Decay experiments by the Nantes-Valencia Collaboration

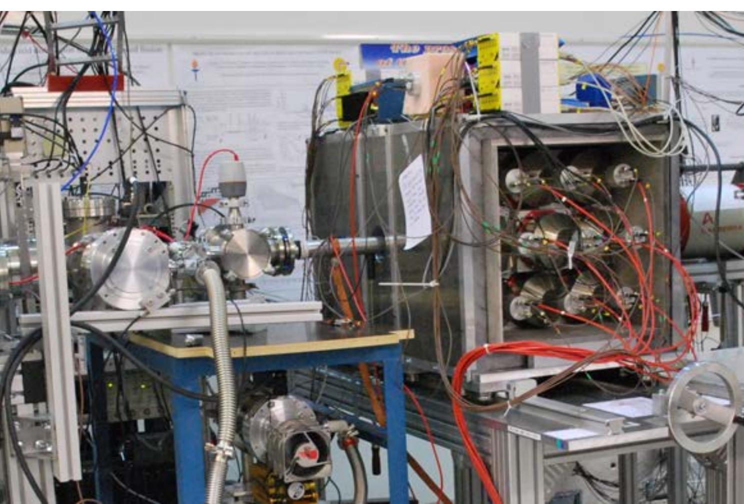
A. Algora

IFIC, CSIC-University of Valencia

and ATOMKI, Debrecen

For the Valencia-Subatech Collaboration

Results from the VTAS, DTAS collaboration experiments at Jyväskylä



A few words on antineutrino spectrum calculations

Two main categories

- Models constrained by Schreckenbach et al. data (measurements of the conglomerate beta spectrum at ILL and converted into antineutrino spectrum using different assumptions)

  Schreckenbach et al. works

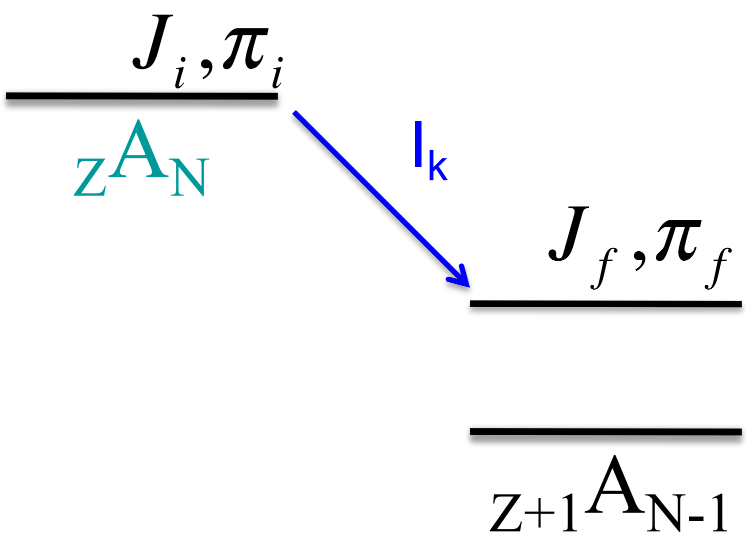
  Huber model (conversion)

  Mueller model (summation-conversion)

- Summation calculations that require again a large amount of nuclear data (high quality beta decay data, fission yields, shape corrections)

Neutrino and decay heat summation calculations

Beta decay (β^-)



Spectrum for each transition

$$J_i,\pi_i \rightarrow J_f,\pi_f$$

$$S(Q-E_k,J_i\pi_i,J_f\pi_f)$$

Spectrum for the decay (n)

$$S_n(E)=\sum_k I_k S(Q-E_k,J_i\pi_i,J_f\pi_f)$$

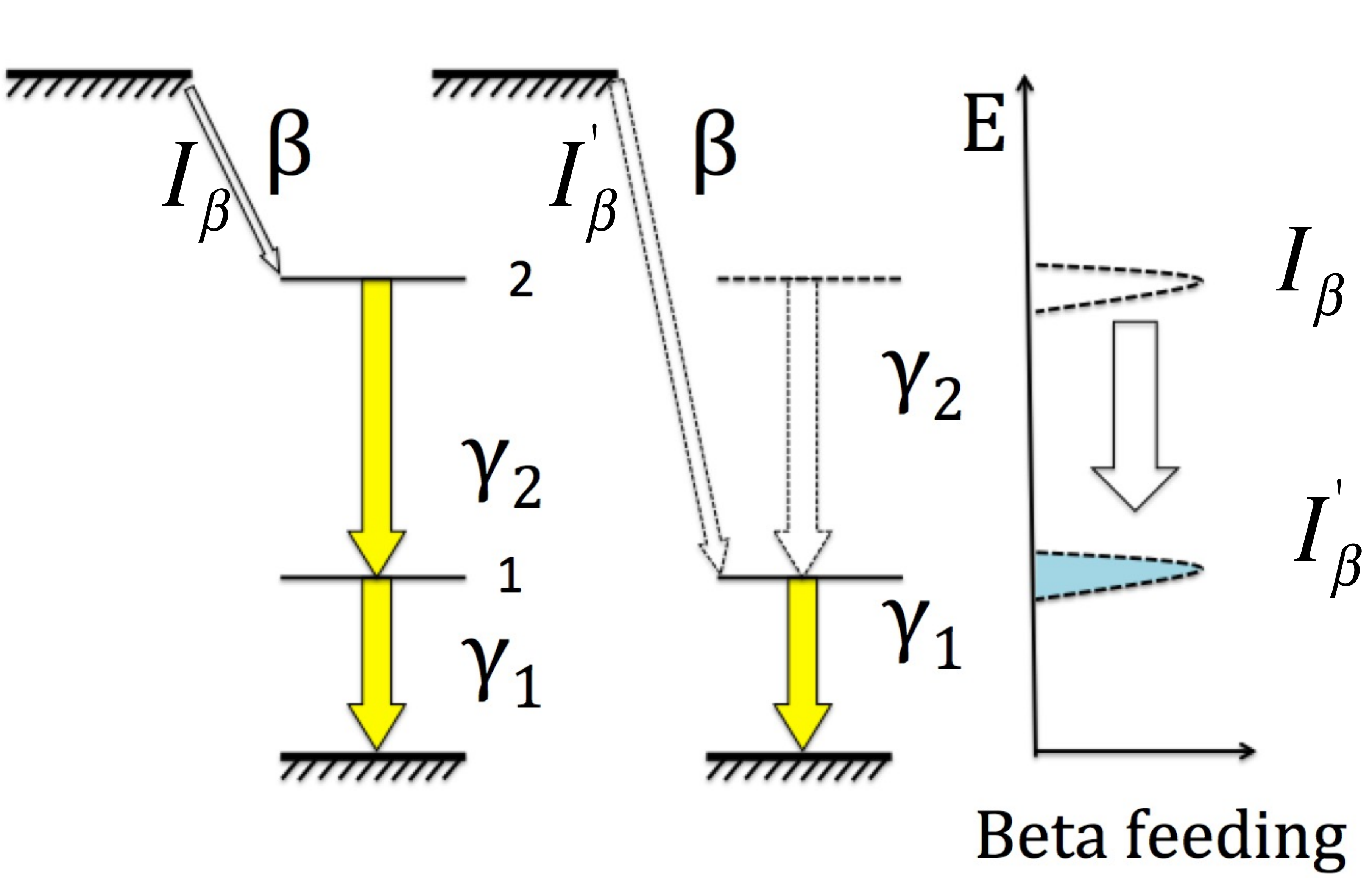
Anti-neutrino rate per fission (Vogel, 1981)

$$S(E)=\sum_n \lambda_n N_n S_n(E)/r=\sum_n CFY_n S_n(E)$$

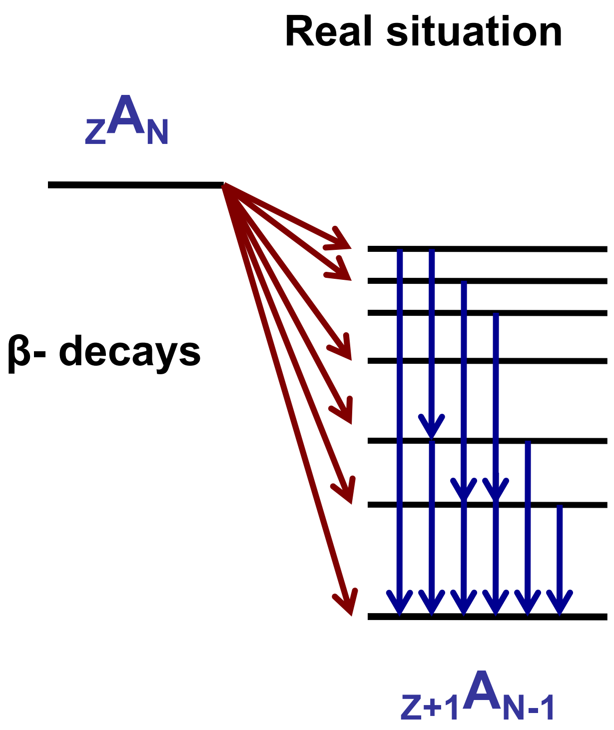
Decay heat summation calculation

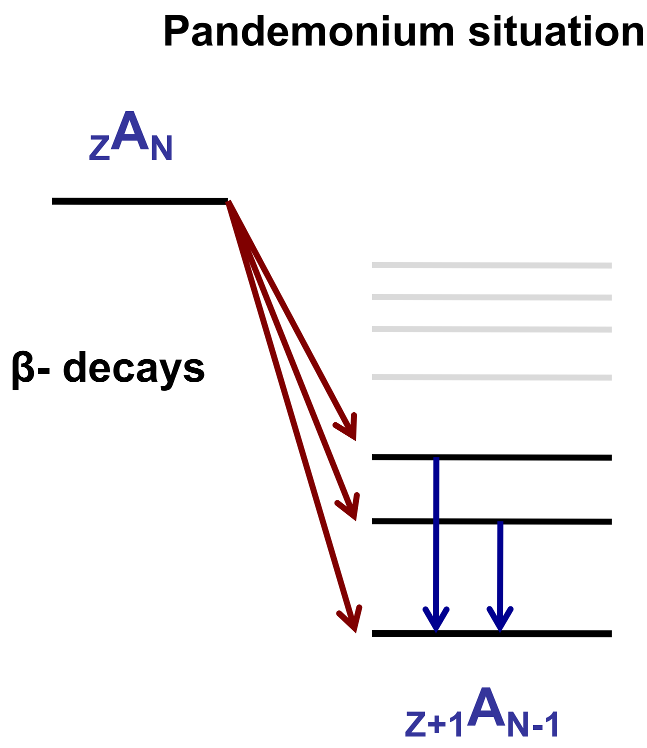
$$f(t)=\sum_i E_i\lambda_i N_i(t)$$

What can go wrong? Pandemonium effect



Effect of Pandemonium on summation calculations (decay heat example)



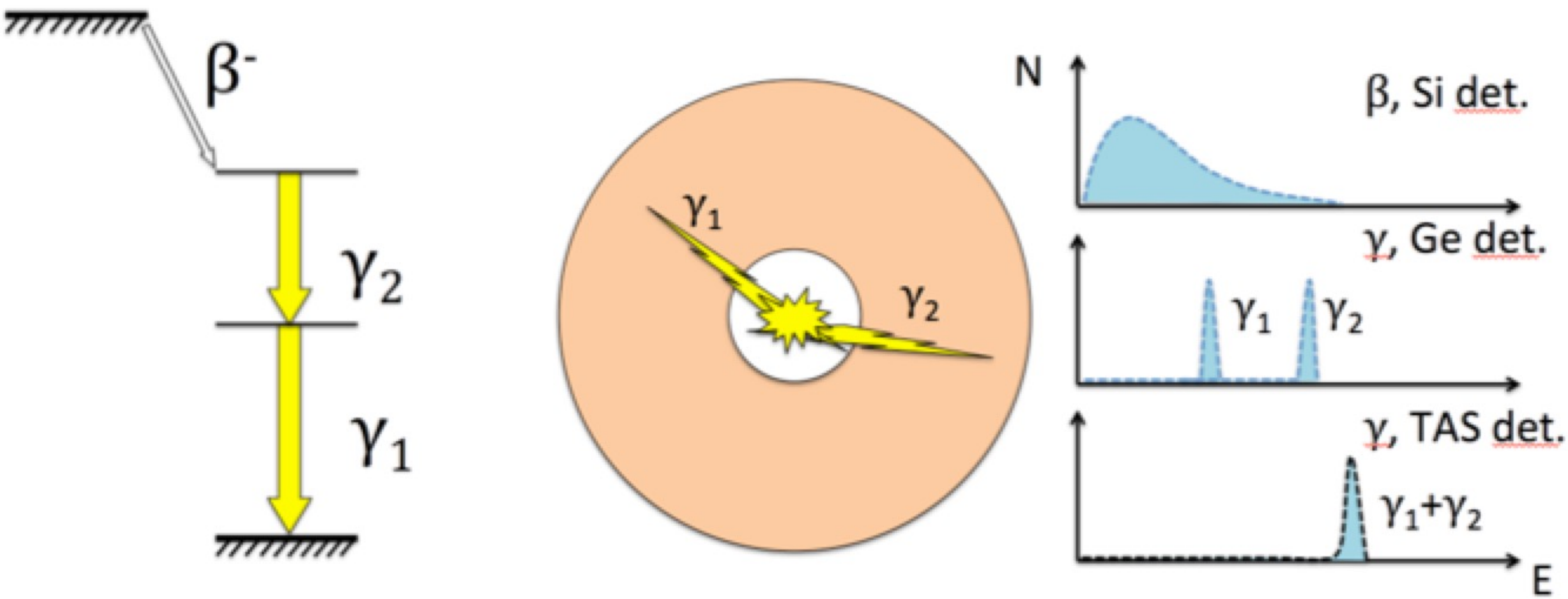


As a result of the Pandemonium, betas are estimated with higher energies from databases. Their spectra is harder. Incomplete level schemes can affect the antineutrino calculations as well.

The gamma mean energies are reduced since you detect less gammas.

This is why you should avoid using data suspicious of suffering from Pandemonium

Total absorption spectroscopy applied to beta decay studies

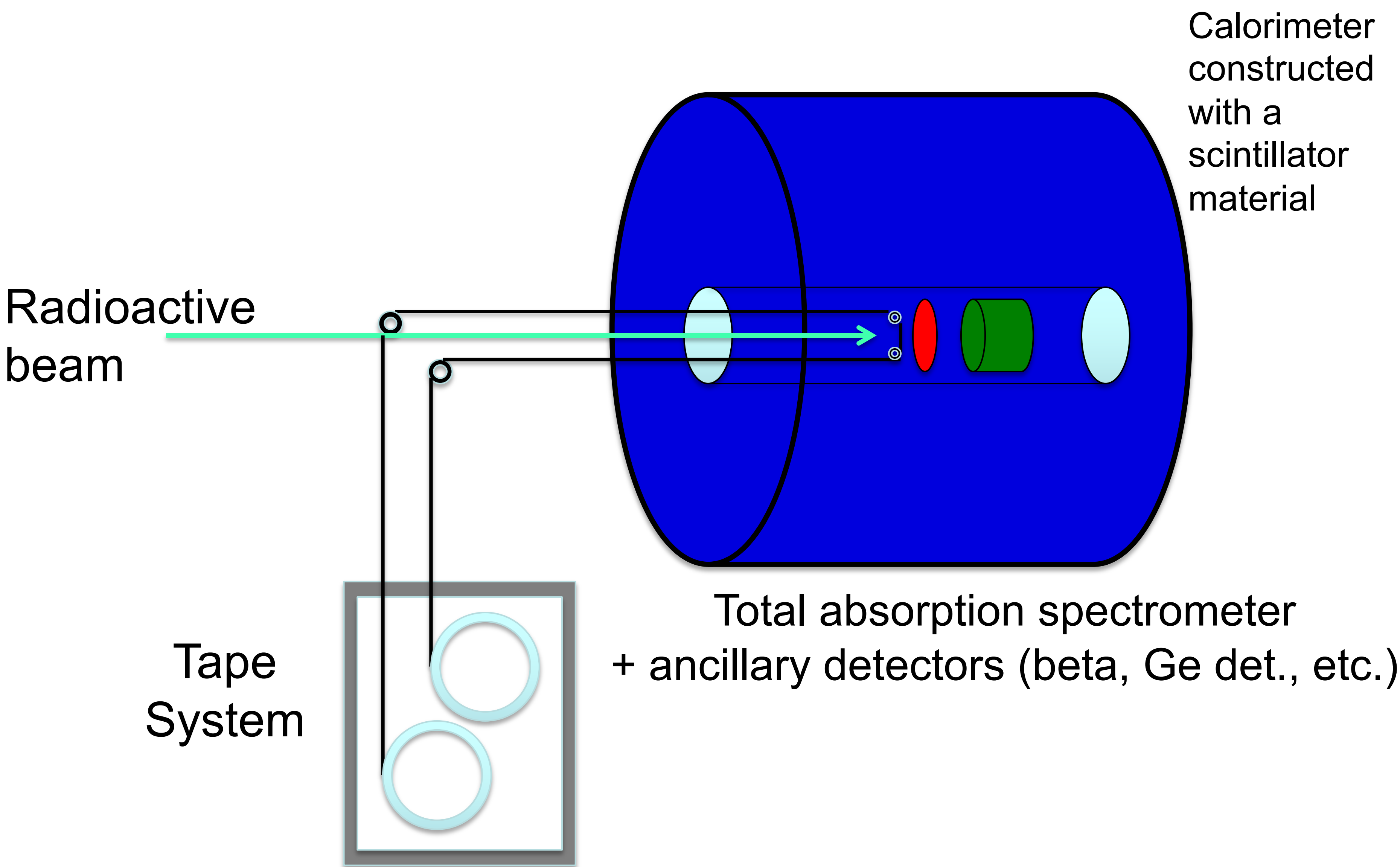


$$d = R(B) \cdot f$$

$(note\ that\ f \equiv I_\beta)$

Requirements: clean spectrum or a proper treatment of the contaminants, some knowledge of decay level scheme of the daughter, etc.

Typical total absorption experiments



What was meant by "high quality decay data" for summation calculations

- Use of beta decay data that does not suffer from the Pandemonium effect
- Proper determination of the ground state to ground state feeding (it affects the global feeding normalization, and can provide absolute gamma branches per decay of relevance for many applications.)
- In the case of beta decays that present beta delayed neutron emission, determination of the gamma/neutron competition above the neutron separation energy

The beginning (for us) …

We got interested in the topic after the work of Yoshida and co-workers (Journ. of Nucl. Sc. and Tech. 36 (1999) 135)

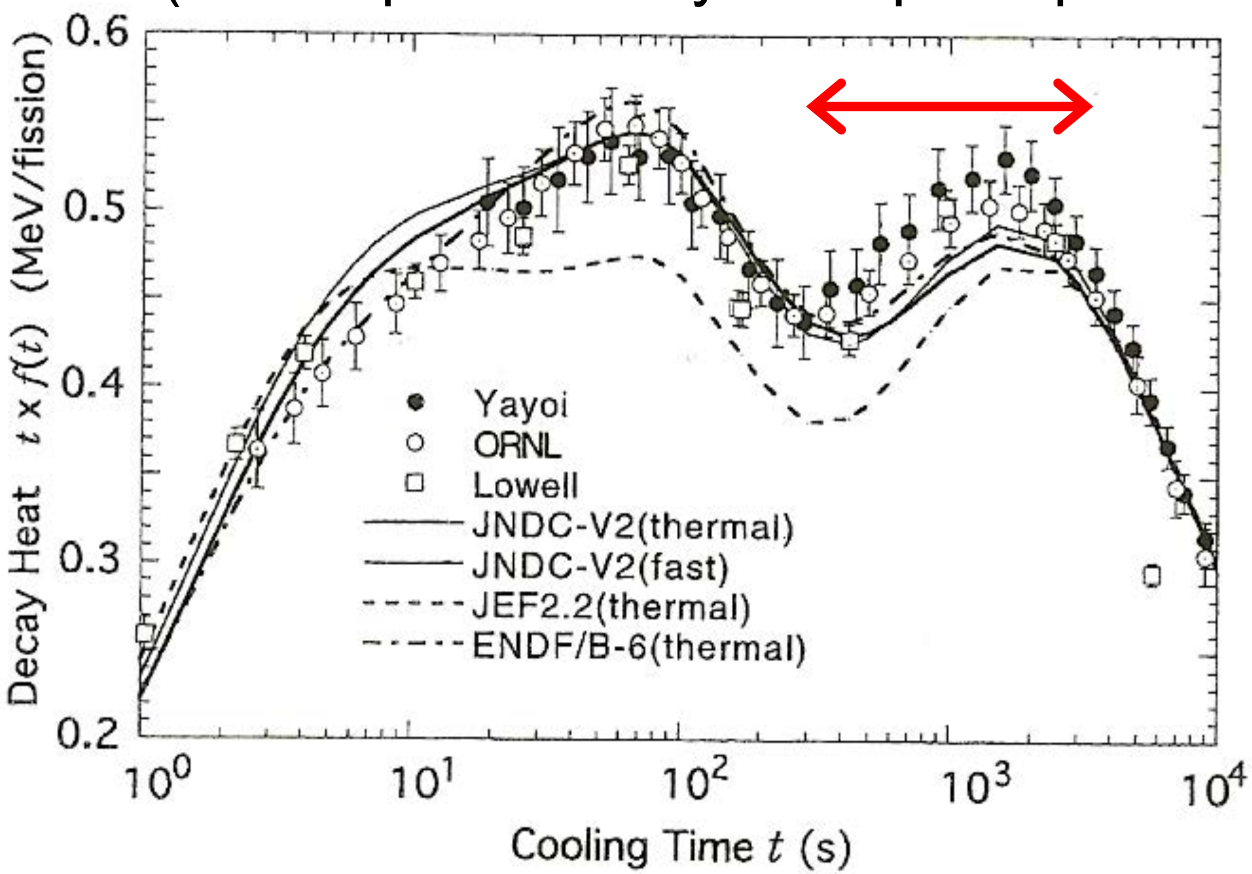
^{239}Pu example (similar situation for ^{235,238}U)

Detective work: identification of some nuclei that could be blamed for the "anomaly" ^{102,104,105}Tc

Suggestion: TAGS measurements !

^{239}Pu example

(fission pulse decay heat γ component)



Some published and on-going cases for Decay Heat and Antineutrino Spectrum calculations

Tables extracted from « Beta-decay studies for applied and basic nuclear physics », Algora et al. Eur. Phys. J. A 57 (2021) 85, 2020

Table 2. List of parent nuclides identified by the WPEC-25 (Nuclear Energy Agency working group) that should be measured using the total absorption technique to improve the predictions of the decay heat in reactors [48,49]. These nuclides are of relevance for conventional reactors based on ^{235}U and ^{239}Pu fission. The list contains 37 nuclides. Rel. (relevance) stands for the priority of the measurement. Isotopes marked with asterisks show the measurements performed by our collaboration. Nuclides marked with † are also relevant for the ^{233}U/^{232}Th fuel, see additional cases in Table 3. The isotopes are identified according to the Z-Symbol-A notation; m stands for metastable or isomeric state.

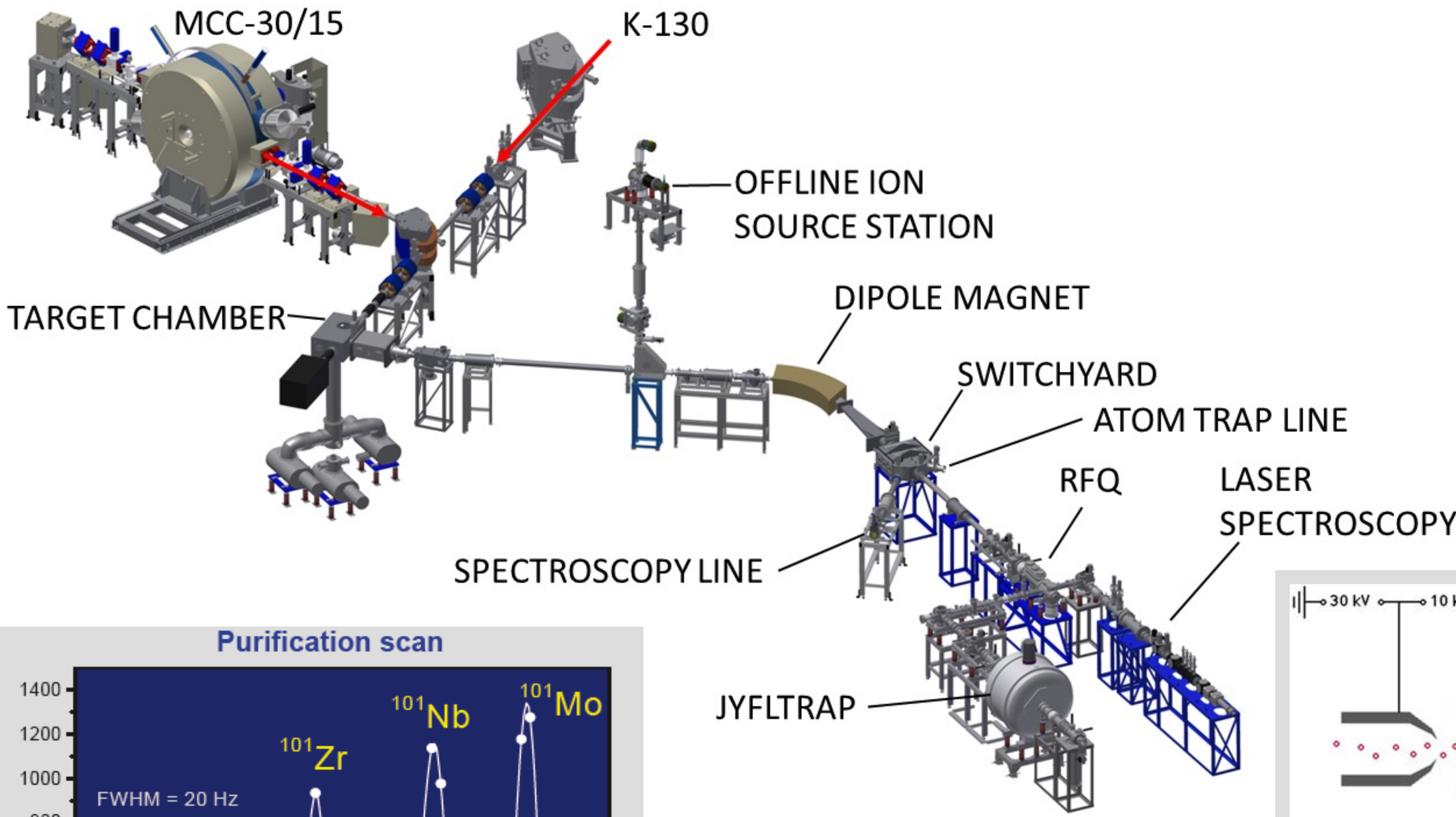
Isotope	Rel.	Isotope	Rel.	Isotope	Rel.
35-Br-86†*	1	41-Nb-99†	1	52-Te-135†	2
35-Br-87†*	1	41-Nb-100†*	1	53-I-136†	1
35-Br-88†*	1	41-Nb-101†*	1	53-I-136m†	1
36-Kr-89†	1	41-Nb-102†*	2	53-I-137†*	1
36-Kr-90†	1	42-Mo-103†*	1	54-Xe-137†	1
37-Rb-90m	2	42-Mo-105*	1	54-Xe-139†	1
37-Rb-92†*	2	43-Tc-102†*	1	54-Xe-140†	1
38-Sr-89	2	43-Tc-103†*	1	55-Cs-142*	3
38-Sr-97	2	43-Tc-104†*	1	56-Ba-145	2
39-Y-96†	2	43-Tc-105*	1	57-La-143	2
40-Zr-99†	3	43-Tc-106*	1	57-La-145	2
40-Zr-100†	2	43-Tc-107*	2		
41-Nb-98†*	1	51-Sb-132†	1		

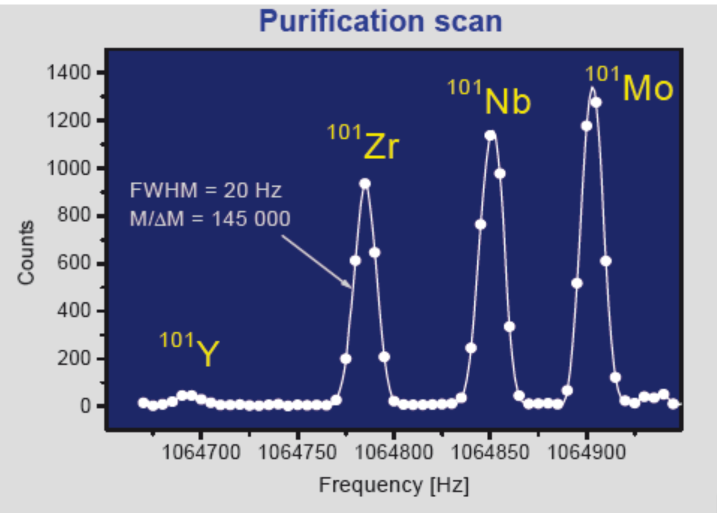
Table 6. List of nuclides identified by the IAEA TAGS Consultants that should be measured using the total absorption technique to improve the predictions of the reactor antineutrino spectra. These nuclides are of relevance for conventional reactors based on ^{235}U and ^{239}Pu nuclear fuels. The list contains 34 nuclides [103]. Relevance (Rel.) stands for the priority of the measurement. Isotopes marked with asterisks show the measurements performed by our collaboration, m stands for metastable or isomeric state.

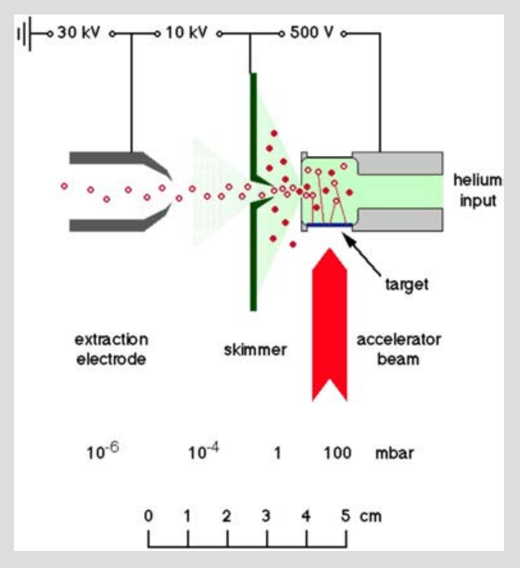
Isotope	Rel.	Isotope	Rel.	Isotope	Rel.
36-Kr-91	2	39-Y-97m	1	53-I-138*	2
37-Rb-88	1	39-Y-98m	1	54-Xe-139	1
37-Rb-90	1	39-Y-99*	1	54-Xe-141	2
37-Rb-92*	1	40-Zr-101	1	55-Cs-139	1
37-Rb-93*	1	41-Nb-98*	1	55-Cs-140*	1
37-Rb-94*	2	41-Nb-100*	1	55-Cs-141	2
38-Sr-95*	1	41-Nb-101*	1	55-Cs-142*	1
38-Sr-96	1	41-Nb-102*	1	57-La-146	2
38-Sr-97	2	41-Nb-104m	2		
39-Y-94	1	52-Te-135	1		
39-Y-95*	1	53-I-136	2		
39-Y-96*	1	53-I-136m	1		
39-Y-97	2	53-I-137*	1		

Courtesy: M. Fallot (with some modifications)

IGISOL IV at Jyväskylä (Ion-guide + JYFL Penning trap)

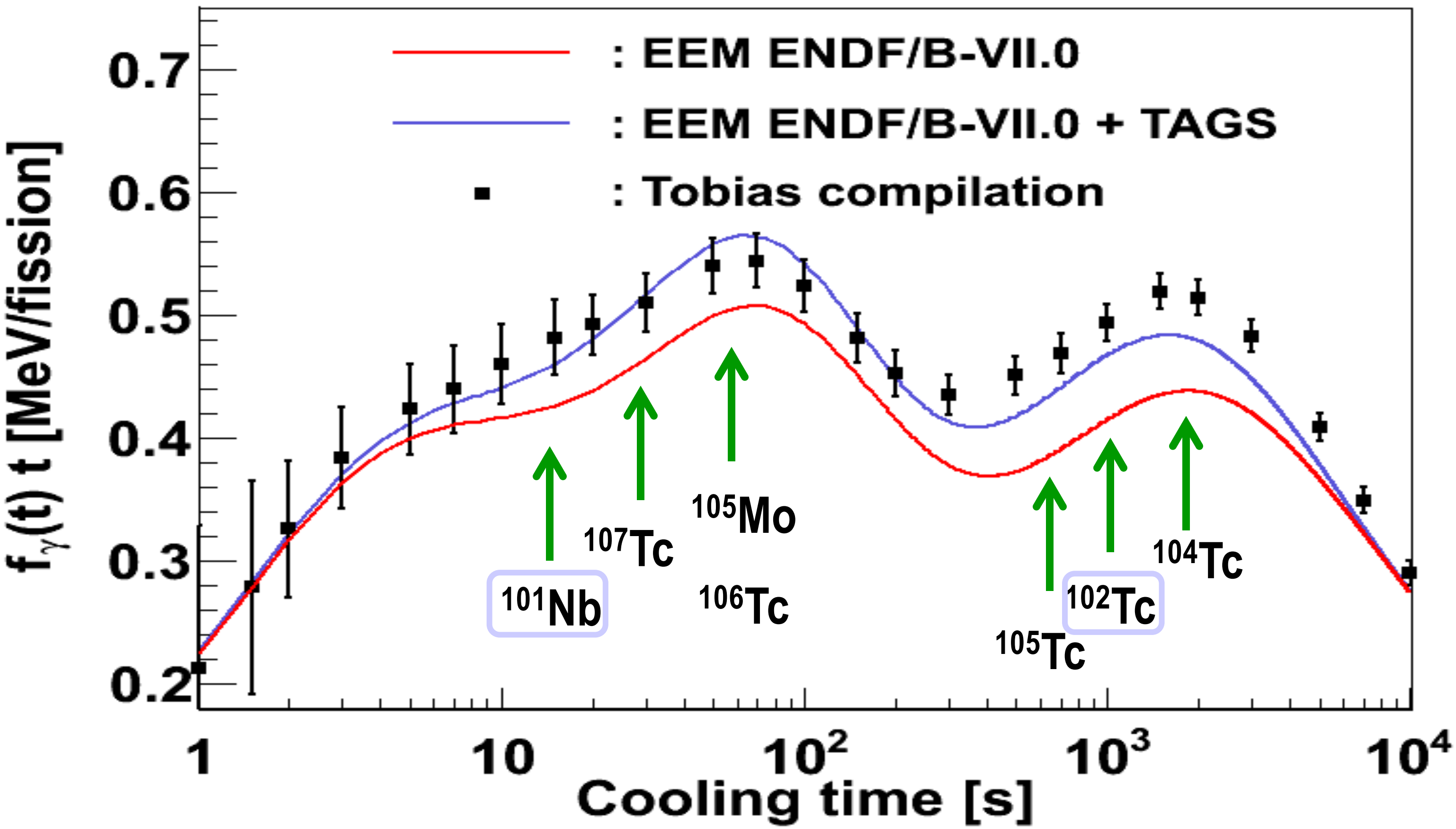






Impact of the earlier results for ^{239}Pu: electromagnetic component

Motivated by Yoshida *et al.* (Journ. of Nucl. Sc. and Tech. 36 (1999) 135) and WPEC-25



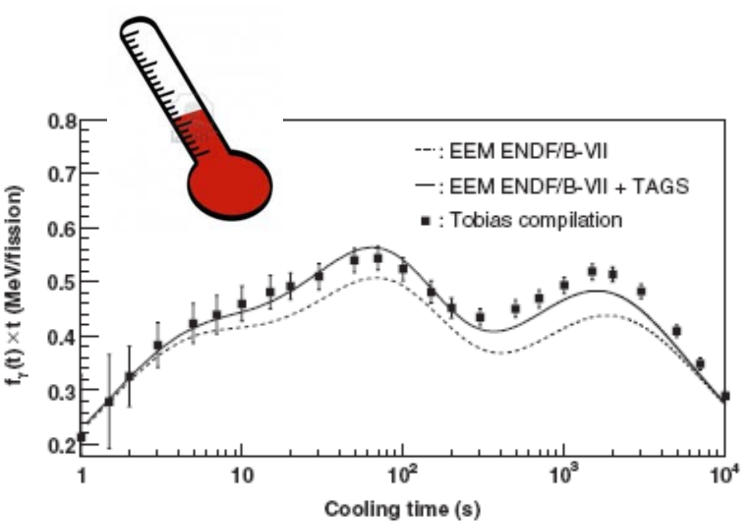
Algora, Phys. Rev. Letts. 105, 202505, PhD Thesis D. Jordan,

K. P. Rykaczewski, Physics 3, 94 (2011)

DH courtesy A. Sonzogni

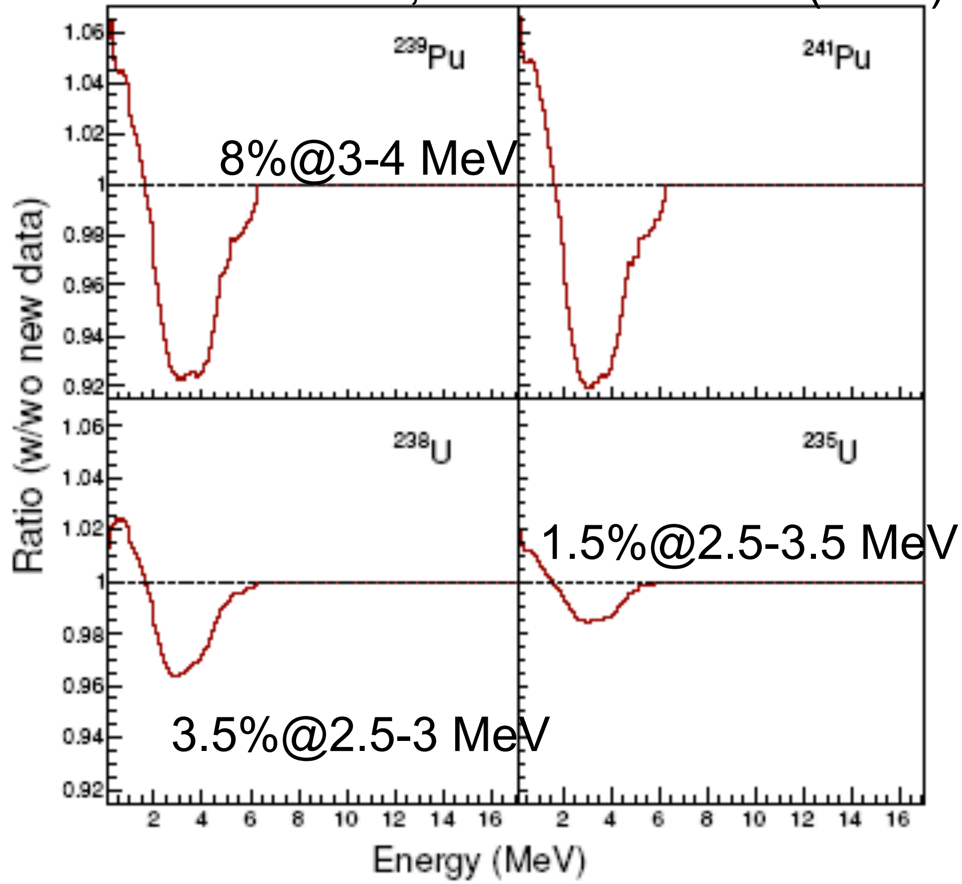
Results also confirmed by R. W. Mills using JEFF 3.1

Impact of some of our earlier data: $^{102,104,105,106,107}Tc$, ^{101}Nb, ^{105}Mo



Dolores Jordan, PhD thesis
Algora et al., PRL 105, 202501 (2010)
D. Jordan PRC 87, 044318 (2013)

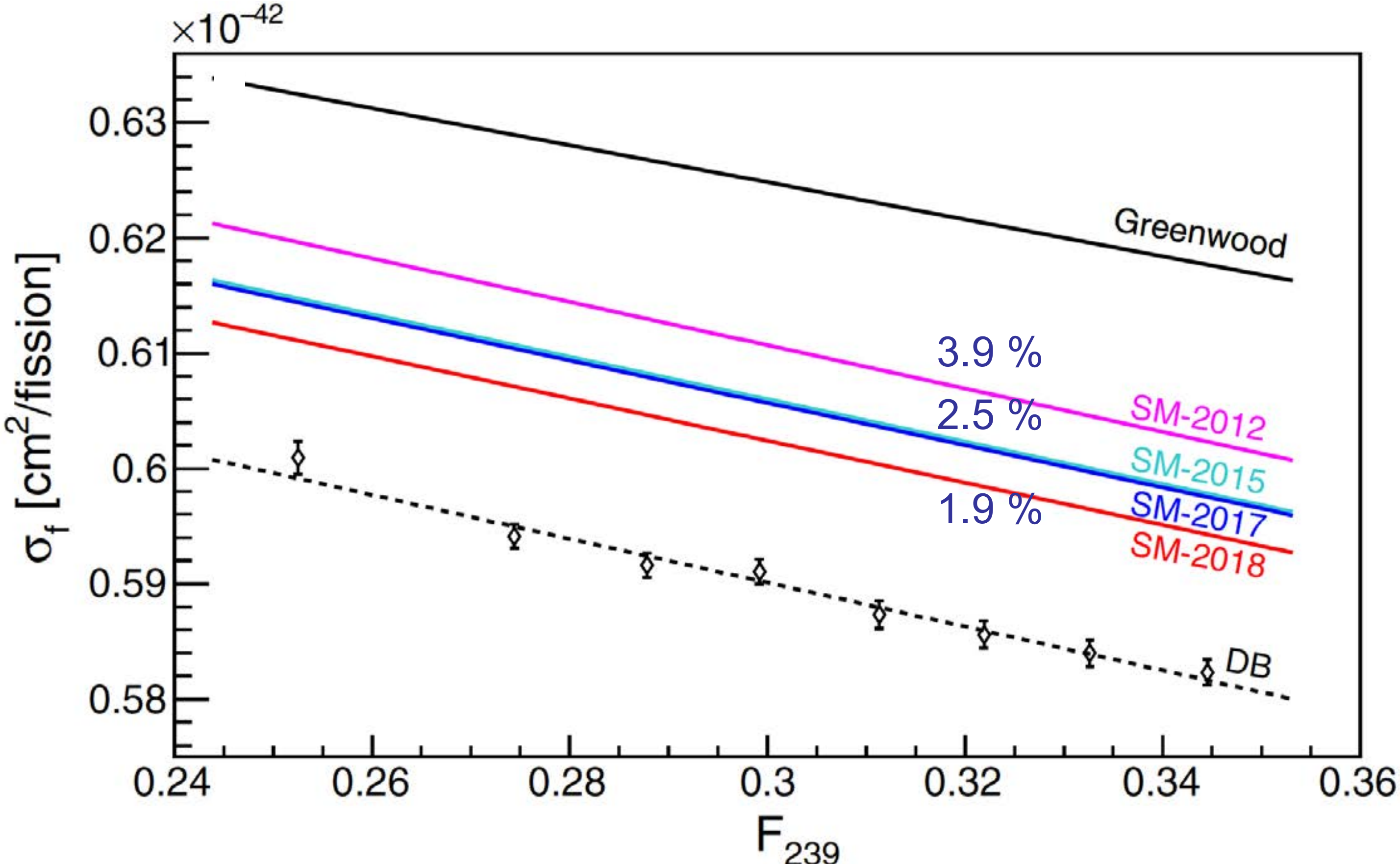
M. Fallot et al., PRL 109.202504 (2012)



Ratio between 2 antineutrino spectra built with and without the $^{102,104,105,106,107}Tc$, ^{105}Mo, ^{101}Nb TAS data. Only 5 Pandemonium cases

TAS impact in summation calculations

Effect of the successive inclusion of TAS data (Pandemonium free data) in the summation model (flux)



Careful selection of the pandemonium free data + TAS data

SM-2012: $^{102,104,105,106,107}Tc$, ^{105}Mo, and ^{101}Nb

SM-2015: $^{92,94}Rb$, and $^{87,88}Br$

SM-2017: ^{91}Rb, ^{86}Br

SM-2018: $^{100,100m,102,102m}Nb$

DB: Daya Bay

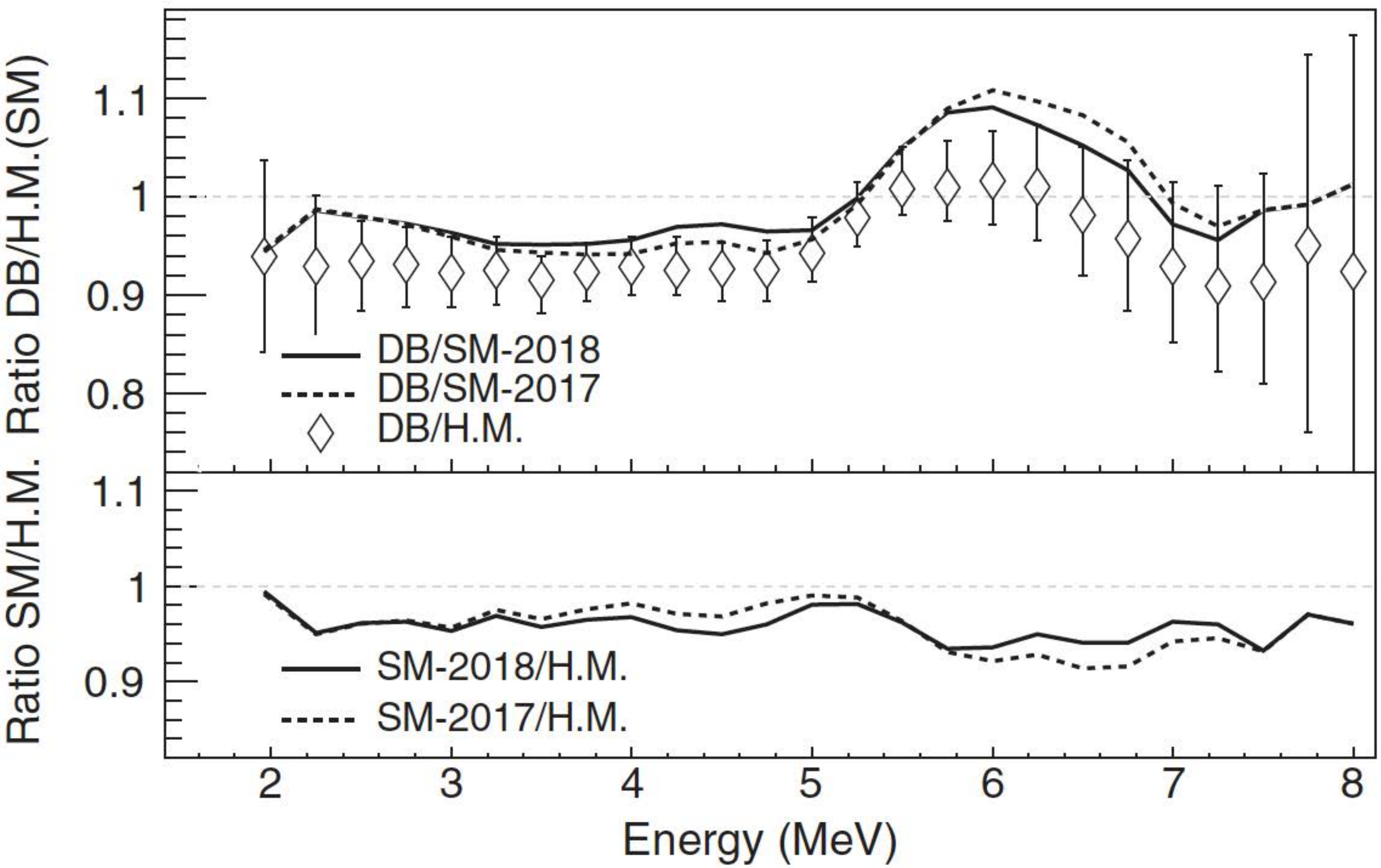
$$\sigma_f(F_{239}) = \bar{\sigma}_f + \frac{d\sigma_f}{dF_{239}}(F_{239} - \bar{F}_{239}).$$

M. Estienne et al. PRL 123, 022502 (2019)

The success of summation calculations

Results from the application of a new summation calculation including all our TAS measurements. The discrepancy with the antineutrino meas. within this model is of the order of 2 %

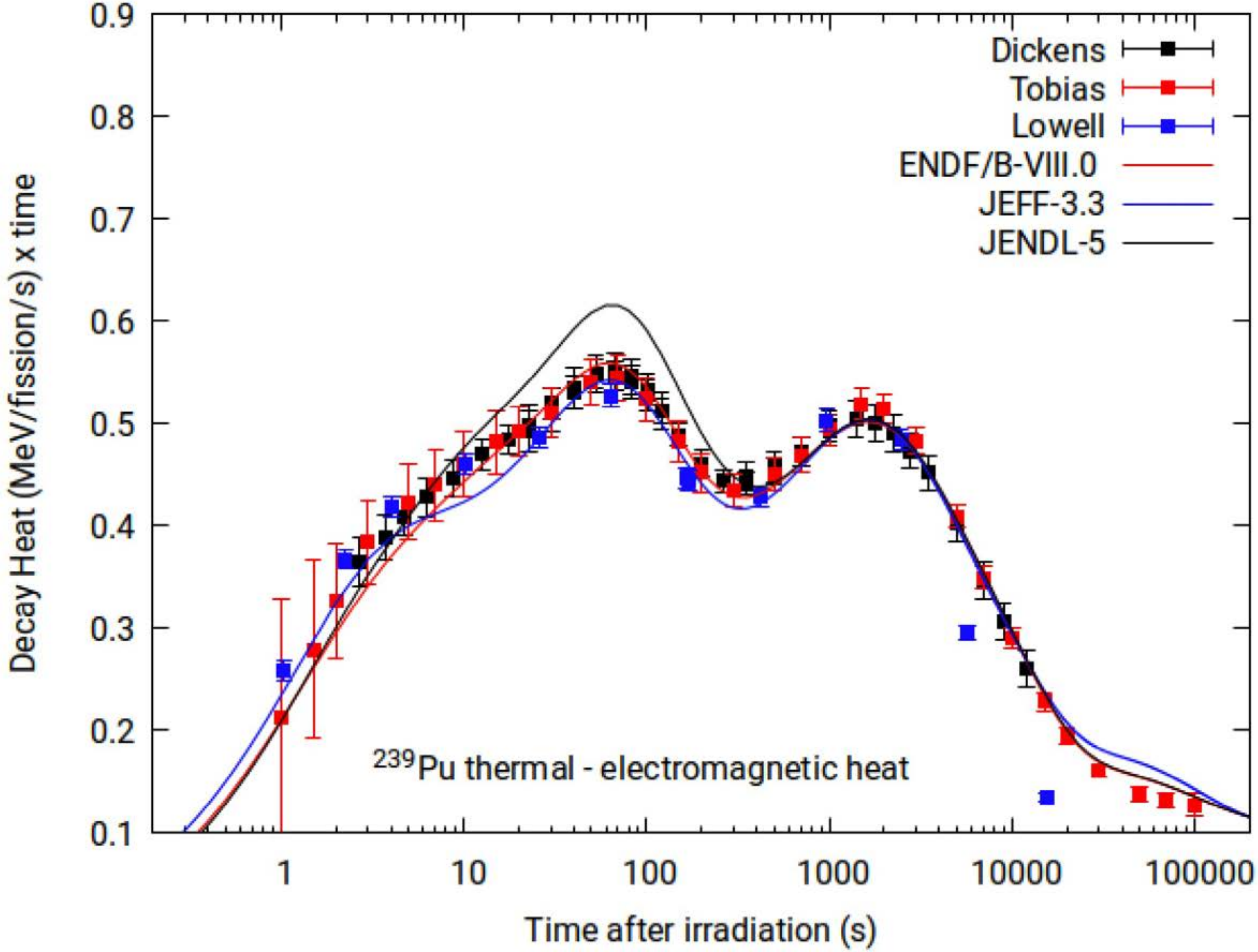
M. Estienne, M. Fallot, A. Algora, et al. PRL 123, 022502 (2019)



Subatech Calculations Estienne, Fallot, et al.

Summation is slightly better than the Hueber-Mueller conversion in the 2-5 MeV range, which dominates the flux

Impact of the measurements for ^{239}Pu

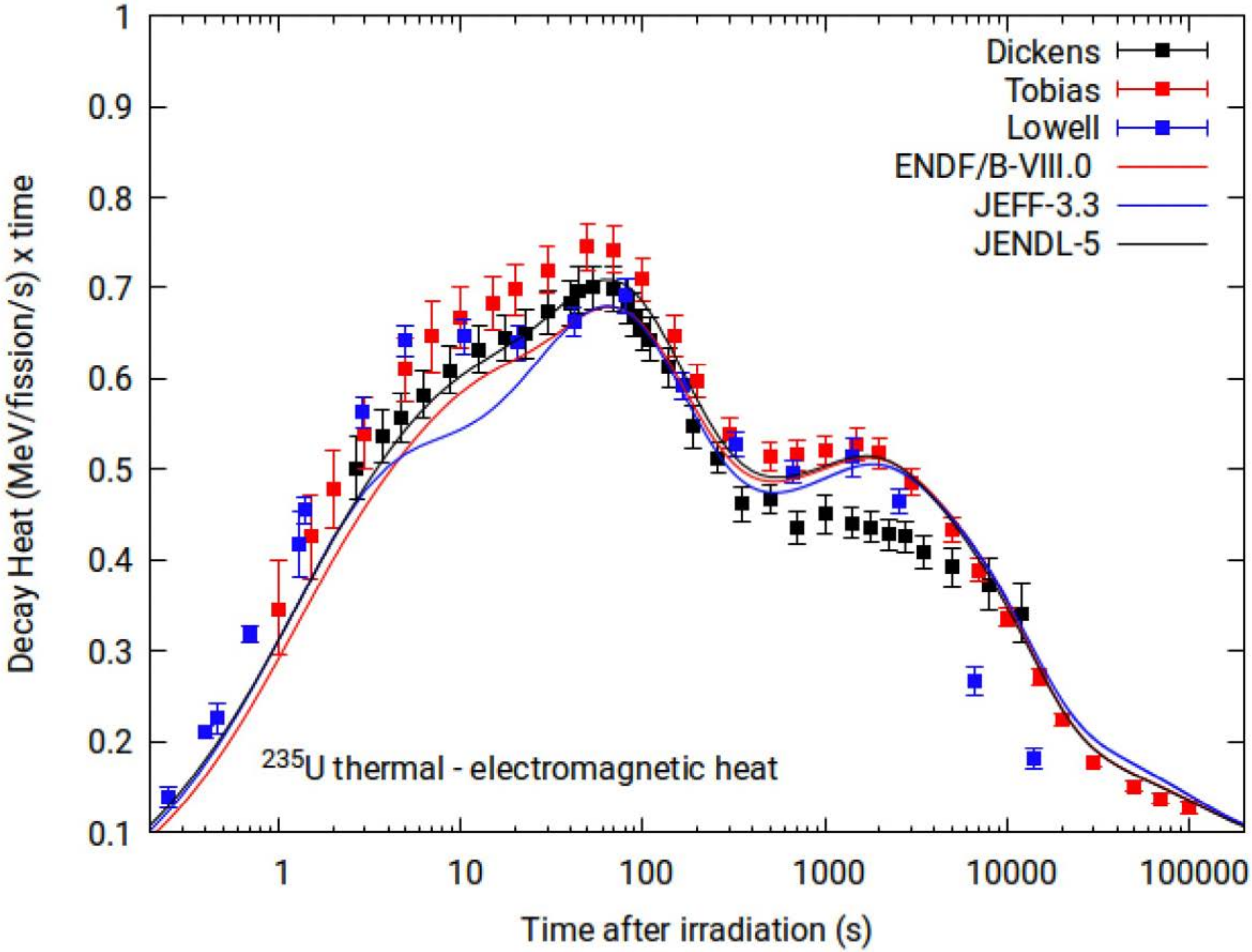


Nichols, Dimitriou, et al., Subm. to Eur. Phys. J. A

https://arxiv.org/abs/2212.10335

Courtesy of L. Giot with SERPENT

Impact of the measurements for ^{235}U

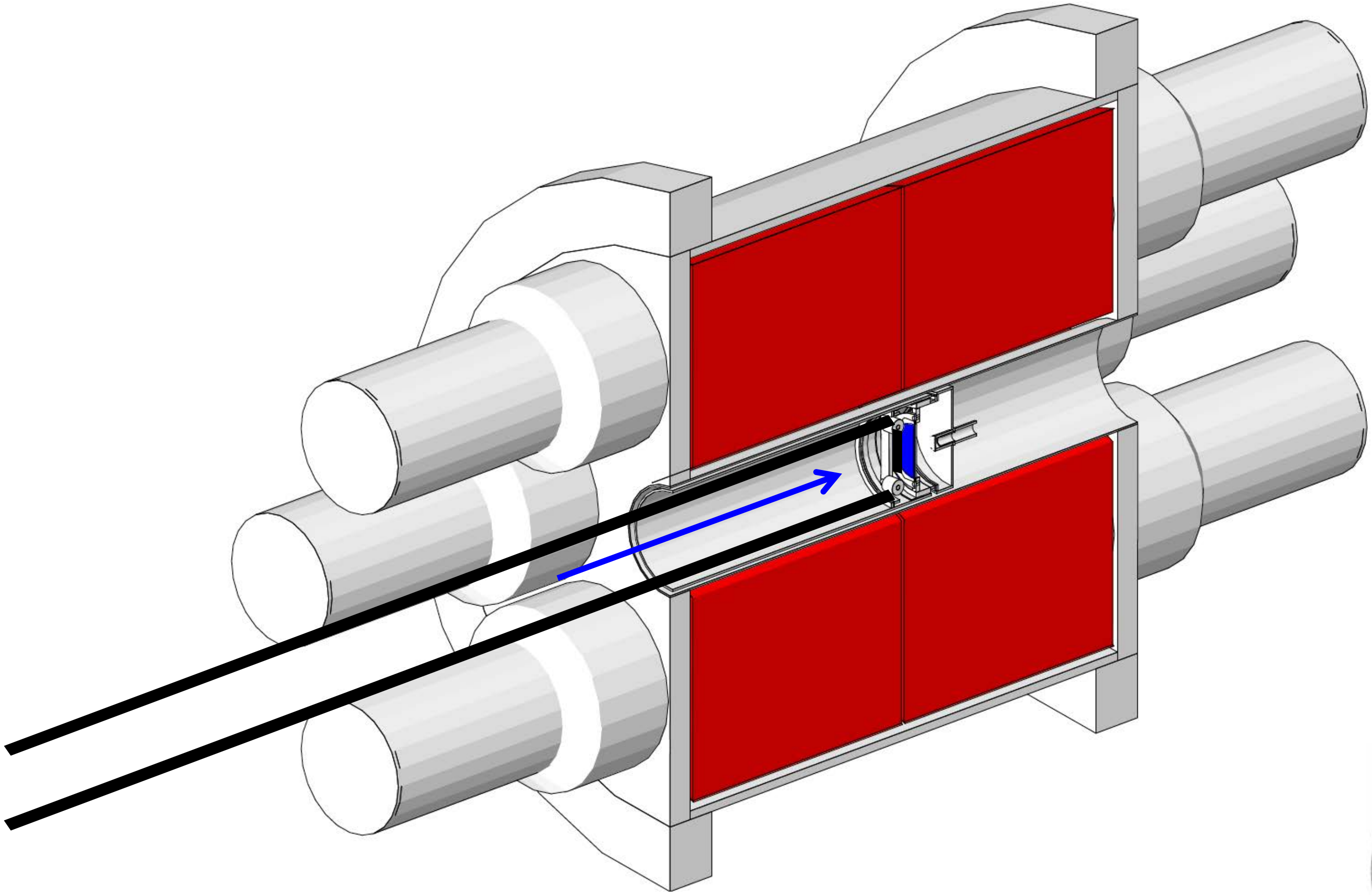


Nichols, Dimitriou, et al., Subm. to Eur. Phys. J. A

https://arxiv.org/abs/2212.10335

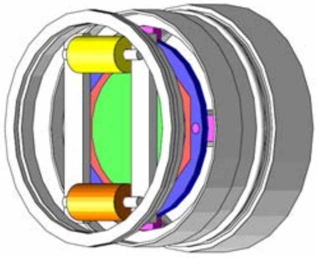
Courtesy of L. Giot with SERPENT

VTAS in Jyväskylä (November 2009)
$^{86,87,88}Br$, $^{91,92,93,94}Rb$



Segmented BaF2 detector
with optically separated crystals

Si detector endcup

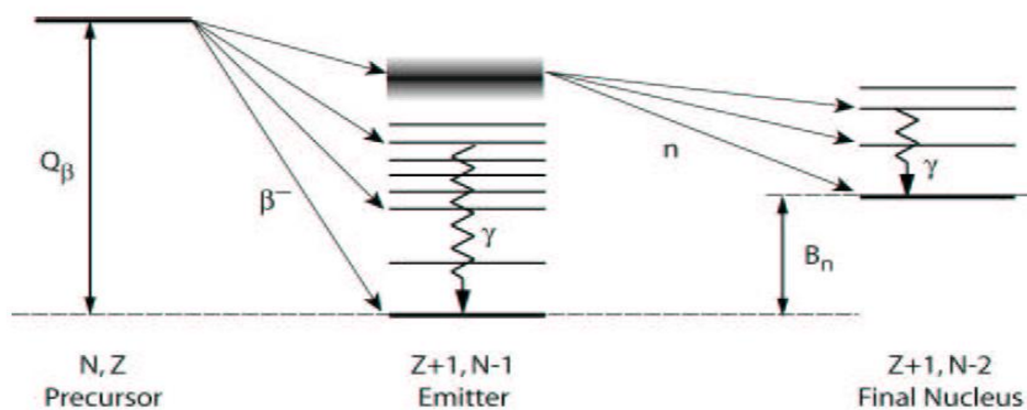


Examples from the second experiment: ^{87}Br, ^{88}Br, ^{94}Rb

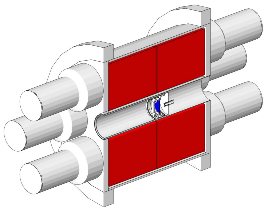
- Measure moderate beta delayed neutron emitters with complementary techniques
- Priority one in the IAEA list (decay heat)
- Moderate fission yields
- Pandemonium cases ?
- Interest from the structure point of view: vicinity of N=50 closed shell
- Competition between gamma and neutron emission above the Sn value

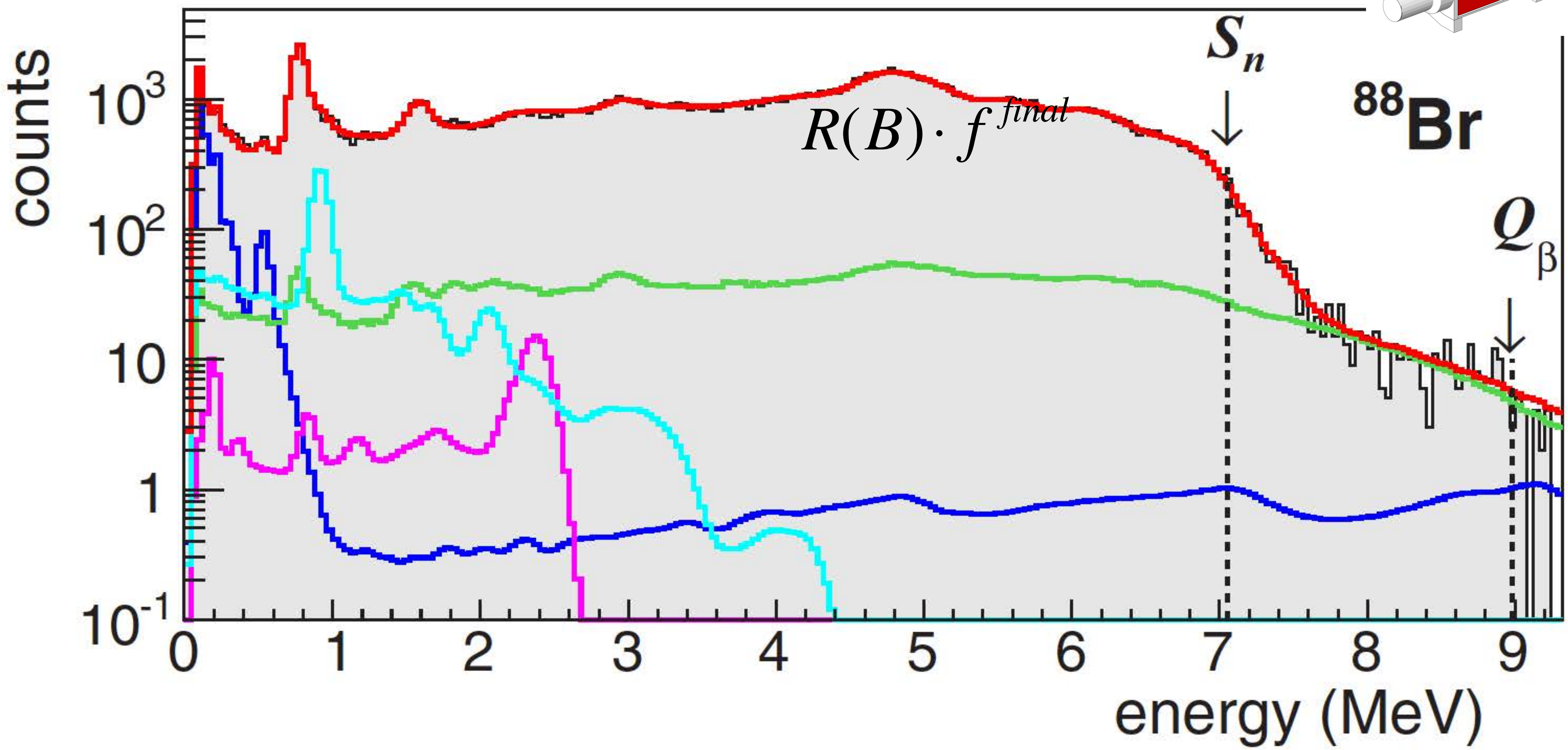
$$\frac{1}{T_{1/2}} = \int_0^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) dE_x$$

$$P_n = \frac{\int_{S_n}^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) \cdot \frac{\Gamma^n}{\Gamma^n + \Gamma^\gamma} dE}{\int_0^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) dE_x}$$



Beta delayed neutron emitters, example: ^{88}Br

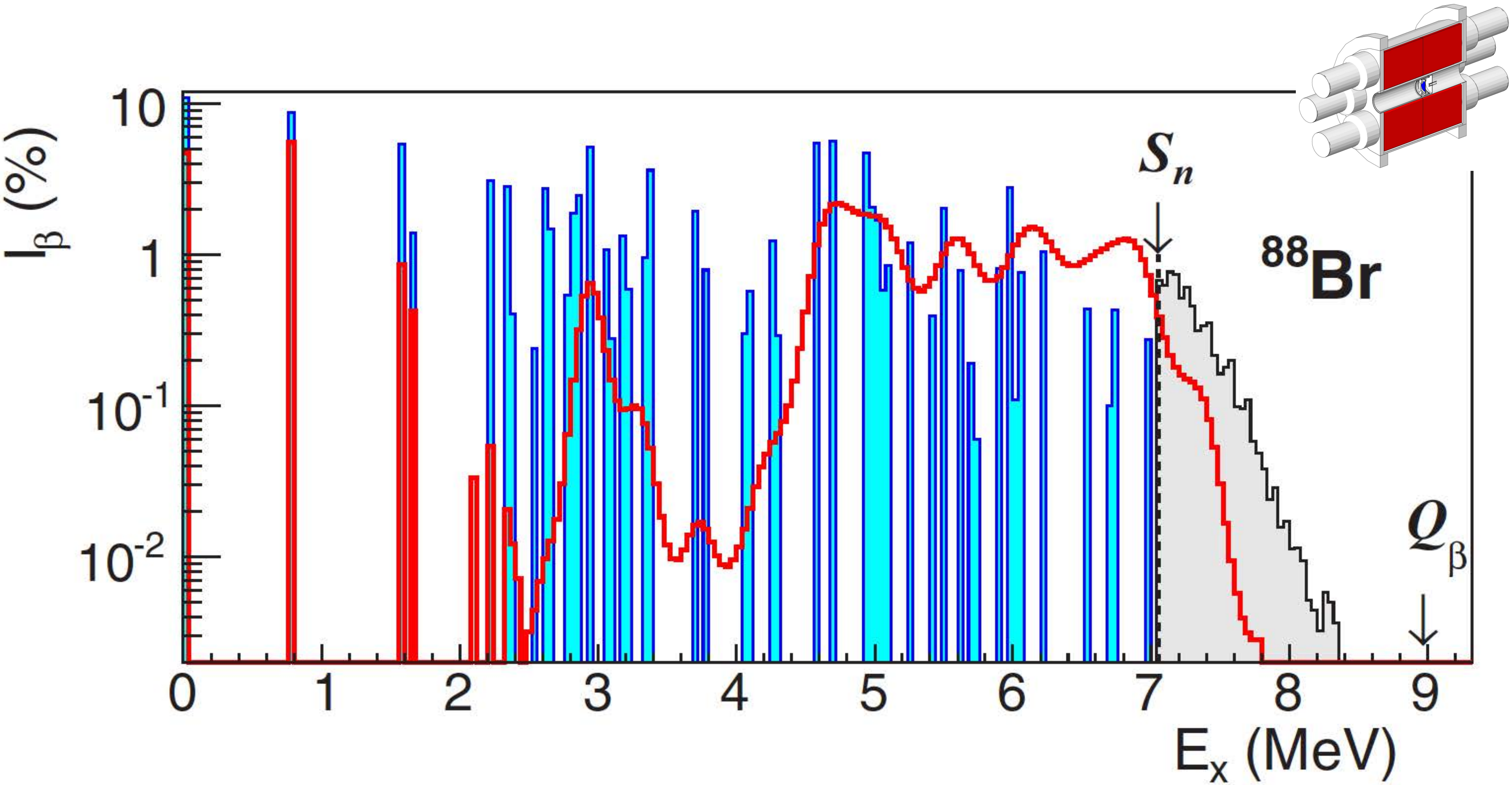




E. Valencia, et al, PRC95, 024320 (2017)
Tain et al. PRL 115, 062502

Pn=6.4 (6) %

Beta delayed neutron emitters, example: ^{88}Br

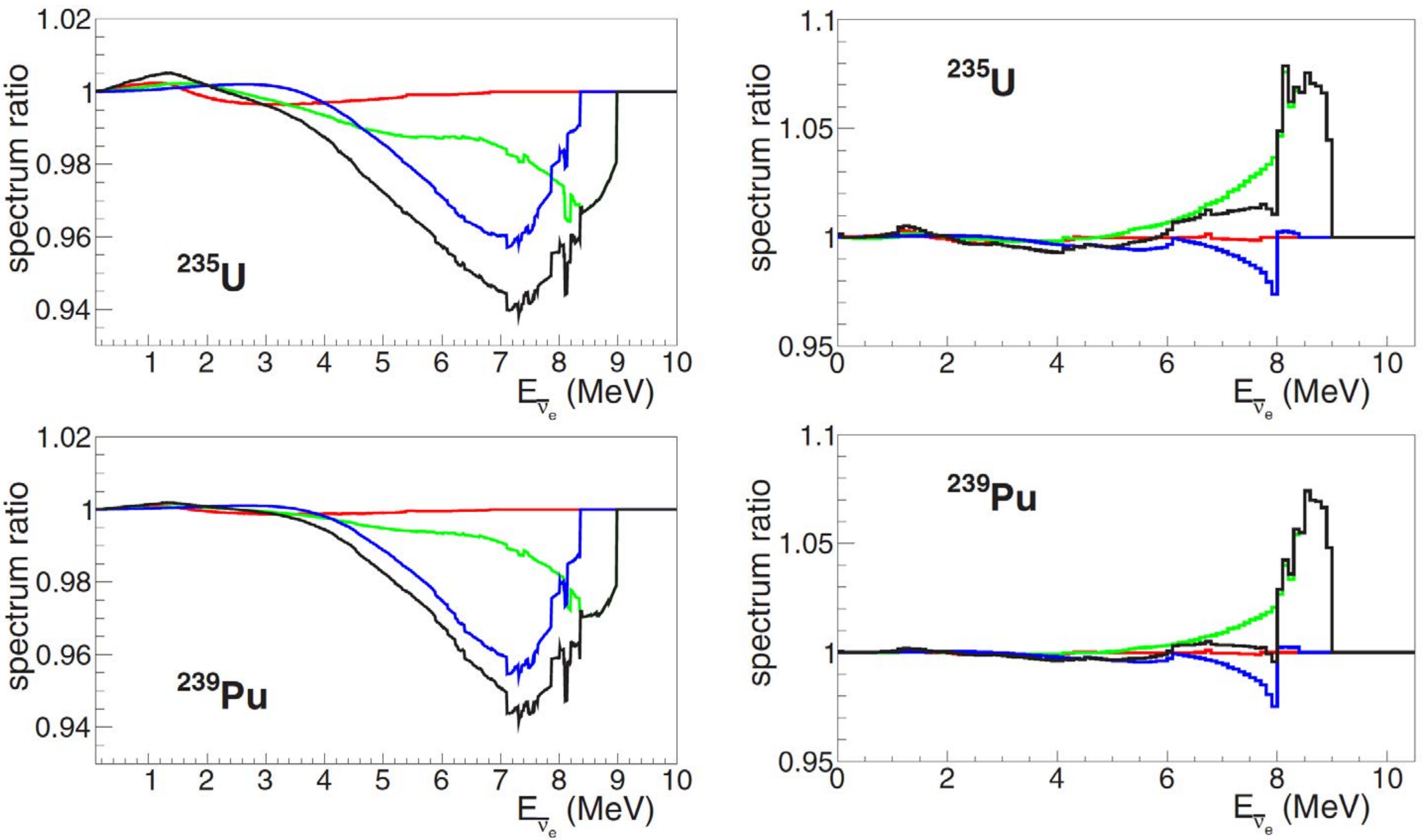


E. Valencia, et al, PRC95, 024320 (2017)
Tain et al. PRL 115, 062502

Pγ=1.59 (+27-22) %

Pn=6.4 (6) %

Antineutrino impact of ^{87,88}Br, ^{94}Rb

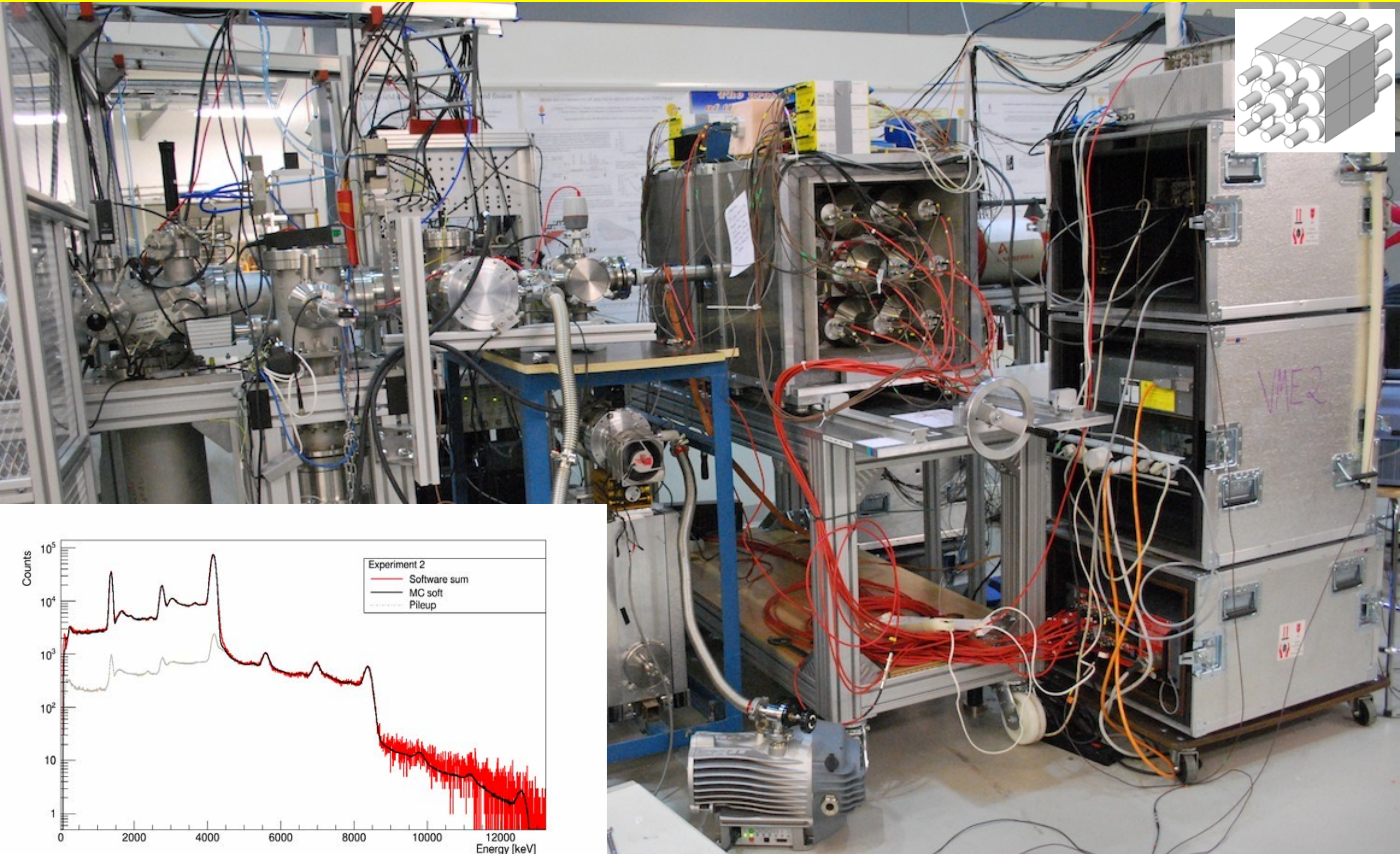


TAS vs high-resolution impact
(^{87}Br red, ^{88}Br green, ^{94}Rb blue, all black)

TAS vs Tengblad impact
Calc. courtesy of A. Sonzogni

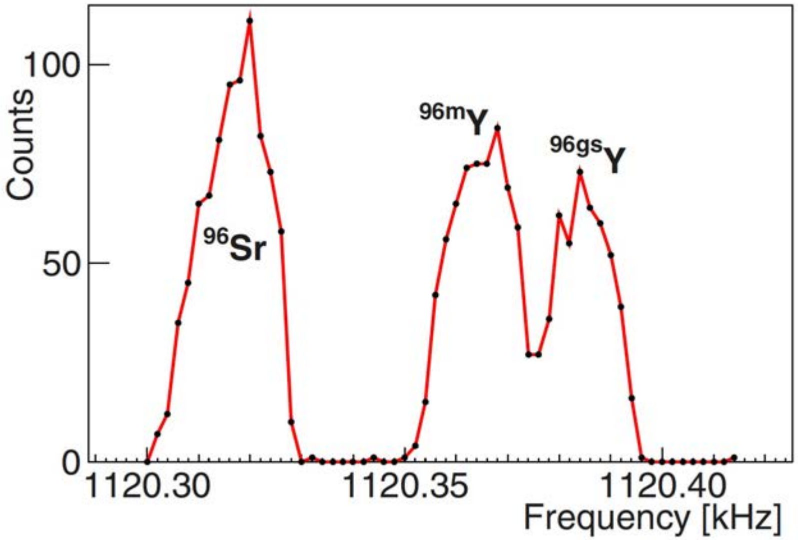
DTAS at Jyväskylä (Feb. 2014)

(Subatech-Valencia Coll., spokespersons: Fallot, Tain, Algora)



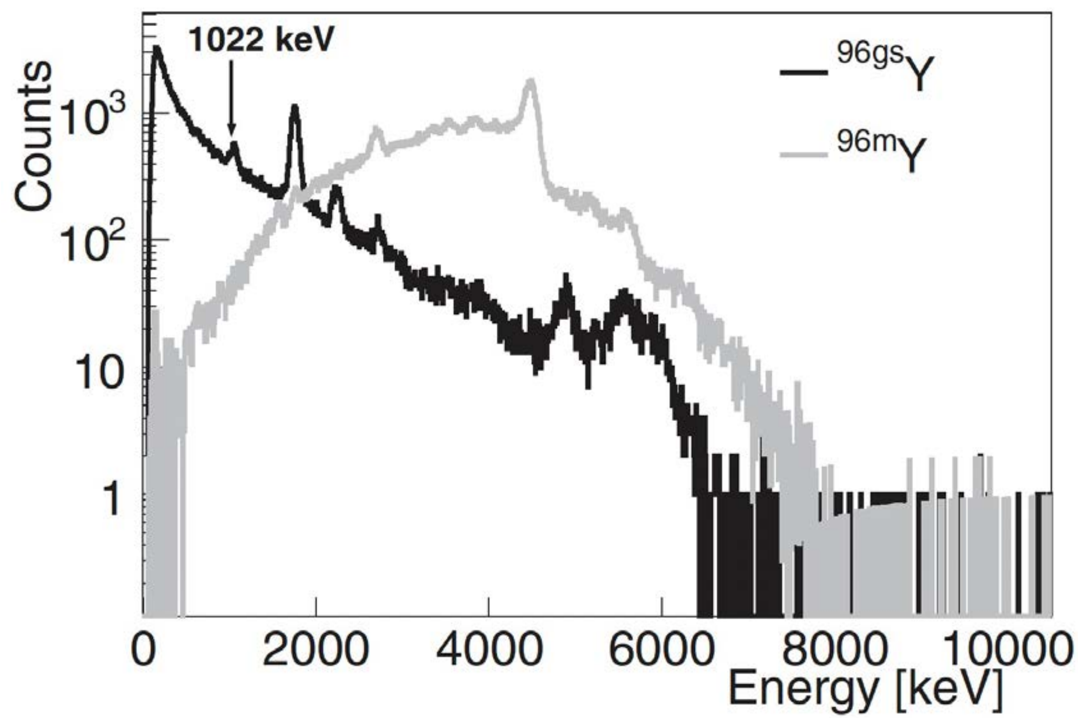
Example: the ^{96gs,96m}Y cases (from 18(+5) relevant decays measured)

^{96gs}Y decay is the second most relevant contributor to the antineutrino spectrum



JYFL Penning trap scan, A=96

TAGS spectra for 96Y isomers



CFY of the order of 4% and ~2 % respectively for 235U; and 2% for 239Pu for both isomers. Major contributor to the spectrum (11% for the 5-6 MeV bin)

Challenge: E0 transition from 1581 keV level in the 96gsY case

Requirement: Special treatment of the response function, because conversion electrons + pair production

Guadilla et al, PRC 106, 014306 (2022)

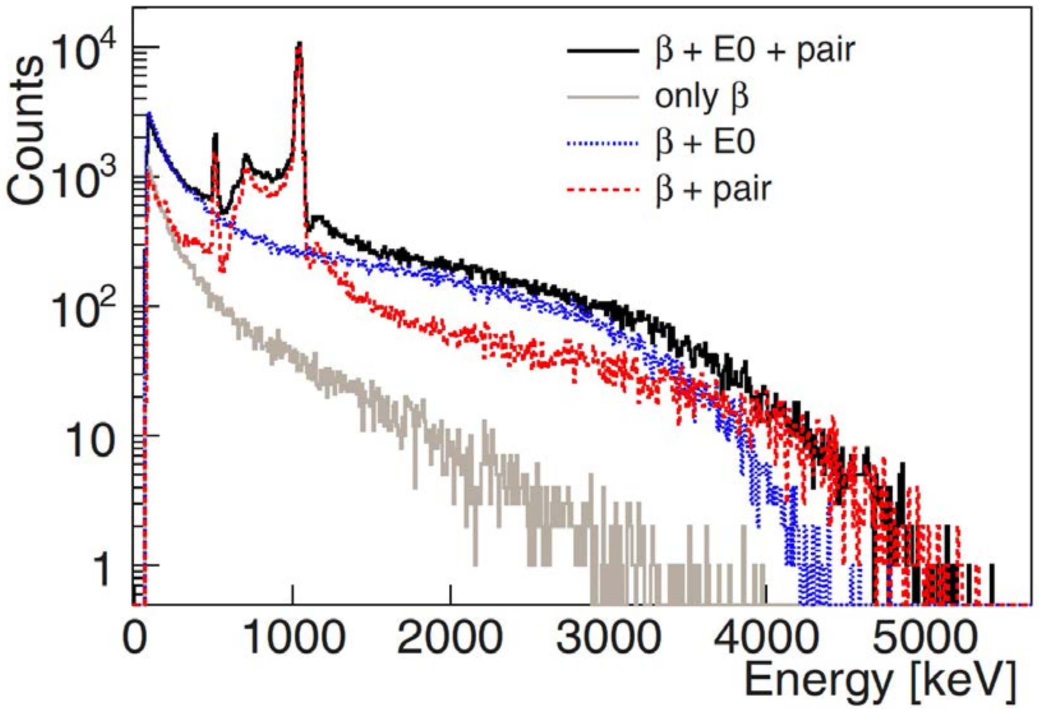
The E0 challenge in the response

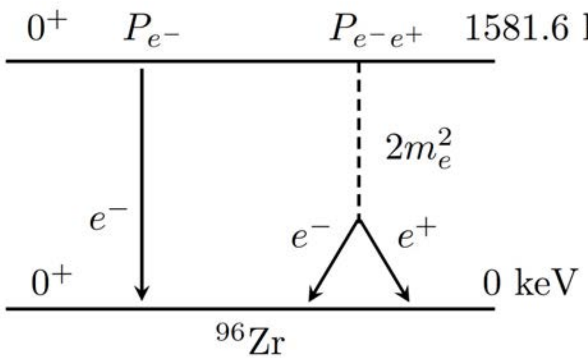
$$d = R(B) \cdot f$$

$$R_j^{\beta-} = b^- \otimes r_k$$

$$r_j = \sum_{k=0}^{j-1} b_{jk} g_{jk} \otimes r_k$$

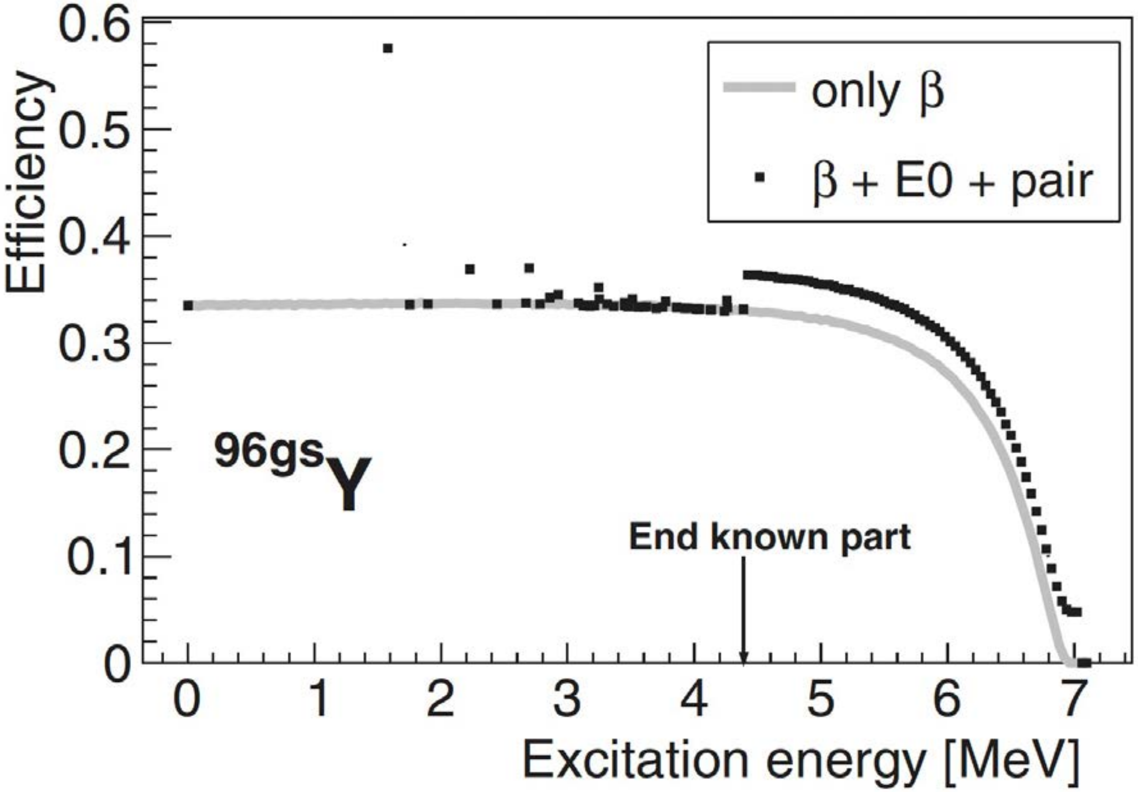
$$g_{jk} \rightarrow \left(\frac{1}{1+\alpha_{jk}^{tot}} g_{jk} + \frac{\alpha_{jk}^{tot}}{1+\alpha_{jk}^{tot}} e_{jk}^{k} \otimes x + \ldots\right)$$

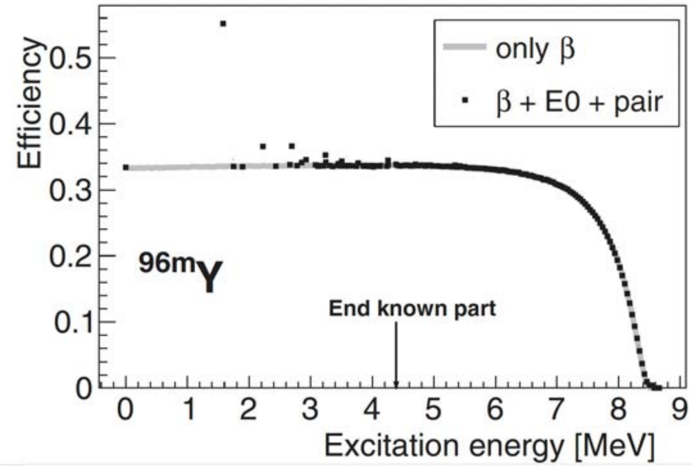




Guadilla et al, PRC 106, 014306 (2022)

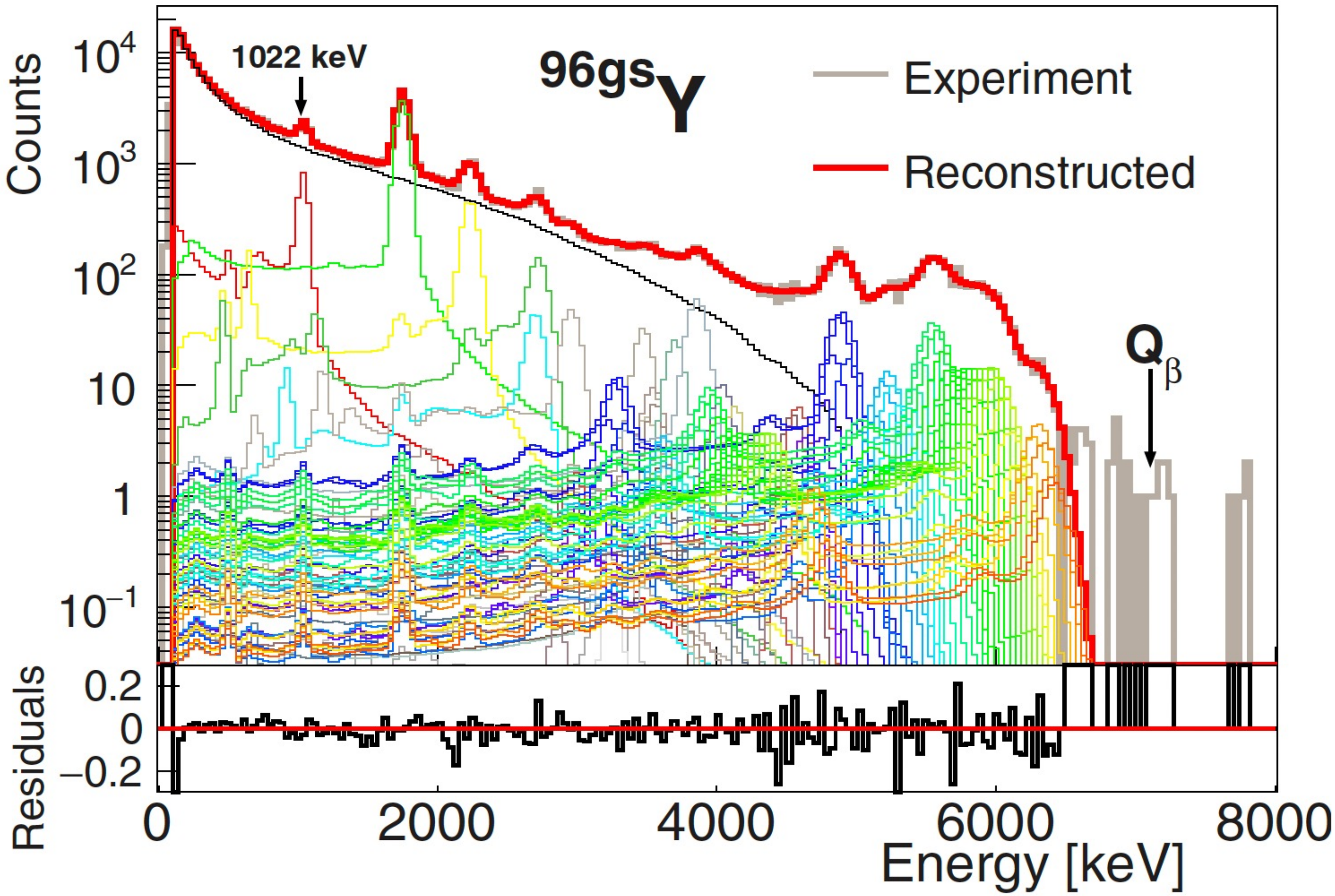
Impact on the beta efficiency





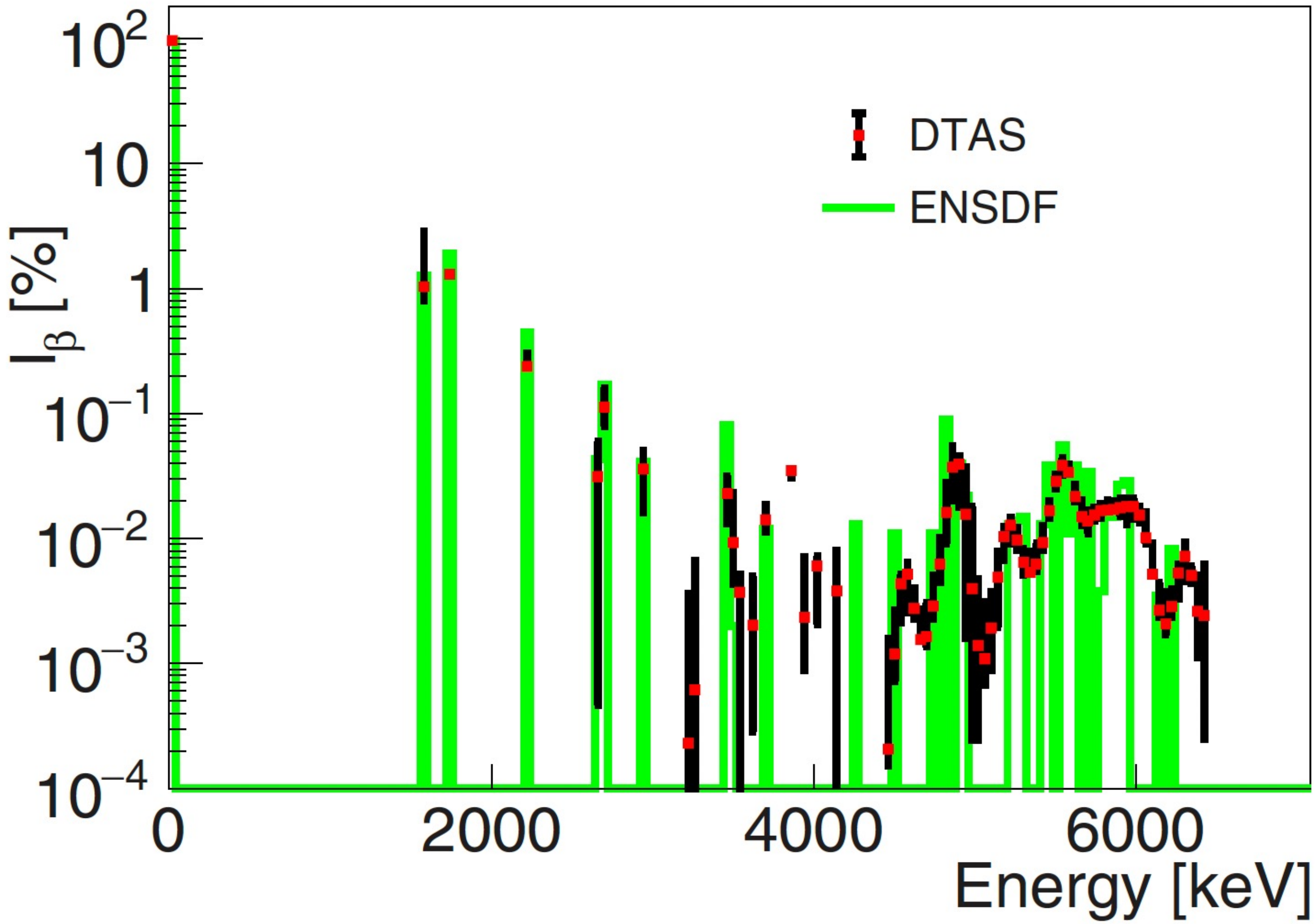
Guadilla et al, PRC 106, 014306 (2022)

TAGS spectrum and analysis for 96gsY



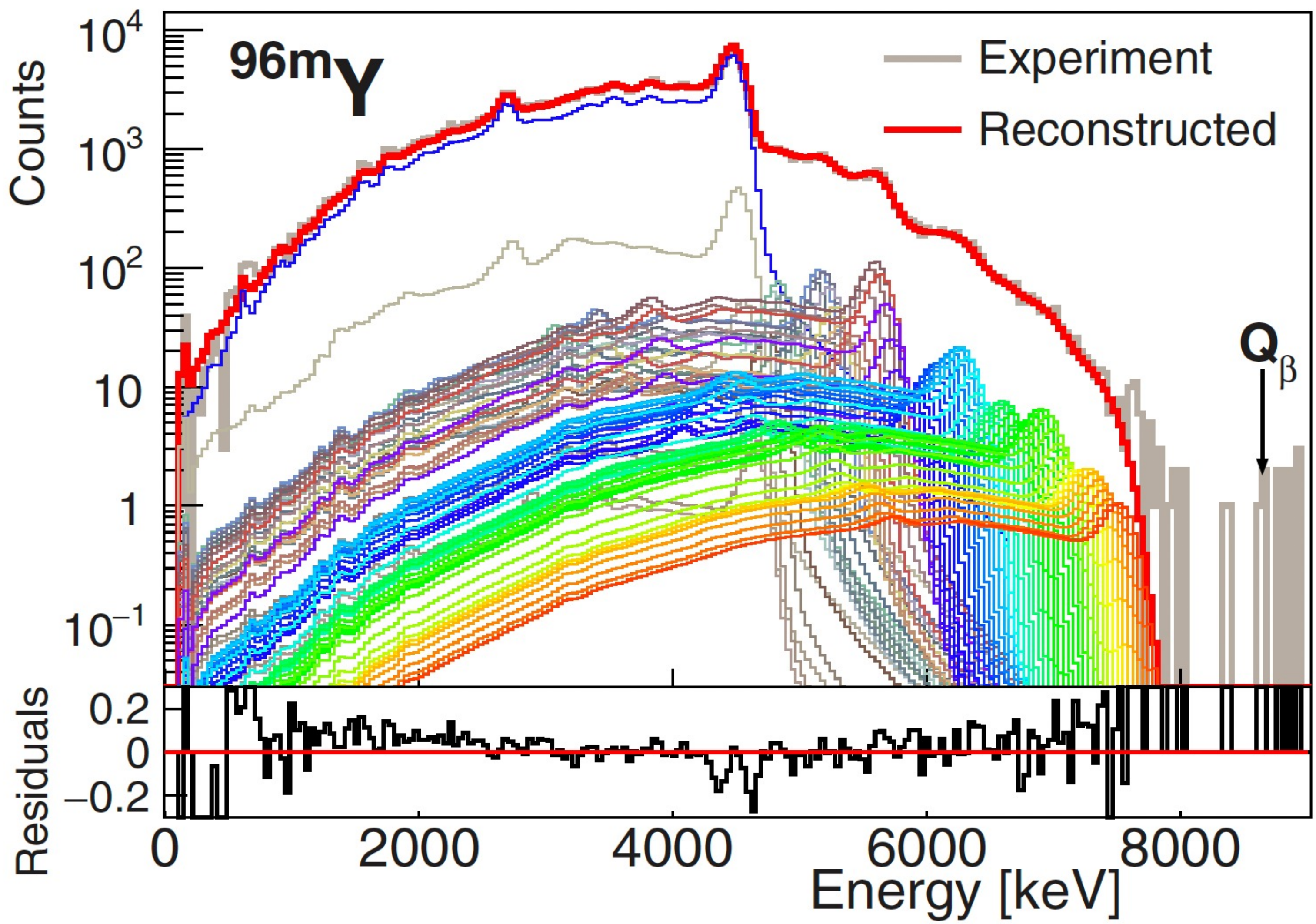
Guadilla et al, PRC 106, 014306 (2022)

Feeding distribution for 96gsY



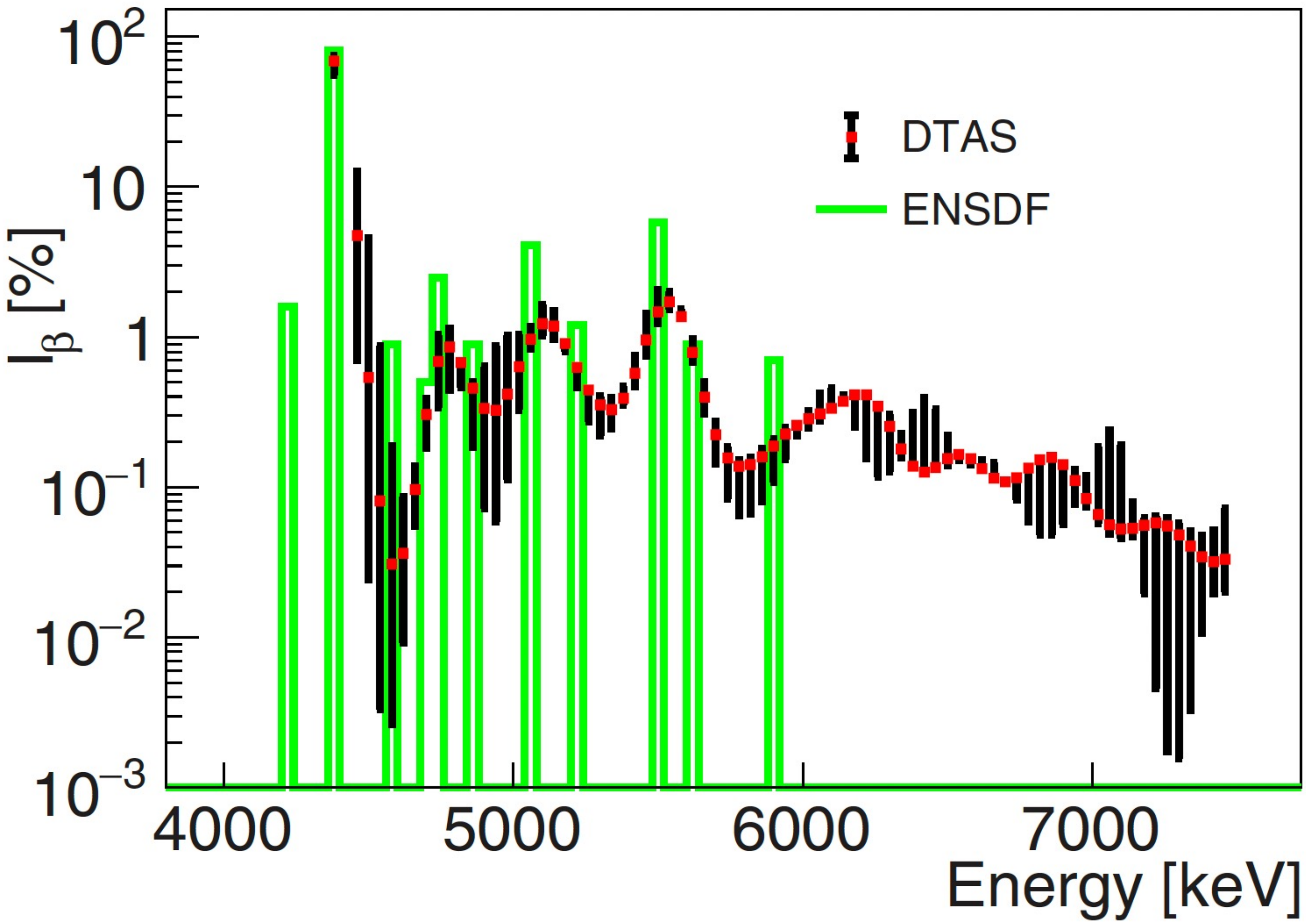
Guadilla et al, PRC 106, 014306 (2022)

TAGS spectrum and analysis 96mY



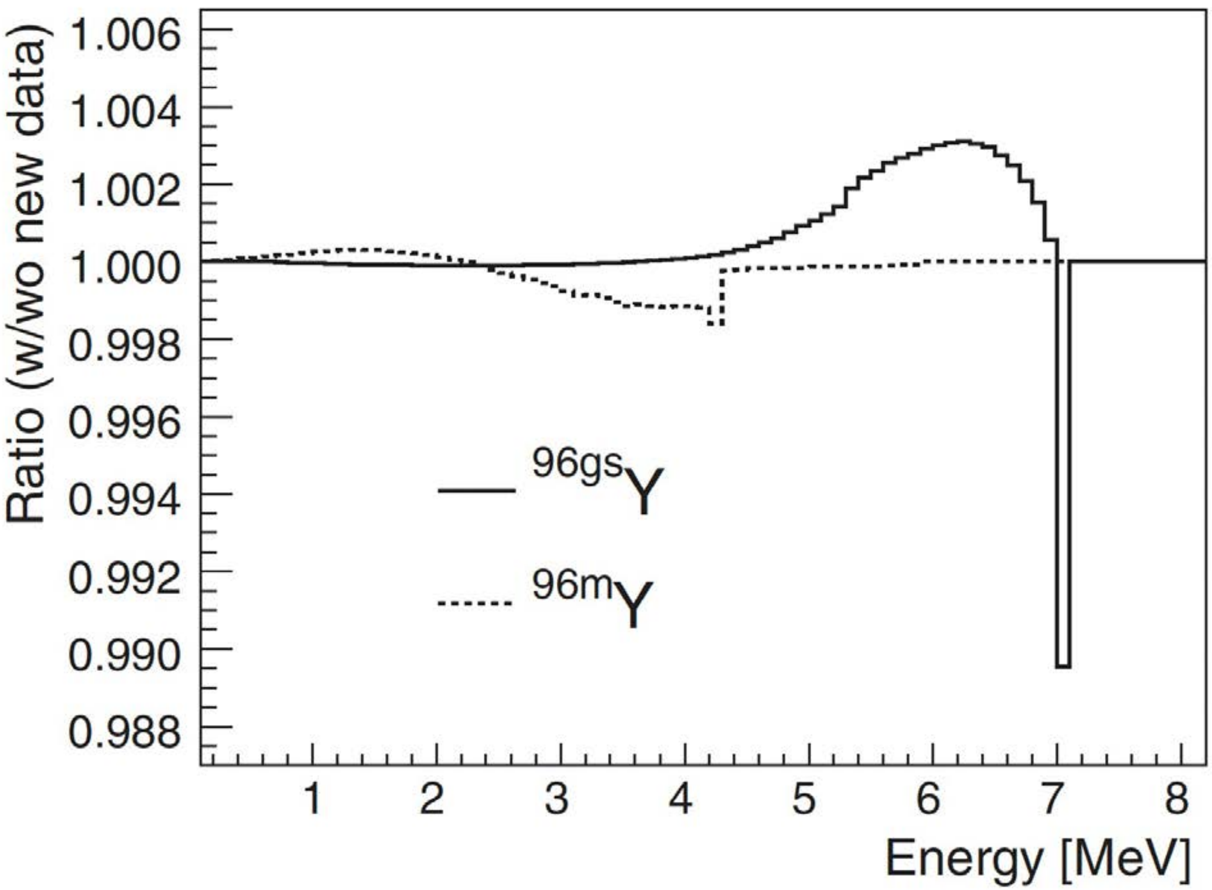
Guadilla et al, PRC 106, 014306 (2022)

Feeding distribution 96mY



Guadilla et al, PRC 106, 014306 (2022)

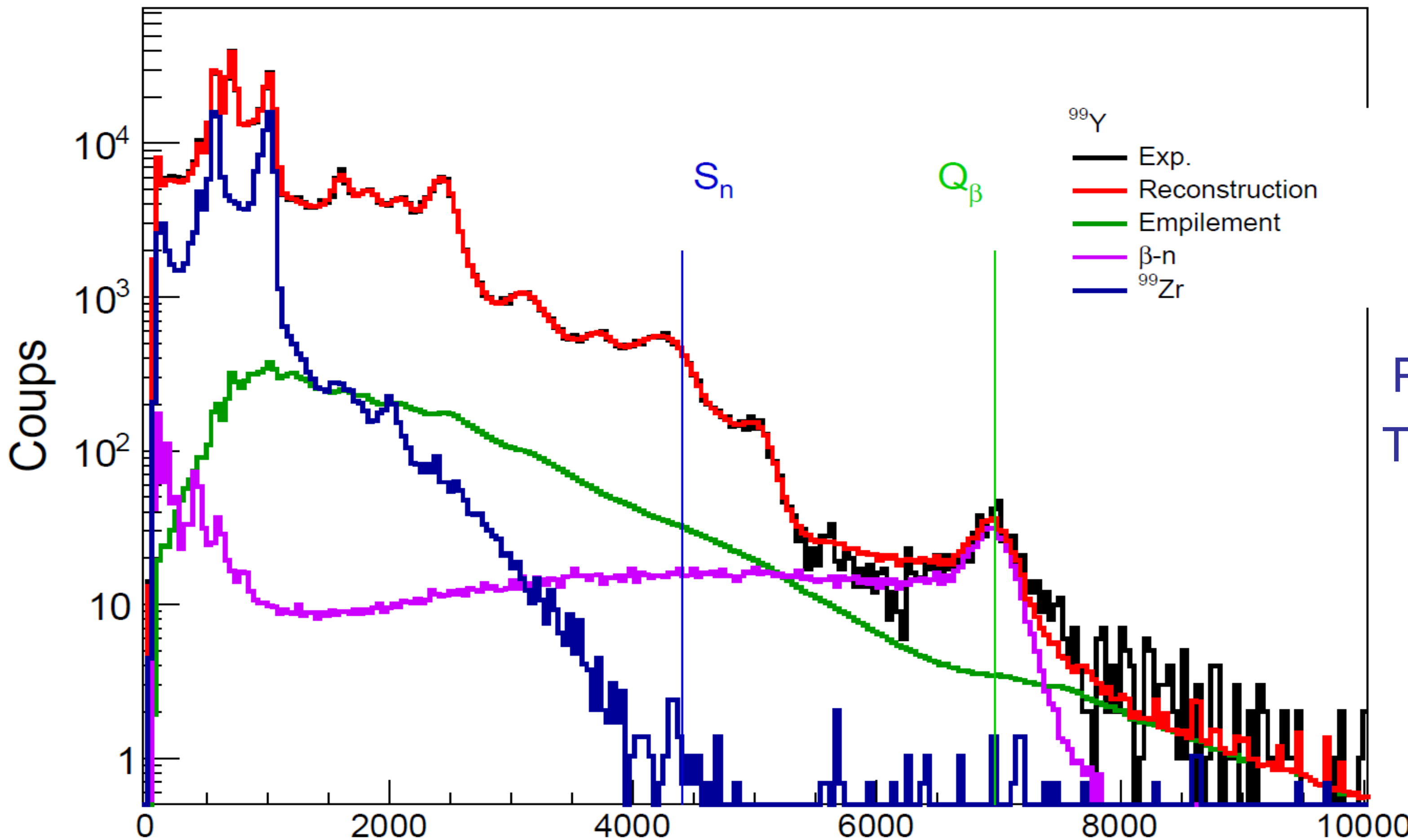
Neutrino impact of the new results



Impact of the new TAGS data with respect to the values used in the summation calculation model. Previosly for 96gsY Rudstam data was used and for 96mY JEFF3.3 data was employed.

Guadilla et al, PRC 106, 014306 (2022)

Another example: ^{99}Y Nantes analysis

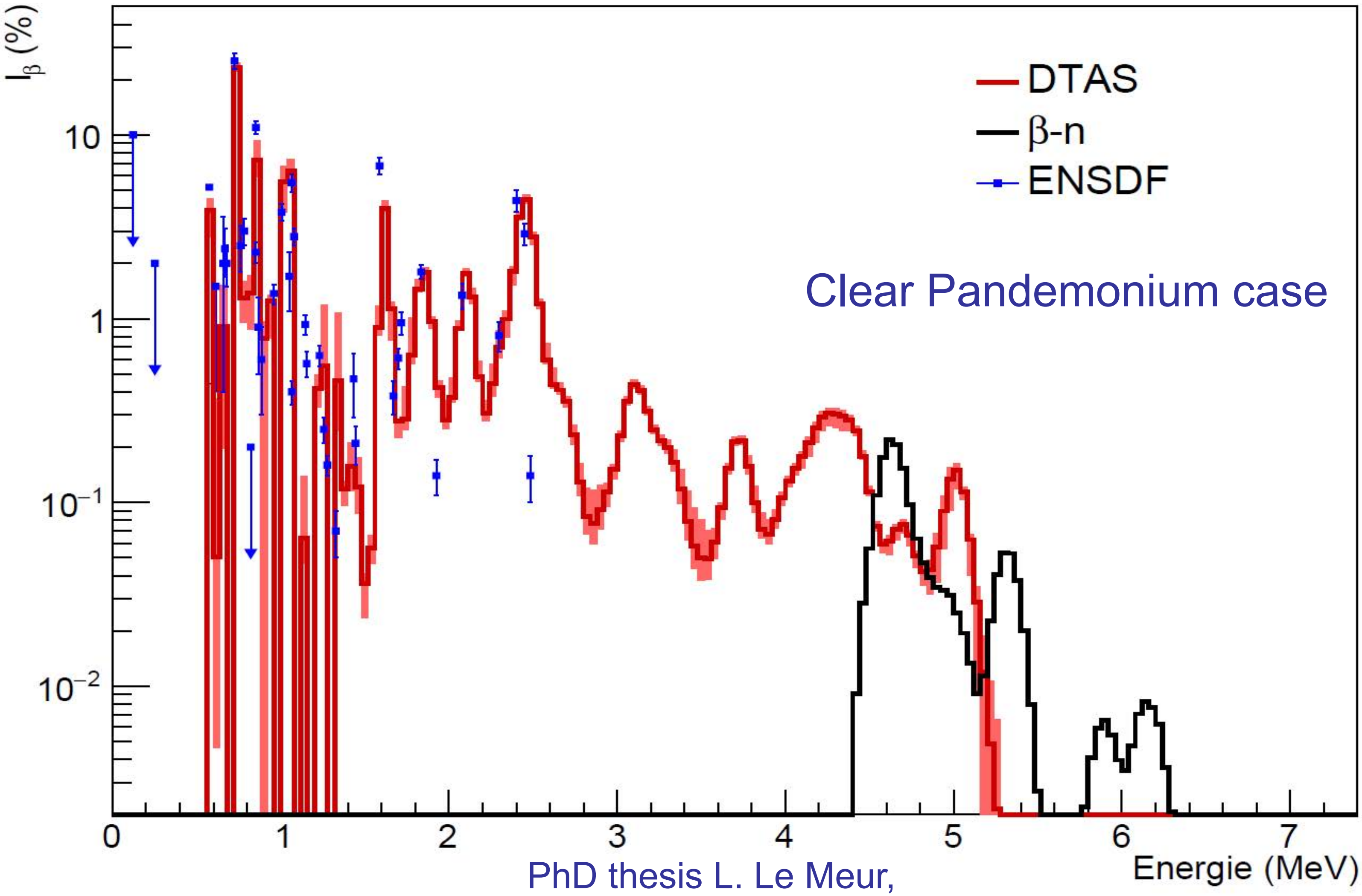


Pn= 2.5(5)

$T_{1/2}$= 1.48 s

PhD thesis L. Le Meur, Subatech, Nantes

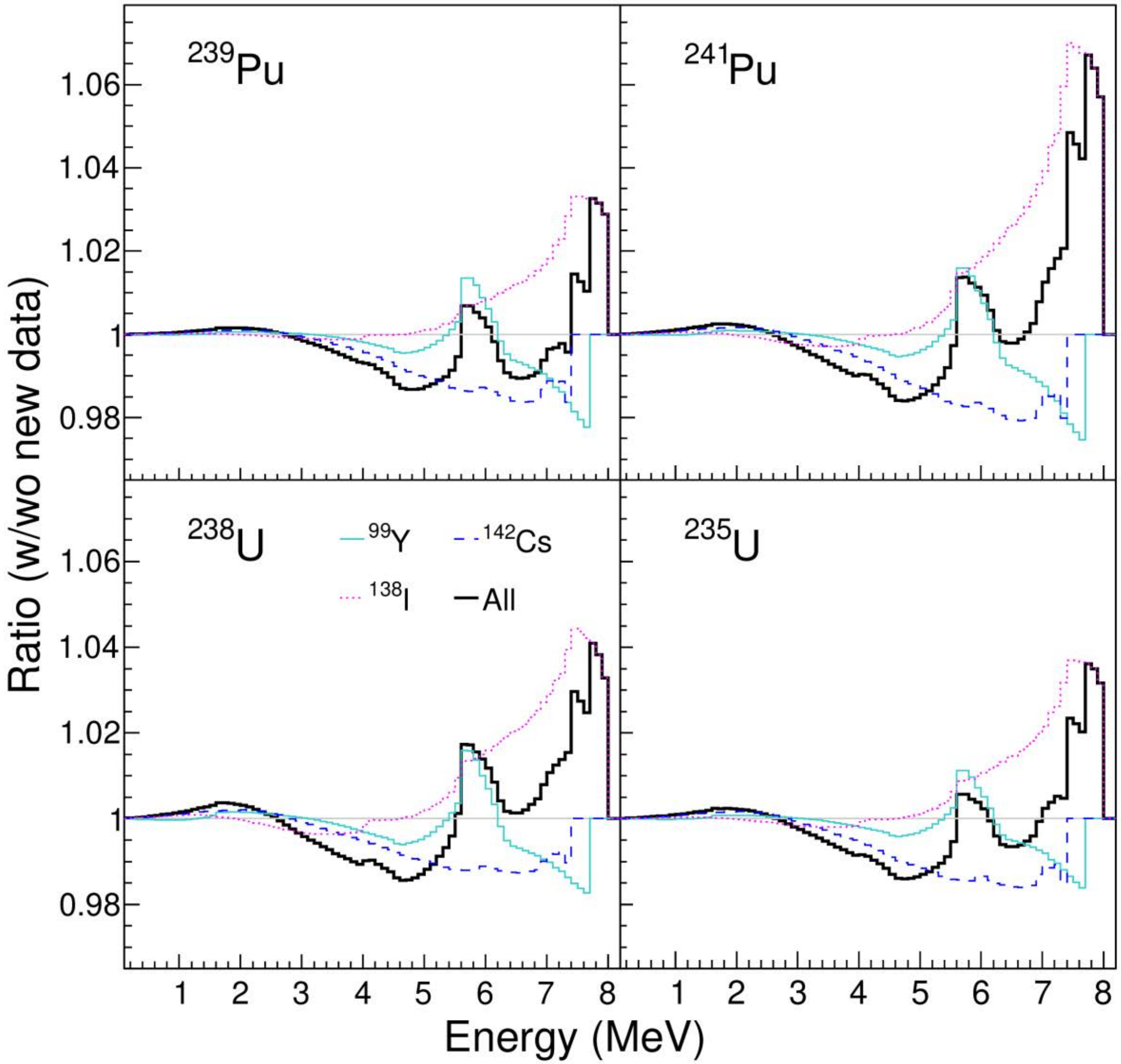
Another example: ^{99}Y
Nantes analysis



PhD thesis L. Le Meur,
Subatech, Nantes

Combined impact of ^{99}Y, ^{138}I, ^{142}Cs Analyses by Nantes

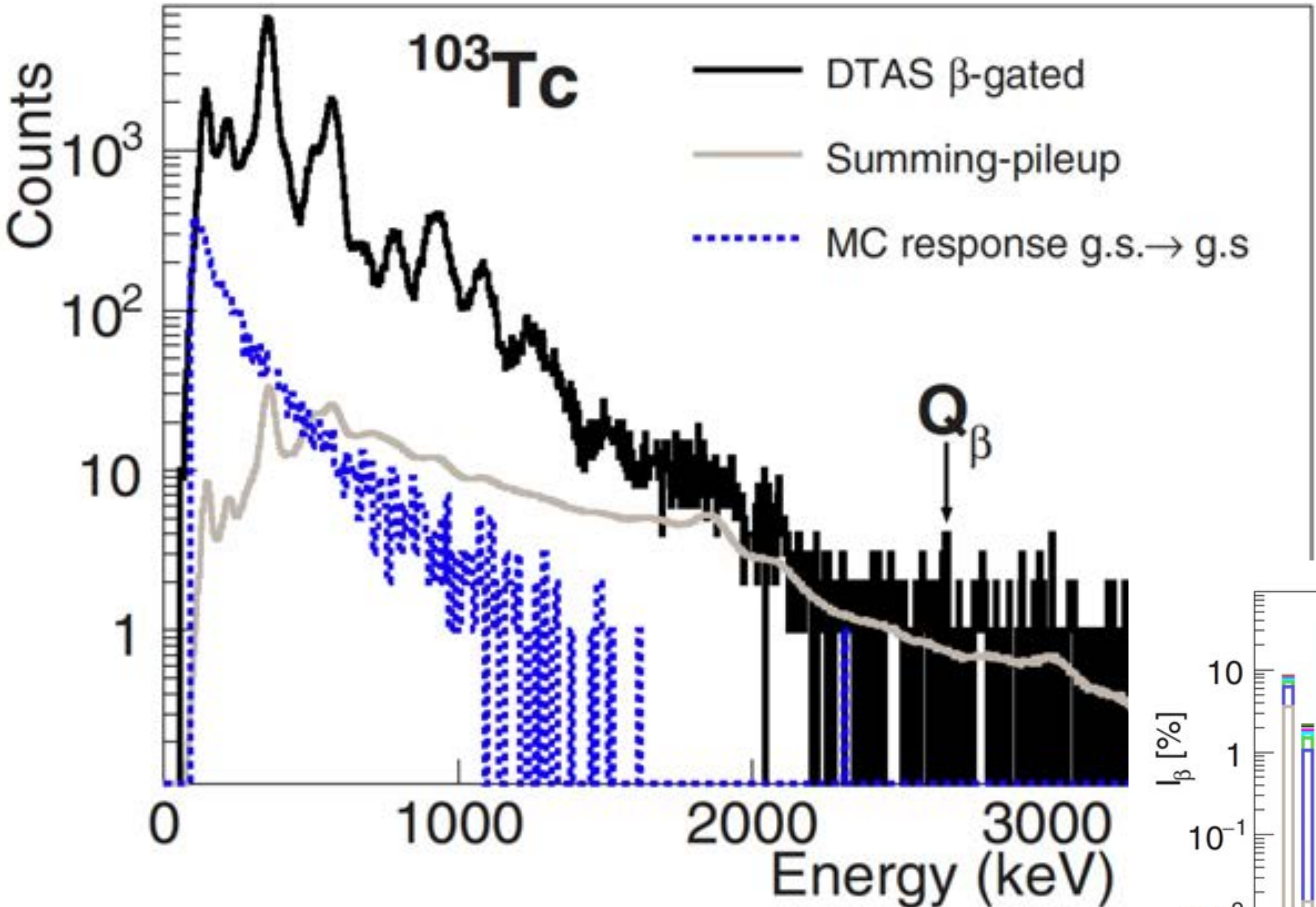
~1.5% impact in the region of the spectral distorsion



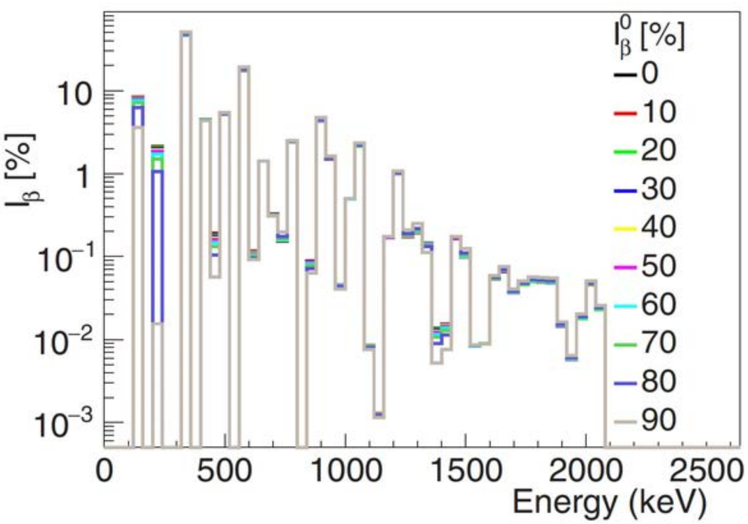
Calculation by M. Estienne

^{103}Tc decay (an odd TAGS case)
Bad luck or serendipity?

$^{103}_{43}Tc \rightarrow \, ^{103}_{44}Ru$



TAGS analysis insensitive to the assumed ground state feeding



Guadilla et al. PRC 102, 064304 (2020)

A new method for determining the gs to gs feeding

Based on a comparison of the number of counts detected in the beta detector (N_β) with the number of counts detected in the TAS in coincidence with the betas ($N_{\beta\gamma}$)

$$I_\beta^0 = \frac{1 - \frac{N_{\beta\gamma}}{N_\beta}\frac{\bar{\varepsilon}_\beta^*}{\bar{\varepsilon}_{\beta\gamma}^*}}{1 + \frac{N_{\beta\gamma}}{N_\beta}\frac{\varepsilon_\beta^0 - \bar{\varepsilon}_\beta^*}{\bar{\varepsilon}_{\beta\gamma}^*} - \frac{\varepsilon_{\beta\gamma}^0}{\bar{\varepsilon}_{\beta\gamma}^*}}.$$

ε_β^*, $\varepsilon_{\beta\gamma}^*$ are average efficiencies to excited states

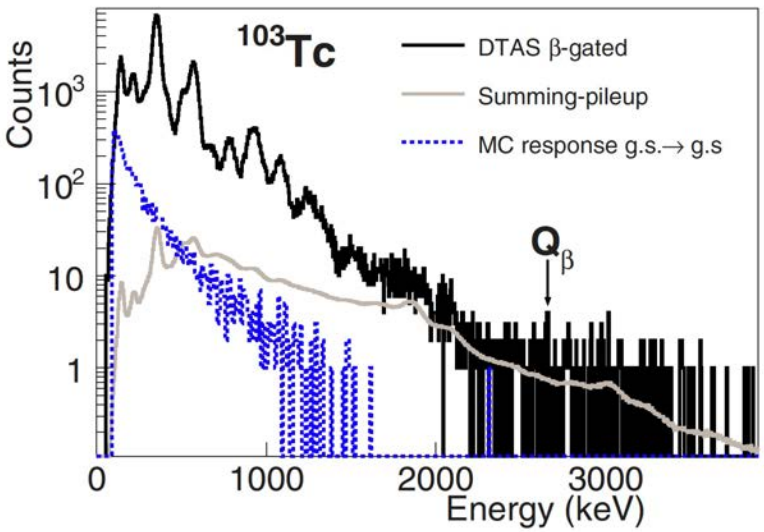
ε_β^0, $\varepsilon_{\beta\gamma}^0$ average efficiencies to gs

Corrected form in comparison with the earlier work of Greenwood et al. NIM A317 (1992) 175

The method was tested with synthetic data.

Guadilla et al. PRC 102, 064304 (2020)

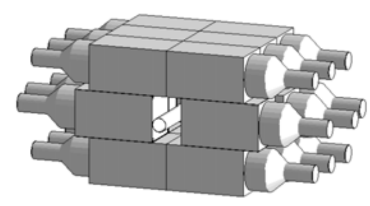
$${}^{103}_{43}Tc \rightarrow {}^{103}_{44}Ru$$



$I_\beta^0(^{103}Tc) = 45.6(+1.5\text{-}0.9)$

ENSDF value is 34(8)

Ground state feedings obtained the new method



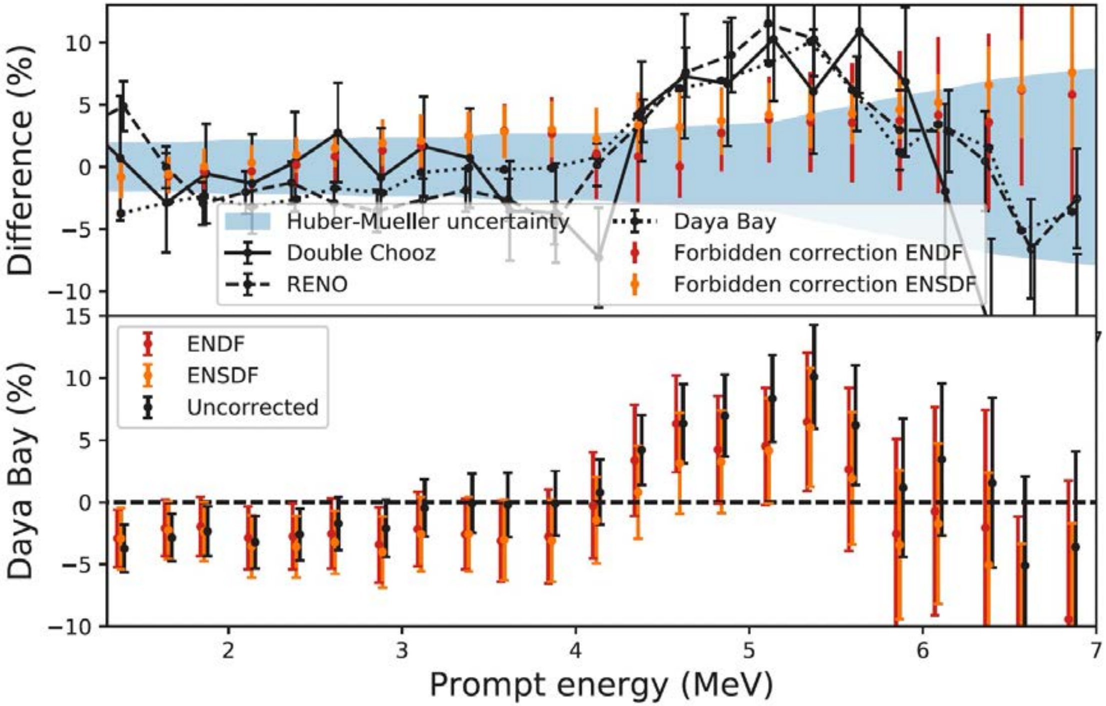
	I_β^0 (%)		
Isotope	ENSDF	TAGS	$4\pi\gamma-\beta$
^{95}Rb	$\leqslant 0.1$	$0.03^{+0.11}_{-0.02}$	$-0.2(42)$
^{100gs}Nb	50(7)	46^{+16}_{-15}	40(6)
^{102m}Nb	—	$42.5^{+9.3}_{-10.0}$	44.3(28)
^{100}Tc	93.3(1)	93.9(5)	92.8(5)
^{103}Tc[a]	34(8)	—	$45.6^{+1.5}_{-0.9}$
^{137}I	45.2(5)	$50.8^{+2.7}_{-4.3}$	45.8(13)
^{140}Cs	35.9(17)	$39.0^{+2.4}_{-6.3}$	36.0(15)

[a]For this decay the I_β^0 numbers include the intensity to the first excited state in ^{103}Ru at 2.81(5) keV.

Reduced uncertainties and consistency with the TAGS results

Guadilla et al. PRC 102, 064304 (2020)

Another relevant problem: the spectrum shape



L. Hayen et al., PRC 99.031301 (2019)

See also:

Hayes et al., 2013

Li, Zhang, 2019

L. Hayen et al., PRC 99.031301 (2019)

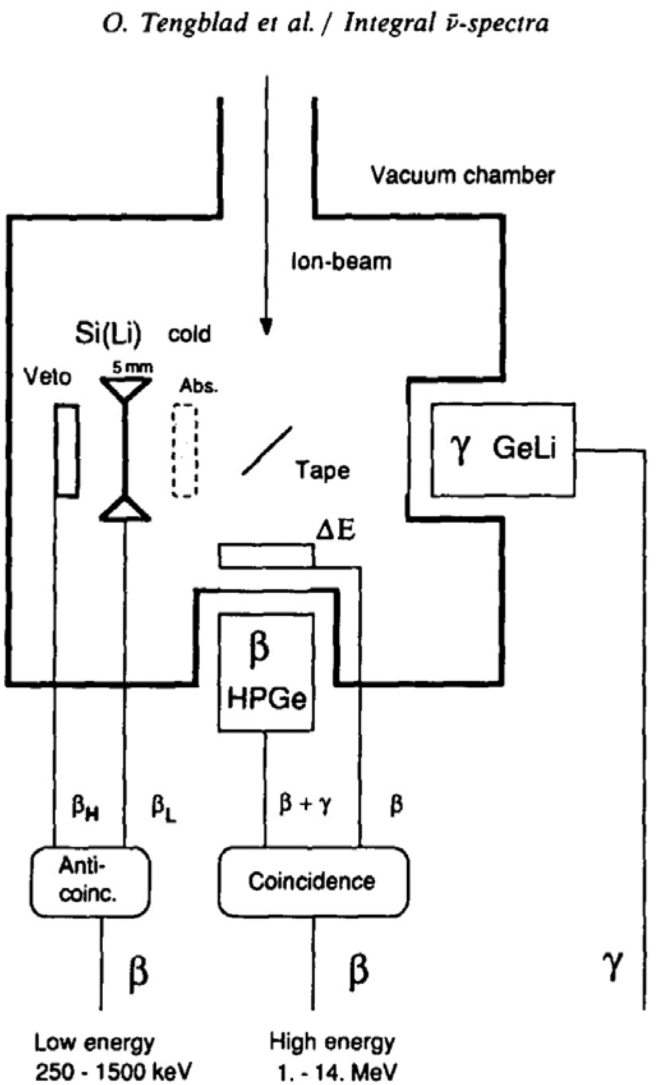
$$\frac{dN}{dW} = pW(W - W_0)^2 F(Z, W) C(Z, W) K(Z, W),$$

$$C_{0^-} = 1 + \frac{2R}{3W} b + \mathcal{O}(\alpha Z R, W_0 R^2),$$

$$C_{1^-} = 1 + aW + \mu_1 \gamma \frac{b}{W} + cW^2,$$

FIG. 5. Top panel: Normalized spectral ratios for three modern experiments relative to the Huber-Mueller predictions [2], and the normalized forbidden spectrum correction described in this work using ENDF and ENSDF decay libraries. The prompt energy of the positron emerging from the inverse β decay is related to the antineutrino energy via $E_{\text{prompt}} \approx E_\nu - 0.782$ MeV. Bottom panel: Difference between Daya Bay spectral data and different theoretical models. Error bars are calculated using experimental, H-M, and forbidden uncertainties and are assumed uncorrelated. Here Uncorrected is relative to the H-M estimate shown in the top panel, and ENDF and ENSDF are the new results.

β-shape measurements Tengblad data



Tengblad et al NPA 503 (1989) 136

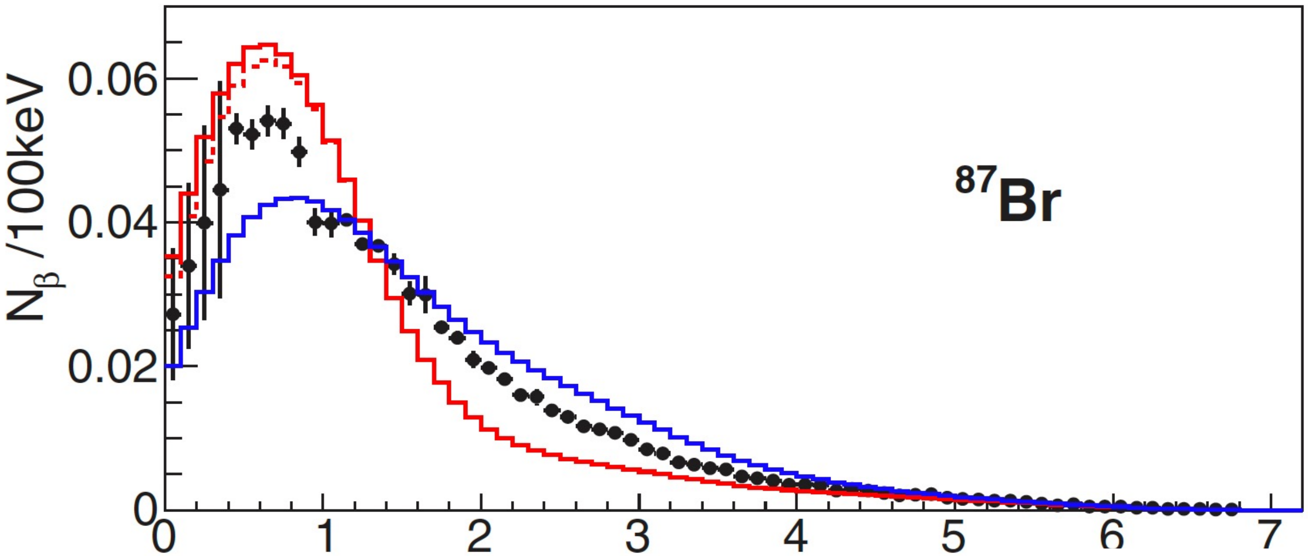
Impressive work: 111 measured beta spectra and deduced the corresponding antineutrino spectrum

Measurements performed at ISOLDE and at OSIRIS on-line isotope separators, not always optimal isotope separation

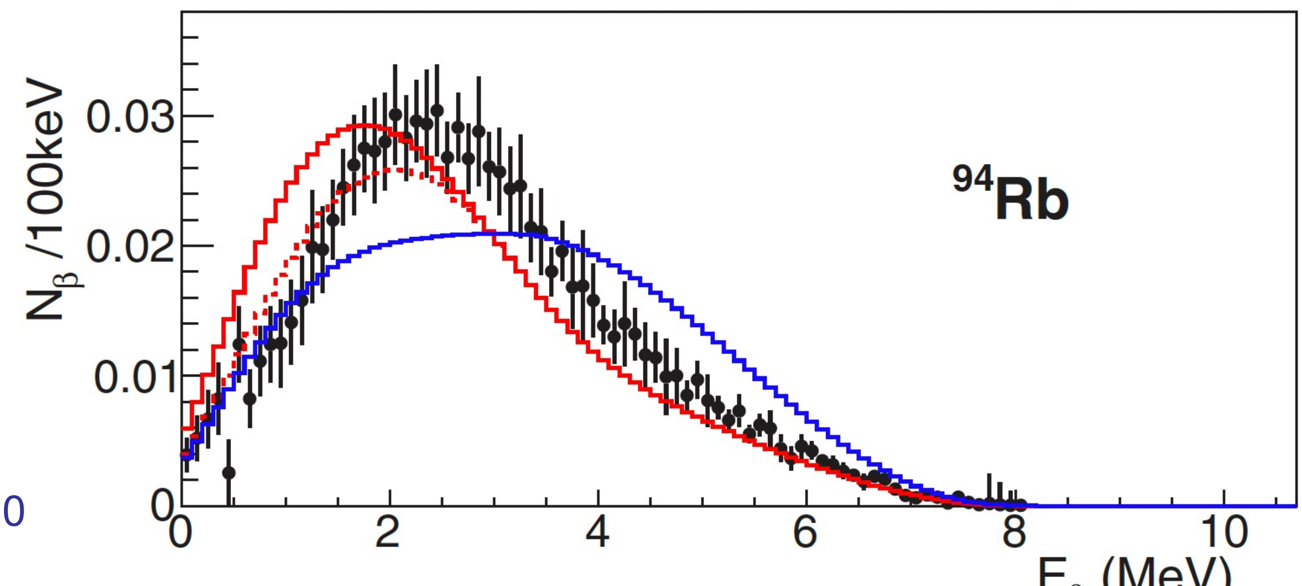
What can be improved:

- Better instrumentation ? Avoid the matching of two regions
- Isotopically pure beams
- Better Monte Carlo codes
- New deconvolution algorithms

Deduced β-shapes from TAGS data vs Tengblad data

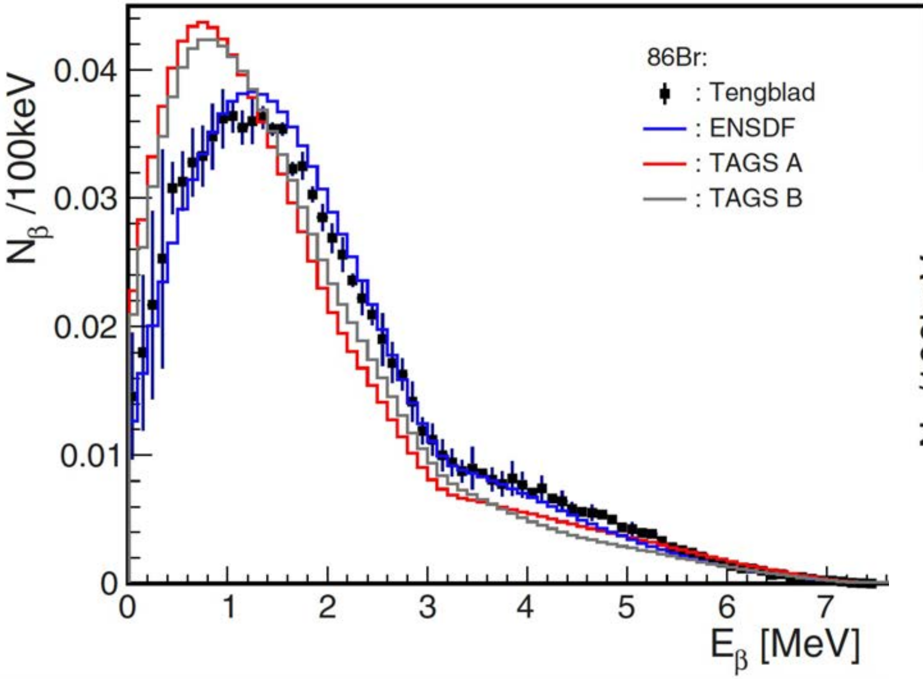


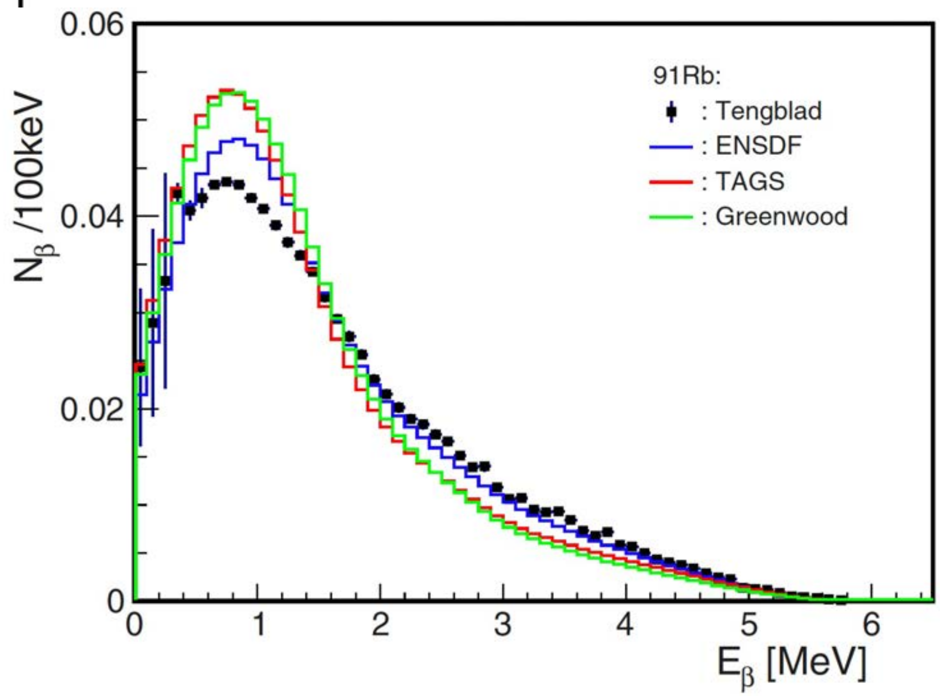
Black dots: Tengblad
Blue line: high-res
Red dotted: TAGS
Red cont. : TAGS +bdn



Valencia et al. PRC 95.024320

Deduced β-shapes from TAGS data vs Tengblad data





Black dots: Tengblad

Blue line: high-res

Red line: TAGS

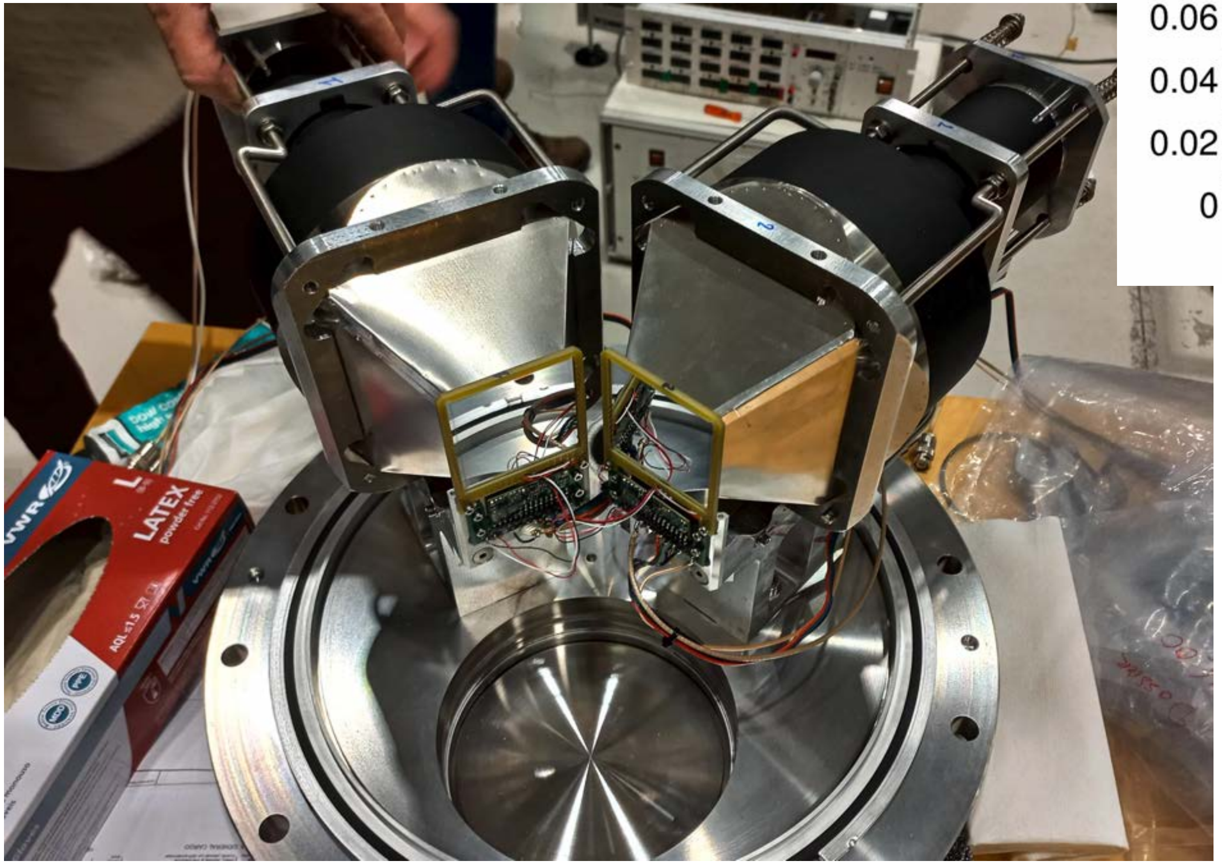
Rice et al. PRC 95.024320

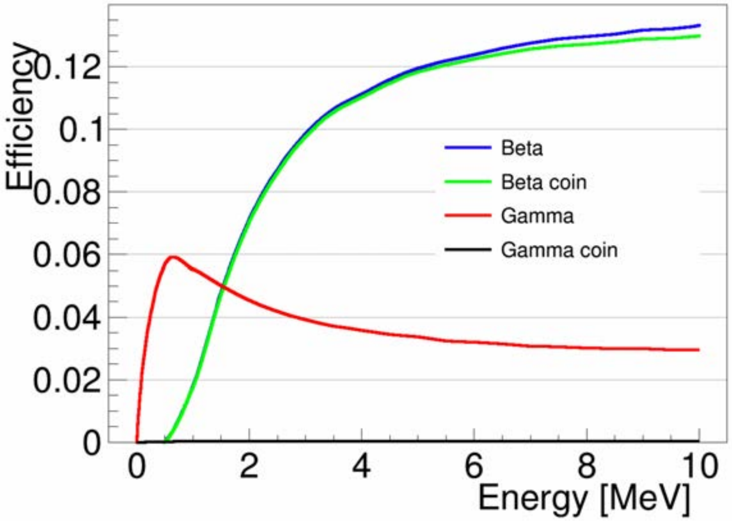
β-Shape project
Nantes-Surrey-Valencia Collaboration

- $\Delta E - E$ telescopes to measure the beta spectrum of selected decays using isotopically pure beams at Jyväskylä
- Si and plastic detectors

β-Shape project

Detail of the experimental chamber (designed by eng. of Nantes)



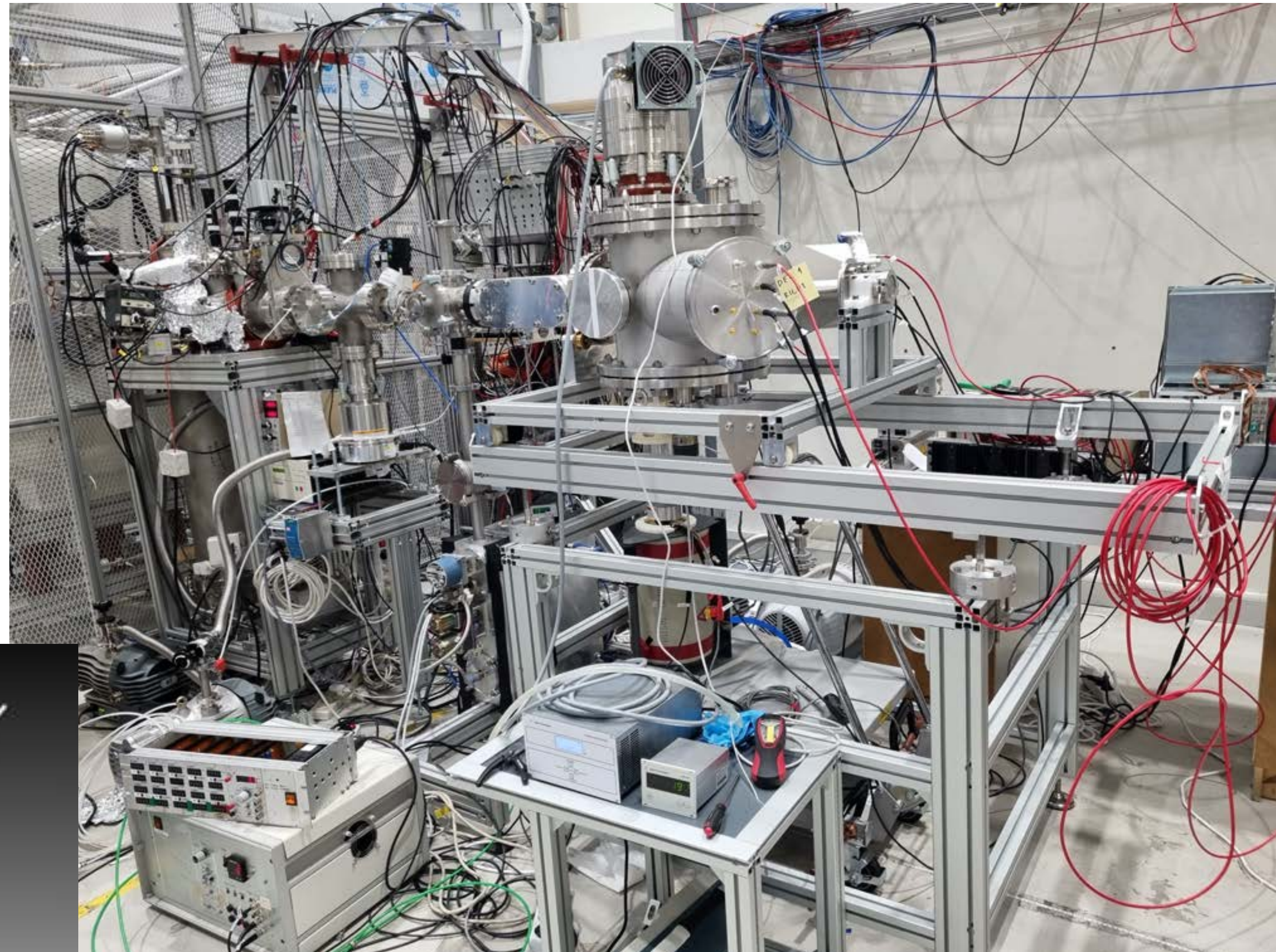


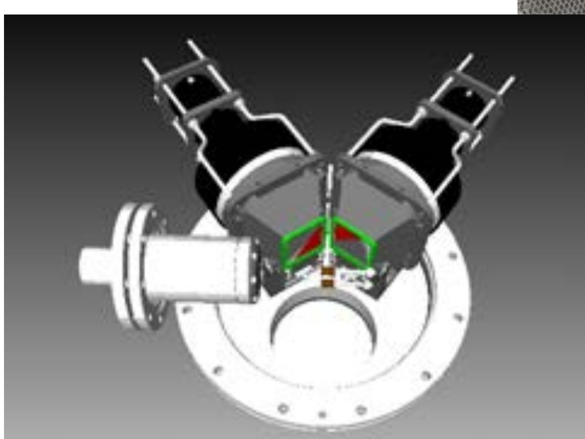
DE-E system provides very high gamma rejection efficiency

β-Shape project
Nantes-Surrey-Valencia Collaboration

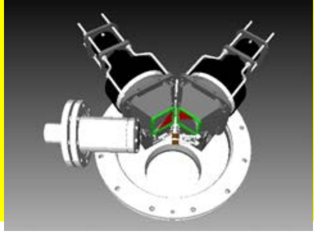
$\Delta E - E$ telescopes to measure the beta spectrum of selected decays using isotopically pure beams at Jyväskylä

Si and plastic detectors





β-Shape project



What we have done for the moment:

- Development of the setup. Comissioning and first experiment

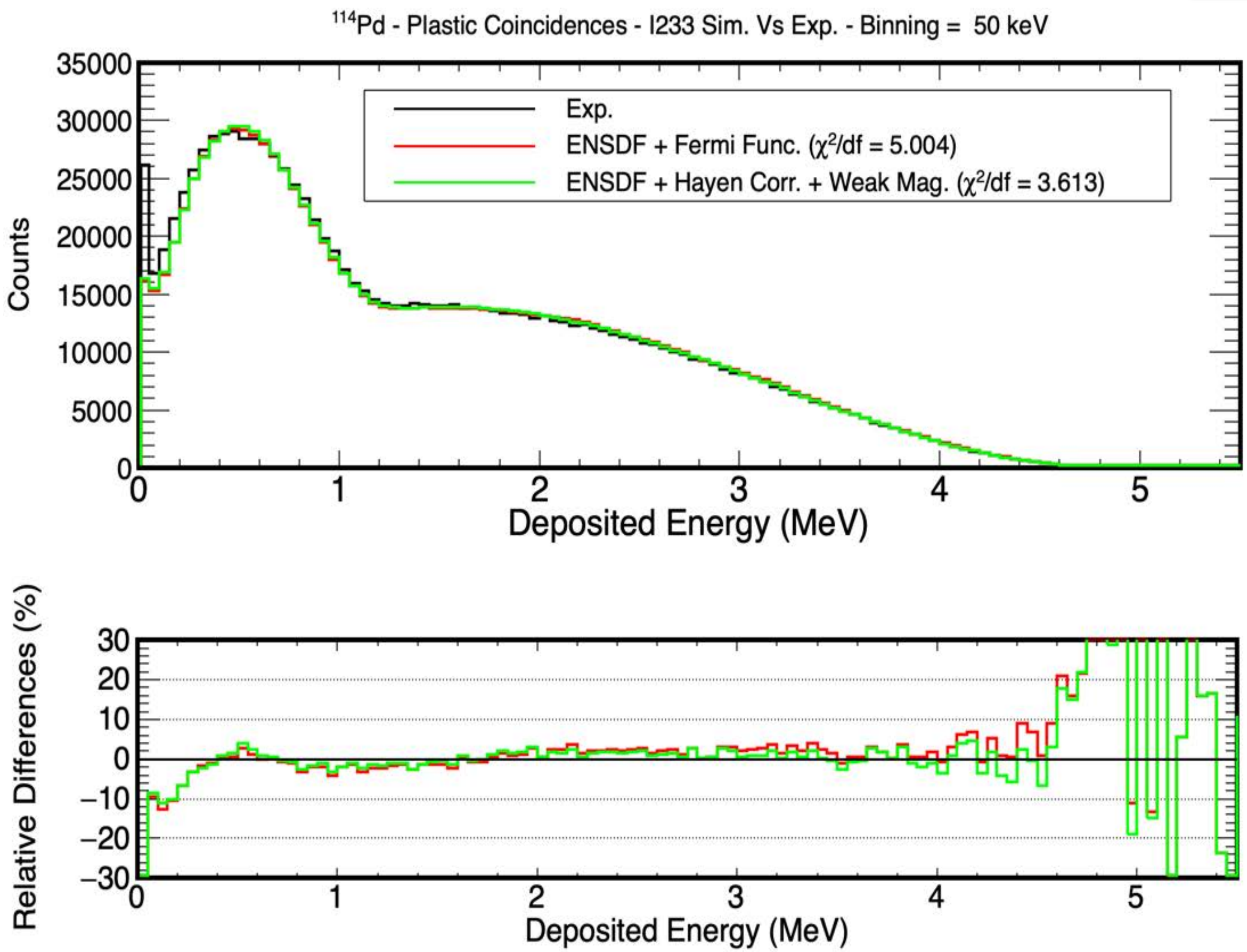
- Implementation of a complex event generator for the simulation of complex beta decays in GEANT4 (Hayen et al. corrections). Validation of the MC for the 114Ag case (see next slide)

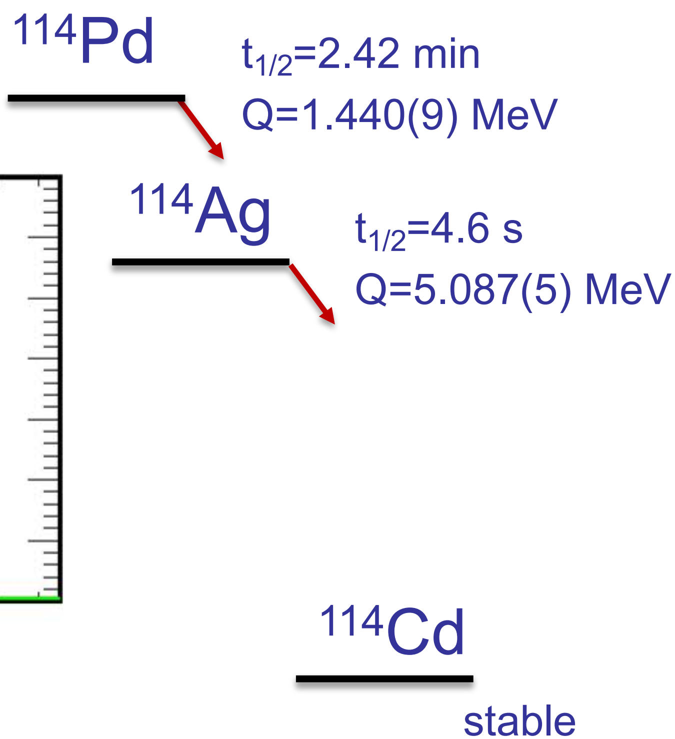
Next steps (d=Ro)

Measurement and analysis using deconvolution techniques of the most relevant contributors using our setup and deduce the spectrum shape for comparison with theoretical predictions. Meaurements using trap assisted spectroscopy

β-Shape project: commissioning

Validation of MC



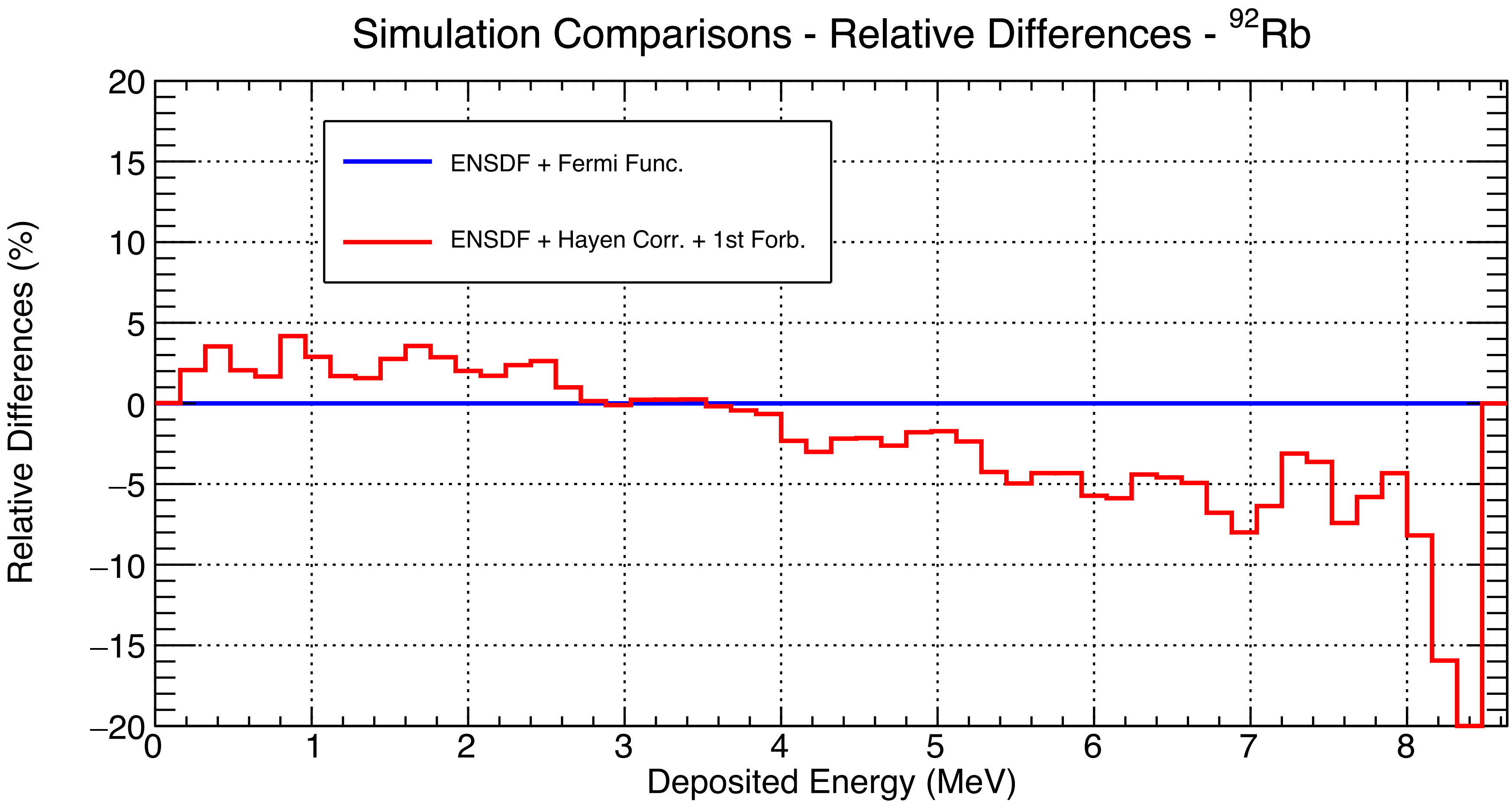


Experiment vs simulations, allowed decays

Courtesy of G. Alcala

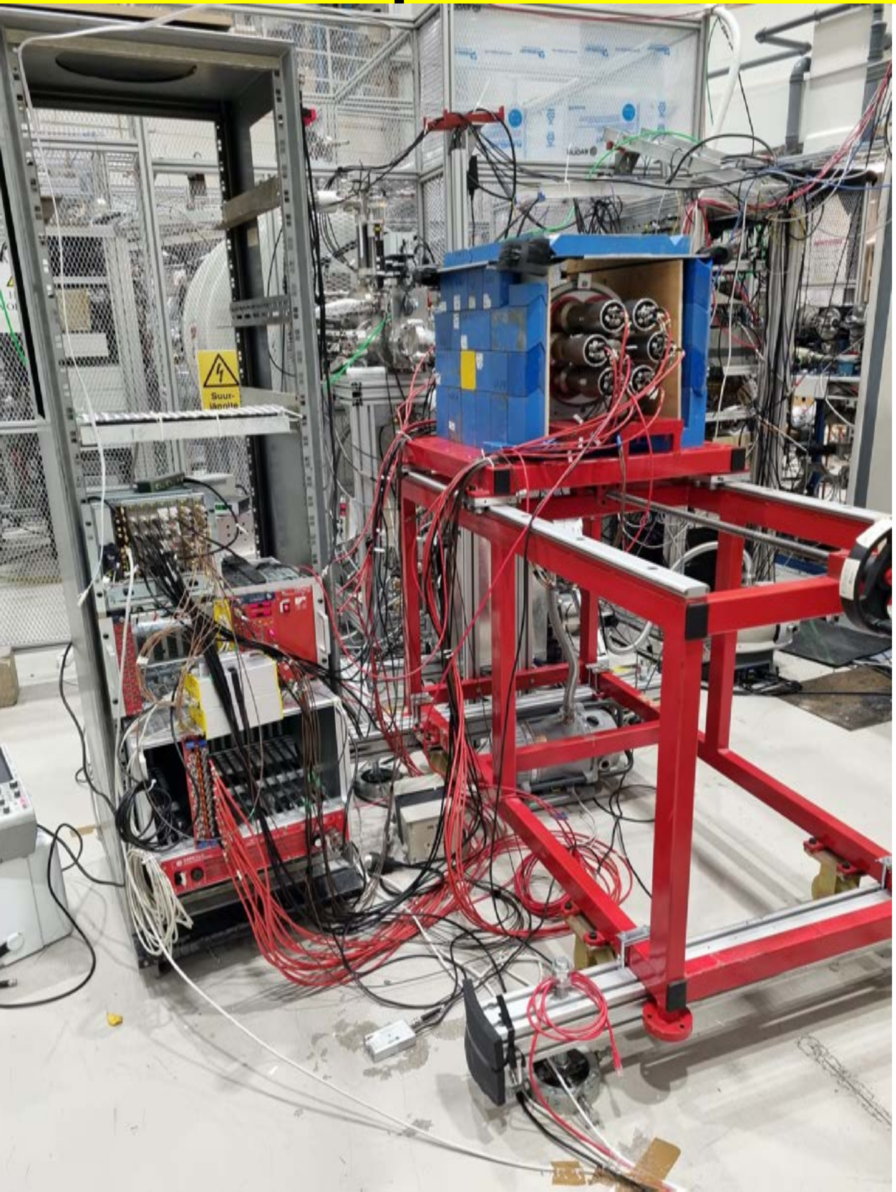
β-Shape project: challenge

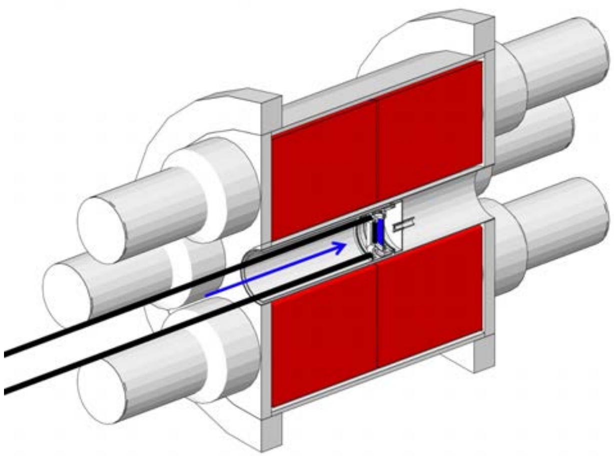
Hayen + 1st forbidden corrections effect on the shapes for 92Rb decay



Courtesy of G. Alcala

New experimental TAGS campaing performed in Sep. 2022





Segmented BaF2 detector

+

Plastic detector

Measured several cases of interest for antineutrino studies that include cases with isomers and beta delayed neutron emission. Approx. 16 cases.

Cases previosly identified as important contributors to the summation calculation.

Summary

- We hope that it was shown that total absorption measurements can provide useful data for applications related to nuclear reactors, in particular for decay heat calculations and for anti-neutrino physics applications
- We are running a research program related to this topic, that can also have an impact in nuclear structure and astrophysics (not discussed here)
- We also have started a new experimental program related to beta spectrum shape studies, that profits from our earlier experience at Jyväskylä and our experience in deconvoluting spectra.
- We thank the IAEA data section for the continous support of the related activities

Collaboration

Univ. of Jyvaskyla, Finland

CIEMAT, Spain

UPC, Spain

Subatech, France

Univ. of Surrey, UK

ATOMKI, Hungary

PNPI, Russia

LPC, France

IFIC, Spain

GSI, Germany

Special thanks to the students who have worked in the project:

D. Jordan, S. Rice, A. -A. Zakari-Issoufou,

E. Valencia, V. Guadilla, L. Le Meur, G. Alcala

Discussions with and slides from: L. Giot, M. Fallot, J. L. Tain, V. Guadilla, G. Alcala are acknowledged

Decay heat calculations: L. Giot, A. Sonzogni, Ch. Sublet, M. Fleming

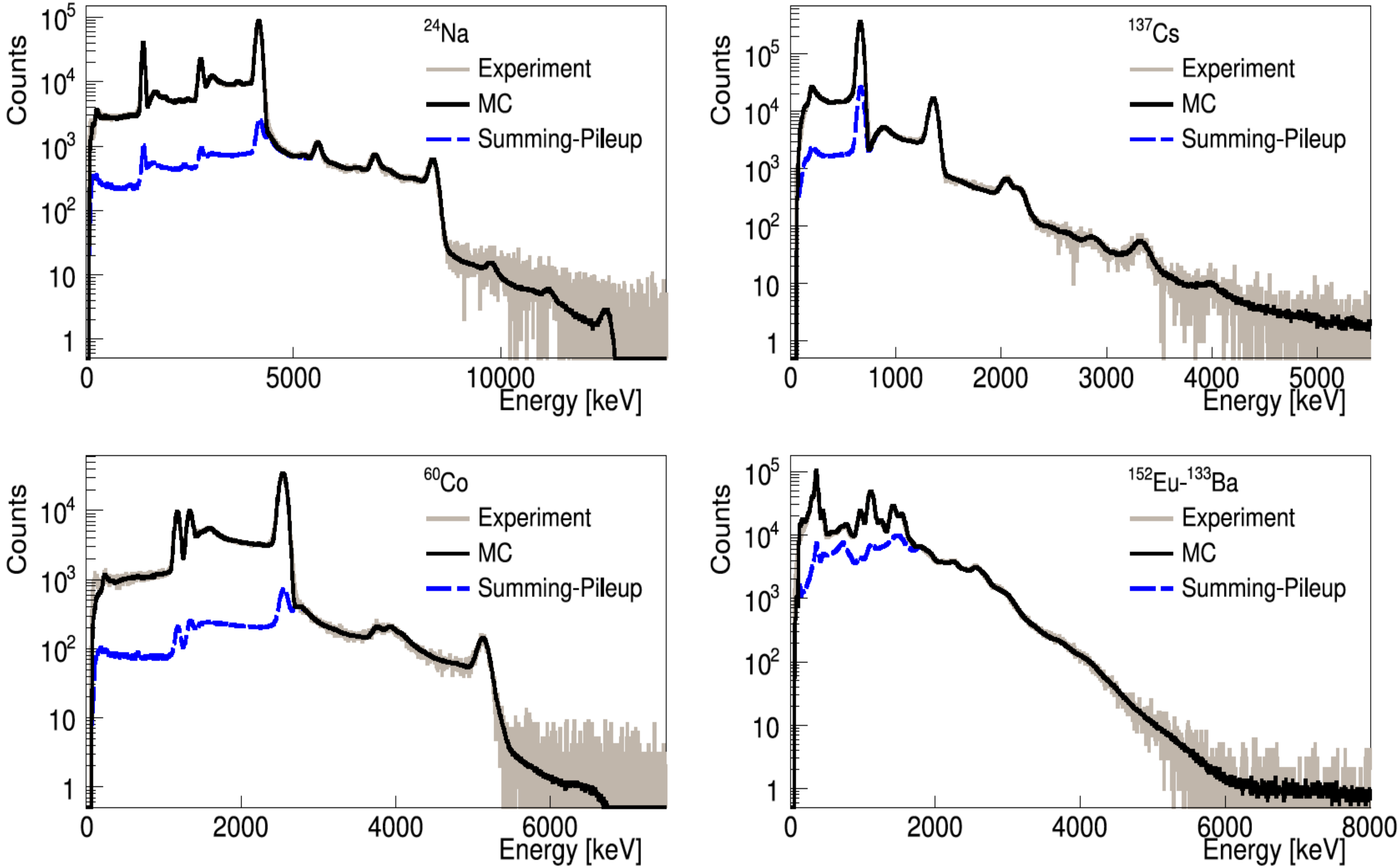
Antineutrino calculations: M. Estienne, M. Fallot, A. Sonzogni

THANK YOU

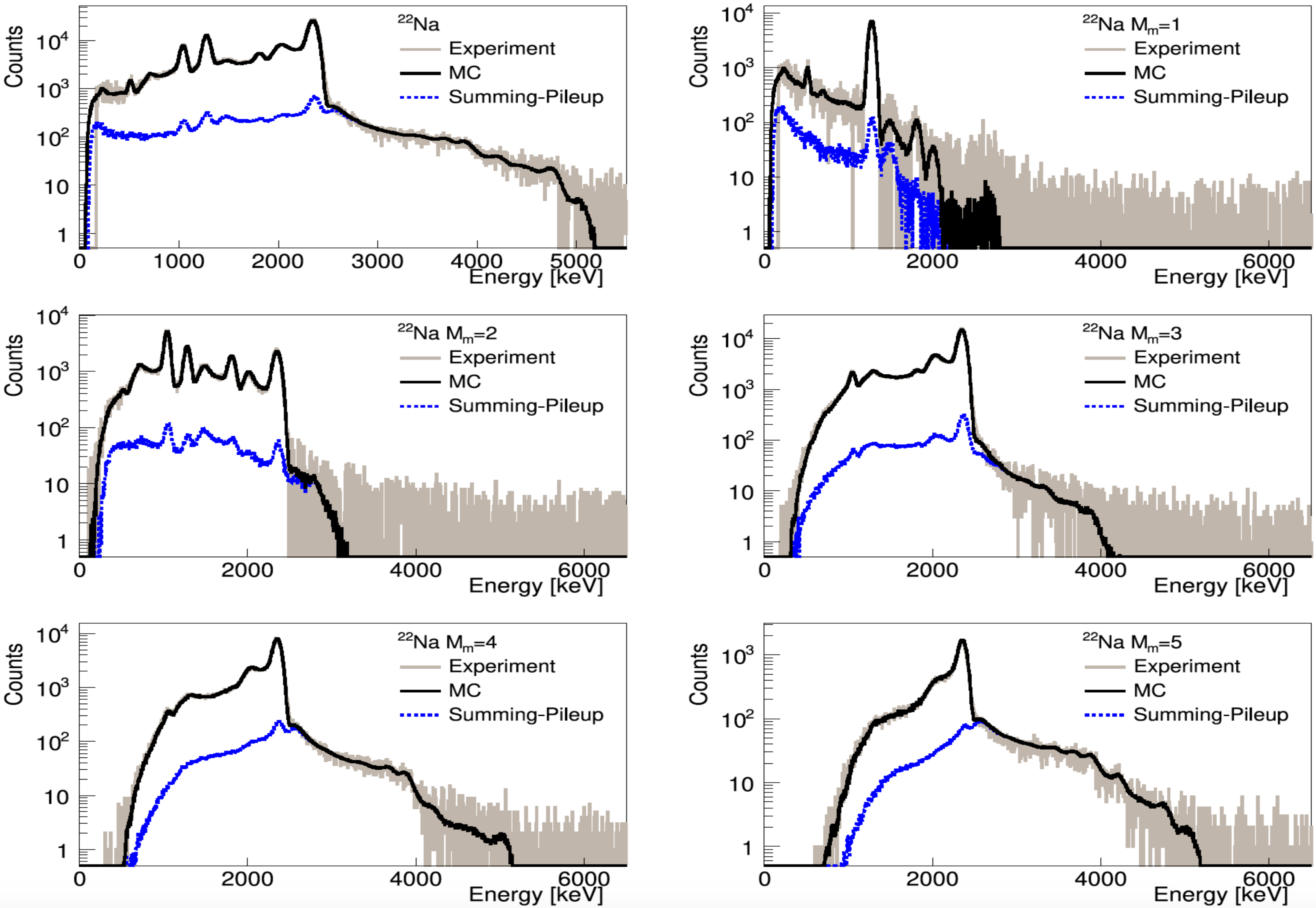
V. Guadilla, L. Le Meur, Z. Issoufou, S. Rice, E. Valencia, M. D. Jordan, A. Beloeuvre, J. L. Tain, M. Fallot, M. Estienne, J. Agramunt, A. Porta, J. A. Briz, A. A. Sonzogni, T. Eronen, G. Alcala, J. A. Victoria, L. M. Fraile, E. Ganioglu, W. Gelletly, D. Gorelov, J. Hakala, A. Jokinen,, V. Kolkinen, J. Koponen, L. Lebois, T. Martinez, A. Montaner, I. Moore, E. Nácher, S. Orrigo, H. Penttilä, I. Pohjalainen, J. Reinikainen, M. Reponen, S. Rinta-Antila, B. Rubio, T. Shiba,, V. Vedia, A. Voss, J. N. Wilson, etc.

A. Algora, …

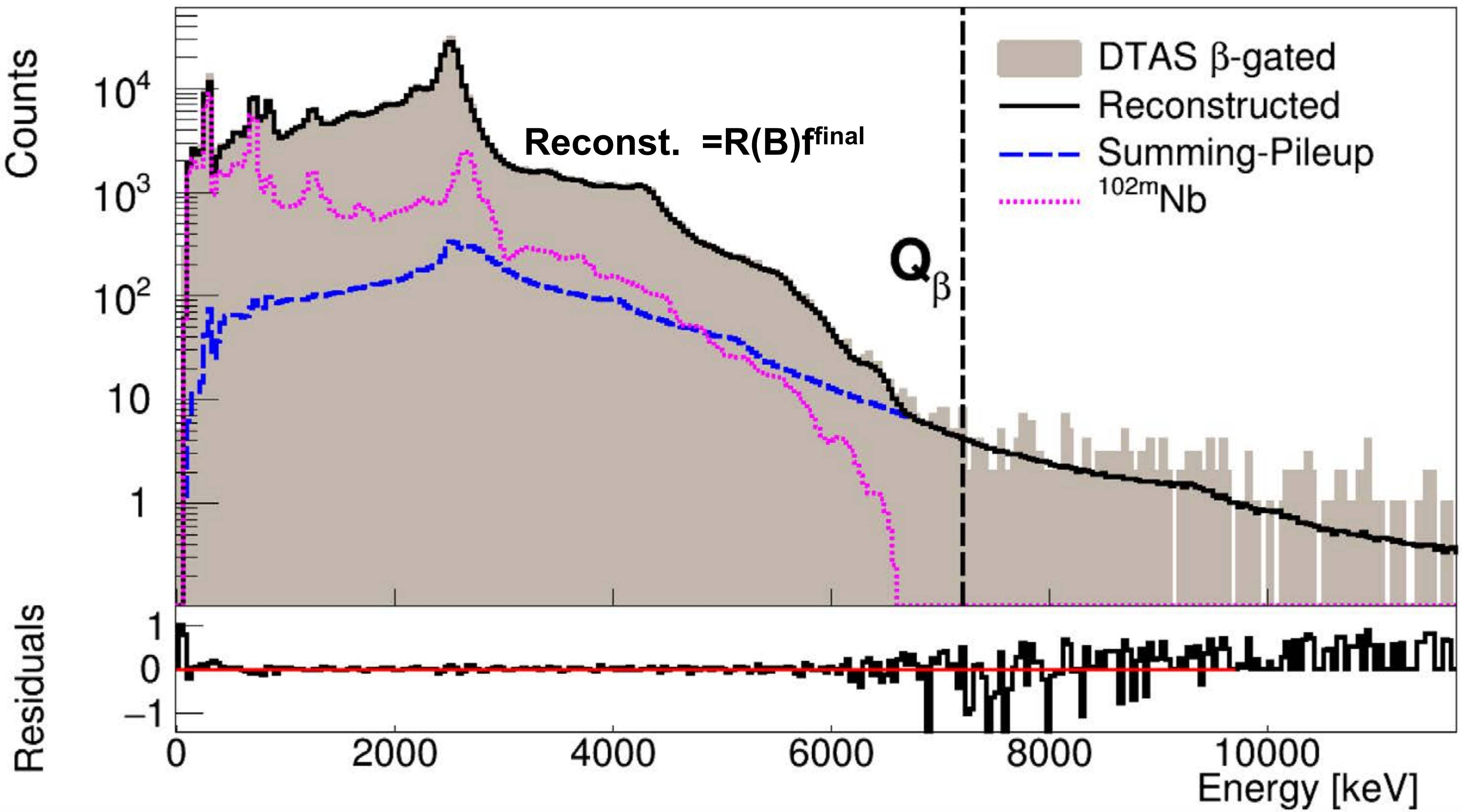
Characterization of the detector I: sources



Characterization of the detector II: reproduction of multiplicities ^{22}Na decay

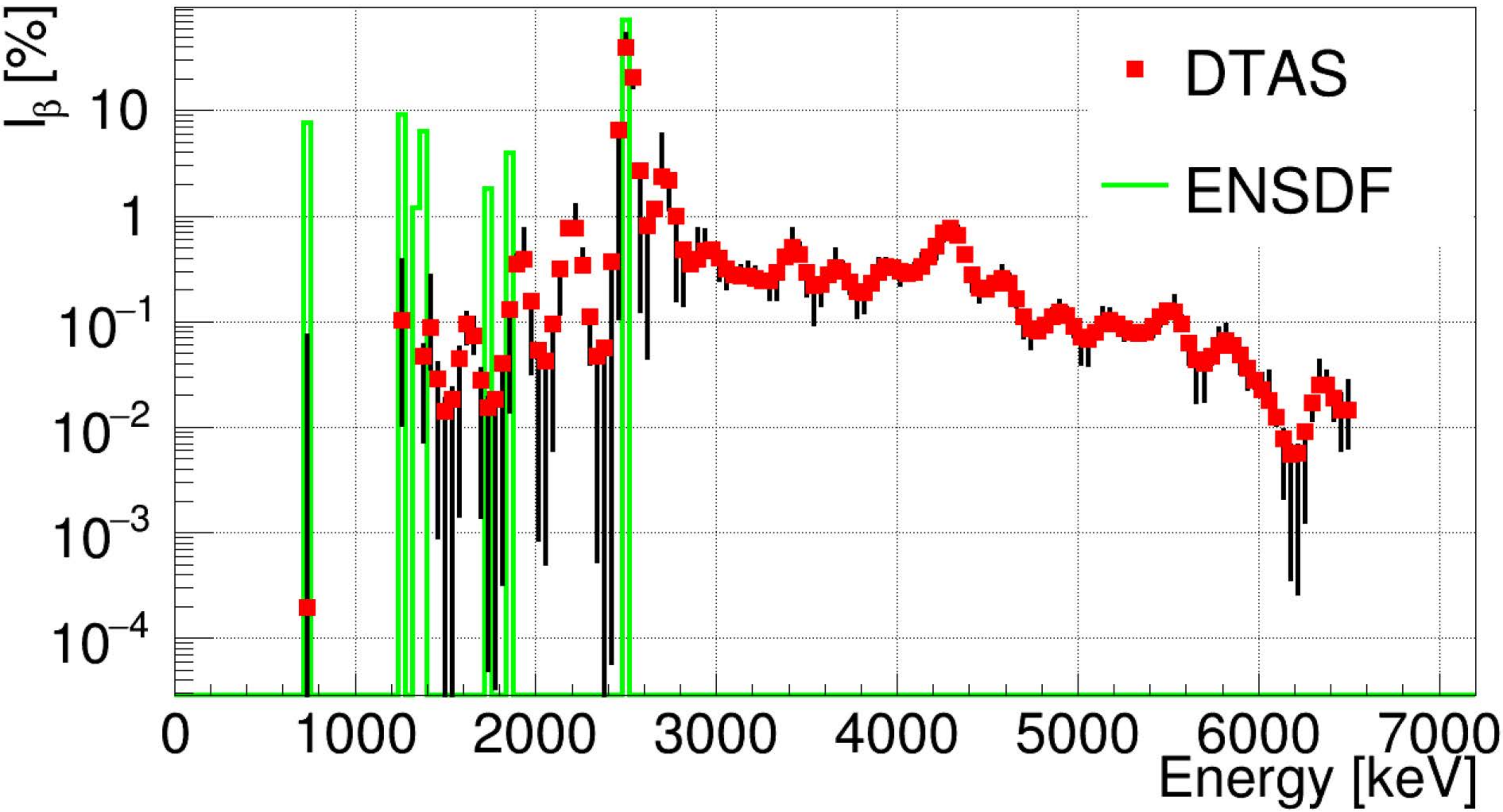


^{102gs}Nb decay (4+ state)



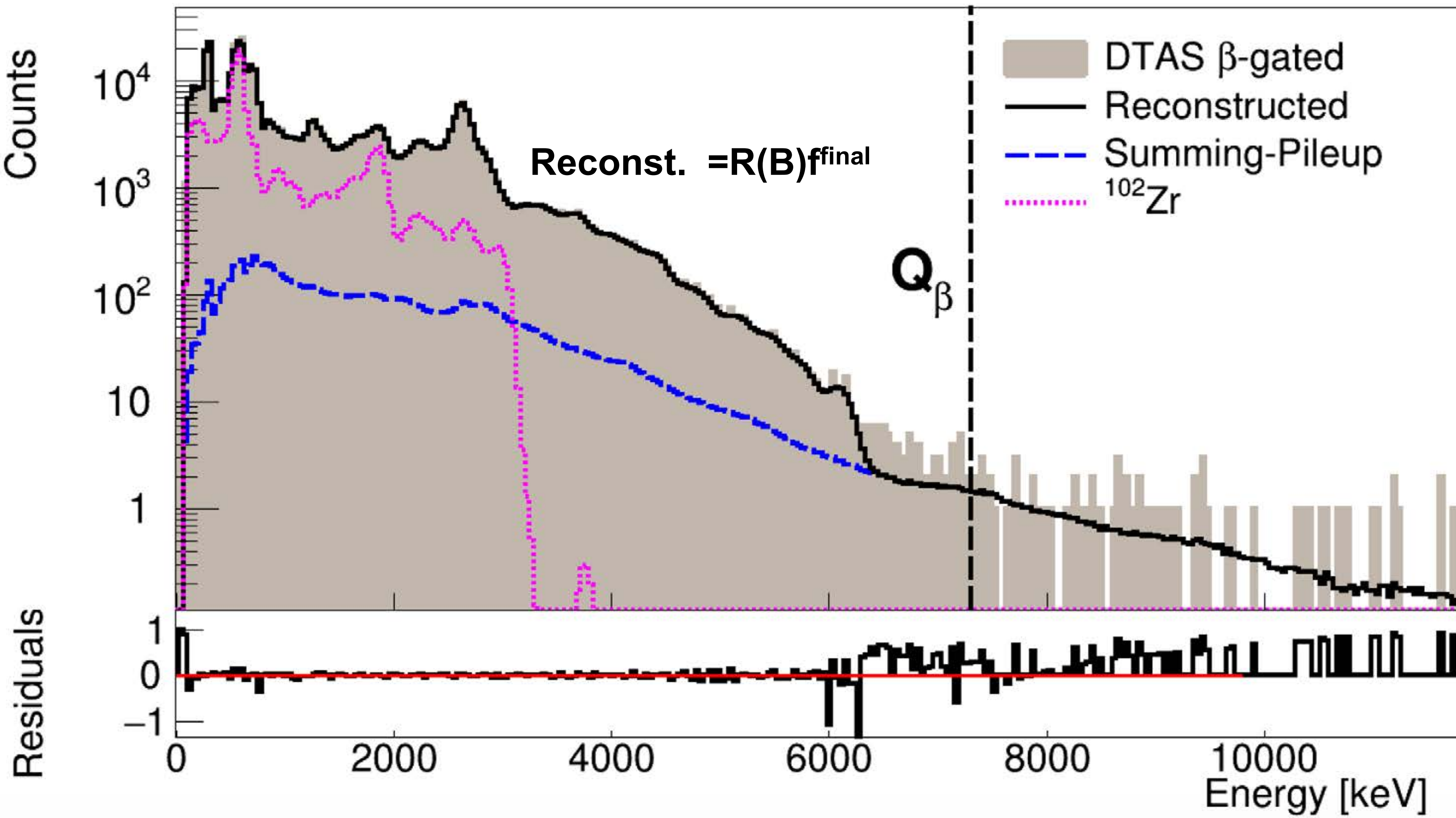
V. Guadilla et al., PhD thesis and V. Guadilla et al., PRL 112.042502

^{102gs}Nb decay (4+ state)



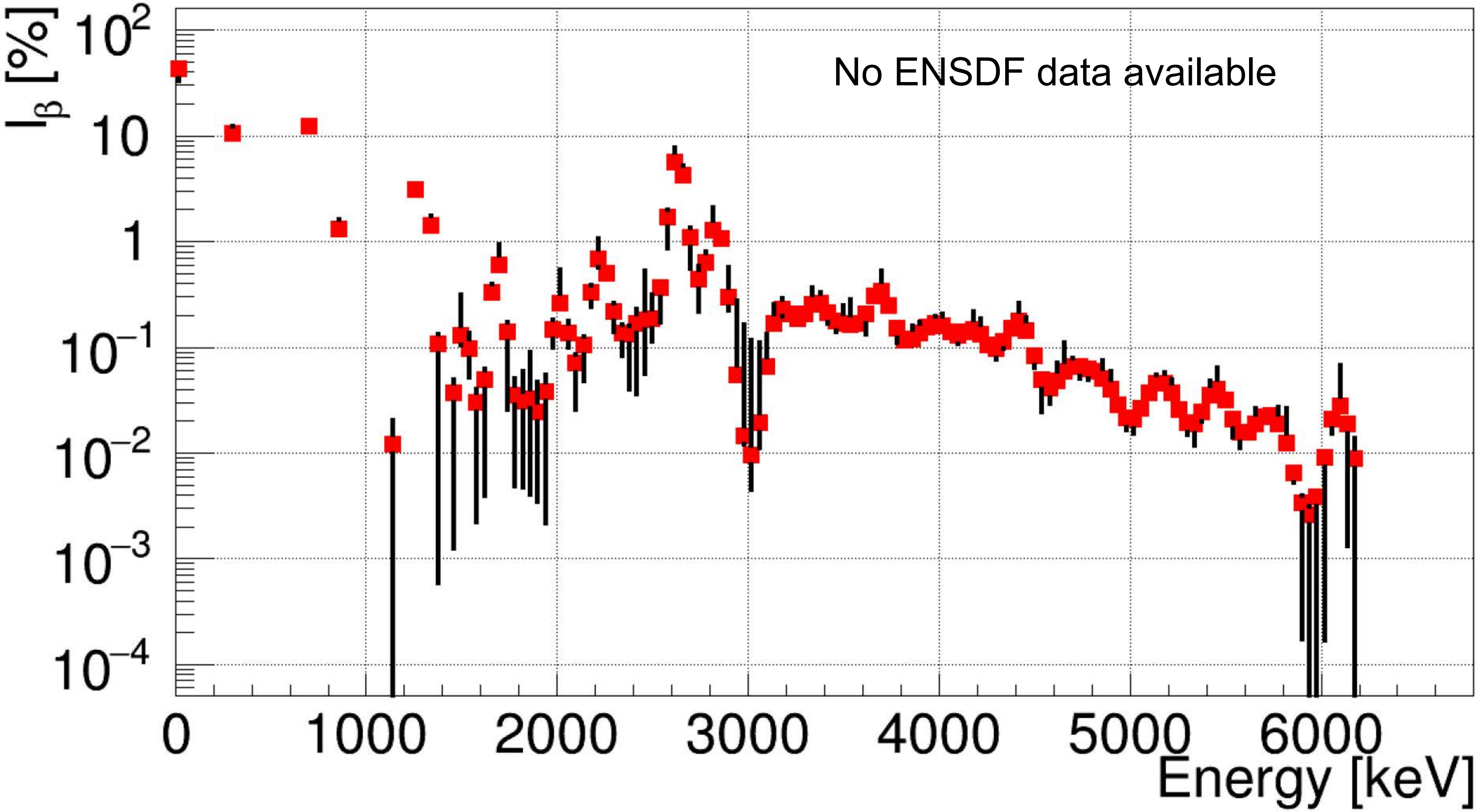
V. Guadilla et al., PRL 112.042502

^{102m}Nb decay (1+ state, 94 keV)



V. Guadilla et al., PhD thesis

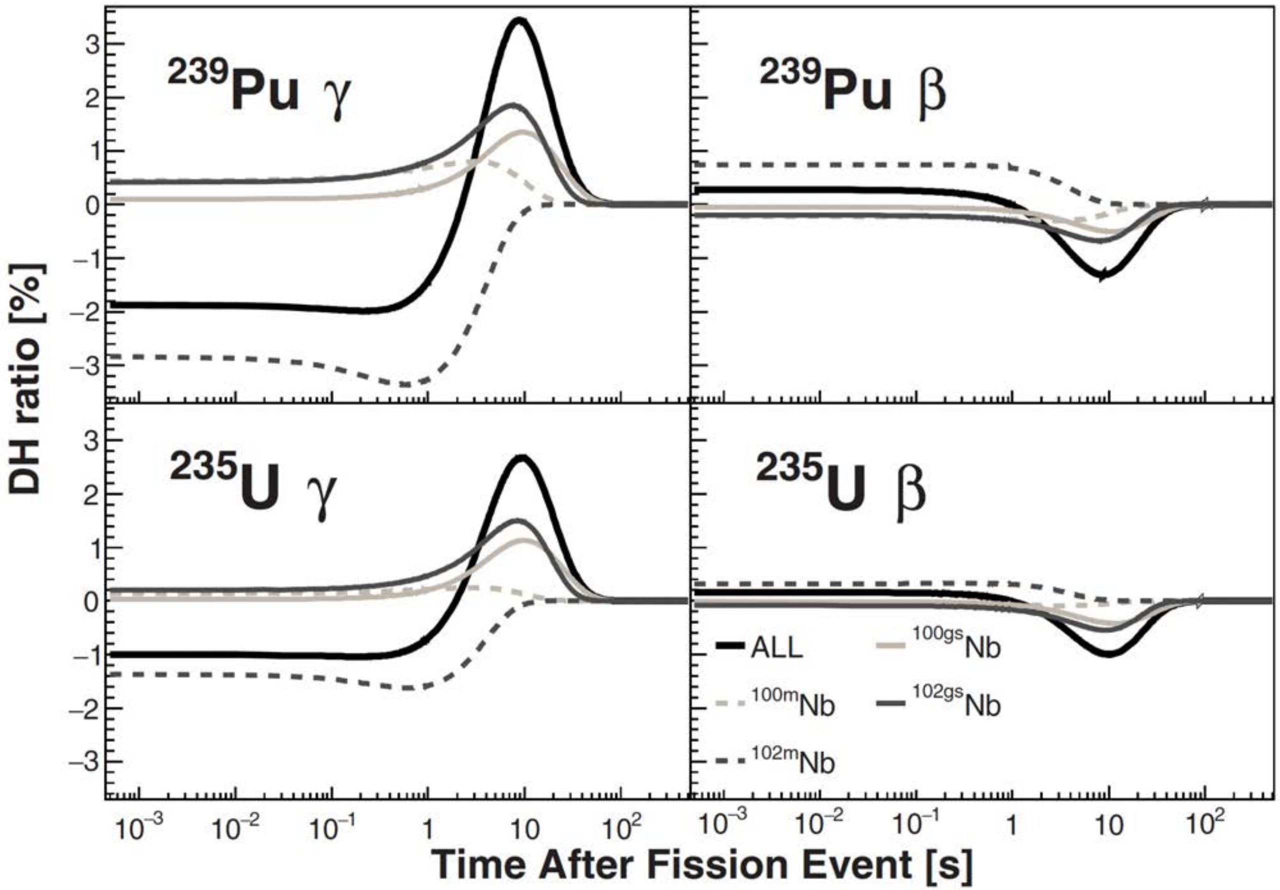
^{102m}Nb decay (1+ state, 94 keV)



V. Guadilla et al., PRL 112.042502

Impact on the decay heat summation calculations

DH summation calculation Courtesy of A. Sonzogni PhD thesis of V. Guadilla



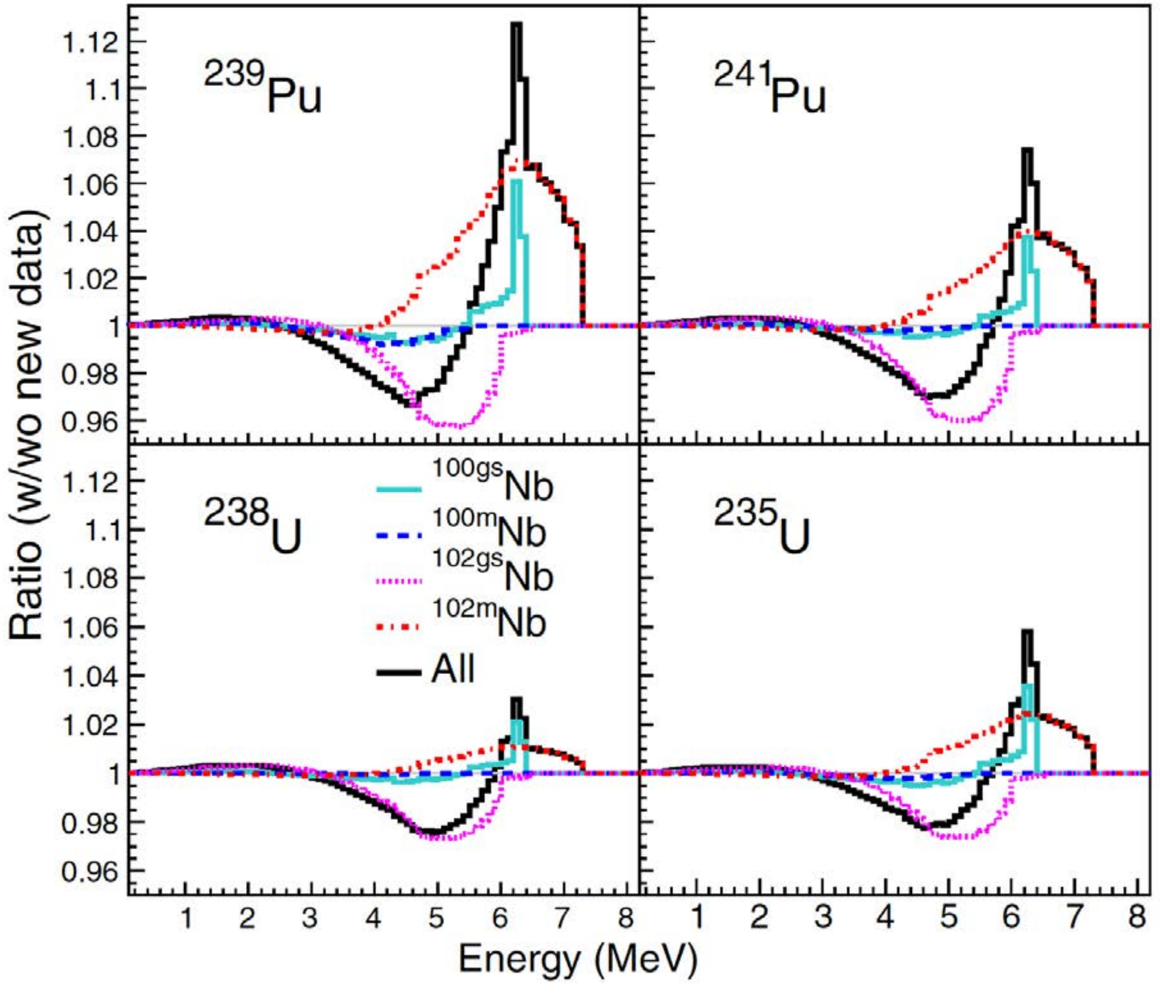
Impact of the 4 new Nb decay studies, with decaying isomers.

V. Guadilla et al., PRC 100, 024311 (2019)

Impact on the neutrino summation calculations

Neutrino summation calculation
Courtesy of M. Fallot, M. Estienne et al,
PhD thesis of V. Guadilla

Impact of the 4 new Nb decay studies, with decaying isomers. **Large impact in the region of the spectral distortion !!!**



V. Guadilla et al., PRL 112.042502