Gamma-Ray Output Spectra from $^{239}$Pu Fission

J.L. Ullmann
(For the DANCE collaboration)

_LANSCE-NS_
_Los Alamos National Laboratory_
_Los Alamos, New Mexico USA_

Workshop on Fission Experiments and Theoretical Advances
_(FIESTA)_
Sept. 10 - 12, 2014
Santa Fe, New Mexico

LA-UR-14-27044
Collaborators and Acknowledgements


Los Alamos National Laboratory

C.-Y. Wu, A. Chyzh, J.A. Becker, J. Gostic, R. Henderson, E. Kwan

Lawrence Livermore National Laboratory

Support provided by
US DOE / NNSA
Contract DE-AC52-06NA25396
(Los Alamos National Security, LLC)
Contract DE-AC52—07-NA27344
(Lawrence Livermore National Security, LLC)
American Reinvestment and Recovery Act
**Fission Physics**

Initial fragment - high spin, excitation

Neutron decay - removes excitation

Gamma decay - (E1) removes spin and energy

Gammas - from fragment decay
  - neutron rich (?) Structure
  - Fragment mass distribution changes with neutron energy

\[ ^{239}\text{Pu}(n,f) \]
\[ \nu_n = 2.9 \]
\[ \nu_\gamma = 7.2 \] (Thermal)
Gamma-ray output from $^{239}\text{Pu}(n,fission)$

- Prompt gamma ray emission from fission not well studied
  - Only 1 published spectrum for $^{239}\text{Pu}(n,f)$ – at thermal
  - (V.V. Verbinski, Phys. Rev. C 7, 1173 (1973) )
  - Other measurements – but do cover wide gamma energy range

- Experiment at LANSCE moderated white neutron source
  - Need fission tagging – use LLNL/LANL PPAC
  - Gammas detected using Detector for Advanced Neutron Capture Experiments (DANCE) “4π” detector
  - Gammas $\pm$ 20 ns from fission event
  - Direct measurement of multiplicity and total energy distribution
DANCE gamma-ray calorimeter

- 160 BaF$_2$ crystals – each 0.75 liter
- Inner radius = 17 cm, crystal depth = 15 cm
- $^6$LiH inner sphere to absorb scattered neutrons
- Internal conversion plus absorption in LiH may affect low-energy gamma spectrum
DANCE and LANSCE

DANCE ball
(Open)
$^6$LiH sphere in center

(Los Alamos Neutron Science Center (LANSCE))
Fission tagging using PPAC

LLNL / LANL / MSI PPAC
4.37 cm dia X 4.77 cm long

Fission efficiency ~ 70%

PPAC Target Assembly

PPAC Params
- Gas = Isobutane
- P = 4.5 Torr
- Flow = 4 cc/m
- $^{239}$Pu (total) = 2.43 mg/cm$^2$
- (0.7 cm dia deposit)
- 99.967% enriched
**DANCE response correction crucial**

Single gamma efficiency $\epsilon = 0.85$

7 gammas $\Rightarrow \epsilon = (0.85)^7 = 0.40$

**Methods of response correction**


“Inverse Methods” – solve $O = R \, I$ for input spectrum $I$

1-dimensional or 2-dimensional $O$ and $I$


1D: $E_\gamma$, Mult each unfolded

2D: Unfold $E_{tot}$ vs Mult Matrix

“Forward Methods” – Assume spectra, simulate response and compare

Iterate spectra until fit

Ultimate - use a real physics model with parameters

Experimental approach - don’t depend on physics model

Parameterize data analytically

**NOT (!!) a physics model (but may be motivated by physics)**
Detector Response Correction

(Preliminary results shown in this figure!)

\[ \epsilon = 0.85 \quad \text{(Geometric)} \]

\[ \epsilon = (0.85)^7 \quad \text{for 7 gamma rays} = 0.40 \]
Parameterized fission spectra

**Multiplicity**
- Sum over two distributions
  \[ M_\gamma = M_1 + M_2 \]
- Assume Multiplicity ~ Spin distributions \( P(J) \sim (2J+1)e^{-J(J+1)/B^2} \) 
  (Wilhelmy, Phys Rev C 5, 2041 (1972) )

**Gamma energy distribution**
- \( P(\varepsilon) \sim T(\varepsilon) \rho(\varepsilon) \quad T(\varepsilon) \sim A \varepsilon^3 \) (E1), \( \rho(\varepsilon) \sim Be^{a(E_0-\varepsilon)} \)
- Best fit:
  \[ P_1(\varepsilon) \sim \varepsilon^2e^{-(a_1+M_\gamma b_1)\varepsilon} \]
  \[ P_2(\varepsilon) \sim \varepsilon^3e^{-(a_2+M_\gamma b_2)\varepsilon} \]  
  (Fit params: \( a_1, a_2, b_1, b_2 \))
- Observed Gamma spectrum is sum over many fission products
  - Different excitation energies, temperatures, multipolarity
- Parameterization - not a physics model
Procedure for fitting parameters

- Generate event consisting of $M_\gamma = M_1 + M_2$ gamma rays with energy $\varepsilon_i$ from $P(M_{1,2})$ and $P_{1,2}(\varepsilon_\gamma)$
- Transport all gammas from event through GEANT4 model of DANCE to produce “experimental” values
- Compare to measured values – calculate $\chi^2$
  - $M_{cr}$ chosen to avoid overlapping clusters
  - $M_{cr}$ easily simulated by GEANT
  - $E_{tot}$ (Tresh = 150 and 400 keV)
  - $M_{cr}$ (150 keV Threshold)
  - $E_\gamma$ (150 keV threshold)
  - More weight to higher threshold
- Vary parameters at random to find minimum $\chi^2$
  - Iterated “Simulated Annealing” technique
  - Vary parameters over range $1 \pm \delta$
  - $P(\Delta) = e^{-\Delta/T}$
Detour: Fission neutron response

DANCE response to fission-spectrum neutrons

Total Efficiency for neutrons 1.3%
\(^{239}\text{Pu}(n,\text{fission})\) Cross Section

Current results gated on 10.93 + 11.89 eV 1\(^+\) resonances
Thermal, 7.82, 22.26, 75.0 1\(^+\) and 32.33 0\(^+\) resonances
have similar spectra
Best-fit parameters

\[ P(M_\gamma) = P(M_1) + P(M_2) \]

\[ P(M_1) = (2M_1 + 1)e^{-M_1(M_1+1)/B_1^2} \]

\[ P(M_2) = (2M_2 + 1)e^{-M_2(M_2+1)/B_2^2} \]

\[ P_1(\varepsilon_\gamma) = \varepsilon^2 e^{-(a_1 + M_\gamma b_1)\varepsilon} \]

\[ P_2(\varepsilon_\gamma) = \varepsilon^3 e^{-(a_2 + M_\gamma b_2)\varepsilon} \]

\[ \begin{align*}
B_1 &= 6.66 \pm 0.15 \\
B_2 &= 2.54 \pm 0.12 \\
a_1 &= 3.80 \pm 0.12 \\
a_2 &= 1.53 \pm 0.14 \\
b_1 &= 0.0428 \pm 0.0019 \\
b_2 &= 0.0522 \pm 0.0034 
\end{align*} \]
Raw Data vs Parametrized fit

(All spectra for 150 keV $E_{cr}$ threshold)
Theoretical Calculations

Monte-Carlo Hauser-Feshbach Model


- Fragment mass distribution semi-empirical
- Radiative strength functions from RIPL-3
- Level density was Gilbert-Cameron formalism
- Spin Distribution:

\[ P(J) = (2J+1)e^{-J(J+1)/2B^2} \]

\[ B = \alpha \frac{I_o T}{\hbar^2} \]

- \( I_o \) = ground-state moment of inertia
- \( T \) = fragment temperature
- \( A \) = adjustable parameter
Results – Gamma-ray Multiplicity

$^{239}$Pu(n,f) Measured
CrystalMultiplicity very sensitive to threshold

Average Raw Crystal Multiplicity

<table>
<thead>
<tr>
<th>Threshold (keV)</th>
<th>&lt;Mcl&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>150</td>
<td>7.2</td>
</tr>
<tr>
<td>300</td>
<td>5.7</td>
</tr>
<tr>
<td>400</td>
<td>4.7</td>
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</tbody>
</table>

$^{239}$Pu(n,f) Corrected $\gamma$ Multiplicity

- 150 keV Threshold
- MCHF from Stetcu, LA-UR-14-23128 ($\alpha = 1.5$)
Results: $^{239}$Pu(n,f) Gamma Energy

Measured (before response correction) cluster energy for Cluster Multiplicities 4, 8, 12

- Response-corrected gamma energies (all multiplicities)
- MCHF Calculation (Stetcu)
- Unfolded 1D (Chyzh)
Results: $^{239}$Pu(n,f) Total gamma-ray energy

- Response-corrected gamma energies (all multiplicities)
- MCHF Calculation (Stetcu, $\alpha = 1.5$)
### $^{239}\text{Pu}(n,f)$ Average $\gamma$ Multiplicity and $E_{\text{tot},\gamma}$

<table>
<thead>
<tr>
<th>Source</th>
<th>$&lt;M&gt;$</th>
<th>$&lt;E_{\text{tot},\gamma}&gt;$</th>
</tr>
</thead>
<tbody>
<tr>
<td>This Work (10.93 eV)</td>
<td>7.15 ± 0.09</td>
<td>7.46 ± 0.06</td>
</tr>
<tr>
<td>Pleasonton (thermal)</td>
<td>6.88 ± 0.35</td>
<td>6.73 ± 0.35</td>
</tr>
<tr>
<td>Verbinski (thermal)</td>
<td>7.23</td>
<td>6.81</td>
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<td>Verbinski (thermal)</td>
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<tr>
<td>Verbinski (thermal)</td>
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<td></td>
</tr>
<tr>
<td>MCHF (Stetcu/Talou)</td>
<td>7.05</td>
<td>6.74</td>
</tr>
<tr>
<td>Madland Summary</td>
<td></td>
<td>6.74</td>
</tr>
<tr>
<td>Chyzh, “1D” Unfolding</td>
<td>7.50</td>
<td>7.30</td>
</tr>
<tr>
<td>Chyzh, “1D” Unfolding</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chyzh “2D” Unfolding</td>
<td>7.93</td>
<td>7.94</td>
</tr>
<tr>
<td>Chyzh “2D” Unfolding</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- Uncertainty in $<E_{\text{tot}}>$ ~ Fitting uncertainty, determined as $\sigma$ of 14 iterations with lowest $\chi^2$
- $<M>$ very sensitive to detection threshold!
Unexpected detail: \((n,\gamma f)\)?

Resonances we studied looked to be quite similar – but is that universally true? (S. Mosby - \(^{239}\text{Pu}(n,\gamma)\) analysis)

\(<n_n>\) dips at several weak \(L=0\) resonances

- 35.5 eV 1+
- 41 1+
- 44.5 1+
- 52.7 1+

Neutron multiplicity measurements

Gamma multiplicity measurements
Resonance properties – Raw data

Gamma multiplicity

Total Gamma Energy
Summary and Conclusions

- Measurements of distribution of multiplicity, gamma energy, and total gamma energy (“forward modeling” parameterization of data) - Needs high-segmentation, $4\pi$ capability !!
- Average multiplicity in agreement with previous measurements - sensitive to thresholds
- Average $E_{\text{tot}} \sim 10\%$ higher than previous
- Theoretical modelling reproducing $^{239}\text{Pu}$ data, but with adjustable parameters
- Still some puzzles: $(n,\gamma f)$?
- “More work to be done!”
Why study fission gammas?

• Applications - need to know distribution of gamma-ray multiplicity, gamma energy, total gamma energy
  – Reactors: heating, decay heat
  – Non proliferation
  – Illicit nuclear materials (portal monitors)

• Physics of fission
  – Fission products - high-spin, neutron-rich
  – Decay properties constrain models
<E_\gamma> Changes with neutron energy

- Fragment mass distribution changes with neutron energy
- Madland formula reflects changing products and J (excitation)
  \[ E_{\text{tot}} = 6.741 + 0.117 T_n \text{(MeV)} - 0.0002 T_n^2 \text{ MeV} \]
- Low energies resonances – no significant change in gamma properties
  (Thermal and 100 keV should have similar <E_\gamma> )

PPAC Performance

**PPAC Pulse Height**

- **h_PPAC_E2**
  - Entries: 86925
  - Mean: 403.8
  - RMS: 166.5

- **PPAC Efficiency**
  - PPAC Threshold = 350

- **Run**:
  - Values range from 27110 to 27150.
Measure Mult ($\nu\gamma$), $E_{\text{gam}}$, $E_{\text{tot}}$ and Neutron energy

No fission tag
(Capture + (1-$\epsilon$) Fission)
Response Correction / Unfolding

• Philosophy – experimenters want to measure something,
  • Not just fit parameters to a theory
• Forward method – not a physics model
  • Parameters motivated by physics
  • but really only parameters
  • Represent data
Detour: Fission neutron response

Fission neutrons – Ave energy ~ 2 MeV
BUT – High energy tail! (Maxwellian)

Fission neutron effects – MCNP by TNT
Transport $^{252}$Cf fission neutron spectrum into DANCE

 DIMENSIONS:

$^6$LiH sphere = 10.50 cm (id)
16.51 cm (od)
0.85 g/cm$^3$

$^{252}$Cf sphere = 17.00 cm (id)
32.00 cm (od)
4.88 g/cm$^3$

$^{252}$Cf point source at center

(MCNP Calculations by T. Taddeucci)
Detector Response Function

Multiplicity

• Sum over two distributions
  \[ M_y = M_1 + M_2 \]

• Spin distributions \( P(J) \sim (2J+1)e^{-J(J+1)/B^2} \)
  (Wilhelmy, Phys Rev C 5, 2041 (1972) )

• Assume \( P(M) = P(J) \) (Number of gammas = spin)
  (Good for E1, M1 roughly)

• 2 fission products \( \Rightarrow P(M_y) = P(M_1) + P(M_2) \)

B’ s are fitted - M’ s (J’ s) are random variables
Detector Response Function

**Gamma energy distribution**

- $P(\epsilon) \sim T(\epsilon) \rho(\epsilon)$
  - For E1 transitions: $T(\epsilon) \sim A \epsilon^3$
  - “Constant Temperature” $\rho(\epsilon) \sim B e^{aE_x} = B e^{a(E_0 - \epsilon)}$
  - $P(\epsilon) \sim \epsilon^3 e^{-a\epsilon}$

- Lemaire calculation: $P(\epsilon) \sim \epsilon^2 e^{-\beta \epsilon}$

- Best fit:
  - $P_1(\epsilon) \sim \epsilon^2 e^{-(a_1 + M_1 b_1)\epsilon}$
  - $P_2(\epsilon) \sim \epsilon^3 e^{-(a_2 + M_2 b_2)\epsilon}$
    (Fit params: $a_1$, $a_2$, $b_1$, $b_2$)

- Observed Gamma spectrum is sum over many fission products
  - Different excitation energies, temperatures, multipolarity

**Details:**
M. Jandel et al., Los Alamos Report LA-UR-12-24975
Multiplicity dependence of $E_{\text{tot}}$ and $E_{\gamma}$

- **$E_{\gamma}$**: no strong multiplicity dependence
- **$E_{\text{tot}}$**: $E_{\text{tot}}$ roughly proportional to multiplicity
Multiplicity sensitive to DANCE threshold

Multiplicity, total energy with different thresholds determined from MCHF calculation
High-energy gammas

Changes to Analysis 29-Aug-12

• Parameterization
• Method fitting: “Metropolis Algorithm” + Simulated Annealing
• Parameters fitted for Chi-square minimization
  • 7 spectra (P9VA2MD)
    • Overestimates high-energy effect
  • 4 Spectra (P9VA2ME)
• Uncertainties in $\langle E \rangle$, $\langle M \rangle$
  • New method of minimization implies cannot easily use previous technique for estimating uncertainties
• Use % Std of 14 best-fit (lowest Chi2) iterations
• BUT - use Value of best-fit iteration!
• Effect of threshold on measured multiplicity
Previous Measurements


Other measurements – incomplete Gamma energy range

• “Unfolding” of measured spectrum critical to results
• Pleasonton also determined fission product ID from Doppler shift.
PPAC Assembly

- Front cover with assembled polyimide rings
- Counter container
- Back cover with the Kapton window and gas feedthrough
- The assembled polyimide ring for one of two anodes
- Flexible cable for the anode signal transmission

Fig 4. Various counter parts before the final assembling.