New measurements of spontaneous fission properties of Pu isotopes

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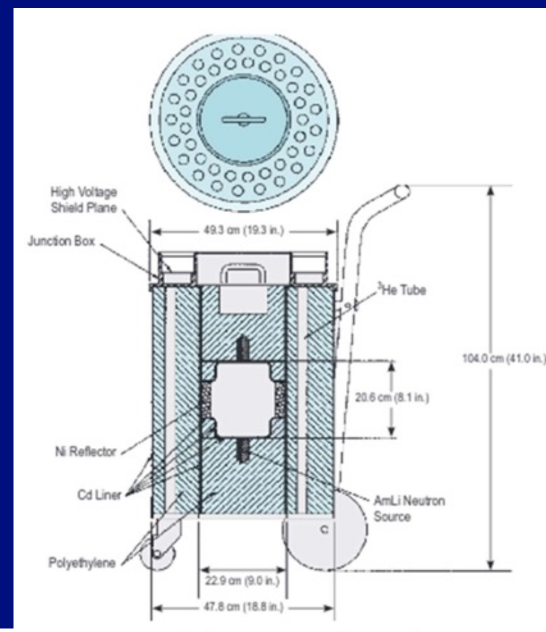
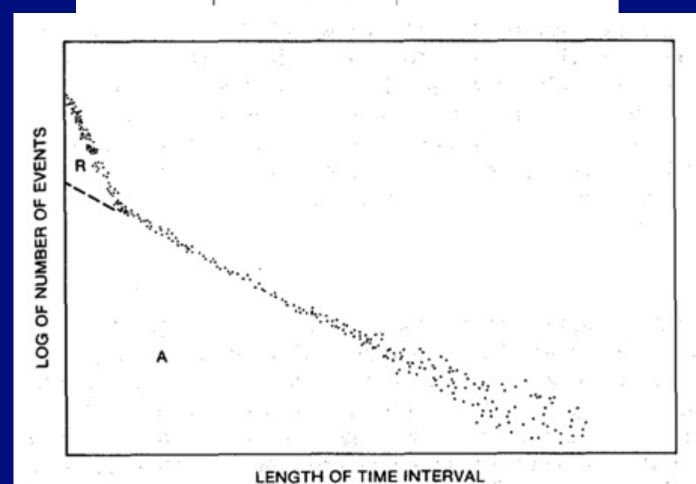
Improved nuclear data can improve NDA for the quantification of Pu content

- Typical application: neutron well counter with a sample inside, measuring the number and time distribution of the neutrons detected
- Spontaneous fission neutrons are a relatively clear signal for even-even Pu isotope quantity, absent any unlikely actinides (Cf, Cm and Am isotopes for example).
- Nuclear data needed to accurately calculate the expected neutron rate expected from a sample include
    - Isotopic sf rates (fission half-lives)
    - Neutron number per fission
    - Neutron number distribution
    - Prompt Fission Neutron Spectra (PFNS)





Pu spontaneous fission properties

isotope	half-life	specific activity (Ci/g)	fission branch per decay	$\bar{\nu}$	spontaneous fission rate (f/(mg·s))
^{238}Pu	87.7(1) y	17	$1.9(0.1) \times 10^{-11}$	2.19(7)	1.2
^{240}Pu	6561(7) y	0.23	$5.7(2) \times 10^{-8}$	2.154(5)	0.49
^{242}Pu	$3.73(2) \times 10^5$ y	0.0039	$5.53(5) \times 10^{-6}$	2.149(8)	0.79
% uncertainties					
^{238}Pu	0.11%		5.3%	3.2%	1.2
^{240}Pu	0.11%		3.5%	0.23%	0.49
^{242}Pu	0.54%		0.90%	0.37%	0.79

Table 1: *Spontaneous fission properties of even-even Pu isotopes. Values and uncertainties for half-lives and fission branches come from ENSDF [8], and values and uncertainties on $\bar{\nu}$ come from Ref. [3]. The lower half of the table shows uncertainies as percentages.*

The Team

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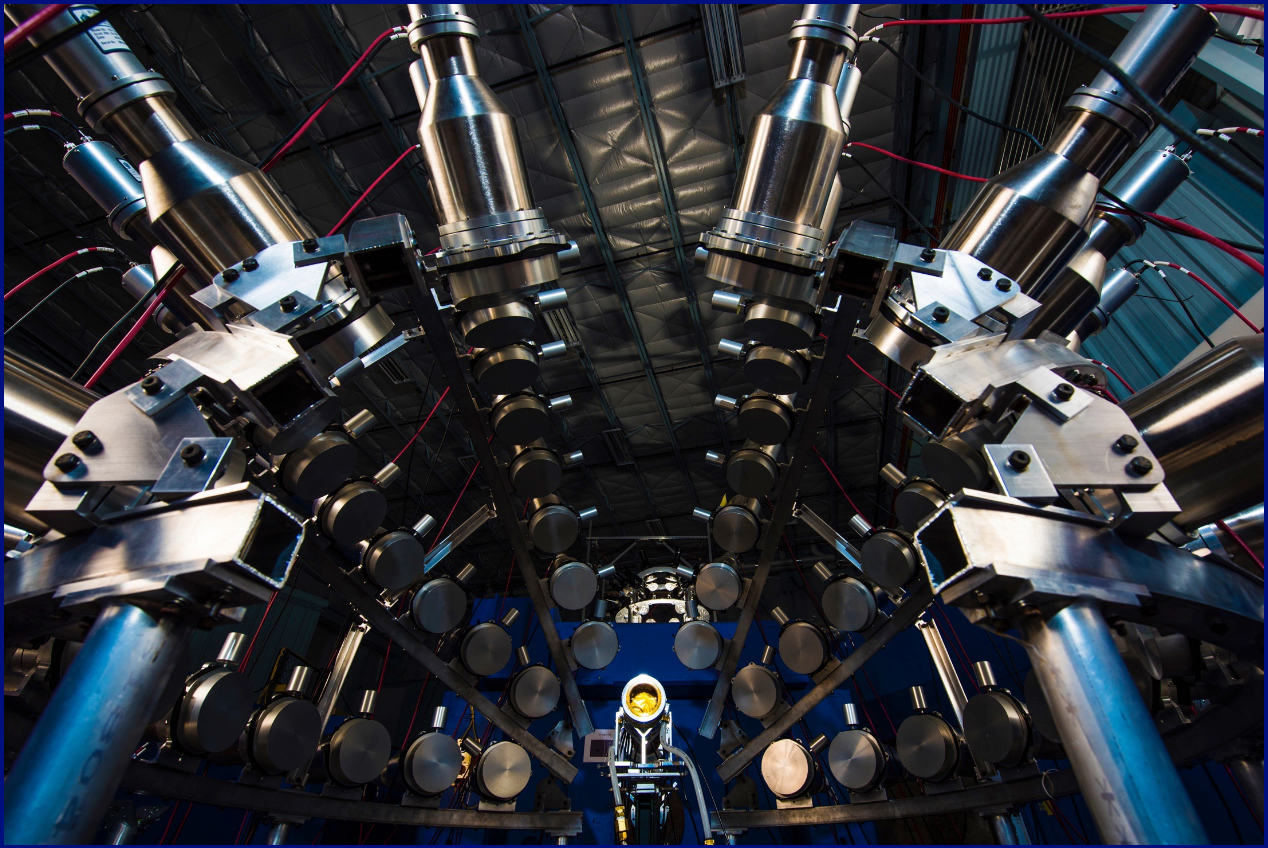
Chi-Nu

Neutron detection:
EJ309 Liquid Scintillator Array

- 1 m distance
- 54 detectors

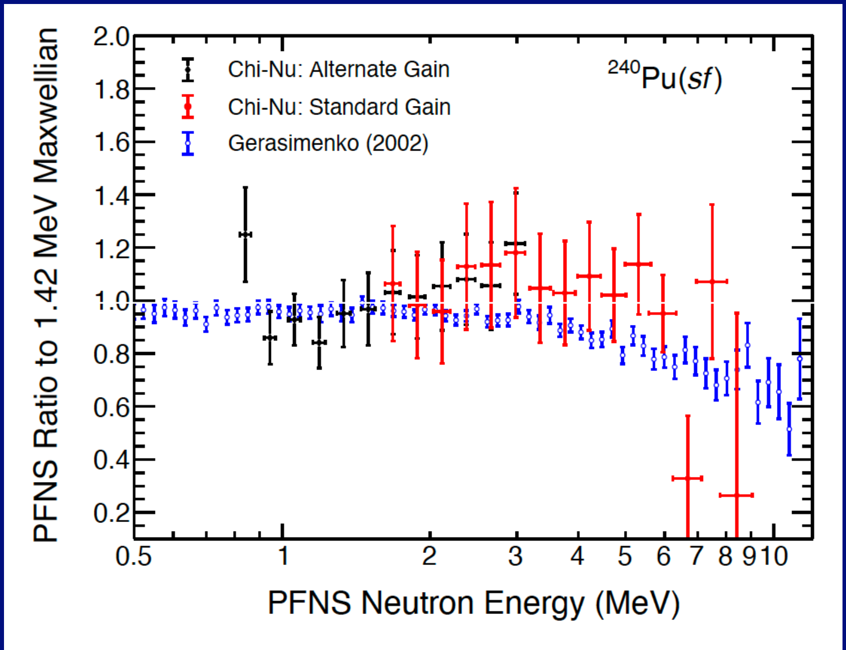
Fission detection:
12-cell Parallel Plate Avalanche Counter (PPAC) built at LLNL with a total of ~18mg of 99.875% ^{240}Pu

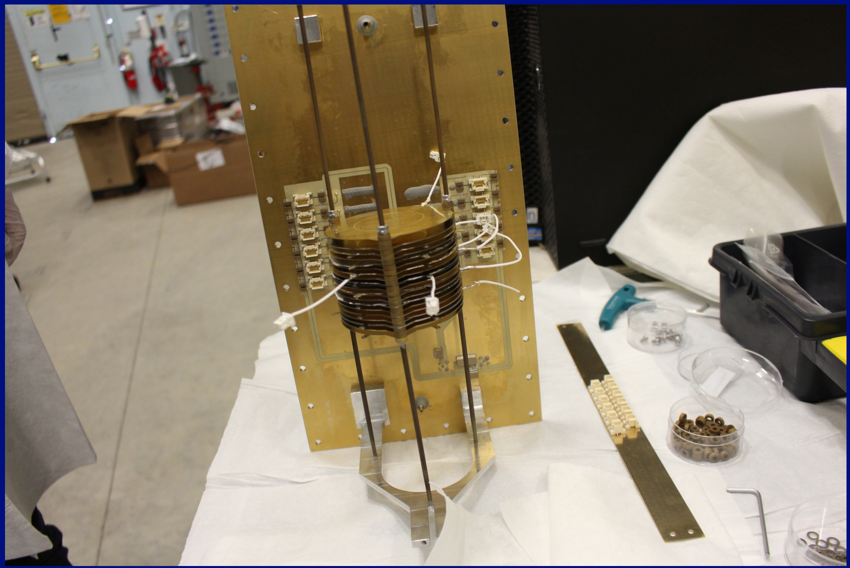


Chi-Nu Liquid Scintillator Detector Array

^{240}Pu(sf) Prompt Fission Neutron Spectrum

- Prior PFNS measurements of Pu(sf) are limited – only one modern PFNS measurement, with incomplete uncertainty quantification
- We measured as part of a NCSP funded effort to measure the PFNS of ^{240}Pu(n,f)
- sf data taken between “macropulses” at LANSCE/WNR
- Used a LLNL Parallel Plate Avalanche Counter
- Do we need more data? Yes, with a better fission trigger...



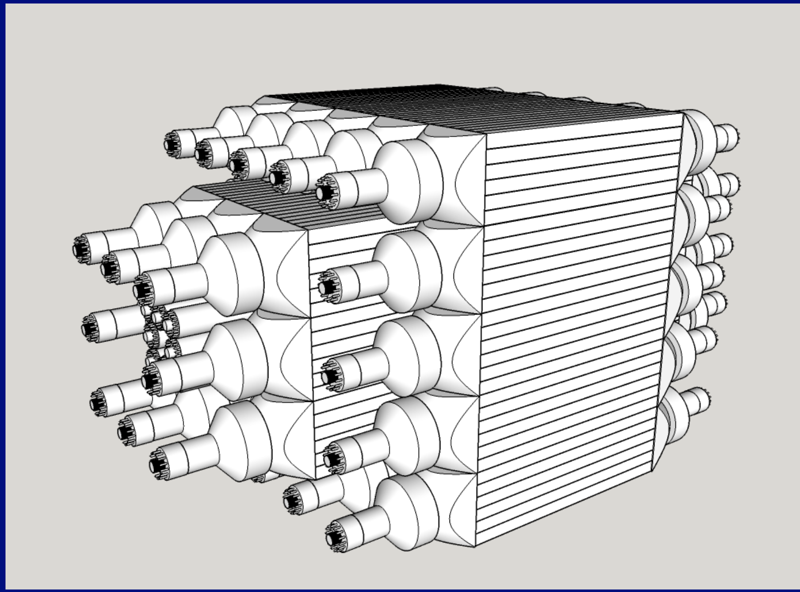


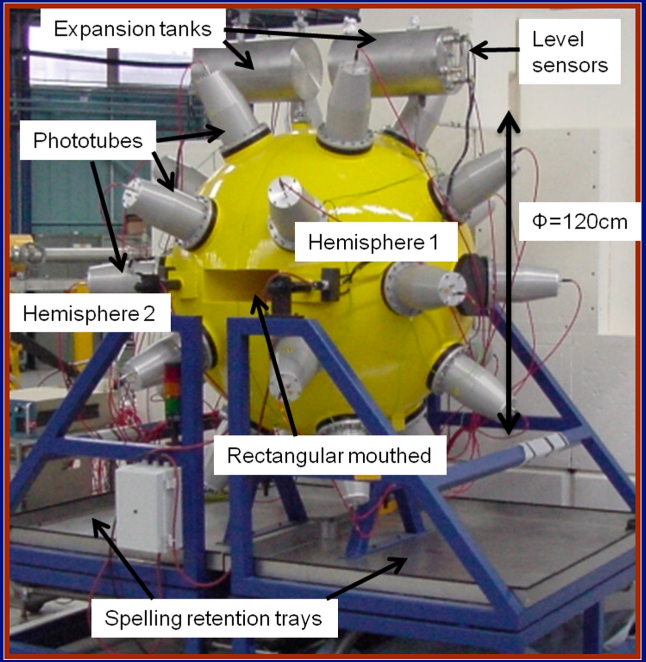
^{242}Pu(sf) PFNS

- Data taken using a legacy foil at LANSCE with the University of Michigan
- A fission chamber was built to house one of these foils (9 mg of ^{242}Pu)
- Data taken during covid over several months, when LANSCE beam was off (there was an extended beam off period)
- n-γ correlations from the data were analyzed and published (S. Marin, et al., *Event-by-event Neutron-Gamma Multiplicity Correlations in ^{242}Pu(sf)*, 2021 INMM 2021 Annual Meeting Proceedings
- These data have now also been shared with the University of New Mexico to extract the Prompt Fission Gamma-ray Spectrum (PFGS)
- The PFNS from these data will be obtained as part of this project

Various high-efficiency neutron counter designs to consider

- Plastic bars with inter-bar thin Gd layers, as in the CEA SCONE array (right)
- Gd-doped liquid scintillator type detectors, like Carmen (below)





Detailed MCNP-Polimi calculations will be used to develop a suitable high-efficiency, economical detector array design.

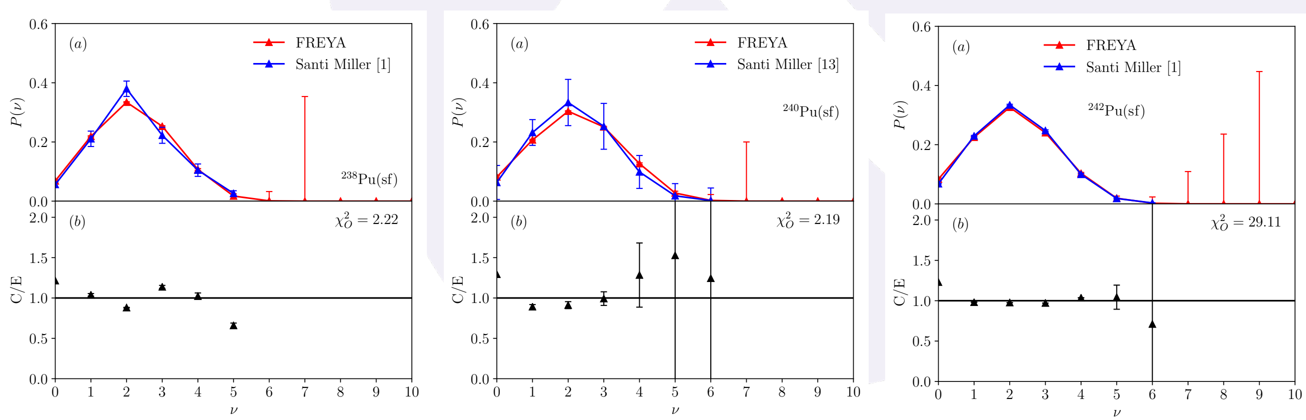
This detector array will then be used to measure the neutron number distribution for ^{240,242}Pu(sf).

Even-even Pu samples at LANSCE

- A total of 18 mg of ^{240}Pu on 12 foils, electrodeposited at LLNL for the (n,f) PFNS measurement, 4 cm in diameter, in a PPAC
- Legacy samples of Pu isotopes from fission earlier fission cross section measurements, each on ~10cm diameter deposit, mounted on 10 inch foil:
  - ^{240}Pu: 0.787 mg
  - ^{242}Pu: 9.918 mg (current ^{242}Pu(sf) data)
  - Other non-even-even and non-Pu samples, plus possibly more ^{240,242}Pu
- The ongoing LANSCE and CEA/DAM/DIF collaboration on (n,f) PFNS measurements plans a ^{240}Pu(n,f) measurement this FY; ~16mg on (22) 5 cm foils mounted in in a fission chamber, from Geel (Belgium). Can be used for sf measurements.
- All of these samples are high isotopic purity, typically greater than 95% with some greater than 99%

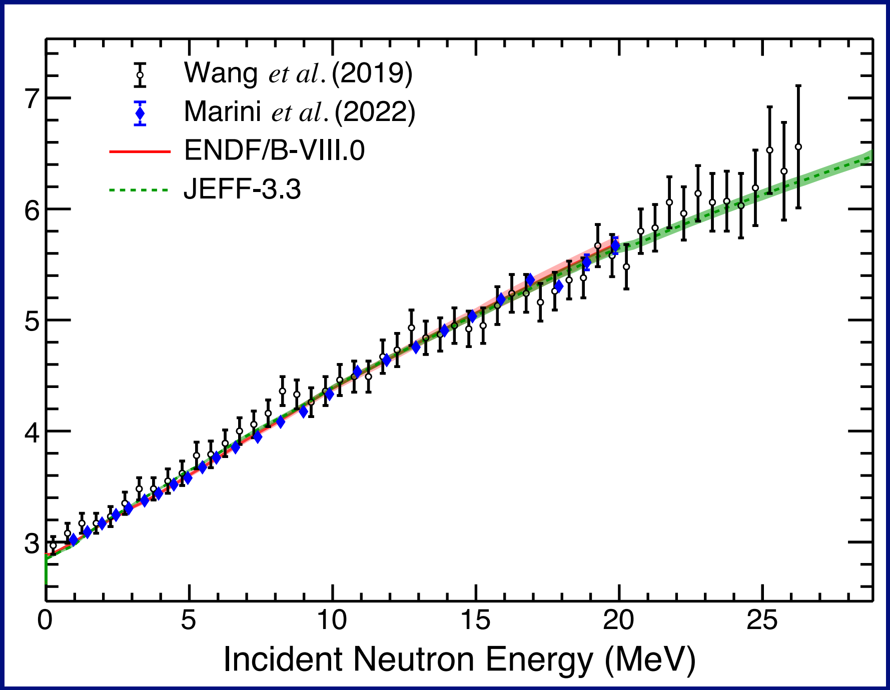
Theory and Evaluation of Pu(sf) Results (LLNL)

- Improvements in modeling of ^{238,240,242}Pu(sf) are needed, especially for the prompt fission neutron spectrum (PFNS), average neutron multiplicity, multiplicity distribution P(ν), branching ratios and half-lives.
- Modeling will be done with the complete event fission generator, FREYA, developed at LBNL and LLNL.
- The average neutron multiplicities of ^{238,240,242}Pu(sf) are small, ~2.14- 2.19, but higher moments of P(ν) are broad, resulting in FREYA fits with an atypically large parameter controlling these moments
- No other data are available for fitting these isotopes, fits could be done much better with additional data, particularly the correlated data taken by Chi-Nu at LANL and FS-3 at UM
- New data could significantly improve physics modeling and produce better evaluations



New neutron number distribution measurements

- The Chi-Nu array, or similar array, could be used to count neutrons as long as the fission tag is adequate.
- At the right is an example of a nubar ("Marini") obtained with Chi-Nu in-beam for $^{239}Pu(n,f)$
- However, higher moments of the distribution are difficult to obtain with high precision
- So, high neutron detection efficiency techniques need to be investigated



P Marini, et al., PLB 835, 137513 (2022)
BS Wang, et al., PRC 100, 064609 (2019)

^{238}Pu Measurements

- Goal is to extend all of these measurements to ^{238}Pu.
- Not included in the plan, yet, due to its much higher specific activity.
- But, LANSCE can handle adequate amounts to make sf measurements, and reasonably pure material is available.
- We would need a very clean fission signal, though, and making a thin ^{238}Pu foil for a fission chamber, say, will take some effort and may fail.
- Alternate fission signals can be investigated.

- We will look into ^{238}Pu measurements during this project, and proceed if they look viable.

Thank You!

and

Any Questions?