Fission studies with with the LOHENGRIN spectrometer

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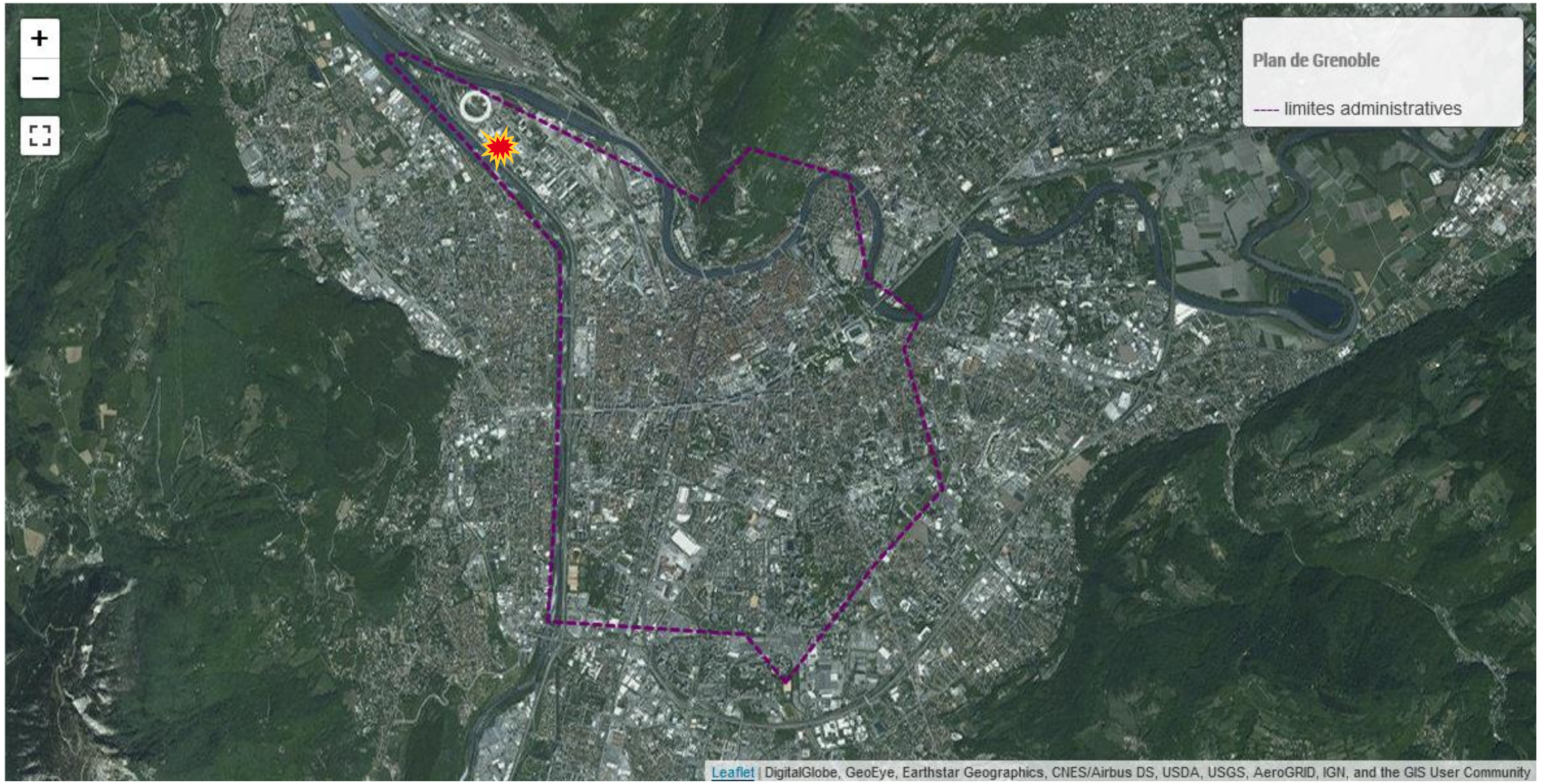
A. Chebboubi, G. Kessedjian, O. Serot, D. Bernard, O. Litaize, M. Houdouin-Quenault, S. M. Cheikh
CEA, DES, IRESNE, DER, SPRC, LEPh, Cadarache, 13108 Saint-Paul-lès-Durance
C. Sage, O. Méplan, M. Ramdhane, A. Vieville, J-F. Muraz, O. Guillaudin, F. Collovati
LPSC, Université Grenoble-Alpes, CNRS/IN2P3, 38026 Grenoble, France
U. Köster, Y. H. Kim, P. Mutti, J-M. Daugas
Institut Laue-Langevin, 38042 Grenoble, France
M. Diakaki
National Technical University of Athens, Athens, Greece

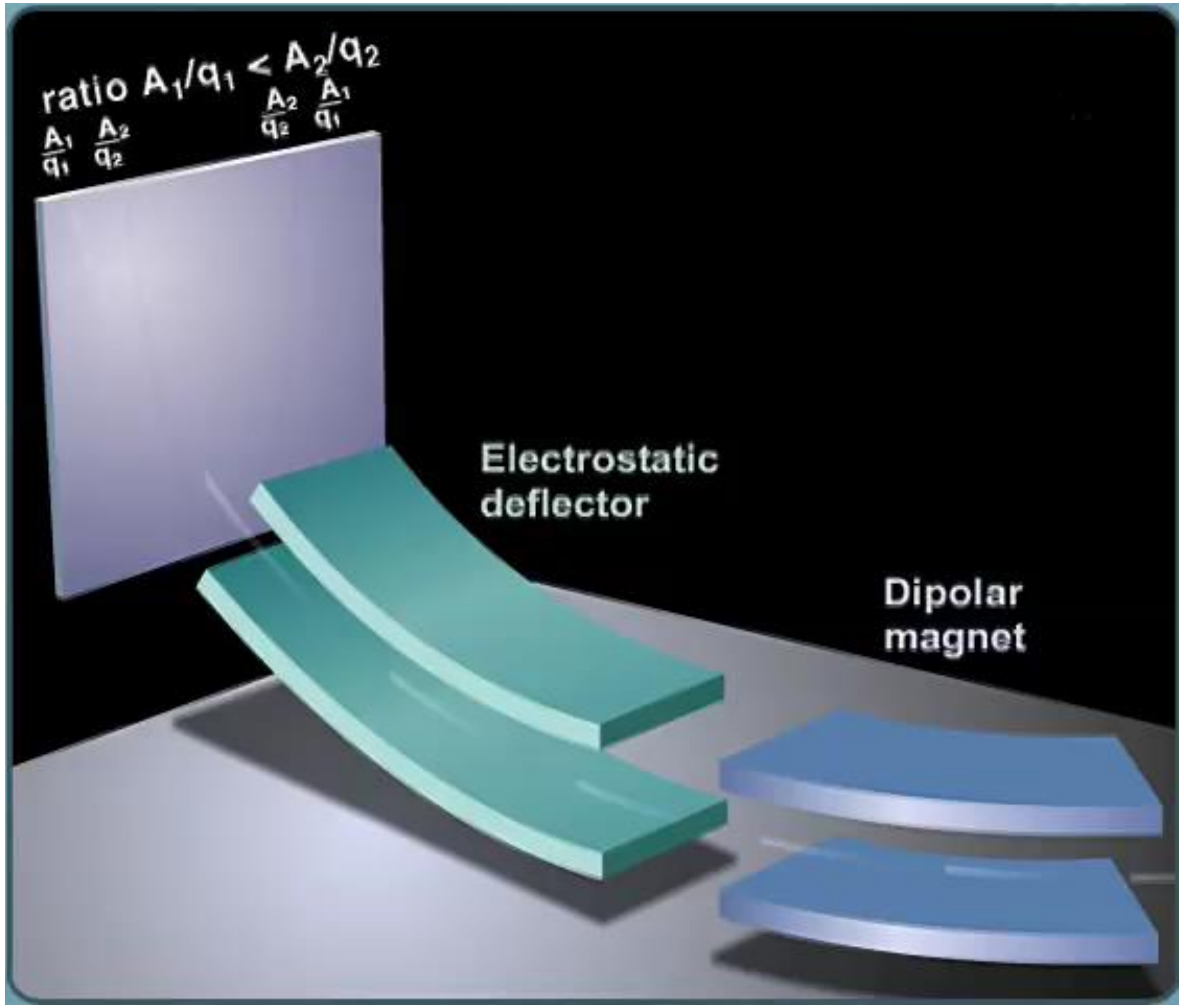
Institut Laue-Langevin

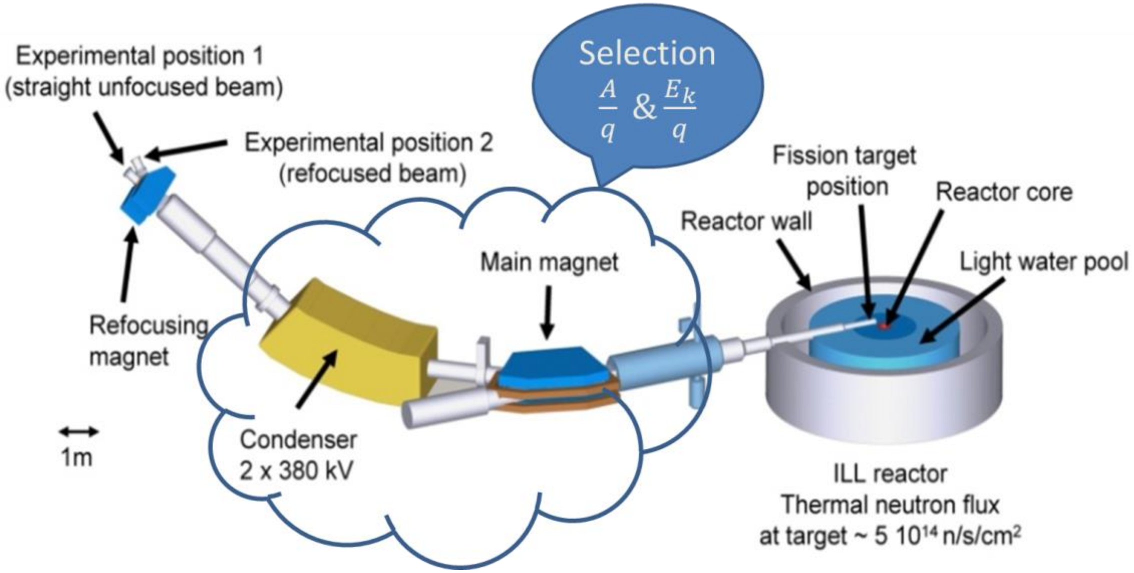
ILL : founded and govern by France, Germany and United Kingdom

- Build in 1967
- 40 instruments (mainly neutron spectroscopy for biology, materials …)
- 540 member staff + 1400 users per year
- 105 M€ per year
- High Flux Reactor : 58.3 MW thermal. New vessel in 1995.



LOHENGRIN working principle



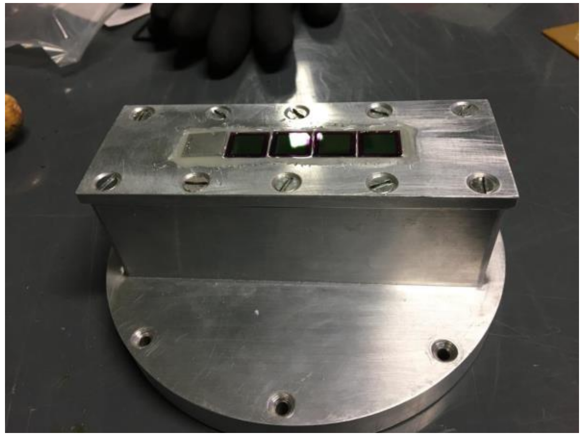


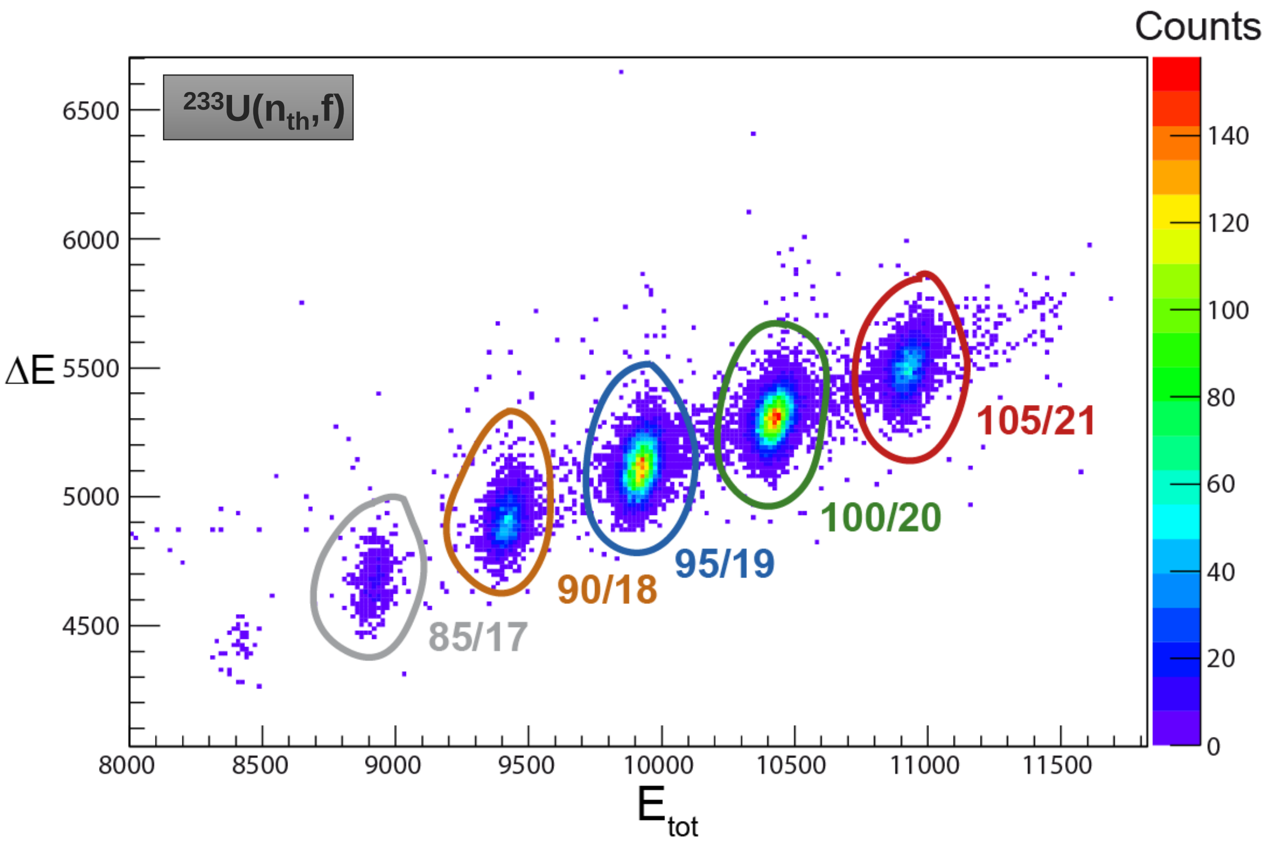
Lohengrin : selection with the mass over ionic charge $\frac{A}{q}$ and Kinetic energy over Ionic charge $\frac{E_k}{q}$ ratios

$$(A_1, E_1, q_1) \equiv (A_2, E_2, q_2) \equiv (A_3, E_3, q_3)$$

1. Measurements of fission fragment mass yields

Measurements of fission product mass yields : experimental setup





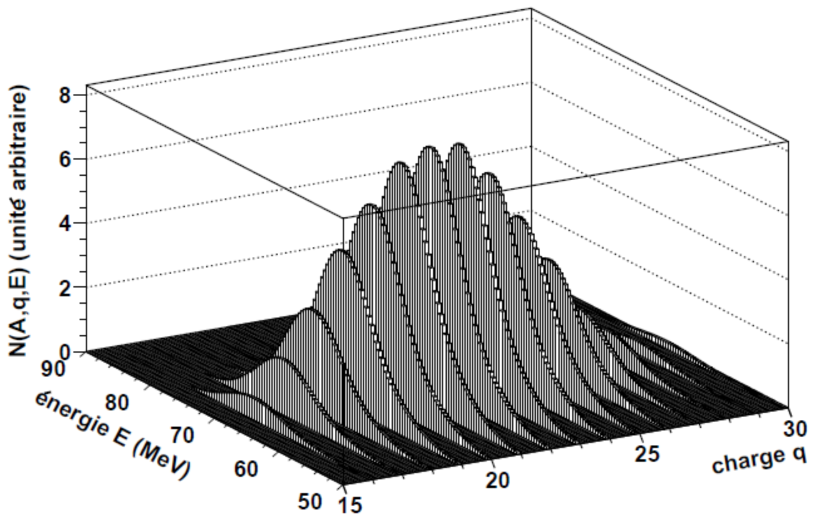
- Split anode Frish Grid chamber
- 30 cm long
- Use of Isobutane @ 30-40 mbar
- Thin SiN entrance windows (100 nm)
- σ_{E_k}~300 keV @ 100 MeV

Ionization chamber $\Delta E - E$

Mass A identification through measurement of kinetic energy E_k

→ Removing mass degeneracy

How to measure mass yields at LOHENGRIN



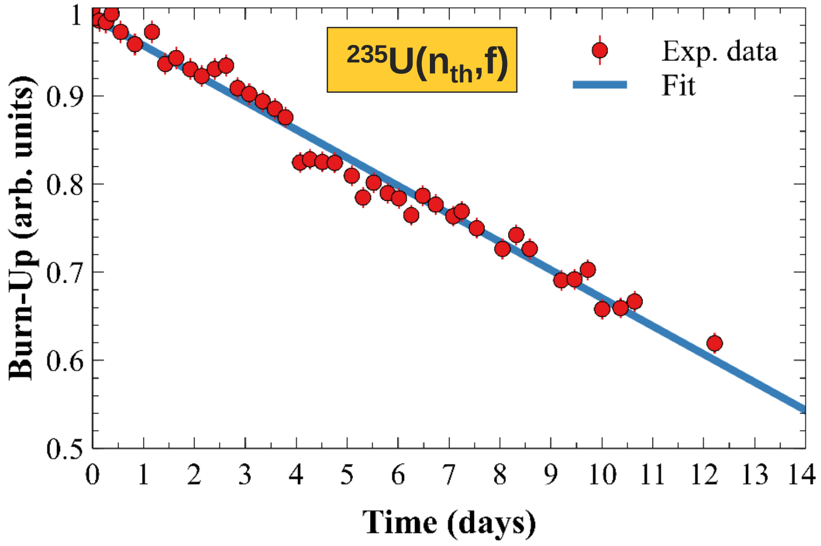
Relative measurements

$$\mathcal{N}(A) = \sum_q \int_{E_k} \frac{\mathcal{N}(A, q, E_k) dE_k}{BU(t)}$$

Absolute assessment

$$Y(A) = \frac{\mathcal{N}(A)}{\sum_A \mathcal{N}(A)}$$

$$\sum_{Heavy\ A} Y(A) = 1$$



Main issue : burning of the target BU(t) and beam time

- Choices E_k, q distributions must be made
- Correlations between E_k and q make the analysis more complex
- Tremendous effort over 15 years to reduce the uncertainties and handle bias !

Current data taking :

- 3 E_k scan & 1 q scan to measure a mass yield (at least)
- For some masses (high electronic conversion) more scan are mandatory

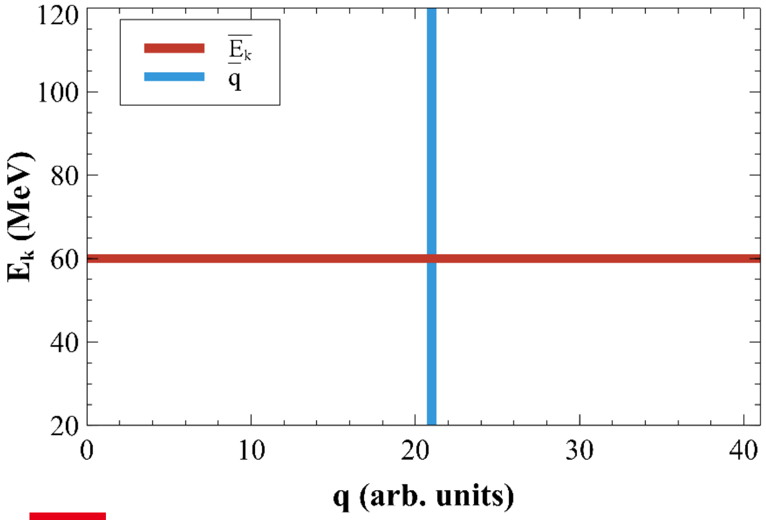
Estimation of mass yields : ideal case

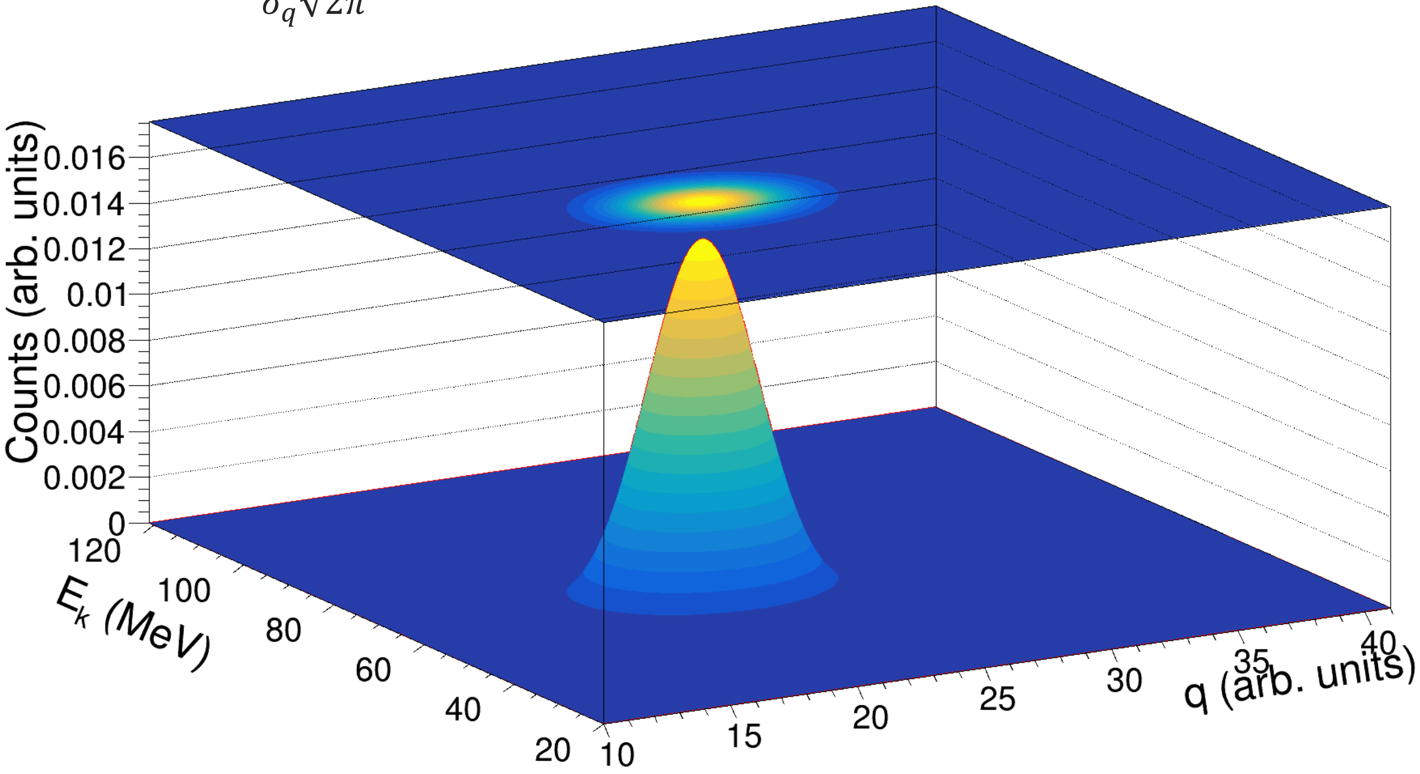
Gaussian shape in (E_k, q) space

$$h(E_k,q) = f\left(E_k|\overline{E_k},\sigma_{E_k}\right) \times g\left(q|\bar{q},\sigma_q\right) = \frac{\exp\left(-\frac{1}{2}\left(\frac{(E_k-\overline{E_k})^2}{\sigma_{E_k}^2}\right)\right)}{\sigma_{E_k}\sqrt{2\pi}} \times \frac{\exp\left(-\frac{1}{2}\left(\frac{(q-\bar{q})^2}{\sigma_q^2}\right)\right)}{\sigma_q\sqrt{2\pi}}$$

Discretization + LOHENGRIN resolution :

$$N(E_k,q) = \int_{E_k-\frac{\Delta E_k}{2}}^{E_k-\frac{\Delta E_k}{2}} h(E_k,q)dE_k$$





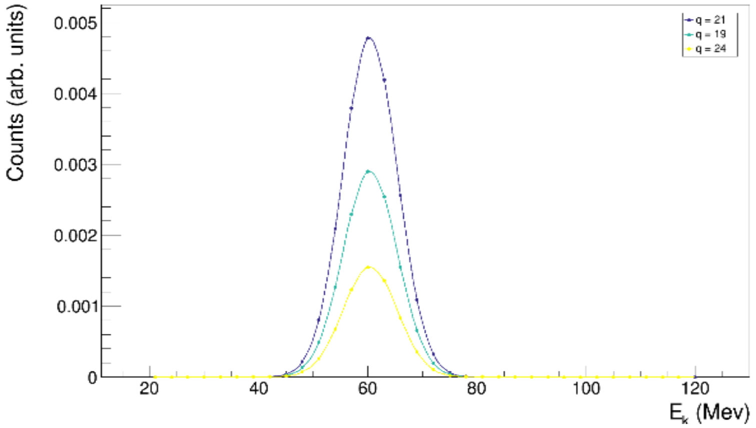
Estimation of mass yields : ideal case

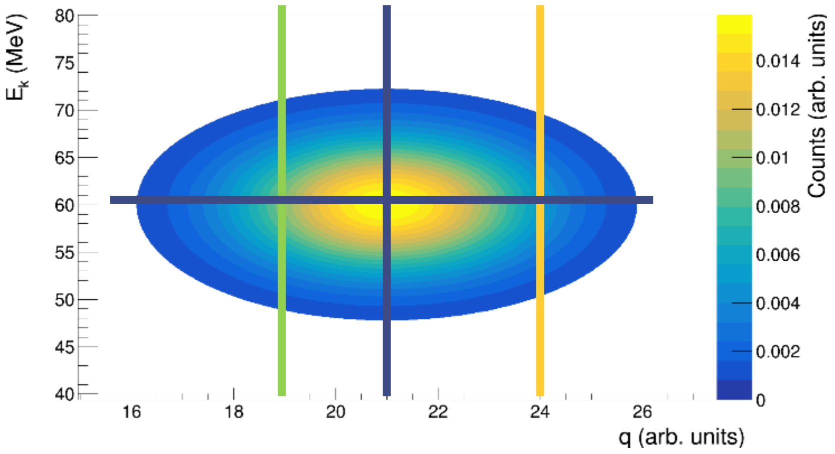
Real mass yield value :

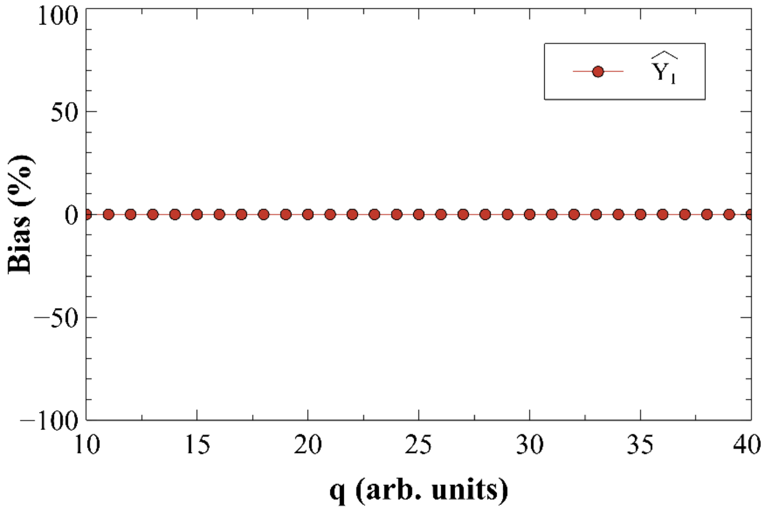
$$Y = \sum_{E_k} \sum_{q} \frac{N(E_k, q)}{\Delta E_k}$$

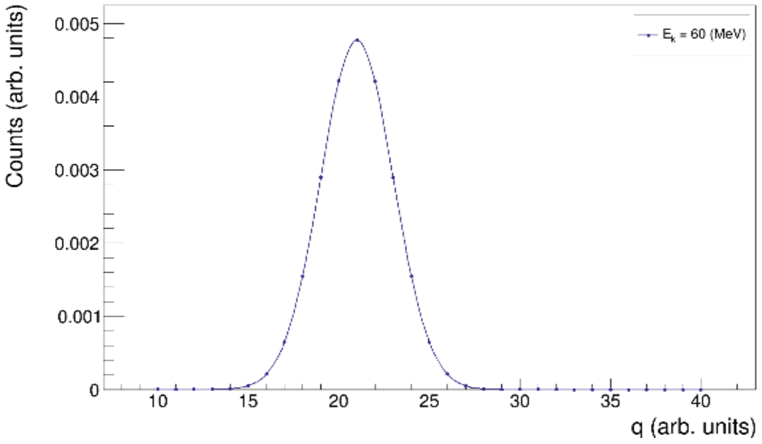
Estimator(s)

$$\widehat{Y_1} = \frac{1}{P(q)} \sum_{E_k} \frac{N(E_k|q)}{\Delta E_k}, \; P(q) = \frac{N(q|E_k^{\times})}{\sum_q N(q|E_k^{\times})}$$









Estimation of mass yields : 1D correlation

Gaussian shape in (E_k, q) space

$$h(E_k,q) = f\left(E_k|\overline{E_k}(q),\sigma_{E_k}\right) \times g\left(q|\bar{q},\sigma_q\right) = \frac{\exp\left(-\frac{1}{2}\left(\frac{\left(E_k-\overline{E_k}(q)\right)^2}{\sigma_{E_k}^2}\right)\right)}{\sigma_{E_k}\sqrt{2\pi}} \times \frac{\exp\left(-\frac{1}{2}\left(\frac{(q-\bar{q})^2}{\sigma_q^2}\right)\right)}{\sigma_q\sqrt{2\pi}}$$

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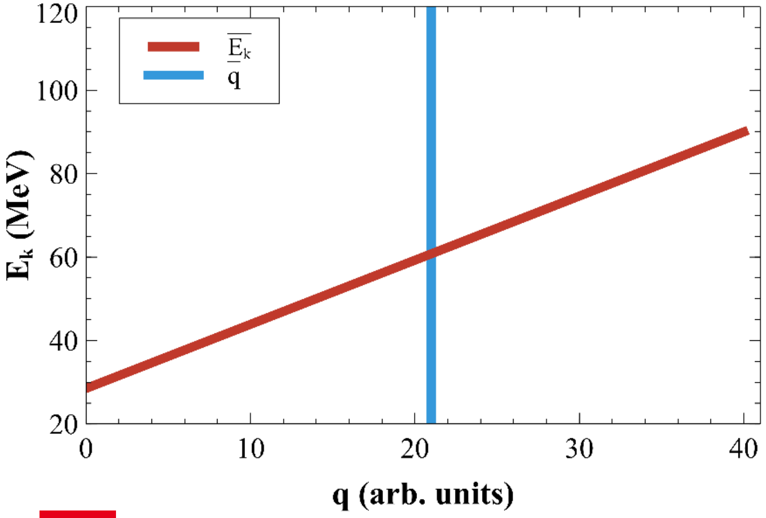

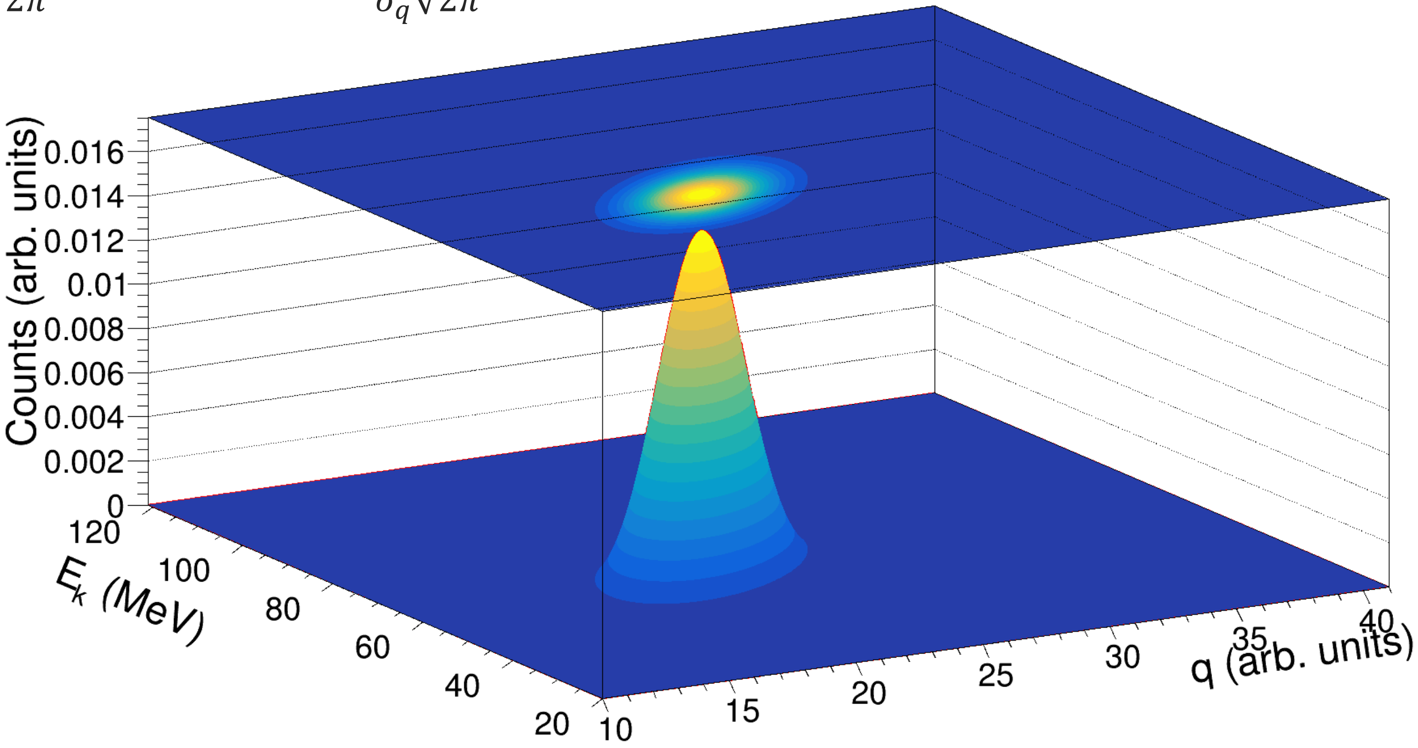

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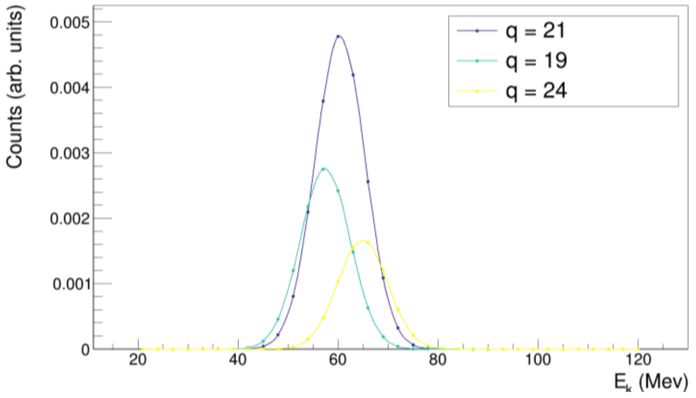
Estimator(s)

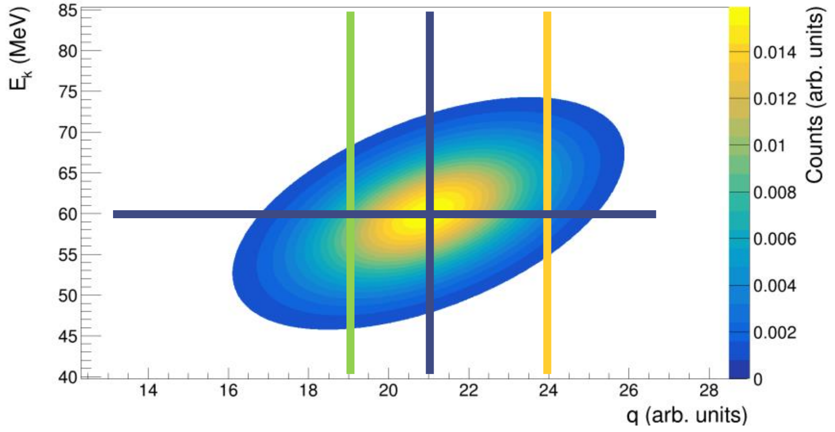
$$\widehat{Y}_1 = \frac{1}{P(q)} \sum_{E_k} \frac{N(E_k|q)}{\Delta E_k}, \; P(q) = \frac{N(q|E_k^{\times})}{\sum_q N(q|E_k^{\times})}$$

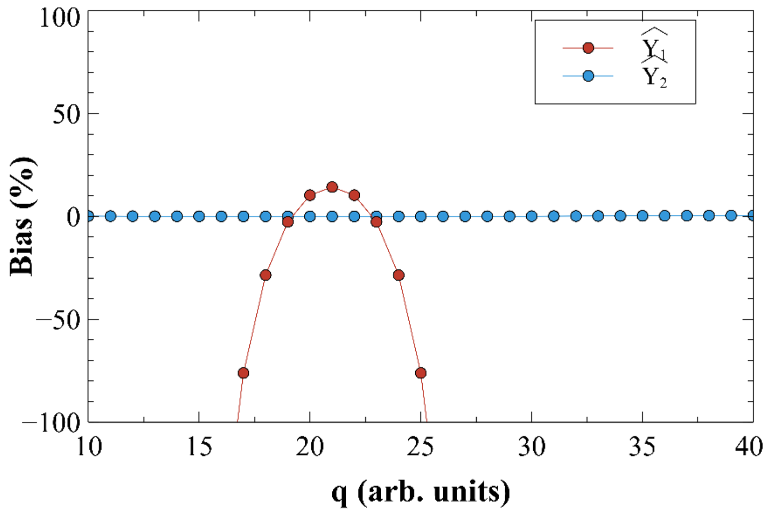
$$\widehat{Y}_2 = \frac{1}{P(q)} \sum_{E_k} \frac{N(E_k|q)}{\Delta E_k},$$

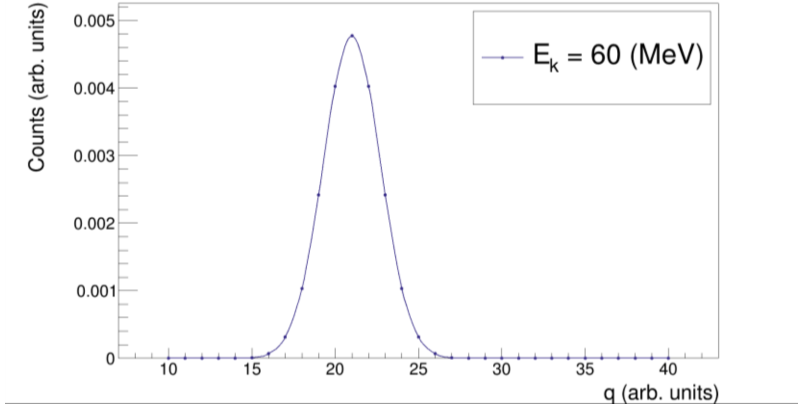
$$P(q) = \frac{N(q|E_k^{\times})/f\left(E_k^{\times}|\overline{E_k}(q), \sigma_{E_k}\right)}{\sum_q N(q|E_k^{\times})\,/f\left(E_k^{\times}|\overline{E_k},(q)\sigma_{E_k}\right)},$$

$$\sum_q P(q) = 1$$





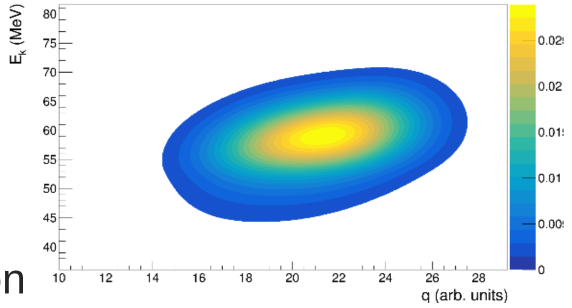


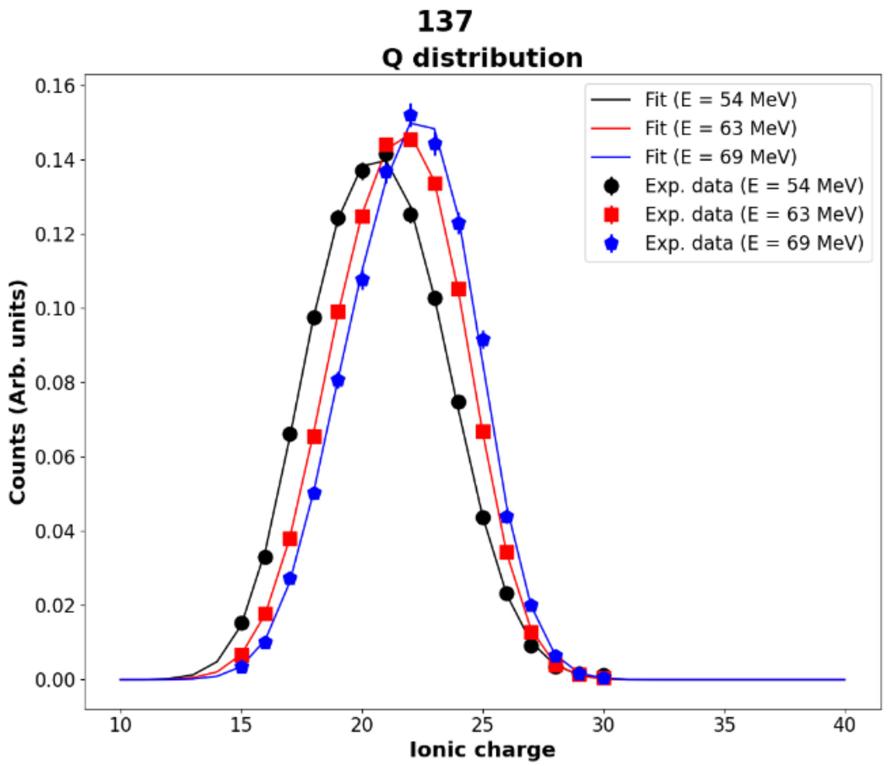


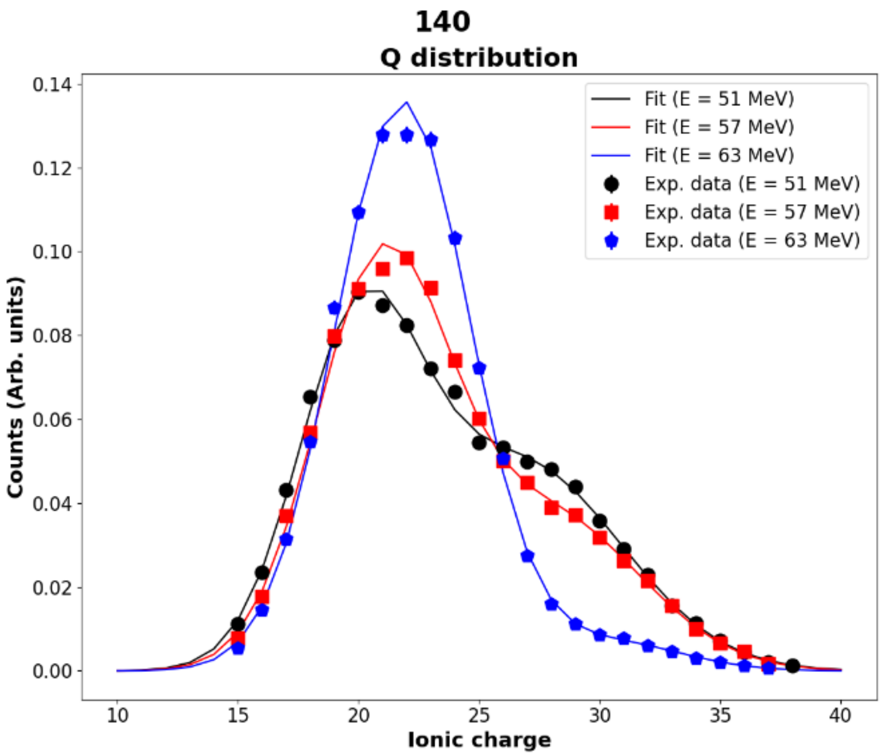
To go further : nanosecond cases

- In region [139-150] ionic charge distribution are deformed (ns isomers)

→ origin of the bias?
- Deformation depends on the kinetic energy
- **Additional correction in order take into account such effect : 2D correction**
- Bias between 1D and 2D corrections are propagated for masses without data for 2D correction







Estimation of mass yields : 2D correlation

Gaussian shape in (E_k, q) space

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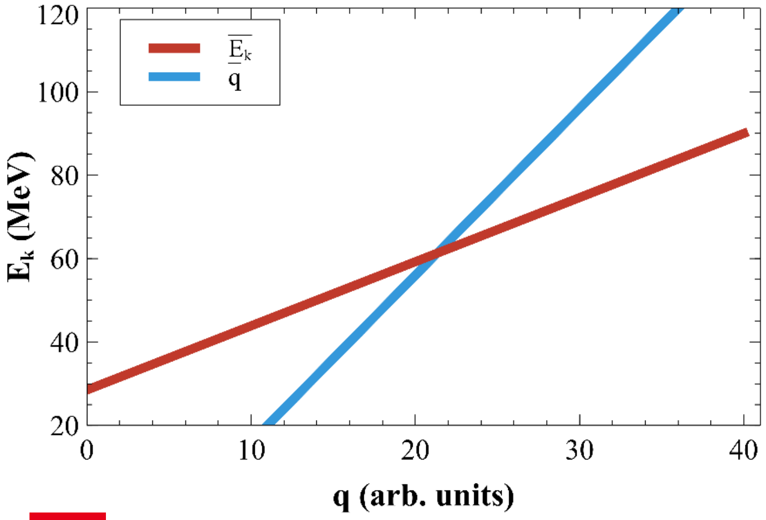

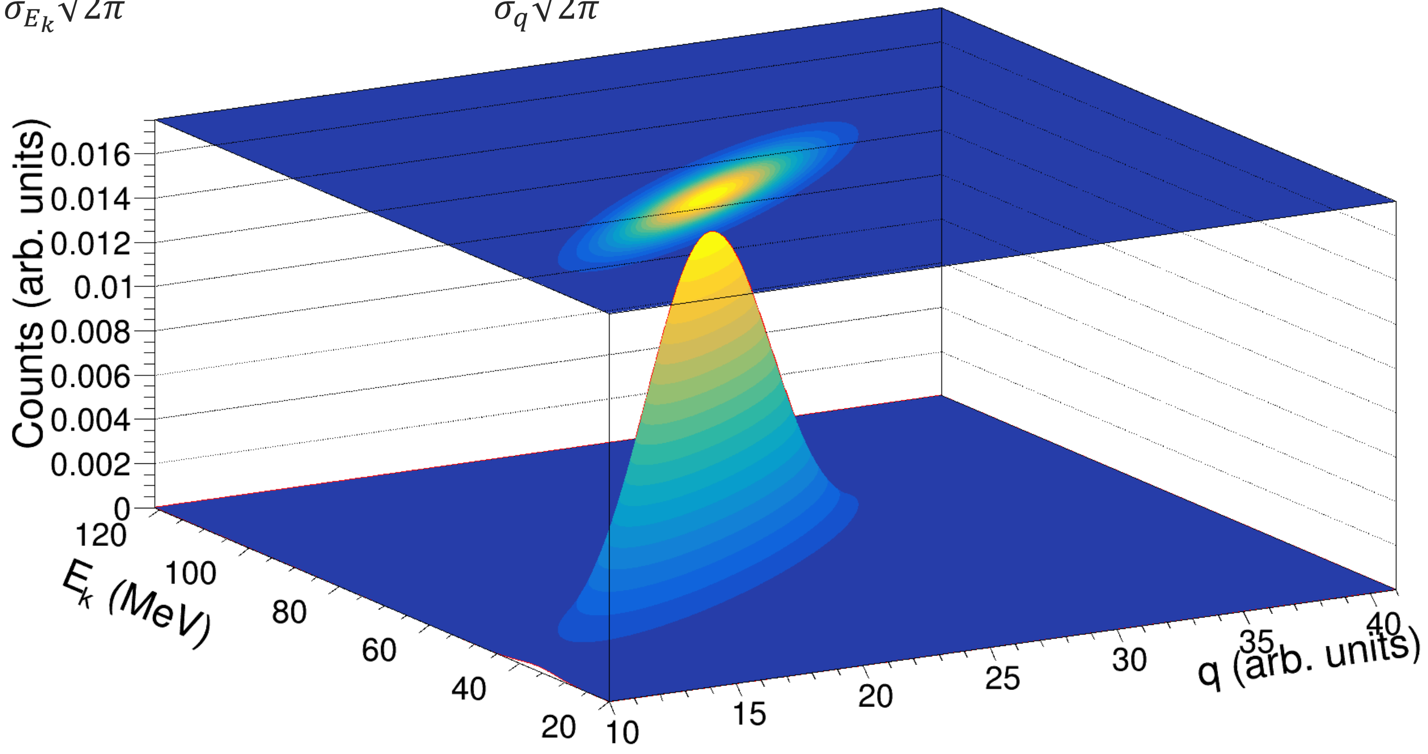

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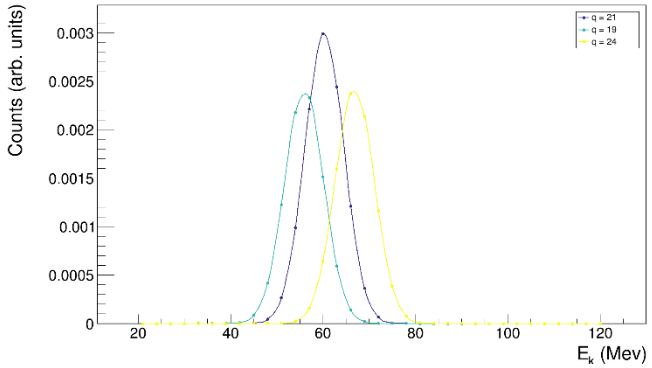
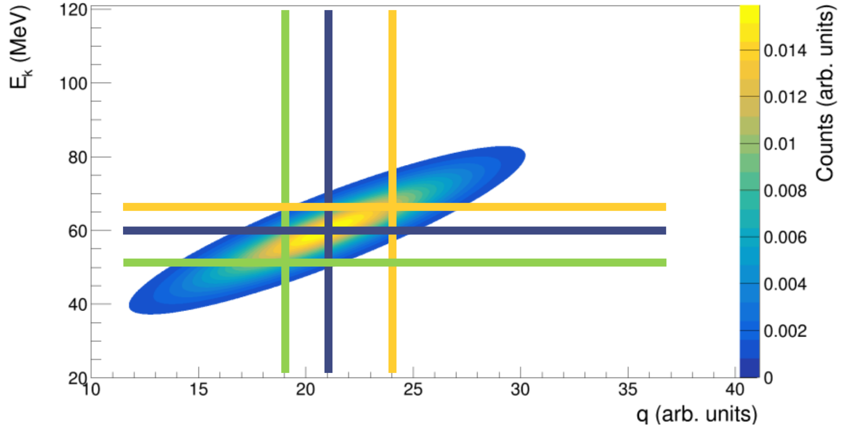
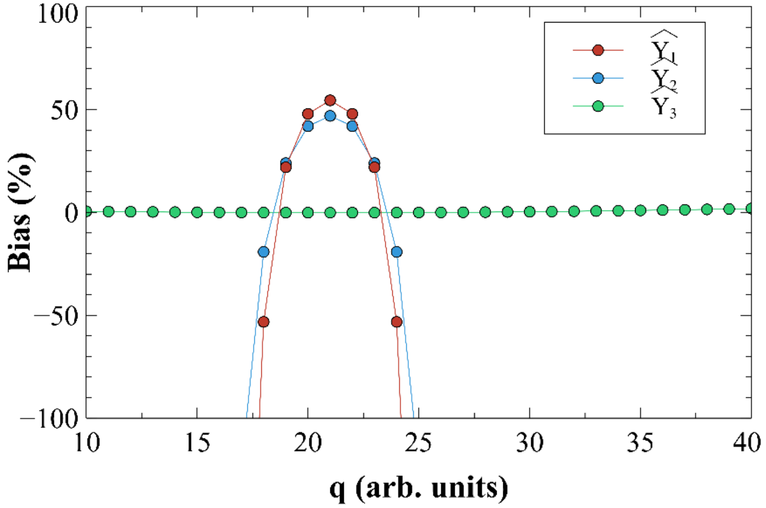
$$\sum_q P(q) = 1$$

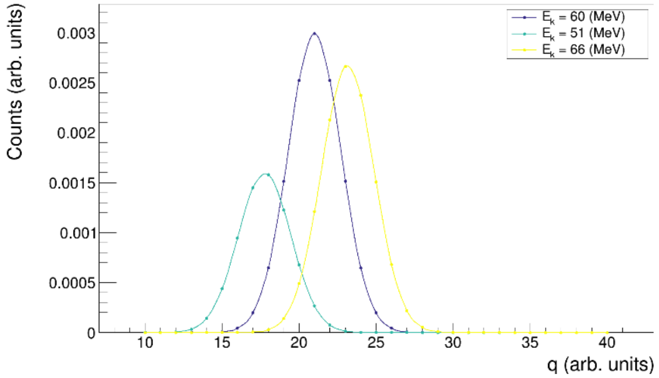
$$\widehat{Y_3} = \frac{1}{P(q)} \sum_{E_k} \frac{N(E_k|q)}{\Delta E_k},$$

$$P(q) = \frac{\dfrac{N(q|E_k^{\times})}{f\left(E_k^{\times}|\overline{E_k}(q), \sigma_{E_k}\right) \times g\left(q|\bar{q}(E_k^{\times}), \sigma_q\right)}}{\dfrac{\sum_q N(q|E_k^{\times})}{f\left(E_k^{\times}|\overline{E_k}(q), \sigma_{E_k}\right) \times g\left(q|\bar{q}(E_k^{\times}), \sigma_q\right)}},$$



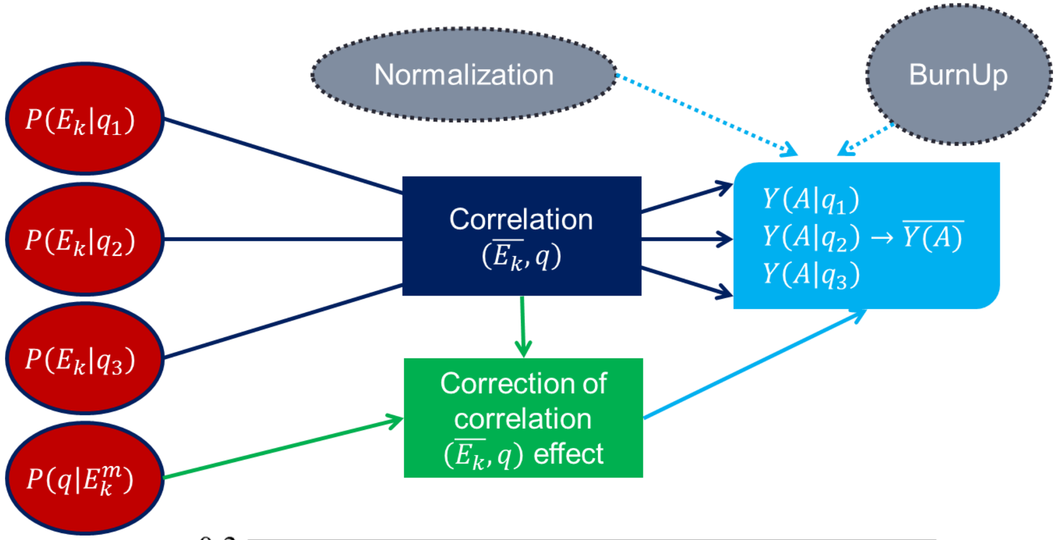


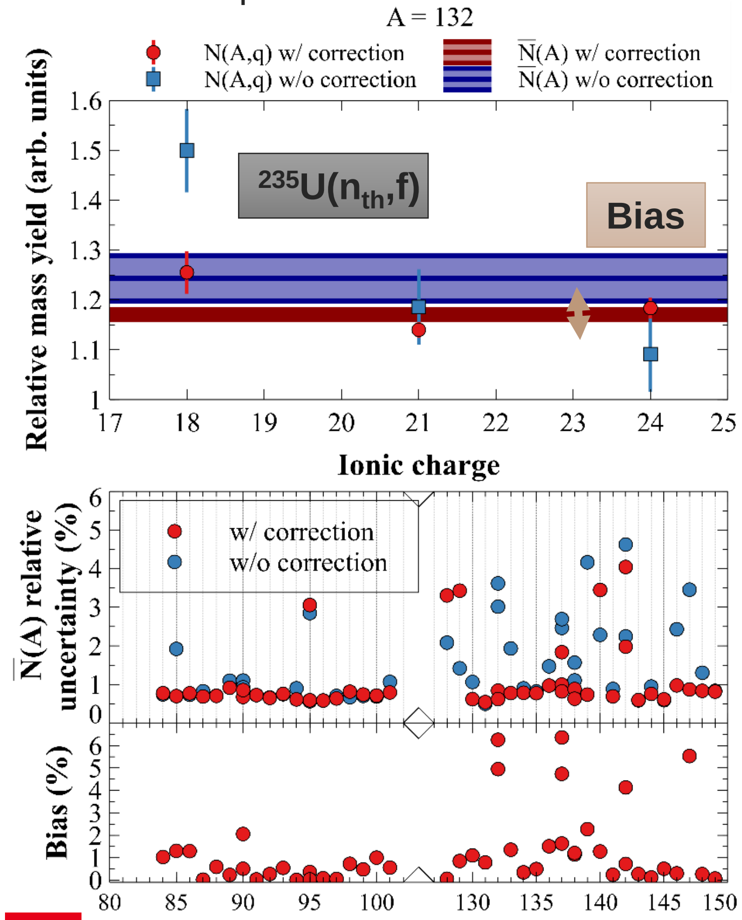




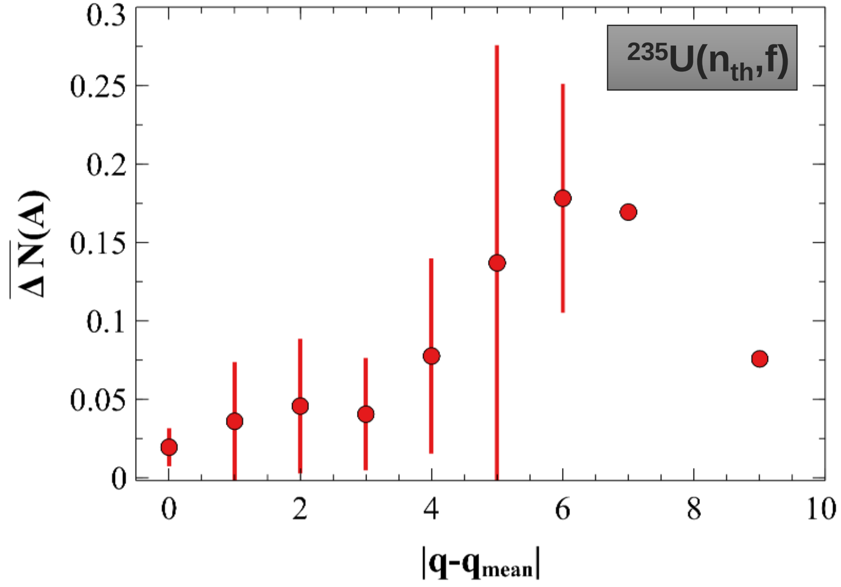
How to analyze such amount of data

~ 350 scans → ~ **7000 points (300 h of beam time)**
~ 15 steps to go from count rate to absolute fission mass yields → uncertainty propagation complex
→ Use of Bootstrap technique : sample count rates and "reroll the experiment"

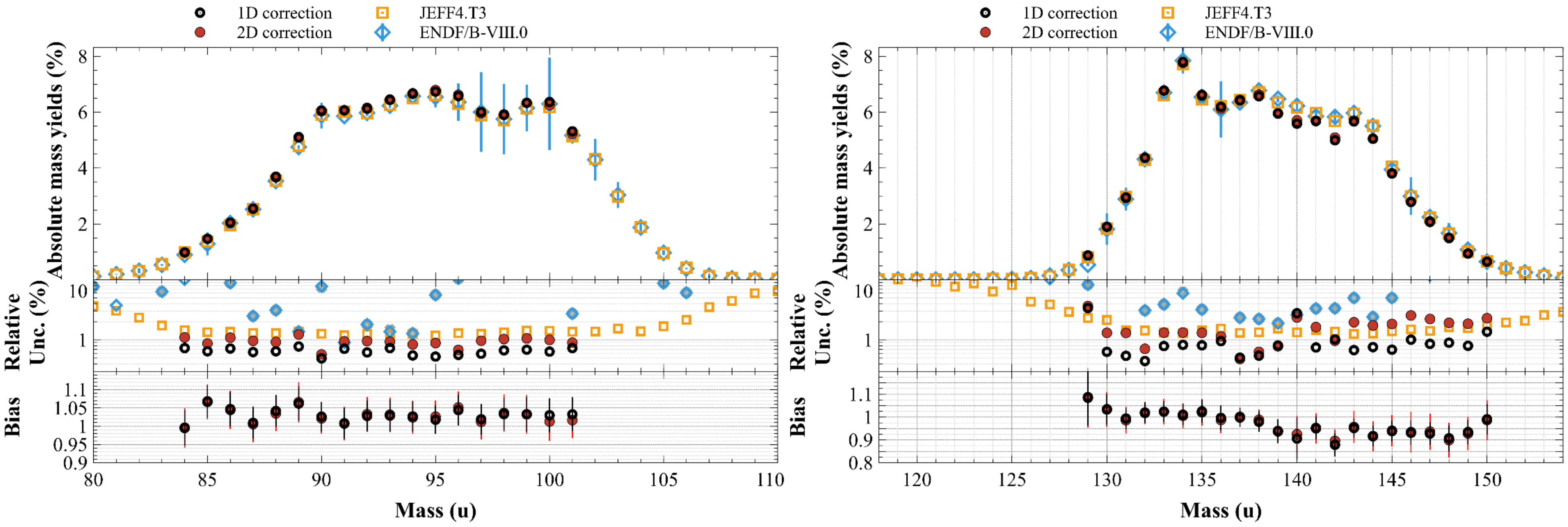


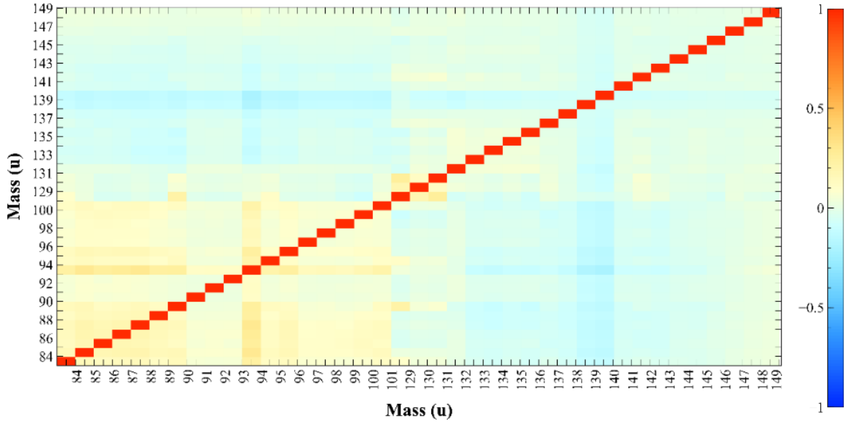


^{233}U,^{241}Pu : uncertainty around 2-3%



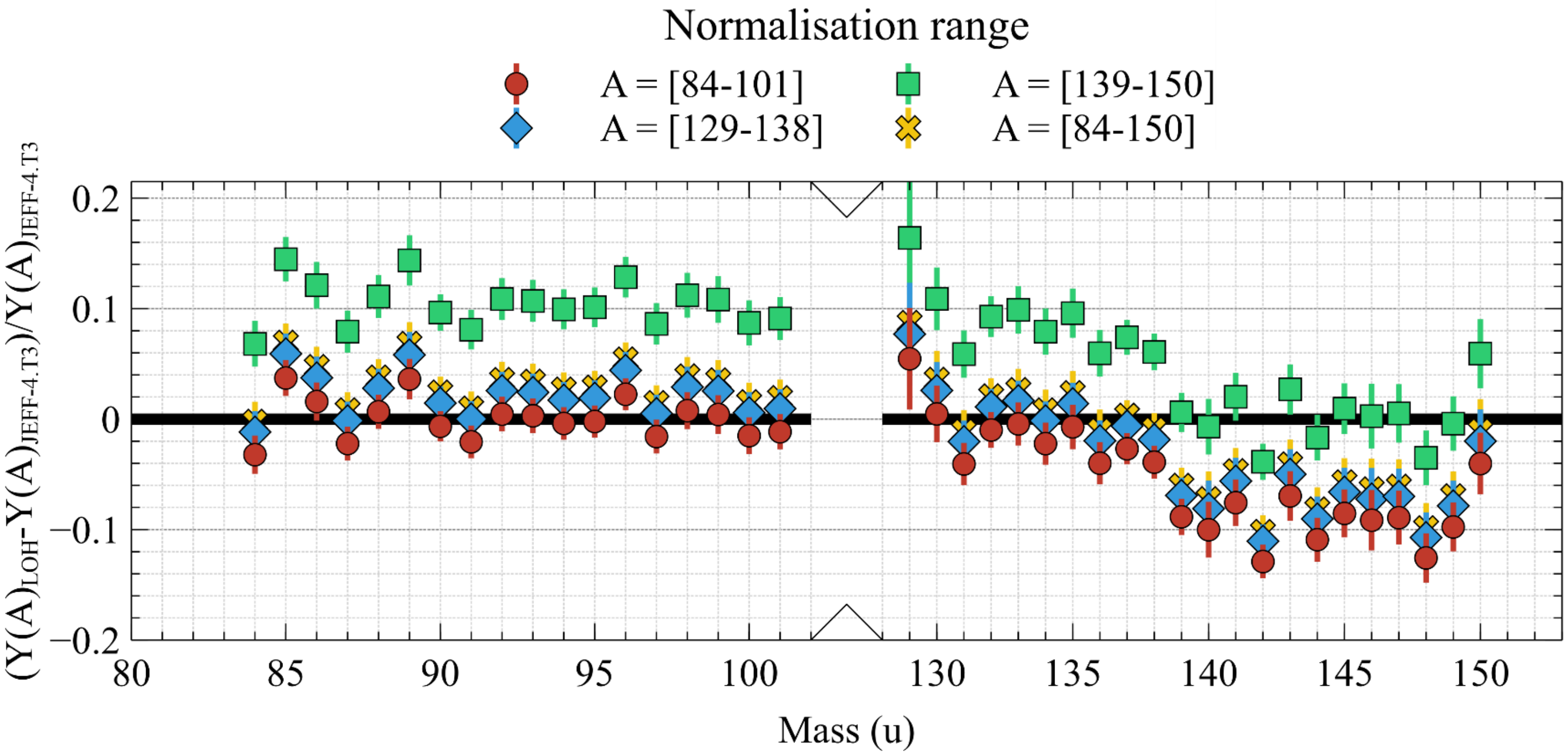
Results





- Heavy peak : ☑
- Reduce uncertainties and handle bias : ☑ Starting with **uncertainties around 6-10%** (^{235}U + ^{241}Am + ^{239}Pu) → 10 years of efforts to reduce uncertainty around **3-5%** (^{233}U + ^{239,241}Pu) and now around **1-3%** (^{235}U)
- Self normalization : ongoing

Impact of the normalization and comparison with JEFF-4.T3

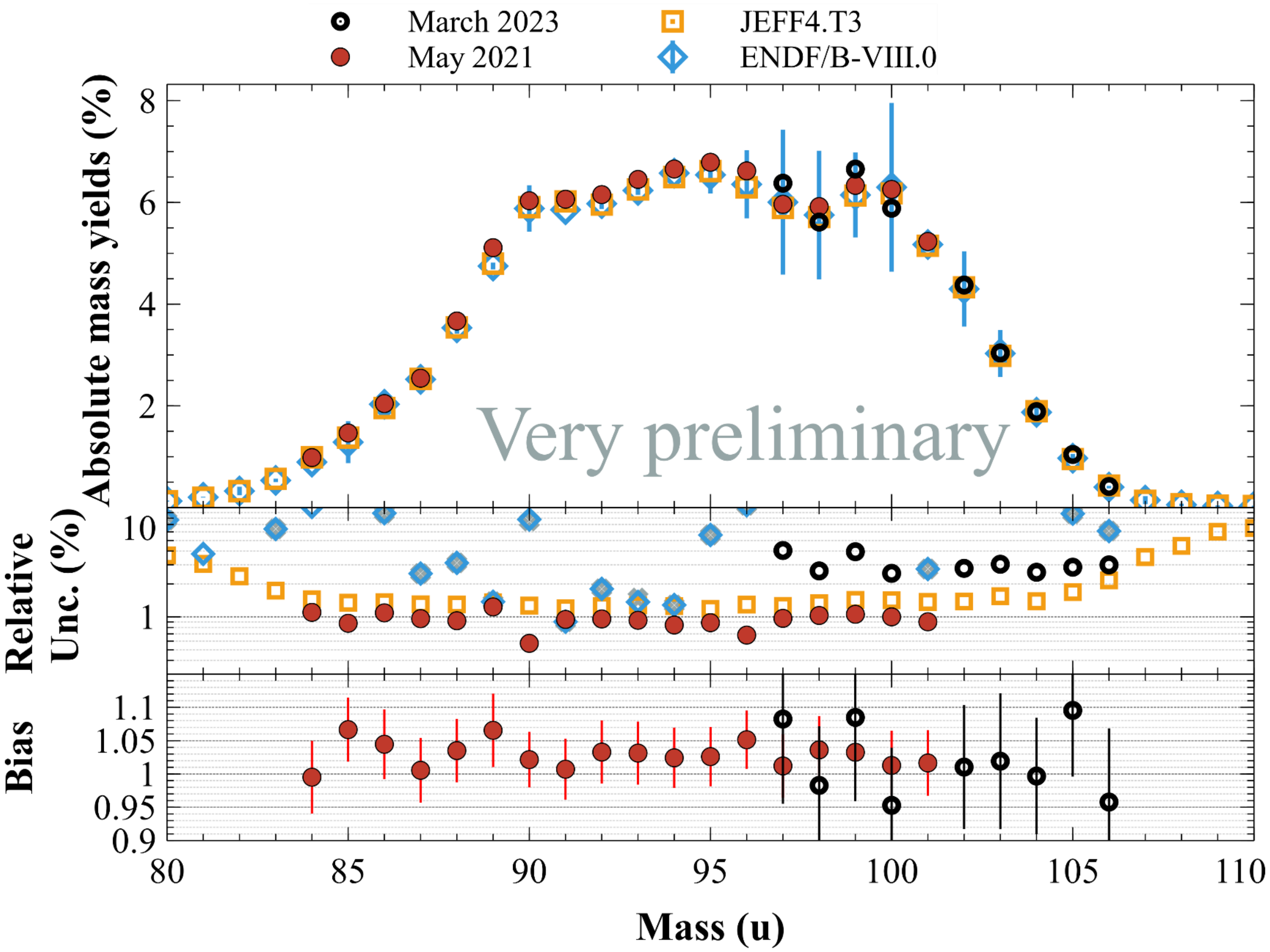


$$Y(A) = k \times N(A) = \left(\frac{\sum_{A'} Y(A')_{JEFF}}{\sum_{A'} N(A')}\right) \times N(A)$$

Test of compatibility at 3 σ		Normalisation range			
$\chi^2(\alpha = 0.997)$	Chi2 range	84-101	129-138	139-150	84-150
37.7	84-101	23.2	29.5	155.5	70.5
25.3	129-138	18.7	10.0	100.9	12.4
28.5	139-150	179.6	130.2	14.4	147.3
68.1	84-150	225.0	186.7	264.2	178.3

- None normalization range can provide a total agreement
- Turns out the “nanosecond” range seems biased → more investigation ongoing
- Nonetheless, good agreement are found for other regions !

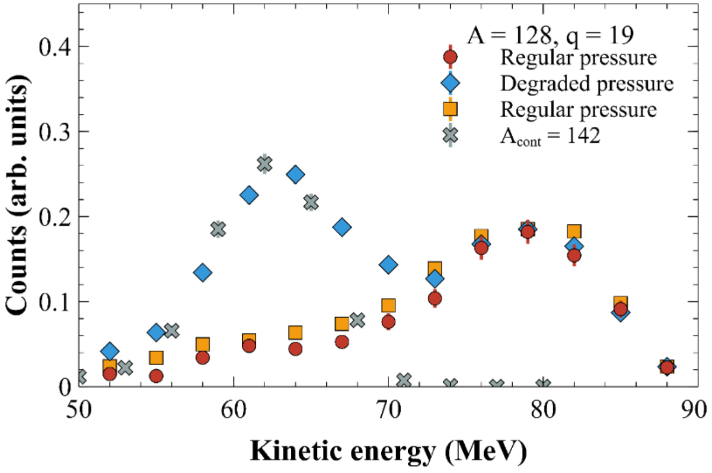
March 2023 campaign

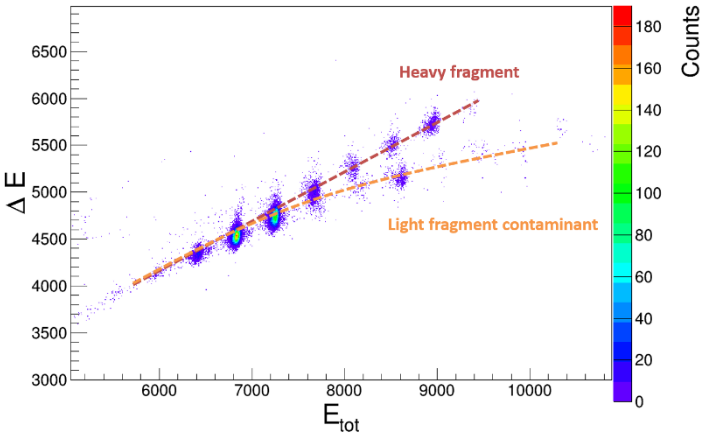


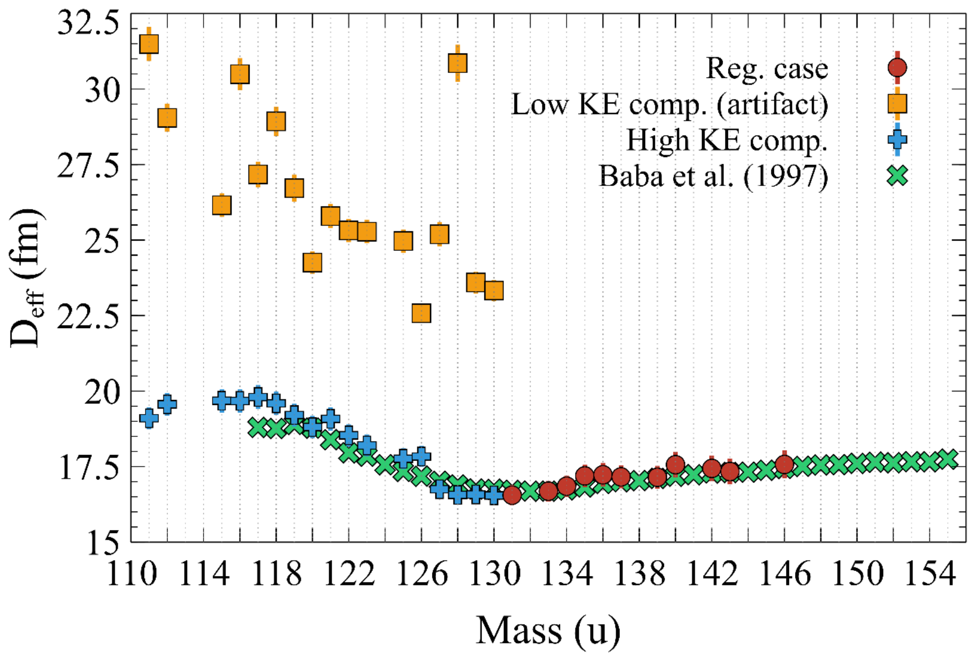
- 1900 files recorded in 5 days
- Very preliminary
- Descent to symmetry seems ok
- Higher uncertainty may come from LOHENGRIN instability

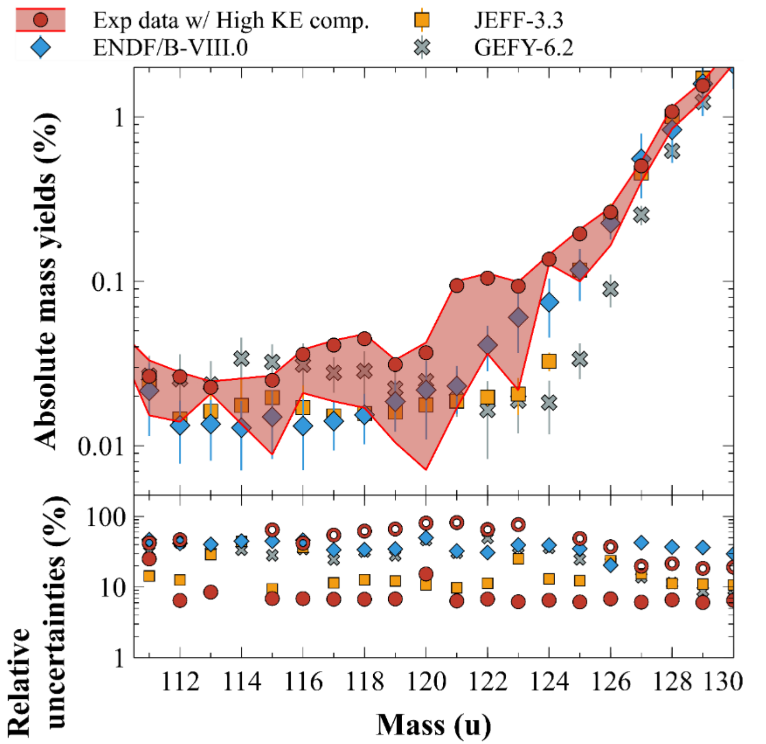
LOHENGRIN limits: symmetry mass region

- Contamination due to charge changing between main magnet and electrostatic deflector
- Dedicated analysis to correct the low energy component. Remaining component seems to be an artefact. New setup mandatory in order to correct these KE distributions more precisely









ToF development

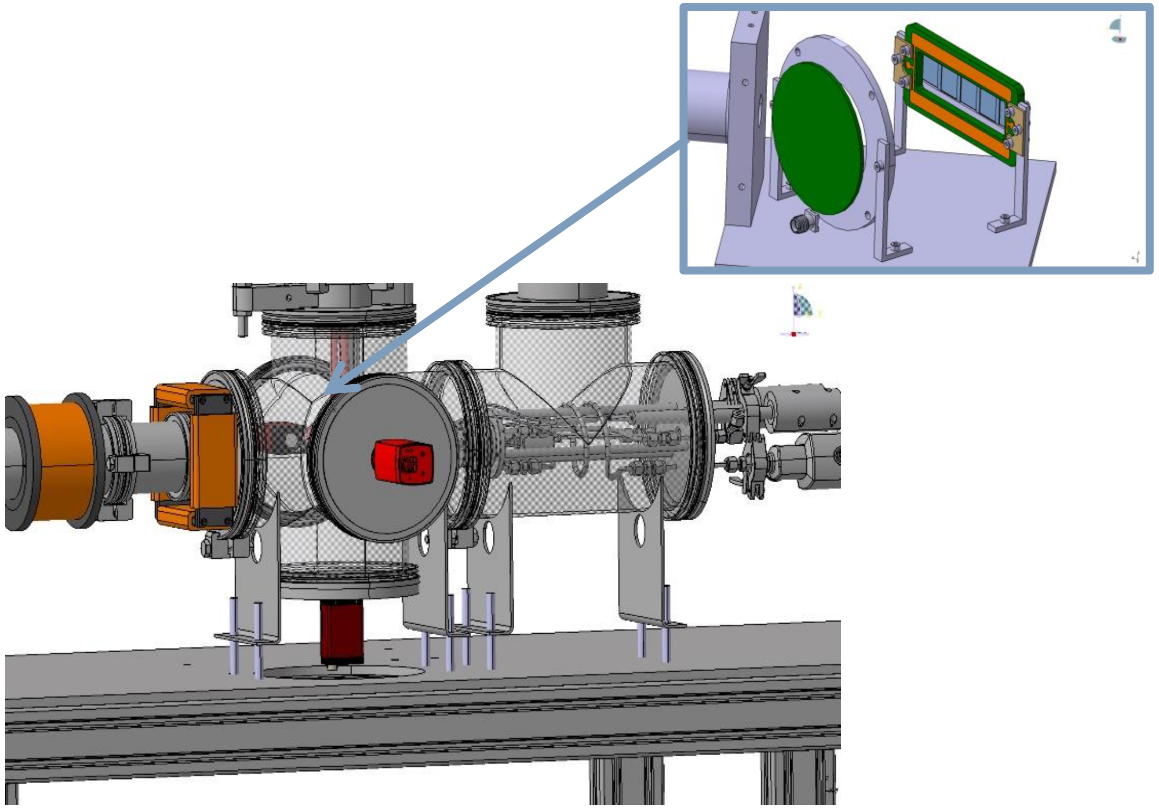
Contaminants have the same energy → measurement of the velocity to disentangle both signals

$$\Delta t = \frac{L}{v} = L \times \sqrt{\frac{m}{2E_k}}$$

Goal: separate contaminants with symmetry mass. $A_{cont} - A_{sym} \cong 20$ u with a distance of $L \cong 60\ cm$
Principle : detect the passage of the fission fragments with the secondary electrons emission from thin foils
Specifications : timing resolution (~ 150 ps) / small energy and angle straggling → SiN foils !

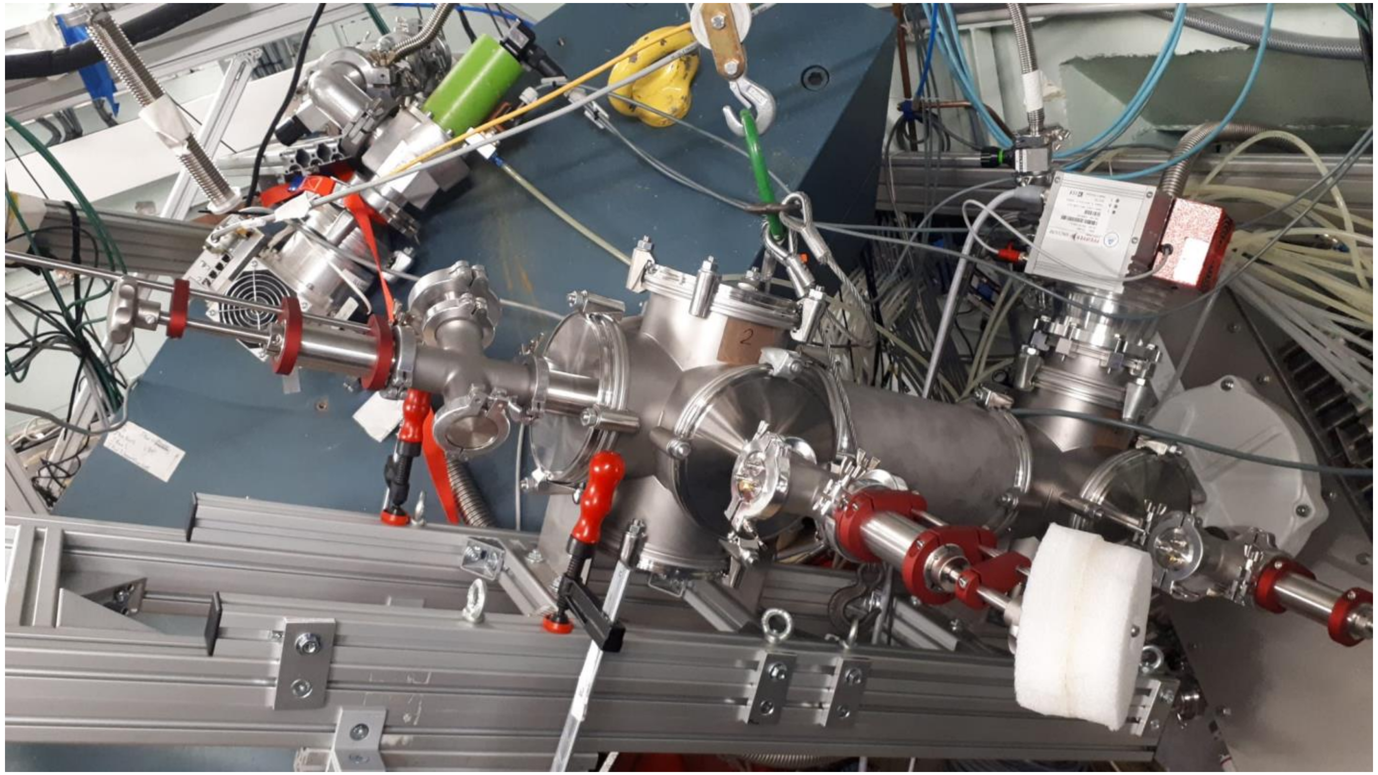
SED	+	-	Results
Channeltron	Work at "high" pressure: 10^{-3} mbar	Efficiency Unknown timing resolution	$\sigma_t = 600\ ps$ Efficiency was ok (FF produces ~ 20 electrons)
SiPM+scintillator	No pressure issue Good time resolution	Need to convert electron to photon Background?	Dark noise too high
MCP	Good time resolution	Work at reduced pressure 10^{-7} mbar	Dedicated ToF line under development. Pressure is stabilized. Tests ongoing

ToF development



PhD thesis of Adrien Vieville (2023-2026) funded by **NACRE** !

December 2023 test



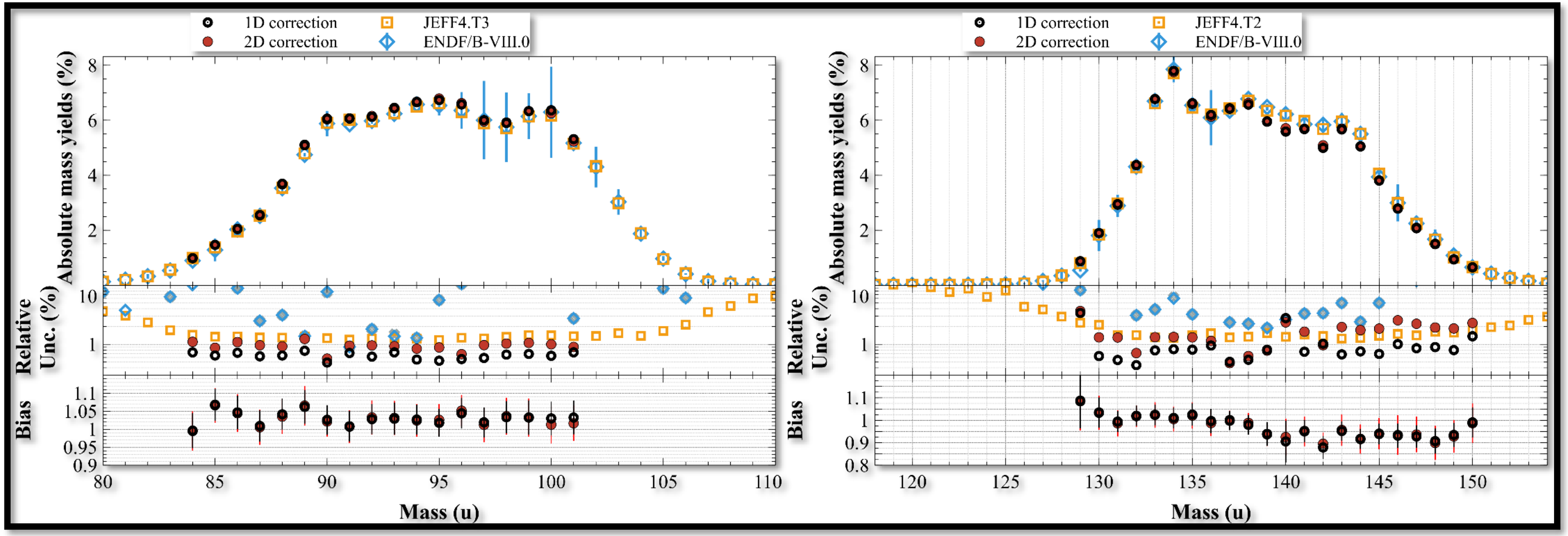
Achievements

- Mechanical coupling
- Pressure <1e-6 mbar
- MCP signals can be read by LOHENGRIN acquisition system

Next steps in 2024

- Data analysis
- New mechanics to have more flexibility to center the beam
- Improve the data acquisition system (MCP are really fast !)
- Angular straggling measurements
- Developments of new geometry (90° vs current 45°)

Conclusion



- Over the past decade, new methodologies were developed in order to improve the accuracy of the measurements
- Analysis of new $^{235}U(n_{th},f)$ mass yields on going
- Underestimation of mass between 139-150 due to nanosecond isomers
- New device to measure symmetry region
- Perform the self normalization
- Measurement of A=153 for $^{239}Pu(n_{th},f)$ reaction in November 2023. Interest for nuclear reactor studies (reactivity loss)

2. Other observables of interest to study the fission process : isomeric ratios

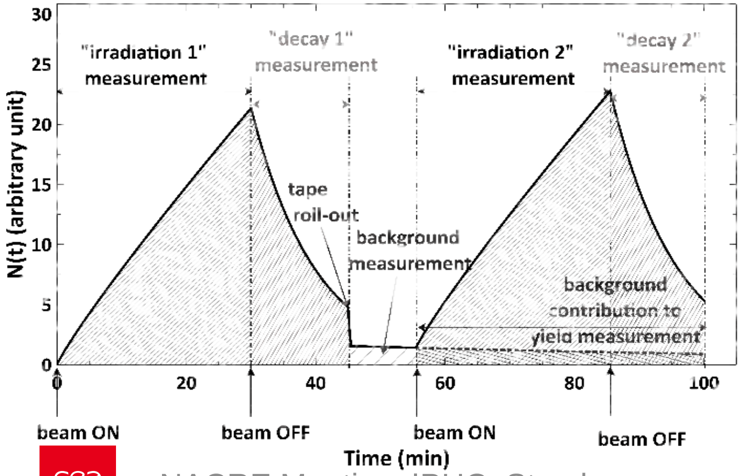
Review of different techniques to assess Isomeric Ratio on LOHENGRIN

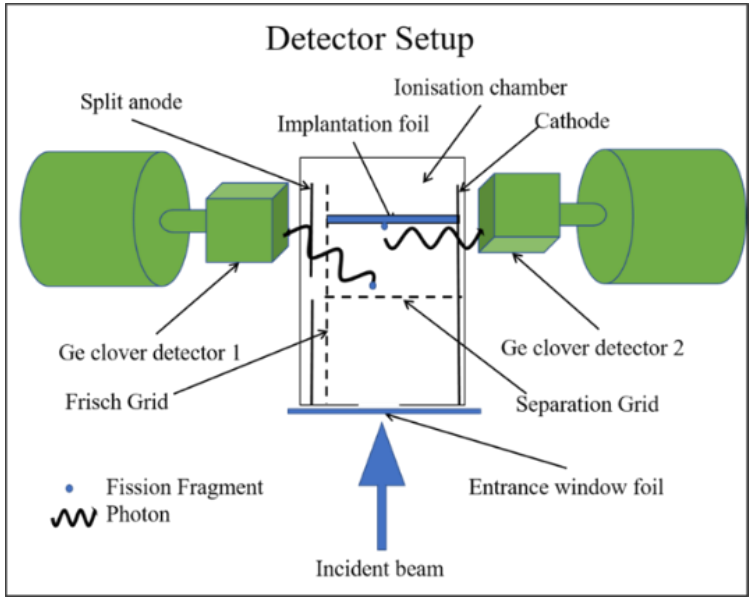
ms to min Isomers

By product of isotopic yield analysis

Difficulty : Some Isomeric states have the same γ lines as ground state

Solution : Measurement of increasing and decreasing count rates of both states





µs Isomers

Difficulty : Isomeric states can be filled by the β decay of the father nuclei

Solution : Coïncidence between ionization chamber and γ detectors

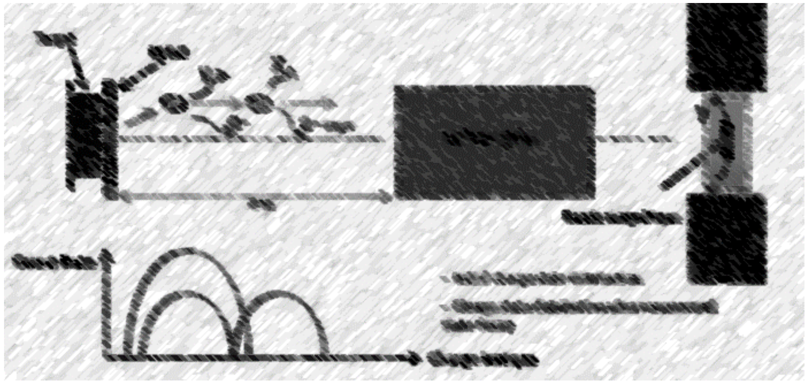
ns Isomers

Difficulty : Isomeric states have a period

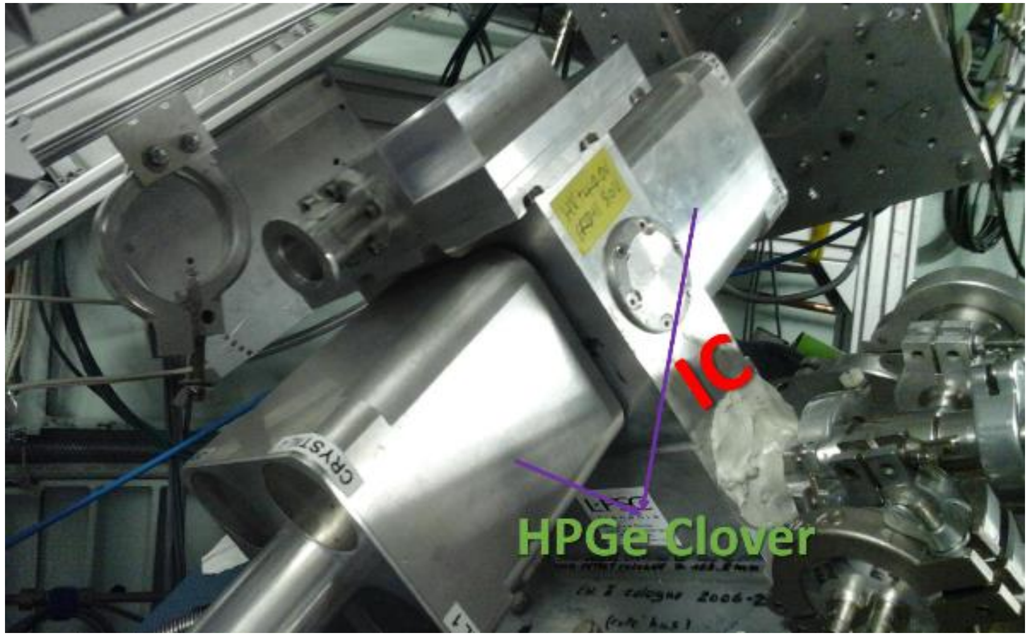
$T_{1/2} \ll tof\ (\sim 2\mu s)$

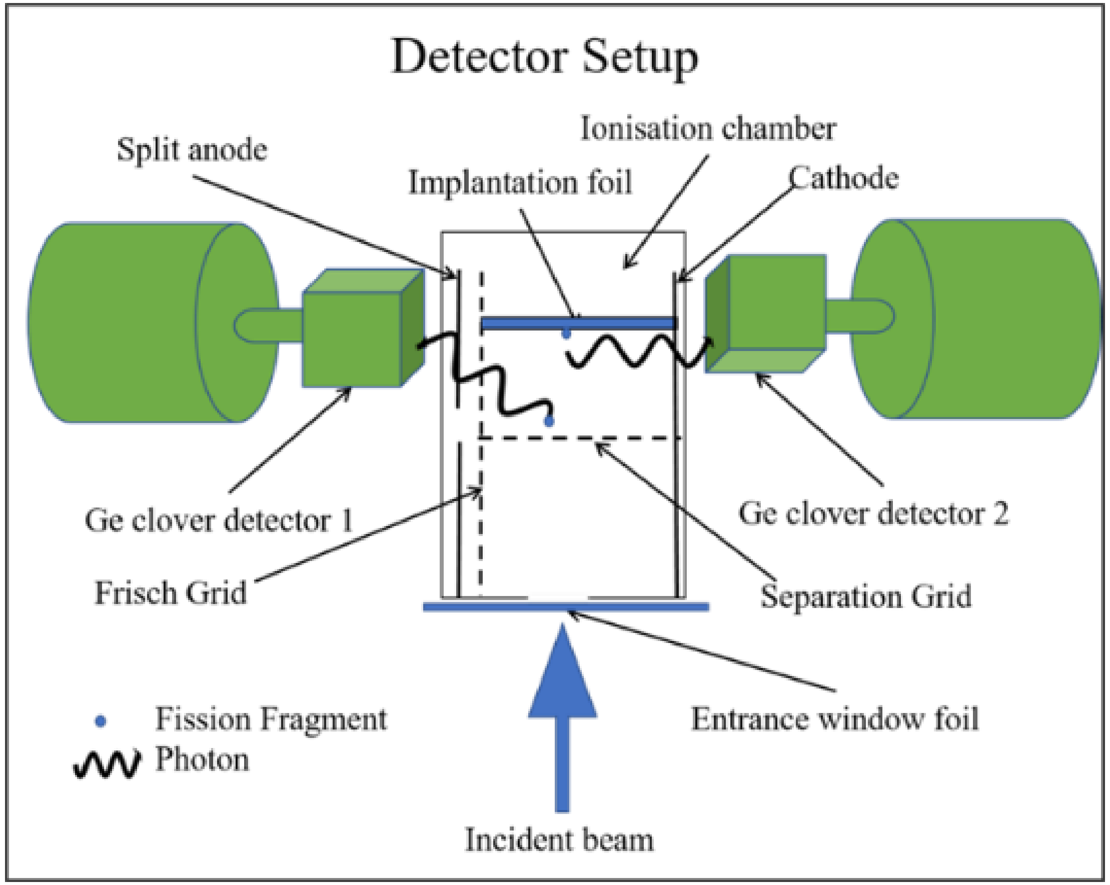
→ no direct measurement

Solution : Statistical analysis of the ionic charge distribution

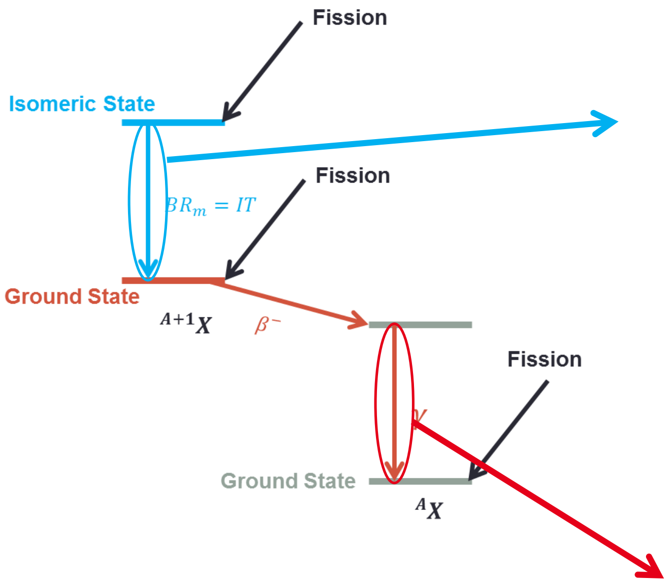


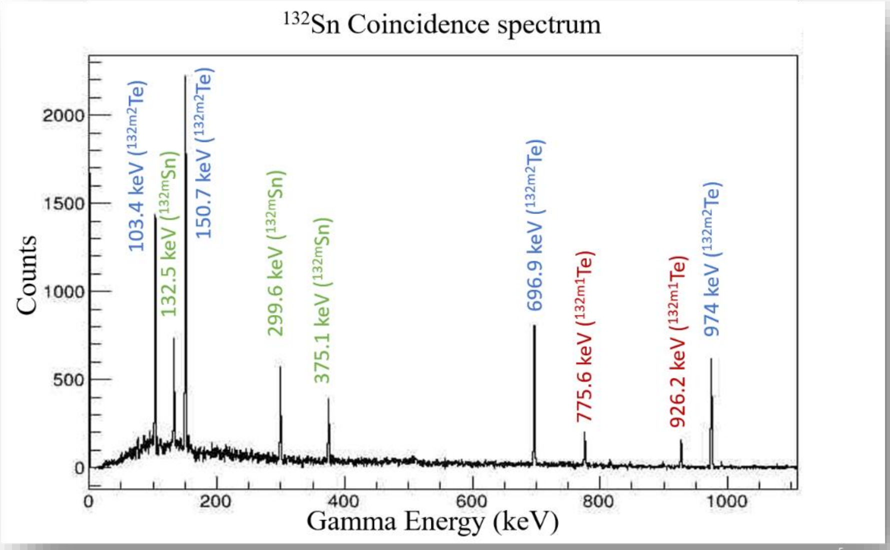
µs isomer experimental setup



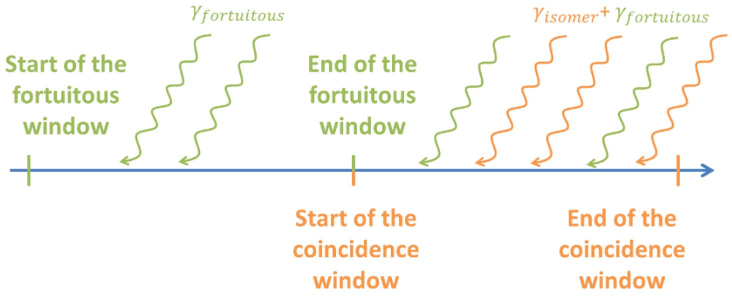


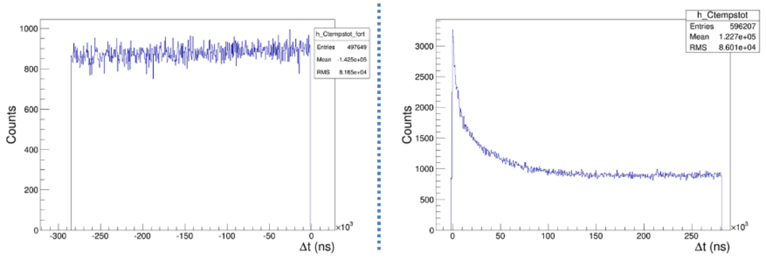
Analysis of µs isomers



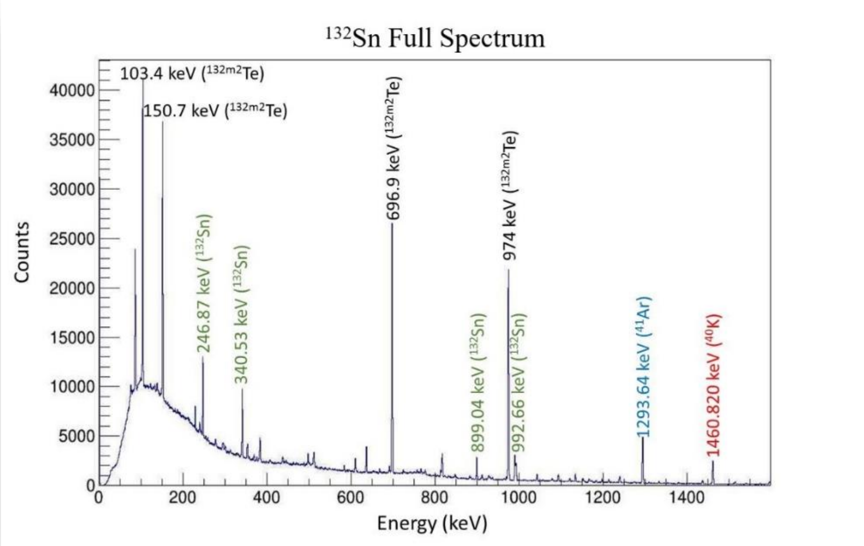


Coincidence between ionization chamber and γ detectors
→ Isomeric state measurement
$\Delta T_{Gate} = 10T_{1/2}$





Ungated γ Spectrum i.e. without coincidence
→ Ground state measurement
→ Extraction more difficult because of the S/B ratio



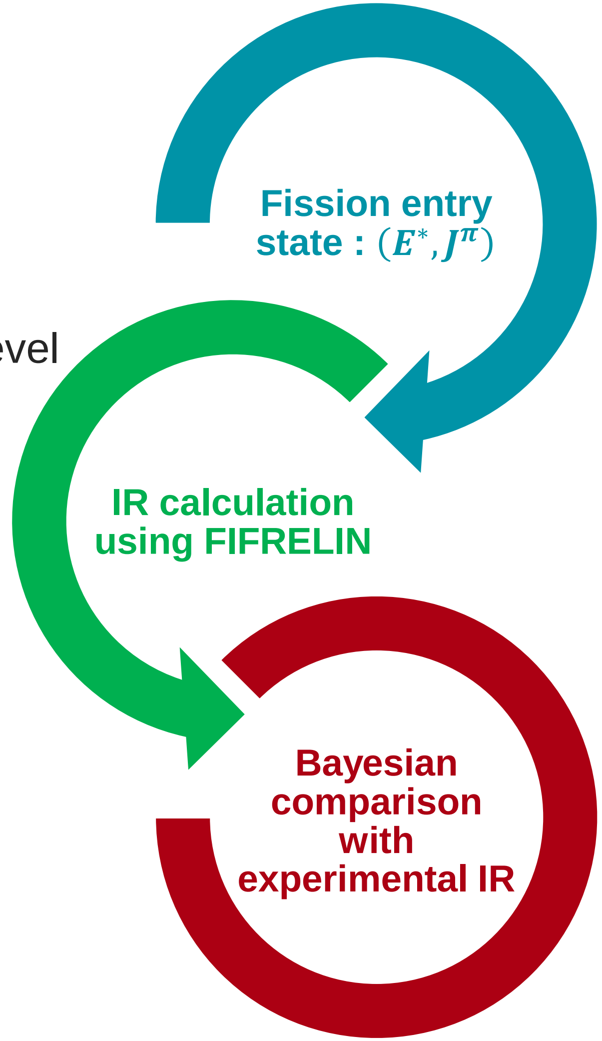
$$IR = \frac{\tau_f(^{132m}Sn)}{\tau_f(^{132m}Sn) + \tau_f(^{132gs}Sn)}$$

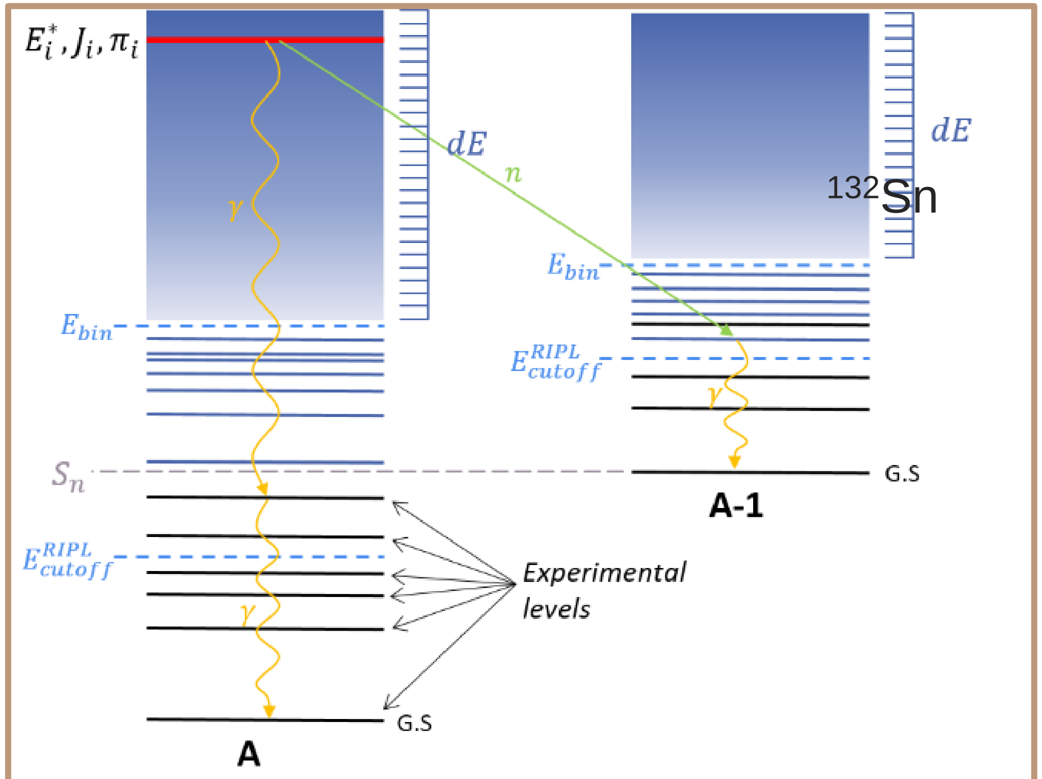
How to assess angular momentum using FIFRELIN

In this work, FIFRELIN (developed by CEA Cadarache) is used **only as a nuclear de-excitation code**

What is required for FIFRELIN :

- experimental level scheme (RIPL-3)
- Model of nuclear density to complete the level scheme (CGCM)
- Model of γ strength function (EGLO)
- Electron conversion coefficients (BrIcc)





O. LITAIZE et al., Eur. Phys. J. A51, 177 (2015)

How to assess angular momentum using FIFRELIN

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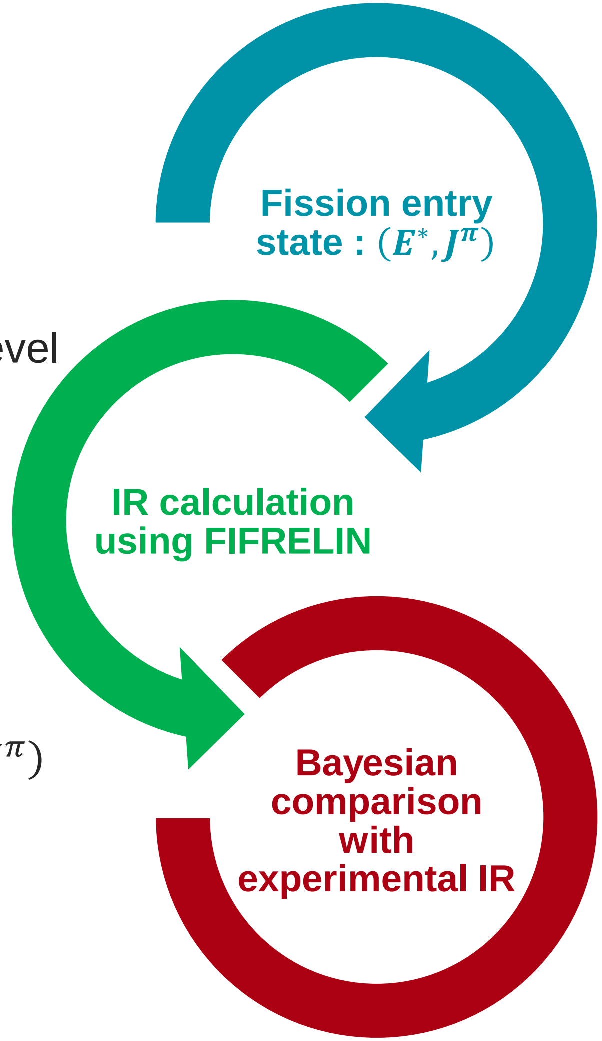
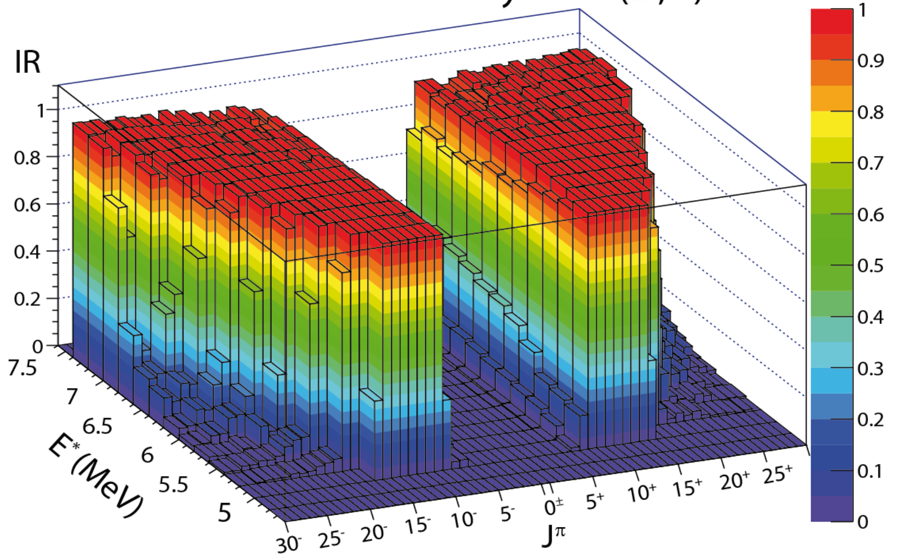
For comparison with experimental results standard spin distribution:

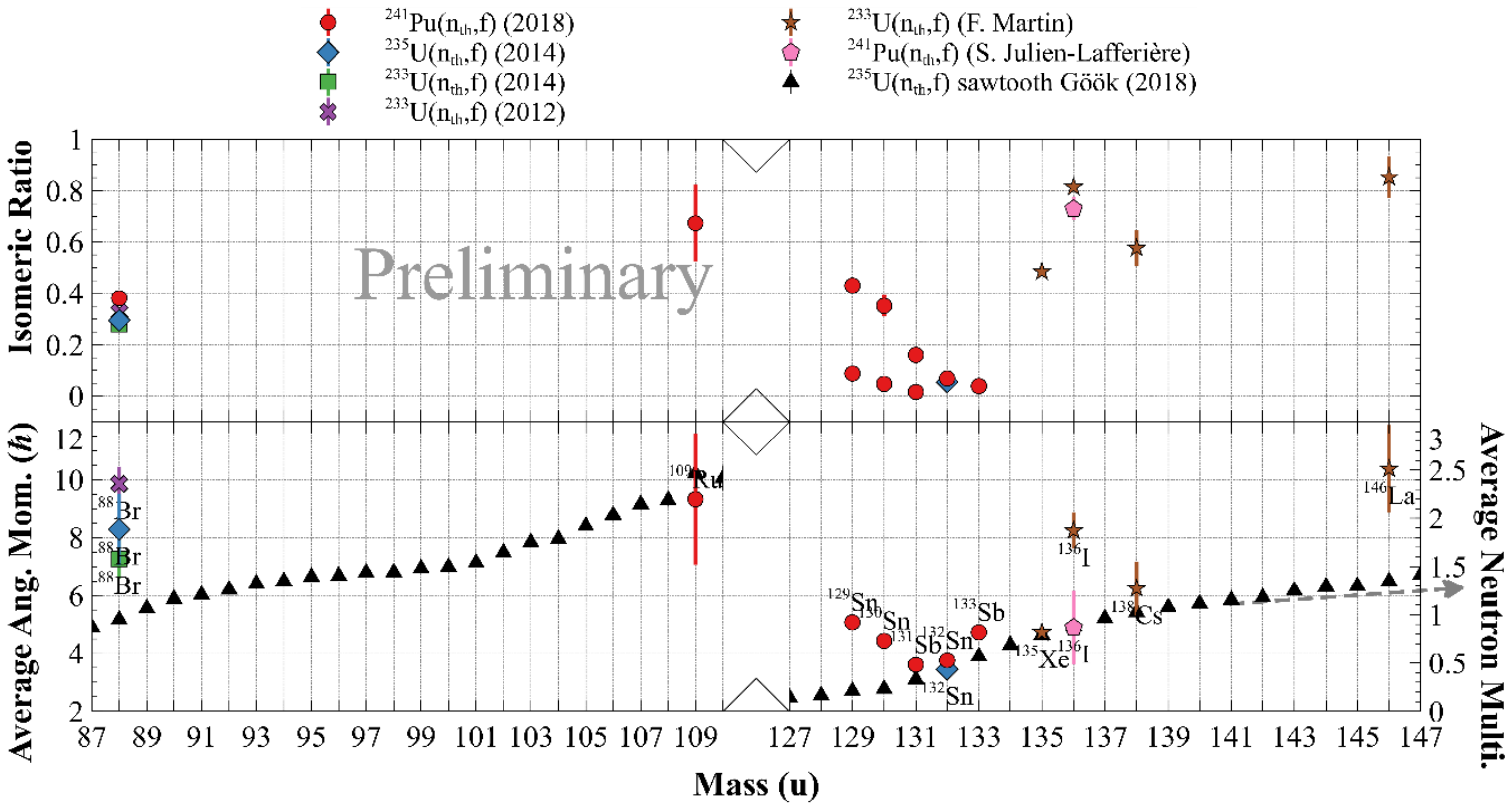
- $IR_{FIF}(E^*, J_{RMS}) = \sum_E \sum_\pi P(\pi) P(J) IR_{FIF}(E^*, J^\pi)$
- $P(J) \propto (2J+1) \exp\left(-\frac{\left(J+\frac{1}{2}\right)^2}{J^2_{cutoff}}\right)$
- $P(\pi) = \frac{1}{2}$



Isomeric Ratio calculated by FIFRELIN as a function of entry state (E^*,J^π) ^{132}Sn

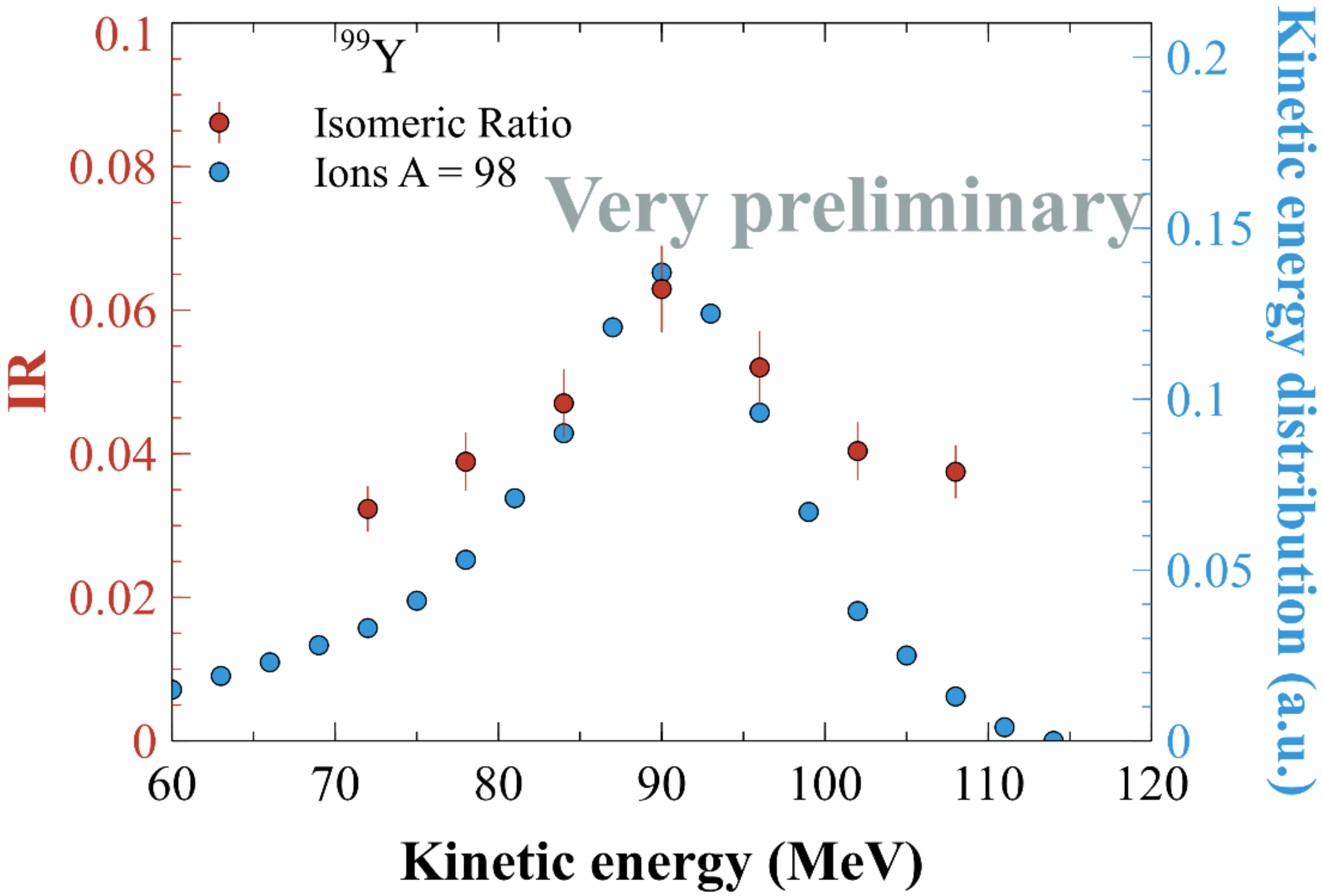


November 2023 campaign: motivations



Goals : complete measurements in the light fragment mass region

November 2023 campaign: first results with ^{239}Pu target



Isotope	Kinetic Energy	Si measurement
^{88}Br	90	
^{94}Y	72/78/84/90/96/102/108	×
^{95}Y	72/78/84/90/96/102/108	×
^{99}Y	72/78/84/90/96/102/108	×
^{100}Nb	72/78/84/90/96/102/108	×
^{133}Sb	60/66/72/80/86/90	×
^{138}I	66/72/80/86	

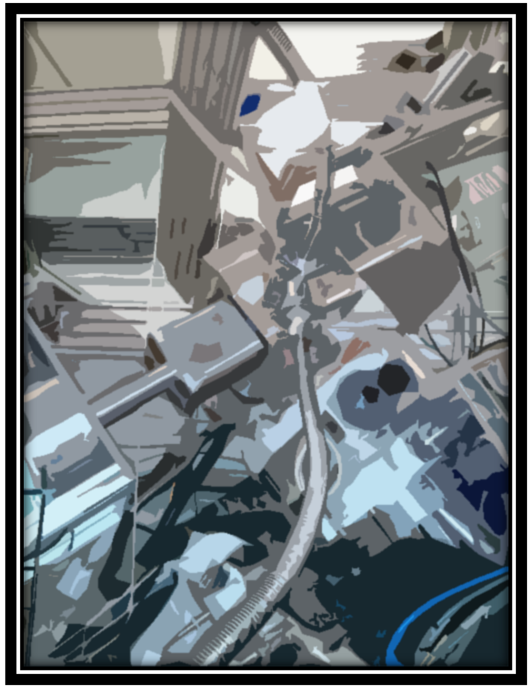
Quite successful campaign despite some difficulties with LOHENGRIN

Will be part of a thesis for 2025

Upcoming measurements

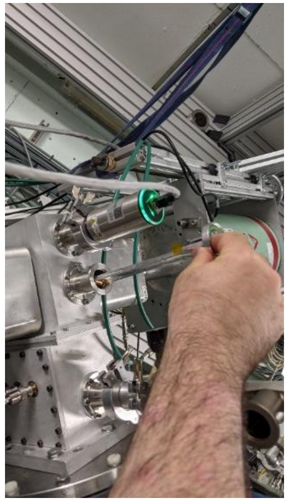
- TOF development
  - Symmetry and far asymmetry region measurements (^{235}U, ^{245}Cm …)
- IR measurements for the light fission fragment mass region for ^{241}Pu (March 2024)
- High resolution Kinetic Energy distribution : indirect measurement of neutron multiplicity, excitation energy …
- Independent fission yields (^{239}Pu) with classic or alternative (activation) technique
- Cumulated fission yields with FIPPS

Thank you for your attention

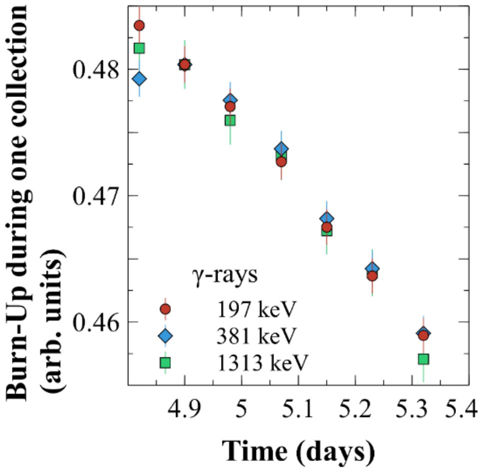


Measurement of long lived isotopes

- Industrials provides us with a list of isotopes which contributes to the total dose rate and to its uncertainty in specific areas of Nuclear Power Plants within accidental conditions.
- Aim : reduce the uncertainty of the associated yields
- Issue : long-lived isotopes
- Solution : new data taking. Already tested on ^{136}Cs



Background phase:
New beam stop aluminum foil is placed inside the vacuum chamber



Implantation phase I
The beam of mass 136 is implanted into the foil during 5-18 hours.

Implantation phase II
The beam of mass 139 is implanted into the foil during 1 hour. ^{139}Ba will be used as a "tracker"

Transfer phase
After 4-6 hours, the residual radioactivity is undetectable with the LOHENGRIN setup or radioprotection probes. The foil is transferred to the **"Laboratoire des basses activités" (LBA)** of LPSC

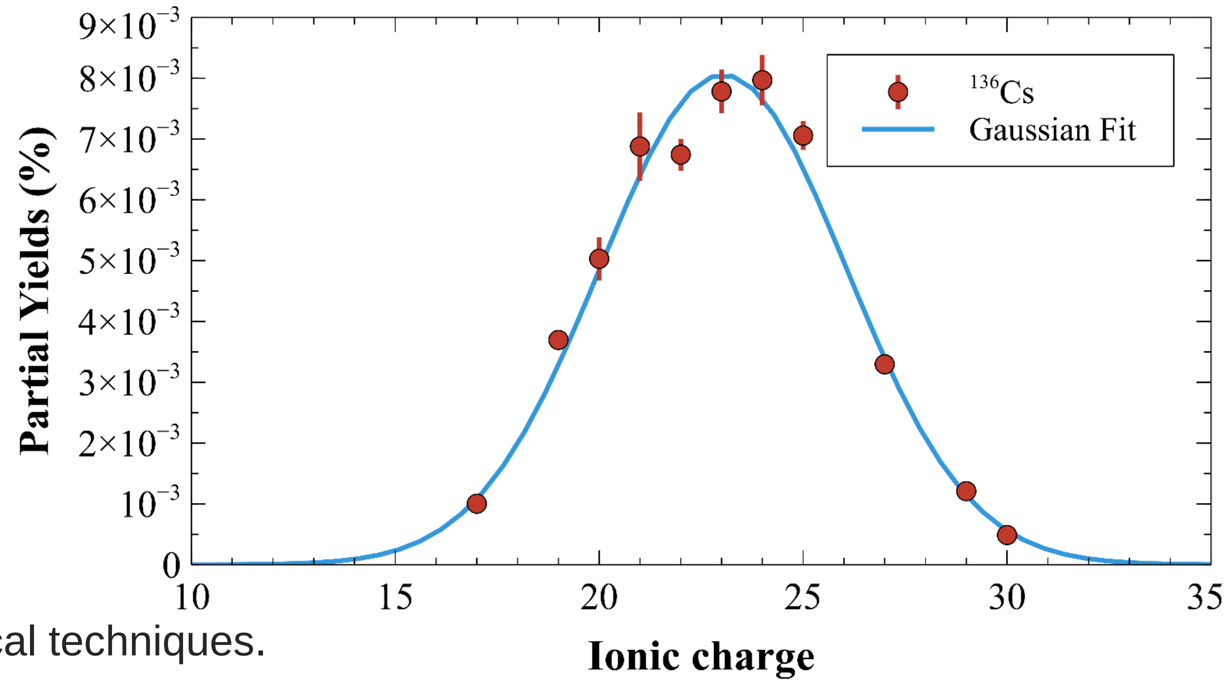
LBA measurement
The decay of ^{136}Cs (and weak residues of ^{139}Ba) is then measured during the next days.

Conclusion

A. Chebbouabi et al., *EPJ Web Of Conf., 284, 08004 (2023)*

- 10 days
- 1600 files
- By definition ^{136}Cs yield is strongly correlated to 139 isobaric chain
- The final uncertainty is mainly coming from normalization factor

^{136}Cs	Yield (%)
JEFF-3.1.1	0.093 ± 0.033
This work (ND)	0.061 ± 0.004
This work (today)	0.059 ± 0.006

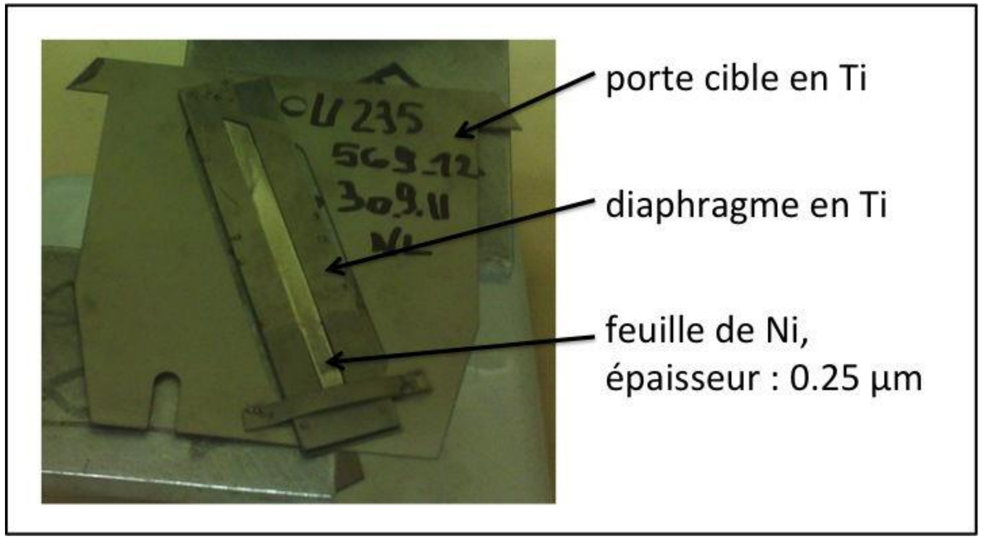


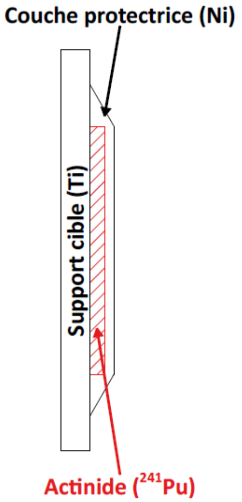
- It is possible to measure inaccessible fission yields by classical techniques.

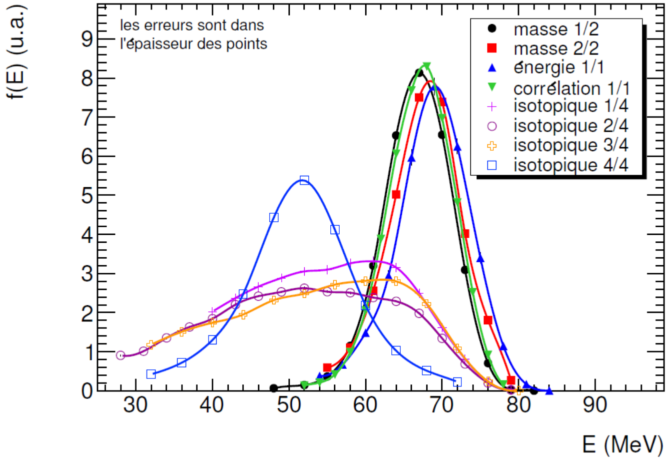
- Combination with a shielded and low background setup of HPGe located at LBA of LPSC permit to achieve such experiments. Preliminary results are promising. **Reduction of uncertainty by factor 5 !**

- A new independent fission yield of ^{136}Cs and its associated uncertainty are expected and may answer to the request of industrial partners on total dose rate calculation in accidental situations. This work will also be an input to the JEFF-4 library.

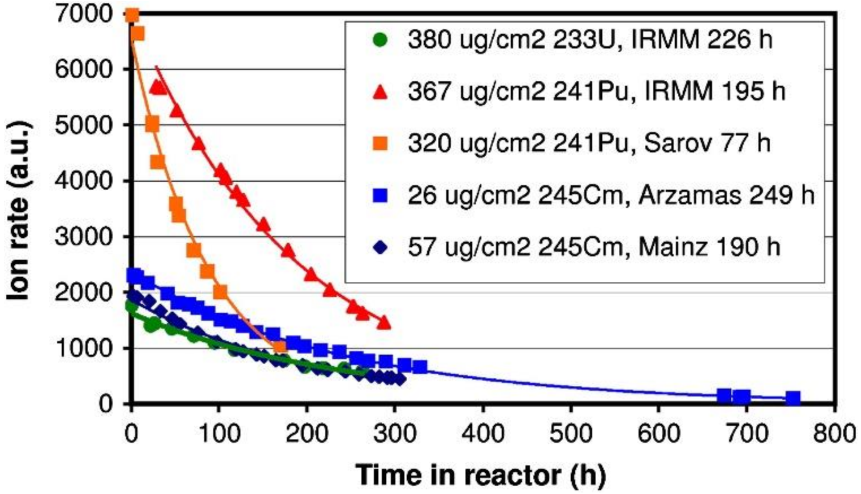
How the target evolves with time

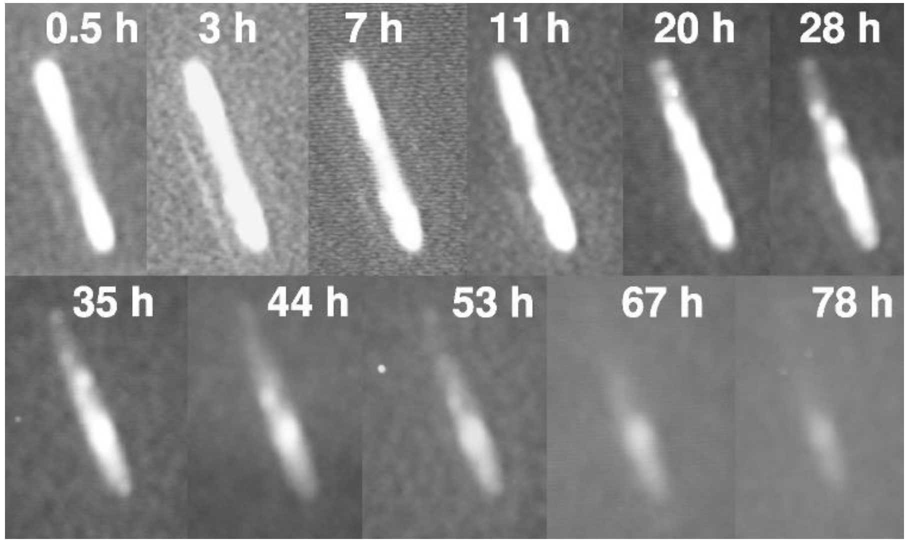




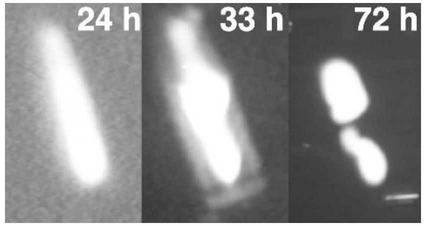


- Legal limit of activity: 3.7 GBq (0.1 Ci) expect for ^{241}Pu and ^{243}Cm : 1 and 2 mg …
- 70 targets over 15 years (6k€/year since 2009)
- Need of spectroscopic quality target





286 µg/cm² ^{241}Pu target



655 µg/cm² ^{239}Pu target