Study of the fission dynamics via Isomeric Ratio Measurements at Lohengrin

-

Development of a new Gas Filled Magnet spectrometer within the FIPPS project

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Outline

- Nuclear fission and usefulness of nuclear data for applications and fundamental physics
- Development of a new spectrometer : Gas Filled Magnet (GFM)
  - Properties of a GFM : experimental outcome
  - Comparison with a Monte Carlo Calculation
- Isomeric Ratio measurements at Lohengrin (ILL)

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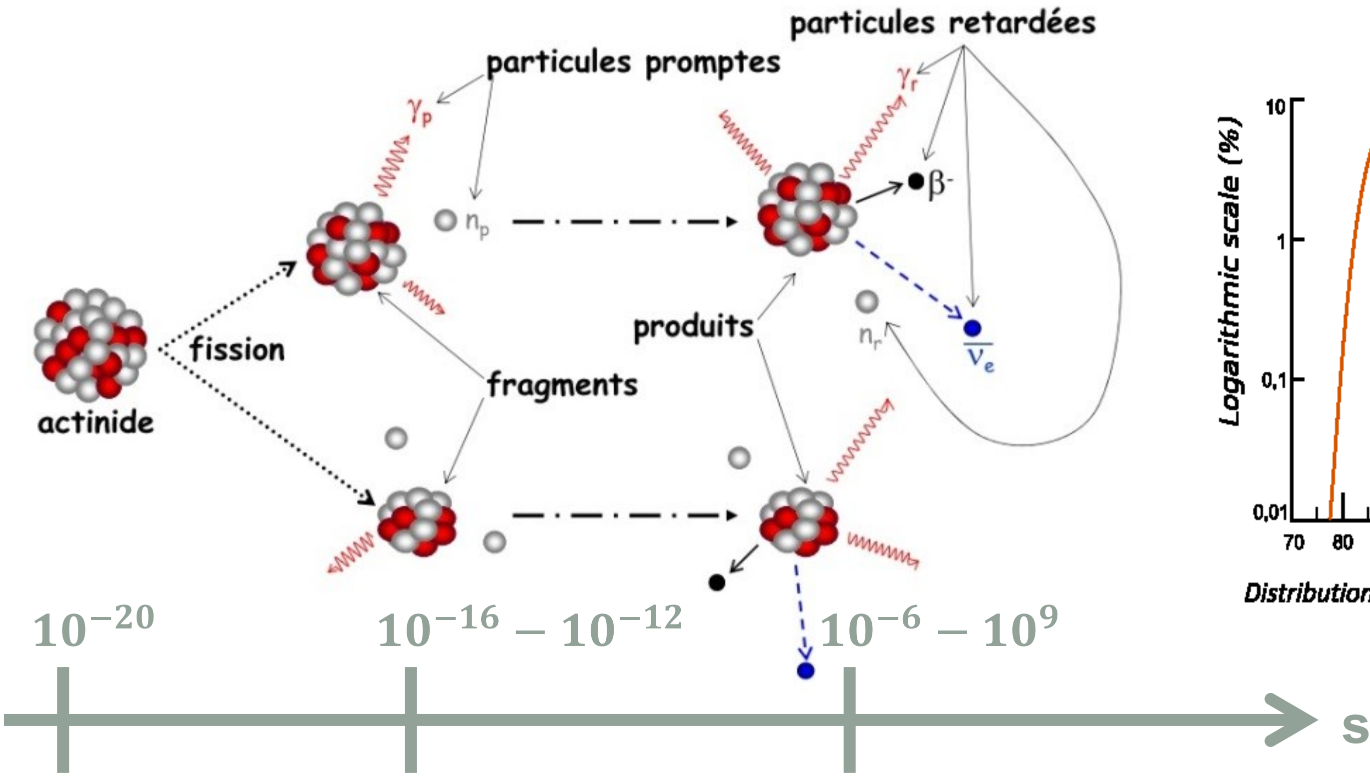
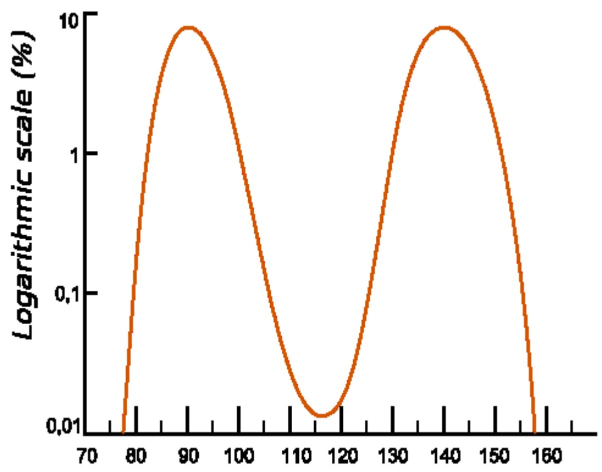
Nuclear Fission Process





Distribution of Uranium-235 fission products (A)

Discovered by Hahn, Strassman and Meitner in 1938 → Chemistry Nobel Prize 1944

A heavy nucleus is broken into two lighter fragments. Emission of a few particles (neutron, γ)

Context of the fission yield studies

➤ Impact of fission yields on the current and innovative fuel cycles

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

- Inventory of spent fuel
- Isotopic composition → Residual power
- Radiotoxicity of spent fuel

} $Y(A, Z)$

- Modeling prompt particle emission (n/γ)
  → foreseen material damage/heating
  in reactor studies

} $P(E^*, J^\pi)$

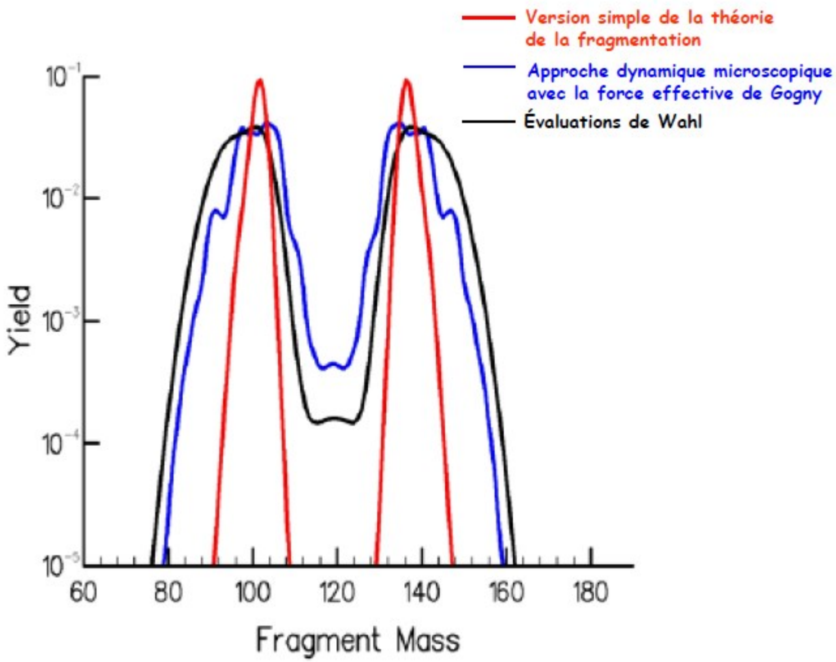
Context of the fission yield studies

$$Y(A,Z,E,J^{\pi}) = Y(A,Z) \times P(E,J^{\pi})$$

➤ Measurements for fission process study

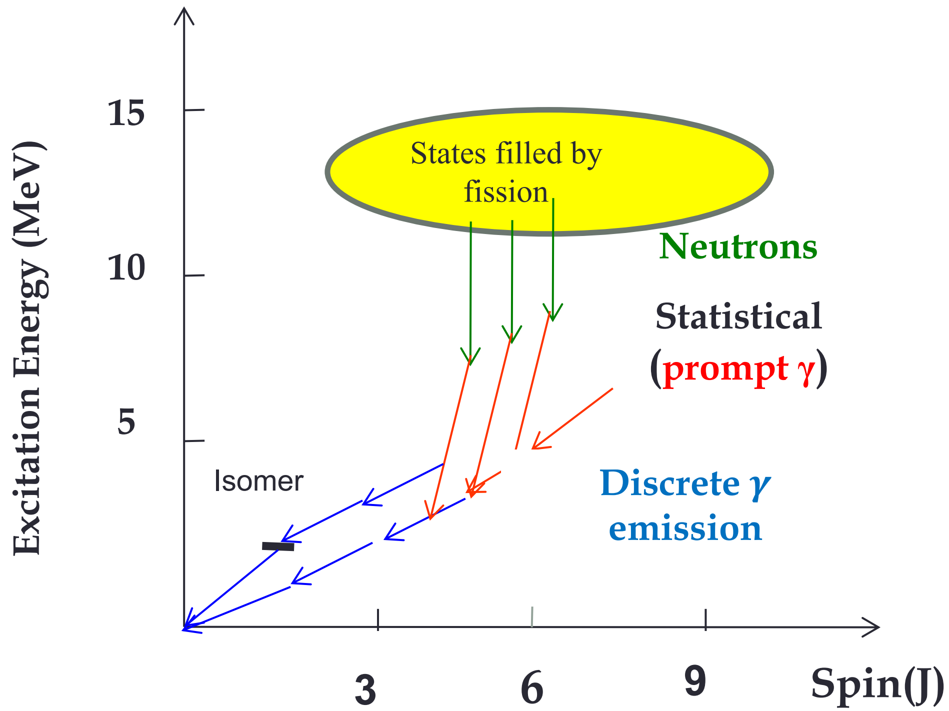
- Improving the predictive power of fission models is necessary for the evaluations at different neutron energies → $Y(A,Z,E,J^{\pi})$
- Lack on dynamical aspect for fission process modelisation
- Inconsistency between Models or evaluations and Experiments for heavy fragments and symmetric region

Disagreement between fission models and experimental data assessment



How to measure Spin Distribution ?

$$Y_{th}(A,Z,E^*,J^\pi) = Y(A) \times P(Z)\Big|_A \times P(E^*)\Big|_{A,Z} \times P(J^\pi)\Big|_{A,Z,E^*}$$



To study Spin distribution, look at $\gamma_{prompt}, n_{prompt}$ structure effect at low excitation energy : Isomeric Ratio

Isomer : state with longer half-life than neighboring states

How to measure Spin Distribution ?

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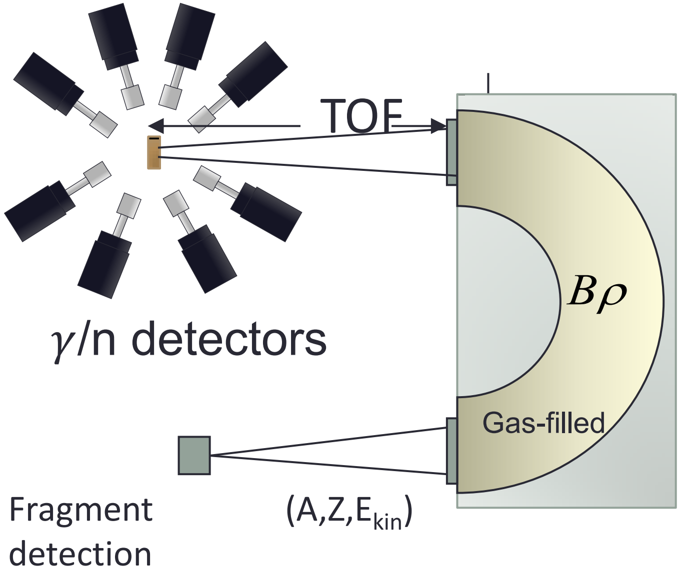
$$P\left(\frac{N_{isomer}}{N_{GS}}\right)\Bigg|_{A,Z,\overline{E_k}} \qquad P(\gamma_{prompt})\Big|_{A,Z,\overline{E^*}}$$

- **Isomeric ratio** : Recent measurements on Lohengrin

Min/s Isomers :
$^{136}I, ^{132}Sb, ^{130}Sb, ^{129}Sb, ^{129}Sn, ^{99}Nb, ^{98}Y$

µs Isomers :
$^{136}Xe, ^{132}Te, ^{129}Sb, ^{99}Y, ^{98}Y, ^{94}Y, ^{88}Br$

ns Isomers :
Almost all isotopes in heavy mass region



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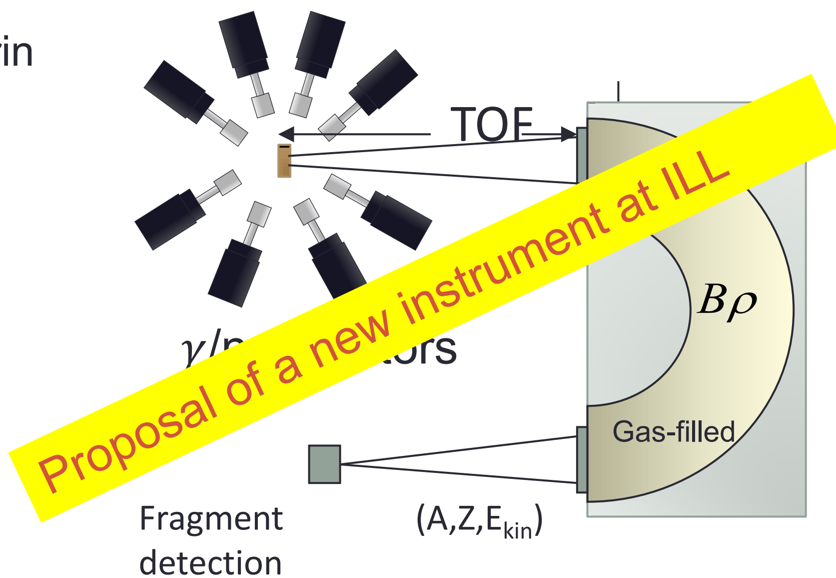

- **FIPPS (Fission Product Prompt gamma-ray Spectrometer) Goals** :
  - Direct measurement of prompt particles ($\gamma_{prompt}, n_{prompt}$)
  - Fission product spectroscopy (astrophysics interest)
  - Neutron emission
  - Short lifetime isomers (ps,ns)

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  - **Properties of a GFM : experimental outcome**
  - **Comparison with a Monte Carlo Calculation**

- Isomeric Ratio measurements at Lohengrin (ILL)

Back to the present : Lohengrin limits

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

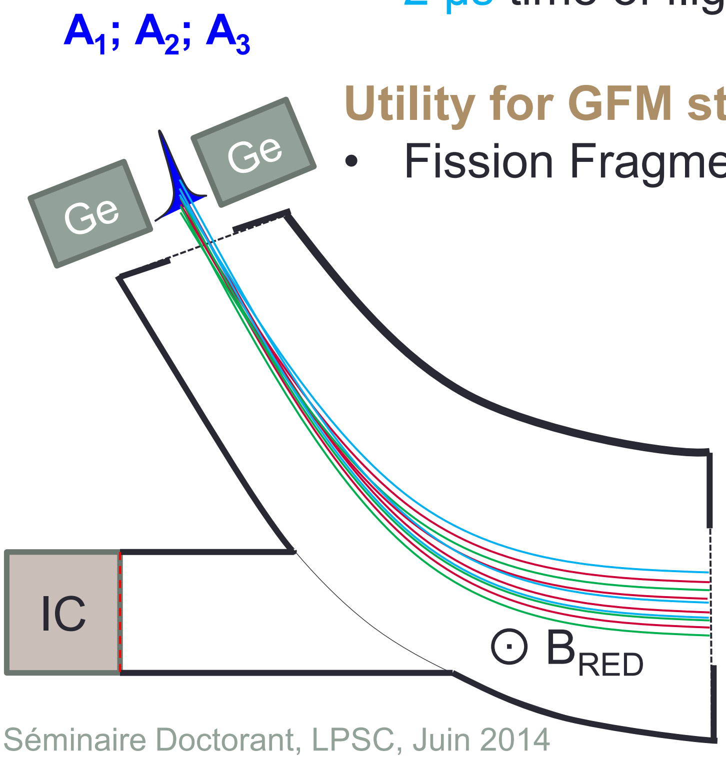
$$(A_1,E_1,q_1) \equiv (A_2,E_2,q_2) \equiv (A_3,E_3,q_3)$$

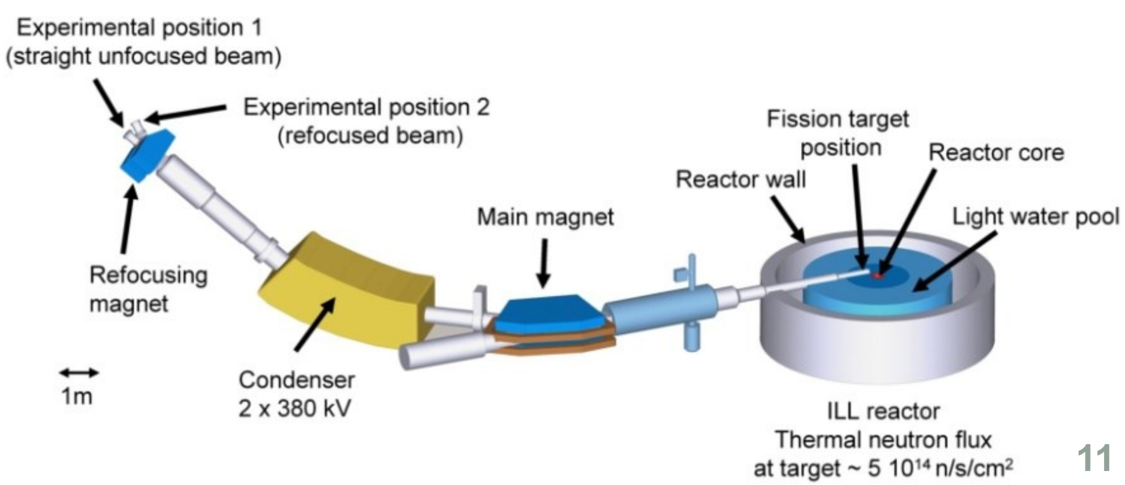
Limits :

- 2 µs time of flight (TOF) → no prompt particle study (10^{-16} s)

Utility for GFM study :

- Fission Fragment Source !!





Experimental Setup for GFM study

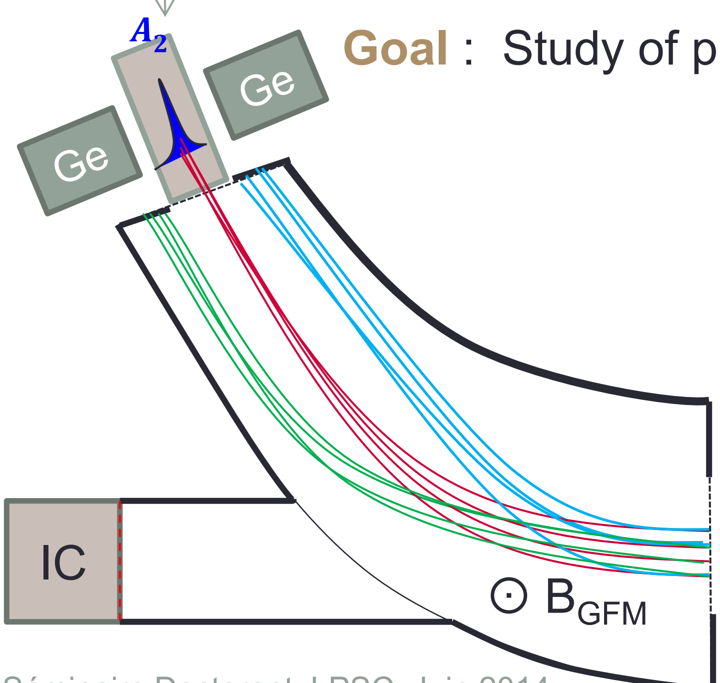
Setup : The RED magnet is now filled with various gases → GFM

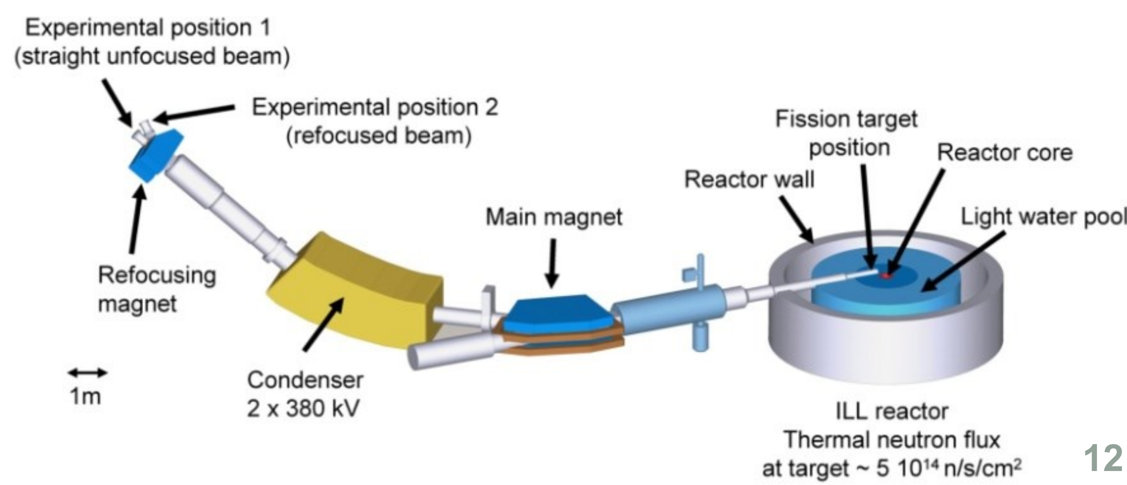
GFM : Spatial dispersion of fission fragments according to the mass A and Nuclear charge Z

$$\begin{cases} B\rho \propto A \left. \left\langle \dfrac{v(z)}{q(Z)} \right\rangle \right|_{P,Gas} \\ B\rho \propto \dfrac{A}{Z^{\frac{1}{3}}} \; [1] \end{cases}$$

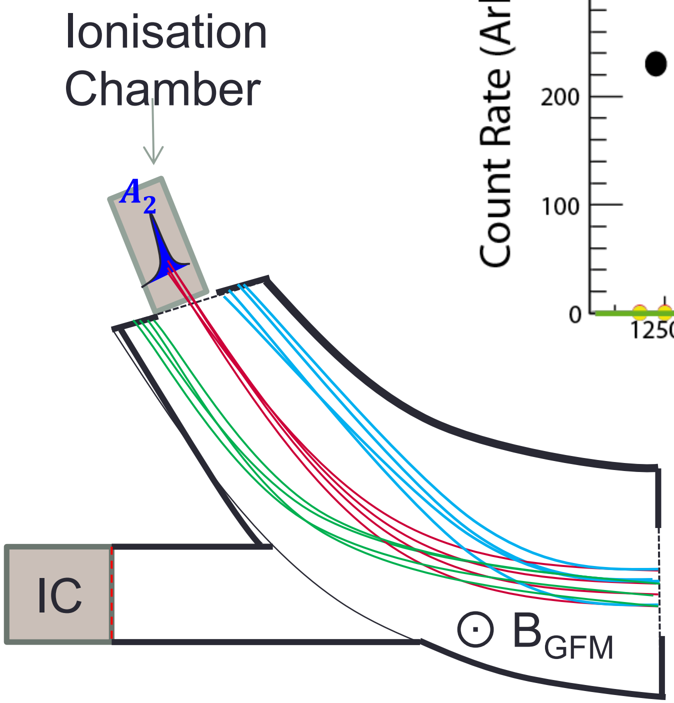
Goal : Study of properties of this device / feasibility

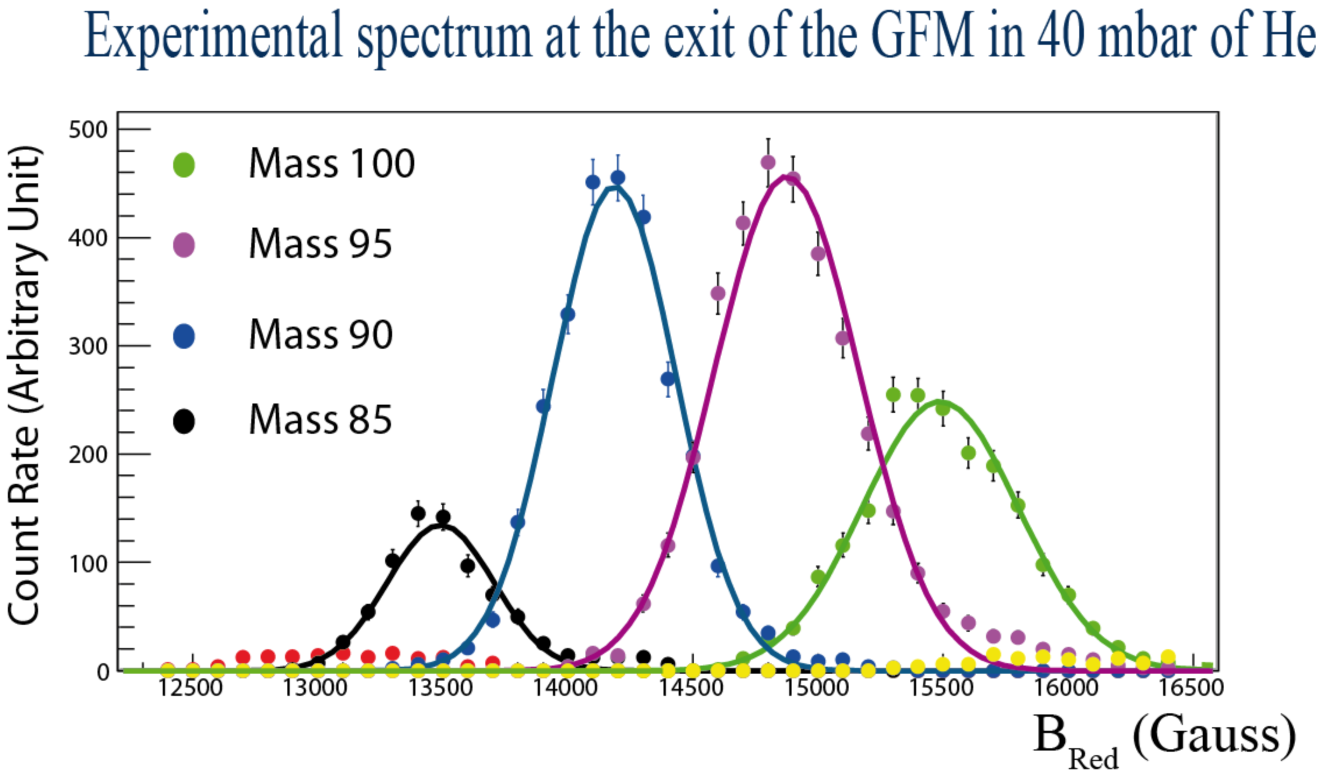
Ionisation Chamber





GFM separation power

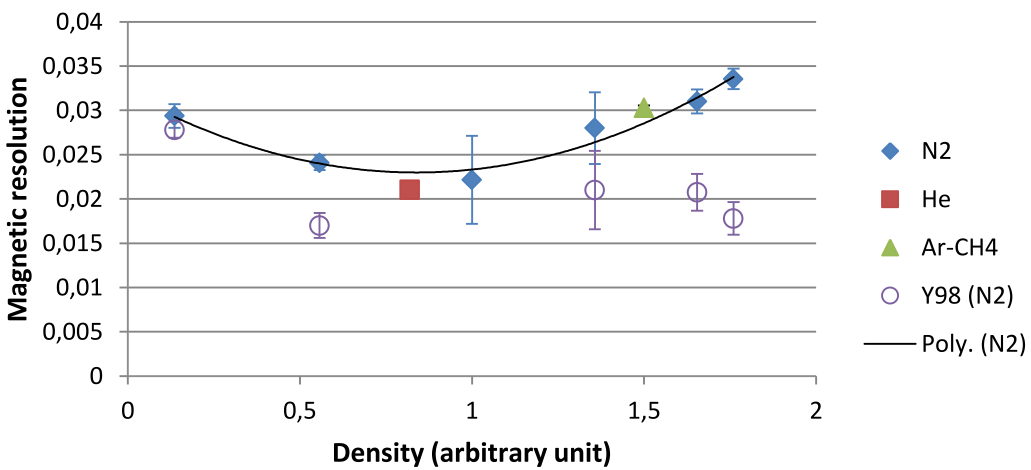




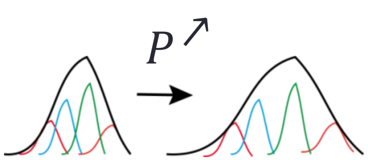
Gas Comparison : Experimental results

Gas	Pressure	Electronic density ratio	Resolution
N_2	7 mbar	1	2,2 %
He	40 mbar	0,82	2,1 %
$Ar - CH_4$	8,5 mbar	1,5	3,0 %

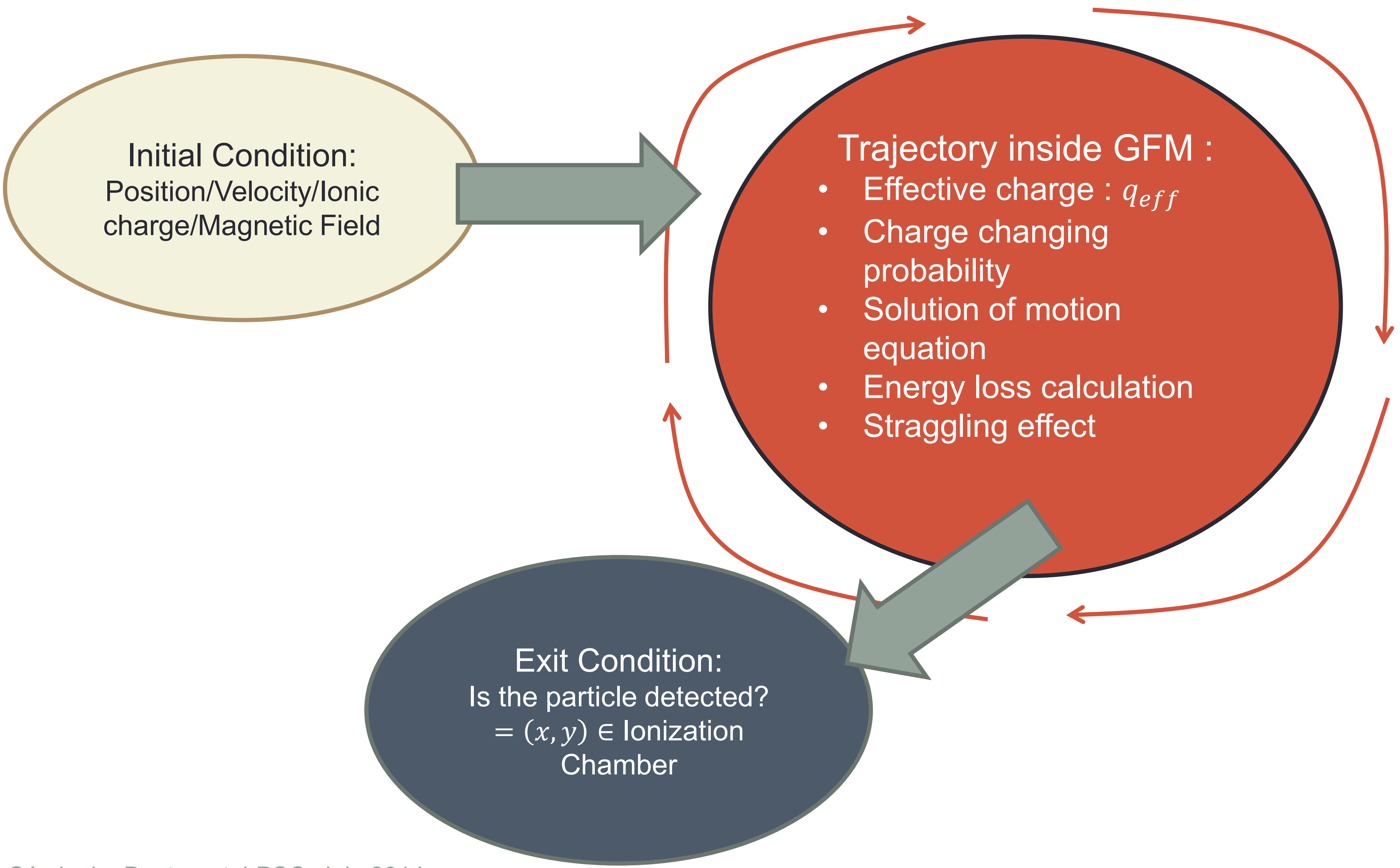
Evolution of the magnetic resolution with density in several gases for A=98



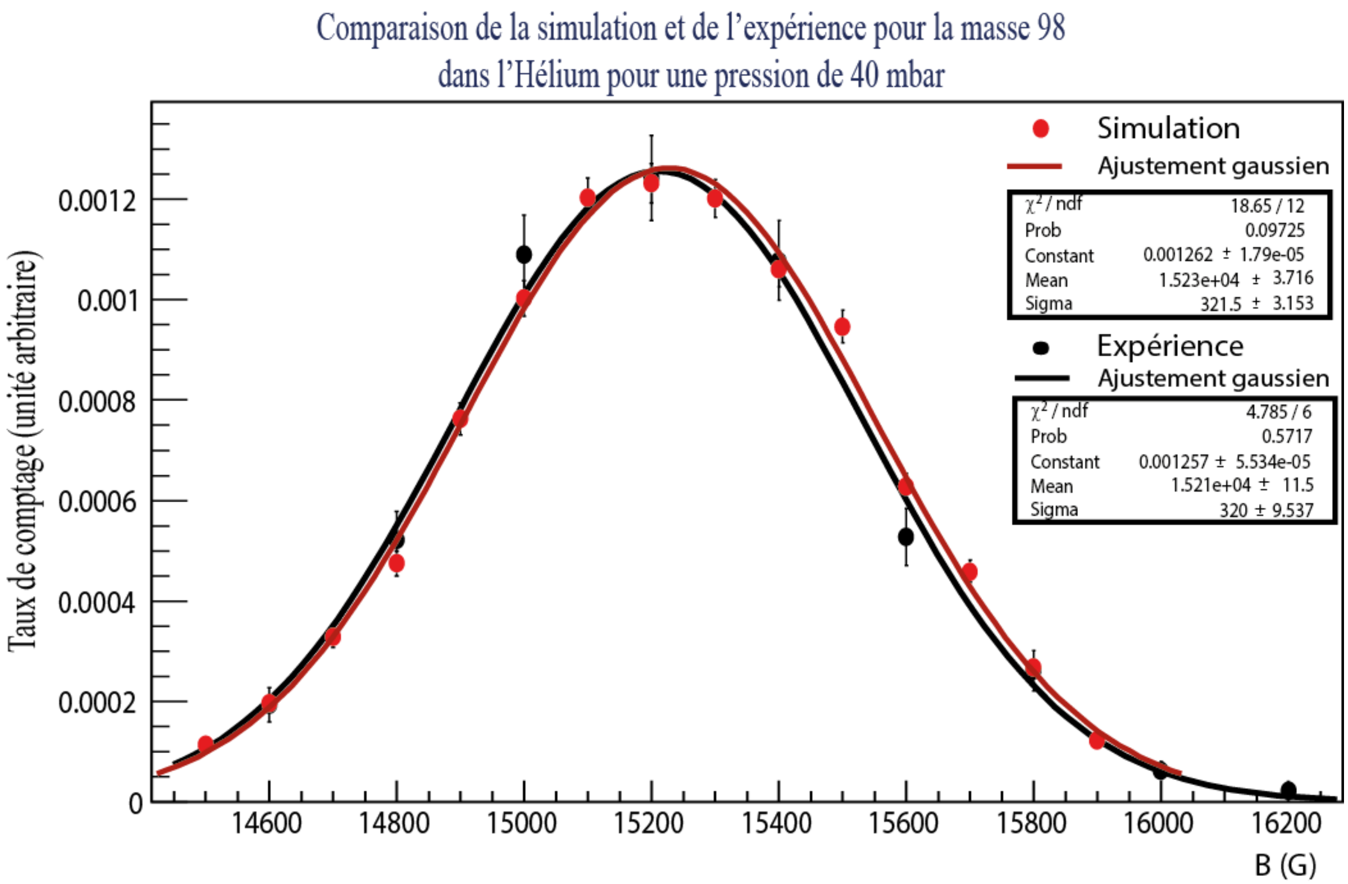
$$Density = \frac{\rho_{e^-}}{\rho_{e^-}(N_2, P = 7mbar)}$$



Monte Carlo Calculation Architecture



Gas Filled Magnet : What to compare

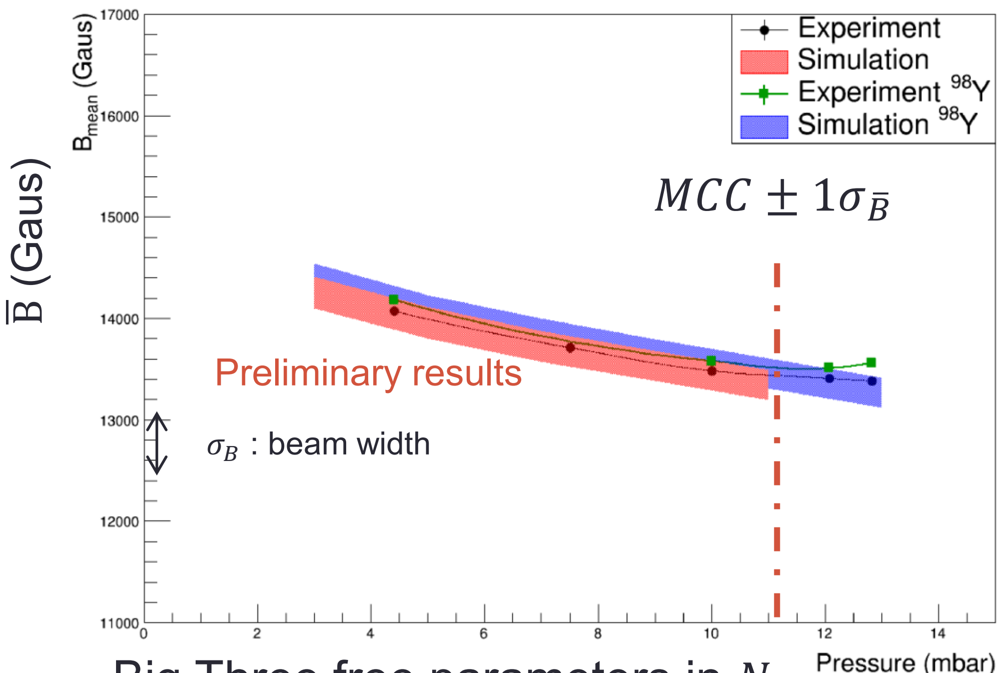


We will compare $\bar{B}$ along the pressure

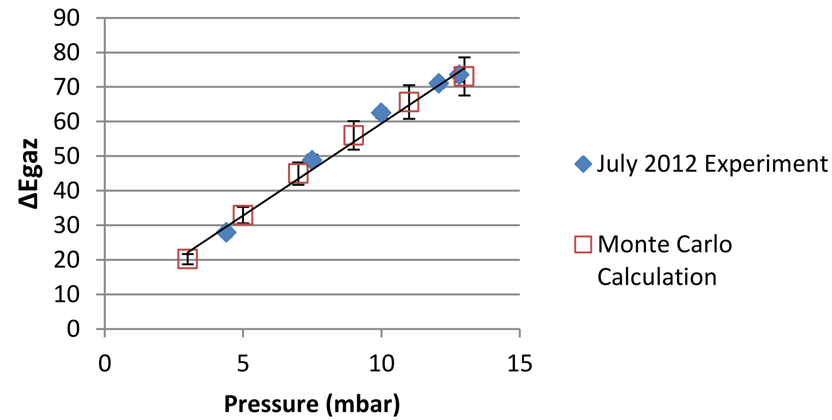
N_2 Gas Filled Magnet : Test

^{98}Y & A=98 / E=90MeV / ^{233}U

Magnetic Field : pressure variation inside N_2



Energy loss in GFM for Mass 98 @ E=90MeV

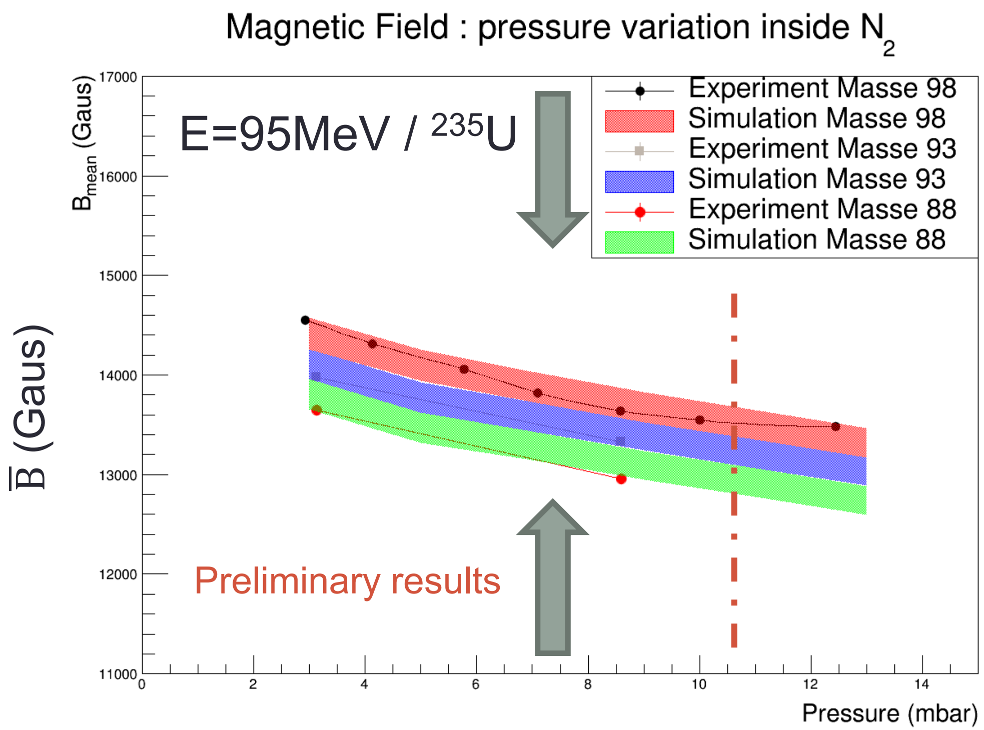


Big Three free parameters in N_2

- $k = \frac{\sigma_{2e^-}}{\sigma_{e^-}} = 0{,}56 \pm 0{,}05$
- $q_{eff} = q_{Betz} + \beta \ln\left(\frac{\rho}{\rho_0}\right) \rightarrow \beta = -0{,}4$
- $\Delta P = \mathrm{P_{meas}} - \mathrm{P_{MCC}} = 0{,}5 \pm 0{,}3$ mbar

Set using A=98 & ^{98}Y experimental data

N_2 Gas Filled Magnet : Comparison



- Agreement for pressure below 10 mbar at 1σ
- $k \rightarrow \bar{B}$ shift
- For $P > 10\ mbar$ inconsistent calculation → Density effect on effective charge calculation

⇨ Predictive calculation for light mass at 1σ

⇨ Design the new spectrometer (find the better geometry)

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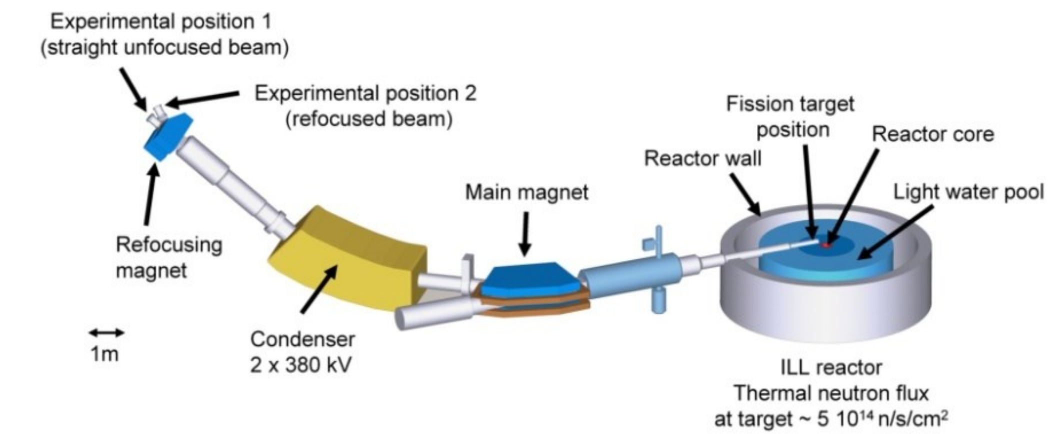
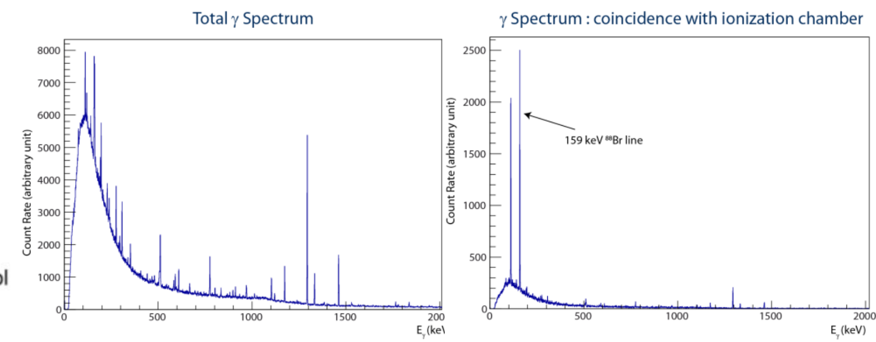
Experimental Set-Up : µs Isomer





Coincidence between ionization chamber and γ detectors

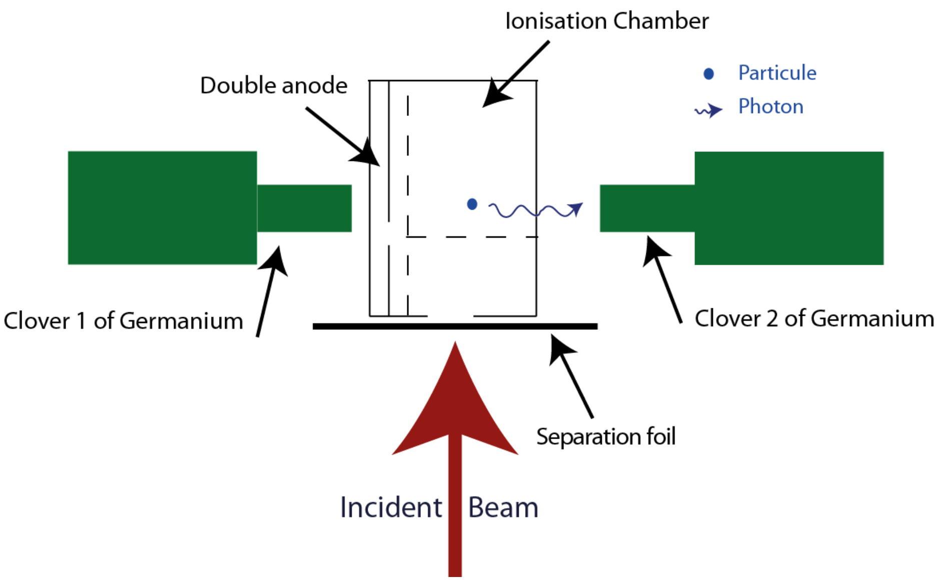
$$\Delta T_{Gate} = 10 T_{1/2}$$

→ Isomeric state measurement

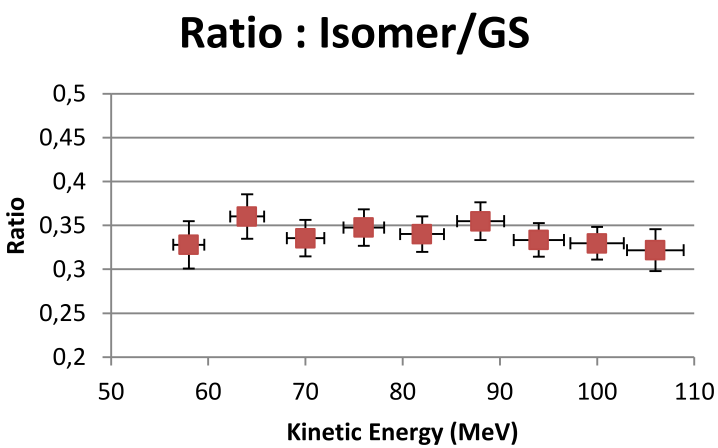
Total γ spectrum → GS measurement. More difficult

For long live GS, loss by gas pumping → correction needed

$$\lambda_{Gas} \ll \lambda_{\beta}$$

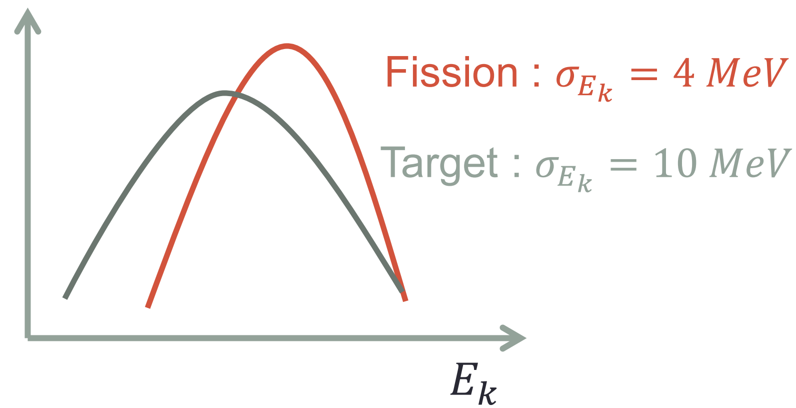


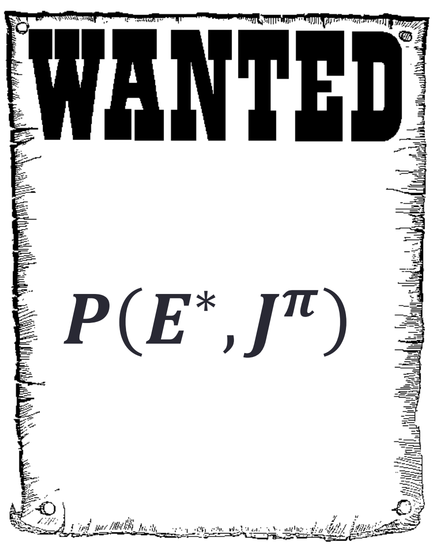
^{88}Br Results

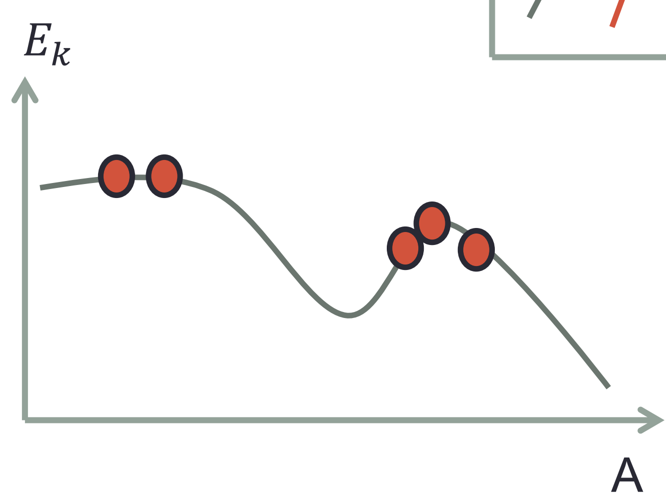


Isomeric Ratio :

$$\overline{RI(E_k)} = \frac{N_f(^{88m}Br)}{N_f(^{88m}Br)+N_f(^{88}Br)} = 0{,}34 \pm 0{,}007$$



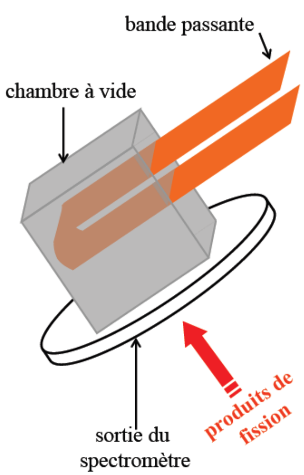




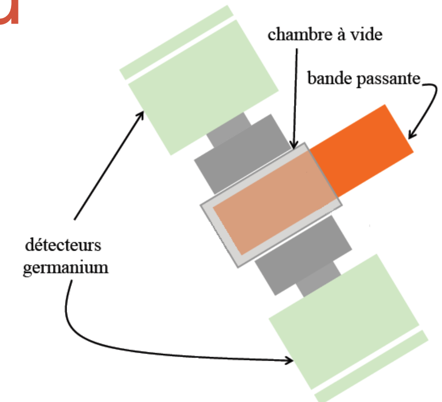
$\overline{E^*} = f(\overline{E_k})$: models

Measurement → $P(\overline{E^*}, J^\pi)$

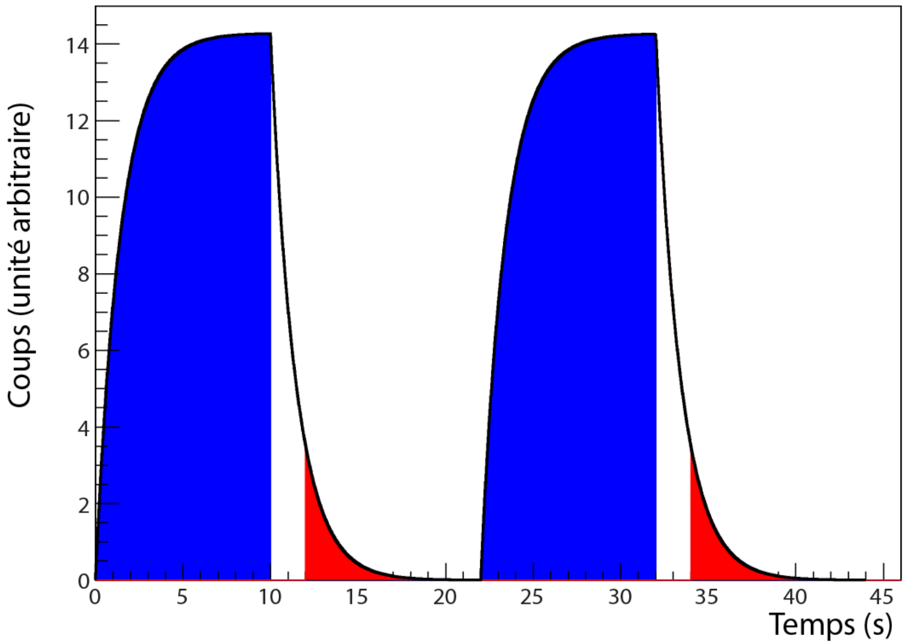
Beam Cut Method



Side view



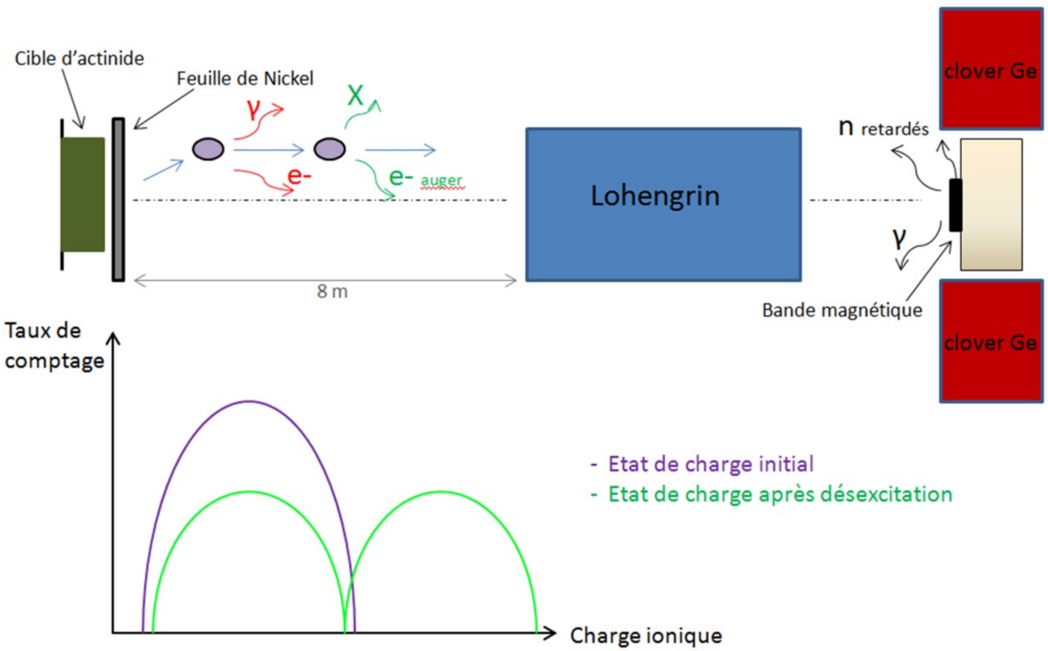
Up view

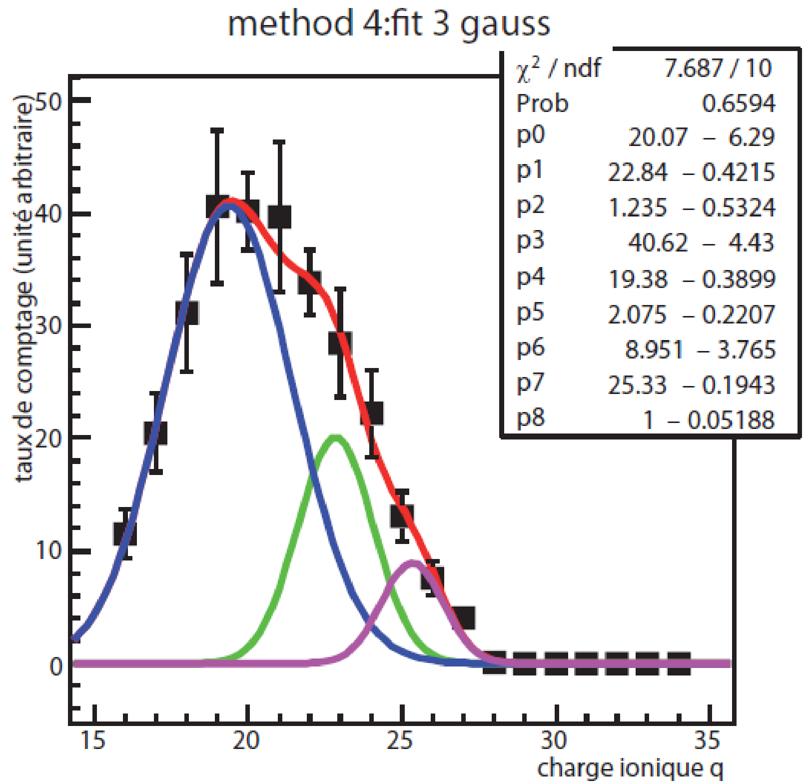


Masse	Isotope	Period	γ
136	^{m}I I Xe	46,9 s 83,4 s 2,95 µs	**197/381/1313**/369 **1313**/1321/2289/ **197/381/1313**
130	^{m}Sb Sb	6,3 m 39,5 m	**839/793/182**/1017 **793/839**/330/**182**
132	^{m}Sb Sb Te	4,10 m 2,79 m 28,1 µs	**974/697/103/150** **974/697/103**/989 **974/697/103/150**

- Allow to separate isomeric state and GS feeding from a same γ line

Nanosecond Isomer





Nikolaev and D. (1968) (2)

18,68126485 1,96827205

Structure effect will deform the ionic charge distribution

Conclusion and Perspectives

GFM studies

- Nowadays :
  - Spectrometer power is bound by the atomic collision in gas $\frac{\sigma_B}{B} = \frac{\sigma_A}{A} \geq 1{,}5\% \; (\frac{A}{\sigma_A} = 66)$
  - Good understanding of physics inside a GFM
- Future:
  - Benchmark between MCC and Geant4
  - Design the FIPPS spectrometer with a validated code
  - Experiment with different gas

Fission yields and spin distribution

- New methods has been developed
- New observables
- Goals : Improvement of fission models

- **Future :**
  - Comparison with theoretical code
  - Analysis …
  - New Experiment

Thank you for you attention

Any questions?

Backup

Context of the fission yield studies

➤ Impact of fission yields on the current and innovative fuel cycles

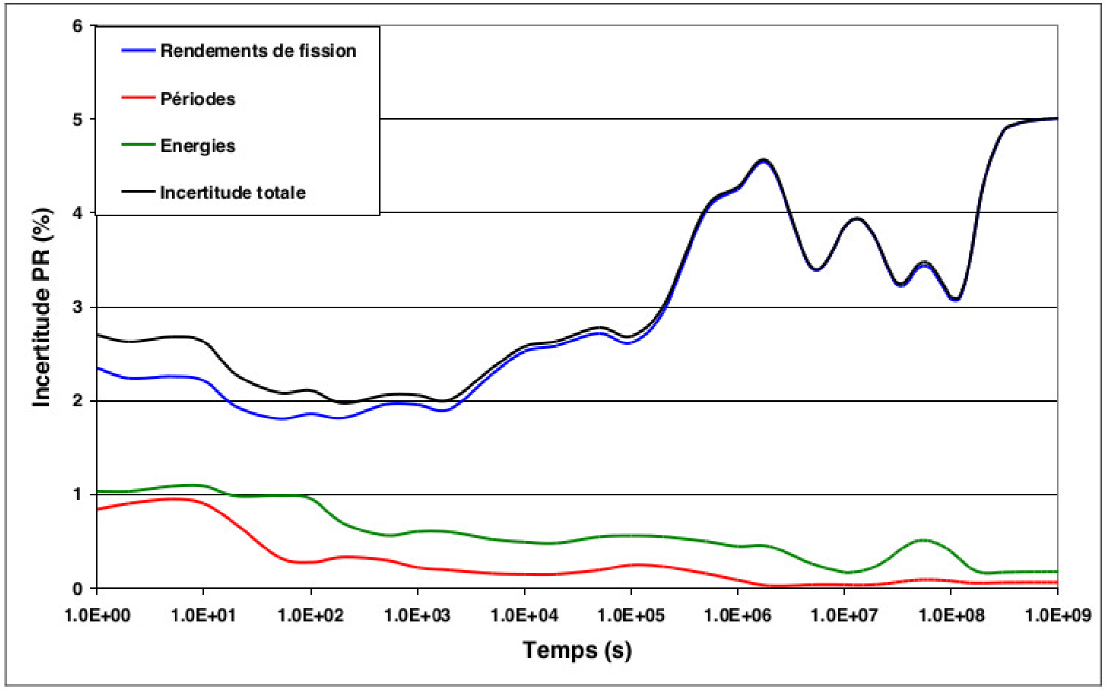
$$Y(A,Z,E^*,J^\pi) = Y(A,Z) \times P(E^*,J^\pi)$$

- Inventory of spent fuel

Isotopic composition → Residual power $RP(t) = \sum_Z Y(A,Z)E_\beta(A,Z)\lambda(A,Z)$

- Radiotoxicity of spent fuel

Uncertainty of residual power calculation come from the uncertainty of fission yields $Y(A,Z)$



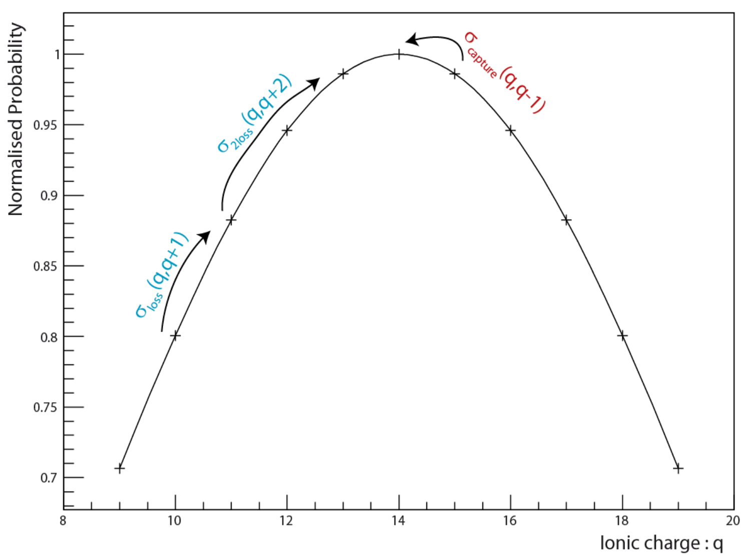
Model involved into the MCC

- Atomic collision model at each step x according to M.Paul & H.Betz :
  - Equilibrium charge state distribution : $F(q) \propto \exp\left(-\frac{(q-\bar{q})}{2d^2}\right)$
  - σ_{tot}^{capt} using experimental measurements : with F(q) we can define A_c, b_c, A_l, b_l
  - Total Loss cross section : $\sigma_{tot}^{loss}(q) = A_l \exp\left(-b_l(q-\bar{q})\right)$
  - Total Capture cross section : $\sigma_{tot}^{capt}(q) = A_c \exp(b_c(q-\bar{q}))$

  - Multiple Capture cross section : negligible
  - Multiple Loss cross section : $\sigma_{2l} = k_0\sigma_l$ (Heavy gas)
    - For N_2, $\boldsymbol{k_0 = \frac{\sigma_{2l}}{\sigma_l} \approx 0,5}$

  - The probability for charge changing through a distance x,
    - $P(q,q') = \frac{\sigma(q,q')}{\sigma_{total}}(1-e^{-\sigma_{total}\rho x})$

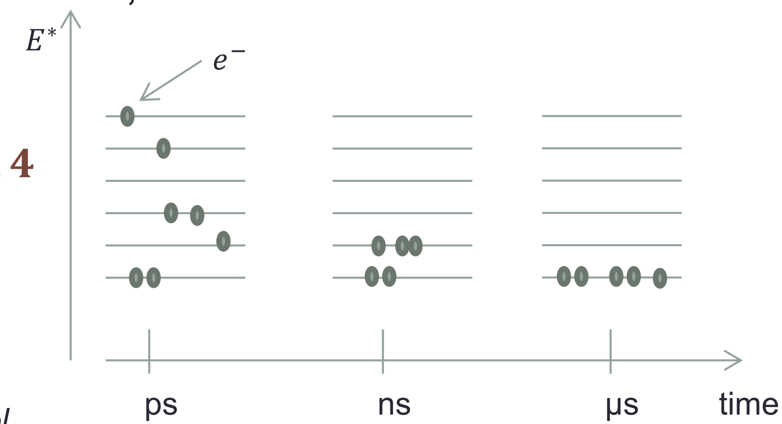
Equilibrium charge state distribution



M. Paul et al, *Nuclear Instruments and Methods in Physics Research A, vol. 277, pp. 418-430, 1989*
H.D. Betz, *Reviews of Modern Physics, vol.44, n° 3, p. 465, July 1972.*

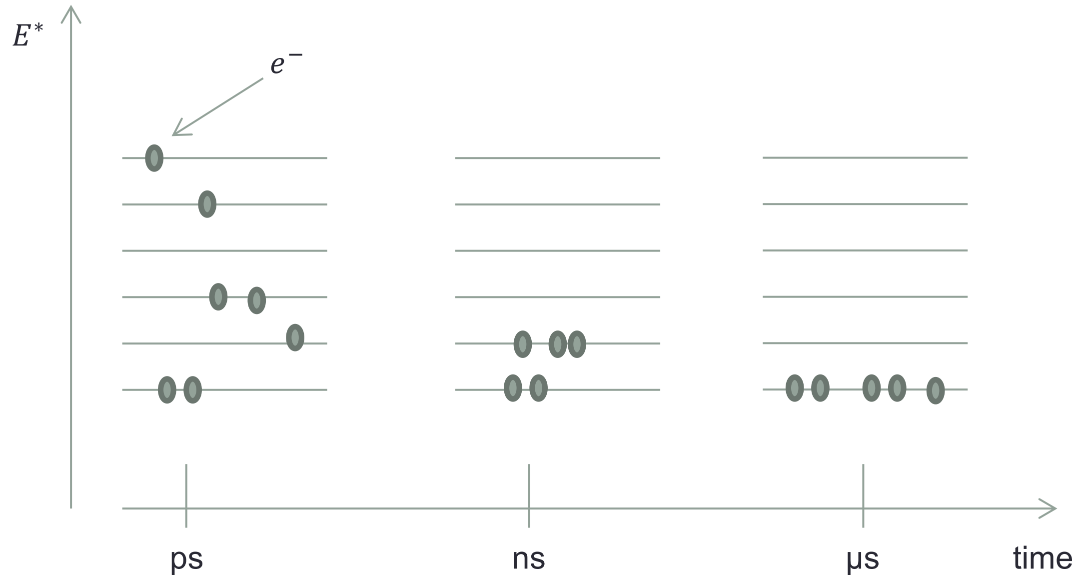
Model involved into the MCC

- According to Pierce, the energy loss,
  - $\left(\frac{dE}{dX}\right)_{Z_{ion},A,E} = \frac{q_{eff}^2}{\gamma_p^2} \times \left(\frac{dE}{dX}\right)_{p,\frac{E}{A}}$ screening effect
- According to Betz,
  - $q_{eff} = Z_{ion}\left(1 - C \times \exp\left(-\frac{v_{ion}}{v_0 Z^{\delta}}\right)\right)$
  - $C = 1{,}032$; $\delta = 0{,}69$; $v_0 = 2{,}19.10^{-8}\ cm.s^{-1}$: Bohr velocity
  - F(q) is symmetric → $\boldsymbol{q_{eff} \equiv \bar{q}}$
- According to Gregorich, the charge density effect,
  - $\Delta\bar{q} \propto \ln\left(\frac{\rho}{\rho_0}\right) \propto \ln\left(\frac{P}{P_0}\right)$
    - $\boldsymbol{q_{eff} = q_{effBetz} + \beta \times \ln\left(\frac{P}{P_0}\right)}$ **with** $\boldsymbol{\beta = -0,4}$
- Straggling effect : Molière potential



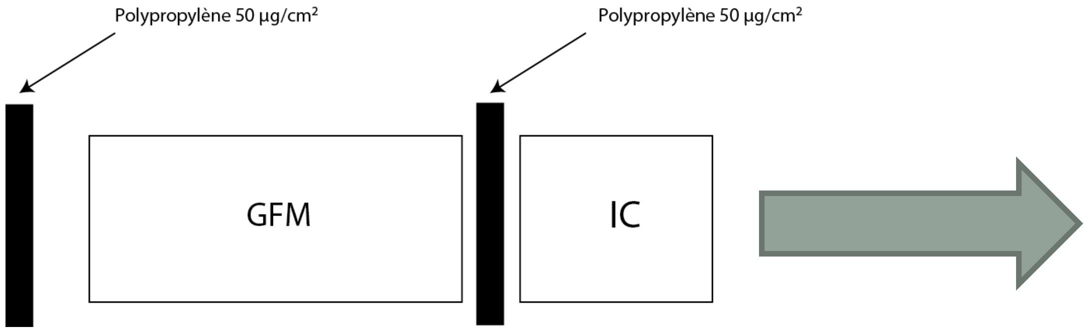
Pierce et al, Physical Review, vol 173, number 2, pp 390-405, 1968
K.E. Gregorich, *Nuclear Instruments and Methods in Physics Research A, vol. 711, pp. 47-59, 2013.*

Charge Density Effect



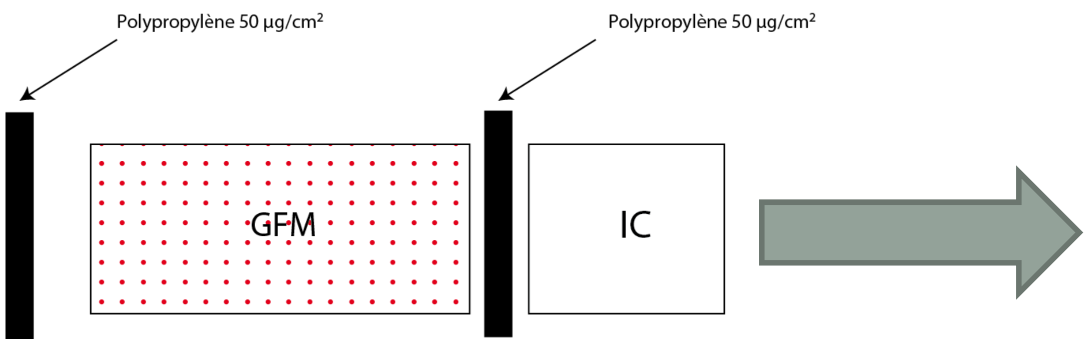
After a capture or a loss of an e^-, the ion can be excited, The relaxation time can be longer than the collision time.
Cross section of an excited state $\neq$ Cross section of the GS

Measurement of the Energy Loss in the GFM



Calibration : Input SRIM inside foils

$$E_{\mathrm{final}} = E_{init} - \Delta E_{foil1}(E_{init}, Z, A) - \Delta E_{foil2}(E_{init} - \Delta E_{foil1}, Z, A)$$

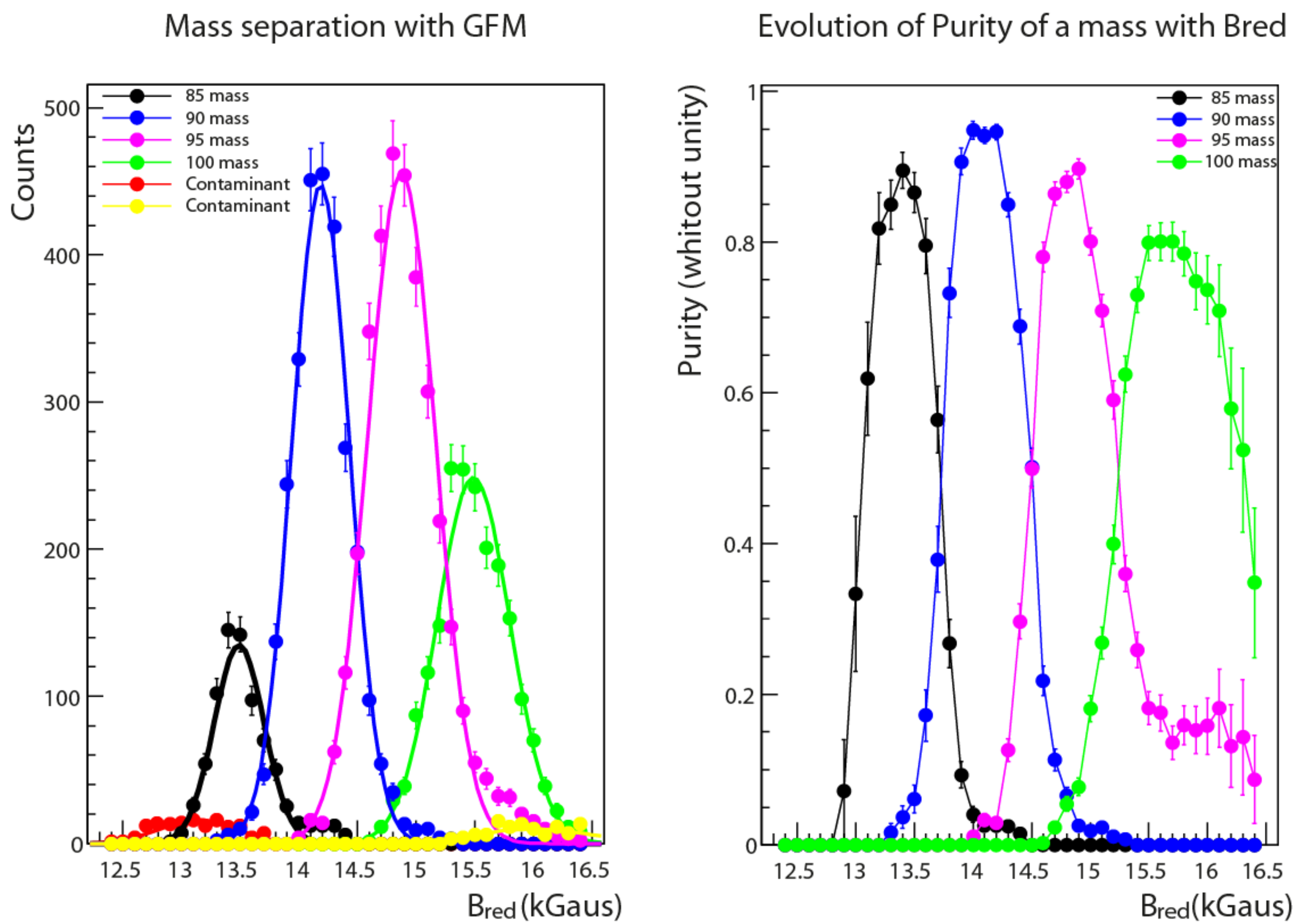


Result : Need SRIM in the second foil

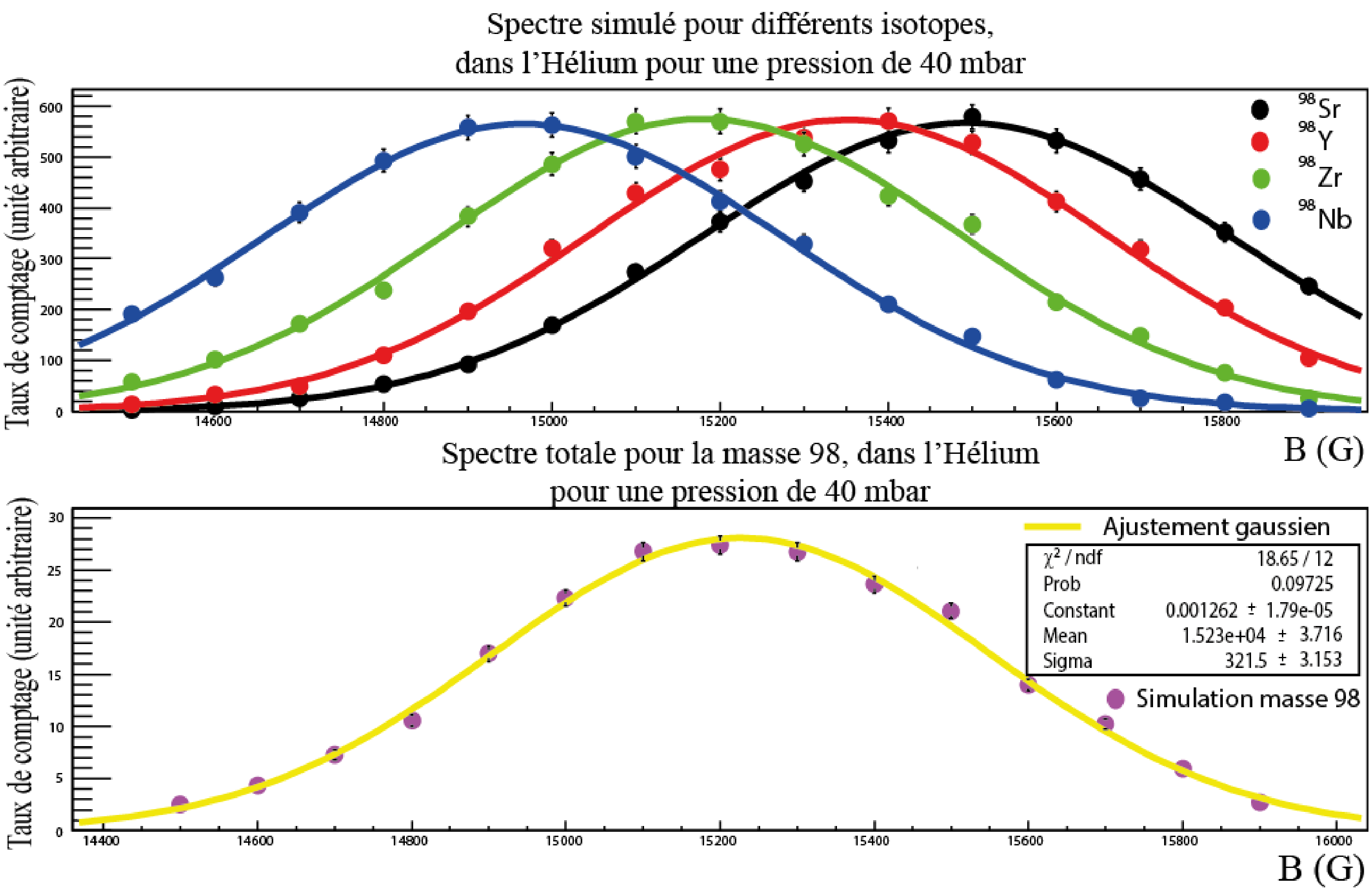
$$E_{\mathrm{final}} = E_{init} - \Delta E_{foil1}(E_{init}, Z, A) - \Delta E_{gas}(E, \mathrm{q_{eff}}, \mathrm{Z}, \mathrm{A}) - \Delta E_{foil2}(E_{init} - \Delta E_{foil1} - \Delta E_{gas}, Z, A)$$

Since we are interested in ΔE_{gas}, the sensitivity of the foil layer is small

GFM @Lohengrin



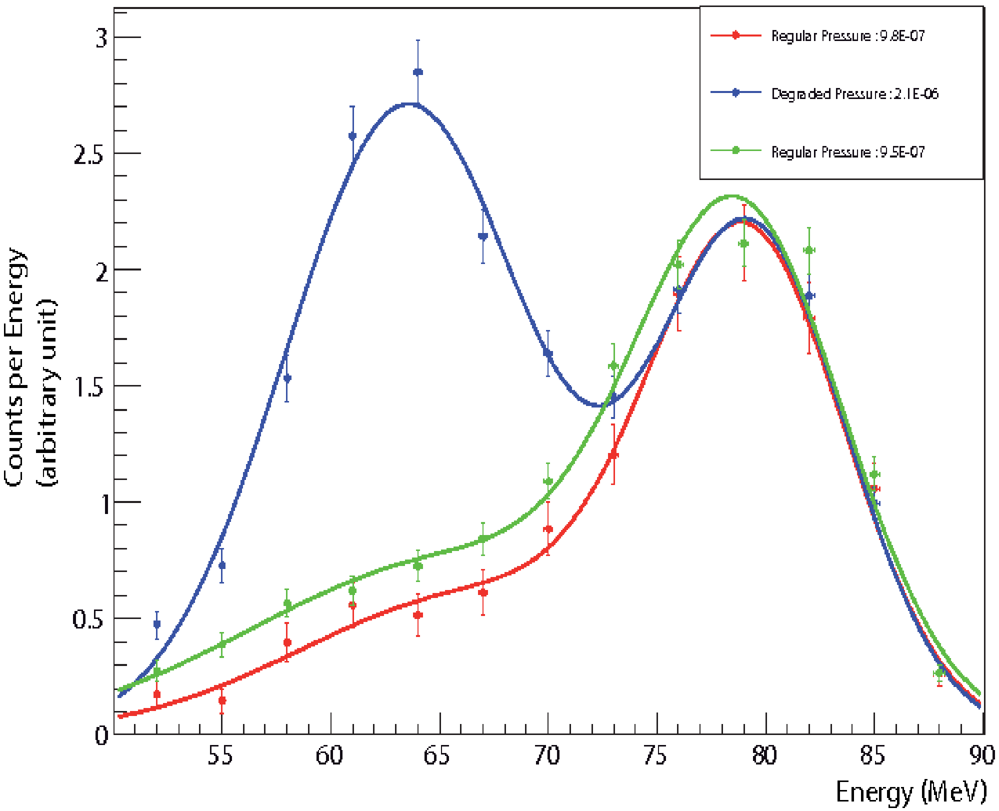
- Isobaric beam could allow to measure the Total β, γ decay energy per mass



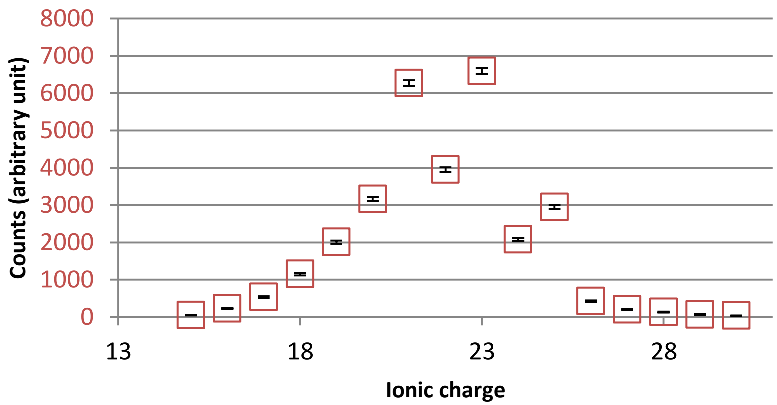
Spectre simulé pour différents isotopes,
dans l'Hélium pour une pression de 40 mbar
Taux de comptage (unité arbitraire)
^{98}Sr
^{98}Y
^{98}Zr
^{98}Nb
B (G)
Spectre totale pour la masse 98, dans l'Hélium
pour une pression de 40 mbar
Taux de comptage (unité arbitraire)
Ajustement gaussien
χ^2 / ndf 18.65 / 12
Prob 0.09725
Constant 0.001262 ± 1.79e-05
Mean 1.523e+04 ± 3.716
Sigma 321.5 ± 3.153
Simulation masse 98
B (G)

Symmetry contamination

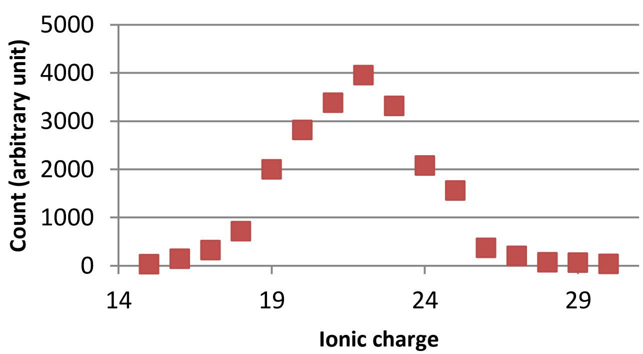
Evolution of the kinetic energy distribution of the 128 Mass with the pressure inside the condenser



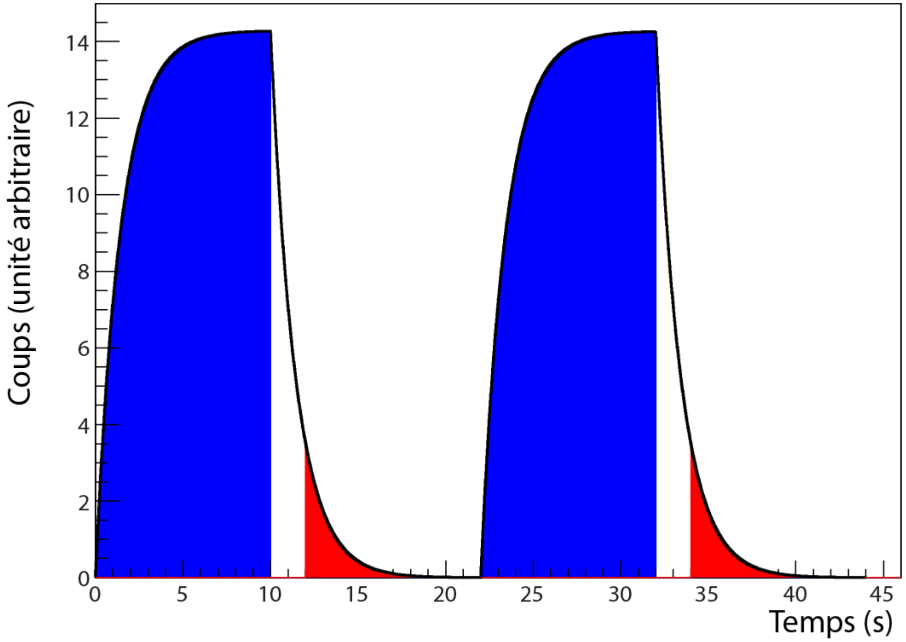
Ionic charge distribution of the mass 124 with an indicator of contamination



Shape of the ionic charge distribution of the mass 124 by removing the artefact



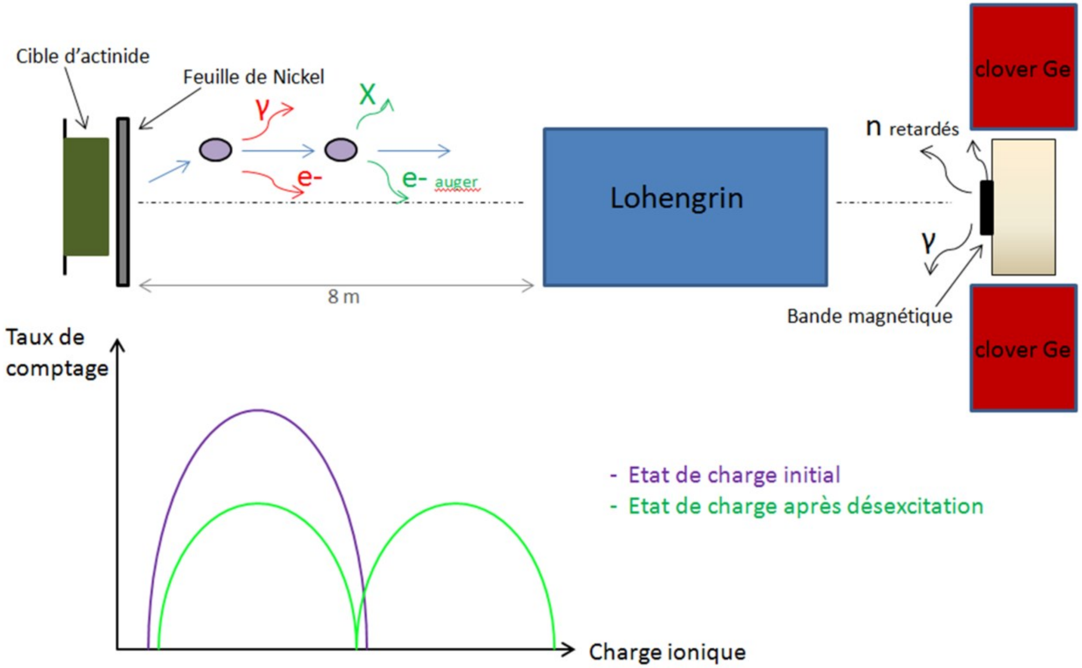
Beam Cut Method



Masse	Isotope	Durée de vie	Raie
136	^{m}I I Xe	46,9 s 83,4 s 2,95 µs	**197/381/1313**/369 **1313**/1321/2289/ **197/381/1313**
130	^{m}Sb Sb	6,3 m 39,5 m	**839/793/182**/1017 **793/839**/330/**182**
132	^{m}Sb Sb Te	4,10 m 2,79 m 28,1 µs	**974/697/103/150** **974/697/103**/989 **974/697/103/150**

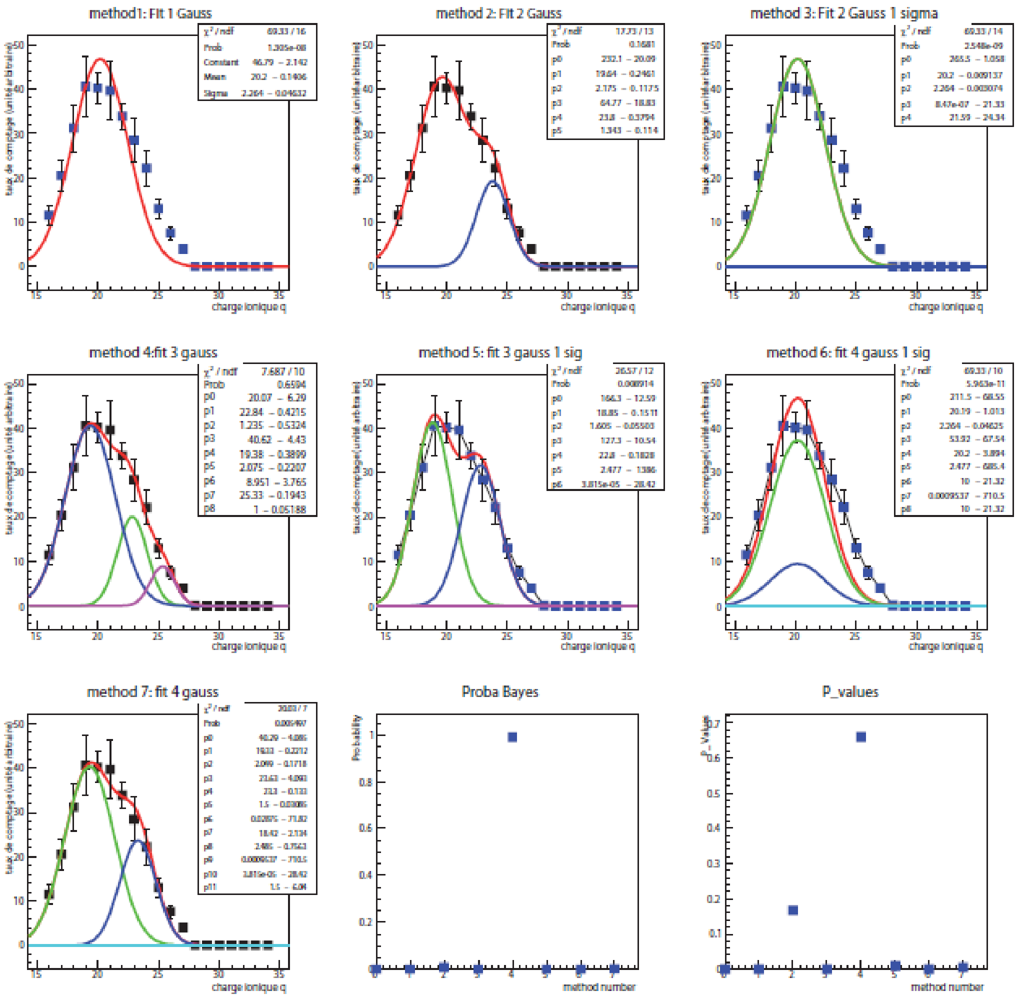
- Temps d'acquisition $\sim 5T_{1/2}$
- Répétition → ↗ stat
- Séparation état métastable/ fondamental (peuplant une même raie)

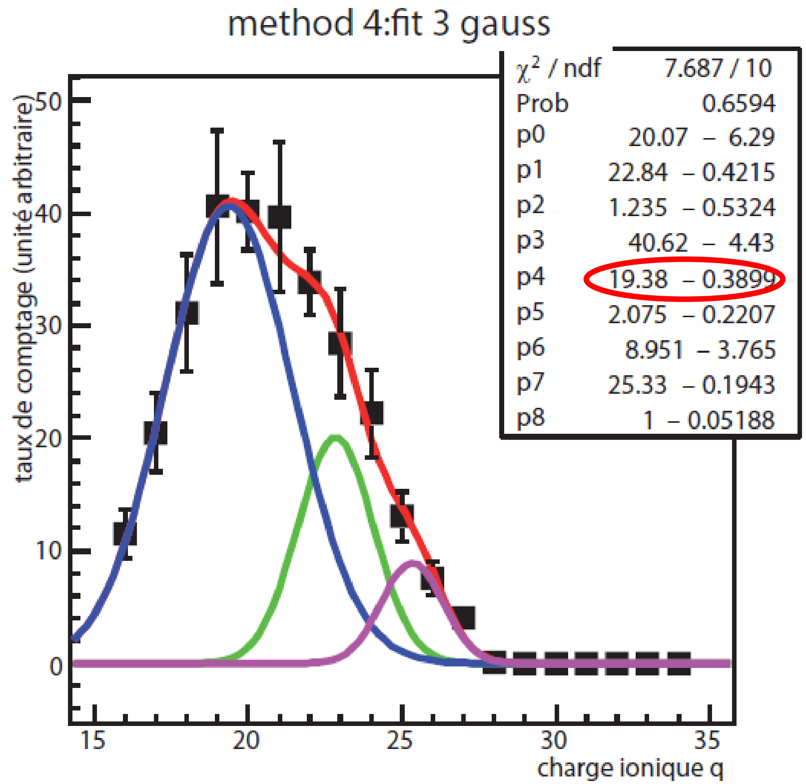
Nanosecond Isomer



- Stabilisation de la charge moyenne dans la cible
- Si nanosecondes isomères :
  - Réarrangement du cortège électronique
  - Distribution en charge plus complexe

Nanosecond Isomer

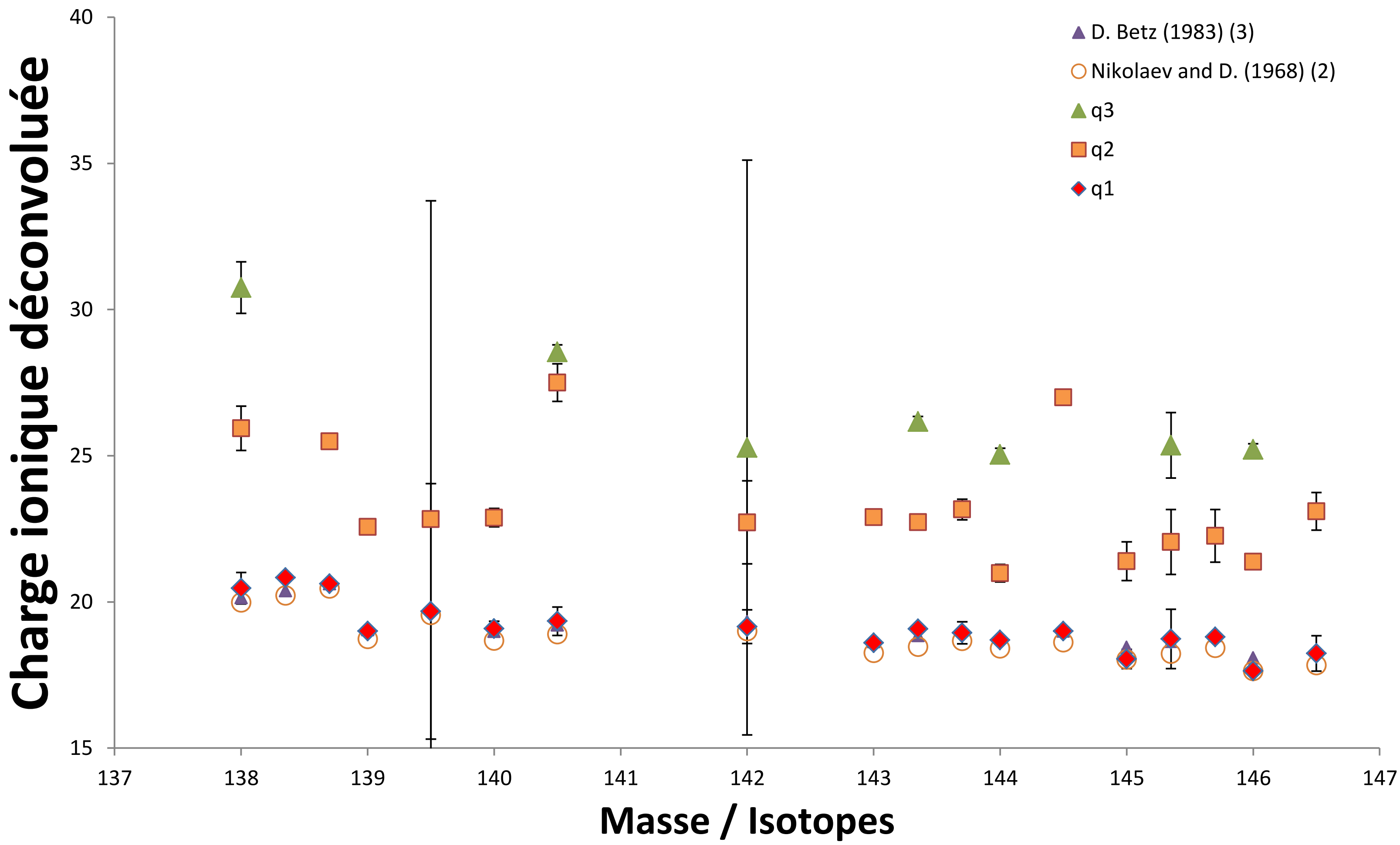




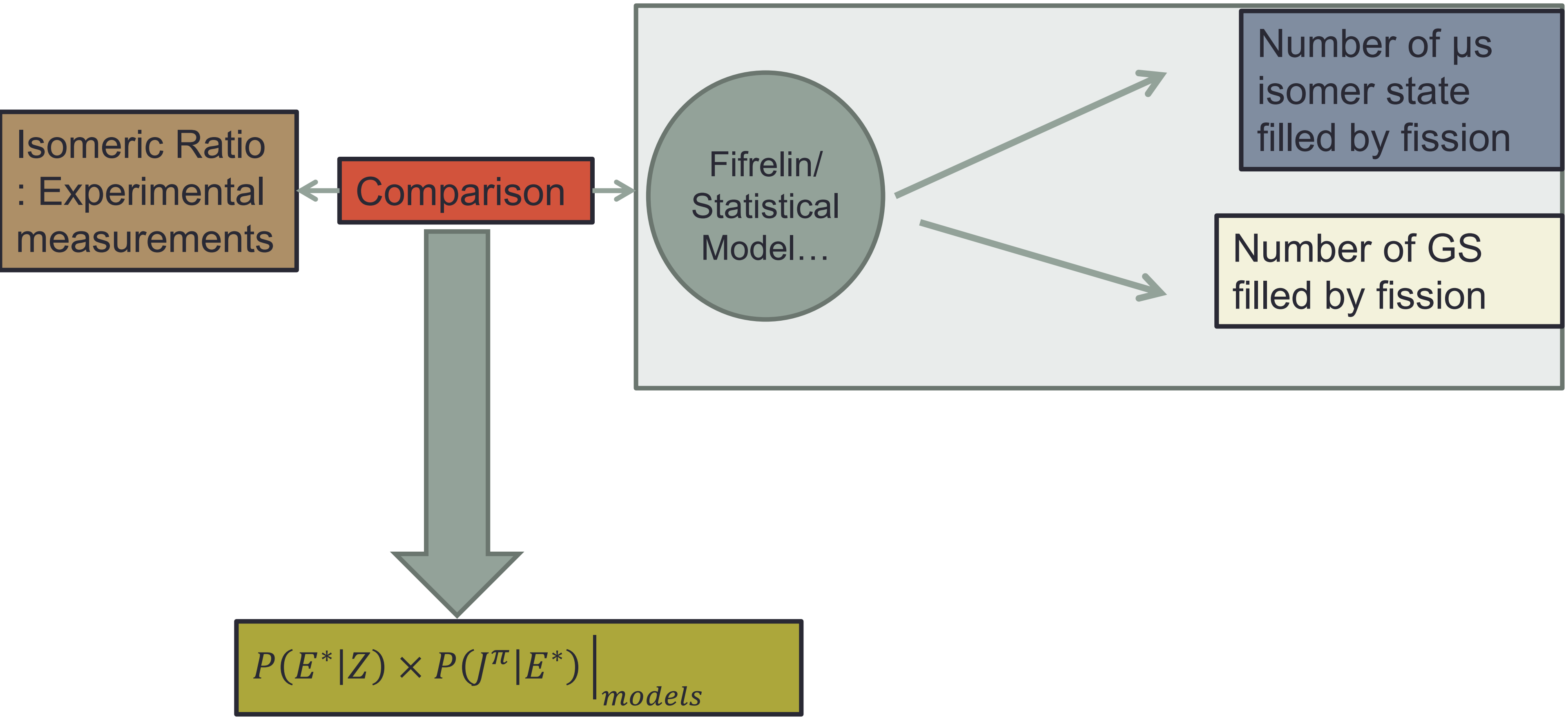
Nikolaev and D. (1968) (2)

18,68126485 1,96827205

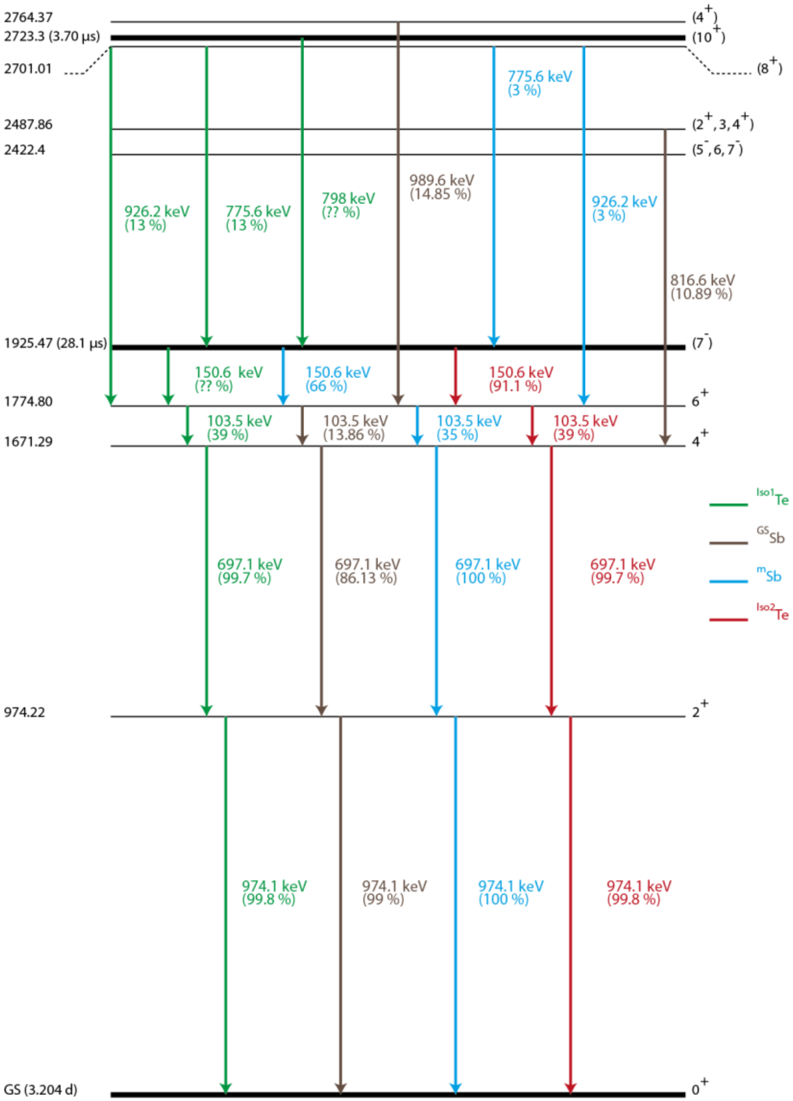
Nanosecond Isomer



Interpretation using theoretical code

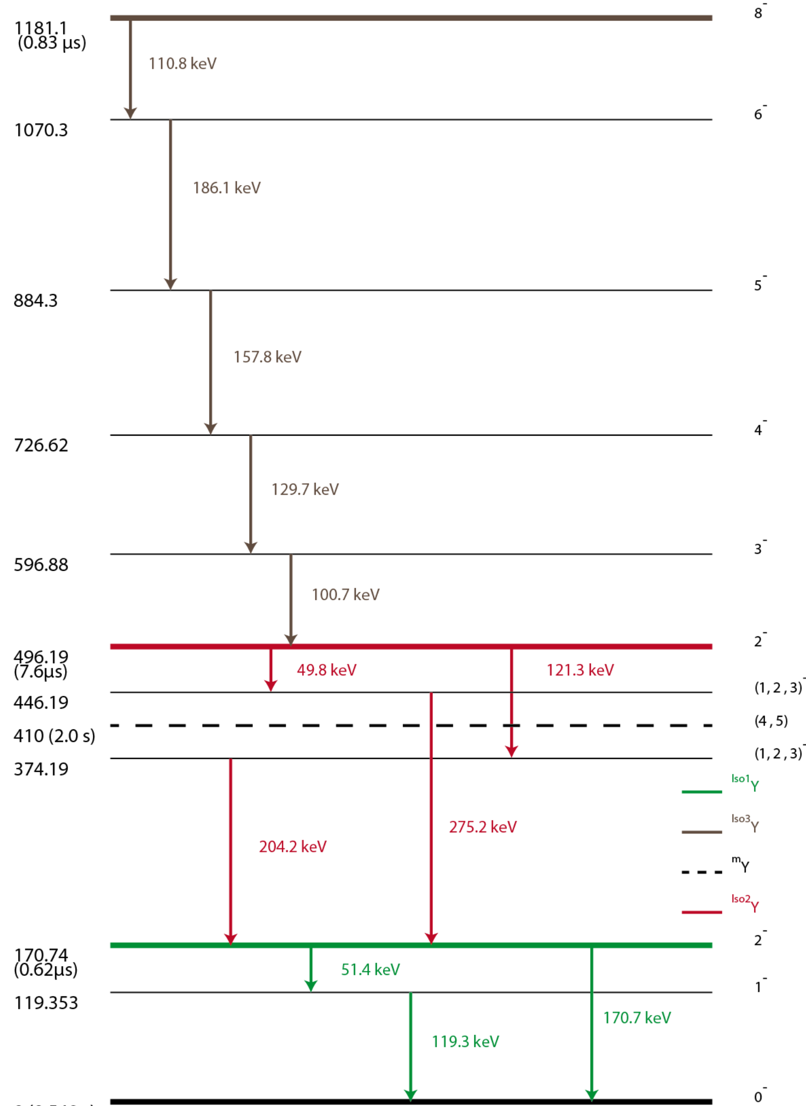


Fifrelin is a Monte Carlo code developed by CEA Cadarache.
Calculation of gamma decay of fission fragments.



2764.37
2723.3 (3.70 µs)
2701.01
2487.86
2422.4
1925.47 (28.1 µs)
1774.80
1671.29
974.22
GS (3.204 d)
(4+)
(10+)
(8+)
(2+,3,4+)
(5-,6,7-)
(7-)
6+
4+
2+
0+
775.6 keV (3 %)
926.2 keV (13 %)
775.6 keV (13 %)
798 keV (?? %)
989.6 keV (14.85 %)
926.2 keV (3 %)
816.6 keV (10.89 %)
150.6 keV (?? %)
150.6 keV (66 %)
150.6 keV (91.1 %)
103.5 keV (39 %)
103.5 keV (13.86 %)
103.5 keV (35 %)
103.5 keV (39 %)
697.1 keV (99.7 %)
697.1 keV (86.13 %)
697.1 keV (100 %)
697.1 keV (99.7 %)
974.1 keV (99.8 %)
974.1 keV (99 %)
974.1 keV (100 %)
974.1 keV (99.8 %)
Iso1Te
GSSb
mSb
Iso2Te

^{132}Te

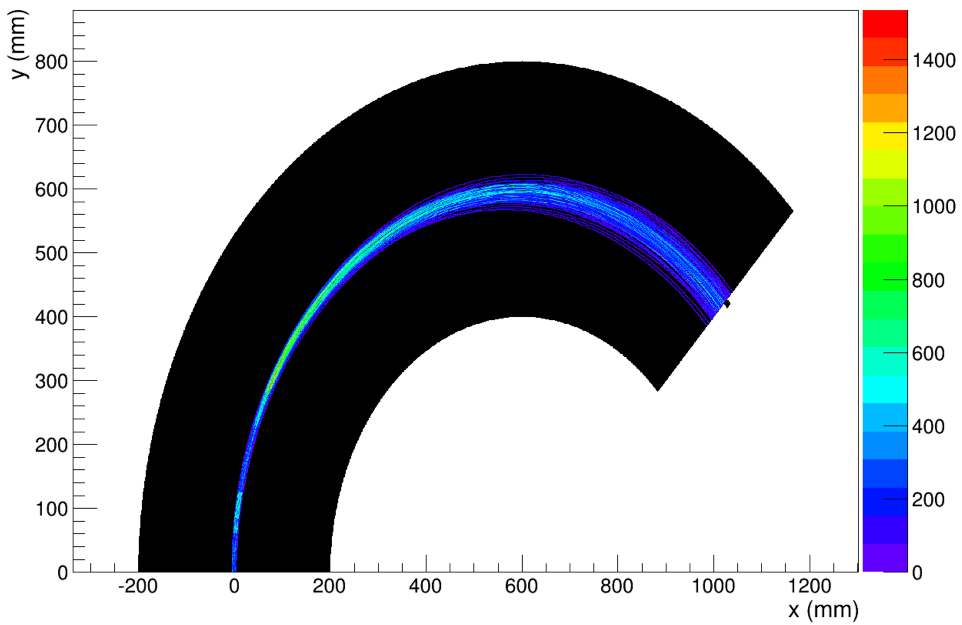


1181.1 (0.83 µs)
1070.3
884.3
726.62
596.88
496.19 (7.6µs)
446.19
410 (2.0 s)
374.19
170.74 (0.62µs)
119.353
0 (0.548 s)
8-
6-
5-
4-
3-
2-
(1,2,3)+
(4,5)
(1,2,3)-
2-
1-
0-
110.8 keV
186.1 keV
157.8 keV
129.7 keV
100.7 keV
49.8 keV
121.3 keV
204.2 keV
275.2 keV
51.4 keV
119.3 keV
170.7 keV
Iso1Y
Iso3Y
mY
Iso2Y

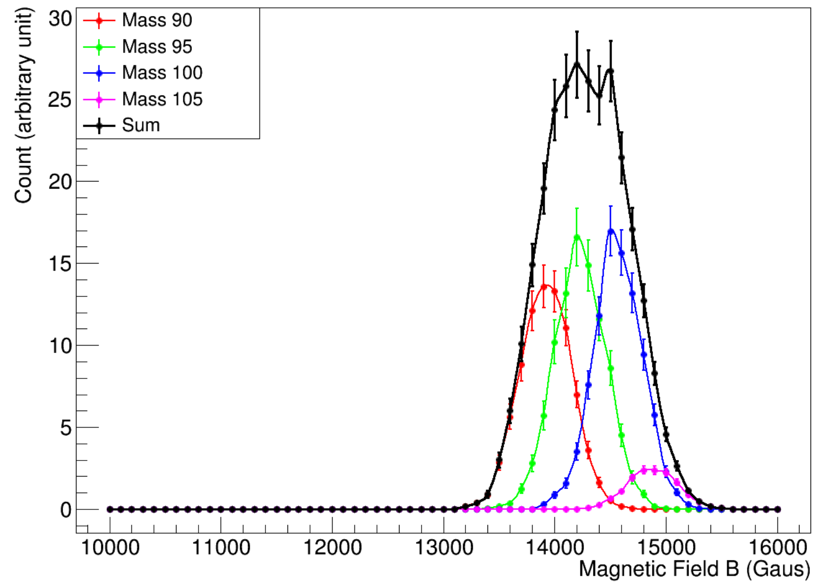
^{98}Y

What about FIPPS

Parallel Beam @ $E = 90$ MeV
1 cm collimator (in & out)



Great separation ≅ 5 $Mass$

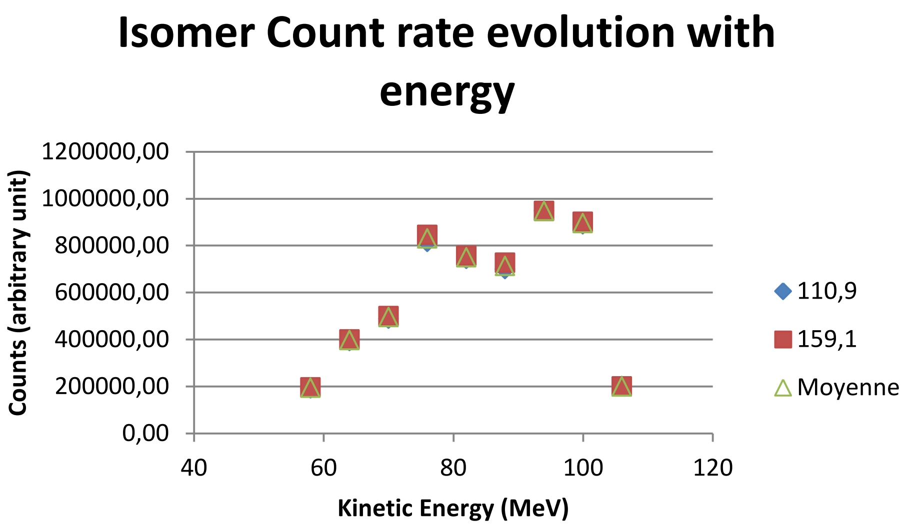


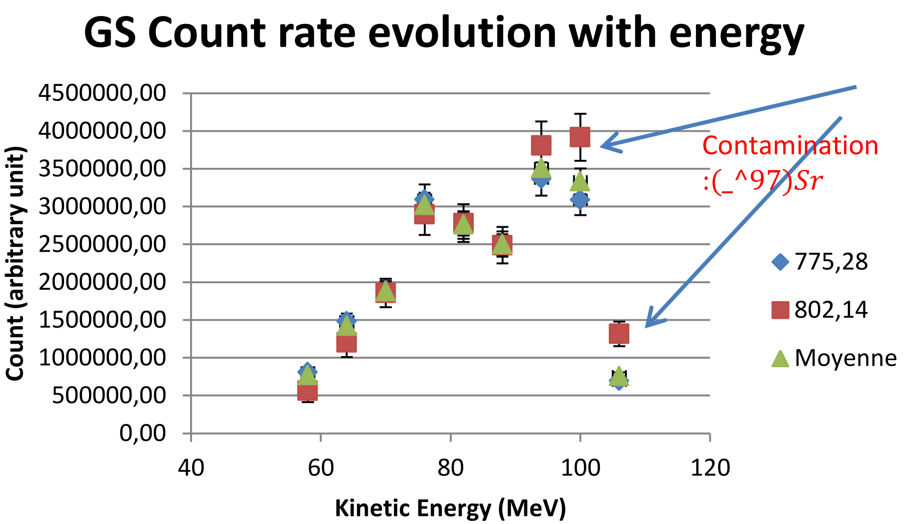
Goal : Find a geometry with the better mass separation

$\sigma_{GFM} < \sigma_{fission}$ Mass resolution is ruled by fission process

$\sigma_{GFM} > \sigma_{fission}$ Convolution between GFM and fission mass resolution

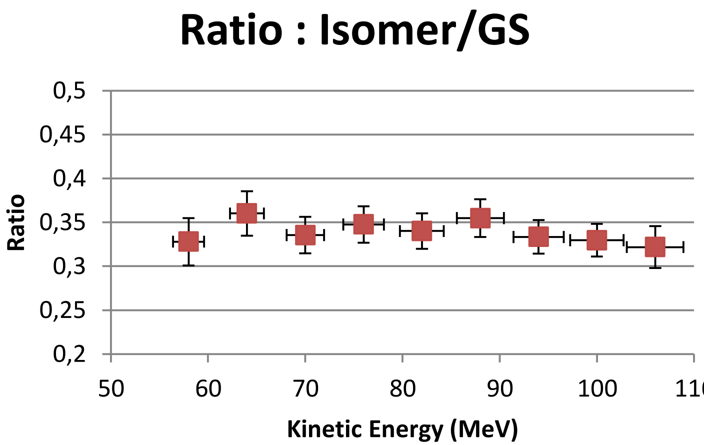
^{88}Br Results





$$N_{\gamma_1} = N_f \epsilon_{\gamma_1} I_{\gamma_1}$$
$$N_{\gamma_2} = N_f \epsilon_{\gamma 2} I_{\gamma_2}$$

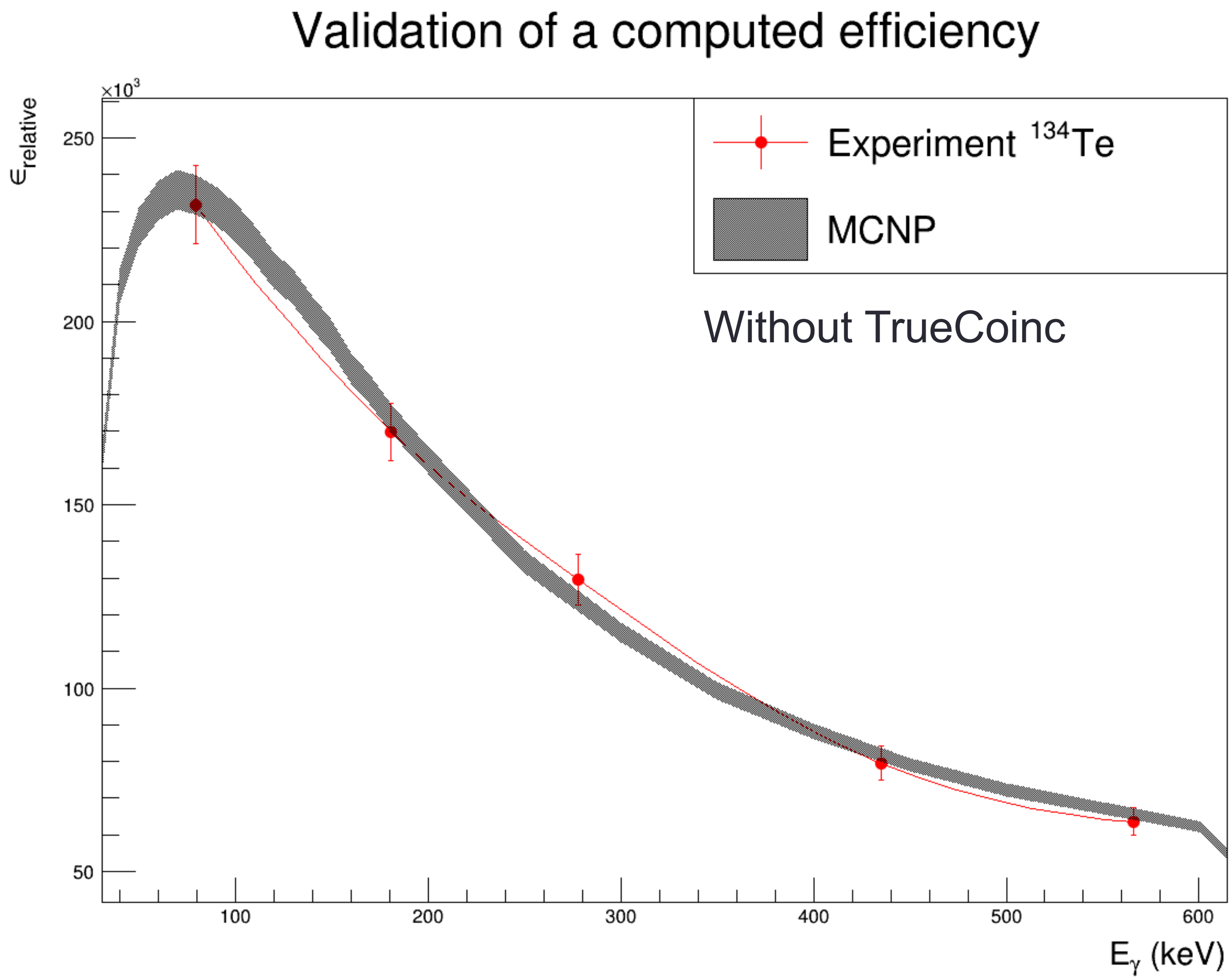
Perfect agreement between γ_1 & γ_2



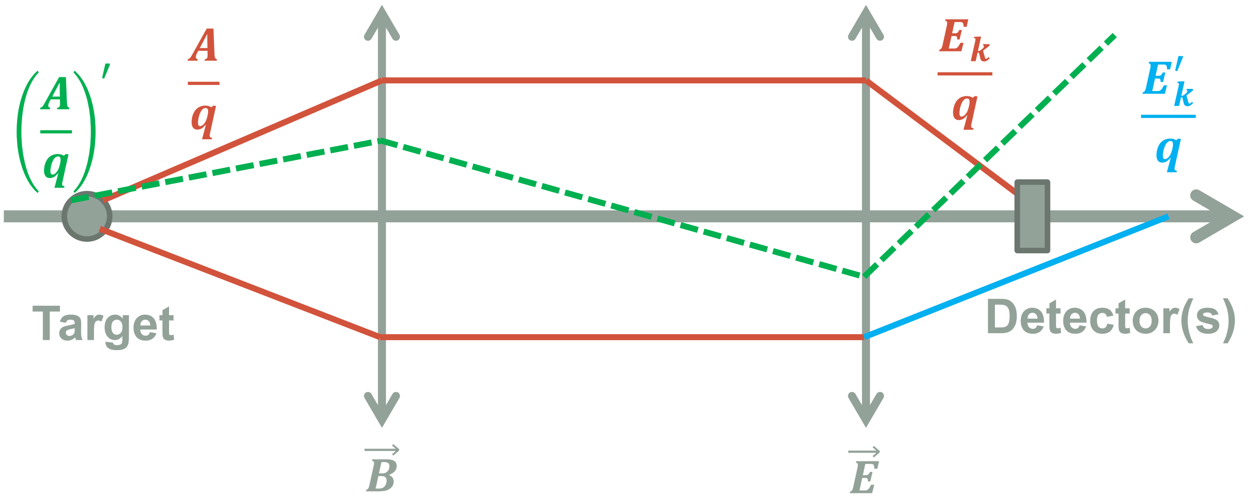
Isomeric Ratio :

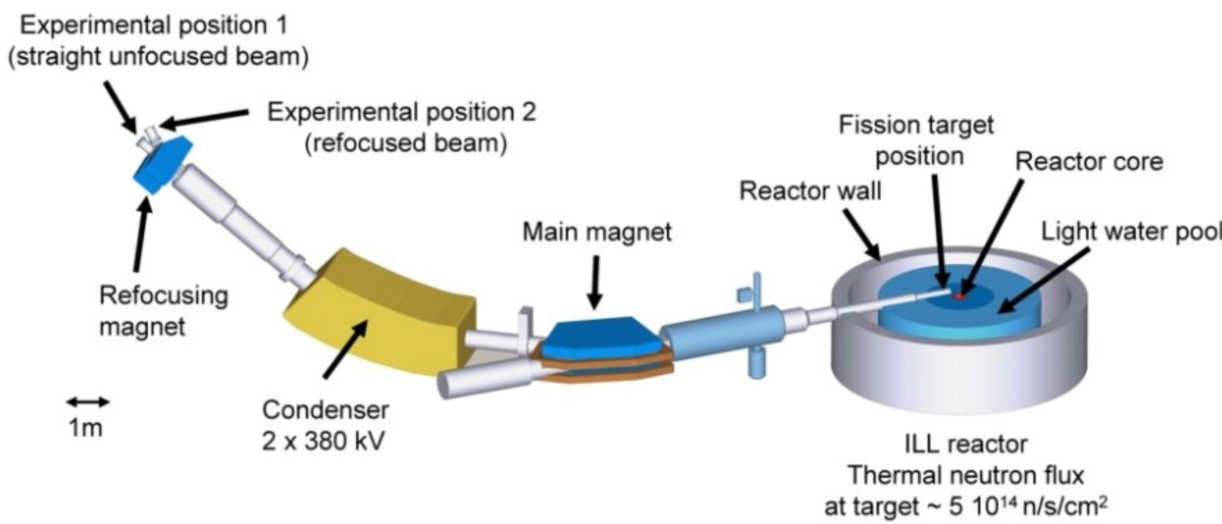
$$RI = \frac{N_f(^{88m}Br)}{N_f(^{88m}Br) + N_f(^{88}Br)} = 0{,}34 \pm 0{,}007$$

^{88}Br Analysis : γ efficiency

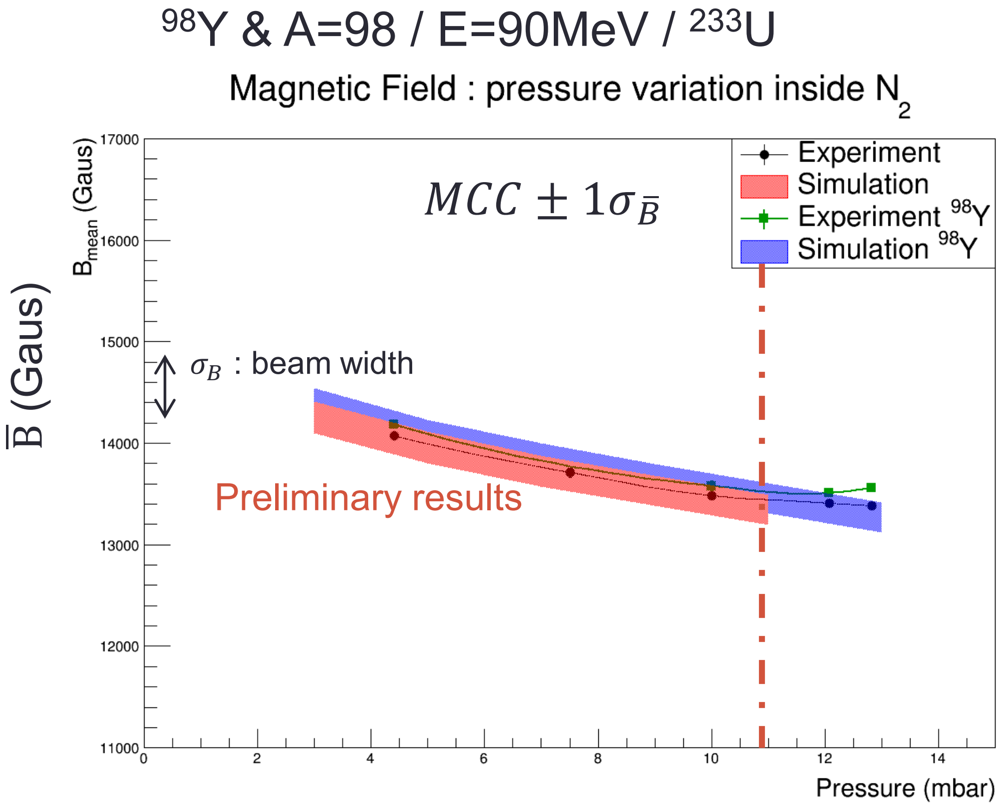


The Lohengrin mass spectrometer





N_2 Gas Filled Magnet : Test



Parameter	Sensibility $\bar{B}$	Sensibility σ_B	ΔE_{gas}
$\theta_0 \to \theta_0 + 1°$	0,346	0,711	0,44
$\theta_0 \to \theta_0 - 1°$	0,395	0,855	0,44
$\theta_1 \to \theta_1 + 1°$	0,040	0,231	0,003
$\theta_1 \to \theta_1 - 1°$	0,033	0,174	0,004
$R_{out} \to R_{out} + 5\,mm$	0,00059	0,0510	0
$R_{out} \to R_{out} - 5\,mm$	0,00088	0,0430	0
$R_{in} \to R_{in} + 5\,mm$	0,00096	0,0183	0
$R_{in} \to R_{in} - 5\,mm$	0,00058	0,0202	0
$T \to T + 10\,K$	0,00236		0,055
$T \to T - 10\,K$	0,00241		0,058
Library (Jeff vs ENDF)	0,00020	0,0028	0
Target (^{233}U vs ^{235}U)	0,00424	0,0097	0,006
$k \to k \pm 0{,}05$	0,063	0,063	0
SRIM	0,0018	0,024	0,059

- $\sigma_{MCC} = \sqrt{\sigma^2_{\bar{B}stat} + \sigma^2_{syst}}$ → Systematic : Geometry/Temperature/Library/Target
- For $P > 10\,mbar$ inconsistent calculation → Density effect on effective charge calculation