



# *Experimental Nuclear Data Activities at ANL*

[kondev@anl.gov](mailto:kondev@anl.gov)

work supported by the Office of Nuclear Physics, Office of Science, US DOE

# Highlights

- ❑ **Two projects funded by the Office of Nuclear Physics, Office of Science (ARRA)**
  - ✓ *Measurement of Actinide Neutronic Transmutation Rates with Accelerator mass spectroscopy (MANTRA)* – in collaboration with G. Youinou, G. Palmiotti, M. Salvatores (INL), G. Imel (ISU) and R. Pardo (ANL-PHY)
  - ✓ *Decay data measurements & evaluations for decay heat calculations* – in collaboration with P. Chowdhury (UML) and C. Lister(ANL-PHY)
  
- ❑ **Commissioning of the CARIBU upgrade of ATLAS – very exciting!**
  - ✓ main trust on Nuclear Structure Physics & Nuclear Astrophysics, BUT ...
  - ✓ opportunities for significant improvements of Nuclear Data for beta-delayed gamma's (decay heat), beta-delayed neutrons (project funded by DOE/ONP - collaboration led by G. Savard (ANL-PHY) and N. Scielzo (LLNL)); fission yields; FP reaction cross sections
  
- ❑ **Properties of long-lived isomeric states and development of novel detectors and techniques for ND research**
  - ✓ Studies of  $^{177m}\text{Lu}$  – inelastic neutron acceleration & medical physics
  - ✓ LaBr<sub>3</sub>(Ce) scintillation array – a new ANL LDRD in collaboration with M. Carpenter, S. Zhu and D. Seweryniak (ANL-PHY)

# MANTRA (Measurement of Actinide Neutronic Transmutation Rates with Accelerator mass spectroscopy)



INL: Gilles Youinou, G. Palmiotti, M. Salvatores

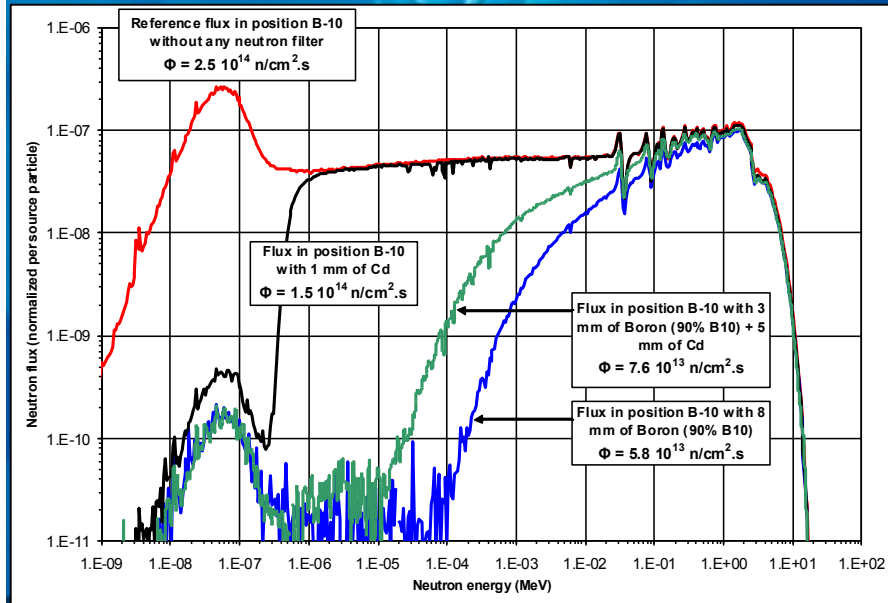
ANL: F.G. Kondev, R. Pardo

ISU: G. Imel.



Basic Concept:

- ✓ irradiating (small) samples of pure MA at the ATR facility at INL
- ✓ measurements using the AMS technique at the ATLAS facility ANL



## Fission for the Future

Argonne, Idaho to study recycling nuclear fuel

[Read the full story »](#)



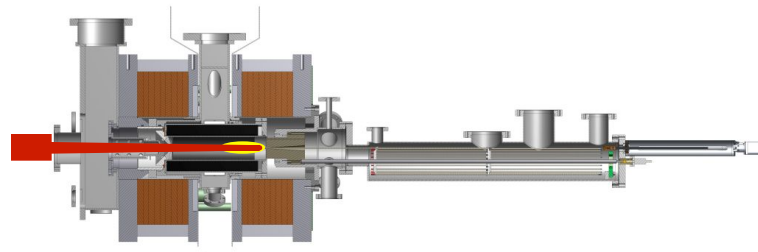
# AMS Challenges for MANTRA

- ❑ Large number of samples:
  - ✓ 13 high purity target materials
  - ✓ 3 different neutron energy spectra
  - ✓ 2-3 samples for each case
  - ✓ non-irradiated material
  - ✓ blanks
  
- ❑ Desired accuracy of results: ~2%!
  - ✓ No cross-talk between samples
  - ✓ Stable, repeatable transmission between source and ion detector
  
- ❑ Small sample size (few mg total, actinide component <1mg)
  - ✓ Reduce radiological problems with samples
  
- ❑ Limited “Z” (elemental) resolution of the detector system



# Improved Configuration for AMS

- ❑ Laser ablation of material into ECR Source
- ❑ Automated multi-sample changer for source



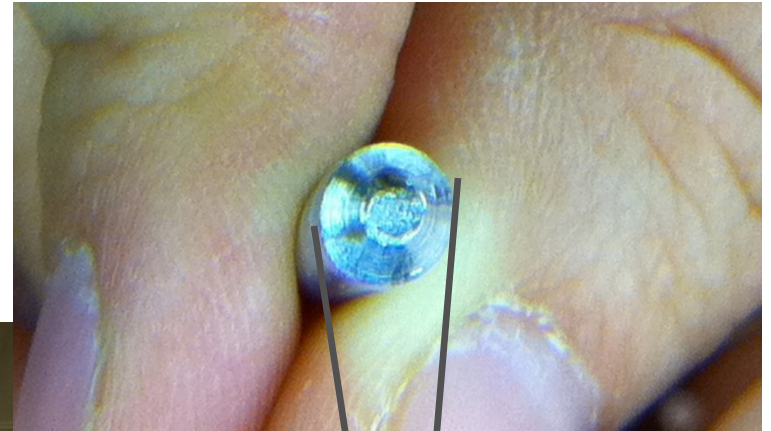
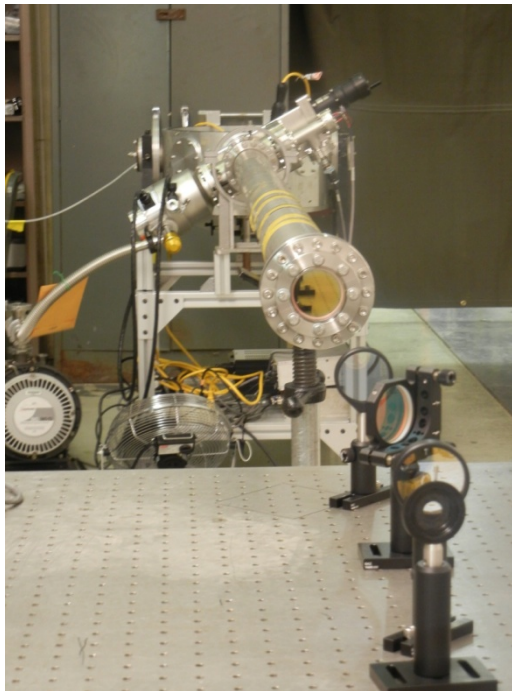
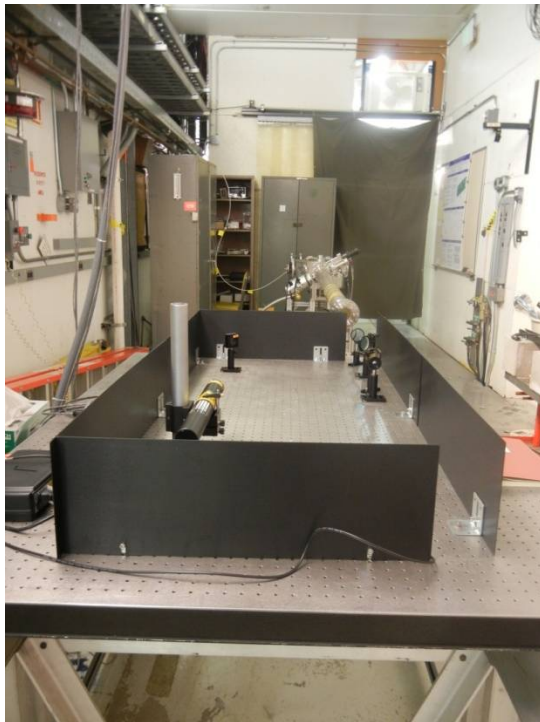
- ❑ Fully automate experiment, much as in  $^{14}\text{C}$  AMS
  - ✓ Automated switching of accelerator between (m/q) configurations
  - ✓ Automated sample changing
  - ✓ Integrated data acquisition
- ❑ Improved detector electronics to improve sensitivity, reduced background and increase in “Z” resolution

# Off line laser ablation test facility

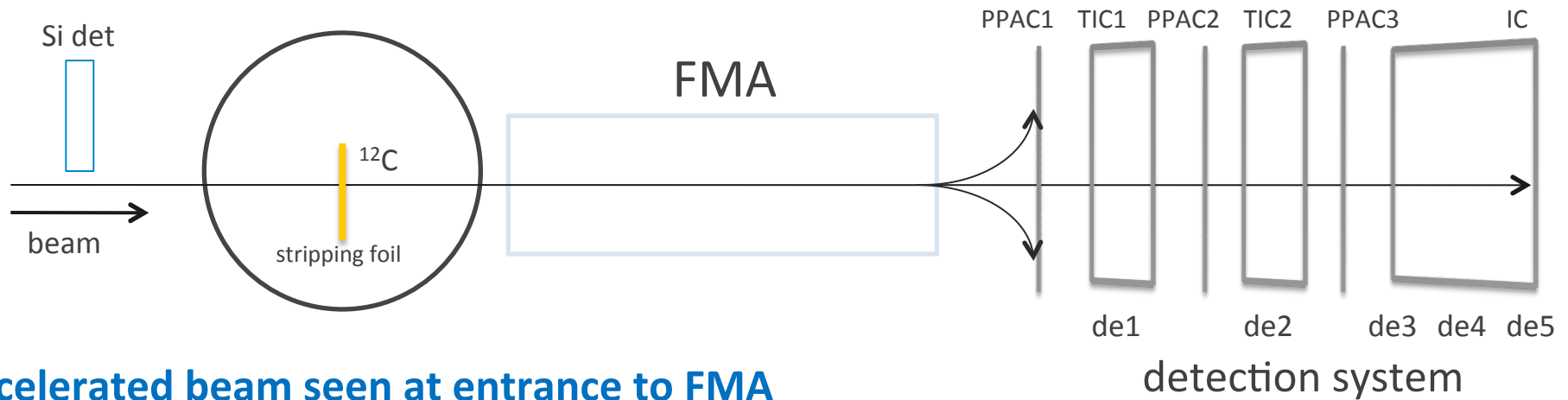
## A Passat Diode-Pumped Solid-State HELPP 1064 Laser

First measurements on stable targets

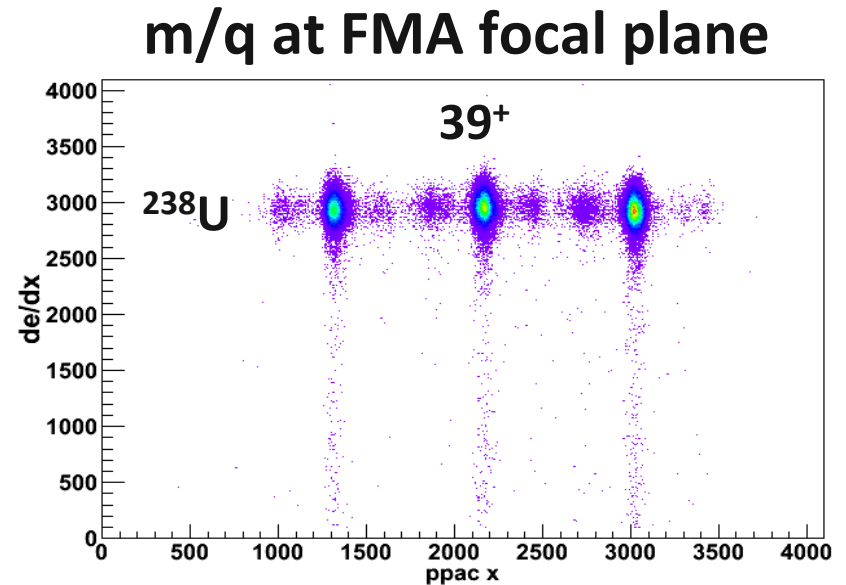
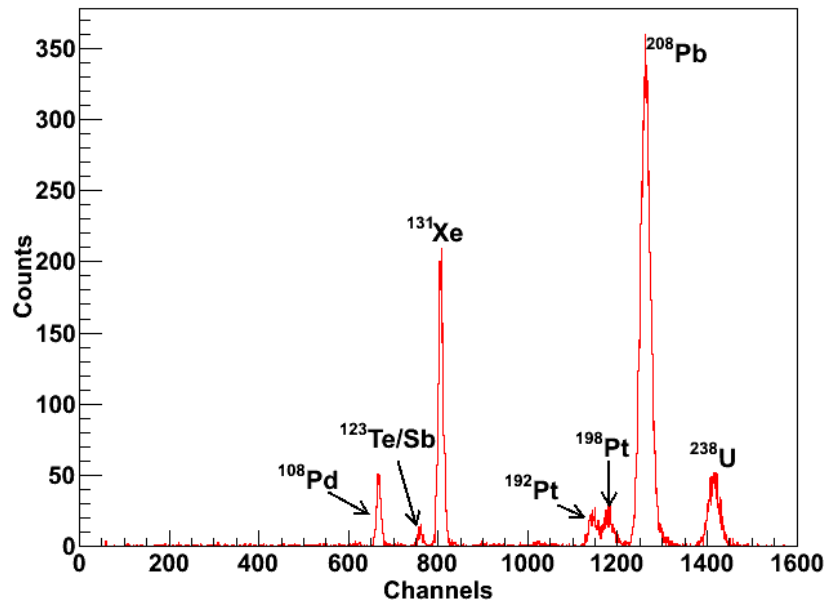
- ✓ Ablation rates appear more than adequate
- ✓ angular distribution of ablated material very sharply peaked at the normal



# MANTRA test run: December 2010

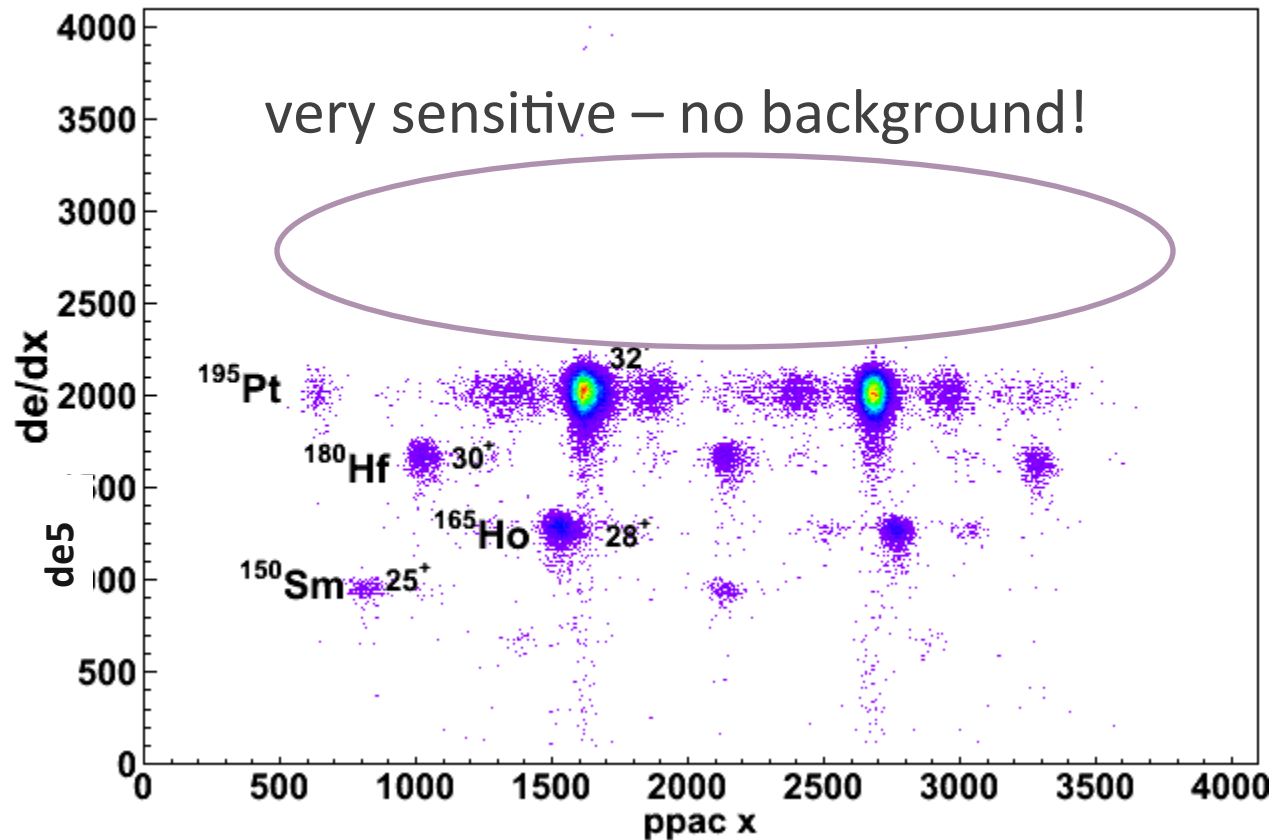


accelerated beam seen at entrance to FMA



# MANTRA test run – cont'd

blank sample: FMA scaled for  $^{240}\text{X}^{39+}$



bring all components together – full test run in March 2012 – irradiated samples in the summer/fall of 2012



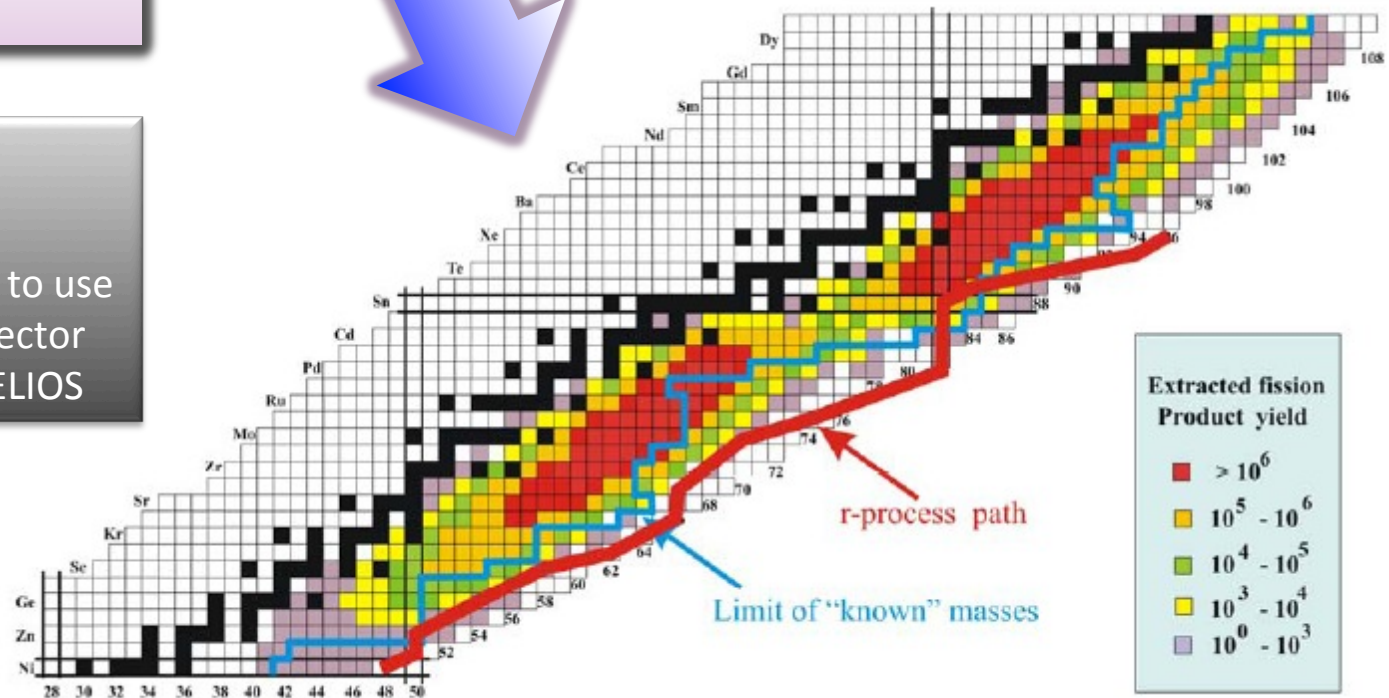
# CARIBU

❑ **C**alifornium **R**are **I**on **B**reeder **U**ppgrade (**CARIBU**) of ATLAS –  
1 Ci  $^{252}\text{Cf}$  spontaneous fission source (~20% of total activity  
extracted as ions) - gas catcher and isobar separator - with or  
without post acceleration

not accelerated - short  $T_{1/2}$  and  
more exotic

sufficient yields for post  
accelerated FP decay  
spectroscopy – capability to use  
some other powerful detector  
equipment – GS, FMA, HELIOS

no stopovers for  
refractory elements!

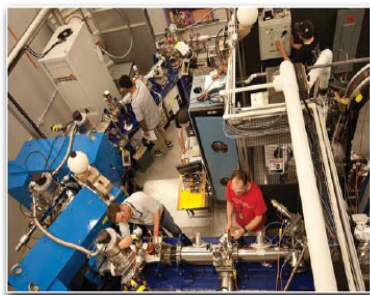




# Decay Workshop at ANL

## Workshop on "Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics"

April 14-16, 2011 at



A workshop on "Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics" will be held at Argonne National Laboratory on April 14-16, 2011.

The aim of the workshop is to discuss opportunities for decay studies at the Californium Rare Isotope Breeder Upgrade (CARIBU) of the ATLAS facility with emphasis on advanced fuel cycle (AFC) applications, nuclear structure and astrophysics research. The workshop will consist of review and contributed talks. Presentations by members of the local groups, outlining the status of relevant in-house projects and available equipment, will also be organized. Time will also be set aside to discuss and develop working collaborations for future decay studies at CARIBU.

Topics of interest include:

- Decay data of relevance to AFC applications with emphasis on reactor decay heat
- Discrete high-resolution gamma-ray spectroscopy following radioactive decay and related topics
- Calorimetric studies of neutron-rich fission fragments using Total Absorption Gamma-ray Spectrometry (TAGS) technique
- Beta-delayed neutron emissions and related topics
- Decay data needs for nuclear astrophysics

### Workshop Organizers

Dr. Michael Carpenter, Argonne National Laboratory  
Prof. Partha Chowdhury, University of Massachusetts Lowell  
Dr. Jason Clark, Argonne National Laboratory  
Dr. Filip Kondev, Argonne National Laboratory  
Dr. Kim Lister, Argonne National Laboratory  
Dr. Dariusz Seweryniak, Argonne National Laboratory

Please visit the Workshop web site for additional information about registration, program, lodging and transportation to Argonne.

<http://www.ne.anl.gov/capabilities/nd/AFC-Apr11/>



<http://www.ne.anl.gov/capabilities/nd/AFC-Apr11/program.shtml>

14-16<sup>th</sup> April 2011

79 Participants from 13 countries  
and 28 institutions

Aimed at engaging the community in  
CARIBU decay (and accelerated  
beam) physics.

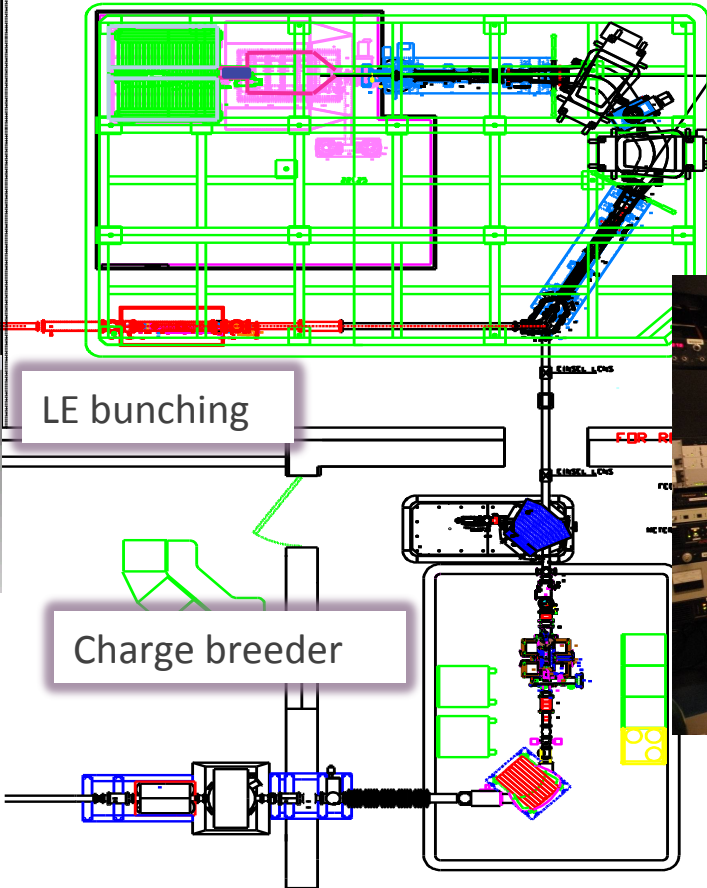
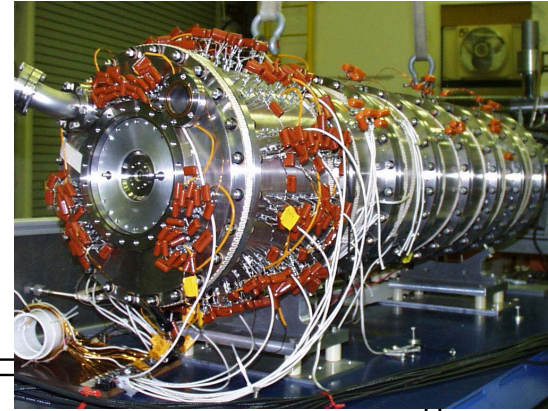
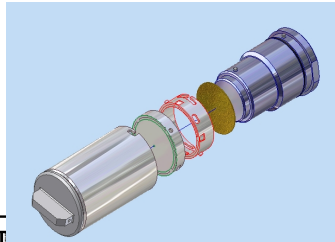
Decay Heat  
Astrophysics  
Nuclear Structure



# CARIBU "front end" layout



1Ci of  $^{252}\text{Cf}$  - 1.9 mg material deposited on a  $\sim 2$  cm disk

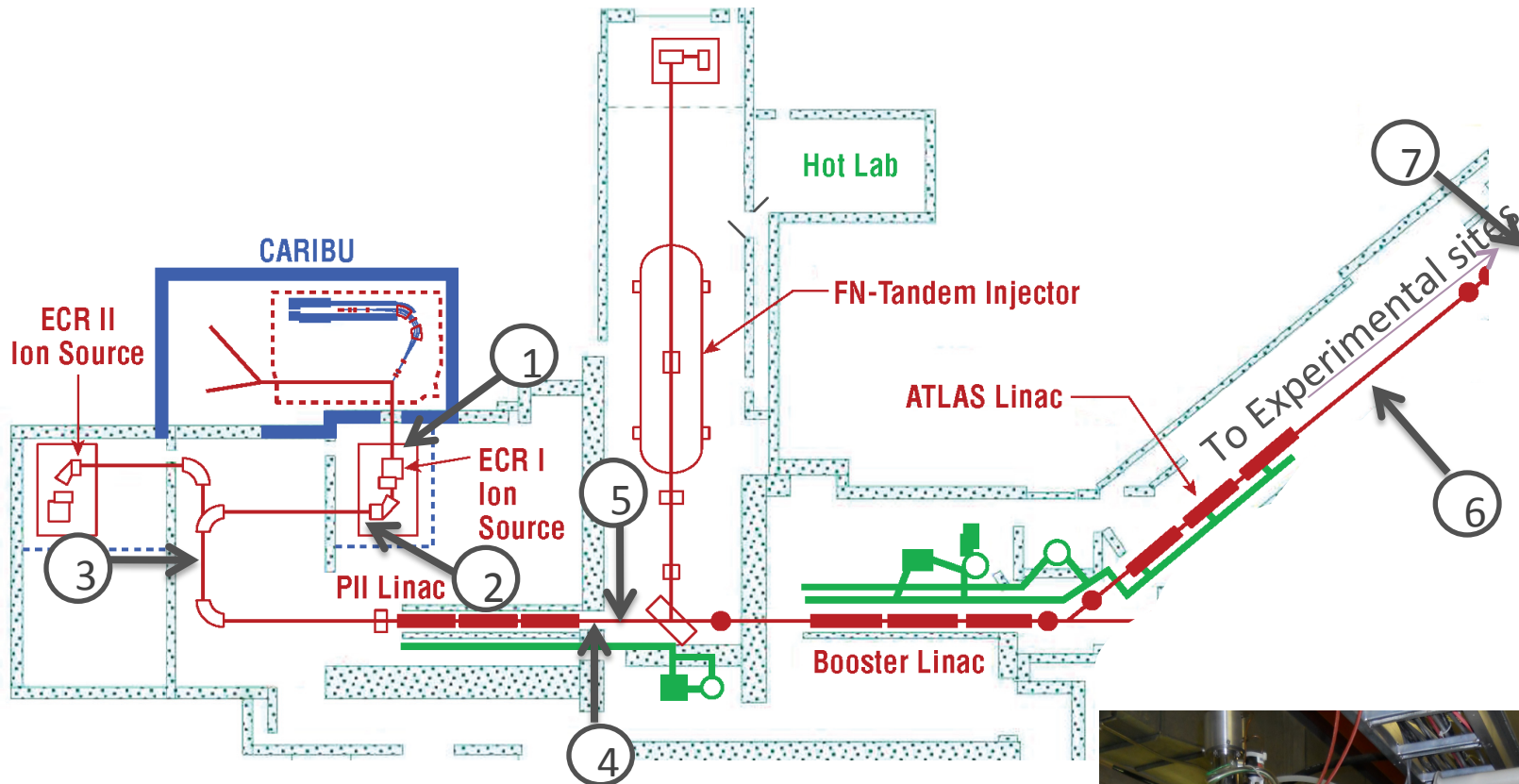


- ✓ FP thermalization
- ✓ quick FP extraction – 5-20 ms with high efficiency -45%
- ✓ form a low-emittance beam for post-acceleration

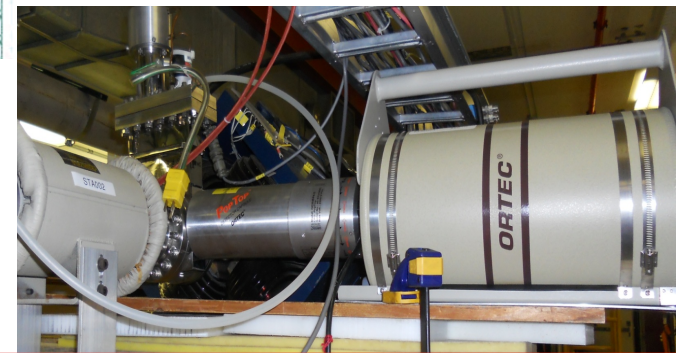


purifying the beam  
 $\Delta M/M \sim 1/20000$

# Beam intensities - “Reference points”



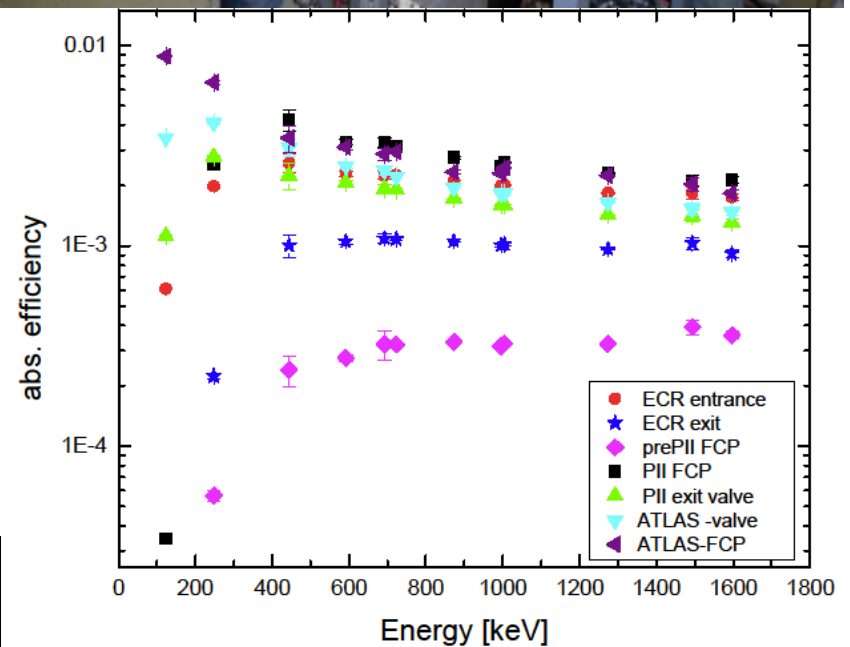
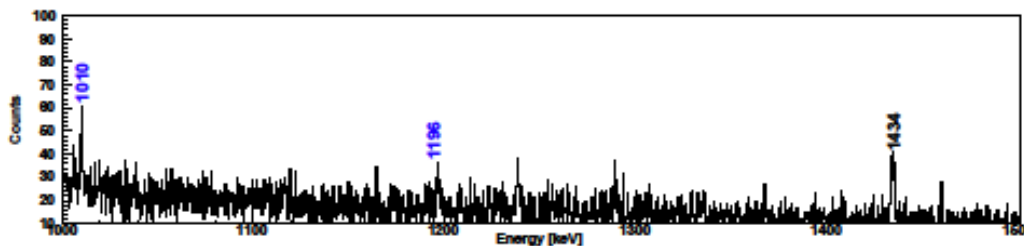
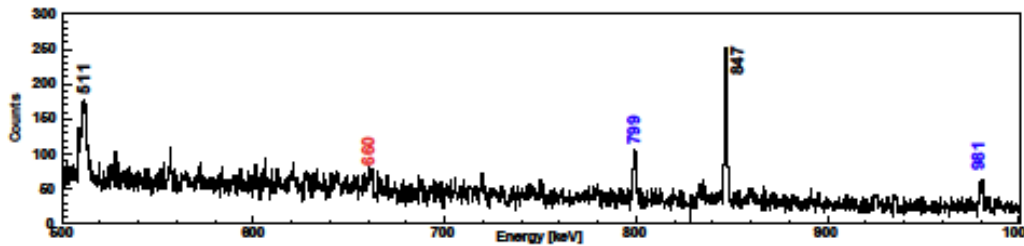
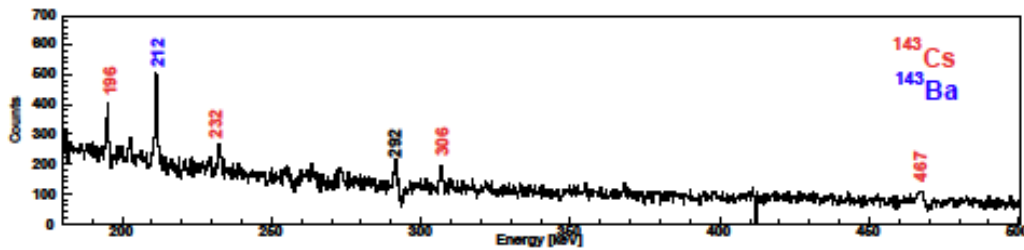
- ✓ 1- ECR entrance
- ✓ 2- ECR exit
- ✓ 3- pre-PII FCP
- ✓ 4 – PII FCP
- ✓ 5- PII exit valve
- ✓ 6- ATLAS valve
- ✓ 7-ATLAS FCP



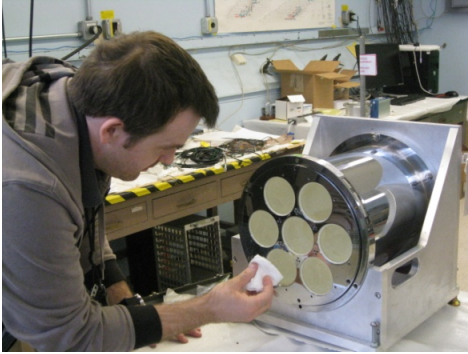


# CARIBU Beam Diagnostics through ATLAS

As always with gamma rays .....  
**ABSOLUTE** yields need careful calibration.....

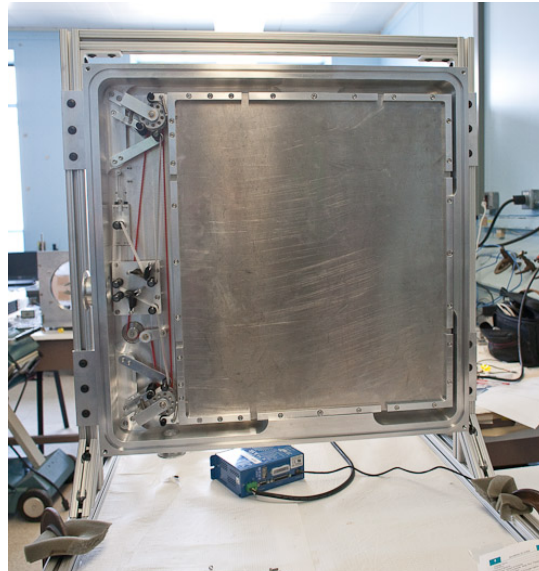


# Decay Station at CARIBU



TAGS

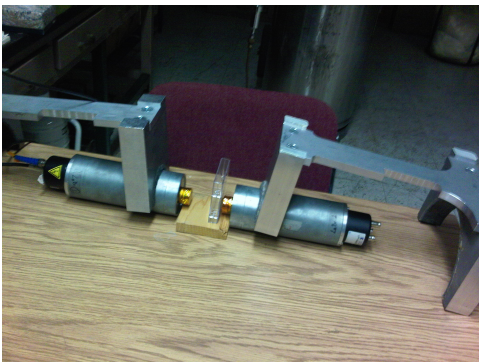
NaI from INL (Greenwood et al.)



Tape Systems (deployed at CARIBU) adopted design from LSU design, but with increased tape-advance rate (~2m in 0.1s)

Construction: P. Bertone / B. DiGiovine

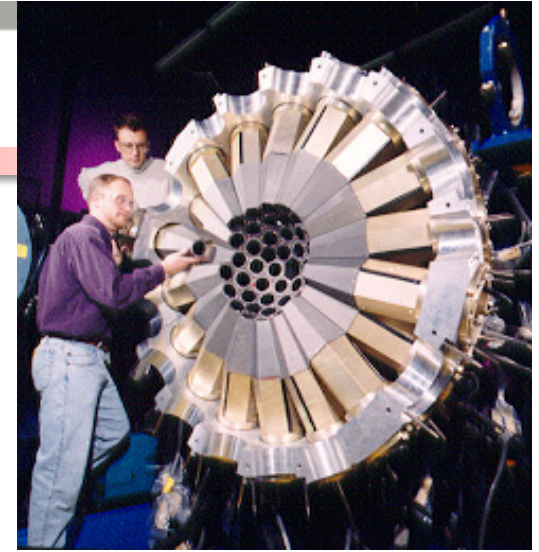
plastic scintillators & Si detectors  
LaBr<sub>3</sub>(Ce) – a new LDRD grant – M.P. Carpenter & F.G. Kondev – up to 10 det.



X-array

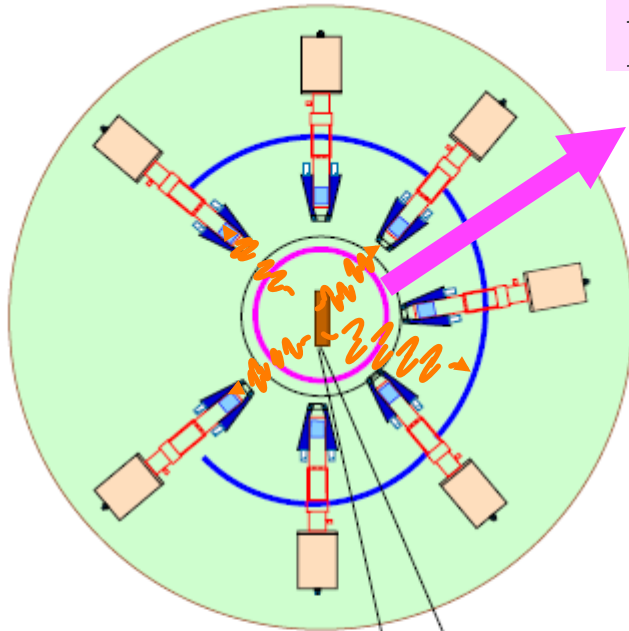
One “Super-Clover” & four 70 X 70 mm crystals - a pack with ~275 % efficiency relative to a 3” x 3” NaI

# $\beta^-$ counting station with GS

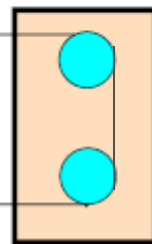


GAMMASPHERE

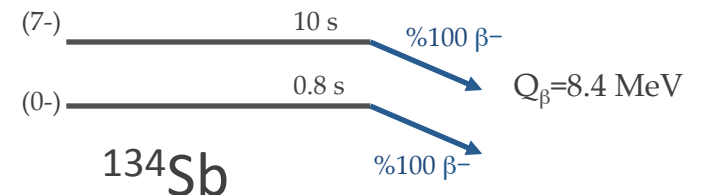
plastic scint. ( $\beta^-$ )



tape system



- ✓ high resolution & sensitivity
- ✓ powerful  $\beta^- \gamma \gamma$  coin – resolving weak cascades & isomers!



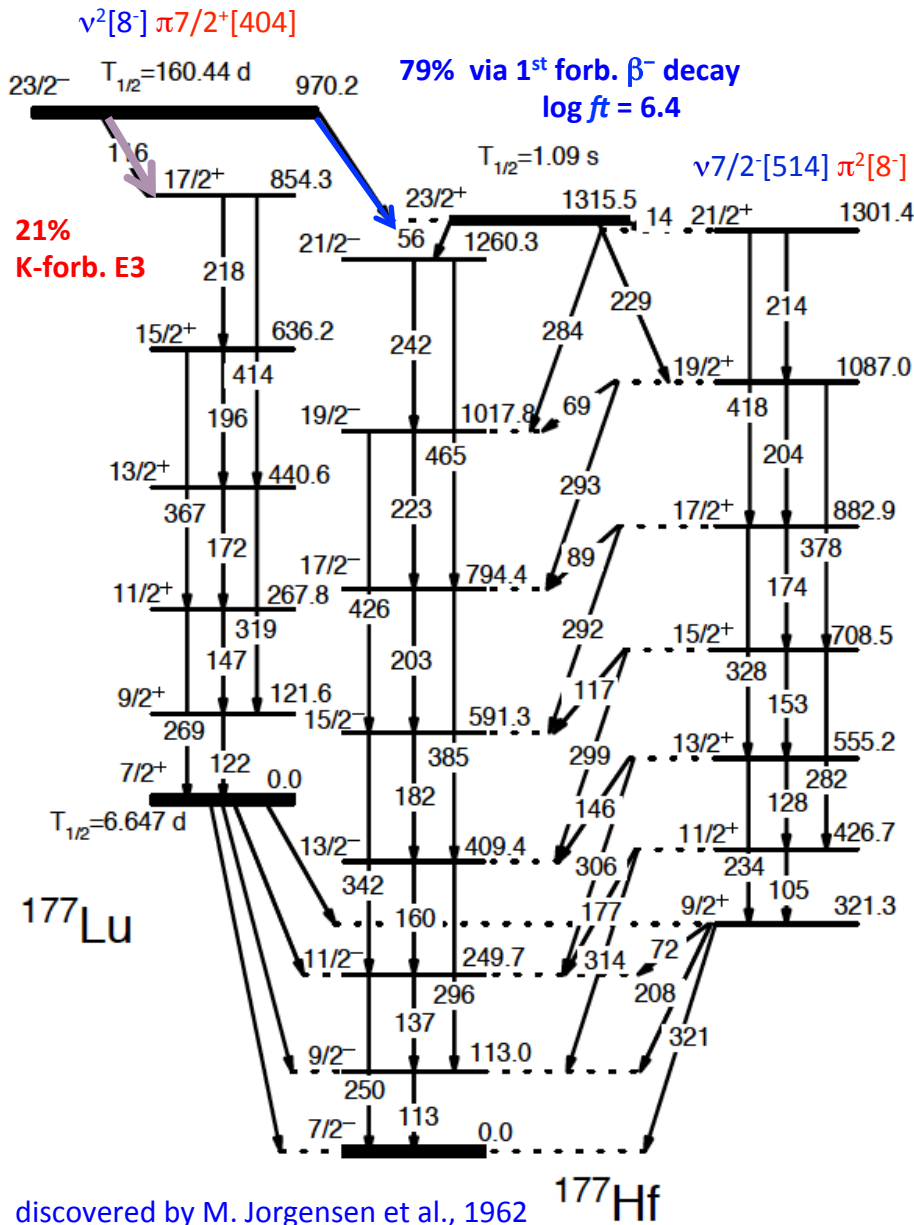
## GS as a calorimeter

- ✓ P. Reiter et al. Phys. Rev. Lett. 84, 3542 (2000)
- ✓ with a modest upgrade – suitable for  $\beta^-$  decay studies – HRGS & TAGS





# $K^\pi=23/2^-$ isomer in $^{177}\text{Lu}$

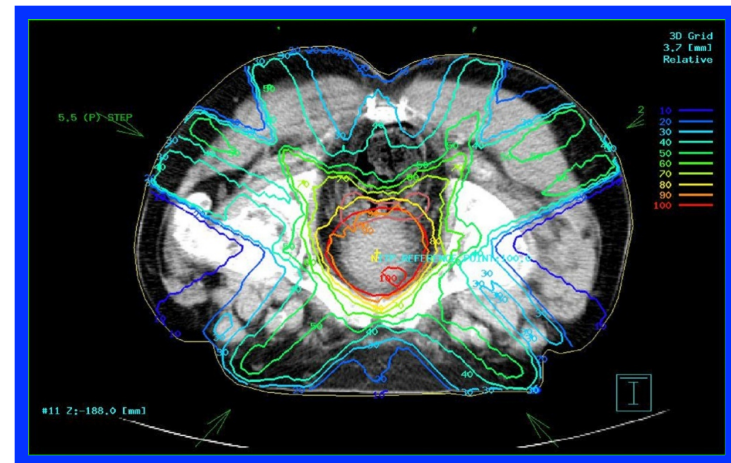


PHYSICAL REVIEW C 83, 064617 (2011)

## Direct evidence for inelastic neutron "acceleration" by $^{177}\text{Lu}^m$

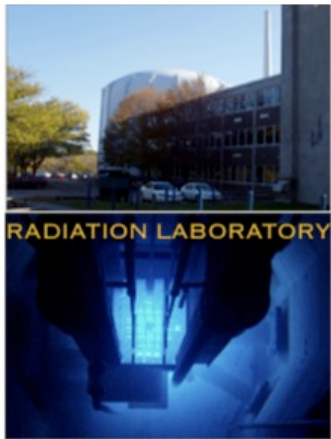
O. Roig,<sup>1,\*</sup> V. Méot,<sup>1</sup> B. Rossé,<sup>1</sup> G. Bélier,<sup>1</sup> J.-M. Daugas,<sup>1</sup> A. Letourneau,<sup>2</sup> A. Menelle,<sup>3,4</sup> and P. Morel<sup>1</sup>

huge cross section  
 $\sigma=146$  (19) b vs 258 (58) b  
 still need explanation



impurity in the production of the  
 medical isotope  $^{177}\text{Lu}$  ( $T_{1/2}=6.647$  d)

# $^{177m}\text{Lu}$ Isomer production



$^{176}\text{Lu}(n,\gamma)^{177m}\text{Lu}$  @ UML

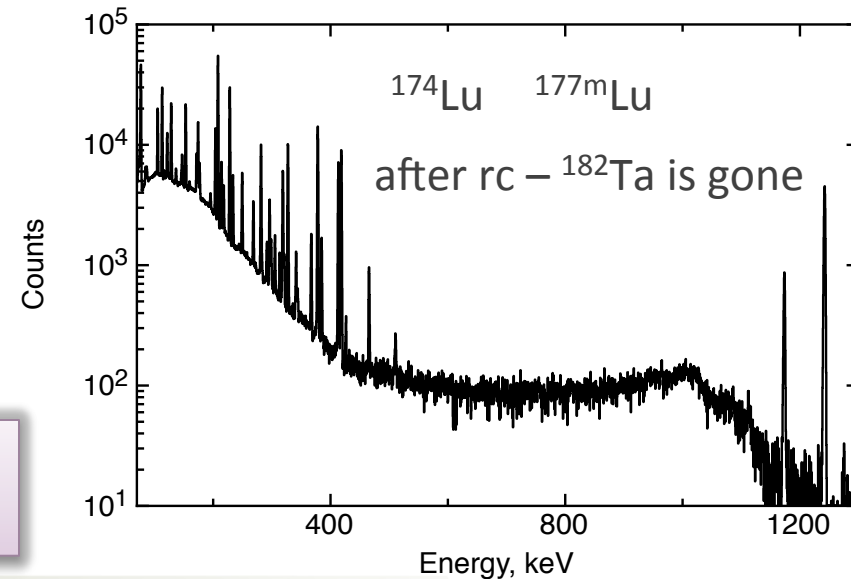
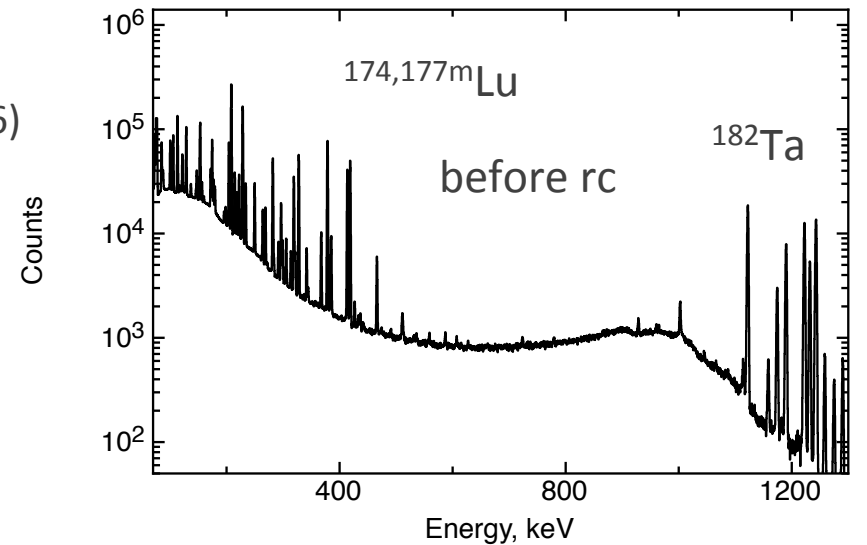
- ✓ natural Lu target material
- ✓ irradiation (4/24/2003 – 4/18/2006)
- ✓ total neutron fluence:  $3.5 \times 10^{19}$
- ✓ ~4 yr of cooling

$^{177}\text{W}$ 132 M ε: 100.00%	$^{178}\text{W}$ 21.6 D ε: 100.00%	$^{179}\text{W}$ 37.05 M ε: 100.00%	$^{180}\text{W}$ 1.8E+18 Y 0.12% α: 100.00%	$^{181}\text{W}$ 121.2 D ε: 100.00%	$^{182}\text{W}$ >8.3E+18 Y 26.50% α	$^{183}\text{W}$ >1.3E+19 Y 14.31% α	$^{184}\text{W}$ >2.9E+19 Y 30.64% α
$^{176}\text{Ta}$ 8.09 H ε: 100.00%	$^{177}\text{Ta}$ 56.56 H ε: 100.00%	$^{178}\text{Ta}$ 9.31 M ε: 100.00%	$^{179}\text{Ta}$ 1.82 Y ε: 100.00%	$^{180}\text{Ta}$ 8.154 H ε: 86.00% β-: 14.00%	$^{181}\text{Ta}$ STABLE 99.98%	$^{182}\text{Ta}$ 114.43 D β-: 100.00%	$^{183}\text{Ta}$ 5.1 D β-: 100.00%
$^{175}\text{Hf}$ 70 D ε: 100.00%	$^{176}\text{Hf}$ STABLE 5.26%	$^{177}\text{Hf}$ STABLE 18.60%	$^{178}\text{Hf}$ STABLE 27.28%	$^{179}\text{Hf}$ STABLE 13.62%	$^{180}\text{Hf}$ STABLE 35.08%	$^{181}\text{Hf}$ 42.39 D β-: 100.00%	$^{182}\text{Hf}$ 8.90E+6 Y β-: 100.00%
$^{174}\text{Lu}$ 3.31 Y ε: 100.00%	$^{175}\text{Lu}$ STABLE 97.41%	$^{176}\text{Lu}$ 3.76E+10 Y 2.59% β-: 100.00%	$^{177}\text{Lu}$ 6.647 D β-: 100.00%	$^{178}\text{Lu}$ 28.4 M β-: 100.00%	$^{179}\text{Lu}$ 4.59 H β-: 100.00%	$^{180}\text{Lu}$ 5.7 M β-: 100.00%	$^{181}\text{Lu}$ 3.5 M β-: 100.00%

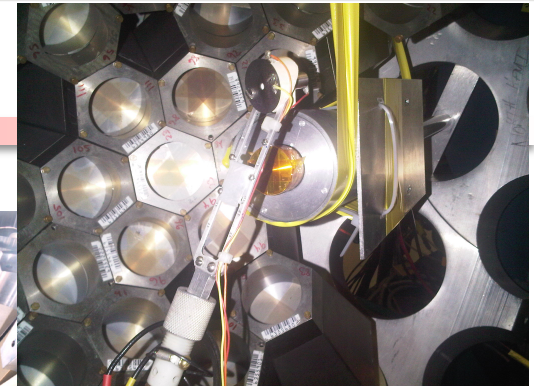
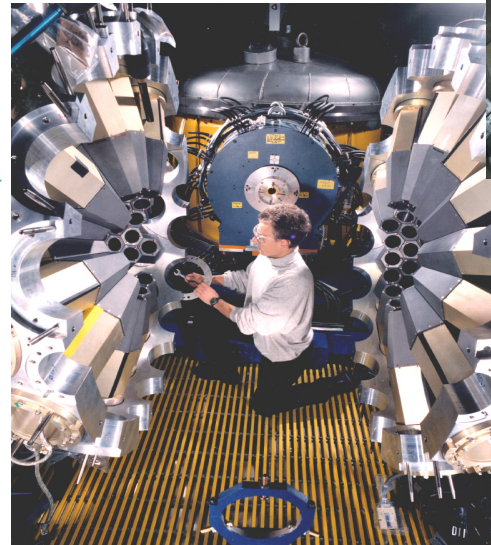
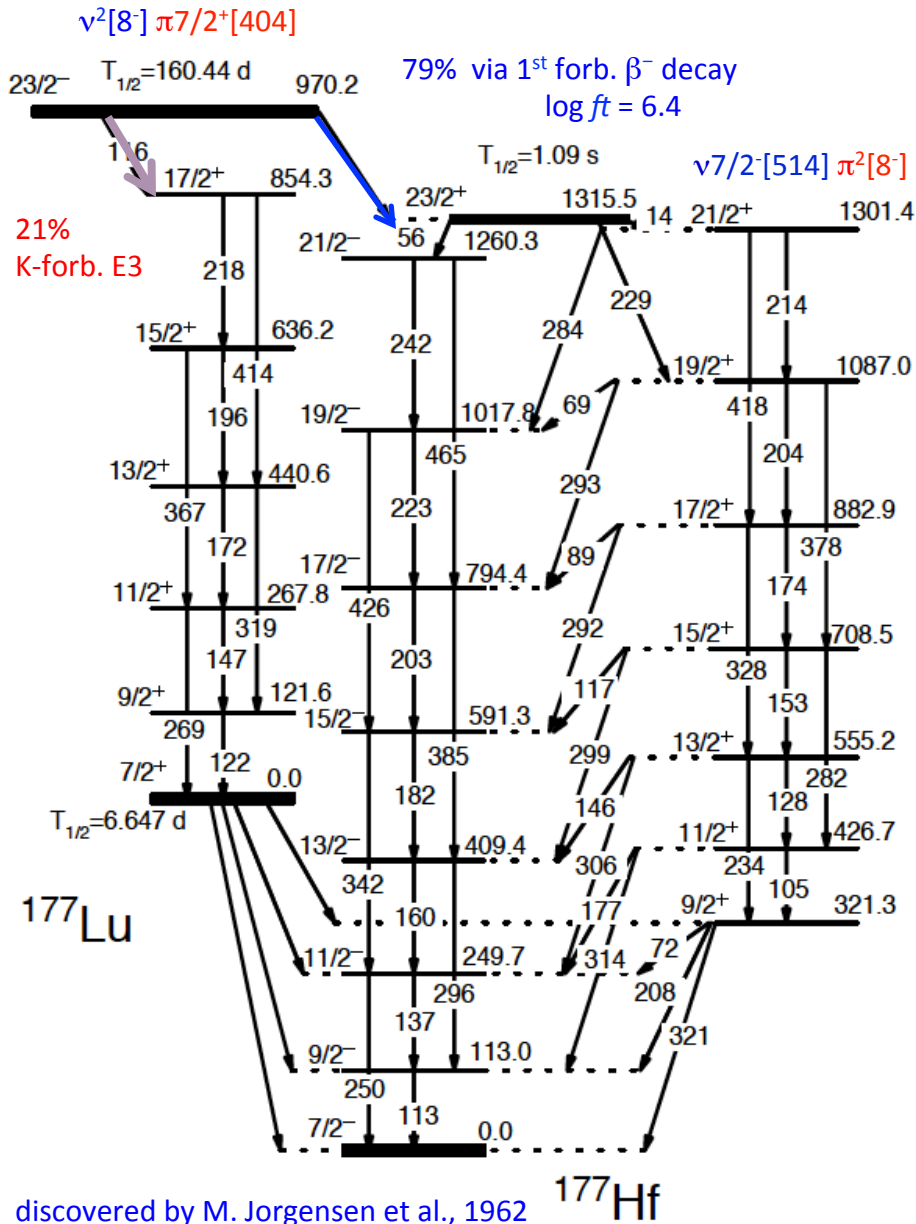
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$  - impurities

radiochemical separation  
John Green & Irshad Ahmad

a single HpGe detector

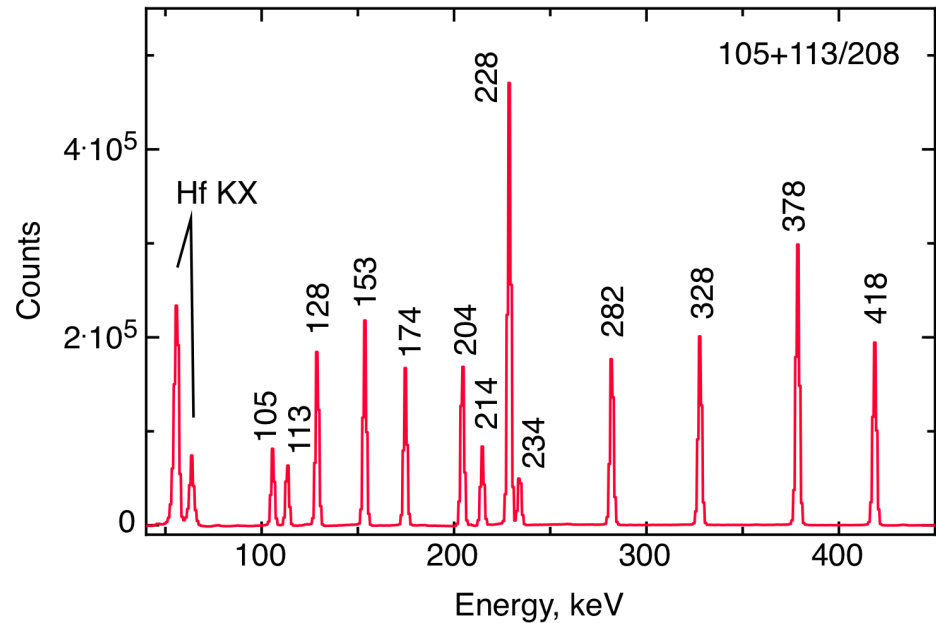


# Decay of $K^\pi=23/2^-$ isomer



*LaBr<sub>3</sub>(Ce)*

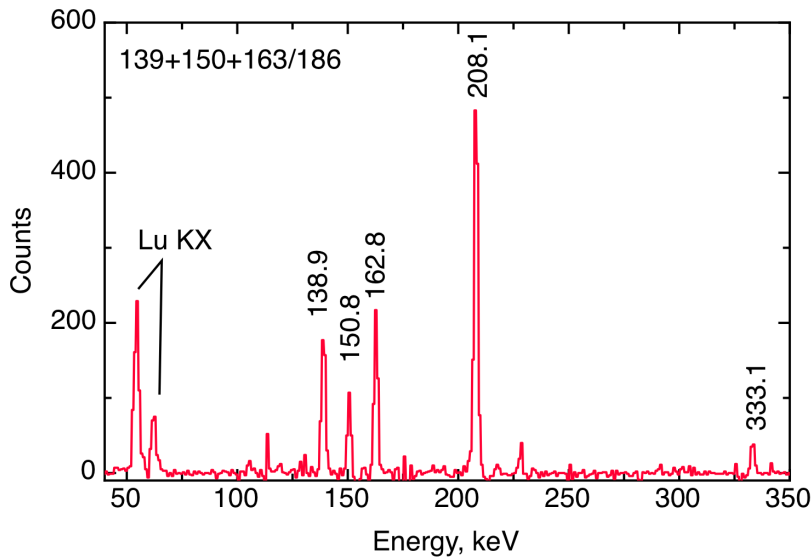
PMT - XP20D0B - Photonis  
collaborators Univ. Sofia  
1" by 1"; 2% at 1.3 MeV



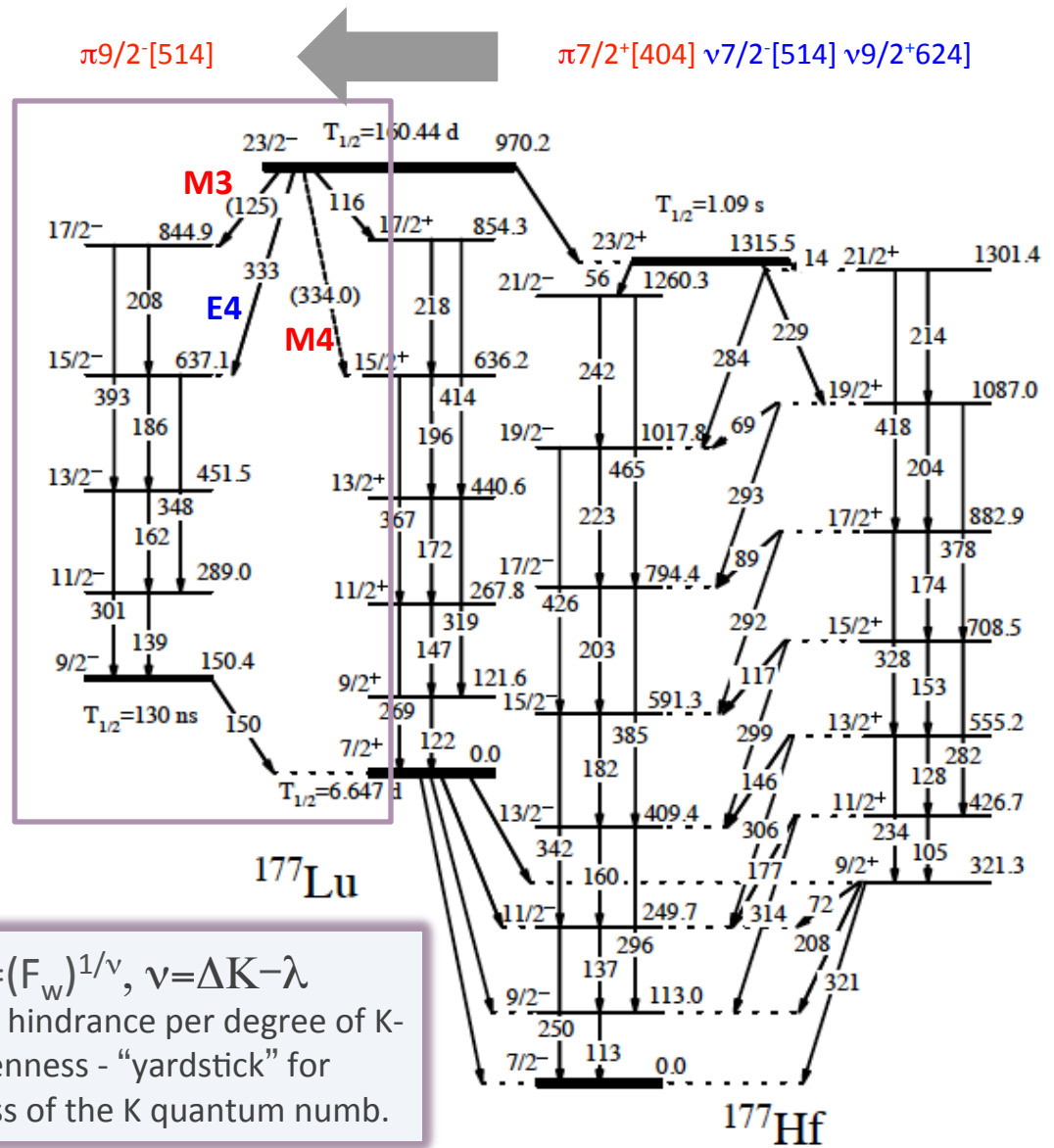


# Decay of $K^\pi=23/2^-$ isomer – cont.

$$\alpha_T(M3)=94 - \alpha_T(E4)=1$$

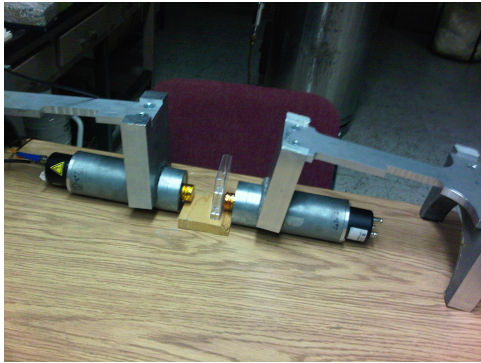


E, keV	Mult	$\nu$	$f_\nu$
116	E3	5	61.5(6) 66 ( $^{178}\text{Hf}$ )
125	M3	4	470 (40) 83 ( $^{177}\text{Hf}$ )
333	E4	3	980 (80)
334	M4	4	>53 72 ( $^{178}\text{Hf}$ )



$f_\nu = (F_w)^{1/\nu}$ ,  $\nu = \Delta K - \lambda$   
 reduced hindrance per degree of K-forbiddenness - "yardstick" for goodness of the K quantum numb.

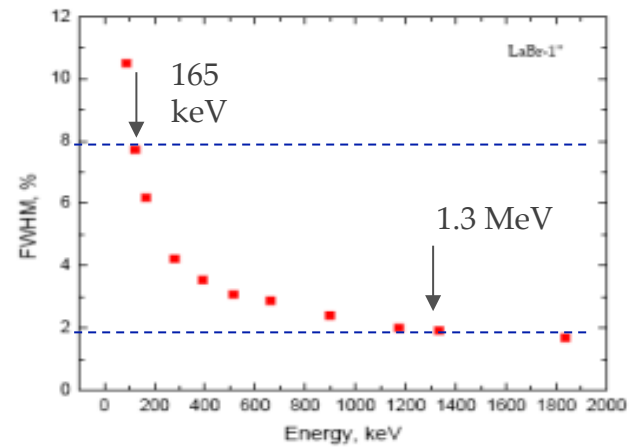
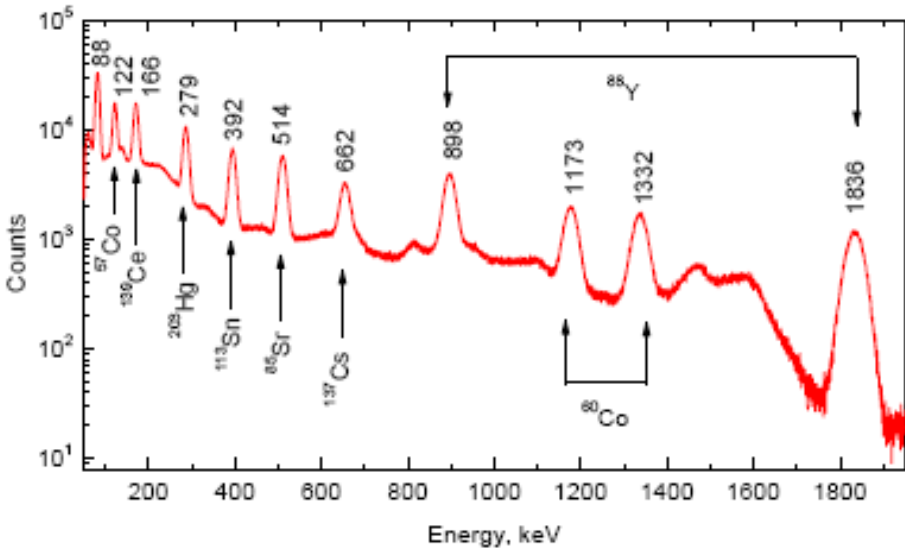
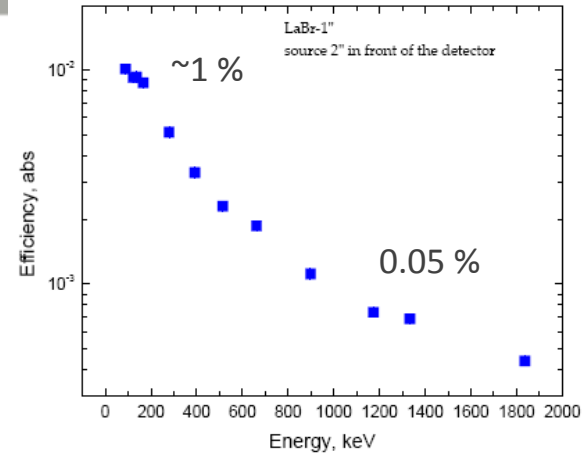




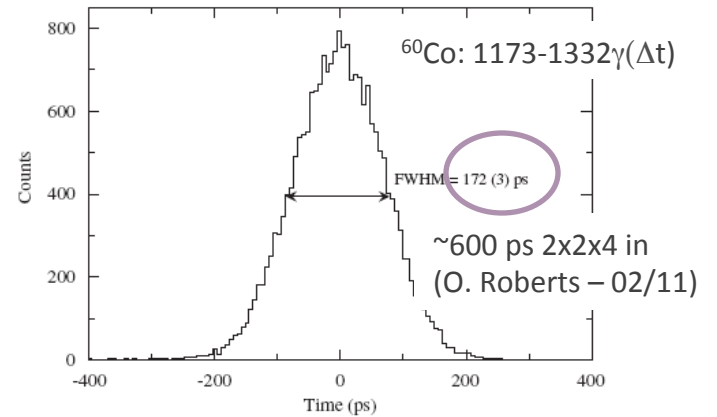
# LaBr<sub>3</sub>(Ce)

	LaBr	NaI	BaF <sub>2</sub>
Density [g/cm <sup>3</sup> ]:	5.29	3.67	4.88
Decay Time [ns]:	16	250	0.7
Light yield [ph/keV]:	63	38	1.8

**PMT - XP20D0B – Photonis**  
collaborators Univ. Sofia



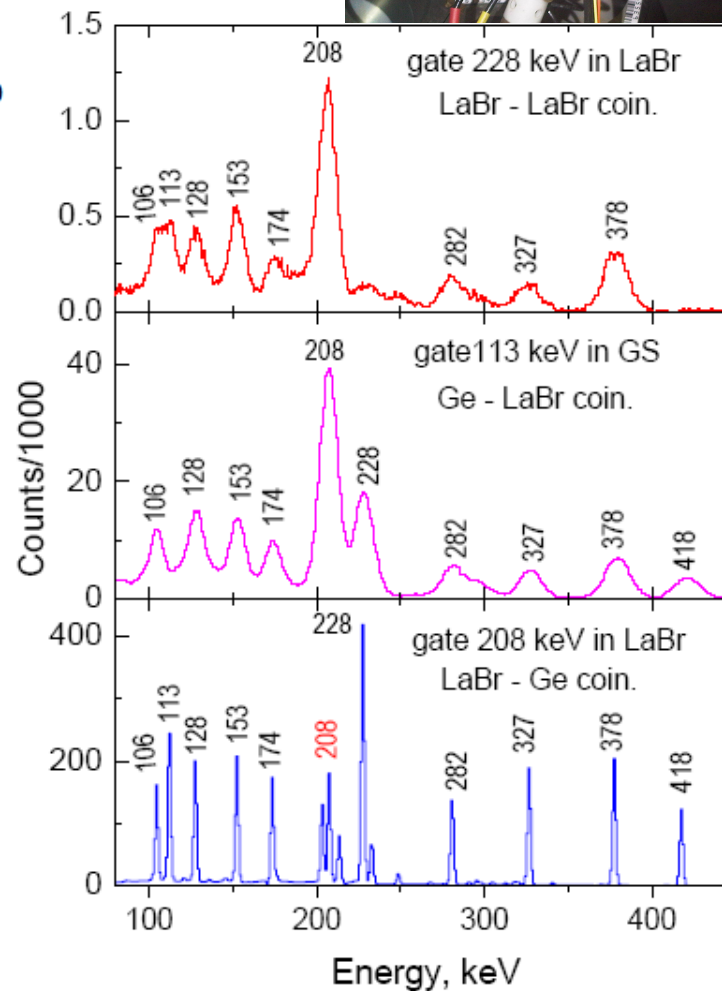
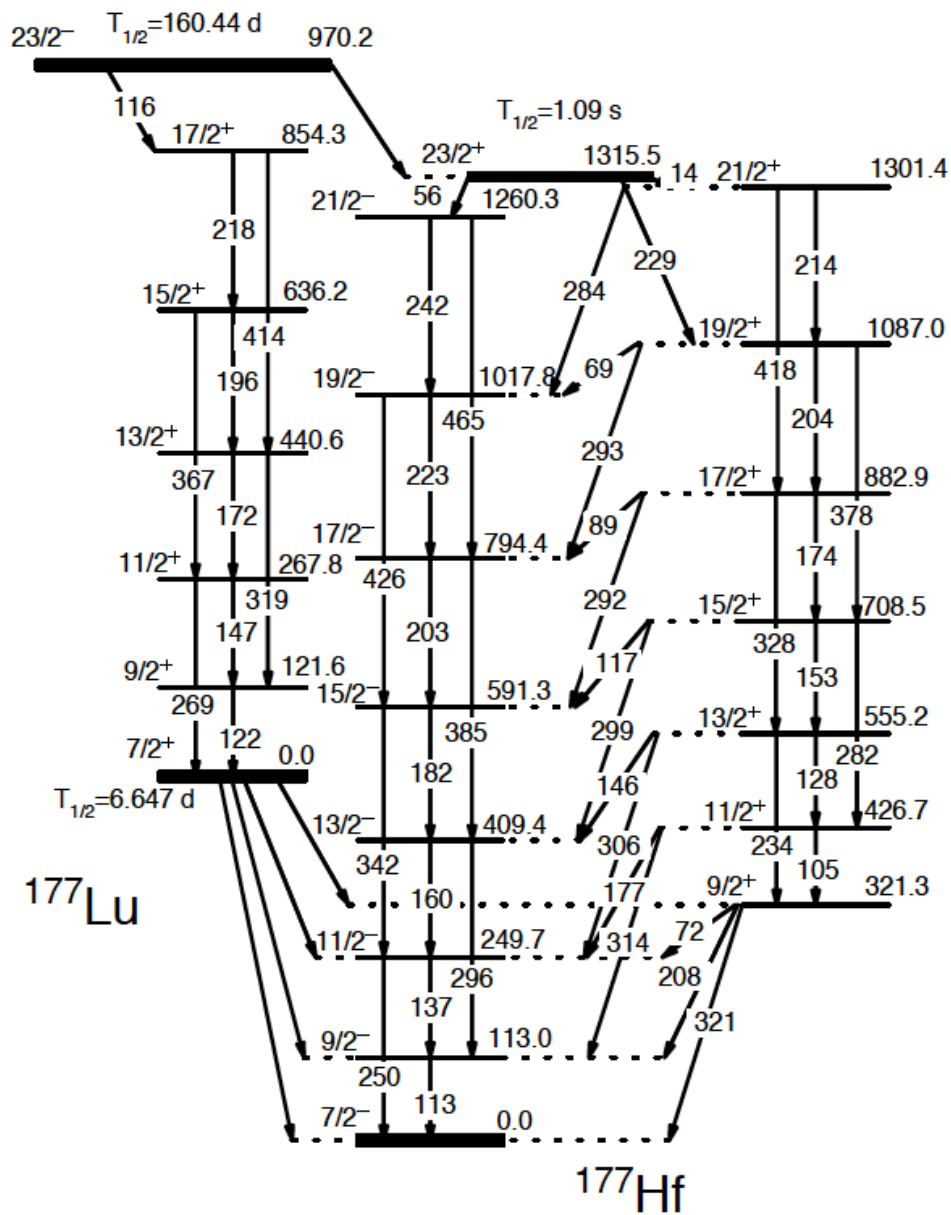
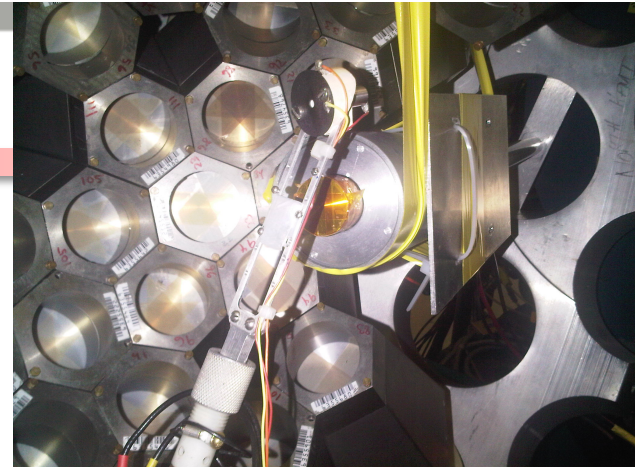
calibrated source & a single HpGe detector



*Good energy resolution & high efficiency*



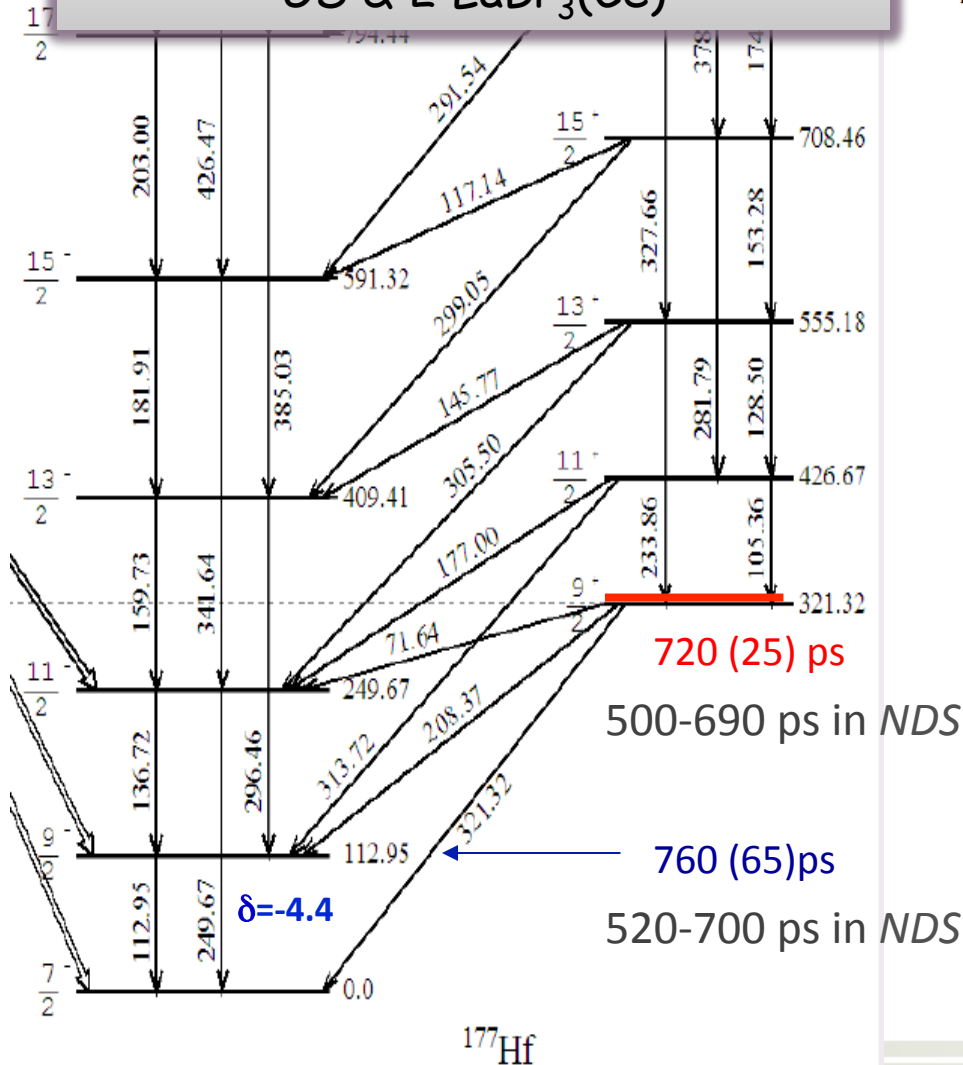
# GS-LaBr<sub>3</sub>(Ce) coincidences



# Fast timing measurements

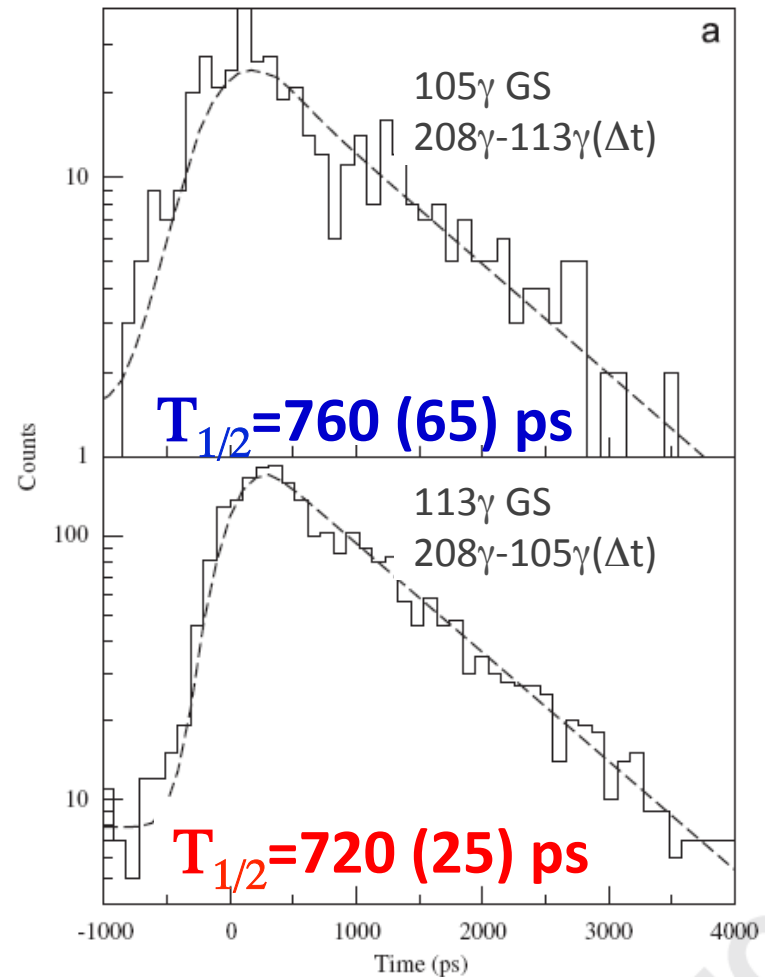


Direct (electronic) timing -  $\gamma$ - $\gamma\gamma(\Delta t)$   
 GS & 2 LaBr<sub>3</sub>(Ce)



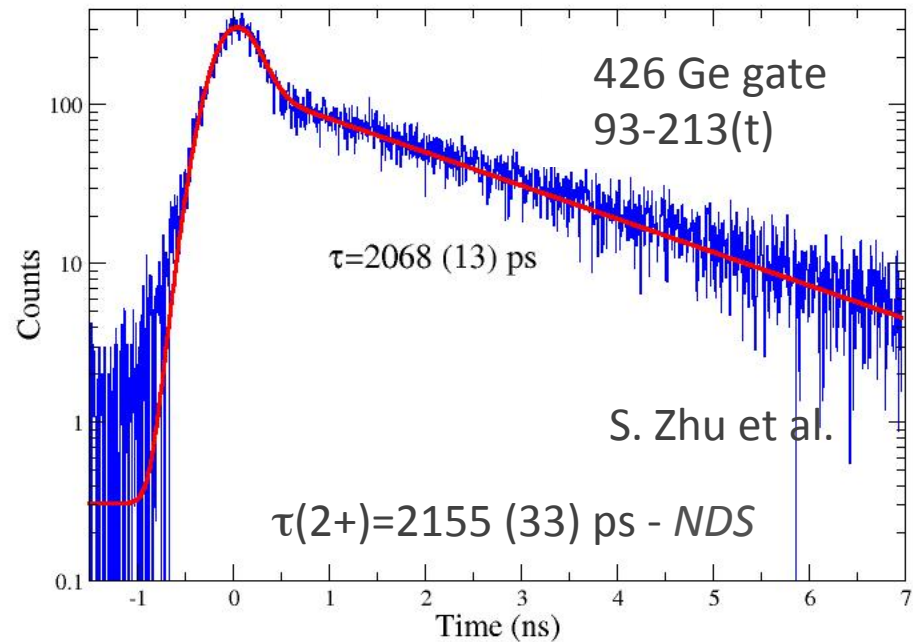
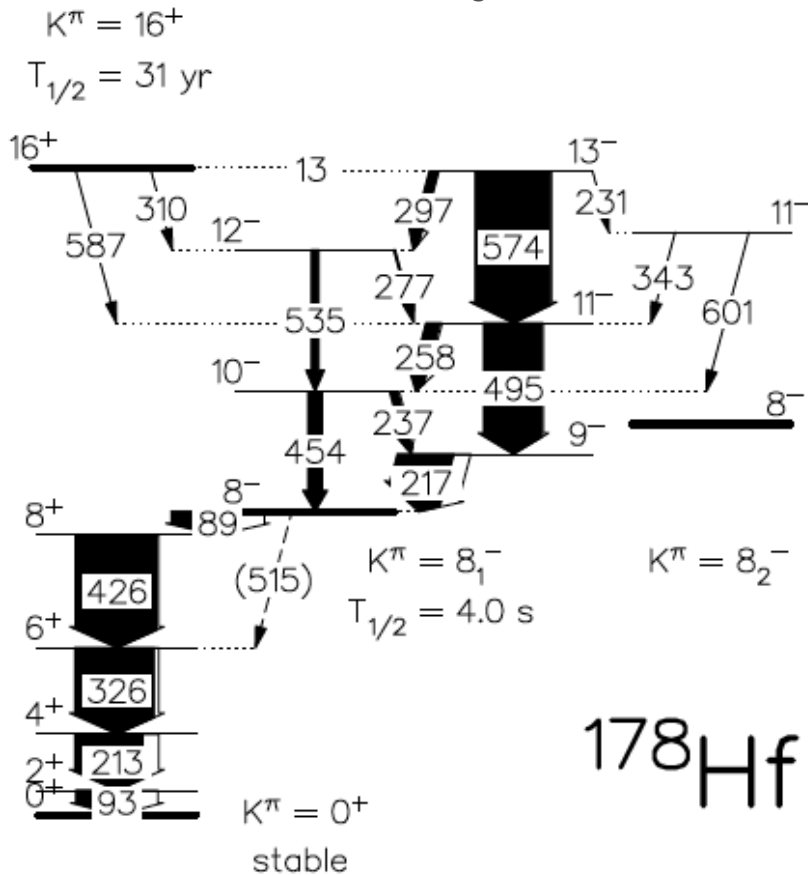
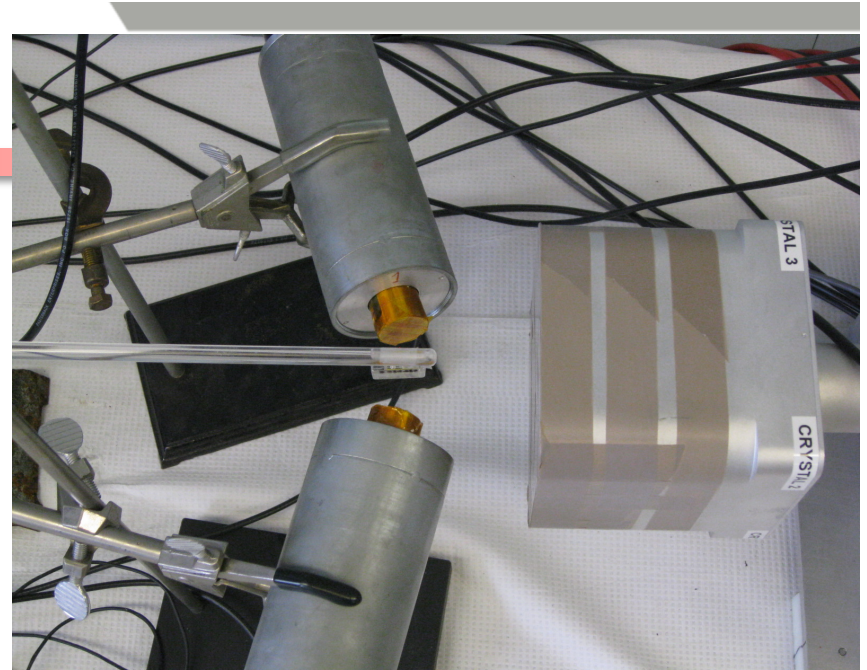
$\gamma$ -ray coincidence and fast-timing measurements using LaBr<sub>3</sub>(Ce) detectors and gammasphere <sup>☆</sup>

S. Zhu <sup>a,\*</sup>, F.G. Kondev <sup>a</sup>, M.P. Carpenter <sup>a</sup>, I. Ahmad <sup>a</sup>, C.J. Chiara <sup>a,b</sup>, J.P. Greene <sup>a</sup>, G. Gurdal <sup>a</sup>, R.V.F. Janssens <sup>a</sup>, S. Lalkovski <sup>c</sup>, T. Lauritsen <sup>a</sup>, D. Seweryniak <sup>a</sup>



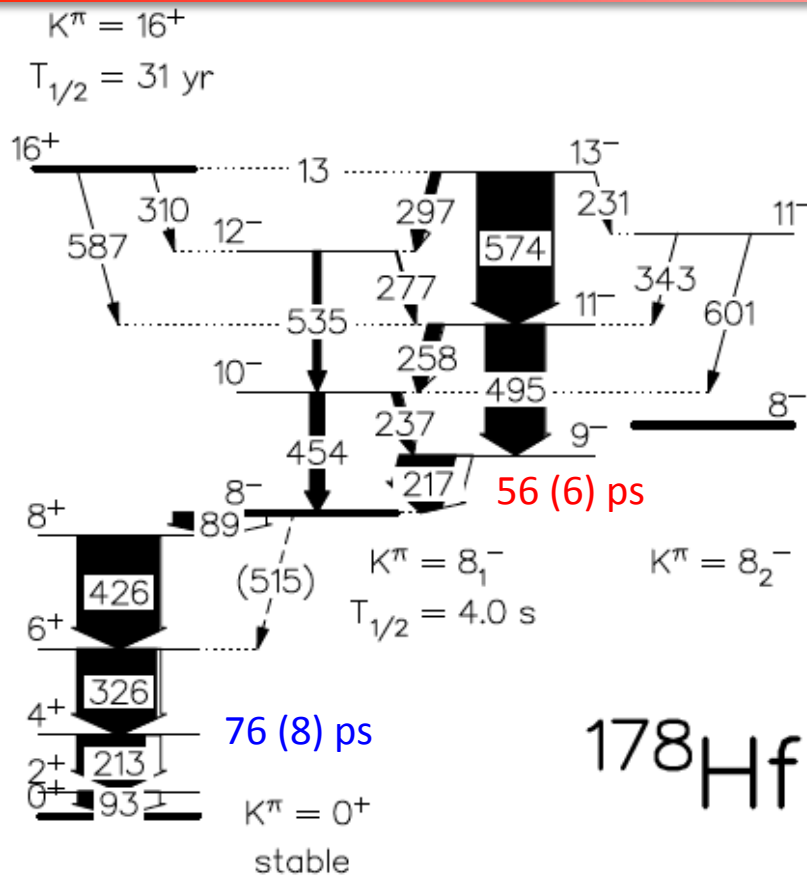
# Studies of $^{178m}\text{Hf}$ decay

$\sim \mu\text{C}$  source produced at LANL  
2 LaBr<sub>3</sub>(Ce) + 1 HpGe clover





# Lifetime of states above the $K^\pi=8^-$ isomer



Configuration mixing between  $\pi^2$  and  $\nu^2$  [8-] bands

- ✓ from log ft values in  $^{178}\text{Lu}$  beta decay
- ✓ from in-band branching ratios
- ✓  $B(M1) \sim (gk-gr)$

