Total Absorption Spectroscopy Measurements for Applications and Nuclear Structure

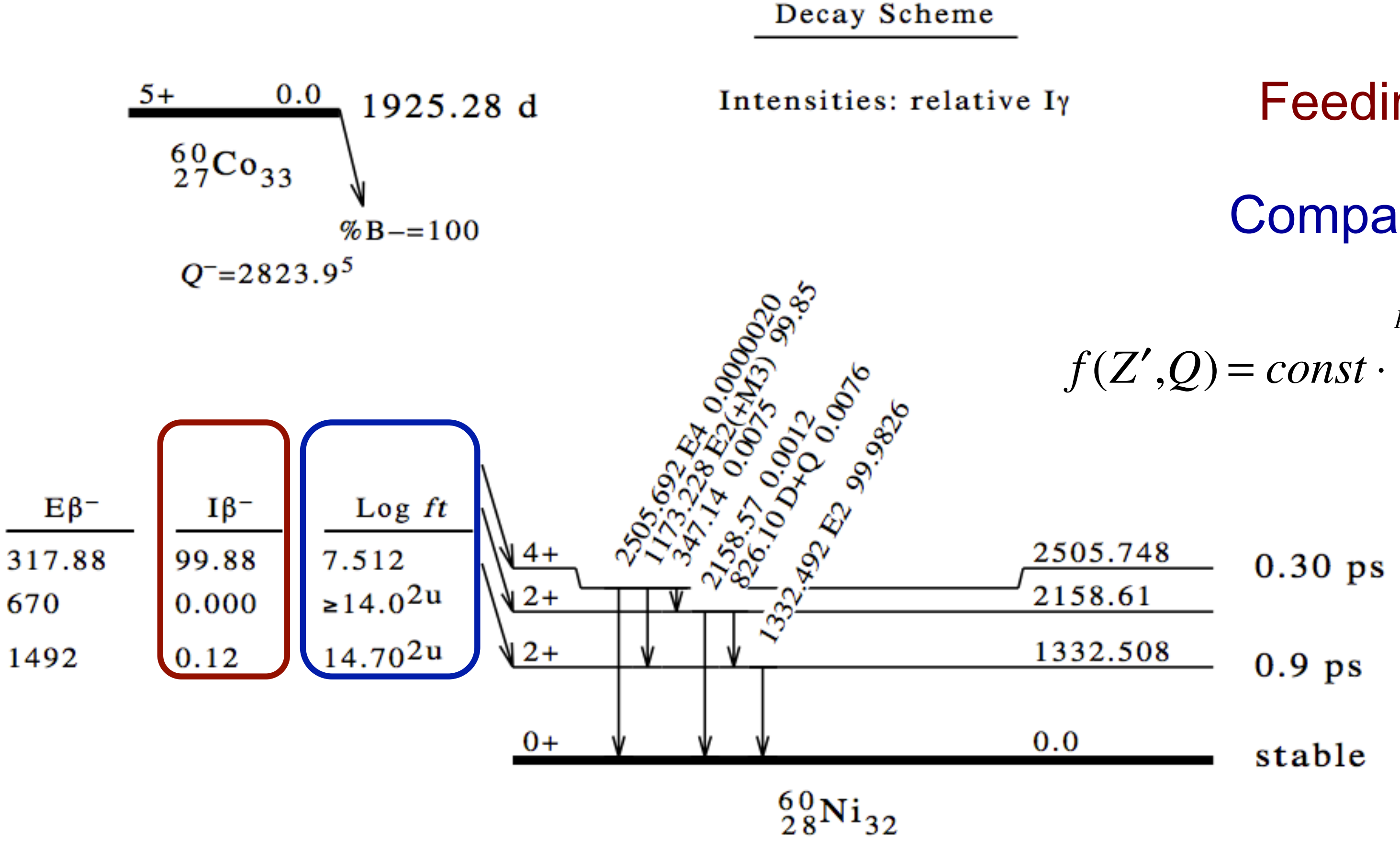
Alejandro Algora

IFIC, CSIC-University of Valencia, Spain

MTA ATOMKI, Debrecen, Hungary

INT Neutrino Workshop, Seattle, November 2013

Example: ^{60}Co decay from http://www.nndc.bnl.gov/



Feeding:= $I_\beta = P_f$*100

Comparative half-life: ft

$$f(Z',Q) = const \cdot \int_0^{p_{\max}} F(Z',p)\, p^2 (Q - E_e)^2\, dp$$

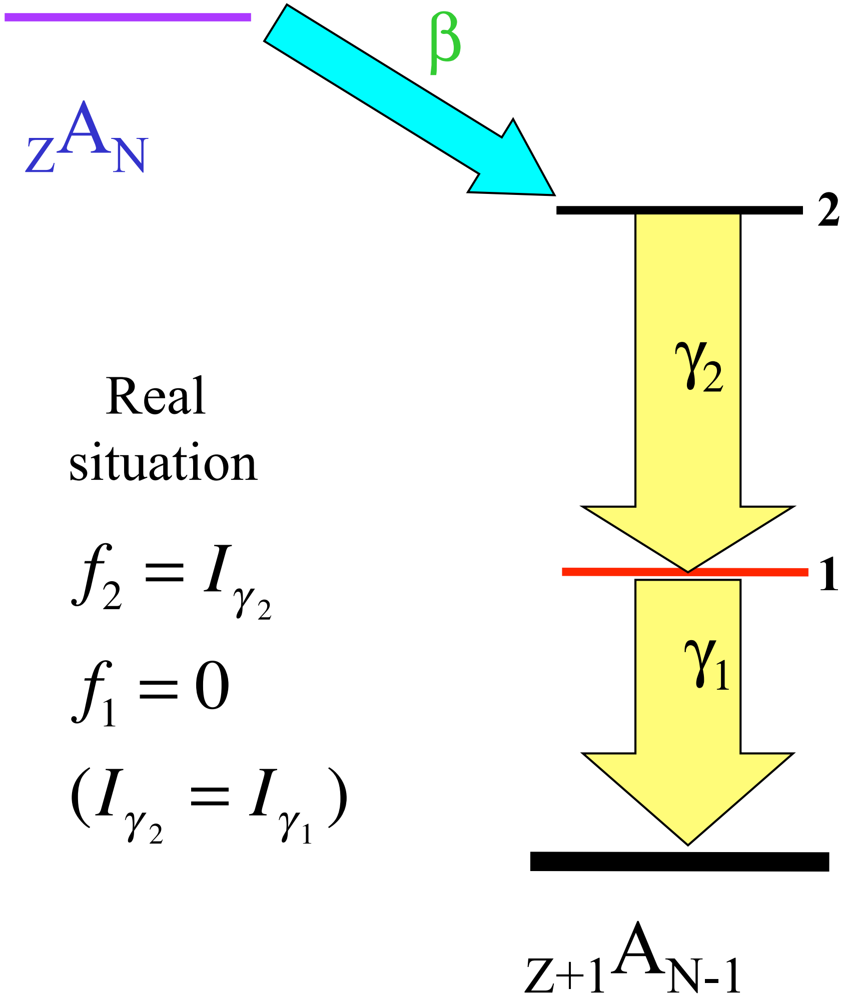
$$ft_f = const' \frac{1}{|M_{if}|^2} = const' \frac{1}{B_{i\to f}}$$

$$S_\beta(E) = \frac{P_\beta(E)}{f(Z',Q_\beta - E)T_{1/2}} = \frac{1}{ft(E)}$$

$$B_{i\to f} = \frac{1}{2J_i + 1} \left| \langle \Psi_f | \tau^{\pm} \; or \; \sigma\tau^{\pm} | \Psi_i \rangle \right|^2$$

$$t_f = \frac{T_{1/2}}{P_f} \qquad T_{1/2} = \frac{\ln(2)}{\lambda} = \tau \ln(2)$$

The problem of measuring the β-feeding

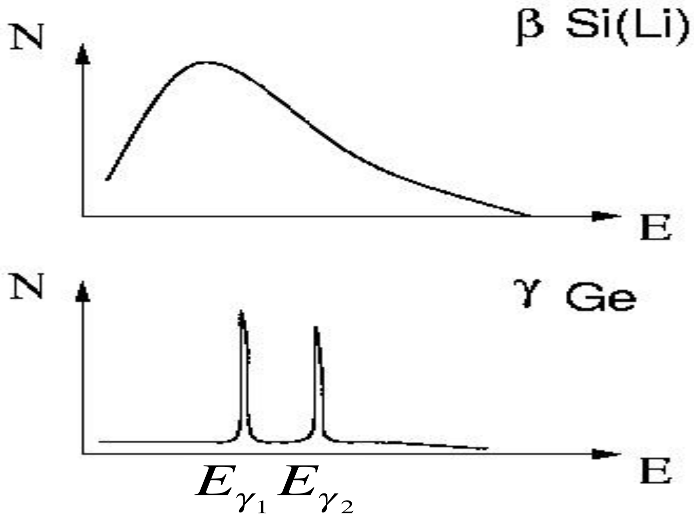


Real situation

$$f_2 = I_{\gamma_2}$$

$$f_1 = 0$$

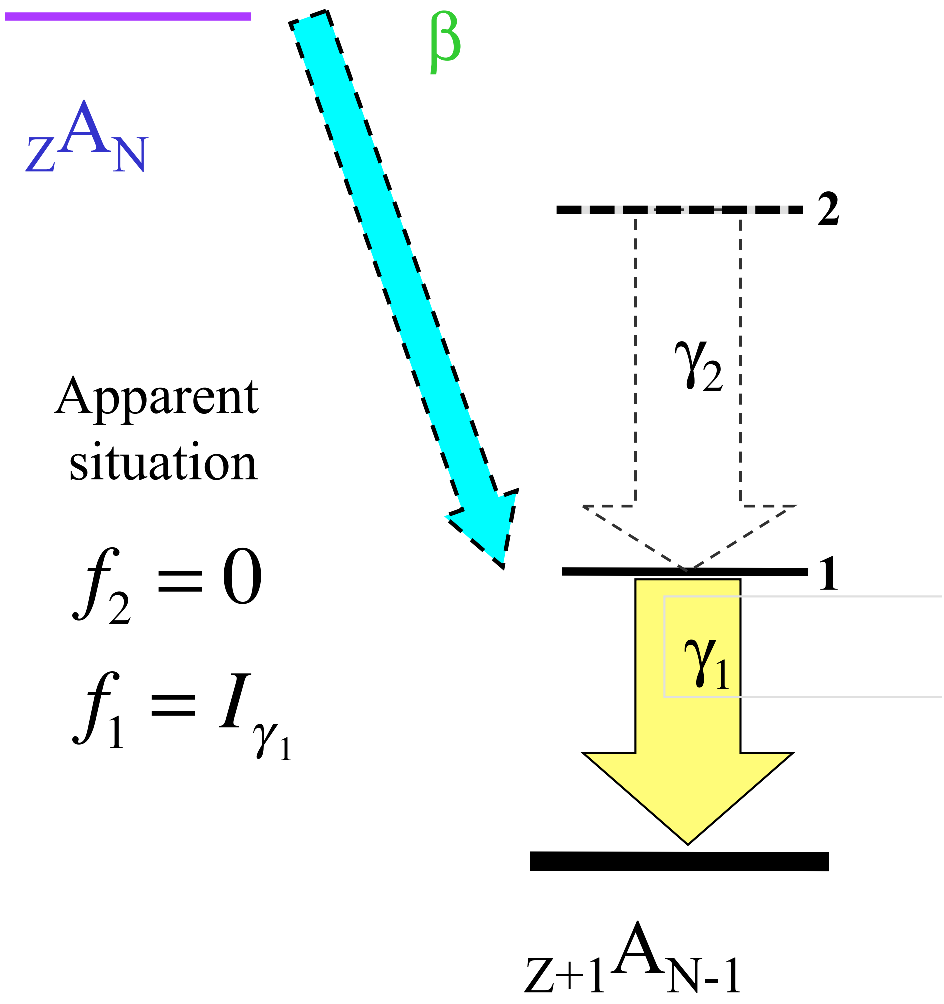
$$(I_{\gamma_2} = I_{\gamma_1})$$



- Ge detectors are conventionally used to construct the level scheme populated in the decay

- From the γ intensity balance we deduce the β-feeding

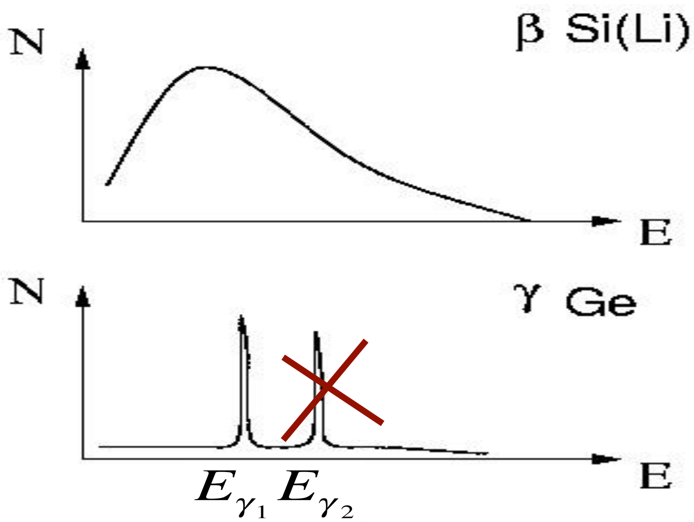
Experimental perspective: the problem of measuring the β- feeding



Apparent situation

$$f_2 = 0$$

$$f_1 = I_{\gamma_1}$$



- What happens if we miss some intensity

$$Single\ \gamma \sim \varepsilon$$

$$Coinc\ \gamma_1\gamma_2 \sim \varepsilon_1\varepsilon_2$$

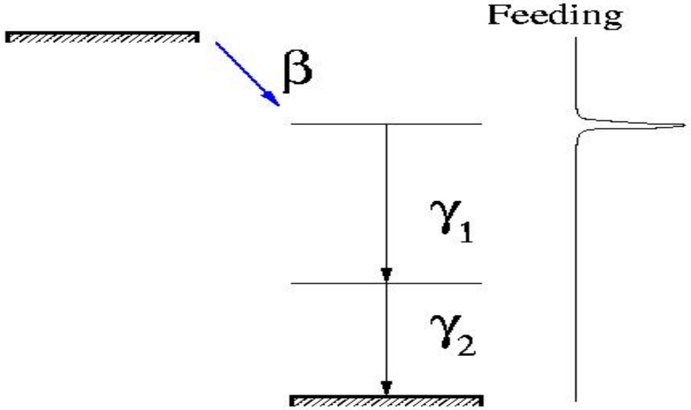
Pandemonium (The Capital of Hell)

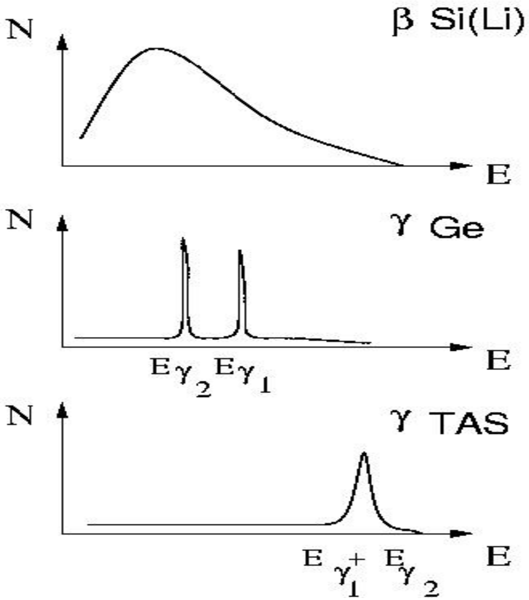
introduced by John Milton (XVII) in his epic poem Paradise Lost

John Martin (~ 1825), presently at Louvre

Hardy et al., Phys. Lett. 71B (1977) 307

TAGS measurements





Since the gamma detection is the only reasonable way to solve the problem, we need a highly efficient device:

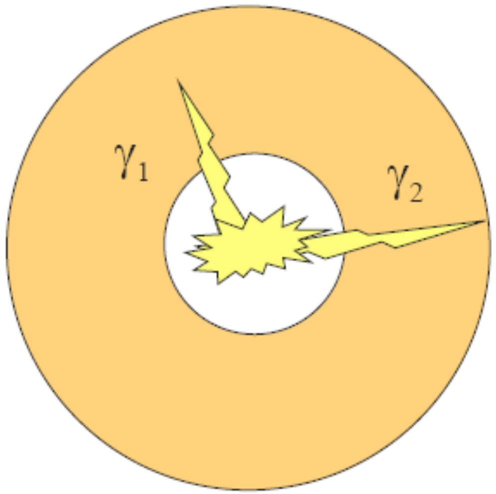
A TOTAL ABSORTION SPECTROMETER

But there is a change in philosophy. Instead of detecting the individual gamma rays we sum the energy deposited by the gamma cascades in the detector.

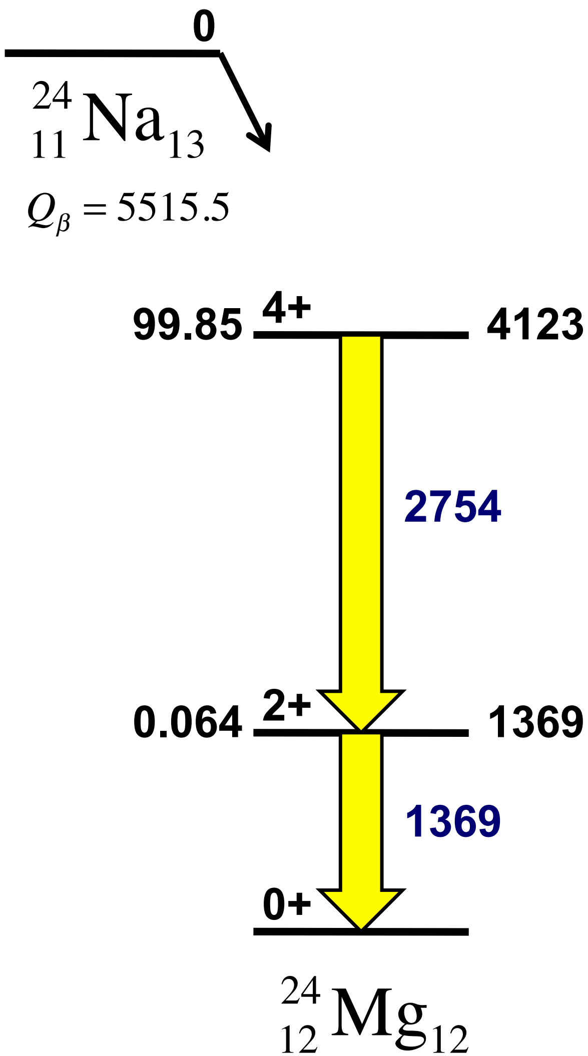
A TAS is like a calorimeter!

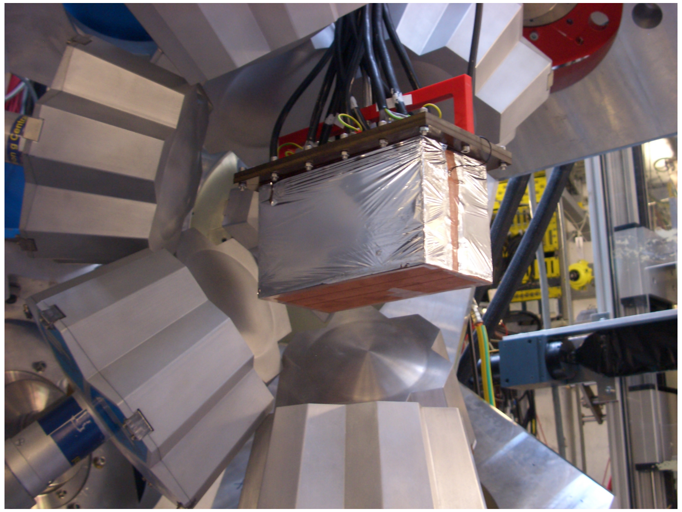
Big crystal, 4π

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



Ge detector case: ^{24}Na decay





RISING

Stopped Beam Configuration:

15 clusters, 105 Ge capsules

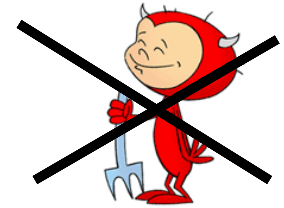
$$\varepsilon_{p1} = 0.10 \qquad \gamma_1 = 1369 \text{ keV}$$

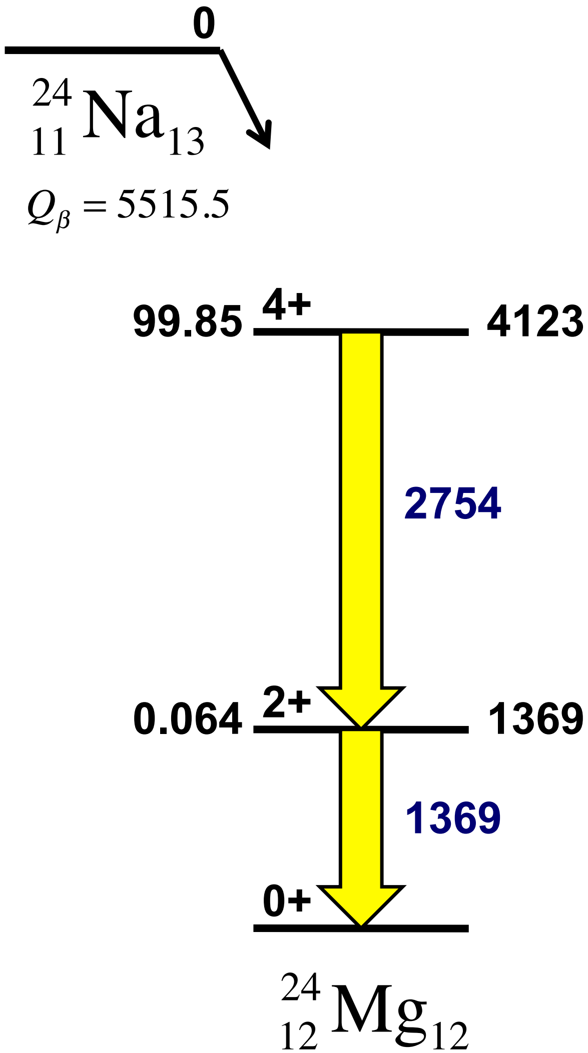
$$\varepsilon_{p2} = 0.06 \qquad \gamma_2 = 2754 \text{ keV}$$

$$\varepsilon_{coinc} = \varepsilon_{p1} \cdot \varepsilon_{p2}$$

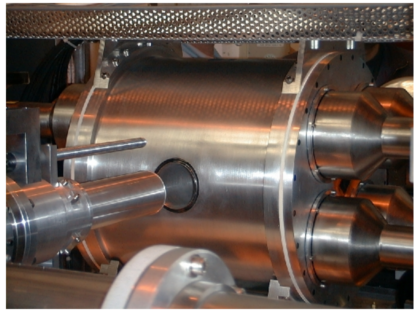
$$\varepsilon_{coinc} = 0.006$$

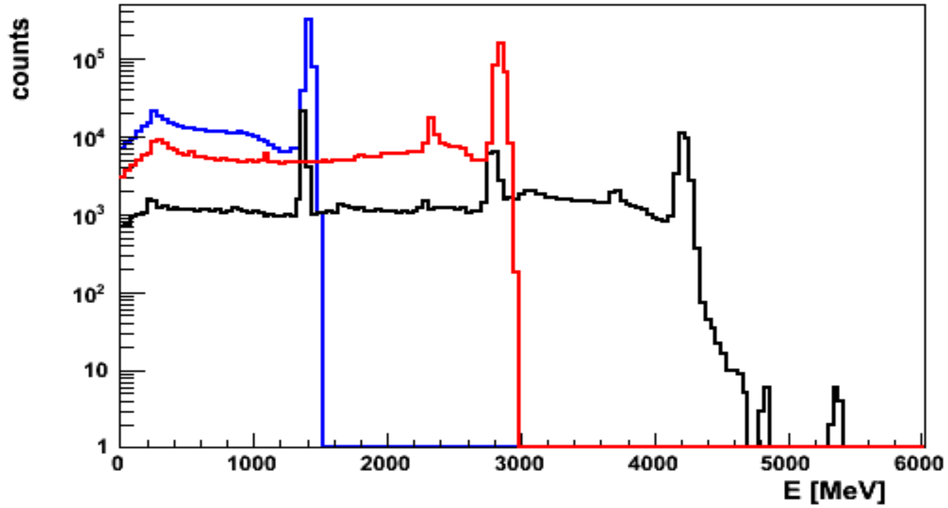
TAS case: ^{24}Na decay





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





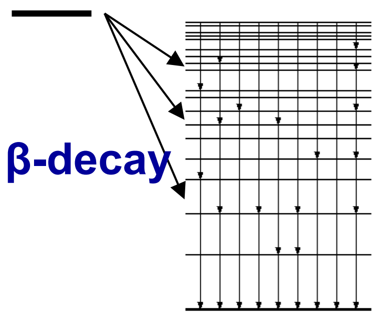
$$\varepsilon_{Total}(1369\ \mathrm{keV}) = 0.81$$

$$\varepsilon_{Total}(2754\ \mathrm{keV}) = 0.72$$

$$\varepsilon_{Total}(cascade) = \varepsilon_{Total}^{\gamma_1}(1-\varepsilon_{Total}^{\gamma_2}) + \varepsilon_{Total}^{\gamma_2}(1-\varepsilon_{Total}^{\gamma_1}) + \varepsilon_{Total}^{\gamma_1}\varepsilon_{Total}^{\gamma_2} = 0.95$$

Analysis

$$d_i = \sum_j R_{ij} f_j \quad or \quad \mathbf{d} = \mathbf{R} \cdot \mathbf{f}$$

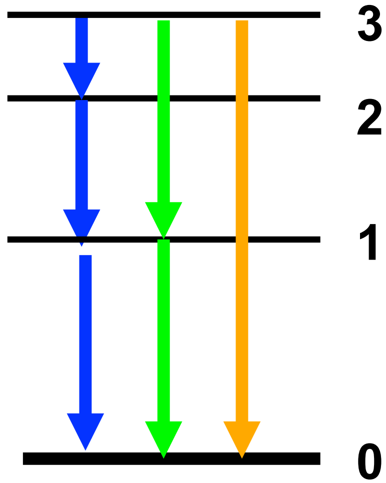


R is the response function of the spectrometer, ***R**_ij_* means the probability that feeding at a level ***j*** gives counts in data channel ***i of the spectrum***

The response matrix $\mathbf{R}$ can be constructed by recursive convolution:

$$\mathbf{R_j} = \sum_{k=0}^{j-1} b_{jk} \mathbf{g_{jk}} \otimes \mathbf{R_k}$$

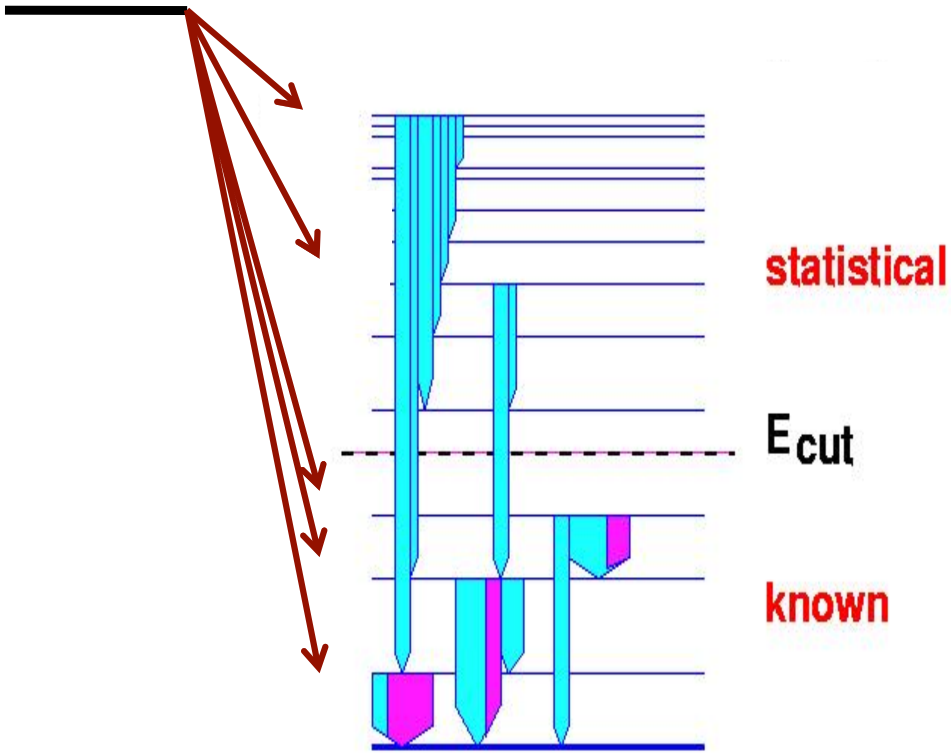
$\mathbf{g_{jk}}$: γ-response for *j* ➔ *k* transition
$\mathbf{R_k}$: response for level *k*
b_{jk}: branching ratio for *j* ➔ *k* transition



Mathematical formalization by Tain, Cano, et al.

The complexity of the TAGS analysis

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



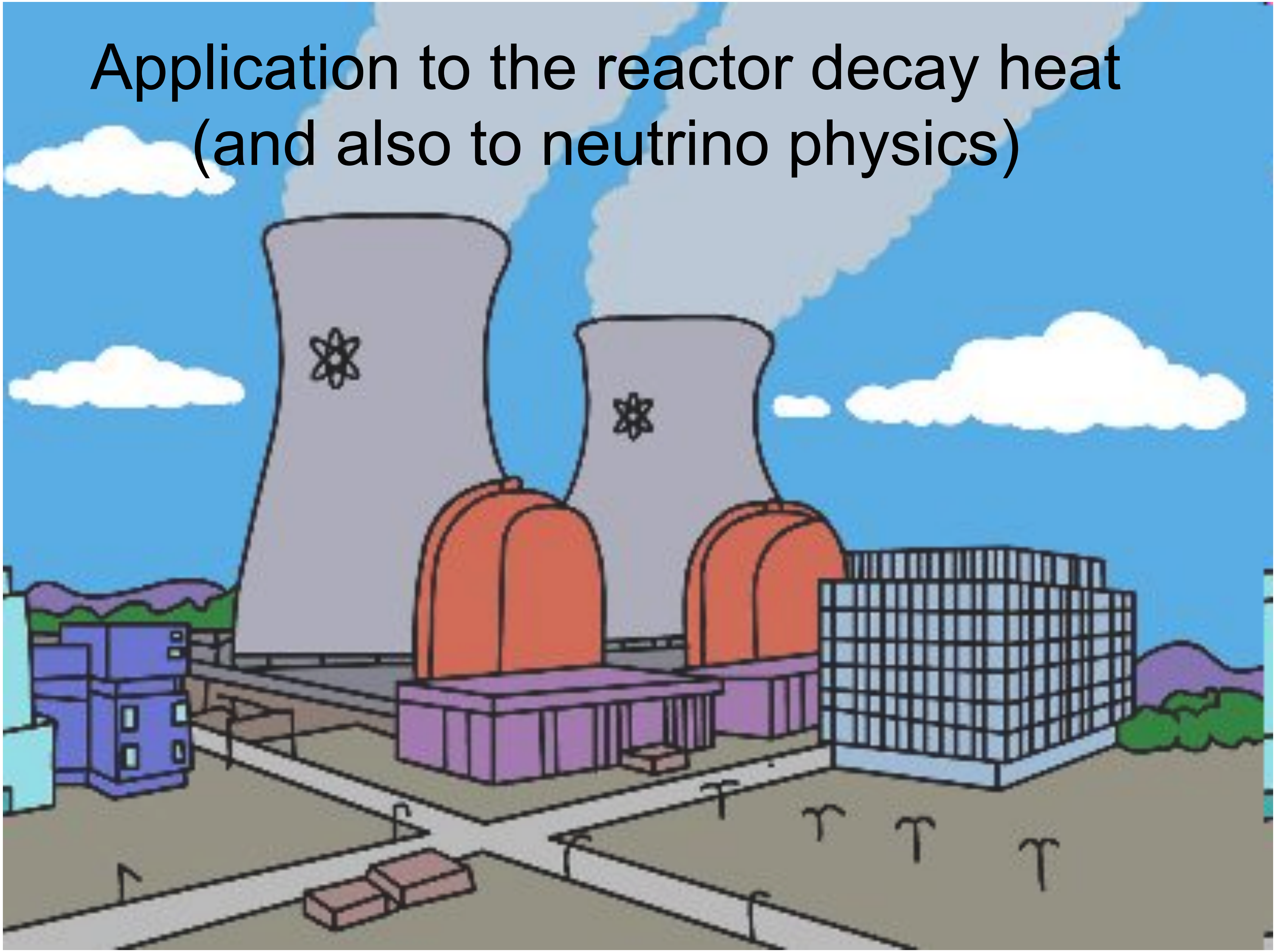
Expectation Maximization (EM) method:
modify knowledge on causes from effects

$$P(f_j \mid d_i) = \frac{P(d_i \mid f_j)P(f_j)}{\sum_j P(d_i \mid f_j)P(f_j)}$$

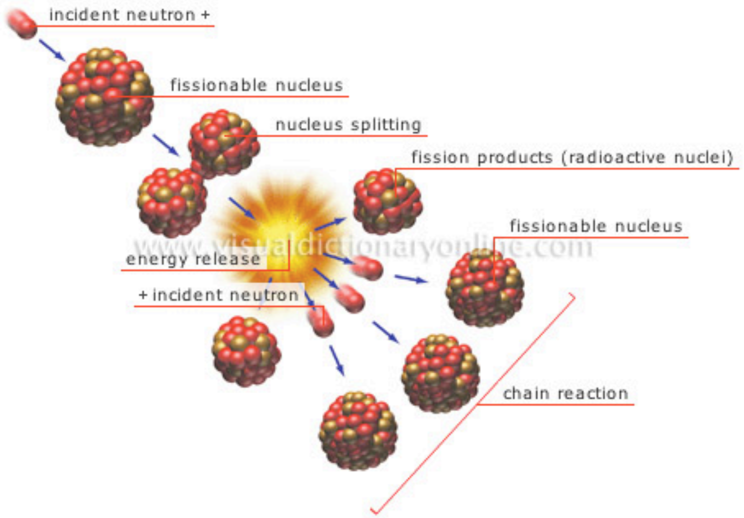
Algorithm:

$$f_j^{(s+1)} = \frac{1}{\sum_i R_{ij}} \sum_i \frac{R_{ij} f_j^{(s)} d_i}{\sum_k R_{ik} f_k^{(s)}}$$

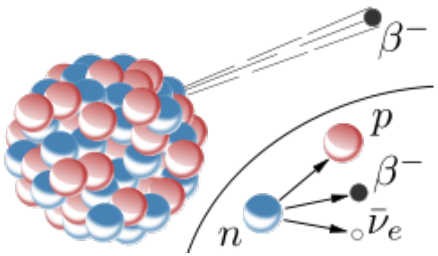
Application to the reactor decay heat (and also to neutrino physics)



Fission process energy balance and beta decay



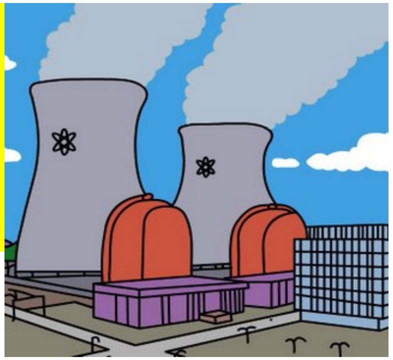
Each fission is approximately followed by 6 beta decays (sizable amount of energy released by the fission products)



Energy released in the fission of ^{235}U	
Energy distribution	MeV
Kinetic energy light fission fragment	100.0
Kinetic energy heavy fission fragment	66.2
Prompt neutrons	4.8
Prompt gamma rays	8.0
Beta energy of fission fragments	7.0
Gamma energy of fission fragments	7.2
Subtotal	192.9
Energy taken by the neutrinos	9.6
Total	202.7

James, J. Nucl. Energy 23 (1969) 517

Decay heat: how to determine it ?



- Measure it (lacks flexibility and it is costly)
- Try to predict or calculate in the best way
  - Statistical method (the first solution)

    Way and Wigner, Phys. Rev. 73 (1948) 1318

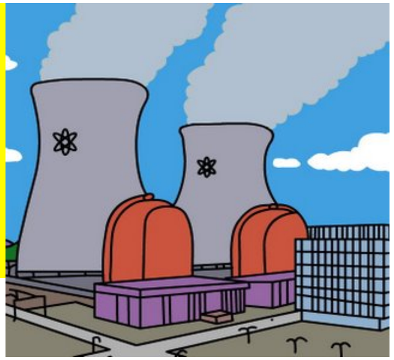
$$B(t) = 1.26 t^{-1.2} MeV/s$$

$$\Gamma(t) = 1.40 t^{-1.2} MeV/s$$

later, Griffin, Phys. Rev. 134 (1964) B817

  - Summation calculations (next slide)

Decay heat: summation calculations



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

E_i Decay energy of the nucleus i (gamma, beta or both)

λ_i Decay constant of the nucleus i $\lambda = \frac{\ln(2)}{T_{1/2}}$

N_i Number of nuclei i at the cooling time t

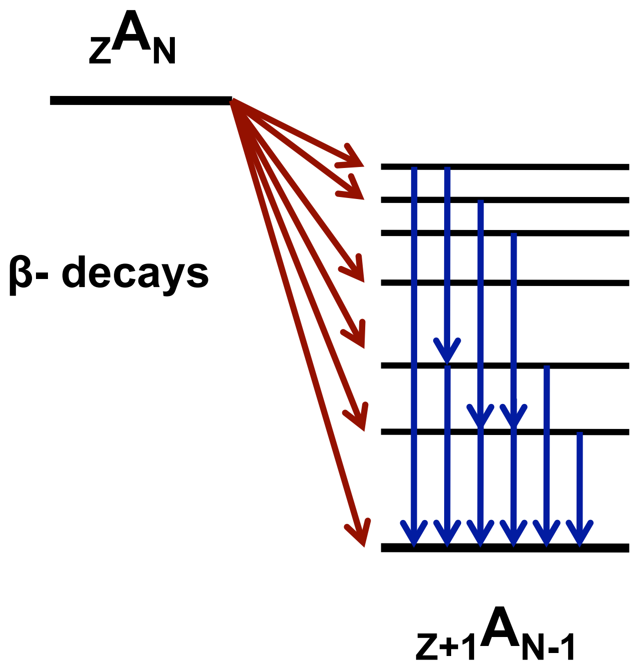
Requirements for the calculations: large databases that contain all the required information (half-lives, mean γ- and β-energies released in the decay, n-capture cross sections, fission yields, this last information is needed to calculate the inventory of nuclides)

How the mean energies are determined ?

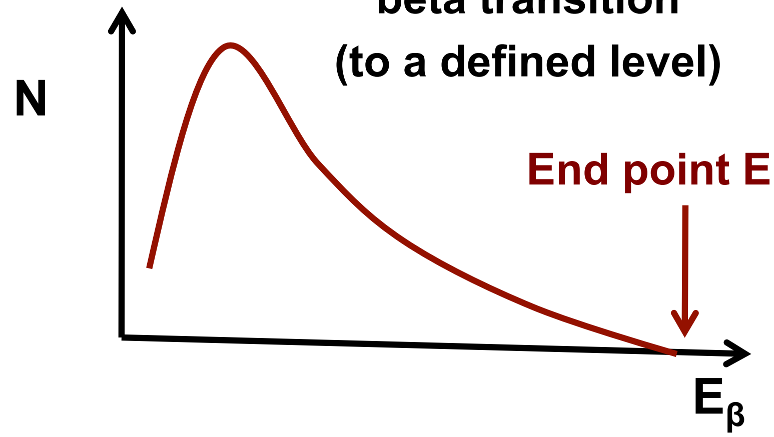
$$f(t) = \sum_i E_i \lambda_i N_i(t)$$

DATABASES:

feeding or beta decay prob. distributions



Beta spectrum for a beta transition (to a defined level)

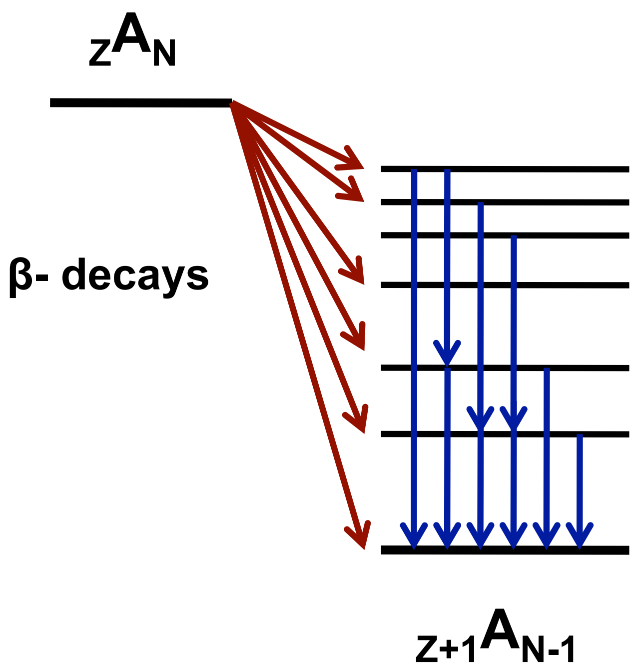


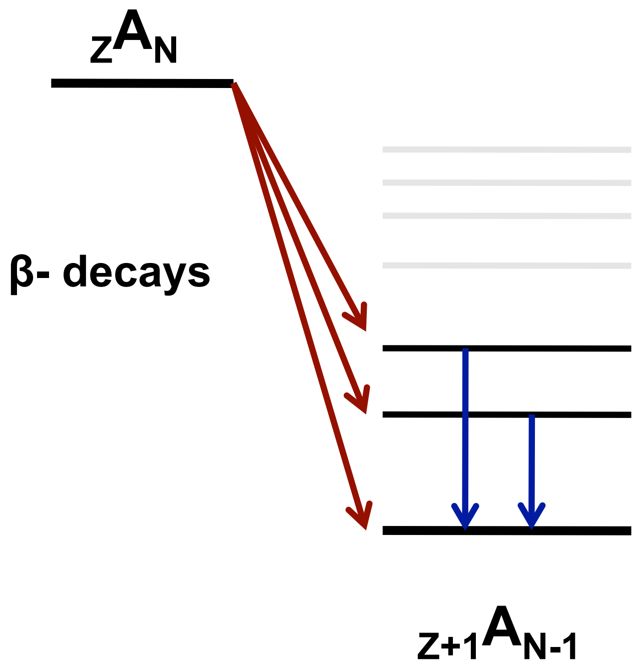
$$\overline{E}_\beta = \sum_i I_\beta(E_i)\langle E_{\beta,i}\rangle$$

$$\overline{E}_\gamma = \sum_i I_\beta(E_i) E_i$$

Mean energies and Pandemonium

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





$$\overline{E}_\beta = \sum_i I_\beta(E_i)\langle E_{\beta,i}\rangle$$

$$\overline{E}_\gamma = \sum_i I_\beta(E_i)E_i$$

$\overline{E}_\beta$ overestimation

$\overline{E}_\gamma$ underestimation

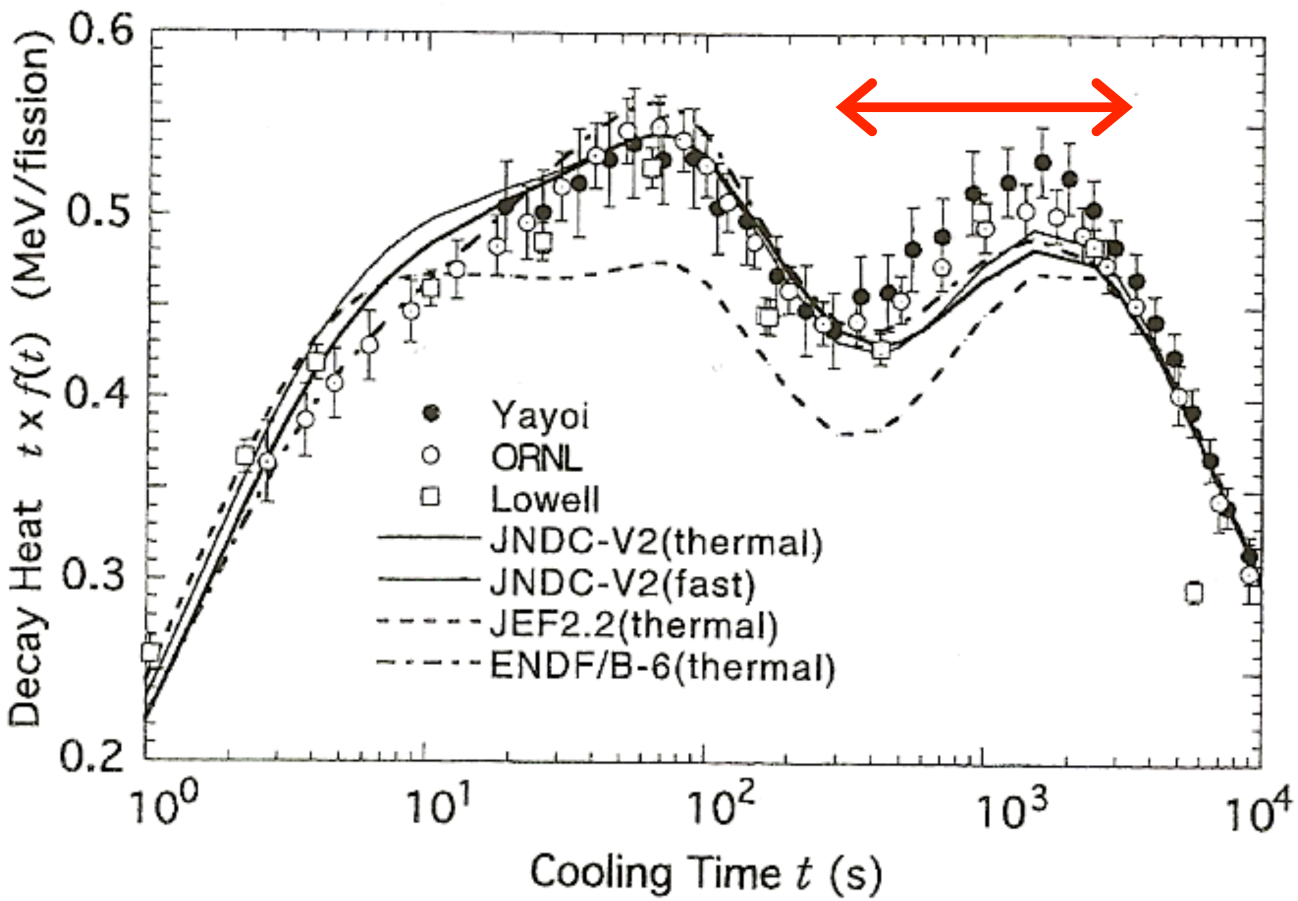
The beginning …

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 $^{102,104,105}Tc$

^{239}Pu example (γ component)

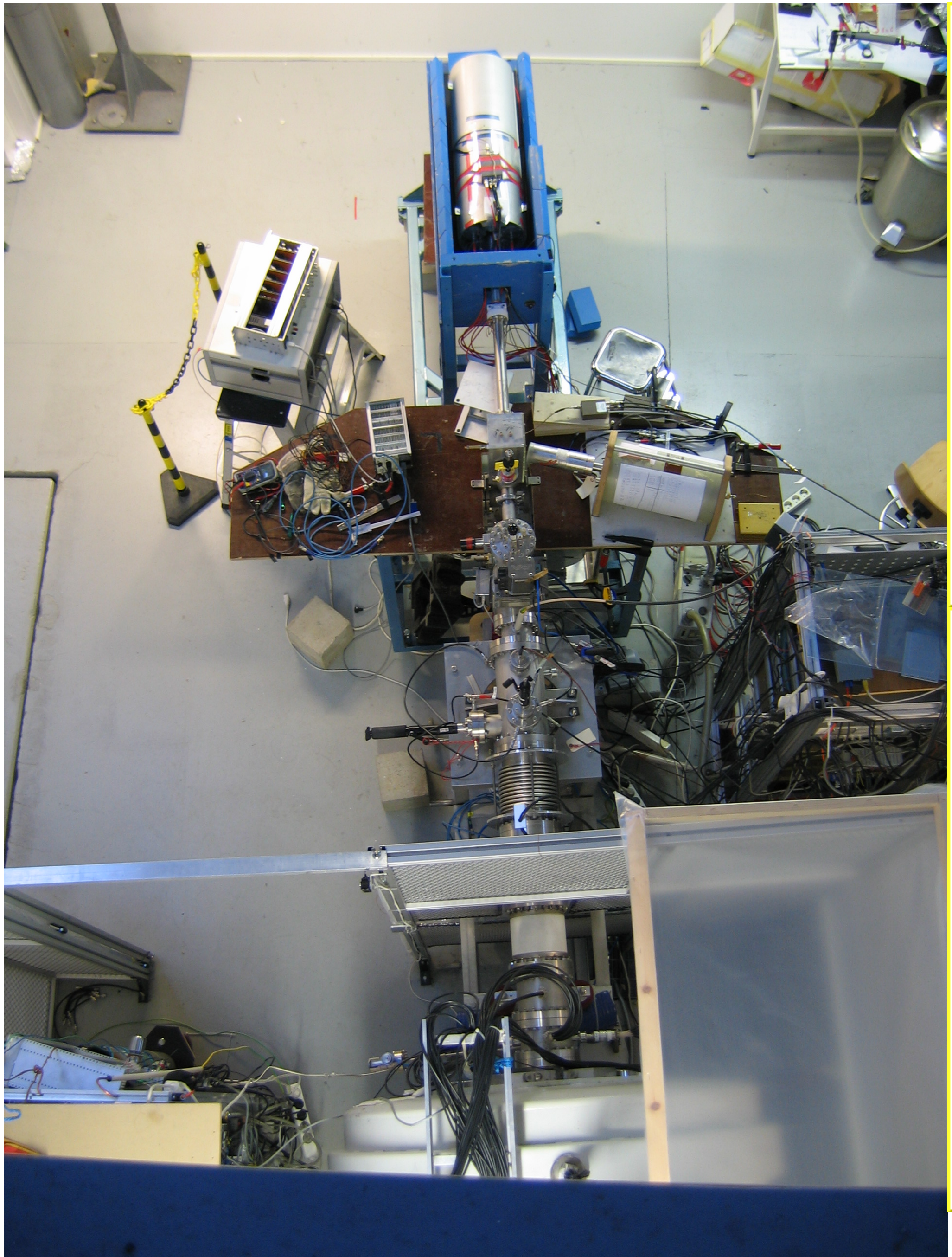


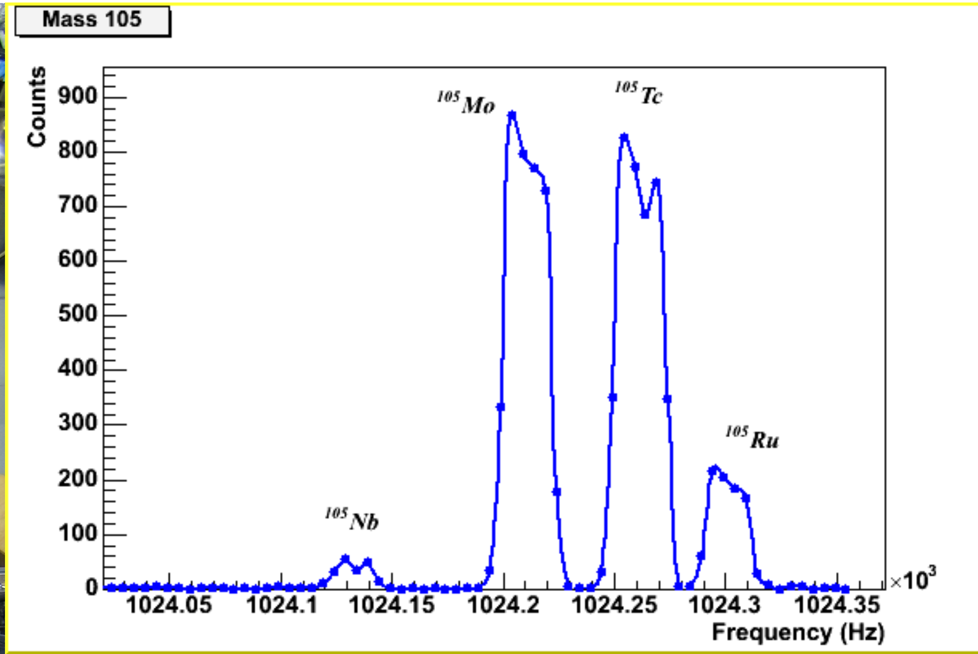
The "famous" list
WPEC-25 (IAEA working group)

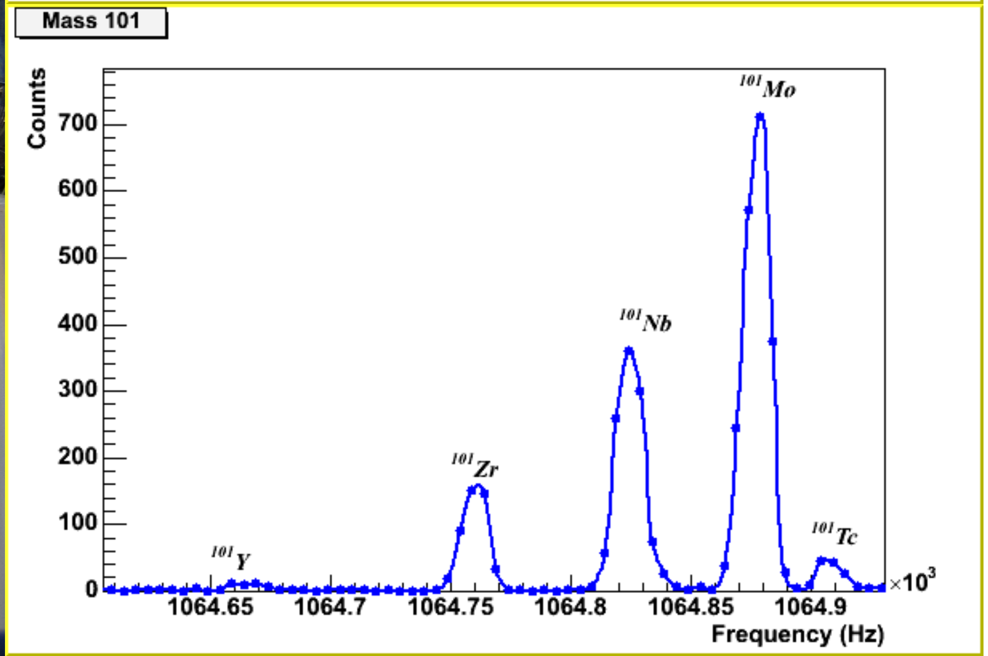
Radionuclide	Priority	Radionuclide	Priority	Radionuclide	Priority
35-Br-86	1	41-Nb-99	1	52-Te-135	2
35-Br-87	1	41-Nb-100	1	53-I-136	1
35-Br-88	1	41-Nb-101	1	53-I-136m	1
36-Kr-89	1	41-Nb-102	2	53-I-137	1
36-Kr-90	1	42-Mo-103	1	54-Xe-137	1
37-Rb-90m	2	42-Mo-105	1	54-Xe-139	1
37-Rb-92	2	43-Tc-102	1	54-Xe-140	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		

37 nuclides, of which 23 were given first priority, reports by A. Nichols et al. (IAEA).

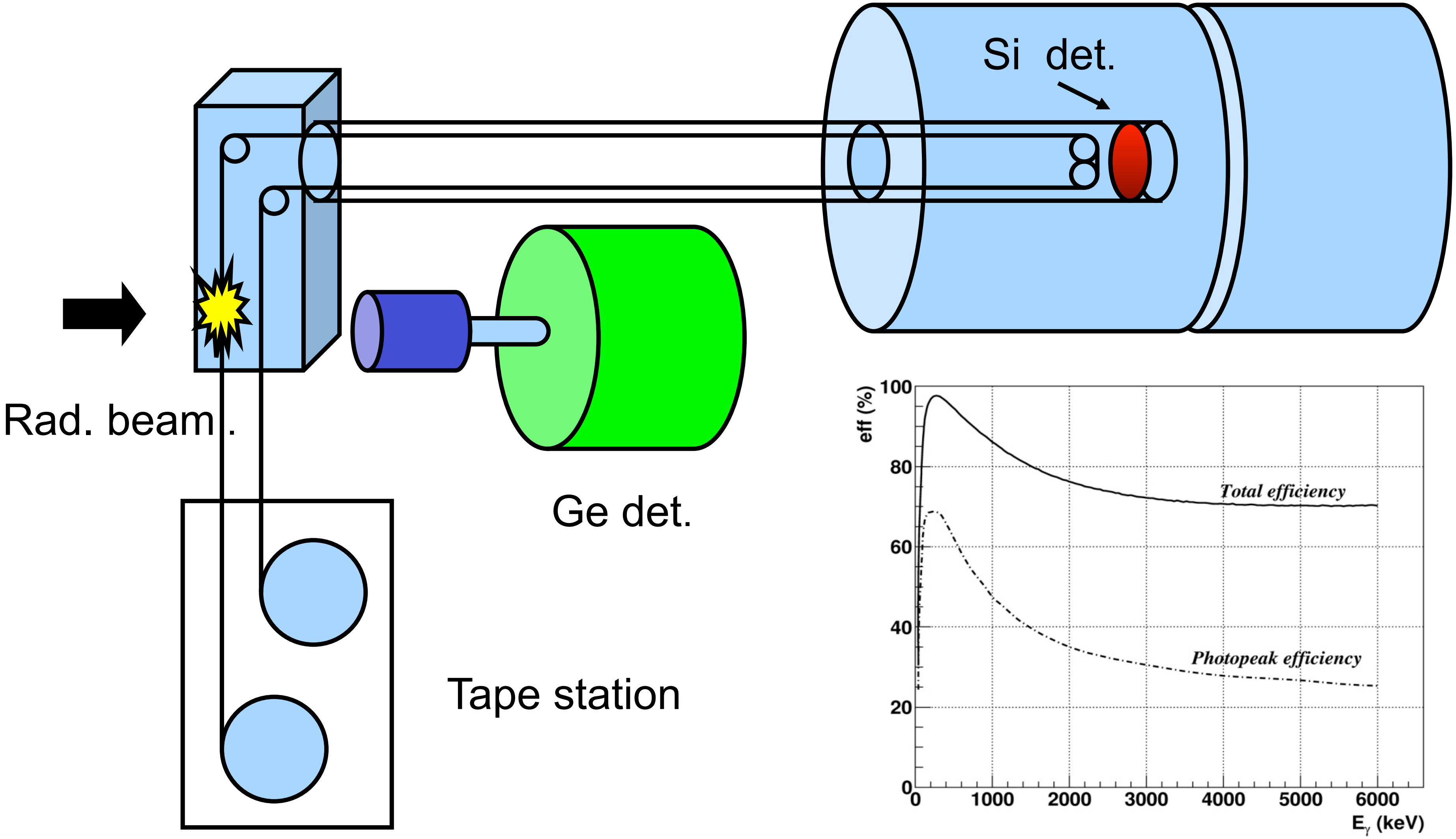
New feature: IGISOL + trap-assisted spectroscopy



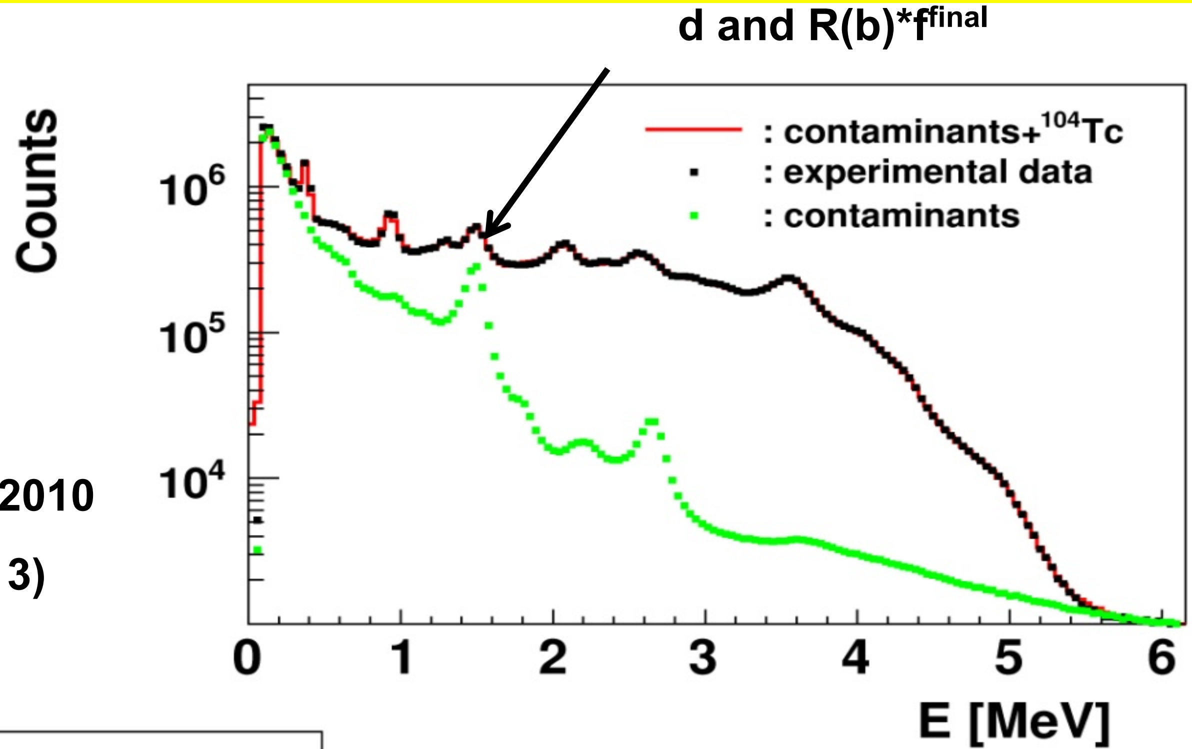




TAS experimental setup at Jyväskylä

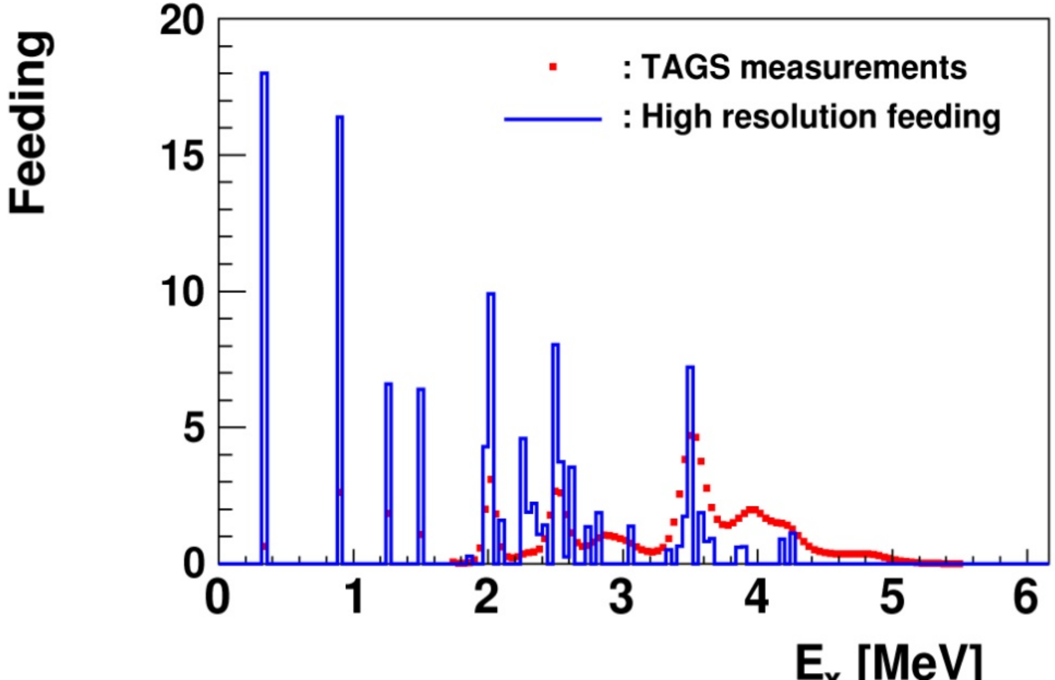


Results of the analysis for ^{104}Tc



D. Jordan, PhD Thesis, Valencia, 2010

D. Jordan, PRC 87, 044318 (2013)



$T_{1/2}$ = 1098(18) s; Q_β= 5516(6) keV

E_β(TAGS) = 931 (10) keV
E_β(JEFF-3.1) = 1595 (75) keV
ΔE_β = -664 keV

E_γ(TAGS) = 3229 (24) keV
E_γ(JEFF-3.1) = 1890 (31) keV
ΔE_γ = 1339 keV

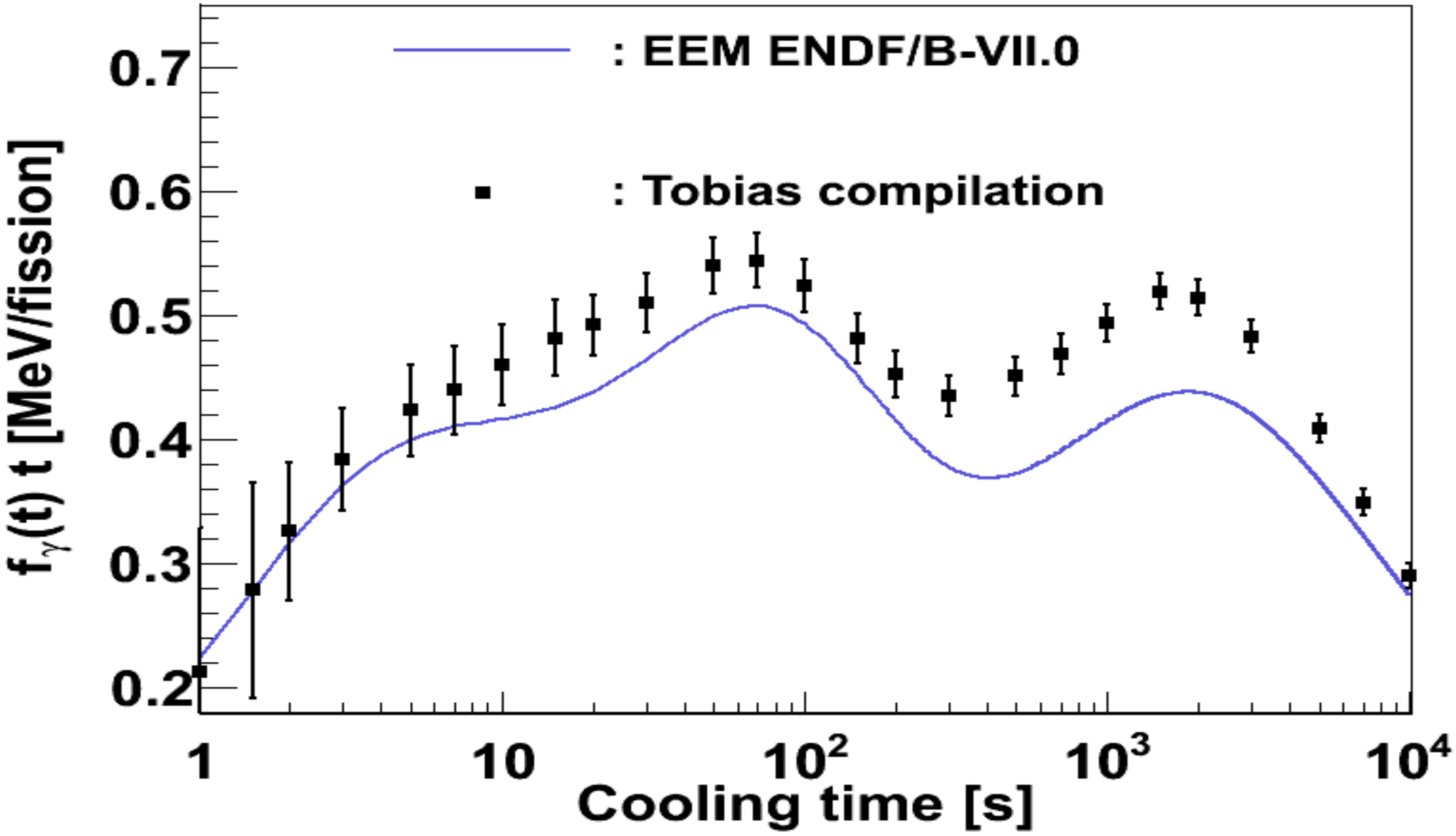
All results published up to now

Isotope	Energy type	TAGS [keV]	JEFF-3.1 [keV]	ENDF/B-VII [keV]	Difference [keV]
^{101}Nb (7.1 s)	beta	1797 (133)	1863 (307)	1966 (307)	-67/-169
	gamma	445 (279)	245 (22)	270 (22)	200/175
^{102}Tc (5.28 s)	beta	1935 (11)	1945 (16)	1945 (16)	-10
	gamma	106 (23)	81 (5)	81 (5)	25
^{104}Tc (1098 s)	beta	931 (10)	1595 (75)	1595 (75)	-664
	gamma	3229 (24)	1890 (31)	1890 (31)	1339
^{105}Tc (456 s)	beta	764 (81)	1310 (173)	1310 (205)	-546
	gamma	1825 (174)	668 (19)	665 (19)	1157/1160
^{105}Mo (35.6 s)	beta	1049 (44)	1922 (122)	1922 (122)	-873
	gamma	2407 (93)	551 (24)	552 (24)	1856/1855
^{106}Tc (35.6 s)	beta	1457 (30)	1943 (69)	1906 (67)	-486/-449
	gamma	3132 (70)	2191 (51)	2191 (51)	941
^{107}Tc (21.2 s)	beta	1263 (212)	2056 (254)	2054 (254)	-793/-791
	gamma	1822 (450)	515 (11)	515 (11)	1307

$Q_\beta(^{102}Tc \rightarrow ^{102}Ru) = 4532 keV$ $\quad$ $Q_\beta(^{101}Nb \rightarrow ^{101}Mo) = 4569 keV$

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

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



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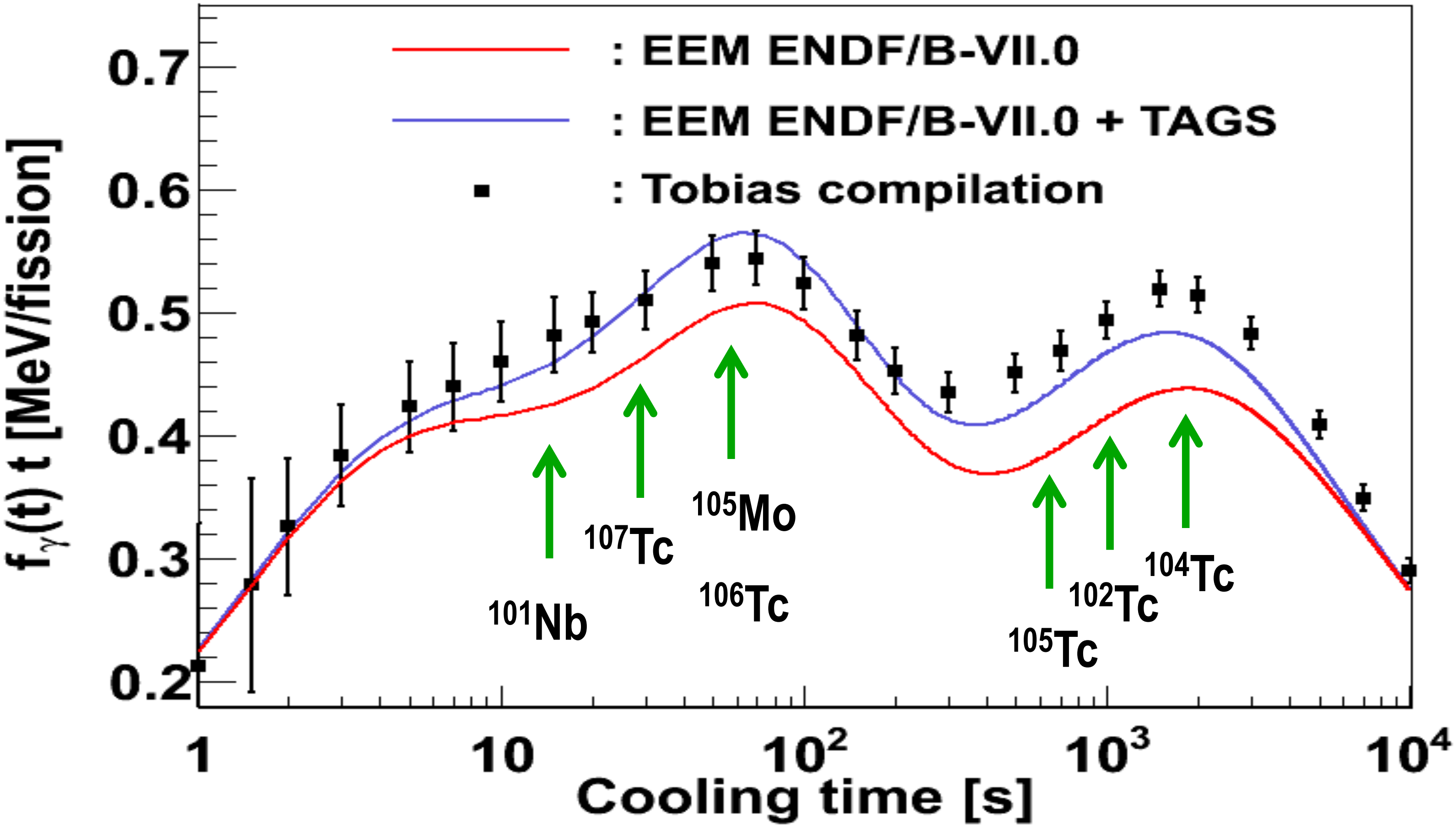


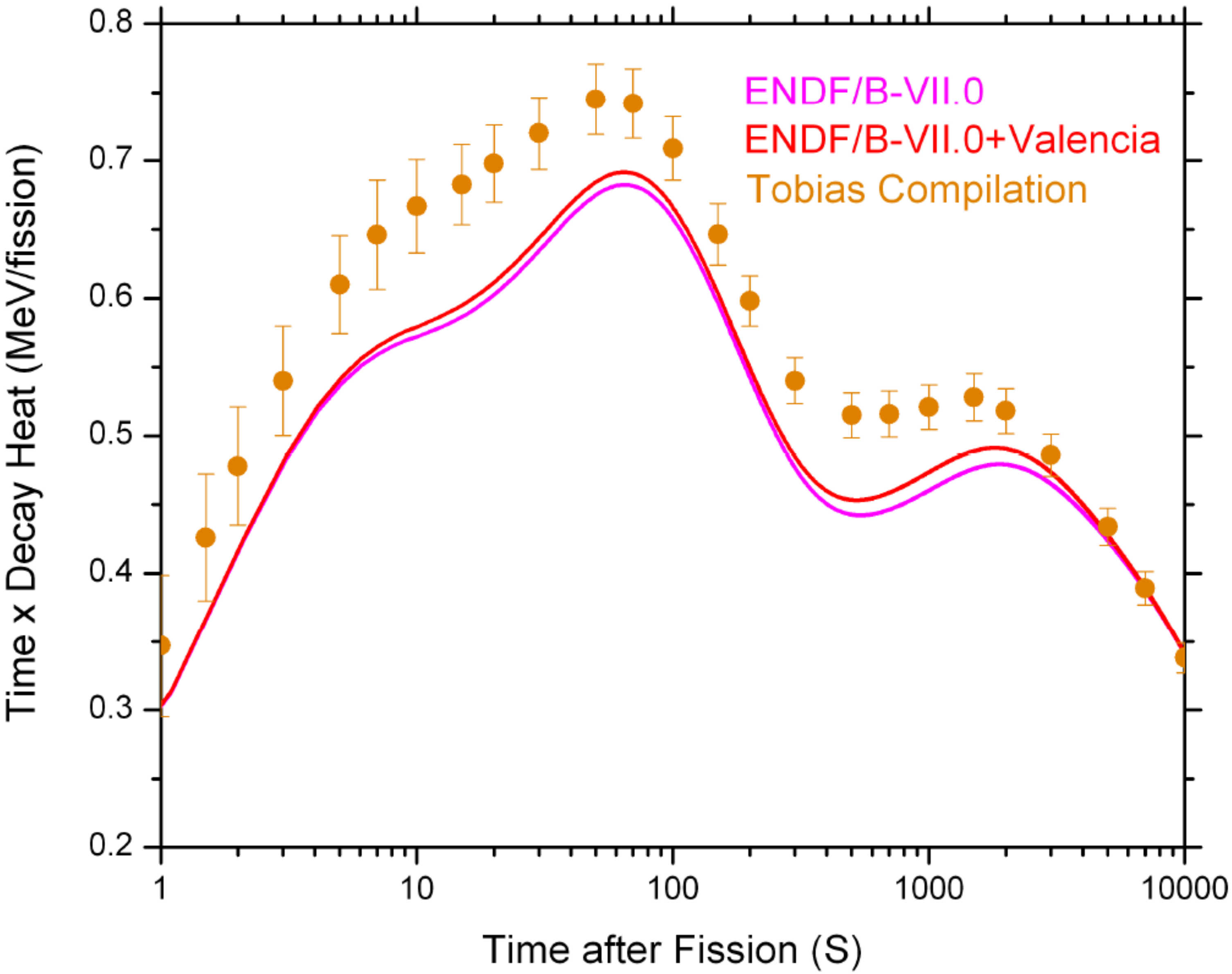
DH Courtesy A. Sonzogni

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

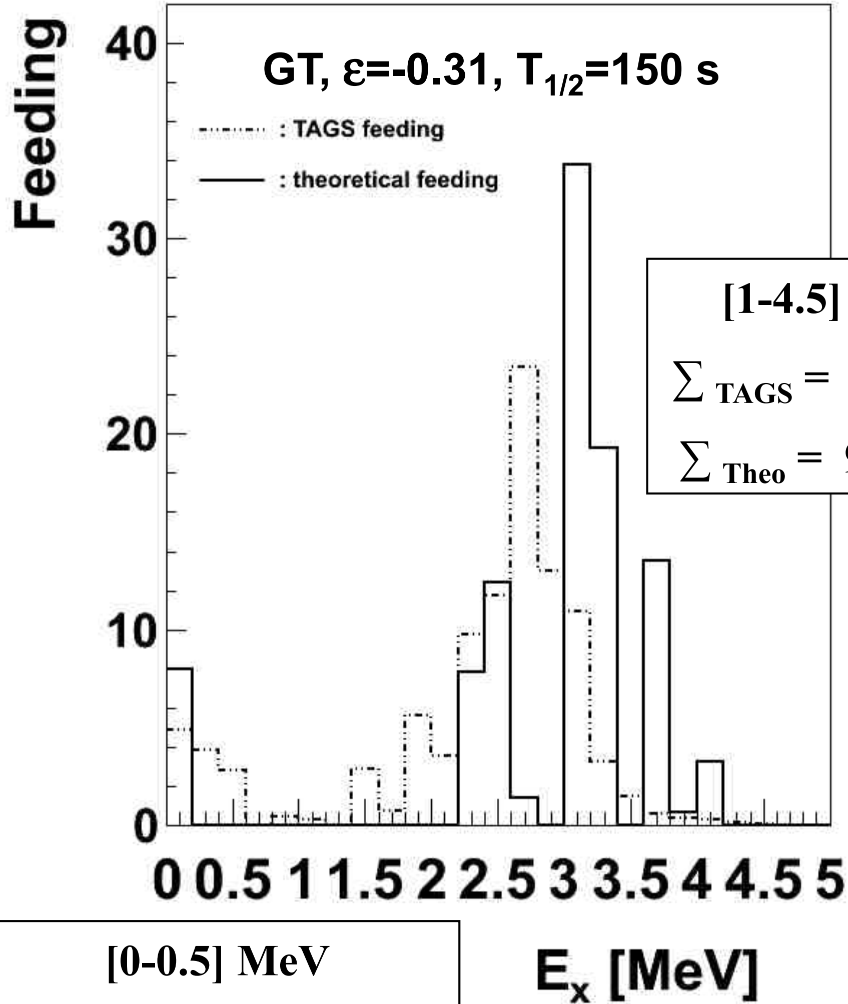
K. P. Rykaczewsky, Physics 3, 94 (2011)

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

Impact of the results for ^{235}U



Results of QRPA calculations
^{105}Mo, $T_{1/2}$(exp) = 35.6 s



[1-4.5] MeV

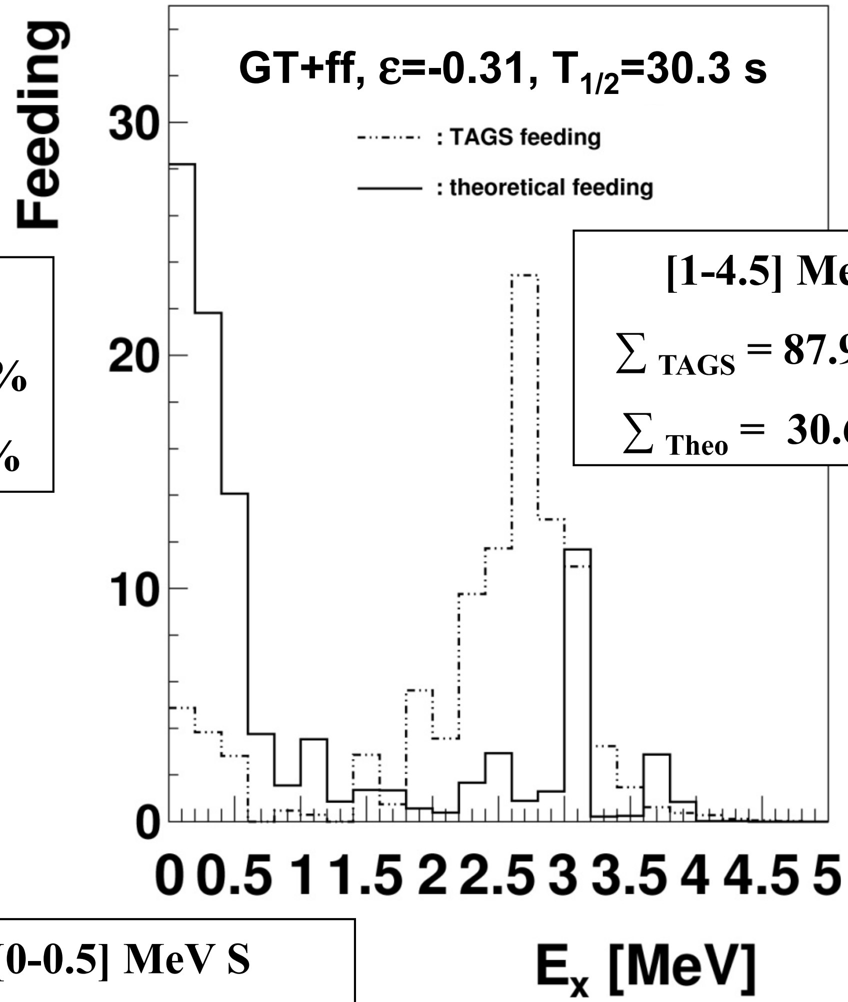
$\sum_{TAGS}$ = 87.99%

$\sum_{Theo}$ = 92.05%

[0-0.5] MeV

$\sum_{TAGS}$ = 11.51%

$\sum_{Theo}$ = 7.94%



[1-4.5] MeV

$\sum_{TAGS}$ = 87.99 %

$\sum_{Theo}$ = 30.62%

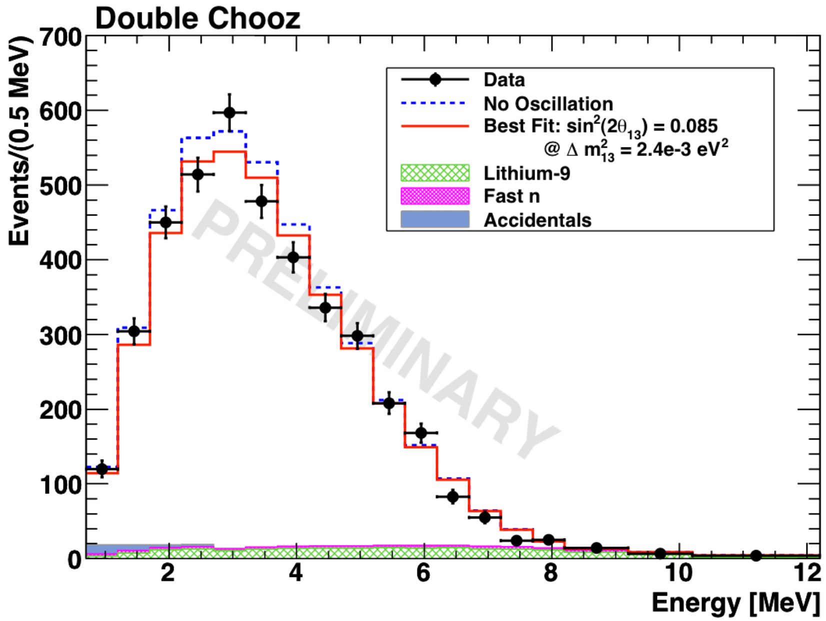
[0-0.5] MeV S

$\sum_{TAGS}$ = 11.51%

$\sum_{Theo}$ = 67.84 %

Kratz *et al.*

Reactor neutrino experiments: summation calculations



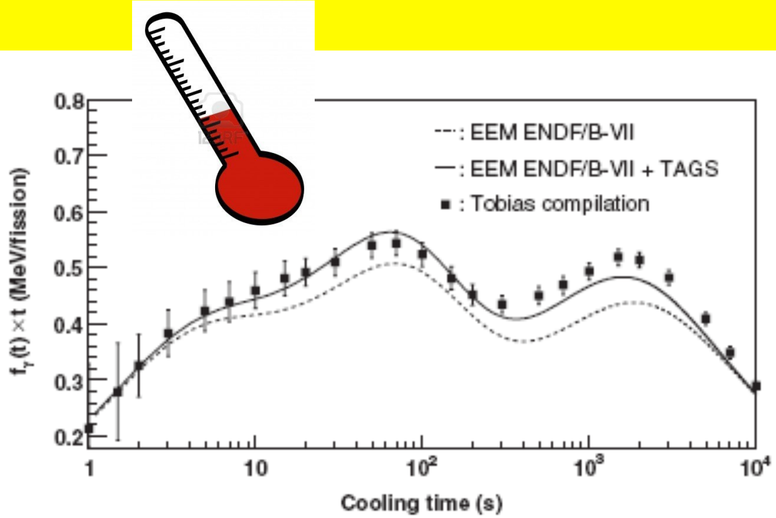
$$N(E_\nu) = \sum_n Y_n(Z,A,t) \cdot \sum_i b_{n,i}(E_0^i) P_\nu(E_\nu, E_0^i, Z)$$

Y_n Number of beta decays per unit time of fragment with Z, A (cumm. Yield)

$b_{n,I}$ branching ratio of the i branch with maximum electron energy E^i_0

$P\nu$ neutrino spectrum of the i branch with maximum electron energy E^i_0

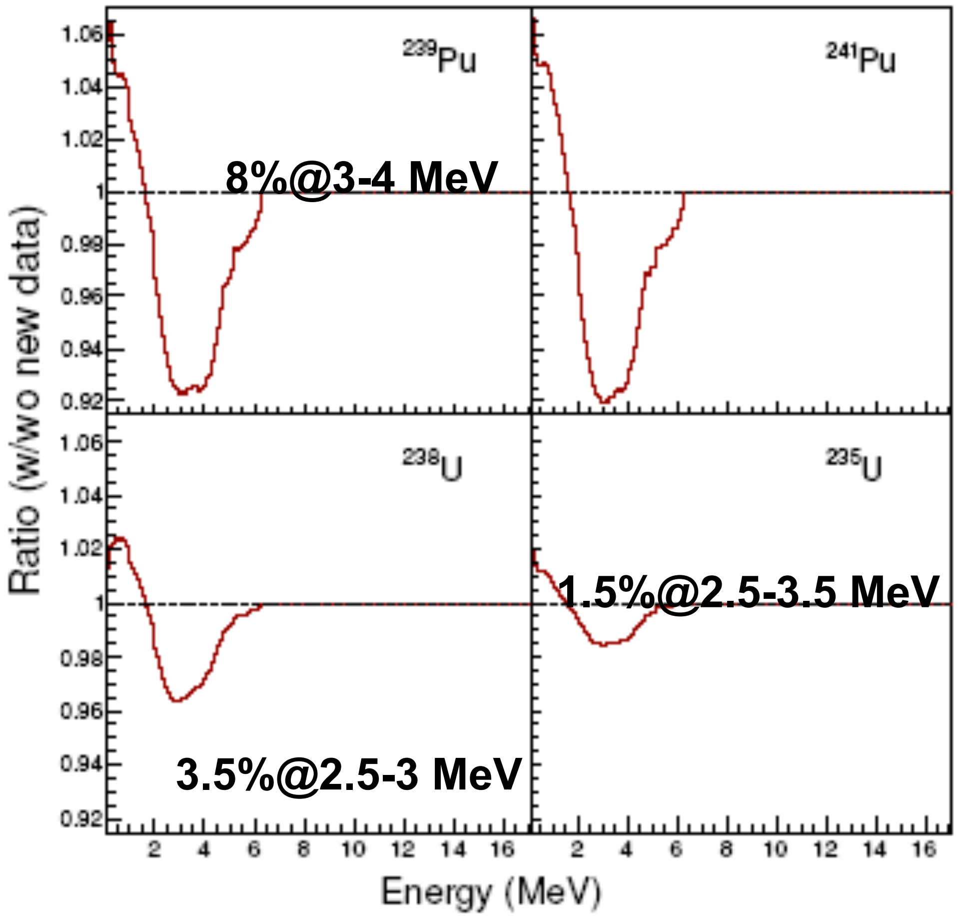
Some additional impact of our data



Algora et al., PRL 105.202501

Dolores Jordan, PhD thesis, 2010

M. Fallot et al., PRL 109.202504

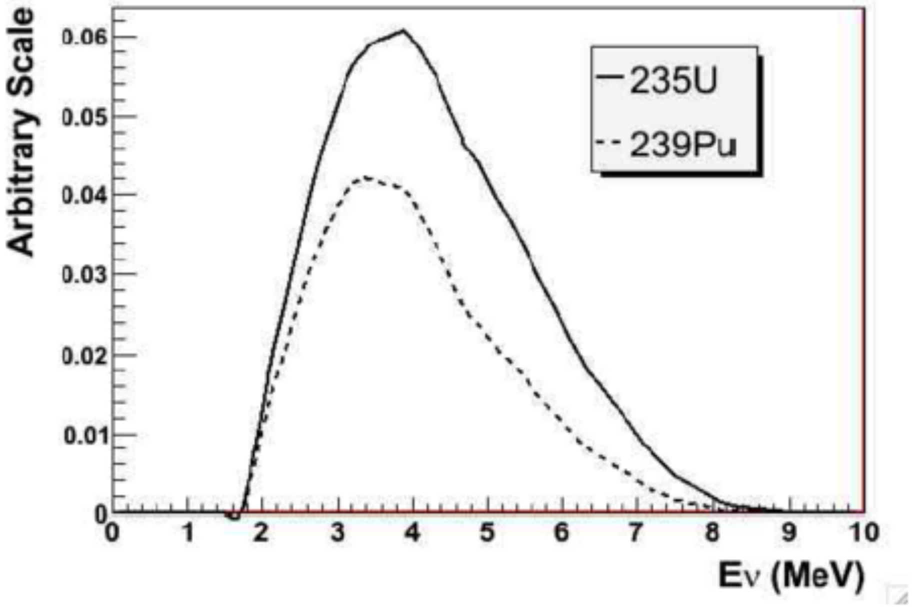


Ratio between 2 antineutrino spectra built with and without the ^{102,104,105,106,107}Tc, ^{105}Mo, ^{101}Nb TAS data

Another application: prediction of the neutrino spectrum from reactors for non-proliferation

	235U	239Pu
Released E per fission	201.7 MeV	210.0 MeV
Mean neutrino E	2.94 MeV	2.84 MeV
Neutrinos/fission >1.8 MeV	1.92	1.45
Aver. Int. cross section	$3.2 \times 10^{-43} cm^2$	$2.8 \times 10^{-43} cm^2$

$$\bar{\nu} + p \rightarrow e^{+} + n$$ (threshold 1.8 MeV)



•Relevance for non-proliferation studies (working group of the IAEA). Neutrino flux can not be shielded. Study to determine fuel composition and power monitoring. Non-intrusive and remote method.

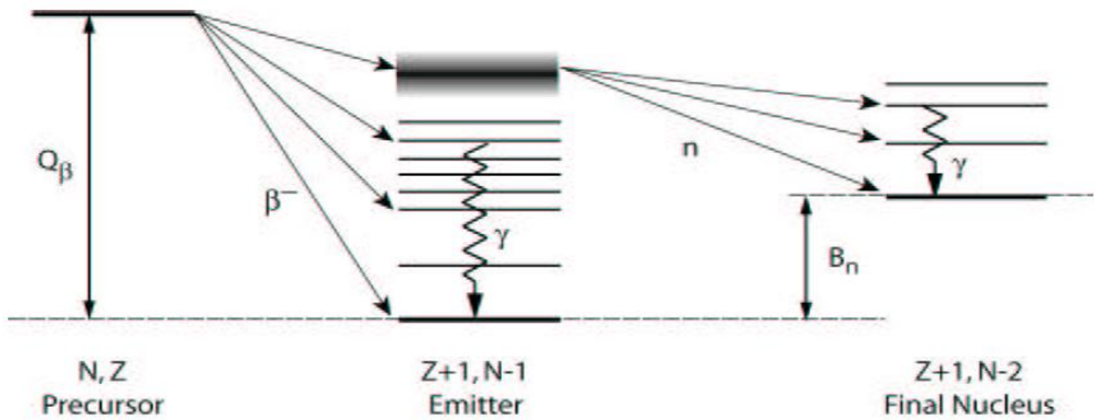
•Approved proposal to study some nuclides related to this problem (IGISOL, trap assisted TAS) (Fallot, Tain, Algora)

Motivation of recently analyzed cases: $^{87}Br, ^{88}Br$

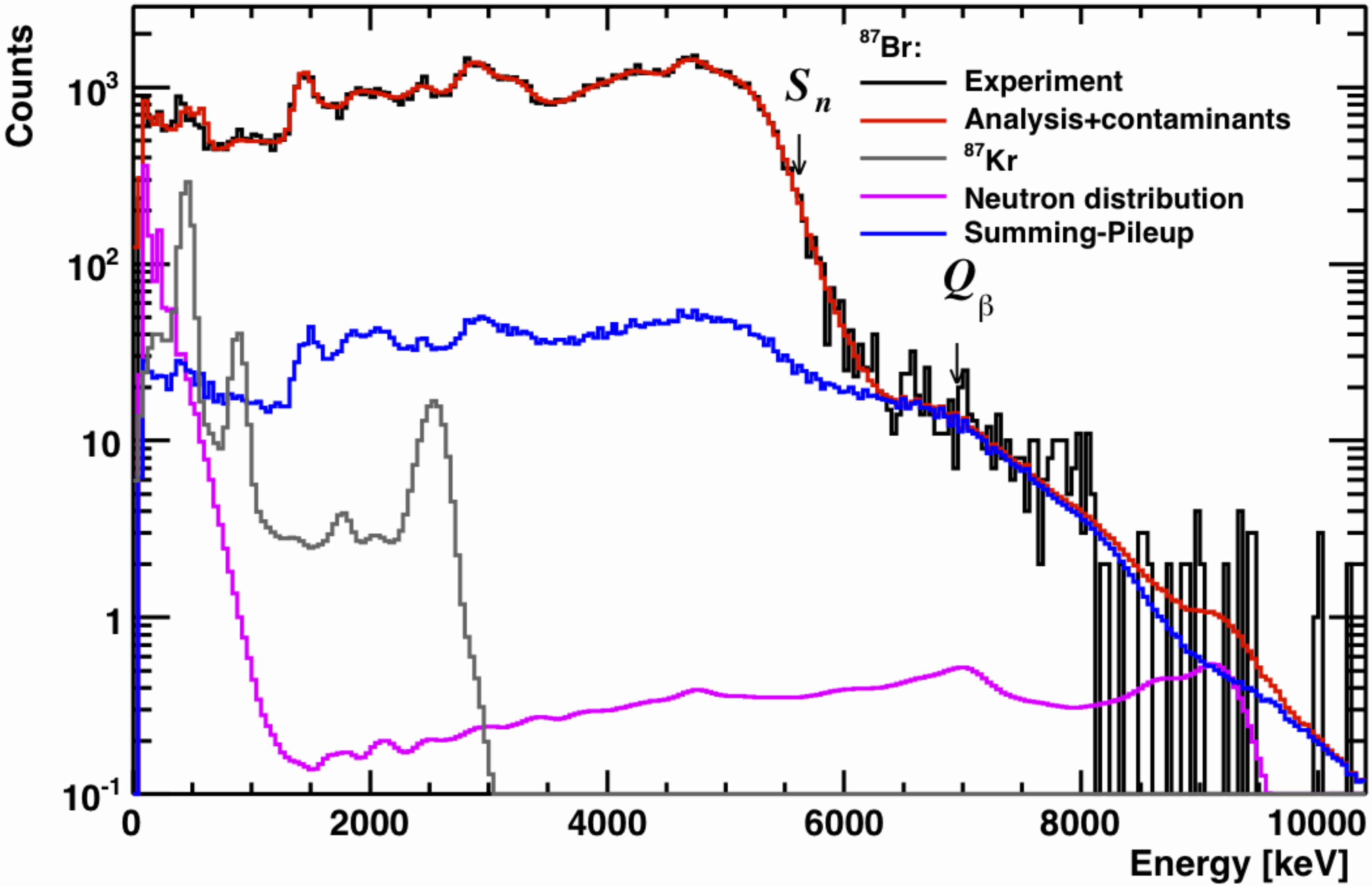
- Priority one in the IAEA list
- Moderate fission yields
- Pandemonium cases ?
- Interest from the structure point of view: vicinity of n closed shell
- Competition between gamma and neutron emission above the Sn value

$$\frac{1}{T_{1/2}} = \int_{0}^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) dE_x$$

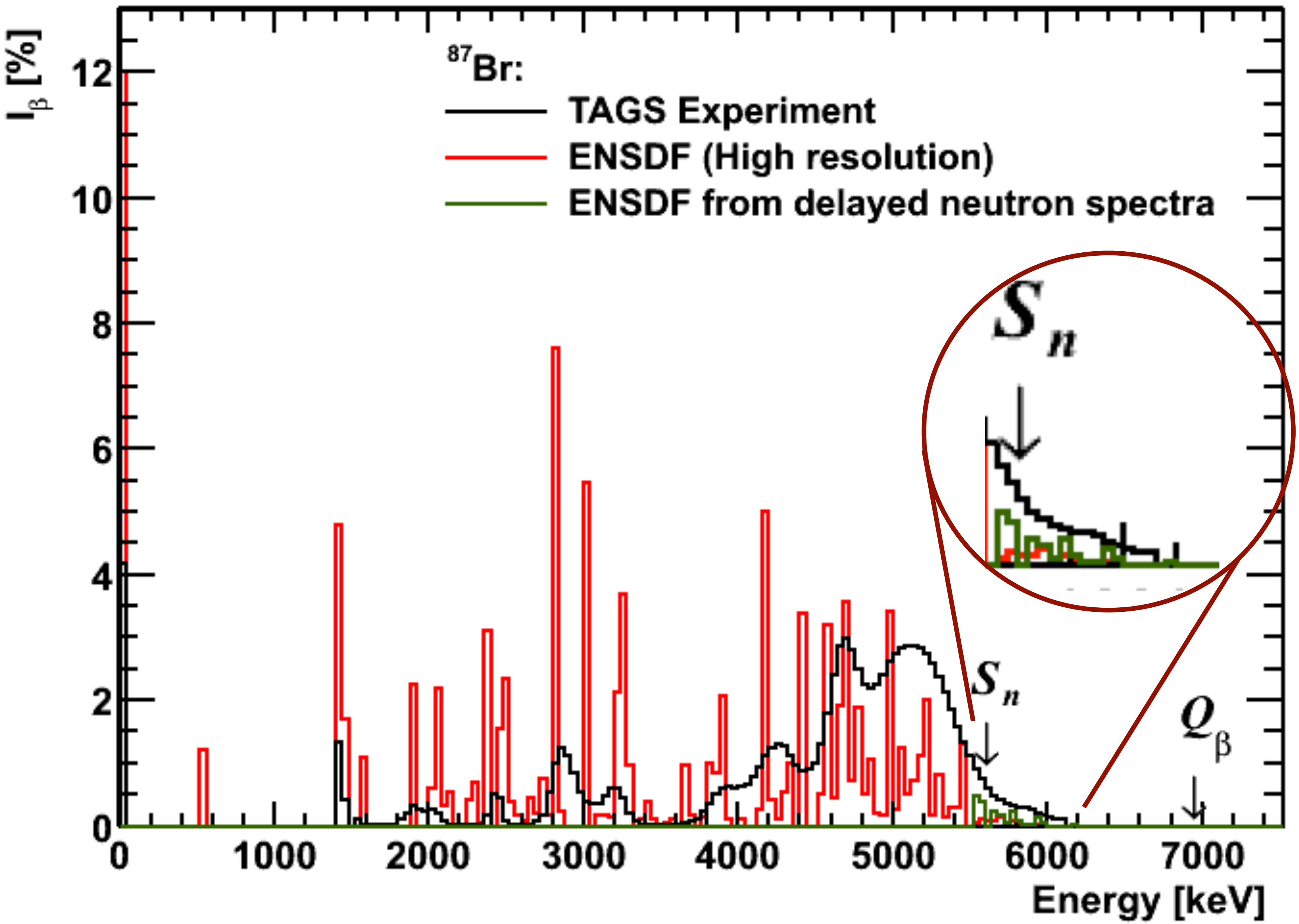
$$P_n = \frac{\int_{S_n}^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) \frac{\Gamma^n}{\Gamma^n + \Gamma^\gamma} dE_x}{\int_{0}^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x) dE_x}$$



^{87}Br: meas. spectrum + contaminants + analysis



Deduced feedings from 87Br decay



^{87}Br feedings and mean energies (very preliminary !)

	ENDF	TAGS
$<E_\beta>$[keV]	1656(75)	1017(16)
$<E_\gamma>$ [keV]	3345(35)	4242(30)
% above Sn	0.58	< 5.4 %

Q_β=6817(5) keV

Sn= 5515.4(8)

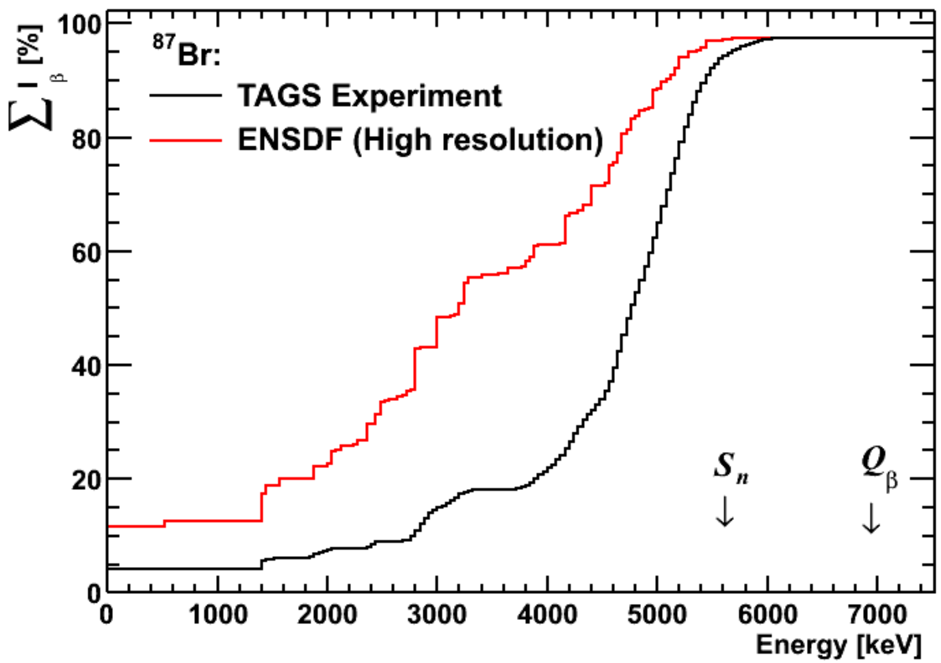
$T_{1/2}$=55.65(13) s

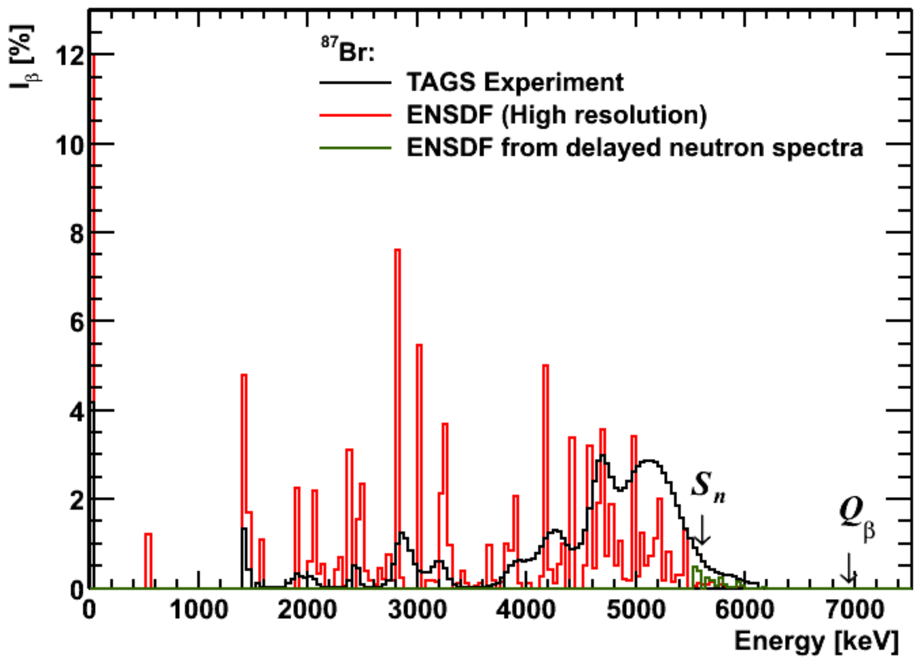
Pn (^{87}Br) = 2.52(7)%

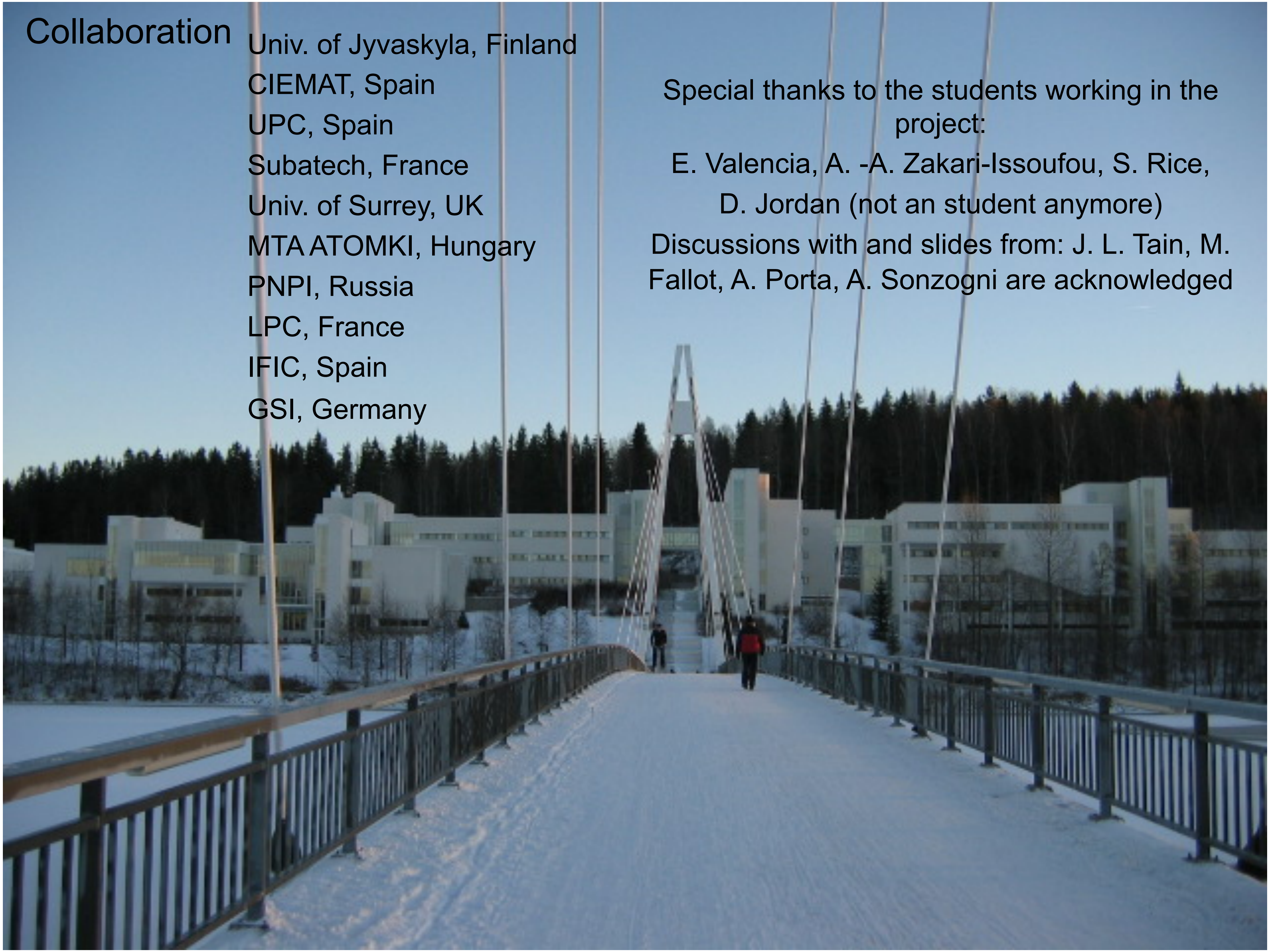
Cum fiss. (^{235}U) =0.02

Cum fiss.(^{239}Pu) =0.005

Nuh et al. I_{gam}/I_n~0.9







Collaboration

- Univ. of Jyvaskyla, Finland
- CIEMAT, Spain
- UPC, Spain
- Subatech, France
- Univ. of Surrey, UK
- MTA ATOMKI, Hungary
- PNPI, Russia
- LPC, France
- IFIC, Spain
- GSI, Germany

Special thanks to the students working in the project:

E. Valencia, A. -A. Zakari-Issoufou, S. Rice,

D. Jordan (not an student anymore)

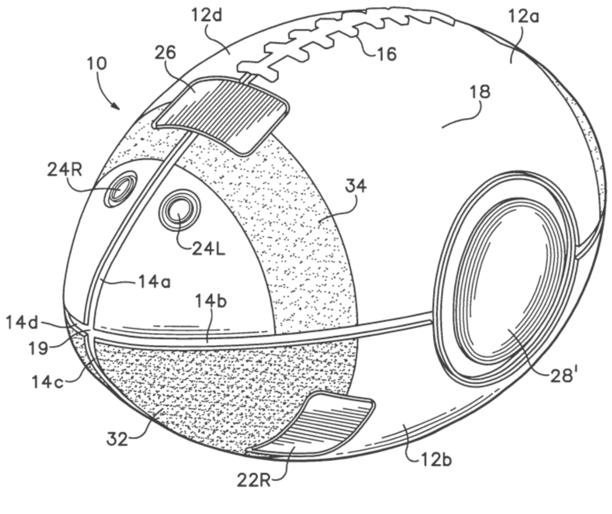
Discussions with and slides from: J. L. Tain, M. Fallot, A. Porta, A. Sonzogni are acknowledged

Nuclear Shapes

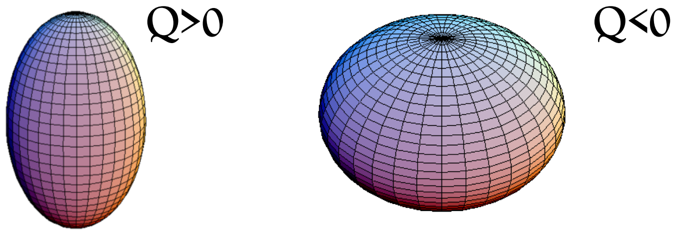
Experimentally how do we deduce nuclear shapes ?

What can beta decay offer ?

In any question related to nuclear shapes, you should remember that the answer is always model dependent

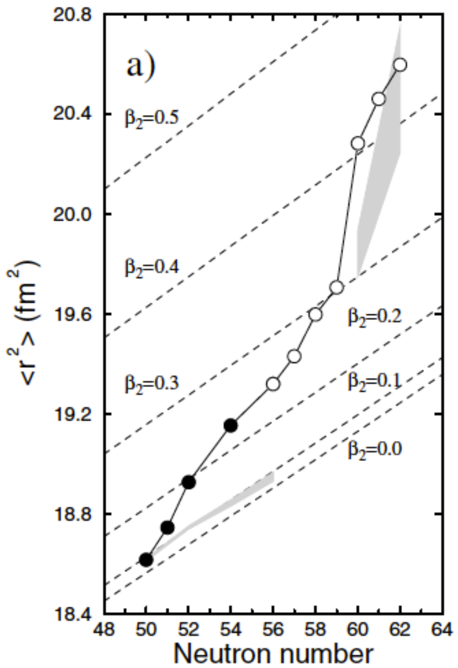


Experimentally
how the shapes of nuclei are determined ?



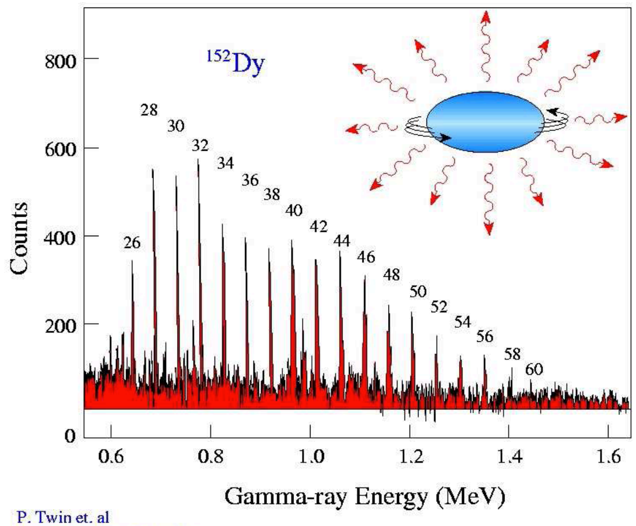
$$Q_s = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} Q_0$$

- Nuclear electric quadrupole moments
- Nuclear radii measurements by means of isotope shifts (muonic atoms, laser spectroscopy)
- Nuclear spectroscopy methods (life-time meas., fast-timing, electron-conversion measurements, etc.)



Campbell
PRL 89, 2002

Laser spectroscopy of cooled Zr fission products (droplet model)

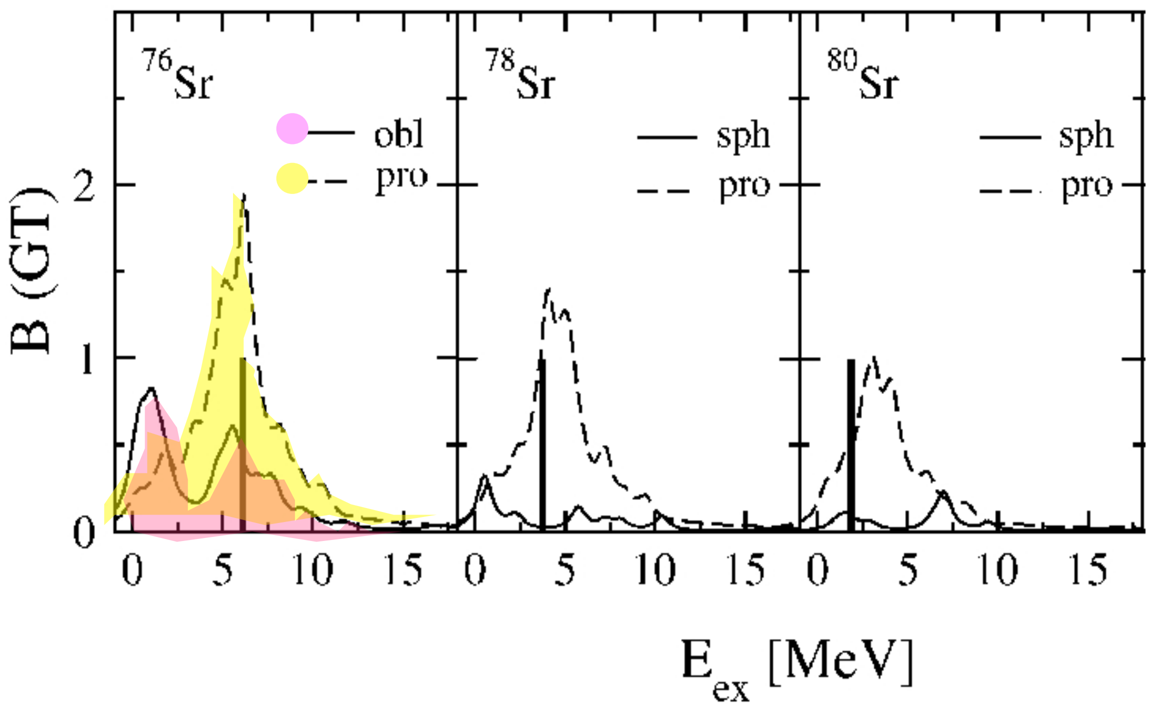


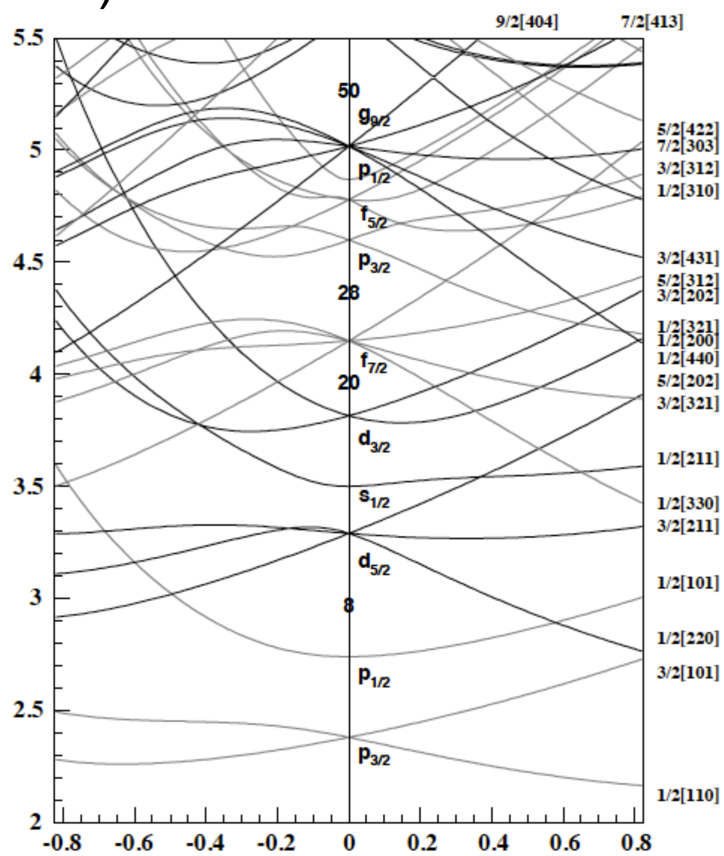
P. Twin et. al
Phys. Rev. Lett. 57 (1986)

What can beta decay offer apart from spectroscopy …

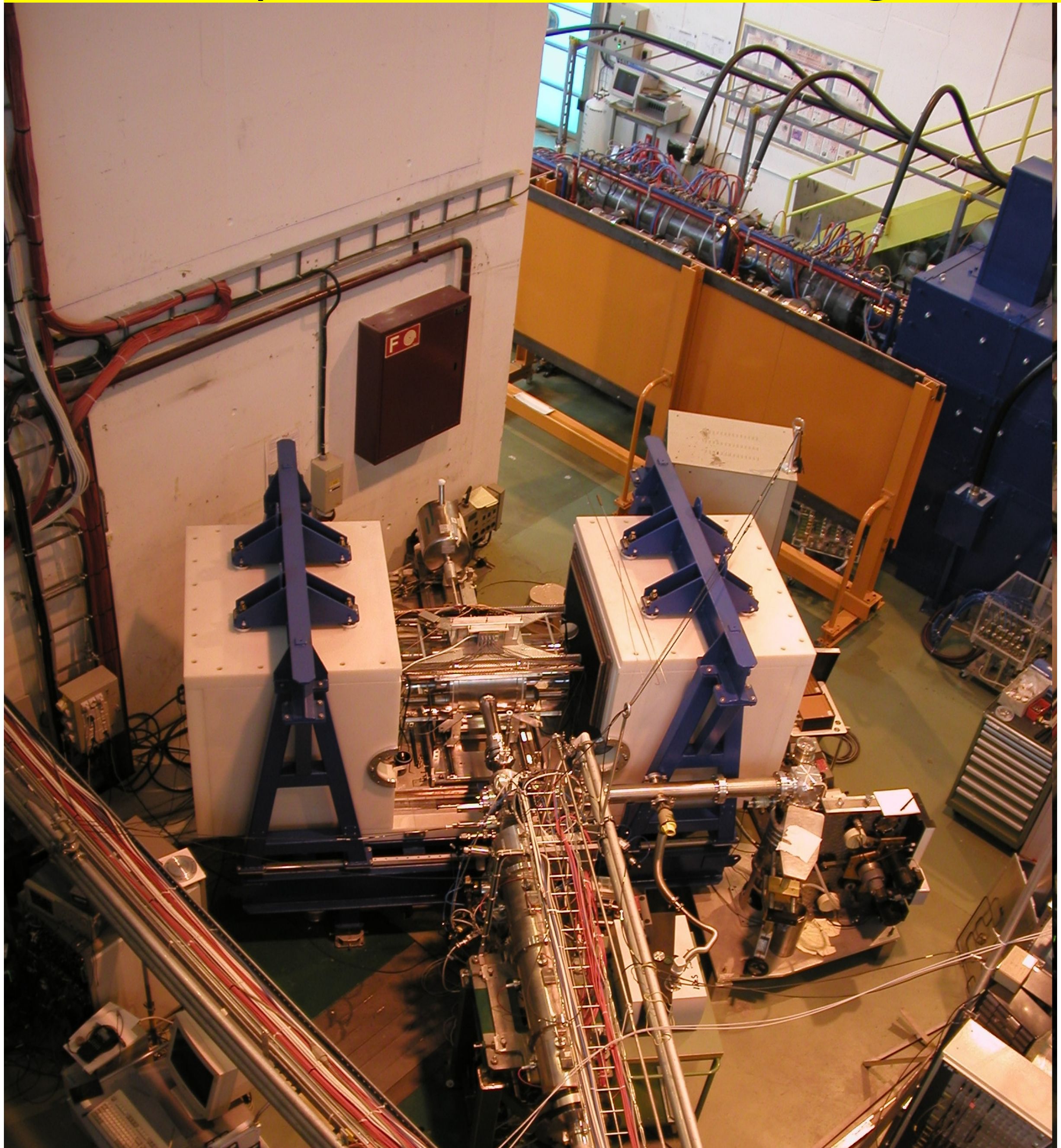
One alternative, based in the pioneering work of I. Hamamoto, (Z. Phys. A353 (1995) 145) later followed by studies of P. Sarriguren *et al*., Petrovici *et al*. is related to the dependency of the strength distribution in the daugther nucleus depending on the shape of the parent. It can be used when theoretical calculations predict different B(GT) distributions for the possible shapes of the ground state (prolate, spherical, oblate).

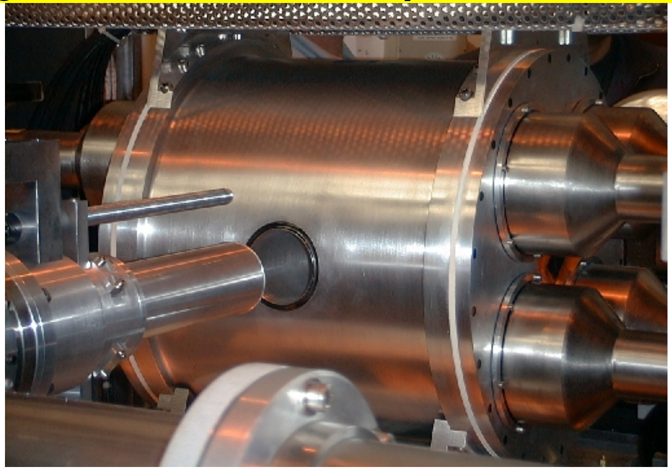
P. Sarriguren *et al*., Nuc. Phys. A635 (1999) 13





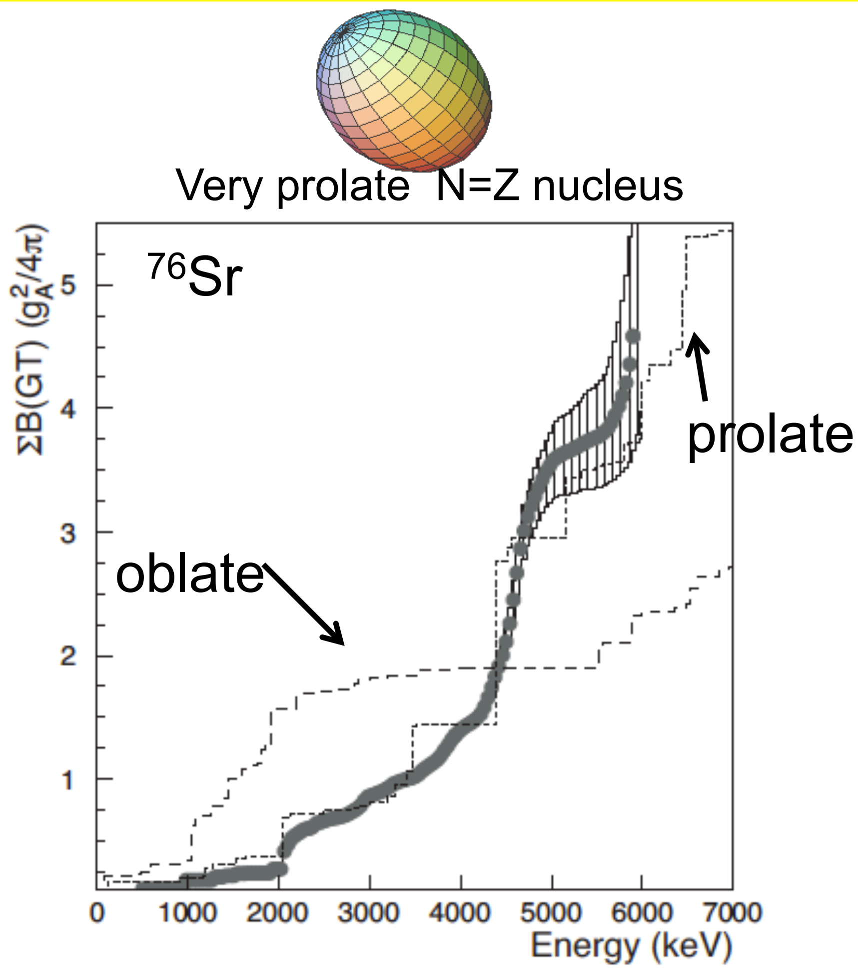
Lucrecia: the TAS at ISOLDE (CERN)
(Madrid-Strasbourg-Surrey-Valencia)

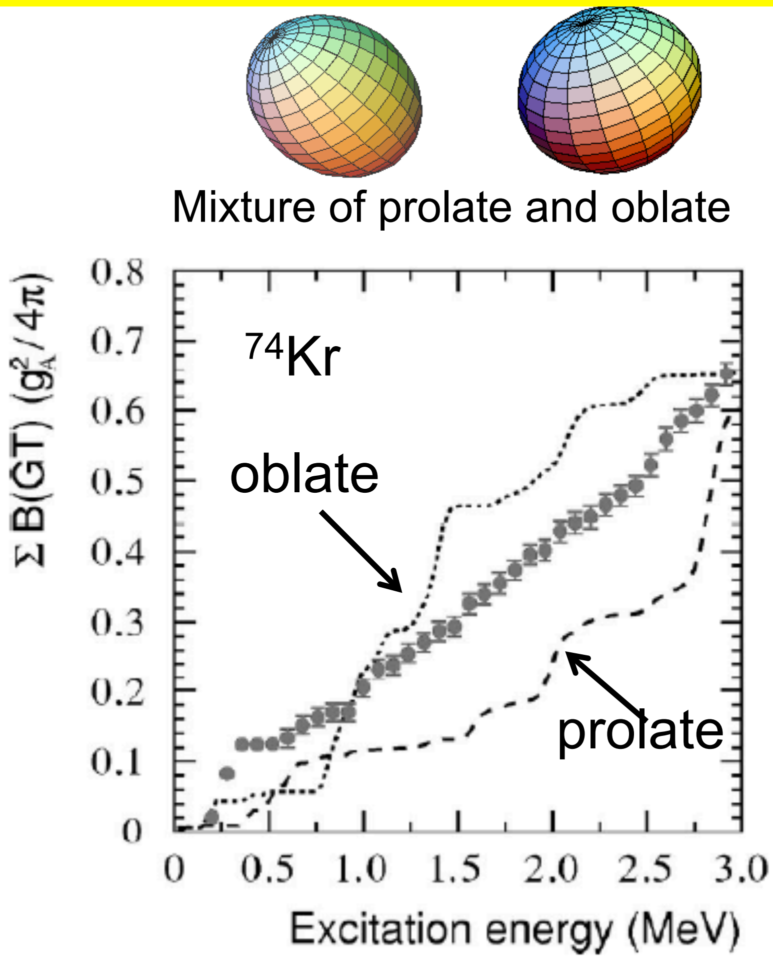




- A large NaI cylindrical crystal 38 cm Ø, 38cm length
- An X-ray detector (Ge)
- A β detector
- Possibility of collection point inside the crystal

Some earlier examples
(proposals of B. Rubio, P. Dessagne, W. Gelletly, et al.)





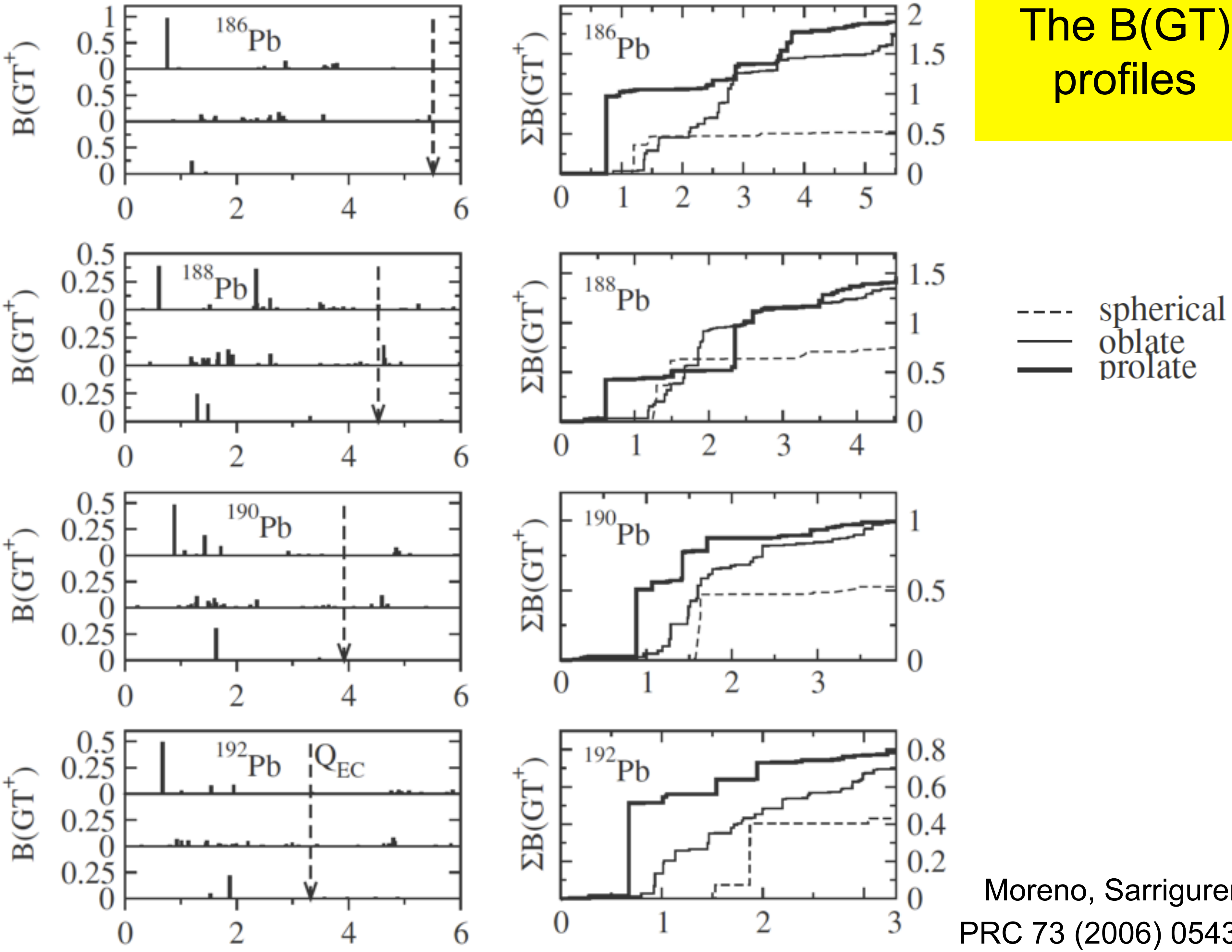
E. Nácher *et al. PRL 92 (2004) 232501 and PhD thesis Valencia*

Ground state of ^{76}Sr prolate (β_2 ~0.4) as indicated in Lister et al., PRC 42 (1990) R1191

E. Poirier *et al., Phys. Rev. C 69, 034307 (2004) and PhD thesis Strasbourg*

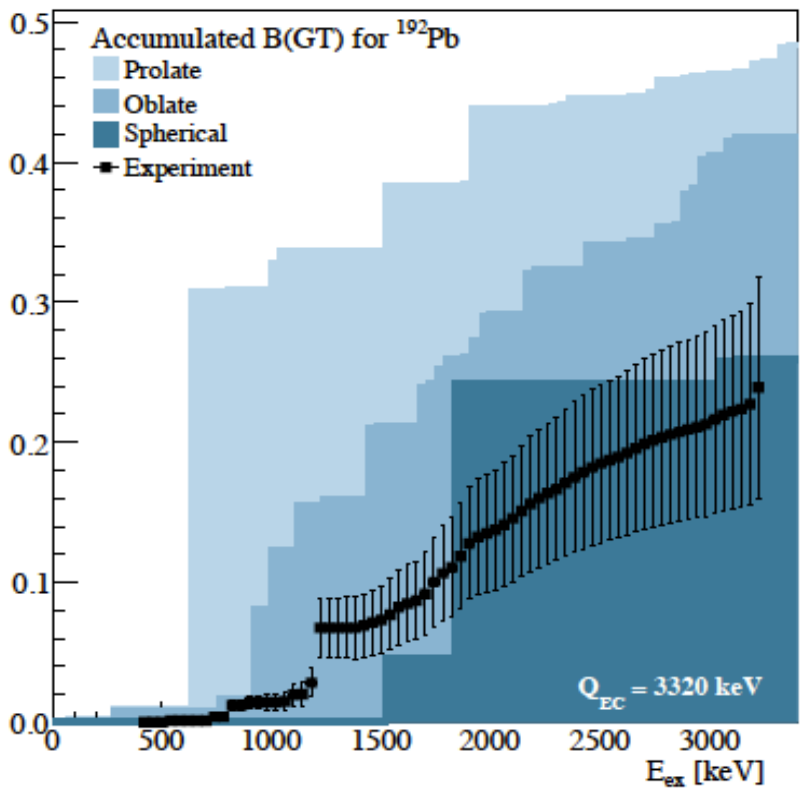
Ground state of ^{74}Kr:(60±8)% oblate, in agreement with other exp results and with theoretical calculations (A. Petrovici et al.)

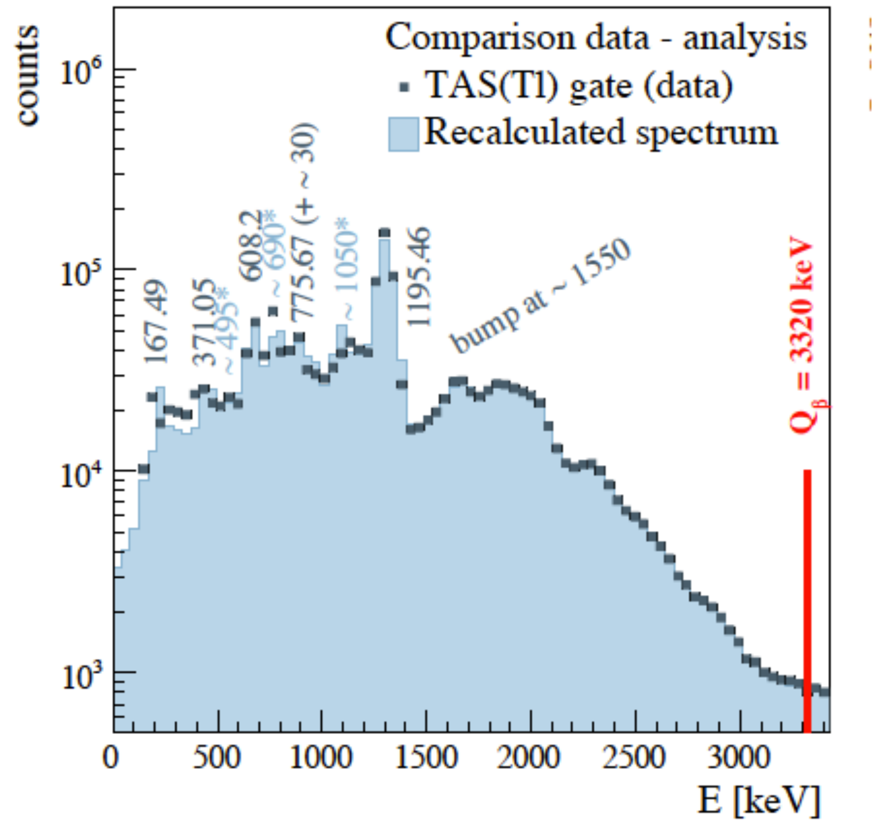
The B(GT) profiles



Moreno, Sarriguren
PRC 73 (2006) 054317

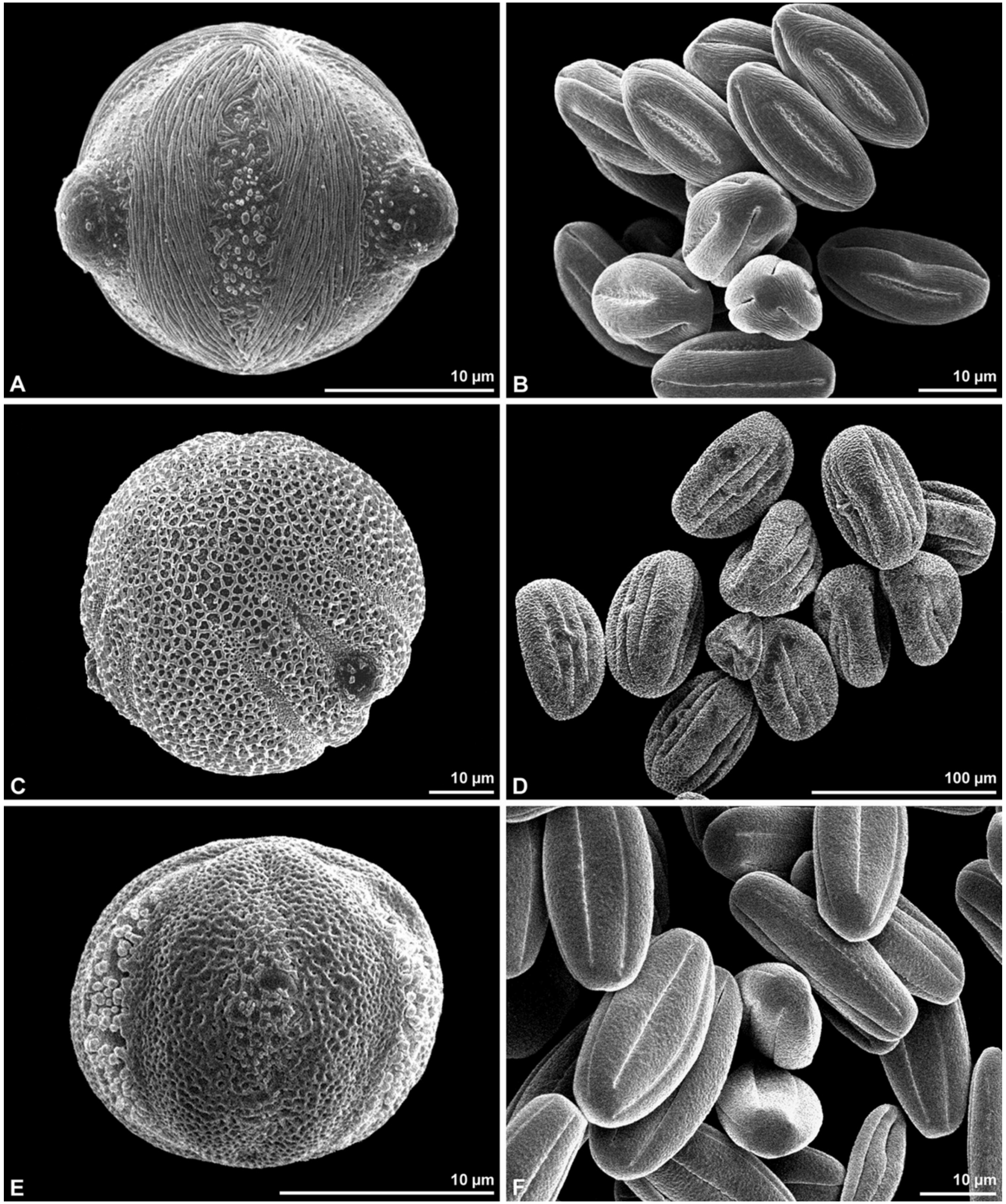
IS440 results: ^{192}Pb example





Thesis work of M. E. Estevez 2012, and M. E. Estevez *et al.* in preparation. Theory from PRC 73 (2006) 054317

Results consistent with spherical picture, but less impressive than in the A≈80 region. Similar situation for ^{190}Pb. *Possible explanation, the spherical character of the Pb nuclei, but requires further testing.*



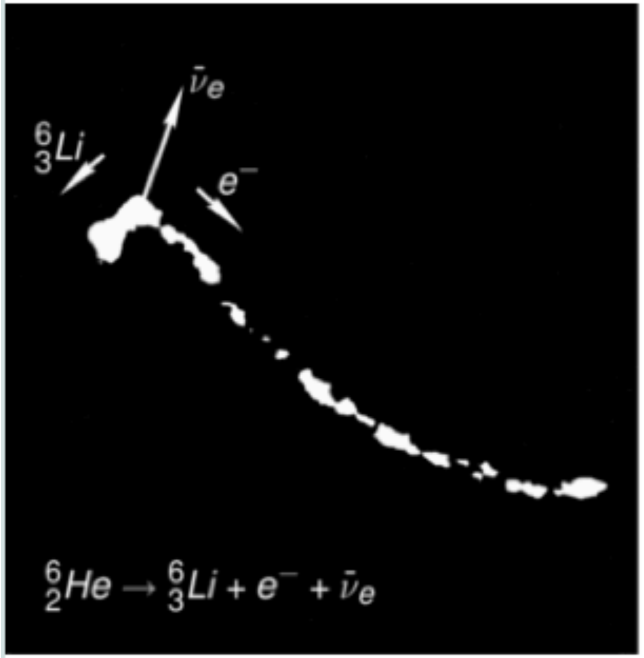
E. Estevez, J .L. Tain, B. Rubio, E.Nácher, J. Agramunt, A. B. Perez, L. Caballero, F. Molina, D. Jordan, A. Krasznahorkay, M. Hunyadi, Zs. Dombrádi, W. Gelletly, P. Sarriguren, O. Moreno, M. J. G. Borge, O. Tengblad, A. Jungclaus, L. M. Fraile, D. Fedosseev, B. A. Marsh, D. Fedorov, A. Frank, A. Algora

Conclusions

- I hope I have shown you that total absorption measurements can contribute to a better assessment of the decay heat in nuclear reactors.
- We are running a research program related to this topic, that can also have an impact in nuclear structure and astrophysics (not discussed here) and in neutrino physics applications
- The technique can be used for testing nuclear models, which can also be of relevance for neutrino physics applications

THANK YOU

Institute of Nuclear Research (MTA ATOMKI), Debrecen, Hungary



European Physical Society – EPS Historic Site

The neutrino experiment at MTA Atomki

USING A CLOUD CHAMBER LOCATED IN THIS BUILDING, IN 1956 J. CSIKAI AND A. SZALAY PHOTOGRAPHED BETA-DECAY EVENTS. IN SOME CASES THE ANGLE BETWEEN THE TRACKS OF THE ELECTRON AND THE RESIDUAL NUCLEUS IMPLIED THE EMERGENCE OF AN UNDETECTED THIRD PARTICLE IN THE DECAY. THUS CONFIRMING THE EXISTENCE OF THE NEUTRINO, THE DEBRECEN NEUTRINO EXPERIMENT LAID A BRICK OF THE FOUNDATION OF MODERN PHYSICS.

$$^{6}He \rightarrow {}^{6}Li + e^{-} + \overline{\nu}$$