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Joint ICTP-IAEA Workshop on Nuclear Reaction Data for Advanced Reactor Technologies

19 - 30 May 2008

Neutron Induced Capture Cross Section Measurements.

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Neutron Induced Capture

Cross Section Measurements

Workshop on Nuclear Reaction Data for Advanced Reactor Technologies

Trieste, Italy, 19 – 30 May 2008

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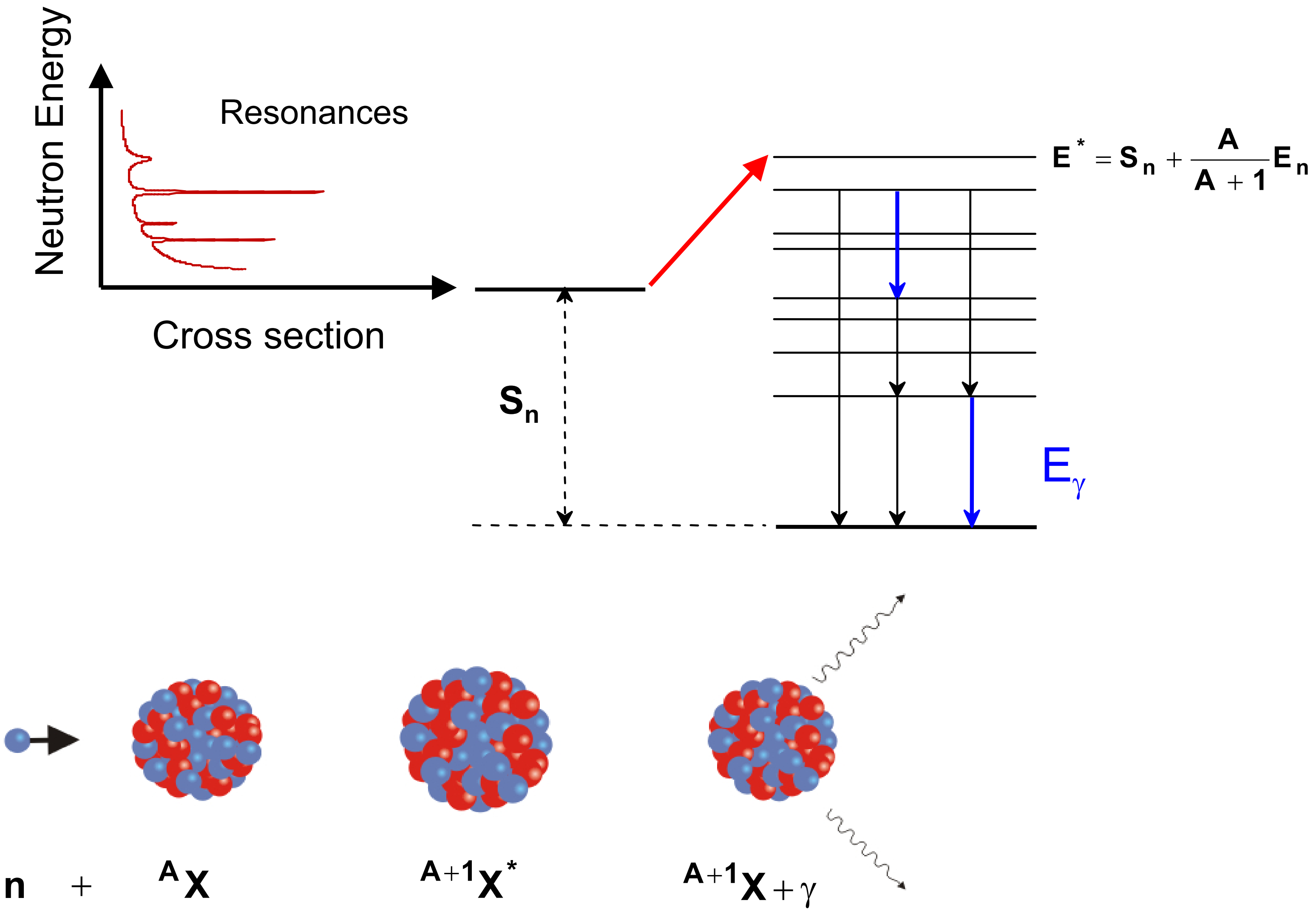
Joint Research Centre (JRC)
IRMM - Institute for Reference Materials and Measurements
Geel - Belgium
http://irmm.jrc.ec.europa.eu/
http://www.jrc.ec.europa.eu/

- **Neutron cross section measurements (Part 1)**
- **Data reduction and uncertainties (Part 2)**
- **Neutron induced capture cross section measurements (Part 3)**
  - Basic principles
  - Spectroscopic measurements (Ge-detectors)
  - Total energy principle with Pulse Height Weighting Technique
  - Normalization + special aspects
  - Applications

    (Unfortunately no reporting of other facilities (see part 1))

Neutron induced capture process



$$E^* = S_n + \frac{A}{A+1} E_n$$

$$n \quad + \quad {}^{A}X \qquad {}^{A+1}X^* \qquad {}^{A+1}X + \gamma$$

Experimental observables

Reaction

$$C_r = \varepsilon_r Y_r A_r \varphi_r$$

- φ_r Neutron fluence rate
- ε_r Detection efficiency
- A_r Effective area
- Y_r Reaction Yield

  Beam fraction undergoing (n,r)

$\Rightarrow$ Complex relation between C_r and Y_r

Y_r related to σ_r

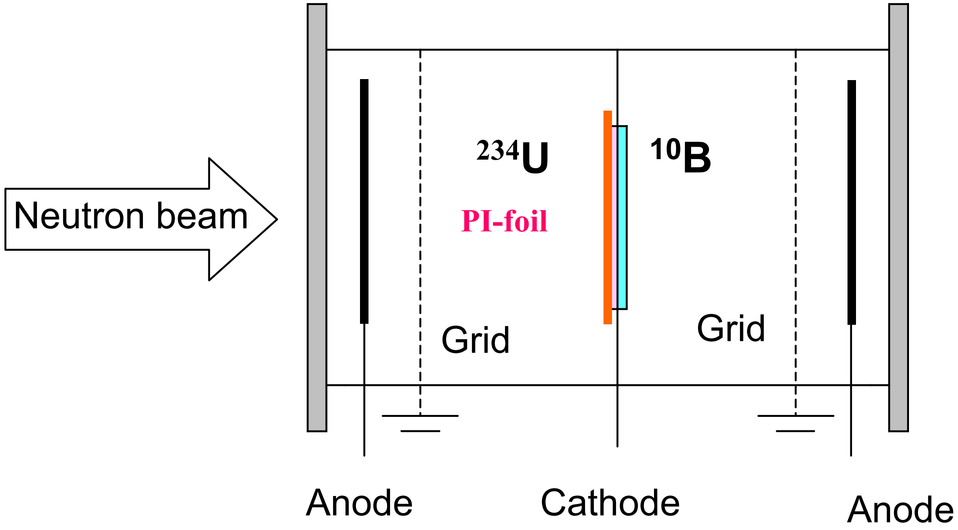
Transmission

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

- Incoming flux cancels
- Detection efficiency cancels
- Good geometry !

$\Rightarrow$ Direct relation between T and σ_{tot}

σ(n,f), σ(n,cp) - measurements



- **Flux** $C_\varphi \cong \varepsilon_\varphi n_\varphi \sigma_\varphi A_\varphi \varphi_\varphi$
- **Fission** $C_f \cong \varepsilon_f n_f \sigma_f A_f \varphi_f$

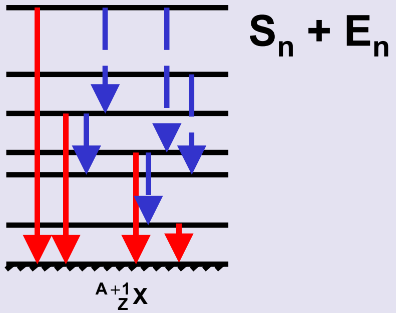
Back to back target $A_\varphi = A_f$

Ionisation chamber $\varepsilon = 100\ \%$

$$\sigma_f \cong \frac{C_f n_\varphi}{C_\varphi n_f} \sigma_\varphi$$

Requirements for Capture Detectors

- **General**
  - (1) ε_c independent of gamma cascade: energy spectrum and multiplicity
  - (2) Good Timing Characteristics
  - (3) Low Neutron Sensitivity



- **Specific applications**
  - (4) Low sensitivity to radioactive decay
  - (5) Distinction between (n,γ) and (n,f) (see N. Colonna)

Capture detectors for $\sigma(n,\gamma)$

- ***Total absorption detectors*** *(N. Colonna)*

  4π & $\varepsilon_\gamma \approx 100\%$

  BaF_2 FzK, DANCE, nTOF
  NaI RPI

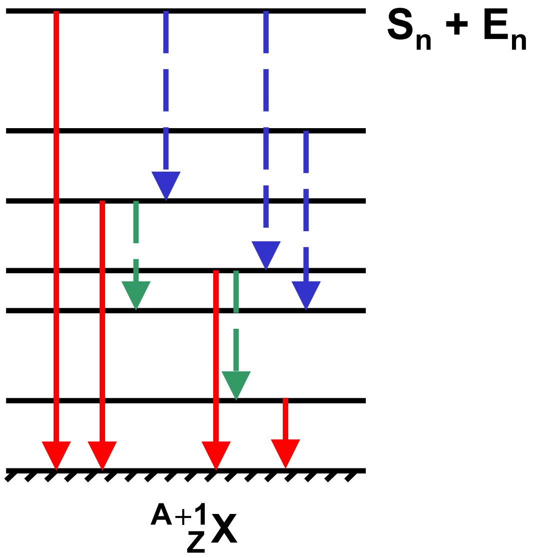
- **Total energy detection systems**

  $\varepsilon_\gamma = kE_\gamma$ and $\varepsilon_\gamma << 1$

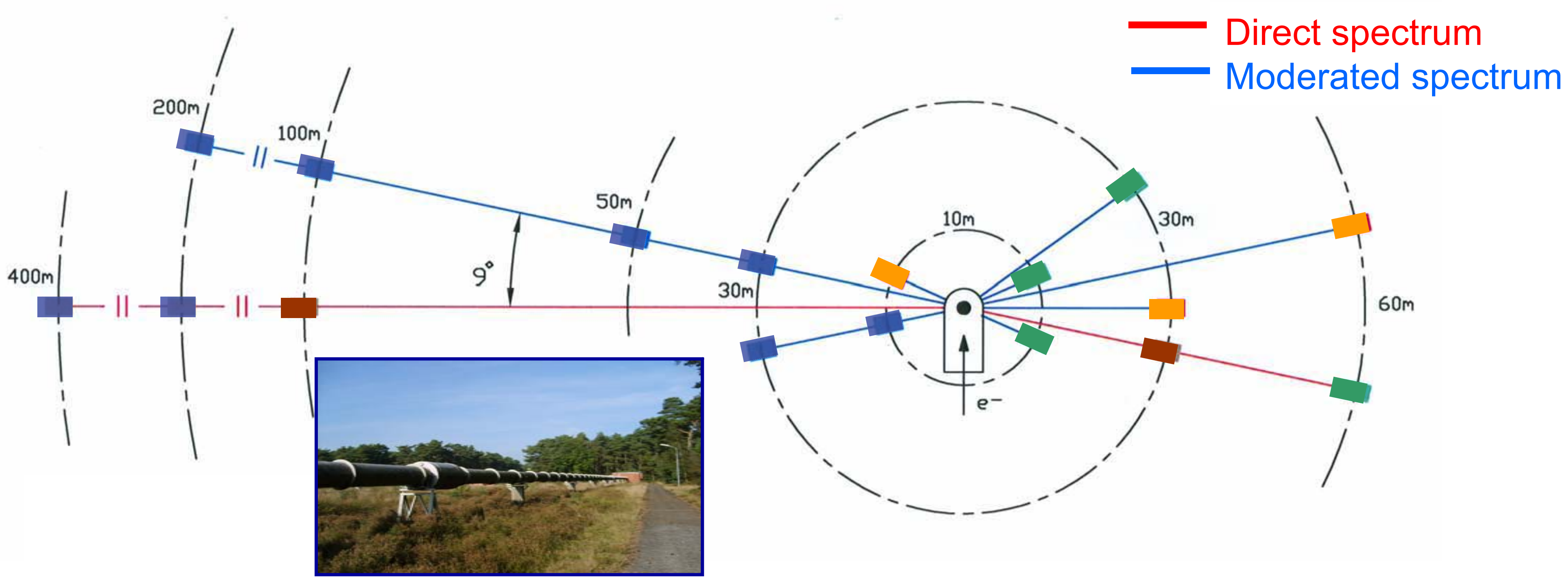
  C_6D_6 GELINA, ORELA, nTOF, KURRI

- **High resolution Ge- detectors**

  $\sigma(n,\gamma)$ only if spectrum is well known
  Special applications



Measurement Stations



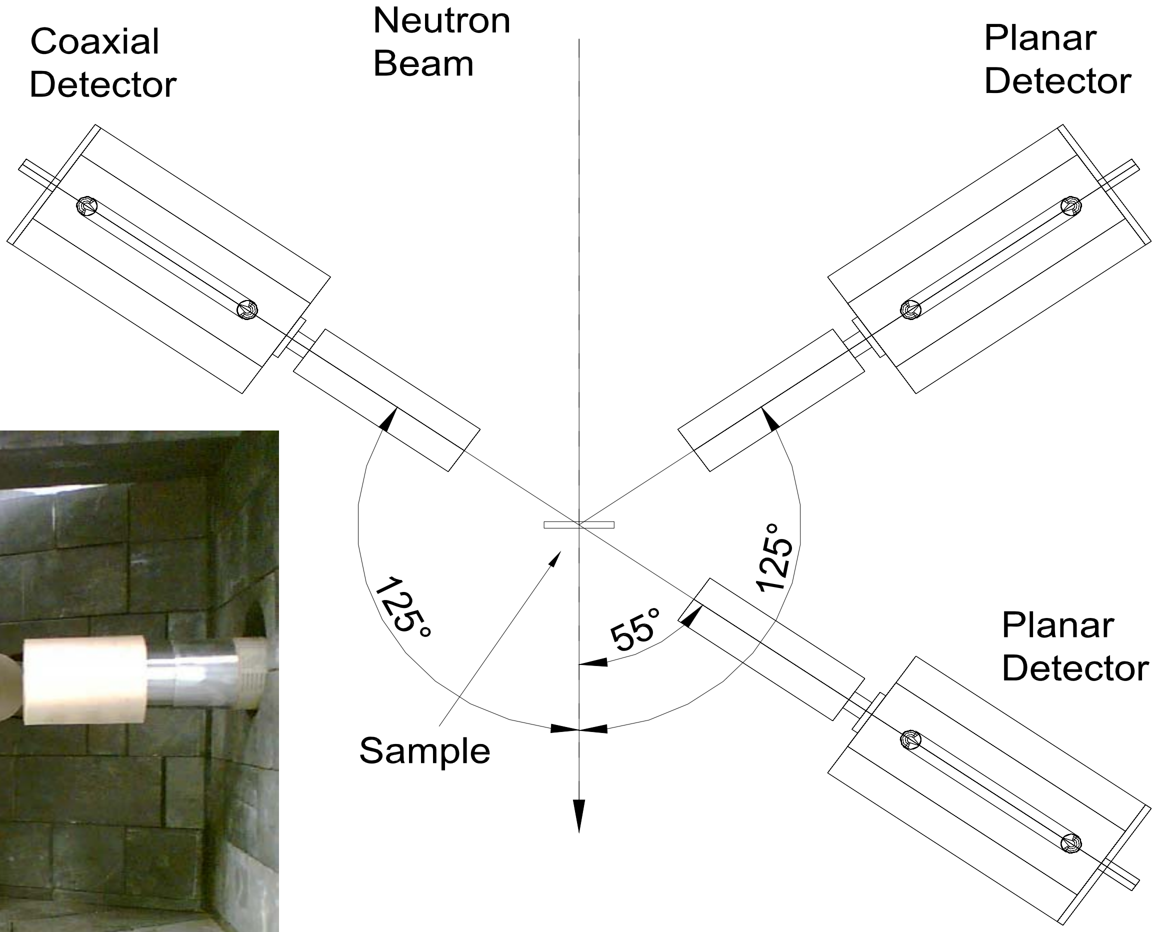
🟧	(n,γ)	NIM A, 577, 626 (2007)
🟦	(n,tot)	NP A 773, 173 (2006)
🟩	(n,f) and (n,cp)	NSE 156, 211 (2007)
🟫	(n,n'γ)	NP A 786, 1 (2007)

- **Ge-detectors (L = 10 m)**
  - Spin and parity PR C 61, 054616 (2000)
  - Partial cross sections & branching ratio
  - Isotope identification
- **C_6D_6 detectors (L = 10, 30 and 60 m)**
  - Parameterisation of σ(n,γ)
  - NRCA (Ancient Charm)

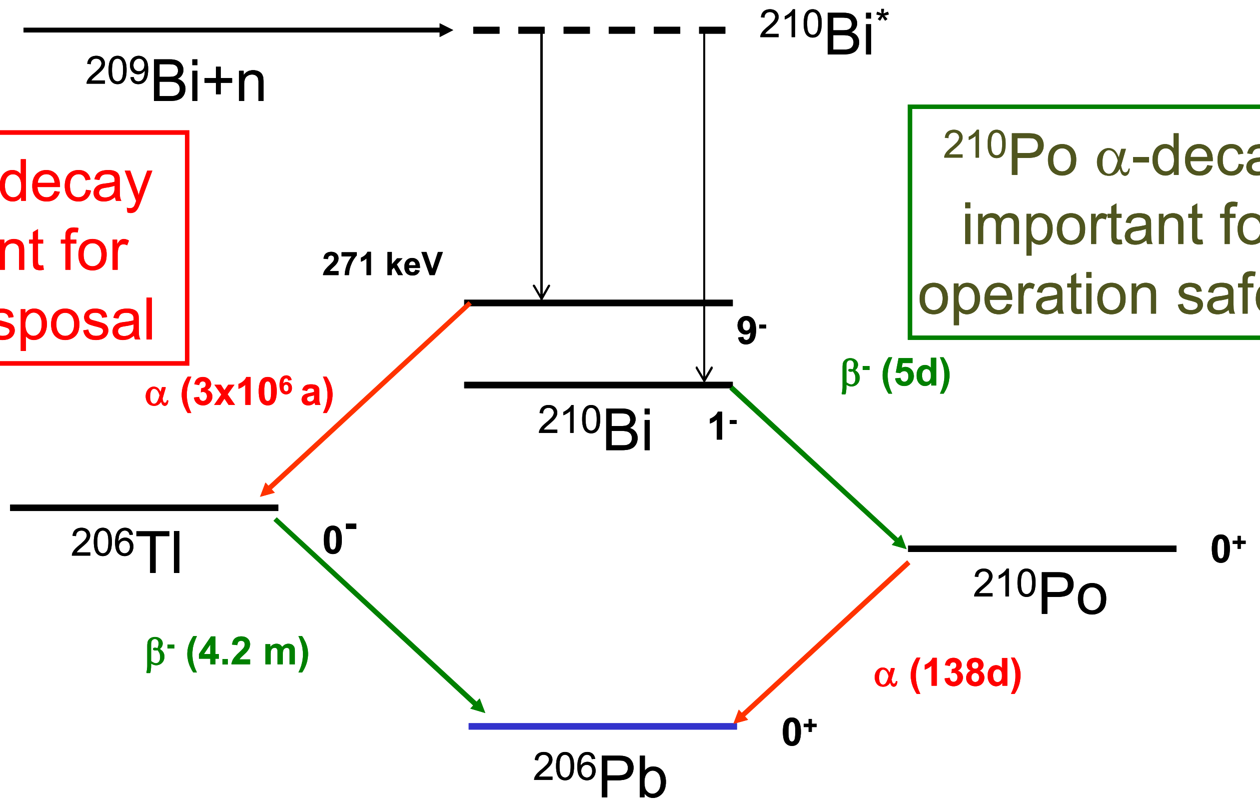
Gamma spectroscopic measurements

- **Partial capture cross sections and branching ratio**
- **Isotope identification**
- **Spin and parity assignment (PR C 61, 054616 (2000))**
- *For $\sigma(n,\gamma)$ in resonance region*
  Only if level-scheme is well-known
  - Neutron sensitivity of Ge-detectors
  - Time resolution

Spectroscopic measurements Ge-detectors at L = 10 m



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



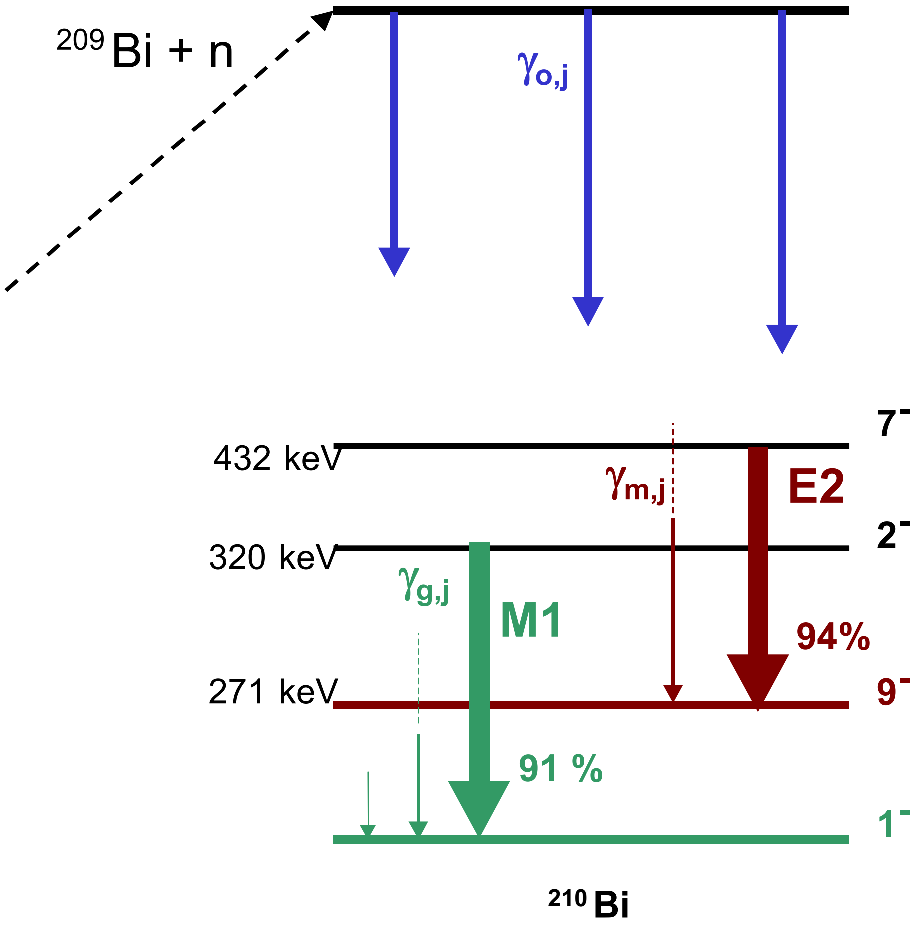
^{210m}Bi α-decay important for waste disposal

^{210}Po α-decay important for operation safety

Important for any system using Bi as coolant, moderator or target (ADS)

Borella et al., JRNC 265, 267 (2005) + ND2007

Principle



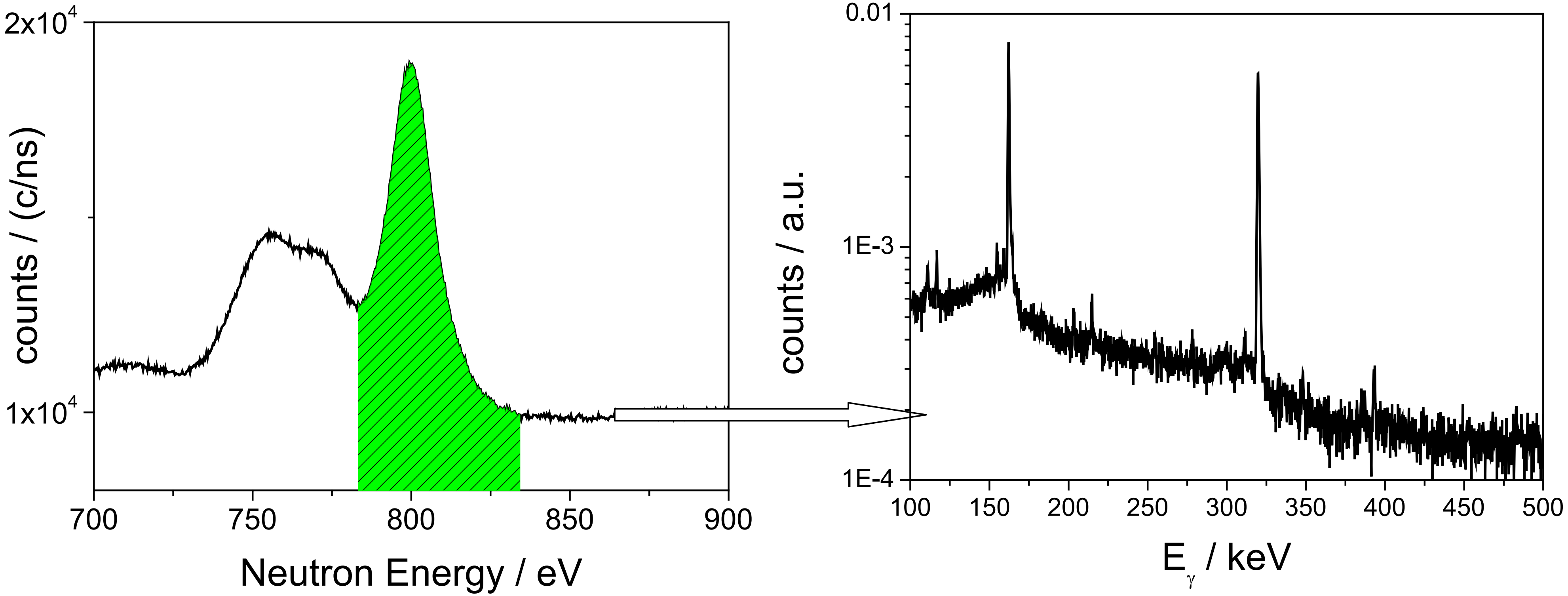
$$\sigma_{m+g} = \sum_j \sigma_{\gamma o,j}$$

$$\sigma_m = \sum_j \sigma_{\gamma m,j}$$

$$\sigma_g = \sum_j \sigma_{\gamma g,j}$$

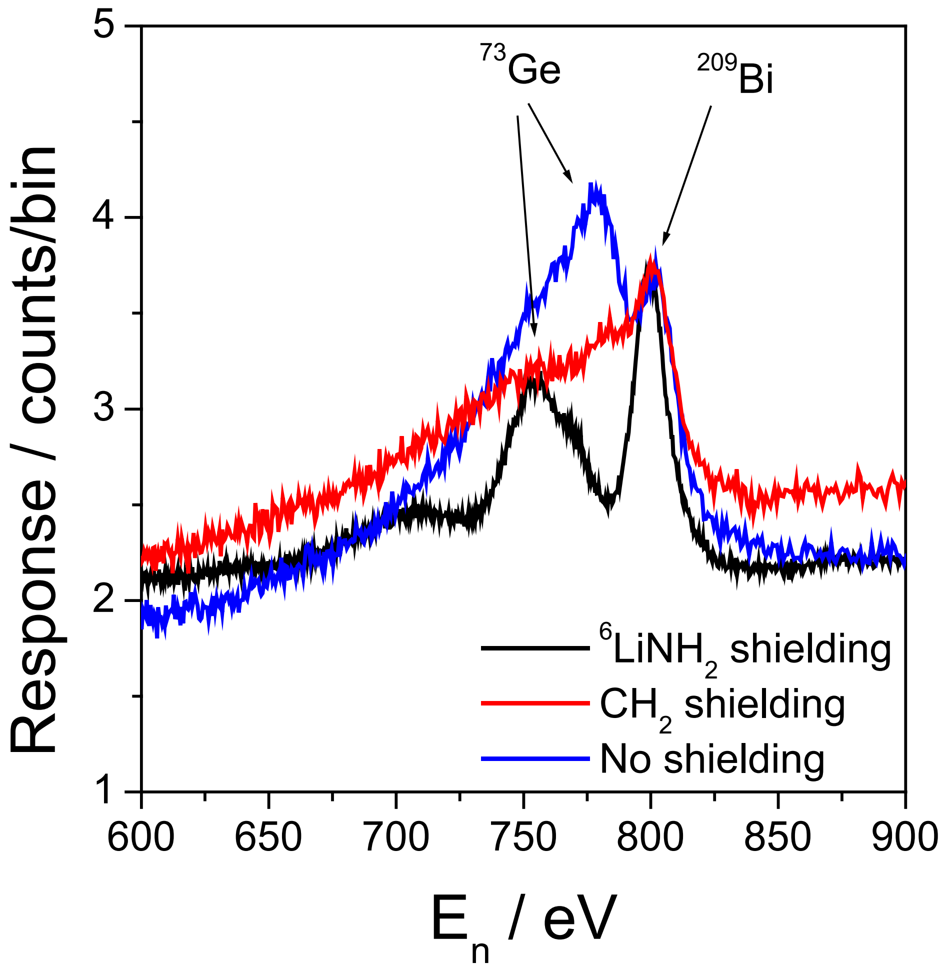
Measurements at GELINA

- **HPGe detectors**
  - 1 Coaxial (E_γ up to 8 MeV), 2 Planar (E_γ from 100 keV to 1.5 MeV)
- **Sample**
  - Metallic Bi 99.99%, ∅ 80 mm, thickness 1.26 mm (4.38×10^{-3} at/b)
- **TOF selection ⇒ γ -ray spectra for a given energy region**

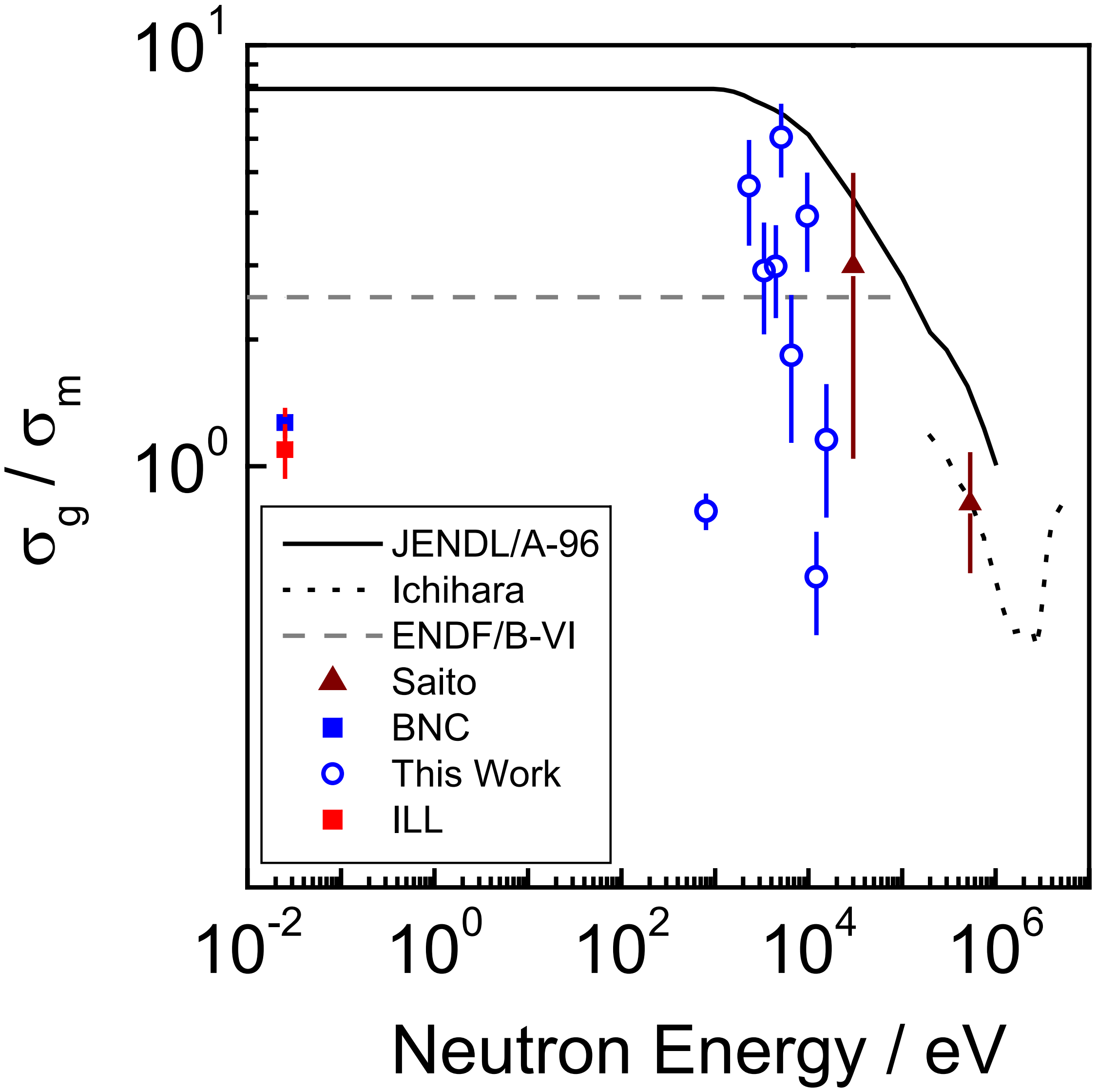


Measurements at GELINA

- **Difficulties**
  - Very low cross section (σ_{g+m} ~ 40 mb at E_{th})
  - Very high scattering to capture ratio (~10^3)
  - Neutron sensitivity of Ge detectors
- **Choice of a short FP**
  - Limits energy range to 20 keV

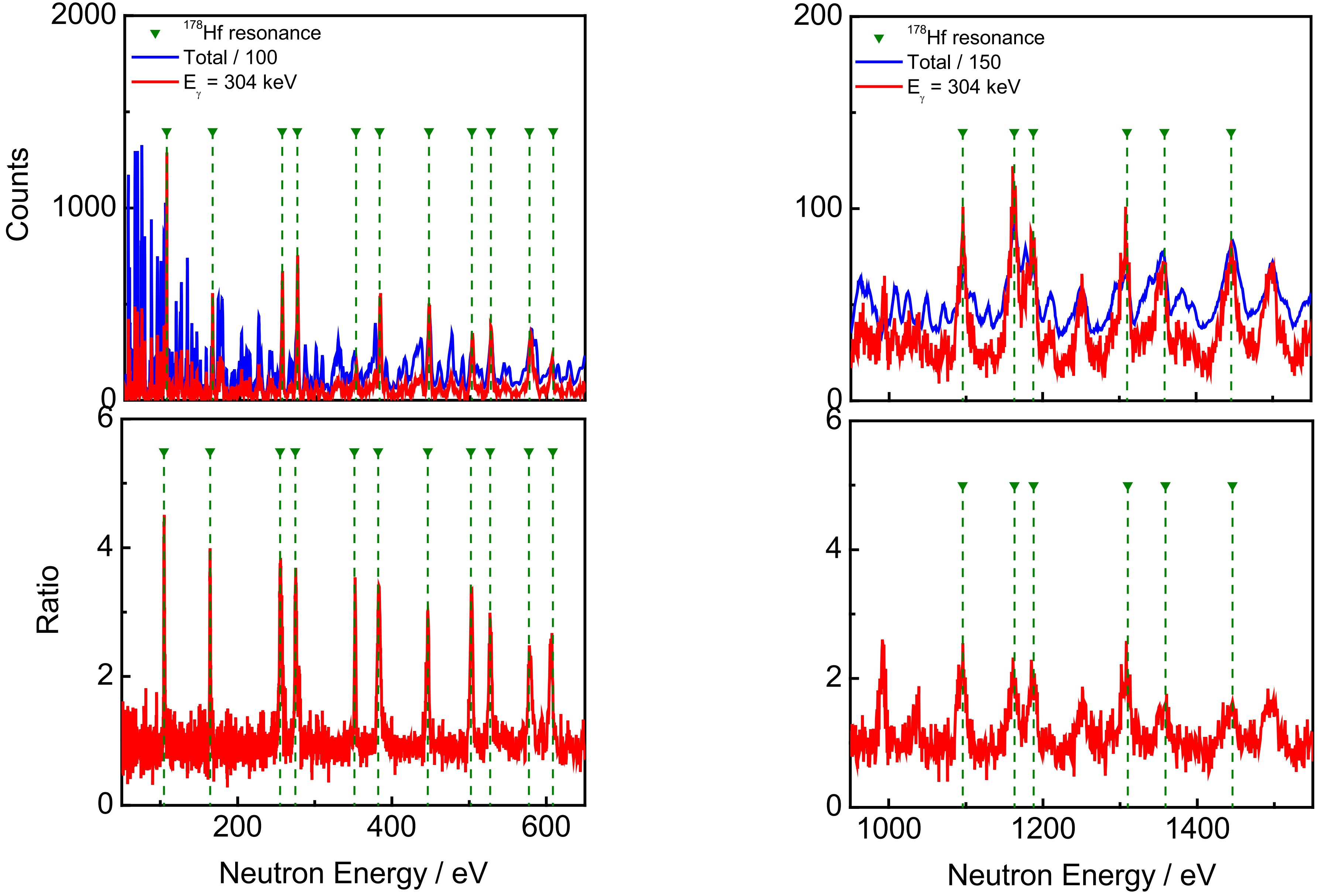


Preliminary results

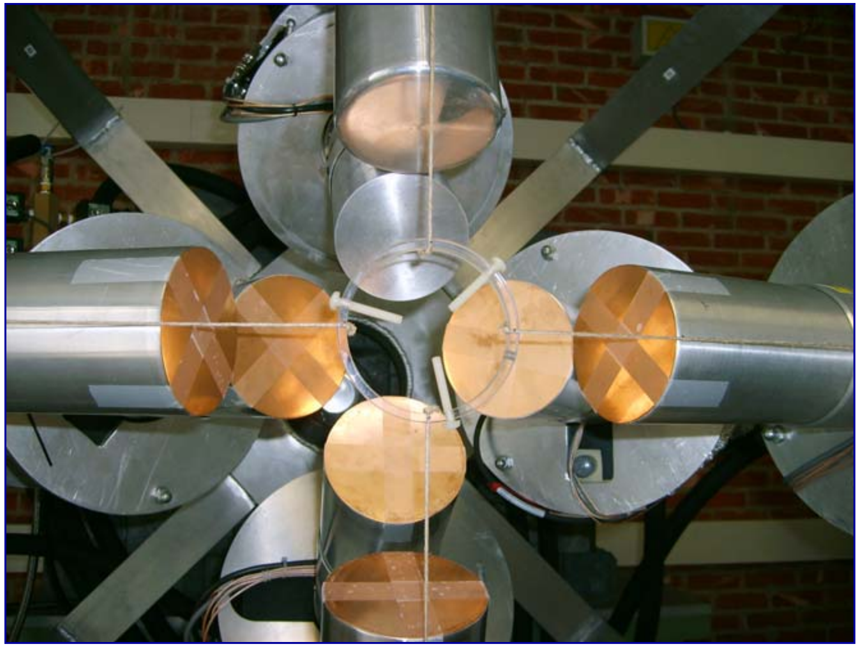


Borella et al., Nice ND2007

Isotope identification, example ^{nat}Hf



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



2001-2006 : 2 – 4 HPGe 100 %

^{56}Ni, ^{52}Cr, ^{209}Bi, $^{206, 207, 208}$Pb

2006 – 2007 : 8 HPGe (100 %)

^{56}Fe, ^{28}Si

Mihailescu et al., NIM A578, 298 (2007)

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

Capture experiments

- **Basic principles**
- **Spectroscopic measurements**

- **Total energy principle with PHWT**

- **Normalization + special aspects**
- **Applications**

Total energy detection principle
$\varepsilon_\gamma << 1$ and $\varepsilon_\gamma = k\,E_\gamma$

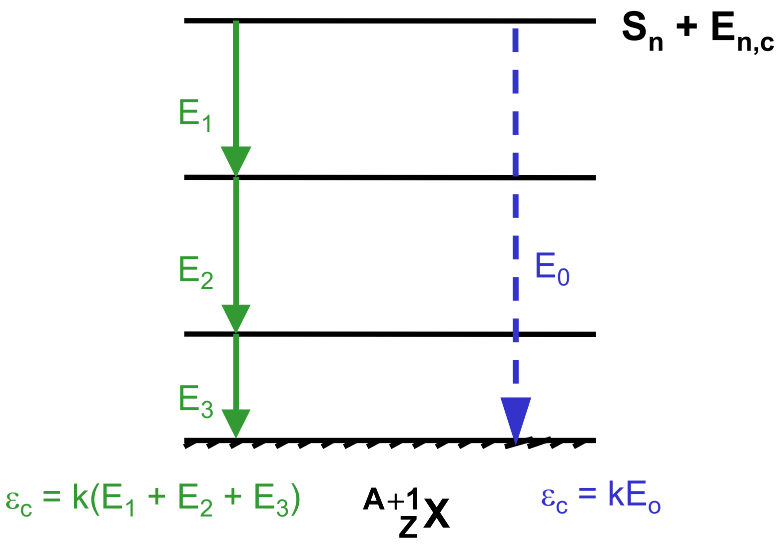
- **Detection efficiency: $\varepsilon_\gamma << 1$**

$$\varepsilon_c = 1 - \prod_i \left(1 - \varepsilon_{\gamma,i}\right)$$

with $\varepsilon_\gamma << 1$

$$\varepsilon_c \approx \sum_i \varepsilon_{\gamma,i}$$

- **Detection efficiency: $\varepsilon_\gamma = k\,E_\gamma$**



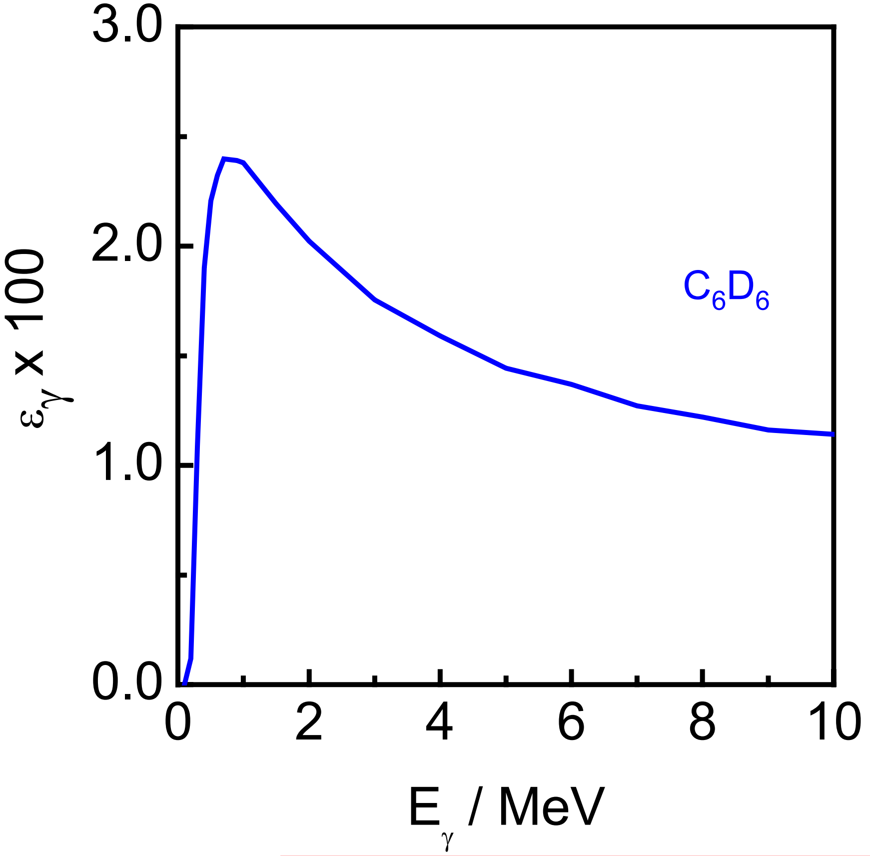
⇒

$$\varepsilon_c \approx \sum_i \varepsilon_{\gamma,i} = k \sum_i E_{\gamma,i} \approx k\,(\,S_n + E_{n,c}\,)$$

independent of cascade

Moxon and Rae, Nucl. Instr. & Meth. 24 (1963) 445

Total energy detection with C_6D_6



- $\varepsilon_\gamma << 1$
- $\varepsilon_\gamma \not\propto E_\gamma$

$\varepsilon_\gamma \not\propto E_\gamma \quad \Rightarrow$ **Pulse Height Weighting Technique (PHWT)**

(Maier – Leibnitz)

Macklin and Gibbons, Phys. Rev. 159 (1967) 1007

Total energy detection with C_6D_6 in combination with PHWT

Pulse Height Weighting Technique (PHWT)

$$\int \mathbf{R(E_d, E_\gamma)\, WF(E_d)\, dE_d} = \mathbf{kE_\gamma}$$

$R(E_d,E_\gamma)$ is response function of detector, with $\int R(E_d, E_\gamma)\, dE_d = \varepsilon_\gamma$

For each event we record:

- time-of-flight T_n
- energy deposited in detector E_d

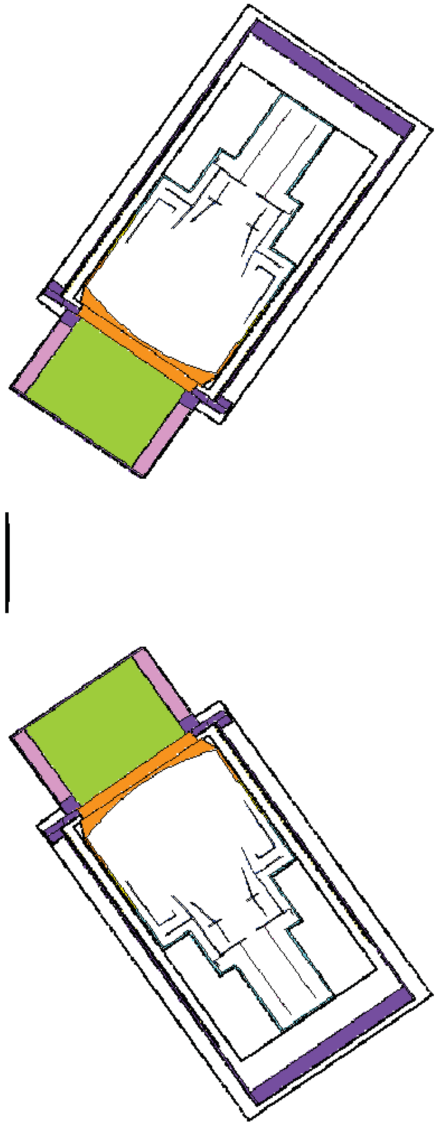
$$\Rightarrow \quad C_w(T_n) = \int C_c(T_n, E_d)\, WF(E_d)\, dE_d$$

R(E_d , E_γ) : by MCNP (Geometry)

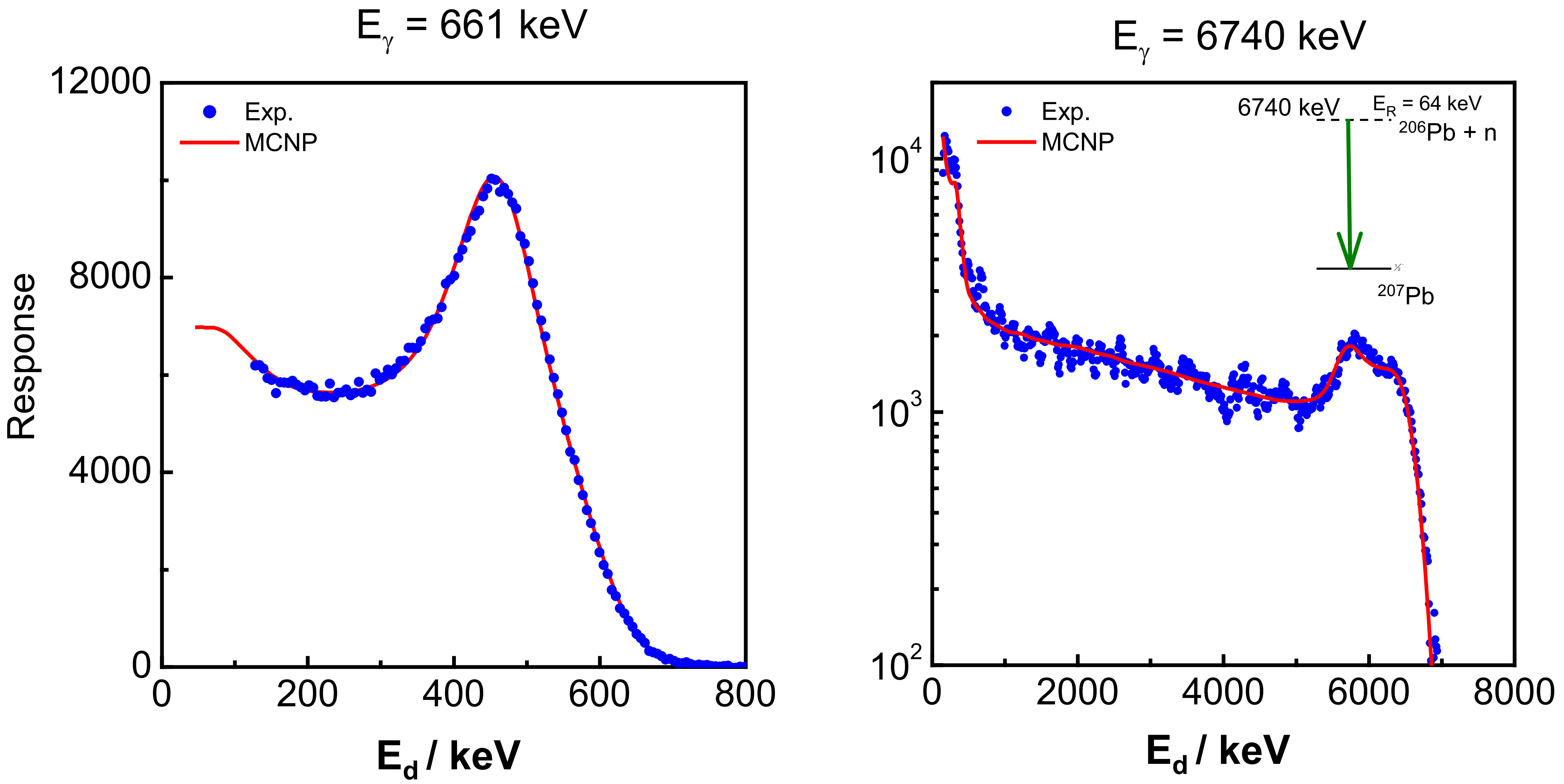
Careful description of the geometry:

- Detectors (C_6D_6)
- Aluminium canning
- Quartz window (boron free)
- Photomultipliers + glass envelope (Si)
- Electromagnetic insulation (μ - metal)
- Expansion tube (TEFLON)

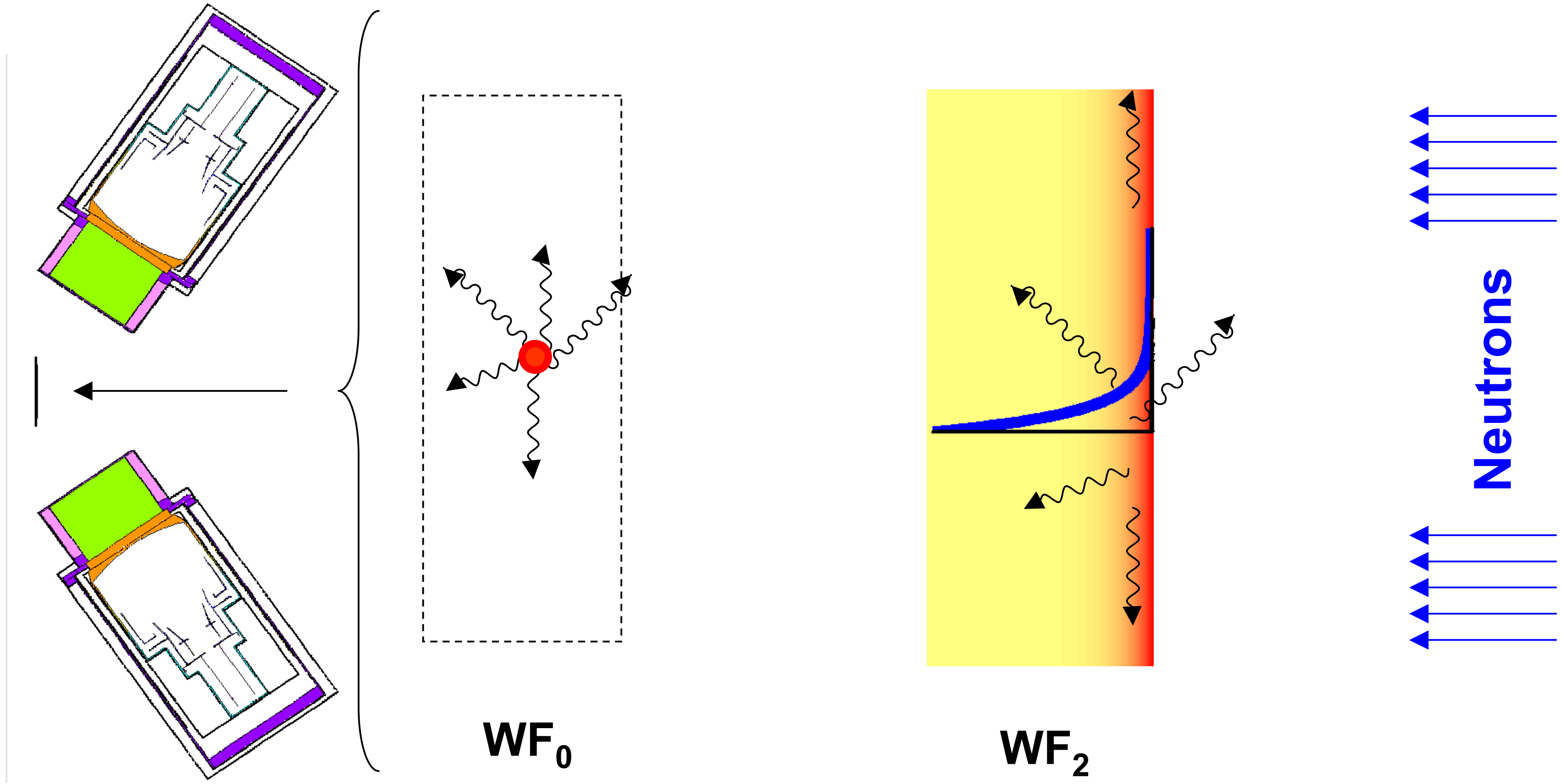
For $\sigma(n,\gamma)$ measurements: + sample



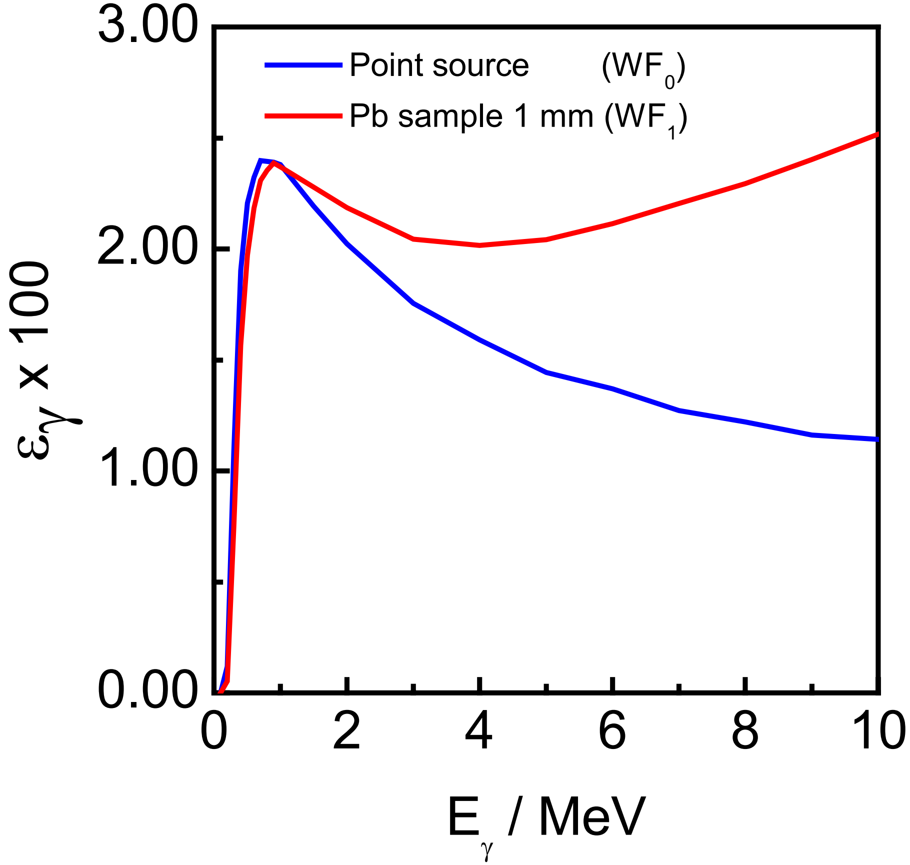
C_6D_6 Response by MCNP

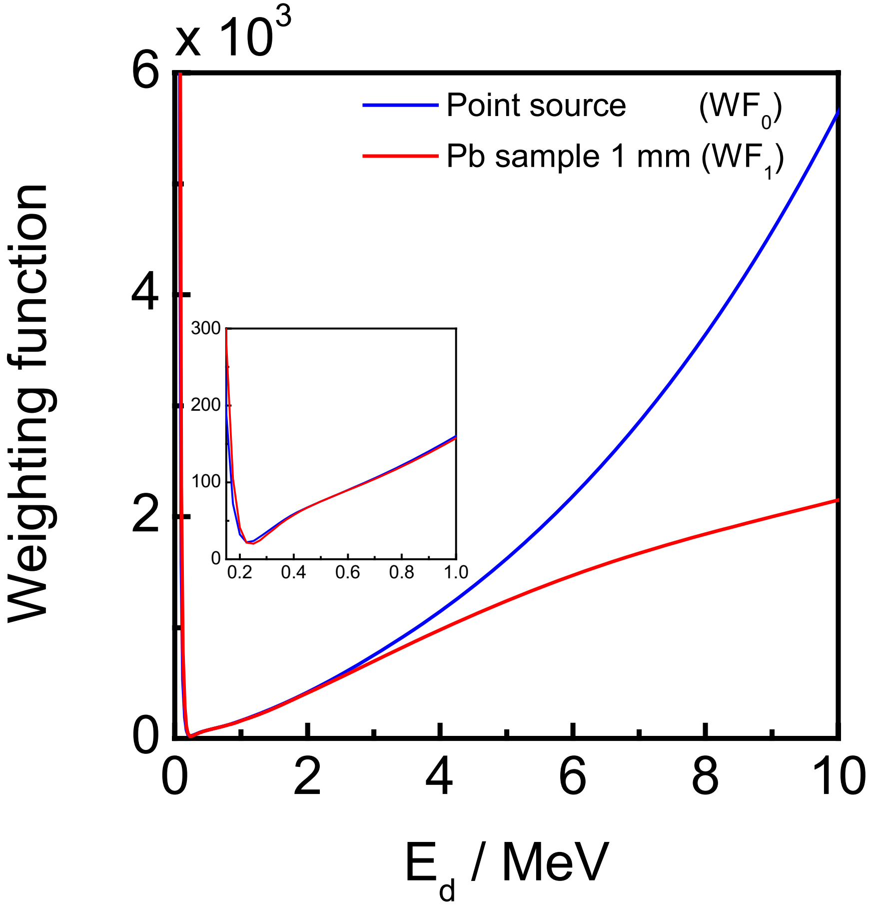


WF – gamma transport in sample + detector



Examples of WF





$$kE_\gamma = \int_{E_{DL}} R(E_m, E_\gamma)\, W(E_m)\, dE_m$$

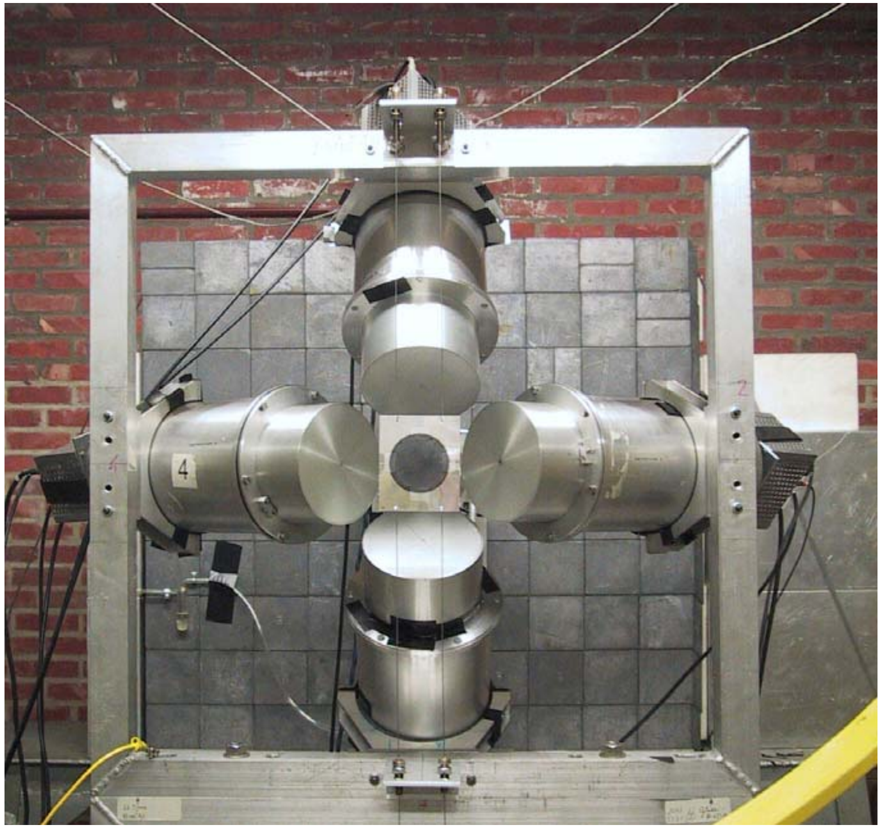
$$C_w = \int_{E_{DL}} C_c(E_m)\, W(E_m)\, dE_m$$

Capture cross section measurements at GELINA

Total energy detection

- **C_6D_6 liquid scintillators**
  - 125^o
  - PHWT
- **Flux measurements (IC)**
  - $^{10}B(n,\alpha)$
  - $^{235}U(n,f)$

L = 10 m, 30 m and 60 m



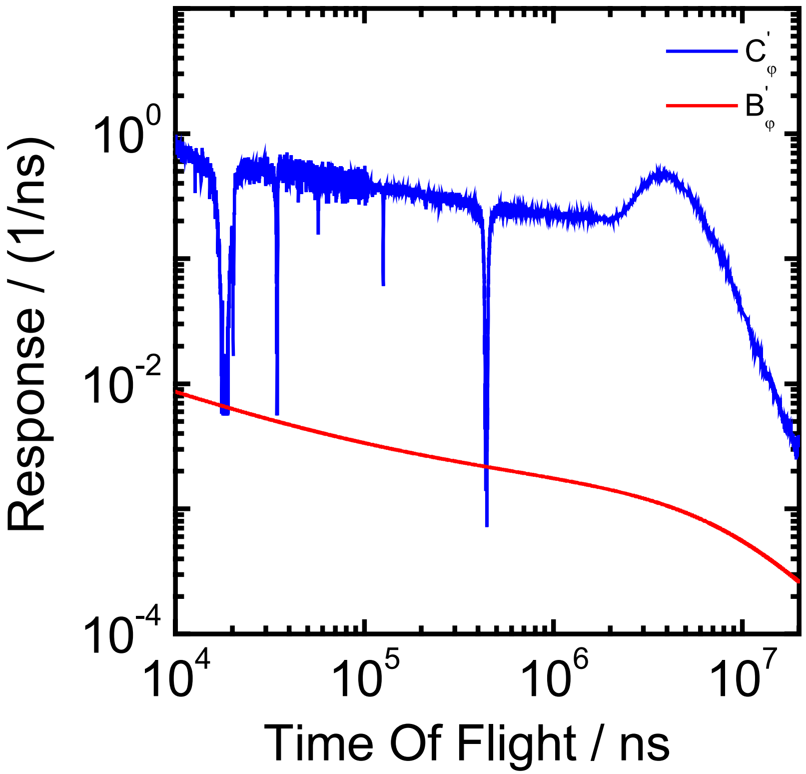
$$Y_{exp} = N\sigma_{\varphi}\frac{C_w - B_w}{C_{\varphi} - B_{\varphi}}$$

WF : from MC simulations

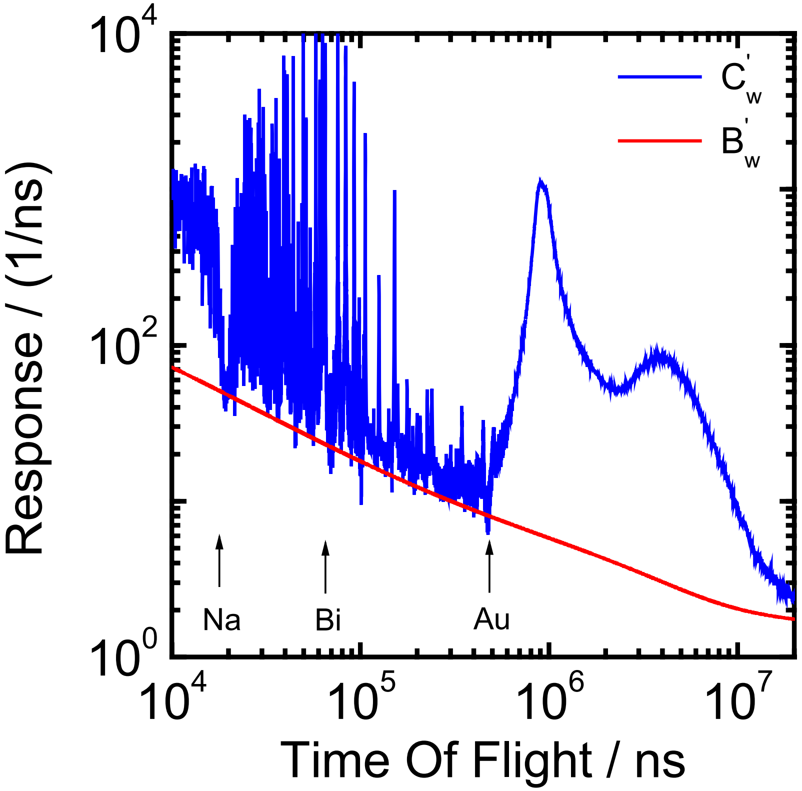
$$C_w(T_n) = \int C_c(T_n, E_d)\, WF(E_d)\, dE_d$$

$\sigma(n,\gamma)$ measurements e.g. $^{103}Rh(n,\gamma)$

Flux measurement



Capture measurement



$$Y_{exp} \propto \sigma_\varphi \frac{C'_w - B'_w}{C'_\varphi - B'_\varphi}$$

Capture experiments

- **Basic principles**
- **Spectroscopic measurements**
- **Total energy principle with PHWT**

- **Normalization + special aspects**

- **Applications**

Normalization

- ***Standard cross sections***
  - $\sigma(n_{th},\gamma)$
  - $^{197}Au(n,\gamma)$ *for* $E_n > 200$ *keV*

- **Well - known resonance**

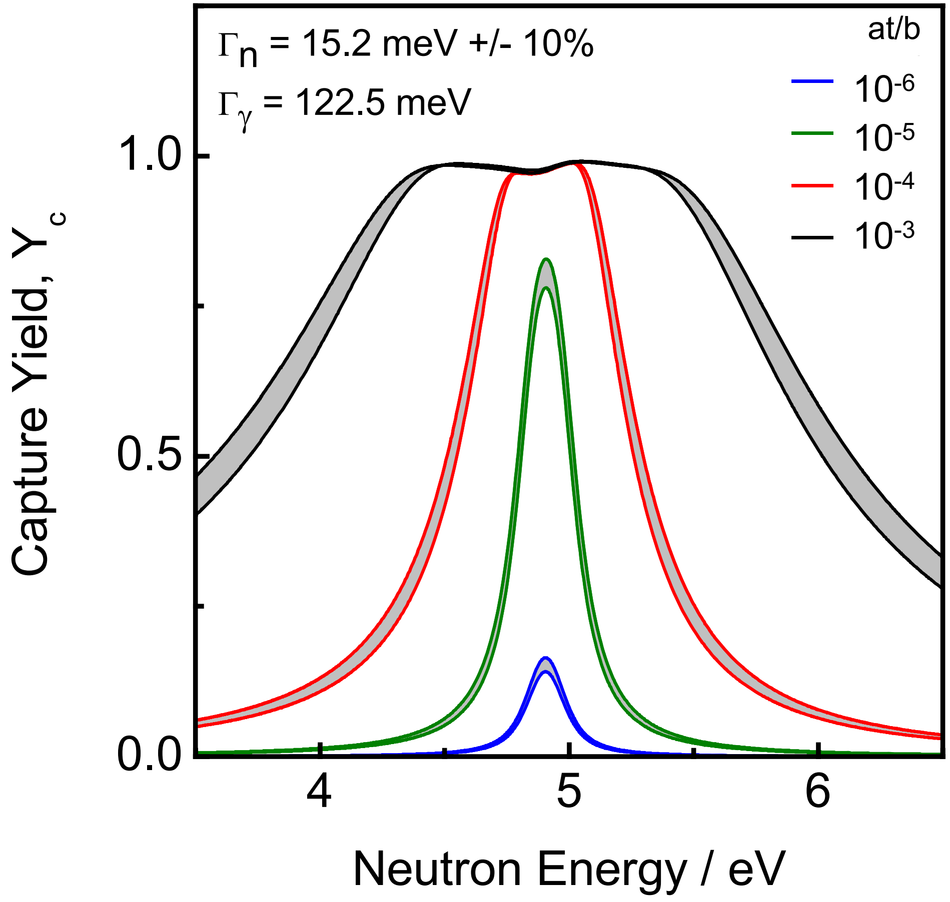
  (with $\Gamma_n << \Gamma_\gamma \Rightarrow Y_c \propto g\Gamma_n$ and $g\Gamma_n$ from transmission)
  - $^{56}Fe(n,\gamma)$ $E_r = 1.15$ keV

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

- **Saturated resonance**
  - $n\sigma_t >> 1 \quad \Rightarrow Y_c \sim (\sigma_\gamma / \sigma_t)$
  - $\Gamma_n << \Gamma_\gamma \quad \Rightarrow Y_c \sim 1$ almost independent of resonance parameters

    e.g. $^{197}Au(n,\gamma)$ at 4.9 eV and $^{109}Ag(n,\gamma)$ at 5.2 eV

Normalization: saturated resonance



^{197}Au(n,γ) : E_r = 4.9 eV

For $n\sigma_t \gg 1$ and $\Gamma_n \ll \Gamma_\gamma$

$\Rightarrow Y_c \sim 1$

Reduction of systematic effects due to:

- **target thickness of reference sample**
- **nuclear data**

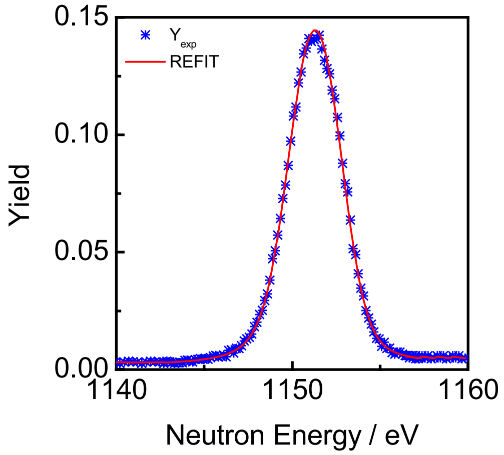
$$Y_{exp} = N\sigma_\varphi \frac{C'_w - B'_w}{C'_\varphi - B'_\varphi}$$

σ_φ : only the relative energy dependence is required
$\Rightarrow$ ^{10}B(n,α) which behaves ~ 1/v

^{56}Fe(n,γ) Γ_n for E_r = 1.15keV

Sample	Fe g/cm^2	X	∅ mm	WF$_0$ Γ_n / meV	WF$_2$ Γ_n / meV
Fe1	0.105		60	53.3 (1.1)	62.6 (1.3)
Fe2	0.394		60	58.0 (1.0)	62.5 (1.1)
Fe3	0.905		60	57.9 (1.0)	60.2 (1.0)
Fe^{206}Pb *	0.394	1.213	60	62.4 (1.0)	63.1 (1.1)
FePb*	0.422	1.103	60	63.1 (1.1)	62.6 (1.1)
FePb*	0.422	2.725	60	59.2 (1.1)	62.6 (1.1)
Fe4	0.202		80	55.4 (1.0)	61.2 (1.1)
Fe5	0.795		80	60.6 (1.1)	60.3 (1.1)
Fe6	0.998		80	61.2 (1.1)	61.2 (1.1)
FeAu	1.708	0.118	80	62.9 (1.1)	61.3 (1.1)
Fe_2O_3	1.404	0.603	80	55.8 (1.0)	59.1 (1.0)
Mean				**59.1**	**61.5**
Std				**3.3**	**1.3**
Std (%)				**5.6**	**2.1**

* Sandwich



Γ_γ = 574 meV

Uncertainties of 2% can be reached

Reference value
(transmission)
Γ_n = 61.7 (0.9) meV

WF : systematic bias effects

- **Count losses due to discriminator level**
- **Imperfect weighting due to internal conversion process**
- **Detection of coincident events**
- **Deviations from linearity ($\varepsilon_\gamma = kE_\gamma$)**
- **Incorrect energy calibration**

Wilson et al. : NIM A511 (2003) 388

nTOF collaboration : NIM A521 (2004) 454

The impact of these effects can be reduced (avoided) by:

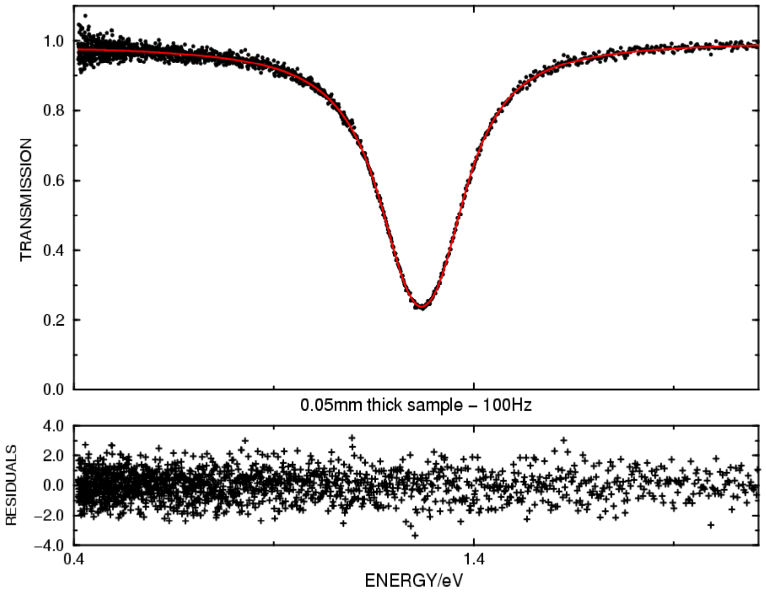
⇒ normalizing at a reference σ_γ with a similar γ - ray cascade

⇒ + internal normalization: identical experimental conditions

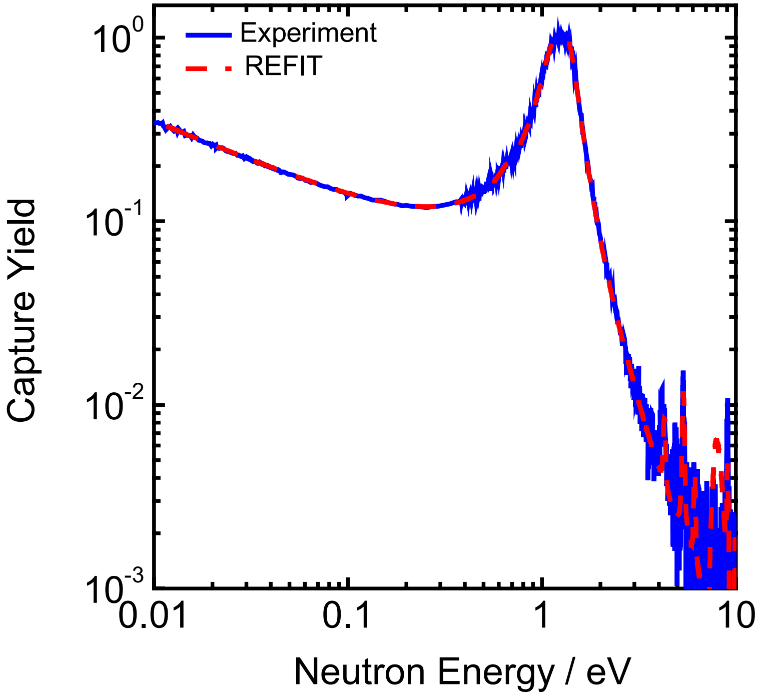
⇒ Resonance with $\Gamma_n << \Gamma_\gamma$: saturated or transmission + capture

Borella et al. Nucl. Instr. Meth. A577, (2007) 626

Internal normalization : ^{103}Rh(n,γ)



REFIT
RP + N
⇔

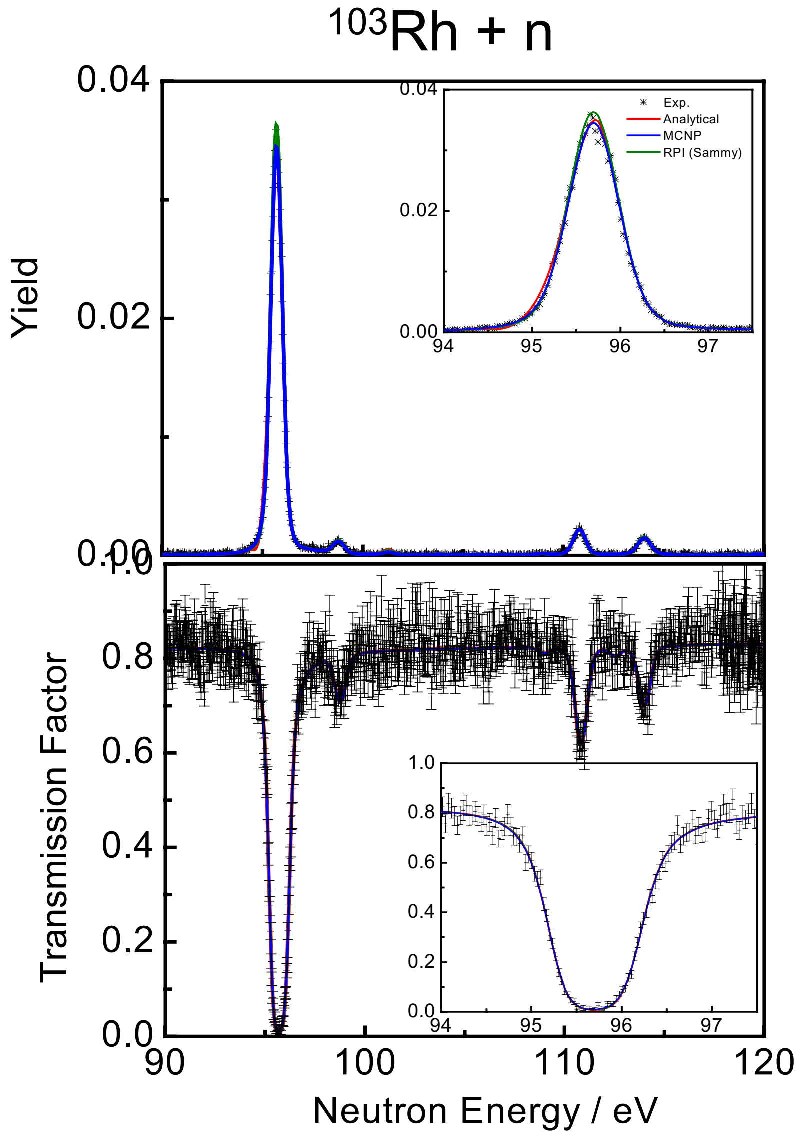


E_r = 1.260 eV
Γ_n = 0.464 meV
Γ_γ = 156.0 meV
g = 3/4

σ(n,γ) at 25.3 meV

WF : 142.0 (1.5) b
1.0 % normalization
0.5 % uncorrelated
Counts : 143.3 b

Impact of the resolution function on RP



Resolution Function (RF)	**E_0 = 95.7 eV Γ_n / meV**
Capture	
Analytical (REFIT)	**2.47 (5)**
MCNP	**2.42 (5)**
RPI (fit MCNP RF)	**2.50 (5)**
Transmission	
Analytical (REFIT)	**2.40 (5)**
MCNP	**2.42 (5)**
RPI (fit MCNP RF)	**2.40 (5)**

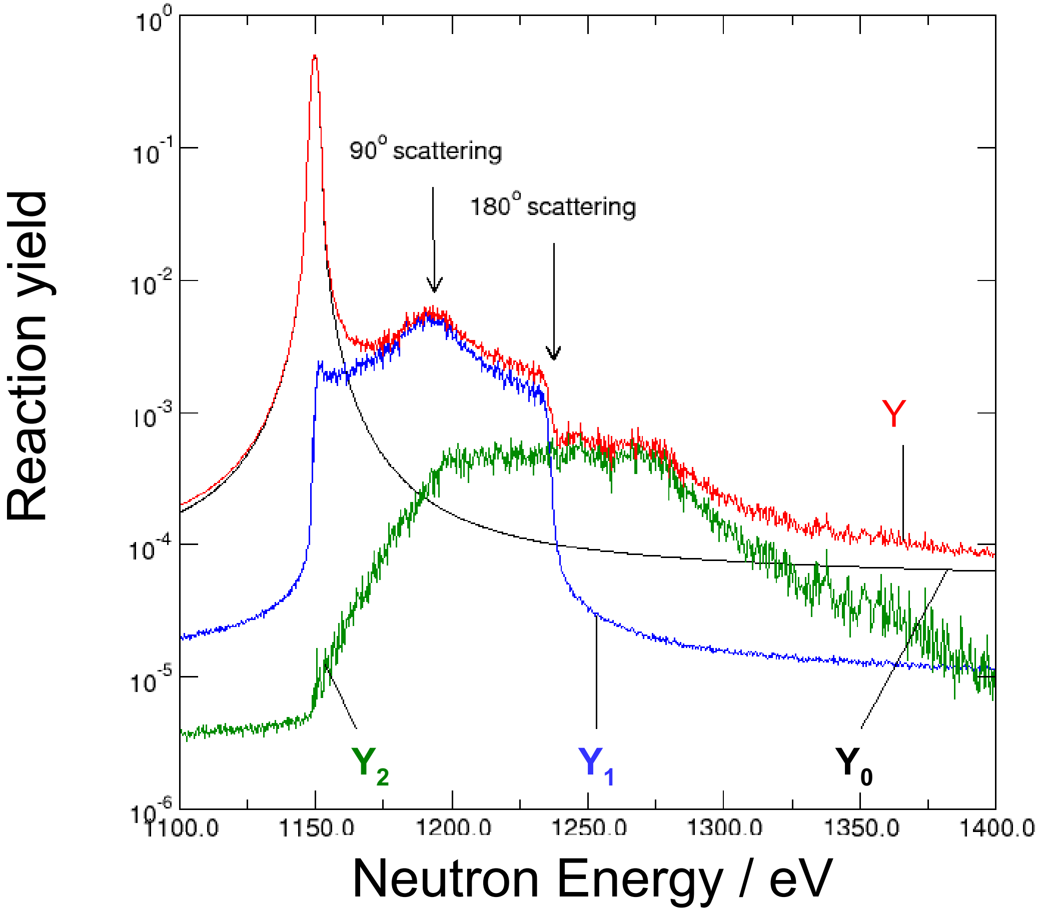
⇒ RF has an impact on RP deduced from RSA

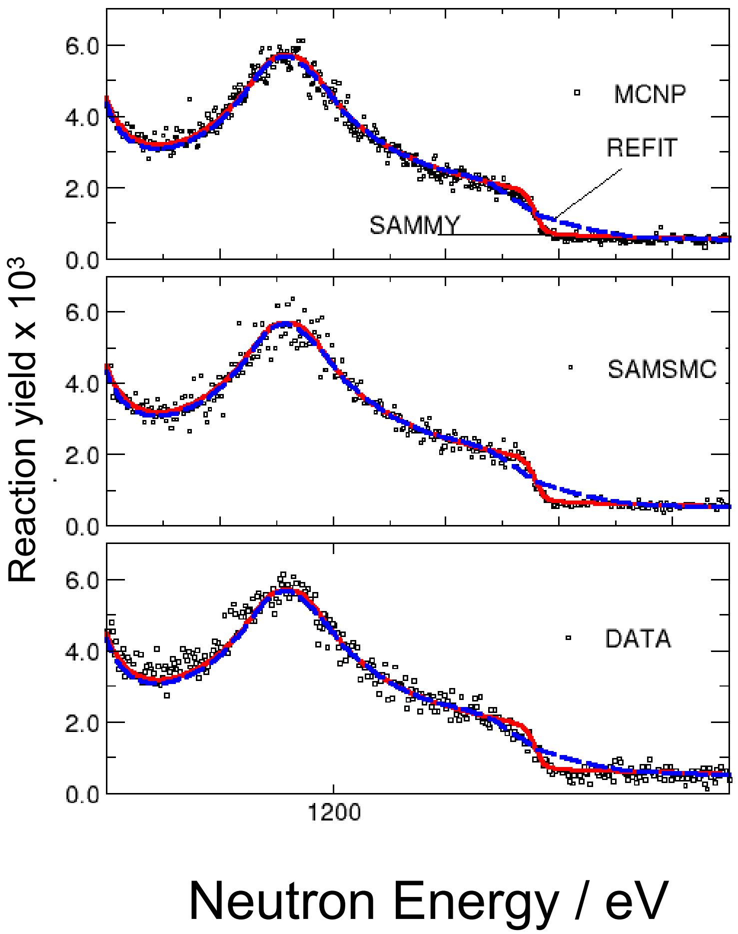
⇒ Transmission less sensitive to RF than capture

Kopecky et al., WONDER 2006

Self-shielding + multiple scattering for ^{56}Fe(n,γ)

n_{Fe} = 1.075 10^{-2} at/b, t = 1.3 mm



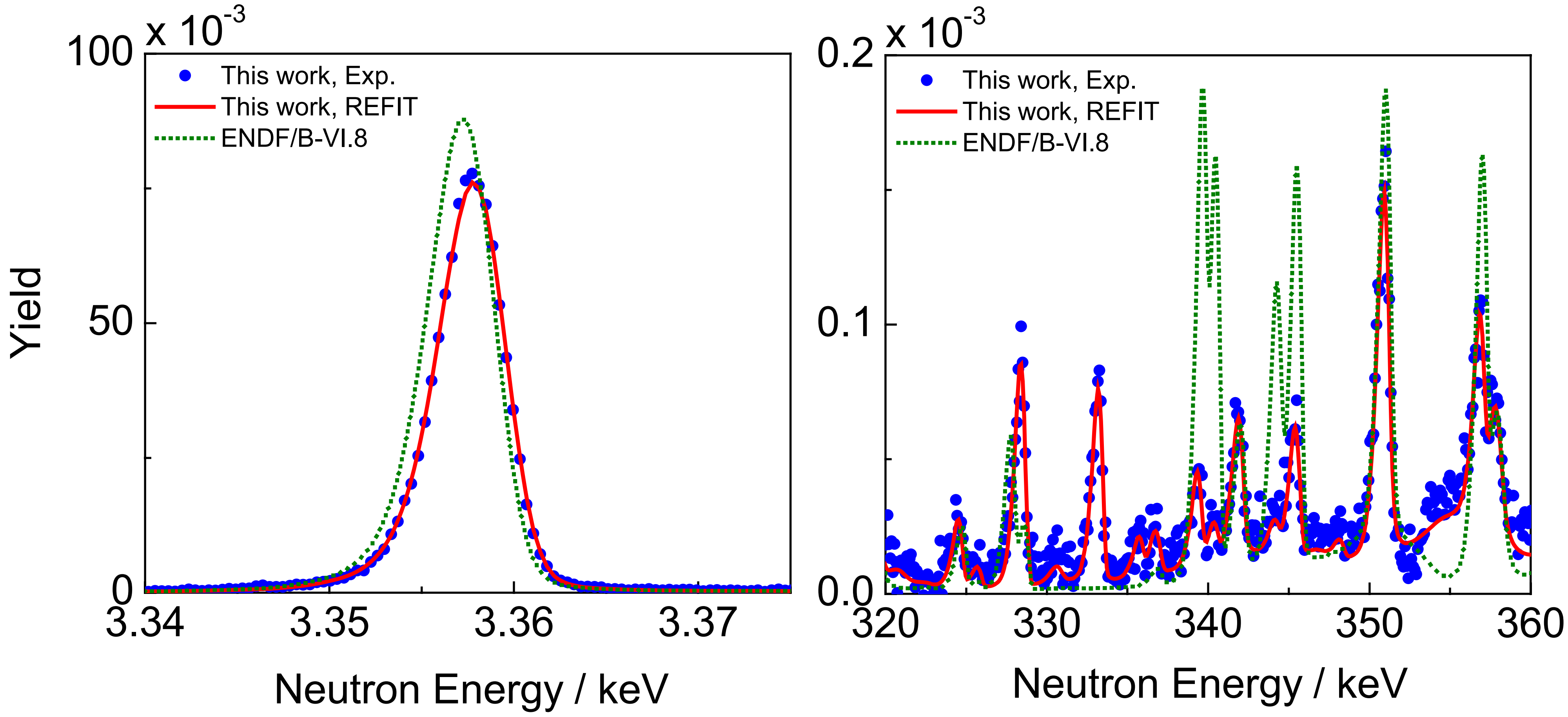


Capture experiments

- **Basic principles**
- **Spectroscopic measurements**
- **Total energy principle with PHWT**
- **Normalization**

- **Applications**

^{206}Pb + n :results



Borella et al., Phys. Rev. C 76 (2007) 014605

New - resonance parameter file for ^{206}Pb+n

- Resonance parameter file (matching thermal values)

  - 304 resonances up to 620 keV (221 in ENDF/B-VI.8)
  - R’ = 9.54 (2) fm
  - $E_o > 80$ keV: Γ_n from ORELA, (Horen et al., PR C20, 478 (1979))

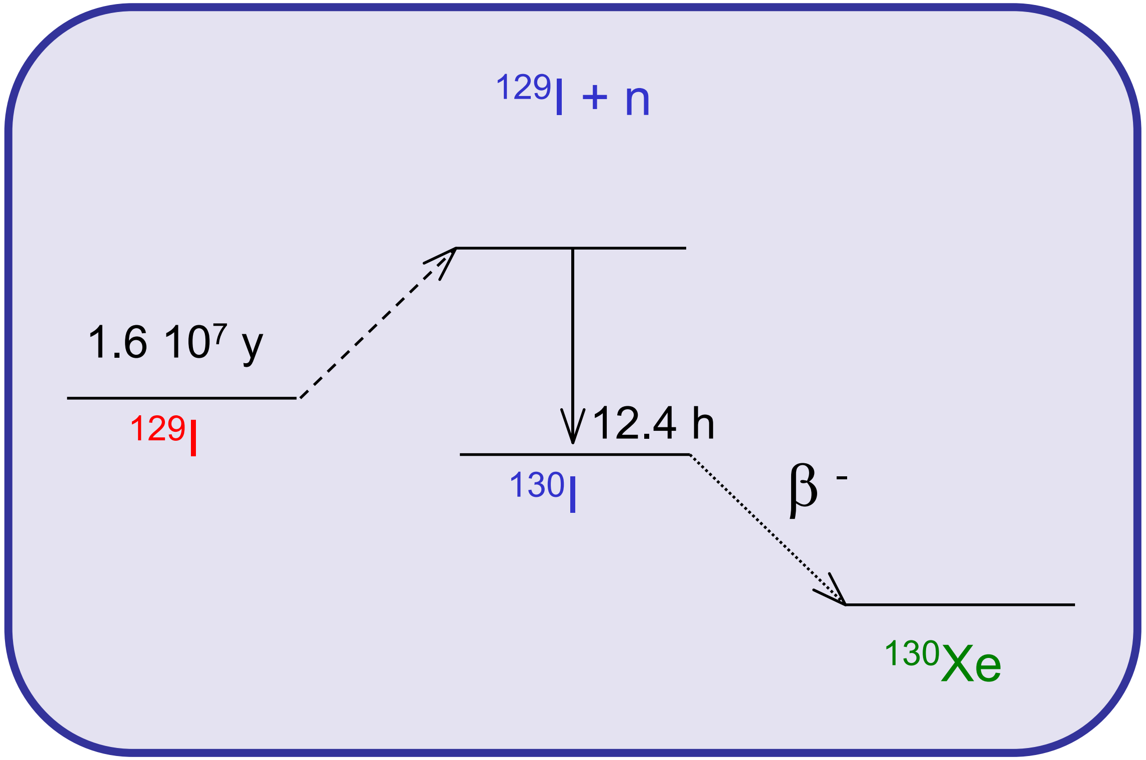
- Photon strength functions from C_6D_6 spectra

  - $f_{E1} < 225$ (5) 10^{-9} MeV^{-3}
  - f_{M1} follows systematics (no M1 enhancement)
  - B(E2)↓

Borella et al., Phys. Rev. C 76 (2007) 014605

Transmutation of LLFP (^{99}Tc and ^{129}I)

$$^{129}\mathrm{I} + n \rightarrow {}^{130}\mathrm{I} \rightarrow {}^{130}\mathrm{Xe}\ \text{(stable)}$$



Iodine sample preparation & characterisation

- **Sample preparation group at IRMM extracted:**

  ~ 150 g Iodine (powder) from 210 liter reprocessed waste (1.3 g/l Iodine and 40 MBq/l)

- **Sample characterisation: (PbI_2 + PbO) powder sample by mass spectrometry , (N)AA and NRTA**

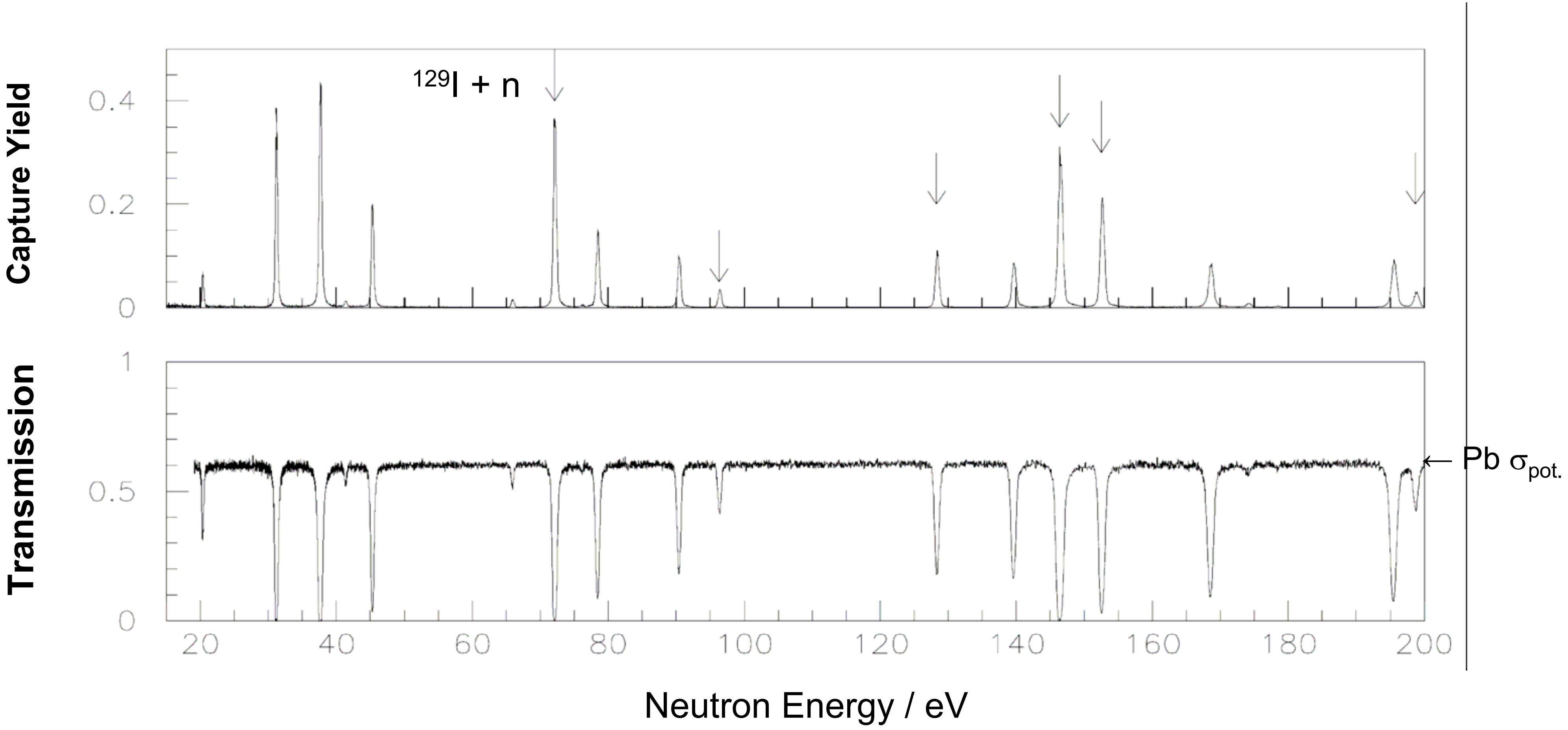
  –PbI_2 (~ 43.2%)
  –PbO (~ 56.8%)

	Weight %
^{129}I	16.5
^{127}I	3.4
$^{129}I/^{127}I$	4.9
Pb	59.5
Na	1.0
O	14.5
N	1.2

G. Noguerre, PhD 2003 and PRC, 2007

Resonance Parameters for ^{129}I + n

Overlap with ^{127}I +n resonances and Pb potential scattering

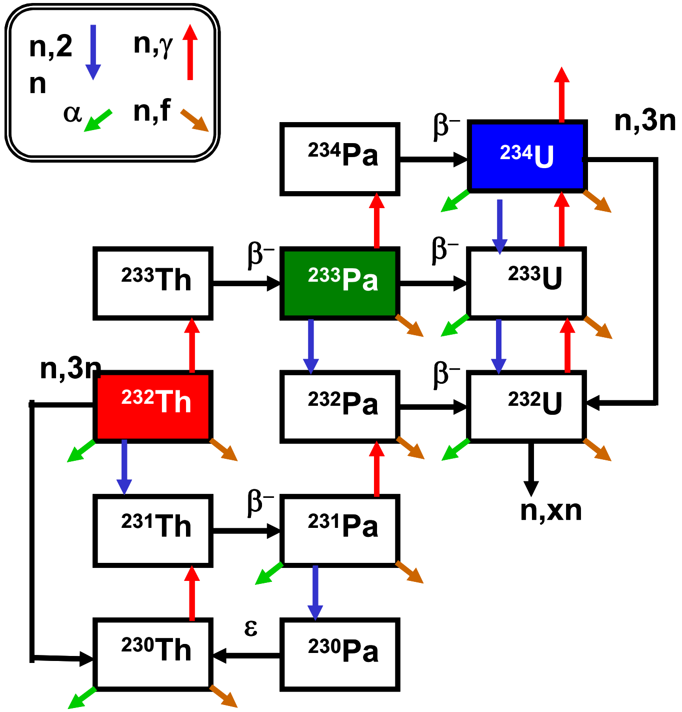


⇒ **Additional measurements on ^{127}I + n**

^{129}I and ^{127}I : in RRR

Data	^{127}I		^{129}I	
	E_{max} (RRR)	N	E_{max} (RRR)	N
BROND 2.2			2.1 keV	66
ENDF/B-VI	1.0 keV	93	0.2 keV	5
JEFF 3.0	2.0 keV	188	3.0 keV	126
JENDL 3.3	4.2 keV	374	3.0 keV	127
IRMM	10.0 keV	719	10.0 keV	400

IAEA CRP: Thorium – Uranium Fuel Cycle



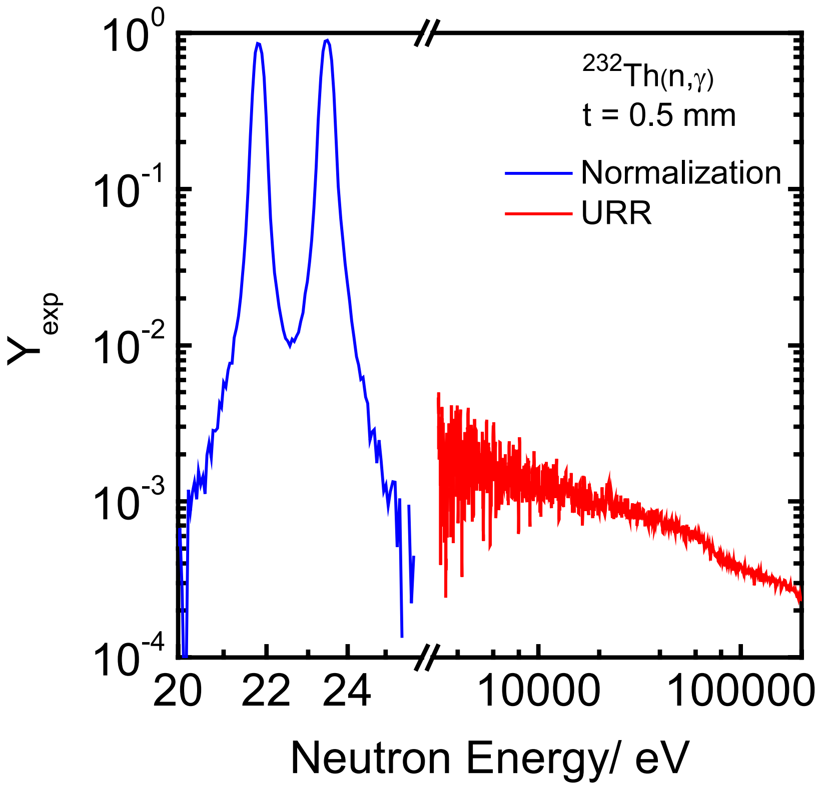
Nuclide	Reaction	Accuracy (%)
^{232}Th	total	3
	(n,γ)	1-2
^{233}Pa	(n,f)	20
^{234}U	(n,f)	3
^{236}U	(n,f)	5

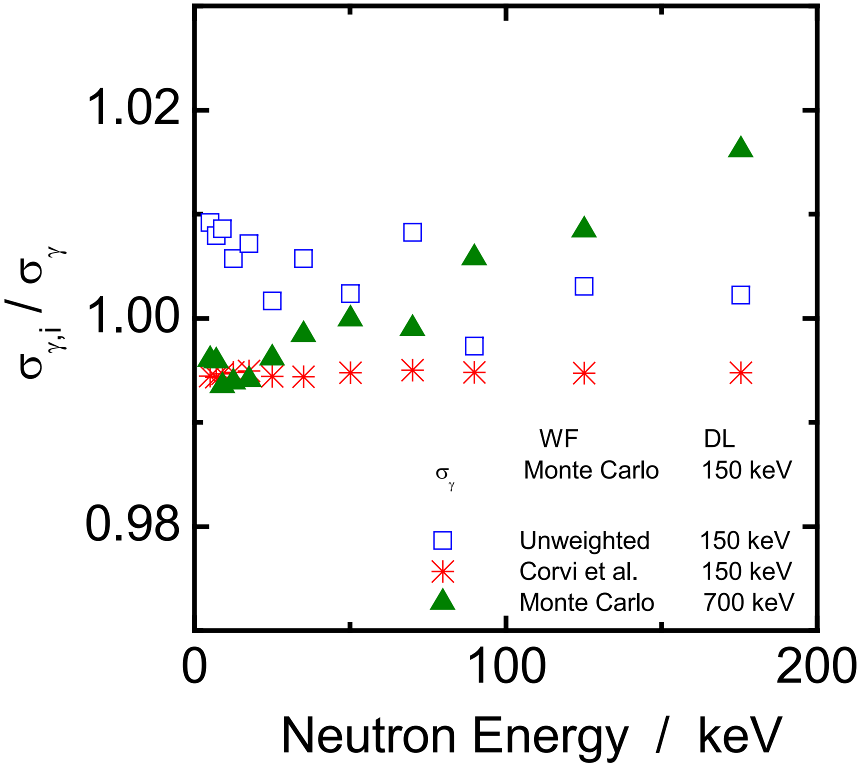
1) 2 % uncertainty on σ_γ for ^{232}Th(n,γ) in URR ⇒ uncertainty on ^{233}U production rate

2) 10 % uncertainty on σ_γ for ^{232}Th(n,γ) ⇒ 30% uncertainty on proton current to operate ADS with a $k_{eff} \approx 0.97$

1) E.T. Cheng and R.D. Mathews, ND1979, p. 834
2) M. Salvatores, ND1997, Part I, p. 3

Internal normalization : ^{232}Th(n,γ) in URR





Ref.	E_r = 21.8 eV		E_r = 23.5 eV		N / 0.339
	Γ_n / meV	Γ_γ / meV	Γ_n / meV	Γ_γ / meV	
Olsen et al.	2.08	25.3	3.82	26.9	1.000
Kobyashi et al.	2.09	25.2	3.88	26.1	1.000
Chrien et al.	2.10	24.0	3.70	26.0	0.994

Borella et al., Nucl. Sci. Eng. 152 (2006) 1-14

Overall uncertainty : 1.7 %

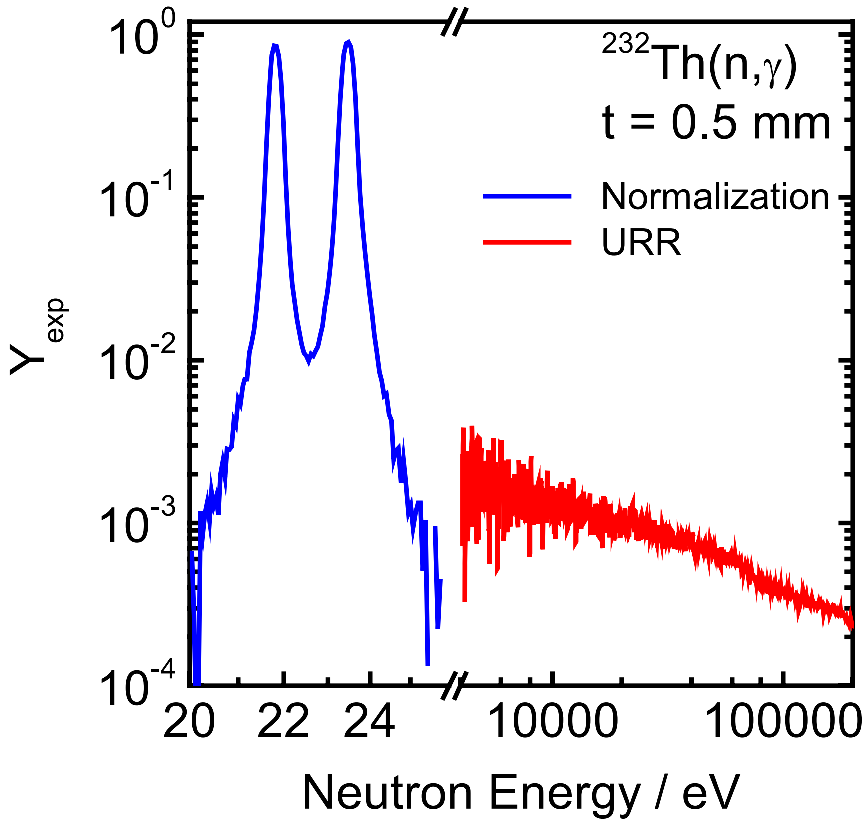
normalization : 1.5 %

uncorrelated : 0.2 %

+ dead time, self-shielding

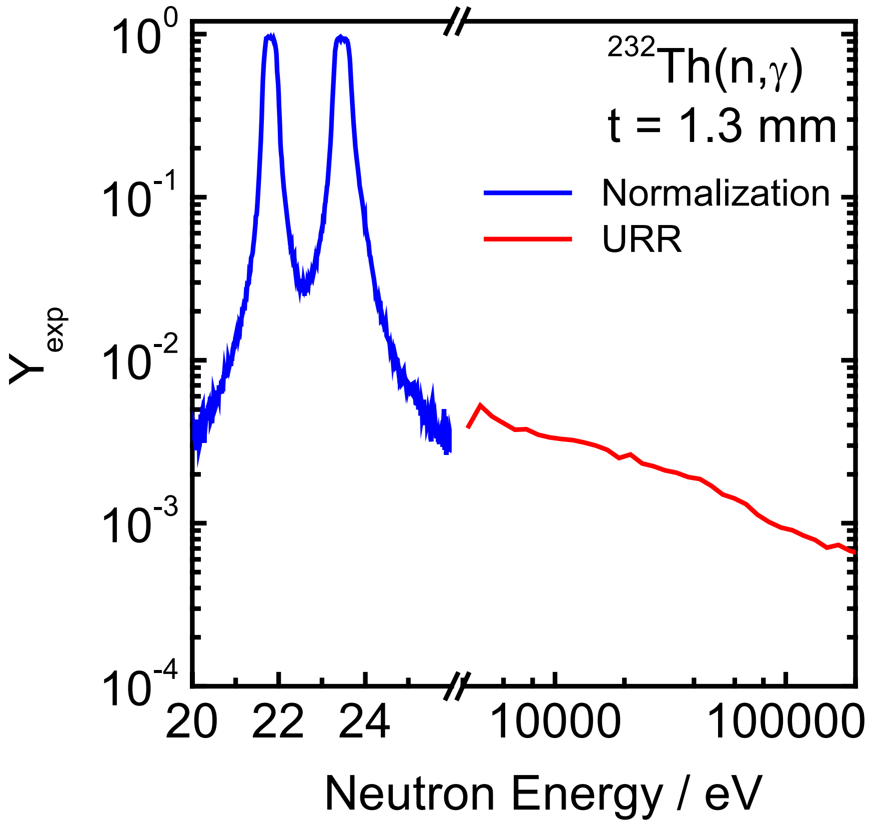
Examples : ^{232}Th(n,γ)

GELINA



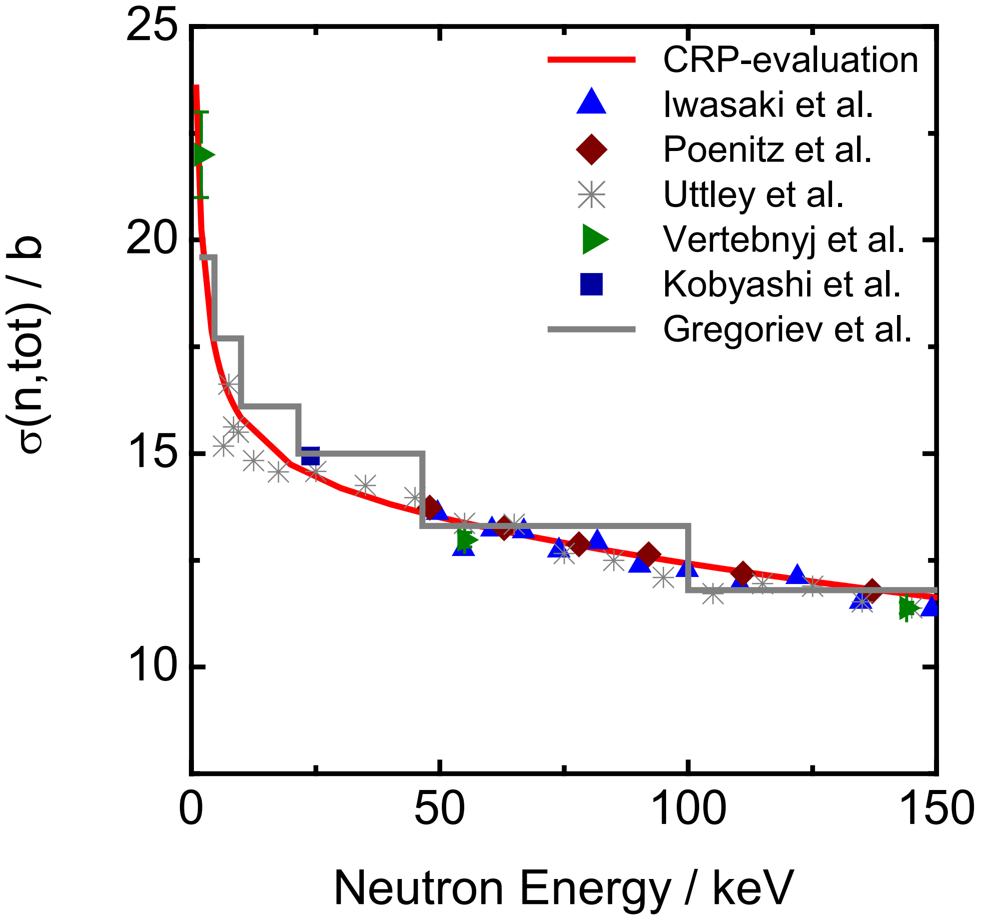
Borella et al., NSE 152, 1 (2006)

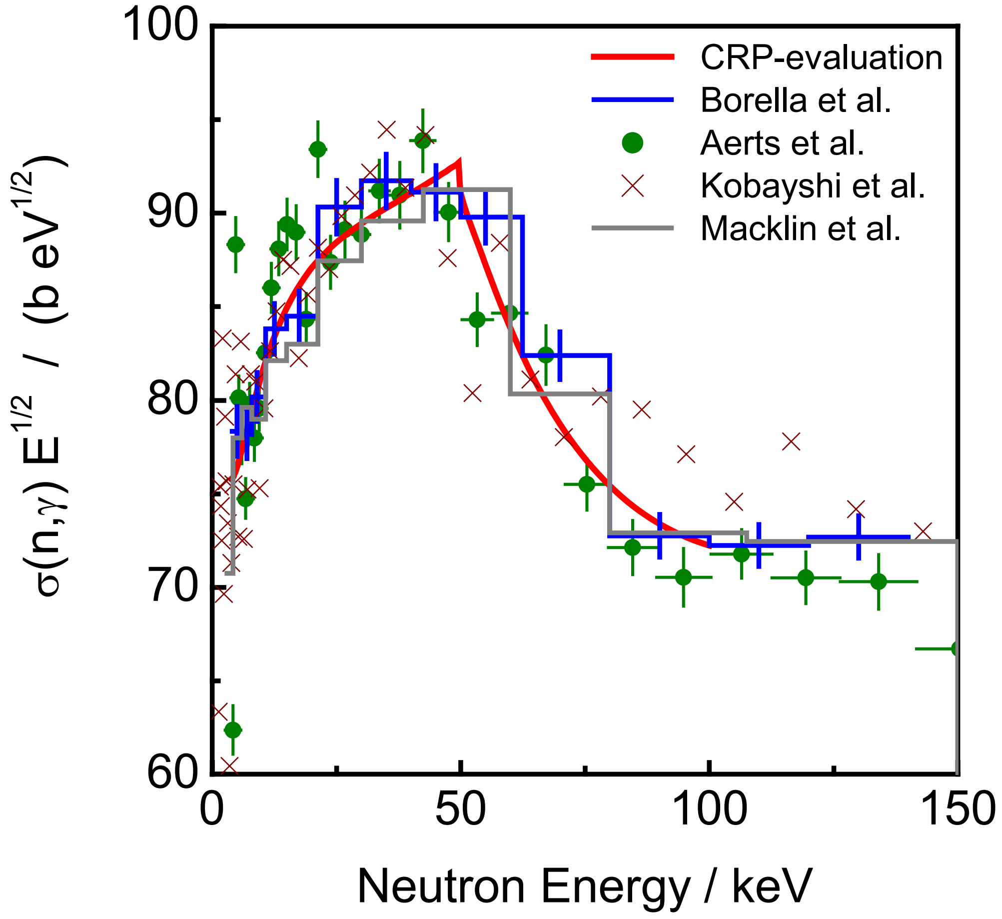
nTOF



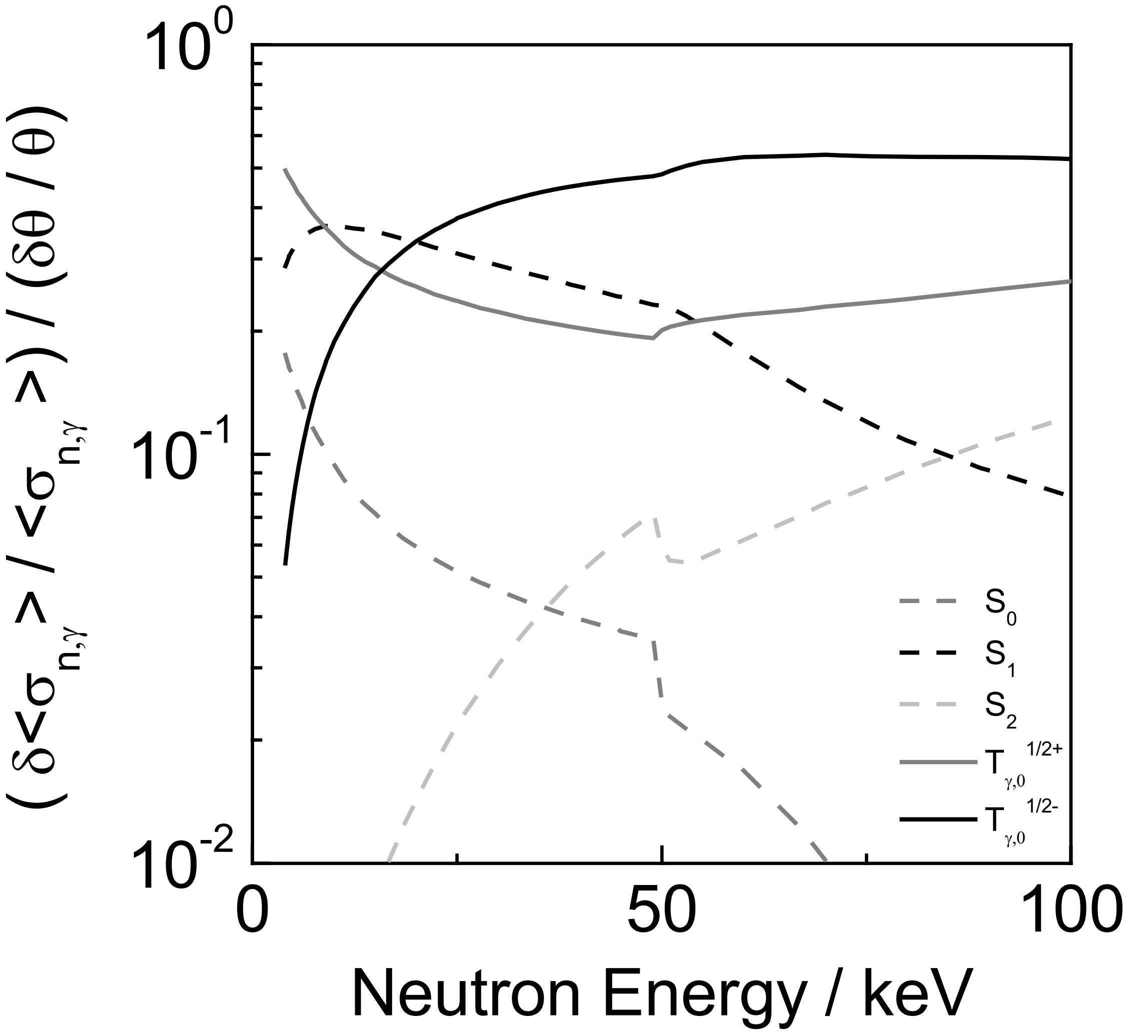
Aerts et al., PR C73, 054610 (2006)

IAEA CRP : Th-U fuel cycle





Sirakov et al., Annals of Nuclear Energy, 2008doi:10.1016/j.anucene.2007.12.008,


10^0
10^{-1}
10^{-2}
0
50
100
Neutron Energy / keV
$(\delta\langle\sigma_{n,\gamma}\rangle / \langle\sigma_{n,\gamma}\rangle) / (\delta\theta / \theta)$
S_0
S_1
S_2
$T_{\gamma,0}^{1/2+}$
$T_{\gamma,0}^{1/2-}$