DE LA RECHERCHE À L'INDUSTRIE

TSL ACTIVITIES IN THE FRAME OF THE NAUSICAA COLLABORATION

G. Noguere, J.P. Scotta, C. De Saint Jean, A. Filhol, J. Ollivier, E. Farhi,
Y. Calzavarra, V. Jaiswal, L. Leal, J.I. Marquez Damian, P. Maldonado

16 May 2018, NEA, Paris

Context

New TSL evaluations of H2O and UO2 (ENDF\B-VIII)

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 0.000000+0 0.000000+0         0         0        54         2
 H(H2O)     ARCAB     EVAL-JUN17 Marquez Damian
                      DIST-
----ENDF/B-VIII.0     MATERIAL 1
-----THERMAL NEUTRON SCATTERING DATA
------ENDF-6 FORMAT

*******************************************************************
*
* Temperatures = 283.6, 293.6, 300.0,
*                323.6, 350.0, 373.6, 400.0,
*                423.6, 450.0, 473.6, 500.0,
*                523.6, 550.0, 573.6, 600.0 K, 623.6 K
*
* Extrapolated temperatures = 650.0, 800.0 K
*
* This evaluation is based on the CAB Model for light water[1]
* in a liquid state, T < T-crit(H2O) = 647.1 K. Two extrapolated
* temperature points at 650 K and 800 K were added for backwards
* compatibility with ENDF/B-VII.1 scripts. The file (MF7) was
* generated using NJOY 99.396[1] with a patch (upcab).
*
* The CAB model is a further improvement of ENDF/B-VII (2006)
* and IKE, Stuttgart (2005) models for light water (incoherent
* inelastic approximation for n + H-in-H2O & vibrational
* spectrum decomposition). The continuous spectrum[2] is based
* on MD GROMACS Calculations[2] and diffusion coefficients
* measured by Yoshida[3] and Mills[4].
*
* For oxygen in H2O, free gas approximation is acceptable.
*
* The evaluation was prepared by:
*
* J.I. Marquez Damian, F. Cantargi, and J.R. Granada
* Nuclear Data Group - Neutron Physics Department
* Centro Atomico Bariloche - Argentina (CAB):
*
*  and
*
* D. Roubtsov
* Canadian Nuclear Laboratories (CNL)
* Chalk River, Canada
*
* References:
*  [1] Ann. Nucl. Energy, 65, 280 (2014)
*      http://dx.doi.org/10.1016/j.anucene.2013.11.014
*  [2] J. Chem. Phys. 139, 024504 (2013)
*      http://dx.doi.org/10.1063/1.4812828
*  [3] J. Chem. Phys. 123, 164506 (2005)
*      http://dx.doi.org/10.1063/1.2056542
*  [4] J. Phys. Chem. 77, 685 (1973)
*      http://dx.doi.org/10.1021/j100624a025
*
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 U(UO2)    LEIP LAB   EVAL-OCT16 J.L. Wormald, Y. Zhu, A.I. Hawari  48 1451     5
                      DIST-                                          48 1451     6
----ENDF/B-VIII.beta3 MATERIAL   48                                  48 1451     7
-----THERMAL NEUTRON SCATTERING DATA                                 48 1451     8
------ENDF-6 FORMAT                                                  48 1451     9
 Temperatures = 296 400 500 600 700 800 1000 1200 K                  48 1451    10
                                                                     48 1451    11
 Background                                                          48 1451    12
 ----------                                                          48 1451    13
 This library was produced by the Low Energy Interaction Physics     48 1451    14
 (LEIP) group at NC State University. The inelastic scattering       48 1451    15
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 from the NJOY code system was used to produce File 7 MT=2 and 4     48 1451    18
 for U in UO2 and O in UO2. Modifications to the LEAPR module were   48 1451    19
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 Per convention, MAT=48 and ZA=148 are used for U in UO2.            48 1451    21
                                                                     48 1451    22
 References                                                          48 1451    23
 ----------                                                          48 1451    24
 1. A.I. Hawari, et al "Ab Initio Generation of Thermal Neutron      48 1451    25
    Scattering Cross Sections," Proceedings of PHYSOR 2004: The      48 1451    26
    Physics of Fuel Cycles and Advanced Nuclear Systems ---          48 1451    27
    Global Developments (2004).                                      48 1451    28
 2. J.L. Wormald, A.I. Hawari "Ab Initio Generation of Thermal       48 1451    29
    Scattering Law for Uranium Dioxide," Transactions of the         48 1451    30
    American Nuclear Society, v 115 (2016)                           48 1451    31
 3. Y. Zhu, A.I. Hawari, "Implementation of a Generalized Coherent   48 1451    32
    Elastic Scattering Formulation for Thermal Neutron Scattering    48 1451    33
    Analysis," Proceedings of ICNC 2015: 35th International          48 1451    34
    Conference on Nuclear Criticality Safety (2015)                  48 1451    35
                                1        451         38          0  48 1451    36
                                7          2       1305          0  48 1451    37
                                7          4      46810          0  48 1451    38
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Experimental validation H2O and UO2

Microscopic and integral data base

Double-differential neutron cross sections and DOS : new experimental program

⇒ Light and heavy water : IN4, IN5 and IN6 at ILL from cold to hot operating conditions

- H20: 285 K to 540 K (P = 1- 600 bar)

⇒ UO2 : IN4 and IN6 from room temperature to Hot Full Power conditions

- T = 294 K to 900 K

Total cross sections: from the EXFOR data base

⇒ Analysis of transmission measurements performed on UO2 samples at the time-of-flight facility GELINA of JRC-Geel

- T = 23 K and 294 K

Integral validation with CEA benchmarks

⇒ Analysis of the MISTRAL programs carried out in the EOLE reactor of CEA Cadarache

- T = 280 K to 354 K

CAB model for light and heavy water

Annals of Nuclear Energy 65 (2014) 280–289

Contents lists available at ScienceDirect

Annals of Nuclear Energy

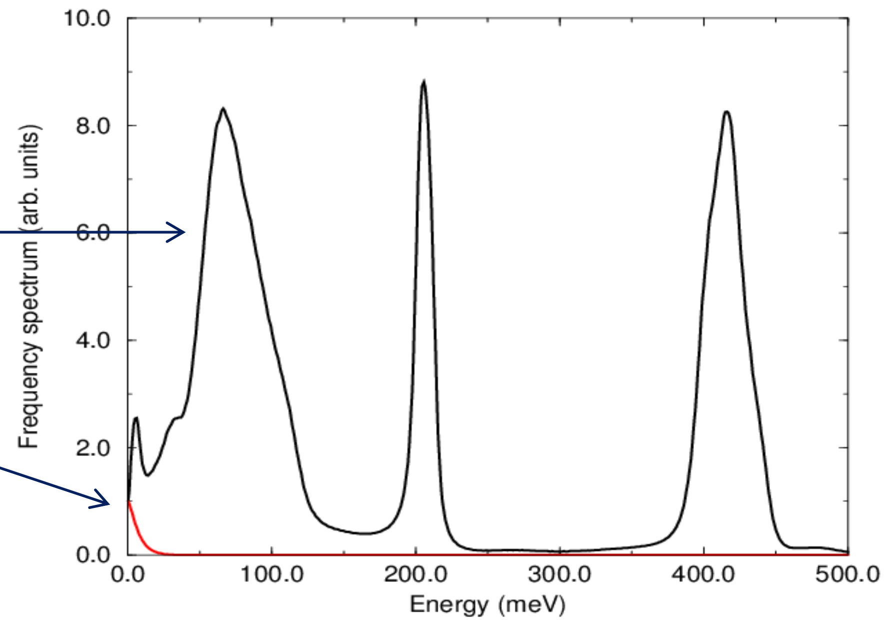
journal homepage: www.elsevier.com/locate/anucene

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

J.I. Márquez Damián [a,*], J.R. Granada [a], D.C. Malaspina [b]

[a] Neutron Physics Department and Instituto Balseiro, Centro Atómico Bariloche, CNEA, Argentina
[b] Department of Biomedical Engineering and Chemistry of Life Processes Institute, Northwestern University, 2145 Sheridan Road, Evanston, IL 60208, United States

- Density of states calculated with GROMACS
- Difusion model from Egelstaf's expression available in LEAPR



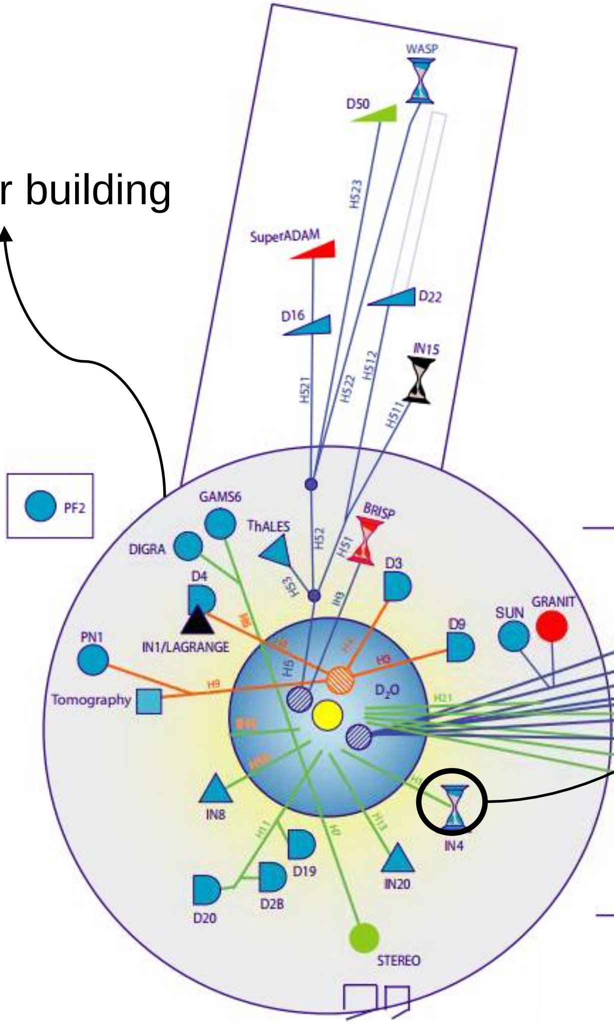
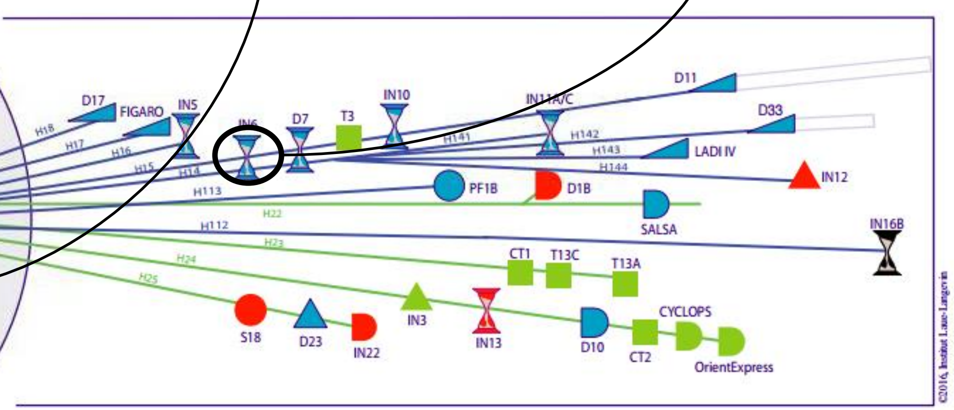
Double-differential neutron cross section H2O

INSTITUTE LAUE-LANGEVIN

Reactor building

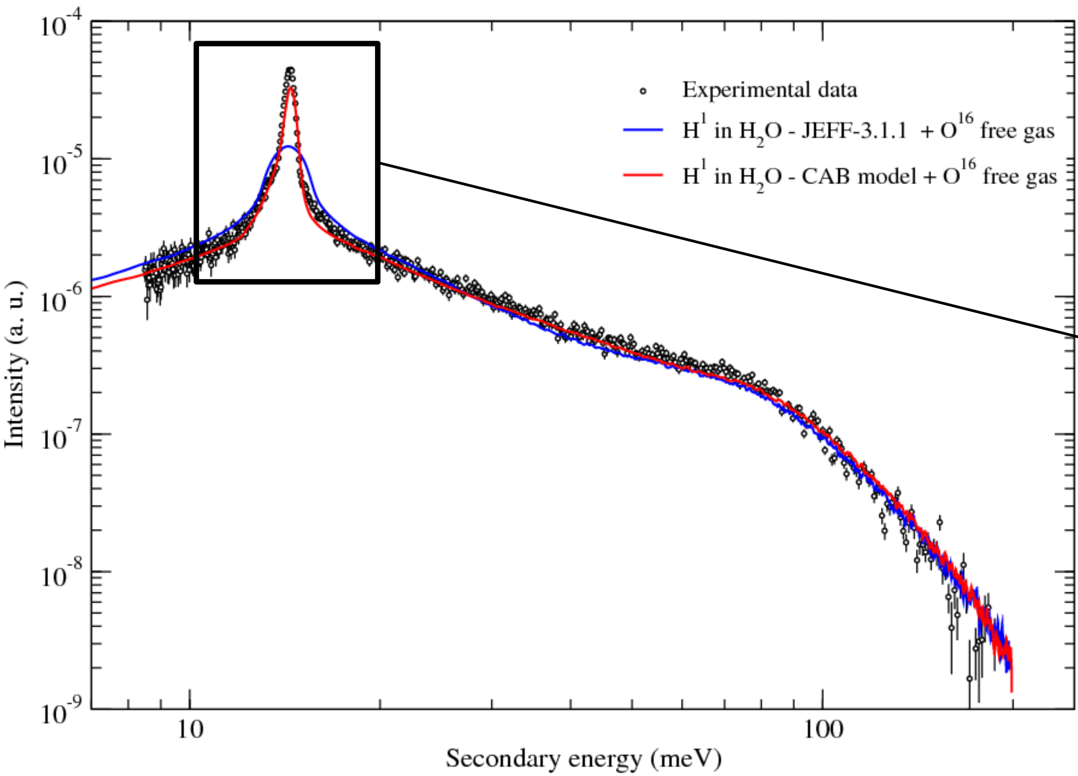
IN4 ToF spectrometer

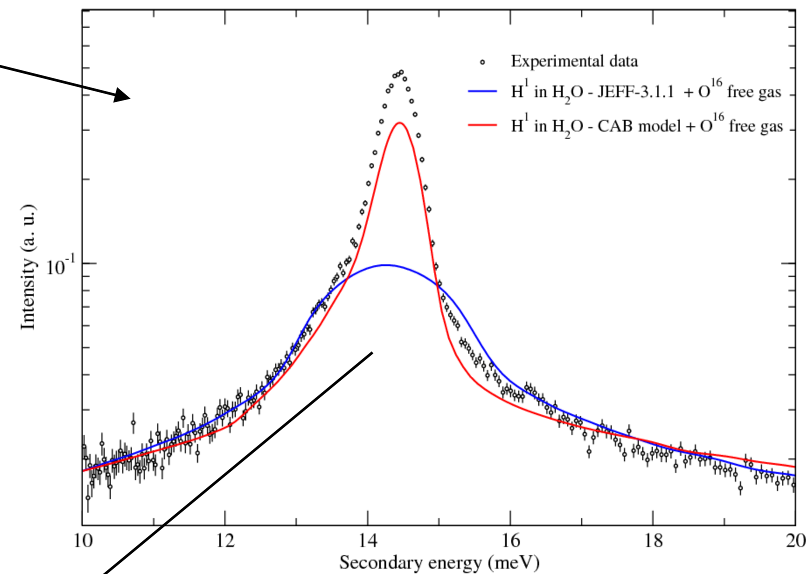
IN6 ToF spectrometer

Double-differential neutron cross section H2O

Incident neutron energy E_0 = 14 meV , θ = 15° and T= 300 K.





Problems in JEFF-3.1.1 $\Rightarrow$ free gas law to account the molecular diffusion, while CAB model uses the Egelstaff expression

Double-differential neutron cross section H2O

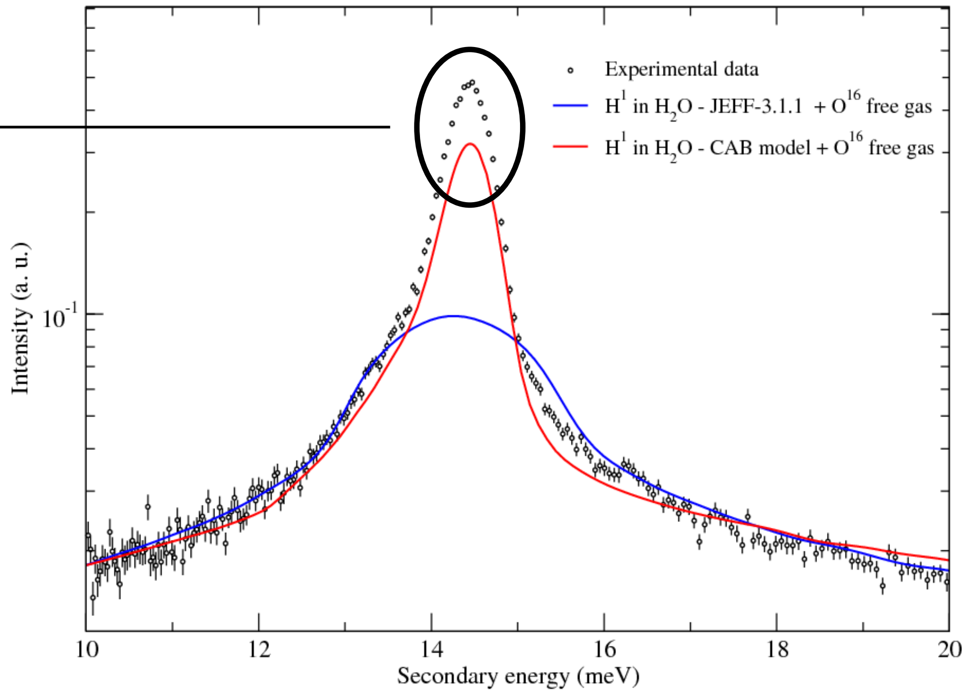
Source of such discrepancies between the CAB model and the data

⇒ non-classical behavior in the diffusion of the water molecules ?

⇒ molecules could diffuse in a step-like movement.

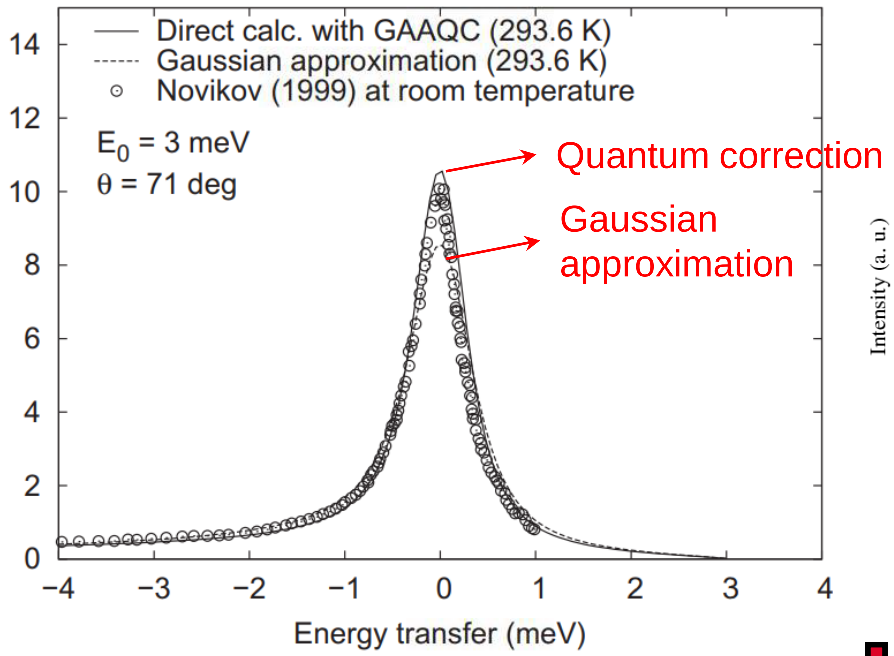
⇩

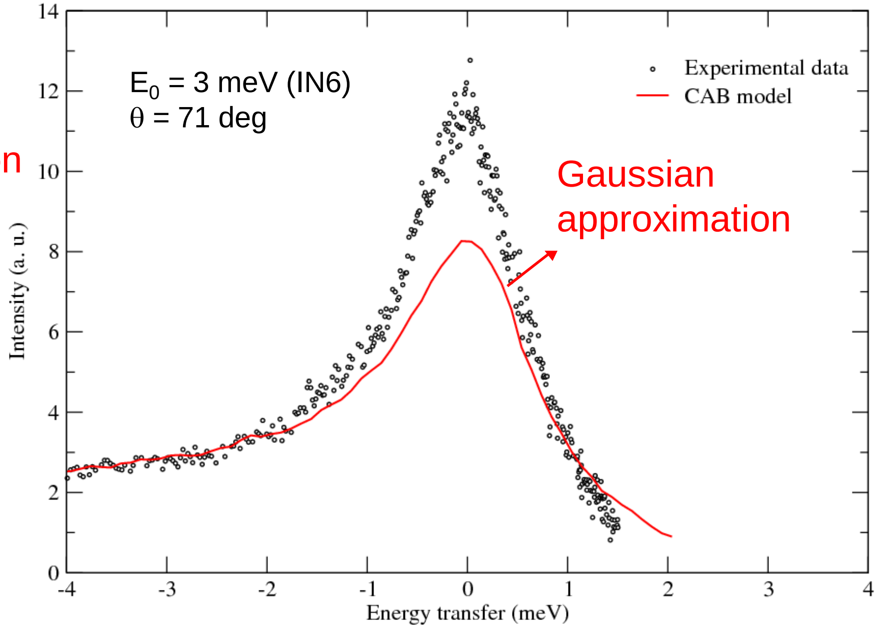
LEAPR module deficiency?



Double-differential neutron cross section H_2O

To compensate the deficiencies of the diffusion model, Abe et al. (ANE 83, 302, 2015) applied a **quantum correction** to the scattering function. Similar work was done by J.I. Márquez Damián (ANE 92, 107, 2016)

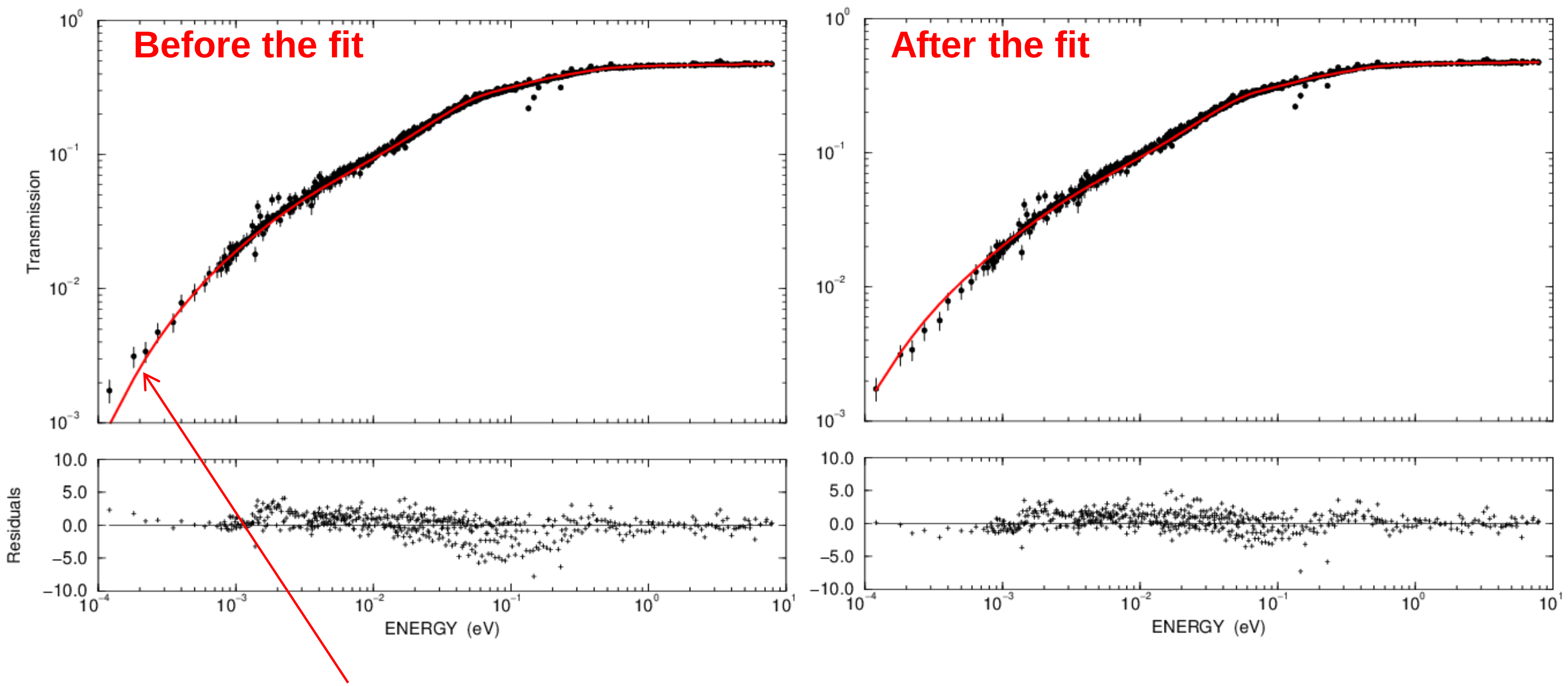




The nature of the discrepancies between the calculation and the ILL data seems to be the same.

Total cross section H2O

Two-step CONRAD calculation for producing Model Parameter Covariance Matrix for GROMACS (CAB model)



prior model parameters from
CAB MODEL

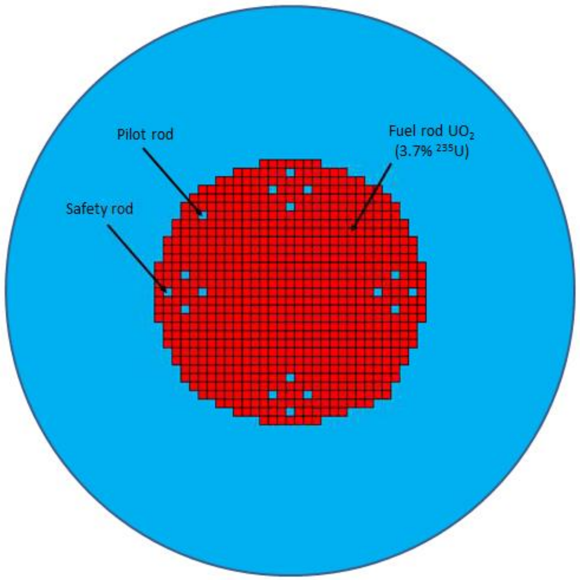
Prior and posterior model parameters are nearly equivalent

$\Rightarrow$ Good agreement with the EXFOR data

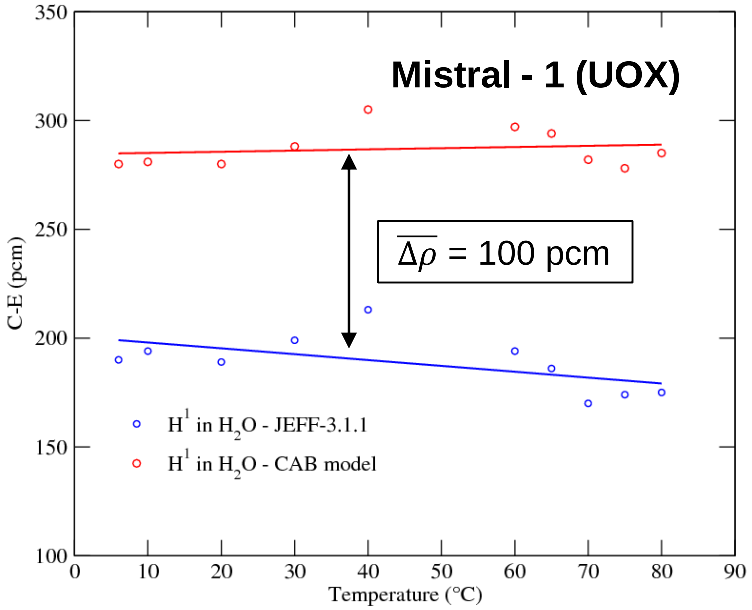
Experimental validation with integral benchmarks H2O

EOLE reactor (CEA Cadarache, France)

Mistral - 1 (UOX core)



- 750 fuel cells (3.7% in ^{235}U)
- 17 guide tubes
- Cell pitch 1.32 cm
- Moderation ratio 1.7
- Moderator: $H_2O+H_3BO_3$
- Reactivity control: [B]



When the CAB model is used in the JEFF-3.1.1 library, the slope as a function of the temperature vanishes

NCSU model for UO2

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0.000000+0 0.000000+0          0          0         0          6  48 1451     2
1.000000+0 5.000000+0          0          0        12          8  48 1451     3
0.000000+0 0.000000+0          0          0        31          3  48 1451     4
U(UO2)     LEIP LAB   EVAL-OCT16 J.L. Wormald, Y. Zhu, A.I. Hawari  48 1451     5
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                                 1        451        38          0  48 1451    36
                                 7          2      1305          0  48 1451    37
                                 7          4     46810          0  48 1451    38
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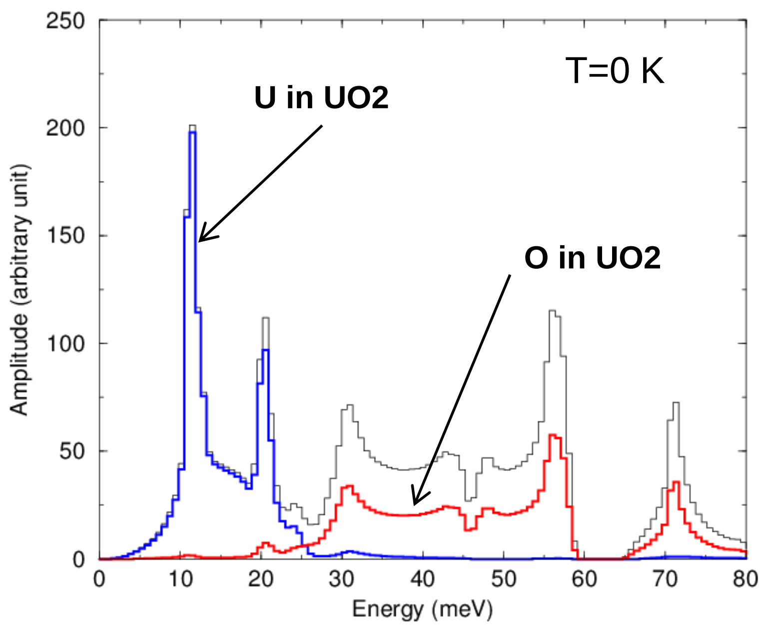
New TSL file from North Caroline South University (NCSU)



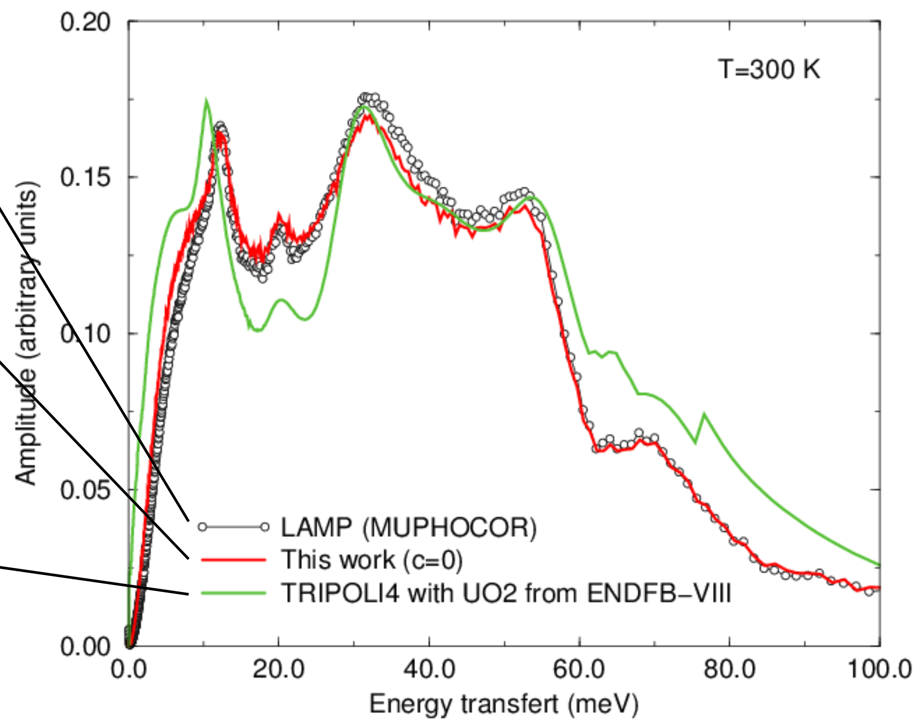
Experimental validation DOS of UO2

Experimental phonon spectrum extracted from ILL data T=300 K

MUPHOCOR (LAMP analysis tool) provide an experimental phonon spectrum by using equations similar to those available in the LEAPR formalism

Nearly similar results can be directly obtained by using the cumulative distribution of $S_{sym}(q,\omega)$

Differences between ILL data and TRIPOLI4 simulations performed with the NCSU model still not well understood

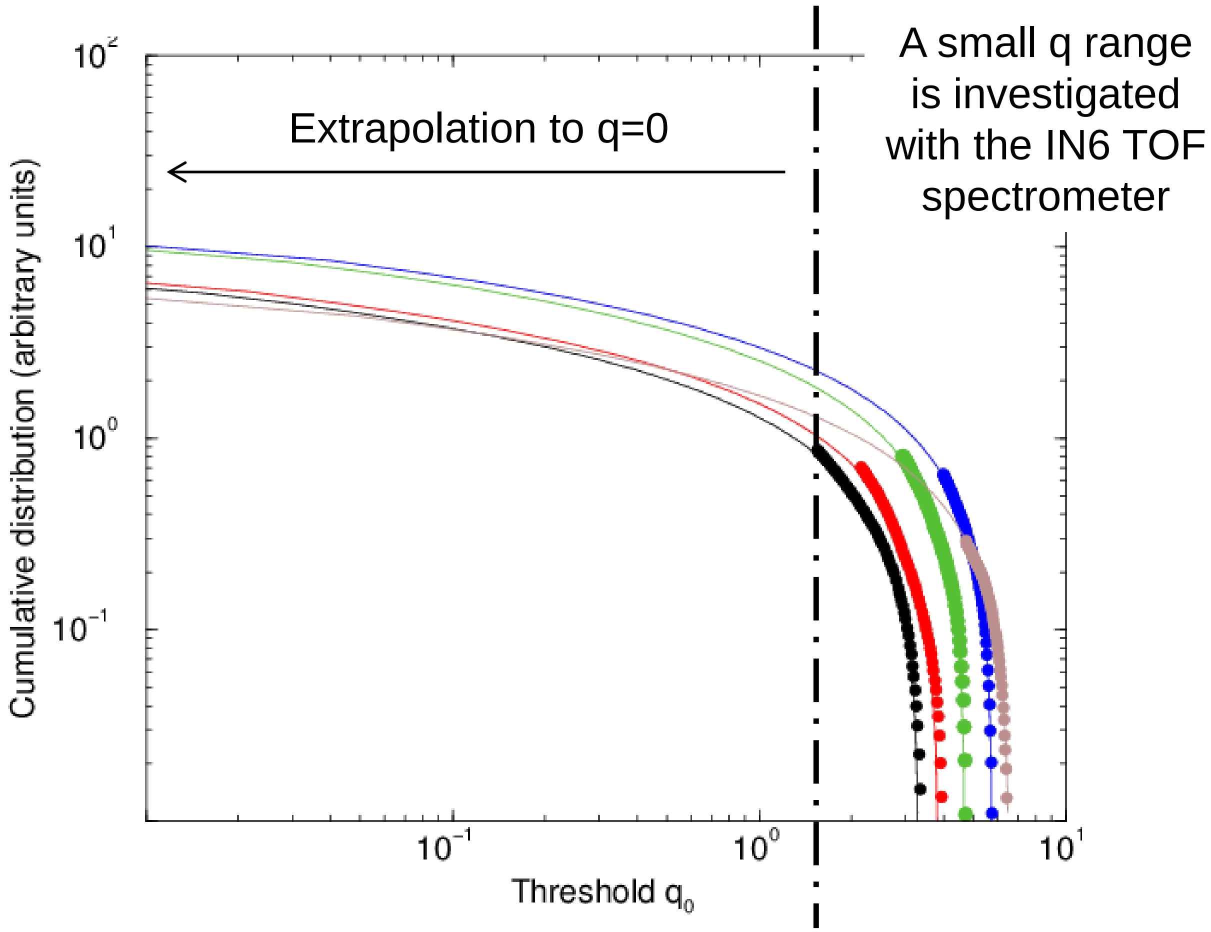


⇩

Difficult to achive a meaningfull experimental validation of the phonon spectrum

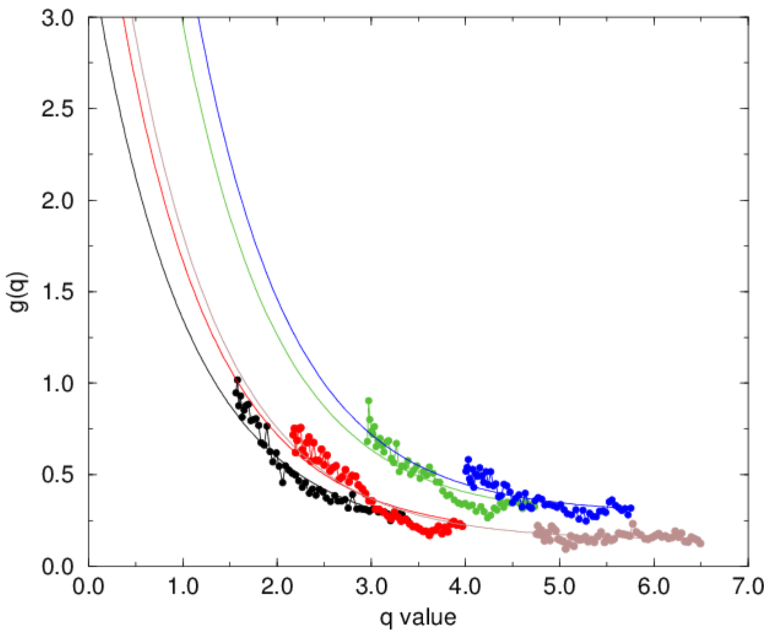
Experimental validation DOS of UO2

Experimental phonon spectrum extracted from ILL data T=300 K



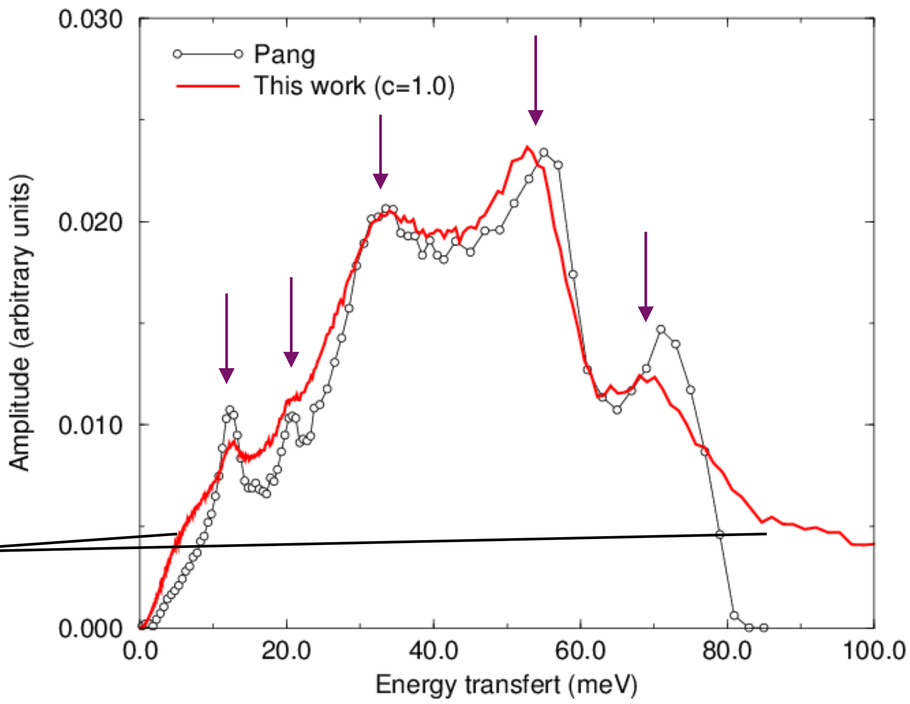
Experimental validation DOS of UO2

Experimental phonon spectrum extracted from ILL data T=300 K



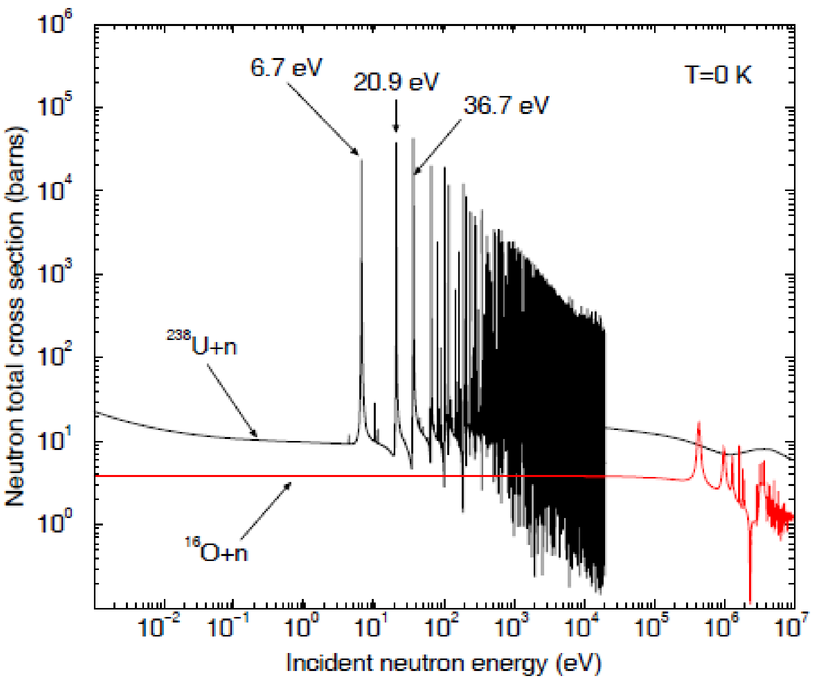
The use of the cumulative distribution of $S_{sym}(q,\omega)$ allows to get an experimental phonon spectrum in reasonable agreement with data from J. Pang (Phys. Rev. B89, 115132, 2014)

« background contribution » in the ILL data which is not taken into account in the TRIPOLI4 simulations



Total cross section UO2

Doppler broadening of neutron-induced resonances using LEAPR formalism



Eur. Phys. J. Plus (2018) **133**: 177
DOI 10.1140/epjp/i2018-12009-y

THE EUROPEAN PHYSICAL JOURNAL PLUS

Regular Article

Doppler broadening of neutron-induced resonances using ab initio phonon spectrum*

G. Noguere[1,a], P. Maldonado[2], and C. De Saint Jean[1]
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[2] Department of Physics and Astronomy, Uppsala University, Box 516, S-75120 Uppsala, Sweden

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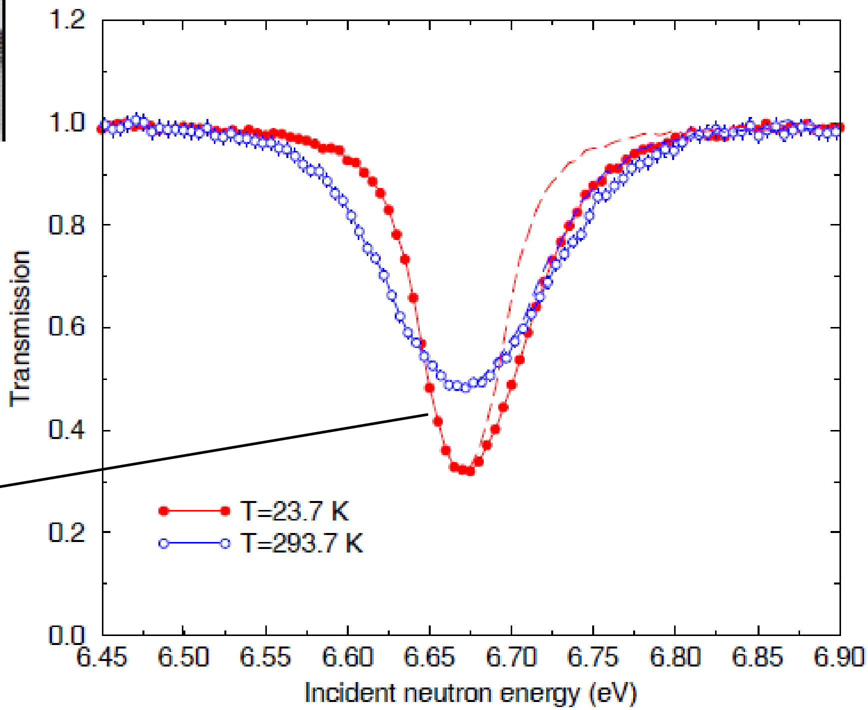
Abstract. Neutron resonances observed in neutron cross section data can only be compared with their theoretical analogues after a correct broadening of the resonance widths. This broadening is usually carried out by two different theoretical models, namely the Free Gas Model and the Crystal Lattice Model, which, however, are only applicable under certain assumptions. Here, we use neutron transmission experiments on UO_2 samples at $T = 23.7$ K and $T = 293.7$ K, to investigate the limitations of these models when an *ab initio* phonon spectrum is introduced in the calculations. Comparisons of the experimental and theoretical transmissions highlight the underestimation of the energy transferred at low temperature and its impact on the accurate determination of the radiation widths $\Gamma_{\gamma\lambda}$ of the ^{238}U resonances λ. The observed deficiency of the model represents an experimental evidence that the Debye-Waller factor is not correctly calculated at low temperature near the Neel temperature ($T_N = 30.8$ K).

we use recent spectra calculated at 23 K and 300 K
by Pablo Maldonado

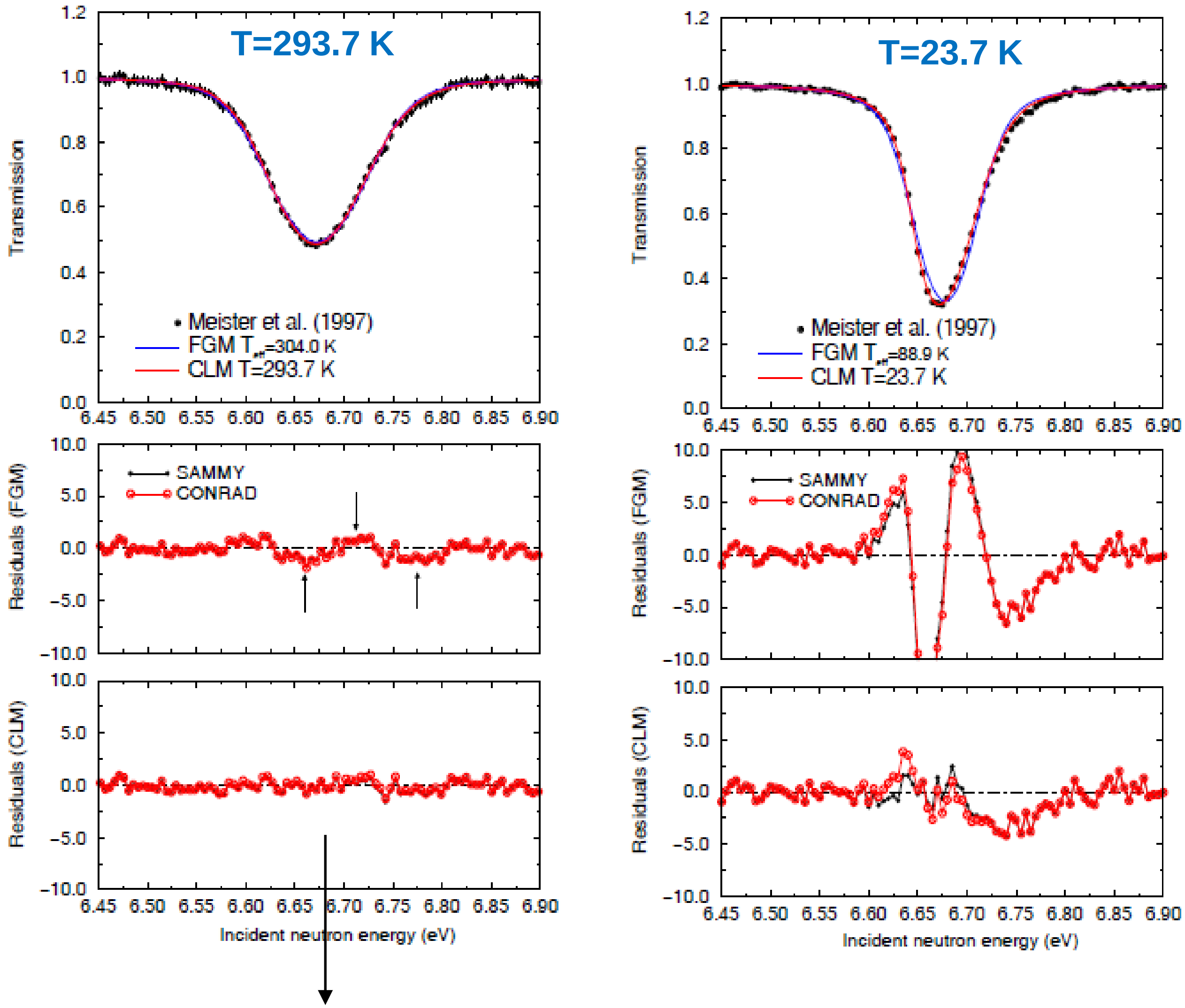
Total cross section UO2

Measurements were performed by Meister et al. (1997) at the GELINA facility (JRC-Geel, Belgique) with the Time Of Flight technique at L=30 m



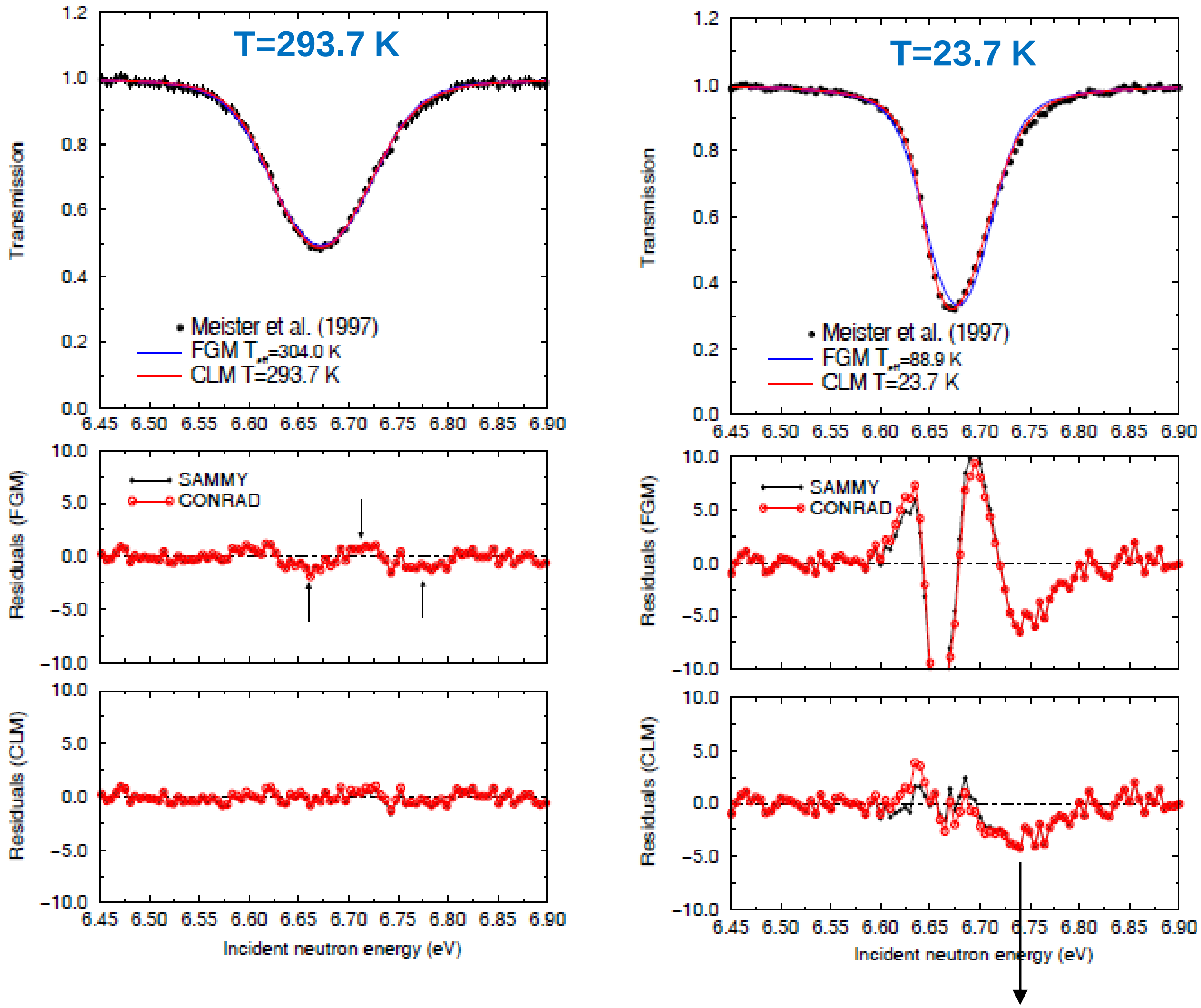
Strong assymetry observed at low temperature (T=23 K)

Total cross section UO2



At room temperature, LEAPR formalism provides an excellent agreement with the data

Total cross section UO2



At T=23.7 K, model not adapted to account for the strong coupling between the lattice vibrations and the magnetic properties of UO_2 (+Jahn-Teller distortion of the oxygen cage)

Conclusions

Double-differential neutron cross sections

- Provide qualitative information
- Difficult to explain the differences between the data and the simulation
- Inter-comparaison exercises between facilities are needed

Neutron total cross section

- Provide quantitative « integrated » information in the thermal and resonance ranges
- Needs for transmission data at high temperature !
- Needs for data below 1 meV
- Difficult to get beam time on the existing cold neutron sources to do « simple » transmission measurements

Integral benchmarks (keff)

- Provide global trends on TSL
- Sensitivity studies are needed to investigate « compensation » effects