



n_TOF

Measurement of the $^{236}\text{U}(n,f)$ cross-section at n_TOF

Raul Sarmento, ITN rsarmento@itn.pt

Instituto Tecnológico e Nuclear, Lisbon, Portugal

Marco Calviani, CERN Marco.Calviani@cern.ch

European Organization for Nuclear Research, Geneva, Switzerland

N. Colonna, I. Gonçalves, P. Vaz & the n_TOF Collaboration www.cern.ch/ntof

Summary

Motivation

The previous measurements
Status of the $^{236}\text{U}(n,f)$ cross-section evaluations

Experimental setup

The n_TOF spectrometer
The Fast Ionization Chamber
The samples

Data analysis

Event reconstruction
Energy calibration and the n_TOF flux
Background subtraction and dead-time correction

Results

The low energy cross-section
The resonance and intermediate resonance regions
The cross-section around the threshold energy

Conclusions

n_TOF

Motivation

The medium-term energy requirements at a global scale have brought renewed interest in the nuclear energy production.

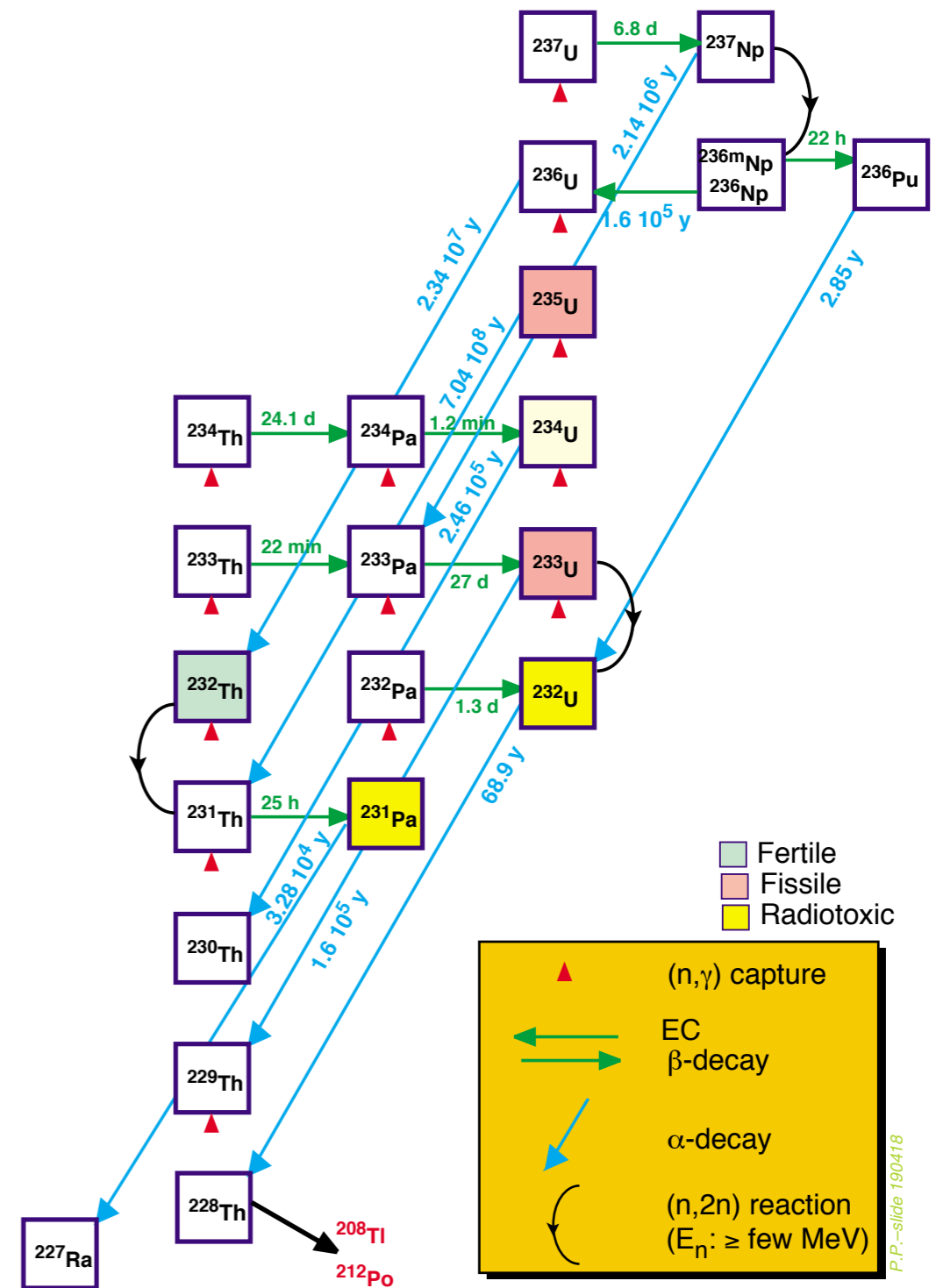
The known issues concerning the nuclear power production, such as the proliferation resistance, the operational safety and the minimization of radiotoxicity waste, need to be addressed.

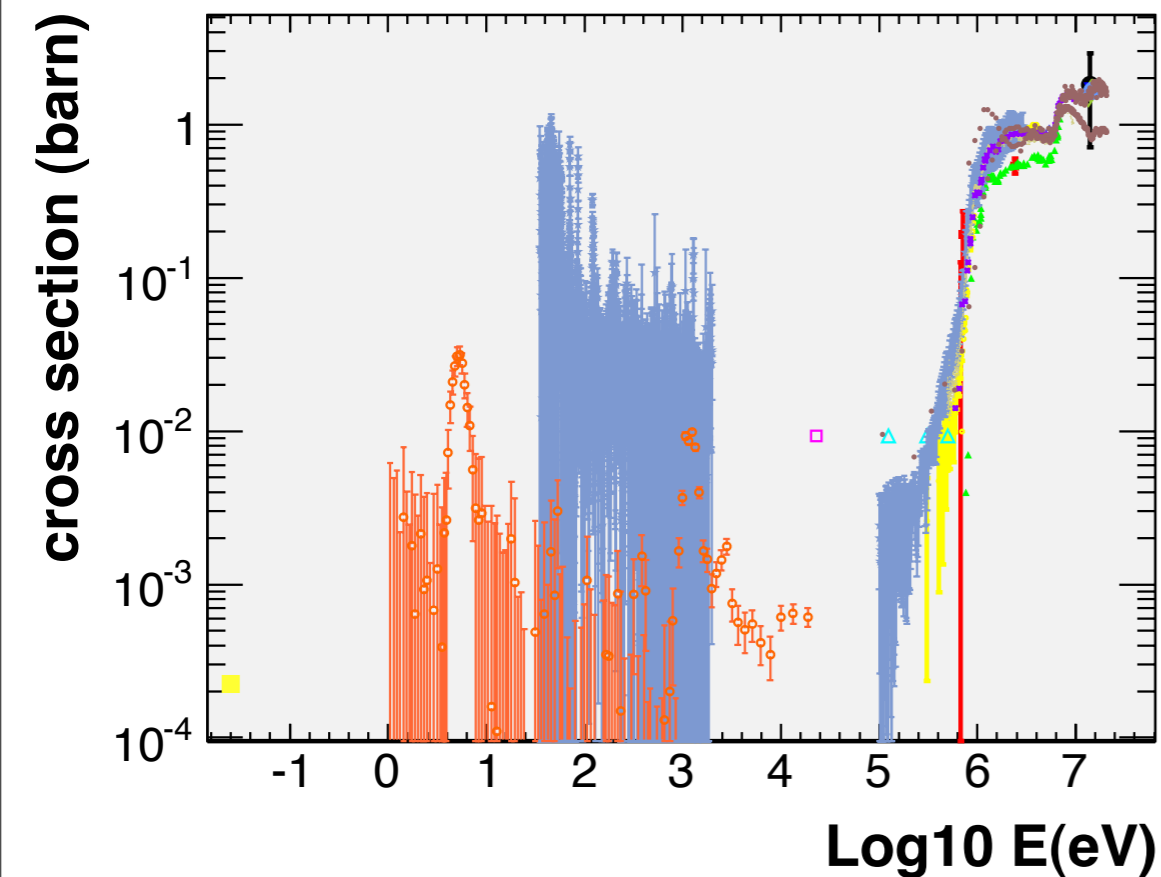
For that purpose, solutions based on ADS systems or Gen-IV reactors with the Th/U fuel cycle and transmutation have been proposed.

The ^{236}U builds-up in the equilibrium fuel composition of the Th/U cycle.

The International Atomic Energy Agency issued the demand for the knowledge of the $^{236}\text{U}(n,f)$ fission cross section data with 5% accuracy for the technology design and development.

The Th-U fuel cycle (figure extracted from Abbondanno et al. CERN-INTC-2001-025).





- W. Nyer (1950)
- W. Nyer (1951)
- R. Henkel (1952)
- J. Wahl, R. Davis (1954)
- R. Lamphere, R. Greene (1956)
- J. Perkin et al. (1965)
- P. White et al. (1965)
- P. White, G. Warner (1967)
- J. Cramer, D. Bergen (1970)
- H. Rosler et al. (1972)
- J. Meadows (1978)
- F. Manabe et al. (1988)
- J.W.MEADOWS (88)
- C.Wagemans et al. (2000)
- B. Lyles et al. (2007)
- C.Wagemans et al. (2008)
- A. Alekseev (2008)

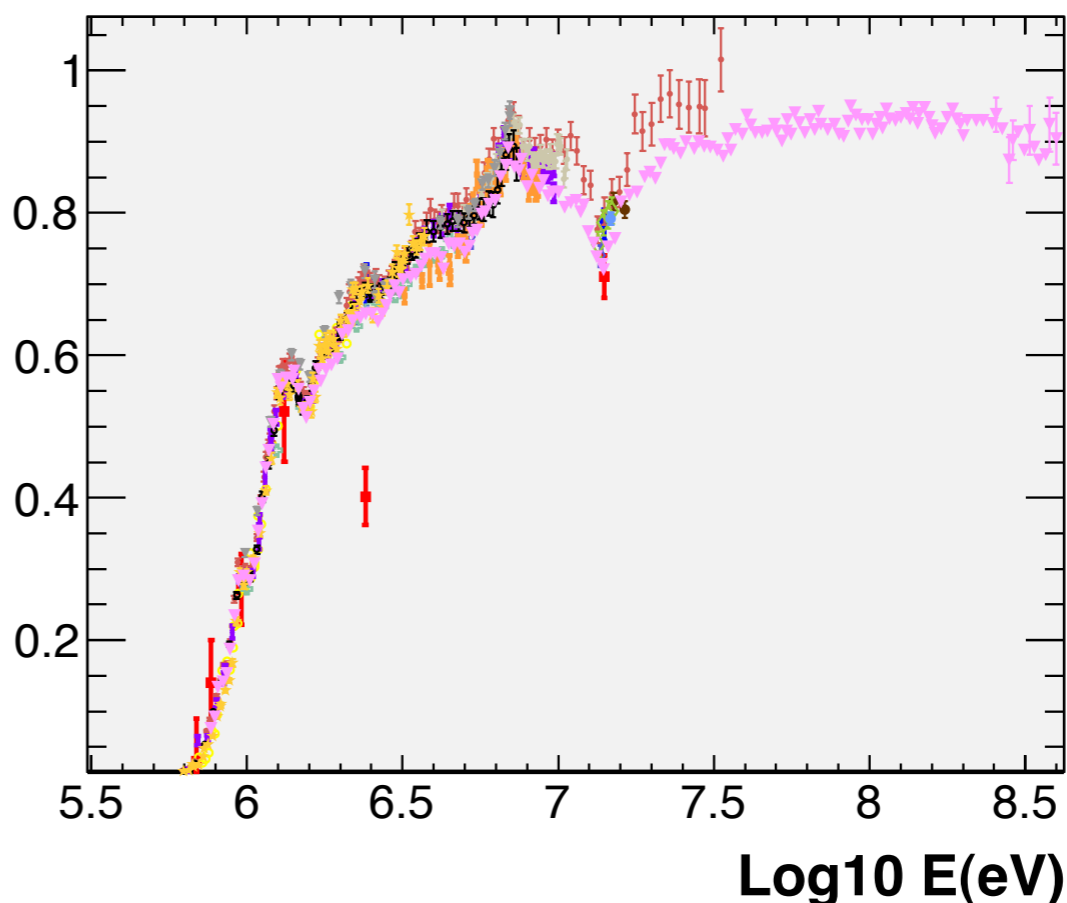


The experimental data on the $^{236}\text{U}(n,f)$ cross-section extracted from the Experimental Nuclear Reaction Data (EXFOR)

The experimental data on the ratio between the $^{236}\text{U}(n,f)$ and the $^{235}\text{U}(n,f)$ cross-sections extracted from the Experimental Nuclear Reaction Data (EXFOR)



$$\sigma(\text{U-236}) / \sigma(\text{U-235})$$



- W. Nyer (1951)
- J. Wahl, R. Davis (1954)
- R. Lamphere, R. Greene (1956)
- P. White et al. (1965)
- P. White, G. Warner (1967)
- W. Stein et al. (1968)
- J.W. Behrens, G. Carlson (1977)
- J. Meadows (1978)
- C. Nordborg et al. (1978)
- A. Goverdovskii et al. (1985)
- B. Fursov, et al. (1985)
- A. Goverdovskii (1986)
- H. Terayama et al. (1986)
- Y.Watanabe,ET.AL. (87)
- F. Manabe et al. (1988)
- J. Meadows (1988)
- Y.Watanabe,ET.AL. (87)
- D. Shpak et al. (1990)
- P. Lisowski et al. (1991)



The $^{236}\text{U}(n,f)$ cross-section resonances observed and presented in the literature.

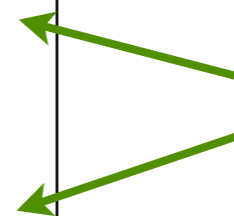
1972 - Theobald et al. in Geel
(the uncertainty in the widths of the resonances is 2.5% of the width value)

1994 - Parker et al. at LANSCE

2008 - Wagemans et al. at GELINA

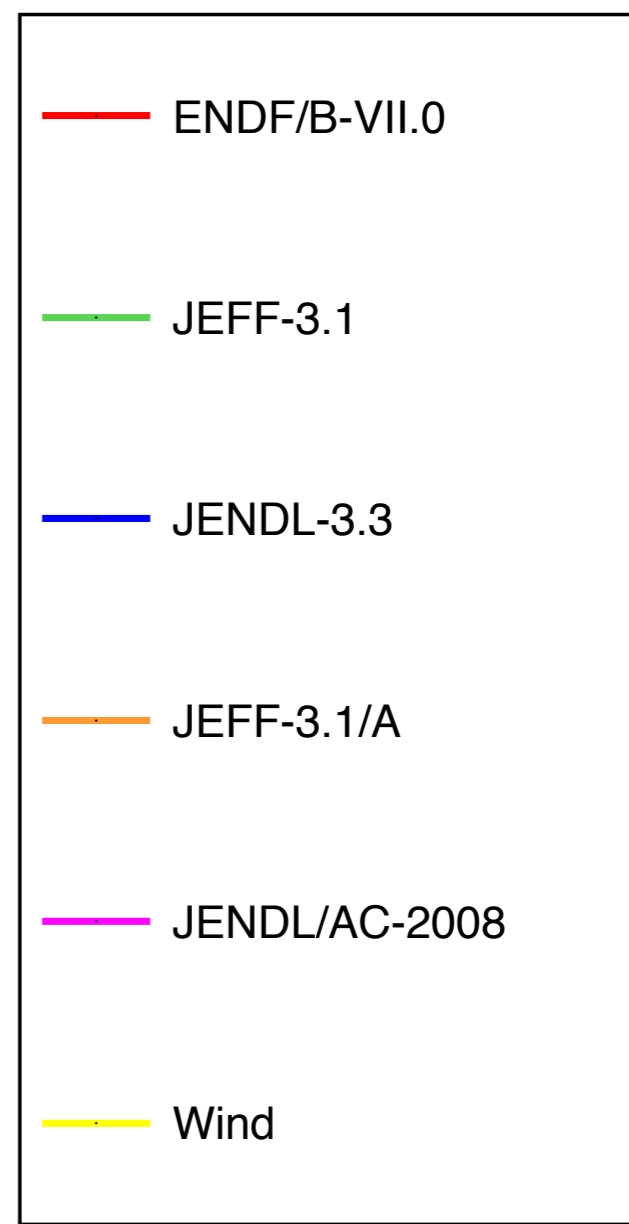
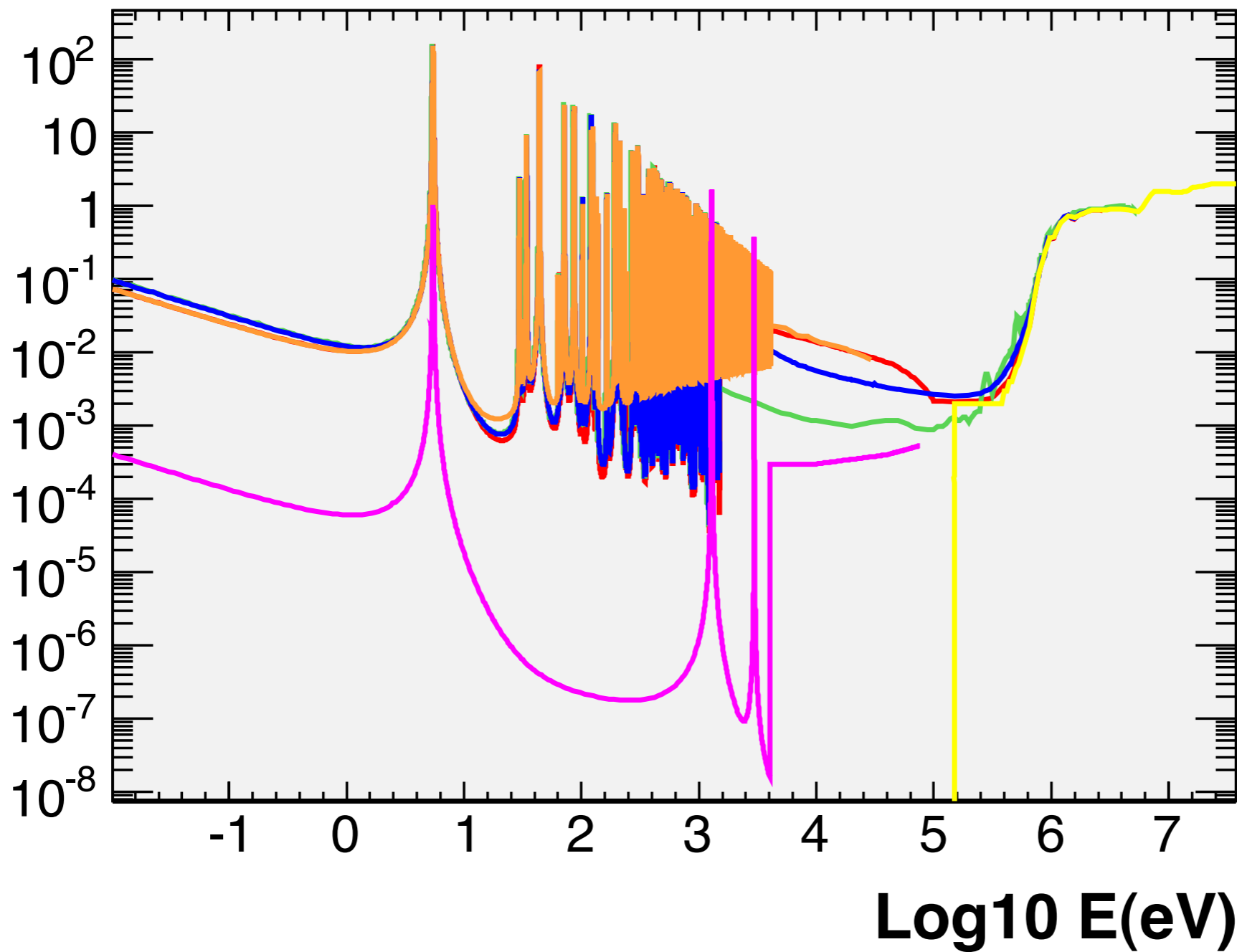
*these resonances were also observed, although no resonance parameters were given

ENERGY EV	WIDTH MILI-EV	WIDTH-ERR MILI-EV	
5.4500E+00	2.9000E-01		
5.4500E+00	1.3000E-03	1.0E-04	
5.4500E+00	1.7000E-03	1.0E-04	
2.9900E+01	1.6000E-01		
3.4000E+01	1.8000E-01		
4.3700E+01	4.3000E-01		
7.1100E+01	2.9000E-01		
8.6400E+01	3.0000E-01		
1.2080E+02	3.4000E-01		
1.2470E+02	2.1000E-01		
1.9400E+02	5.0000E-01		
2.1400E+02	3.2000E-01		
2.7240E+02	4.0000E-01		
2.8820E+02	4.8000E-01		
3.0250E+02	4.6000E-01		
3.7100E+02	4.2000E-01		
3.7900E+02	3.0000E-01		
4.1500E+02	5.9000E-01		
1.2688E+03	8.2000E-01	3.0E-01	*
1.2817E+03	7.7000E+00	5.0E+00	*
1.2917E+03	9.3000E-01	1.1E-01	*
2.9589E+03	1.4000E+00	6.0E-01	
6.3000E+03	1.0800E+01	6.0E+00	*
1.0400E+04	4.6000E+00	2.6E+00	*



The evaluated data on the $^{236}\text{U}(n,f)$ cross-section
extracted from the Evaluated Nuclear Data File (ENDF)

cross-section (barn)



Summary of the shortcomings attributed to the $^{236}\text{U}(n,f)$ cross-section evaluated data which are inconsistent with the present demands of nuclear data for the design of ADS systems

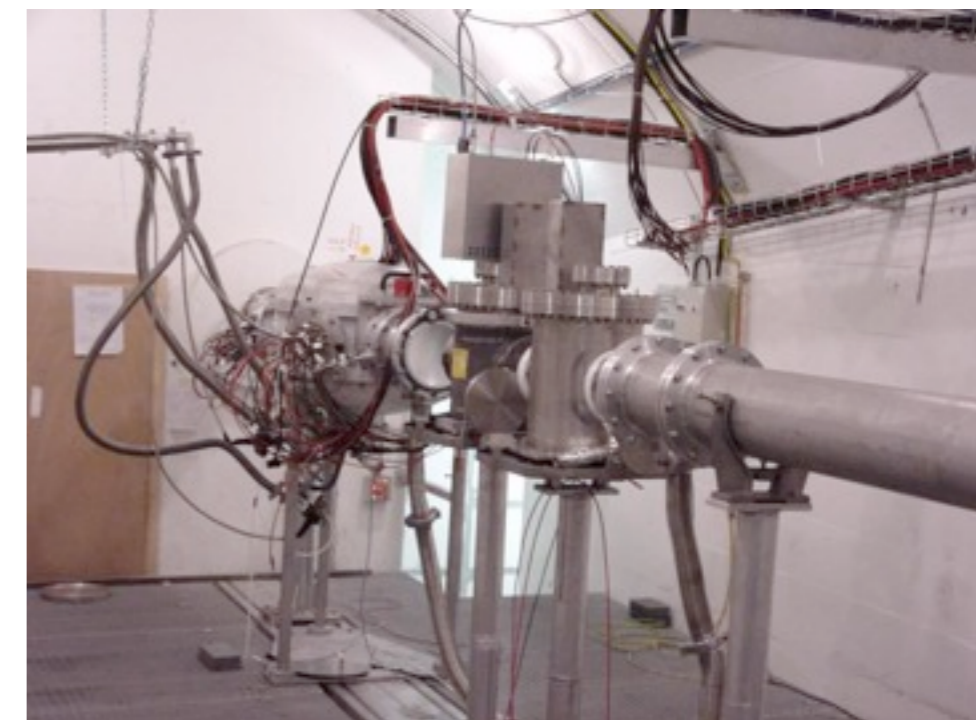
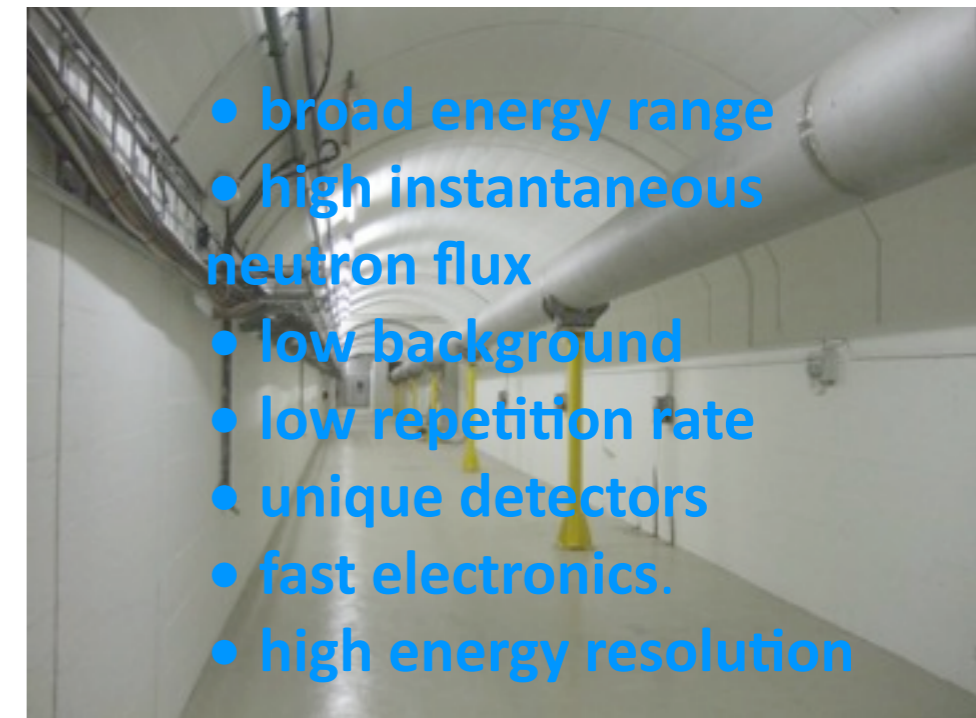
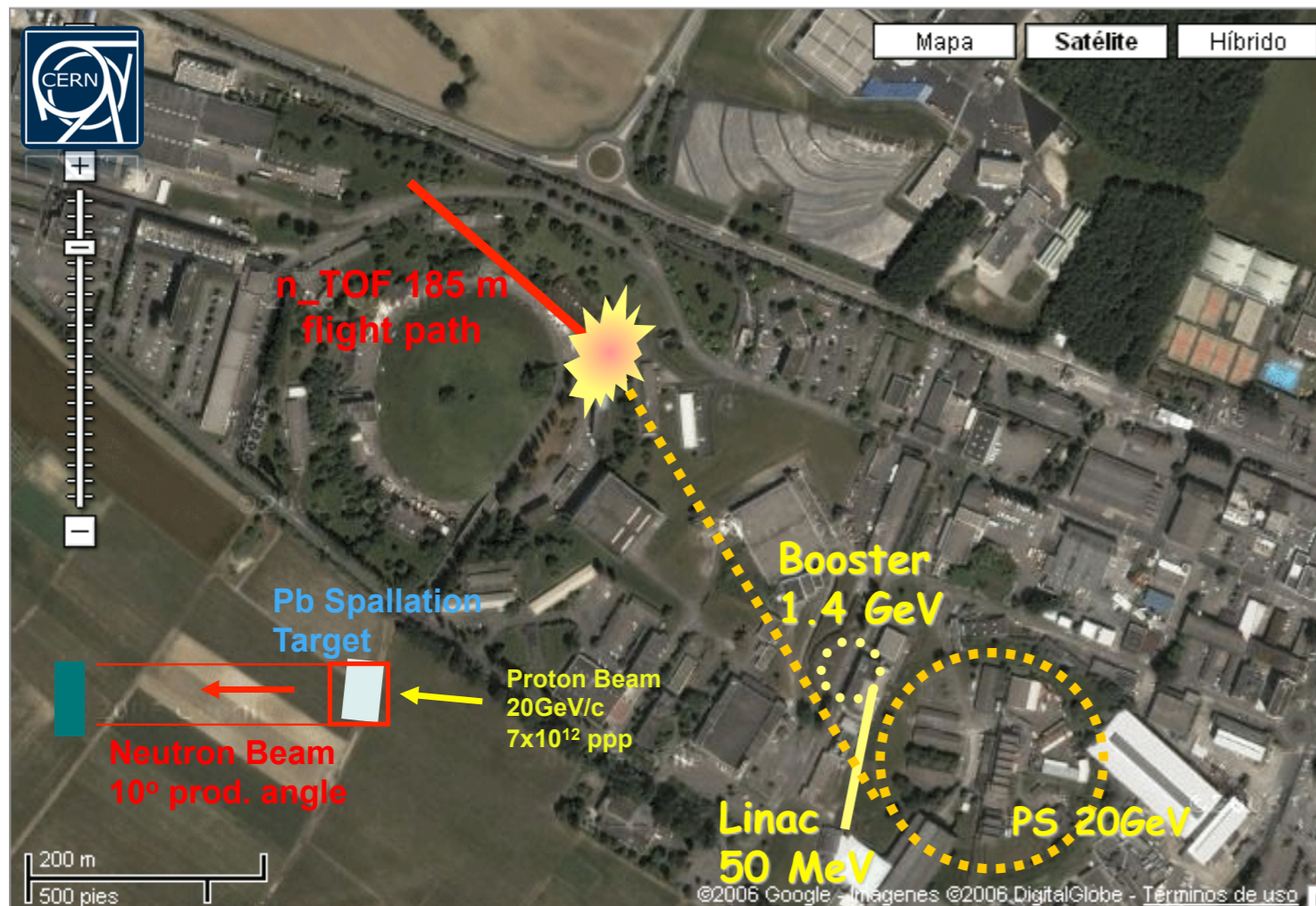
- 1. The thermal cross-section - overestimation by two orders-of-magnitude**
- 2. The 5.45 eV resonance width - overestimation by two orders-of-magnitude**
- 3. The resonance region is filled with false ^{236}U resonances**
- 4. Intermediate energy resonances are missing**
- 5. No agreement on the absolute value up to few hundred keVs**

n_TOF

The n_TOF spectrometer and the experimental setup

The n_TOF facility at CERN is a spallation target facility to measure neutron capture and fission cross-sections by the time-of-flight method.

Characteristics of the facility:



- ➔ Beams of protons were accelerated to 20 GeV by the PS proton synchrotron.
- ➔ A 60x60x80 cm³ Pb block was used as the spallation target.
- ➔ Neutron beams go through the 185 m long flight tunnel - the neutron tube which has the collimators - to reach the experimental area.

Measurement of the $^{236}\text{U}(n,f)$ cross-section at n_TOF
rsarmento@itn.pt & Marco.Calviani@cern.ch on behalf of the n_TOF Collaboration

The ^{236}U fission data taking - experimental conditions:

The collimation system was adjusted to the fission setup - the diameter of the beam at the entrance of the experimental area was 8 cm. The neutron flux at the experimental area was $10\text{E}+05$ neutrons/cm² ppp.

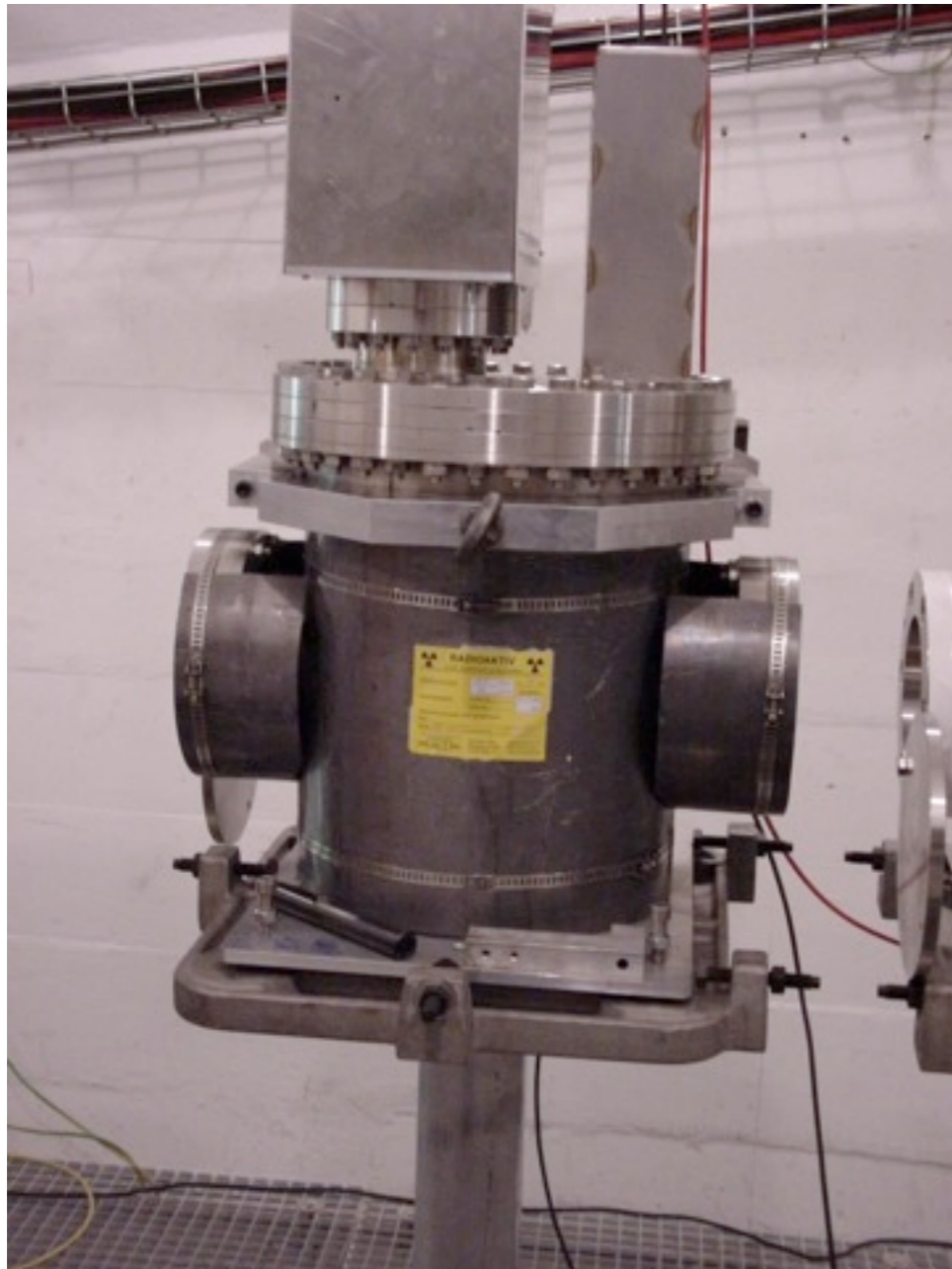
The experimental area was mounted with the fission setup, consisting of a fast ionization chamber containing ^{236}U and ^{235}U samples.

The signal was amplified by a Current Feedback Operational Amplifier AD844 and was digitized with Flash Analog-to-Digital Converter channels with 8-bit resolution working at a 250 MHz sampling rate.

A total of $1.56\text{E}+18$ protons from dedicated PS beams generating $2.30\text{E}+05$ spallation events plus $4.52\text{E}+17$ protons from parasitic PS beams generating $1.76\text{E}+05$ spallation events was used for the measurement.

A run with no neutron beam was also taken to obtain data for background corrections.

The detector used for the measurement was a Fast Ionization Chamber - **FIC**



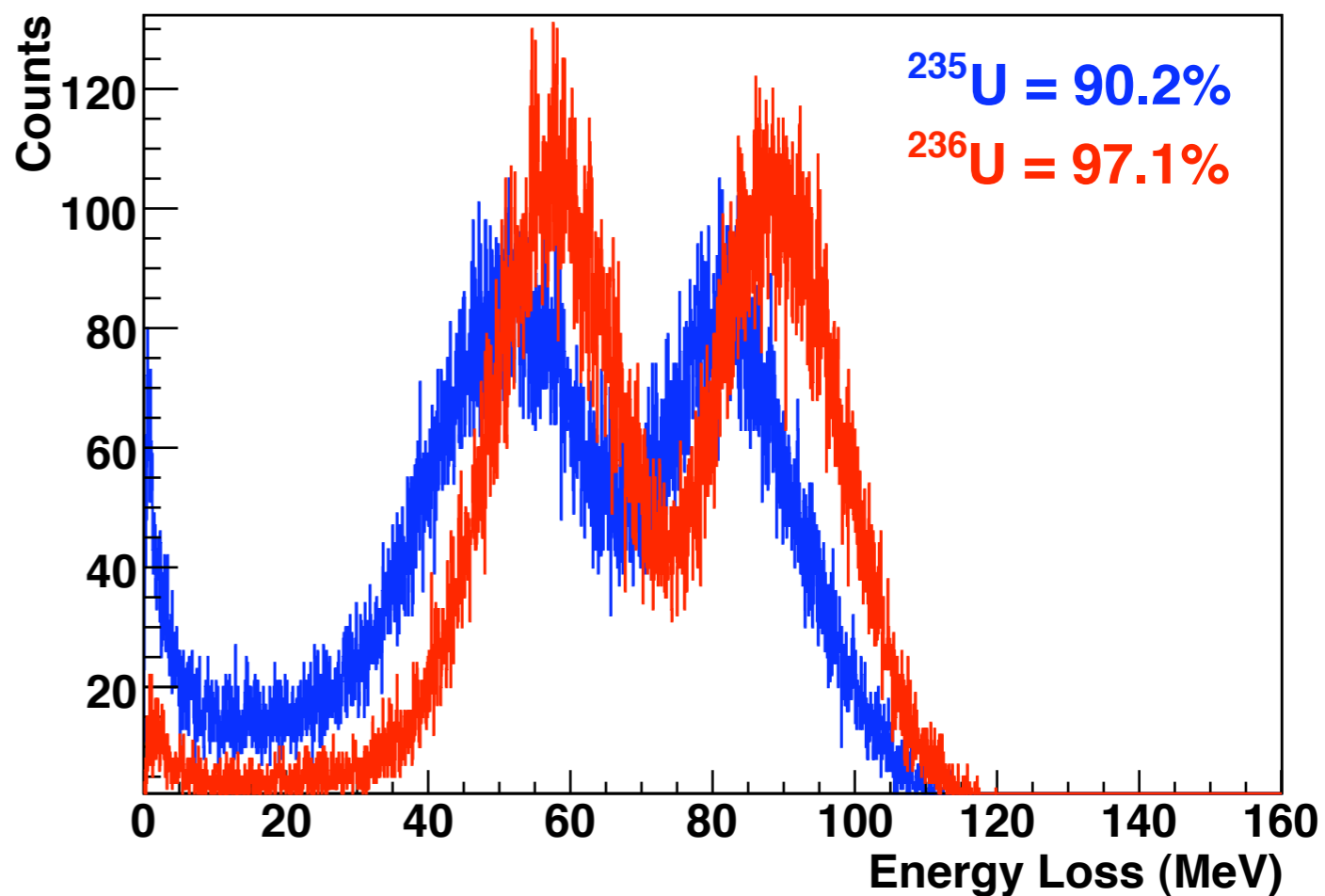
- ➔ The ionization chamber operates in the ion saturation region - no avalanche multiplication occurs.
- ➔ The fission cross-section is measured by detecting the fission fragments (FF) - electrons and ion pairs are produced in the gas by the FF and this charge is collected by applying a voltage between the electrodes.
- ➔ One single fragment is detected per fission event - 2π detection efficiency. The other fission fragment is absorbed in the Al electrode backing the sample to measure.
- ➔ The gas was chosen by its fast timing properties to avoid pile-up problems.

Complementary information on the FIC detector.

Gas	90% Ar + 10% CF₄
Gas pressure	720 mbar
Gap between the electrodes	20 mm
Electric field	550 V/cm
Electron drift velocity	12 cm/μs

The intrinsic **detection efficiency** was calculated by performing computational simulations with FLUKA. The values were obtained for setting a threshold on the energy deposited by the fission fragments at 35 MeV.

Energy Loss FF



^{236}U samples information

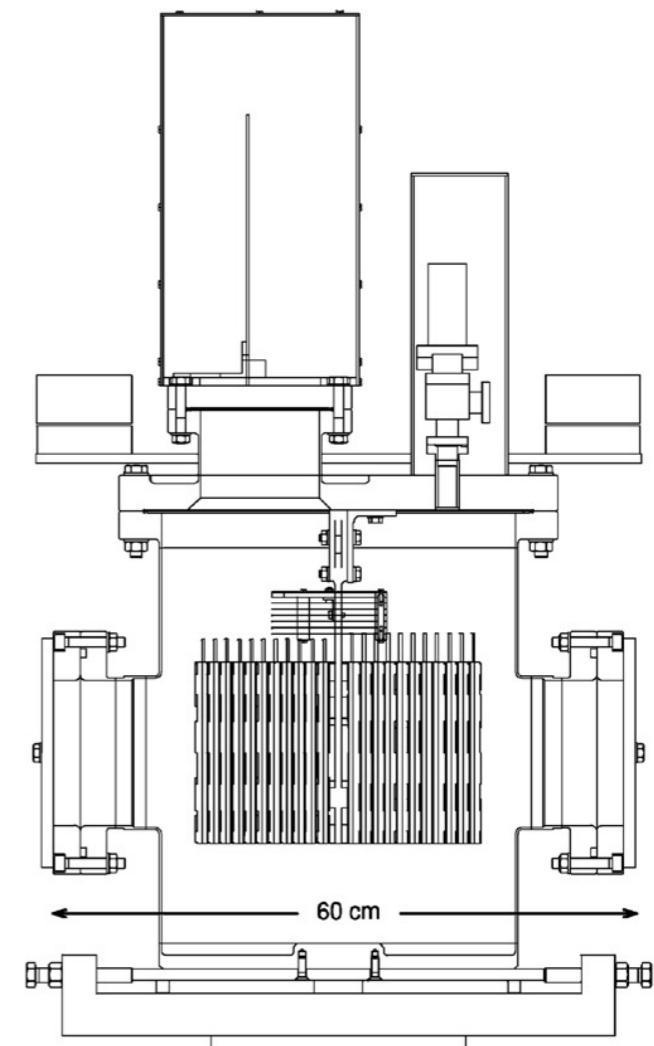
Isotopic composition (mass %)	U-235	0.05
	U-236	99.85
	U-238	0.1
Mass (mg) sample 1 sample 2	$11.1 \pm 1.4\%$	
	$10.2 \pm 1.4\%$	
Diameter (mm)	80	
Activity of alpha-radiation at 04/02/04 (mCi)	$2.6\text{E-}02$	

The samples were produced by the painting technique. The ^{236}U samples thicknesses are $5.66\text{E-}07$ atoms/barn and $5.18\text{E-}07$ atoms/barn respectively.

The samples were placed at a 186.4 m flight-path.

A stack of cells mounted along the beam allows the simultaneous measurement of the multiple samples.

The cross-section was measured relative to the reference $^{235}\text{U}(n,f)$ cross-section, known as standard, using simultaneously a $35.6 \pm 1.4\%$ mg and 80 mm diameter sample.

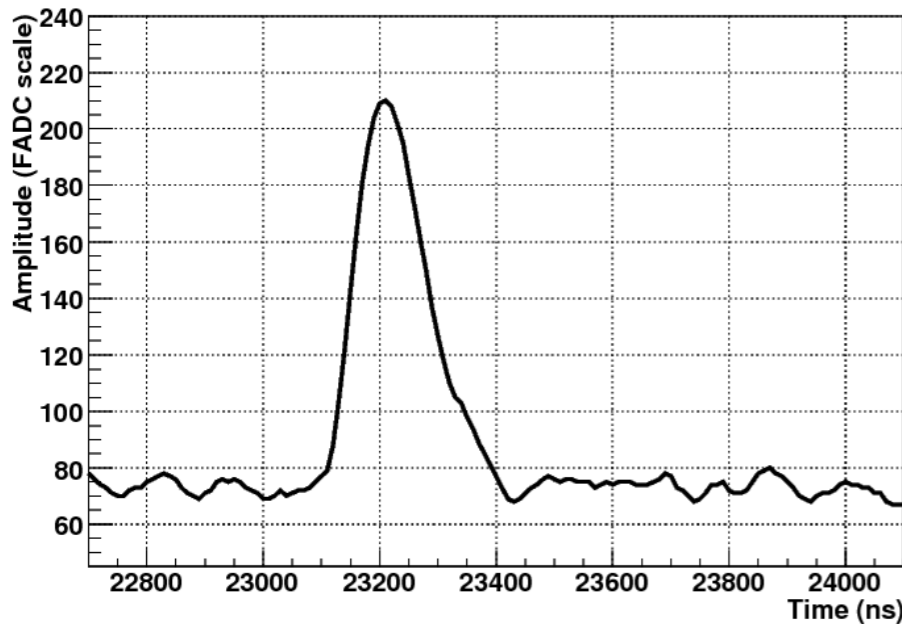


n_TOF

The data analysis

The event reconstruction

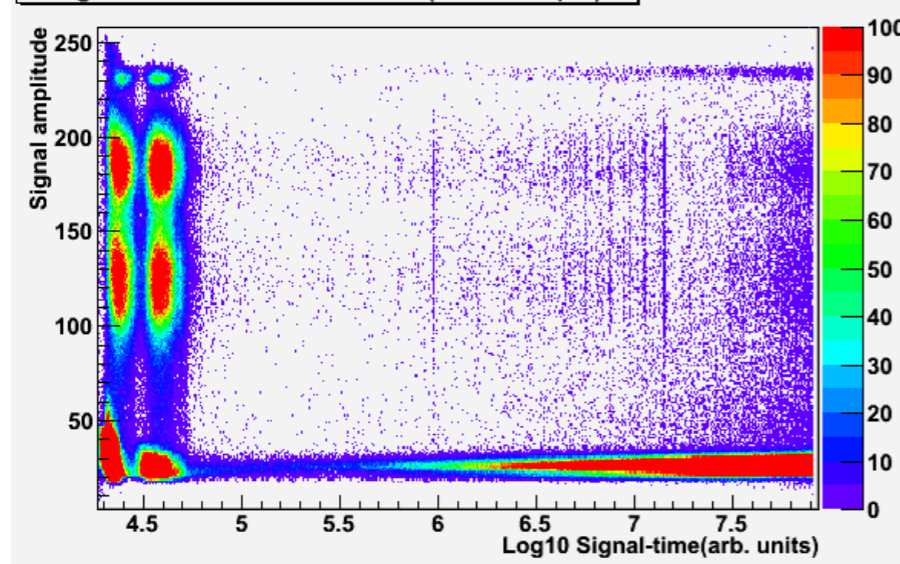
The digitized waveform of a typical FF signal.



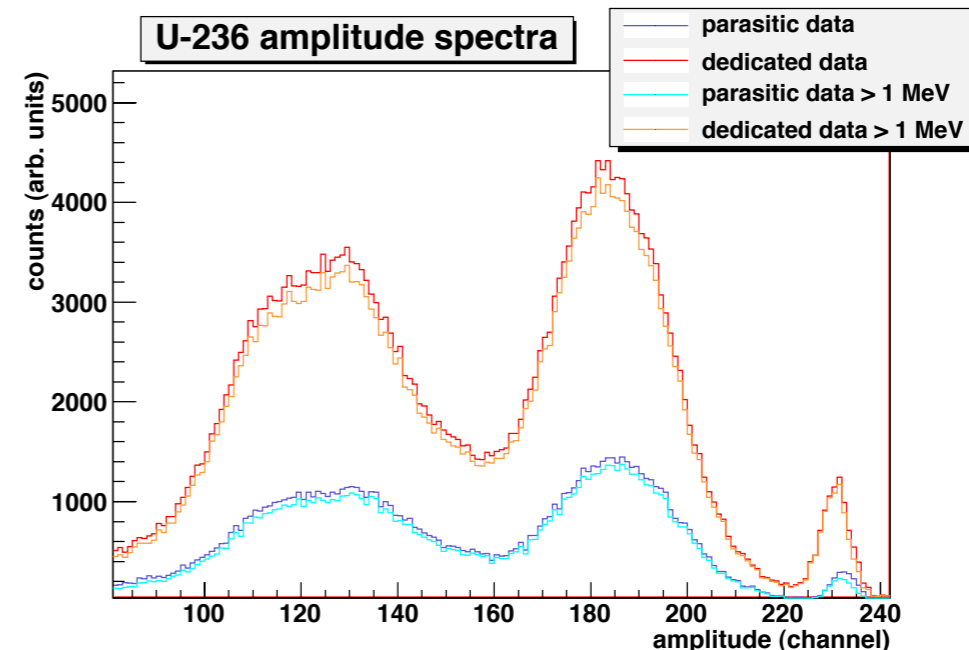
The signal reconstruction from the raw data was made by applying C++ routines developed using the ROOT framework and based on the Advanced Spectra Processing Function class TSpectrum.

Dedicated files are created storing the **amplitude, baseline, time information and area** of the signal peaks, as well as the **time and intensity of the proton bunch** originating the respective spallation neutrons.

Signal counts from detector 1 (236U sample)



U-236 amplitude spectra



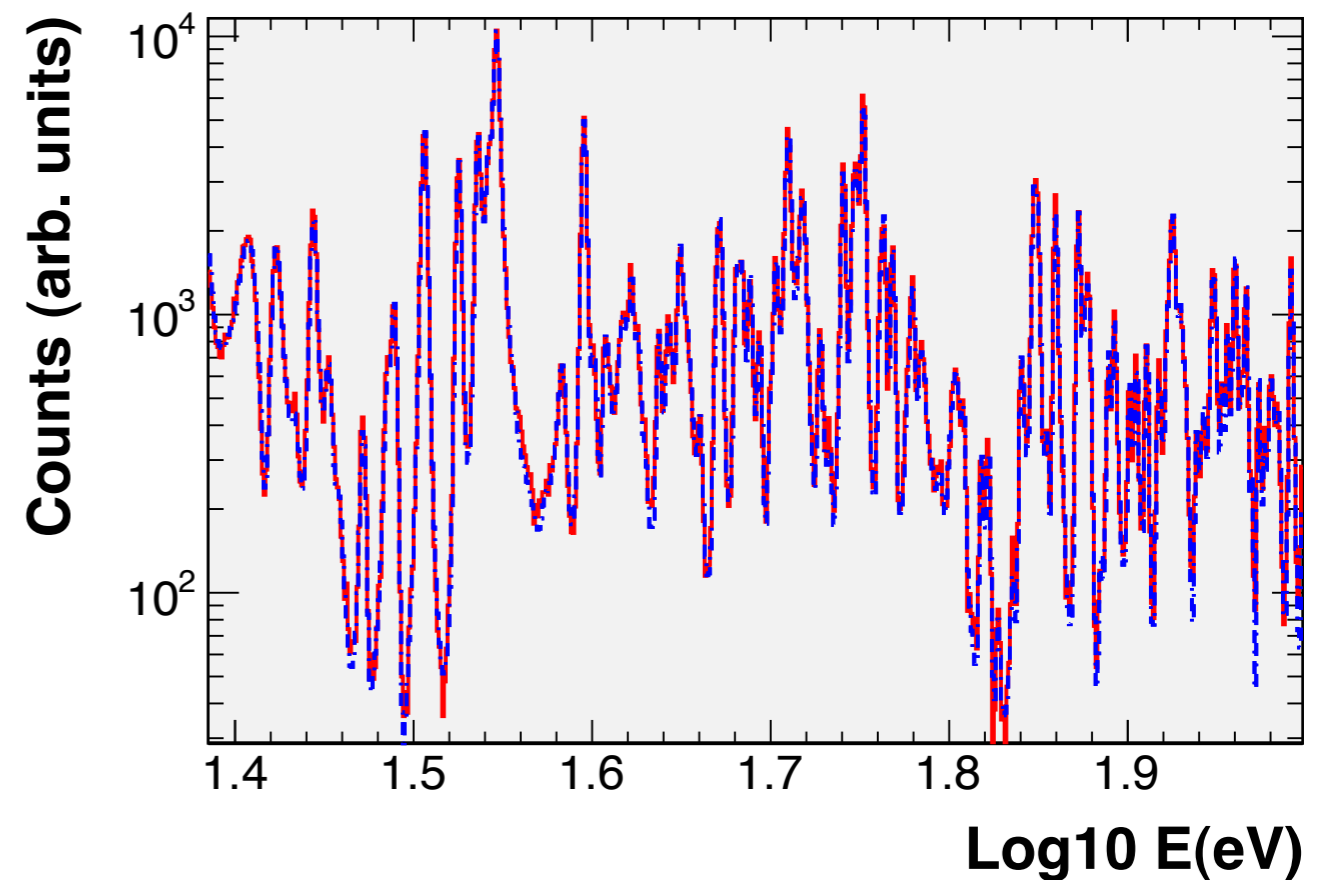
$$E(\text{eV}) = \frac{1}{\sqrt{1 - \frac{L^2}{TOF^2 c^2}}} m_n(\text{eV})$$

$$TOF = \frac{\text{time} - \text{time}_\gamma}{\text{samplerate}} + \frac{L}{c} - \text{peakshift} + \text{timeoffset}$$

The energy calibration

The energy calibration of the signal was made from the ^{235}U data and then applied to the ^{236}U data.

A perfect agreement between the latest $^{235}\text{U}(n,f)$ cross-section of ENDF and the data obtained allowed to check good time reconstruction of the signals.



The comparison of the **n_TOF ^{235}U fission counts** after performing the TOF-to-energy calibration and the superimposed **ENDF $^{235}\text{U}(n,f)$ cross-section data (scaled)**.

The neutron flux

$$\Phi_{FIC-0} = \frac{(c - b)^{235U}}{(N \epsilon \Delta_{dt})^{235U}} \frac{1}{\sigma_{ENDF/B-VII.0}^{235U}}$$

The neutron flux is obtained from the ^{235}U data, using the $^{235}\text{U}(n,f)$ cross-section extracted from the ENDF evaluation, for control purposes.

c : FFs detected

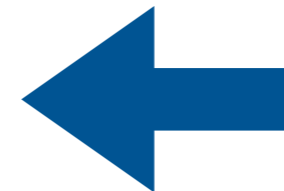
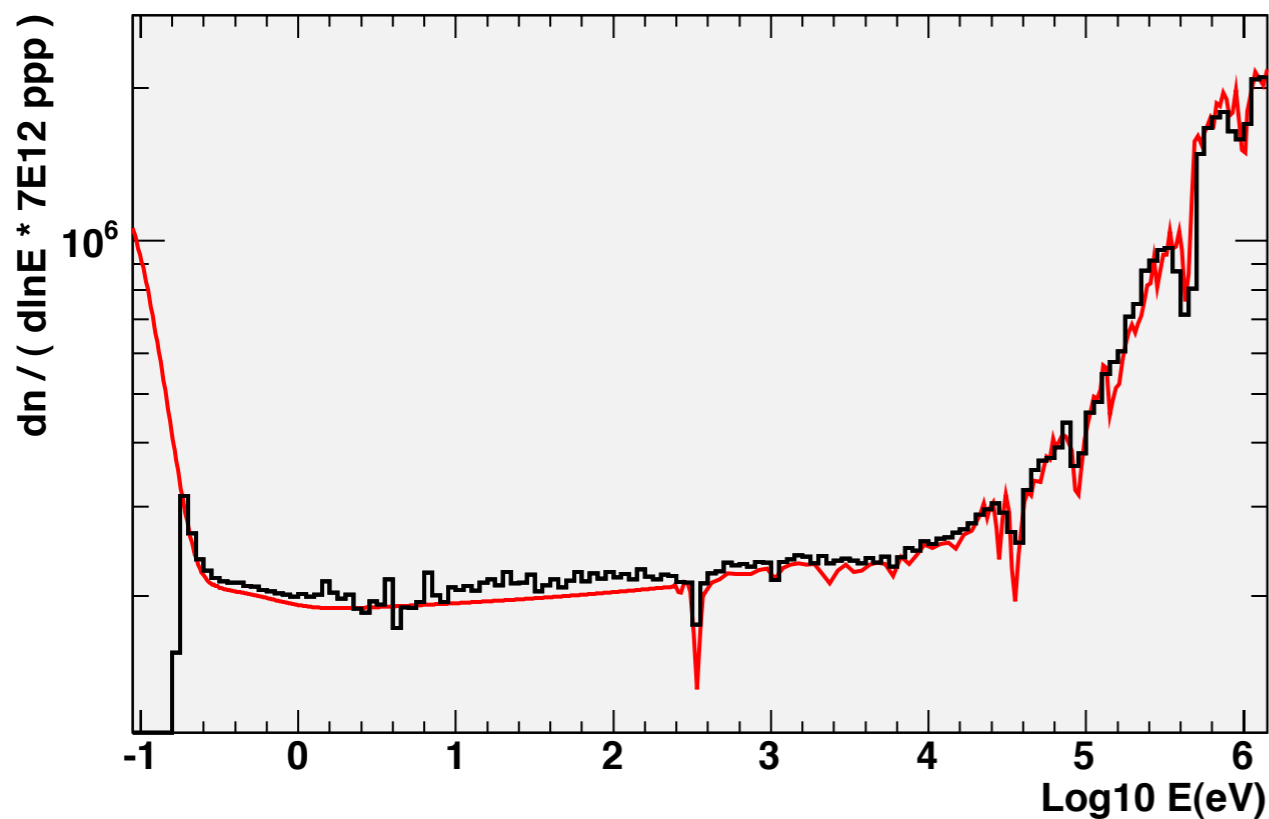
b : background

ϵ : detection efficiency

Δ_{dt} : dead-time correction factor

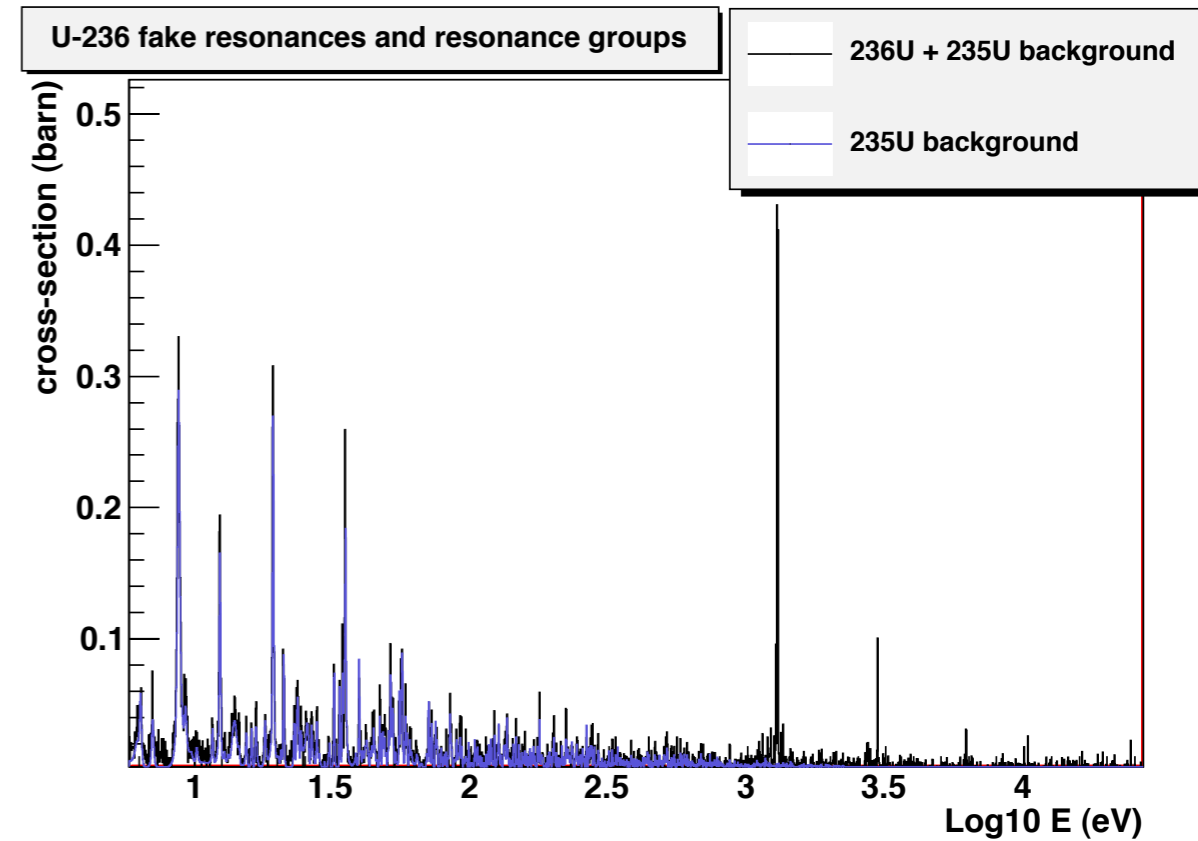
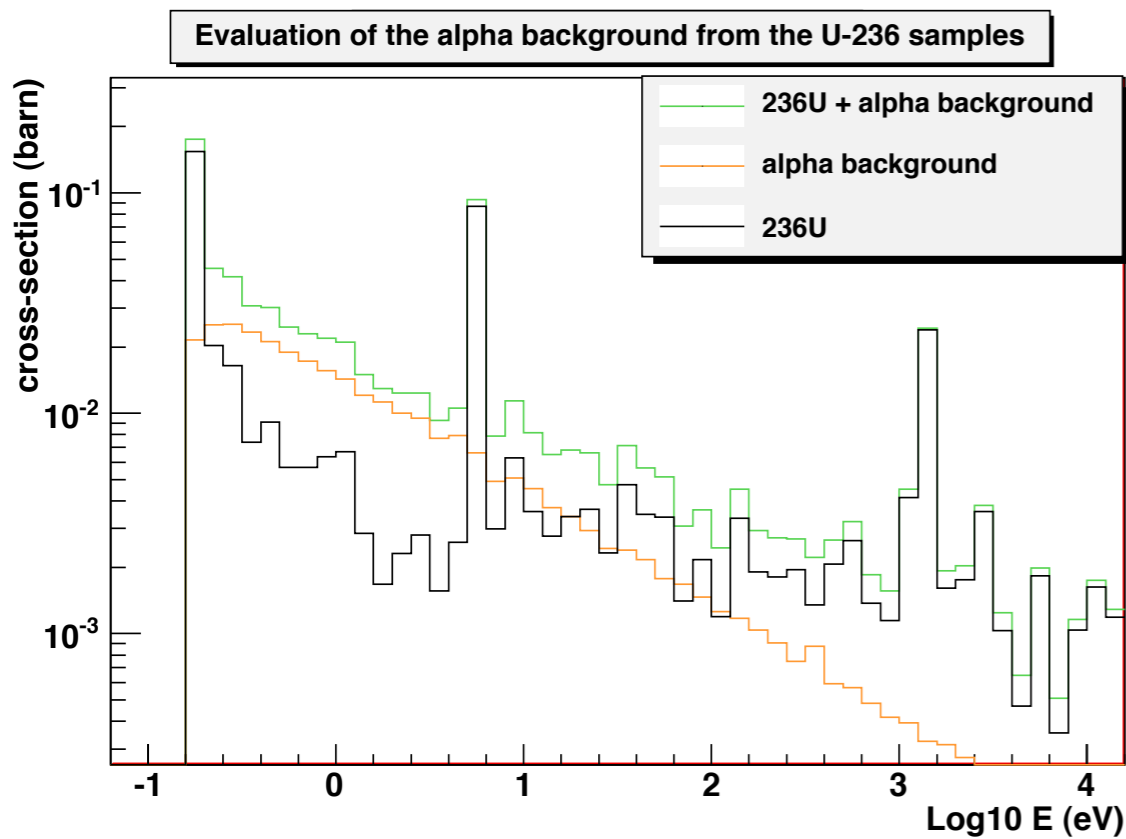
N : sample thickness

σ : evaluated cross-section



In **black**, the neutron flux obtained from the data (correctly scaled to the position of the measurement) and compared with the flux **the flux obtained from a different fission measurement.**

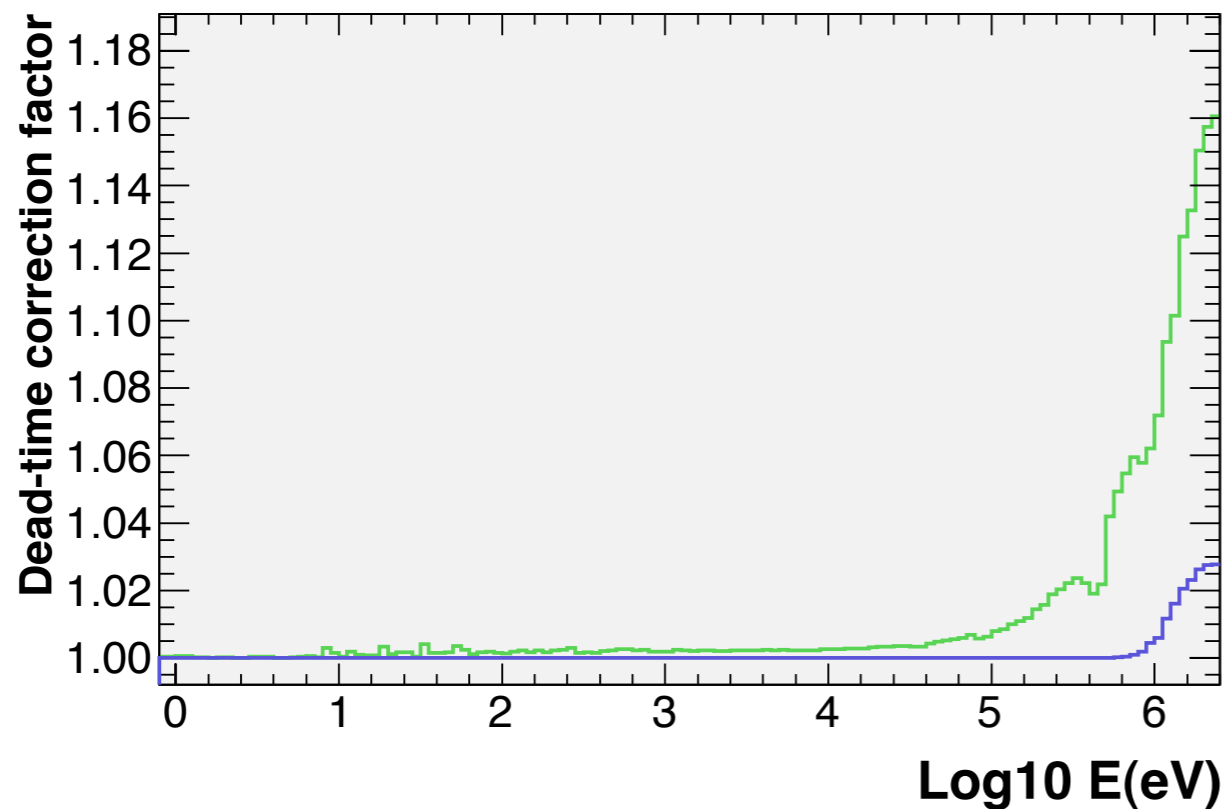
The background subtraction



- The alpha background removal was made by setting an amplitude threshold - particles depositing fewer than 35 MeV were cut off from the analysis.
- Data from runs with no neutron beam was also subtracted (after normalization) from the signal.

- The removal of the background due to the contamination of ^{235}U in the ^{236}U samples is a **critical step** in the data analysis!
- Most of the resonant contribution above 10 eV disappears at this step.

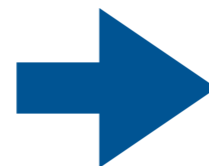
The dead-time correction



The dead-time correction becomes important for the ^{236}U above the energy threshold and for the ^{235}U above 100 keV.

This correction is higher for the dedicated data than for the parasitic data, as there are higher count-rates for the dedicated pulses.

THE DEAD-TIME
CORRECTION FACTOR



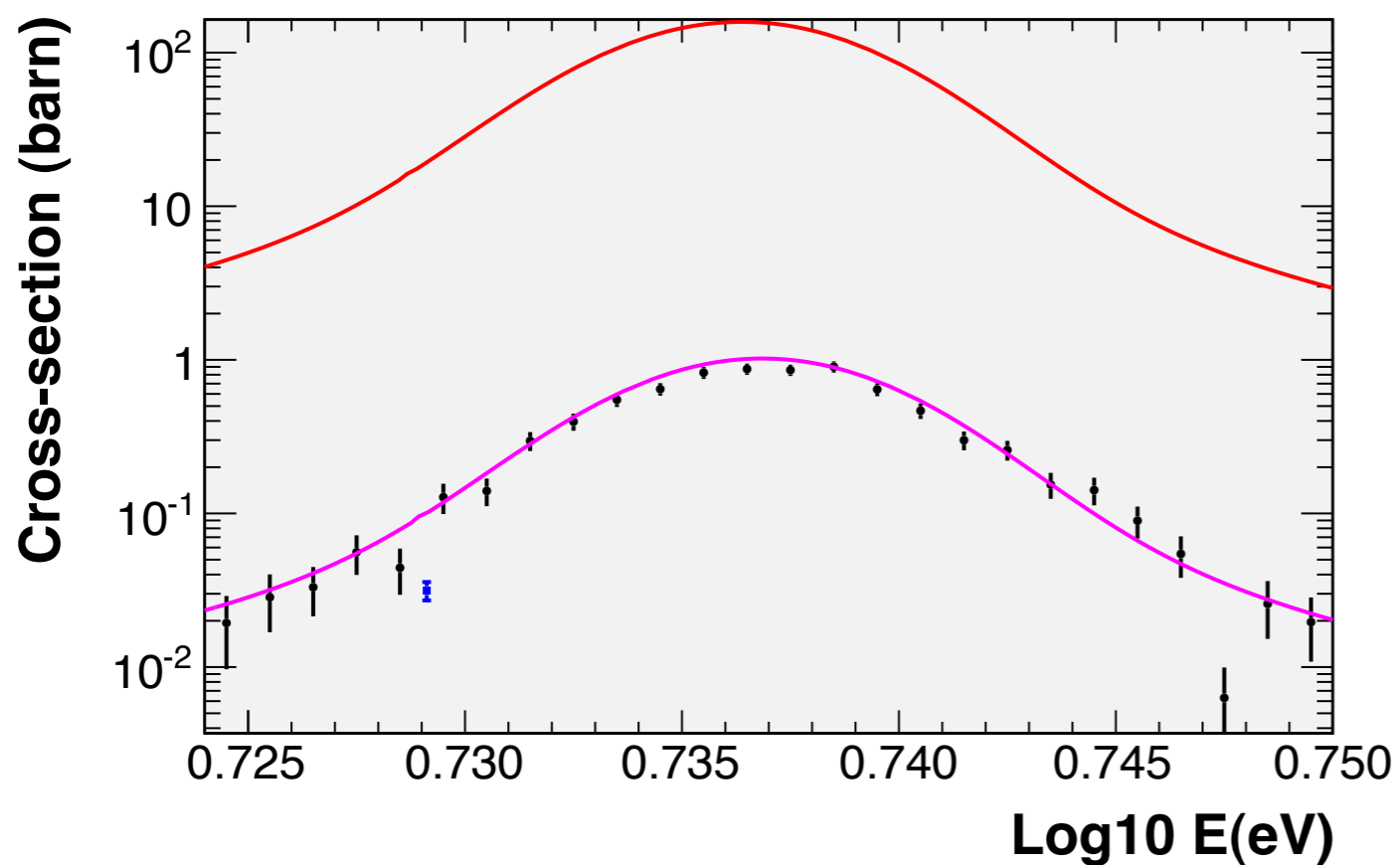
$$\Delta_{dt} = \frac{1}{1 - c_r \delta t}$$

n_TOF

Results

The low energy cross-section

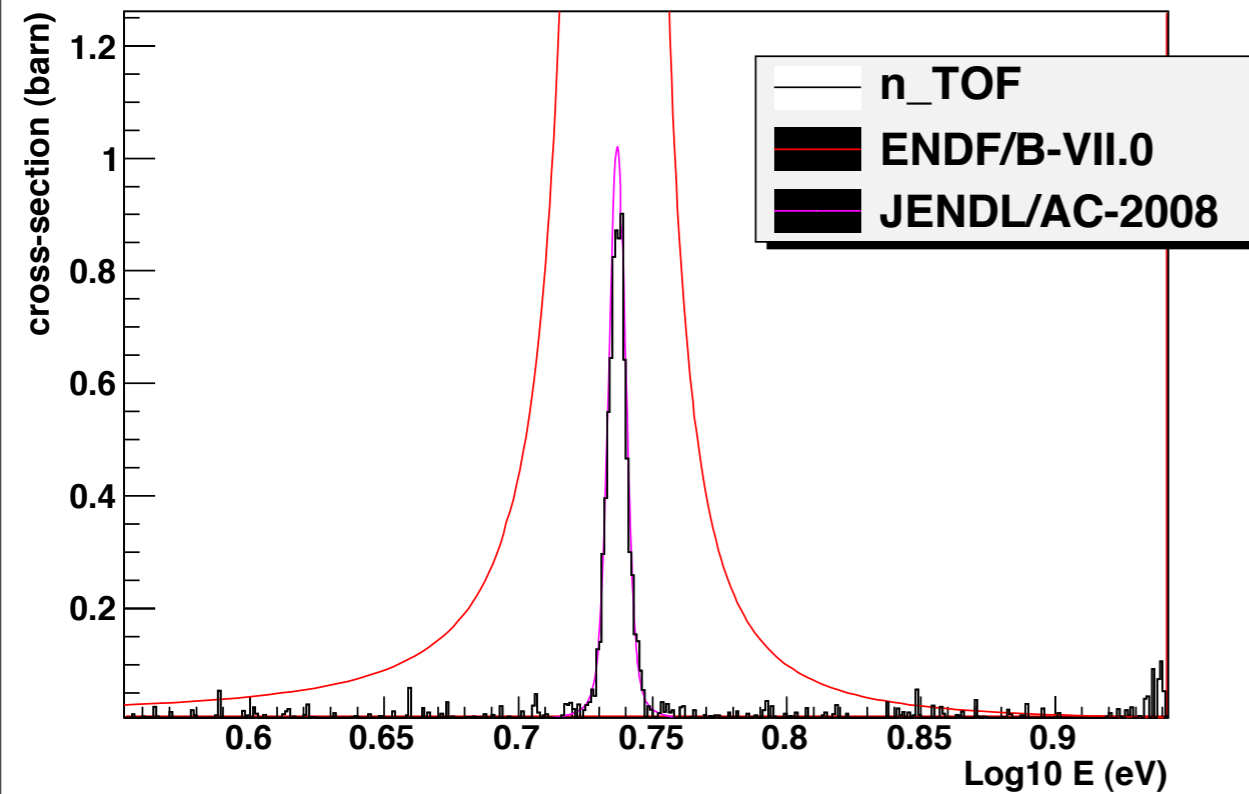
$^{236}\text{U}(n,f)$ resonance at 5.45 eV



- ENDF/B-VII.0 evaluation
- JENDL/AC-2008 evaluation
- Alekseev et al. data
- nTOF data

The low energy cross-section

5.45 eV resonance of the U-236 (n,f) cross-section



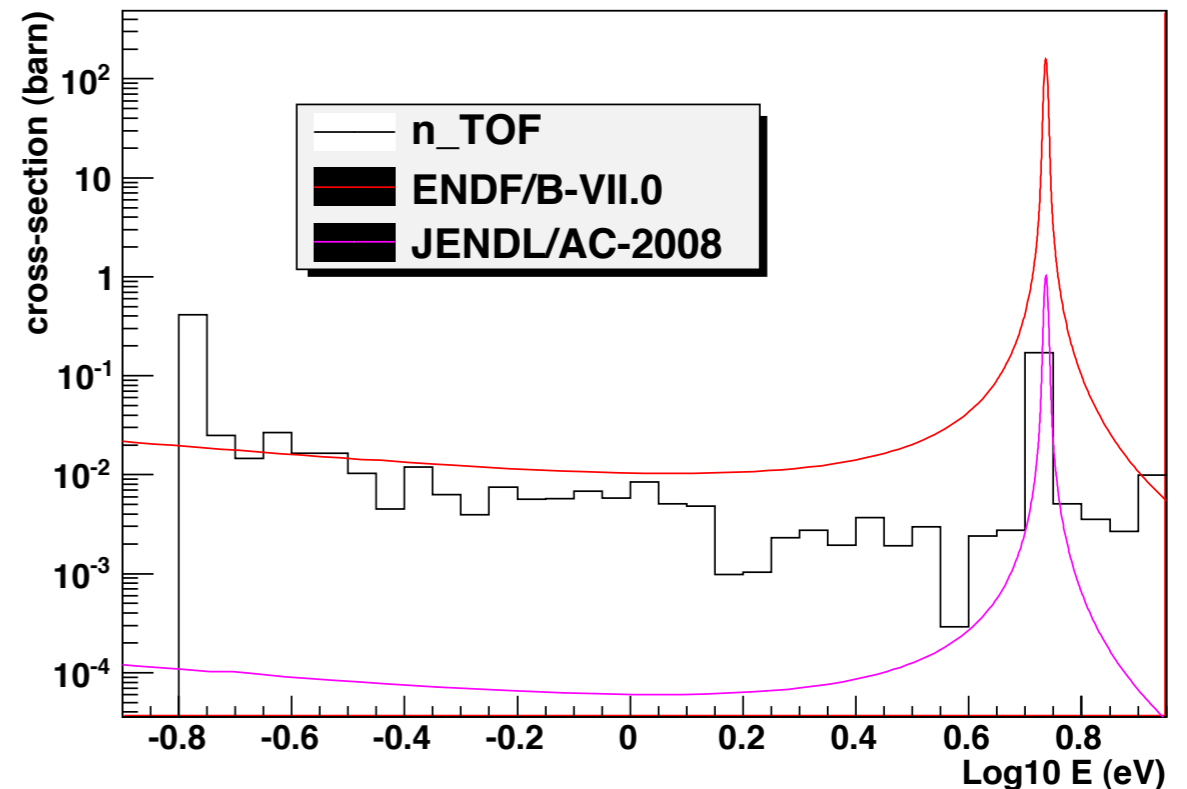
$$\sigma^{236U} = \frac{(c - b)^{236U}}{(c - b)^{235U}} \frac{(N \epsilon \Delta_{dt})^{235U}}{(N \epsilon \Delta_{dt})^{236U}} \sigma_{ENDF/B-VII.0}^{235U}$$

c : FFs detected **b** : background
ε : detection efficiency **Δ_{dt}** : dead-time correction factor
N : sample thickness **σ** : evaluated cross-section

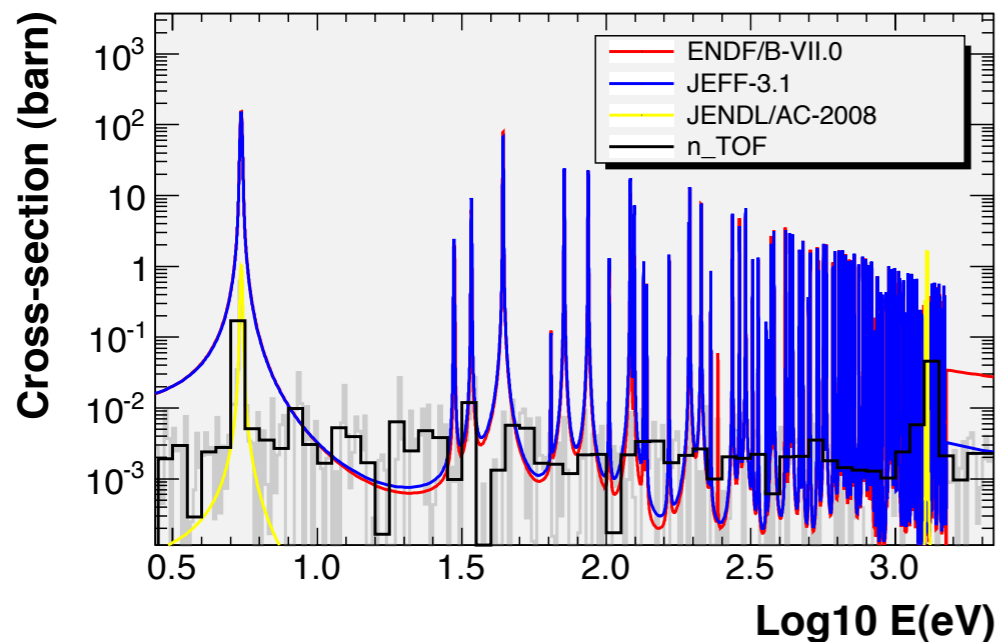
- The first resonance at 5.45 eV was measured with excellent resolution and was found to have a much smaller width than evaluated by ENDF.

- The absolute value of the cross-section below the resonance resulted with a more pronounced slope in the range 10^{-3} - 10^{-2} barn.

Low energy U-236 (n,f) cross-section

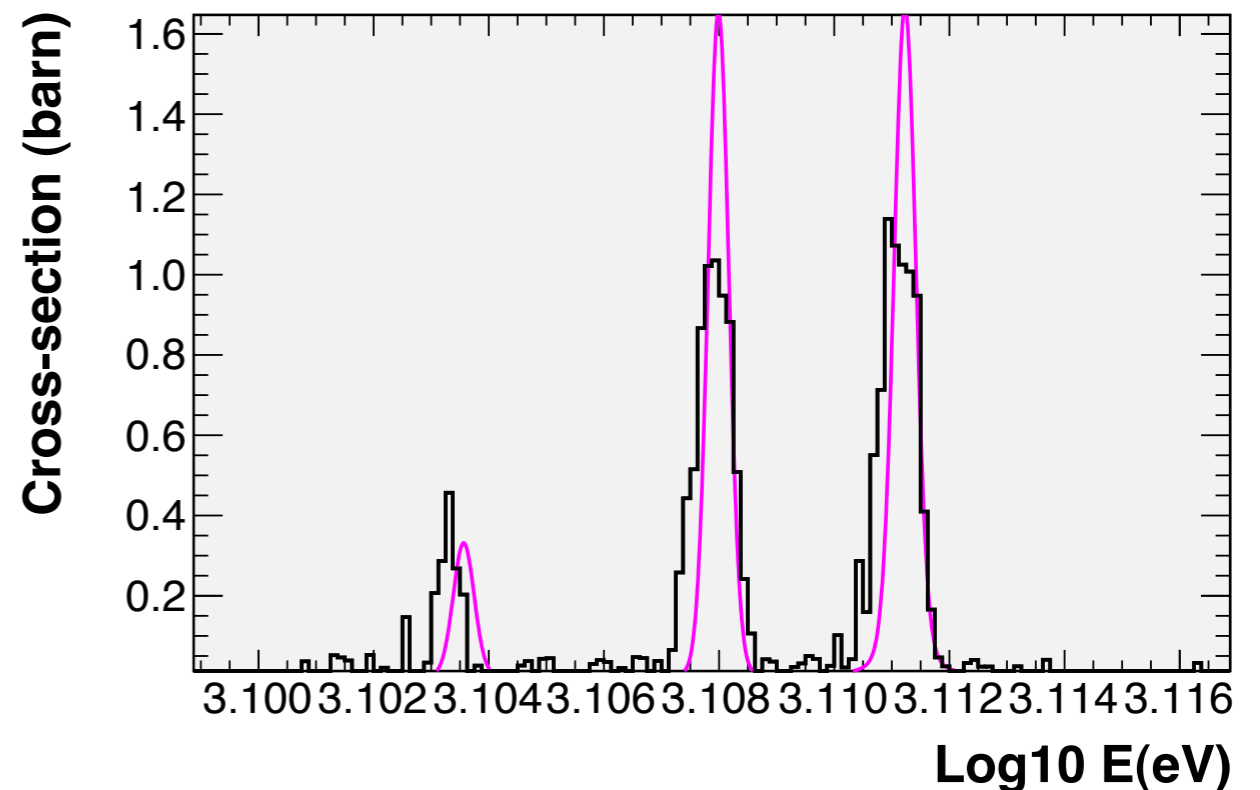


The resonance and intermediate resonance regions



- No $^{236}\text{U}(n,f)$ resonances were observed between 5.45 eV and the 1.25 keV.

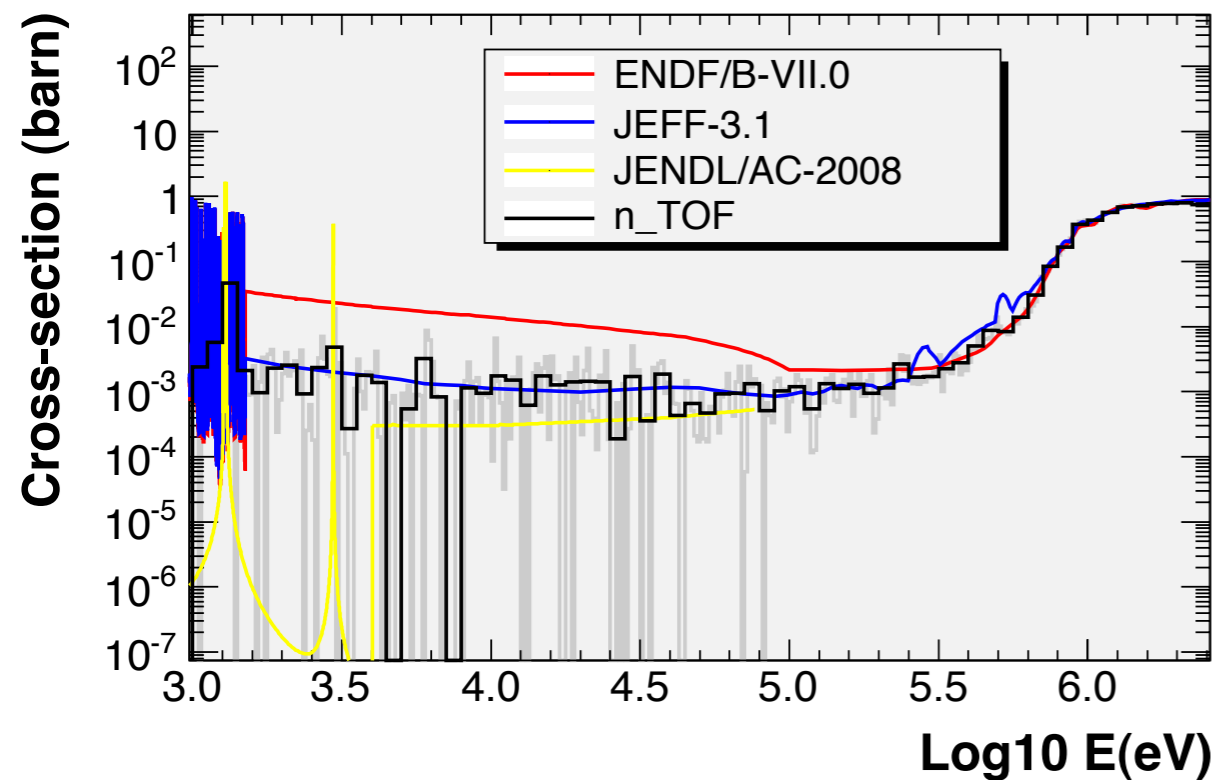
- The triple resonance at 1.25 keV was observed with very good resolution.
- The **energies of the triple resonance are below** the values presented in the literature.
- Resonances at **2.95 keV, 6.17 keV, 10.3 keV and 36.5 keV** were observed.



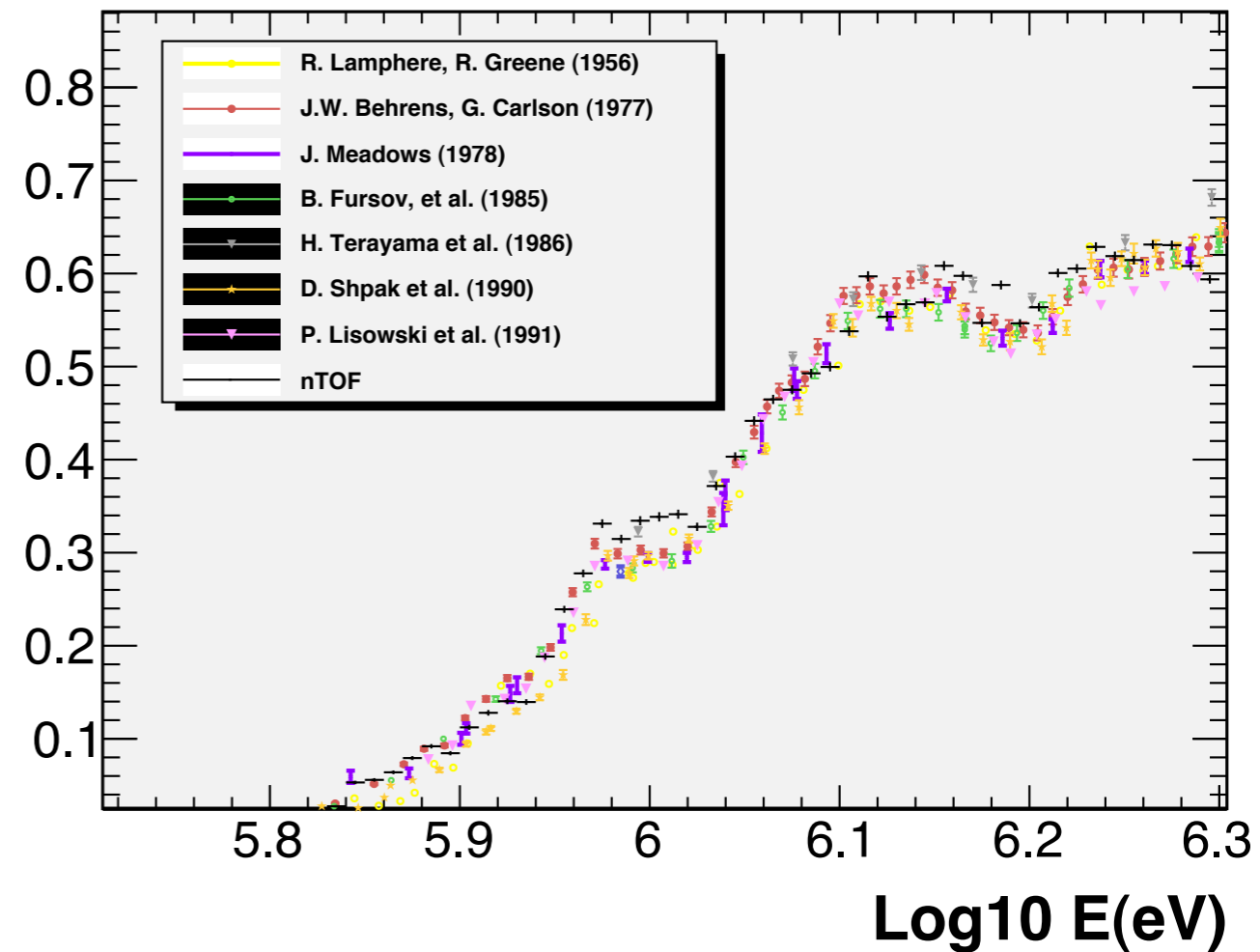
- JENDL/AC-2008 evaluation
- nTOF data

The cross-section around the threshold energy

The cross-section below the energy threshold



$\sigma_{-36} / \sigma_{-35}$



The experimental cross-section ratio to the ²³⁵U cross-section

n_TOF

Conclusions

- The cross-section was extracted **from 200 meV to 2 MeV** in a single measurement for the first time.
- The **5.45 eV resonance** was measured with good energy resolution and **fission width** similar to the measured by Wagemans et al. at GELINA (2008). This is the third measurement confirming that the cross-section value at this energy **in most evaluated data libraries is highly overestimated**.
- As on the most recent measurements, **no ^{236}U resonances were observed from 10 eV up to 1 keV**. The structure, if existing in this energy region, does not provide any significant or wide cross-section resonance. Mean values of the cross-section were taken after removing the contamination in this energy region.
- **Intermediate energy resonances were observed which are missing in the evaluated data**. These were measured with high resolution and are consistent with the latest results at LANSCE (1994), confirming the existence of intermediate structure in this nucleus.
- **Below the energy threshold, the results obtained confirm the approach followed in the JEFF evaluation**.

Final Scientific EFNUDAT Workshop
August 31, 2010 Geneva, Switzerland



n_TOF

Thank you.

Raul Sarmento, ITN rsarmento@itn.pt

Instituto Tecnológico e Nuclear, Lisbon, Portugal

Marco Calviani, CERN Marco.Calviani@cern.ch

European Organization for Nuclear Research, Geneva, Switzerland

N. Colonna, I. Gonçalves, P. Vaz & the n_TOF Collaboration www.cern.ch/ntof