Improving the Nuclear Data on Fission Product Decays at CARIBU

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Joint Proposal of LLNL and ANL

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**BNL:** S. Zhu

**UTK:** K. Siegl

**LBNL:** R. Orford
The Goal of this Project is to Perform Detailed Studies of Key Fission-Product Decay Properties

Nuclear Data Impacts Understanding of Nuclear Events:

**Fission-product isomer-to-g.s. ratios and masses**
Understanding of fission dynamics and angular momentum

**Measurements of short-lived fission products**
- $^{132m}/^{132}$Sb isomer to g.s. ratio & mass measurements
- $^{128m}/^{128}$Sn and $^{128m}/^{128}$Sb isomer to g.s. ratios & mass measurements

**Beta-delayed γ-ray branching ratios**
Nuclear data for fission yields - impacts nuclear forensics

**Precision branching ratio of long-lived fission products of the importance to nuclear forensics**
- $^{156}$Eu precision decay measurement
- Improved Geant4 simulations
- Planning to collect $^{161}$Tb & $^{111}$Ag in the spring

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life [days]</th>
<th>γ-ray energy [keV]</th>
<th>Current branching ratio [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{156}$Eu</td>
<td>15.2</td>
<td>811.8</td>
<td>(9.7±0.8)</td>
</tr>
<tr>
<td>$^{161}$Tb</td>
<td>6.9</td>
<td>74.6</td>
<td>(10.2±0.5)</td>
</tr>
<tr>
<td>$^{111}$Ag</td>
<td>7.5</td>
<td>342.1</td>
<td>(6.68±0.33)</td>
</tr>
<tr>
<td>$^{127}$Sb</td>
<td>3.85</td>
<td>685.5</td>
<td>(36.8±2.0)</td>
</tr>
</tbody>
</table>

Uncertainties of the order of 5-10% need to be remeasured
CARIBU Opens Up Opportunities to Study Decays of Fission Products

CAlifornium Rare Isotope Breeder Upgrade

$^{252}$Cf spontaneous fission source

Mass-separated beams of any fission product with $t_{1/2} > 25$ ms

1.0 Ci - $^{252}$Cf ($10^9$ fissions/sec)

We’re Working with ANL to perform Isomer Mass Measurements

→ Identify isomer and ground state by mass, this method is independent on decay properties

\[ E = mc^2 \]

\[ E + \Delta E_{isomer} = (m + \Delta m_{isomer})c^2 \]

Isomer mass measurements of $^{132,132m}$Sb

→ ions ejected from the trap and transported to a position-sensitive detector

→ ions in the strong magnetic field of a Penning trap, where the frequency of the ion’s cyclotron motion depends on the mass of the ion

Preliminary result:

$^{132m}$Sb $\Delta E = 145.6(1.1)$ keV
We’re Working with ANL to perform Isomer Mass Measurements

→ Identify isomer and ground state by mass, this method is independent on decay properties

Mass and energy: \( E = mc^2 \)

\[ E + \Delta E_{\text{isomer}} = (m + \Delta m_{\text{isomer}})c^2 \]

Isomer mass measurements of \( ^{128,128_m}\text{Sb} \) and \( ^{128,128_m}\text{Sn} \)

→ ions ejected from the trap and transported to a position-sensitive detector

→ ions in the strong magnetic field of a Penning trap, where the frequency of the ion’s cyclotron motion depends on the mass of the ion

Canadian Penning Trap

Precise Trap Diagram

Preliminary result:
\( ^{128_m}\text{Sb} \Delta E = 47(2) \text{ keV} \)

First Measurement!
Precision Branching Ratio Measurements of Long-lived Fission Products

Many long-lived fission products measured in 60s and 70s have high uncertainties on decay branching ratios.

- $^{156}\text{Nd}$: 5.5 s
- $^{156}\text{Pm}$: 26.7 s
- $^{156}\text{Sm}$: 9.4 h
- $^{156}\text{Eu}$: 15.2 d
- $^{156}\text{Gd}$: stable

We use radioactive beams produced at CARIBU, $\beta$-detection, and $\gamma$-ray spectroscopy to improve nuclear data.

$^{156}\text{Eu}$ strongest $\gamma$-ray intensities have ~10% uncertainties.

Sample harvesting
- Implant mass-separated radioactive ion beam on thin carbon foil at CARIBU (ANL)

Decay measurement
- $\beta$ detection and $\gamma$-ray spectroscopy at TAMU

Method described in K. Kolos et al. “New approach to precisely measure $\gamma$-ray intensities …” NIM A 1000, 165240 (2021)
We Collected High-quality Data for $^{156}$Eu

→ Data collected with precisely calibrated β-γ coincidence detection setup

→ Data collected during 7 days

→ Uncertainties >5%

→ Previous measurements used neutron capture from reactors i.e. much dirtier spectrum.

<table>
<thead>
<tr>
<th>γ-ray energy</th>
<th>$I_\gamma$ (NNDC)</th>
<th>Uncert. [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>88.97</td>
<td>8.4(11)</td>
<td>13.1</td>
</tr>
<tr>
<td>599.47</td>
<td>2.08(17)</td>
<td>8.2</td>
</tr>
<tr>
<td>646.29</td>
<td>6.3(5)</td>
<td>7.9</td>
</tr>
<tr>
<td>723.47</td>
<td>5.4(4)</td>
<td>7.4</td>
</tr>
<tr>
<td>811.77</td>
<td>9.7(8)</td>
<td>8.3</td>
</tr>
<tr>
<td>1065.14</td>
<td>4.9(4)</td>
<td>8.2</td>
</tr>
<tr>
<td>1079.16</td>
<td>4.6(4)</td>
<td>8.7</td>
</tr>
<tr>
<td>1153.67</td>
<td>6.8(6)</td>
<td>8.8</td>
</tr>
<tr>
<td>1154.08</td>
<td>4.7(4)</td>
<td>8.5</td>
</tr>
<tr>
<td>1230.71</td>
<td>8.0(7)</td>
<td>8.7</td>
</tr>
<tr>
<td>1242.42</td>
<td>6.6(5)</td>
<td>7.6</td>
</tr>
<tr>
<td>1965.95</td>
<td>3.9(3)</td>
<td>7.7</td>
</tr>
<tr>
<td>2026.65</td>
<td>3.3(3)</td>
<td>9.1</td>
</tr>
<tr>
<td>2186.71</td>
<td>3.5(3)</td>
<td>8.6</td>
</tr>
</tbody>
</table>
We Collected High-quality Data for $^{156}$Eu

→ Data collected with precisely calibrated $\beta$-$\gamma$ coincidence detection setup
→ Data collected during 7 days

$^{156}$Eu provides a unique simulation challenge

→ Simulations now tracks gamma rays emitted allowing for easier calculations and run on LLNL supercomputers
We Collected High-quality Data for $^{156}$Eu

- Data collected with precisely calibrated $\beta$-$\gamma$ coincidence detection setup
- Data collected during 7 days

With our technique we were able to improve $^{156}$Eu decay data and reach sub-1% precision!

Comparison of current (NNDC) evaluated data with our results for $\gamma$-ray branching ratios.
CARIBU Will Be Changing to nuCARIBU

- $^{252}$Cf source will be replaced by neutron-induced fission on actinide foils
- Significant gain
- Operationally easier to maintain and operate
- More reliable source of fission products
- Plan to collect $^{161}$Tb & $^{111}$Ag this Spring before switch
- Simulations already finished for $^{161}$Tb

Fission Yields

$^{161}$Tb

$^{111}$Ag

Significant loss

 deposited energy in gas counters

$\beta$-decay

JAEA Nuclear Data Center
We publicize our results at workshops, conferences, reviews:
- Independent Review in November 2020
- APS DNP 2021/2022

Publications:
K. Siegl, K. Kolos, N. D. Scielzo et al. “Beta-decay half-lives of $^{134,134m}$Sb and their isomeric yield ratio produced by the spontaneous fission of $^{252}$Cf” PRC 98, 054307 (2018)

K. Kolos, A. M. Hennessy, N. D. Scielzo et al. “New approach to precisely measure $\gamma$-ray intensities for long-lived fission products, with results for the decay of $^{95}$Zr” NIM A 1000, 165240(2021)

+ Working on publication of $^{156}$Eu (D.E.M. Hoff)
+ More from CPT measurements

Worked with many graduate students:
- Kevin Siegl (Notre Dame), B. Champine and Tyler Nagel (UC Berkeley), A. Hennessy and E. Heckmaier (UC Irvine), Erin Good (LSU), Benjamin Schroeder (TAMU)

Looking for new graduate students!

→ More experiments to come: isomeric-to-g.s. with the CPT and precision decay studies of long-lived FP branching ratios