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PRELIMINARY COVARIANCES OBTAINED WITH CONRAD/GROMACS FOR H(H20)

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Outlines

Two main objectives:

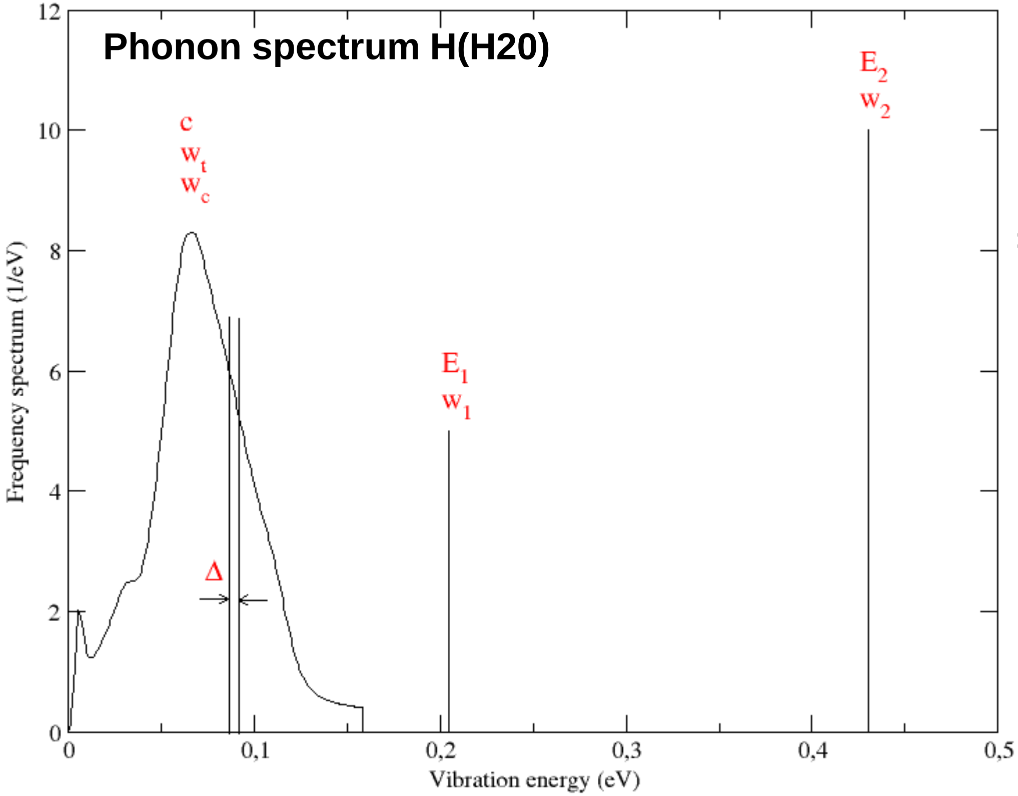
- Create covariance matrix between the **LEAPR** parameters for JEFF-3.1.1
- Create covariance matrix between the **GROMACS** parameters for the CAB model

Propagation of the TSL uncertainties to integral calculations by using:

- Direct perturbation of the model parameters
- Random $S(\alpha,\beta)$ tables (TMC calculation)
- Sensitivity calculations

Covariance matrix between the LEAPR model

The first step was to determine the covariance matrix between the LEAPR parameters



⇒ **2 main parameters were identified:**

- The energy bin Δ uses to reconstruct the phonon spectrum
- The weight w_t of the translational vibration mode

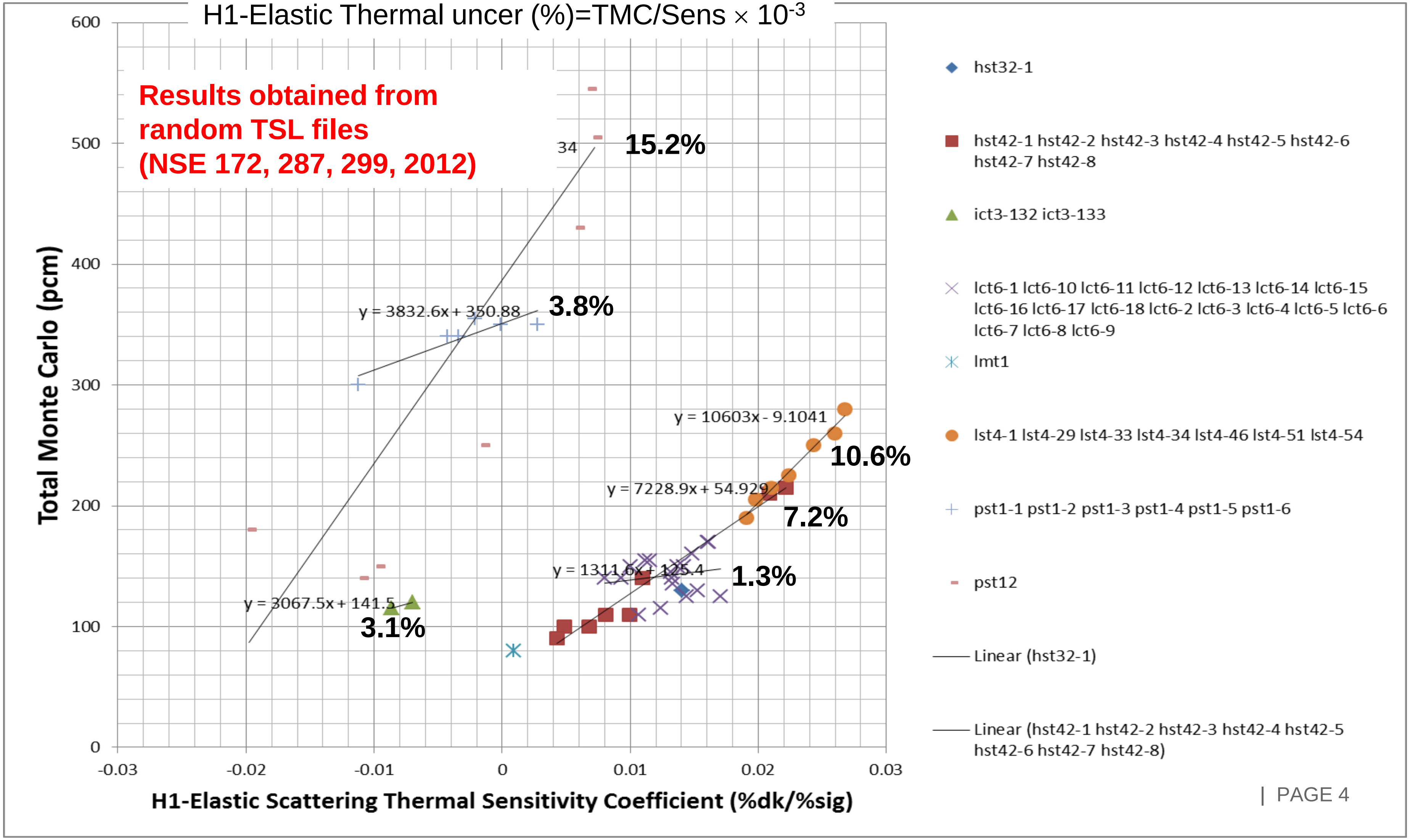
https://www.oecd-nea.org/science/wpec/sg42/

Random TSL files for TMC calculations

1000 H in H2O files based on JEFF-311 (access to these documents is restricted to SG42 members)

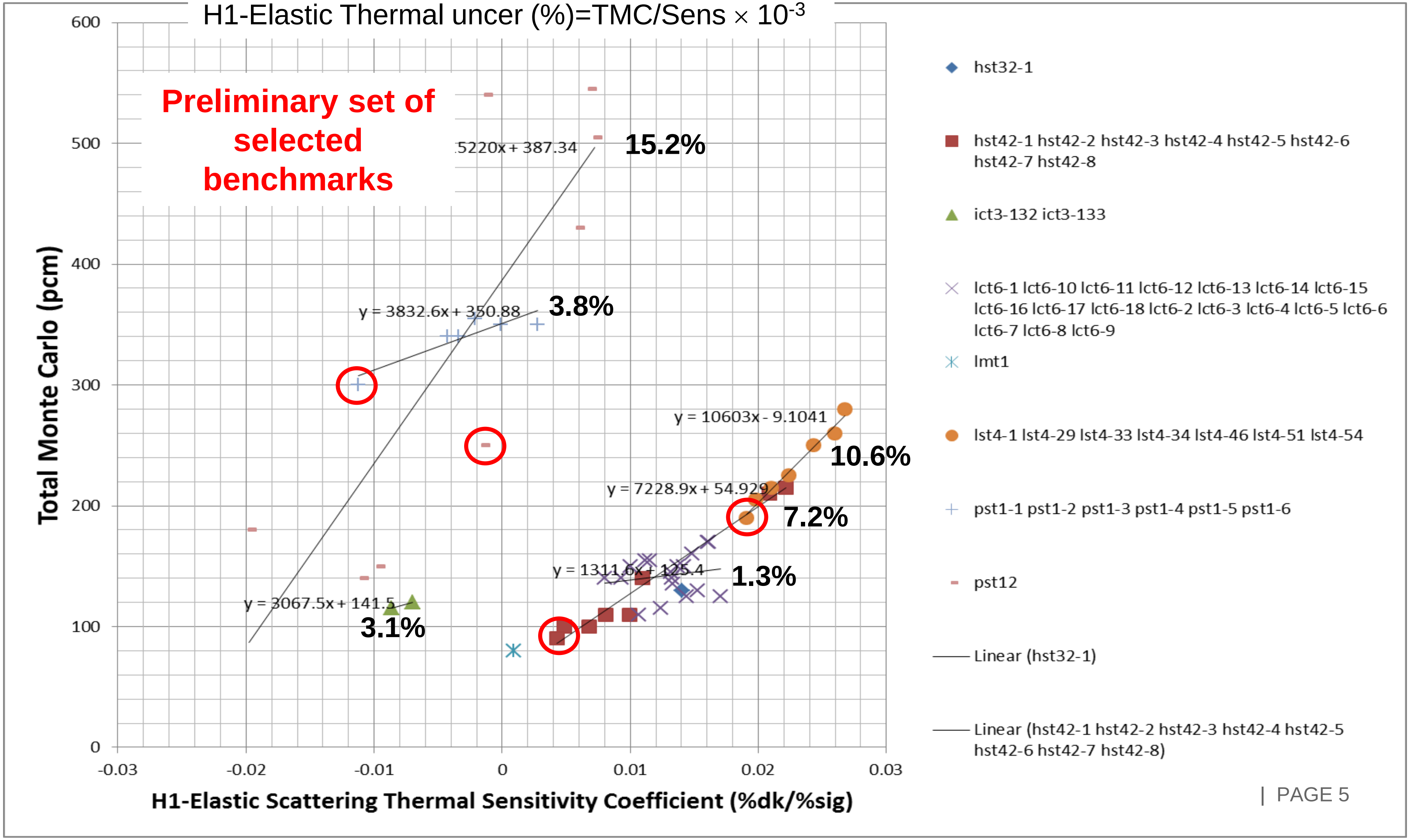
Propagation of the TSL uncertainties

TMC . vs. H1-Elastic Scattering Thermal Sensitivity Coefficient



Propagation of the TSL uncertainties

TMC . vs. H1-Elastic Scattering Thermal Sensitivity Coefficient



Propagation of the TSL uncertainties of JEFF-3.1.1

Random files can be found in the WPEC/SG42 web site
https://www.oecd-nea.org/science/wpec/sg42/

Low sensitivity to H(H2O)

Benchmarks	EALF (eV)	EAFG (eV)	TMC		Direct perturbation of the LEAPR parameters
			SERPENT	MCNP	TRIPOLI4
HEU-SOL-THERM-042-001	0.0320	0.0388		35 pcm	
LEU-SOL-THERM-004-001	0.0421	0.0735		53 pcm	79 pcm
PU-SOL-THERM-012-014	0.0783	0.1440		132 pcm	
PU-SOL-THERM-001-001	0.0885	0.1660	157 pcm	160 pcm	145 pcm
LEU-COMP-THERM-007-001	0.2870	0.5910		110 pcm	
MISTRAL-2 (MOX)	-	-			199 pcm

Cf. presentation of Juan Pablo Scotta on MISTRAL-1

Excellent agreement between the different codes and methods

Covariance matrix between the GROMACS parameters

THE JOURNAL OF CHEMICAL PHYSICS **135**, 224516 (2011)

A flexible model for water based on TIP4P/2005

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Water potential
TIP4P/2005f

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CAB model for water

CAB models for water: A new evaluation of the thermal neutron scattering laws for light and heavy water in ENDF-6 format

CrossMark

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NUCLEAR SCIENCE AND ENGINEERING: **172**, 164–179 (2012)

Zero Variance Penalty Model for the Generation of Covariance Matrices in Integral Data Assimilation Problems

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Zero Variance Penalty model
in CONRAD

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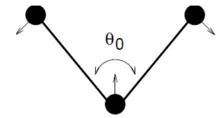
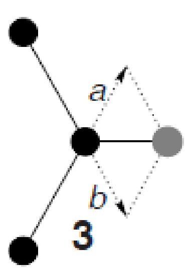
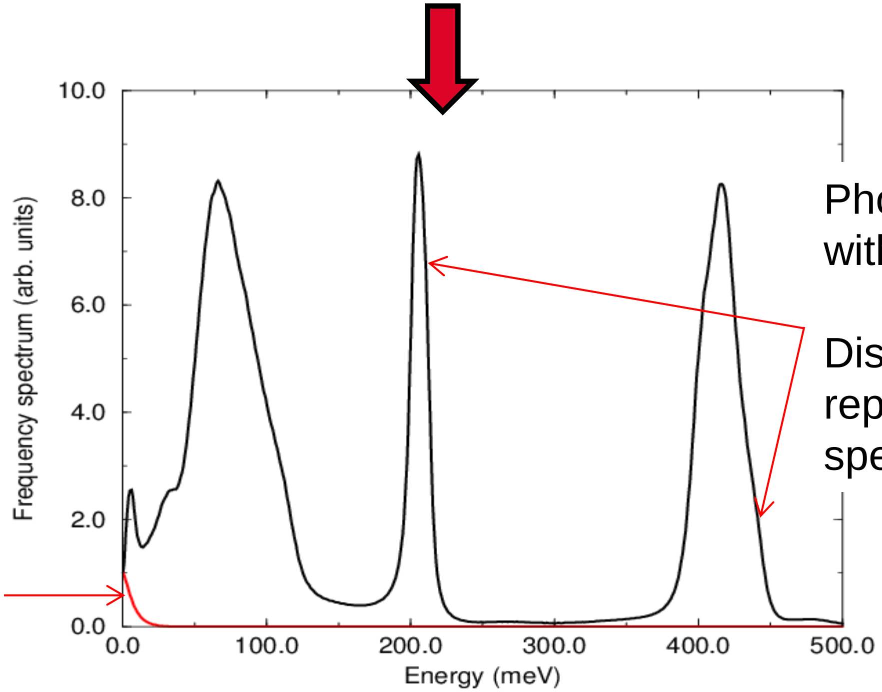
GROMACS parameters for water

Contributions	GROMACS functions	Parameters
Lennard-Jones potential	$V_{LJ}(r_{ij}) = 4\epsilon_{ij}\left(\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right)$	**sigma** **epsilon**
Electrostatic interaction	$V_c(r_{ij}) = f\frac{q_i q_j}{\varepsilon_r r_{ij}}$	**e**
Morse potential	$V_{morse}(r_{ij}) = D_{ij}[1 - \exp(-\beta_{ij}(r_{ij} - b_{ij}))]^2$,	**b** **D** **Beta**
Harmonic angle potential	$V_a(\theta_{ijk}) = \frac{1}{2}k_{ijk}^{\theta}(\theta_{ijk} - \theta_{ijk}^{0})^2$ 	**theta0** **k**
« dummy » atom		**a=b**

GROMACS parameters for water

Prior values, variances and correlations

sigma	= 3.16440E-01	+/- 3.16440E-03	(1.0%)	100									
epsilon	= 7.74907E-01	+/- 7.74907E-03	(1.0%)	0	100								
e	= 0.55640E+00	+/- 0.55640E-02	(1.0%)	0	0	100							
b	= 0.09419E+00	+/- 0.09419E-02	(1.0%)	0	0	0	100						
D	= 432.581E+00	+/- 432.581E-02	(1.0%)	0	0	0	0	100					
beta	= 22.8700E+00	+/- 22.8700E-02	(1.0%)	0	0	0	0	0	100				
theta0	= 107.400E+00	+/- 107.400E-02	(1.0%)	0	0	0	0	0	0	100			
k	= 367.810E+00	+/- 367.810E-02	(1.0%)	0	0	0	0	0	0	0	100		
a	= 0.13288E+00	+/- 0.13288E-02	(1.0%)	0	0	0	0	0	0	0	0	100	



Phonon spectrum generated with the GROMACS results

Discrete oscillators are replaced by a continuuous spectrum

Diffusion constant (translational mode) is deduced from the GROMACS results

Fitting model of CONRAD

Linear least-squares : « best » estimate of the GROMACS parameters are found iteratively by solving the following generic equations

iteration 1

$$(M_x')^{-1}=M_x^{-1}+G^tC^{-1}G$$

$$X'=X_0+M_x'G^tC^{-1}(Y-Y_{th}(X_0))$$

with $M_x = \mathrm{diag}(\mathrm{var}(x_{01}) \ldots \mathrm{var}(x_{0n}))$

and $G_{ij} = \left.\frac{\partial y_{th\,i}}{\partial x_j}\right|_{X=X_0}$ for i = 1,m and j = 1,n

iteration 2

$$(M_x'')^{-1}=M_x^{-1}+G^tC^{-1}G$$

$$X''=X'+M_x''G^tC^{-1}(Y-Y_{th}(X'))$$

with $G_{ij} = \left.\frac{\partial y_{th\,i}}{\partial x_j}\right|_{X=X'}$ for i = 1,m and j = 1,n

iteration 3 …

Fitting model of CONRAD

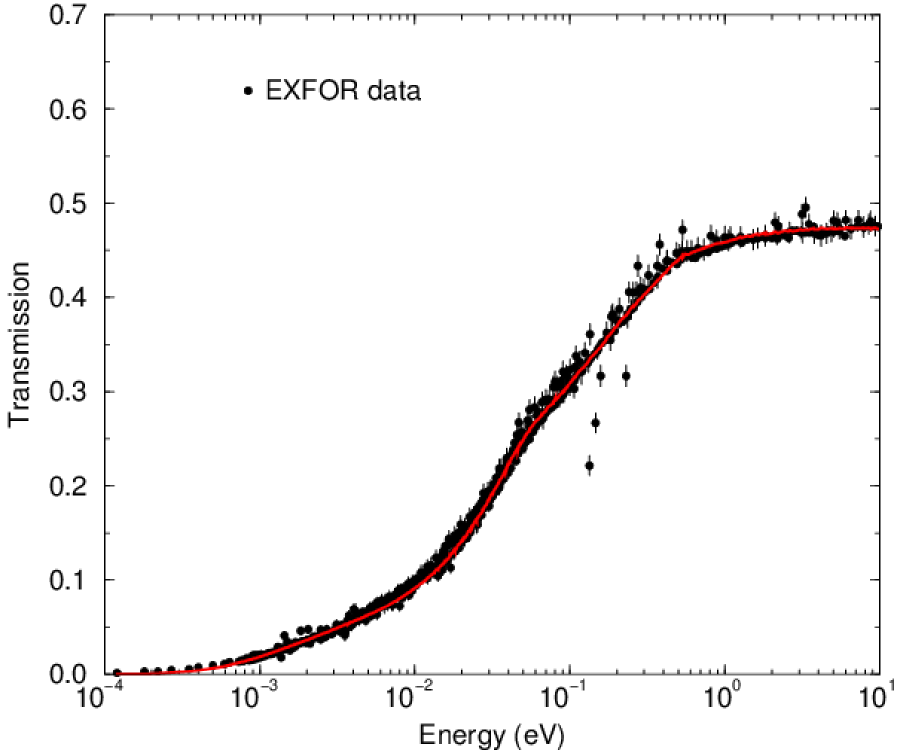
Such a fitting model was not designed to account for uncertainties of “systematic” origin.

This problem was solved in CONRAD by introducing **marginalization techniques**

These methods account for uncertainties of the “**nuisance**” and “**latent**” model parameters

Nuissance parameter definition

Example : H20 transmission is calculated as follows at a temperature ***T***



$$T_{th}(E) = NT_R(E) + B(E)$$

$$T(E) = \exp[-n\sum_i r_i \sigma_{T,i}(E)]$$

Where ***N*** is the normalisation, ***B*** a « speudo background » and ***n*** the aerial density

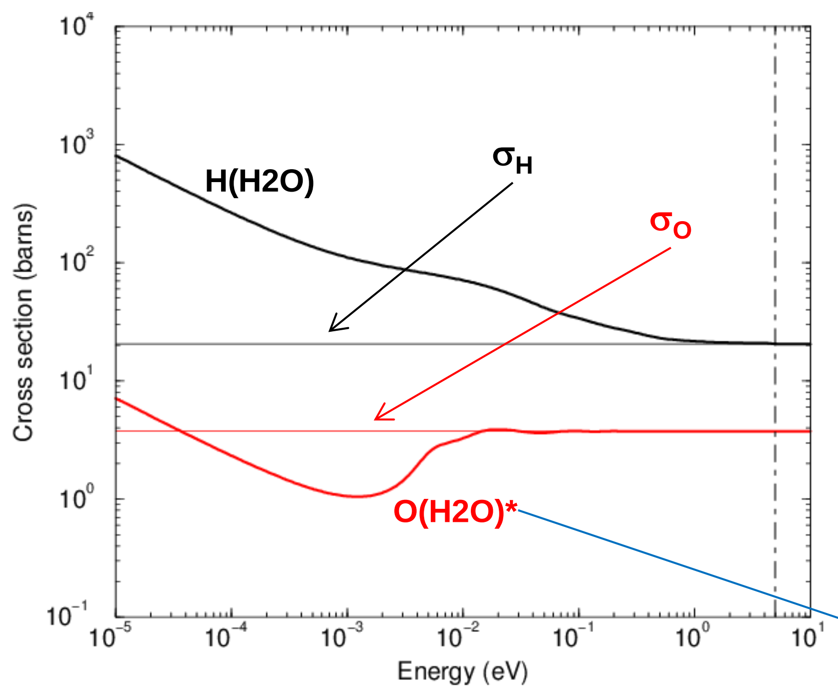
+ resolution function ***R_E(E')*** for double-deferential data

⇒ nuissance parameters of interest are *N*, *B*, *n* , *T*, *R_E(E')* …

Latent model parameter definition

Latent variables (as opposed to observable variables which can be fitted on the experimental data) may define redundant parameters or hidden variables that cannot be observed directly. This term reflects the fact that such variables are really there, but they cannot be observed or measured for practical reasons.

Example : contributions of O(H2O) and free atom cross sections of H and O



uncertainties are unknown for the moment

*O(H20) from Jose Ignacio Marquez Damian

Latent model parameter definition

Latent variables (as opposed to observable variables which can be fitted on the experimental data) may define redundant parameters or hidden variables that cannot be observed directly. This term reflects the fact that such variables are really there, but they cannot be observed or measured for practical reasons.

Example : contributions of O(H2O) and free atom cross sections of H and O

References	σ_H	σ_O
IKE model (JEFF-3.1.1)	20.478 b	3.761
Atlas of Neutron Resonances	20.491±0.014 b	3.761±0.006 b
STD2006 (IAEA)	20.436±0.041 b	
JEFFDOC-1488	20.474 ±0.012 b	3.761 ± 0.005 b

±0.2%

±0.2%

Latent model parameter definition

Latent variables (as opposed to observable variables which can be fitted on the experimental data) may define redundant parameters or hidden variables that cannot be observed directly. This term reflects the fact that such variables are really there, but they cannot be observed or measured for practical reasons.

Example : contribution of the translational weight ω_t

In the CAB model, the translational weight is deduced from the measurements performed by Novikov

Diffusion masses for light water, from measurements by Novikov et al. (1990).

T [K]	m_{diff}/m_{H_2O}
300	6.49
400	2.10
500	2.25
600	1.44

$$\omega_t = \frac{m_H}{m_{diff}} = \frac{m_H}{m_{H2O}} \left(\frac{m_{diff}}{m_{H2O}} \right)^{-1}$$

⇒ a relative uncertainty of **10%** is assumed in the calculations

Governing equations

After the fitting procedure, the covariance matrix between the parameters μ is defined as:

$$\mu = \begin{pmatrix} x \\ \theta \end{pmatrix} \quad \Sigma = \begin{pmatrix} M_x & 0 \\ 0 & M_\theta \end{pmatrix}$$

M_x Covariance Matrix between the GROMACS parameters provided by the fitting model

M_θ Covariance matrix between the auxiliary parameters (nuissance and latent parameters)

$\Rightarrow$ under this fitting model, the cross-covariance matrix between x and θ contained only **zeros**

Governing equations

Non-zero elements can be calculated by using « Variance Penalty » terms:

D.W . Muir, « The contribution of individual correlated parameters to the uncertainty of integral quantities», Nucl. Inst. Meth. A 644, 55 (2011)

The model parameters and the full covariance matrix has to be partitioned as:

$$\mu = \begin{pmatrix} x \\ \theta \end{pmatrix} \qquad \Sigma = \begin{pmatrix} \Sigma_{11} & \Sigma_{12} \\ \Sigma_{21} & \Sigma_{22} \end{pmatrix}$$

By definition, the observable parameters *x* are the « **passive** » parameters and the latent parameters θ are the « **active** » parameters

An estimation of Σ can be obtained by adding two covariance matrices

$$\Sigma = M_x + H_\theta$$

The « statistical » contribution M_x can be estimated via a fitting model and the « systematic» contribution H_θ can be obtained analytically by introducing **variance penalty terms *P(z,θ)***

Governing equations

$P(z,\theta)$ is a measure of the contribution of the uncertainty of the latent and nuisance variables to the variance of a given calculated quantity z. By definition

$$P(z,\theta) = G H_\theta G^T$$

The covariance matrix H_θ has the following form

$$H_\theta = \begin{pmatrix} M_{x,\theta} M_\theta^{-1} M_{x,\theta}^T & M_{x,\theta}^T \\ M_{x,\theta} & M_\theta \end{pmatrix}$$

For a vector quantity z of general dimension k, the derivative matrix $G=(G_x,G_\theta)$ of the quantity z to the parameters x and θ is defined as:

$$G_x = \begin{pmatrix} \frac{\partial z_1}{\partial x_1} & \cdots & \frac{\partial z_1}{\partial x_n} \\ \vdots & \ddots & \vdots \\ \frac{\partial z_k}{\partial x_1} & \cdots & \frac{\partial z_k}{\partial x_n} \end{pmatrix} \qquad G_\theta = \begin{pmatrix} \frac{\partial z_1}{\partial \theta_1} & \cdots & \frac{\partial z_1}{\partial \theta_m} \\ \vdots & \ddots & \vdots \\ \frac{\partial z_k}{\partial \theta_1} & \cdots & \frac{\partial z_k}{\partial \theta_m} \end{pmatrix}$$

Governing equations

By using the partitioned form of G, we have

$$P(z,\theta) = G_x M_{x,\theta} M_\theta^{-1} M_{x,\theta}^T G_x^T + G_x M_{x,\theta} G_\theta^T + \left(G_x M_{x,\theta} G_\theta^T\right)^T + G_\theta M_\theta G_\theta^T = 0$$

The « zero variance penalty » condition lead to

$$M_{x,\theta} = -(G_x^T G_x)^{-1} G_x^T G_\theta M_\theta$$

According to the definition of Σ, we have

$$\Sigma = \begin{pmatrix} \Sigma_{11} & \Sigma_{12} \\ \Sigma_{21} & \Sigma_{22} \end{pmatrix}$$

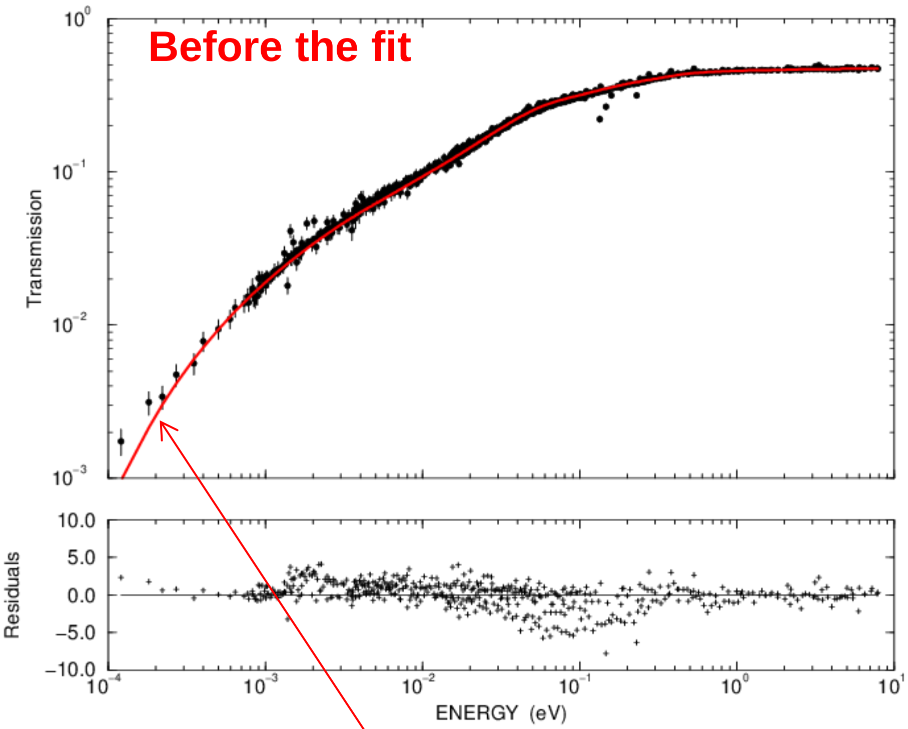
$$\begin{cases} \Sigma_{11} = M_x + \left(G_x^T G_x\right)^{-1} G_x^T G_\theta M_\theta G_\theta{}^T G_x \left(G_x^T G_x\right)^{-1} \\ \Sigma_{12} = -(G_x^T G_x)^{-1} G_x^T G_\theta M_\theta \\ \Sigma_{22} = M_\theta \end{cases}$$

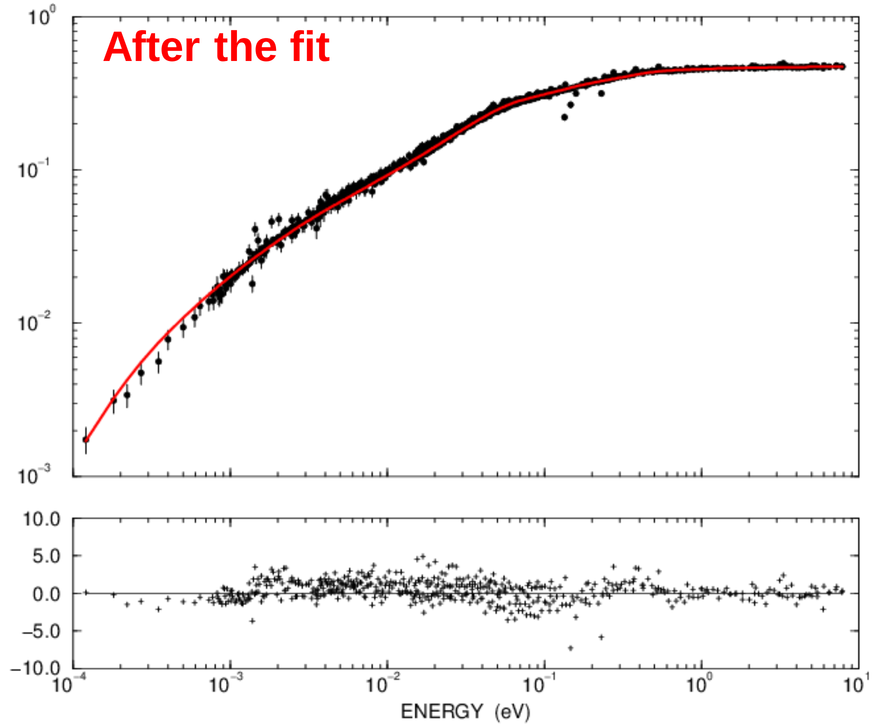
Covariance matrix between the parameters:

- GROMACS parameters
- Experimental parameters
- LEAPR parameters

Results for H(H20)

Two-step CONRAD calculation for producing Model Parameter Covariance Matrix for GROMACS





prior model parameters from **CAB MODEL**

Prior and posterior model parameters are nearly equivalent

Results for H(H20)

Two-step CONRAD calculation for producing Model Parameter Covariance Matrix for GROMACS

After the fit

													M_x
sigma	= 0.3239E+00	+/- 0.1509E-02	(0.5%)	100									
epsilon	= 0.7683E+00	+/- 0.6790E-02	(0.9%)	-4	100								
e	= 0.5647E+00	+/- 0.3998E-02	(0.7%)	49	-5	100							
b	= 0.9606E-01	+/- 0.6061E-03	(0.6%)	35	-5	-47	100						
D	= 0.4256E+03	+/- 0.3807E+01	(0.9%)	-18	-24	-4	-7	100					
beta	= 0.2401E+02	+/- 0.1099E+00	(0.5%)	-16	23	-7	-9	-21	100				
theta0	= 0.1053E+03	+/- 0.9130E+00	(0.9%)	-1	-22	-7	-38	-19	11	100			
k	= 0.3713E+03	+/- 0.3202E+01	(0.9%)	-1	7	-2	-23	4	-39	-14	100		
a	= 0.1320E+00	+/- 0.1266E-02	(1.0%)	-13	-10	-1	-5	-10	-8	-16	-9	100	

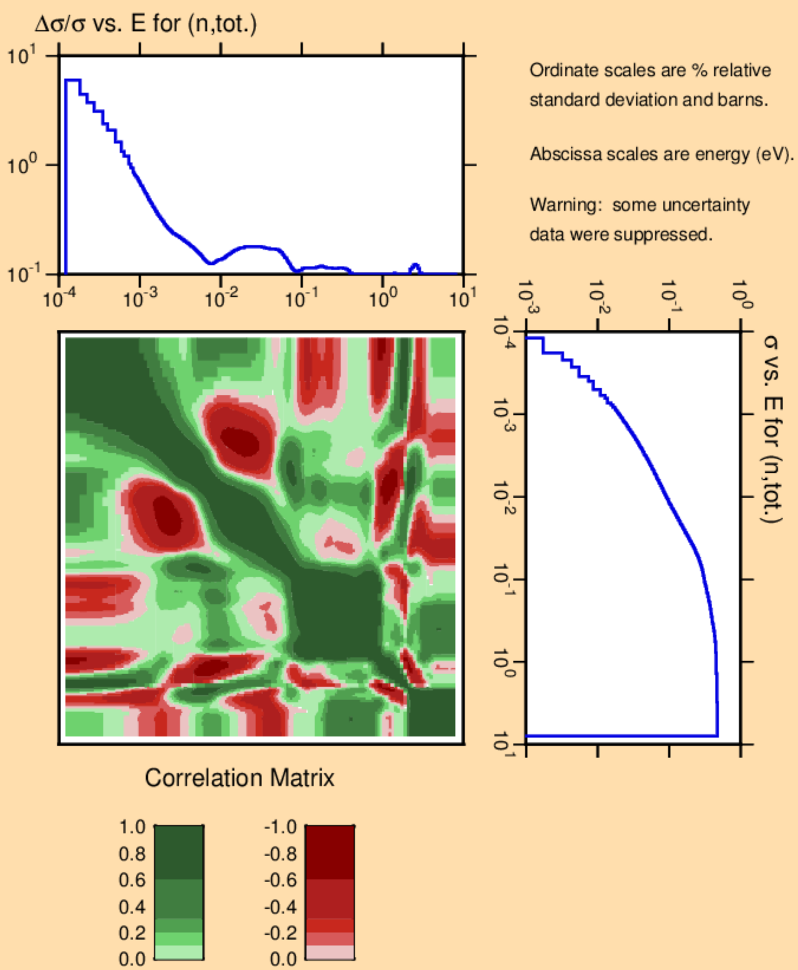
After marginalization

													Σ_{11}
sigma	= 0.3239E+00	+/- 0.7864E-02	(2.4%)	100									
epsilon	= 0.7683E+00	+/- 0.7445E-01	(9.7%)	-13	100								
e	= 0.5647E+00	+/- 0.1222E-01	(2.2%)	57	-81	100							
b	= 0.9606E-01	+/- 0.6473E-02	(6.7%)	98	-19	58	100						
D	= 0.4256E+03	+/- 0.3064E+02	(7.2%)	88	-41	74	93	100					
beta	= 0.2401E+02	+/- 0.2476E+01	(10.3%)	-68	43	70	-74	-91	100				
theta0	= 0.1053E+03	+/- 0.1865E+02	(17.7%)	-93	31	-69	-97	-98	87	100			
k	= 0.3713E+03	+/- 0.1157E+02	(3.1%)	79	33	66	82	92	-90	-91	100		
a	= 0.1320E+00	+/- 0.3529E-01	(26.7%)	71	64	85	77	93	-95	-89	87	100	

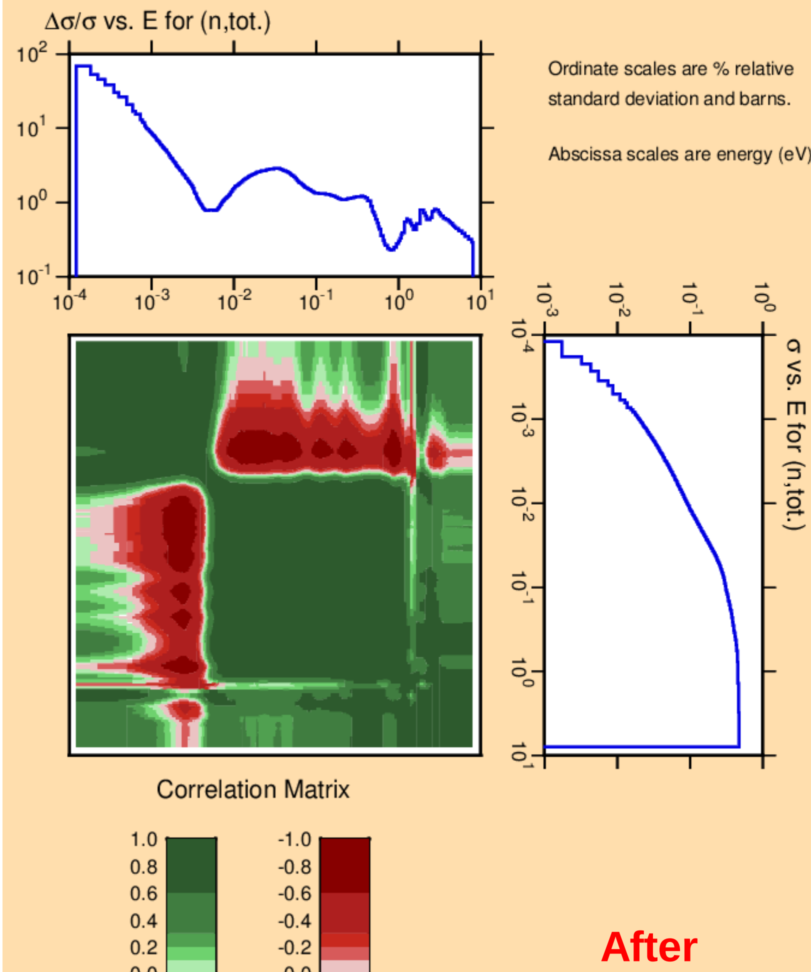
$$\Sigma_{11} = M_x + \left(G_x^T G_x\right)^{-1} G_x^T G_\theta M_\theta G_\theta^{\,T} G_x \left(G_x^T G_x\right)^{-1}$$

Results for H(H20)

Relative uncertainties and correlation matrix for H2O (transmission)

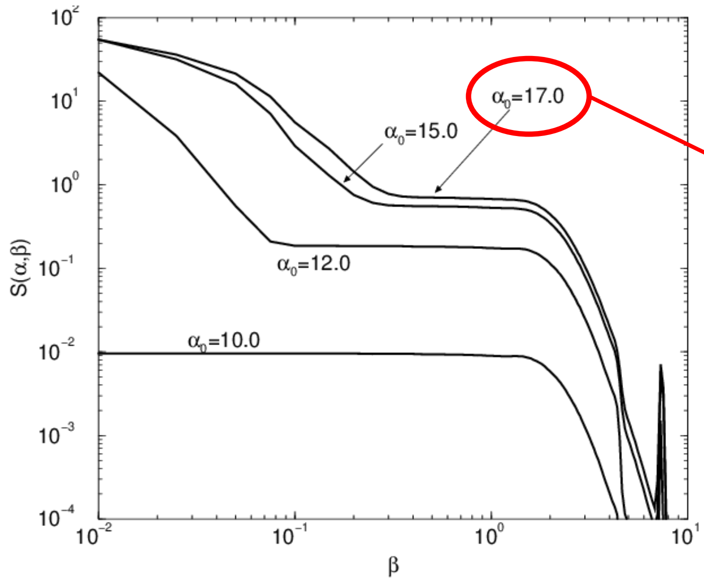


After the fit



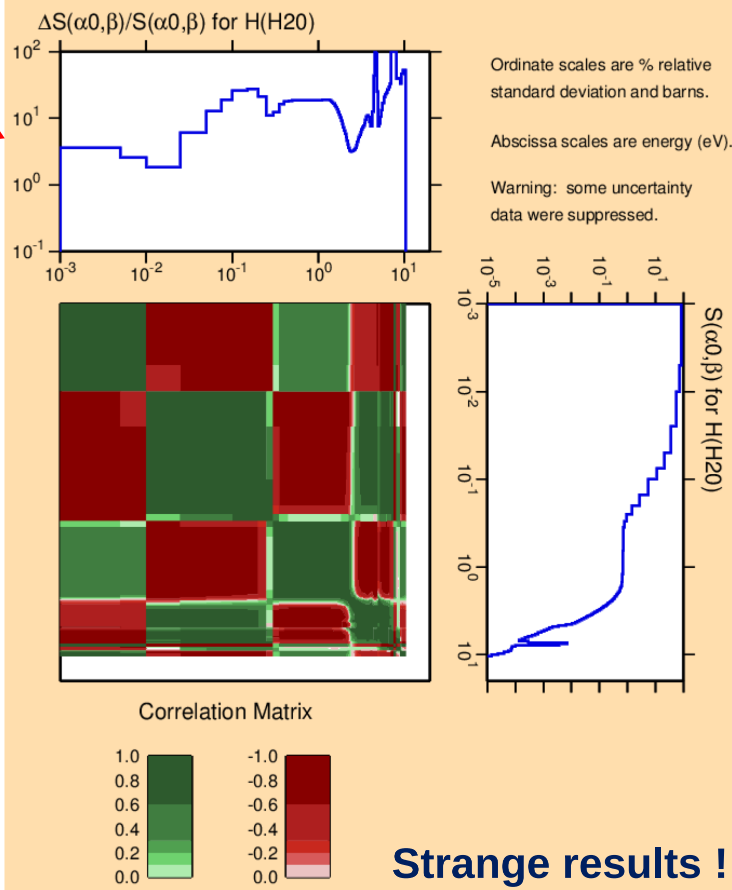
After marginalization

Relative uncertainties and correlation matrix on S(α,β)



Calculated with the Model Parameter Covariance Matrix obtained after marginalization

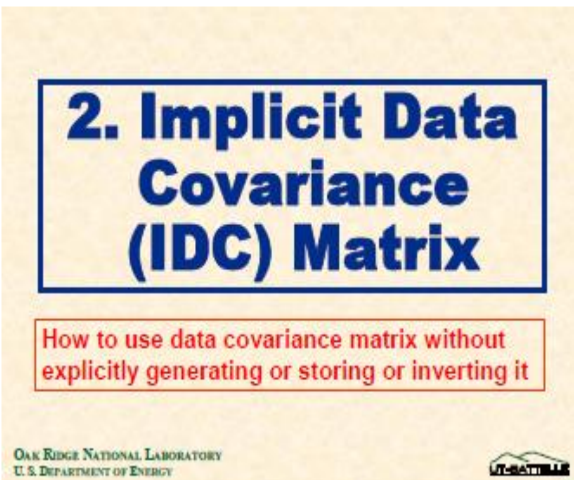
How to store large covariance matrix for S(α,β) ?



Strange results !
To be confirmed

IDC : a solution to store large Resonance-Parameter Covariance Matrix

« A concise method for storing and communicating the Experimental Covariance Matrix », N.M. Larson, ORNL/TM-2008/104, 2008



- Available in SAMMY and CONRAD to read AGS covariance file (used at the IRMM)
- Cf. WPEC/SG-36 «Evaluation of experimental data in the resolved resonance region»

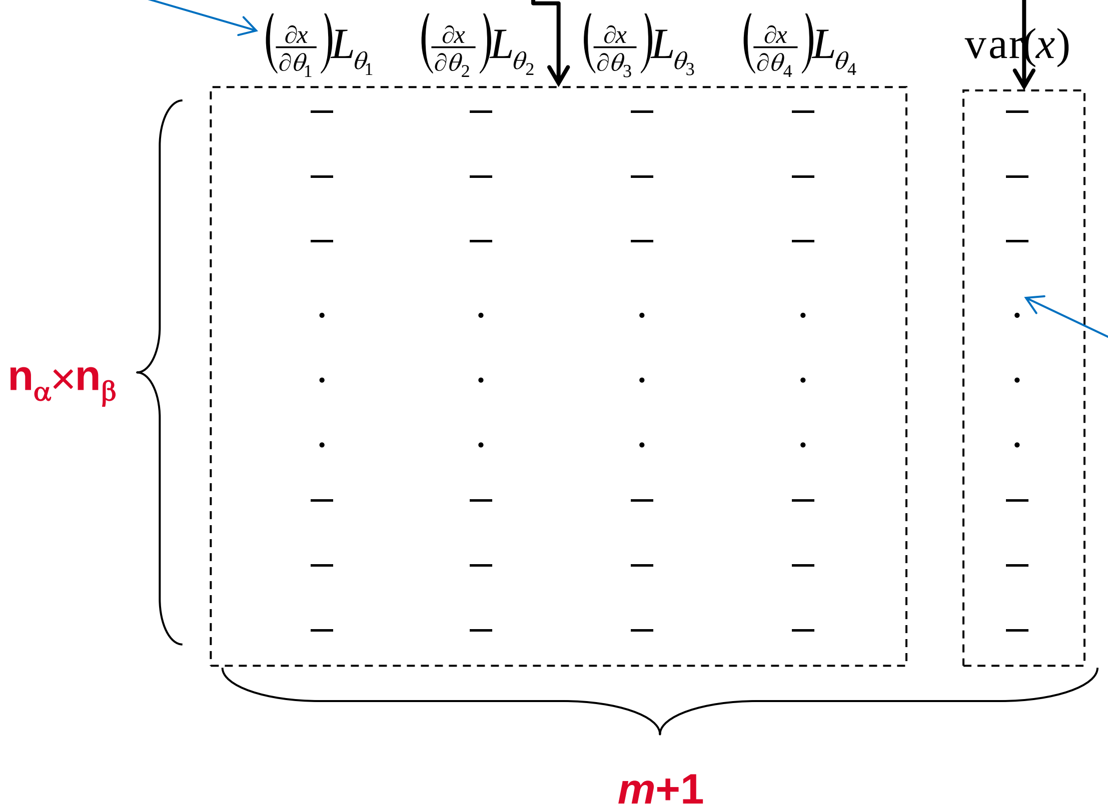
⇒ IDC format can be also used to store covariance matrix for S(α,β)

IDC format to store large covariance matrix

Derivatives of S(α,β) to the nuissance parameters introduced in the marginalization procedure

$$D = S_\theta S_\theta^T + M$$

diagonal matrix



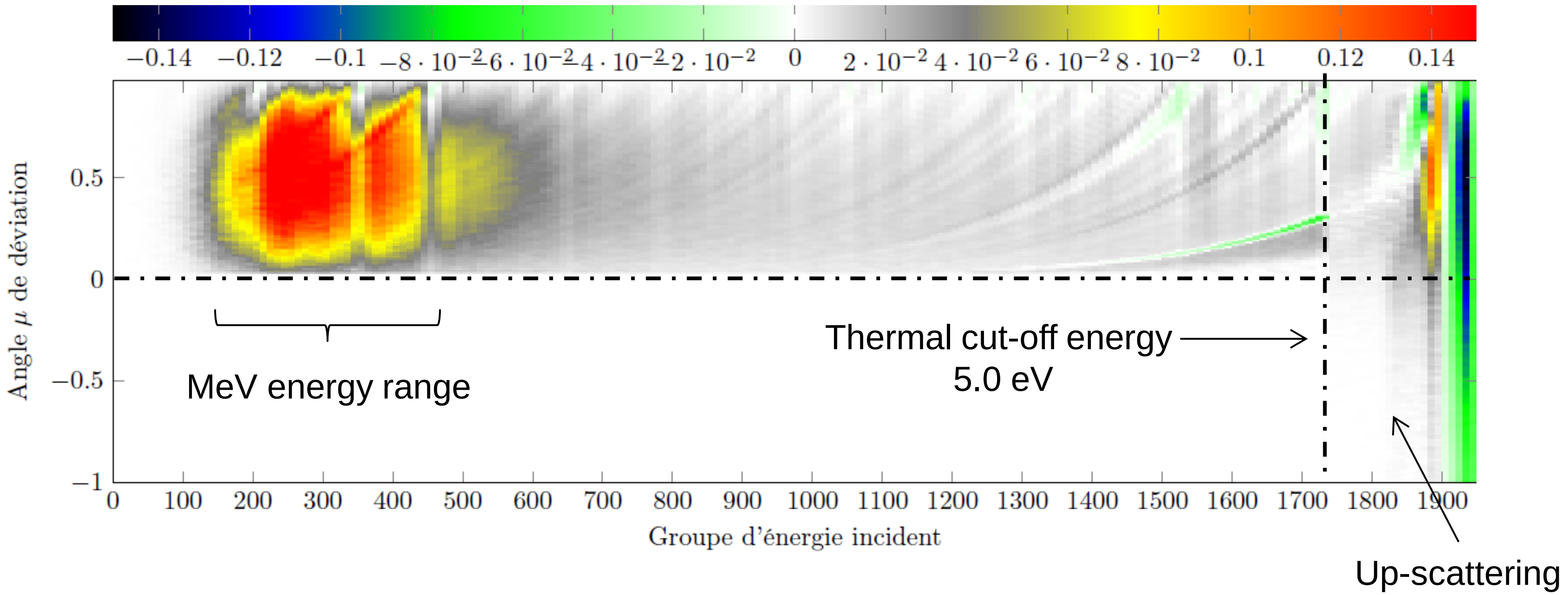
Variance on S(α,β) from the fitting procedure
Correlations from the fit are neglected

For a given integral quantity z, the variance is calculated as follow:

$$Var(z)=SDS^T$$

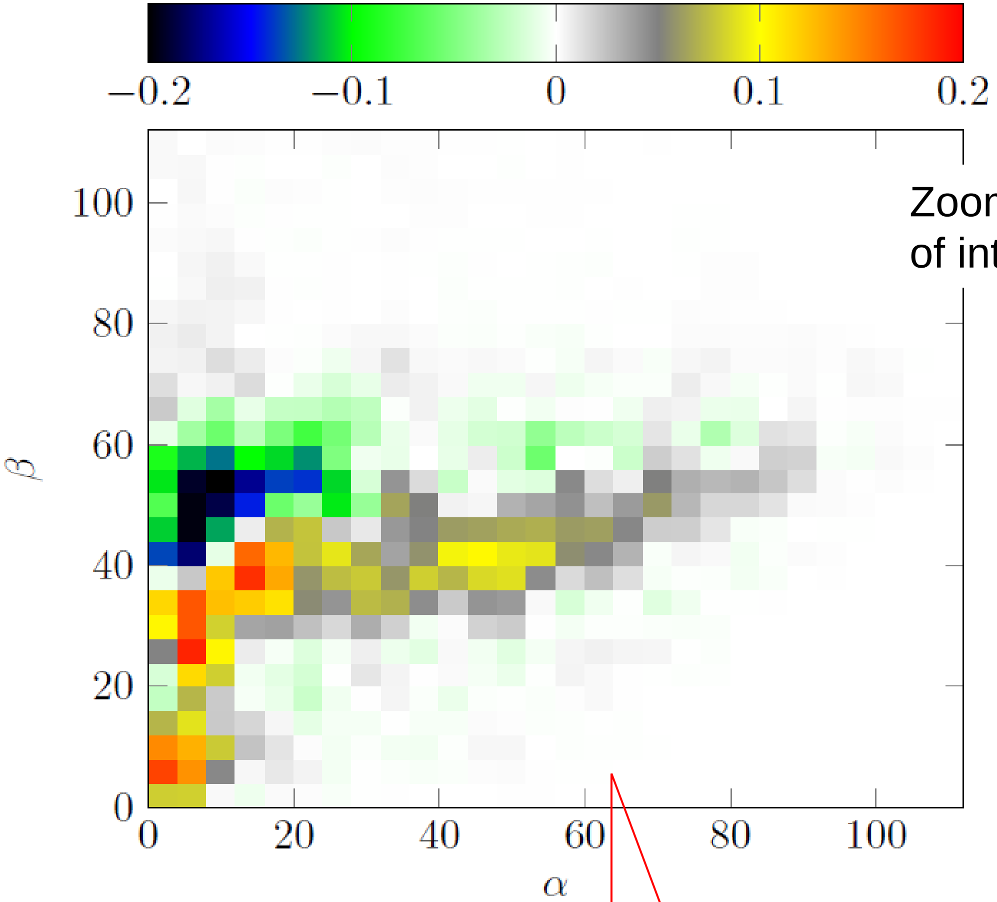
Where *D* is the covariance matrix for S(α,β) and *S* contains the derivatives of *z* to S(α,β)

Exemple: Sensitivity to the elastic scatering of H(H20) in **PST001.1** as a function of the incident energy and μ obtained with TRIPOLI4-IFP (PhD thesis G.Truchet, CEA Cadarache)



Propagation of TSL uncertainties to integral calculations

Exemple: Sensitivity to S(α,β) of H(H20) in **PST001.1** obtained with TRIPOLI4-IFP (PhD thesis G.Truchet, CEA Cadarache)



Zoom on the (α,β) region of interest for this work

⇒ Final calculations Var(z)=SDS^T not yet performed !

Conclusions

Present results on TSL uncertainties

☑ **CONRAD** can be use to produce Model Parameter Covariance Matrix for LEAPR

☑ **CONRAD** can be use to produce Model Parameter Covariance Matrix for GROMACS

☑ Production of large covariance matrix for S(α,β) is possible with the **IDC formalism**

☑ **TRIPOLI4-IFP** can be used to propagate TSL uncertainties to integral data

Next steps

☐ Include double-differential and mu-bar data in the fitting procedure

☐ Generate random files for the CAB model

☐ Finalize the propagation of the uncertainties with TRIPOLI4-IFP

Acknowledgment

Special thanks to Jose Ignacio Marquez Damian for his help on using GROMACS and for the fruitful discussions and advices.