Neutron-induced capture-to-fission cross section ratio measured at LANSCE

CNR-24

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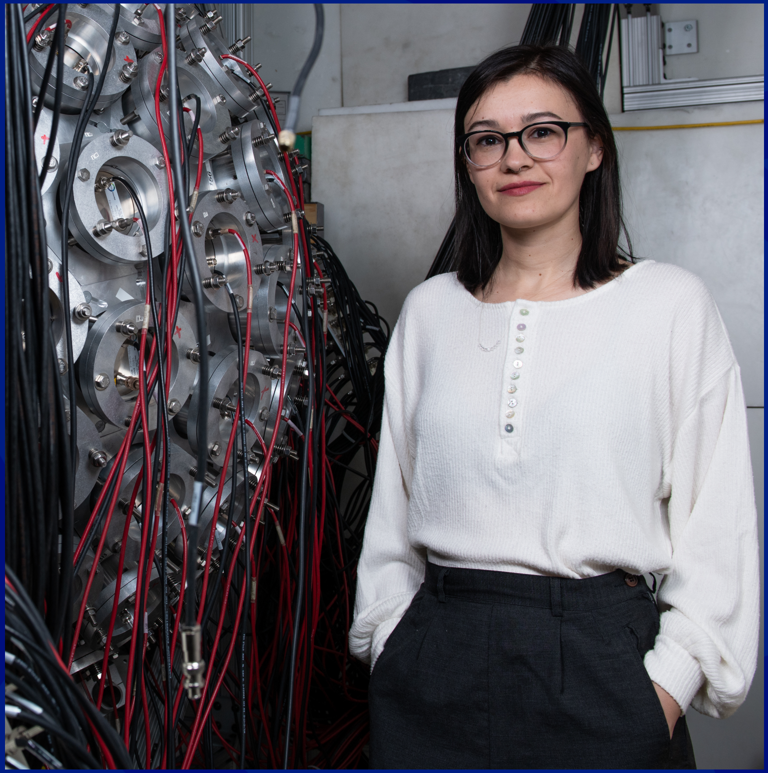
Presented by Ingrid Knapova

July 8-12, 2024



LA-UR-24-26643

Motivation

- There is a need of reduced uncertainties for applications in nuclear technology, including the design of advanced reactors.
- Also, uncertainties in the capture cross section impact our understanding of the criticality of U/Pu systems and transmutation rates.
- Isotopes involved in the Th-U and U-Pu fuel cycles of interest for these studies.
  - ^{235}U, ^{233}U and ^{239}Pu measured at LANSCE.
- The capture-to-fission ratio measurement eliminates the systematic uncertainties derived from:
  - Neutron flux, self-shielding and sample mass.

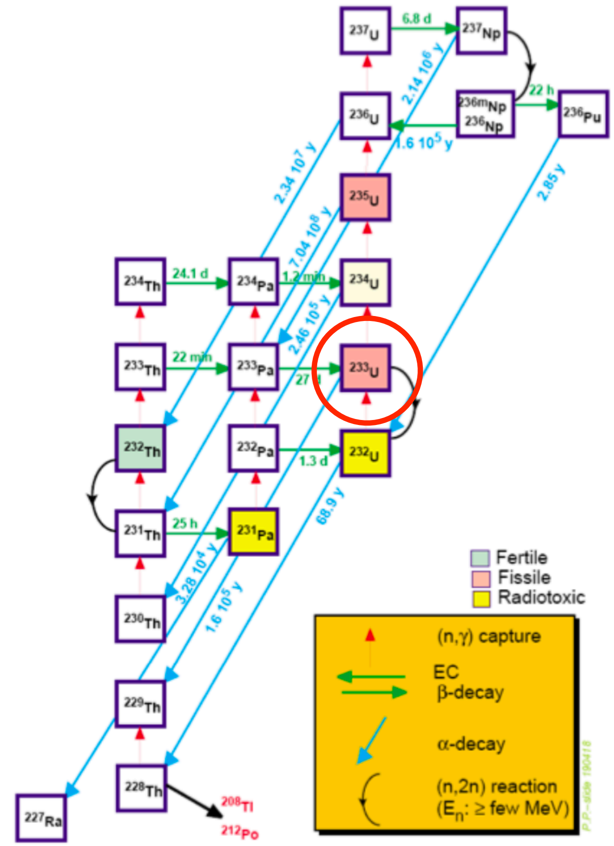


Figure 1 Illustration of the thorium fuel cycle where ^{233}U plays a role.

Motivation

- Experimental cross section data in the literature are sometimes scarce and measured decades ago with the detecting technologies available at the time (detectors, neutron flux and electronics).
- New data are constantly required to update the nuclear data evaluations.
- For some isotopes, the fission rate is considerable compared to capture.
- Good discrimination between γ-rays coming from capture and fission is required.
- New measurements proposed at LANL combining a fission detector and DANCE.

The ^{233}U measurement was the first ND measurement funded at LANSCE by NCSP!

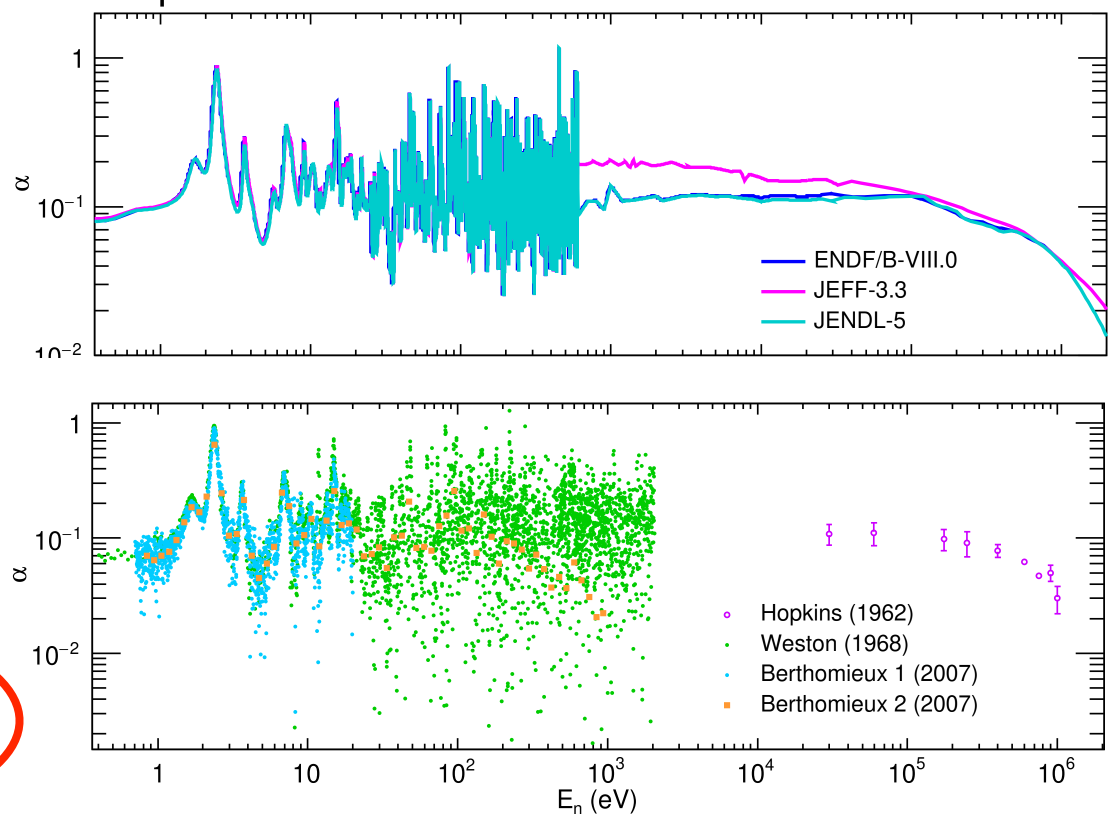


Figure 2 ^{233}U capture-to-fission ratio from the literature

LANSCE facility

- Neutrons produced by proton spallation on a W target.
- DANCE:
  - Mark-III spallation target.
  - Flight path 14 (20 m).
  - White neutron spectrum (En = 10 meV - 500 keV).

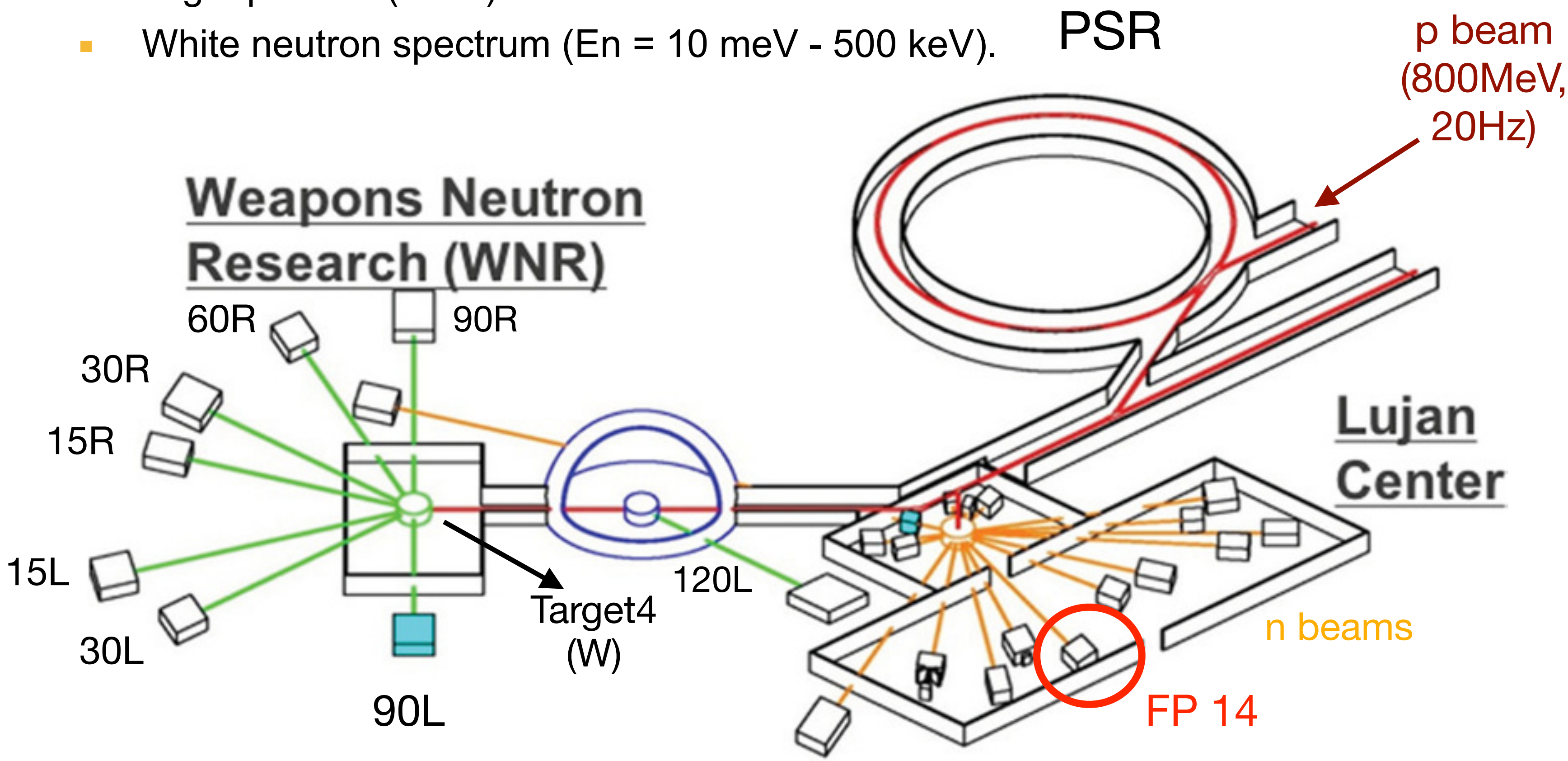
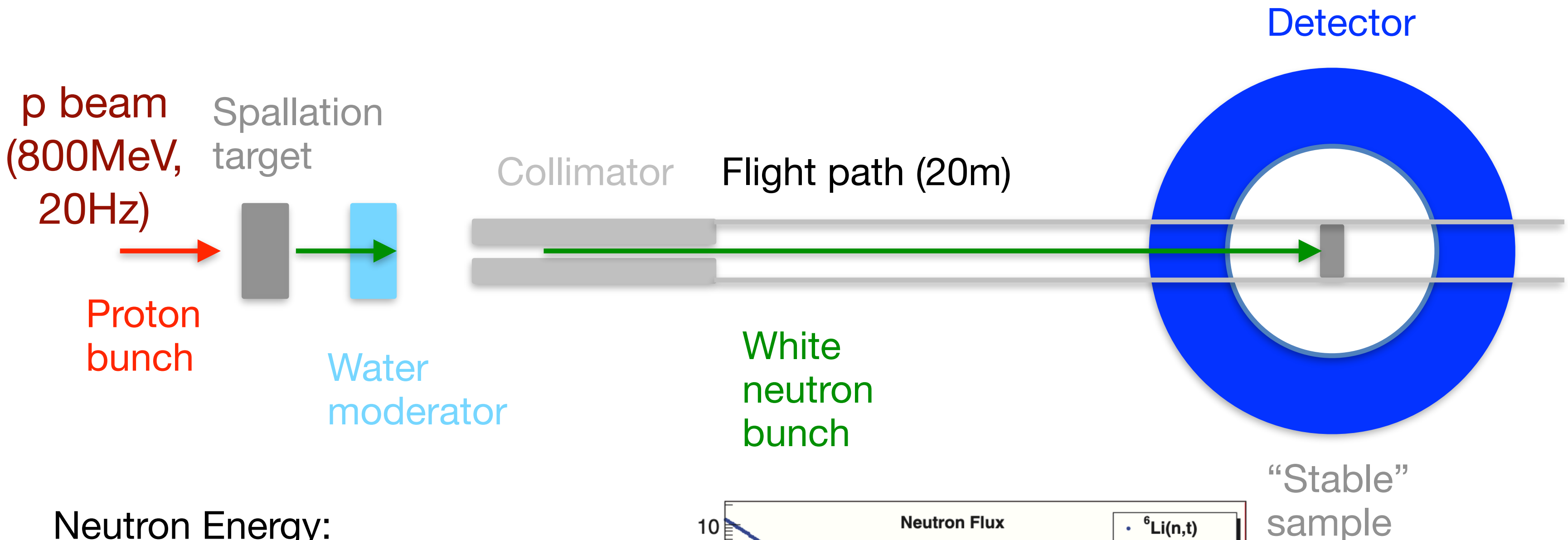


Figure 3 LANSCE facility illustration

Time-of-flight measurements

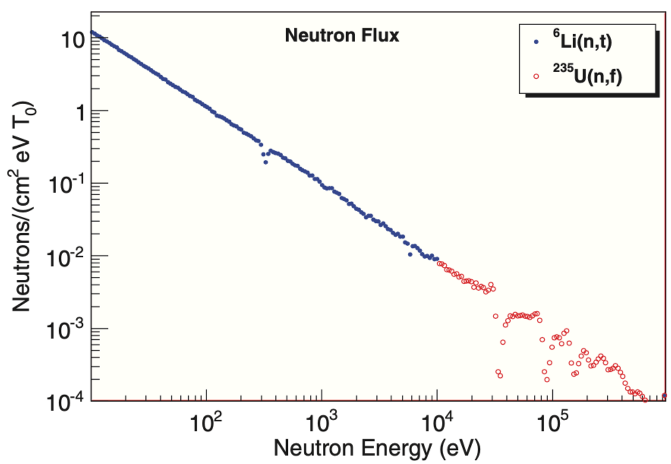


Neutron Energy:

$$E_n = m_n c^2 \frac{1}{\sqrt{1-(\frac{v}{c})^2}} - 1$$

with:

$$v = L/T$$



$Flux_n = 3 * 10^5$ n/s/cm^2/dec

Detectors

DANCE (**D**etector for **A**dvanced **N**eutron **C**apture **E**xperiments)

- $4\pi BaF_2$ γ-ray calorimeter composed by 160 crystals with an inner cavity of 17 cm radius [1].
- Used to measure neutron capture cross section data on small quantities of radioactive isotopes. Single γ-ray detection efficiency of 85%.
- We can measure En, Esum, Ecl, and Mcl, providing more information than with C6D6 detectors.
- A LiH ball is placed inside around the sample to absorb scattered neutrons.

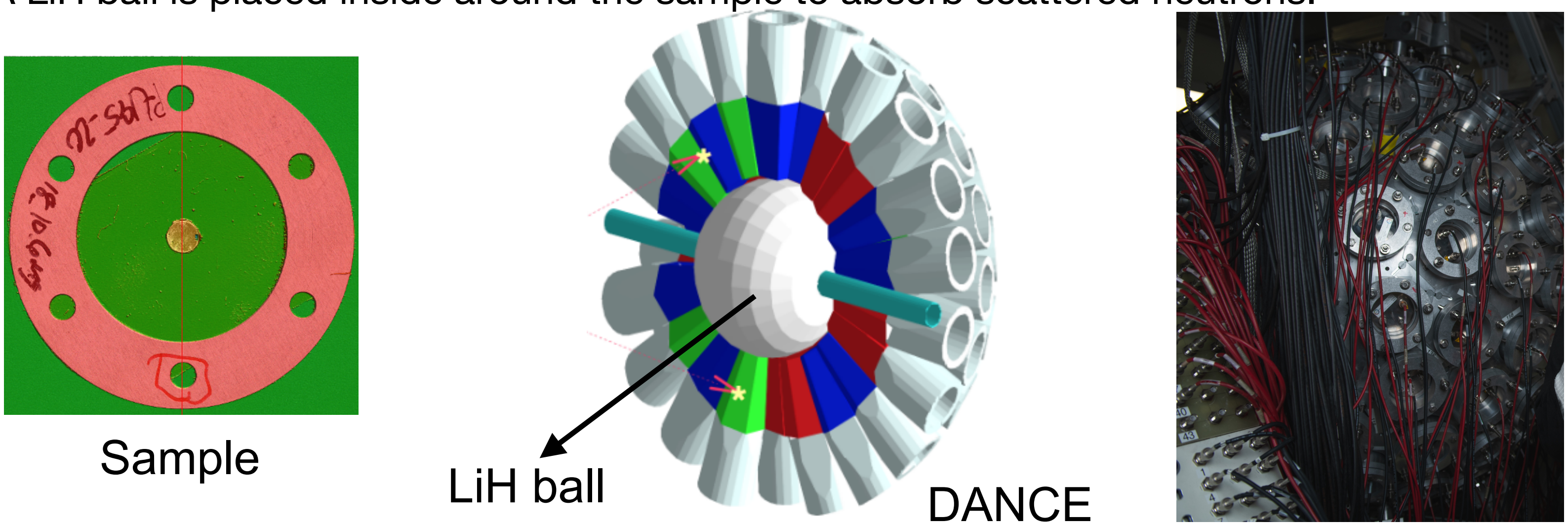


Figure 4 ^{233}U DANCE sample (left), DANCE + LiH ball (center) and DANCE picture (right)

[1] M. Heil et al., Nucl. Instrum. Methods Phys. Res. A **459**, 229 (2001).

Detectors

NEUANCE (**NEU**tron detector array at d**ANCE**)

- Neutron detector array that consists in 21 stilbene crystals arranged in a cylindrical geometry around the beam pipe [2].
- Used to detect neutrons coming from fission and determine by coincidence with DANCE, the fission γ-rays.
- NEUANCE detects neutrons with energies above 500 keV (fission neutrons have these energies), therefore **low energy scattered neutrons** that are below this threshold are **discriminated**.
- Single fission neutron efficiency of 12.5%.
- Possibility to use a thick target -> higher statistics/lower measuring time required.
- NEUANCE can also detect γ-rays.

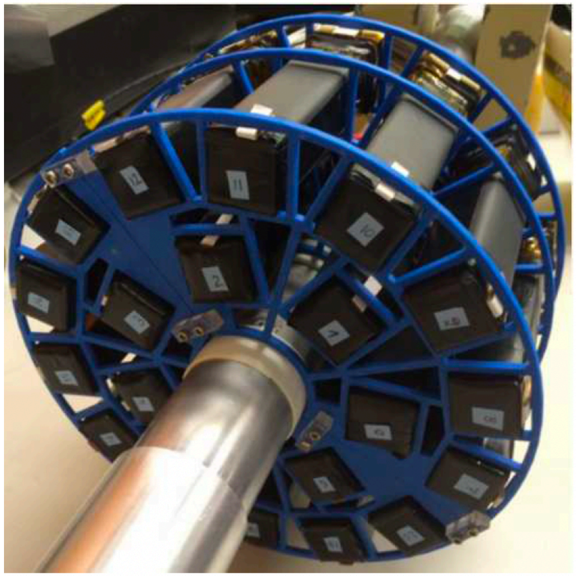


Figure 5 NEUANCE instrument

[2] M. Jandel et al. Nuclear Inst. and Methods in Physics Research, A **882** (2018) 105-113.

Detectors

PPAC (**P**arallel **P**late **A**valanche **C**ounter)

- 4π detection range [3].
- Used to detect Fission Fragments (FF) and determine by coincidence with DANCE, the fission γ-rays.
- Need to use a thin sample to achieve a high fission fragment detection efficiency -> lower statistics/larger measuring time required.
- Charged particle detection efficiency of ~65%.

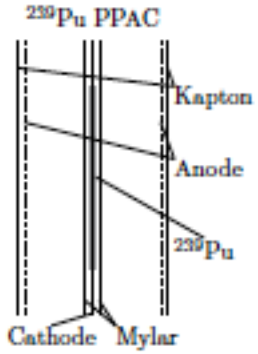


Figure 6 PPAC geometry

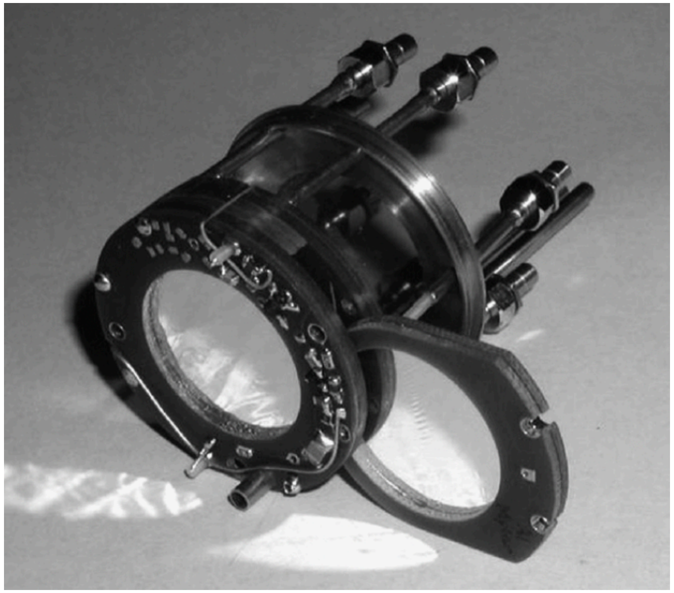


Figure 7 PPAC for DANCE

[3] M. Jandel et al. PRL **109**, 202506 (2012).

DANCE calibrations

- Intrinsic radioactivity of BaF_2 used to calibrate the DANCE crystals.
- Using the Alpha-decay chain of the ^{226}Ra present in the BaF_2.

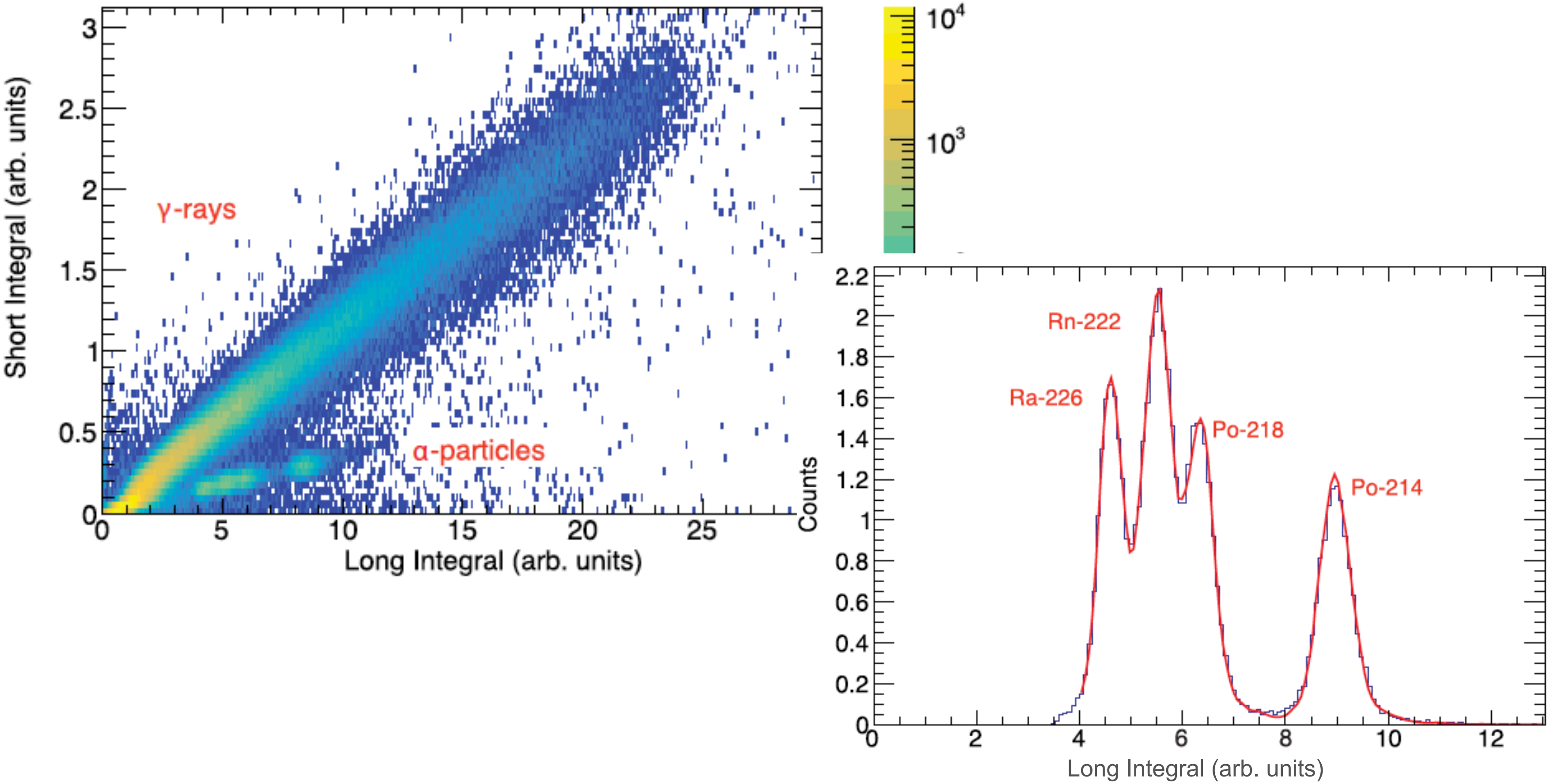


Figure 8 Pulse shape discrimination spectrum for DANCE crystals

Fission tagging process

- Search for coincidences between the two detectors.
- The DANCE γ-rays in coincidence with the fission particles (FFs/ neutrons) are tagged as fission gammas.
- The purpose of tagging is to define the shape of the fission γ-ray spectrum that can be subtracted from the total spectrum.

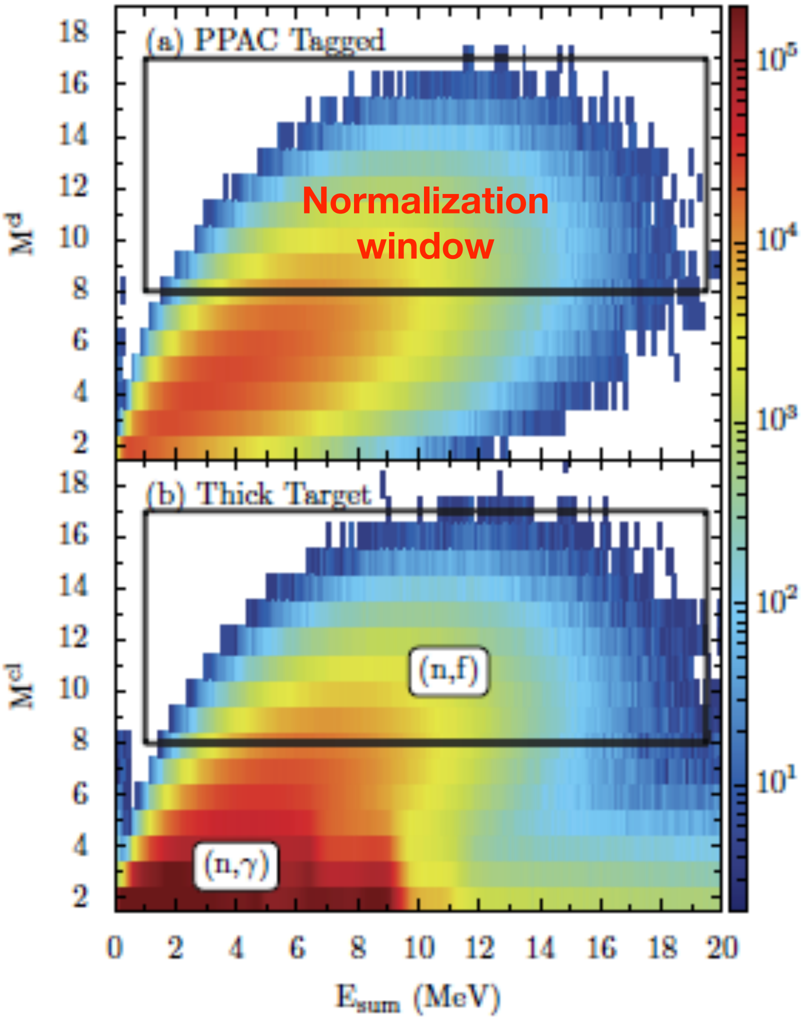
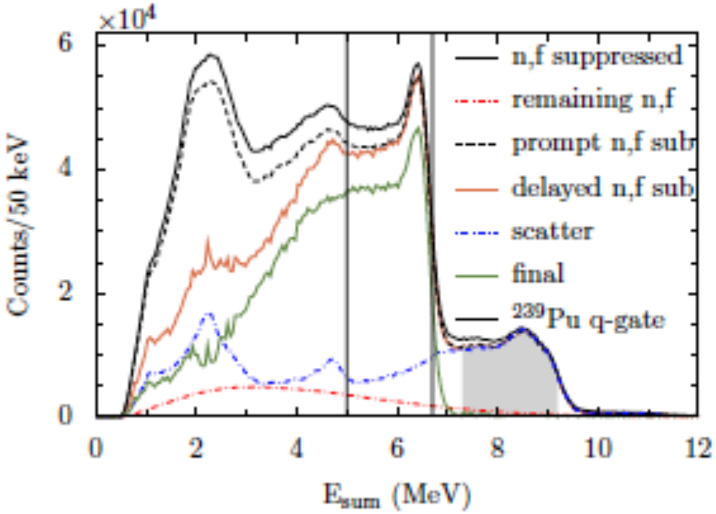


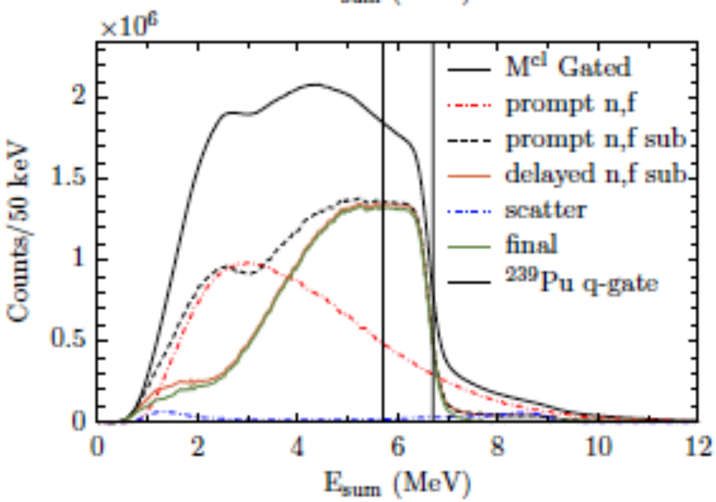
Figure 9 Fission tagging process

Background studies

- The background varies with the neutron energy, therefore it is subtracted per En bin.



Mcl = 4
Q = 6.53 MeV
Esum = (5.7-6.7) MeV



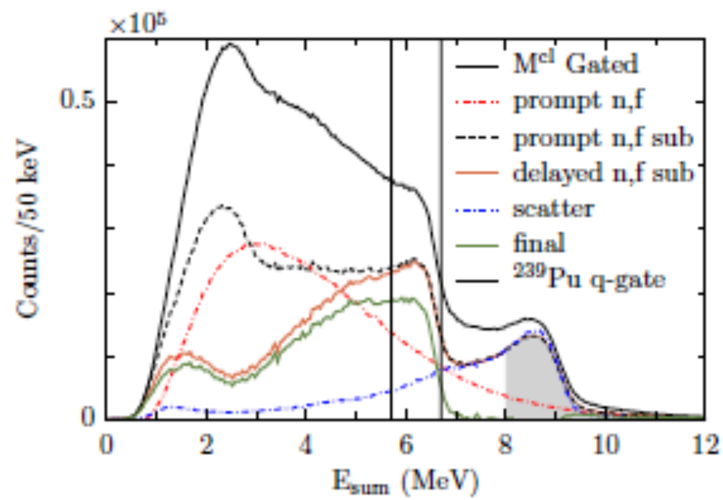


Figure 10 Background subtraction process for ^{239}Pu analysis

Capture-to-fission ratio

The capture-to-fission ratio is given by:

$$\alpha(E_n) \equiv \frac{\sigma_\gamma(E_n)}{\sigma_f(E_n)}$$

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Hence:

$$\alpha(E_n) = \frac{1}{k} \frac{C_\gamma(E_n)}{C_f(E_n)}$$

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Capture-to-fission ratio

- Experimental advantages of the **capture-to-fission ratio**:
  - It is much simpler and more reliable to determine experimentally as many of the systematic questions:
    - Sample mass
    - Self-shielding
    - Neutron exposure

  will cancel out in an appropriately designed experiment.
- The relative capture cross section can be obtained multiplying the ratio by the evaluated fission cross section.

Measurements at LANSCE

^{235}U capture-to-fission ratio

- Measured using the DANCE + PPAC setup from (4 eV to 1 MeV) .
- Three independent measurements for high precision in capture cross section:
  - Measurement with a thick sample -> to increase statistics at high neutron energies.
  - Measurement with a thin sample inside the PPAC -> for fission tagging.
  - Measurement of the neutron scattering background using a ^{208}Pb sample.
- Targets:
  - Thick target of 26 mg/cm^2 of 94% enriched ^{235}U.
  - Thin sample 130 ug/cm^2 of 99.9% enriched ^{235}U.
  - Scattering sample of ~120 mg/cm^2 of 99% enriched ^{208}Pb.
- Results published in [4].

[4] M. Jandel et al., Phys. Rev. Letters **109**, 202506 (2012).

^{235}U relative capture cross section

- The **alpha-derived capture cross section** was calculated by multiplying the capture-to-fission ratio by the ENDF/B-VII.0 fission cross section.
- The broadened cross section was used in the Resolved Resonance Region.

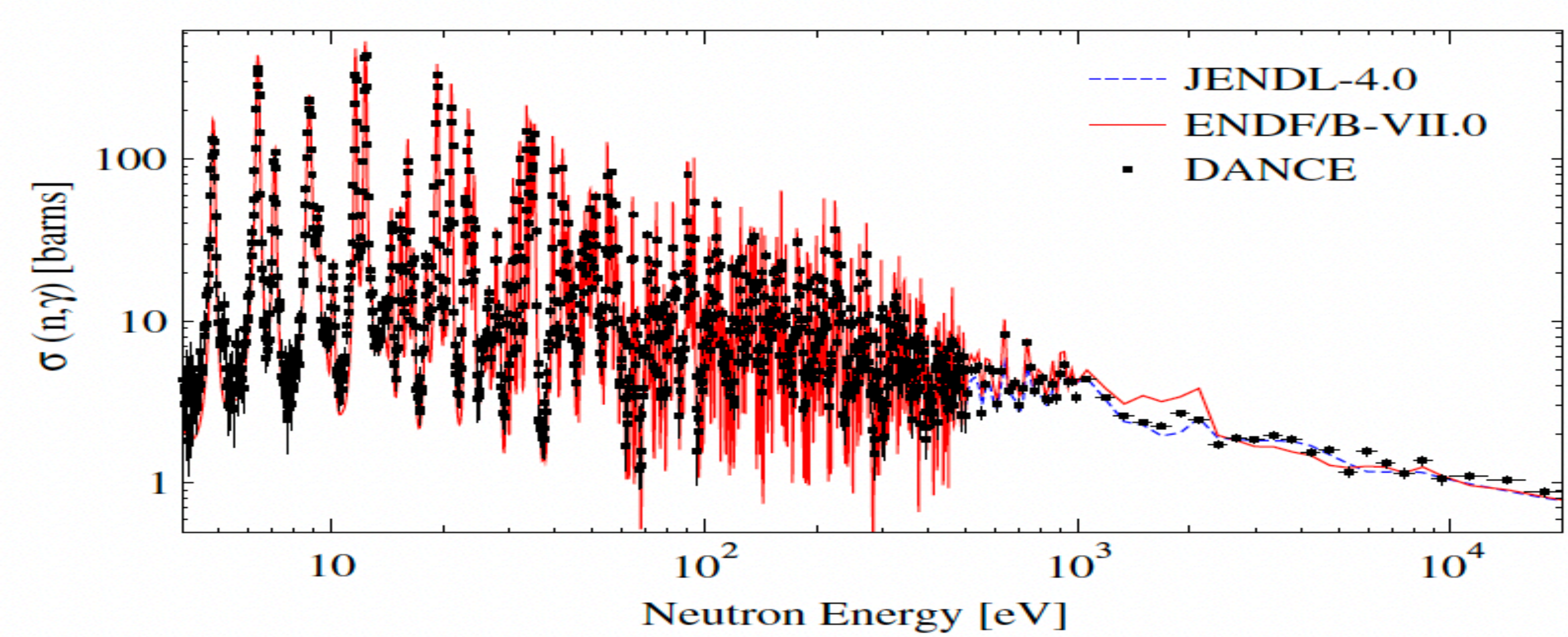

Figure 11 Results of the alpha-derived ^{235}U(n,γ) cross section

Measurements at LANSCE

^{239}Pu capture-to-fission ratio

- Measured using the DANCE + PPAC setup from (10 eV to 1.3 MeV) [5] - [7].
- Three independent measurements for high precision in capture cross section.
- Targets:
  - Thin 937 ug of 99.97% enriched ^{239}Pu.
  - Thick ~50 mg of ^{239}Pu.
  - Scattering 200 mg/cm^2 ^{208}Pb.

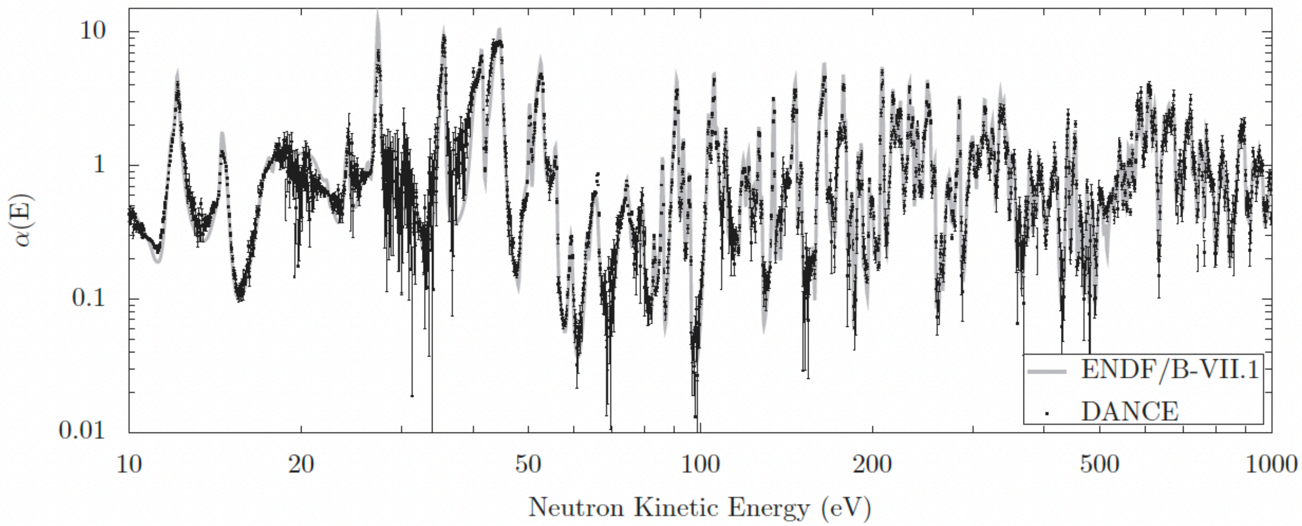

Figure 12 Results of the ^{239}Pu capture-to-fission ratio from 10 eV to 1 keV

[5] S. Mosby et al. Phys. Rev. C **89**, 304610 (2014).

[6] S. Mosby et al. Phys. Rev. C **97**, 041601 (2018).

[7] S. Mosby et al. Nucl. Data Sheets **148**, (2018) 312-321.

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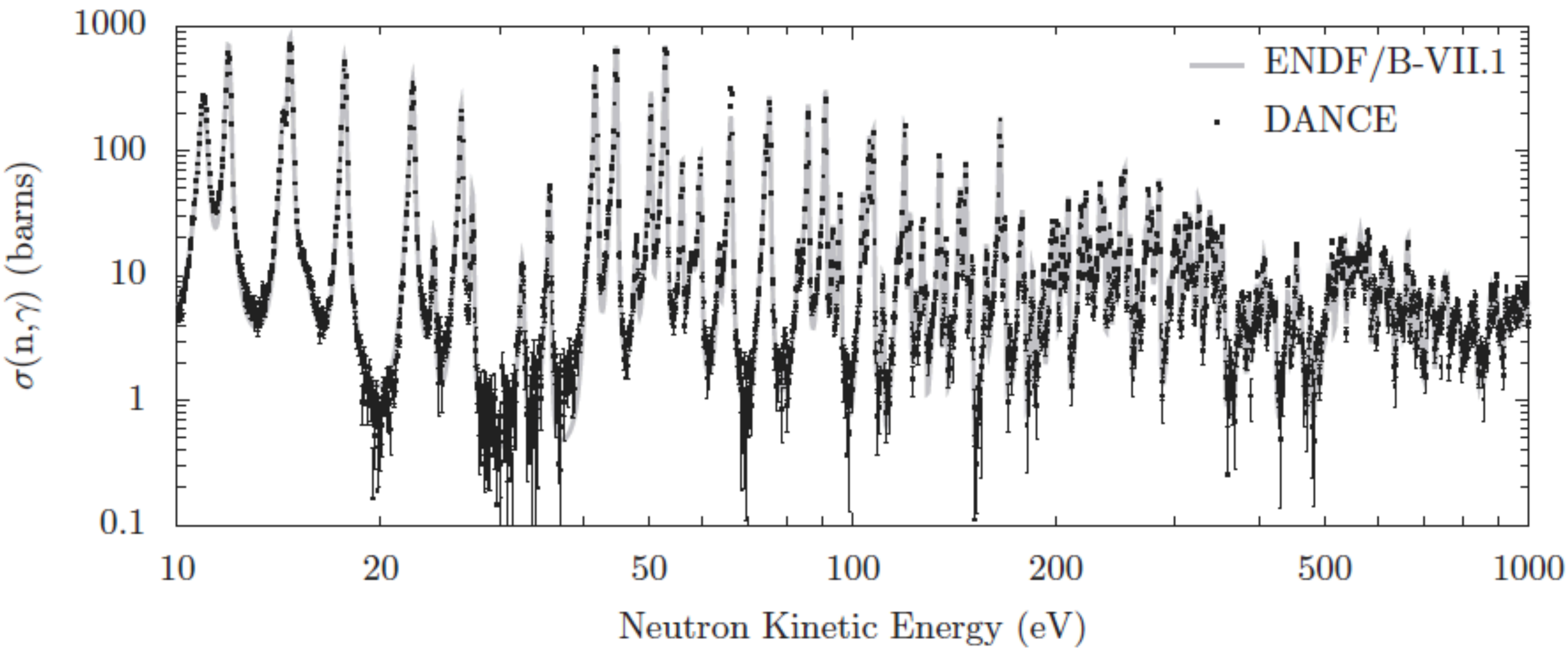

Figure 13 Results of the alpha-derived ^{239}Pu(n,γ) cross section from 10 eV to 1 keV

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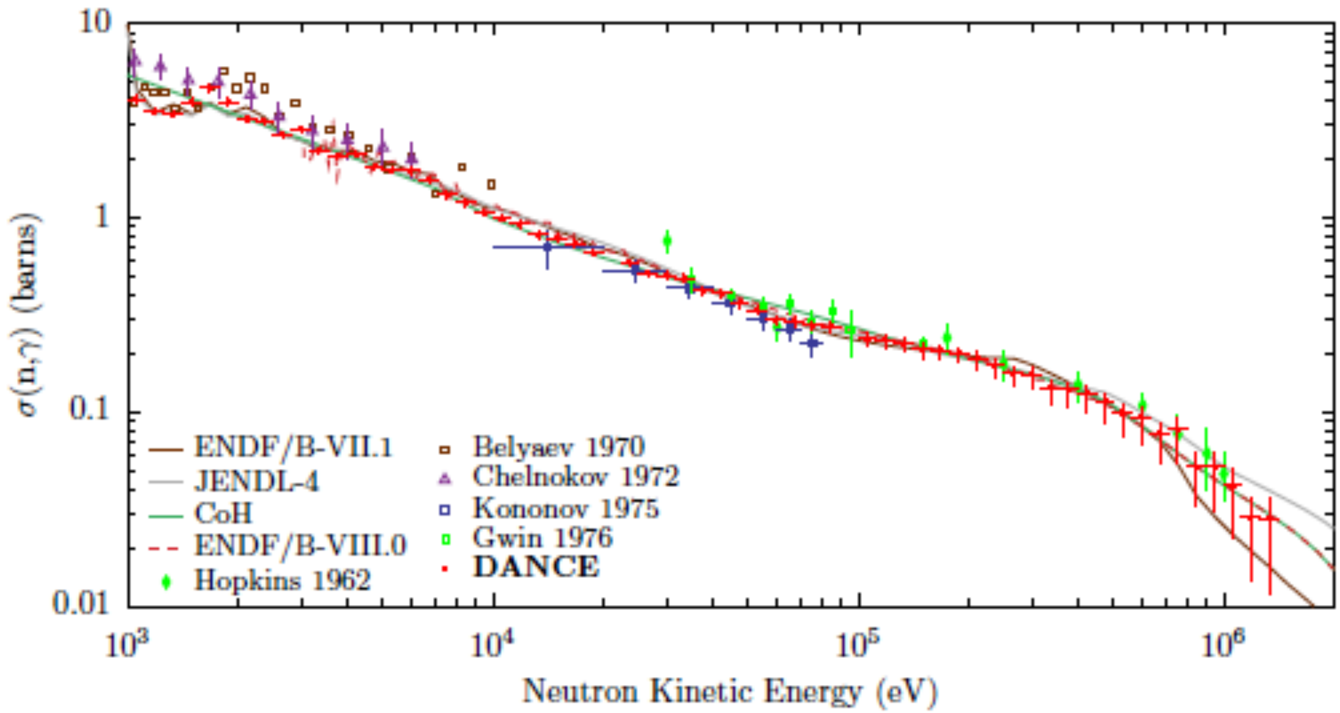

Figure 14 Results of the alpha-derived ^{239}Pu(n,γ) cross section from 1 keV to 1.3 MeV

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Measurements at LANSCE

^{233}U capture-to-fission ratio

- Measured using the DANCE + NEUANCE setup from (0.7 eV to 250 keV).
- Normalization to **ENDF/B-VIII.0** broadened cross section ratio in the neutron energy region (8.1-14.7) eV:
- Results published in [7].

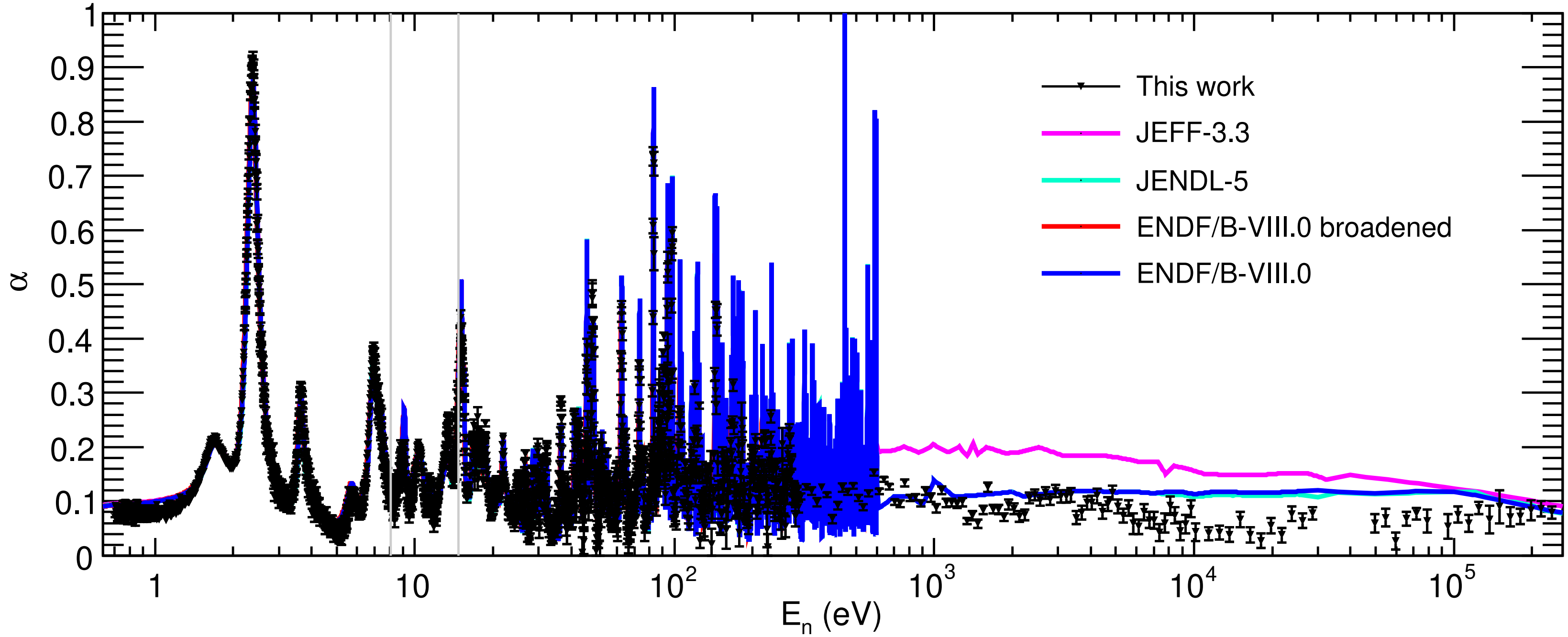
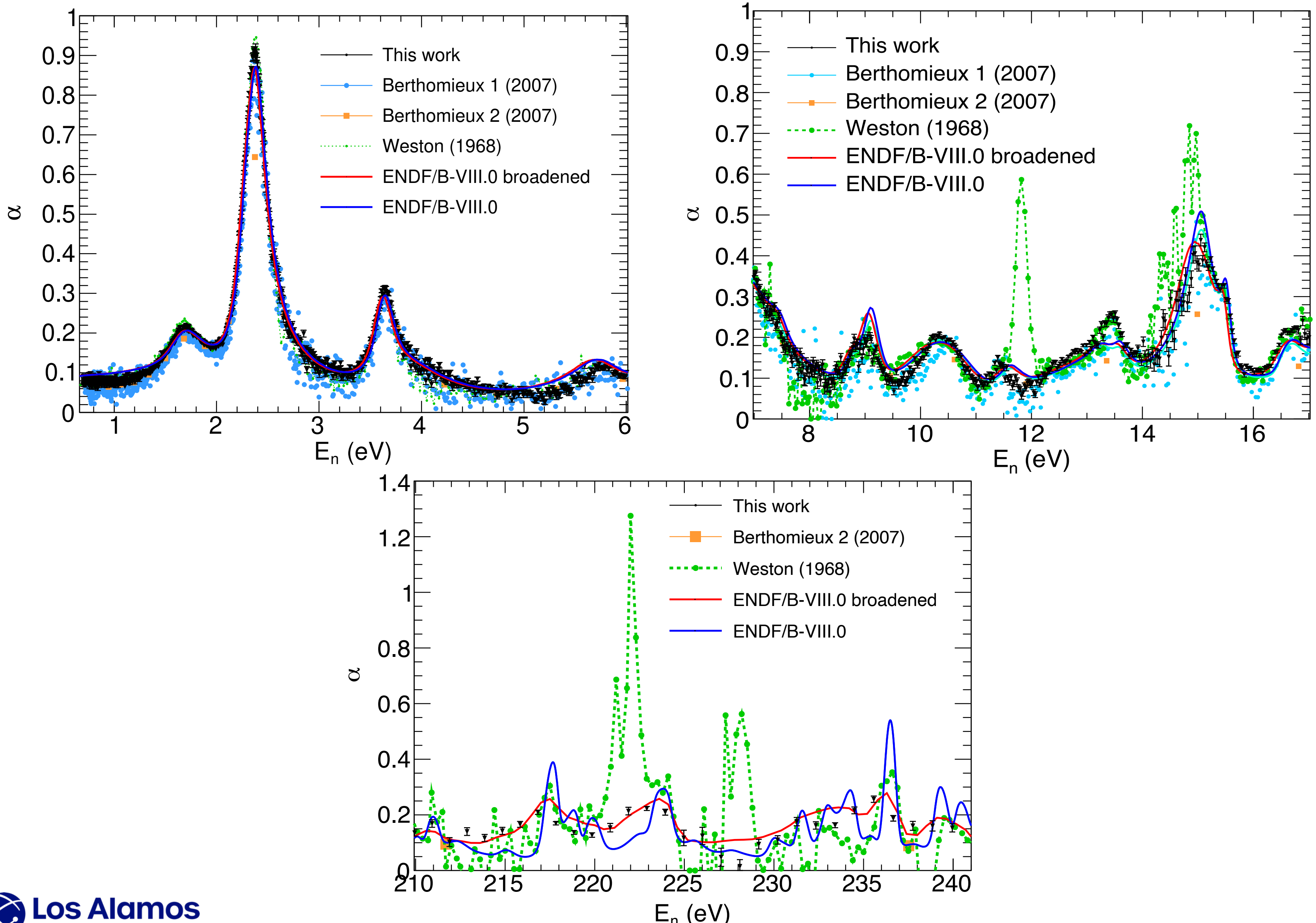


Figure 15 Results of the ^{233}U capture-to-fission ratio

[7] E. Leal-Cidoncha et al., Phys. Rev. C **108** 014608 (2023)



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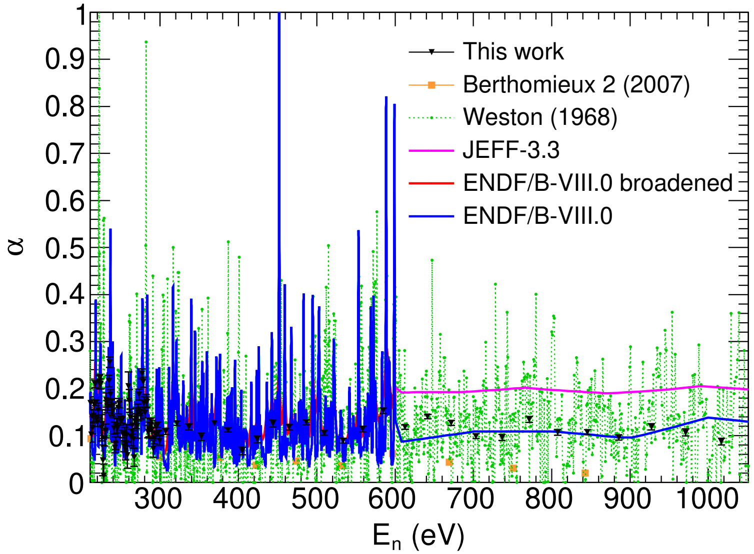
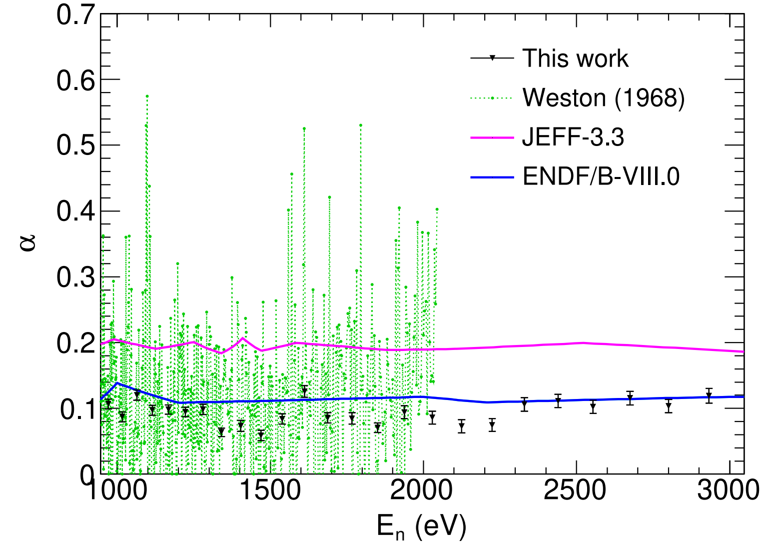


This work
Berthomieux 2 (2007)
Weston (1968)
JEFF-3.3
ENDF/B-VIII.0 broadened
ENDF/B-VIII.0
α
E_n (eV)



This work
Weston (1968)
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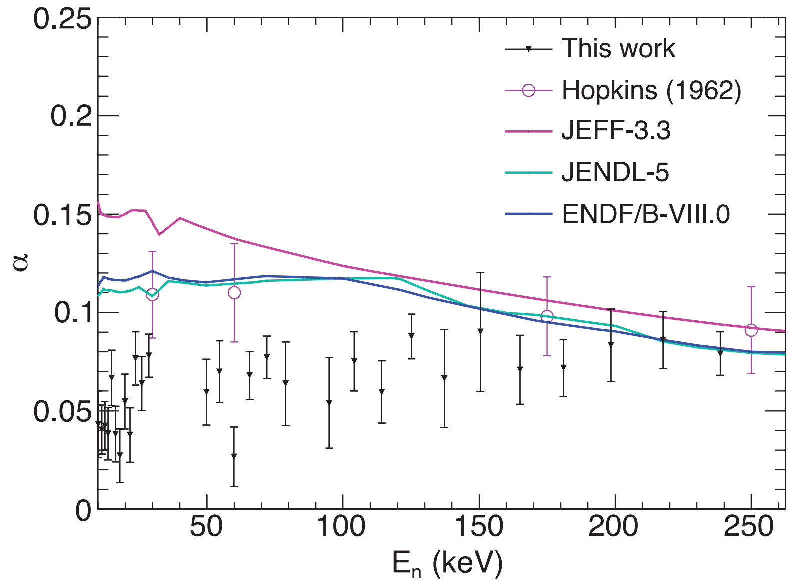
α
E_n (eV)



This work
Hopkins (1962)
JEFF-3.3
JENDL-5
ENDF/B-VIII.0
α
E_n (keV)

^{233}U relative capture cross section

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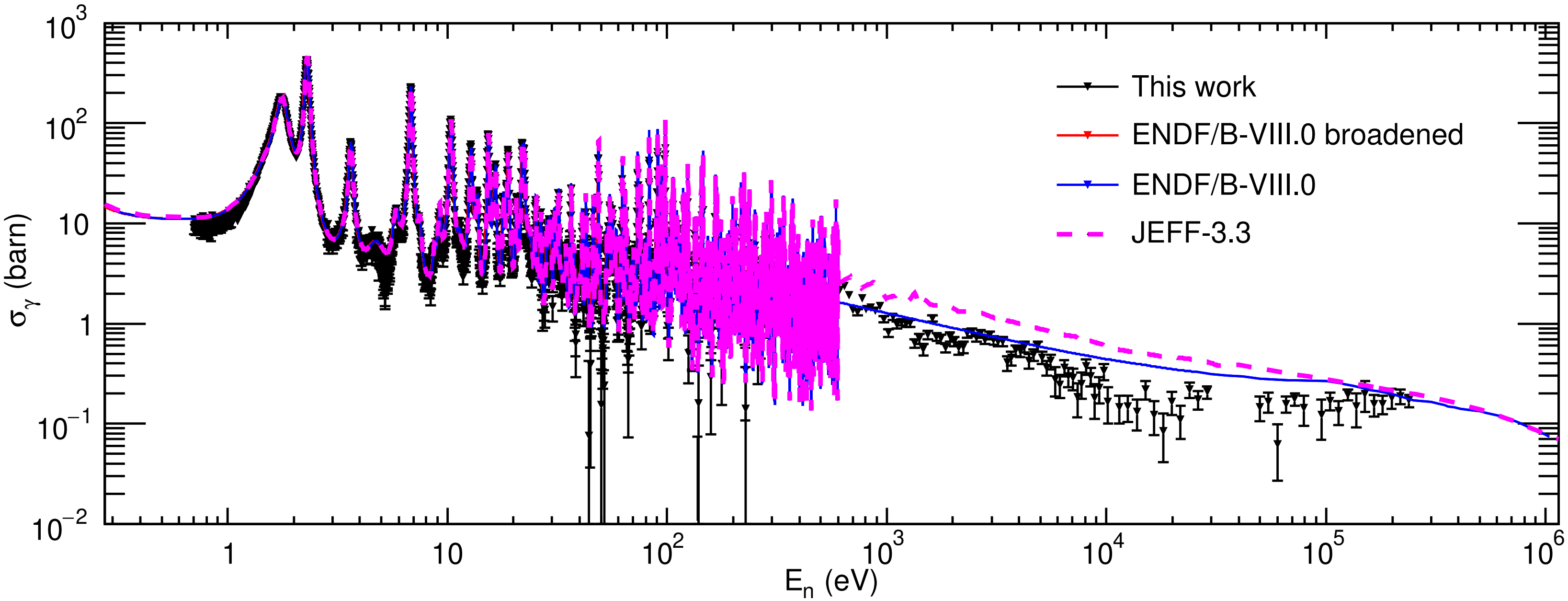

Figure 16 Results of the alpha-derived ^{233}U(n,γ) cross section

Statistical Model Calculations

- Statistical model calculations were performed by I. Stetcu, T. Kawano and A. Lovell with the CoH3 code [4] from 1 keV to 5 MeV (Energy for which only the first fission chance is involved).
- This code combines the coupled-channels optical model and the statistical Hauser-Feshbach model calculations by performing the Engelbrecht-Weidenmüller transformation of the penetration matrix.
- Different values of the **average γ-ray width** have been tried by adjusting the M1 γ-ray strength function for the scissors mode.
- Mughabghab gives 40 meV.
- To reproduce the data from Hopkins it had to be reduced to 24 meV.
- A smaller value would be needed to reproduce this work.

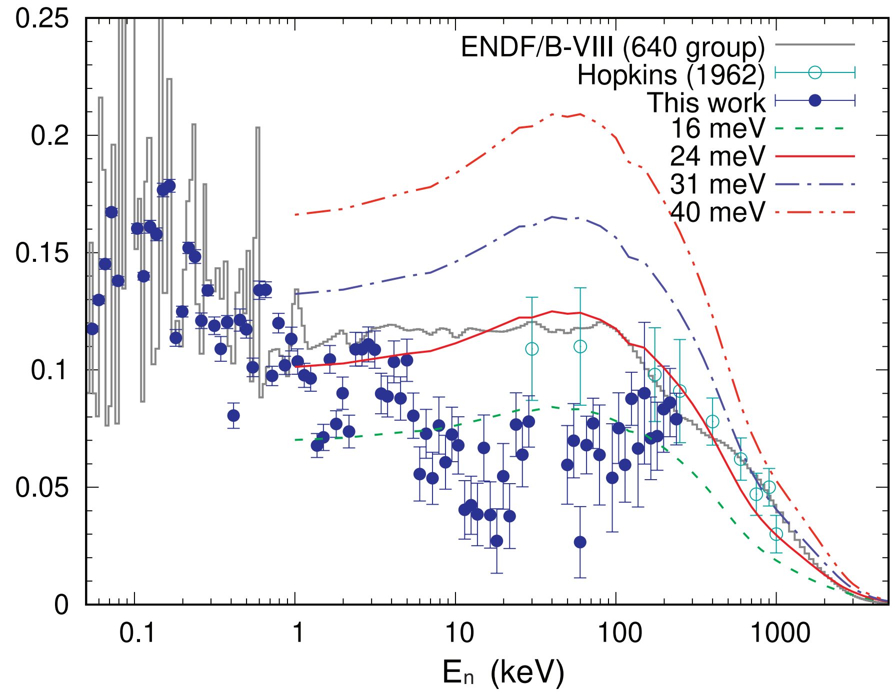

Figure 17 Statistical model calculations for ^{233}U

[4] T. Kawano, Springer Proceedings in Physics **254**, 27 (2021)

Conclusions

- The capture-to-fission cross section ratio eliminates the uncertainties associated to the neutron flux, sample mass and self-shielding.
- It can be measured at LANSCE combining DANCE and a fission detector.
- A PPAC and NEUANCE have been used in combination with DANCE to detect FF and fission neutrons.
- Measurements of the ^{235}U, ^{233}U and ^{239}Pu in the neutron energy region from eV to 100s keV - 1 MeV have been performed in the last years.
- The alpha-derived cross sections have been also calculated multiplying the ratio by the evaluated fission cross section.
- Other detectors could be used in combination with DANCE to tag fission events in the future.

Acknowledgements

Thanks to our collaborators:

Aaron Couture, Evelyn M. Bond, Todd A. Bredeweg, Cathleen Fry, Marian Jandel, Toshihiko Kawano, Amy E. Lovell, Shea Mosby, Gencho Rusev, Ionel Stetcu, and John Ullmann
Los Alamos National Laboratory

Luiz Leal, and Marco Pigni
Oak Ridge National Laboratory