β-Decay Measurements of Neutron Rich Nuclei at the Holifield Radioactive Ion Beam Facility (HRIBF)

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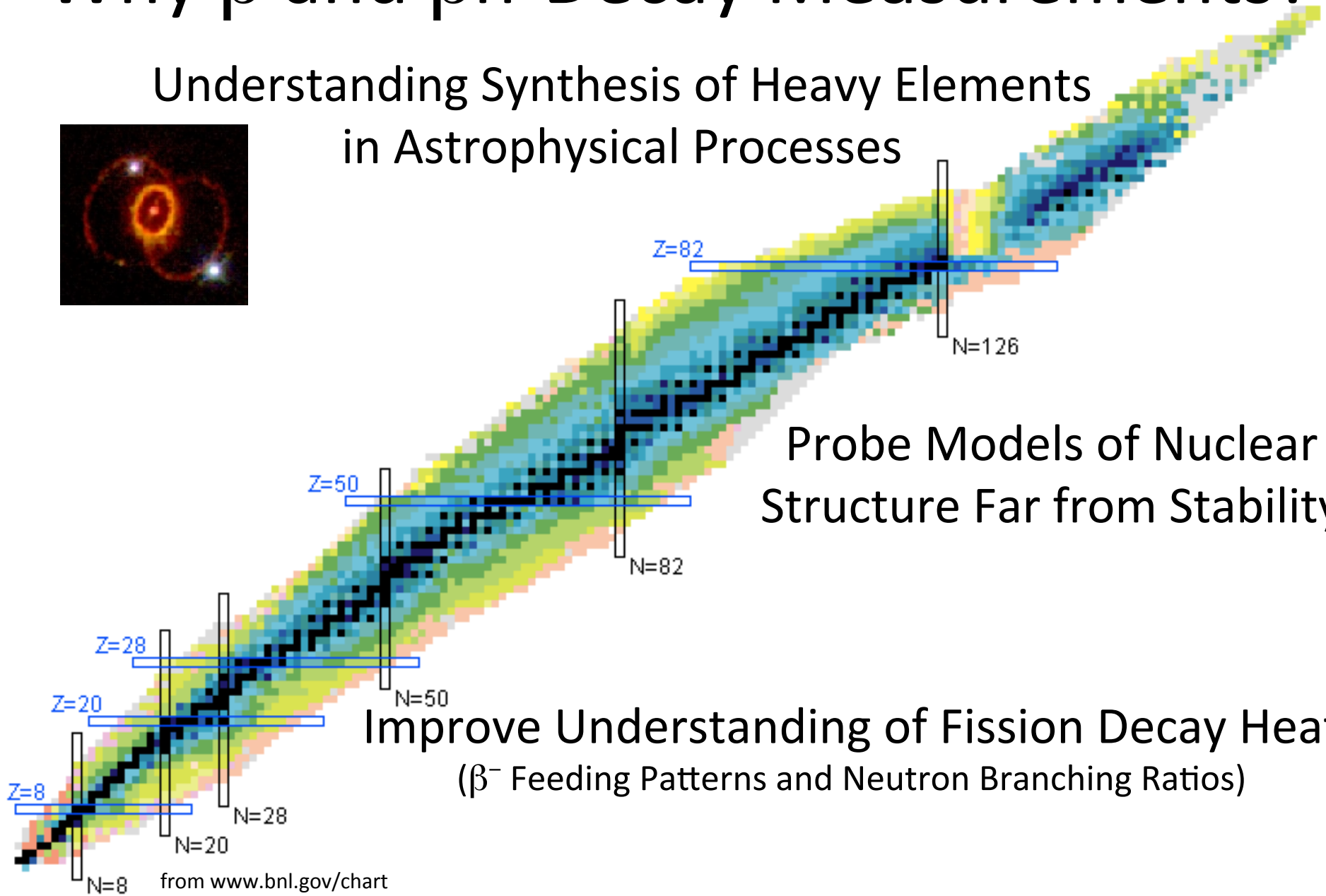
Louisiana State University

RIKEN June 12, 2012

Why β and βn-Decay Measurements?

Understanding Synthesis of Heavy Elements in Astrophysical Processes



Probe Models of Nuclear Structure Far from Stability

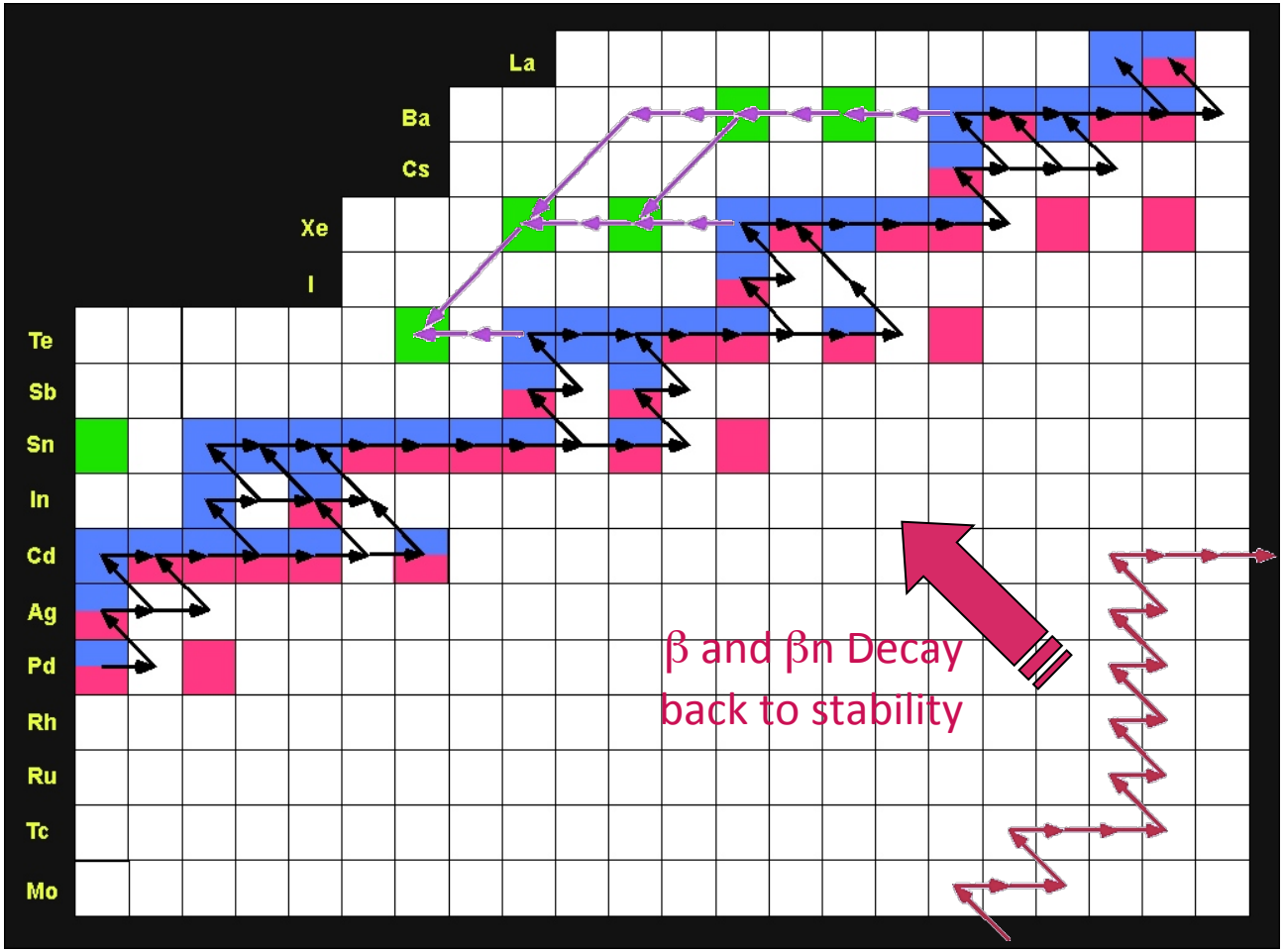
Improve Understanding of Fission Decay Heat

(β^- Feeding Patterns and Neutron Branching Ratios)

from www.bnl.gov/chart

Origin of the Elements

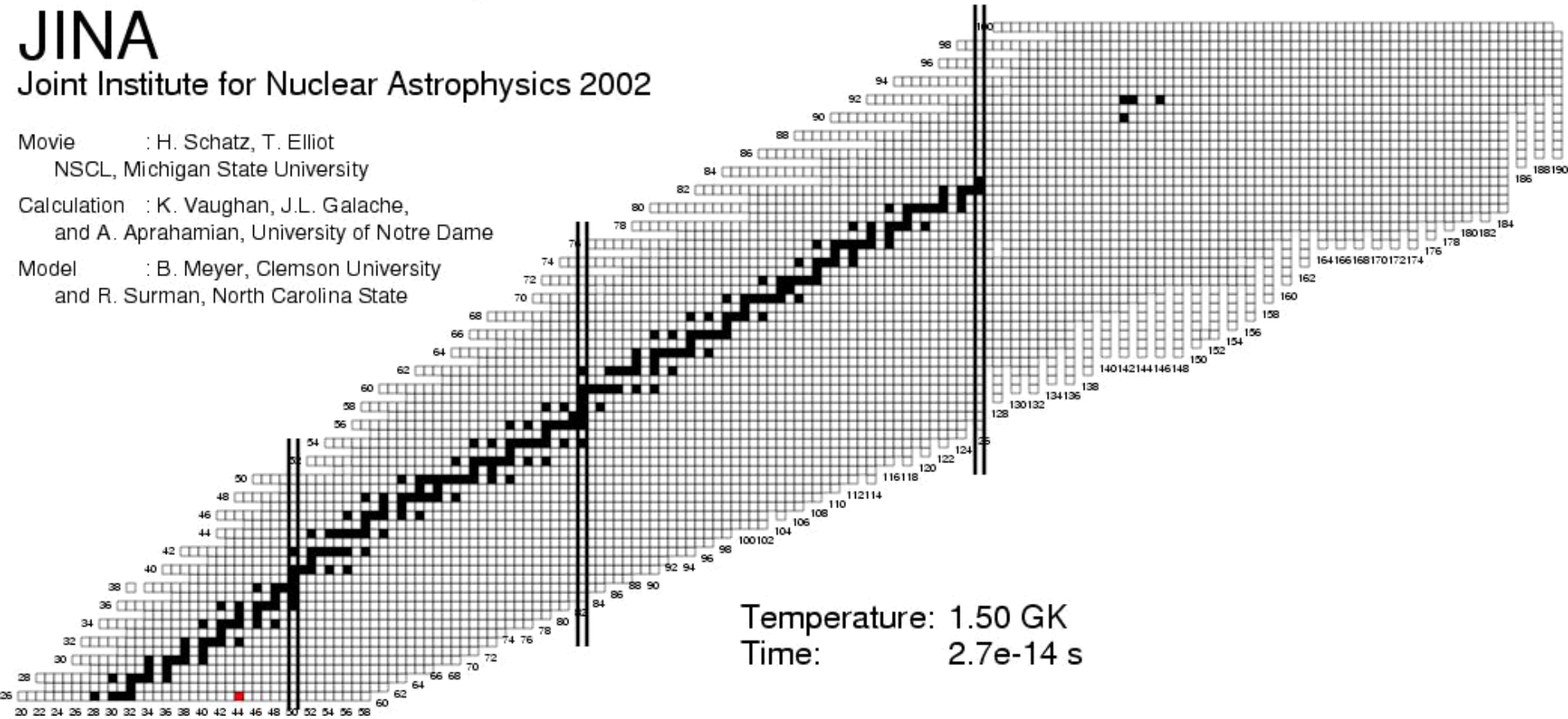
It is believed that there are three main types of processes that account for the observed elements in the universe



- s-Process
  - ~80% of Isotopes
  - (n,γ) MACs Needed
  - Occurs in Asymptotic Giant Branch (AGB) Stars
  - Branch Points Crucial
- r-Process
  - ~70% of Isotopes
  - Far from Stability
  - Occurs Where?
- p-Process
  - ~10% of Isotopes
  - Secondary Process

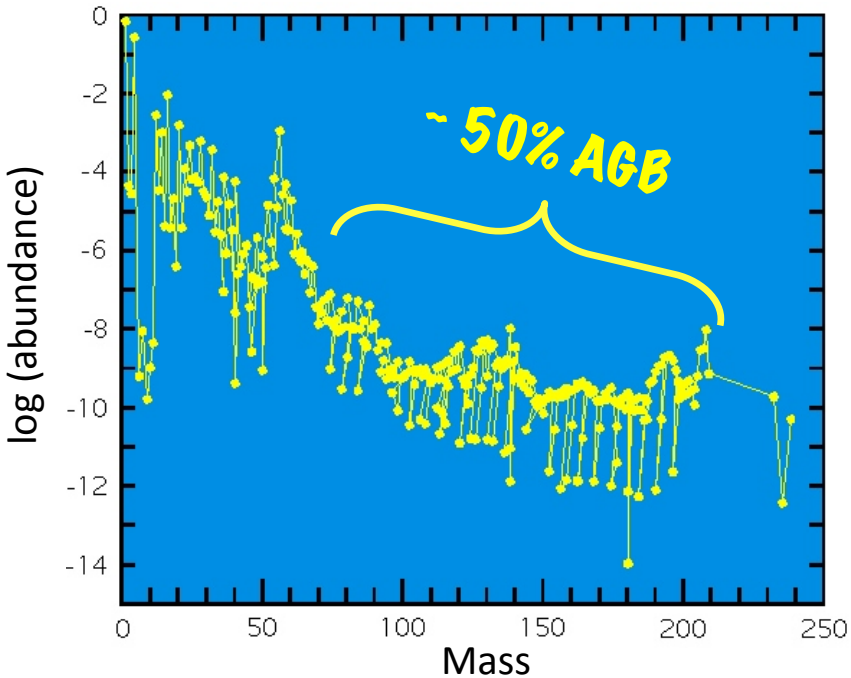
r-Process Example

Nucleosynthesis in the r-process



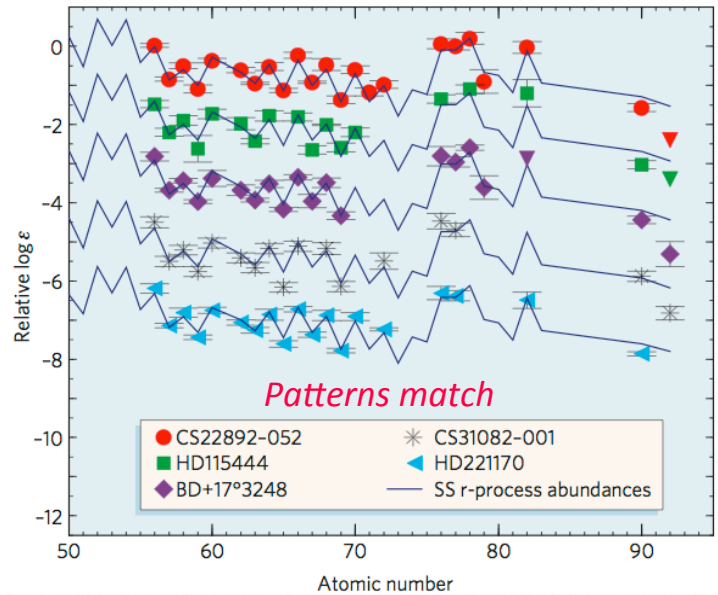
$T_{1/2}$, Nuclear Masses, and P_n (Neutron Emission Probability) Most Important to Measure in Order to Predict Final Abundances of Elements Generated with the r-Process

Origin of the Elements

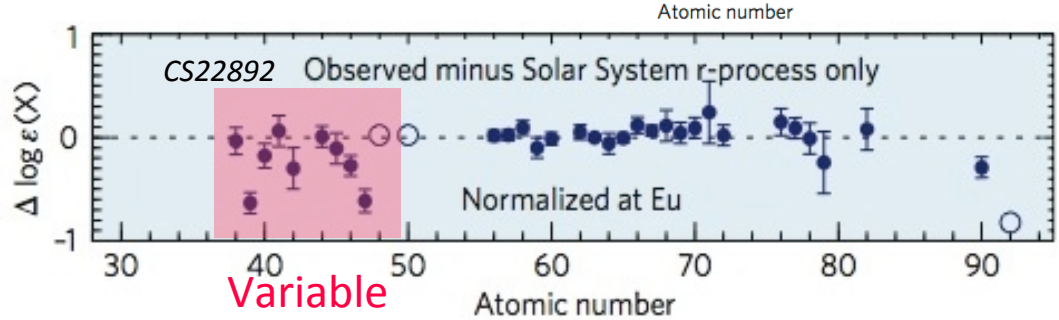


- Changing paradigm
  → Spectra from old halo stars

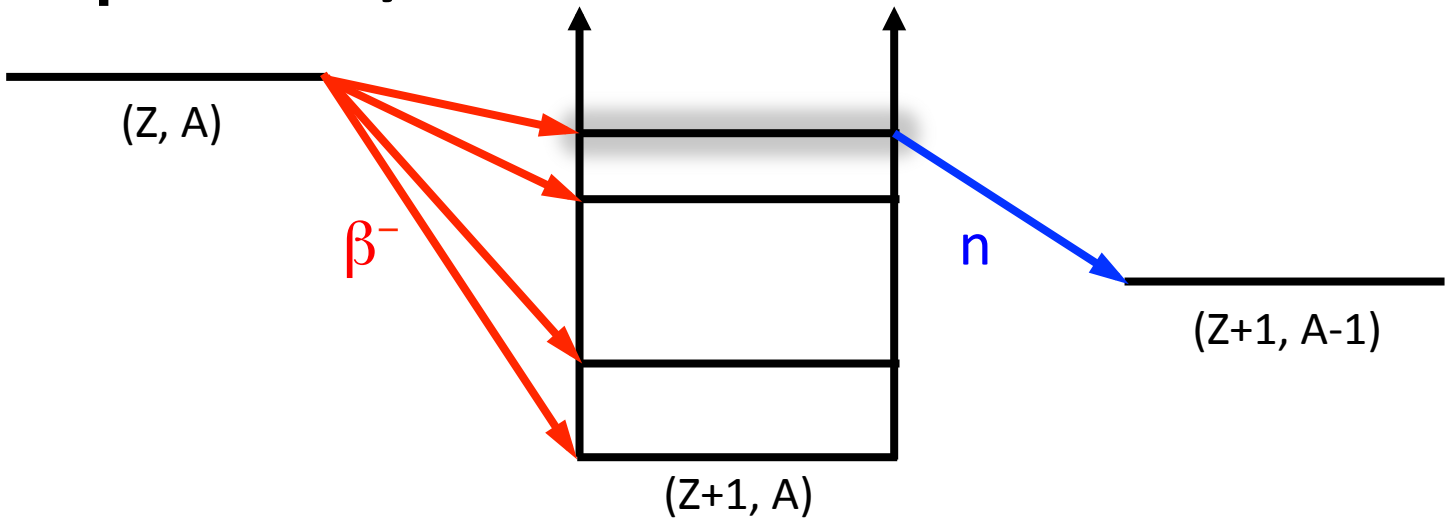
Cowan & Sneden, Nature (2006).



- Universal abundance pattern for heavy elements
  →r process
- Light elements vary
  → Other process?
  → Light Element Primary Process



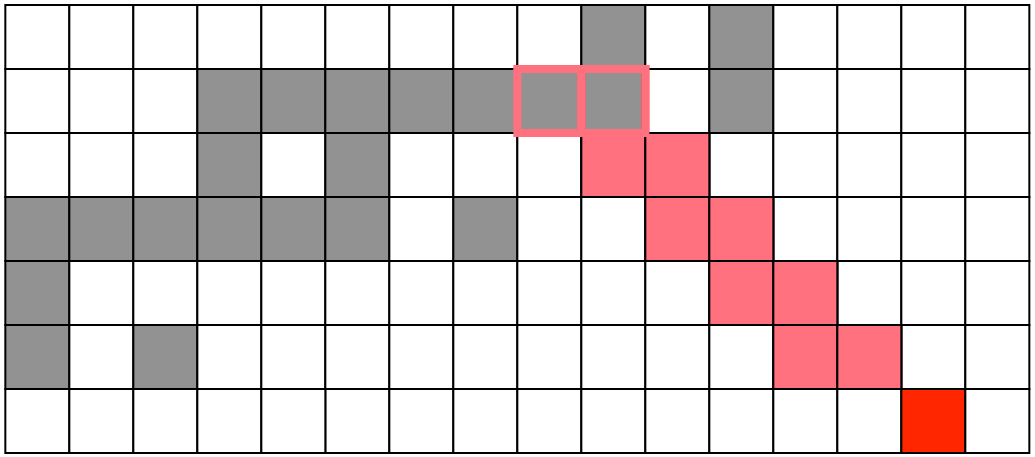
β Delayed Neutron Emission



Beta decay proceeds to a wide range of states in the daughter nucleus

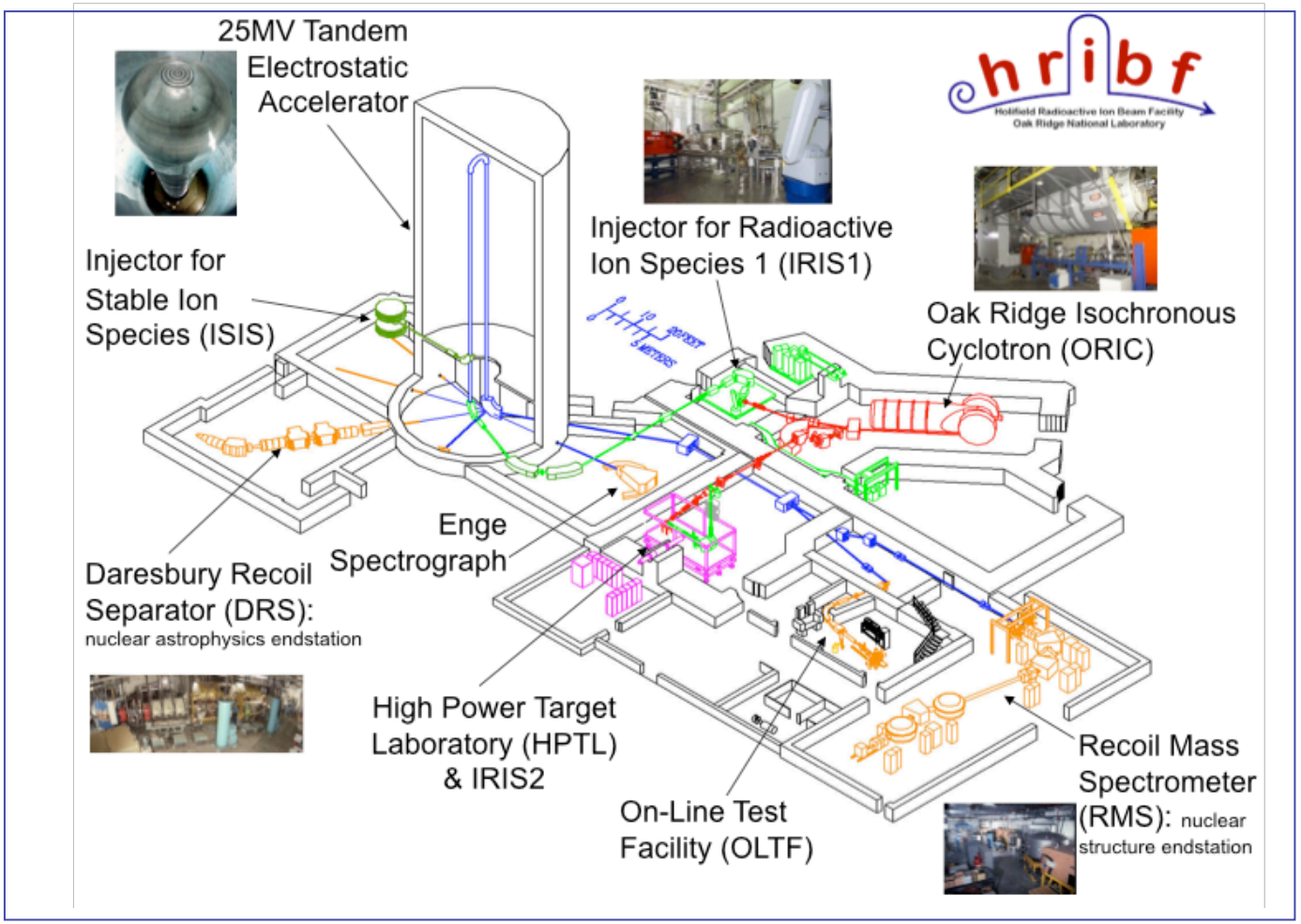
Lower energy states are favored due to Fermi factor, but the nuclear matrix element plays an equally important role

Neutron emission changes the final abundance in a given mass chain and replenishes neutron density of environment



Holifield Radioactive Ion Beam Facility(HRIBF) at Oak Ridge National Lab (ORNL)

Unfortunately the HRIBF has Ceased Operations
Further Studies at Argonne National Lab in the US and TRIUMF in Canada are Possible



Useful Detectors Used in β Decay Studies

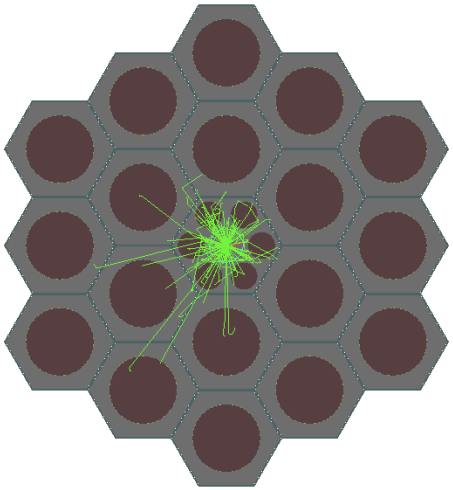
Gamma Detector Limitations: **Low Efficiency**, Poor Energy Resolution (NaI)

Neutron Detector Limitations: Low Efficiency, Poor Energy Resolution, γ Discrimination

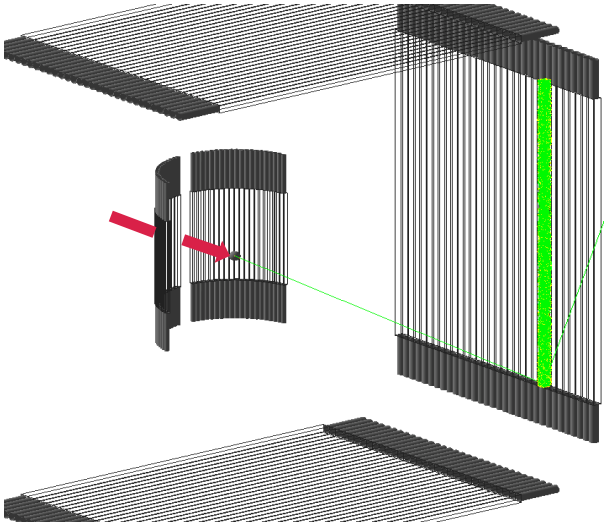
Electron Detector Limitations: Energy Threshold, γ Discrimination

Three New Detectors at ORNL Address these Detector Limitations

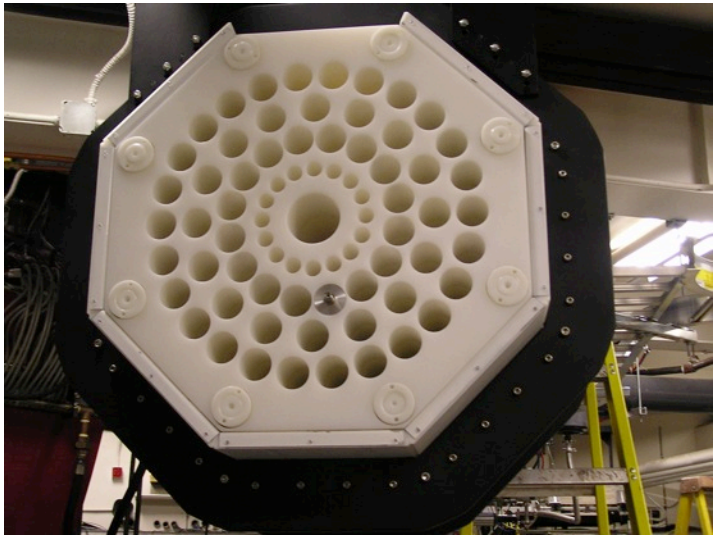
MTAS



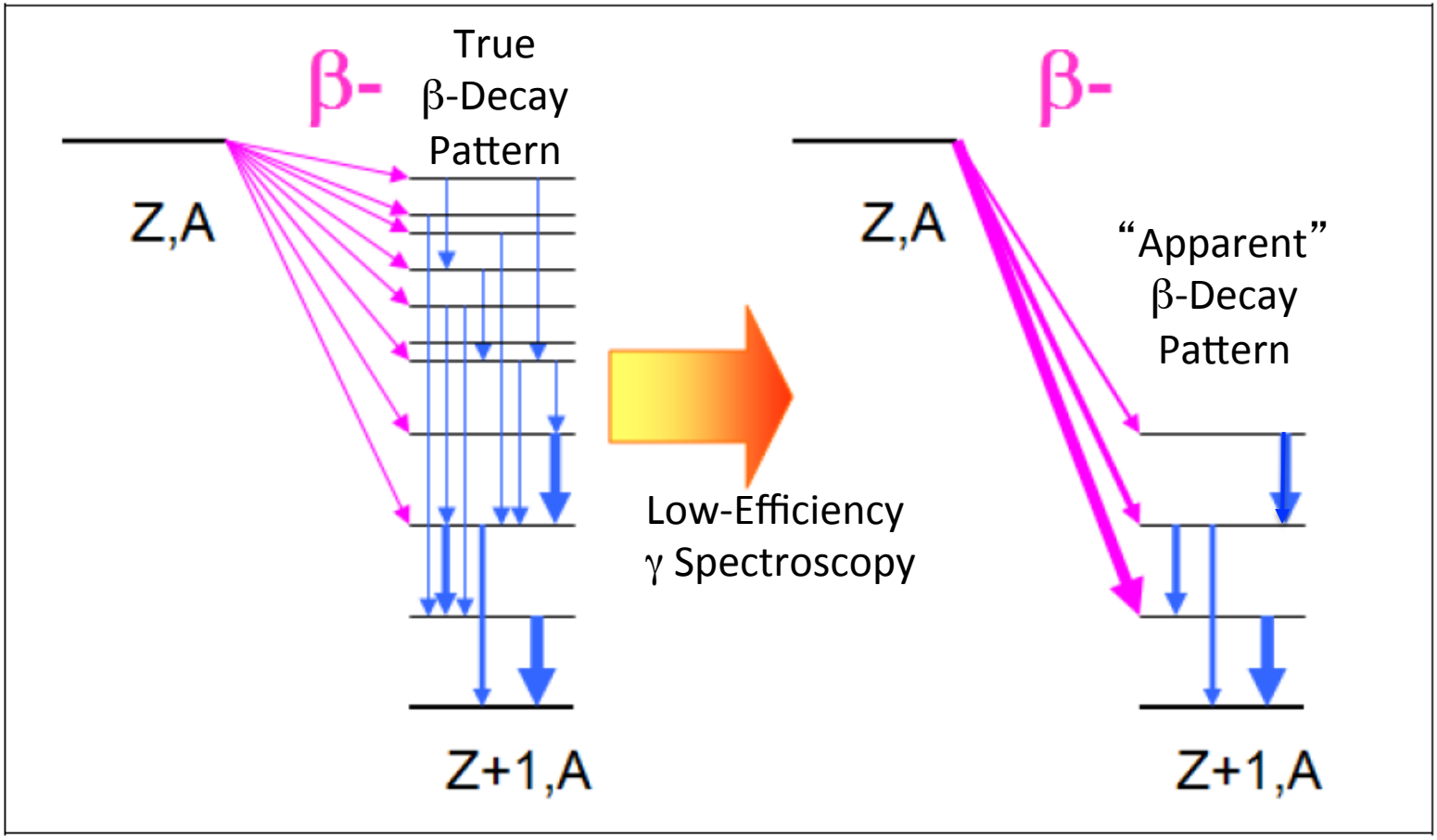
VANDLE



3Hen



The Pandemonium Effect

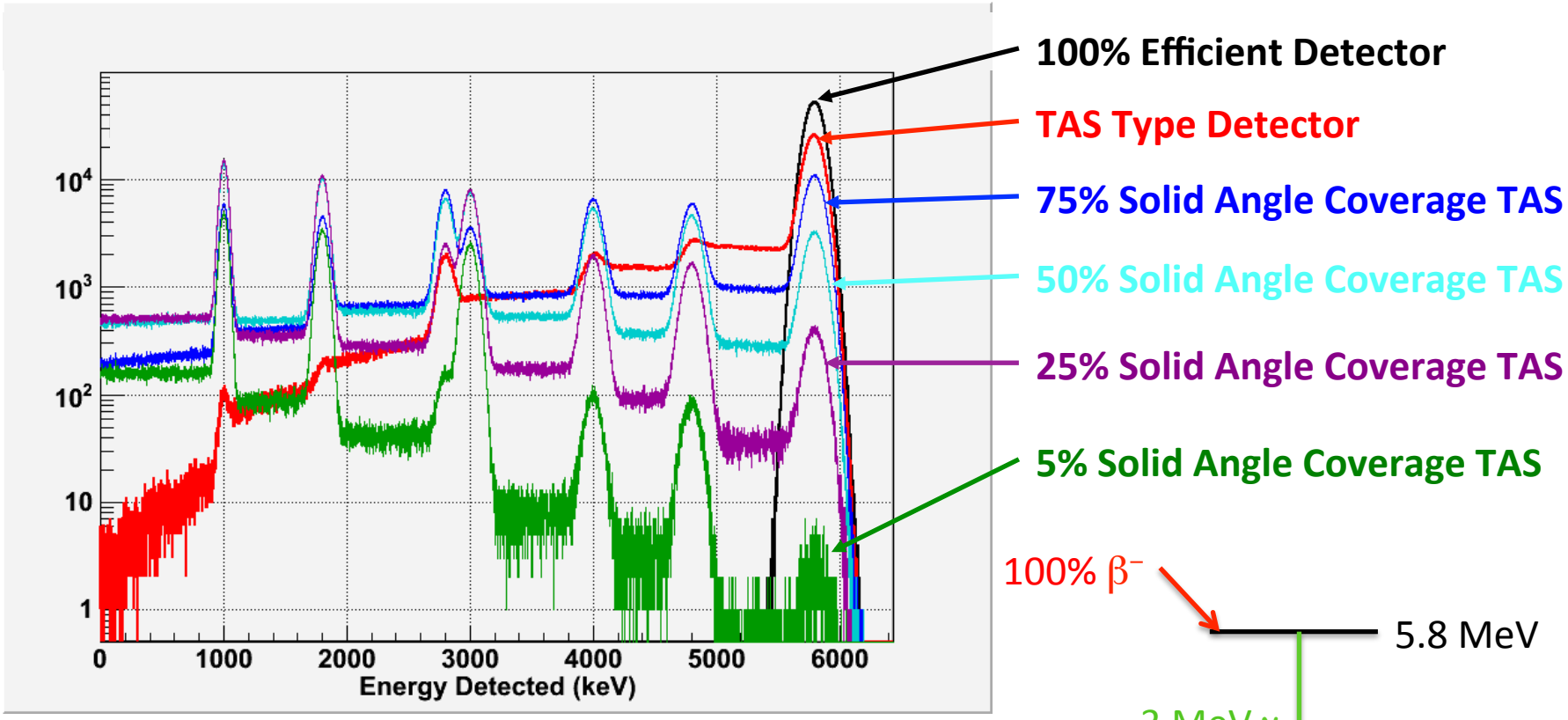


A.A. Sonzogni and M. Herman, NNDC 2011

Adopting "Apparent" decay pattern results in a wrong interpretation of β decay properties and respective nuclear structure. The energies and intensities of emitted γ-rays are underestimated while β-electron and neutrino energies are over estimated.

Total Absorption Spectrometry (TAS) is the Solution!

Illustration of the Pandemonium Effect



Simulated 3 γ Decay from a Hypothetical 5.8 MeV Level

(E_γ=1 MeV, 1.8 MeV, and 3 MeV)

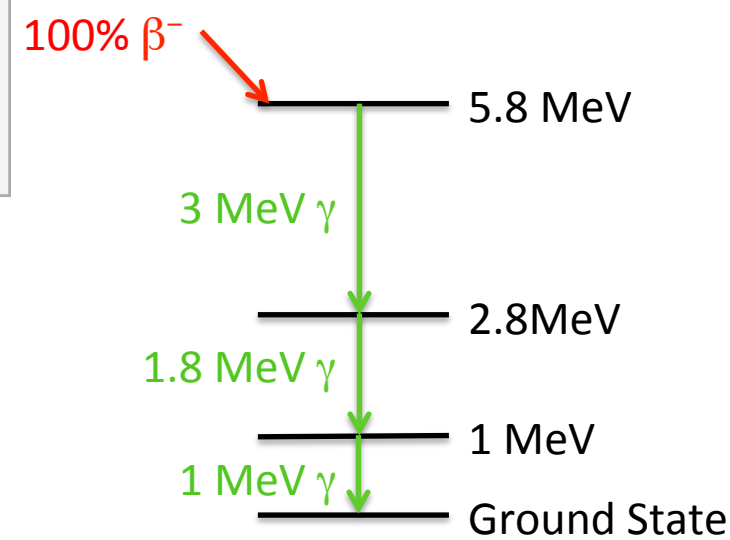
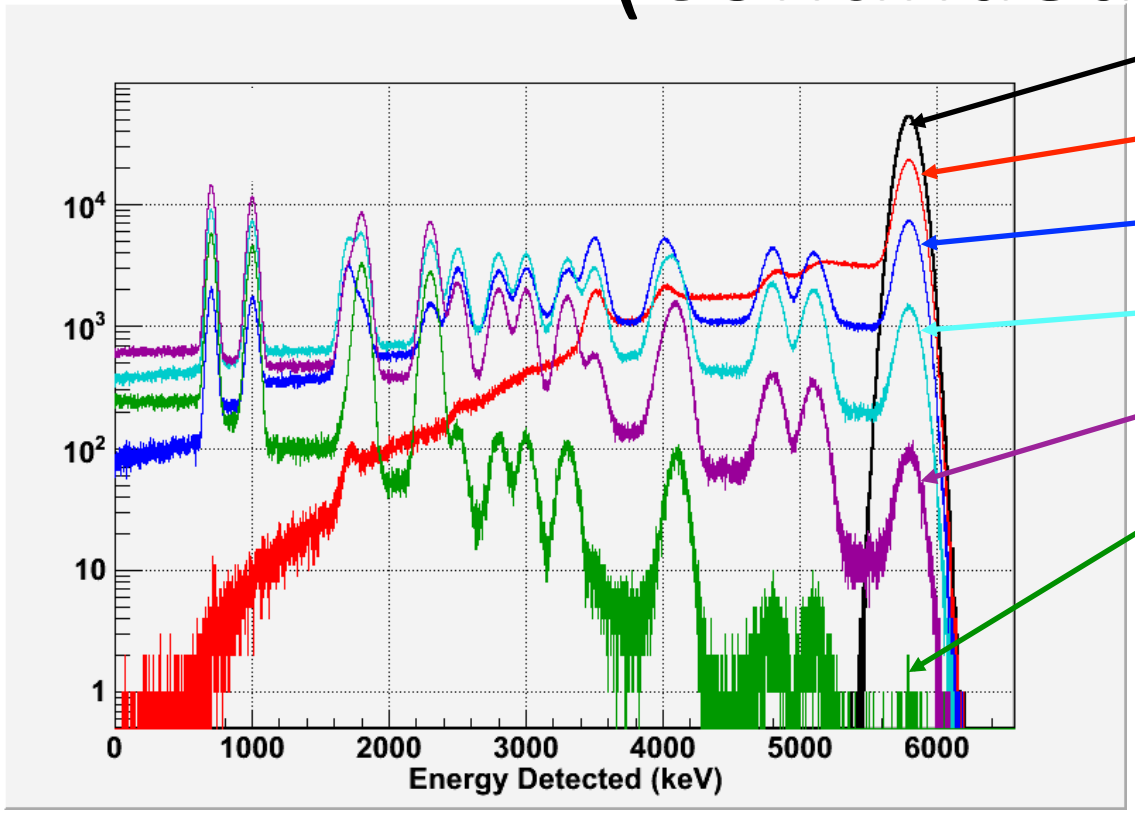
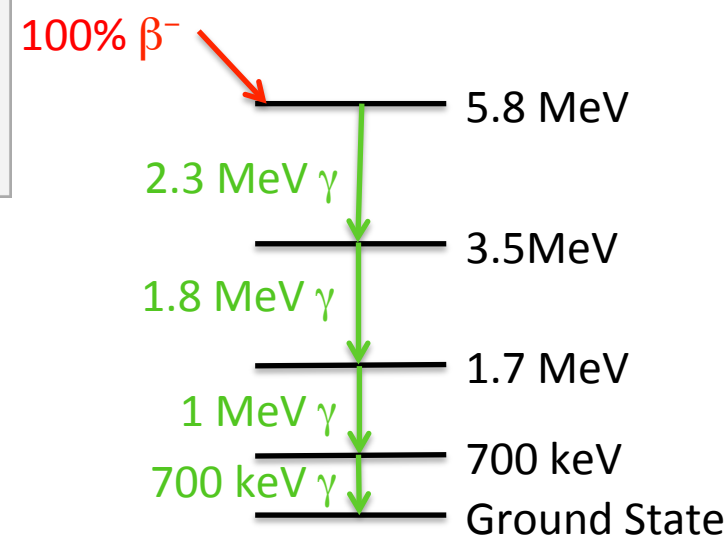


Illustration of the Pandemonium Effect (Continued)





Simulated 4 γ Decay from a Hypothetical 5.8 MeV Level

(E_γ=700 keV, 1 MeV, 1.8 MeV, and 2.3 MeV)

Current and Previous TAS Detectors

Modular Total Absorption Spectrometer (MTAS)
ORNL (since ~2011)

Idaho TAS : (1990's → ANL)
MTAS has 17 times larger volume

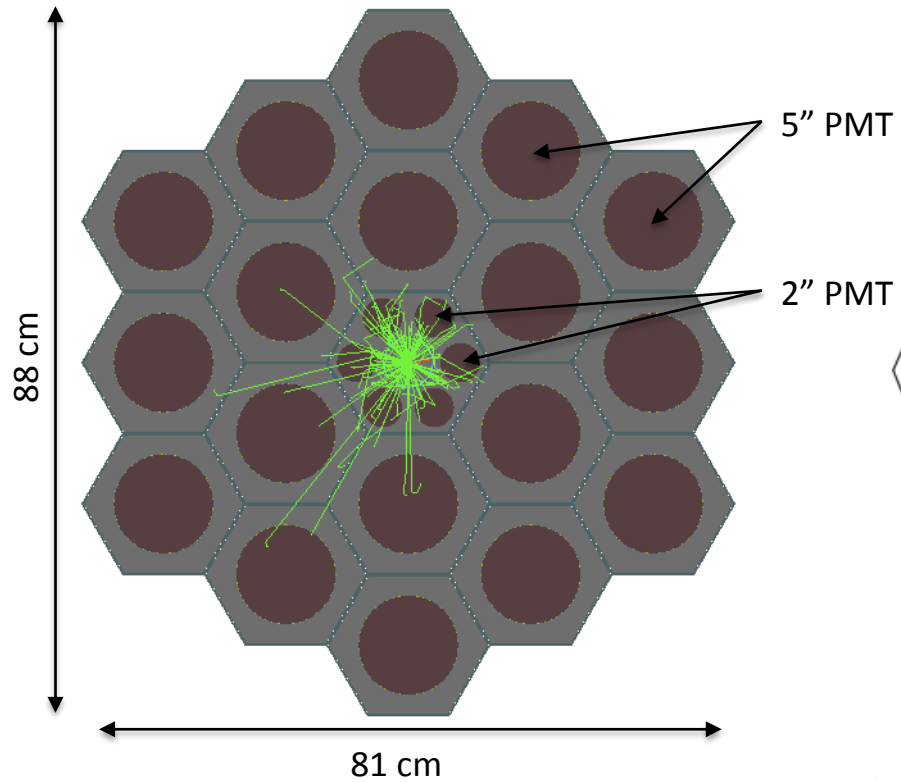
LBNL TAS : GSI (1995 – 2003) → LBNL
MTAS is 7 times larger

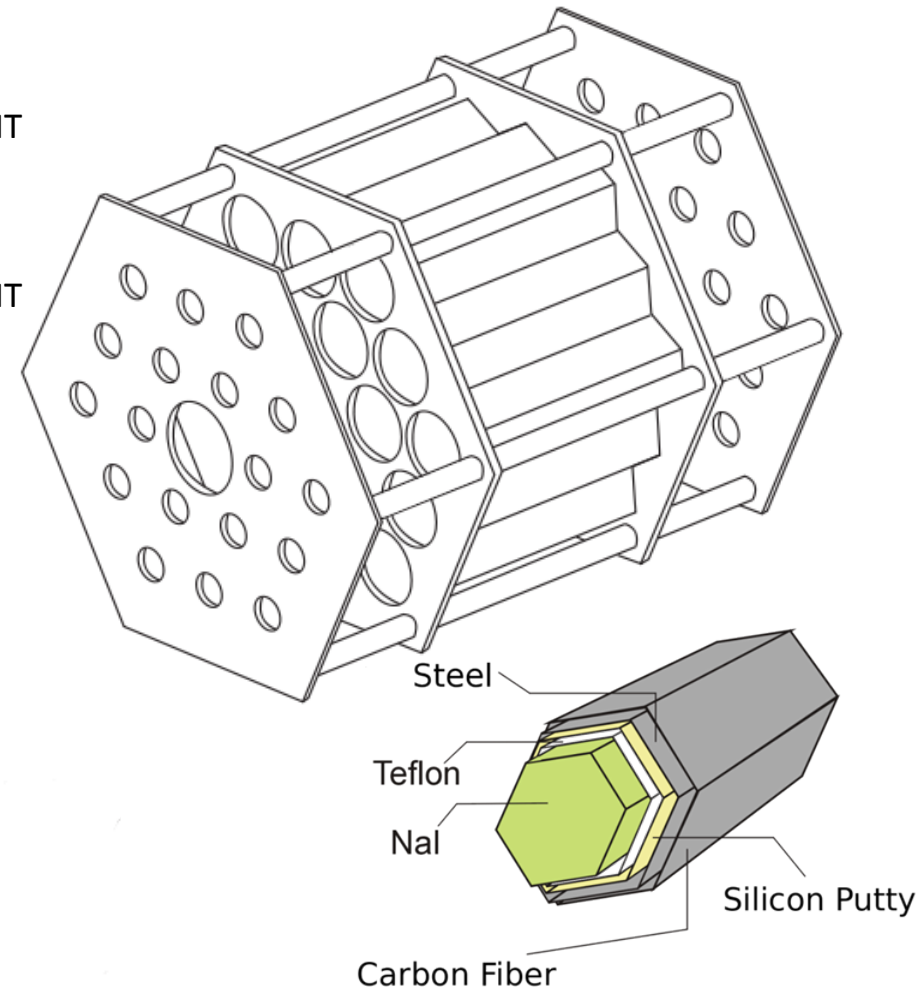
Valencia-ISOLDE "Lucrecia" TAS (since ~2003)
MTAS is 8 times larger

MSU-Notre Dame "SuN" TAS (since ~2011)
MTAS is 5 times larger

Modular Total Absorption Spectrometer

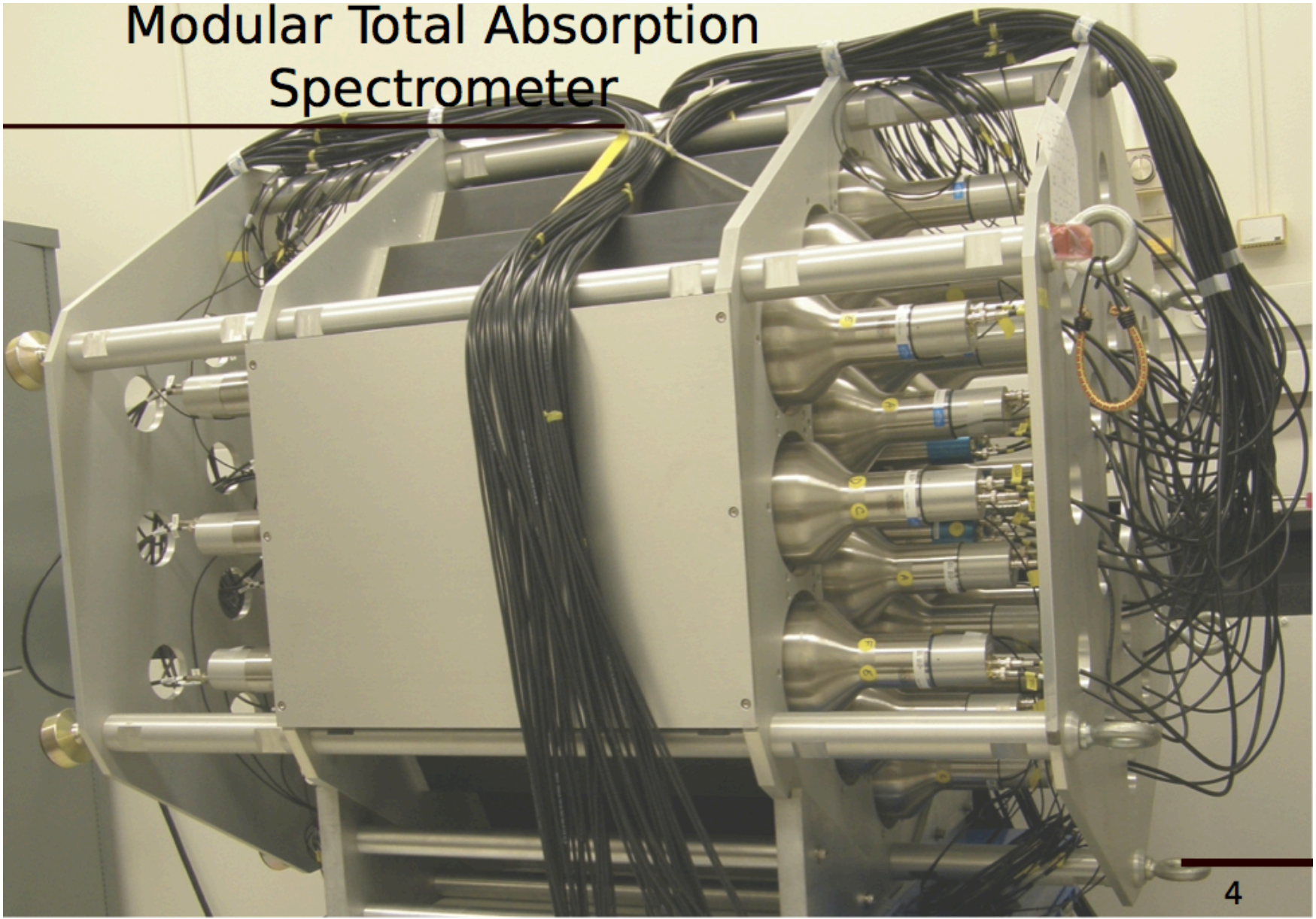
19 Hexagonal NaI(Tl) Modules (53.3 cm x 17.6 cm Face to Face) with Carbon Fiber Housing
Each Regular Module has 2-5” PMT, while the Center Module has 12-2” PMT
Center Module has 6.35 cm Hole to Accommodate Beam Line and Auxiliary β-Detectors
Approximately **One Ton** of NaI(Tl)!



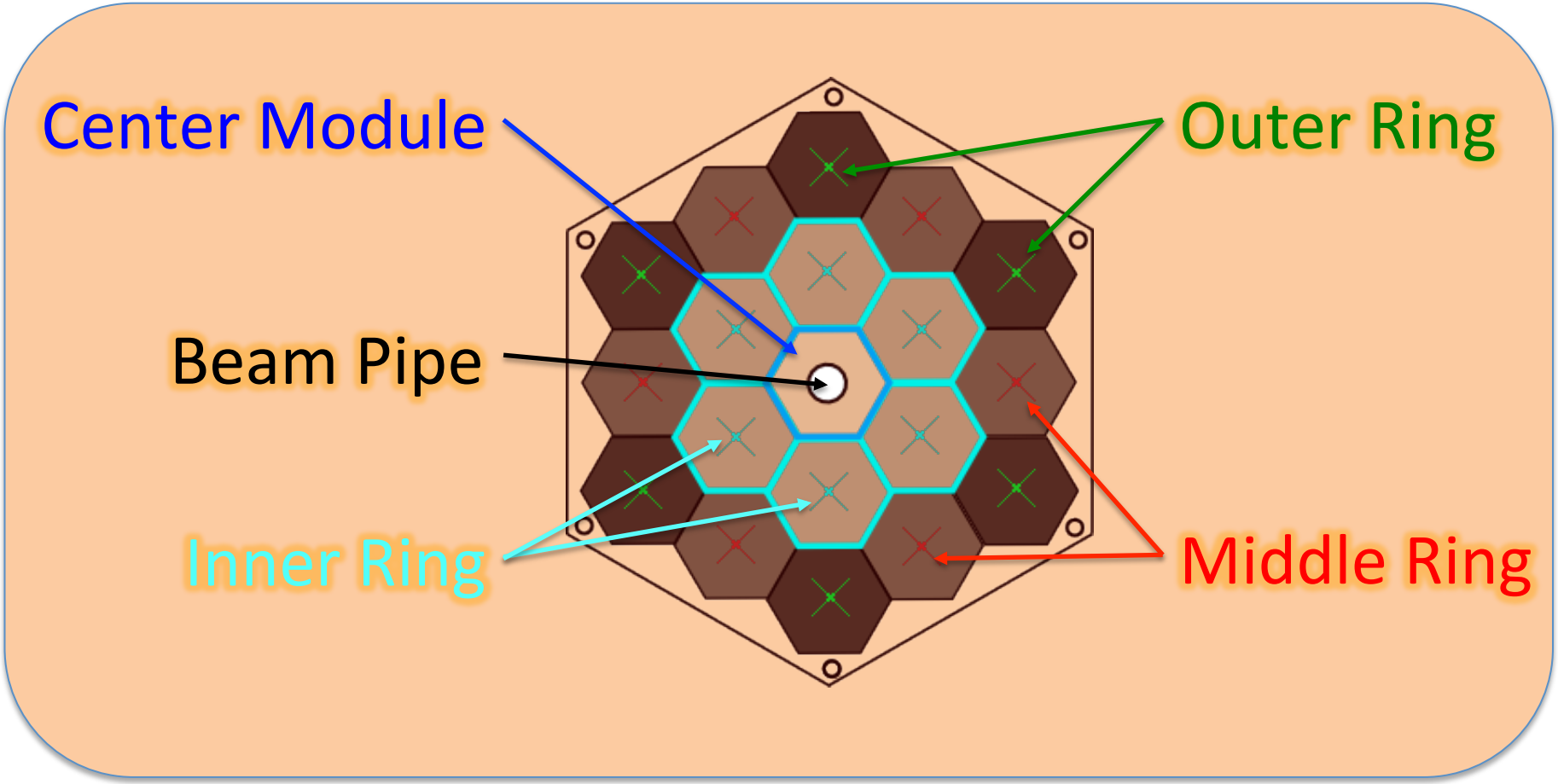


Modular Total Absorption Spectrometer with
100 ^{137}Cs Simulated Decays

MTAS

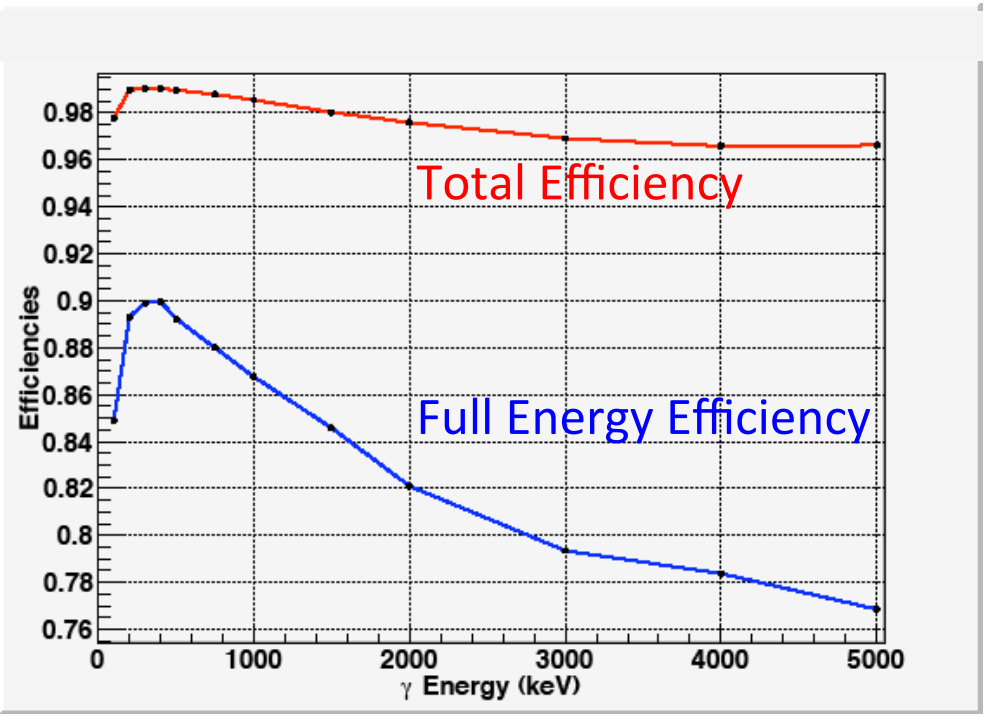


MTAS Ring Identification

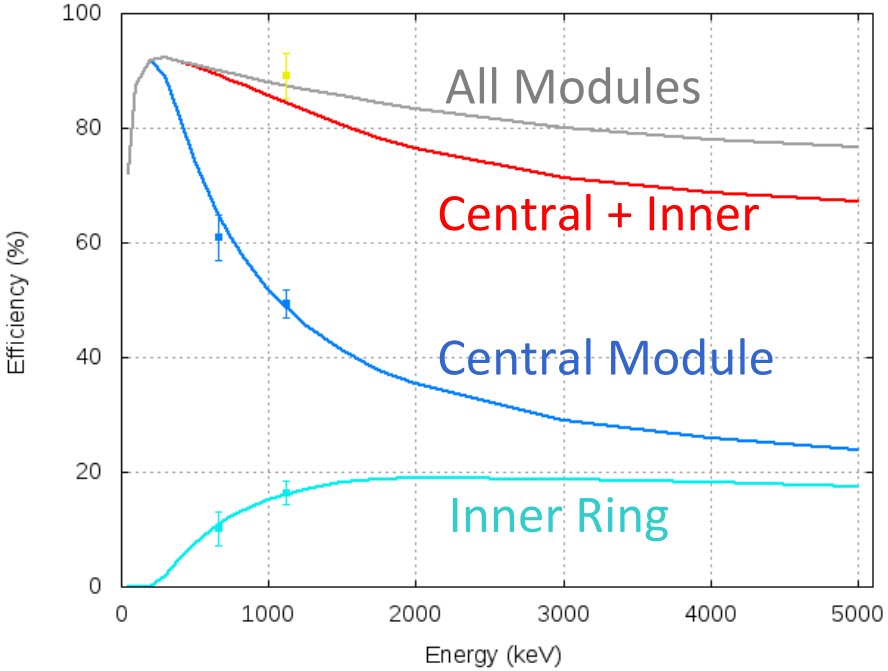


Efficiency of MTAS

Single γ Total Efficiency and Single γ Full Energy (Peak) Efficiency are Critical Parameters for Total Absorption Spectrometry (TAS)

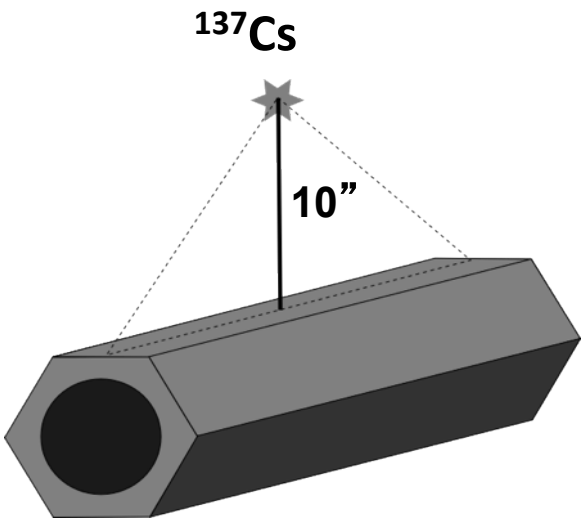


Simulated Single γ-Ray Efficiency versus Energy
Total Efficiency: Any Energy Deposited
Full Energy (Peak) Efficiency: All Energy Deposited

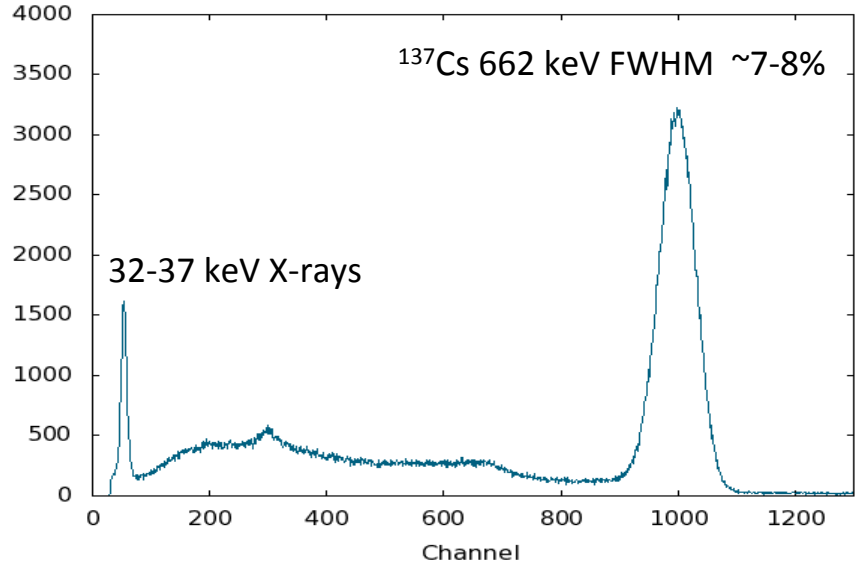


Single γ-Ray Full Energy Efficiency by Ring
Also Shown are ^{137}Cs and ^{65}Zn Data

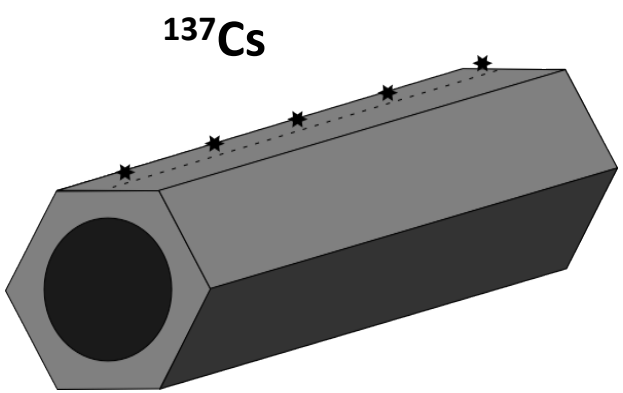
Test Measurements with ^{137}Cs Source

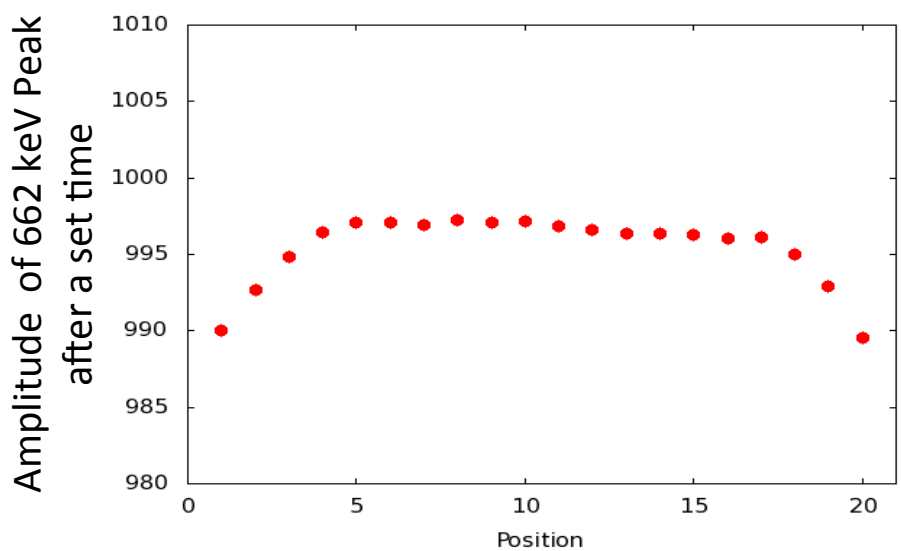


St Gobain specifications: FWHM < 8.5%

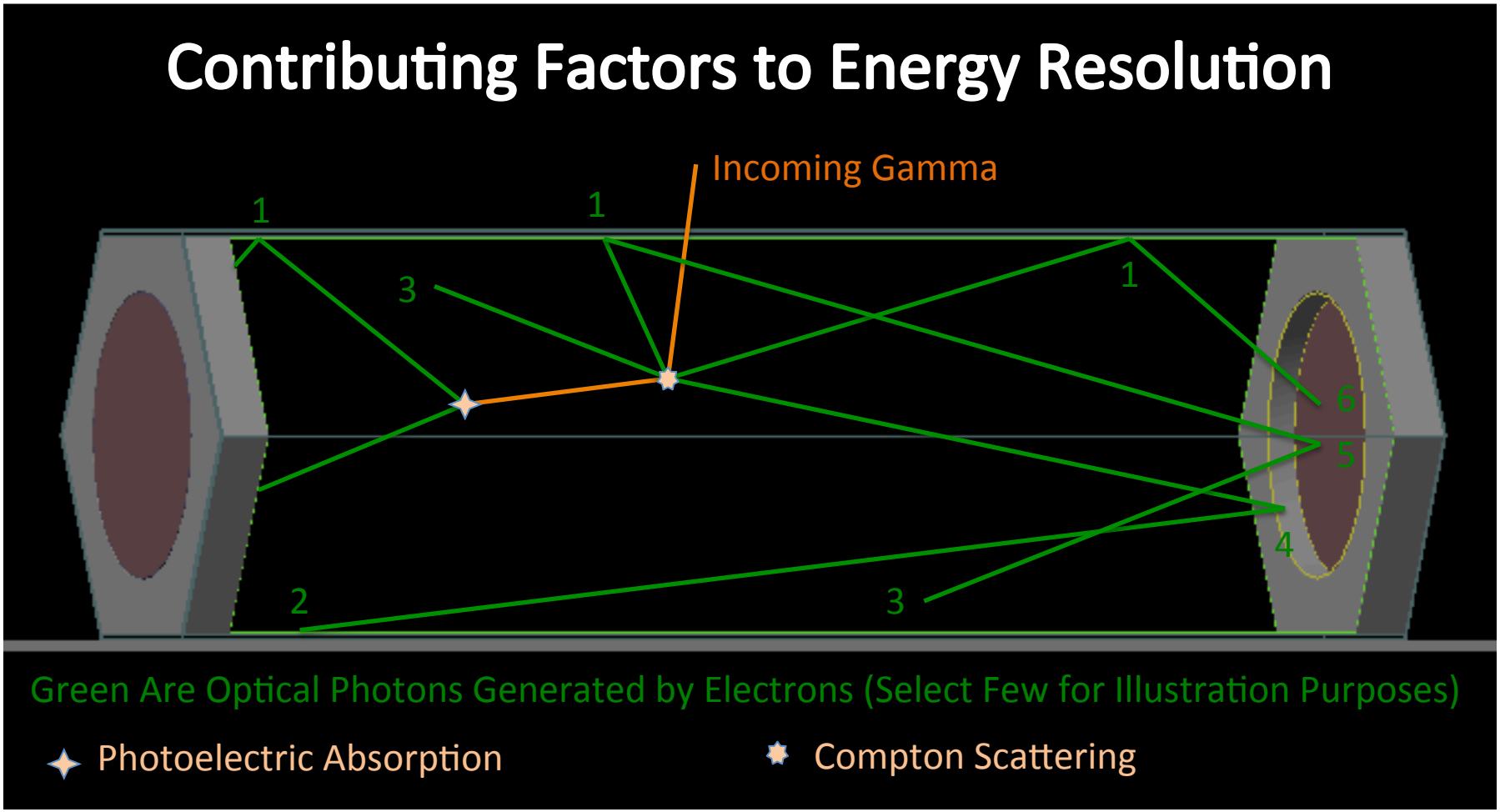


Uniformity of modules was measured with ^{137}Cs source placed along each side (19 measurements per each side, 1“ steps)





Simulation of Light Production in NaI(Tl)



1 – Reflection at Teflon NaI Boundary

2 – Absorption at Teflon NaI Boundary

3 – Absorption by NaI Crystal

4 – Reflection at NaI Window Glass Boundary

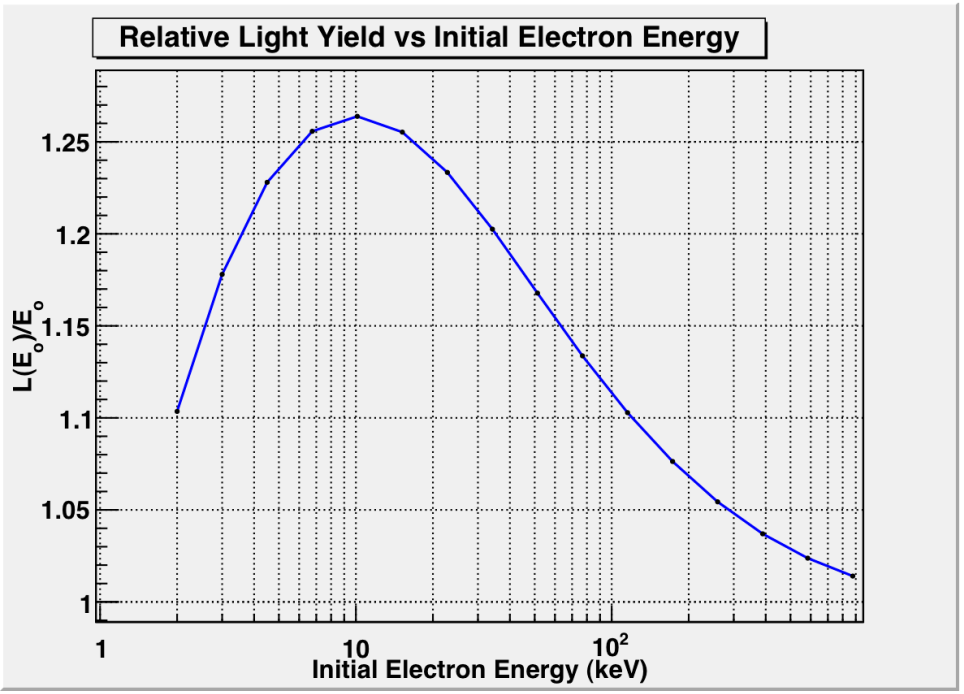
5 – Reflection at PMT Cathode Face

6 – Absorption with PE Creation on PMT Face

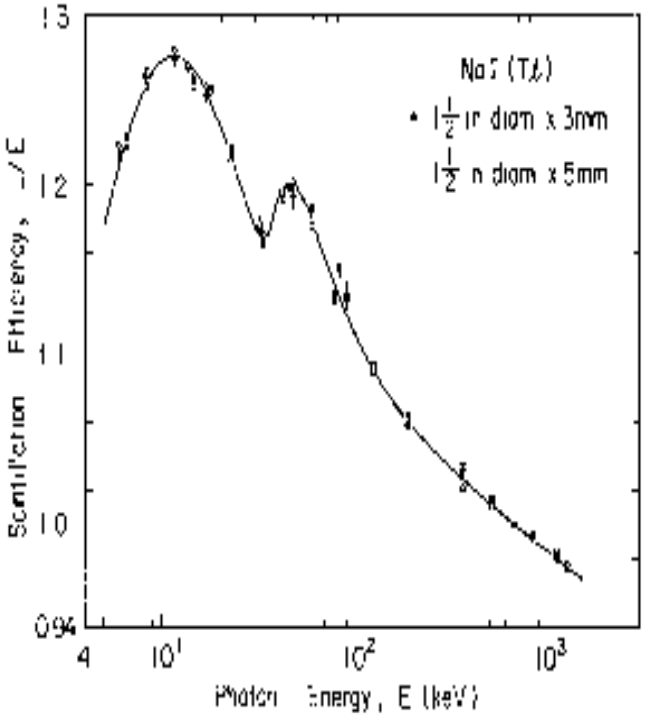
+ Nonlinear Light Production!

Nonlinear Light Generation in NaI(Tl)

Scintillators generate light approximately proportional to the energy deposited. But at low energy this is not necessarily the case. Organic scintillators often see a decrease in light output per energy deposited. NaI(Tl) actually generates an excess of light for energies less than ~1 MeV.

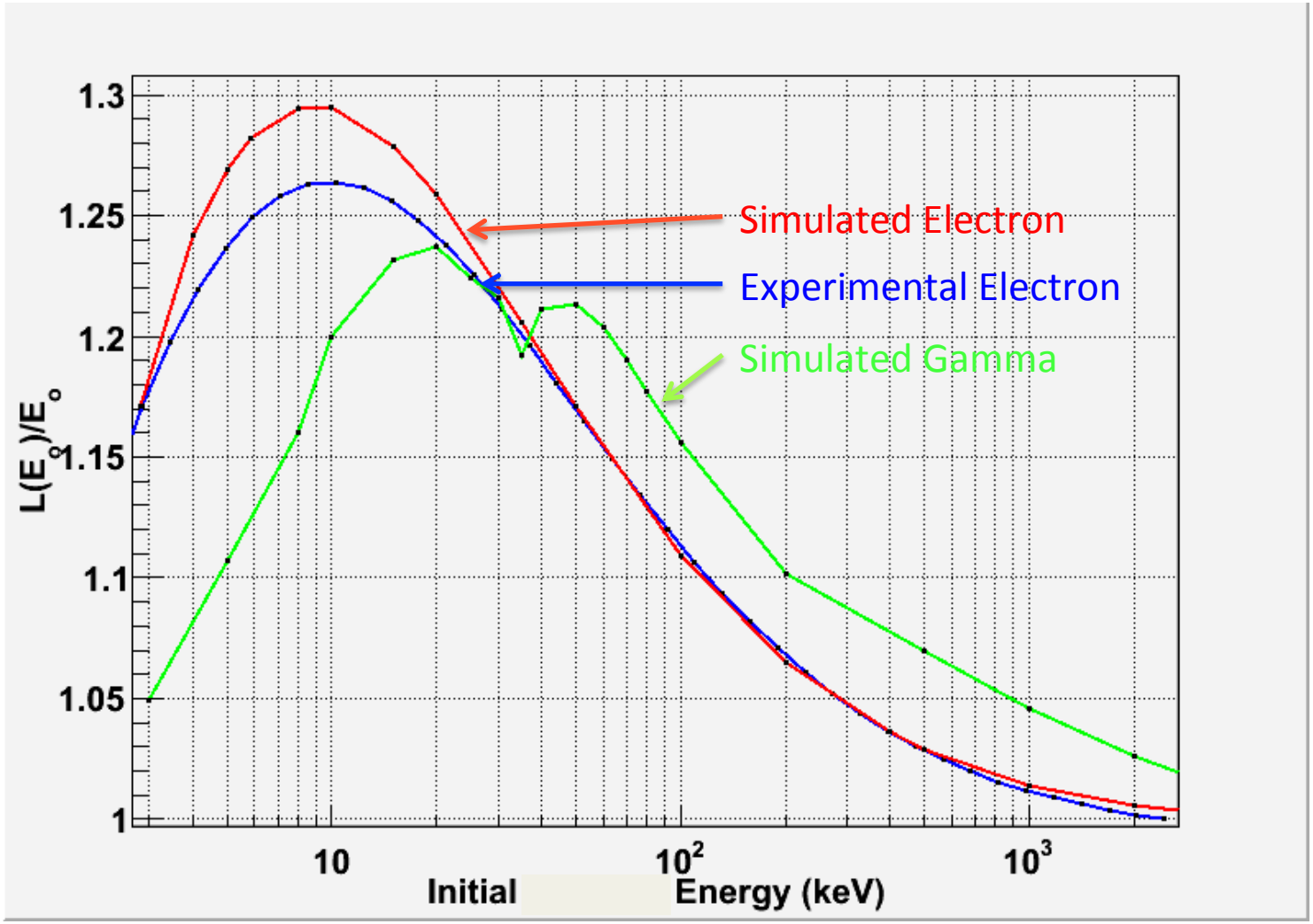


Parameterized experimental light output of electrons of a specified initial energy. The curve is normalized to unity for 3 MeV electrons.



Experimental light output of γ rays versus initial energy. The curve is normalized to unity at 662 keV. T. Tojo, NIM A238 (1985)

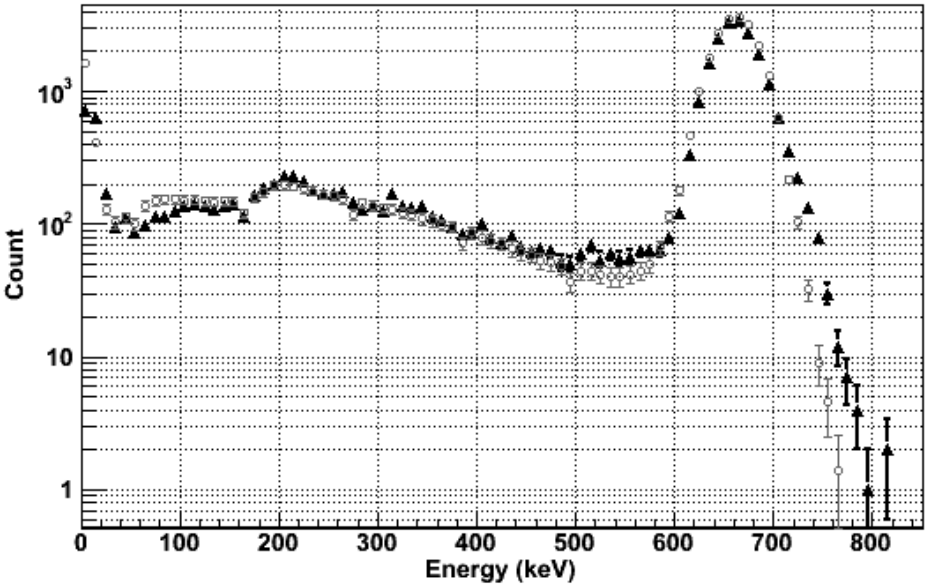
Simulation of Light Production in NaI(Tl) Nonlinear Light Generation



Results from dE/dx based light production in GEANT4. Good results for particles above ~20keV.

Simulation Compared to Measurement

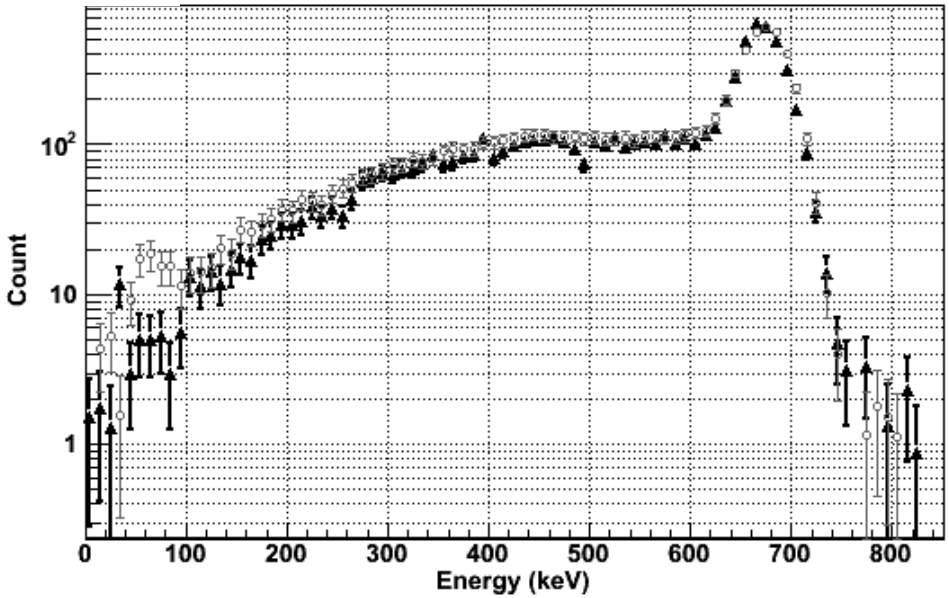
Center Module



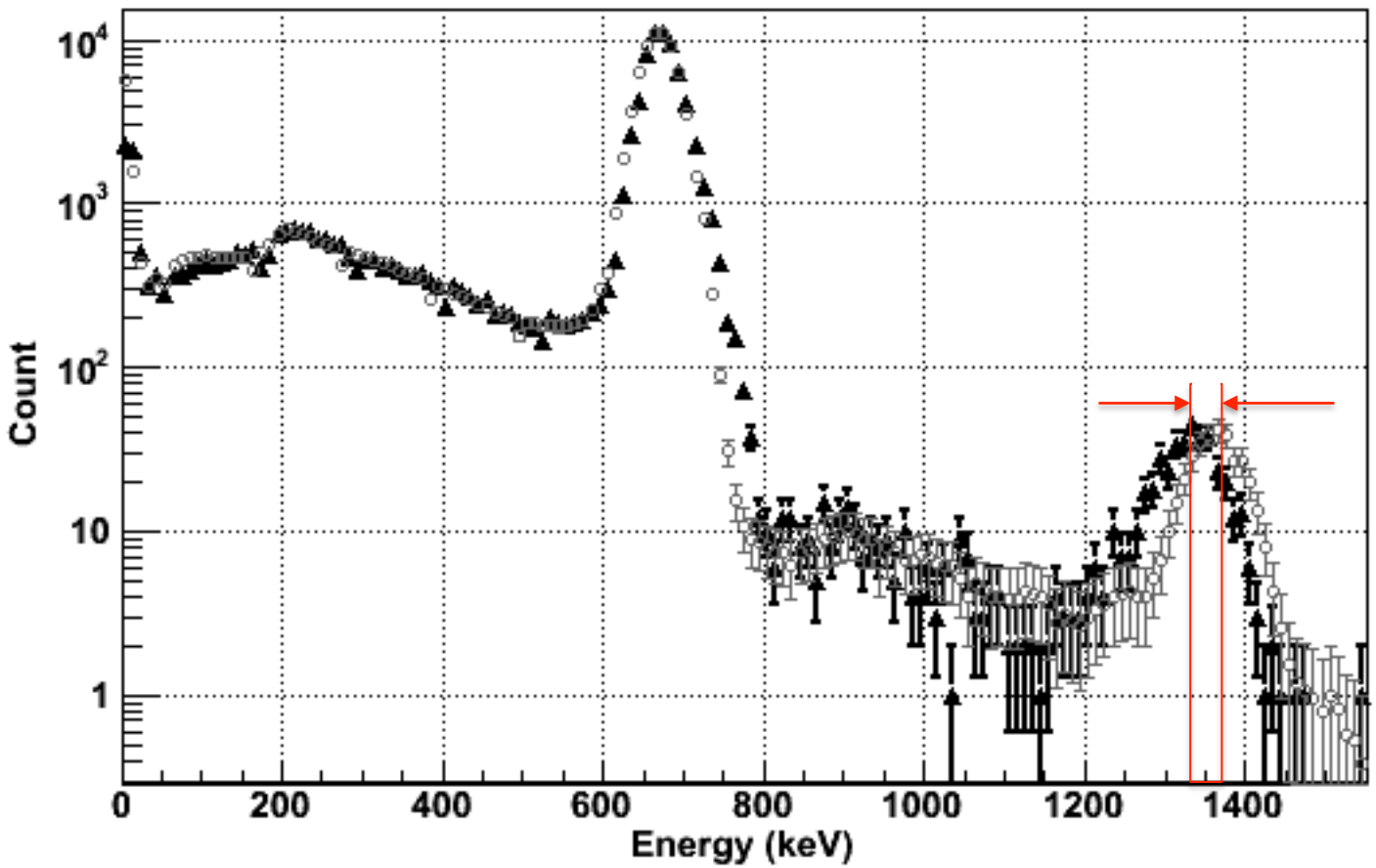
Low Activity ^{137}Cs

Triangles – Simulation
Open Circles - Data

Inner Ring



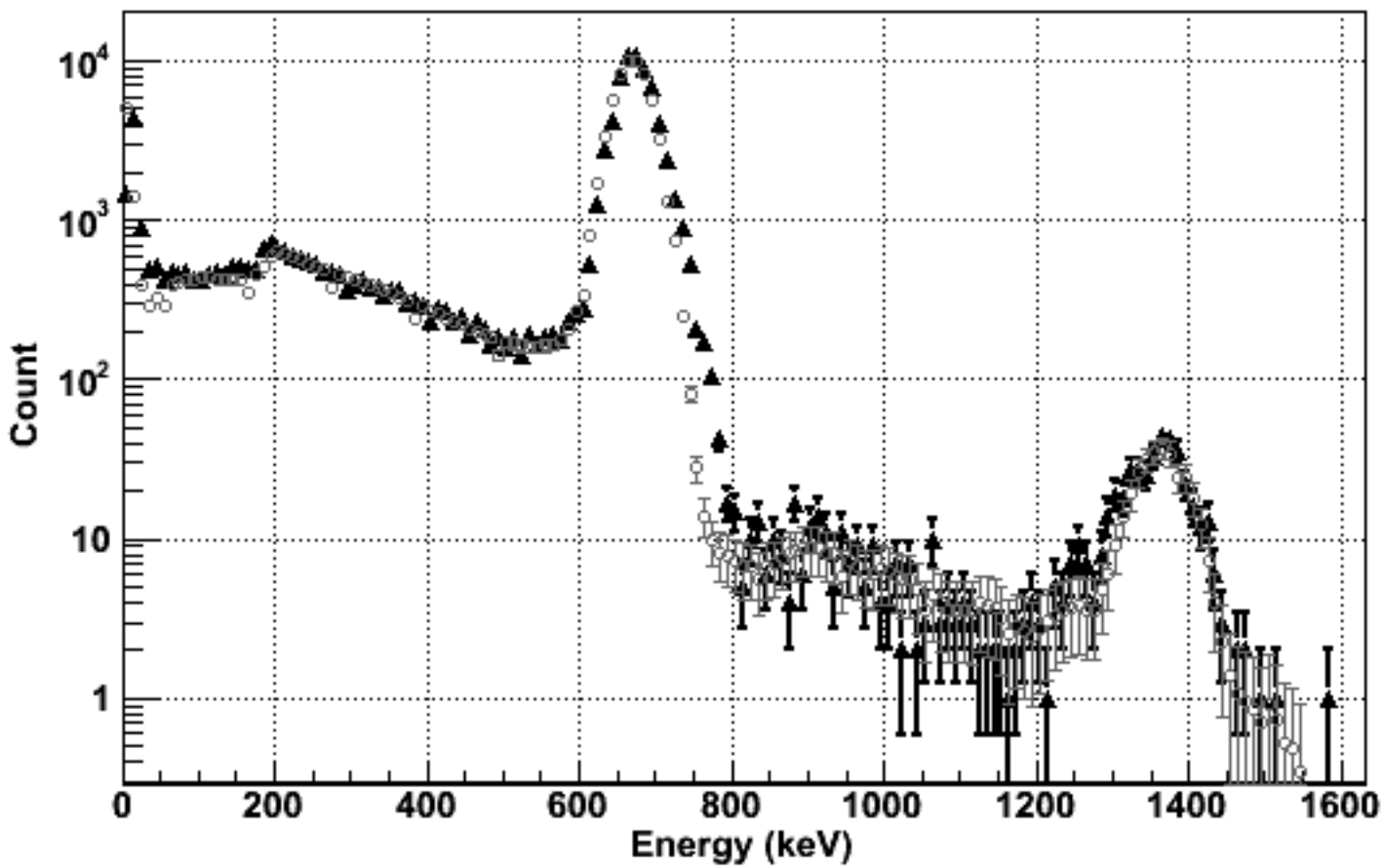
Simulation Compared to Measurement High Activity ^{137}Cs Center Module



Triangles – Simulation
Open Circles - Data

Per PMT Channel for Each Additional γ in Multi-γ Decays
~3 keV Shift per PMT per extra γ in Center Module (2.5" PMT)
~7 keV Shift per PMT per extra γ for Other Modules (5" PMT)

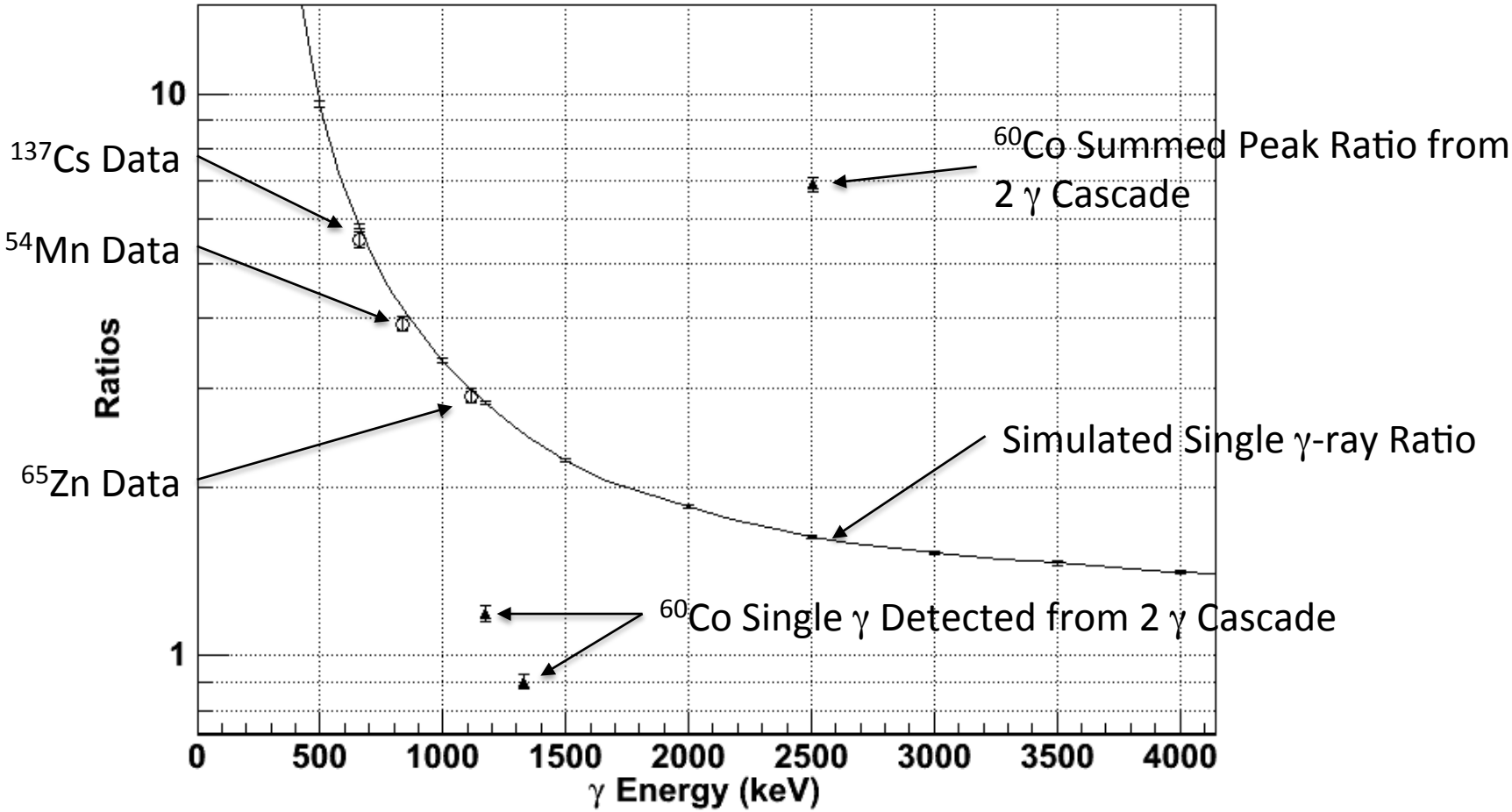
Simulation Compared to Measurement High Activity ^{137}Cs Center Module



Triangles – Simulation
Open Circles - Data

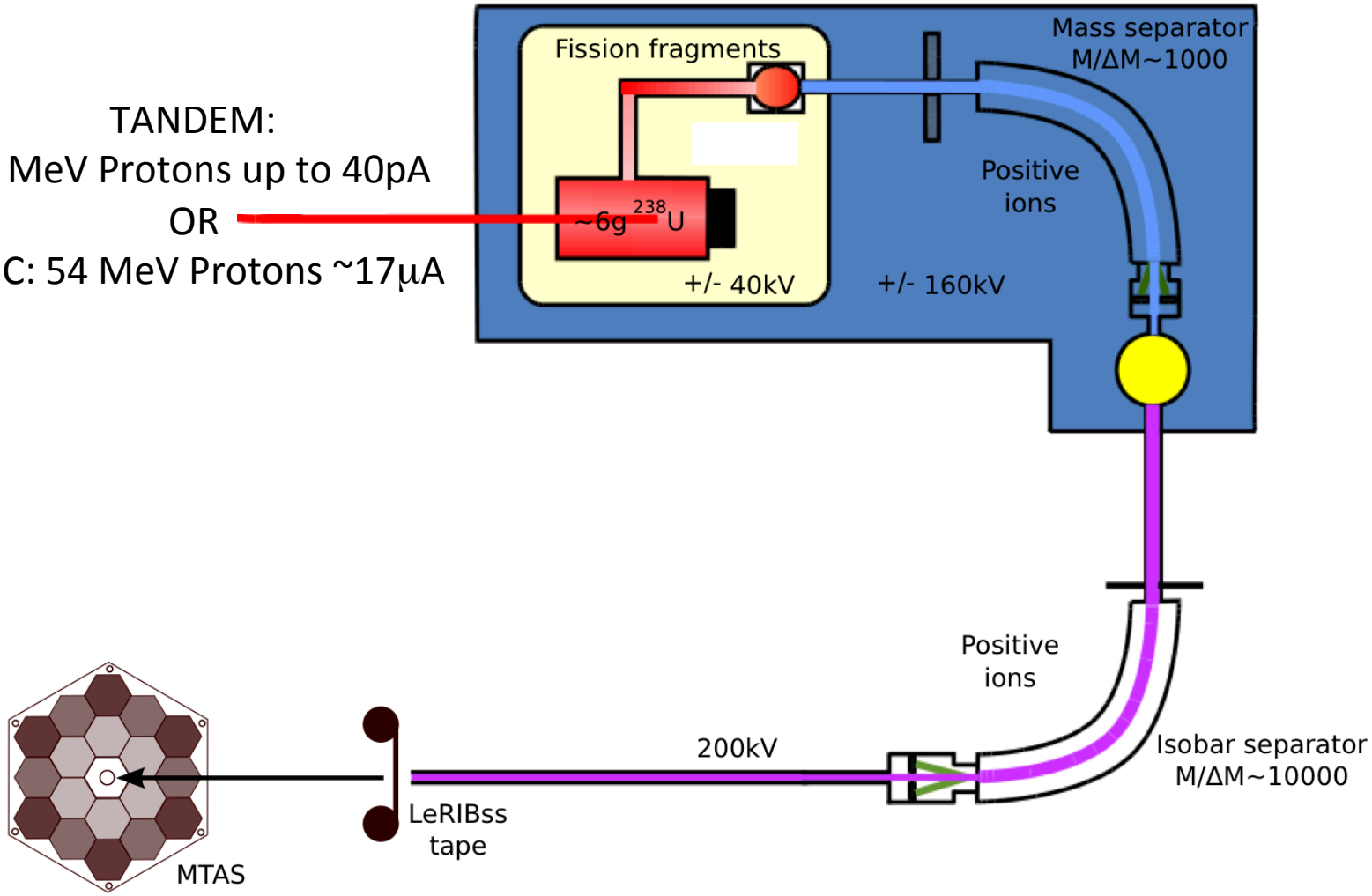
Modularity Uses of MTAS

Ratio of the counts in the central detector versus the inner ring of modules gives a measure of the γ multiplicity of the event



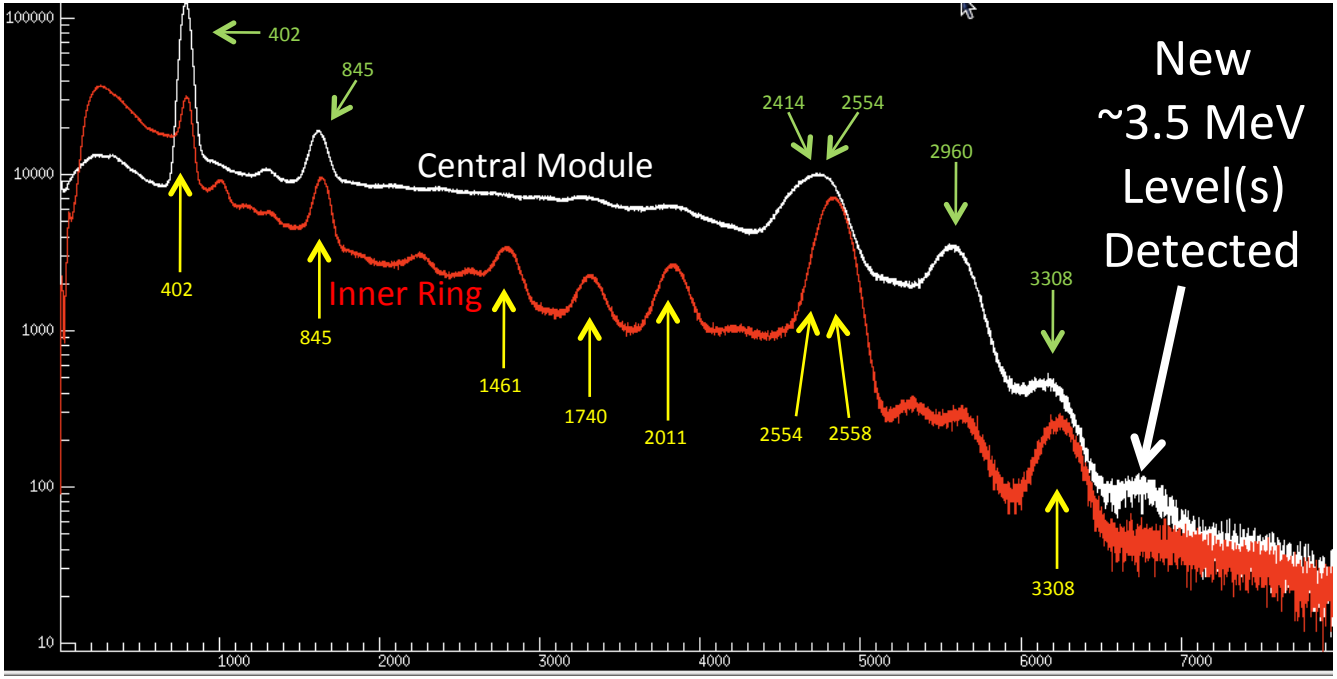
MTAS Experimental Setup

TANDEM:
40 MeV Protons up to 40pA
OR
ORIC: 54 MeV Protons ~17μA



First MTAS measurement of ^{87}Kr radioactivity ($T_{1/2}$=76 min) at the HRIBF

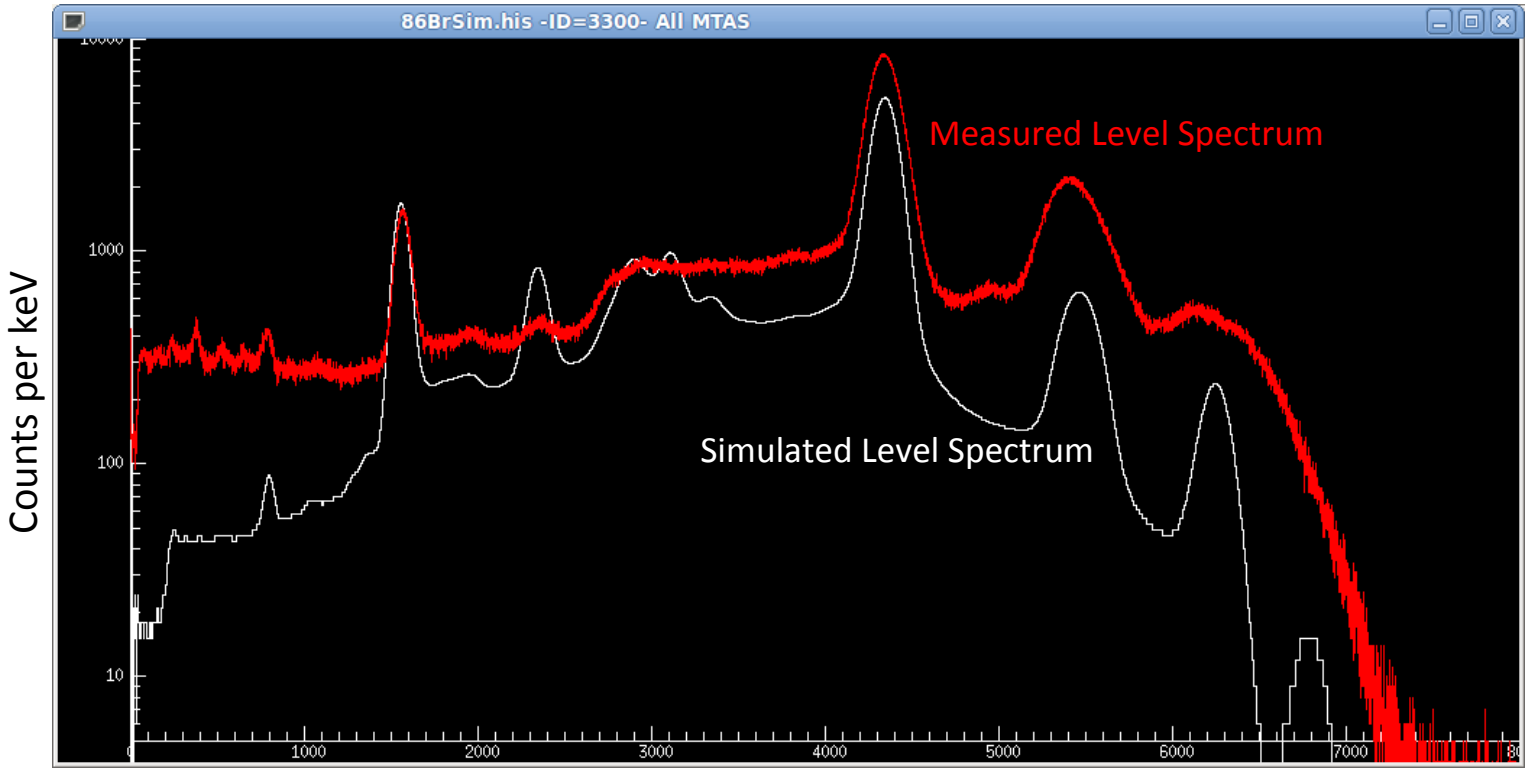
A=87 mass chain ending with ^{87}Kr decay is produced in thermal neutron fission at the rate of 2.5% per ^{235}U and 1% per ^{239}Pu



Channel Number (2 keV/channel)

MTAS at the HRIBF's OLTF, January 2012

Example of Collected Data: Priority "1" Decay of $^{86}Br \rightarrow {}^{86}Kr + \beta^-$



The measured MTAS gamma spectrum (red line) is compared to the simulation of MTAS response (white line) to the decay of ^{86}Br listed in the adopted data files. The normalization is made to match the feeding of the 1.6 MeV excited state in ^{86}Kr. An excess of high energy gamma radiation release is clearly detected

(A. Kuźniak et al., January 2012, preliminary)

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International Evaluation Co-operation

VOLUME 25

ASSESSMENT OF FISSION PRODUCT DECAY DATA FOR DECAY HEAT CALCULATIONS

A report by the Working Party on International Evaluation Co-operation of the NEA Nuclear Science Committee

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International Atomic Energy Agency
AUSTRIA

Red Arrows Indicate Recent MTAS-OLTF Measurements

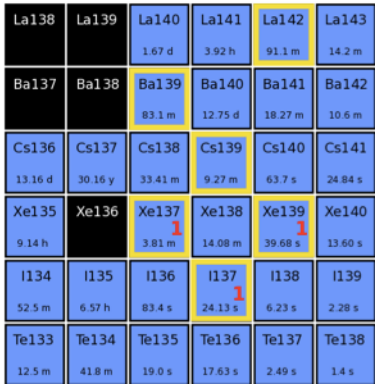

Table 3. Requested TAGS measurements

Radionuclide	Priority	Q_β-value (keV)	Half-life	Comments
35-Br-86	1	7626(11)	55.1 s	
35-Br-87	1	6852(18)	55.65 s	Extremely complex decay scheme with substantial gamma component; large uncertainties in the mean gamma energy arises from significant disagreements between the various discrete gamma-ray measurements. Also (β^-,n) branch.
35-Br-88	1	8960(40)	16.36 s	(β^-,n) branch.
36-Kr-89	1	4990(50)	3.15 min	Incomplete decay scheme.
36-Kr-90	1	4392(17)	32.32 s	Incomplete decay scheme.
37-Rb-90m	2	6690(15)	258 s	Repeat of INL TAGS measurement; data check.
37-Rb-92	2	8096(6)	4.49 s	Small (β^-,n) branch.
38-Sr-89	2	1493(3)	50.53 d	
38-Sr-97	2	7470(16)	0.429 s	Extremely short half-life (0.429 s), and possible (β^-,n) branch.
39-Y-96	2	7096(23)	5.34 s	
40-Zr-99	3	4558(15)	2.1 s	
40-Zr-100	2	3335(25)	7.1 s	
41-Nb-98	1	4583(5)	2.86 s	
41-Nb-99	1	3639(13)	15.0 s	
41-Nb-100	1	6245(25)	1.5 s	
41-Nb-101	1	4569(18)	7.1 s	
41-Nb-102	2	7210(40)	1.3 s	
42-Mo-103	1	3750(60)	67.5 s	
42-Mo-105	1	4950(50)	35.6 s	
43-Tc-102	1	4532(9)	5.28 s	
43-Tc-103	1	2662(10)	54.2 s	
43-Tc-104	1	5600(50)	18.3 min	
43-Tc-105	1	3640(60)	7.6 min	
43-Tc-106	1	6547(11)	35.6 s	
43-Tc-107	2	4820(90)	21.2 s	
51-Sb-132	1	5509(14)	2.79 min	
52-Te-135	2	5960(90)	19.0 s	

Radionuclide	Priority	Q_β-value (keV)	Half-life	Comments
53-I-136	1	6930(50)	83.4 s	Incomplete decay scheme.
53-I-136m	1	7580(120)	46.9 s	
53-I-137	1	5877(27)	24.13 s	(β^-,n) branch.
54-Xe-137	1	4166(7)	3.82 min	Incomplete decay scheme.
54-Xe-139	1	5057(21)	39.68 s	
54-Xe-140	1	4060(60)	13.6 s	
55-Cs-142	3	7308(11)	1.69 s	(β^-,n) branch.
56-Ba-145	2	5570(110)	4.31 s	Repeat of INL TAGS measurement; data check.
57-La-143	2	3425(15)	14.2 min	Repeat of INL TAGS measurement; data check.
57-La-145	2	4110(80)	24.8 s	Repeat of INL TAGS measurement; data check.

MTAS Collaborators

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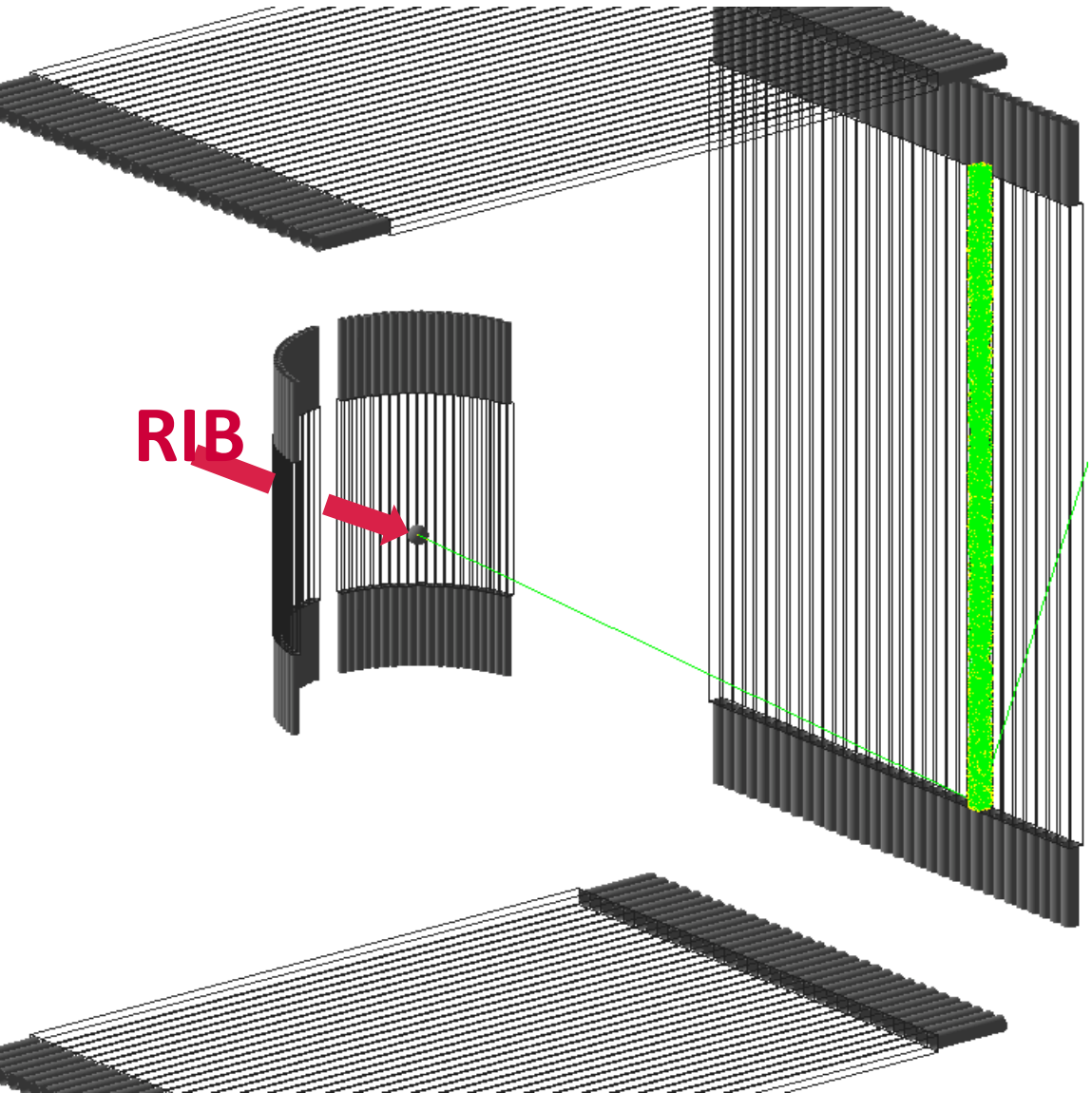
[3]University of Tennessee, Knoxville, TN 37996, USA,
[4]ORAU, Oak Ridge, TN 37830, USA,
[5]Louisiana State University, Baton Rouge, LA 70803, USA

Versatile Array of Neutron Detectors at Low Energy (VANDLE)



Neutron Time of Flight Detector

~250 Configurable Scintillator (BC408) Bars

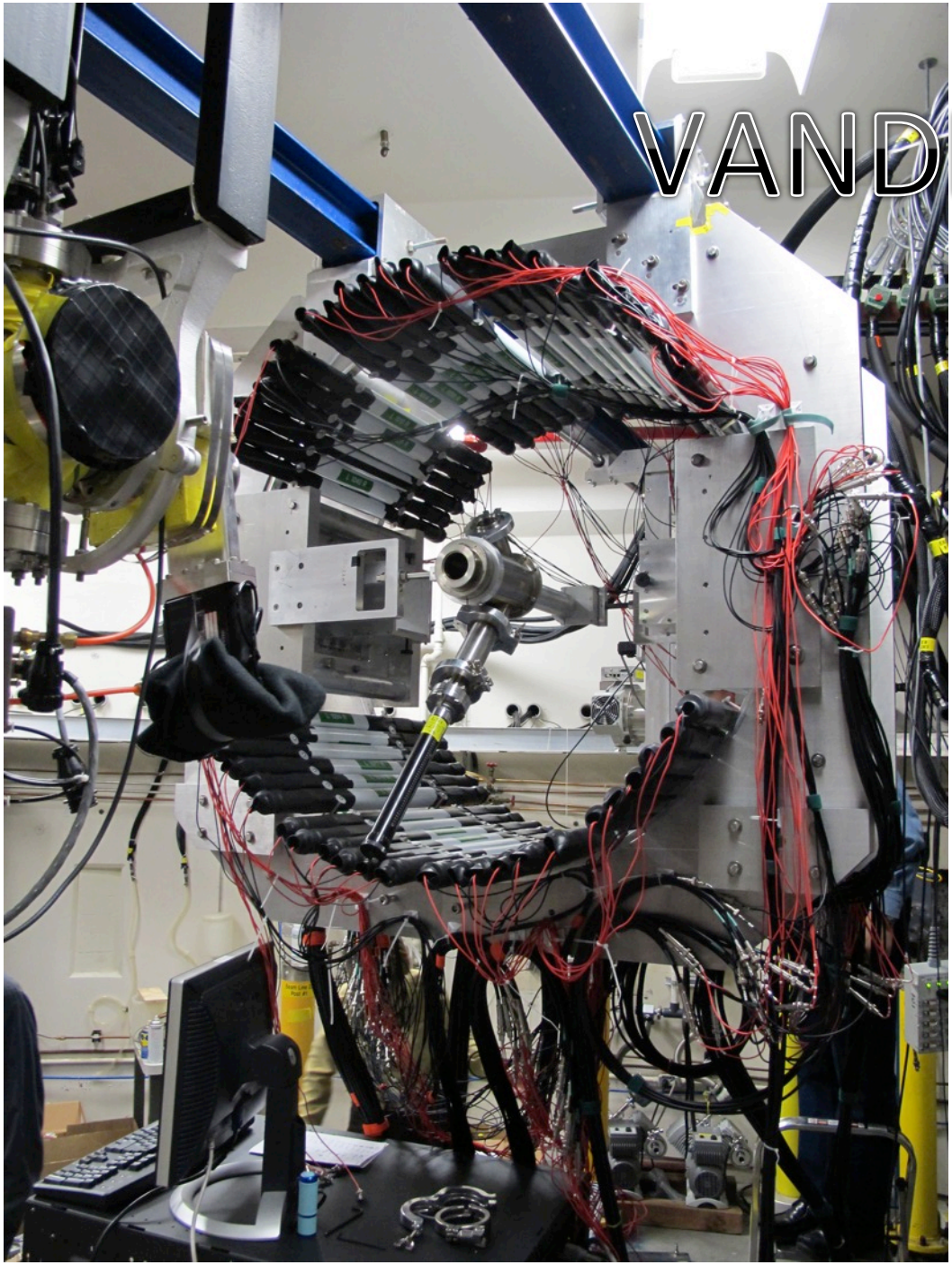
Two sizes of Scintillator Bars:

200 cm length (5^2 cm^2 area)

60 cm length (2.5^2 cm^2 area)

Bars can be arranged in different configurations that are optimized for particular types of measurements including transfer reactions like (d,n) and neutron emission following β decay

VANDLE



First VANDLE Experimental Setup

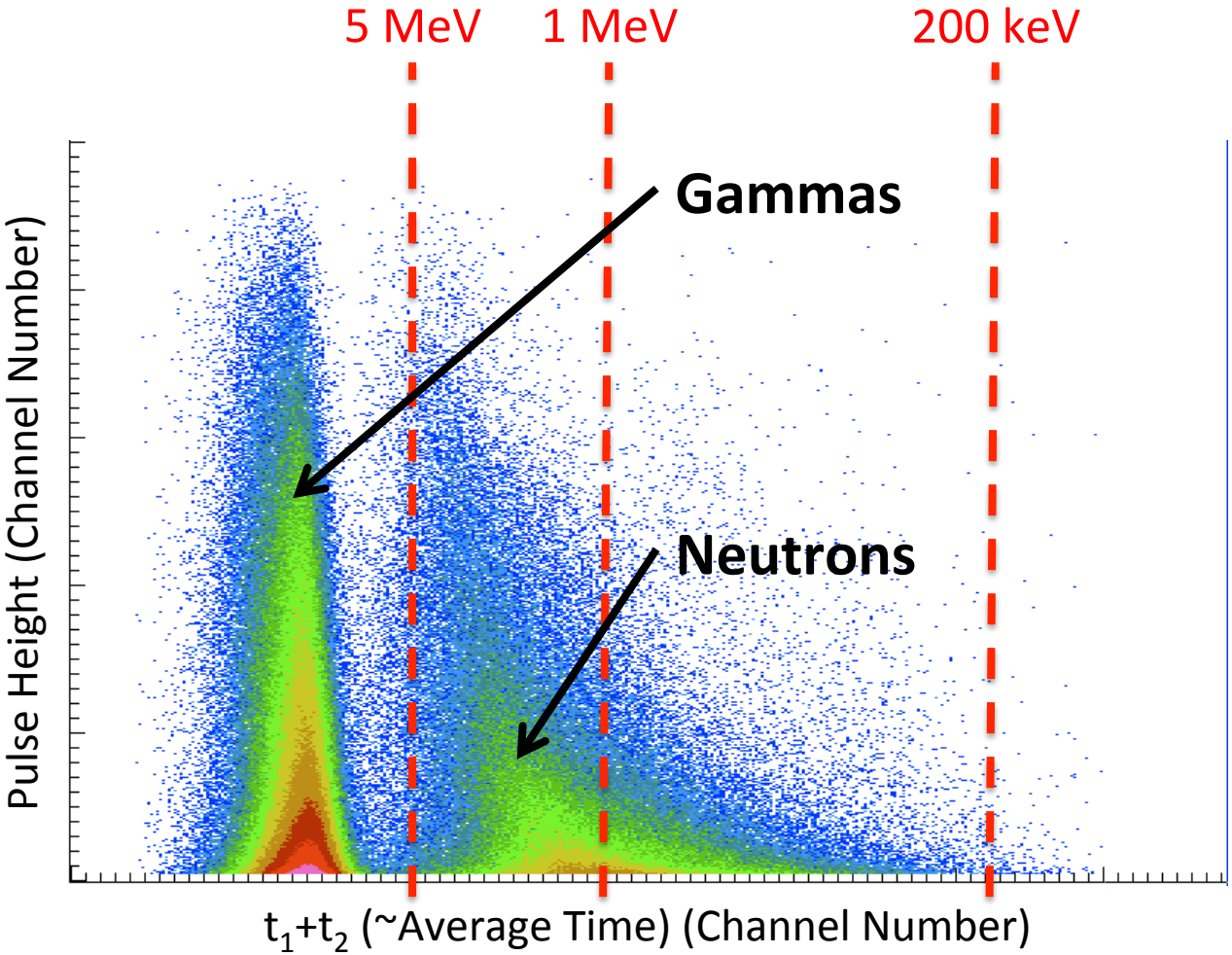
Time of Flight Neutron and γ Detector

40 – 60 x 2.5 x 2.5 cm^3 Scintillator Bars

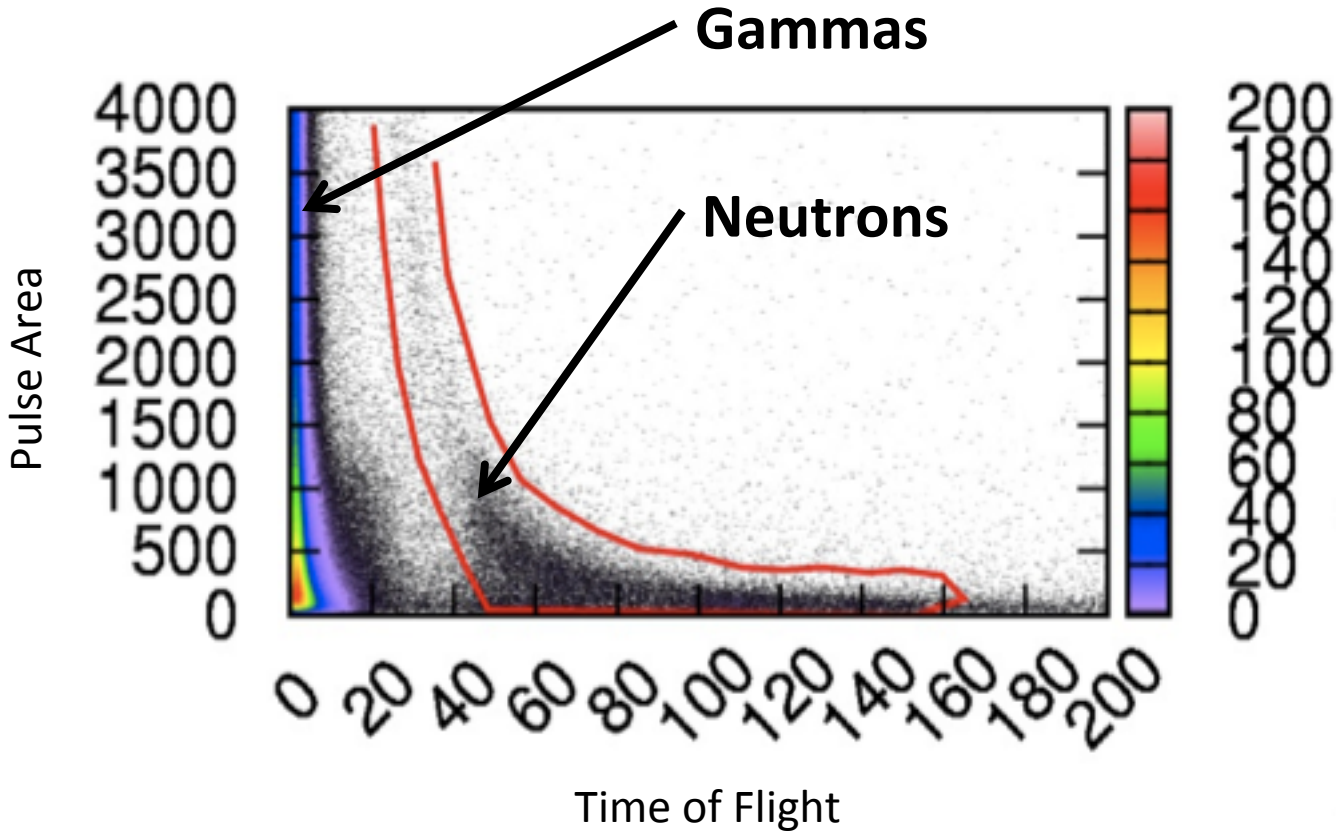
2 Clover Germanium Detector

2 Plastic Scintillator β Detectors

Single VANDLE Bar ^{252}Cf Measurements

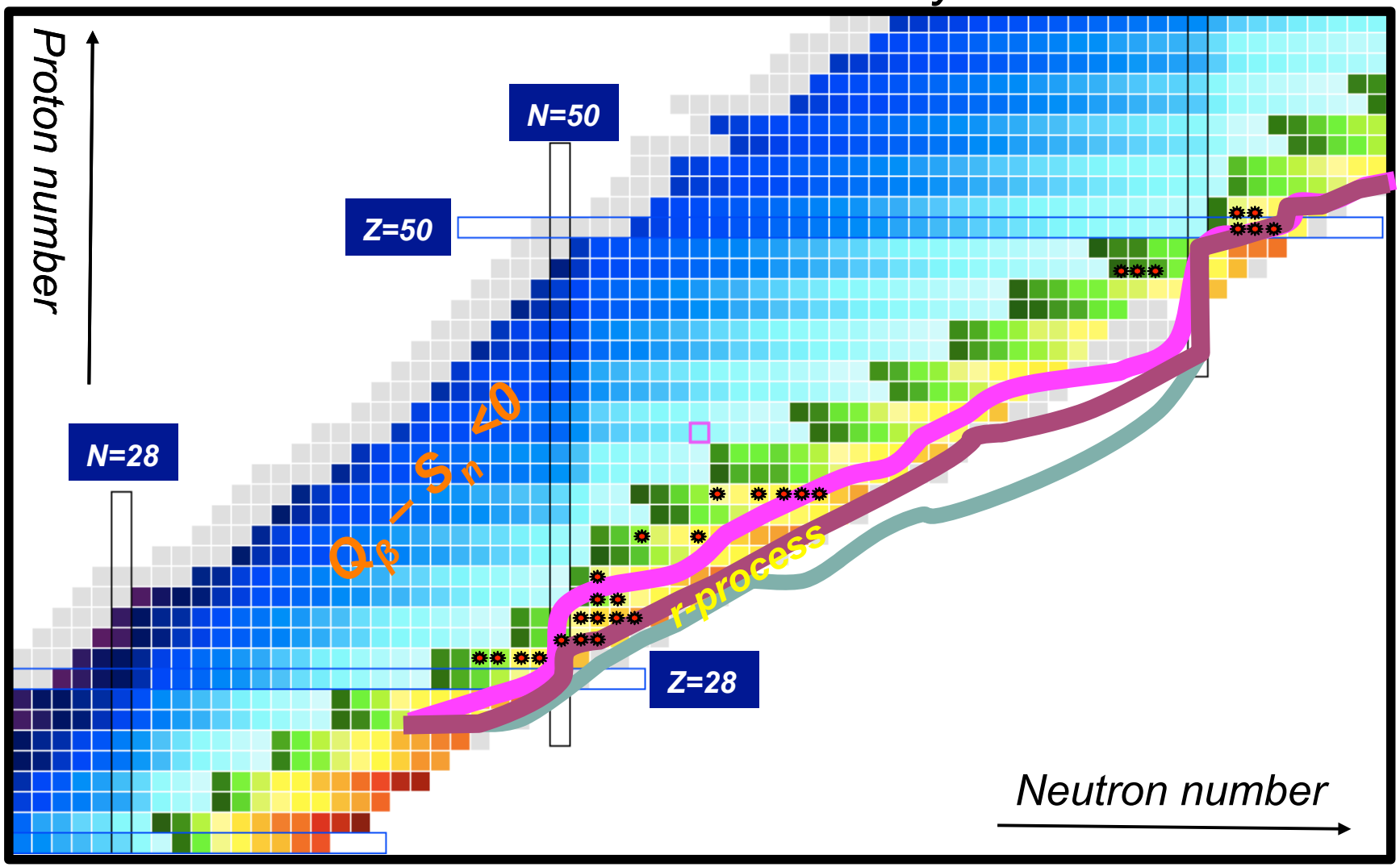


Raw VANDLE Data from ^{77}Cu



The Versatile Array of Neutron Detectors at Low Energy

Beta-delayed neutron emitters near r-process path studied at HRIBF/LeRIBSS in February 2012



VANDLE Collaborators

R. Grzywacz[1, 2], S.V. Paulauskas[1], M. Madurga[1], W.A Peters[3], C.J. Gross[2], D.T. Miller[1]
M. Al-Shudifat[1], D.W. Bardayan[2], J.C. Batchelder[3], J. Blackmon[4], N.T. Brewer[5], L. Cartegni[1],
K. Chipps[8], J.A. Cizewski[6], P.A. Copp[7], M.E. Howard[6], S. Ilyushkin[8], C. Jost[2], A. Kuzniak[9], S. Liu[3],
B. Manning[6], C. Matei[3], M. Matos[6], A.J. Mendez II[2], K. Miernik[2], P.D. O'Malley[6], S. Padgett[1],
B.C. Rasco[4], D. Stracener[2], F. Raiola[8], A. Ratkiewicz[6], K. Rykaczewski[2], F. Sarazin[8], K.T. Schmitt[2],
I. Spassova[3], D. Walter[6], E.H. Wang[5], M. Wolinska-Cichocka[3], E. Zganjar[4]

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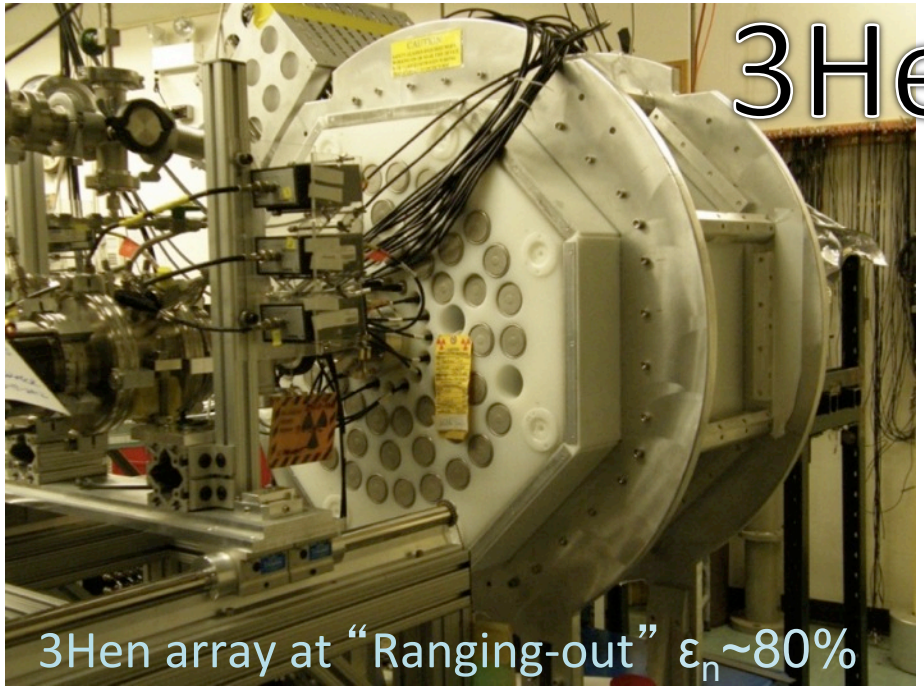
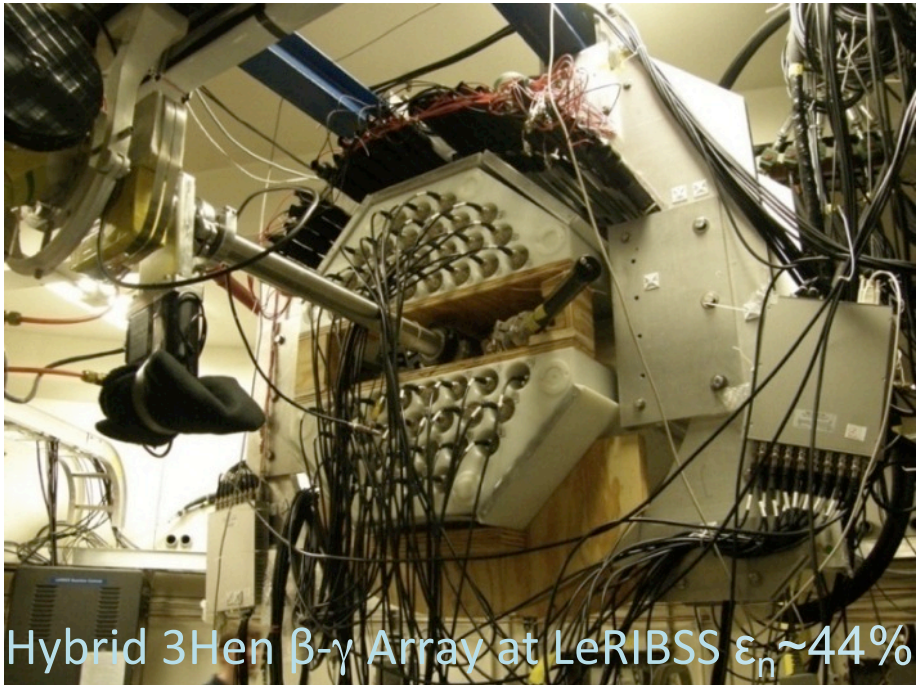
3Hen



3Hen array at "Ranging-out" ε_n~80%



Hybrid 3Hen β-γ Array at LeRIBSS ε_n~44%

Thermalized Neutron Detector

Hybrid 3Hen Details in ()

74 (48) High Efficiency ^{3}He Gas Tubes

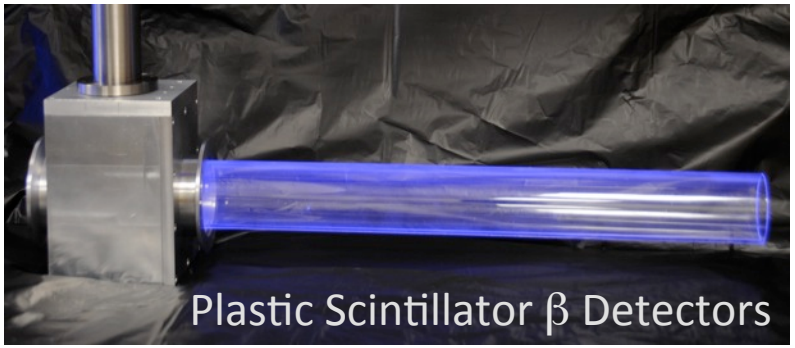
(2 Clover Germanium Detectors)

2 Plastic Scintillator β Detectors

^{3}He Tubes Surrounded by High Density Polyethylene to Help Thermalize Neutrons

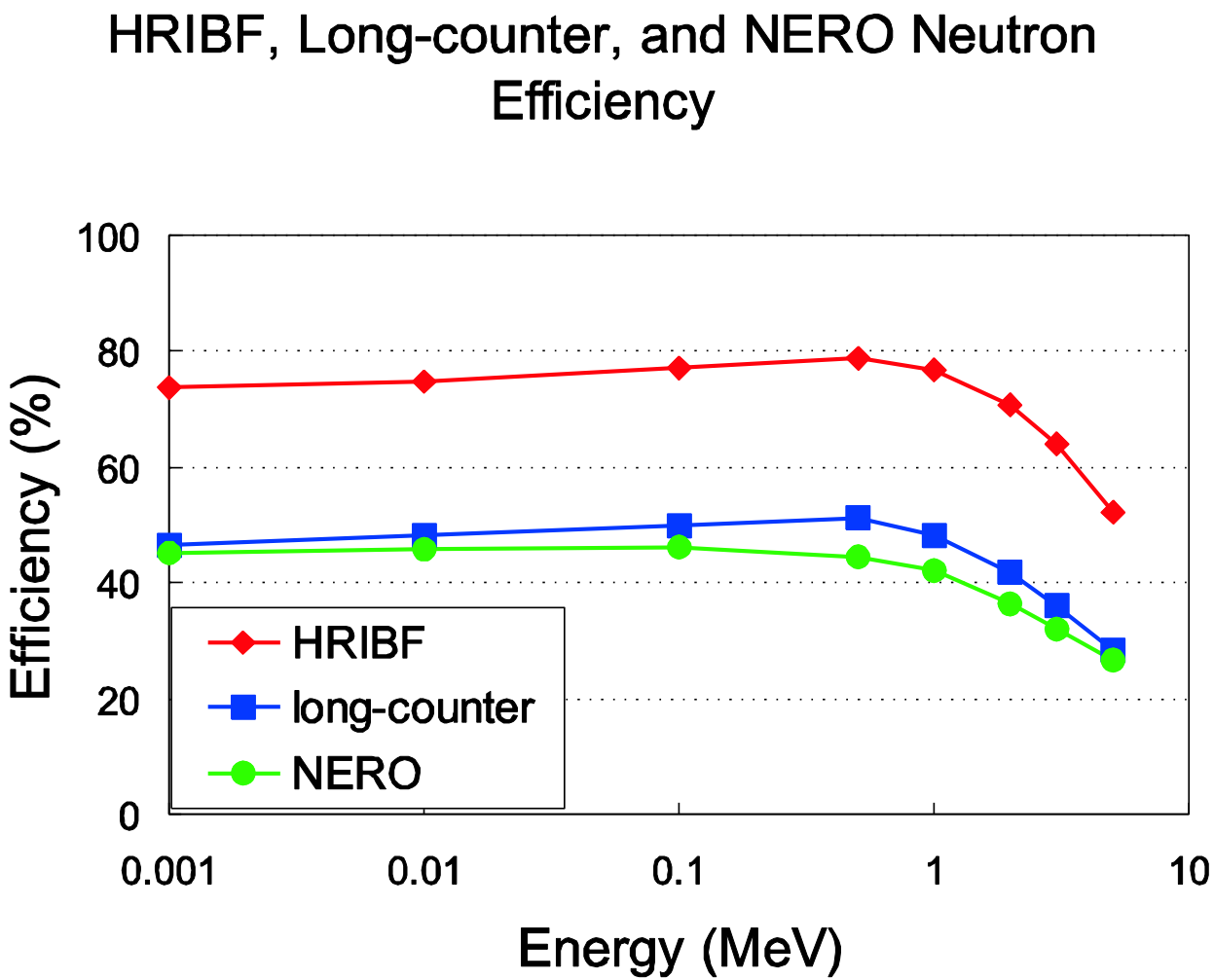
Cadmium Layer Around Edge to Absorb Background Thermal Neutrons

Nuclei Ranged Out in Gas Chamber (First Experiment with Laser Ion Source High Purity Exotic ^{86}Ga Delivered)



Plastic Scintillator β Detectors

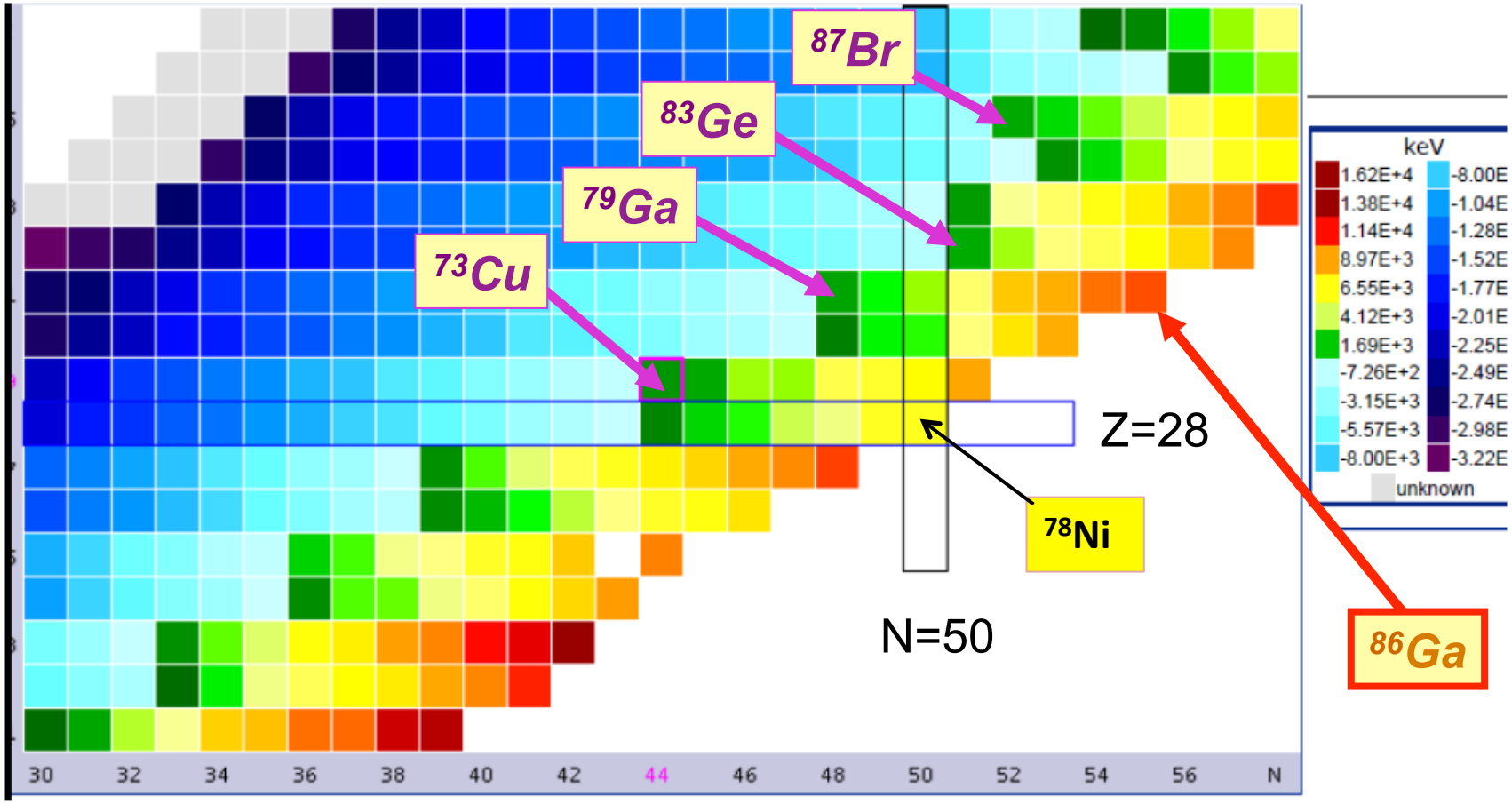
Comparison of 3Hen to Other Detectors



Efficiency curve of full 3Hen detector array (74 3He tubes) compared to the efficiencies of other existing neutron counters like the long-counter of K.L. Kratz and of the NSCL Neutron Observer NERO.

From NNDC :

Energy Window (Q_β - S_n)(Q value – Neutron Separation Energy) for β-Delayed Neutron Emission Precursors



Blue – βn emission not possible

Green – 3Hen $I_{\beta n}$ measurements of low energy βn's

Red-Brown – β-delayed 2n emission (low energy?)

3Hen Collaborators

K.P. Rykaczewski, C.J. Gross, K. Miernik (ORNL Physics Division),
R.K. Grzywacz, K.C. Goetz, M. Madurga, D. Miller, S.V. Paulauskas (UTK),
M. Wolińska-Cichocka (ORAU/ORNL),
I. Gauld (ORNL NST Division),
B.C. Rasco, E. Zganjar (LSU),
J.A. Winger (Mississippi),
A. Fijałkowska, M. Karny, A. Korgul, C. Mazzocchi (University of Warszaw)

Conclusion

Beta decay of neutron rich nuclei is important for understanding nuclear structure, the origins of the elements, and reactor decay heat.

New detector capabilities allow us to study more neutron-rich nuclei with low energy beams

A wealth of new data was acquired at the HRIBF over the last year – now under analysis

Unfortunately the HRIBF has now ceased operations as user facility

ORIC cyclotron being decommissioned

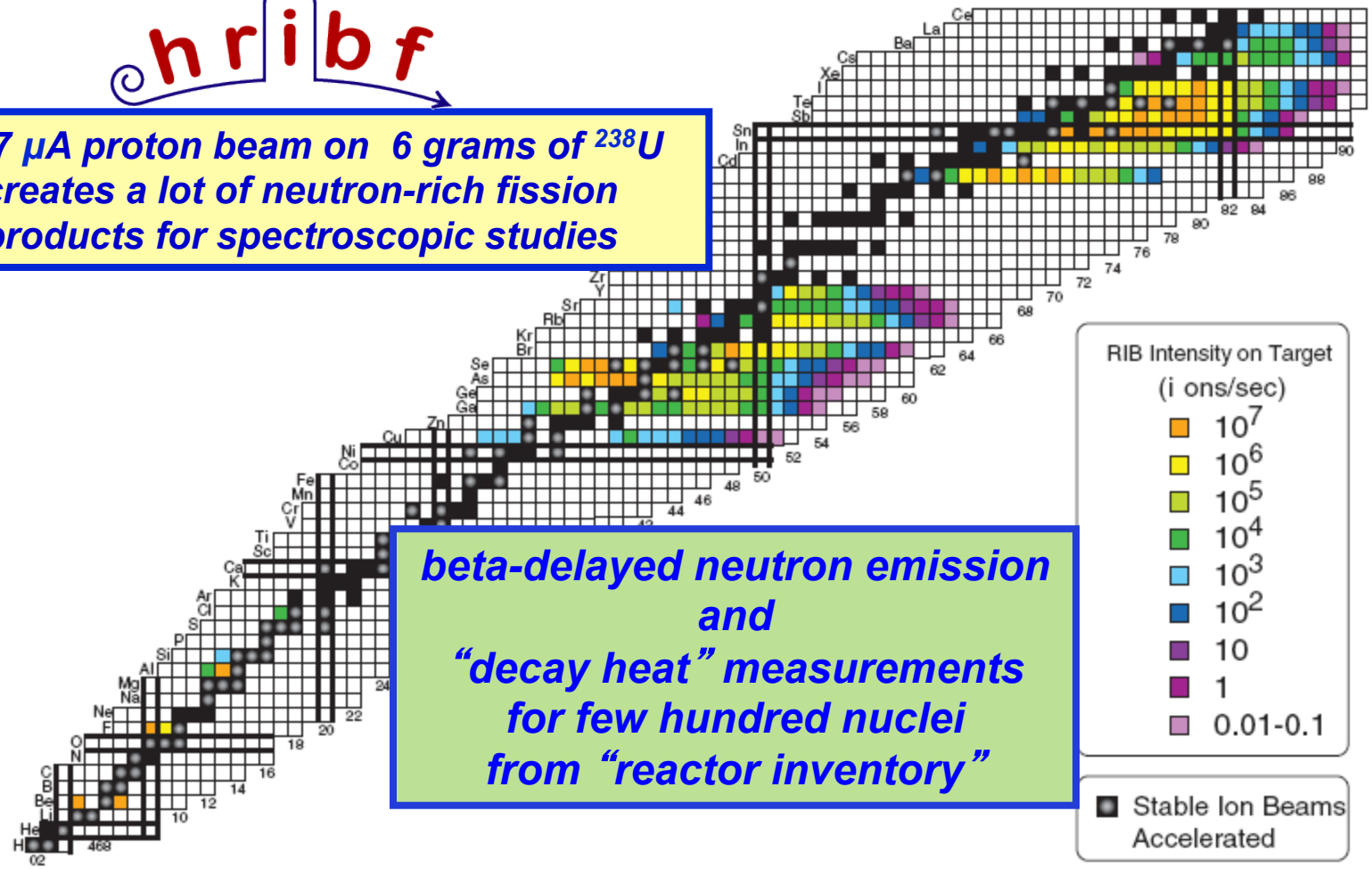
Further Studies using the CARIBU facility at Argonne National Lab and ARIEL at TRIUMF in Canada are Possible

THANK YOU

Backup Slides…

hribf

~ 17 μA proton beam on 6 grams of ^{238}U creates a lot of neutron-rich fission products for spectroscopic studies



beta-delayed neutron emission and "decay heat" measurements for few hundred nuclei from "reactor inventory"

Figure 3. Radioactive and stable beams presently accelerated at the HRIBF. Additional radioactive species such as Zn, Kr, Cd, and Xe are available up to 200 keV as positive ions at LeRIBSS. Stable beams from our new SNICS source [11] are used for RIB development. They are also used to simulate RIBs for tuning and developing new techniques as well as to calibrate detectors.

poster Dan Stracener

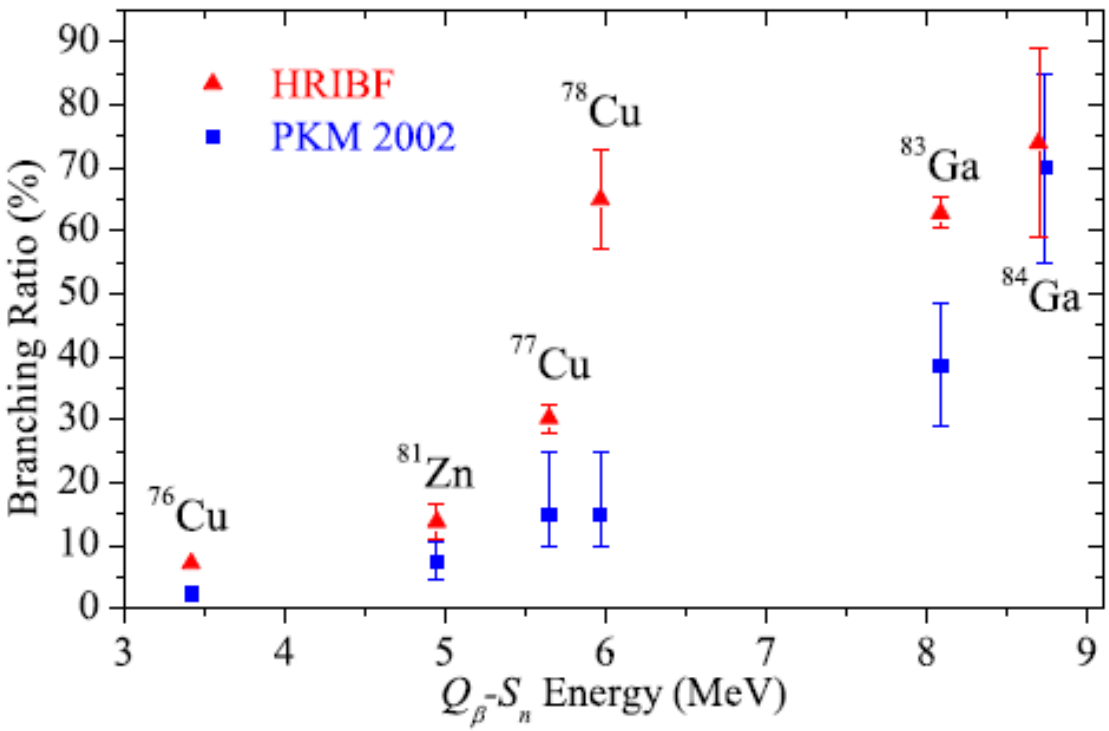
Recent HRIBF data

Winger et al., PRL 102, 142501 (2009); *PR C 80, 054304 (2009); PR C81, 044303 (2010), PR C82, 064314 (2010), PR C83, 014322 (2011)*

are pointing to much higher βn-branching ratios in the ***^{78}Ni region***

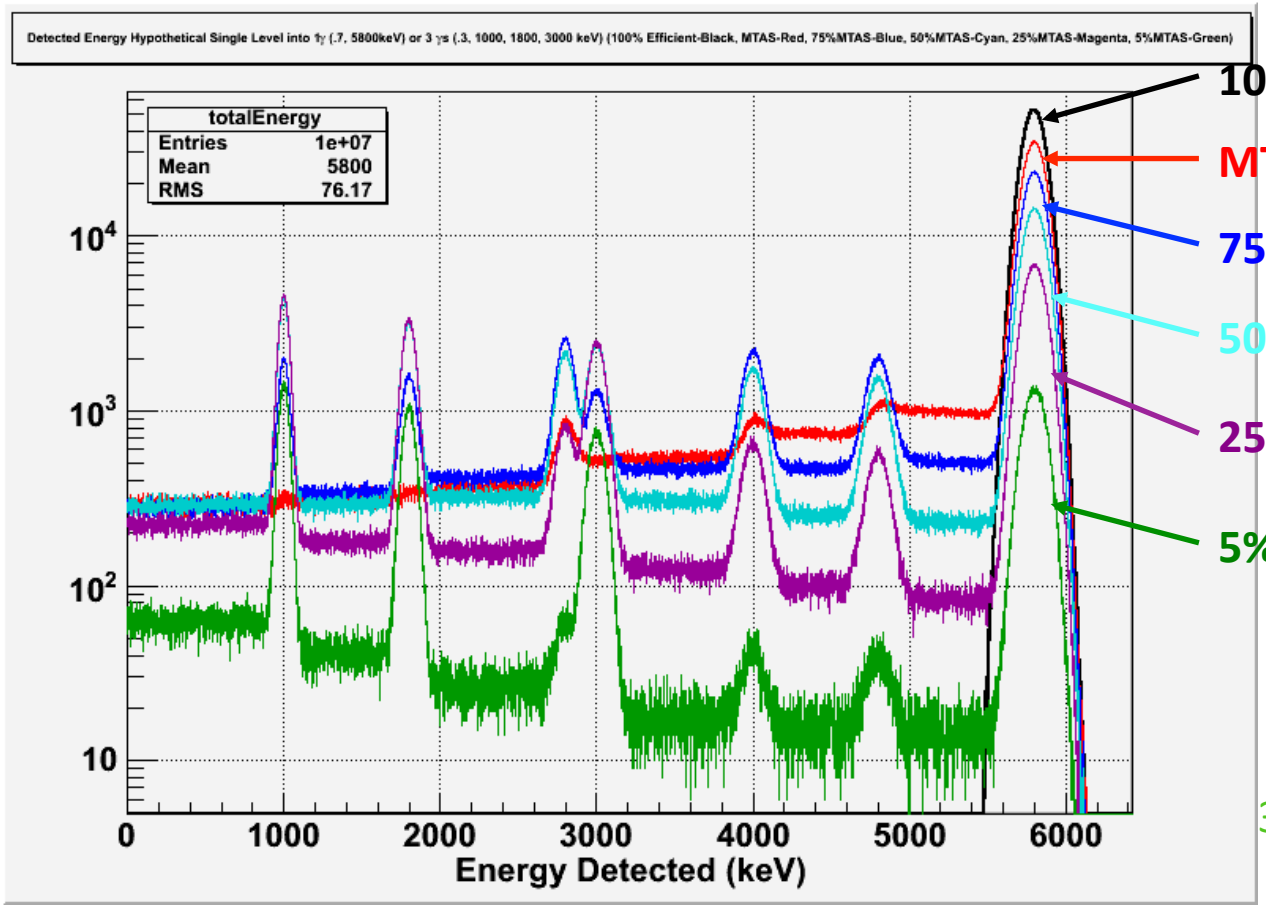
in comparison to earlier measurements and calculations,

see, e.g., ***Pfeiffer, Kratz, Moeller*** **(PKM 2002)** ***Prog. Nucl. Energy, 41, 5 (2002)***



Similar conclusion from a recent NSCL paper basing on NERO results
P. Hosmer, H. Schatz et al., PR C82 , 025806, 2010

starts to be relevant for nuclear fuel cycle ?!



100% Efficient Detector

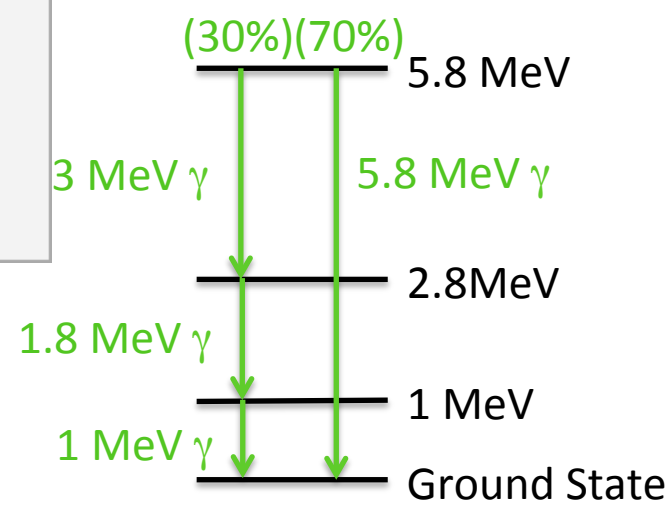
MTAS

75% Solid Angle Coverage MTAS

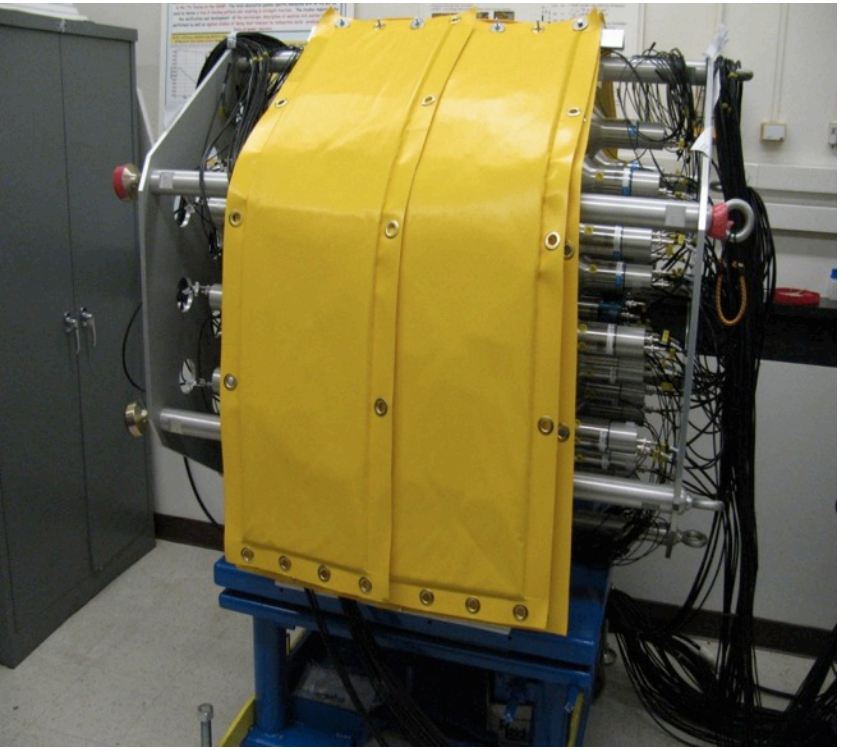
50% Solid Angle Coverage MTAS

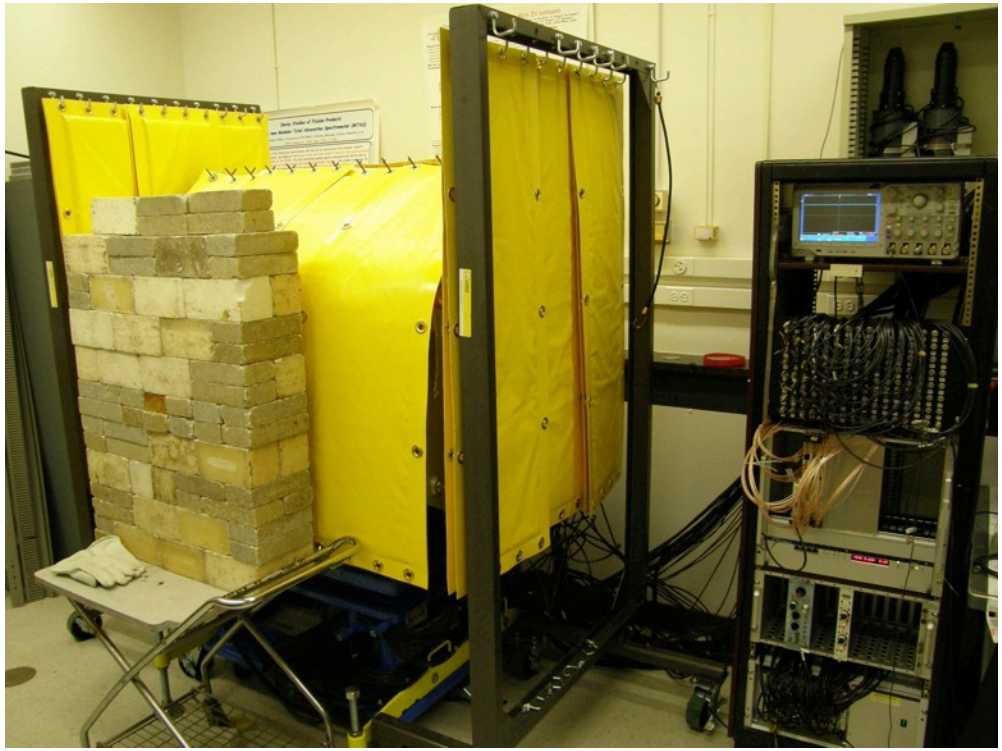
25% Solid Angle Coverage MTAS

5% Solid Angle Coverage MTAS



MTAS Shielding



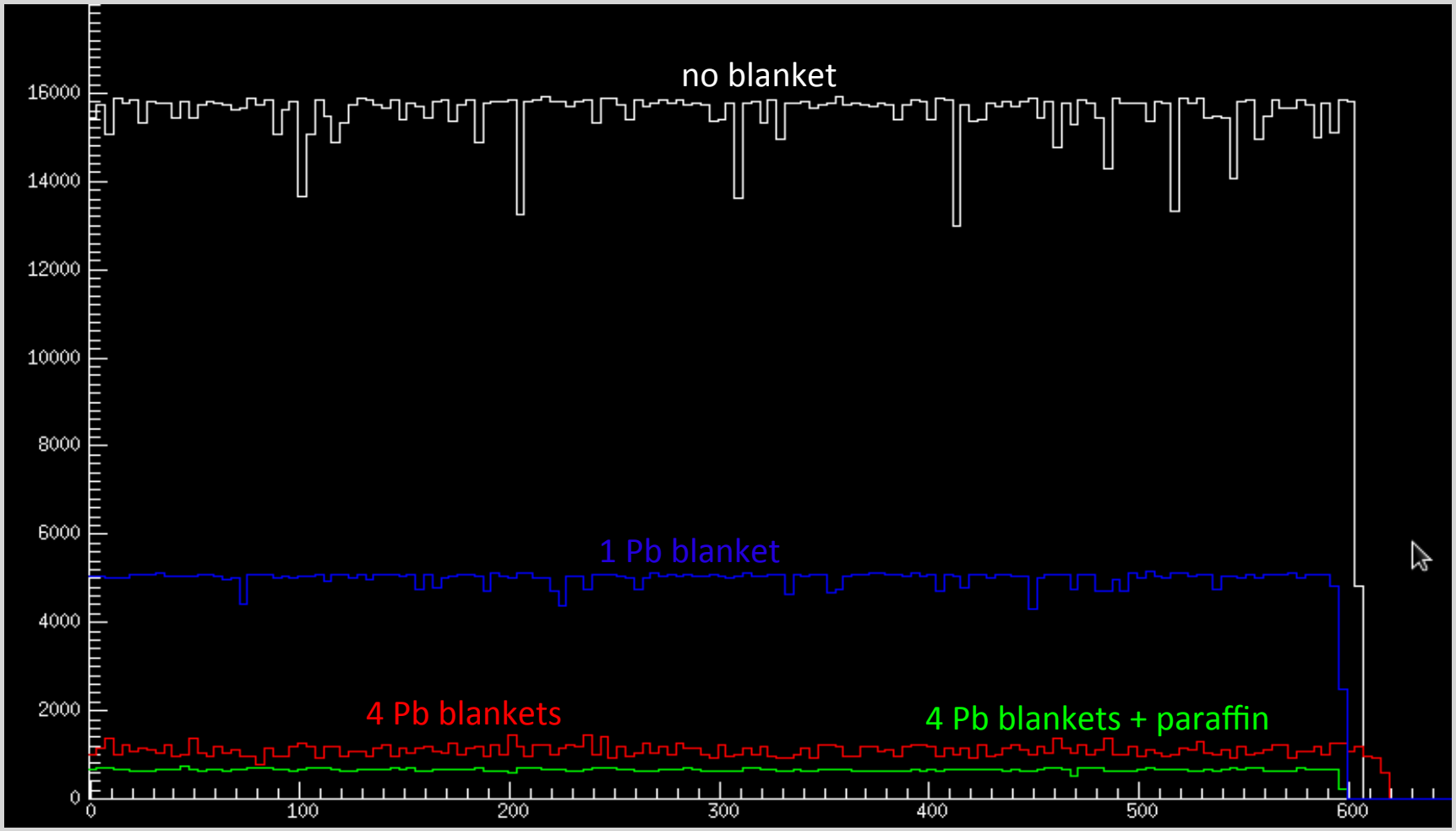


"light" lead shielding
lead wool blankets
~ ¾" lead equivalent

MTAS test stand was assembled
next to the radioactivity storage room
in order to test data transfer speed

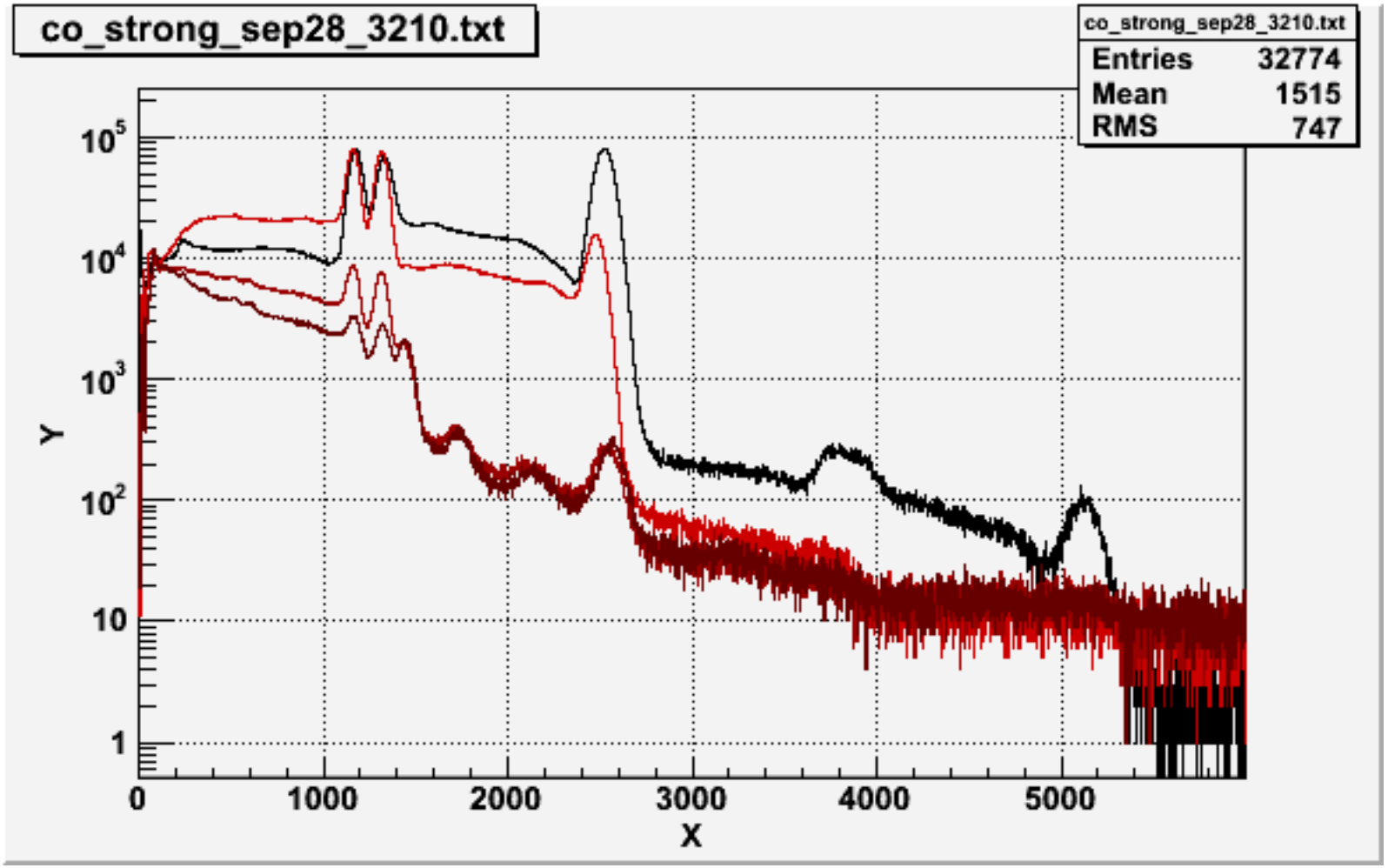
Shielding Tests

Background level per module went down from ~ 16000 Hz to ~600 Hz

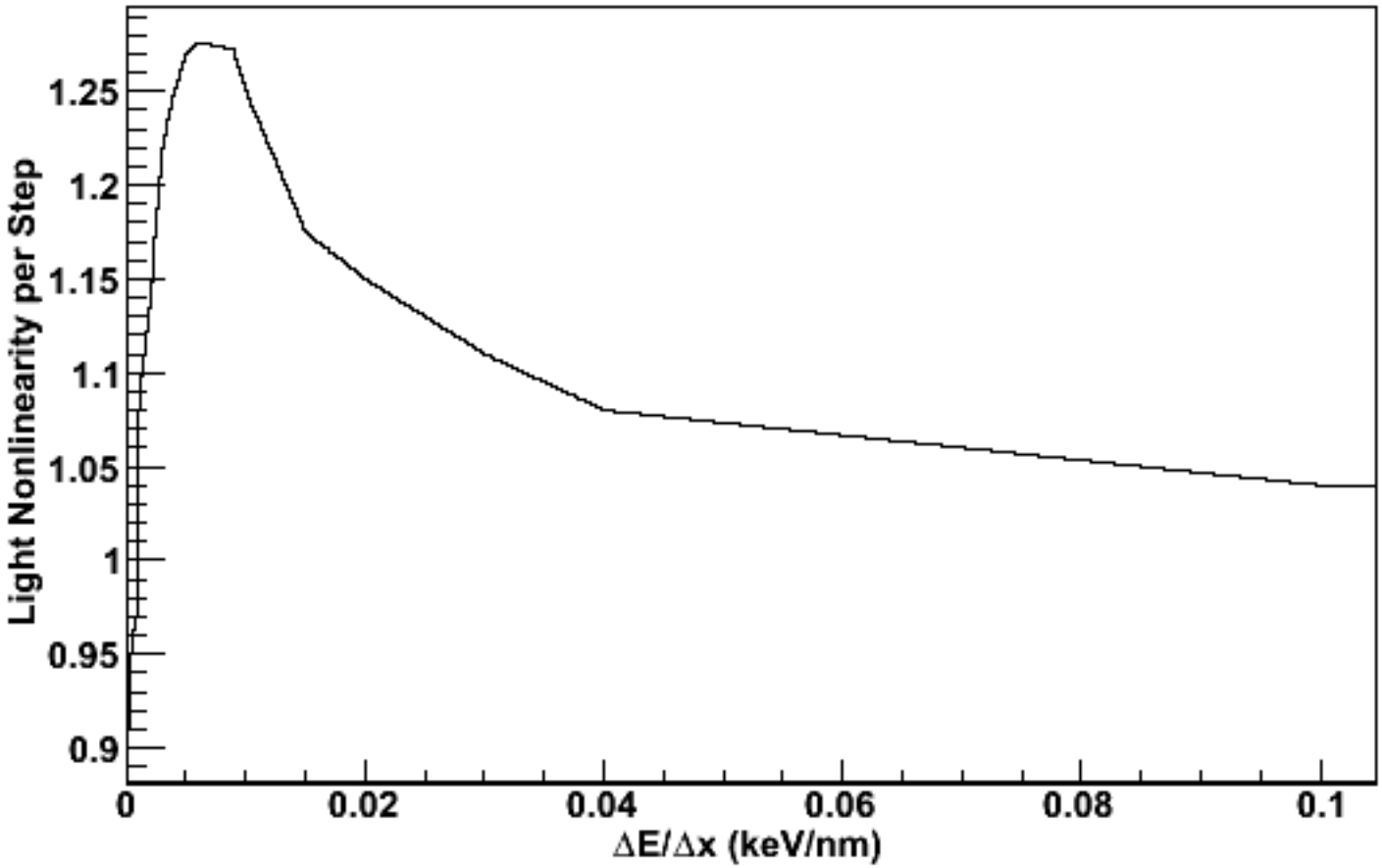


MTAS Tape Drive System

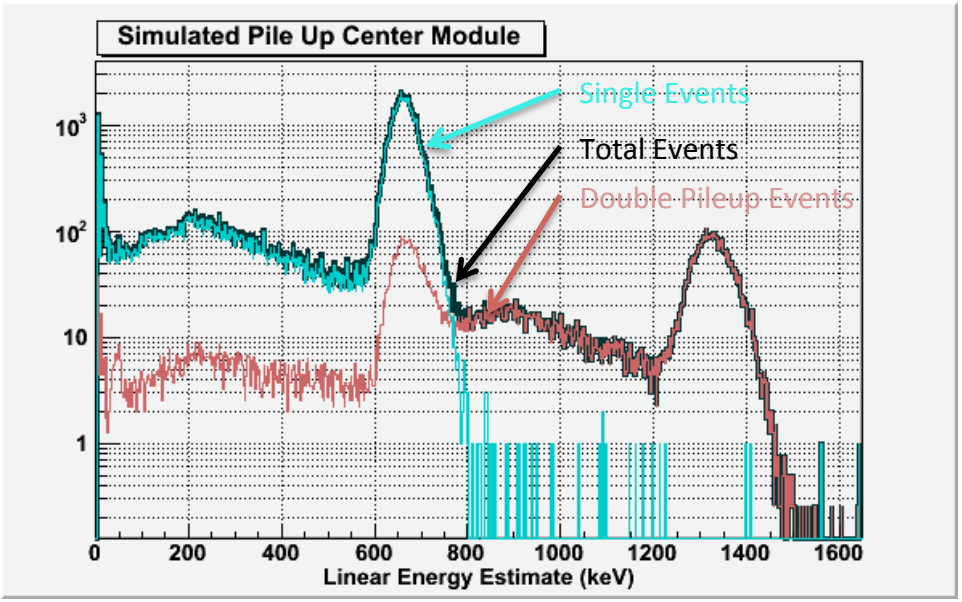
^{60}Co Data By Ring



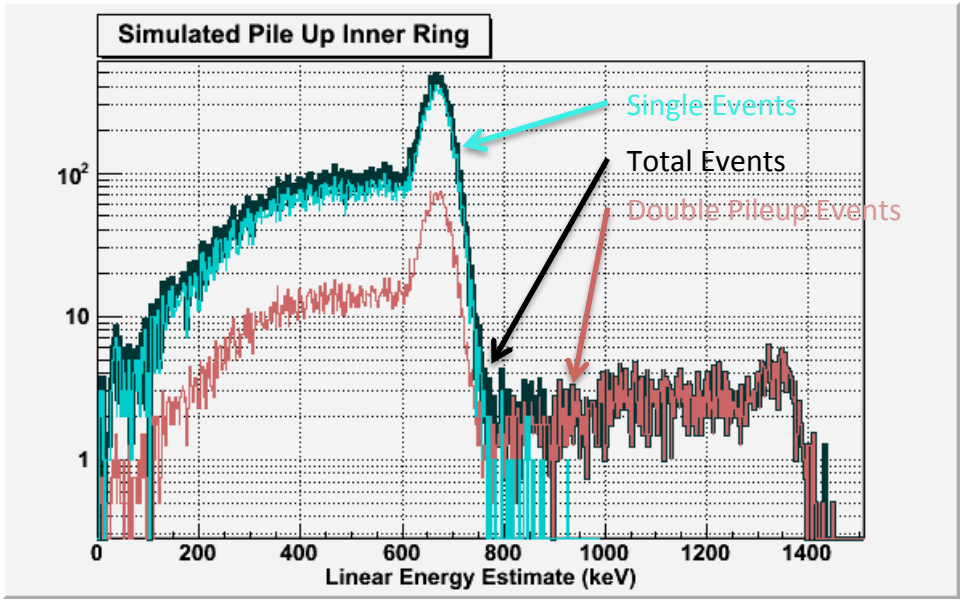
dE/dx Curve Calculated from and Used in GEANT



Simulation of Light Production in NaI(Tl) Event Pile Up



Simulated pileup of one and two event ^{137}Cs in the central module



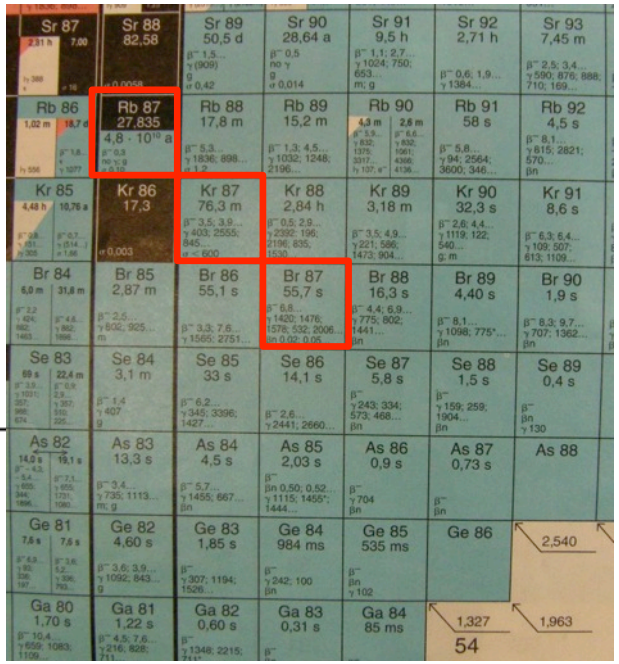
Simulated pileup of one and two event ^{137}Cs in the inner ring

The probability for the next event to occur is given by $P(t) = e^{-\lambda t}$ λ-source activity

If this time is less than the digitizer acquisition time then a pileup has happened (with no event rejection)

For decays with changing activity, which includes most of the planned MTAS experiments, pileup peak heights will decrease as the activity decreases.
Comparison of relative peak heights versus time distinguishes pile up peaks from real peaks.

^{87}Kr Info



^{87}Kr β− Decay 1971Sh01,1973BlZH,1973GeYV

Decay Scheme

Intensities: I(γ+ce) per 100 parent decays