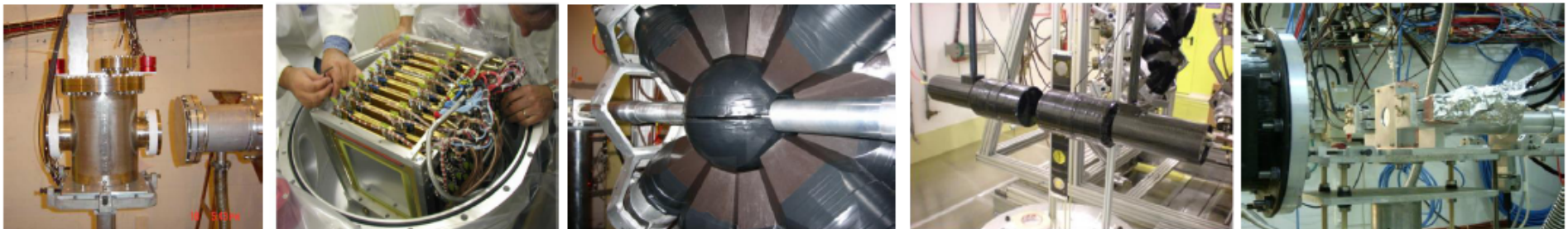




Past, Present and Future Research Activities on Nuclear Fission at n_TOF

Nicola Colonna

Istituto Nazionale Fisica Nucleare, Sezione di Bari



The **n_TOF** Collaboration

(~100 Researchers from 30 Institutes)

CERN

Technische Universität Wien

Austria

Univ. of Canberra

Australia

IRMM EC-Joint Research Center, Geel
Belgium

Charles Univ. (Prague)

Czech Republic

IN2P3-Orsay, CEA-Saclay

France

Univ. of Athens, Ioannina, Demokritos

Greece

INFN Bari, Bologna, LNL, Trieste, ENEA – Bologna

Italy

BARC

India

Univ. of Tokio

Japan

Univ. of Lodz

Poland

ITN Lisbon

Portugal

IPPE-Obninsk, JINR-Dubna

Russia

IFIN – Bucarest

Rumania

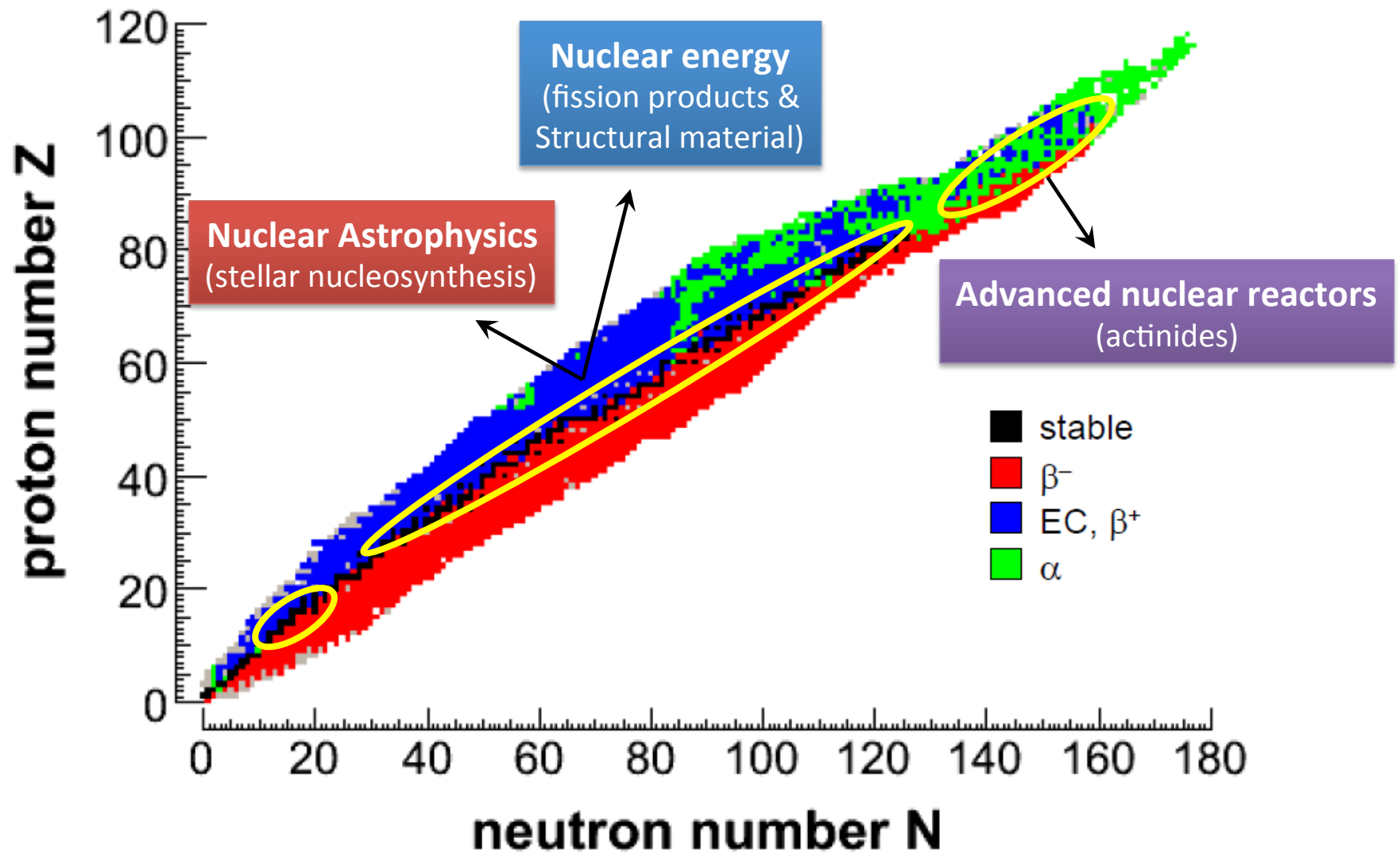
**CIEMAT, Univ. of Valencia, Santiago de Compostela,
University of Cataluna, Sevilla**

Spain

Univ. of Manchester, Univ. of York, Hearthfordshire

UK

Neutron cross sections



The nuclear waste problem

	Cm 238 2,4 h	Cm 239 3 h	Cm 240 27 d	Cm 241 32,8 d	Cm 242 162,94 d	Cm 243 29,1 a	Cm 244 18,10 a	Cm 245 8500 a	Cm 246 4730 a
Am 236 ? 3,7 m	Am 237 73,0 m	Am 238 1,63 h	Am 239 11,9 h	Am 240 50,8 h	Am 241 432,2 a	Am 242 16 h	Am 243 7370 a	Am 244 10,1 h	Am 245 2,05 h
Pu 235 25,3 m	Pu 236 2,858 a	Pu 237 45,2 d	Pu 238 87,74 a	Pu 239 2,411 · 10 ⁴ a	Pu 240 6563 a	Pu 241 14,35 a	Pu 242 3,750 · 10 ⁵ a	Pu 243 4,956 h	Pu 244 8,00 · 10 ⁷ a
Np 234 4,4 d	Np 235 396,1 d	Np 236 22,5 h	Np 237 2,144 · 10 ⁶ a	Np 238 2,117 d	Np 239 2,355 d	Np 240 7,22 m	Np 241 13,9 m	Np 242 2,2 m	Np 243 1,85 m
U 233 1,592 · 10 ⁵ a	U 234 0,0055	U 235 0,7200	U 236 2,345 · 10 ⁷ a	U 237 75 d	U 238 99,2745	U 239 23,5 m	U 240 14,1 h		U 242 16,8 m
Pa 232 1,31 d	Pa 233 27,0 d	Pa 234 1,17 m	Pa 235 24,2 m	Pa 236 9,1 m	Pa 237 8,7 m	Pa 238 2,3 m			
Th 231 25,5 h	Th 232 100	Th 233 22,3 m	Th 234 24,10 d	Th 235 7,1 m	Th 236 37,5 m	Th 237 5,0 m			

244, 245Cm
1.5 Kg/yr

241Am: 11.6 Kg/yr
243Am: 4.8 Kg/yr

239Pu: 125 Kg/yr

237Np: 16 Kg/yr

LLFP
76.2 Kg/yr

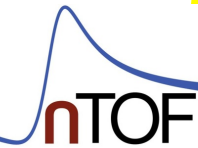
Quantities refer to yearly production in 1 GW_e LW reactor

LLFP



The Th/U fuel cycle

	Cm 238 2,4 h	Cm 239 3 h	Cm 240 27 d	Cm 241 32,8 d	Cm 242 162,94 d	Cm 243 29,1 a	Cm 244 18,10 a	Cm 245 8500 a	Cm 246 4730 a	
Am 236 ? 3,7 m	Am 237 73,0 m	Am 238 1,63 h	Am 239 11,9 h	Am 240 50,8 h	Am 241 432,2 a	Am 242 141 a	Am 243 7370 a	Am 244 26 m	Am 245 2,05 h	
Pu 235 25,3 m	Pu 236 2,858 a	Pu 237 45,2 d	Pu 238 87,74 a	Pu 239 2,411 · 10 ⁴ a	Pu 240 6563 a	Pu 241 14,35 a	Pu 242 3,750 · 10 ⁵ a	Pu 243 4,956 h	Pu 244 8,00 · 10 ⁷ a	
Np 234 4,4 d	Np 235 396,1 d	Np 236 22,5 h	Np 237 2,144 · 10 ⁶ a	Np 238 2,117 d	Np 239 2,355 d	Np 240 7,22 m	Np 241 13,9 m	Np 242 2,2 m	Np 243 1,85 m	
U 233 1,592 · 10 ⁵ a	U 234 0,0055	U 235 0,7200	U 236 120 ns	U 237 4,75 d	U 238 99,2745	U 239 23,5 m	U 240 14,1 h		U 242 16,8 m	
Pa 232 1,31 d	Pa 233 2,0 d	Pa 234 1,17 m	Pa 235 24,2 m	Pa 236 9,1 m	Pa 237 8,7 m	Pa 238 2,3 m	148		150	
Th 231 25,5 h	Th 232 1,405 · 10 ¹⁰ a	Th 233 22,3 m	Th 234 24,10 d	Th 235 7,1 m	Th 236 37,5 m	Th 237 5,0 m				



FP

LLFP

Nuclear energy and the need of nuclear data – EU programs

Objectives of the nuclear industry/research in Europe:

- improve safety and efficiency of current reactors (LWR)
- develop a new generation of reactors (Gen IV fast reactors and Accelerator Driven Systems)

Strategic Research Agenda of the European Sustainable Nuclear Energy Technology Platform (SNETP)

Availability of **accurate nuclear data** (cross sections, decay constants, branching ratios, etc.) is the basis **for precise calculations** (for safety, efficiency, and life extension) and **new generation reactors**. **Need to measure fission cross section of actinides (from Th to Cm) with half-life from a few years up to 10¹⁰ years.** This is particularly true **for fuels containing minor actinides** for their transmutation in fast spectra.

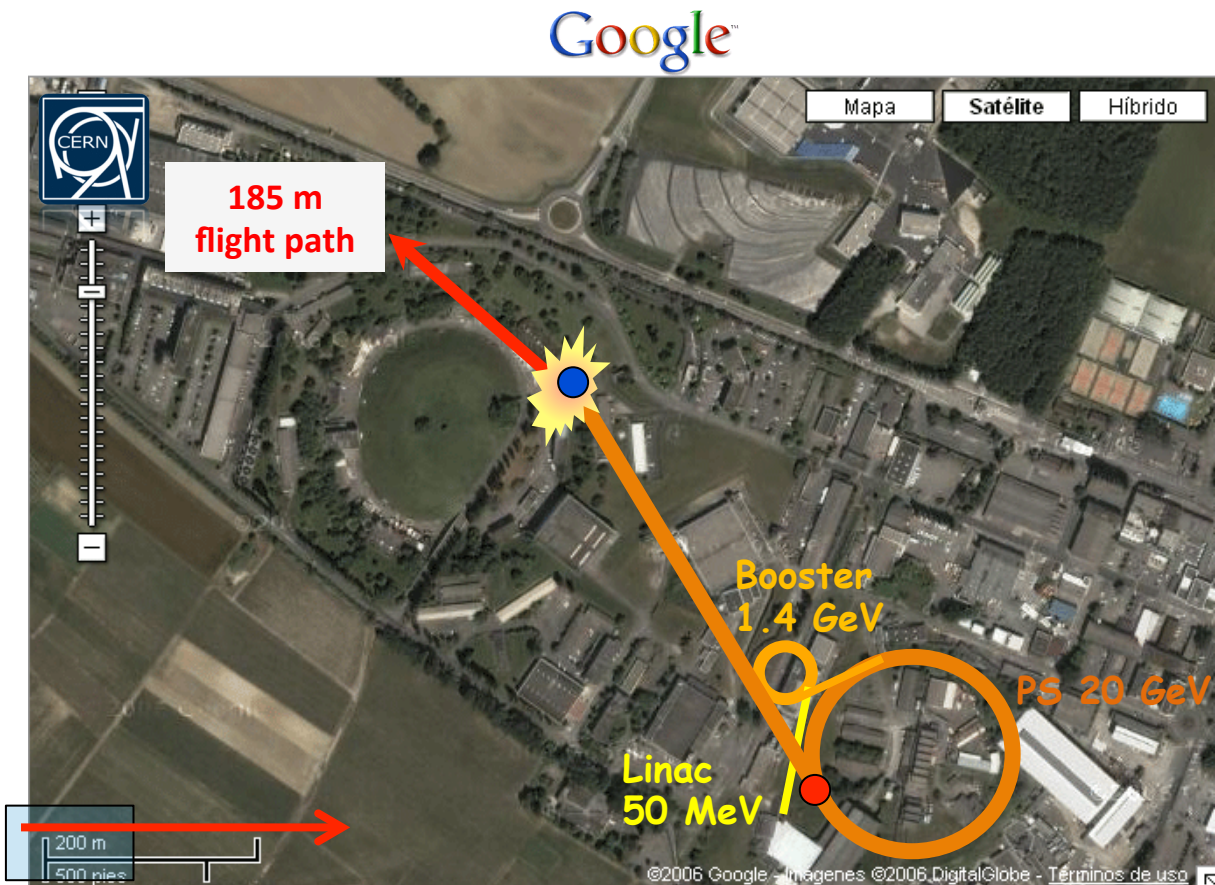
NEA/WPEC-26 (ISBN 978-92-64-99053-1)

UNCERTAINTY AND TARGET ACCURACY
ASSESSMENT FOR INNOVATIVE SYSTEMS USING
RECENT COVARIANCE DATA EVALUATIONS

The overall list of requirements is rather long:

- capture cross sections of ^{235}U , ^{238}U , ^{237}Np , $^{238-242}\text{Pu}$, $^{241,242\text{m},243}\text{Am}$, ^{244}Cm
- fission cross sections of ^{234}U , ^{237}Np , $^{238,240-242}\text{Pu}$, $^{241,242\text{m},243}\text{Am}$, $^{242-246}\text{Cm}$

The n_TOF facility at CERN

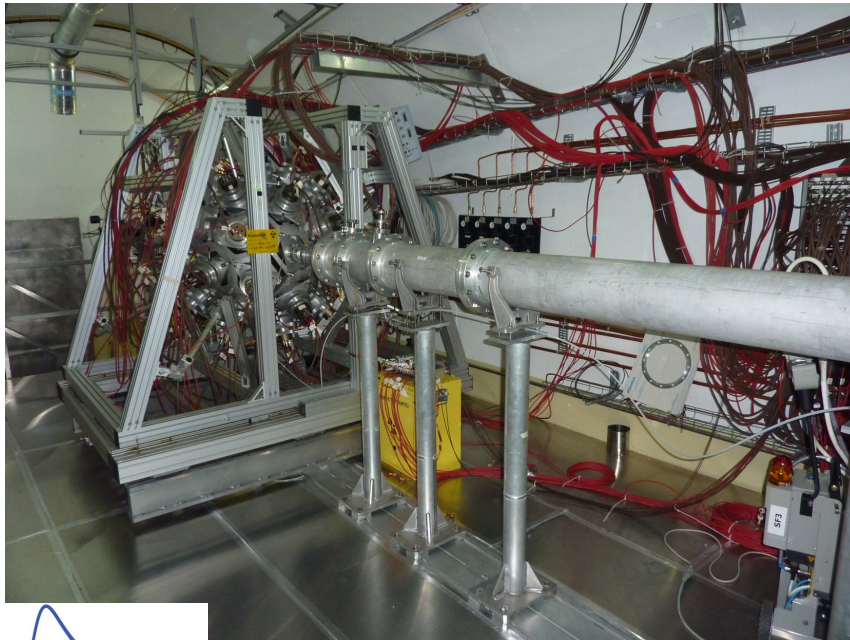


n_TOF is a **spallation** neutron source based on **20 GeV/c protons** from the CERN PS hitting a **Pb block** (~360 neutrons per proton).

Experimental area at **185 m** (now a second one at **20 m**)



The real thing



The spallation target(s)

Phase 1: 2001 – 2004

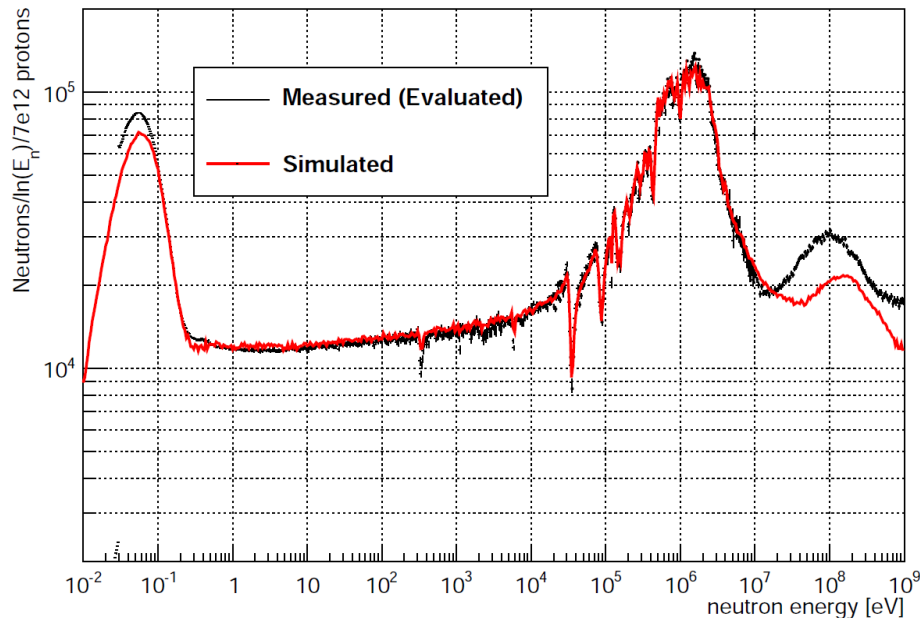


Phase 2: 2008 – today



The n_TOF facility

Advantages of the **PS proton beam**: high energy, high peak current, low duty cycle.



Main feature: **high instantaneous neutron flux** (10^6 n/pulse).

- very convenient for measurements of **radioactive isotopes** (maximizes signal-to-background ratio)
- ideal facility for **actinides** (nuclear technology)

Other features of the neutron beam:

- high **resolution in energy** ($\Delta E/E = 10^{-4}$) study **resonances**
- wide **energy range** ($25 \text{ meV} < E_n < 1 \text{ GeV}$) measure **fission** from thermal to GeV
- low **repetition rate** ($< 0.8 \text{ Hz}$) no **wrap-around**

The n_TOF measurements

Phase 1 (2001-2004)

Capture

^{151}Sm

^{232}Th

$^{204,206,207,208}\text{Pb}$, ^{209}Bi

$^{24,25,26}\text{Mg}$

$^{90,91,92,94,96}\text{Zr}$, ^{93}Zr

$^{186,187,188}\text{Os}$

$^{233,234}\text{U}$

^{237}Np , ^{240}Pu , ^{243}Am

Fission

$^{233,234,235,236,238}\text{U}$

^{232}Th , ^{209}Bi , ^{237}Np

$^{241,243}\text{Am}$, ^{245}Cm

Phase 2 (2009-2012)

Capture

^{25}Mg , ^{88}Sr

$^{58,60,62}\text{Ni}$, ^{63}Ni

$^{54,56,57}\text{Fe}$

$^{236,238}\text{U}$, ^{241}Am

Fission

$^{240,242}\text{Pu}$

$^{235}\text{U}(n,\gamma/f)$

^{232}Th , ^{234}U

^{237}Np (FF ang.distr.)

(n, α)

^{33}S , ^{59}Ni

Phase 3 (just started)

Capture

....

....

Fission

....

....

(n, p) (n, α)

....

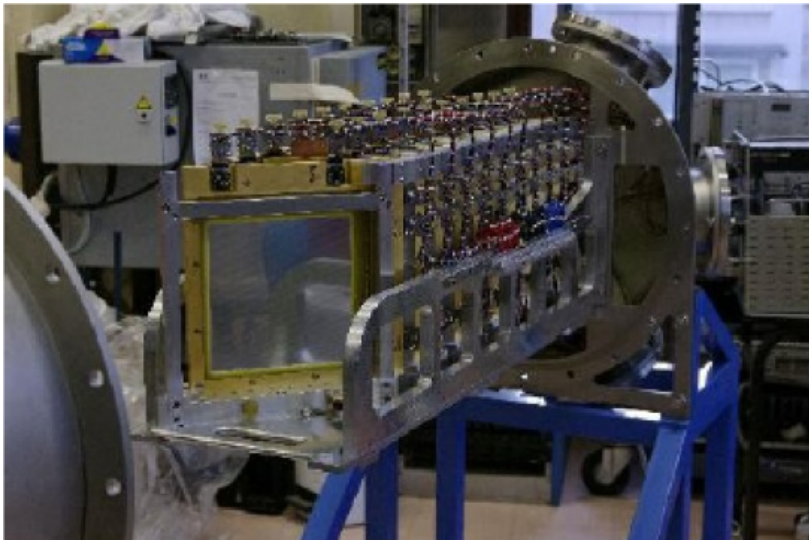
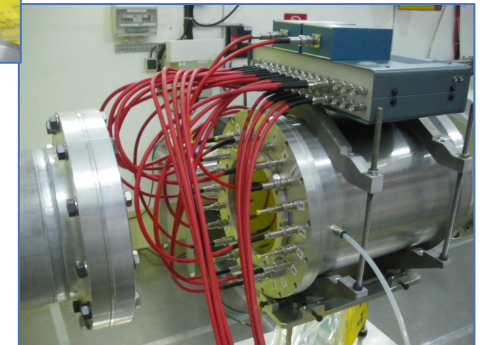
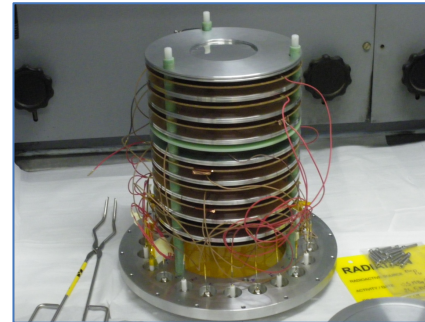
....

Experimental setups for fission

Over the years, **several systems** have been used for detecting fission fragments, with **two different techniques**.

Micromegas chamber

- low-noise, high-gain, radiation-hard detector



Parallel Plate Avalanche Counters (PPAC)

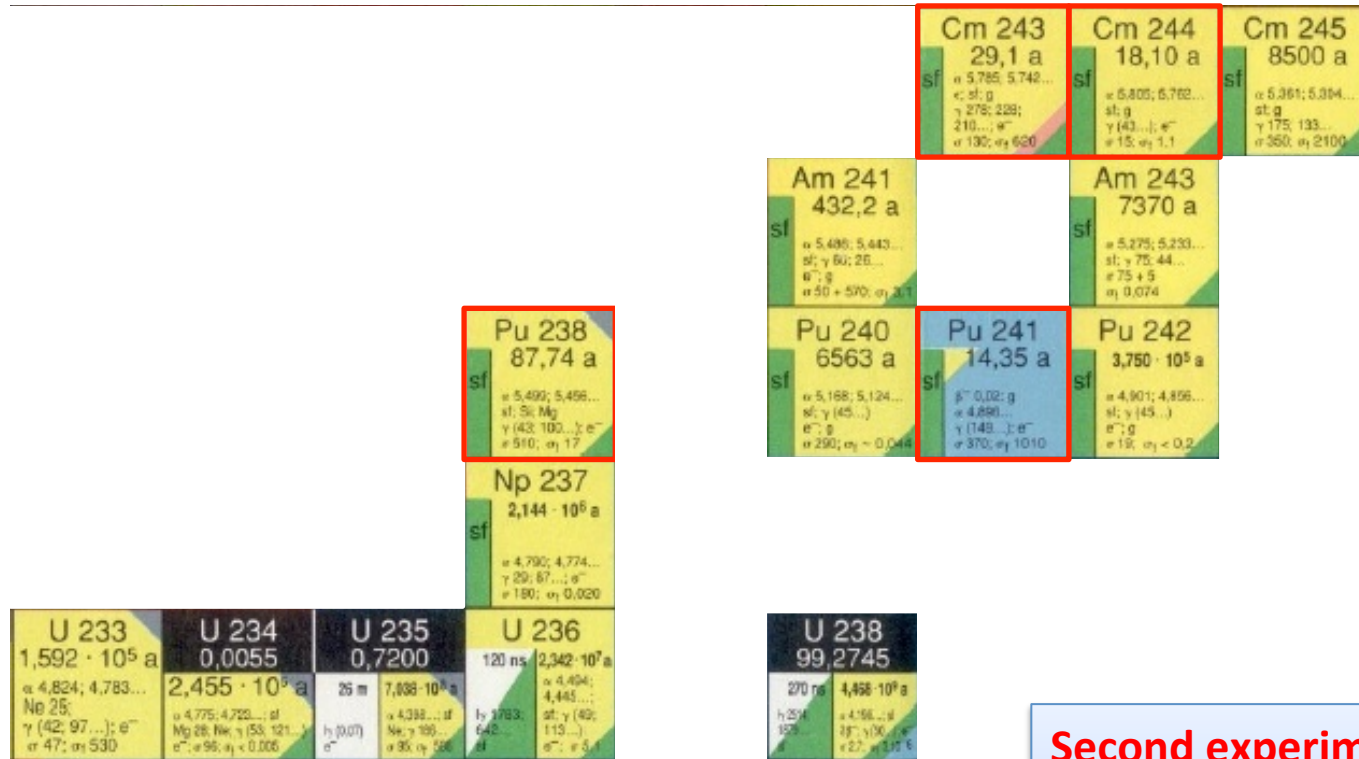
- Fission fragments detected **in coincidence**
- Very good rejection of **α -background**
- Low sensitivity to **γ -flash**
- **Position sensitive** to measure angular distribution

Measured fission reactions



In the two experimental campaigns, measured capture and fission cross sections for **most long-lived actinides** (432 y and above).
For some of them, measured FF anysotropy as well.

More to be measured



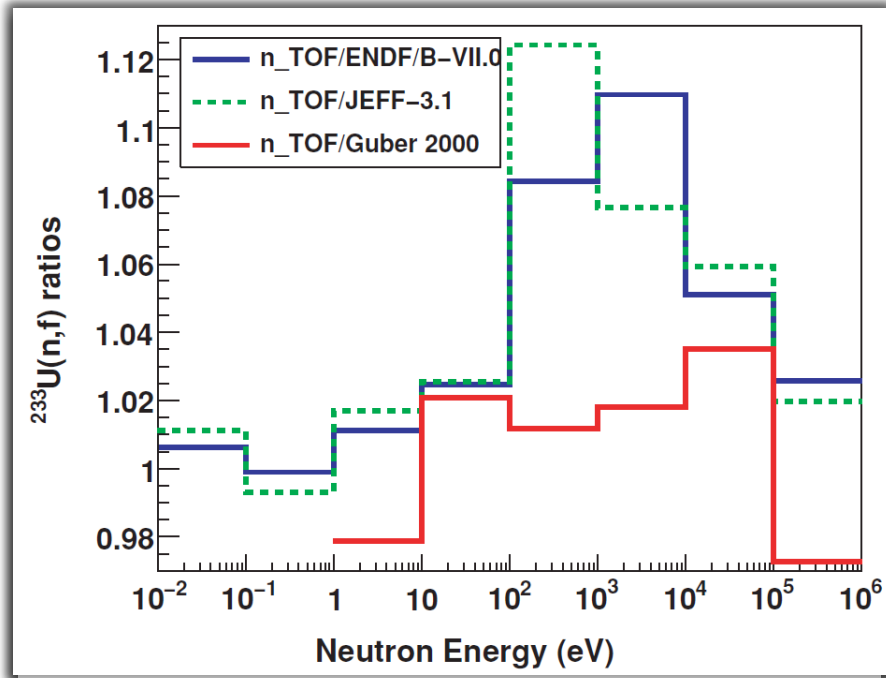
Second experimental area at n_TOF will allow to measure also some short-lived actinides.

²³⁰Th

Th 232
100
1,405 · 10¹⁰ a

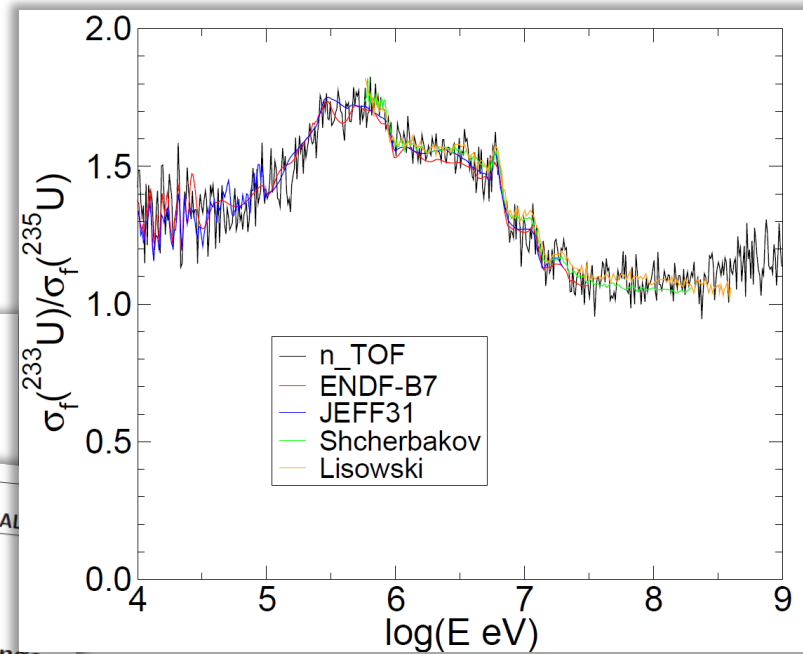


The $^{233}\text{U}(n,f)$ cross section



Half-life: 1.59×10^5 y
 Sample: 29 mg (/4)
 Activity: 2.6 MBq (each sample)

Fission cross-section on ^{233}U measured in a single measurement from thermal to 20 MeV, with **5 % accuracy, and high resolution.**



PHYSICAL REVIEW C 80, 044604 (2009)

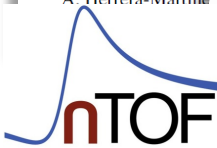
High-accuracy fission cross-section measurement at the white-neutron source n_TOF from
 Eur. Phys. J. A (2011) 47: 2
 DOI 10.1140/epja/i2011-11002-y

Regular Article – Experimental Physics

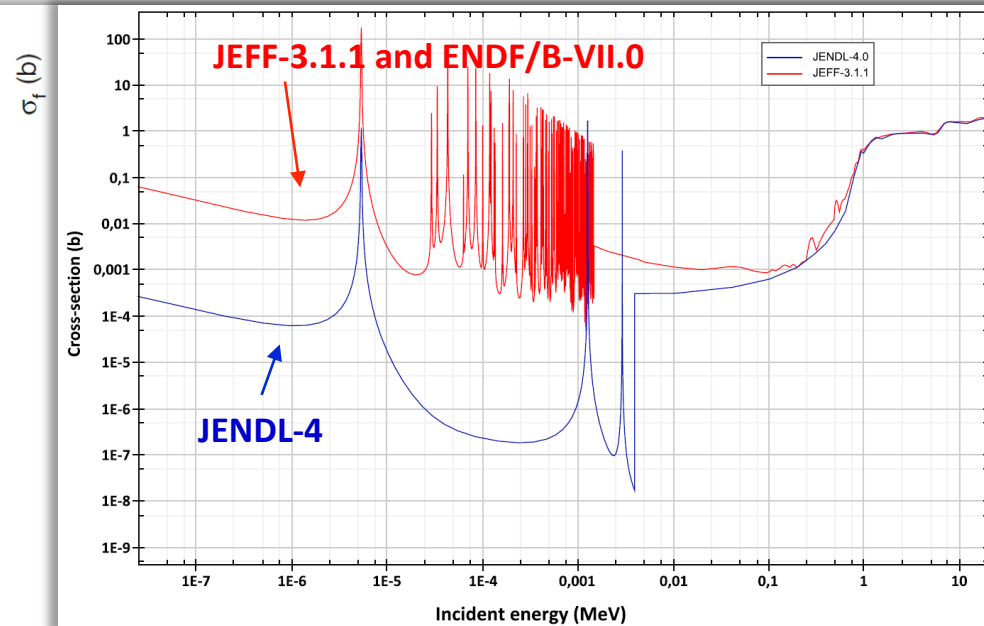
THE EUROPEAN PHYSICAL JOURNAL

M. Calviani,^{1,2,*} J. Prato,³
 P. Assimakopoulos,^{8,†}
 F. Calviño,¹⁴ D. Cano-Ott,¹⁵
 A. Couture,²² J. Cox,¹⁶
 M. Embid-Segura,¹⁷
 E. González-Romero,¹⁸
 A. Herrera-Martín,¹⁹

Neutron-induced fission cross-section of ^{233}U in the energy range
 $0.5 < E_n < 20$ MeV



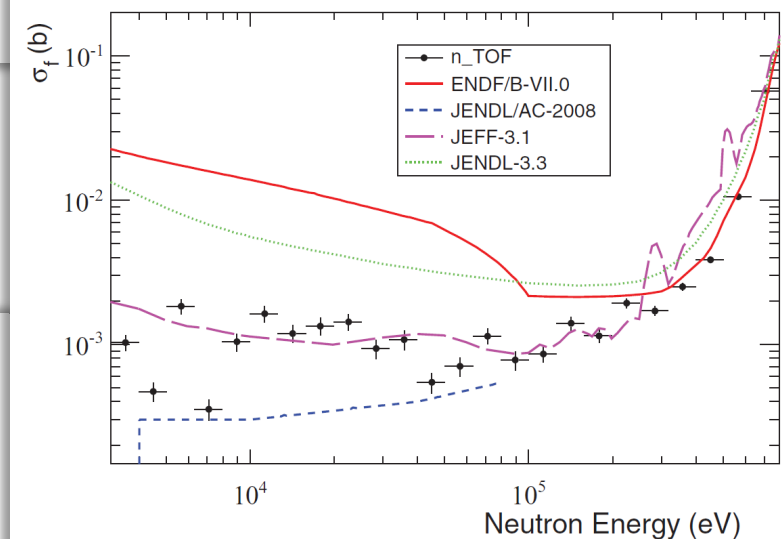
The fission cross-section of ^{236}U



Half-life: 2.34×10^7 y
 Sample: 21.4 mg (/4)
 Activity: 13 kBq (each sample)
 Contamination of ^{235}U 0.05%

n_TOF data **confirm** results from GELINA (C. Wagemans et al.).
 Below a few keV, **ENDF and JEFF overestimate** cross section (x100).

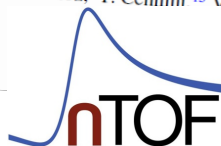
Resonances in ENDF and JEFF are **from ^{235}U !!**
JENDL-4 is (mostly) correct



PHYSICAL REVIEW C 84, 044618 (2011)

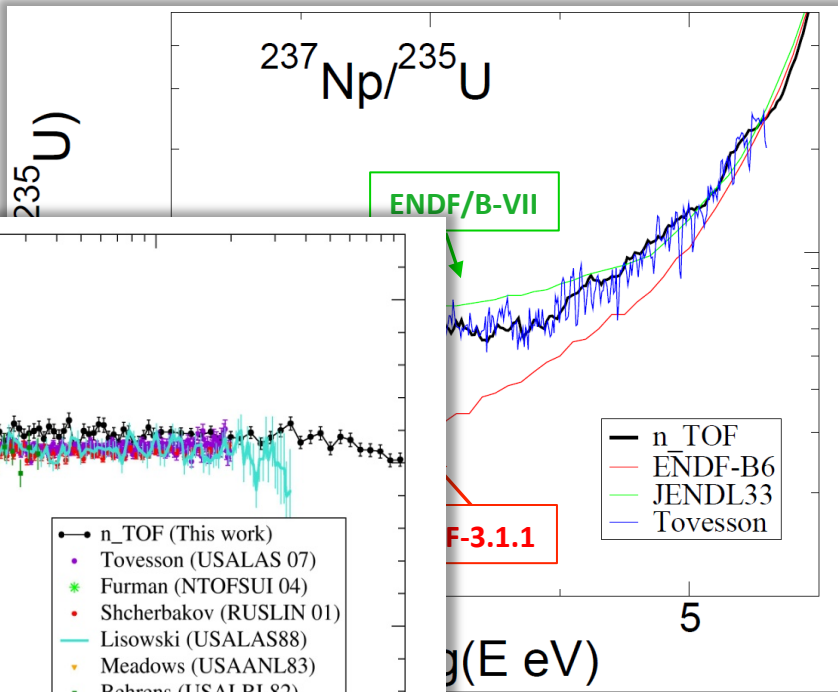
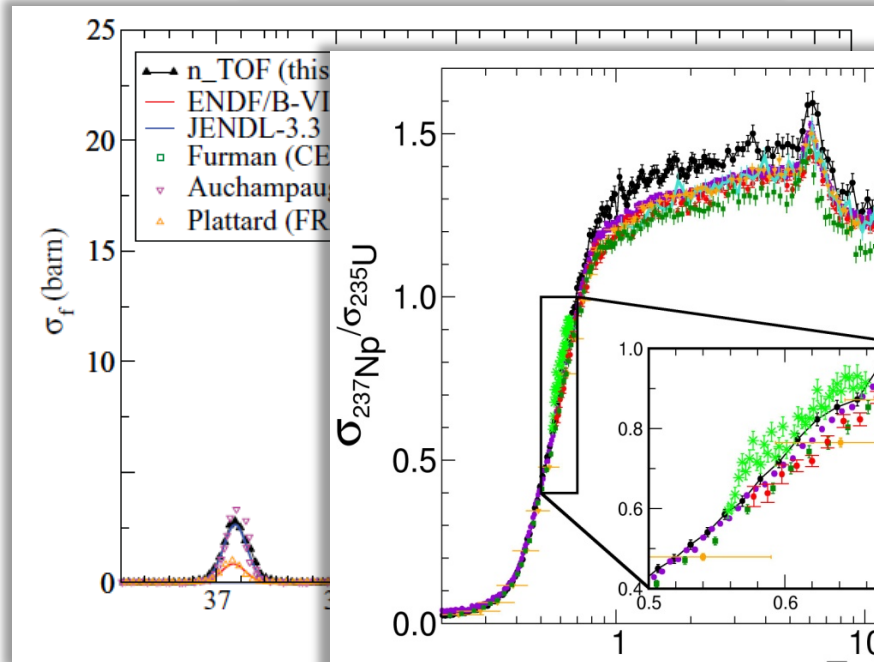
Measurement of the $^{236}\text{U}(n,f)$ cross section from 170 meV to 2 MeV at the CERN n_TOF facility

R. Sarmiento,¹ M. Calviani,² J. Praena,¹⁴ N. Colonna,³ F. Belloni,⁴ I. F. Gonçalves,¹ P. Vaz,¹ G. Aerts,¹⁷ H. Alvarez,²⁶
 F. Alvarez-Velarde,⁵ S. Andriamonje,² J. Andrzejewski,⁶ P. Assimakopoulos,⁷ L. Audouin,⁸ M. Barbagallo,³ G. Badurek,⁹
 P. Baumann,¹⁰ F. Becvar,¹¹ E. Berthoumieux,¹⁷ F. Calvino,¹² D. Cano-Ott,⁵ R. Capote,^{13,14} C. Carrapico,¹ A. Carrillo de
 Albornoz,¹ P. Cennini,¹⁵ V. Chepel,¹⁶ E. Chiaveri,² G. Cortes,¹⁸ A. Couture,¹⁹ J. Cox,¹⁹ M. Dahlfors,¹⁵ S. David,⁸ M. Diakaki,³⁴
²¹ C. Domingo-Pardo,²² W. Dridi,¹⁷ I. Duran,²⁶ C. Eleftheriadis,²³ L. Ferrant,⁸ A. Ferrari,¹⁵
 rais-Koelbl,¹³ K. Fuji,⁴ W. Furman,²⁴ E. Gonzalez-Romero,⁵ A. Guerrero,^{5,2} F. Gunsing,¹⁷ D. ...



The $^{237}\text{Np}(n,f)$ reaction

Half-life: 2.14×10^6 y
 Sample: 63 mg (/4)
 Activity: 0.41 MBq (each sample)



Below threshold, some corrections on current library
 recent

Controversial result, still being checked (not confirmed by other n_TOF datasets).
 Still needs confirmation !!

EW C 82, 034601 (2010)
 CERN Neutron Time-of-Flight (n_TOF) facility
 C. Parada, L. Tassan-Got, L. Audouin, D. Tarrío, ...
 C. Le Naour, C. Stephan, ...
 M. Calviani, D. Cano-Ott, ...
 A. Mengoni, P. M. Milazzo, C. Moreau, ...



$^{240,242}\text{Pu}(n,f)$ at n_TOF

See next talk:

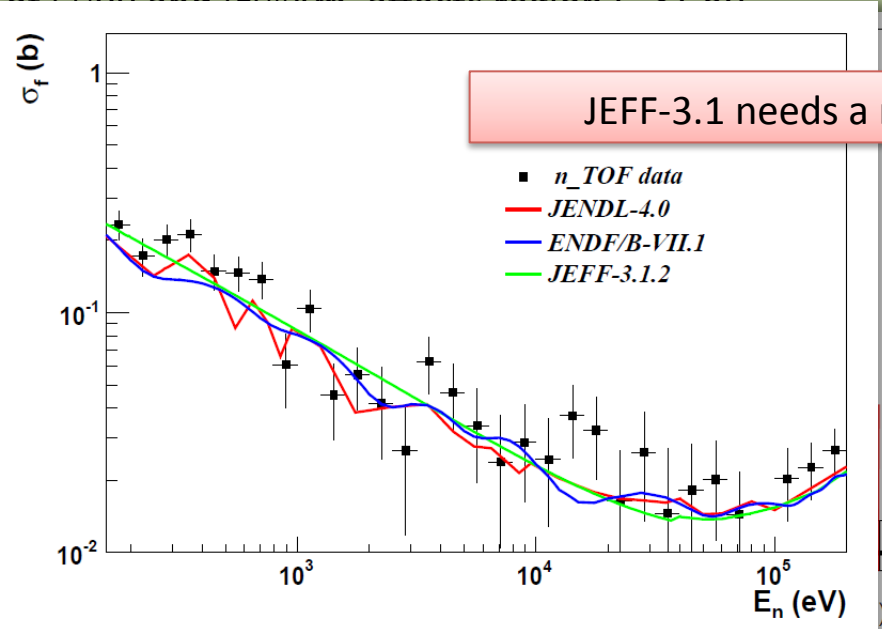
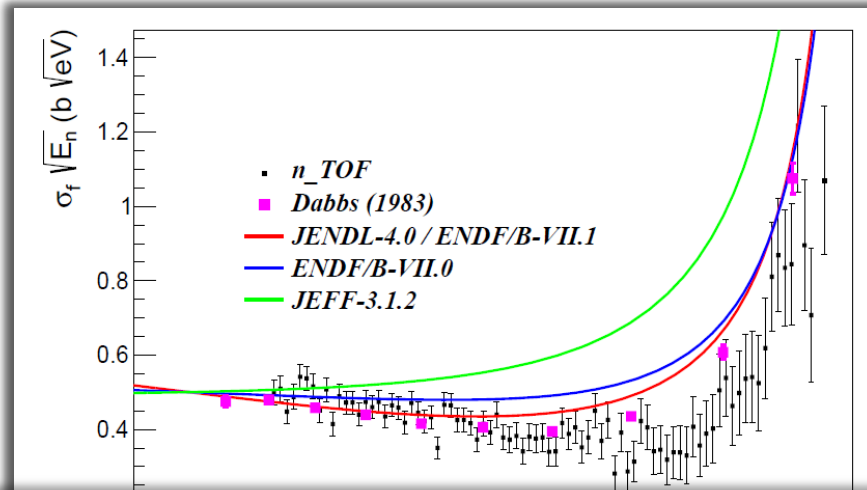
Andrea Tsinganis (NTUA/CERN)

The $^{241}\text{Am}(n,f)$ at n_TOF

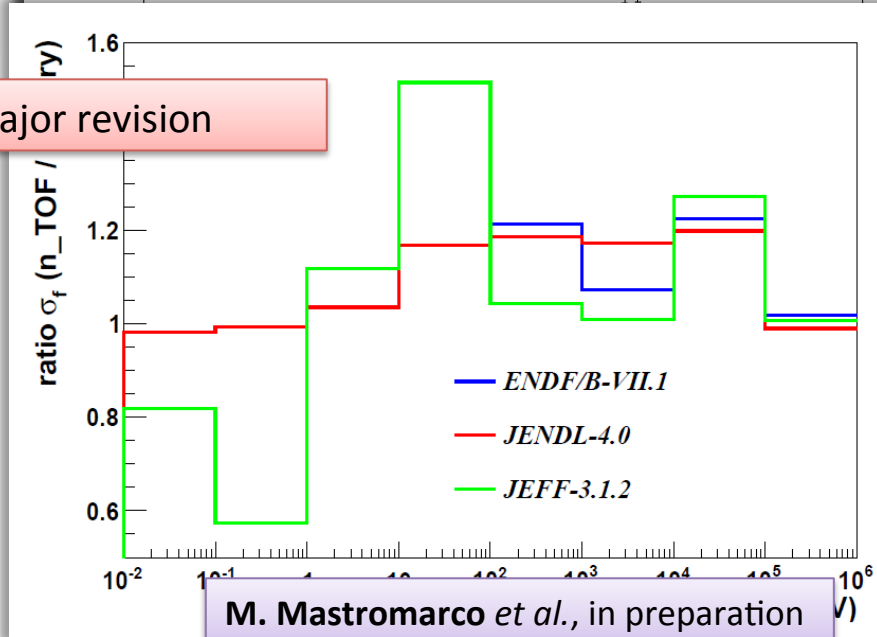
Half-life: **432 y**
 Sample: **2.26 mg (/8)**
 Activity: **35 MBq** (each sample)
 Contamination (undeclared): ^{239}Pu and $^{232\text{m}}\text{Am}$

Efficiency corrections not very accurate (due to high threshold). Normalized to Dabbs (1983), 3rd resonance.

Small (but important) **undeclared contamination** of ^{239}Pu and $^{242\text{m}}\text{Am}$ affects region $E_n < 10^4$ eV



JEFF-3.1 needs a major revision

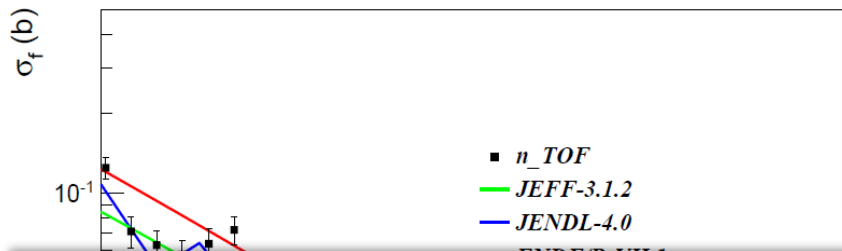
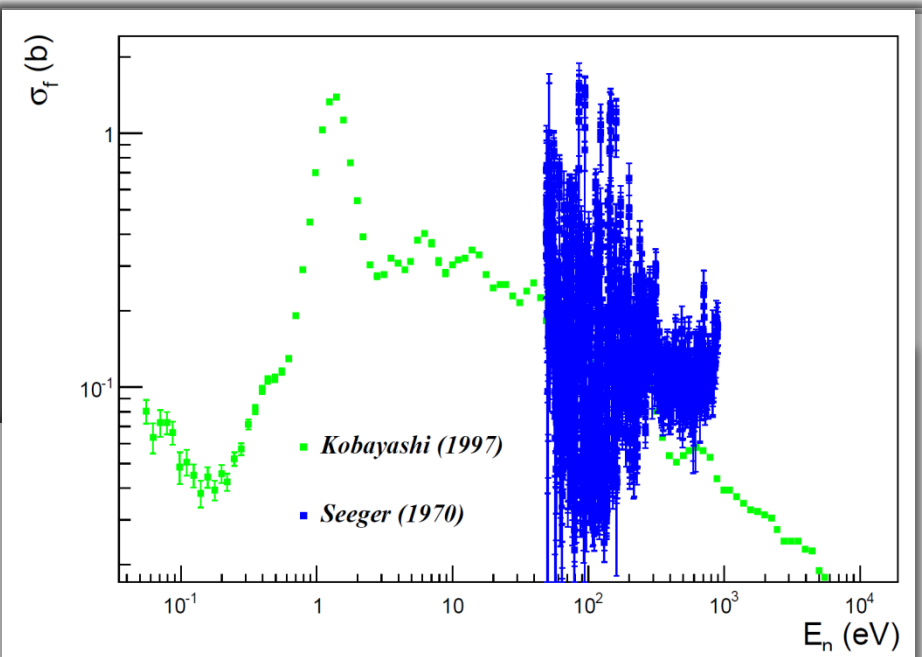


M. Mastromarco *et al.*, in preparation

The $^{243}\text{Am}(n,f)$ at low energy

Half-life: **7370 y**
 Sample: **4.8 mg (/8)**
 Activity: **4.4 MBq** (each sample)
 Contamination (declared): ^{241}Am **2.5%**
 Contamination (undeclared): ^{239}Pu , $^{242\text{m}}\text{Am}$

M. Mastromarco *et al.*, in preparation



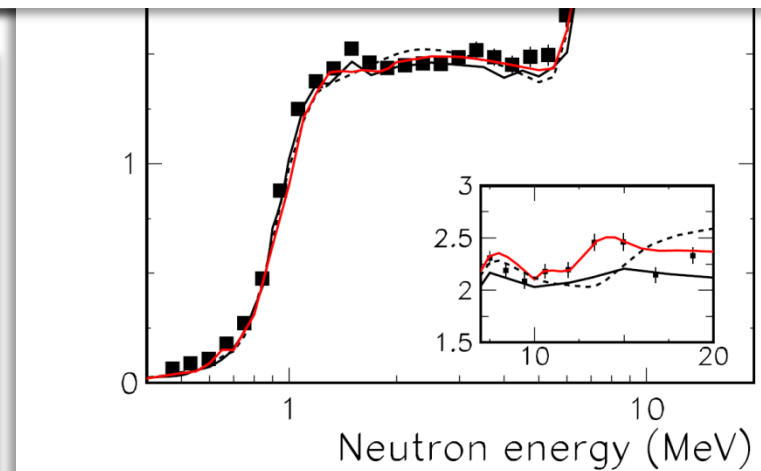
Eur. Phys. J. A (2011) 47: 160
 DOI 10.1140/epja/i2011-11160-x

Regular Article – Experimental Physics

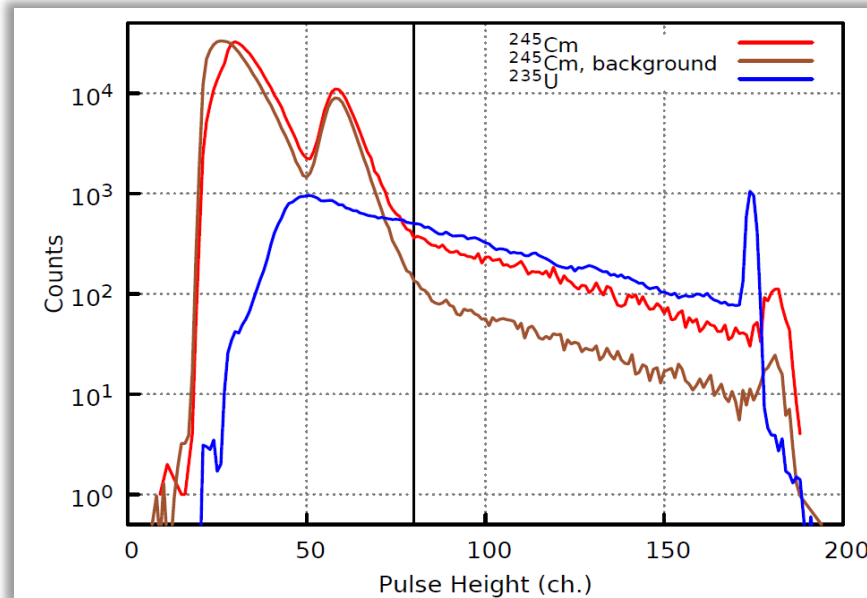
THE EUROPEAN
 PHYSICAL JOURNAL A

Measurement of the neutron-induced fission cross-section of ^{243}Am relative to ^{235}U from 0.5 to 20 MeV

F. Belloni¹, M. Calviani^{2,3}, N. Colonna⁴, P. Mastinu², P.M. Milazzo^{1,a}, U. Abbondano¹, G. Aerts⁵, H. Álvarez⁶, F. Alvarez-Velarde⁷, S. Andriamonje⁵, J. Andrzejewski⁸, L. Audouin⁹, G. Badurek¹⁰, M. Barbagallo⁴, P. Baumann¹¹, F. Bečvář¹², E. Berthoumieux⁵, F. Calviño¹³, D. Cano-Ott⁶, R. Capote^{14,15}, C. Carrapico¹⁶, P. Cennini³, V. Chepel¹⁷, E. Chiaveri³, G. Cortes¹³, A. Couture¹⁸, J. Cox¹⁸, M. Dahlfors³, S. David¹¹, I. Dillmann⁹, C. Domingo-Pardo¹⁹, W. Dridi⁵, I. Duran⁶, C. Eleftheriadis²⁰, M. Embid-Segura⁶, A. Ferrari³, R. Ferreira-Marques¹⁷, K. Fujii¹, W. Furman²¹, I. Goncalves¹⁶, E. Gonzalez-Romero⁶, A. Goverdovski²², F. Gramegna², C. Guerrero⁷, F. Gunsing⁵, B. Haas²³, R. Haight²⁴, M. Heil⁹, A. Herrera-Martinez³, M. Igashira²⁵, E. Jericha¹⁰, F. Käppeler⁹, Y. Kadi³,



The $^{245}\text{Cm}(n,f)$ reaction



Half-life: **8500 y (18.1 y)**
 Sample: **1.71 mg (/4)**
 Activity: **87 MBq** (each sample)
 Contamination (declared): ^{244}Cm **6.6%**

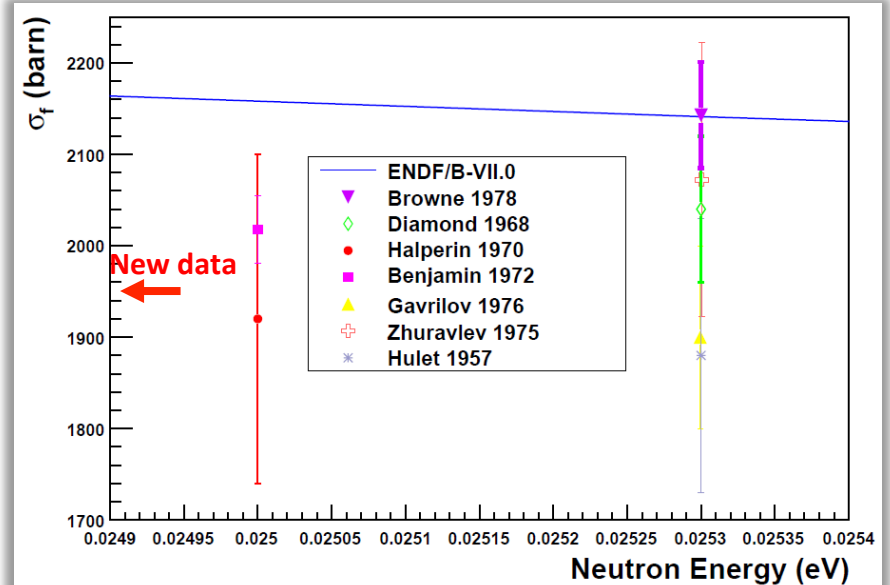
Very large α background (0.1 GBq)

High thresholds necessary (large uncertainty in efficiency corrections). Only cross section shape with good accuracy (3%).

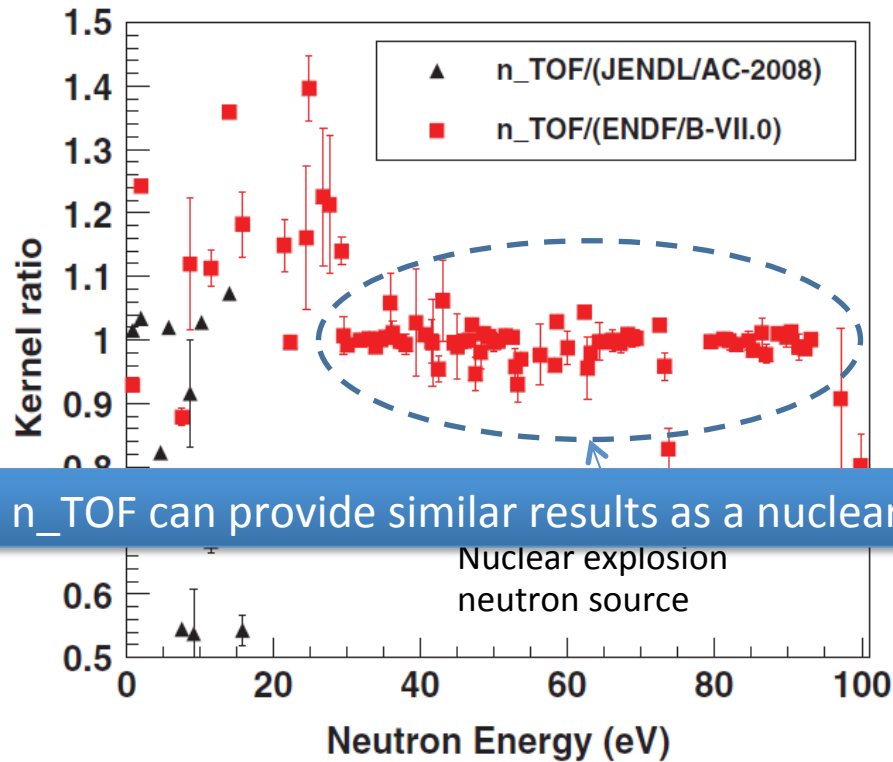
For absolute cross section, need to normalize to “recommended value” at thermal energy.

However, **large uncertainty (30%)** on thermal data.

Used two recent measurements of the thermal cross section (ILL and SCK-Mol) that agree within 5%.



The fission cross section of ^{245}Cm



Half-life: **8500 y**
 Sample: **1.71 mg**
 Activity: **87 MBq** (each sample)
 Contamination (declared): ^{244}Cm **6.6% (18.1 y)**

Below 30 eV, two (**very old**) measurements exist, showing **large discrepancies**.
 Above 30 MeV, **only one** measurement with neutrons from a **nuclear test**.

n_TOF can provide similar results as a nuclear explosion (but with fewer side effects ...)

From thermal energy to 30 eV a **revision of the evaluations** is needed.

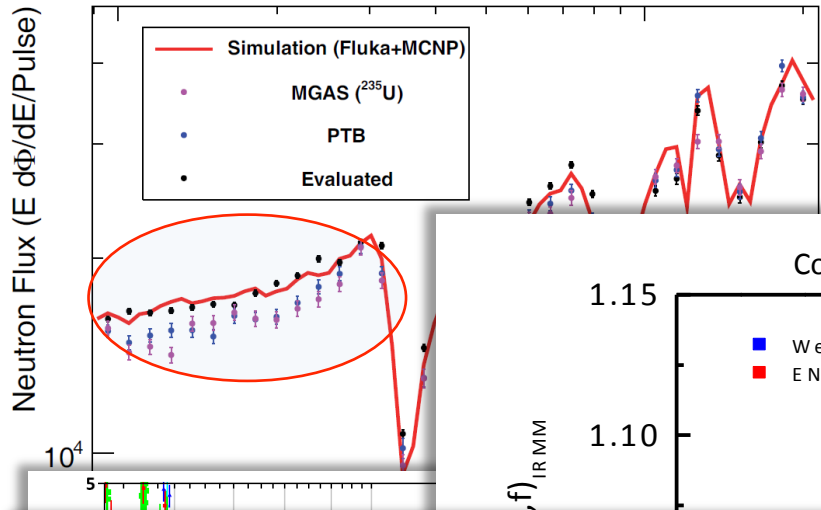
Above 30 eV, n_TOF confirm previous data and evaluations.

PHYSICAL REVIEW C **85**, 034616 (2012)

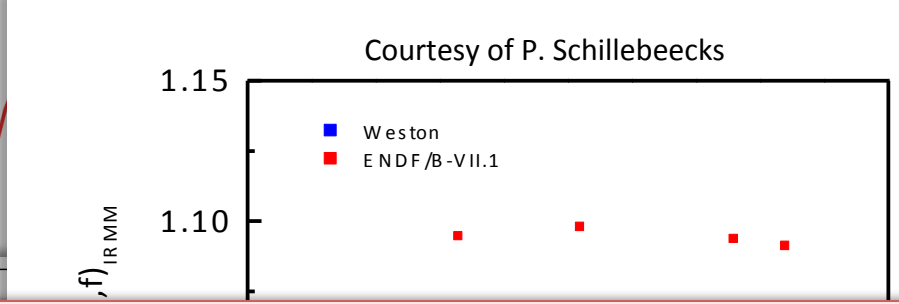
Neutron-induced fission cross section of ^{245}Cm : New results from data taken at the time-of-flight facility n_TOF

M. Calviani,^{1,2,4} M. H. Meaze,^{3,†} N. Colonna,³ J. Praena,⁴ U. Abbondanno,⁵ G. Aerts,⁶ H. Alvarez,⁷ F. Alvarez-Velarde,⁸ S. Andriamonje,^{2,6} J. Andrzejewski,⁹ P. Assimakopoulos,^{10,‡} L. Audouin,¹¹ G. Badurek,¹² M. Barbagallo,³ P. Baumann,¹³ F. Bečvář,¹⁴ F. Belloni,^{5,6} B. Berthier,¹¹ E. Berthoumieux,⁶ F. Calviño,¹⁵ D. Cano-Ott,¹⁶ R. Capote,^{4,17} C. Carrapiço,^{6,18} P. Cennini,² V. Chepel,¹⁹ E. Chiaveri,² G. Cortes,¹⁵ A. Couture,¹⁹ J. Cox,¹⁹ M. Dahlfors,² S. David,¹¹ I. Dillmann,²⁰ C. Domingo-Pardo,²¹ W. Dridi,⁶ J. Duran,⁷ C. Eleftheriadis,²² M. Embid-Segura,⁸ J. Ferrant,^{11,‡} A. Ferrari,²

$^{235}\text{U}(n,f)$ between 10 and 30 keV



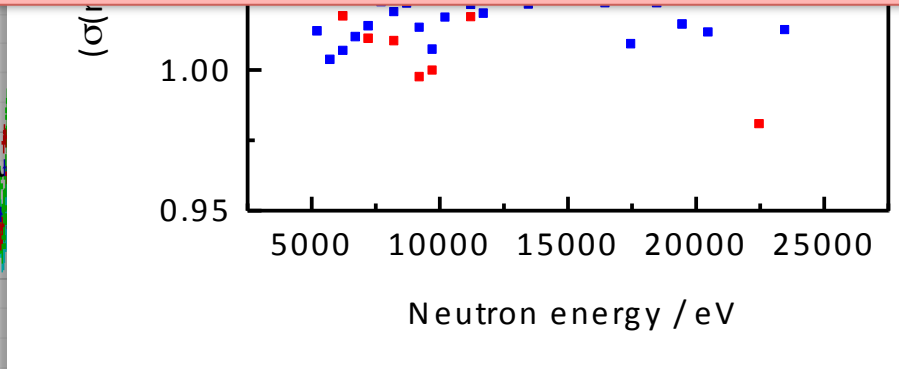
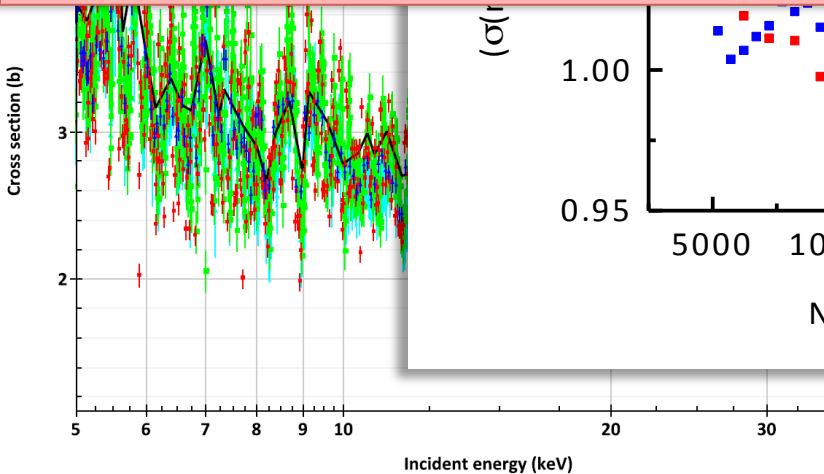
The **flux** calculated on the basis of the $^{235}\text{U}(n,f)$ cross section **found systematically lower** than “expected” in the 10-30 keV range (*M. Barbagallo et al., Eur. Phys. J A 49 (2013) 156*).



ge potentially

week ending
16 NOVEMBER 2012

Several evidences of a problem in the $^{235}\text{U}(n,f)$ cross section between 10 and 30 keV
Need to investigate it further (a new measurement is planned at n_TOF)



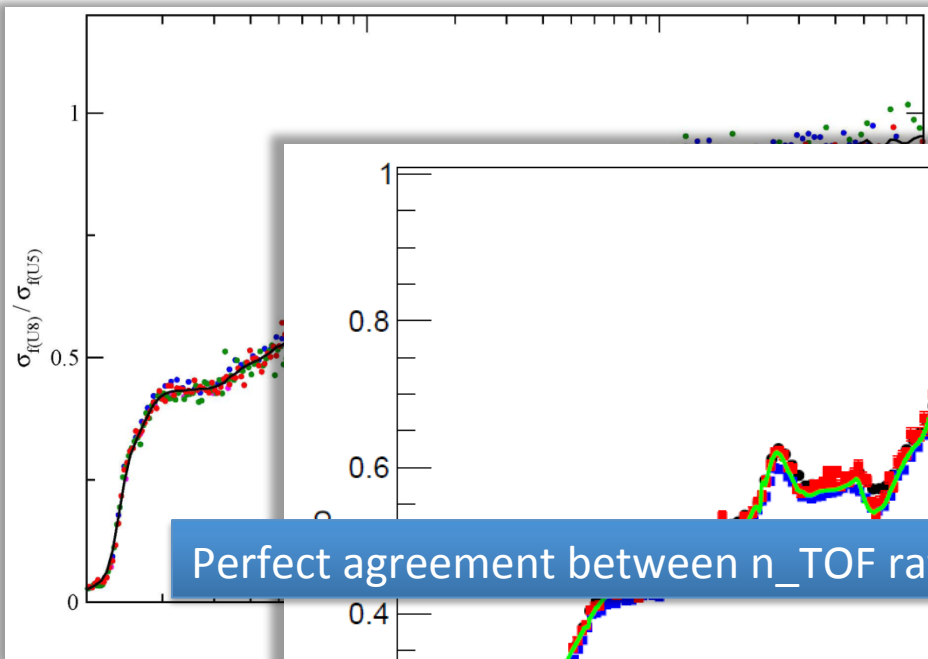
5, USA
550, USA

cross sections are
and JENDL-4.0
are observed

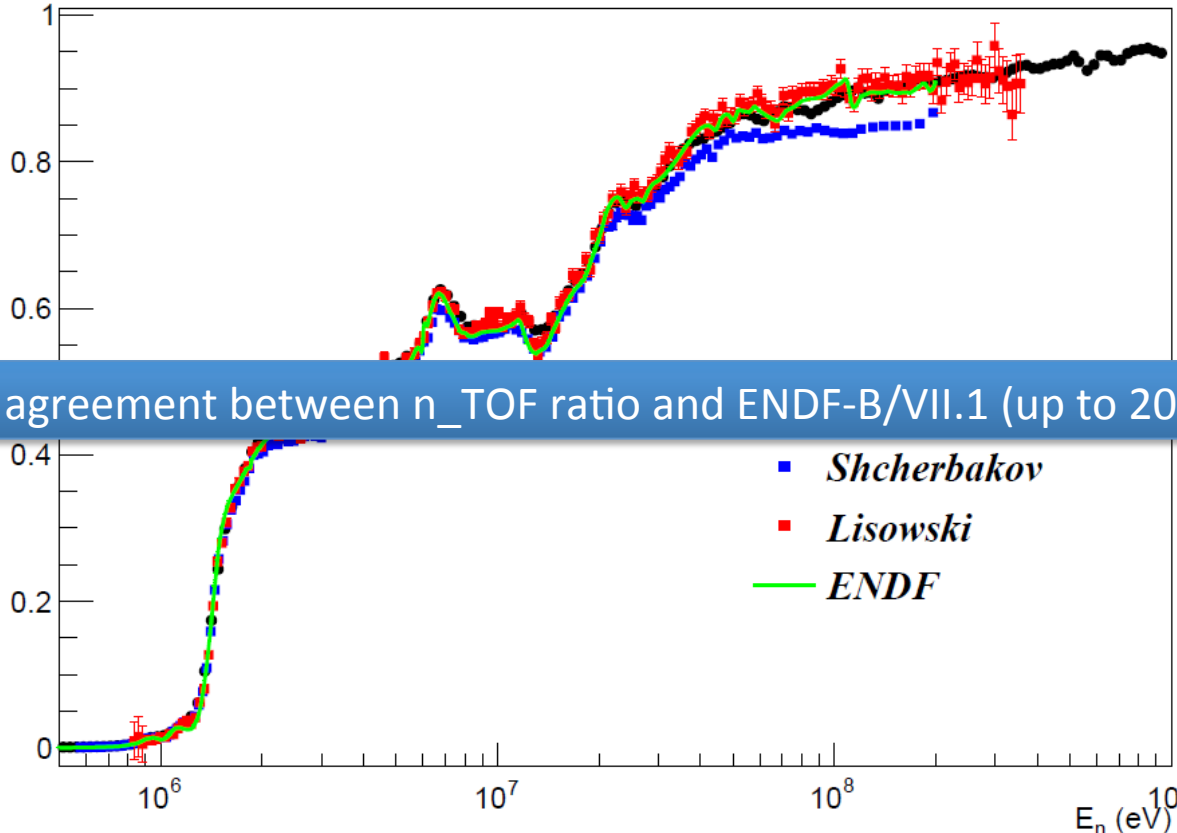
among other measurements [14–18]. Neutron flux at

$^{238}\text{U}/^{235}\text{U}(n,f)$ ratio up to 1 GeV

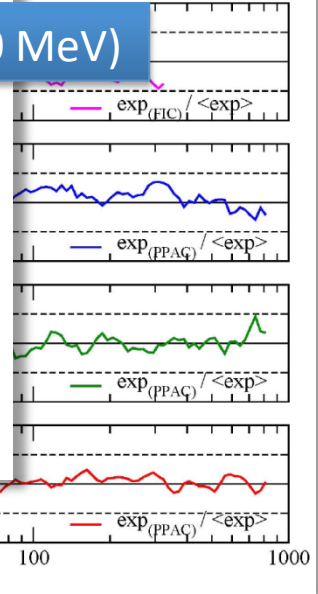
The $^{238}\text{U}/^{235}\text{U}$ cross section ratio has been measured at n_TOF up to 1 GeV. Collected and detectors and within 3%.



Perfect agreement between n_TOF ratio and ENDF-B/VII.1 (up to 200 MeV)



- *Shcherbakov*
- *Lisowski*
- *ENDF*



All n_TOF data are in good order to collect $^{238}\text{U}/^{235}\text{U}$ cross section ratio.

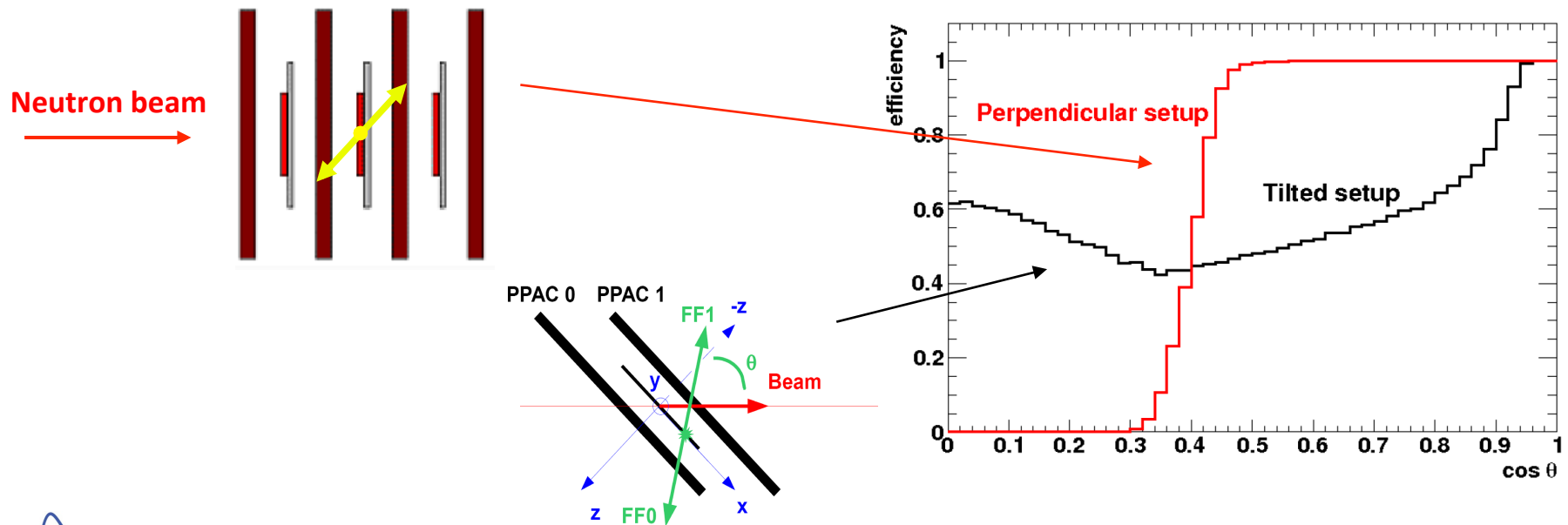


Angular distribution of FF

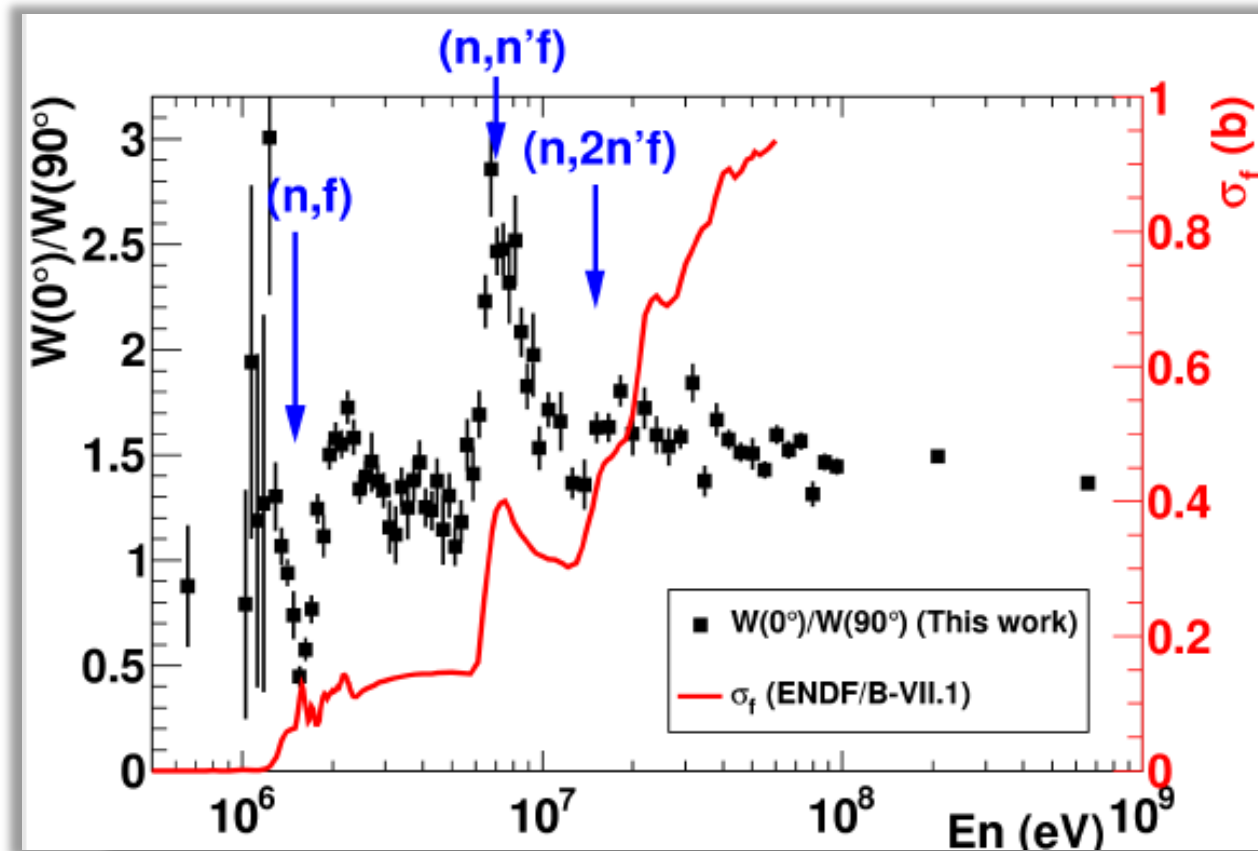
Fission Fragment angular distribution important to:

- obtain information on the state of the nucleus at saddle point (spin, parity, ...) and on the **fission dynamics**
- calculate more **reliable detection efficiency**, thus improving accuracy of cross sections.

The effect on the cross section is particularly important **for coincidence technique (PPAC,** due to backing the angular acceptance is limited to 65°)

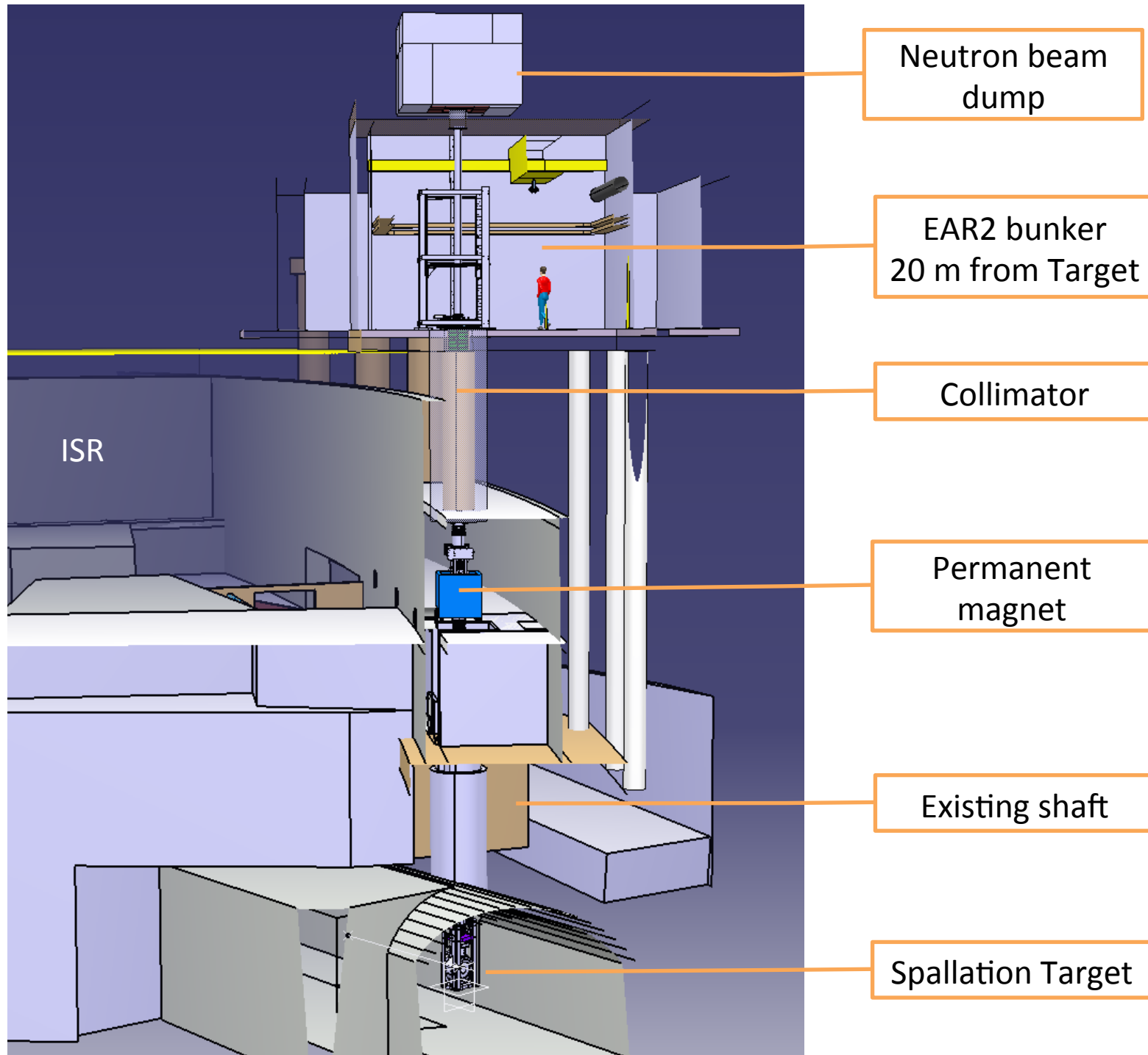


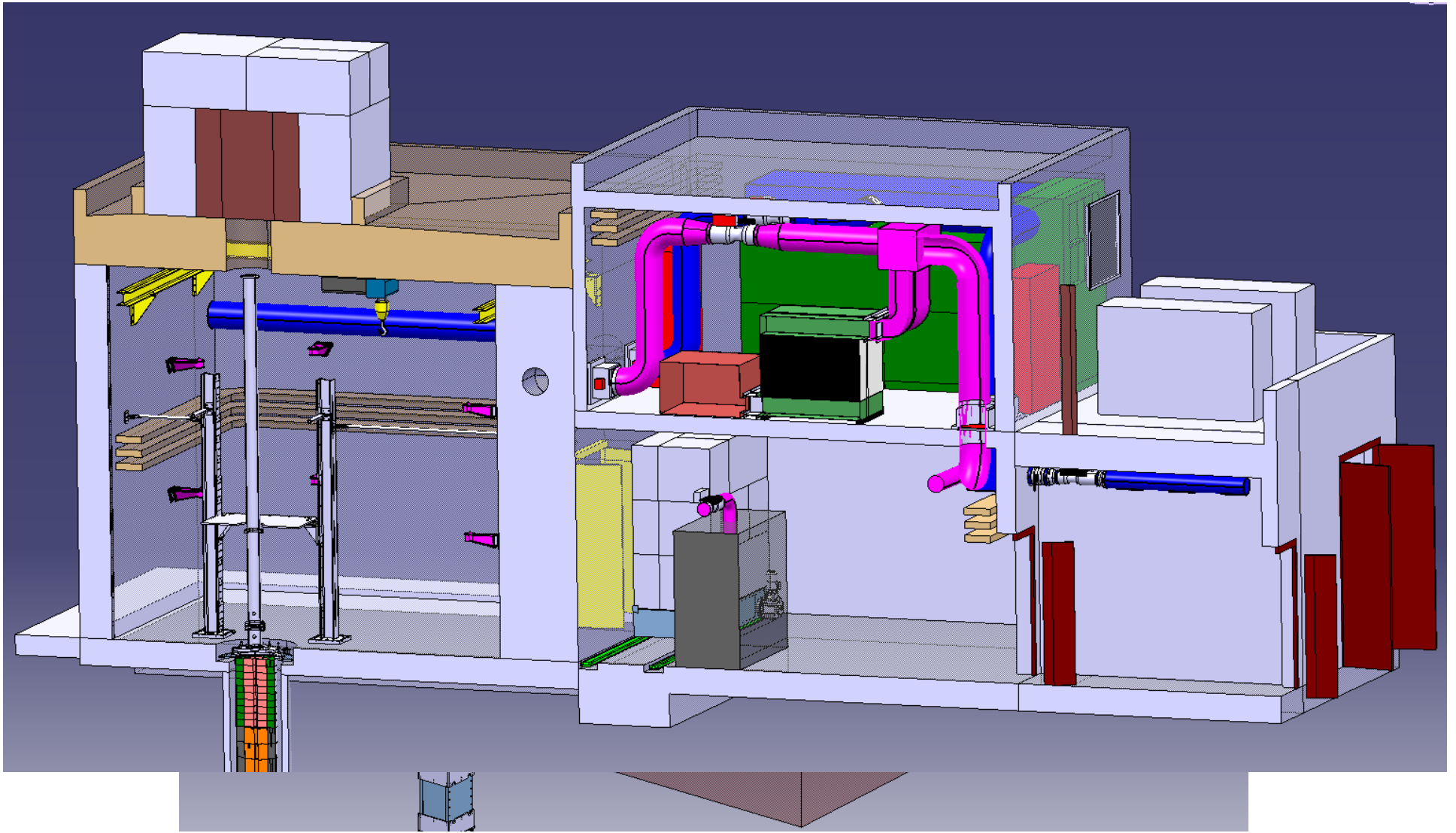
Angular anisotropy in ^{232}Th fission reaction



Measured anisotropy from fission threshold to 1 GeV !!!

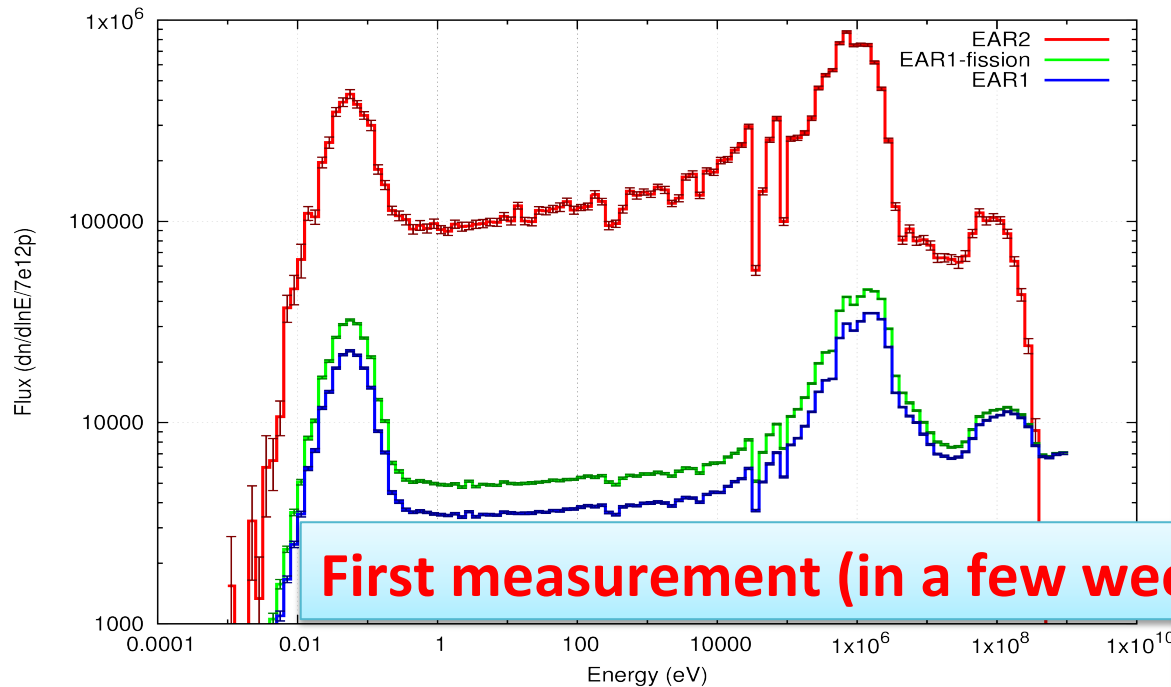
The second experimental area at n_TOF







Main features of EAR 2



Higher fluence, by a factor of 25, relative to EAR1.

The **shorter flight path** implies a factor of 10 smaller time-of-flight.

Global gain by a factor of **250 in the signal/background ratio** for radioactive isotopes !!!!!

The huge gain in signal-to-background ratio in EAR2 allows to measure cross sections of **radioactive isotopes as low as a few years.**

First measurement (in a few weeks): $^{240}\text{Pu}(n,f)$

Fission measurements foreseen in EAR2:

- ^{238}Pu (87.7 y), ^{241}Pu (14.1 y), ^{244}Cm (18.1 y)
- ^{232}U (70 y) - cross section and FF angular distribution
- ^{230}Th (available in small amount)

Conclusions



- There is need of **accurate new data** on neutron-induced fission cross-sections for **advanced nuclear technology**.
- Since 2001, **n_TOF@CERN** has provided an important contribution to the field, with several measurements on **long-lived actinides**.
- Results obtained so far can help improve current evaluated databases and models of fission reactions.
- A second **experimental area at 20 m** has just been built and is now being commissioned.
- It will open **new perspectives** for frontier measurements on short-lived radionuclides.



WORK IN PROGRESS

Thank you