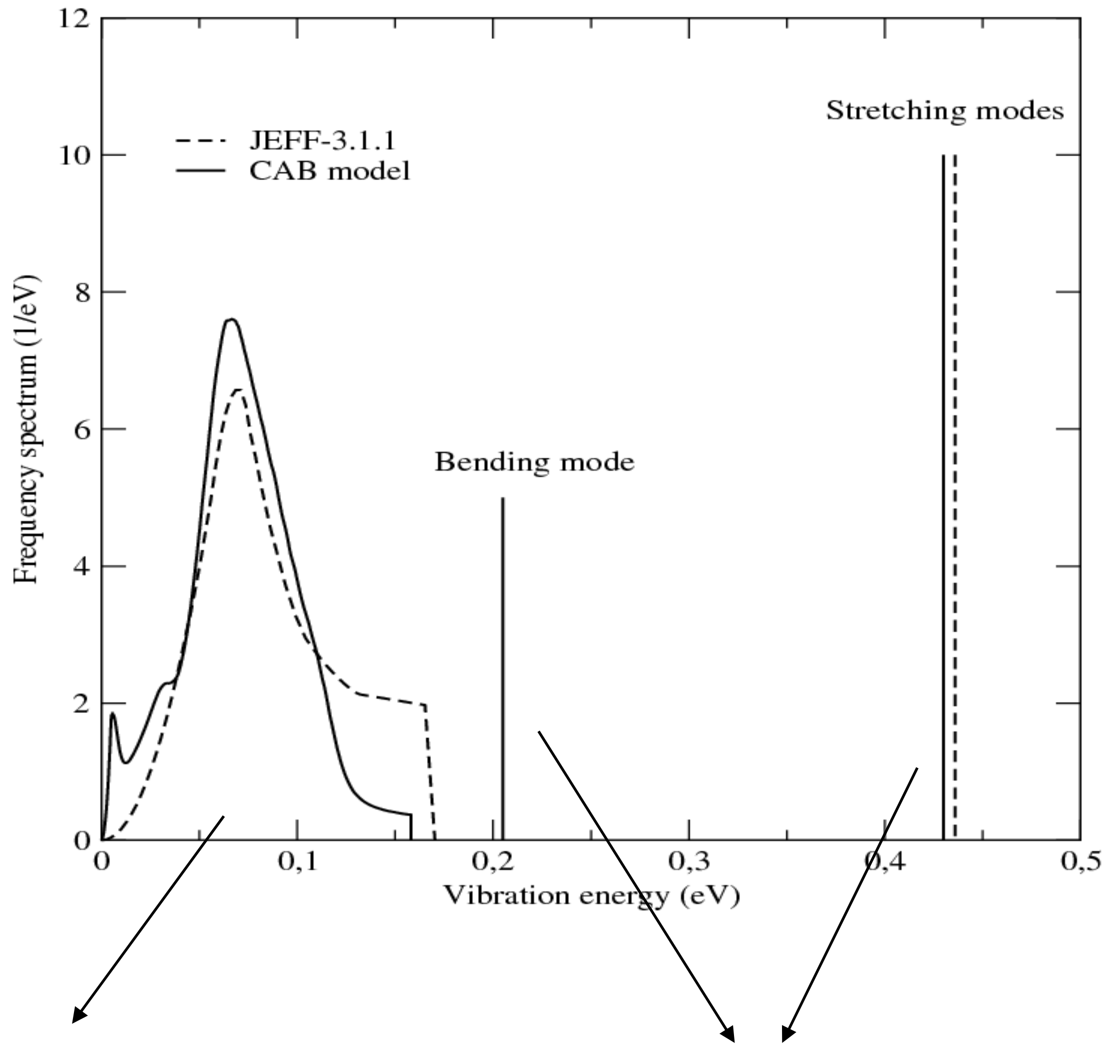
Frequency spectrum

Thermal scattering function

Double differential inelastic scattering cross section

FREQUENCY SPECTRUM OF ^1H IN H_2O AT 294 K

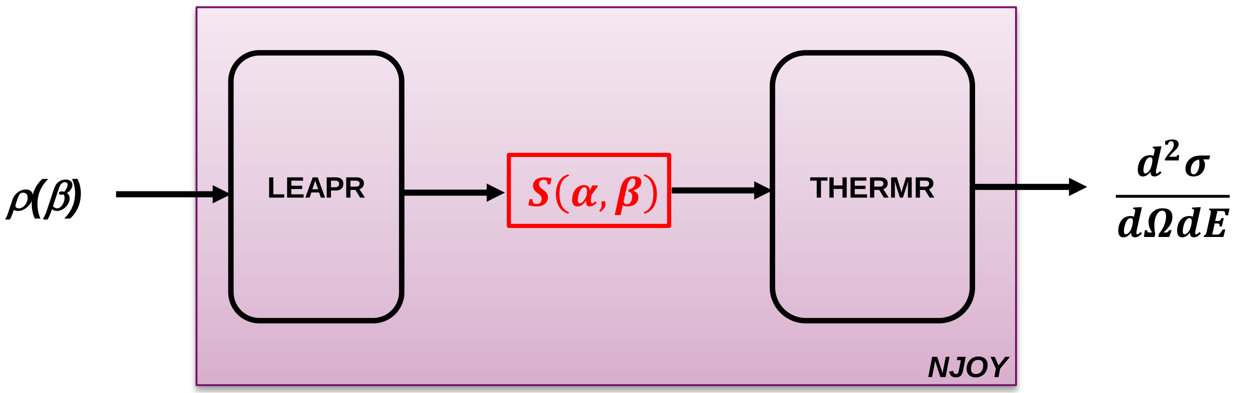
- The contribution of ^{16}O in H_2O is negligible for light water



Intramolecular vibration modes

Internal modes of the molecule

COMPARISON OF TSL MODELS FOR ¹H IN H_2O



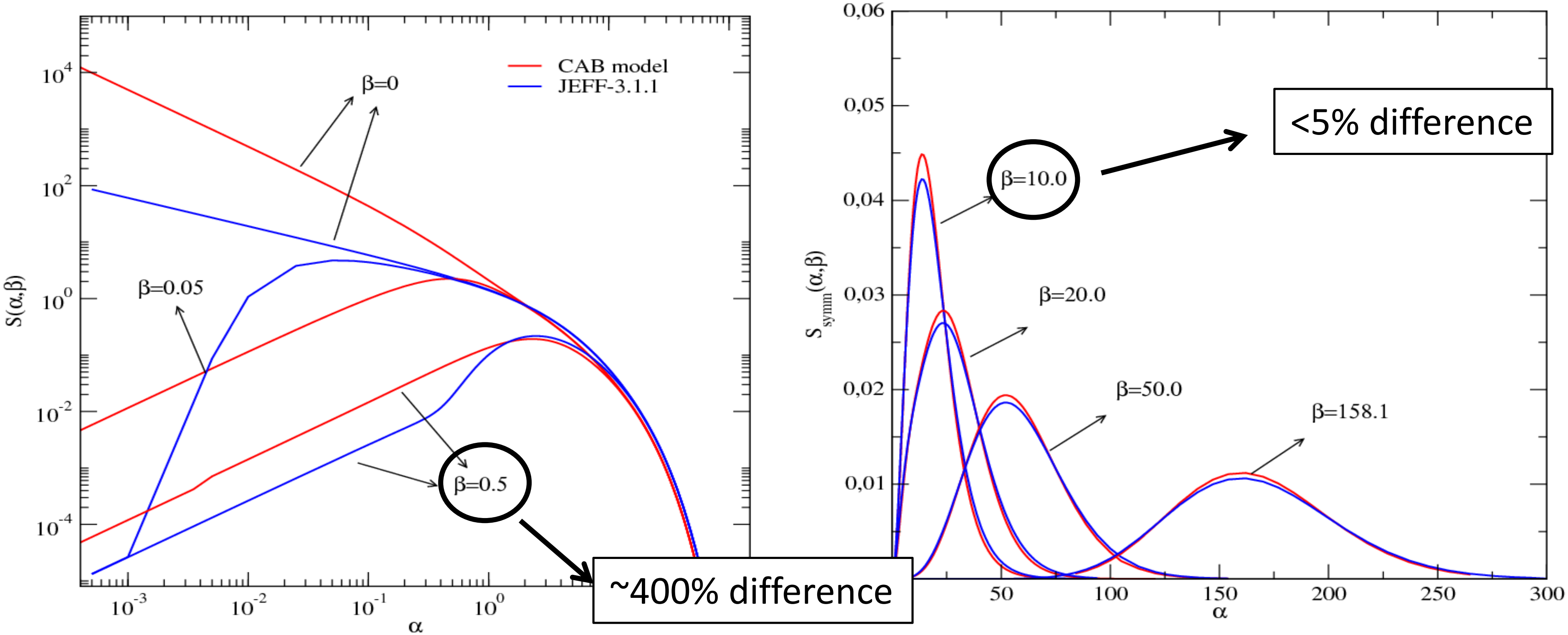
Frequency spectrum

Thermal scattering function

Double differential inelastic scattering cross section

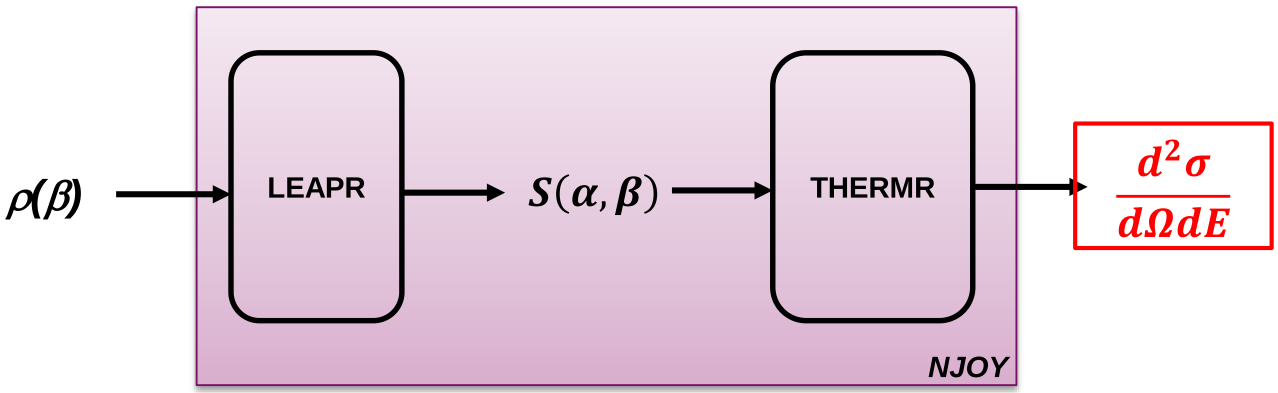
S(α,β) FUNCTION PARAMETRIZED WITH β AT 294 K

S(α,β) output from LEAPR ⟶ $S_{sym}(\alpha,\beta) = S(\alpha,\beta)e^{\beta/2}$



No visible differences at large energy transfer. On the other hand, an impact is expected at very small energy exchange < 15 meV (β = 0.5)

COMPARISON OF TSL MODELS FOR ^1H IN H_2O



Frequency spectrum

Thermal scattering function

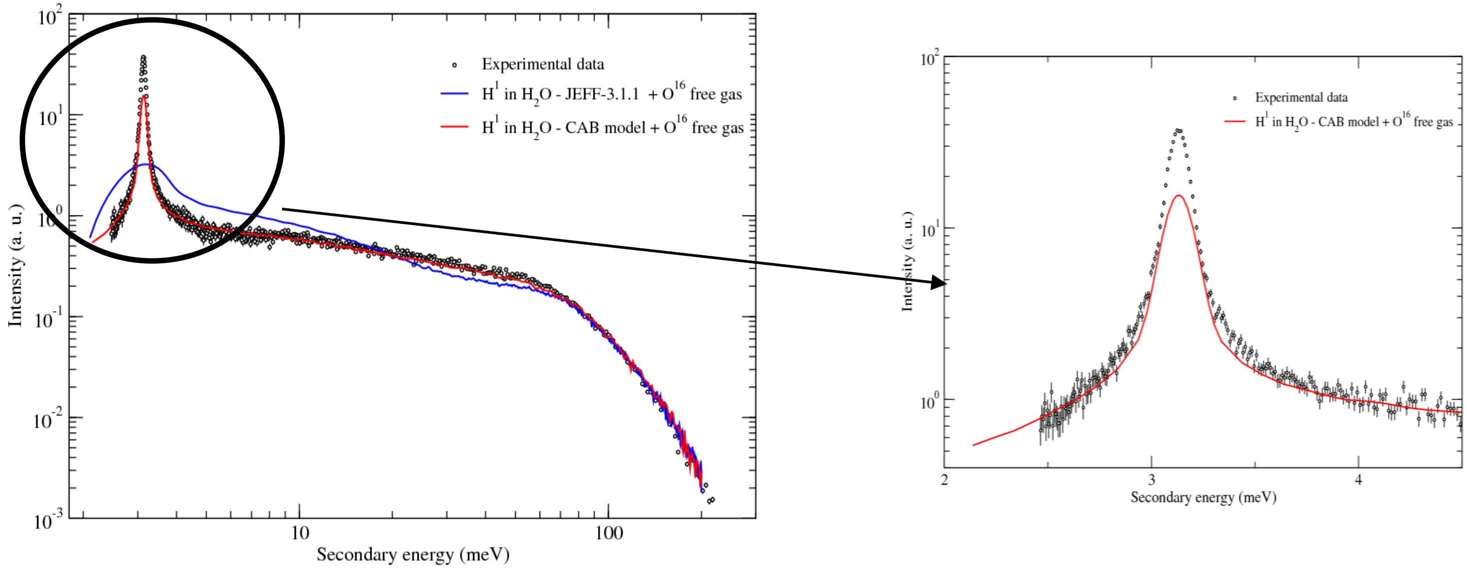
Double differential inelastic scattering cross section

DOUBLE DIFFERENTIAL CROSS SECTION AT 294 K

We have performed experimental measures of the H_2O double differential cross section at the ILL Institute (Refer to Gilles Noguere Talk). Here are the results for:

- Incident neutron energy E_0 = 3 meV
- Scattering angle θ = 15°

The continuous lines were obtained with a ToF simulation in the Monte Carlo code TRIPOLI4, using the TSL evaluation files of JEFF-3.1.1 and CAB model.



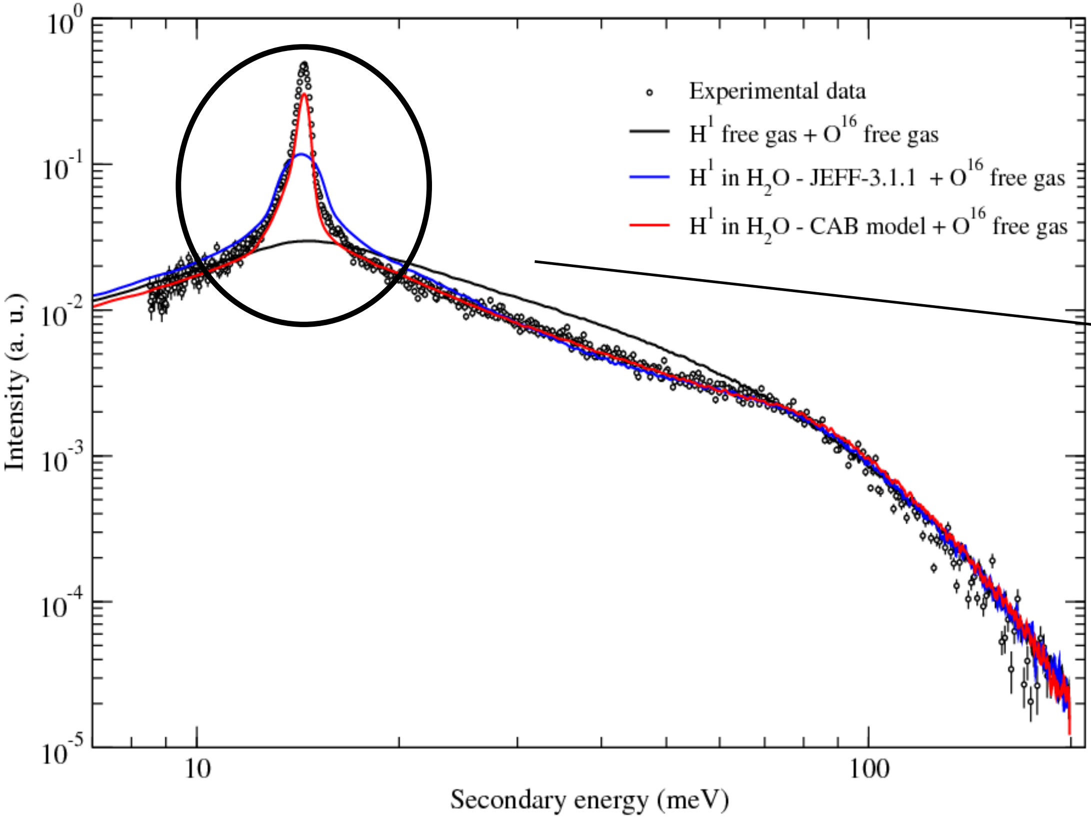
- JEFF-3.1.1 has problems in reproducing the experimental data. The cold incident neutron energy reveals the weaknesses of the model, mainly driven by the use of a free gas model to describe the molecular translational.

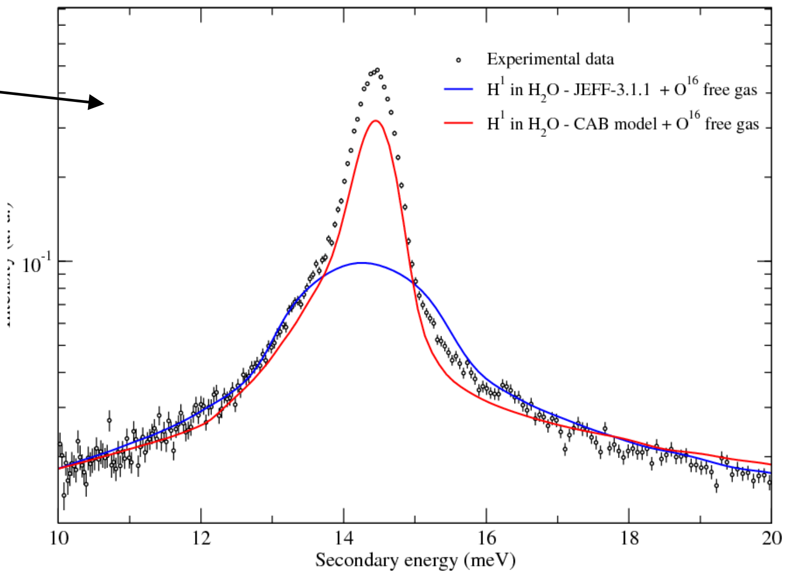
- Better agreement of CAB model (incorporation of a diffusion model). But not fully suitable to account the whole shape of the quasi-elastic peak?? Background problems?? Multiple scattering correction??. Still, the comparison between models is valid.

DOUBLE DIFFERENTIAL CROSS SECTION AT 294 K

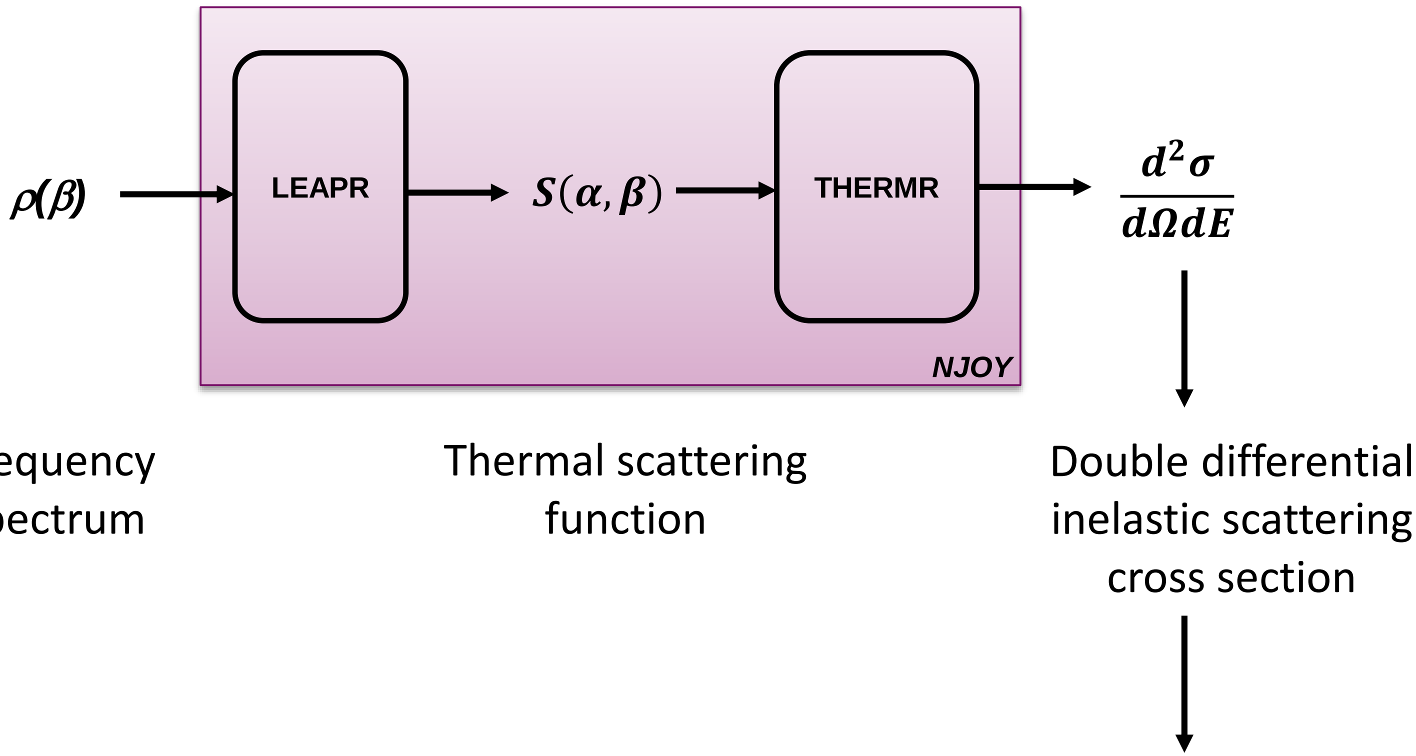
IN4c

- Incident neutron energy E_0 = 14 meV
- Scattering angle θ = 15°



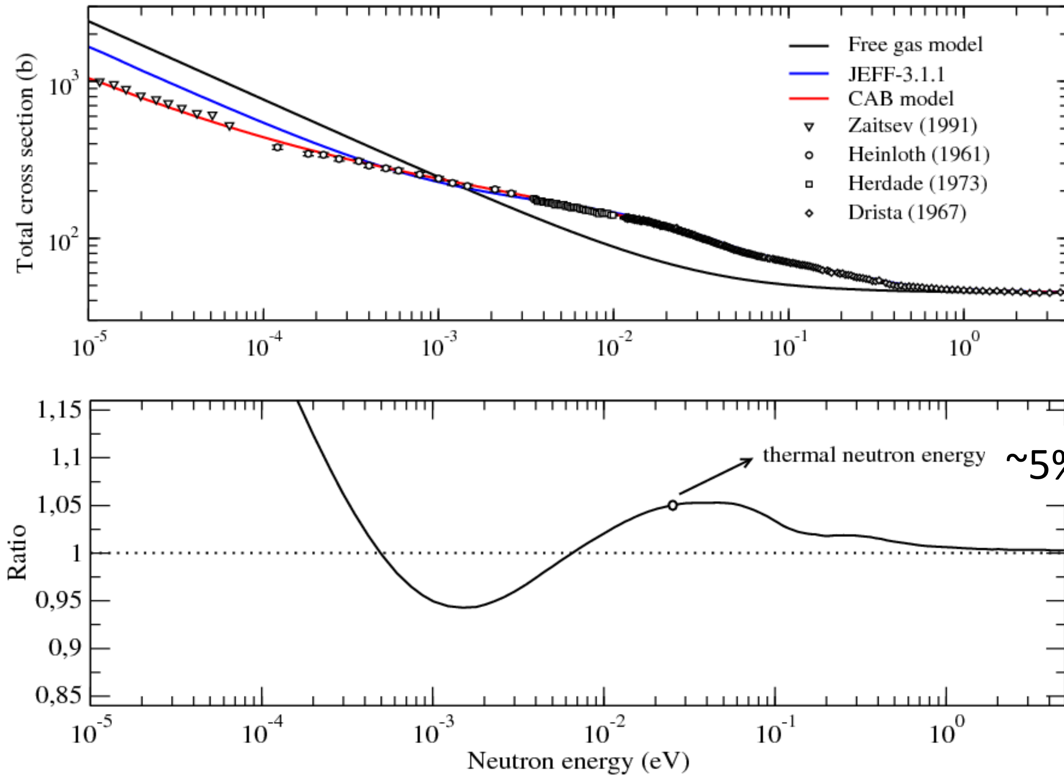


COMPARISON OF TSL MODELS FOR ^{1}H IN H_2O



Total cross section $\sigma_{tot}(E) = \sigma_\gamma(E) + \sigma_n(E)$

H_2O TOTAL CROSS SECTION AT 294 K



~5% difference

- Good agreement between the CAB model and the experimental data throughout all the neutron energy.
- Sizeable discrepancies between the CAB model and JEFF-3.1.1 for the cold neutron energy range (E<0.2 meV).

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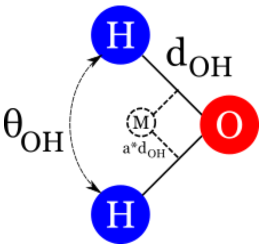
TSL UNCERTAINTIES QUANTIFICATION

- The present methodology was applied for JEFF-3.1.1 and for CAB model. Results for covariance matrices of H1 in H2O for JEFF-3.1.1 library are already published in *G. Noguere et al. Ann. Nucl. Ener. 104, 132-145 (2017).*

- The method will be explained for CAB model

✓ The quantification of the CAB model uncertainties implies the generation of the covariance matrix between the model parameters.

Model parameter vector (TIP4P/2005f water potential) $\boldsymbol{\overline{p} = (\epsilon_0, \sigma_0, q, D_{OH}, \beta_{OH}, d_{OH}, k_\theta, \theta_0, a)}$



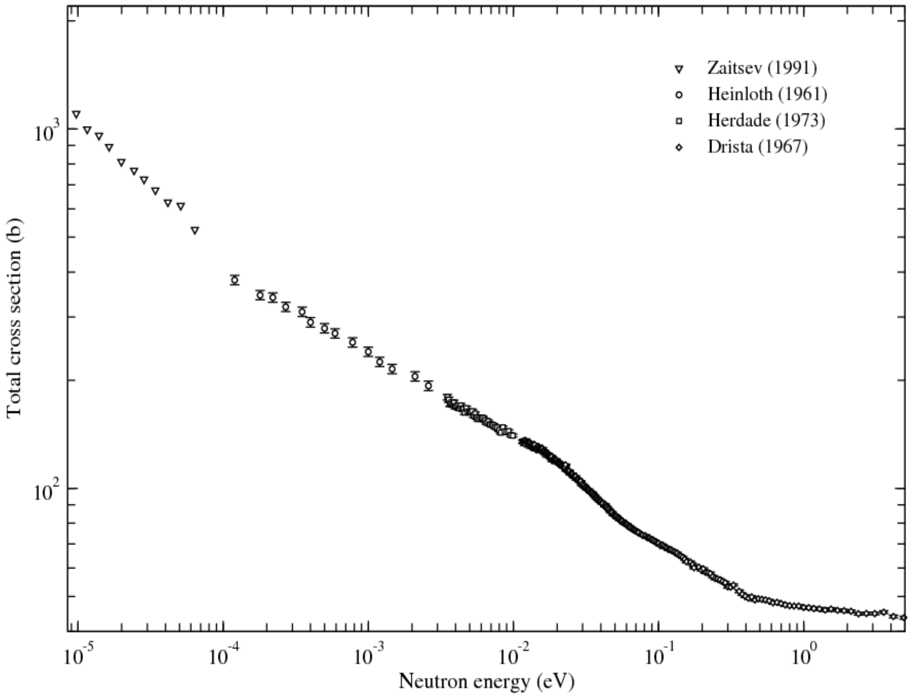
M. A. Gonzalez and J. L. F. Abascal, Jour. Chem. Phys. 135, 224516 (2011)

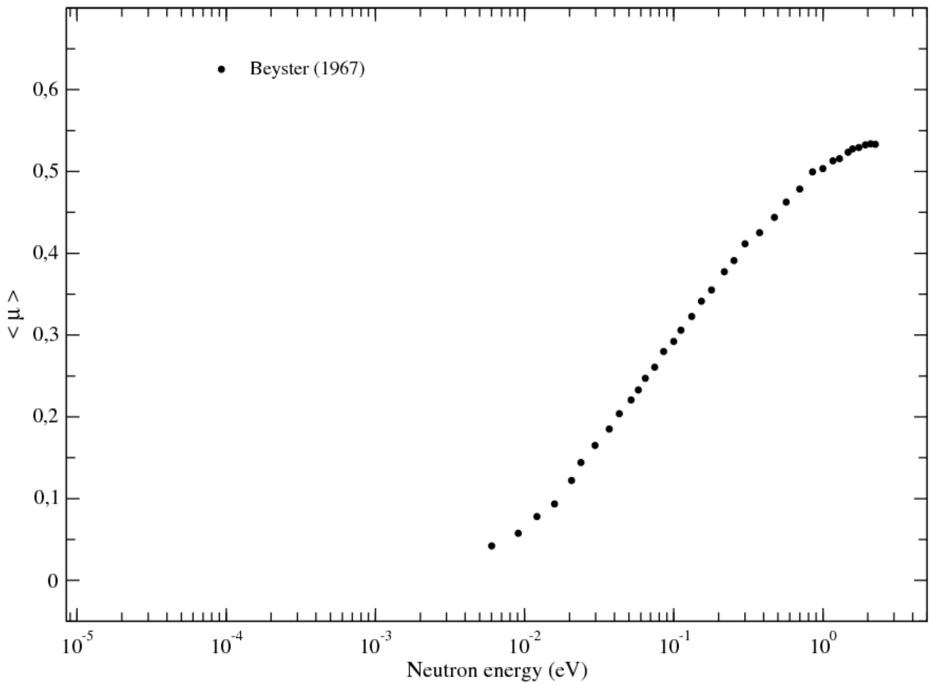
✓ The uncertainties will be quantified by fitting the parameters with an “appropriate” set of experimental data. The new parameters will give a theory that will reproduce as close as possible the selected data (**Generalized Least Square Method GLS**)

CONRAD code
(code for nuclear reaction analysis and data assimilation)

TSL UNCERTAINTIES QUANTIFICATION

Relevant experimental data included in the fitting procedure: the **H_2O cross section** and the **average cosine of the scattering angle $\bar{\mu}$** (important physical quantities for reactor applications).





TSL UNCERTAINTIES QUANTIFICATION FOR CAB MODEL

Definition of the “nuisance” parameters

Experimental parameters :

- ✓ Generally, the experimental uncertainties are poorly published. So we don’t have access to clear information about the normalization **N** used or the measured background **B** and the fluctuation of the temperature **T**.

- ✓ The sample thickness **t** used in the experiment (related to the numerical density) is another parameter to be treated separately and normally the expérimentateur provides this information.

Fixed model parameters :

- ✓ In CAB model the diffusion mass (translational weight $\boldsymbol{\omega_t}$ in LEAPR) was deduced experimentally from “*A. G. Novikov et al., Journal of Structural Chemistry 31,77 (1990)*”. This parameter is penalized with 10% of uncertainty (no published uncertainty on these data).

- ✓ The uncertainty of the ^{1}H free atom cross section $\boldsymbol{\sigma_\infty}$ was taken from the neutron cross section standards of IAEA (2006).

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b}{4\pi kT}\sqrt{\frac{E'}{E}}S(\alpha,\beta)$$

$$\sigma_b = \left(1+\frac{1}{A}\right)^2 \sigma_\infty$$

$$\sigma_\infty = 20{,}436 \pm 0{,}2\%\ b$$

TSL UNCERTAINTIES QUANTIFICATION FOR CAB MODEL

Calculation of the final covariance matrix

Model parameter vector (TIP4P/2005f water potential)

$$\overline{\mathbf{p}} = (\boldsymbol{\epsilon}_0, \boldsymbol{\sigma}_0, \mathbf{q}, \mathbf{D}_{\mathbf{OH}}, \boldsymbol{\beta}_{\mathbf{OH}}, \mathbf{d}_{\mathbf{OH}}, \mathbf{k}_{\boldsymbol{\theta}}, \boldsymbol{\theta}_0, \mathbf{a})$$

"Nuissance" parameter vector

$$\overline{\boldsymbol{\theta}} = (\mathbf{N}, \mathbf{B}, \mathbf{t}, \mathbf{T}, \boldsymbol{\omega}_{\mathbf{t}}, \boldsymbol{\sigma}_{\infty})$$

It can be then demonstrated that the full covariance matrix $\boldsymbol{\Sigma}$ between *all the parameters* can be expressed as:

$$\boxed{\Sigma = \begin{pmatrix} \Sigma_{11} & \Sigma_{12} \\ \Sigma_{21} & \Sigma_{22} \end{pmatrix}}
\quad
\begin{cases}
\Sigma_{11} = M_x + \left(G_x{}^T G_x\right)^{-1} G_x{}^T G_\theta M_\theta G_\theta{}^T G_x \left(G_x{}^T G_x\right)^{-1} \\
\Sigma_{12} = -\left(G_x{}^T G_x\right)^{-1} G_x{}^T G_\theta M_\theta = \Sigma_{21} \\
\Sigma_{22} = M_\theta
\end{cases}$$

$\mathbf{M_x}$ = covariance matrix between the parameter $\overline{\mathbf{p}}$ given by the fit
$\mathbf{M_\theta}$ = covariance matrix between the parameters $\overline{\boldsymbol{\theta}}$
G_x = derivate matrix with respect to $\overline{\mathbf{p}}$
G_θ = derivate matrix with respect to $\overline{\boldsymbol{\theta}}$

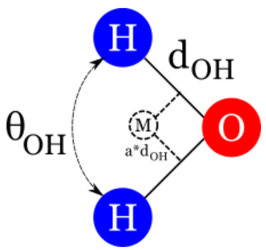
$$G_x = \begin{pmatrix} \frac{\partial z_1}{\partial \varepsilon_0} & \cdots & \frac{\partial z_1}{\partial a} \\ \vdots & \ddots & \vdots \\ \frac{\partial z_k}{\partial \varepsilon_0} & \cdots & \frac{\partial z_k}{\partial a} \end{pmatrix}
\qquad
G_\theta = \begin{pmatrix} \frac{\partial z_1}{\partial N} & \cdots & \frac{\partial z_1}{\partial \sigma_\infty} \\ \vdots & \ddots & \vdots \\ \frac{\partial z_k}{\partial N} & \cdots & \frac{\partial z_k}{\partial \sigma_\infty} \end{pmatrix}$$

For more information: G. Noguere et al. Nucl. Sci. Eng. 172, 164-179 (2012).

TSL UNCERTAINTIES QUANTIFICATION FOR CAB MODEL

Final correlation matrix and relative uncertainties for all parameters

Paramter	Value	Results after the fit									
		Relative posterior uncertainty	Posterior correlation matrix								
σ_O [nm]	0,31644	2,3%	100	-77	93	69	33	-18	-64	83	-14
ε_O [KJ/mol]	0,77491	14,6%		100	-71	-98	-85	53	97	-54	-32
q_H [e]	0,5564	3,2%			100	59	28	-2	-60	81	-18
d_{OH} [nm]	0,09419	6,3%				100	89	-63	-96	44	38
D_{OH} [KJ/mol]	432,58	6,2%					100	-63	-88	6	57
β [1/nm]	22,87	4,2%						100	51	-11	-28
θ_0 [°]	107,4	6,4%							100	-45	-49
k_θ [KJ/mol/rad²]	367,81	3,8%								100	-14
a [nm]	0,13288	2,7%									100



The correlation matrix between the model parameters and their uncertainty

Variance - Covariance matrix between the parameters Σ

Uncertainty propagation

- Microscopic data: $S(\alpha,\beta), \frac{d^2\sigma}{d\Omega dE'}; \frac{d\sigma}{d\Omega}; \sigma(E)$, …
- Integral measurements: k_{eff},…

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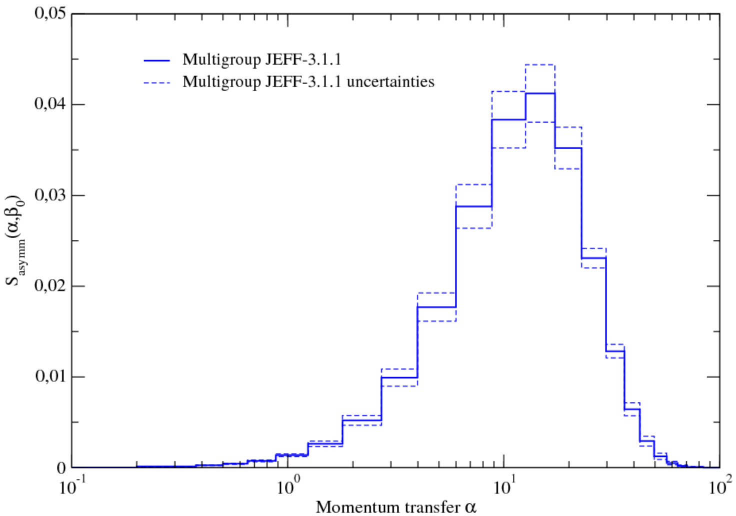
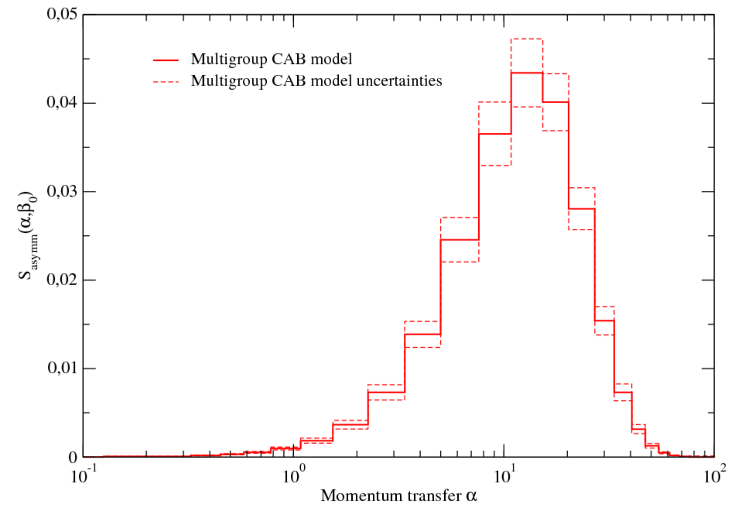
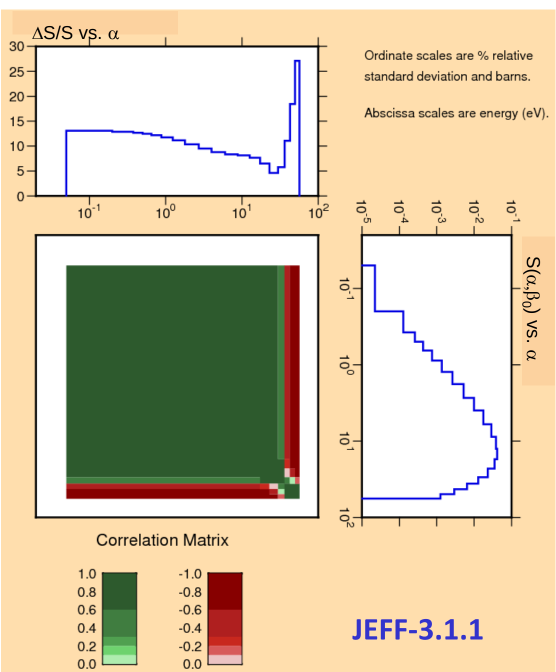
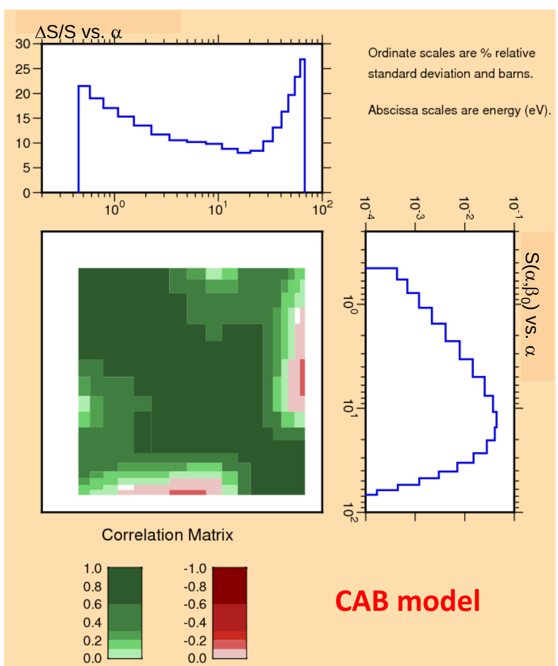
UNCERTAINTIES PROPAGATION TO $S(\alpha,\beta_0)$ SCATTERING FUNCTION (294 K)



Example for β = 10.0





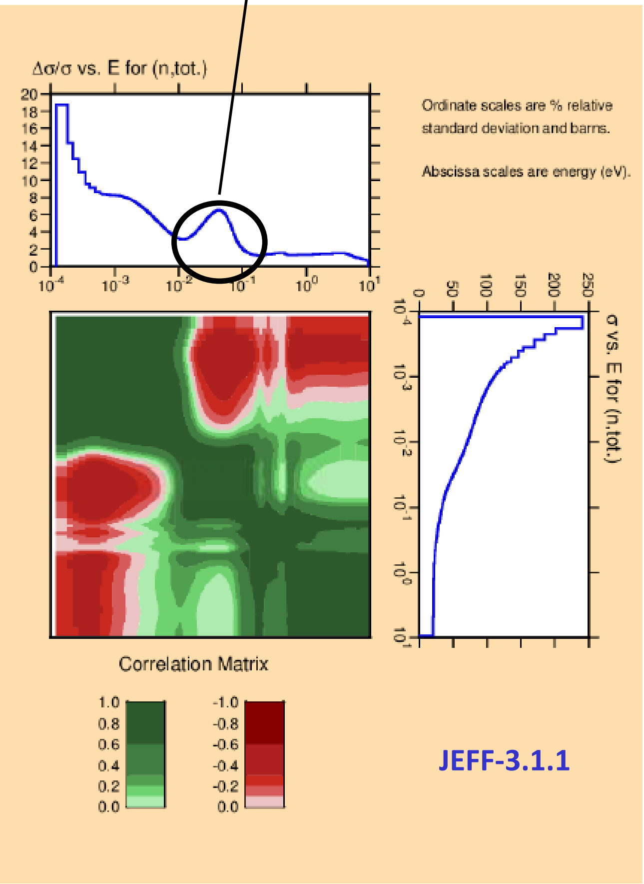


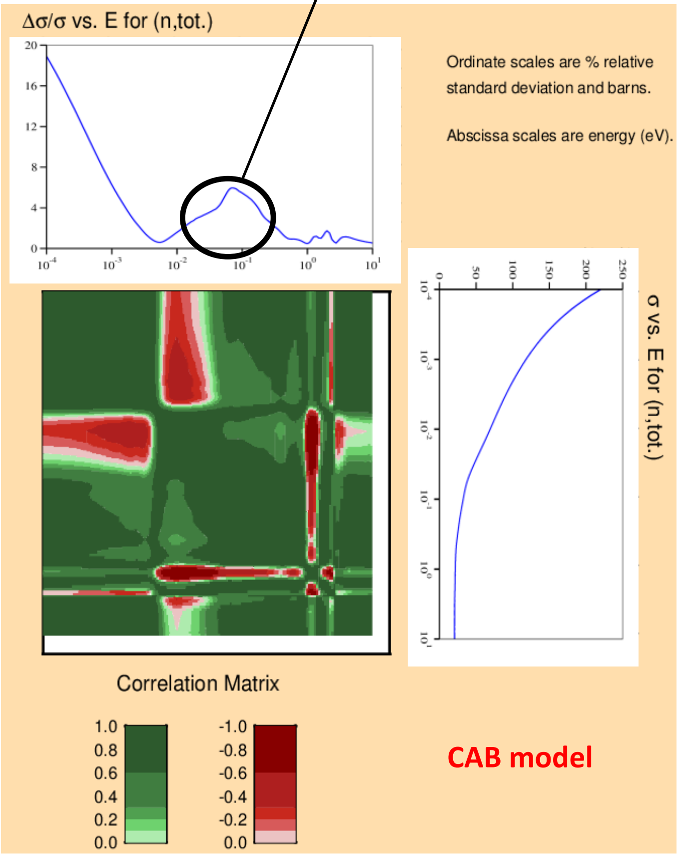
UNCERTAINTIES PROPAGATION TO ^1H IN H_2O SCATTERING CROSS SECTION (294 K)

Relative uncertainty at E_0 = 25.3 meV (thermal energy)

$\Delta\sigma/\sigma$ = 5.0%

$\Delta\sigma/\sigma$ = 3.3%





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CONCLUSIONS

1 – The thermal scattering models of ^1H in H_2O of IKE model (JEFF-3.1.1) (experimental frequency spectrum) and CAB model (simulated with molecular dynamics frequency spectrum) were investigated.

- Big differences (~400%) at low energy transfer in the $S(\alpha,\beta)$ function
- Our experimental measures of the ddxs at cold neutron energy show the limitations of JEFF-3.1.1 library. CAB model agrees with the data but there are problems in the quasi-elastic peak of the distribution.
- For the total cross section, CAB model has also good agreement with the data while JEFF-3.1.1 presents discrepancies for cold neutron energies.

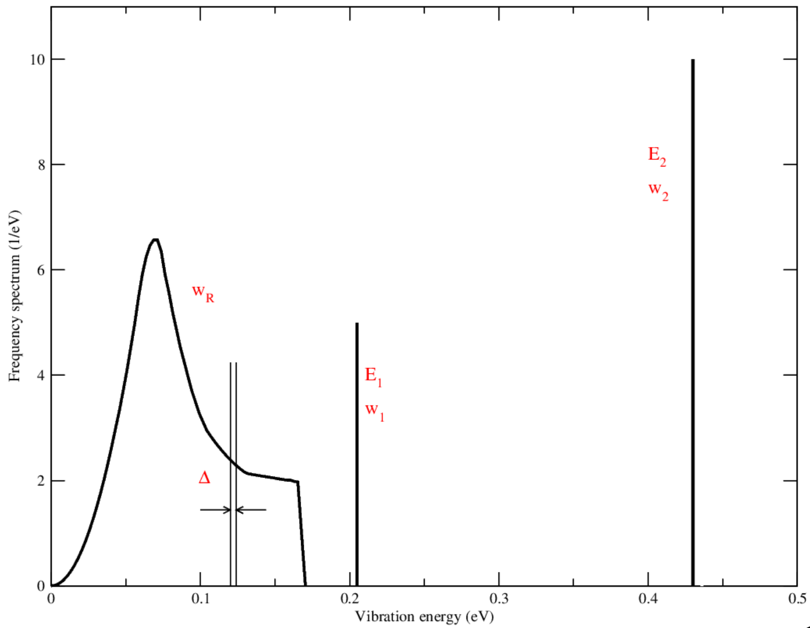
2 – Covariance matrices for both models using CONRAD code were obtained.

3 – It was done an uncertainty propagation to the $S(\alpha,\beta)$ scattering function and to the scattering ^1H in H_2O cross section.

$\Delta\sigma / \sigma$ (25.3 meV) = 5.0%
JEFF-3.1.1

$\Delta\sigma / \sigma$ (25.3 meV) = 3.3%
CAB

IKE MODEL (JEFF-3.1.1) PARAMETERS



LEAPR module parameters

- ω_t: translational mode weight
- ω_R: rotational mode weight
- ω_1: bending mode weight
- ω_2: stretching mode weight
- Δ: energy binning of continuous spectrum
- E_1: bending mode energy
- E_2: stretching mode energy

ω_t	0.021739
ω_c	0.489131
E_1	205.0
ω_1	0.163043
E_2	436.0
ω_2	0.326087

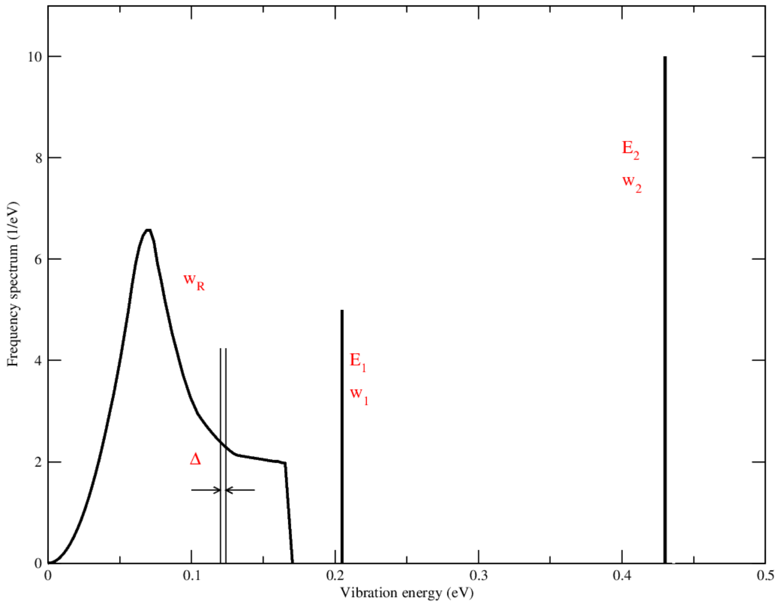
(T = 293.6 K)

The sum of the weights of each contribution to the total spectrum must be equal to 1:

$$\omega_t + \omega_R + \omega_1 + \omega_2 = 1$$

TSL UNCERTAINTIES QUANTIFICATION FOR JEFF-3.1.1

The same methodology was applied as for CAB model to generate a covariance matrix between IKE model parameters (LEAPR parameters)



ω_t: translational mode weight

ω_R: rotational mode weight

ω_1: bending mode weight

ω_2: stretching mode weight

Δ: energy binning of continuous spectrum

E_1: bending mode energy

E_2: stretching mode energy

Paramter	Value	Relative posterior uncertainty	Posterior correlation matrix					
Δ [meV]	1,0	24,1%	100	-1	2	15	-1	0
E1 [meV]	205,0	2,9%		100	0	6	0	0
E2 [meV]	436,0	8,3%			100	46	0	0
ω_T	0,0217	18,6%				100	14	0
σ_H [b]	20,436	0,2%					100	0
c	0	±1,5						100