Gamma-ray cascade study in the abundant fission fragments with the EXILL experiment and FIFRELIN simulation

MICHAIŁ RĄPAŁA

Varenna, 12 June 2018
MOTIVATIONS

Generation III+ and IV reactors
- More accurate simulation
- Higher safety

Gamma heating process
- More precise simulation

HTMR Asia Development Limited,
MOTIVATIONS

Gamma heating process
- More precise simulation

Fission fragment deexcitation simulation code
- Improvement of implemented models

Fission
- Prompt gamma-ray cascade

Generation III+ and IV reactors
- More accurate simulation
- Higher safety

HTMR Asia Development Limited,
Experimental data
• Array of Ge-detectors around a fissile target in an intense cold neutron beam at ILL
  • 15 days with $^{235}\text{U} (575 \text{ mg/cm}^2) + \text{Sn/Zr}$
  • 5 days with $^{235}\text{U} (675 \mu\text{g/cm}^2) + \text{Be}$
  • 15 days with $^{241}\text{Pu} (300 \mu\text{g/cm}^2) + \text{Be}$

• Around $10^5$ fissions per second
• Detection efficiency $\sim 6\%$ at 1MeV
• Fragments stopped in the target (no Doppler)
• Data analysis $\rightarrow \gamma\gamma\gamma$ coincidences

M. Jentschel et al, JINST 12, P11003 (2017)
TRIPLE GAMMA COINCIDENCE IN PRACTICE (1/2)


Kr

Ba

Fragment pair selection

observable

M. Rapala, IRFU/DPhN/LERN

June 12, 2018
Coincidence spectrum with the 359.5 keV and 769.0 keV transitions

- 475 keV (4<sup>+</sup> → 2<sup>+</sup>) $^{142}$Ba
- 631.1 keV (6<sup>+</sup> → 4<sup>+</sup>) $^{142}$Ba
- 1034 keV (4<sup>+</sup> → 2<sup>+</sup>) $^{92}$Kr
- 1296.7 keV (4<sup>+</sup> → 2<sup>+</sup>) $^{92}$Kr

Counts per bin width [c]

Counts per bin width [c]
Problems with analysis and solutions
• Results depends on the gates and baseline selection

Gates at 401.6keV ($^{90}$Kr) and 199.2keV ($^{144}$Ba)

330.7keV ($4^+ \rightarrow 2^+$) $^{144}$Ba
• Redefinition of background treatment
  - NO DIRECT BACKGROUND SUBTRACTION → GATE SCANNING

2D gate scan → Semi-automatic spectra fitting → Gate fitting
FIFRELIN simulations
**FIFRELIN Ingredients**

- Deexcitation with the Hauser-Feshbach treatment using
  - Experimental nuclear level scheme at low excitation energy,
  - Nuclear level density at high excitation energy,
  - Gamma strength functions,
  - Neutron transmission from optical potential

- \((E^*,J)\) distribution of the primary fragment

  - constant spin cut-off model (Constant)
  - energy dependent spin cut-off model (BSFGM)

100\(^{100}\)Zr gamma-ray cascade simulated with FIFRELIN
**Impact of the (E,J) Distribution on the Evolution of the Cascade with the Partner (1)**

**Evolution of the γ-ray intensities in $^{100}$Zr**

- Strong dependence with the Fission partner
- Constant spin cut-off model
- Strong dependence with the Fission partner

→ Te mass number
← Neutron evaporation
← Excitation energy
IMPACT OF THE \((E,J)\) DISTRIBUTION ON THE EVOLUTION OF THE CASCADE WITH THE PARTNER (2)

- No dependence with the fission partner

BSFGM model

Evolution of the \(\gamma\)-ray intensities in \(^{100}\text{Zr}\)

- \(\text{Te mass number}\)
- \(\text{Neutron evaporation}\)
- \(\text{Excitation energy}\)
The intensities of the main transitions do not seem to depend on the fission partner (i.e. on the number of evaporated neutrons)

→ Rejection of the “Constant” model
Feeding of the high spin states is overestimated in Fifrelin

M. Rapala, IRFU/DPhN/LERN
“BSFGM model”

Primary fragment distribution

\[ n(J) = f_\sigma \cdot \rho(J) \]

Where \( \rho(J) = \frac{2J+1}{2\sigma^2} \exp\left(-\frac{(J+\frac{1}{2})^2}{2\sigma^2}\right) \)

is the spin dependence of the nuclear level density

Idea: Scan of \( f_\sigma \) and comparison of simulations with data

\( \chi^2 \) of simulations with different \( f_\sigma \)

(\( \chi^2 \) built on the intensities of the main transitions in the GS band of \(^{100}\)Zr)

The spin distribution of the fission fragments is close to the spin distribution of the nuclear level density
Initial simulation

\[ \nu_L = 1.42 \]
\[ \nu_H = 1.01 \]
\[ \nu_T = 2.43 \]

Simulation with modified \( f_s \)

- Better intensities but:

\[ \nu_L = 1.53 \]
\[ \nu_H = 1.2 \]
\[ \nu_T = 2.73 \]

Idea: Scan of the other free parameters
Conclusions
CONCLUSIONS

• Experimental data (EXILL experiment)
  • Measurement of the prompt-gamma ray cascades coming from the deexcitation of the fission fragments
  • Semi-automatic analysis technique (γγγ coincidences)
  • Determination of the prompt-gamma ray intensities

• Simulation data (FIFRELIN code)
  • Analysis of the spin cutoff models (rejection of the “Constant” model)
  • Optimization of spin (the nuclear level density spin distribution)
  • Prompt-neutron multiplicity optimization needed
COLLABORATION

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\textsuperscript{4} Institut Laue-Langevin, Grenoble, France
\textsuperscript{5} LPSC, CNRS/IN2P3, Grenoble, France
\textsuperscript{6} INFN, Legnaro, Italy
\textsuperscript{7} GANIL, Caen, France
• Europium source data used for calibration
\[ ^{152}\text{Eu} \rightarrow ^{152}\text{Gd} \text{ or } ^{152}\text{Sm} \]

• Three types of detectors with different performance

• Total coincidence efficiency formula simplification

\[
\varepsilon_{\text{coinc}}(E_1, E_2, E_3) = \sum_{i=1}^{N} \sum_{j \neq i}^{N} \sum_{k \neq i, j}^{N} \varepsilon_i(E_1) \varepsilon_j(E_2) \varepsilon_k(E_3)
\]

\[
\varepsilon_{\text{app}}(E_1, E_2, E_3) = \frac{(N - 1)(N - 2)}{N^2} \varepsilon_T(E_1) \varepsilon_T(E_2) \varepsilon_T(E_3)
\]

\[
\varepsilon_T(E) = \sum_{i=1}^{N} \varepsilon_i(E) = N \cdot \varepsilon(E)
\]

**DIFFERENCE LOWER THAN 2% IN RANGE BETWEEN 100keV TO 1.5MeV**
• Europium source data used for calibration
  $^{152}\text{Eu} \xrightarrow{} ^{152}\text{Gd}$ or $^{152}\text{Sm}$
• Three types of detectors with different performance
• Total coincidence efficiency formula simplification
• Correction of true coincidence effect
• Better value of the peak/background ratio

Gates at 769.0keV \(^{82}\text{Kr}\) and 359.5keV \(^{142}\text{Ba}\)

- 2 bins gate width
- 1 bin gate width
• Better visibility of contaminants
• More precise contamination positioning
• More precise results
• Lower uncertainty

• 2 bins gate width

• 1 bin gate width
• Dependence on a complementary fragment (neutron evaporation)

- Effect caused by dependence between excitation energy and level density
- More evaporated neutrons
- Lower excitation energy of the fission fragments after neutron evaporation
- Lower probability of high-spin transition production

FIFRELIN simulated transition intensities in $^{142}$Ba
• Three-dimensional histogram → γγγ-cube

• Fixed time window

• Transitions coming within the time window limit are considered to be in coincidence
GAMMA HEATING PROCESS IN A NUCLEAR REACTOR

• Result of the gamma-ray energy deposition


Areva, UK-EPR, Fundamental safety overview Vol. 1, Chapter A, Page 63

Almost 100% of total heating in the reflector
What we want to study:

- Fission Process
- Prompt Gamma-Ray Cascade in Fission Fragments
The continuum

The part partially completed from the models

The experimental nuclear levels

The experimental levels
Additional levels (from theoretical law)

E_{\text{bin}}
E_{\text{RIPL3}}^{\text{cut-off}}
S_n
E_i, J_i, \pi_i

O. Litaize et al., ND-2016, Bruges
Fission fragment de-excitation

Simulation code
- FIFRELIN
  - Fission Models
  - Nuclear structure models
  - De-excitation models
  - Databases

Observables
- Neutrons and prompt gamma-rays spectra and multiplicity
- Post-neutron yields, released energy
- Gamma-ray cascades in the fragments

Results useful for simulation of:
- Residual power
- Gamma-heating
- Radiation embrittlement of reactor vessel
Main questions concerning de-excitation process:

What happens after the scission point?
How is the excitation energy shared between the two fragments?
What are the initial spin distributions?
Are they correlated?
What is the process that generates high spin in the fragment?
Three-dimensional histogram → γγγ-cube

Time shift correction for all Ge crystals
Energy calibration of all Ge crystals (run after run)
Coincidence time window
  - FIXED TO 200NS

Cube created in Poland at Warsaw University
  - TIME NEEDED: 6 TO 12 MONTHS
• Gating process

  • SELECTING TRANSITION OF A PARTICULAR ENERGY

  • SELECTING FISSION FRAGMENTS
SPECTRA FITTING PROBLEMS - PEAK SHAPE

- Simple Gaussian

- Bad resolution
  - SUM OF ALL DETECTORS
  - SUM OF ALL RUNS
  - COMPTON – TAILS
• $^{91}\text{Zr}(n,\gamma)$, $^{92}\text{Zr}(n,\gamma)$
  • ADJUSTED ON $^{92}\text{Zr}$ AND $^{93}\text{Zr}$ CLEAN PEAKS

• Fit function
  • SUM OF 3 GAUSSIANS
    • Same center
    • COMPTON – TAILS
• Triple Gaussian + Compton tails
• Gating with subtracted background
  • Gate background close to the peak

Spectrum without gates
• Gating with subtracted background
  
  • Gate background close to the peak
  
  Gate at 401.6keV ($^{90}$Kr)
ANALYSIS PROCEDURE

- Fitted background
- Precise contamination identification and removal
- Reproducible results
• Gamma-ray cascade

Gamma-ray cascade in $^{142}$Ba according to FIFRELIN
T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)
M. Rapala, IRFU/DPhN/LERN

Gamma-ray cascade in $^{142}$Ba according to EXILL data
T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)
PRELIMINARY RESULTS

- Gamma-ray cascade

Gamma-ray cascade in $^{142}$Ba according to FIFRELIN
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M. Rapala, IRFU/DPhN/LERN

June 12, 2018
**Preliminary Results**

- **Gamma-ray cascade**
  - **Good agreement for low spin transitions**
  - **High spin transitions overestimated by FIFRELIN code**
  - **Probably wrong estimation of a mean value of the fission fragment spin distribution**

<table>
<thead>
<tr>
<th>E (keV)</th>
<th>J1→J2</th>
<th>EXILL*</th>
<th>FIFR.**</th>
<th>Ratio(/%)</th>
</tr>
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<tbody>
<tr>
<td>359.6</td>
<td>2^+→0^+</td>
<td>100 (3)</td>
<td>100.0 (1)</td>
<td>1.00 (3)</td>
</tr>
<tr>
<td>475.2</td>
<td>4^+→2^+</td>
<td>80 (2)</td>
<td>91.4 (2)</td>
<td>1.14 (3)</td>
</tr>
<tr>
<td>631.2</td>
<td>6^+→4^+</td>
<td>42 (2)</td>
<td>66.3 (1)</td>
<td>1.58 (8)</td>
</tr>
<tr>
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<tr>
<td>766.5</td>
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<td>2.4 (7)</td>
<td>3.60 (3)</td>
<td>1.5 (4)</td>
</tr>
<tr>
<td>706.8</td>
<td>5^-→4^+</td>
<td>10 (1)</td>
<td>10.17 (5)</td>
<td>1.0 (1)</td>
</tr>
<tr>
<td>486.7</td>
<td>7^-→6^+</td>
<td>8 (1)</td>
<td>15.60 (6)</td>
<td>1.9 (2)</td>
</tr>
<tr>
<td>354.4</td>
<td>9^-→8^+</td>
<td>4.4 (5)</td>
<td>8.40 (4)</td>
<td>1.9 (2)</td>
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<tr>
<td>561.1</td>
<td>9^-→7^-</td>
<td>4.0 (5)</td>
<td>7.52 (4)</td>
<td>1.9 (2)</td>
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<td>11^-→9^-</td>
<td>2 (1)</td>
<td>6.22 (3)</td>
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<td>8^-→6^+</td>
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<td>585.6</td>
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**Preliminary Results**

- Gamma-ray cascade

**Gamma-ray cascade in $^{142}$Ba according to $^{248}$Cm data**

Urban et al., Nucl. Phys A 613 (1997) 107

M. Rapala, IRFU/DPhN/LERN

**Gamma-ray cascade in $^{142}$Ba according to EXILL data**

T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)

M. Rapala, IRFU/DPhN/LERN

June 12, 2018
**Preliminary Results**

- **Gamma-ray cascade**
  - Transition intensity values very close especially in ground state band
  - Probably initial spins of the primary fission fragments in both systems are similar

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<th>E (keV)</th>
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<th>( I_\gamma ) (^{248}\text{Cm}^{**} )</th>
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PRELIMINARY RESULTS

- Dependence on a complementary fragment (neutron evaporation)

FIFRELIN simulated transition intensities in $^{142}$Ba

EXILL data transition intensities in $^{142}$Ba
Gamma-ray cascade in $^{92}$Kr according to EXILL data
(in coincidence with $^{142}$Ba)
T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)

Gamma-ray cascade in $^{92}$Kr according to FIFRELIN
(old RIPL-3 version, without low intensity transitions, in coincidence with $^{142}$Ba)
PRELIMINARY RESULTS

- RIPL-3 update
- Strong dependence of the FIFRELIN simulation output on spin values

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Gamma-ray cascade in $^{92}$Kr according to EXILL data
(in coincidence with $^{142}$Ba)
T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)

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Gamma-ray cascade in $^{92}$Kr according to FIFRELIN
(new RIPL-3 version, without low intensity transitions,
in coincidence with $^{142}$Ba)
T. Materna et al., accepted for publ. In EPJ Web of Conferences (2017)
• Simulations validity criterion:
  - AVERAGE PROMPT-NEUTRON MULTIPLICITY

\[
\bar{v}_L = 1.41 \\
\bar{v}_H = 1.01 \\
\bar{v}_T = 2.42
\]

• \(\sigma_L, \sigma_H\) (CONSTANT) or \(f_\sigma\) (BSFGM)
• **Free parameters:**
  
  1. \( R_T^{\text{min}} \)
  2. \( R_T^{\text{max}} \)

\[
E^* = aT^2
\]

\[
X E_L = E_L^* + E_L^{rot}
\]
Ratio of $\gamma$-transition intensities in $^{100}$Zr (EXILL data/$^{252}$Cf data)
• In CONSTANT model initial spin is unchanged
• Neutron evaporation follows the mean level density
RESULTS: COMPARISON WITH SPONTANEOUS FISSION

- $^{248}\text{Cm}$ – good agreement
- $^{252}\text{Cf}$ – discrepancy

<table>
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<tr>
<th>E (keV)</th>
<th>J1→J2</th>
<th>$^{235}\text{U}(n_{\text{th}},f)$ (this work)</th>
<th>$^{252}\text{Cf}$</th>
<th>$^{248}\text{Cm}$</th>
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<tr>
<td>212.5</td>
<td>2$^+$$\rightarrow$0$^+$</td>
<td>100</td>
<td>100</td>
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<tr>
<td>352.0</td>
<td>4$^+$$\rightarrow$2$^+$</td>
<td>78(3)</td>
<td>84(4)</td>
<td>81(8)</td>
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<tr>
<td>497.1</td>
<td>6$^+$$\rightarrow$4$^+$</td>
<td>44(2)</td>
<td>65(3)</td>
<td>49(5)</td>
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<td>625.6</td>
<td>8$^+$$\rightarrow$6$^+$</td>
<td>15(1)</td>
<td>26(1)</td>
<td>14(3)</td>
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<td>2.5(6)</td>
<td>5.2(3)</td>
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<tr>
<td>841.7</td>
<td>12$^+$$\rightarrow$10$^+$</td>
<td>1.2(5)</td>
<td>2.1(6)</td>
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<tr>
<td>219.8</td>
<td>7$^+$$\rightarrow$6$^+$</td>
<td>4.1(6)</td>
<td>5.5(3)</td>
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<tr>
<td>250.3</td>
<td>8$^+$$\rightarrow$7$^+$</td>
<td>1.8(4)</td>
<td>2.4(7)</td>
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<tr>
<td>267.3</td>
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<td>0.8(4)</td>
<td>1.0(3)</td>
<td></td>
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<td>3.0(9)</td>
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<tr>
<td>845.2</td>
<td>6$^+$$\rightarrow$4$^+$</td>
<td>3.0(6)</td>
<td>8.8(4)</td>
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<tr>
<td>1695.2</td>
<td>6$^+$$\rightarrow$4$^+$</td>
<td>2.8(4)</td>
<td>5(1)</td>
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<td>665.98</td>
<td>2$^+$$\rightarrow$2$^+$</td>
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<td>7.0(4)</td>
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<td>850.1</td>
<td>4$^+$$\rightarrow$4$^+$</td>
<td>5.0(6)</td>
<td>9.5(5)</td>
<td></td>
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RESULTS - DIFFERENT SPIN CUTOFF MODELS

- The main band
  - TOO HIGH INTENSITIES

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### Preliminary Results

- **Scanning** $R_T^{min}$ and $R_T^{max}$

- **Too high** $\bar{\nu}_T \approx 2.7$

- **Single element testing**
  - **Prompt-neutron multiplicity** of $^{100}$Zr ($1.4 \div 1.65$)

- **Valid** $\bar{\nu}_{Zr}$
  - **Low** $R_T^{max} = 1.00$

<table>
<thead>
<tr>
<th>E (keV)</th>
<th>J1→J2</th>
<th>$I_\gamma$</th>
<th>EXILL</th>
<th>$\bar{\nu}_{Zr} = 1.78$</th>
<th>$\bar{\nu}_{Zr} = 1.49$</th>
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<tr>
<td>212.5</td>
<td>2⁺→0⁺</td>
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<tr>
<td>352.0</td>
<td>4⁺→2⁺</td>
<td>78(3)</td>
<td>84.8(9)</td>
<td>86.3(9)</td>
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<tr>
<td>497.1</td>
<td>6⁺→4⁺</td>
<td>44(2)</td>
<td>54.9(7)</td>
<td>57.7(7)</td>
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<tr>
<td>625.6</td>
<td>8⁺→6⁺</td>
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<td>14.9(3)</td>
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<td>739.2</td>
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<tr>
<td>841.7</td>
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<td>219.8</td>
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<td>250.3</td>
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<td>1.14(8)</td>
<td>1.02(8)</td>
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<tr>
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<td>3.0(1)</td>
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</table>
• Measurement of the prompt gamma-ray cascades of the fission fragment pairs
  • Zr-Te, Kr-Ba, Sr-Xe, Mo-Sn
• COMPARISON TO OTHER EXPERIMENTAL DATA
  • $^{252}\text{Cf}$, $^{248}\text{Cm}$