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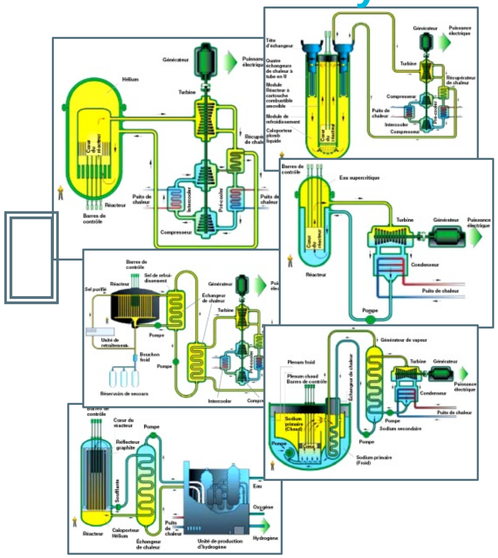
(n,xn γ) reaction cross section measurements for (n,xn) reaction studies

M. Kerveno

A. Bacquias, C. Borcea, Ph. Dessagne, J.C. Drohé, N. Nankov, M. Nyman
A. L. Negret, A. Plompen, C. Rouki, G. Rudolf, M. Stanoiu, J.C. Thiry

Gen IV reactor systems



New concepts of reactors
-> fast reactors
-> accelerator driven systems

New fuel cycle
-> ^{238}U / ^{239}Pu
-> ^{232}Th / ^{233}U

Important needs of **new nuclear data** over a wide range of nuclei, energy and reactions

One of the challenges is **measurement's accuracy**

NEA Nuclear Data High Priority Request List

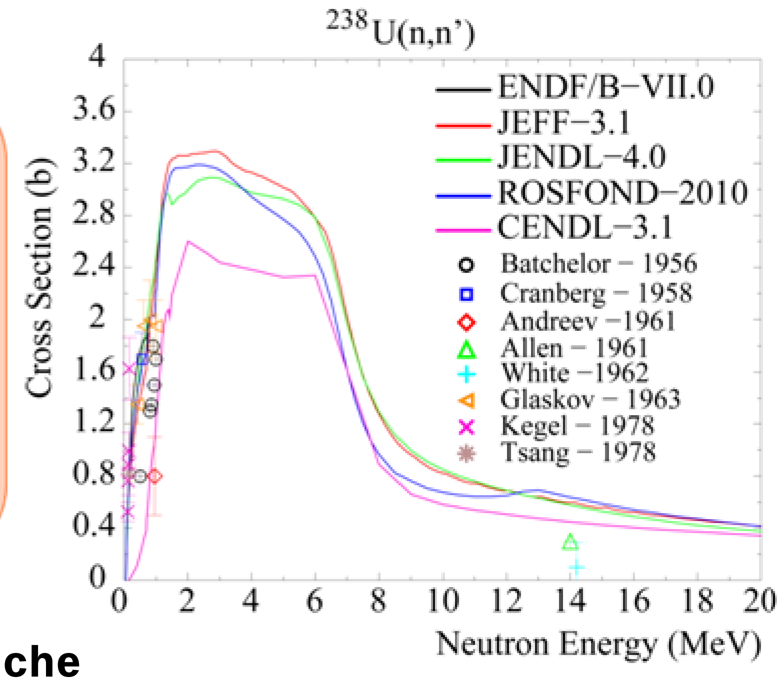
In reactor, **(n,xn) reactions** (x ≥ 1) contribute to
-> Energy loss mechanism
-> Neutron multiplication
-> Production of radioactive isotopes

Bibliography in data bases shows that **improvement** of the **knowledge of (n,xn)** process is **necessary**.

Example of ^{238}U(n,n')

State of the art :

40 measurements in **EXFOR** from 1956 to 2009
8 total cross section measurements
=> **large discrepancies**
between experimental data
and
between evaluated cross sections
=> **^{238}U(n,n') XS uncertainty = 20 %**



Precise requirement from NEA and CEA/Cadarache

Salvatores et al. ; A.Santamarina et al.
Current **uncertainties on ^{238}U(n,n') impact the accuracy** of the **k_{eff}**, of **the power** and of **the β_{eff} calculations** of large core reactors (PWR, FR).

Target accuracy on ^{238}U(n,n') :
-> PWR : **± 10 %**
-> SFR : **± 5 %**

How to study (n,xn) reactions?

-> Direct measurement of secondary neutrons
-> Activation technique
-> **prompt γ-ray spectroscopy**

detection of the **γ-rays** stemming from the **decay of excited states** of nucleus created by the **(n,xn) reaction**.

(n,xn γ) cross sections :
- can also **impact the k_{eff}** calculation,
- can be measured using **"white" neutron beam** with the TOF technique,
- provide **exclusive measurements very restrictive** for testing models.

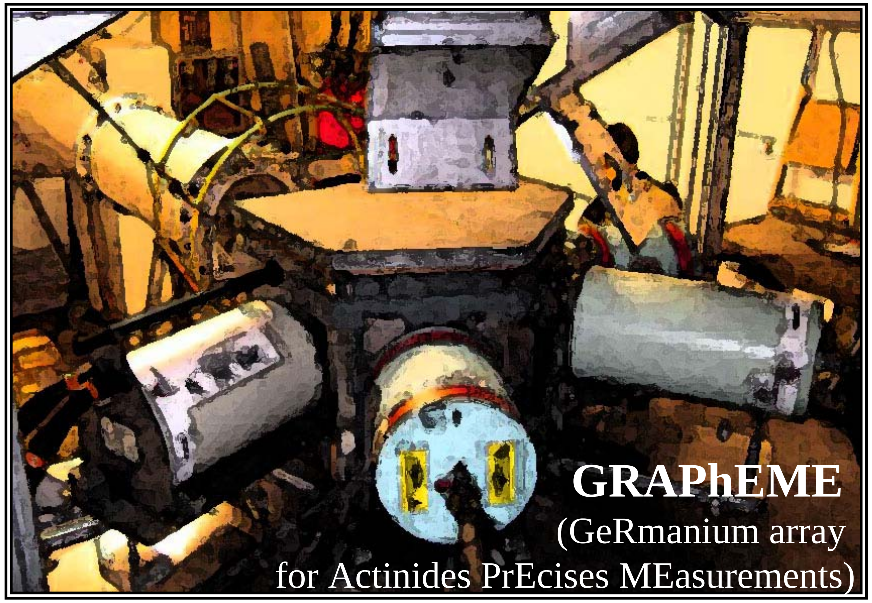
Example of $^{238}U(n,n'\gamma)$
D.Bernard et al.
For small reactor core :
k_{eff} -> **50 % of its sensitivity from first inelastic threshold**
For large reactor core :
radial power -> **sensitivity to the first inelastic levels**

From (n,xn γ) cross section measurements **to total (n,xn)** cross section :
Need of structure parameters and theoretical model...

Introduction : IPHC / IRMM / IFIN-HH experimental project

(n,xn γ) reaction cross sections measurements

IPHC (France) / IRMM (Belgium) / IFIN-HH (Romania) collaboration
=> development of an **experimental set-up GRAPhEME**
dedicated to the **precise measurement**
of the **(n,xn γ) reaction cross sections** on actinides
@ **GELINA facility (IRMM-Belgium)**

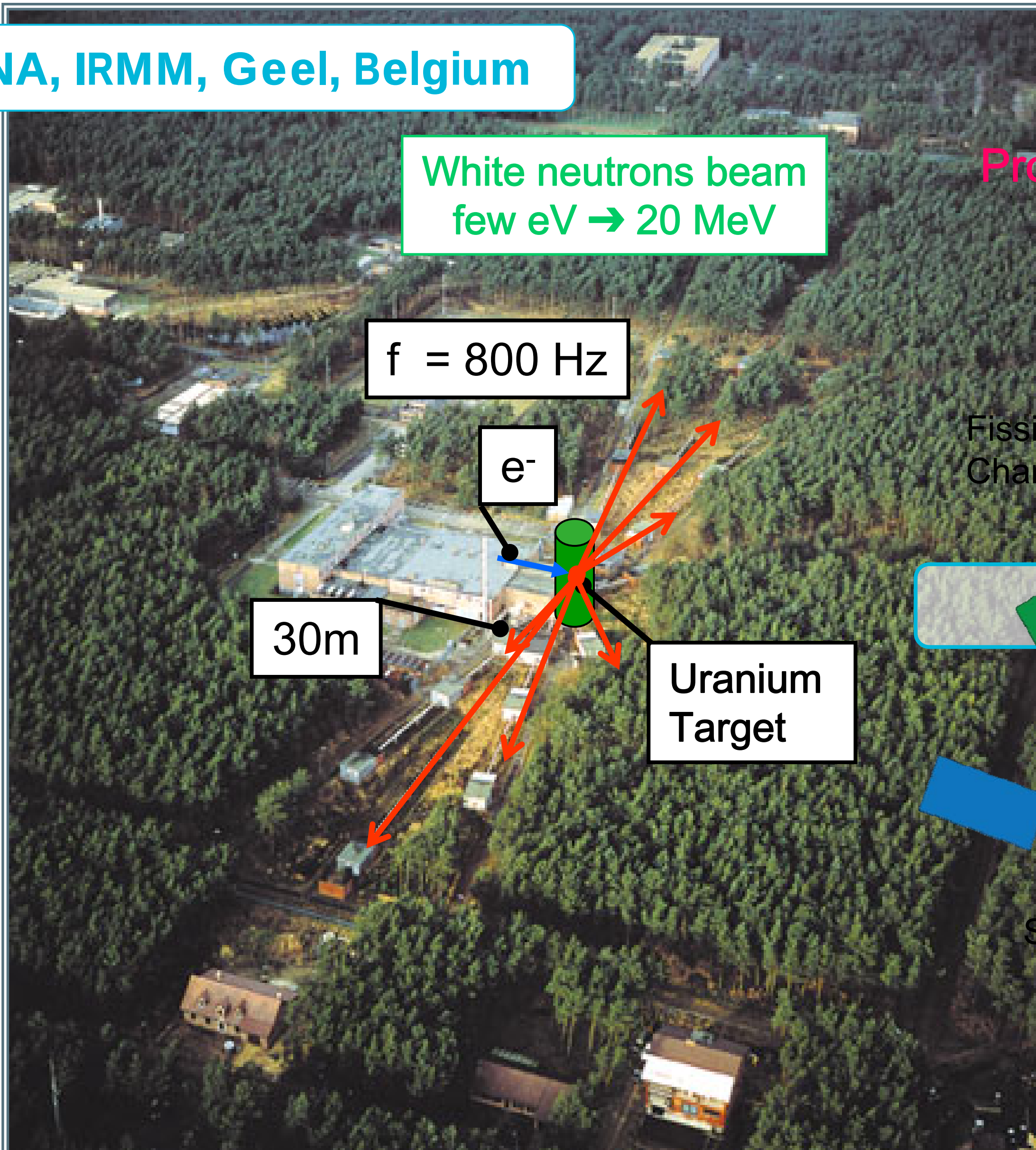


2005 – 2010 :
^{235}U campaign
2009 – 2010 :
^{232}Th campaign
2009 – 2012 :
^{nat,182,183,184,186}W campaign
2011 – 2012 :
^{238}U campaign

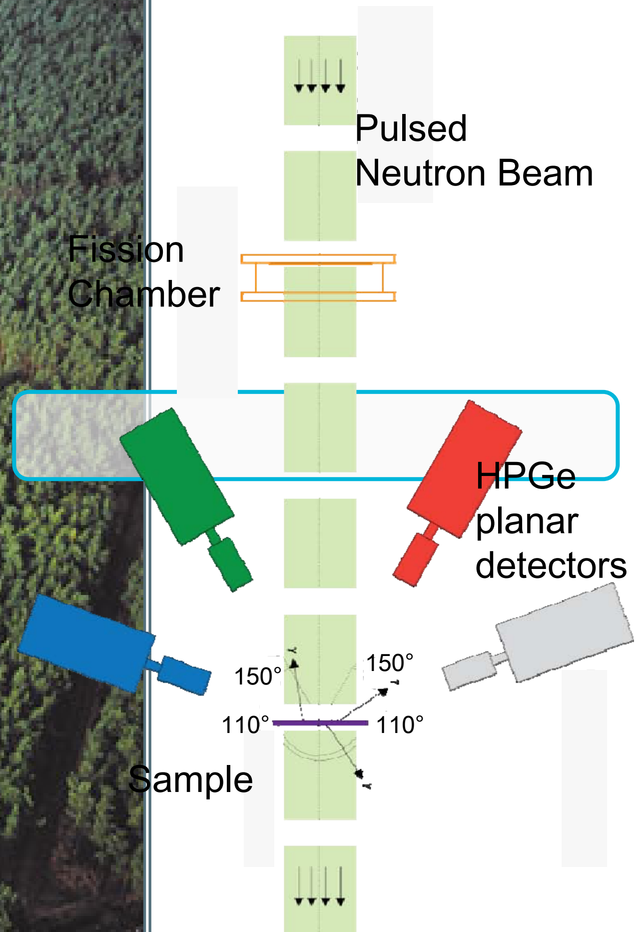
Collaboration with **theoreticians** and **evaluators** to **improve** the **quality** and the **description** of our **measured cross sections**

Experimental set-up @ GELINA : GRAPHEME

GELINA, IRMM, Geel, Belgium

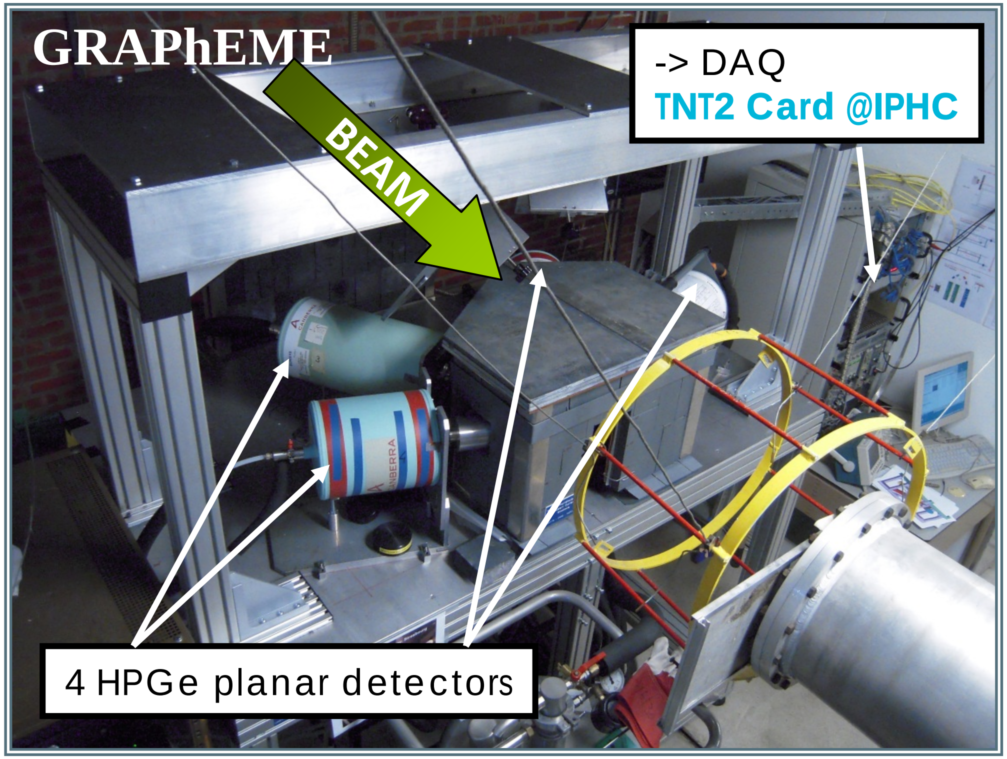


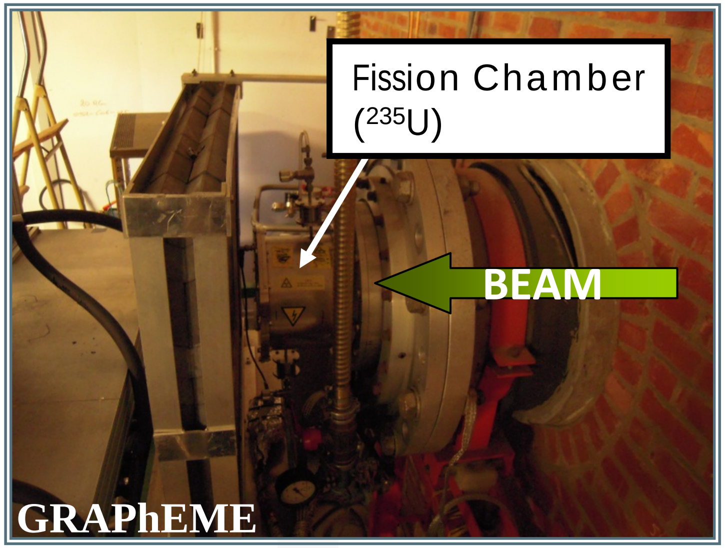
Prompt γ spectroscopy

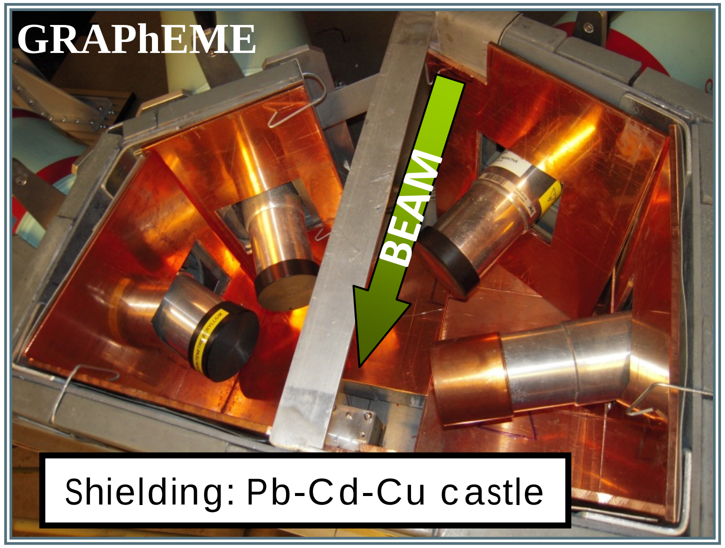


Experimental set-up @ GELINA : GRAPHEME

FP16 – 30 m



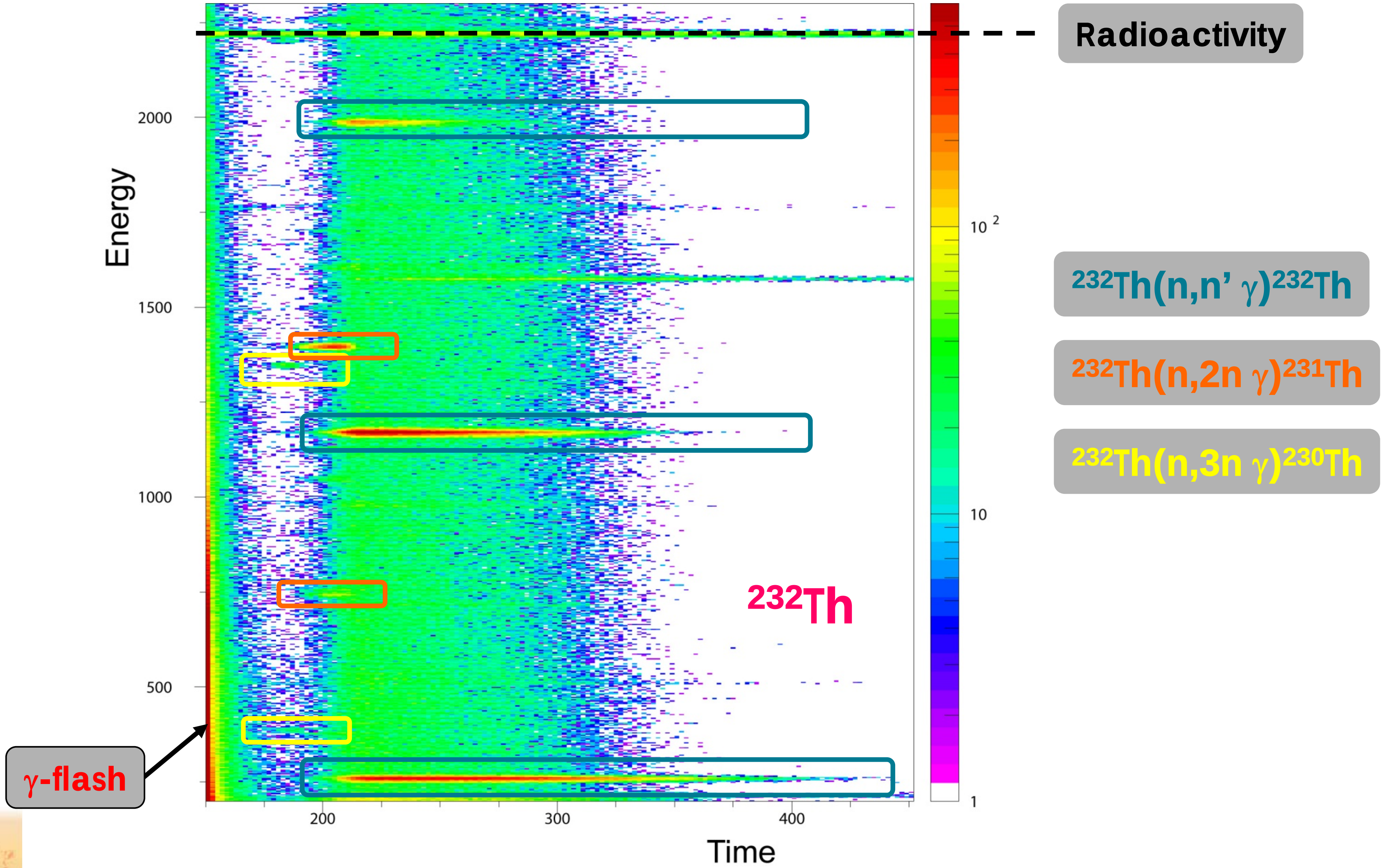




Noise insulation
(electromagnetic field from the accelerator)
and
γ background reduction

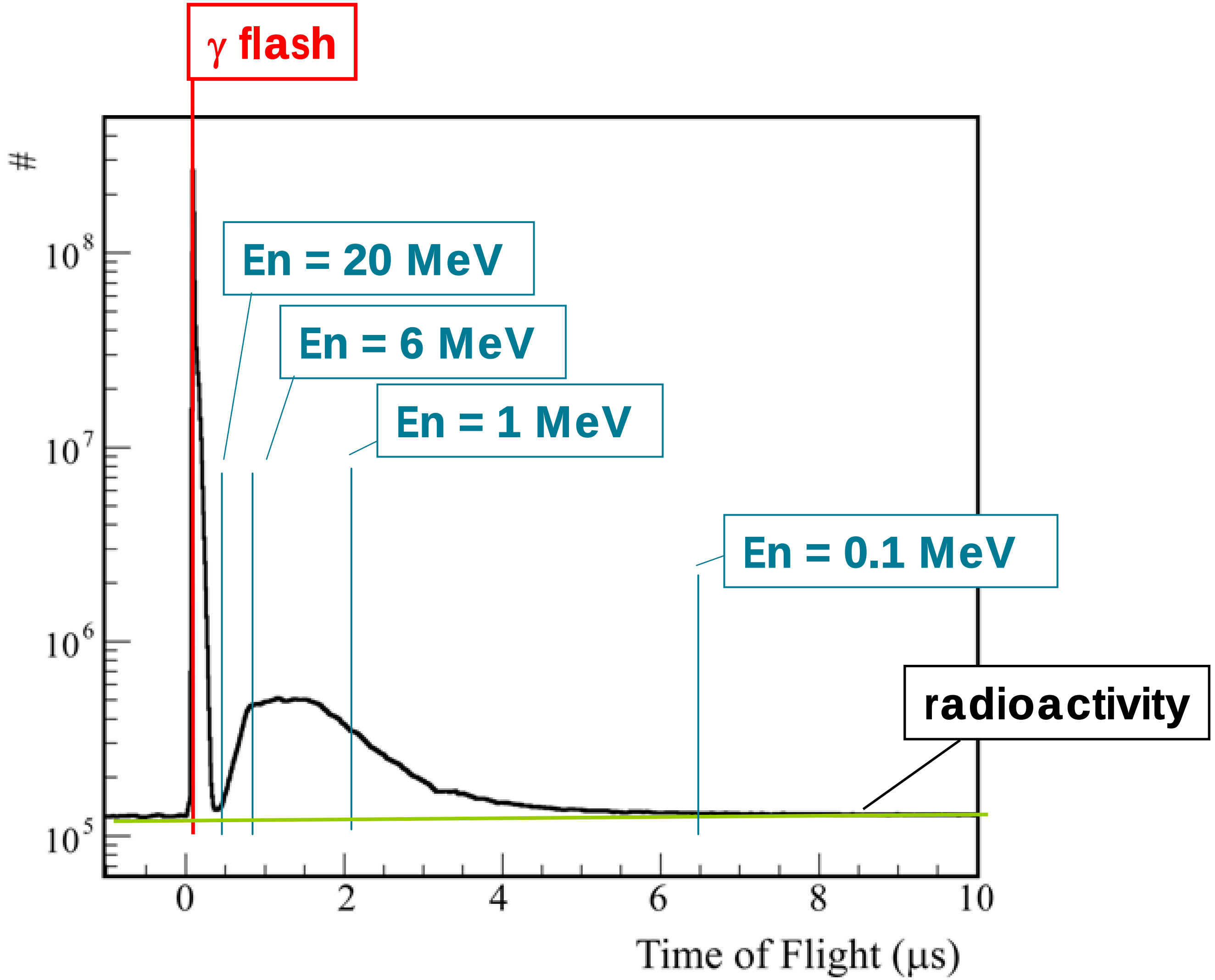
Data Analysis

TOF and γ spectra



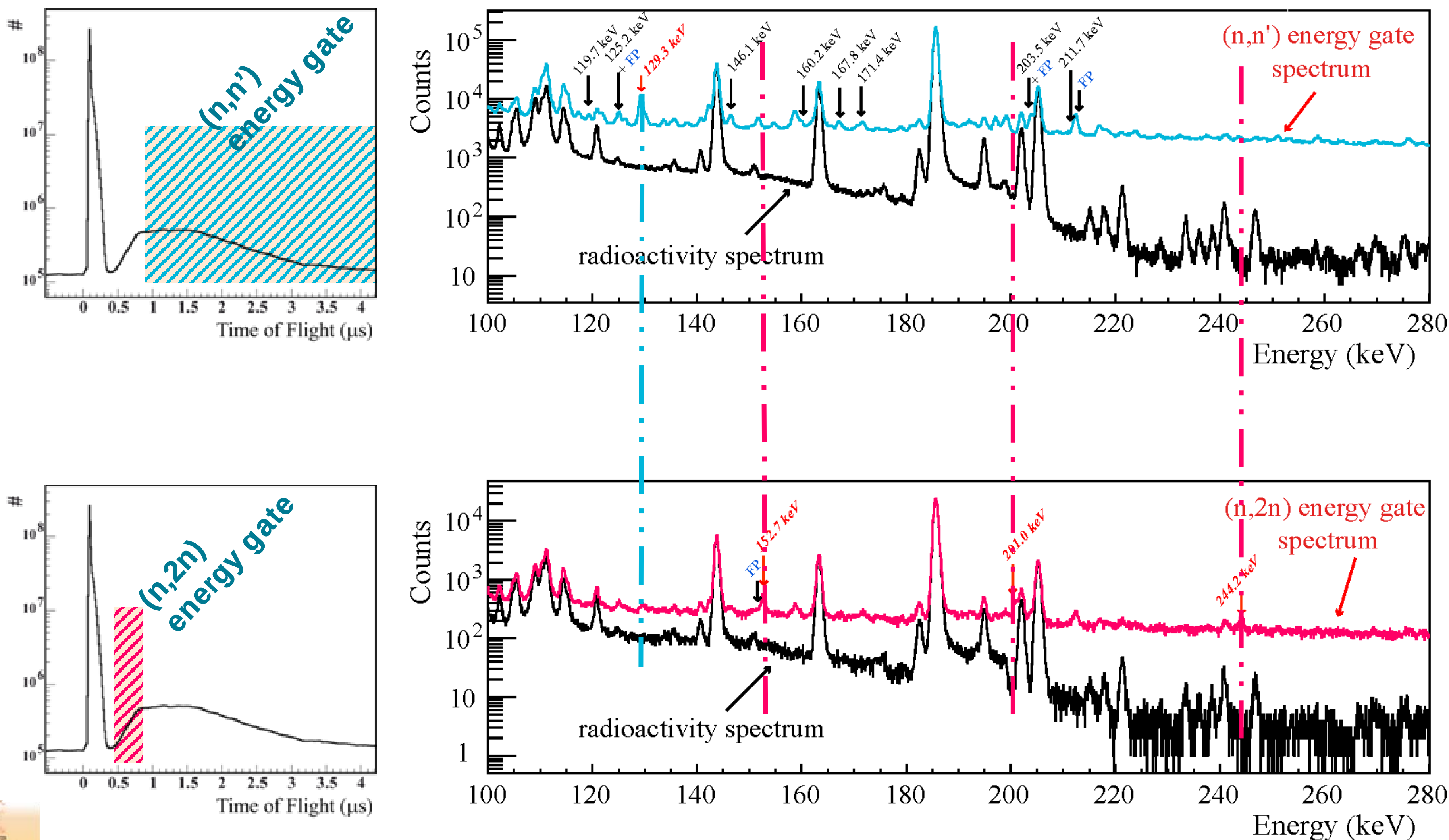
TOF spectrum

^{235}U case

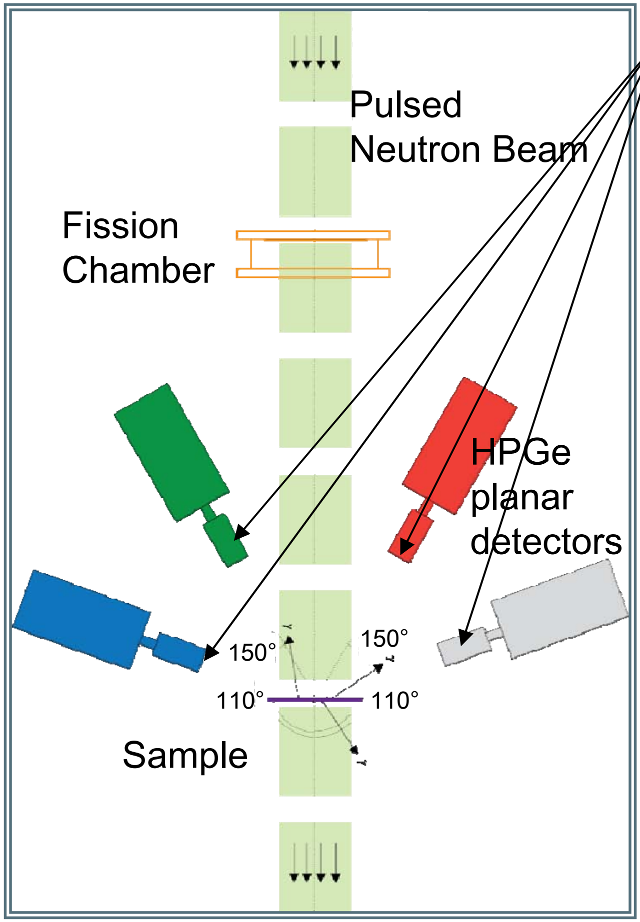


TOF and γ spectra

^{235}U case



Cross section calculation



$$\frac{d\sigma^{(n,xn\gamma)}}{d\Omega}(\theta) = \frac{n_{det}}{N_{at} \cdot \phi_n \cdot \varepsilon \cdot t}$$

n_{det} : number of detected γ
N_{at} : number of atoms in the sample
ϕ_n : neutron flux
ε : HPGe efficiency
t : measurement time

Differential cross section can be expressed by a finite **sum of Legendre polynomials** :

$$\frac{d\sigma^{(n,xn\gamma)}}{d\Omega}(\theta) = \frac{\sigma_{tot}}{4\pi}\sum_{i=0}^{\infty} a_i P_i(\cos\theta)$$

Measurement at the **polynomial nodes**
Gauss quadrature :

$$\sigma_{tot}^{(n,xn\gamma)} = 4\pi\left[w_1^* \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta_1^*) + w_2^* \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta_2^*) \right]$$

w_1 = 0.3479 for Θ_1 = 30.56° or 149.44° and
w_2 = 0.6571 for Θ_2 = 70.12° or 109.88°

What about uncertainties?

$\Delta\sigma/\sigma$: **5% to 7%** E_n= **0.5 – 9 MeV**
But can **reach up to 20 %** E_n> **9 MeV**
(where the neutron flux is low)

Cross section of a given γ ray transition [b]

$$\frac{d\sigma^{(n,xn\gamma)}}{d\Omega}(\theta) = \frac{n_{\det}}{N_{at} \cdot \phi_n \cdot \varepsilon_\gamma \cdot t}$$

Detected hits in a given ray:
- **peak identification** (possible contamination)
- **Good statistics**
- **Low background**

Acquisition time [s]
- **Long time measurements** (stability)

Number of atoms in target
- **Sample composition**
- **Precise** measurements (**size, weight**)

γ detection efficiency
- **Measurements**
- **Simulations** (attenuation, oxidation)

Incident neutron flux [$s^{-1}\cdot cm^{-2}$]
Fission chamber characteristics

Results

$^{235}U(n,xn\ \gamma)$

Beam time: 1466 hours
Sample: enrichment ^{235}U 93.2 %
mass 37.43 g
diameter 12.00 cm
thickness 0.21 mm

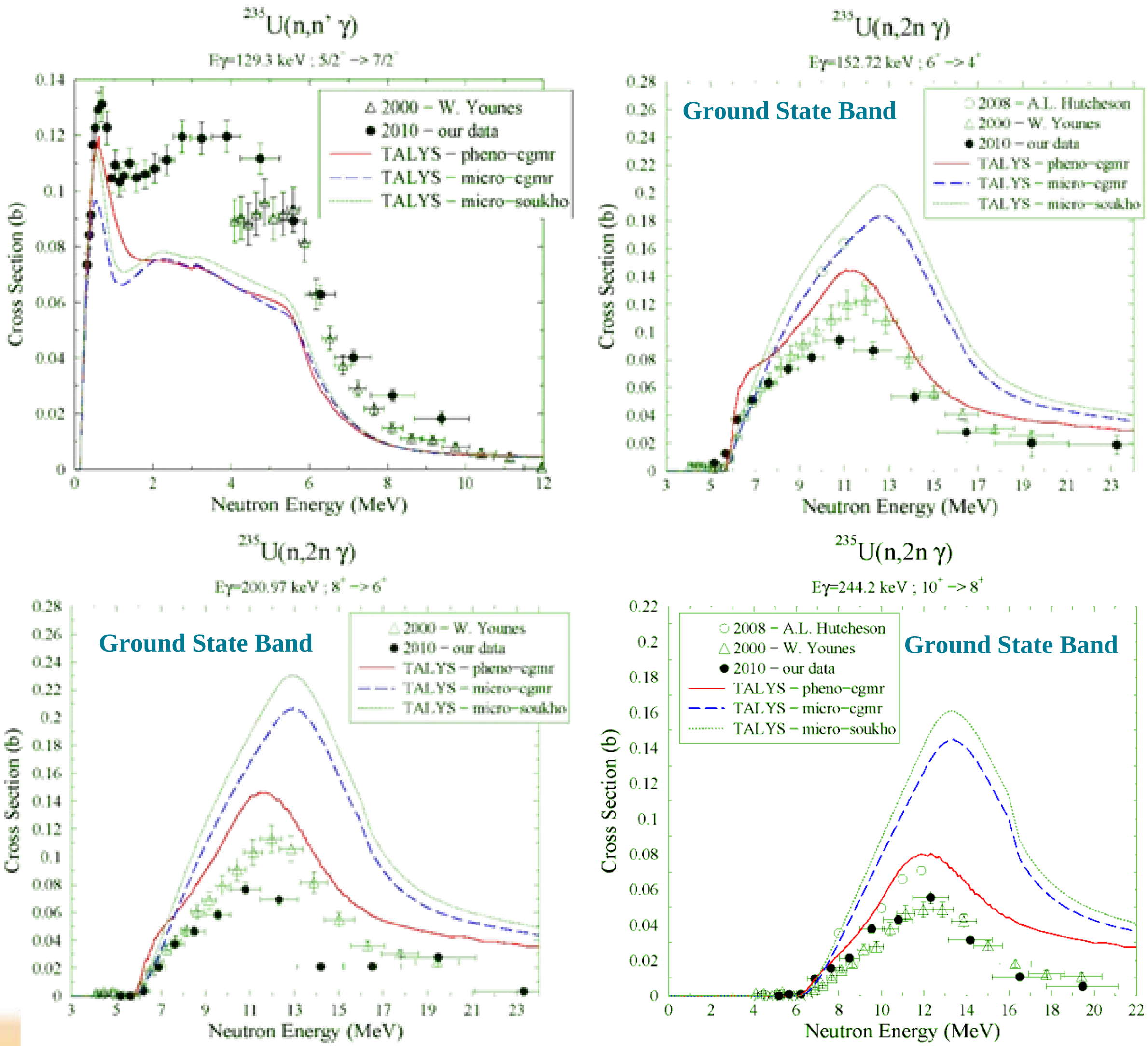
1 γ transition in ^{235}U
3 γ transitions in ^{234}U
-> Compare to **TALYS calculations**
(P.Romain, CEA/DAM, FRANCE)

Bibliography:
Very few measurements in **EXFOR** :
4 σ(n,n') measurement (1961-1969)
1 σ(n,n' γ) meas. (*2000 Younes et al.*)

2 σ(n,2n) measurement (1972-1980)
1 σ(n,2n γ) meas. (*2000 Younes et al.*)

+ σ **(n,xn γ)** in *A.L. Hutcheson Thesis* (2008)

$^{235}U(n,xn\ \gamma)$*



exp data % Our exp data

* **discrepancies with Hutcheson** data.
* **agreement with Younes** data for the **244 keV** γ transition but **discrepancies at high neutron energies** for the 2 other (n,2n γ) transitions.

TALYS % Exp data

* **pheno-cgmr** is the **best parameterization .**
* **(n,n' γ):** shape and amplitude are not well reproduced.
* **(n,2n γ) :** quite good agreement in the shape but factor 1.5 to 1.9 in amplitude.

*J.C Thiry et al. paper submitted soon

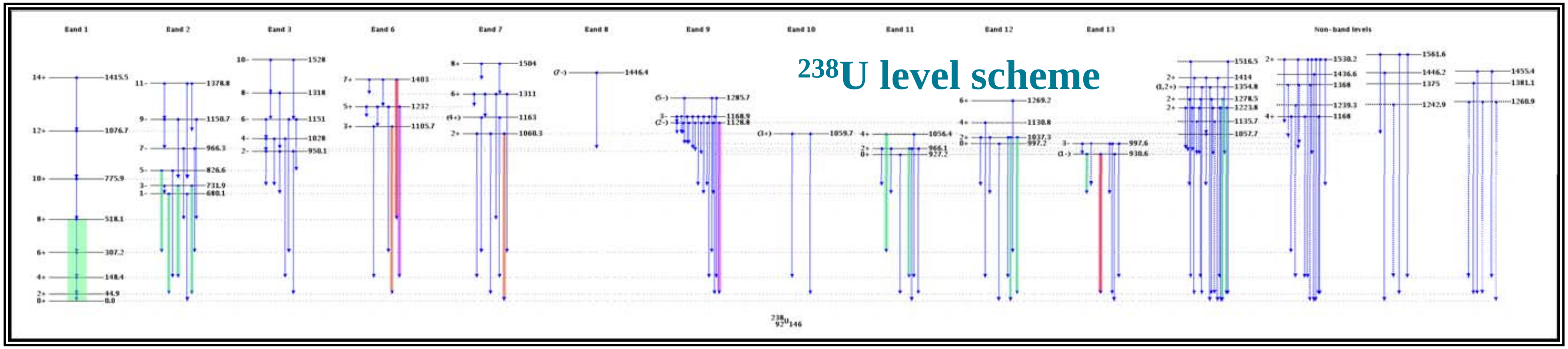
Results

^{238}U(n,xn γ) : preliminary

NEA Nuclear Data
High Priority Request List

Beam time: 1200 hours
Sample: purity ^{nat}U 99.9 %
mass 10.62 g
diameter 7.02 cm
thickness 0.18 mm

About 30 (n,xn γ) transitions "spotted"
20 preliminary γ transitions in **^{238}U**
including **5** going **to the ground state.**
-> Compare to **TALYS calculations**
(P.Romain, CEA/DAM, FRANCE)

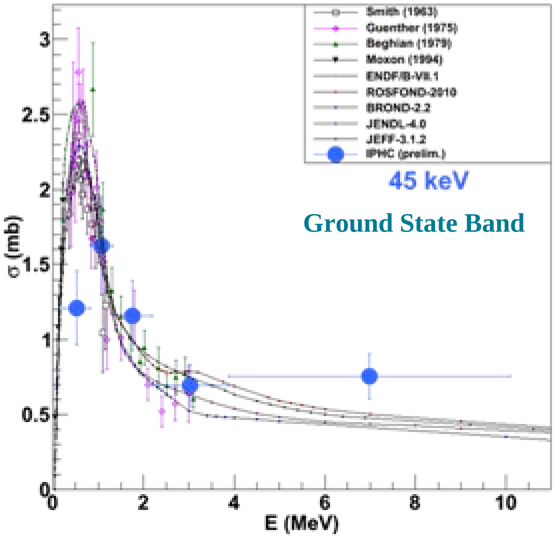


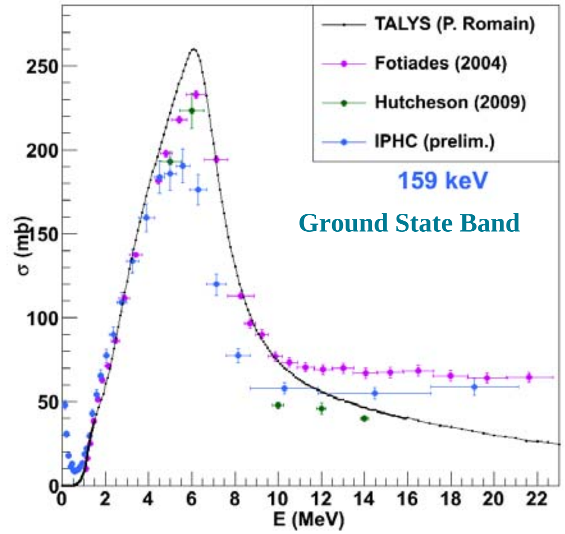
Bibliography:
lot of total cross section measurements in **EXFOR** but :
4 σ(n,n' γ) meas. (*1976 Voss et al., 1979 olsen et al., 2004 Fotiades et al., 2009 Hutcheson et al.*)

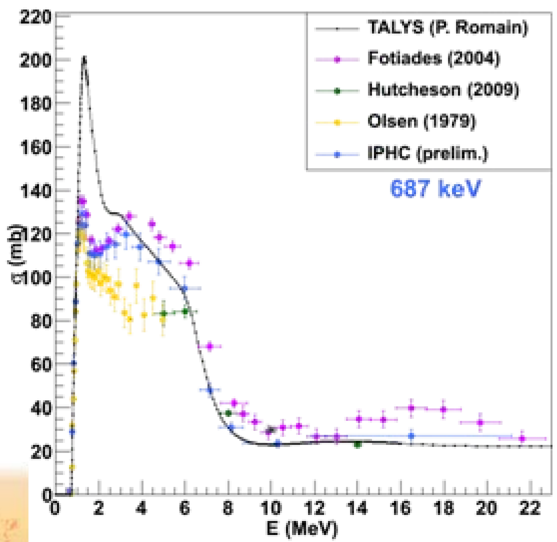
2 σ(n,2n γ) meas. (*2009 Hutcheson et al., 2004 Fotiades et al.,*)

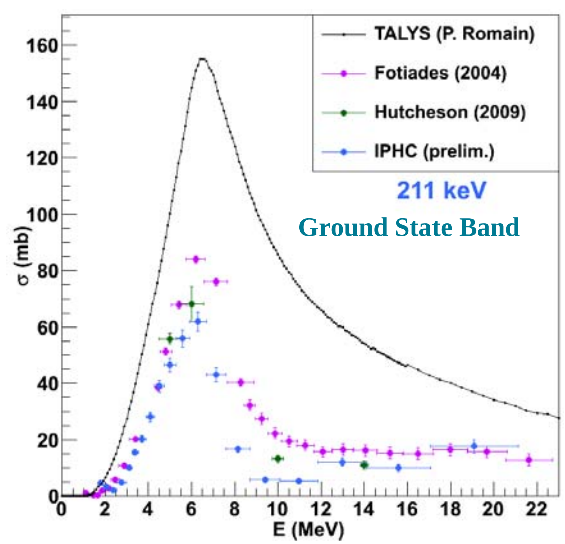
Results

$^{238}U(n,n'\gamma)$: preliminary









N.B. we are able to **measure** the **de-excitation** of **the first level in ^{238}U**

exp data % Our exp data

* Fotiades data slightly **higher** than our data but **good agreement** in **shape**.

TALYS % Exp data

* Shape is **well reproduced** **except** in some case **when a direct component** appears : the relative proportion of the two components are not well calculated.
* In **amplitude**, **discrepancies** depend on the γ-transition.

Results

^{232}Th(n,xn γ) : very preliminary

Beam time: 375 hours
Sample: enrichment ^{232}Th 99.5 %
mass 11.99 g
surface 36.46 cm²
thickness 0.30 mm

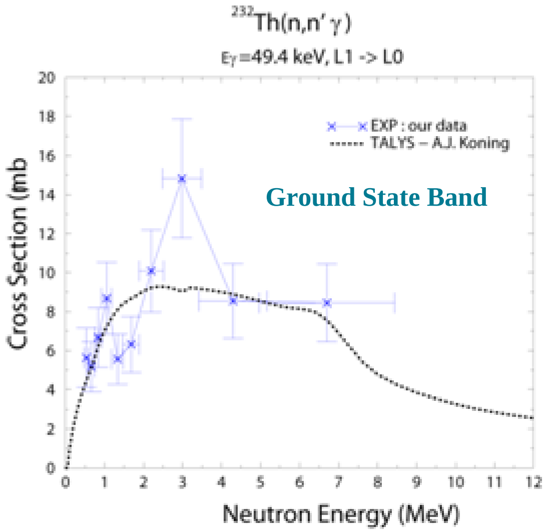
12 γ transitions in **^{232}Th**
1 γ transition in **^{231}Th**
-> Compare to **TALYS calculations**
(A.Koning, NRG, The Netherland)

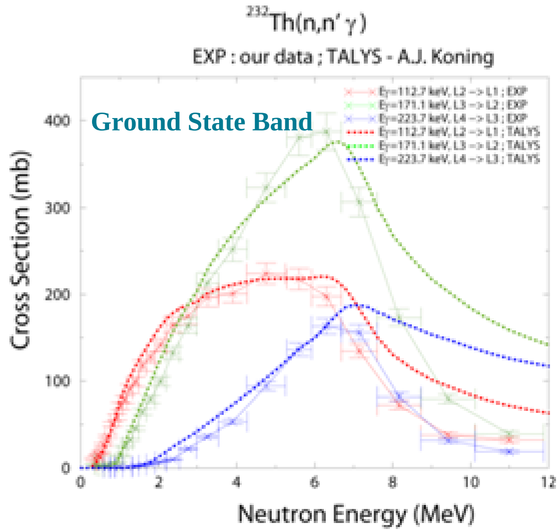
Bibliography:
Several measurements in **EXFOR** :
3 σ(n,n') measurement (1962-1983)
12 σ(n,n') level production measurement (1962-2001)
1 σ(n,n' γ) meas. (*1985 Dave et al.*)

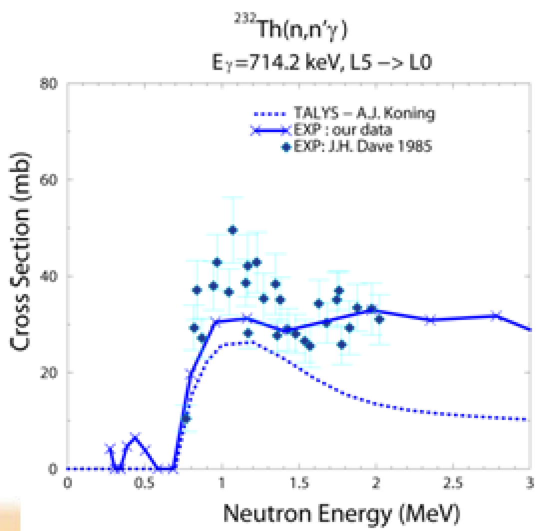
21 σ(n,2n) measurement (1956-2011)
0 σ(n,2n γ) meas.

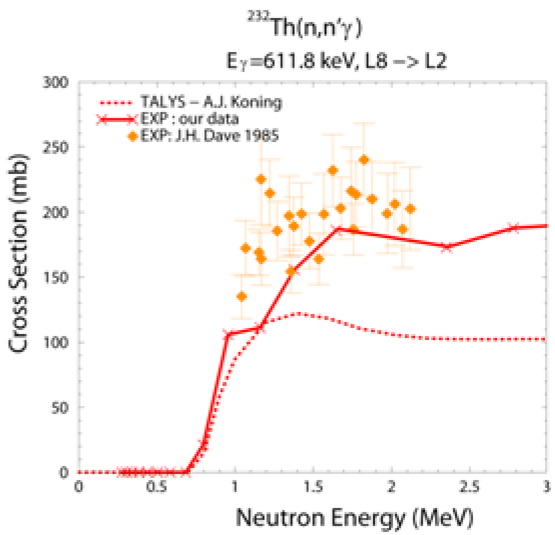
Results

^{232}Th(n, n' γ) : very preliminary









N.B. we are able to **measure** the **de-excitation** of **the first level in ^{232}Th**

exp data % Our exp data

* **agreement** is **very good** up to E_n=2 MeV (high limit of the J.H. Dave exp data).

TALYS % Exp data

* **amplitude** is **well reproduced** for states in ground_state band but **overestimation above E_n = 7 MeV.**
* for other γ-transitions the **agreement** is **less good**.

$^{186,184,183,182}W(n,xn\ \gamma)$: very preliminary

Beam time: 300 hours $^{186,184,183}W$
500 hours ^{182}W
Sample: enrichment $^{186,184,182}W$ ~ 94.5 %
^{183}W ~ 83.75 %
mass ~ 45 to 49 g
diameter ~ 6.6 to 7.1 cm
thickness ~ 0.13 to 1.30 mm

Bibliography:
Few measurements in **EXFOR** :

^{182}W : **2 σ(n,n')** measurement (1967 – 1999)
7 σ(n,2n) measurement(1959 – 1997)

^{183}W : **2 σ(n,n')** measurement (1982 – 1996)
1 σ(n,2n) measurement (1980)

^{184}W : **3 σ(n,n')** measurement (1967 – 2003)
4 σ(n,2n) measurement (1966 – 1982)

^{186}W : **3 σ(n,n')** measurement (1967 – 1996)
14 σ(n,2n) measurement (1959 – 1999)

^{182}W sample
27 γ transitions in **^{182}W**
4 γ transitions in **^{181}W**

^{183}W sample
17 γ transitions in **^{183}W**
5 γ transitions in **^{182}W**

^{184}W sample
15 γ transitions in **^{184}W**
4 γ transitions in **^{183}W**

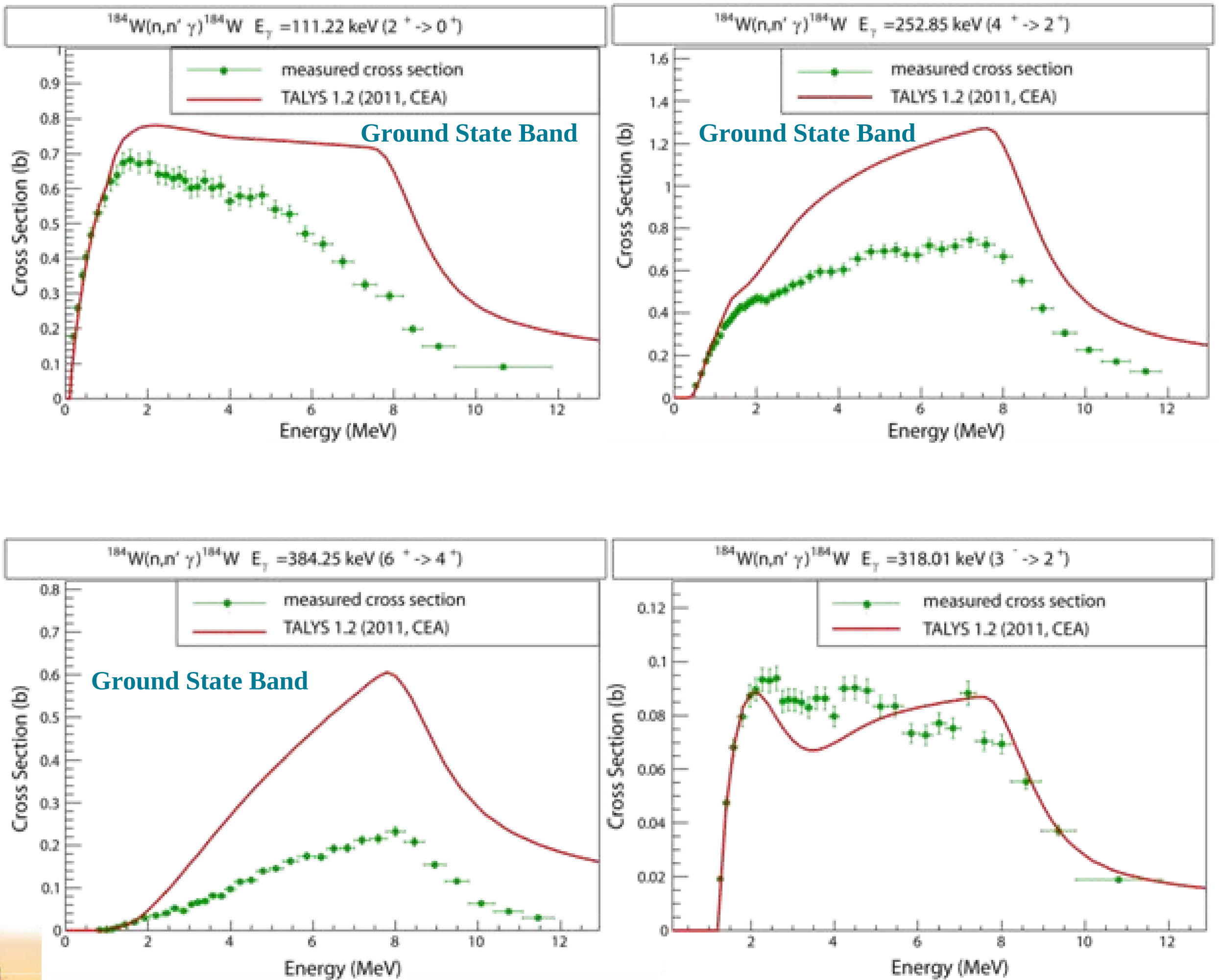
^{186}W sample
15 γ transitions in **^{186}W**
3 γ transitions in **^{185}W**

-> Compare to **TALYS calculations**
(P.Romain, CEA/DAM, FRANCE)

Results

$^{186,184,183,182}W(n,xn\ \gamma)$: very preliminary

$^{184}W(n,n'\ \gamma)$



Preliminary conclusions

TALYS % Exp data

* in most cases the **shape** is **well reproduced** but the **discrepancies** for the **amplitude** are **different** for **each γ transition.**

* **branching ratio** data bases play an **important role**.

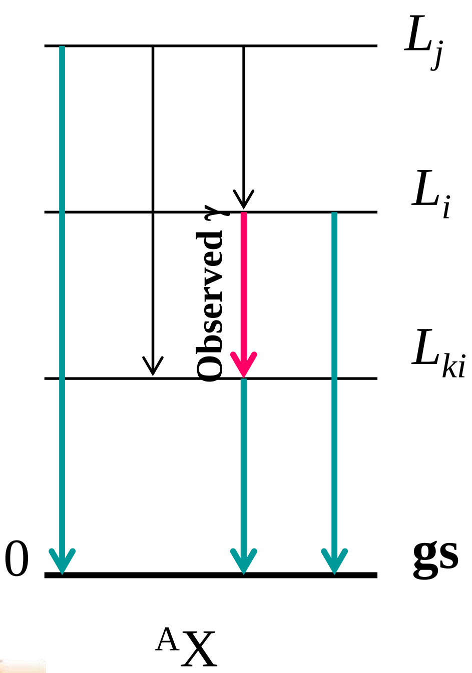
From (n,xn γ) to (n,xn) cross sections?

Measured
(n,xn γ)
cross section

Theoretical model

Spectroscopic Parameters

total
(n,xn)
cross section



Total inelastic scattering cross section is the **sum** of the cross section carried by **all transitions** that **directly decay to the ground-state**

In general case:

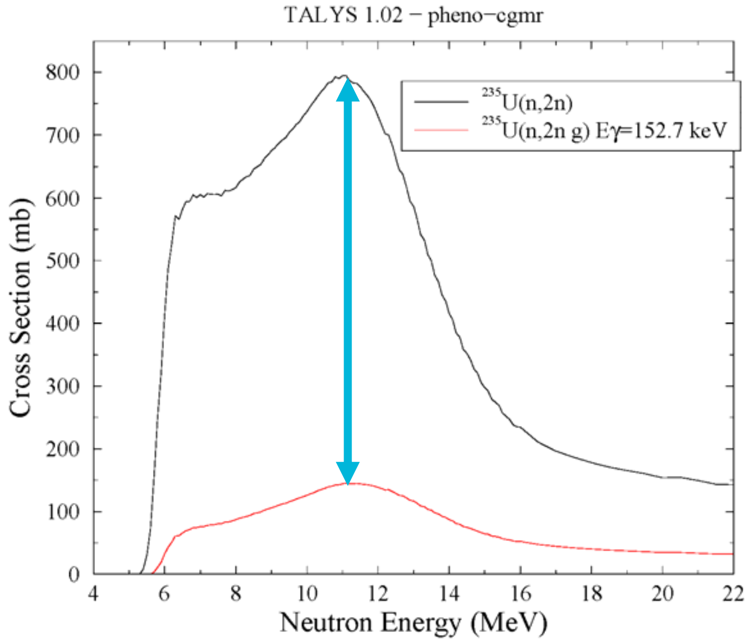
$$\sigma_{n'}(E) = \sum_{i=1}^{E_x(Li)\leq E} \sigma_{n',\gamma}(E, L_i \rightarrow L_{k_i}) \frac{p(L_i \rightarrow g.s.)}{p_\gamma(L_i \rightarrow L_{k_i})}$$

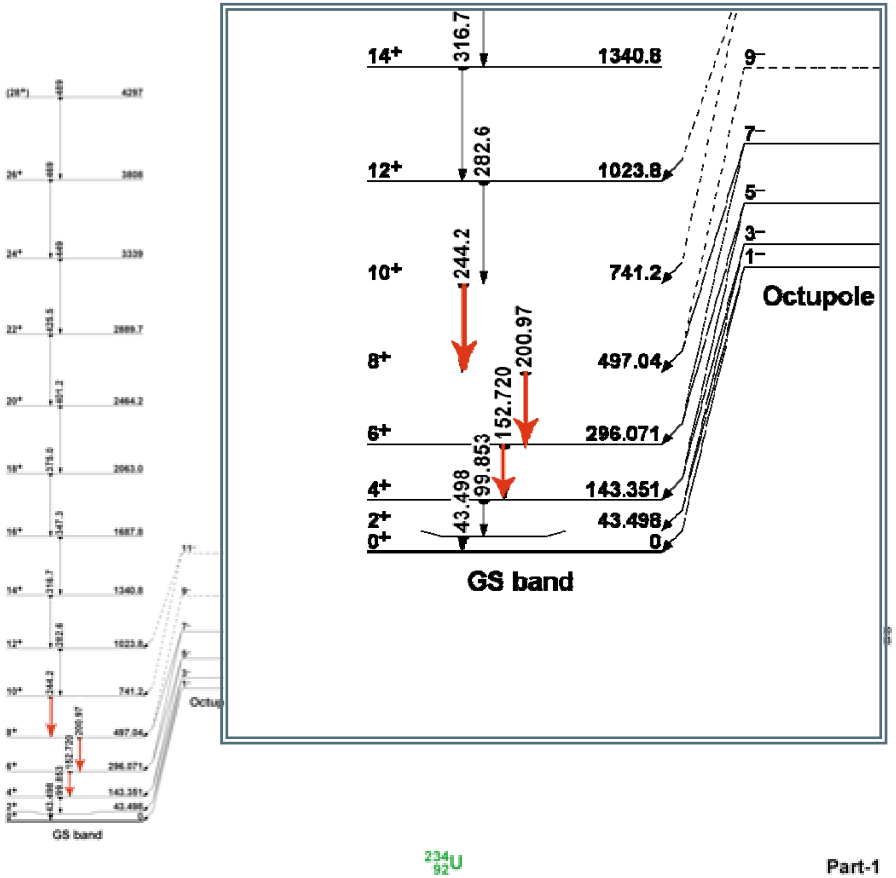
-Requires a good knowledge of spectroscopic parameters

-Practically, the **deduced inelastic cross section is a lower limit** for the **total inelastic cross section-> model prediction**

From (n,xn γ) to (n,xn) cross sections?

^{235}U(n,2n) case



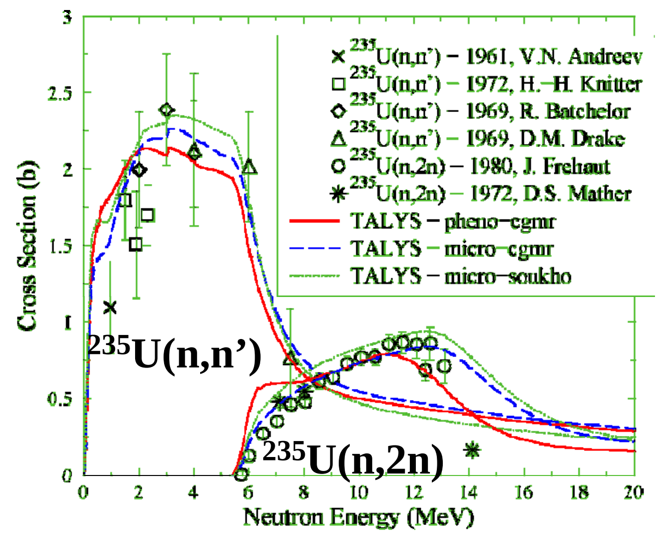


We have measured **only ~20%** of the total cross section …
Strong model dependence

What did we learn?

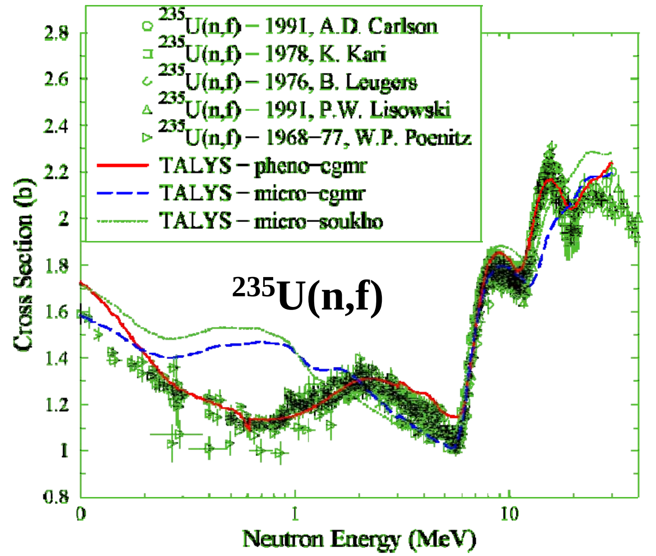
Experimental point of view :

- **control** and **minimize** all source of **error**
- **matrix covariance** calculation
- **measurement** of a **maximum of γ-transitions**
- **efforts** have to be done **to measure** the **γ-transitions to the ground state**



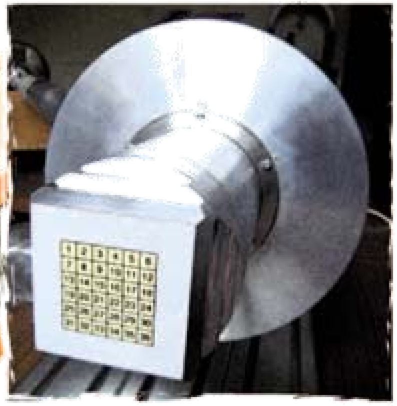
Theoreticians and evaluators point of view :

- for fissionable nuclei, **σ(n,f)** must be **well described**
- nucleus **structure, branching ratios** and **internal conversion coefficients** play an **important role**
- **another approach** than the **exciton** model (TALYS-1.2) has to be **tested** to model the **pre–equilibrium** reactions



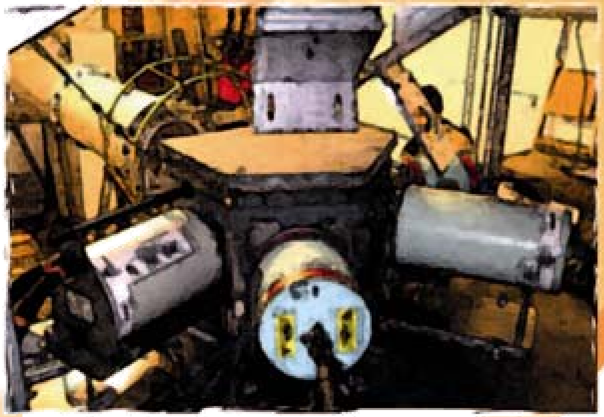
Conclusion

Branching ratio
Conversion electron
measurements



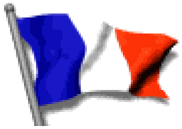
precise σ(n,xn γ)
^{235}U, ^{238}U, ^{232}Th,
^{186,184,183,182}W
Covariance matrix

precise σ(n,xn γ)
highly radioactive targets
^{233}U (^{232}Th cycle)
Segmented HPGe



Collaboration with
evaluators:
quality of experimental data
theoreticians:
quality of model predictions

Collaboration, Financial support, Acknowledgement

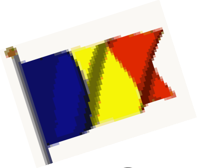


A. Bacquias, Ph. Dessagne, M. Kerveno, G.Rudolf

IPHC Strasbourg, France

A. Plompen, J.C. Drohé, M. Nyman

irmm Geel, Belgium



C. Borcea, A.L. Negret

IFIN-HH Bucharest, Romania

PACEN cnrs

NUDAME Neutron Data Measurements

EUFRAT

Accurate Nuclear Data for nuclear Energy Sustainability

The authors thank the team of the GELINA facility for the preparation of the neutron beam and for their strong support day after day....