Development of a Gas Filled Magnet spectrometer coupled with the Lohengrin spectrometer for fission study

Isomeric population ratio measurements for the fission of $^{233}\text{U}(n,f)$ on the Lohengrin spectrometer

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Context of the fission yield studies

- Impact of fission yields in the actual and innovative fuel cycles
  - Inventory of used fuel: isotopic composition
  - Residual power: minor actinides and fission products
  - Radiotoxicity of used fuel
  - Experimental fuel studies: reaction cross sections and isotope yields are needed to comparison Calculation/Experiment (C/E)
  - Calculation of prompt $\gamma$ rays emitted per fissile nucleus

- Sensitivities to residual power
  - Independent measurements: uncertainties from 2.5% to 5%
  - Total correlations in data uncertainties from 8% to 16%
  - Uncertainties due to the fission yields are greater than the mean $\beta/\gamma$ energy released or the periods with a factor 2.5 to 800.

J.Ch. Benoit, PhD Thesis CEA Cadarache

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Measurements for fission process study

- Fission models are necessary for the evaluations but poor prediction power (e.g. Wilkins (scission point), Wahl ($A_p/Z_p$), Microscopic approach (Bruyères Le Châtels) …)

- Incoherence between Models or evaluations and Experiments for heavy fragments and symmetric region

Needs of new measurements

- Structure in mass and nuclear charge distributions (e.g. Fifrelin, neutron emission, $\gamma$ prompt)

- Isotopic distributions near symmetric region ➤ Nuclear charge polarization

- Spin distributions of the fission fragments as a function of the excitation energy ➤ e.g. modeling des prompt $\gamma$ emission
Progression

- Development of a Gas Filled Magnet (GFM) coupled to the Lohengrin spectrometer → Goal: Isobaric beam

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

Setup:
- IC & \( A/\Delta A \big|_{\text{Lohengrin}} = 400 \) ➤ mass yields up to \( A = 155 \) (at 3\( \sigma \))
- Ge Clover ➤ Isotopic yields with \( \gamma \) spectrometry
  ➤ for low yields or low \( \gamma \) intensities, signal/background ratio is so poor to obtain sufficient accuracy
Development of a Gas Filled Magnet spectrometer@ Lohengrin

- Progression
  - Development of a Gas Filled Magnet (GFM) coupled to the Lohengrin spectrometer \( \rightarrow \) Goal : Isobaric beam

\[
\text{Lohengrin : selection with the mass on ionic charge ratios } \frac{A}{q} \\
\text{and Kinetic energy on Ionic charge } \frac{E}{q} \\
(A_1, E_1, q_1) \equiv (A_2, E_2, q_2) \equiv (A_3, E_3, q_3)
\]

\[
\text{GFM : Spatial dispersion of fission fragments according to the mass } A \text{ and Nuclear charge } Z
\]

\[
B \cdot \rho \propto \frac{A \cdot \langle v(Z) \rangle}{\langle q(Z) \rangle} \\
B \cdot \rho \propto \frac{A}{Z^{1/3}} \quad \text{Gaz,P} \tag{1}
\]
Development of a Gas Filled Magnet spectrometer@ Lohengrin

- Progression
  - Development of a Gas Filled Magnet (GFM) coupled to the Lohengrin spectrometer → Goal: Isobaric beam

  **Lohengrin**: selection with the Mass on Ionic charge ratios $A/q$ and Kinetic energy on Ionic charge $E/q$
  
  $$\left( A_1, E_1, q_1 \right) \equiv \left( A_2, E_2, q_2 \right) \equiv \left( A_3, E_3, q_3 \right)$$

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

  $$B \cdot \rho \propto \frac{A \cdot \left\langle v(Z) \right\rangle}{\left\langle q(Z) \right\rangle} \left| \begin{array}{c} \text{Gaz,P} \\ A \\ Z^{1/3} \end{array} \right| [1]$$

  - Gas → Ionic charge is function of ion velocity
  - Magnet field → spread of extracted mass from Lohengrin

**Goal of Instrument:**
- Improve the separation power → Isobaric beam
- Increase the sensitivity in symmetric region

**Scope:**
- Fission, nuclear structure and astrophysical interest
4He Gas Filled Magnet spectrometer@ Lohengrin

Mass separation with GFM

Evolution of Purity of a mass with Bred

\[ A \rightarrow 85 \quad 90 \quad 95 \quad 100 \]

\[ A/\Delta A \rightarrow 63 ; \quad 58 ; \quad 52 ; \quad 50 \]

exit collimator 1cm $\equiv$ 100 Gauss

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**$^4$He Gas Filled Magnet spectrometer@ Lohengrin**

**Monte Carlo calculations:**

- Initial conditions: Distributions in position, Energy and Velocity $\vec{v}$
- Effective charge distribution ($q_{\text{eff}}$) according to the Betz model [1]
  - Motion equation step $\mathcal{E} < \lambda_{q\rightarrow q'} \rightarrow$ new position and velocity $\vec{v}'$
  - Bethe-Block energy loss $\rightarrow |\vec{v}'| \rightarrow |\vec{v}''|$
  - $e^-$ Capture or Loss probabilities according to the Paul model [2]
    $\rightarrow$ mean free path $\lambda_{q\rightarrow q'} \rightarrow$ stochastic charge
- Exit condition: in/out collimator

---

### Monte Carlo calculations:

- **Initial conditions**: Distributions in position, Energy and Velocity $\vec{V}$
- **Effective charge distribution** ($q_{\text{eff}}$) according to the Betz model [1]
  - motion equation step $\epsilon < \lambda_{q \rightarrow q'} \rightarrow$ new position and velocity $\vec{V}'$
  - Bethe-Block energy loss $\rightarrow |\vec{V}|'' \rightarrow \vec{V}''$
- e$^-$ Capture or Loss probabilities according to the Paul model [2]
  - mean free path $\lambda_{q \rightarrow q'} \rightarrow$ stochastic charge
- **exit condition**: in/out collimator

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**Stochastic Trajectory Calculation**

- A=100 E=90 MeV
  - $P_{\text{He}} = 30\text{mbar}$
  - Y($^{233}\text{n,f}$)$^{100}\text{Y};^{100}\text{Zr};^{100}\text{Nb};^{100}\text{Mo}$

- A=98 E=90 MeV
  - $P_{\text{He}} = 40\text{mbar}$
  - Y($^{233}\text{n,f}$)$^{98}\text{Sr};^{98}\text{Y};^{98}\text{Zr};^{98}\text{Nb}$

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**4He Gas Filled Magnet spectrometer@ Lohengrin**

### Monte Carlo calculations:
- Initial conditions: Distributions in position, Energy and Velocity $\vec{\nu}$
- Effective charge distribution ($q_{\text{eff}}$) according to the Betz model [1]
  - motion equation step $\mathcal{L} \lambda_{q \rightarrow q'} \rightarrow$ new position and velocity $\vec{\nu}'$
  - Bethe-Block energy loss $\rightarrow |\vec{\nu}''| \rightarrow \vec{\nu}''$
  - $e^-$ Capture or Loss probabilities according to the Paul model [2]
    $\rightarrow$ mean free path $\lambda_{q \rightarrow q'} \rightarrow$ stochastic charge
- Exit condition: in/out collimator

### Results:
- Mass acceptance of GFM as a function of $B$ is needed for mass and Isotopic Yield measurements
- Magnet field $B(\text{Imax}) = 1700 \text{G}$
- $^4\text{He}$ GFM separation up to $A \approx 130$

$$B \cdot \rho \propto \frac{A \cdot \langle \nu(Z) \rangle}{\langle q(Z) \rangle} \bigg|_{\text{Gaz,P}}$$

---

A = 98  E = 90MeV  
$P_{He} = 40 \text{mbar}$  
$Y( ^{235}\text{(n,f)} ^{98}\text{Sr}; ^{98}\text{Y}; ^{98}\text{Zr}; ^{98}\text{Nb})$
**N₂ Gas Filled Magnet spectrometer@ Lohengrin**

**isotopic resolution** in the GFM spectrometer
- μs Isomer used to tag isotope
N₂ Gas Filled Magnet spectrometer@ Lohengrin

isotopic resolution in the GFM spectrometer

- µs Isomer used to tag isotope

![Graph showing isotopic resolution and mass spectrometry results](image)

- Monte Carlo Calculation
- Experiment

![Mass resolution graph](image)

- A (uma)
- Bmean (Gauss)

- Mass resolution
- Mean B Cal - Exp

- 88Br, 98Y, 109Rh, 109Ru, 132Te, 136Xe

Lohengrin mass spectrometer

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N₂ Gas Filled Magnet spectrometer@ Lohengrin

**Isotopic resolution** in the GFM spectrometer

- µs Isomer used to tag isotope

According to Betz and unlike M. Paul et al., multi electron loss cross section have to be considered in N₂: \( \sigma_{2e^-} = 70\% \sigma_{e^-} \)

Validity of Betz assumptions? Gaussian distribution of ionic charge is not correct for heavy masses \( q_{exp} > q_{cal} \)

ns Isomer states in heavy region mass can disturb the ionic charge due to the internal conversion effect

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GFM perspectives

- Nowadays, the instrument allows to change quickly the setup; then comparison between vacuum magnet and Gas Filled Magnet will allow to determine the energy loss in the gas. This parameter is the last constraint to compare Experiment to Monte Carlo calculation.

- The GFM will be test with heavy noble gases (Ar, Kr, Xe) to determine the mass and isotopic resolution and then the mass/Isotope acceptance
  - Heavy gas > electronic density increases > q(Z) increases > magnet rigidity Bρ decreases > heavy mass
  - pressure > change the electronic density > maximum of resolution i.e. minimum of ΔBρ / Bρ = f(Gas, P, E_{ion}) > Best resolution for loss Energy in the gas ~70% E_{ion}

- For heavy mass, nowadays calculations are not sufficient to extract precisely the mass or isotopic acceptance > Description of the ionic charge distributions in heavy mass region have to be improved

- The GFM coupled to the Lohengrin spectrometer could limit the γ ray contaminants for the isotopic yield measurements

\[
Y(A,Z) = Y(A) \cdot P(Z|A)
\]

\[
P(Z|A) = \frac{N(A,Z)}{\sum_{Z} N(A,Z) / A_{c}(A,Z)}
\]

- γ Spectrometry after β decay
- Relative measurements

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Isomeric ratio with $\gamma$ spectrometry method

\[ Y(A, Z, \text{Isomer}) = Y(A) \cdot P(Z|A) \]

\[ \frac{R(m/\text{GS}; E_k)}{1 + R(m/\text{GS}; E_k)} \]

\[ \frac{1}{1 + R(m/\text{GS}; E_k)} \]

Ionisation Chamber

IC measurements

$\gamma$ Spectrometry measurements

Mean isomeric ratio on energy kinetic distribution

Lohengrin $^{233}$U(n,f)

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$^{98}$Y Isomeric populations : GS – m states

$0^+ \text{(GS)}$ → $\beta$ decay → $98\text{Sr}$

$0^-$ (GS) → $\beta$ decay → $98\text{Y}$

$0^+$ (GS) → $\beta$ decay → $98\text{Zr}$

$10^-$ (0.83 µs)

$4^- \text{ (7.6 µs)}$

$(4,5) \text{ (2.0 s)} m$

$2^- \text{ (0.62 µs)} \beta$ decay

$0^-$ (GS) $548s$ $98\text{Y}$

$0^+ \text{ (GS)}$ $30.7s$ $98\text{Zr}$
\( ^{98}\text{Y} \) Isomeric populations: GS – m states

\( \gamma \) raies after \( \beta \) decay of \( ^{98}\text{Y}_{\text{GS}} \) & \( ^{98}\text{Y}_{\text{m}} \):

- 1590 keV & 1223 kev → \( Y_{\text{GS}} \) & \( Y_{\text{m}} \)
- 621 keV et 648 keV → \( Y_{\text{m}} \)

Measurements without time coincidence between IC and Ge clovers

\( T_{\text{exp}} = 120 \text{ mn for each kinetic energy} \)

\[
N^{^{98}Y}_{\text{GS}}|^{\text{Fission}} = N^{^{98}Y}_{\text{GS}}|^{\text{tot}} - N^{^{98}\text{Sr}}_{\gamma} \rightarrow Y^{^{98}Y}_{\text{GS}}
\]

\[
N^{^{98}Y}_{m}|^{\text{fission}} = N^{^{98}Y}_{m}|^{\text{tot}} - N^{^{98}\text{Sr}}_{\gamma} \rightarrow Y^{^{98}Y}_{m}
\]

No event from \( ^{98}\text{Sr} \)

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\[ N(\mu s \ Y)_{t=tof}^f = N(\mu s \ Y)_{t=tof}^{tot} - \sum_{gate} N(\mu s \ Y)_{\beta; 98 \ Sr} \]

\[ N(\mu s \ Y)_{t=tof}^f = N(\mu s \ Y)_{t=tof}^{tot} - \tau(98 \ Sr) \cdot N_{gate} \cdot \Delta t_{gate} \]

Sr decay rate: \[ \tau(98 \ Sr) = \frac{N(98 \ Sr)}{T_{exp}} \]

\[ \gamma \text{ events from Sr} \]

Life time Sr << T_{exp}

\[ \gamma \text{ Raies after beta decay of 98Sr} \rightarrow 444 \text{ keV and 428 keV} \]
\[ N(\mu s \text{ Y})_{t=tof}^f = N(\mu s \text{ Y})_{t=tof}^{tot} - \sum_{gate} N(\mu s \text{ Y})_{\beta; 98\text{ Sr}} \]

\[ N(\mu s \text{ Y})_{t=tof}^f = N(\mu s \text{ Y})_{t=tof}^{tot} - \tau(98\text{ Sr}) \cdot N_{gate} \cdot \Delta t_{gate} \]

\text{Sr decay rate: } \tau(98\text{ Sr}) = \frac{N(98\text{ Sr})}{T_{\text{exp}}} \rightarrow \gamma \text{ Raies after beta decay of 98Sr}

\rightarrow 444 \text{ keV and } 428 \text{ keV}
$^{98}$Y Isomeric populations : $\mu$s isomer

Sensitive parameters :

- Gamma Intensity per $\beta$ decay
- Relative intensity
- Internal conversion coefficients \cite{1}
- Relative gamma efficiency $\rightarrow ^{96}$Y

\[ N(\mu s \ Y)_{t=tof}^f = N(\mu s \ Y)_{t=tof}^{tot} - \sum_{gate} N(\mu s \ Y)_{\beta; \ ^{98}Sr} \]

\[ N(\mu s \ Y)_{t=tof}^f = N(\mu s \ Y)_{t=tof}^{tot} - \tau(\ ^{98}Sr) \cdot N_{gate} \cdot \Delta t_{gate} \]

Sr decay rate : \[ \tau(\ ^{98}Sr) = N(\ ^{98}Sr)/T_{exp} \]

\[ \gamma \] Raies after beta decay of $^{98}$Sr $\rightarrow 444 \text{ keV and } 428 \text{ keV}$

[1] IAEA data base

\cite{Wonder Workshop, Sept. 2012}
### 98Y Isomeric ratios in the fission of $^{233}$U(n,f)
#### Preliminary results

<table>
<thead>
<tr>
<th>I Ratio (t=tof)</th>
<th>GS 0-</th>
<th>m (4,5)</th>
<th>$\mu_1$ 2-</th>
<th>$\mu_2$ 4-</th>
<th>$\mu_3$ 10-</th>
</tr>
</thead>
<tbody>
<tr>
<td>GS (548s)</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>m (2s)</td>
<td>0.158 ±0.008</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\mu_1$ (0.62\mu s)</td>
<td>0.023 ±0.011</td>
<td>0.146 ±0.071</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\mu_2$ (7.6\mu s)</td>
<td>0.409 ±0.018</td>
<td>2.585 ±0.078</td>
<td>17.8 ±8.6</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>$\mu_3$ (0.83\mu s)</td>
<td>0.014 ±0.001</td>
<td>0.087 ±0.008</td>
<td>0.60 ±0.29</td>
<td>0.034 ±0.001</td>
<td>1</td>
</tr>
</tbody>
</table>

- In fine, 10 Isomeric ratios have been determined for the mean kinetic energy of 98Y
- These Isomeric Ratios depend of the spin distribution at excitation energy
- six Isomeric ratios at $E_k$ have been measured for A =88; 94 98; 99; 129; 132 at mean kinetic energy associated
Conclusions and perspectives on the Isomeric ratio measurements for the fission of $^{233}\text{U}(n,f)$

• Isomeric populations have been measured for 6 masses at different mean kinetic energy $\rightarrow R(A, Z, \overline{E}_k)$

• For $^{98}\text{Y}$, 10 ratios have been measured. The goal is now to interpret these measurements as a spin probability distribution at $\overline{E}_k$: $P(J|\overline{E}_k)$

The spin distribution depends on:

▶ level density as a function of $E^*$
▶ Excitation energy repartition between the two fragments
▶ 10 degrees of freedom will allow to describe several momenta of the spin distribution

• Considering the isomeric ratio measurements on 6 masses at different mean kinetic energies, the spin distribution can be explored as a function of excitation energy $P(J; E^*)$

• To limit the systematic effects due to the models, a new campaign for the same isotopes with a target of $^{235}\text{U}$ has been proposed to compare the spin distributions
References:
