Decay Studies of Fission Products with a new Modular Total Absorption Spectrometer (MTAS)

$ 1.58 M project, FY 2010 – FY 2012

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(ORNL and UTK)
PhDs: *B.C. Rasco (LSU), M. Wolinska (ORAU/ORNL), M. Karny (ORAU/Warsaw)*
PhD students: *A. Kuzniak (UTK/Warsaw) , K. Goetz (UTK, Center for Interdisciplinary Research and Graduate Education – CIRE)*

GOALS :

*A **Modular Total Absorption Spectrometer (MTAS**)*
will be constructed, characterized
and applied to the decay studies
with pure beams of neutron-rich nuclei.

*The studies important for the **verification and development of nuclear structure models** will be performed as well as applied measurements of*
decay heat
released by radioactive nuclei produced in nuclear fuels at power reactors.

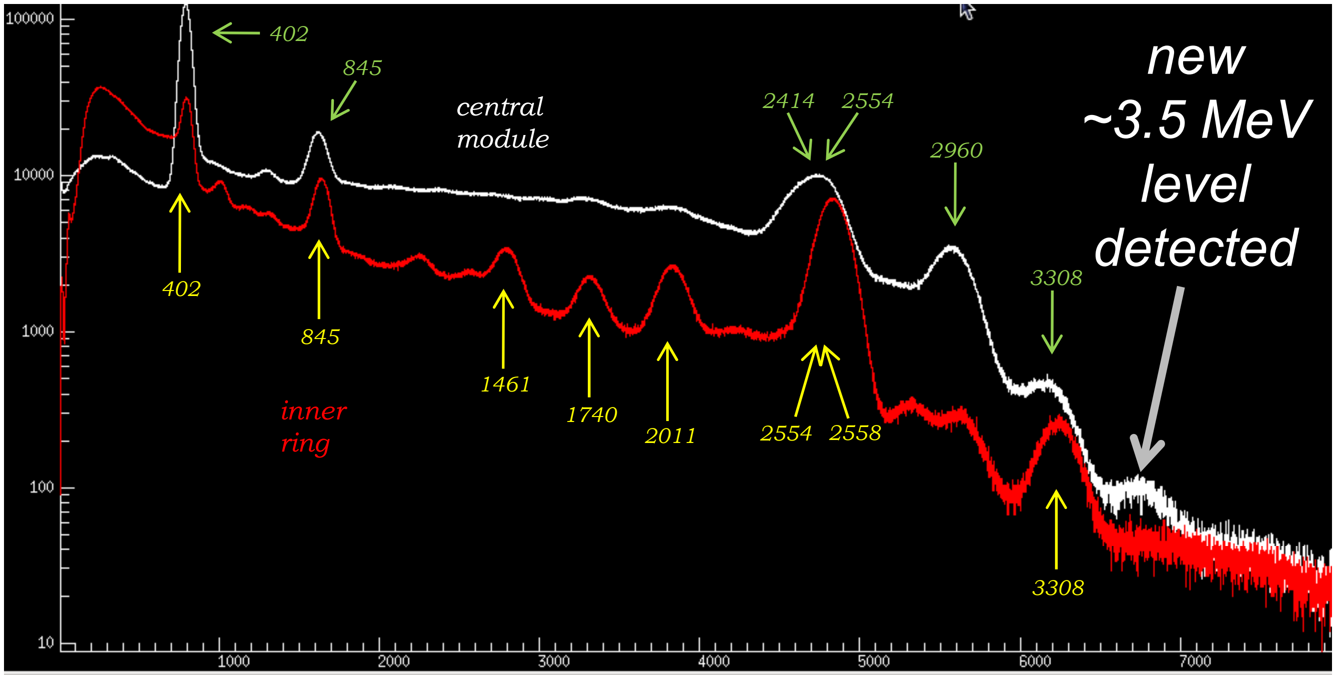
U.S. DEPARTMENT OF ENERGY

Holifield Radioactive Ion Beam Facility
Oak Ridge National Laboratory

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

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



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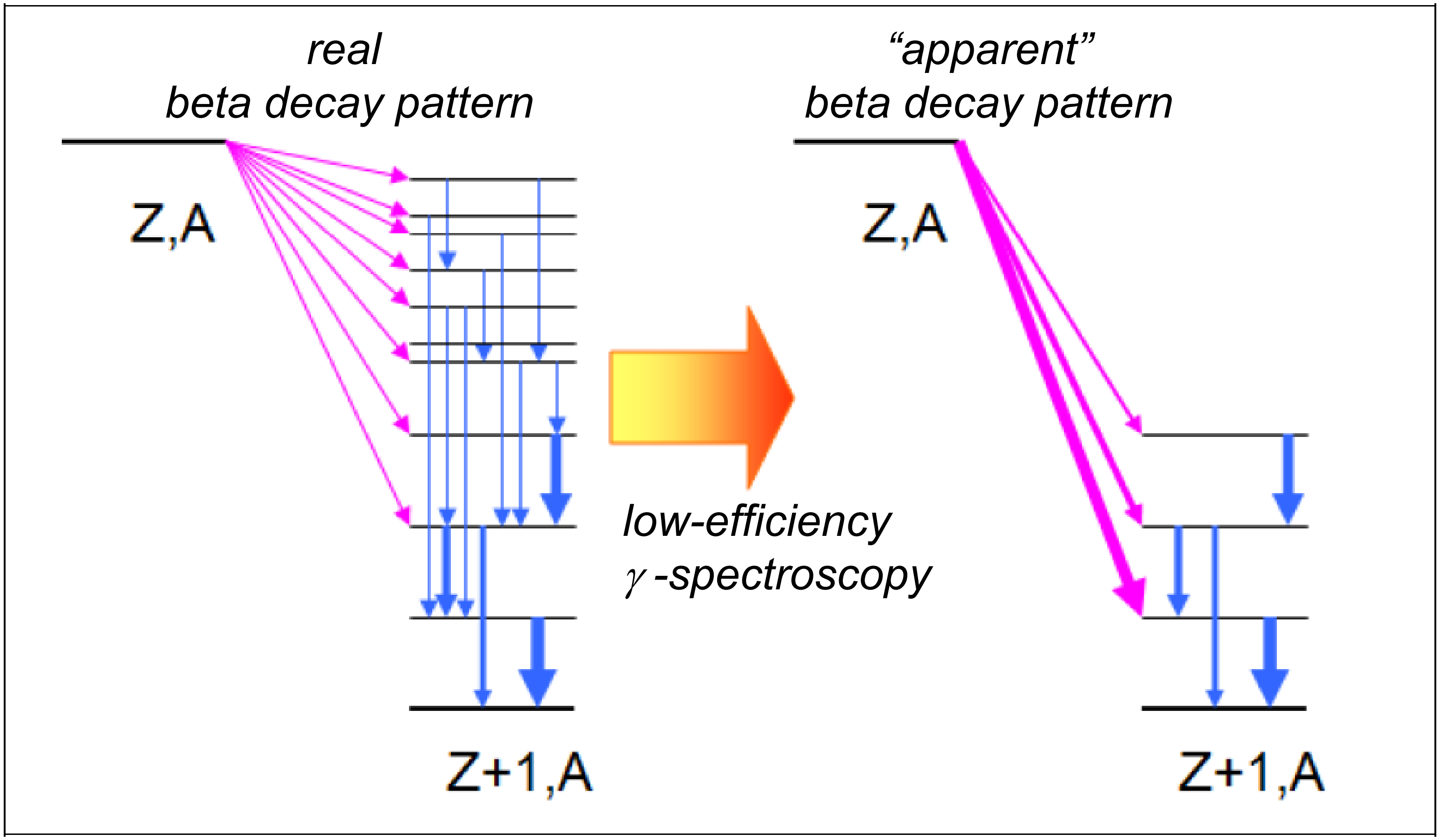
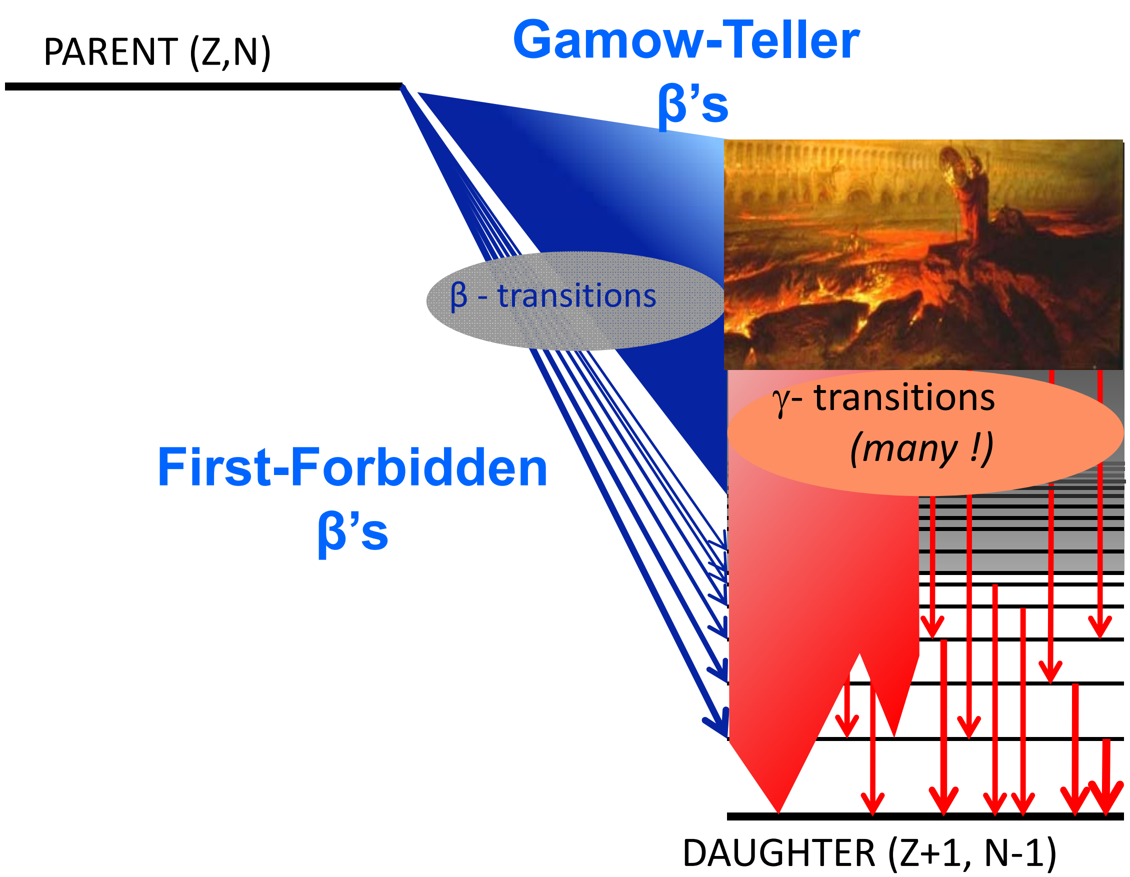
Why do we need Total Absorption Gamma Spectroscopy to study the decays of neutron-rich nuclei (fission products) ?



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

Adopting "apparent" decay pattern results in a wrong interpretation of beta decay properties and respective nuclear structure. The energies and intensities of emitted γ-rays and electrons (decay heat) are wrongly estimated !

In fact, the true βγ-decay pattern is even more dramatic for very neutron-rich nuclei



The illustration of
"Pandemonium Effect"
as discussed by
J. Hardy et al. ,
Phys. Lett. 71 B, 307, 1977

The true decay picture
for the neutron-rich
parent nucleus (Z,N),
with many weak β-transitions
and
following low intensity
γ-transitions.

"Pandemonium" credits: J. Milton (1667) and J. Martin (1825)
(see K. Rykaczewski, Physics 3, 94, 2010)

Total Gamma Absorption Spectroscopy is needed
to correctly establish such complex decay scheme

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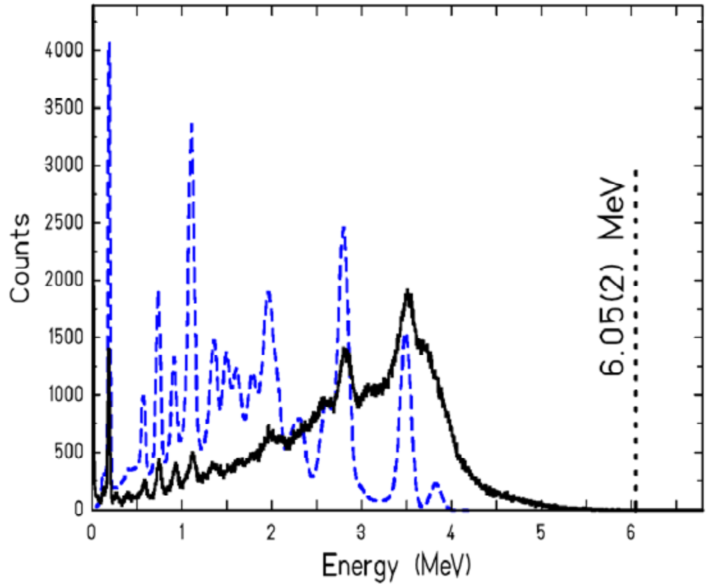
The problem and its importance is well recognized in the nuclear structure studies and reactor physics



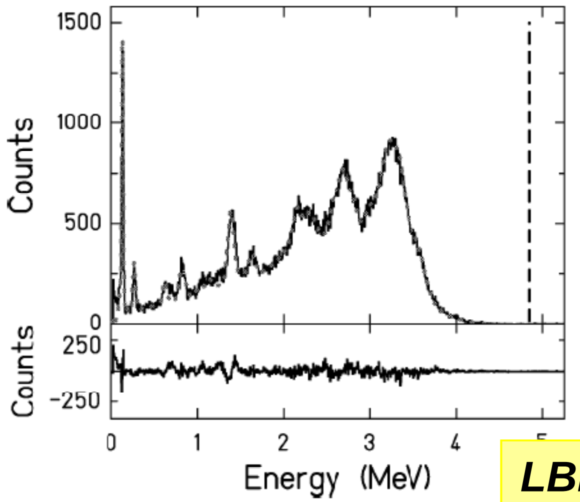
γ - ray spectra for the decay of ^{103}In
M. Karny.., Grzywacz.., Rykaczewski, Nucl. Phys. A640, 3 (1998).

*The **blue line** represents the simulated TAGS spectrum, calculated using a decay scheme derived from the high-resolution, but low-efficiency measurement.*

*Gamma spectrum measured with the LBNL Total Absorption Gamma Spectrometer (TAGS) is given as a **black line**.*

Note large missing strength at high excitation energies !

and there is a solution: total absorption gamma spectroscopy



*The gamma energy spectra **measured** with TAGS (solid lines) and **calculated** (dotted lines) after the de-convolution procedure leading to the true Electron Capture transitions feeding patterns and to the correct decay scheme of ^{105}In*

M. Karny , .., Grzywacz, .. Rykaczewski, Nucl. Phys. A690, 367 (2001).

The lower panel shows notably small differences between measured and calculated spectra.

***LBNL TAGS at GSI 1997-2003: M.Karny et al., NIM B126 (1997) 411 and over 20 papers with physics results** (^{100}Sn, ^{146}Gd regions)*

B. Rubio and W. Gelletly, Lectures at the Euroschool on Exotic Nuclei :

During the operation of power reactors some 7–8% of the total power comes from the β-decay of the very large number of neutron–rich fission products.

A knowledge of how the decay heat varies with time is important because

(a) it is necessary for economic reasons to optimize the re-fuelling procedure,

(b) one needs to maintain cooling because of the decay heating,

see I.C. Gauld (ORNL) – Decay heat predictions for LOCA analysis (2006)
recent exp data : Fukushima - Daichi nuclear power plant (2011)

(c) when the fuel is finally removed from the reactor it must be properly shielded.

Licensing processes involved in nuclear fuel cycle !

Assessment of fission product decay data for decay heat calculations

coordinator: T. Yoshida, monitor: A.L. Nichols,

*Nuclear Science NEA/WPEC-25, ISBN 978-92-64-99034-0, **NEA** No. 6284,*

Organization for Economic Cooperation and Development OECD 2007

DOE NUCLEAR ENERGY RESEARCH AND DEVELOPMENT ROADMAP (April 2010)

5.4 Interfaces and Coordination (page 45):

The use of a "science-based" approach to develop innovative nuclear energy systems and components requires a strong collaboration between NE and the Office of Science (SC) to employ the tools developed for science in engineering applications. Such tools include advanced experimental techniques, a fundamental understanding of materials behavior, and advanced computational sciences.

ORNL is a home of first nuclear energy innovation hub,

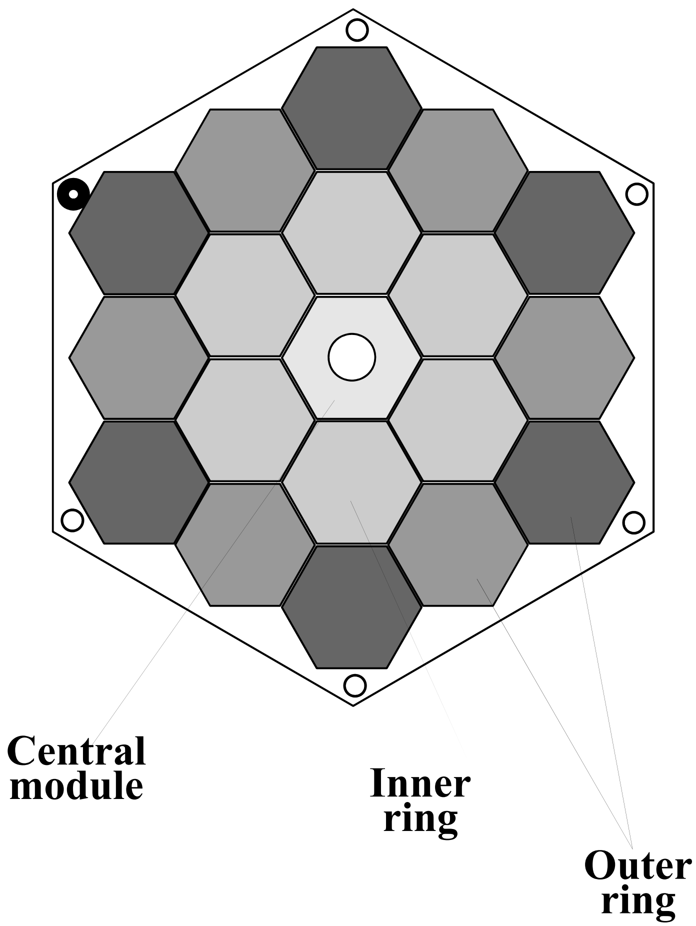
*the **Consortium for Advanced Simulation of Light Water Reactors (CASL, 2010).***

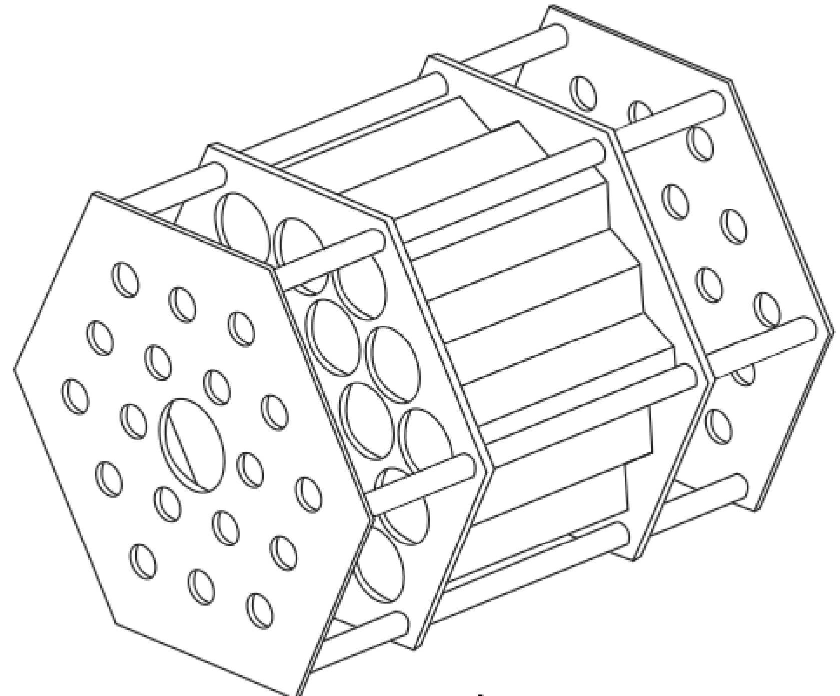
Also, the nuclear energy related codes,

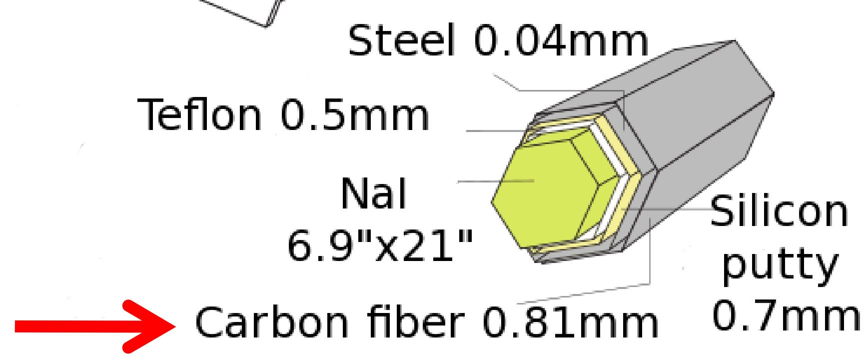
***SCALE** and **ORIGen**,*

*are maintained at ORNL since **1976.***

MTAS final design: 19 hexagonal modules, with a central module having a 2.5” hole to accommodate a beam line and auxiliary beta detectors. Each module is 21” long, 8” diameter (6.9” face-to-face) and has custom made carbon fiber housing. Weight ~120 lbs/module, ~2200 lbs total.







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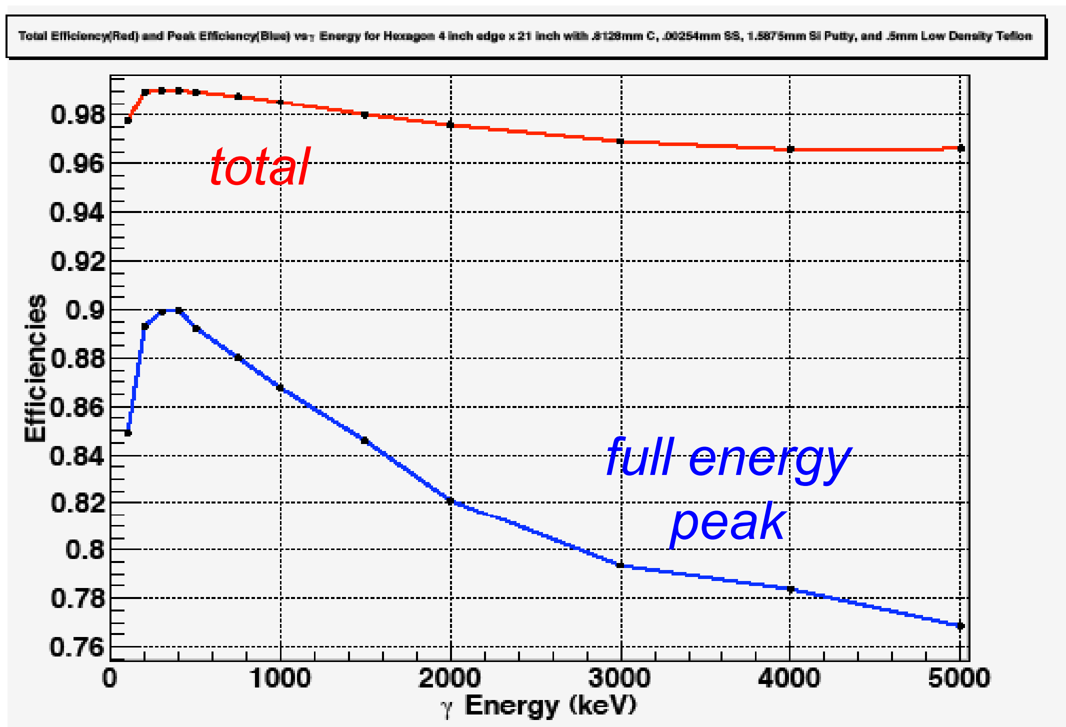
Efficiency simulations for MTAS

Total efficiency and full energy γ-efficiency (related to the active volume) are critical parameters for **Total Absorption Spectrometry**

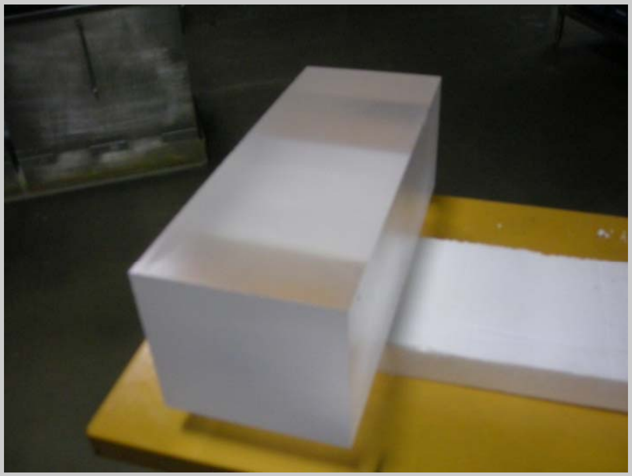


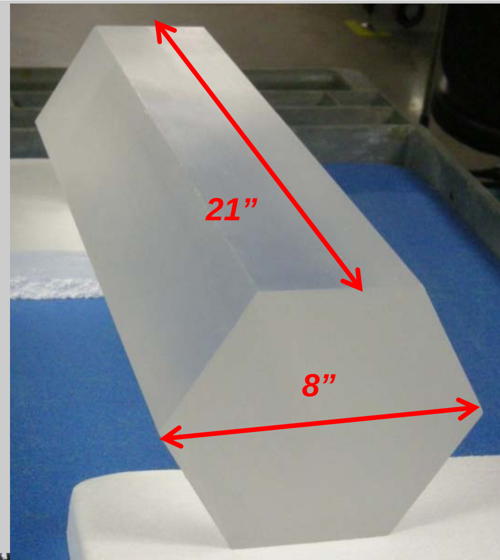
MTAS γ-efficiency
GEANT4, B. C. Rasco (LSU)

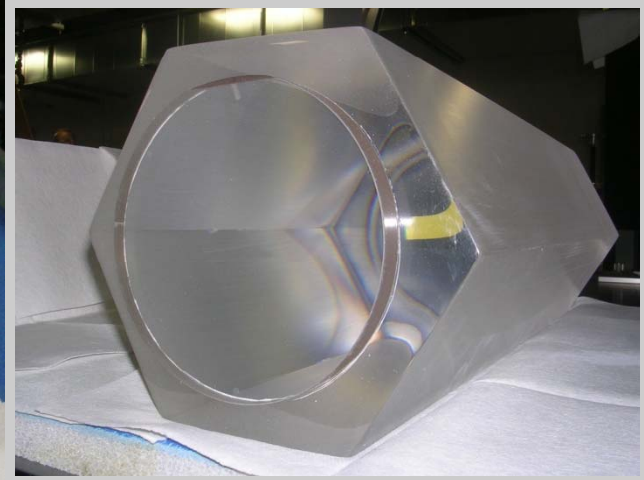
Idaho TAS : (1990's → ANL) : ***MTAS has 17 times larger volume***
LBNL TAS : GSI (1995 – 2003) → LBNL : ***MTAS is 7 times larger***
Valencia-ISOLDE "Lucrecia" TAS (since ~ 2003) : ***MTAS is 8 times larger***

PRODUCTION CYCLE OF MTAS MODULES

All MTAS crystals were grown the U.S., encapsulated and initially tested at the Saint Gobain Crystal plant in Hiram (OH).







test station SGC ~ August 2010

ASSEMBLING MTAS ARRAY

All NaI(Tl) modules were assembled in the support frame at SGC (December 2010). The array was shipped to Oak Ridge (January 2011) on air-conditioned truck. At ORNL, MTAS has been tilted by 90 degrees (February 2011) and placed at the heavy-duty adjustable-height support cart.

SGC, Hiram (OH), October 2010

HRIBF, Oak Ridge (TN), February 2011

SCIENCE OF MTAS RESPONSE FUNCTION

Many processes add to the width σ of the γ-line in MTAS spectrum , with the spread due to the light propagation and non-linear light generation being the dominant factors.

$$\sigma^2_{total} = \sigma^2_0 + \sigma^2_1 + \sigma^2_2 + \sigma^2_3 + \sigma^2_4$$

inherent spread in number of photons

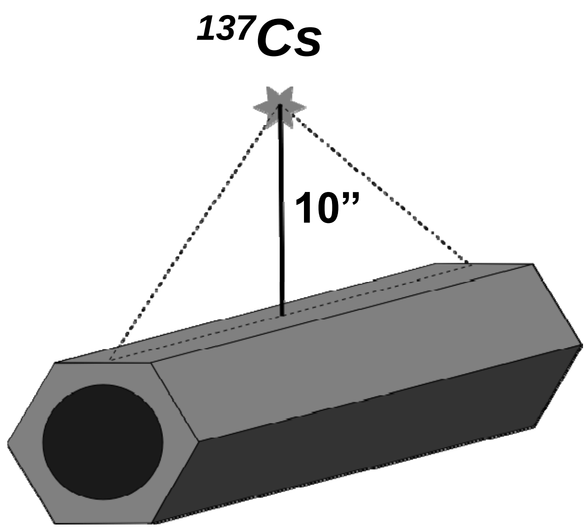
spread due to light propagation that is dependent on the location of energy deposit

Nonlinear generation of light (quenching)

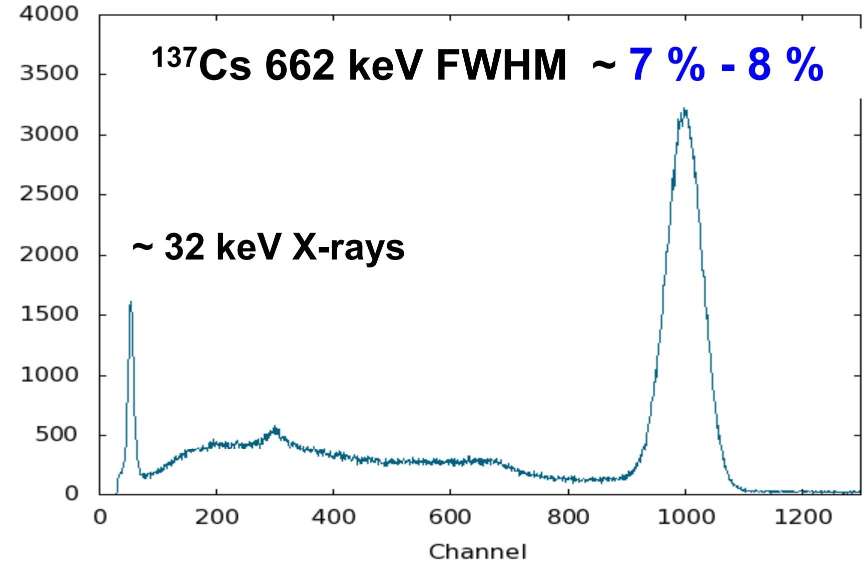
PMT amplification

spread due to electronics

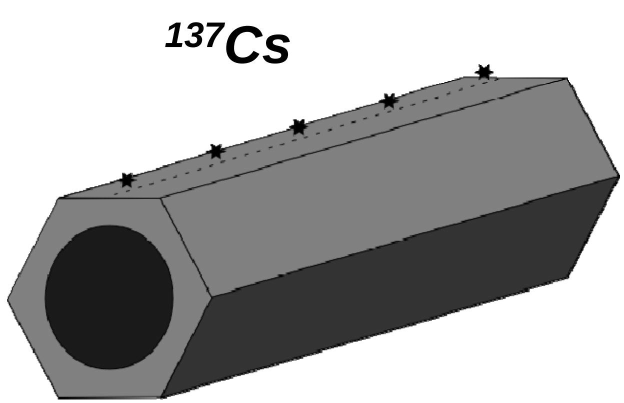
TEST MEASUREMENTS WITH UNCOLLIMATED ^{137}Cs SOURCE

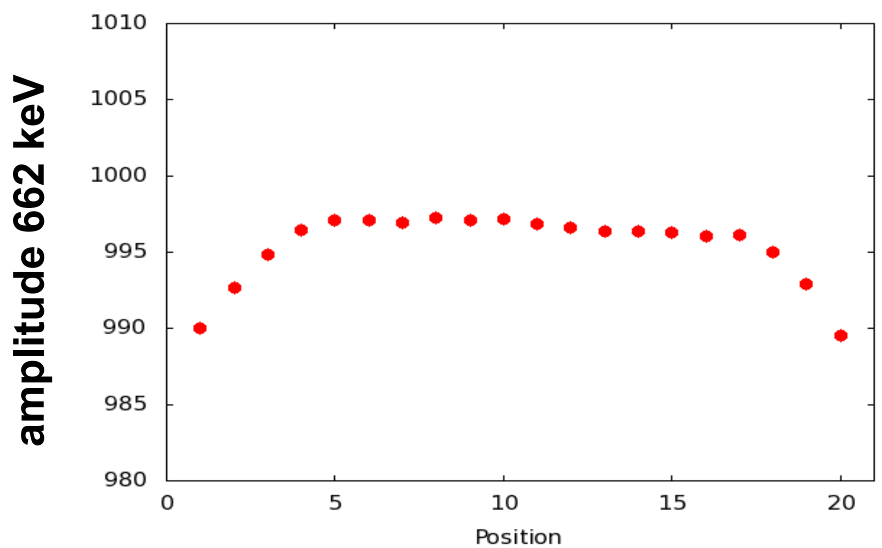


SGC specifications: ***FWHM < 8.5%***

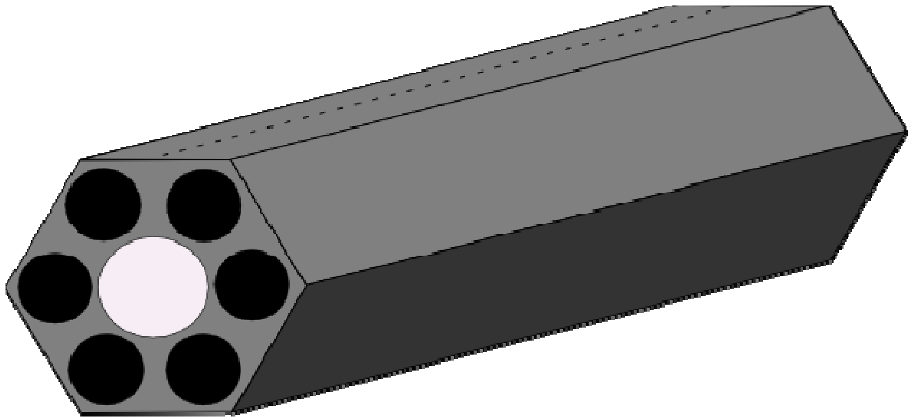


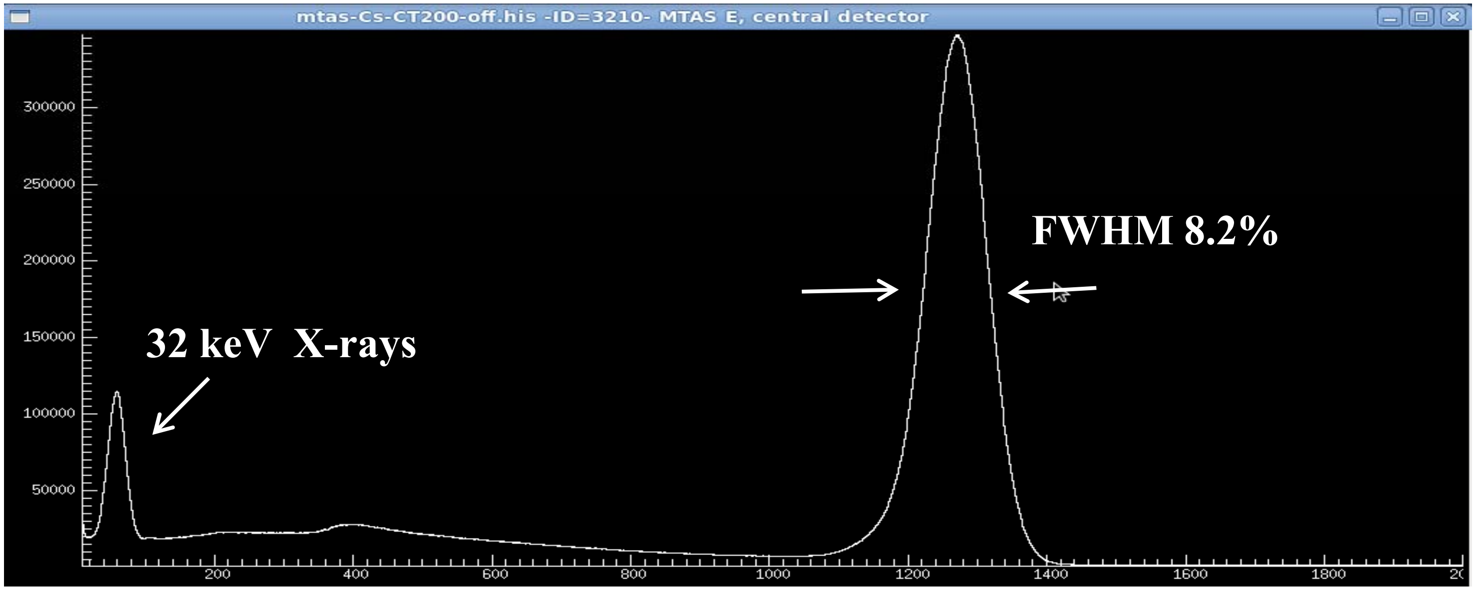
Uniformity of each crystal was measured with ^{137}Cs source placed close to each side (19 measurements per each side, 1” steps)





The FWHM for 662keV in MTAS central module is about 8.2 % and the energy threshold for all 12 summed signals is below 30 keV





SGC manufacturer specs:

FWHM (662 keV) < 15% with a goal to achieve <11.5 %

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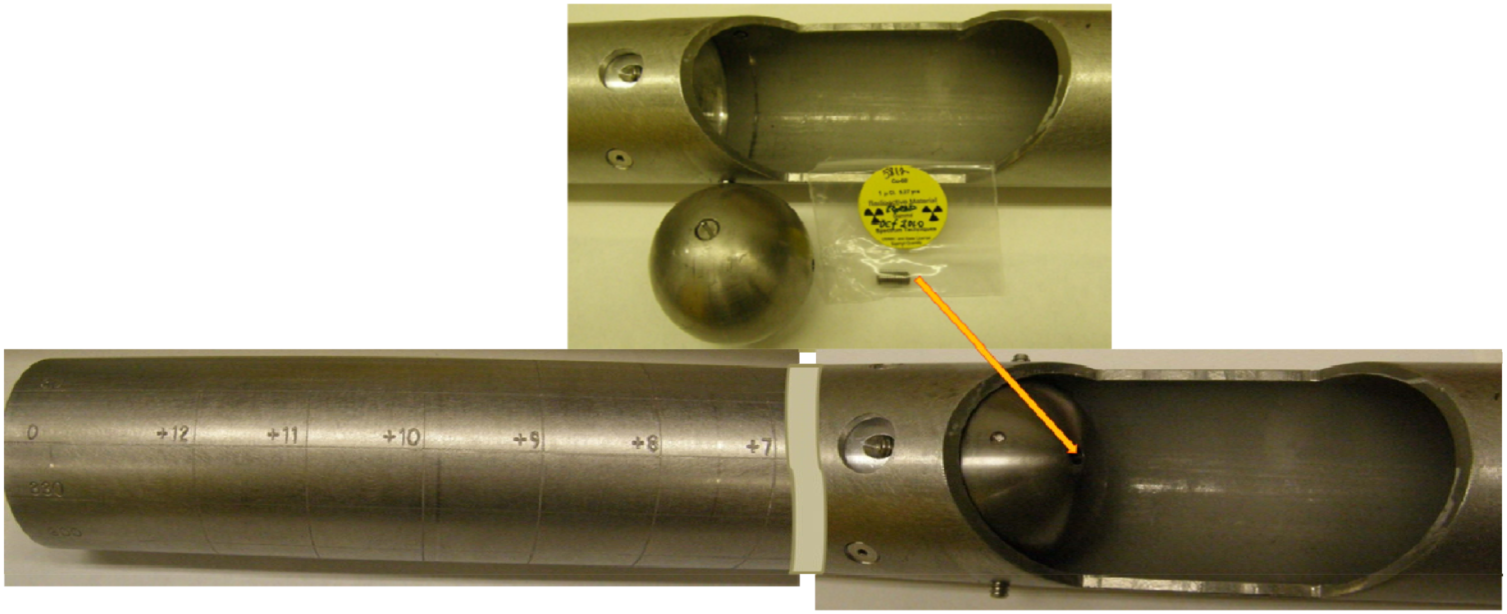
MTAS scanner device (~ 80” long, 2” diameter) with collimated custom-made ^{137}Cs and ^{60}Co sources



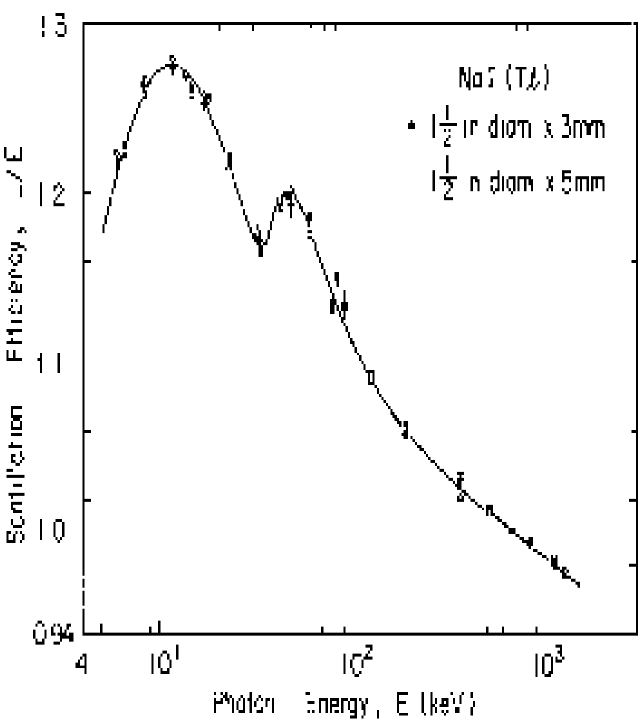
^{137}Cs and ^{60}Co activities are centrally mounted
in the middle of adjustable-position heavy-metal ball.

Measurements covering the angle of radiation
and the positions along the MTAS axis
are used to establish/test the position dependence of MTAS response

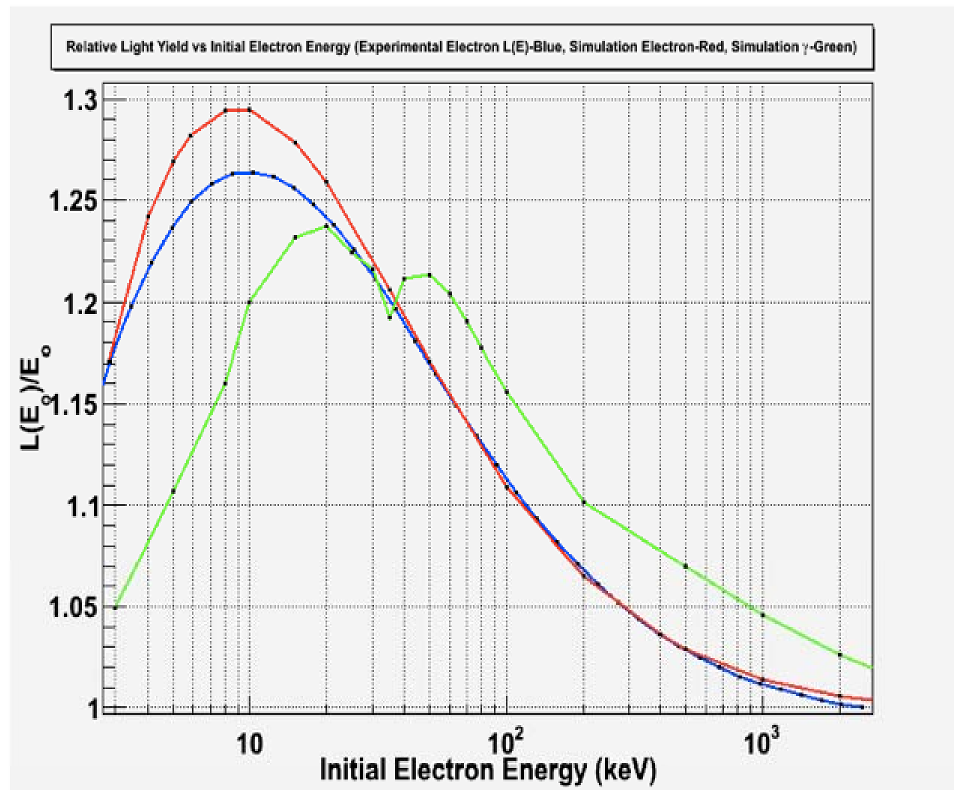
INTRODUCING NONLINEAR LIGHT GENERATION INTO GEANT4

Simulated light production function for gamma rays resembles the experimental shape.

T. Tojo, NIM A238 (1985)

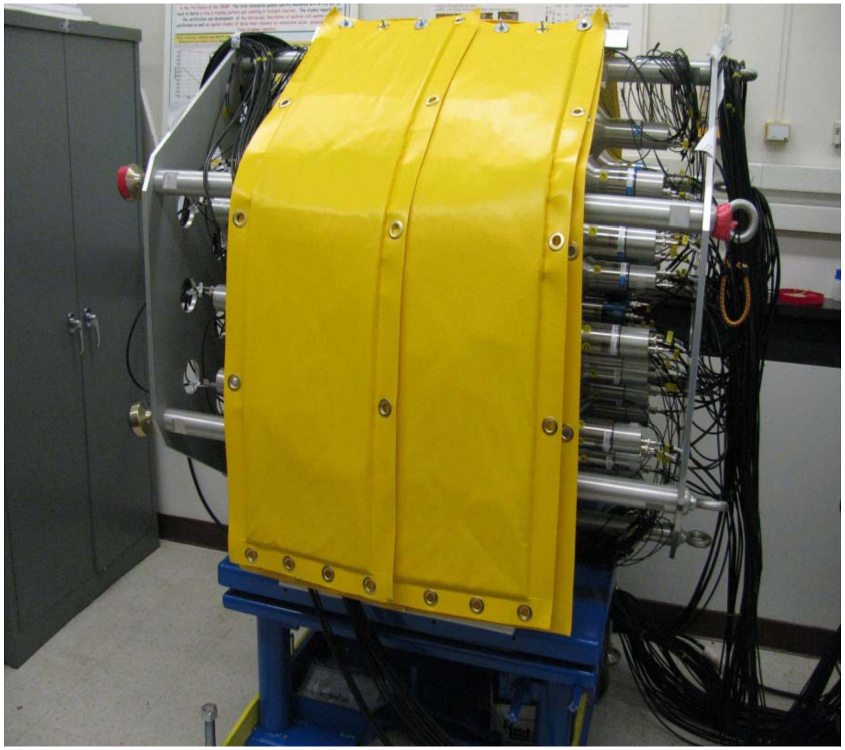


B.C. Rasco (LSU, 2011)



photon energy

SHIELDING



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

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

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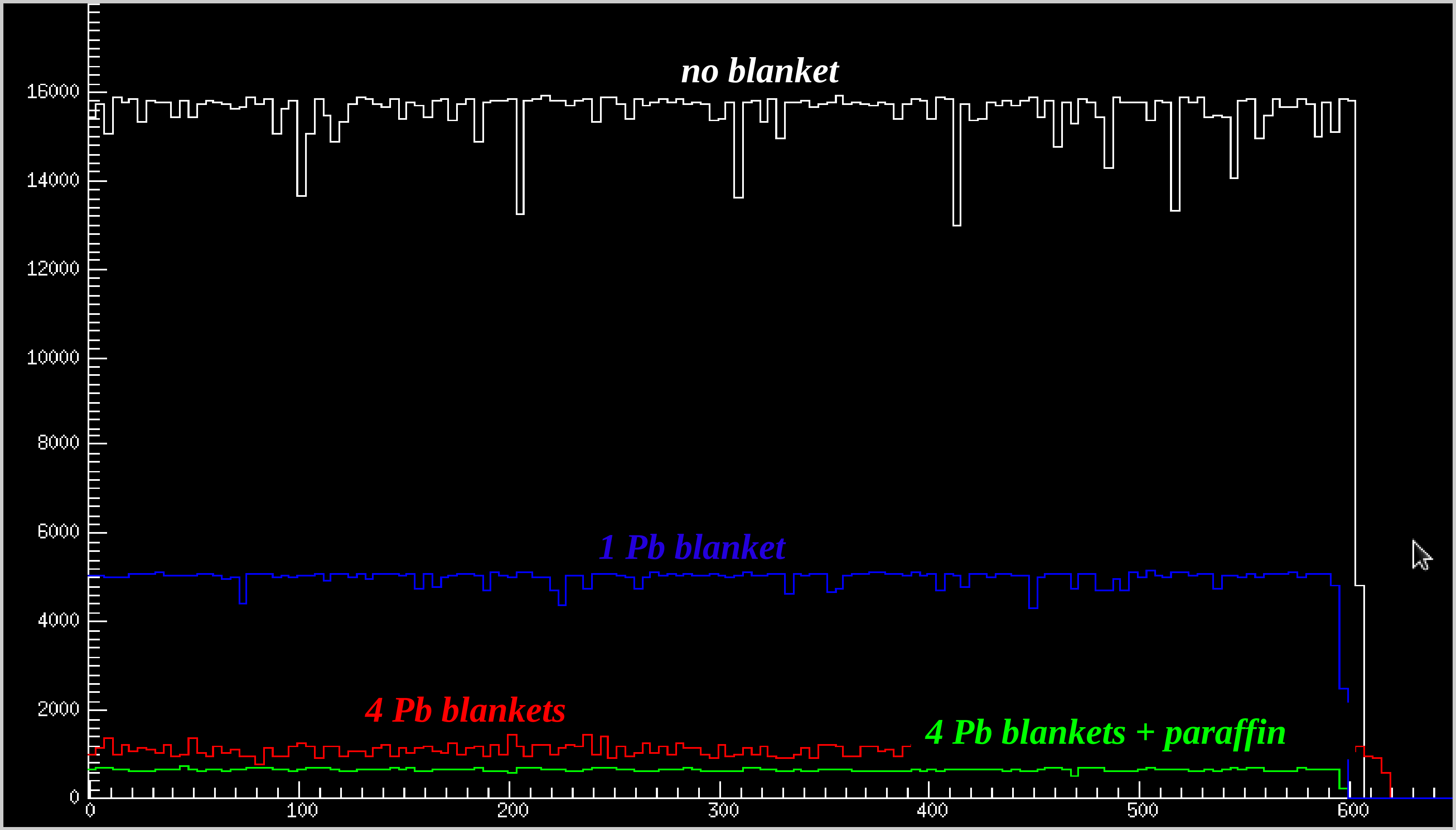
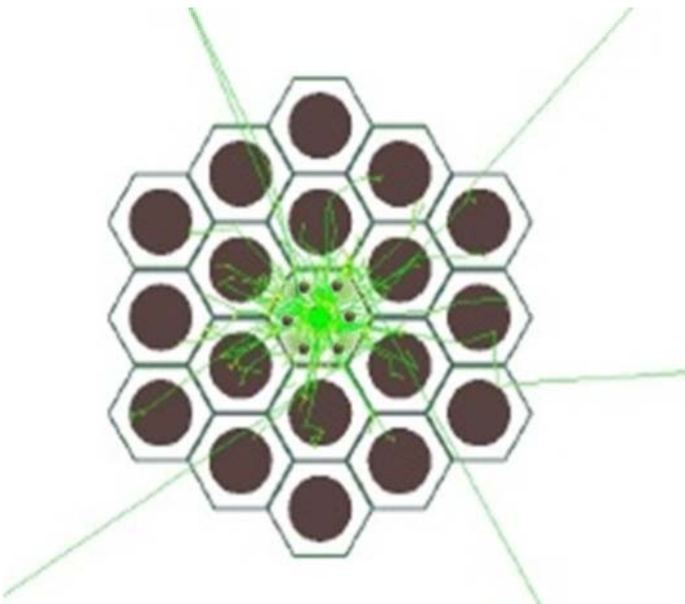
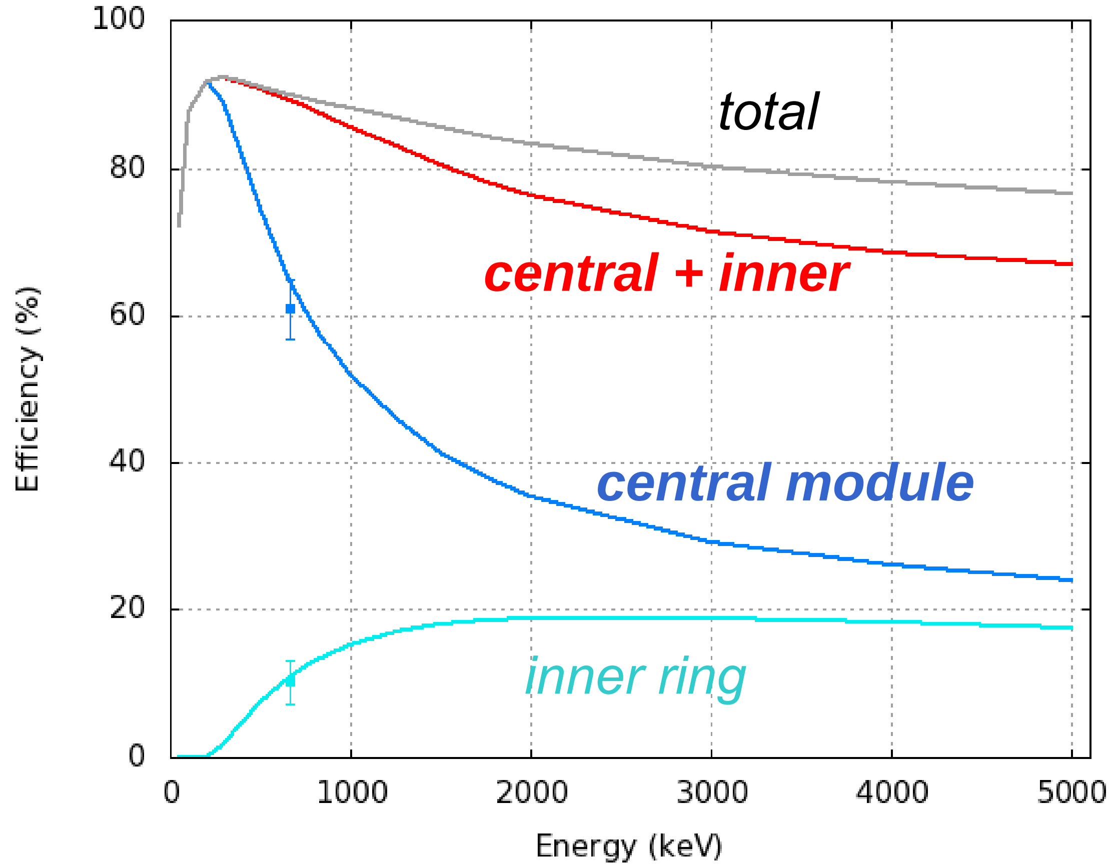
"LIGHT" SHIELDING TESTS

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



First tests of MTAS full energy γ-efficiency with calibrated ^{137}Cs source





"simulations team"

A. Kuzniak, B.C. Rasco, M. Karny

So far, so good… !

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SHIELDING (cont.)

Movable 3-sector support frame
for an additional 1" lead layer
Design: J. Johnson (6', ORNL)
partial use of ORNL-owned lead

delivery of full shielding structure
is expected during this week

Final shielding will have
total lead thickness ~ 1.75

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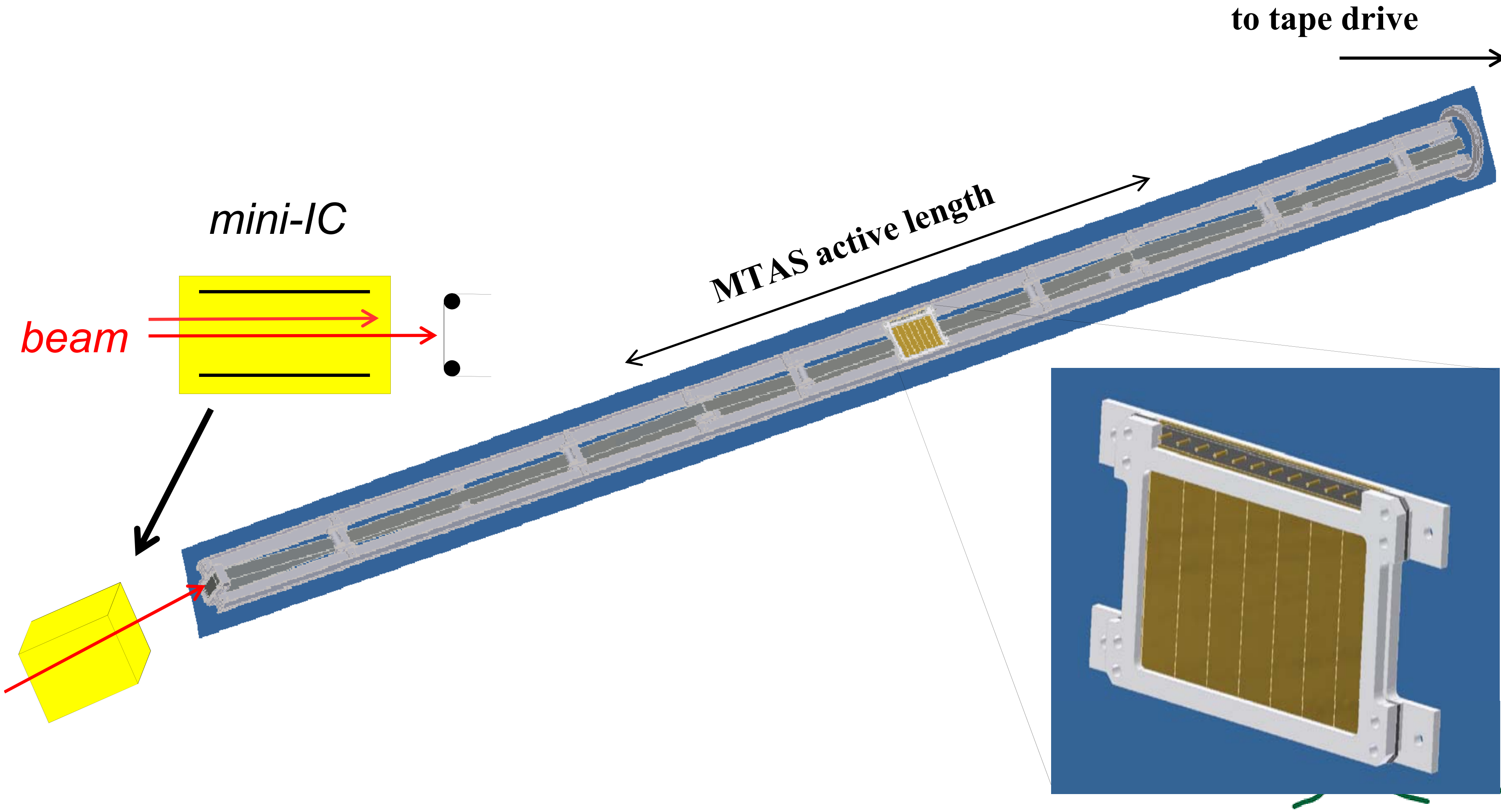
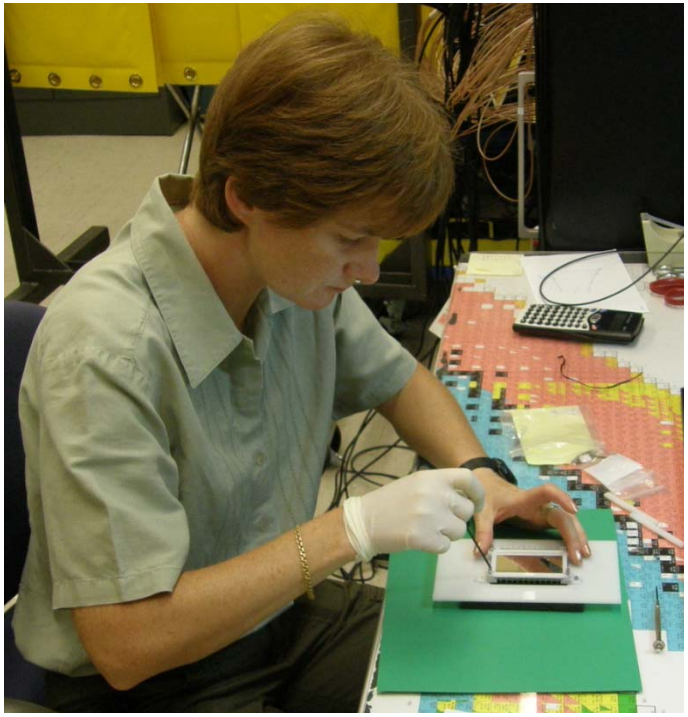
AUXILLIARY DETECTORS and MOVING TAPE DRIVE

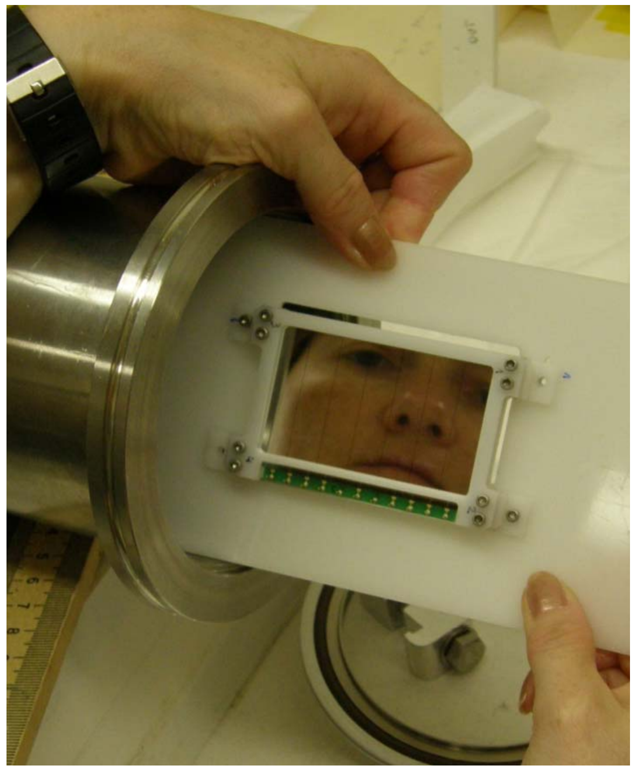
mini Ionization Chamber (mini-IC) for identifying and counting radioactive ions
7-strip, 1 mm thick Si SSDs – detectors surrounding the tape-collected sample
tape drive (Ed Zganjar, LSU)



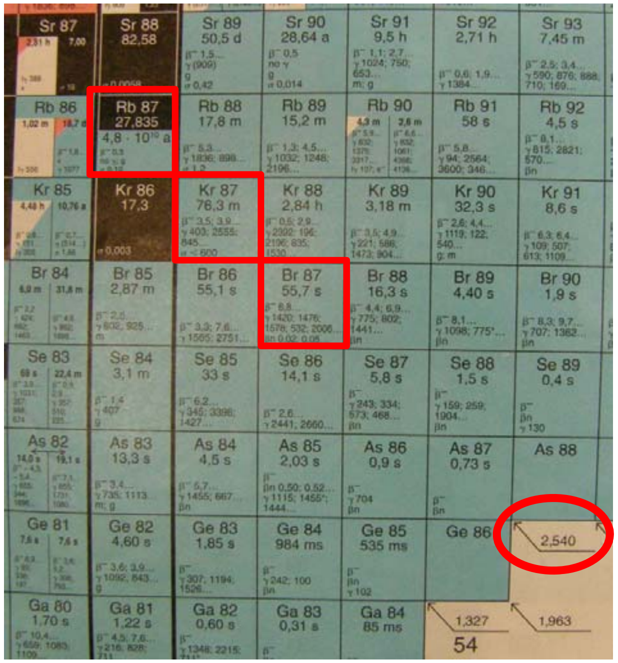
MICRON I-1000 Si-detectors (7- strip, 60 mm by 40 mm active area)
MESYTEC preamps + 100 MHz Pixie16 (re-used)

FWHM (1 MeV electrons) ***~ 25 keV,*** *energy threshold* ***< 50 keV***

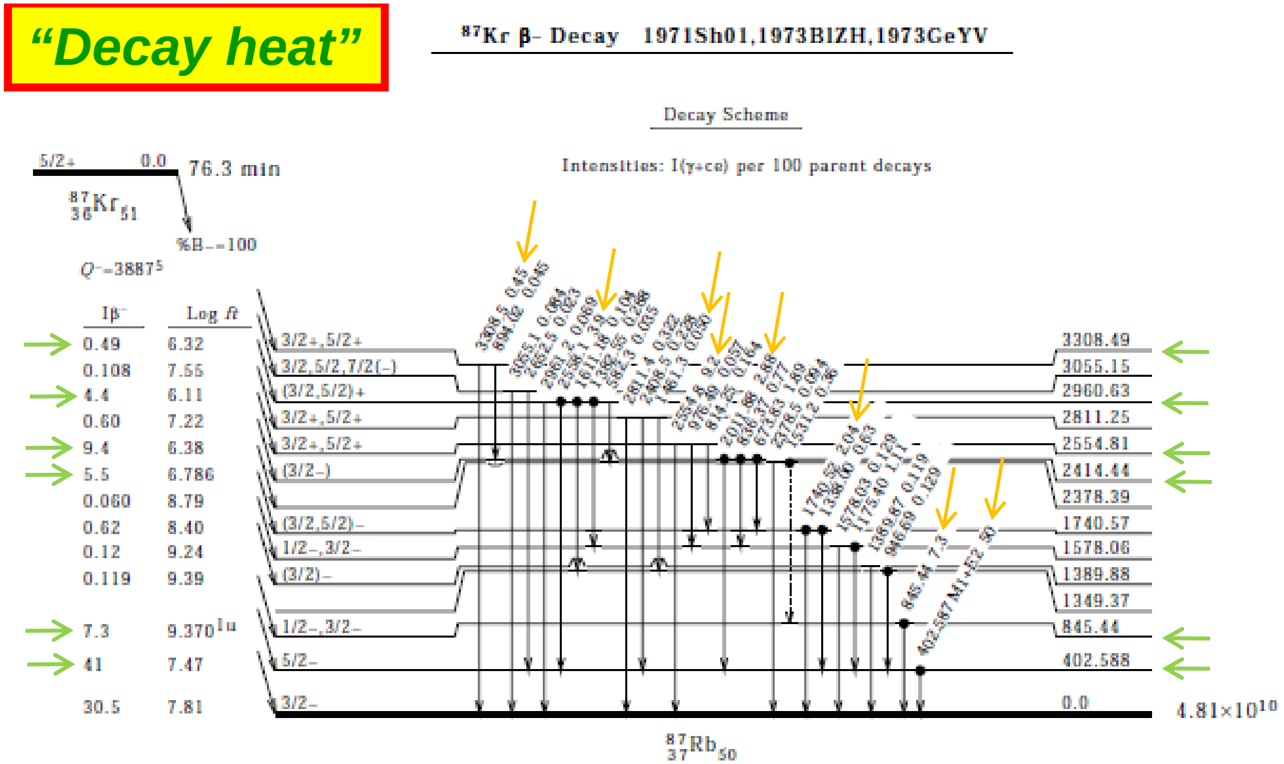




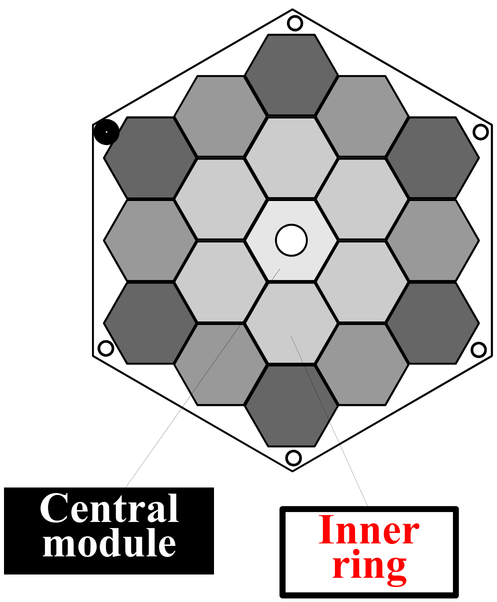
"Decay heat"



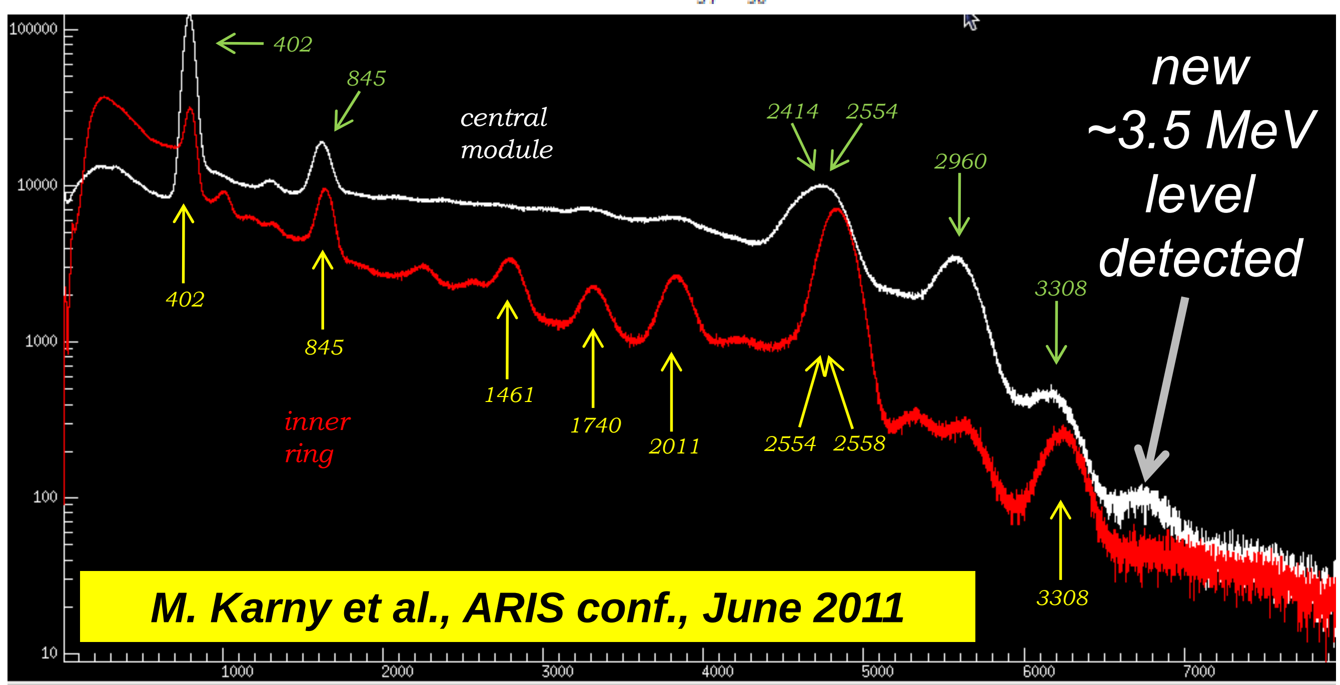
Sr 87
Sr 88
82,58
Sr 89
50,5 d
Sr 90
28,64 a
Sr 91
9,5 h
Sr 92
2,71 h
Sr 93
7,45 m
Rb 86
Rb 87
27,835
Rb 88
17,8 m
Rb 89
15,2 m
Rb 90
Rb 91
58 s
Rb 92
4,5 s
Kr 85
Kr 86
17,3
Kr 87
76,3 m
Kr 88
2,84 h
Kr 89
3,18 m
Kr 90
32,3 s
Kr 91
8,6 s
Br 84
Br 85
2,87 m
Br 86
55,1 s
Br 87
55,7 s
Br 88
16,3 s
Br 89
4,40 s
Br 90
1,9 s
Se 83
Se 84
3,1 m
Se 85
33 s
Se 86
14,1 s
Se 87
5,8 s
Se 88
1,5 s
Se 89
0,4 s
As 82
As 83
13,3 s
As 84
4,5 s
As 85
2,03 s
As 86
0,9 s
As 87
0,73 s
As 88
Ge 81
Ge 82
4,60 s
Ge 83
1,85 s
Ge 84
984 ms
Ge 85
535 ms
Ge 86
2,540
Ga 80
1,70 s
Ga 81
1,22 s
Ga 82
0,60 s
Ga 83
0,31 s
Ga 84
85 ms
1,327
1,963
54



87Kr β- Decay 1971Sh01,1973BlZH,1973GeYV
Decay Scheme
Intensities: I(γ+ce) per 100 parent decays
5/2+ 0.0 76.3 min
87 36 Kr 51
%B-=100
Q-=3887 5
Iβ-
Log ft
0.49 6.32 3/2+,5/2+ 3308.49
0.108 7.55 3/2,5/2,7/2(-) 3055.15
4.4 6.11 (3/2,5/2)+ 2960.63
0.60 7.22 3/2+,5/2+ 2811.25
9.4 6.38 3/2+,5/2+ 2554.81
5.5 6.786 (3/2-) 2414.44
0.060 8.79 2378.39
0.62 8.40 (3/2,5/2)- 1740.57
0.12 9.24 1/2-,3/2- 1578.06
0.119 9.39 (3/2)- 1389.88
1349.37
7.3 9.370 1u 1/2-,3/2- 845.44
41 7.47 5/2- 402.588
30.5 7.81 3/2- 0.0 4.81×10^10 y
87 37 Rb 50



Central module
Inner ring



100000
10000
1000
100
10
1000
2000
3000
4000
5000
6000
7000
402
845
central module
2414
2554
2960
3308
new ~3.5 MeV level detected
402
845
1461
1740
2011
2554
2558
inner ring
3308
M. Karny et al., ARIS conf., June 2011

We should remember that even the very best simulations of nuclear fuel cycles require correct experimental input data.

"Conquering nuclear pandemonium",
K. Rykaczewski, Physics, 3, 94, 2010
(credit to A. Algora et al., PRL 105, 202501, 2010)

SUMMARY

Modular Total Absorption Spectrometer MTAS was constructed, characterized and already applied to the initial decay studies of fission products *at ORNL.*

The final deliveries of array components (part of Pb-shielding, customized tape drive) are expected in August/September 2011.

The initial studies important for the ***verification and development of nuclear structure models***
and in particular applied measurements of
decay heat released by radioactive nuclei produced in nuclear fuels at power reactors
will be performed in 2012.

Budget status *(see quarterly reports):*
Total of ***$ 1580 K*** *(1^{st} Oct 2009 – 30^{th} Sept 2012)*
Costs and commitments : ***$ 1430 K*** *(with* ***$ 700 K hardware*** *)*
include post-docs and students in 2012