Nuclear Science and Security Consortium

**September Workshop and Advisory Board Meeting**

# Demonstration of an Approach to Precisely Measure Gamma-ray Branching Ratios for Long-Lived Beta Emitters

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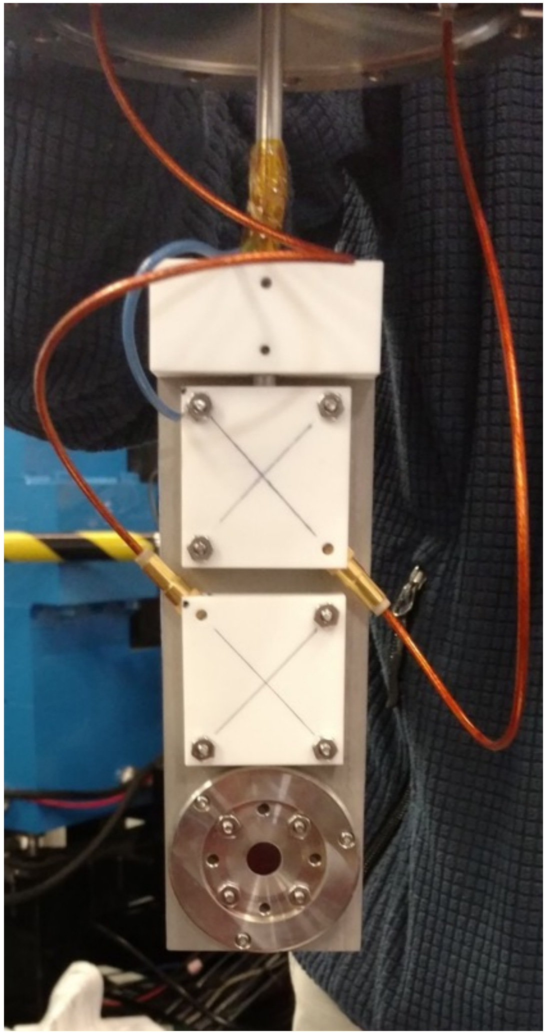
**Modeling and Simulation, Radiation Detection and Measurement, Nuclear Data**

# Presentation Outline

- Project Overview (Specifically $^{95}Zr$)
- Sample Production – CARIBU
- γ-ray and βγ Coincidence Measurement
- β Detector Simulation – GEANT4
- Beta Efficiency Calculation
- Branching Ratio Calculation
- Future Plans ($^{144}Ce$ and $^{147}Nd$)

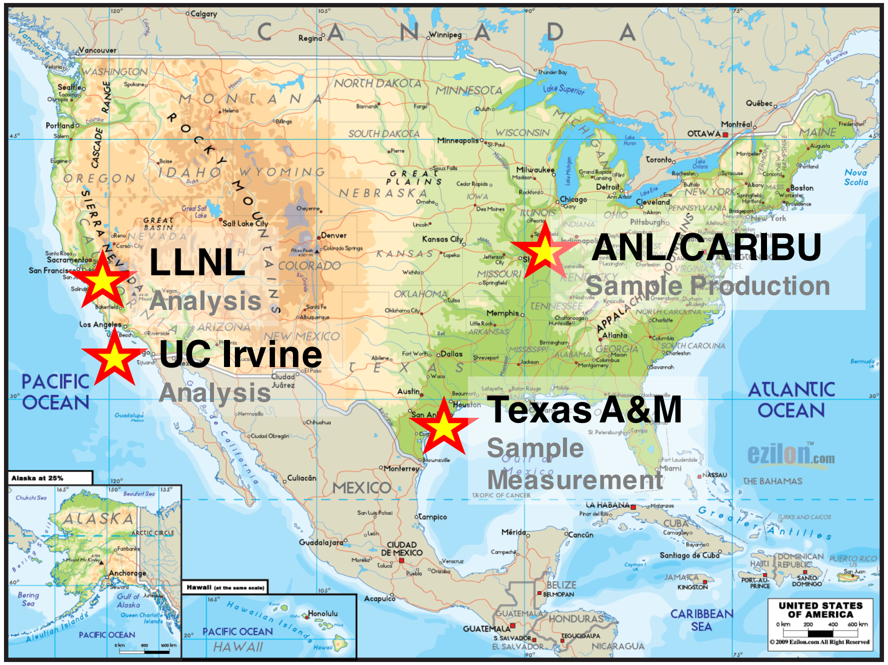


# Project Overview

## Reduce uncertainties in fission product γ-ray branching ratios

Collaboration between UC Irvine, Lawrence Livermore National Lab (LLNL), Argonne National Lab (ANL), and Texas A&M University (TAMU)

- Create ultra-pure radioactive sample at the CARIBU facility at ANL
  - $^{95}$Zr (calibration), $^{147}$Nd, $^{144}$Ce
- Measure β particles and γ-rays in coincidence at TAMU
  - **β particles** measured with custom-built 4π gas-flow proportional counter
  - **γ-rays** measured with precision HPGe
- Simulate β detector response to confirm experimental measurements

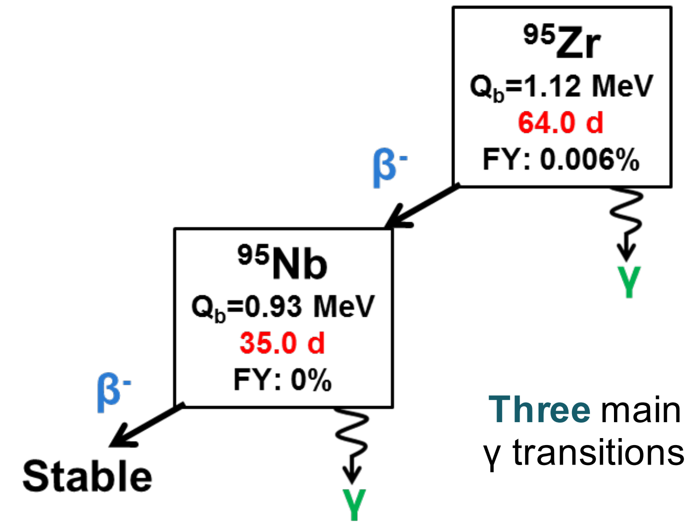


# Branching Ratio

- **Branching ratio is the fraction of a specific decay emitted over all the decays**

$$BR = \frac{specific\ decay\ mode}{total\ decay}$$

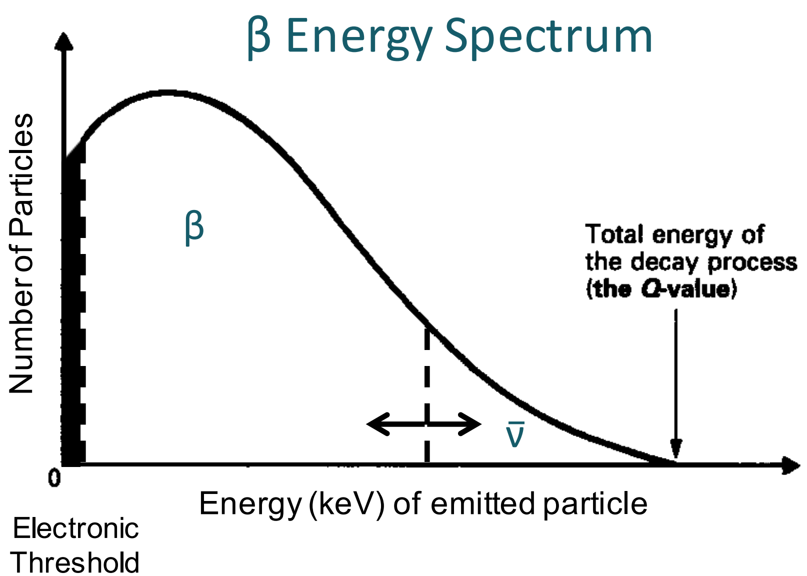
| $^{95}Zr$ / $^{95}Nb$ γ-rays | | |
|---|---|---|
| | Energy (keV) | Branching Ratio (%) |
| $^{95}Zr$ | 724.2 | 44.27 (22) |
| $^{95}Zr$ | 756.7 | 54.38 (22) |
| $^{95}Nb$ | 765.8 | 99.808 (7) |



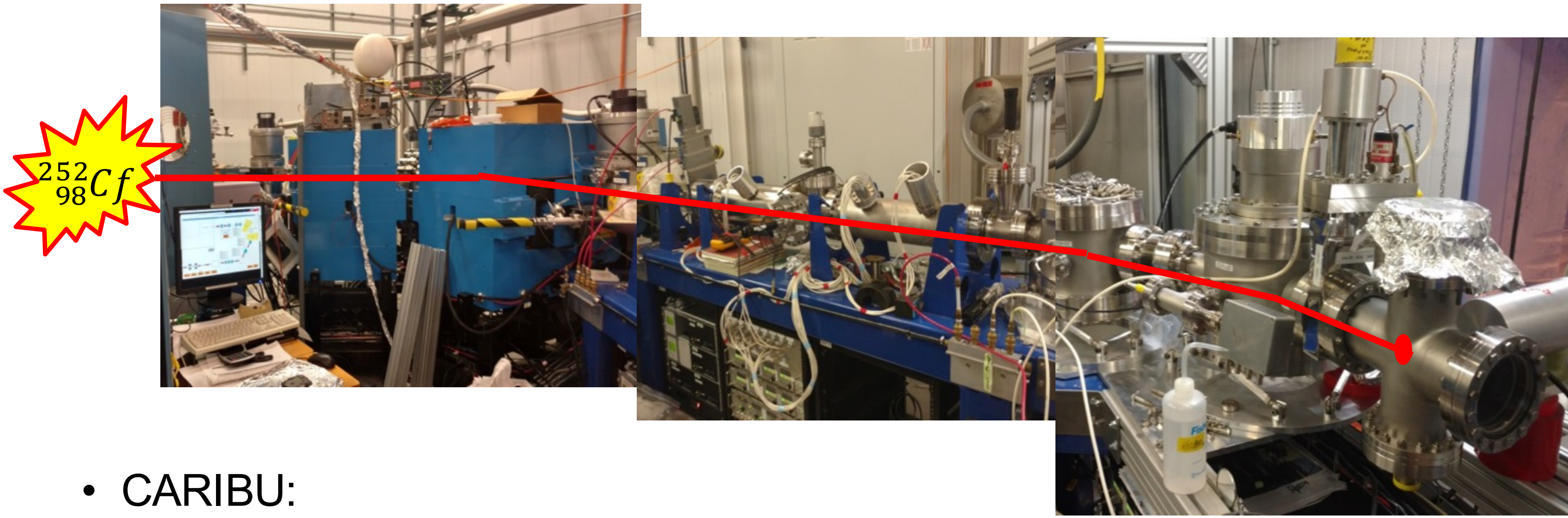

- **$^{95}Zr$ BR is well known**
  - Used as a calibration source, test of the method
- **BR can be used to determine the number of decays**
  - $N_{decays} = \frac{N_\gamma}{BR_\gamma * \varepsilon_\gamma}$
  - From the number of decays, one can determine the number of fissions with more certainty

# Difficulties

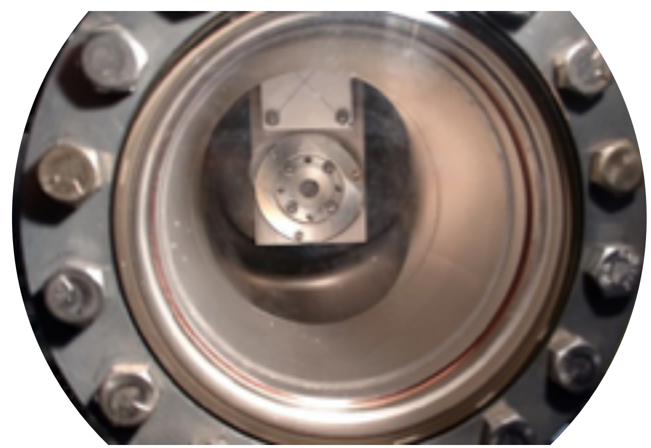
- **“Dark” Decays ($^{147}$Nd, $^{144}$Ce)**
  - Not all γ-rays are emitted (internal conversion electrons instead)
  - All emitted γ-rays, β particles, and CE need to be measured
- **Purity of Sample**
  - Difficult to separate interested β particle decays from contaminants
- **Self-Attenuation**
  - Low Q value energies
    - 399, 367, and 160 keV
  - Charged particle gets absorbed in foil and not detected
- **Precision Measurement**



# CARIBU – Source Production

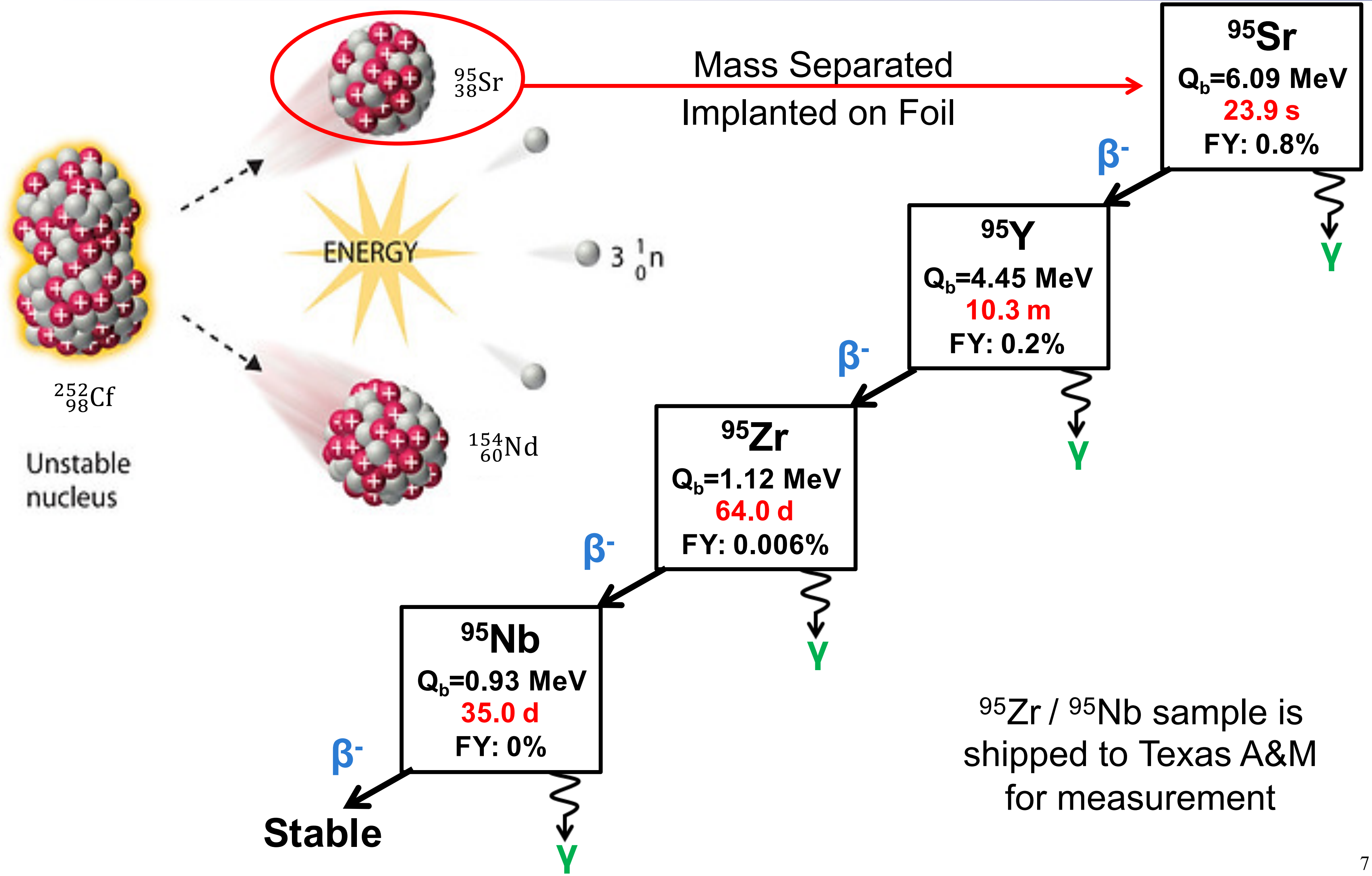



- CARIBU:
  - $^{252}$Cf spontaneous fission source
  - Ionized to +2 charge
  - Mass separated (A=95)
  - Implanted onto ultra-thin carbon foil
- Reduces effects from sample purity and self-attenuation



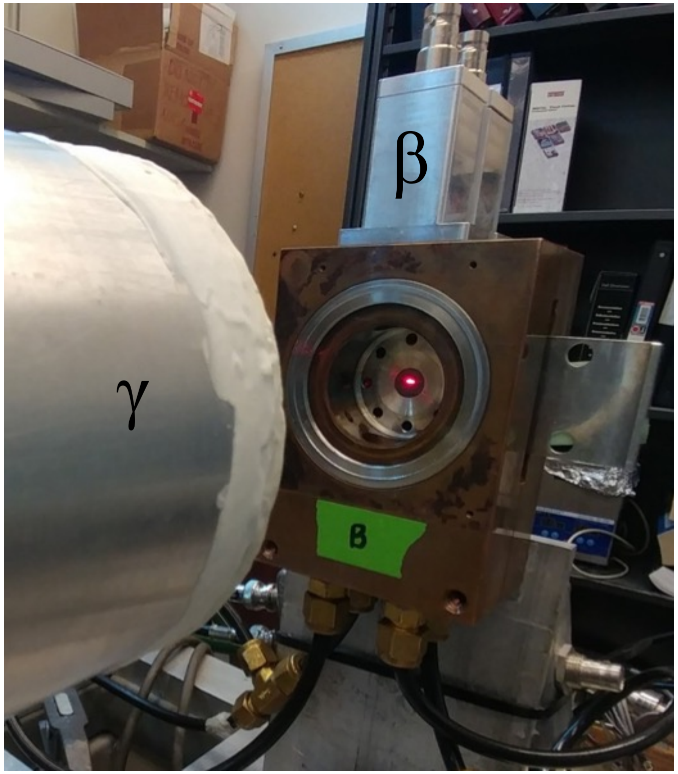
0.2 µm Carbon Foil

# Isotope Details – $^{95}Zr$, A = 95



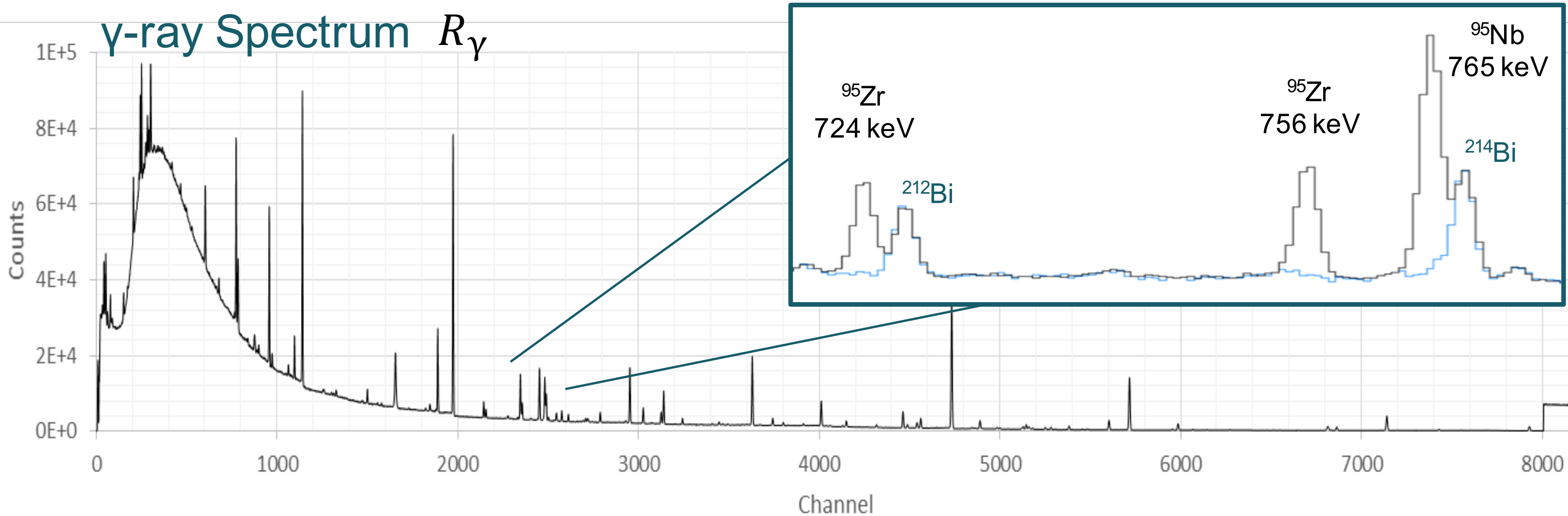

$^{95}Zr$ / $^{95}Nb$ sample is shipped to Texas A&M for measurement

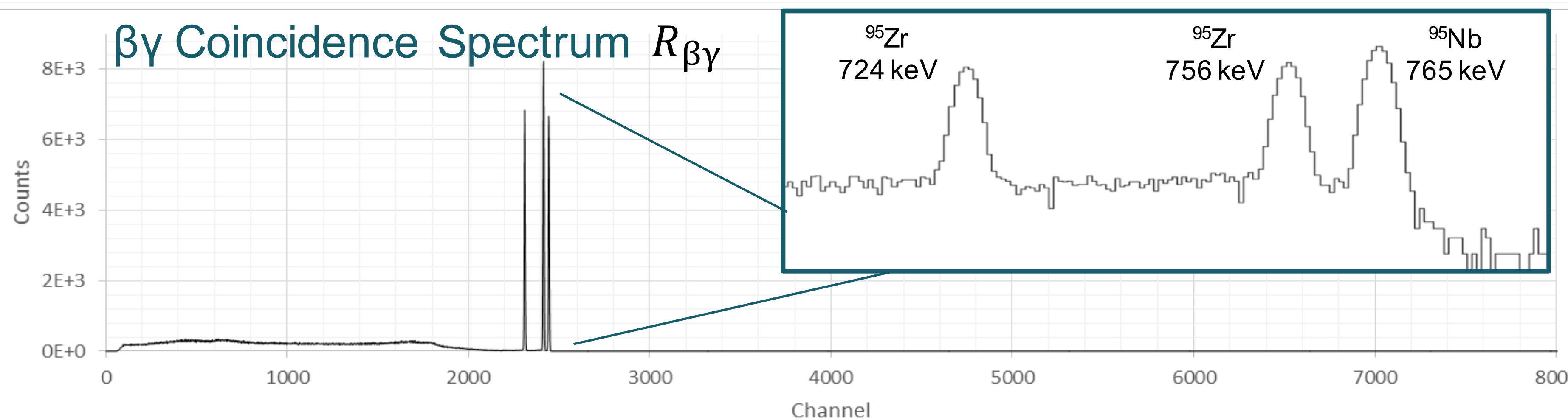
# Experimental Measurement (TAMU)




- **HPGe (γ-ray):**
  - Detector efficiency known with uncertainty of 0.2%
- **β Detector**
  - 4π gas flow proportional counter
  - High detector efficiency for CARIBU samples
    - Dependent on the isotope and electronic threshold
- **Coincidence Measurement**
  - Must detect both a β and γ within 2 μs
  - Creates clean γ spectrum with little interference from background

# γ-ray Spectrum and βγ Coincidence Spectrum



# β Efficiency of β Detector

$$R_{\beta\gamma} = R * \varepsilon_\beta * \varepsilon_\gamma * BR$$

$$R_\gamma = R * \varepsilon_\gamma * BR$$

$$\frac{R_{\beta\gamma}}{R_\gamma} = \varepsilon_\beta \longleftarrow \text{Transition Specific}$$

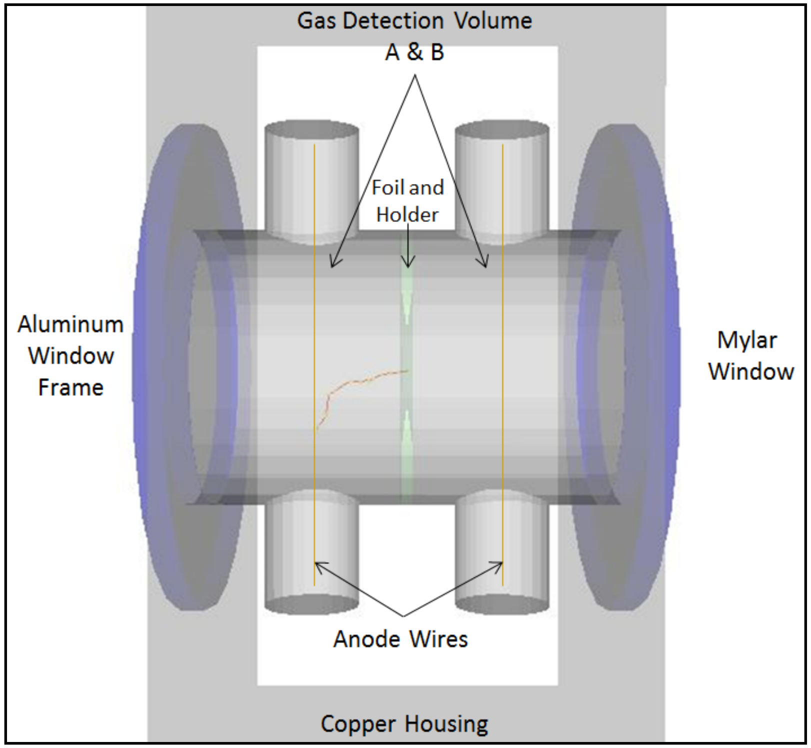
- $R_{\beta\gamma}$ = rate of βγ coincidence
- $R$ = rate of isotopic decay
- $R_\gamma$ = rate of γ coincidence
- $\varepsilon_\beta$ = transition beta efficiency
- $\varepsilon_\gamma$ = peak gamma efficiency
- $BR$ = γ-ray branching ratio

- **β detector efficiency**
  - Dependent on
    - Energy of β particle
    - Electronic threshold of detector

**Experimental measurements can be confirmed with simulation results!**

# GEANT4 Simulations of β Detector Efficiency

- **Simulate:**
  - 4π β detector design
  - Isotope specific
    - β energy spectrum
    - Fermi function (+ nucleus vs. – β interaction)
    - Nuclear size
  - Transition specific
    - β particle
    - γ-ray
    - Conversion electron
- **Compare experimental β detector efficiencies for specific transitions**




**Beta Efficiencies for $^{95}Zr$ and $^{95}Nb$**

| | | Simulations with a 4.6 keV Threshold | | Measured Values | |
|---|---|---|---|---|---|
| Isotope | β Energy (keV) | β Efficiency (%) | Uncertainty (%) | β Efficiency (%) | Uncertainty (%) |
| $^{95}Zr$ | 366.9 | 76.90 | 0.80 | 76.85 | 0.76 |
| $^{95}Zr$ | 399.4 | 78.70 | 0.77 | 79.89 | 0.69 |
| $^{95}Nb$ | 159.8 | 86.14 | 1.24 | 86.27 | 1.25 |

# Branching Ratio Calculation

$$BR = \frac{R_{\beta\gamma}}{\varepsilon_{\gamma} * R_{\beta_{isotope}}} * \frac{\varepsilon_{\beta_{isotope}}}{\varepsilon_{\beta_{peak}}}$$

- $R_{\beta\gamma}$ = rate of βγ coincidence
- $R_{\beta_{isotope}}$ = rate of emitted beta particles
- $\varepsilon_{\gamma}$ = efficiency of γ-rays
- $\varepsilon_{\beta_{isotope}}$ = efficiency of isotope beta particles
- $\varepsilon_{\beta_{peak}}$ = efficiency of specific transition

| $^{95}Zr$ Branching Ratios | | | | | |
|---|---|---|---|---|---|
| Isotope | Energy (keV) | Literature (Nudat, %) | Absolute Uncertainty (%) | Measured (%) | Absolute Uncertainty (%) |
| $^{95}Zr$ | 724.2 | 44.27 | 0.22 | 44.16 | 0.47 |
| $^{95}Zr$ | 756.7 | 54.38 | 0.22 | 54.16 | 0.56 |
| $^{95}Nb$ | 765.8 | 99.81 | 0.01 | 99.45 | 1.01 |

# Future Plans

- **Applying these calculations to other, more complex data sets:**
  - $^{147}$Nd data
  - $^{144}$Ce data
- **Apply more complex corrections**
  - Decay correct differences in collection time of γ-ray and βγ coincidence measurements
  - Correct for feeding of higher excited states
  - Correct for large influence of conversion electrons
- **Determination of uncertainty contributions**
  - Normalization of background γ-ray spectra
  - Gain shifts over time
  - Peak fitting abilities
- **Determine if repeat experiments are necessary**
  - Measurements of $^{95}$Zr and $^{147}$Nd

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