ISOLDE Workshop and Users Meeting 2024

RECENT RESULTS ON THE BETA DECAY OF 152TB AND ITS RELEVANCE IN THERANOSTICS

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IS722 collaboration

Motivation

When performing simulation codes and therapy planners, it is essential to calculate the dose administered to the patient and its spatial distribution, both in diagnosis using **PET** or **SPECT** cameras and in treatment (brachytherapy and theranostics).

The beta decay of radioisotopes of use or potential use in medicine must be well studied and the $I_\beta(E)$ **must be accurate and reliable.**

TAS technique

Contribution to "Diamond Jubilee of RCA"

Alan L. Nichols*

Status of the decay data for medical radionuclides: existing and potential diagnostic γ emitters, diagnostic β^+ emitters and therapeutic radioisotopes

https://doi.org/10.1515/ract-2022-0004
Received January 6, 2022; accepted April 10, 2022;
published online May 18, 2022

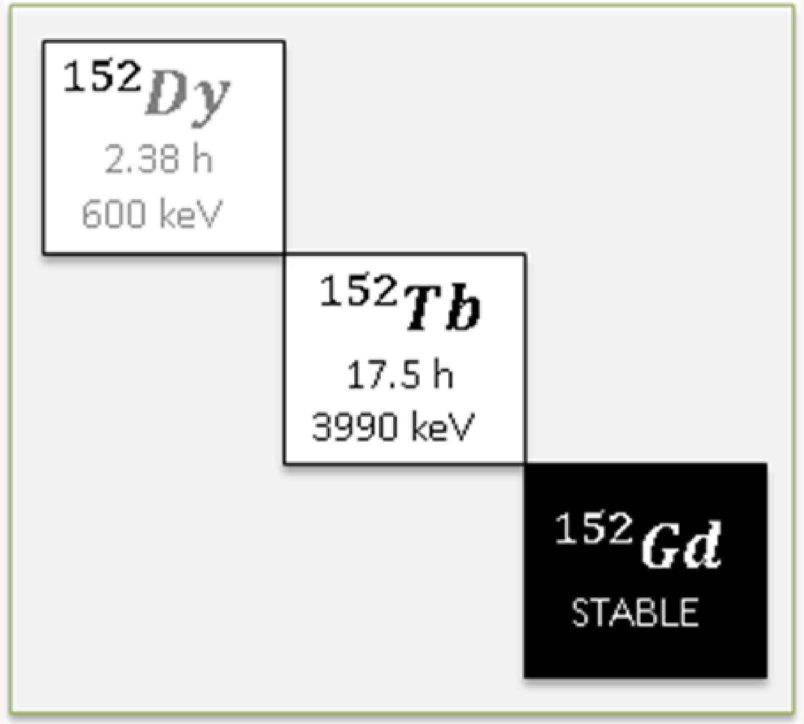
adequate quantification of the required decay data (i.e., dose calculations include half-lives, energies and emission probabilities of α, β^+, various electron particles, γ and X-rays,

Nichols, A. L. Radiochimica Acta, 110(6-9), 609-644.(2022).

Isotope of interest

^{152}Tb is a promising candidate to be used for PET imaging based on the emission of β^{+}-particles

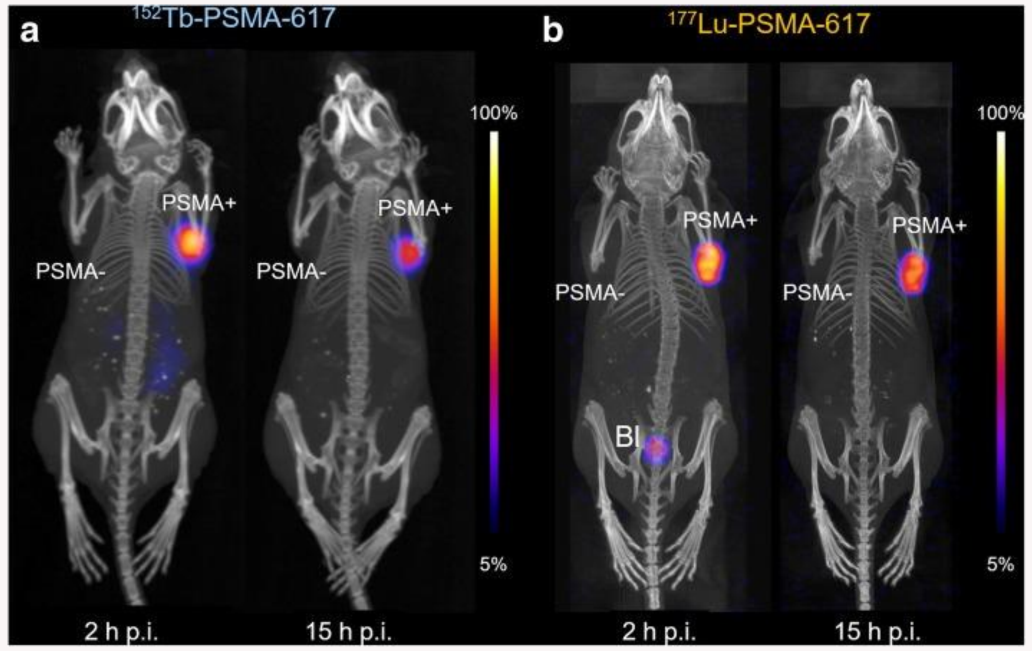
Thera/peutic + diag/**nostic** **--->Theranostic pair**



β+: 20.3%
EC: 79.7%

Extremely extensive and complex decay scheme.

^{161}Tb ^{152}Tb



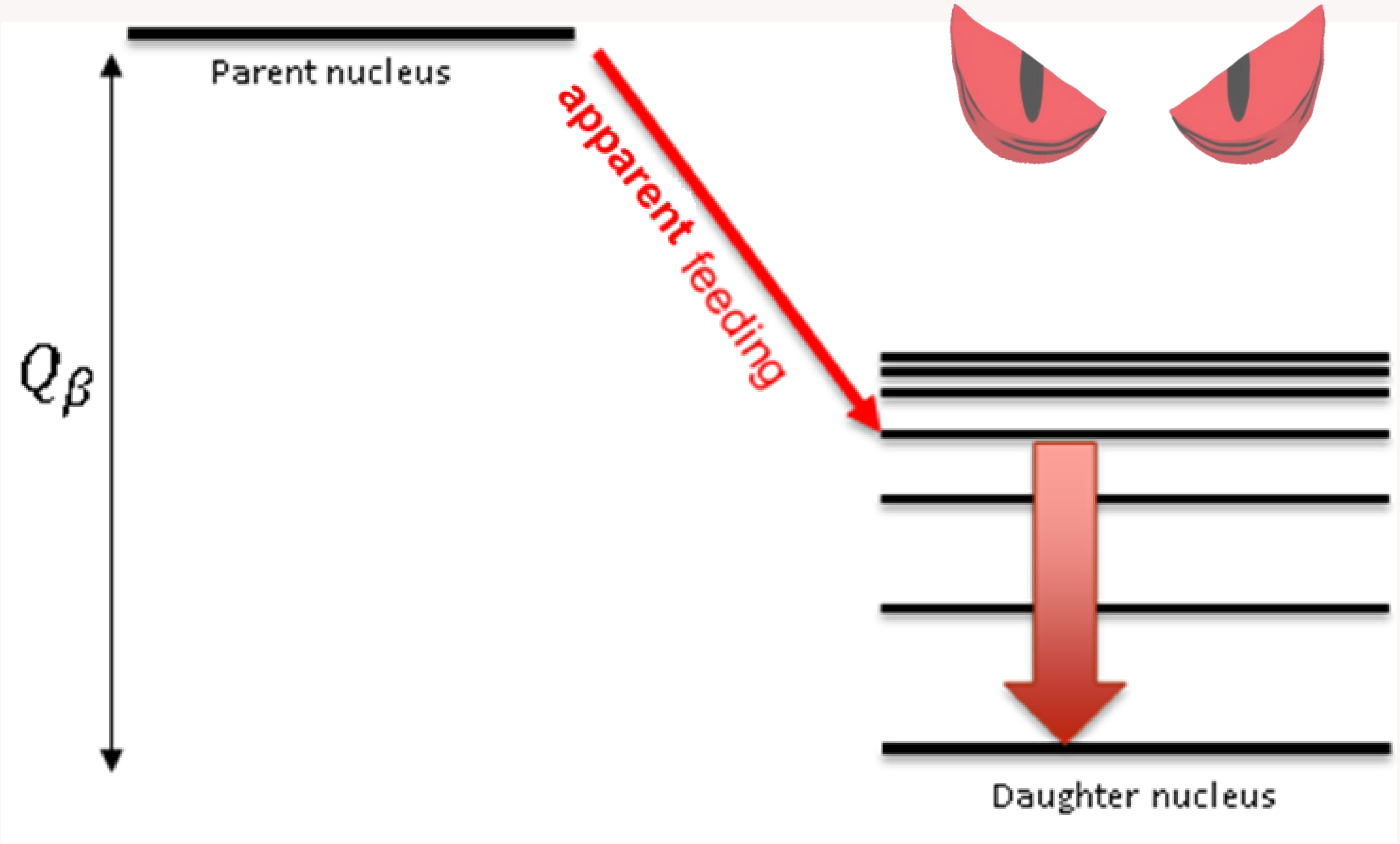
What do we want to measure?

- It is important to determine accurately the beta intensity distribution $I_\beta(E)$.
- The total EC/β^+ ratio. (Especially relevant for PET isotopes)
- The ratio between energy per decay in the form of γ-rays and the energy per decay in the form of β particles. (Relevant for all isotopes used in nuclear medicine)

Very often the only information available about the beta intensity has been measured with high resolution (HR) gamma detectors.

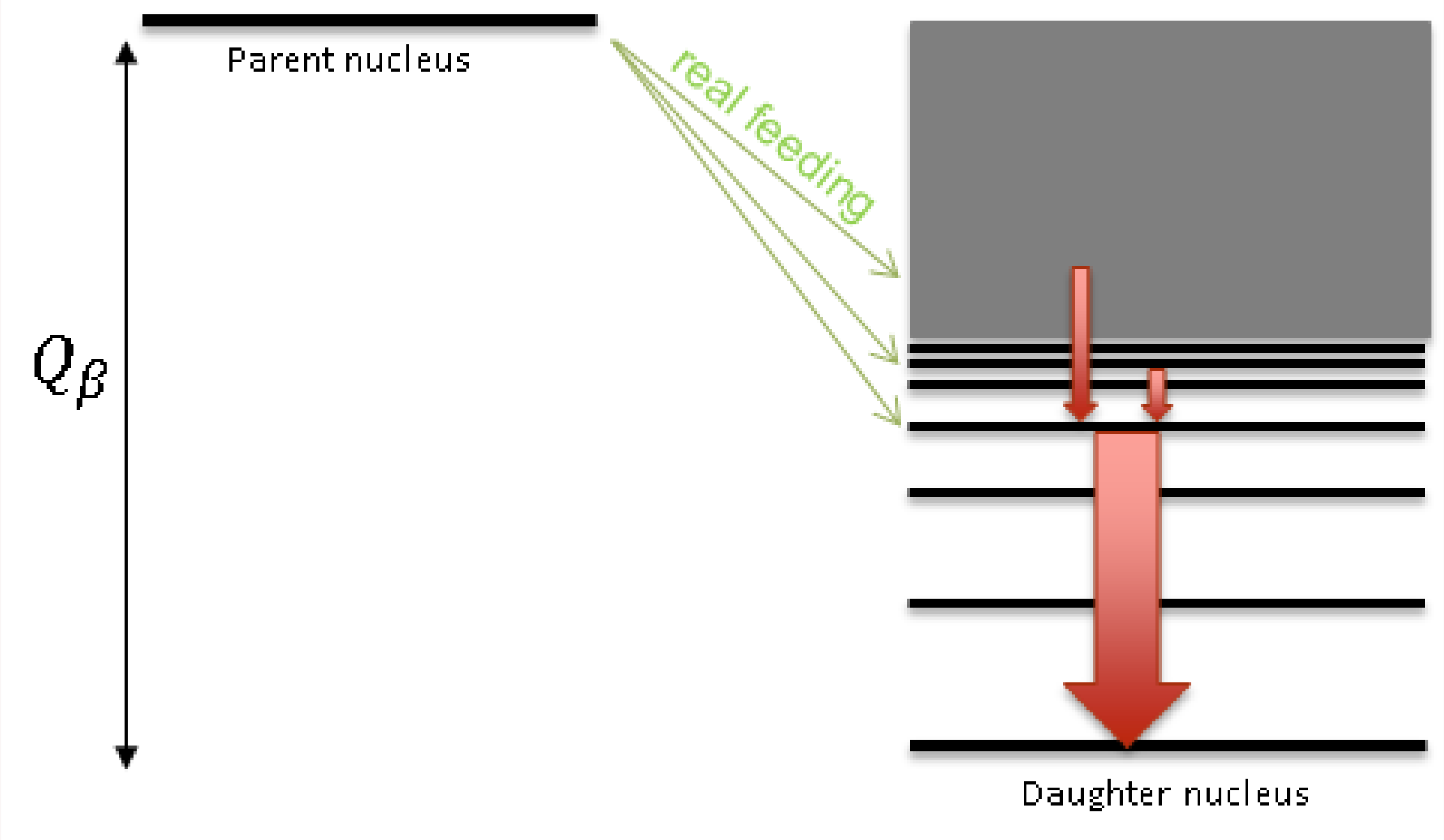
Why don't we just measure it with Ge detectors (e. g. IDS)?

Pandemonium Effect



Decays with large Q_β values may lead to populate in a high density of levels close to Q-value in the daughter nucleus.

High resolution spectra may suffer from Pandemonium effect, which is the ***overestimation*** of β feeding at low energy levels and the ***underestimation*** at higher energy.

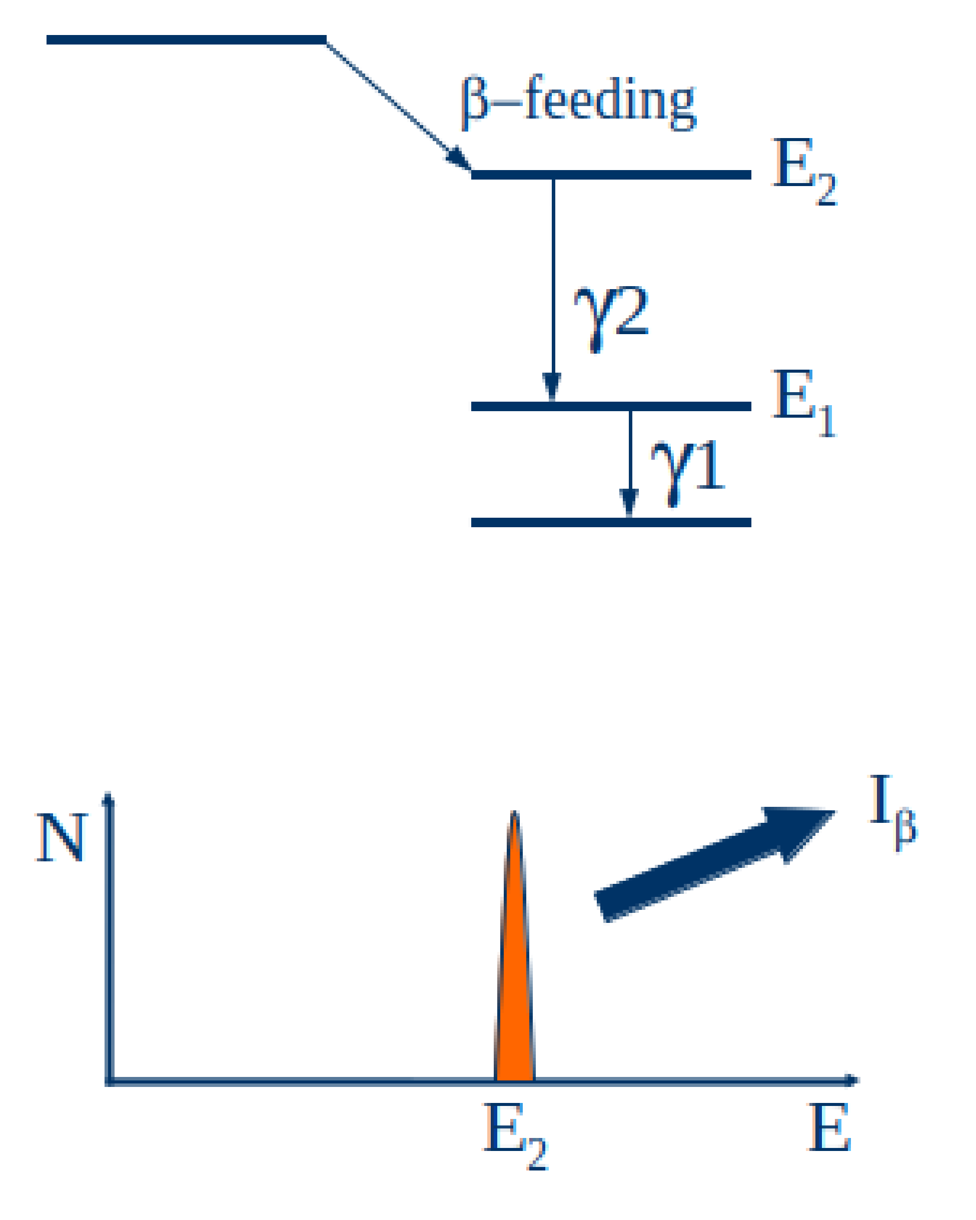


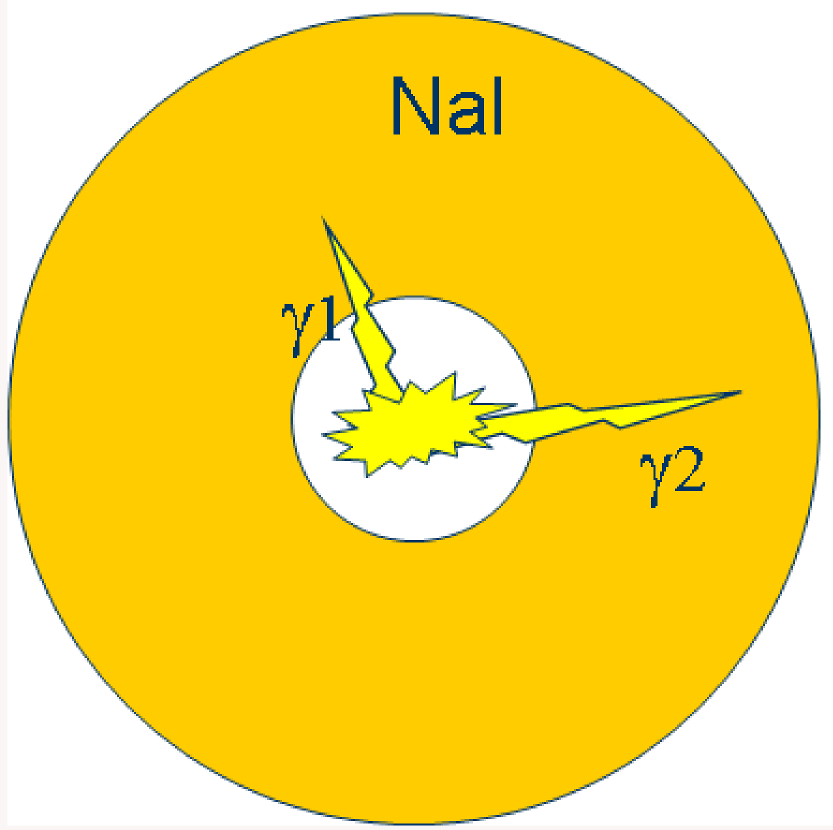
Hardy et al., Physics letters B 71. (1977) Pandemonium

The experimental technique

Total Absorption Spectroscopy (Ideal case)

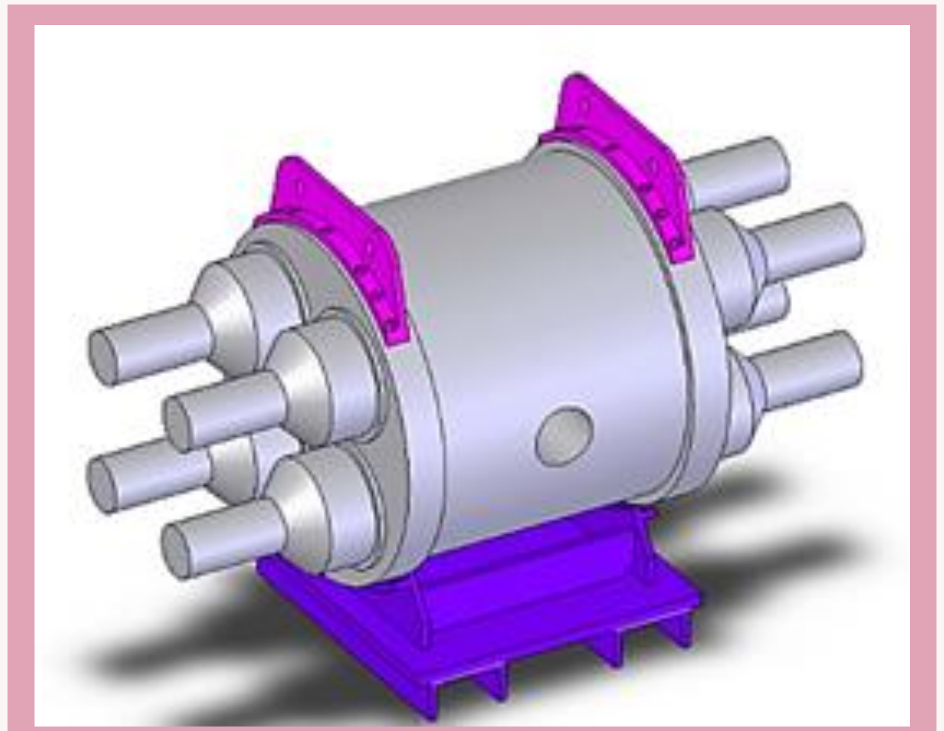
An ideal TAS is a detector with an ideal 100% efficiency (An Electromagnetic calorimeter), registers the total deposited energy in each individual event from any gamma or beta decay. Therefore, decay's information can be extracted from the total sum spectrum.

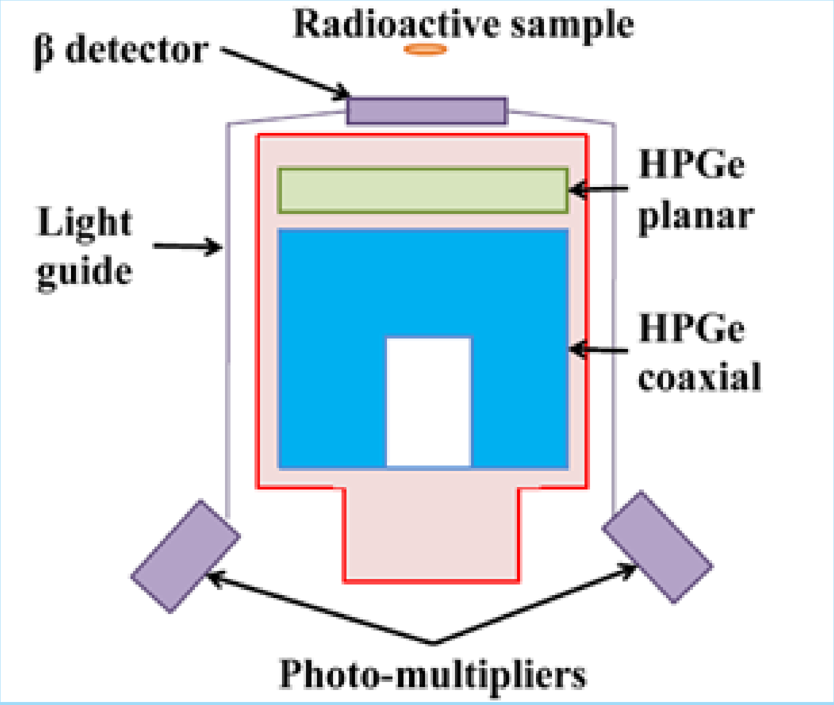


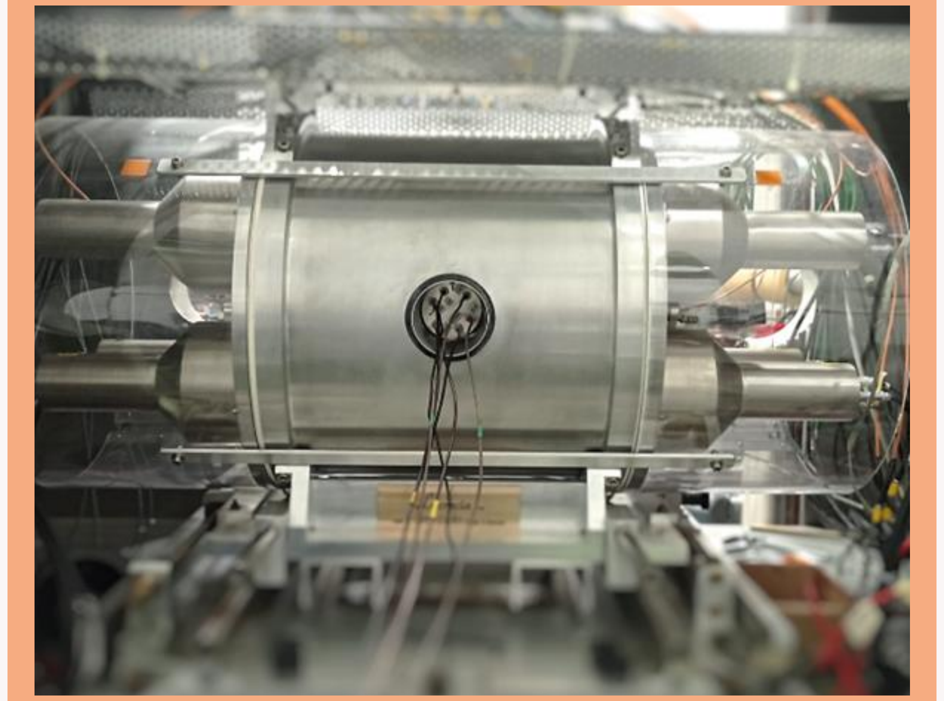


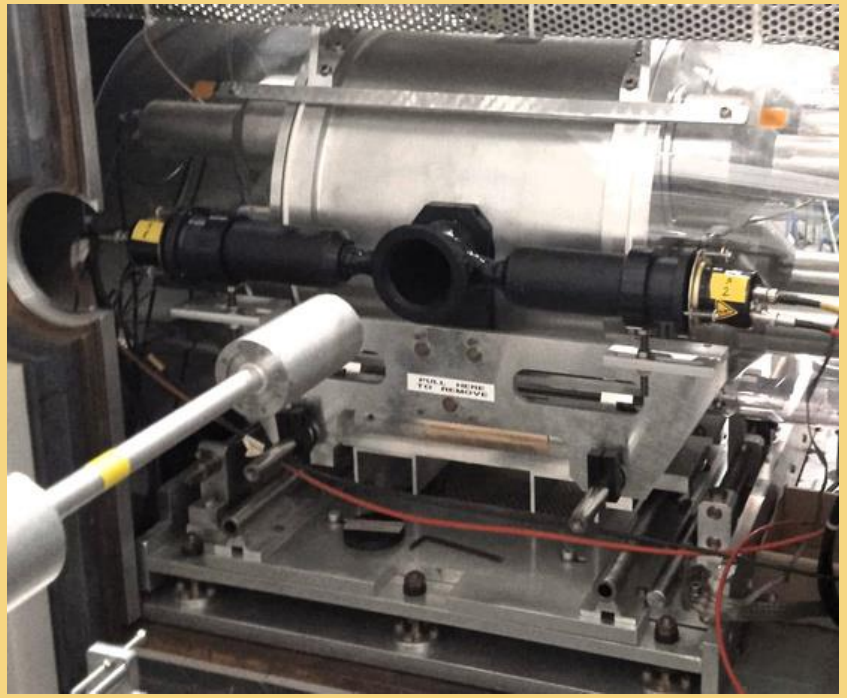
Development of TAS analysis techniques: Taín et al., NIM A571 (2007)

Lucrecia Detector









TAS

- NaI(Tl) Crystal Scintillator.
- Diameter 38 cm x length 38 cm.
- Hole is used for isotope implantation and the installation of ancillary detectors.

Ancillary Detectors

- Plastic beta detector
- HPGe telescope (Coaxial and planar)

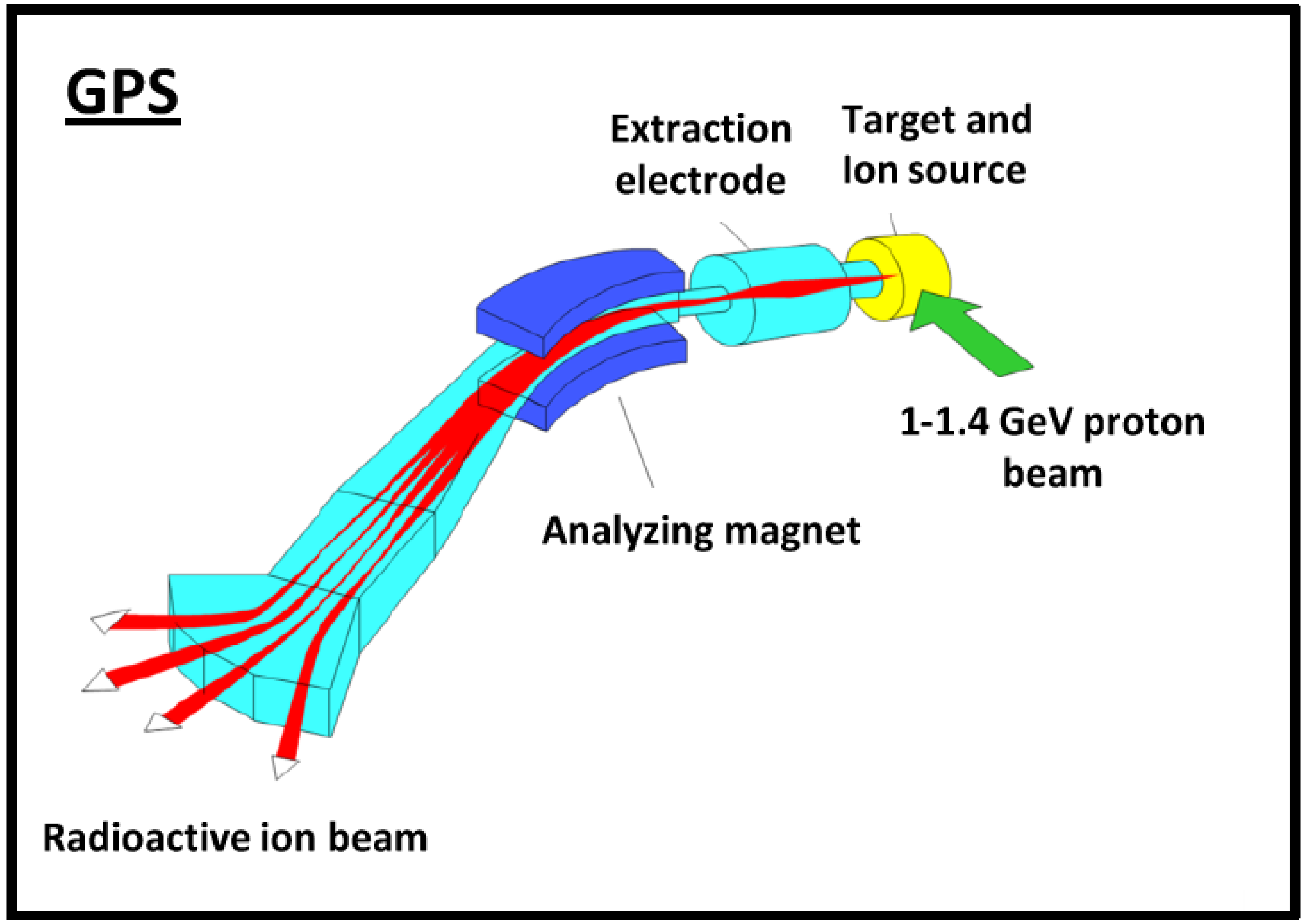
B Rubio et al 2017 J. Phys. G: Nucl. Part. Phys. 44 084004

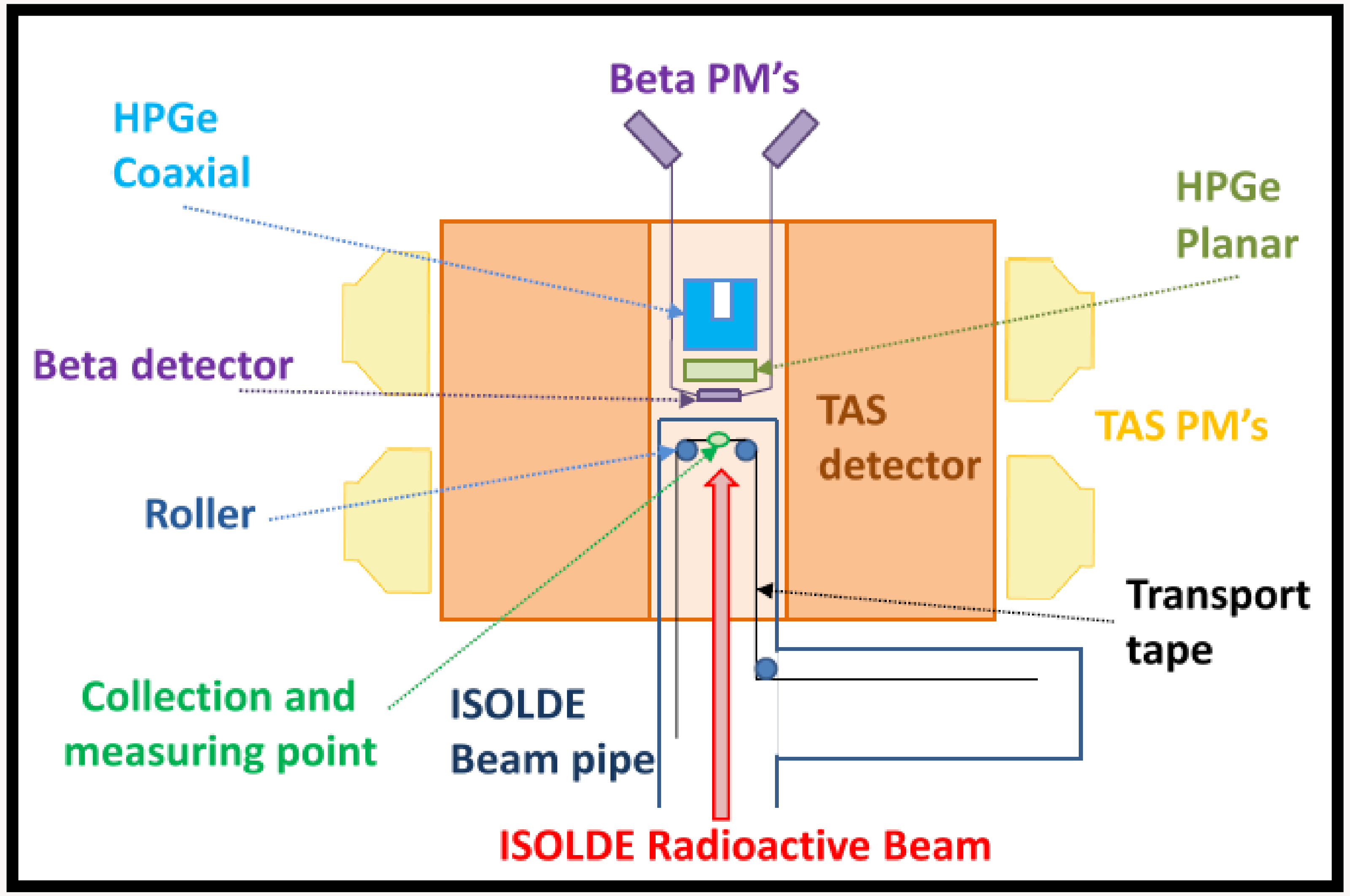
The same simples were brought to ILL(Grenoble) and mesaured at the FIPPS Ge Array

GLM beam line

thanks to Ulli Köster!

Radioactive isotopes can be obtained from an **Isotope Separation On-Line DEvice** (ISOLDE).



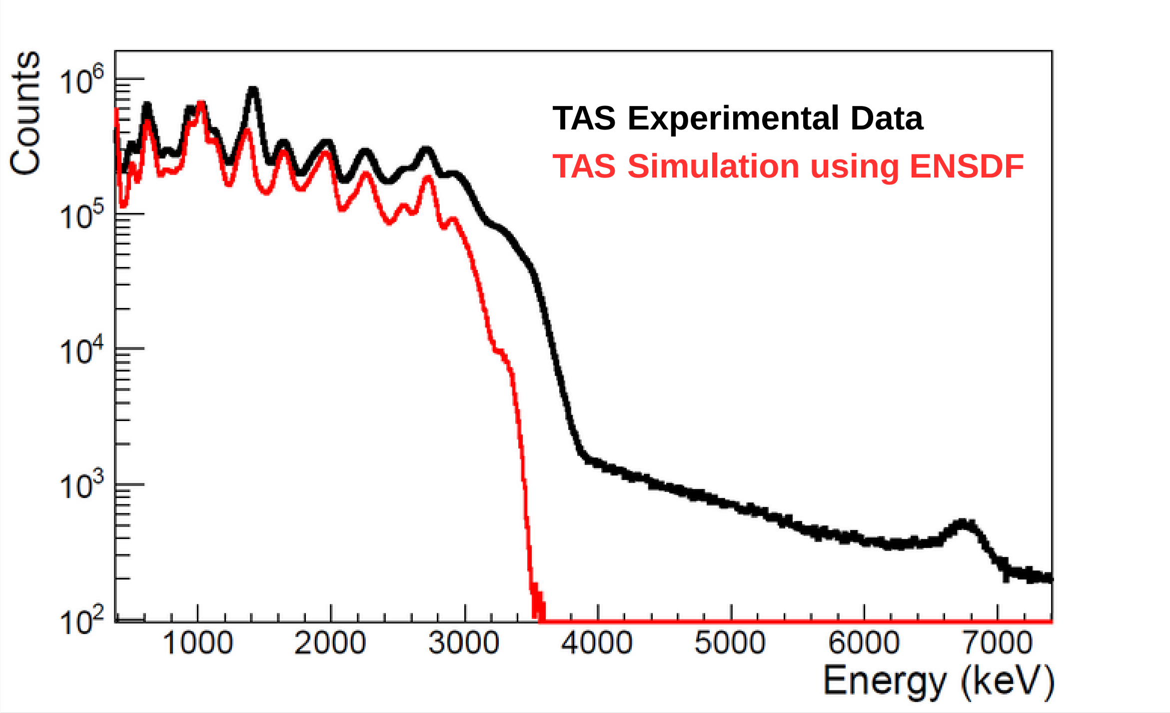


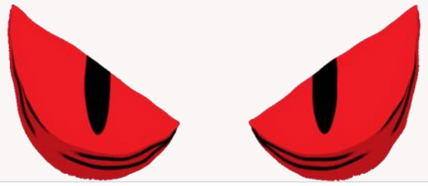
The radioisotopes of interest are extracted and implanted in a magnetic tape inside the TAS detector.

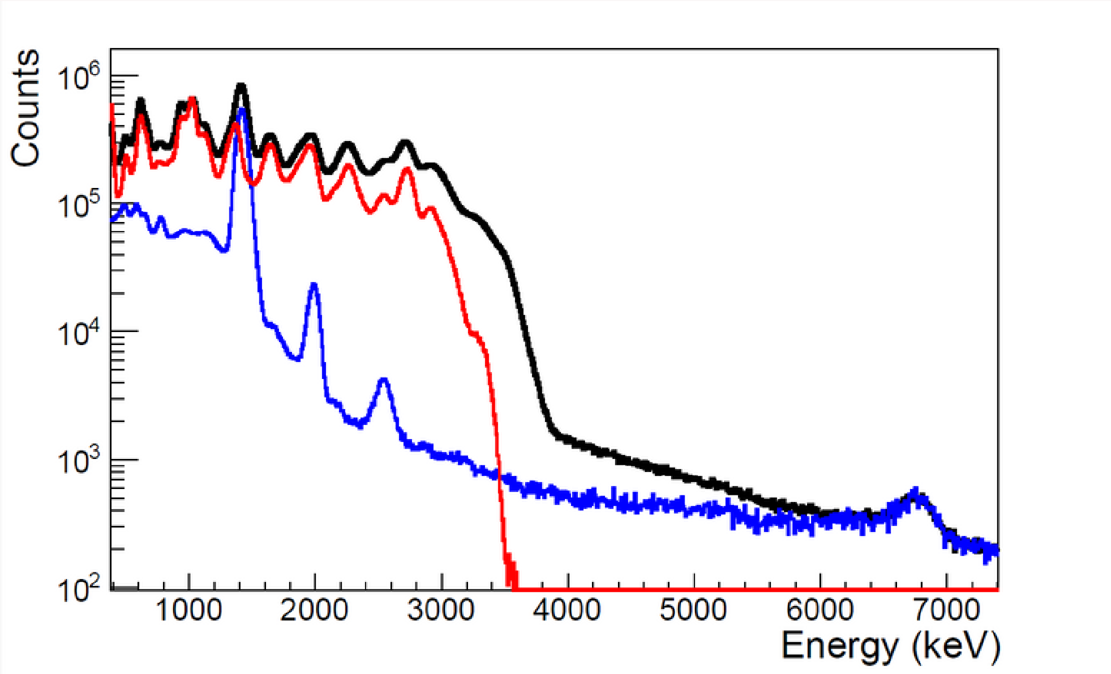
PRELIMINARY ANALYSIS

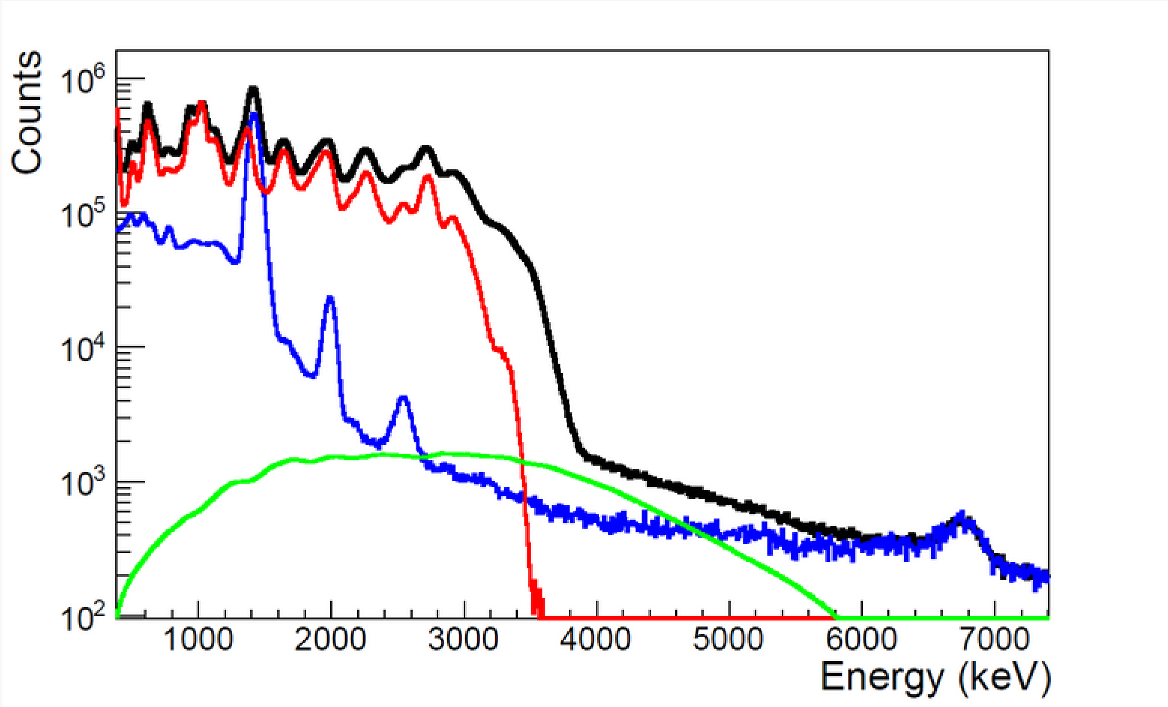
Preliminary Analysis

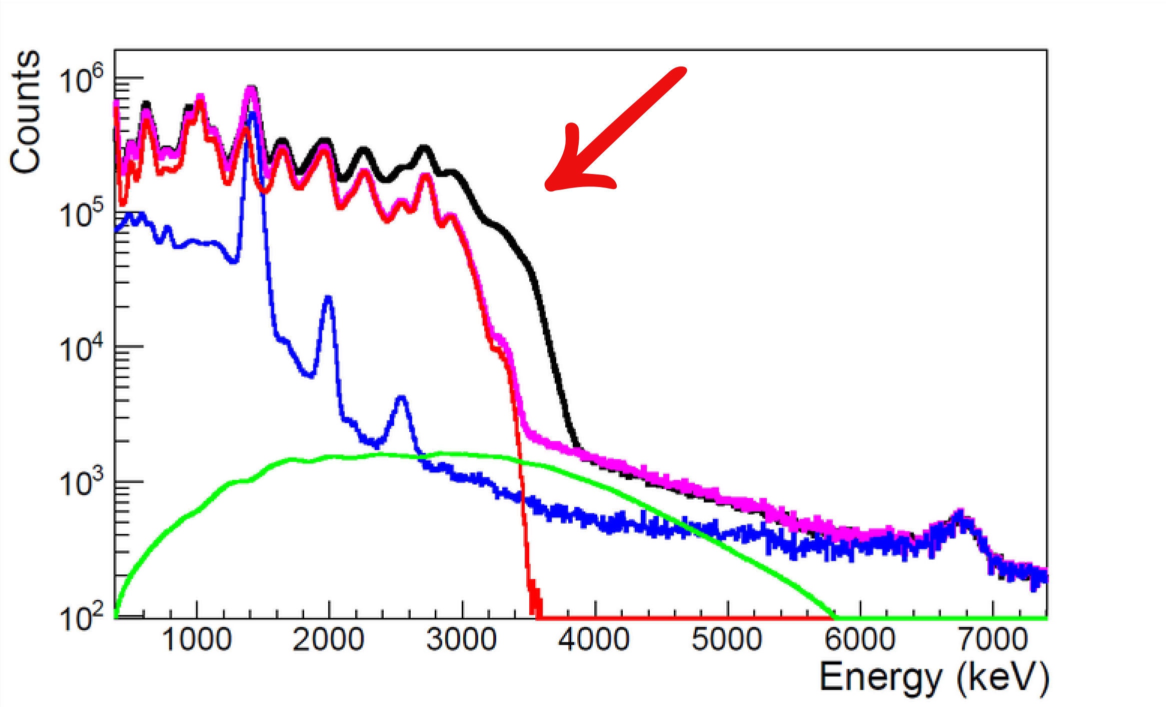
^{152}Tb







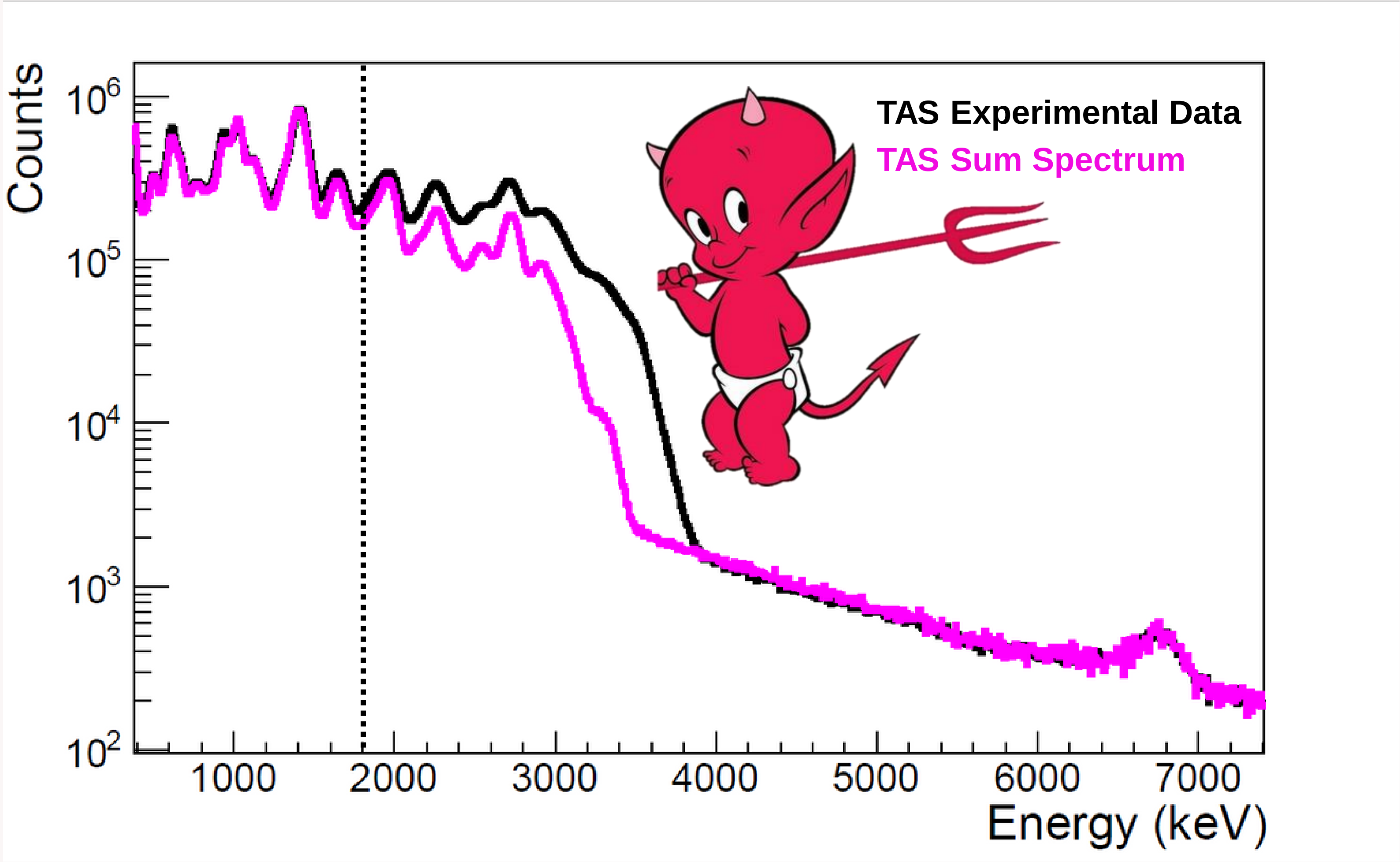




Sum Spectrum = Simulation + Background + Pile-up

Preliminary Analysis

^{152}Tb



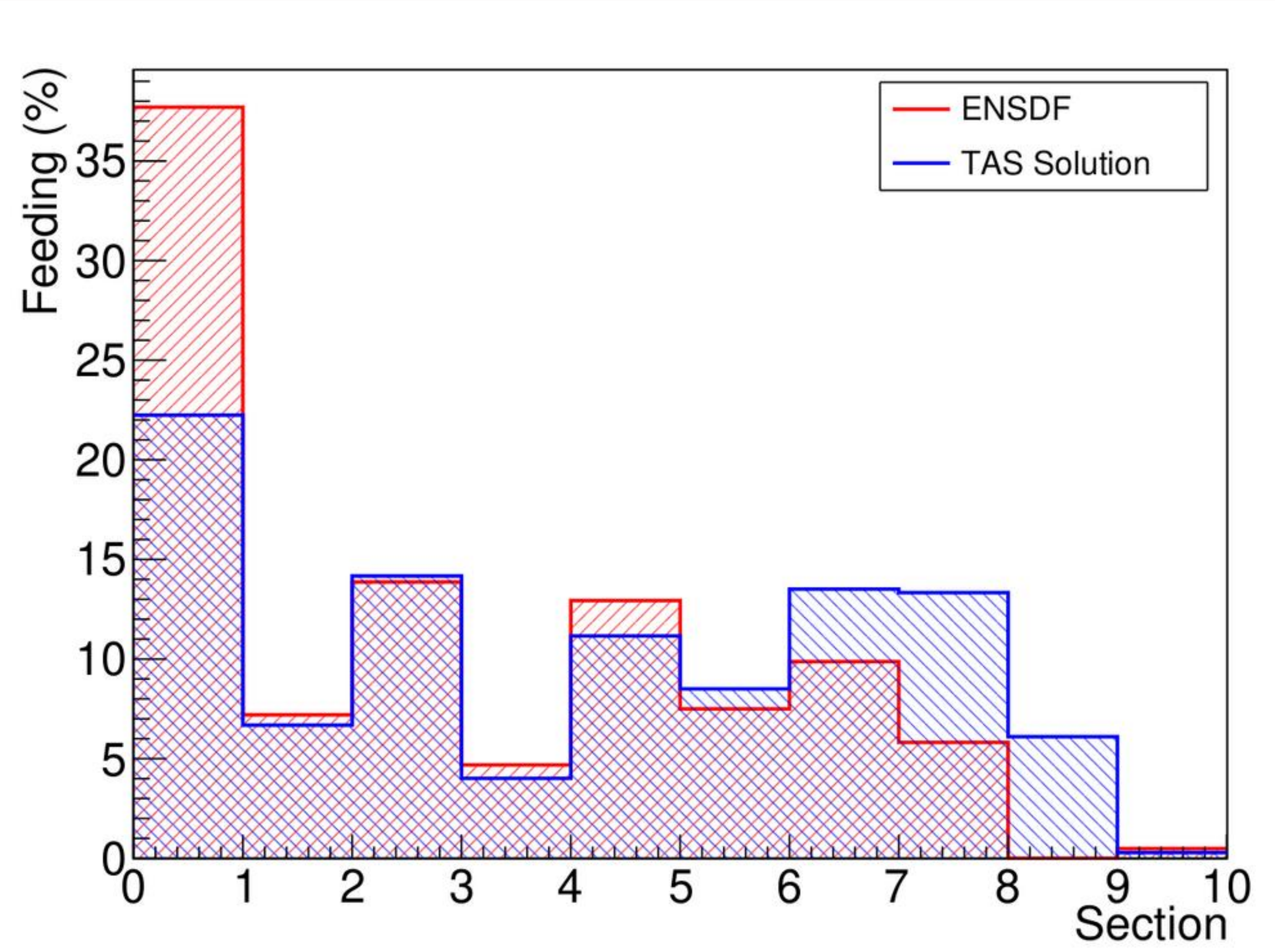
When everything in the gamma spectrum is well understood then is time for the **unfolding algorithm (EM)**

» Distribution of probability of the beta intensity

RESULTS

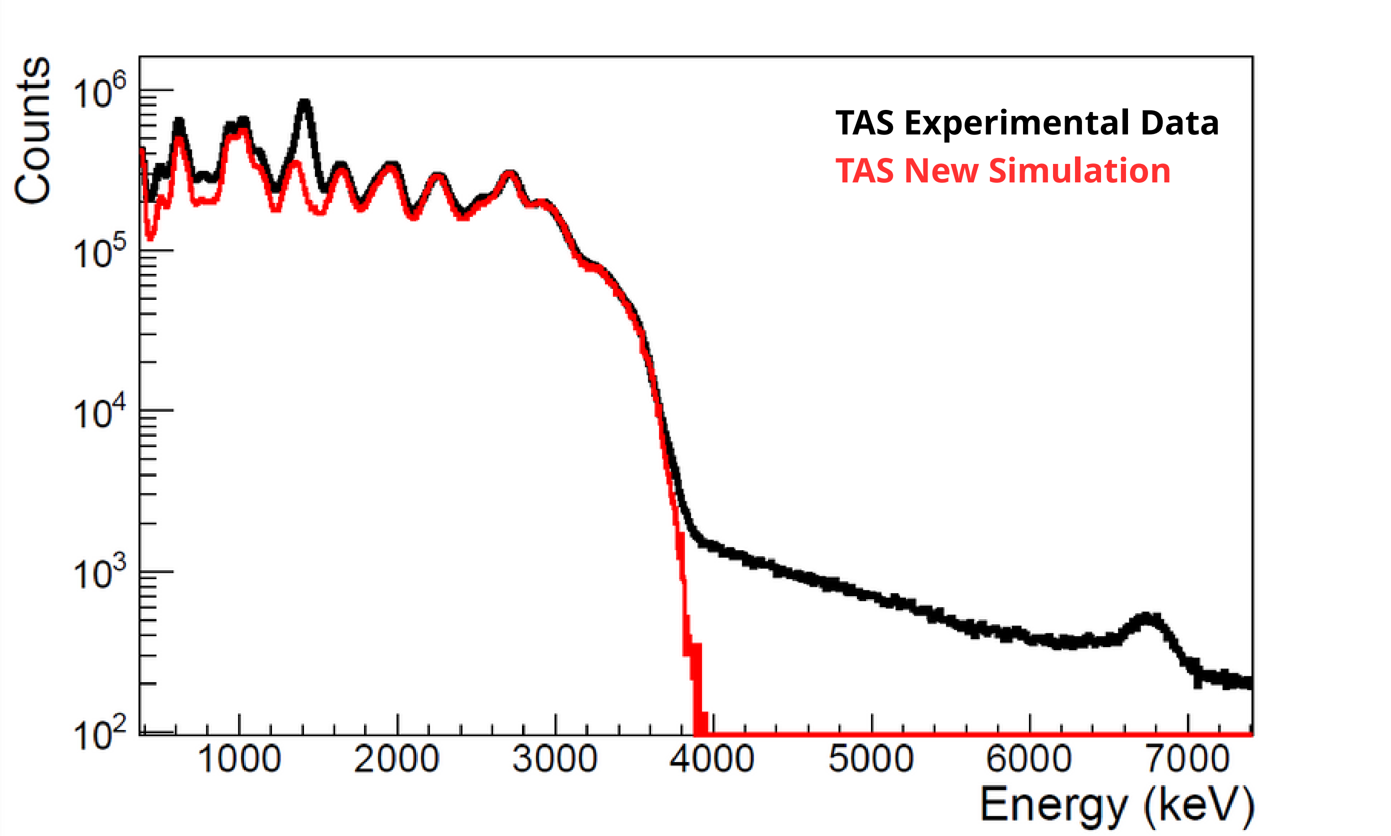
Results ^{152}Tb

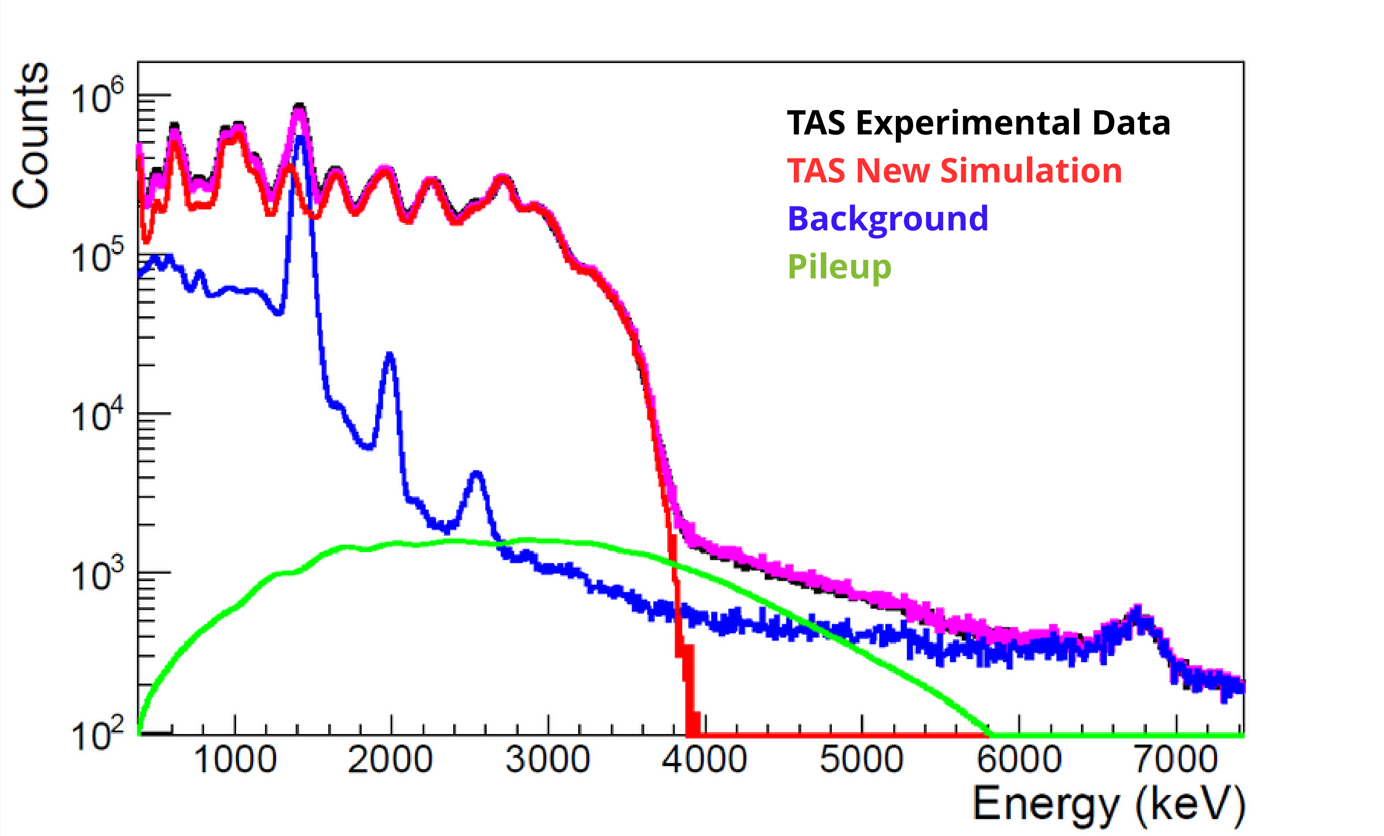
Section	Energy (keV)	ENSDF β Feeding (%)	TAS Solution β Feeding (%)
1	0 – 399	37.7	22.24
2	400 – 799	7.20	6.68
3	800 – 1199	13.87	14.17
4	1200 - 1599	4.69	4.01
5	1600 – 1999	12.93	11.16
6	2000 – 2399	7.50	8.50
7	2400 – 2799	9.87	13.52
8	2800 – 3199	5.81	12.64
9	3200 – 3599	0.00	6.56
10	3600 – 3990	0.49	0.52

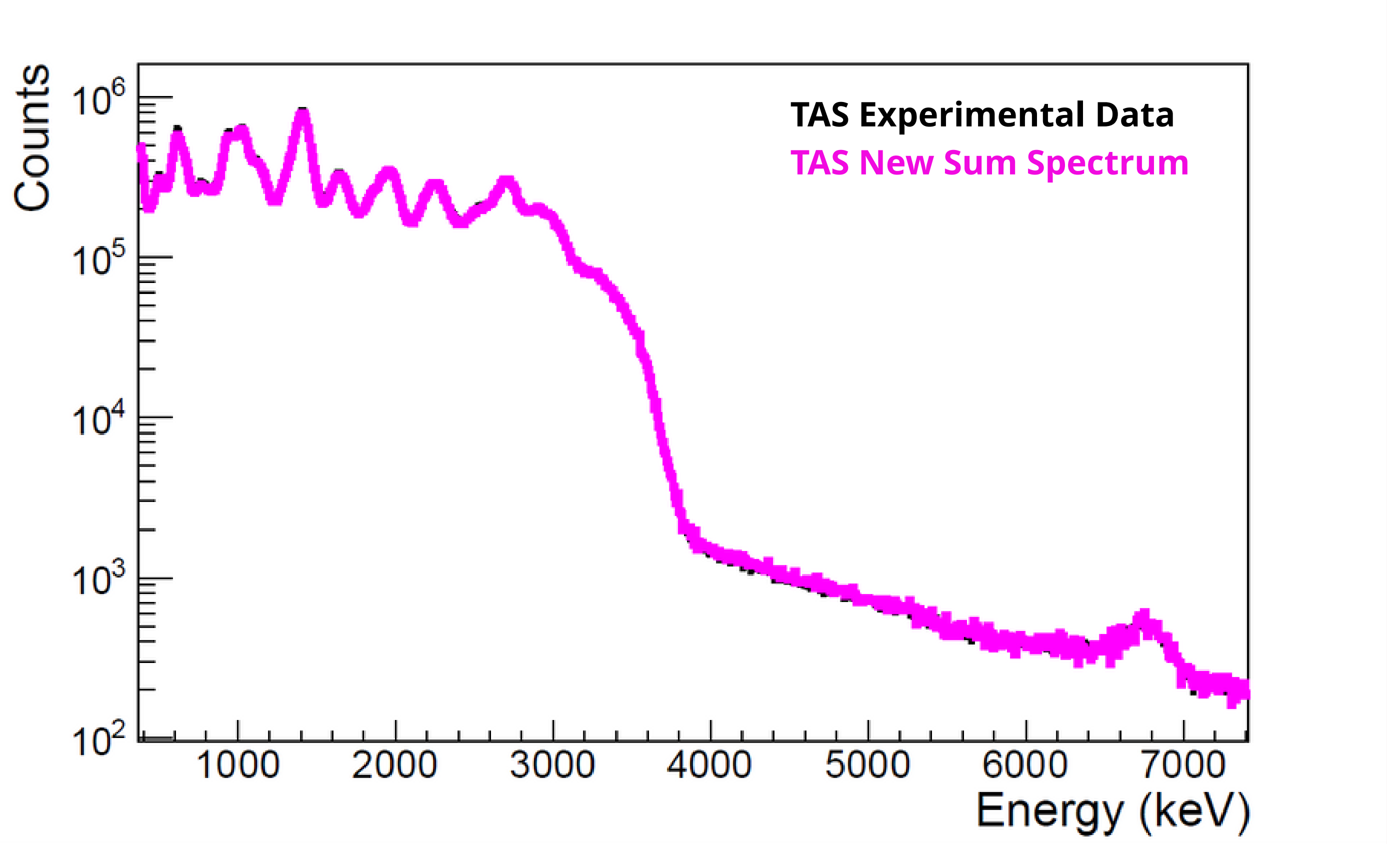


Finally, with the **new feeding** obtained by the unfolding algorithm, we can recalculate a new gamma spectrum and compare it with the **TAS** data.

Results ^{152}Tb

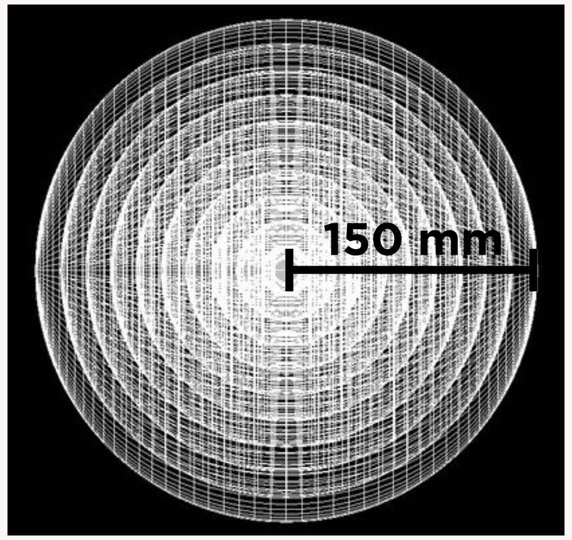






Results ^{152}Tb

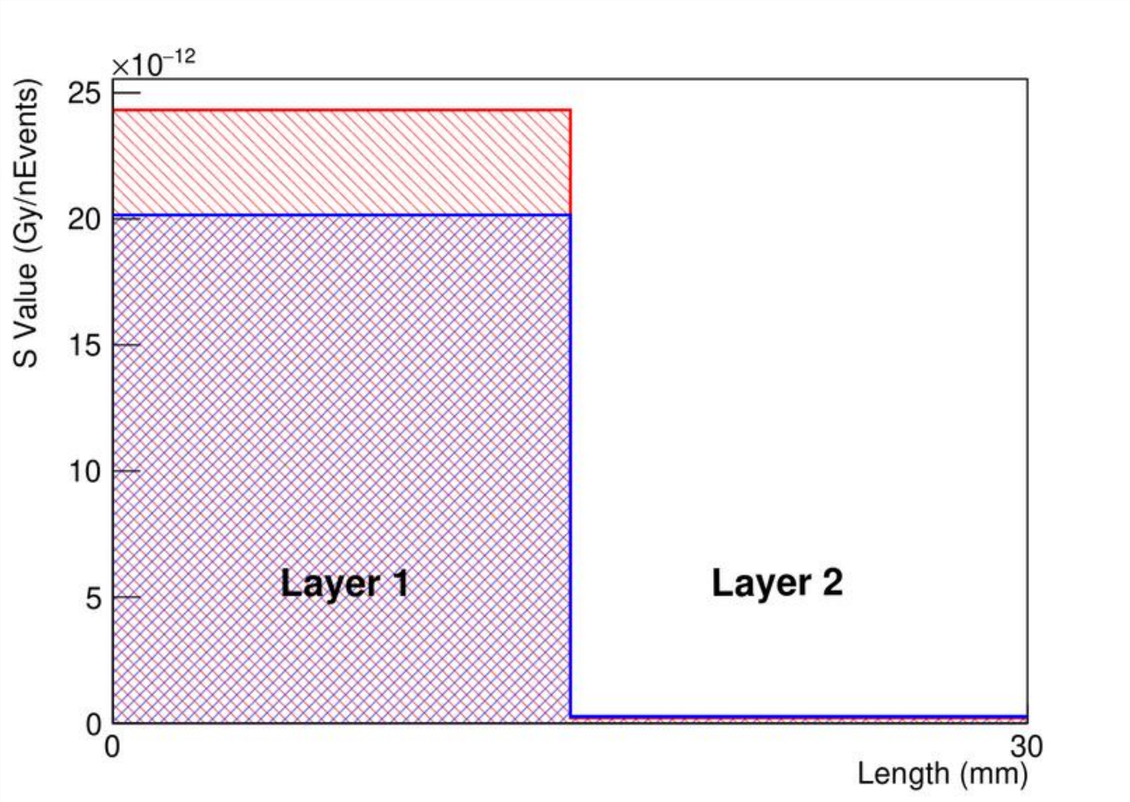
Simulation Dose

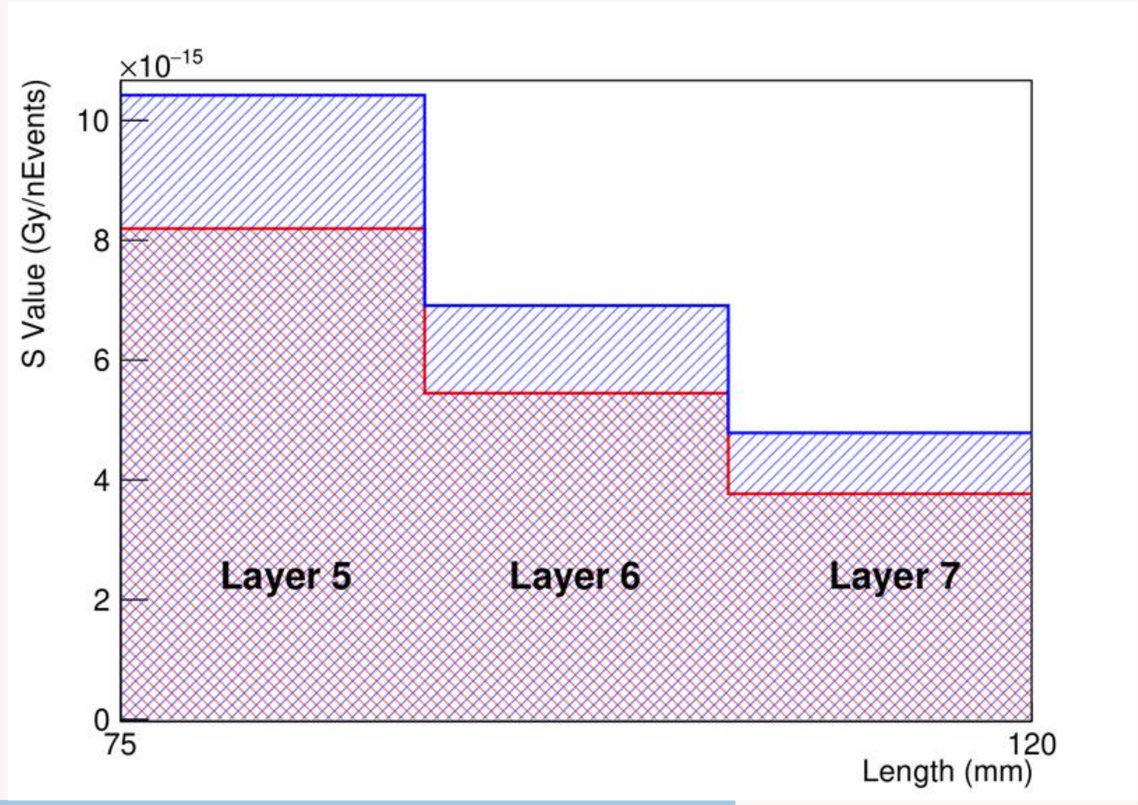


- 10 concentric spheres of water
- Source 152Tb

Simulation ENSDF Feeding
Simulation new Feeding TAS solution

Layer	S value (Gy/Bq^{-1} s^{-1})	S value (Gy/Bq^{-1} s^{-1})	Difference (%)
1	3.17e-12	2.61e-12	17,55
2	1.03e-13	1.30e-13	-26,63
3	3.72e-14	4.73e-14	-27,22
4	1.87e-14	2.36e-14	-26,15
5	1.09e-14	1.39e-14	-26,91
6	7.07e-15	8.98e-15	-27,13
7	4.85e-15	6.14e-15	-26,69
8	3.41e-15	4.35e-15	-27,61
9	2.47e-15	3.18e-15	-28,78
10	1.79e-15	2.32e-15	-29,03





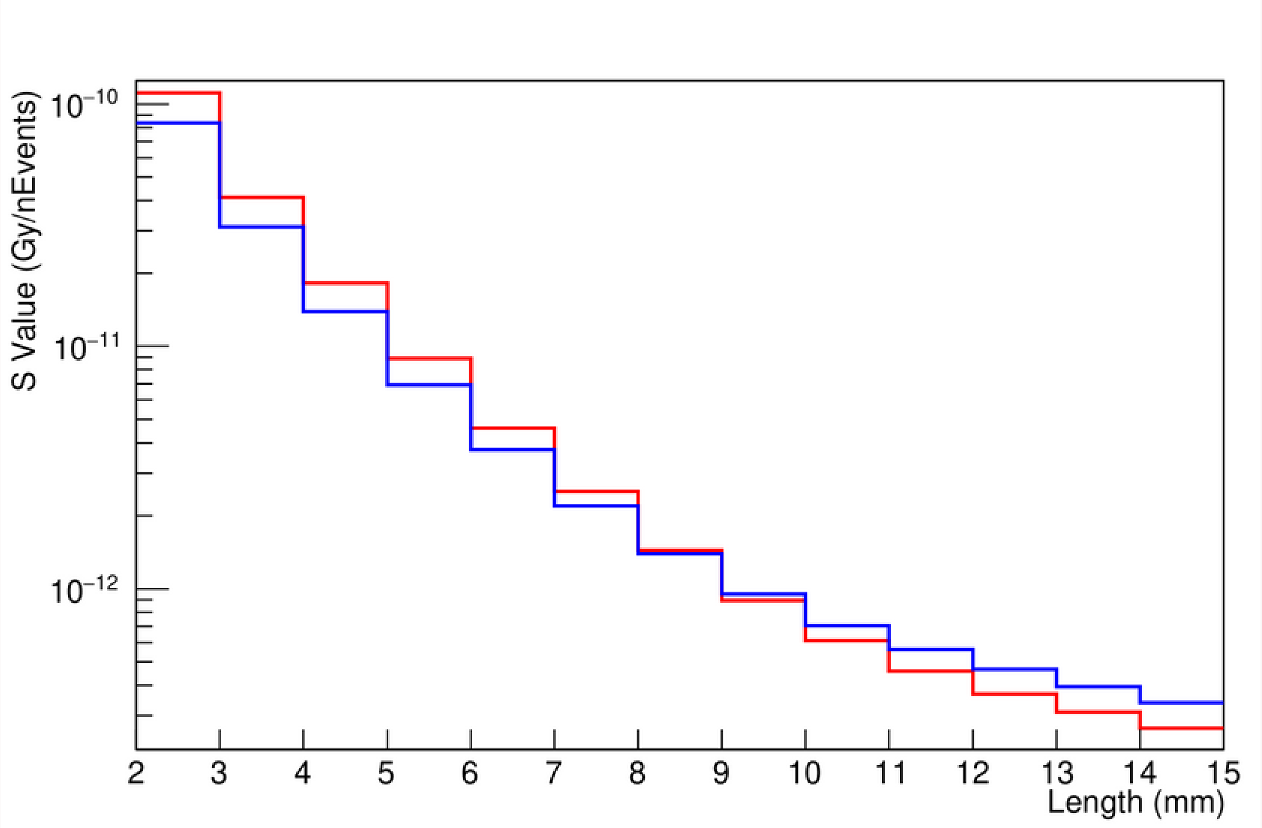
Results ^{152}Tb

Simulation Dose

Simulation ENSDF Feeding
Simulation new Feeding TAS solution

mm	S value (Gy/Bq^{-1} s^{-1})	S value (Gy/Bq^{-1} s^{-1})	Difference (%)
1	1.90e-08	1.58e-08	16.85
2	5.11e-10	3.75e-10	26.73
3	1.11e-10	8.36e-11	24.88
4	4.12e-11	3.11e-11	24.48
5	1.82e-11	1.39e-11	23.63
6	8.90e-12	6.92e-12	22.34
7	4.61e-12	3.75e-12	18.58
8	2.52e-12	2.20e-12	12.71
9	1.44e-12	1.40e-12	3.20
10	8.95e-13	9.51e-13	-6.25
11	6.12e-13	7.05e-13	-15.27
12	4.57e-13	5.62e-13	-22.99
13	3.68e-13	4.65e-13	-26.43
14	3.10e-13	3.94e-13	-27.19
15	2.66e-13	3.39e-13	-27.51

- Water sphere
- Radio 15 mm
- Source 152Tb



Number of positrons per decay:

ENDSF = 0.20

TAS = 0.14

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Summary

- Medium mass and heavy nuclei: one never knows whether there is Pandemonium...We need to measure them!! Especially cases with particular sensitivity for structure/astro, reactor or medicine.

- In order to calculate the correct dosage, it is fundamental to know the deficiencies in the nuclear data, more specifically the distribution of probability of the beta intensity

- We observed the presence of the pandemonium effect in 152Tb. In the simulation made with the data reported by the ENSDF there are discrepancies in the intensity in energies between 2 MeV and the Q value.

- The deconvolution algorithm corrected these deficiencies, adjusting feeding at low energy levels and close to the Q_value, getting a better fit with the experimental data. These corrections show significant changes in the dose simulation, underscoring the importance of adjusting nuclear data for applications such as dosimetry.

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ISOLDE

THANKS

TO THE AUDIENCE AND TO THE ISOLDE CREW WHO HAVE ORGANIZED THIS EVENT

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Off-line TAS measurements of the long-lived nuclei ^{152,155}Tb and ^{76,77}Br for their relevance in medicine and neutrino physics

Sep - 27, 2022

E. Nácher[1], U. Köster[2], J.A. Briz[3], J. Agramunt[1], A. Algora[1], B. Rubio[1], J.L. Taín[1], W. Gelletly[4], Z. Favier[5], L.M. Fraile[3], MJG. Borge[6], V. Guadilla[7], K. Johnston[5], S. Parra[1], O. Tengblad[6], J. Vijande[1], Z. Yue[5]

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2 Institut Laue-Langevin, Grenoble, France
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4 University of Surrey, Guildford, UK
5 CERN, Geneve, Switzerland
6 Instituto de Estructura de la Materia (CSIC), Madrid, Spain
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Extra Slides

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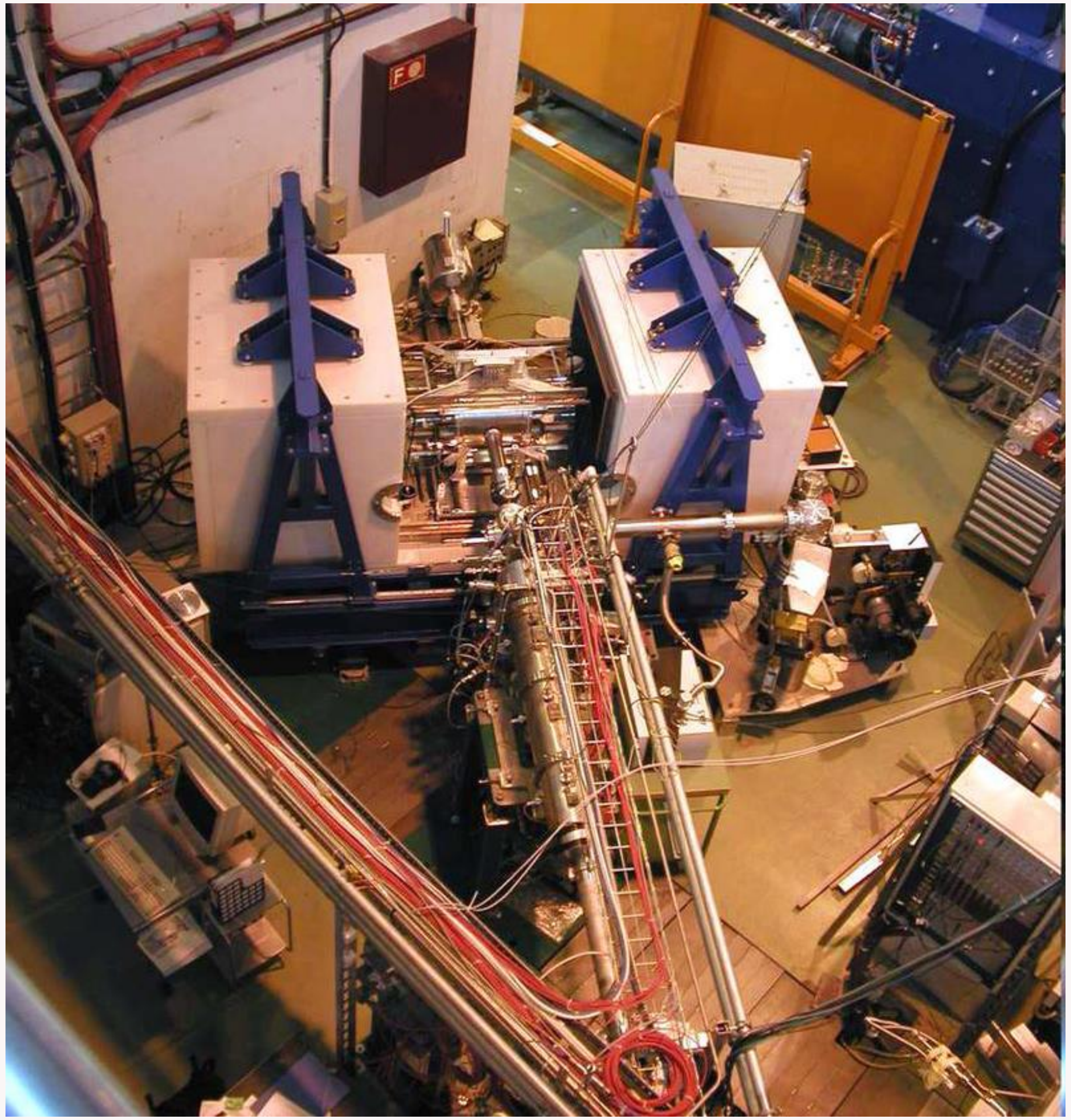
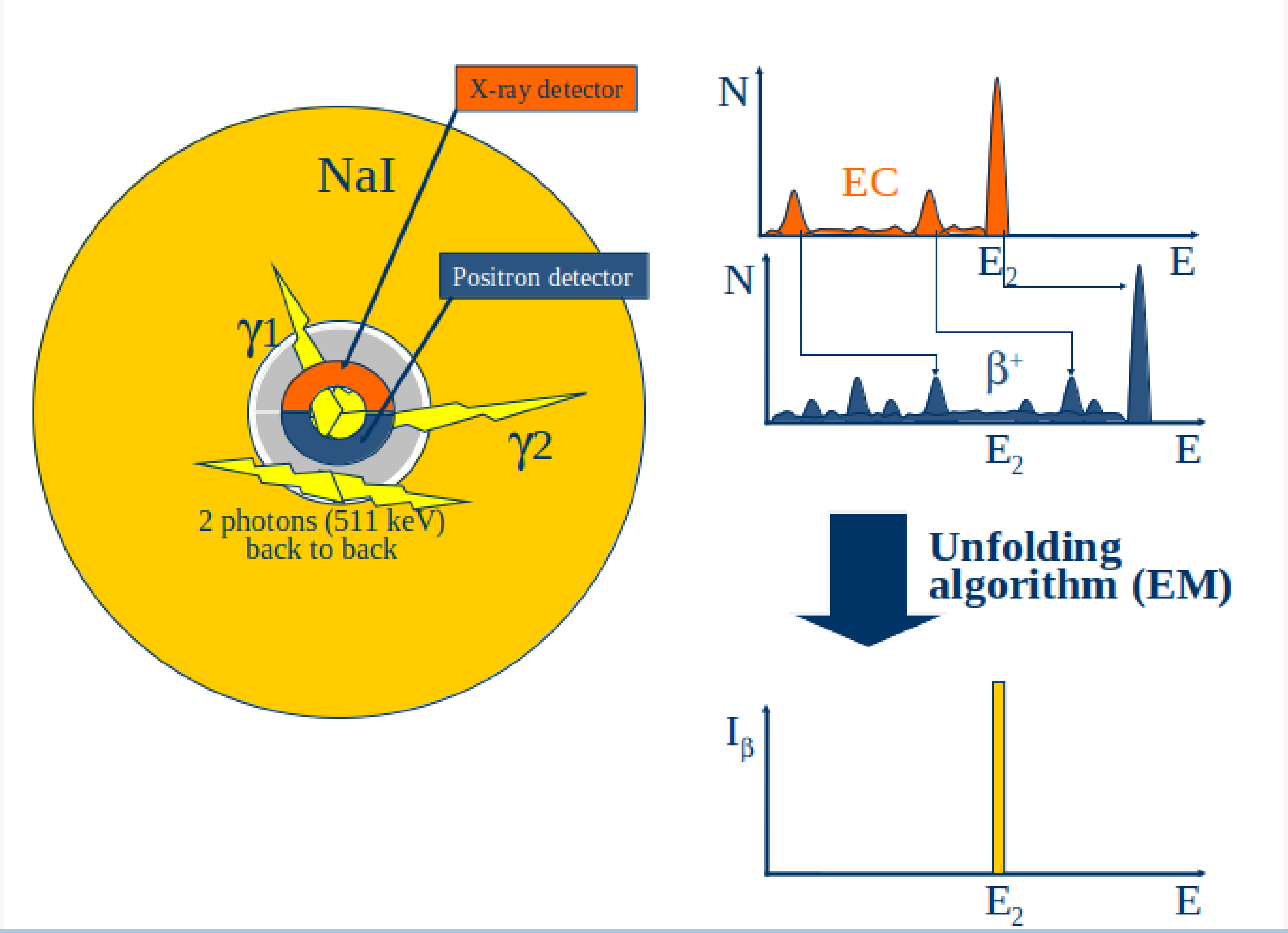
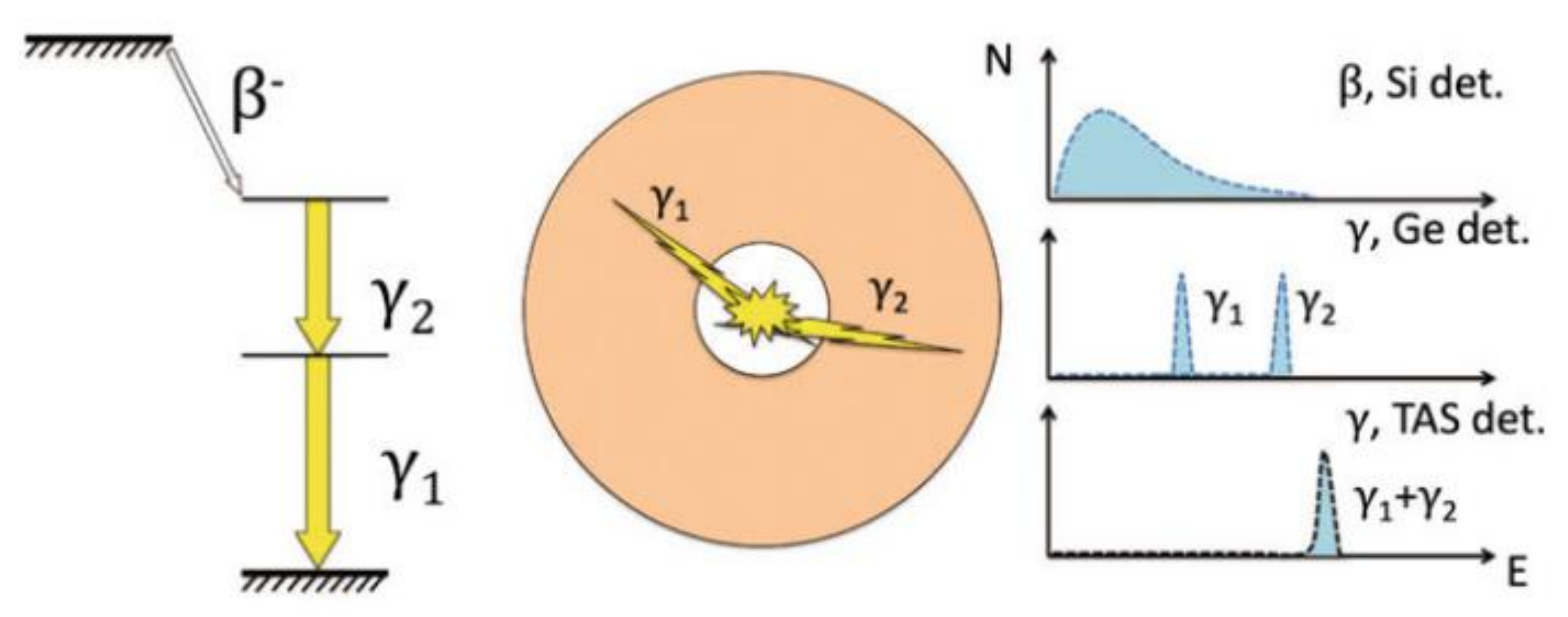


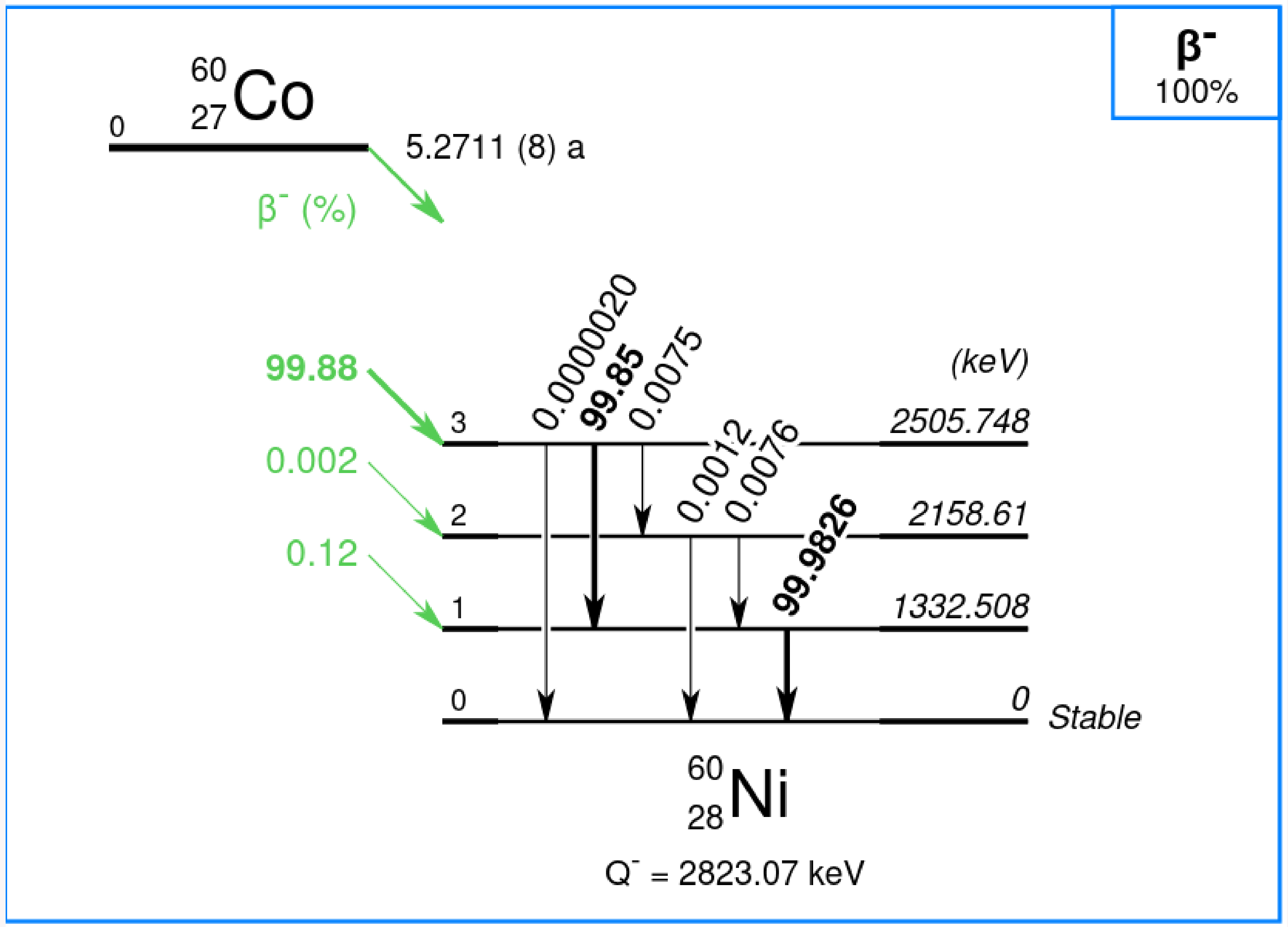
Total Absorption Spectroscopy (Ideal case)

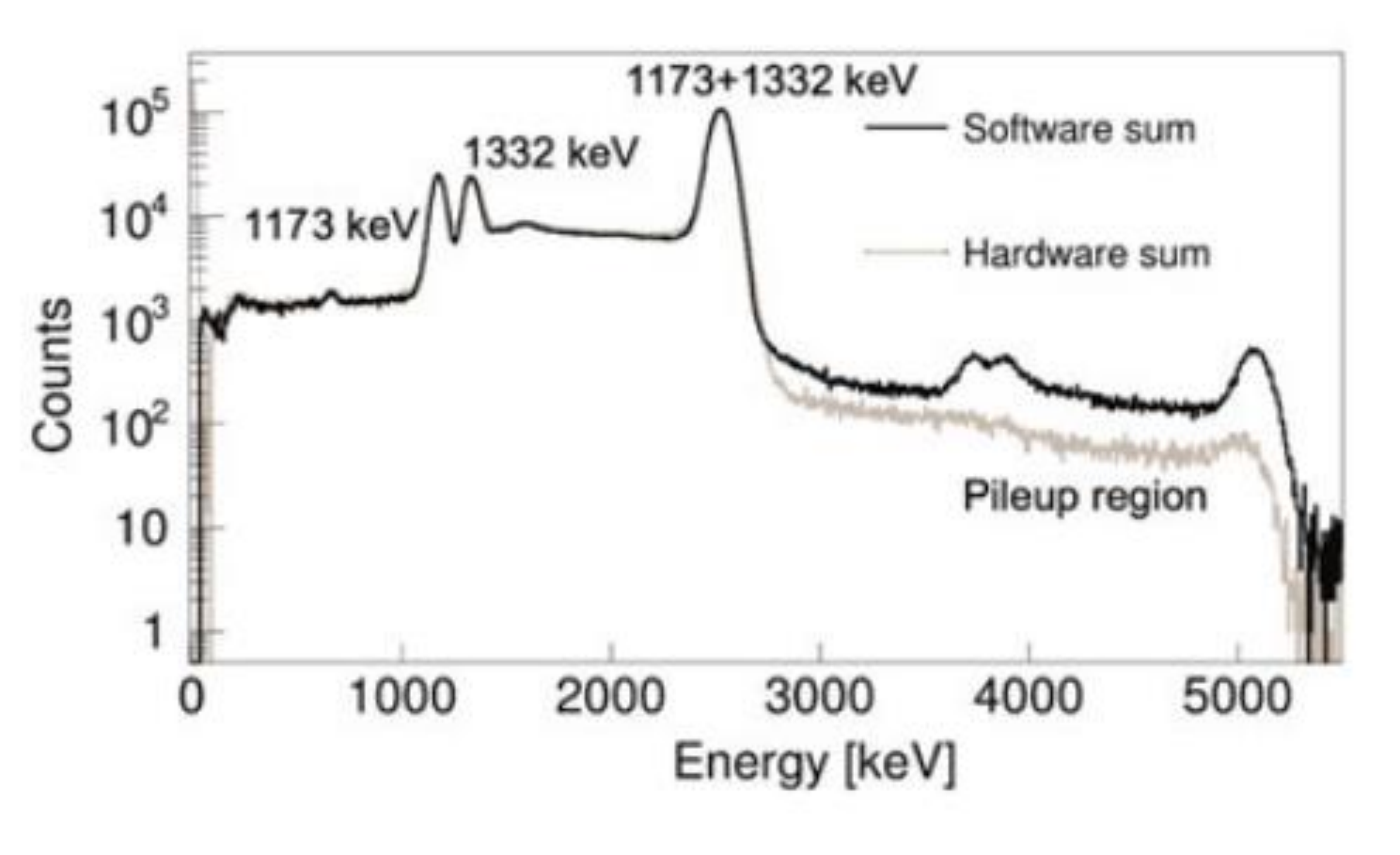


Total Absorption Spectroscopy (Ideal case)



So, with a total absorption spectrometer we do not see the individual gamma rays emitted in the decay, we see the sum of the energy of the gamma cascades that follows the beta decay.

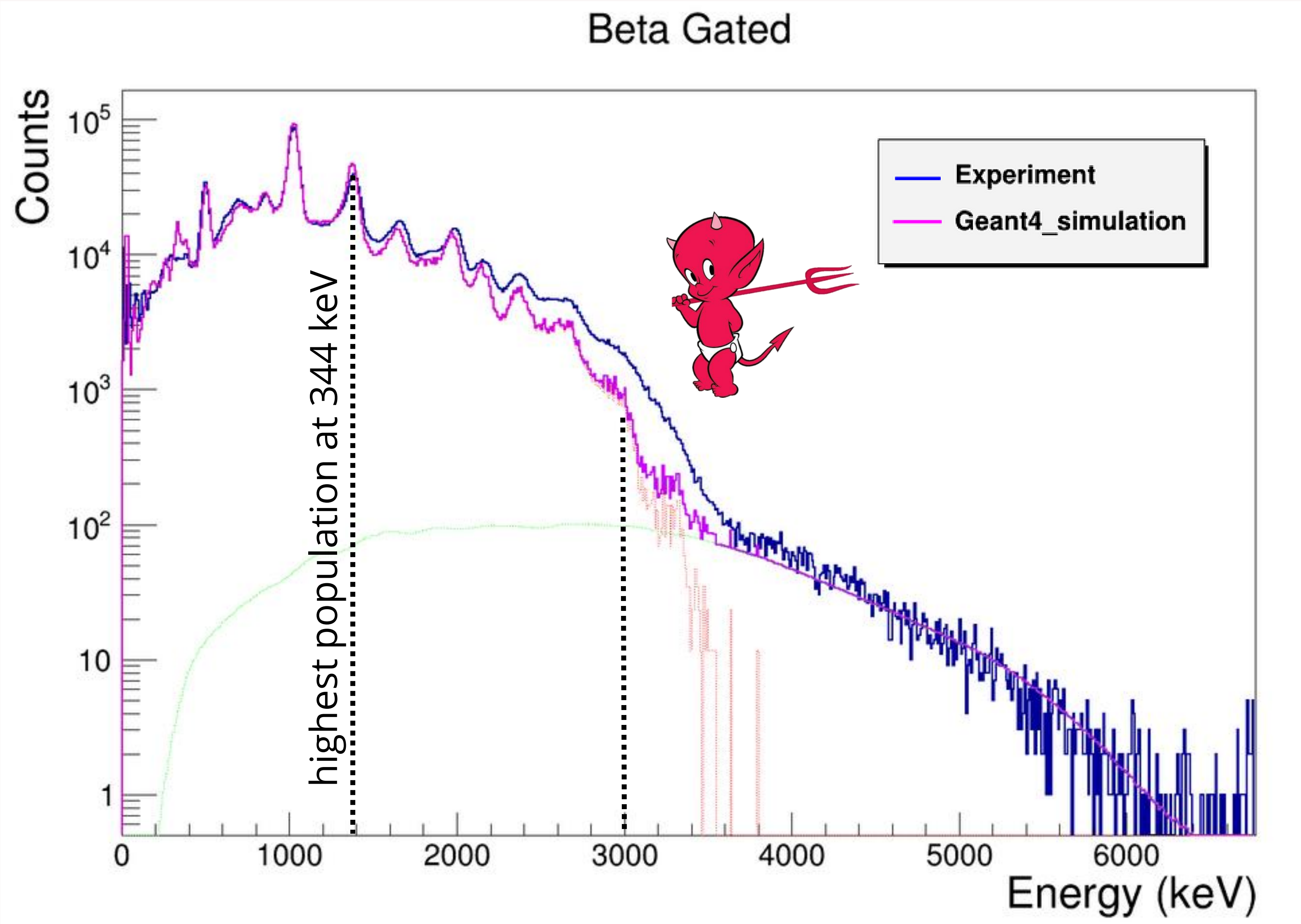

β-
100%
60
27 Co
0
5.2711 (8) a
β- (%)
99.88
0.002
0.12
0.0000020
99.85
0.0075
0.0012
0.0076
99.9826
(keV)
3
2
1
0
2505.748
2158.61
1332.508
0
Stable
60
28 Ni
Q- = 2823.07 keV

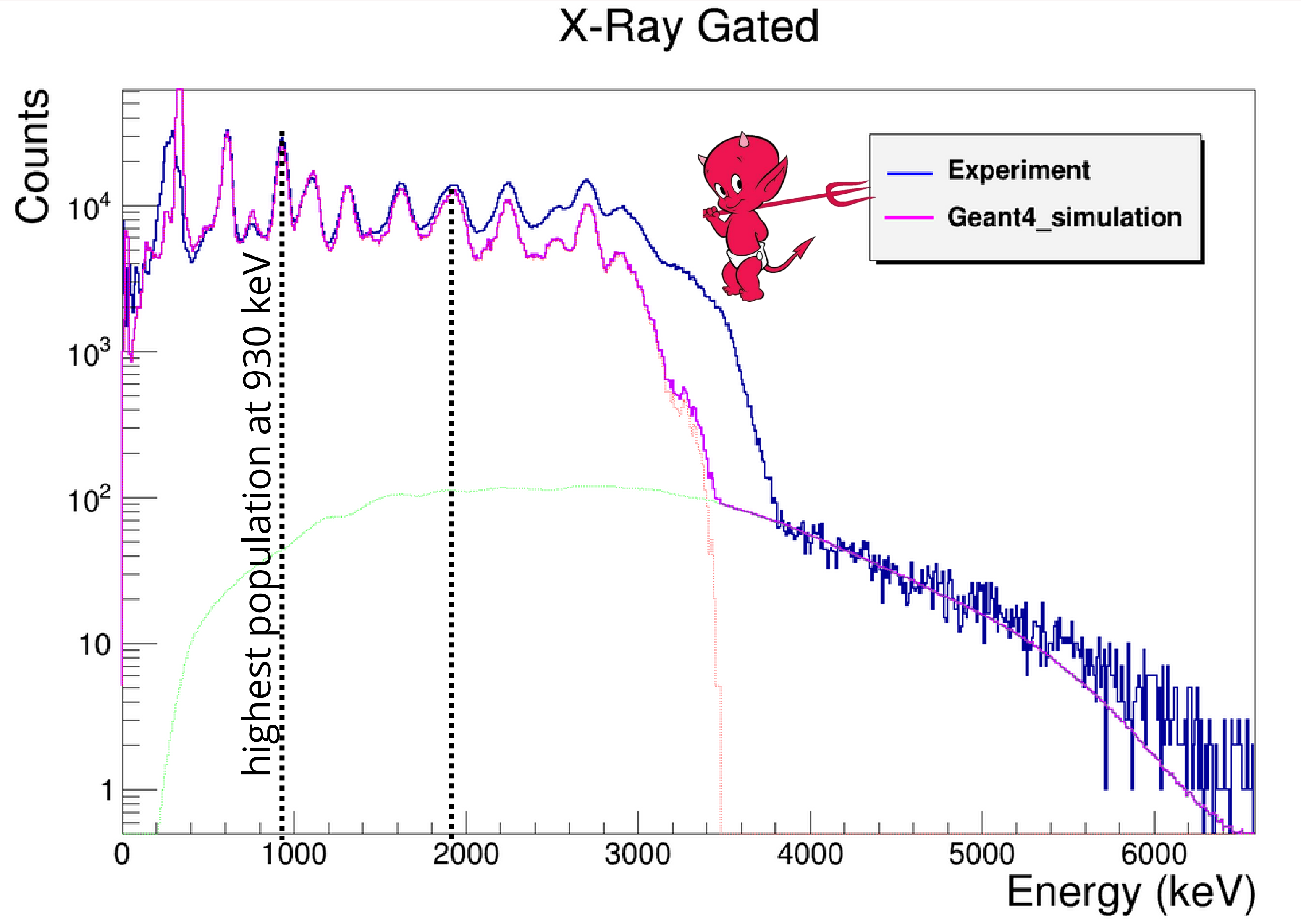

1173+1332 keV
1332 keV
1173 keV
Software sum
Hardware sum
Pileup region
Counts
Energy [keV]
0
1000
2000
3000
4000
5000

^{152}Tb

Larger fraction of level meissed by the HPGe detectors. Considerably large pandemonium. Parallel measurement with FIPPS at ILL (Grenoble)

^{152}Tb is a good isotope for PET imaging, theranostic pair of ^{149}Tb, ^{161}Tb used for therapy, e.g. labeled PSMA-617 (prostate) or DOTATOC (neuroendocrine)





$$d_i = \sum_{j=0}^{j_{max}} R_{ij}(B) f_j + C_i$$

di is the content of bin ***i*** in the measured TAGS spectrum

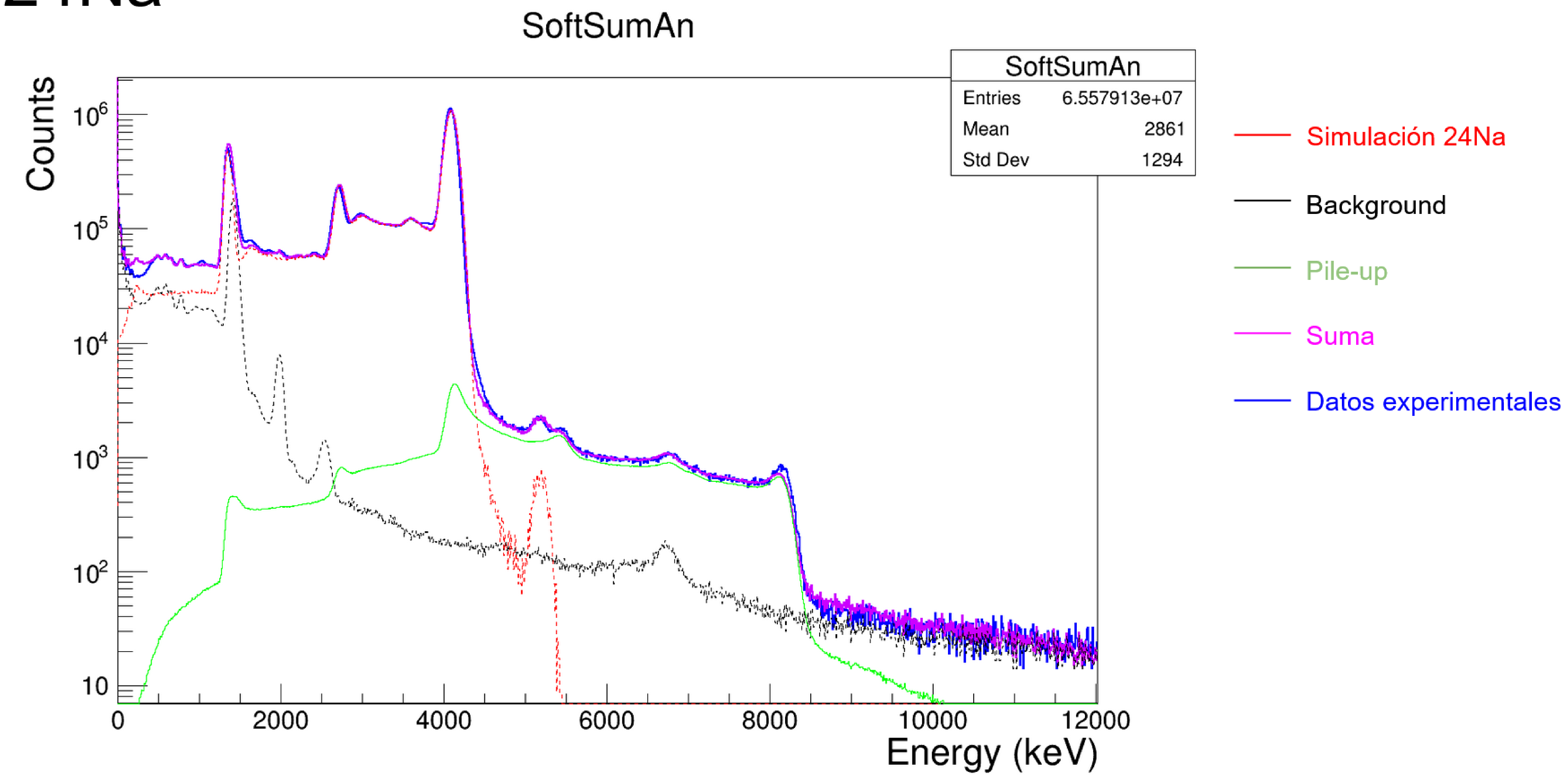
Rij is the response matrix of the TAGS setup and represents the probability that a decay that feeds level ***j*** in the level scheme of the daughter nucleus gives a count in bin ***i*** of the TAGS spectrum and

fj is the β feeding to the level ***j***

Expectation Maximisation Algorithm (EM)

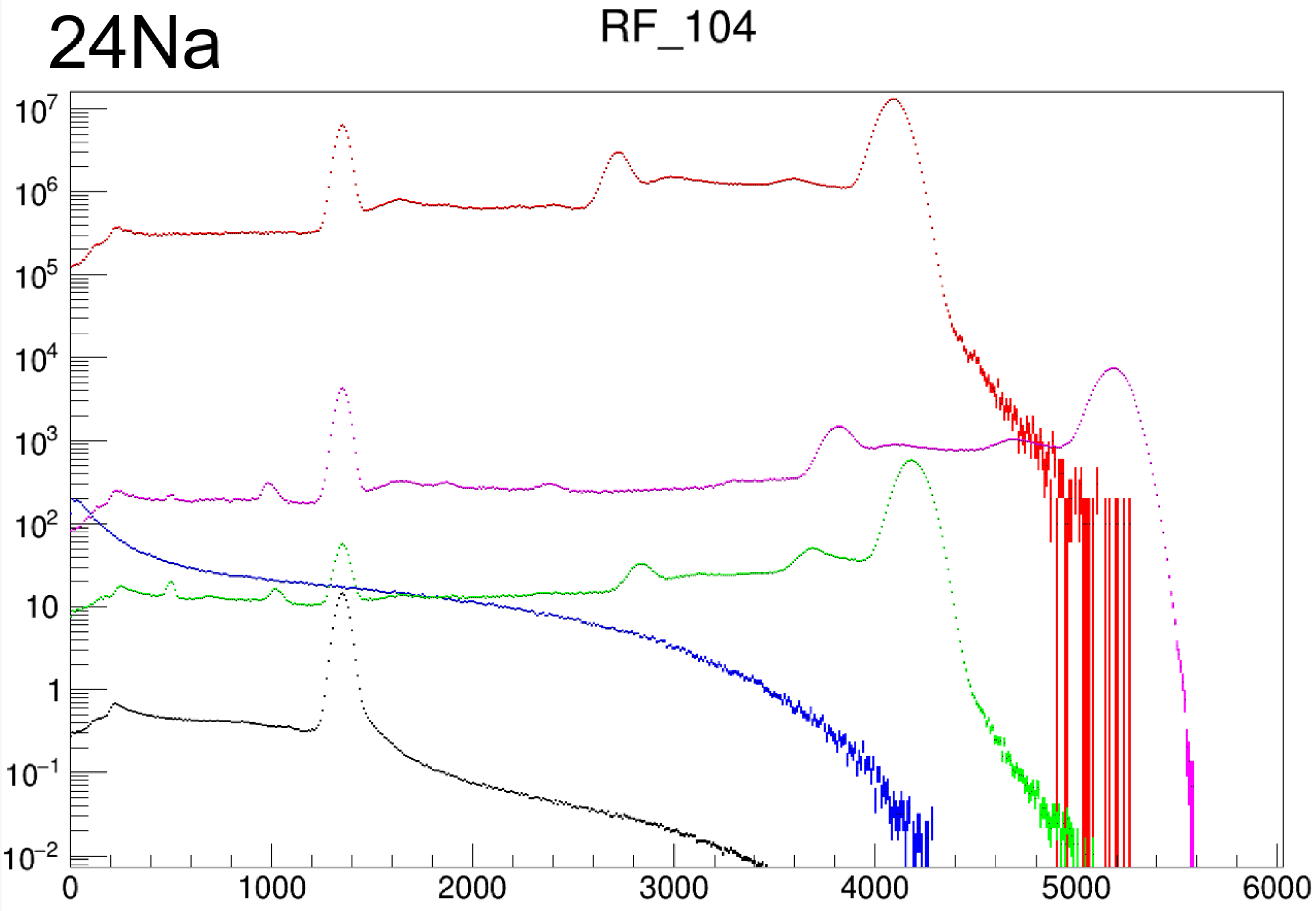
$$f_j^{(s+1)} = \frac{1}{\sum_{i=1}^{n} R_{ij}} \sum_{i=1}^{n} \frac{R_{ij} f_j^{(s)} d_i}{\sum_{k=1}^{m} R_{ik} f_k^{(s)}}, \quad j = 1, \ldots, m$$

24Na



SoftSumAn
SoftSumAn
Entries 6.557913e+07
Mean 2861
Std Dev 1294
Counts
Energy (keV)
10
10^2
10^3
10^4
10^5
10^6
0
2000
4000
6000
8000
10000
12000
Simulación 24Na
Background
Pile-up
Suma
Datos experimentales

24Na



RF_000	0	1
RF_001	1368	35
RF_002	4122	104
RF_003	4238	106
RF_004	5235	131
RF_005	5260	132
RF_006	5300	133
RF_007	5340	134
RF_008	5380	135
RF_009	5420	136
RF_010	5460	137

```
root [5] CheckSolution()
Energy level:  0      feeding:  0.00541198
Energy level:  1360     feeding:  3.54733e-05
Energy level:  4120     feeding:  99.8991
Energy level:  4200     feeding:  0.00349092
Energy level:  5200     feeding:  0.0687819
Energy level:  5240     feeding:  0.00888182
Energy level:  5280     feeding:  0.00262065
Energy level:  5320     feeding:  0.00124902
Energy level:  5360     feeding:  0.00124219
Energy level:  5400     feeding:  0.00234428
Energy level:  5440     feeding:  0.00686428
```

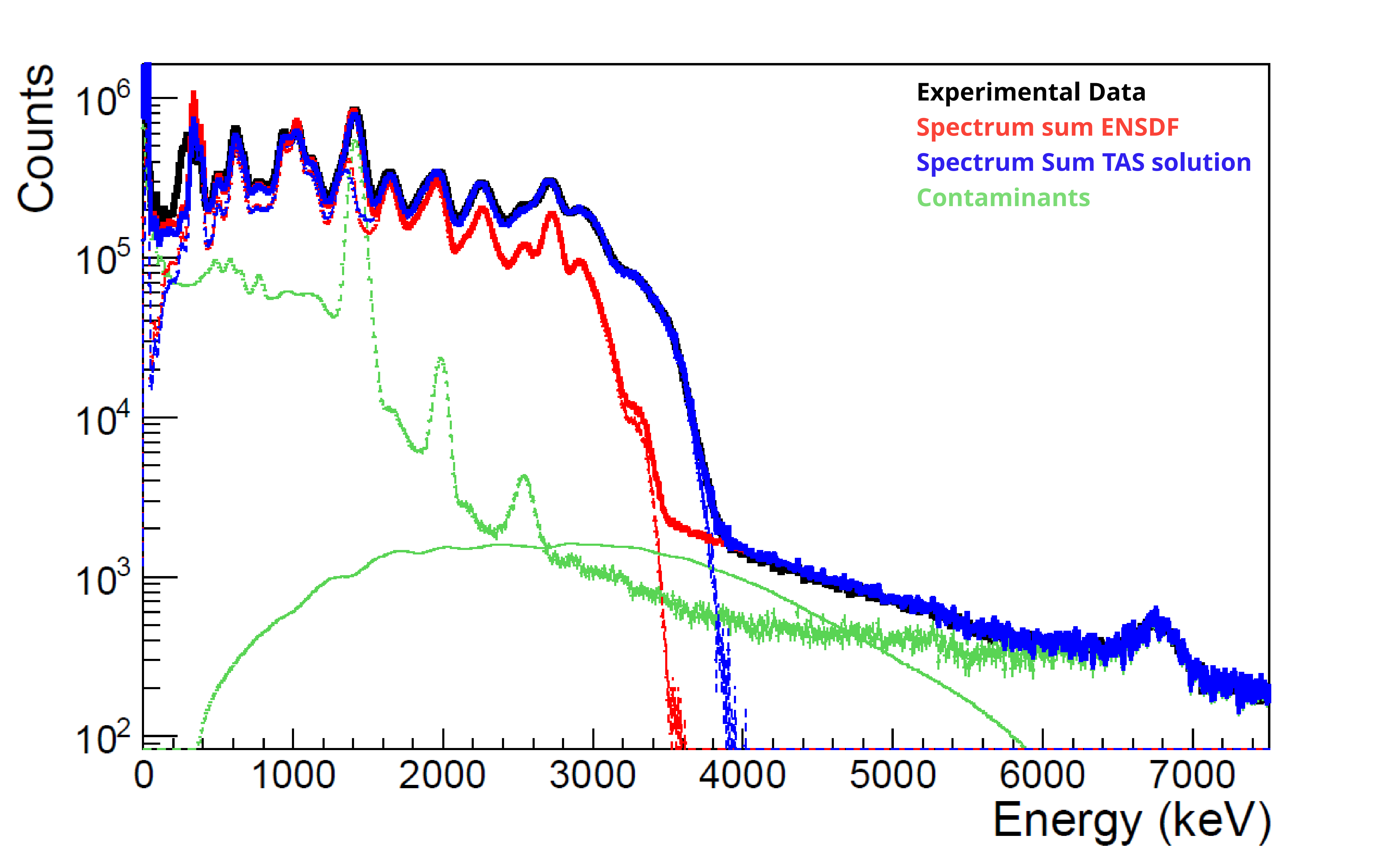
RF_001 * 0.00541198

RF_ 035 * 3.54733e-05

RF_104 * 99.89.91

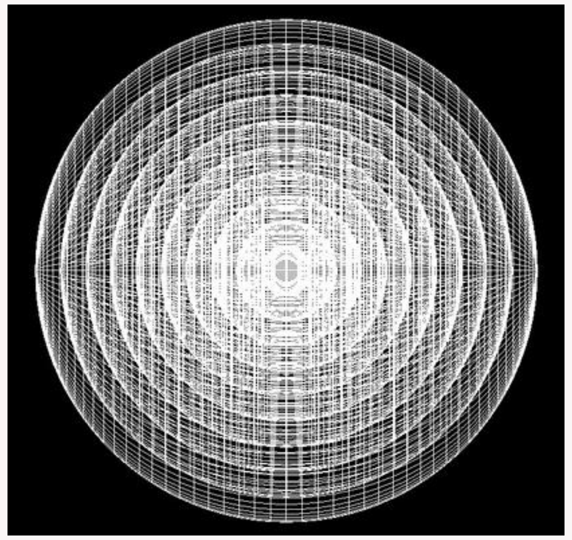
RF_106 * 0.00349092

RF_131 * 0.068789

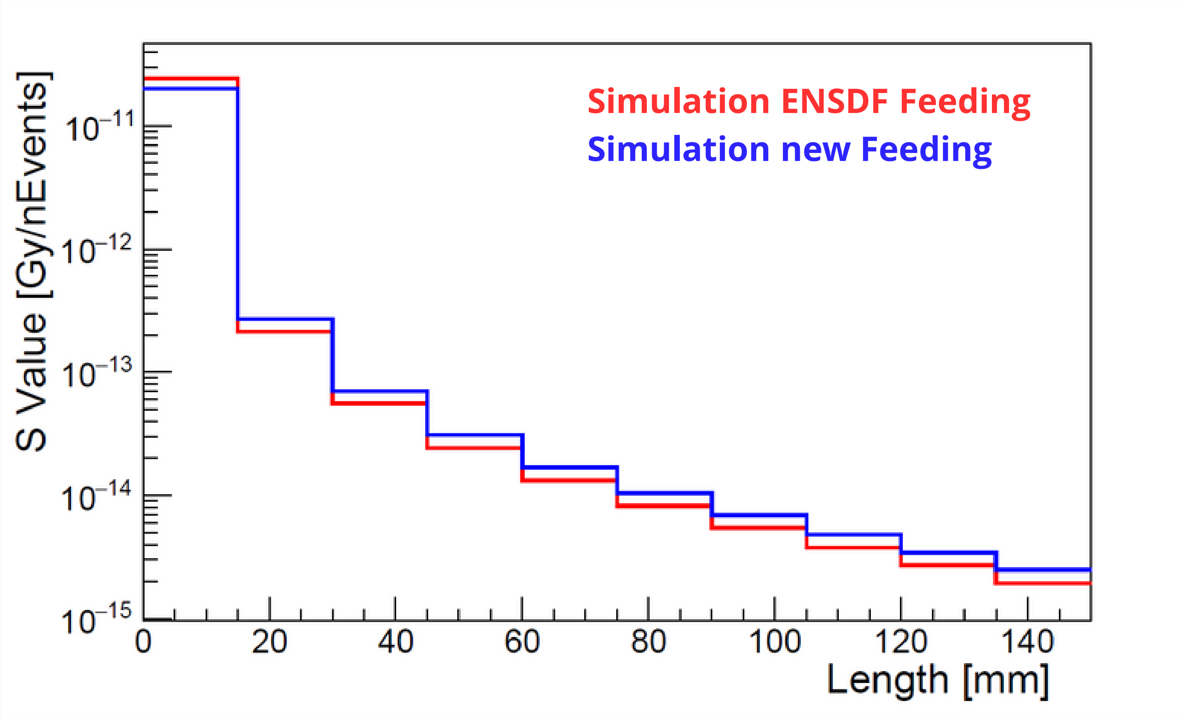

Counts
Experimental Data
Spectrum sum ENSDF
Spectrum Sum TAS solution
Contaminants
10^6
10^5
10^4
10^3
10^2
0
1000
2000
3000
4000
5000
6000
7000
Energy (keV)

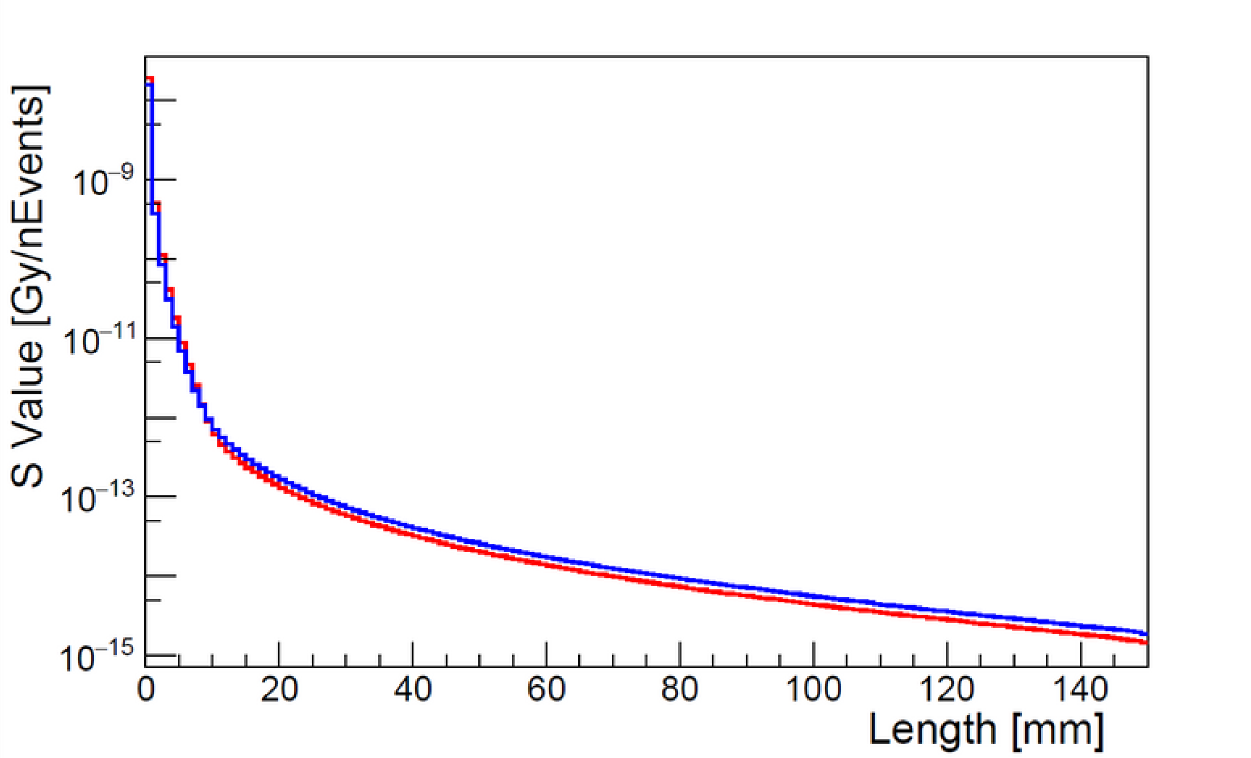
Results ^{152}Tb

Simulation Dose



- Water sphere
- Radio 150 mm
- Source 152Tb

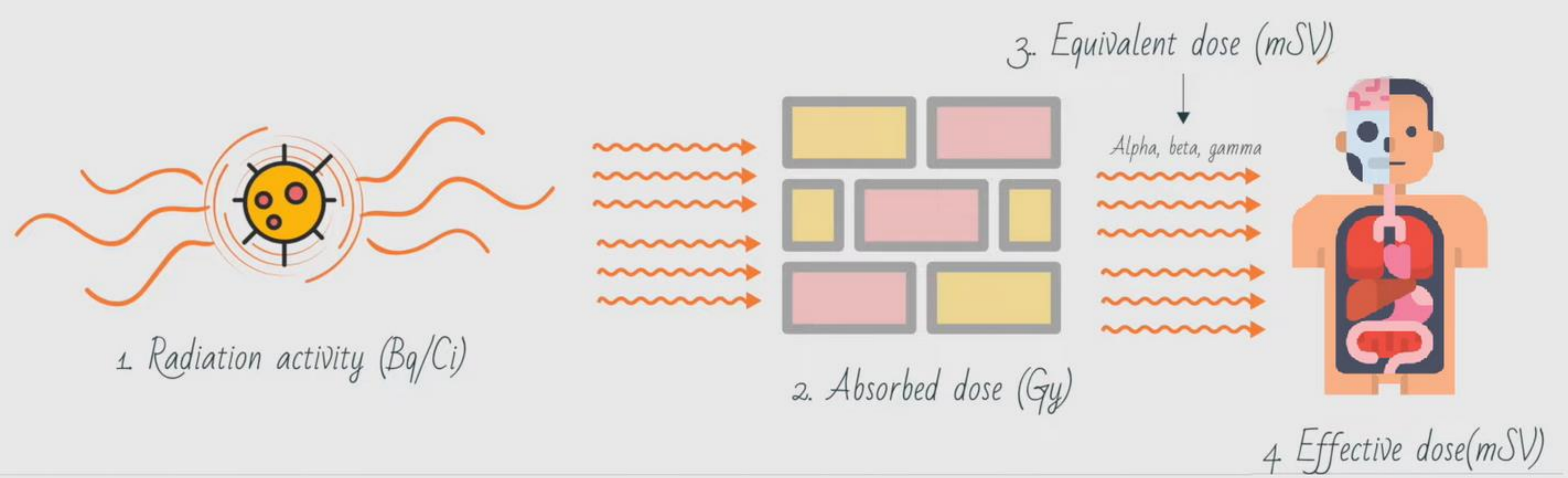




1 Millon events

Beta+ : 20,19 %

Beta+ : 14,14 %



$$S_{(Target \leftarrow Source)} = \frac{D}{N_{events}}$$

$$S_{(Target \leftarrow Source)} = \frac{E_{dep}}{m_T N_{events}}$$

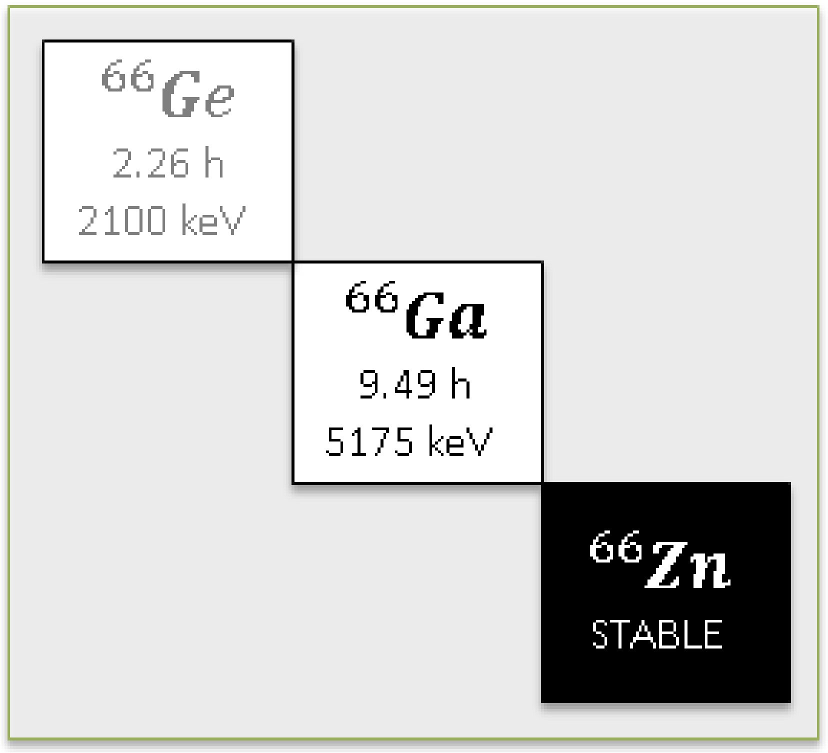
D : absorbed dose to target in **Gy** (J/kg)

m : mass of target (kg)

E : Energy dep of target (kg)

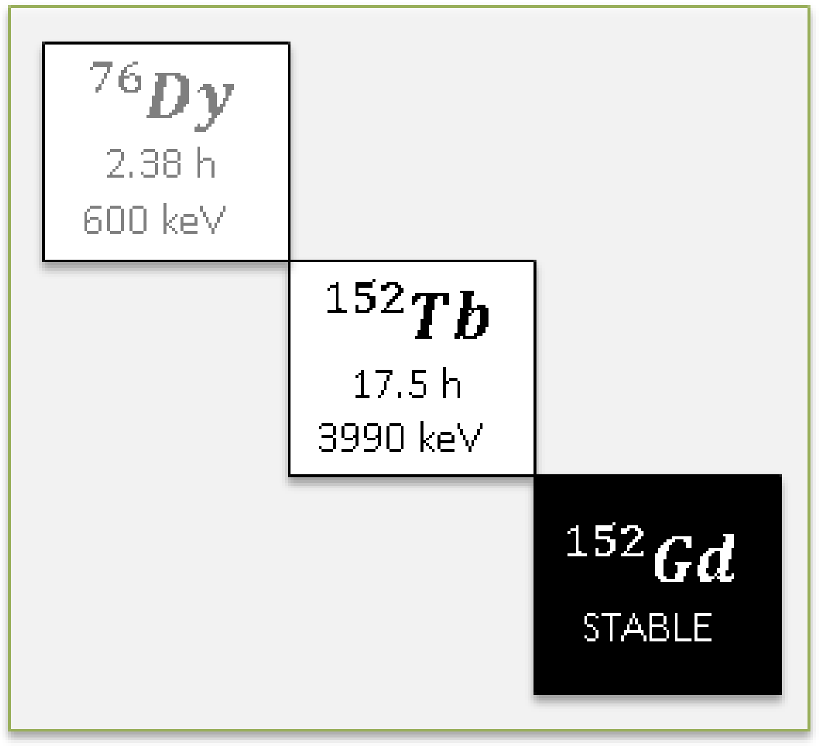
Isotopes of interest

Potentially useful non-standard β^+ emitters.



Complex and well defined decay scheme.
4γ rays unplaced

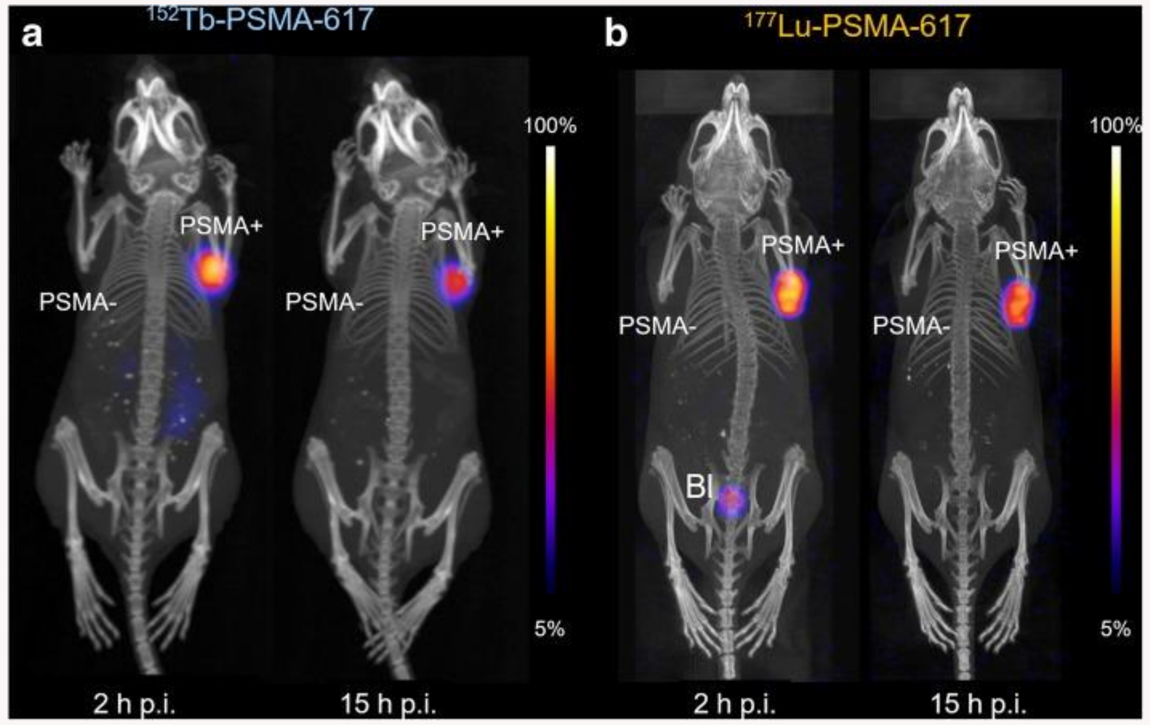
Theranostic Pair



Extremely extensive and complex decay scheme.
248γ rays unplaced

Theracnostics pairs

thera/peutic + diag/**nostic** --->**theranostic** pair



^{66}**Ga** — ^{67}**Ga** — SPECT + microdosimetry

^{152}**Tb** — ^{161}**Tb** — Radiotherapy + microdosimetry

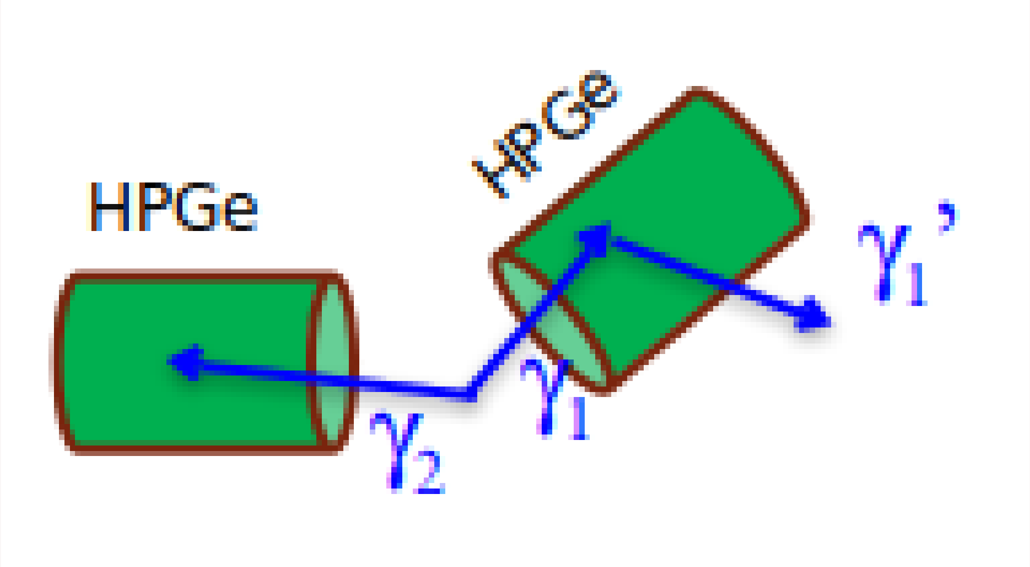
The isotopes of interest

152Tb is used in medicine within radiolabelled molecules for PET and SPECT imaging or as theranostic pairs for treatment.

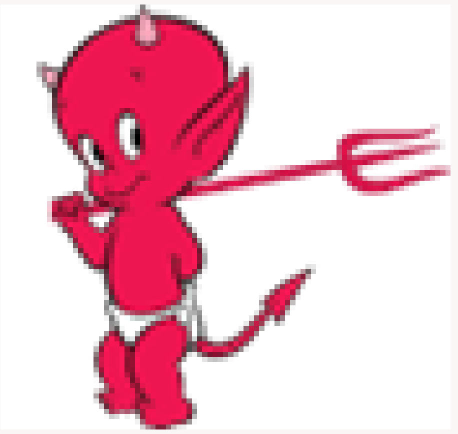
These isotope, among others, have been identified by Nichols [NIC22] as needing for a TAGS measurement

^{152}Tb use with ^{161}Tb as theranostic pair	17.5(1) h	< (7.0×10^{-7})% α 100% EC/β^+: 20.3(15)% β^+ 79.7(15)% EC	95 to 100EC/β^+, 387γ + 248γ unplaced $E_{\beta+}^{\text{end point}}$: 2037, 2353, 2624, 2968 keV E_γ: 271.09, 344.279, 586.27 keV	Most significant EC/β^+ decay directly to the ground (25% (8.0% β^+)), first (12.7% (5.9% β^+)), second (6.85% (1.20% β^+)) and fourth (8.06% (2.30% β^+)) excited states of ^{152}Gd, along with significant depopulation by 271.09, 344.279- and 586.27-keV γ rays and over 380 lower-intensity γ emissions from 117.25 to 3140.20 keV – *extremely extensive and complex decay scheme that includes as many as 387 γ rays of which the placement of only seven are in doubt, while a further 248 γ rays remain unplaced (113.5 to 3621.7 keV); suitable candidate for TAGS*

High resolution (HR) technique.



The spectrum can suffer from the **Pandemonium effect.**



The efficiency drops as we look to higher energies, which means that it has a bias to γ-rays at high energy.

Hardy et al., Physics letters B 71. (1977)
Pandemonium