



**HAL**  
open science

## La fission dans tous ses états

Abdelaziz Chebboubi

► **To cite this version:**

Abdelaziz Chebboubi. La fission dans tous ses états. Physique [physics]. Université Aix Marseille, 2024. tel-04591668

**HAL Id: tel-04591668**

**<https://cea.hal.science/tel-04591668>**

Submitted on 29 May 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



# La fission dans tous ses états

Soutenance HDR, 27/05/2024, Aix-en-Provence

A. Chebboubi

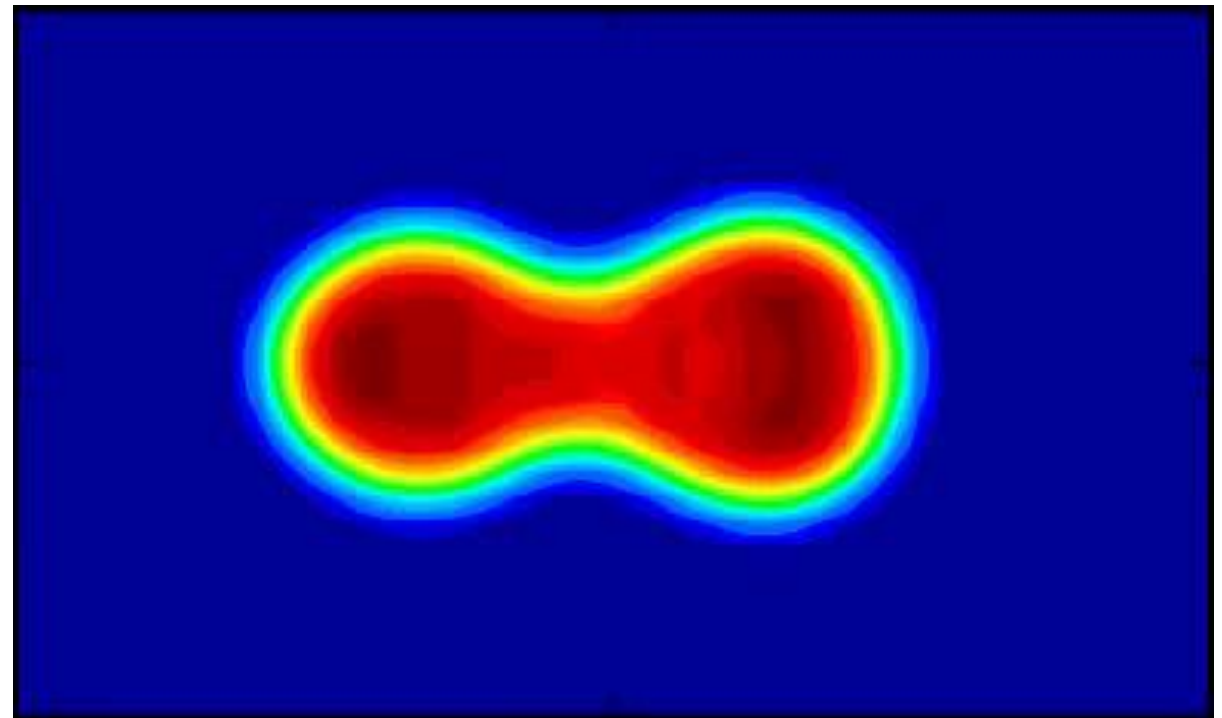
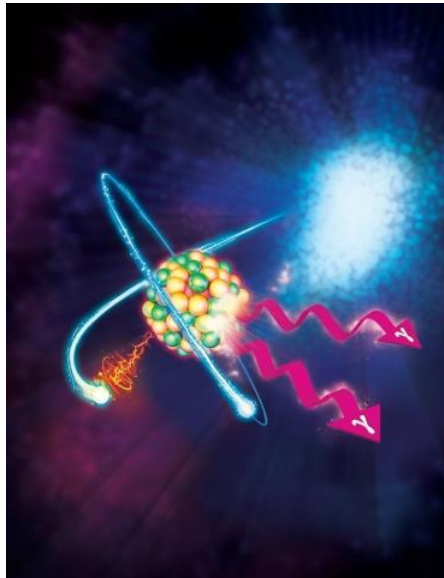
CEA, DES, IRESNE, DER, SPRC, LEPH, Cadarache, 13108 Saint-Paul-lès-Durance



# Discovery of the nuclear fission process

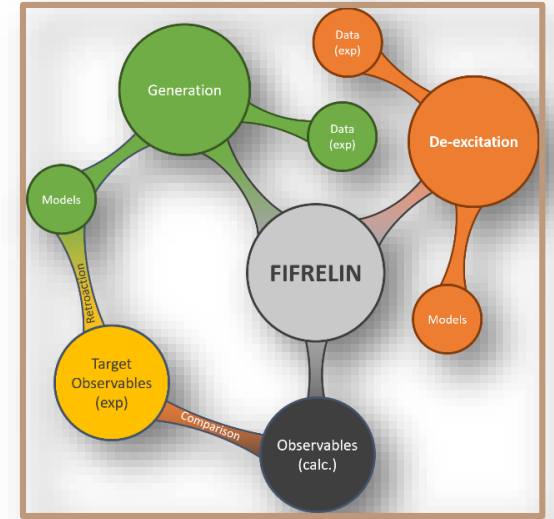
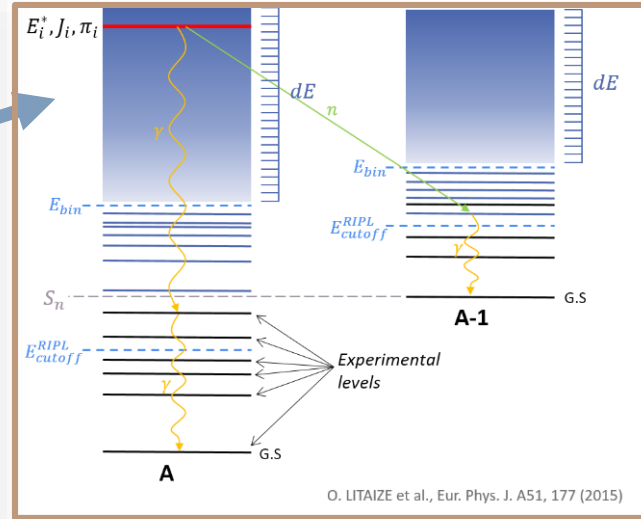
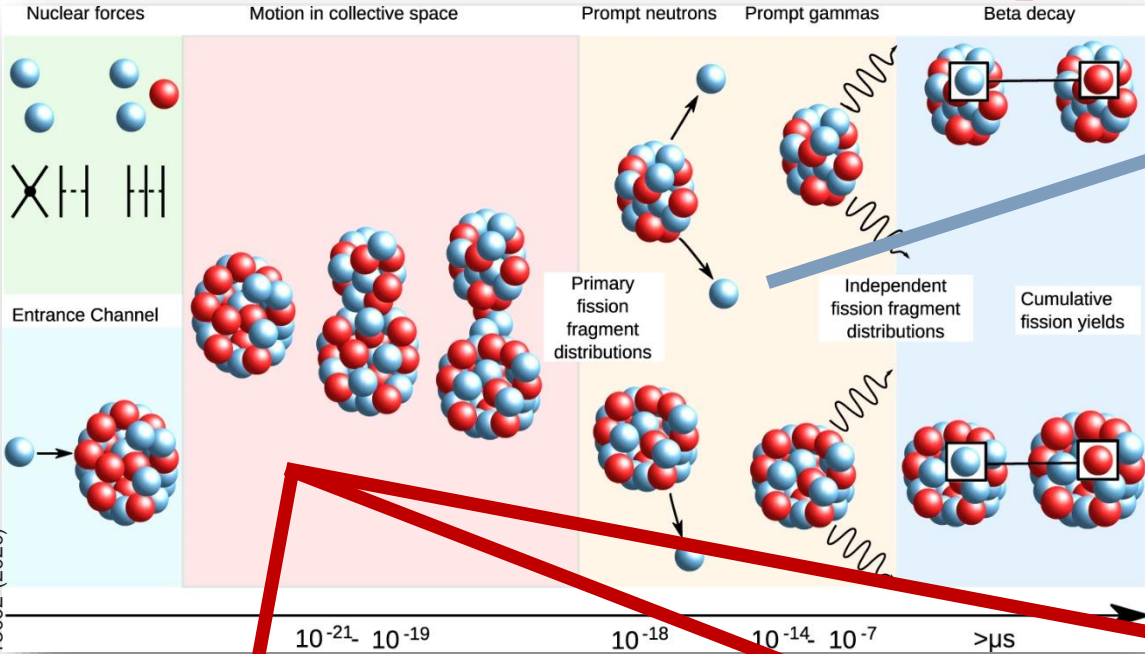


- Discovery of nuclear fission in 1938 by Fritz Strassmann, Lise Meitner et Otto Hahn.
- Chemistry Nobel prize 1944 : Otto Hahn.
- Principle : splitting of heavy nucleus, in general an actinide, into smaller nuclei.
- Secondary particles (neutrons,  $\gamma$ -rays, electrons ...) are emitted.

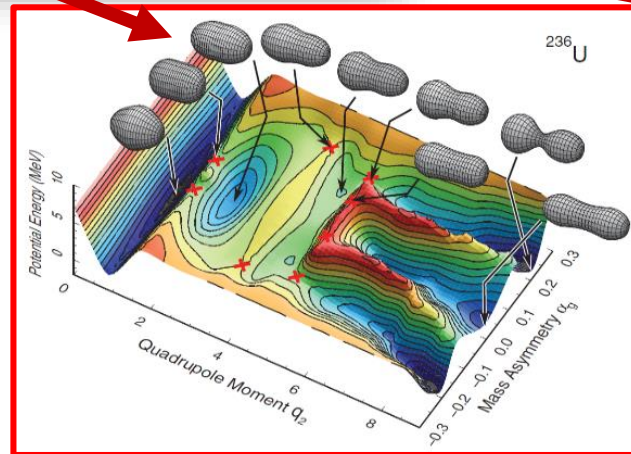
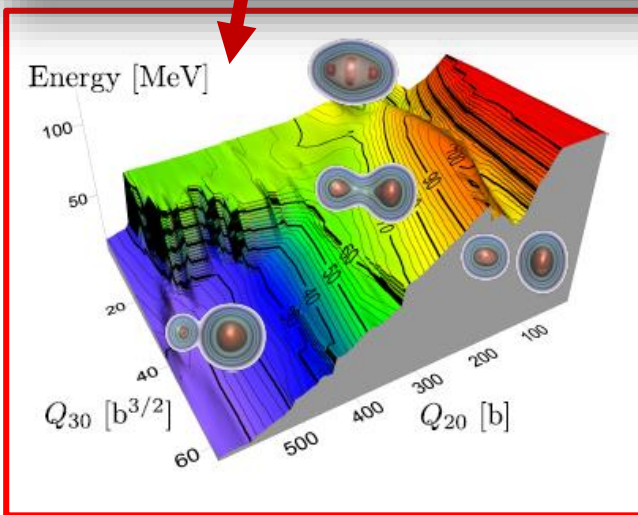


© S. Umar *et al.*, *Density-constrained TDDFT with application to fission*, Quantitative large amplitude shape dynamics: fission and heavy ion fusion, International workshop (2013)

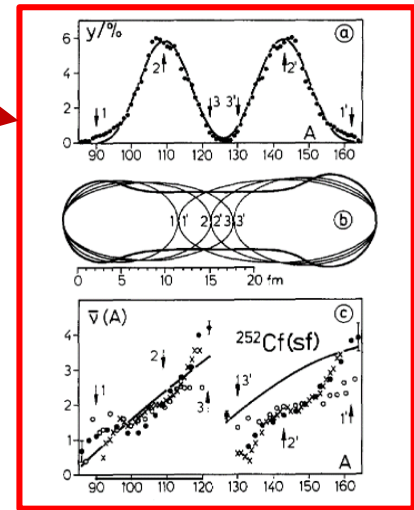
# The nuclear fission process



FIFRELIN : generation and de-excitation of fission fragment. Developed by DES/IRENE



© Ichikawa et al., Contrasting fission potential energy structure of actinides and mercury isotopes, Phys. Rev. C 40, 770 (1989)





# Looking for fission yields

Fission yields = **production rate** of fission fragment for a given mass  $A$ , nuclear charge  $Z$ , excitation energy  $E^*$ , kinetic energy  $E_k$ , angular momentum  $J$ , parity  $\pi$ , and isomeric state  $m$

$$Y(A, Z, E_k, E^*, J^\pi) = Y(A) \times P(Z|A) \times P(E_k|A, Z) \times IR(m|A, Z, E^*, E_k)$$

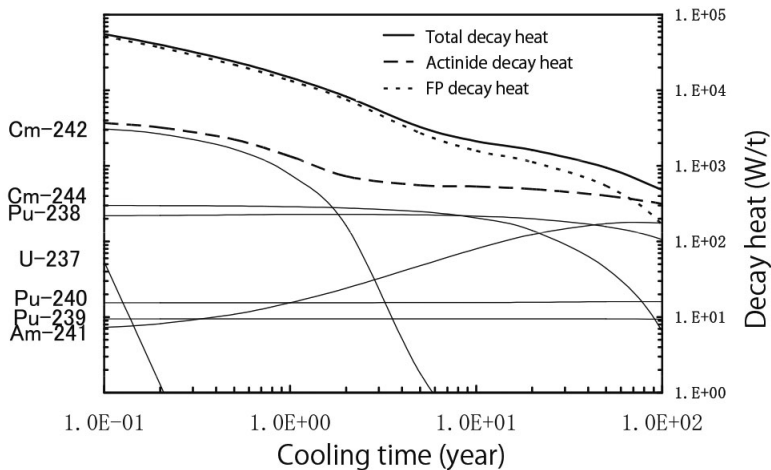
**Independent fission yields**  $Y(A, Z) = Y(A) \times P(Z|A)$  are used in nuclear reactor studies

Isotopic composition

→ Residual power

→ Radiotoxicity of spent fuel

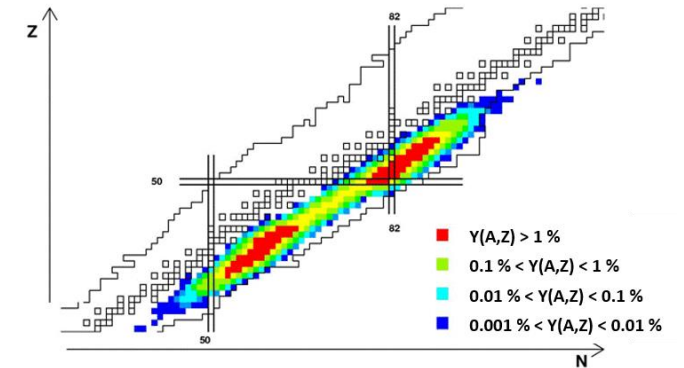
Decay Heat



© T. Yoshida, Atomic Energy Society of Japan, Vol.1.88 (2021)

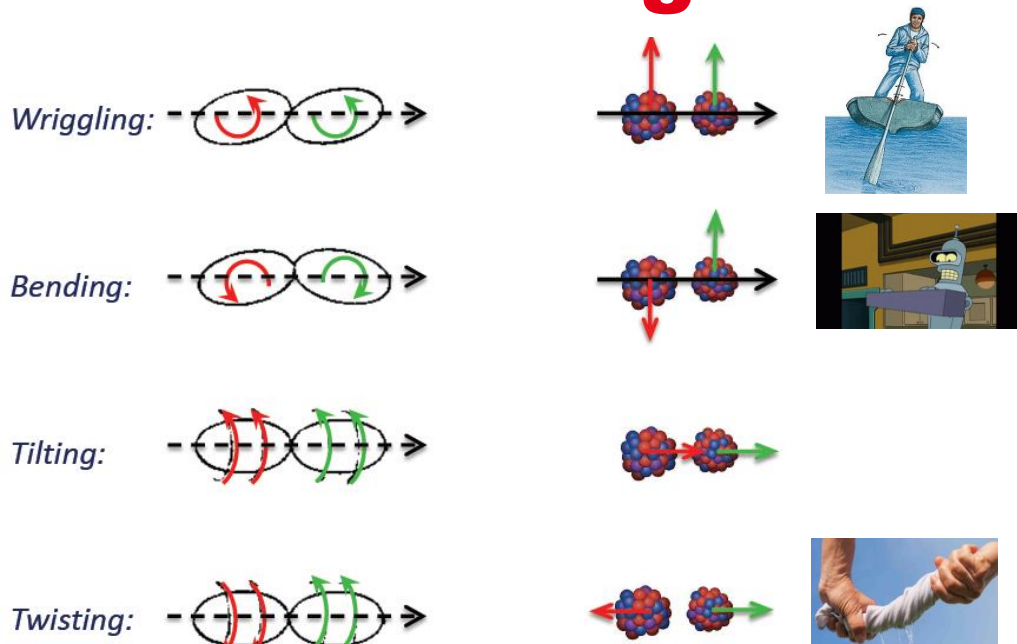


©Orano, La Hague

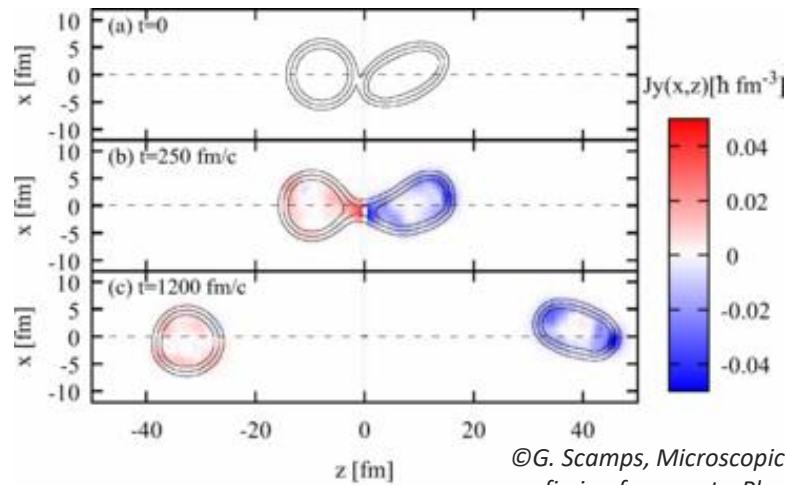
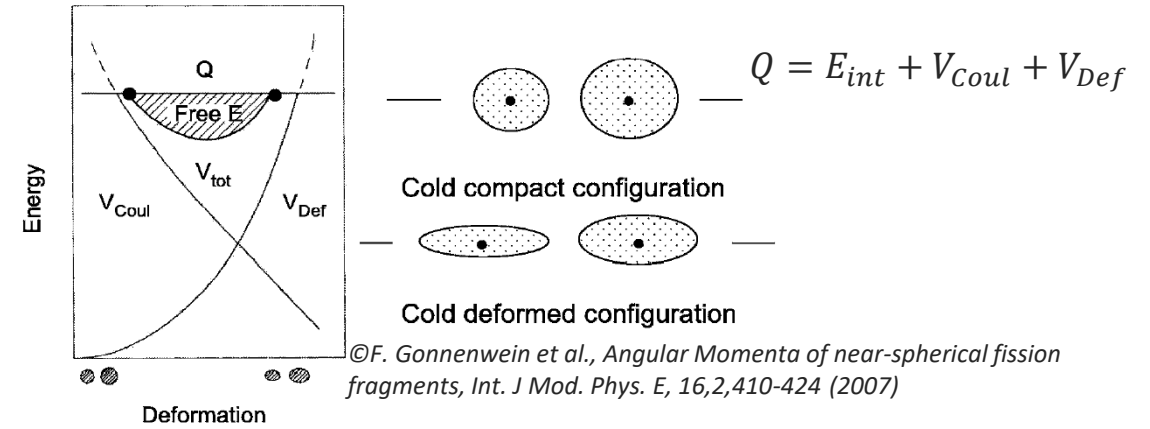


Radioprotection

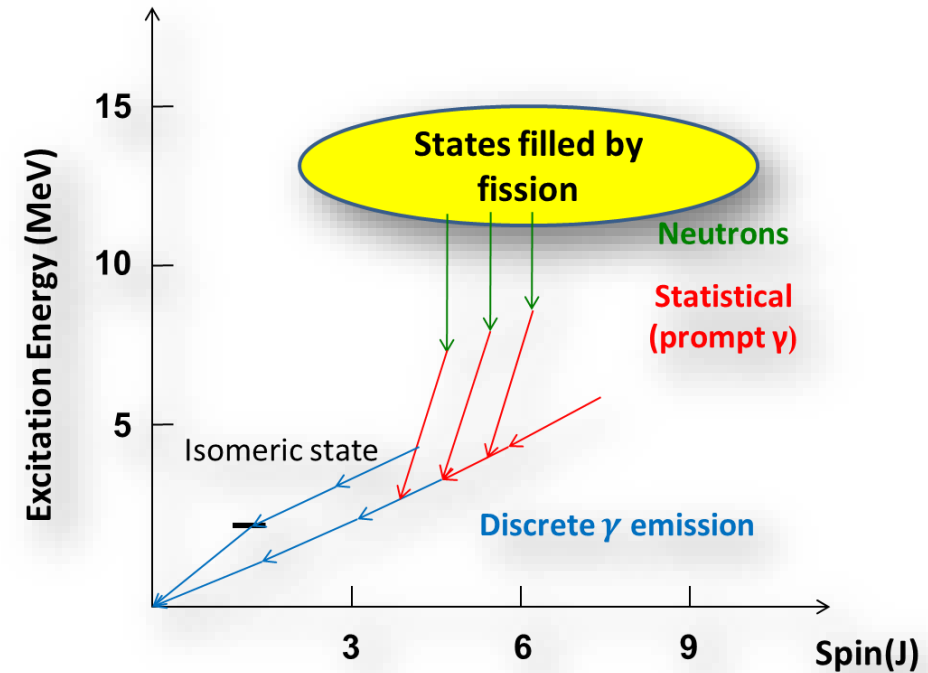
# Focus on angular momentum generation




©J. Randrup et al., Generation of Fragment Angular Momentum in Nuclear Fission, EPJ WoC, 284,04004 (2023)



©G. Scamps, Microscopic description of the torque acting on fission fragments, Phys. Rev. C 106, 054614 (2022)





# 1 ■ Isomeric ratio measurements with the LOHENGRIN spectrometer

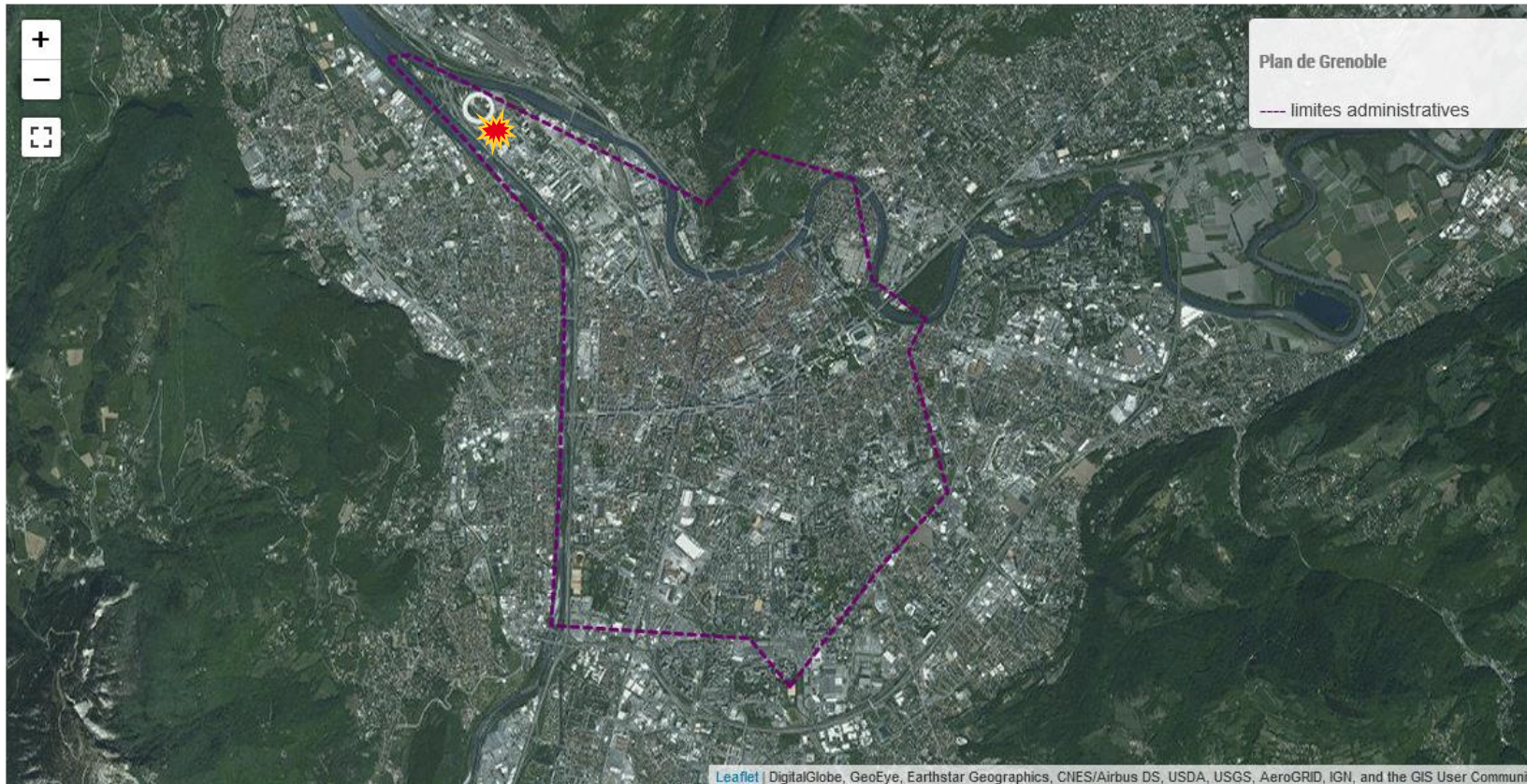
J. Nicholson thesis



# Institut Laue-Langevin



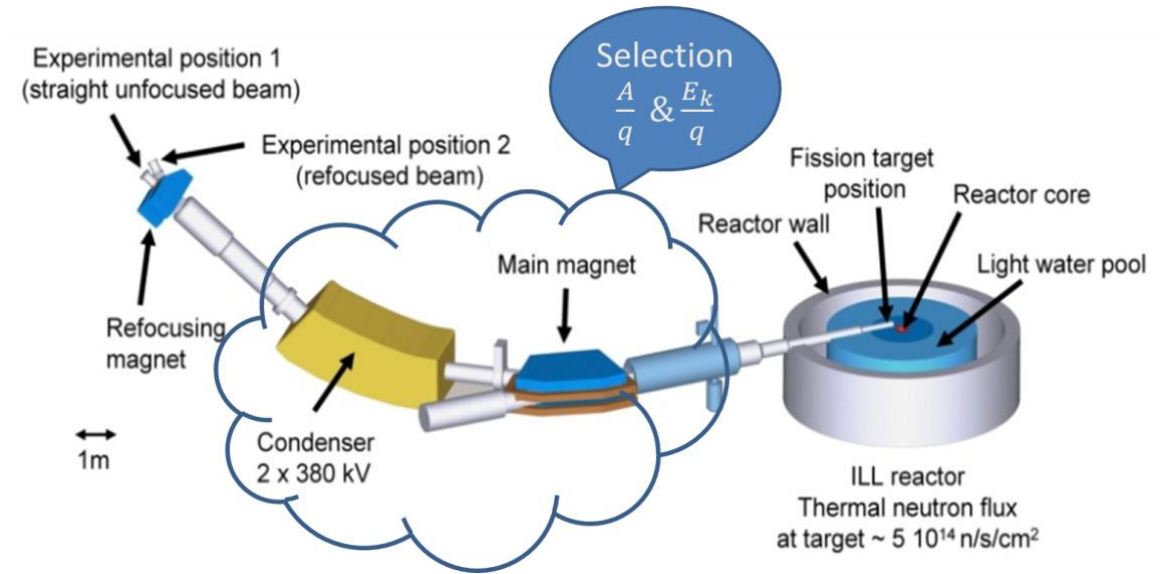
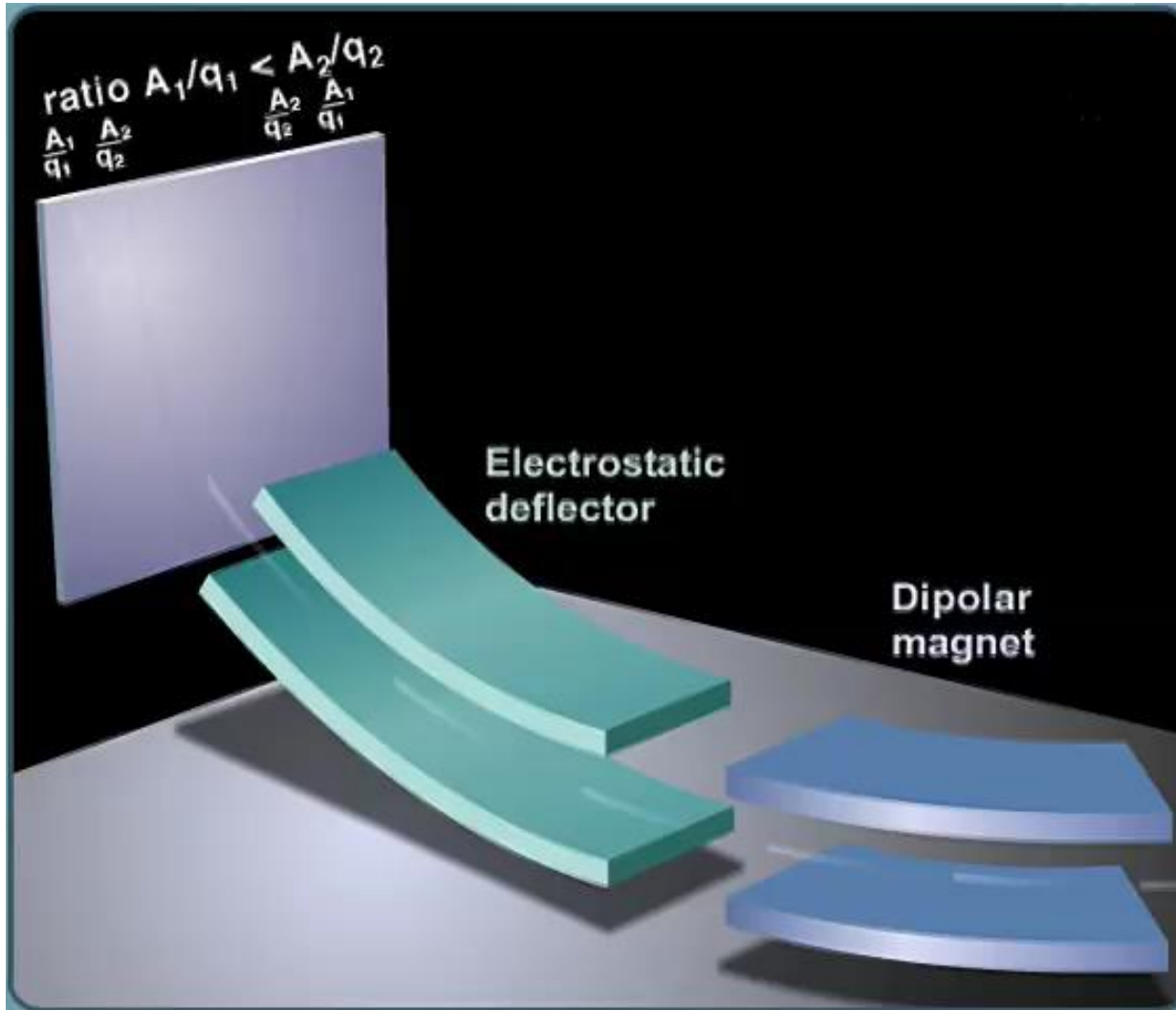
- ✦ ILL : founded and govern by France, Germany and United Kingdom
  - Build in 1967
  - 40 instruments (mainly neutron spectroscopy for biology, materials ...)
  - 540 member staff + 1400 users per year
  - 105 M€ per year
  - High Flux Reactor : 58.3 MW thermal. New vessel in 1995.







# 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

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



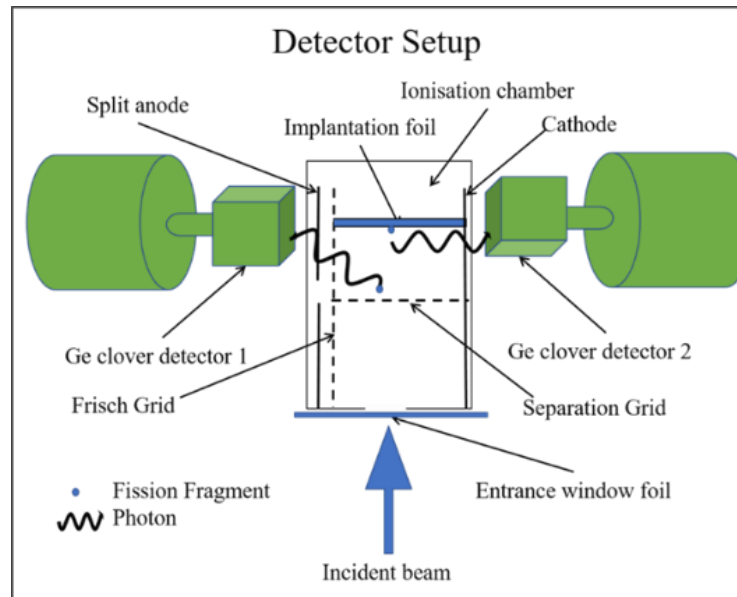
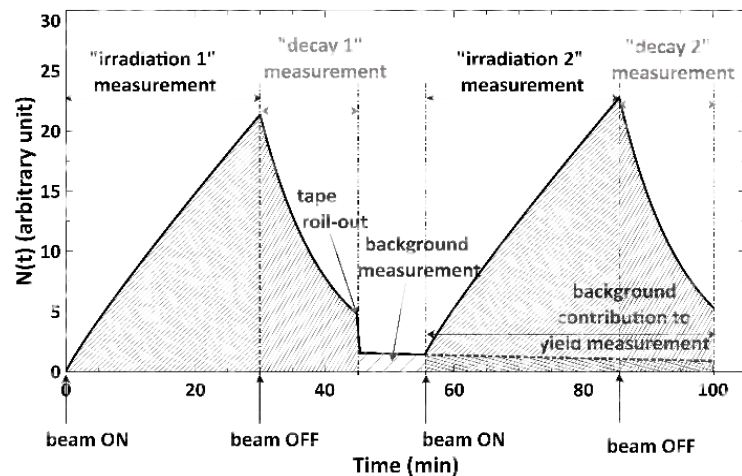
# Review of different techniques to assess Isomeric Ratio on LOHENGRIN

## ms to min Isomers

By product of isotopic yield analysis

Difficulty : Some Isomeric states have the same  $\gamma$  lines as ground state

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



## $\mu$ s Isomers

Difficulty : Isomeric states can be filled by the  $\beta$  decay of the father nuclei

Solution : Coincidence between ionization chamber and  $\gamma$  detectors

## ns Isomers

Difficulty : Isomeric states have a period

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

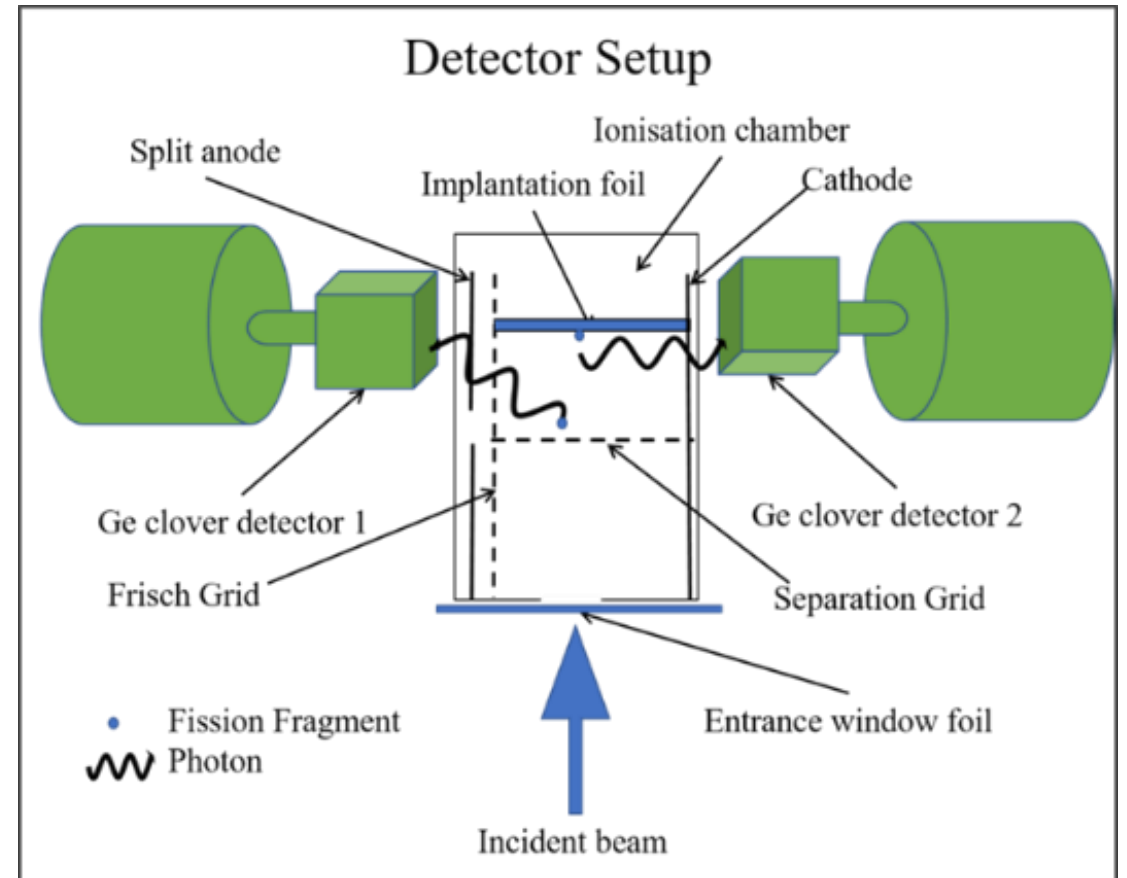
→ no direct measurement

Solution : Statistical analysis of the ionic charge distribution

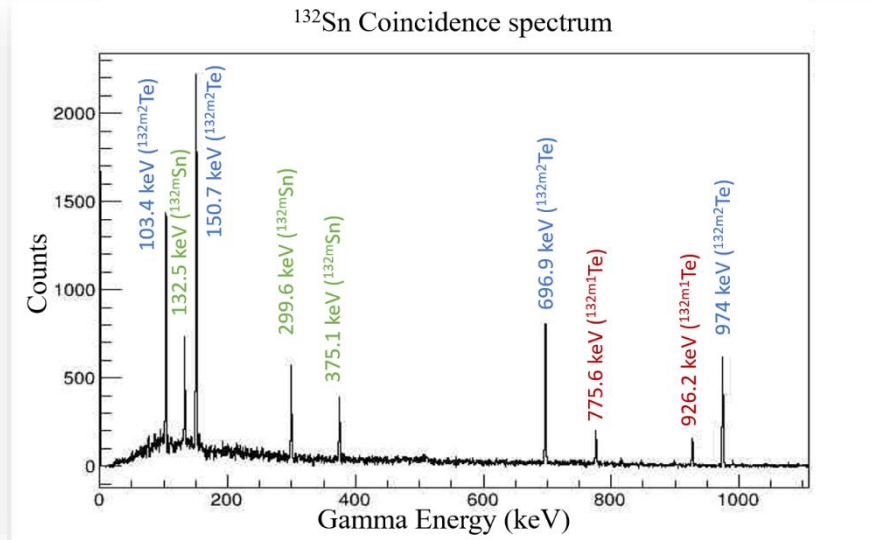
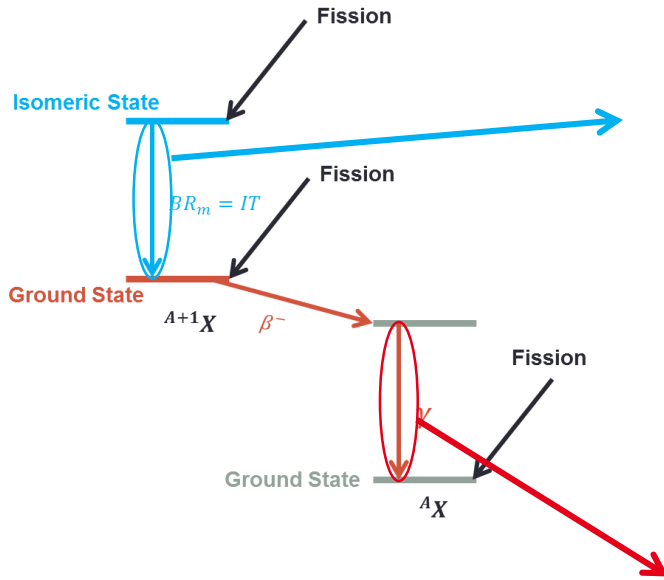




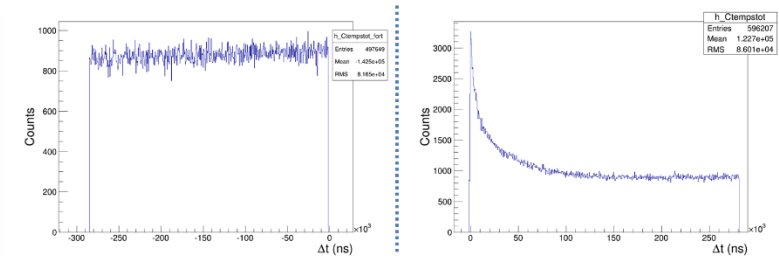
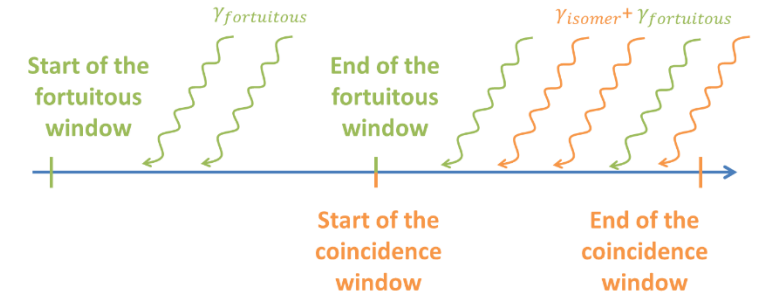
# $\mu$ s isomer experimental setup



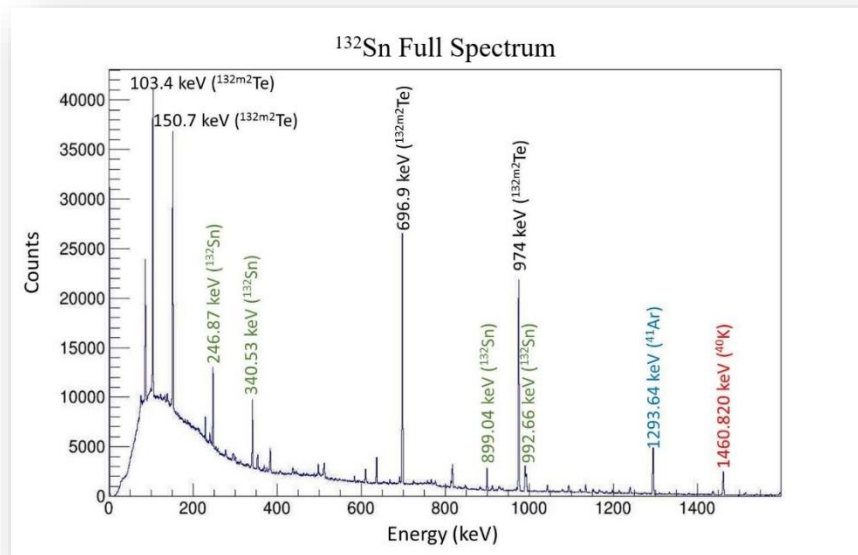
# Analysis of $\mu$ s isomers



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

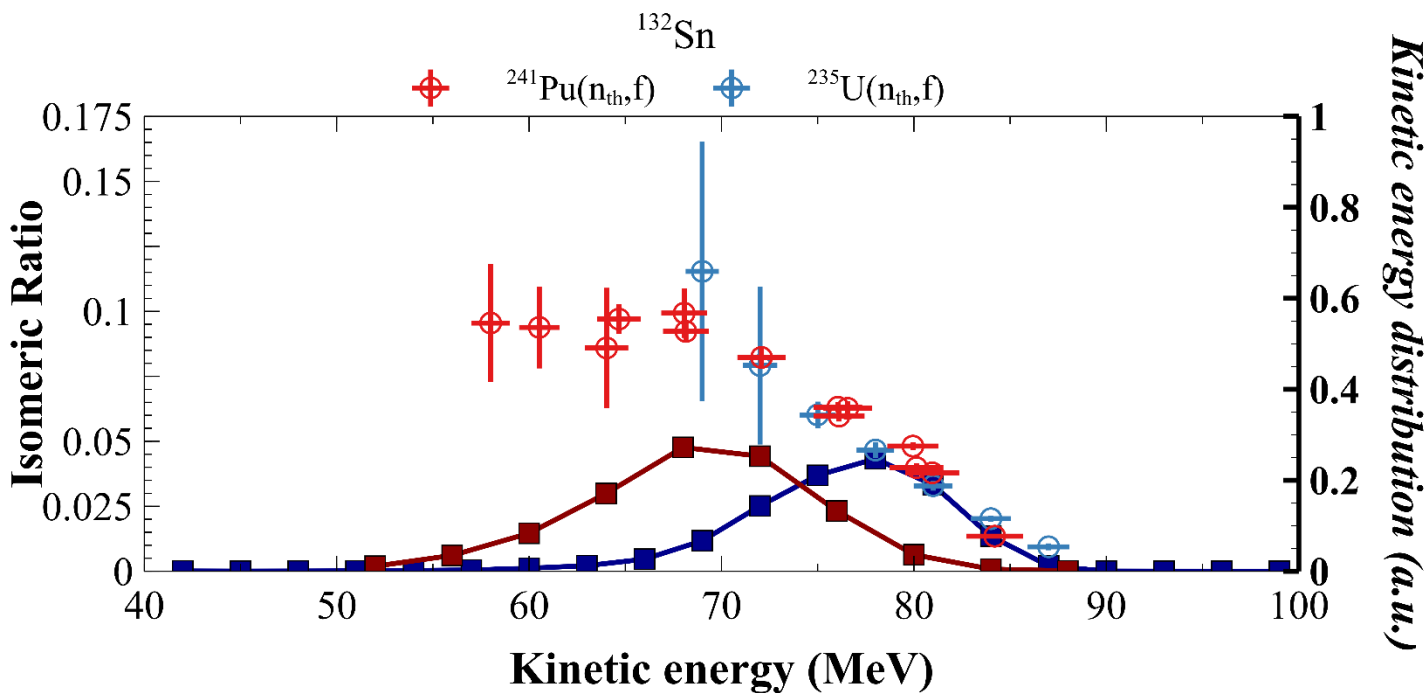


$$IR = \frac{\tau_f(^{132m}\text{Sn})}{\tau_f(^{132m}\text{Sn}) + \tau_f(^{132gs}\text{Sn})}$$

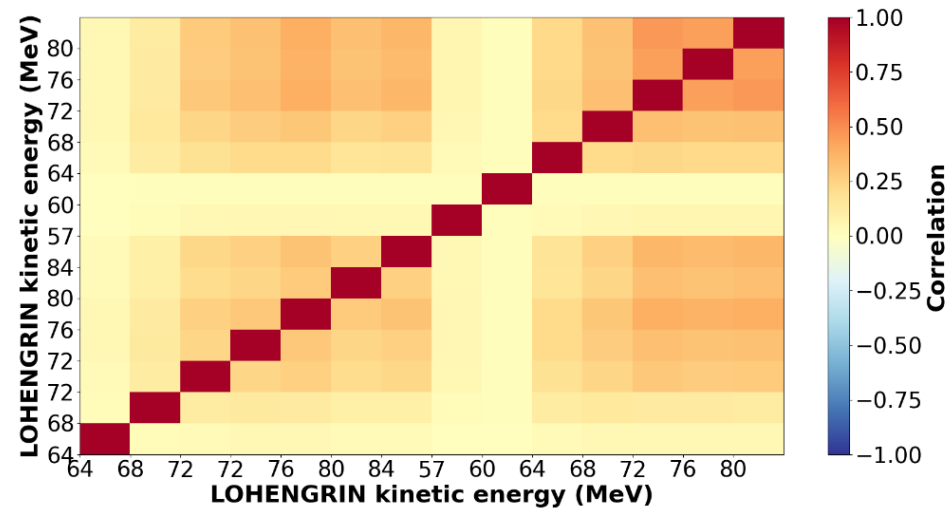
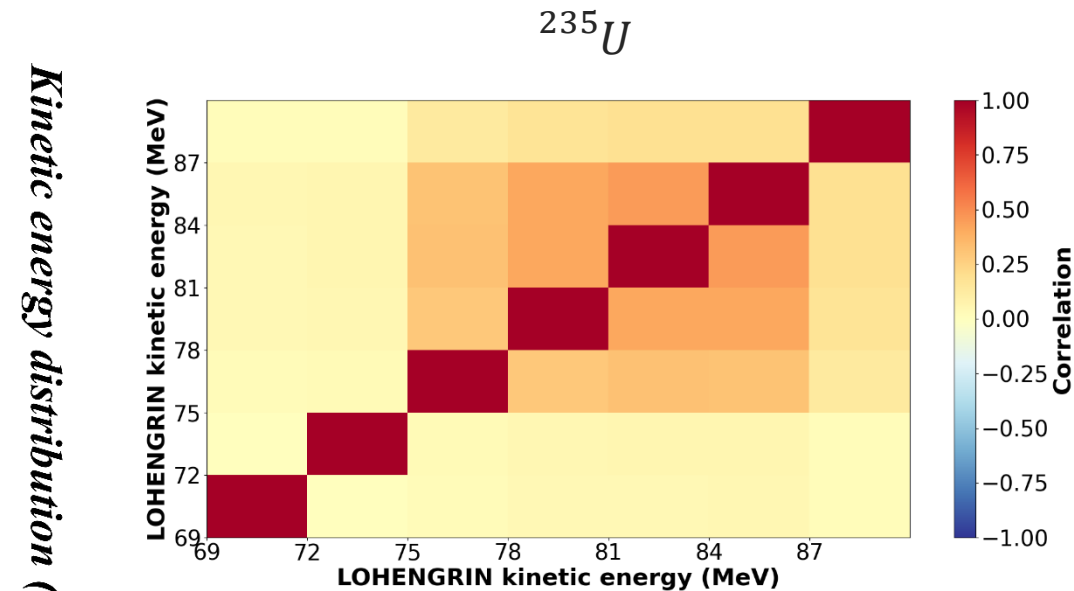


Ungated  $\gamma$  Spectrum i.e. without coincidence  
 → Ground state measurement  
 → Extraction more difficult because of the S/B ratio

# 132Sn case : results



Reaction	$\overline{IR}$ : $^{132}\text{Sn}$	$\sigma_{\overline{IR}}$ : $^{132}\text{Sn}$
$^{235}\text{U}(n_{\text{th}},f)$	0.054	0.006
$^{241}\text{Pu}(n_{\text{th}},f)$	0.068	0.002



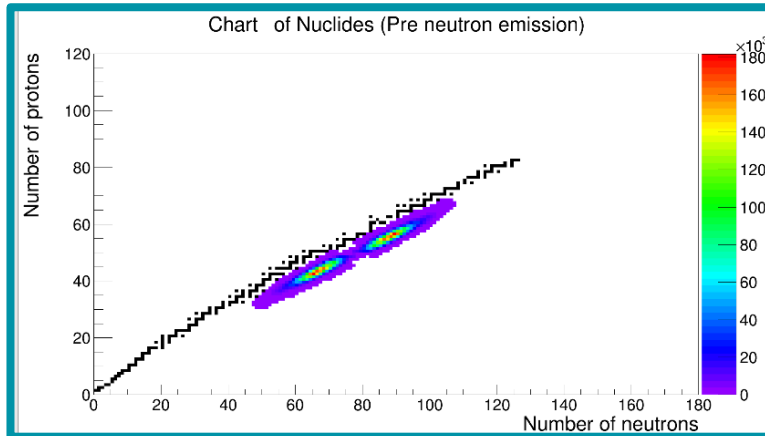


# FIFRELIN : a bridge between experimental and theoretical worlds

FIFRELIN (Fission Fragments Evaporation modeLING)

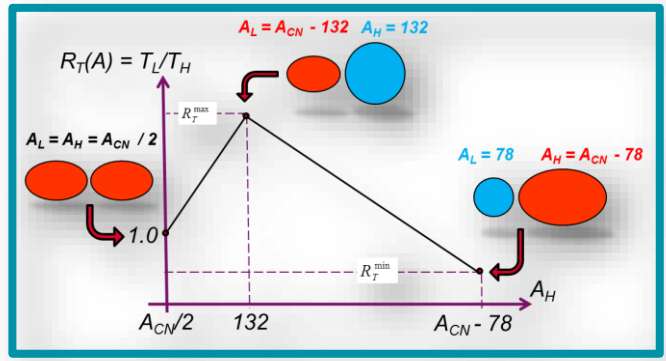
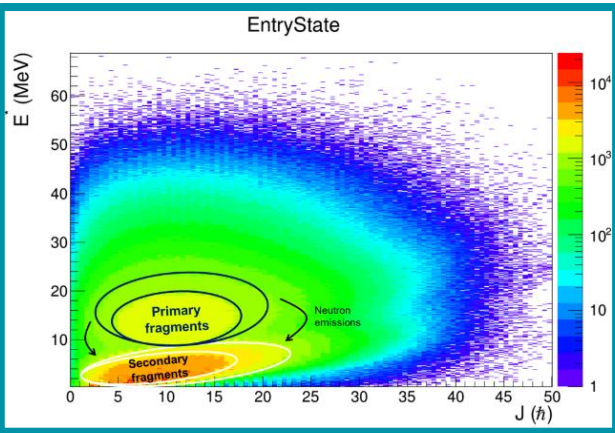
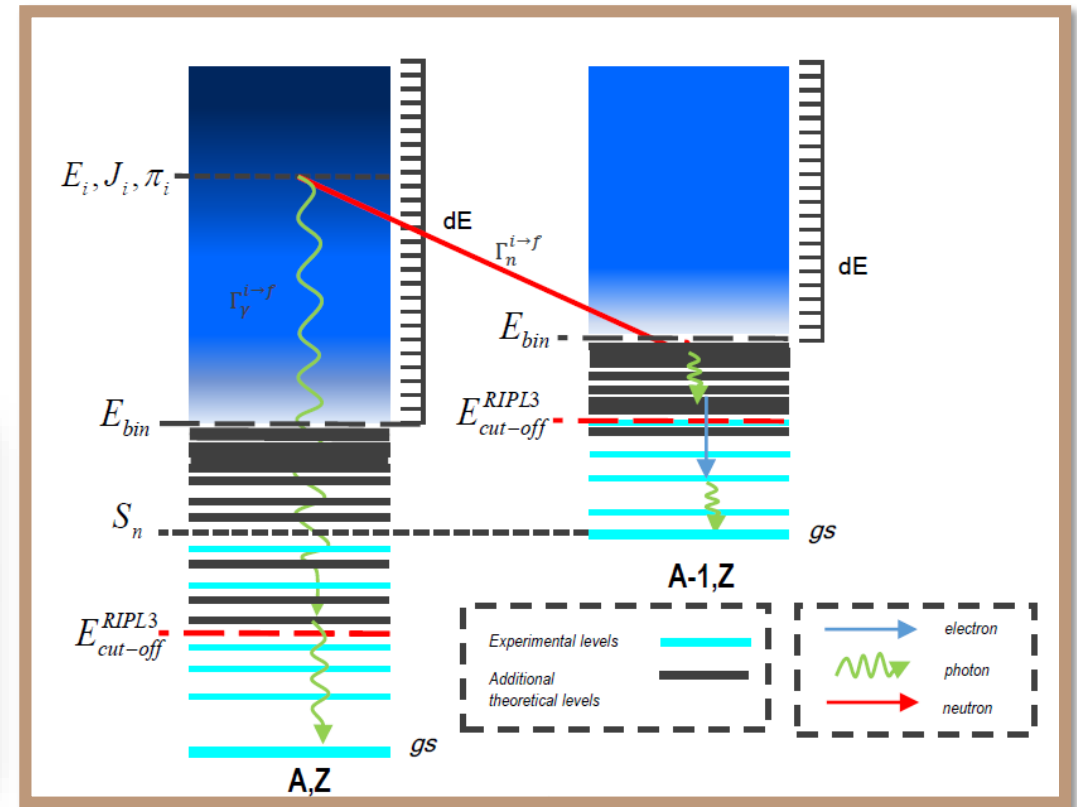
- Goal : characterize fission fragment since its creation ( $\sim$  scission) until  $\beta$  decay (not included)

## Fission Fragments Generation

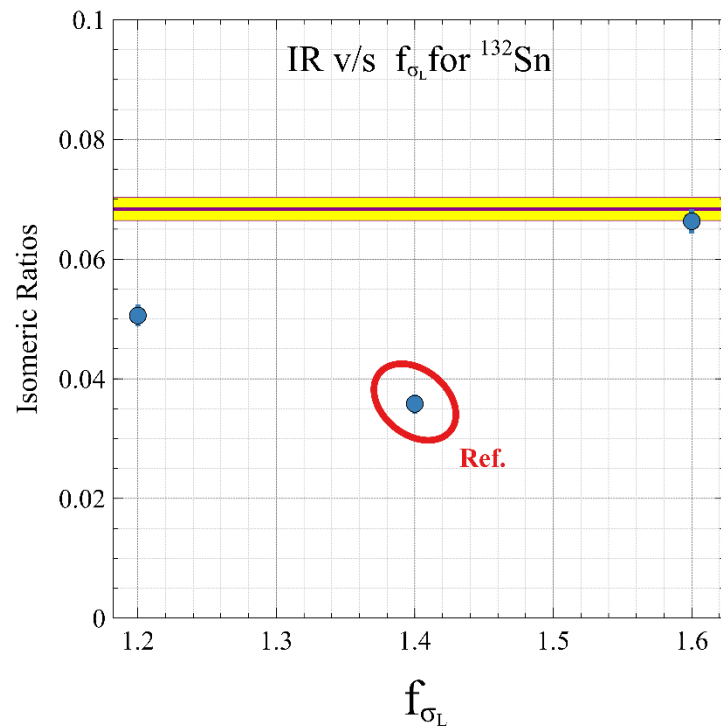
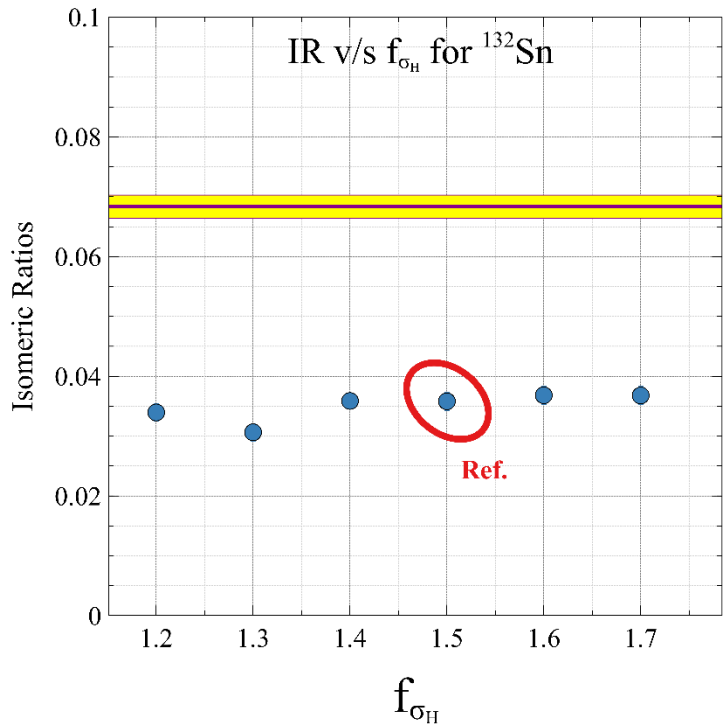


2 steps  
code

Creation of nuclear levels for given fission fragments and daughters



# Comparison with classic FIFRELIN calculation



●  $f_{\sigma_L}=1.4$     ■ Exp.  $\langle\text{IR}\rangle$

●  $f_{\sigma_H}=1.5$     ■ Exp.  $\langle\text{IR}\rangle$

- Level density model : CGCM
- Model of  $\gamma$  strength function (EGLO)
- The impact of  $f_{\sigma_L}$  is more important than  $f_{\sigma_H}$
- Can be explained by nucleons exchange at scission (TDHFB)  $\rightarrow$  role of deformation energy?
- Thermal excitation ?
- Role of the rotational energy in the total energy excitation sharing ?
- Correlation between both fragment arise naturally !

# Direct determination of angular momentum with FIFRELIN decay

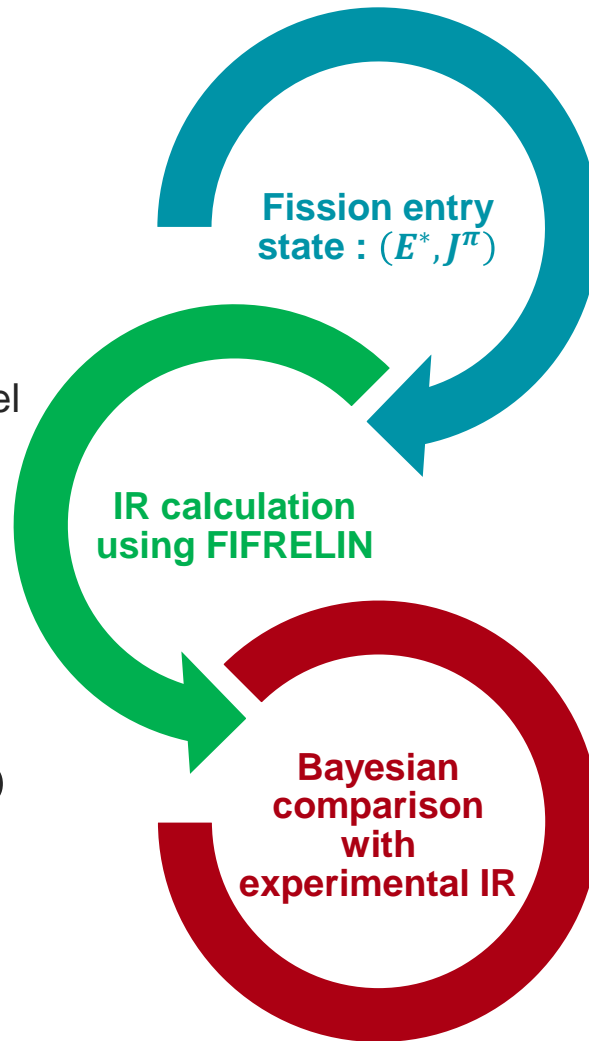
In this work, FIFRELIN (developed by CEA Cadarache) is used **only as a nuclear de-excitation code (step 2)**

What is required for FIFRELIN :

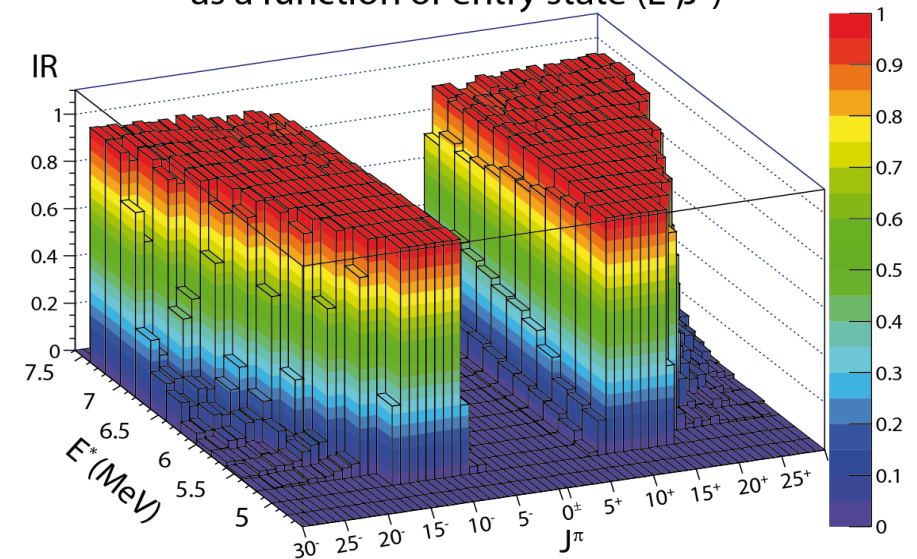
- experimental level scheme (RIPL-3)
- Model of nuclear density to **complete** the level scheme (CGCM)
- Model of  $\gamma$  strength function (EGLO)
- Electron conversion coefficients (Brlcc)

For comparison with experimental results standard spin distribution:

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

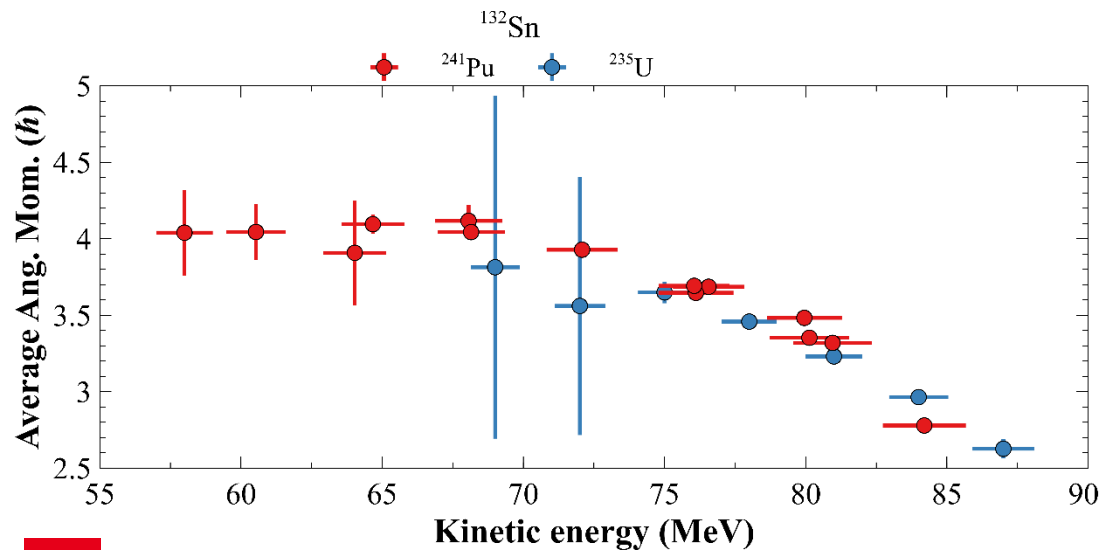
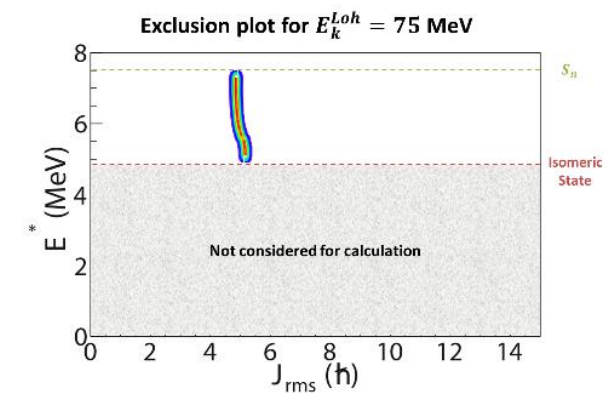
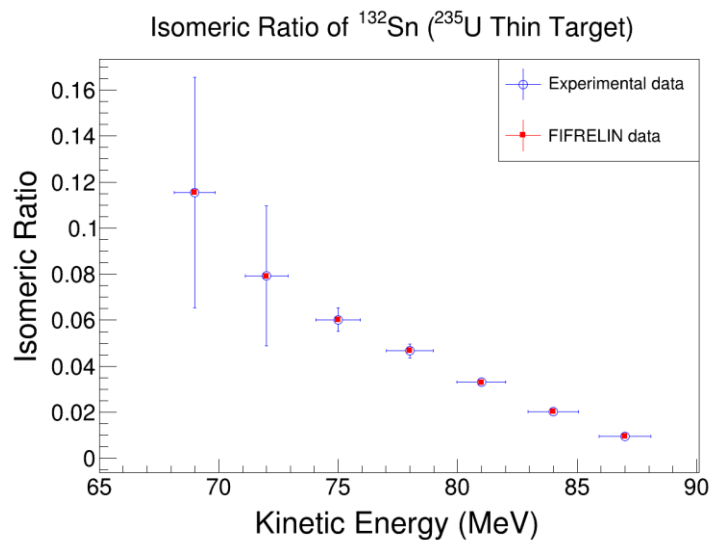
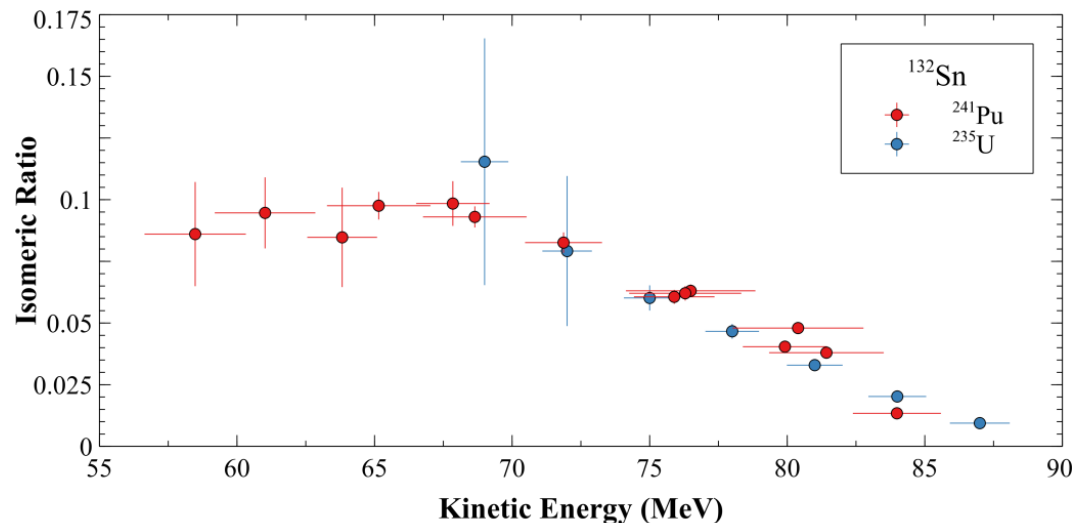


Isomeric Ratio calculated by FIFRELIN as a function of entry state ( $E^*, J^{\pi}$ )



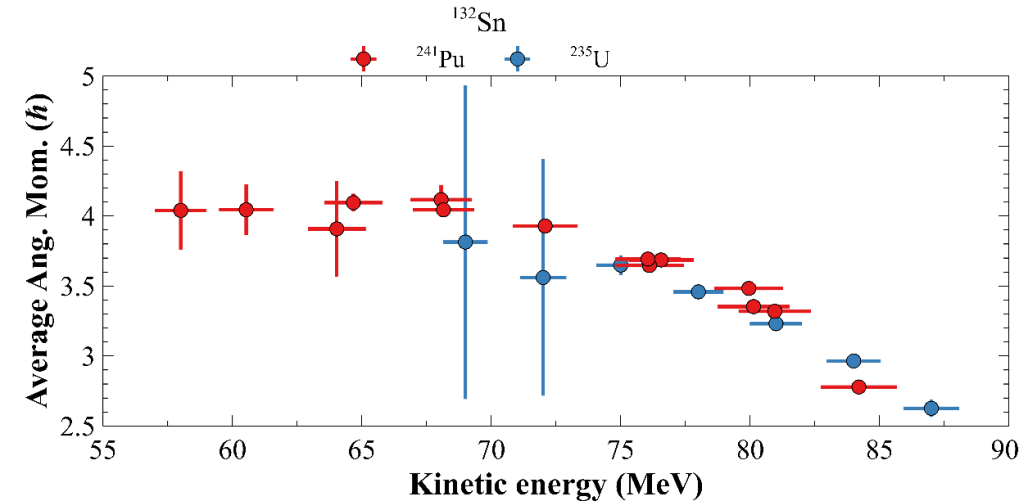
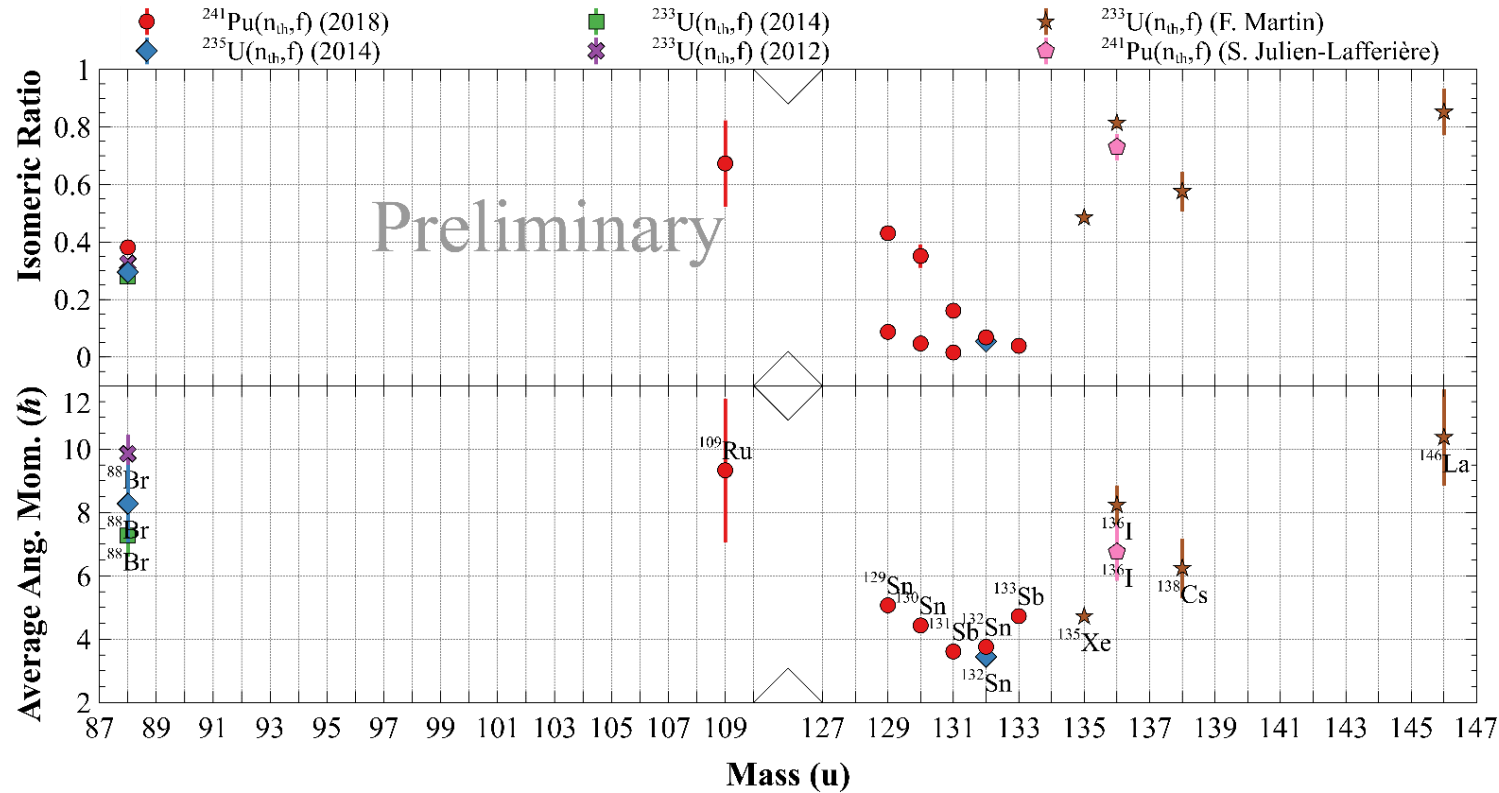


# Synthesis on $^{132}\text{Sn}$ work



- IRs are compared with FIFRELIN calculations starting from arbitrary initial nuclear state
- The derived **average angular momentum** is dependent on the fission fragment **kinetic energy**
- HFB (from P. Marevic et al.) predicts  $2.5 \hbar$  (with  $^{239}\text{Pu}$ )
  - Neutron emission ?
  - Thermal excitation ?

# Synthesis



- Isomeric ratios : probe for fission fragment angular momentum
- **Dependency of the derived average angular momentum with the fission fragment kinetic energy**
- Next step : measurement in the light fragment region (part of a thesis 2025-2028)

# Some applications


- Needs from industrial partners to improve knowledge (central value and uncertainty) for some nuclides for radioprotection (human and environment) purposes
- IR can be directly useful for such requests !

Nuclide	Nuclear data	Uncertainty	Part (%) on total uncertainty
$^{134}\text{I}$	$^{239}\text{Pu} \rightarrow ^{134}\text{Te}$	14.1	52
	$^{239}\text{Pu} \rightarrow ^{134}\text{I}$	24.2	15
	$^{239}\text{Pu} \rightarrow ^{134\text{m}}\text{I}$	24.2	8
	$^{241}\text{Pu} \rightarrow ^{134}\text{Te}$	11.0	16

©V. Vallet





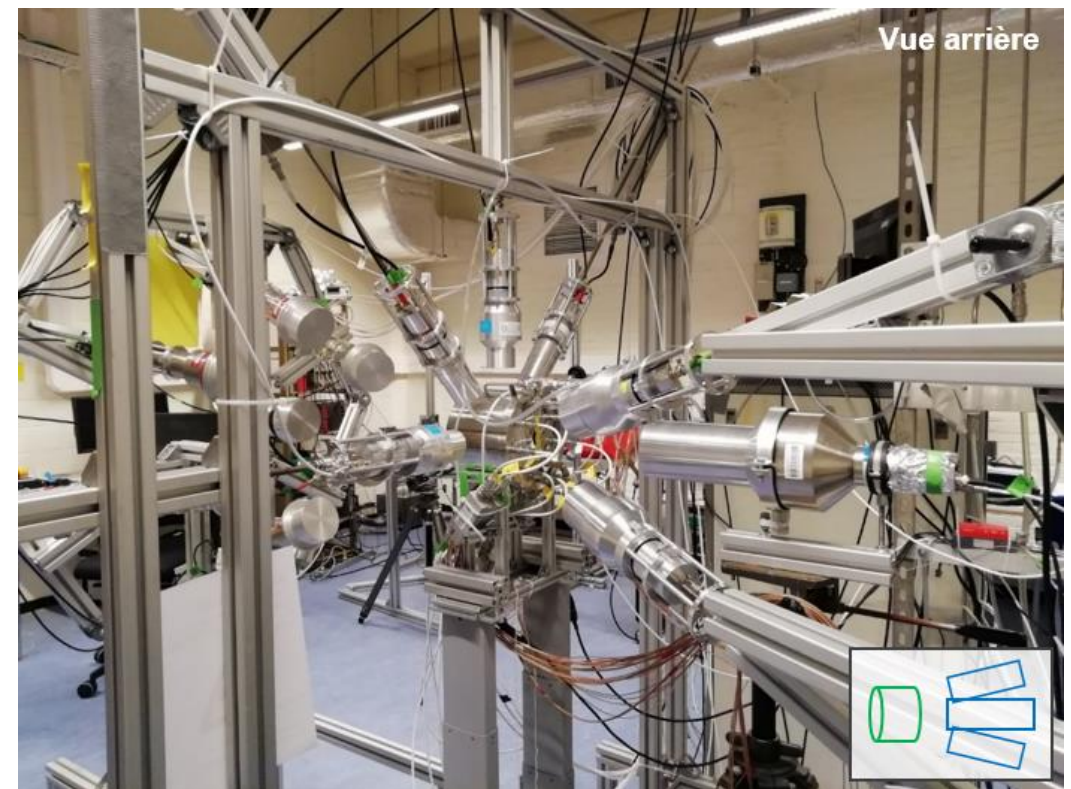
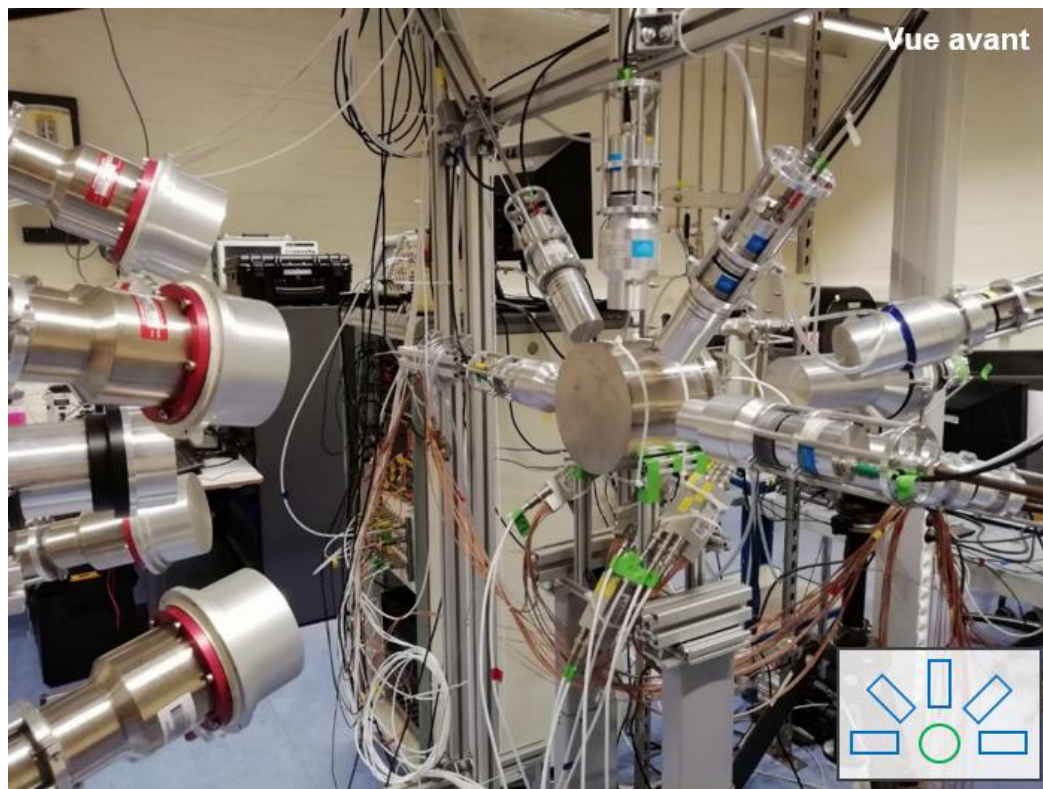


# 2 ■ Measurement of prompt $\gamma$ -rays characteristics with the VESPA setup

V. Piau thesis

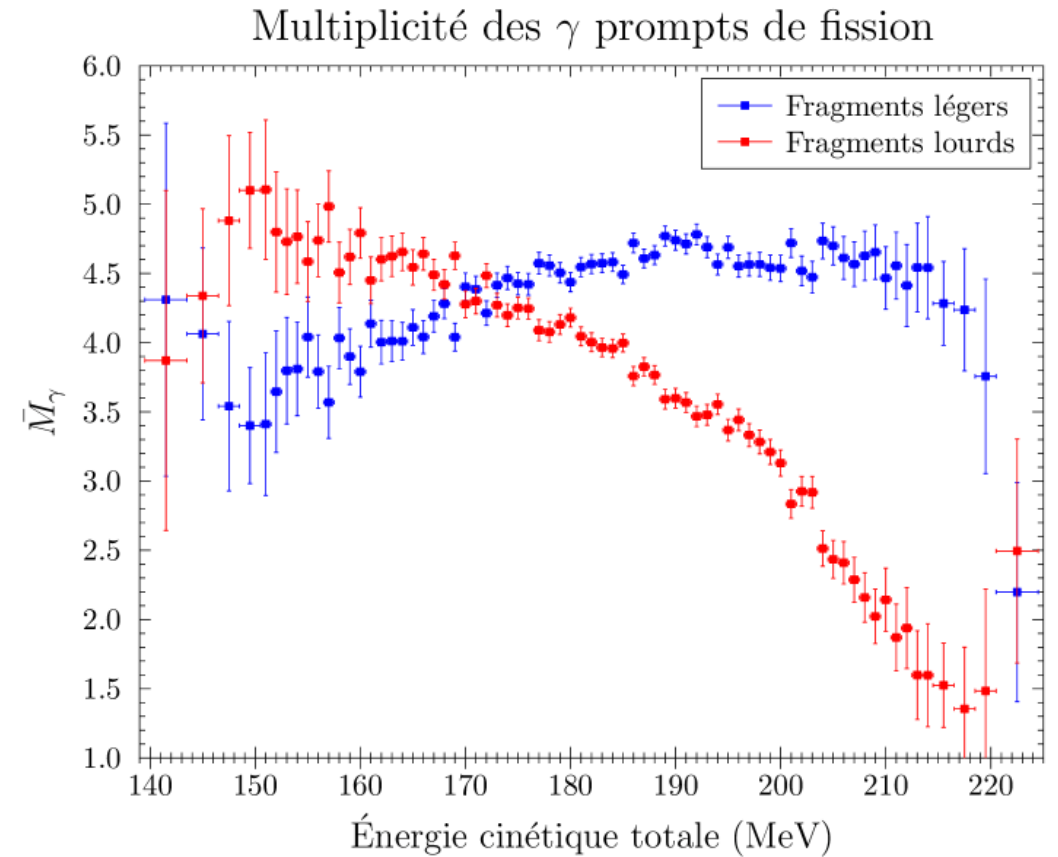
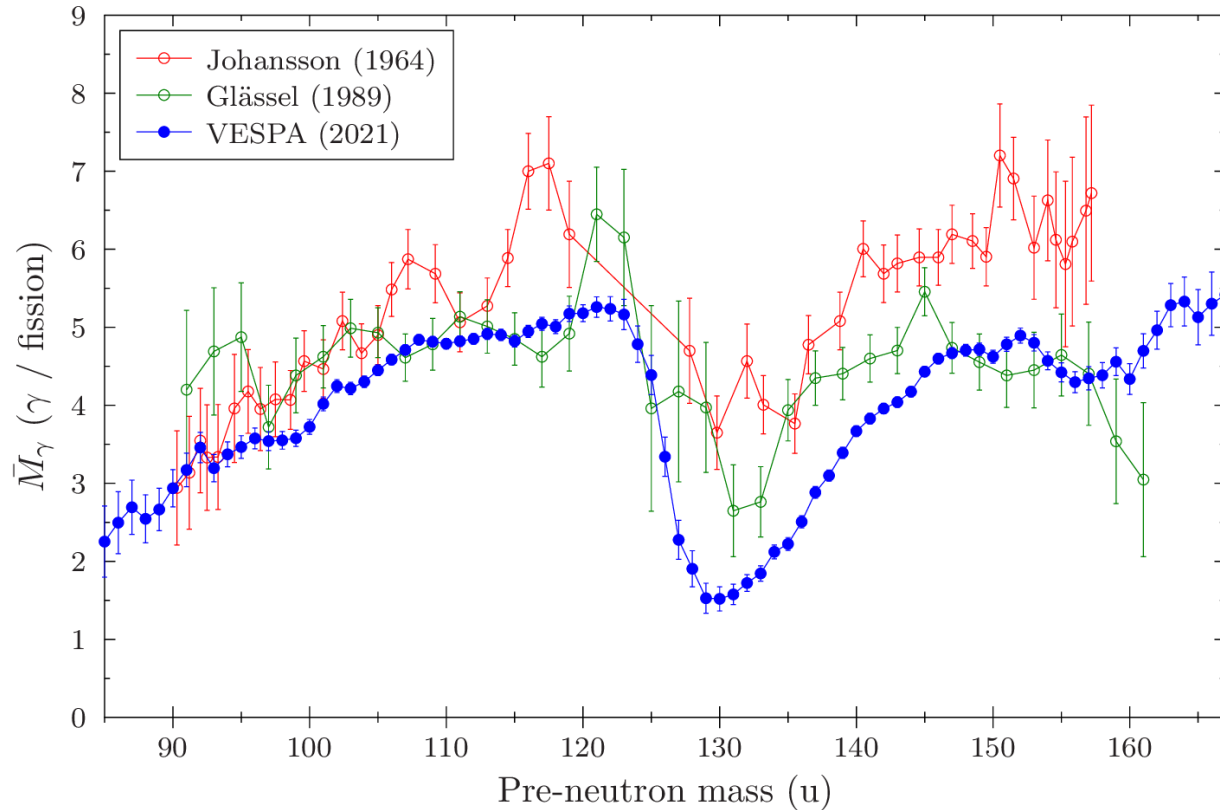
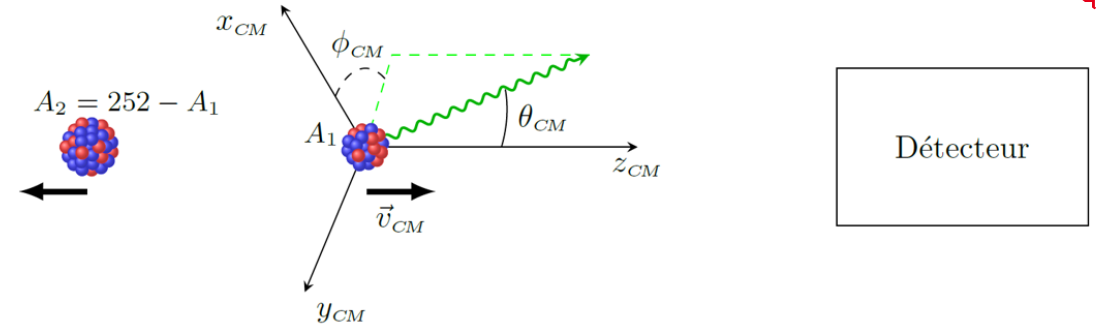
# VESPA setup

- Twin ionization chamber with Frisch Grid with  $^{252}\text{Cf}$  in the middle (Ni support)
- 8 LaBr<sub>3</sub> for  $\gamma$  detections
- 7 organic scintillators for neutron detections



# Correlation with mass

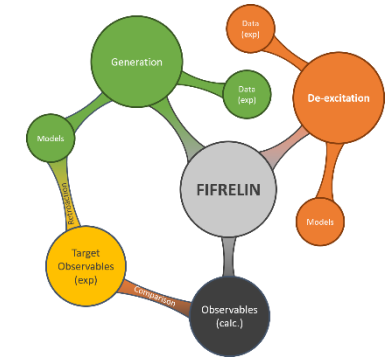
- Use of Doppler effect to associate  $\gamma$ -rays to one fragment !
- Only 3 detectors can be used.  
Here, the time windows is  $\pm 3$  ns and  $0.8 \leq E_\gamma \leq 8$  MeV
- Two detectors are normalized to the total multiplicity derived from A11218.



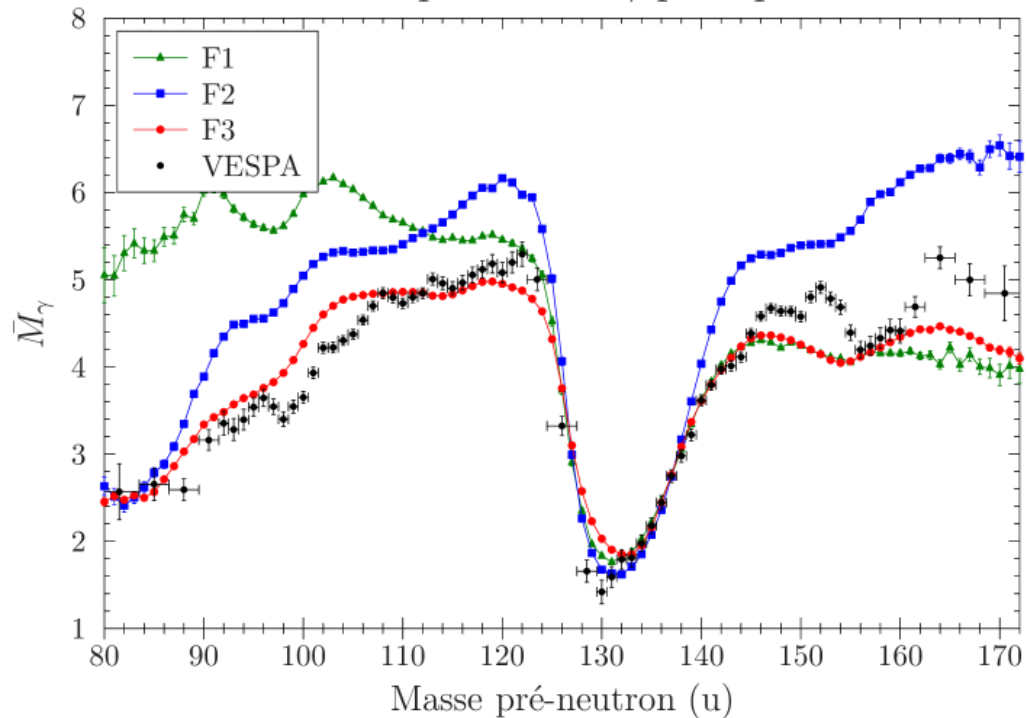
# Interpretation using FIFRELIN

Combination	Model of initial angular momentum	Model of level density	[ $RT_{\min}$ ; $RT_{\min}$ ]	[ $k_L$ ; $k_H$ ]
F1	Constant	CGCM	[0.45;1.40]	[10.5;8.0]
F2	Energy dependent	CGCM	[0.5;1.40]	[1.7;1.5]
F3	Energy dependent	HFB14	[0.5;1.45]	[1.4;1.3]

$$\overline{\nu}_L = 2.07; \overline{\nu}_H = 1.70$$

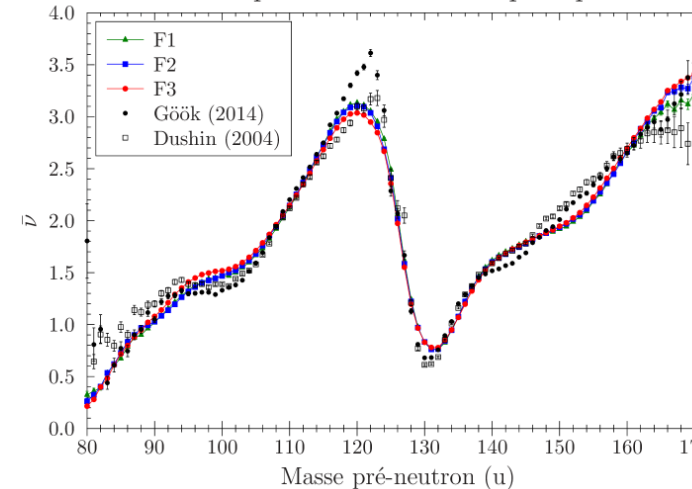


Multiplicité des  $\gamma$  prompts

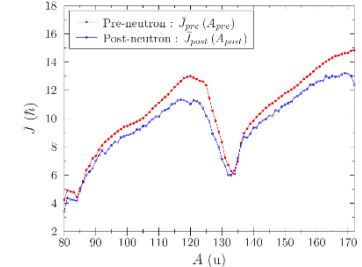


- F3 is the best combination of models
- Sawtooth behavior seems to raise from energy dependent model
- The scaling is better using HFB14 instead of CGCM level density

Multiplicité des neutrons prompts



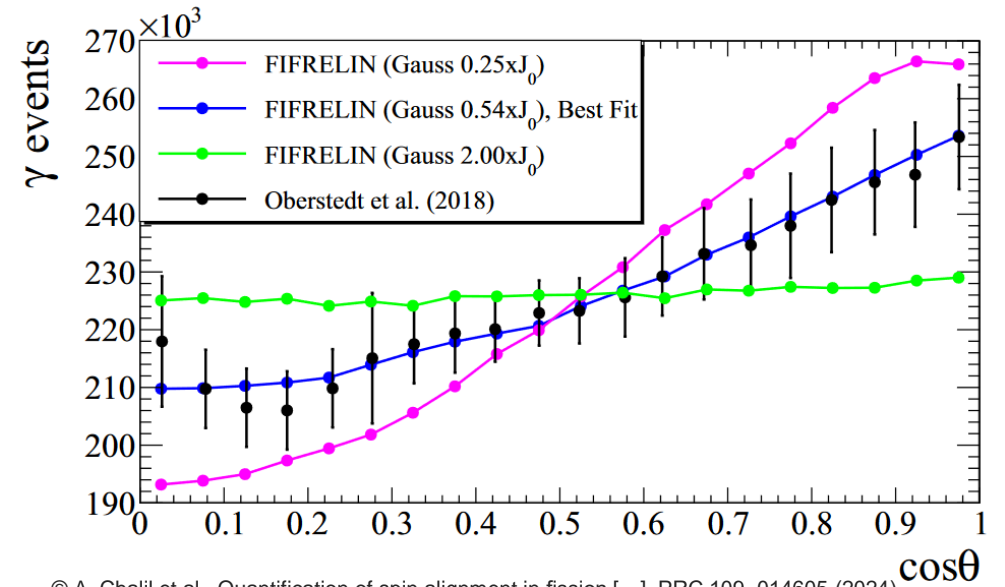
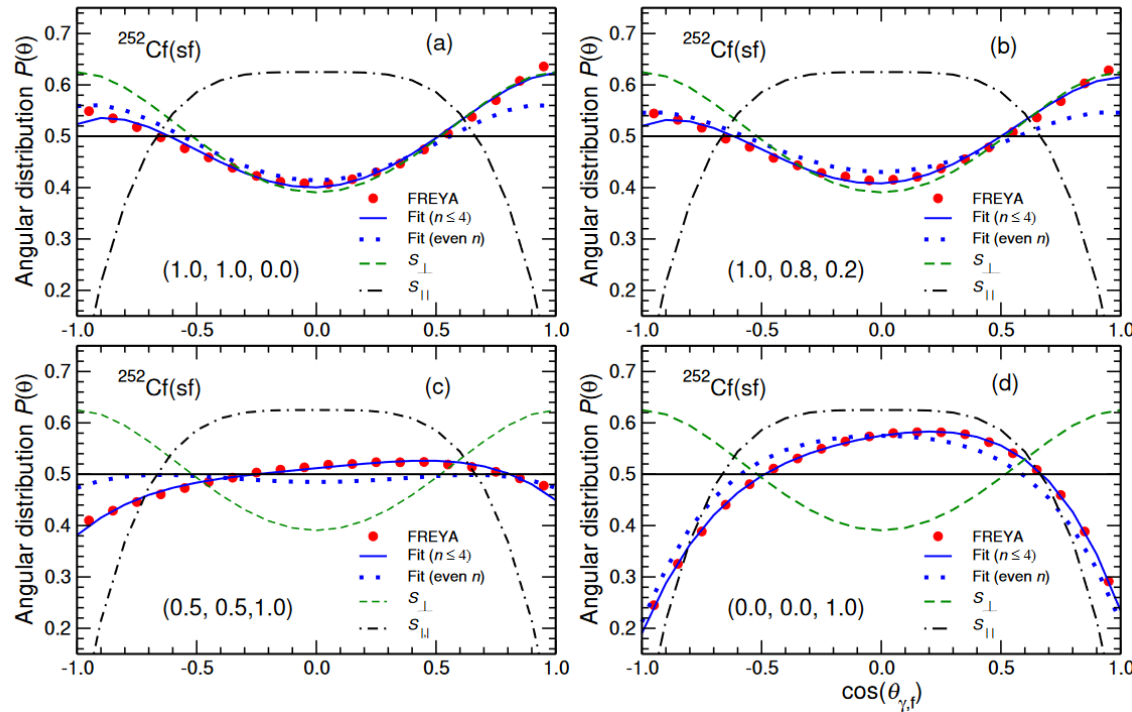
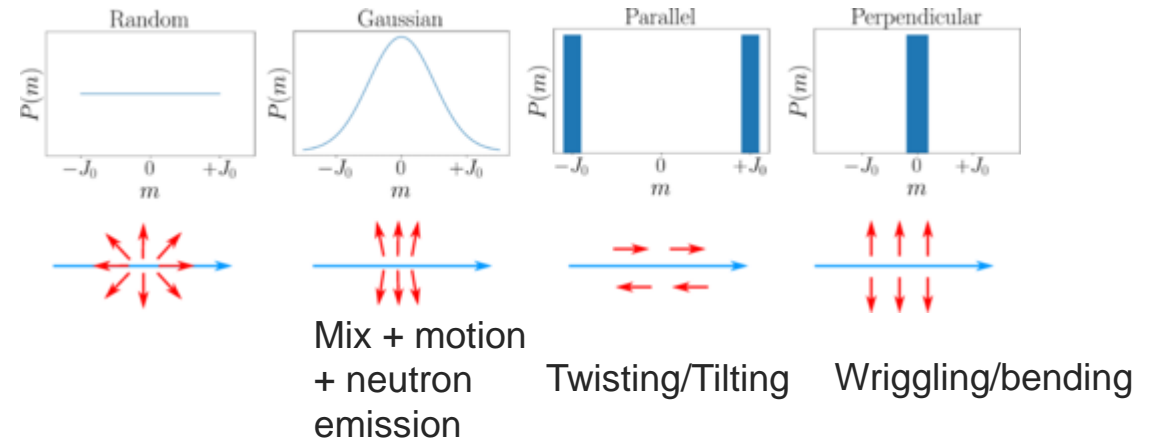
Moment angulaire total des FF





# To go further : angular correlation !

Simulated  $\gamma, FF$  angular correlation from E2 transition and different wiggling/bending/twisting feeding !



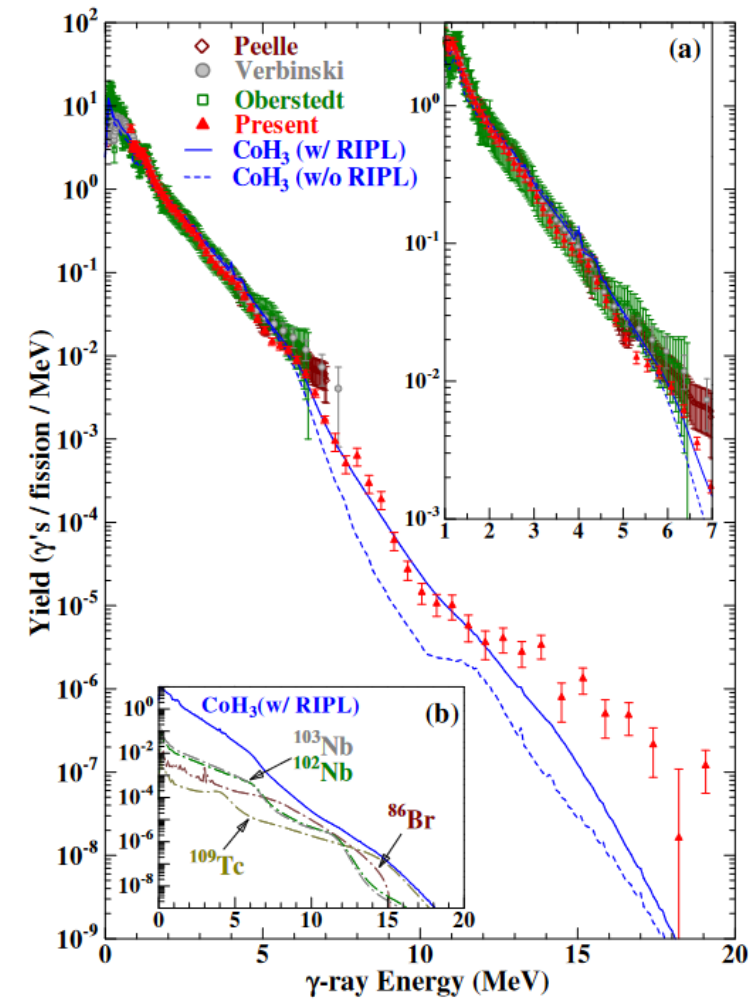
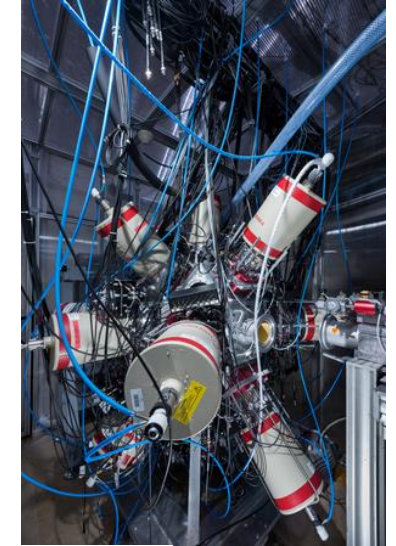
© R. Vogt and J. Randrup, The role of angular momentum in fission, EPJ Web of Conf. 292,08006 (2024)

© A. Chalil et al., Quantification of spin alignment in fission [...], PRC 109, 014605 (2024)

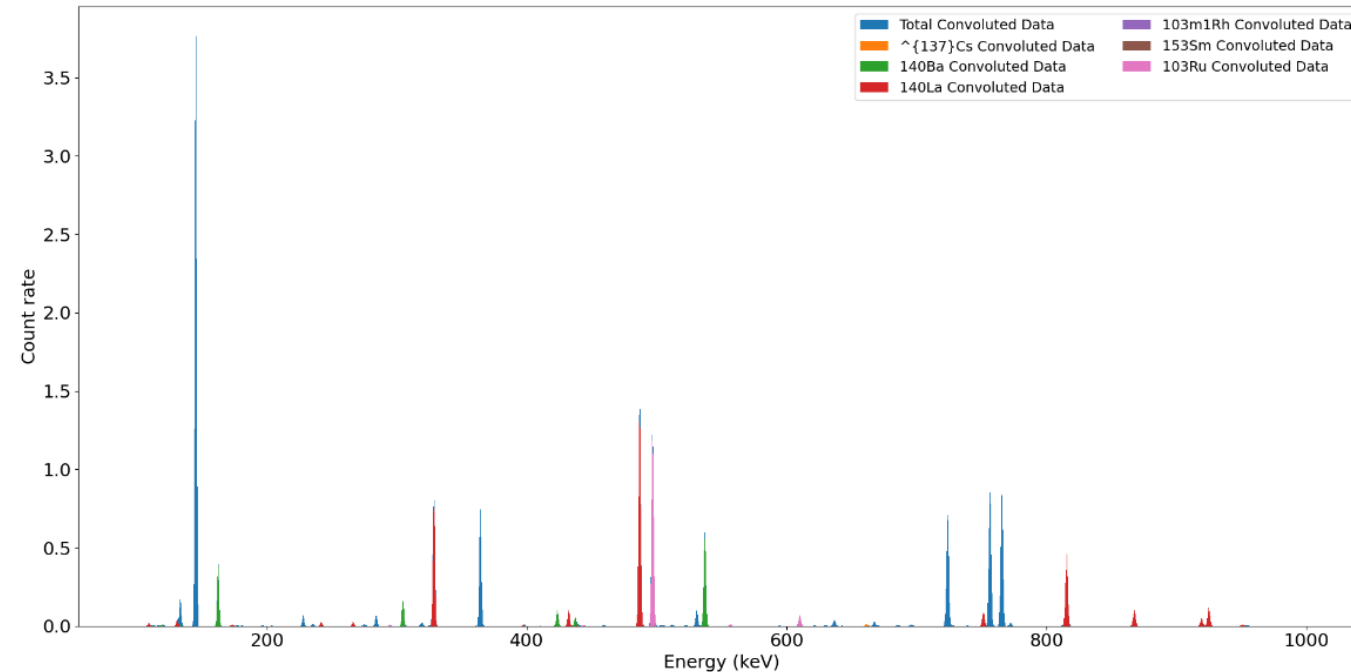
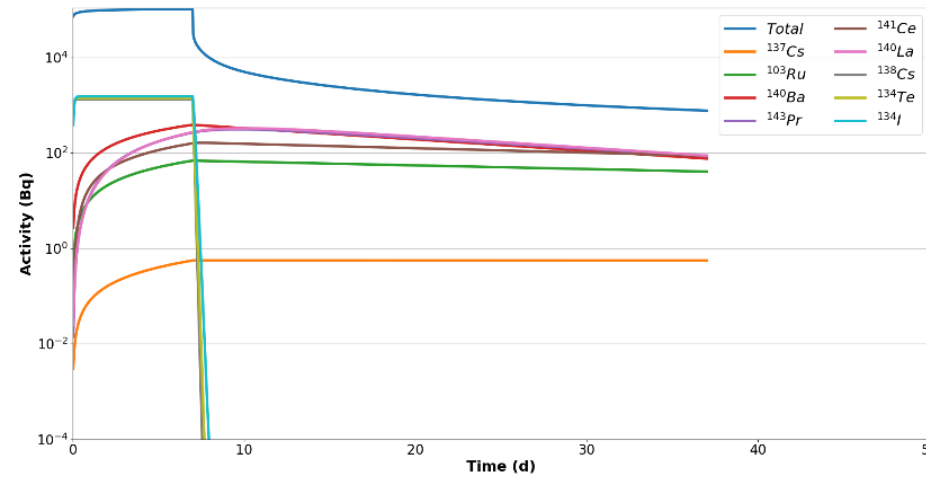


# 3 ■ Next programs

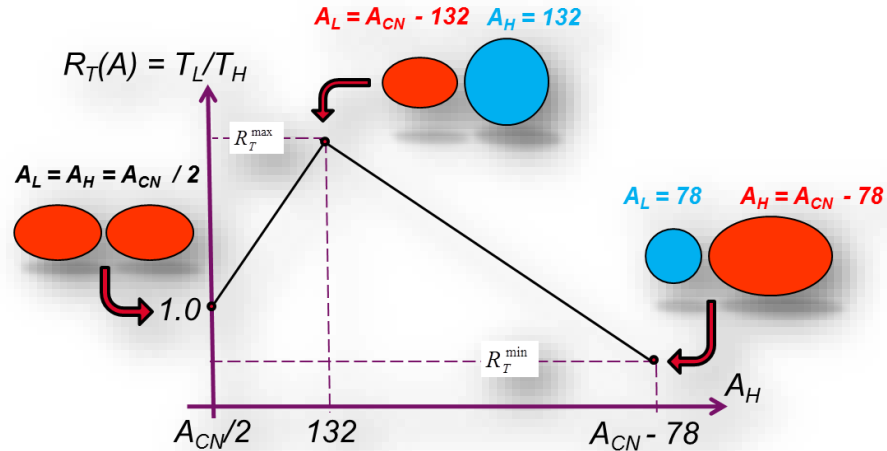
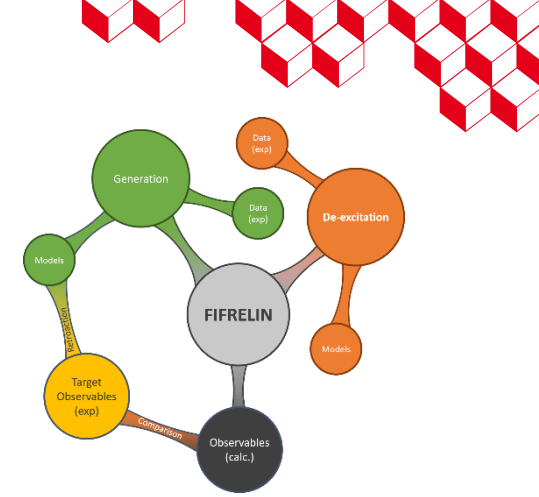
# New measurement incoming with FIPPS



From Makii et al., PRC, 100, 044610 (2019)

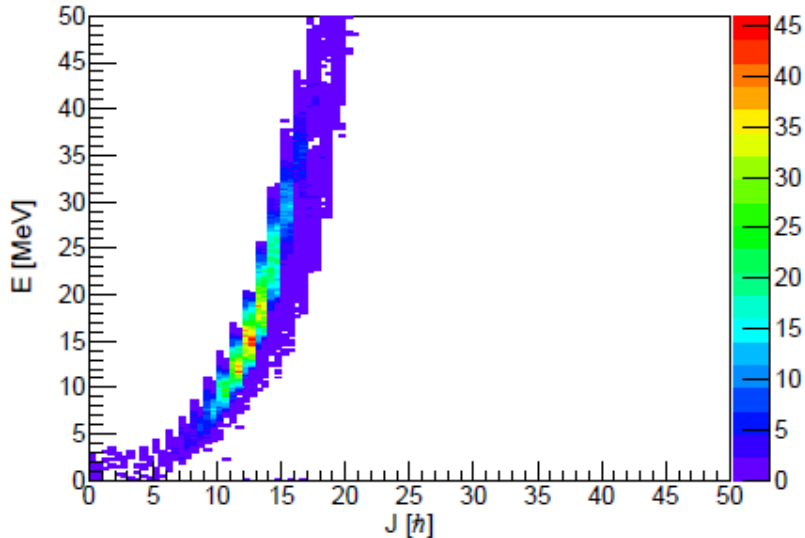


# Artificial Intelligence with FIFRELIN



How to determine these 4 parameters ?

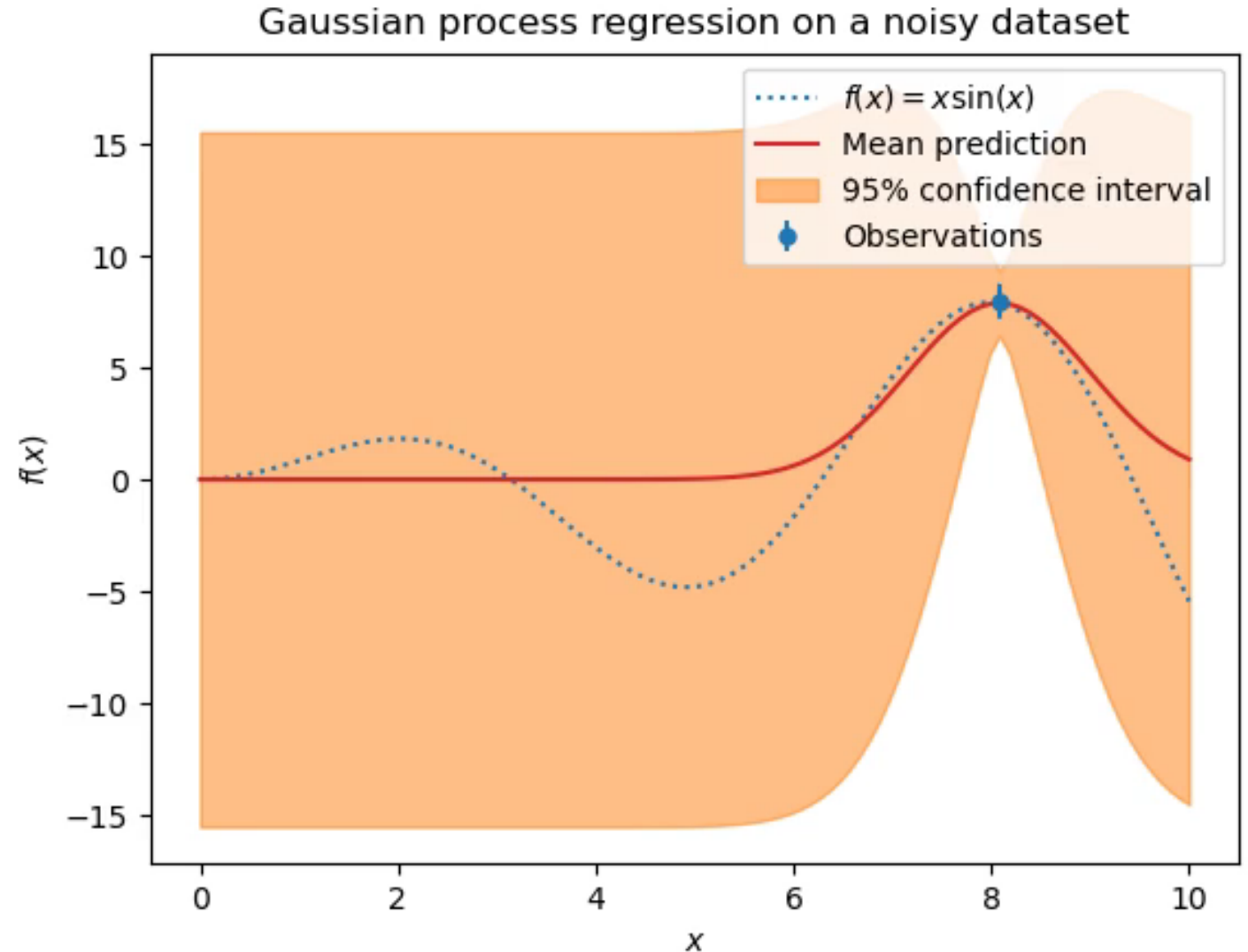
- If we consider each of them as independent, and at least 10 steps to find their best values, we need  $10^4 = 10000$  simulations
- One way to reduce the number of simulations (time consuming) : Machine Learning
- In this work (of G. Bazelaire), we started with Kriging algorithm



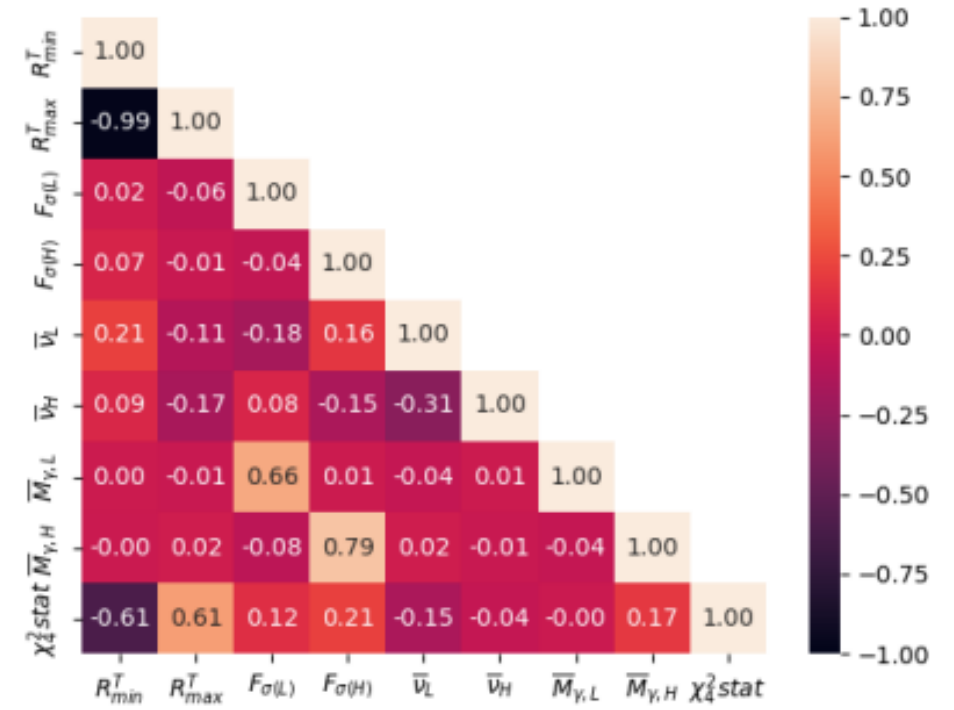
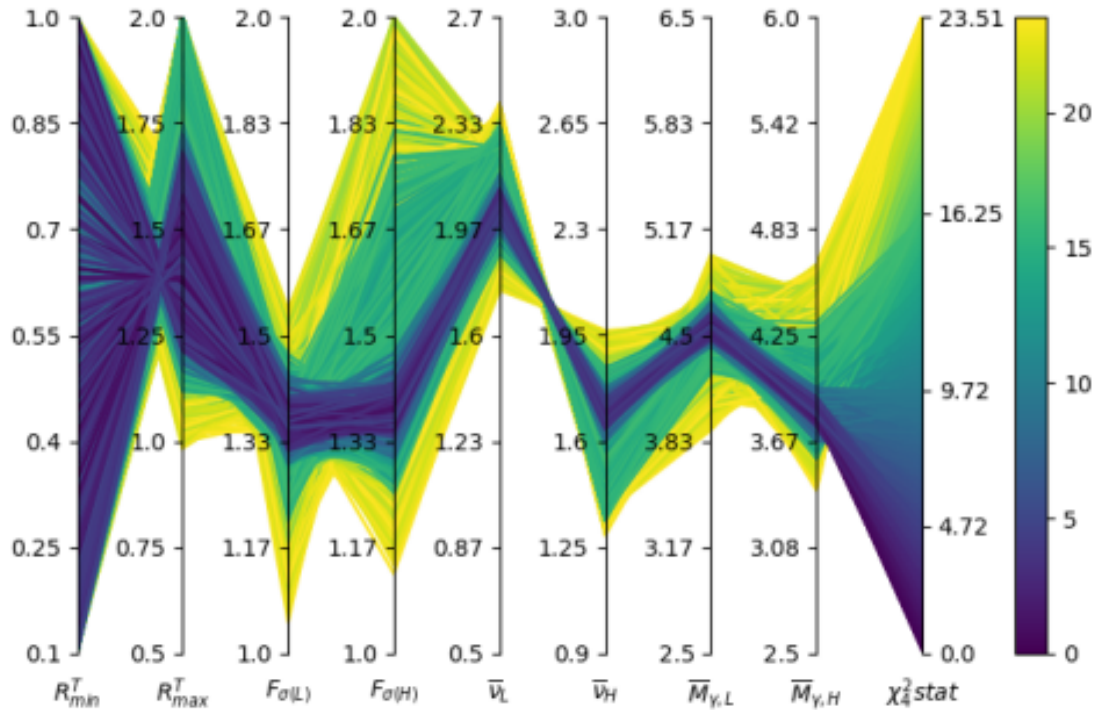


# Kriging

- Can be seen as a more complex way to make interpolation
- Suitable for linear problems!
- Based on prior covariance between data : hyper-parameters are fitted and control the smoothness of the interpolated function
- Not the best option for really high dimensions !

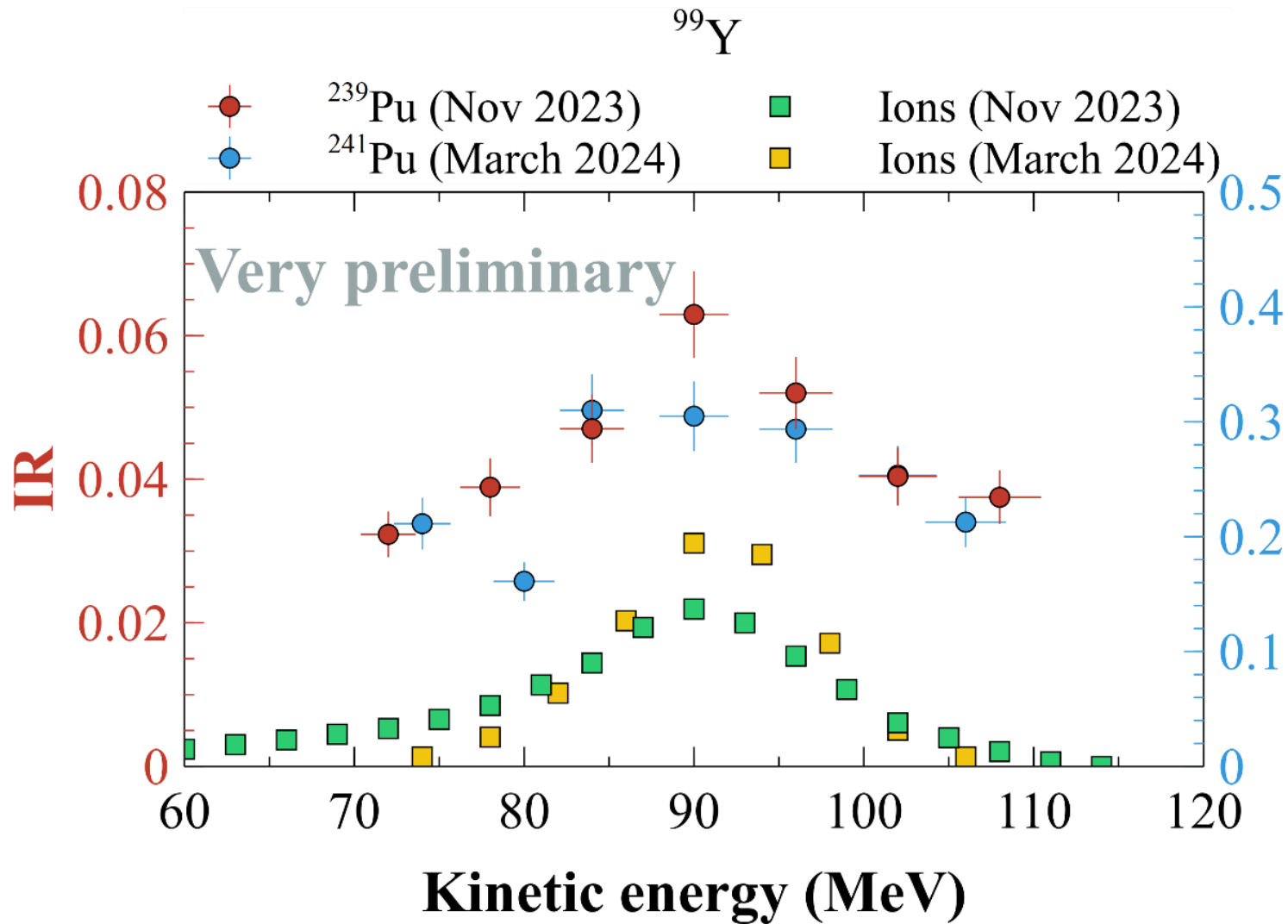


# Some results



- Target :  $[\bar{v}_L, \bar{v}_H, \bar{M}_{Y,L}, \bar{M}_{Y,H}] = [2.06, 1.70, 4.56, 3.82]$
- In 2h, from scratch, we found an optimum :  $[RT_{min}, RT_{max}, f_L, f_H] = [0.18, 1.58, 1.38, 1.37]$  thanks to 100 simulations

# Isomeric ratios in the light fragment mass region

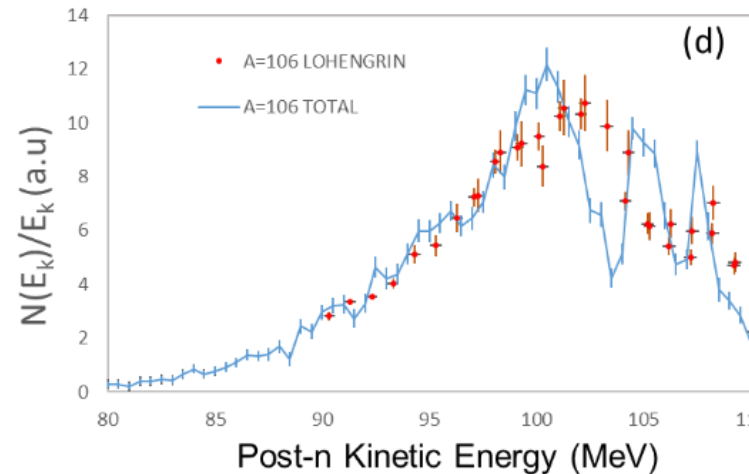
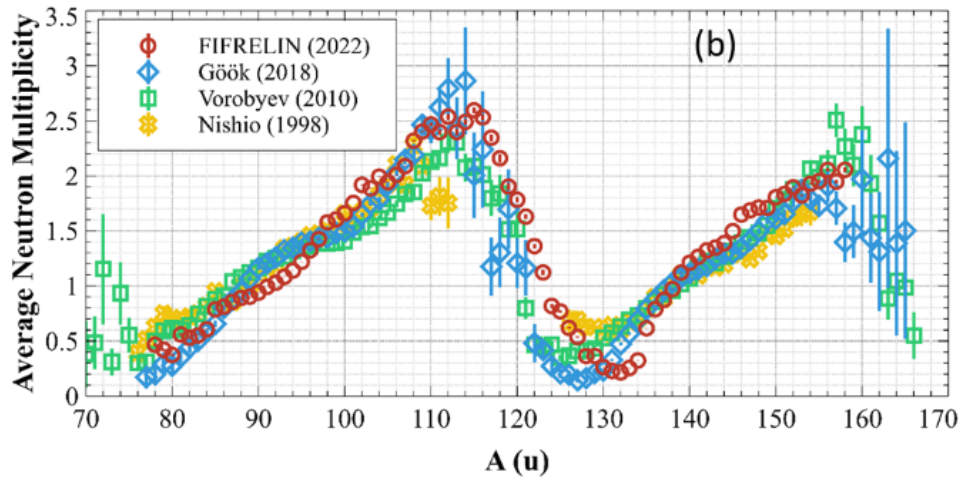
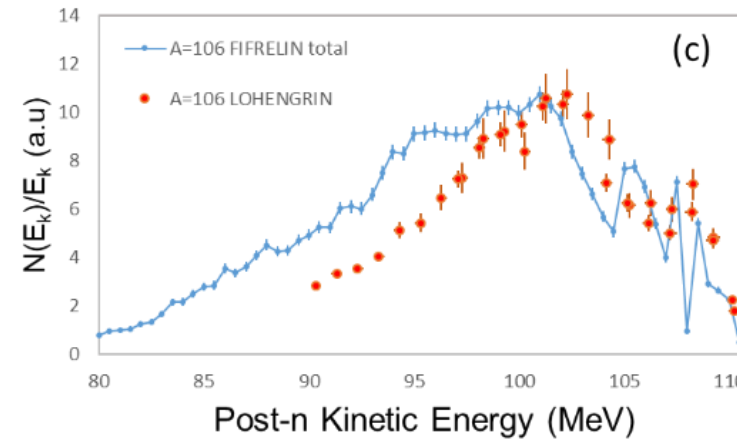
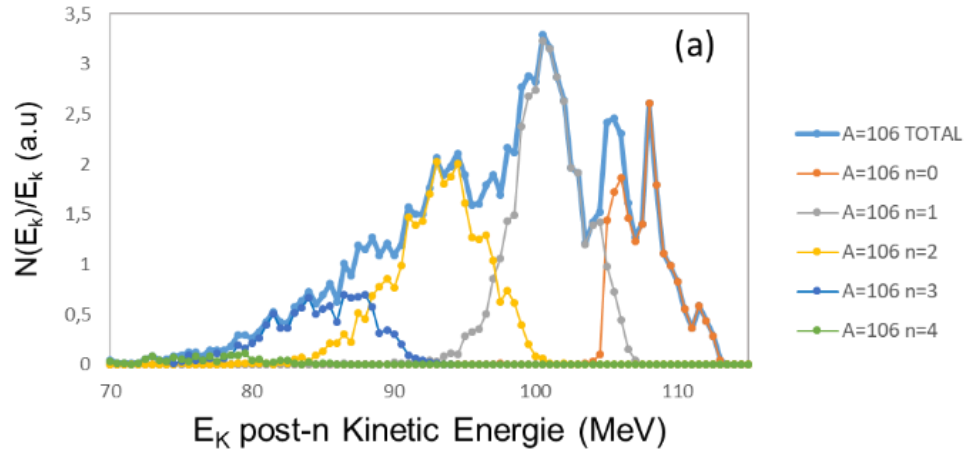


Isotope	$^{239}\text{Pu}$ (Nov 2023)	$^{241}\text{Pu}$ (March 2024)
$^{88}\text{Br}$	90	82/86/90/94/98/102
$^{94}\text{Y}$	72/78/84/90/96/102/108	78/84/90/93/96/100/104
$^{95}\text{Y}$	72/78/84/90/96/102/108	74/78/82/86/90/102/106/110
$^{99}\text{Y}$	72/78/84/90/96/102/108	74/80/84/90/96/102/106
$^{100}\text{Nb}$	72/78/84/90/96/102/108	78/82/86/90/94/98/102/106
$^{133}\text{Sb}$	60/66/72/80/86/90	
$^{138}\text{I}$	66/72/80/86	
$^{109}\text{Ru}$		82/90/102

Kinetic energy distribution (a.u.)

- Quite successful campaigns
- Will be part of a thesis for 2025 - 2028
- Next :  $^{241}\text{Am}/^{243}\text{Cm}/^{235}\text{U}$

# Estimation of neutron emission through accurate kinetic energy distribution measurements

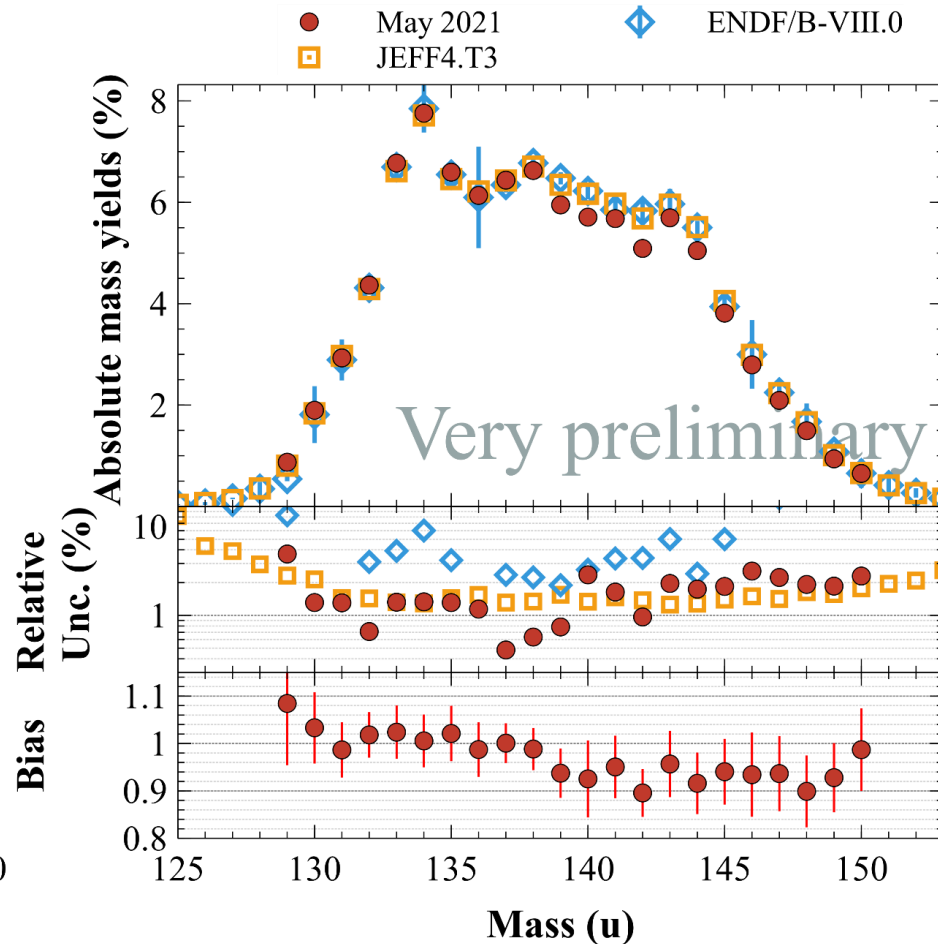
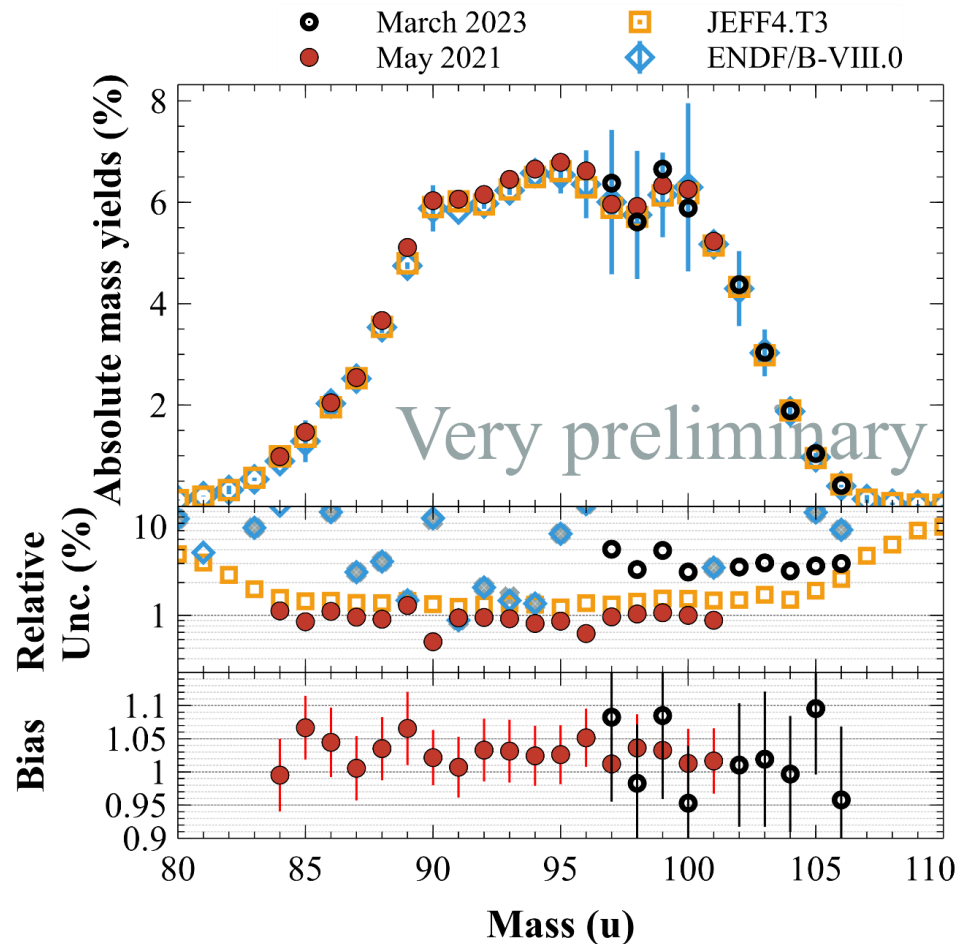


- New experiment planned in July 2024
- Part of a thesis in 2025 - 2028



# High precision mass yield measurements

- Context : new fission mass yield evaluation provide uncertainty around 2% by taking into account ~ 20 experiments
- Aim : provide new independent data set in order to test such new evaluation
- Next : Symmetry mass region with new setup !





# 4. Conclusions

# Conclusions

Nuclear structure drives the fission process

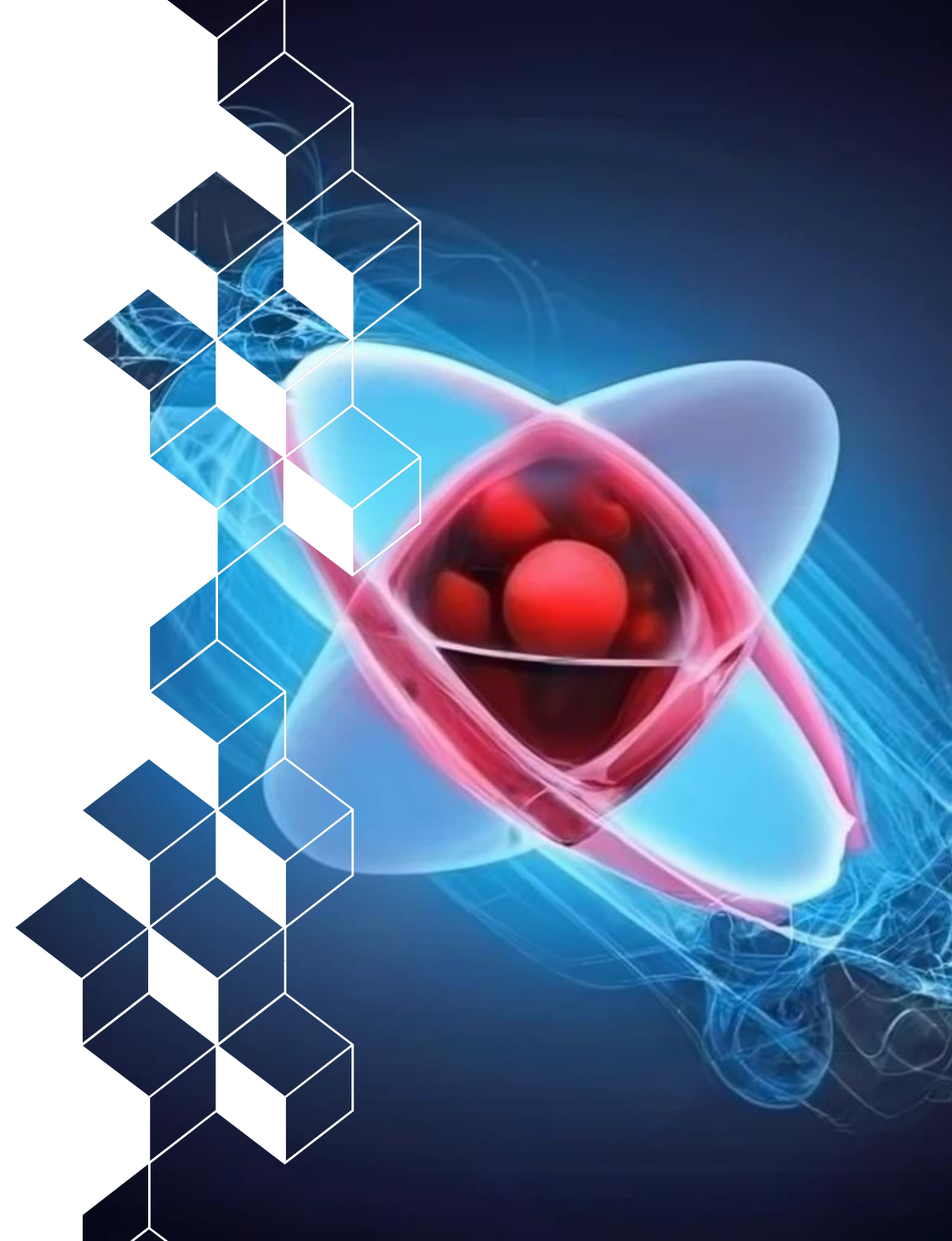
- Impact of level densities ( $\gamma$  multiplicity)
- Competition between Spherical/Quadrupole/Octupole deformation (structures in mass yields)

The generation of fission fragment angular momentum is a combination of several mechanisms

- Collective modes due to deformation
- Thermal excitation
- Nucleons exchange at scission
- Coulomb repulsion
- Strong connection with excitation energy (intrinsic?)

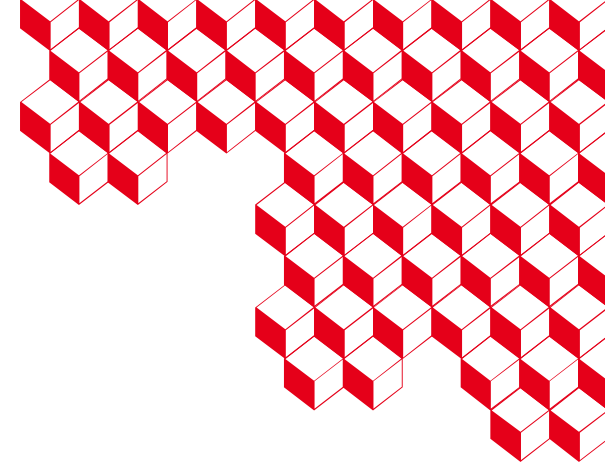
Use of machine learning to go further into the use of FIFRELIN (link between theoretical and experimental fields)

The research of precision, permit to feed both application and fundamental research !





# Thank you for your attention



**A. Chebboubi, G. Kessedjian, O. Serot, D. Bernard, V. Vallet, O. Litaize, S. Julien-Laferrrière, J. Nicholson, V. Piau, M. Houdouin-Quenault, S. M. Cheikh, F. Géhin, G. Bazelaire**

CEA, DES, IRESNE, DER, SPRC, LEPH, Cadarache, F-13108 Saint-Paul-lès-Durance

**C. Sage, O. Méplan, M. Ramdhane, A. Vieville**

LPSC, Université Grenoble-Alpes, CNRS/IN2P3, F-38026 Grenoble, France

**U. Köster, A. Blanc, Y. H. Kim, J-M. Daugas, P. Mutti, C. Michelagnoli**

Institut Laue-Langevin, F-38042 Grenoble, France

**M. Diakaki**

National Technical University of Athens, Athens, Greece

**S. Oberstedt, A. Göök**

European Commission, Joint Research Centre (JRC), 2440 Geel, Belgium