Evaluation of the Resolved Resonance Range

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APRENDE Experimentalists-Evaluators workshop (WP2-WP4), 26-27/02/2025, IPHC Strasbourg

Context

WP2 New nuclear data measurements

Task4.2 (n,f) cross section
Task4.3 (n,γ) cross section

Pu239(n,tot)
Pu239(n,f)
ν_p(Pu239)
Pu241(n,γ)
Pu241(n,f)
α(Pu241)
U238(n,γ)
Bi209(n,γ)
Er166(n,γ)
Er167(n,γ)
Cu63(n,γ)
Cu65(n,γ)

Resonance analysis

WP4 Nuclear data evaluation

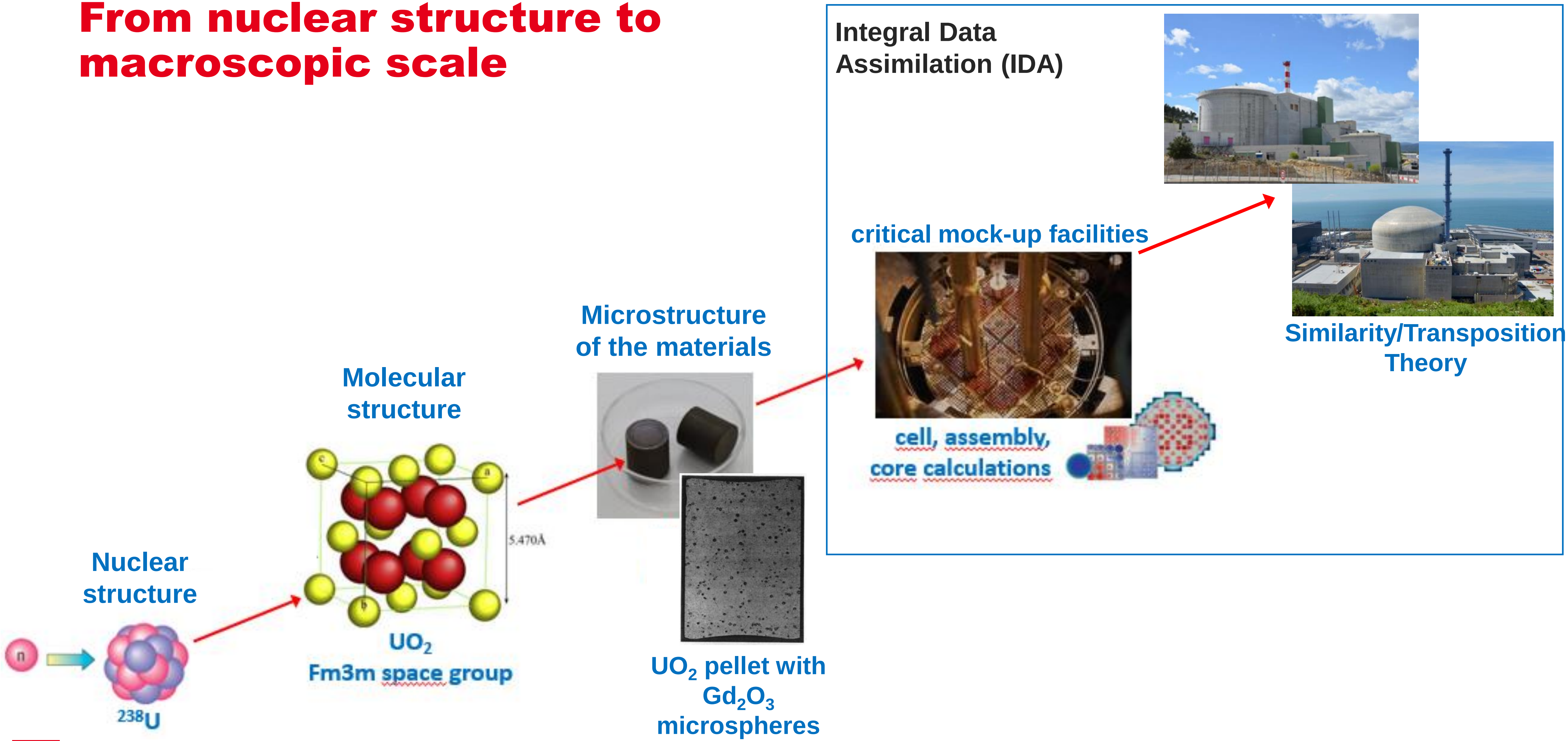
Task4.2 Thermal Scattering Laws
Task4.4 Cross section evaluation

U233
U234
U235
U238
Pu239
Pu240
Pu241
Zr

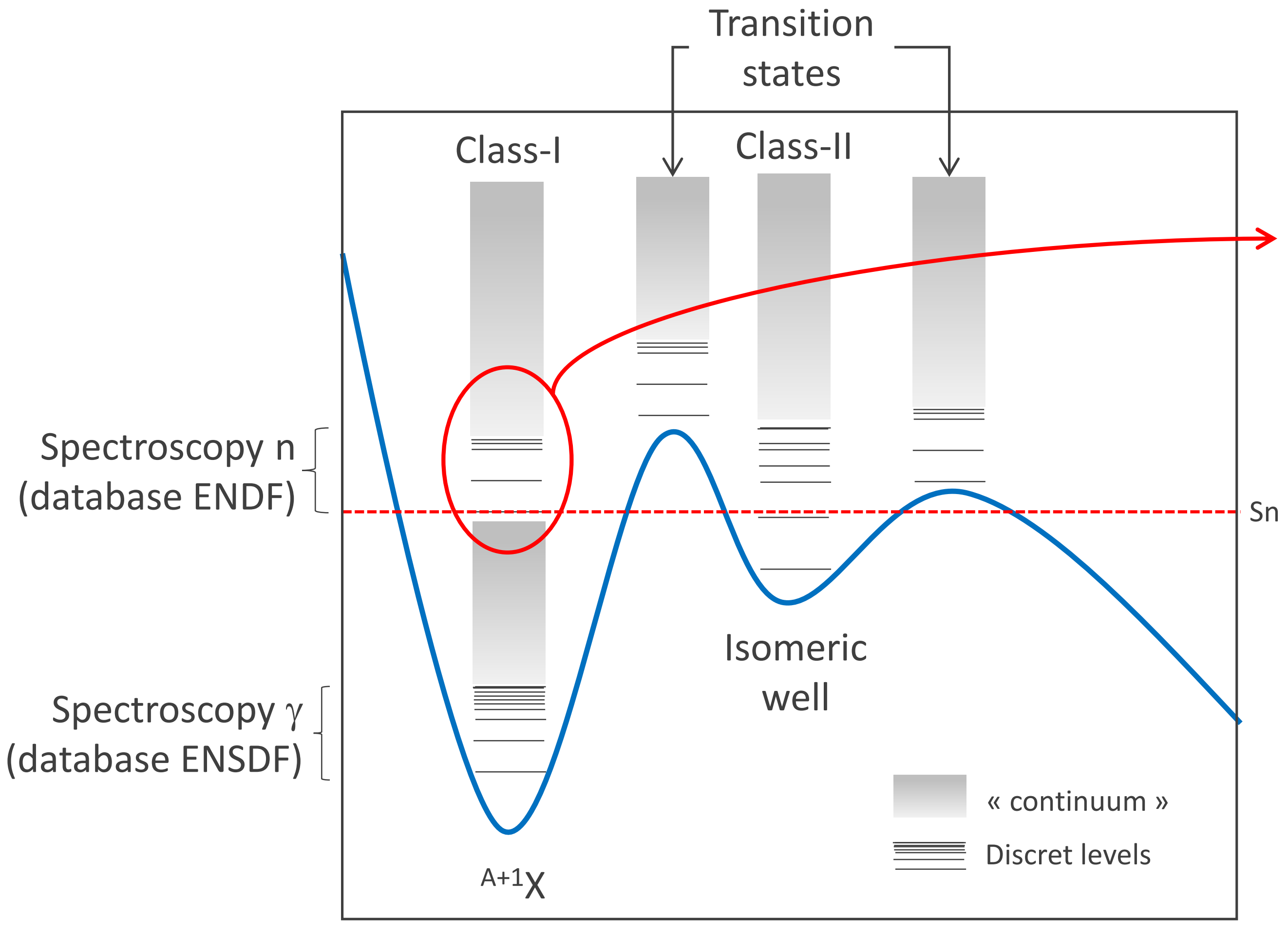
TSL from ESS
ZrH_x

Evaluation of the Resolved Resonance Range

From nuclear structure to macroscopic scale



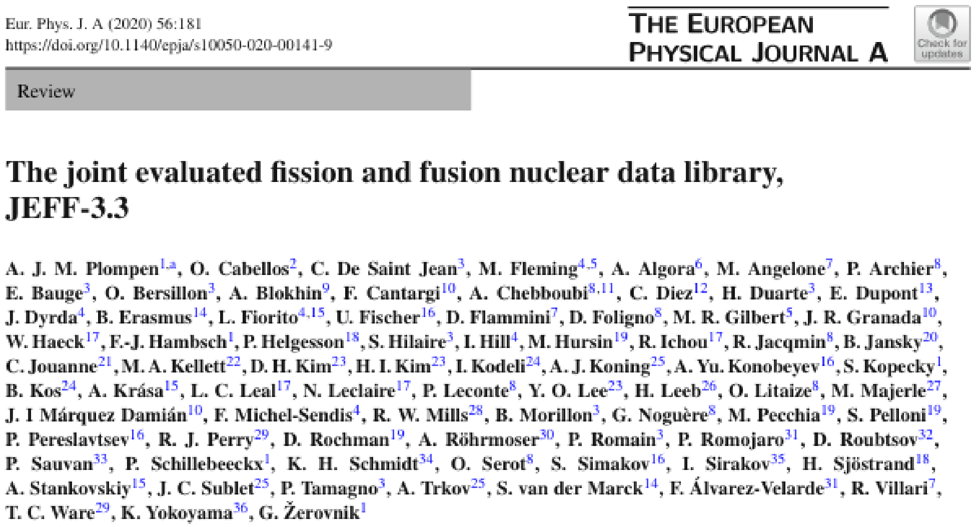
Nuclear structure



Potential energy as a function of the nucleus deformation

The Resonance Shape Analysis relies on the **R-Matrix** theory to extract properties of the compound nucleus states (resonance parameters) from time-of-flight measurements

Prior resonance parameters comes from evaluated nuclear data libraries in ENDF-6 format



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THE EUROPEAN PHYSICAL JOURNAL A

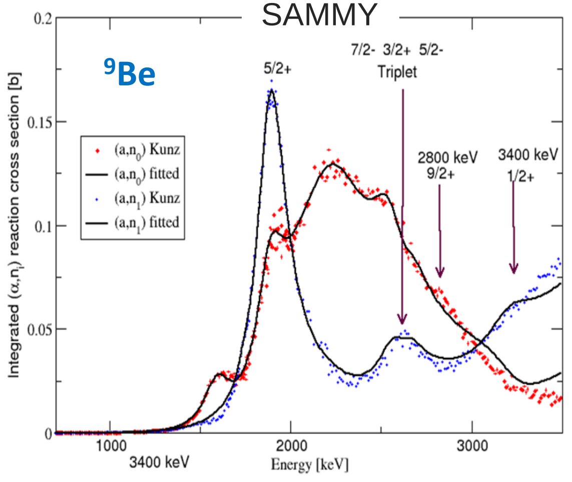
Review

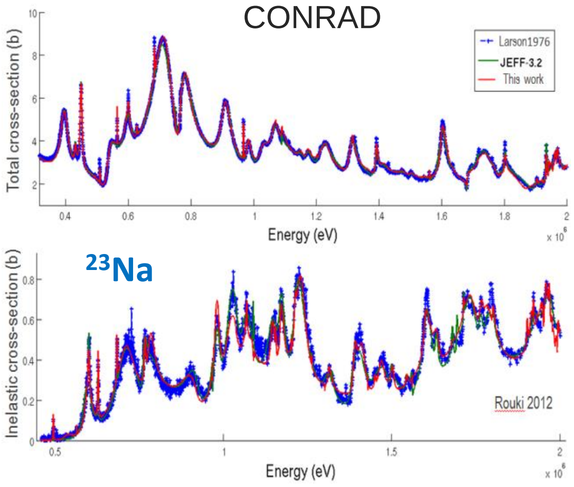
The joint evaluated fission and fusion nuclear data library, JEFF-3.3

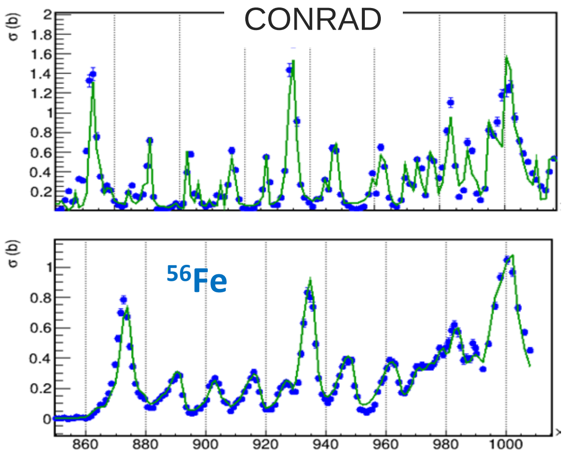
A. J. M. Plompen[1,a], O. Cabellos[2], C. De Saint Jean[3], M. Fleming[4,5], A. Algora[6], M. Angelone[7], P. Archier[8], E. Bauge[3], O. Bersillon[3], A. Blokhin[9], F. Cantargi[10], A. Chebboubi[8,11], C. Diez[12], H. Duarte[3], E. Dupont[13], J. Dyrda[4], B. Erasmus[14], L. Fiorito[4,15], U. Fischer[16], D. Flammini[7], D. Foligno[8], M. R. Gilbert[5], J. R. Granada[10], W. Haeck[17], F.-J. Hambsch[1], P. Helgesson[18], S. Hilaire[3], I. Hill[4], M. Hursin[19], R. Ichou[17], R. Jacqmin[8], B. Jansky[20], C. Jouanne[21], M. A. Kellett[22], D. H. Kim[23], H. I. Kim[23], I. Kodeli[24], A. J. Koning[25], A. Yu. Konobeyev[16], S. Kopecky[1], B. Kos[24], A. Krása[15], L. C. Leal[17], N. Leclaire[17], P. Leconte[8], Y. O. Lee[23], H. Leeb[26], O. Litaize[8], M. Majerle[27], J. I Márquez Damián[10], F. Michel-Sendis[4], R. W. Mills[28], B. Morillon[3], G. Noguère[8], M. Pecchia[19], S. Pelloni[19], P. Pereslavtsev[16], R. J. Perry[29], D. Rochman[19], A. Röhrmoser[30], P. Romain[3], P. Romojaro[31], D. Roubtsov[32], P. Sauvan[33], P. Schillebeeckx[1], K. H. Schmidt[34], O. Serot[8], S. Simakov[16], I. Sirakov[35], H. Sjöstrand[18], A. Stankovskiy[15], J. C. Sublet[25], P. Tamagno[3], A. Trkov[25], S. van der Marck[14], F. Álvarez-Velarde[31], R. Villari[7], T. C. Ware[29], K. Yokoyama[36], G. Žerovnik[1]

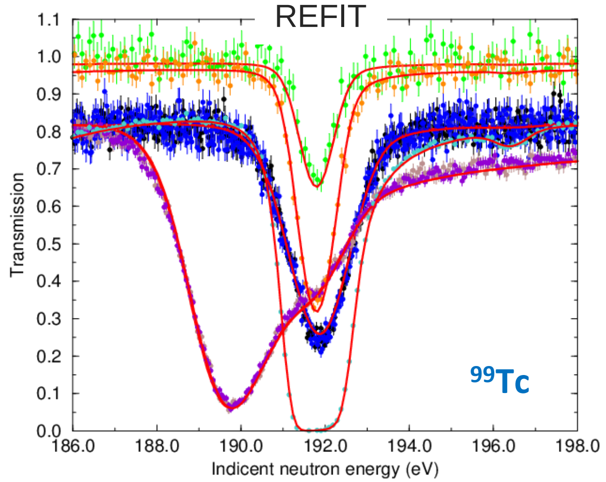
Nuclear structure

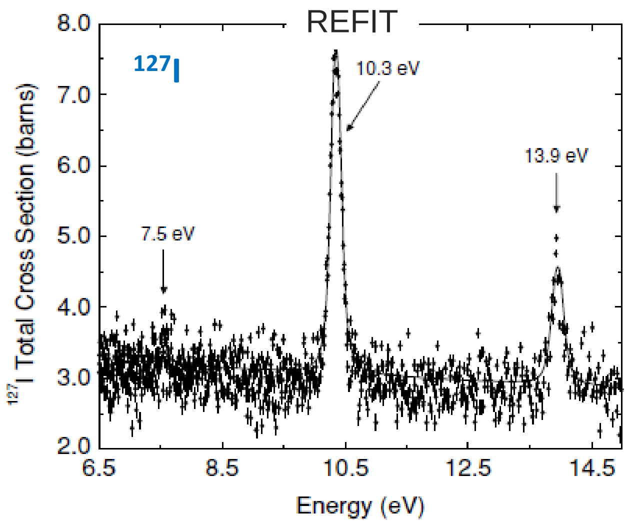
R-Matrix formalism is a well established formalism for studying nuclear properties, from light to heavy nuclei

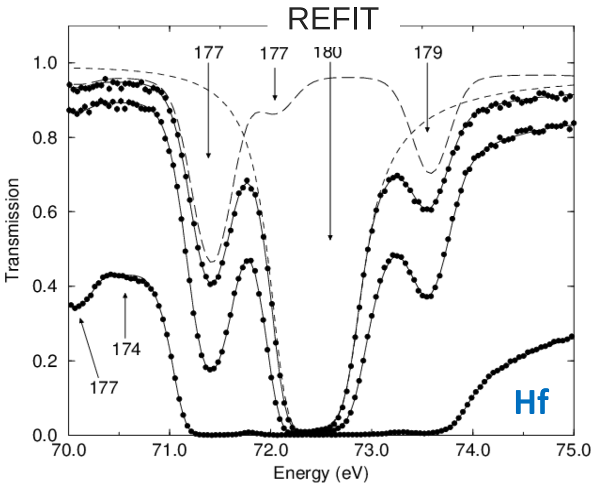


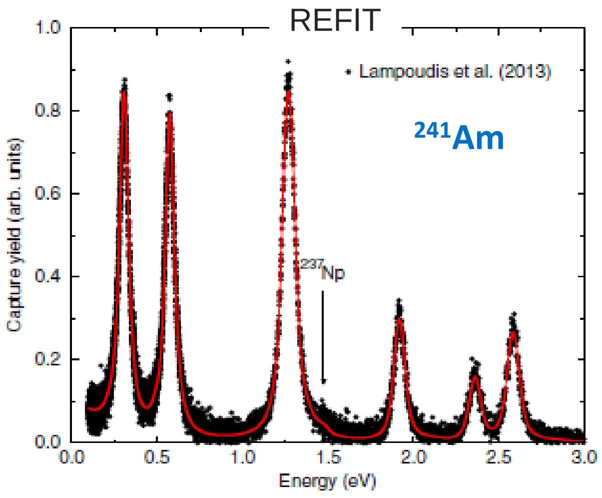


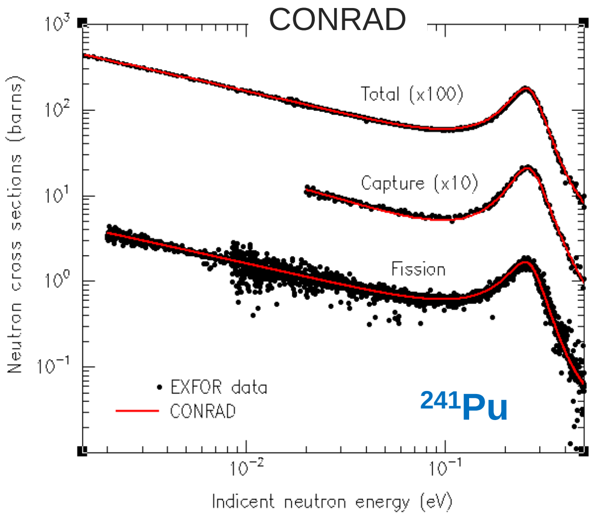






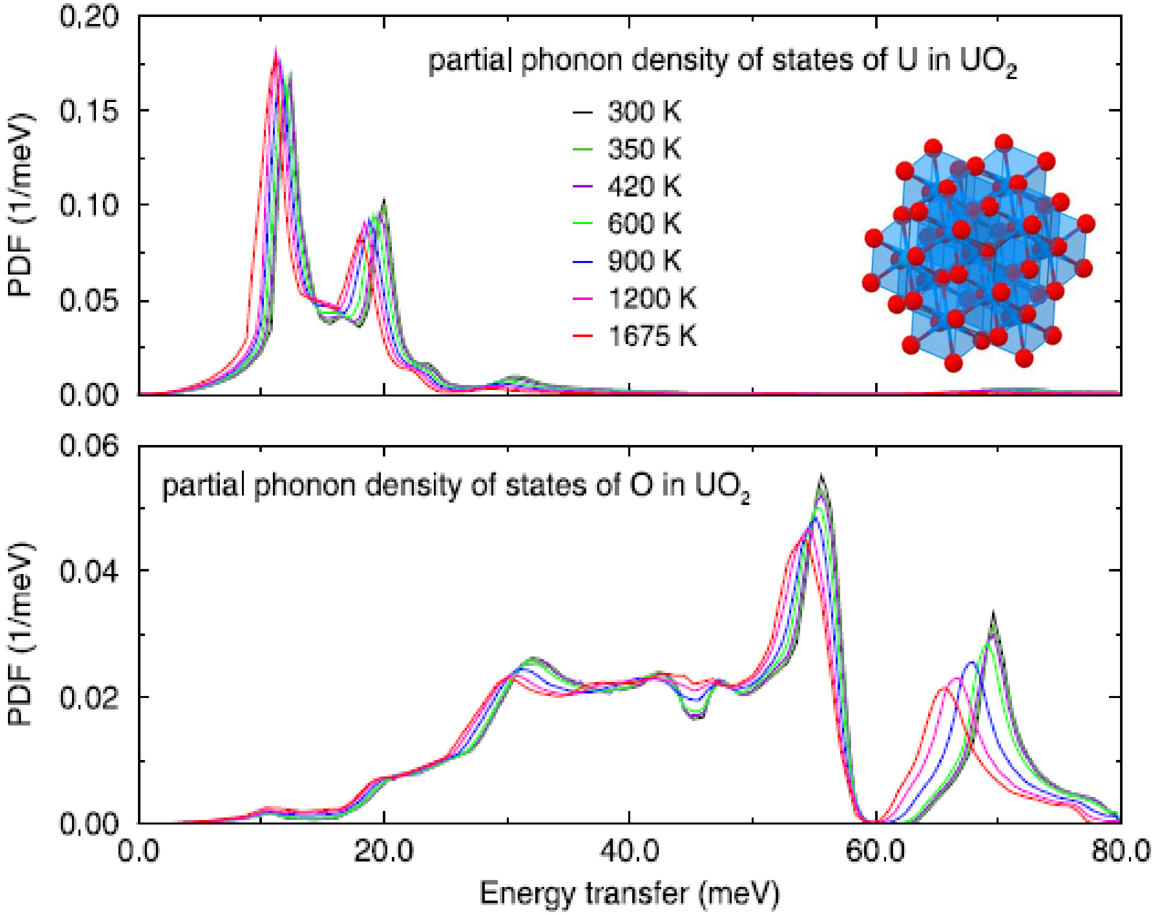




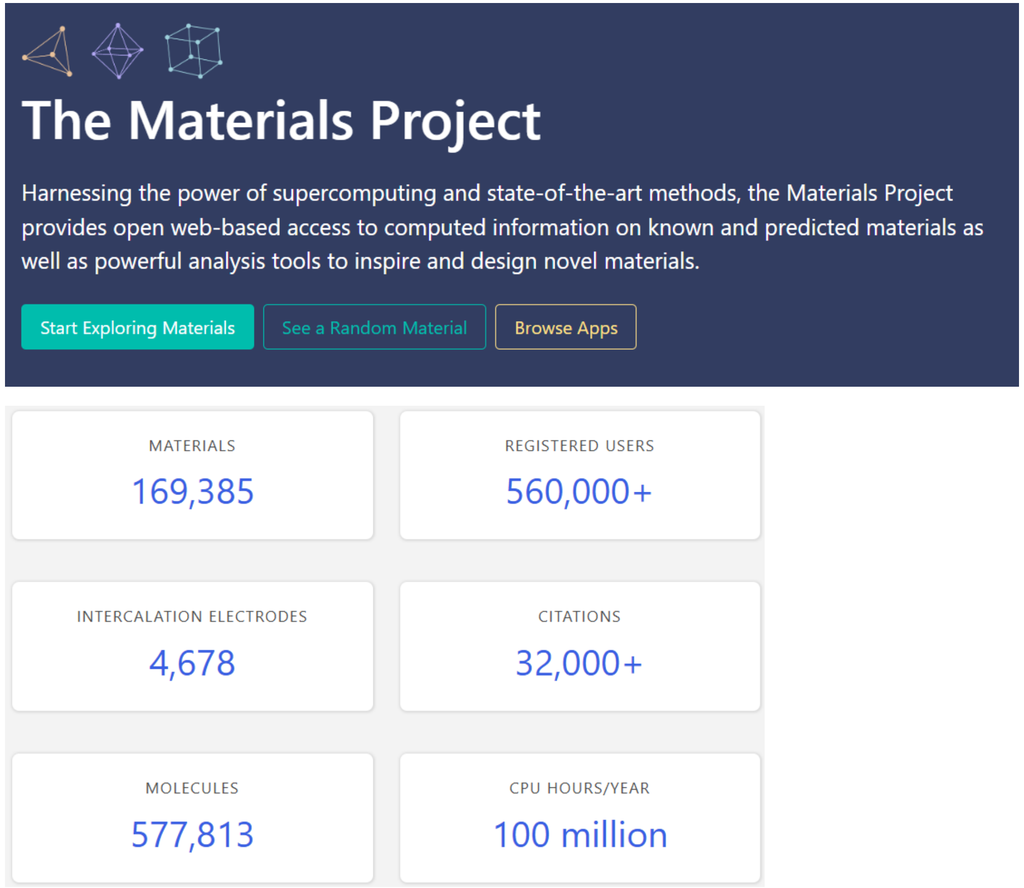


Molecular structure and dynamics

Nuclear model codes (**FLASSH**, **nCRYSTAL**, **CINEL**) take into account crystal structure properties (space group, lattice parameter) and vibrations of the target nuclei (partial phonon density of states)



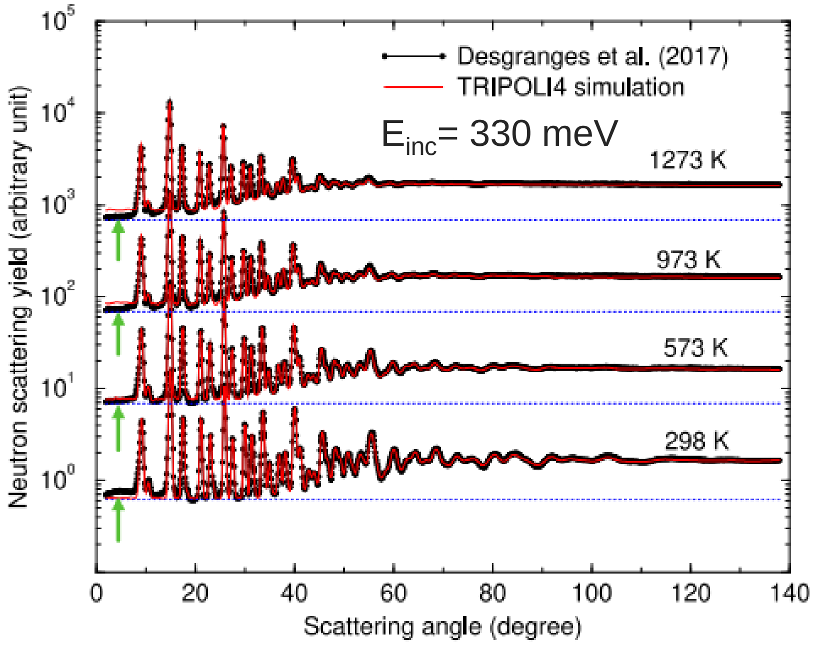
Prior material properties can be retrieved from the Materials Project database



Molecular structure and dynamics

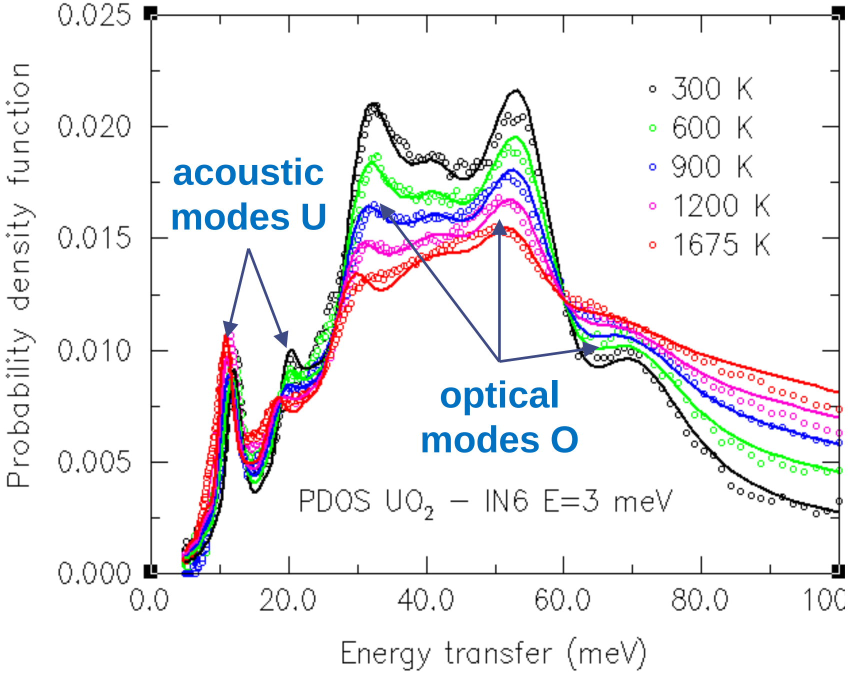
Temperature-dependence of the material properties can be accessed either by Ab Initio Molecular Dynamics (AIMD) or from diffraction, inelastic neutron scattering (INS) and neutron Compton scattering (NCS) experiments

UO_2 diffraction pattern measured at ILL with D4

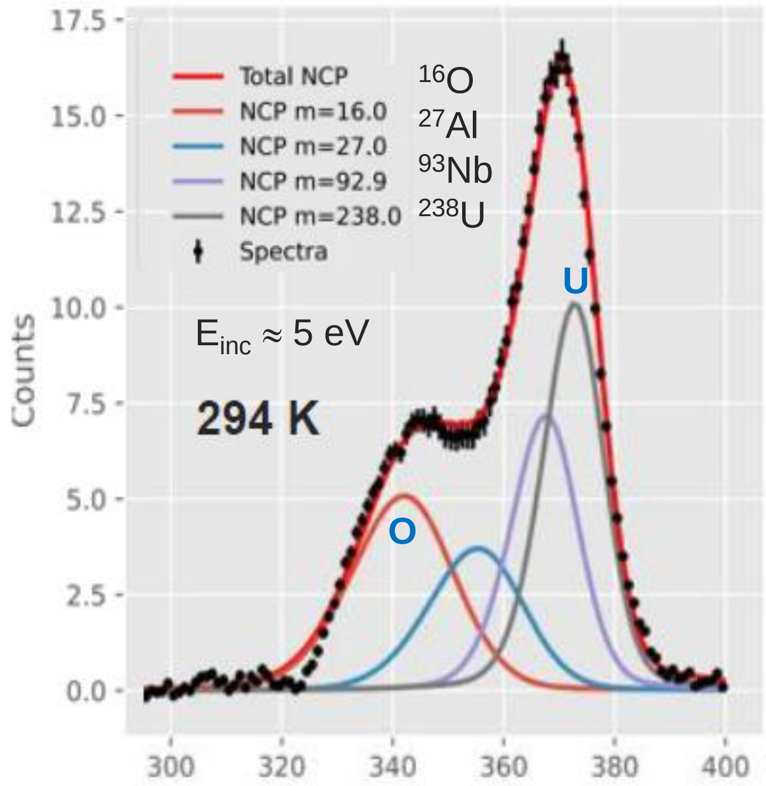


Fm3m group space

PDOS of UO_2 measured at ILL with IN6

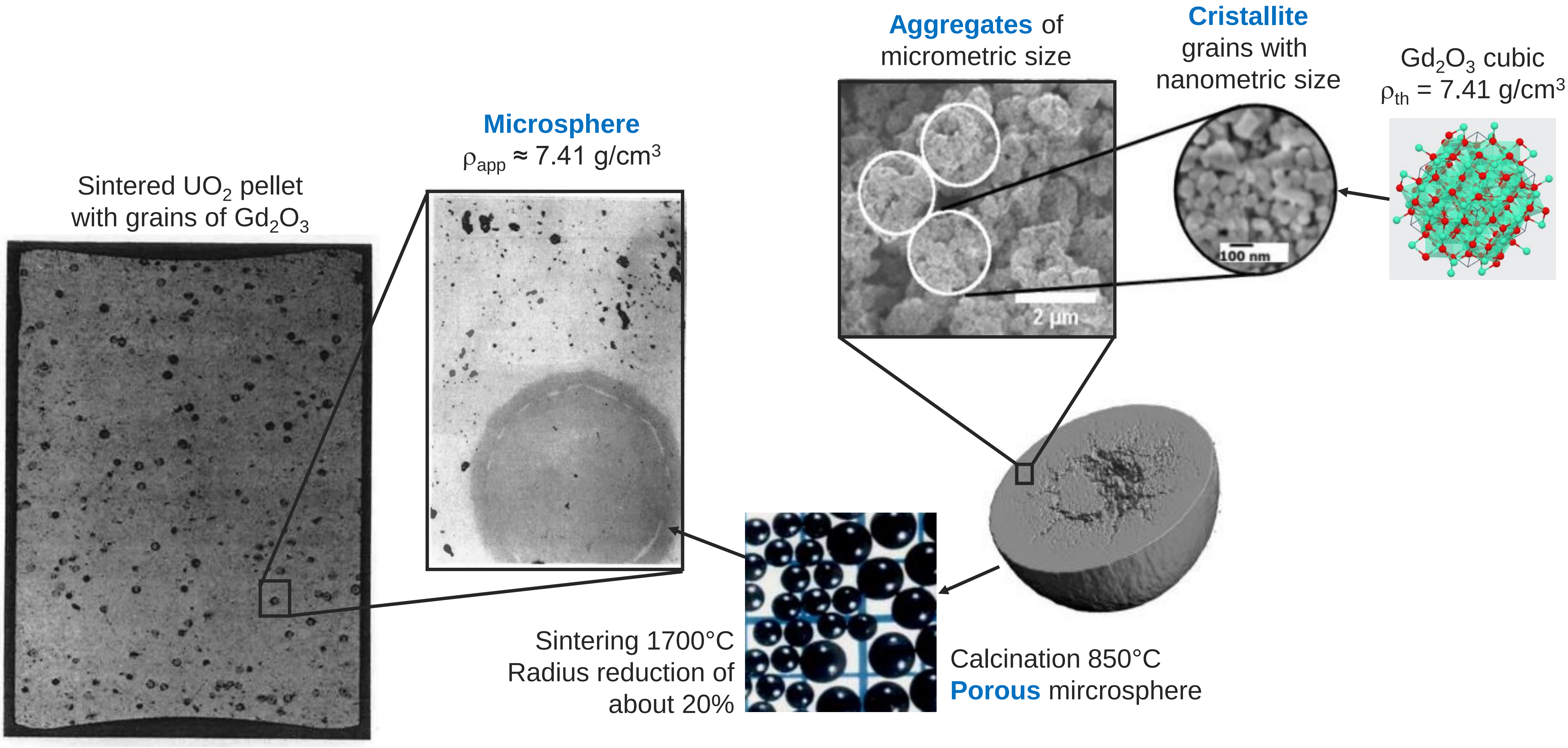


NCS of UO_2 measured at ISIS (UK) with VESUVIO



Neutron Compton scattering (or deep inelastic neutron scattering) measures atomic momentum distributions as a function of the energy transfer

Microstructure of the materials



Microstructure of the materials

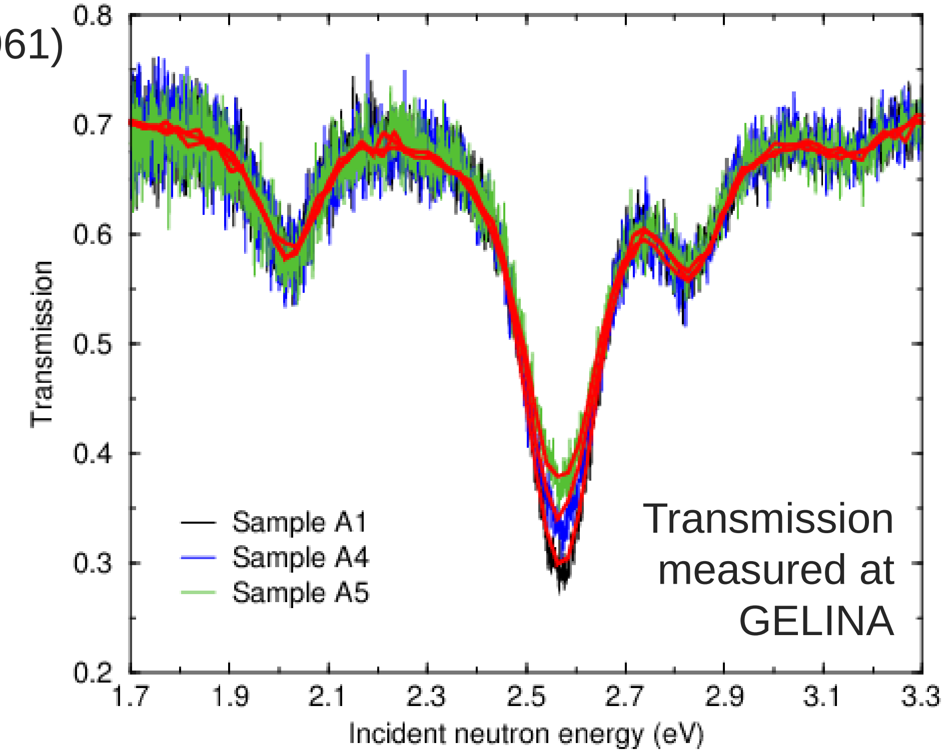
Particle self-shielding correction from Doub (NSE, 10, 299, 1961)

$$\bar{\sigma}_{x,i}(E) = f(E)\sigma_{x,i}(E) \quad \text{with} \quad f = \frac{1}{\frac{2}{3}y\left(\frac{V}{g}\right)} \ln\left(\frac{1}{1-\left(\frac{V}{g}\right)(1-\bar{t})}\right)$$

Expression of *t* derived from Case et al. , Introduction to the theory of neutron diffusion, 1953

$$\bar{t} = \frac{2}{y^2}(1-(1+y)e^{-y}) \quad \text{with} \quad y = 2r\Sigma$$

In which *r* is the radius of the mircosphere and *V* depends on the number of microspheres



Transmission measured at GELINA

UO2 Sample	Gd2O3 Mircrophere diameter	Number of mircrospheres in a pellet
A1	-	UO2 sample containing homogenously mixed Gd2O3 powder
A4	195(10) µm	5955
A5	380(19) µm	814

Evaluation of the Resolved Resonance Range

From nuclear structure to macroscopic scale

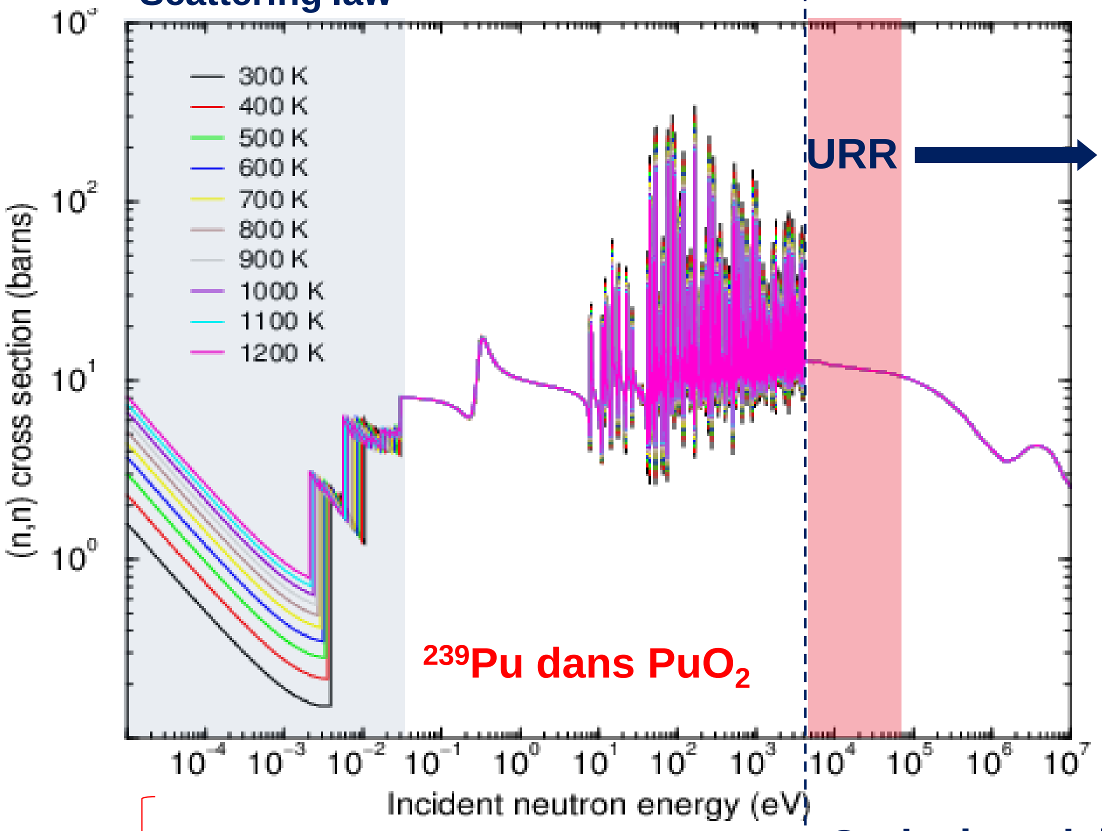
Mix nuclear models and experimental corrections

- Resolution broadening
- Doppler broadening
- Multiple scattering correction

Conclusions

Mix nuclear models and experimental corrections

Thermal Scattering law | RRR | « Continuum »



Average parameters calculated with the urr option of TALYS

⬇

Probability Tables

+ Experimental corrections

Standard models implemented in CONRAD

R-Matrix formalism
MLBW, RM, LRF7, Brune parametrization, (n,γf),$P_f(E)$,$R_{eff}(E)$, R∞…

Optical model (ECIS, CCCP)
Statistical model (TALYS, CONRAD)

ESTIMA method | SPRT method

Convert results in ENDF-6 file and processing for TRIPOLI

Mix nuclear models and experimental corrections

Most popular corrections in resonance analysis

Theoretical transmission

$$T_R(E) = \int_0^\infty R(E,E')\, T_{th}(E')\, \mathrm{d}E'$$

Resolution function

$$T_{th}(E) = \mathrm{e}^{-\sum_i n_i \sigma_{T,i}(E)}$$

Doppler broadened cross section

Theoretical reaction yield

$$Y_R(E) = \int_0^\infty R(E,E')\, Y_{th}(E')\, \mathrm{d}E'$$

$$Y_{th}(E) = (1+\alpha(E))\underbrace{(1-T_{th}(E))\frac{\sum_i n_i \sigma_{\gamma,i}(E)}{\sum_i n_i \sigma_{T,i}(E)}}_{Y_0(E)}\varepsilon_{\mathrm{cw}}(E_{\mathrm{n}}) + \varepsilon_{\mathrm{nw}}(E_{\mathrm{n}})Y_{\mathrm{n}}(E_{\mathrm{n}})$$

Neutron sensitivity

Multiple scattering correction

$Y_{th}(E) = Y_0(E)+Y_1(E)+Y_n(E)$

+ γ-ray attenuation in the sample …

Mix nuclear models and experimental corrections

See P. Schillebeeckx et al. NDS 113, 3054 (2012)

Available online at www.sciencedirect.com

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Nuclear Data Sheets

Nuclear Data Sheets 113 (2012) 3054–3100

www.elsevier.com/locate/nds

Determination of Resonance Parameters and their Covariances from Neutron Induced Reaction Cross Section Data

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Evaluation of the Resolved Resonance Range

From nuclear structure to macroscopic scale

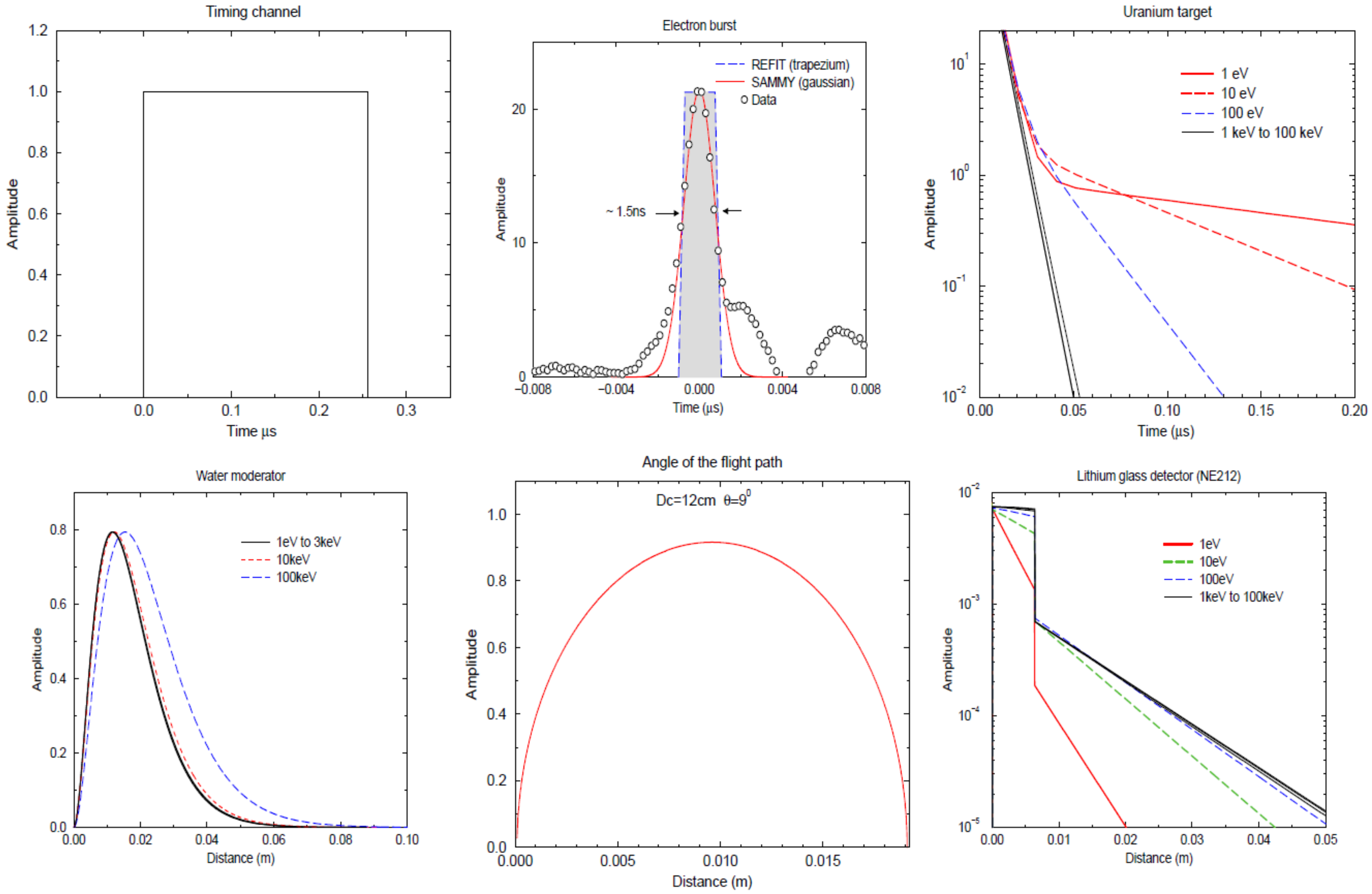
Mix nuclear models and experimental corrections

- **Resolution broadening**
- Doppler broadening
- Multiple scattering correction

Conclusions

Resolution function

Monte-Carlo distributions or **simple functions** can be used to describe the neutron burst (Gaussian or Lorentzian), the neutron target decay (sum of exponentials), the moderator (chi-square distribution with υ degree of freedom), the flight path angle (half-circle) and the detector in the case of transmission



Resolution function

Monte-Carlo distributions or **simple functions** can be used to describe the neutron burst (Gaussian or Lorentzian), the neutron target decay (sum of exponentials), the moderator (chi-square distribution with υ degree of freedom), the flight path angle (half-circle) and the detector in the case of transmission

- **Contribution of the neutron source (uranium target, …)**

$$I_t(t) \approx \frac{\ln(2)}{\tau(E)}\left(k_1 + k_2 E^{k3}\right)e^{-\frac{\ln(2)}{\tau(E)}t} \qquad \tau(E) = \lambda_\tau \frac{72.298}{\sqrt{E}}$$

- **Contribution of the water moderator**

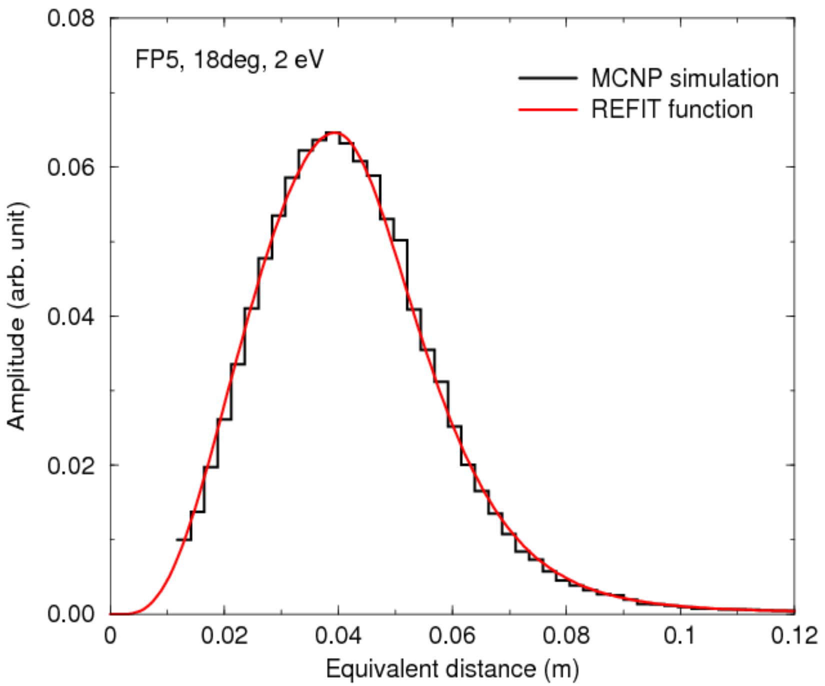
$$I_m(t) = \frac{1}{2}\left(\frac{\sqrt{E}}{72.298\lambda_0}\right)^3 t^2 e^{-\frac{\sqrt{E}}{72.298\lambda_0}t}$$

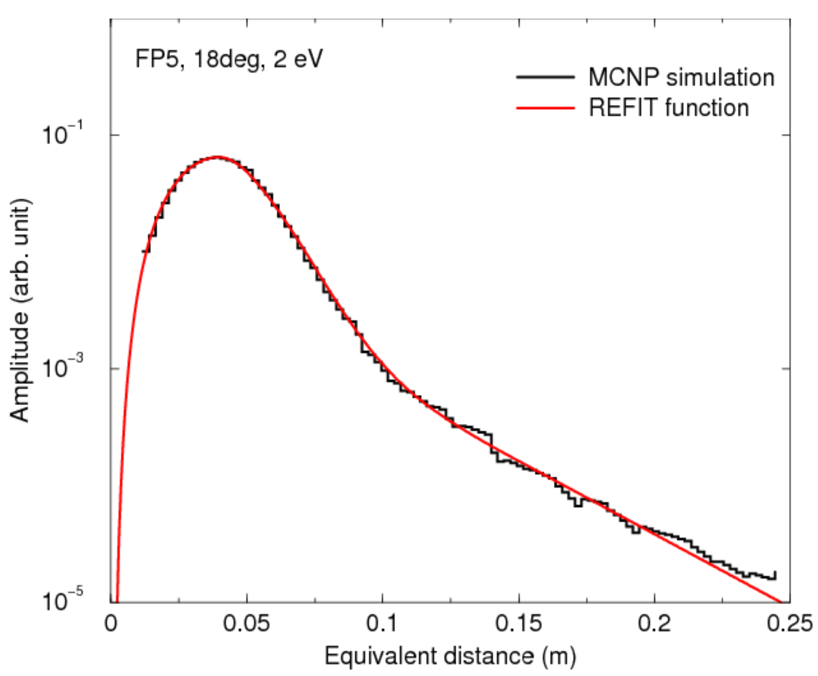
- **Contribution of the flight path angle**

$$I_\theta(t) \propto \sqrt{T_m^2 + \left(T_m - 2t\right)^2} \qquad T_m = \frac{72.298 D_c \tan(\theta)}{\sqrt{E}}$$

Resolution function

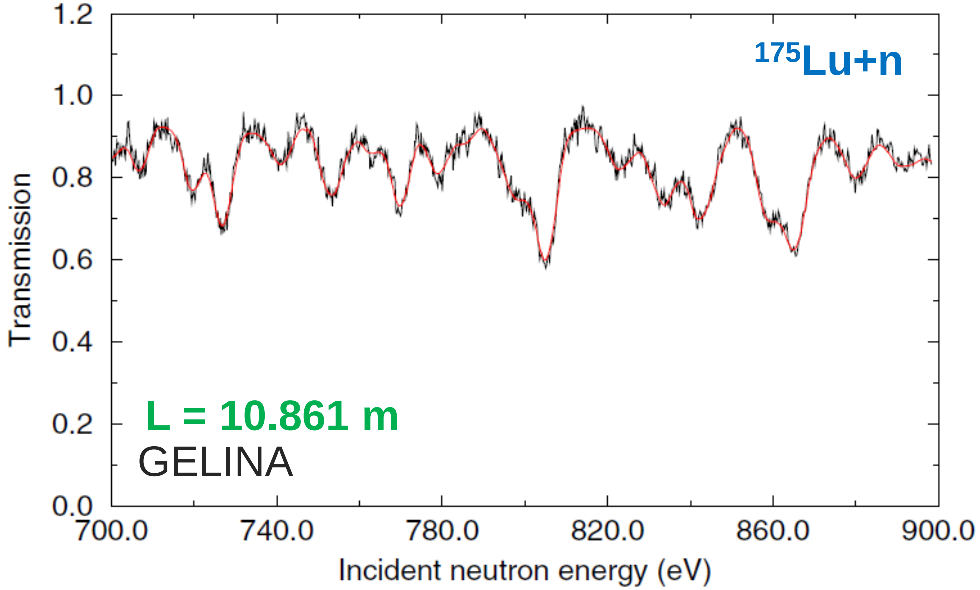
Monte-Carlo distributions or **simple functions** can be used to describe the neutron burst (Gaussian or Lorentzian), the neutron target decay (sum of exponentials), the moderator (chi-square distribution with υ degree of freedom), the flight path angle (half-circle) and the detector in the case of transmission

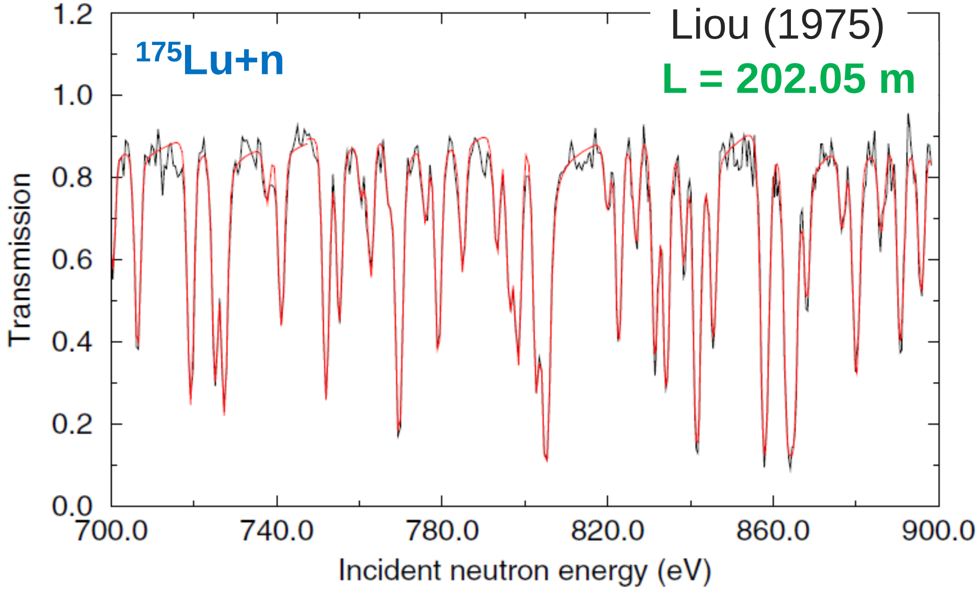




⇒ **Convolution of simple functions** is able to correctly reproduce the Monte-Carlo distributions (GELINA facility)

Resolution broadening





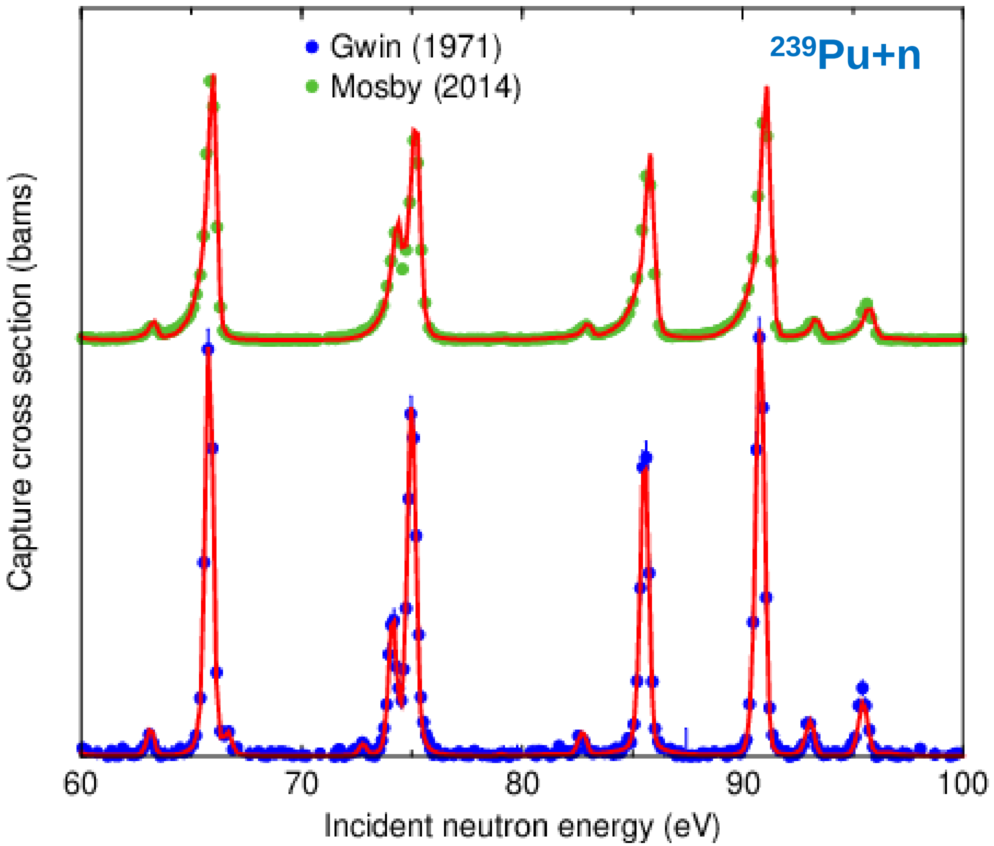
High resolution measurements ⇒ long flight length

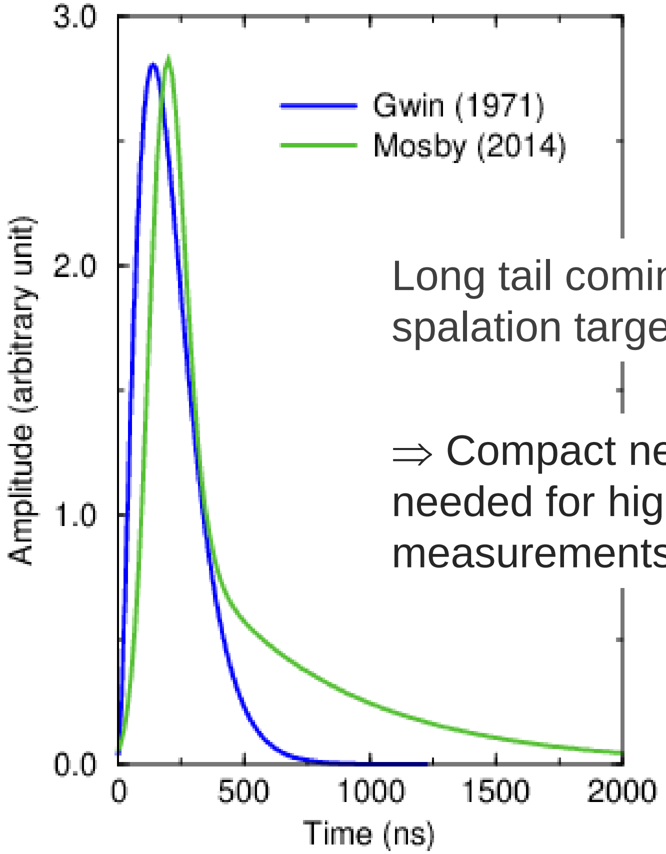
$$\frac{\Delta E}{E} = \frac{2}{L}\sqrt{\frac{E}{t^2(1eV)}\Delta T^2 + \Delta L^2}$$

$$\Delta T^2 = \Delta T_{CW}^2 + \Delta T_{burst}^2$$

$$\Delta L^2 = \Delta L_{mod}^2 + \Delta L_{det}^2$$

Resolution broadening





Long tail coming from the thick spalation target (LANCE)

⇒ Compact neutron source are needed for high resolution measurements

⇒ Warning ! the fitting procedure will accommodate values of the **radiative widths** in order to compensate possible deficiencies of the resolution function

Evaluation of the Resolved Resonance Range

From nuclear structure to macroscopic scale

Mix nuclear models and experimental corrections

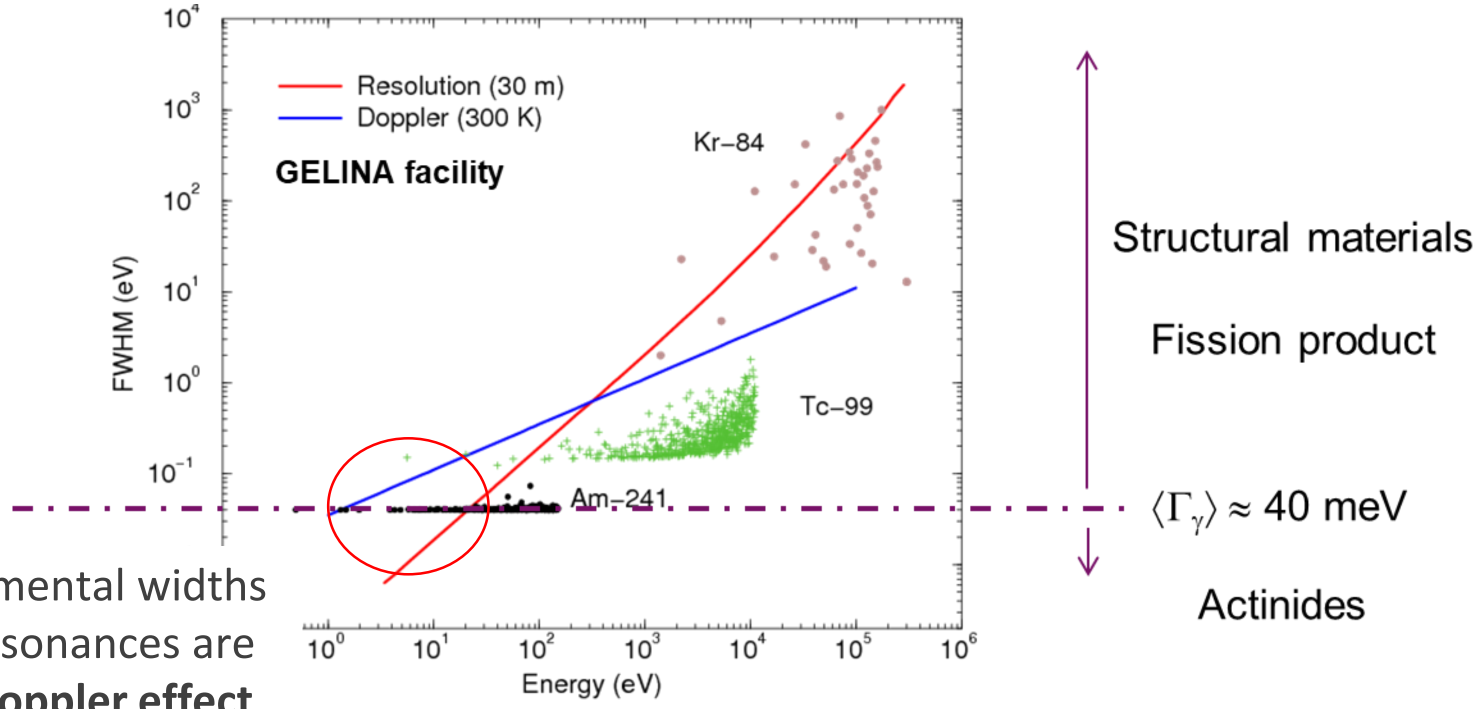
- Resolution broadening
- **Doppler broadening**
- Multiple scattering correction

Conclusions

Doppler effect

Energy dependence of the Full Width at Half Maximun (FWHM) of the Doppler and resolution contributions, compared to the natural widths of ^{241}Am+n, ^{99}Tc+n and ^{84}Kr+n

$$\Gamma^2_{obs} \approx \Delta_R^2 + \Delta_D^2 + \Gamma^2_{tot}$$



For actinides, experimental widths of the low energy resonances are dominated by the **Doppler effect**

Free Gas Model with T_{eff}

The Doppler broadened cross sections are calculated by averaging the zero Kelvin cross section over the target velocity distribution. In other words, the Doppler effect can be reduced to the convolution of the unbroadenened cross-section by a **dynamic structure factor *S*** dependent on the incident neutron energy E, outgoing energy E' and temperature T:

$$\sigma_x(E,T) = \int_0^\infty \sigma_x(E',0)S(E,E',T)\mathrm{d}E'$$

In the framework of the **Free Gas Model (FGM)**, velocities of the target nuclei follow a Maxwell Boltzmann distribution, that lead to :

$$S(E,E',T) = \frac{1}{\Delta_\mathrm{D}\sqrt{\pi}}\sqrt{\frac{E'}{E}}\left[e^{-\frac{4}{\Delta_\mathrm{D}^2}\left(E-\sqrt{EE'}\right)^2} - e^{-\frac{4}{\Delta_\mathrm{D}^2}\left(E+\sqrt{EE'}\right)^2}\right]\mathrm{d}E'$$

The Doppler width is given by

$$\Delta_\mathrm{D} = \sqrt{\frac{4mk_\mathrm{B}T}{M}E}$$

Lamb suggests replacing the thermodynamic temperature T with an **effective temperature T_{eff}** to account for crystal lattice effects. The effective temperature can be estimated via the Debye temperature or the PDOS as follow:

$$T_{eff} \simeq \frac{3}{8}\theta_D \coth\left(\frac{3}{8}\frac{\theta_D}{T}\right) \qquad T_\mathrm{eff} = \frac{\hbar}{2k_\mathrm{B}}\int_0^\infty \omega\rho(\omega)\coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)\mathrm{d}\omega$$

Crystal Lattice Model with phonon density of state

Double-differential neutron scattering cross section using a modified version of the Courcelle model

4PCF model

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{1}{4\pi}\frac{k_f}{k_i} S(\Delta\vec{k},\omega)\sigma^{T^*}(E^*)$$

In the case of CLM with phonon expansion **p ≥ 1**

Angle integrated neutron scattering cross section using the α' model of Aitor Bengoechea (PhD Thesis)

S(α,β) calculated with
- Phonon expansion model (CLM-LEAPR)
- Short Time Approximation (SCT)
- Free Gas Model with T_{eff} (FGM)

Equivalent to DBRC !!!

α' model

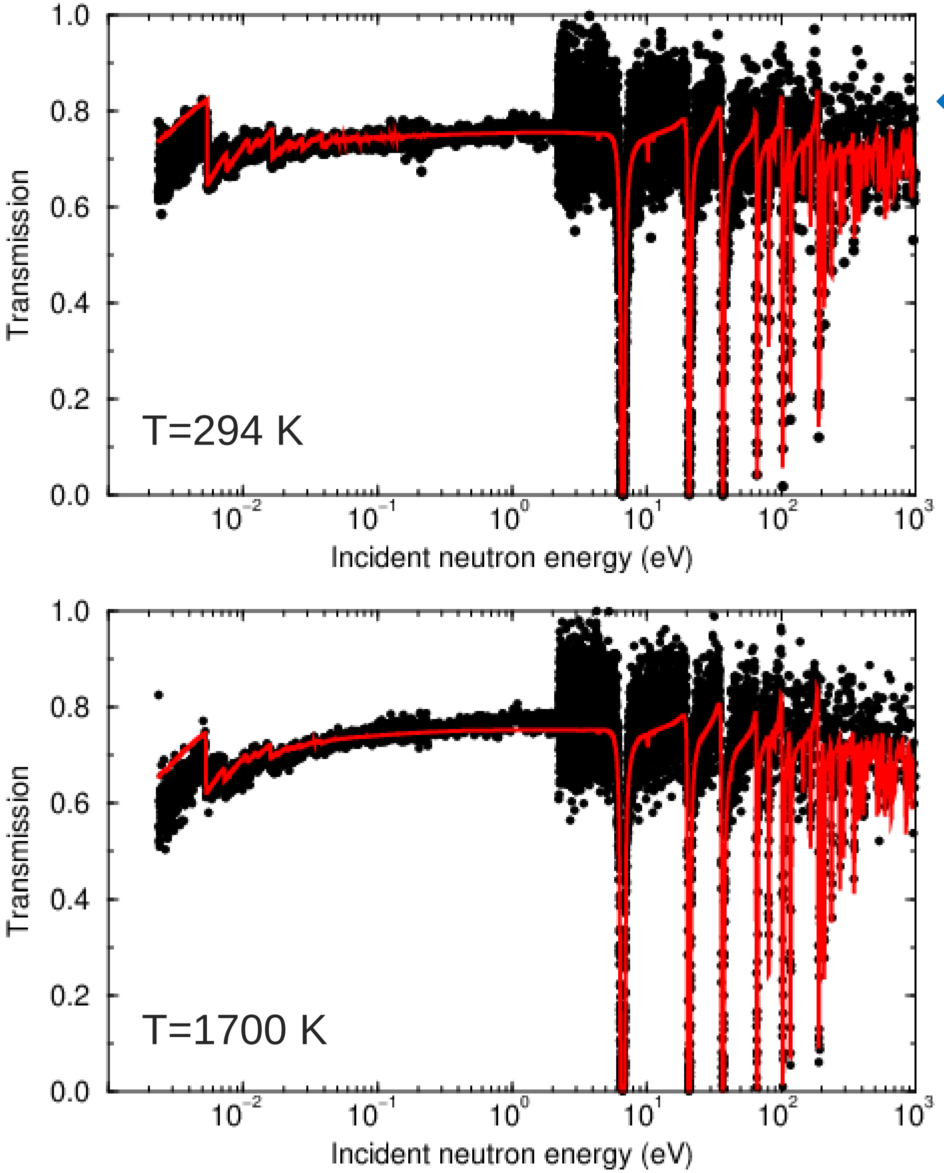
$$\sigma^T(E) = \frac{1}{2k_B T}\int\int\sqrt{\frac{E'}{E}}\, S(\alpha',\beta)\sigma_0(E' + \alpha' k_B T)dE'd\mu$$

In the case of CLM with phonon expansion **p ≥ 0**

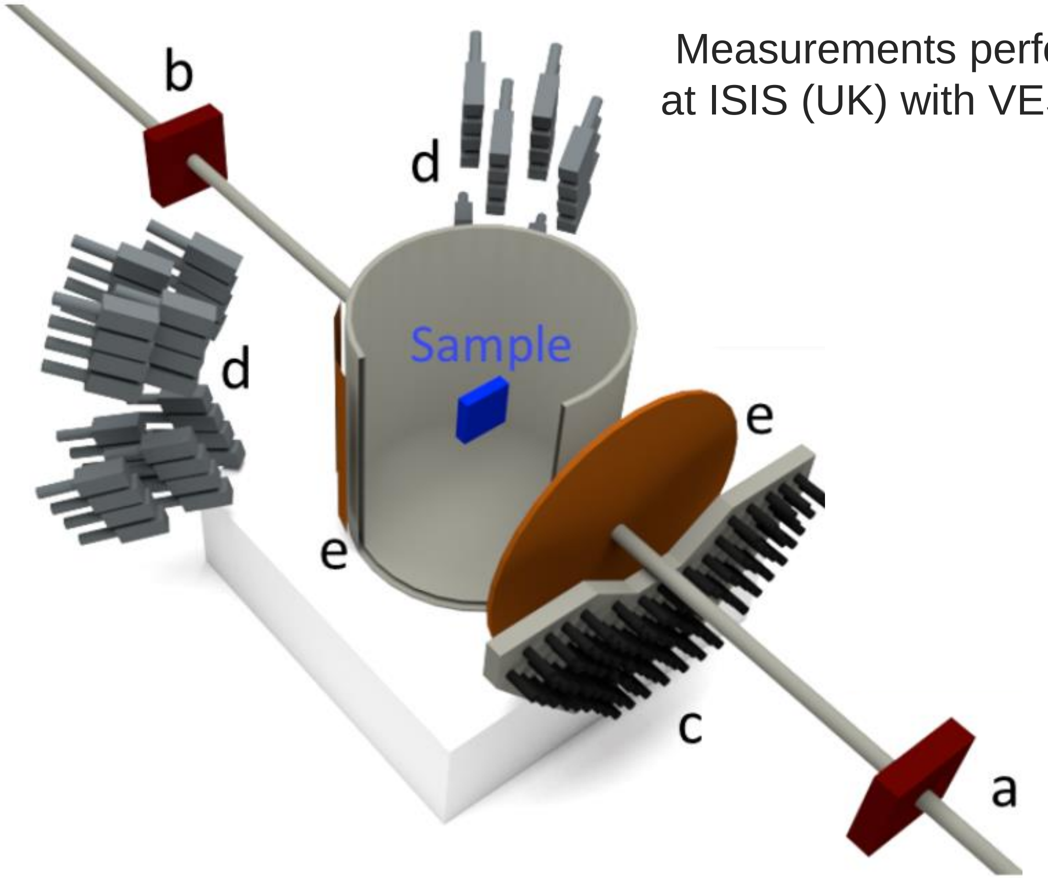
Average momentum transfer α'

$$\alpha' = \alpha_{cap} + \frac{E' - 2\mu\sqrt{EE'}}{Ak_B T}\cdot e^{\frac{\lambda_S T_{eff}}{T}} \text{ with } \boldsymbol{\alpha_{cap}} = \frac{\boldsymbol{E}}{\boldsymbol{Ak_bT}}$$

Crystal Lattice Model with phonon density of state



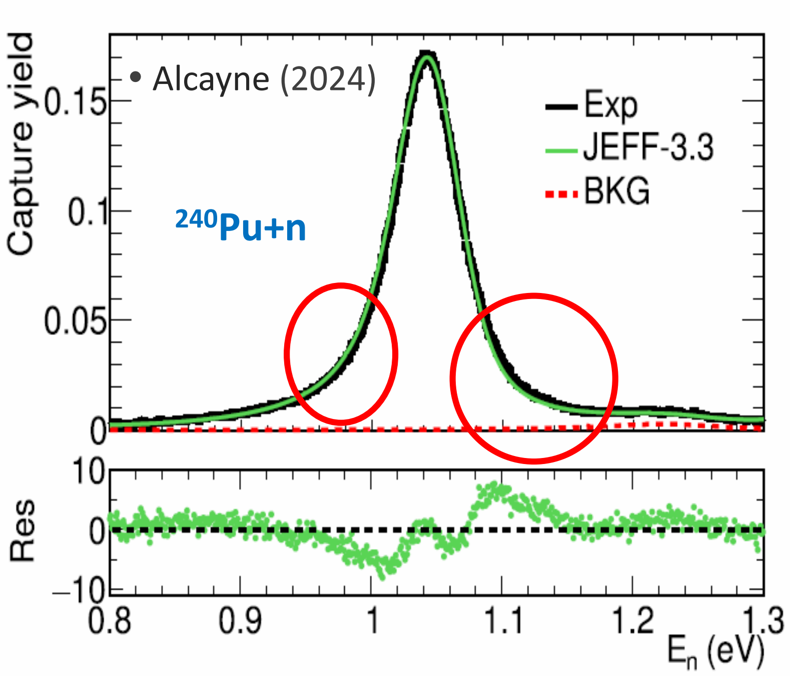
The objective of the **4PCF model** is to unified the Doppler broadening formalisms between the thermal and resonance ranges of the neutron cross sections in order to use S(α,β) model over the entire neutron energy range

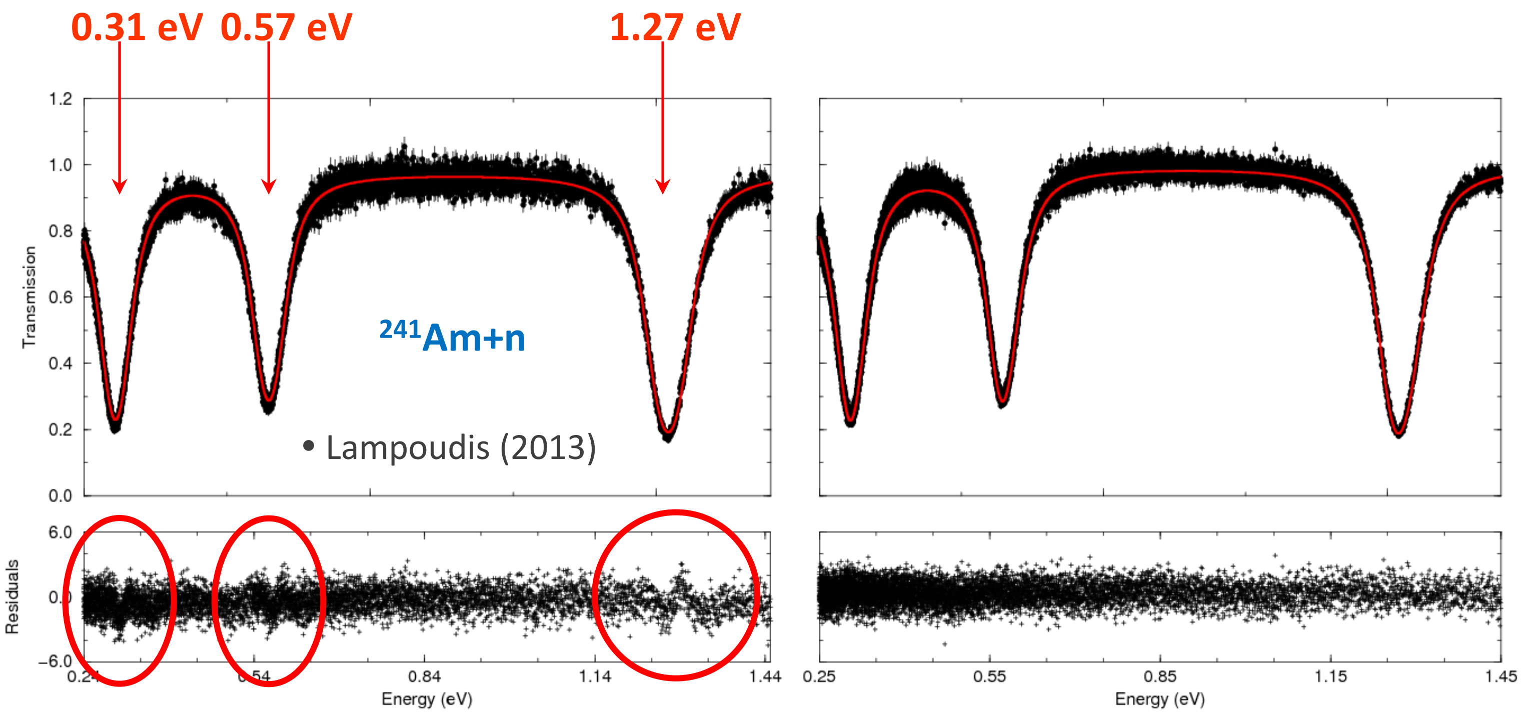


Measurements performed at ISIS (UK) with VESUVIO

Doppler broadening

The impact of the cubic structure (Fm3m symmetry) of PuO_2 and AmO_2 can be observed at low energy even at room temperature



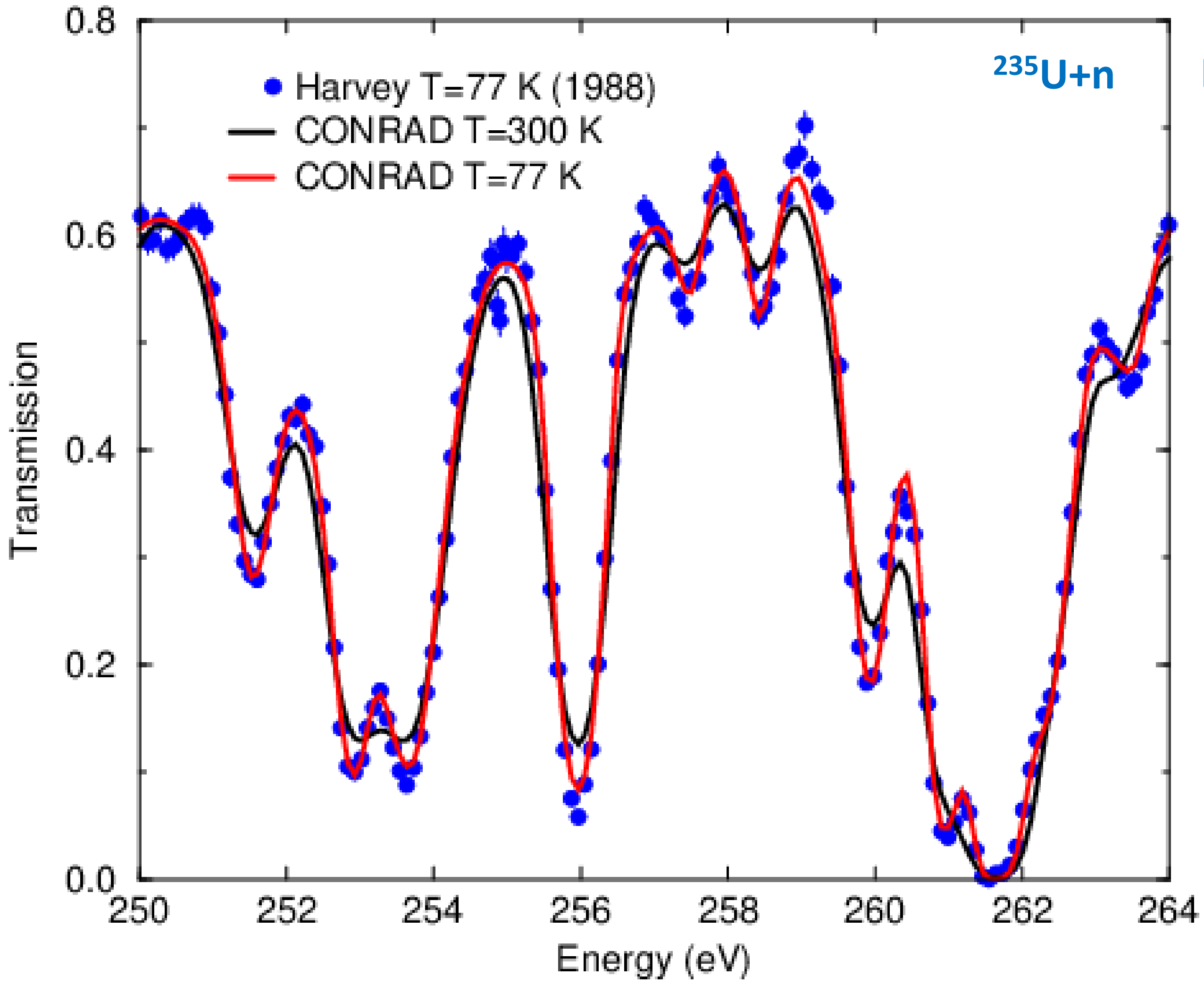


Free Gas Model (FGM)
with an effective temperature

Cristal Latice Model (CLM)
with a phonon density of states

⇒ Warning ! For actinides, the fitting procedure will accommodate values of the **radiative widths** in order to compensate deficiencies of the Doppler model

Doppler broadening



$^{235}U+n$

High-resolution measurements ⇒ low temperature

- ❑ reduce the contribution of the Doppler effect
- ❑ allow identifying multiplets of resonances
- ❑ useful for extending the resolved resonance range

Evaluation of the Resolved Resonance Range

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Mix nuclear models and experimental corrections

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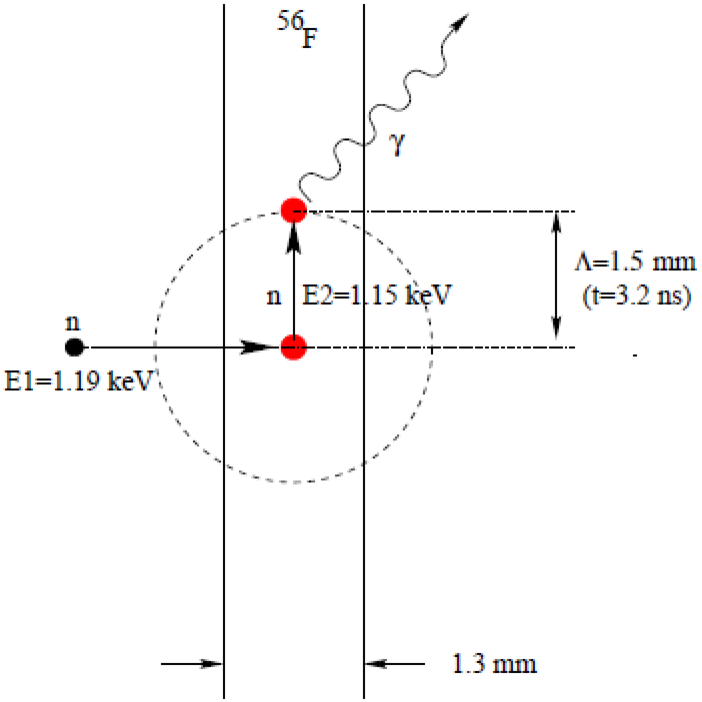
Conclusions

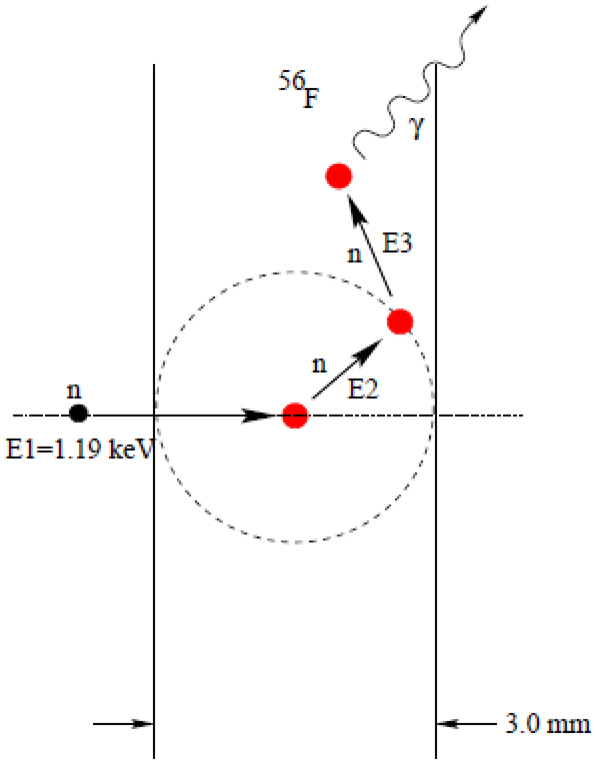
Multiple scattering correction

Thin sample



single 90° scattering

Thick sample

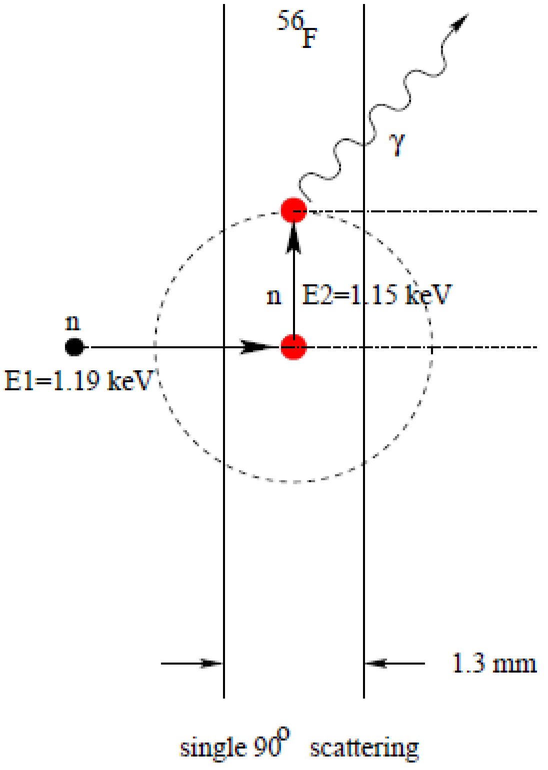


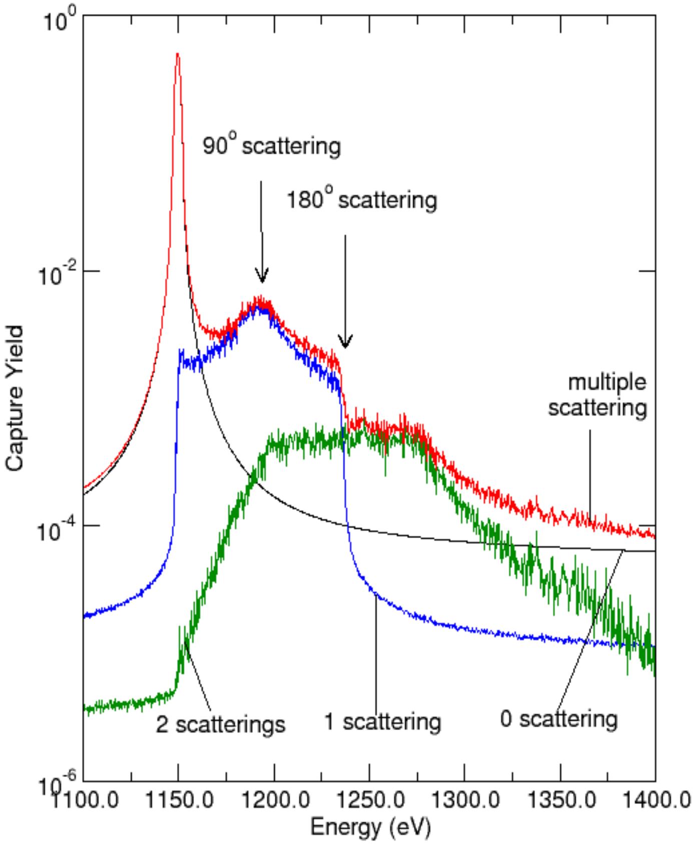
multiple scattering

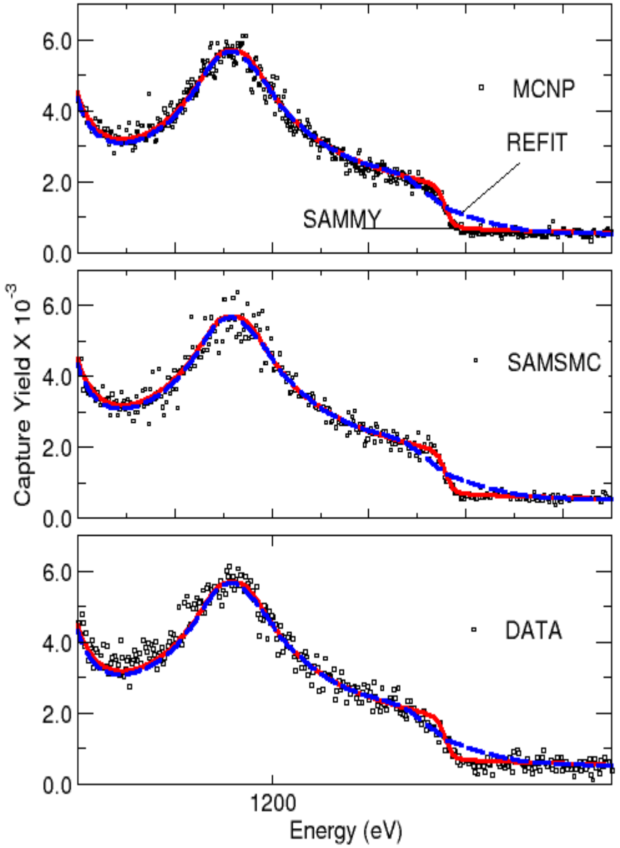
Analytical or Monte-Carlo models are available in the Resonance Shape Analysis codes

Multiple scattering correction in ^{56}Fe sample

Thin sample

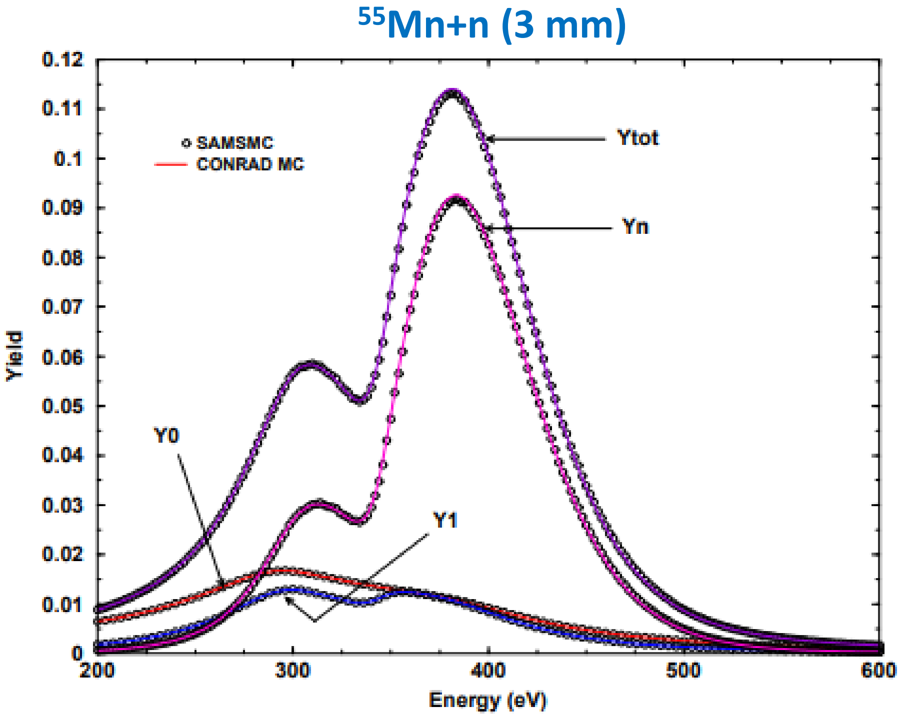


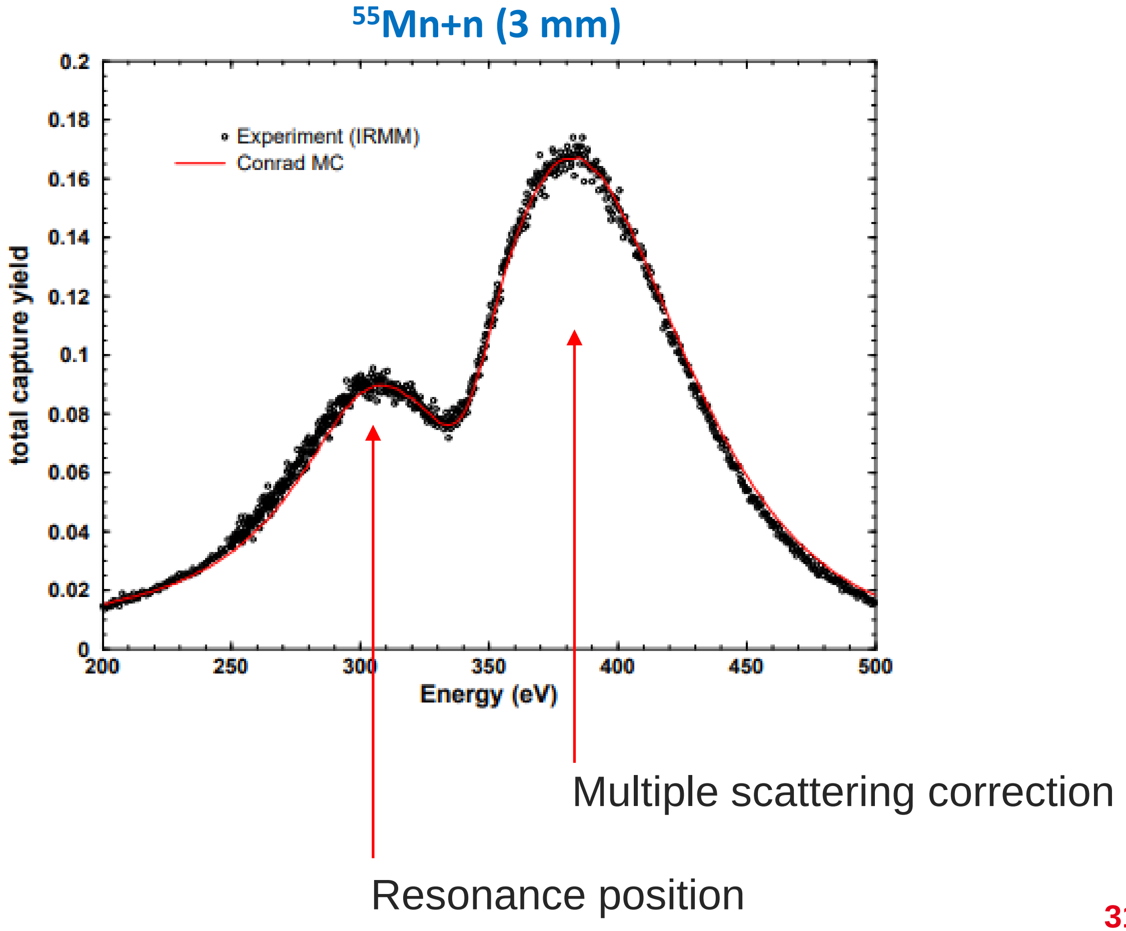




Multiple scattering correction in ^{55}Mn sample

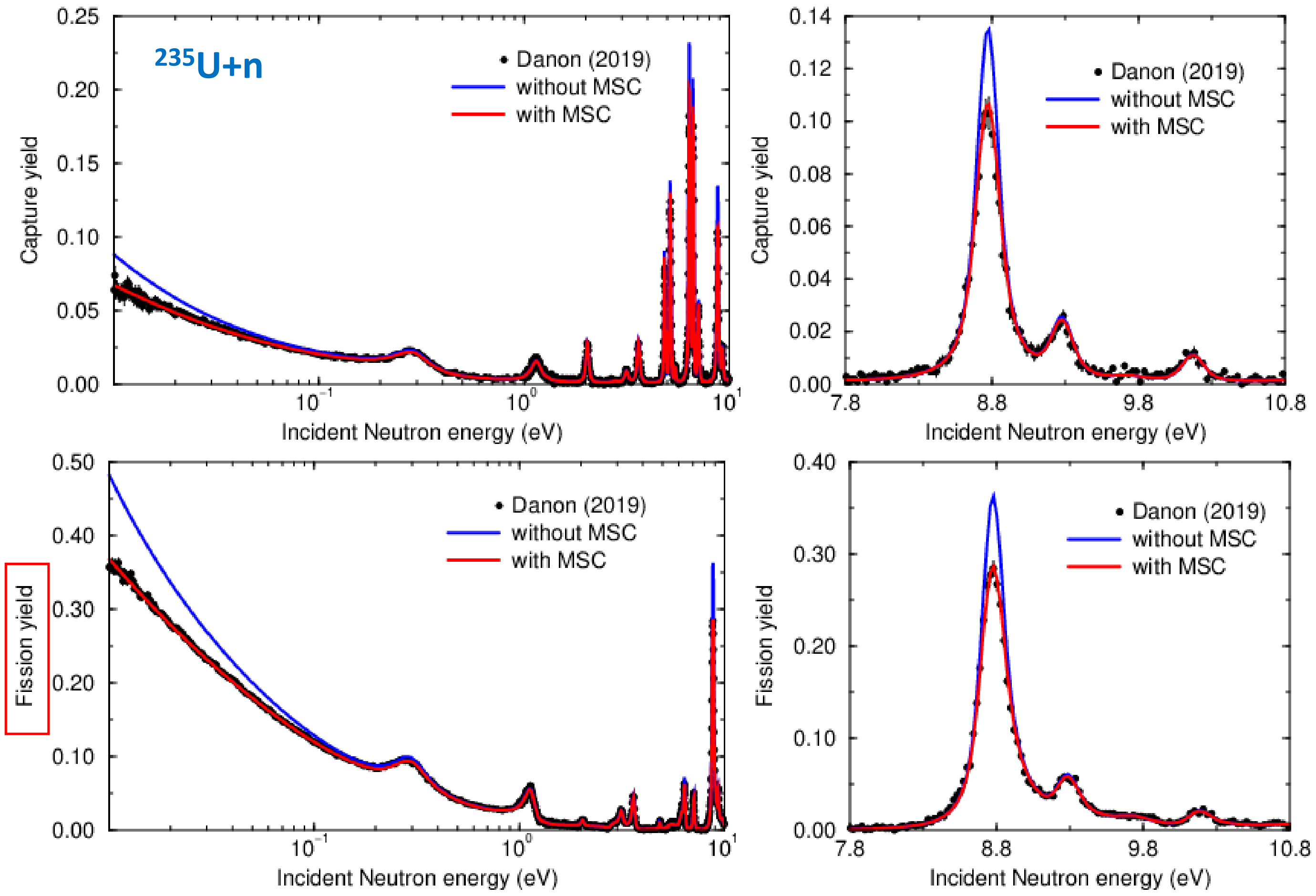
Example of CONRAD calculations for a thin disc of Mn55 (3 mm thick). The huge multiple scattering contribution Yn due to the Mn55 resonance can only be reproduced by Monte-Carlo.





Multiple scattering correction in ^{235}U sample

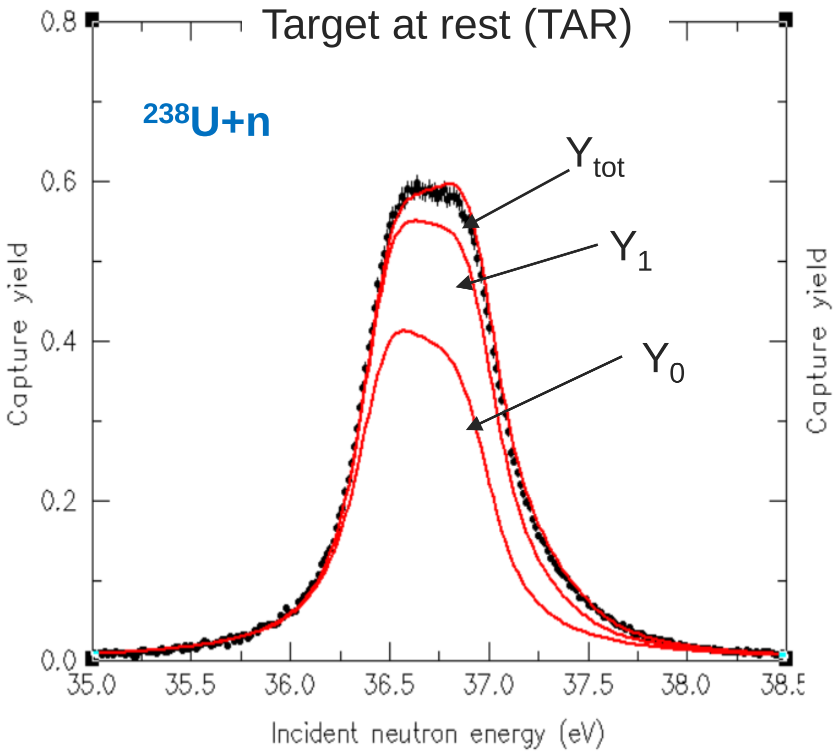
Sizeable impact of the correction in the thermal energy range and in the energy range [7.8-11 eV] of the recommended fission integral for the U235 capture and fission yields measured at the RPI facility

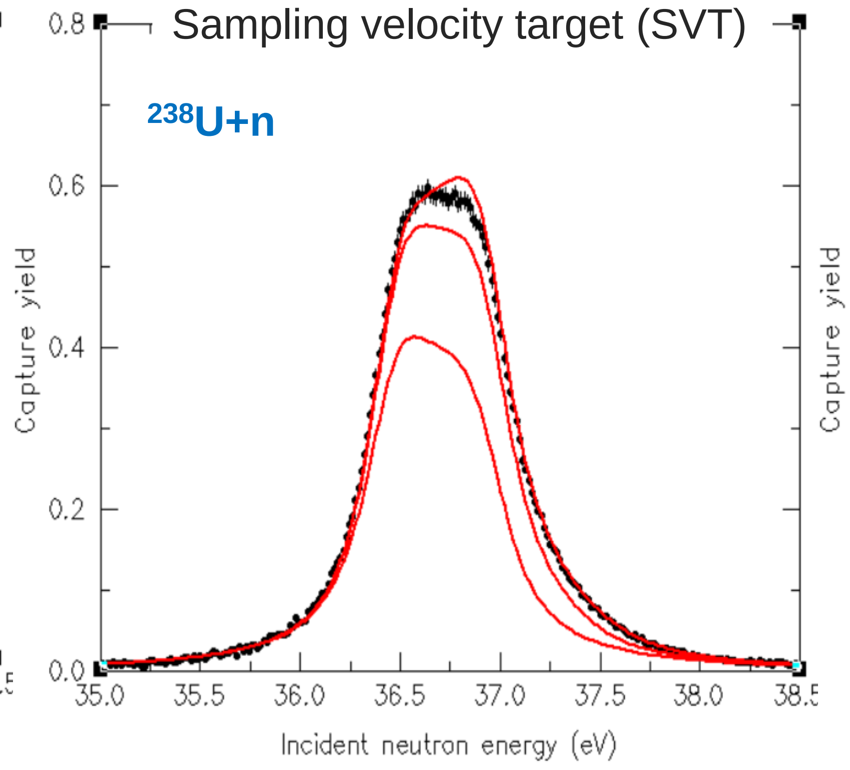


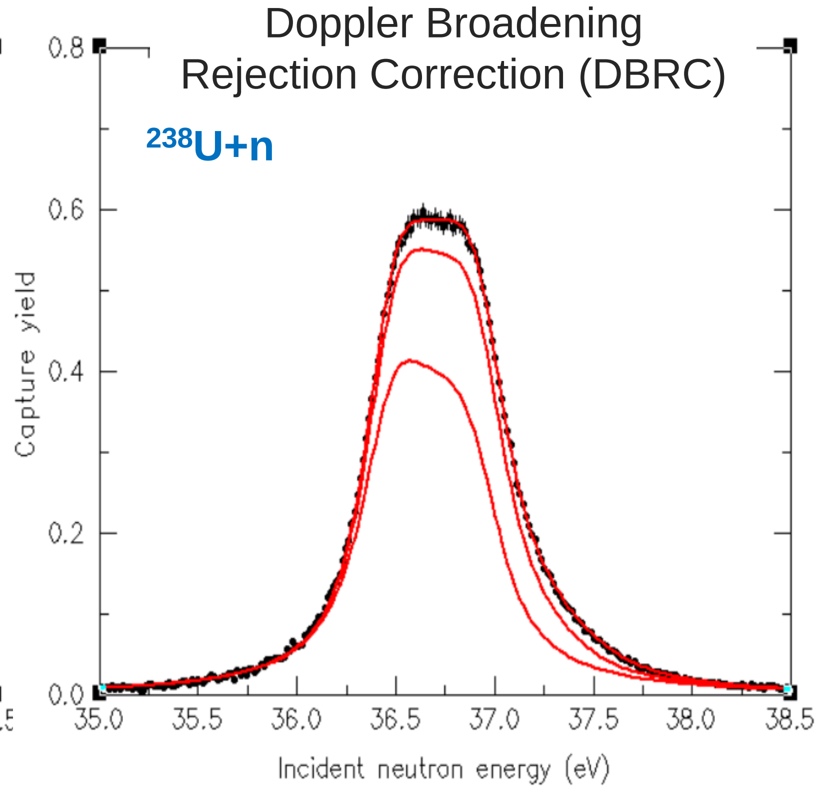
Monte-Carlo model is recommended for a precise description of the multiple scattering and an accurate determination of the resonance parameters and normalization

Multiple scattering correction in ^{238}U sample

Monte-Carlo model in the CONRAD code can account for target velocity and up-scattering effects (DBRC model) due to temperature



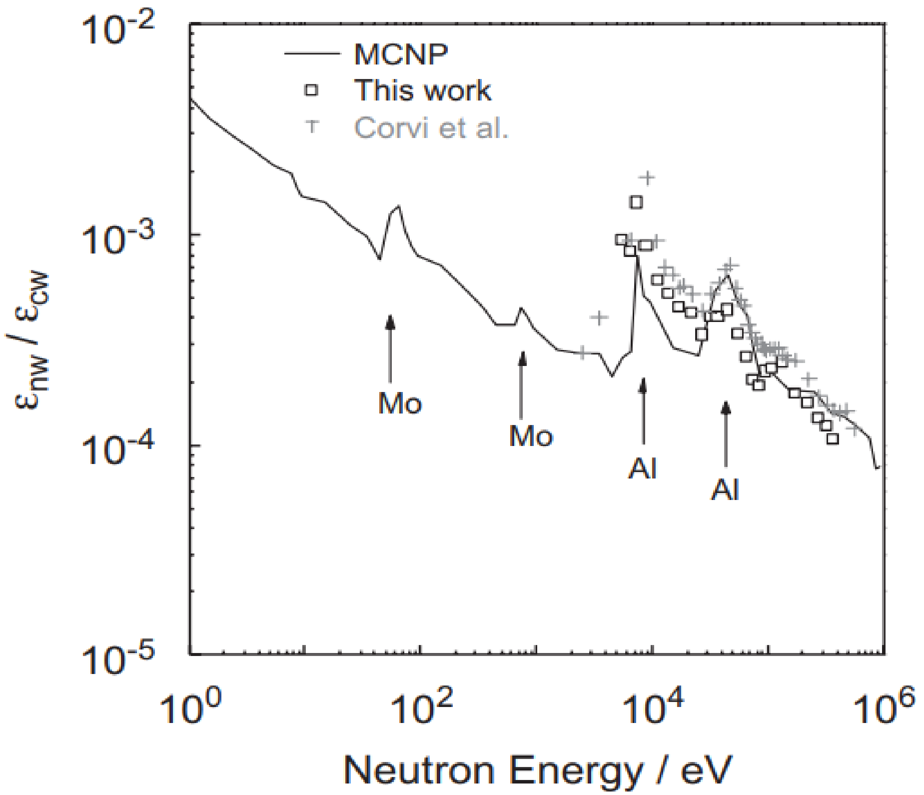


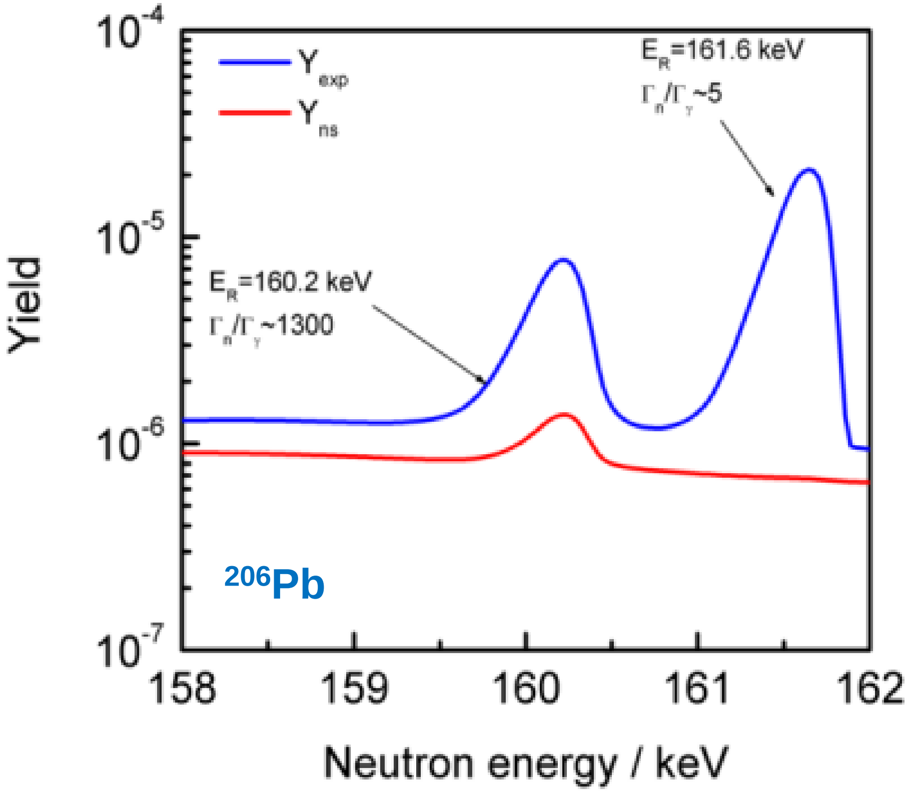


⇒ Warning ! the fitting procedure will accommodate values of the resonance parameters to compensate possible deficiencies of the multiple scattering correction

Neutron sensitivity correction

$$Y_{th}(E) = (1+\alpha(E))\,(1-T_{th}(E))\,\frac{\sum_i n_i\,\sigma_{\gamma,i}(E)}{\sum_i n_i\,\sigma_{T,i}(E)}\,\varepsilon_{\rm cw}(E_{\rm n}) + \boxed{\varepsilon_{\rm nw}(E_{\rm n})\,Y_{\rm n}(E_{\rm n})}$$





⇒ Important for “scattering resonances”

Evaluation of the Resolved Resonance Range

From nuclear structure to macroscopic scale

Mix nuclear models and experimental corrections

- Resolution broadening
- Doppler broadening
- Multiple scattering correction

Conclusions

Conclusions

R-Matrix codes for resonance analysis

- Resonance Shape Analysis codes REFIT, SAMMY and CONRAD

Need for a unified Doppler model to cover the thermal and resonance ranges

- Free Gas Model with an effective temperature is an approximation not valid at low neutron energy

Multiple scattering correction

- Monte-Carlo model is recommended to account for target velocity and up-scattering effects (DBRC)

Simultaneous data analysis

- Use various data sets measured with samples of different thicknesses to establish a consistent set of parameters