Inelastic scattering on $^{232}\text{Th}$: measurements and beyond

E. Party
3rd year PhD in DNR team

Plan

• I. Context
• II. Method
• III. Experiment
• IV. Analysis
• V. Results
• VI. Additional works
I. Context – Inelastic scattering on $^{232}$Th and reactor applications

Why inelastic scattering?
Nuclear reactions occurring in reactors:
- $(n,g);(n,f);(n,\text{el});(n,\text{inl});(n,x)$
- **Inelastic scattering**: main slowing down factor in fast neutron reactor
- Reactor neutronic simulation needs precise nuclear data
- Experimental data on some reactions lack precision, and may be inexistant
- **Realistic covariances** are needed for **evaluation** process and propagation of uncertainty

Why Thorium?
Material exposed to neutrons:
- Nuclear fuel
- Moderator
- Structural materials
- $^{232}$Th: fertile isotope, possible nuclear fuel in combination with $^{233}$U
- Even for non-Th fueled reactor, measurements may be of use for a systematic theory of inelastic scattering
I. Context – State of measurements on $^{232}\text{Th}$

- Scarce measurement
- Almost no points for neutron energy $> 3$ MeV for partial (level or gamma) cross sections
- There is a need for more measurement at higher energy
II. Method – Principle of measurement

\((n,n')\) can be measured using:

1. Neutron scattered – Direct detection
   - Most useful for reactor application, as energy loss and angular distribution are directly observable
   - Difficulties to measure precisely and separate contributions from \((n,e)\), \((n,f)\),...
   - Energy resolution is poor

2. Deexcitation of nucleus – Gamma spectroscopy
   - Relatively easy to observe
   - High energy resolution using HPGe counters allows to distinguish close levels
   - As \(Z\) is high, unseen deexcitation by conversion electron may be a problem
   - May not allow direct measure of excitation energy (~neutron energy loss), then it is dependent on nuclear structure to obtain level- and total XS
III. Experiment – GELINA neutron production facility

GELINA neutron production facility:

- Located at EC-JRC Geel, Belgium
- 800 Hz pulsed neutron beam
- Electron induced photofission neutron from uranium target
- Neutron energy range from few keV to 20 MeV
- 12 flight path from 5 to 400 meter long

Birdview of GELINA facility
III. Experiment – GRAPhEME setup for $^{232}\text{Th}$ campaign

GRAPhEME (GeRmanium array for Actinides PrEcise Measurements):

- 4 HPGe detectors placed at 110° and 150° of beam axis detect gamma rays produced
- One fission chamber placed ahead of the target determine neutron beam fluence
- Digital acquisition cards TNT2-100 MHz
- $^{232}\text{Th}$ target of width $e=0.3$ mm located at circa 30 m from neutron source
- Neutron energy obtained using time of flight measurement (resolution 0.8 MeV at 20 MeV, 10 keV at 1 MeV)

GRAPhEME as used for $^{232}\text{Th}$ data collection (2009-2013)
III. Experiment – Cross section measurements

Differential cross section formula

\[
\frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(\theta_i,E_n) = \frac{N\gamma(\theta_i,E_n)\tau_{pup,i}/\epsilon\gamma,i}{4\pi N_n(E_n) \rho_{Th}}
\]

\(N\gamma(\theta_i,E_n)\) : Number of \(\gamma\)-rays of interest detected at angle \(\theta_i\) for a time of flight equivalent to \(E_n\)
\(\tau_{pup,i}\) : Corrective factor due to pile up dead time
\(\epsilon\gamma,i\) : Efficacy of HPGe counter for considered \(\gamma\)-ray energy
\(\rho_{Th}\) : Areal atomic density of thorium target
\(N_n(E_n)\) : Number of impinging neutrons on thorium target for a time of flight equivalent to \(E_n\)

\[
N_n(E_n) = \frac{N_{fiss}(E_n)/\epsilon_{FC}}{\sigma_{f,U235}(E_n) \rho_{U235}}
\]

\(N_{fiss}(E_n)\) : Number of fissions detected for a time of flight equivalent to \(E_n\)
\(\epsilon_{FC}\) : Efficacy of fission chamber
\(\sigma_{f,U235}(E_n)\) : Fission cross section of U-235 for neutron energy \(E_n\)
\(\rho_{U235}\) : Areal atomic density of U-235 foil in fission chamber
IV. Analysis – Identification

Identification of gamma peaks, in nuclear structure databases, using:

- 1) Gamma energy centroid
- 2) Time of flight where it has been observed

- Bidimensional spectrum is projected on several time of flight windows, each corresponding to a neutron energy interval
- Area under peak for each projection is needed to compute cross section on corresponding neutron energy interval
IV. Analysis – Total cross section

Combination of 4 HPGe measures:

- 2 times 2 partial cross sections at 110° et 150° from beam axis
- We need angle integrated cross section
- As transitions' multipolarity is always less than 3, following Gauss quadrature formula gives exact angle integrated cross section:
  \[
  \sigma_{(n,xn \gamma)} = 4\pi \left[ w_{110} \frac{d \sigma_{(n,xn \gamma)}^{(110^\circ, E_n)}}{d\Omega} + w_{150} \frac{d \sigma_{(n,xn \gamma)}^{(150^\circ, E_n)}}{d\Omega} \right]
  \]
- This formula is the reason why HPGe were placed at 110° and 150°

Mean of 2 measures at one angle:

- Measures from 2 HPGe for one angle
- They are correlated \((\rho_{th}, N_n(E_n)\) terms are same, \(\varepsilon_{det, r}\) may be correlated too).
- To maximise information, we need to use ponderated mean, using covariances
- Minimal uncertainty is achieved by using the following estimator \(\sigma(\theta)\) to combine two measures \(\sigma_A, \sigma_B\) at the same angle \(\theta\):

\[
\sigma(\theta) = \frac{1}{\text{Var}(\sigma_A) - \text{covar}(\sigma_A, \sigma_B)} \frac{\sigma_A + \frac{1}{\text{Var}(\sigma_B) - \text{covar}(\sigma_A, \sigma_B)} \sigma_B}{\text{Var}(\sigma_A) - \text{covar}(\sigma_A, \sigma_B)} \frac{1}{\text{Var}(\sigma_B) - \text{covar}(\sigma_A, \sigma_B)}
\]
IV. Analysis – Uncertainties

Typical uncertainties (1σ values):
1. From U-235 foil width $\rho_{\text{U}235}$: 0.55 %
2. On target aeral density $\rho_{\text{Th}}$: 1.3 %
3. From fission chamber efficacity $\varepsilon_{\text{FC}}$: 2.1 %
4. On HPGe photopeak efficacity $\varepsilon_{\gamma,i}$: 2 %
5. From $\sigma_{f,\text{U}235}(E_{n})$: 0.5 to 1 %
6. Statistical and fit uncertainty on $N_{\gamma}(\theta_{i},E_{n})$: 2 to 20 %, depending on $\gamma$-ray observed and neutron energy
7. On correction factor due to pile up $\tau_{\text{pup},i}$: 0.5 %
8. From statistical error on $N_{\text{fiss}}(E_{n})$: 0.3 to 2 %

→ Minimal total uncertainty: 3.5 %
→ Including 3.3 % fully correlated over all $E_{n}$

Differential cross section formula

\[
\frac{d\sigma_{(n,xn\gamma)}}{d\Omega}(\theta_{i},E_{n}) = \left(\frac{N_{\gamma}(\theta_{i},E_{n})/\varepsilon_{\gamma,i}}{\tau_{\text{pup},i}\rho_{\text{U}235}\sigma_{f,\text{U}235}(E_{n})}\right) \cdot 4\pi \left(\frac{N_{\text{fiss}}(E_{n})/\varepsilon_{\text{FC}}}{\rho_{\text{Th}}}\right)
\]

Different measures' points are identified by:
- Neutron energy range observed $E_{n}$
- $\gamma$-ray considered
- HPGe counter number $i=\{1,2,3,4\}$

Variance is obtained by classic error propagation

Covariance between two measure points has been obtained the covariance of each terms in both points formula
V. Results – level scheme of $^{232}$Th

\[ 0.0(0+) \quad 49.4(2+) \quad 162.1(4+) \quad 333.3(6+) \quad 556.9(8+) \quad 1137.1(12+) \]

\[ 714.4(12+) \quad 826.8(10+) \quad 774.4(3-) \quad 774.1(2+) \quad 730.6(12+) \]

\[ 883.8(5-) \quad 960.8(5+) \quad 1050.9(6+) \quad 1042.9(7-) \]

\[ 1053.9(2+) \quad 1078.6(0+) \quad 1094.4(3+) \]

\[ 1072.4(2-) \quad 1077.9(1-) \quad 1072.4(2-) \]

\[ 1146.3(7-) \quad 1146.8(7+) \quad 1148.3(9+) \quad 1145.3(4-) \]

\[ 1182.6(3-) \quad 1218.1(4+) \quad 1208.8(5-) \quad 1218.1(4+) \]

\[ 1222.1(6+) \quad 1258.7(8+) \quad 1328.6(5-) \quad 1322.2(2+) \]

\[ 1387(2+) \quad 1489(1?) \quad 1490(5+) \quad 1553(2+)? \]

\[ 1609(2?) \quad 1480(?) \quad 1647(?) \quad 1578(2?) \quad 1581(1,2+) \]

\[ 1609(2?) \quad 1480(?) \quad 1647(?) \quad 1578(2?) \quad 1581(1,2+) \]

\[ 1573(1,2+) \quad 1578(2?) \quad 1618(?) \]

\[ 1498.7(11-) \quad 1469.3(10+) \]

\[ 1370(9+) \quad 1249.6(9-) \quad 1222.1(8+) \quad 1258.7(8+) \]

\[ \sigma(n,n'\gamma) \text{ obtained} \]

\[ \text{Weak or unseen} \]

\[ \text{E0} \]

\[ \text{From Demidov (Phys. At. Nucl. 71, 1839 (2008))} \]

\[ 30 \text{ first levels} \]

\[ \gamma: 4^+4 \]

\[ \text{Non-band levels} \]

Rotational bands from Demidov (2008)
V. Results – level scheme of $^{232}$Th

Spin assignment uncertain

Missing branching ratio in NNDC

Erroneous branching ratio

1.2 MeV

30 first levels

J=0  J=1  J=2  J=3  J=4  J=5  J=6  J=7  J=8  J=9  J=10

1561(1,2+)  1573(1.2+)  1489(1?)  1450(?)  1303(?)

1578(2+)  1585(2+)  1609(?)

1480(?)  1387(2+)  1322(2+)

1553(2+)  1618(?)  1647(?)

1490(5+)

1413.8(4+)

1328.8(5+)

1208.8(5-)

1121.7(2+)

1106.7(3-)  1133.3(4+)

1293(+)

1237.6(5+)

1208.8(5-)

966.2(5+)

883.8(5+)

1050.9(6+)

1023.3(6+)

1042.9(7-)

1122.1(8+)

1222.1(8+)

1249.6(9+)

1237.6(5+)

1146.3(7+)

1370(9+)

1469.3(10+)

1498.7(11+)

1482.2(14+)

1413.8(4+)

1490(5+)

1121.7(2+)

1106.7(3-)  1133.3(4+)

1293(+)

1237.6(5+)

1208.8(5-)

966.2(5+)

883.8(5+)

1050.9(6+)

1023.3(6+)

1042.9(7-)

1122.1(8+)

1222.1(8+)

1249.6(9+)

1237.6(5+)

1146.3(7+)

1370(9+)

1469.3(10+)

1498.7(11+)

1482.2(14+)

1413.8(4+)

1490(5+)

1121.7(2+)

1106.7(3-)  1133.3(4+)

1293(+)

1237.6(5+)

1208.8(5-)

966.2(5+)

883.8(5+)

1050.9(6+)

1023.3(6+)

1042.9(7-)

1122.1(8+)

1222.1(8+)

1249.6(9+)

1237.6(5+)

1146.3(7+)

1370(9+)

1469.3(10+)

1498.7(11+)

1482.2(14+)
V. Results – spectroscopic problems

Lacking branching ratio:

- 5 transitions at 347, 407, 959, 1072 and 1122 keV for decay of level 1122 keV(2+) (from NNDC)
- Branching ratio for 347, 407 and 959 keV gamma emissions only
- From our data, $I_{\gamma}=300(30)$ for 1072 keV gamma emission
- Dominant branching ratio is not mentionned!
- Other studies support dominance of 1072 keV deexcitation: [Demidov (2008), McGowan (Nuc. Phys. A562-1993)]
- Moreover, some assigned branching ratio seem erroneous

Unidentified γ-rays and levels:

- Non-registered γ-ray observed at 1075.5 keV
- Deduced cross section is compatible with inelastic scattering origin
- Observed by Demidov (Phys. Atomic Nuclei, 2008) who assigned it to a 4- state at 1237.6 keV
- Many gamma peaks are still unidentified
V. Results – Ground state rotational band

- At higher spin → weaker cross section; maximum for higher neutron energy
- Default Talys prediction maximum cross section position is at higher energy
- Strong internal conversion for low energy transitions decrease cross section (IC = 332 for transition $2^+ \rightarrow 0^+$ de 49 keV)
V. Results – Some other transitions

- Diverse shapes depending on excited levels
- Good agreement between our measurements with the ones from Dave et al. (1985)
- Varying agreement with default TALYS prediction depending on level concerned
V. Results – Summary

After 800 beam hours:

- 81 (n,n'γ), 11 (n,2nγ), 7 (n,3nγ) cross sections obtained for neutron energy from 0.2 MeV to 20 MeV
- Maximum cross section from 10 mb to 400 mb
- Good agreement with Dave(1985) for neutrons from 0.7 to 2.2 MeV
- Dozens of cross sections never registered in EXFOR before

On uncertainty quantification:

- 3 % for stronger transitions (ground band 4⁺ → 2⁺, 6⁺ → 4⁺… ) mainly from HPGe efficacity, fully correlated between neutron energies
- Up to 20 % for weaker transitions, mainly from statistic uncertainty, uncorrelated between neutron energies
- Covariances obtained between all measure points
VI. Additional works – Theory

Collaboration with theoreticians from CEA/DAM/DIF have been engaged for other isotopes ($^{238}\text{U}$, $^W$ ...), is beginning on $^{232}\text{Th}$

Issues:

- Is there a need of better nuclear structure knowledge for theory?
- Can theory reproduce our measurements?
- To what extent reaction mechanisms are constrained by our partial measurements?

Example of $^{238}\text{U}$:

- In $^{238}\text{U}$, some features have been reproduced by calculation
- Drop of XS at high spin reproduced using QRPA calculated spin distribution of residual nucleus instead of ad hoc prescription exciton (Dupuis et al., ND2016 - EPJ Web of Conf.146, 12002 (2017))
VI. Additional works – Sensitivity studies

• We stressed inelastic scattering importance in reactor applications

• How exactly do it matters?

• Sensitivity studies allow to quantify effect of variation of XS on reactor parameters

• To compare importance relative to other reactions, all reactions on both $^{232}$Th and $^{233}$U are studied

On sensitivity:

• Sensitivity of A to B is the quotient of relative variation of a variable A stemming from the relative variation of parameter B

• Its unity is « % / % »

• It is similar to a first order derivative

• Linear approximation appliable only for small variations

• Could be used to propagate moderate uncertainties
VI. Additional works – Sensitivity studies

Objectives:

- **Compute reactor's parameters sensitivity** to nuclear cross section of Th-232 and U-233 (neutron multiplication coefficient $k_{\text{eff}}$, effective delayed neutron fraction $\beta_{\text{eff}}$, radial power distribution...)
- **Validation** of these results by calculation of variance using:
  - Sensitivities and covariance matrices
  - Total Monte Carlo (TMC) method
- **Apply** this method to different reactor core design (Pressurized Water Reactor, Sodium-cooled Fast Reactor, Molten Salt Fast Reactor)

Tools used:

- **Transport codes**:
  - MCNP6 and SERPENT2
  - Both Monte Carlo neutron transport code
  - Sensitivity calculation tools recently added (some years ago)

- **Nuclear data library**:
  - 300 randomized TENDL-2013 files used for TMC calculations
  - ENDFB and JEFF library files used for reference sensitivity calculations

- **Computational power**:
  - Computation resources of CC-IN2P3
VI. Additional works – Sensitivity studies

Sensitivities of $k_{eff}$ to cross sections in PWR:

- Capture on $^{233}$U and $^{232}$Th, and fission on $^{233}$U at low energy ($E_n<1$ eV) are most sensitive reactions.
- $k_{eff}$ sensitivity to inelastic scattering is less than 100 time lower.
- Work in progress for other parameters and reactors.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Reaction</th>
<th>$\text{SENS SERPENT2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th-232</td>
<td>capture</td>
<td>$-0.2951$ %/%</td>
</tr>
<tr>
<td></td>
<td>fission</td>
<td>$0.0056$ %/%</td>
</tr>
<tr>
<td></td>
<td>elastic</td>
<td>$0.0083$ %/%</td>
</tr>
<tr>
<td></td>
<td>inelastic</td>
<td>$-5.57e-4$ %/%</td>
</tr>
<tr>
<td></td>
<td>n2n</td>
<td>$0.0014$ %/%</td>
</tr>
<tr>
<td>U-233</td>
<td>capture</td>
<td>$-0.0754$ %/%</td>
</tr>
<tr>
<td></td>
<td>fission</td>
<td>$0.3668$ %/%</td>
</tr>
<tr>
<td></td>
<td>elastic</td>
<td>$-2.27e-4$ %/%</td>
</tr>
<tr>
<td></td>
<td>inelastic</td>
<td>$-3.45e-5$ %/%</td>
</tr>
<tr>
<td></td>
<td>n2n</td>
<td>$1.09e-5$ %/%</td>
</tr>
</tbody>
</table>

Energy integrated sensitivities
VI. Additional works – Future measurements

- Deexcitation by internal conversion not observed with GRAPhEME (ground state band $2^+ \rightarrow 0$ gamma emission is very weak due to $^{332}$ IC factor)

- DELCO project aim to conceive a new instrument to allow internal conversion electrons detection

- Work in progress in the team

- $^{233}$U experimental data collected, to be analyzed

- $^{239}$Pu target in development to be employed within GRAPhEME

- Dedicated nuclear structure studies of actinides by combining GRAPhEME and GAINS setup is considered
Conclusion

Realized:
- Numerous cross sections obtained, soon to be in EXFOR
- Covariances of cross sections have been derived
- Sensitivity studies are in progress

To come:
- Preparation of new measurements (electron, $^{239}$Pu)
- Collaboration with theoreticians on collected cross sections

Thanks for your attention