Improving the Nuclear Data on Fission Product Decays at CARIBU

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Lawrence Livermore National Laboratory

Joint Proposal of LLNL and ANL

FY18-22

Funded by DOE/NNSA/ DNN R&D (NA-22)

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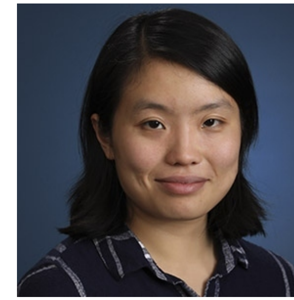

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Funded by DOE Office of Nuclear Physics/ Nuclear Data

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Our Collaborators

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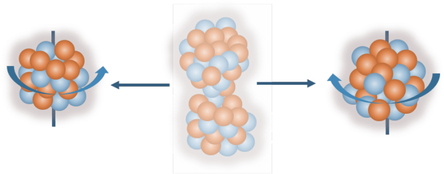
LBNL: R. Orford

The Goal of this Project is to Perform Detailed Studies of Key Fission-Product Decay Properties

Nuclear Data Impacts Understanding of Nuclear Events:

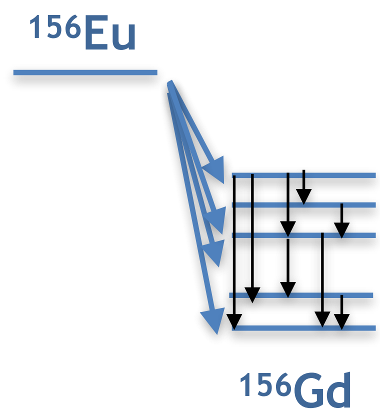


Fission-product isomer-to-g.s. ratios and masses

Understanding of fission dynamics and angular momentum

Measurements of short-lived fission products

- $^{132m/132}$Sb isomer to g.s. ratio & mass measurements
- $^{128m/128}$Sn and $^{128m/128}$Sb isomer to g.s. ratios & mass measurements



Beta-delayed γ-ray branching ratios

Nuclear data for fission yields – impacts nuclear forensics

Precision branching ratio of long-lived fission products of the importance to nuclear forensics

- ^{156}Eu precision decay measurement
- Improved Geant4 simulations
- Planning to collect ^{161}Tb & ^{111}Ag in the spring

Isotope	Half-life [days]	γ-ray energy [keV]	Current branching ratio [%]
^{156}Eu	15.2	811.8	(9.7±0.8)
^{161}Tb	6.9	74.6	(10.2±0.5)
^{111}Ag	7.5	342.1	(6.68±0.33)
^{127}Sb	3.85	685.5	(36.8±2.0)

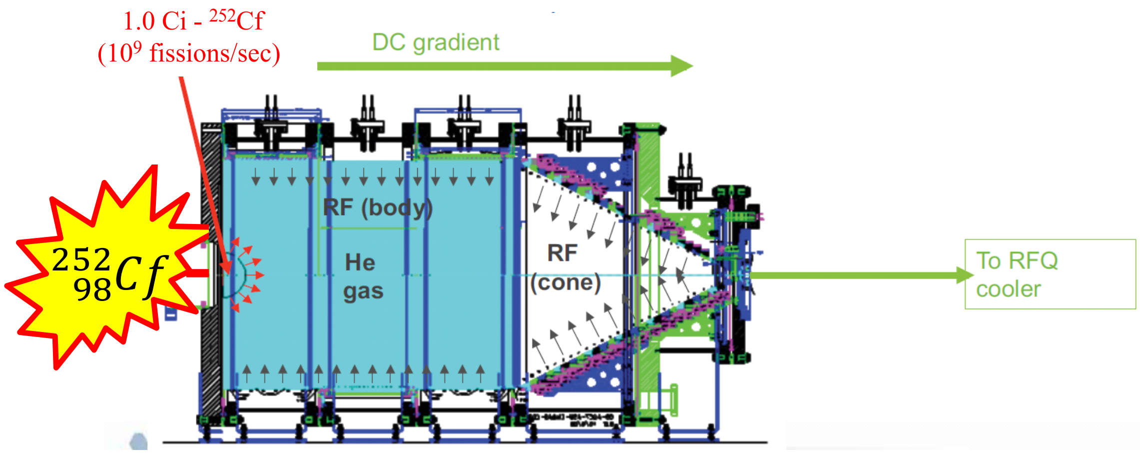
Uncertainties of the order of 5-10% need to be remeasured

CARIBU Opens Up Opportunities to Study Decays of Fission Products

CAlifornium **R**are **I**sotope **B**reeder **U**pgrade

^{252}Cf spontaneous fission source

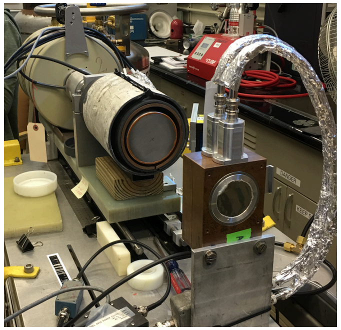
Mass-separated beams of any fission product with $t_{1/2}$ > 25 ms



G. Savard, R. Pardo. Proposal for the ^{252}Cf source upgrade to the ATLAS facility. (2005)

CPT Measurements

Precision Branching Ratio Measurements



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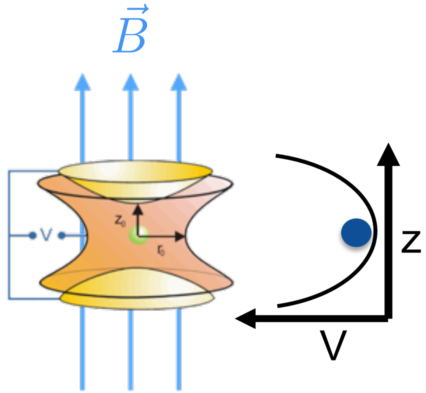
We're Working with ANL to perform Isomer Mass Measurements

→ Identify isomer and ground state by mass, this method is independent on decay properties

Mass and energy: $E = mc^2$

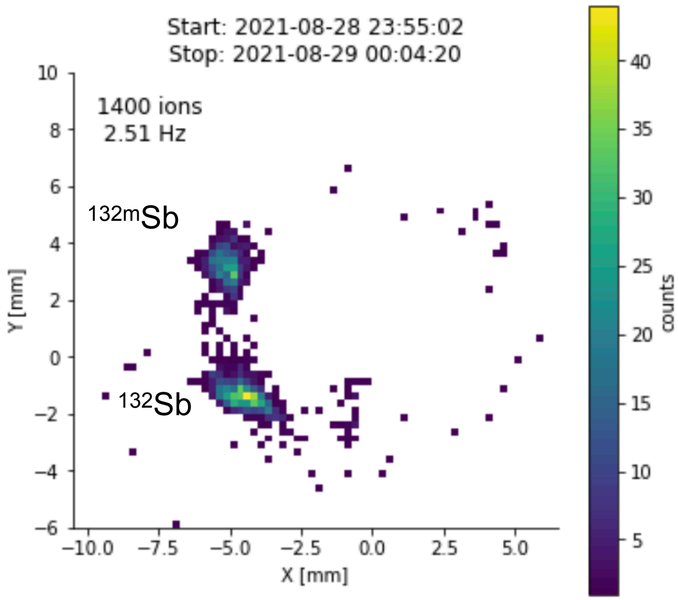
$$E + \Delta E_{isomer} = (m + \Delta m_{isomer})c^2$$



→ ions in the strong magnetic field of a Penning trap, where the frequency of the ion's cyclotron motion depends on the mass of the ion

Isomer mass measurements of ^{132,132m}Sb

→ ions ejected from the trap and transported to a position-sensitive detector



Preliminary result:

^{132m}Sb ΔE =145.6(1.1) keV

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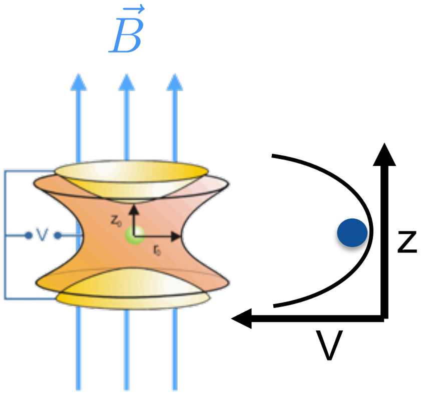
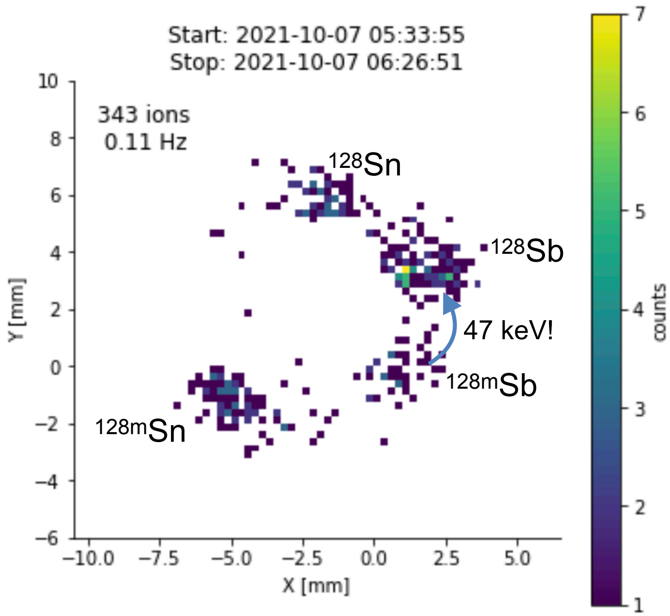


→ ions in the strong magnetic field of a Penning trap, where the frequency of the ion's cyclotron motion depends on the mass of the ion

Isomer mass measurements of ^{128,128m}Sb and ^{128,128m}Sn

→ ions ejected from the trap and transported to a position-sensitive detector



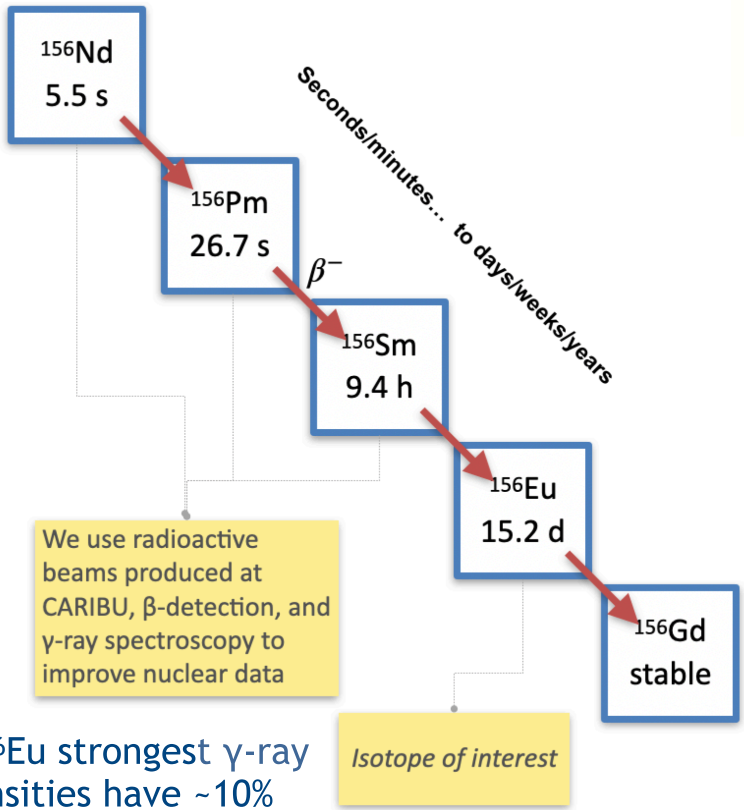
Preliminary result:

^{128m}Sb ΔE = 47(2) keV

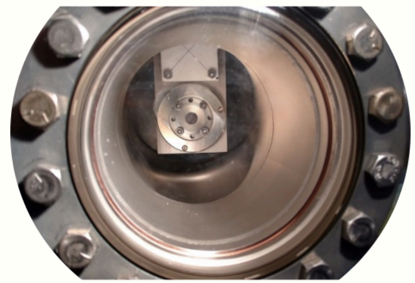
First Measurement!

Precision Branching Ratio Measurements of Long-lived Fission Products

→ Many long-lived fission products measured in 60s and 70s have high uncertainties on decay branching ratios



→ ^{156}Eu strongest γ-ray intensities have ~10% uncertainties



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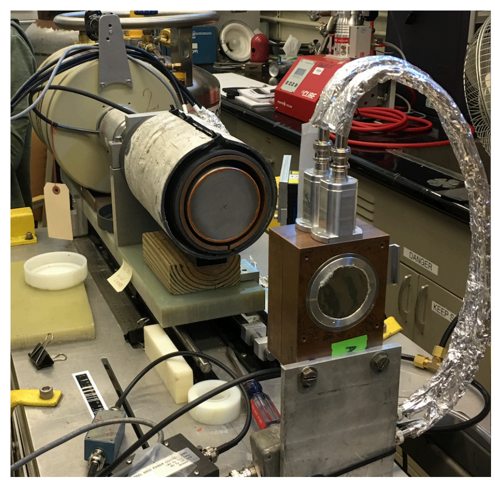
Sample harvesting

→ Implant mass-separated radioactive ion beam on thin carbon foil at CARIBU (ANL)

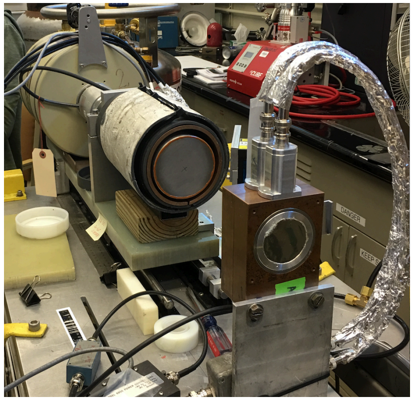
Decay measurement

→ β detection and γ-ray spectroscopy at TAMU



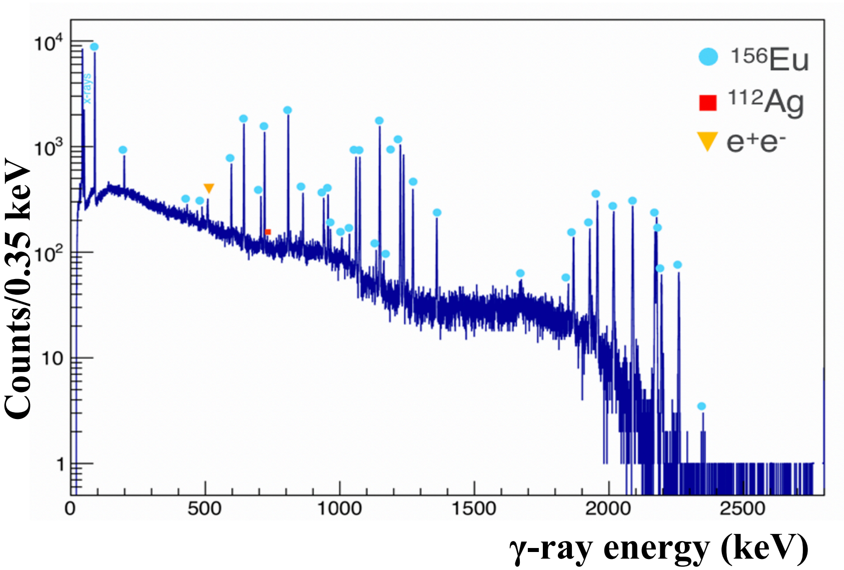
Method described in K. Kolos et al. "New approach to precisely measure γ-ray intensities …" NIM A 1000, 165240 (2021)

We Collected High-quality Data for ^{156}Eu



→ Data collected with precisely calibrated β-γ coincidence detection setup

→ data collected during 7 days



γ-ray energy	I_γ (NNDC)	Uncert. [%]
88.97	8.4(11)	13.1
599.47	2.08(17)	8.2
646.29	6.3(5)	7.9
723.47	5.4(4)	7.4
811.77	9.7(8)	8.3
1065.14	4.9(4)	8.2
1079.16	4.6(4)	8.7
1153.67	6.8(6)	8.8
1154.08	4.7(4)	8.5
1230.71	8.0(7)	8.7
1242.42	6.6(5)	7.6
1965.95	3.9(3)	7.7
2026.65	3.3(3)	9.1
2186.71	3.5(3)	8.6

→ Uncertainties >5%

→ Previous measurements used neutron capture from reactors i.e. much dirtier spectrum.

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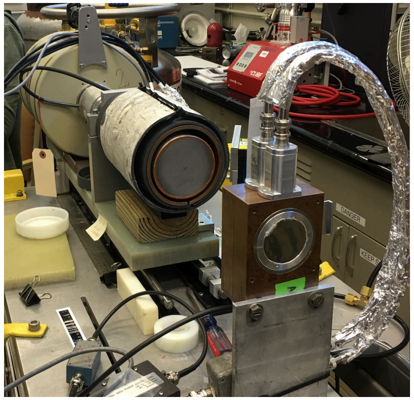

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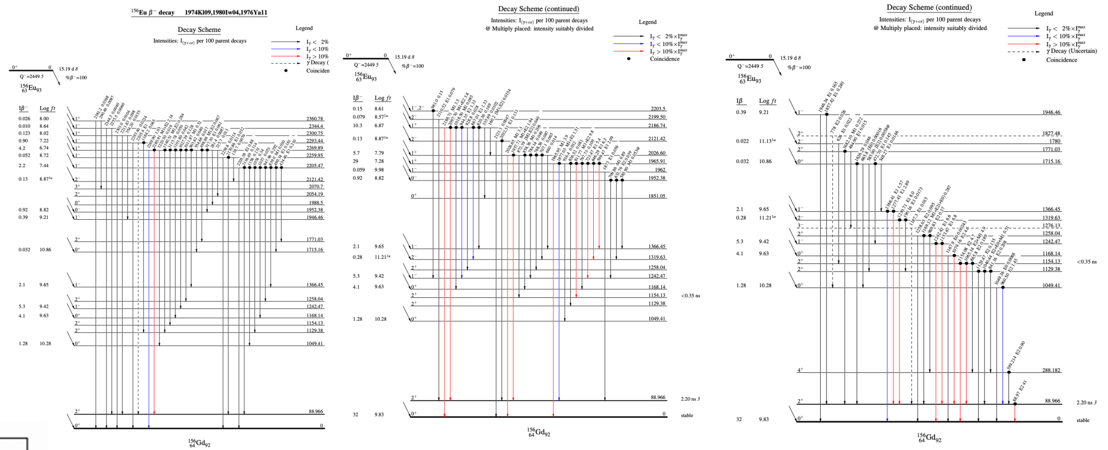
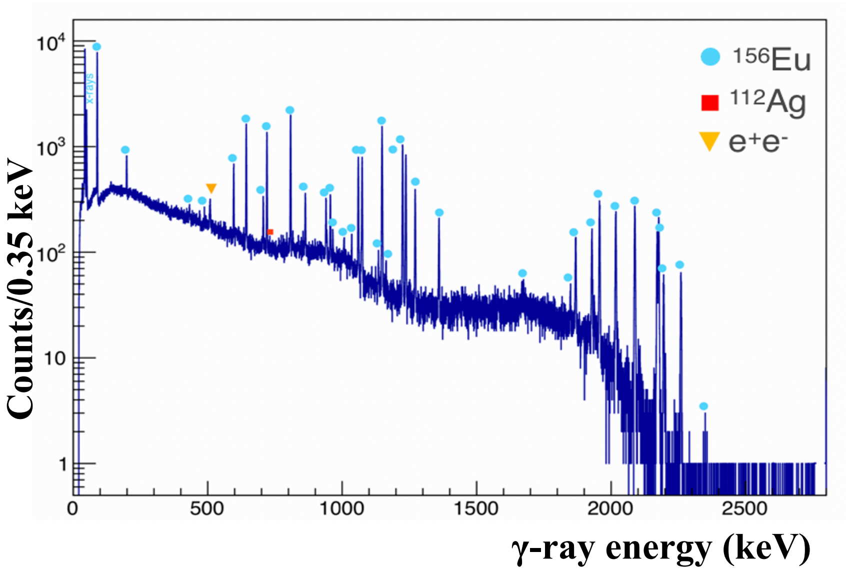
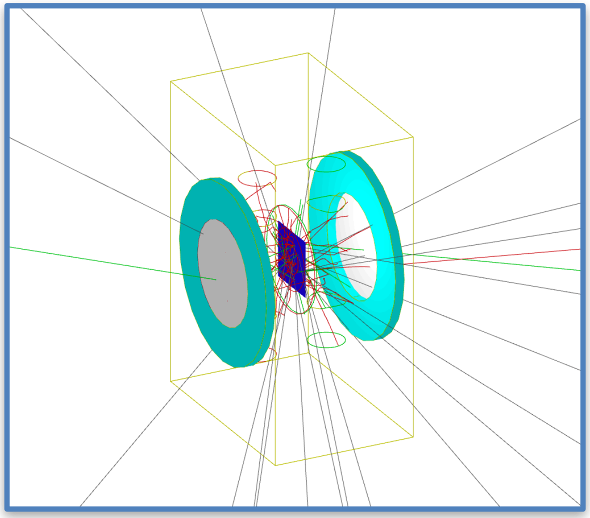




- → ^{156}Eu provides a unique simulation challenge
- → Simulations now tracks gamma rays emitted allowing for easier calculations and run on LLNL supercomputers



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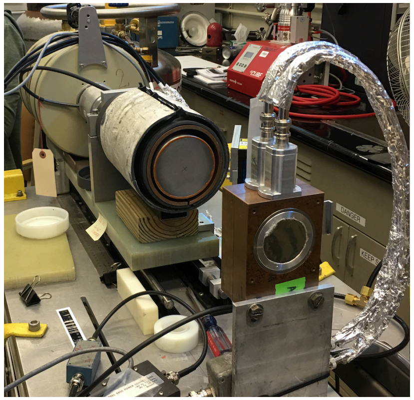

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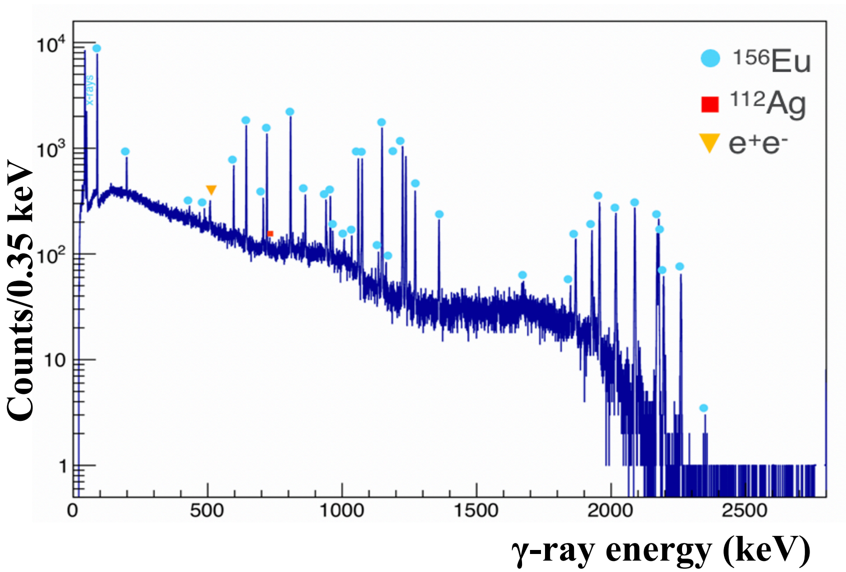
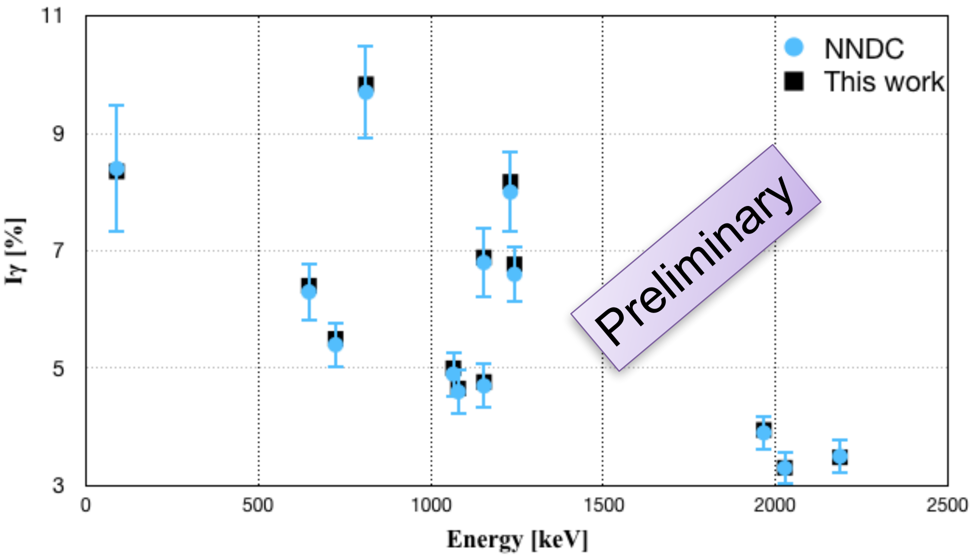


With our technique we were able improve ^{156}Eu decay data and reach sub-1% precision!

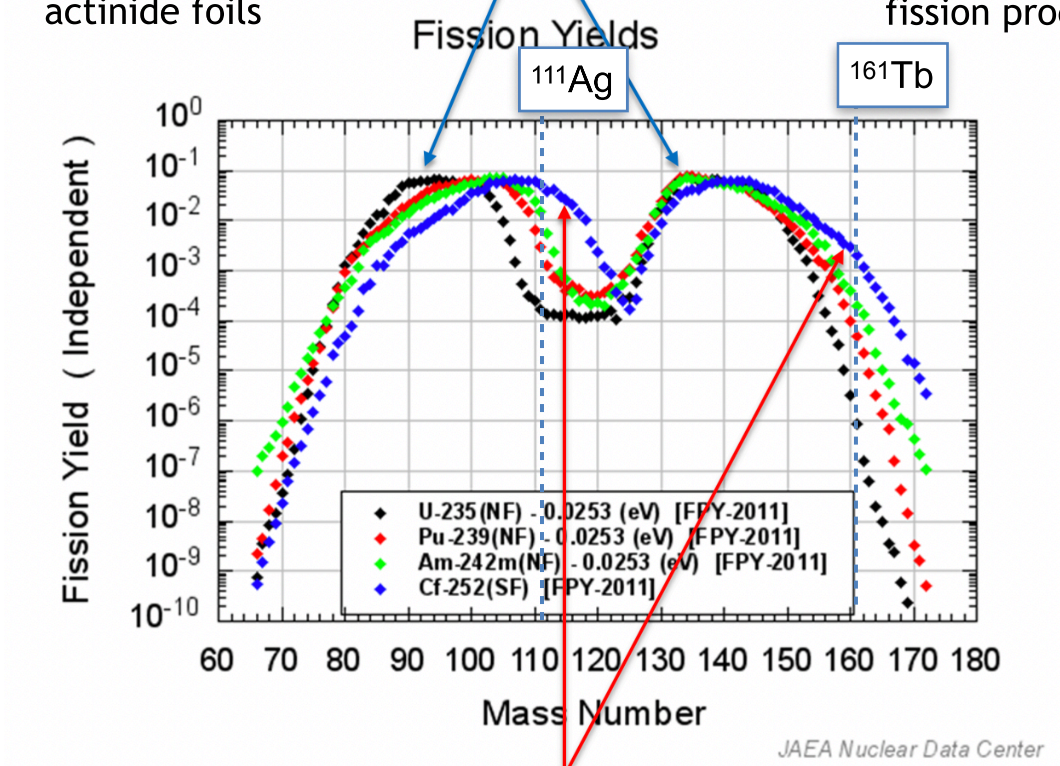


→ Comparison of current (NNDC) evaluated data with our results for γ-ray branching ratios

CARIBU Will Be Changing to nuCARIBU

→ ^{252}Cf source will be replaced by neutron-induced fission on actinide foils

Significant gain



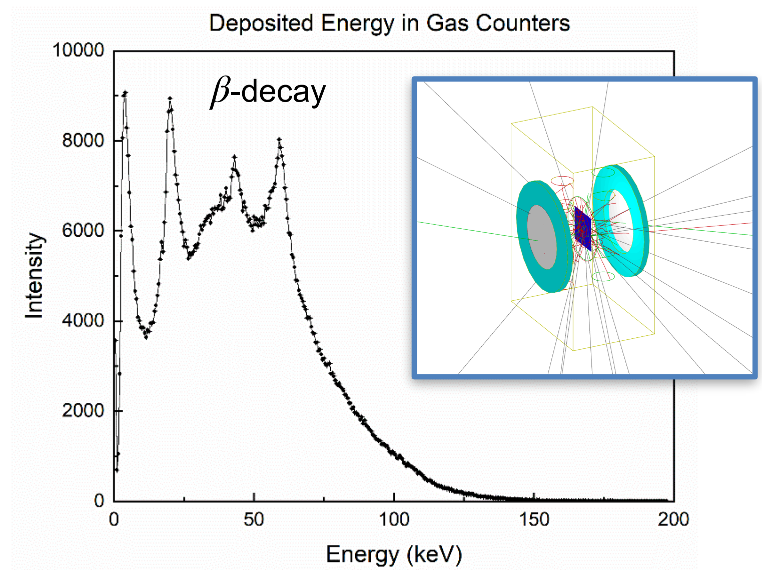
Significant loss

→ Operationally easier to maintain and operate

→ More reliable source of fission products

→ Plan to collect ^{161}Tb & ^{111}Ag this Spring before switch

→ Simulations already finished for ^{161}Tb



Results Dissemination and Outlook

We publicize our results at workshops, conferences, reviews:

-Independent Review in November 2020
-APS DNP 2021/2022

Publications:

K. Siegl, K. Kolos, N. D. Scielzo et al. "Beta-decay half-lives of $^{134,134m}Sb$ and their isomeric yield ratio produced by the spontaneous fission of ^{252}Cf" PRC 98, 054307 (2018)

K. Kolos, A. M. Hennessy, N. D. Scielzo et al. "New approach to precisely measure γ-ray intensities for long-lived fission products, with results for the decay of ^{95}Zr" NIM A 1000, 165240(2021)

+ Working on publication of ^{156}Eu (D.E.M. Hoff)
+ More from CPT measurements

Worked with many graduate students:

- Kevin Siegl (Notre Dame), B. Champine and Tyler Nagel (UC Berkeley), A. Hennessy and E. Heckmaier (UC Irvine), Erin Good (LSU), Benjamin Schroeder (TAMU)

Looking for new graduate students!

→ More experiments to come: isomeric-to-g.s. with the CPT and precision decay studies of long-lived FP branching ratios