



Neutron-induced reaction cross section studies at GELINA

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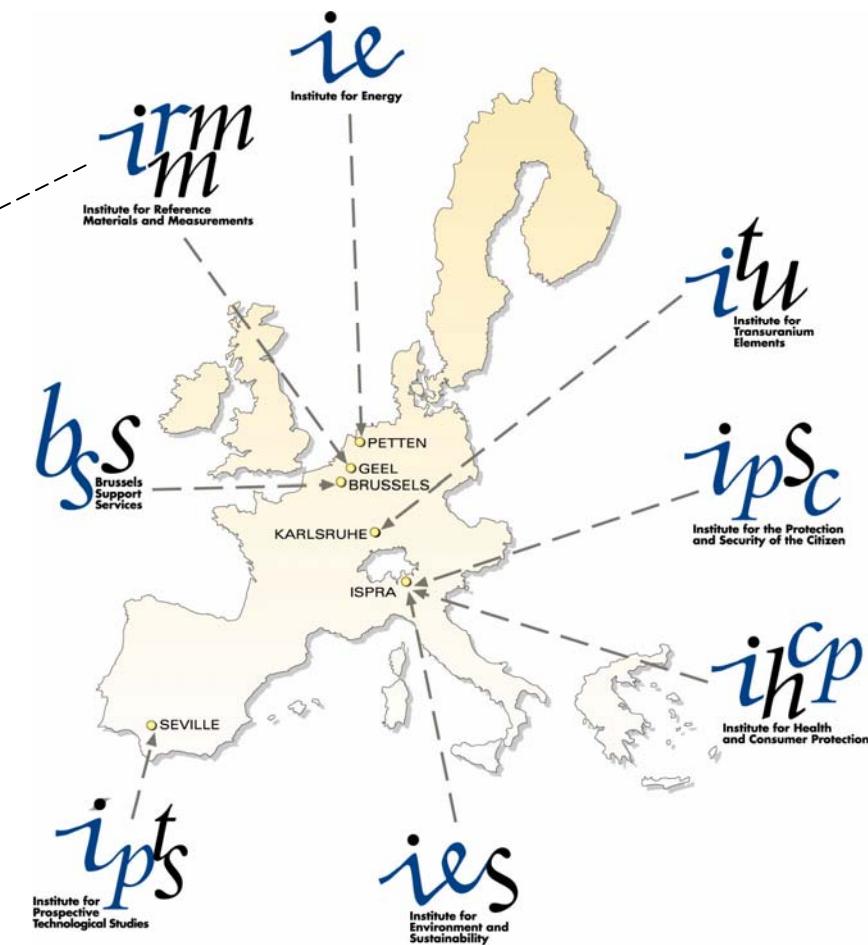
Institute for Reference Materials and Measurements (IRMM), Geel, Belgium

<http://www.irmm.jrc.be>
<http://www.jrc.ec.europa.eu>

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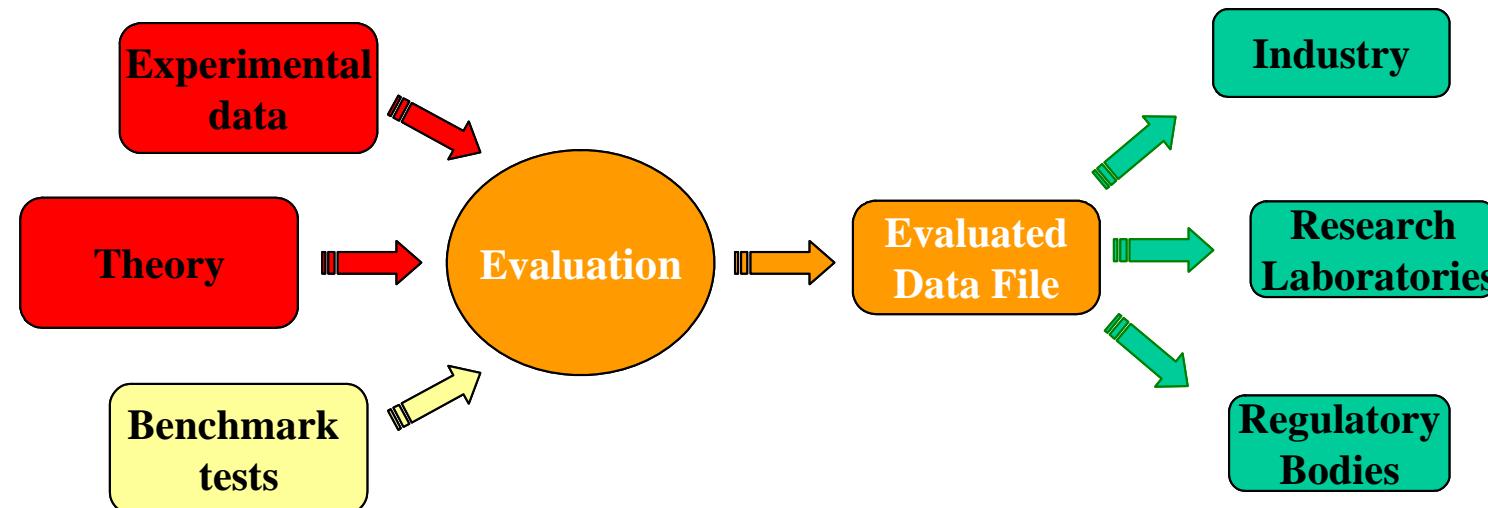
**Institute for Reference
Materials and Measurements**



The Mission of the Neutron Physics Unit

Provide European safety authorities and industry with neutron reaction data for:

- The *safety assessment* of nuclear installations and the nuclear fuel cycle
- The *feasibility study and development* of waste transmutation facilities and innovative reactor systems



⇒ GELINA and VdG

7 MV Van de Graaff accelerator

- Single-user facility
- 6 different beam lines
- 100h/w
- 0.1 - 10 & 13-21 MeV
- Li(p,n), T(p,n), D(d,n), T(d,n)

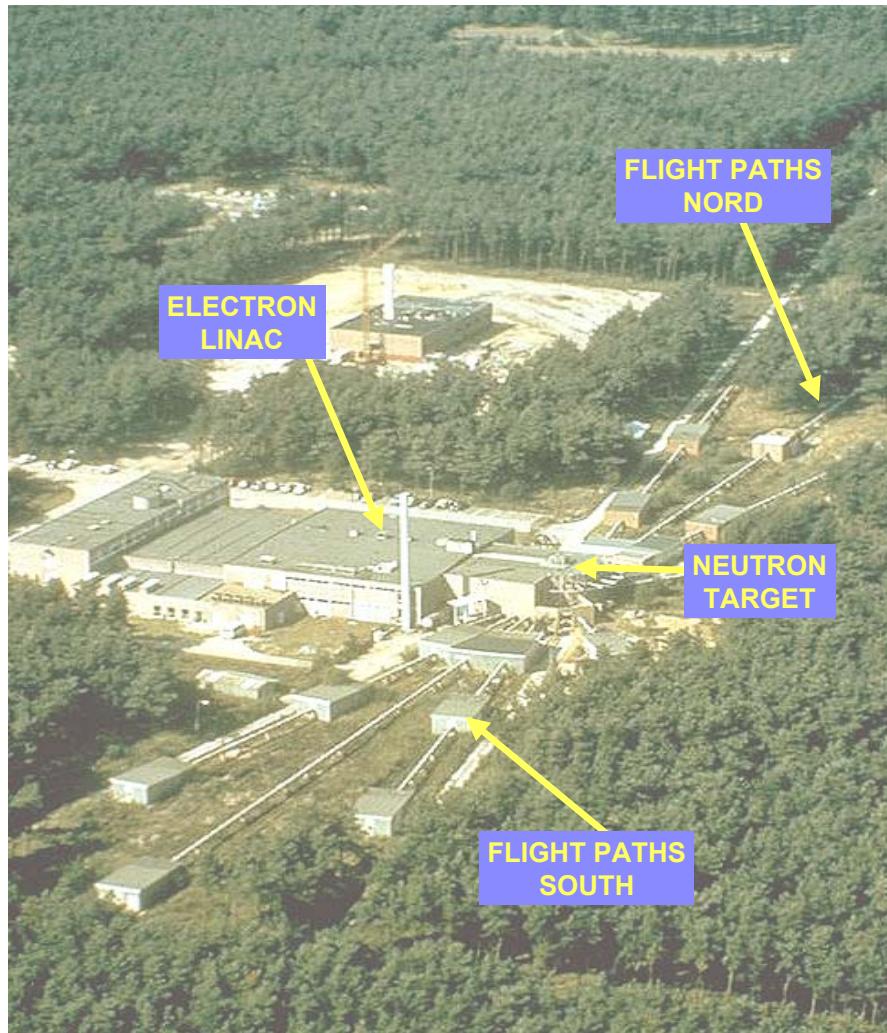


Measurement types

- Fission
- Activation measurements
 $n \rightarrow$ light charged particles
- Flux (BIPM)
- Inelastic scattering
- Calibration of detectors

- **GELINA – TOF Facility**
- **Measurement stations (capabilities)**
- **Research Activities**
- **Cross Section Measurements for Applications**

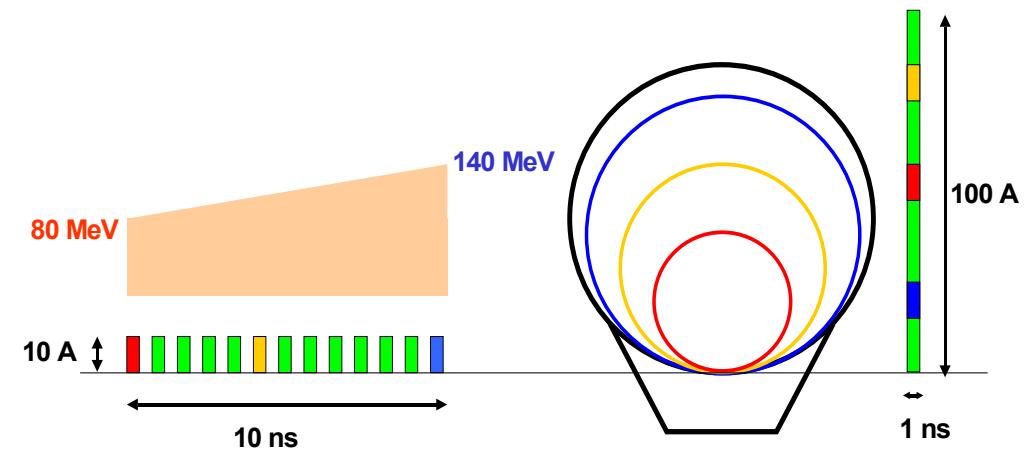
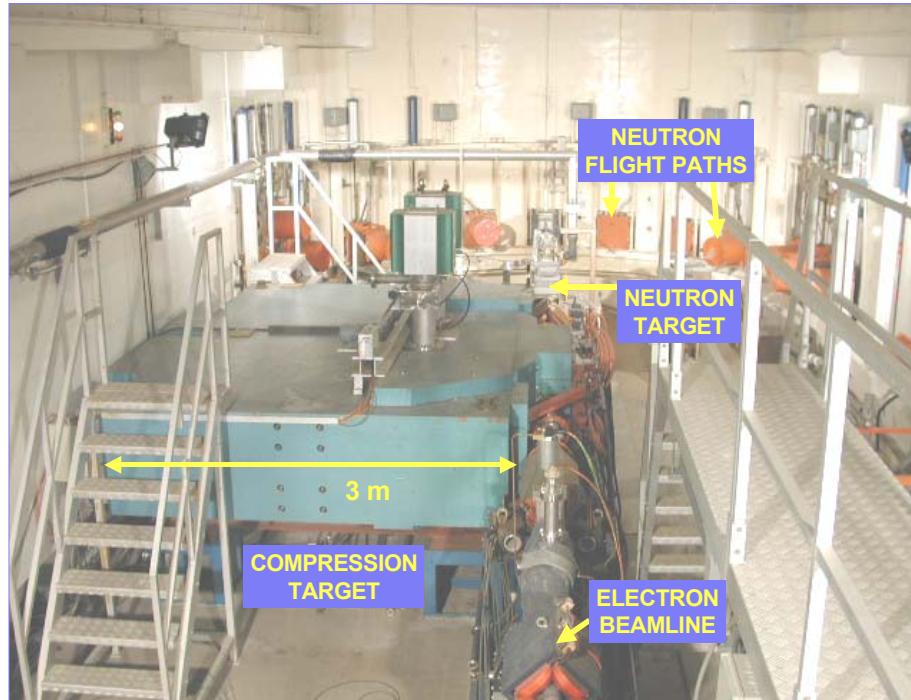
GELINA



- Time-of-flight facility
- Pulsed white neutron source
($1 \text{ meV} < E_n < 20 \text{ MeV}$)
- Multi-user facility with 10 flight paths
(10 m - 400 m)
- The measurement stations have special equipment to perform:
 - Total cross section measurements
 - Partial cross section measurements

Pulse Width	:	1ns
Frequency	:	50 Hz – 800 Hz
Average Current	:	4.7 μA – 75 μA
Neutron intensity	:	$1.6 \cdot 10^{12} \text{ n/s}$ – $2.5 \cdot 10^{13} \text{ n/s}$

Compression Magnet



$$B\rho = \frac{p}{q}; E \approx pc; q = e$$

$$\Rightarrow \rho = \frac{1}{B} \frac{E}{qc}$$

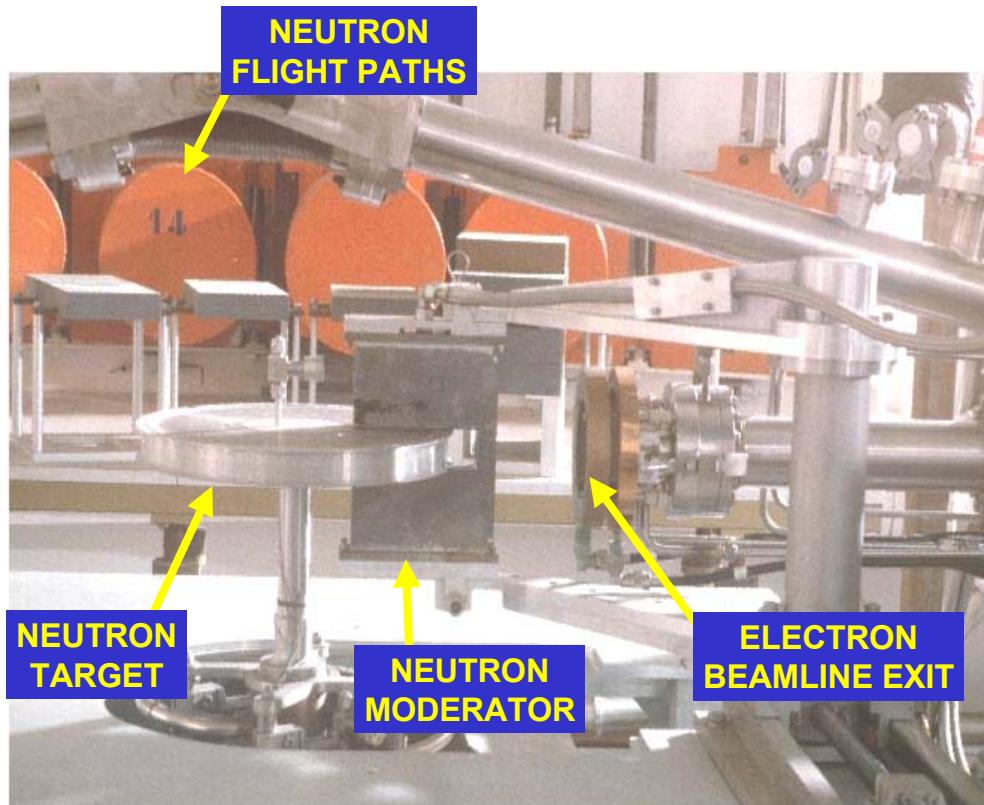
$$\Rightarrow B = \frac{2\pi}{qc^2} \frac{\Delta E}{\Delta t}$$

$$\Delta E = 60 \text{ MeV}$$

$$\Delta \tau = 10 \text{ ns}$$

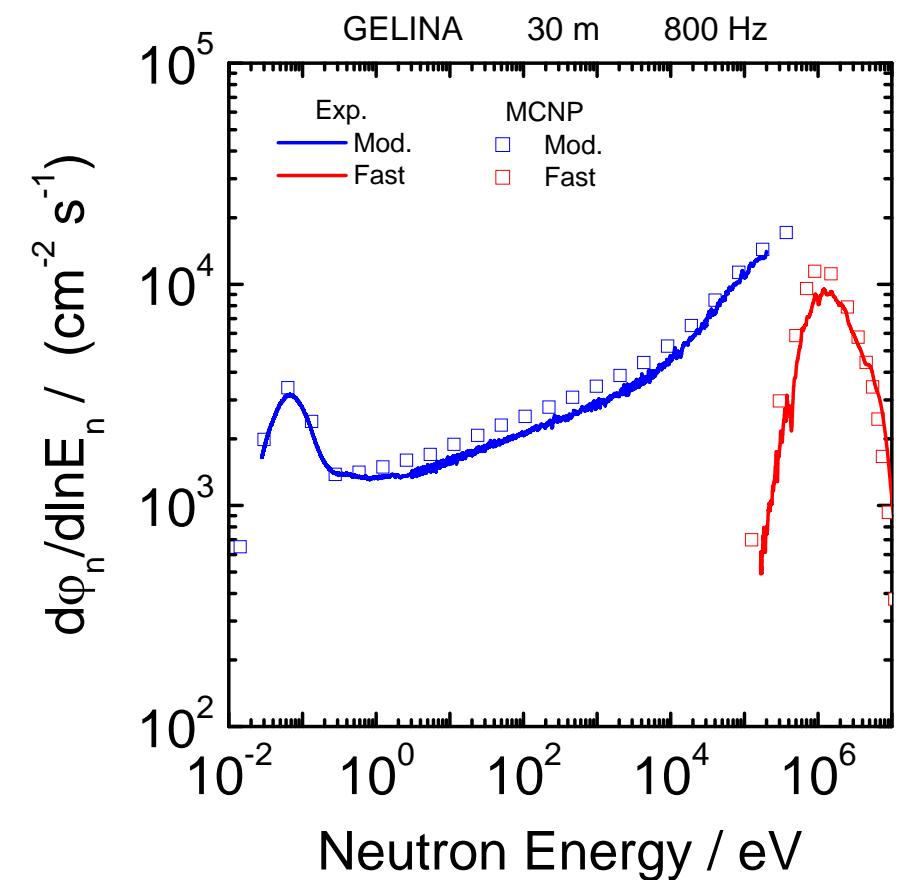
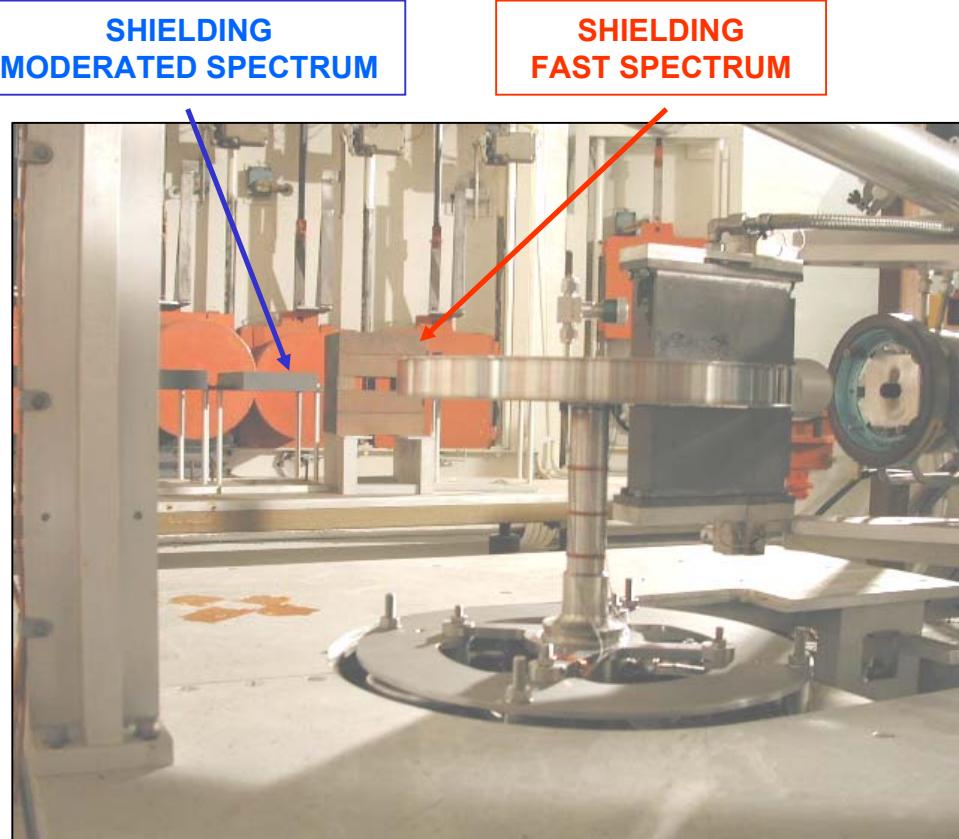
→ compressed pulse length ~ 1 ns

Neutron Production

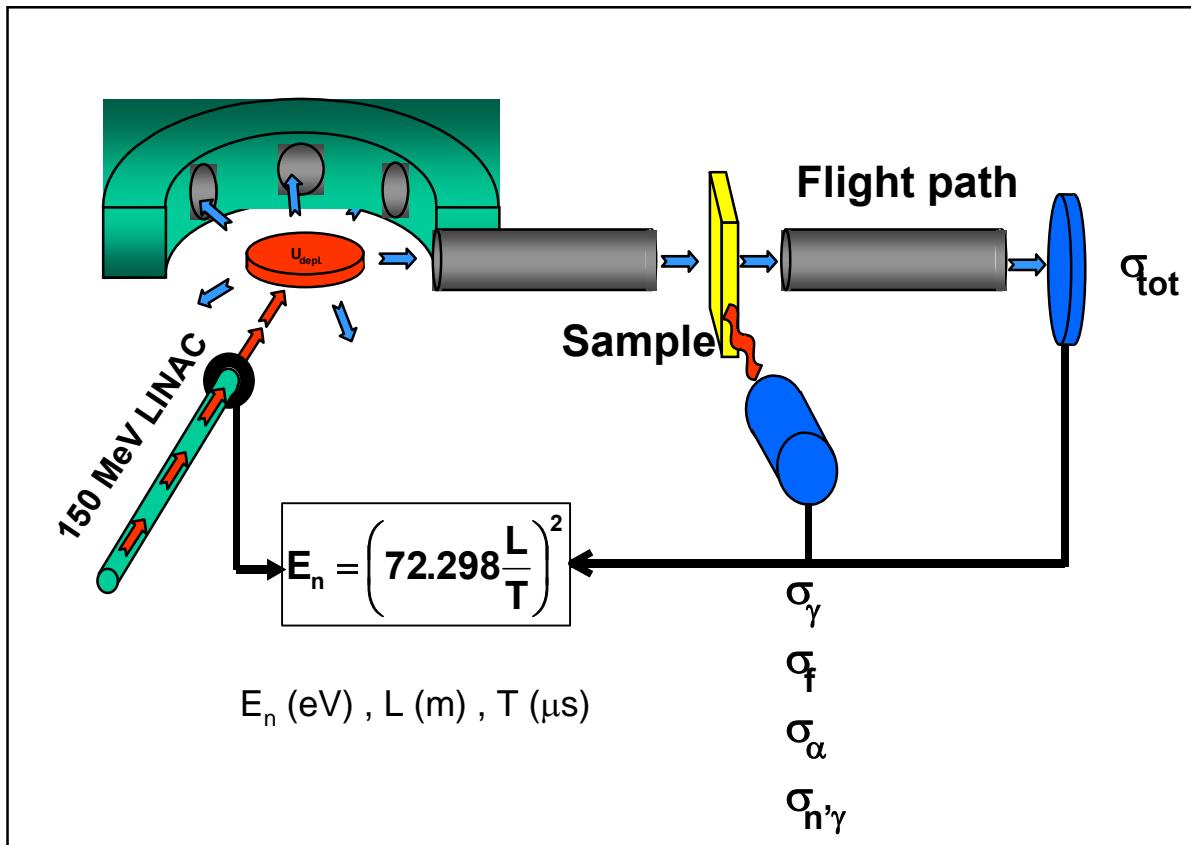


- e^- accelerated to $E_{e^-,max} \approx 140$ MeV
- (e^-, γ) Bremsstrahlung in U-target (rotating & cooled with liquied Hg)
- (γ, n) , (γ, f) in U-target
- Low energy neutrons by water moderator in Be-canning

Neutron Production



GELINA and ORELA TOF-Facilities for High Resolution Total and Partial Cross-Sections



- Neutron Flux $\Rightarrow L \downarrow$

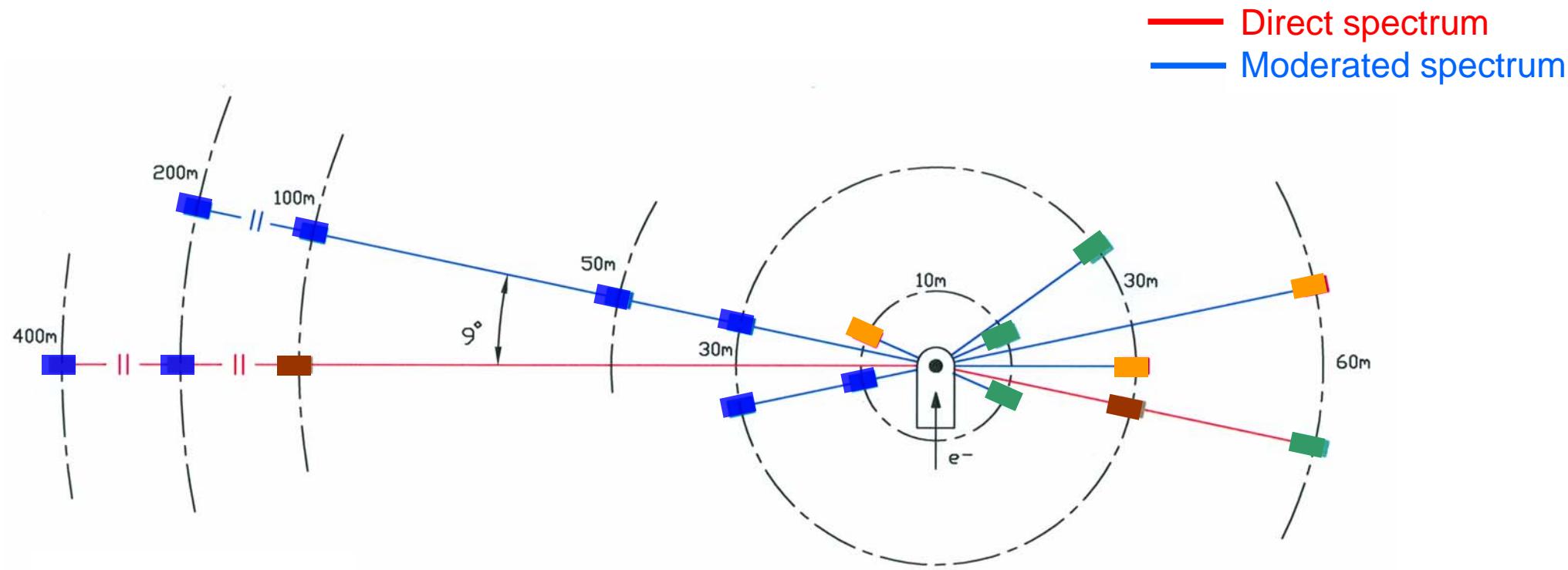
$$\varphi_n(L) \propto \frac{1}{L^2}$$

- Resolution $\Rightarrow L \nearrow$

$$\frac{\Delta E_n}{E_n} = \frac{1}{L} \sqrt{\frac{E_n \Delta T^2}{\alpha} + \Delta L^2}$$

We need to adapt the flight path length and operation conditions to the required resolution and neutron flux

Measurement Stations



orange square	(n,γ)	NIM, j.nima.2007.03.034
blue square	(n,tot)	NP A 773, 173 (2006)
green square	(n,f) and (n,cp)	NSE 156, 211 (2007)
brown square	$(n,n'\gamma)$	NP A 786, 1 (2007)

Transmission Measurements

Sample & Background Filters



Detector



Detector stations

Moderated : $L = 30 \text{ m}, 50 \text{ m}, (100 \text{ m}, 200 \text{ m})$

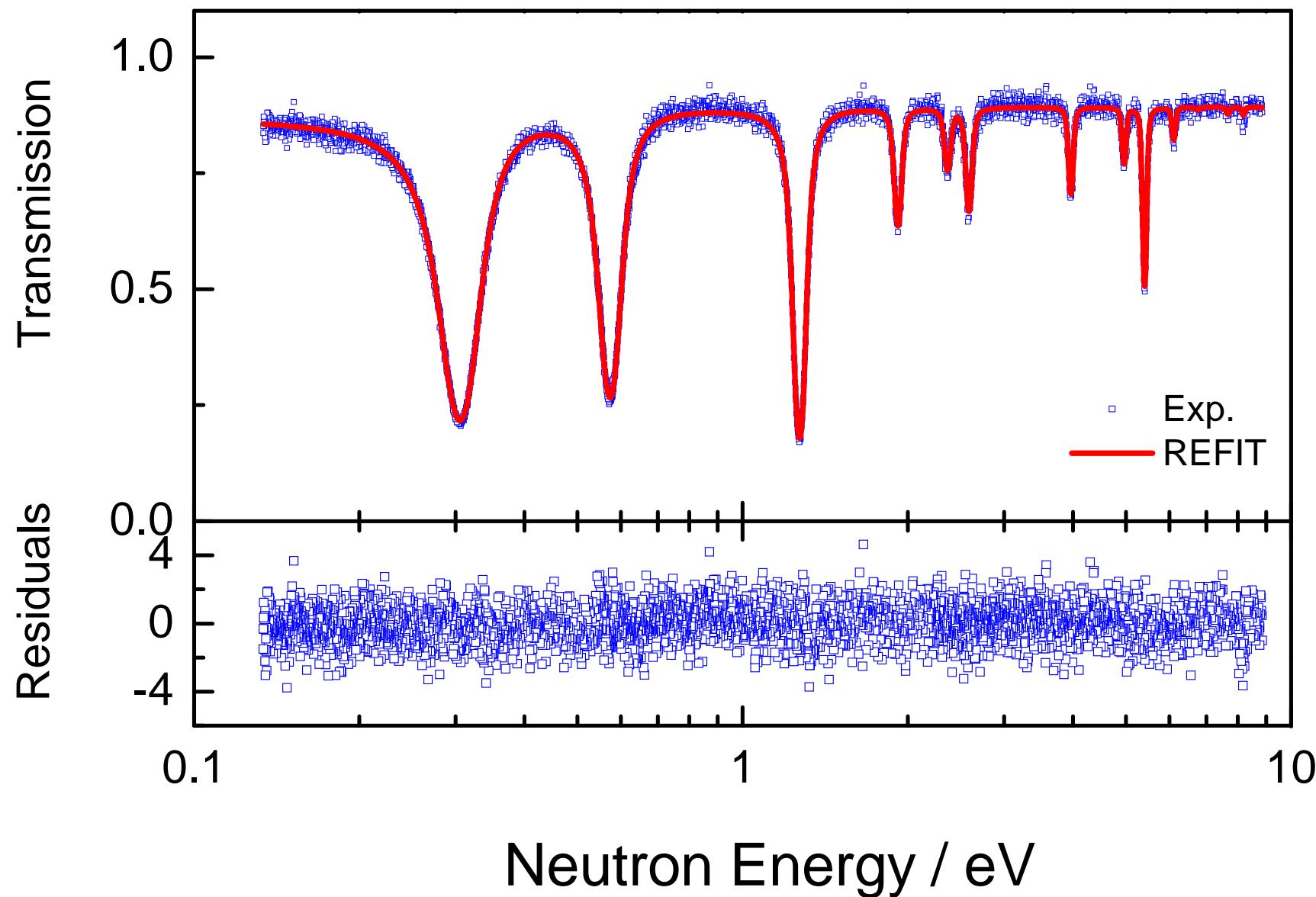
Fast : $L = 400 \text{ m}$

Low energy : ${}^6\text{Li}(n,t)\alpha$ Li-glass

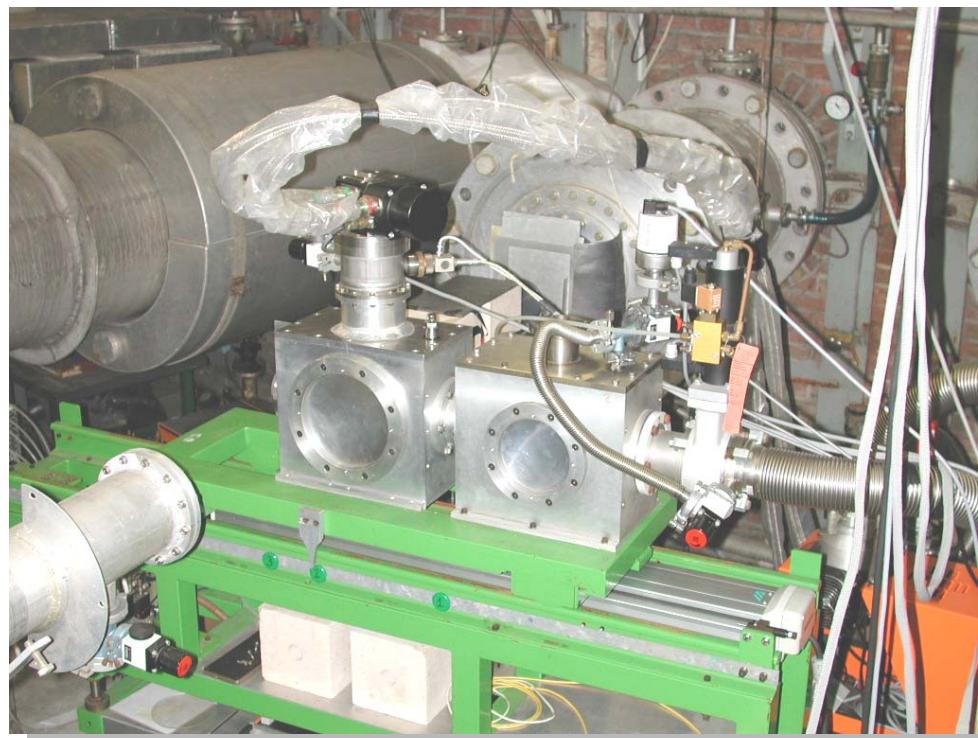
High energy : $H(n,n)H$ Plastic scintillator

$$T = \frac{C_{in}}{C_{out}} \cong e^{-n\sigma_{tot}}$$

$^{241}\text{Am}(\text{n,tot})$ at 30 m

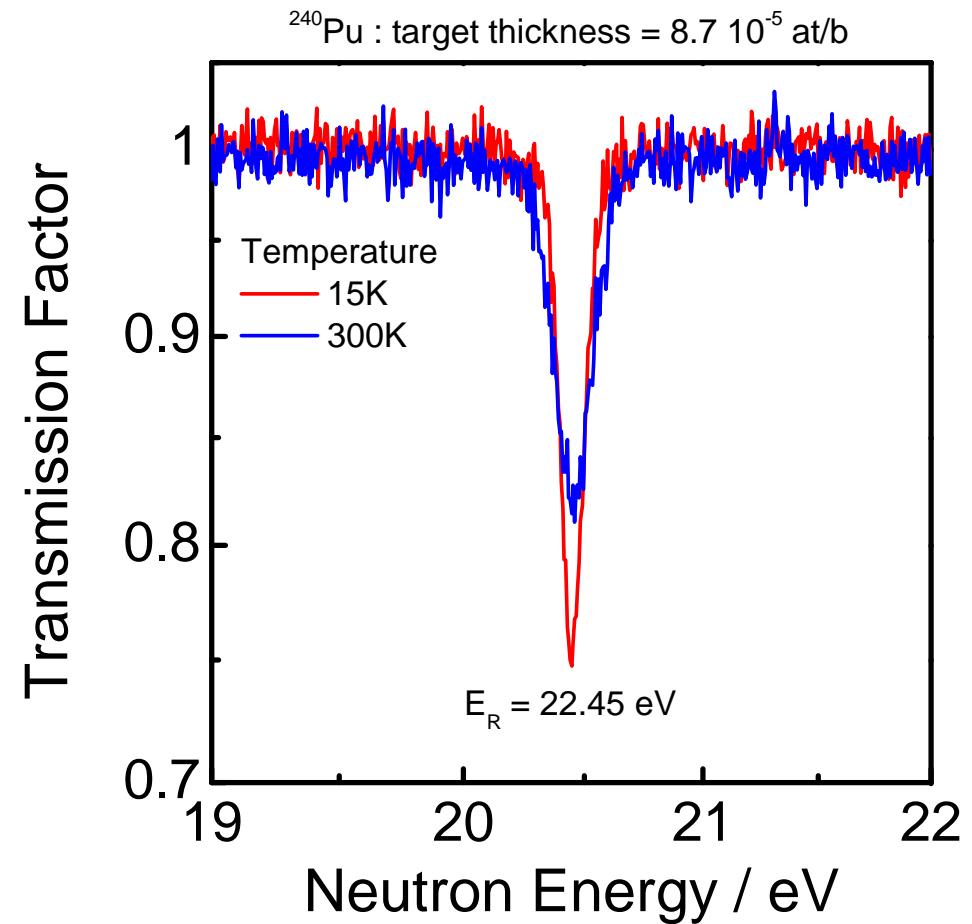


Doppler Measurements



Detector station at : L = 30m

Temperatures from 10 K - 300 K



(n,f) and (n, cp) Measurements at 10 m and 30 m

- Flux $^{10}\text{B}(\text{n},\alpha)$ or $^{235}\text{U}(\text{n},\text{f})$

$$C_\phi = \varepsilon_\phi n_\phi \sigma_\phi A_\phi \phi_\phi$$

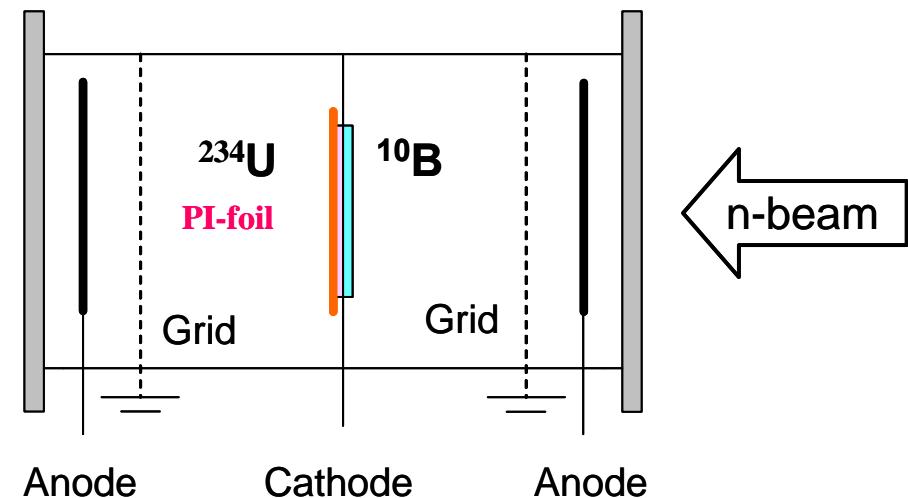
- Fission

$$C_f = \varepsilon_f n_f \sigma_f A_f \phi_f$$

+ Back to back target : $A_\phi = A_f$

+ Ionisation chamber : $\varepsilon_\phi = 100\%$

$\varepsilon_f = 100\%$

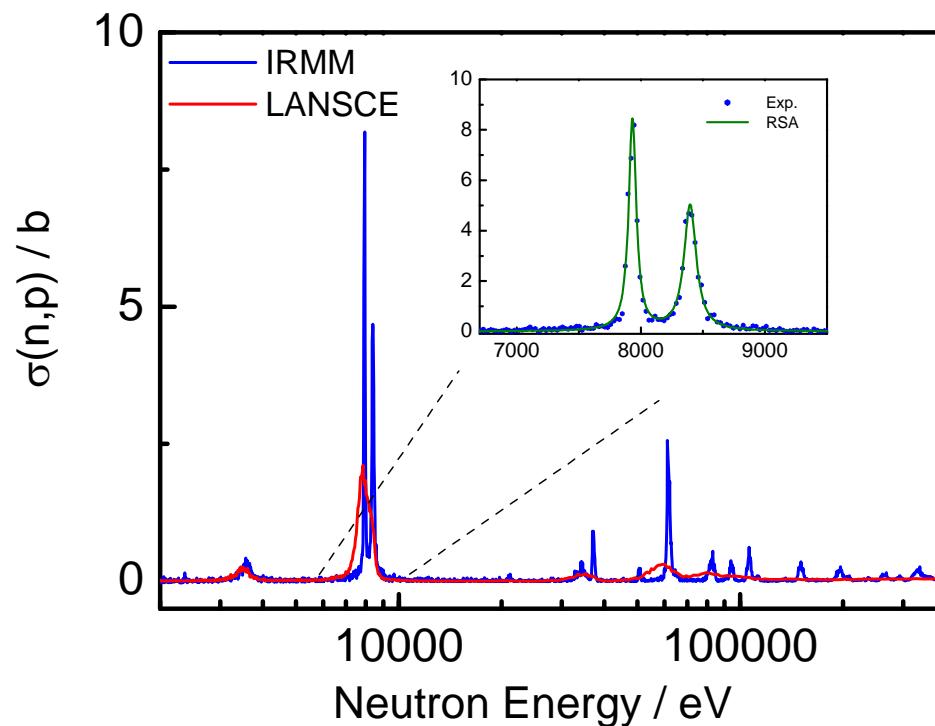


$$\Rightarrow \sigma_f = \frac{C_f}{C_\phi} \frac{n_\phi}{n_f} \sigma_\phi$$

Detector station at : L = 10 m and 30m
Moderated spectrum

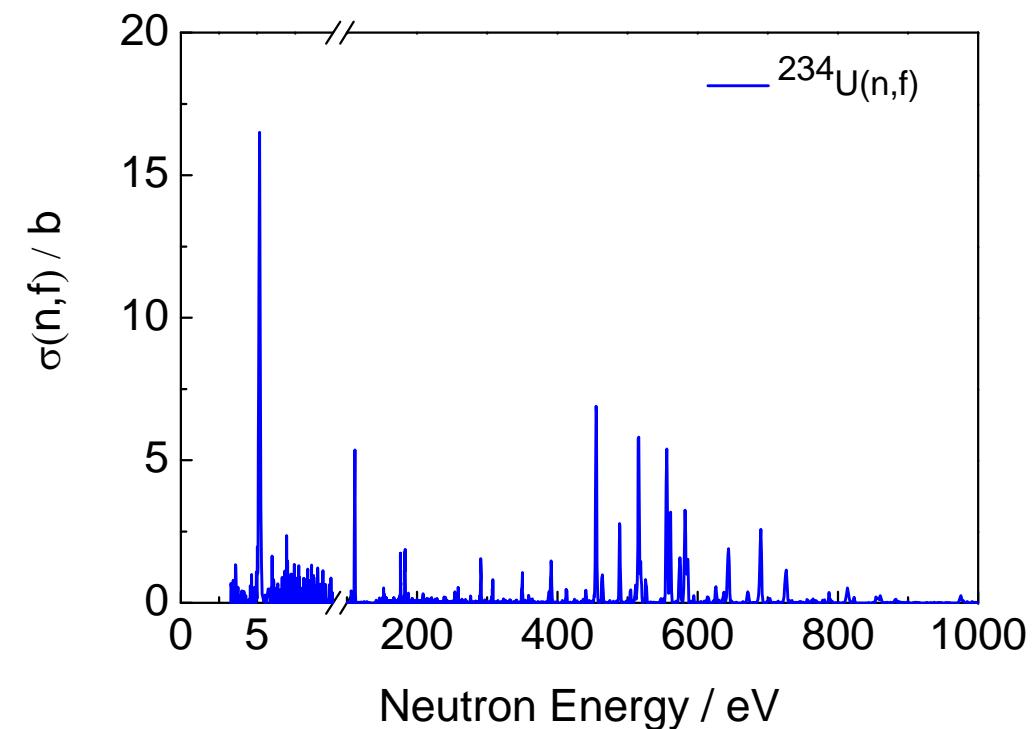
(n,f) and (n,cp) : IRMM - UGent

$^{36}\text{Cl}(\text{n},\text{p})^{36}\text{S}$



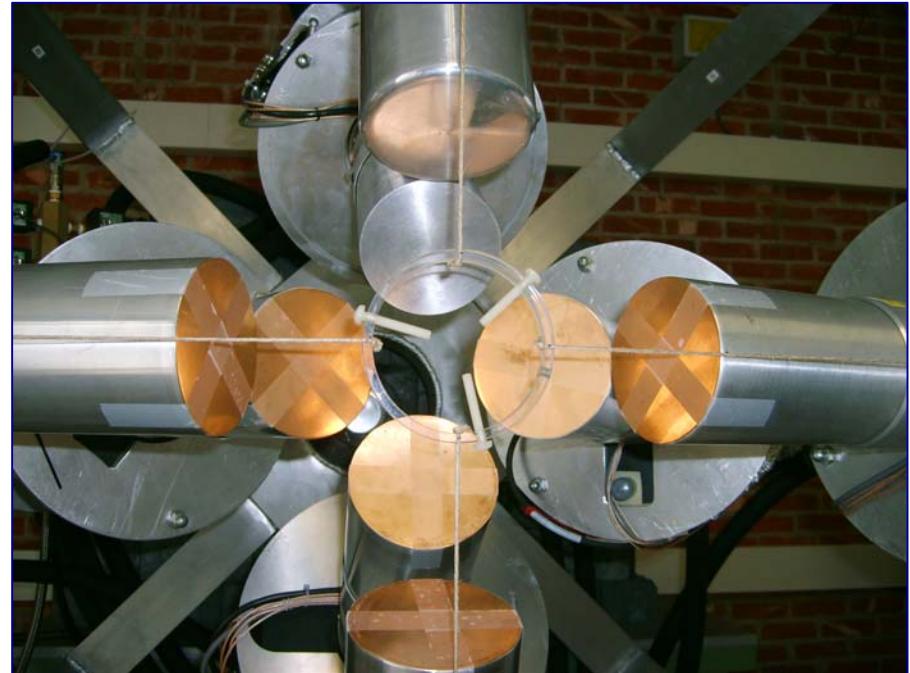
De Smet et al., Accepted PRC

$^{234}\text{U}(\text{n},\text{f})$



Heyse et al., NSE 156, 211 (2007)

In-elastic scattering at 30 m and 200 m Direct spectrum

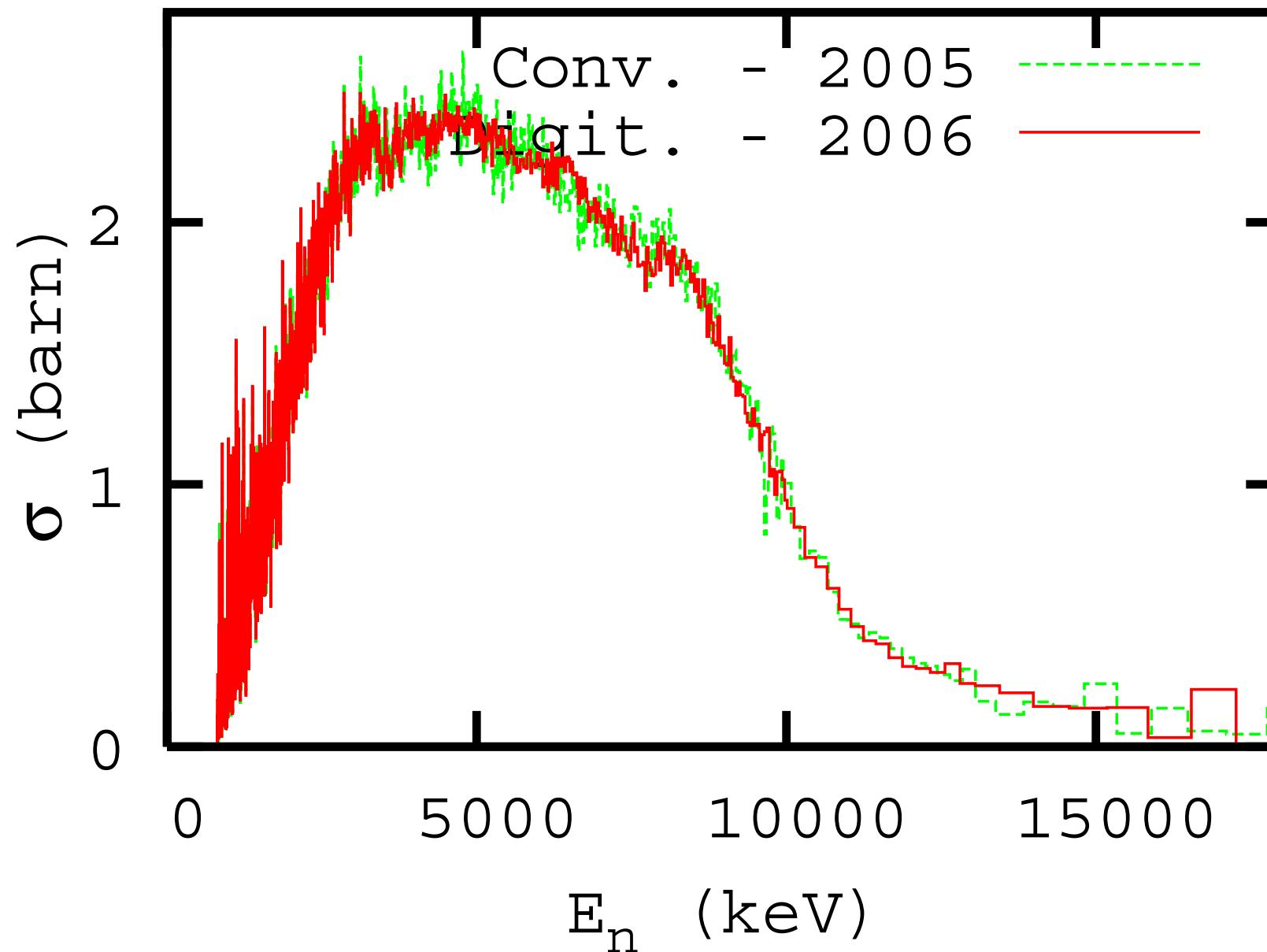


2001-2006 : 2 – 4 HPGe 100 %
 ^{56}Ni , ^{52}Cr , ^{209}Bi , $^{206, 207, 208}\text{Pb}$

2006 – 2007 : 8 HPGe (100 %)
 ^{56}Fe , ^{28}Si

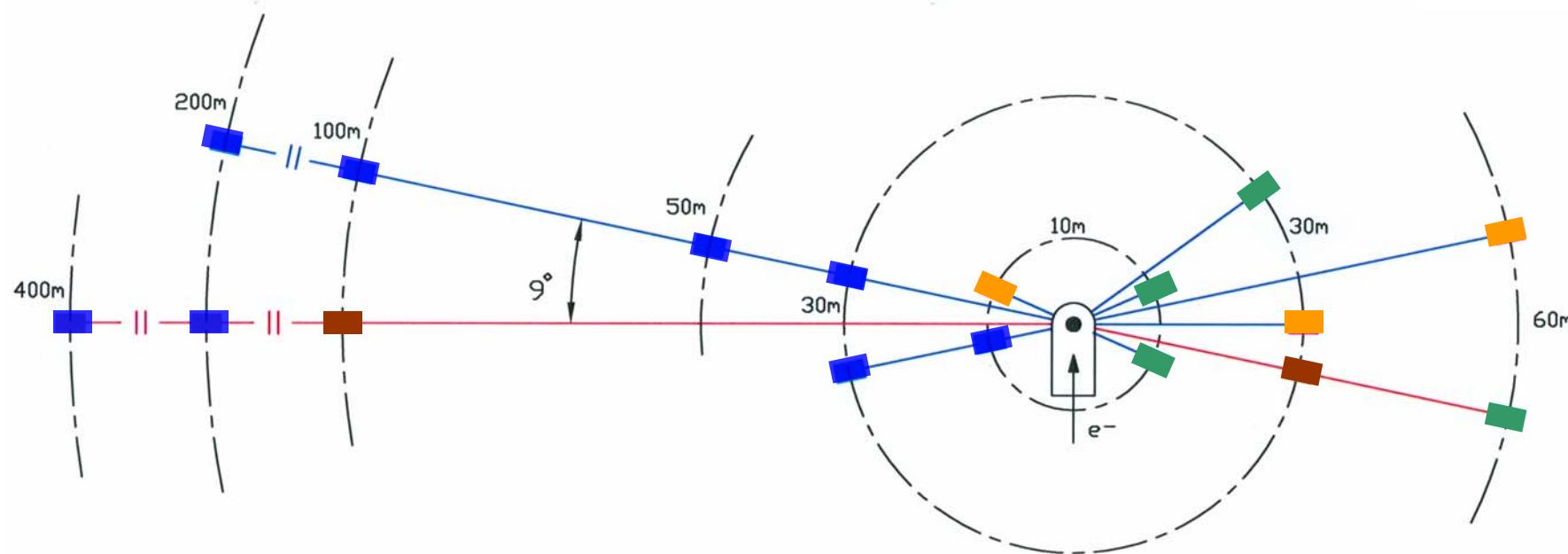
Mihailescu et al., NP A786, 1 (2007)

$^{206}\text{Pb}(n, n'\gamma)$ in-elastic scattering cross section



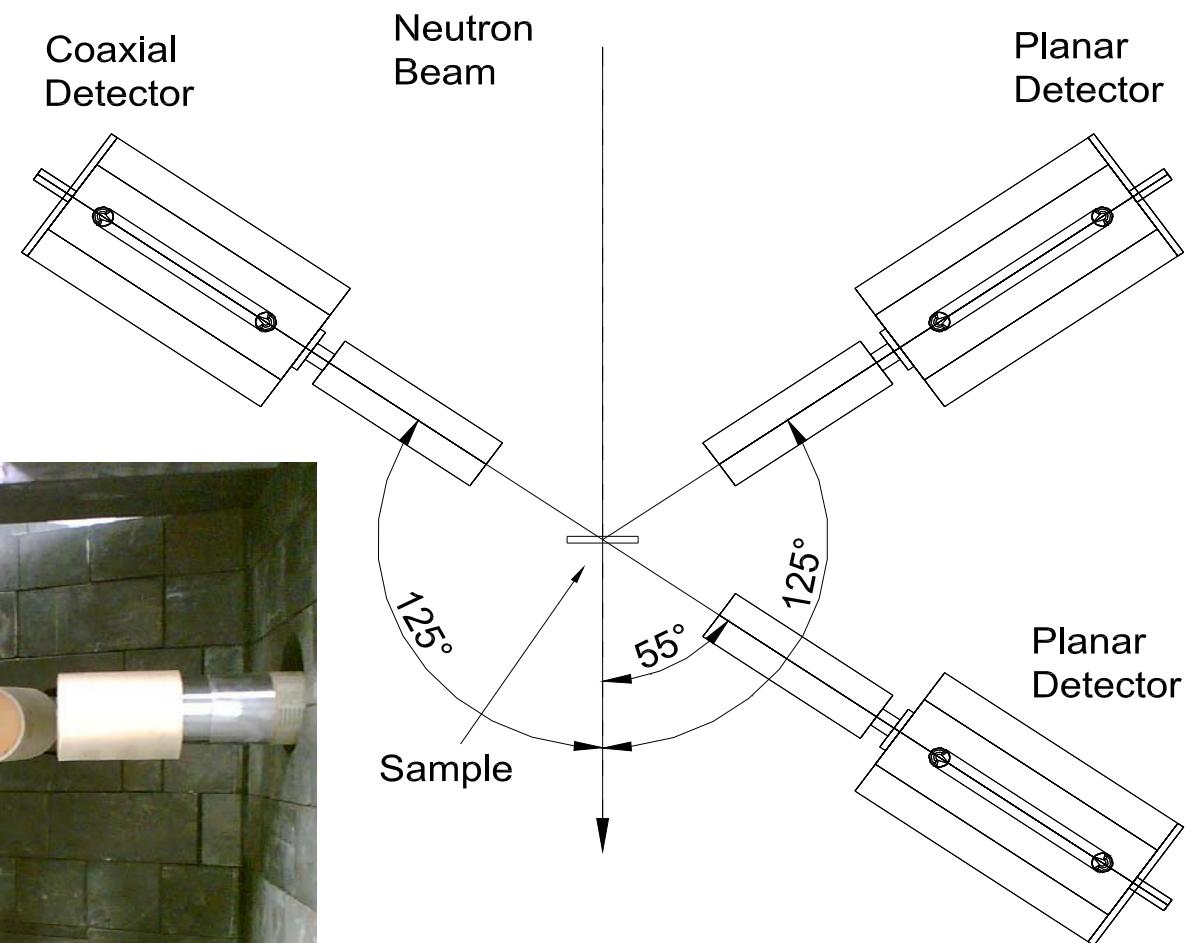
Capture Measurement Stations

— Direct spectrum
— Moderated spectrum



- **C_6D_6 detectors ($L = 10, 30$ and 60 m)**
 - Study of resonance structured $\sigma(n,\gamma)$ from thermal up to 1 MeV
- **Ge-detectors ($L = 10$ m)**
 - Spin and parity PR C 61, 054616 (2000)
 - Partial capture cross sections & branching ratio
 - Isotope identification

Ge-detectors at L = 10 m

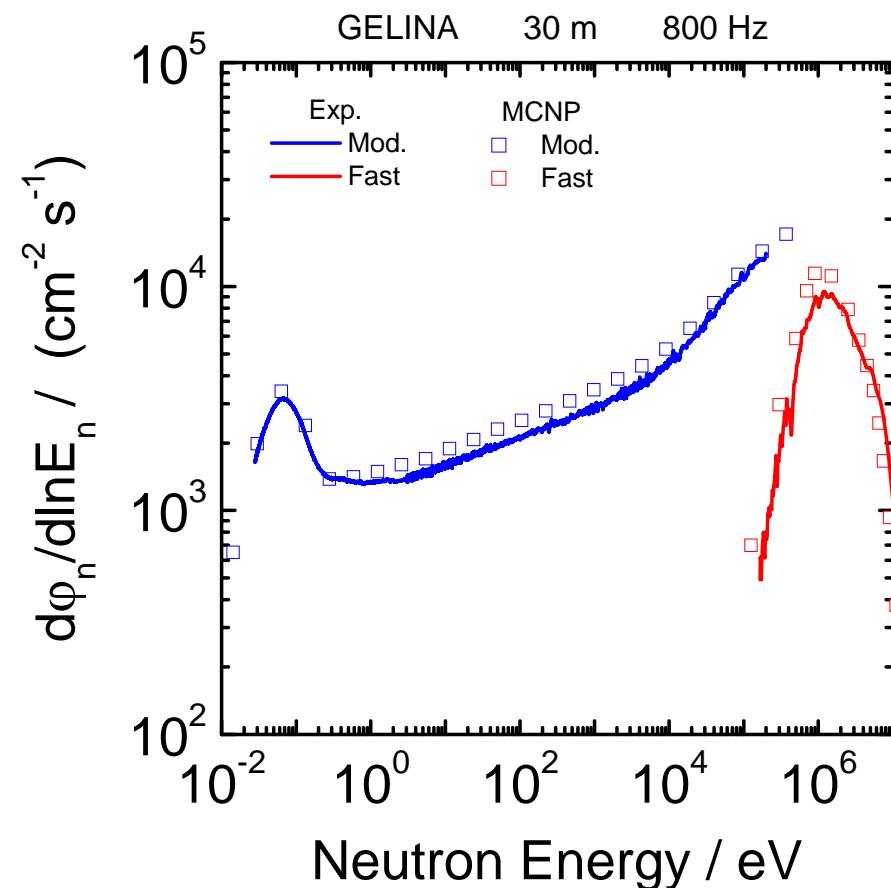


Improve the quality of the cross section data obtained at GELINA:

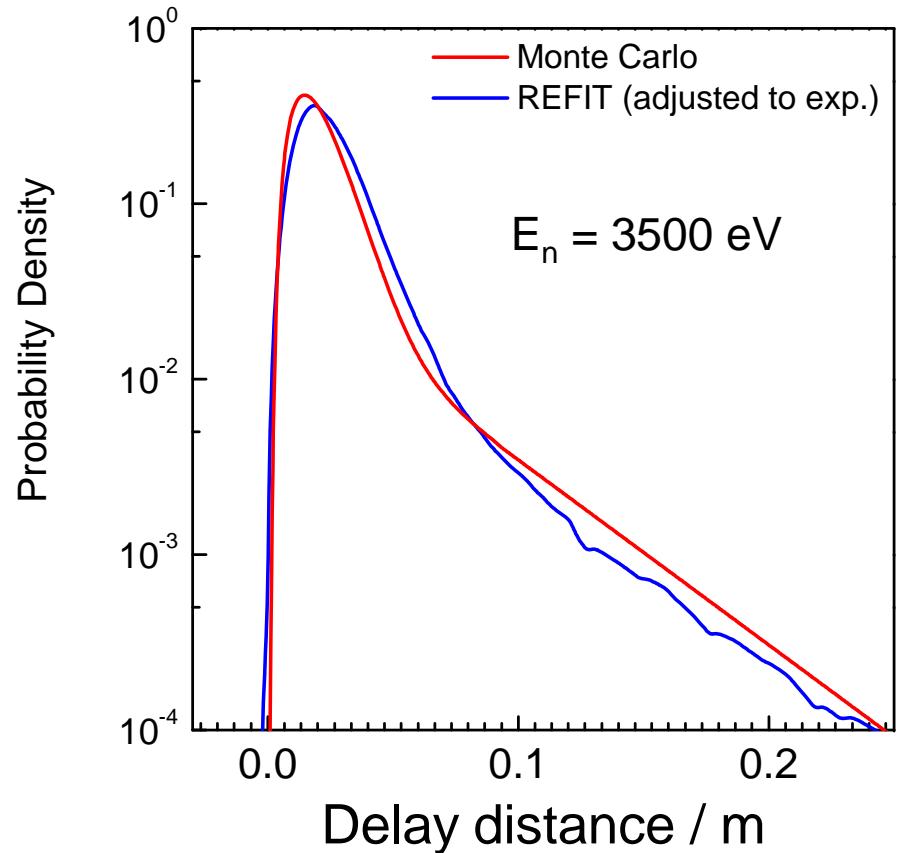
- Better understanding of neutron producing target by MC-simulations
- Use of digitizers to replace conventional analog electronics
- Data reduction procedures with full covariance information
- Improved data reduction for capture measurements
- Improved RSA for thin sample transmission data
- Support the development of analysis codes for RRR and URR

Characterisation of GELINA : TOF-Facility

Neutron Flux



Resolution Function

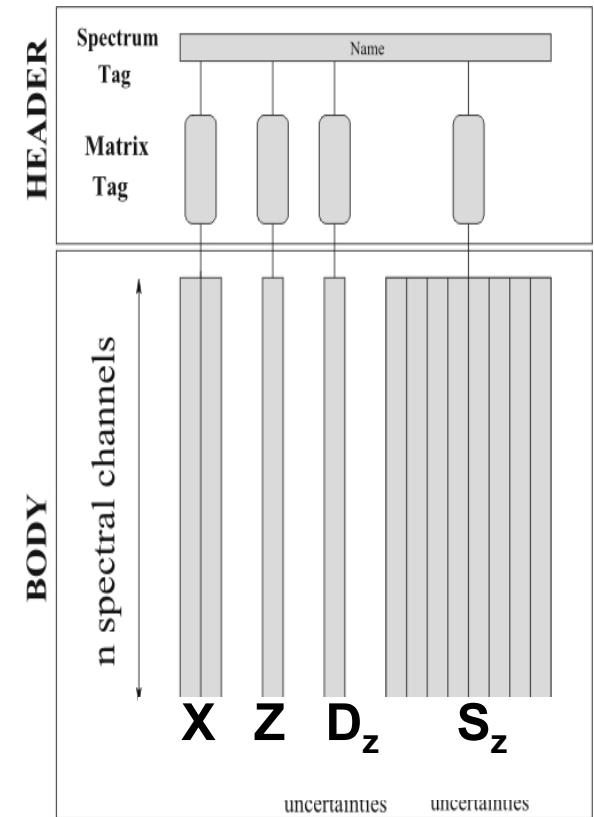


Flaska et al., NIM , A531, 394 (2004)

Data reduction with full covariance information

Analysis of Generic tof Spectra (AGS)

- Transform count rate spectra into observables (transmission factors, partial reaction yields)
- Full propagation of uncertainties starting from counting statistics
- Output: complete covariance matrix
- Special format for covariance matrix
- Due to the special format used in AGS:
 - Reduce space for data storage ([EXFOR](#))
 - Verify and document the sources of uncertainties in each step of the reduction process



Observable Z (dim. n) with k sources of correlated uncertainties \Rightarrow

D_z : uncorrelated part
n values

S_z : correlated part
dim. (n x k)

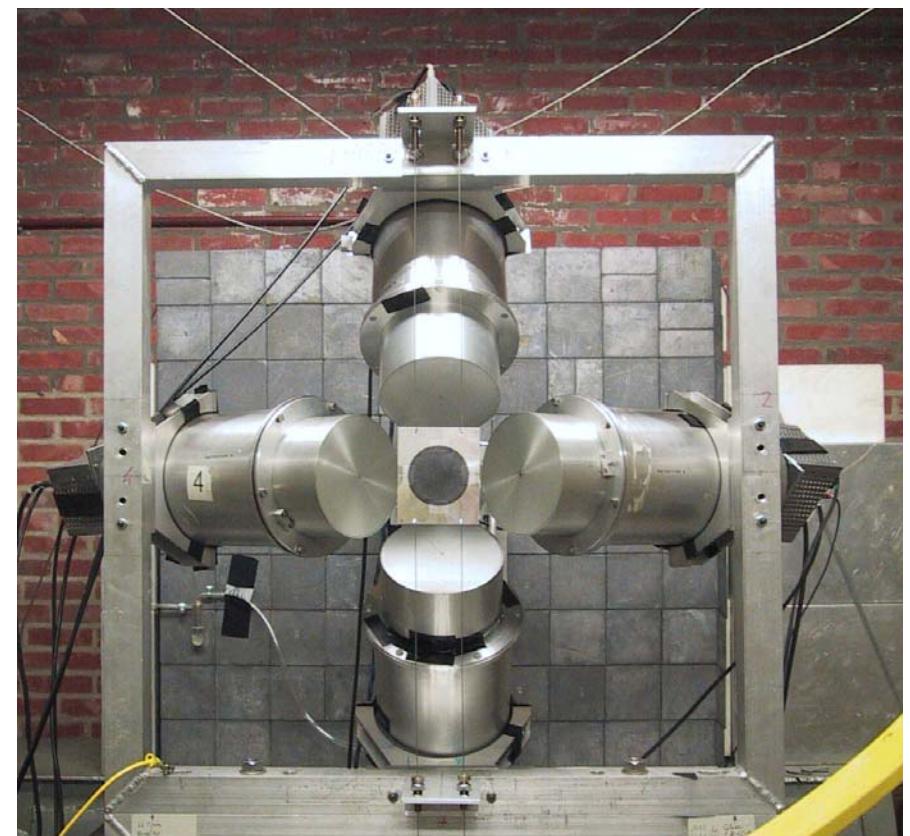
$$C_z = D_z + S_z S_z^T$$

Total energy detection

- **C_6D_6 liquid scintillators**
 - 125°
 - PHWT $\int R(E_d, E_\gamma) WF(E_d) dE_d = kE_\gamma$
- **Flux measurements (IC)**
 - $^{10}B(n,\alpha)$
 - $^{235}U(n,f)$



$L = 10 \text{ m}, 30 \text{ m}$ and 60 m



$$Y_{\text{exp}} = \sigma_\varphi \frac{C_w - B_w}{C_\varphi - B_\varphi}$$

WF : from MC simulations

$$C_w(T_n) = \int C_c(T_n, E_d) WF(E_d) dE_d$$

Improved Data Reduction for (n,γ)

Reliable WF's can be obtained by Monte Carlo simulations provided that the geometry input reflects the experimental conditions, i.e. accounts for γ -ray transport in sample

(started with Perey et al. at ORELA)

⇒ Weak resonance : WF1

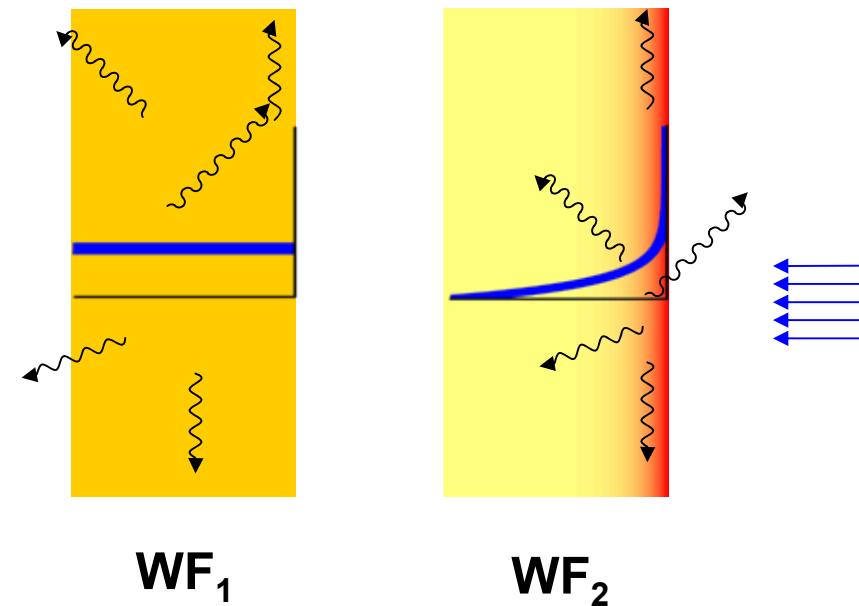
⇒ Strong resonance : WF2

(Affects also the observed shape)

Procedure :

(1) Apply WF1 on experimental data

(2) Correction factor on calculated yield



WF₁

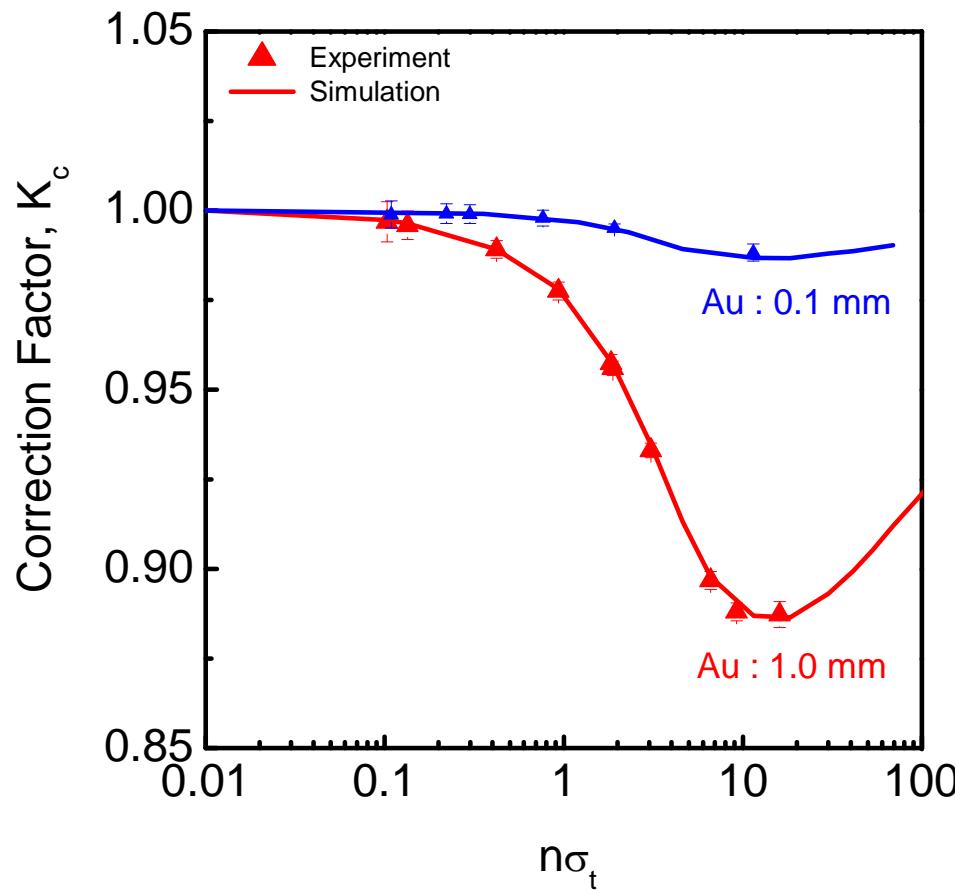
WF₂

$$K_c(n\sigma_t) = \frac{\langle WF_1 \rangle}{\langle WF_2 \rangle}$$

$$Y_{\text{exp}} = N \int R(T_n, E_n) (K_c \varepsilon_{\text{cw}} Y_c + \varepsilon_{\text{nw}} Y_n) dE_n$$

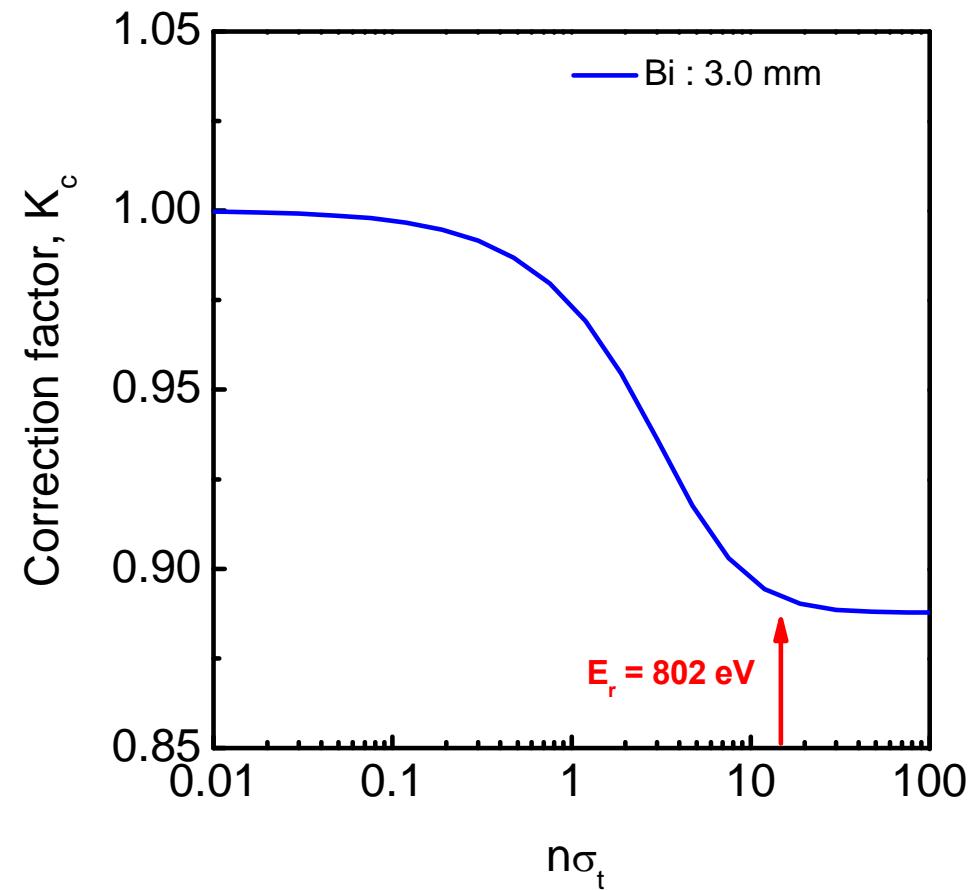
Examples : Au & Bi

Au



MC: γ -ray spectrum from DICEBOX

Bi



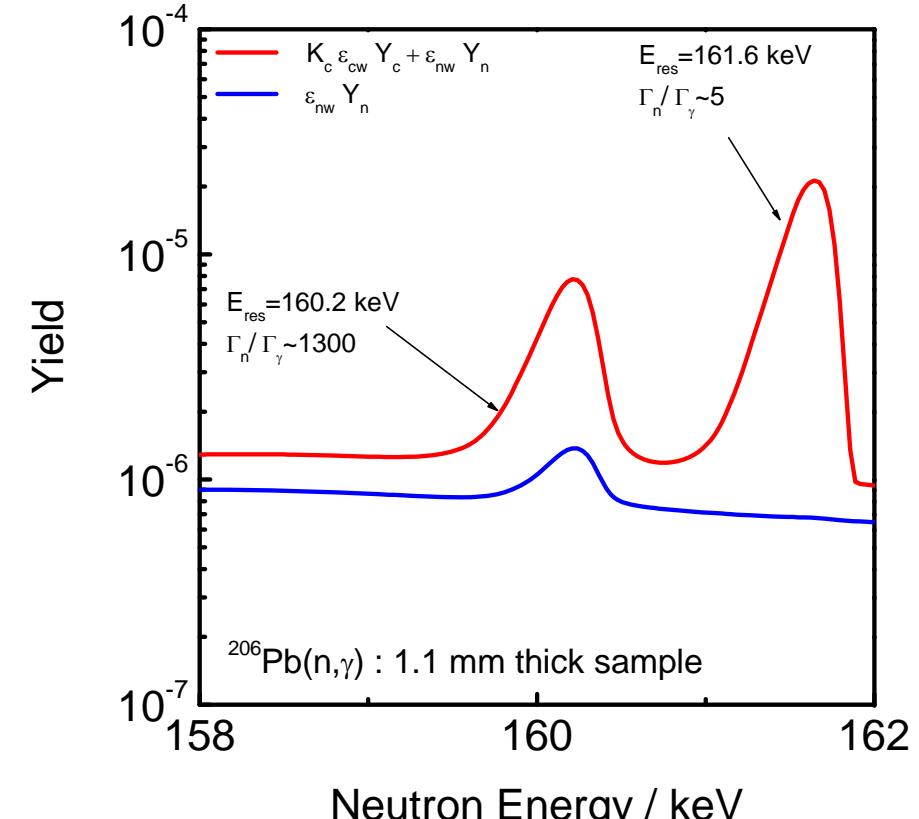
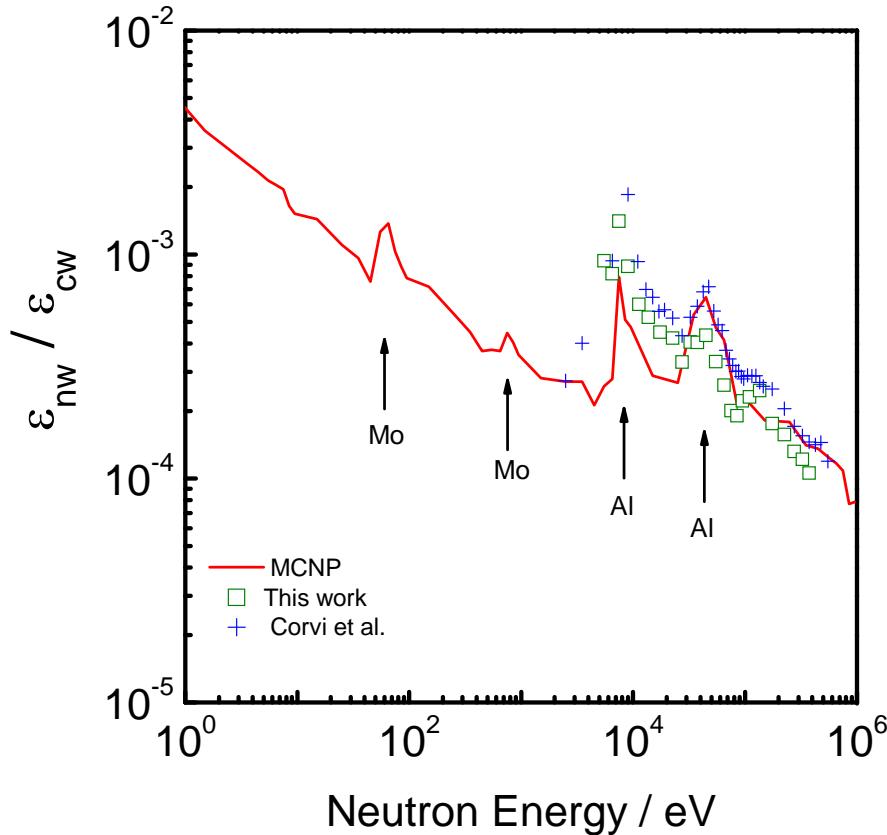
MC: γ -ray spectrum from measurements at BNC

Neutron sensitivity

$$Y_{\text{exp}} = N \int R(T_n, E_n) (K_c \varepsilon_{\text{cw}} Y_c + \varepsilon_{\text{nw}} Y_n) dE_n$$

$$Y_c = (1 - e^{-n\sigma_t}) \frac{\sigma_\gamma}{\sigma_t} + Y_m$$

$$Y_n = (1 - e^{-n\sigma_t}) \frac{\sigma_n}{\sigma_t} - Y_m$$



Borella et al., j.nima.2007.03.034

REFIT

$\Rightarrow \Gamma_\gamma = 53.5 \text{ meV}$

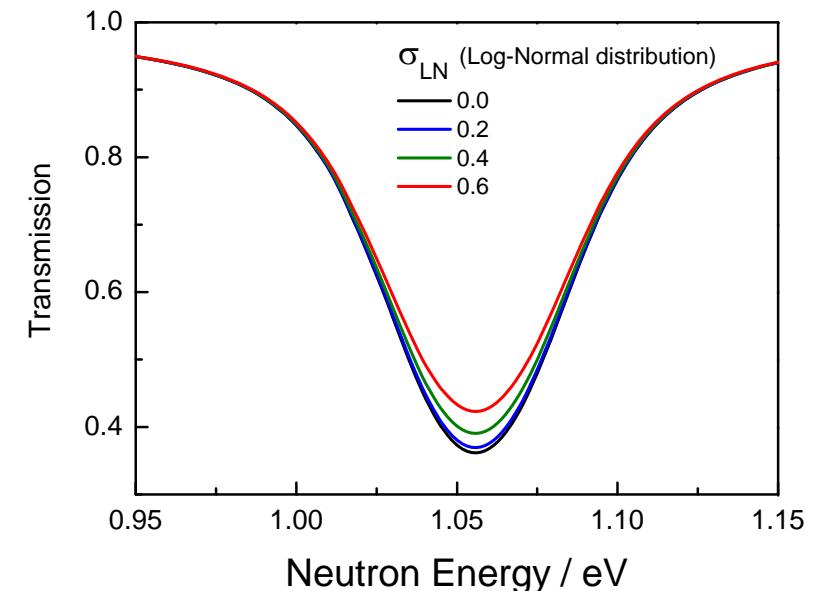
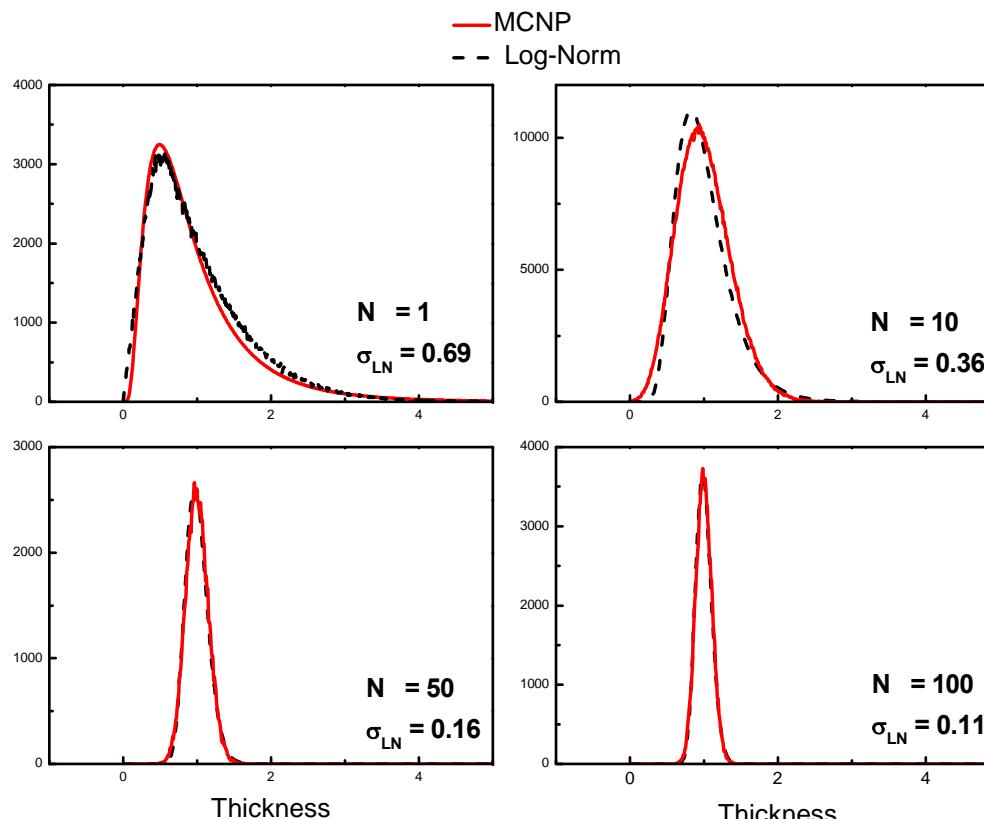
$$\Gamma_\gamma' = \Gamma_\gamma - \frac{\varepsilon_n}{\varepsilon_c} \Gamma_n$$

$$\Rightarrow \Gamma_\gamma = 48.3 \text{ meV}$$

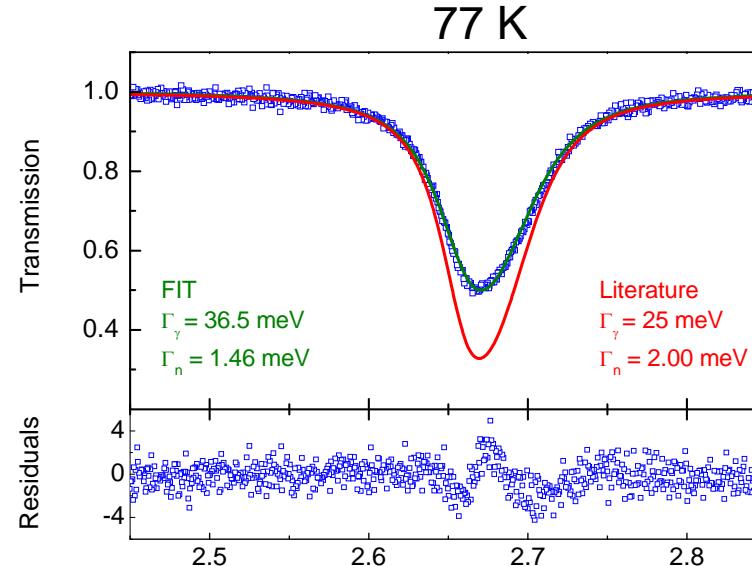
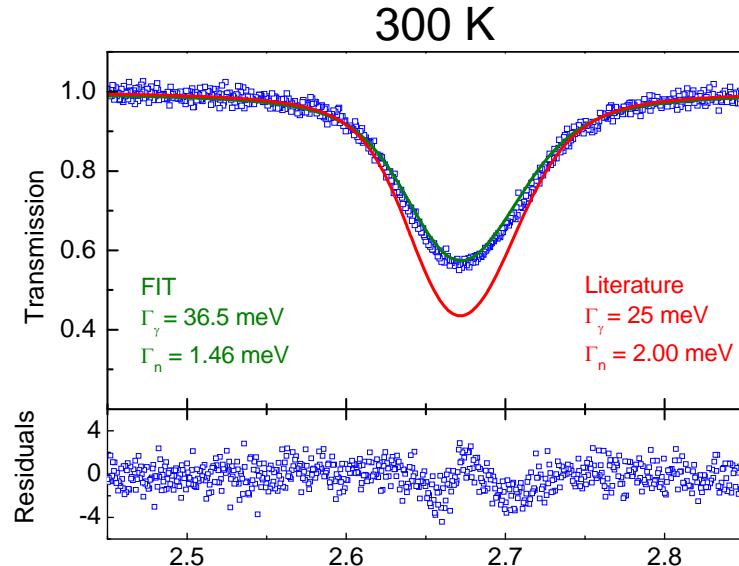
Improved RSA of thin sample transmission data



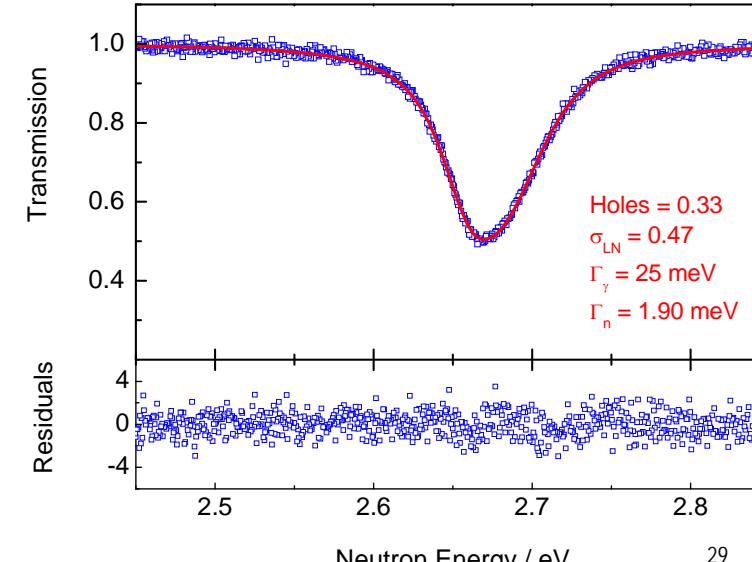
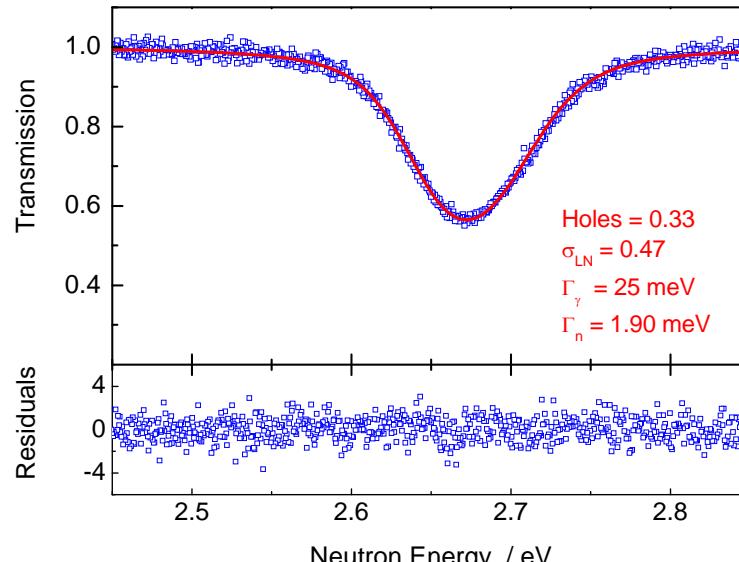
Variation of sample thickness can be described by a Log-Normal distribution with free parameter σ_{LN}



Thin sample transmission data : ^{242}Pu $2.5 \cdot 10^{-5}$ at/b



Variation of thickness described by Log-Normal distribution



Support the development of analysis codes for the parameterisation of experimental data in terms of RP

- **Resolved resonance region : REFIT**

(Moxon, WONDER 2006)

- Tested and documented version
(collaboration with Serco Assurance: C. Dean)
- Implement improvements (EFNUDAT support for scientific visit M. Moxon)
 - Improved analysis of capture data
 - Treatment of variation in sample thickness for transmission
(Kopecky et al. ND2007)
 - Accommodate different resolution functions
(Kopecky et al., WONDER 2006)

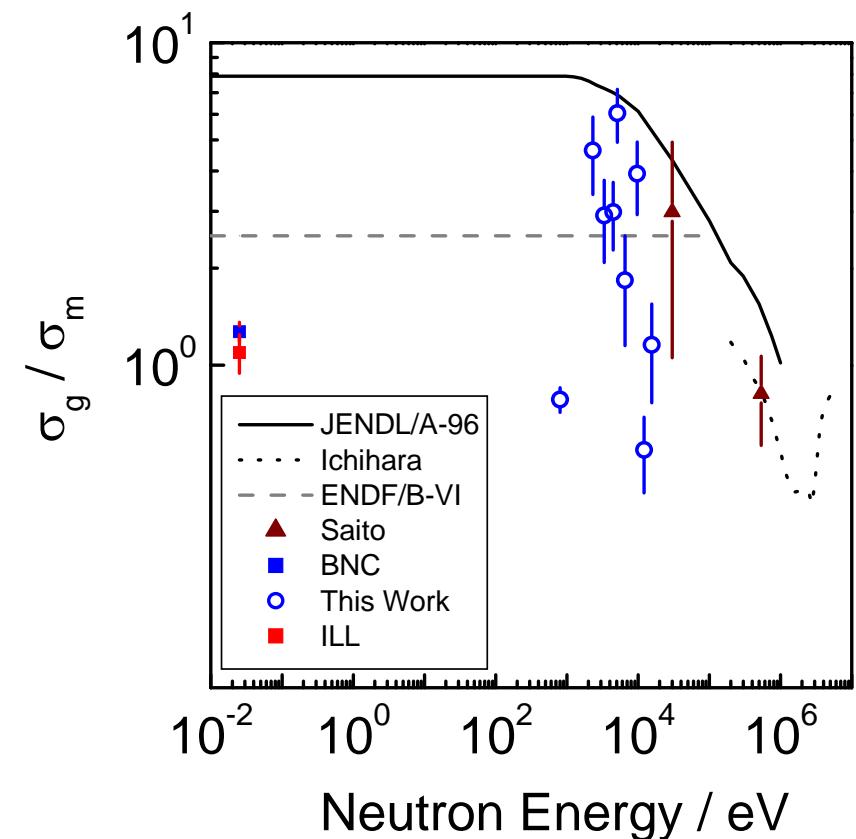
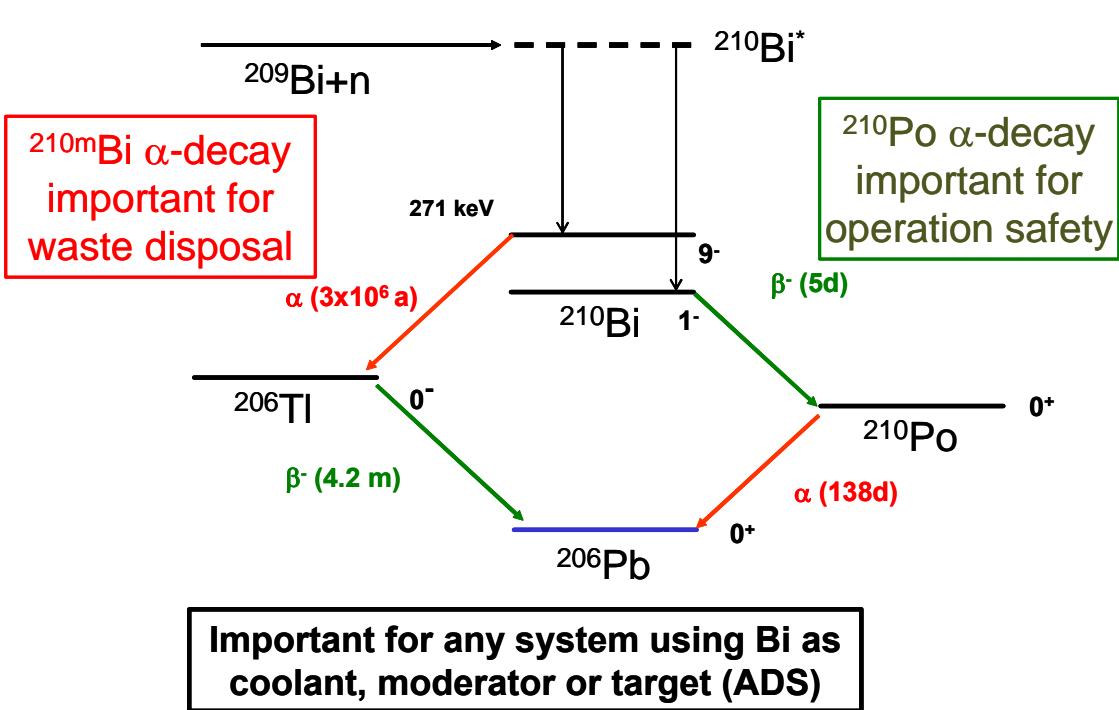
- **Unresolved resonance region (collaboration with IAEA)**

(Sirakov et al., WONDER 2006)

- HF + WF
- Link to optical model (DCCOM) (Capote et al.)
- Covariance data
- ENDF-6 compatible output

- **Transmutation of nuclear waste**
 - RP for ^{99}Tc , ^{129}I , ^{237}Np , ^{241}Am
- **High Temperature Reactors**
 - $^{240,242}\text{Pu}$ – Doppler effect
- **Development of ADS systems**
 - ^{209}Bi -branching
 - $^{206,207,208}\text{Pb}$ and ^{209}Bi (n,tot), (n,γ), ($n,n'\gamma$)
- **Th-U fuel cycle**
 - $^{234,236}\text{U}(n,f)$
 - RP for ^{232}Th (RRR + URR)
- **U-Pu fuel cycle**
 - Fission products
 - ^{55}Mn

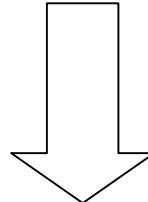
$^{209}\text{Bi}(n,\gamma)$ - branching ratio at L = 10 m with Ge-detectors



Borella et al., ND2007

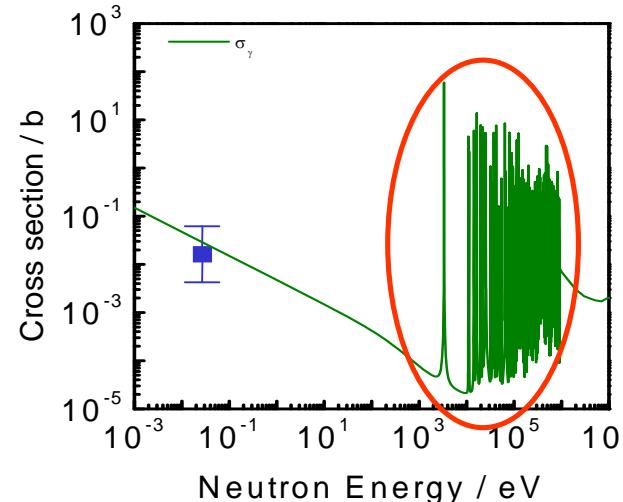
ADS : $^{206}\text{Pb} + n$ resonance parameter file

Thermal
neutron
energy



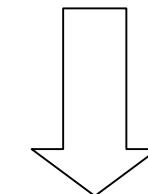
BNC

$$\sigma(n_{\text{th}}, \gamma) = 27.3 (8) \text{ mb}$$



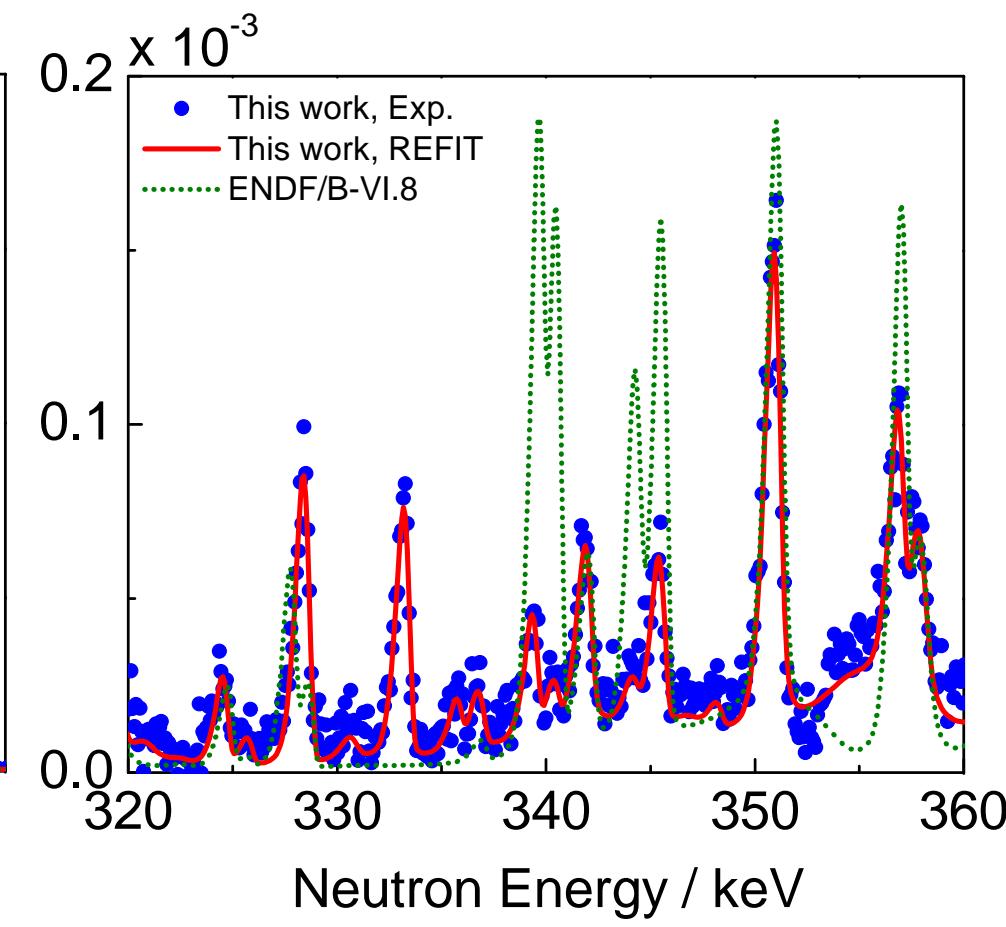
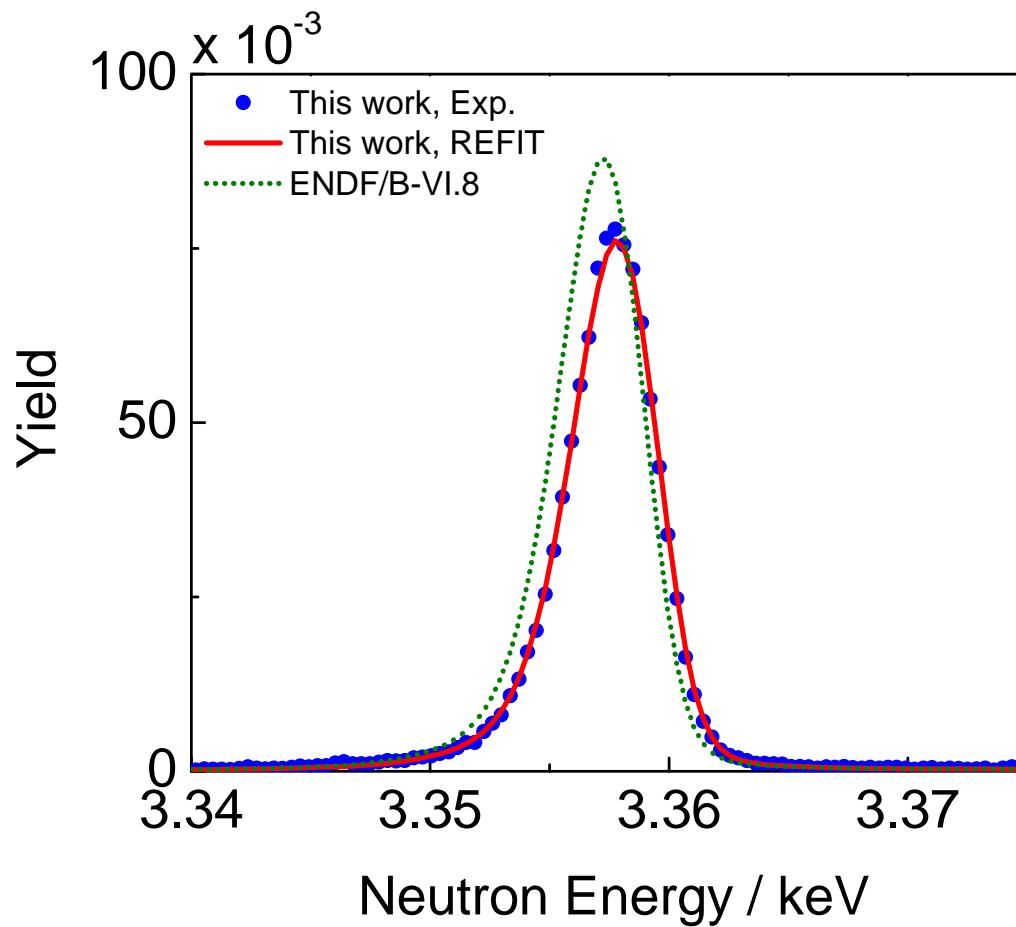
RRR

GELINA



- Resonance parameter file (matching thermal values)
 - 304 resonances up to 620 keV (221 in ENDF/B-VI.8)
 - $R' = 9.54 (2) \text{ fm}$
 - $E_o > 80 \text{ keV}$: Γ_n from ORELA, (Horen et al., PR C20, 478 (1979))
- MAC cross sections (based on 304 resonances !)
- Photon strength function from C_6D_6 spectra
 - $f_{E1} < 225 (5) 10^{-9} \text{ MeV}^{-3}$
 - f_{M1} follows systematics (no M1 enhancement)
 - $B(E2) \downarrow$

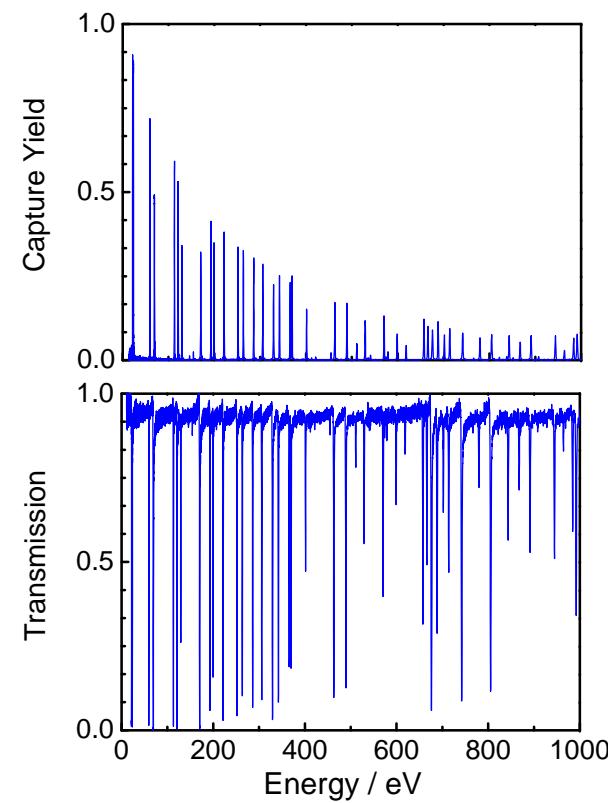
$^{206}\text{Pb} + \text{n} : \text{results}$



⇒ new evaluation : re-analyse together with ORELA data
(transmission and scattering)

IAEA CRP: Thorium – Uranium Fuel Cycle

Resonance parameters (RRR & URR) for $^{232}\text{Th} + \text{n}$



Facility	Measurement Type	Flight Path Length	Target Thickness at/b
ORELA	Transmission	180 m	$1.68 \cdot 10^{-4}$ $14.00 \cdot 10^{-4}$ $38.70 \cdot 10^{-4}$ $193.1 \cdot 10^{-4}$
GELINA	Transmission	50 m	$8.00 \cdot 10^{-4}$ $34.0 \cdot 10^{-4}$ $61.0 \cdot 10^{-4}$
GELINA	Capture Borella et al., NSE 152, 1 (2006)	15 m 60 m	$15.8 \cdot 10^{-4}$ $34.0 \cdot 10^{-4}$
n-TOF	Capture	200 m	$41.0 \cdot 10^{-4}$

RRR (ORNL) + URR (IRMM) \Rightarrow ENDF/B-VII

Optimise the U - Pu fuel cycle

More accurate nuclear data are needed for:

- 1) Tendency to operate conventional nuclear power plants at increased fuel burn-up
- 2) Fuel cycles based on reprocessed fuel
- 3) Spent fuel storages (ORNL – RADWASTE)

Nuclide	Priority Level	
	Storrer et al.	Salvatores
	Burnup	Reprocessing
¹⁰³ Rh	1	
¹³³ Cs	1	
¹⁵⁵ Gd		1
¹⁴⁷ Sm		1
¹⁴⁹ Sm	1	
¹⁵² Sm		1
¹⁴³ Nd	2	1
¹⁴⁵ Nd		1
¹⁵³ Eu	2	1
¹⁵⁵ Eu	1	

1 & 2 : Collaboration with CEA Saclay (F)

$$\sigma_\gamma < 2-3\%$$

3 : Collaboration with ORNL (US)

M. Salvatores, ND2002, J. Sci. & Techn. 2 (2002) p. 4
 F. Storrer et al., ND2002, J. Sci. & Techn. 2 (2002) p. 1357
 NEA High Priority List, March 2001

Collaboration ORNL – IRMM

Isotope	Transmission				Capture			
	Flight Path	Frequency	Target Thick.	Data Red.	Flight Path	Frequency	Target Thick.	Data Red.
⁵⁵ Mn	30 m	800 Hz	$1.2 \cdot 10^{-1}$ at/b			30 m	$4.4 \cdot 10^{-4}$ at/b	
	50 m	800 Hz	$4.4 \cdot 10^{-4}$ at/b	Done		30 m	$1.9 \cdot 10^{-2}$ at/b	
	50 m	800 Hz	$1.5 \cdot 10^{-3}$ at/b	Done		60 m	$1.0 \cdot 10^{-2}$ at/b	
	50 m	800 Hz	$1.0 \cdot 10^{-2}$ at/b	Done		60 m	$1.9 \cdot 10^{-2}$ at/b	Done
	50 m	800 Hz	$1.9 \cdot 10^{-2}$ at/b	Done				
¹⁰³ Rh	50 m	100 Hz	$3.4 \cdot 10^{-4}$ at/b	Done		15 m	40 Hz	$1.8 \cdot 10^{-3}$ at/b
	50 m	100 Hz	$2.2 \cdot 10^{-3}$ at/b	Done		30 m	800 Hz	$1.8 \cdot 10^{-3}$ at/b
	50 m	400 Hz	$4.6 \cdot 10^{-2}$ at/b	Done		30 m	800 Hz	$3.4 \cdot 10^{-4}$ at/b
	50 m	800 Hz	$4.6 \cdot 10^{-2}$ at/b	Done		60 m	800 Hz	$1.8 \cdot 10^{-3}$ at/b
¹³³ Cs	30 m	800 Hz	$2.3 \cdot 10^{-4}$ at/b			15 m	50 Hz	$3.8 \cdot 10^{-4}$ at/b
	30 m	800 Hz	$1.0 \cdot 10^{-2}$ at/b			15 m	50 Hz	$1.5 \cdot 10^{-3}$ at/b
	50 m	800 Hz	$2.3 \cdot 10^{-4}$ at/b	Done		15 m	50 Hz	$5.2 \cdot 10^{-3}$ at/b
	50 m	800 Hz	$5.2 \cdot 10^{-3}$ at/b	Done		30 m	800 Hz	$2.3 \cdot 10^{-4}$ at/b
						30 m	800 Hz	$3.8 \cdot 10^{-4}$ at/b
^{nat} Gd	30 m	800 Hz	$1.5 \cdot 10^{-3}$ at/b			30 m	800 Hz	$1.5 \cdot 10^{-3}$ at/b
	50 m	800 Hz	$6.0 \cdot 10^{-4}$ at/b	Done		30 m	800 Hz	$5.2 \cdot 10^{-3}$ at/b
	50 m	800 Hz	$1.5 \cdot 10^{-3}$ at/b	Done		30 m	800 Hz	$1.0 \cdot 10^{-2}$ at/b
						15 m	50 Hz	$6.0 \cdot 10^{-4}$ at/b
						30 m	50 Hz	$6.0 \cdot 10^{-4}$ at/b
¹⁵⁵ Gd	30 m	800 Hz	$3.4 \cdot 10^{-4}$ at/b			15 m	50 Hz	$6.0 \cdot 10^{-3}$ at/b
	30 m	800 Hz	$6.0 \cdot 10^{-3}$ at/b			30 m	800 Hz	$3.4 \cdot 10^{-4}$ at/b
						30 m	800 Hz	$6.0 \cdot 10^{-3}$ at/b

Criticality safety

RADWASTE

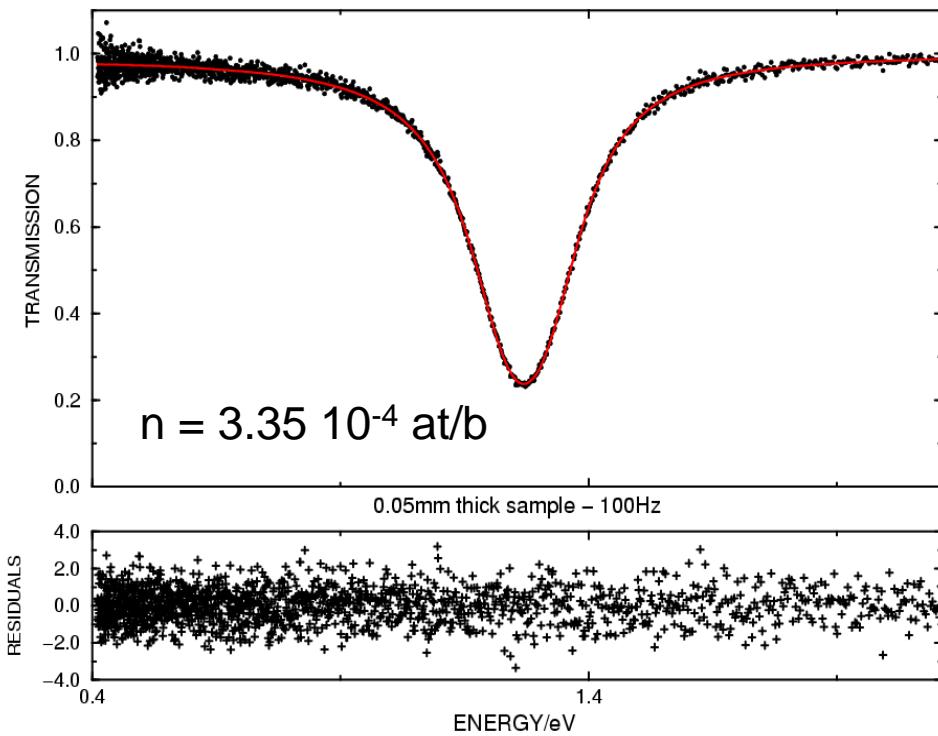
Combine transmission & capture
Different flight paths & facilities
Use of samples with different properties



Reduction of bias effects due to:
Resolution
Sample properties
Normalization of capture data

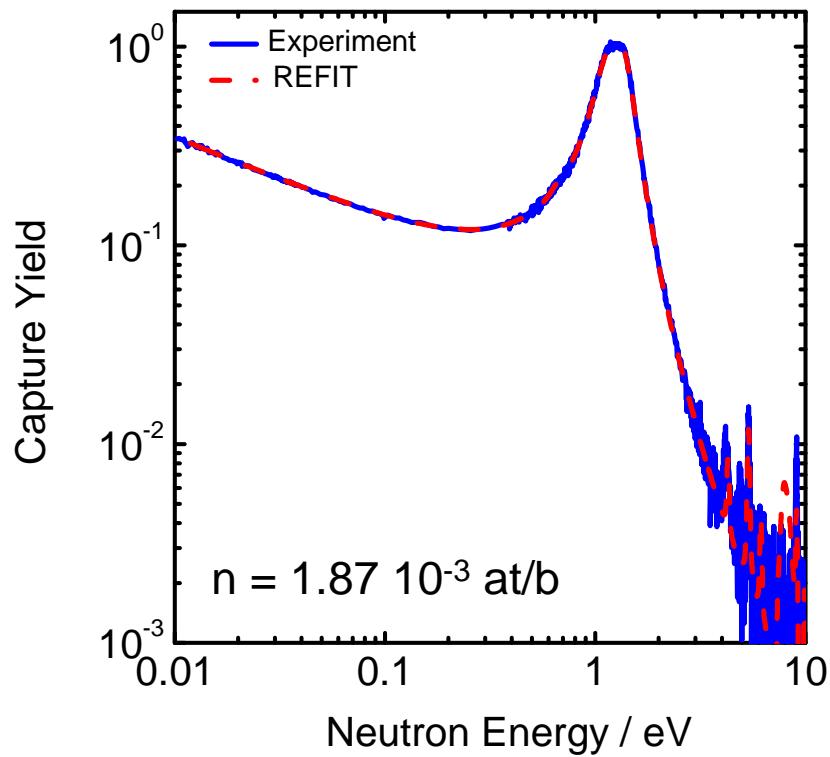
^{103}Rh : Thermal region

Transmission: 50 m 100 Hz



$E_r = 1.260 \text{ eV}$
 $\Gamma_n = 0.464 \text{ meV}$
 $\Gamma_\gamma = 156.0 \text{ meV}$
 $g = 3/4$

Capture : 15 m 40 Hz



$\sigma(n,\gamma) \text{ at } 25.3 \text{ meV}$

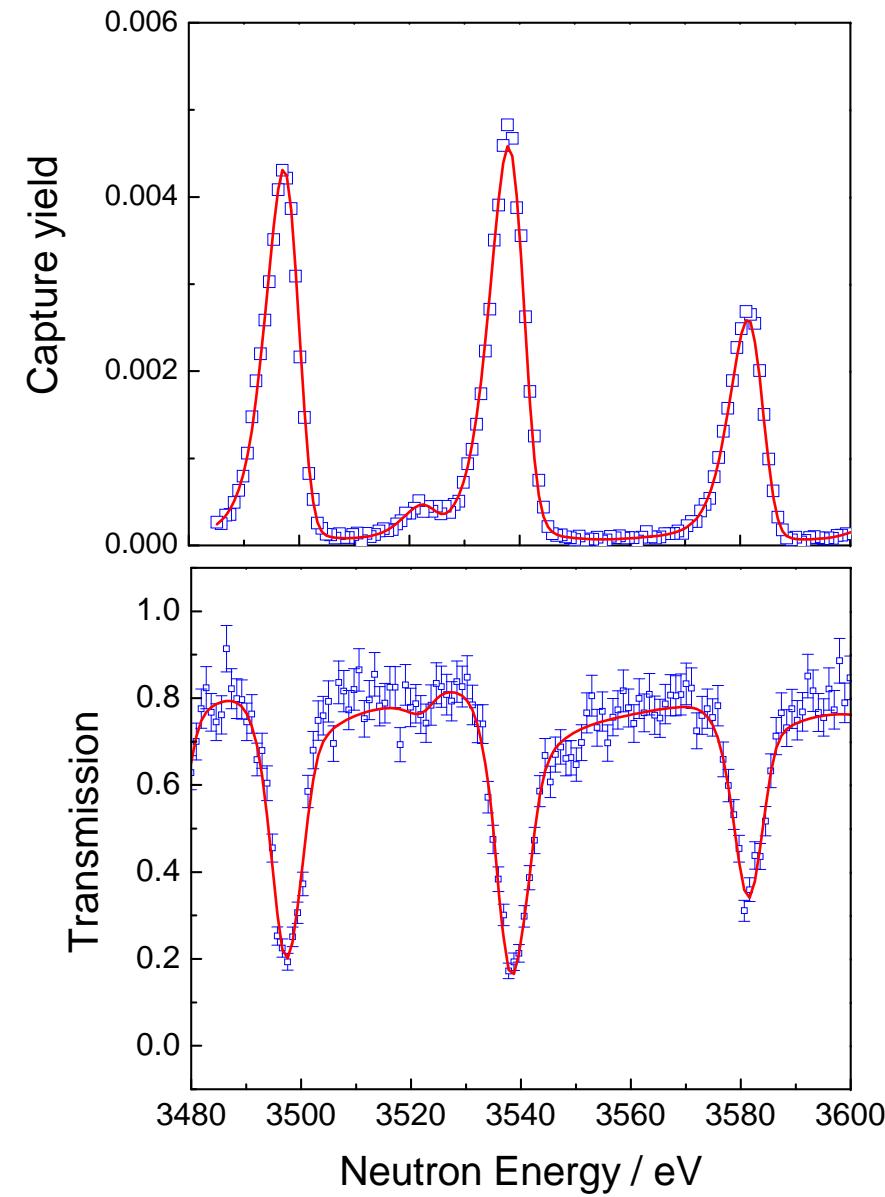
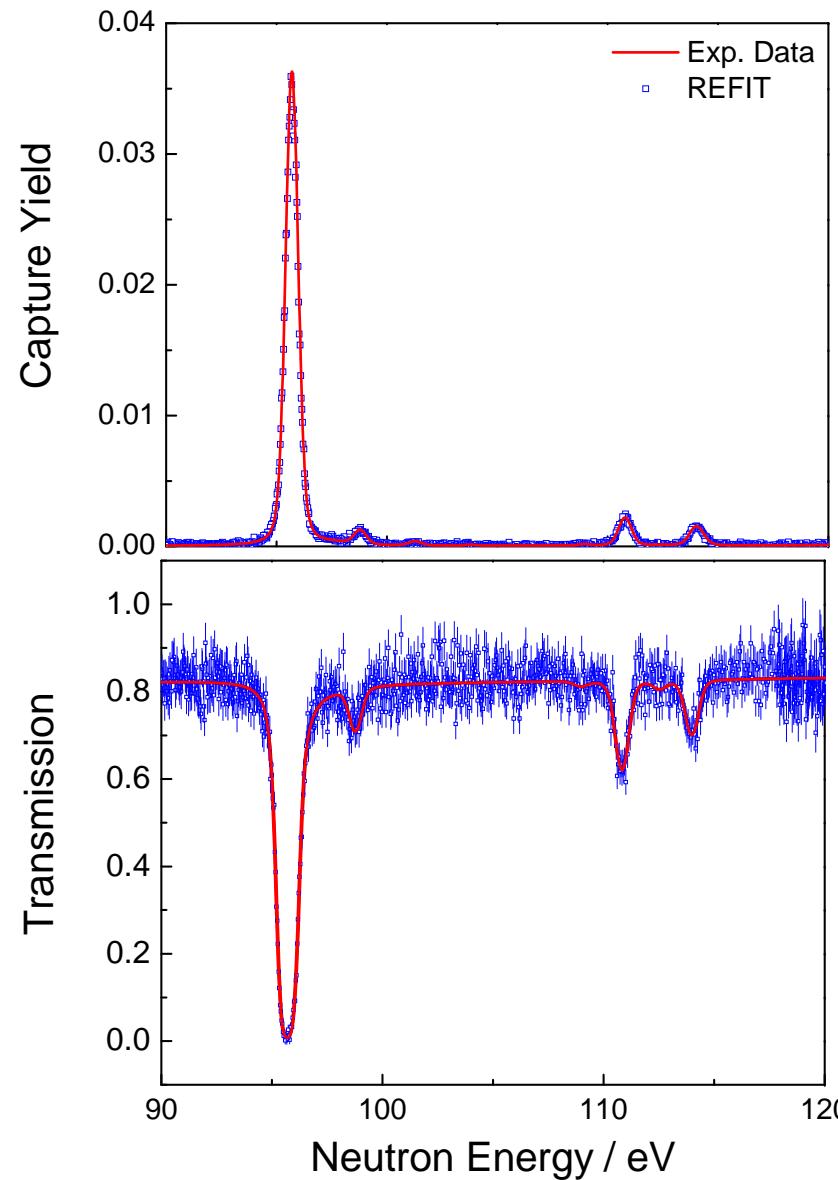
WF : 142.0 (1.5) b

1.0 % normalization

0.5 % uncorrelated

Counts : 143.3 b

^{103}Rh : Resolved Resonance Region



^{103}Rh : Unresolved Resonance Region

Improved the experimental data in URR :

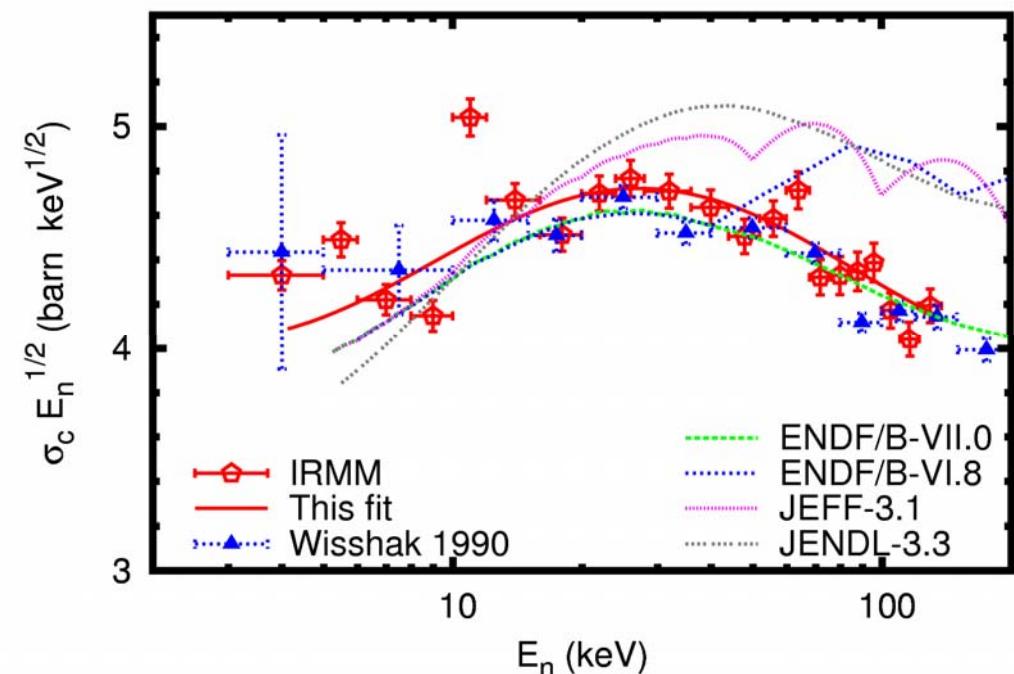
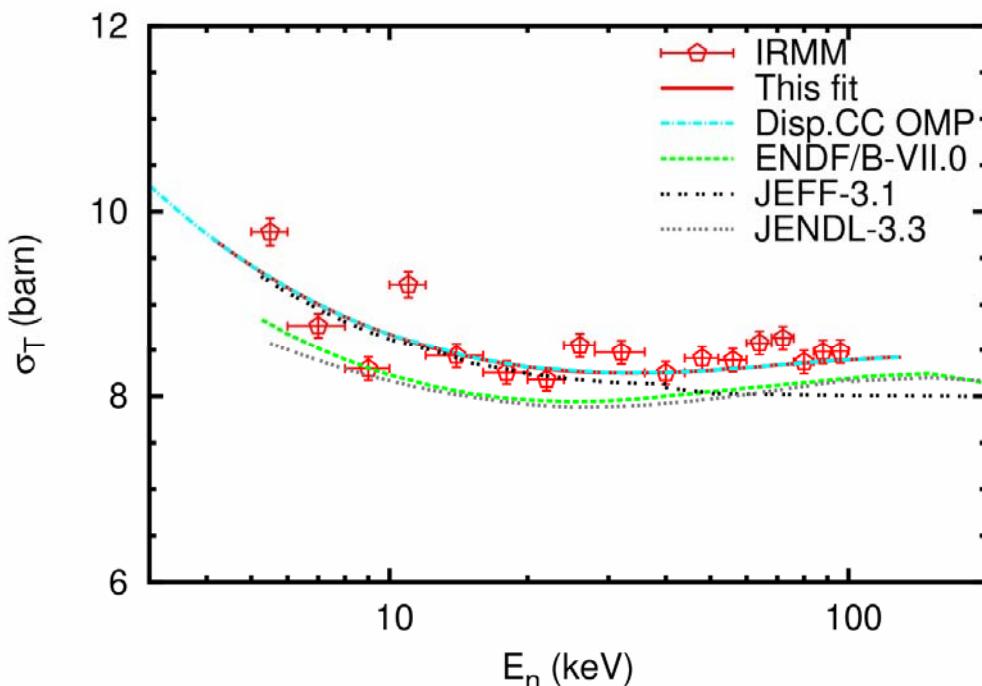
Transmission : 50 m, 800 Hz, $n = 4.49 \cdot 10^{-2}$

Capture : 30 m, 800 Hz, $n = 1.87 \cdot 10^{-3}$

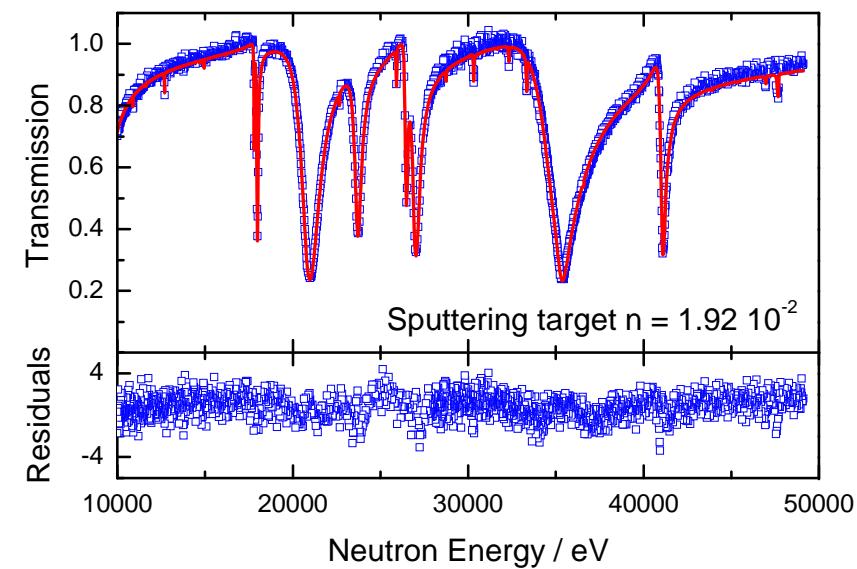
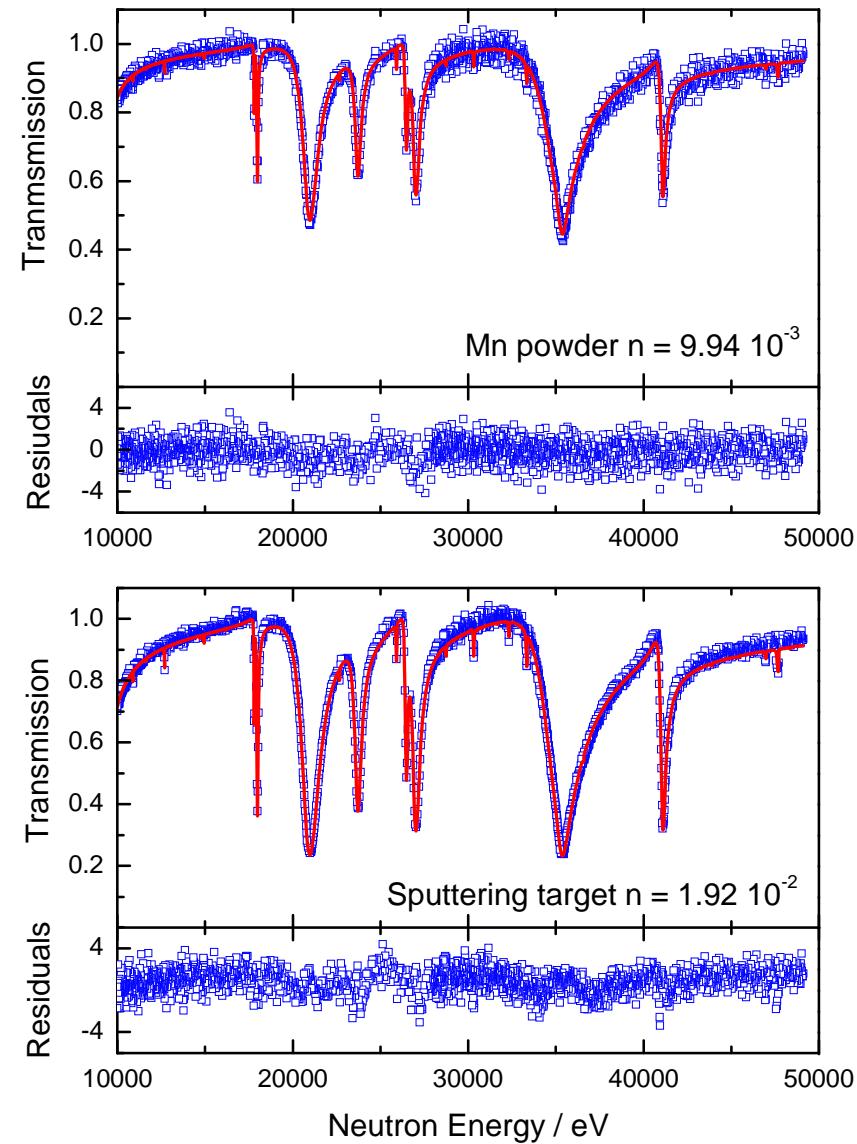
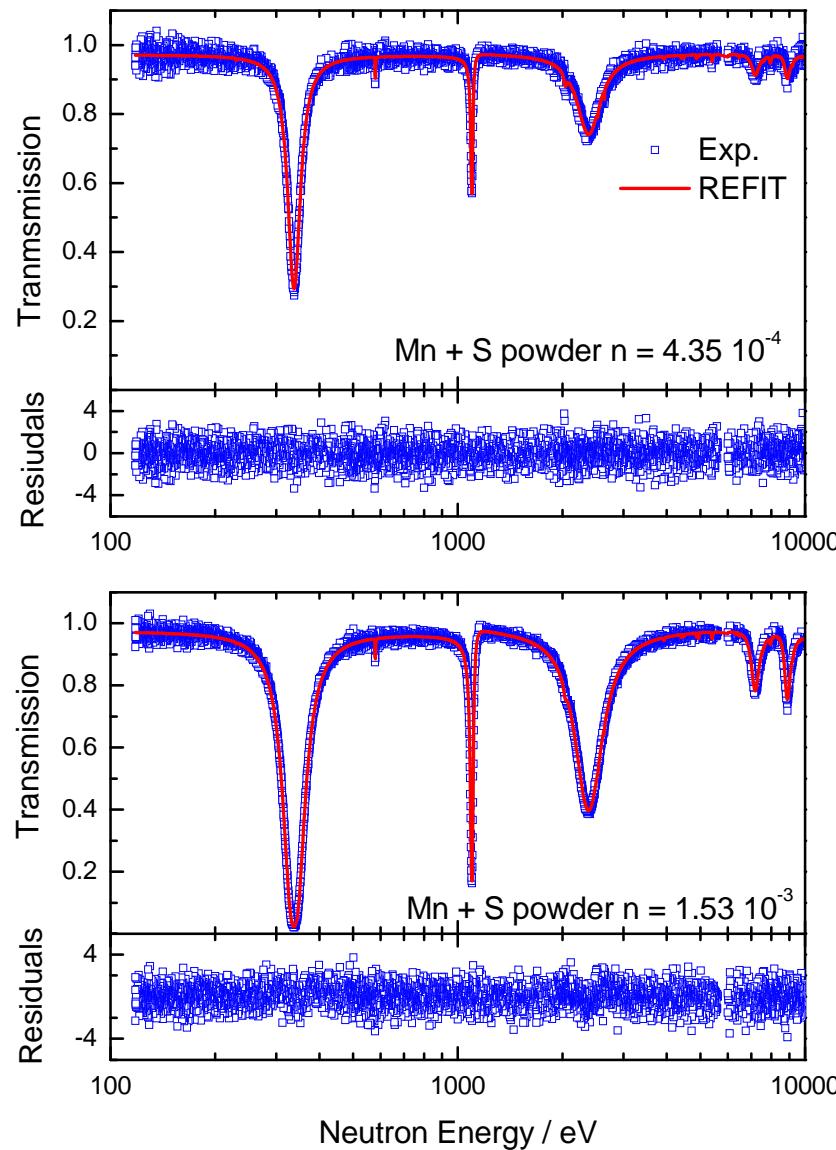
Analysis based on Sirakov et al. (WONDER 2006):

HF + WF with link to OM : Sirakov (IRMM) & Capote (IAEA)

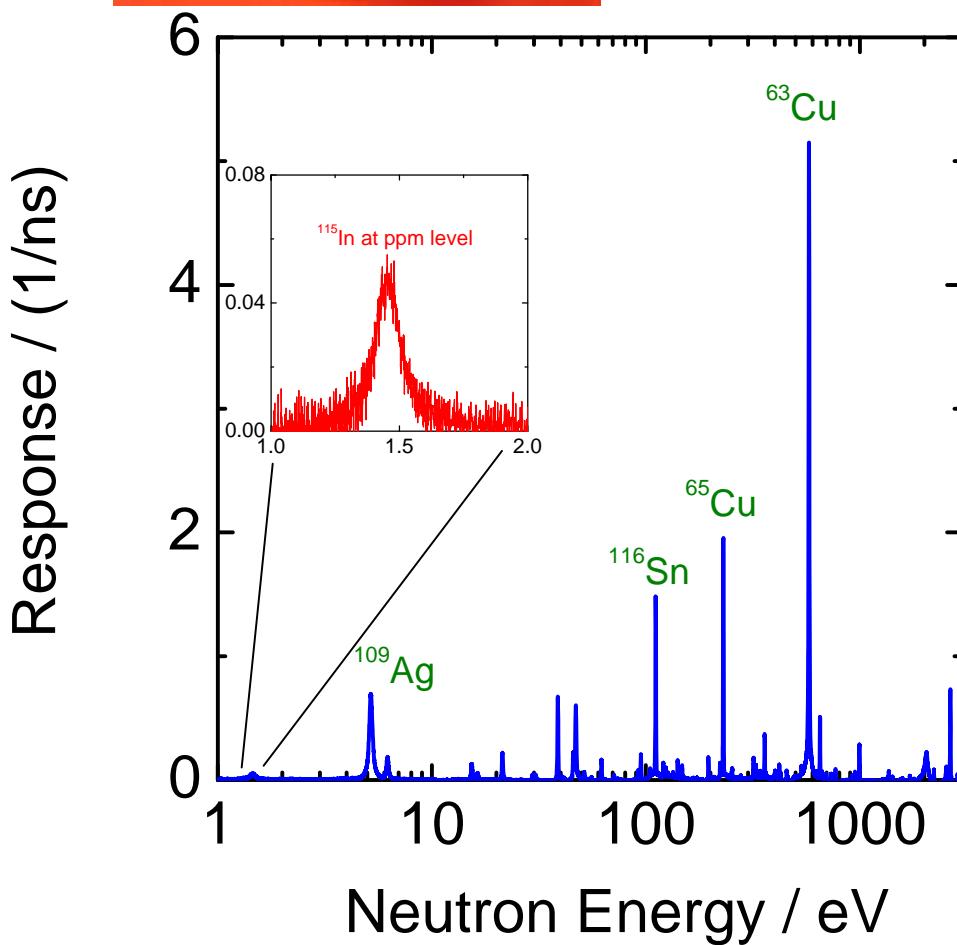
S_ℓ and R^∞ from Dispersive Coupled Channel Optical Model Potential



^{55}Mn Transmission measurements at 50 m

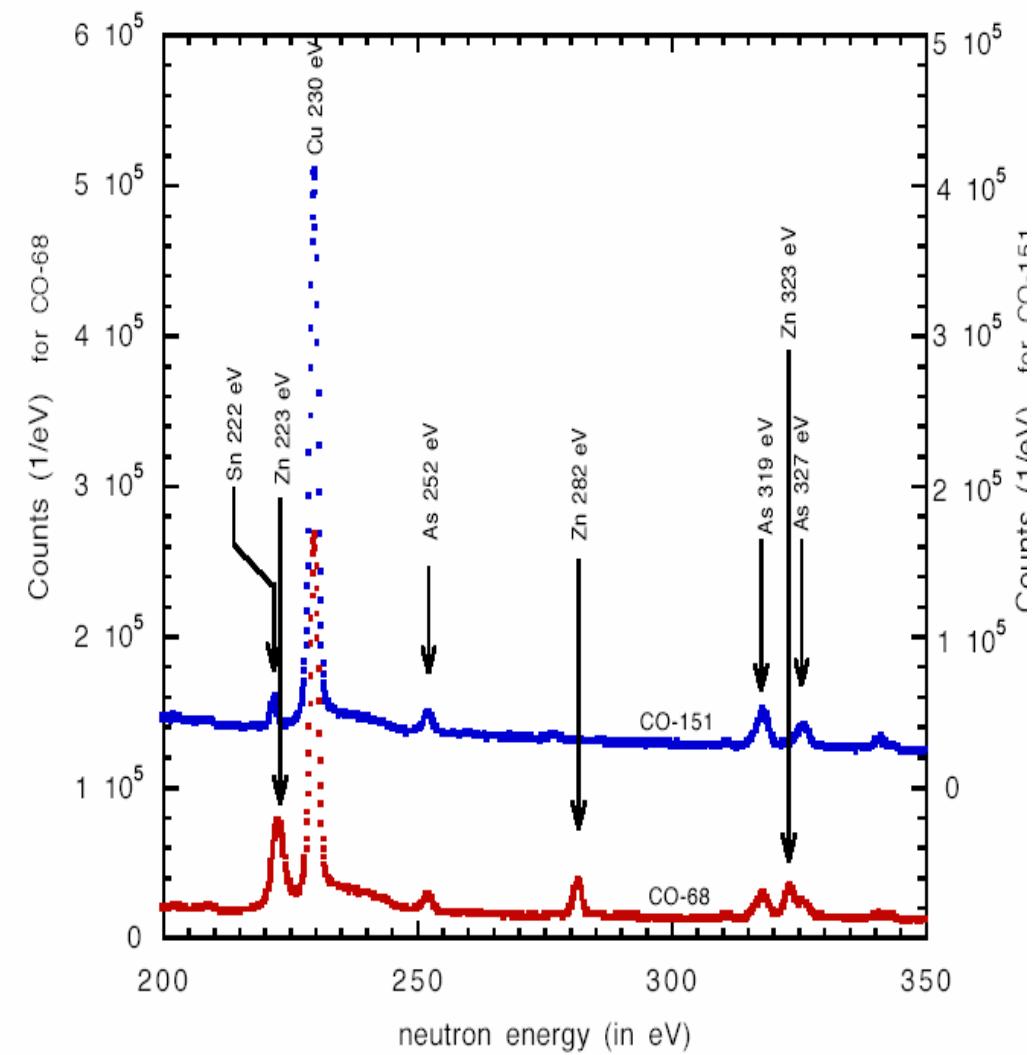


Neutron Resonance Capture Analysis : NRCA

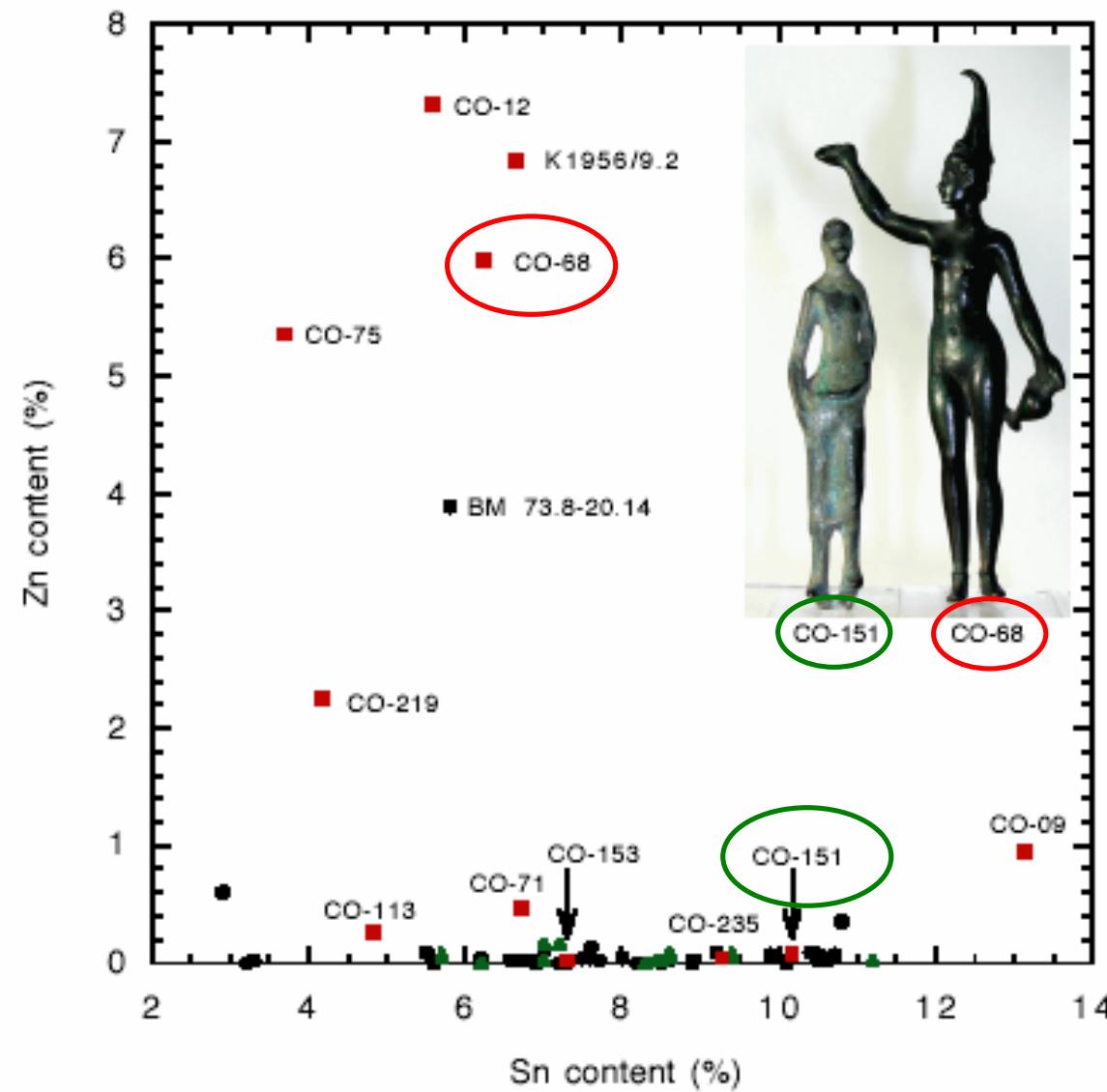


Element	Fractions (%)	Isotope	Resonance (eV)
Cu	77.76 (0.11)	^{63}Cu ^{65}Cu	579.0 230.0
Sn	20.85 (0.10)	^{112}Sn ^{116}Sn ^{117}Sn ^{118}Sn ^{119}Sn ^{120}Sn ^{122}Sn ^{124}Sn	94.8 111.2 38.8 45.7 222.6 427.5 1756.0 62.0
As	0.34 (0.01)	^{75}As	47.0
Sb	0.196 (0.021)	^{121}Sb ^{123}Sb	6.24 21.4
Ag	0.090 (0.01)	^{107}Ag ^{109}Ag	16.3 5.2
Fe	0.770 (0.09)	^{56}Fe	1147.4
In	0.0061 (0.0003)	^{115}In	1.46

NRCA : Authenticity of Etruscan artefacts



NRCA : Authenticity of Etruscan artefacts



Conclusions

- **GELINA : a TOF-facility similar to ORELA for high resolution cross section measurements in the resonance region.**
- **Importance of collaboration between ORNL and IRMM for production and analysis of cross section data in the resonance region.**
- **Progress in measurement capabilities, data reduction and analysis has improved the quality of the data.**
- **Accurate neutron induced cross section data, including full covariance information, can be obtained at these facilities from thermal up to the unresolved resonance region with improved uncertainty levels.**

EC-JRC-IRMM (B)

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