

Nuclear Resonance Fluorescence and Potential Uses in Active Inspection Technologies

Alan W. Hunt (IAC, Physics)

Mathew T. Kinlaw (Idaho National Laboratory)

Glen A. Warren (Pacific Northwest National Laboratory)

Edward T. E. Reedy (Physics, Ph.D. Student)

Heather A. Seipel (Physics, Ph.D. Student)

James Johnson (Idaho National Laboratory)

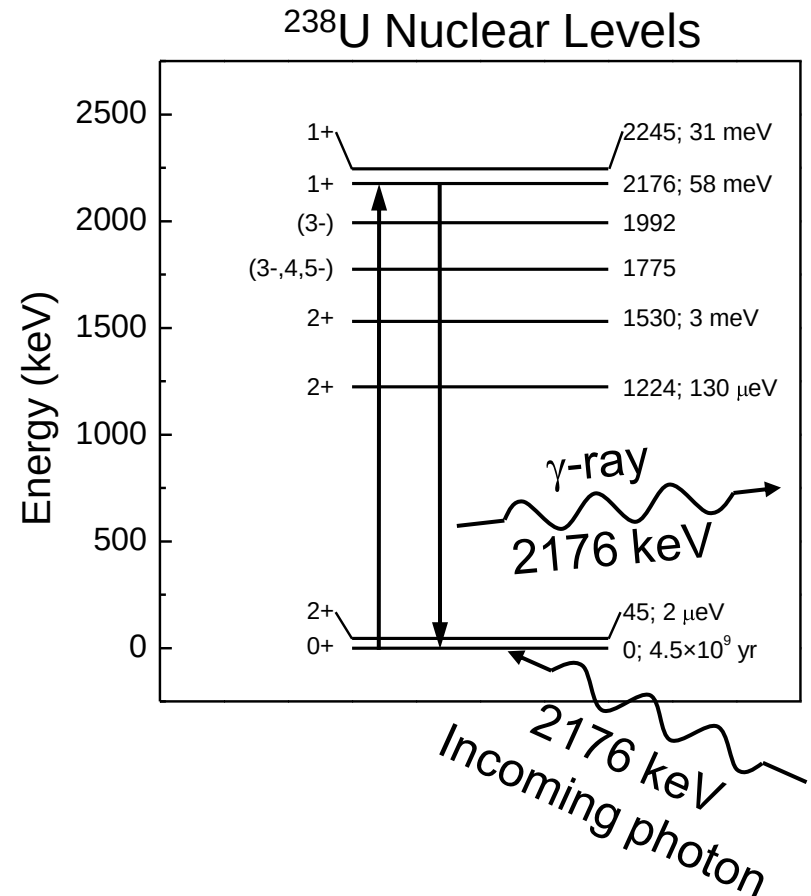
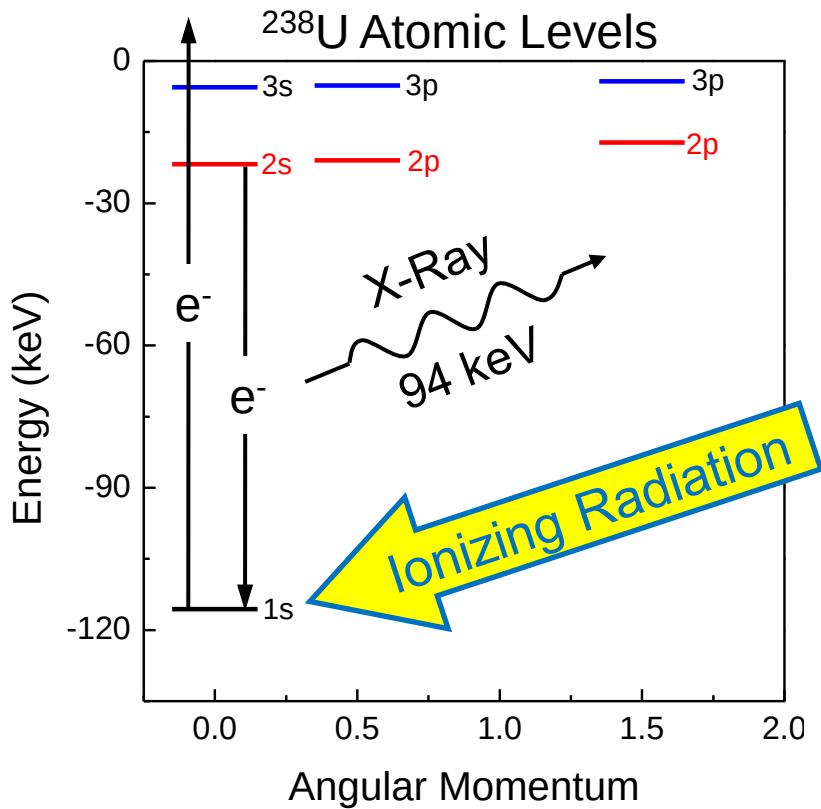
Scott M. Watson (Idaho National Laboratory)



Funding NA-22



NRF is Fluorescence Process Similar to XRF



- XRF: Elemental fingerprint
- NRF: Isotopic fingerprint
- Nuclear resonance fluorescence
 - High-energies 100's of keV to $\sim$ MeV
 - Probe photon must be on resonance

NRF Cross Section are Large and Narrow

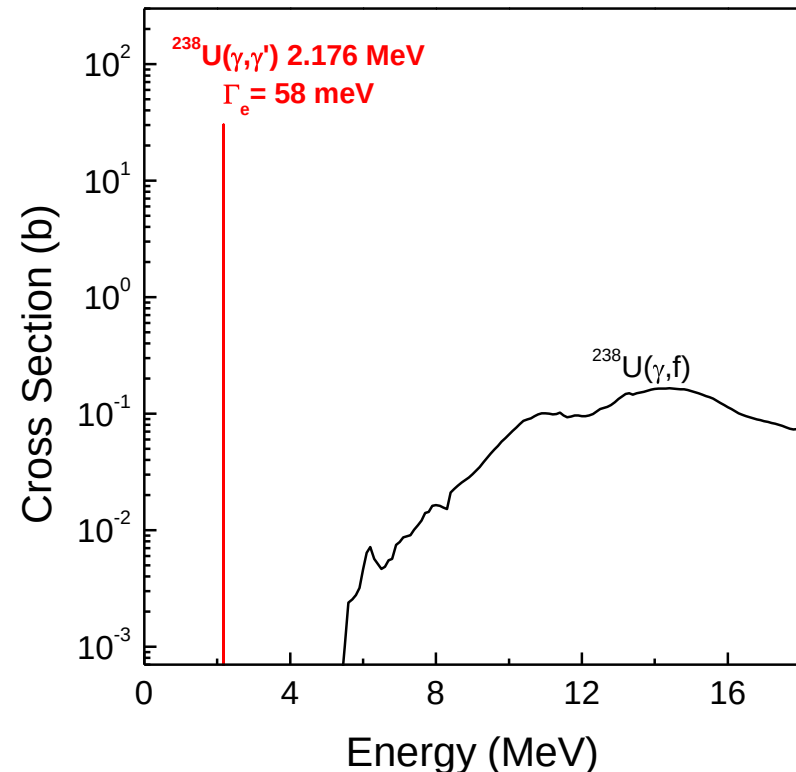
- NRF cross section is simple Lorentzian

$$\sigma_{nrf}(\gamma) = \left(\frac{c\hbar}{E_e} \right)^2 \cdot \frac{2J_e + 1}{2J_o + 1} \cdot \frac{\Gamma_{e0}^2}{(E_\gamma - E_e)^2 + \frac{1}{4}\Gamma_e^2}$$

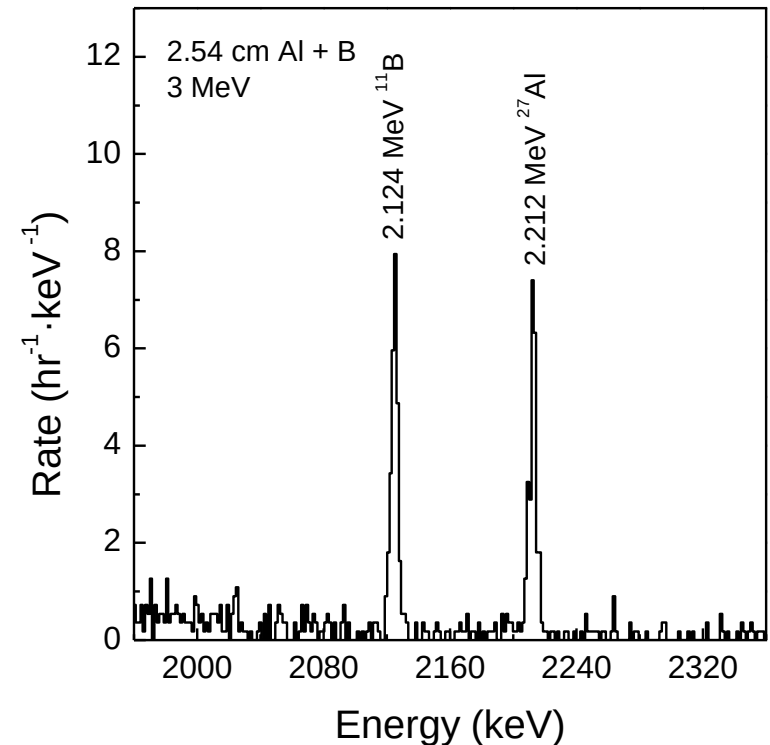
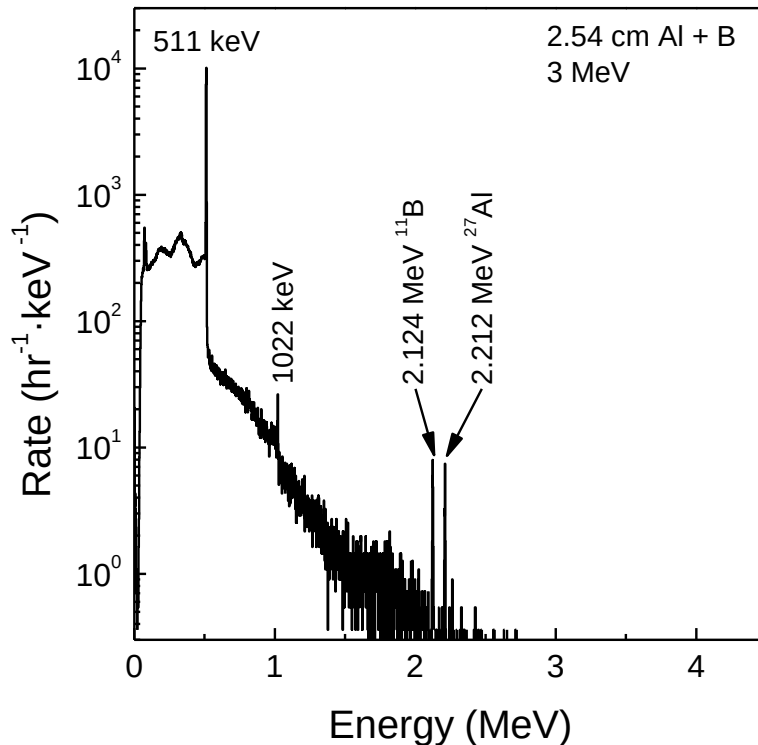
← Partial Width
← Total Width

Excitation Energy

- Large widths → big cross sections
- Typical widths μeV , meV , eV
- Thermal motion broadening important
- Typical peak cross section
 - ^{208}Pb : 263 b, ^{238}U : 30 b, ^{11}B : 6.0 b
- Broad band probe source (i.e. brems)
 - Flux constant across width
 - Integrated cross section more relevant
 - ^{238}U : 58 b·eV, ^{11}B : 51 b·eV
 - Integrated fission σ : 1.1×10^6 b·eV

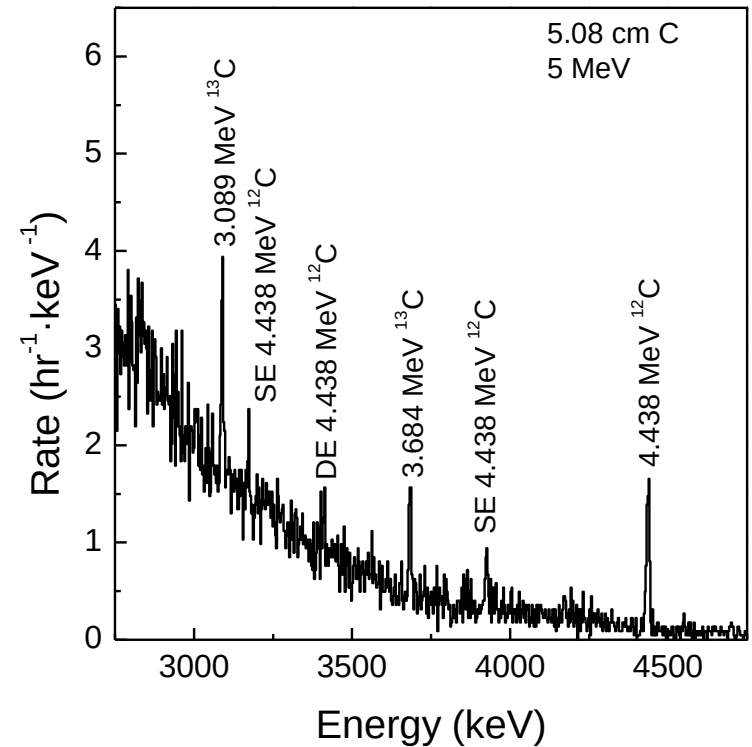
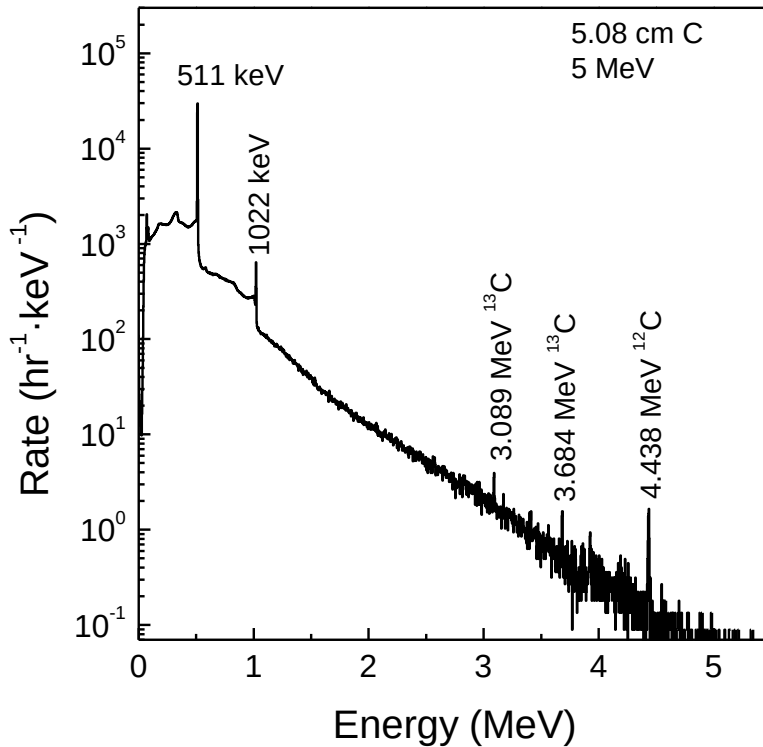


NRF Isotopic Fingerprints are Unique: B + ^{27}Al



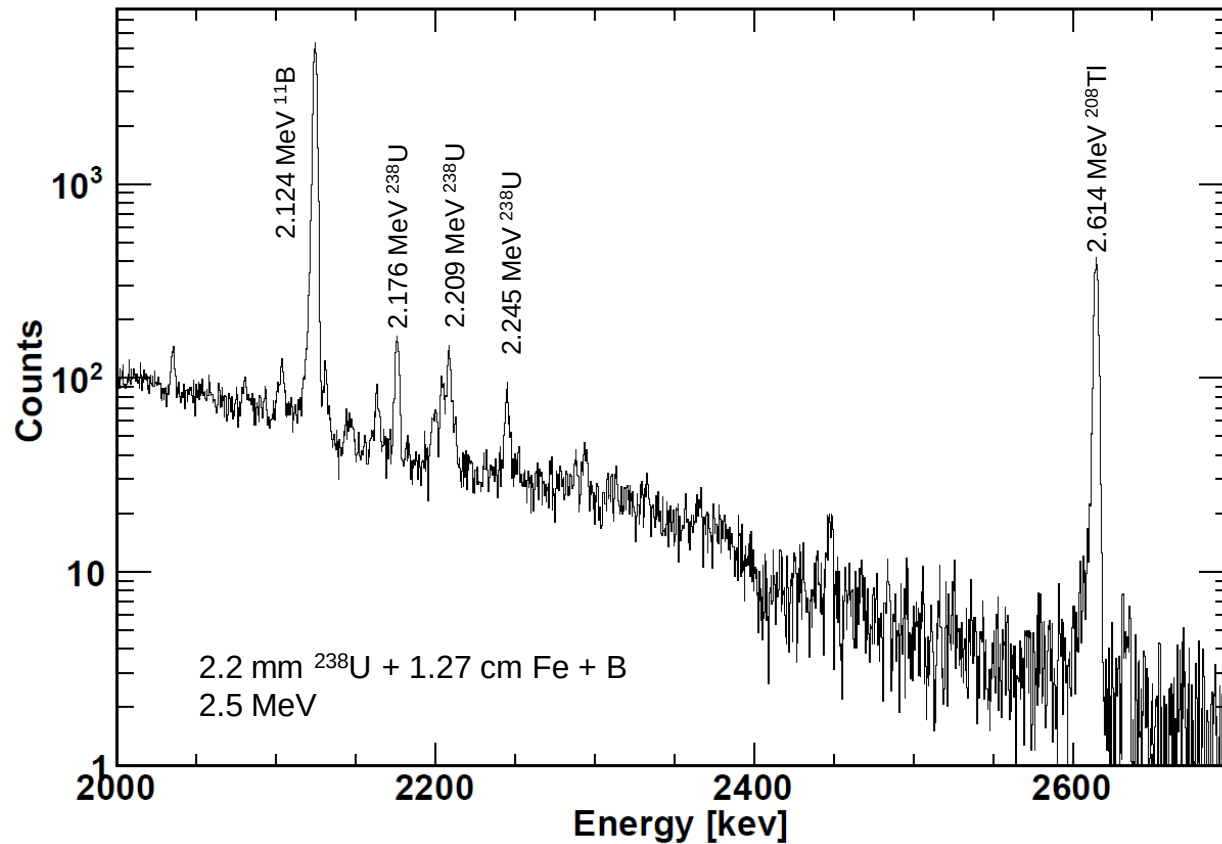
- 3 MeV bremsstrahlung
 - 3 kHz pulsed accelerator; $\sim 15 \mu\text{A } e^-$
- Low-Z targets little background
- Cross sections
 - ^{11}B (80%) 51 b·eV
 - ^{27}Al (100%) 18 b·eV

NRF Isotopic Fingerprints are Unique: C



- 5 MeV bremsstrahlung
 - 3 kHz pulsed accelerator; $\sim 8 \mu\text{A } e^-$
- Higher-energy + thick target $\rightarrow$ background
- Cross sections
 - ^{12}C (98.9%) 10 b·eV
 - ^{13}C (1.1%) 235 b·eV

NRF Isotopic Fingerprints are Unique: ^{238}U + Fe + B



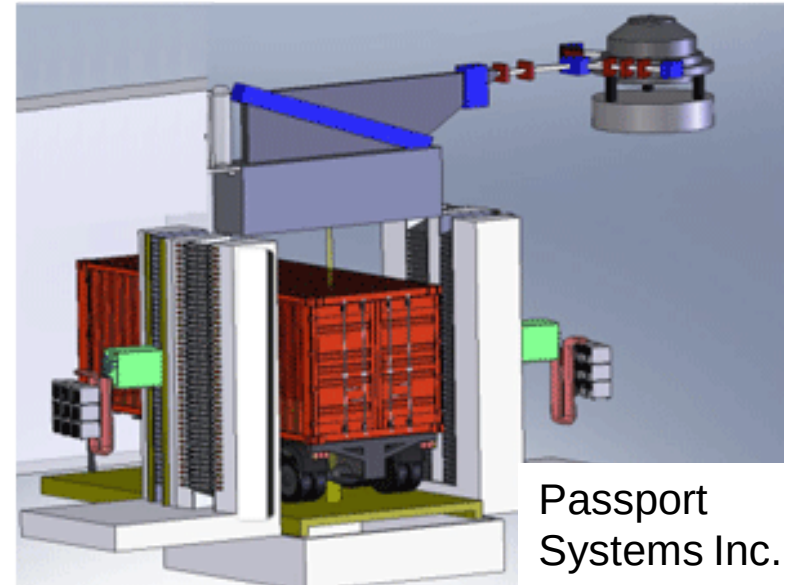
G. A. Warren et. al.

201

- 2.5 MeV bremsstrahlung
 - CW accelerator; $\sim 27 \mu\text{A } e^-$
- High-Z + shielding $\rightarrow$ background
- Cross sections
 - ^{11}B (80%) 51 b·eV
 - ^{238}U (99.8%) 58 b·eV

Isotope Identification Key to Potential Applications

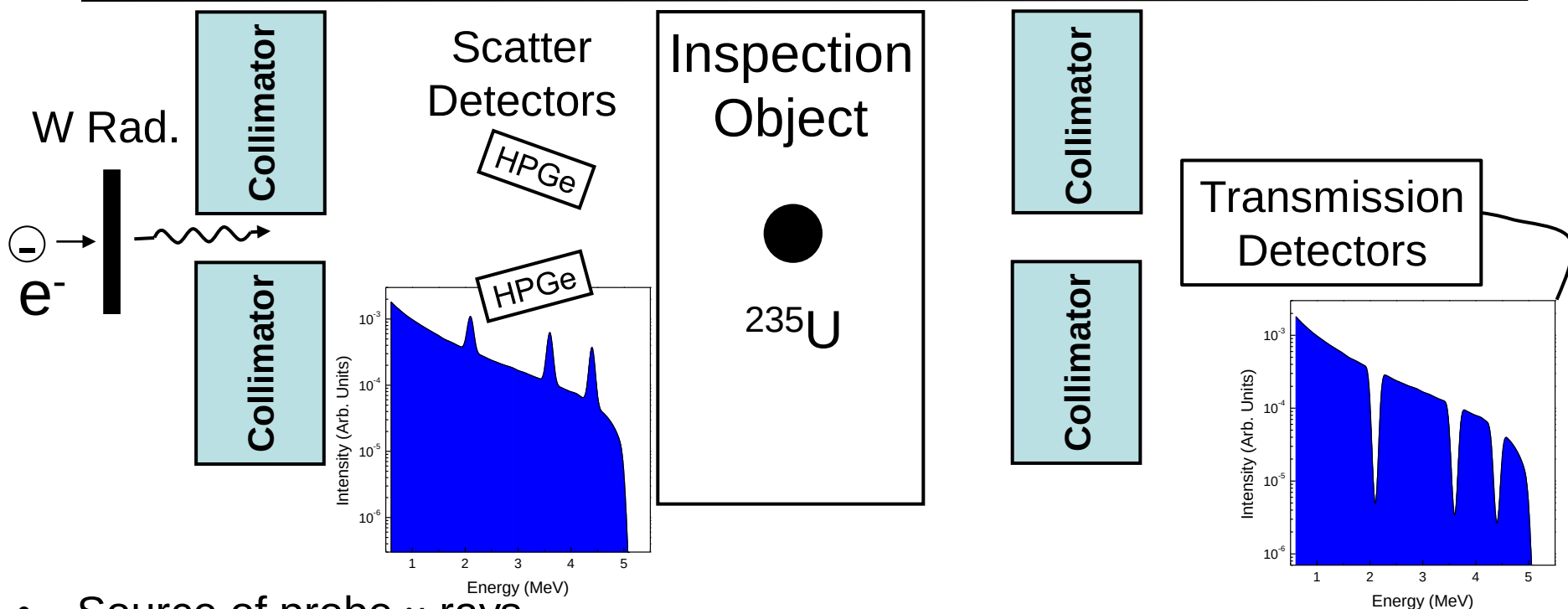
- **Safeguards**
 - Nuclear energy
 - Specificity, accuracy, speed
- **Nuclear Forensics**
 - Military, law enforcement
 - Specificity, speed, accuracy
- **Waste Assaying**
 - Nuclear energy
 - Sensitivity, specificity, accuracy
- **Cargo Screening**
 - Security, customs
 - Sensitivity, speed, environment
- Treaty Verification
 - Nonproliferation, Diplomacy
 - Sensitivity, specificity
- Long Standoff
 - Military
 - Sensitivity, speed, environment



Nondestructive Key!

- **S**ensitivity – Minimal detectable mass
- **A**ccuracy – Mass determination
- **S**peed – How fast is inspection
- **S**pecificity – Isotope identification
- **E**nvironment – How much dose

Experiment or Inspection Setup Straightforward



- Source of probe γ -rays
 - Bremsstrahlung: Look for many NRF lines, majority of beam “unused”
 - Monoenergetic (e.g. LCS): Majority of beam “used”, observe a single NRF line
- Collimation and Shielding (probe beam and detectors)
- Detectors
 - Scatter: Observe transition
 - Transmission: Measure absorption of probe beam (tuned to specific NRF lines)

Messy when Schematic Meets Experimental Reality

Collimator

90% HPGe
Detector

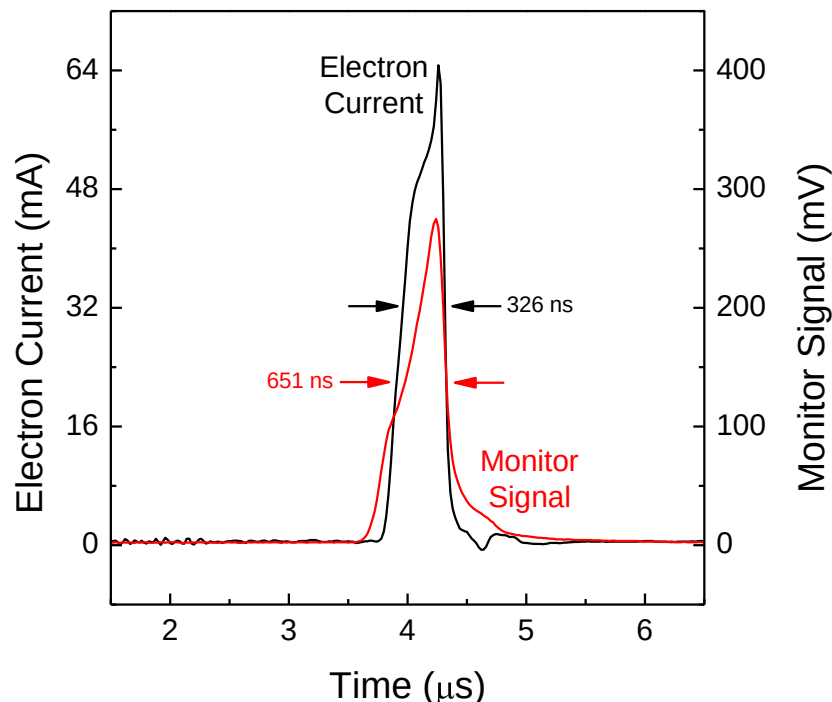
^{238}U
Target



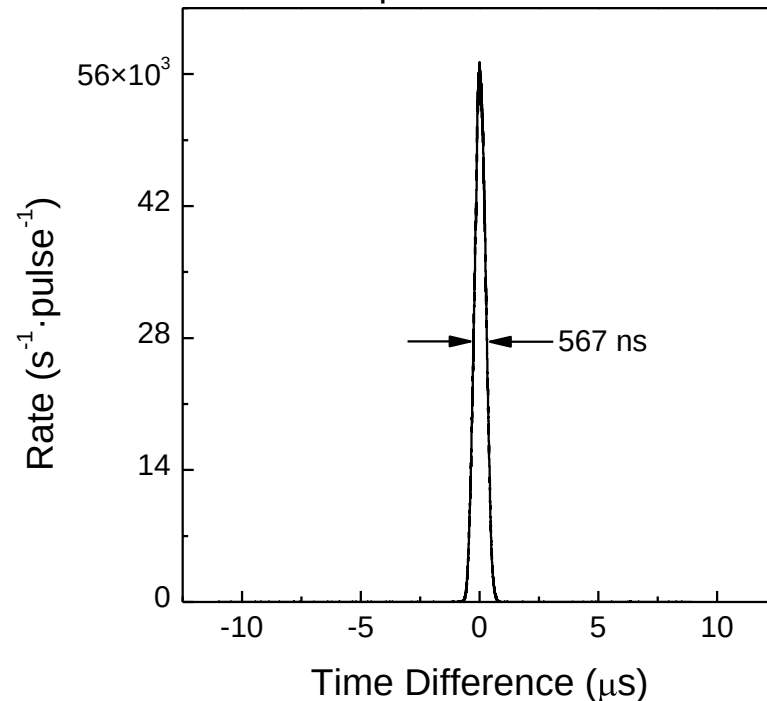
Beam
Monitor

Pulsed Linacs are “Off” Most of the Time

Electron and Beam Monitors



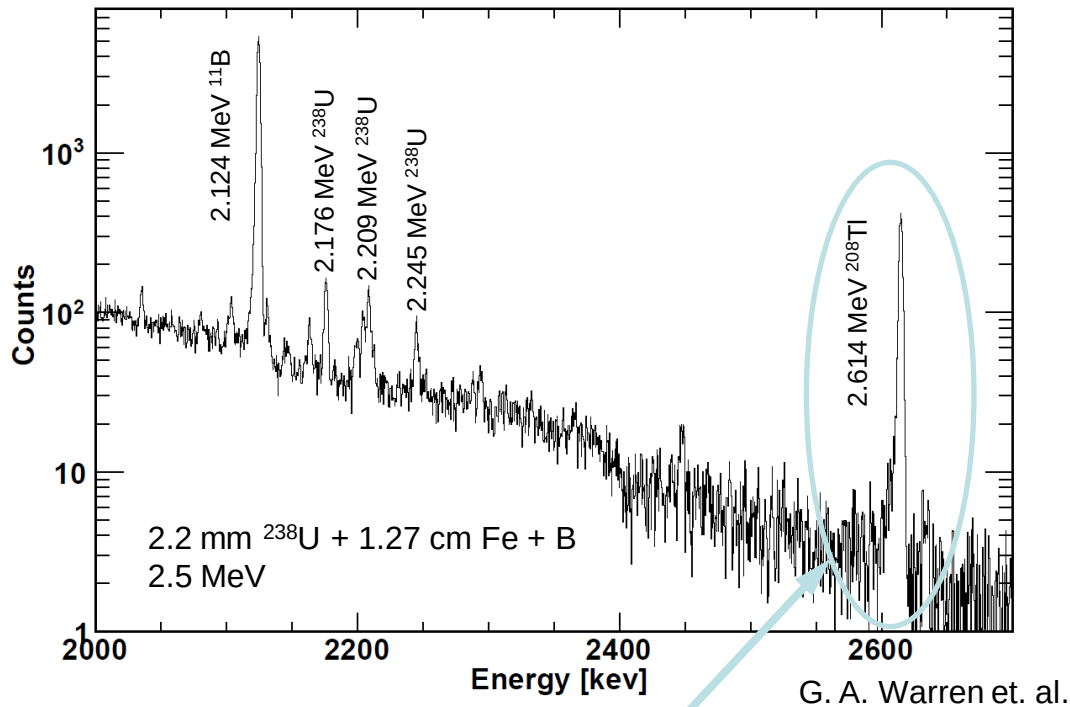
Coincidence Spectrum HPGe vs. Brem.



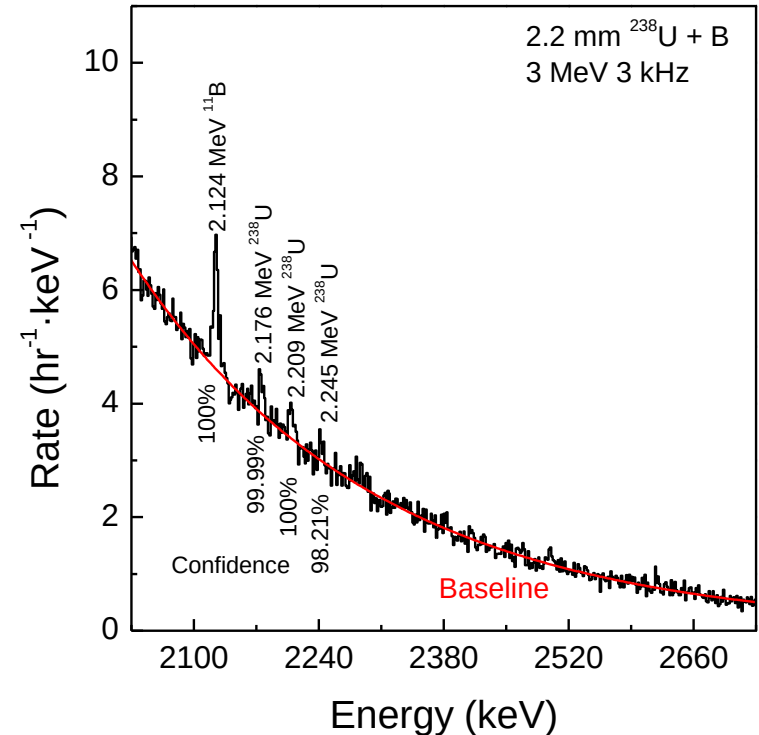
- Repetition rates are low
 - Typical linac (i.e. medical): 300 Hz
 - Enhanced medical: 1 to 3 kHz
- IAC linac “on” 0.059% to 0.195% of time
- Must avoid pulse pileup in HPGe
 - Detector rate ~ 100 to 300 Hz
- CW Accelerator
 - Detector rate ~ 15 kHz
 - Can this be pushed higher?
- Do not use pulsed linacs?

Low Rep. Rate → Statistically Starved Spectra

CW Accelerator NRF Spectrum



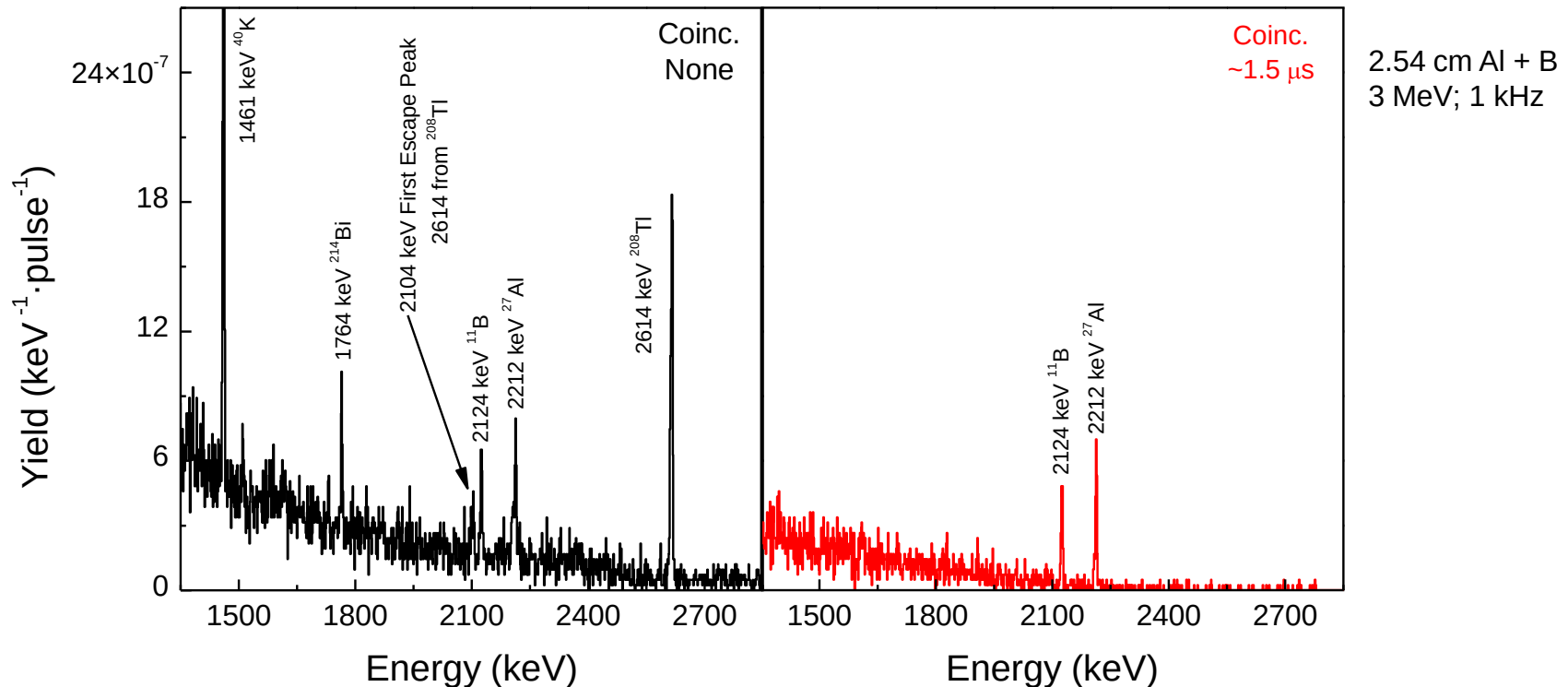
Pulsed Linac NRF Spectrum



Passive Background

- CW Experiment
 - Shielded ^{238}U plate
 - 27 μA e^- current
 - Detector rate ~ 15 kHz
- Pulsed Experiment
 - Bare ^{238}U plate (same as in CW)
 - 14 μA e^- current
 - Detector rate ~ 300 Hz

Pulsed Brems Eliminates Natural Background

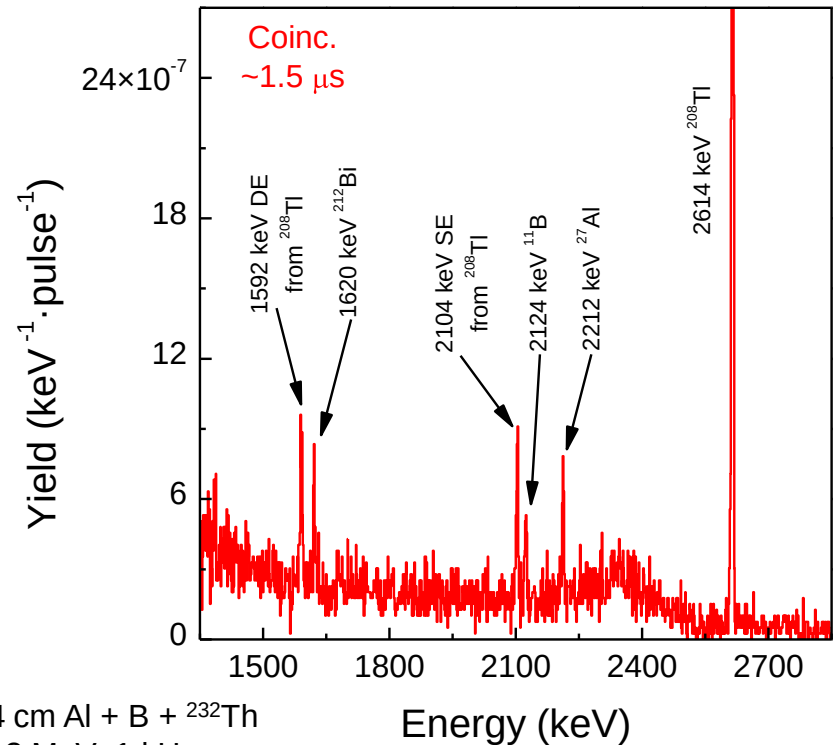
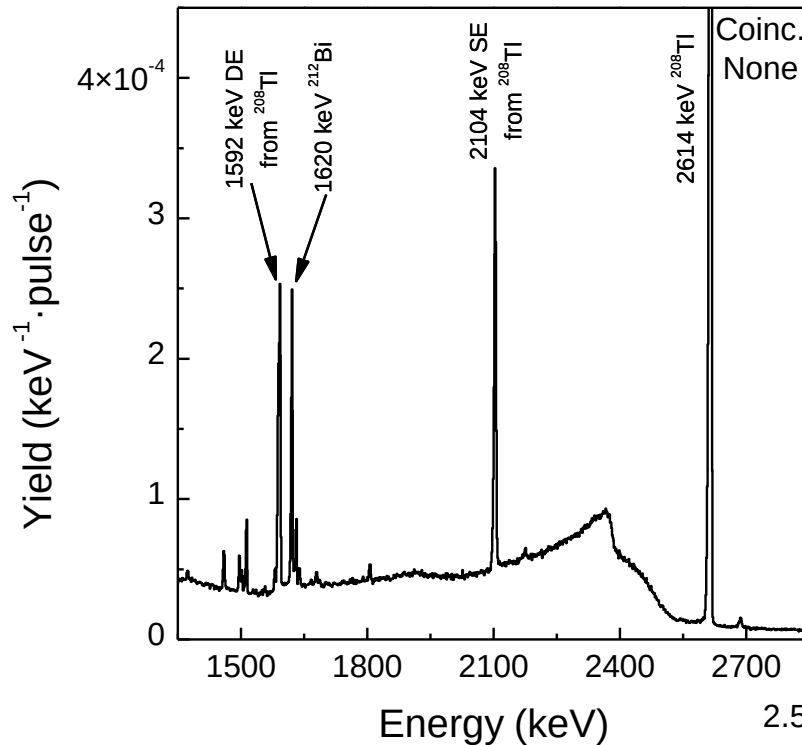


- Without coincidence requirement
 - Passive background: $\sim 10^{-5}$ pulse⁻¹
 - NRF yield: $(5.2 \pm 0.6) \times 10^{-6}$ pulse⁻¹
- 1.5 μs coincidence requirement
 - Passive background: $\sim 10^{-8}$ pulse⁻¹
 - NRF yield: $(3.4 \pm 0.4) \times 10^{-6}$ pulse⁻¹

Important to Minimize Passive Background

- Low mass or shielded materials
 - Natural background is significant
 - Nuclear forensics
 - Waste assaying
 - Cargo screening
- High passive backgrounds
 - Passive background obscures NRF transitions
 - Safeguards
 - Assaying of spent fuel for ^{239}Pu (UCB + LBL currently investigating)
 - Reprocessing monitor
 - Waste assaying
 - Fissile content assay for waste classification
- Minimized background → Maximized sensitivity

Coinc. to Minimize Large Passive Background



Additional ^{232}Th increased 2614 keV rate by 10^3

- Without coincidence requirement
 - Passive: $\sim 10^{-2} \text{ pulse}^{-1}$
 - NRF yield: Unmeasurable
- 1.5 μs coincidence requirement
 - Passive: $\sim 10^{-5} \text{ pulse}^{-1}$
 - NRF yield: $(2.9 \pm 0.6) \times 10^{-6} \text{ pulse}^{-1}$

Conclusions

- NRF provides isotopic fingerprint
 - Applications requiring isotope specificity
- NRF cross sections are large and narrow
 - Broad energy probe beam: Identify many isotopes; Majority of beam “unused”
 - Monoenergetic probe beam: Identify single isotope; Majority of beam “used”
- Advantages of pulsed bremsstrahlung
 - Minimize passive background by 10^3
 - Maximize sensitivity
- Issues with pulsed bremsstrahlung sources
 - Low repetition rates → small NRF rates
 - Need high rate accelerator
 - Enhanced pulse pile up
 - Need deconvolution algorithms (exploit time information)

Path Forward for Pulsed Bremsstrahlung

- Current CW experiments
 - 15 kHz detector rate
- Desired pulsed accelerator
 - Repetition rate: 150 kHz
 - Charge in pulse: Match 15 kHz detector rate
 - Pulse width: ~ 10 ns take full advantage of coincidence timing
- Capabilities of pulsed NRF system
 - NRF intensities matching current CW experiments
 - $\sim 10^3$ passive background reduction
- Higher detector rate?
 - Increase repetition rate $\rightarrow$ decrease passive background reduction
 - Increase hit fraction $\rightarrow$ increased pulse pileup