Joint Research Centre

the European Commission's in-house science service

Serving society
Stimulating innovation
Supporting legislation

Neutron resonance experiments

Design and analysis

Peter Schillebeeckx

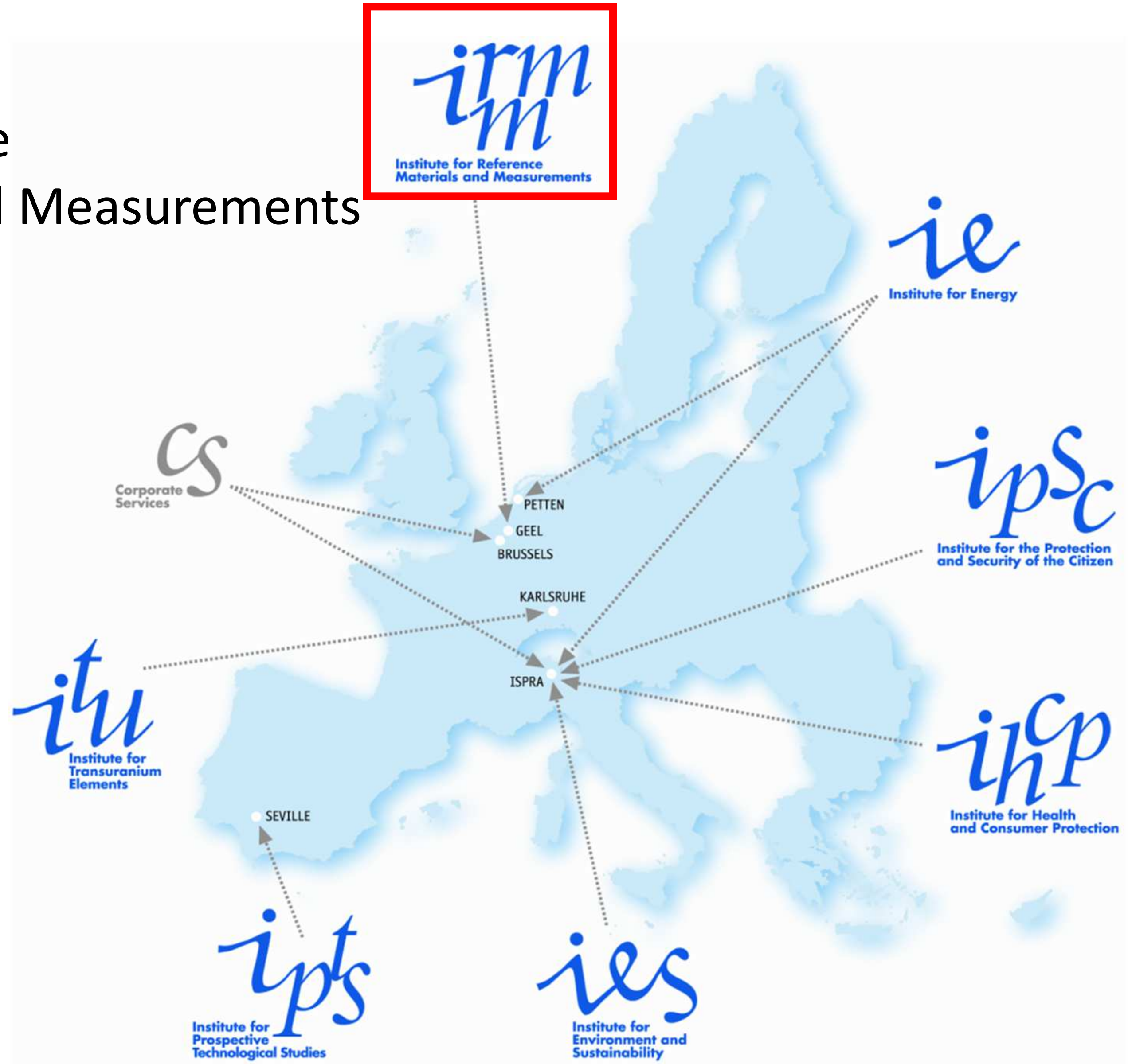
Summer school on Neutron Detectors and Related Applications
29 June – 2 July 2016
Riva del Garda, Trento, Italy

European Commission

EC – JRC – IRMM

Joint Research Centre
Institute for Reference Materials and Measurements

After re-organisation
JRC Geel



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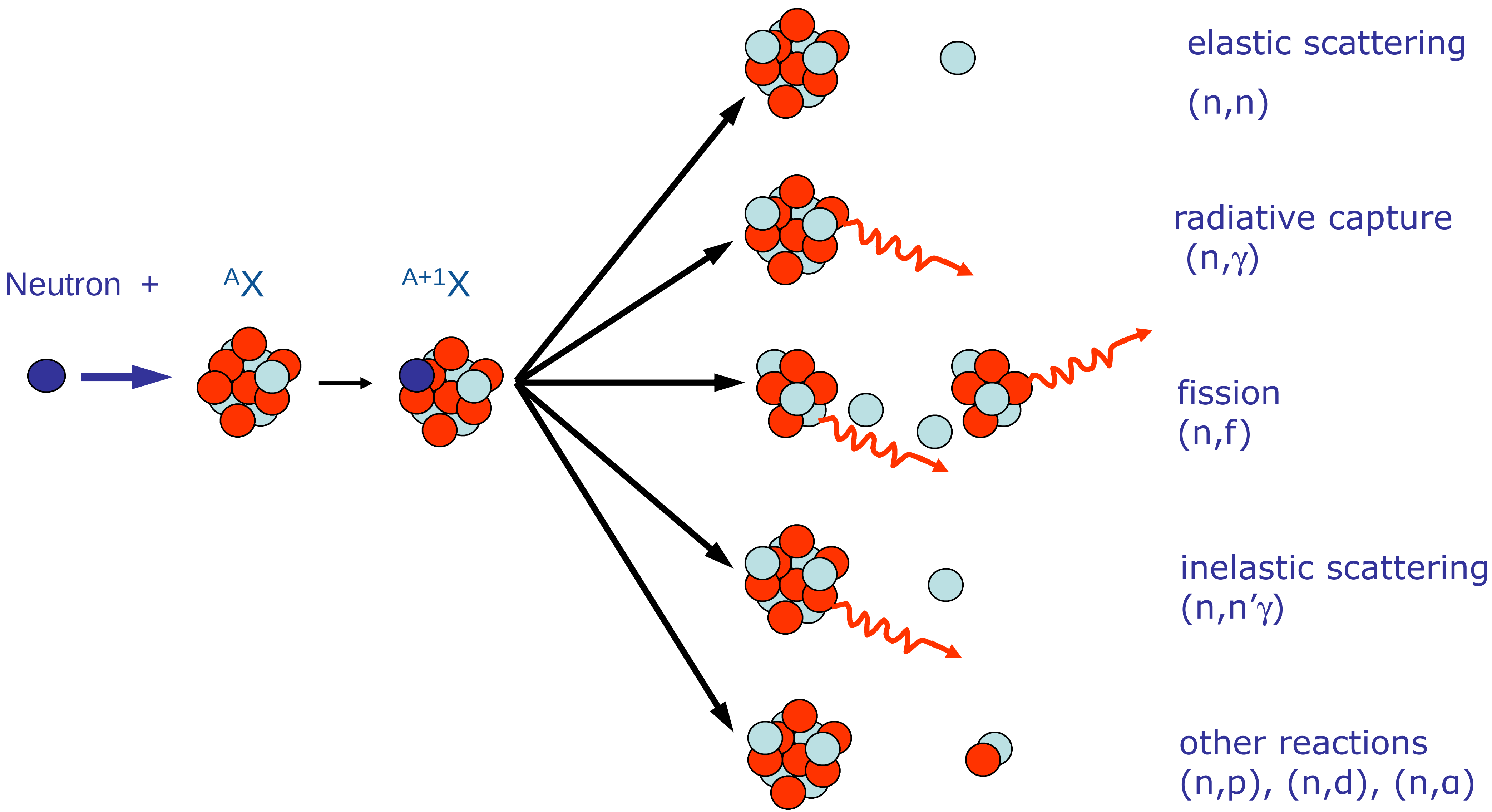
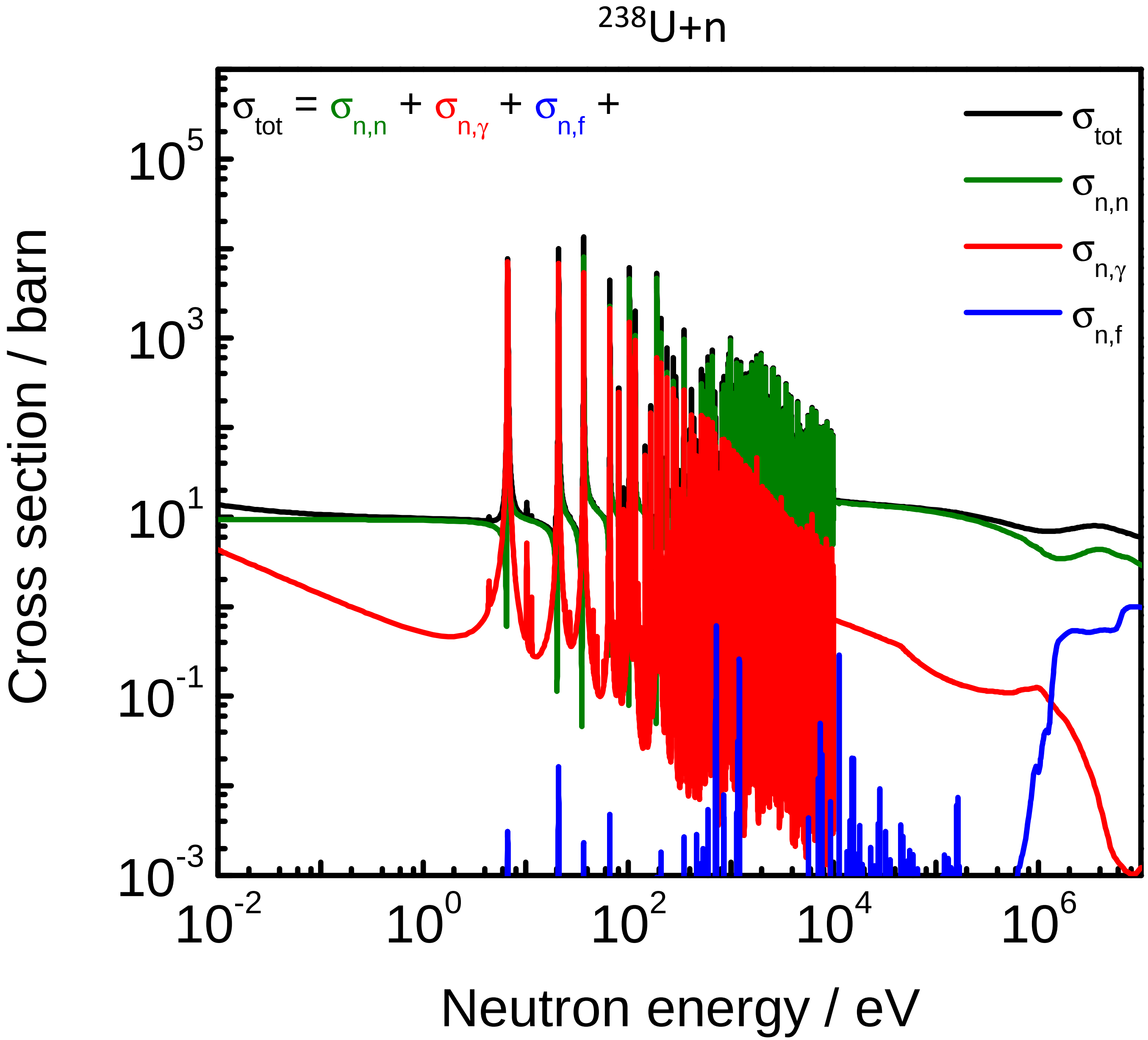
- Neutron induced reaction cross sections
- Neutron sources
  - Mono-energetic neutron beams by (cp,n) reactions
  - Time-of-flight measurements at white neutron sources (response function)
- Cross section measurements
  - Total cross section (transmission)
  - Reaction cross sections
- Neutron resonance transmission and capture analysis (NRTA& NRCA)
  - Archaeological applications
  - Nuclear applications (melted fuel from severe nuclear accidents)

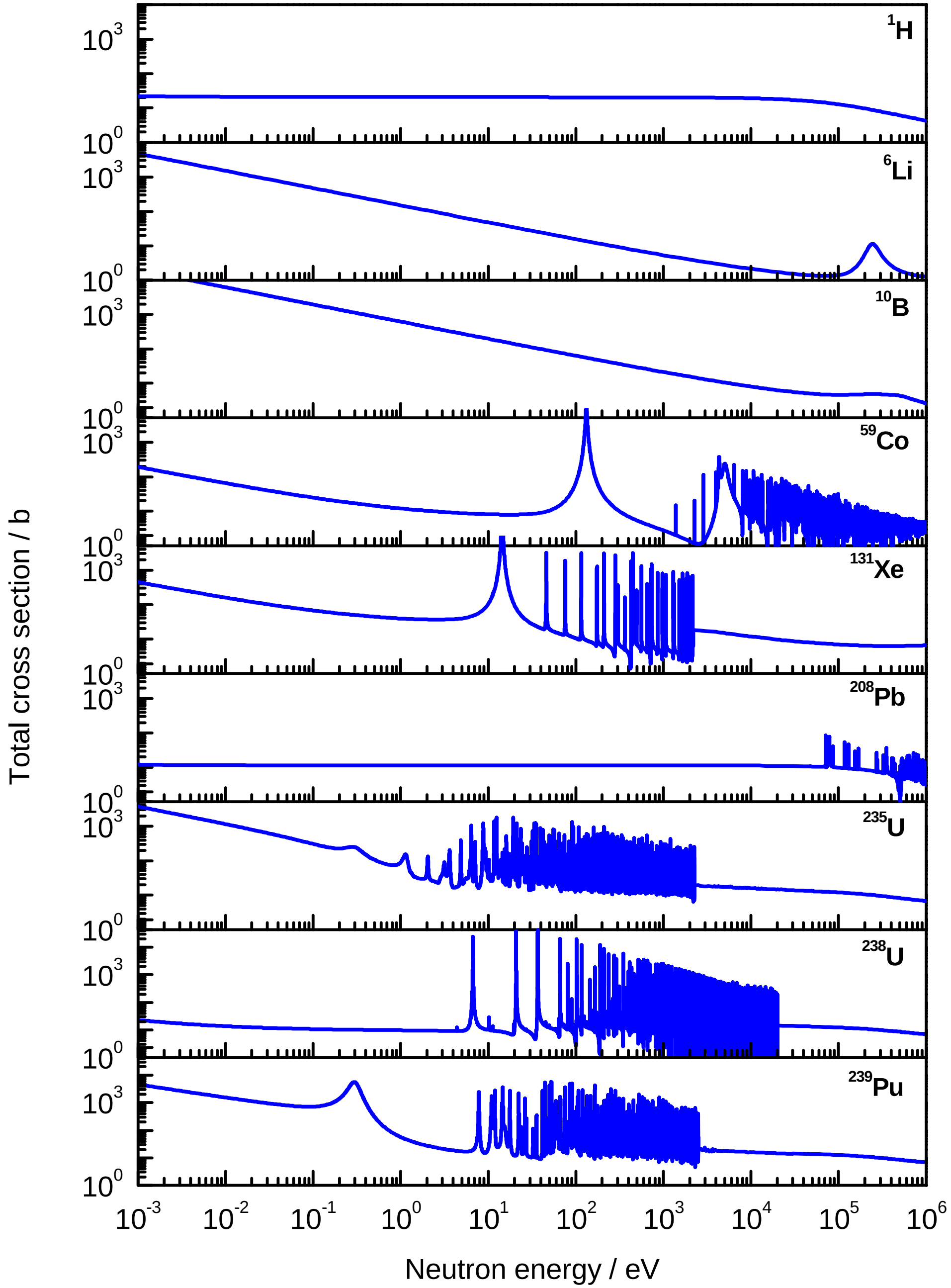
Neutron induced reactions



Neutron induced reaction cross sections

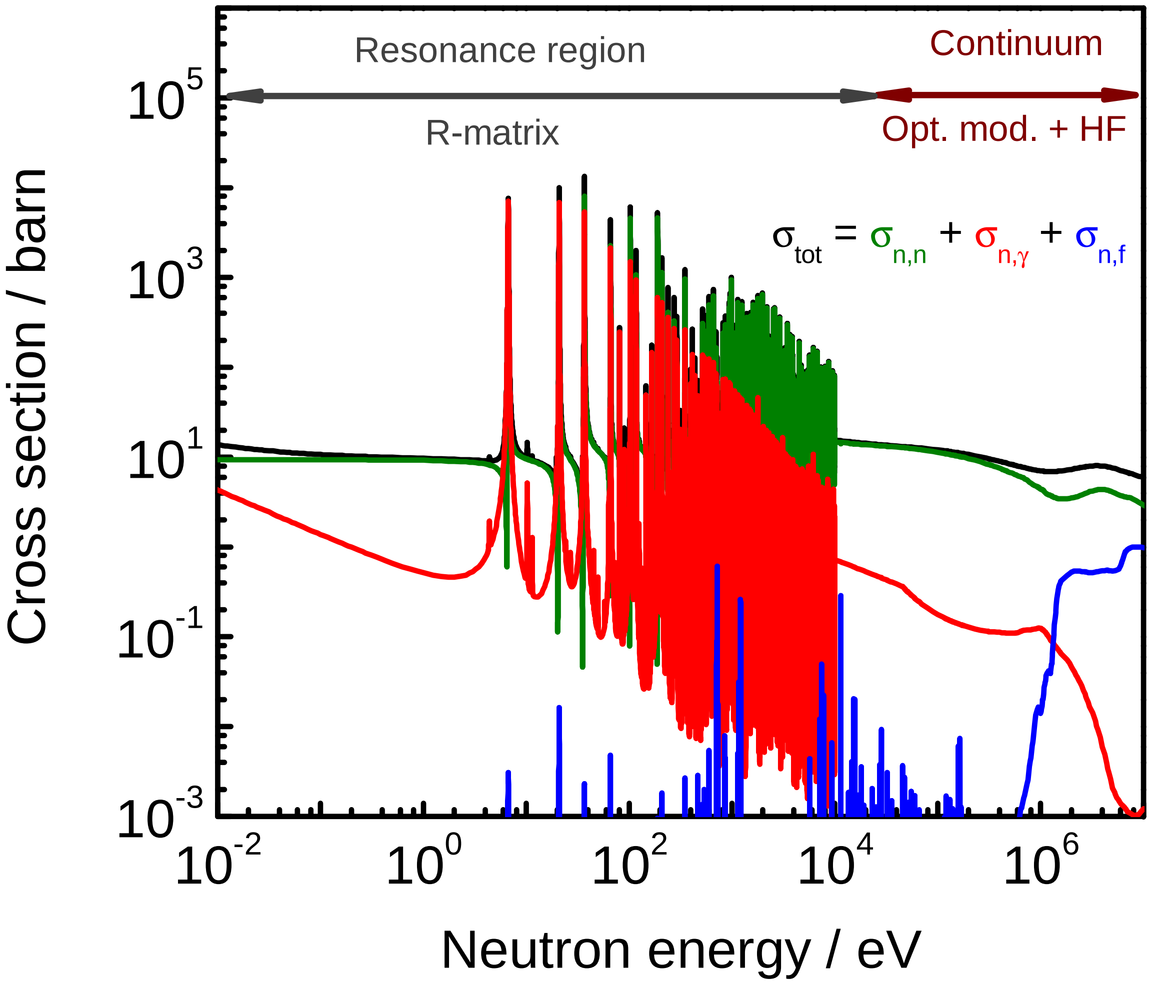


Neutron induced reaction cross sections



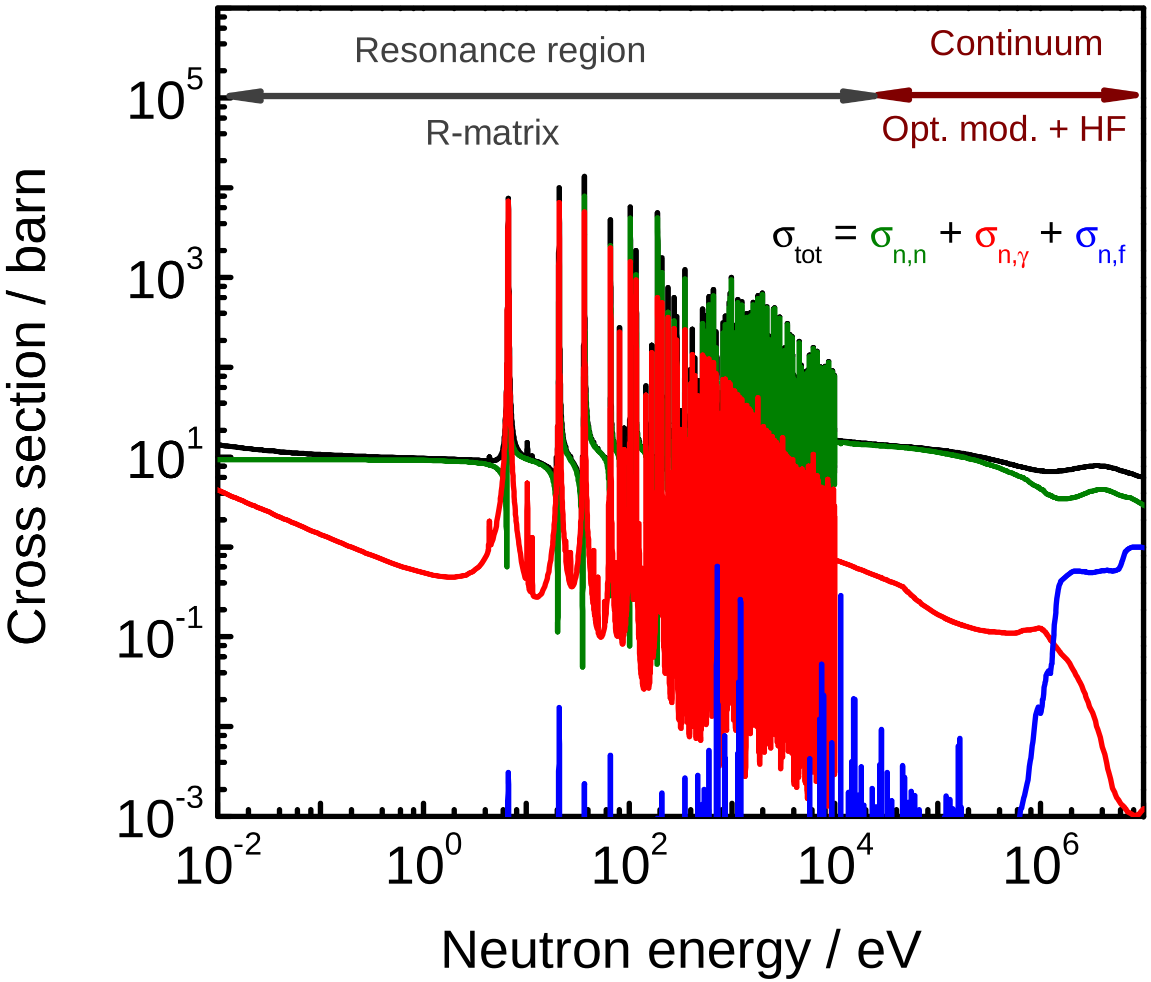
- Calculated cross sections are required for nuclear energy applications
  - Ensures full consistency, e.g. $\sigma_{tot} = \sum_i \sigma_{r,i}$
  - Doppler broadening
  - Inter- and extrapolation in regions were no data is available
- Cross sections are strongly energy dependent
- Cross sections are different for each nuclide

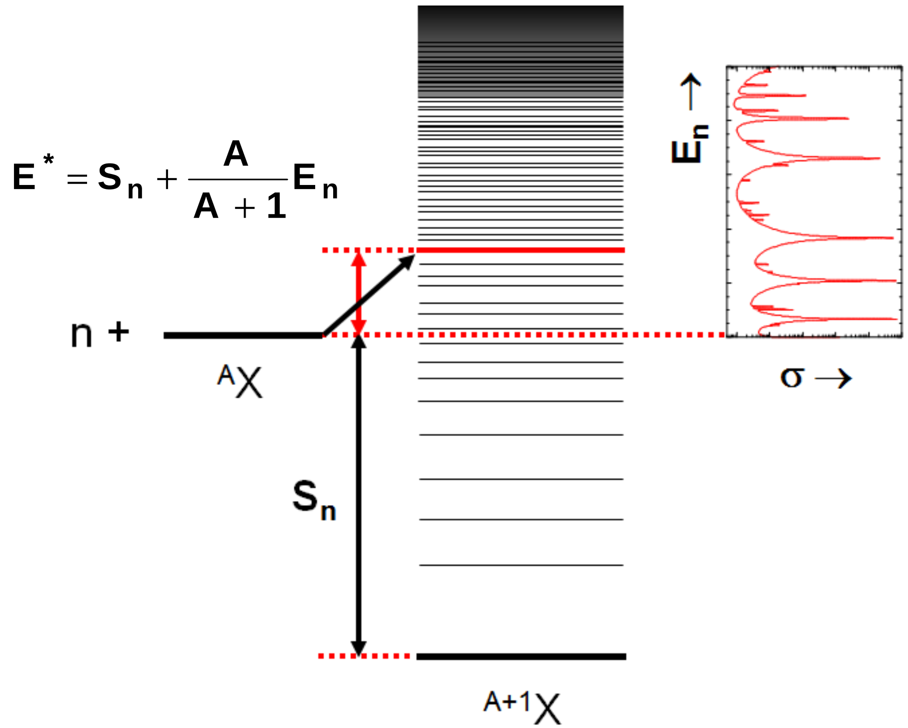
Neutron induced reaction cross sections



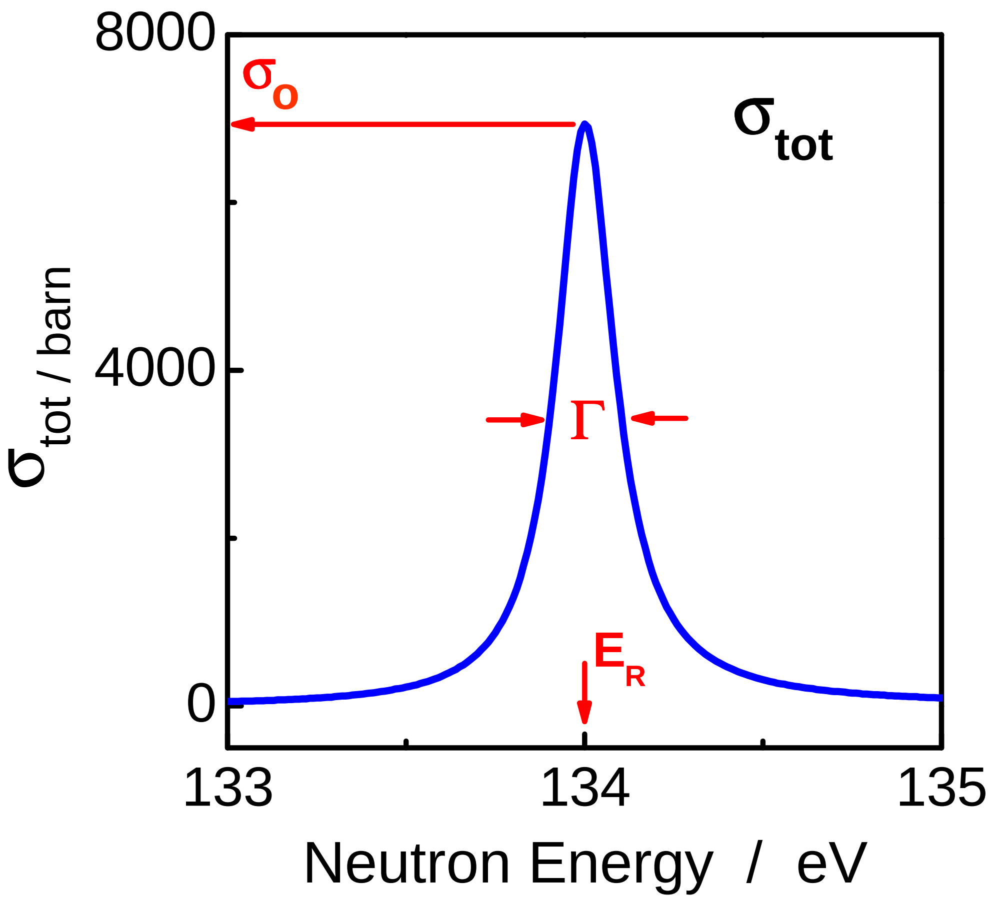
- Parameterized by nuclear reaction model
- Different models in different energy regions

Parameterisation in resonance region





Parameterisation in resonance region



The resonant structure can be described by a Breit-Wigner shape :

$$\sigma_{tot} \sim \frac{\Gamma}{(E_n - E_R)^2 + (\Gamma/2)^2}$$

with

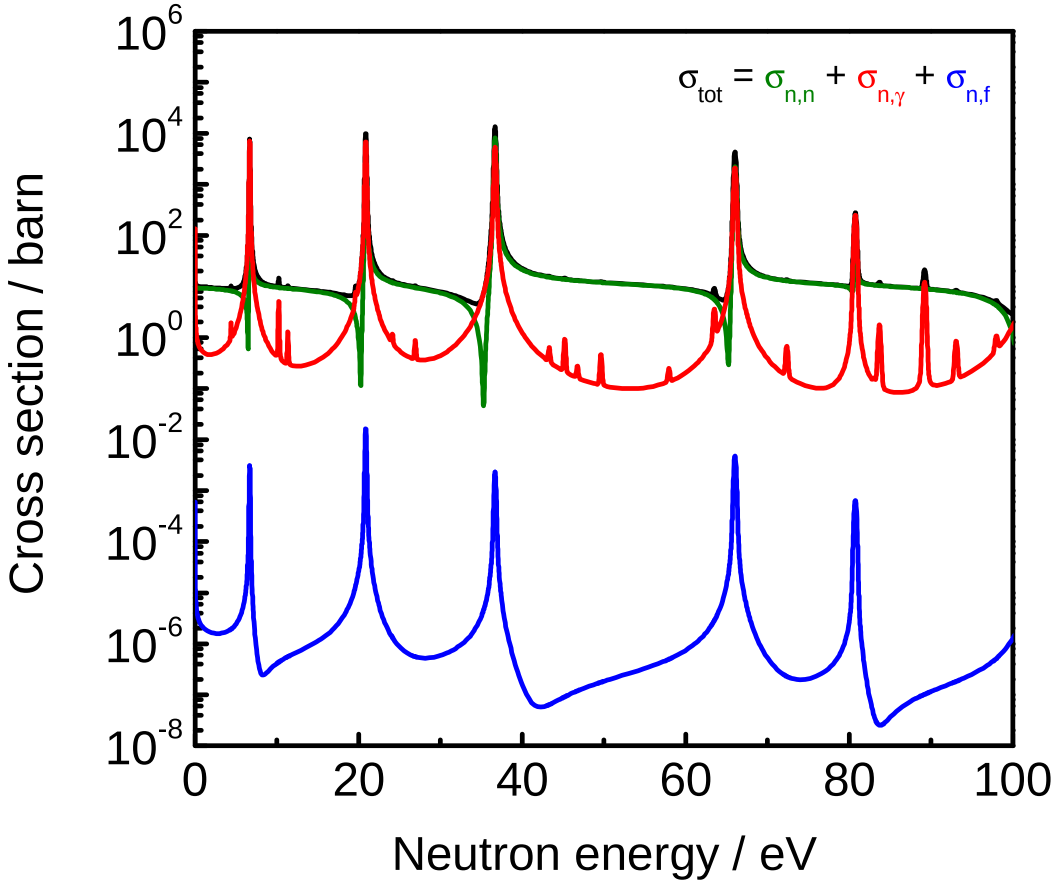
Γ total width (FWHM)

E_R resonance energy

Heisenberg uncertainty principle

$$\Delta E \Delta t = \frac{\hbar}{2} \qquad \Gamma \Delta t = \frac{\hbar}{2}$$

Parameterisation in resonance region



Model parameters

R and $(E_R, J^{\pi}, \Gamma_n, \Gamma_{\gamma}, \ldots,)_j$

E_R	resonance energy
$\Gamma_n, \Gamma_{\gamma}, \Gamma_f$	partial widths
Γ	total width
	$(\Gamma = \Gamma_n + \Gamma_{\gamma} + \Gamma_f \ldots)$
R	scattering radius

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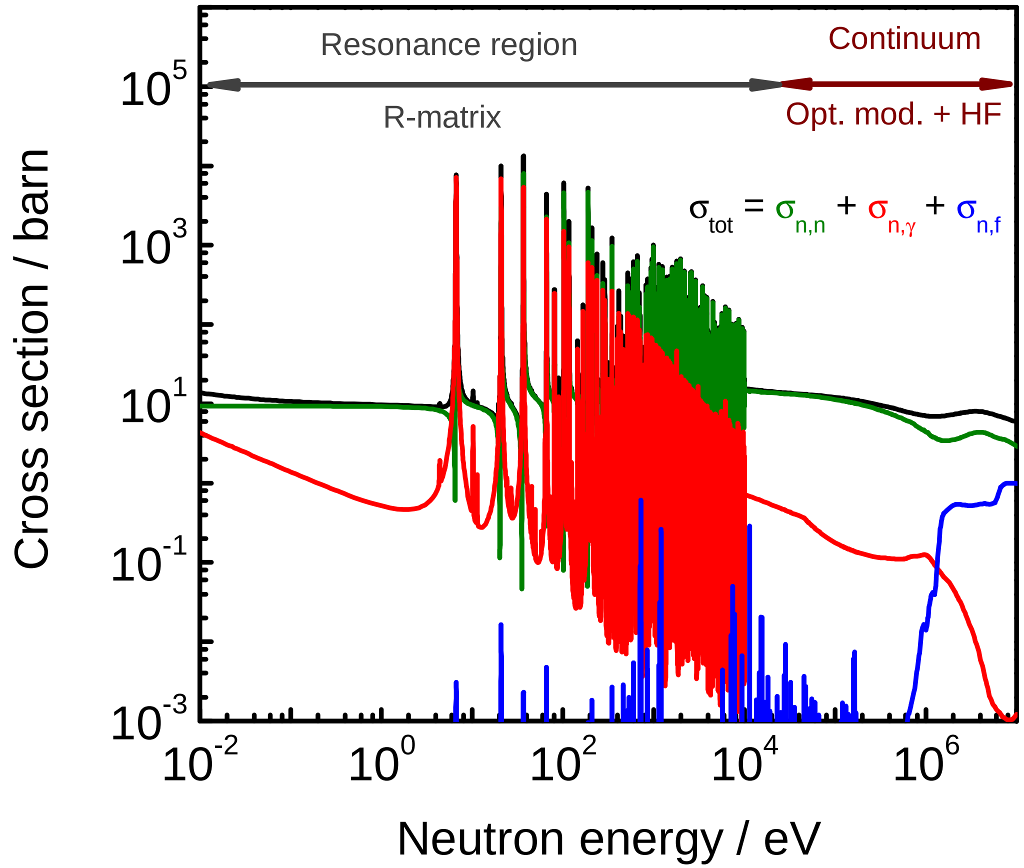
Neutron induced reaction cross sections



- Cross sections cannot be predicted by nuclear theory from first principles
- Model parameters are adjusted to experimental data

$\Rightarrow$ Experimental data are required

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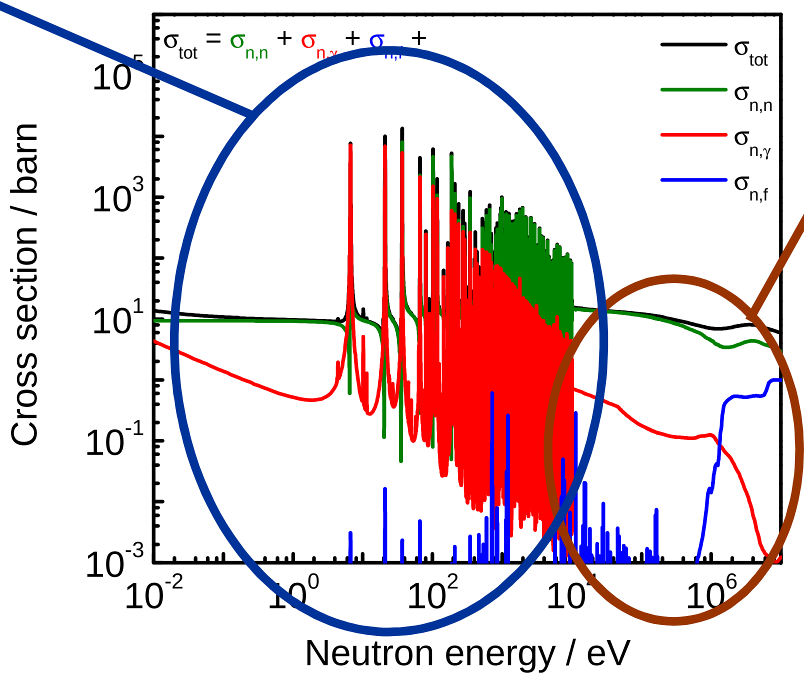
Neutron induced reaction cross sections

GELINA

Van de Graaff

$^{238}U+n$



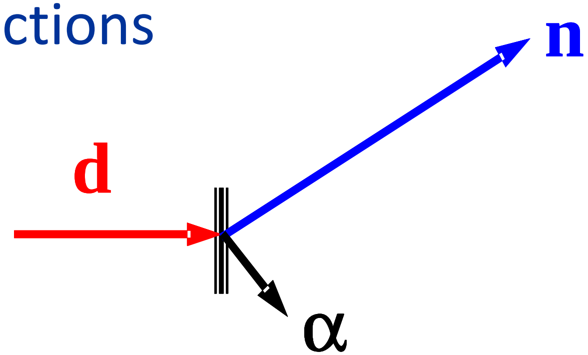
White neutron source
+
Time-of-flight (TOF)

Mono-energetic neutrons
(cp,n) reactions

Mono-energetic neutron beams by (cp,n) reactions

quasi mono-energetic neutrons produced via nuclear reactions

e.g. T(d,n)^{4}He

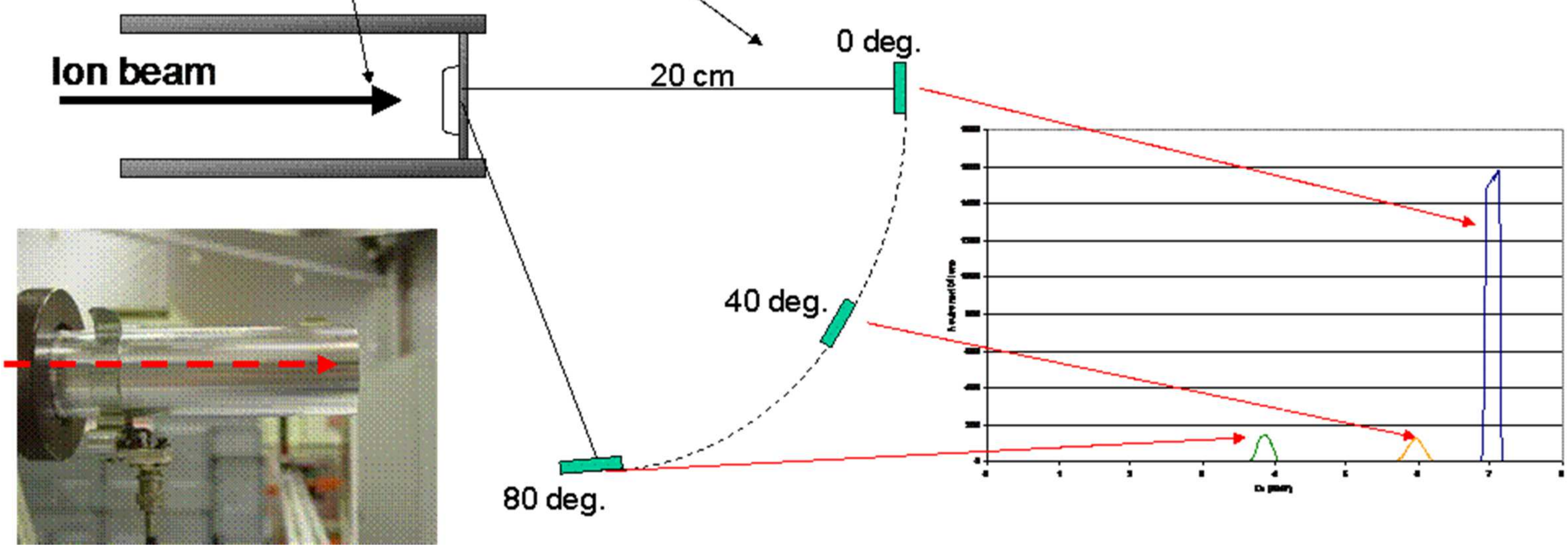


^{7}Li(p,n)^{7}Be	E_n: 0 - 5.3 MeV
T(p,n)^{3}He	E_n: 0 - 6.2 MeV
D(d,n)^{3}He	E_n: 1.8 - 10.1 MeV
T(d,n)^{4}He	E_n: 12.1 - 24.1 MeV

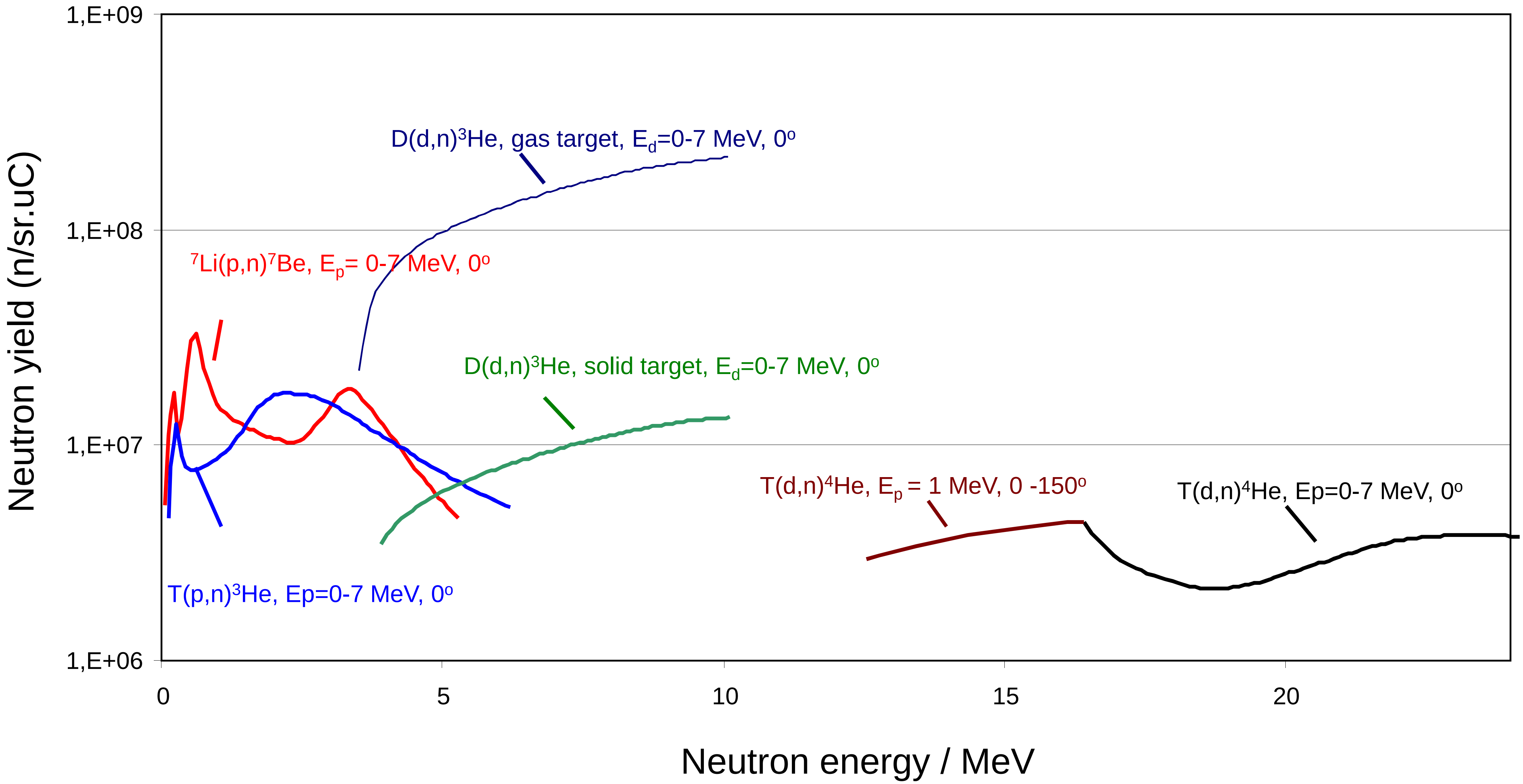
Mono-energetic neutron beams by (cp,n) reactions

- nuclear reaction,
- neutron emission angle,
- ion energy.

$^{7}Li(p,n)^{7}Be$	E_n: 0 - 5.3 MeV
$T(p,n)^{3}He$	E_n: 0 - 6.2 MeV
$D(d,n)^{3}He$	E_n: 1.8 - 10.1 MeV
$T(d,n)^{4}He$	E_n: 12.1 - 24.1 MeV



Mono-energetic neutron beams by (cp,n) reactions



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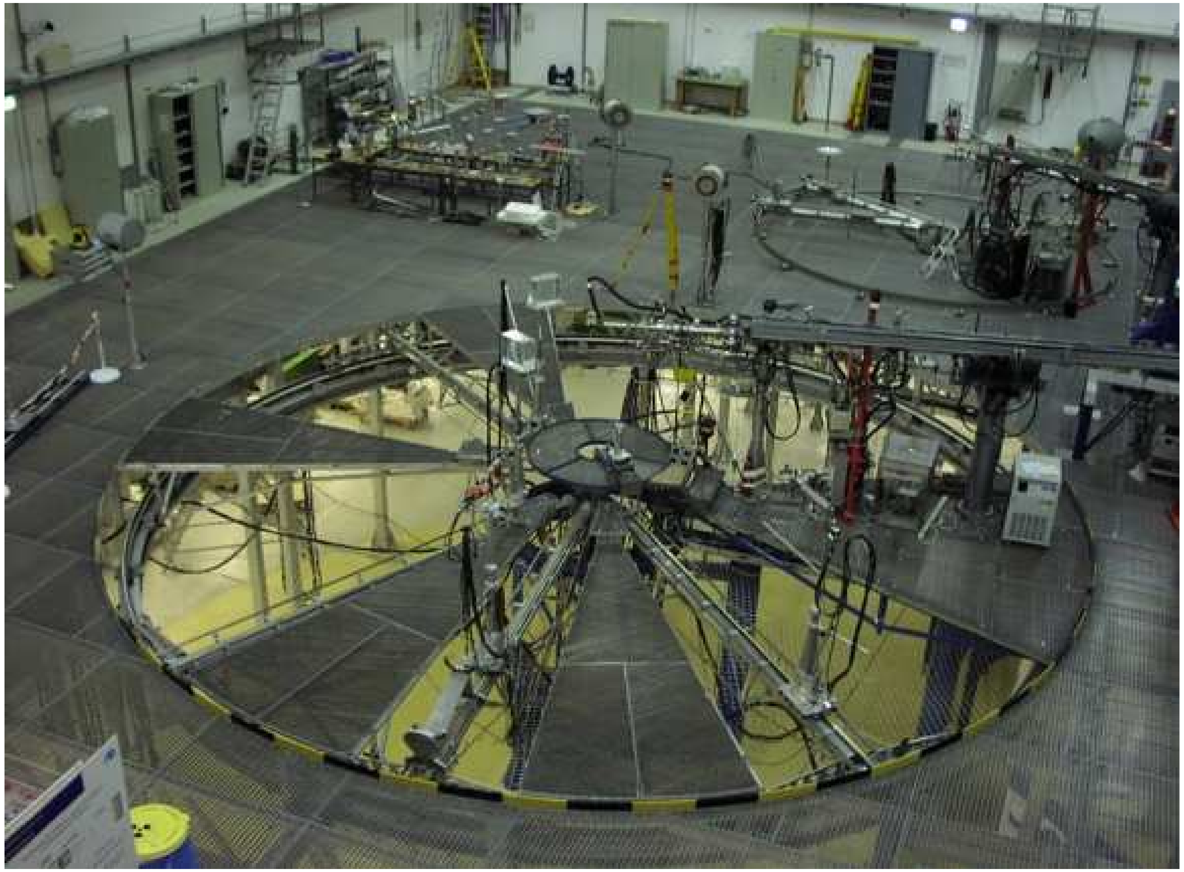
Low-scatter experimental hall at PTB Braunschweig

- Neutron cross section measurements
- Neutron activation measurements
- Fission studies
- Neutron fluence measurements
- **Calibration of detectors**

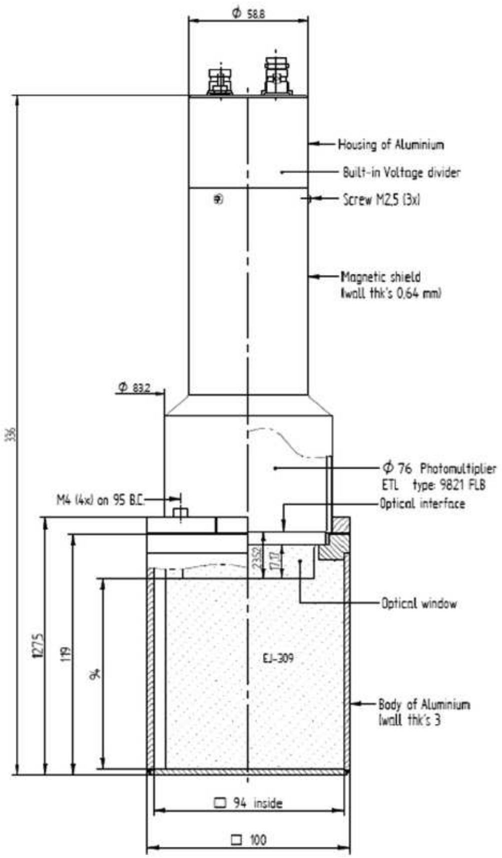


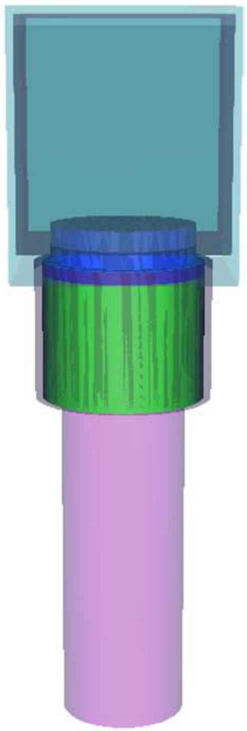
Response functions of EJ-309 scintillator at PTB

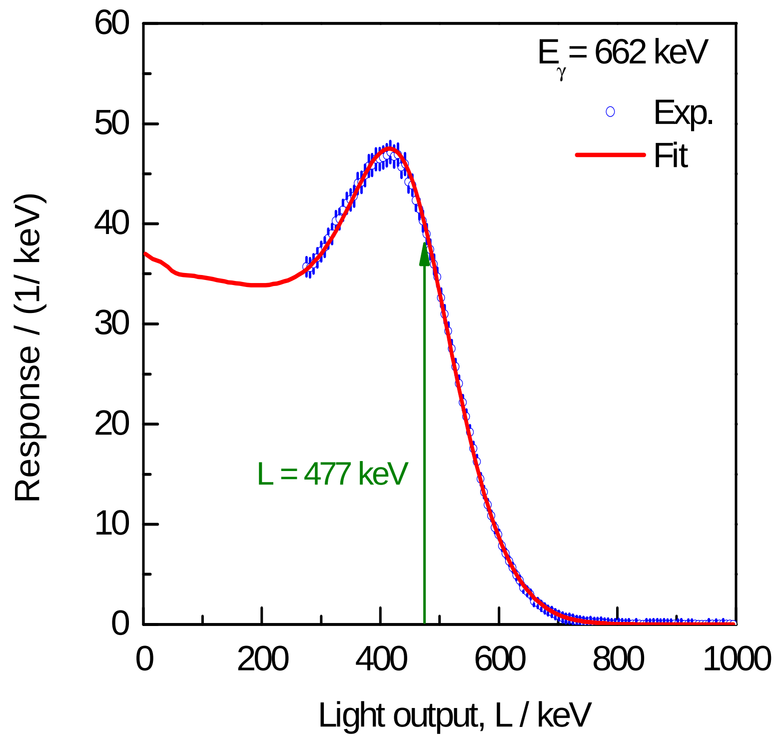
Conversion of light output into energy units based on energy deposition of electrons:

Light output for electrons is ≈ linear

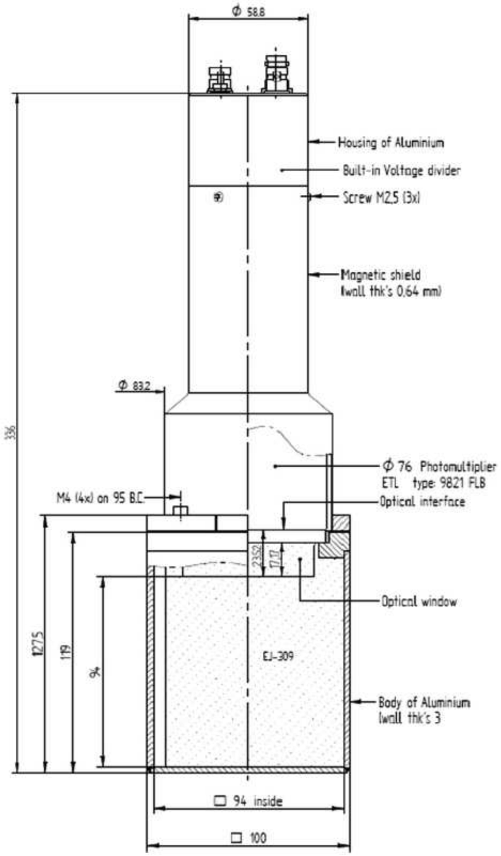
$$L_e = a\ P_e$$

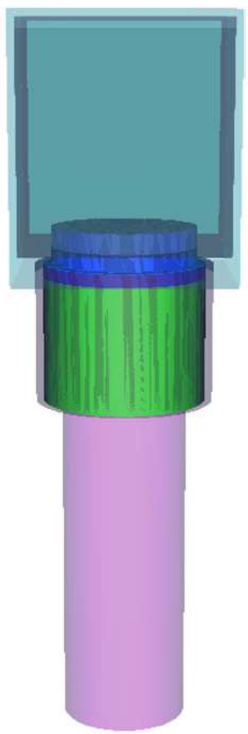


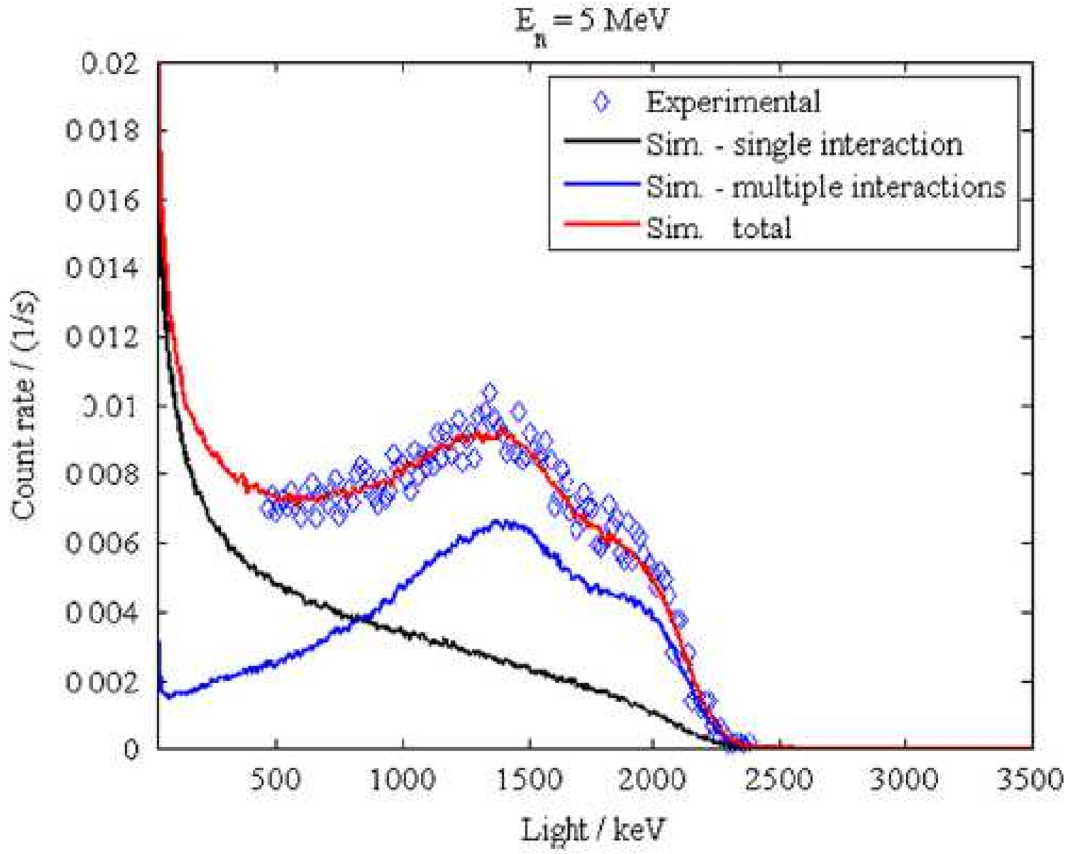




Response functions of EJ-309 scintillator at PTB

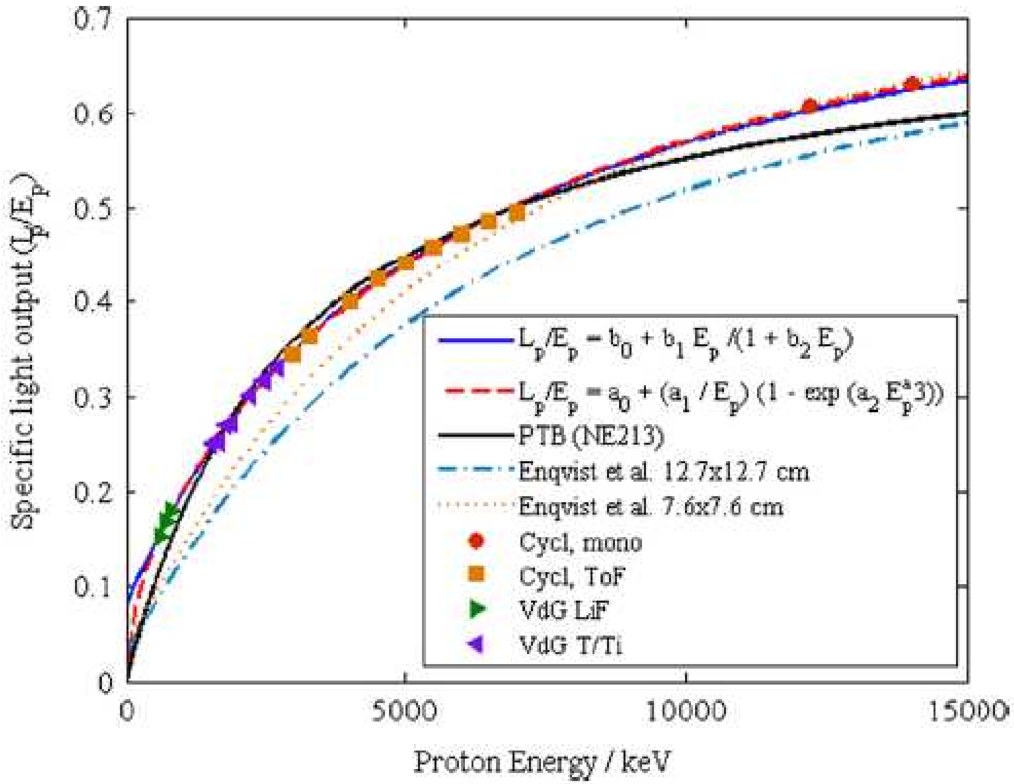


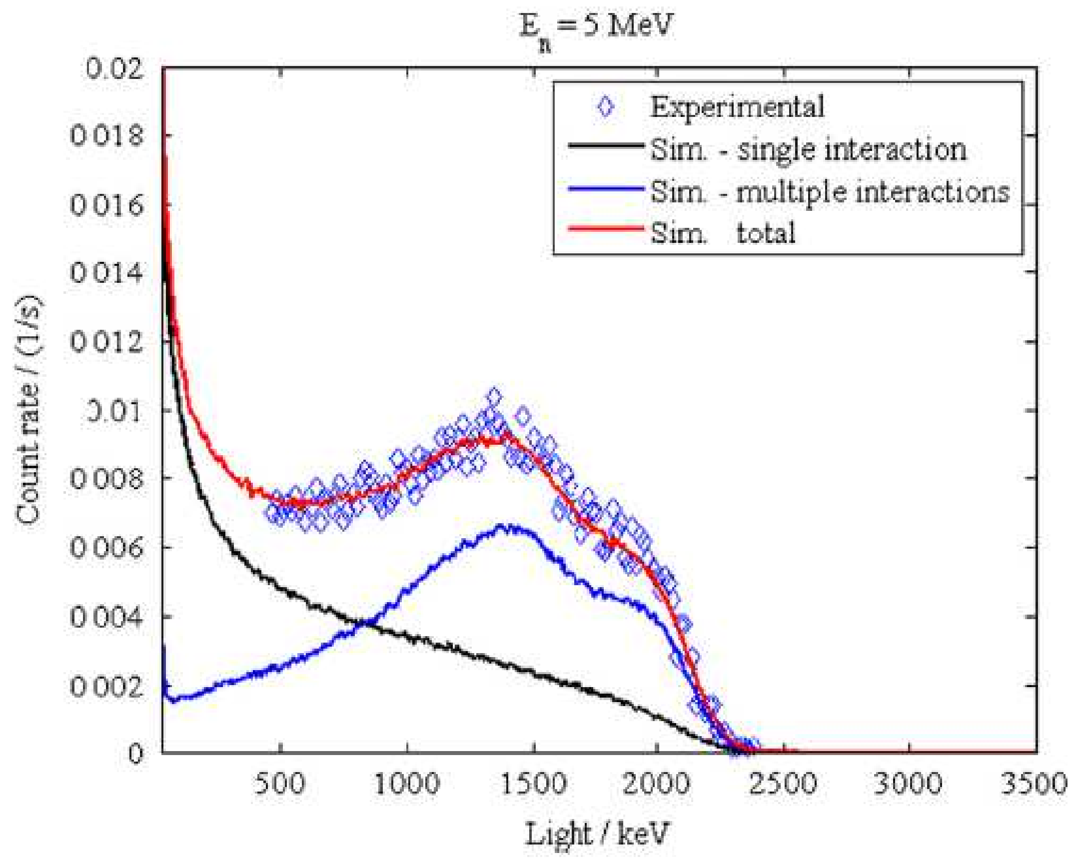




Response functions of a EJ-309 scintillator at PTB

Light output function



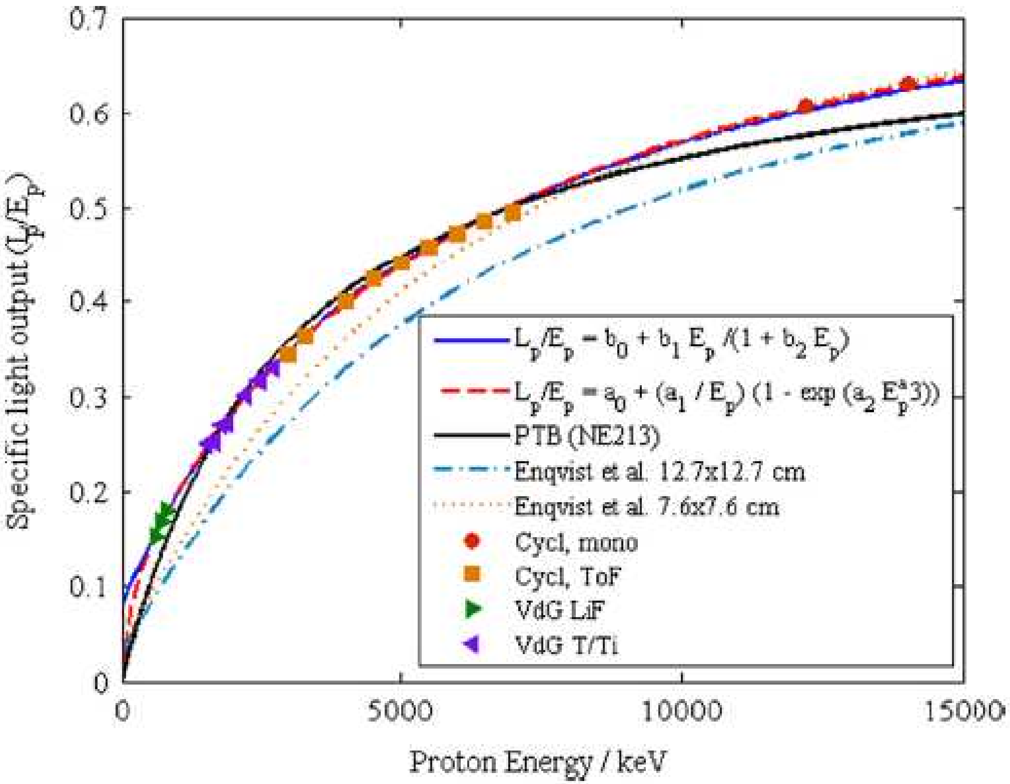


- Ideally : $L_p / E_p = 1$
- L_p : strongly non-linear (quenching)

Tomanin et al., NIMA 756 (2014) 45

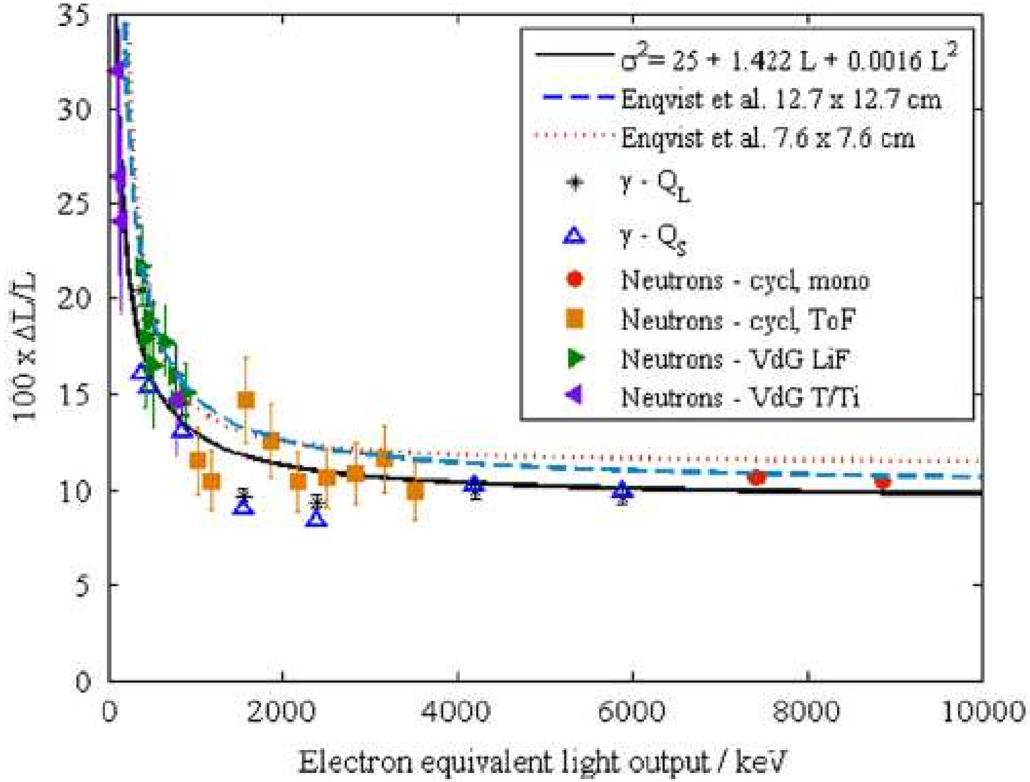
Response functions of a EJ-309 scintillator at PTB

Light output function



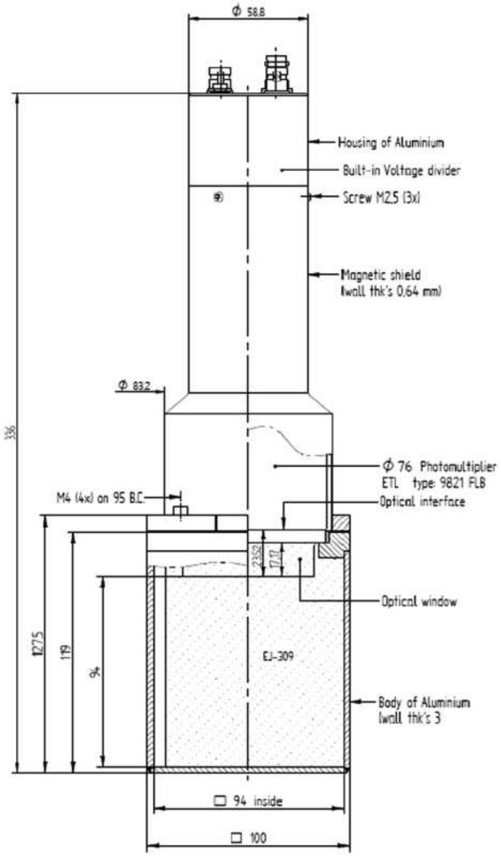
- Ideally : $L_p / E_p = 1$
- L_p : strongly non-linear (quenching)

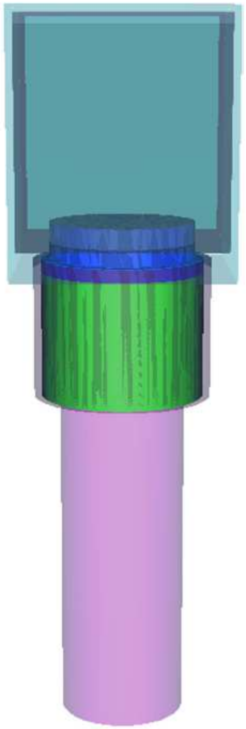
Resolution



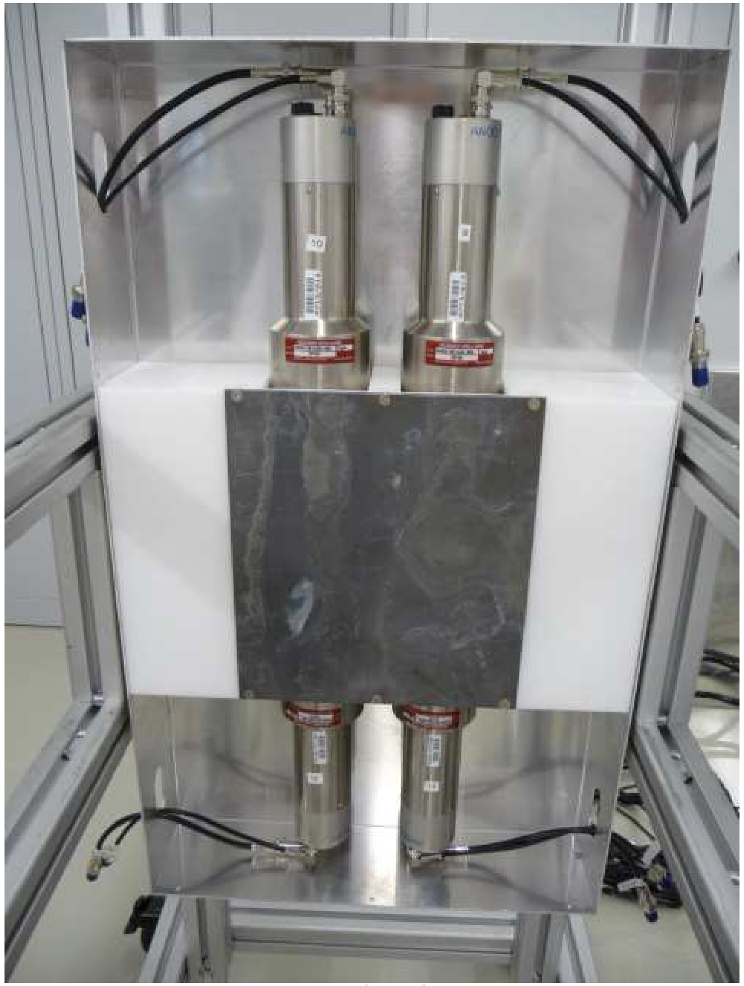
- $\Delta L_p / E_p \approx \Delta L_e / E_e$

Use of EJ-309 scintillator to verify fuel assemblies

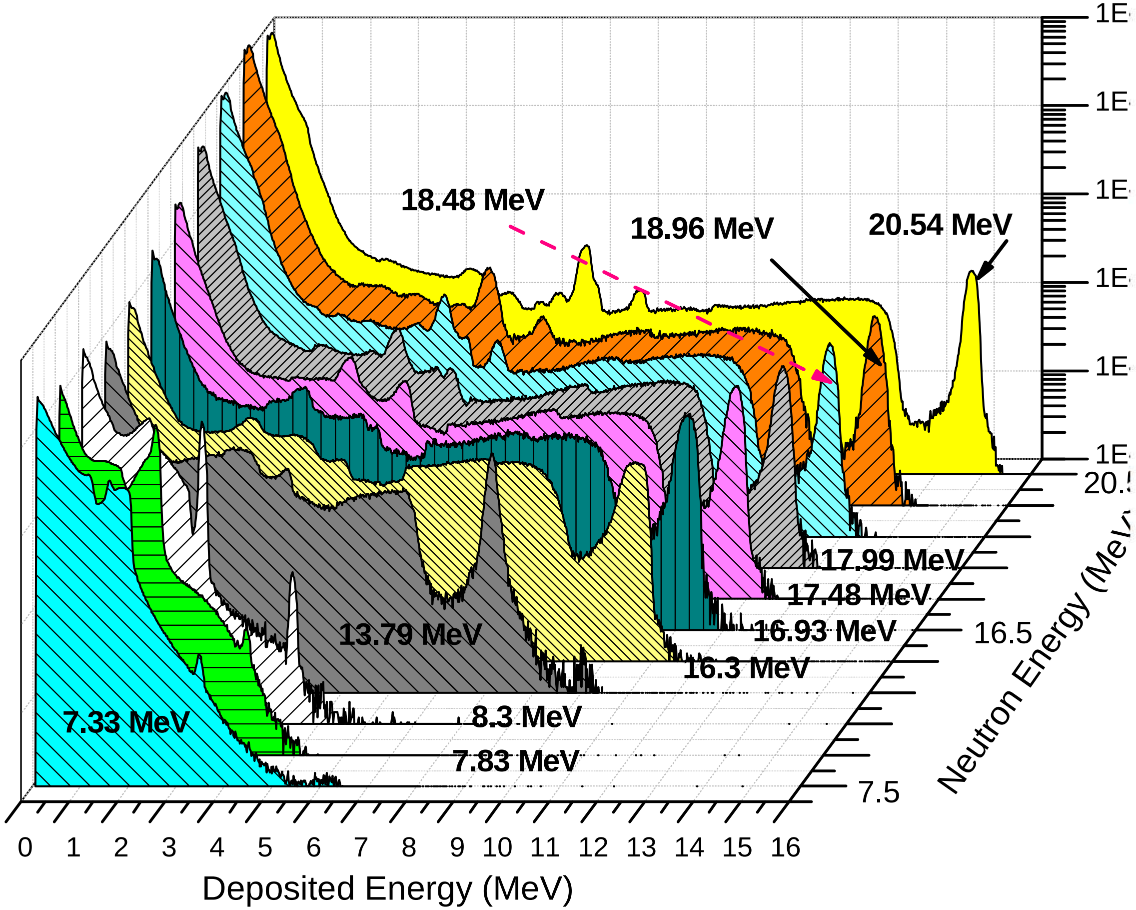


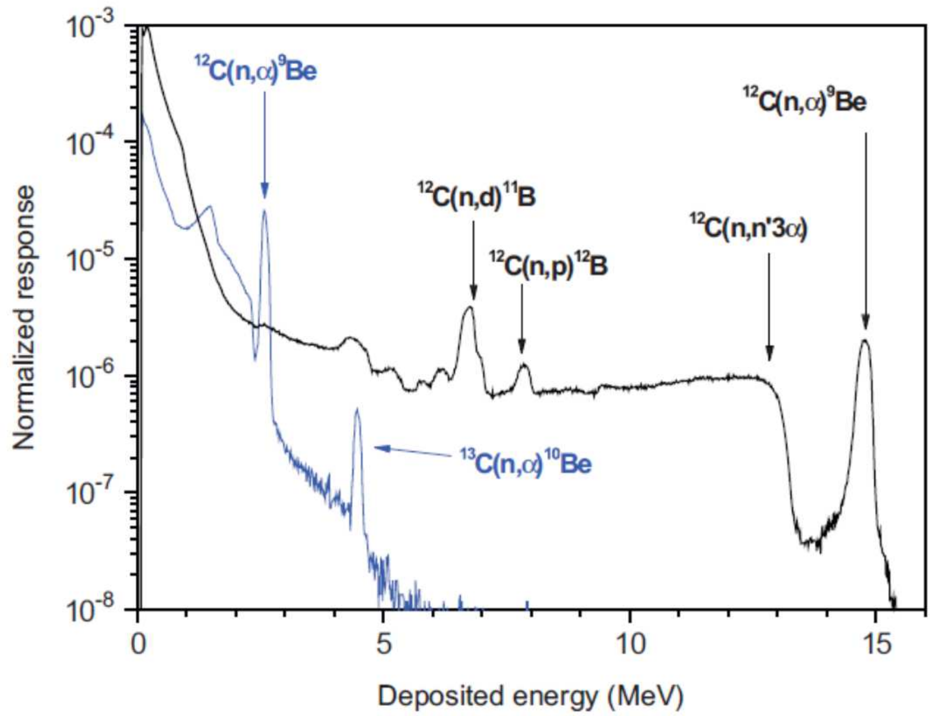


Verification of fuel assemblies
Detection system based on a EJ309 scintillator
^{3}He-alternative



Response functions of diamond detectors at JRC Geel





Pillon et al., NIMA 640 (2011) 185

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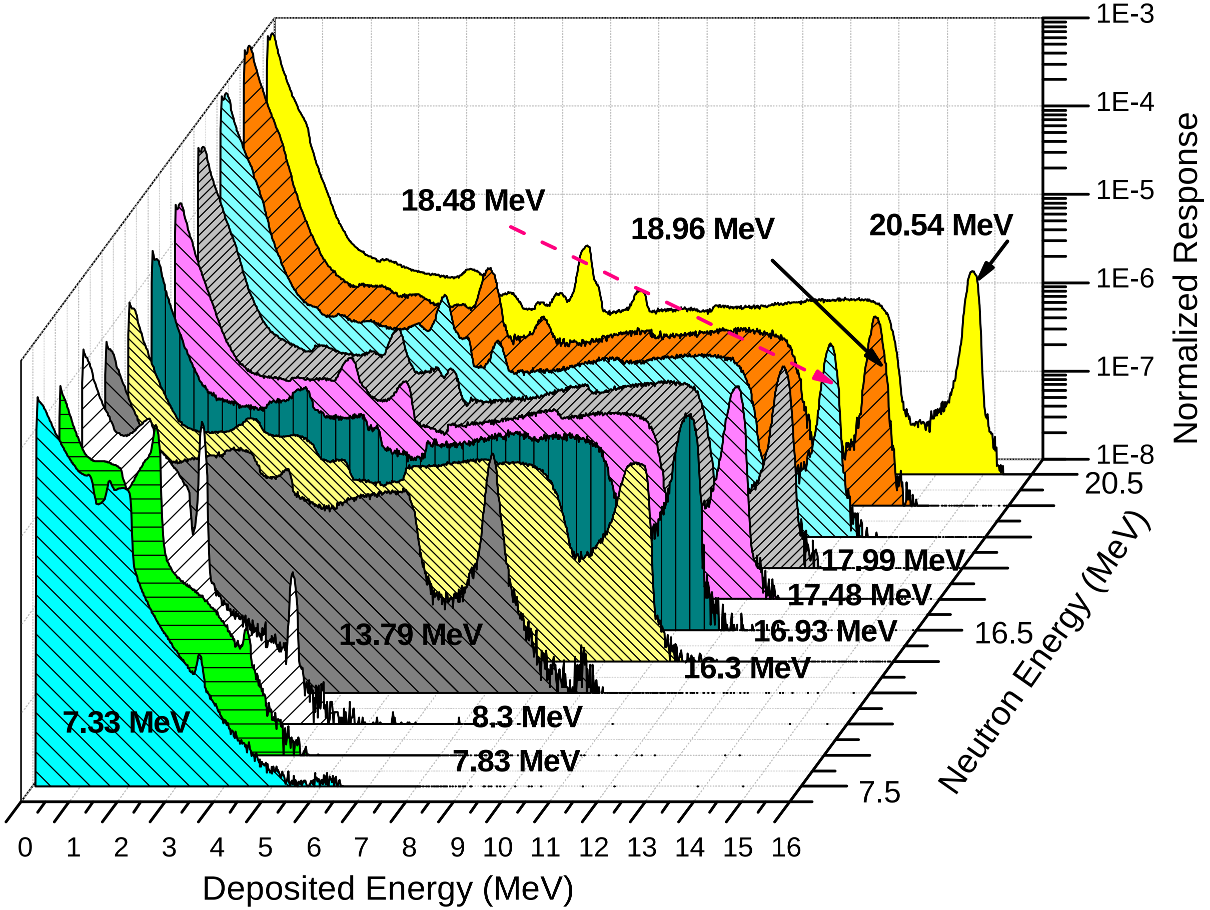
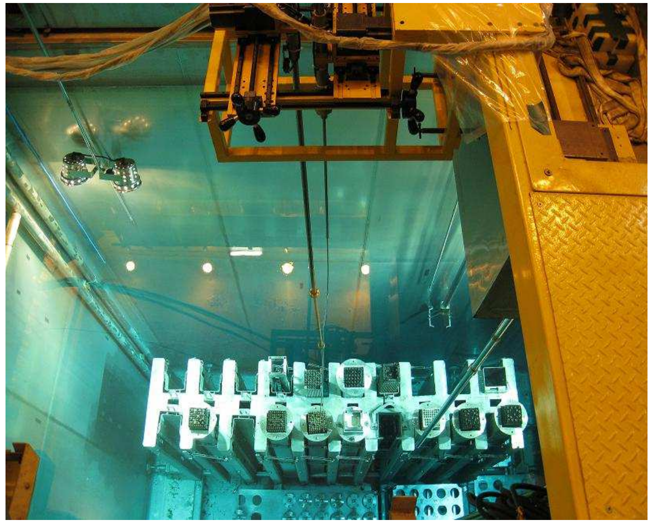
Diamond detectors



Use of diamond detectors for verification of spent fuel



- Nuclear safeguards
- Final disposal of spent fuel: Finland and Sweden

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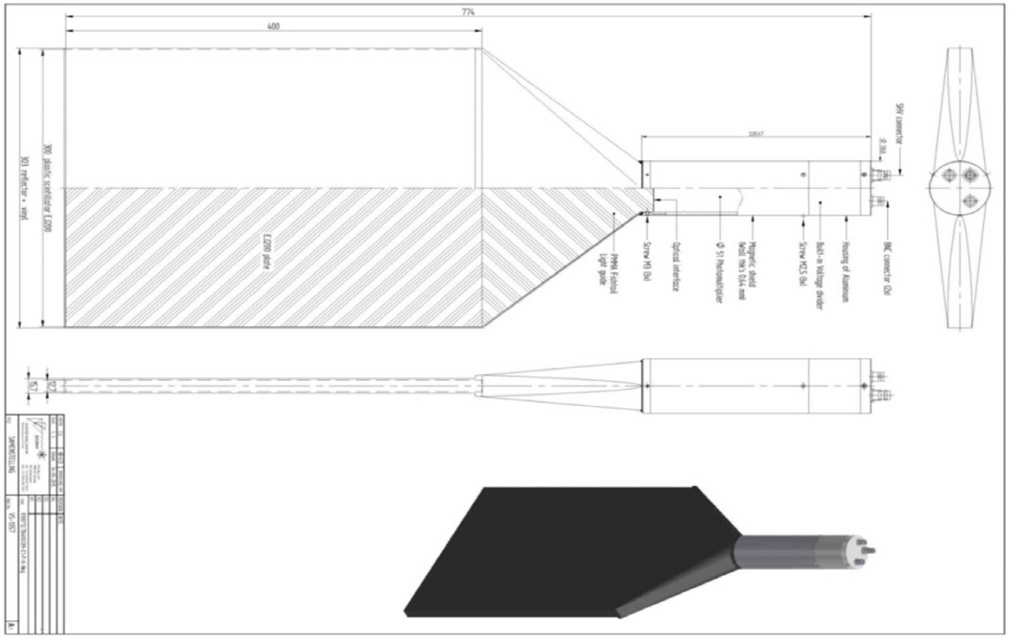
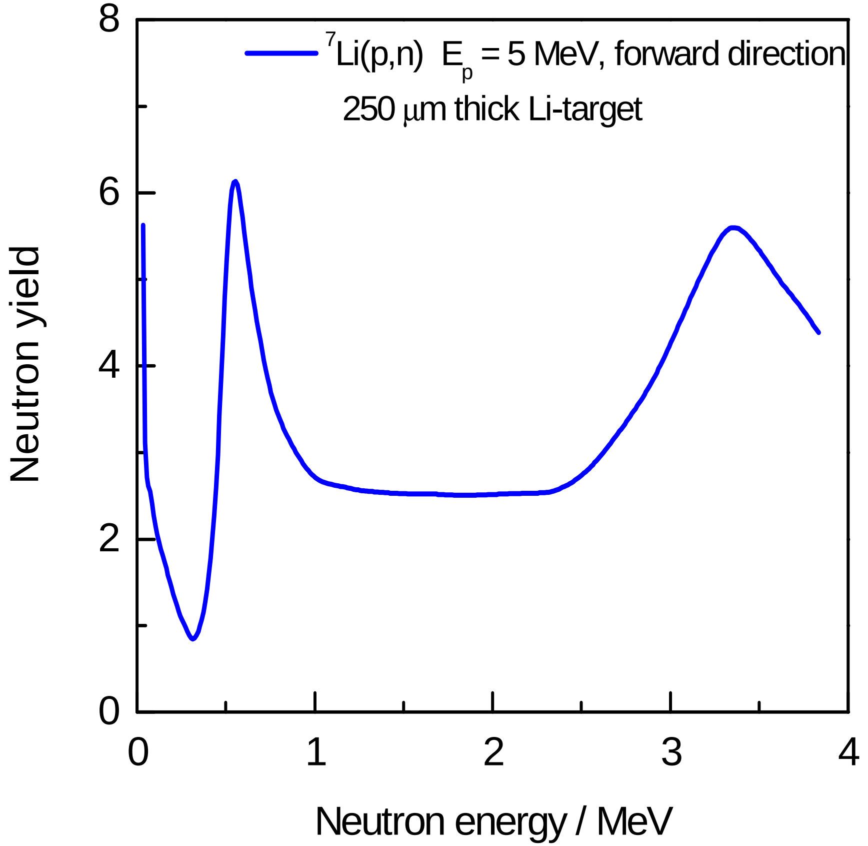
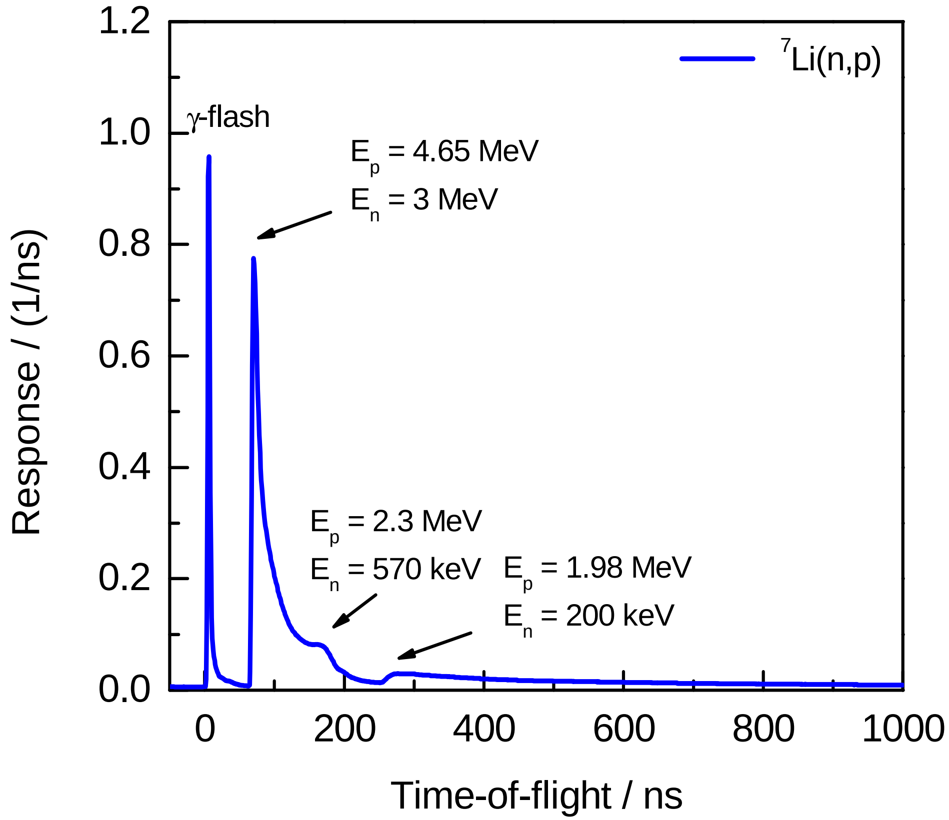
Response functions of a plastic scintillator at INFN Legnaro

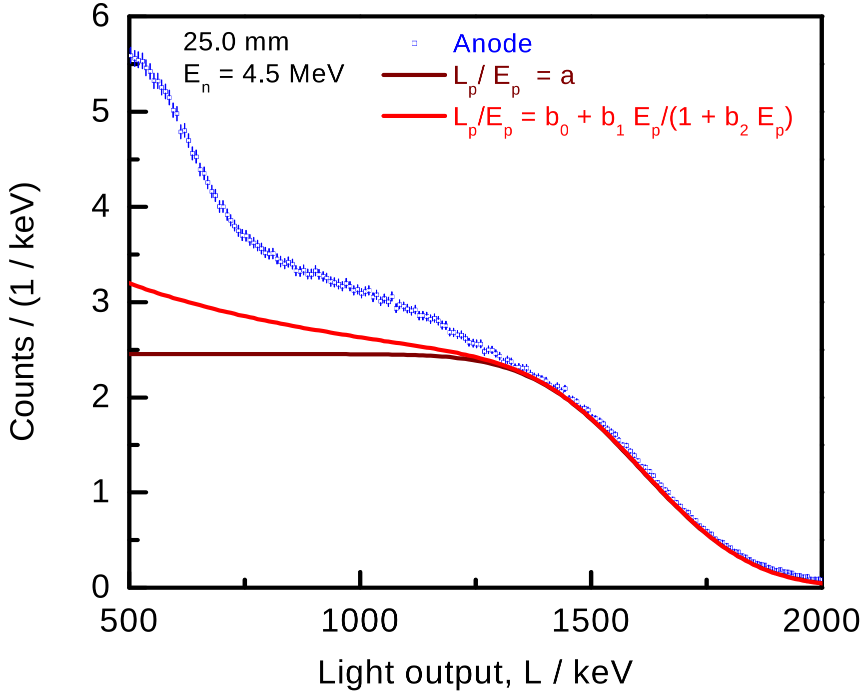


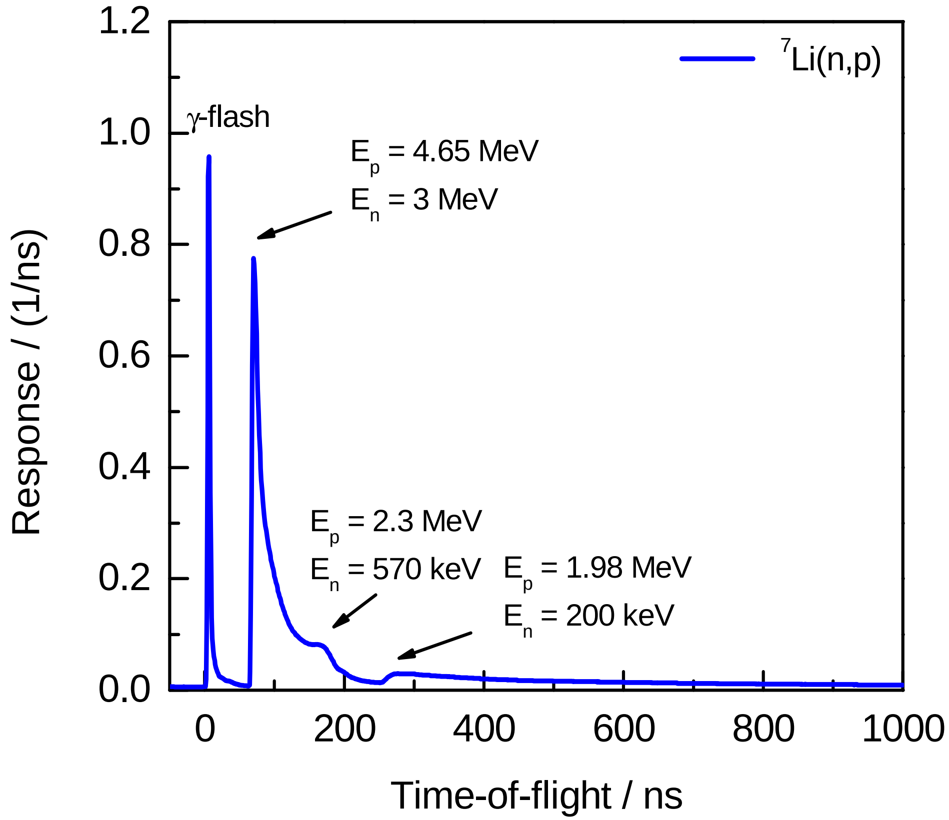
Response functions of a plastic scintillator at INFN Legnaro





Response functions of a plastic scintillator at INFN Legnaro





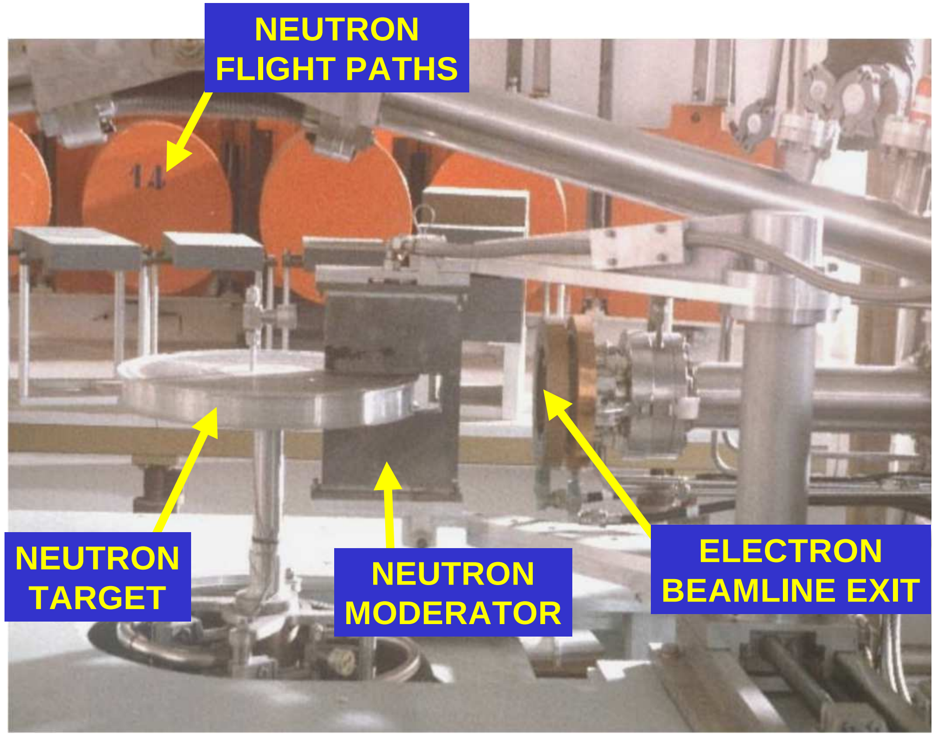
TOF – facility GELINA

- Pulsed white neutron source

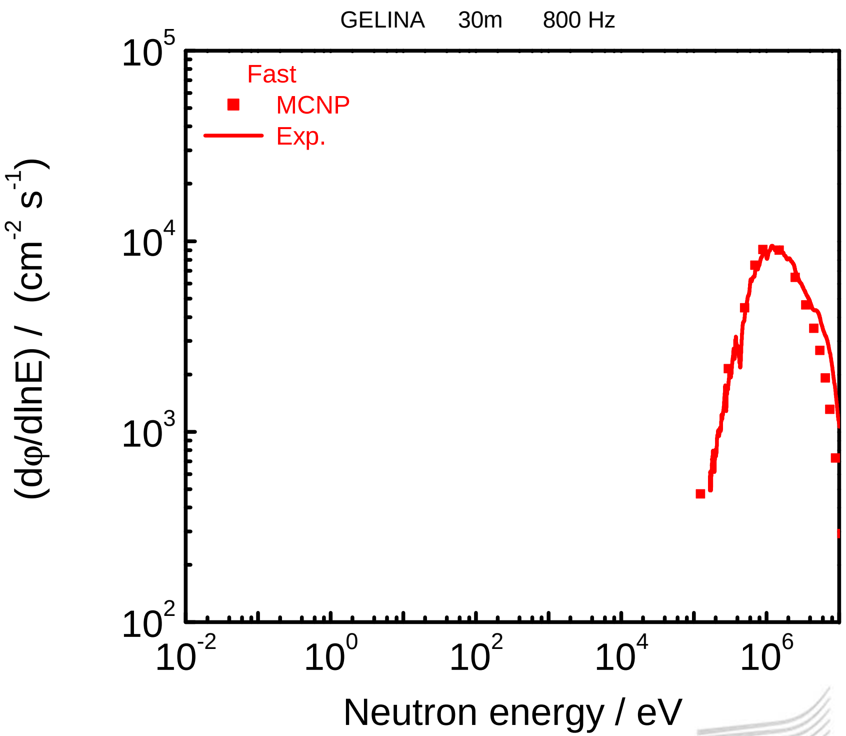
  (10 meV < E_n < 20 MeV)

- Neutron energy : time – of – flight (TOF)
- Multi-user facility: 10 flight paths (10 m – 400 m)
- Measurement stations with special equipment:
  - Total cross section measurements
  - Partial cross section measurements

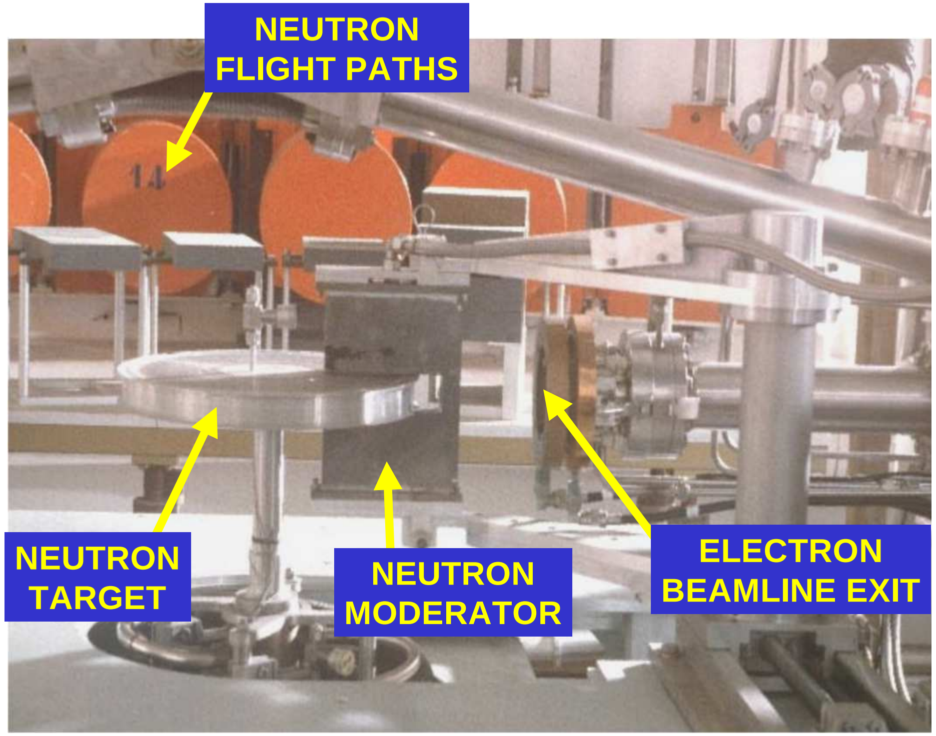
GELINA : neutron production



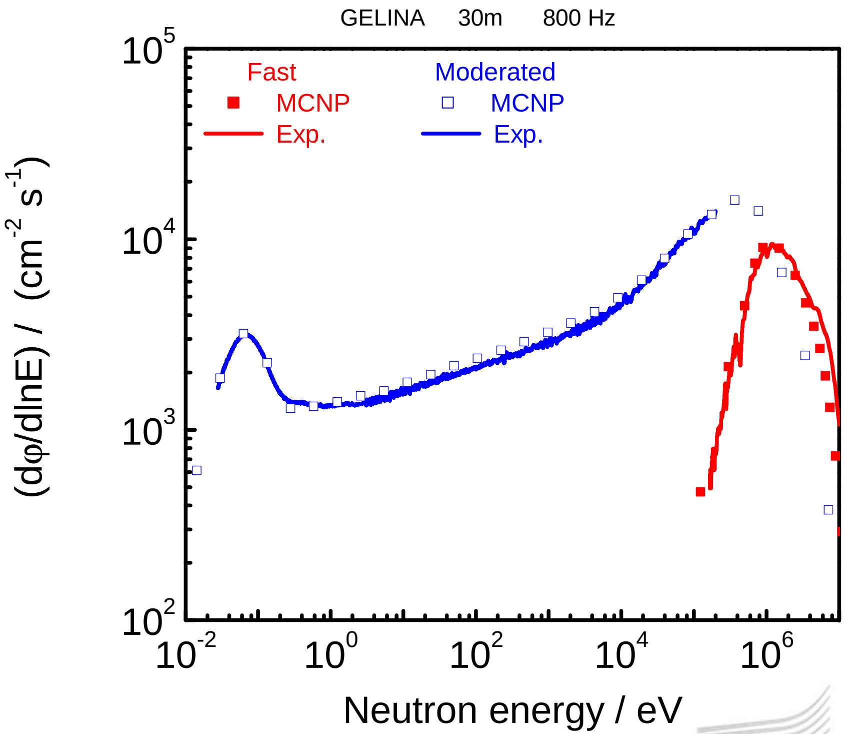
- e^- accelerated to $E_{e-,max} \approx 140$ MeV
- Bremsstrahlung in U-target (rotating & cooled with liquid Hg)
- (γ , n) , (γ , f) in U-target



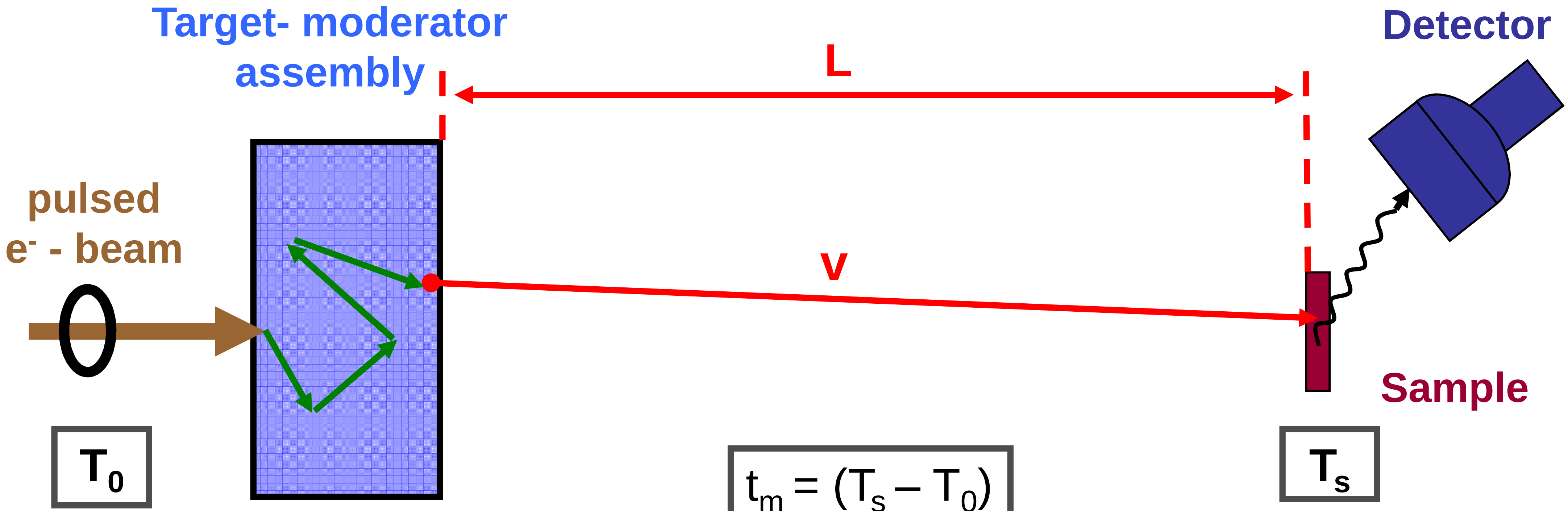
GELINA : neutron production



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



TOF - measurements

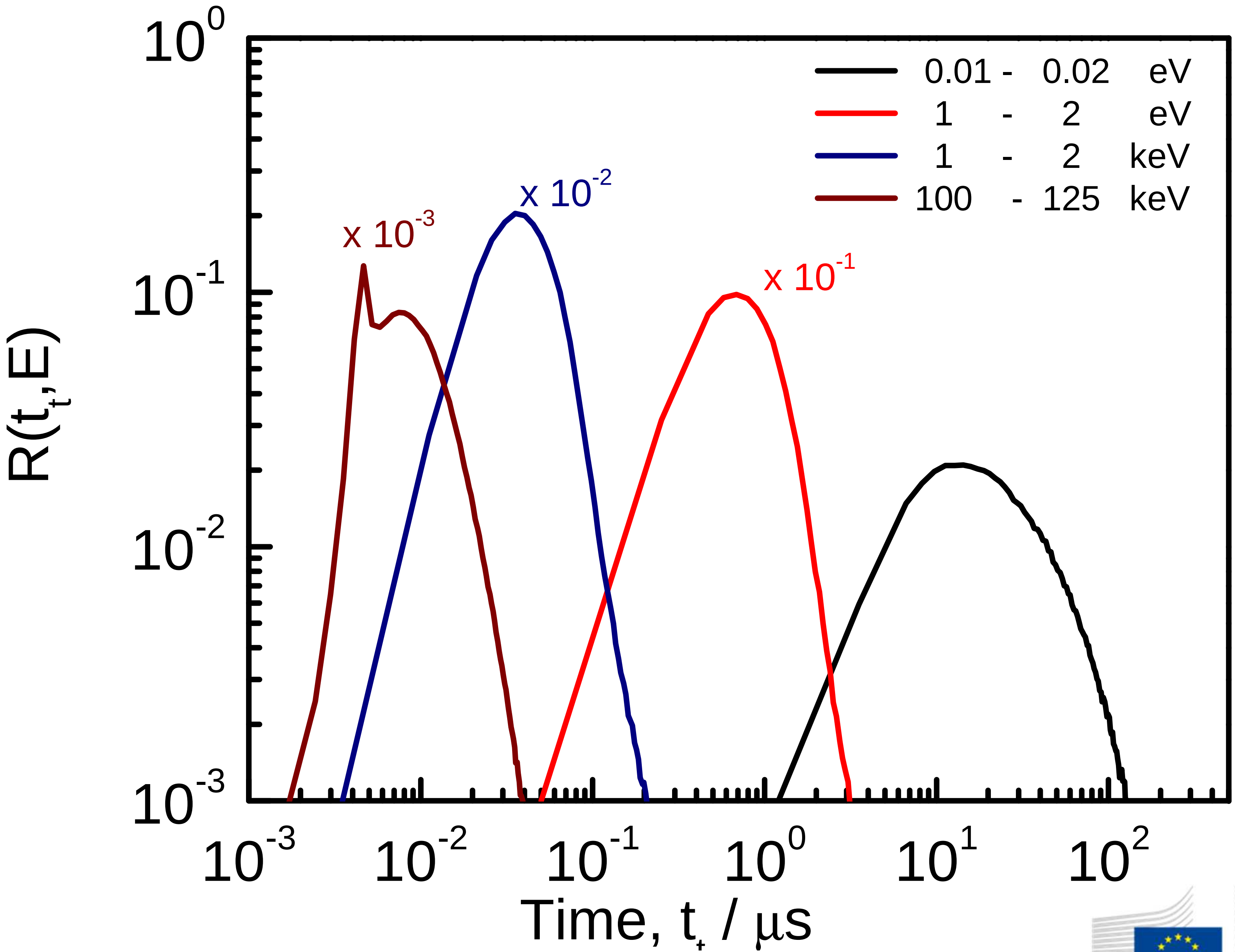


$$t_m = (T_s - T_0)$$

$$t = t_m - (t_t + t_d)$$

$$v = \frac{L}{t}$$

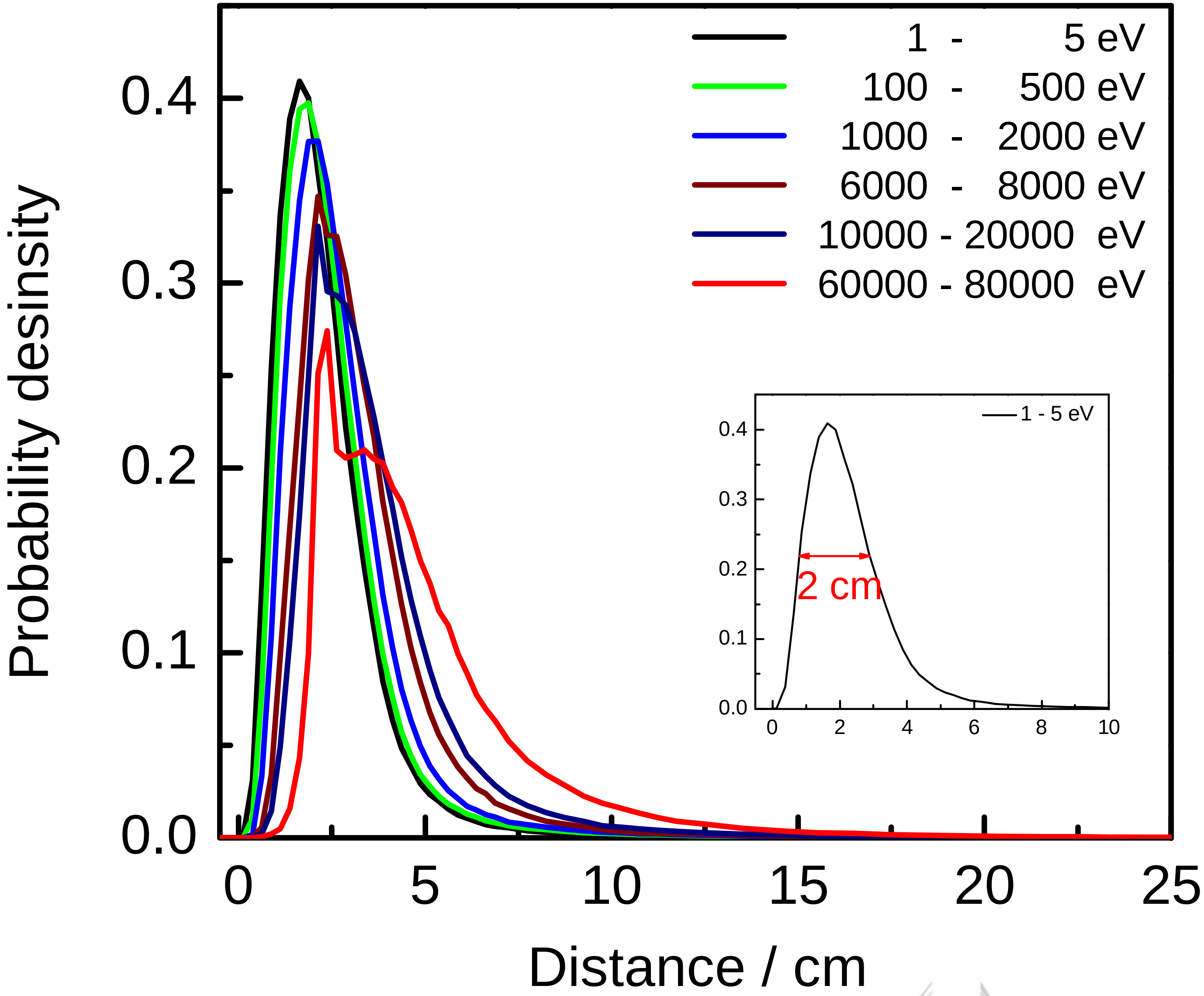
Probability distribution of t_t : GELINA



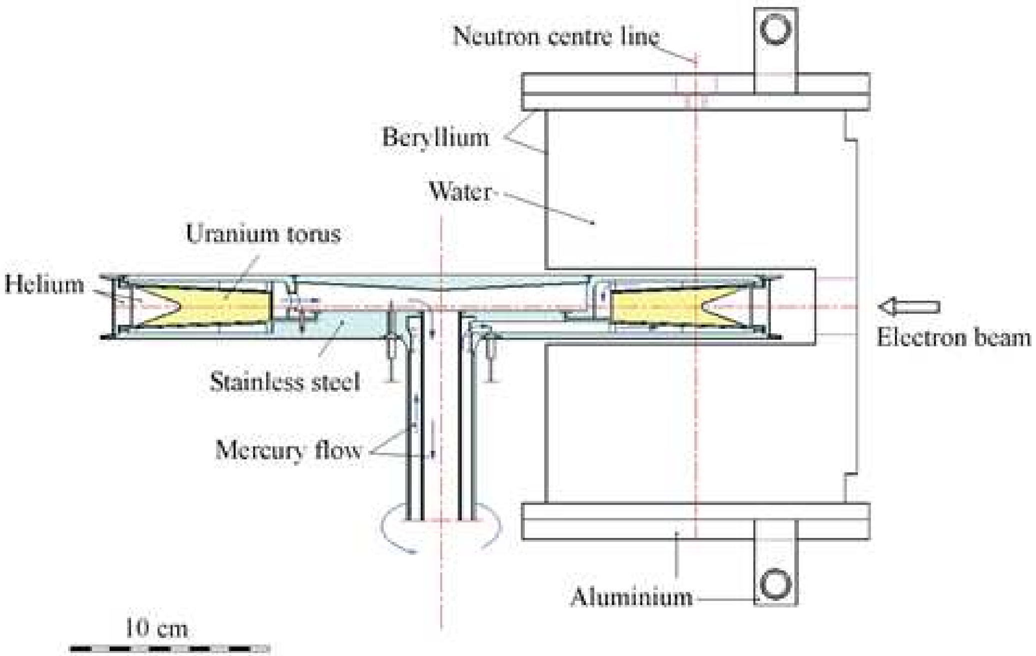
Equivalent distance : $L_t = v\, t_t$

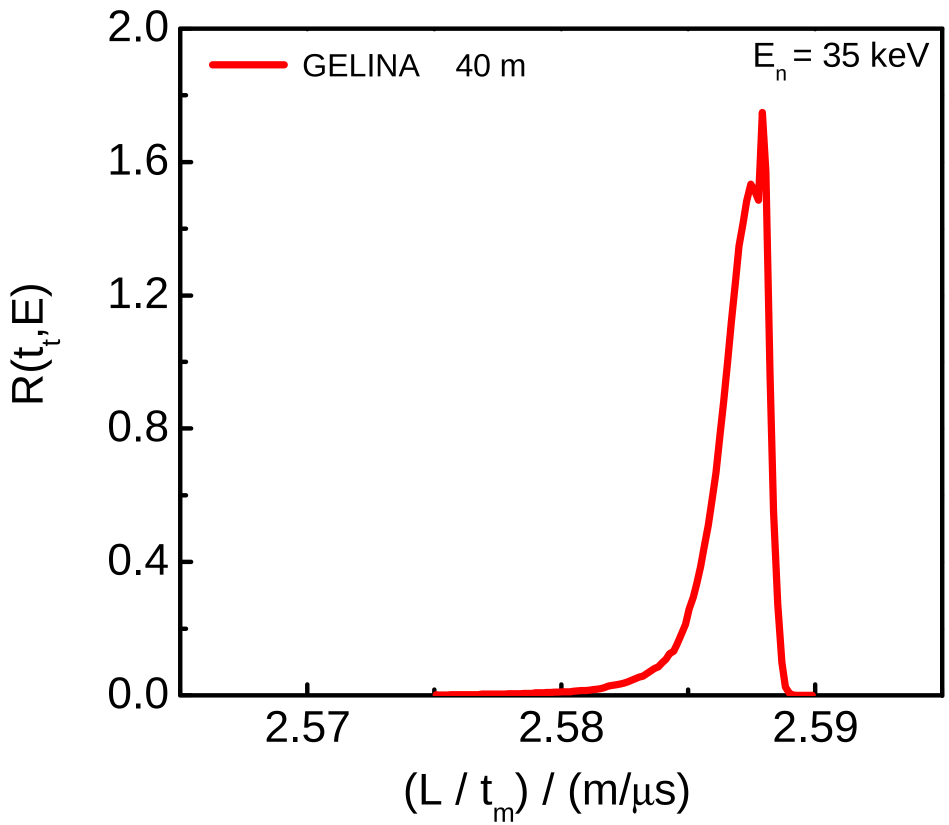
$$R(t_n, E_n) = R'(L(t_n), E_n)\left|\frac{dL}{dt_n}\right|$$

Flaska et al., NIMA 531(2004) 392



$P(t_t, E)$: photonuclear

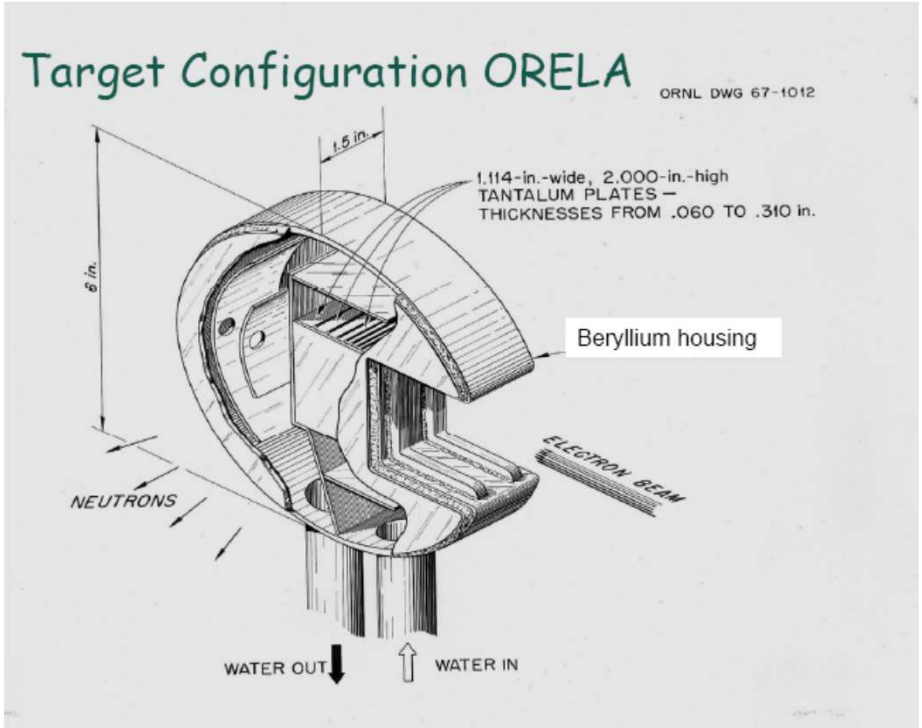


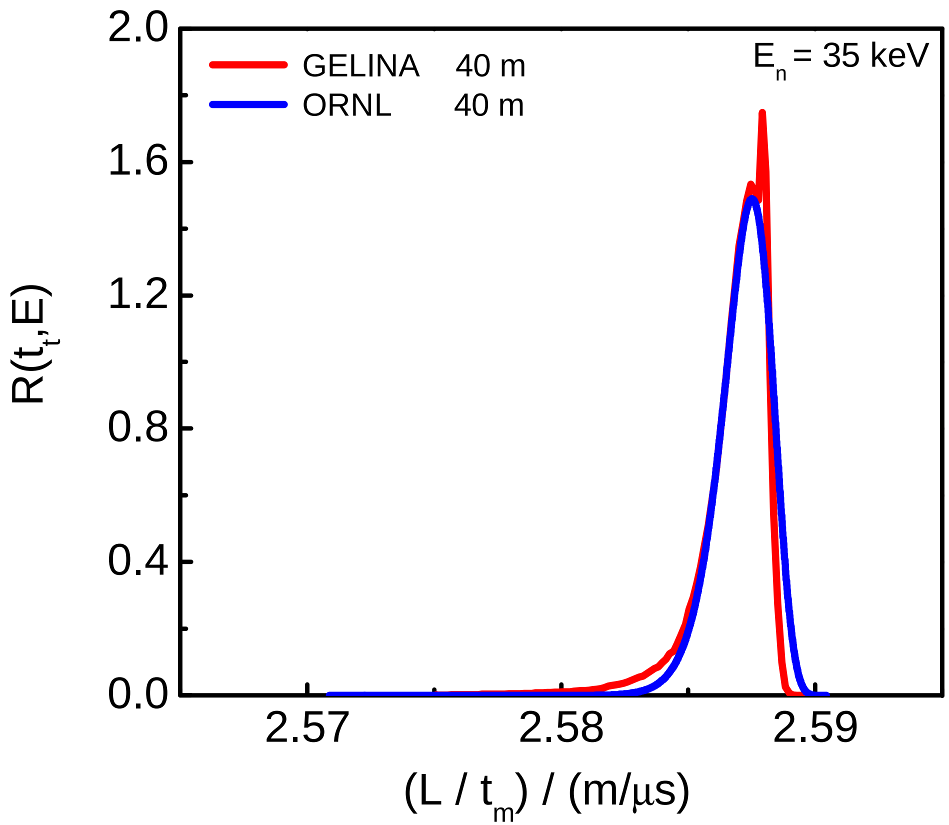


Resolution : ΔL

GELINA : 2 cm

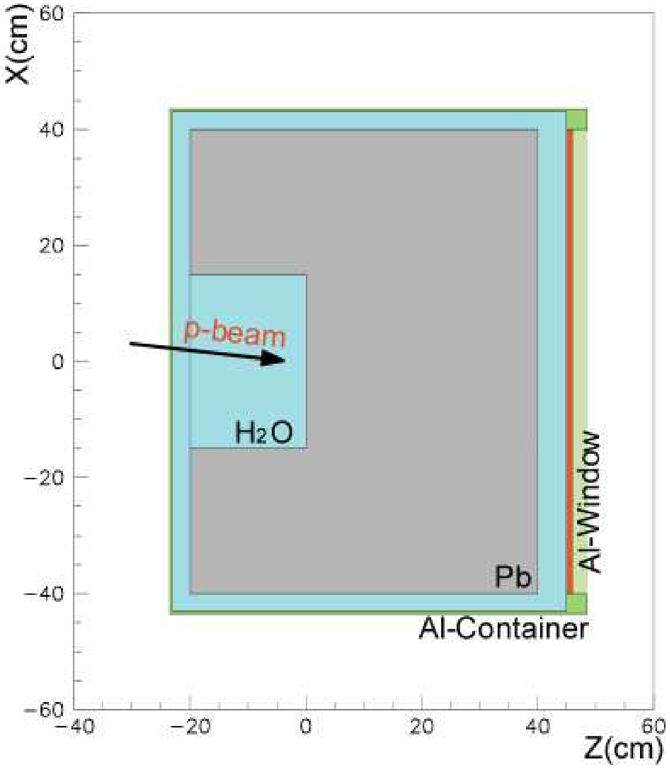
$P(t_t, E)$: photonuclear



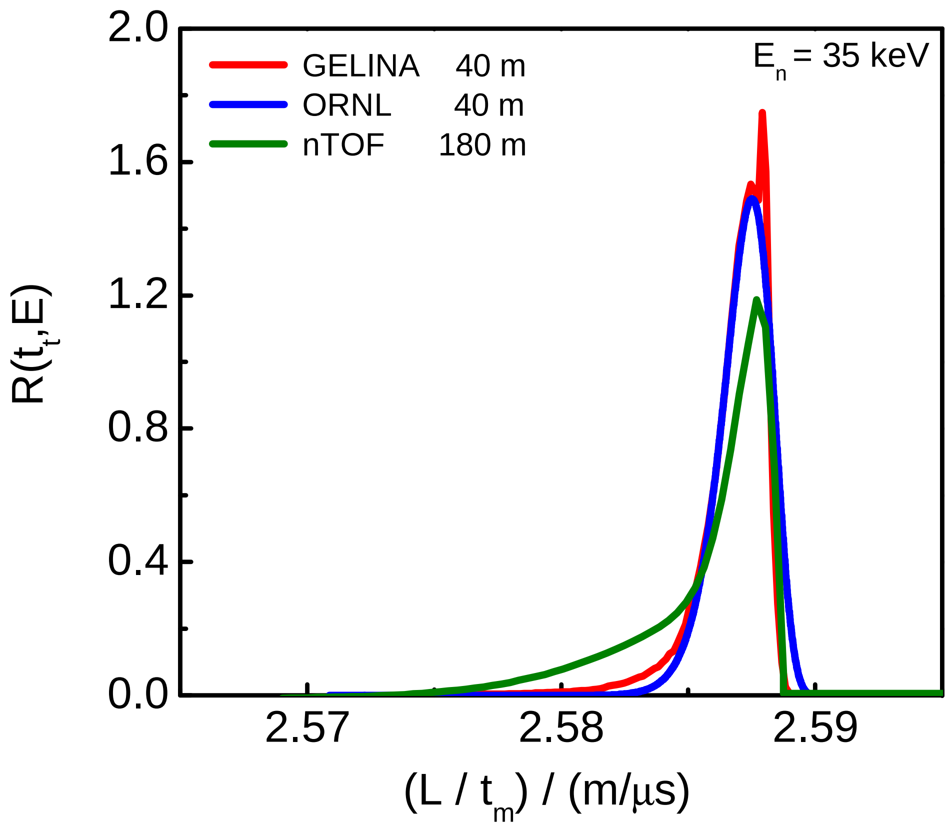


Resolution	:	ΔL
GELINA	:	2 cm
ORELA	:	2 cm

$P(t_t, E)$: photonuclear ⇔ spallation reactions



Dimensions	: $80 \times 80 \times 60$ cm^3
Pure Lead	: 4 t
H_2O moderator	: 5 cm
Al-window	: 1.6 mm
Al-container	: 140 l



Resolution	:	ΔL
GELINA	:	2 cm
ORELA	:	2 cm
nTOF	:	10 cm

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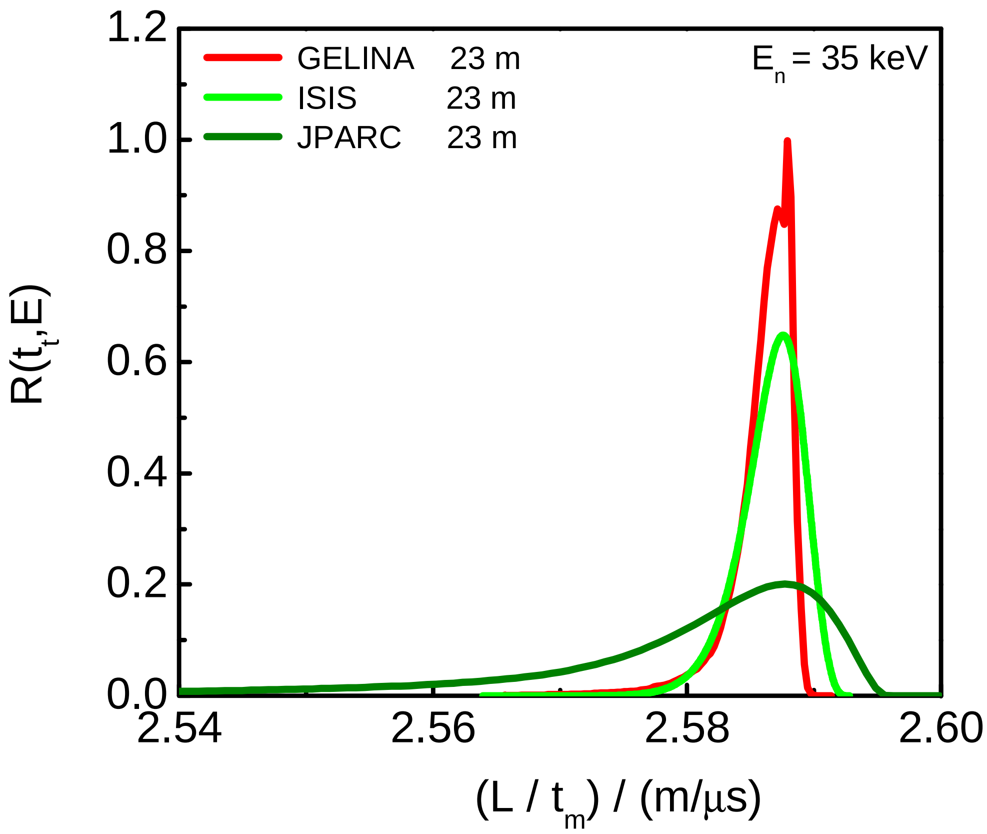
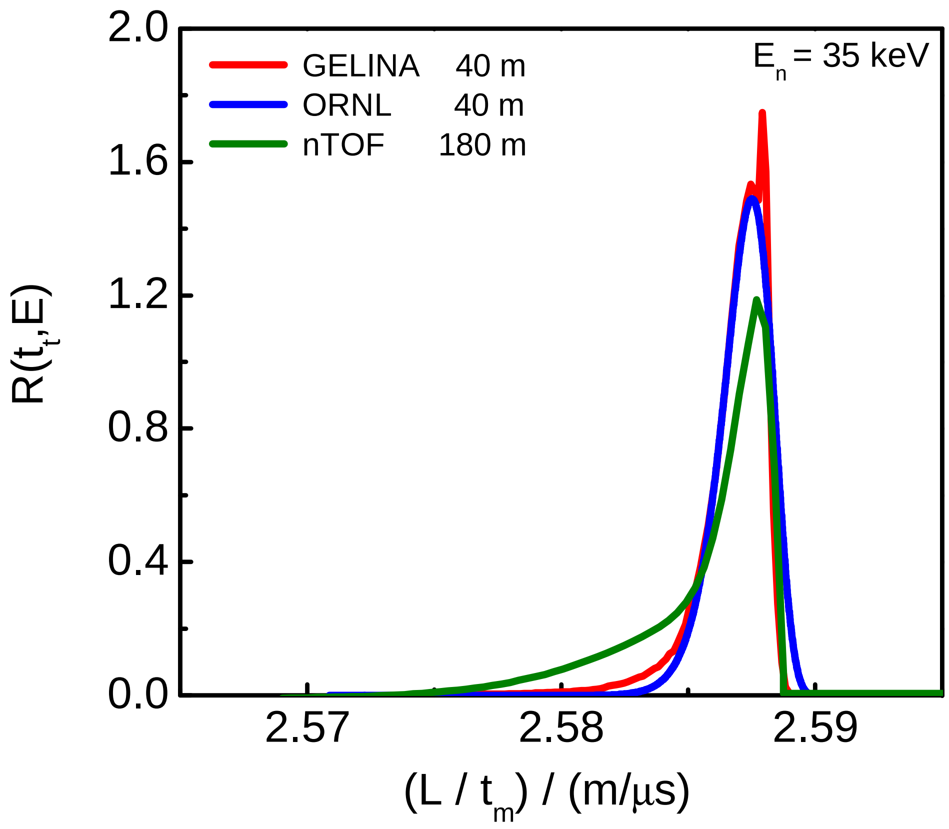
$P(t_t, E)$: photonuclear ⇔ spallation reactions

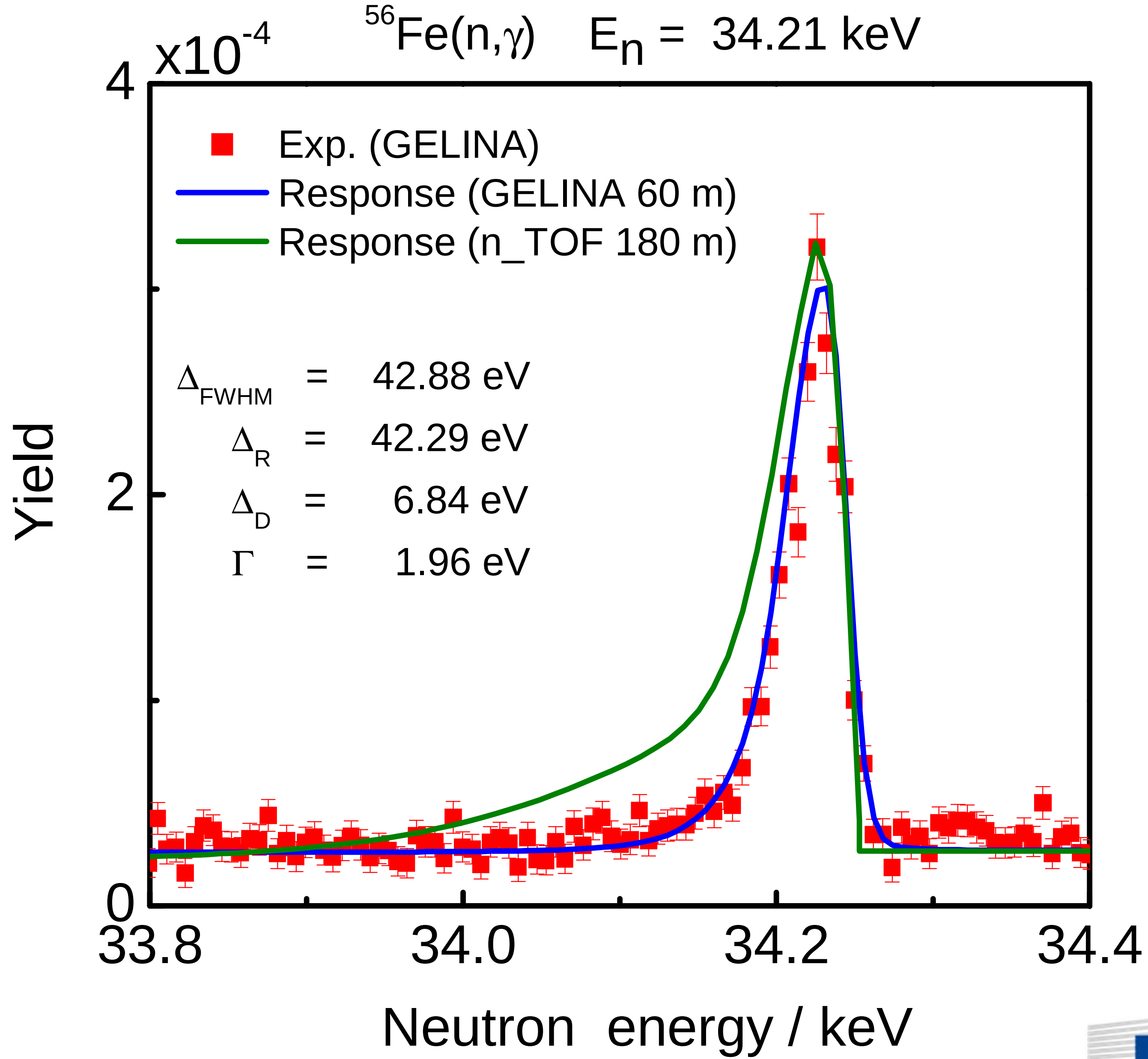




Resolution : ΔL
ISIS (INES) : 5 cm
J-PARC (MLF/ANNRI) : 13 cm
Strongly depend on target/moderator configuration (coupled/decoupled)

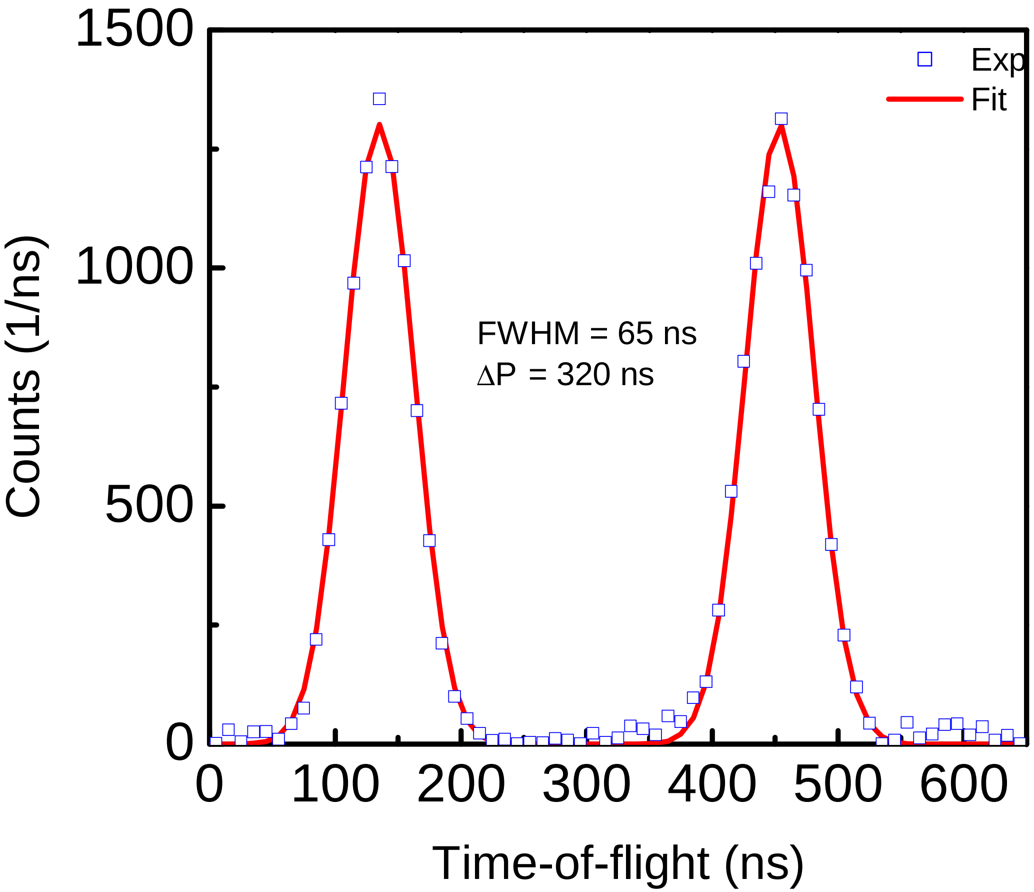
Resolution : ΔL
GELINA : 2 cm
ORELA : 2 cm
nTOF : 10 cm

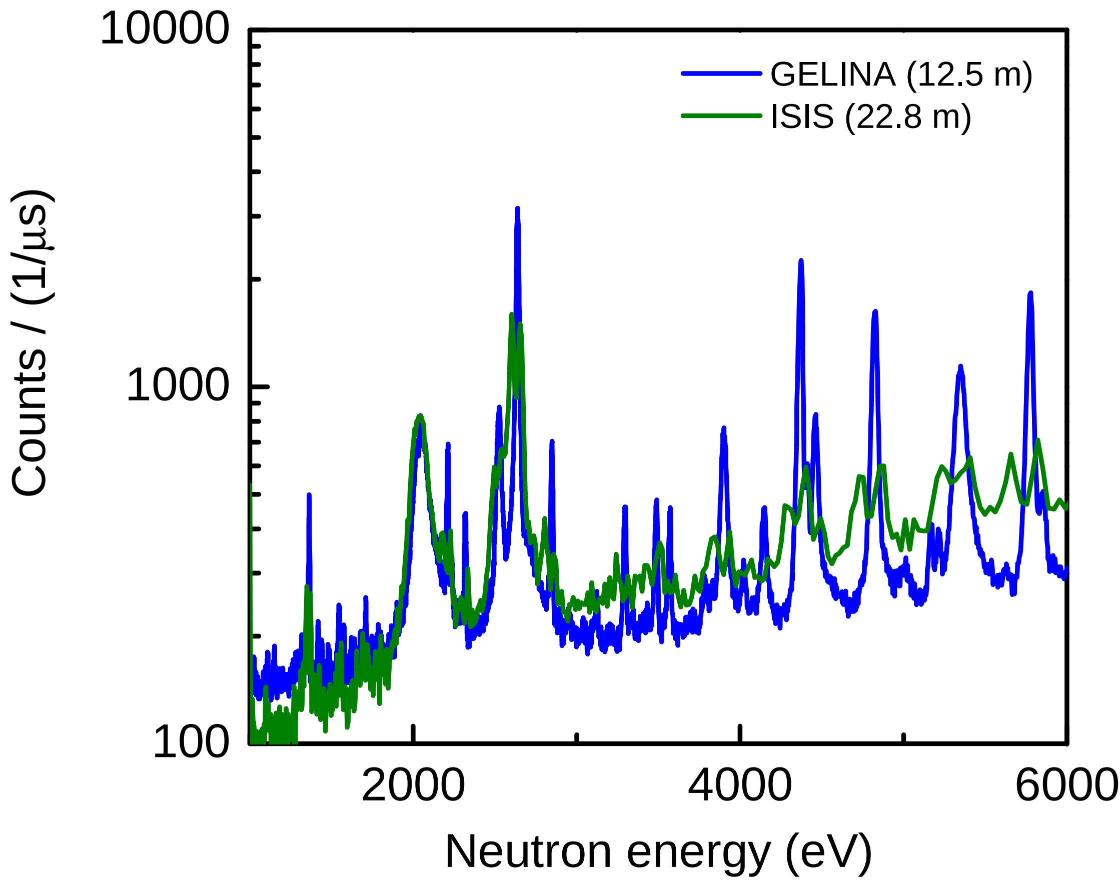
Response GELINA (60 m) < - > n_TOF (180 m)



Response GELINA (12.5 m) < - > ISIS (22.8 m)

Double pulse proton beam at ISIS



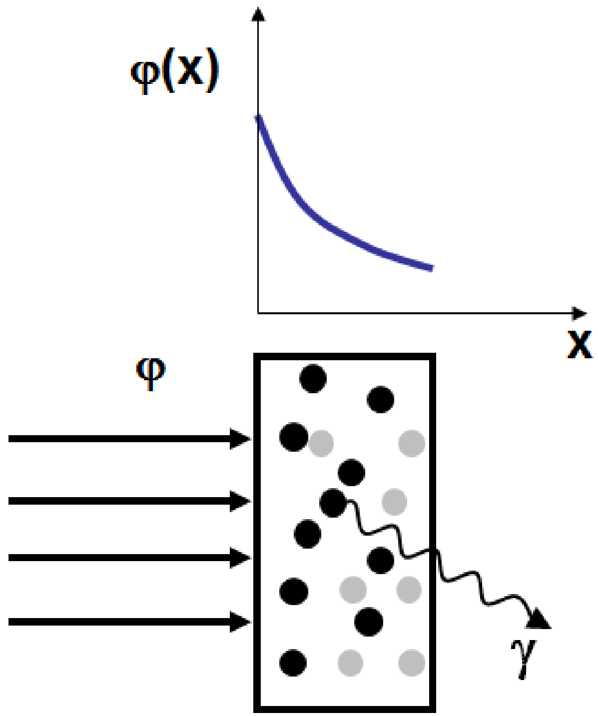


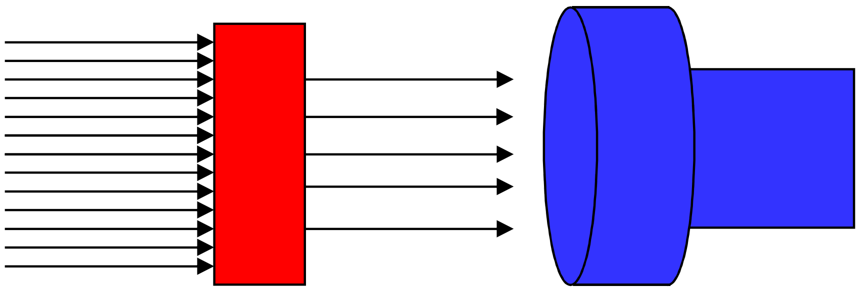
Cross section measurements

Transmission

$$T \cong e^{-n\sigma_{tot}}$$

T : transmission
Fraction of the neutron beam traversing the sample without any interaction





$$n = \frac{N_A m}{m_a A}$$

- n : areal density
- N_A : Number of Avogadro
- m : material mass
- A : sample area
- m_a : atomic mass

European Commission

Cross section measurements

Transmission

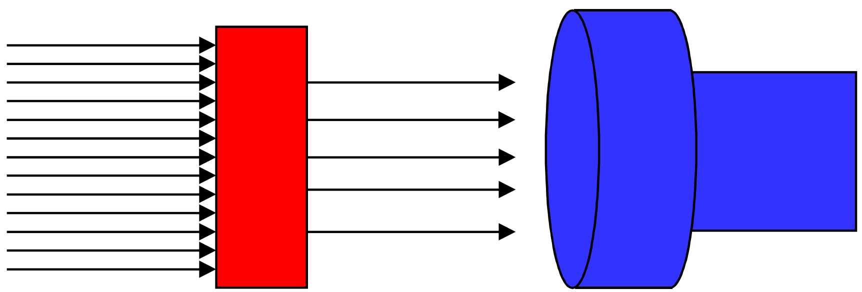
$$T \cong e^{-n\sigma_{tot}}$$

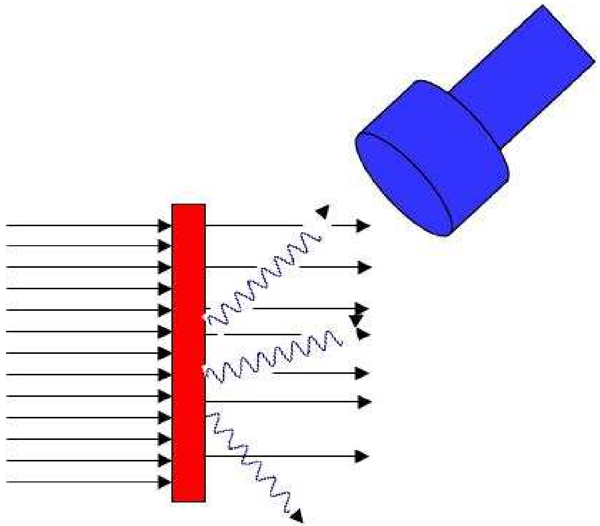
T : transmission
Fraction of the neutron beam traversing the sample without any interaction

Reaction

$$Y_r \approx (1 - e^{-n\sigma_{tot}})\frac{\sigma_r}{\sigma_{tot}}$$

Y_r : reaction yield
Fraction of the neutron beam creating a (n,r) reaction in the sample





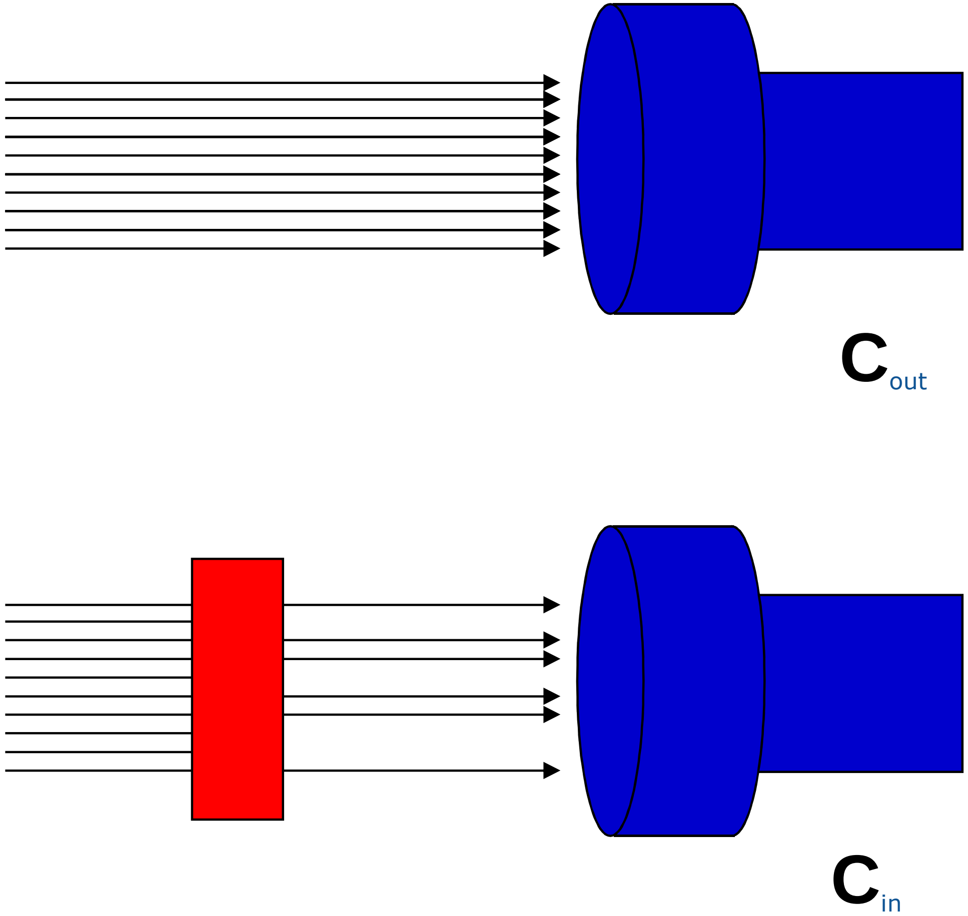
Transmission measurements

Transmission

$$T_{exp} = \frac{C_{in}}{C_{out}} \propto e^{-n\sigma_{tot}}$$

- Incoming neutron flux cancels
- Detection efficiency cancels

$\Rightarrow$ **Direct relation between T_{exp} and σ_{tot}**



Transmission : principle

$$T_{exp} = \frac{C_{in}}{C_{out}} \propto e^{-n\sigma_{tot}}$$

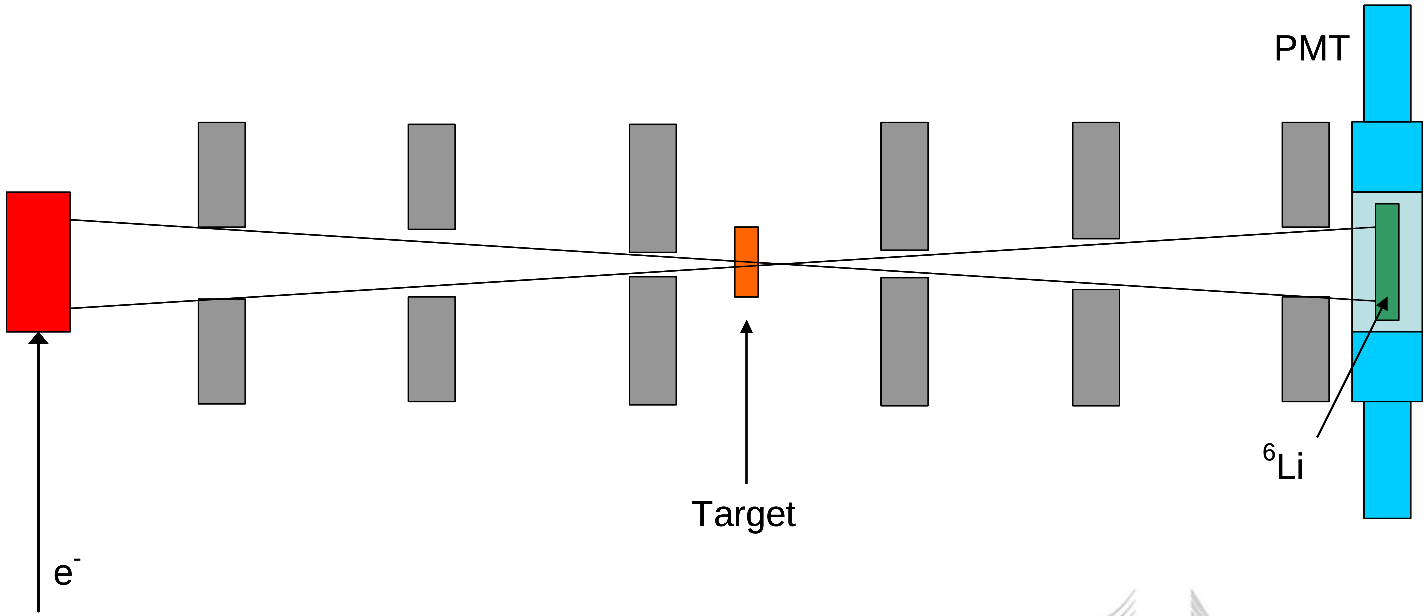
(1) All detected neutrons passed through the sample

(2) Neutrons scattered in the target do not reach detector

(3) Sample perpendicular to parallel neutron beam

$\Rightarrow$ Good transmission geometry (collimation)

(4) Homogeneous target (no spatial distribution of n)



European Commission

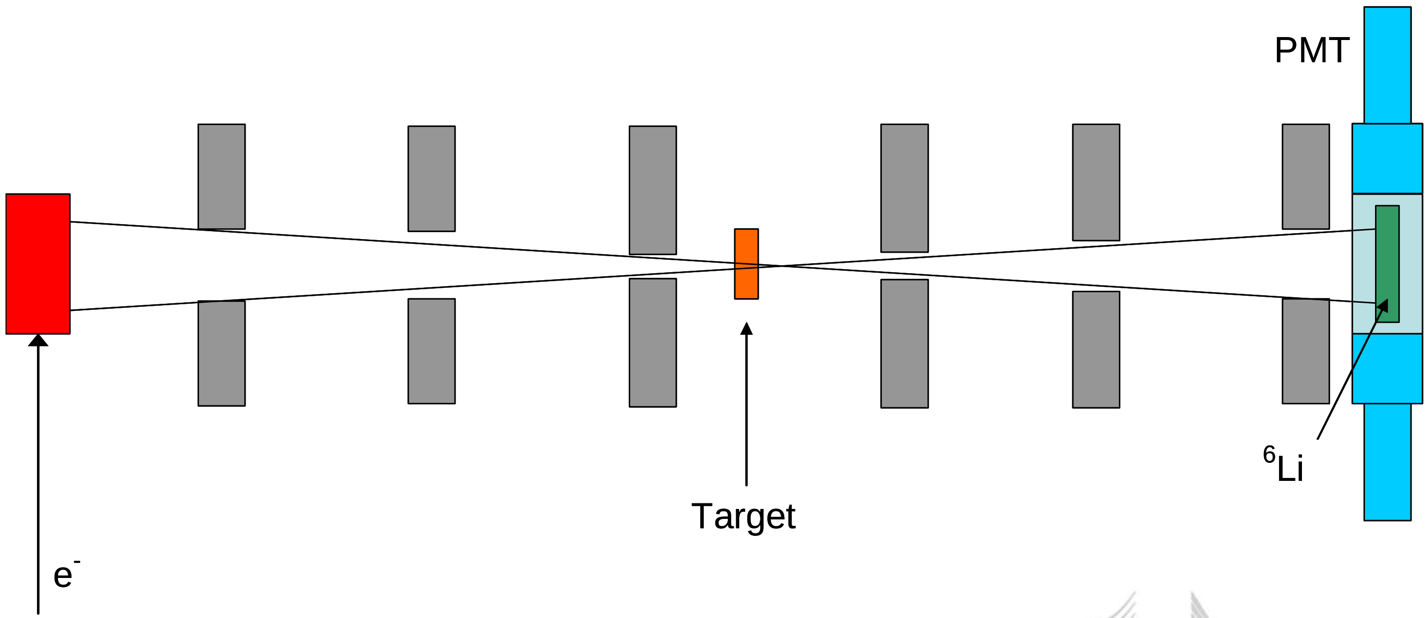
Transmission : principle

$$T_{exp} = \frac{C_{in}}{C_{out}} \propto e^{-n\sigma_{tot}}$$

Detectors

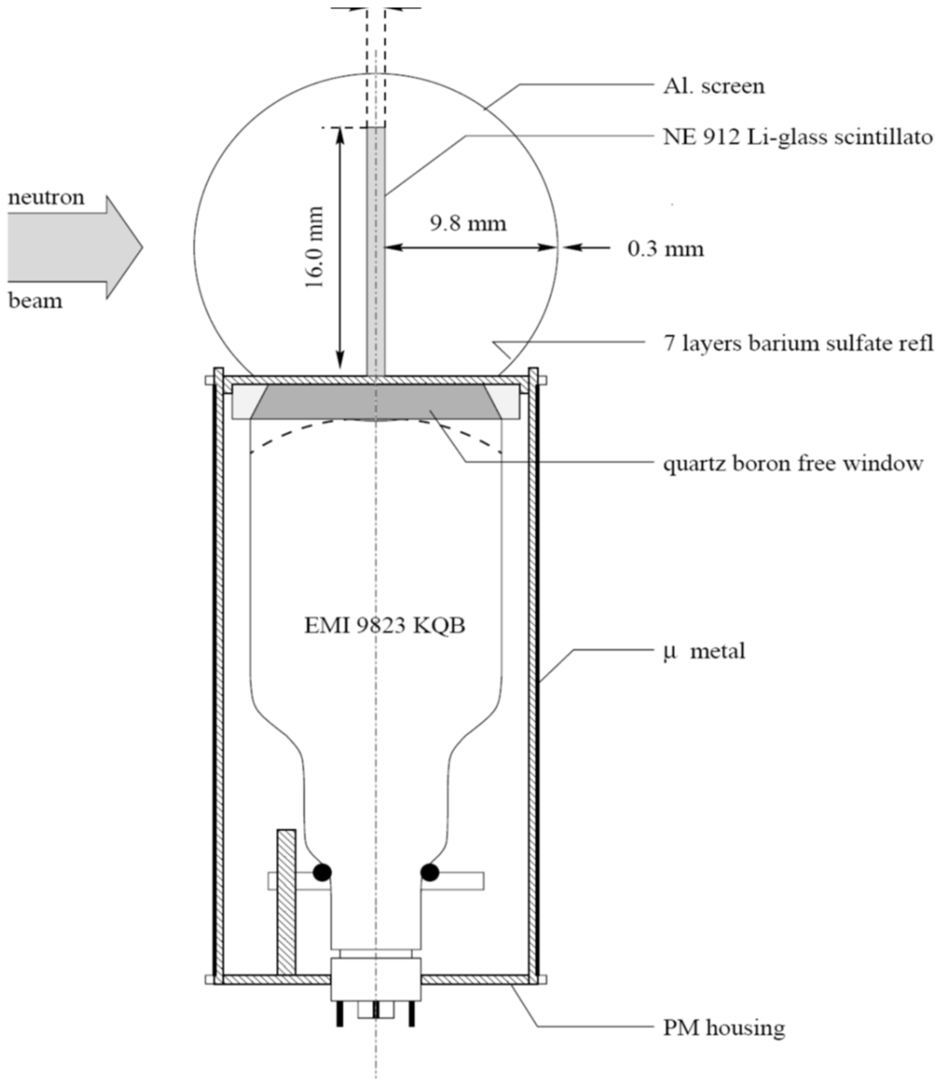
Low energy : $^{6}Li(n,t)\alpha$ Li-glass

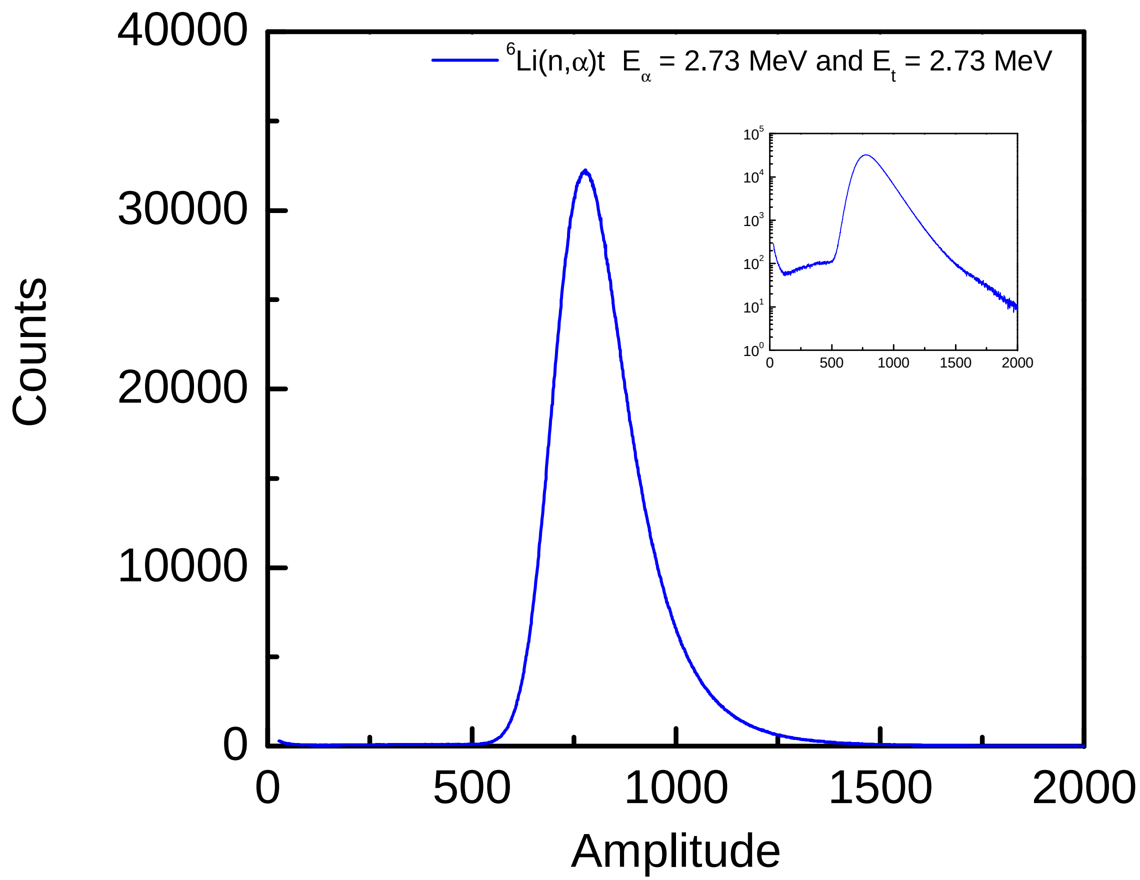
High energy : H(n,n)H NE213 type, plastic scintillator



Lithium-glass scintillator : energy deposition

$^{6}Li(n,t)\alpha$
Scintillator + PMT

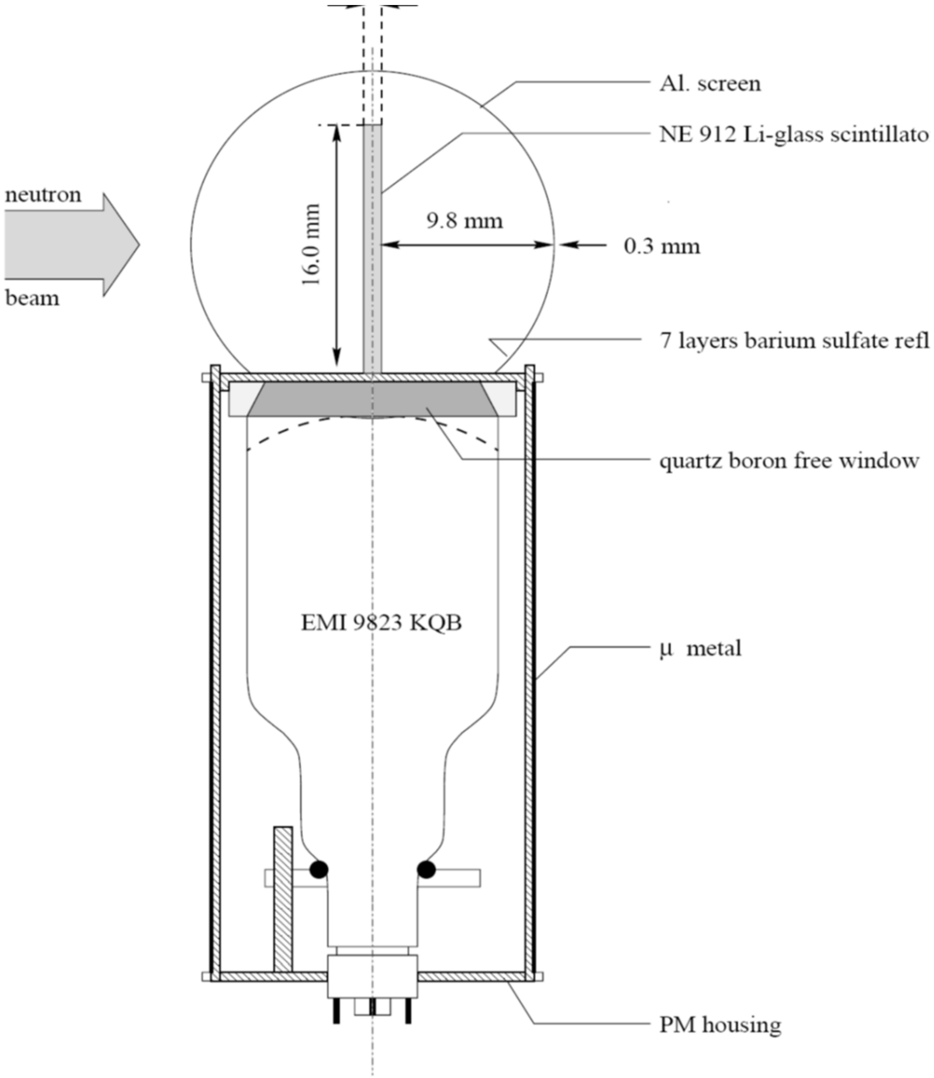


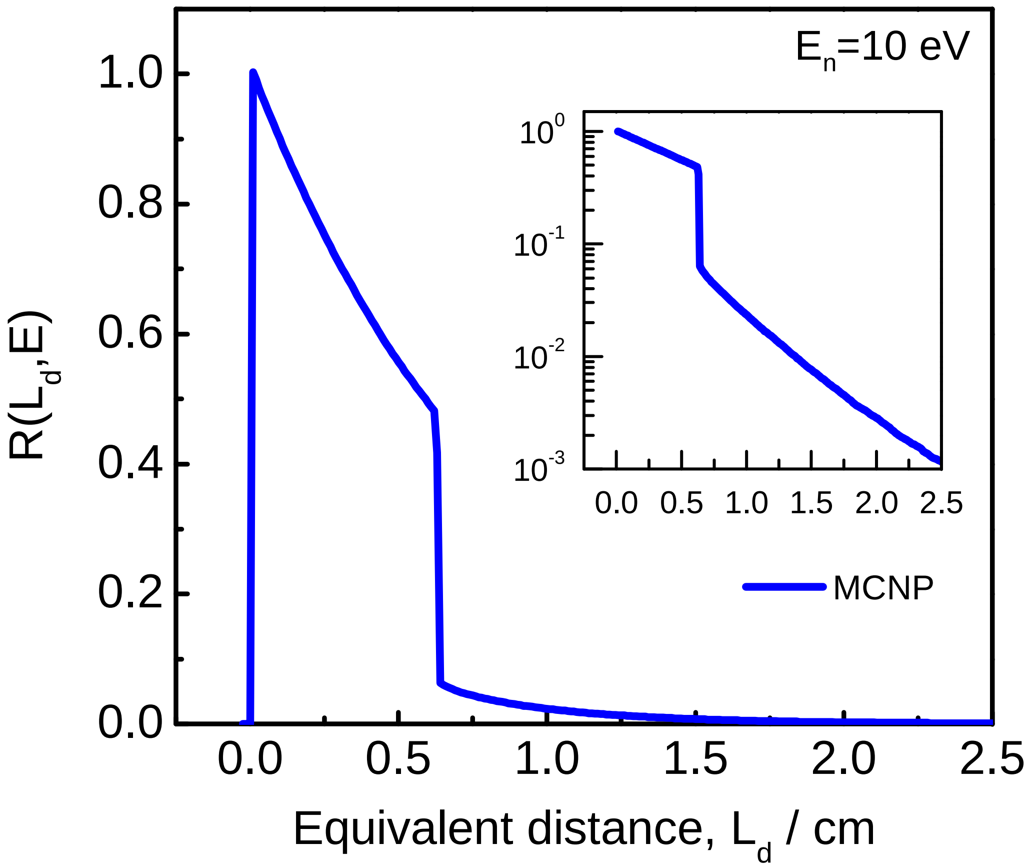


Lithium-glass scintillator : resolution

$^{6}Li(n,t)\alpha$

Scintillator + PMT





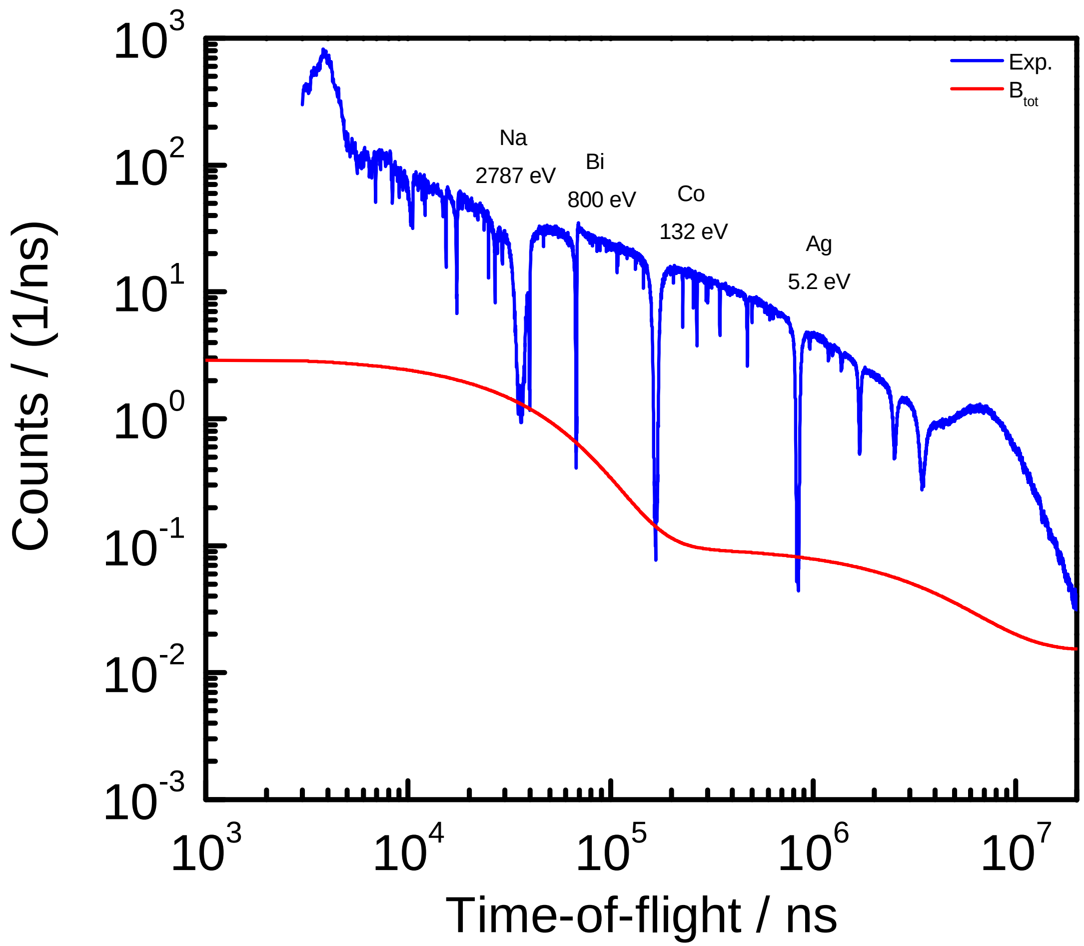
Transmission station at GELINA

^{6}Li detector

Castle

Neutron target + moderators

Background: black resonance technique



Black resonance filter

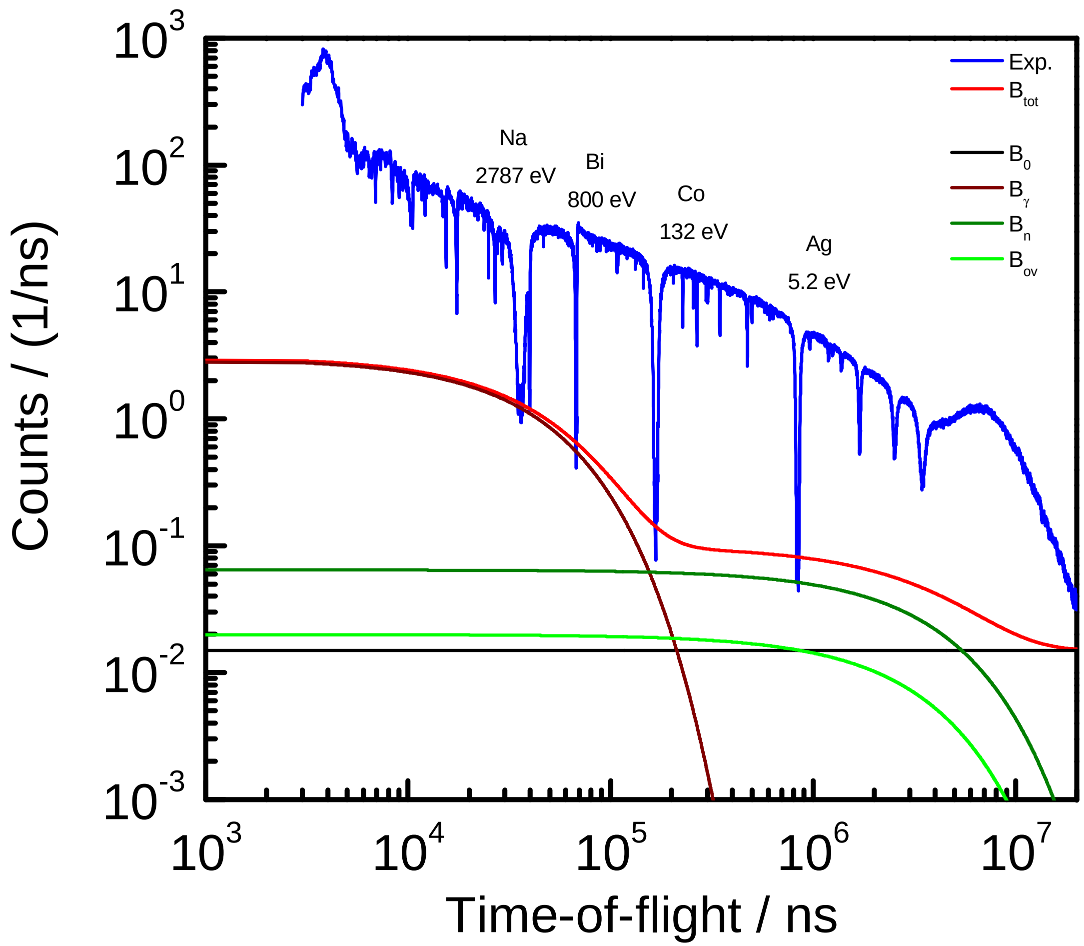
- strong resonance at E_r

$$T = e^{-n\sigma_{tot}} \approx 0$$

- removes all neutrons at TOF corresponding to E_r

$\Rightarrow$ Remaining counts are due to background

Background: black resonance technique

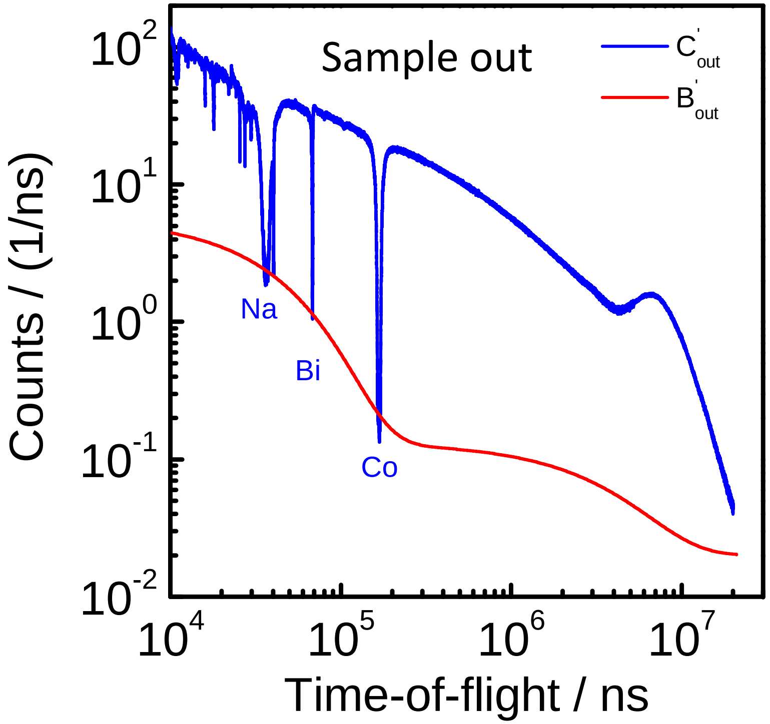


$B(t) = B_0 + B_\gamma(t) + B_n(t) + B_{ov}(t)$

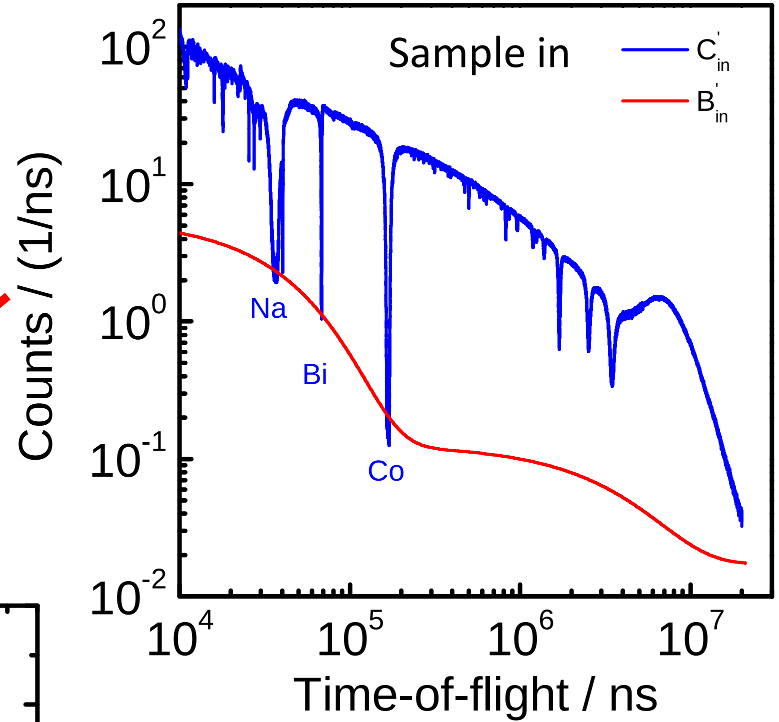
- B_o time independent
- $B_\gamma(t)$ $^1H(n, \gamma)$ $E_\gamma = 2.2$ MeV
  $b_1 e^{-\lambda_1 t}$
- $B_n(t)$ scattered neutrons
  $b_2 e^{-\lambda_2 t}$
- $B_{ov}(t)$ overlap neutrons
  $b_3 e^{-\lambda_3 (t+\tau_0)}$

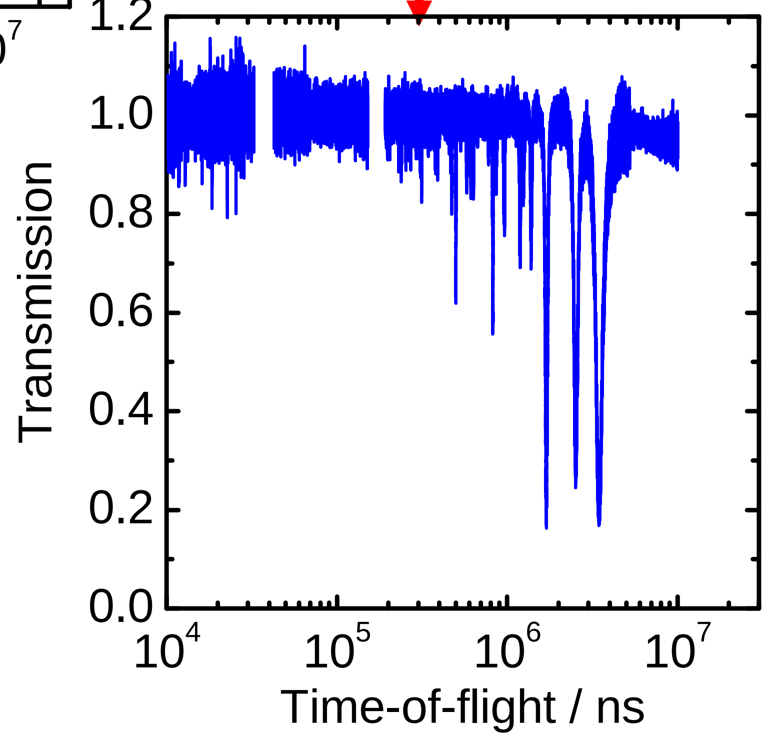
Shape from measurements at lower frequency or extrapolation at high TOF-values

Transmission data : ^{241}Am + n



$$T_{exp} = \frac{C'_{in} - B'_{in}}{C'_{out} - B'_{out}}$$





$$\frac{u_{T_{exp}}}{T_{exp}} \approx 0.25\%$$

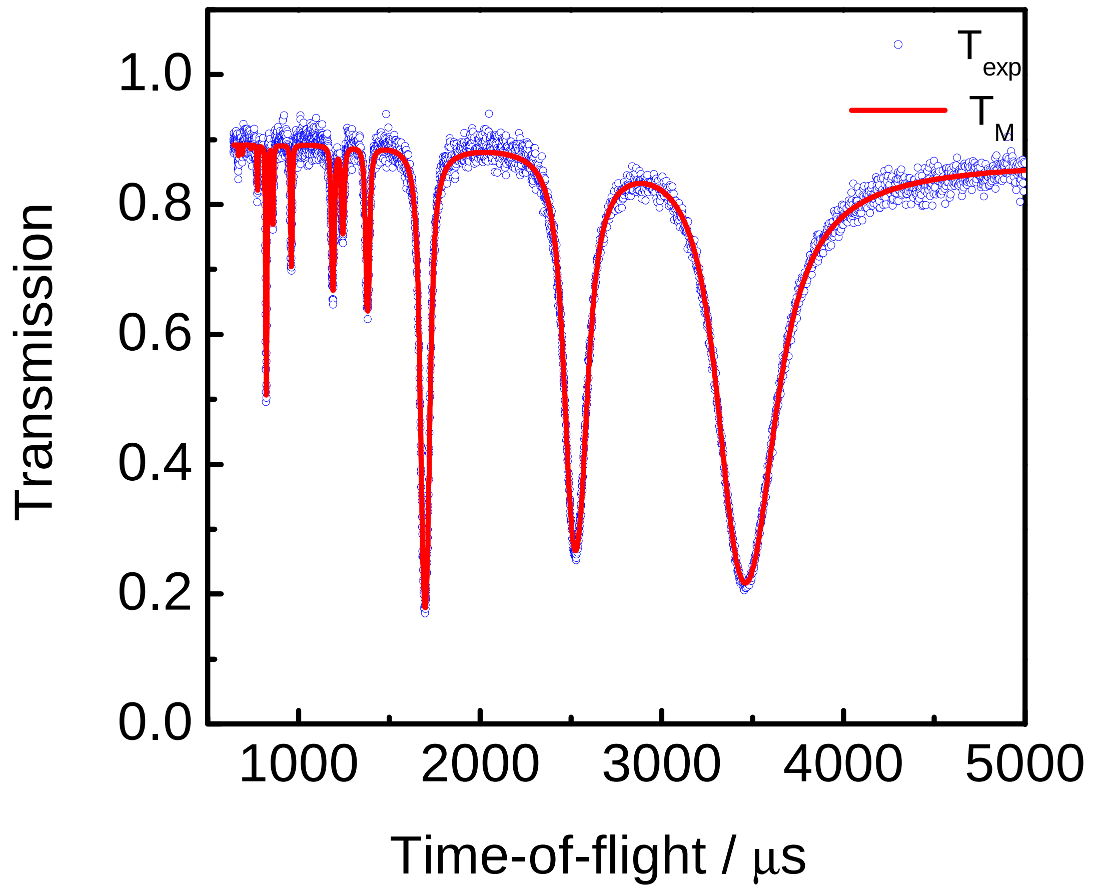
Transmission data : ^{241}Am + n

Transmission : fraction of neutron beam traversing without any interaction the sample

$$T_{exp} = \frac{C'_{in} - B'_{in}}{C'_{out} - B'_{out}} \qquad \frac{u_{T_{exp}}}{T_{exp}} < 0.25\%$$

$$\chi^2(RP) = (T_{exp} - T_M)^T \, V_{T_{exp}}^{-1} \, (T_{exp} - T_M)$$

⇒ **absolute measurement**
⇒ **no calibration measurement required**

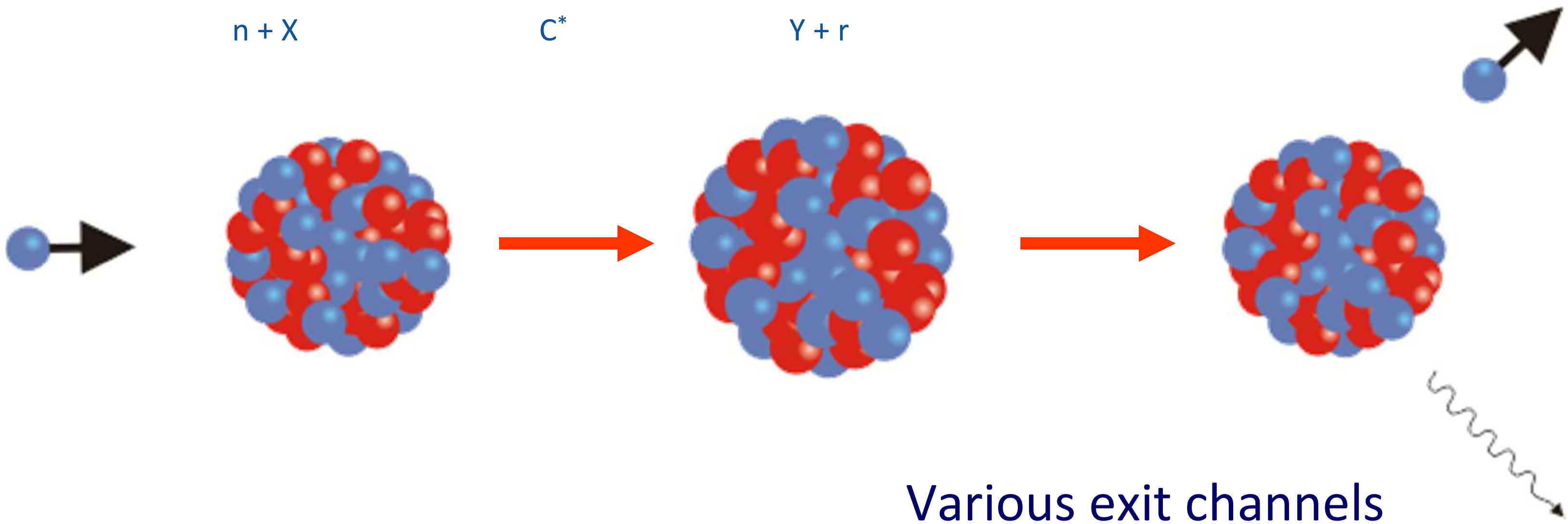


$$T_M(t) = \int R(t, E)\, e^{-n\,\sigma_{tot}(E)}\, dE$$

R(t,E)	:	response of TOF-spectrometer
σ_{tot}	:	total cross section
n	:	areal number density
		total number of atoms per unit area

Schillebeeckx et al. ,Nucl. Data Sheets, 113 3054 (2012)

Neutron Induced reactions



Various exit channels

$n + X \rightarrow$	C^*	$\rightarrow$	$X + n$	(n,n) elastic scattering	σ_n
		$\rightarrow$	$Y + \gamma$	(n,γ) capture	σ_γ
		$\rightarrow$	$Y_1 + Y_2$	(n, f) fission	σ_f
		$\rightarrow$	$Y + p$	(n,p)	σ_p
		$\rightarrow$	$Y + \alpha$	(n,α)	σ_α
		$\rightarrow$	...		

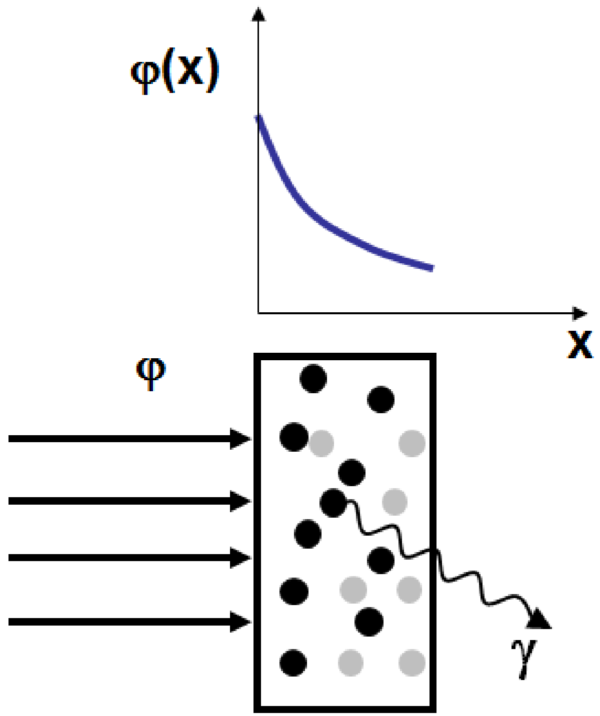
All channels together: σ_{tot}

European Commission

Reaction cross section measurement

Reaction yield

$$Y_r = (1 - e^{-n\sigma_{tot}})\frac{\sigma_r}{\sigma_{tot}} + \ldots$$



Y_r: reaction yield

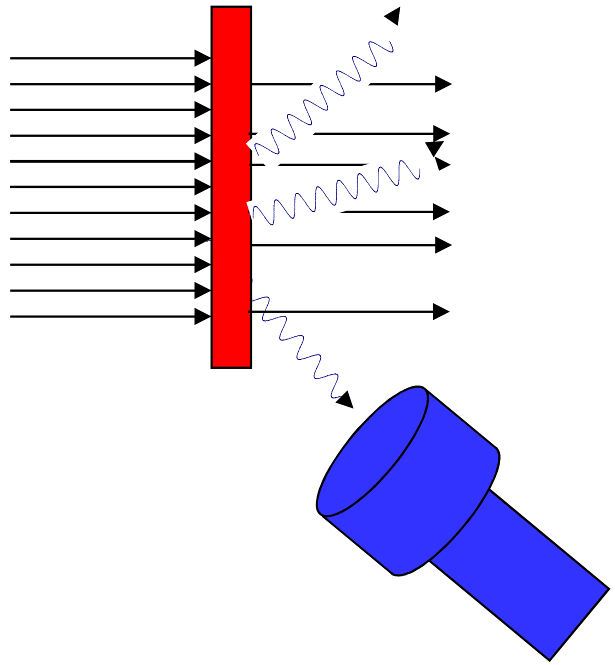
- n : areal density
- σ_r : cross section for (n,r) reaction
- σ_{tot} : total cross section

Fraction of the neutron beam creating a (n,r) reaction in the sample

Reaction cross section measurement

Experimental response ⇔ yield

$$C_r = \varepsilon_r \, \Omega_r \, P_r \, Y_r \, A \, \varphi$$

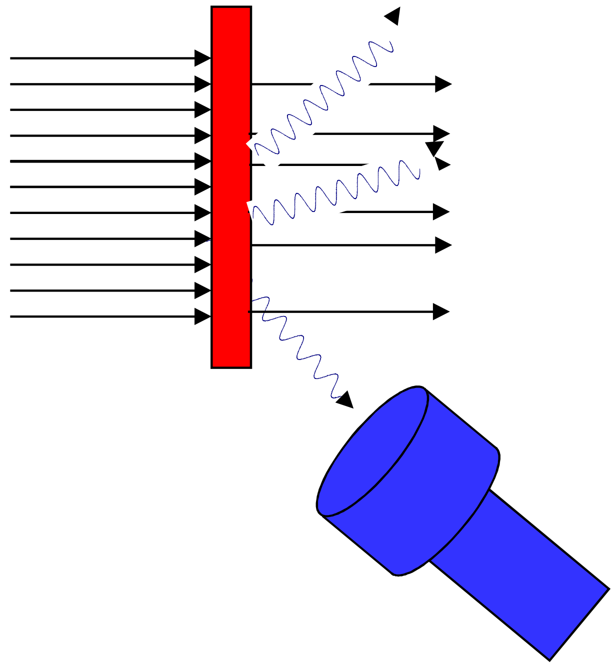


- C_r experimental response
- φ neutron flux
- A effective area (beam/sample intersection)
- Y_r reaction yield
- P_r escape probability
- Ω_r solid angle
- ε_r detection efficiency

Reaction cross section measurement

Experimental response ⇔ yield

$$Y_{r,exp} = \frac{1}{AP_r\Omega_r\varepsilon_r}\frac{C_r}{\varphi}$$



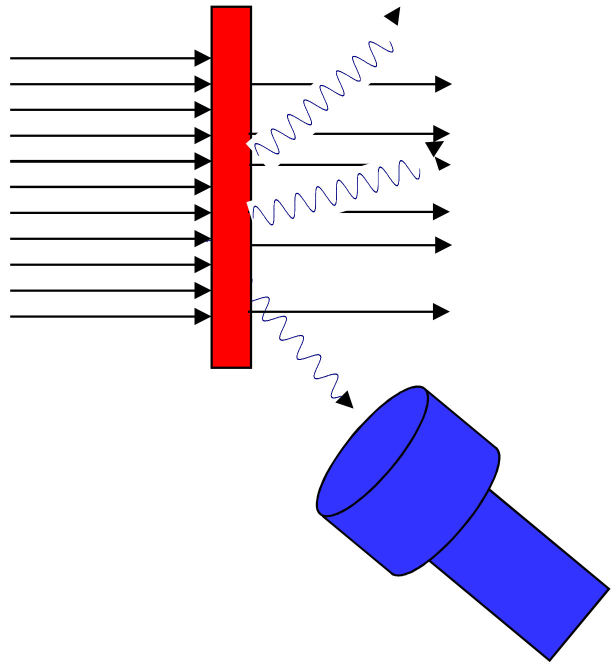
- φ neutron flux

- A effective area
- P_r escape probability
- Ω_r solid angle
- ε_r detection efficiency

Reaction cross section measurement

Experimental response ⇔ yield

$$Y_{r,exp} = \frac{1}{N_r} \frac{C_r}{\varphi}$$



- φ neutron flux

- A effective area
- P_r escape probability
- Ω_r solid angle
- ε_r detection efficiency

$$\mathbf{N_r = A\, P_r\, \Omega_r\, \varepsilon_r}$$

Cross section measurements

Transmission

$$T = e^{-n\,\sigma_{tot}}$$

$$T_{exp} = \frac{C_{in}}{C_{out}}$$

- Absolute measurement
- No additional measurements

+ direct relation: T $\Leftrightarrow$ σ_{tot}
  good geometry
  homogeneous sample

Reaction

$$Y_r \cong (1 - e^{-n\sigma_{tot}})\,\frac{\sigma_r}{\sigma_{tot}}$$

$$Y_{r,exp} = \frac{1}{N_r}\frac{C_r}{\varphi}$$

- Normalization required
- Additional flux measurement

+ complex relation : $Y_r \Leftrightarrow \sigma_r$
  $Y_r = f\,(\sigma_r, \sigma_{tot}\ \&\ \sigma_n)$
  only for $n\sigma_{tot} \ll 1$: $Y_r \cong n\,\sigma_r$

Cross section measurements

Transmission

$$T = e^{-n\,\sigma_{tot}}$$

$$T_{exp} = \frac{C_{in}}{C_{out}}$$

- Absolute measurement
- No additional measurements

σ_{tot} : most accurate cross section

$^{197}Au(n,\gamma)$ from transmission

$\sigma(n_{th}, \gamma) = (98.7 \pm 0.1)$ b

Reaction

$$Y_r \cong (1 - e^{-n\sigma_{tot}})\,\frac{\sigma_r}{\sigma_{tot}}$$

$$Y_{r,exp} = \frac{1}{N_r}\frac{C_r}{\varphi}$$

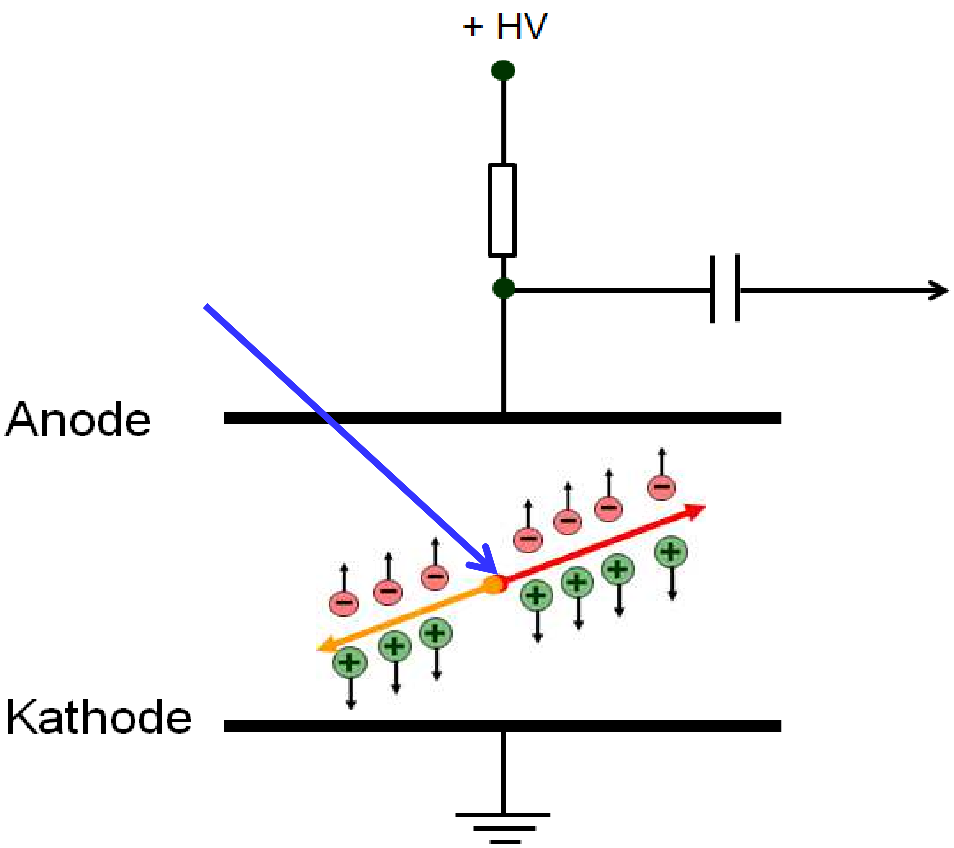
- Normalization required
- Additional flux measurement

\+ complex relation : $Y_r \Leftrightarrow \sigma_r$

$Y_r = f(\sigma_r, \sigma_{tot} \,\&\, \sigma_n)$

only for $n\sigma_{tot} << 1$: $Y_r \cong n\,\sigma_r$

Flux measurement : neutron detection

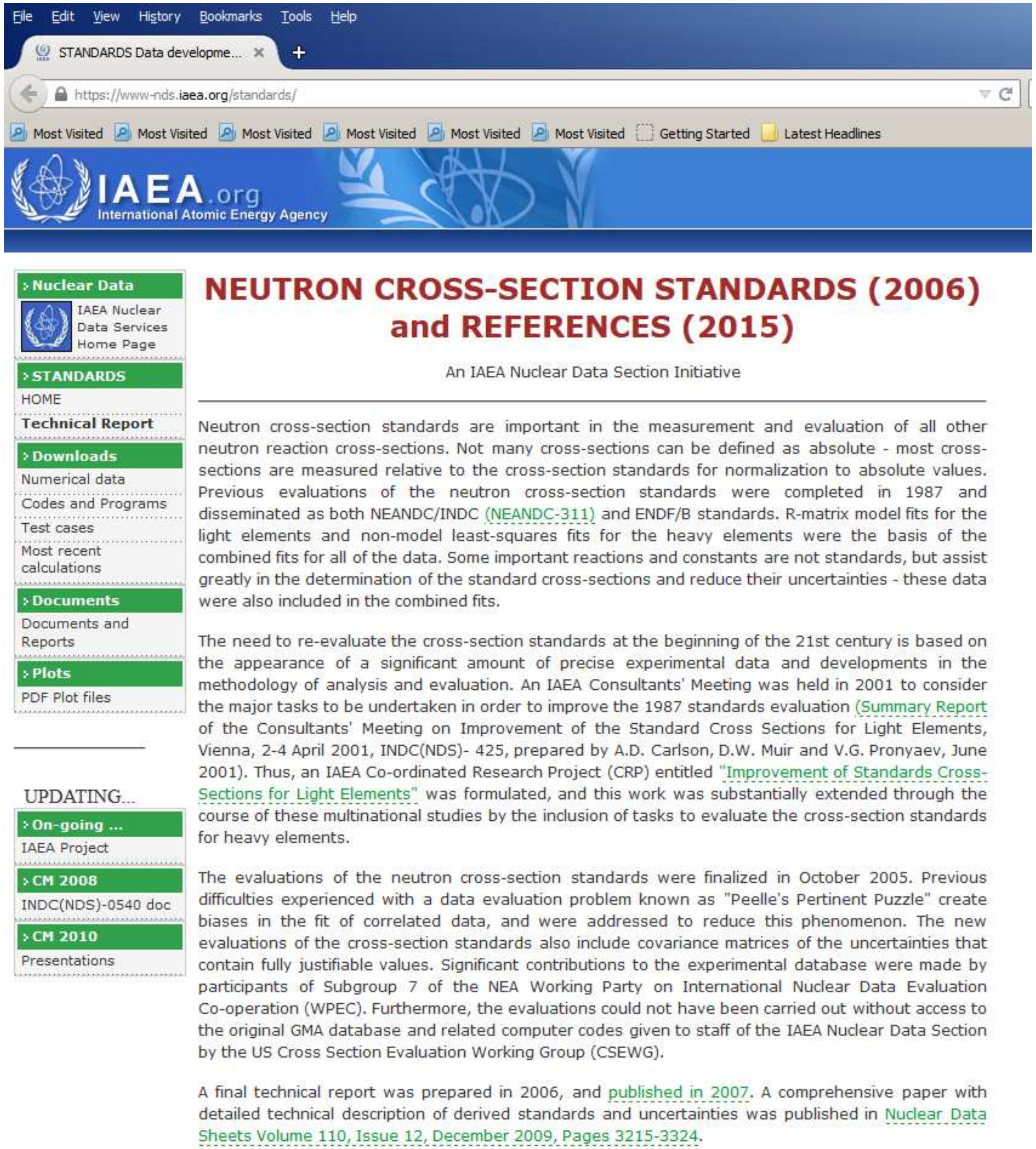


$$n + {}^{3}_{2}He \rightarrow p + {}^{3}_{1}H$$

Neutron: electrical neutral

- No direct ionisation or excitation
- **Transfer reaction is required to transform neutron energy into charged particle energy**
- **Create ionisation or excitation**

Neutron flux measurements: standard reactions



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NEUTRON CROSS-SECTION STANDARDS (2006) and REFERENCES (2015)

An IAEA Nuclear Data Section Initiative

Neutron cross-section standards are important in the measurement and evaluation of all other neutron reaction cross-sections. Not many cross-sections can be defined as absolute - most cross-sections are measured relative to the cross-section standards for normalization to absolute values. Previous evaluations of the neutron cross-section standards were completed in 1987 and disseminated as both NEANDC/INDC (NEANDC-311) and ENDF/B standards. R-matrix model fits for the light elements and non-model least-squares fits for the heavy elements were the basis of the combined fits for all of the data. Some important reactions and constants are not standards, but assist greatly in the determination of the standard cross-sections and reduce their uncertainties - these data were also included in the combined fits.

The need to re-evaluate the cross-section standards at the beginning of the 21st century is based on the appearance of a significant amount of precise experimental data and developments in the methodology of analysis and evaluation. An IAEA Consultants' Meeting was held in 2001 to consider the major tasks to be undertaken in order to improve the 1987 standards evaluation (Summary Report of the Consultants' Meeting on Improvement of the Standard Cross Sections for Light Elements, Vienna, 2-4 April 2001, INDC(NDS)- 425, prepared by A.D. Carlson, D.W. Muir and V.G. Pronyaev, June 2001). Thus, an IAEA Co-ordinated Research Project (CRP) entitled "Improvement of Standards Cross-Sections for Light Elements" was formulated, and this work was substantially extended through the course of these multinational studies by the inclusion of tasks to evaluate the cross-section standards for heavy elements.

The evaluations of the neutron cross-section standards were finalized in October 2005. Previous difficulties experienced with a data evaluation problem known as "Peelle's Pertinent Puzzle" create biases in the fit of correlated data, and were addressed to reduce this phenomenon. The new evaluations of the cross-section standards also include covariance matrices of the uncertainties that contain fully justifiable values. Significant contributions to the experimental database were made by participants of Subgroup 7 of the NEA Working Party on International Nuclear Data Evaluation Co-operation (WPEC). Furthermore, the evaluations could not have been carried out without access to the original GMA database and related computer codes given to staff of the IAEA Nuclear Data Section by the US Cross Section Evaluation Working Group (CSEWG).

A final technical report was prepared in 2006, and published in 2007. A comprehensive paper with detailed technical description of derived standards and uncertainties was published in Nuclear Data Sheets Volume 110, Issue 12, December 2009, Pages 3215-3324.

For neutron flux measurements a transfer reaction with a well-known cross section is required:

Standard and reference reactions

https://www-nds.iaea.org/standards/

Neutron flux measurements: standard reactions

Reaction	2200 m/s	Energy region E_{low}		Energy region E_{high}	
$^{1}H(n,n)$		1 keV	-	20 MeV	
$^{3}He(n,p)$	X	25.3 meV	-	50 keV	
$^{6}Li(n,t)$	X	25.3 meV	-	1 MeV	
$^{10}B(n,\alpha)$	X	25.3 meV	-	1 MeV	
$^{10}B(n,\alpha_1\gamma)$	X	25.3 meV	-	1 MeV	
$^{nat}C(n,n)$		25.3 meV	-	1.8 MeV	
$^{197}Au(n,\gamma)$	X	0.2 MeV	-	2.5 MeV	
$^{235}U(n,f)$	X	0.15 MeV	-	200 MeV	(1 GeV)
$^{238}U(n,f)$	X	2 MeV	-	200 MeV	(1 GeV)
$^{239}Pu(n,f)$	X	0.15 MeV	-	300 MeV	
$^{209}Bi(n,f)$		34 MeV	-	1 GeV	
$^{nat}Pb(n,f)$		34 MeV	-	1 GeV	

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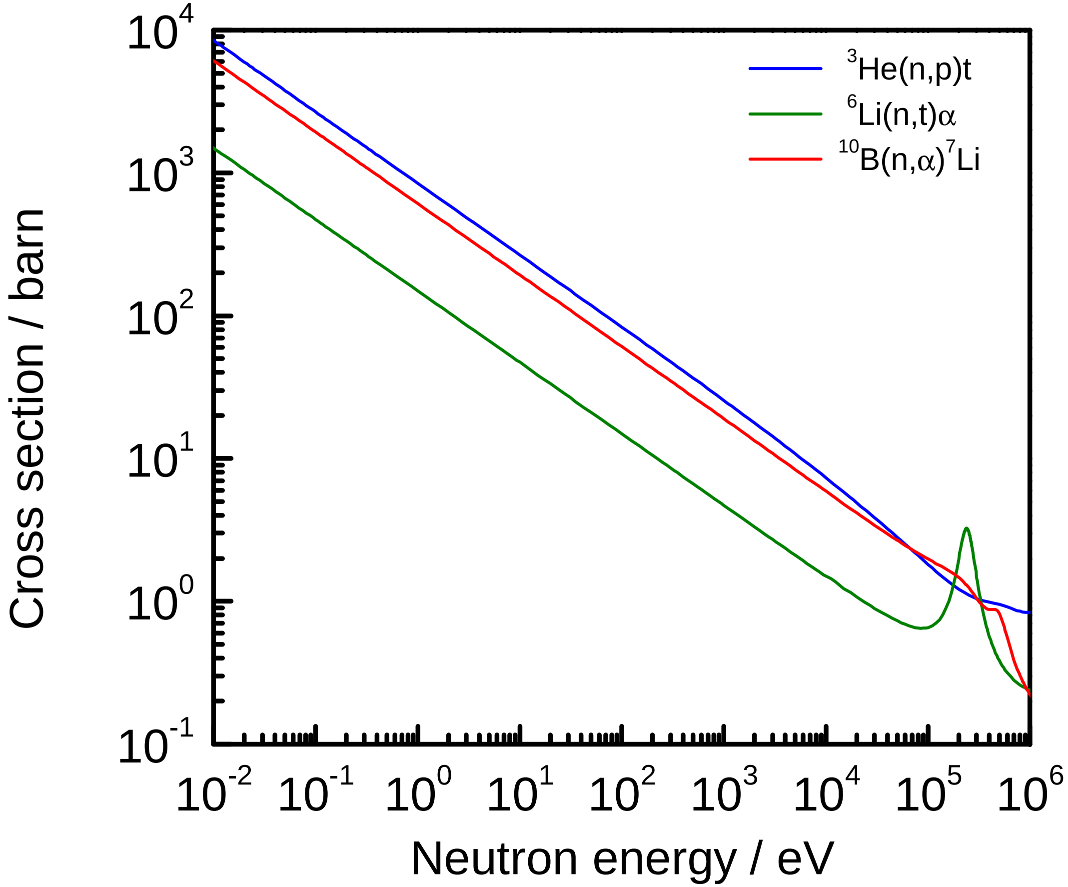

European Commission

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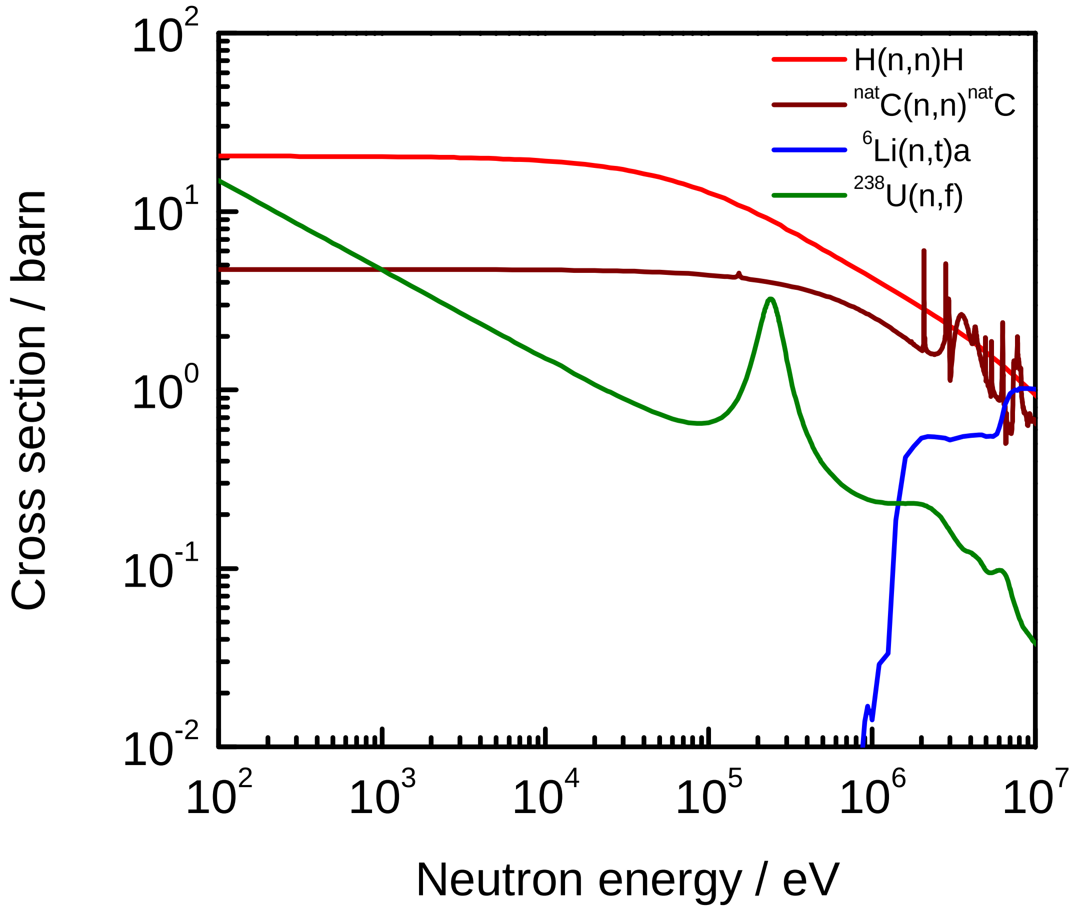

Neutron flux measurements: standard reactions

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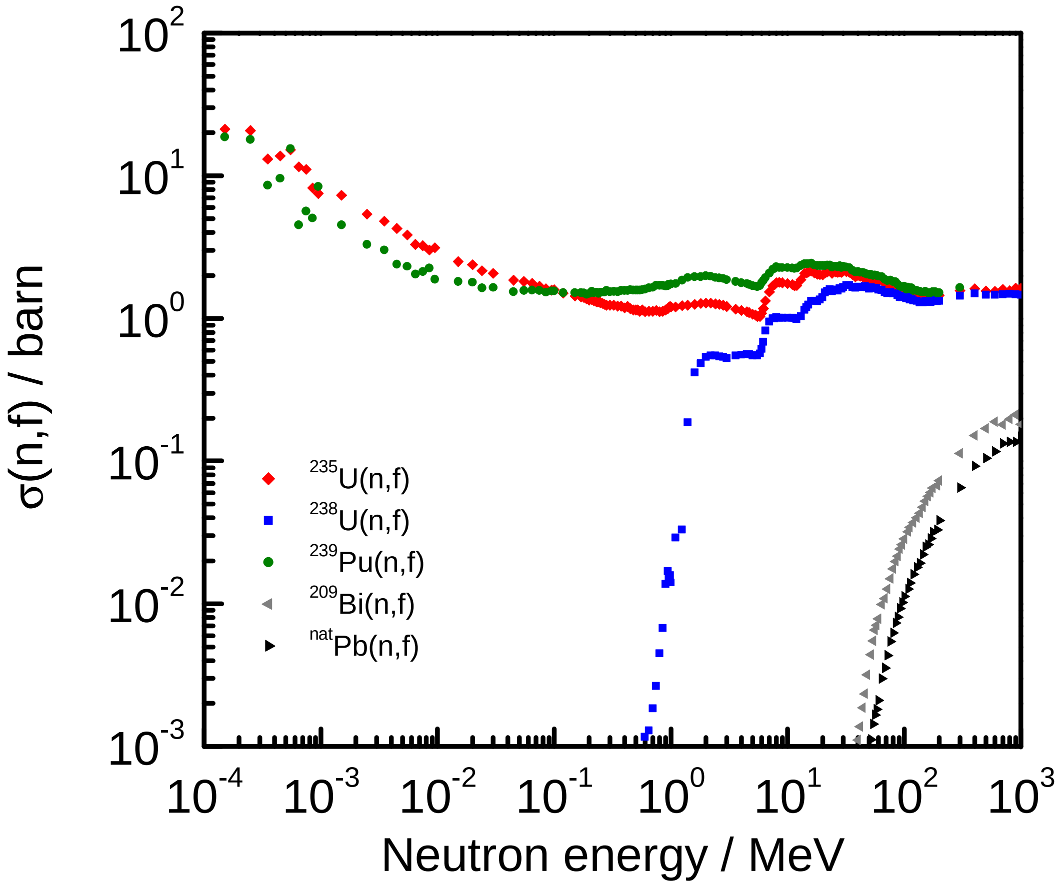
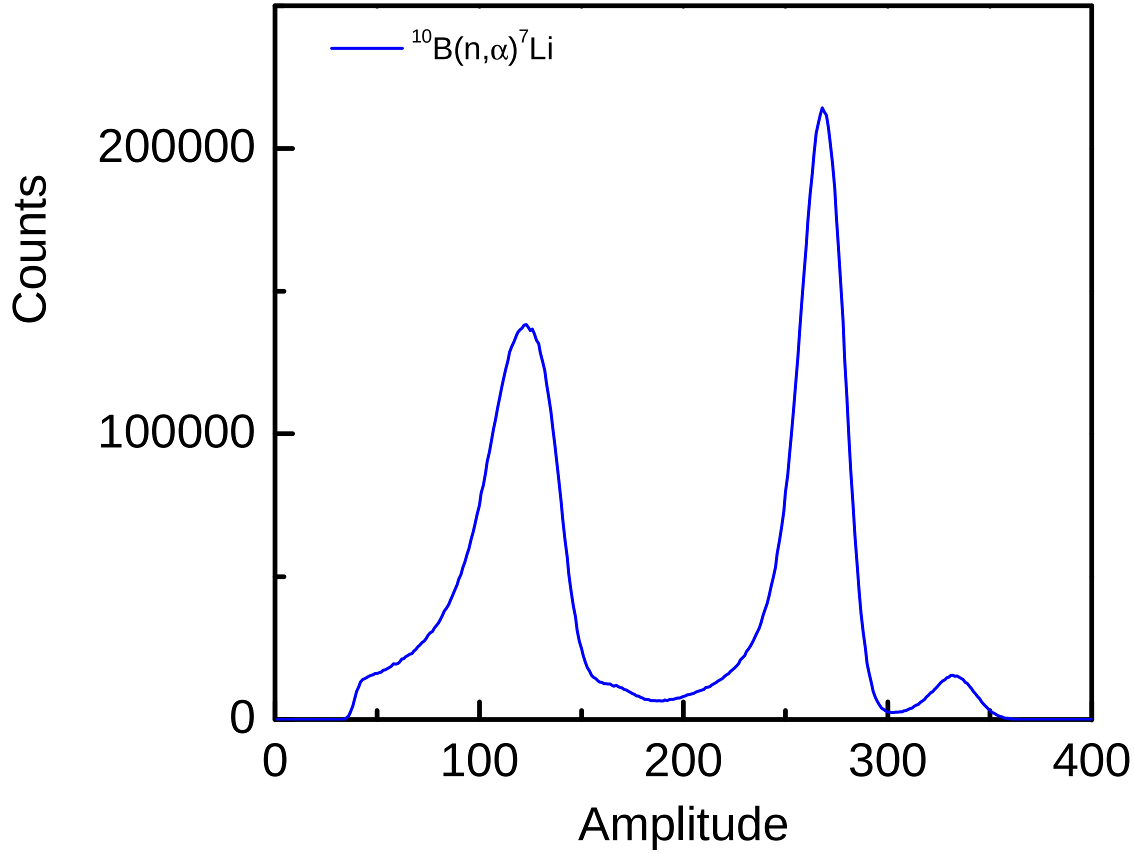


Neutron flux measurements

Frisch-gridded ionisation chamber loaded with ^{10}B

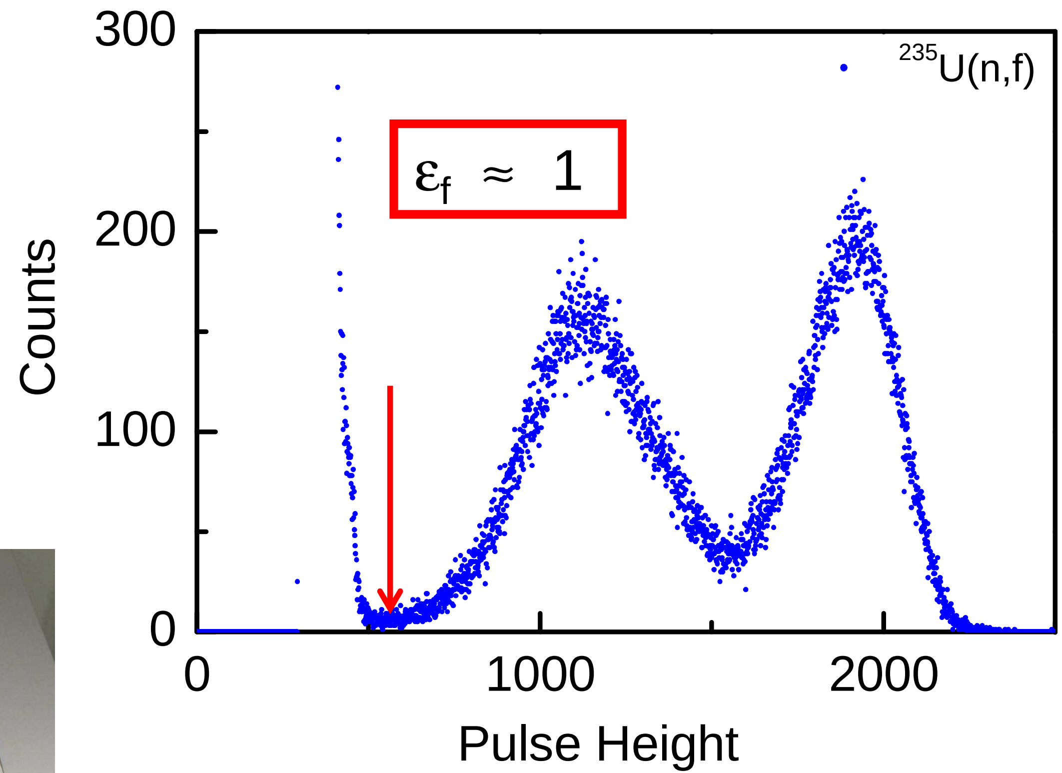
standard reaction: $^{10}B(n,\alpha)^{7}Li$



Neutron flux measurements

Frisch-gridded ionisation chamber loaded with ^{235}U

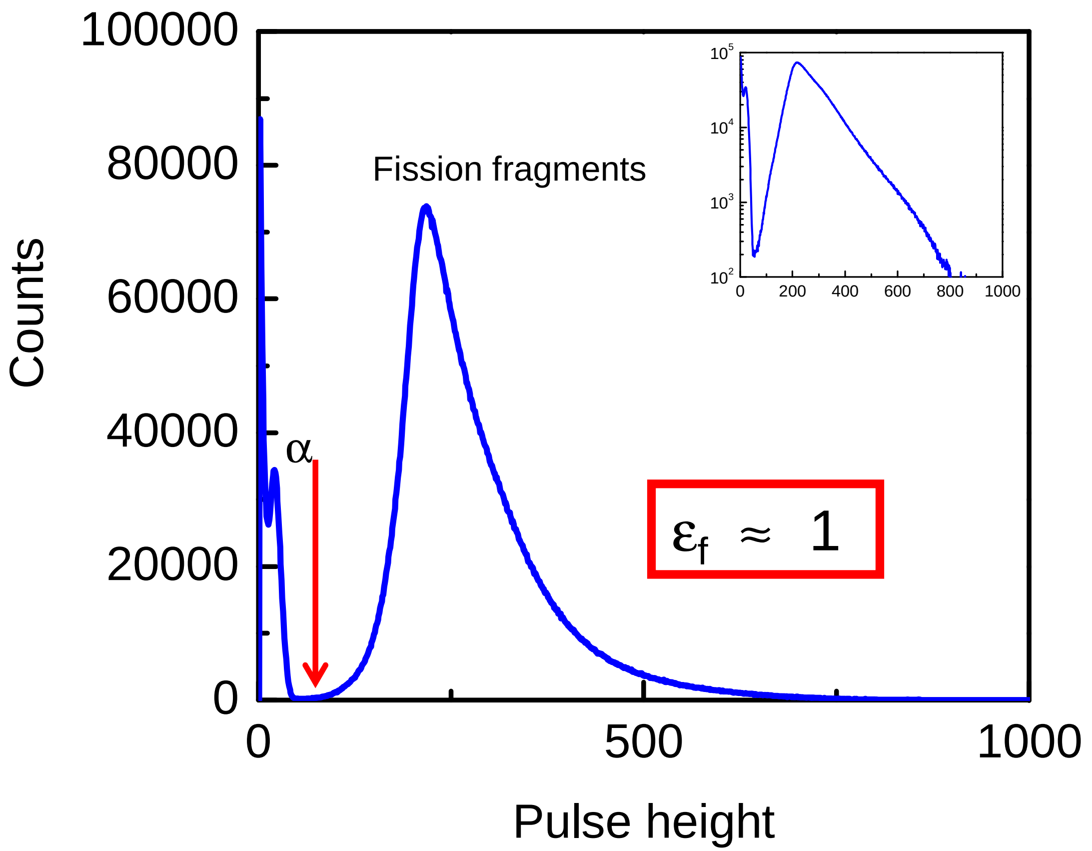
standard reaction: $^{235}U(n,f)$



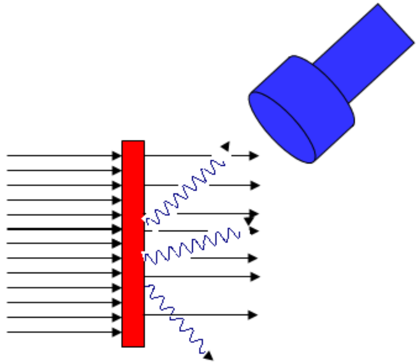
Neutron flux measurements

Parallel plate ionisation chamber loaded with ^{235}U

standard reaction: $^{235}U(n,f)$



Reaction cross section measurements



1. Reaction measurement $C_r = \varepsilon_r \, \Omega_r \, P_r \, Y_r \, A_r \, \varphi$

2. Flux measurement $C_\varphi = \varepsilon_\varphi \, \Omega_\varphi \, P_\varphi \, Y_\varphi \, A_\varphi \, \varphi$
   (mostly thin target)

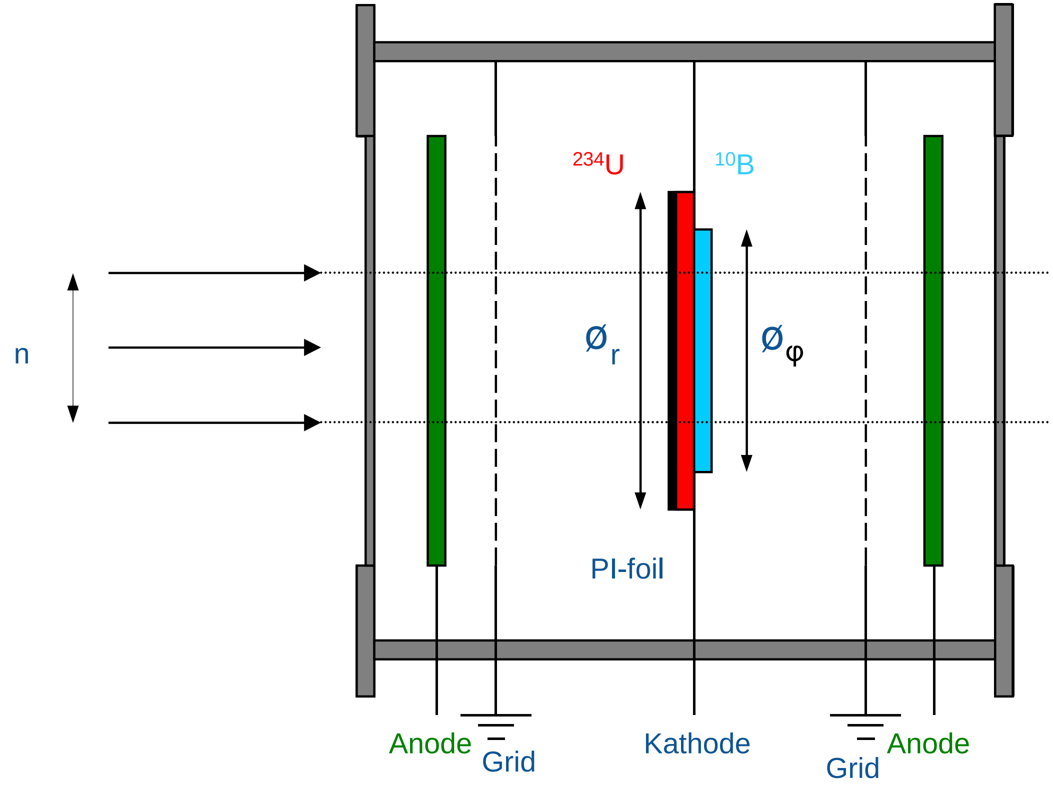
$$Y_{r,exp} = \frac{\varepsilon_\varphi}{\varepsilon_r} \frac{\Omega_\varphi}{\Omega_r} \frac{P_\varphi}{P_r} \frac{A_\varphi}{A_r} \frac{C_r}{C_\varphi} Y_\varphi = N \frac{C_r}{C_\varphi} Y_\varphi$$

$\Rightarrow$ $Y_{r,exp}$ is the ratio of results of 2 reaction cross section measurements

$\Rightarrow$ Y_φ is required $Y_\varphi \cong (1 - e^{-n\sigma_{tot}}) \dfrac{\sigma_\varphi}{\sigma_{tot}}$

neutron standard reaction

Example : fission cross section



$$A_\varphi = A_r$$
$$P_\varphi = P_r \quad (\approx 1)$$
$$\Omega_\varphi = \Omega_r$$
$$\varepsilon_\varphi = \varepsilon_r \quad (\approx 1)$$

$$Y_{r,exp} = \frac{\varepsilon_\varphi}{\varepsilon_r}\frac{\Omega_\varphi}{\Omega_r}\frac{P_\varphi}{P_r}\frac{A_\varphi}{A_r}\frac{C_r}{C_\varphi}Y_\varphi$$

Geometry (back to back)

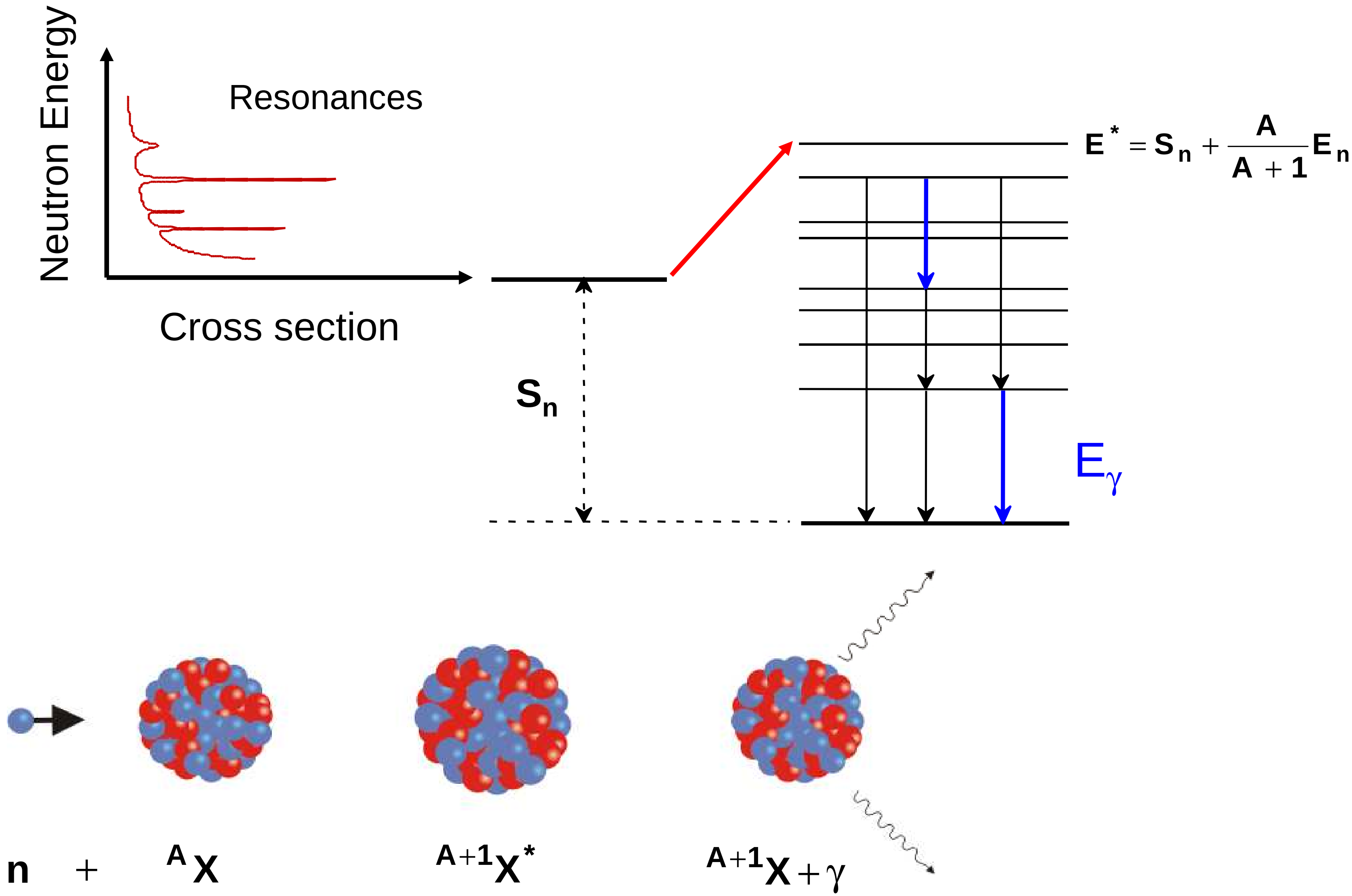
$$Y_r \cong \frac{C_r}{C_\varphi}Y_\varphi$$

Thin target approximation

$Y \cong n\sigma \qquad n\sigma_{tot} << 1$

$$\sigma_r \cong \frac{C_r}{C_\varphi}\frac{n_\varphi}{n_X}\sigma_\varphi$$

Neutron induced capture process



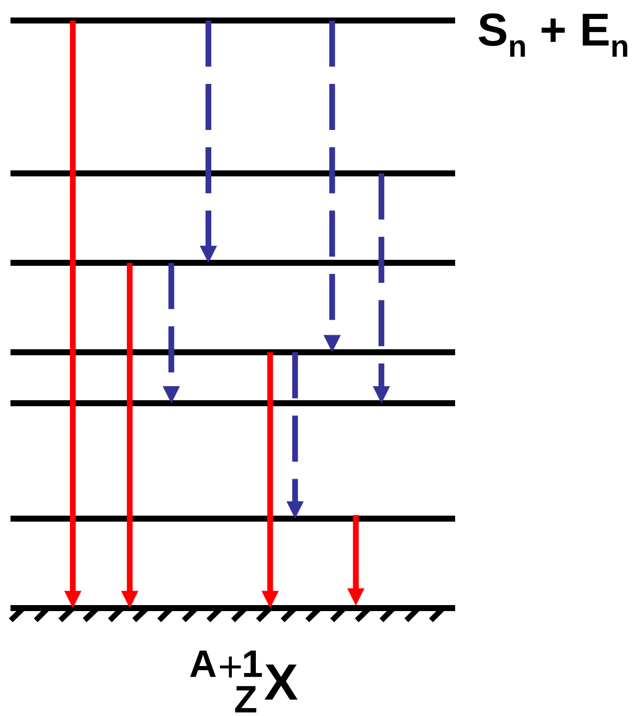
$\sigma(n,\gamma)$ measurements

Efficiency to detect capture event independent of gamma-ray cascade

- Gamma-ray spectroscopy
  good resolution for E_γ e.g. Ge-detectors

- Total absorption detectors
  4π & $\varepsilon_\gamma \approx 100\%$ e.g. BaF_2

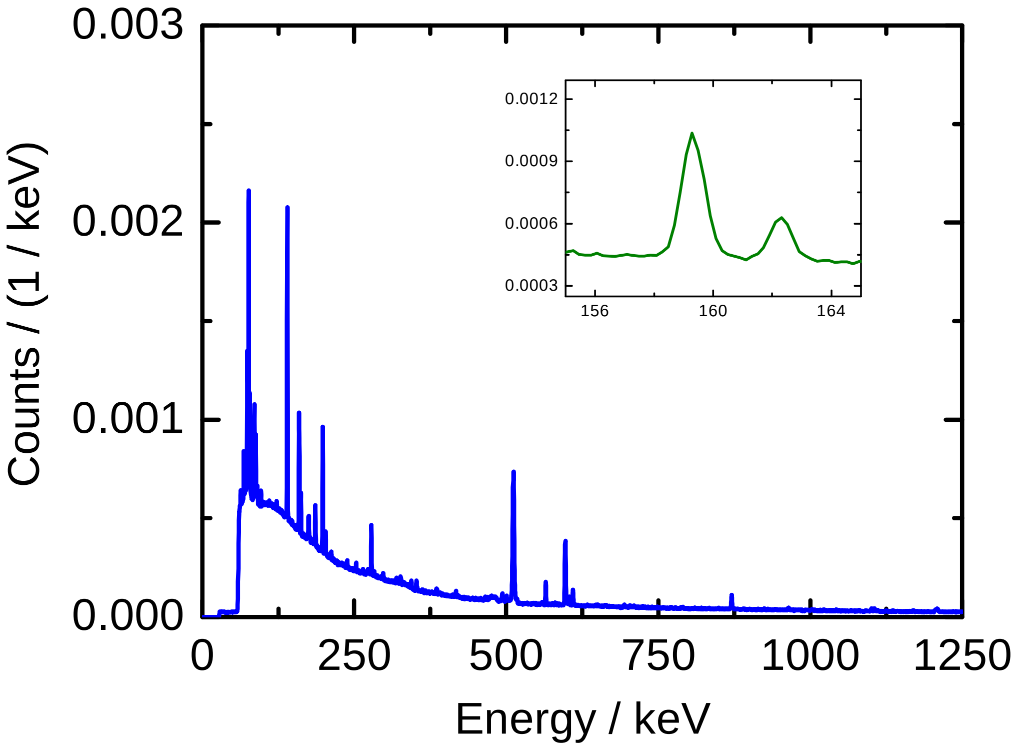
- Total energy detection principle
  $\varepsilon_\gamma \propto E_\gamma$ & $\varepsilon_\gamma << 1$ e.g. C_6D_6 scintillators



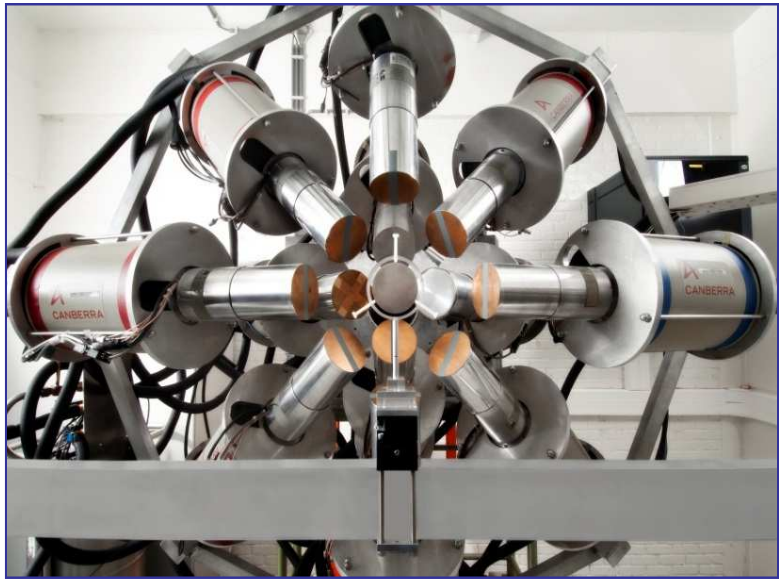
Schillebeeckx et al., Nucl. Data Sheets 113 (2012) 3054

σ(n,γ) : γ -ray spectroscopy

- Gamma-ray spectroscopy
  high resolution & simple known decay scheme

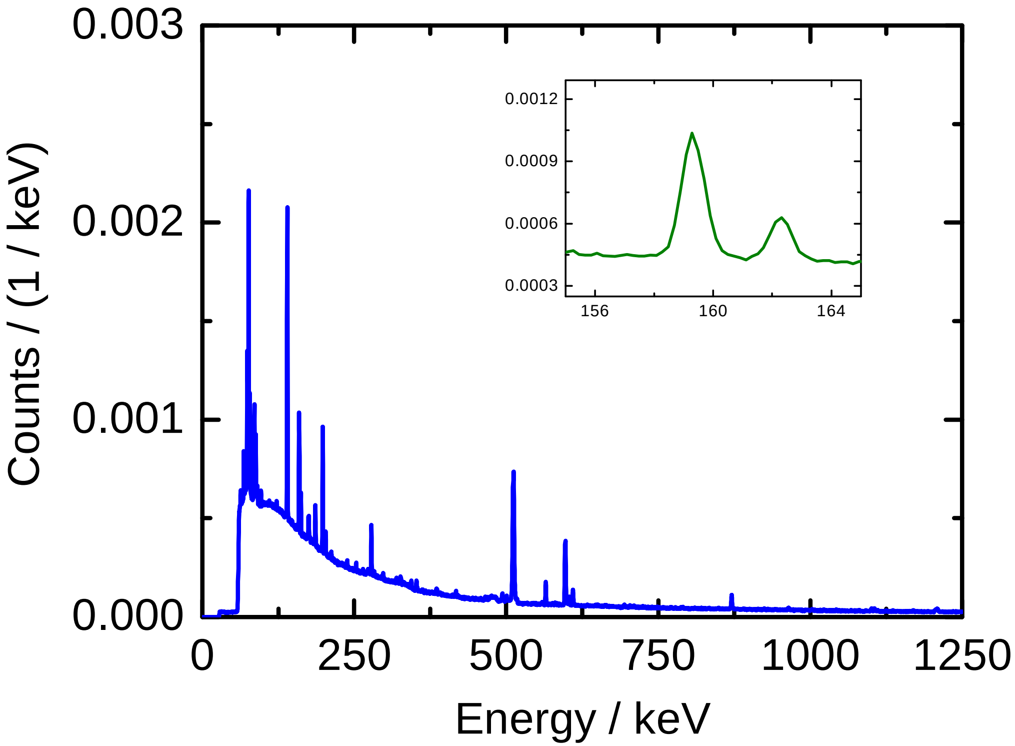


Ge-detectors



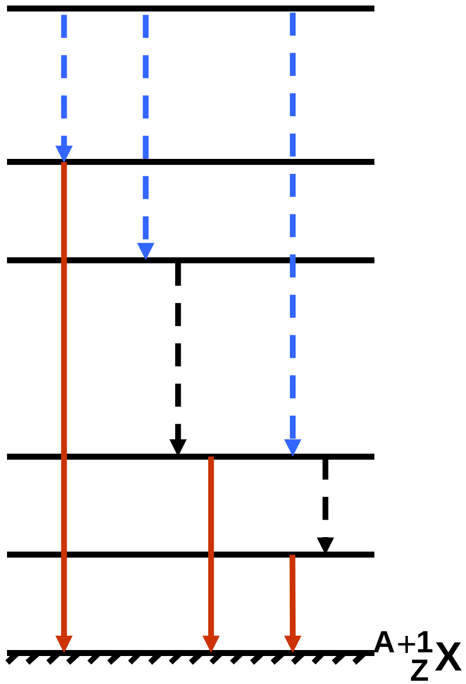
σ(n,γ) : γ -ray spectroscopy

- Gamma-ray spectroscopy
  high resolution & **simple well-known decay scheme**



$$\sigma_\gamma = \sum_{J_0} \sigma_{\gamma,J}$$

$$\sigma_\gamma = \sum_{J_{gs}} \sigma_{\gamma,J}$$



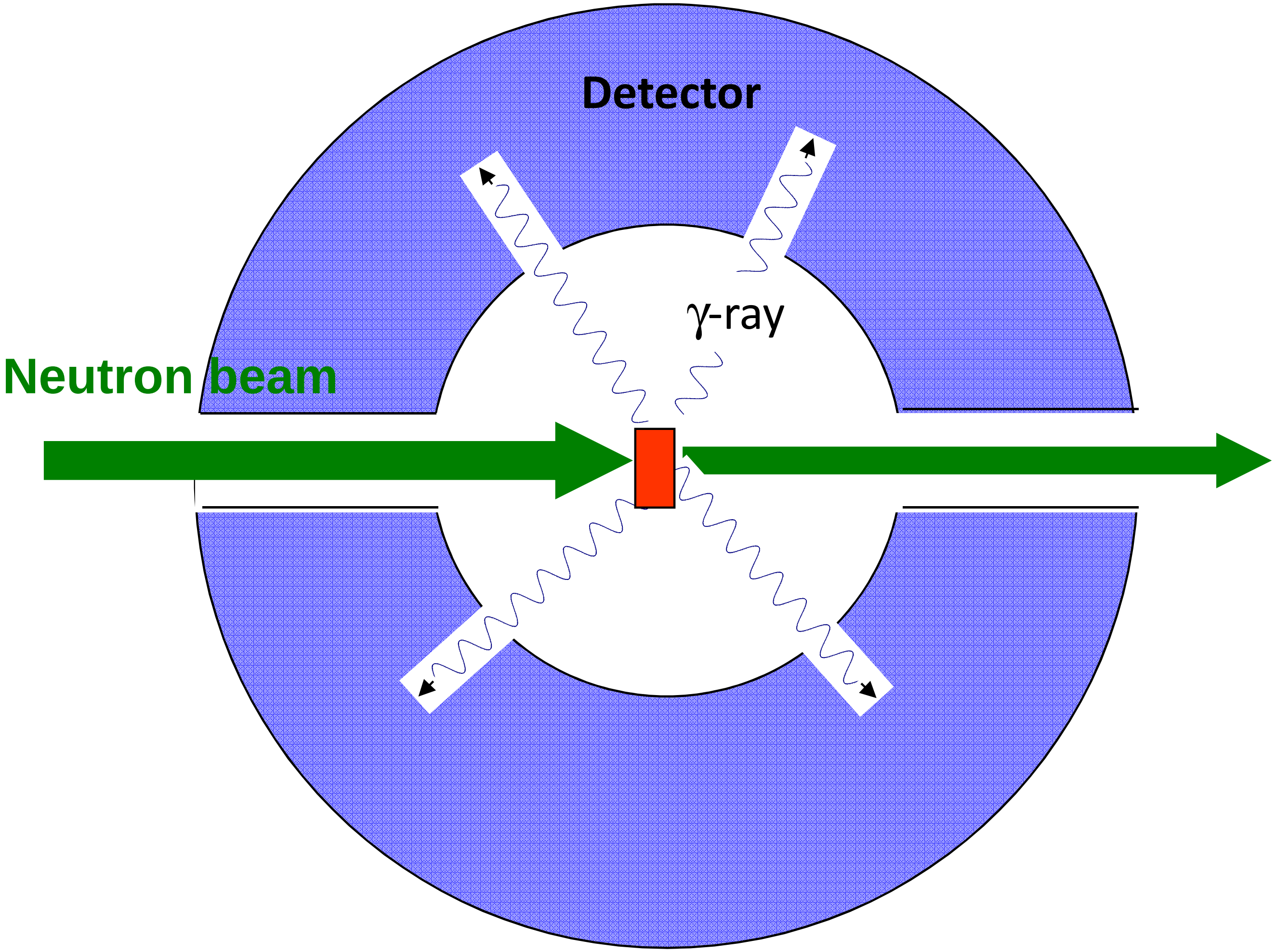
Borella et al., Nucl. Phys. A 850 (2011) 1

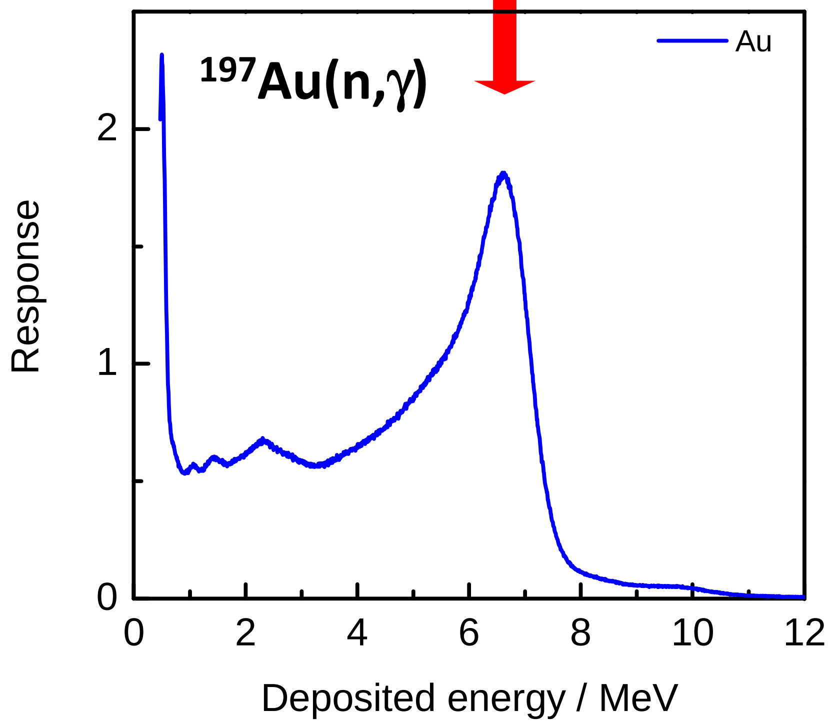
σ(n,γ) : total absorption detector

- Total absorption detector

  $\Omega \approx 4\pi$ & $\varepsilon_\gamma \approx 100\%$

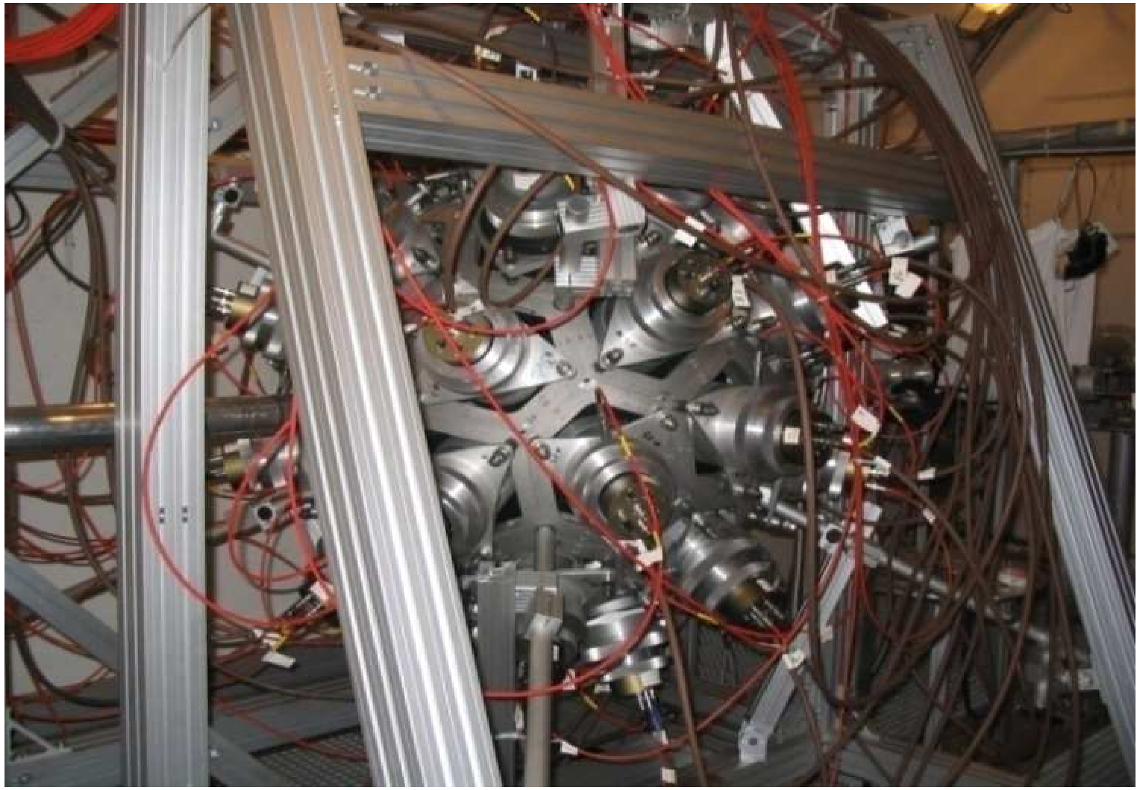
$$E_d = \sum_{i=1}^{n} E_{\gamma i}$$

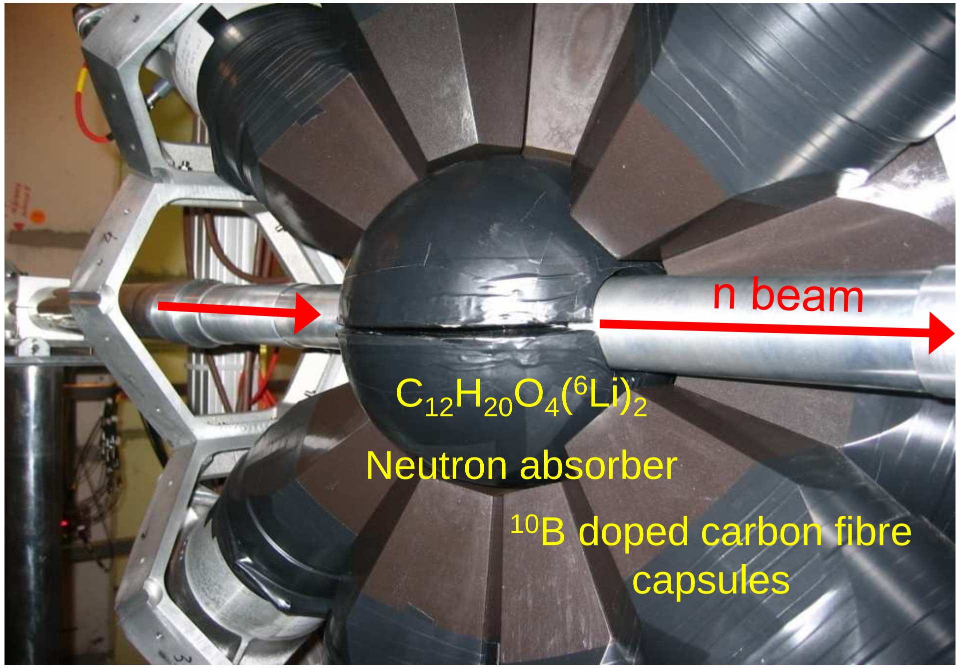




Total absorption detector at nTOF (CERN)

- Total absorption detector
  $\Omega \approx 4\pi$ & $\varepsilon_\gamma \approx 100\%$





Guerrero et al, NIMA 608 (2009) 424

σ(n,γ) : total energy detection principle

- Total energy detection principle

Probability to detect a capture event = efficiency to detect at least one γ - ray

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

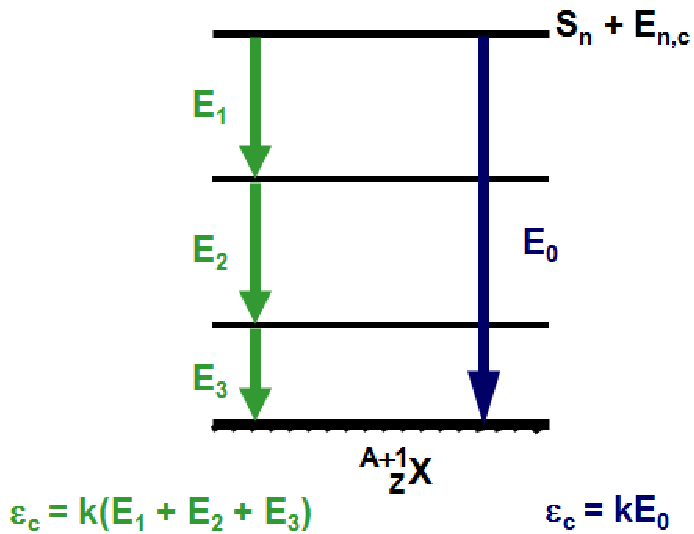
1) Detection efficiency : $\varepsilon_\gamma << 1$

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

2) Detection efficiency: $\varepsilon_\gamma = k\, E_\gamma$

$$\Rightarrow \quad \varepsilon_c \approx \sum_i \varepsilon_{\gamma,i} = k \sum_i E_{\gamma,i} \approx k\,(S_n + E \frac{m_X}{m_X + m_n})$$

independent of γ-ray cascade



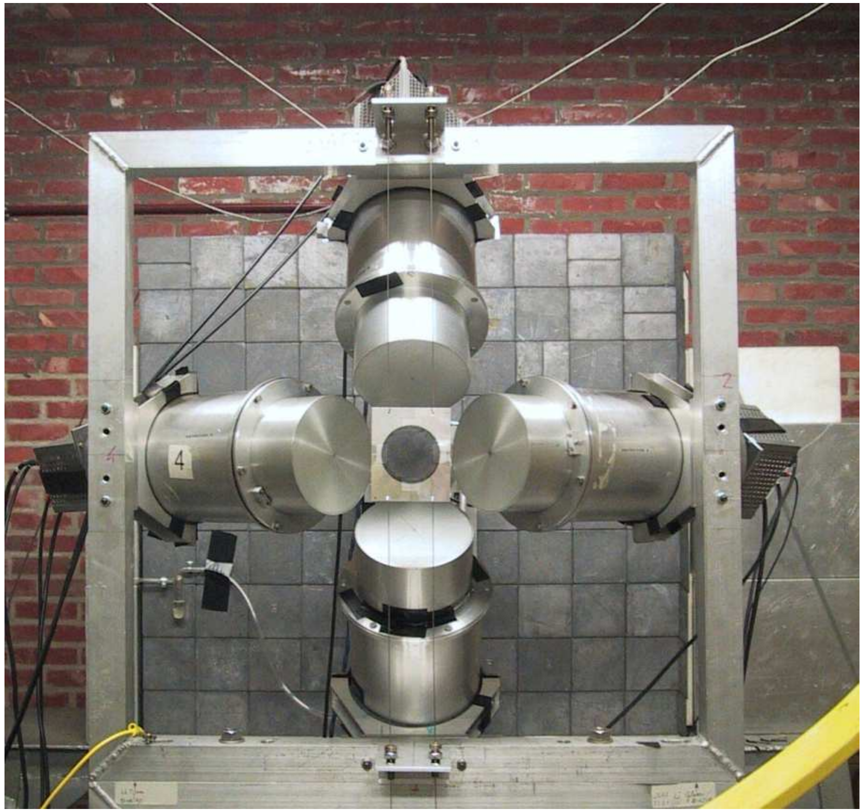
European Commission

$\sigma(n,\gamma)$ measurements at GELINA

Total energy detection principle

- C_6D_6 liquid scintillators
  $\varepsilon_\gamma << 1$ & $\varepsilon_\gamma \propto E_\gamma$ (PHWT)
- Flux measurements (IC): $^{10}B(n,\alpha)$

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

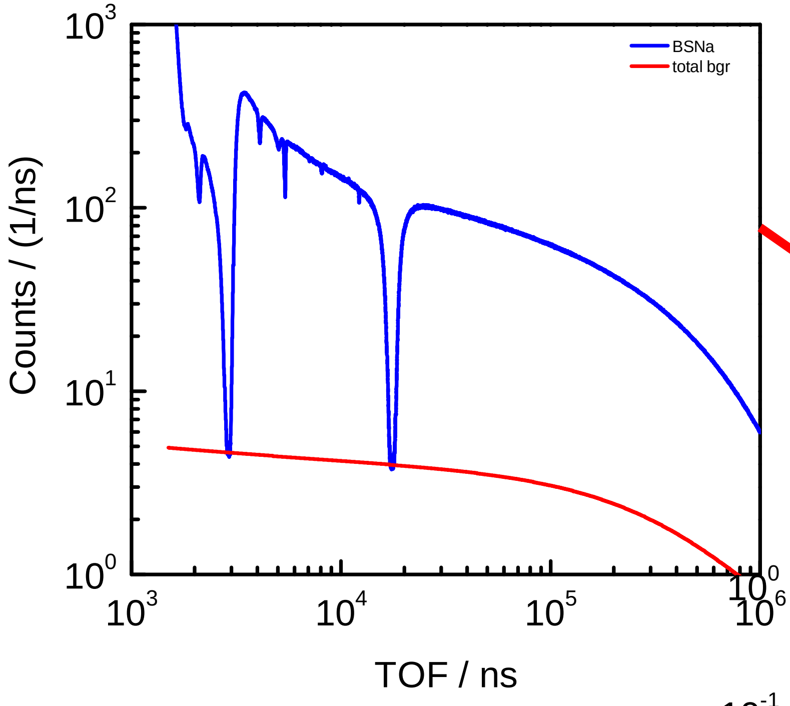


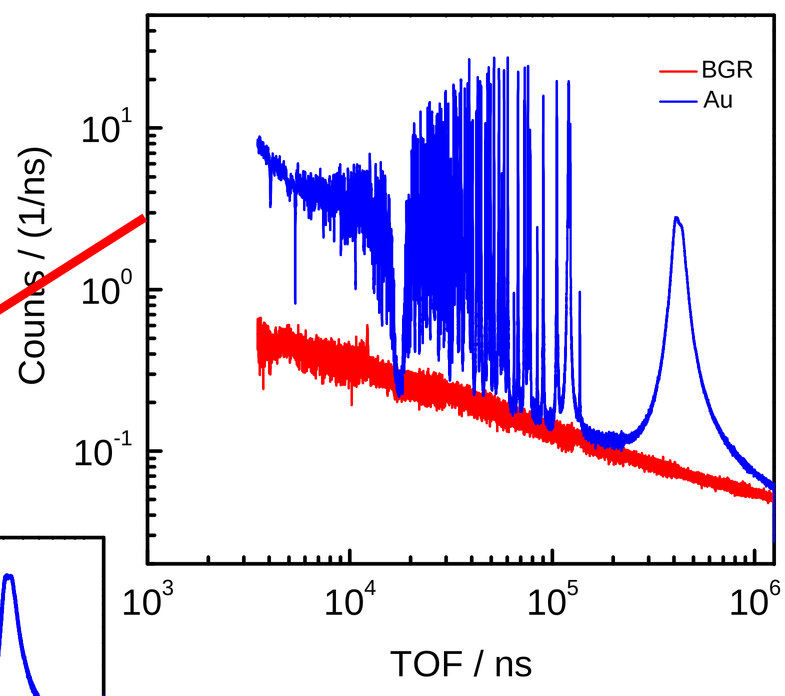
Pulse Height Weighting Technique

$$E_\gamma = \int WF(E_d)\ R(E_d, E_\gamma) dE_d$$

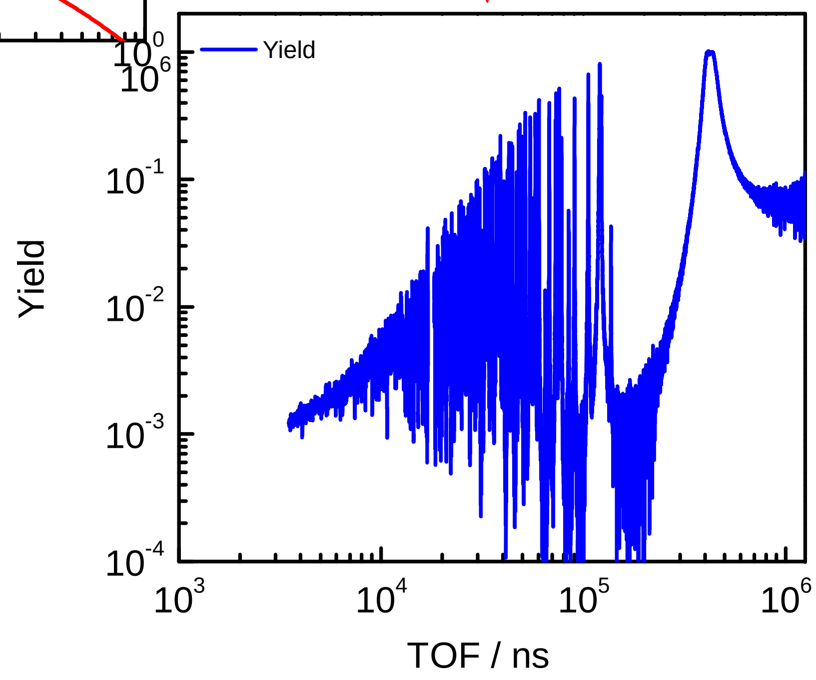
Borella et al., NIMA 577 (2007) 626

Y_{exp} for $^{197}Au(n,\gamma)$

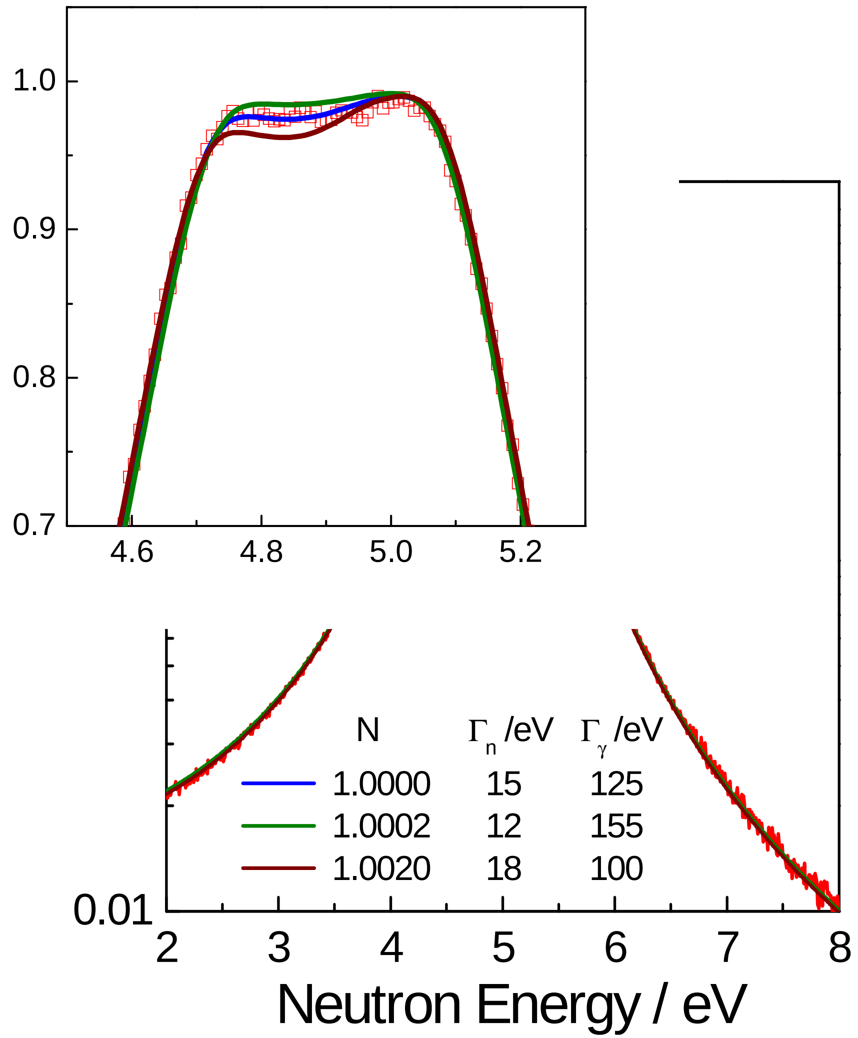




$$Y_{exp} = N \frac{C'_{\gamma} - B'_{\gamma}}{C'_{\varphi} - B'_{\varphi}} Y_{\varphi}$$



Normalization: saturated resonance



$n\sigma_{tot} >> 1$ and $\sigma_\gamma \approx \sigma_{tot}$

$$Y_\gamma \cong \frac{\sigma_\gamma}{\sigma_{tot}}(1 - e^{-n\sigma_{tot}}) + \ldots$$

$$Y_\gamma \cong 1$$

$$\Rightarrow N \cong \frac{C'_\varphi - B'_\varphi}{C'_\gamma - B'_\gamma} \frac{1}{Y_\varphi}$$

N is independent of :

- sample thickness
- nuclear data

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

$$\frac{u_{Y_{exp}}}{Y_{exp}} \leq 2\,\%$$

Experimental observables

$$T_{exp} = \frac{C_{in} - B_{in}}{C_{out} - B_{out}}$$ $$\frac{u_{T_{exp}}}{T_{exp}} \leq 0.25\%$$

$$Y_{exp,\gamma} = K_\gamma \frac{C_{in} - B_{in}}{C_\varphi - B_\varphi} Y_\varphi$$ $$\frac{u_{Y_{exp,\gamma}}}{Y_{exp,\gamma}} \leq 2.0\%$$ (without fission)

$$Y_{exp,f} = K_f \frac{C_{in} - B_{in}}{C_\varphi - B_\varphi} Y_\varphi$$ $$\frac{u_{Y_{exp,f}}}{Y_{exp,f}} \leq 2.0\%$$

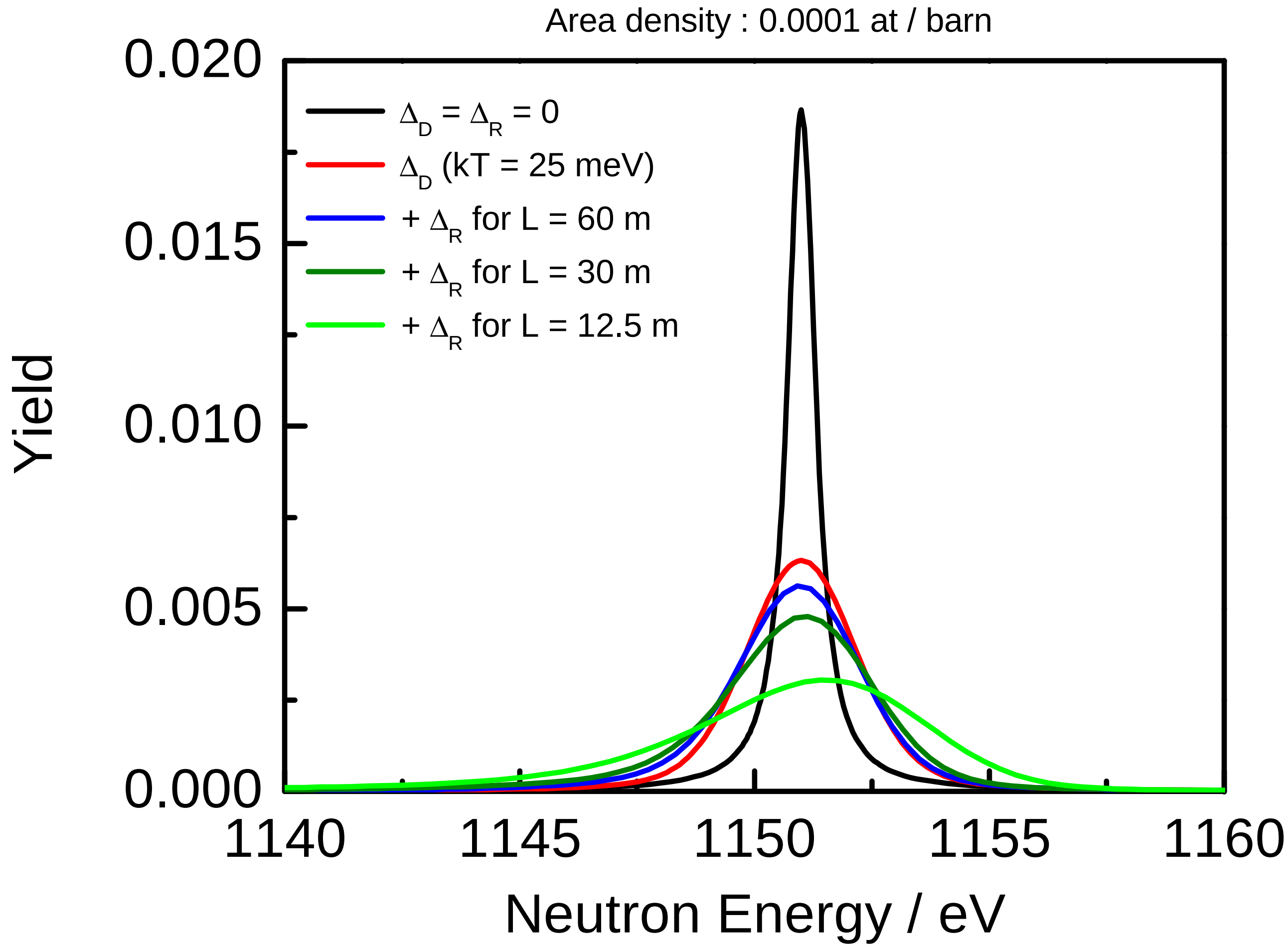
Methodologies to determine (Z_{exp},$V_{Z_{exp}}$) are well established

Nuclear Data Sheets 113 (2012) 3054 - 3100

+ Doppler + Response

$^{56}Fe(n,\gamma)$

n_{Fe} = 1 10^{-4} at/b (0.012 mm thick)



$$Y_\gamma \cong \frac{\bar{\sigma}_\gamma}{\bar{\sigma}_{tot}}(1 - e^{-n\bar{\sigma}_{tot}}) + \ldots$$

$$\bar{\sigma}(E) = \int dE' S(E') \sigma(E - E')$$

$$Y_{exp} = \int R(t,E) Y_\gamma(E) dE$$

L = 60 m

L = 30 m

L = 12.5 m

$$\frac{\Delta E}{E} = 2\sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}$$

European Commission

Resonance shape analysis

$$\chi^2(\eta) = (Z_{exp} - Z_M(t_m,\eta,\kappa))^T \, V_{Z_{exp}}^{-1} \, (Z_{exp} - Z_M(t_m,\eta,\kappa))$$

Z_{exp} : experimental observable

$Z_M(t,\eta,\kappa)$: model for theoretical estimate of Z_{exp}

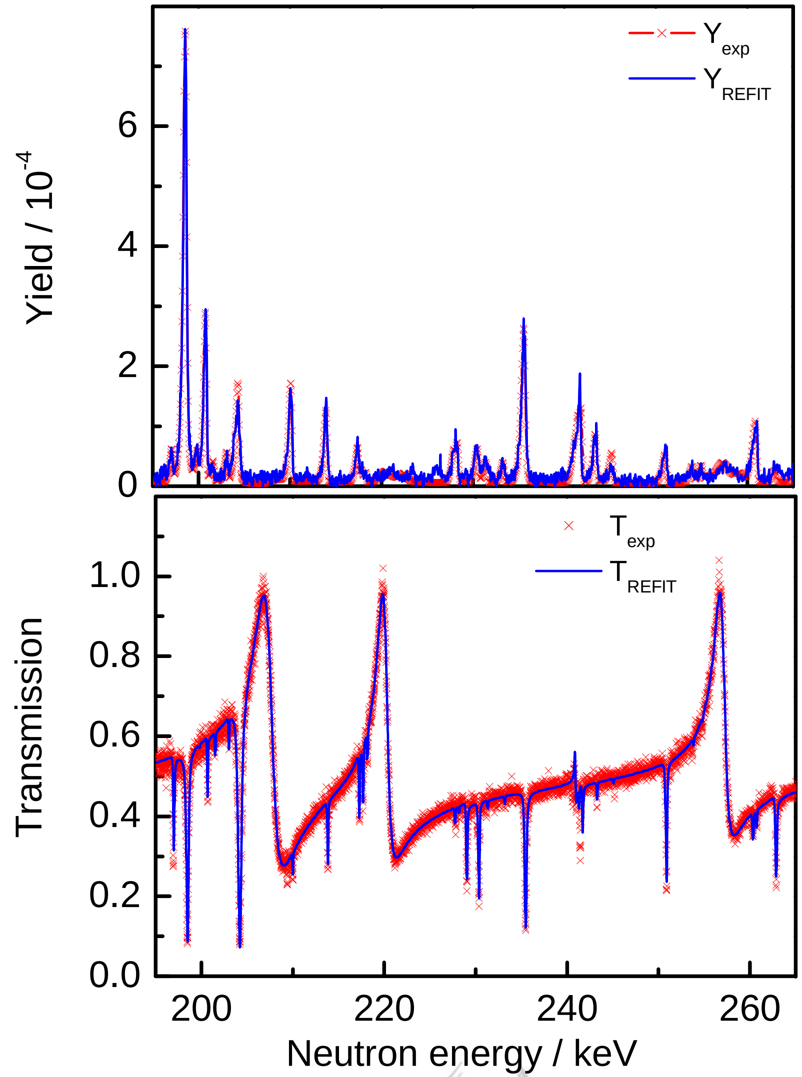
Model implemented in REFIT (M. Moxon):

- R-matrix theory : parameterisation of σ by RP (η)
- Experimental conditions : parameter vector κ

$$Z_M(t_m) = \int R(t_m,E)\; Z_M(E)dE$$

$$Z_M(E) = \begin{cases} T(E) = e^{-n\bar{\sigma}_{tot}} \\ Y_r(E) == (1 - e^{-n\bar{\sigma}_{tot}})\dfrac{\bar{\sigma}_r}{\bar{\sigma}_{tot}} + \dots \end{cases}$$

$^{206}Pb + n$



Resonance shape analysis

$$\chi^2(\eta) = (Z_{exp} - Z_M(t_m, \eta, \kappa))^T \, V_{Z_{exp}}^{-1} \, (Z_{exp} - Z_M(t_m, \eta, \kappa))$$

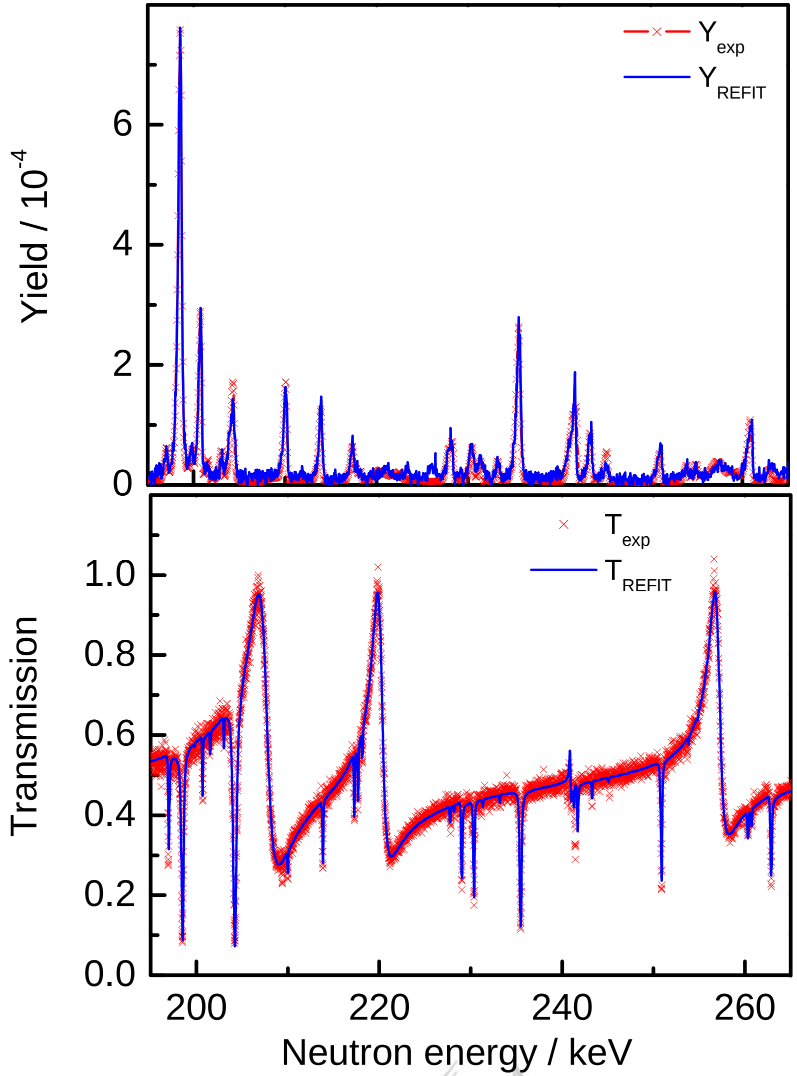
Z_{exp} : experimental observable

$Z_M(t, \eta, \kappa)$: model for theoretical estimate of Z_{exp}

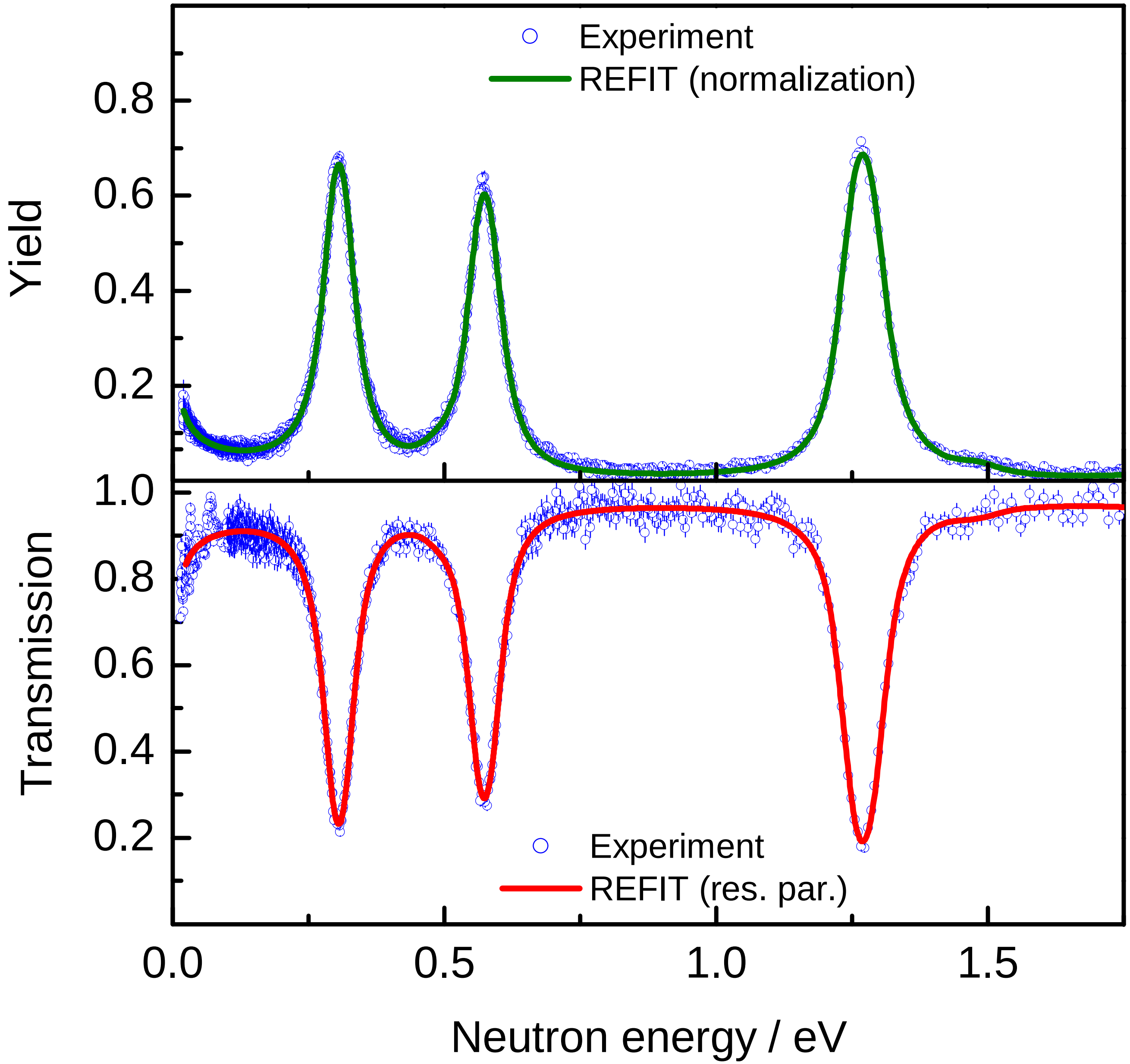
Experimental conditions:

- Doppler broadening
- Response TOF-spectrometer
- Multiple interaction
- Sample characteristics
- Detector characteristics

$^{206}Pb + n$



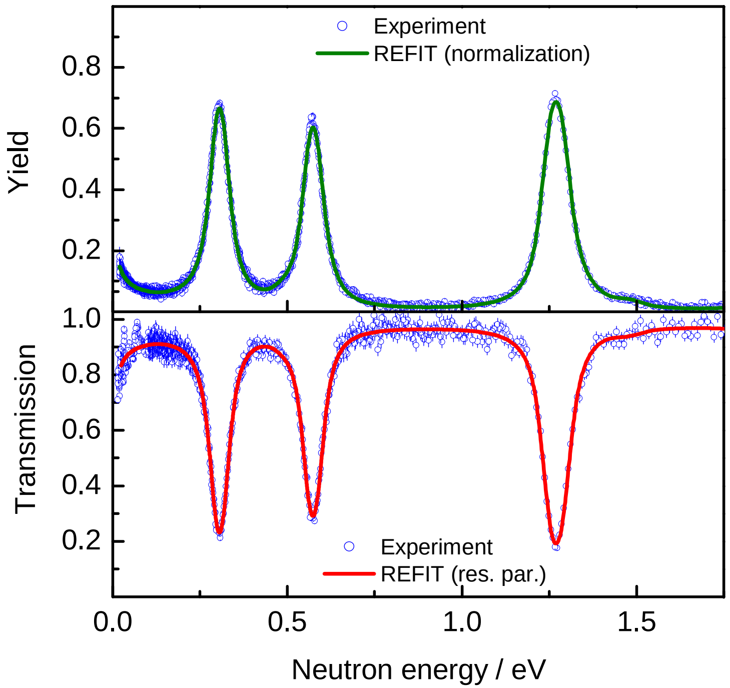
Transmission + capture at GELINA

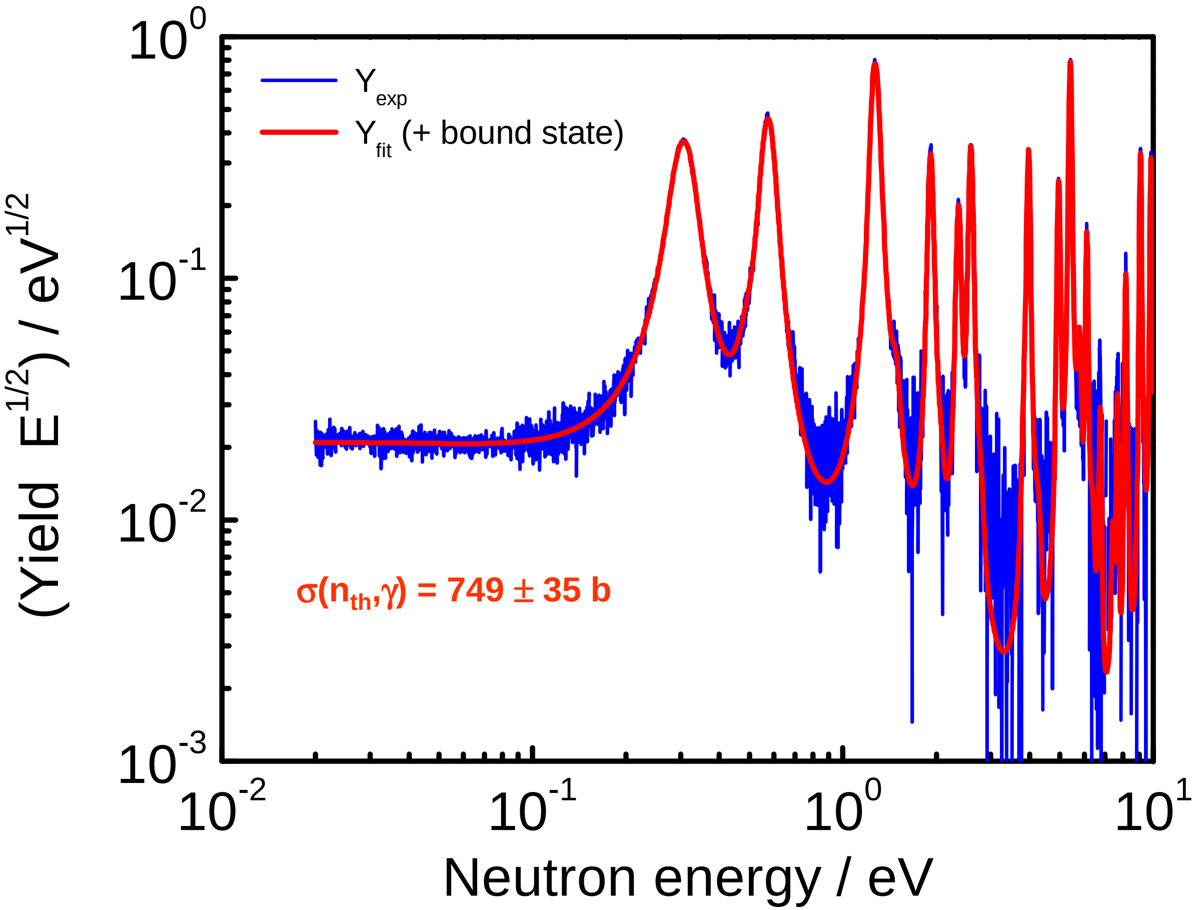


$N_c = 1.00 \pm 0.02$

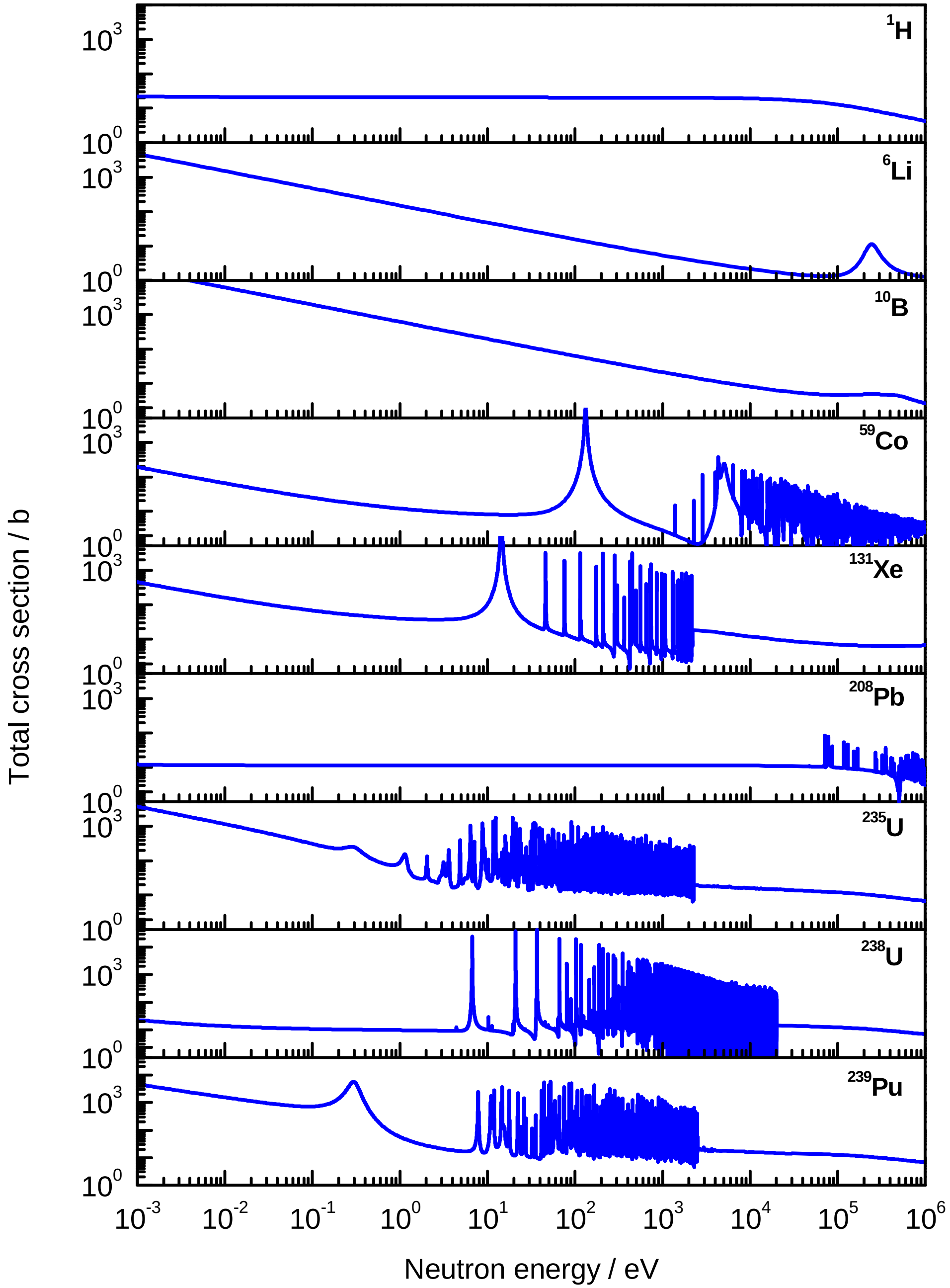
Energy / eV	Γ_n / meV		Γ_γ / meV	
0.306	0.064	(0.0004)	41.55	(0.39)
0.574	0.110	(0.0009)	42.11	(0.63)
1.272	0.373	(0.0035)	41.68	(0.79)

Transmission + capture at GELINA

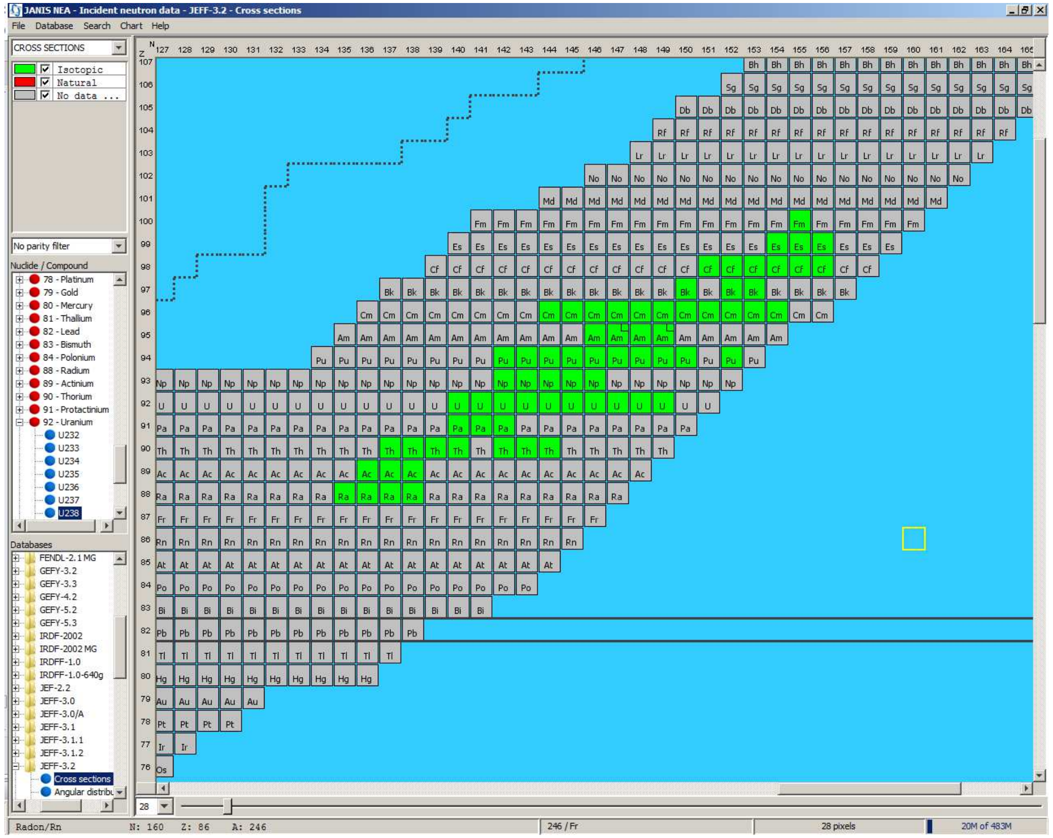




Cross sections for neutron induced reactions



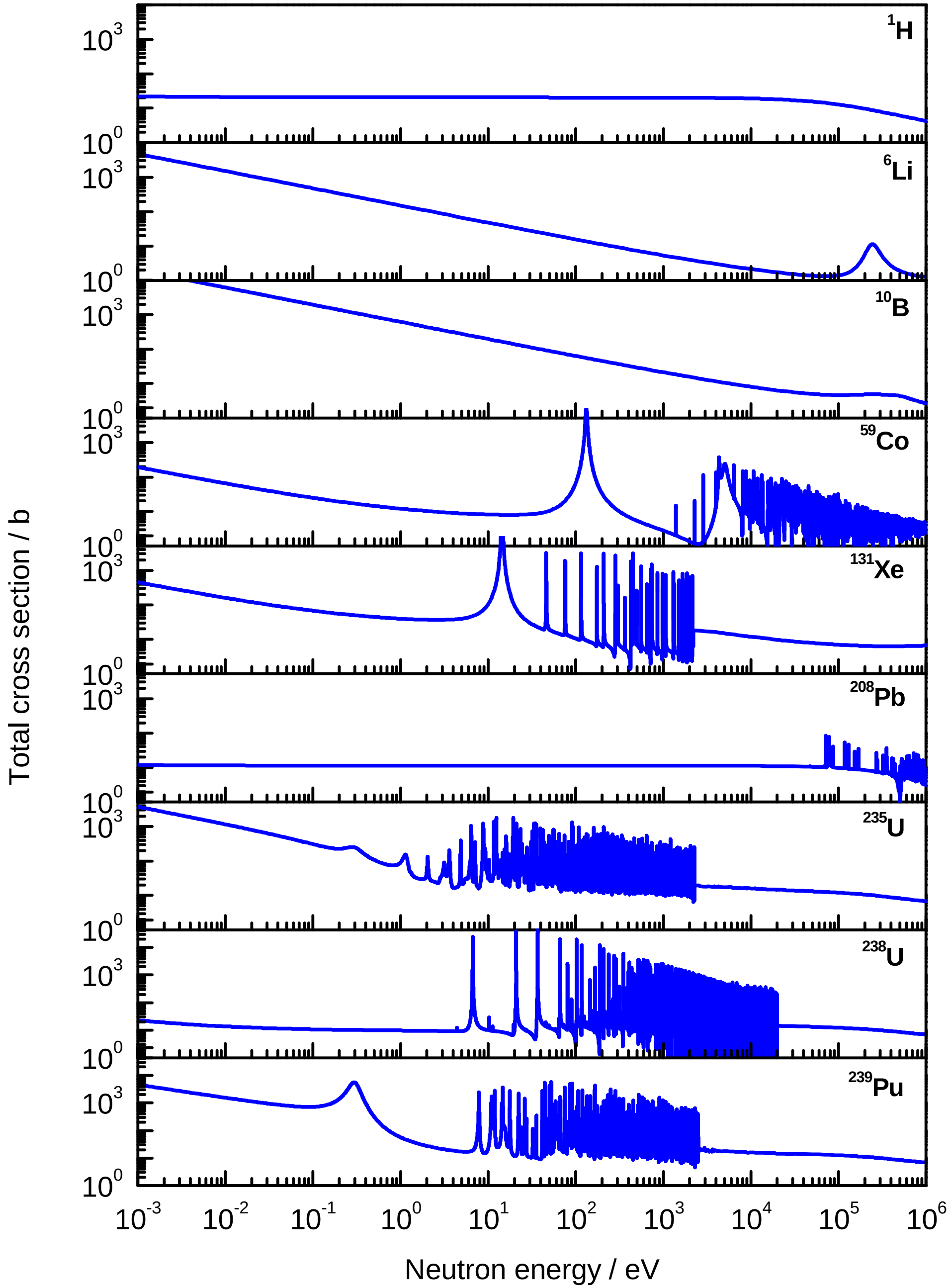
Evaluated data libraries



https://www.oecd-nea.org/janis/

European Commission

Neutron Resonance Analysis (NRCA&NRTA)



- Resonances appear at energies that are specific for each nuclide
- Position and amplitude of resonances can be used as fingerprints to
  - identify and quantify nuclides
  - elemental & isotopic composition
- Neutron Resonance Analysis (developed at JRC)
  - Non-Destructive Analysis (NDA)
  - sensitive to almost all nuclides (except light)
  - no sample preparation required
  - requirements:
    TOF-measurements at a white neutron source

Schillebeeckx et al., EUR Report 26848 EN

Cross section measurements

Total cross section

$$T \cong e^{-n\sigma_{tot}}$$

Capture cross section

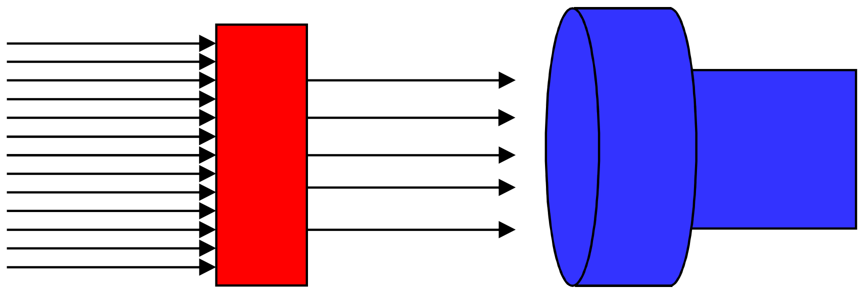
$$Y_\gamma \approx (1 - e^{-n\sigma_{tot}})\frac{\sigma_\gamma}{\sigma_{tot}}$$

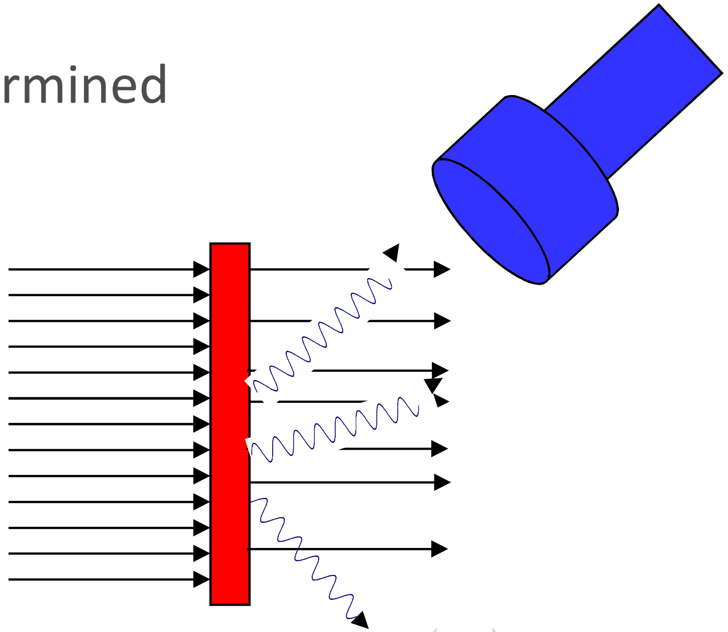
Well-characterised samples

n: total number of atoms per unit area is well-known

⇓

accurate cross-sections can be determined





Neutron resonance analysis

NRTA

$$T \cong e^{-n\sigma_{tot}}$$

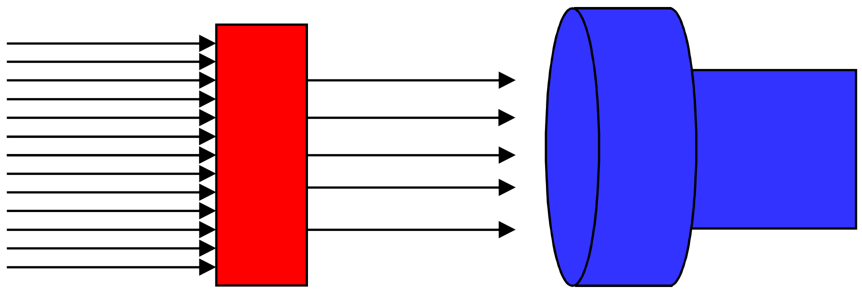
NRCA

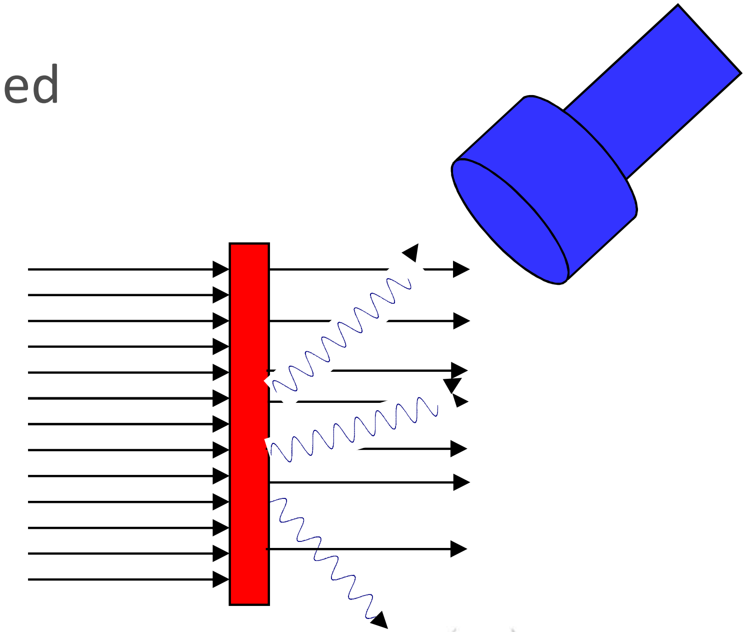
$$Y_\gamma \approx (1 - e^{-n\sigma_{tot}})\frac{\sigma_\gamma}{\sigma_{tot}}$$

Well-known cross sections

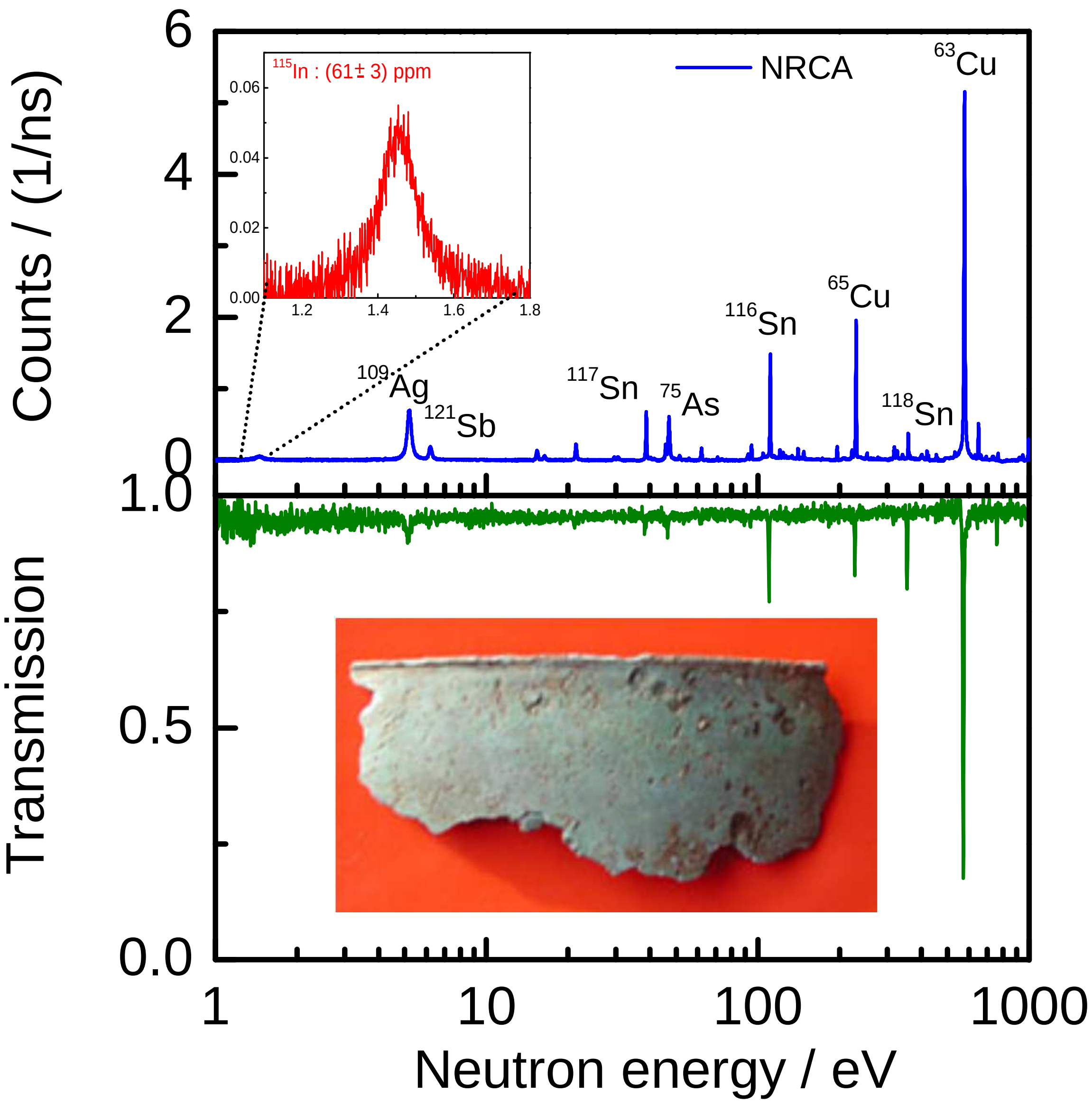
$\Downarrow$

areal density can be determined





NRTA and NRCA

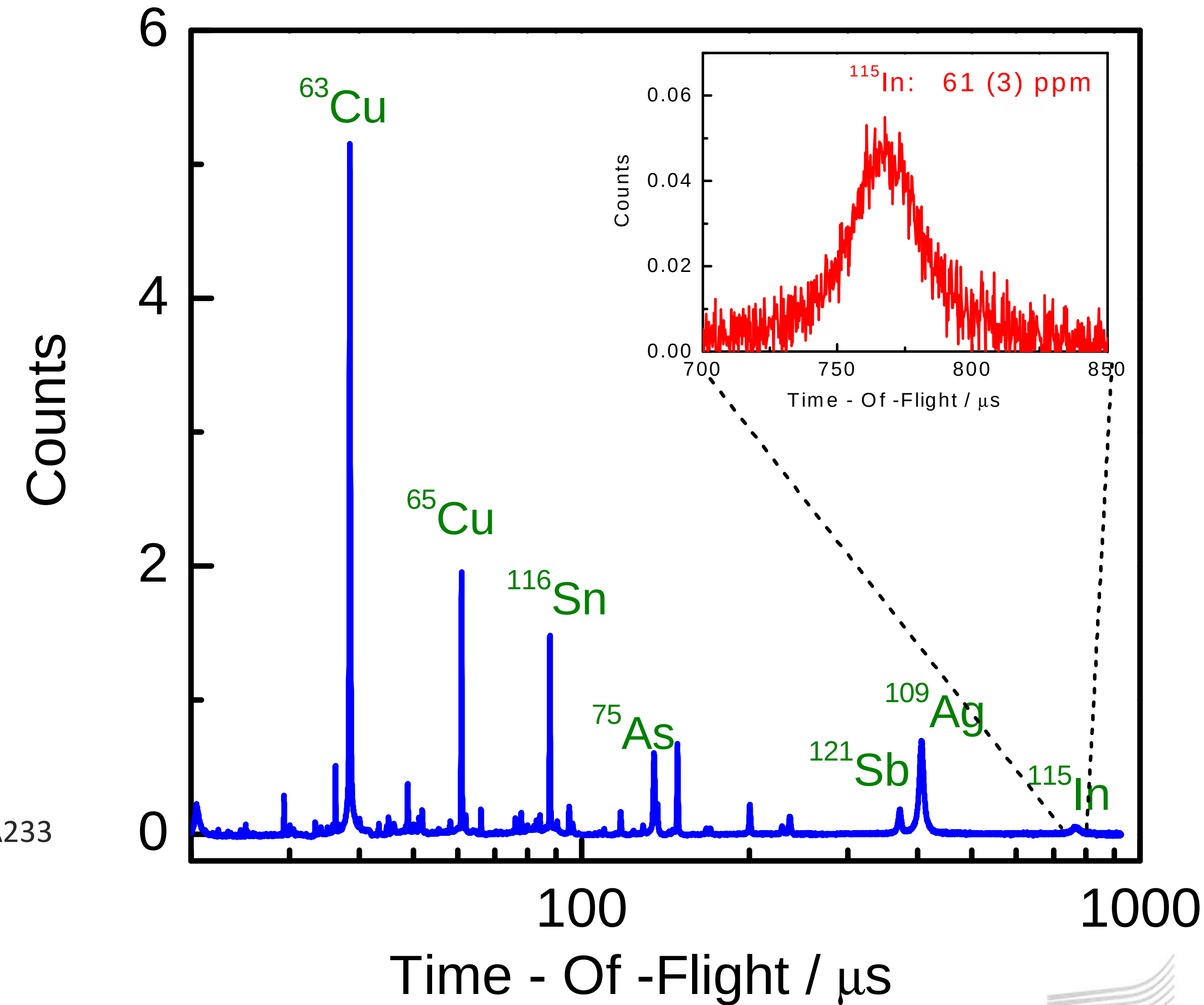


Element	wt%			Isotope	E_r / eV
Cu	**77.76**	±	**0.11**	^{63}Cu	579.0
				^{65}Cu	230.0
Sn	**20.85**	±	**0.10**	^{112}Sn	94.8
				^{116}Sn	111.2
				^{117}Sn	38.8
				^{118}Sn	45.7
				^{119}Sn	222.6
				^{120}Sn	427.5
				^{124}Sn	62.0
As	**0.34**	±	**0.01**	^{75}As	47.0
Sb	**0.20**	±	**0.02**	^{121}Sb	6.24
				^{123}Sb	21.4
Ag	**0.09**	±	**0.01**	^{107}Ag	16.3
				^{109}Ag	5.2
Fe	**0.77**	±	**0.10**	^{56}Fe	1147.4
In	**0.0061**	±	**0.0003**	^{115}In	1.46

NRCA ⇔ PGAA

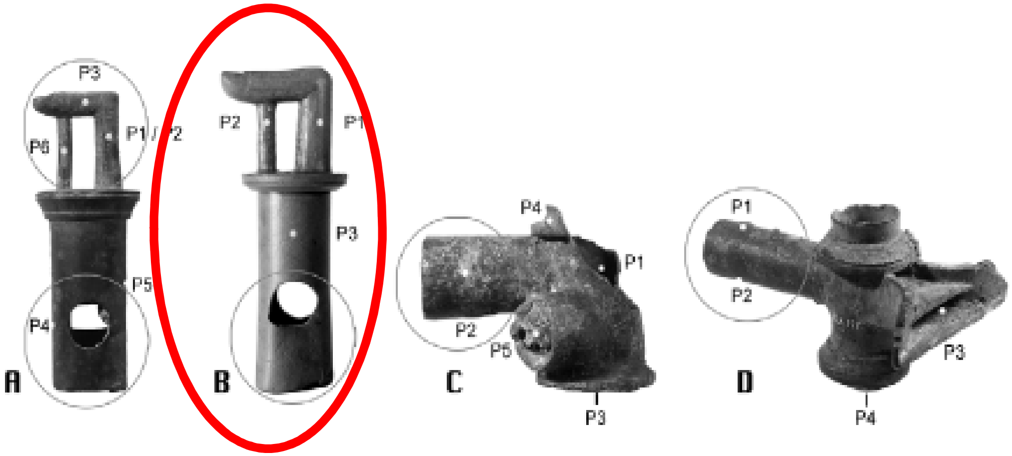
Typical dead time : 2.5 μs

⇒ detection of In is not hindered by strong resonances of other elements (e.g. Cu)

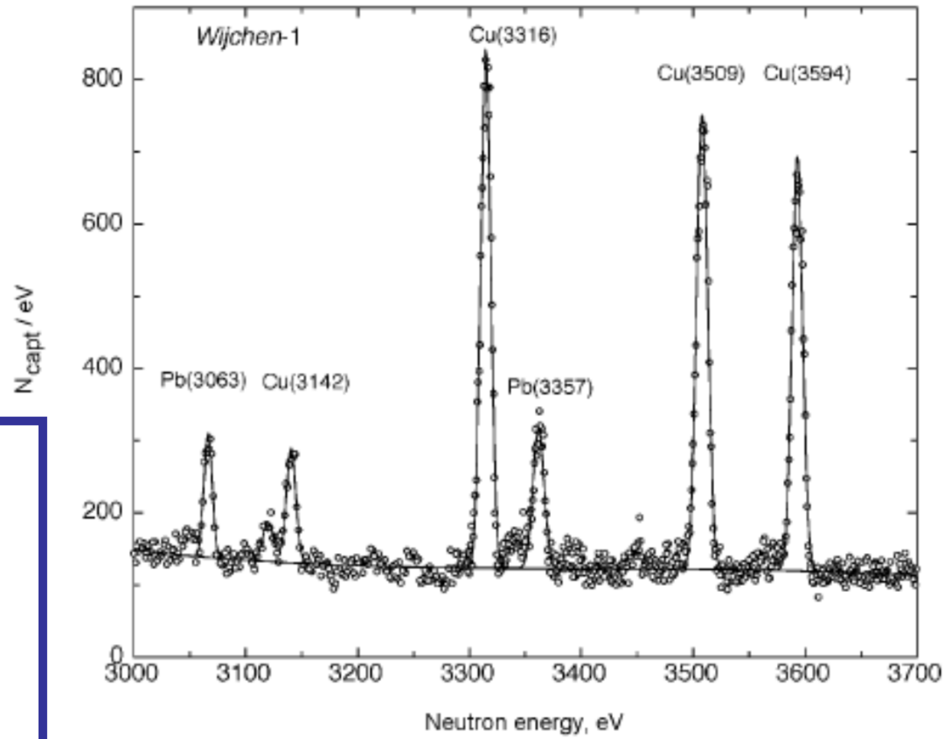


Postma et al., Czech. J. Phys. 53 (2003) A233

Roman mixer tap : hot – cold water (1/2)



	NRCA	Neutron Diffraction		
	materials used	fabrication process		
Cu	(4.8 ± 0.5) g/cm^2			
		P1	P2	P3
Sn/Cu	0.0868 ± 0.0025	0.094	0.0977	0.0940
Pb/Cu	0.335 ± 0.34	0.415	0.337	0.402
Sb/Cu	0.00167 ± 0.00003			
As/Cu	0.00098 ± 0.00003			
Ag/Cu	0.00096 ± 0.00003			
Zn/Cu	0.0036 ± 0.0003			
Fe/Cu	0.0012 ± 0.0003			

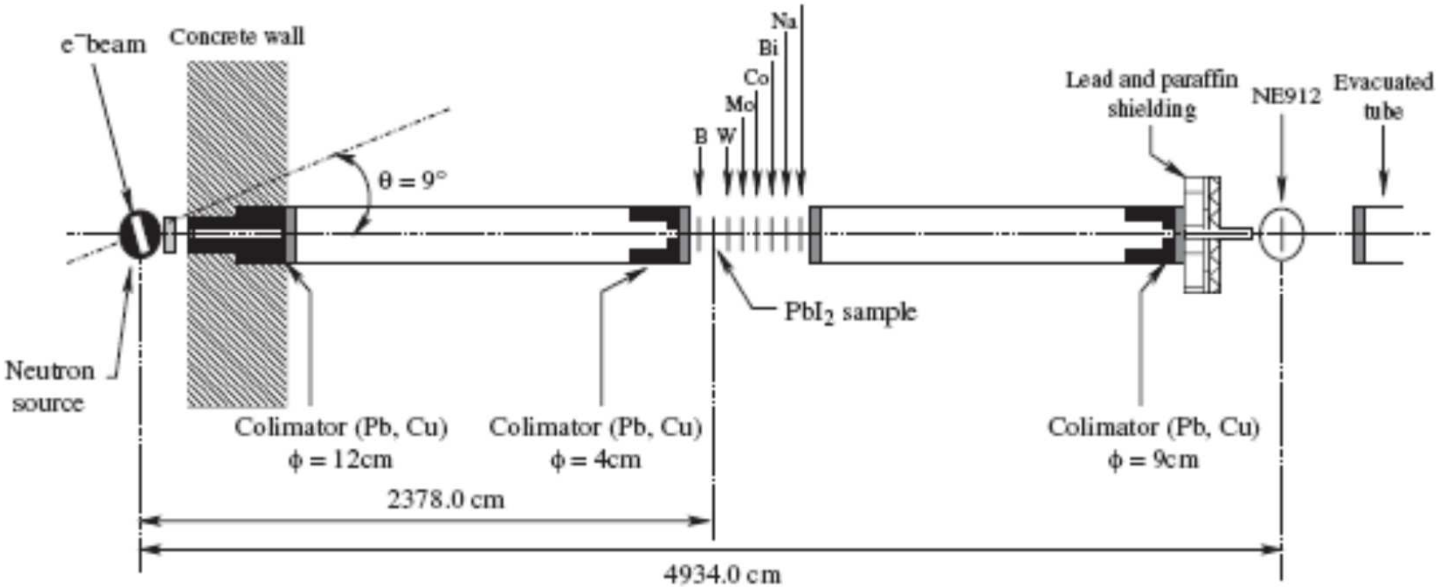


Schut et al., J. Radioanal. Nucl. Chem. 278 (2008) 151

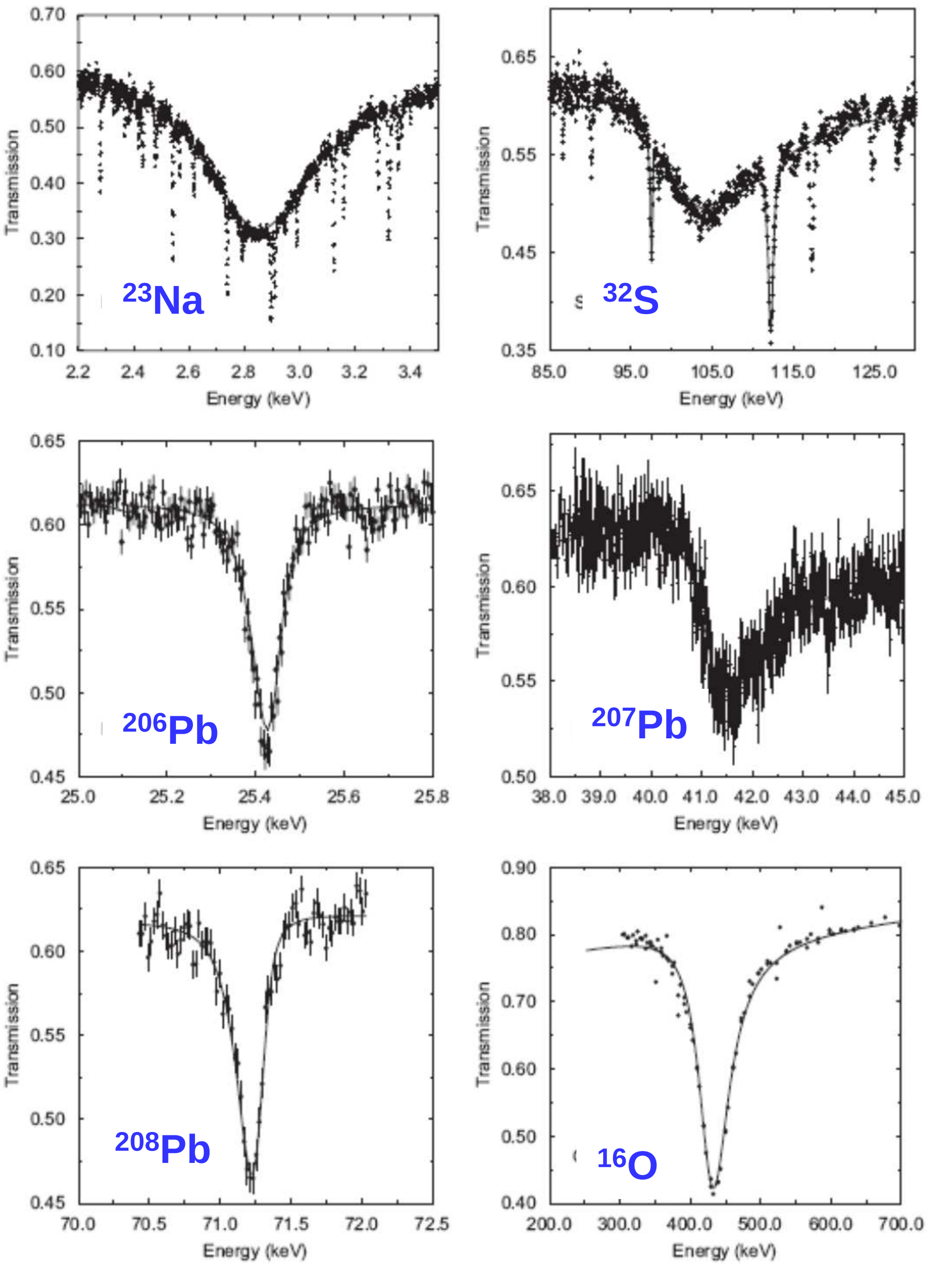
Characterisation of PbI_2 by NRTA at GELINA

- JRC Geel preparation group extracted:
  - 150 g Iodine (powder) from 210 liter from reprocessed waste (Le Hague)
  - (1.3 g/l Iodine and 40 MBq/l)
- Sample characterisation: by mass spectrometry , (N)AA and **NRTA**

Fig. 3. Addition of $Pb(NO_3)_2$ to the iodide solution to precipitate lead iodide.

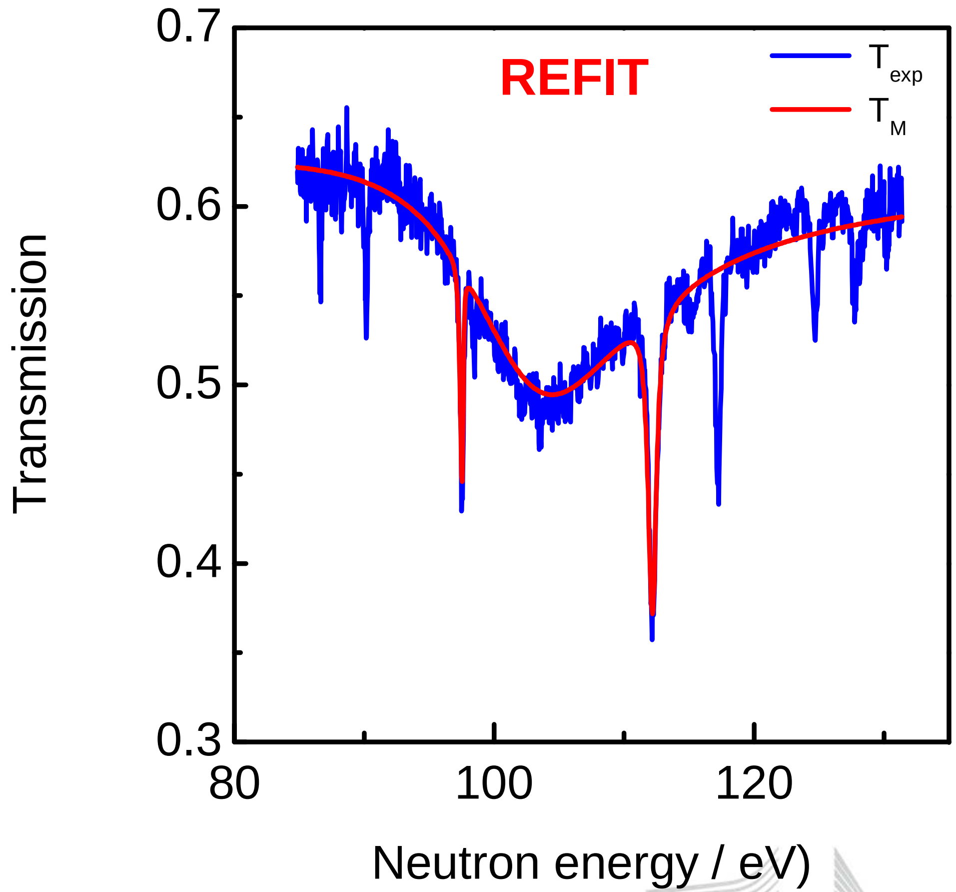


Characterisation of PbI_2 by NRTA at GELINA



$$T_M(t_m, n) = \int R(t_m, E)\, e^{-n\,\sigma_{tot}(E)} dE$$

$$\chi^2(n) = \left(T_{exp} - T_M(t_m, n)\right)^T V_{T_{exp}}^{-1} \left(T_{exp} - T_M(t_m, n)\right)$$



NRTA compared with NAA and ICP-MS

Element		NRTA (GELINA)		NAA		Mass spectrometry (PSI)	
Iodine	total	20.24	(0.41)	19.75	(0.61)	19.86	(0.41)
	^{127}I	3.44	(0.05)	3.35	(0.10)	3.36	(0.08)
	^{129}I	16.80	(0.40)	16.40	(0.60)	16.50	(0.40)
Lead	total	52.30	(1.70)	51.10	(1.80)		
	^{206}Pb	12.80	(0.50)				
	^{207}Pb	11.50	(0.10)				
	^{208}Pb	27.10	(1.70)				
Sulfur		5.44	(0.03)				
Sodium		0.72	(0.02)			1.00	(0.15)
Oxygen		13.92	(0.05)			14.50	(1.50)
Hydrogen		< 0.13				0.02	(0.002)
Nitrogen						1.20	(0.40)

NRTA :

- more elements have been analysed
- isotopic composition of Pb

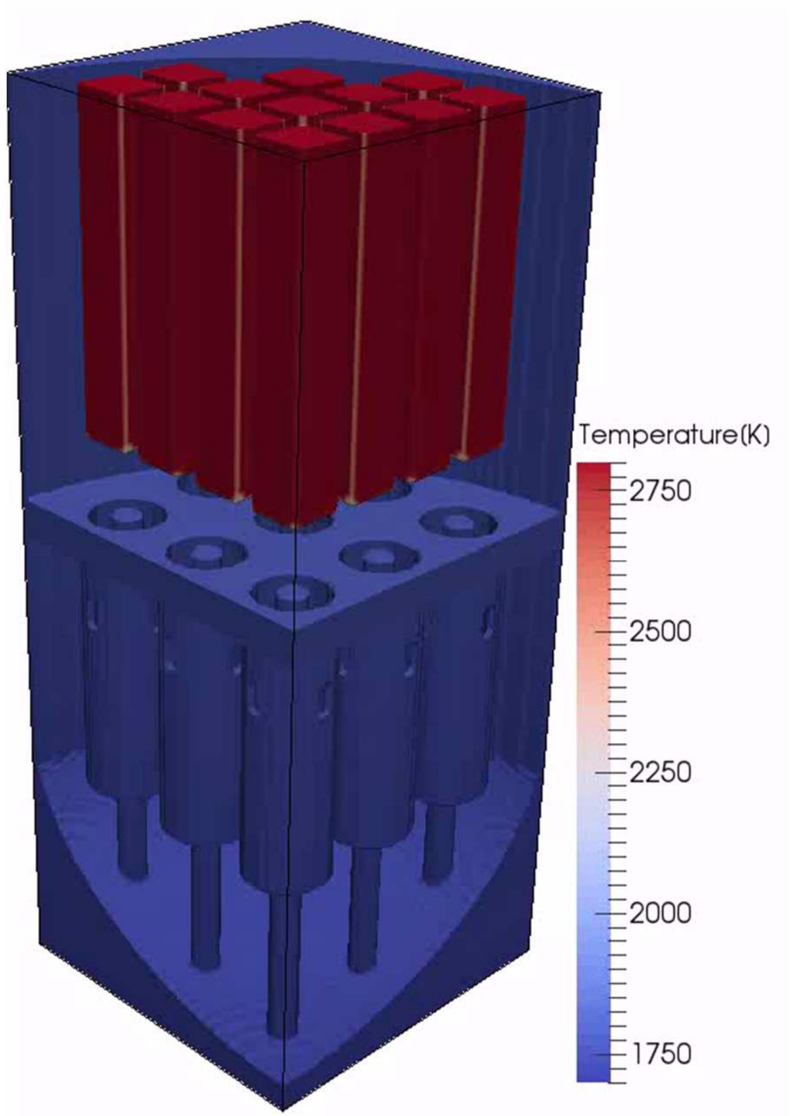
Noguere et al., NIMA 575 (2007) 476

European Commission

Fukushima accident

Earthquake followed by a Tsunami (15 m)

core meltdown (units 1,2,3)

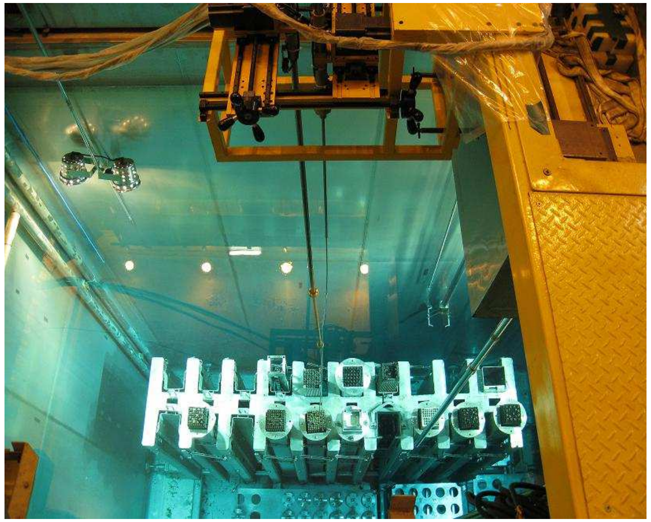


Melted fuel:

Complex mixture of materials in fuel and control/safety rods, contains

i.e. U, Pu, fission products, structural materials and neutron absorbers (^{10}B)

Decommissioning of Fukushima nuclear site



- **Removal of fresh and spent fuel assemblies (undamaged)**
  - started November 2013
- **Removal of melted fuel**
  - start removal within 10 yrs
  - completed after 20 ~ 25 yrs

- **Dismantling of the power plants**
  - completed after 30 ~ 40 yrs

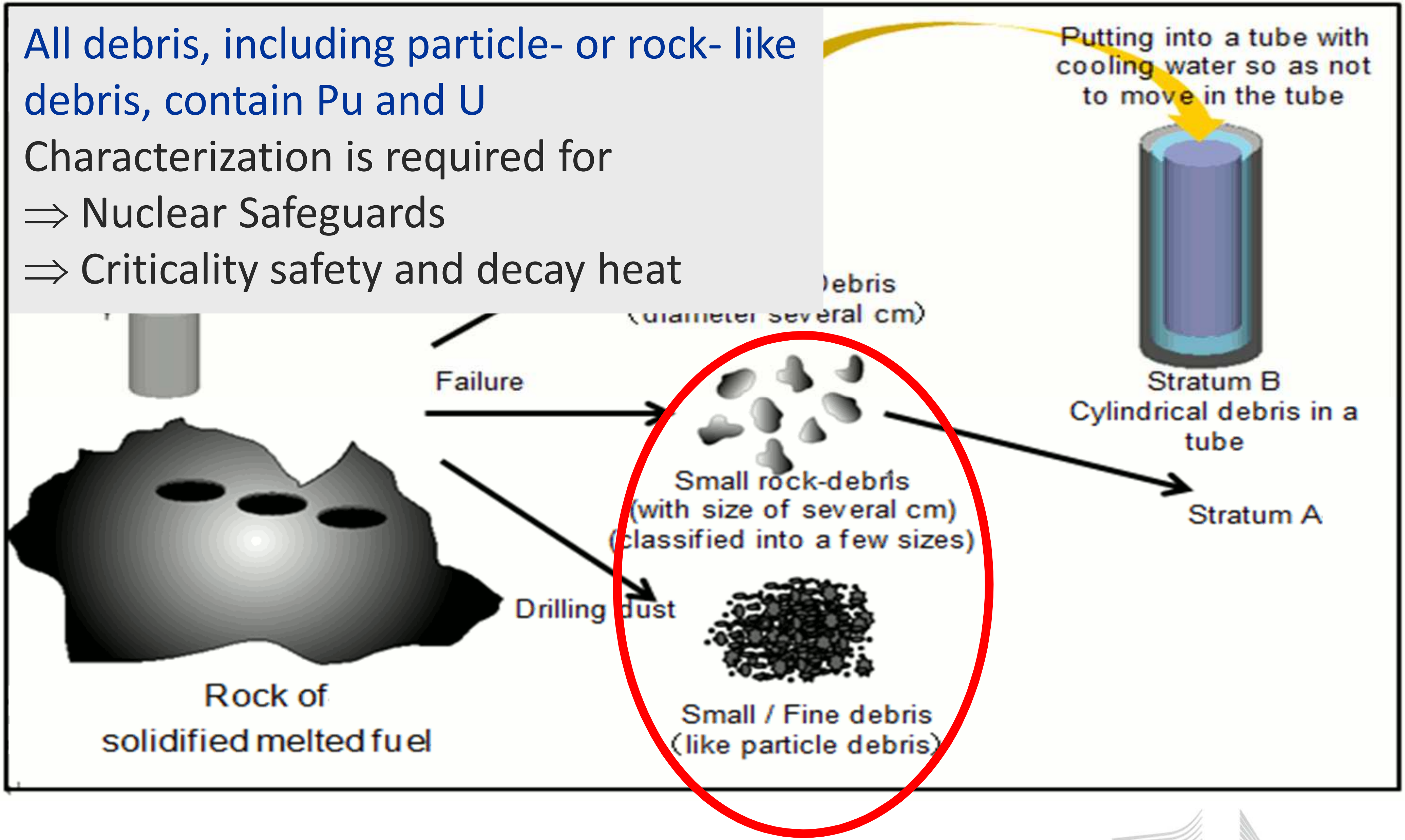
Removal of melted fuel: substantial amount of debris

All debris, including particle- or rock- like debris, contain Pu and U

Characterization is required for

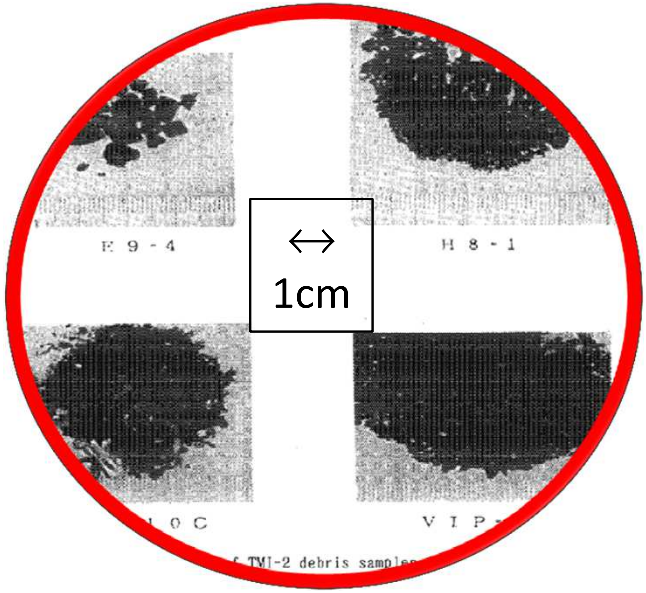
⇒ Nuclear Safeguards

⇒ Criticality safety and decay heat



Complex material to be verified

- Huge amounts of particle-& rock - like debris
- Technical Challenges:
  - Materials only remotely accessible
  - Unknown shape of target materials
  - High temperature
  - Complex mixture of materials
  - Unknown components: ^{10}B
  - Strong radioactivity: ^{137}Cs (about 10^8 Bq/g)



2012: At the time of the first decommissioning studies **no technique existed** to quantify the amount of U and Pu in such **particle - and rock - like debris of melted fuel**

Characterization of debris of melted fuel by NRTA

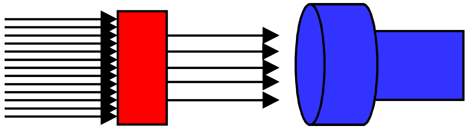
Target value: uncertainty on Pu and U content ≤ 2%

Challenges due to the material characteristics:

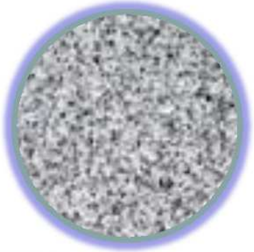
- **Inhomogeneity of the samples**: due to diversity in shape and size of the particle - & rock-like debris samples
- **Impact of impurities**: structural material and neutron absorbers, i.e. ^{10}B (control rods and borated water)
- **Complex transmission spectra due to fission products**

Impact of particle size distribution

Transmission is a non-linear function of n



- Homogeneous sample : $T = e^{-n\,\sigma_{tot}}$
- Heterogeneous sample : $< T > = < e^{-n\,\sigma_{tot}} > \neq e^{-<n>\,\sigma_{tot}}$



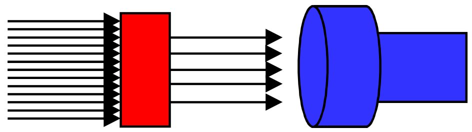
<n> is the quantity of interest

Homogeneous model can lead to an error of 15%

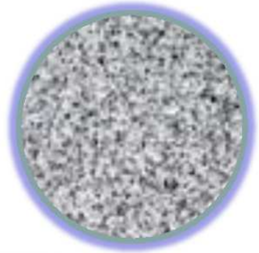
$\Rightarrow$ Dedicated model is required

Impact of particle size distribution

Transmission is a non-linear function of n



- Homogeneous sample : $T = e^{-n\,\sigma_{tot}}$



- Heterogeneous sample : $\langle T \rangle = \langle e^{-n\,\sigma_{tot}} \rangle \neq e^{-\langle n \rangle\,\sigma_{tot}}$

Levermore-Pomraning model (J. Math. Phys. 27, 2526, (1986))

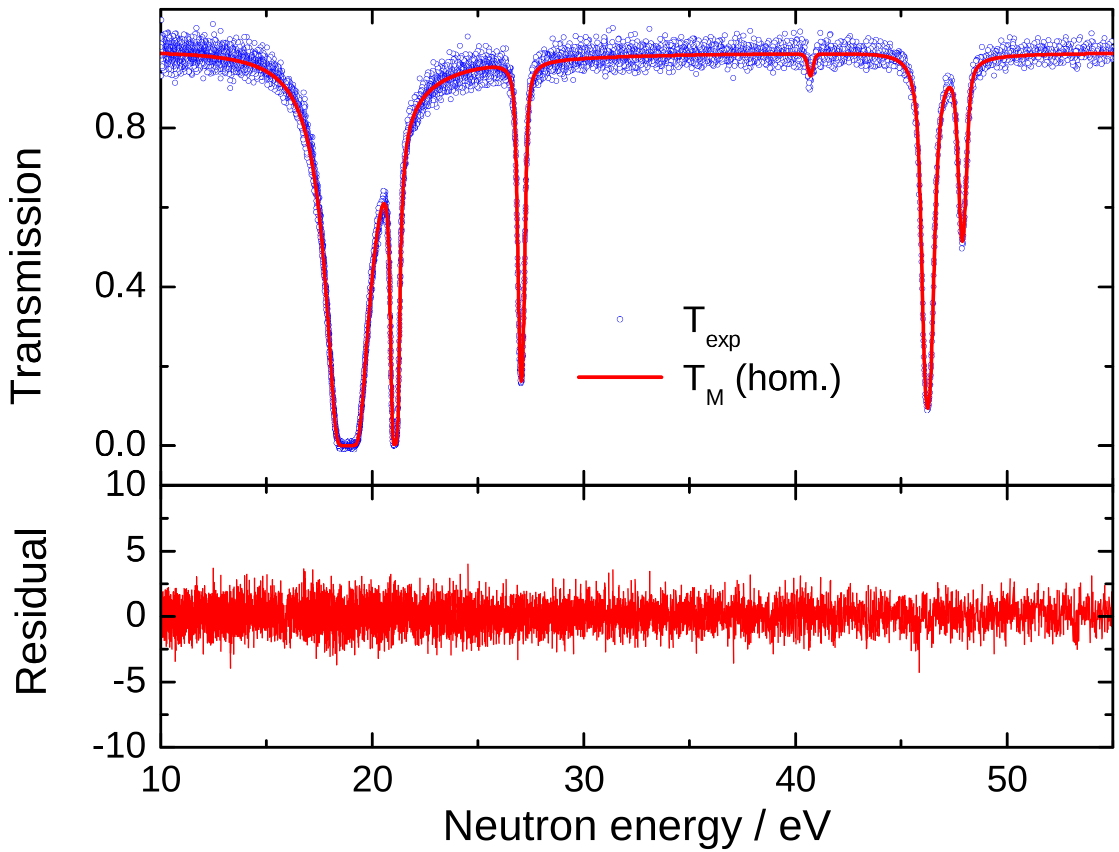
- widely used for other problems dealing with radiation transport through stochastic media, e.g. scattering of sunlight in clouds
- starts from microscopic properties of the sample such as grain size
- in particular applicable for powder samples

implemented in REFIT and validated by experiments at GELINA

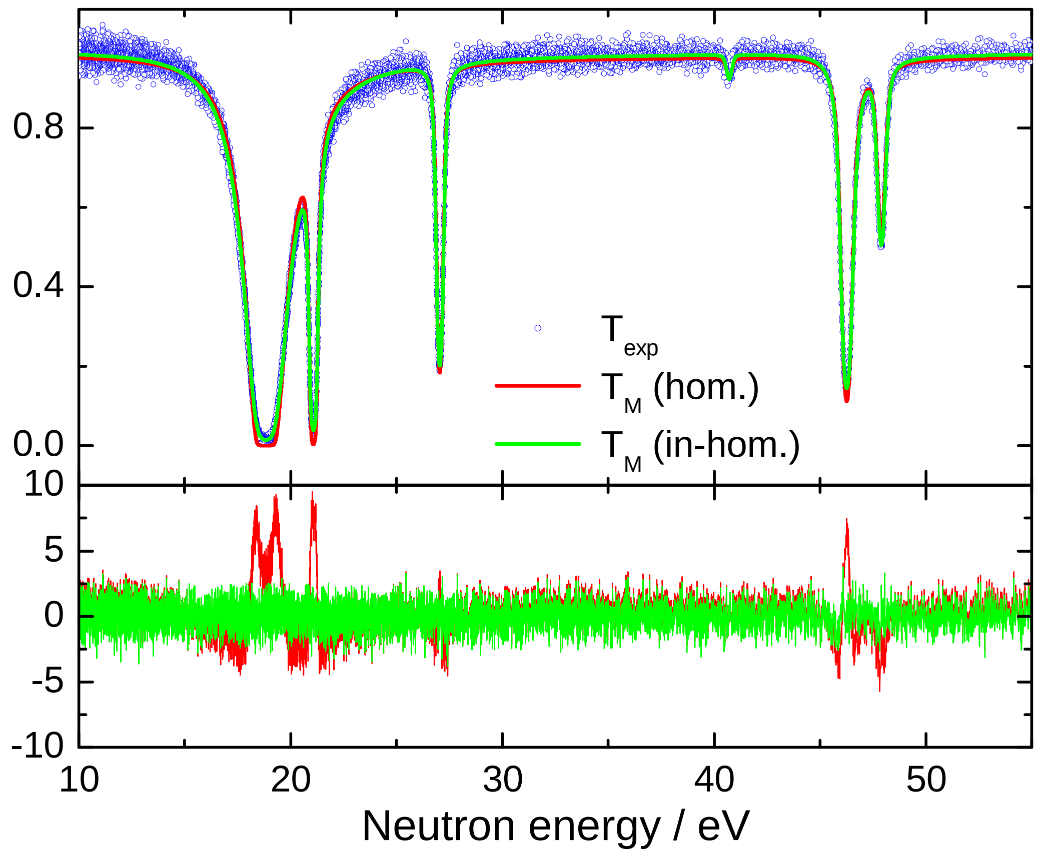
Becker et al., Eur. Phys. J. Plus 129 (2014) 58

Experimental validation of LP-model at GELINA

Declared : n_W = 9.38 10^{-4} at/barn
T_M (hom.) : n_W = 9.36 10^{-4} at/barn



Declared : n_W = 1.03 10^{-5} at/barn
T_M (inhom.) : n_W = 1.05 10^{-5} at/barn



^{nat}W-metal disc
⇒ bias < 1%

^{nat}W-powder mixed with ^{nat}S-powder
LP - model ⇒ bias ≤ 2 %

Becker et al., EPJ Plus 129, 58 (2014)

NRTA demonstration experiment at GELINA

Samples
- 18 different samples
- 8 different elements

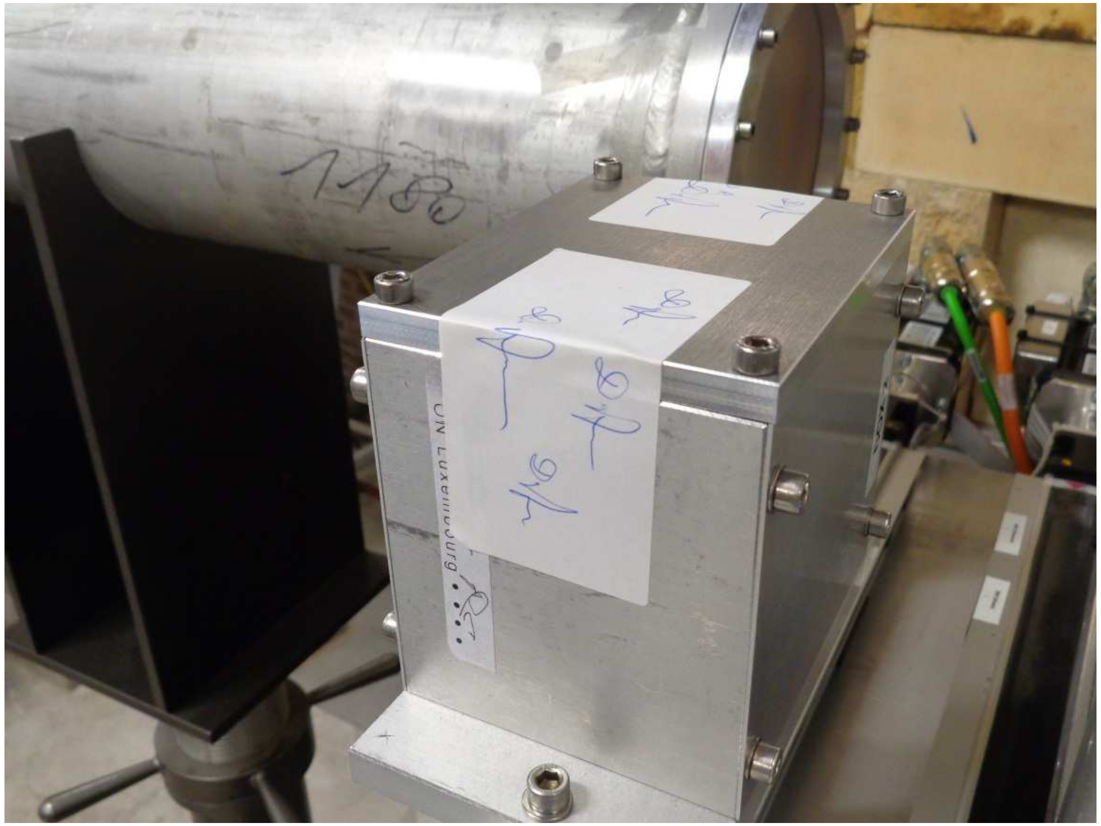
Black box: 8 slots

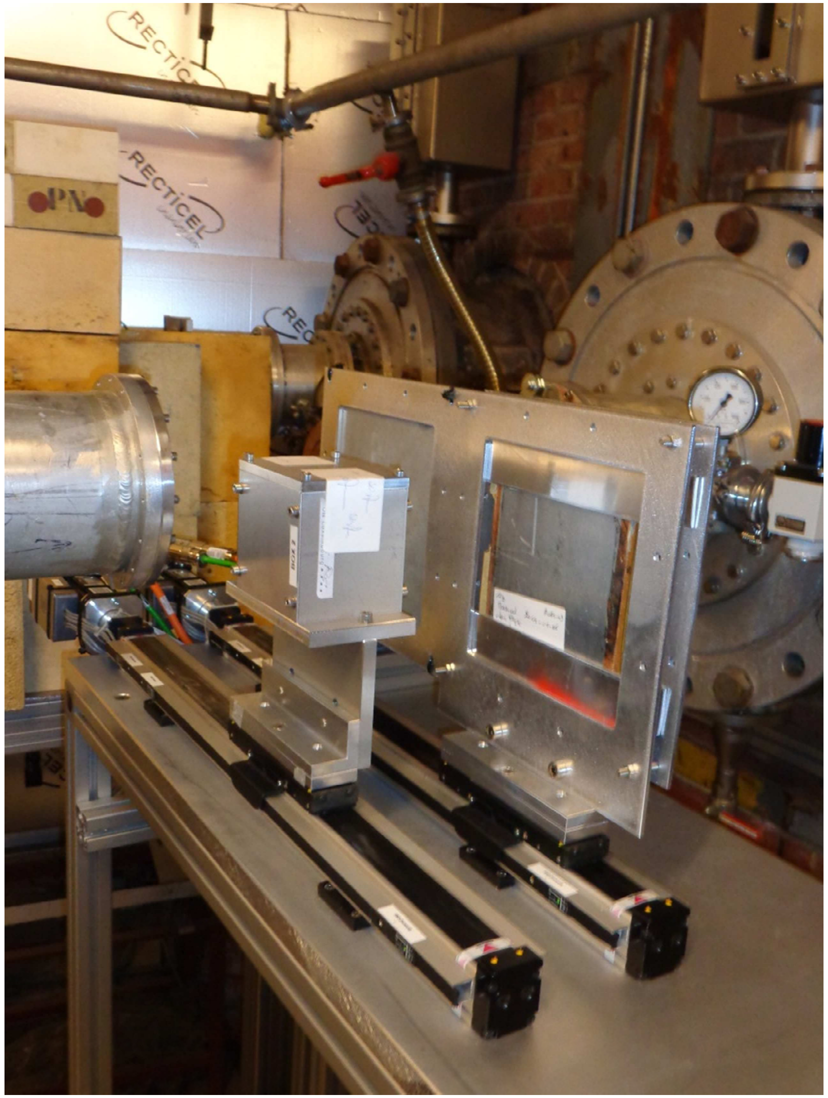
B, Mn, Co, Cu, Nb, Rh, W, Au samples with different thicknesses

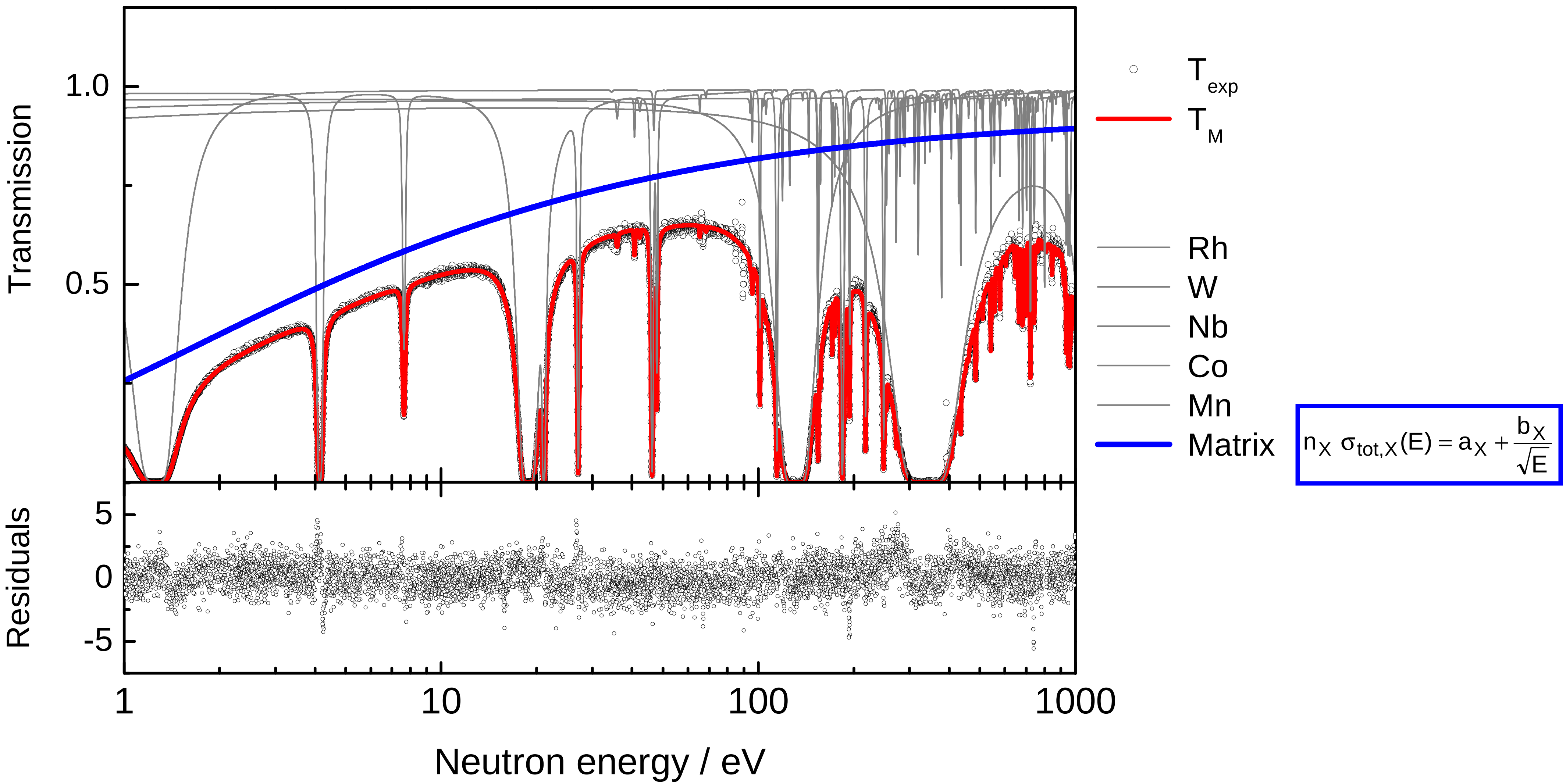
Selection of samples by DG-ENER, IAEA and DOE representatives

NRTA demonstration experiment at GELINA

NRTA station at 10 m

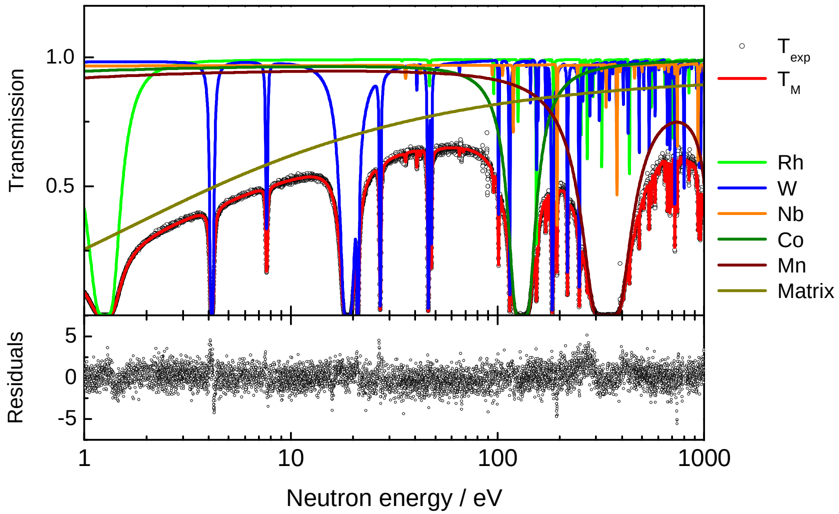





Transmission
1.0
0.5
Residuals
5
0
-5
1
10
100
1000
Neutron energy / eV
T_{exp}
T_M
Rh
W
Nb
Co
Mn
Matrix
$n_X\ \sigma_{tot,X}(E) = a_X + \frac{b_X}{\sqrt{E}}$

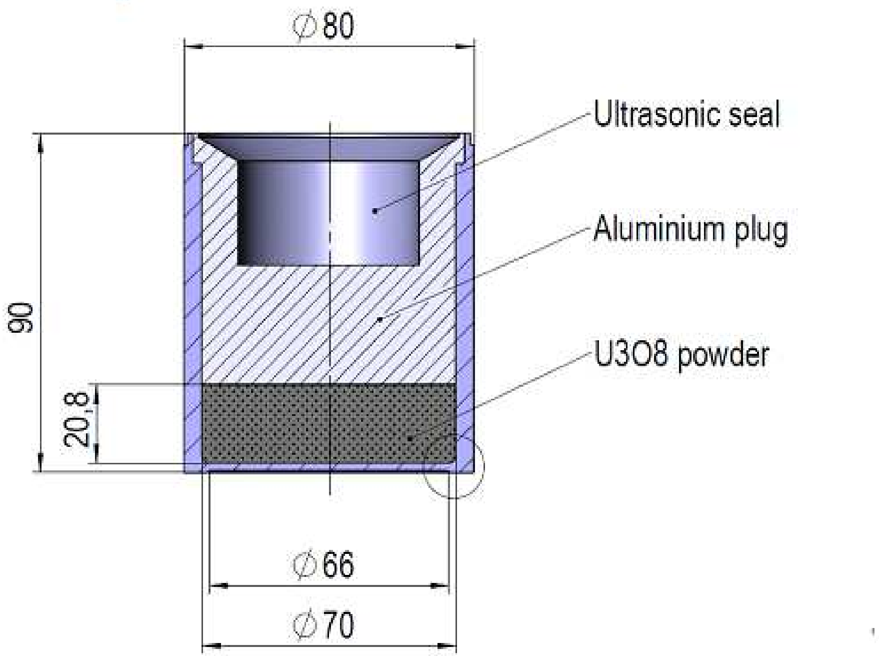
NRTA demonstration experiment at GELINA

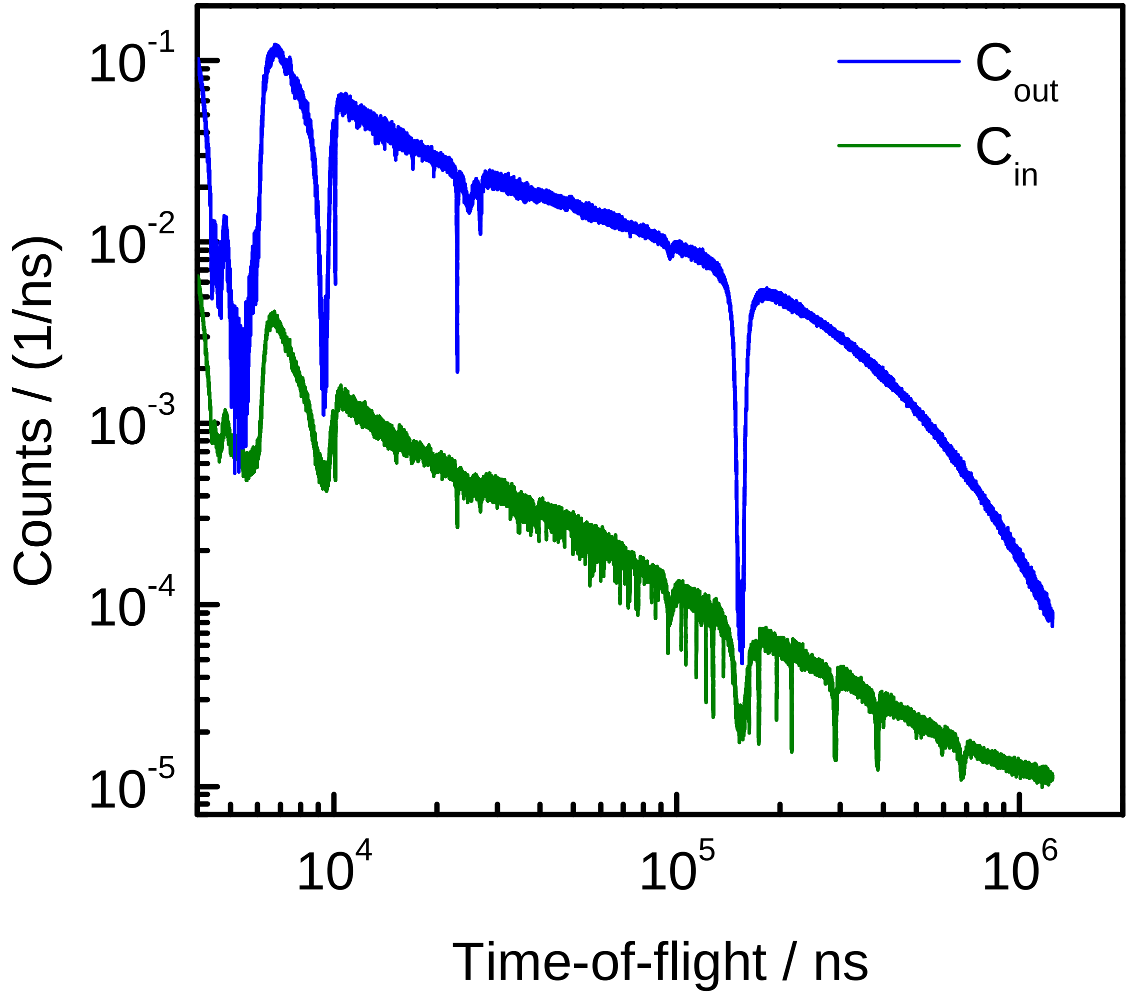
Element	Areal number density (at/barn)		n_{Ref}/n_{NRTA}
	n_{Ref}	n_{NRTA}	
Mn	1.901×10^{-2}	$(1.928 \pm 0.003) \times 10^{-2}$	1.014 ± 0.002
Co	4.583×10^{-3}	$(4.509 \pm 0.015) \times 10^{-3}$	0.984 ± 0.003
Cu	0	0	
Nb	5.485×10^{-3}	$(5.382 \pm 0.010) \times 10^{-3}$	0.981 ± 0.002
Rh	1.856×10^{-3}	$(1.891 \pm 0.003) \times 10^{-3}$	1.019 ± 0.002
W	2.269×10^{-3}	$(2.250 \pm 0.002) \times 10^{-3}$	0.992 ± 0.001
Au	0	0	



NRTA: nuclear material GELINA

U_3O_8 reference sample
EC NRM 171





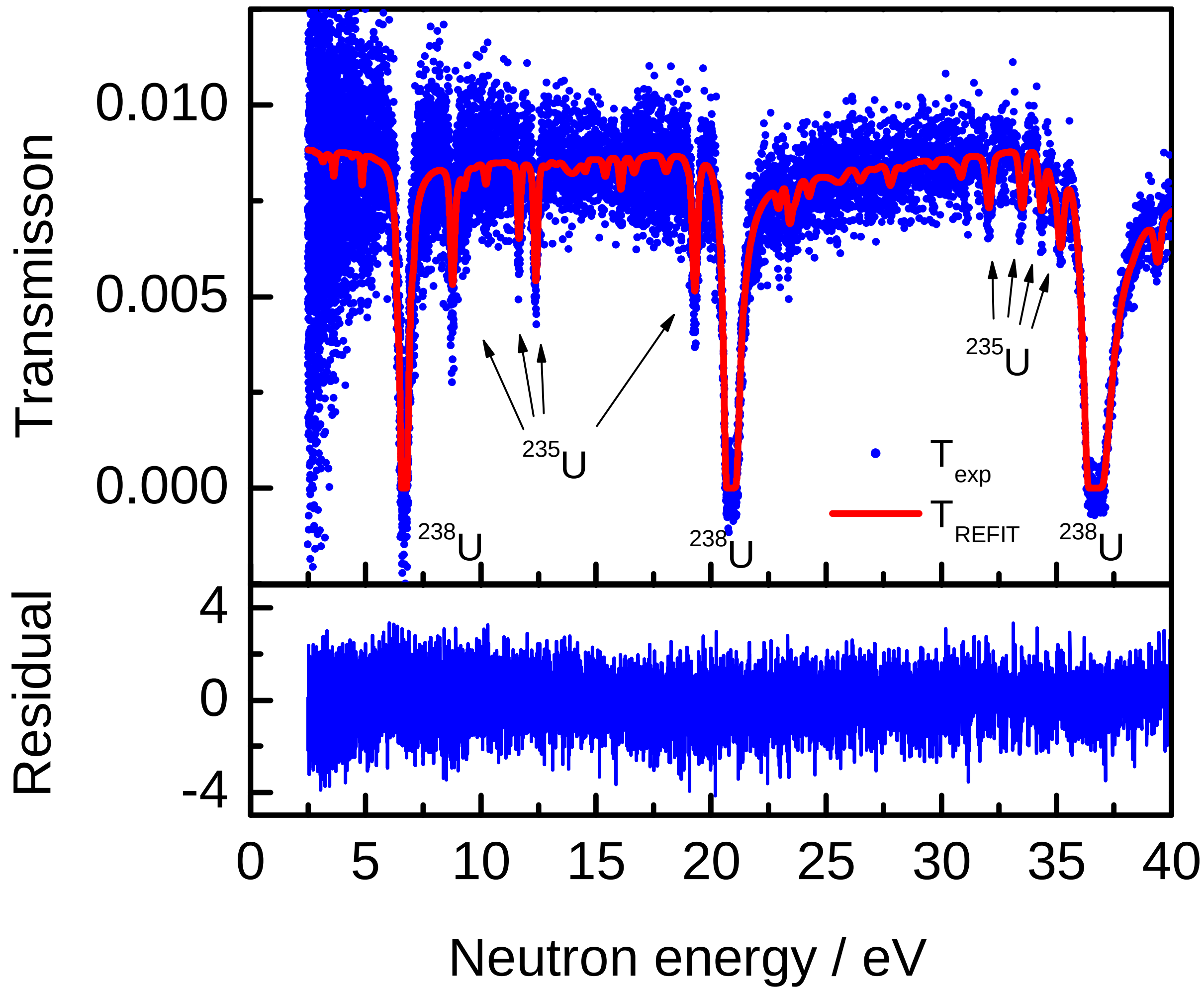
- Strong impact of matrix material
- **Beam attenuation due to matrix ~ 99%**

NRTA of nuclear material at GELINA

U_3O_8 reference sample
EC NRM 171

Fit for ^{235,238}U areal density
+

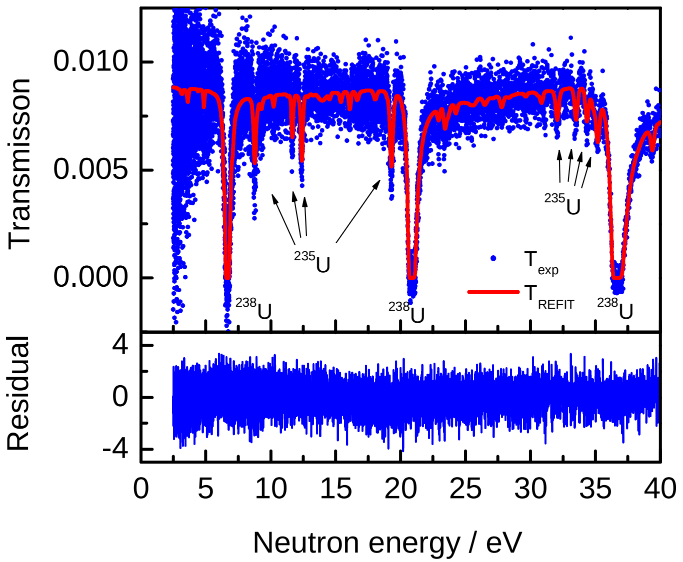
$$n_X \, \sigma_{tot,X}(E) = a_X + \frac{b_X}{\sqrt{E}}$$



NRTA of nuclear material at GELINA

U_3O_8 reference sample
EC NRM 171

U-isotope	Areal number density (at/b)		Ratio
	Declaration	NRTA	
^{235}U	$(5.0326 \pm 0.0080) \times 10^{-4}$	$(5.063 \pm 0.09) \times 10^{-4}$	**1.006**
^{238}U	$(1.0628 \pm 0.0015) \times 10^{-2}$	$(1.062 \pm 0.01) \times 10^{-2}$	**0.999**



$\Rightarrow$ bias < 1.0 %

Schillebeeckx et al., JRC Science and Policy Report, EUR 26848 EN

Thank you for your attention

Transnational access to nuclear facilities at JRC Geel

https://ec.europa.eu/jrc/en/eufrat/contacts

Course on

"Nuclear data processing and use in nuclear applications"

http://gentleproject.eu/courses/schedule/

JRC Geel , 14 – 18 November 2016