Gamma-Ray Emission Spectra as a Constraint on Calculations of $^{234,236,238}$U Neutron-Capture Cross Sections


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Capture cross-sections and spectra important

- Neutron capture cross sections in the “continuum” region $\gtrsim 1\text{ keV}$ and gamma-emission spectra of importance to basic science and many applied fields
  - Defense
  - Nuclear security, safeguards, and forensics
  - Nuclear power
  - $r$- and $s$- process nucleosynthesis
- Careful measurements have been made on most common stable nuclides
- But must rely on calculations (or “surrogate” reactions) for rare or unstable nuclides
- Must benchmark calculations against measurements
  - Cross sections
  - Another observable: gamma-ray spectrum
  - $\langle \Gamma_\gamma \rangle$
Digression: Some theory background

Ultimate Goal: Calculate capture cross sections
(Especially true – unstable nuclides)

Capture cross section calculated by Hauser-Feshbach approximation;

$$\sigma_\gamma \approx \frac{T_{in}}{k^2} \pi W \frac{\sum_{\text{Discrete}} T_{out}(\varepsilon_\gamma)}{\sum T_{out}} + \int_{E_{crit}} T_{out}(E - E_x) \rho(E_x) dE_x$$

$$T_{XL}(\varepsilon_\gamma) = 2\pi \varepsilon_\gamma^{2L+1} f_{XL}(\varepsilon_\gamma)$$

Shape usually OK, magnitude often X2 to X10 error

Strength function normalized to measured average capture width \(\langle \Gamma_\gamma \rangle\)
> Need to know \(\langle \Gamma_\gamma \rangle\) !!

$$\frac{2\pi}{D_o} \langle \Gamma_\gamma \rangle = C \int_0^E d\varepsilon_\gamma (2\pi \varepsilon_\gamma^3) f_{E1}(\varepsilon_\gamma) \rho(E - \varepsilon_\gamma)$$
The DANCE array

Detector for Advanced Neutron Capture Experiments

- 162 segments with 4 different shape crystals (160 segments with crystals)
- Calorimetric detector (nearly $4\pi$) - detects full energy of decay cascade
- High efficiency and high neutron flux allows measurements on milligram samples
- Highly segmented to allow detection of radioactive targets
- Inner radius = 17 cm
- Crystal depth = 15 cm
- State-of-the-art fast transient digitizers for data acquisition - CAEN VX1730B
- $^6$LiH inner sphere to absorb scattered neutrons

Half of DANCE array with $^6$LiH ball
Neutron Capture

Detect ALL gammas-
- Summed energy is $Q$ value ($S_n$) + neutron cm $T$
- $E_x = T_n(1+M_n/M_A)+Q$

$^{138}$Ba($n,\gamma$) $Q = 4.72$ MeV

$^{134,136}$Ba($n,\gamma$) $Q = 6.9$ MeV

$^{135,137}$Ba($n,\gamma$) $Q = 9.11,8.62$ MeV

$^{197}$Au($n,\gamma$) $Q = 6.51$ MeV

$T_{\text{neut}} = 1$ to 10 keV
Neutron Flux Measurement

3 Neutron Monitors
$^6$Li(n,αt) 22.60 m
$^{10}$B(n,α) 22.76 m
$^{235}$U(n,f) 22.82 m

Flux from $^6$Li:
~547 E$^{-1.04}$
\( \text{n/cm}^2/\text{eV/To} \)
(at monitor)
I = 100 \( \mu \text{A} \)
(5-Aug-10)

Monitors are 2 m downstream of target
• Attenuation by target
• Geometric solid angle
• Targets often < beam
Los Alamos Neutron Science Center (LANSCE)
Summary – measurements on $^{234,236,238}$U(n,γ)

- Gamma-ray spectrum measurements from resolved resonances made with 1 - 2 mg/cm² thick targets
- Cross sections > 1 keV measured using thicker targets

### Uranium Measurements

<table>
<thead>
<tr>
<th>Isotope</th>
<th>234</th>
<th>236</th>
<th>238</th>
</tr>
</thead>
<tbody>
<tr>
<td>Q window (MeV)</td>
<td>5.30 ± 0.50</td>
<td>5.13 ± 0.50</td>
<td>4.81 ± 0.65</td>
</tr>
<tr>
<td>γ-spectrum target</td>
<td>1.0 mg/cm²</td>
<td>1.29 mg/cm²</td>
<td>2.27 mg/cm²</td>
</tr>
<tr>
<td>σ target</td>
<td>(2015)</td>
<td>2014 – Analysis In progress</td>
<td>48 mg/cm² PRC 89,034603</td>
</tr>
</tbody>
</table>
Uranium s-wave resonances

\[ ^{234}\text{U}(n,\gamma) \]
\[ ^{236}\text{U}(n,\gamma) \]
\[ ^{238}\text{U}(n,\gamma) \]

Counts

Neutron Energy (eV)

\[ \ell=0 \quad \frac{1}{2}^+ \]

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$^{238}$U Background subtraction – 36 eV resonance (M=2)

**Principal Backgrounds**

- $\beta^-$ decay $^{226}$Ra chain
  - $^{214}$Bi $Q=3.27$ MeV
  - $^{214}$Pb $Q=1.03$ MeV
- $^{138}$Ba $Q=4.72$ MeV

- $^{138}$Ba $Q=4.72$ MeV

**E$_{\text{cluster}}$ (gated by Q,T$_{\text{neut}}$)**

**E$_{\text{sum}}$ (gated by T$_{\text{neut}}$)**
Multiplicity = 2 Gamma ray spectra
Calculate capture gamma-ray spectrum

Formula for spectrum - Monte Carlo DICEBOX algorithm

Level density - Constant Temperature
Parameters from von Egidy and Bucurescu Phys Rev C 72, 044311 (2005)

Radiative strength function - Simplest model is E1 only
• Parameters usually taken from (γ,n)
• Q value (Sn) ≈ 4.81 MeV (²³⁸U)
• What is function between GS and Sn?
• Different models - (Kopecky and Uhl, Phys Rev C41, 1941 (1990))
• Use “Modified Generalized Lorentzian” (MGLO)

\[ f(\varepsilon_\gamma) = \frac{\sigma_r \Gamma_r}{3\pi^2 (\hbar c)^2} \left[ \frac{\varepsilon_\gamma \Gamma_\gamma(\varepsilon_\gamma, T)}{(\varepsilon_\gamma^2 - \varepsilon_r^2)^2 + \varepsilon_\gamma^2 \Gamma_\gamma^2(\varepsilon_\gamma, T)} + \frac{(0.7)4\pi^2 \Gamma_r T^2}{\varepsilon_r^5} \right] \]
Calculate capture gamma-ray spectrum

Nucleus rich in different collective modes of vibration
- M1 Scissors
- M1 Gamow-Teller
- E1 Pygmy

Usually parameterized as “Standard Lorentzian” (SLO)

\[ f(\varepsilon_\gamma) = \frac{1}{3\pi^2(\hbar c)^2} \frac{\sigma_0 \varepsilon_\gamma \Gamma_r^2}{(\varepsilon_\gamma^2 - \varepsilon_r^2)^2 + \varepsilon_\gamma^2 \Gamma_r^2} \]

- Lots of work to characterize these modes using different probes
  Probes have varying sensitivity
- Parameters determined by “Oslo Method” appear to provide consistent description of neutron capture (MK)
- Comparing results from different probes leads to question: “Is there a universal strength function?”
Gamma spectrum provides constraint on RSF

Test radiative strength function by “Spectrum Fitting Method”

• Assume form of Radiative Strength Function

• Generate gamma cascades using Monte Carlo
  • DICEBOX (F. Bečvář, NIM A 417, 434 (1998).)
  • Uses level density above $E_{\text{crit}}$ (~ 0.5 MeV)
  • Tabulated level information below $E_{\text{crit}}$

• Process cascades through GEANT4 model of DANCE
  • Use formulation of Jandel
    (M. Jandel, et al., NIM B 261, 1117 (2007).)
  • Based on original model by Reifarth and Heil
    (M. Heil, R. Reifarth, et al., NIM A 459, 229(2001).)

• Compare to observed gamma-ray spectra − qualitative!!
  • Consider low-lying resolved resonances and gate on Q-value (summed energy) spectrum to reduce backgrounds
  • Background subtraction still needed
$^{238}$U Capture Results – $M_{cl} = 2$

$^{238}$U($n,\gamma$) $M_{cl}=2$ Spectrum

CT Level Density

<table>
<thead>
<tr>
<th>Source</th>
<th>$\Gamma$</th>
<th>$\sigma_1$</th>
<th>$\Gamma_1$</th>
<th>$\Gamma_2$</th>
<th>$\sigma_2$</th>
<th>$\Gamma_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>GDR Berman (RIPL-3)</td>
<td></td>
<td></td>
<td>2.48</td>
<td></td>
<td>384.0</td>
<td>4.25</td>
</tr>
<tr>
<td>M1 Scissors Oslo PRL 109 ($^{233}$Th)</td>
<td></td>
<td></td>
<td>0.85</td>
<td>0.60</td>
<td>0.70</td>
<td></td>
</tr>
<tr>
<td>M1 Scissors Oslo PRC 89 ($^{237}$U)</td>
<td></td>
<td></td>
<td>0.80</td>
<td>0.40</td>
<td>0.60</td>
<td></td>
</tr>
<tr>
<td>M! Spin Oslo PRC 89 ($^{237}$U)</td>
<td></td>
<td></td>
<td>4.00</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>E1 Pygmy Oslo PRC 89 ($^{237}$U)</td>
<td></td>
<td></td>
<td>2.00</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Counts vs. \gamma-Ray Energy (MeV)

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$M_{cl} = 2$ Gamma-ray spectra Calculations

- $^{236}\text{U}$
- $^{234}\text{U}$

<table>
<thead>
<tr>
<th></th>
<th>$E_1$</th>
<th>$\sigma_1$</th>
<th>$\Gamma_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>&quot;Consistent&quot; M1 Scissors</td>
<td>2</td>
<td>0.60</td>
<td>4.7</td>
</tr>
<tr>
<td>&quot;Consistent&quot; M1 GT</td>
<td>7</td>
<td>3.00</td>
<td>4.7</td>
</tr>
</tbody>
</table>

+ SLO E1 GDR

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Background subtraction – 1 – 10 KeV

The graph shows the distribution of gamma energy (MeV) with counts on the y-axis. Different materials are represented with distinct markers and line styles:

- **Solid line**: $^{238}$U
- **Dotted line**: $^{208}$Pb
- **Red dot**: No target
- **Green triangle**: No beam

The y-axis range is from $10^{-4}$ to $10^4$ counts, and the x-axis represents gamma energy from 0 to 12 MeV.
Normalize to weak resonances

Graph

R12792 Normalization Resonances (n=1.39)

Energy

10^3

10^2

10^1

1

10

10^{-1}
Cross Sections

$^{236}\text{U}(n,\gamma)$

$^{234}\text{U}(n,\gamma)$

Cross Section (barns)

- ENDF/B-VII.1
- CoH$_4$ w/PRL 109
- CoH$_4$ w/PRC 89
- Adamchuk _88
- Buleeva _88
- Carlson _70
- Guiskov _66

Cross Section (barns)

- ENDF/B-VII.1
- CoH$_4$ w/PRL 109
- CoH$_4$ w/PRC 89
- Rundberg _2005
Results

- Shape of capture cross section vs neutron energy is not sensitive to form of strength function (although magnitude is).

- Generalized Lorentzian E1 strength function is not sufficient to describe shape of observed gamma-ray spectra.

- $MGLO$ + “Oslo M1” parameters produces quantitative agreement with measured $^{238}U(n,\gamma)$ cross section.

- Additional strength at low energies ($\sim 3$ MeV) – likely M1- is required (See also Guerrero (nTOF): Jour. Korean Phys. Soc. 59, 1510 (2011) (ND2010)).

- Careful study of complementary results on low-lying giant resonance strength is needed to consistently describe observations.
  - M1 - (Heyde, Rev. Mod. Phys. 82, 2365, 2010)
  - Pygmy E1 dipole
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