Neutron capture and fission reactions on ^{235}U: cross sections, α-ratios and prompt fission γ-rays

C. Guerrero and E. Berthoumieux — *CERN (Geneva, Switzerland)*

D. Cano-Ott, E. Gonzalez and E. Mendoza — *CIEMAT (Madrid, Spain)*

M. Sabate — *Universidad de Sevilla (Seville, Spain)*

(*The n_TOF Collaboration*, www.cern.ch/nTOF)

Wonder 2012: 3rd International Workshop on Nuclear Data Evaluation for Reactor Applications

Organised by CEA and NEA | 25-28 September 2012 | Aix-en-Provence, France

Motivation: capture cross sections and α-ratios

The criticality of current and fast future reactors must be known within 0.3-0.5% for operation/safety.

NEA Nuclear Energy Agency
Nuclear Data Services
NEA Nuclear Data High Priority Request List

(FCA) Fast Critical Assembly (JAEA)

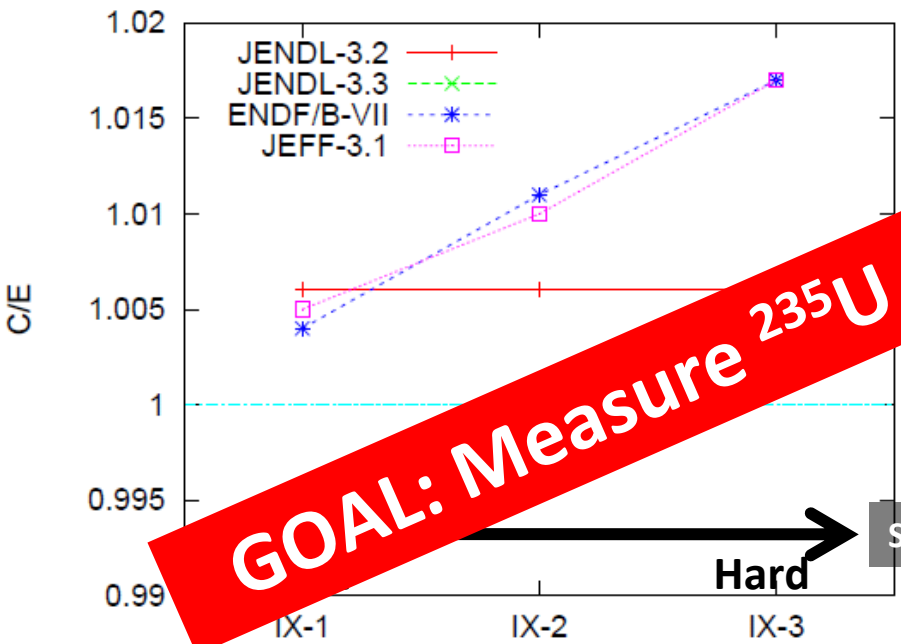
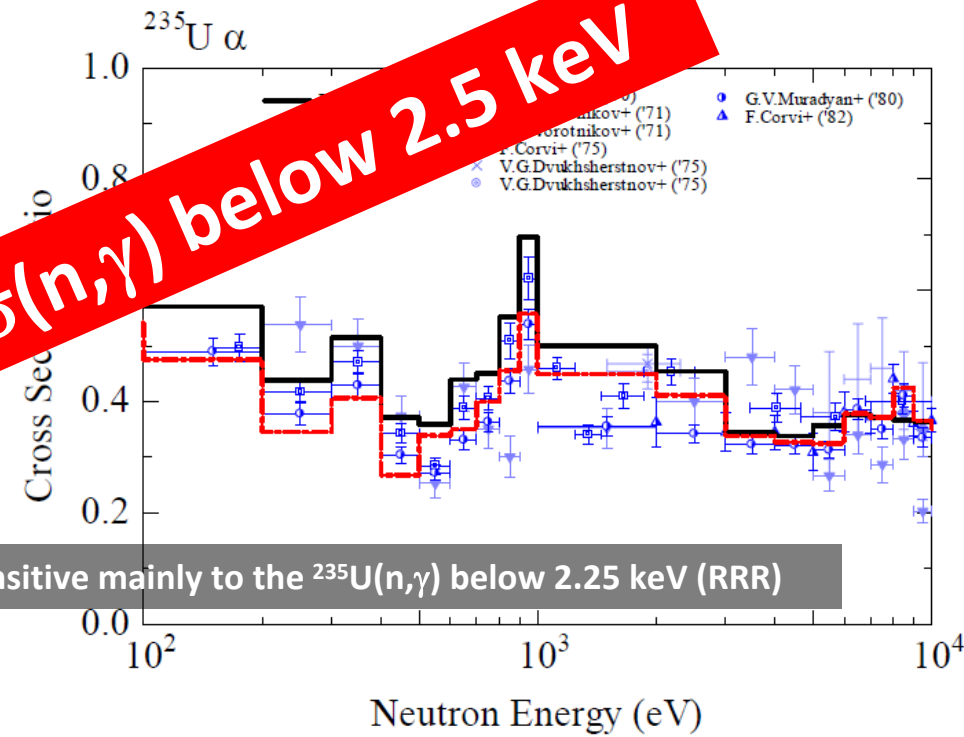


Figure 2: C/E values of criticality of FCA IX assemblies



GOAL: Measure ^{235}U σ(n,γ) below 2.5 keV

Sensitive mainly to the ^{235}U(n,γ) below 2.25 keV (RRR)

Differences up to 2% in the measured and calculated criticality values for FCA (JAERI, Japan) assemblies with different hardness are due to ^{235}U σ(n,γ) below 2.5 keV.

Motivation: prompt fission γ-rays

NEA
Nuclear Energy Agency

Nuclear Data Services

NEA Nuclear Data High Priority Request List

IMPACT

The four fast reactor systems of GenIV feature innovative core characteristics for which **gamma-ray heating estimates for non-fuel zones require an uncertainty of 7.5%. A similar requirement appears for the experimental Jules Horowitz Reactor (RJH) at Cadarache**. Recent studies show evidence of discrepancies on integral measurement in MASURCA, EOLE and MINERVE, from which it is clear that the expectations for GenIV systems and the RJH thermal reactor are not met. Gamma-ray energy release is dominated by ^{239}Pu and ^{235}U.

ACCURACY

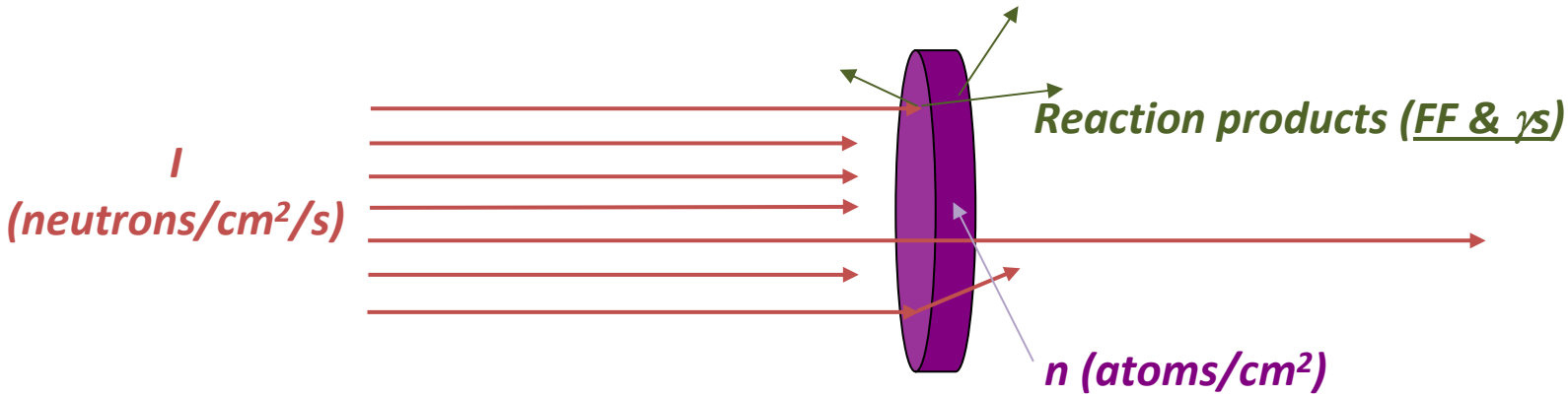
Observed: Discrepancies for C/E ratios in various benchmarks range from 10 to 28%.
Target: 7.5% on the total gamma energy and multiplicity
Target: Best accuracy achievable for the gamma spectrum shape

COMMENT FROM REQUESTER

Forty percent of the total gamma-ray energy release results from prompt decay of fission products. No comprehensive analytic expressions exist and Hauser-Feshbach model calculations are involved and presently lack sufficient knowledge to warrant a solution of the problem. **New measurements would be needed to guide new evaluation efforts**. Present evaluations are based on measurements from the seventies.

Measuring technique

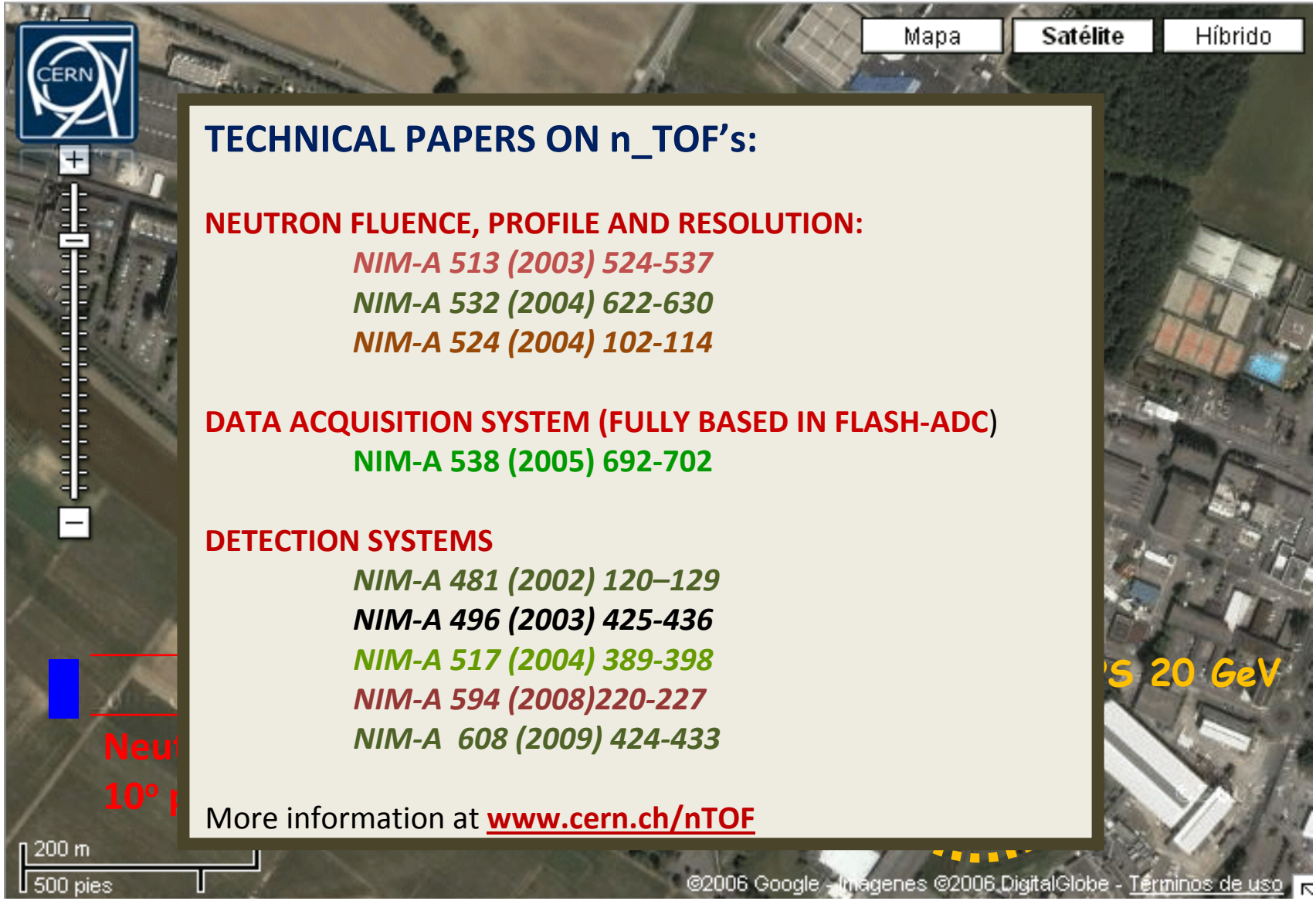
Thermal-epithermal neutrons induce both (n,γ) and (n,f) reactions, both emitting γ-rays



MEASURING THE NEUTRON CROSS SECTIONS & γ-RAY EMISSION REQUIRES

- A facility providing a neutron beam (The n_TOF facility).
- A highly pure sample.
- A detection system for **detecting simultaneously fission fragments and γ-rays**
- The analysis tools to determine the measured cross sections with the required accuracy.

A Google view of n_TOF



TECHNICAL PAPERS ON n_TOF's:

NEUTRON FLUENCE, PROFILE AND RESOLUTION:

NIM-A 513 (2003) 524-537
NIM-A 532 (2004) 622-630
NIM-A 524 (2004) 102-114

DATA ACQUISITION SYSTEM (FULLY BASED IN FLASH-ADC)

NIM-A 538 (2005) 692-702

DETECTION SYSTEMS

NIM-A 481 (2002) 120–129
NIM-A 496 (2003) 425-436
NIM-A 517 (2004) 389-398
NIM-A 594 (2008)220-227
NIM-A 608 (2009) 424-433

More information at **www.cern.ch/nTOF**

Experimental set-up: The TAC and MGAS detectors

We need to detect capture and fission reactions simultaneously!

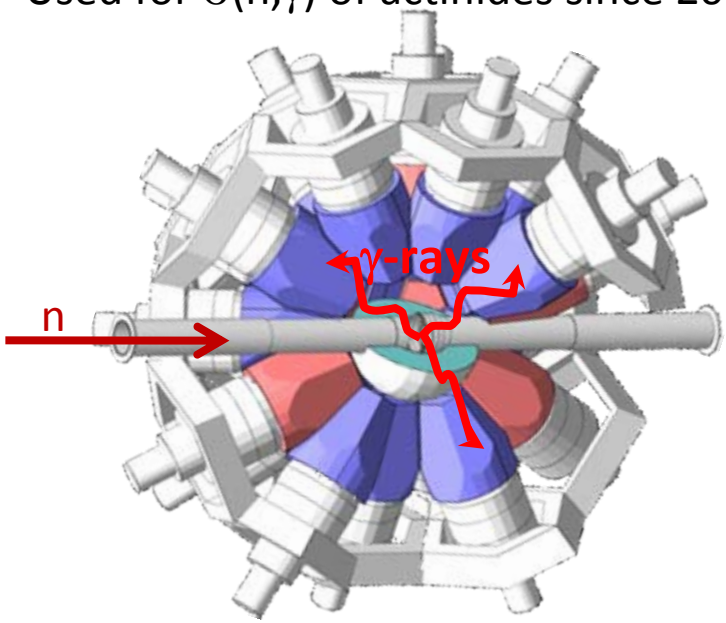
Total Absorption Calorimeter (TAC) for (n,γ)

40 BaF_2 crystals

4π geometry (95% coverage)

16% energy resolution at 662 keV

Used for σ(n,γ) of actinides since 2004



Results: distributions E_{sum}, m_{cr} & E_n

C. Guerrero et al., NIM-A 608 (2009) 424-433

MicroMegas (MGAS) for (n,f)

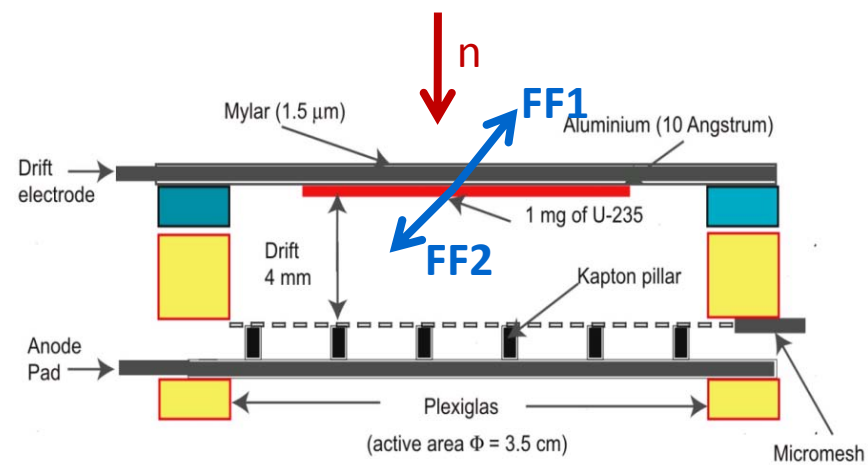
Based on Bulk technology

Double stage gas detector:

conversion +amplification

~90% efficiency for FF. FF.

Used for neutron monitoring since 2009

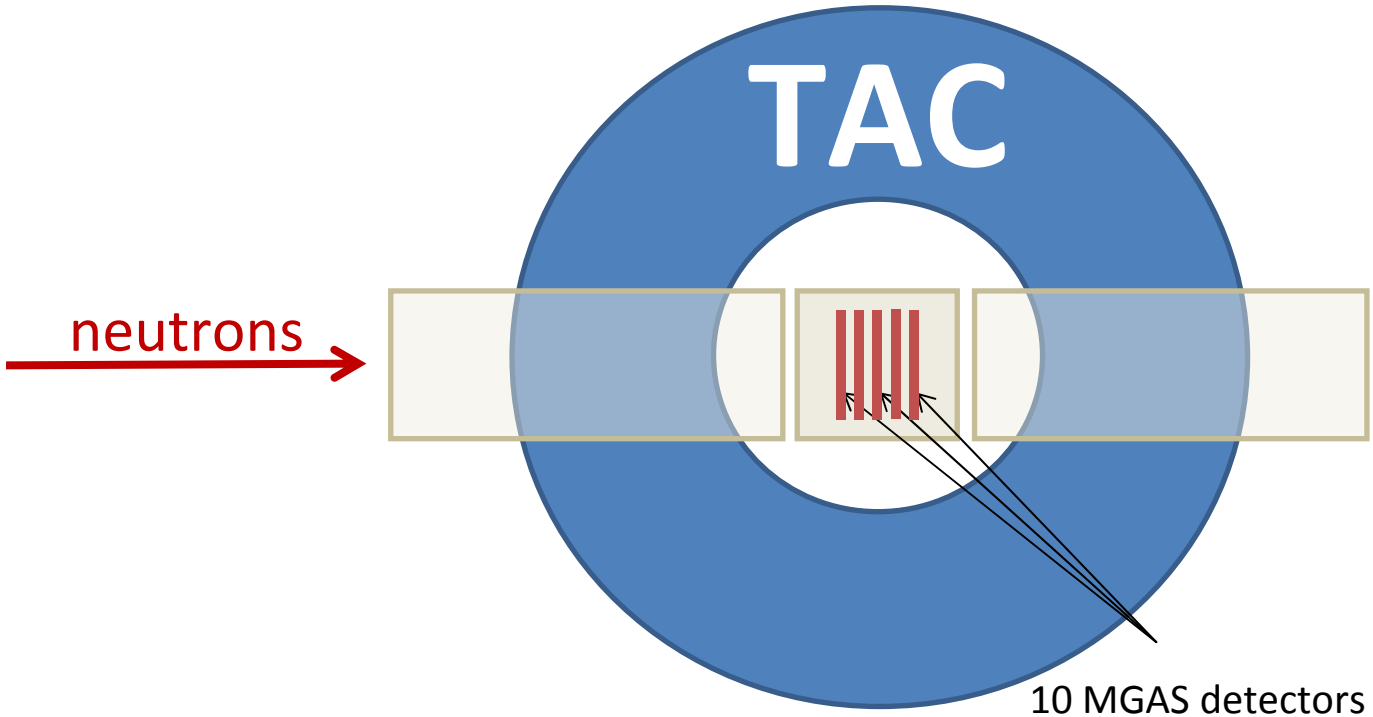


Results: distributions Amp. & E_n

S. Andriamonje et al., NIM-A 481 (2002) 120–129

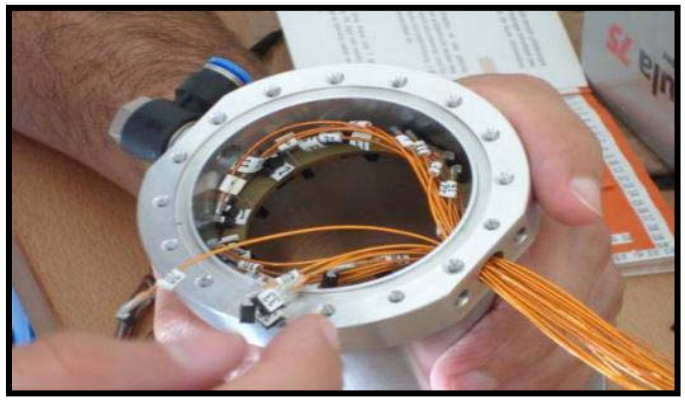
Experimental set-up (2012): Combination of the TAC and MGAS

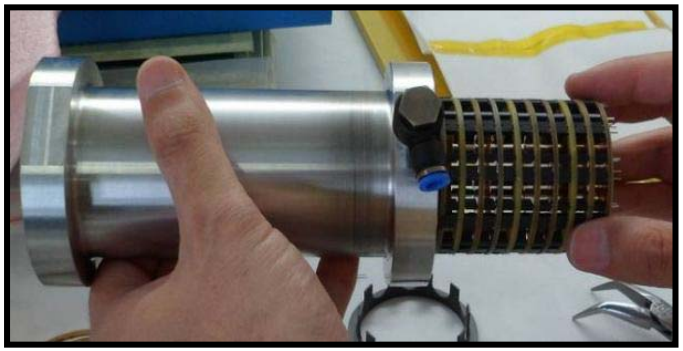
- 10 ^{235}U samples of 300 μg/cm^2 (42 mm diameter)
- MGAS filled with with Ar/CF4/isobutane at 1 atm
- TAC and MGAS signals digitized at 250 MS/s and 100 MS/s, respectively.

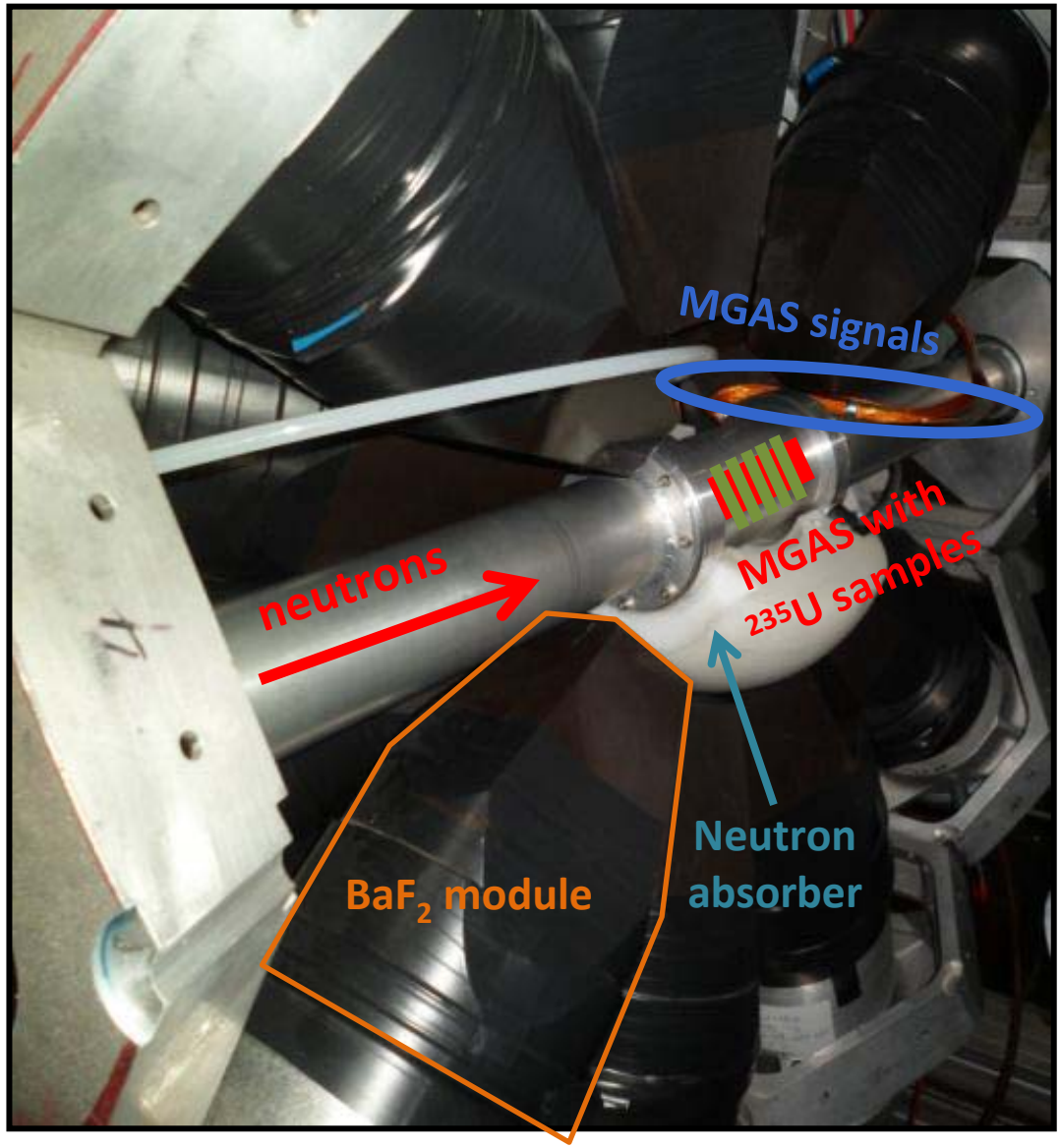


10 MGAS detectors (5 back-to-back assemblies) each equipped with a **4.15 mg ^{235}U** sample (supplied by JRC-IRMM)

Experimental set-up (2012): Combination of the TAC and MGAS







CERN-INTC-2010-037 / INTC-I-105
21/05/2010

Validation of simultaneous measurement of capture and fission reactions at n_TOF

Letter Of Intent to the ISOLDE and Neutron Time-of-Flight Committee

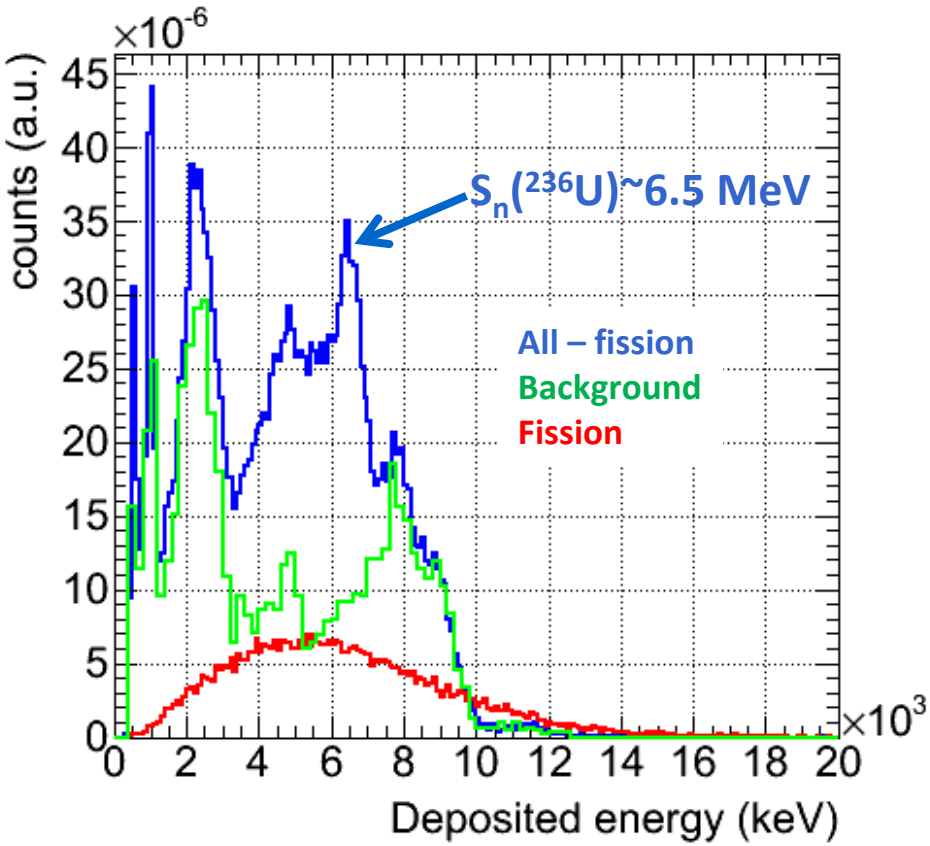
Spokespersons: C. Guerrero[1] and E. Berthoumieux[2]
Technical coordinator: V. Vlachoudis[3]

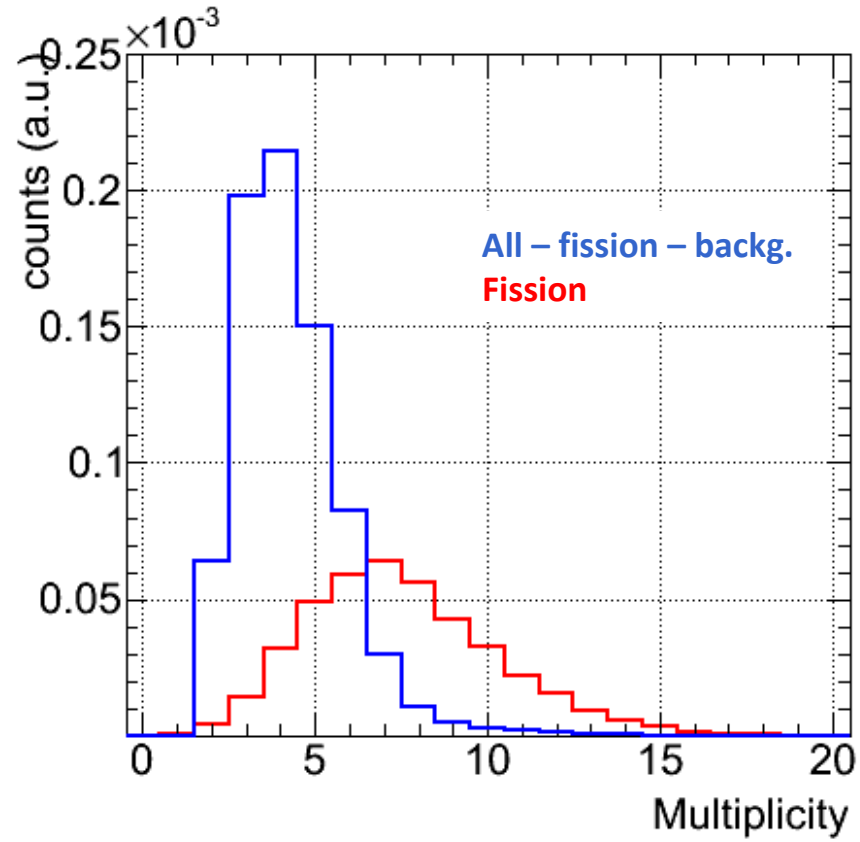
C. Guerrero[1], E. Berthoumieux[2], S. Andriamonje[3], D. Cano-Ott[1], E. Gonzalez-Romero[1], F. Gunsing[2], T. Martinez[1], E. Mendoza[1], M. Calviani[3] and The n_TOF Collaboration (http://cern.ch/n TOF/)

[1]Centro de Investigaciones Energéticas Medioambientales y Tecnológicas – CIEMAT, Madrid, Spain
[2]CEA Saclay, IRFU, F-91191 Gif-sur-Yvette, France
[3]CERN, Geneva, Switzerland

Measurement (2010): E_{sum} and $m_{crystal}$ distributions

Deposited energy (m_{cr}>2) and multiplicity (E_{sum}>3)distributions corresponding to resonances:





Measurement (2010): Detection efficiencies

With two different detectors and two different types of reactions to detect, it is important to define clearly the different efficiencies that play a role in the measurement and their interrelations.

$\varepsilon_{MGAS}(n,f)$, $\varepsilon_{TAC}(n,f)$ and $\varepsilon_{TAC}(n,\gamma)$

When a fission reaction occurs, it can be detected:

a) in both detectors, → $\varepsilon_{MGAS}(n,f) \cdot \varepsilon_{TAC}(n,f)$

b) in none of them, → $(1- \varepsilon_{MGAS}(n,f)) \cdot (1- \varepsilon_{TAC}(n,f))$

c) only in the MGASs → $\varepsilon_{MGAS}(n,f) \cdot (1- \varepsilon_{TAC}(n,f))$

d) only in the TAC. → $(1- \varepsilon_{MGAS}(n,f)) \cdot \varepsilon_{TAC}(n,f)$

When a neutron capture occurs, it can only be detected in the TAC → $\varepsilon_{TAC}(n,\gamma)$

The efficiency for detecting fission reactions in each detector is independent from the other, but the calculation from experimental data requires that these four probabilities are properly taken into account.

Measurement (2010): $\varepsilon_{MGAS}(n,f)$, $\varepsilon_{TAC}(n,f)$ and $\varepsilon_{TAC}(n,\gamma)$

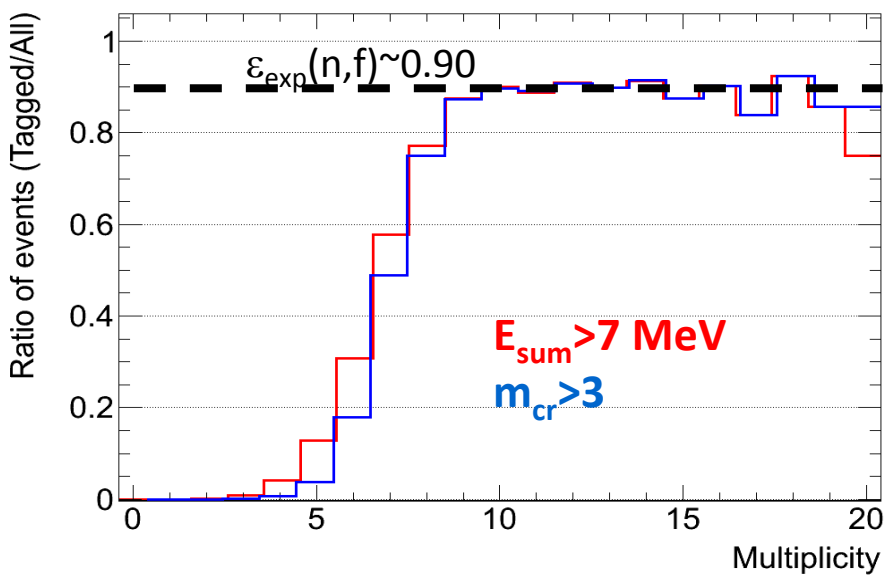
Calculation of $\varepsilon_{MGAS}(n,f)$

MC simulations

Samples are 318 μg/cm^2, nearly identical to those of the ^{235}U samples (316 mg/cm^2) used in FIC, for which simulations with FLIKA give **$\varepsilon_{MC}(n,f)$~0.94** (6% losses due to absorption in the sample).

Experimentally:

Fission events produce high-energy, high-multiplicity TAC events. Assumption → ε_{TAC} ~100% for such events. Then, the detection efficiency of the MGASs can be calculated as the **ratio of *tagged* to *all*** events for multiplicities higher than ~10 (no capture events).



$$\underbrace{\varepsilon_{MC}(n,f)\sim0.94 \;\&\; \varepsilon_{exp}(n,f)\sim0.90}_{\varepsilon_{MGAS}(n,f)\sim0.92}$$

Measurement (2010): $\varepsilon_{MGAS}(n,f)$, $\varepsilon_{TAC}(n,f)$ and $\varepsilon_{TAC}(n,\gamma)$

Calculation of $\varepsilon_{TAC}(n,f)$

A coincident event in the TAC is found for 97% of the MGAS events (MGASamp>20 channels). This value represents the TAC efficiency for fission events, $\varepsilon_{TAC}(n,f)$, and is very similar to the efficiency of $\varepsilon_{TAC}(n,\gamma)$=0.974(4) for capture events in ^{197}Au (from GEANT4 Monte Carlo simulations).

The efficiency $\varepsilon_{TAC}(n,f)$ depends on the analysis conditions for the deposited energy and multiplicity values.

	$m_{cr}>0$	$m_{cr}>1$	$m_{cr}>2$	$m_{cr}>3$		$0<m_{cr}<9$	$1<m_{cr}<9$	$2<m_{cr}<9$	$3<m_{cr}<9$
$0.1<E_{sum}$	96.9	94.3	90.1	83.5		71.0	68.3	64.1	57.5
$1<E_{sum}$	93.3	92.7	89.7	83.4		67.3	66.7	63.7	57.4
$2<E_{sum}$	87.9	87.7	86.0	81.8		62.0	61.8	60.2	55.8
$3<E_{sum}$	79.9	79.8	79.0	76.5		53.9	53.8	53.1	50.5
$0.1<E_{sum}<7$	64.8	62.2	57.9	51.5		58.3	55.7	51.5	45.0
$1<E_{sum}<7$	61.1	60.6	57.6	51.4		54.7	54.1	51.1	45.0
$2<E_{sum}<7$	55.8	55.6	54.0	49.8		49.3	49.1	47.6	43.3
$3<E_{sum}<7$	47.7	47.6	46.9	44.5		41.2	41.2	40.4	38.0

Efficiency of the TAC for detecting fission events under different conditions in deposited energy and crystal multiplicity.

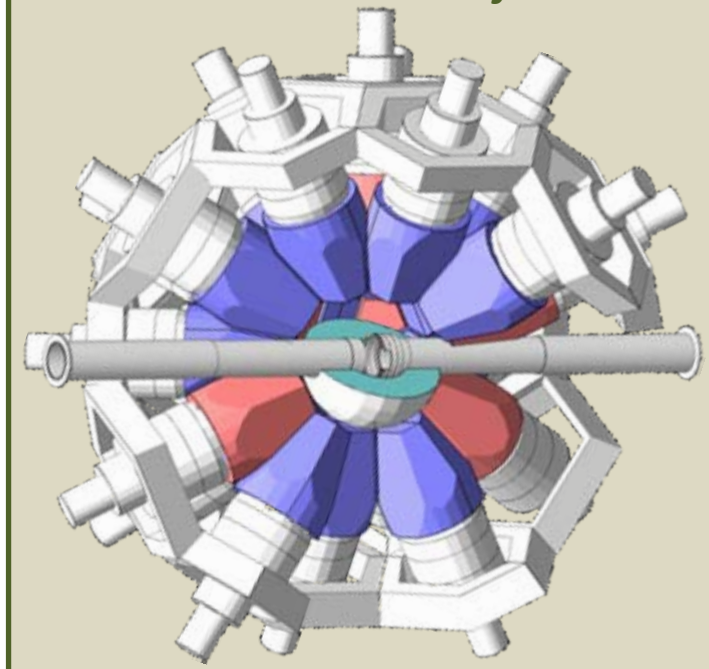
Measurement (2010): $\varepsilon_{MGAS}(n,f)$, $\varepsilon_{TAC}(n,f)$ and $\varepsilon_{TAC}(n,\gamma)$

Calculation of $\varepsilon_{TAC}(n,\gamma)$

The detection efficiency $\varepsilon_{TAC}(n,\gamma)$ can be calculated accurately by means of Monte Carlo simulations when both the experimental set-up and the details of the capture cascades are properly considered.

- Already done for ^{237}Np, ^{240}Pu, ^{241,243}Am, and ^{233}U.
- ^{235}U(n,γ) still to be done

Geant4 simulation of the TAC



C. Guerrero et al., NIM-A 671 (2012) 108-117

Approximation:

^{235}U is very similar to ^{237}Np
- Odd nuclei
- Similar level spacing (~0.5 eV)
- Similar Binding Energies (S_n~6 MeV)
- Cut at 2.5 MeV (0.46*S_n) is to ^{237}Np like 3 MeV is to ^{235}U

$\varepsilon_{TAC}(n,\gamma)$=0.70(3)
[E_{sum}>3 MeV and m_{cr}>2]

Measurement (2010): Results and publication

Test of TAC+MGAS with ^{235}U@ n_TOF

- ✓ Discrimination (n,γ) vs. (n,f)
- ✓ Normalization to σ(n,f)
- ✓ Efficiency correction
- ✓ Background subtraction
- ✓ Identification of impurities

<u>Agreement with evaluations at low E_n</u>

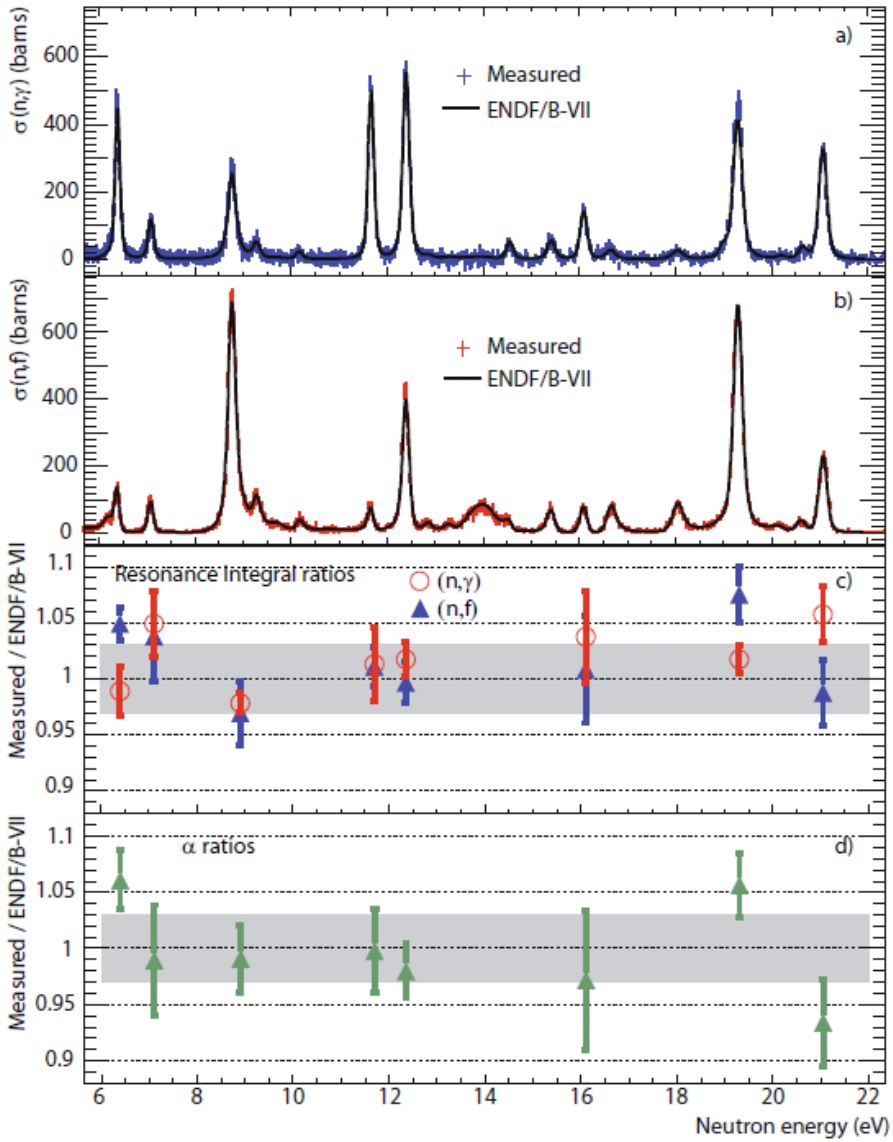
Eur. Phys. J. A (2012) **48**: 29
DOI 10.1140/epja/i2012-12029-2

THE EUROPEAN PHYSICAL JOURNAL A

Tools for Experiment and Theory

Simultaneous measurement of neutron-induced capture and fission reactions at CERN

C. Guerrero[1,2a], E. Berthoumieux[1,7], D. Cano-Ott[2], E. Mendoza[2], S. Andriamonje[1], J. Andrzejewski[4], L. Audouin[3], M. Barbagallo[10], V. Bécares[2], F. Bečvář[5], F. Belloni[6], J. Billowes[26], M. Brugger[1], M. Calviani[1], F. Calviño[8], C. Carrapiço[9], F. Cerutti[1], E. Chiaveri[1], M. Chin[1], N. Colonna[10], G. Cortés[8], M.A. Cortés-Giraldo[11], M. Diakaki[25], I. Dillmann[12], C. Domingo-Pardo[13], I. Duran[14], C. Eleftheriadis[28], M. Fernández-Ordóñez[2], A. Ferrari[1], S. Ganesan[15], G. Giubrone[16], M.B. Gómez-Hornillos[8], I.F. Gonçalves[9], E. González-Romero[2], F. Gramegna[17], E. Griesmayer[18], F. Gunsing[7], D. Jenkins[27], E. Jericha[18], Y. Kadi[1], F. Käppeler[18], D. Karadimos[25], J. Kroll[5], M. Krtička[5], E. Lebbos[1], C. Lederer[20], H. Leeb[18], R. Losito[1], M. Lozano[11], A. Manousos[28], J. Marganiec[2], S. Marrone[10], T. Martinez[2], C. Massimi[21], P.F. Mastinu[17], M. Meaze[10], A. Mengoni[22], P.M. Milazzo[6], C. Paradela[14], A. Pavlik[20], J. Perkowski[4], R. Plag[13], J. Praena[11], J.M. Quesada[11], T. Rauscher[23], R. Reifarth[13], F. Roman[1,24], C. Rubbia[1], R. Sarmento[9], G. Tagliente[10], J.L. Tain[16], D. Tarrío[14], L. Tassan-Got[3], A. Tsinganis[251], G. Vannini[21], V. Variale[10], P. Vaz[9], A. Ventura[22], M. Vermeullen[27], V. Vlachoudis[1], R. Vlastou[25], A. Wallner[20], T. Ware[26], C. Weiß[18], and T. Wright[26]



CERN-INTC-2011-045 / INTC-P-309
06/10/2011

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurements of neutron-induced capture and fission reactions on ^{235}U: cross sections and α ratios, photon strength functions and prompt γ-ray from fission

October, 5 2011

C. Guerrero[1], E. Berthoumieux[2], D. Cano-Ott[3] and the n_TOF Collaboration[4]

[1] CERN, Geneve, Switzerland
[2] CEA, Saclay, France
[3] CIEMAT, Madrid, Spain
[4] www.cern.ch/nTOF

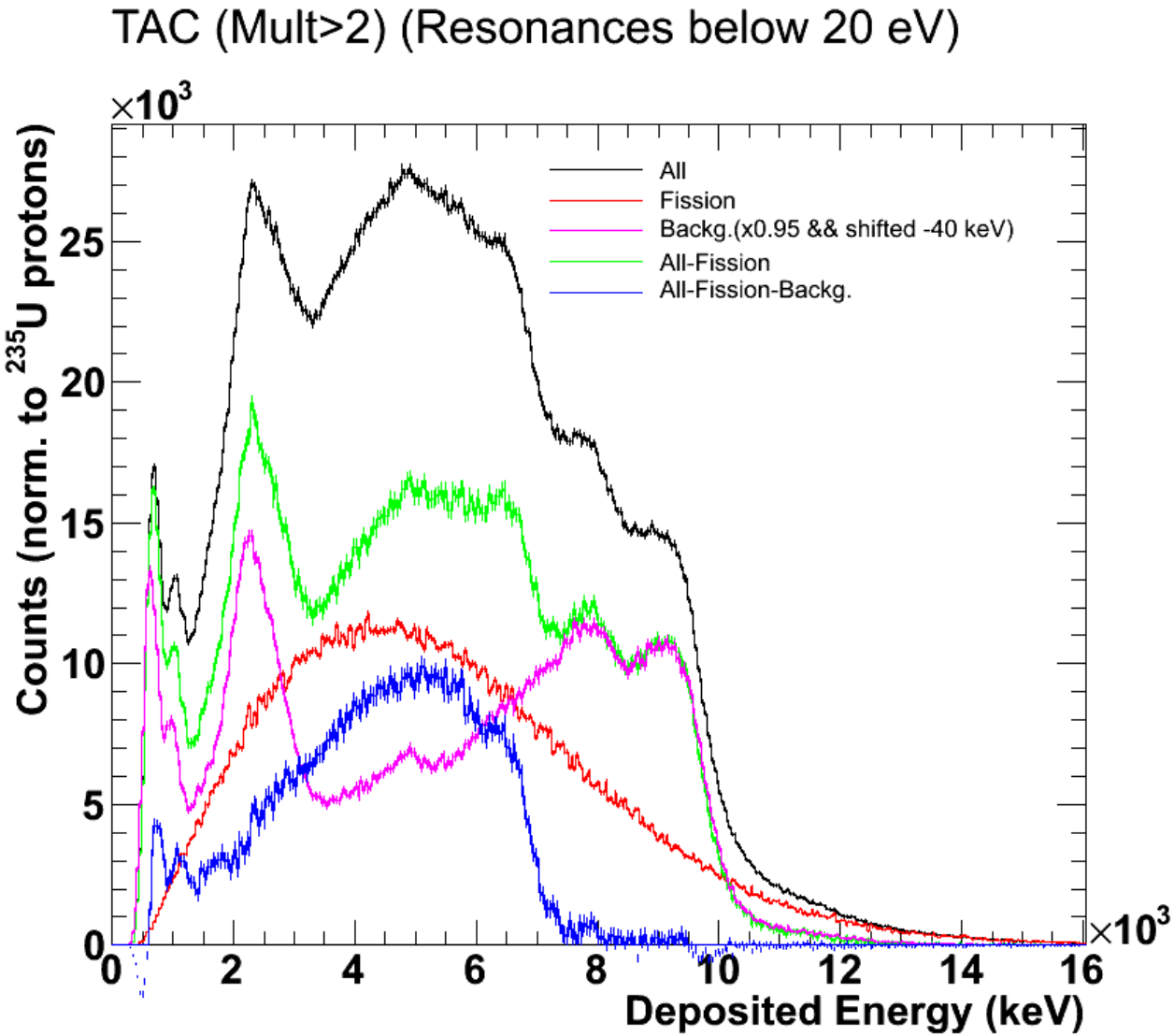
Spokesperson(s): C. Guerrero (carlos.guerrero@cern.ch), E. Berthoumieux (eric.berthoumieux@cern.ch), D. Cano-Ott (daniel.cano@ciemat.es)
Technical coordinator: E. Berthoumieux (eric.berthoumieux@cern.ch)

Data taking ongoing at CERN !!!

Compared to 2010 test measurement:

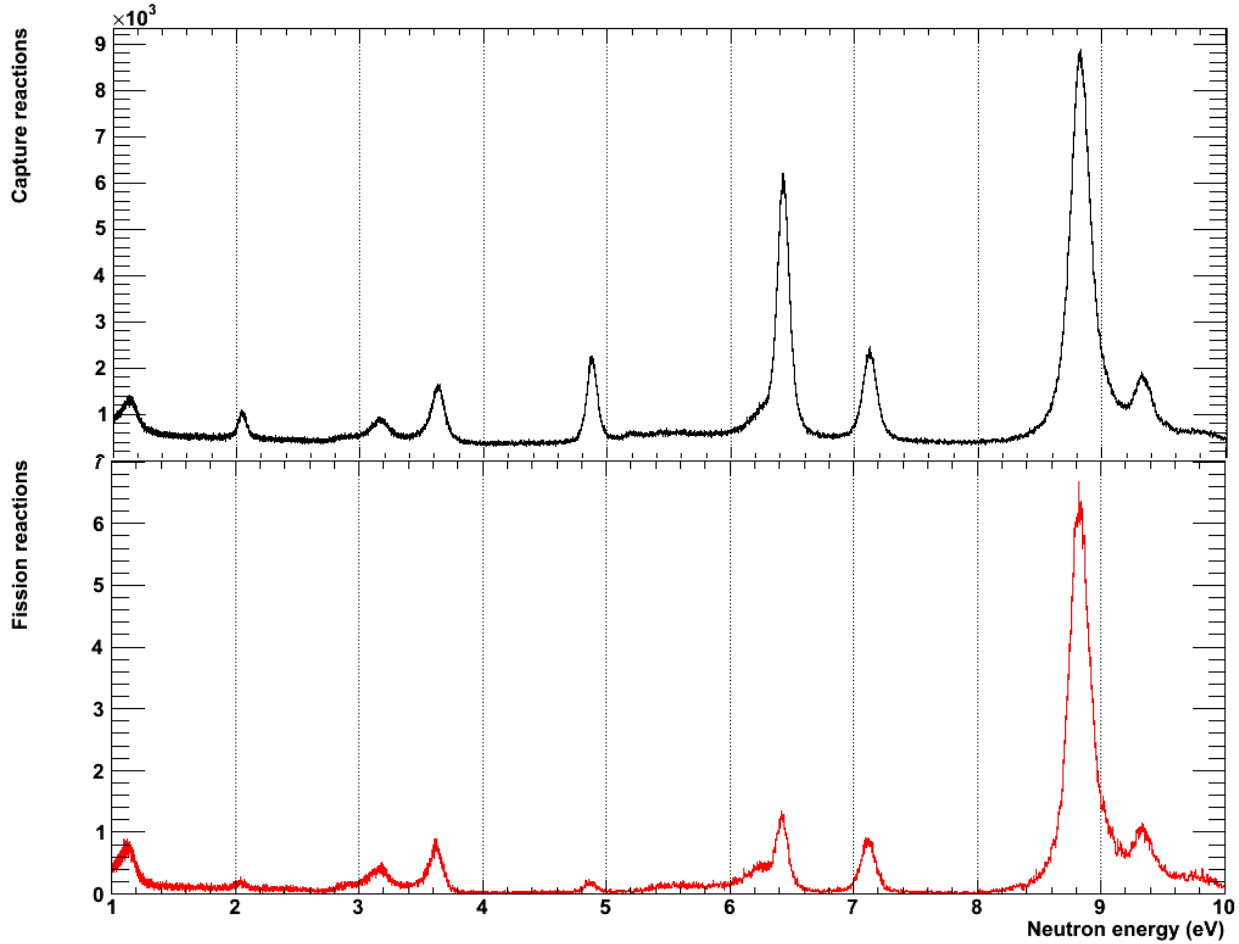
10 samples of 4.15 mg each, instead of 3 samples of 1 mg each → **x10 in mass**
Samples 42 mm in diameter instead of 20 mm → **full beam coverage**
Configuration with neutrons absorber → **x0.2 in neutron scattering background**
More beam time, 9 weeks instead of 1 → **x9 statistics**

σ(n,γ)/(n,f) measurement (2012): deposited energy distributions



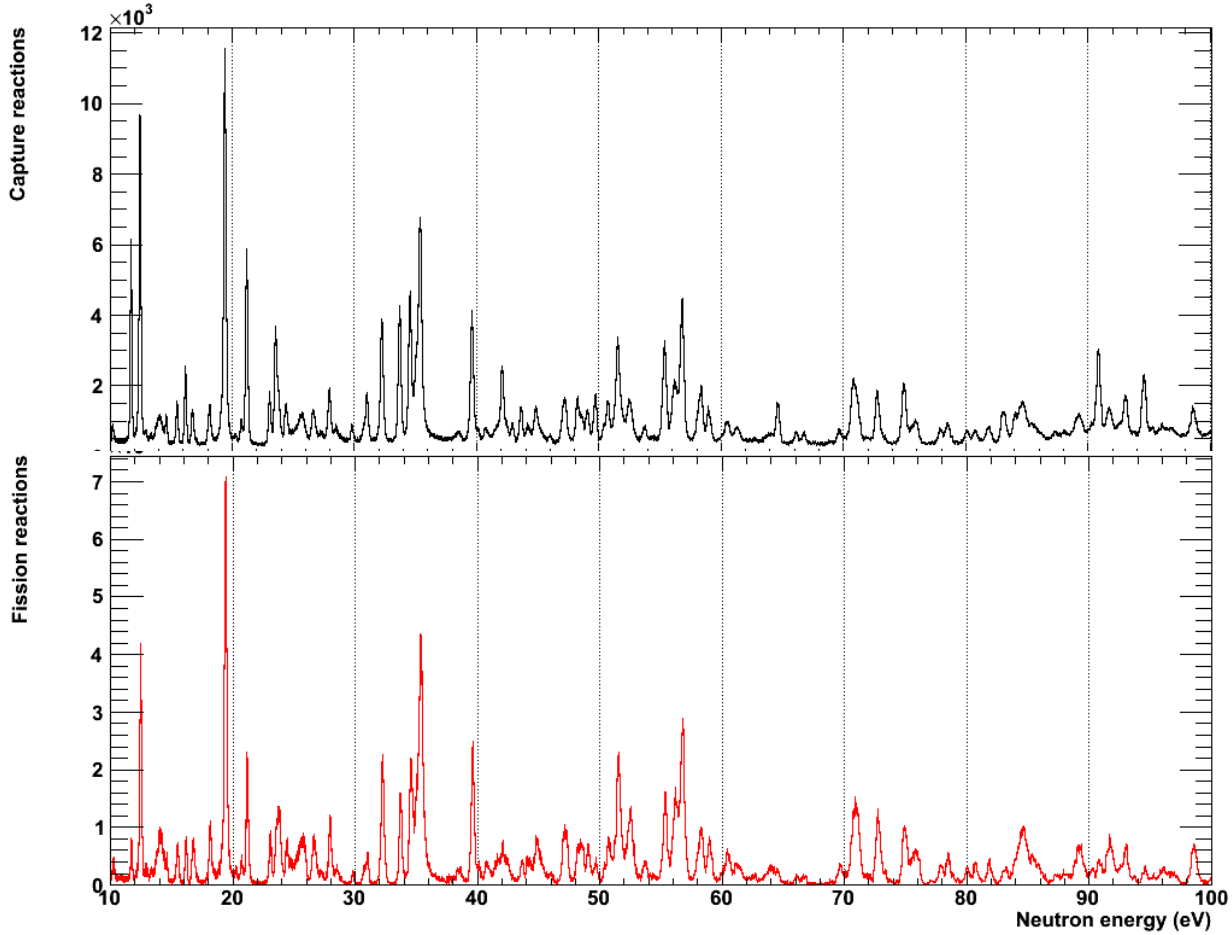
Very Preliminary

σ(n, γ)/(n,f) measurement (2012): neutron energy distributions



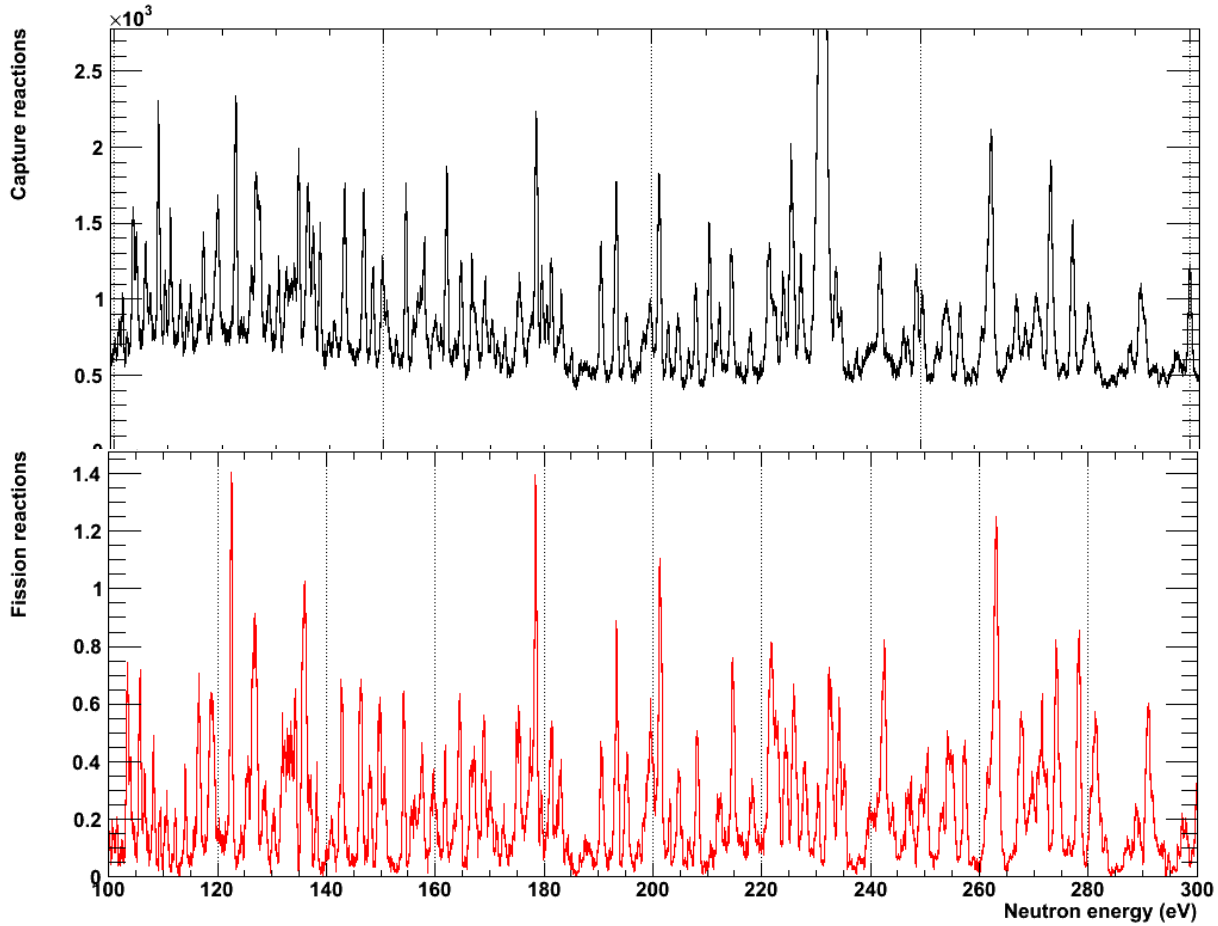
Very Preliminary

σ(n, γ)/(n,f) measurement (2012): neutron energy distributions



Very Preliminary

σ(n, γ)/(n,f) measurement (2012): neutron energy distributions



Very Preliminary

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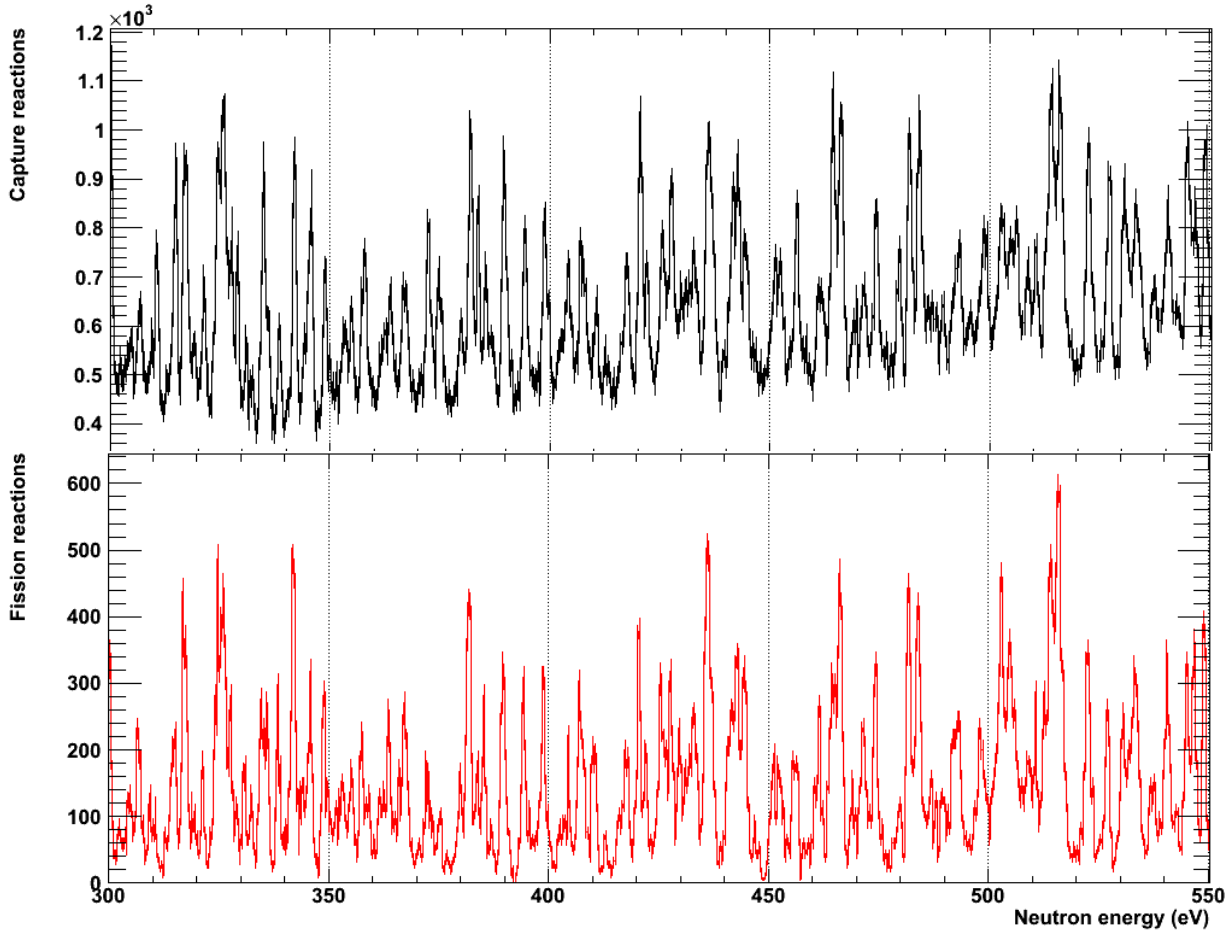

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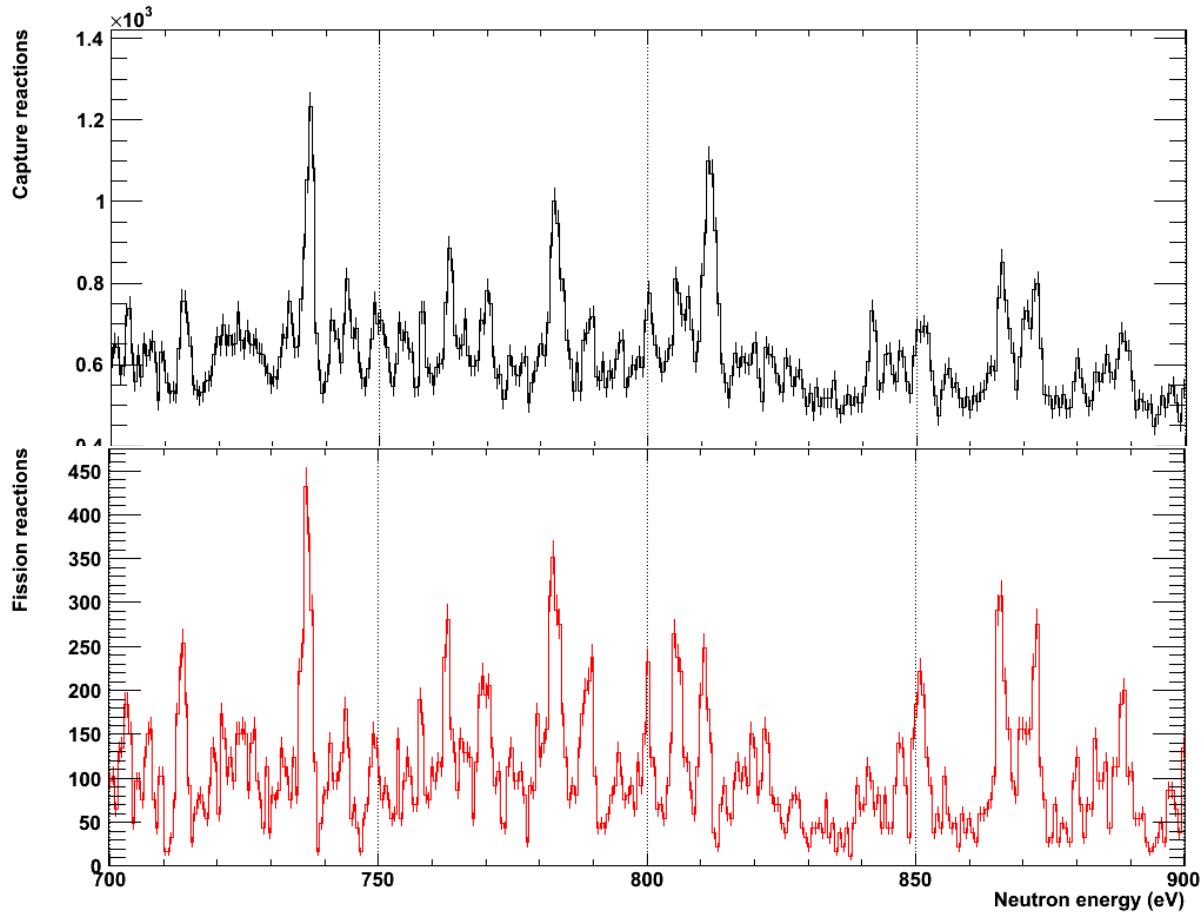
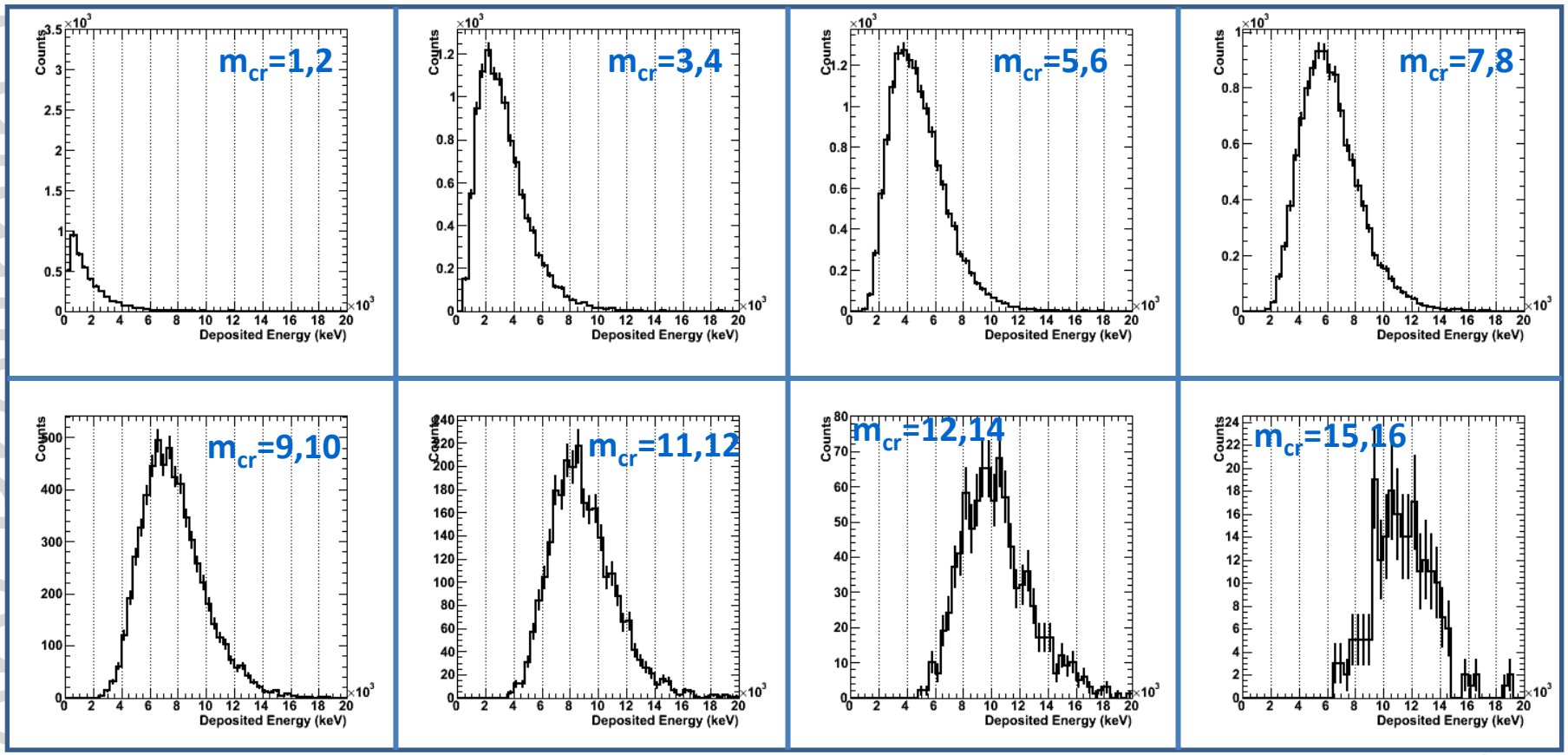


Very Preliminary

Prompt γ-rays from fission in ^{235}U

The 4π BaF_2 Total Absorption Calorimeter (TAC) provides information on the multiplicity and energy of the **prompt g-ray emission following fission** reactions.



Prompt γ-rays from fission in ^{235}U

The 4π BaF_2 Total Absorption Calorimeter (TAC) provides information on the multiplicity and energy of the **prompt g-ray emission following fission** reactions.

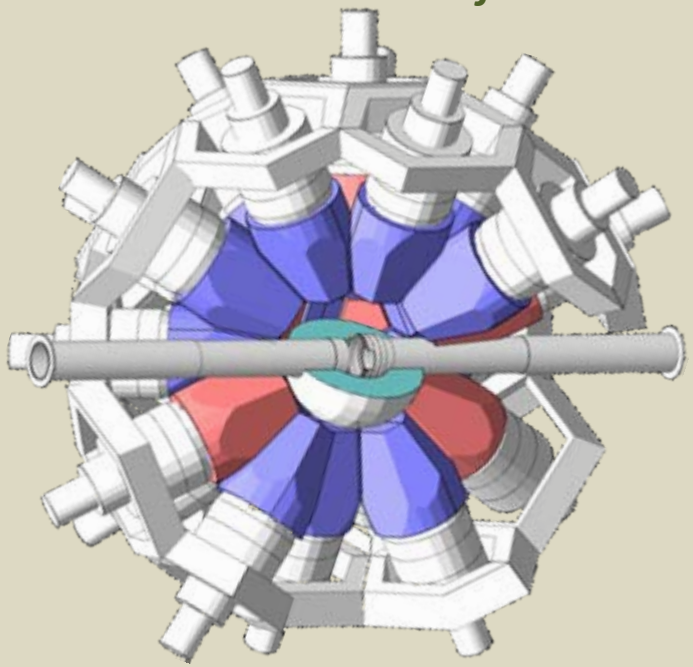
PLAN

Direct measurements of γ-ray emission

+

Benchmark models through:
γ-rays models + simulation
&
comparison to TAC data

Geant4 simulation of the TAC



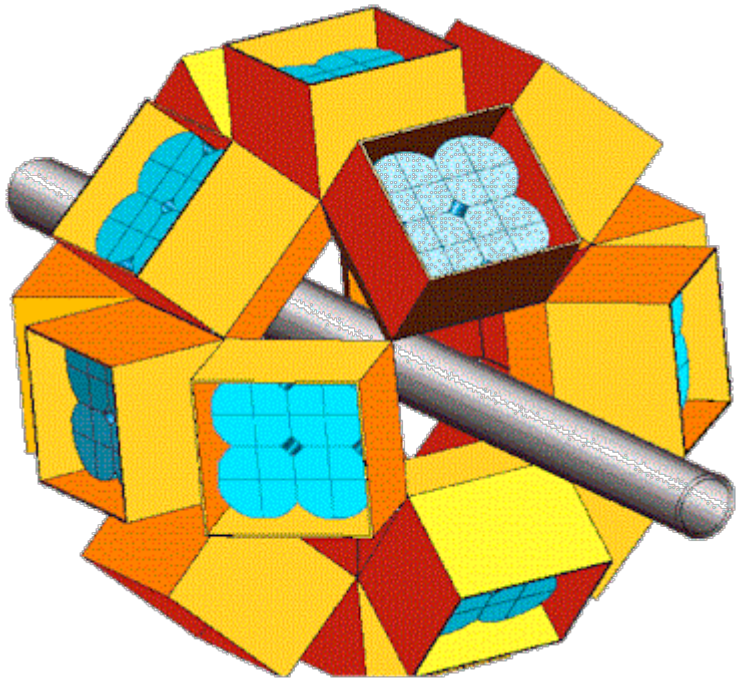
C. Guerrero et al., NIM-A 671 (2012) 108-117

Prompt γ-rays from fission in U & Pu

Independently of n_TOF, a new campaign will take place in **2013** for measuring prompt fission γ-rays from **^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu** through the combination of:

- PBF1 cold neutron beam from ILL
- EXOGAM high resolution HPGe detector array

EXOGAM @ ILL



Conclusions and perspectives

Measuring the capture cross sections and prompt γ-rays of fissile isotopes is of upmost importance for the development of present and future (ADS & Gen-IV) nuclear reactors.

FOLLOWING A SUCCESFUL TEST, A NEW EXPERIMEN IS RUNNING FOR MEASURING:

- **Capture cross section in the Resolved Resonance Region (RRR) [below 2.25 keV]**
- **Resonance parameters in the full RRR**
- **Alpha ratio in the full RRR**
- **Prompt fission γ-rays @thermal and as function of E_n**

Preliminary results from this very fresh data (now being collected) will be presented @ ND-2013 (New York, March 4-8 2013)

D. Cano-Ott et al., Measurement of the neutron capture cross section of the fissile isotope ^{235}U with the CERN n_TOF Total Absorption Calorimeter and a fission tagging based on MicroMegas detectors