

High purity germanium (HPGe) detector R&D for applications from Particle Physics to Nuclear Safeguards

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Pacific Northwest National Laboratory
U. Nagoya Colloquium



Introduction

- ▶ PNNL Overview
- ▶ Three HPGe Detector System Examples
 - MARS (Multi-sensor Airborne Radiation System)
 - High efficiency, rugged, thermally efficient
 - RN LABS (Radionuclide Laboratory System)
 - High efficiency, ultra-low background
 - UHRGe (Ultra-high rate Germanium)
 - High resolution gamma spectroscopy at $>1\text{Mcps}$
- ▶ Summary



PNNL Quick Facts

- ▶ DOE Office of Science laboratory
- ▶ ~4,700 staff
- ▶ Business volume ~\$1B/year
- ▶ Key facilities
 - Environmental & Molecular Science Laboratory, a Department of Energy national scientific user facility
 - Sequim Marine Sciences Laboratory
 - Applied Process Engineering Laboratory—a research, development, and demonstration user facility and technology business incubator
 - Radiochemical Processing Laboratory for nuclear science and engineering
 - Biological Sciences Facility and Computational Sciences Facility
 - Physical Sciences Facility – 5 building complex including underground lab



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Proudly Operated by **Battelle** *Since 1965*

PNNL's presence in Washington



**Pacific Northwest Center for
Global Security**



Marine Research Operations
Sequim, Washington

Richland campus



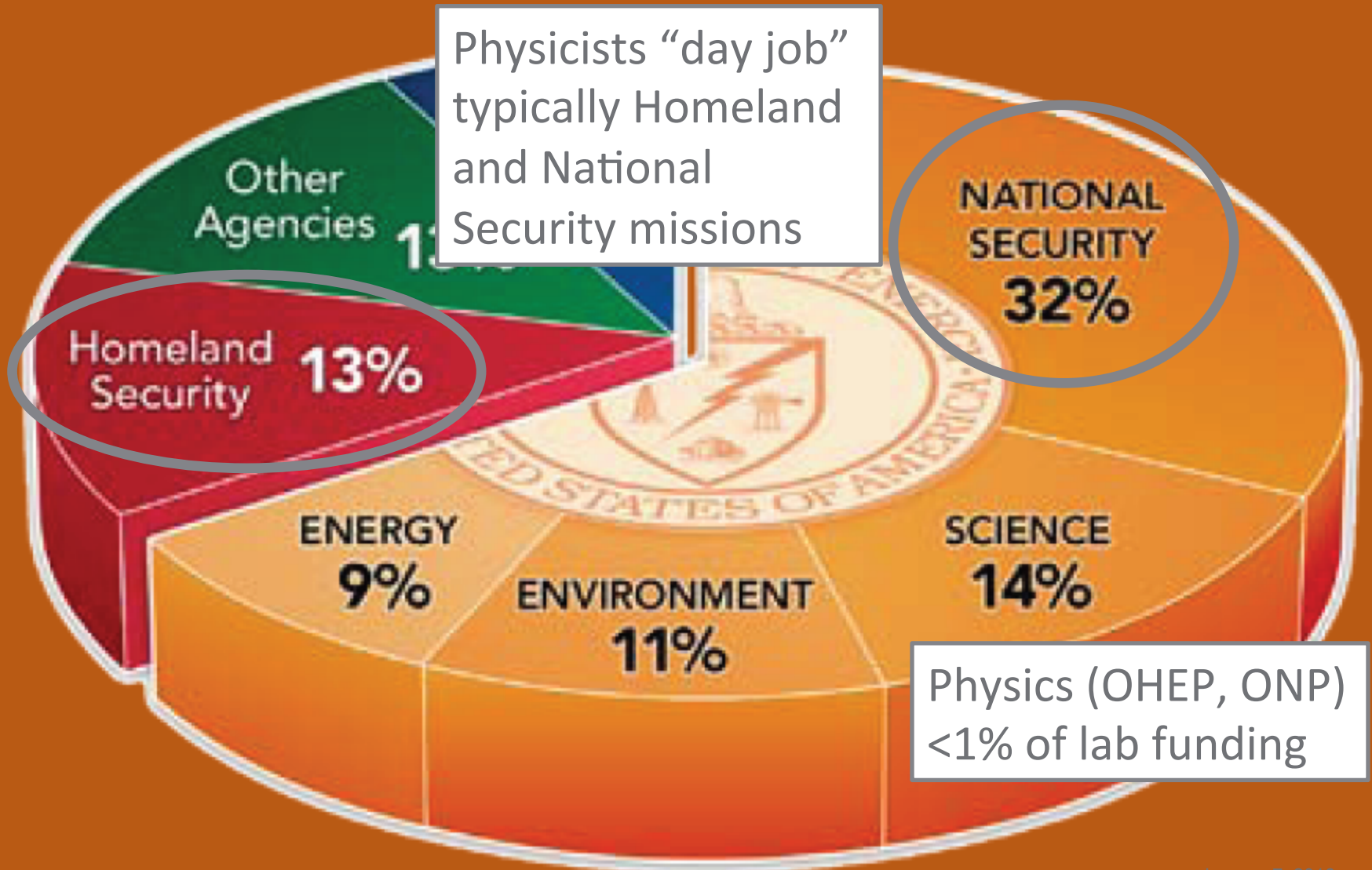
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PNNL Business Areas



Physicists “day job” typically Homeland and National Security missions

NATIONAL SECURITY
32%

Homeland Security
13%

ENERGY
9%

ENVIRONMENT
11%

SCIENCE
14%

Physics (OHEP, ONP)
<1% of lab funding

Why the focus on High Purity Germanium (HPGe) detectors at PNNL?



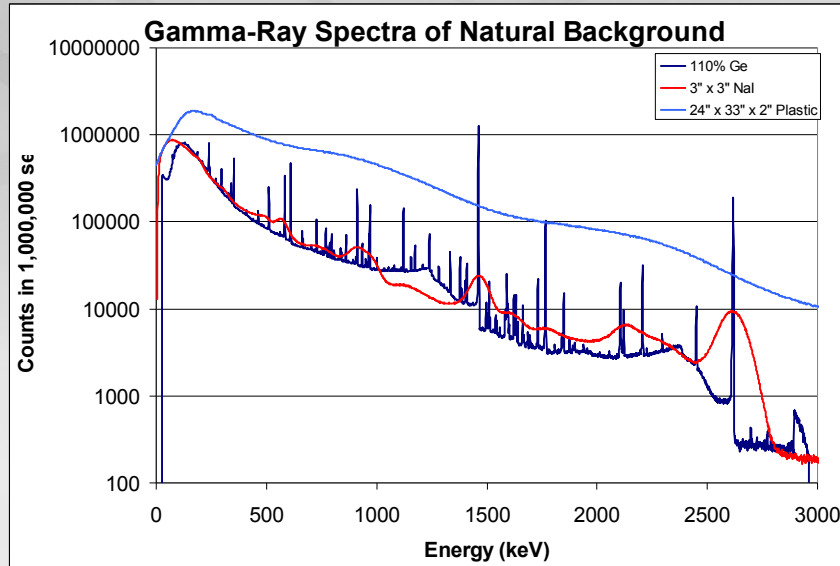
PNNLs HPGe Legacy

- ▶ Unique HPGe detectors developed from the 1960's for gamma assay
- ▶ Pioneers in low-background counting for environmental radioisotopes
 - Fate and transport of isotopes in the environment
 - Understanding radiation on the moon (assay of moon rocks)
 - Treaty Verification
 - Nuclear Forensics
- ▶ Dark Matter searches dating back >25 years
 - Strong synergy with security and environmental science technologies
- ▶ Applications outside the laboratory
 - Airborne systems for emergency response and other applications
 - High rate systems for nuclear safeguards

HPGe is the “gold standard” for gamma-ray spectroscopy



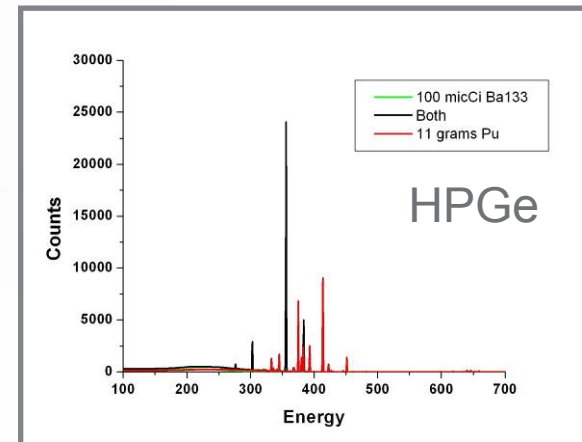
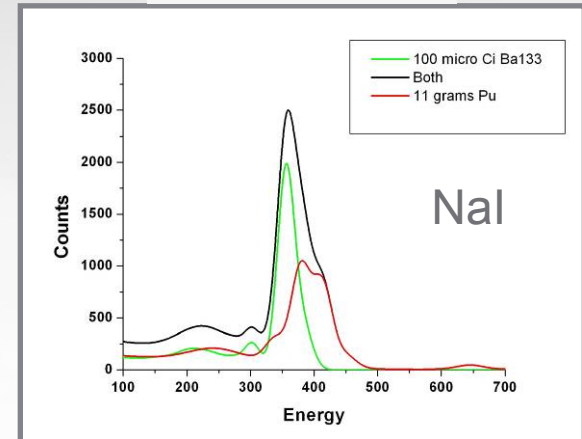
Resolution and Why it Matters



Comparison of Different Resolution Gamma Ray Spectrometers:

- PVT (used in most portal monitors)
 - NaI (used in most backpacks)
- HPGe (used in gamma spectroscopy labs)

Simulation



Resolution = Isotopic Identification
Isotopic Identification = Actionable Information

Multi-sensor Airborne Radiation System

Work sponsored by DOE/NNSA

Office of Defense Nuclear Nonproliferation R&D (DNN R&D)

MARS Development Cycles

What is MARS?

A high resolution gamma ray spectrometer large enough for stand-off applications and ruggedized for use on a variety of mobile platforms

▶ MARS R&D

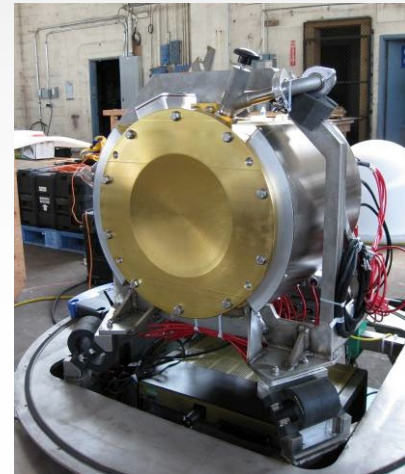
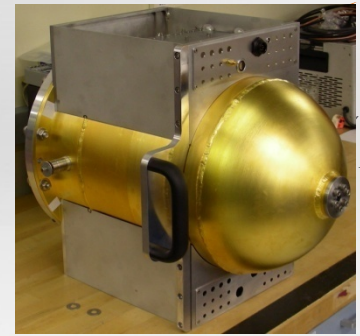
- Multi-sensor Airborne Radiation Survey R&D
- DOE DNN R&D project FY2006-2008
- Initial technology research and development

▶ MARS Flight

- Multi-Sensor Airborne Radiological System Flight
- DOE DNN R&D project FY2008-2011
- Development of technology into rugged field system

▶ Demonstrations

- Demonstration in a maritime environment in FY10-11
- Demonstrated for aerial search/survey in FY12



MARS R&D Project: Advancing the State of the Art

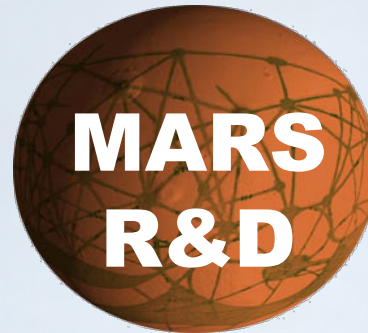


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Where we were: System of commercial detectors

- Too large; not integrated
- Limited mission time (4 hrs)
- Heavy system: ~700 kg
- One-off, fixed configuration
- Limited real-time analysis



Where we arrived: Well integrated monolithic array

- Integrated system-in-a-pod
- Increased mission time (2+ days)
- HPGe module ~25 kg (10kg HPGe) enables much lighter system
- Modular system-of-systems with multi-platform capability
- Real-time actionable data analysis

~1987 ERSS System



Instrument racks with sensor pod against

Filling the gap requires:

- Improved thermal, mechanical, and electronic engineering
- Applying lessons from previous PNNL HPGe projects



2010 MARS System

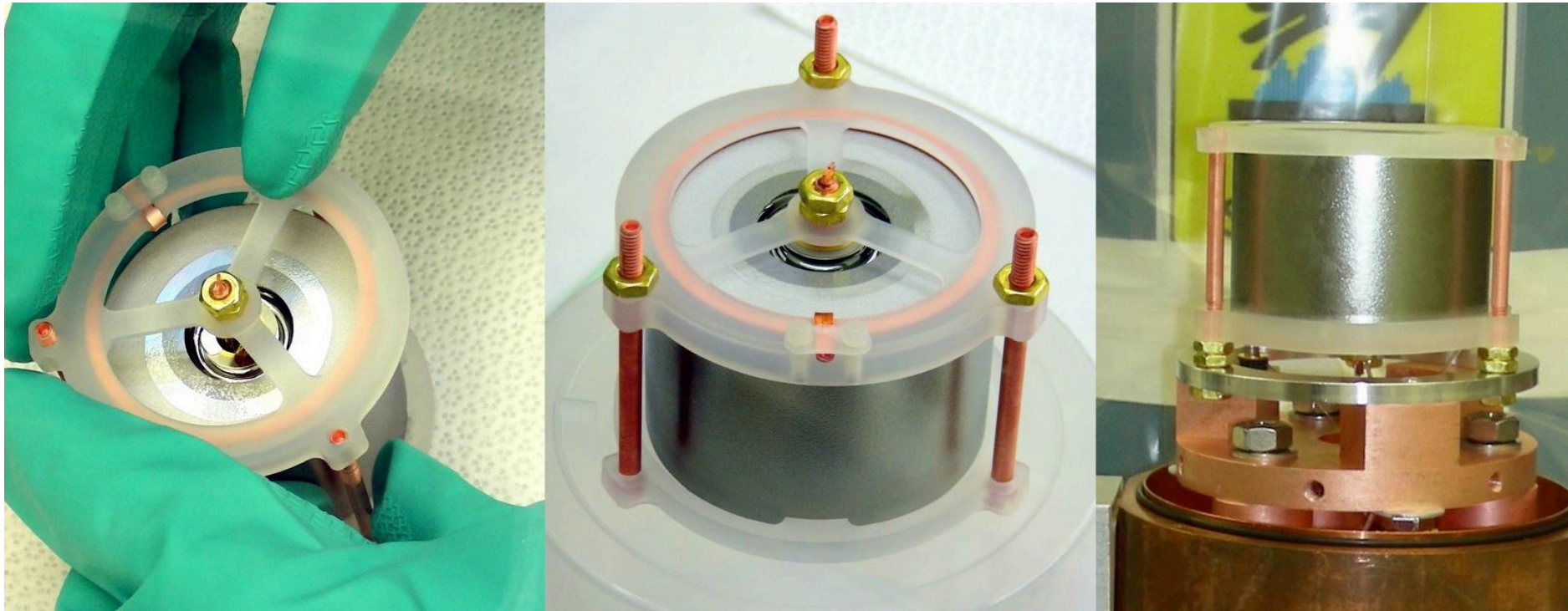




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HPGe Detectors and Detector Mounting

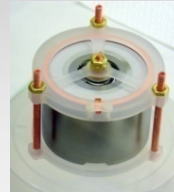


- ▶ PNNL designs start from bare HPGe diodes
- ▶ Control materials for low background applications
- ▶ Control electro-mechanical design for low microphonics

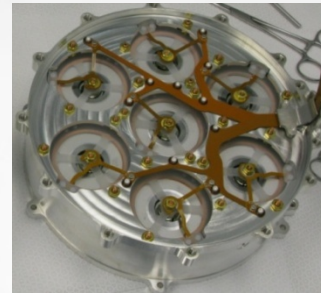


MARS Flight Detector System

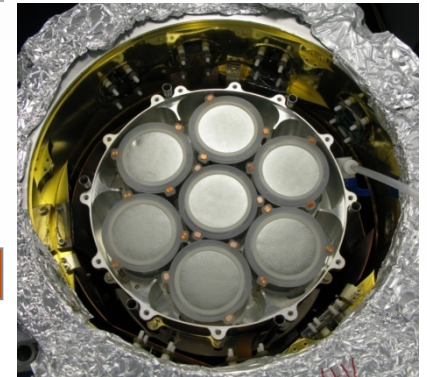
- ▶ *It is an array of 14 high resolution gamma ray spectrometers, each larger than that used in commercial handheld instruments, for high quality, actionable, isotopic identification in the field*
- ▶ Close-packed array improves detection efficiency by 30-50%
- ▶ **This is the first successful close-packed array HPGe system ever built**



HPGe detector

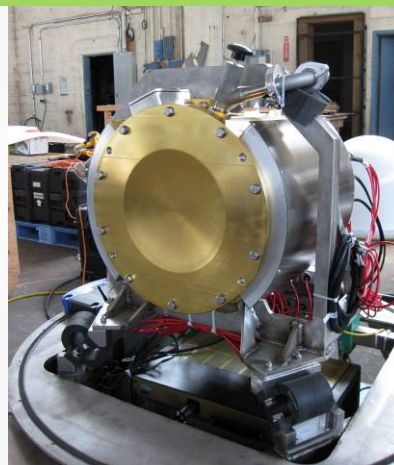


1 of 2 spools, each with 7-detectors



detectors in cryostat

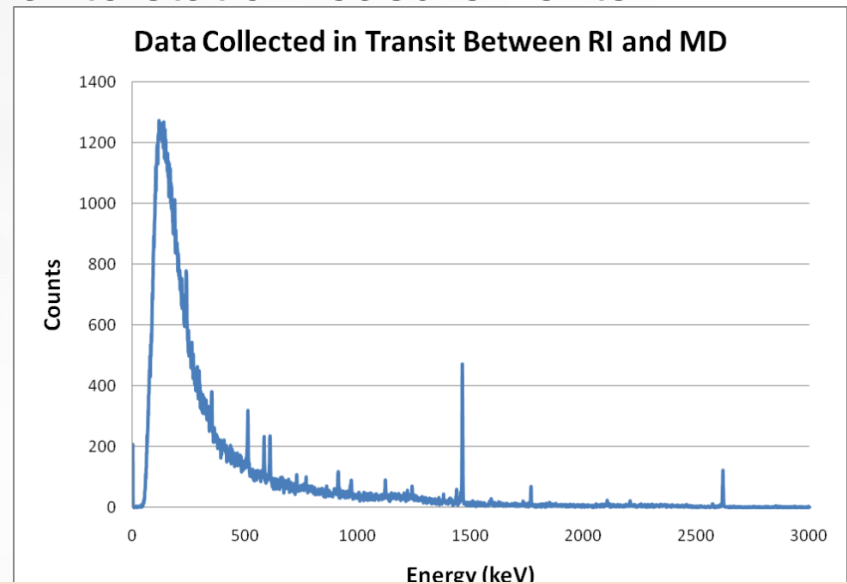
Cryostat mounted



System in enclosure

Cross country journey to field demonstration

- ▶ MARS was transported from Richland, WA to Newport, RI and on to Charleston, SC in a 22 ft Penske truck to participate in Maritime demonstration activities
- ▶ During transit the system was operated off of a large UPS
- ▶ Data were collected in several states at ~60-65 mph
 - Detector performance was similar to static measurements



Dominant spectral features include the 1460 keV line from K-40. Features from the U-238 and Th-232 chains include the 239, 352, 511, 583, 609, 911, 969, 1120, 1239, 1764 and 2614 keV lines.

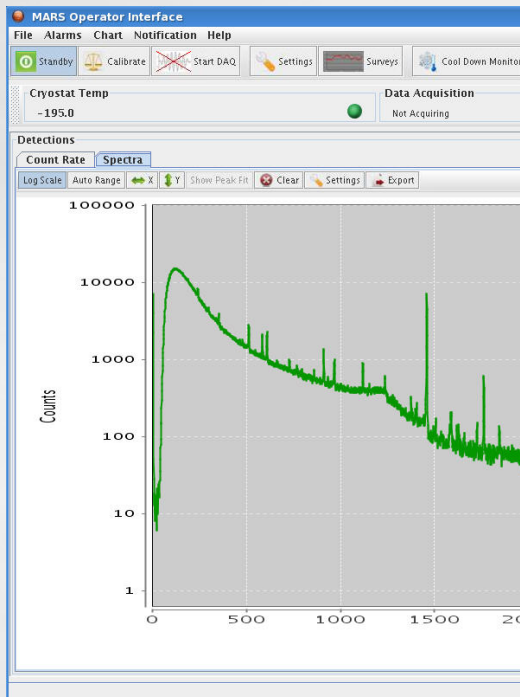
MARS Field Test data – 10 hour Background



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- ▶ ~7 MeV dynamic range
- ▶ ~3 keV energy resolution
- ▶ Rich spectrum from U/Th decays

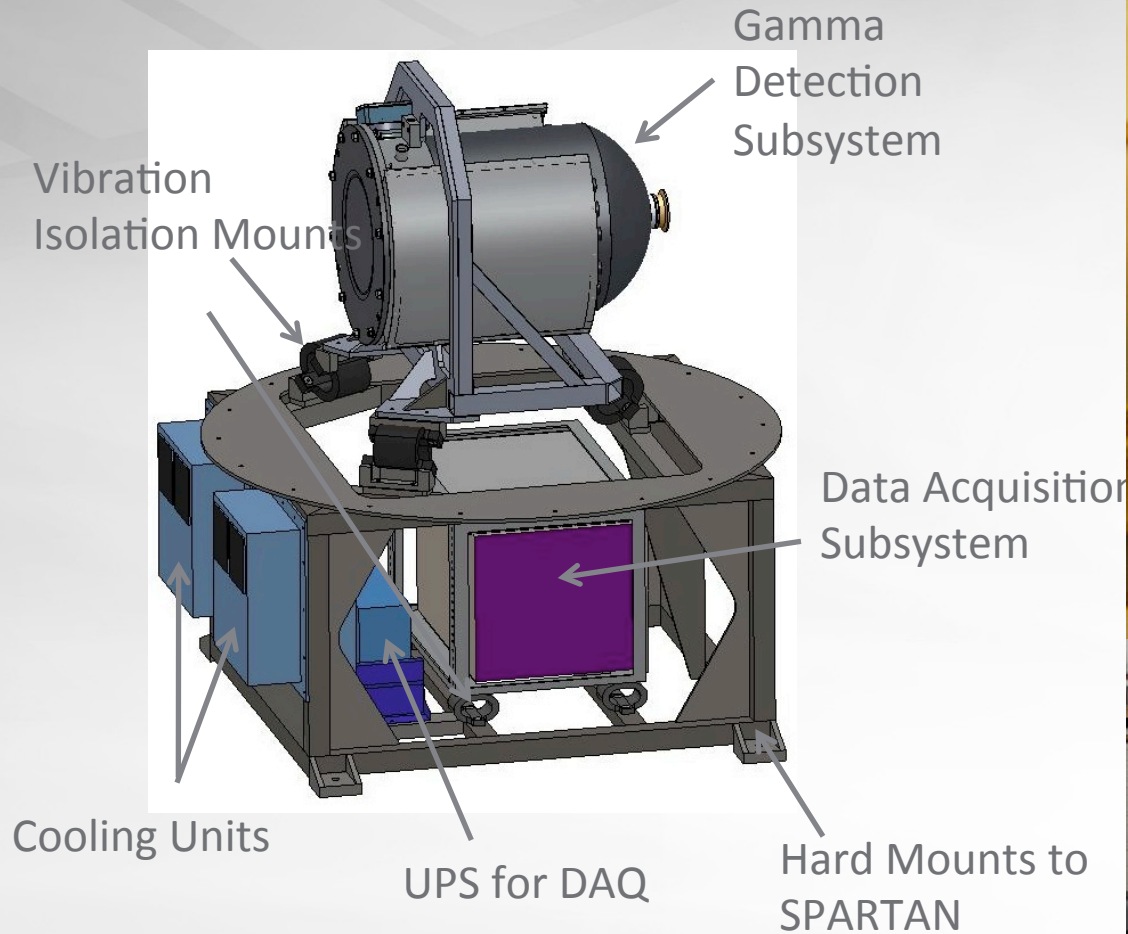


Truck mounted, static measurement



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MARS System on watercraft

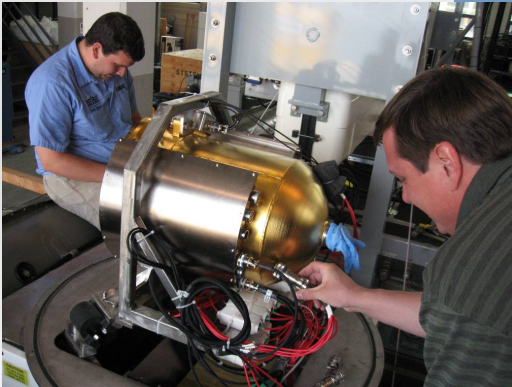




MARS Maritime Demonstration

- ▶ Successfully operated on three boats with similar system architecture

MARS w/o
Radome



MARS on
Local Platform



USV data,
communications



Deployable
Control Station

MARS Technical Demonstration with US Armed Forces and Coast Guard operators



USCG personnel and West Point cadet operating MARS



MARS deployed on USV for maritime interdiction testing



MARS Reconfigured for Flight

- MARS HPGe array modified for installation in Bell 412 helicopter
- Flew 5 test flights from June 15-20, 2012
 - Initial checkout flight
 - Altitude spiral
 - Area 3 survey
 - Point source flyovers
 - Government Wash survey
- Simultaneous data taken with RSI NaI(Tl) system (3 logs)

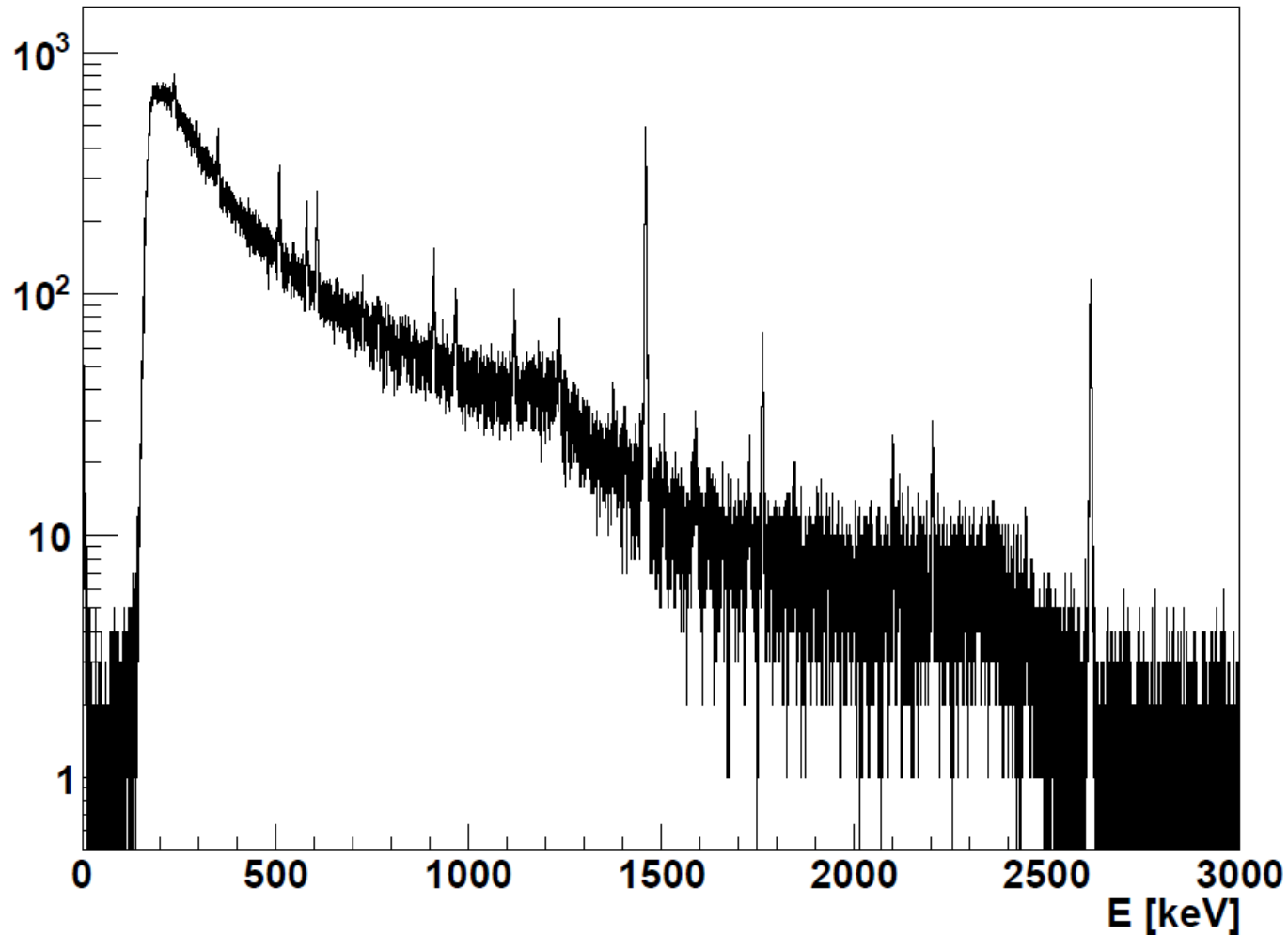
MARS as mounted in helicopter





Sample Spectrum from Test Flight

MARS 30 minute Test Flight





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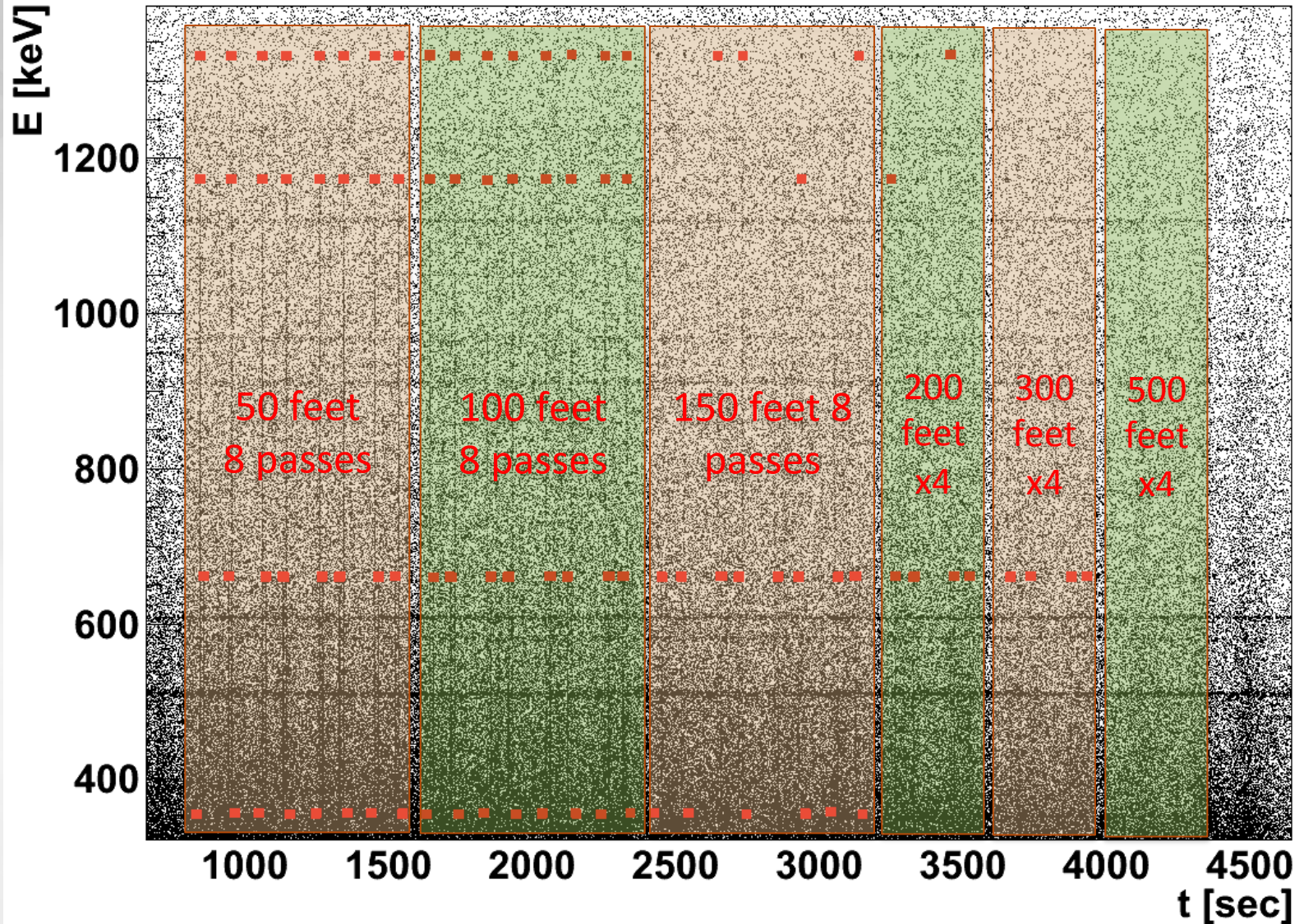
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Source Flights: Alarming $> 3.3\sigma$

Co-60 peaks

Cs-137 peak

Ba-133 peak



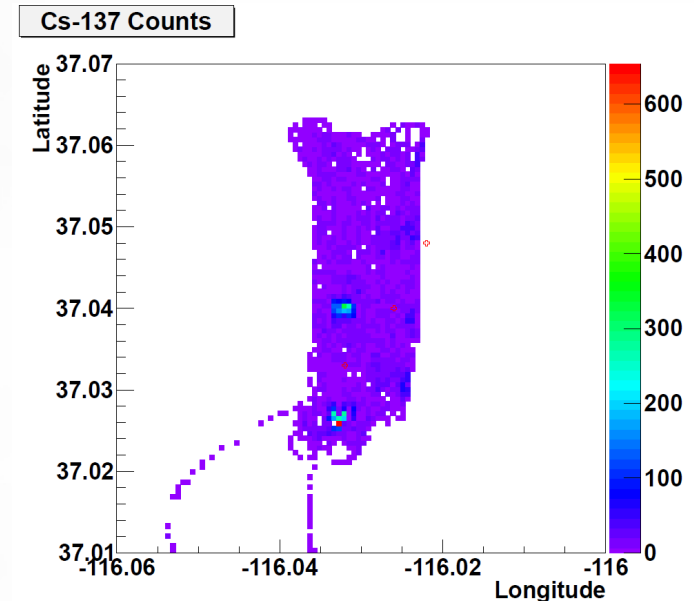


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Performance Summary

- ▶ No problems from vibration
- ▶ UPS adequate to bridge gaps in power from ground to APU to aircraft
- ▶ GPS and radar altimeter successfully integrated into data stream
- ▶ Heat was tolerated but the electronics crate did warn on over temperature
- ▶ Very small shifts in gain and resolution before and after flights appear to be due to temperature (~ 1 keV shift in gain, 0.4 keV increase in FWHM)
- ▶ Liquid nitrogen supply sufficient for two back-to-back 2.5 hour flights without need to refill (helicopter was refueled twice)
- ▶ Second-by-second monitoring with 5 second window was sufficient to alarm on sources



Radio-Nuclide Labs / CASCADES

Work sponsored by DOE/NNSA/NA-22

Ground-based Nuclear Explosion Monitoring R&D (GNEM)

PI: Martin Keillor

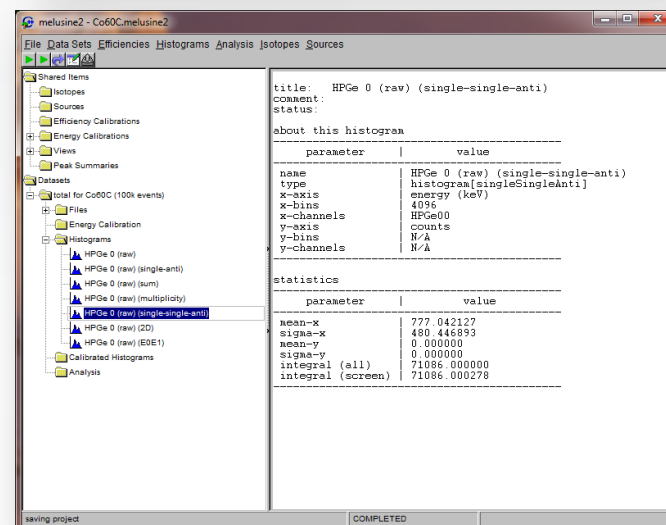
Project Goals



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- ▶ Project funded by DOE GNEM R&D
 - Ground-based nuclear explosion monitoring
- ▶ Build a high-efficiency, ultra-low-background HPGe array that can:
 - Provide high sensitivity laboratory-based capability to detect fission products
 - Produce useful results for very low activity samples
- ▶ Develop automated analysis routines
- ▶ Demonstrate system capabilities

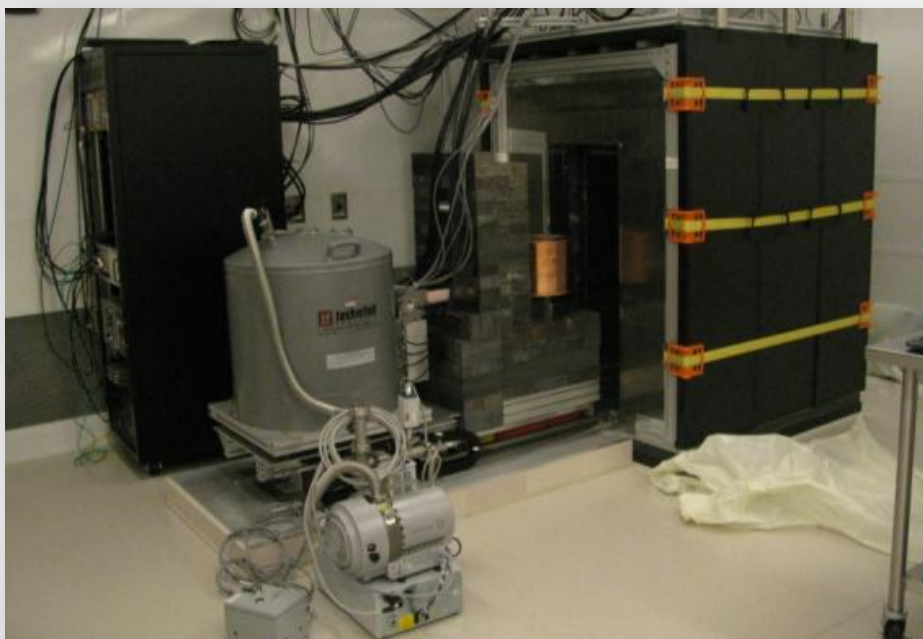


Radionuclide Laboratory System

RN Labs / CASCADES

- ▶ High Efficiency
- ▶ High Selectivity
- ▶ Ultra-Low Background

- ▶ Atmospheric filter paper assay
- ▶ Resolve anomalous results seen in field measurement or normal laboratory assay
- ▶ Low level environmental samples
- ▶ Physics measurements
- ▶ Potential application to International Monitoring System samples





The HPGe Array and Shield System

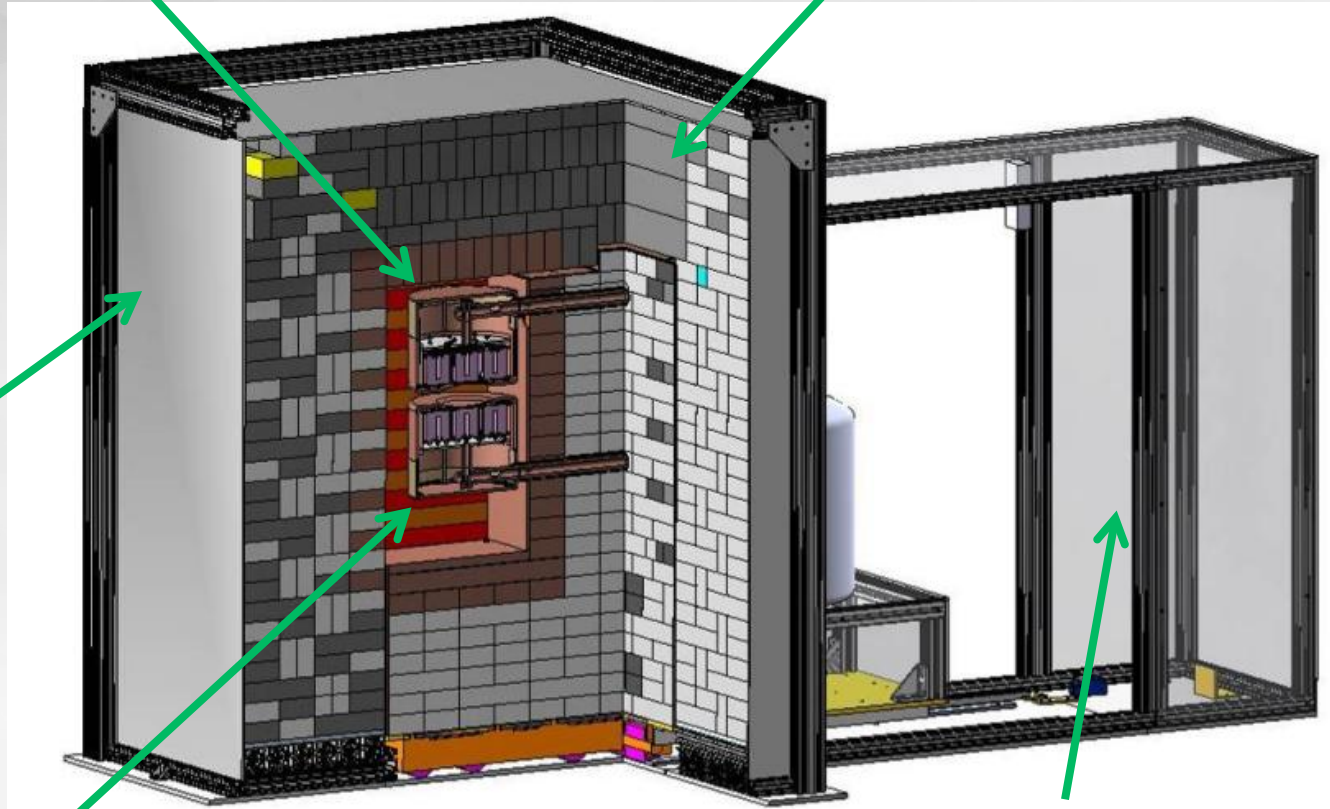
2nd cryostat in cryogenic testing
(sans crystals)

8" Pb shield

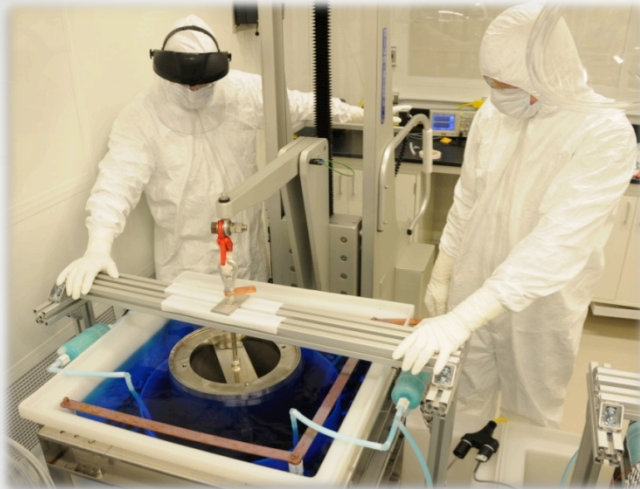
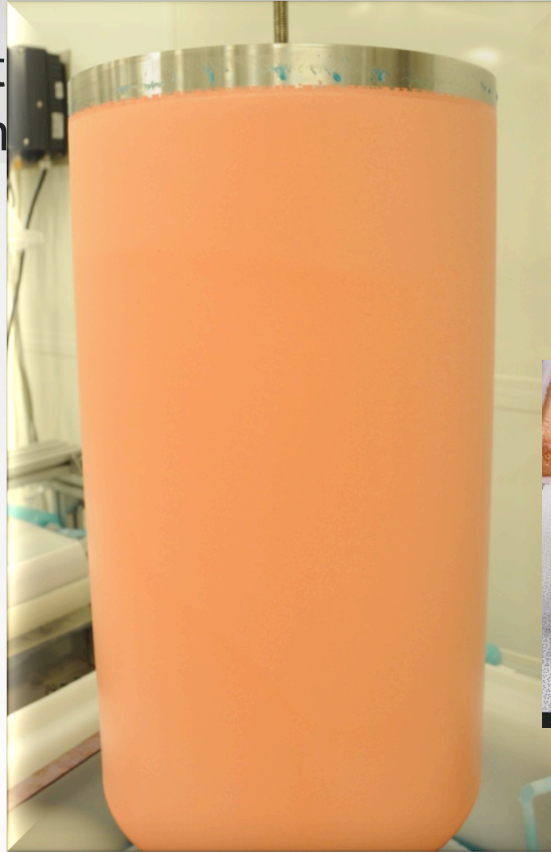
2" BC408
scintillator
active anti-
Cosmic

Operating 7 crystal array

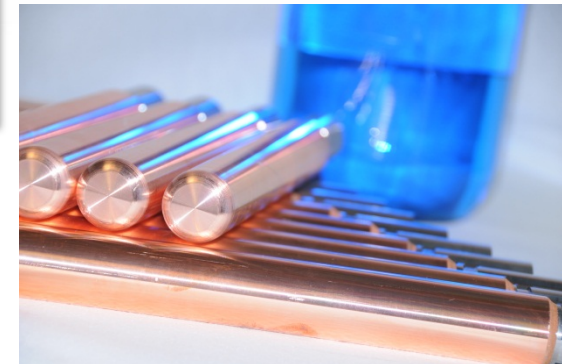
Radon exclusion



Ultra-low background detectors require special materials



Electroforming ultra-high purity copper in a variety of shapes and sizes



Ultra-low background detectors must be shielded from cosmic radiation

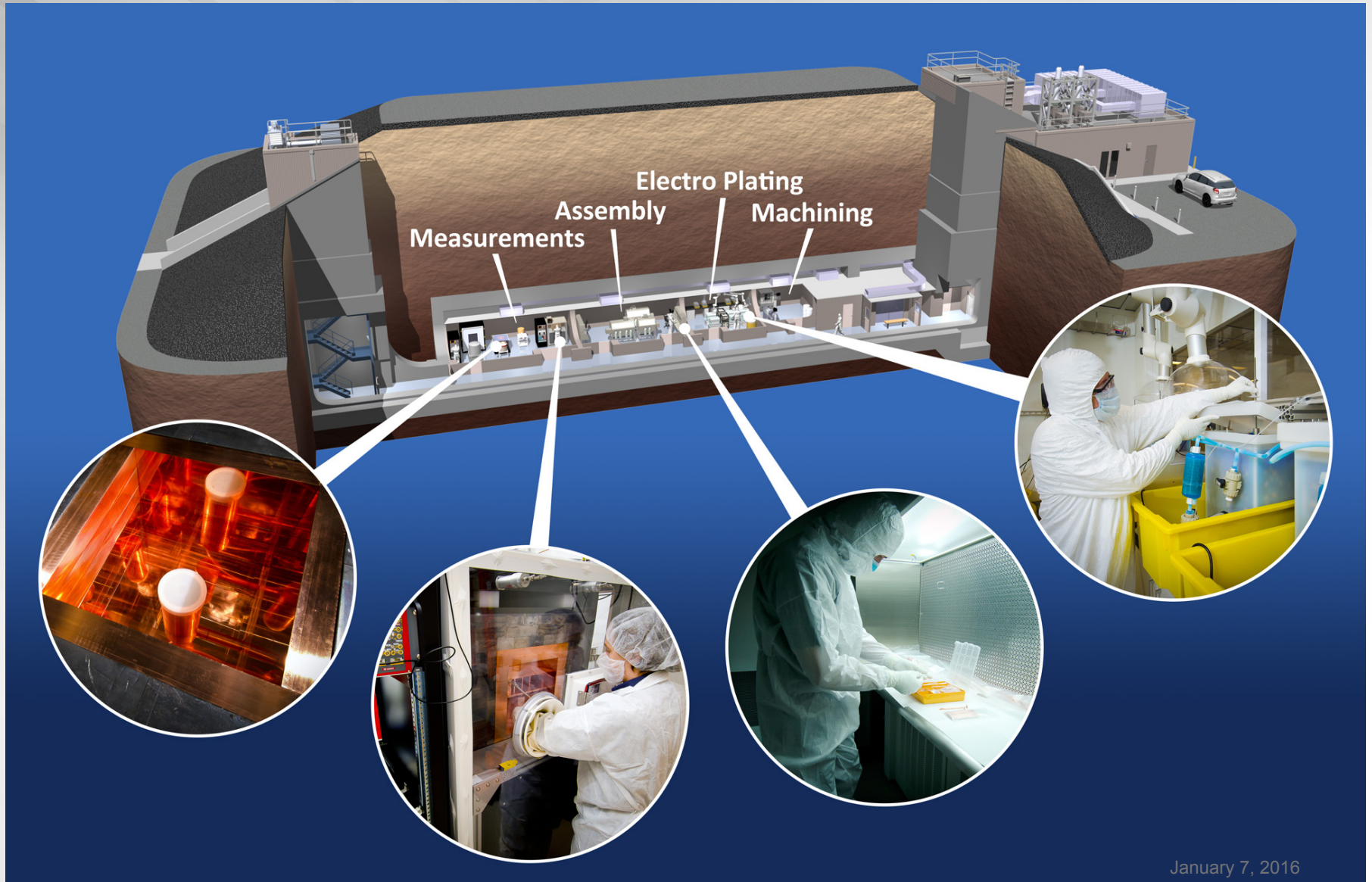
- ▶ Shallow underground lab at PNNL
- ▶ ~30 mwe depth
- ▶ ~2200 sq ft clean space



Virtual Tour of Underground Lab: <http://tour.pnnl.gov/shallow-lab.html>


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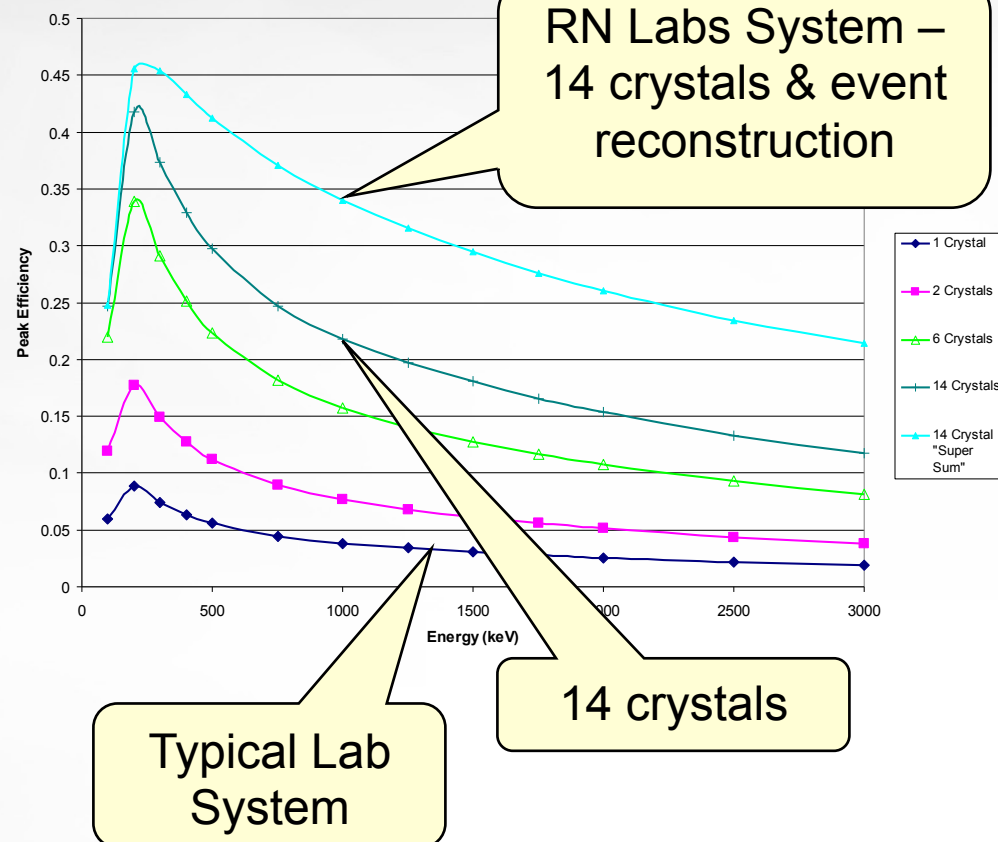
January 7, 2016



High Efficiency from Array Geometry

- ▶ Increasing efficiency provides most direct sensitivity enhancement
- ▶ Crucial for γ - γ coincidence due to product of two efficiency factors
- ▶ Significant gain by summing multiple-crystal interactions (event reconstruction)

MCNP5 Simulated HPGe Crystal Array Efficiencies (single γ)



High Selectivity from Coincidence Counting



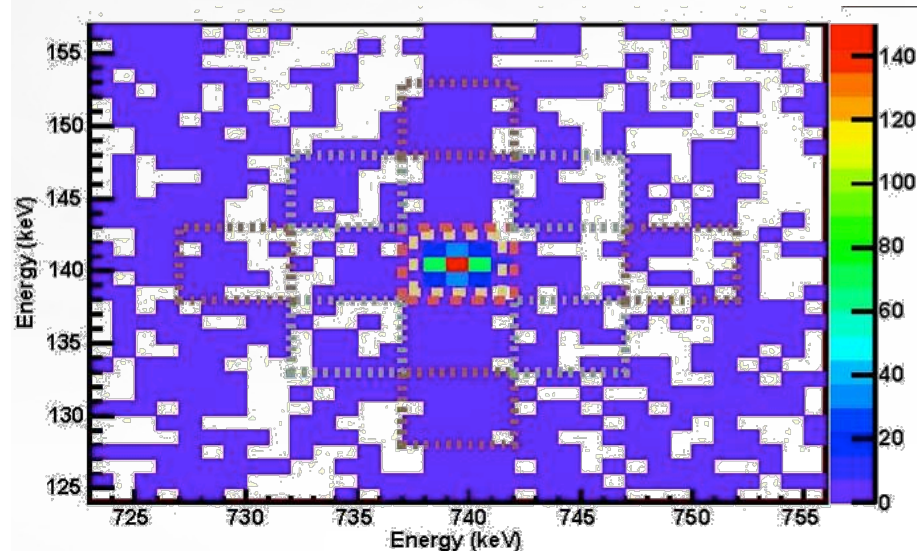
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- ▶ Energy resolution of HPGe provides excellent selectivity
- ▶ γ - γ coincidence further enhances selectivity
 - Targets unique signatures
 - Can resolve interferences found in “singles” spectroscopy
 - Provides substantial background reduction for appropriate isotopes
- ▶ Targets measurement with little or no sample prep

⁹⁹Mo Gamma-Gamma Signal Example

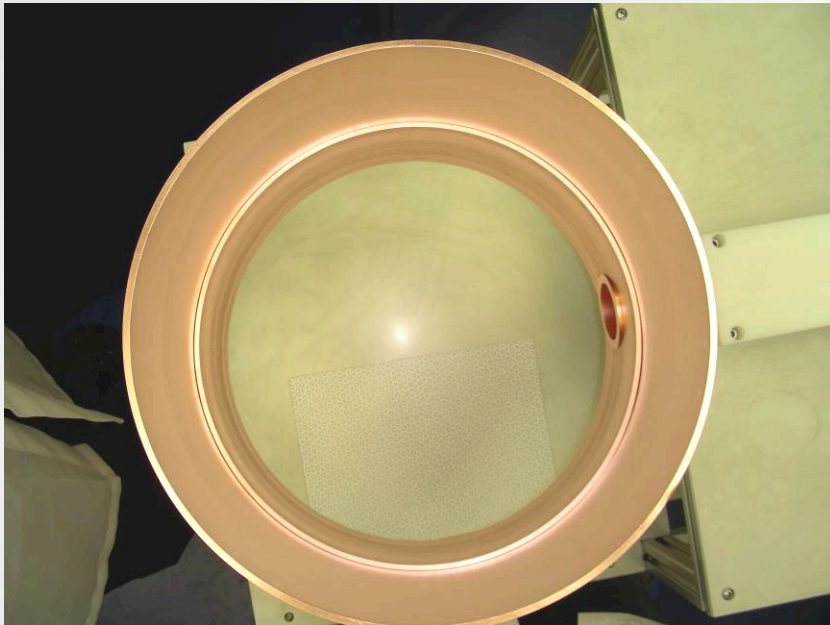
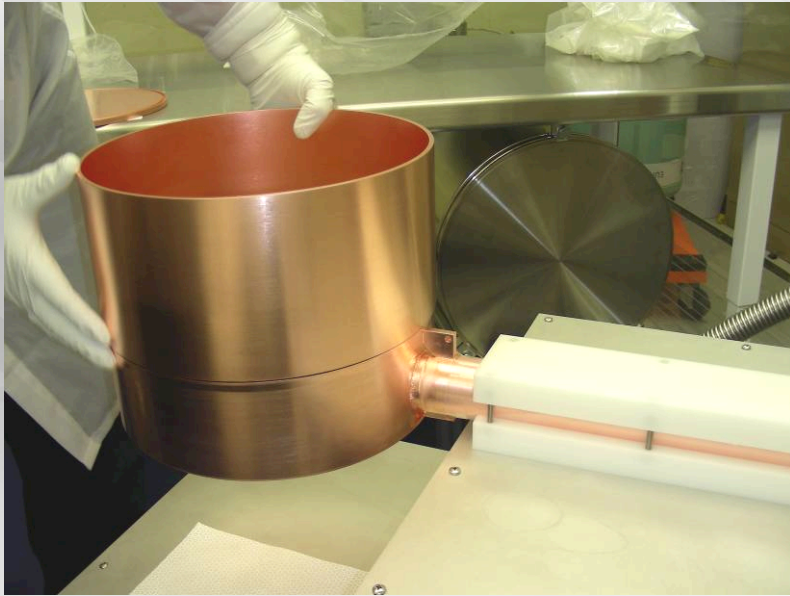
(²³⁵U irradiation, all mixed fission products,
1e8 fissions, age=5days, 24 hour count,
GEANT4 Monte Carlo data)





Electroformed Copper Cryostat Parts



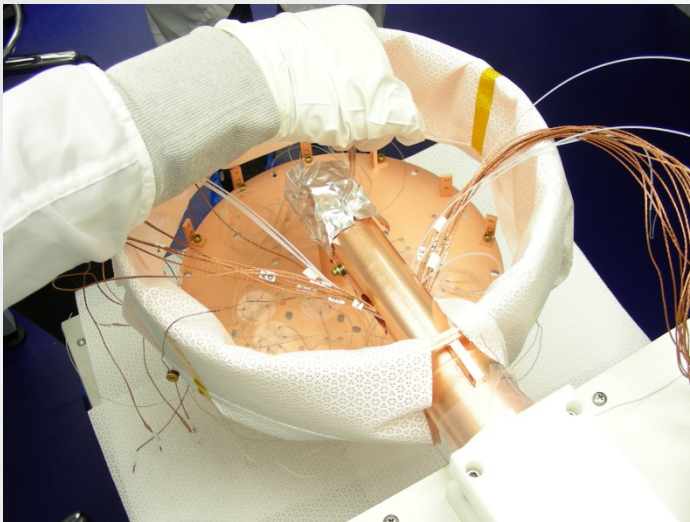
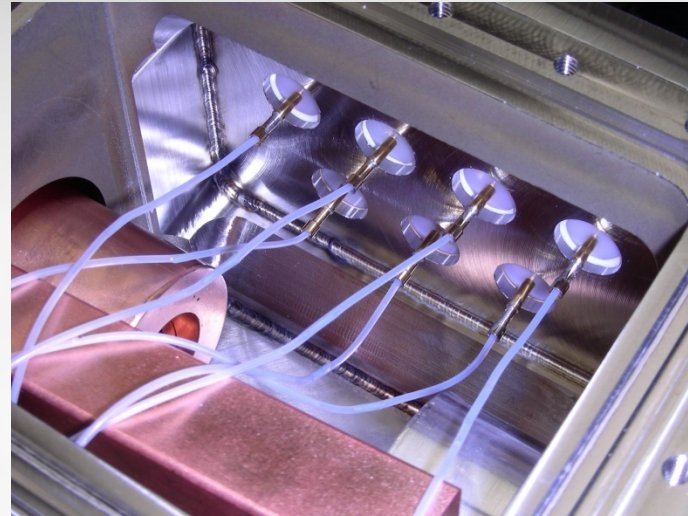
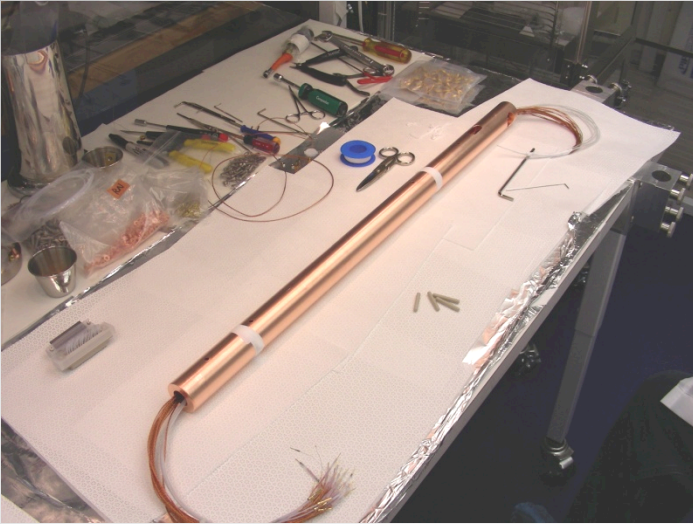




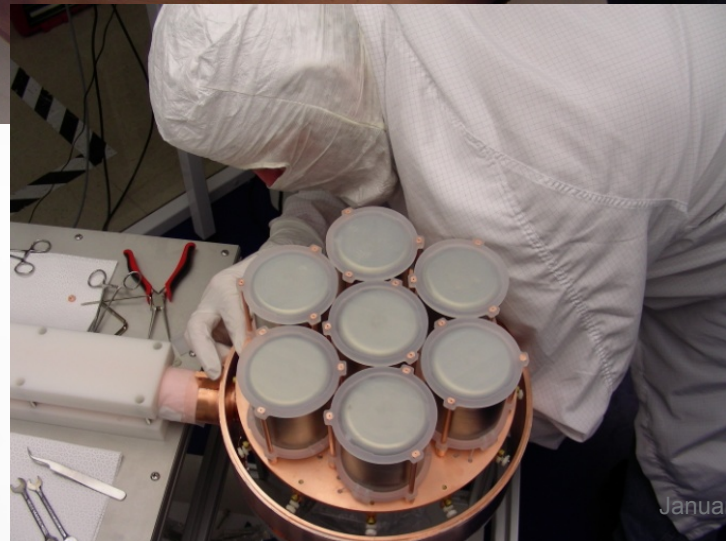
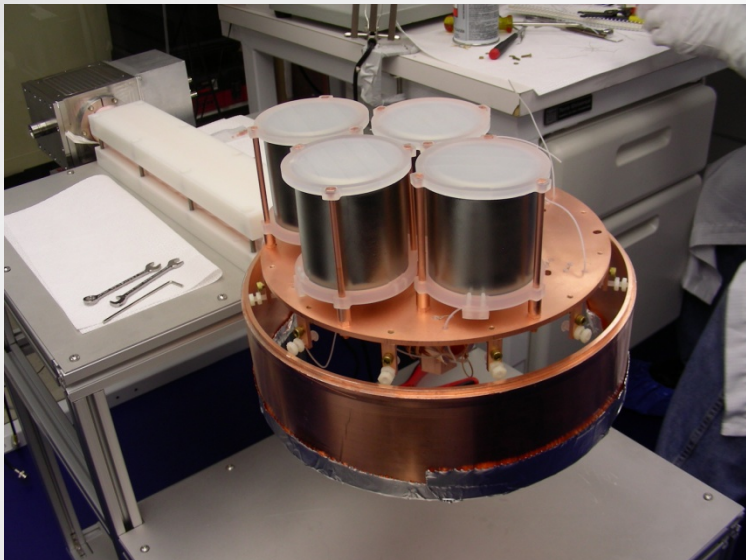
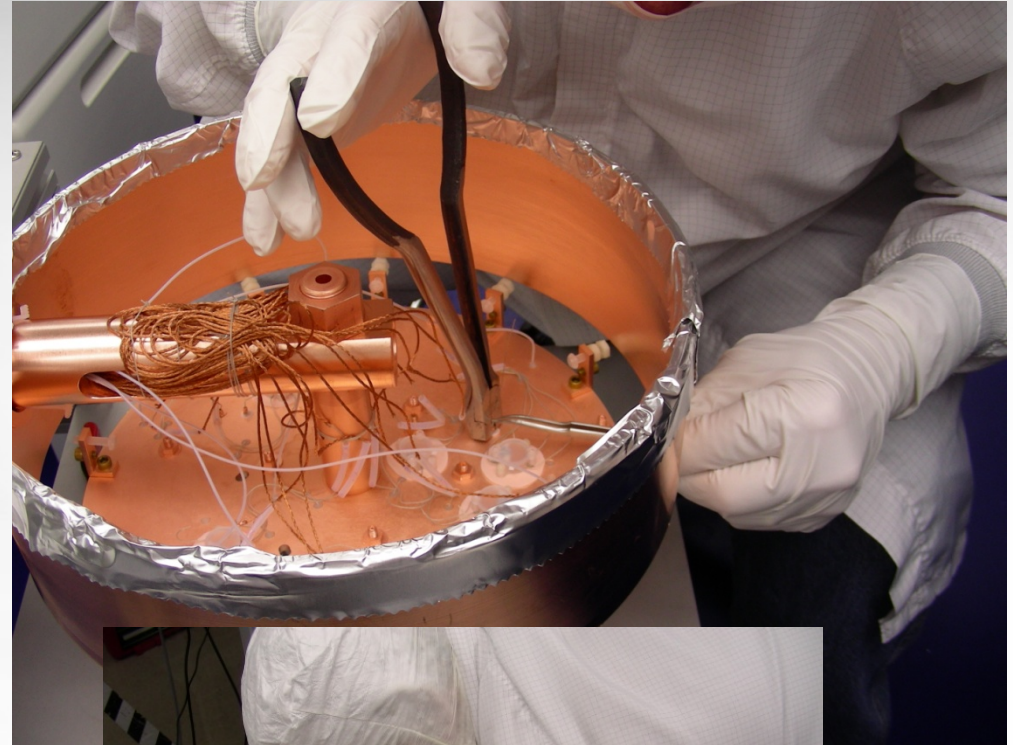
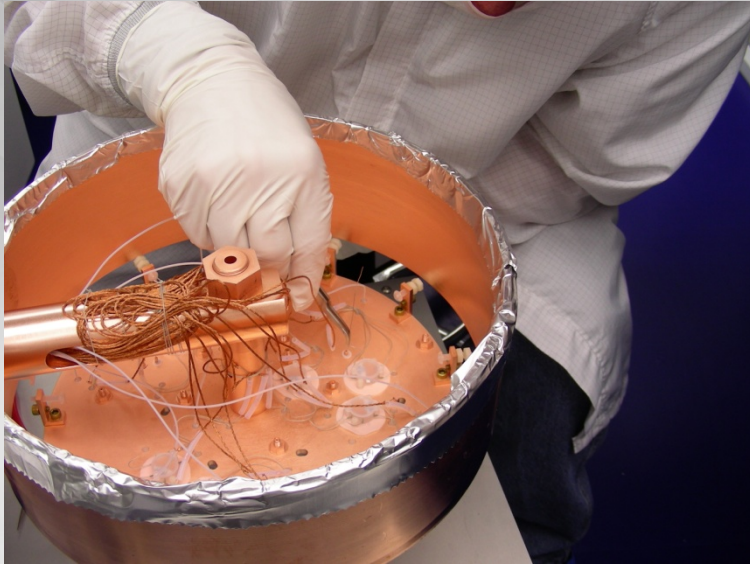
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Installation of Wiring, Front-End Electronics



Installing HPGe and hand-wiring electrical connections – no commercial connectors!



CASCADES 14 HPGe Crystal Gamma Spectrometer



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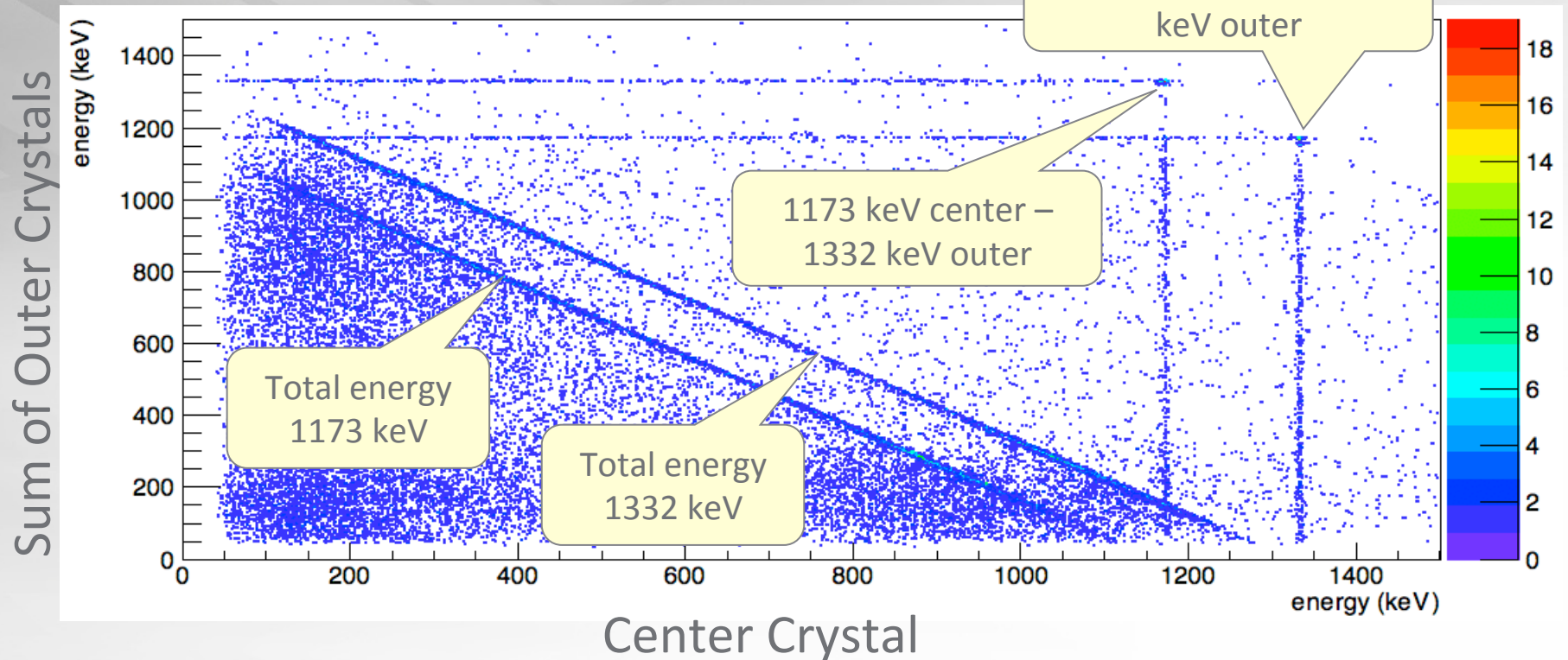
LABORATORY

Since 1965



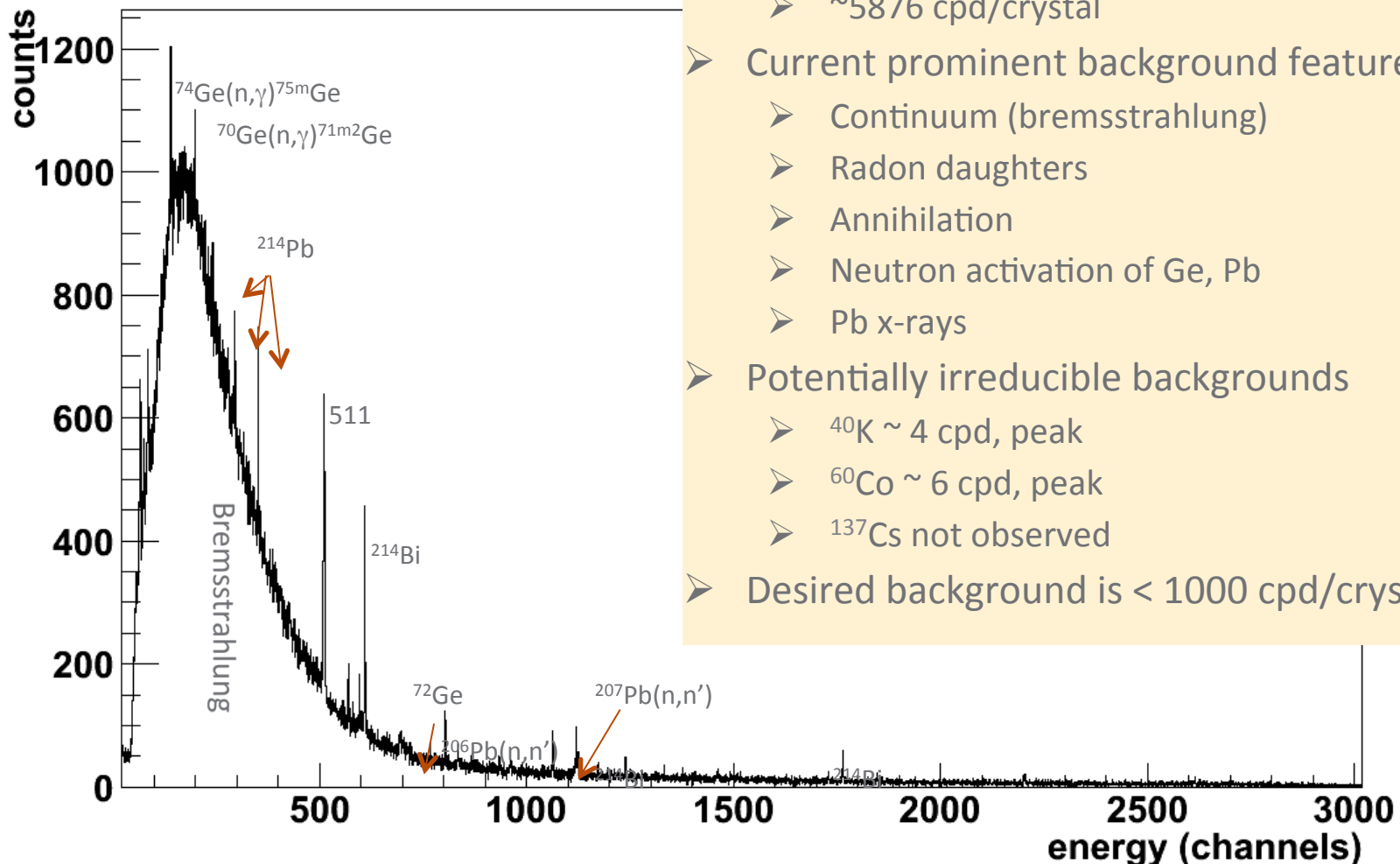
January 7, 2016

Coincidence spectrum from short count of ^{60}Co



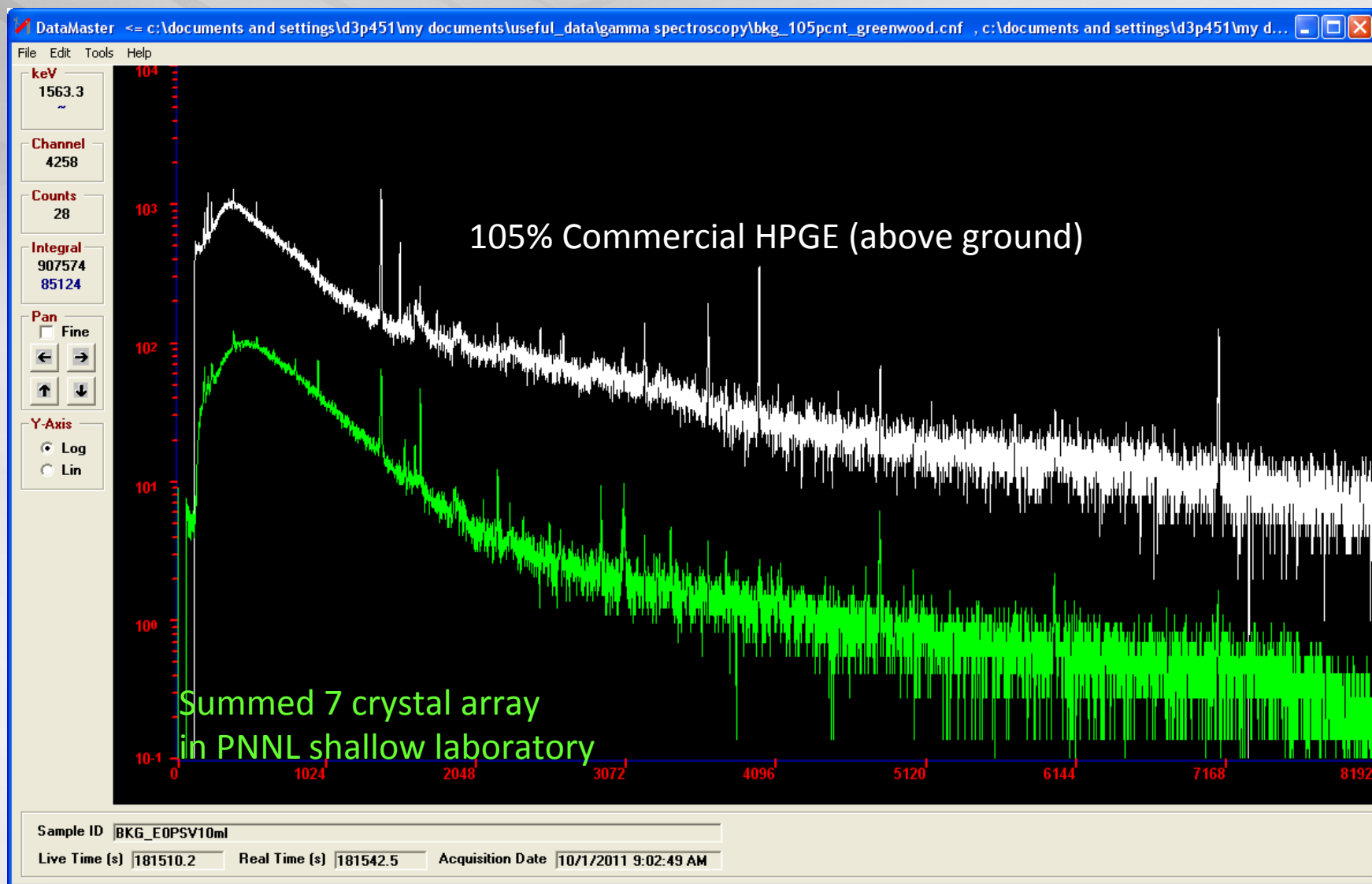
- ▶ Automated analysis tools under development
- ▶ Using C++, ROOT, Coincidence Lookup Library (CLL)
- ▶ First step was developing cascade sum correction framework for 2-D histograms [NIM A 560 (2006) 360–365]

15 day background run, 7 crystals



- 616,970 counts (40-2700 keV)
 - ~5876 cpd/crystal
- Current prominent background features
 - Continuum (bremsstrahlung)
 - Radon daughters
 - Annihilation
 - Neutron activation of Ge, Pb
 - Pb x-rays
- Potentially irreducible backgrounds
 - ^{40}K ~ 4 cpd, peak
 - ^{60}Co ~ 6 cpd, peak
 - ^{137}Cs not observed
- Desired background is < 1000 cpd/crystal

Background Comparison



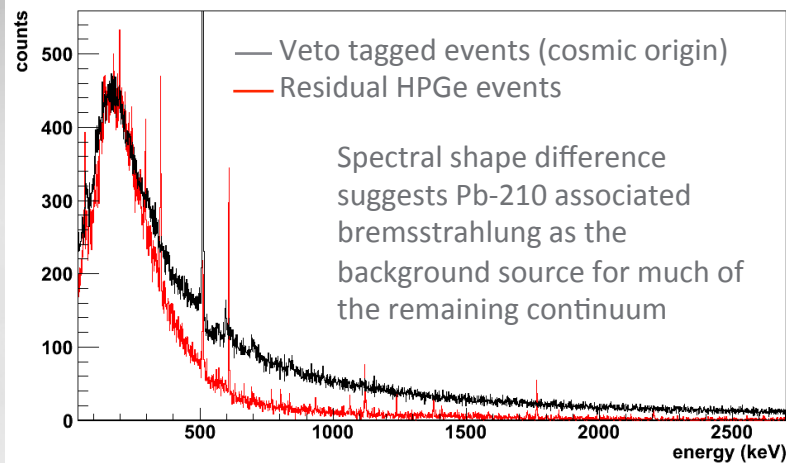


Path to further reduced background

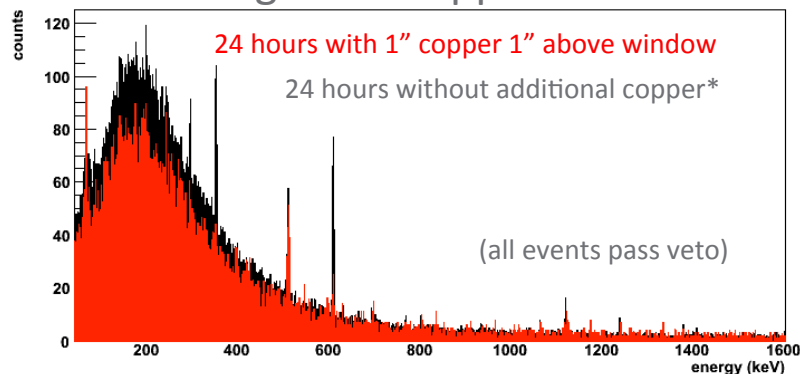
Evidence of

^{210}Pb associated bremsstrahlung

Difference in continuum shape



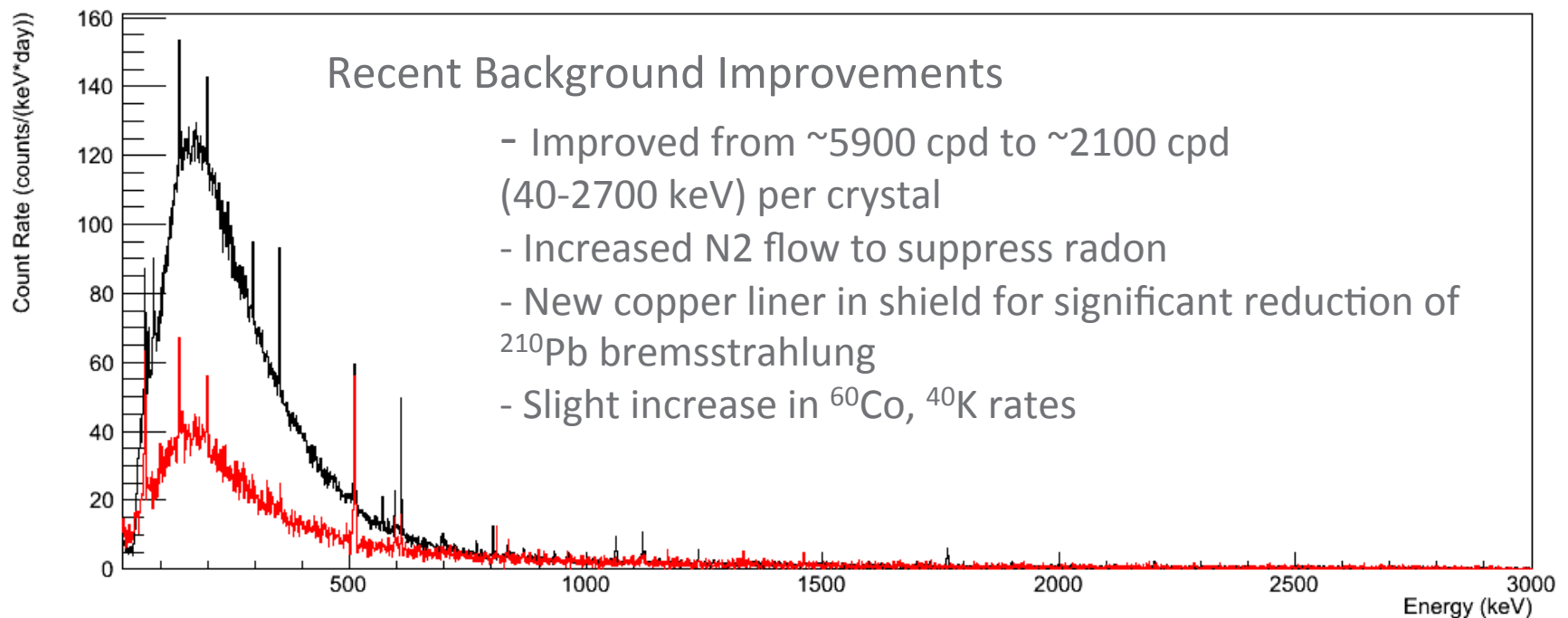
Background suppression



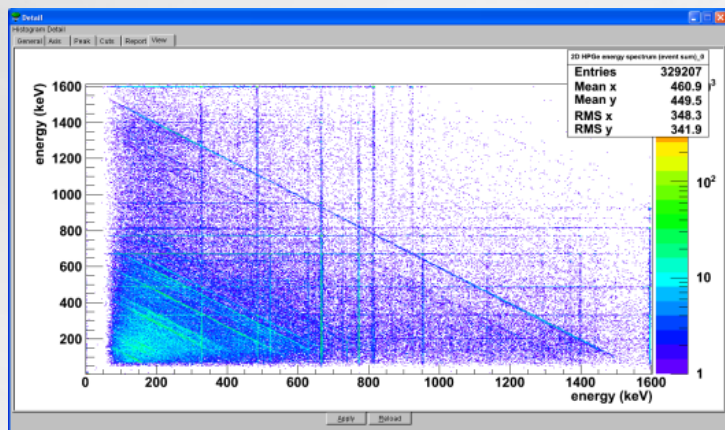
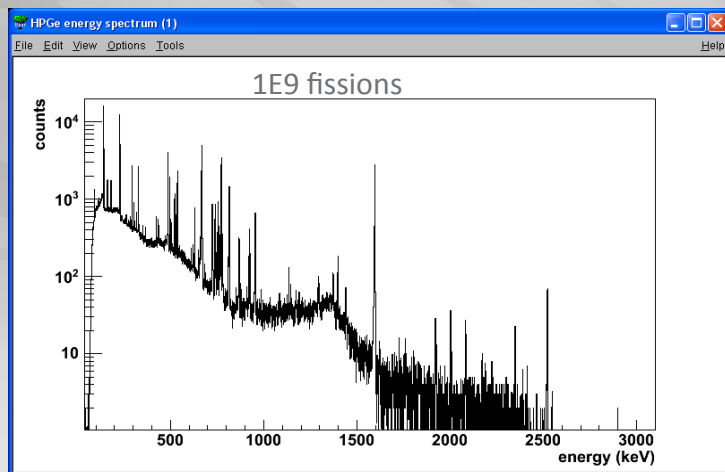
*4.7 days scaled to 24 hours.

- ▶ Complete radon exclusion
- ▶ Complete, optimize active anti-cosmic system
 - ▶ Possible loss of overflow events in PVT
 - ▶ Coincidence timing may not be optimum
- ▶ Verify, eliminate source of continuum
 - ▶ Pb-210 related bremsstrahlung
 - ▶ Solution: inner lining with copper or better lead
 - ▶ Possible cosmic related bremsstrahlung
 - ▶ Reduce shield thickness

Status of array background after improvements implemented



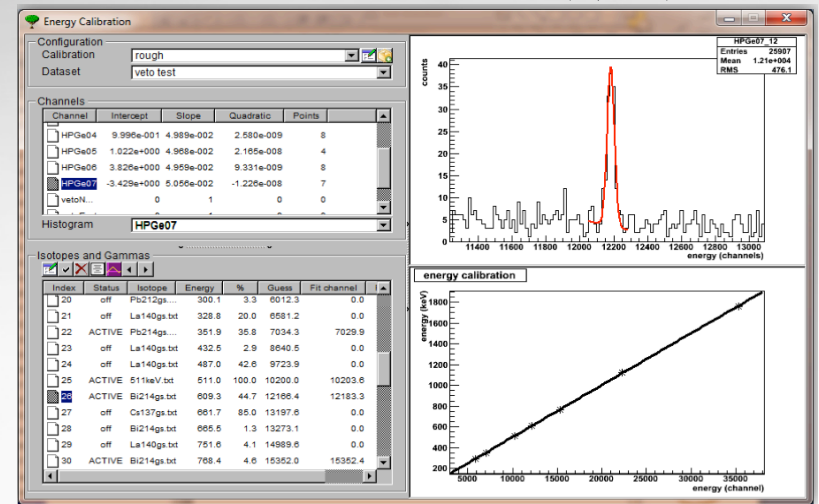
Development of supporting analysis



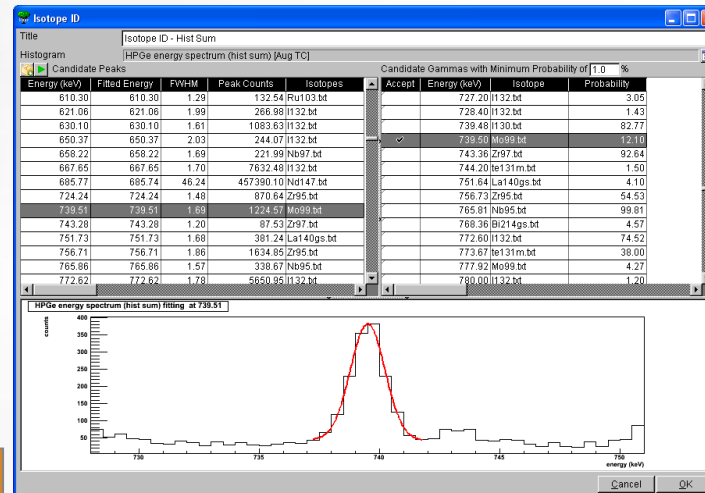
“Singles” and coincidence spectra from thermal irradiation of ^{235}U

Research Team

Craig Aalseth, Anthony Day, Luke Erikson, Brian Glasgow, Eric Hoppe, Brian Hyronimus, Todd Hossbach, Leila Mizouni, Martin Keillor, Allan Myers, Cory Overman, Allen Seifert, Timothy Stavenger



Energy calibration panel



Isotope ID



CASCADES Summary

- ▶ 14 Crystal Array
 - ▶ Low Background Construction
 - ▶ Active anti-cosmic shield
 - ▶ Radon exclusion system
 - ▶ Operates in PNNL Shallow Underground Lab
 - ▶ XIA Pixie-4 Digitizers
- ▶ 72 hour MDA
 - $^{40}\text{K} \sim 40 \text{ mBq}$
 - $^{60}\text{Co} \sim 4 \text{ mBq}$
 - $^{137}\text{Cs} \sim 2 \text{ mBq}$
 - $^{228}\text{Ac} \sim 20 \text{ mBq}$
 - ▶ Sample Geometry
 - $< 4.3 \text{ cm thick}$
 - $< \sim 15 \text{ cm diameter}$
 - Larger diameter can be measured, but detection efficiency is poor outside of 15 cm

NDA of Spent Fuel Ultra-High-Rate Germanium (UHRGe)

Work sponsored by DOE/NNSA

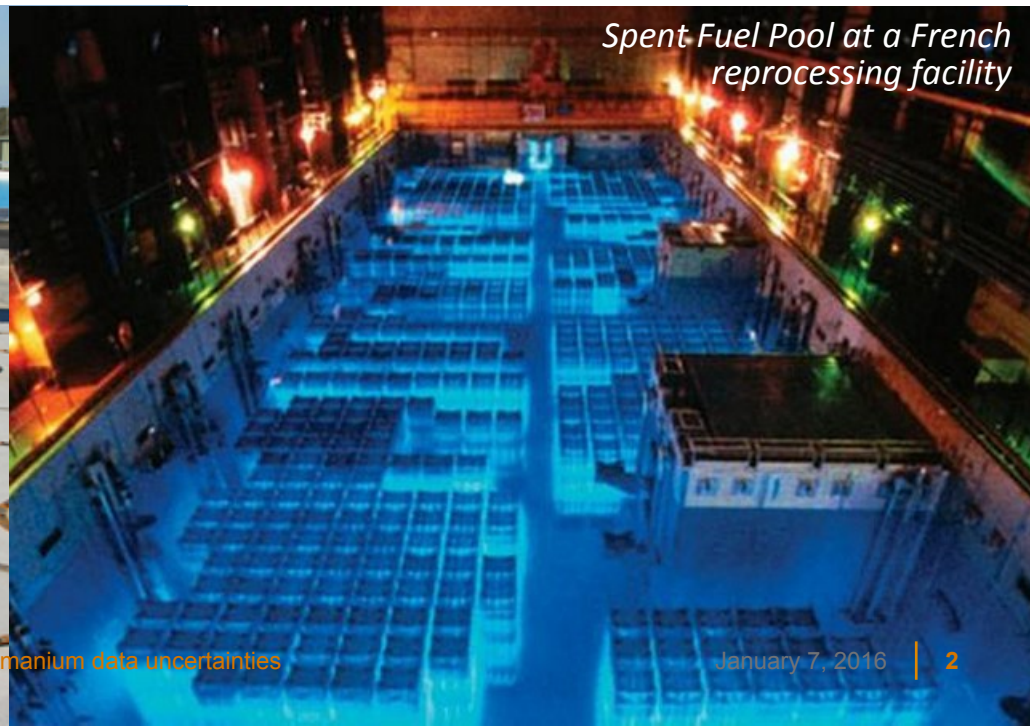
Office of Defense Nuclear Nonproliferation R&D (DNN R&D)



The Spent Nuclear Fuel Concern

- ▶ **One** spent fuel assembly can contain about $\frac{1}{2}$ of IAEA “**concerning**” **amount** of plutonium.¹
- ▶ Want to **non-destructively** determine spent fuel properties to confirm operator declarations or prepare for final disposition.

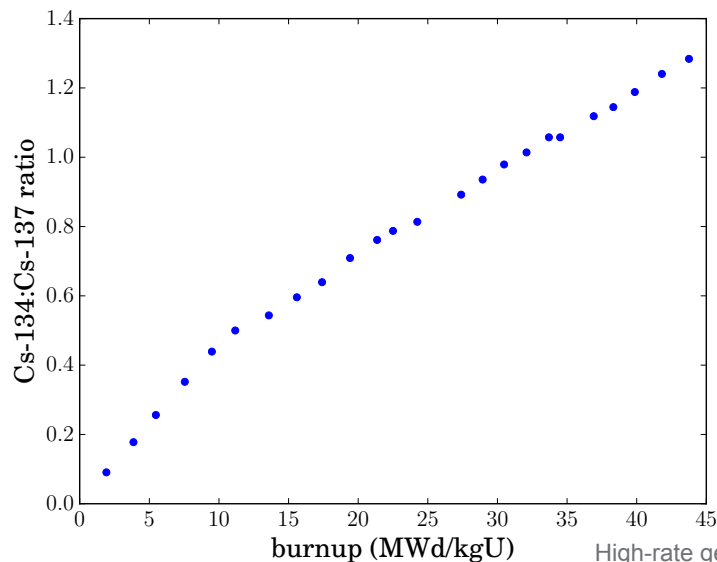
¹Y. Abushady, “Can light water reactors be proliferation resistant?” IAEA-SM-367/15/08





NDA Assay Challenge

- ▶ Assay spent nuclear fuel noninvasively with high-rate, high-purity germanium (HPGe) data
 - Signatures of gamma spectrum are significantly muted in spent fuel with long cool-down times (>20 years)
 - Here, HPGe detectors, which have good energy resolution, are modified to function at high event rates (in excess of 10^6 events/s)²
- ▶ The Cs-134:Cs-137 ratio is used to determine fuel history (i.e., burnup and cooling time)³



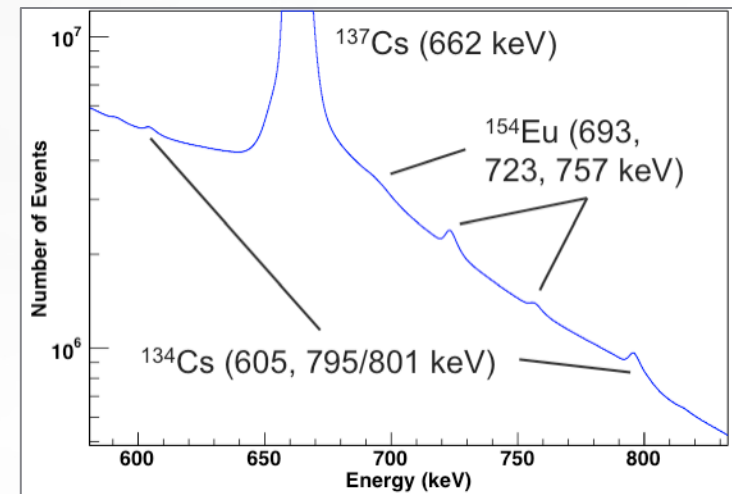
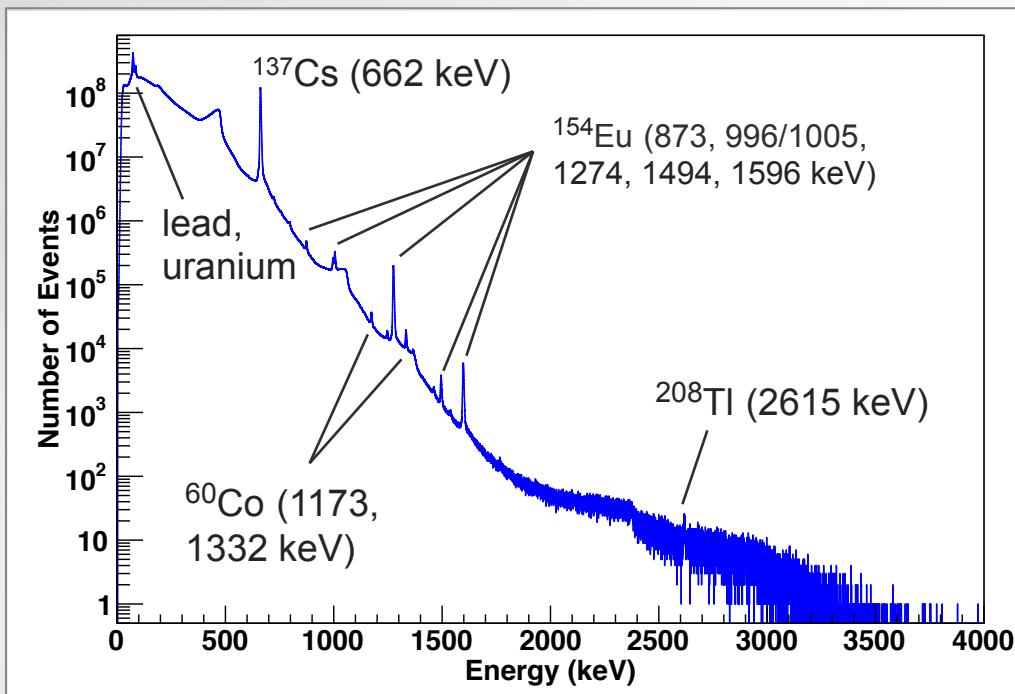
Cooling time (yrs)	Cs-134:Cs-137
0	1.0
5	2.1×10^{-1}
10	4.4×10^{-2}
20	1.9×10^{-3}
30	8.4×10^{-5}

² B. A. VanDevender, et. al., "High-Purity Germanium Spectroscopy at Rates in Excess of 10^6 Events/s," *IEEE Transactions on Nuclear Science*, 2014.

³ G. Kirchner, et. al., "Radioactivity from Fukushima Dai-ichi in air over Europe; part 2," *Journal of Environmental Radioactivity*, vol. 114, pp. 35-40, 2012.

Extracting weak signatures at long cooling times

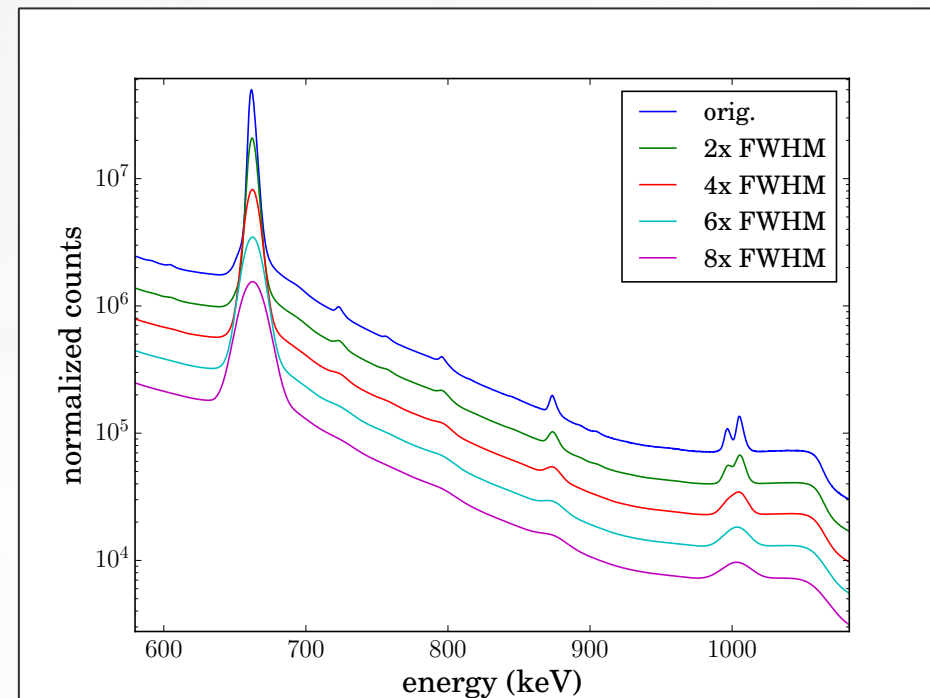
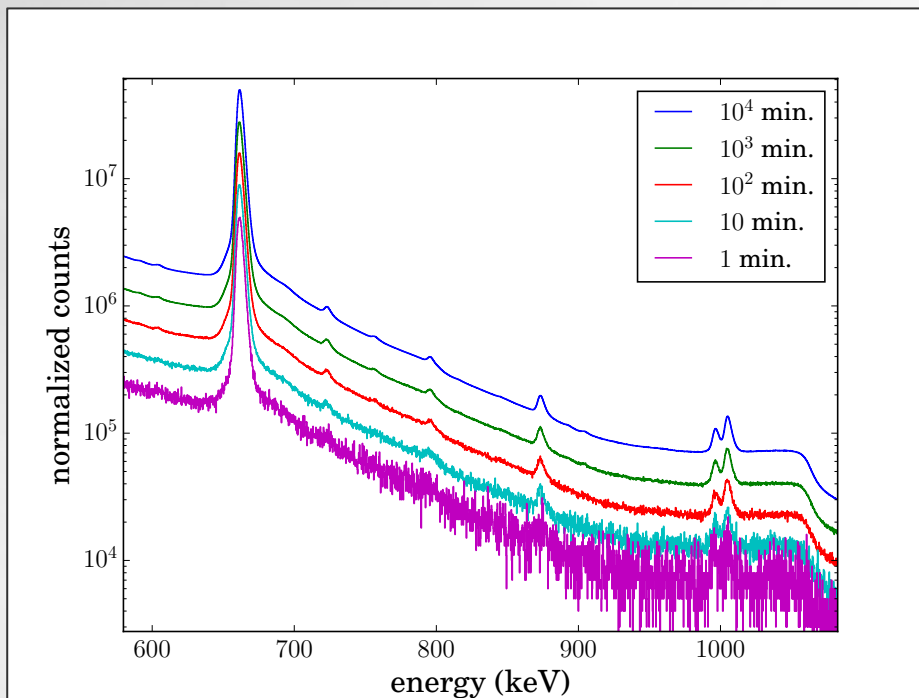
- ▶ At long cooling times, the activity from Cs-134 is quite low compared to the background (dominated by Cs-137)
- ▶ High-rate detector response (pileup!) doesn't help





Measurement time dictated by operators

- For compatibility with facility operations, measurements on spent fuel will likely have to be **completed on the** order of minutes,⁴ requiring high-count rates ($>10^6$ counts/s) and good energy resolution

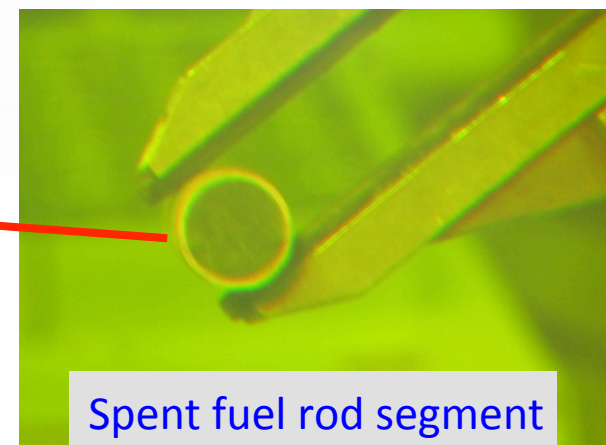
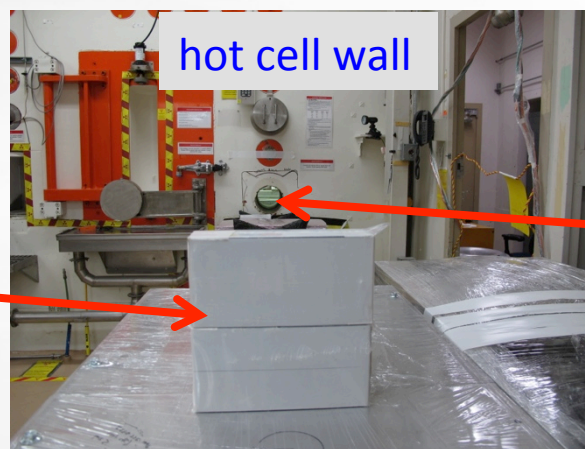
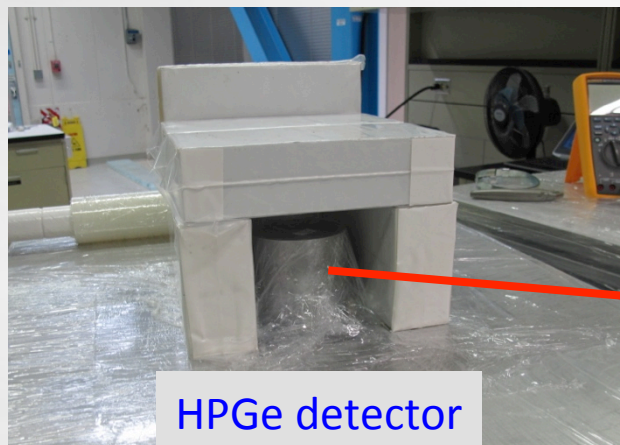


⁴ N. Lundkvist, *et. al.*, "Investigation of possible nondestructive assay (NDA) techniques for the future Swedish encapsulation facility," 53rd Annual INMM, 2012.



Measurements on SNF rod segment

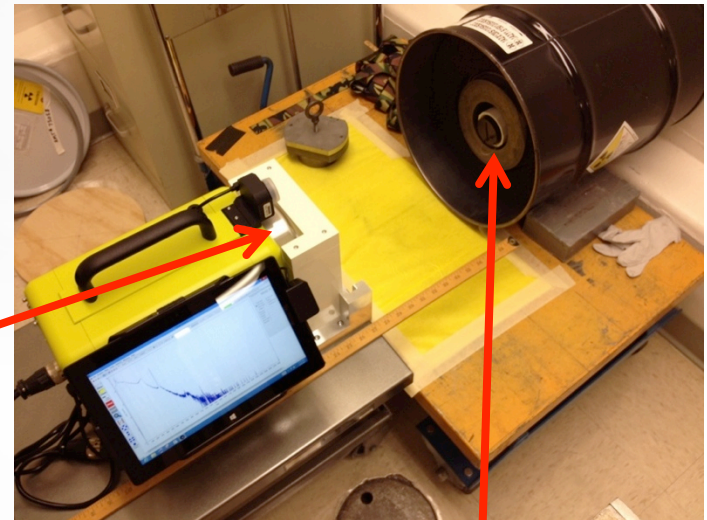
- ▶ ATM-109 is high burnup BWR fuel from Quad Cities I
 - 60-70 GWd/tU, irradiated 1979-87 and 1989-92
 - 58 grams (~55 cm) rod segment with cladding
- ▶ Ultra high-rate germanium (UHRGe) system used to measure fuel
 - Fuel viewed through open port in back of hot cell
 - Input count rates up to 1.4 Mcps
 - 180 second exposures recorded





Collecting high statistics with fuel slice

- ▶ ATM-109 is high burnup BWR fuel from Quad Cities I
 - <60 GWd/tU, irradiated 1979-87 and 1989-92
 - 0.5 g rod segment (~0.5 mm) with cladding
- ▶ A modified version of the PHDS, Co. germanium gamma imager (GeGI)⁵ was used for a long, 2-week count with a smaller section of ATM-109
 - Segmented germanium with 32 channels
 - 10 cm diameter, 1 cm thickness



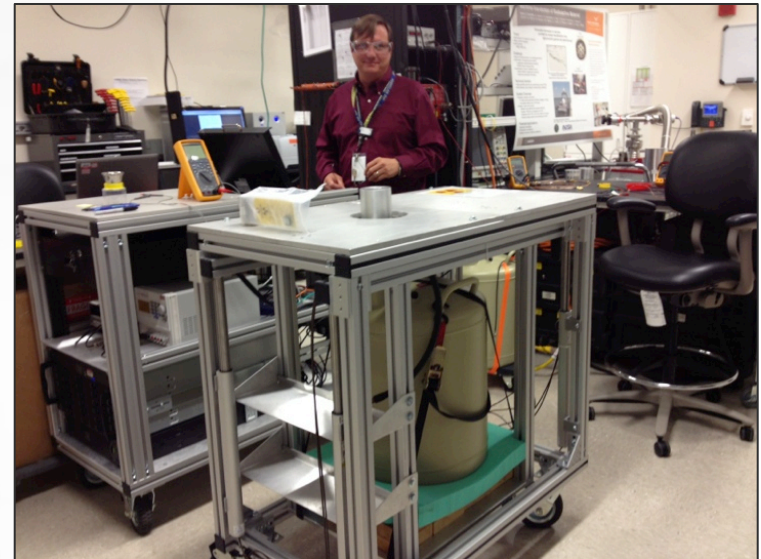
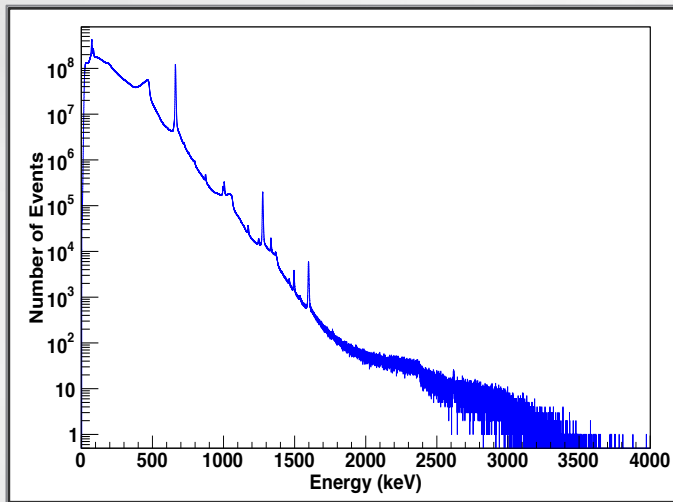
HPGe detector

Spent fuel rod segment



Selection of detector system

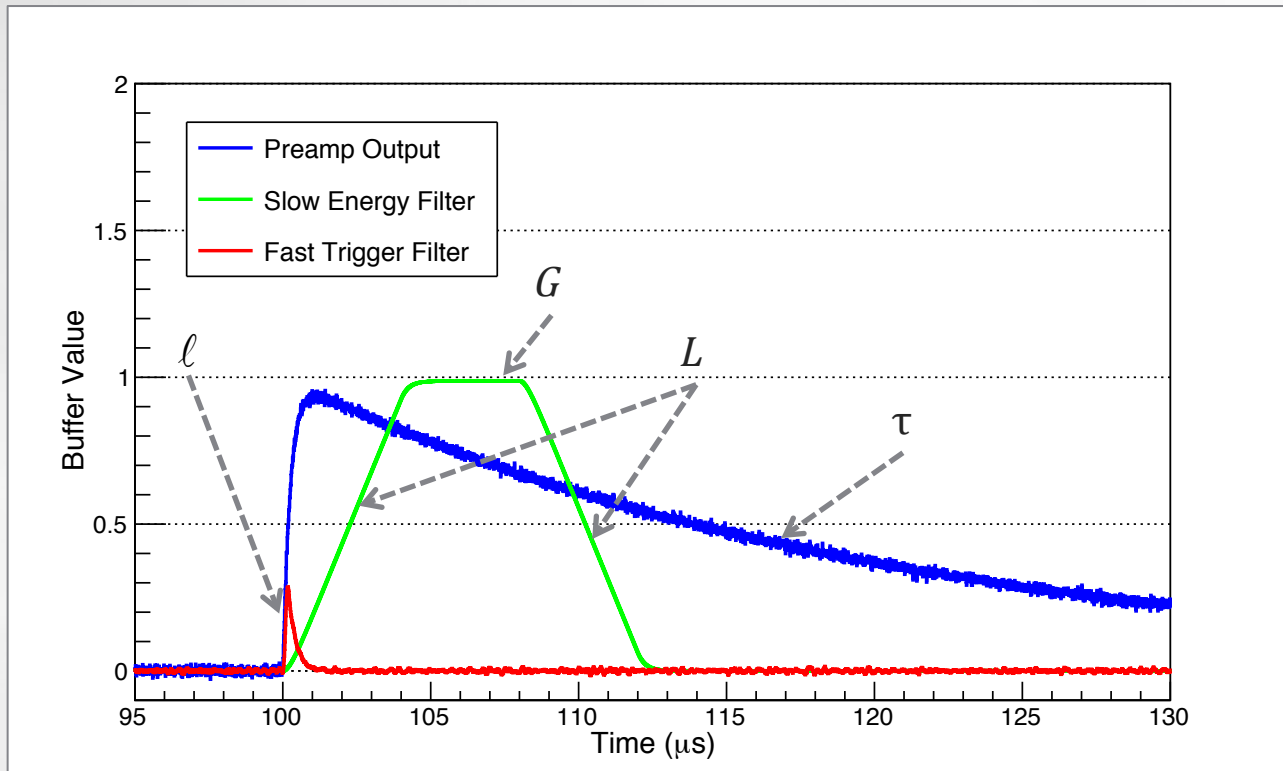
- ▶ Data from the PNNL-developed ultra-high-rate germanium (UHRGe) detector system is used here due to the unique, direct access to the preamp output, even at high-rates
 - Can modify slow and fast filter parameters
 - Can modify pileup rejection methods
 - Can answer not just what, but why





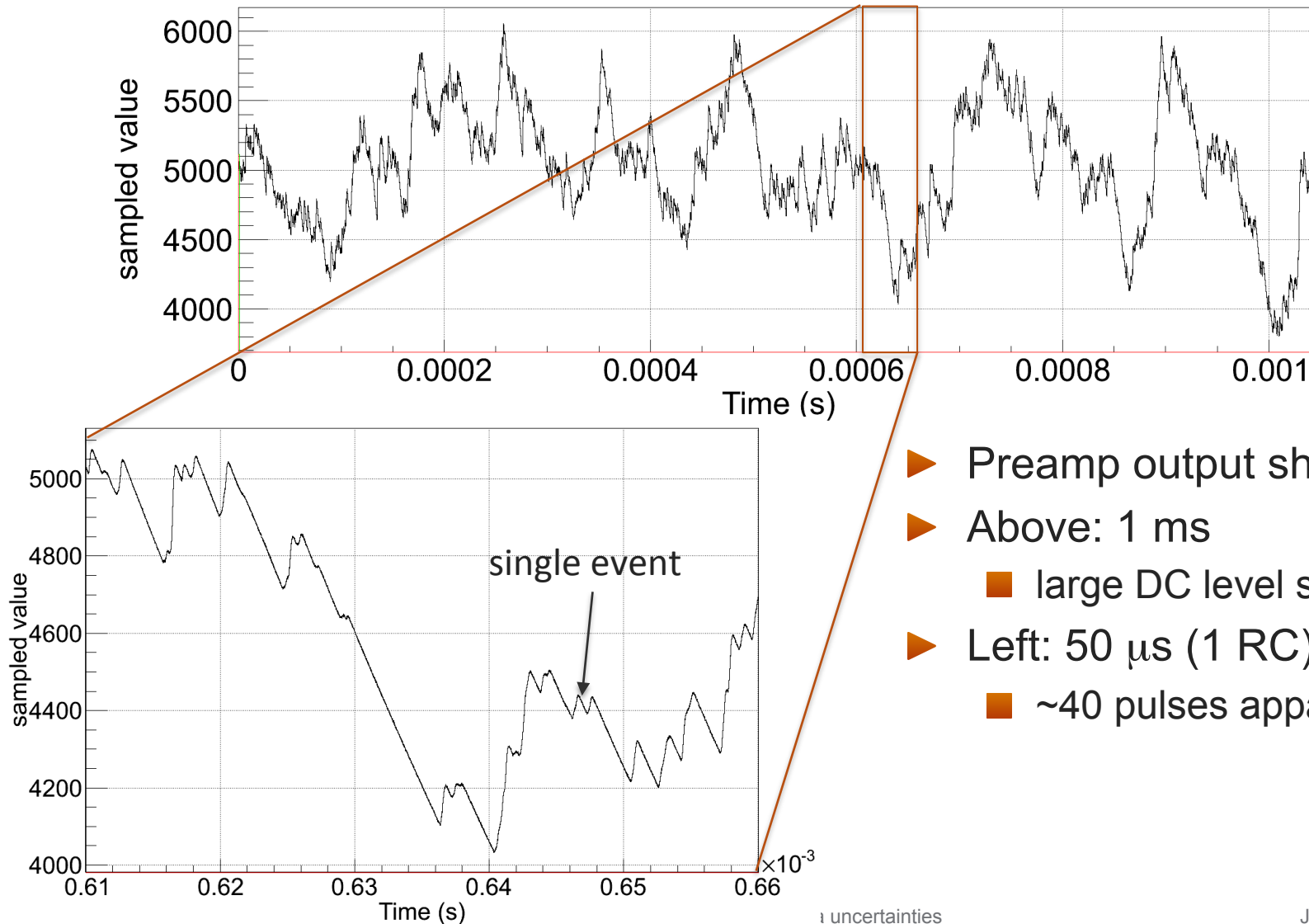
Detector response – a trapezoidal filter

- ▶ Fast filter response, ℓ
- ▶ Slow filter rise time L and gap G
- ▶ Preamplifier decay constant τ



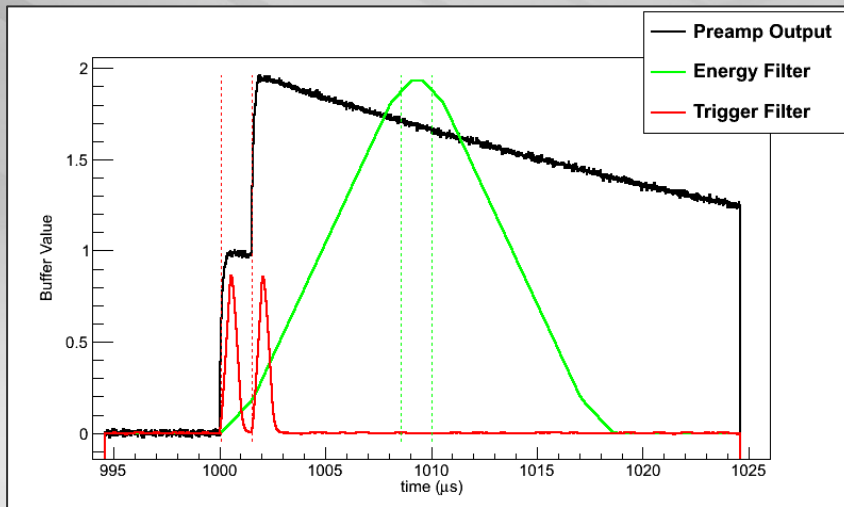


Detector response – very high-rate

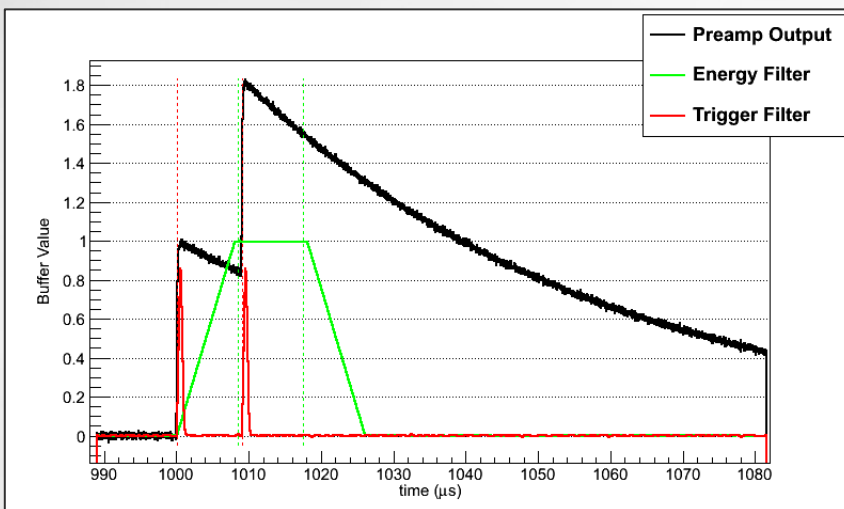
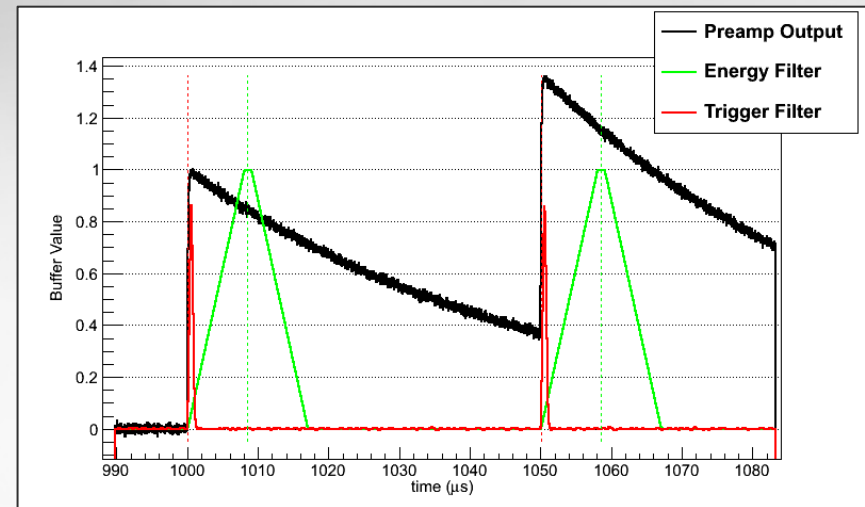


- ▶ Preamp output shown
- ▶ Above: 1 ms
 - large DC level shift
- ▶ Left: 50 μ s (1 RC)
 - ~40 pulses apparent

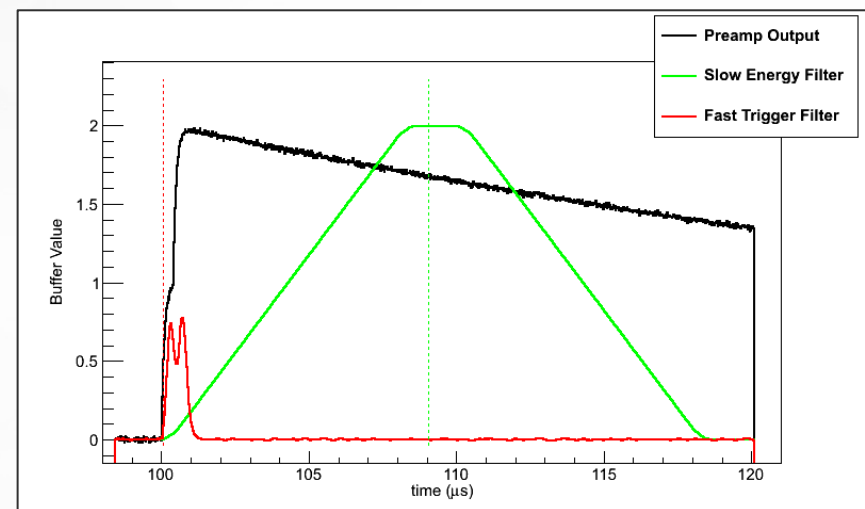
Pileup possibilities



(a) Pileup reject

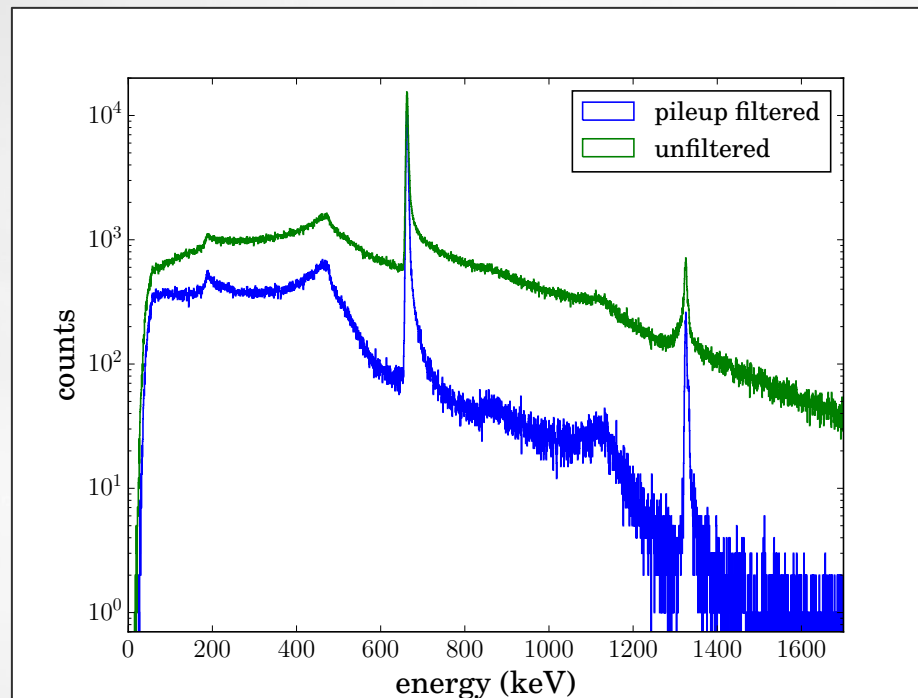


(c) Pileup maybe allow



Results

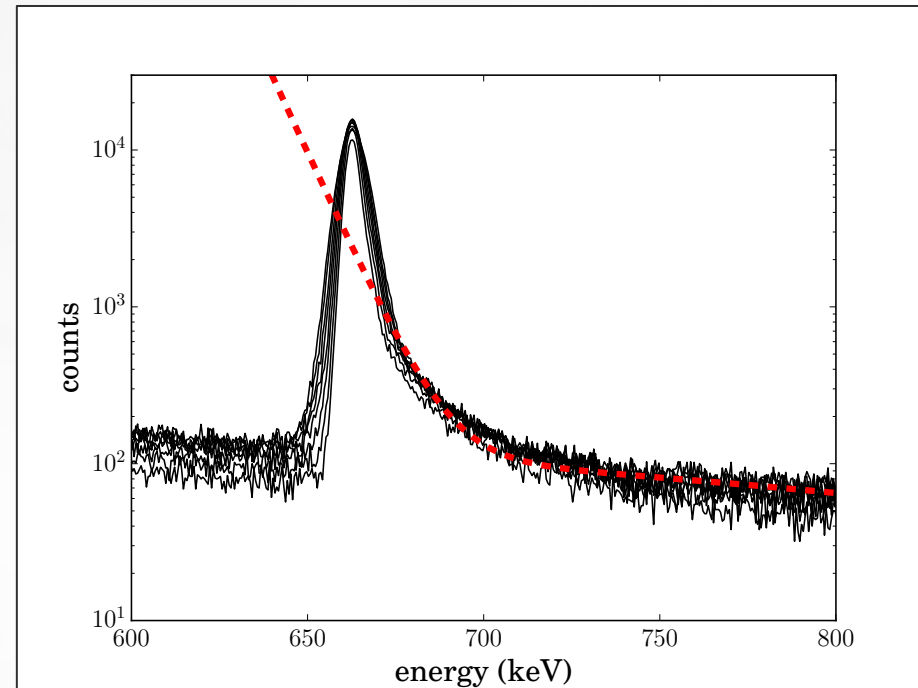
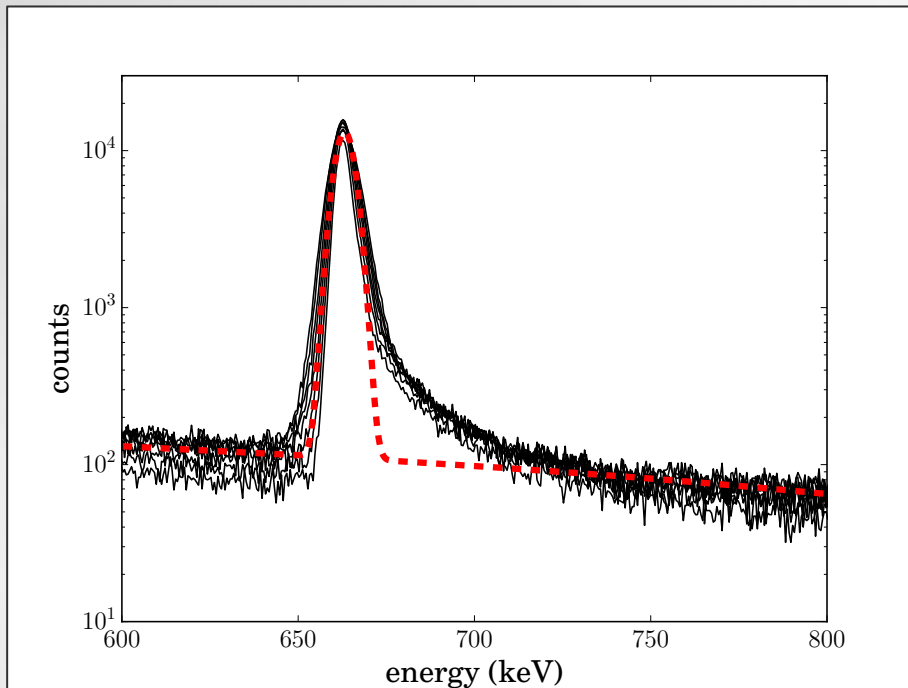
- ▶ Filtering out events that occur within the slow filter rise time and gap ($L+G$) removes much of the pileup.
 - However...





Peak shape curve fitting

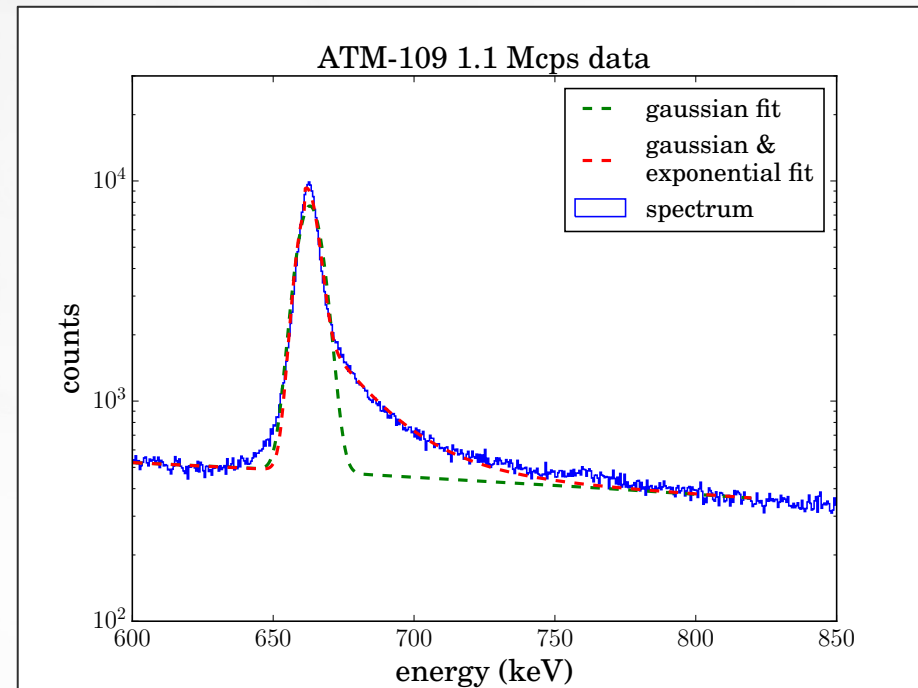
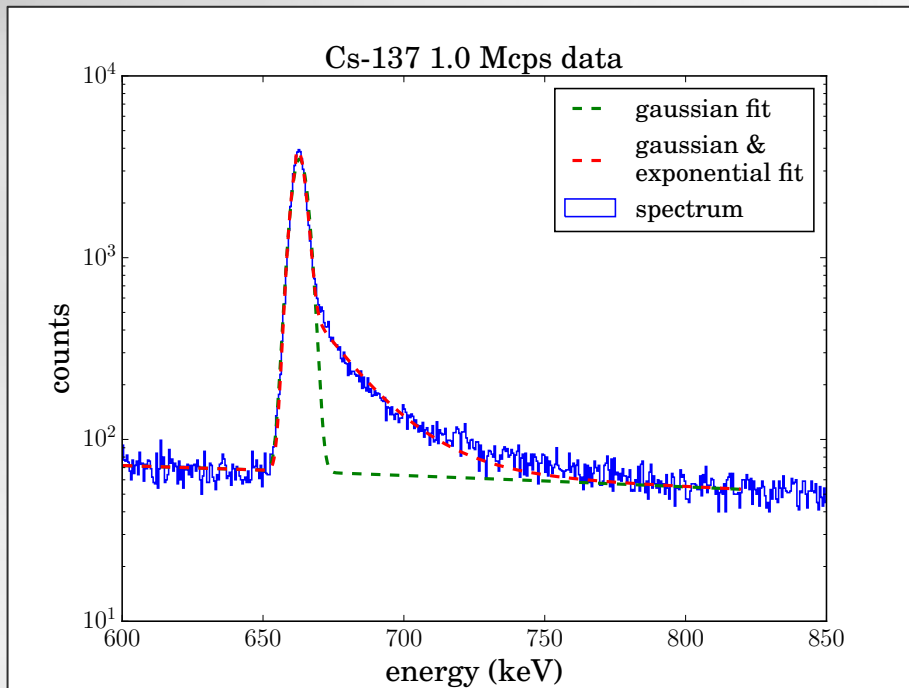
- ▶ There is still a significant tail on the peak
- ▶ A Gaussian clearly does not capture the peak shape
 - It underestimates the peak area by about 25%





Peak shape curve fitting

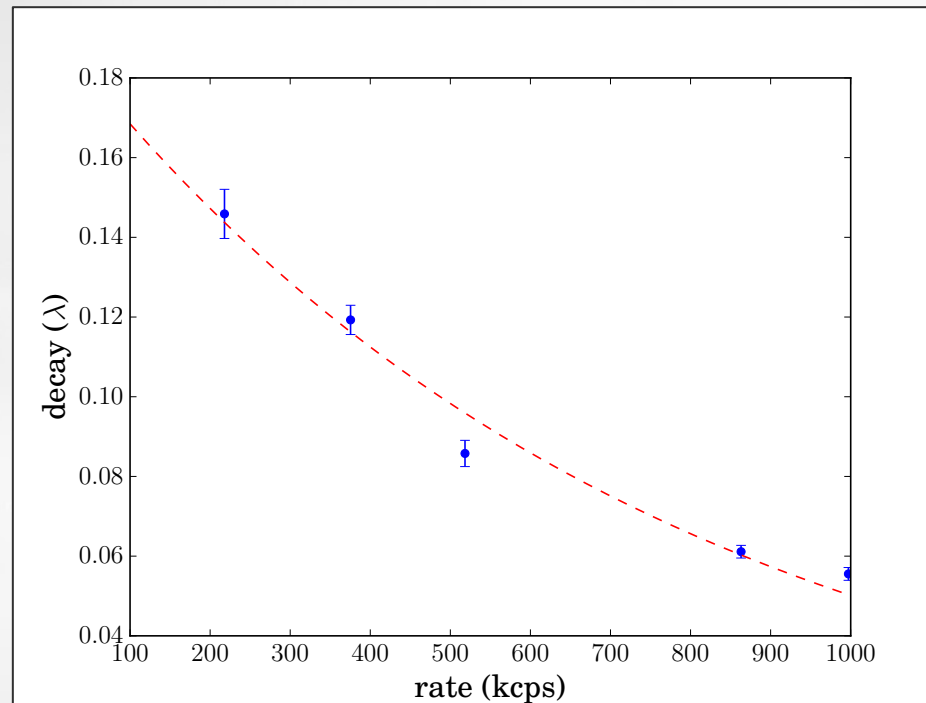
- ▶ A Gaussian plus an exponential fits the peak shape well
 - Consistent shaping at a number of rates and inspected objects





Exponential decay constant vs. rate

- ▶ As expected, the tail on the high-energy side of the photopeak is longer at higher rates
 - An indication that increased rate is linked to increased pileup



High-rate germanium data uncertainties



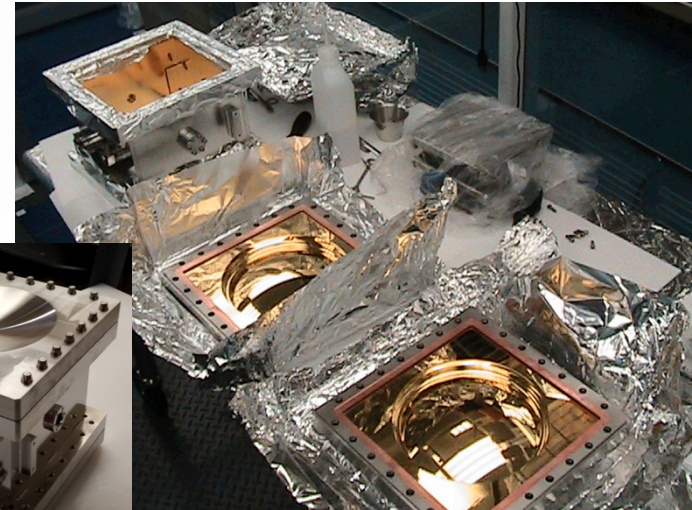
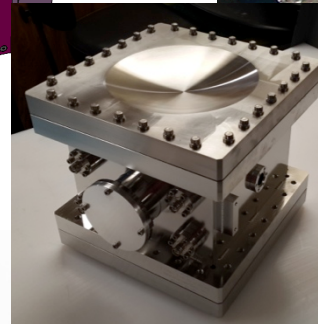
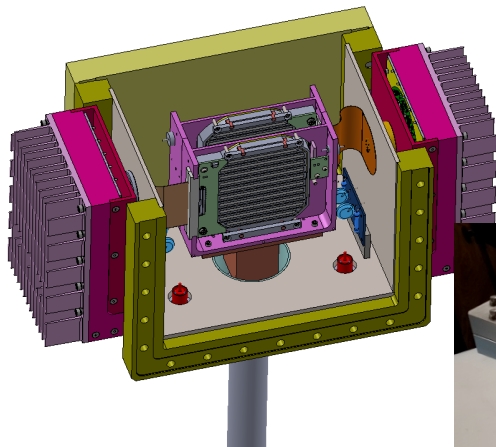
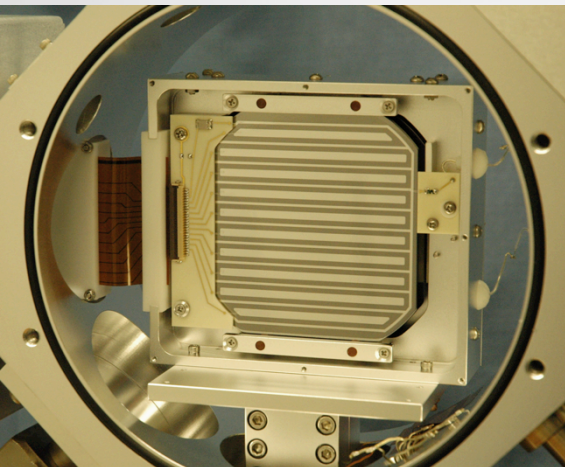
Relevance to particle physics

- ▶ COMET and mu2e experiments seek to measure muon to electron conversion in the presence of a nucleus (lepton flavor violation)
- ▶ This is an extremely rare process – seek sensitivities of 10^{-18}
- ▶ Experiments stop muons in a target and look for the distinct electron emitted in the process with a spectrometer
- ▶ But what if they see signals?
 - Then the “denominator”, the number of stopped muons becomes critical
 - When muons capture they do so in excited states and emit characteristic muonic x-rays as they de-excite to the ground state
 - These x-rays are in the 300-1500 keV range, but terrestrial backgrounds are an issue for detectors with poor energy resolution – HPGe?
 - Rates are extreme due to bremsstrahlung flash with each beam pulse and comes too close in time to clear from detector before delayed muon x-ray signal arrives
 - Precisely the “rare signal in high rate environment” scenario UHRGe was designed to tackle!



Next steps for UHRGe

- ▶ Artifacts at high event rates cannot be completely removed
 - However, analysis algorithms can work around this
- ▶ Reduction in systematic uncertainties leaves only statistical uncertainties, which can be reduced with longer count times (useful for facility operations)
- ▶ The next generation system will utilize segmented, strip detectors⁶
 - Events are split across multiple channels → faster response
 - Thinner detectors mean faster charge collection → faster response



⁶ R. J. Cooper, *et. al.*, "A prototype High Purity Germanium detector for high resolution gamma-ray spectroscopy at high count rates," NIMA, 2015.



Summary

- ▶ In the last 10 years, PNNL has extended work HPGe in three areas
 - Use of arrays of detector elements to greatly increase detection efficiency and acceptance
 - Extended ultra-low-background techniques to create a one-of-a-kind large acceptance coincidence counting system
 - Pushing rate capability of HPGe an order of magnitude further than prior efforts to operate at high rates
- ▶ Most of this development has been driven by nuclear security and nuclear treaty requirements
- ▶ Technology from this work has made its way back to basic science, e.g. the Majorana ^{76}Ge $0\nu \beta\beta$ -decay search and dark matter efforts (CoGeNT) – strong connection to dark matter and neutrino physics
- ▶ Increasingly, both nuclear security and physics applications drive detector R&D at PNNL