Inelastic scattering on ^{232}Th: measurements and beyond

E. Party
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Plan

- I. Context
- II. Method
- III. Experiment
- IV. Analysis
- V. Results
- VI. Additional works

I. Context – Inelastic scattering on ^{232}Th and reactor applications

Why inelastic scattering ?

Nuclear reactions occuring in reactors :

- (n,g);(n,f);(n,el);(n,inl);(n,x)
- Inelastic scattering : main slowing down factor in fast neutron reactor

- Reactor neutronic simulation needs precise nuclear data
- Experimental data on some reactions lack precision, and may be inexistant
- **Realistic covariances** are needed for **evaluation** process and propagation of uncertainty

Why Thorium ?

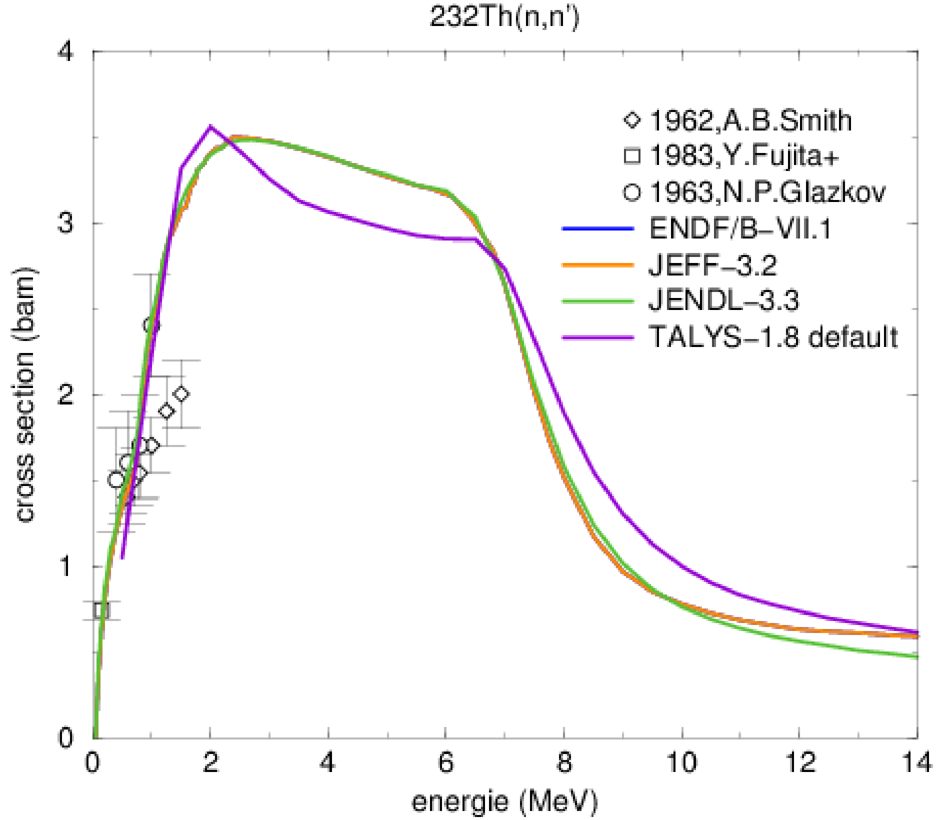
Material exposed to neutrons:

- Nuclear fuel
- Moderator
- Structural materials

- ^{232}Th : fertile isotope, possible nuclear fuel in combination with ^{233}U
- Even for non-Th fueled reactor, measurements may be of use for a systematic theory of inelastic scattering

I. Context – State of measurements on ^{232}Th

- Scarce measurement
- Almost no points for neutron energy > 3 MeV for partial (level or gamma) cross sections
- There is a need for more measurement at higher energy



^{232}Th(n,n') measurements and evaluations

II. Method – Principle of measurement

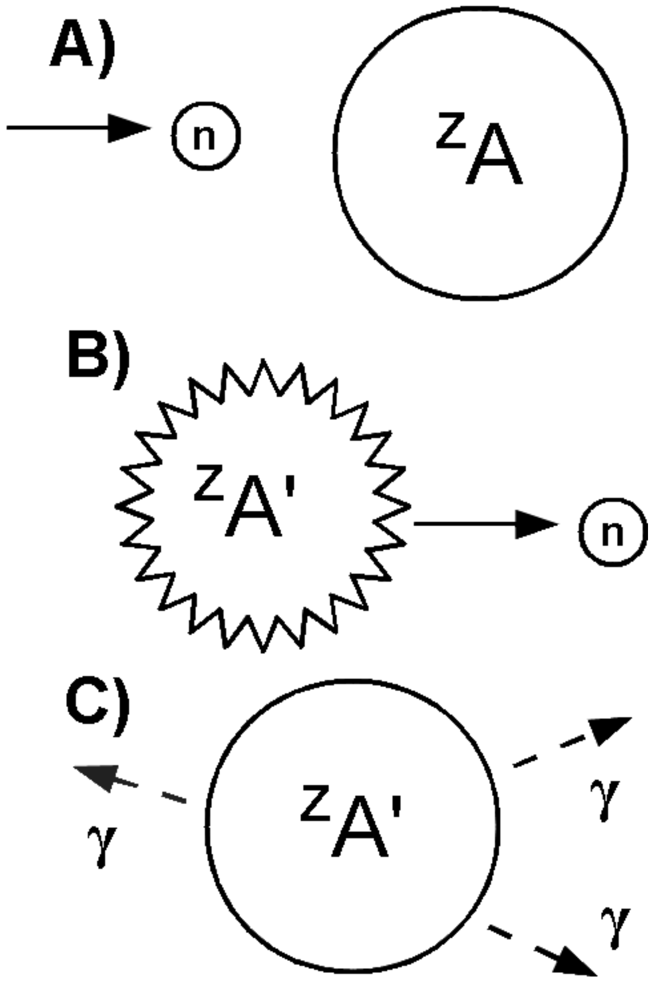
(n,n') can be measured using :

<u>1. Neutron scattered – Direct detection</u>

- Most useful for reactor application, as energy loss and angular distribution are directly observable
- Difficulties to measure precisely and separate contributions from (n,el), (n,f)…
- Energy resolution is poor

<u>2. Deexcitation of nucleus – Gamma spectroscopy</u>

- Relatively easy to observe
- High energy resolution using HPGe counters allows to distinguish close levels
- As Z is high, unseen deexcitation by conversion electron may be a problem
- May not allow direct measure of excitation energy (~neutron energy loss), then it is dependent on nuclear structure to obtain level- and total XS

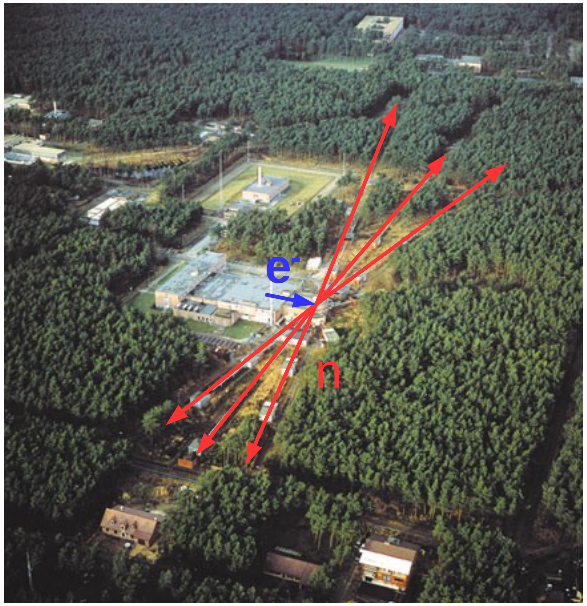


Steps of inelastic scattering

III. Experiment – GELINA neutron production facility

GELINA neutron production facility :

- Located at EC-JRC Geel, Belgium
- 800 Hz pulsed neutron beam
- Electron induced photofission neutron from uranium target
- Neutron energy range from few keV to 20 MeV
- 12 flight path from 5 to 400 meter long

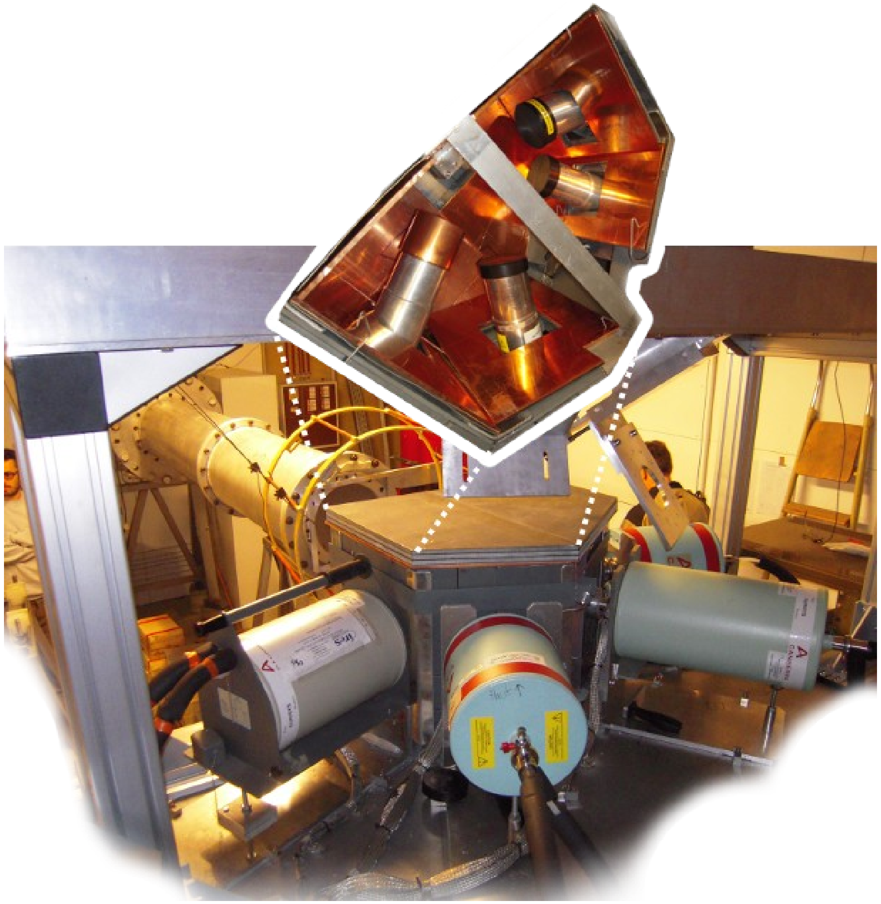


Birdview of GELINA facility

III. Experiment – GRAPhEME setup for ^{232}Th campaign

GRAPhEME (GeRmanium array for Actinides PrEcise Measurements) :

- 4 HPGe detectors placed at 110° and 150° of beam axis detect gamma rays produced
- One fission chamber placed ahead of the target determine neutron beam fluence
- Digital acquisition cards TNT2-100 MHz
- 232Th target of width e=0,3 mm located at circa 30 m from neutron source
- Neutron energy obtained using time of flight measurement (resolution 0.8 MeV at 20 MeV, 10 keV at 1 MeV)



GRAPhEME as used for ^{232}Th data collection (2009-2013)

III. Experiment – Cross section measurements

Differential cross section formula

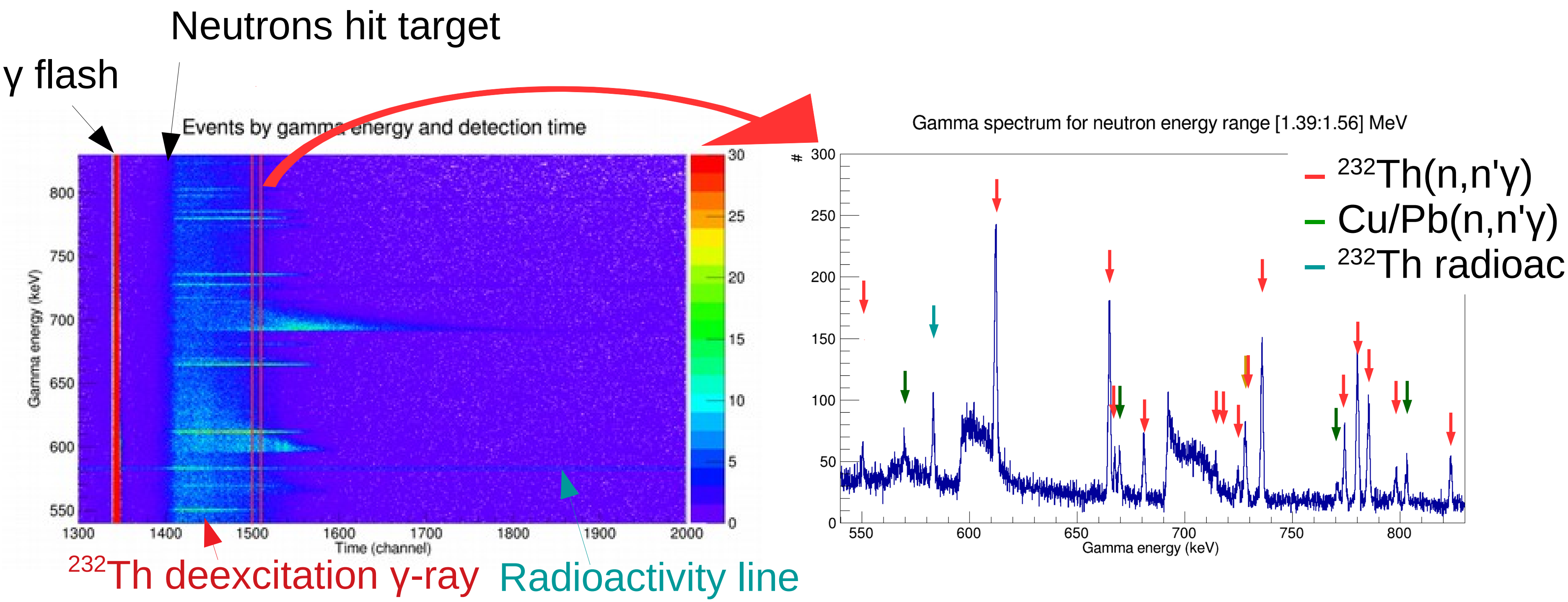
$$\frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(\theta_i, E_n) = \frac{N_\gamma(\theta_i, E_n)\,\tau_{pup,i}/\epsilon_{\gamma,i}}{4\pi N_n(E_n)\rho_{Th}}$$

$N_\gamma(\theta_i,E_n)$: Number of γ-rays of interest detected at angle θ_i for a time of flight equivalent to E_n
$\tau_{pup,i}$: Corrective factor due to pile up dead time
$\varepsilon_{\gamma,i}$: Efficacity of HPGe counter for considered γ-ray energy
ρ_{Th} : Areal atomic density of thorium target
$N_n(E_n)$: Number of impinging neutrons on thorium target for a time of flightl equivalent to E_n

$$N_n(E_n) = \frac{N_{fiss}(E_n)/\epsilon_{FC}}{\sigma_{f,U235}(E_n)\rho_{U235}}$$

$N_{fiss}(E_n)$: Number of fissions detected for a time of flight equivalent to E_n
ε_{FC} : Efficacity of fission chamber
$\sigma_{f,U235}(E_n)$: Fission cross section of U-235 for neutron energy E_n
ρ_{U235} : Areal atomic density of U-235 foil in fission chamber

IV. Analysis – Identification



Histogram ToF-E_γ for one HPGe **Projection for one time window**

Identification of gamma peaks, in nuclear structure databases, using :

- 1) Gamma energy centroid
- 2) time of flight where it has been observed

- Bidimensional spectrum is projected on several time of flight windows, each corresponding to a neutron energy interval
- Area under peak for each projection is needed to compute cross section on corresponding neutron energy interval

IV. Analysis – Total cross section

Combination of 4 HPGe measures :

- 2 times 2 partial cross sections at 110° et 150° from beam axis
- We need angle integrated cross section
- As transitions' multipolarity is always less than 3, following Gauss quadrature formula gives exact angle integrated cross section :
- $\sigma_{(n,xn\gamma)} = 4\pi [w_{110} \frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(110°, E_n) + w_{150} \frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(150°, E_n)]$
- This formula is the reason why HPGe were placed at 110° and 150°

Mean of 2 measures at one angle :

- Measures from 2 HPGe for one angle
- They are correlated (ρ_{Th}, $N_n(E_n)$ terms are same, $\varepsilon_{det,\gamma}$ may be correlated too).
- To maximise information, we need to use ponderated mean, using covariances
- Minimal uncertainty is achieved by using the following estimator σ(θ) to combine two measures σ_A,σ_B at the same angle θ

$$\sigma(\theta) = \frac{\frac{1}{Var(\sigma_A) - covar(\sigma_A, \sigma_B)} \sigma_A + \frac{1}{Var(\sigma_B) - covar(\sigma_A, \sigma_B)} \sigma_B}{\frac{1}{Var(\sigma_A) - covar(\sigma_A, \sigma_B)} + \frac{1}{Var(\sigma_B) - covar(\sigma_A, \sigma_B)}}$$

IV. Analysis – Uncertainties

Typical uncertainties (1σ values) :

1. From U-235 foil width ρ_{U235} : 0.55 %
2. On target aeral density ρ_{Th} : 1.3 %
3. From fission chamber efficacity ε_{FC} : 2.1 %
4. On HPGe photopeak efficacity $\varepsilon_{\gamma,i}$: 2 %
5. From $\sigma_{f,U235}(E_n)$: 0.5 to 1 %
6. Statistical and fit uncertainty on $N_\gamma(\theta_i,E_n)$: 2 to 20 %, depending on γ-ray observed and neutron energy
7. On correction factor due to pile up $\tau_{pup,i}$: 0.5 %
8. From statistical error on $N_{fiss}(E_n)$: 0.3 to 2 %

→ Minimal total uncertainty : 3.5 %

→ Including 3.3 % fully correlated over all E_n

$$\frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(\theta_i,E_n)=\frac{\left(N_\gamma(\theta_i,E_n)/\epsilon_{\gamma,i}\right)\tau_{pup,i}\rho_{U235}\sigma_{f,U_{235}}(E_n)}{4\pi\left(N_{fiss}(E_n)/\epsilon_{FC}\right)\rho_{Th}}$$

Differential cross section formula

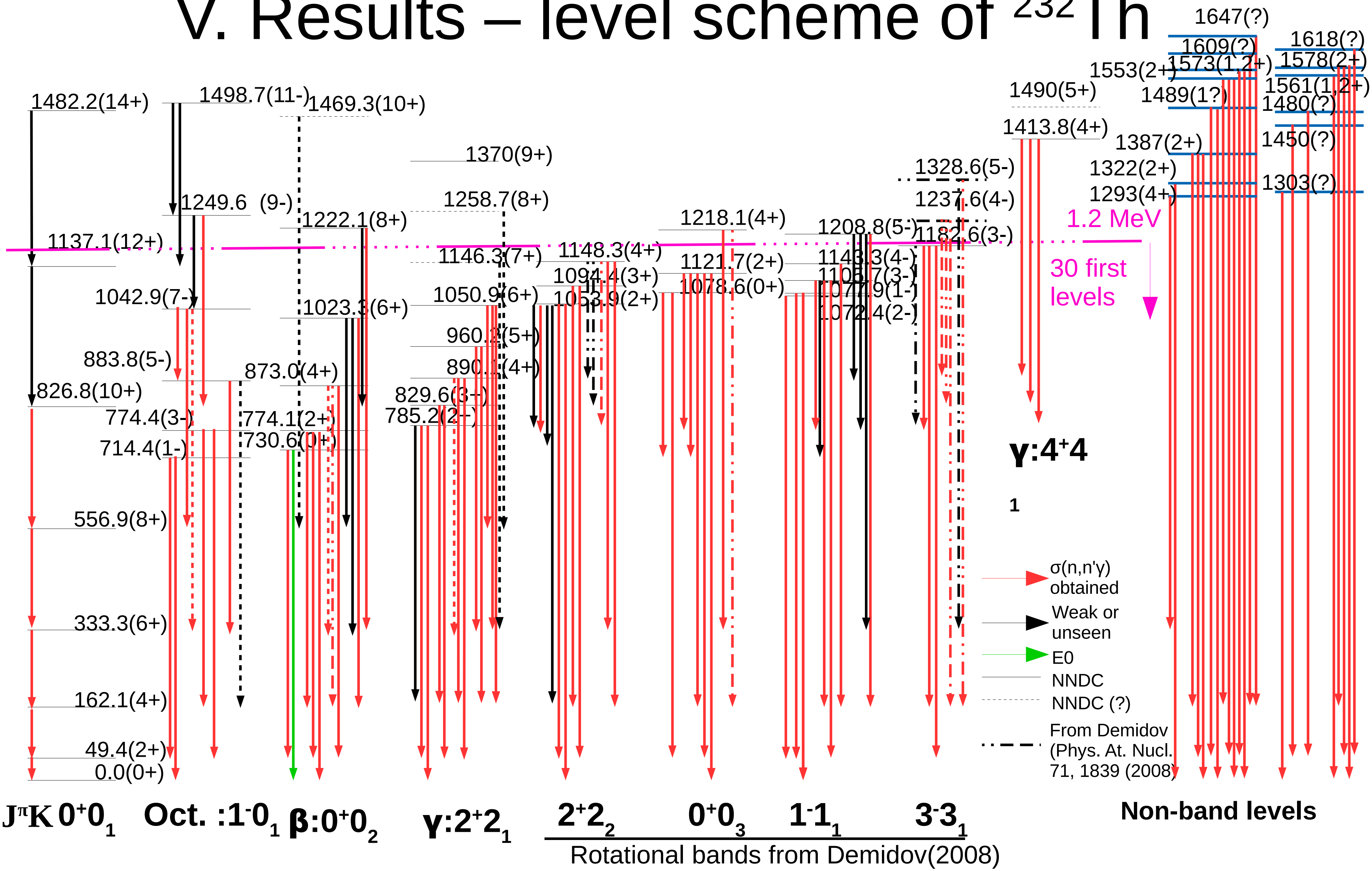
Different measures' points are identified by :

- Neutron energy range observed E_n
- γ-ray considered
- HPGe counter number i={1,2,3,4}

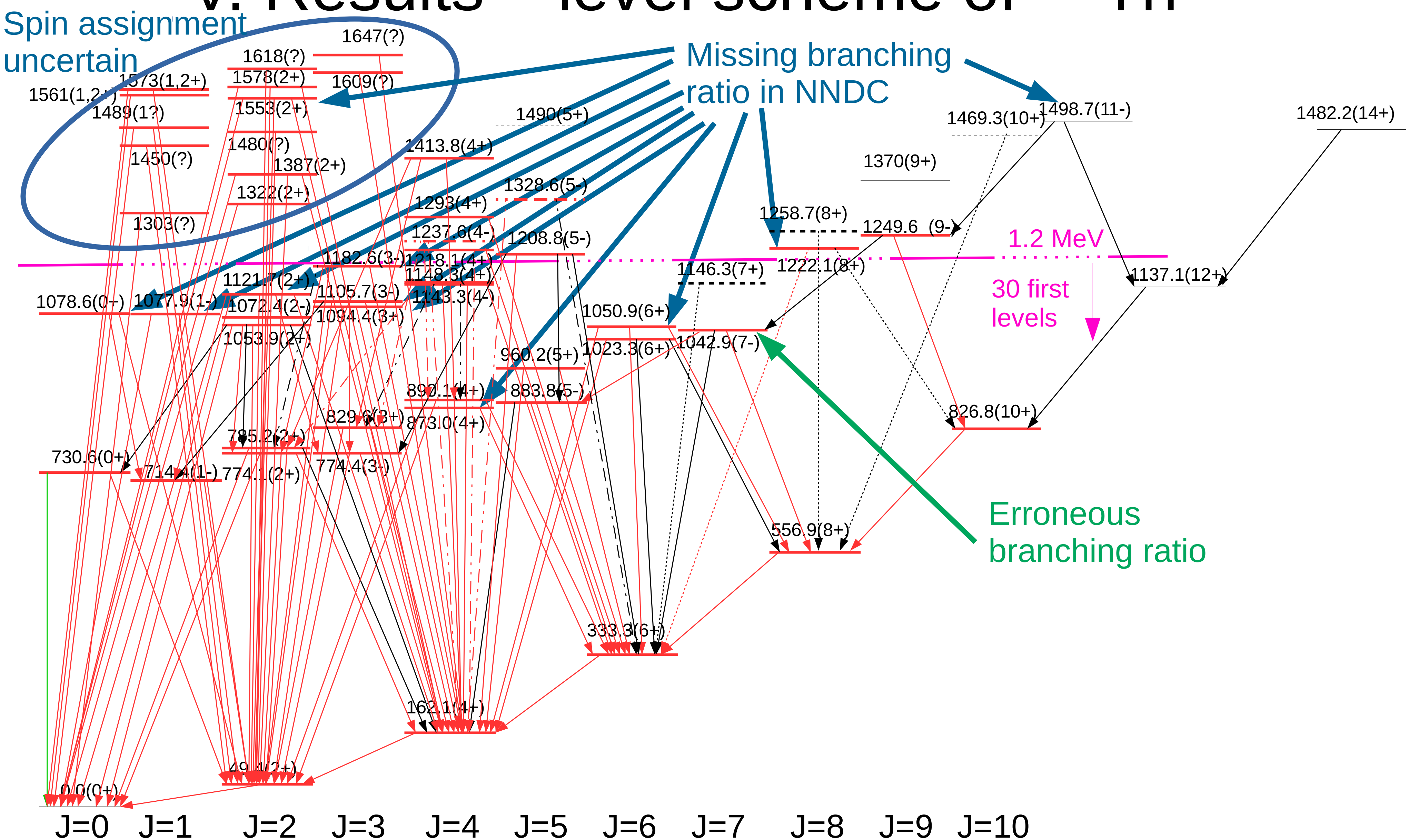
Variance is obtained by classic error propagation

Covariance between two measure points has been obtain the covariance of each terms in both points formula

V. Results – level scheme of ^{232}Th



V. Results – level scheme of ^{232}Th



V. Results – spectroscopic problems

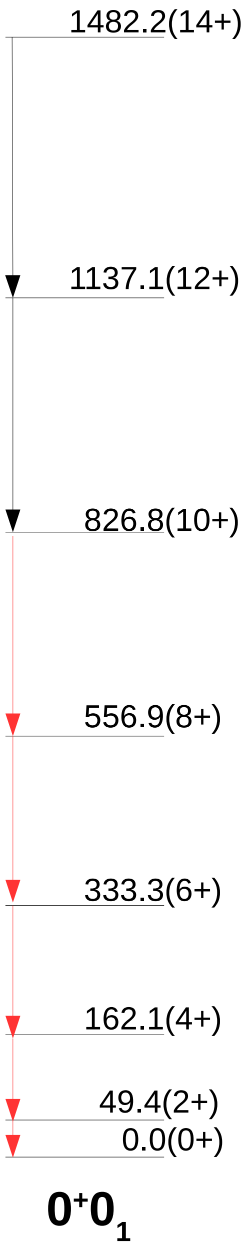
Lacking branching ratio :

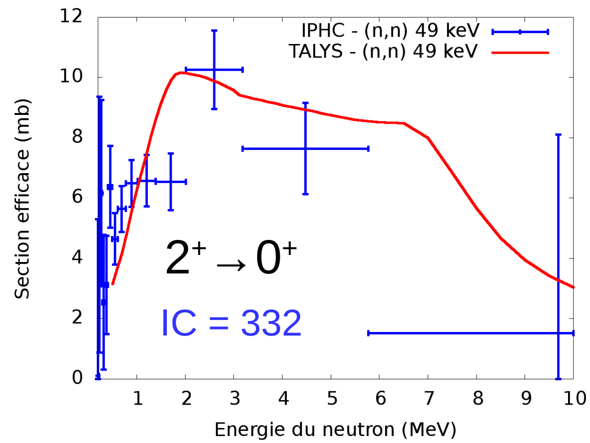
- 5 transitions at 347, 407, 959, 1072 and 1122 keV for decay of level 1122 keV(2+) (from NNDC)
- Branching ratio for 347, 407 and 959 keV gamma emissions only
- From our data, I_γ=300(30) for 1072 keV gamma emission
- Dominant branching ratio is not mentionned !
- Other studies support dominance of 1072 keV deexcitation : [Demidov (2008), McGowan (Nuc. Phys. A562-1993)]
- Moreover, some assigned branching ratio seem erroneous

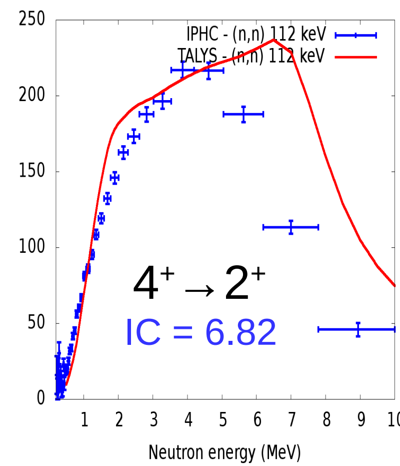
Unidentified γ-rays and levels :

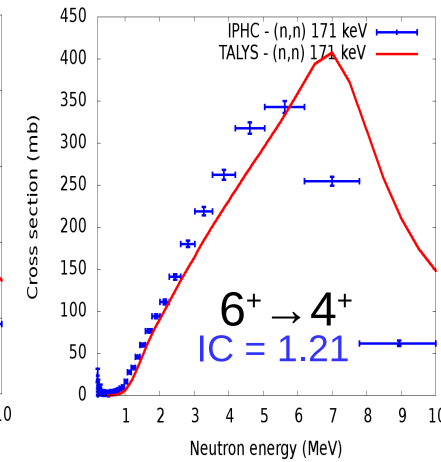
- Non-registered γ-ray observed at 1075.5 keV
- Deduced cross section is compatible with inelastic scattering origin
- Observed by Demidov (Phys. Atomic Nuclei, 2008) who assigned it to a 4- state at 1237.6 keV
- Many gamma peaks are still unidentified

V. Results – Ground state rotational band

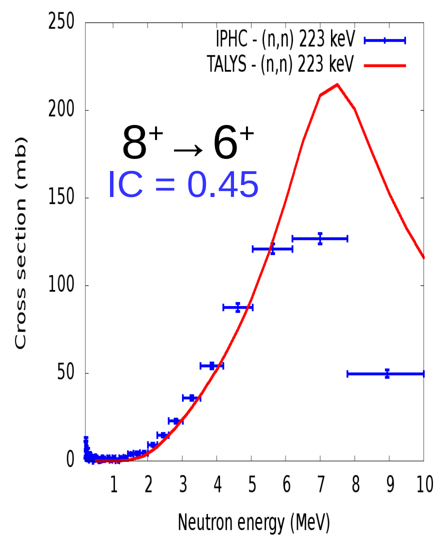


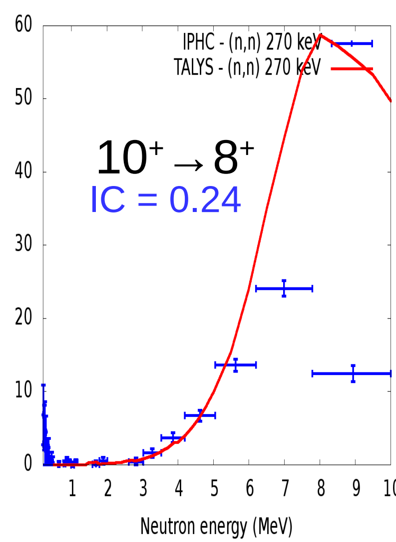




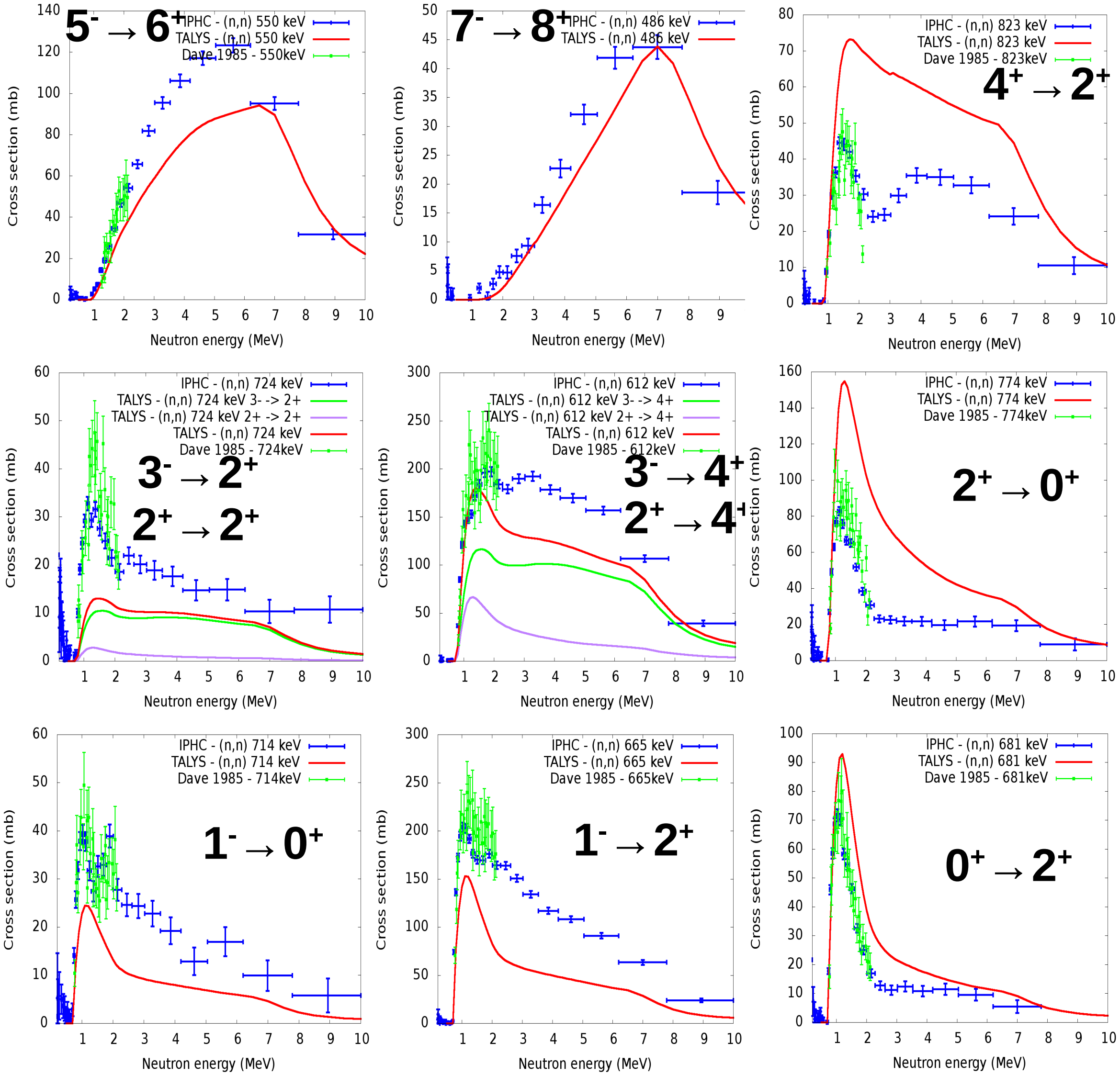


- At higher spin → weaker cross section ; maximum for higher neutron energy
- Default Talys prediction maximum cross section position is at higher energy
- Strong internal conversion for low energy transitions decrease cross section (IC =332 for transition $2^+ \rightarrow 0^+$ de 49 keV)





V. Results – Some other transitions



- Diverse shapes depending on excited levels
- Good agreement between **our measurements** with the ones from **Dave et al.** (1985)
- Varying agreement with default TALYS prediction depending on level concerned

V. Results – Summary

After 800 beam hours :

- 81 (n,n'γ), 11 (n,2nγ), 7 (n,3nγ) cross sections obtained for neutron energy from 0.2 MeV to 20 MeV
- Maximum cross section from 10 mb to 400 mb
- Good agreement with Dave(1985) for neutrons from 0.7 to 2.2 MeV
- Dozens of cross sections never registered in EXFOR before

On uncertainty quantification :

- 3 % for stronger transitions (ground band $4^+ \rightarrow 2^+, 6^+ \rightarrow 4^+$…) mainly from HPGe efficacity, fully correlated between neutron energies
- Up to 20 % for weaker transitions, mainly from statistic uncertainty, uncorrelated between neutron energies
- Covariances obtained between all measure points

VI. Additional works – Theory

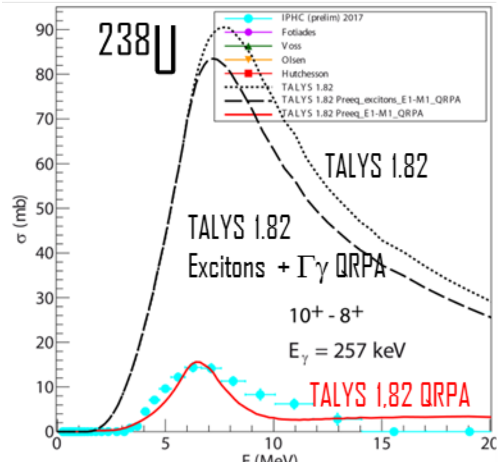
Collaboration with theoreticians from CEA/DAM/DIF have been engaged for other isotopes (^{238}U, W …), is beginning on ^{232}Th

<u>Issues :</u>

- Is there a need of better nuclear structure knowledge for theory ?
- Can theory reproduce our measurements ?
- To what extent reaction mechanisms are constrained by our partial measurements ?

<u>Example of ^{238}U :</u>

- In ^{238}U, some features have been reproduced by calculation
  - Drop of XS at high spin reproduced using QRPA calculated spin distribution of residual nucleus instead of ad hoc prescription exciton (Dupuis et al., ND2016 - EPJ Web of Conf.146, 12002 (2017))



^{238}U measurements and theoretical predictions

VI. Additional works – Sensitivity studies

- We stressed inelastic scattering importance in reactor applications
- How exactly do it matters ?
- Sensitivity studies allow to quantify effect of variation of XS on reactor parameters
- To compare importance relative to other reactions, all reactions on both ^{232}Th and ^{233}U are studied

On sensitivity :

- Sensitivity of A to B is the quotient of relative variation of a variable A stemming from the relative variation of parameter B
- Its unity is « % / % »
- It is similar to a first order derivative
- Linear approximation appliable only for small variations
- Could be used to propagate moderate uncertainties

VI. Additional works – Sensitivity studies

Objectives :

- Compute **reactor's parameters sensitivity** to nuclear cross section of Th-232 and U-233 (neutron multiplication coefficient k_{eff}, effective delayed neutron fraction β_{eff}, radial power distribution…)
- **Validation** of these results by calculation of variance using :

  ➔Sensitivities and covariance matrices

  ➔Total Monte Carlo (TMC) method
- **Apply** this method **to different reactor core design** (Pressurized Water Reactor, Sodium-cooled Fast Reactor, Molten Salt Fast Reactor)

Tools used :

<u>Transport codes :</u>

- MCNP6 and SERPENT2
- Both Monte Carlo neutron transport code
- Sensitivity calculation tools recently added (some years ago)

<u>Nuclear data library :</u>

- 300 randomized TENDL-2013 files used for TMC calculations
- ENDFB and JEFF library files used for reference sensitivity calculations

<u>Computational power :</u>

- Computation resources of CC-IN2P3

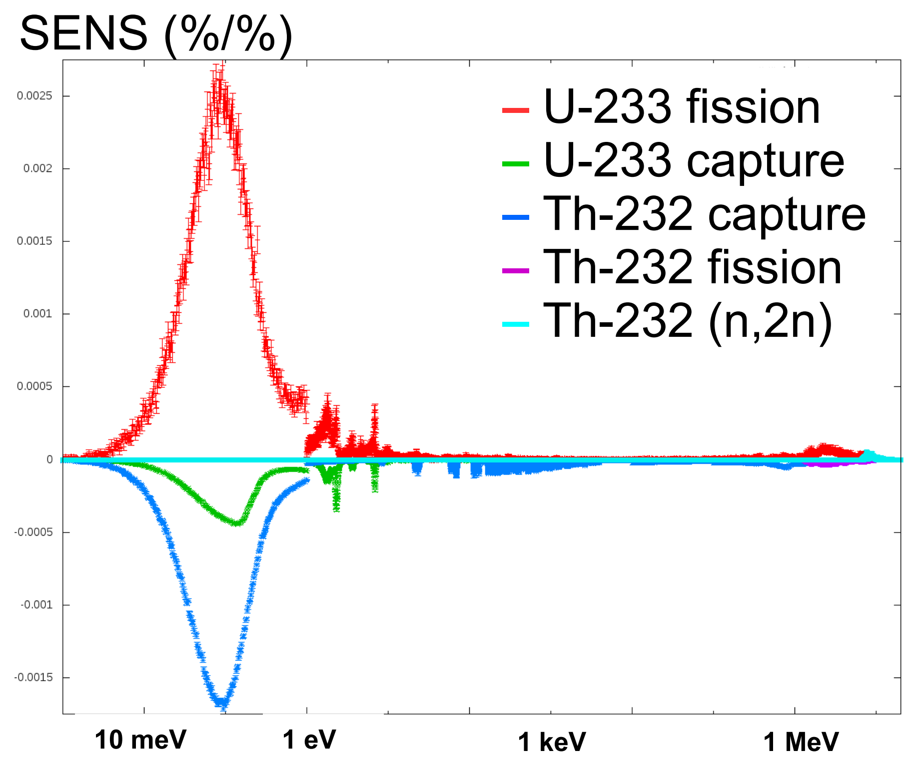
VI. Additional works – Sensitivity studies

Sensitivities of k_{eff} to cross sections in PWR :

- Capture on ^{233}U and ^{232}Th, and fission on 233U at low energy (E_n< 1 eV) are most sensitives reactions
- k_{eff} sensitivity to inelastic scattering is less than 100 time lower
- Work in progress for other parameters and reactors

Isotope	Reaction	SENS SERPENT2
Th-232	capture	**-0.2951** %/%
	fission	0.0056 %/%
	elastic	0.0083 %/%
	inelastic	-5.57e-4 %/%
	n2n	0.0014 %/%
U-233	capture	-0.0754 %/%
	fission	**0.3668** %/%
	elastic	-2.27e-4 %/%
	inelastic	-3.45e-5 %/%
	n2n	1.09e-5 %/%

Energy integrated sensitivities



Sensitivities for U3/Th mix fueled PWR depending on neutron energy

VI. Additional works – Future measurements

- Deexcitation by internal conversion not observed with GRAPhEME (ground state band $2^+ \rightarrow 0$ gamma emission is very weak due to 332 IC factor)
- DELCO project aim to conceive a new instrument to allow internal conversion electrons detection
- Work in progress in the team

- ^{233}U experimental data collected, to be analyzed
- ^{239}Pu target in development to be empolyed within GRAPhEME
- Dedicated nuclear structure studies of actinides by combining GRAPhEME and GAINS setup is considered

Conclusion

Realized :

- Numerous cross sections obtained, soon to be in EXFOR
- Covariances of cross sections have been derived
- Sensitivity studies are in progress

To come :

- Preparation of new measurements (electron, ^{239}Pu)
- Collaboration with theoreticians on collected cross sections

Thanks for your attention