MEASUREMENT OF RADIATIVE CAPTURE CROSS SECTION ON ^{238}U AT THE n_TOF CERN FACILITY

Federica Mingrone
on behalf of the n_TOF Collaboration

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Budapest – October 28, 2013

Motivations

- Improvement in the design of *advanced nuclear systems* and *new nuclear fuel cycles* + simulation of *existing Gen.III reactors*

Fast Reactors: present uncertainties (%)

Isotope	Cross-section	ABTR	SFR		GFR		LFR	ADMAD		
		k_{eff}	k_{eff}	Void	k_{eff}	Power peak	k_{eff}	k_{eff}	Power peak	Void
^{238}U	Inelastic	0.69	0.23	1.96	1.41	1.54	0.73	–	–	–
	Capture	0.26	0.07	1.24	0.41	0.30	0.25	–	–	–
^{56}Fe	Inelastic	0.24	0.53	4.14	–	–	0.24	0.93	7.22	5.43
Na	Inelastic	0.07	0.25	13.43	–	–	–	–	–	–
^{28}Si	Inelastic	–	–	–	0.22	0.25	–	–	–	–
C	Elastic	–	–	–	0.31	0.28	–	–	–	–
^{206}Pb	Inelastic	–	–	–	–	–	0.18	–	–	–
^{207}Pb	Inelastic	–	–	–	–	–	0.16	–	–	–
^{208}Pb	Elastic	–	–	–	–	–	0.13	–	–	–
Pb [(a)]	Inelastic	–	–	–	–	–	–	0.04	0.28	2.28
	Elastic	–	–	–	–	–	–	0.05	0.09	2.84
	Capture	–	–	–	–	–	–	0.07	0.46	1.53
^{209}Bi	Inelastic	–	–	–	–	–	–	0.31	2.23	12.01

[(a)] For the ADMAB calculations have been performed with the JEF-2.2 library (see Section 4) that do not distinguish the Pb isotopes

Pu density at the end of fuel cycles: present uncertainties (%)

Uncertainty on → / Due to ↓		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
^{238}U	Capture	–	1.1	0.2	0.1	–
^{238}Pu	Capture	1.7	0.1	–	–	–
	Fission	4.6	–	–	–	–
^{239}Pu	Capture	–	0.8	1.3	0.7	0.1
	Fission	–	0.2	–	–	–
^{240}Pu	Capture	0.2	–	1.5	6.0	1.0
	Fission	–	–	0.8	0.4	–
^{241}Pu	Capture	–	–	–	0.8	1.5
	Fission	0.2	–	–	5.0	0.7
^{242}Pu	Capture	–	–	–	–	3.9
	Fission	–	–	–	–	2.2
^{241}Am	Capture	1.3	–	–	–	0.2
	Fission	0.2	–	–	–	–
Total		5.1	1.3	2.1	7.9	4.9

Thermal Reactors: present uncertainties (%)

Isotope	Cross-section	PWR	VHTR
		k_{eff} EOC	k_{eff} EOC
^{235}U	ν	0.17	0.27
^{238}U	Inelastic	0.17	0.00
	Capture	0.26	0.19
^{230}Pu	Fission	0.18	0.10
	Capture	0.07	0.11
^{240}Pu	Capture	0.12	0.06
^{241}Pu	Fission	0.34	0.18
	Capture	0.13	0.13
O	Capture	0.43	0.01

Final Report – Subgroup 26 of Organisation for Economic Co-operation and Development (**OECD**) and Nuclear Energy Agency (**NEA**)

Motivations

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Fast Reactors: present uncertainties (%)

		ABTR	SFR		GFR		LFR	ADMAD		
Isotope	Cross-section	k_{eff}	k_{eff}	Void	k_{eff}	Power peak	k_{eff}	k_{eff}	Power peak	Void
^{238}U	Inelastic	0.69	0.23	1.96	1.41	1.54	0.73	–	–	–
	Capture	0.26	0.07	1.24	0.41	0.30	0.25	–	–	–
^{206}Pb	Inelastic	–	–	–	–	–	0.18	–	–	–
^{207}Pb	Inelastic	–	–	–	–	–	0.16	–	–	–
^{208}Pb	Elastic	–	–	–	–	–	0.13	–	–	–
Pb [(a)]	Inelastic	–	–	–	–	–	–	0.04	0.28	2.28
	Elastic	–	–	–	–	–	–	0.05	0.09	2.84
	Capture	–	–	–	–	–	–	0.07	0.46	1.53
^{209}Bi	Inelastic	–	–	–	–	–	–	0.31	2.23	12.01

(a) For the ADMAB calculations have been performed with the JEF-2.2 library (see Section 4) that do not distinguish the Pb isotopes

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Uncertainty on → / Due to ↓		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
^{238}U	Capture	–	1.1	0.2	0.1	–
^{239}Pu	Capture	–	0.8	1.3	0.7	0.1
	Fission	–	0.2	–	–	–
^{240}Pu	Capture	0.2	–	1.5	6.0	1.0
	Fission	–	–	0.8	0.4	–
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^{241}Am	Capture	1.3	–	–	–	0.2
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n_TOF

Motivations

- EXFOR DATABASE: more than 25 datasets in the resolved resonance region and a few less in the unresolved BUT still *inconsistencies for the capture cross section*

NEA High Priority Request List

UNCERTAINTY IN THE CROSS SECTION BELOW 2%

- EC-JRC-IRMM Gelina with C_6D_6 detection system
- n_TOF with TAC detection system
- n_TOF with C_6D_6 detection system

Table 32. Summary of Highest Priority Target Accuracies for Fast Reactors

		Energy Range	Current Accuracy (%)	Target Accuracy (%)
U238	σ_{inel}	6.07 ÷ 0.498 MeV	10 ÷ 20	2 ÷ 3
	σ_{capt}	24.8 ÷ 2.04 keV	3 ÷ 9	1.5 ÷ 2

Table 34. PWR: uncertainty reduction requirements needed to meet integral parameter target accuracies

Isotope	Cross-Section	Energy range	Uncertainty (%)		
			Initial	Required	
				$\lambda=1$	$\lambda\neq1$ [(a)]
U235	σ_{capt}	67.4 - 2.03 keV	33	12	10
U238	σ_{capt}	24.8 - 9.12 keV	9	5	4
		454 - 22.6 eV	2	1	1
	σ_{scatt}	6.07 - 1.35 MeV	20	5	5

Radiative capture on ^{238}U campaign

SAMPLE	DATE (YEAR 2012)	PROTONS
^{238}U	**10 d:** March 29 – April 03, April 25 – 27, April 30 – May 02; **Filters** – **7 d:** April 5 – 6, April 11 – 16	7.85×10^{17} + 1.2×10^{18}
Sample Out	**6 d:** March 28 – 2; Filters – 3 d: April 9 – 1[illegible]	6.271×10^{16} + [illegible].64 $\times 10^{17}$
^{238}U packing	**2 d:** April 21 – 22	9.65×10^{16}
Beam off	**0.25 d:** March 28, April 5, April 11	
Calibrations	**0.25 d:** March 28, April 5, April 11	
Pb	**2d:** April 03 – 04; **Filters** – **2 d:** April 8 – 9	1.50×10^{17} + 1.53×10^{17}
C (5 mm)	**3 d:** April 4 – 5, April 17; **Filters** – **2 d:** April 7 – 8	1.46×10^{17} + 1.66×10^{17}
C (10 mm)	**2 d:** April 19 – 20; **Filters** – **3 d:** April 28 – 30	9.71×10^{16} + 1.11×10^{17}
Au (50 μm)	**2 d:** April 5, April 16	4.86×10^{16}
Au (300 μm)	**2 d:** April 17 – 18; **Filters** – **2 d:** April 27 – 28	8.33×10^{16} + 9.93×10^{16}
Ag	**1 d:** May 02	1.60×10^{16}

^{238}U(n,γ) – TOT: 17 days

Radiative capture on ^{238}U campaign

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^{238}U	**10 d:** March 29 – April 03, April 25 – 27, April 30 – May 02; Filters – **7 d:** April 5 – 6, April 11 – 16	7.85×10^{17} + 1.2×10^{18}
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Ag	**1 d:** May 02	1.60×10^{16}

BACKGROUND – TOT: 14.5 days

Radiative capture on ^{238}U campaign

SAMPLE	DATE (YEAR 2012)	PROTONS
^{238}U	**10 d:** March 29 – April 03, April 25 – 27, April 30 – May 02; Filters – **7 d:** April 5 – 6, April 11 – 16	7.85×10^{17} + 1.2×10^{18}
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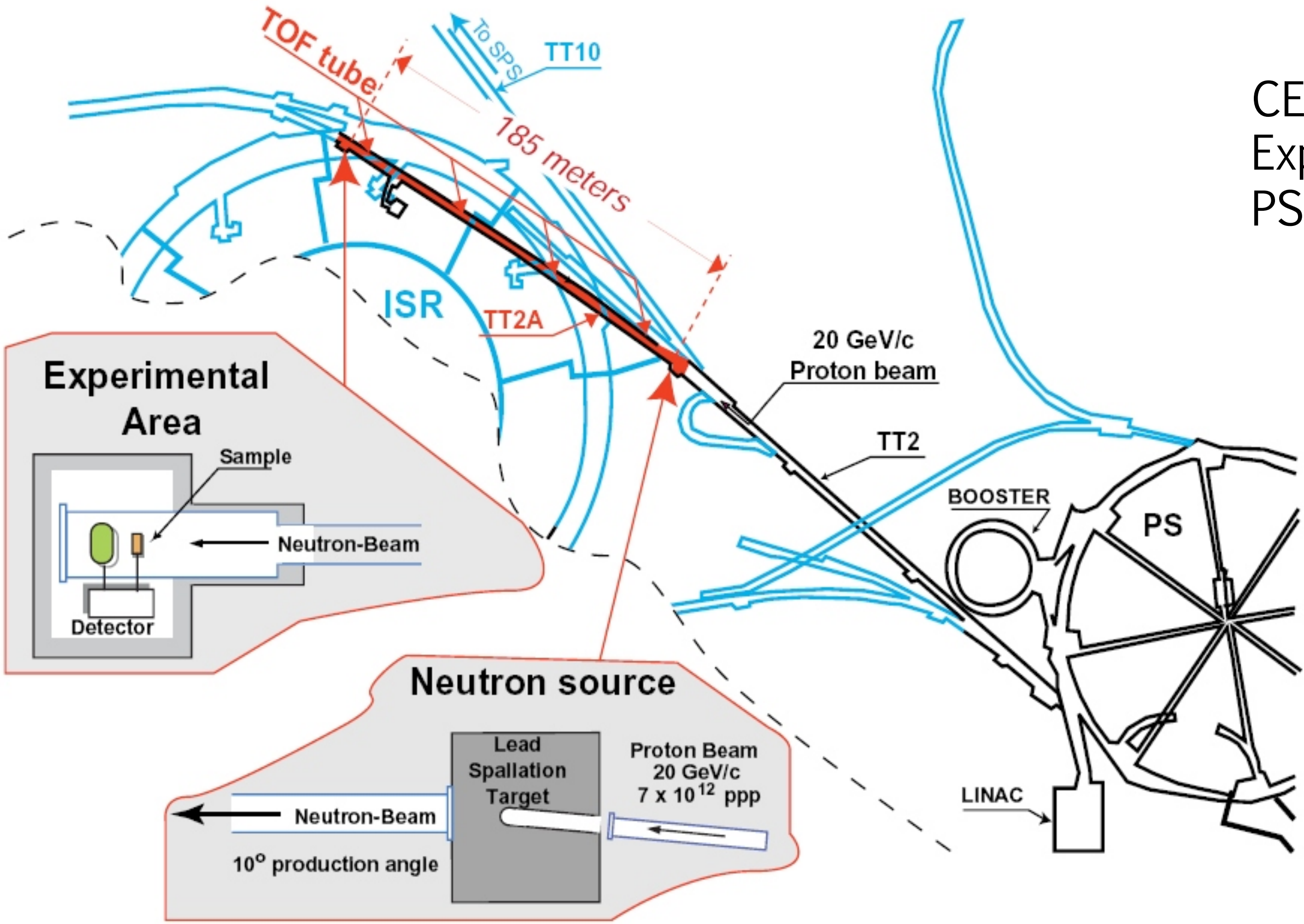
Ag	**1 d:** May 02	1.60×10^{16}

NORMALIZATION – TOT: 3.5 days

n_TOF facility at CERN



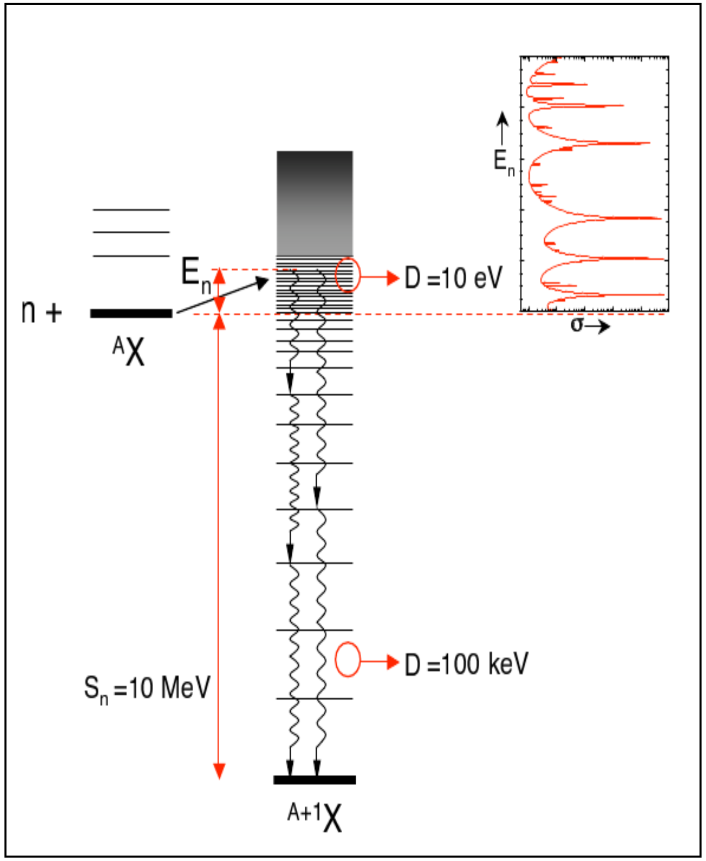
CERN
Experiment
PS213

n_TOF facility: characteristics

Proton beam momentum	20 GeV/c
Intensity (dedicated mode)	7×10^{12} protons/pulse
Repetition frequency	1 pulse/2.4 s
Pulse width	7 ns (rms)
Neutrons/proton	300
Lead target dimensions	∅=60 cm, h=40 cm
Cooling material	H_2O
Moderation material	Borated Water (H_2O + 1.28% H_3BO_3)
Moderator thickness in the exit face	1 cm cooling material + 4 cm moderator

Experimental Technique

Using a Time of Flight (TOF) technique, capture cross section is determined through the measurement of **the reaction yield $Y_R(E_n)$**:



$$Y = N\frac{C-B}{\varepsilon\,\Phi}$$

N: normalization factor to obtain an absolute capture yield
C: capture counts
B: background counts
Φ: incident neutron fluence
ε = k x E_c: detection efficiency

The experimental yield can be expressed in terms of **total (σ_{tot})** and **capture (σ_γ)** cross section:

$$Y(E_n) = (1 - e^{-n\sigma_{tot}})\frac{\sigma_\gamma}{\sigma_{tot}}$$

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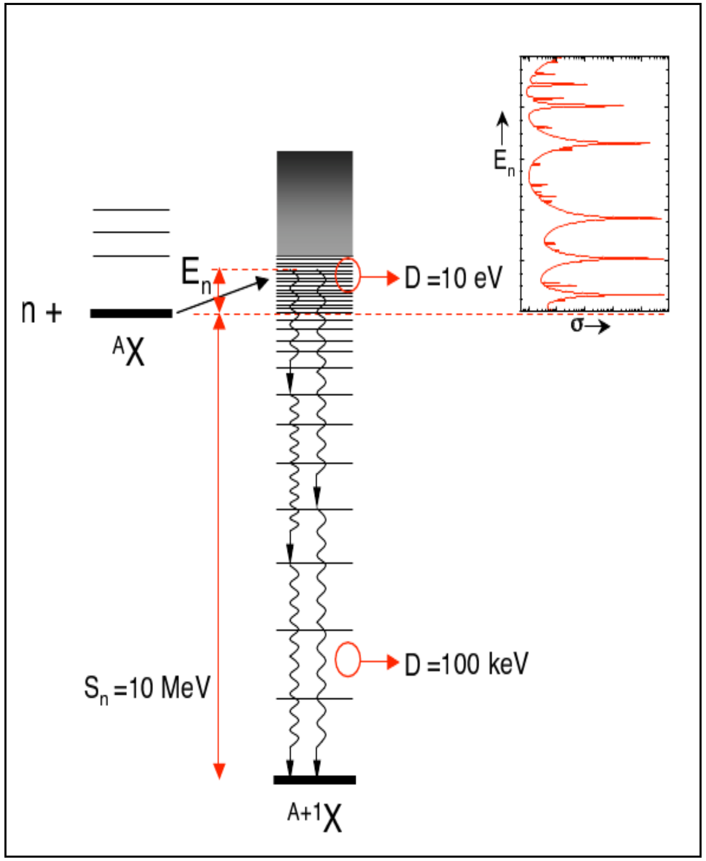
Experimental Technique

For capture measurements, detection efficiency have to be independent from γ-spectrum multiplicity and from γ-ray energy distribution

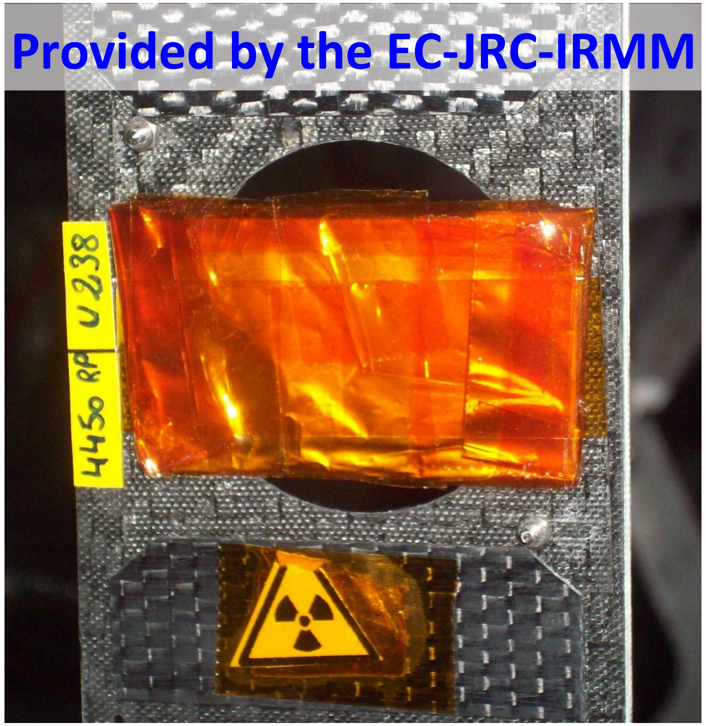


- **Total Absorption Detection**
  - $\varepsilon_\gamma \sim 100\ \%$
  - 4π geometry detectors
- **Total Energy Detection**
  - Low detection efficiency ε_γ
  - Low solid angle

$$\varepsilon_c = \sum \varepsilon_\gamma = k \cdot \left(B_n + E_n^{(cm)}\right)$$

Samples

Enriched metallic uranium rectangular plate (53×30 mm^2, 235 μm thick) enveloped inside a 20 μm aluminum and a 25 μm kapton thick foils.

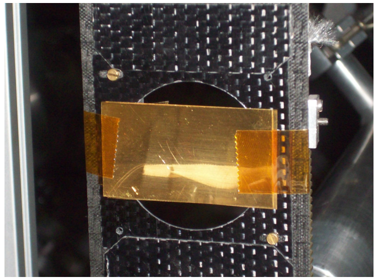


MASS	6.125 ± 0.002 g 9.6×10^{-4} atoms/barn
% 238 U	99.99 %
% 235 U	< 11 ppm
% 233-234-236 U	< 1 ppm each
EXPECTED RADIOACTIVITY	12.4 kBq/g (76 kBq)

Isotopic analysis done @ IRMM in 1984

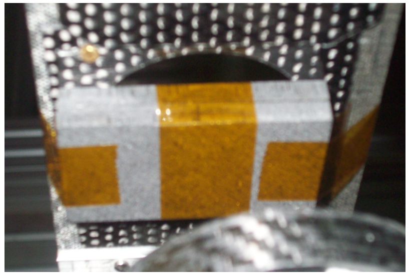
Samples

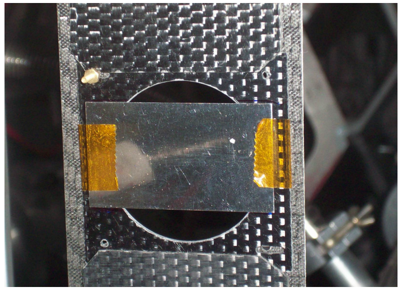
The other samples analyzed have been chosen with geometry as similar as possible to that of the ^{238}U.



^{197}**Au**

C

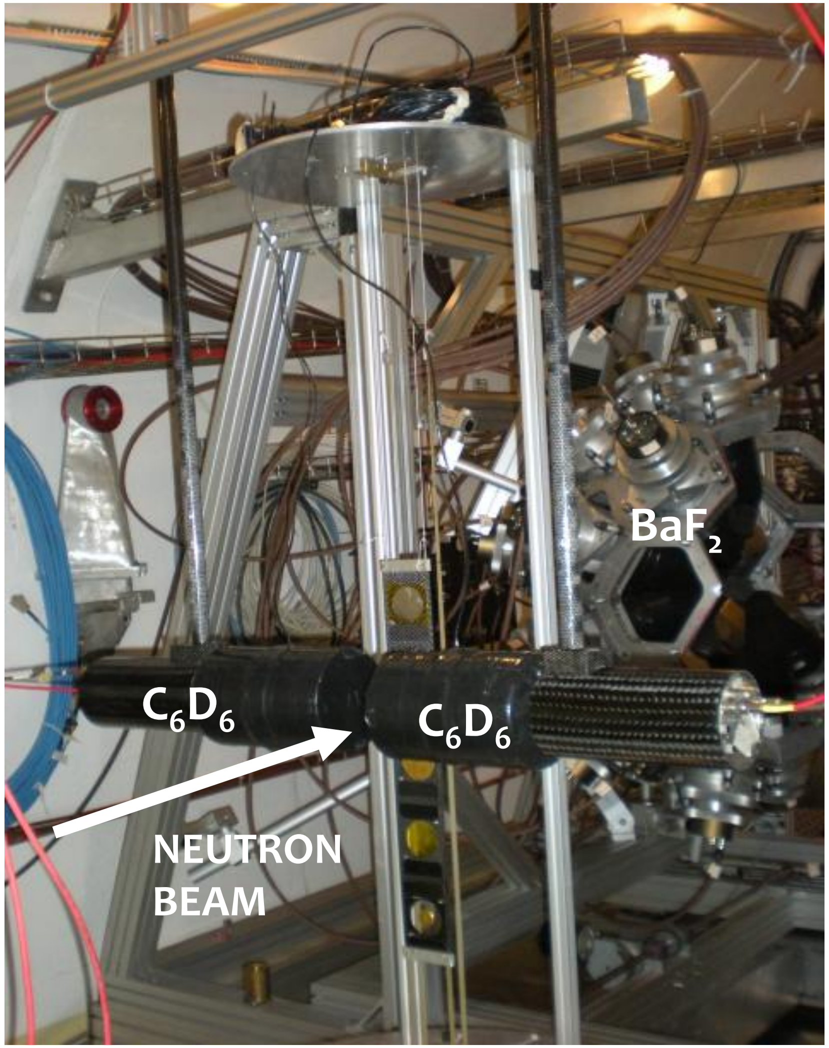




^{nat}**Pb**

SAMPLE	SIZE [mm]	MASS [g]	ATOMIC DENSITY [atoms/barn]
^{197}Au	53.30 × 29.65	9.213	1.773 10^{-3}
Pb	53.77 × 30.19	9.44	1.725 10^{-3}
C	53.35 × 30.20	28.89	8.94 10^{-2}
Fe	53.71 × 30.22	3.225	2.13 10^{-3}
Ag	53.75 × 30.30	4.620	1.59 10^{-3}

Experimental Set–up



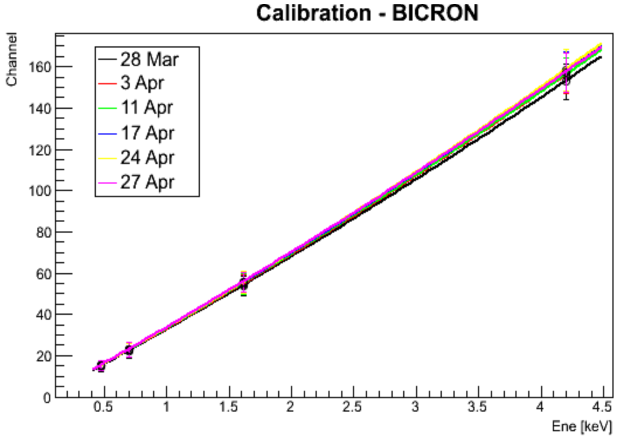
Two different setups for capture measurements

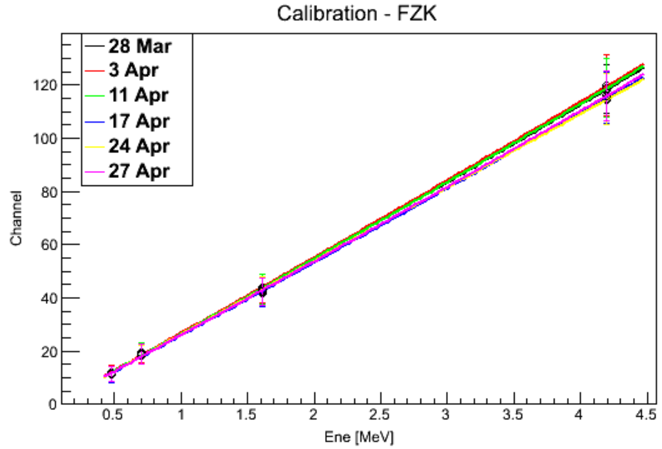
- **Total Absorption Calorimeter**
  - 40 BaF_2 crystals in a 4π geometry
  - Detects the entire γ cascade (together with background n)
- **Two C_6D_6 scintillation detectors**
  - Optimized for an extremely low neutron sensitivity $(\varepsilon_n/\varepsilon_\gamma < 4 \cdot 10^{-5})$
  - Only one γ–ray detected per cascade

    → *Total Energy Detection Technque* with PHWT

Data Reduction

CALIBRATIONS





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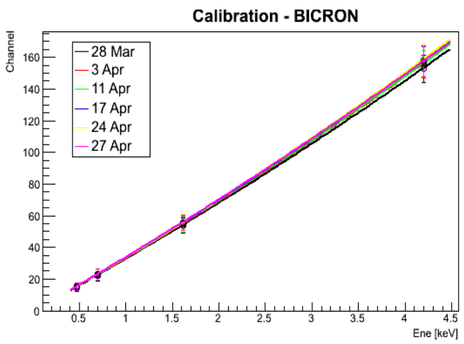
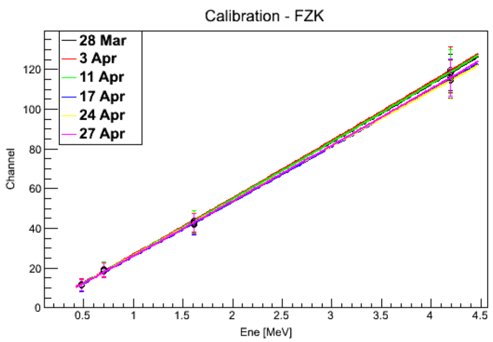
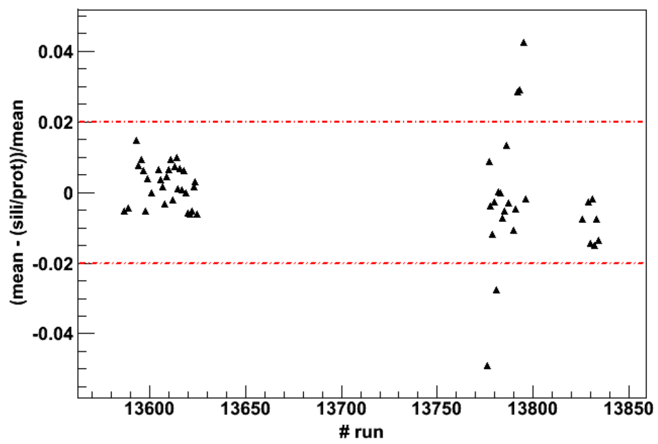
Data Reduction

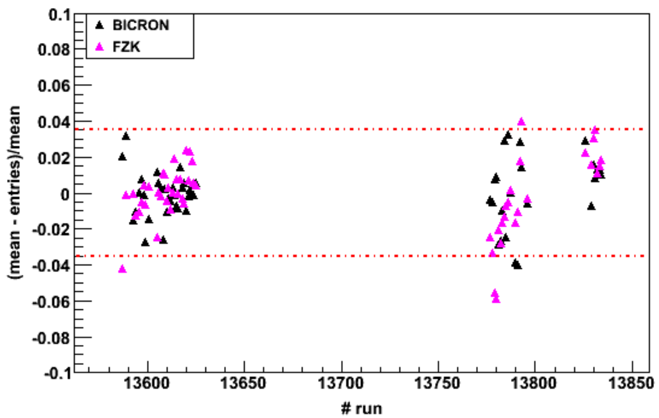
CALIBRATIONS





STABILITY CHECK





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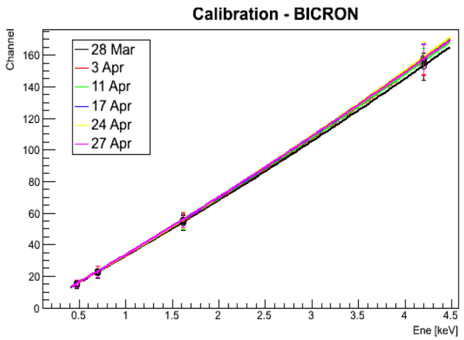
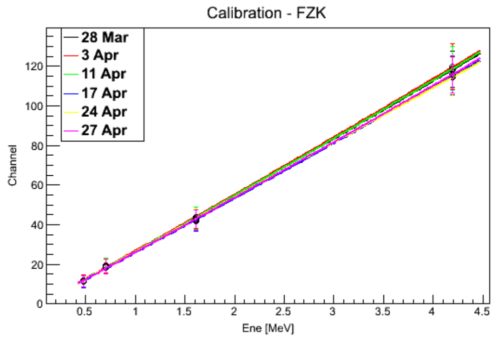
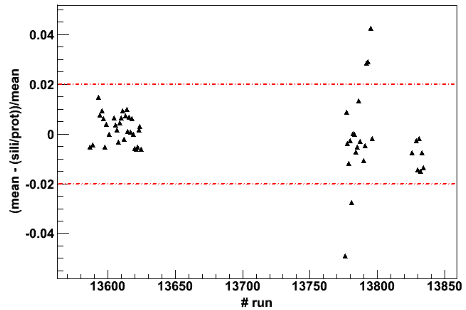
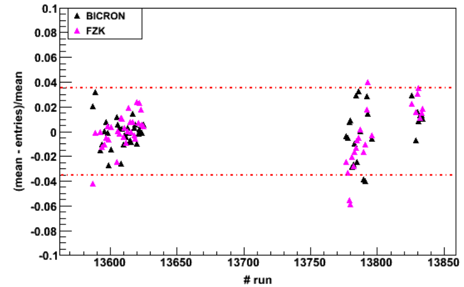


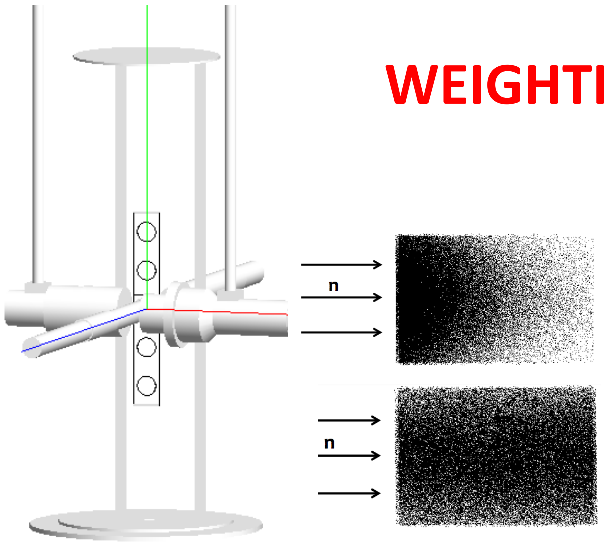


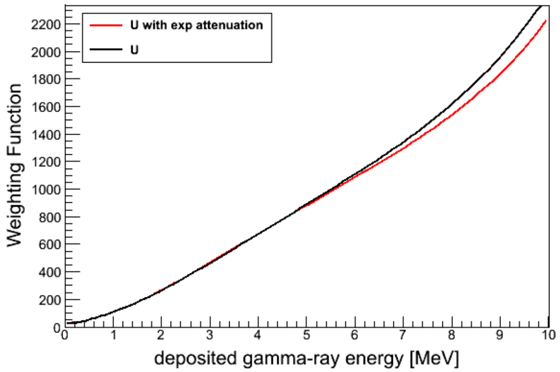
STABILITY CHECK



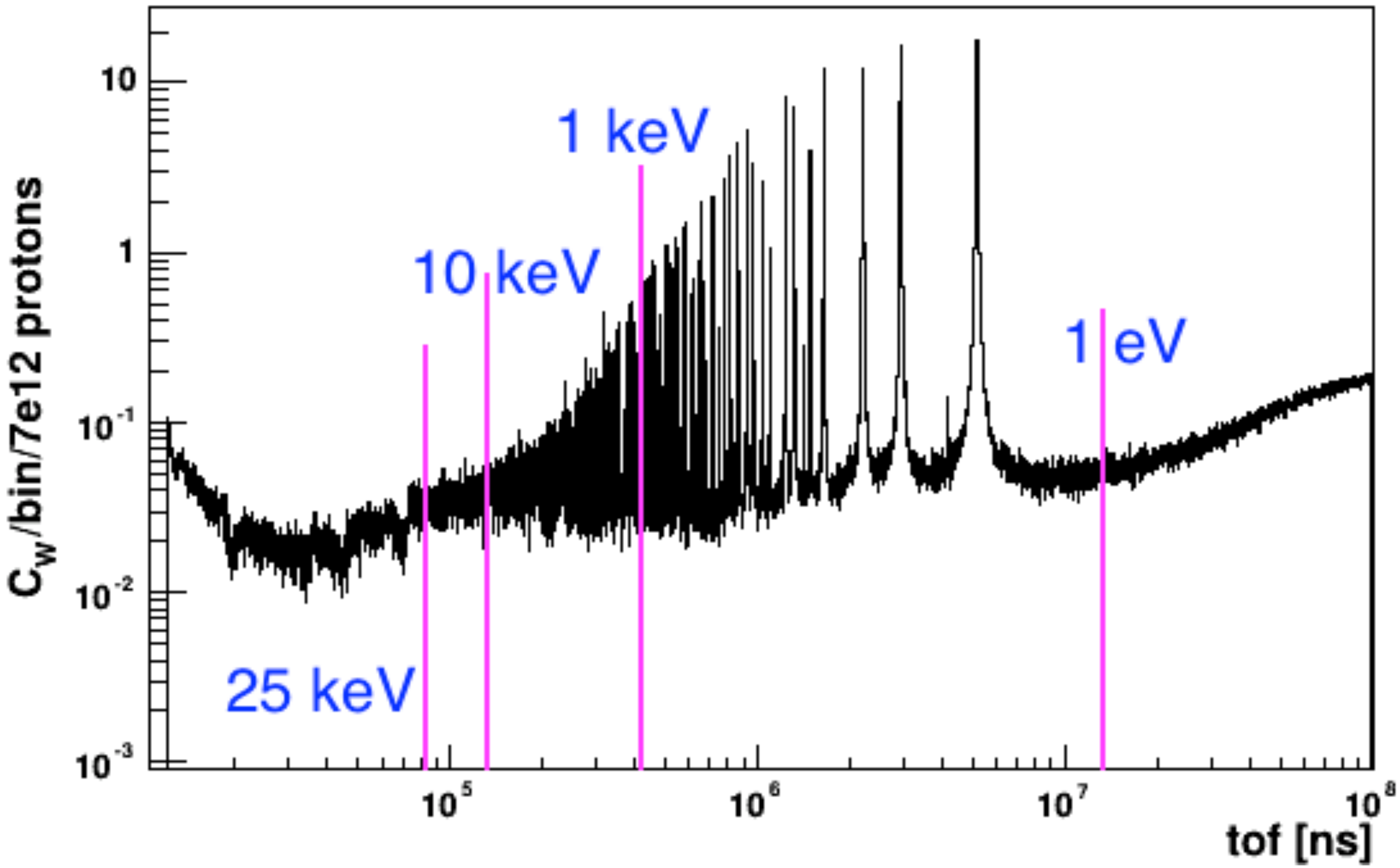


WEIGHTING FUNCTIONS

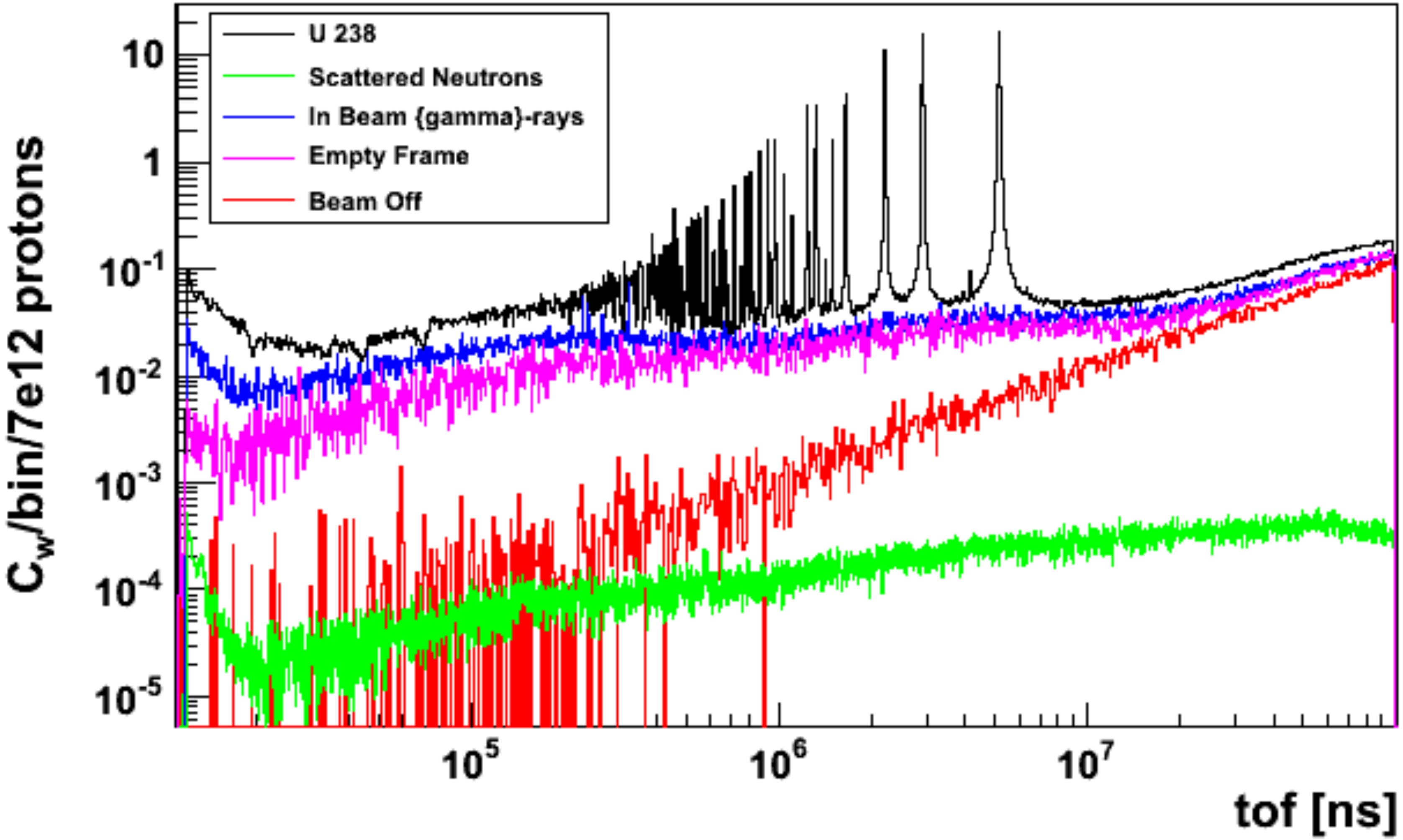




$^{238}U(n,\gamma)$ TOF spectrum

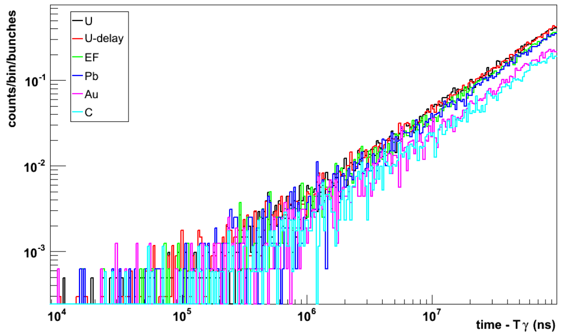


Background subtraction

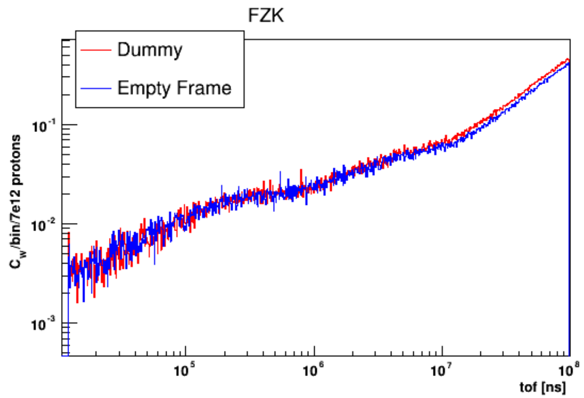


Background subtraction

- **Beam Off**: Natural + ^{238}U radioactivity

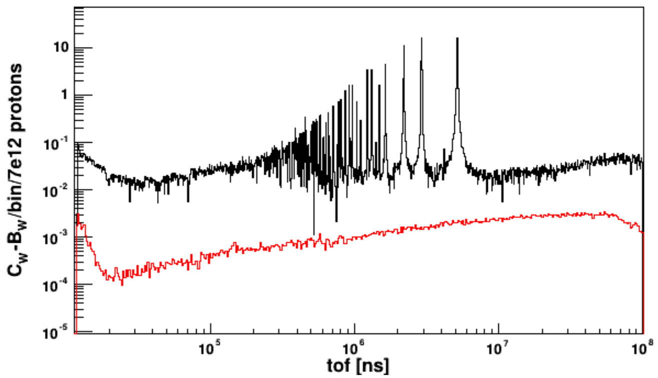


- **Sample Out** and ^{238}U packaging

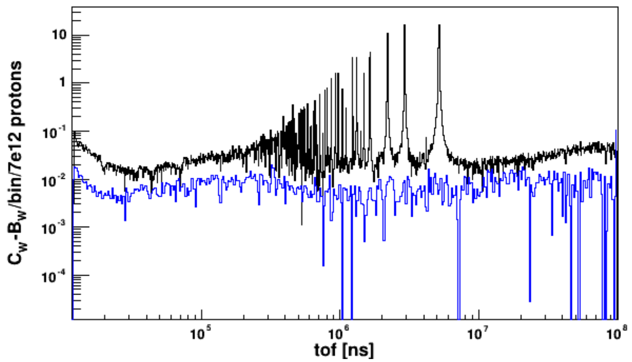


- **Scattered neutrons**: Carbon sample

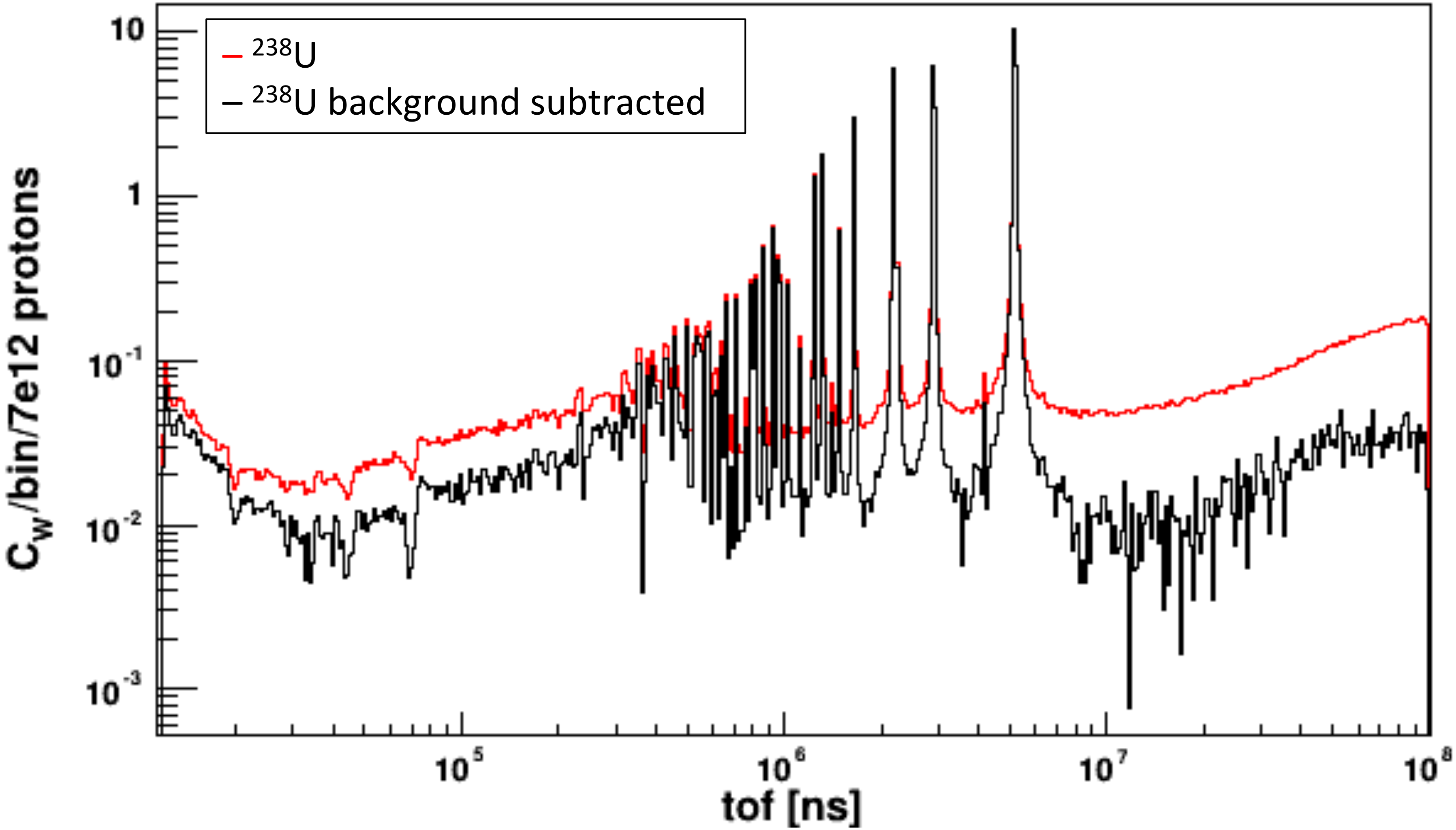
$$Y_{sc}^{U} = \frac{n_U}{n_C} \frac{\sigma_{el}^{U}}{\sigma_{el}^{C}} Y_C$$



- **In-beam gamma rays**: Lead sample (similar Z)



Background subtraction



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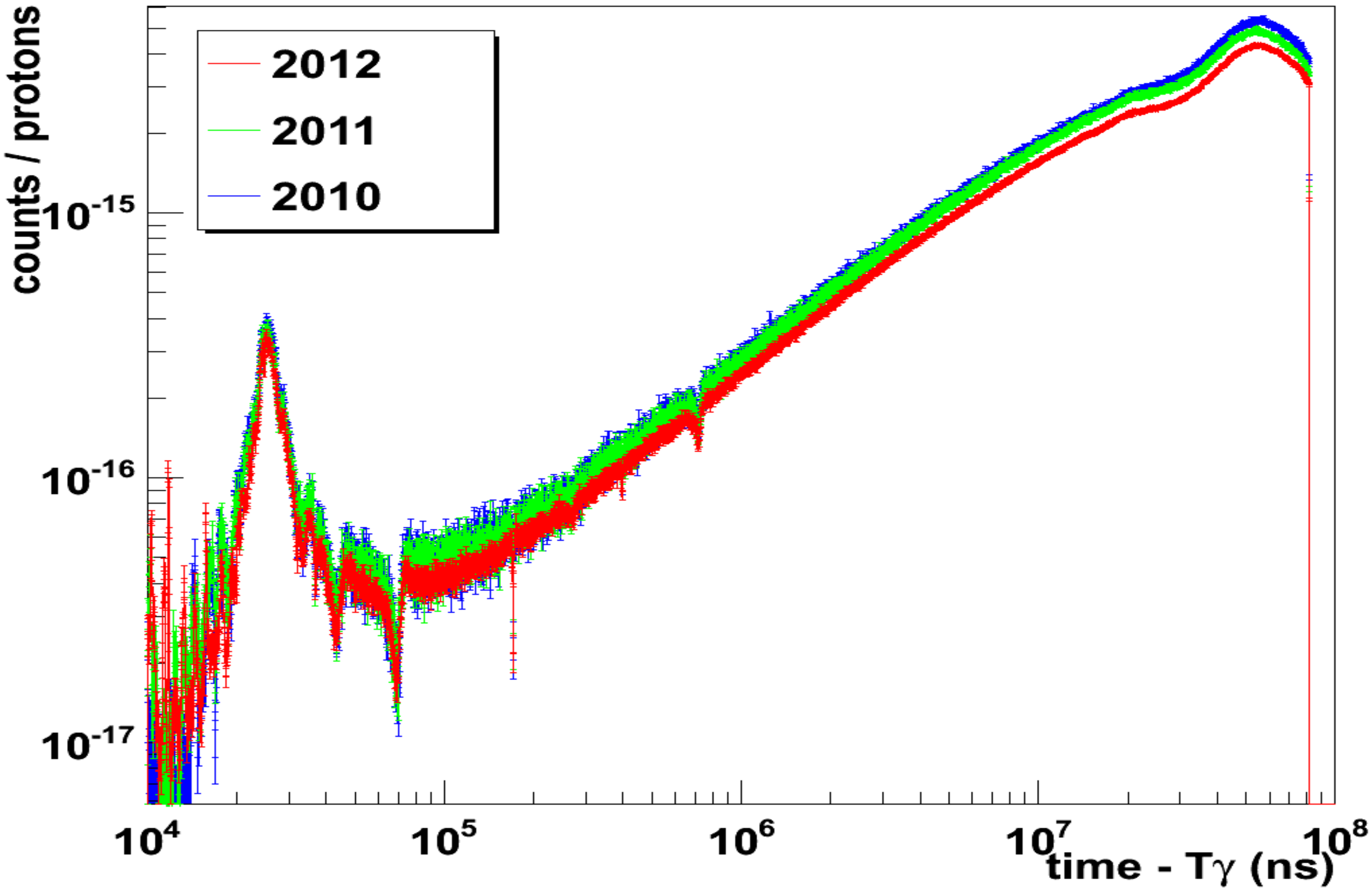
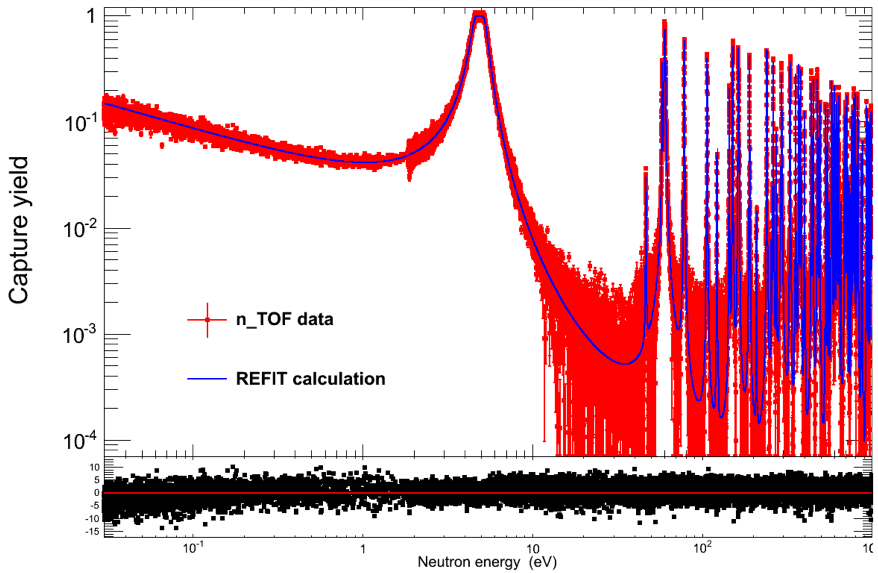
n_TOF

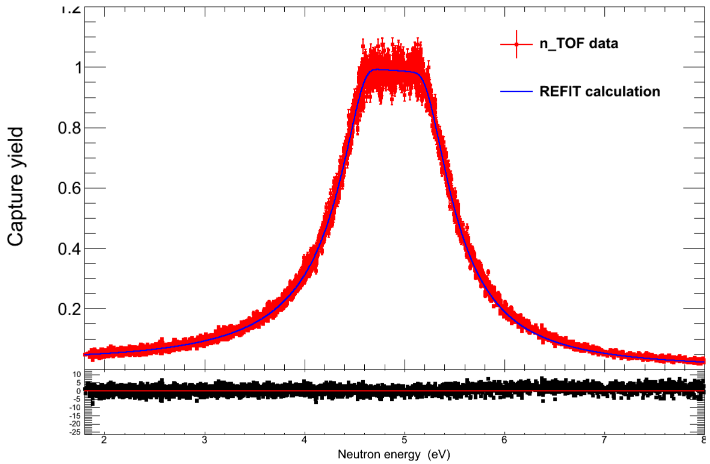
n_TOF flux

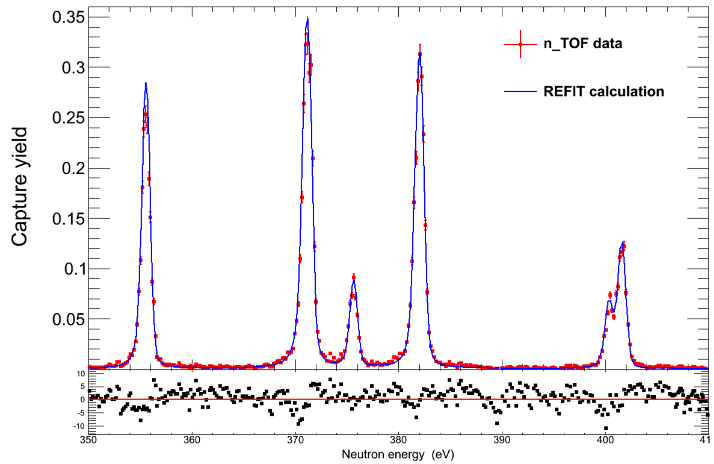


counts / protons
2012
2011
2010
10^{-15}
10^{-16}
10^{-17}
10^{4}
10^{5}
10^{6}
10^{7}
10^{8}
time - Tγ (ns)

^{197}Au YIELD: analysis validation







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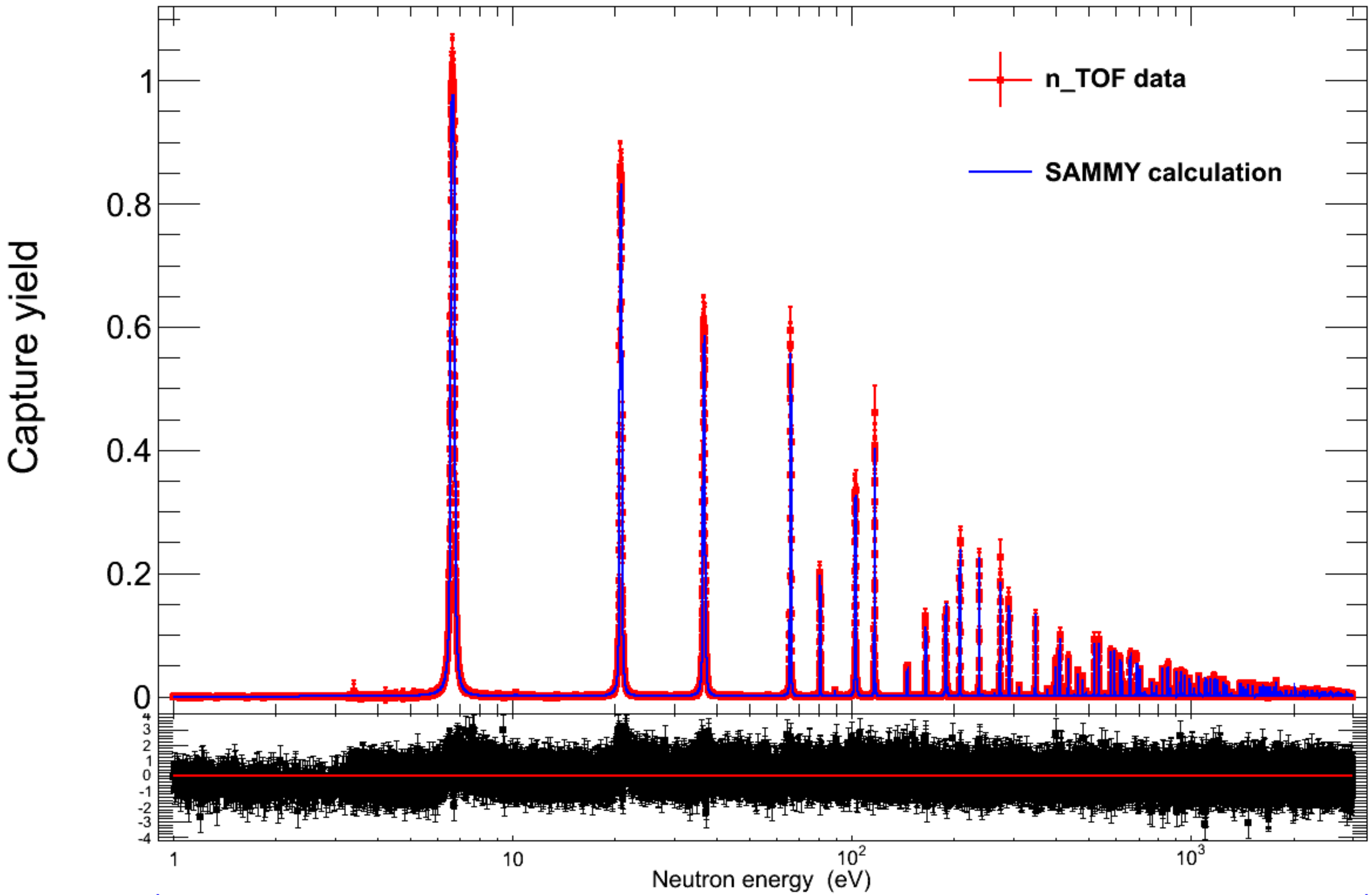
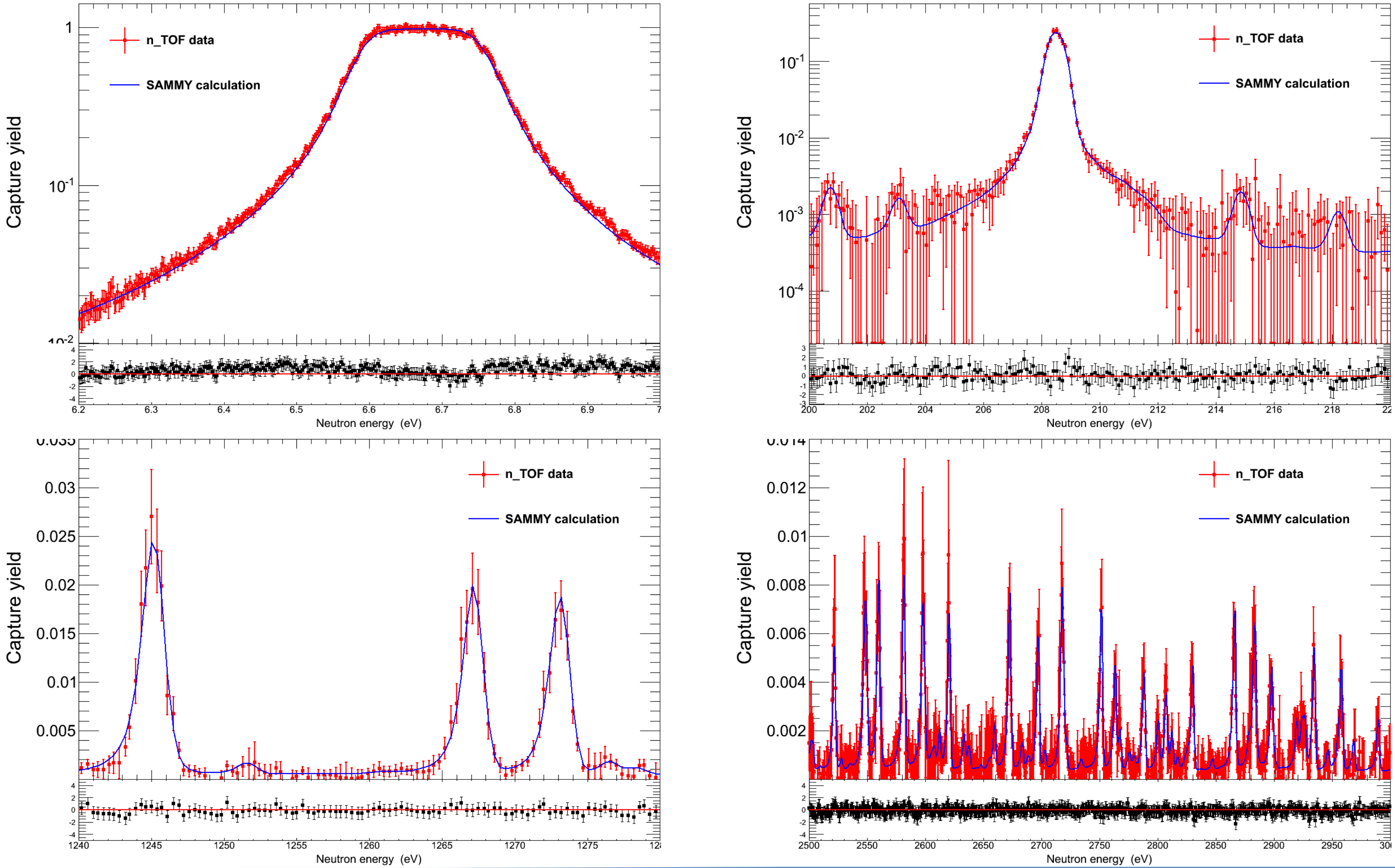
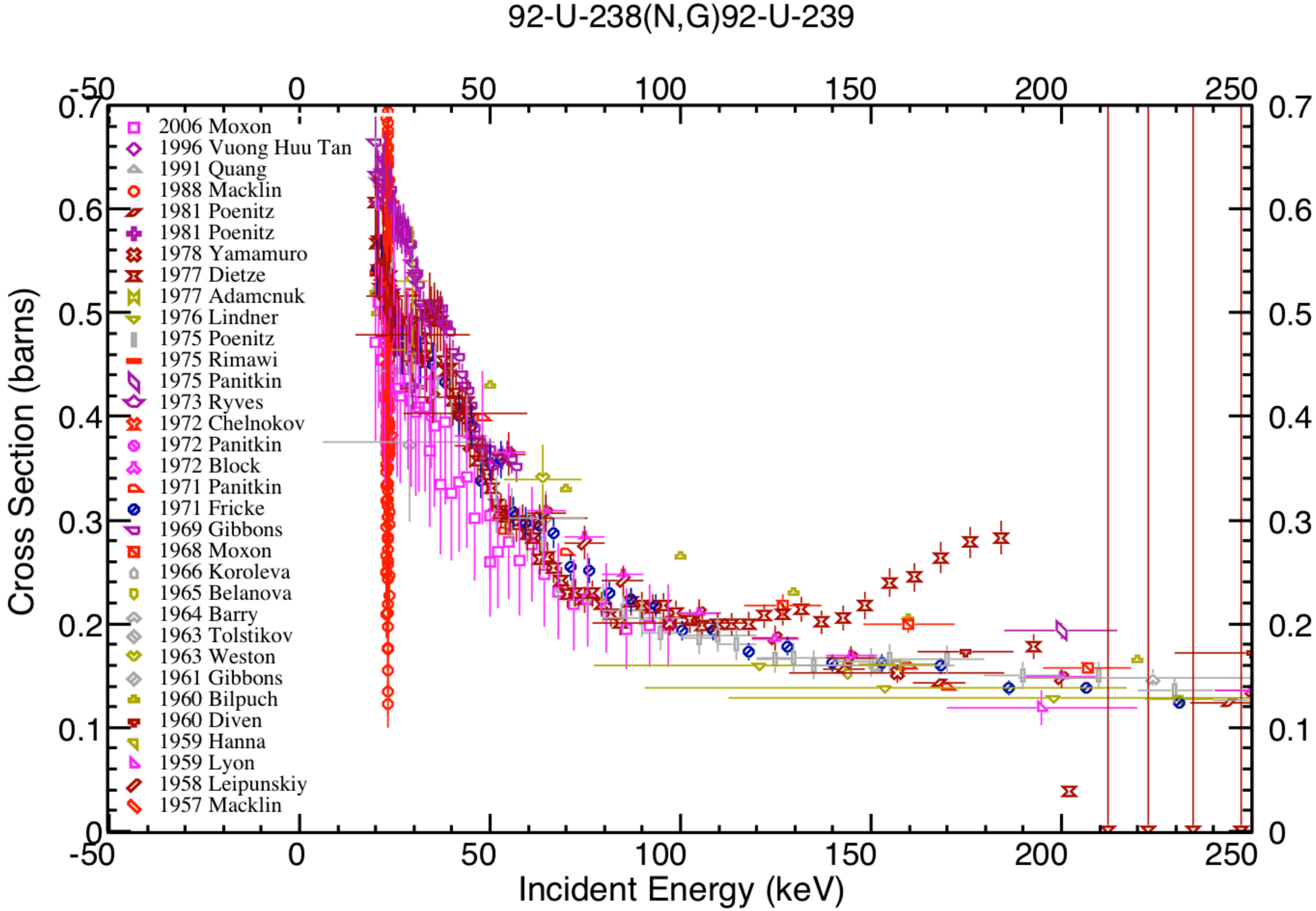
^{238}U CAPTURE YIELD



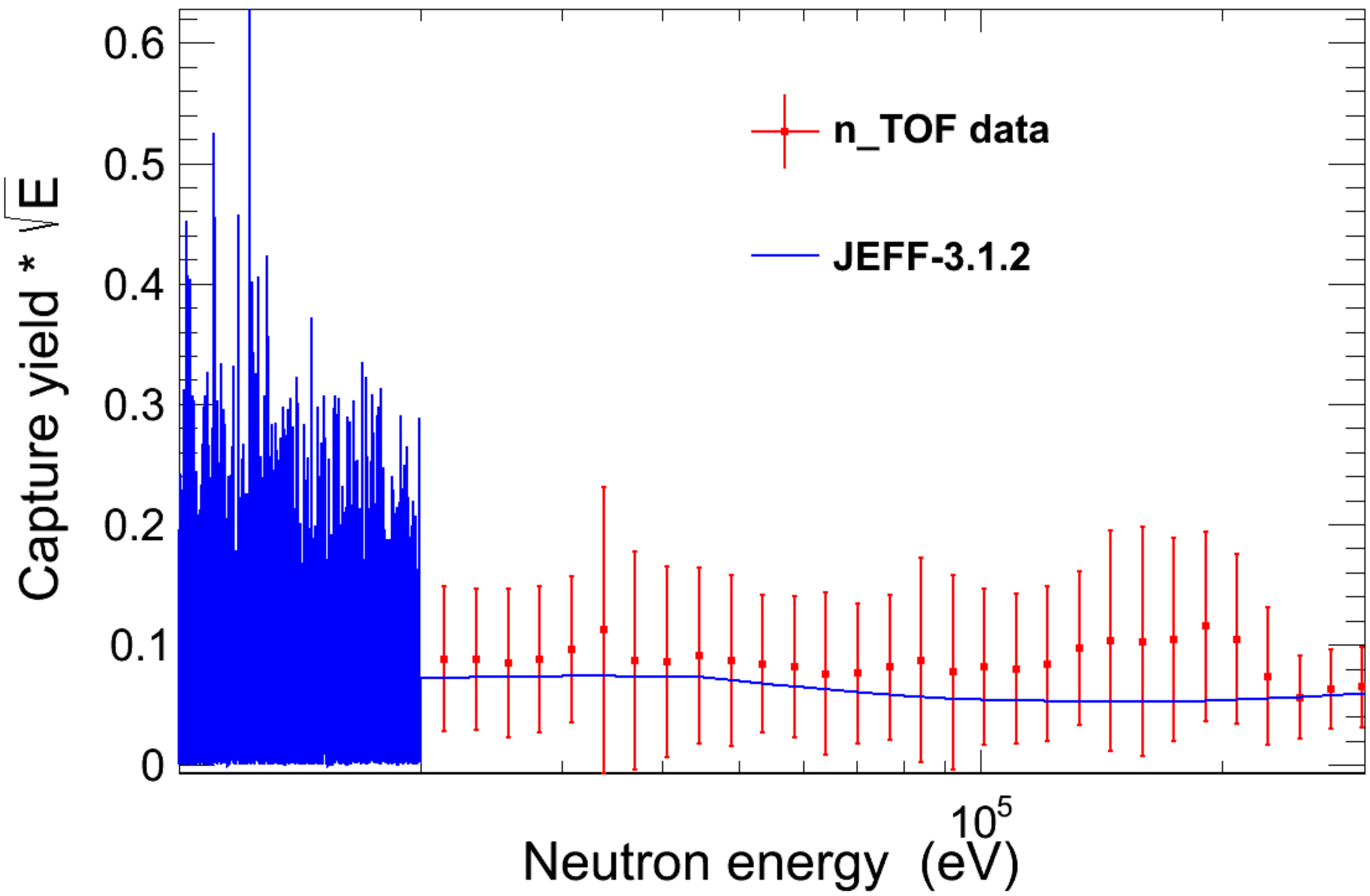
^{238}U CAPTURE YIELD



^{238}U CAPTURE YIELD – URR



^{238}U CAPTURE YIELD – URR



Conclusions

- Accurate data reduction: calibrations, stability check, weighting functions
- Accurate background subtraction (analysis of the background level with filters ongoing)

SOURCE OF UNCERTAINTY	1 eV < En < 1-3 keV	THERMAL + URR
Sample mass	0.03%	0.03%
Neutron flux (shape in E_n)	~1%*	<3%*
Neutron flux (abs. value) [i.e. normalization]	<1%	<1%
Background	0-1% (resonance peak)	<3%
Overall	**1.1-1.5%**	**~4%**

*C. Guerrero et al., Eur. Phys. J. **A** (2013)

Thank you!

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BACKUP

Radiative capture on ^{238}U campaign

SAMPLE	DATE (YEAR 2012)	PROTONS
^{238}U	**10 d:** March 29 – April 03, April 25 – 27, April 30 – May 02; **Filters** – **7 d:** April 5 – 6, April 11 – 16	7.85×10^{17} + 1.2×10^{18}
Sample Out	**6 d:** March 28 – 2; **Filters** – **3 d:** April 9 – 11	6.271×10^{16} + 1.64×10^{17}
^{238}U packing	**2 d:** April 21 – 22	3.65×10^{16}
Beam off	**0.25 d:** March 28, April 5, April 11	
Calibrations	**0.25 d:** March 28, April 5, April 11	
Pb	**2d:** April 03 – 04; **Filters** – **2 d:** April 8 – 9	1.50×10^{17} + 1.53×10^{17}
C (5 mm)	**3 d:** April 4 – 5, April 17; **Filters** – **2 d:** April 7 – 8	1.46×10^{17} + 1.66×10^{17}
C (10 mm)	**2 d:** April 19 – 20; **Filters** – **3 d:** April 28 – 30	9.71×10^{16} + 1.11×10^{17}
Au (50 µm)	**2 d:** April 5, April 16	4.86×10^{16}
Au (300 µm)	**2 d:** April 17 – 18; **Filters** – **2 d:** April 27 – 28	8.33×10^{16} + 9.93×10^{16}
Ag	**1 d:** May 02	1.60×10^{16}

ENTIRE CAMPAIGN: 35 days

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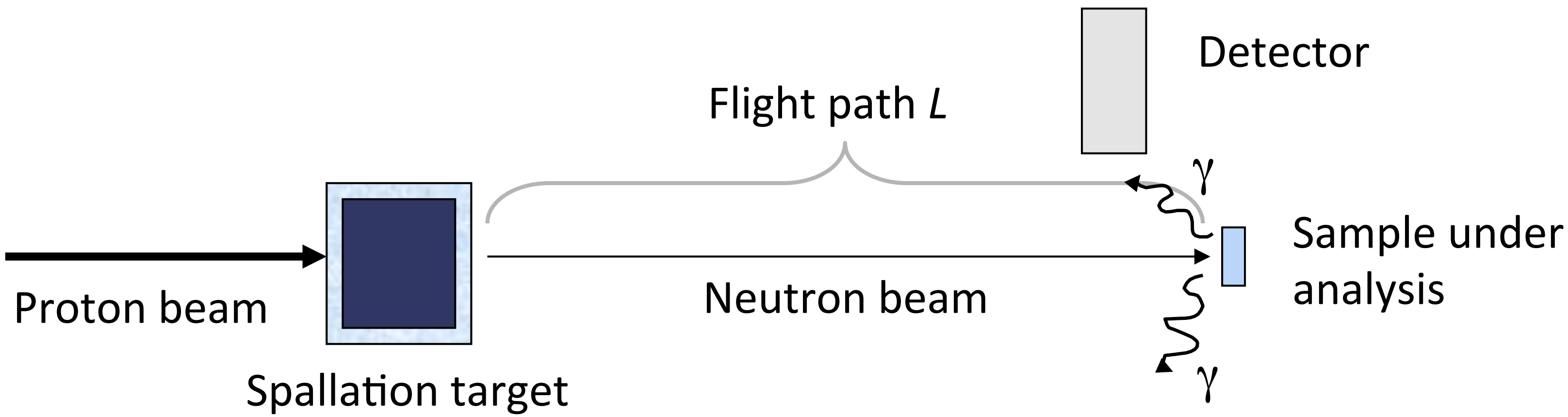
n_TOF

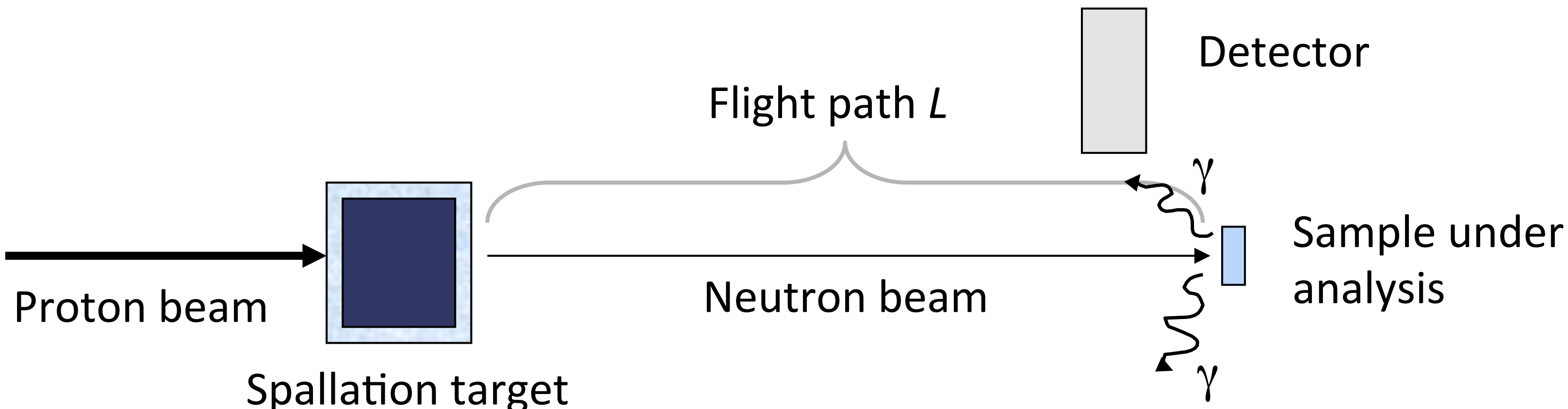
EXPERIMENTAL TECHNIQUE



$$tof = (t_{det} - t_{start}) + t_0$$

$$E_n[eV] = \left(\frac{72.2983 \times L[m]}{tof[\mu s]}\right)^2$$

EXPERIMENTAL TECHNIQUE



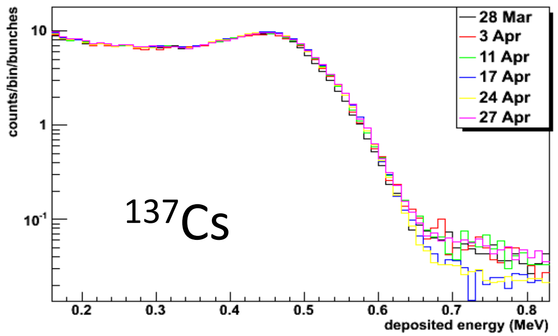
$$tof = (t_{det} - t_{start}) + t_0$$

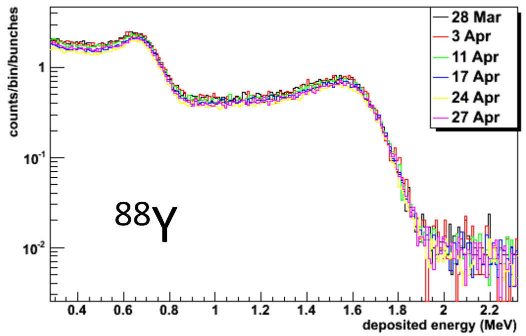
$$E_n[eV] = \left(\frac{72.2983 \times L[m]}{tof[\mu s]} \right)^2$$

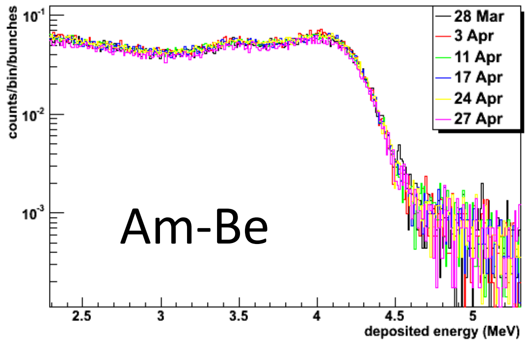
$$\Longrightarrow \quad \frac{\Delta E_n}{E_n} = (\gamma + 1)\gamma \frac{\Delta v}{v}$$

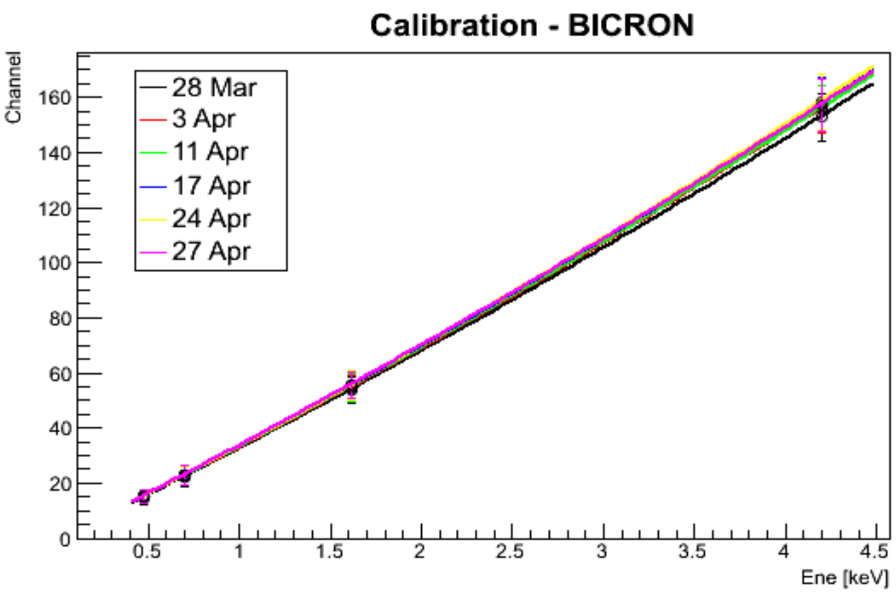
E_n [eV]	FWHM [cm]	ΔE_n [eV]
1	3	3.2×10^{-4}
10	3	3.2×10^{-3}
10^2	4	4.3×10^{-2}
10^3	5	5.4×10^{-1}
10^4	10	11
10^5	27	2.9×10^2
10^6	49	5.3×10^3

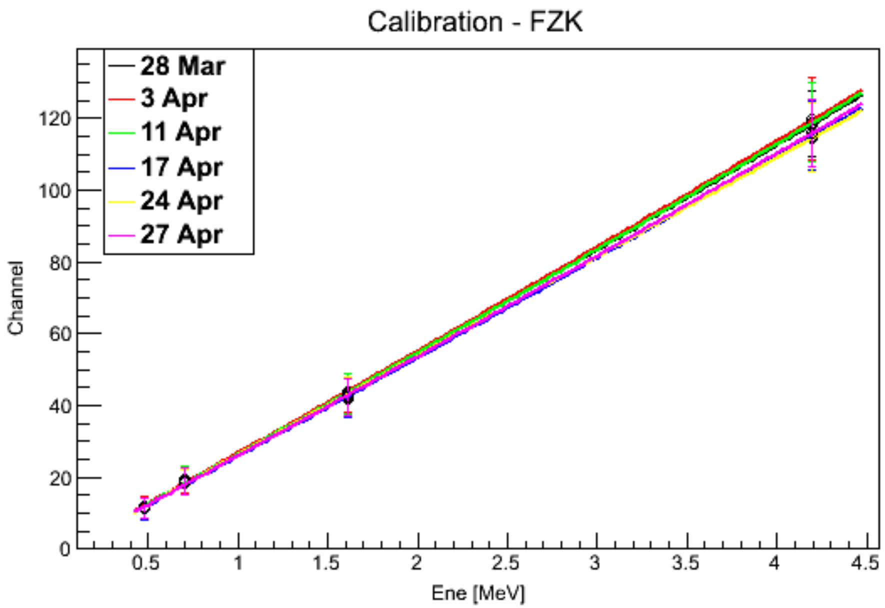
Calibrations



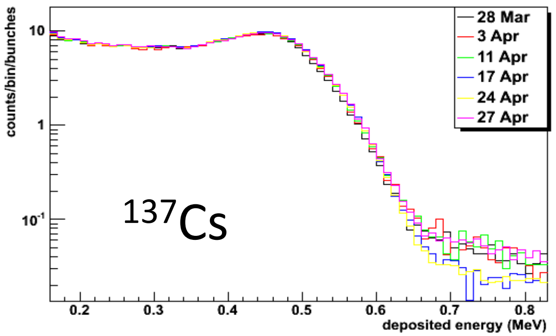


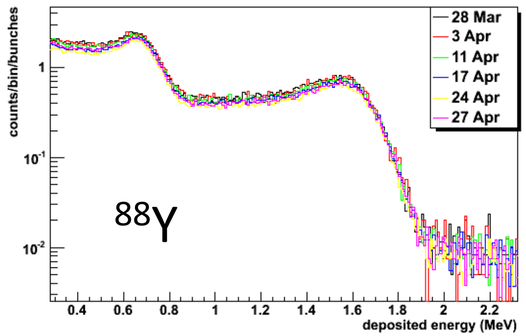


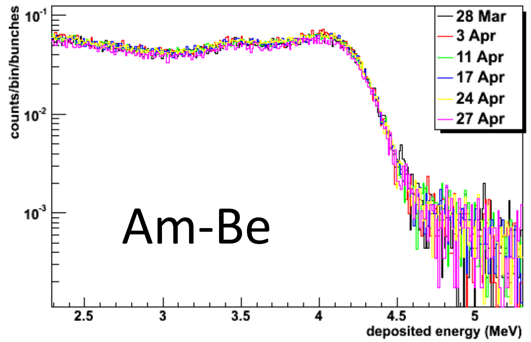


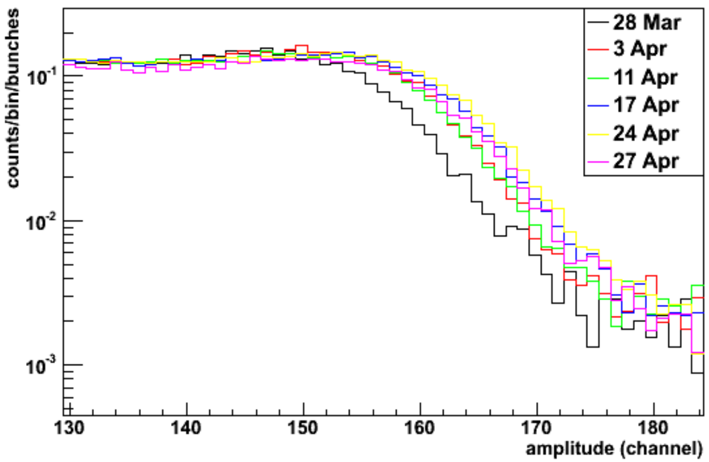


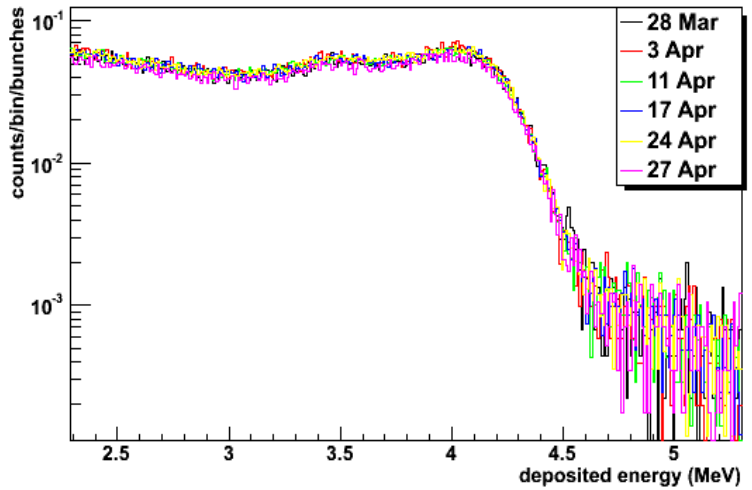
Calibrations



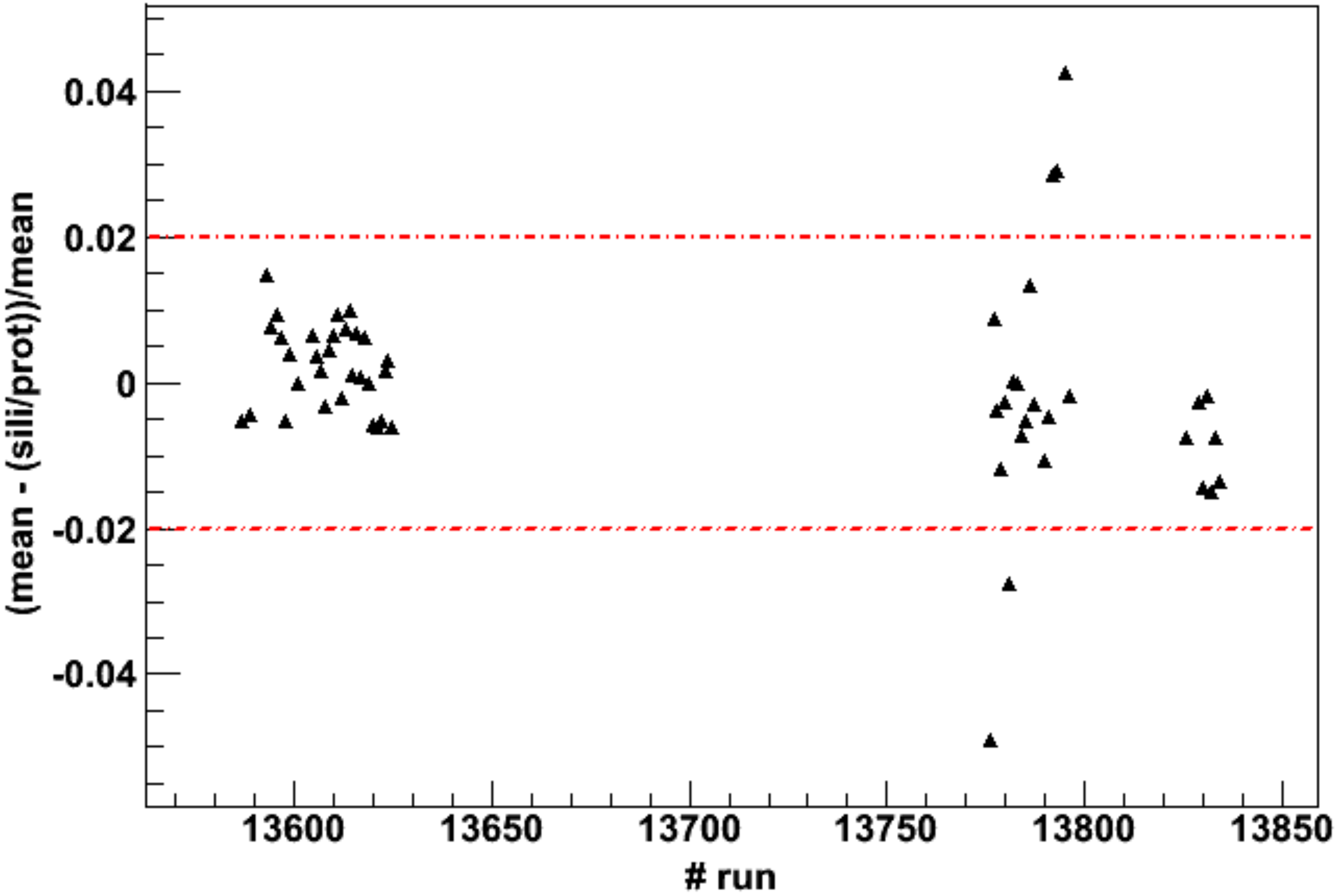




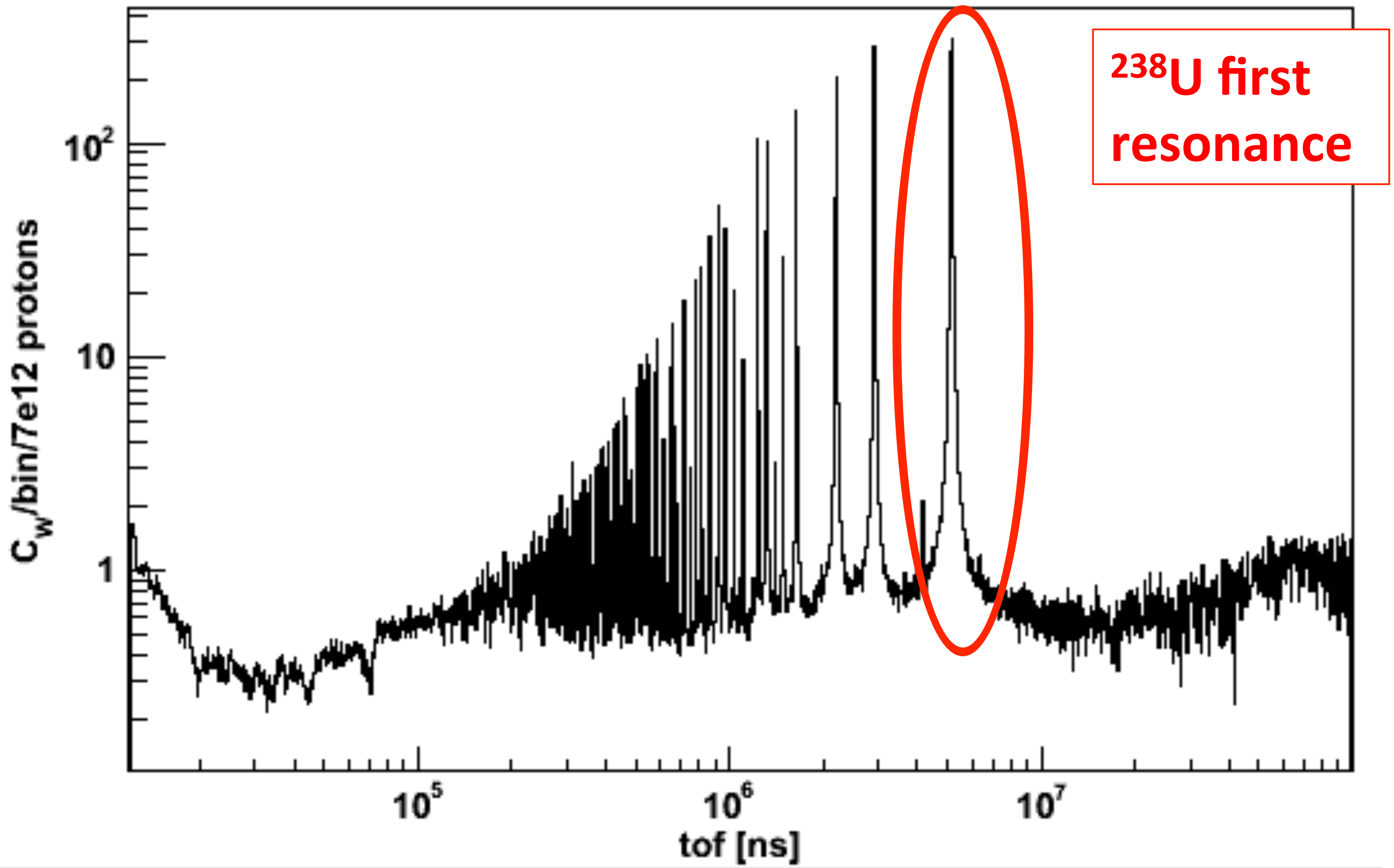




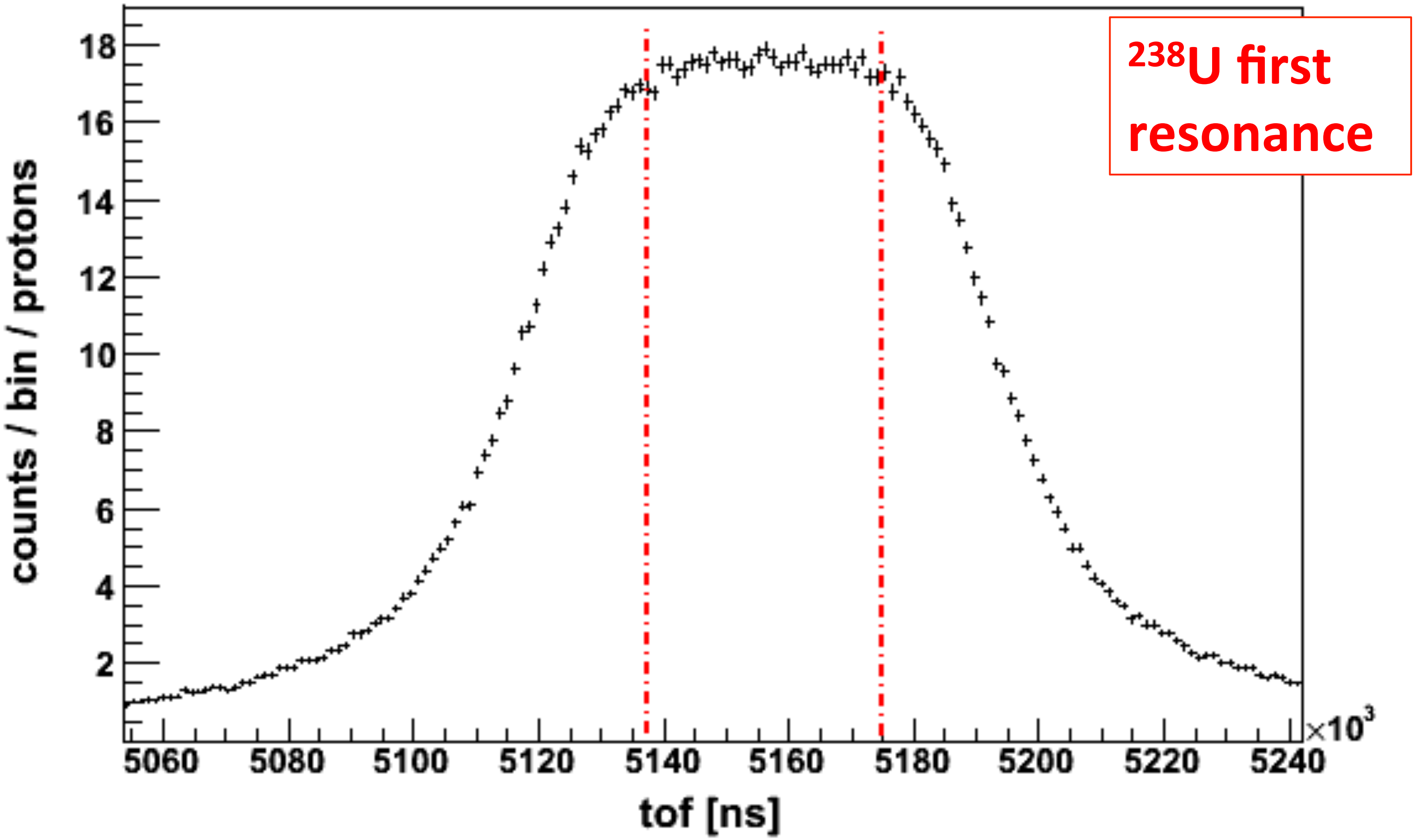
Flux & C6D6 stability



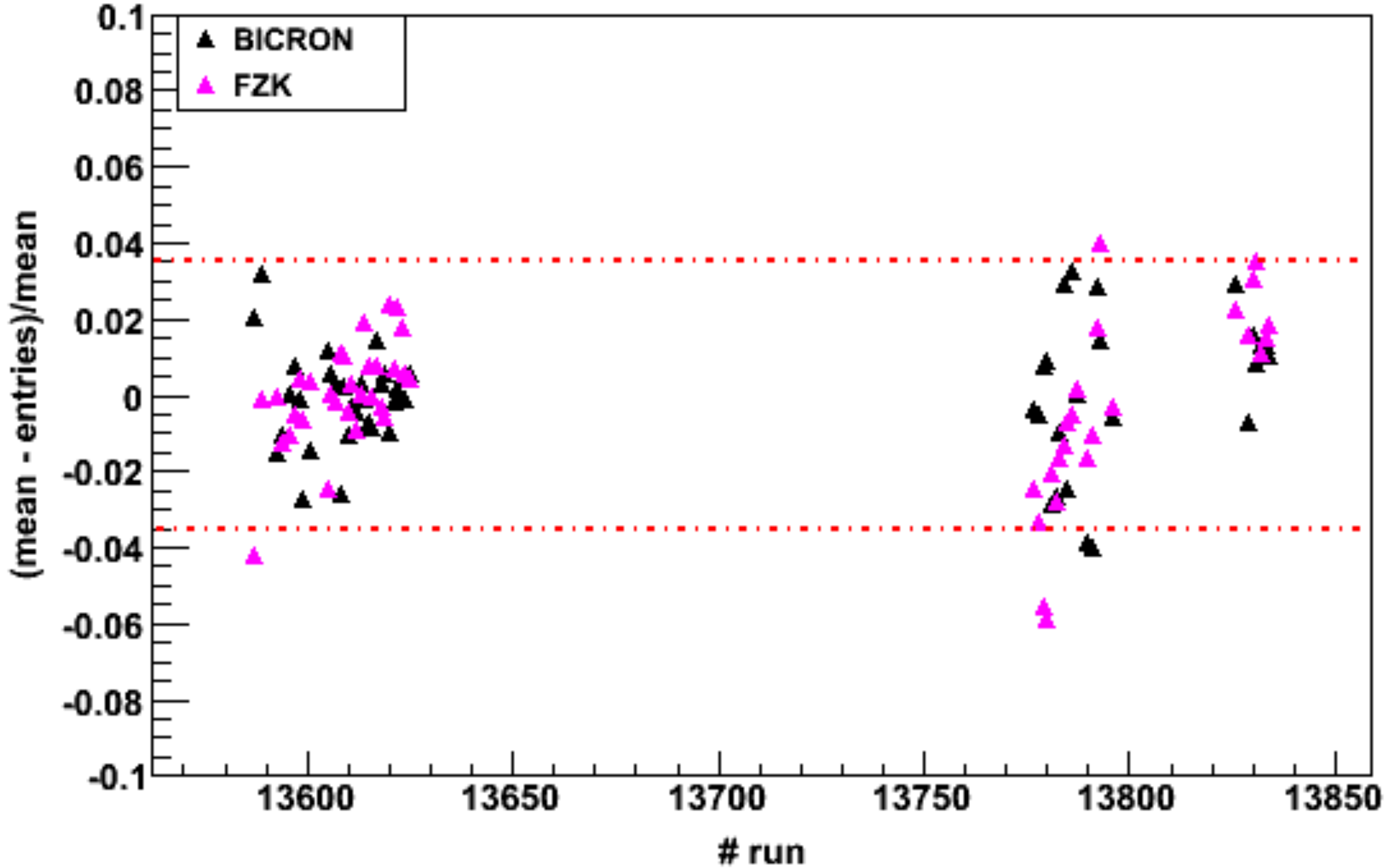
C_6D_6 counts stability



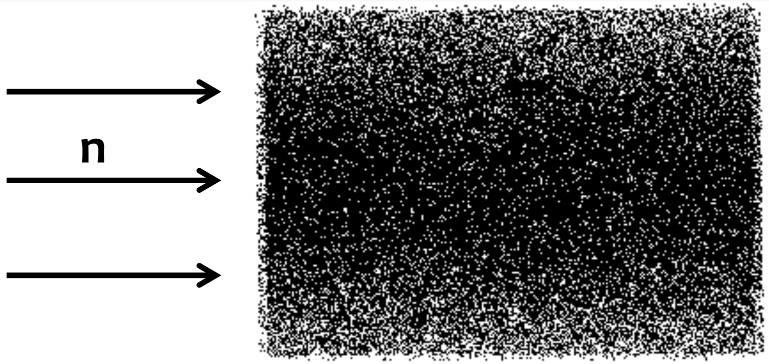
C_6D_6 counts stability

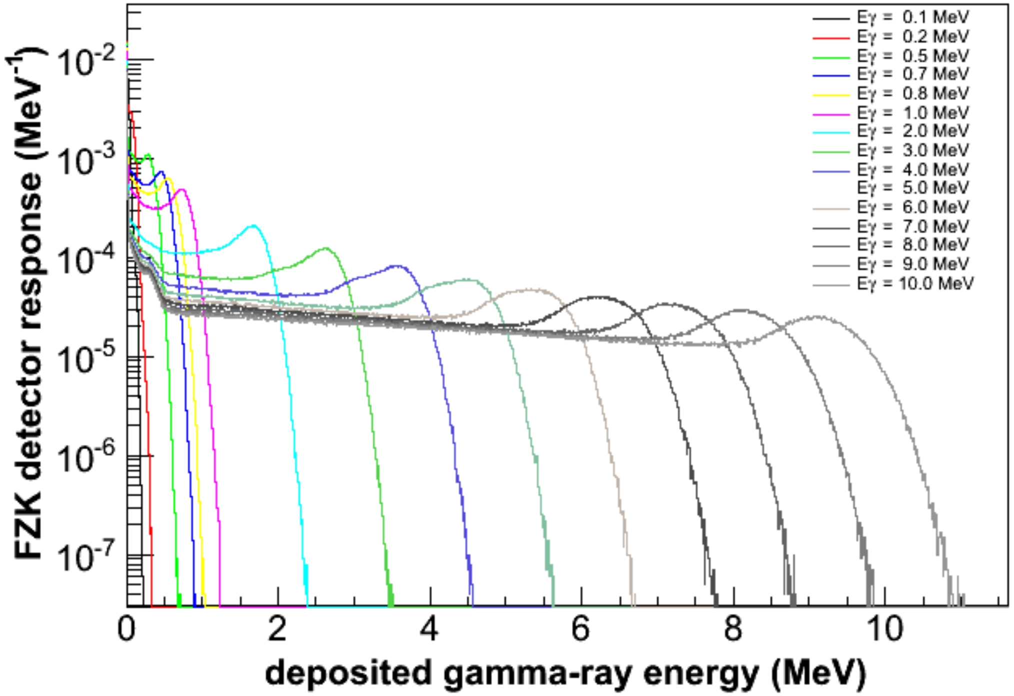


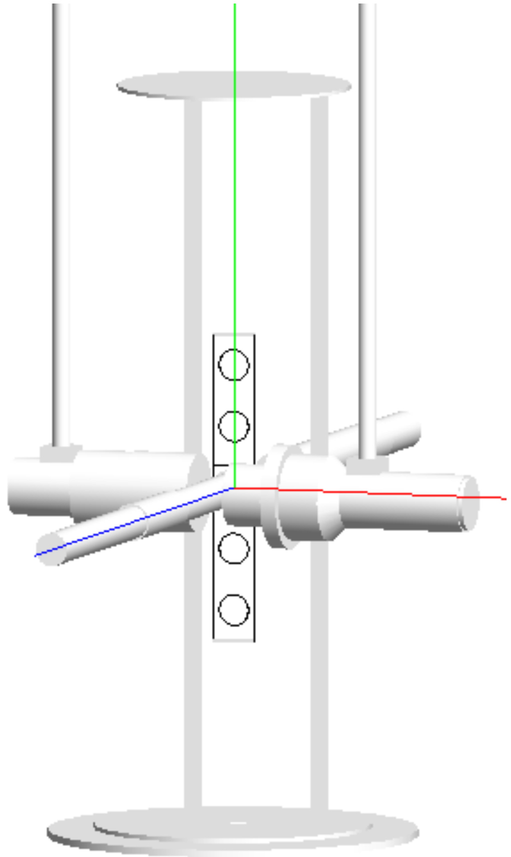
C_6D_6 counts stability



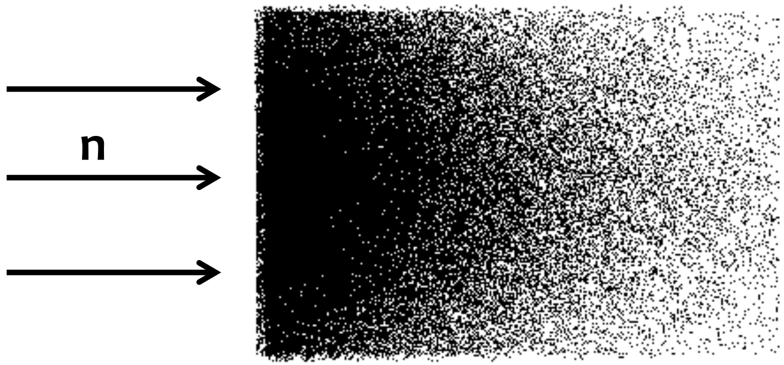
Simulated Detectors Response: Uniform emission

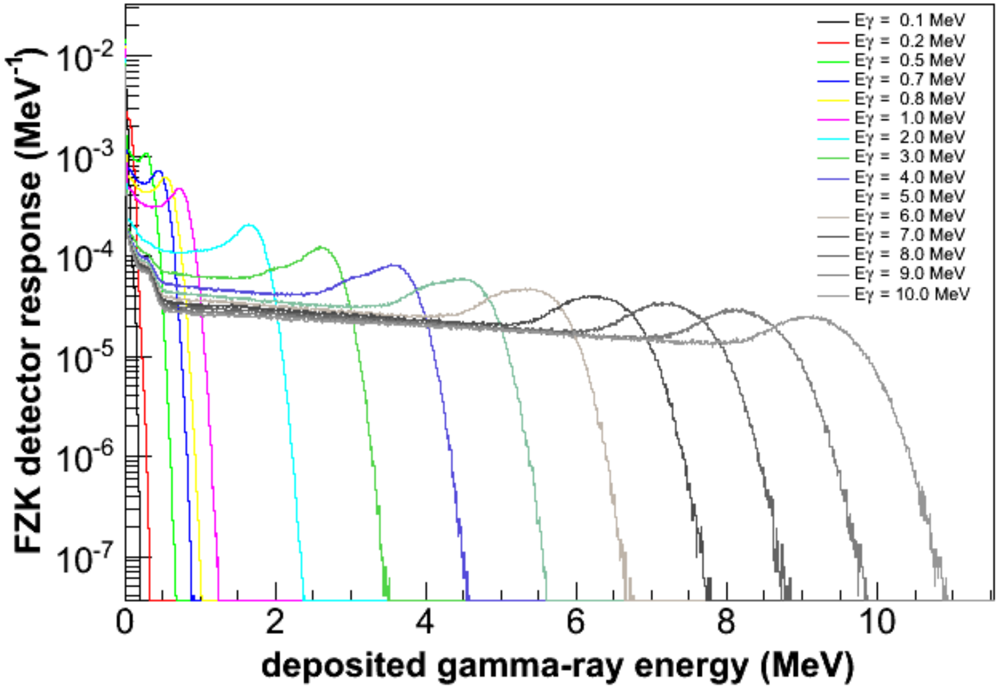


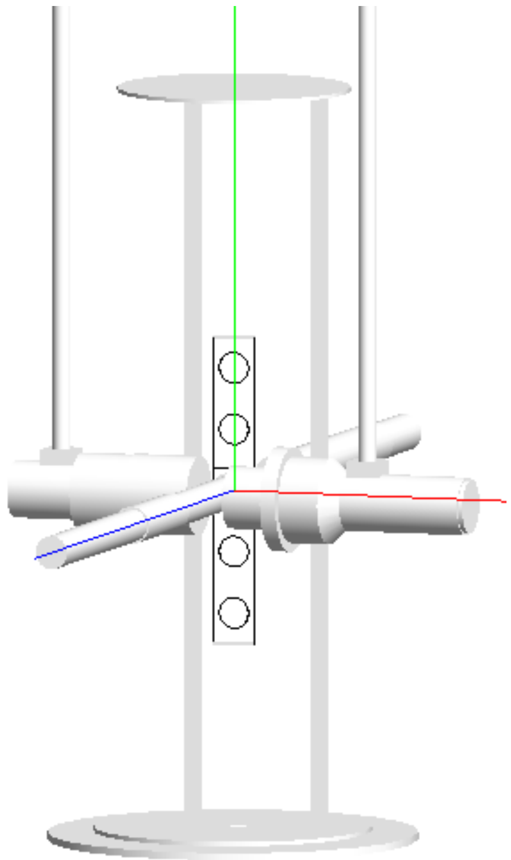




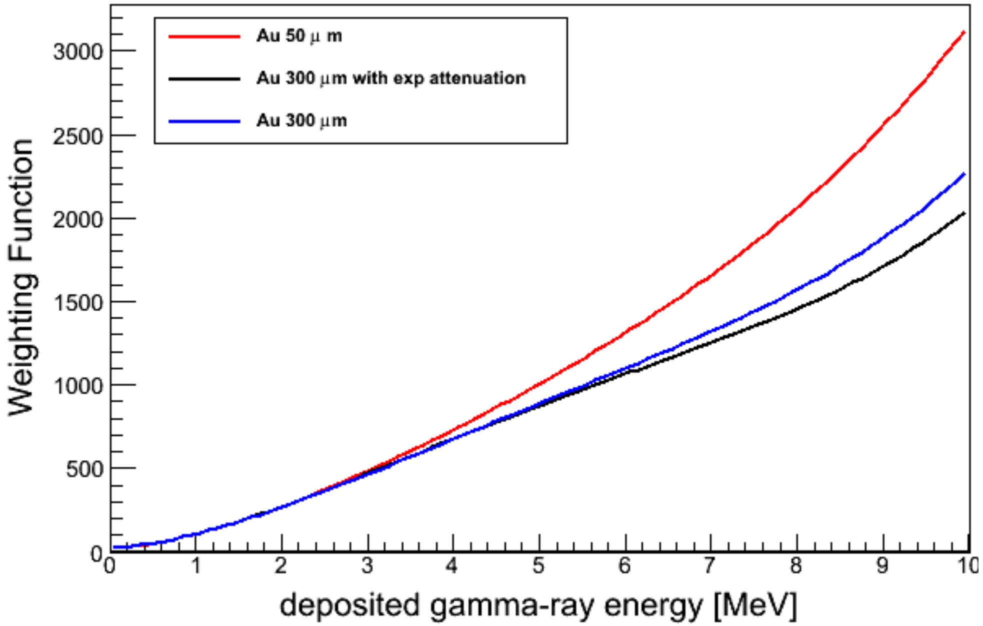
Simulated Detectors Response: Exponential attenuation





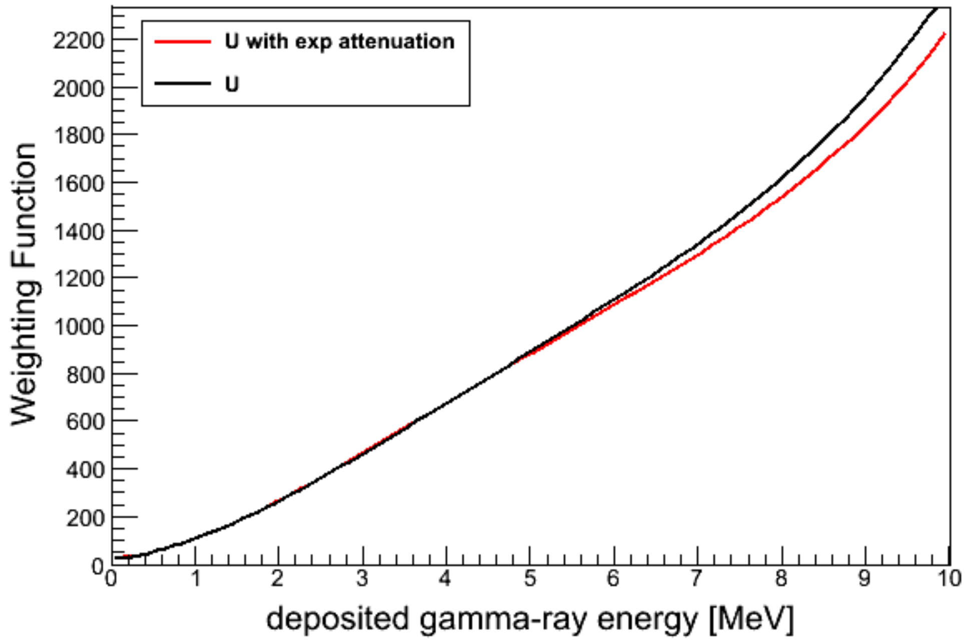


Weighting Functions

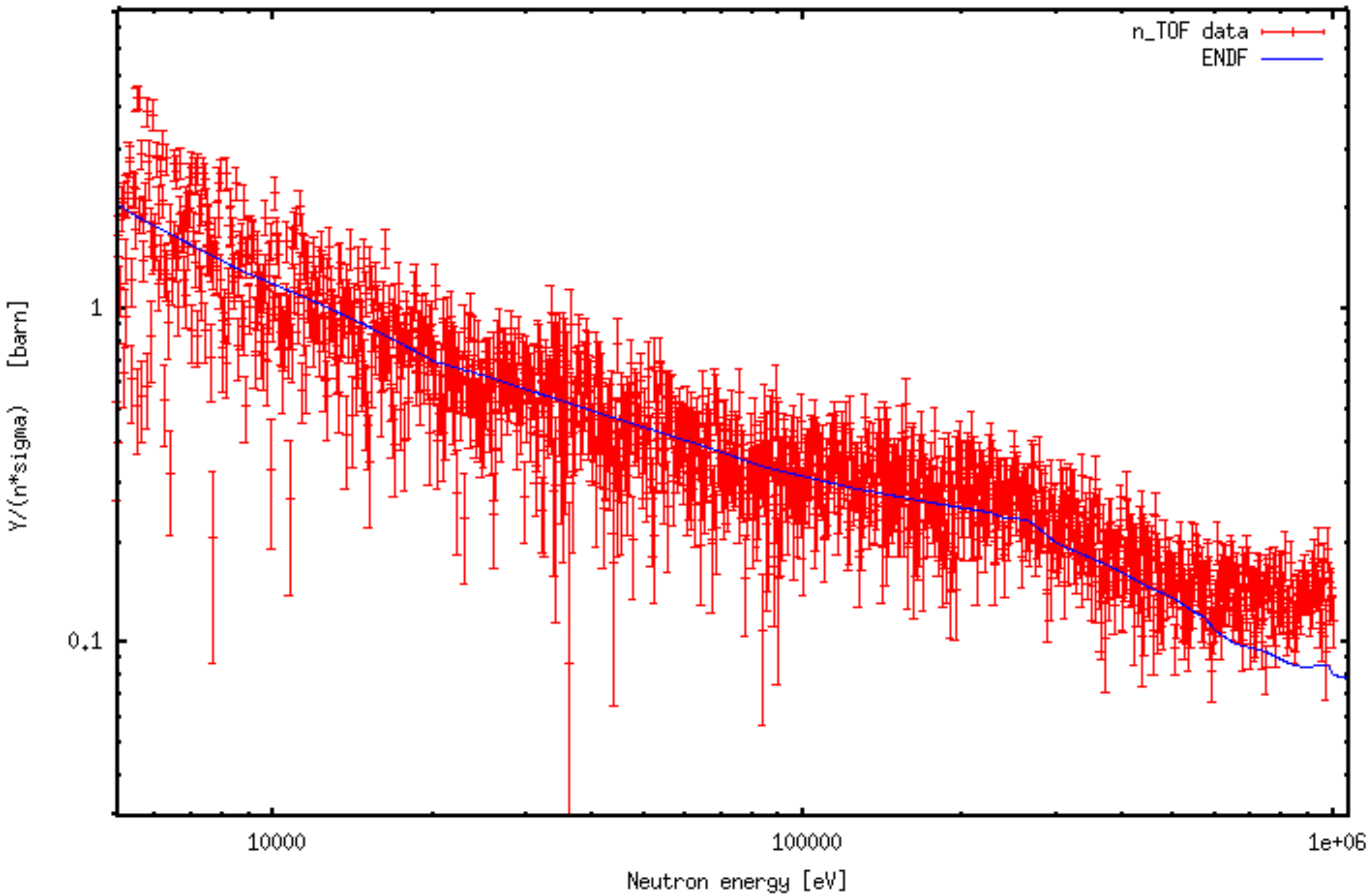


^{197}Au

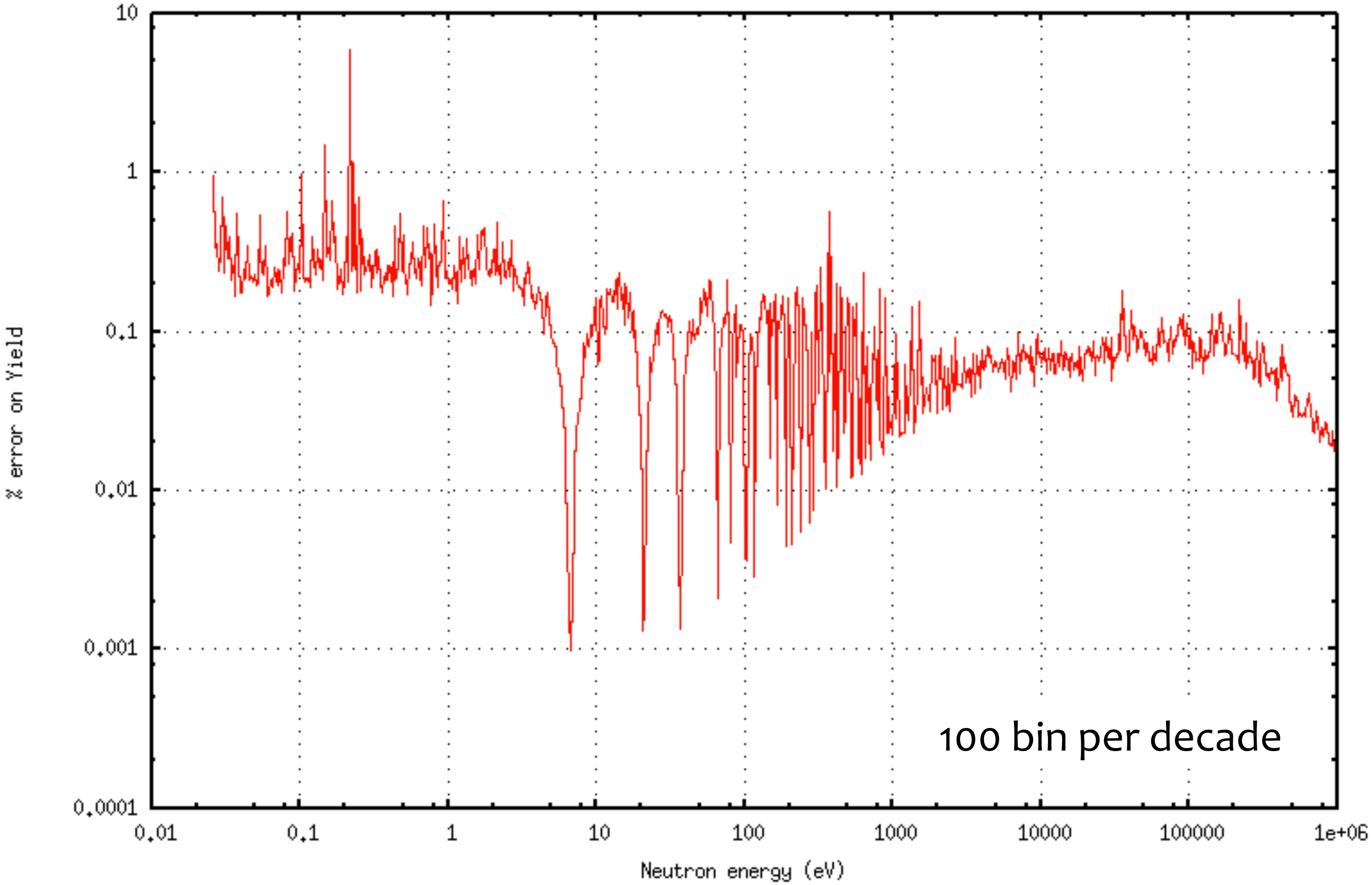
^{238}U



Unresolved Resonance Region: GOLD



Counting statistic uncertainties



Normalization: saturated resonance technique