Los Alamos NATIONAL LABORATORY

New measurements of spontaneous fission properties of Pu isotopes

Matt Devlin
Keegan Kelly
Los Alamos National Laboratory

Sara Pozzi
Shaun Clarke
University of Michigan

Adam Hecht
University of New Mexico

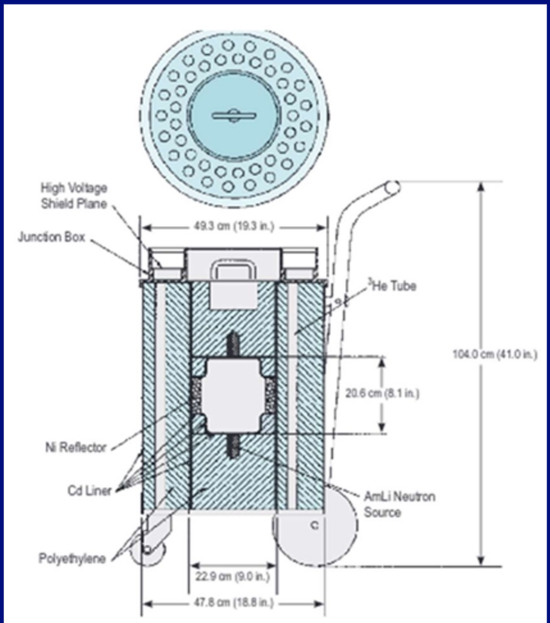
Ramona Vogt
Lawrence Livermore National Laboratory and University of California Davis

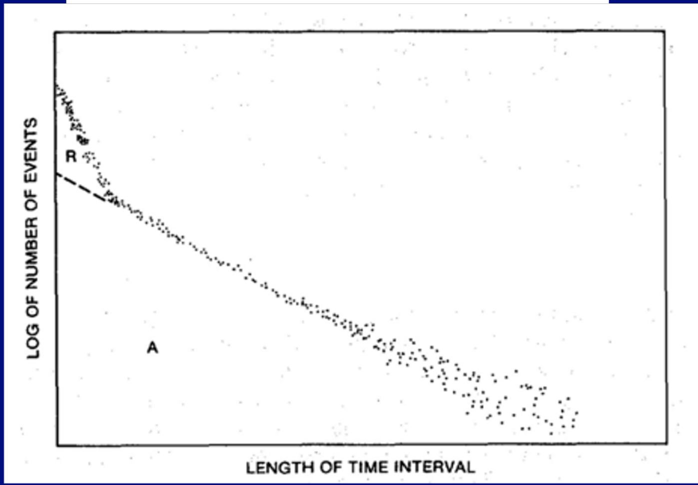
February 13, 2025

LA-UR-25-21262

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

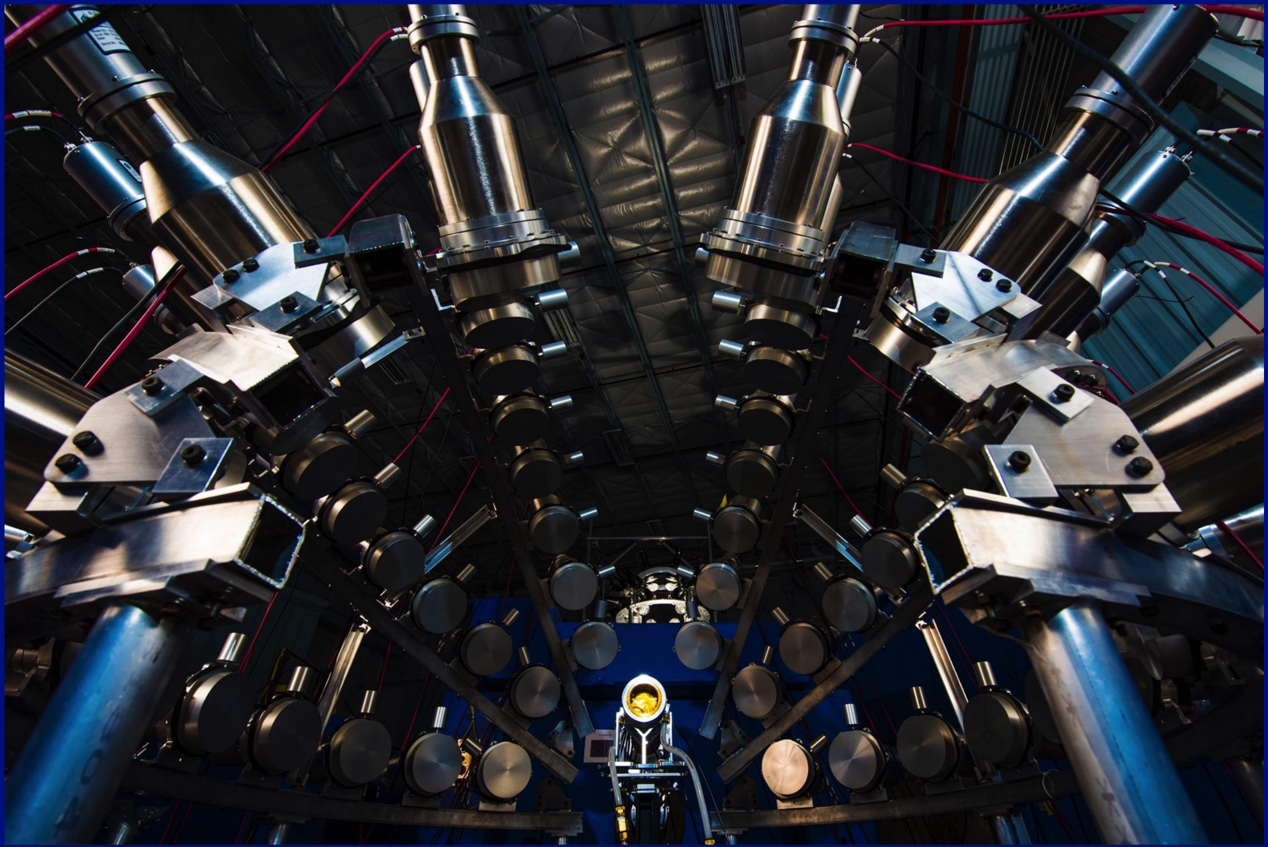
Table 1: *Spontaneous fission properties of even-even Pu isotopes. Values and uncertainties for half-lives and fission branches come from ENSDF [8], and values and uncertainties on $\bar{\nu}$ come from Ref. [3]. The lower half of the table shows uncertainies as percentages.*

^{242}Pu(sf) PFNS data from Chi-Nu analyzed by UNM

Chi-Nu:
54 EJ309 Liquid Scintillator Detector Array, 1 m from the samples

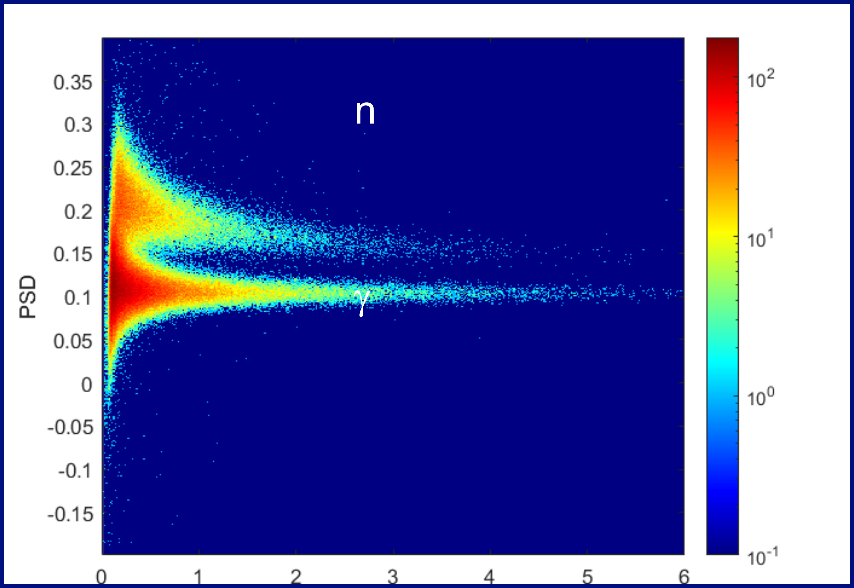
Fission detection: single-cell Fission Chamber, built with a legacy sample AT LANSCE, with ~9.9 mg of 99.92% ^{242}Pu

Measured during COVID, during a long LANSCE beam outage

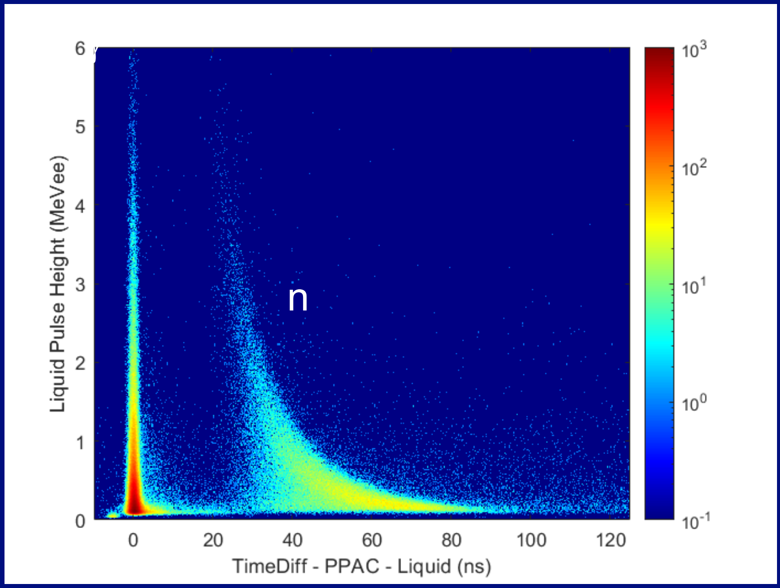


Chi-Nu Liquid Scintillator Detector Array

^{242}Pu(sf) PFNS progress



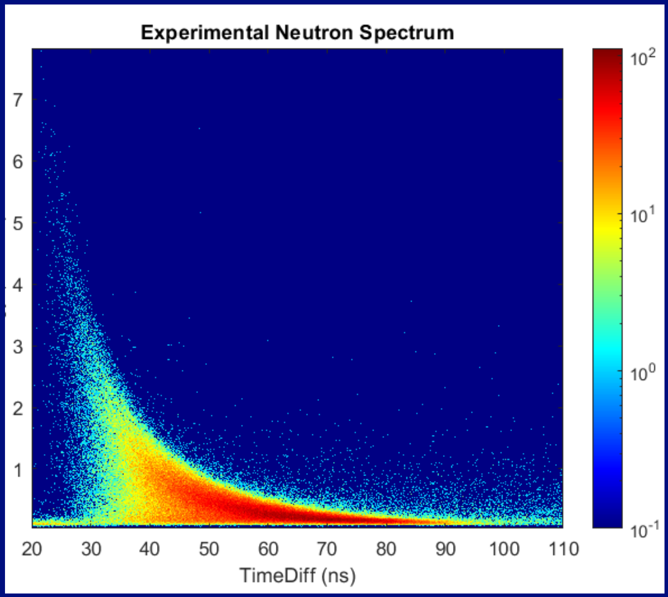
Sample pulse shape discrimination (PSD) plot showing neutron and γ-rays in the liquid scintillators

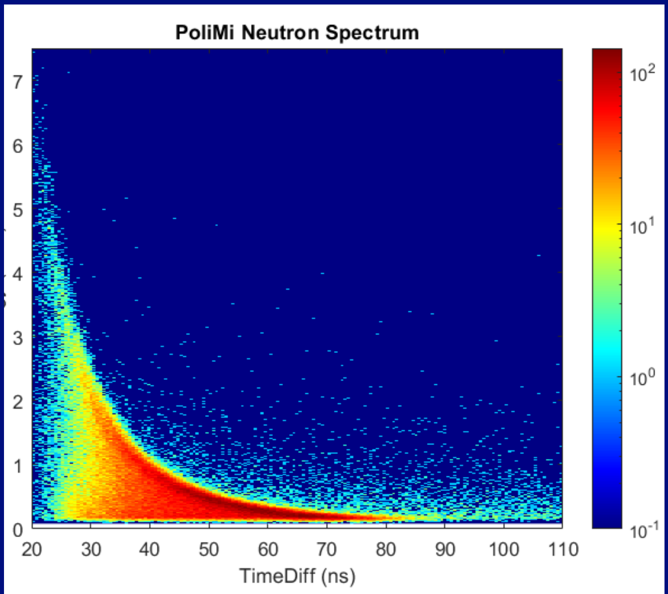


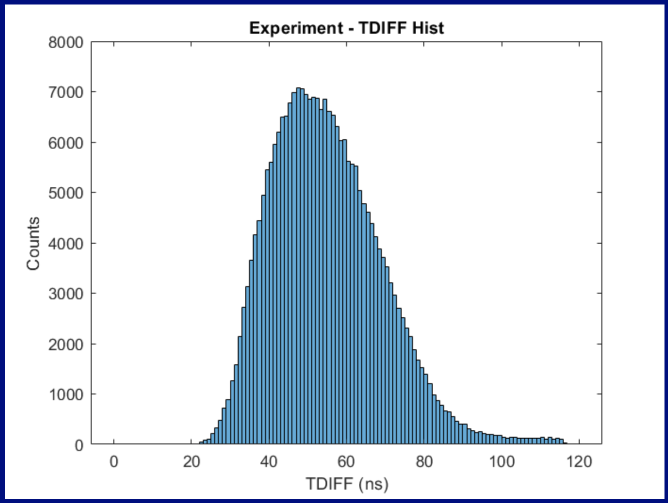
Sample time-of-flight (tof) plot fission showing prompt γ-rays iand neutrons in the liquid scintillators

Mark Wetzel, U New Mexico

^{242}Pu(sf) PFNS progress







Comparing Experiment with Simulation

Watt –Fission Spectrum is calculated in MCNP with the formula:

$$f(E) = ce^{-\frac{E}{a}} \sinh(\sqrt{bE})$$

Where:

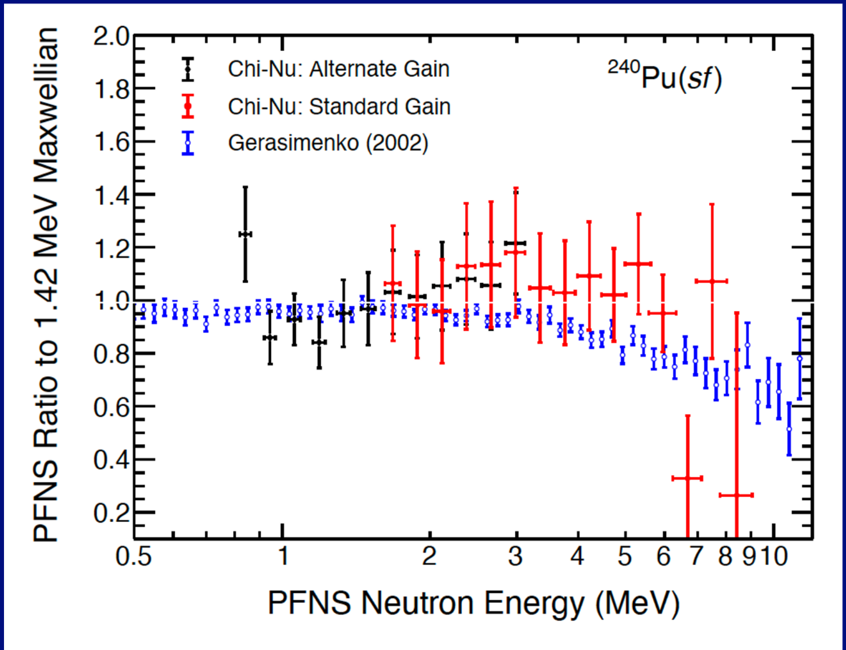
a = 0.819150

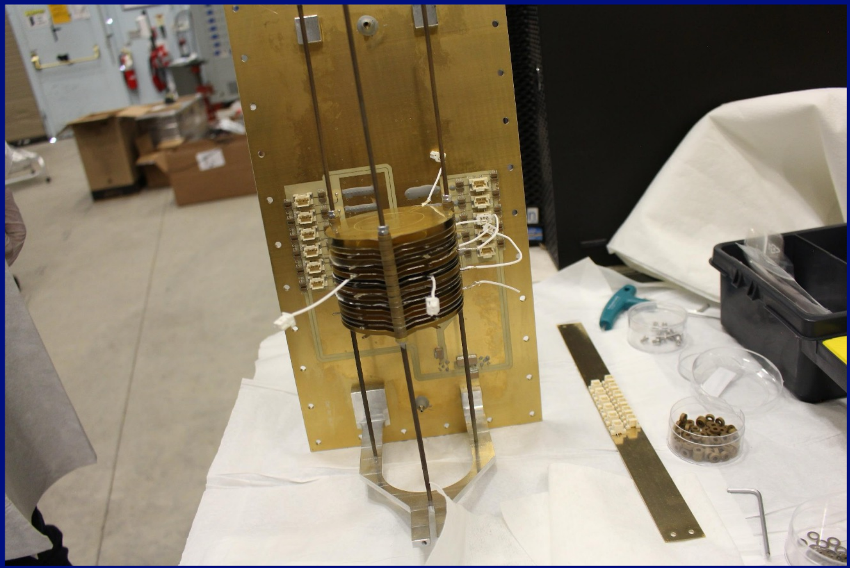
b = 4.36668

Mark Wetzel, U New Mexico

^{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...

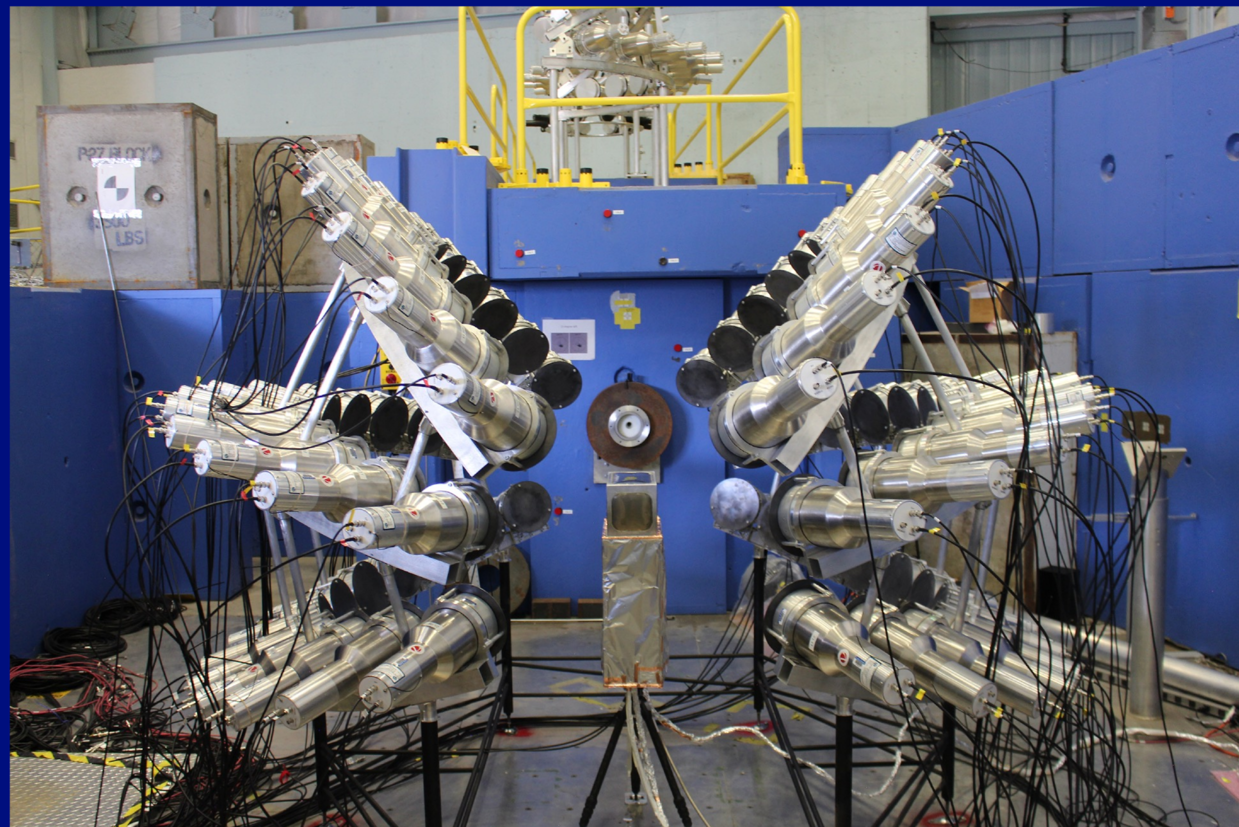




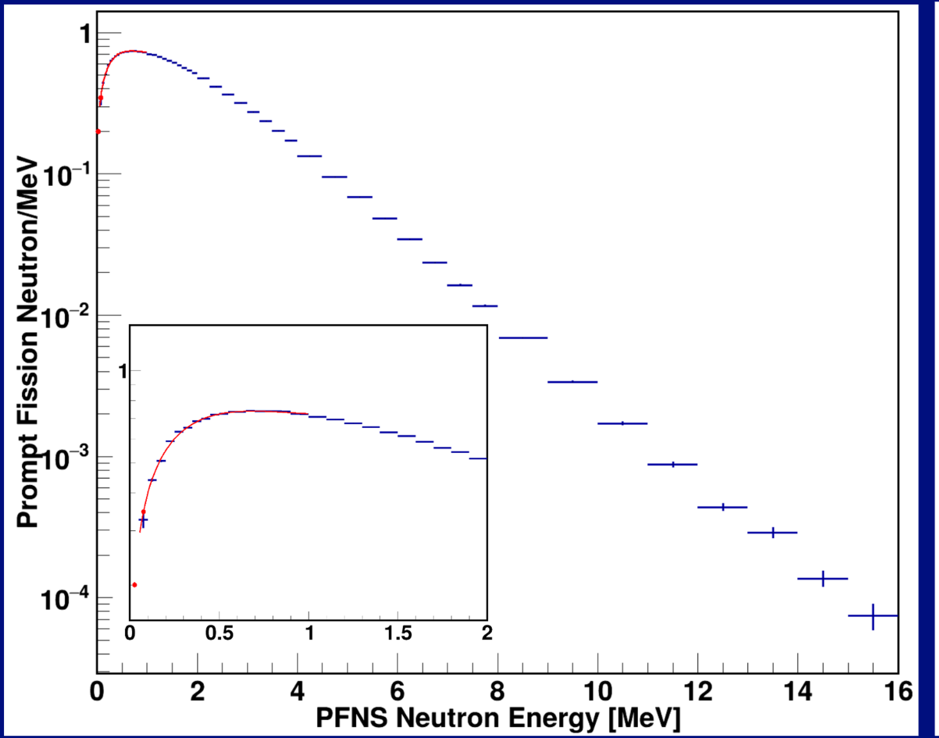
KJ Kelly et al., PRC 109, 064611 (2024)

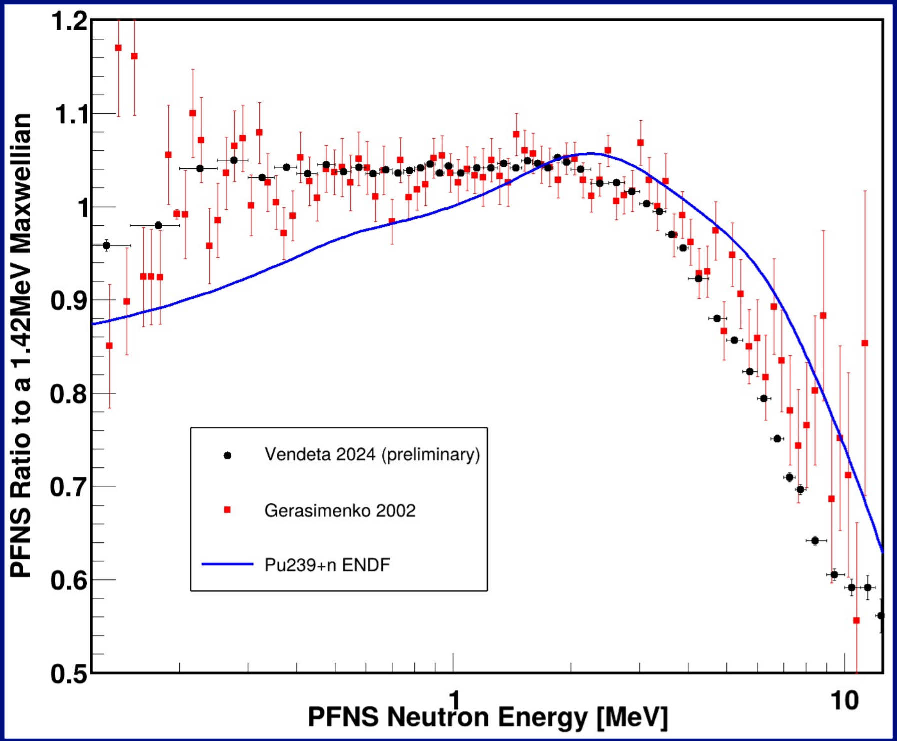
^{240}Pu(sf) PFNS measurement recently completed at LANSCE

- Used the VENDETA array of 72 EJ309 liquid scintillator detecotrs, on loan from the CEA
- Used a fission chamber with fast timing, with 22 foils of ^{240}Pu with a total mass of 11 mg
- Approximately six months of sf data taken



$^{240}Pu(sf)$ PFNS preliminary result

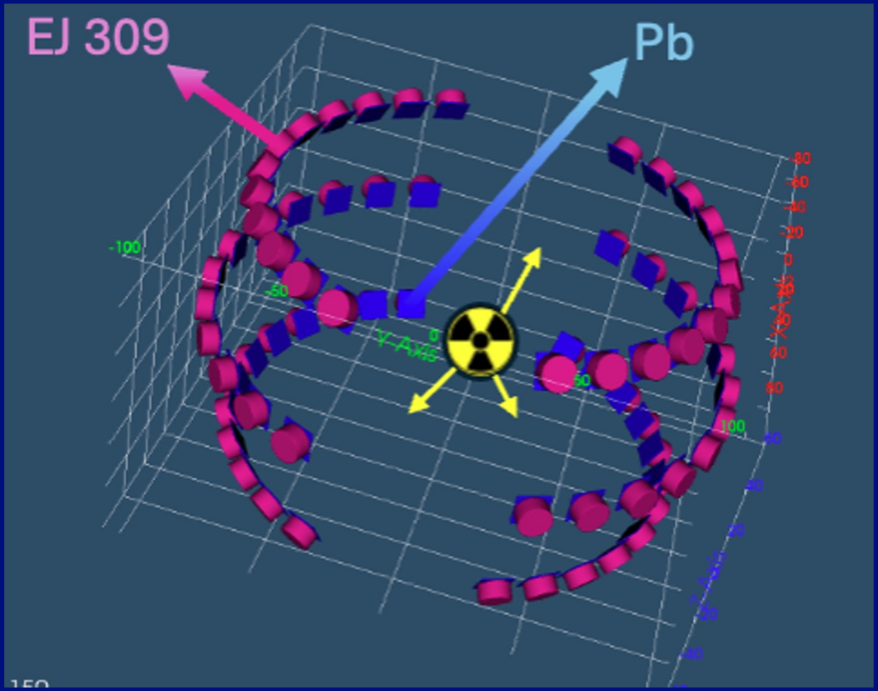




- Data analysis done by Owen Syrett (CEA)

MCNP Simulation Pipeline

- **Goal**: Obtain nn efficiency
- The VENDETA array detectors were modeled using MCNPX-PoliMi[1]
  - 72 EJ 309 detectors and 5 mm of Pb
  - 1 m distance from origin
  - Pu-240 Polimi source, neutron only
- **MCNPX_PoliMi** creates a file with collision data in all detectors that is processed by **MP_Post[2]**
  - Birks' response function taken from literature[3] and 50 keVee light output threshold
- Outputs Coincidence events for detector pairs
  - Used with number of fissions to calculate efficiency

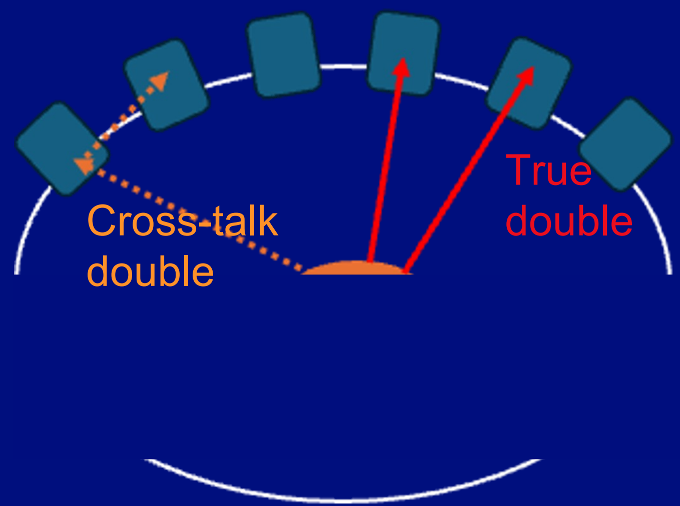


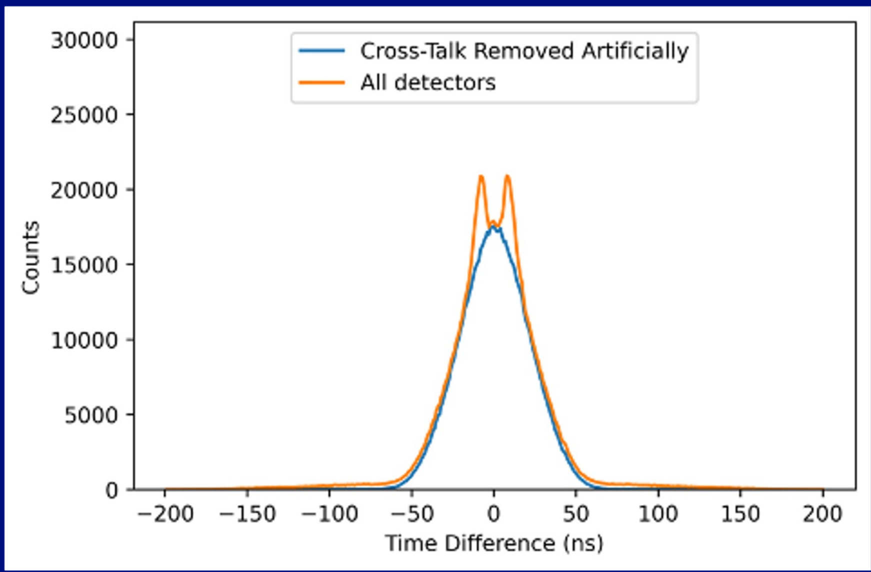
Ethan Schneider, U of Michigan

[1] Pozzi, Sara A., Enrico Padovani, and Marzio Marseguerra, *NIM A* **513** (2003): 550-558.
[2] Miller, Eric C., et al., *J of Nucl. Mat. Man.* **40** (2012): 34-41.
[3] Enqvist, Andreas, et al., NIM A **715** (2013): 79-86.

Simulated Neutron Doubles Efficiency

- Detected neutron doubles are a combination of true doubles and cross-talk doubles
- MPPost has the capability to remove cross-talk events
- Removing cross talk events reduces the neutron doubles efficiency by 15%

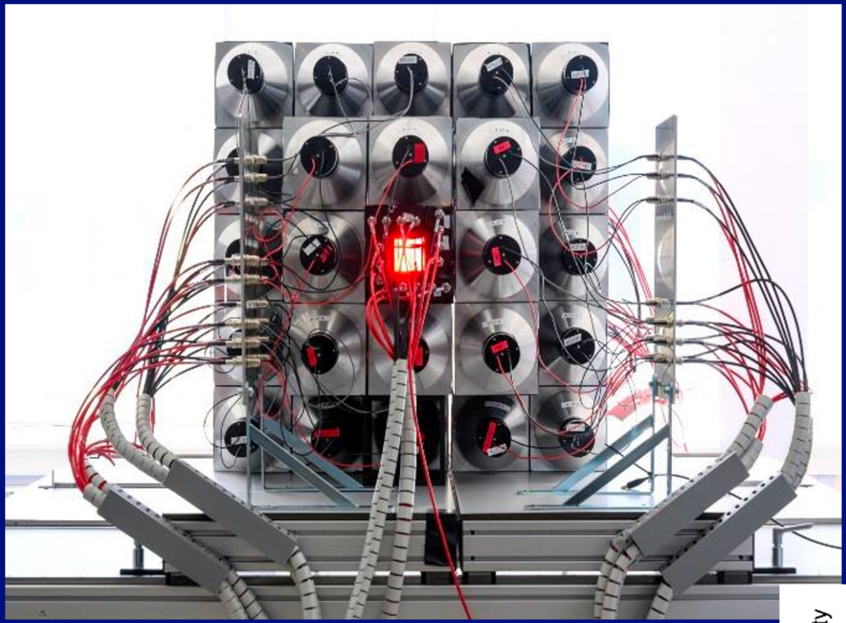


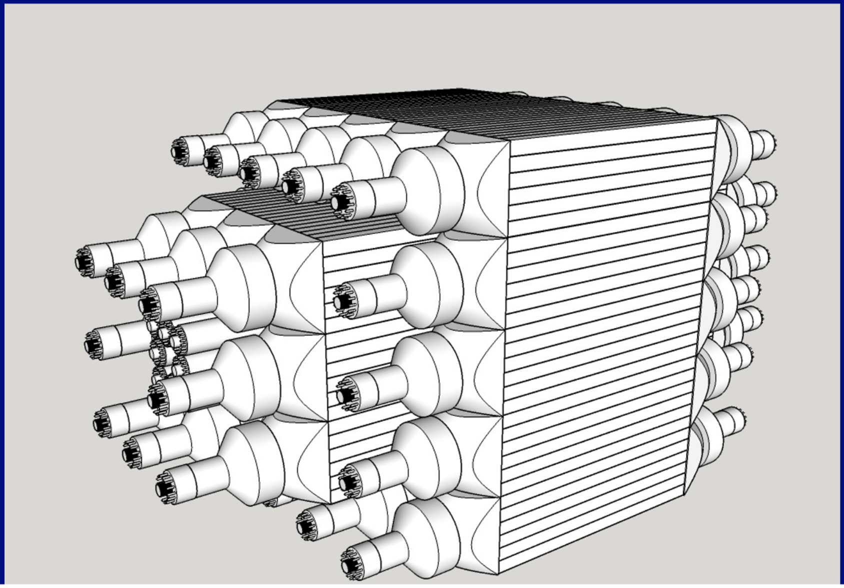


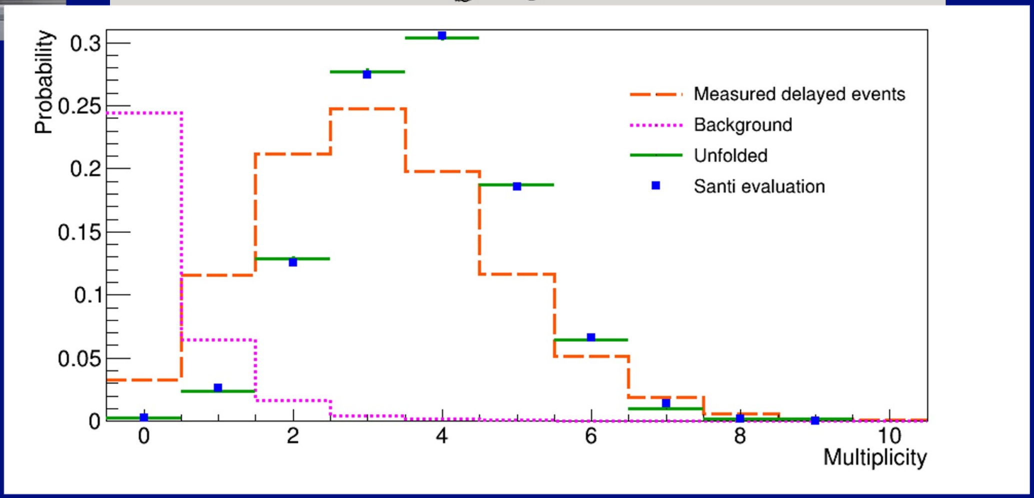
Total Efficiency	Efficiency with cross talk Removed by MP_Post
0.217(2)%*	0.184(2)%*

***Uncertainty is purely statistical error from counts ($\sqrt{N}$/# fissions)**

Various high-efficiency neutron counter designs were considered, and we chose to use SCONE







Plastic scintillator bars with thin inter-bar Gd layers

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

Update on Pu(sf) simulations with FREYA

- Complete fission model FREYA simulates spontaneous fission
- FREYA can take new data in this project and apply it to produce a more rigorous optimization and, eventually, a new evaluation

$$E^*_{sc} = aT^2_{sc} \quad \text{where} \quad a \approx A_0/e_0$$

$$\overline{E}^*_L = x\acute{E}^*_L$$

$$\sigma^2_{E_i} = c\overline{E_i}^* T_i$$

$$T_S = c_S T_{sc}$$

FREYA parameters:
e_0 in level density a;
related to neutron spectrum, fragment de-excitation
x, excitation advantage for light fragment;
related to $\nu(A)$, n-n correlations
c, fluctuations in intrinsic excitation energy;
related to $P(\nu)$, width of TKE distribution
c_S, multiplies scission temperature;
related to photon observables, rotational energy
dTKE, used to better fit average neutron multiplicity

$$\chi^2_O = \frac{1}{n-5}\sum_{i=1}^{n}\frac{(O_i - E_i)^2}{\sigma_i^2}$$

$$\chi^2 = \sum_O \chi^2_O$$

$$P(\chi^2_0, \chi^2_1) = \exp\left(\frac{\chi^2_0 - \chi^2_1}{\chi^2_0 T}\right)$$

Optimization procedure:

- Pick 5 FREYA parameters randomly
- Run 1M FREYA events for each set
- Compare outputs to data to obtain χ^2
- Use simulated annealing to make random walk in parameter space to update parameters
- When χ^2 is reduced sufficiently use grid search in small region to obtain minimum χ^2

Preliminary results for ^{240}Pu(sf)

- Reduced χ^2 relative to VanDyke et al., NIM A **922**, 24 (2019)
- Included new data in fits to improve optimized parameters, thus improving FREYA's description of ^{240}Pu(sf)

Optimized values:

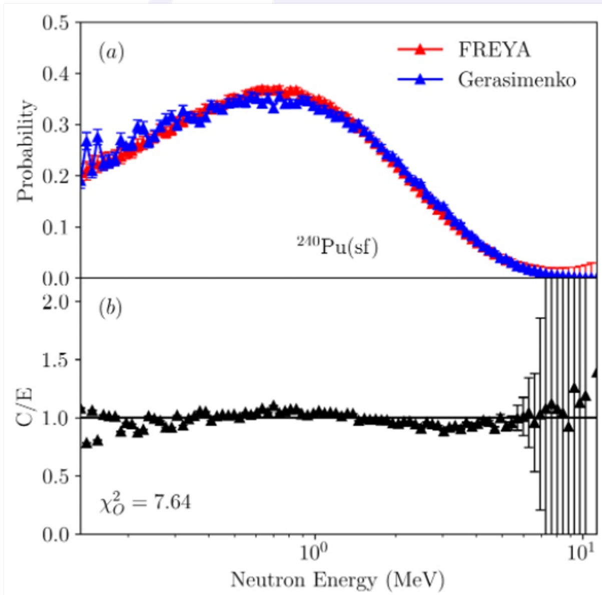
e_0 = 11.83 ± 0.13 MeV^{-1}

x = 1.14 ± 0.08

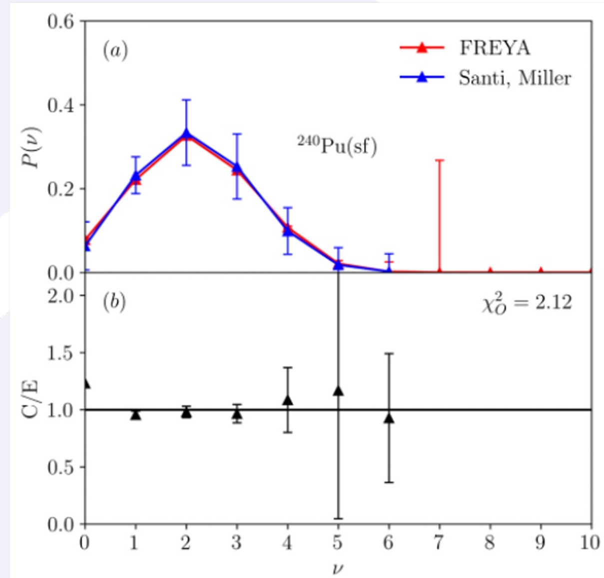
c = 3.07 ± 0.71

c_S = 0.94 ± 0.01

dTKE = -2.90 ± 0.09 MeV



PFNS from Nucl. Sci. Tech. **39**, 362 (2002)



P(ν) from Nucl. Sci. Eng. **160**, 190 (2008); uncertainties increased to keep from dominating fit

New data from this project could replace P(ν) evaluation above with more realistic data, yielding a value of c closer to unity (as expected)

Thank You!

and

Any Questions?