2141-25

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

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Cross section measurements and uncertainties of cross section data

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Cross section measurements and uncertainties of cross section data

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http://www.jrc.ec.europa.eu/

Overview

General introduction

Some detailed measurement examples

Uncertainties in measurement

Some highlights of new possibilities

Measurement design

Quantity to measure (measurand)

cross section(s)
reaction parameter(s)

Measurement principle

activation, emitted particle detection, …

Expression of the quantity in terms of control and influence quantities

Identification of possible influence quantities (sources of error)

Method of measurement

Sequence of logical steps
how to fix control quantities
how to correct for other influence quantities

Measurement procedure

Detailed prescription
- *Physical operations*
- *Data manipulations*

Arriving at
- *Measurement value*
  - *Corrected*
- *Uncertainties*
  - *Complete*
  - *Correlations*

Evaluation of measurement uncertainty – Guide to the expression of uncertainty in measurement, JCGM 100:2008, www.bipm.org (2008)

Method of measurement

‘Hardware’

- **Neutron source/collimation**
- **Sample**
- **Detection equipment fluence or normalization**
- **Detection equipment process rate**
- **Data acquisition**
- **Peripheral control**
- **Ancillary measurements**

‘Software’

Measurement sequence
(foreground, background, iterate over samples, other experimental conditions, sample characterization, calibration)

Evaluation of data
Selection criteria
Data reduction
Determination of values, uncertainties and correlations

Measurements

There is a large variety even for one particular quantity

Specific examples worked out in more detail

Highlights to show the range of possibilities

Specific examples

Transmission in the resonance range

Capture

Inelastic scattering by the (n,n'γ)-technique

Activation
Uncertainties for activation

Drawn from experience at the IRMM neutron sources GELINA and the 7 MV VdG accelerator

Total cross section & transmission + time-of-flight

$$T \equiv \frac{C_{\text{sample in}}}{C_{\text{sample out}}} = e^{-N d \sigma_T} = \frac{Y^c_{\text{in}} - B^c_{\text{in}}}{Y^c_{\text{out}} - B^c_{\text{out}}}$$

Attenuation measurement

σ_T = the total cross section

T = the transmission factor

C = Corrected counts in the detector

N = the nuclide concentration

d = the sample thickness

Y^c = Total counts

B^c = Background counts

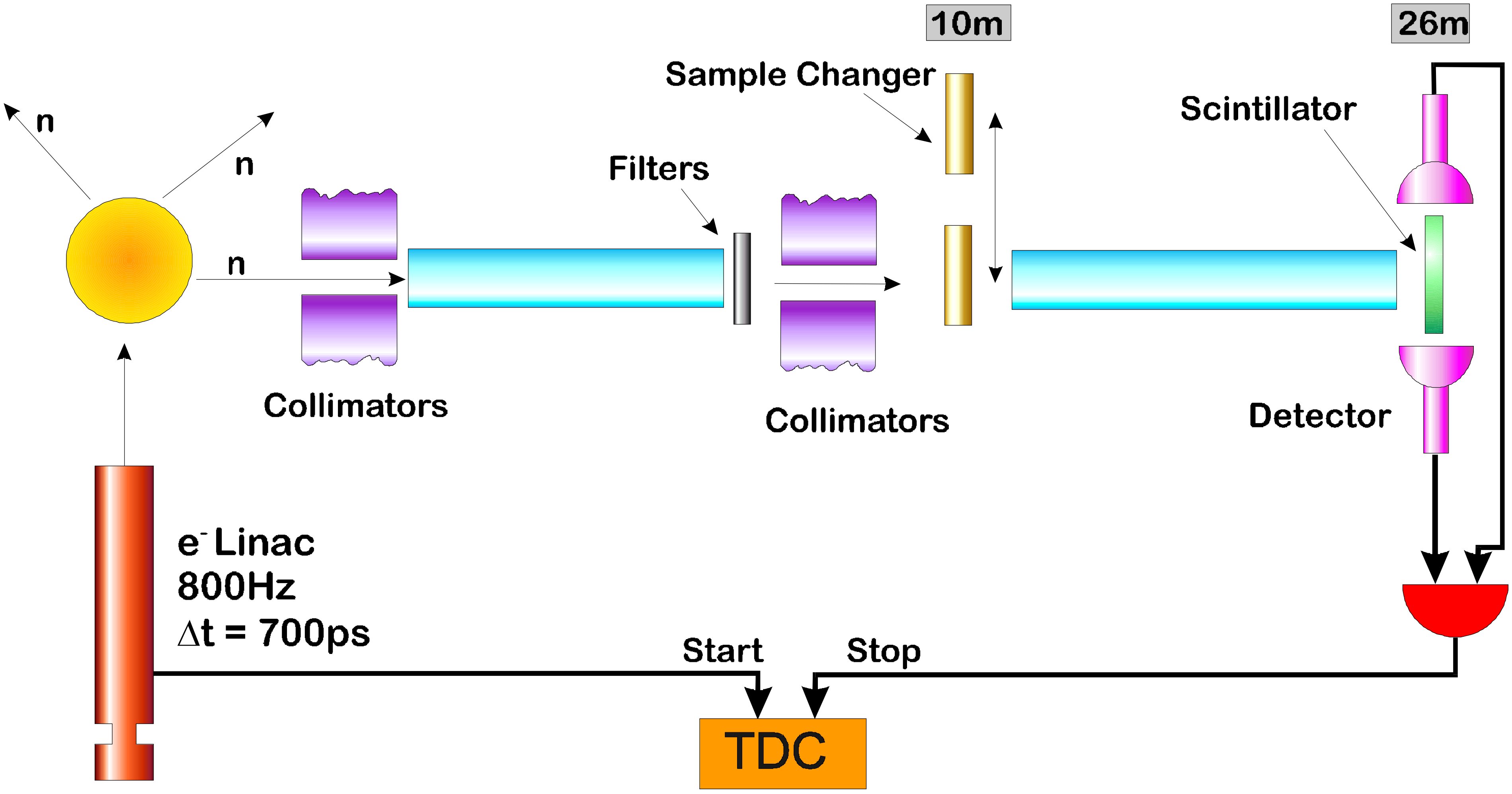
$\circ^c$ = deadtime corrected&normalized

Influence quantities

- *N d: nuclides per unit area*
- *background*
- *other nuclides in sample*
- *sample container wall*
- *sample homogeneity*
- *collimation*
- *temperature*
- *detector+monitor stability*
- *flight path length*
- *resolution functions*
    - *neutron source*
    - *detector*
- *deadtime*

Total cross section & transmission + time-of-flight

Transmission method



GELINA, a multi-user facility

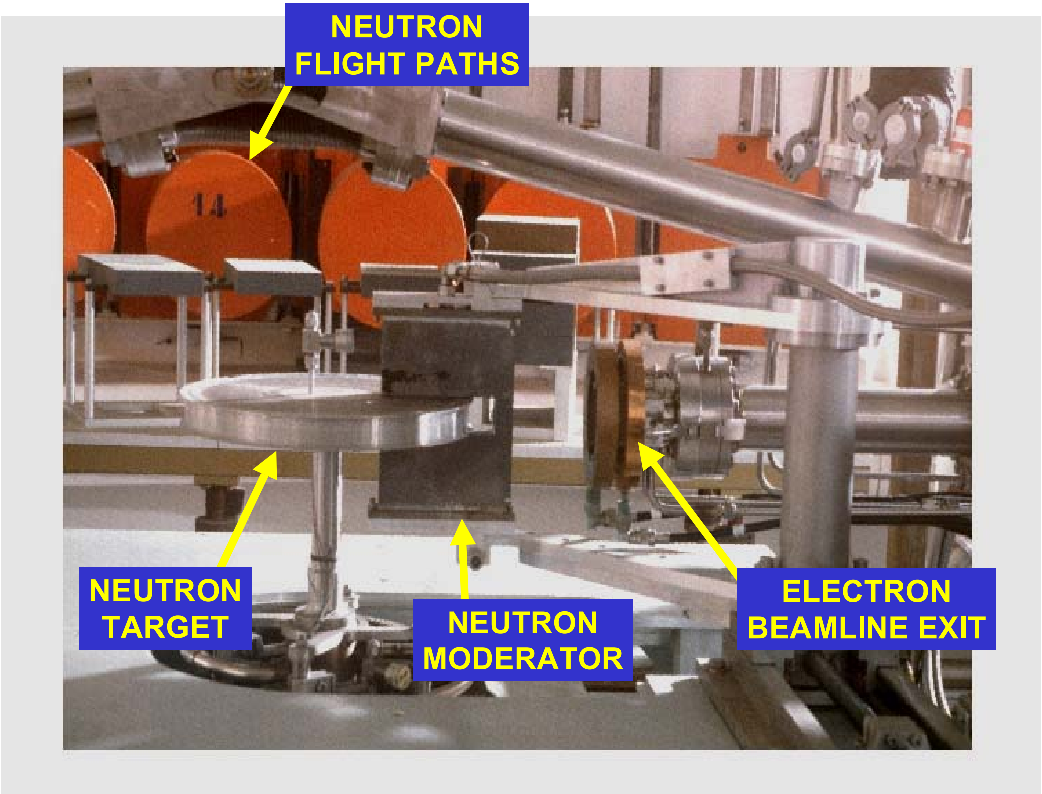
Neutron conversion target

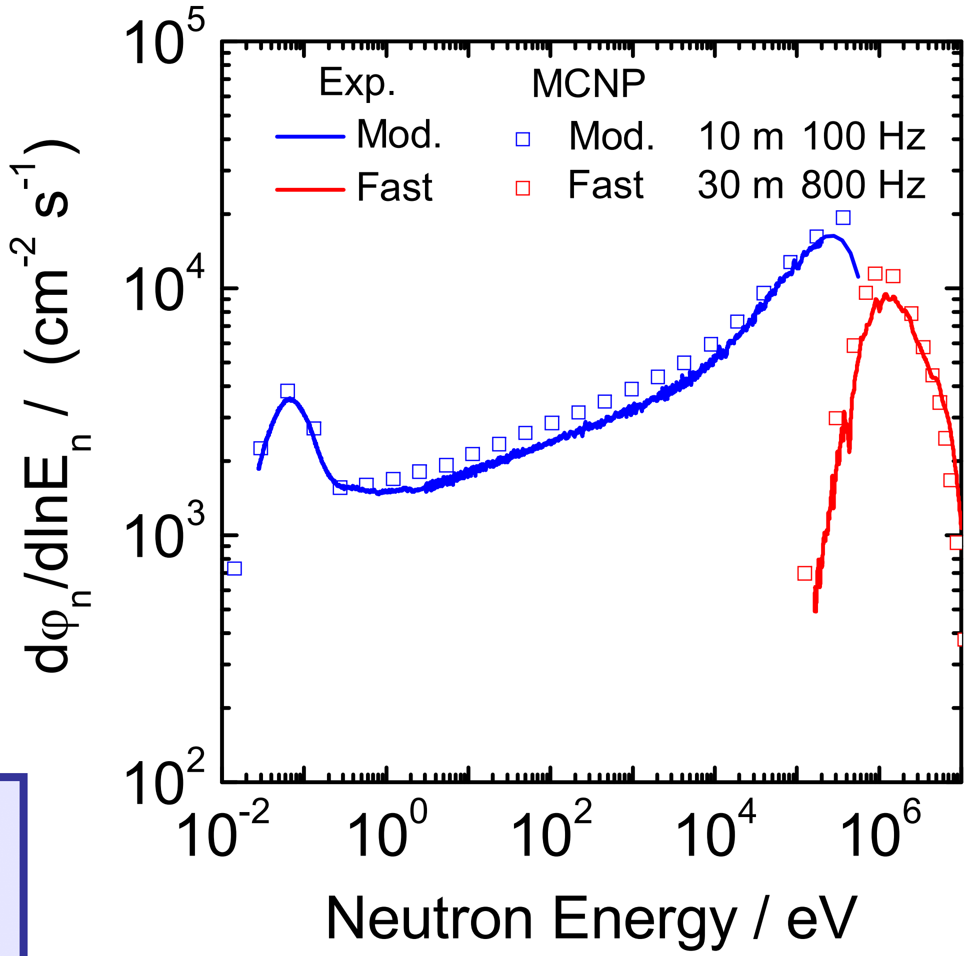
Uranium target (e$^-$ ⇨ γ ⇨ n)
rotating, mercury cooled
4 10^{10} neutrons / burst
Moderated or fast neutron spectrum
24 h/d, 100h/w, 12 parallel FPs

12 Flight paths, 8 to 400 m

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

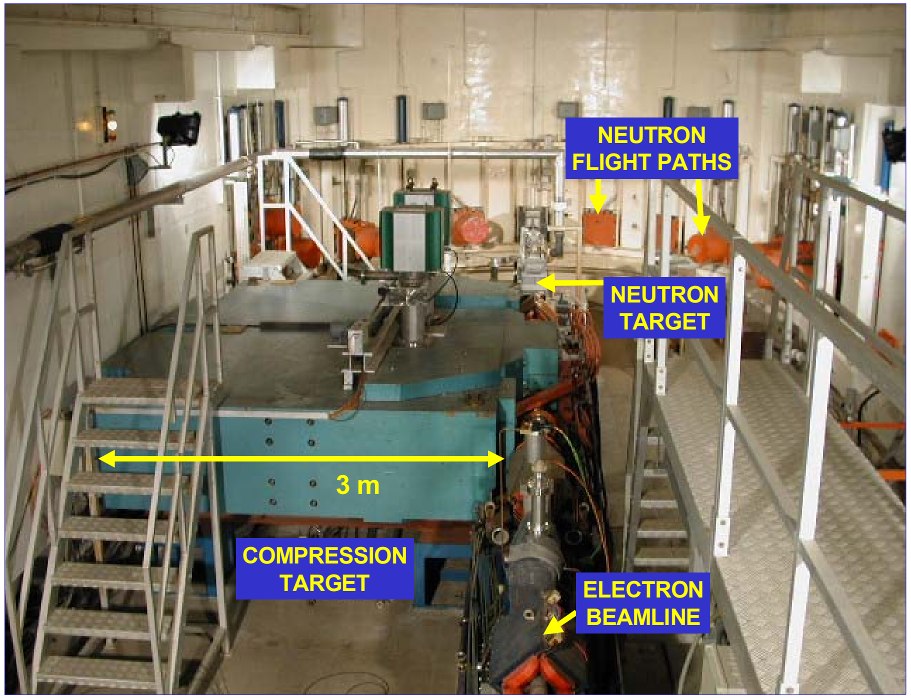
Neutron Production

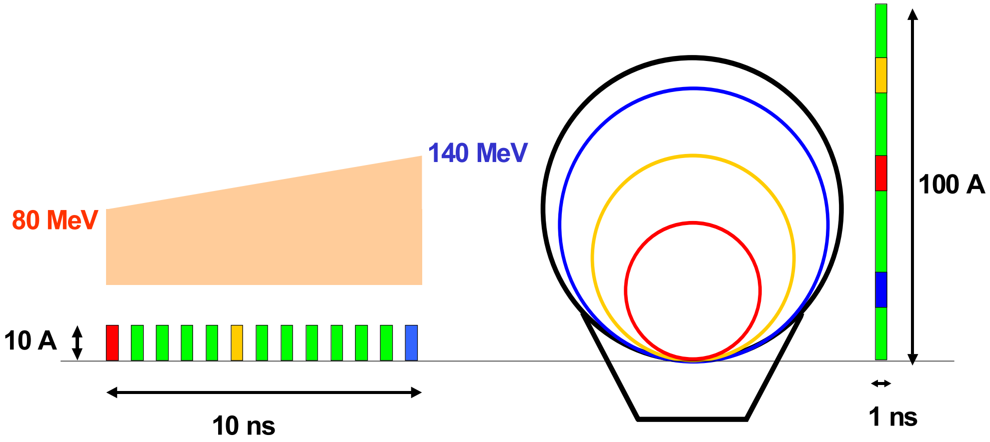




- (e^-, γ) Bremsstrahlung in U-target
- (γ , n) , (γ , f) in U-target
- Low energy neutrons by water moderator in Be-can

Compression Magnet





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

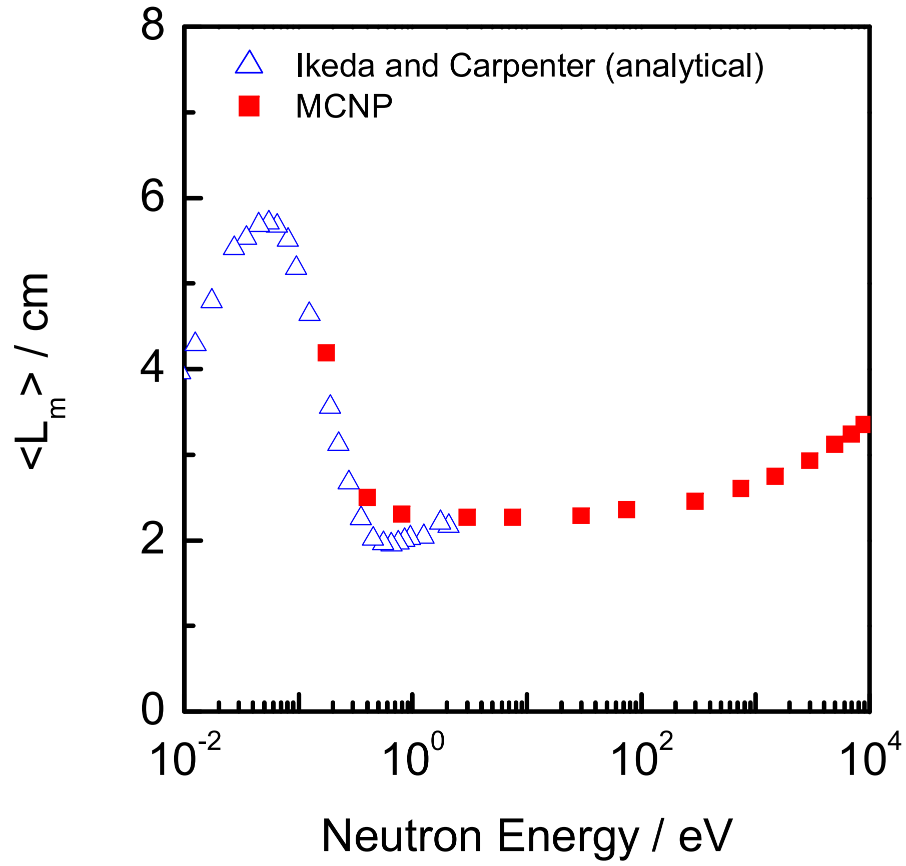
$$\Rightarrow \quad \rho = \frac{1}{B}\frac{E}{qc}$$

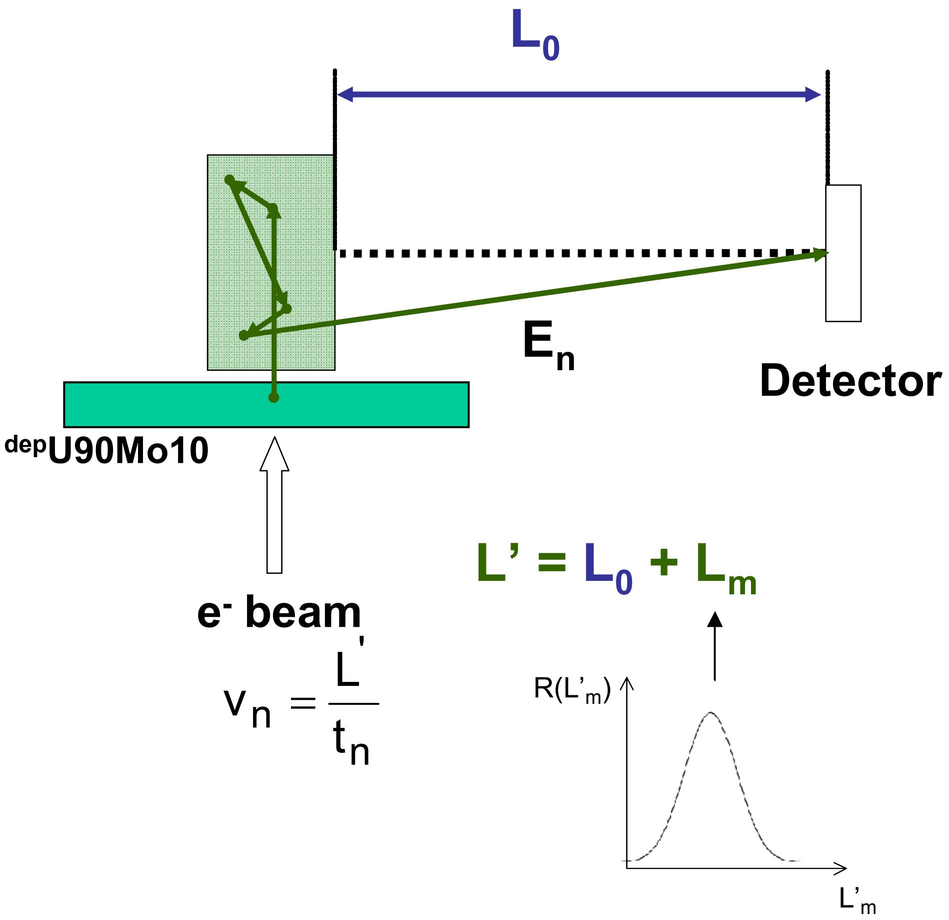
$$\Rightarrow \quad B = \frac{2\pi}{qc^2}\frac{\Delta E}{\Delta\tau}$$

ΔE = 60 MeV
$\Delta\tau$ = 10 ns

➔ compressed pulse length ~ 1 ns

From time-of-flight to energy: effective flight path



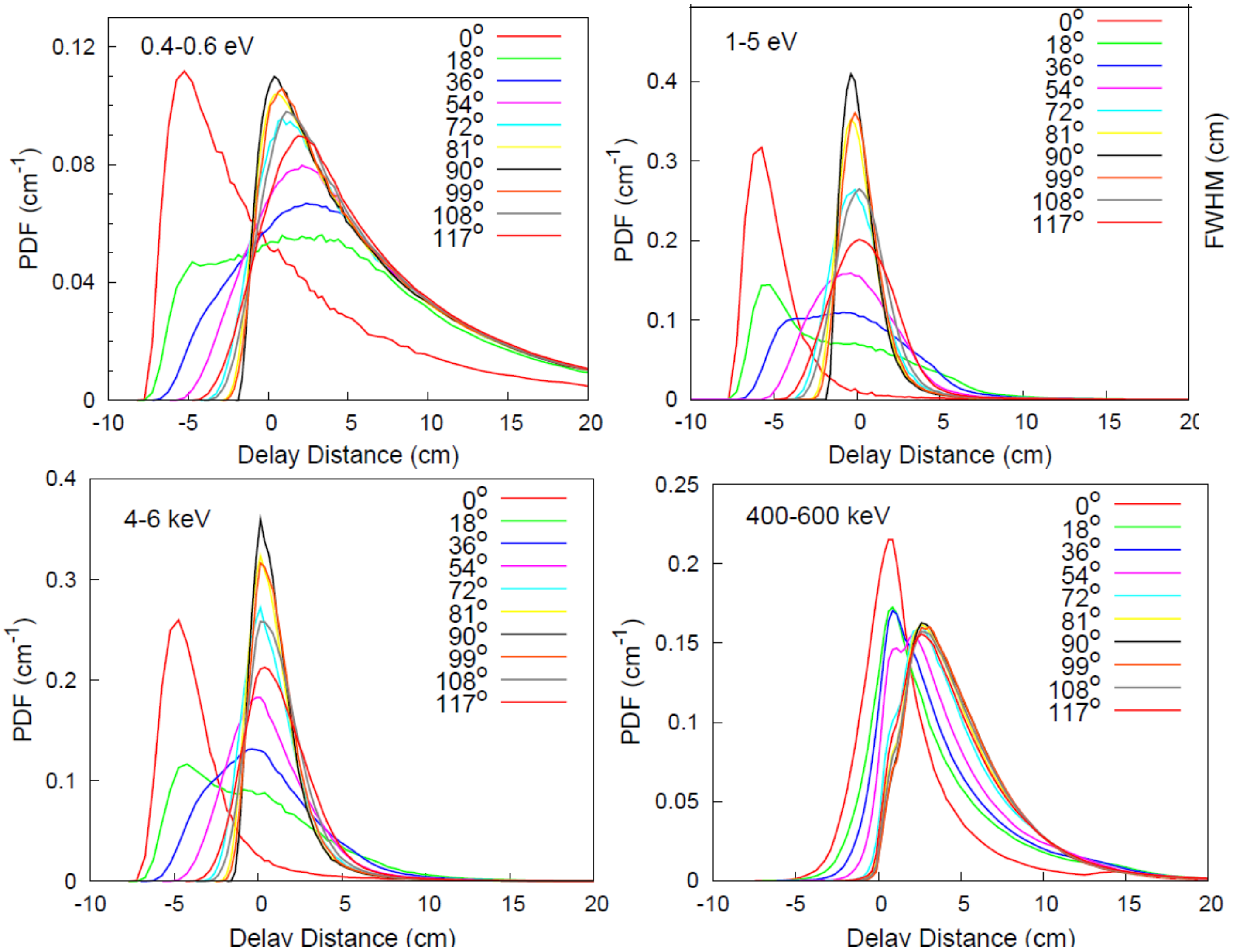


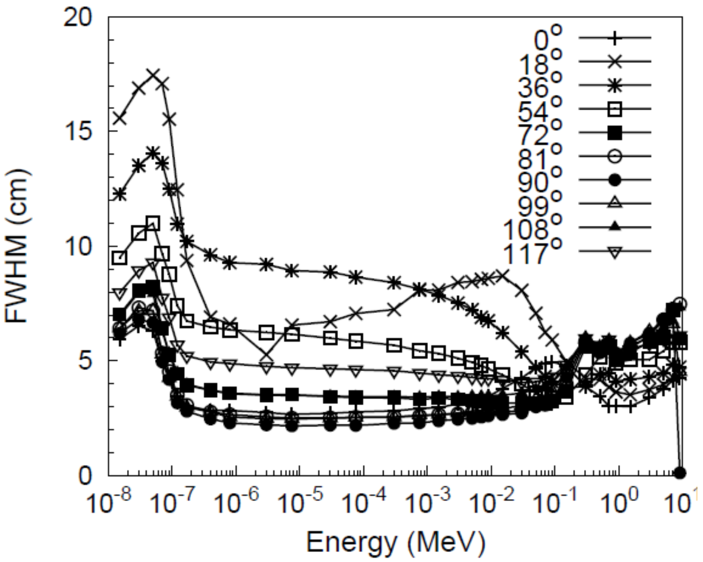
$$v_n = \frac{L'}{t_n}$$

$$L' = L_0 + L_m$$

Analytical expressions in REFIT include storage term of Ikeda & Carpenter

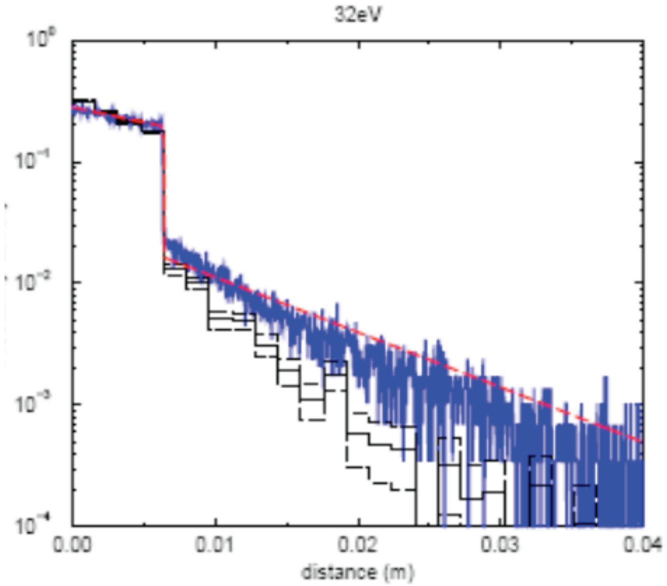
Moderator resolution function





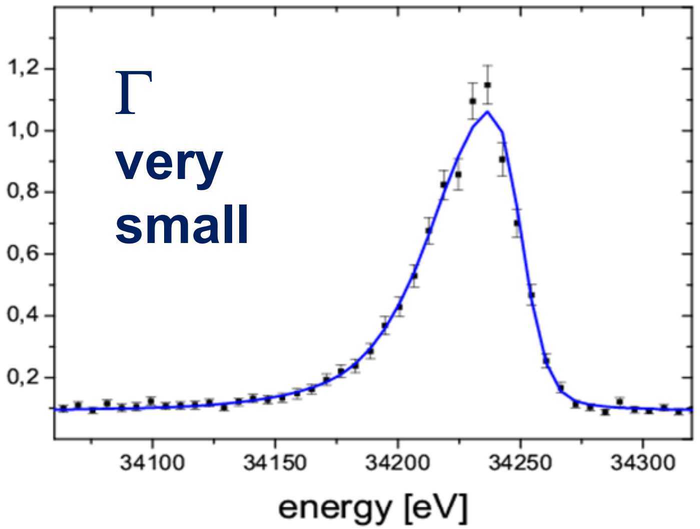
**Ene et al. 2010
Monte Carlo
simulations**

Resonance broadening

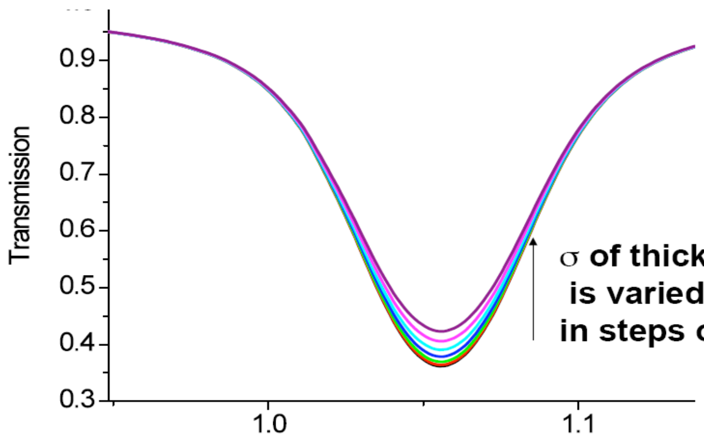


Detector resolution

Neutron source width



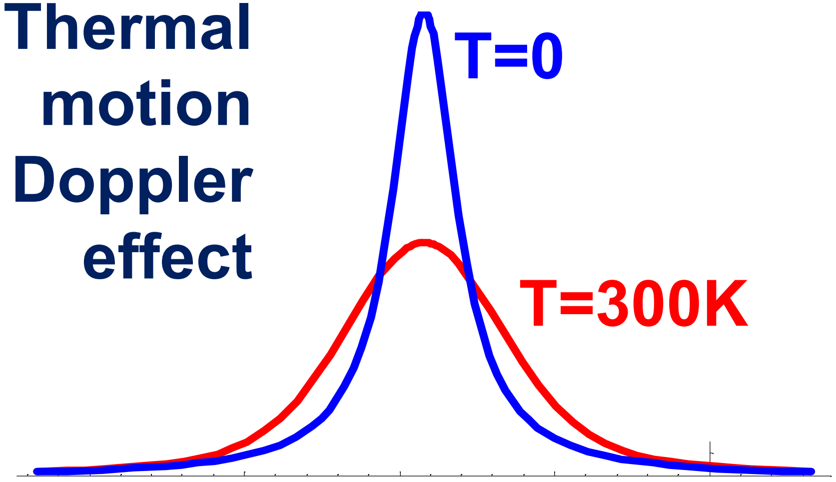
Γ very small



σ of thickness distribution is varied between 0 to 0.6 in steps of 0.1

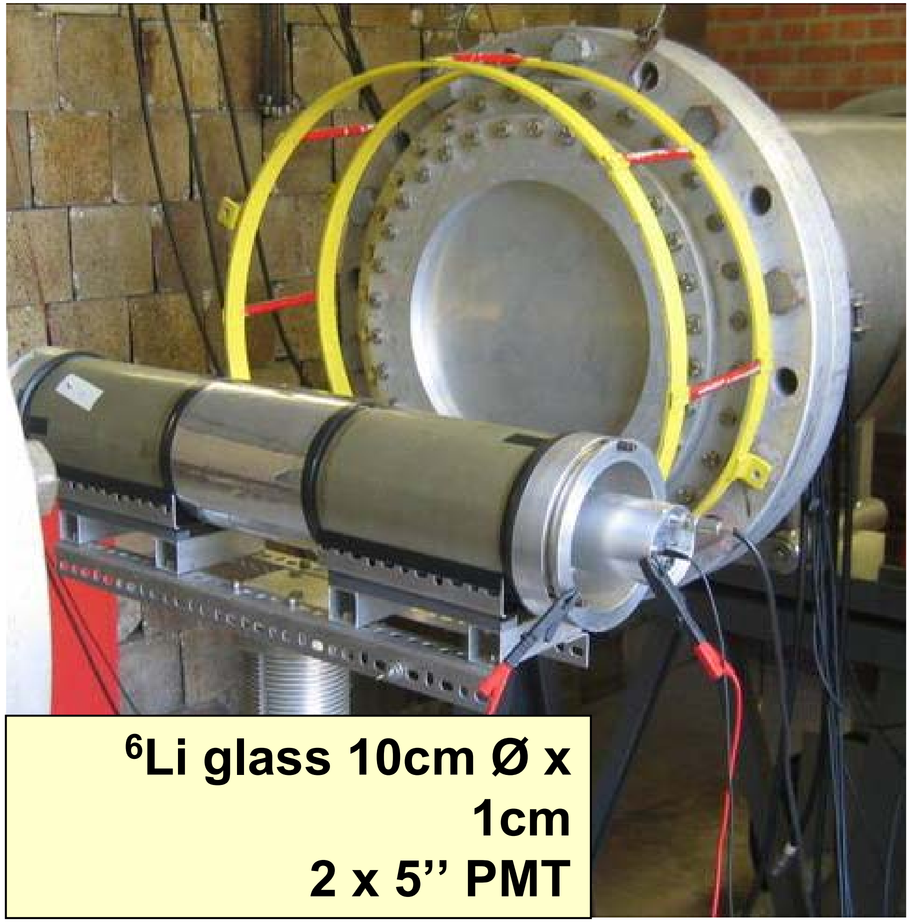
Sample thickness distribution

Thermal motion Doppler effect



Total cross section & transmission + time-of-flight

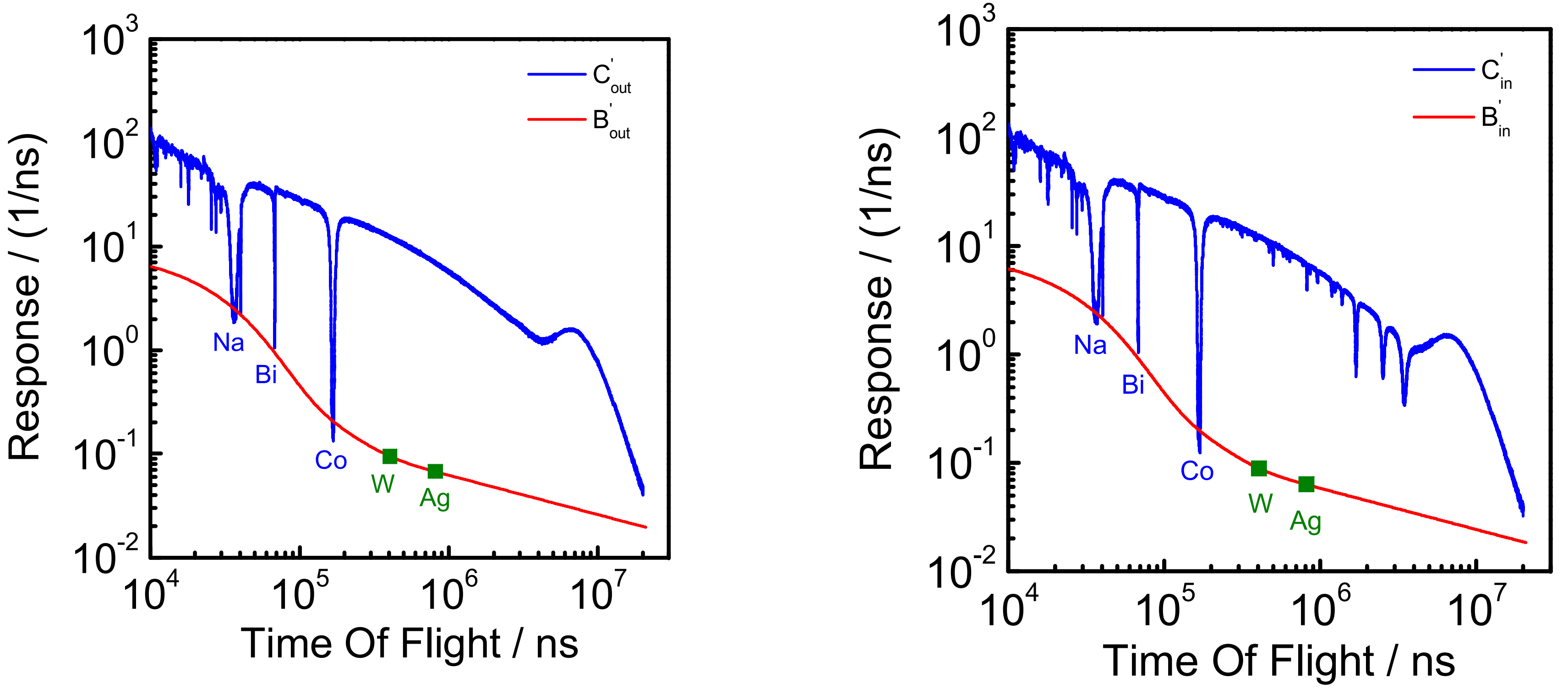
Transmission setup, sample holder and neutron detector



^{6}Li glass 10cm Ø x 1cm
2 x 5’’ PMT

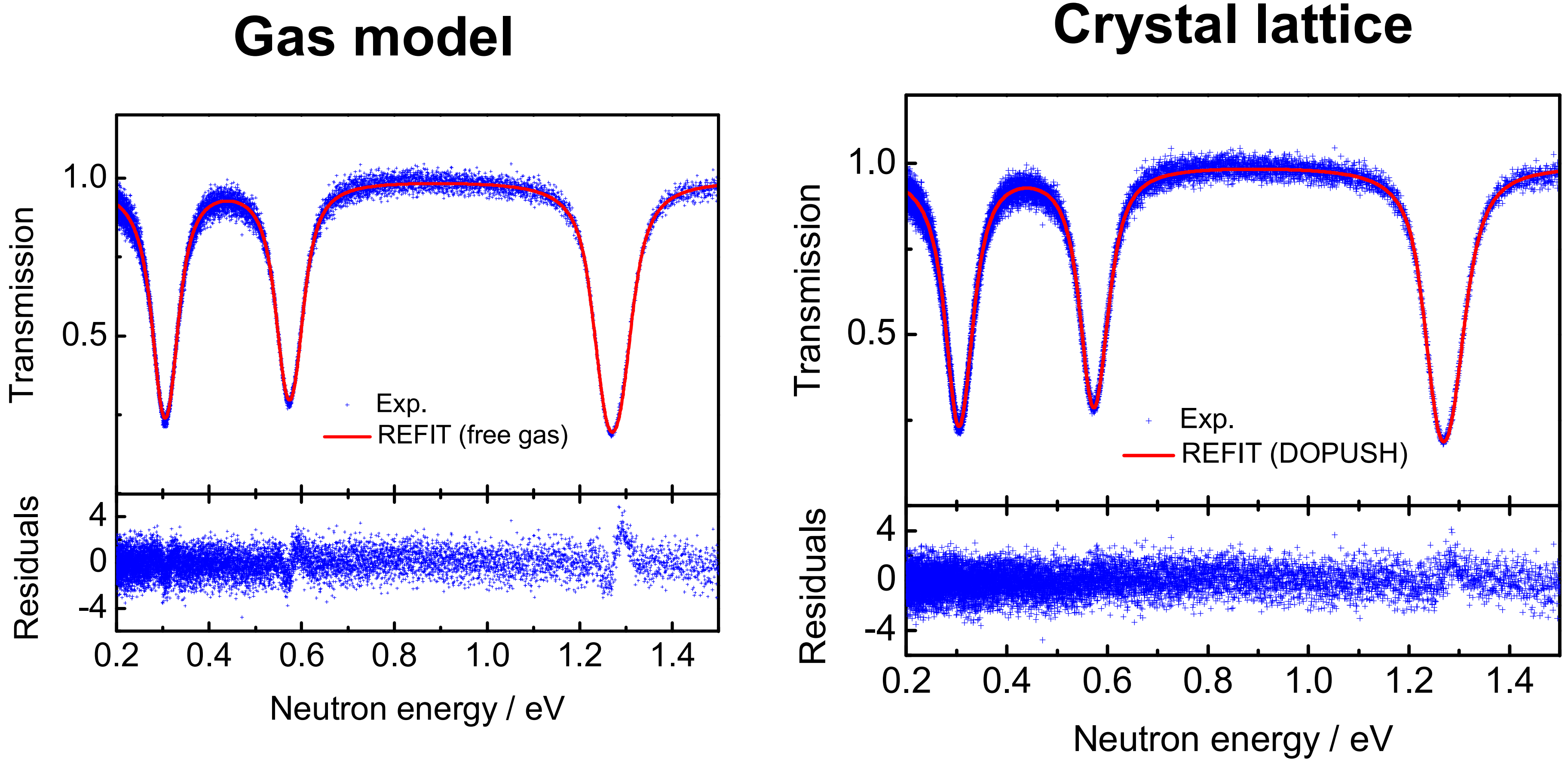
Sample changer

background filters
Fixed and variable: black resonance technique

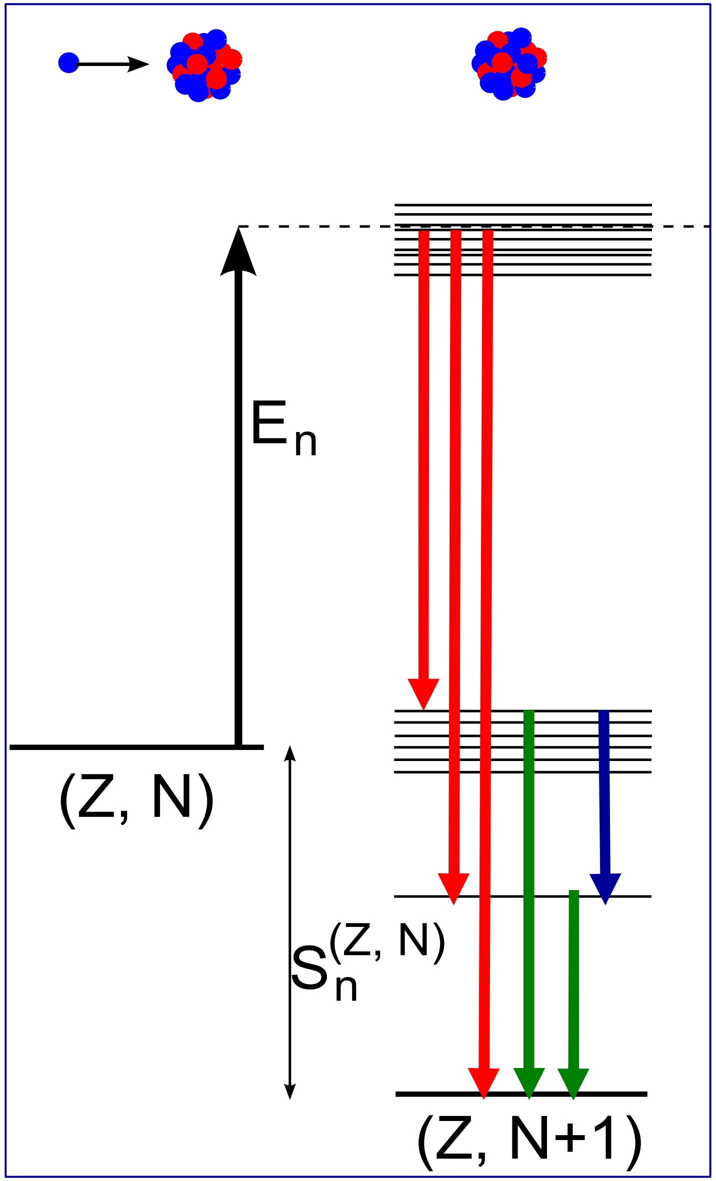


Transmission data and (REFIT, ^{241}Am)

Resonance analysis to obtain resonance parameters from which cross section may be reproduced under any required circumstances



Neutron capture and time-of-flight



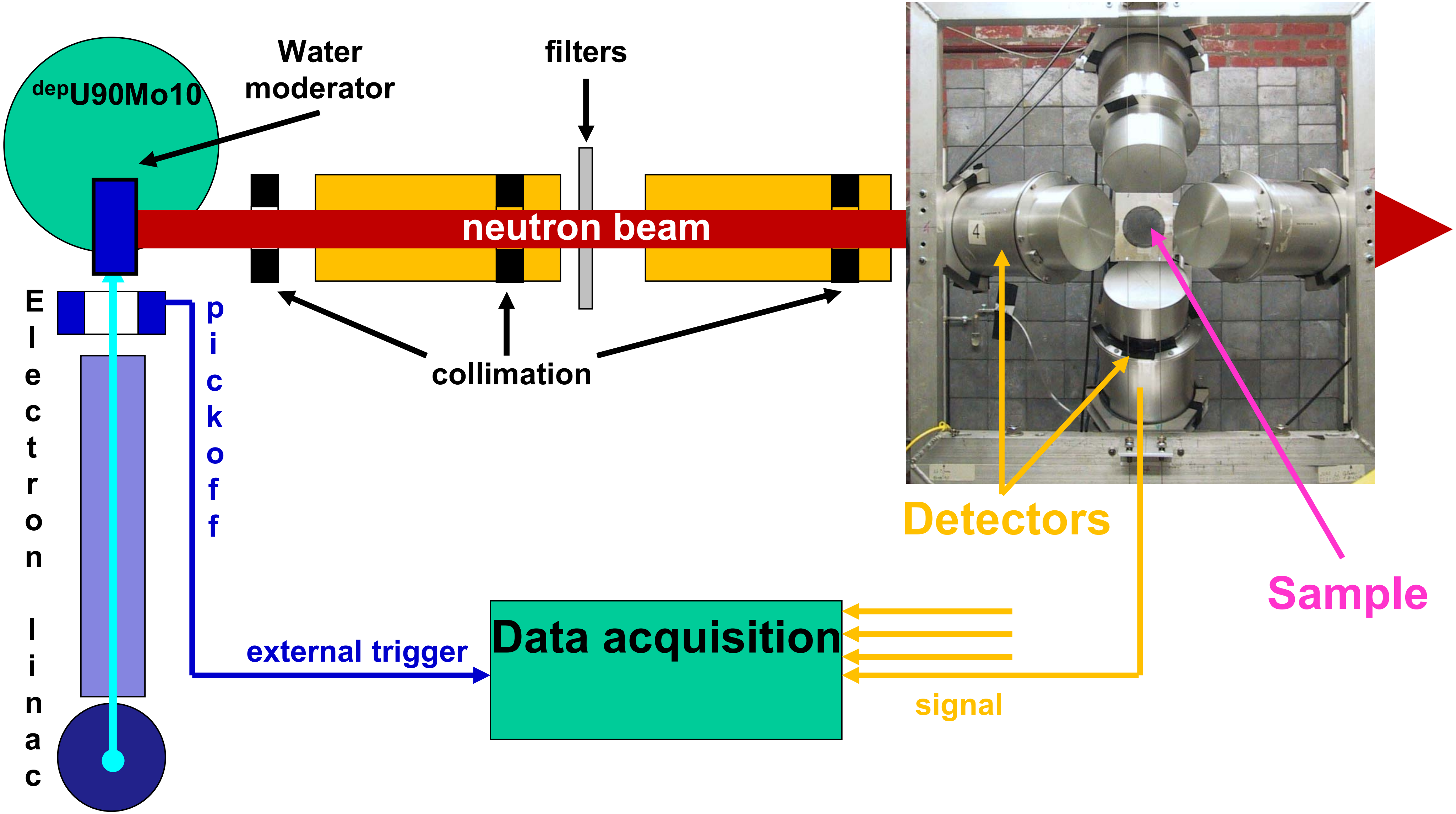
A neutron is absorbed
Nucleus decays: cascade of gamma-rays

Principle: detection of gamma-rays
Needed: gamma detectors, detection efficiency
Concerns: gamma-cascades vary with energy
Gamma-ray angular distribution

Normalization/fluence measurement
fluence distribution as function of energy
Reference cross section or black resonance

Otherwise the issues also shown for transmission (background, deadtime, resolution functions, etc).

Neutron capture and time-of-flight



^{dep}U90Mo10

Water moderator

filters

neutron beam

collimation

Electron linac

pickoff

external trigger

Data acquisition

signal

Detectors

Sample

Neutron capture and time-of-flight

Total energy principle

C_6D_6 liquid scintillators

125°

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

Flux measurements (IC)

$^{10}B(n,\alpha)$

$^{235}U(n,f)$

These are standards with well known energy dependence and cross sections

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

Borella et al., NIMA 577(2007) 626

Modify response with a weighing function so detection efficiency is proportional to the gamma-energy

Then detection efficiency depends on the total excitation energy (not on the details of the cascade)

Efficiency is independent of E_n

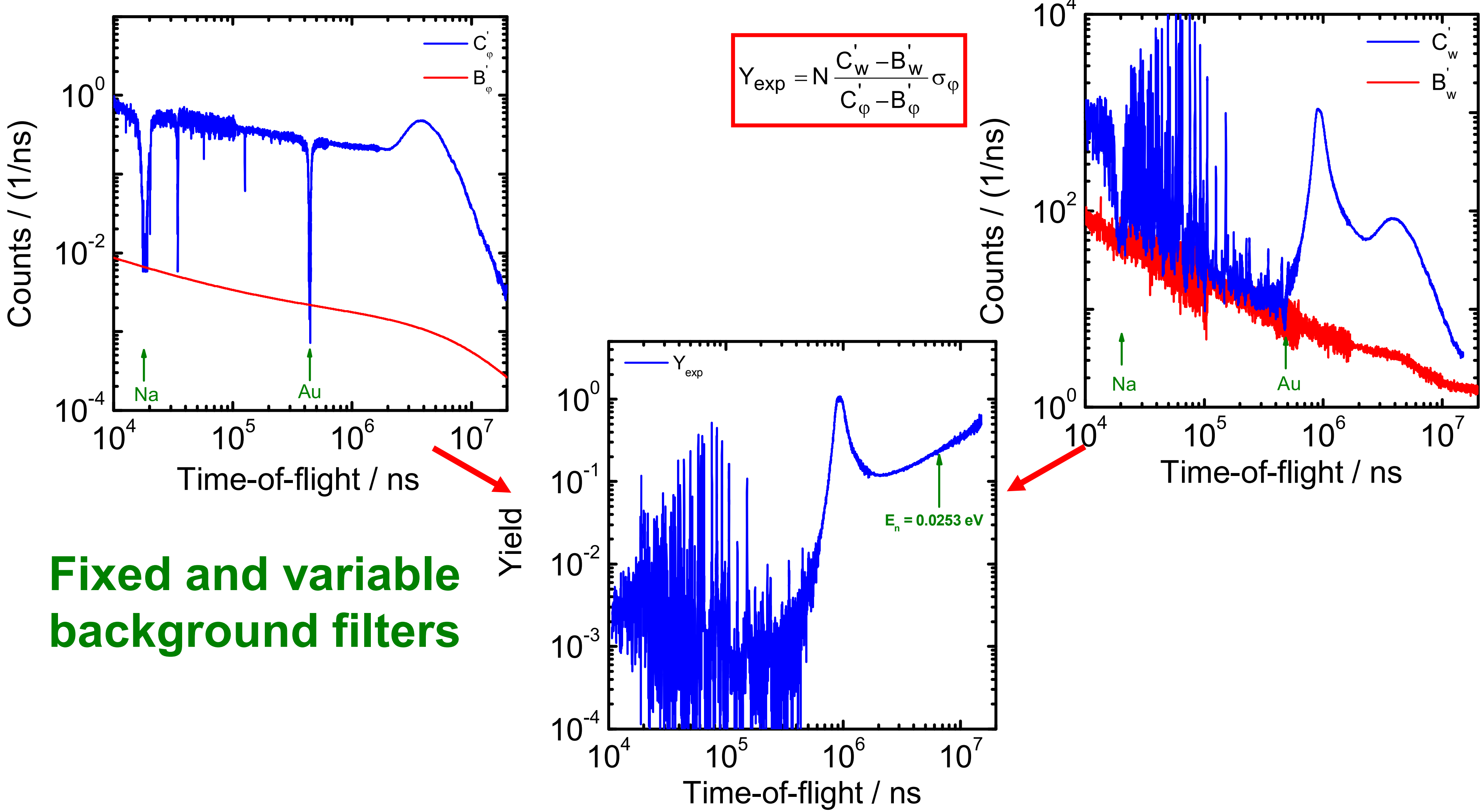
For dipole transitions: possible gamma-angular distribution.

No impact at 125 degrees!

WF : from MC simulations

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

Capture measurements: e.g. ^{103}Rh(n,γ)



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

Fixed and variable background filters

Normalization of capture data

- **C_6D_6 liquid scintillators** : $C'_w - B'_w$
- **Flux measurements (IC)** : $C'_\varphi - B'_\varphi$
  - **$^{10}B(n,\alpha)$ < 150 keV**
  - **$^{235}U(n,f)$ > 150 keV**

$$Y_{exp} = \frac{C'_w - B'_w}{C'_\varphi - B'_\varphi} \boxed{\frac{\varepsilon_\varphi}{\varepsilon_r}\frac{\Omega_\varphi}{\Omega_r}\frac{F_\varphi}{F_r}\frac{A_\varphi}{A_r}} \sigma_\varphi$$

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

Normalization constant N

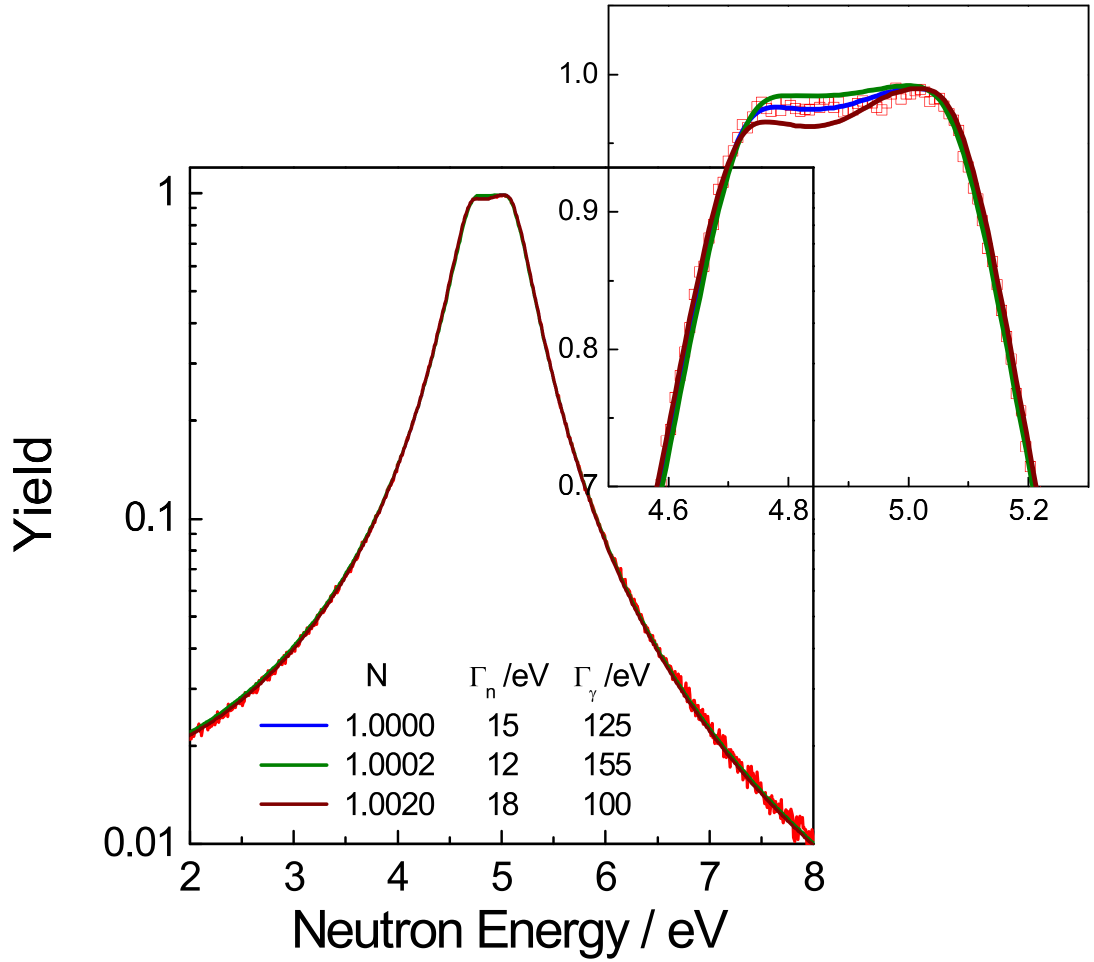
- **Saturated resonance**
  - **^{197}Au : 4.9 eV**
  - **^{109}Ag : 5.2 eV**
  - **....**
- **Resonance with : $\Gamma_n << \Gamma_\gamma$**
  - **Γ_n from transmisson**
  - **^{56}Fe : 1.15 keV**

Internal normalization:

⇒Reduction of systematic effects

Borella et al., NIM A, 577 (2007) 626

Normalization: saturated resonance



$$n\sigma_{tot} \gg 1 \text{ and } \sigma_\gamma \approx \sigma_{tot}$$

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

$$Y_\gamma \cong 1$$

$$\Rightarrow N \cong \frac{C'_\varphi - B'_\varphi}{C'_w - B'_w}\,\frac{1}{\sigma_\varphi}$$

N is independent of :

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

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

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

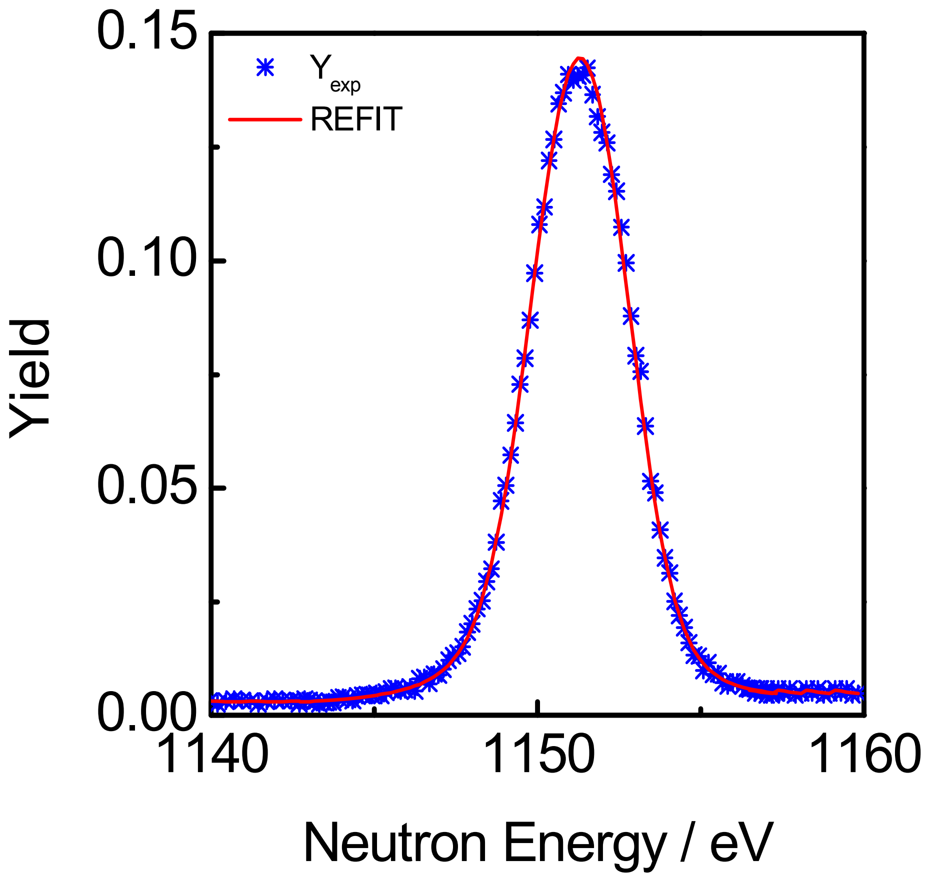
Normalization to 1.15 keV resonance of ^{56}Fe(n,γ)

ORELA Γ_γ = 574 meV

Transmission
Perey et al. : Γ_n = 61.7 $\pm$0.9 meV

Capture (thin + thick sample)
Macklin : Γ_n = 61.8 $\pm$ 1.9 meV

Use the well known iron resonance at 1.15 keV for which Γ_n is very well established.
Iron alloyed or sandwiched with the sample of interest to minimize differences in detection efficiency

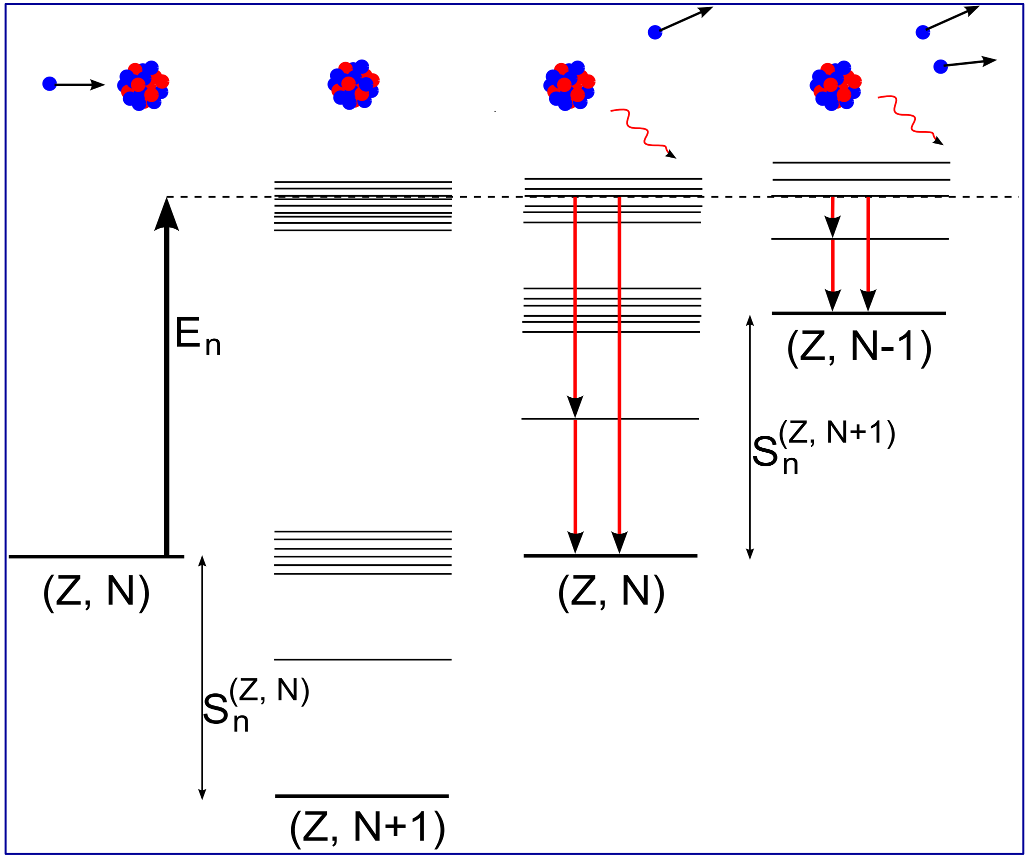


⇒ Uncertainties of 2% can be reached

Measurement of the associated gamma-rays

Very selective (indirect)

Access to angle-integrated cross section



Primary experimental result:

Gamma production cross section

Databases:

Level scheme

Level cross section

Total inelastic cross section

Method

Germanium detectors 8 x 8 cm Ø by 8 cm long, Δt~9.7 ns

GELINA, time-of-flight, 200 m flight path, $\Delta E_n/E_n$=1 keV @ 1 MeV, direct flux configuration, 800 Hz, 550 n/cm2/s

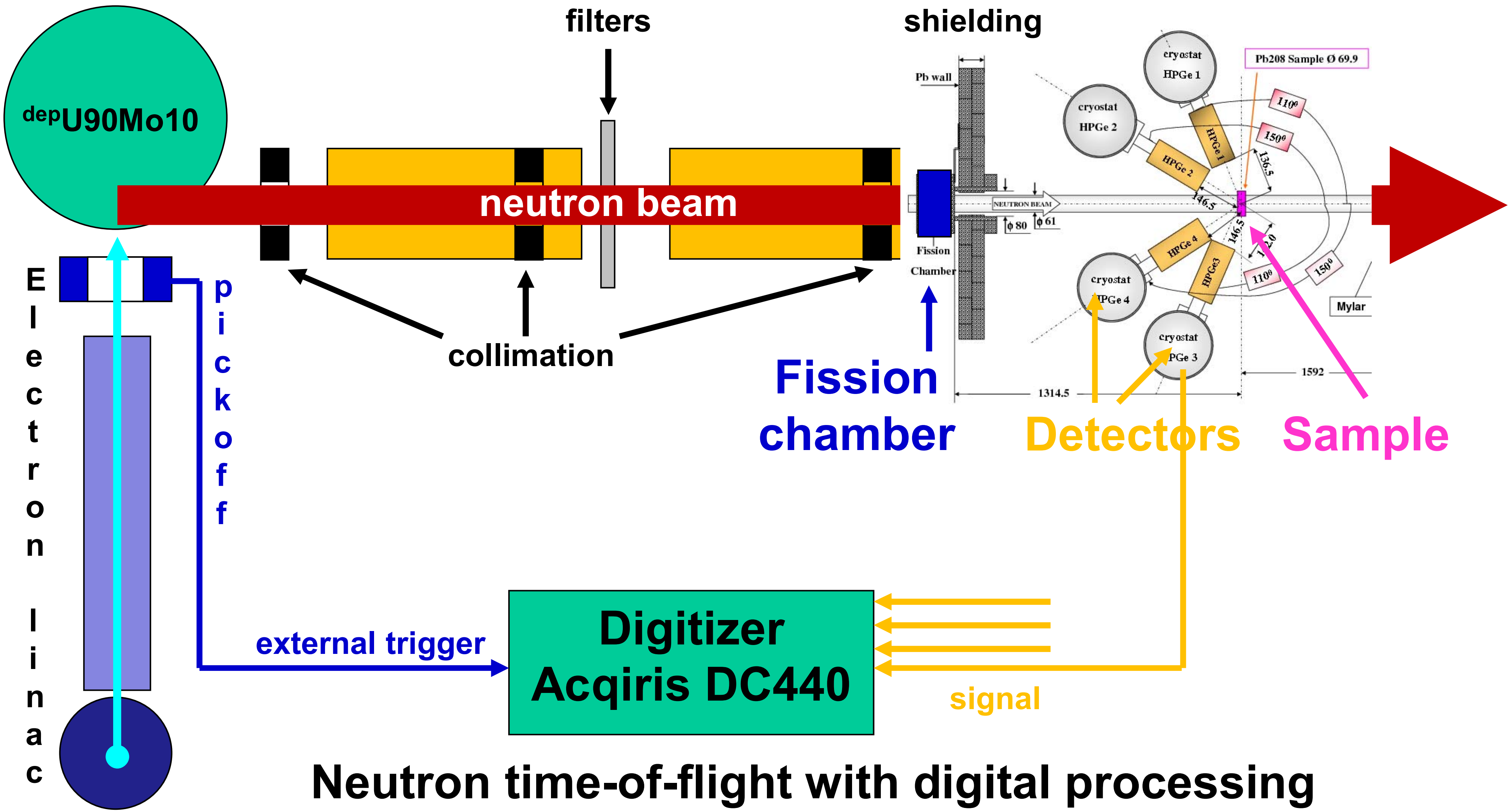
Measurement normalized to ^{235}U(n,F)

Compensation for angular distribution by Gaussian quadrature using weighted sum of $d\sigma/d\Omega$ at 110 and 150 degrees

Elemental sample produced at IRMM

Aiming for $\Delta\sigma/\sigma$ = 5% below 5 MeV

Method



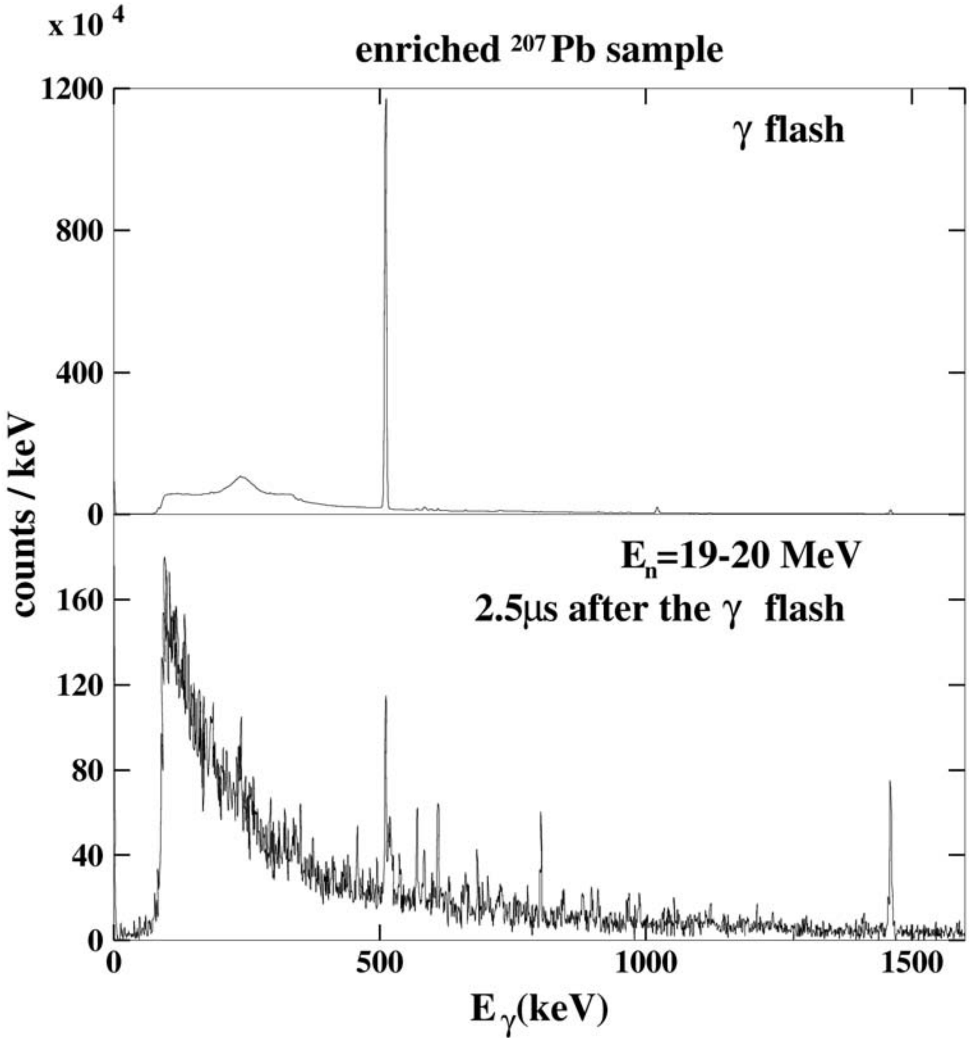
Neutron time-of-flight with digital processing

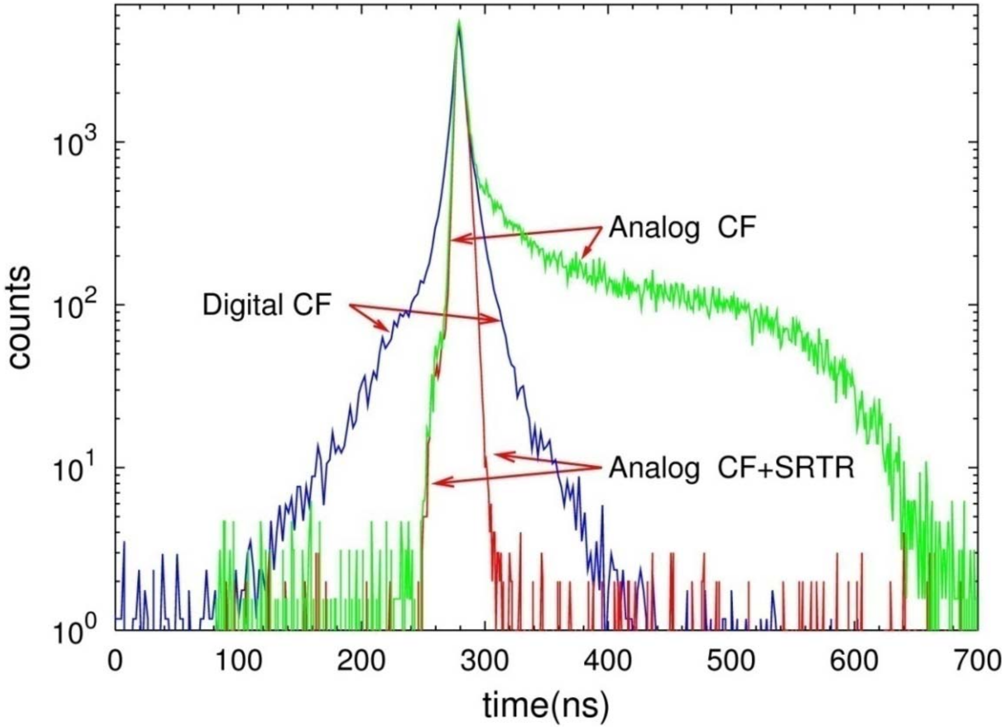
Setup

GAINS @ FP3/200m

GELINA

Use of digitizers w. HPGe



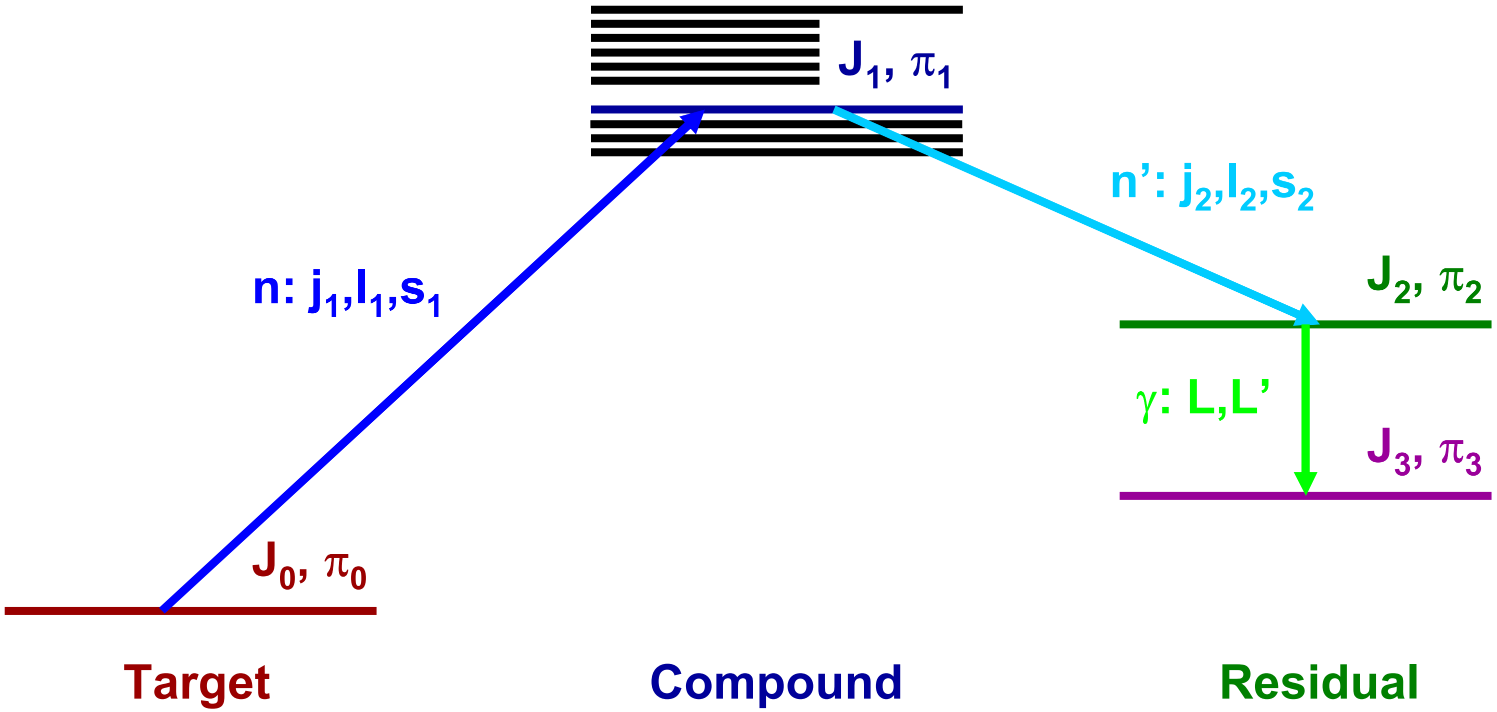


Dead time reduction (< 2.5 μs)

Time response improvement

Time range: 22 μs

Inelastic angular distributions



$$\frac{d\sigma}{d\Omega_\gamma}(\theta) = \frac{1}{8k^2}\sum_{j_1,j_2}\sum_{\nu=0,even}^{\nu_{max}} N'C'W'M(\delta)\tau P_\nu(\cos\theta_\gamma)$$

$$0 \le \nu \le 2j_1, 2J_1, 2J_2, 2L'$$

E. Sheldon and D.M. van Patter,
Rev.Mod.Phys. 38(1966)143

Angular distribution of gammas

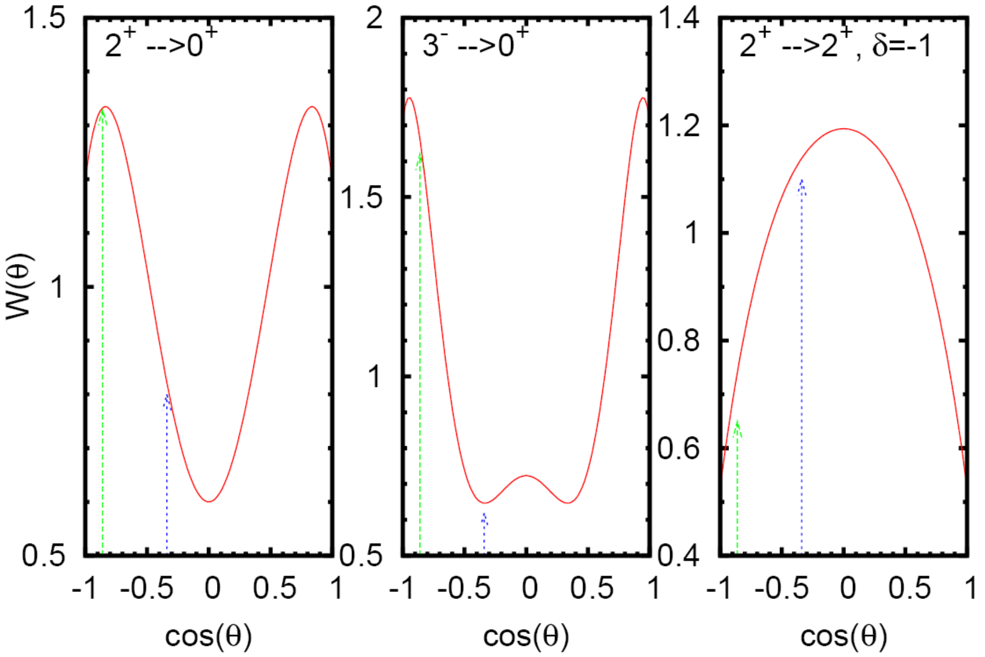
Exact angle integration

$1 \leq L \leq 3$

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{\sigma}{4\pi} W(\theta) = \frac{\sigma}{4\pi} \sum_{k=0}^{k_{\max}} c_{2k} P_{2k}(\cos(\theta))$$

$$\sigma = 2\pi \int_{-1}^{1} \frac{d\sigma}{d\Omega}(x)dx = 2\pi \sum_{i=1}^{2} w_i \frac{d\sigma}{d\Omega}(x_i)$$

$$x_i = \cos(\theta_i)$$



θ_1=110°, 70° → w_1=1.30429
θ_2=150°, 30° → w_2=1.69571

Sample TP-NP 08/10

Metrological information Na disc Ø80 x 4mm (**Can N°2**)

Total mass:	19,44± 0.04 g
Ø	79,80 ± 0.08 mm
Thickness:	4.23 ± 0.08 mm
Area:	50,01 cm^2
Mass/area	0,389 g/cm^2
Density	0,92 g/cm^3 (Theoretical density: 0.97 g/cm^3)

Metallic sodium sample prepared by André Moens at IRMM

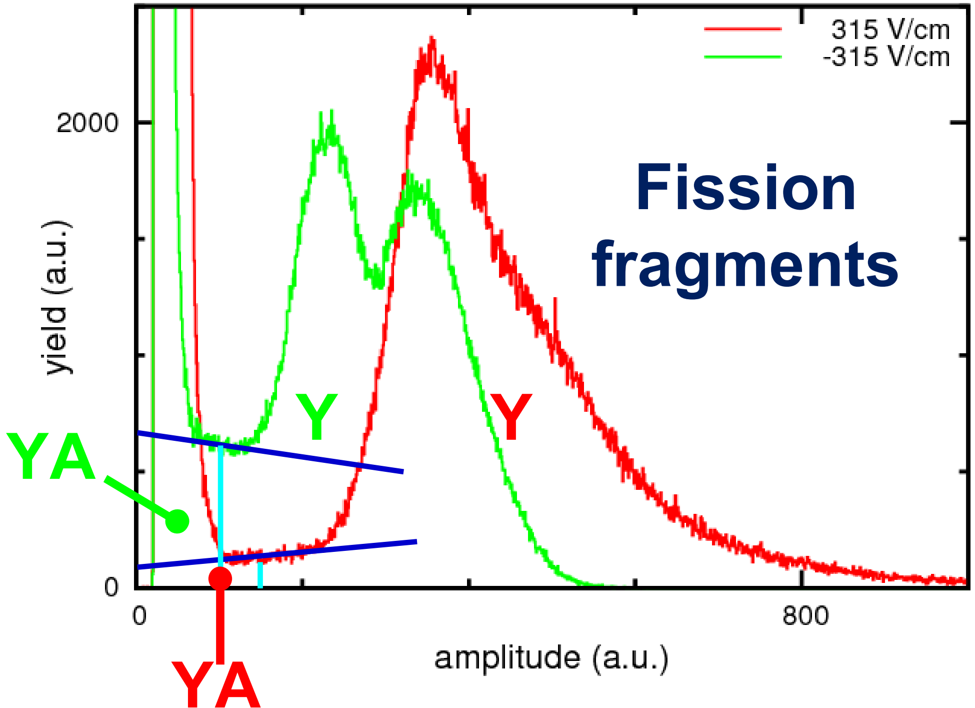
Preparation method

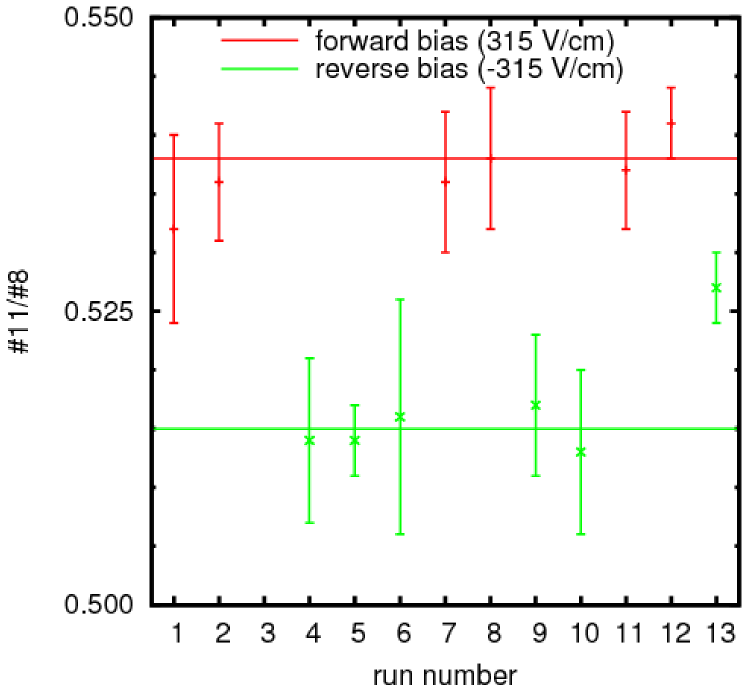
Cutting, Pressing, Mechanical Rolling (sandwich-method) and Punching.
Note: All mechanical transformations done under petrol.
Determination dimensions + weighing under inertgas atmosphere.
Canning Na-disc in Al-can under Ar-atmosphere. Closure of the cans was performed by a gluing method using "UHU plus".

Chemical analyses

Alfa Aesar, Lot D11S201 – Sodium ingot, 99.8% (metals basis)

Normalization to fission





Efficiency = Y / (Y+YA+YB)

Y+YA forward versus backward bias

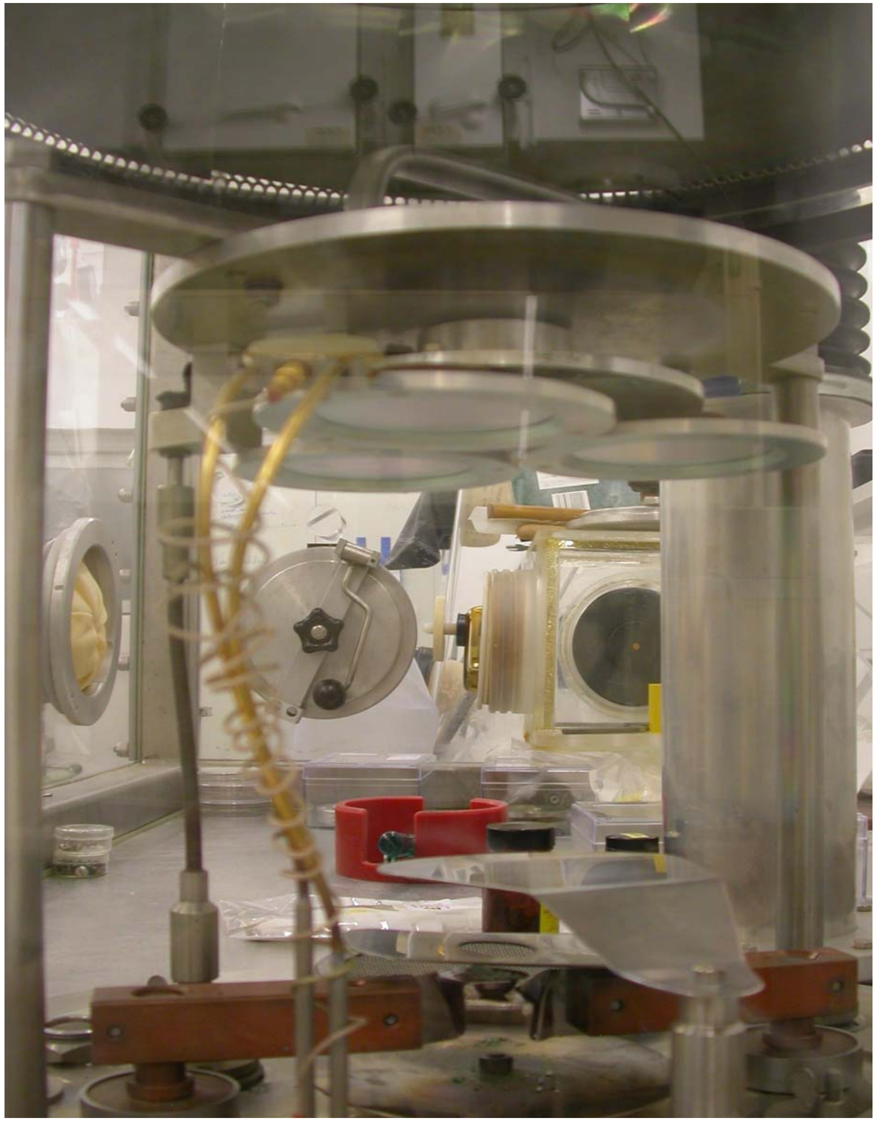
4.4% effect for 0.475 mg/cm^2 UF$_4$ evaporated

Y: yield above threshold, YA: yield below threshold (linear extrapolation)

YB: fragments stopped in the deposit (not shown) YB/(Y+YA+YB) = 0.105(7) t/(mg/cm2)

YB: measured by Budtz-Jörgensen Nucl.Instrum.Meth. 236(1985)630

Normalization: sample preparation



^{235}U

Fission deposit

Evaporated UF_4

Alpha-counting

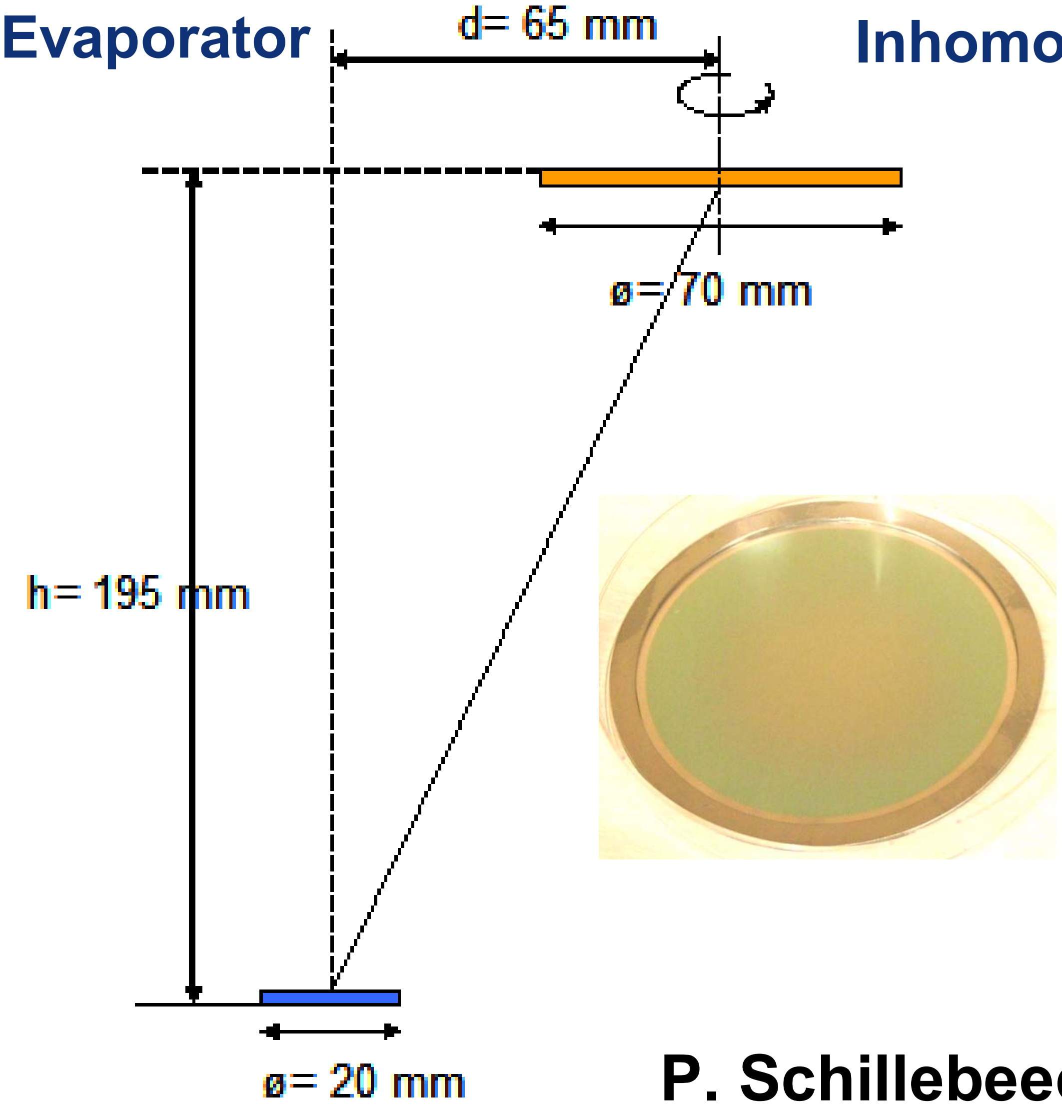
High purity

Roger Eykens
Andre Moens
Marc Peeters
Anna Stolarz

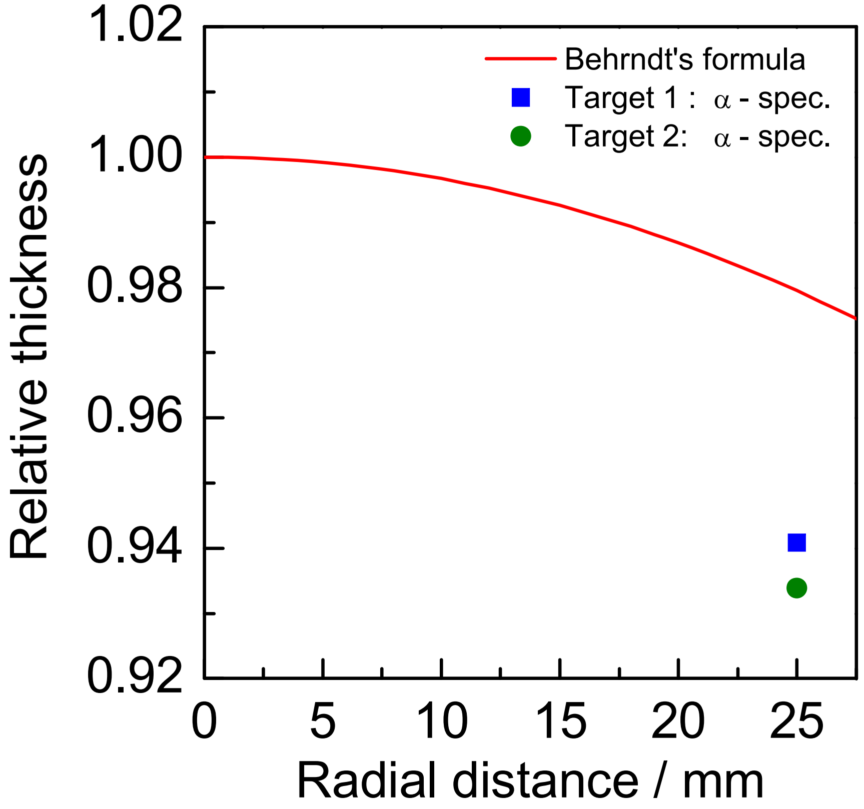
Peter Schillebeeckx

Sample preparation and characterization

Evaporator



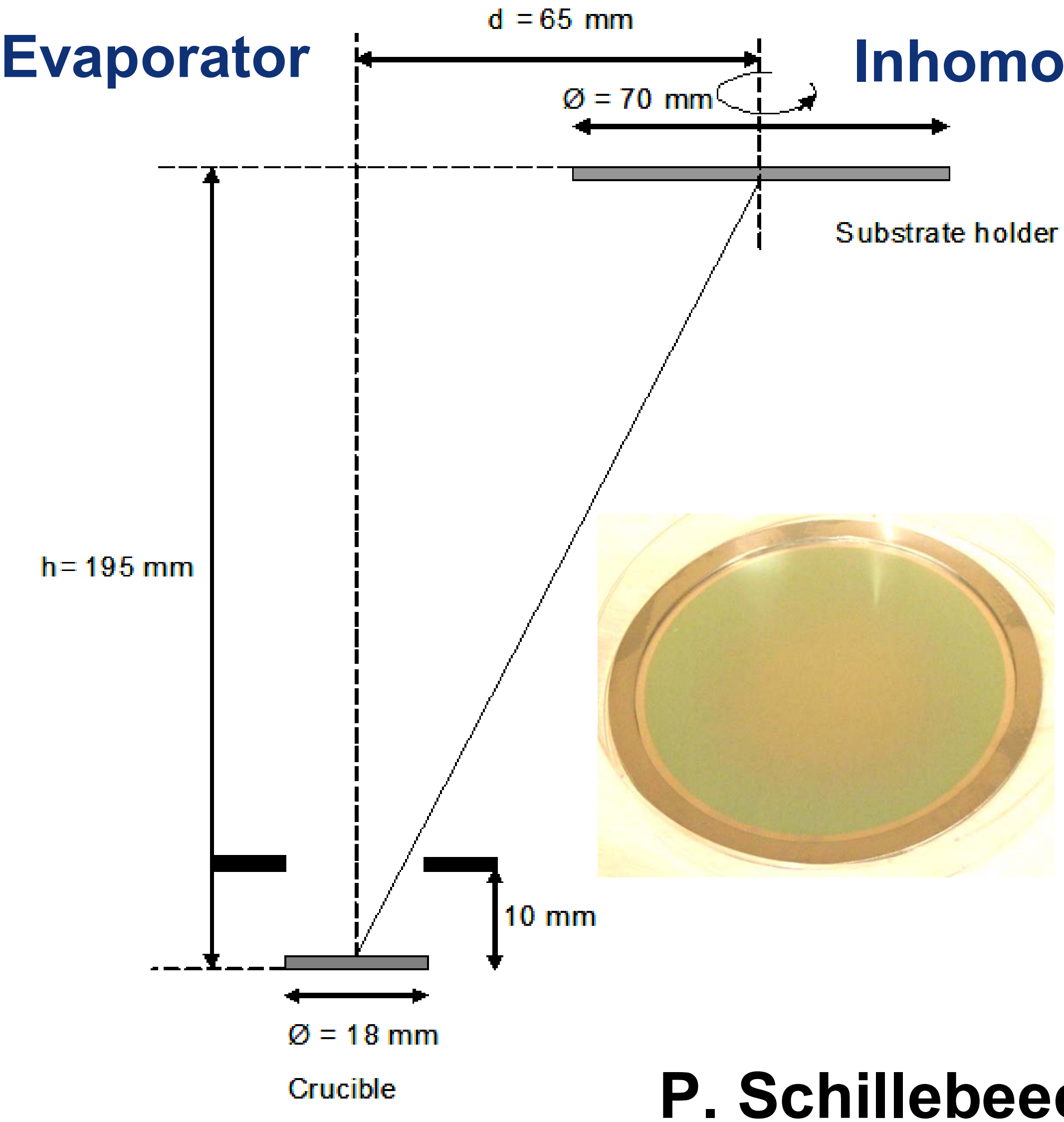
Inhomogeneity: +2% areal mass density 61 mm out of 70 mm



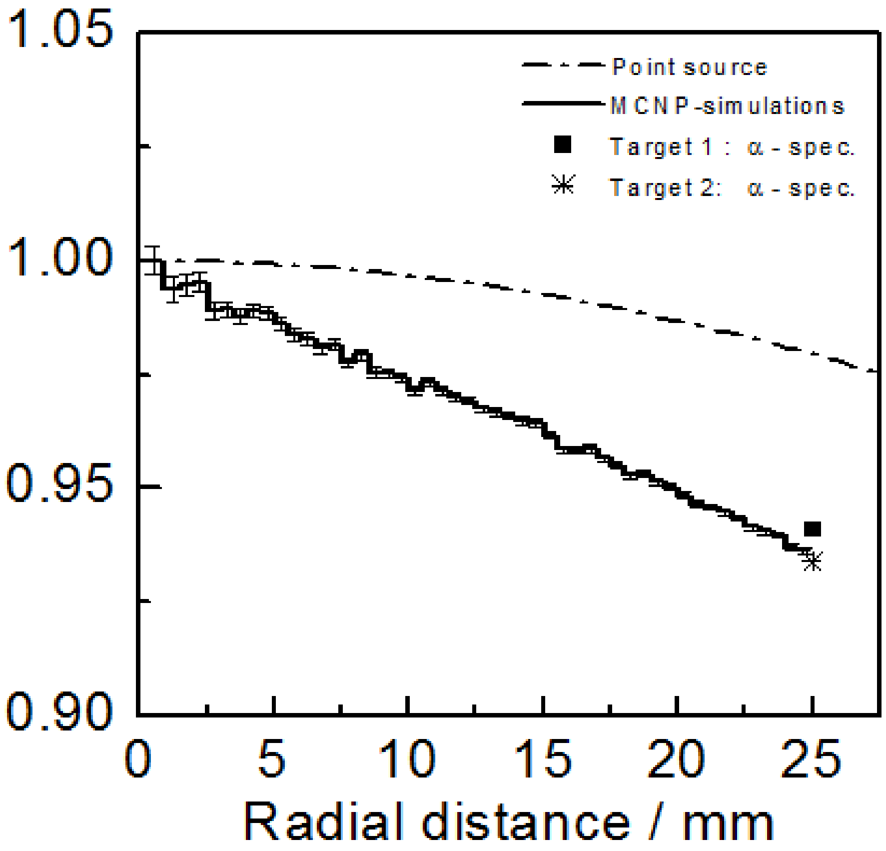
P. Schillebeeckx et al., NIM A 613(2010)378

Sample preparation and characterization

Evaporator



Inhomogeneity: +2% areal mass density
61 mm out of 70 mm



P. Schillebeeckx et al., NIM A 613(2010)378

Sample production and characterization

Activity to atoms
Mass spectrometry
65% activity: U-234

Callet& De Bièvre 15 November 1985	Atom%	Acc (2s)
U-234	0.0626	0.0025
U-235	99.8266	0.0044
U-236	0.0365	0.0027
U-238	0.0739	0.0025

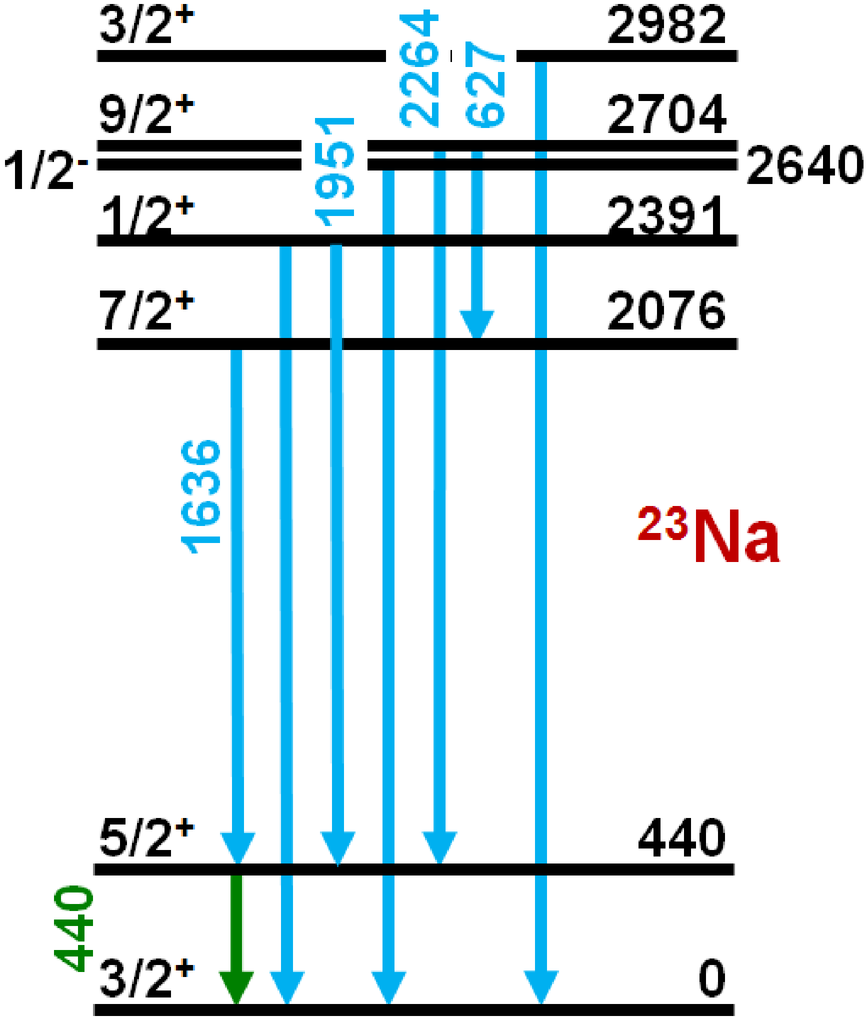
Quantity	Value (1s)
Decay constant	8.78(10) 10^{-17} U/s

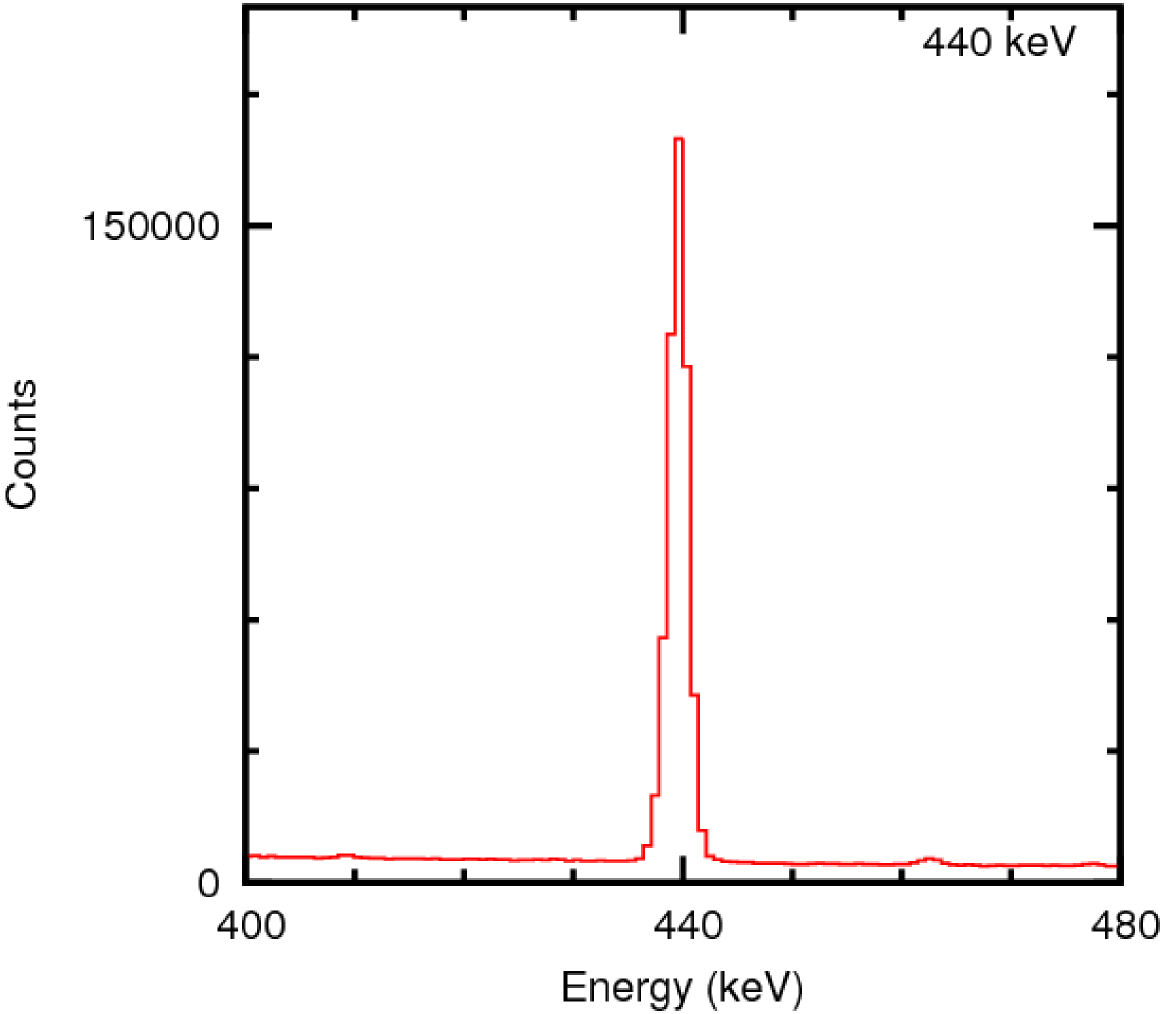
Richter 23 March 2009	Atom%	Acc (2s)
U-234	0.06389	0.00014
U-235	99.82275	0.00020
U-236	0.03768	0.00007
U-238	0.07568	0.00013

Quantity	Value (1s)
Decay constant	8.88(2) 10^{-17} U/s

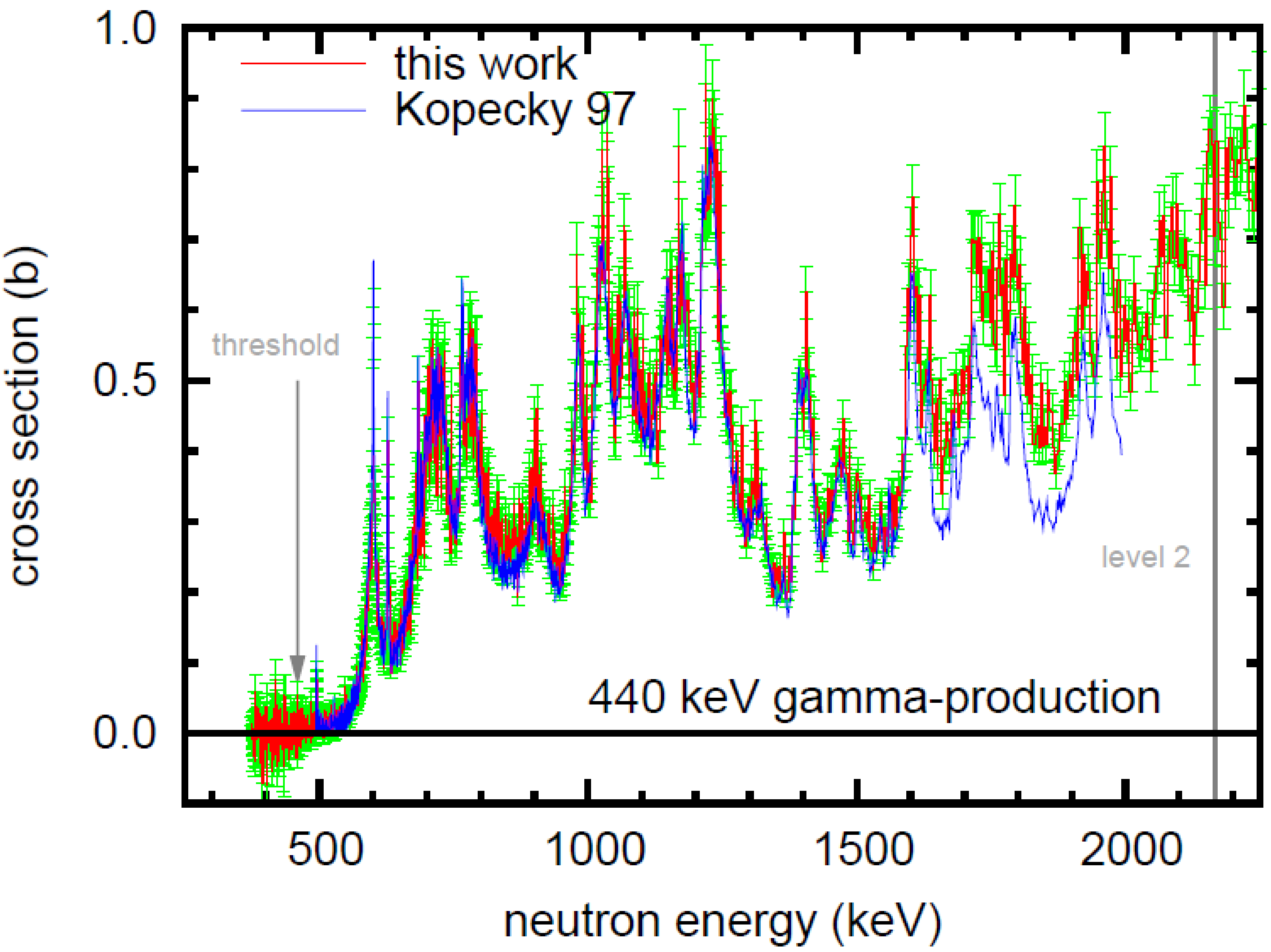
More accurate new value: +1% and 0.3% uncertainty

Example ^{23}Na

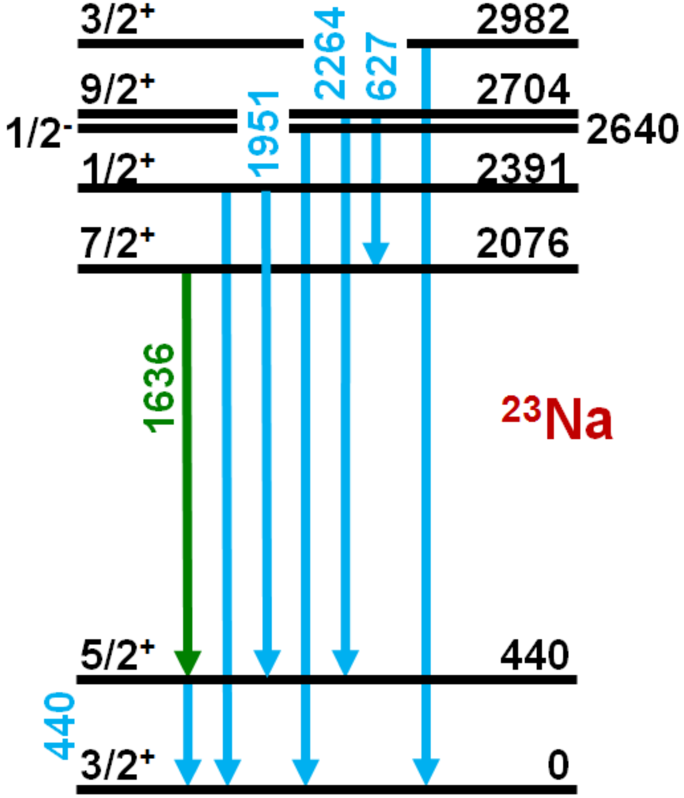


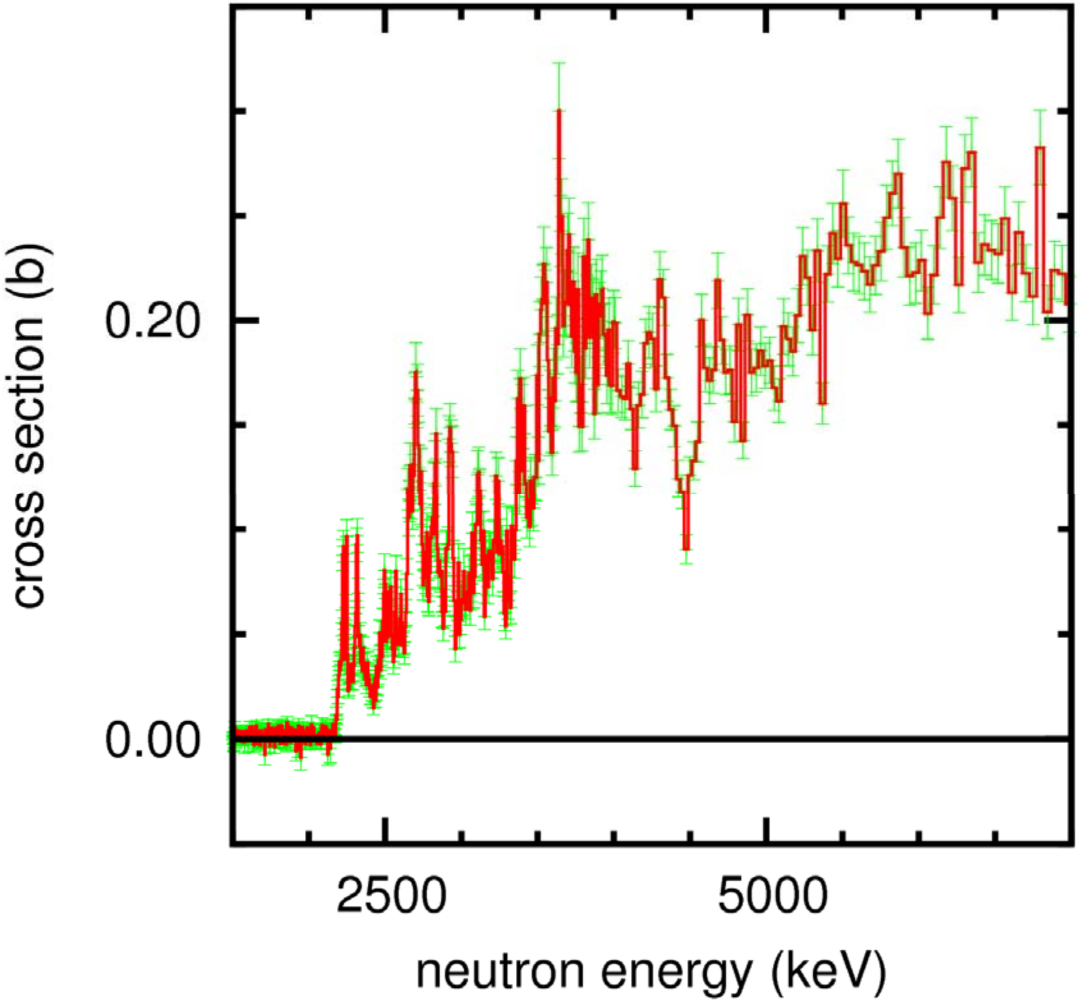


Example ^{23}Na, 440 keV



Example ^{23}Na, 1636 keV





Activation cross section measurements

Neutron source

7MV Van de Graaff accelerator

Binary reactions for quasi mono-energetic neutrons $^{7}Li(p,n)^{7}Be$, $^{3}H(p,n)^{3}He$, $^{3}H(d,n)^{4}He$ reaction

High intensity compared with time-of-flight, but only one energy per measurement

Solid-state Ti/T

Samples

Both natural and enriched

Example $^{241}Am(n,2n)^{240}Am$

Neutron energy and flux monitoring

The neutron fluence rate was determined by the $^{27}Al(n,\alpha)^{24}Na$ ENDF/B-VI standard cross section

The neutron flux density distribution were determined by the spectral index method

$^{115}In(n,n')^{115m}In$, $^{58}Ni(n,p)^{58}Co$, $^{27}Al(n,p)^{27}Mg$, $^{27}Al(n,\alpha)^{24}Na$, $^{56}Fe(n,p)^{56}Mn$, and $^{93}Nb(n,2n)^{92m}Nb$

distinct energy thresholds
time-of-flight spectrum measurements.

7 MV Van de Graaff accelerator

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

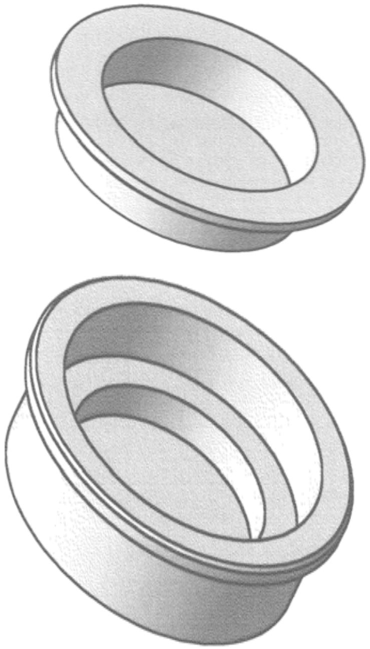
Fission
Activation measurements
light charged particles
Flux (BIPM)
Calibration of detectors

Irradiation setup
L3 beamline with Ti/T target
Light weight sample holder

Sample preparation

9 samples

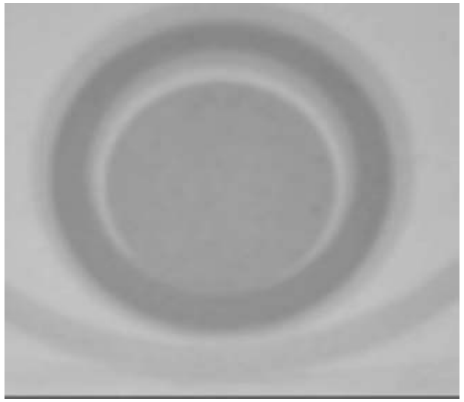
- **32.2 to 42.2 mg ^{241}Am (AmO$_2$)**
- **0.3 to 0.4 g Al$_2$O$_3$ matrix**
- **Ø =12.2 mm, height=1.6 to 2.1 mm**
- **5 GBq/piece, 10 mSv/h contact**



Infiltration technique (JRC-ITU, Karlsruhe) :

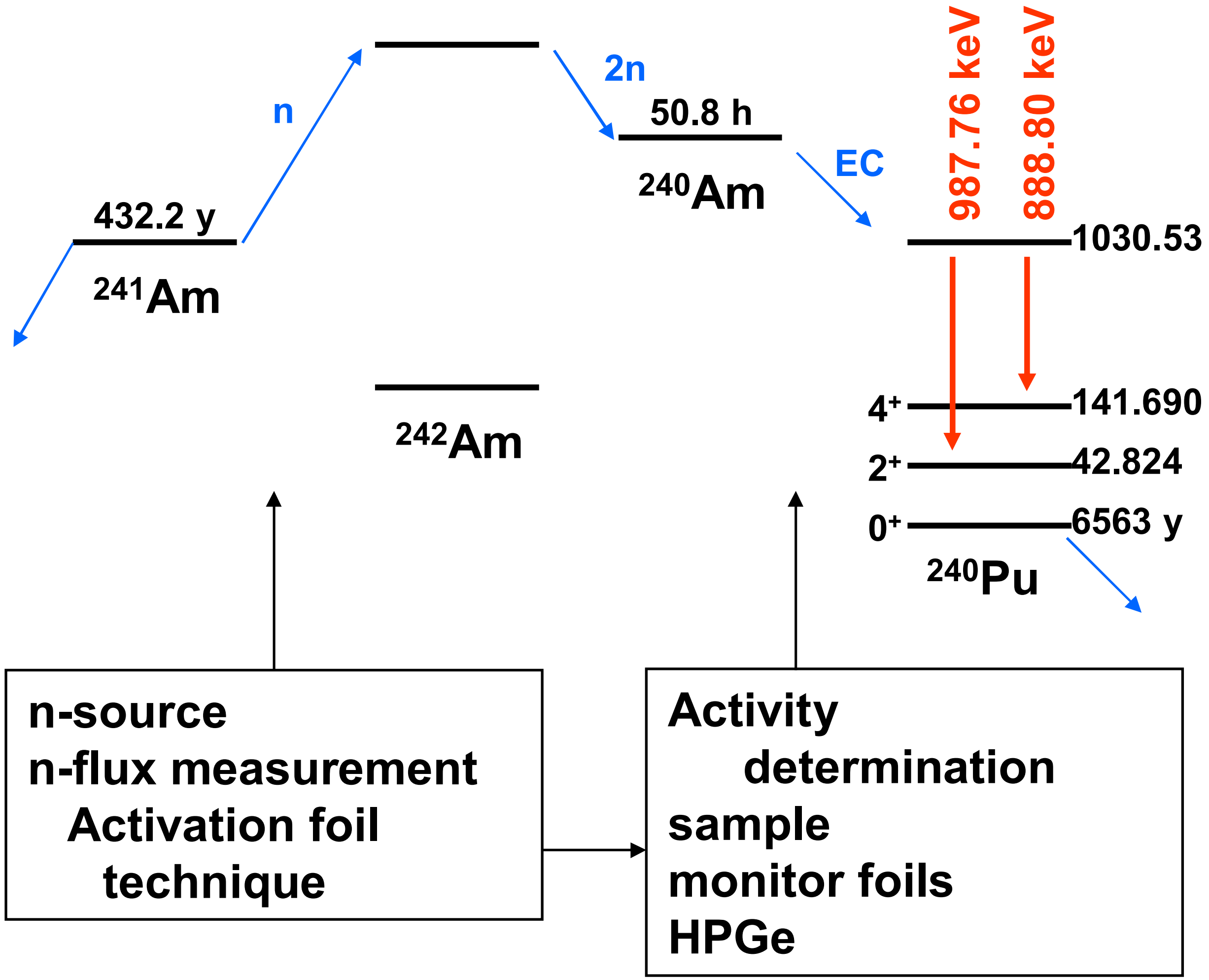
- **Porous alumina granules made by powder metallurgy**
- **Am infiltration with a nitrate solution**
- **Drying/calcination**

Nästren, Holzhäuser, Fernandez, Brossard, Wastin, Ottmar, Somers



Experimental procedure

Schematic (n,2n) process and level scheme



n-source
n-flux measurement
Activation foil technique

Activity determination
sample
monitor foils
HPGe

Relevant gamma rays

Nucleus	Half life	E_γ	I_γ
		(keV)	(%)
^{239}Am	11.9(1) h	278	15.0(17)
		228	11.3(13)
^{240}Am	50.8(3) h	**988**	**73(4)**
		889	**25.1(13)**
^{241}Am	432.1(7) y	60	35.9(5)
		233	4.6(3) 10^{-6}
		276	6.6(4) 10^{-6}
		278	4.4 10^{-7}
		887	**2.2(5) 10^{-7}**
		922	1.9(4) 10^{-7}

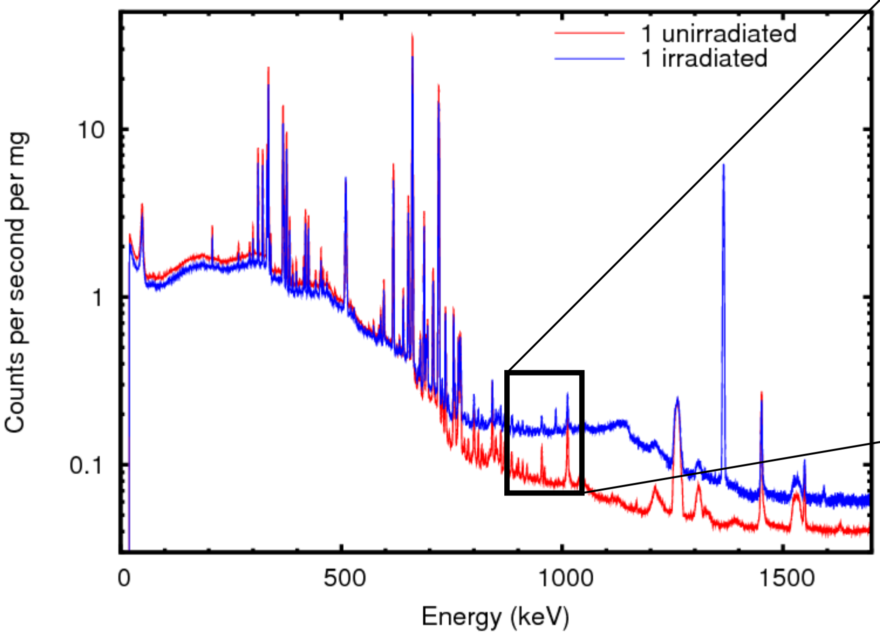
Activity determination

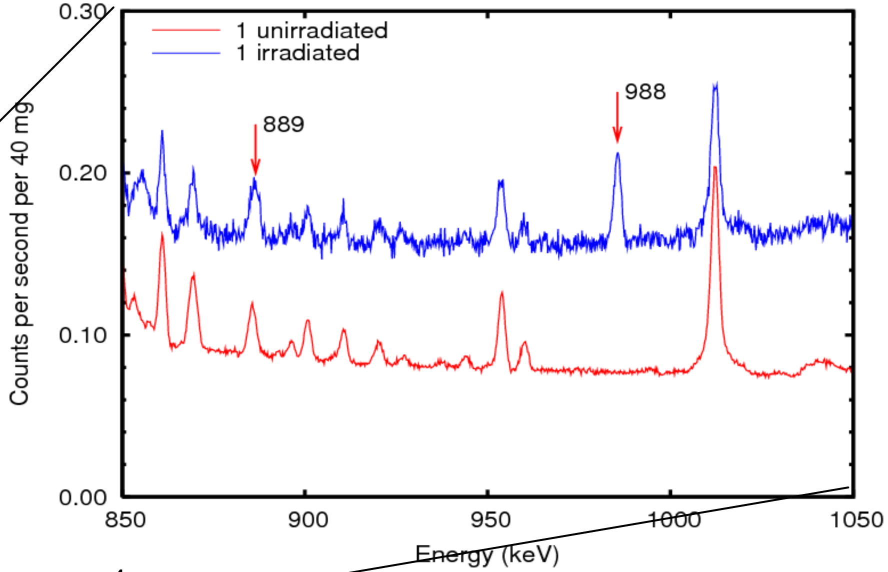
^{240}Am $T_{1/2}$ 50.8(3) h

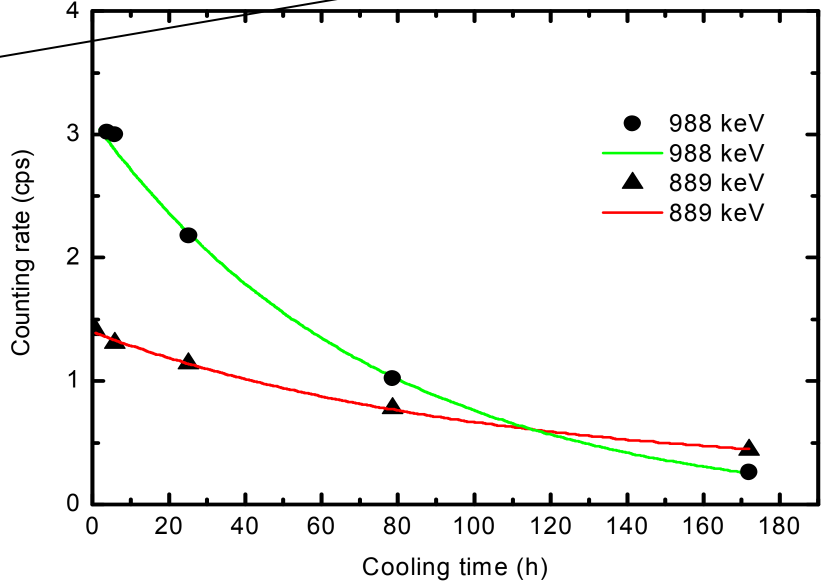
988 keV 73(4)%

889 keV 25.1(1.3)%

fitted $T_{1/2}$(988 keV)= 50.88 h





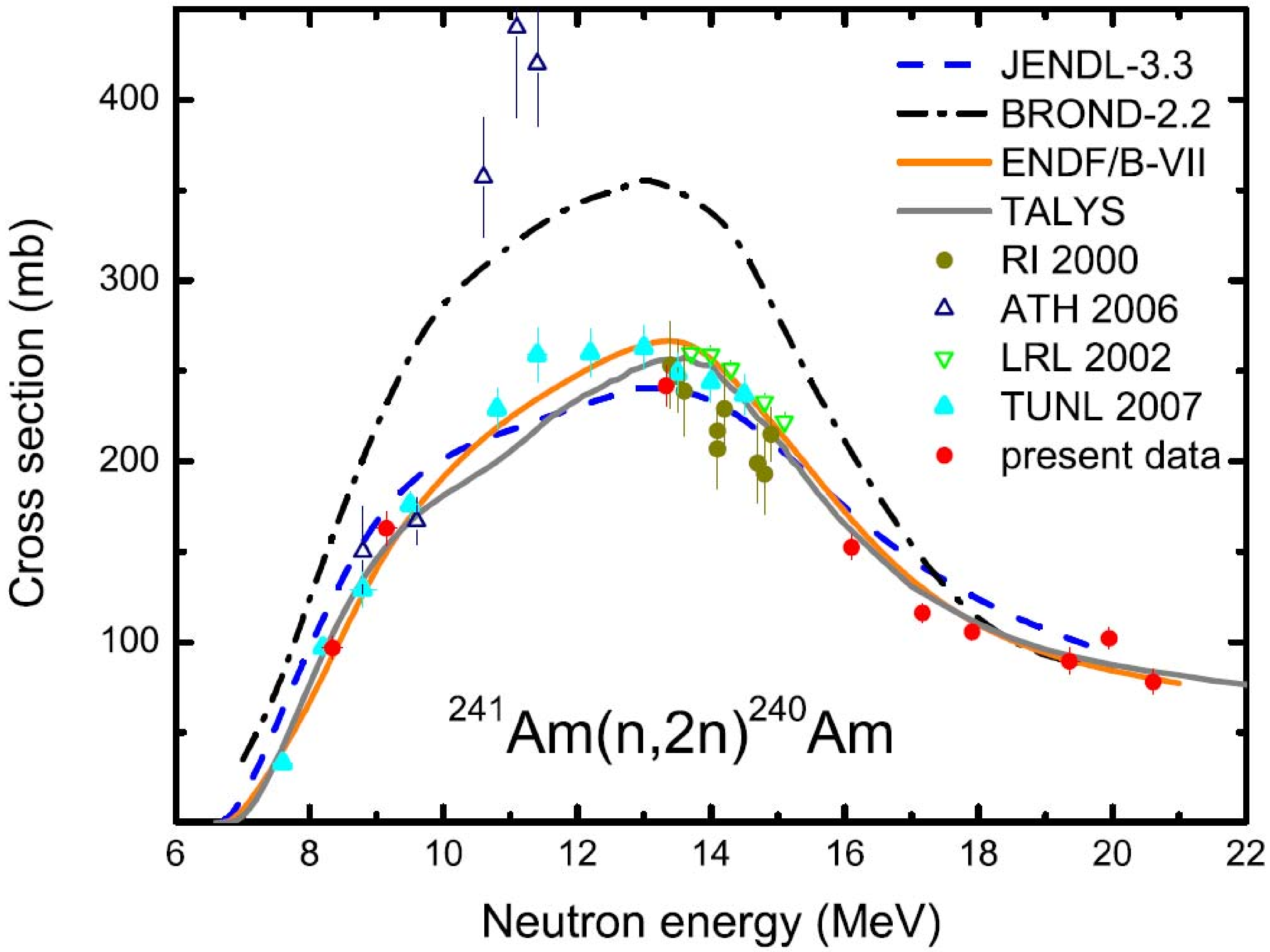


Shielding: 5 mm Pb + 2 mm Sn + 1 mm Cu

^{27}Al(n,α)^{24}Mg from container and matrix

Dead time ~ 10%

Results


Cross section (mb)
100
200
300
400
Neutron energy (MeV)
6
8
10
12
14
16
18
20
22
JENDL-3.3
BROND-2.2
ENDF/B-VII
TALYS
RI 2000
ATH 2006
LRL 2002
TUNL 2007
present data
$^{241}Am(n,2n)^{240}Am$