

Low-Temperature Detectors for High-Precision Measurements

Mark Croce
on behalf of the LANL-NIST-U. Colorado collaboration

ORNL Physics Division Seminar
December 2, 2021

LA-UR-21-24545

Outline

- Introduction to LTDs
- X-ray Spectroscopy and Hyperspectral X-ray Imaging
- Gamma Spectroscopy
- Decay Energy Spectroscopy



What do we mean by “low temperature”?

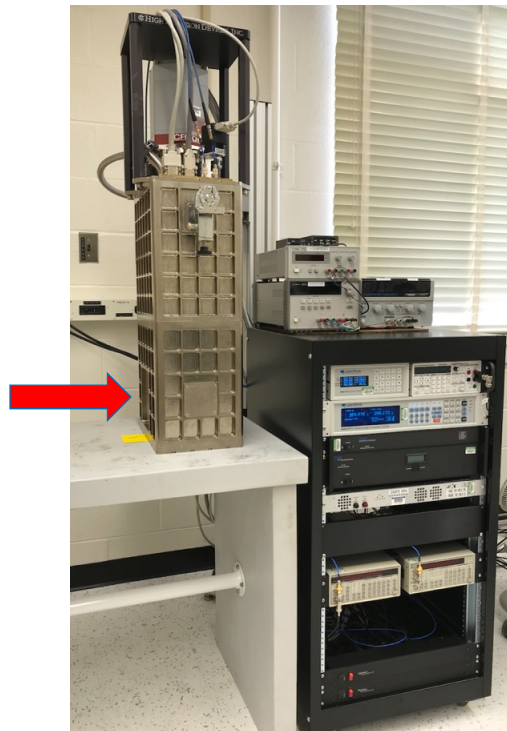
Room temperature: ~ 300 K

Liquid nitrogen: ~ 77 K

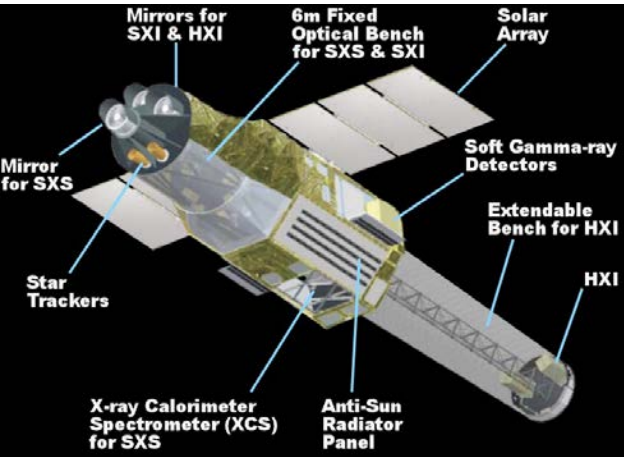
Liquid helium: ~ 4.2 K

Average temperature of universe
(CMB radiation): ~ 2.7 K

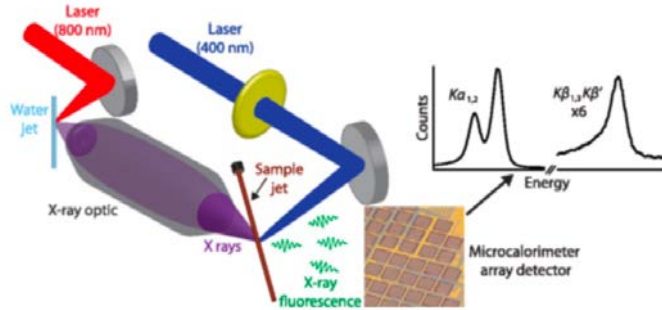
Our detectors: ~ 0.1 K



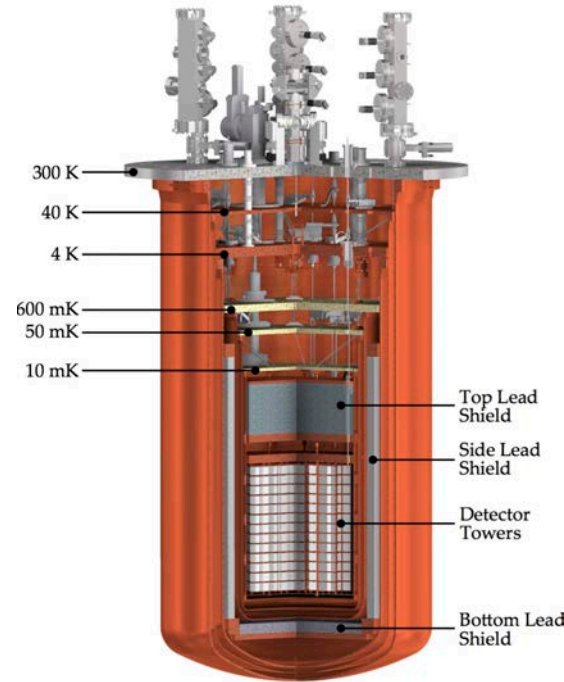
Many applications...



Hitomi (Astro-H) Soft X-ray Spectrometer (credit: JAXA/NASA)



*Picosecond tabletop X-ray spectroscopy to watch chemical reactions
L. Miaja-Avila et al., Phys. Rev. X, 2016*

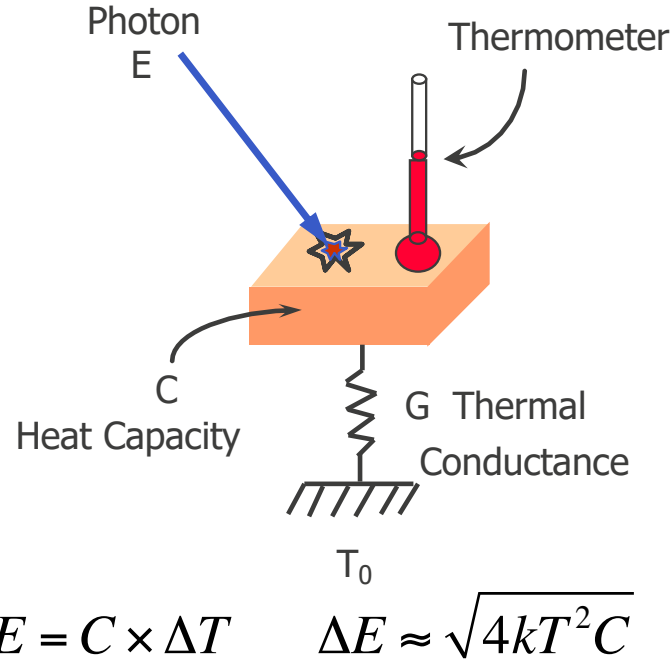


CUORE ^{130}Te $0\nu\beta\beta$ search (credit: CUORE collab.)

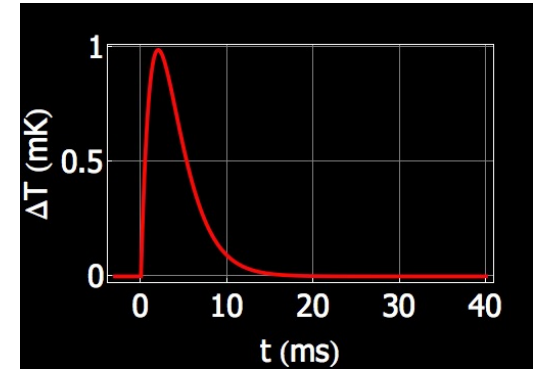
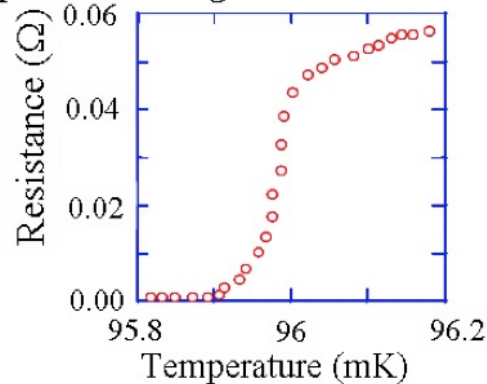


Why so cold? To overcome energy resolution limits.

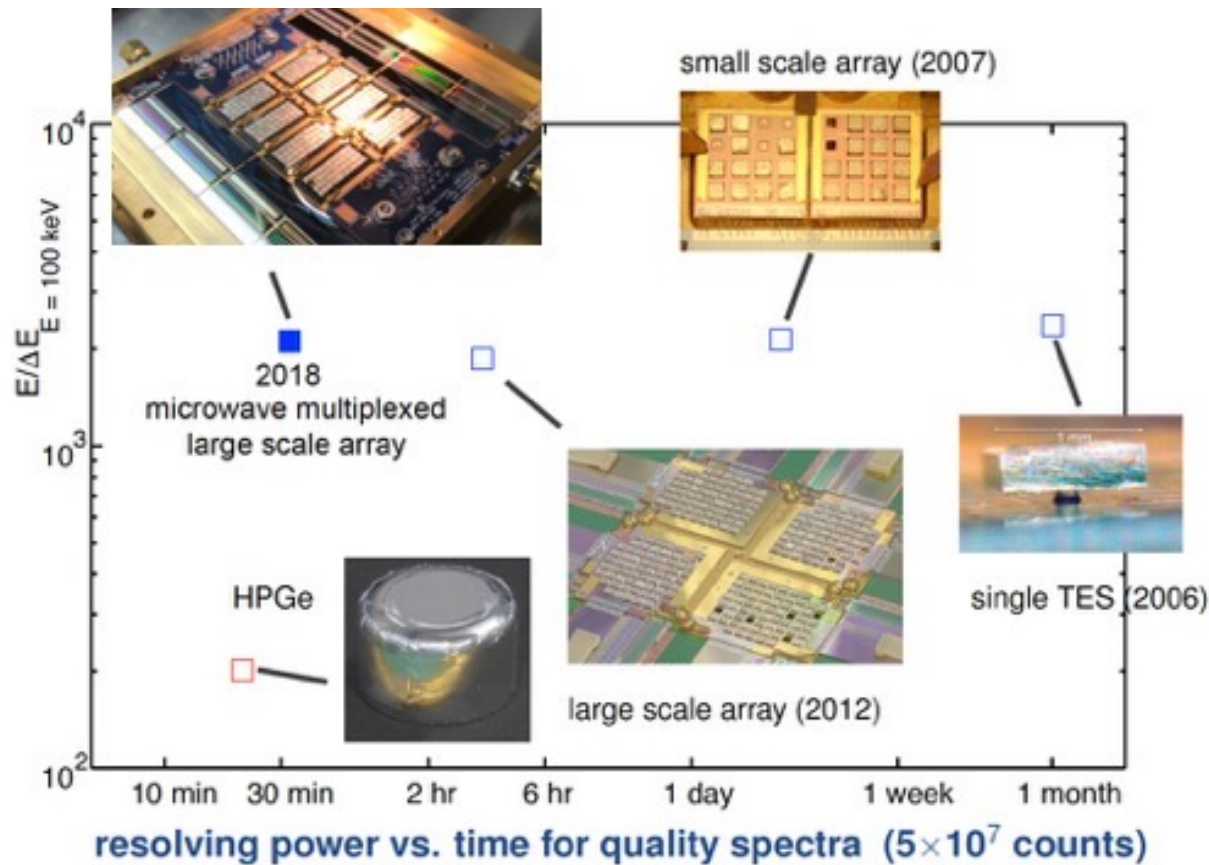
A microcalorimeter measures the thermalized energy of individual photons, nuclear decays, etc. to create an ultra-high resolution energy spectrum (10-50x better than semiconductors)



Superconducting-Normal Transition

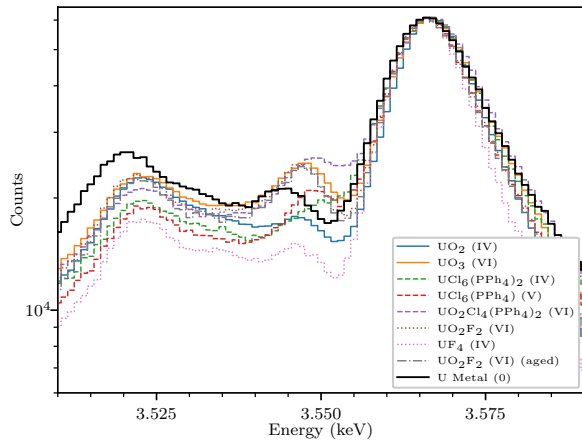


Microcalorimeter technology advancement

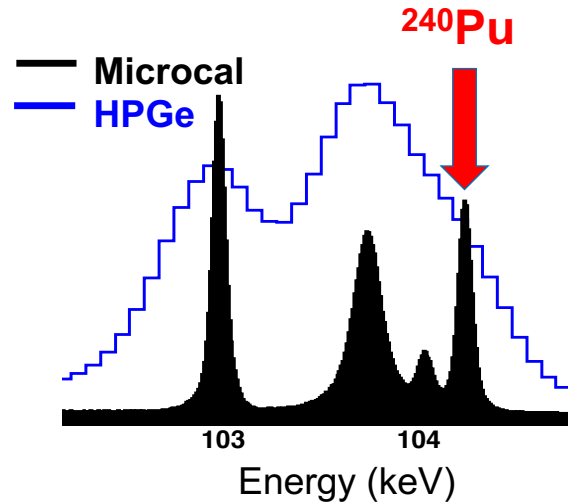


Our focus: nuclear material analysis

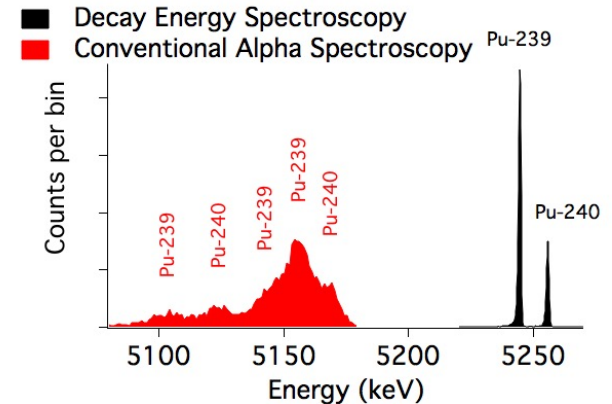
Low-temperature detectors are now creating real measurement capabilities that would be impossible with conventional detectors



X-ray spectroscopy



Gamma-ray spectroscopy



Decay energy spectroscopy



Hyperspectral X-ray Imaging and X-ray Spectroscopy

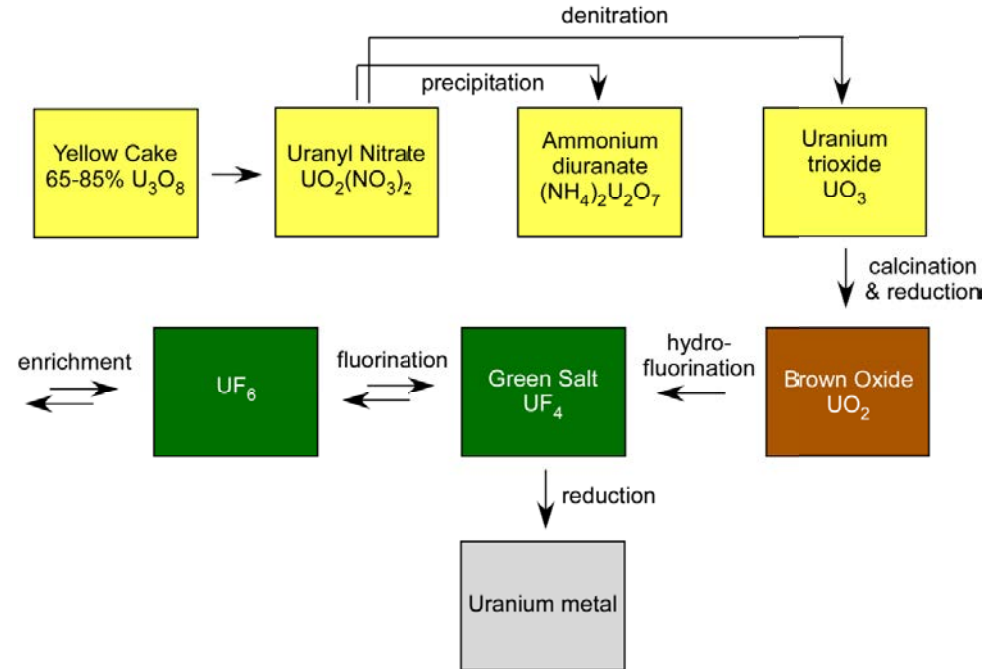
IAEA: Environmental sampling is “the measure confirming the absence of undeclared nuclear material or nuclear activities”

A. Bosko (IAEA), presentation at LANL Oct. 15, 2019

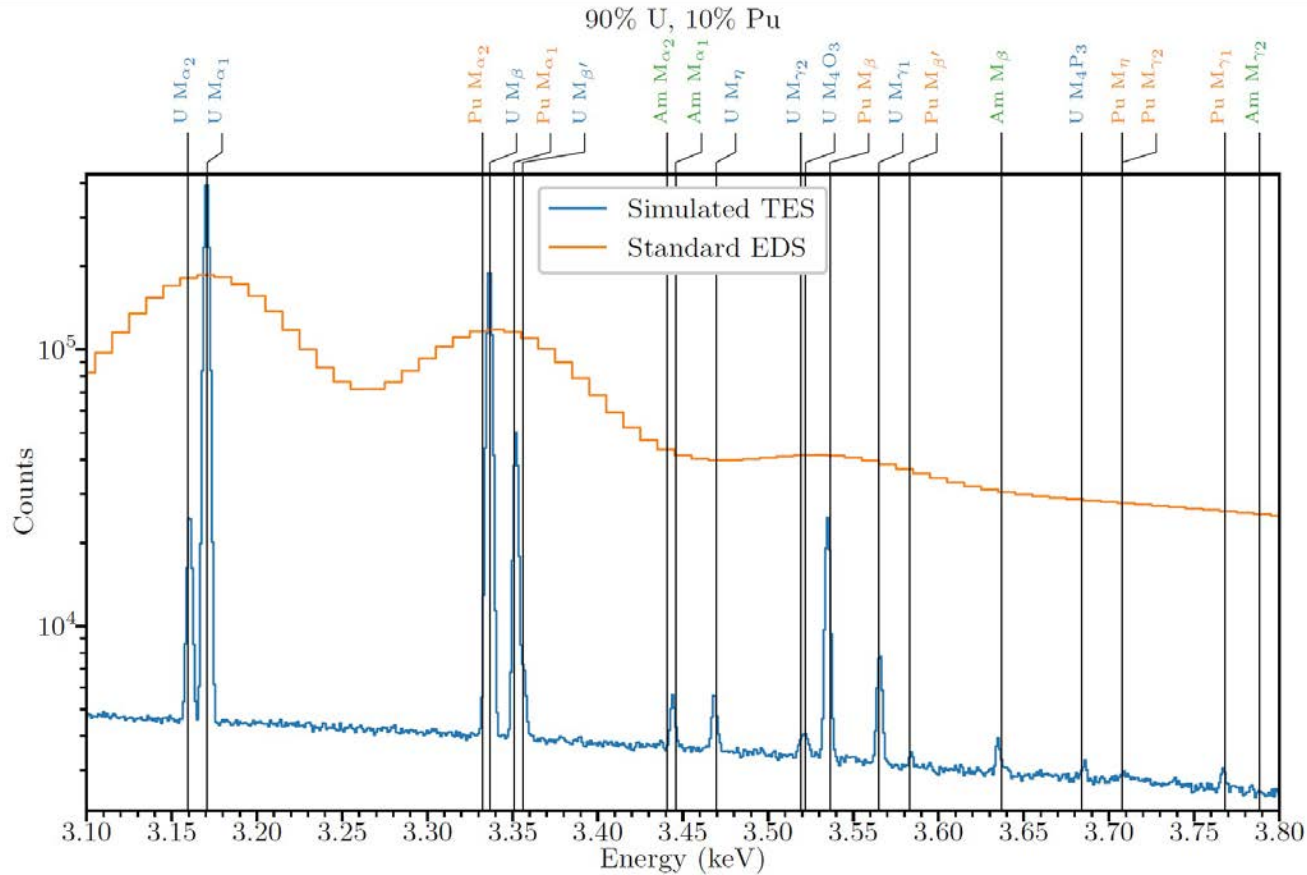


Photo from IAEA

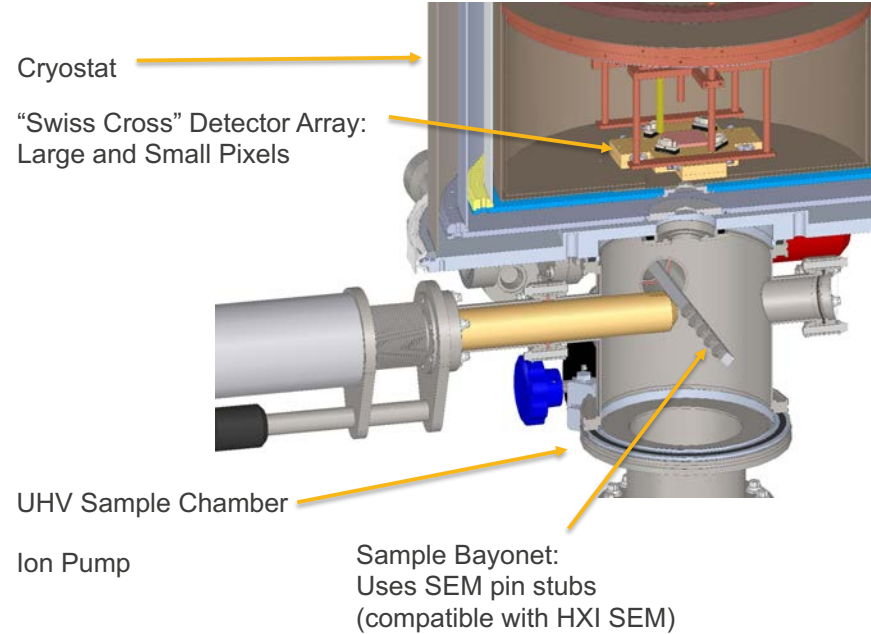
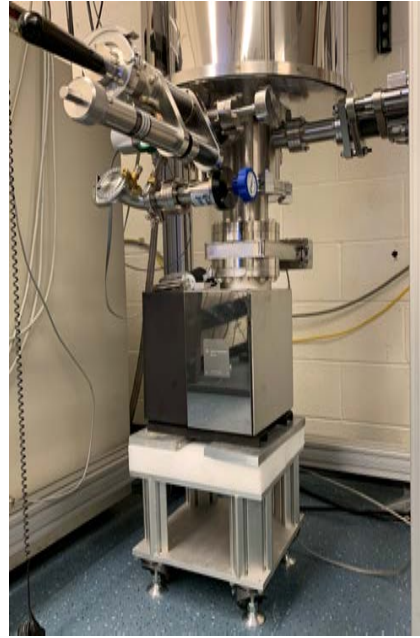
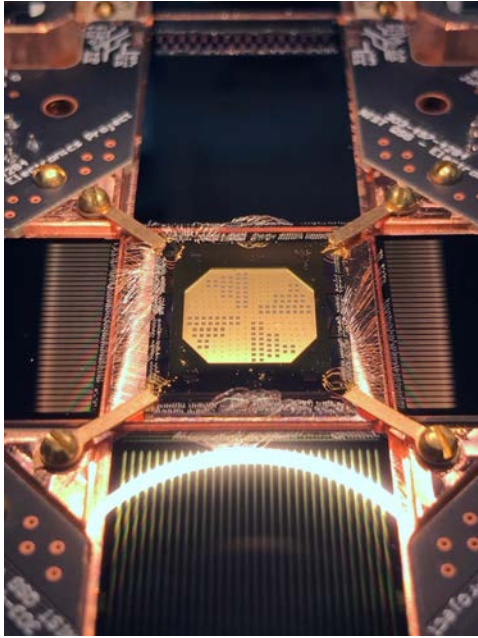
Where in the fuel cycle did a sample originate?
Chemical speciation is a major clue.



SEM/EDS with Si detectors is a valuable tool, but has enough trouble just distinguishing U from Pu



X1 X-ray Fluorescence Instrument for mg-scale samples

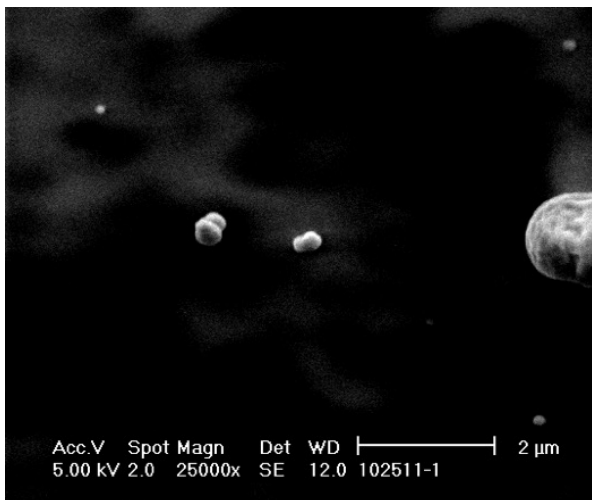


Calibration concept: sample + calibration standards on same pin stub for in-situ calibration

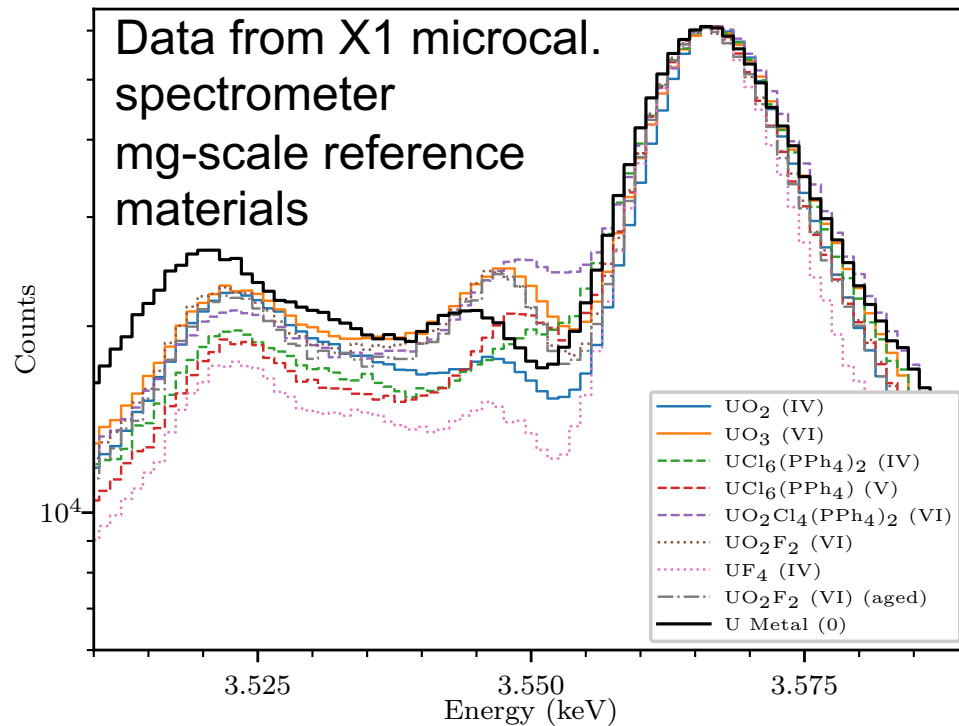


What chemical species are in these small particles?

Too small for optical spectroscopy...



(Hydrolyzed UF_6)



XAFS or HXI



Photo from Advanced Photon Source
www.aps.anl.gov

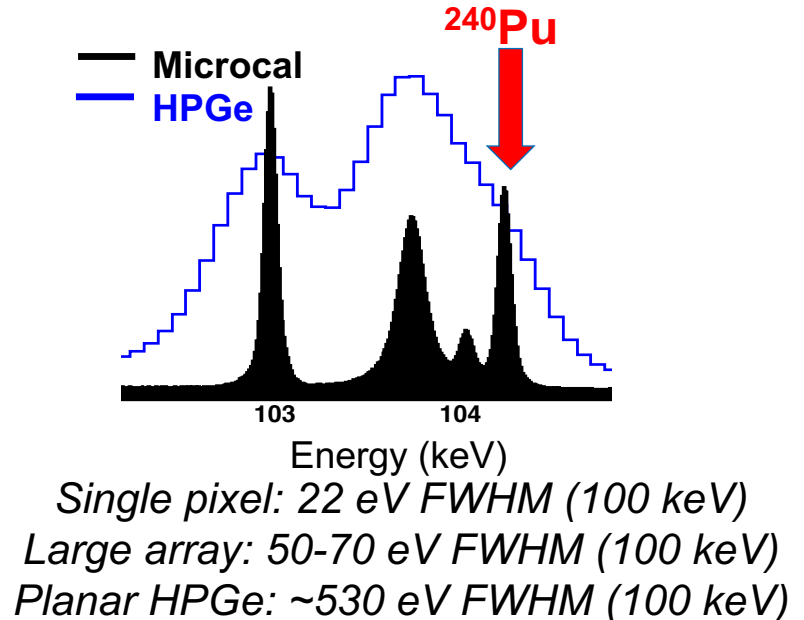
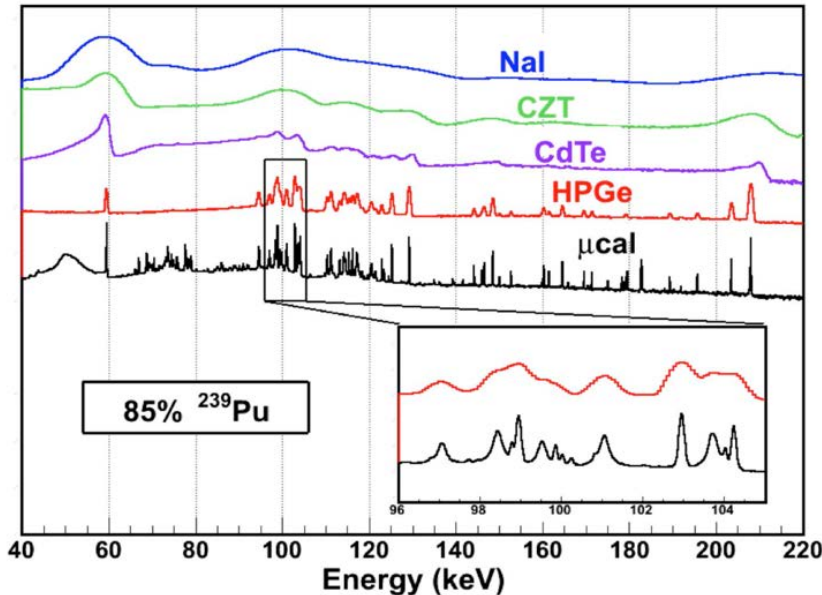


HXI instrument at LANL

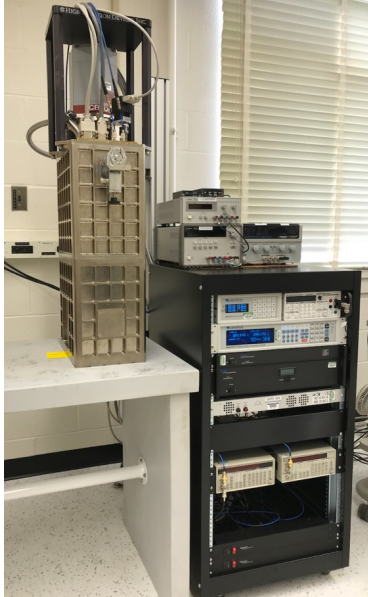


Gamma Spectroscopy

- Overcome uncertainty limits in nondestructive isotopic analysis
- Reduce reliance on sampling, mass spec to improve economics and throughput of safeguards and material accounting measurements (Croce et al. JNMM 2020)
- New determination of gamma ray branching ratios (Yoho et al. NIM A 2020)



This year: first deployed instruments!



SOFIA: TA-55, ORNL
measurement tour

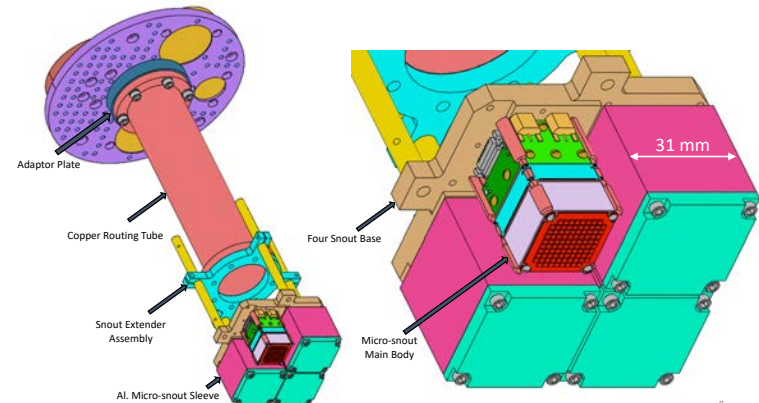
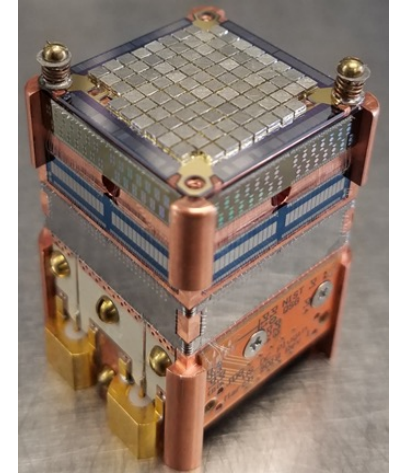


INL Spectrometer to be
permanently installed in
Analytical Laboratory



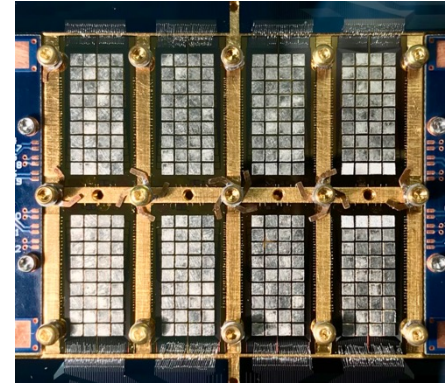
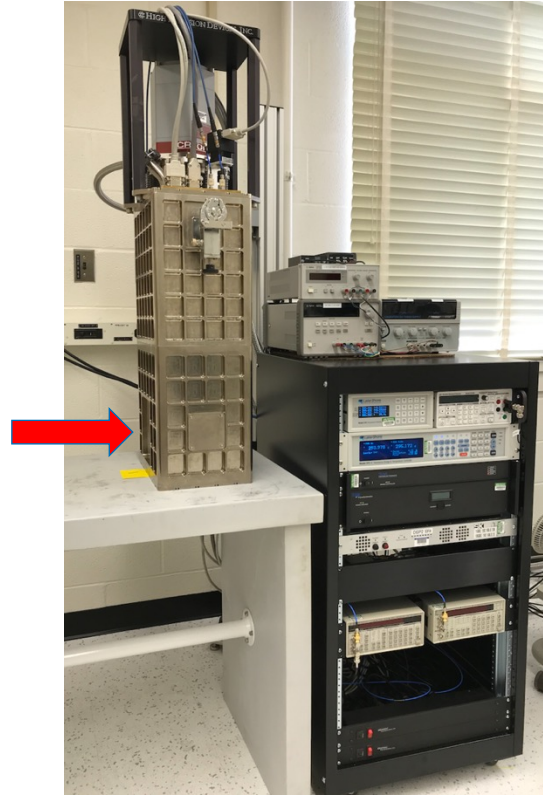
Idaho National Laboratory Spectrometer

- Dilution refrigerator for continuous operation and extremely high counting statistics
- “Microsnout” detector assemblies with ~400 total pixels
- To be installed at INL Materials and Fuels Complex Analytical Laboratory this year

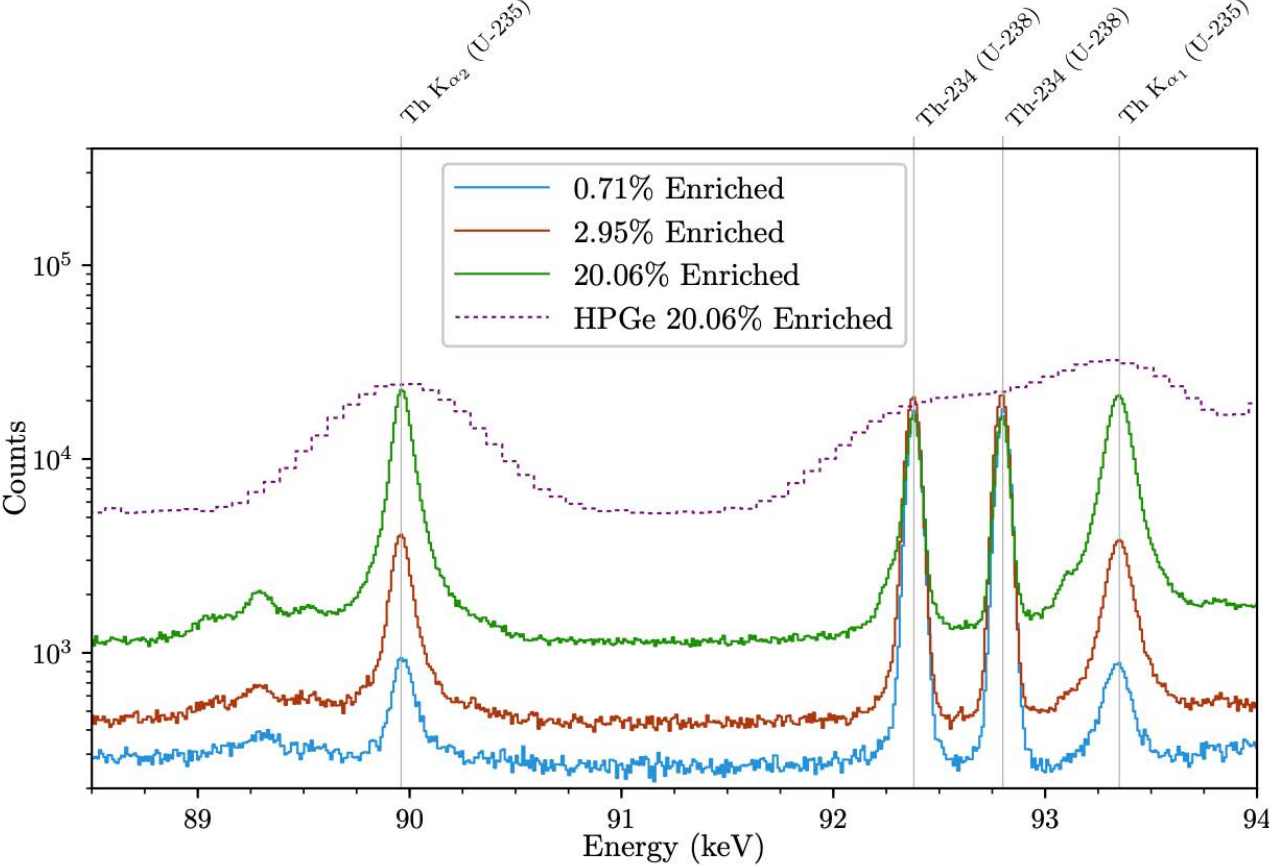


SOFIA: Spectrometer Optimized for Facility Integrated Applications

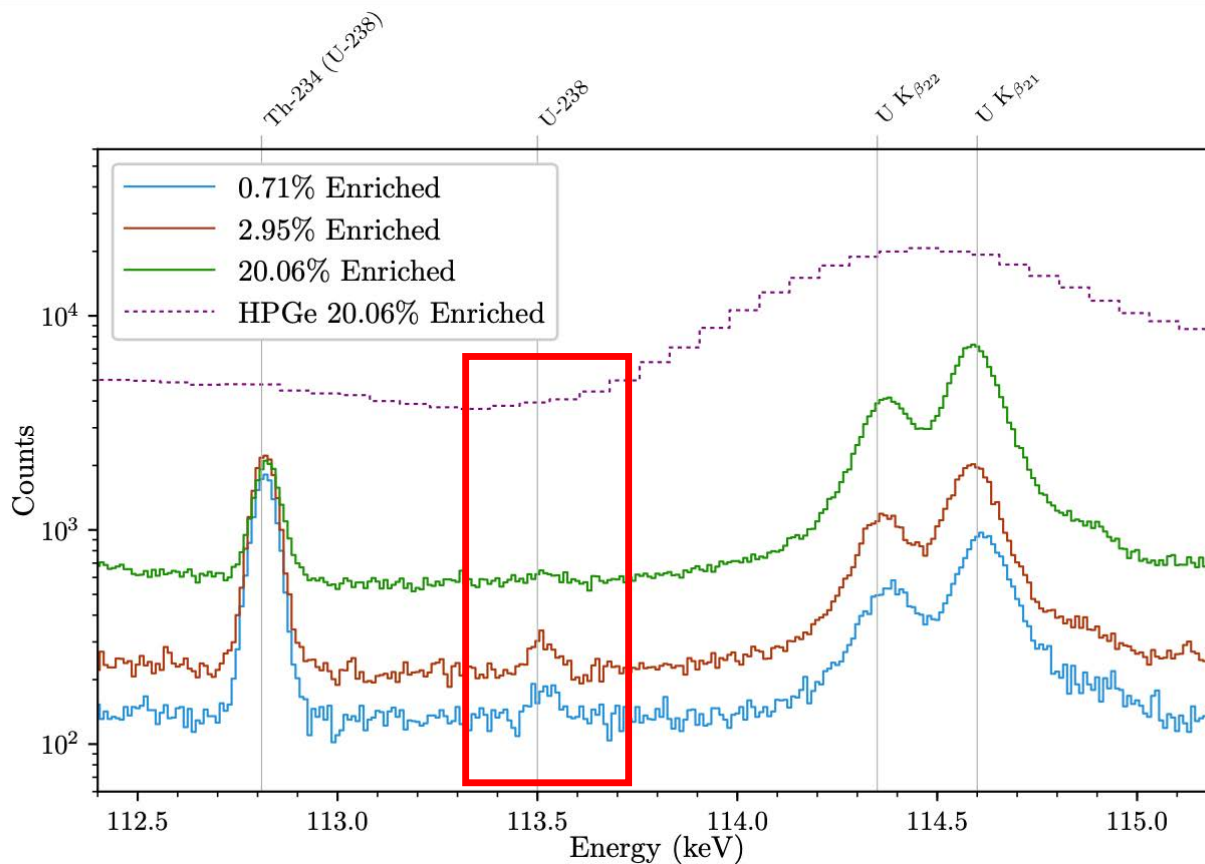
- Samples are placed outside the cryostat, or the instrument can be moved next to a glove box or hot cell
- Power requirements similar to a large window air conditioner
- 256-pixel microwave-multiplexed transition-edge sensor array
- 65 eV FWHM typical resolution at 129 keV
(~8x better than HPGe)
- Capable of > 5000 counts per second



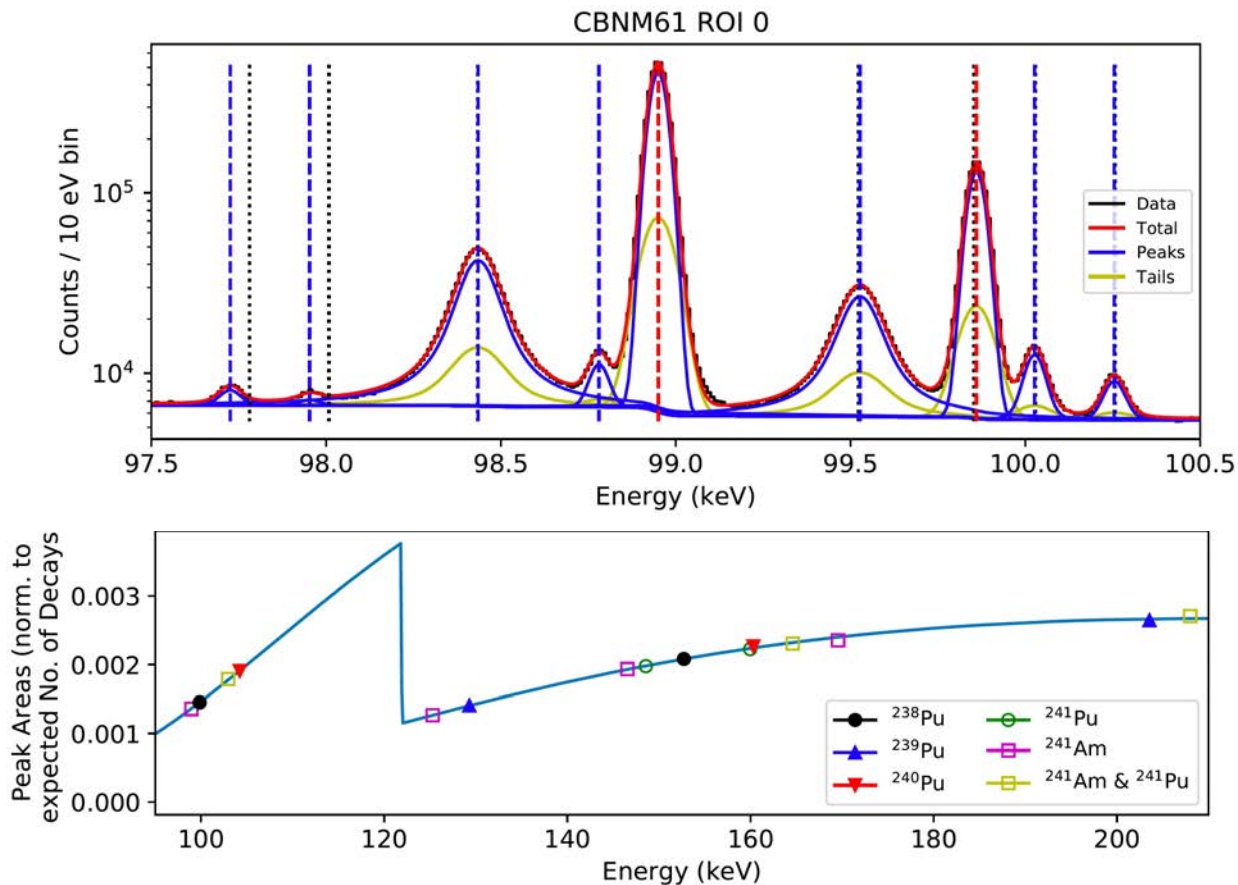
Improving nondestructive measurements of ^{235}U enrichment



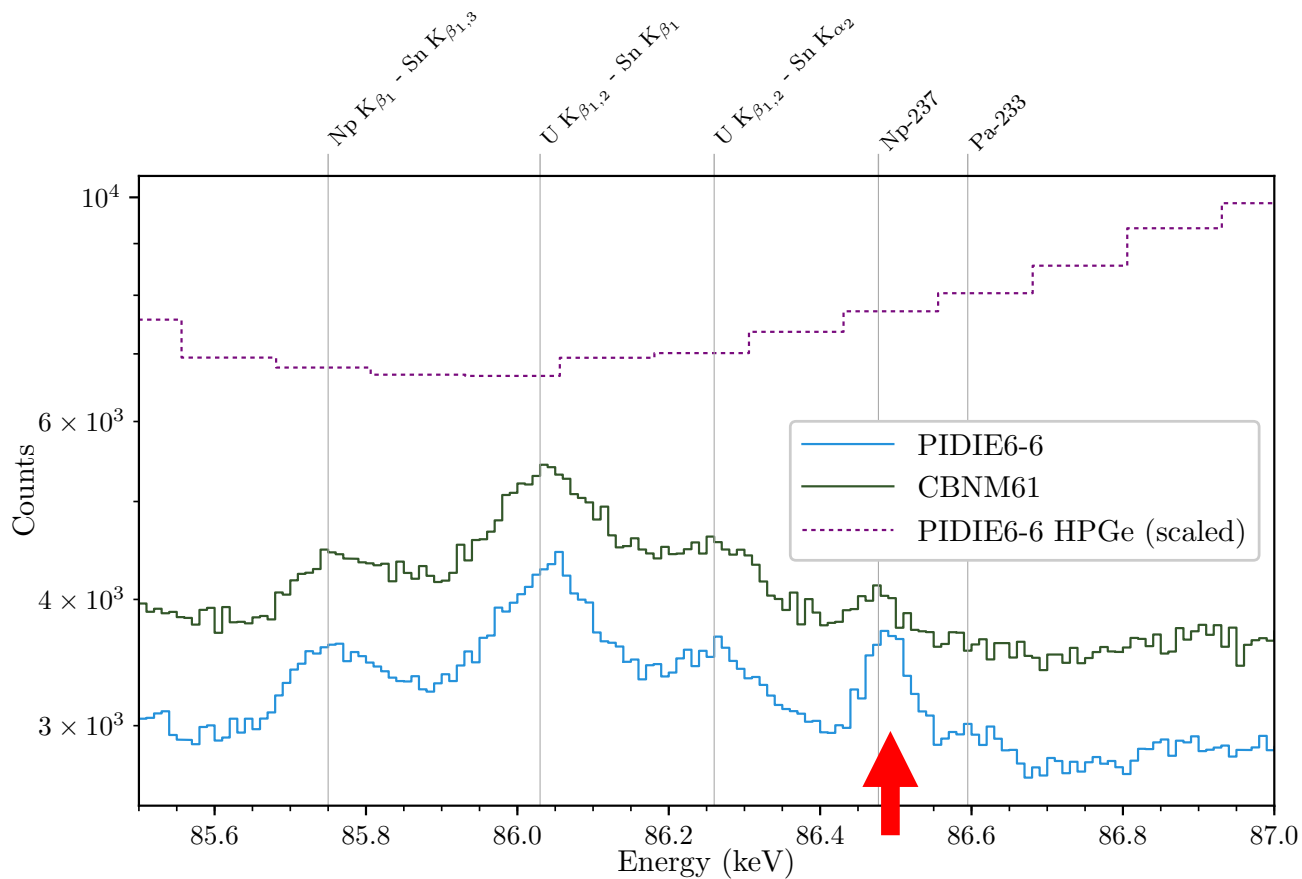
Potential for direct ^{238}U measurement in freshly purified material



Improving nondestructive Pu isotopic analysis

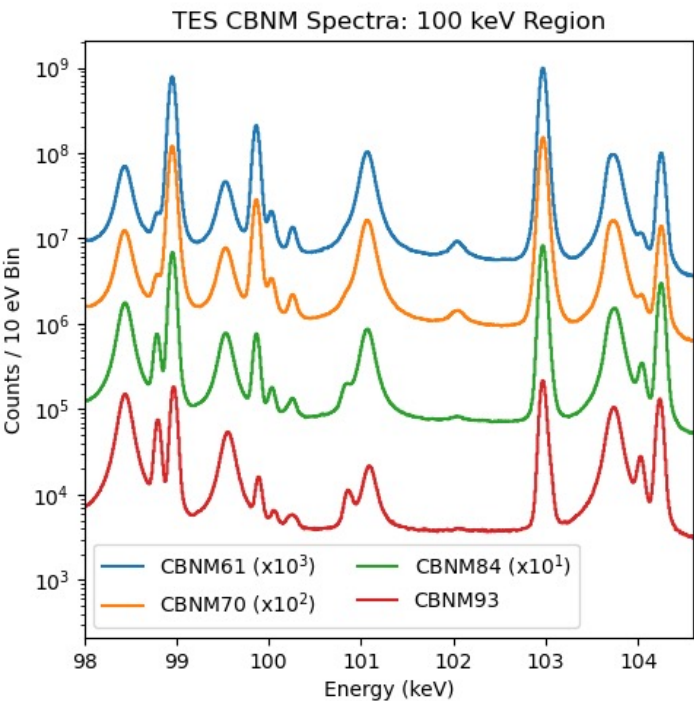


Direct measurement of ^{237}Np in Pu



Quantitative comparison with HPGe for Pu isotopics (in progress)

Example: CBNM84



Isotope	Declared Mass Ratio to Pu239	Measured Mass Ratio to Pu239	Difference	Statistical Uncertainty	Total Uncertainty
Pu238	0.000634	0.000621	-2.05 %	0.48 %	2.04 %
Pu239	1.00000	1.00000	N/A	N/A	N/A
Pu240	0.168	0.168	0.05 %	0.39 %	2.52 %
Pu241	0.00227	0.0228	0.59 %	0.48 %	1.14 %
Am241	0.0120	0.0122	1.30 %	0.36 %	1.52 %

Limited by reference data!



Turn the analysis around to improve the reference data...

M. D. Yoho et al.,
NIM A 2020

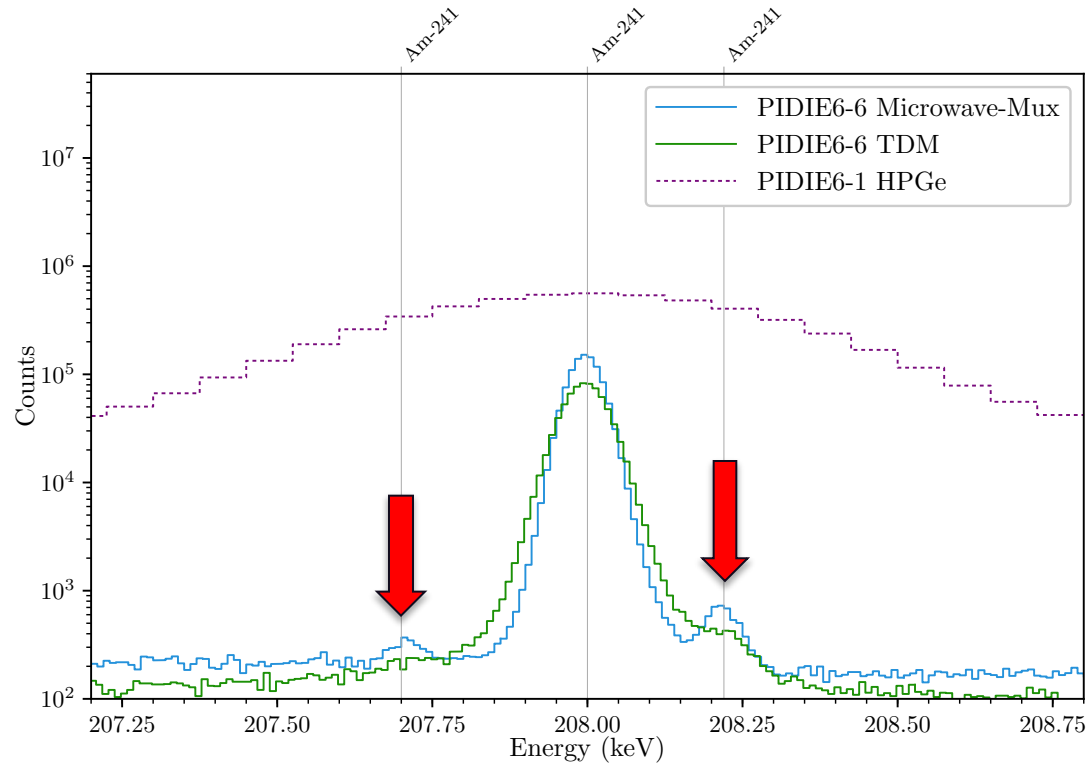
*Greater than 2x
reduction in
uncertainty for five Pu
and Am branching
ratios needed for
isotopic analysis*

Energy [keV]	Isotope	NNDC BR	μ_{BR} [%]	This work BR	μ_{BR} [%]	μ_{BR} Agreement
125.21	²³⁹ Pu	5.63×10^{-5}	2.7	5.51×10^{-5}	13	-0.2
125.3	²⁴¹ Am	4.08×10^{-3}	2.5	4.08×10^{-3}	1.0	0.0
144.201	²³⁹ Pu	2.83×10^{-4}	2.1	2.87×10^{-4}	1.0	0.6
146.094	²³⁹ Pu	1.19×10^{-4}	2.5	1.22×10^{-4}	1.4	0.7
146.55	²⁴¹ Am	4.61×10^{-4}	2.6	4.75×10^{-4}	0.75	1.2
150.04	²⁴¹ Am	7.40×10^{-5}	3.0	7.76×10^{-5}	1.3	1.5
152.72	²³⁸ Pu	9.29×10^{-4}	0.75	9.46×10^{-4}	0.78	1.7
159.955	²⁴¹ Pu	6.68×10^{-6}	1.1	6.87×10^{-6}	2.0	1.2
160.19	²³⁹ Pu	6.20×10^{-6}	19	5.82×10^{-7}	331	-2.5
161.45	²³⁹ Pu	1.23×10^{-4}	1.6	1.20×10^{-4}	1.6	-1.1
161.54	²⁴¹ Am	1.50×10^{-6}	20.0	3.52×10^{-6}	19.9	2.7
164.61	²⁴¹ Pu	4.56×10^{-5}	1.6	4.46×10^{-5}	2.0	-0.9
164.69	²⁴¹ Am	6.67×10^{-5}	3.7	7.78×10^{-5}	4.9	2.4
169.56	²⁴¹ Am	1.73×10^{-4}	2.3	1.72×10^{-4}	0.9	-0.3
171.393	²³⁹ Pu	1.10×10^{-4}	1.8	1.12×10^{-4}	1.4	0.9
175.07	²⁴¹ Am	1.82×10^{-5}	5.5	1.85×10^{-5}	2.8	0.3
188.23	²³⁹ Pu	1.09×10^{-5}	10	8.63×10^{-6}	10.8	-1.6
189.36	²³⁹ Pu	8.30×10^{-5}	1.2	7.91×10^{-5}	1.4	-2.6
191.96	²⁴¹ Am	2.16×10^{-5}	4.6	2.01×10^{-5}	2.8	-1.3
208.005	²⁴¹ Pu	5.19×10^{-4}	1.4	5.34×10^{-4}	1.9	1.2
208.01	²⁴¹ Am	7.91×10^{-4}	2.4	8.08×10^{-4}	5.4	0.4

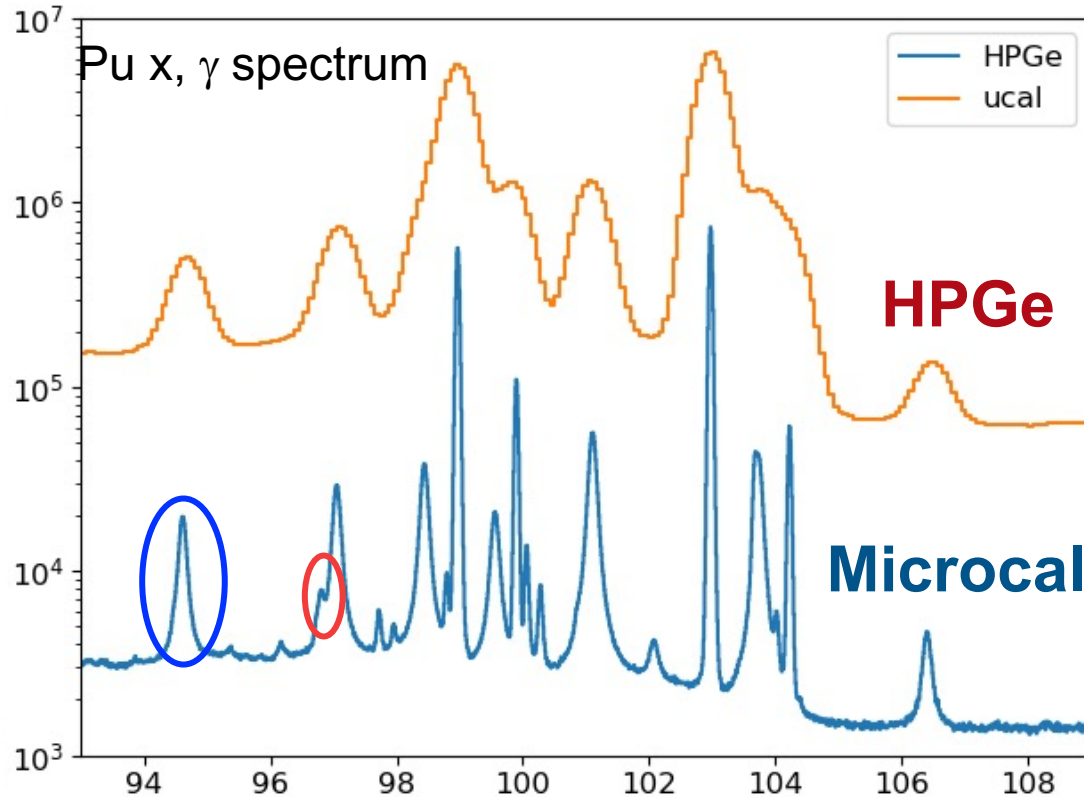


New observation of two ^{241}Am peaks near 208 keV

K E. Koehler et al.,
arXiv:2103.15893

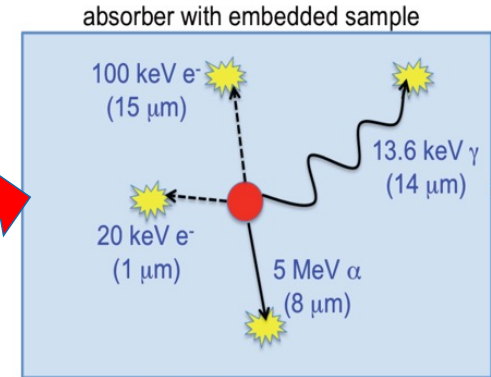
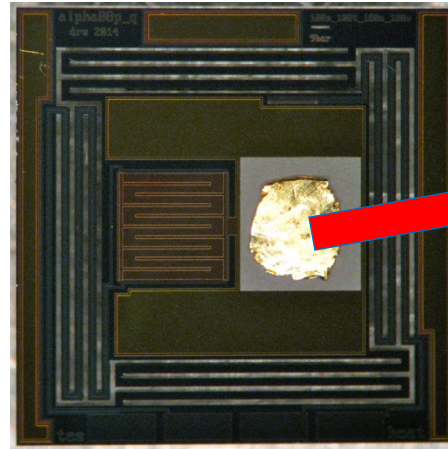
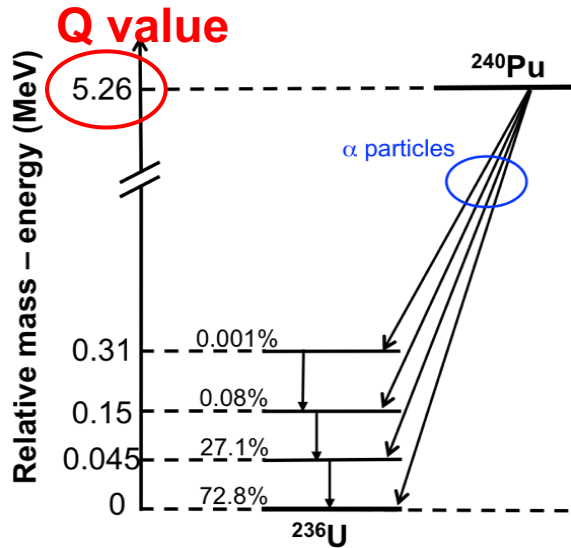


Now investigating X-ray line widths, additional branching ratios, peak energies

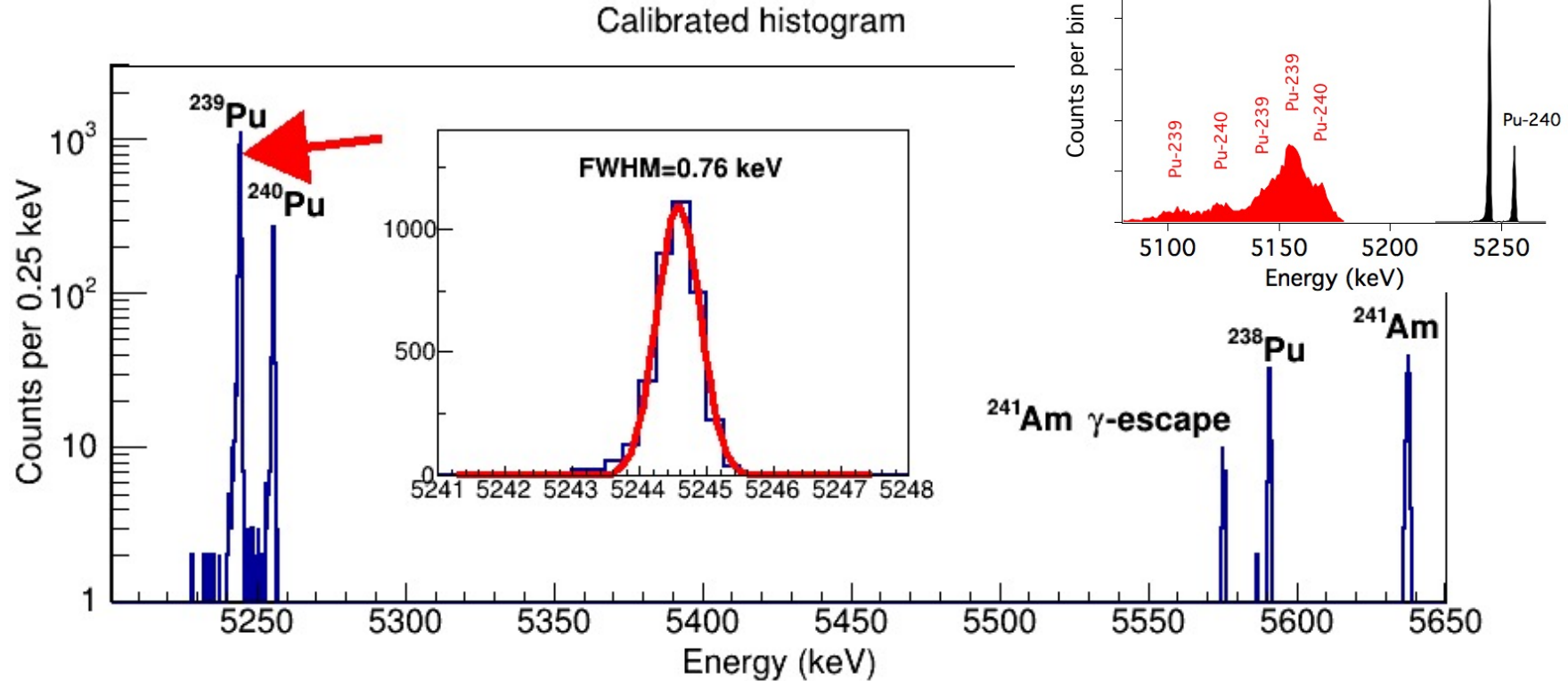


Decay Energy Spectroscopy

- Radiometric measurement with sensitivity and precision to complement mass spectrometry
- Under evaluation for safeguards, certified reference materials, medical isotope production, nuclear forensics, basic nuclear physics

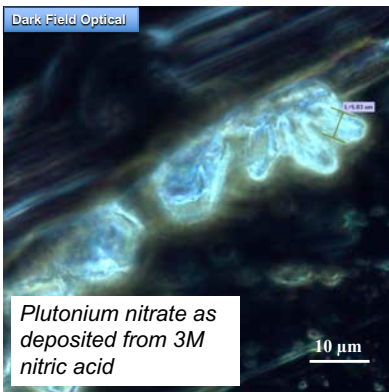


>10x better than limit of Si alpha detectors 0.76 keV FWHM at 5.2 MeV

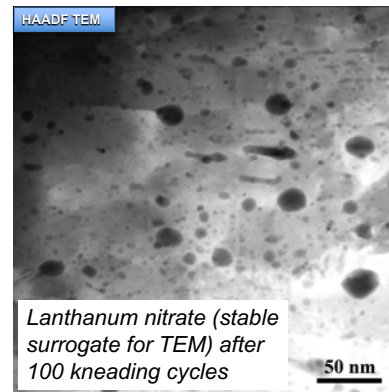


An unusually direct measurement of energy transfer

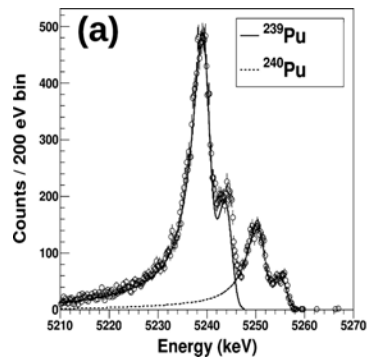
A. S. Hoover et al., Analytical Chemistry 2013



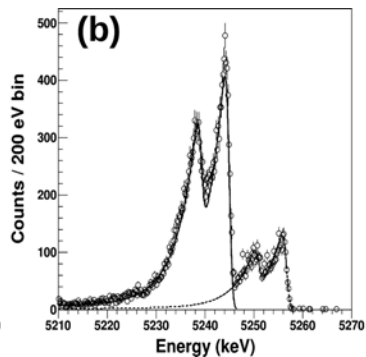
Spectral artifacts nearly eliminated by “kneading” of absorber to fracture crystals in plutonium nitrate deposit



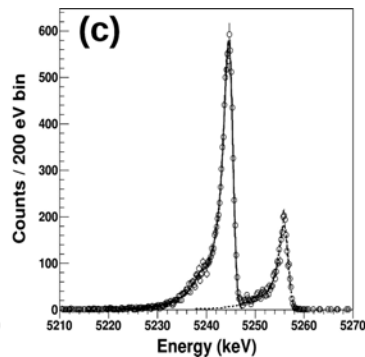
As-deposited Pu nitrate in folded gold foil absorber



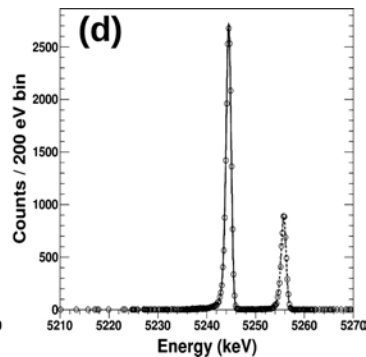
Kneaded for 3 cycles



Kneaded for 6 cycles



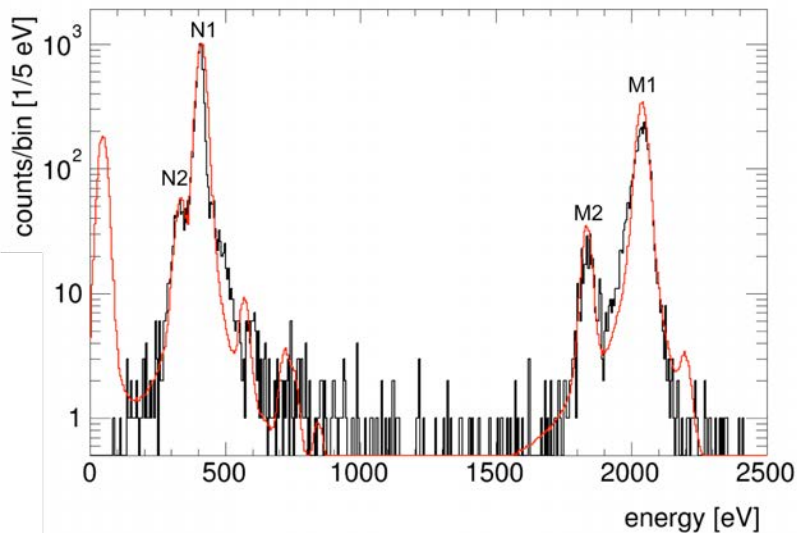
Kneaded for 100 cycles



Electron capture spectroscopy for neutrino mass

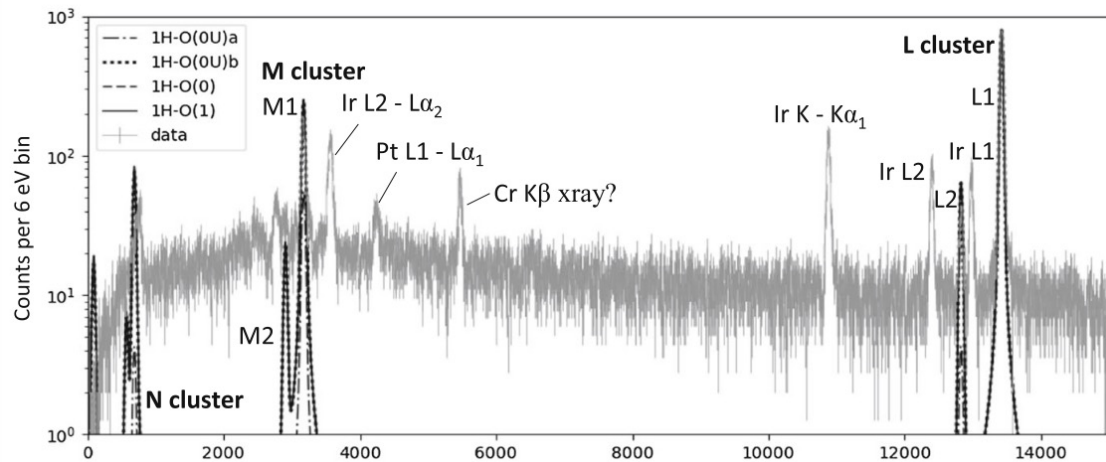
M. P. Croce et al., JLTP. 2016.

^{163}Ho electron capture



K.E. Koehler, JLTP. 2018.

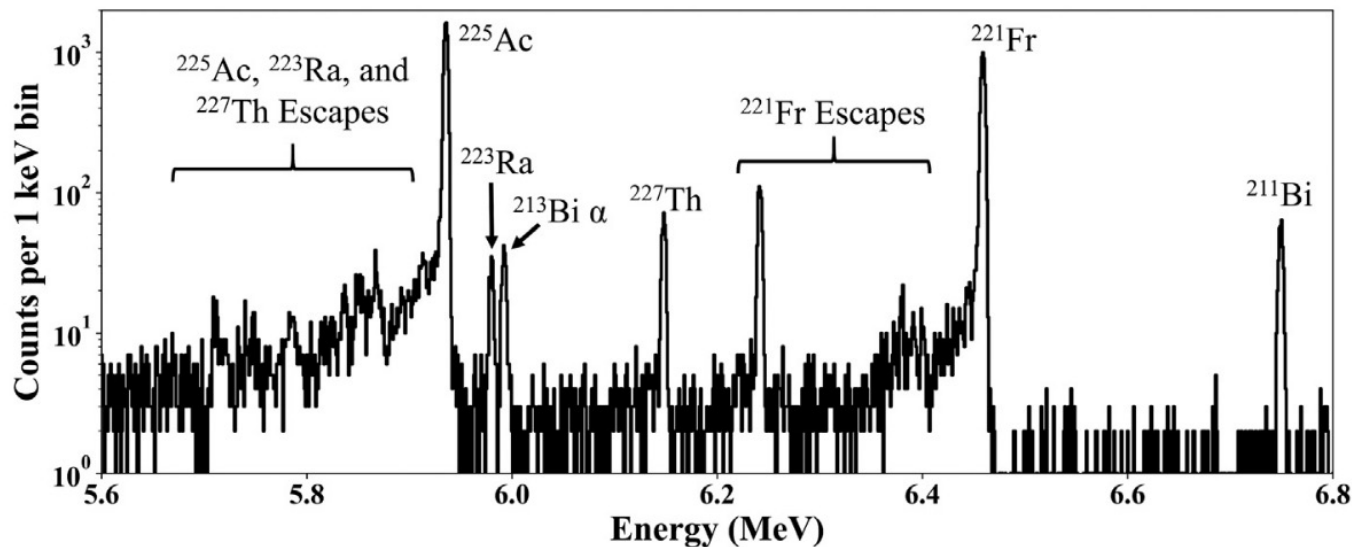
^{193}Pt electron capture



^{225}Ac characterization for targeted alpha therapy

A.D. Tollefson et. al., Applied Radiation and Isotopes (2021)

Accurately quantified end of bombardment $^{227}\text{Ac}/^{225}\text{Ac}$ activity ratio



^{225}Ac production sample spectrum with clear indication of ^{227}Ac impurity visible from ^{223}Ra , ^{227}Th , and ^{211}Bi daughters



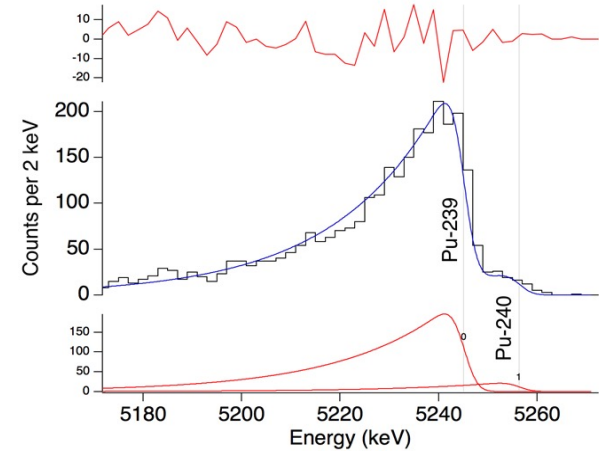
Pu in Trinitite

M.P. Croce et. al, WRL Trinity Issue (2020), arxiv 2103.12215

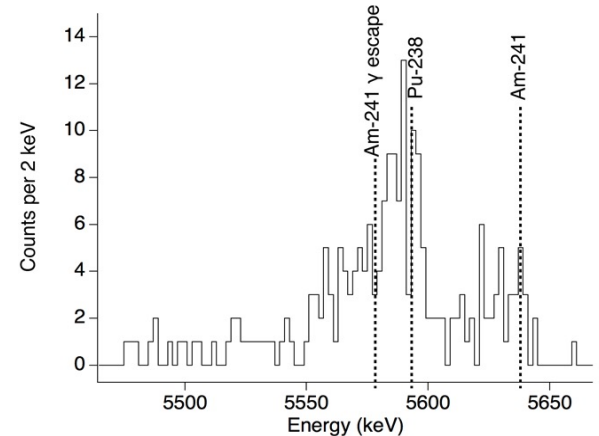
DES was able to identify ^{238}Pu , ^{239}Pu , ^{240}Pu , and ^{241}Am and obtain a decay-corrected $^{240}\text{Pu}/^{239}\text{Pu}$ mass ratio of 0.03 in a trinitite bead



Top: Peaks from ^{239}Pu and ^{240}Pu observed in trinitite. The middle plot shows the fitted spectrum, the top plot shows residuals, and the bottom plot shows the fit components.

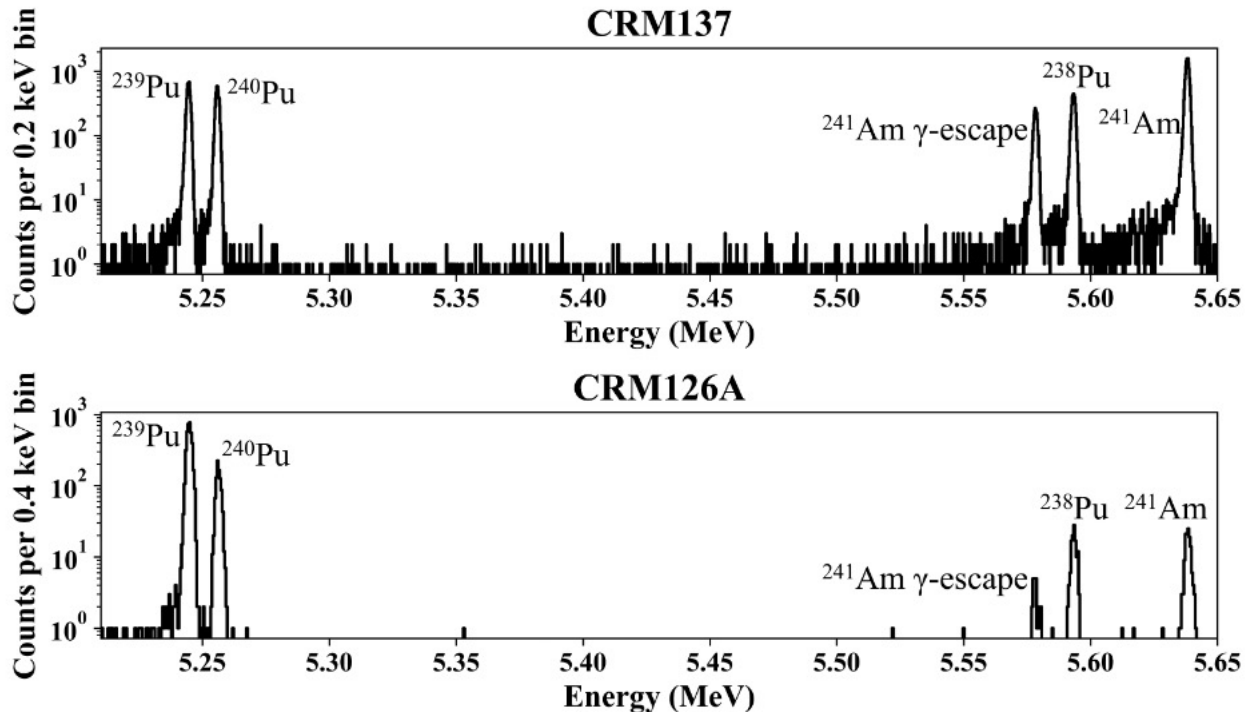


Bottom: The presence of ^{238}Pu and ^{241}Am are also identified. Dashed lines indicate the expected peak locations



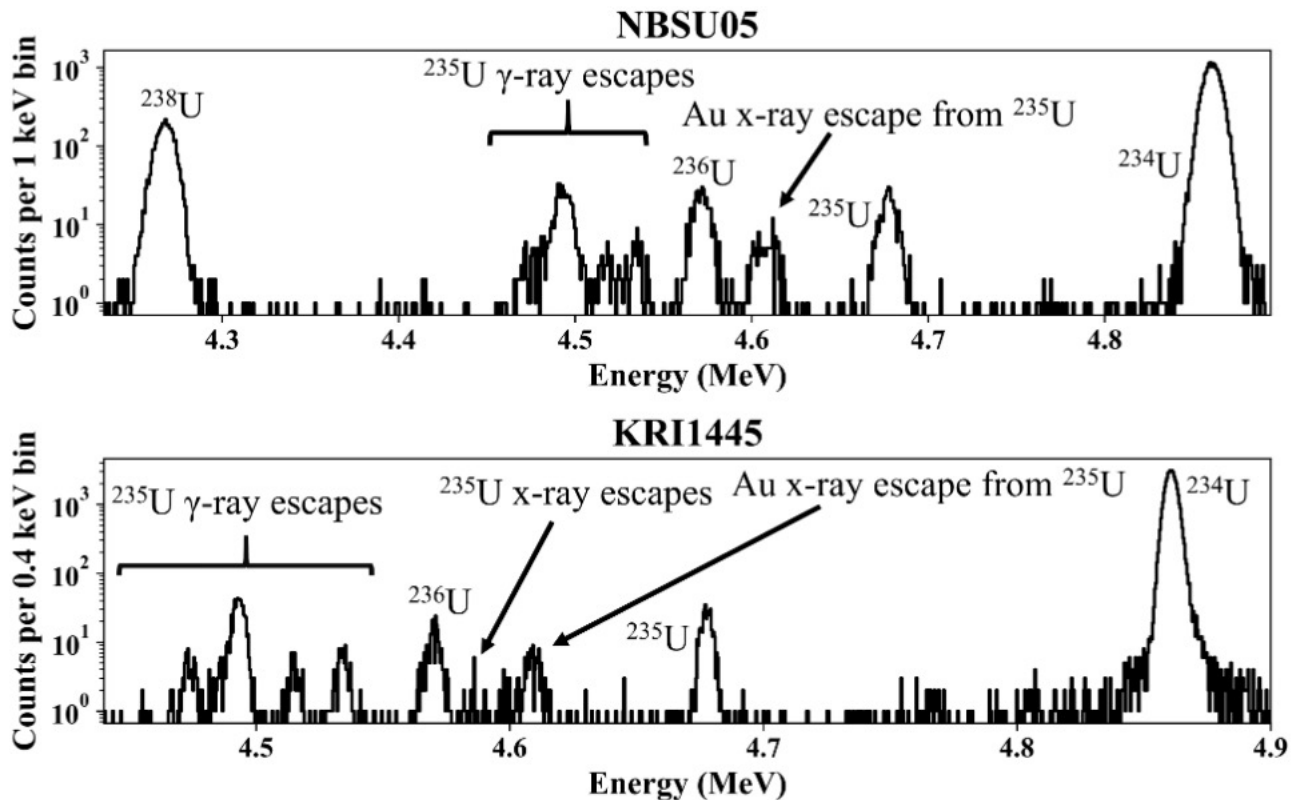
Plutonium – quantitative analysis under evaluation

C. M. Smith et. al., LA-UR-20-23448



Uranium – quantitative analysis under evaluation

C. M. Smith et. al., LA-UR-20-23448



Acknowledgements

Much of this work is done in collaboration with University of Colorado and the National Institute of Standards and Technology

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- LANL Laboratory Directed Research and Development (LDRD)
- LANL Technology Evaluation and Demonstration Program (TED)
- DOE Office of Nuclear Energy MPACT, NEUP, and ARS programs
- DOE NNSA Office of International Nuclear Safeguards
- Strategic Partnership Programs with Idaho and Sandia National Laboratories



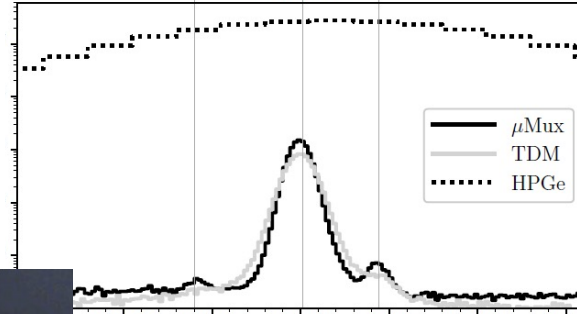
An exciting time for low temperature detectors!

Contact: Mark Croce
mpcroce@lanl.gov

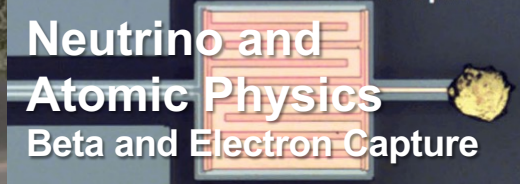
Nuclear Safeguards and Metrology

Nuclear forensics

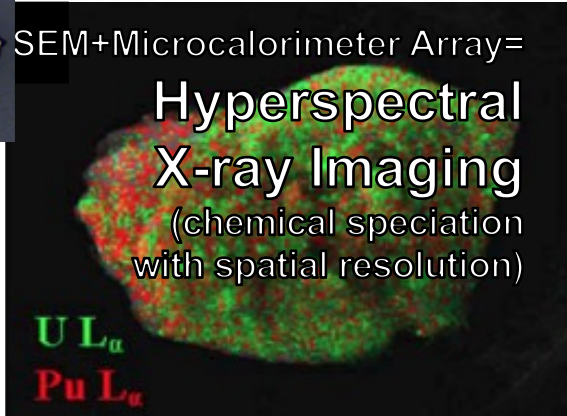
- Treaty verification
- Environmental monitoring



Neutrino and Atomic Physics
Beta and Electron Capture



SEM+Microcalorimeter Array=
Hyperspectral X-ray Imaging
(chemical speciation with spatial resolution)



Cancer Research
(quantification of impurities)

