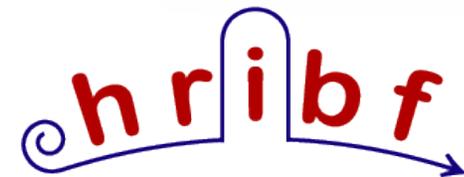


***Decay Spectroscopy of Fission Products
for Nuclear Fuel Cycle
at the HRIBF***

Krzysztof P. Rykaczewski

*Physics Division, Oak Ridge National Laboratory
Oak Ridge, Tennessee*





<http://www.ornl.gov/sci/casl/>

May 2010 : the Department of Energy creates the first nuclear energy innovation hub -- the **Consortium for Advanced Simulation of Light Water Reactors (CASL)** -- headquartered at Oak Ridge.

The first task will be to develop **computer models that simulate nuclear power plant operations, forming a "virtual reactor" for the predictive simulations of light water reactors.** Other tasks include using **computer models** to reduce capital and operating costs per unit of energy, safely extending the lifetime of existing U.S. reactor and reducing nuclear waste volume generated by enabling higher fuel burn-ups.

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

**“Conquering nuclear pandemonium”
KR’s Viewpoint in Physics, 3, 94, 2010**

h r i b f = **Holifield Radioactive Ion Beam Facility
at Oak Ridge**

*proton-induced fission of ^{238}U
creates neutron-rich nuclei
for spectroscopic studies*

*beta-delayed neutron emission
and
“decay heat” measurements*

Z



N



^{78}Ni

ISOL RIBs

^{132}Sn

*$\beta\gamma$ and $\beta n\gamma$
decays*

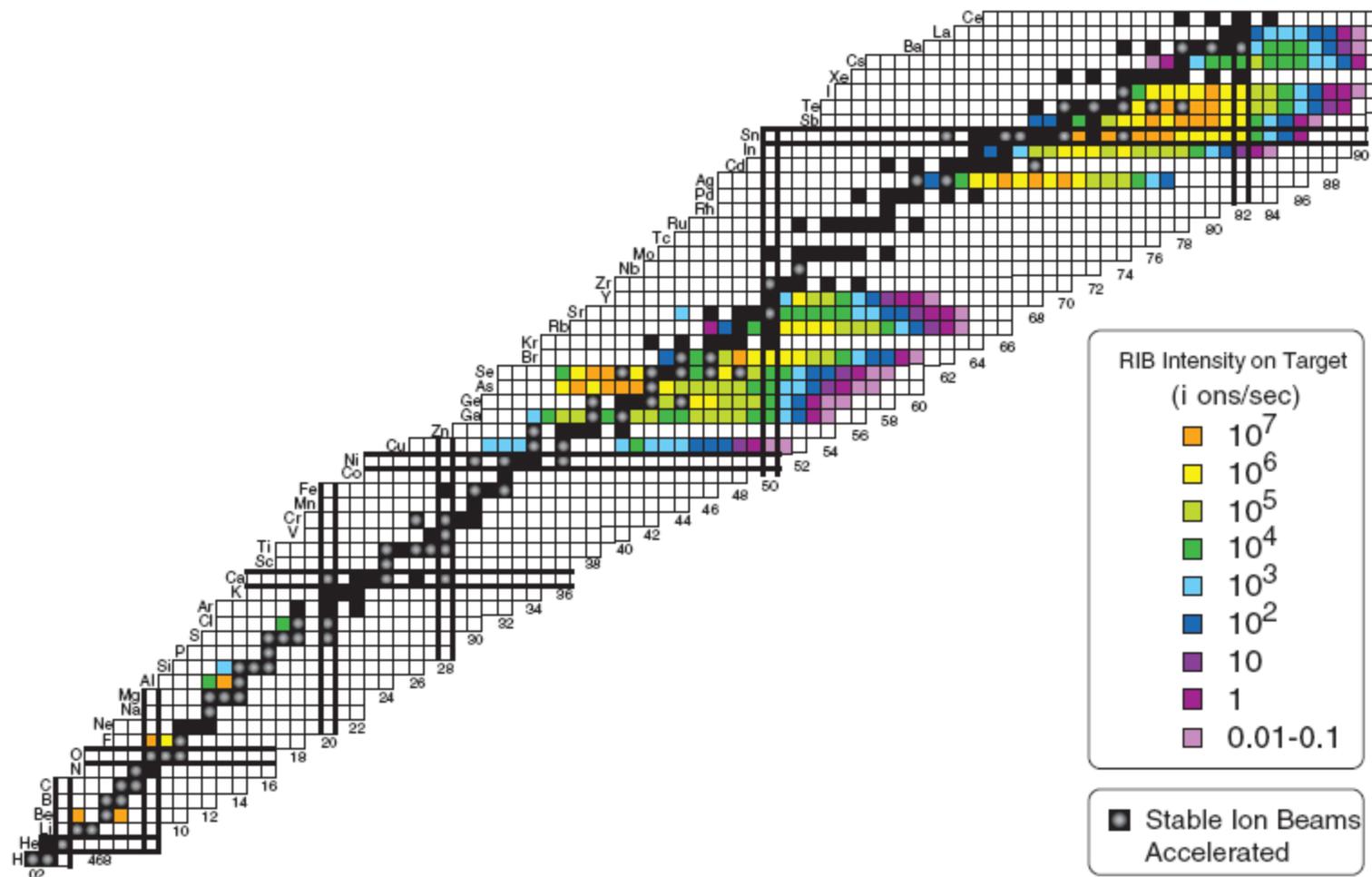


Figure 3. Radioactive and stable beams presently accelerated at the HRIBF. Additional radioactive species such as Zn, Kr, Cd, and Xe are available up to 200 keV as positive ions at LeRIBSS. Stable beams from our new SNICS source [11] are used for RIB development. They are also used to simulate RIBs for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



***among motivations of the HRIBF
decay studies of fission products :***

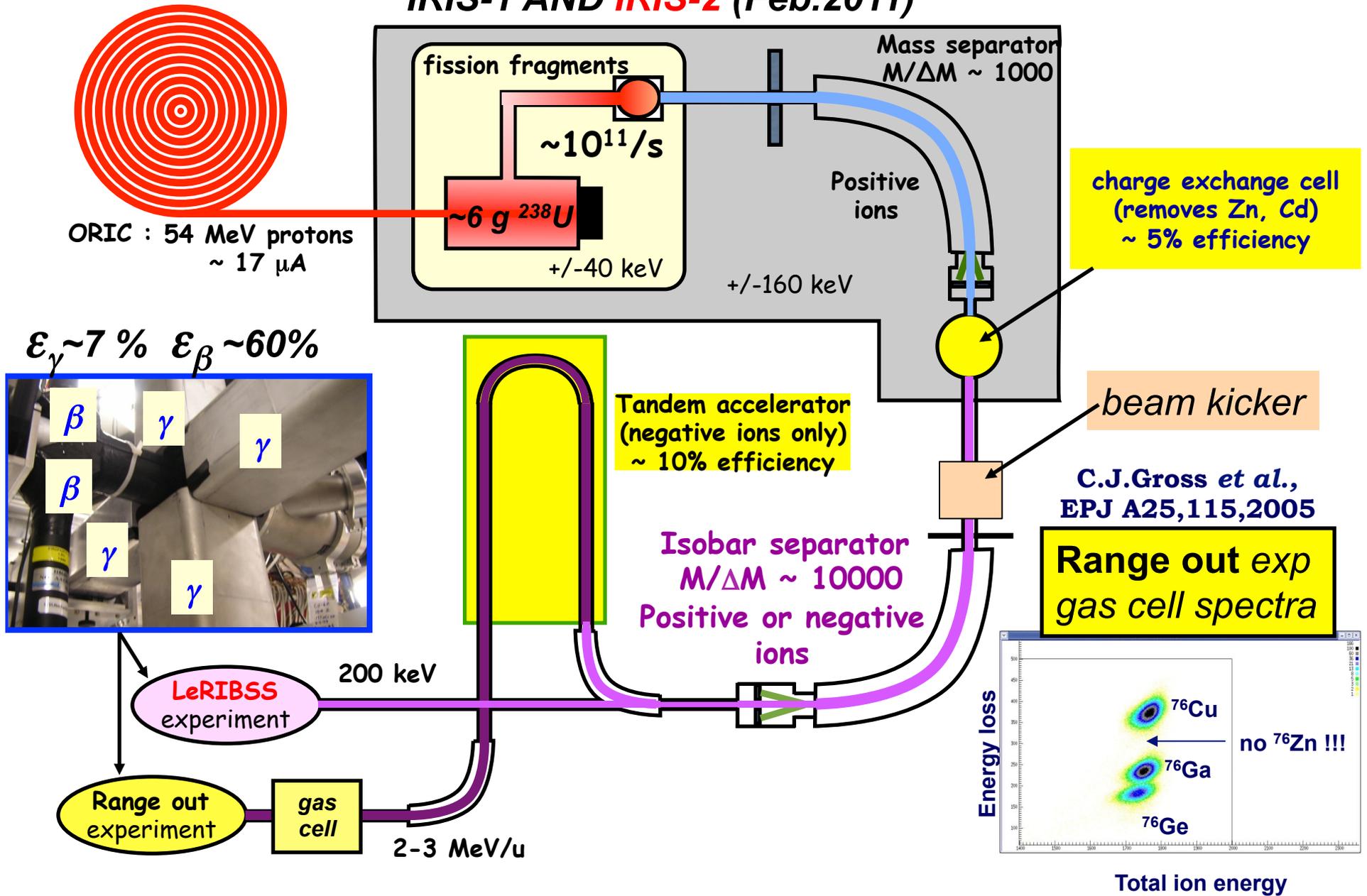
- ***understanding the evolution of nuclear structure***
- ***beta-decay properties are needed for the analysis of post r-process isotopic distributions (half-lives, beta-delayed neutron rates, low- energy isomers ...)***

***the decay properties of fission products
are among the parameters needed for :***

***the operation of nuclear reactors,
reactor shut-down process,
the nuclear spent fuel/nuclear waste handling***

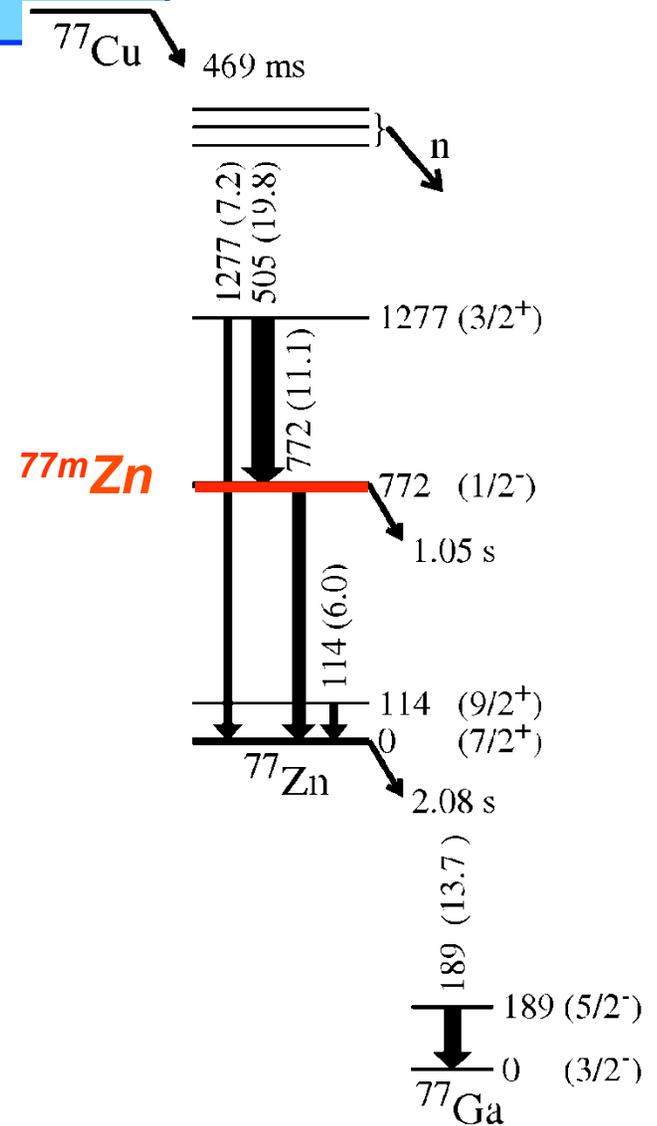
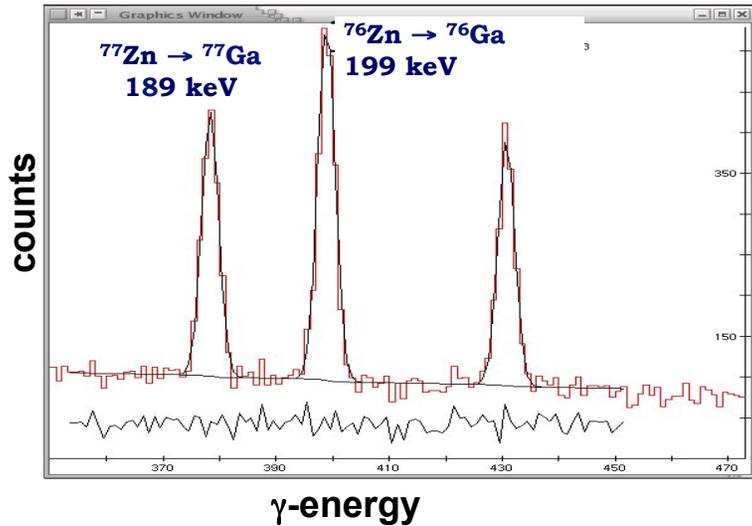
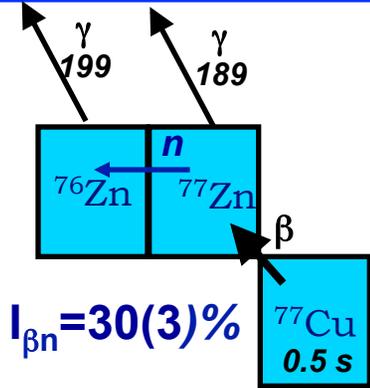
Decay studies of fission products at the **n r i b f**

IRIS-1 AND IRIS-2 (Feb.2011)



hrif

^{77}Cu : 18 hours of "ranging-out" exp with 15 pps
Winger et al., Phys. Rev. Lett., 102, 142502, 2009
Ilyushkin et al., Phys. Rev. C 80, 054304 (2009)



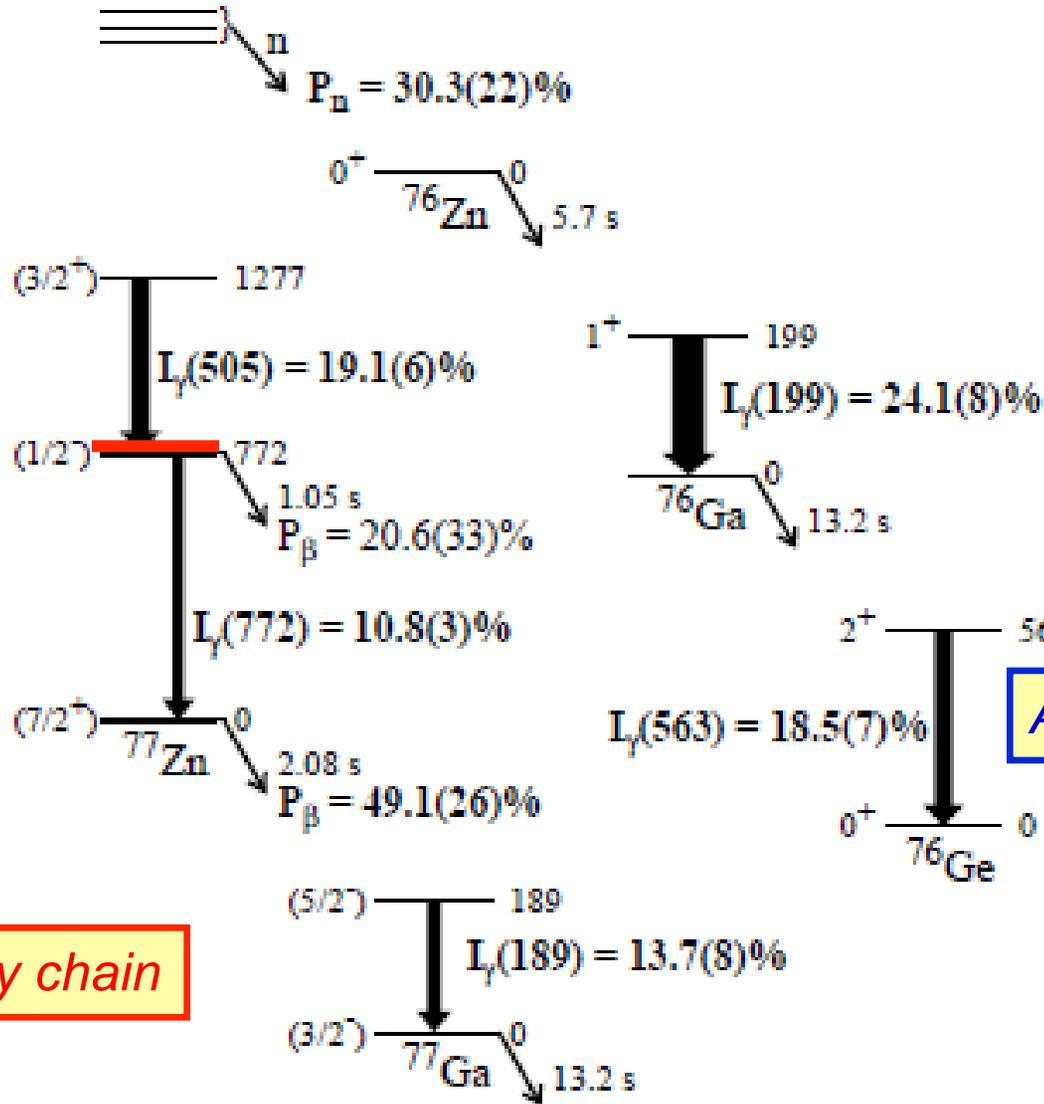
**no Zn in separated beam,
pure beam of Cu ions identified and counted !**

OAK RIDGE NATIONAL LABORATORY
U.S. DEPARTMENT OF ENERGY

watch out :
 $^{77}\text{Cu} \rightarrow ^{77m}\text{Zn} \rightarrow ^{77gs}\text{Ga}$

S. Ilyushkin et al., Phys. Rev. C 80, 054304, 2009
we can solve very complex decay schemes !

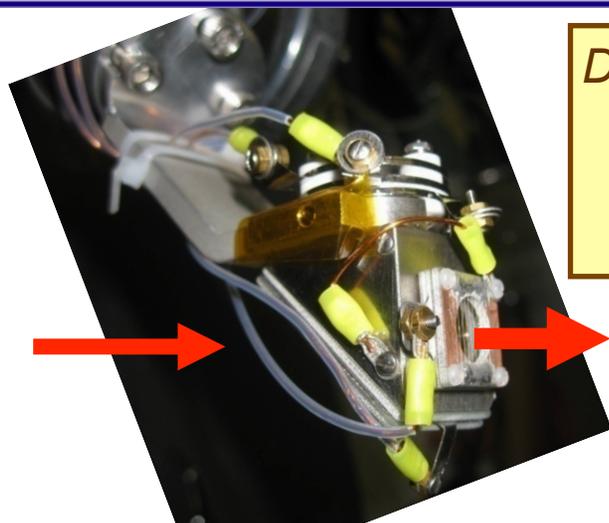
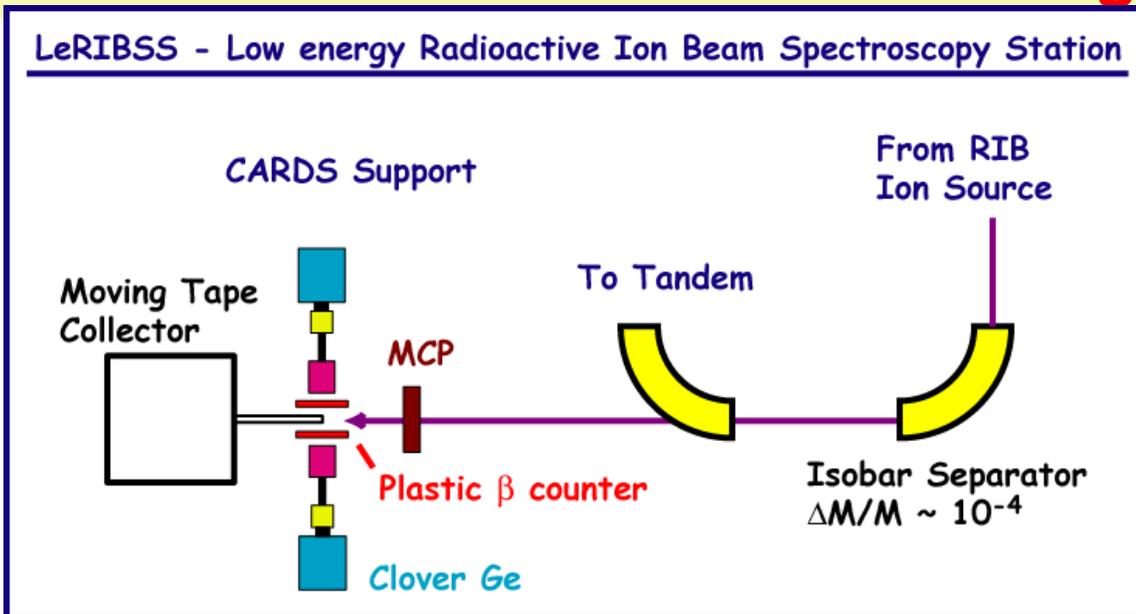
$\left(\frac{5}{2}^- \right)$
 ^{77}Cu ← pure, identified and counted ^{77}Cu ions
 480(9) ms



A=76 decay chain

A=77 decay chain

Decay studies : HRIBF as a powerful ISOL facility
very high rates + isobaric separation + ion “ranging-out” and tagging
+ efficient detectors and digital data acquisition
and the collaborations of contributing users !



*Dan Shapira's MCP at LeRIBSS counting and tagging ions
 5 μg/cm² Carbon-foil,
 ~87% transmission*

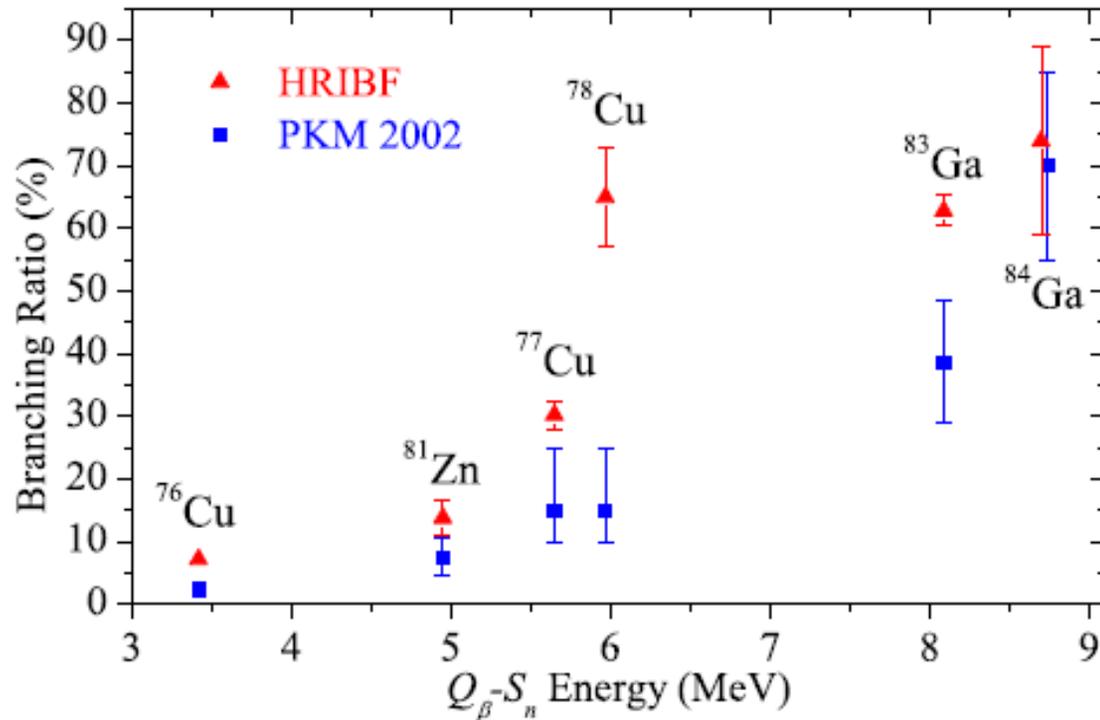
*200-250 keV
 isobarically separated
 radioactive ions*



*β, γ, X-ray, CE, neutron
 detector station*

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)**

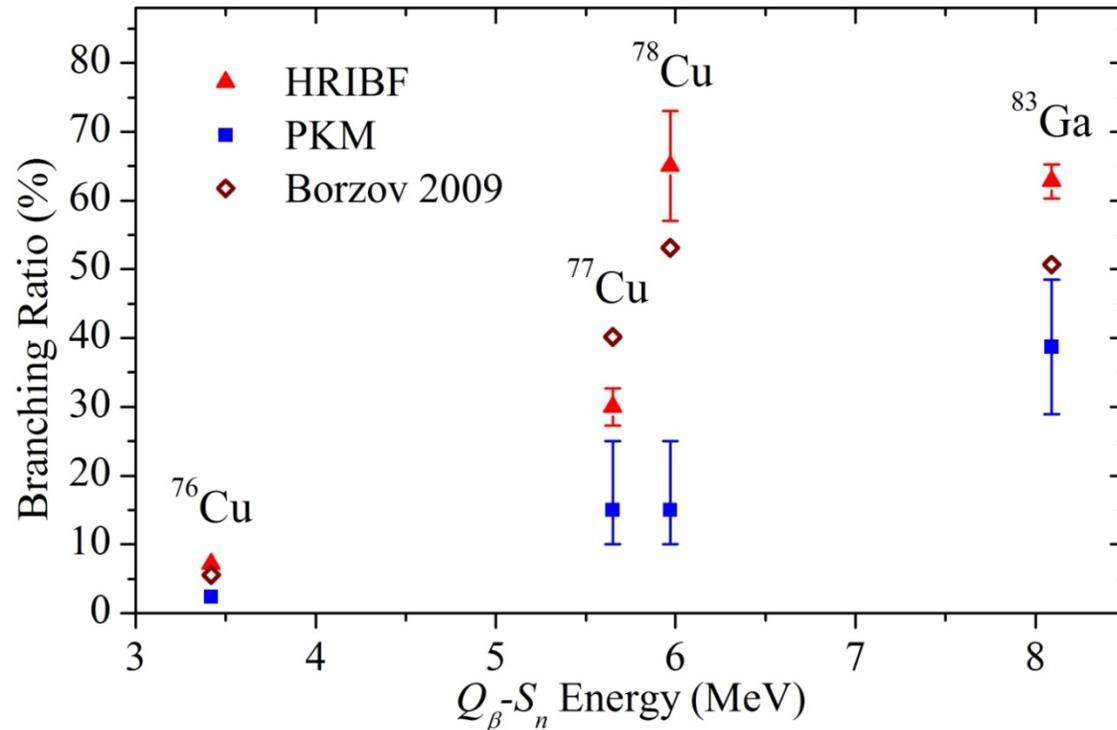


Similar conclusion from a recent NSCL paper basing on NERO results
P. Hosmer, H. Schatz et al., PR C82, 025806, 2010

**starts to be relevant for
nuclear fuel cycle ?!**



J.A. Winger et al.,
PRL, 102, 142502, 2009



New βn -calculations of Borzov closer to the HRIBF “reference values”!

New modeling accounts for :

- new mass measurements (Hakala et al., PRL 101, 052502, 2008)
- an inversion of proton orbitals occurring near ^{78}Ni , from $2p_{3/2}$ to $1f_{5/2}$,

^{75}Cu : Flanagan et al, PRL 103,142501, 2009

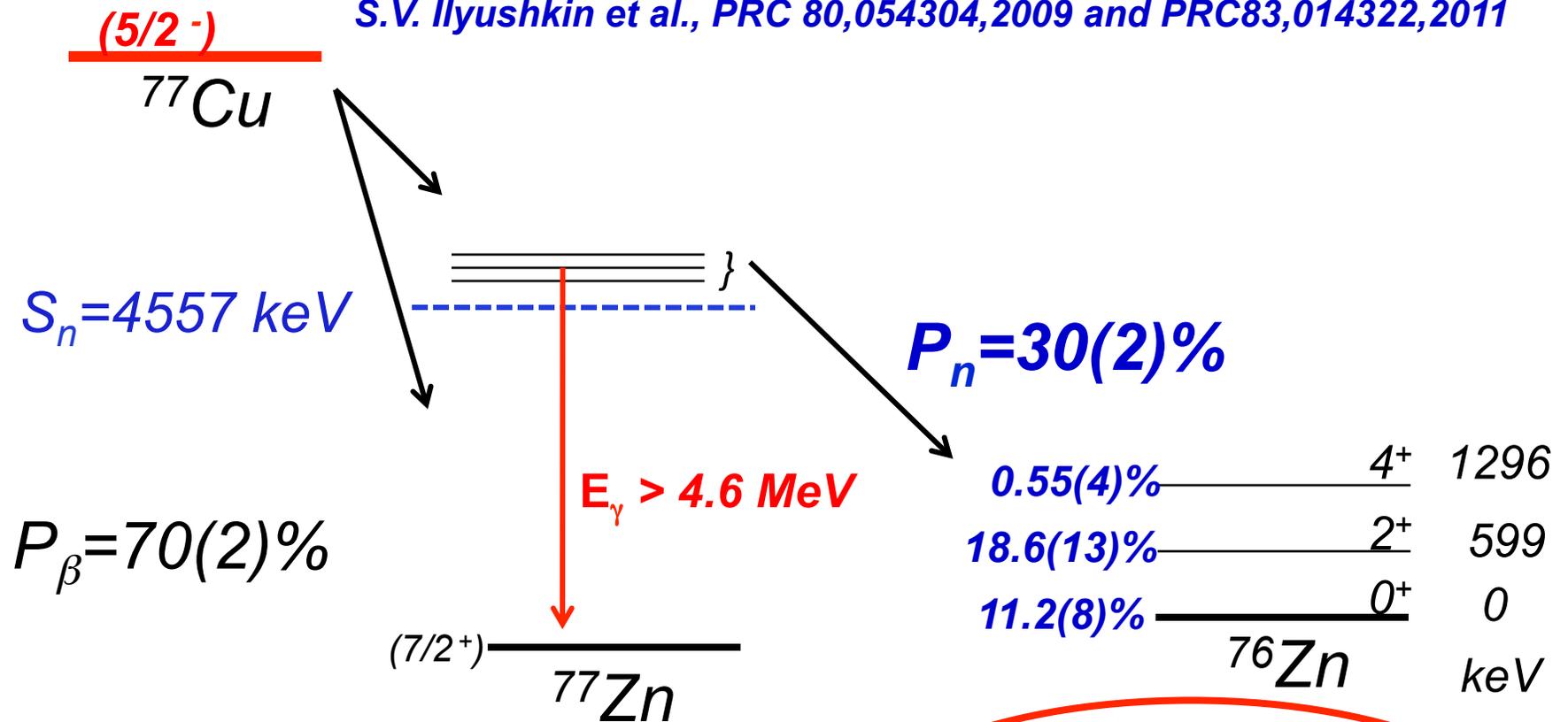
 ^{77}Cu : Ilyushkin et al, PR C83, 014322, 2011

 ^{77}Cu : Ilyushkin et al, PR C80, 054304, 2009

 ^{83}Ga : Winger et al., PR C 81, 044303, 2010

proton orbital inversion plays an important role in Borzov’s modeling!

Fine structure in beta-delayed neutron emission
 S.V. Ilyushkin et al., PRC 80,054304,2009 and PRC83,014322,2011



Could we learn about precursor wave function from βn fine structure ??

from Ian C. Gauld (Reactor Physics Group, ORNL)
the manager for the Oak Ridge Isotope Generation code ORIGEN and associated
nuclear data decay libraries for the past 11 years

- **SCALE Nuclear Analysis Code System**

Standardized Computer Analyses for Licensing Evaluation, ORNL, starting 1976

- **ORIGEN** decay and irradiation analysis code

/Oak Ridge Isotope Generation and Decay Code/

- Nuclear data (e.g., ENDF)

- **Nuclear decay data requirements for nuclear applications**

- **Decay heat for postulated reactor accident/shut down analysis**

- Gamma ray spectroscopy for nuclear safeguards

- **Advanced reactor design and fuel cycle safety**

(slow and fast neutrons !)

- National security

from Ian C. Gauld (2010)

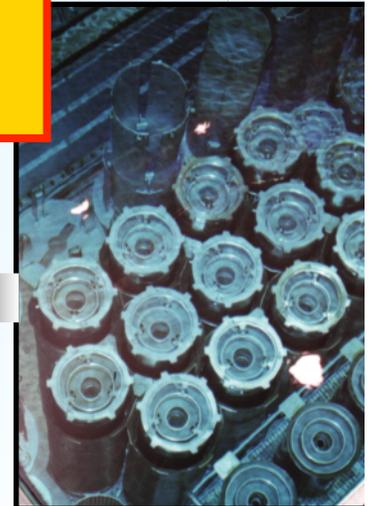
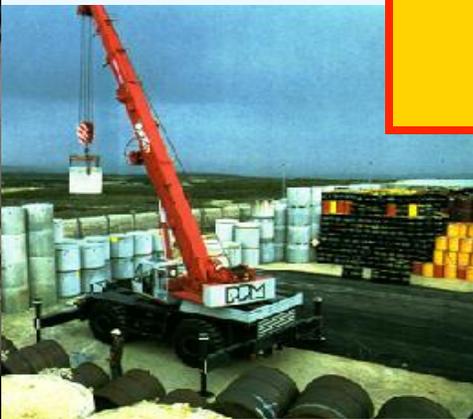
Material processing and fabrication



Commercial and research reactors



SCALE is modular code system used throughout the world for reactor and fuel cycle applications



Reprocessing

Disposal

Transport

Storage

Modeling of beta-delayed neutron emission for applications purposes (“six group formula”)

see, e.g., Dore et al., 4th Int. Workshop on Nuclear Fission and Fission-Product Spectroscopy, Cadarache, France, 13-16 May 2009

“ ..beta-delayed neutron precursors are generally lumped into six groups according to their half-lives [Keepin 1965] . The time dependence of β -delayed neutron yield

$Y_{\beta n}$ is then expressed by

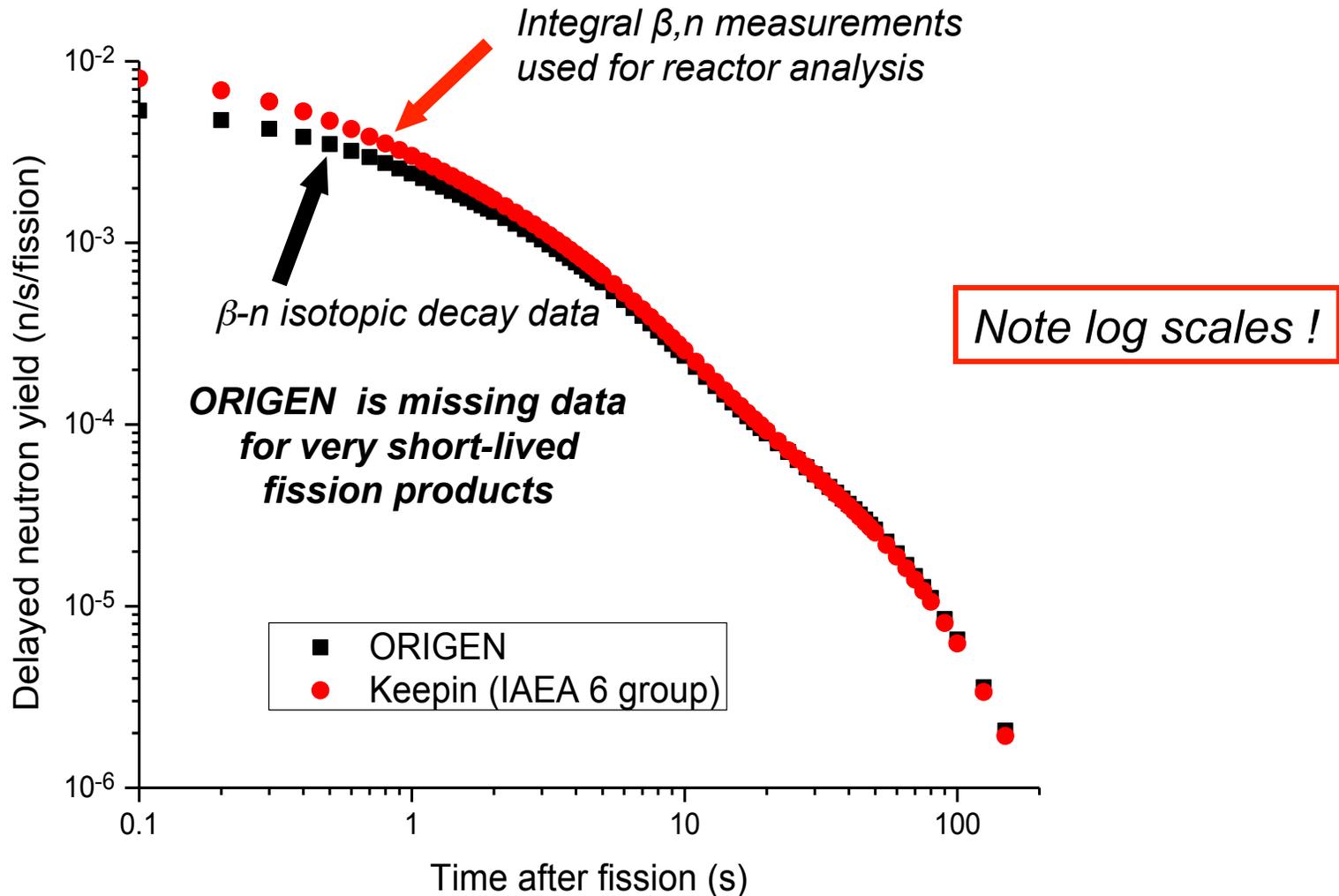
$$Y_{\beta n}(t) = \sum_{i=1}^6 a_i \exp(-\lambda_i t),$$

$i= 1$ to 6 , a_i, λ_i are parameters fitted to “integral observations”

I. C.Gauld, March 2010 :

- **Current fission product β -n decay data do not accurately match integral experiments and delayed neutron parameters can not be calculated from basic decay data**
- **As the U.S. studies new reactor designs and advance fuel cycle options, existing integral β -n data may not be applicable**
 - **Deep burn actinide transmutation studies, etc.**
 - **Improved decay data will enable analyses beyond range of current designs using fundamental physics data**

Delayed Neutron Yield following ^{235}U fission



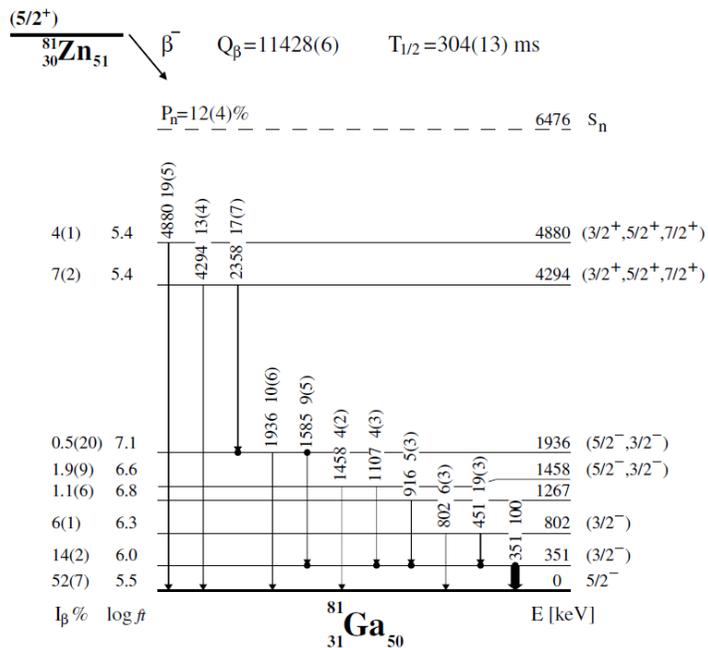
from Ian C. Gauld (2010)

LeRIBSS Decay Spectroscopy of Fission Products: $^{82,83}\text{Zn}$, $^{85,86}\text{Ga}$

February 2011

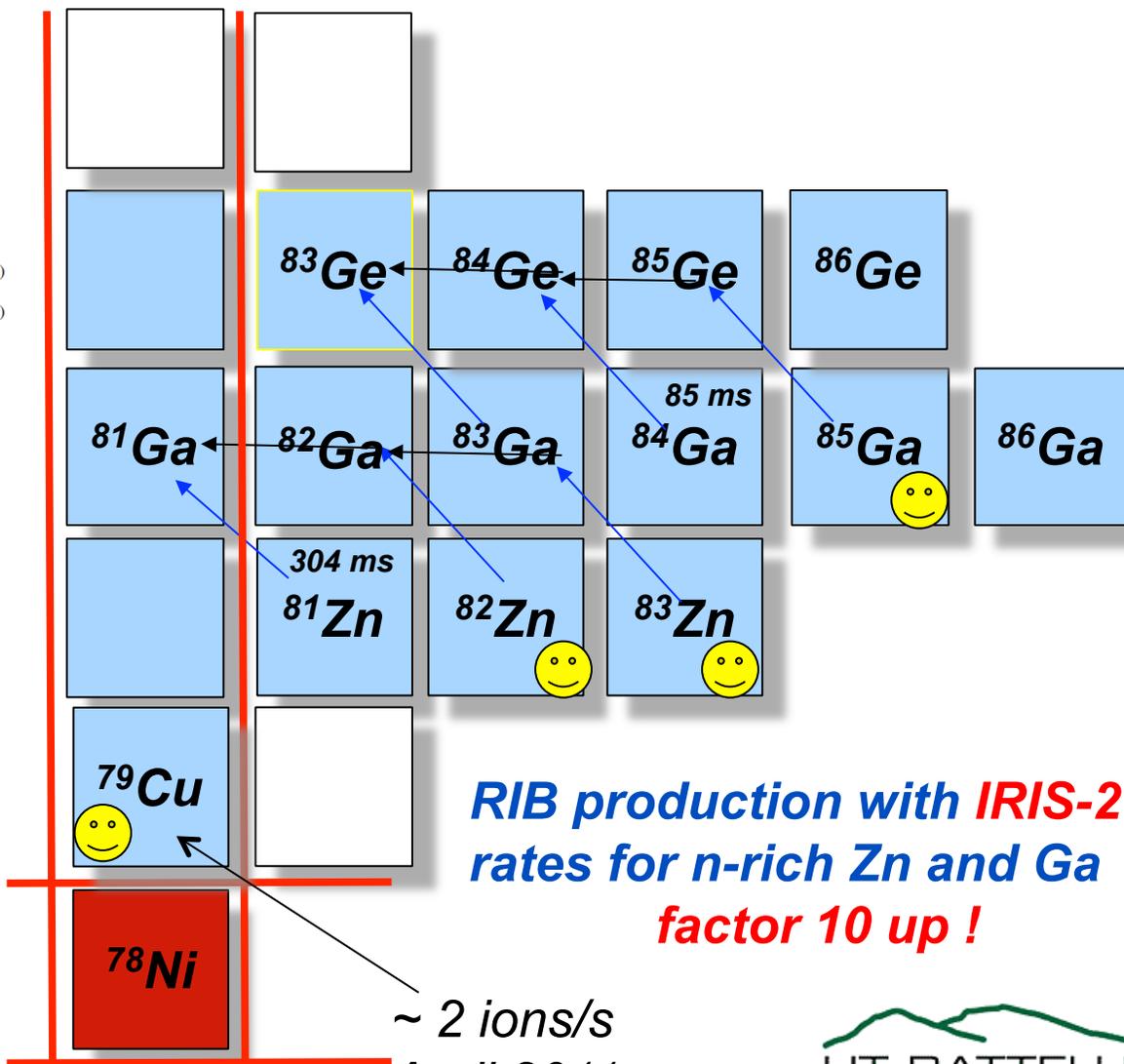
→ talk by Miguel Madurga (UTK)

Summer 2008



^{81}Zn decay

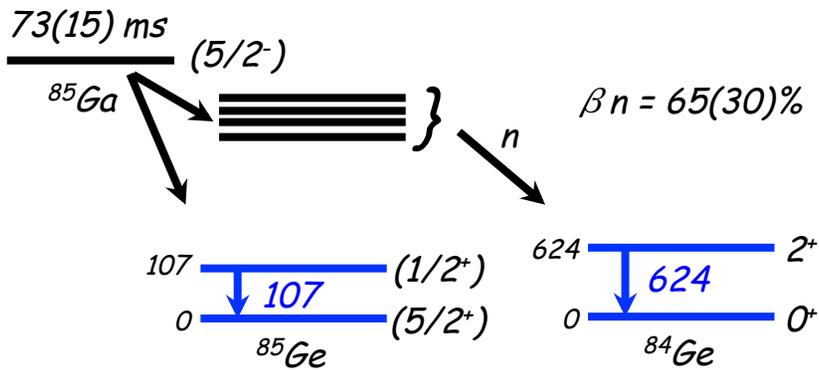
S. Padgett (UTK) et al., PR C 82, 064314, 2010
 exp with 30 pps of ^{81}Zn beam
 10^6 pps of ^{81}Ga → ~ 0 pps
 here $M : \Delta M \sim 6400$



RIB production with IRIS-2 :
 rates for n-rich Zn and Ga
 factor 10 up !

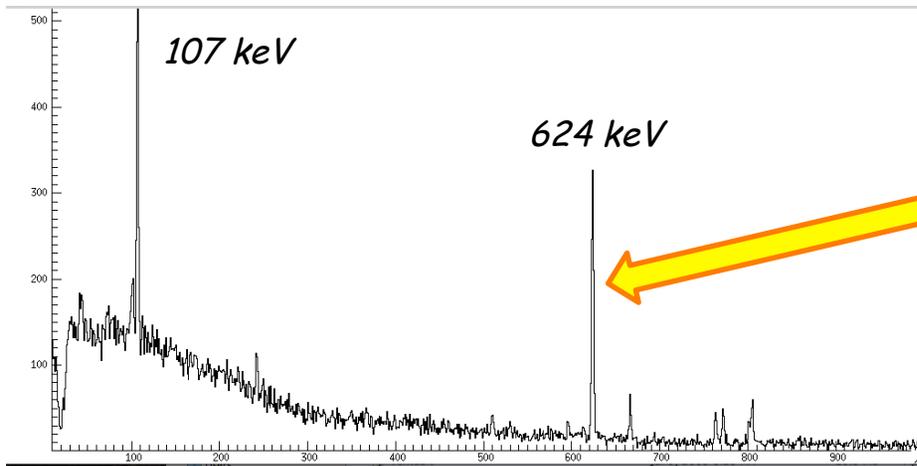
~ 2 ions/s
 April 2011

^{85}Ga February 2011 (online data)

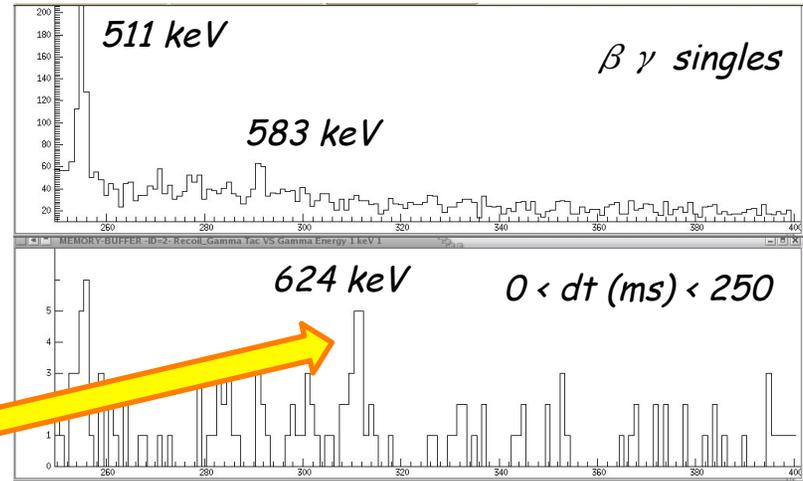


$^{85}\text{Ga}_{54}$

February 2011

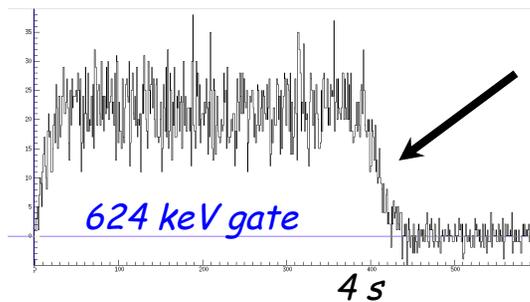
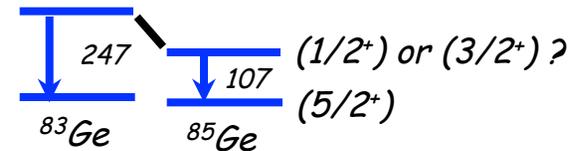


Summer 2006



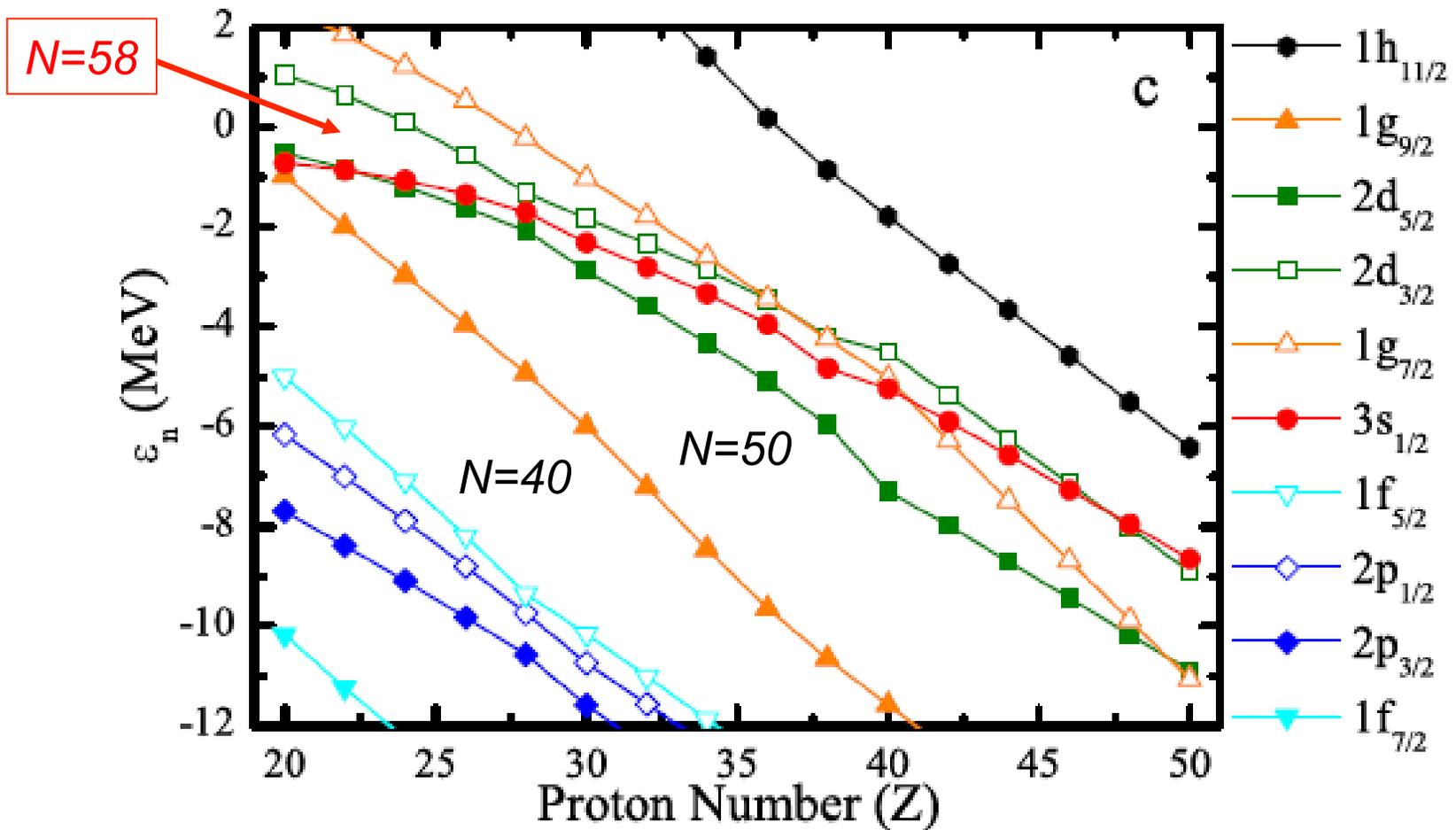
Winger et al., Phys. Rev. C 81, 044303 (2010)

Emerging $N=58$ $d_{5/2}-s_{1/2}$ subshell?



Beam on: 4 s
Beam off: 2 s
Tape move: 0.45 s

“New shell closure at $N=58$ emerging in neutron-rich nuclei beyond ^{78}Ni ”
J.A. Winger et al., Phys. Rev. C81, 044303 (2010).

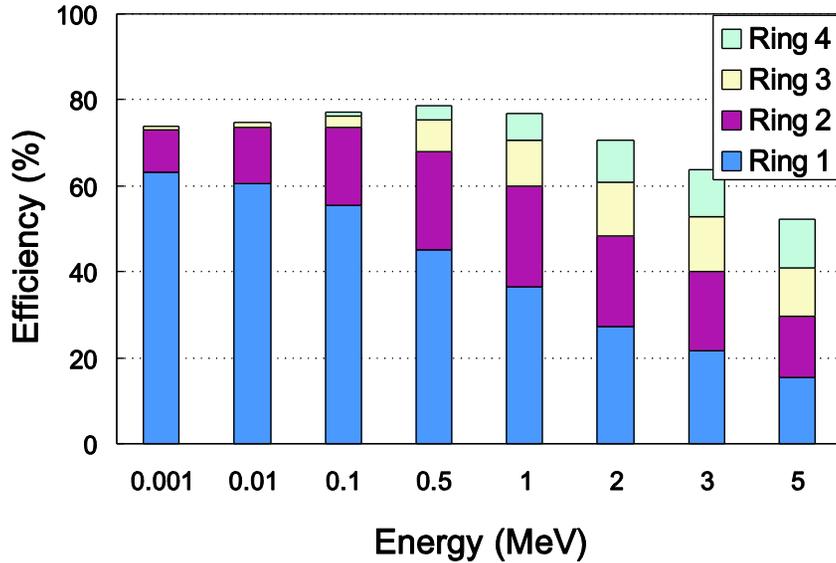


calculations for N=50 isotones : J.Dobaczewski

nearly 80% efficient and segmented neutron counter *3Hen* enhancing decay spectroscopy capabilities at the HRIBF

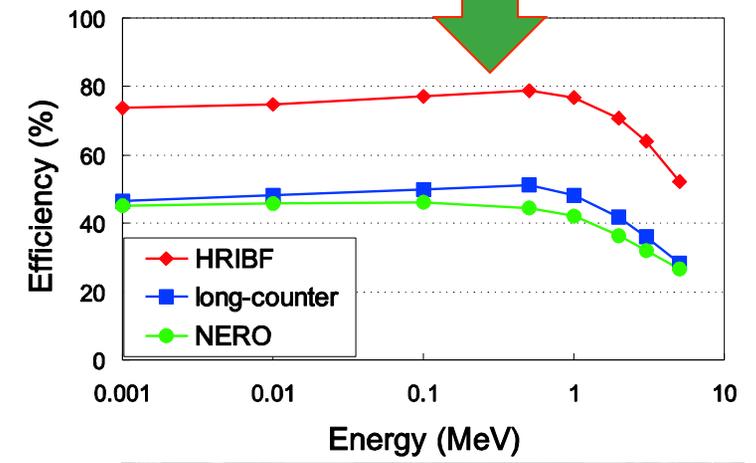


Neutron Efficiency by Ring



*GEANT4 simulations:
Sean Liddick*

HRIBF, Long-counter, and NERO Neutron Efficiency



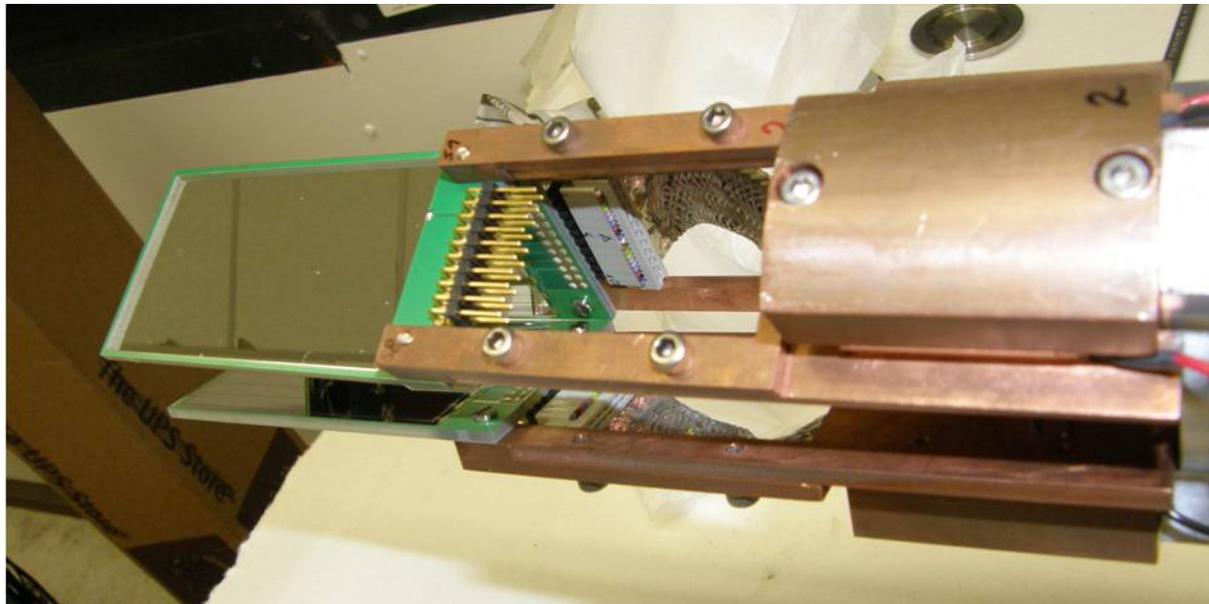
**ORNL, UTK
LSU, Mississippi,
NSCL, UNIRIB**

**discovery experiments
(HRIBF, NSCL, FRIB)
and
applied studies
(HRIBF)**



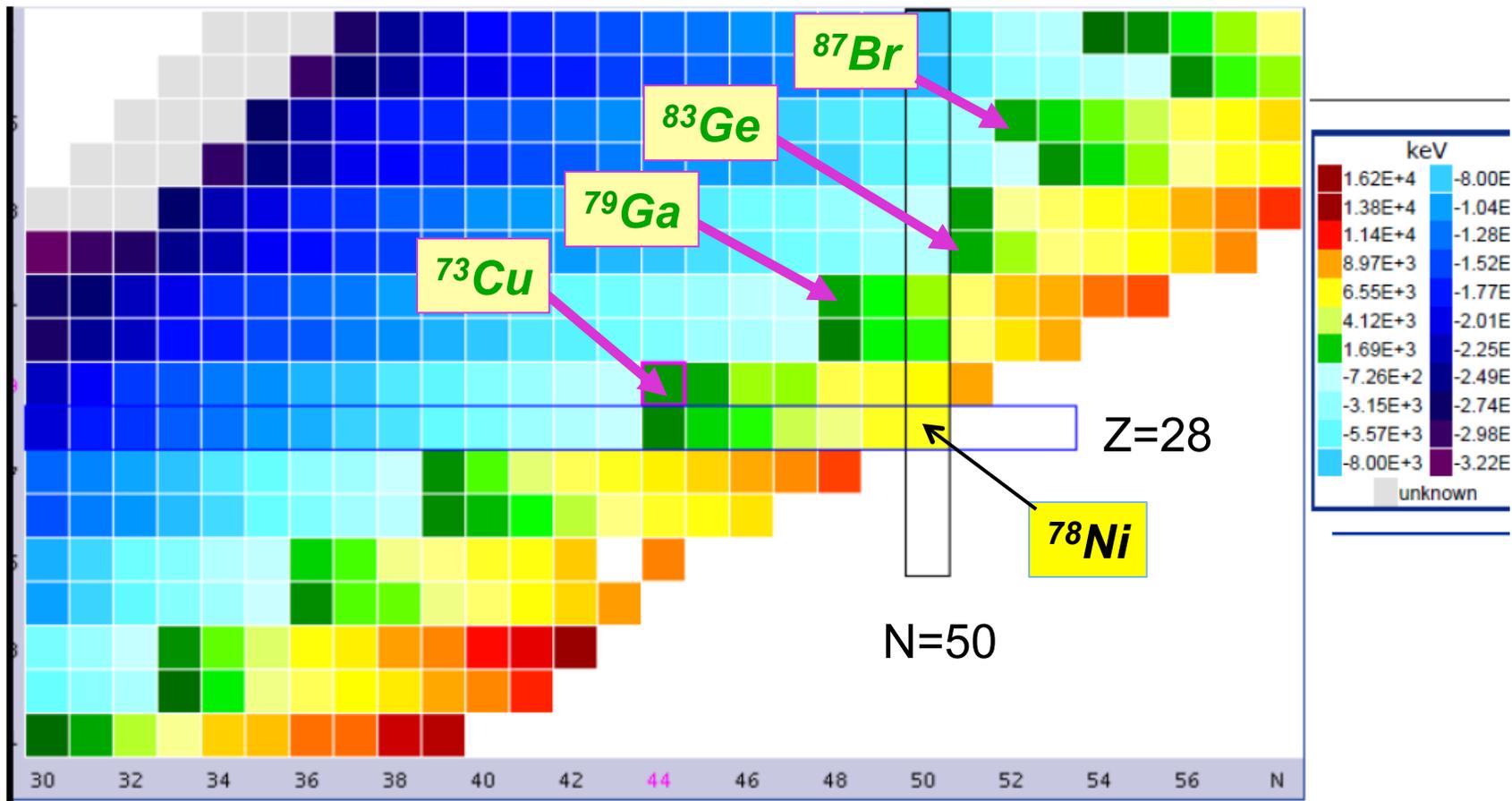
Si-triangle inside 3Hen – beta energy-loss trigger

- three 1-mm thick 8-strips SSDs, ~ 80 mm long
- ~ 20 keV fwhm within 400 -1000 keV electron energy range
- ~ 35 keV energy threshold, ~ 70 % efficiency



beta detectors surrounding the activity collection point on movable tape

from NNDC :
 energy window ($Q_{\beta} - S_{\beta n}$) for β -delayed neutron emission precursors

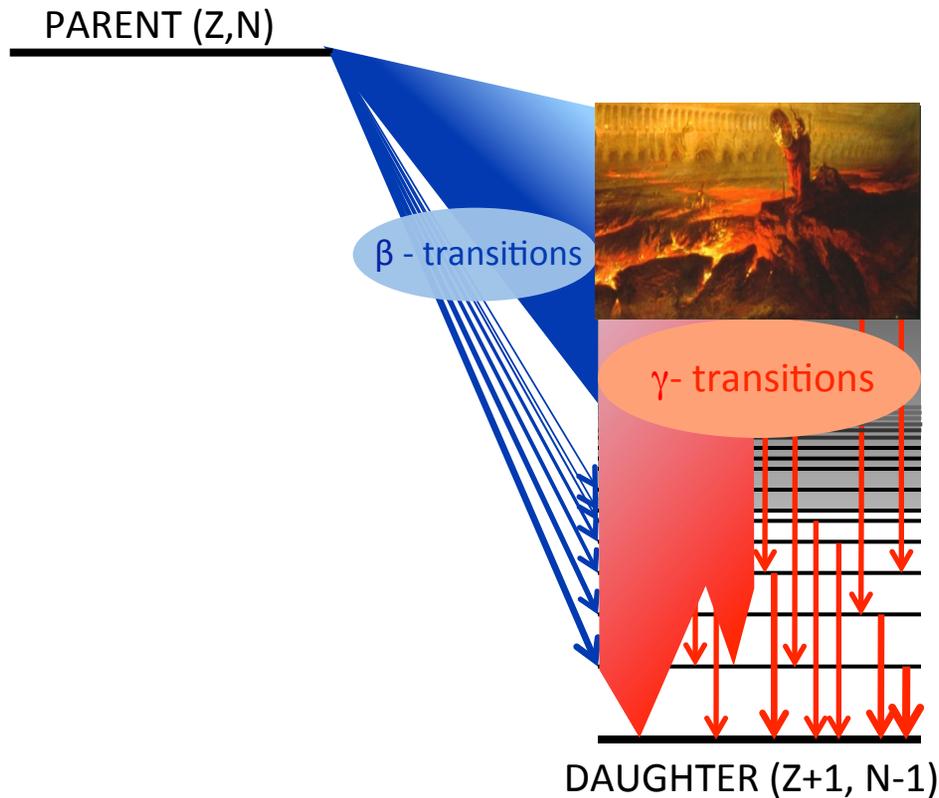


blue color – βn emission not possible

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

red-brown colors – beta-delayed $2n$ emission (low energy?)

Why Total Absorption Gamma Spectroscopy for fission products?



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

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

Credits: J. Milton (1667) and J. Martin (1825)
KR, Physics 3, 94, 2010

ASSESSMENT OF FISSION PRODUCT DECAY DATA FOR DECAY HEAT CALCULATIONS

©OECD 2007
NEA No. 6284

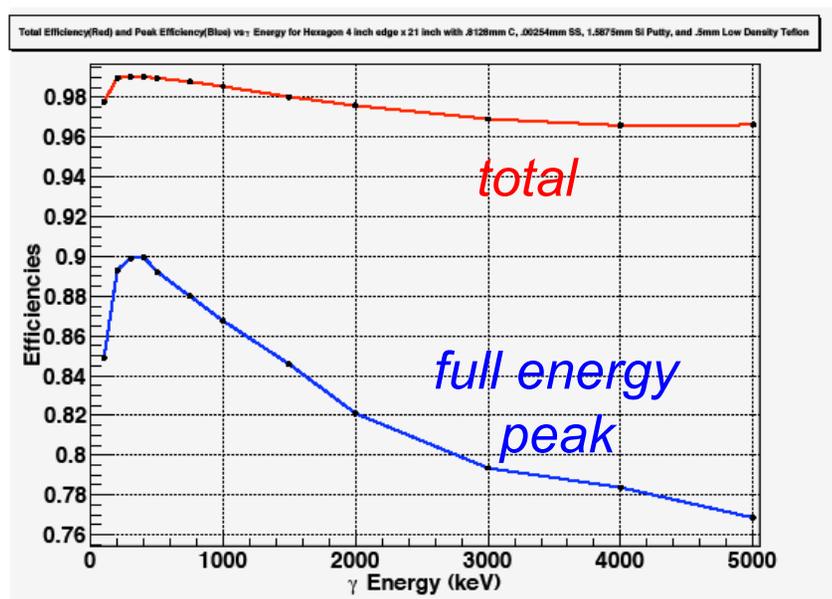
There are obvious needs (β -strength, decay heat) to detect weak, high energy γ -transitions and to establish the true decay pattern for neutron-rich nuclei.

Solution : high-efficiency total absorption spectrometer, see recent example
A. Algora et al., PRL 105, 202501, 2010

New Modular Total Absorption Spectrometer MTAS

(talk by Marek Karny)

A **Modular Total Absorption Spectrometer (MTAS)** has been constructed and will be applied to the **decay studies with pure beams of neutron-rich nuclei produced in the ^{238}U fission at the HRIBF.** The studies important for the verification and development of **the microscopic description of neutron-rich matter** will be performed as well as **applied studies of decay heat released by radioactive nuclei produced in nuclear fuels at power reactors /ARRA funded project/.**



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



Idaho TAS : (1990's \rightarrow ANL) : **MTAS has 17 times larger volume**
LBNL TAS : GSI (1995 – 2003) \rightarrow LBNL : **MTAS is 7 times larger**
Valencia-ISOLDE "Lucrecia" TAS (since \sim 2003) : **MTAS is 8 times larger**
St. Petersburg (Russia) - Jyvaskyla-Valencia TAS

Decay studies for Nuclear Fuel Cycle at HRIBF

- 1. High energy resolution measurements with pure beams
of known intensities (when post accelerated)**

*ranging-out technique plus gamma-beta-conversion electron detectors
basic decay scheme + βn -branching ratio*

- 2. Measurements with Modular Total Absorption Spectrometer **MTAS****

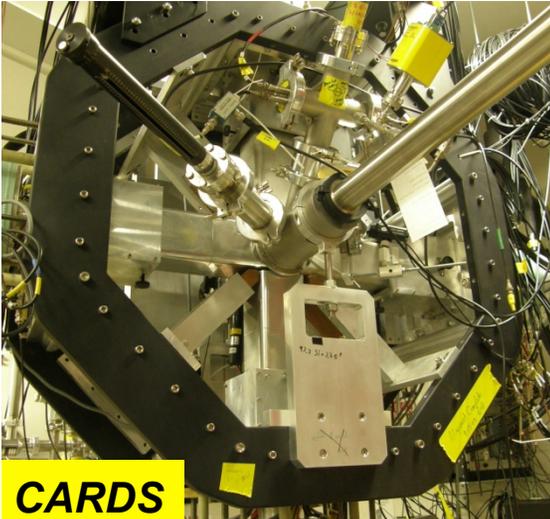
*MTAS gamma spectra → beta strength function within $\beta\gamma$ -window (decay heat)
(MTAS starting with “ranging-out” exps, further studies possibly at **ORISS**)*

- 3. Measurements with **3Hen** and **VANDLE** → β -delayed neutrons**

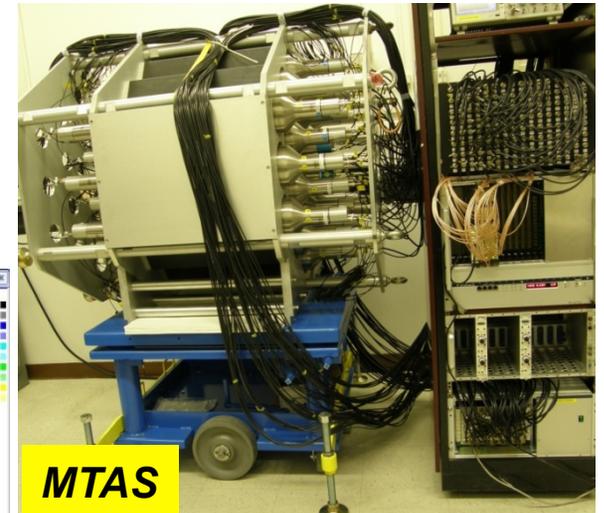
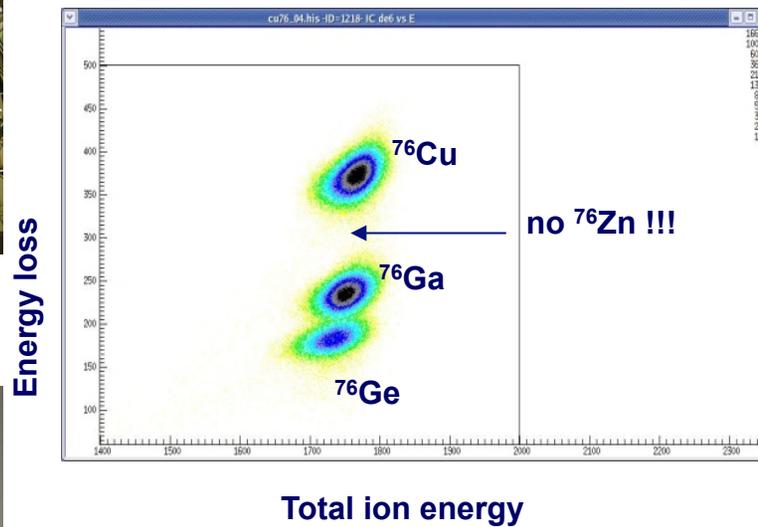
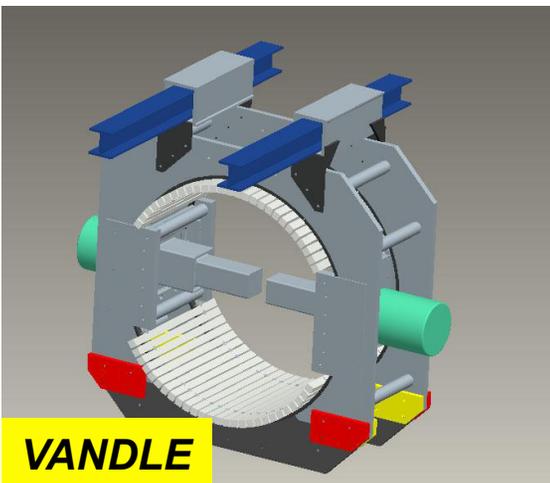
- 4. Combining high-res γ -data, **3Hen**, **MTAS**, **VANDLE**
→ determination of a full β -strength function
(and its consequences)**

Summary

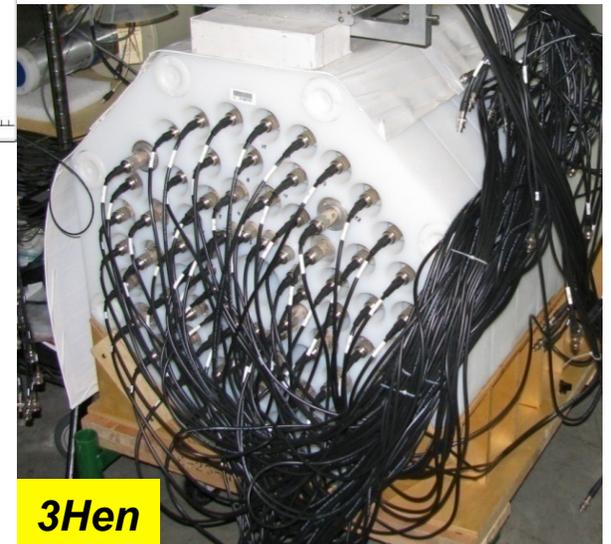
HRIBF has the capabilities to provide reliable input data for the theoretical analysis of β -strength function and for the analysis of processes occurring in nuclear fuels.



Talk by **Miguel Madurga**

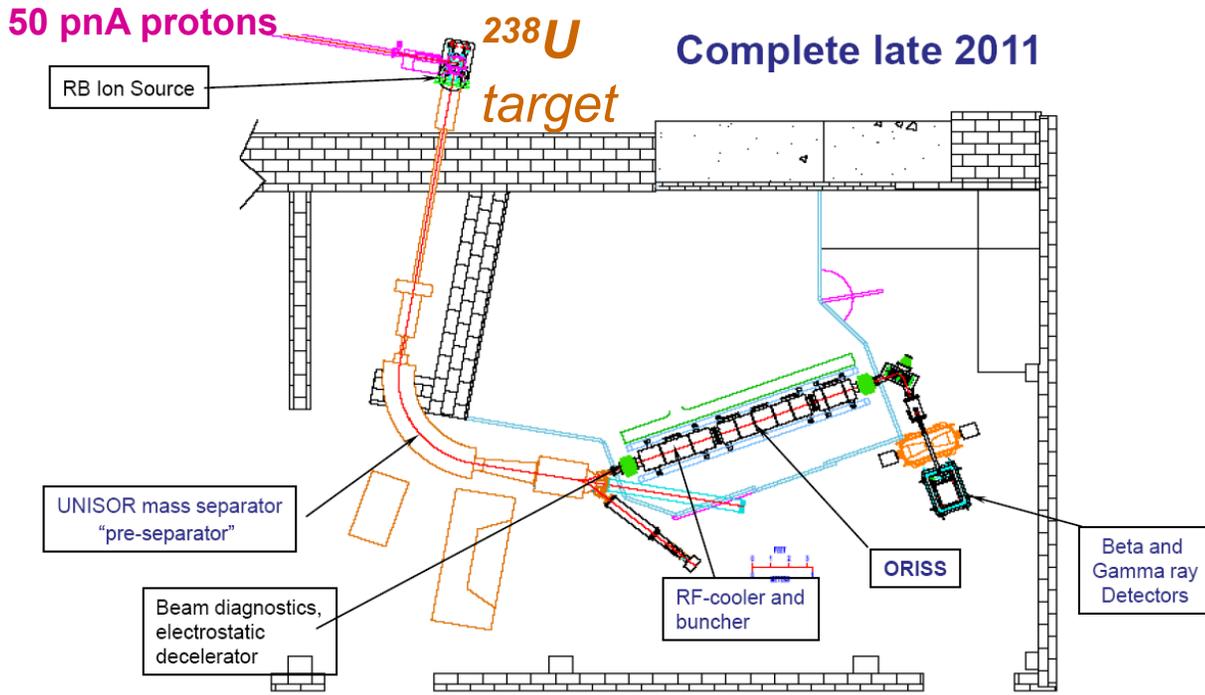


Talk by **Marek Karny**

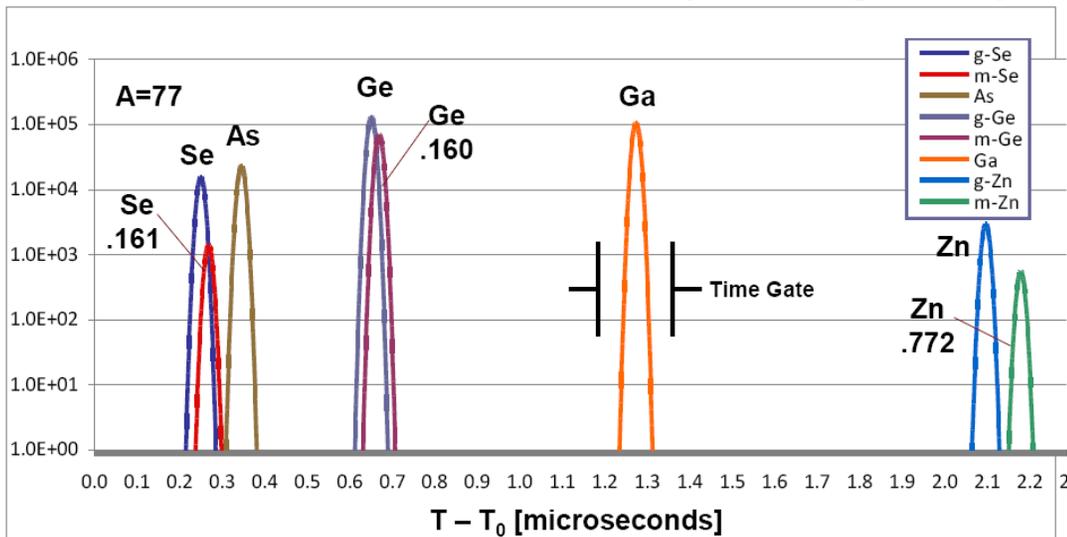
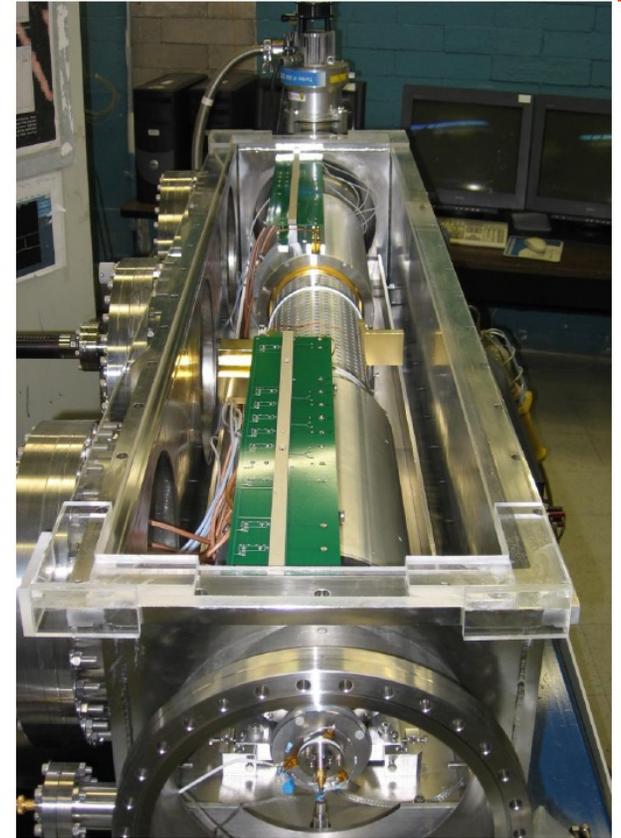


Oak Ridge Isomer Separator and Spectrometer (ORISS)

K. Carter, A. Piechaczek, E. F. Zganjar, J. C. Batchelder and UNIRIB



**based on the
Multi-pass Time of Flight
principle**



**$\Delta M/M \sim 1: 400,000 !!$
efficiency $\sim 50\%$**