β-delayed neutron emission: Measurement of emission probabilities

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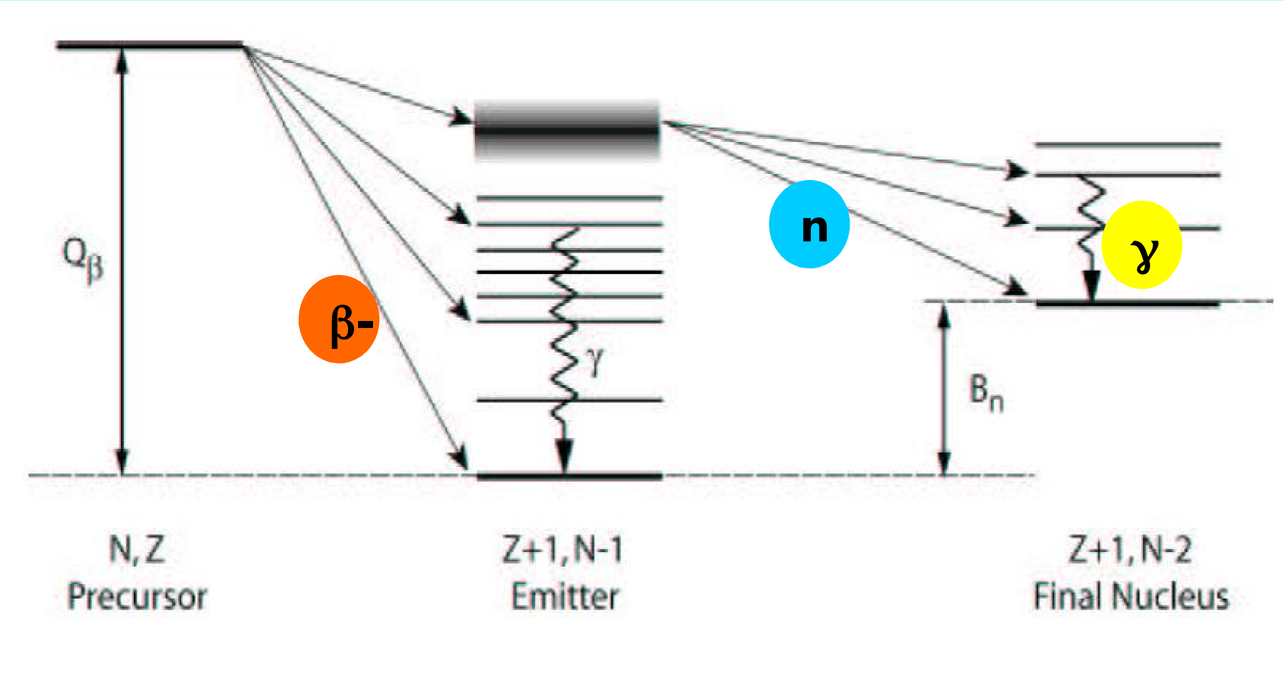
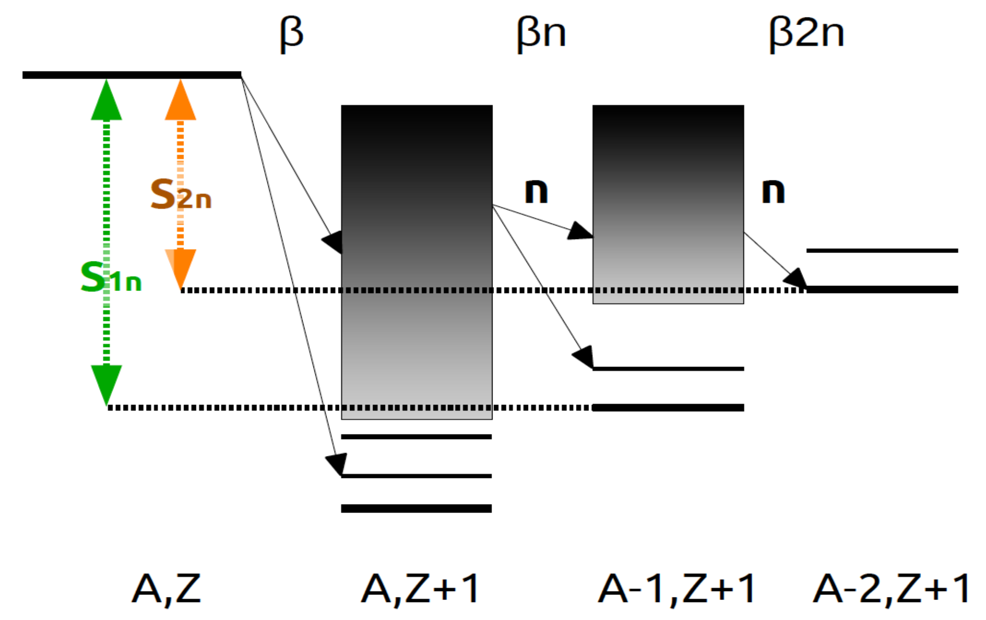
Grupo de Espectroscopia
Gamma y Neutrones

Beta-delayed neutron emission is an exotic process that occurs in neutron rich nuclei whenever the neutron separation energy in the daughter S_n is smaller than the available decay energy window Q_β



Beta-delayed multiple neutron emission can also occur whenever S_{2n}, S_{3n}, ... are smaller than Q_β



P_{xn}: x-neutron emission probability

$$P_{1n} = \frac{\int_{S_n}^{Q_\beta} \frac{\Gamma_{1n}(E_x)}{\Gamma_{tot}(E_x)} S_\beta(E_x) \cdot f(Q_\beta - E_x) \cdot dE_x}{\int_0^{Q_\beta} S_\beta(E_x) \cdot f(Q_\beta - E_x)\, dE_x}$$

$$\Gamma_{tot} = \Gamma_\gamma + \Gamma_{1n} + \Gamma_{2n} + \ldots$$

Total neutron emission probability

$$P_n = \sum_{x>0} P_{xn}$$

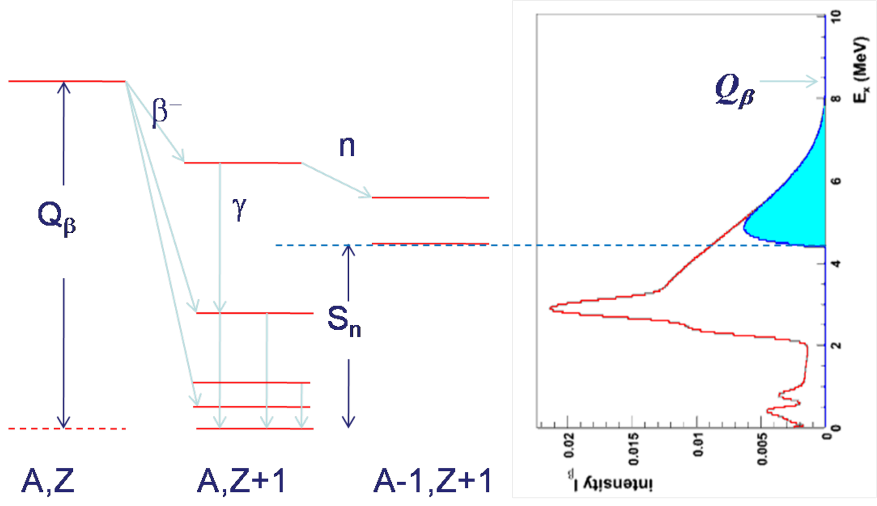
Neutron emission multiplicity

$$\langle n \rangle = \sum_{x>0} x P_{xn}$$

Beta strength:

$$S_\beta(E_x) = \frac{1}{D} \frac{g_A^2}{g_V^2} \frac{1}{2J_i + 1} \left| \langle f \| M_{\lambda\pi}^\beta \| i \rangle \right|^2$$

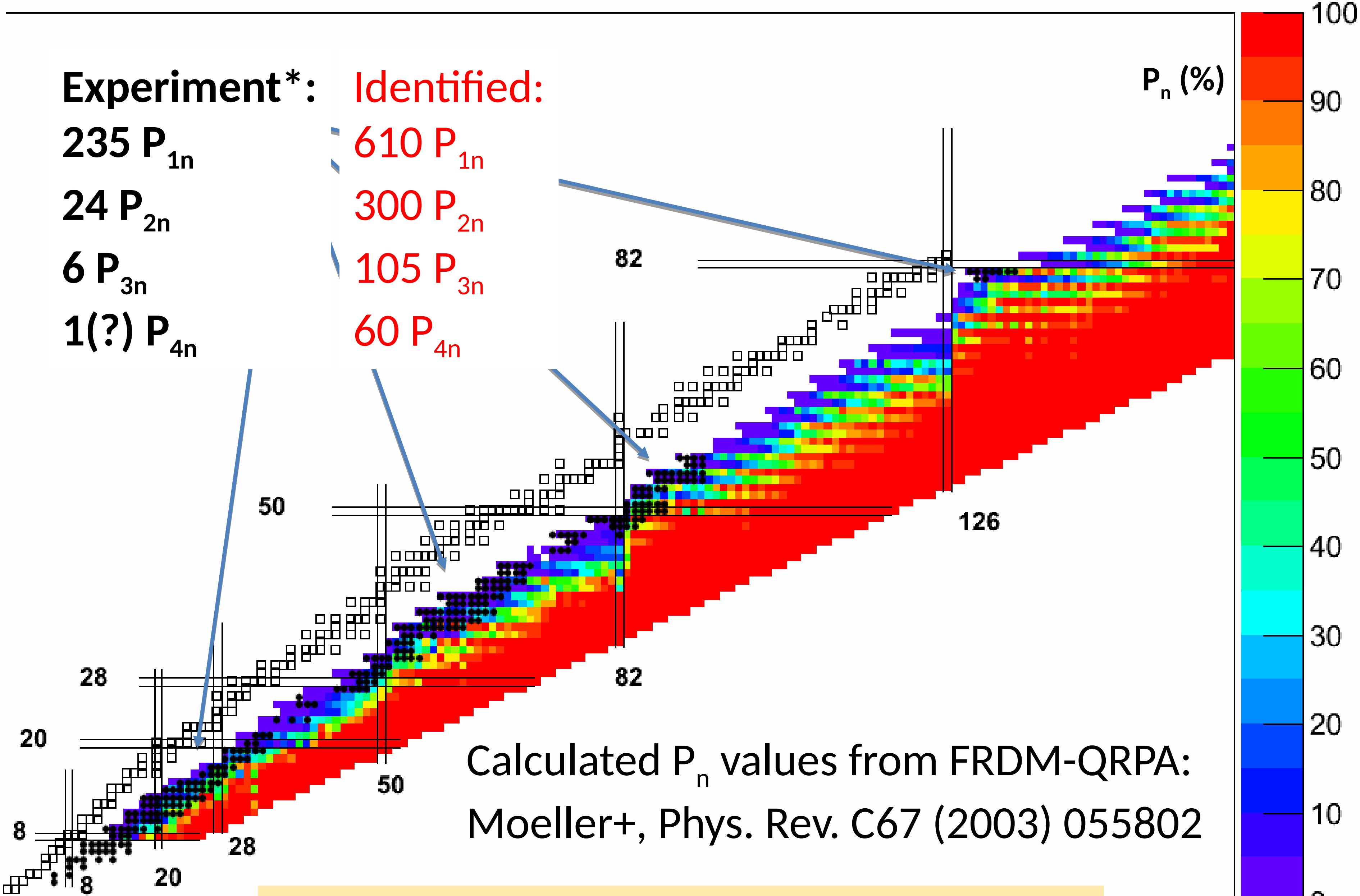
$$= \frac{I_\beta(E_x)}{T_{1/2} f(Q_\beta - E_x)}$$



Measured P_n values versus expected

Experiment*:
235 P_{1n}
24 P_{2n}
6 P_{3n}
1(?) P_{4n}

Identified:
610 P_{1n}
300 P_{2n}
105 P_{3n}
60 P_{4n}



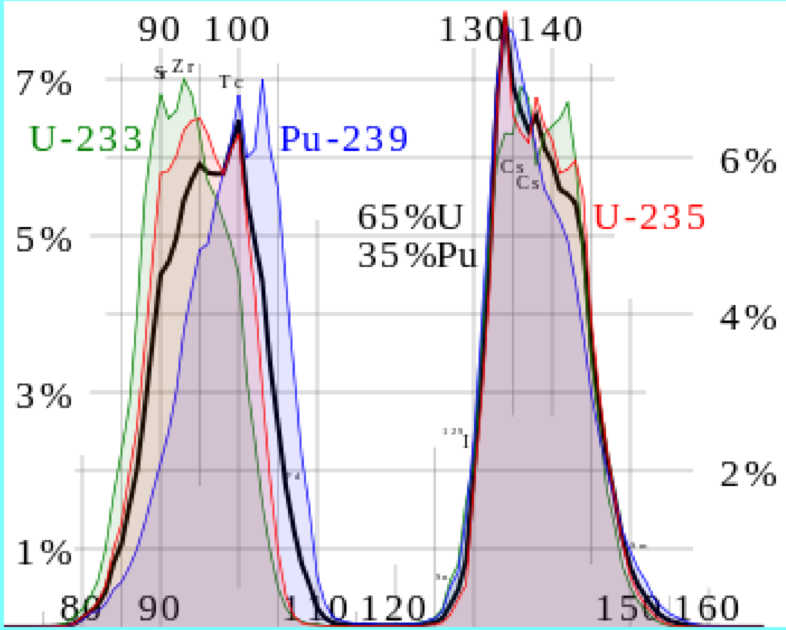
Calculated P_n values from FRDM-QRPA:
Moeller+, Phys. Rev. C67 (2003) 055802

(*) https://www-nds.iaea.org/beta-delayed-neutron/

Nuclear power reactors: delayed neutron fraction

- Some fission products are βn emitters
- They contribute with a small fraction ($\beta<1\%$) to the total number of neutrons in a reactor
- They are however essential for the mechanical control of reactor power

Fission yields as a function of A



Prompt neutrons vs. delayed neutrons

Isotope	fission cross-section 0.025eV / 2MeV	prompt neutrons 0.025eV / 2MeV	delayed neutrons 0.025eV / 2MeV
235U	585 / 1.27	2.42 / 2.63	0.0162 / 0.0165
238U	0.000027 / 0.57	2.36 / 2.60	0.0478 / 0.0478
233U	531 / 1.98	2.48 / 2.63	0.0067 / 0.0077
239Pu	747 / 1.93	2.87 / 3.16	0.0065 / 0.0067
241Pu	1 012 / 1.76	2.92 / 3.21	0.0160 / 0.0160

Thermal energies

- The time evolution of delayed neutron fraction is represented by six (eight) “groups of isotopes”
- The group parameters are fissile nucleus dependent and neutron energy dependent
- They are obtained from integral measurements

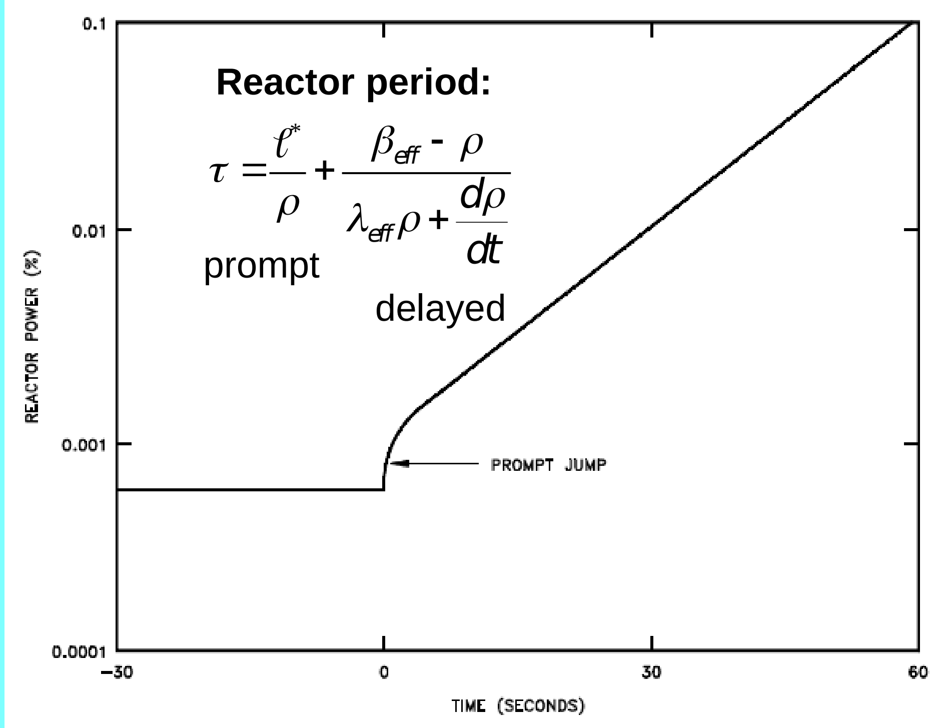
i	Possible precursor nuclei	Mean energy (MeV)	Average half-life of the group [s]			Delayed neutron fraction [%]		
			235U	239Pu	233U	235U	239Pu	233U
1	87Br, 142Cs	0.25	55.72	54.28	55.0	0.021	0.0072	0.0226
2	137I, 88Br	0.56	22.72	23.4	20.57	0.140	0.0626	0.0786
3	138I, 89Br, (93,94)Rb	0.43	6.22	5.60	5.00	0.126	0.0444	0.0658
4	139I, (93,94)Kr 143Xe, (90,92)Br	0.62	2.3	2.13	2.13	0.252	0.0685	0.0730
5	140I, 145Cs	0.42	0.61	0.618	0.615	0.074	0.018	0.0135
6	(Br, Rb, As etc.)	-	0.23	0.257	0.277	0.027	0.0093	0.0087
		Total				0.64	0.21	0.26

Thermal energies

- The time evolution of reactor power after sudden variations of the reactivity ρ is modulated by the delayed neutron fraction β

$$\rho = \frac{k_{eff} - 1}{k_{eff}}$$

k_{eff}: effective multiplication factor



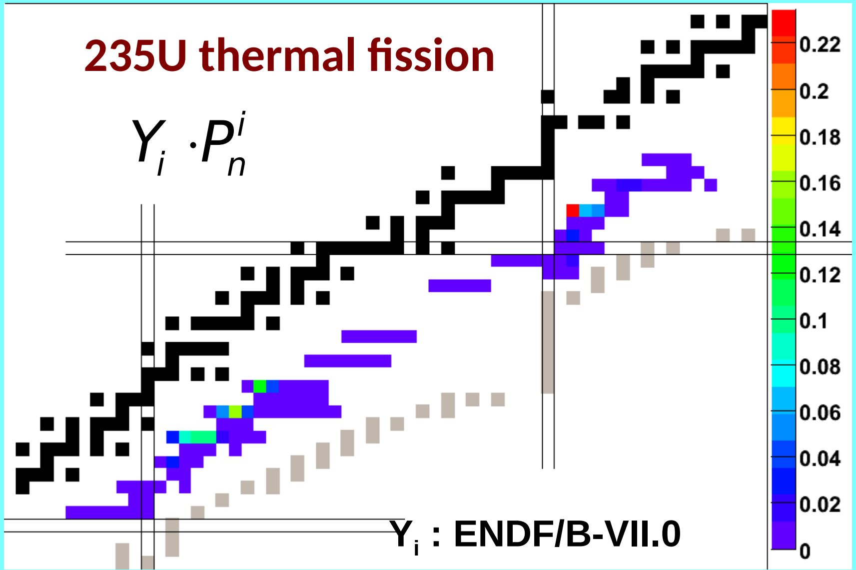
Microscopic summation calculations of $\bar{\nu}_d$

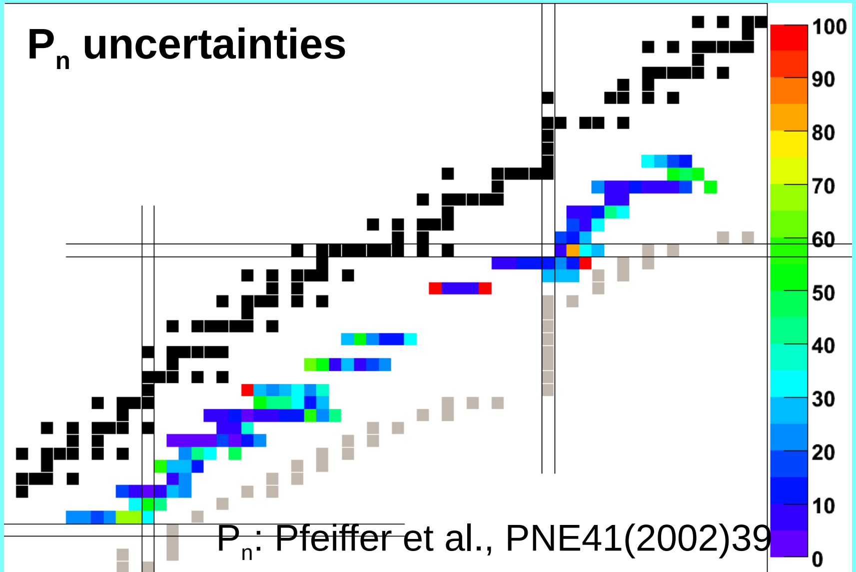
- A more fundamental and generic approach to the estimation of β_{eff}
- Microscopic summation calculations lack still the accuracy of Keepin six-group formula
- Reason: **inaccuracies** in fission yields Y and **delayed neutron emission probabilities** $\boldsymbol{P_n}$
- Improvement of P_n values and comparison with integral measurements can constrain Y

Number of delayed neutrons per fission

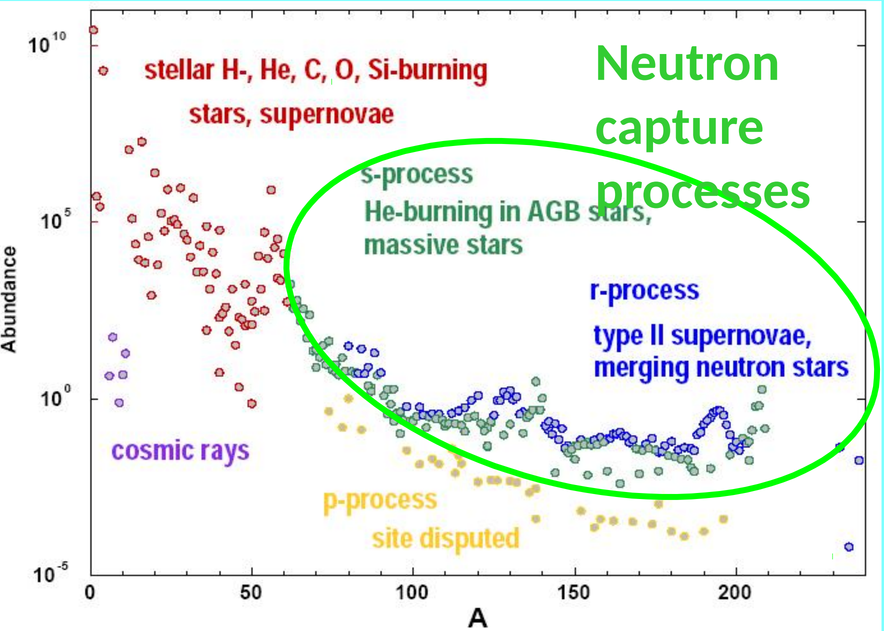
$$\bar{\nu}_d = \sum_i Y_i \cdot P_n^i$$

Can be used to identify P_n values that should be revisited

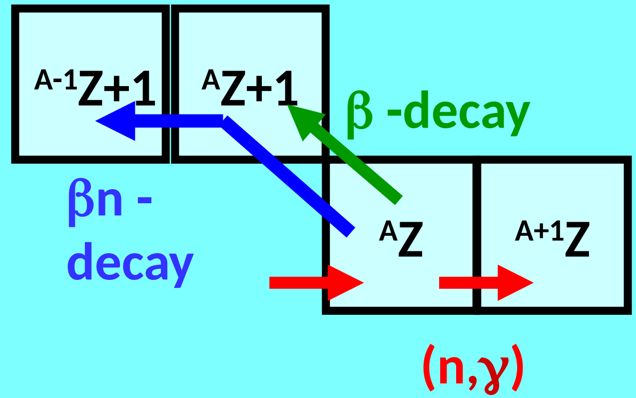


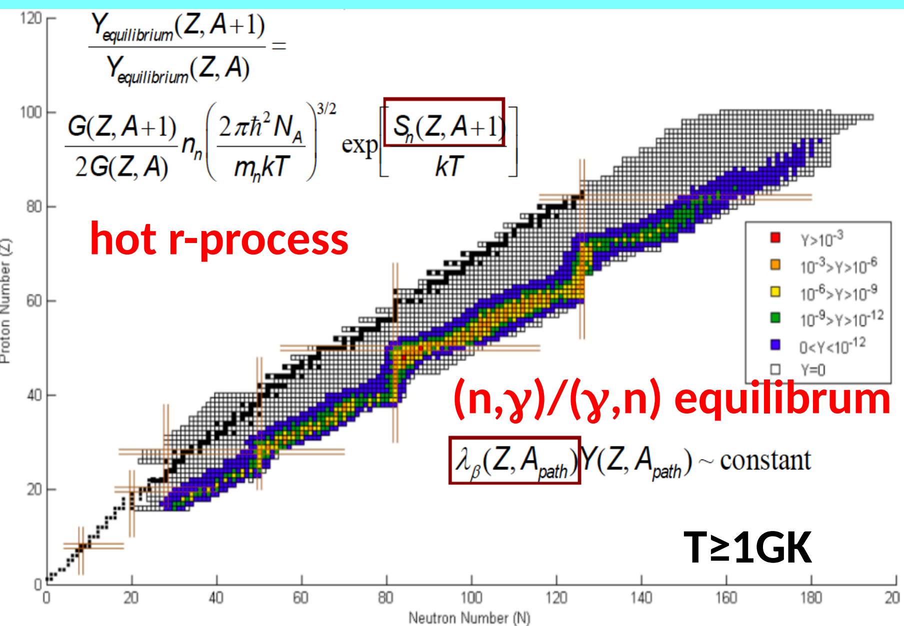


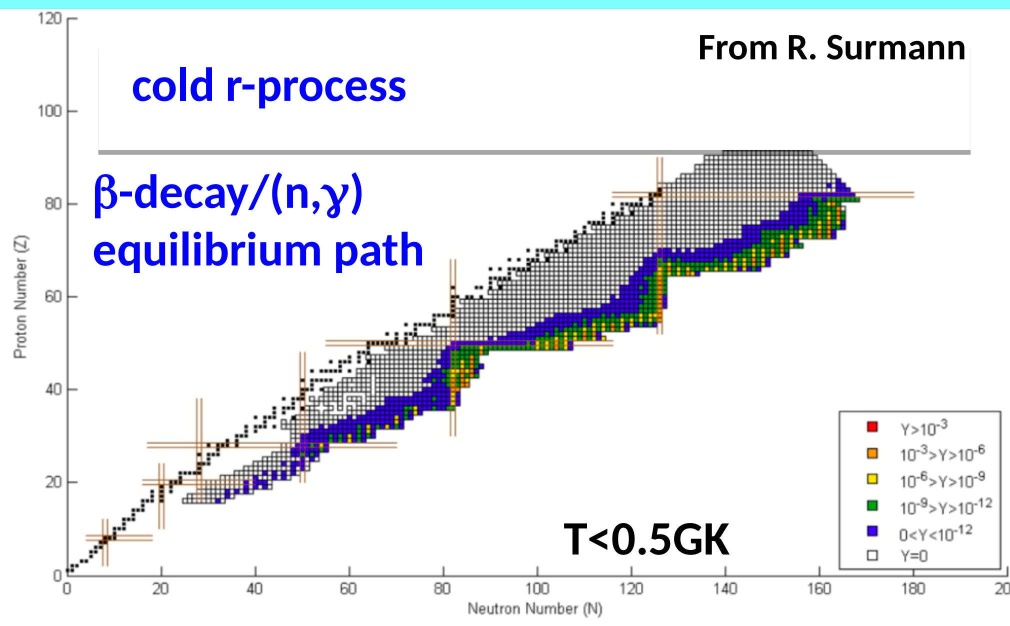
Astrophysics: The r-process



R-process: A short and very high neutron flux ($n_n > 10^{20}$ g/cm³) produces very neutron-rich nuclei by successive neutron captures in a short time, which then decay to stability.

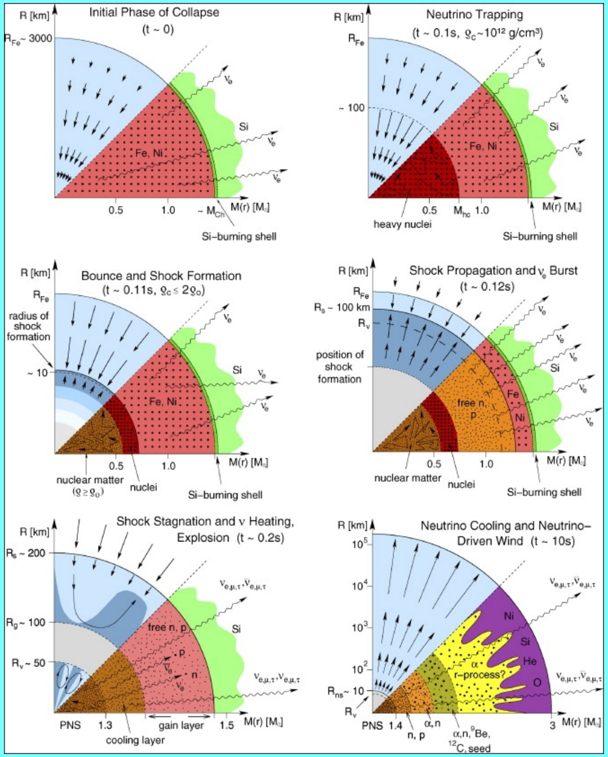






The r-process astrophysical site: Core Collapse Supernova Event

Complex explosion mechanism

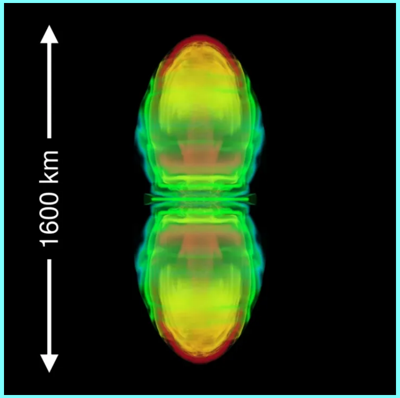


3D simulation

Burrows, Nature 589 (2021) 29

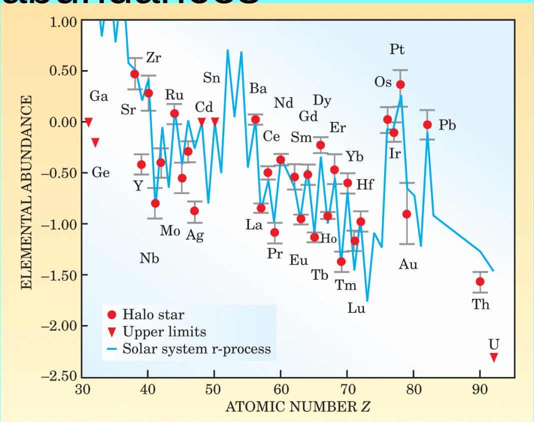
Conditions for synthesis of heaviest elements are not found in simulations

Are very large magnetic fields and rotations needed?



NS or BH remnant

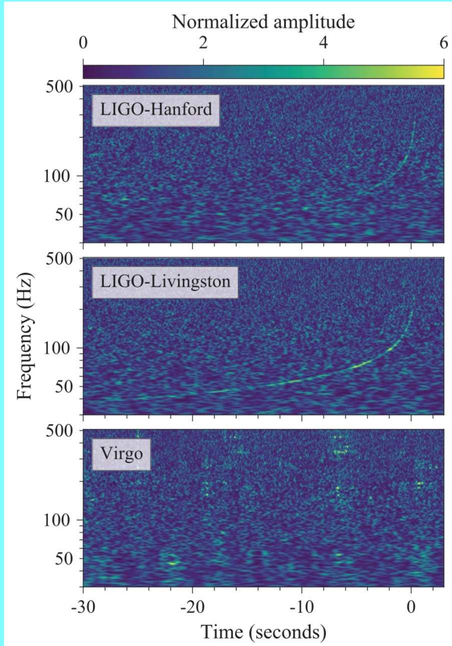
Extremely old stars show solar r-process abundances

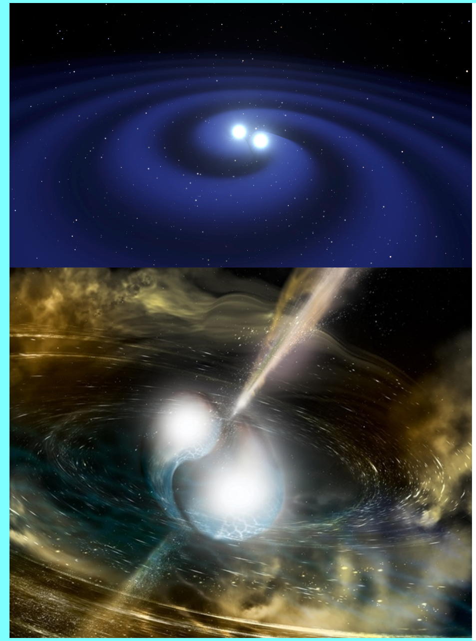


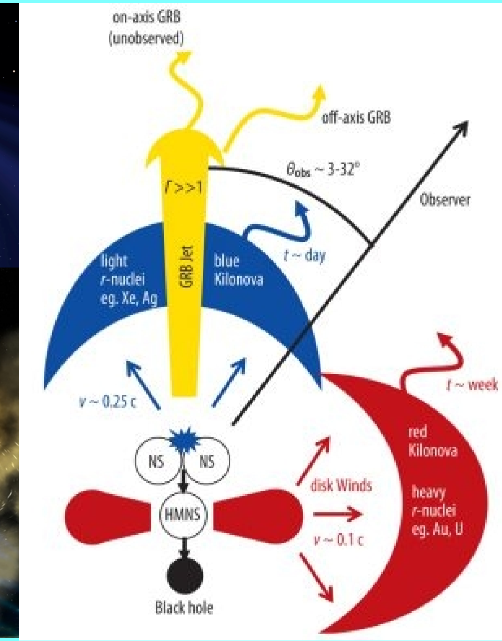
The r-process astrophysical site: Neutron Star Merger

GW170817: First detection of GW from NSM

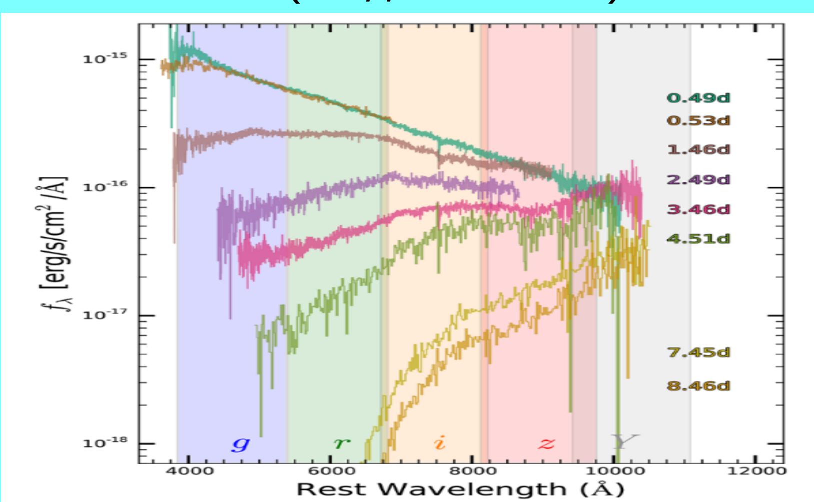
Abbott+, PhysRevLett 119 (2017) 161101

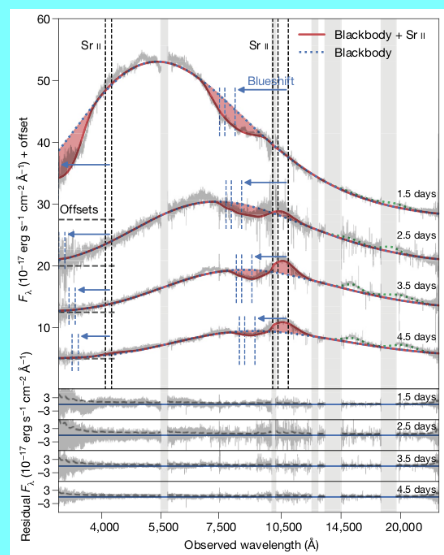






Followed by detection of kilonova EM emission (*Shappee+, Sience*)



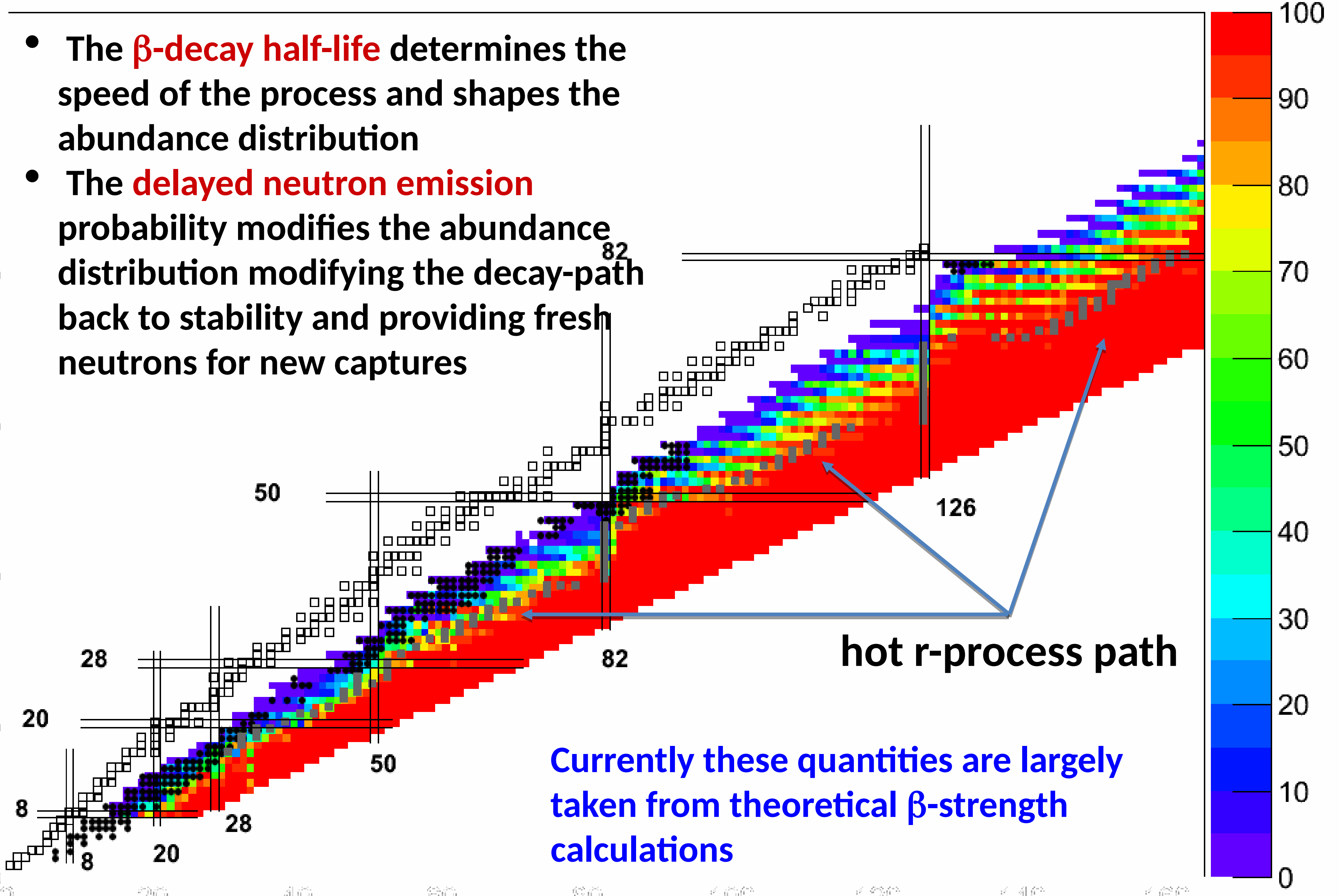


First indication of r-process nucleo-synthesis

Watson+, Nature 574 (2019) 497

Importance of $T_{1/2}$ and P_n values in r-process nucleosynthesis

- The β-decay half-life determines the speed of the process and shapes the abundance distribution
- The delayed neutron emission probability modifies the abundance distribution modifying the decay-path back to stability and providing fresh neutrons for new captures

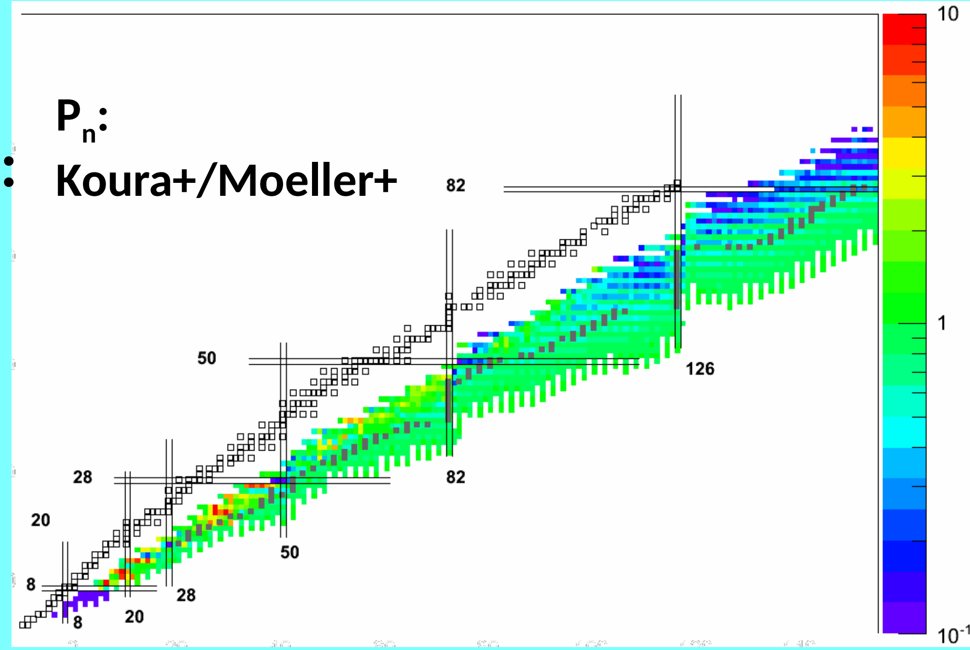


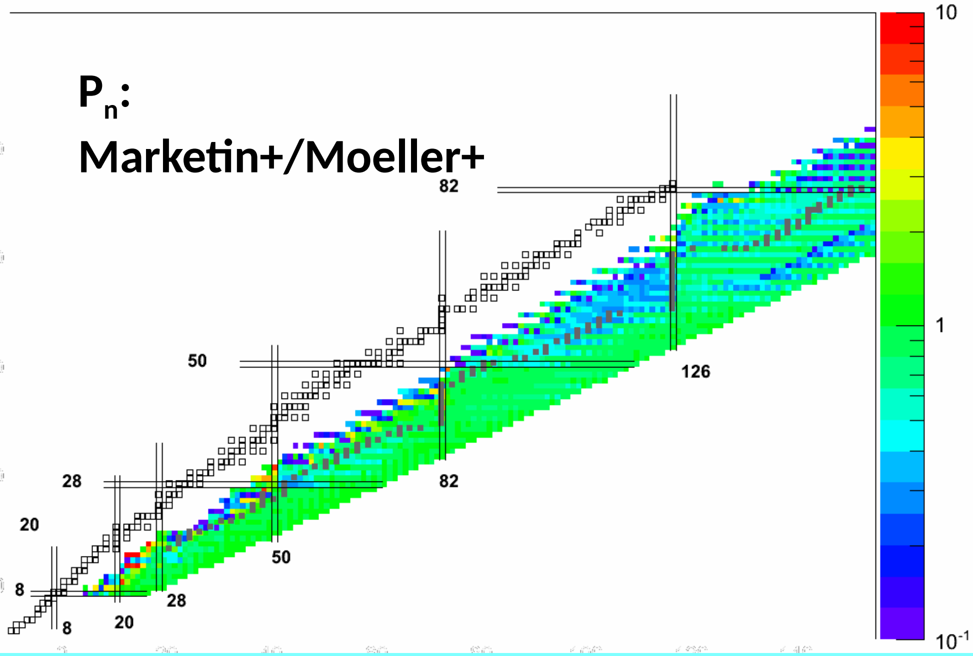
Currently these quantities are largely taken from theoretical β-strength calculations

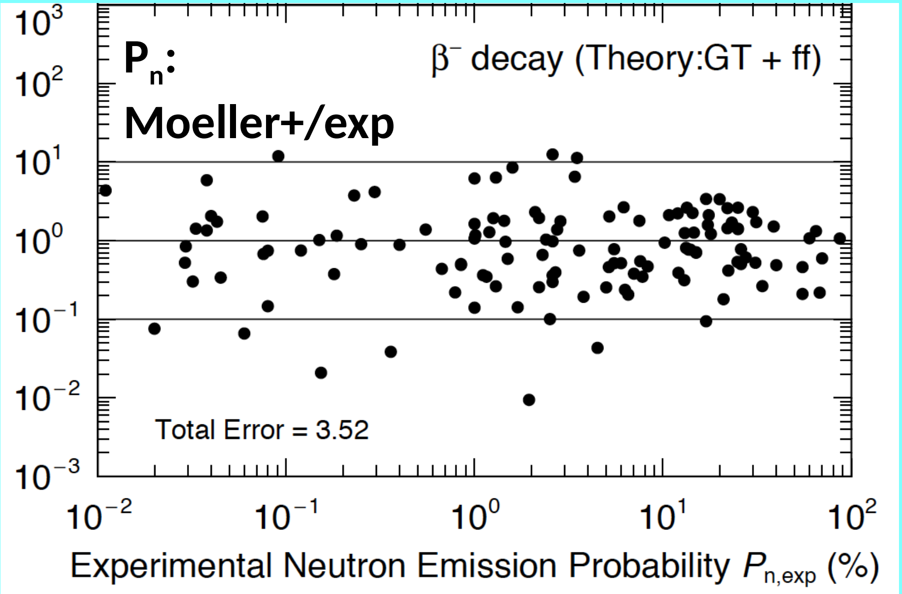
Comparison of global calculations: P_n

How reliable are the calculations?

- **Moeller+, PRC67 (2003) 055802: FRDM+QRPA**
- **Marketin+, PRC93 (2016) 025805: RHB+RQRPA**
- **Koura+, PTP113(2005)305 & PTP84(1990)641 : KTUY+GT2**

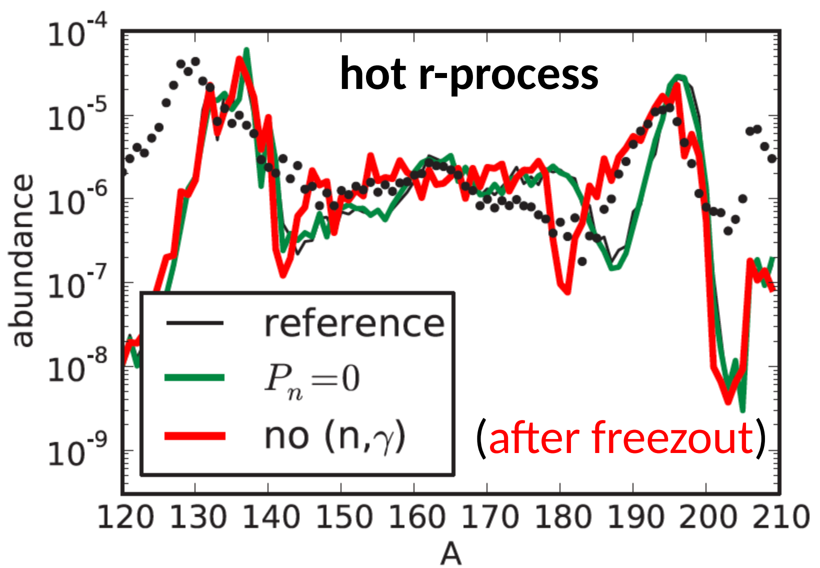


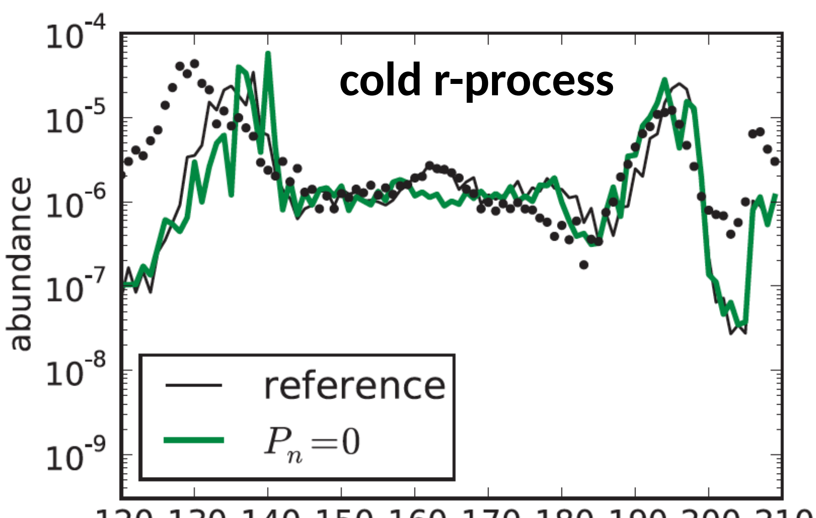




Impact of P_n

Arcones+, PRC83(2011)045809

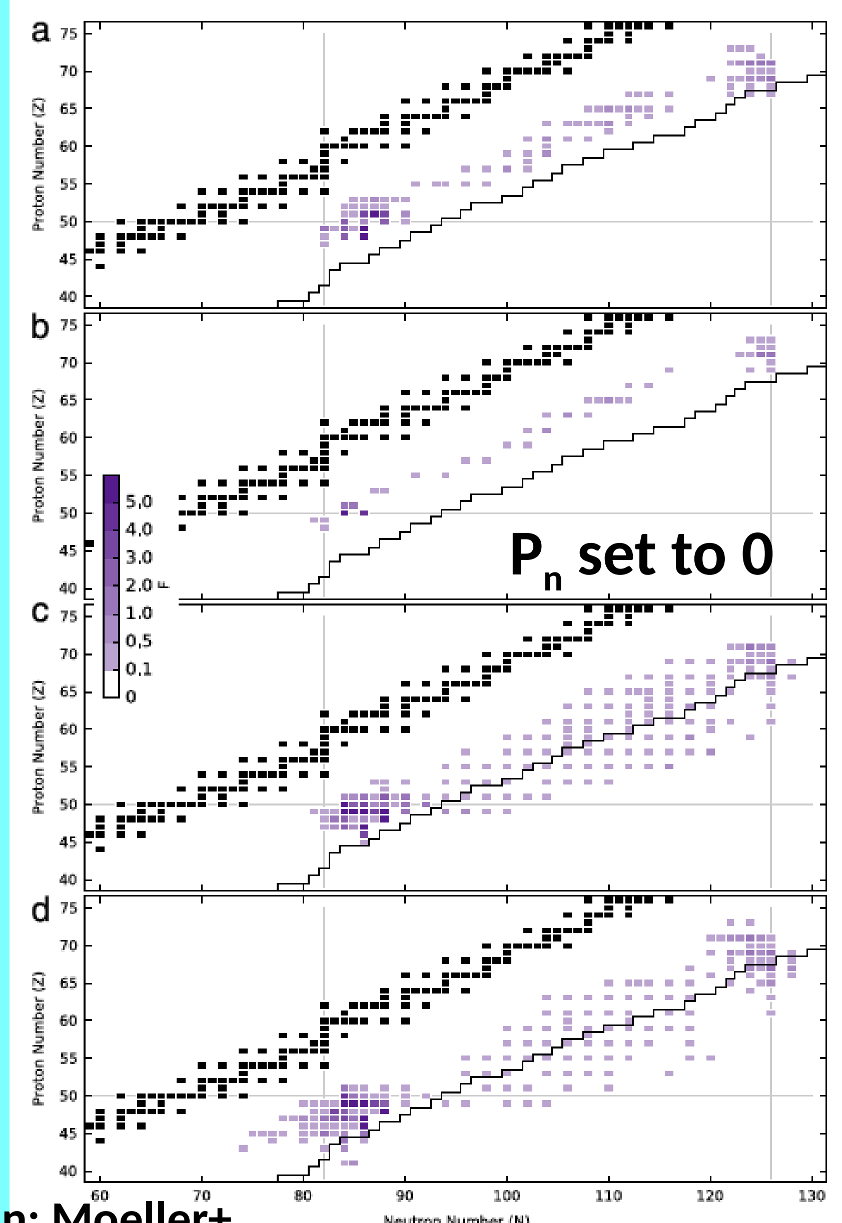




Sensitivity check:

$$F = 100 \sum_A |X(A) - X_b(A)|$$

Mumpower+, ProgPartNucPhys86 (2016) 86



low entropy hot wind

high entropy hot wind

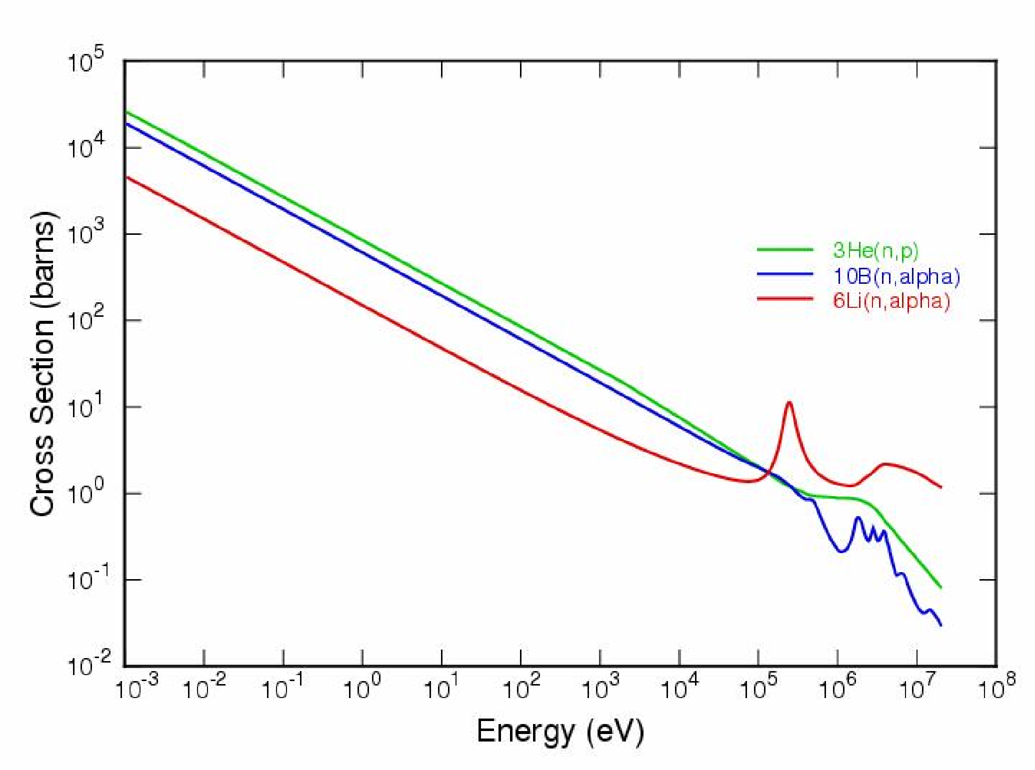
cold wind

neutron star merger

Base calculation: Moeller+

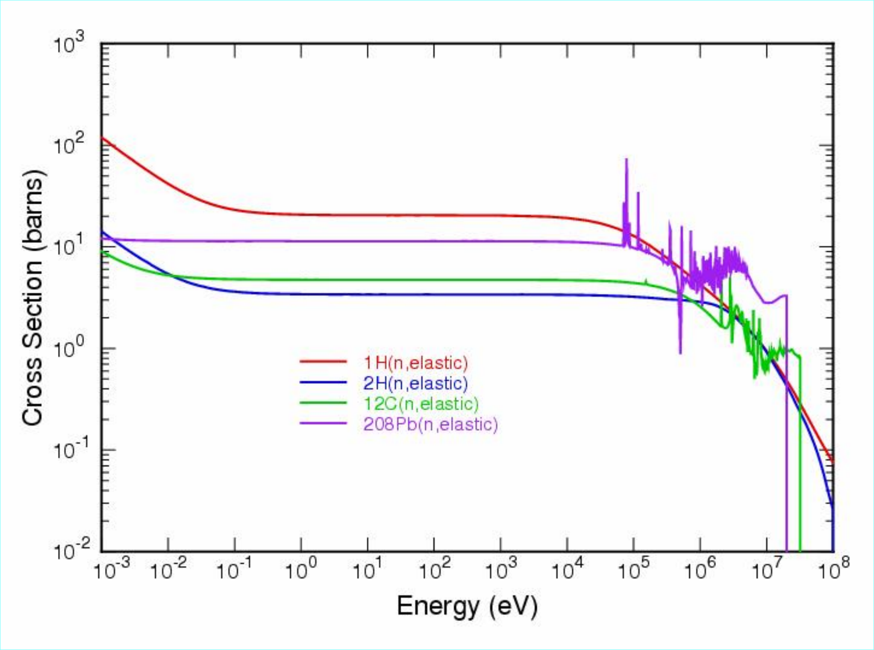
Measurement of P_n values

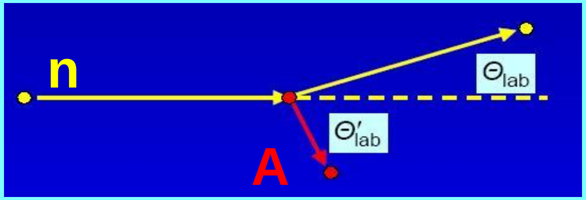
- The best method is by direct detection of the neutrons emitted
- Neutrons are neutral particles thus their detection require the production of electromagnetically interacting particles
- Reactions used: nucleus scattering, charged particle producing reactions, radiative capture, fission
- A useful reaction is $^3He(n,^3H)^1H$, with Q=+764keV which has a large cross-section at thermal energies



- ^{3}He is a (rare) gas that can be used as the sensitive gas of proportional counters

- Moderation of neutron energy by scattering on hydrogen is very useful to thermalize its energy





$$\left[\frac{E}{E_0}\right]_{max} = \frac{(A^2-1)^2}{(A+1)^2} = \alpha$$

Maximum energy loss: 1-α

Slowing down parameter:

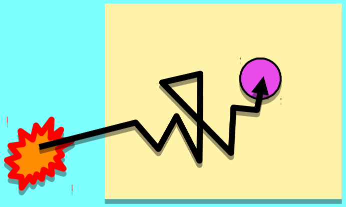
$$\xi = \left\langle \ln\frac{E_0}{E} \right\rangle = 1 + \frac{(A-1)^2}{2A} \ln\frac{A-1}{A+1}$$

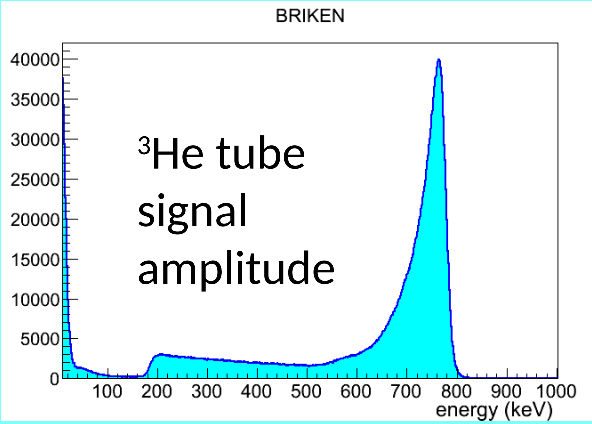
Nucleus	1-α	ξ	N
^{1}H	1	1	18
^{2}H	0.889	0.725	24
^{4}He	0.640	0.425	41
^{12}C	0.284	0.158	111
^{56}Fe	0.069	0.035	500
^{208}Pb	0.019	0.010	1823

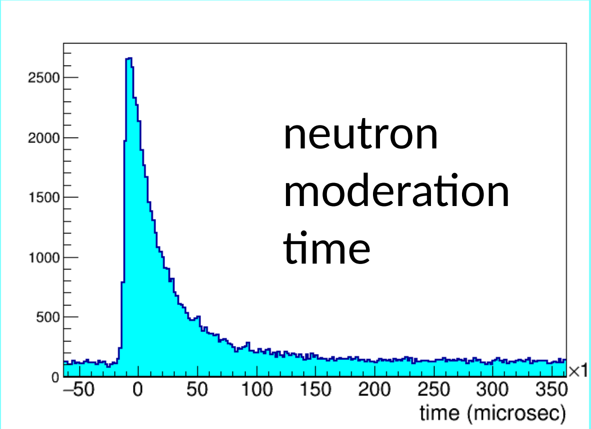
N: cumber of collisions to bring E_n from 1MeV to 25meV

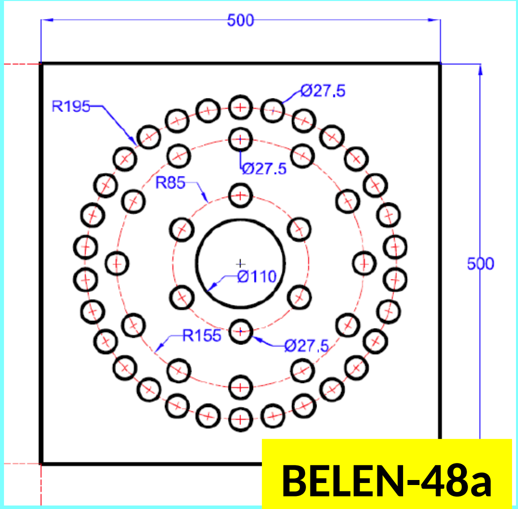
Moderated neutron neutron counter

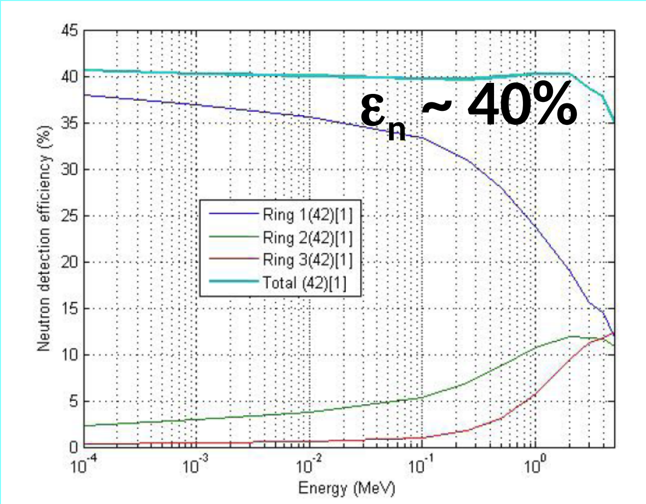
- Array of ^{3}He filled proportional tubes inside a neutron energy moderator polyethylene (PE) matrix

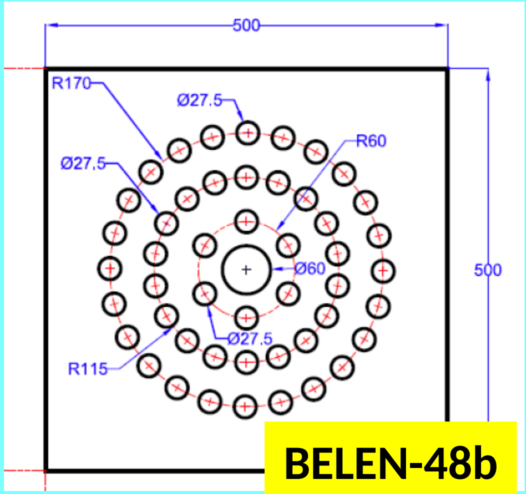


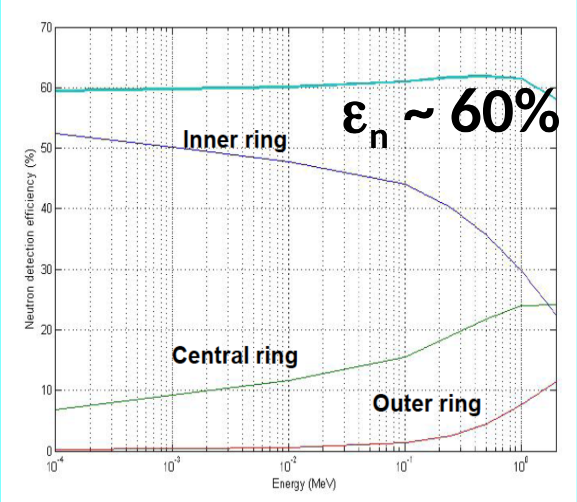










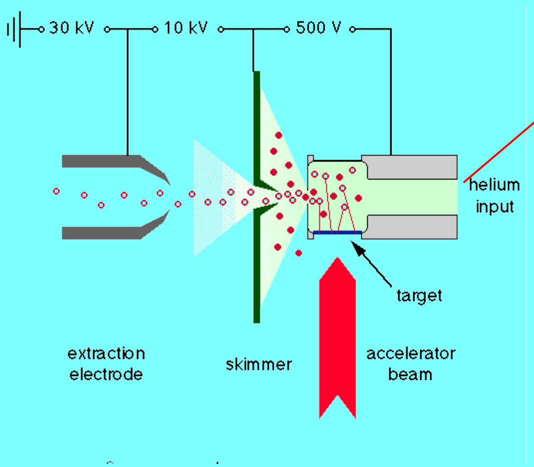


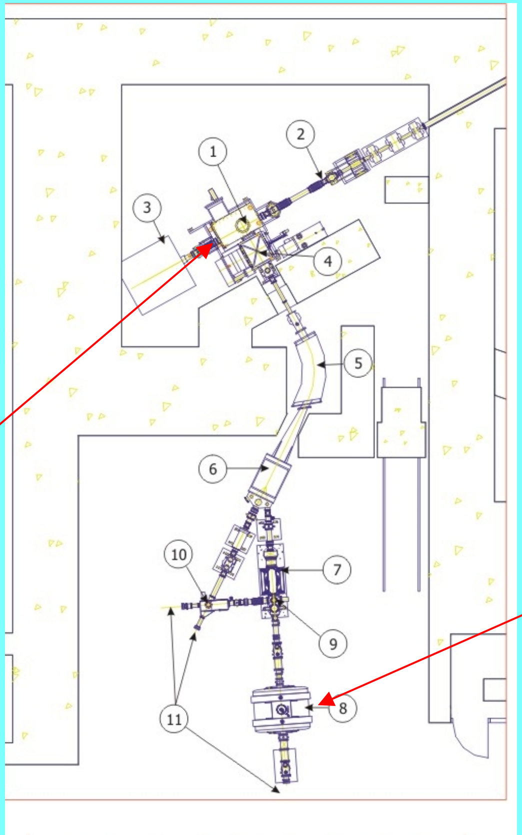
Experiment at ISOL facility: production and selection of isotopes

JYFL Accelerator Laboratory

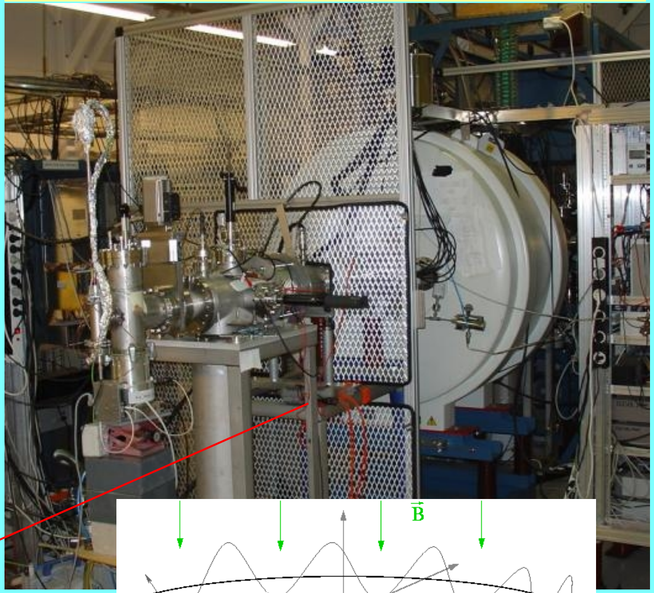
IGISOL separator **+ ion guide** source: refractory elements

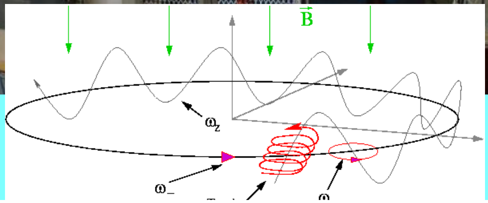
p(25MeV) + Th => FF

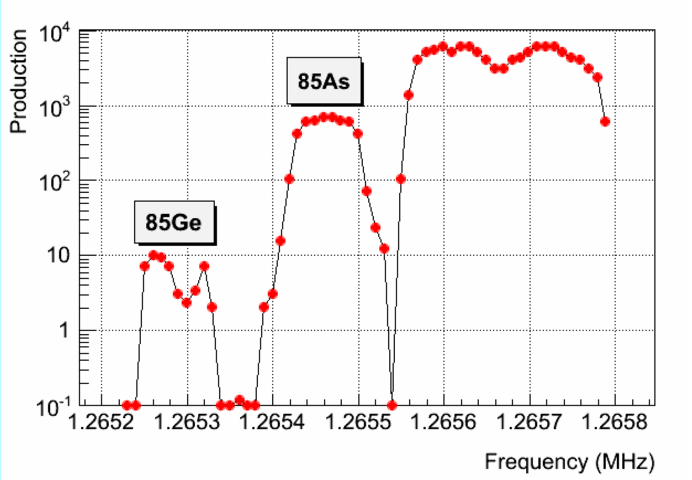




JYFLTRAP Penning trap: isotopic purification







Isotope	Rate (s^{-1})	Isotope	Rate (s^{-1})
^{88}Br	1450	^{85}Ge	6
^{94}Rb	1030	^{85}As	175
^{95}Rb	760	^{86}As	30
^{137}I	100	^{91}Br	80

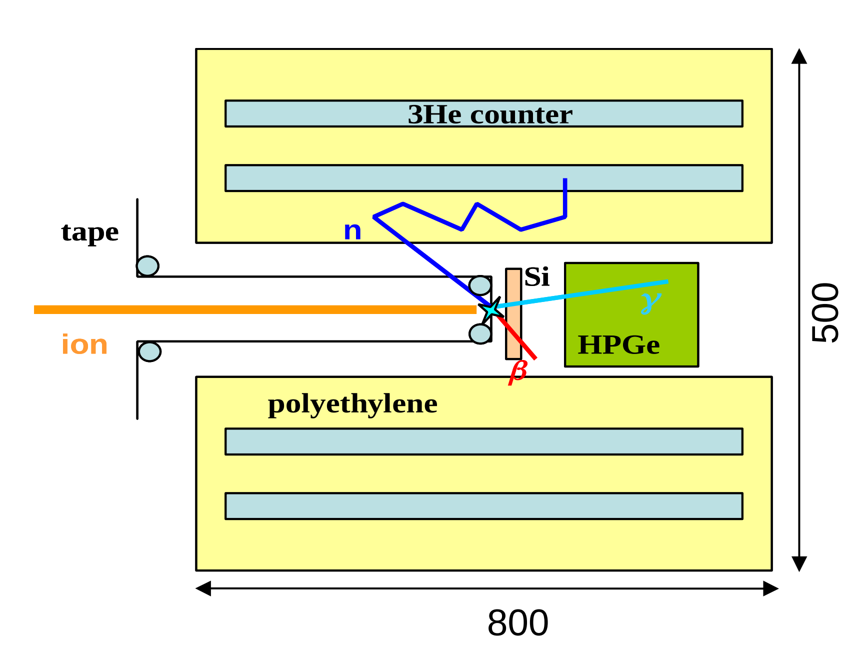
Pure isotopic beams

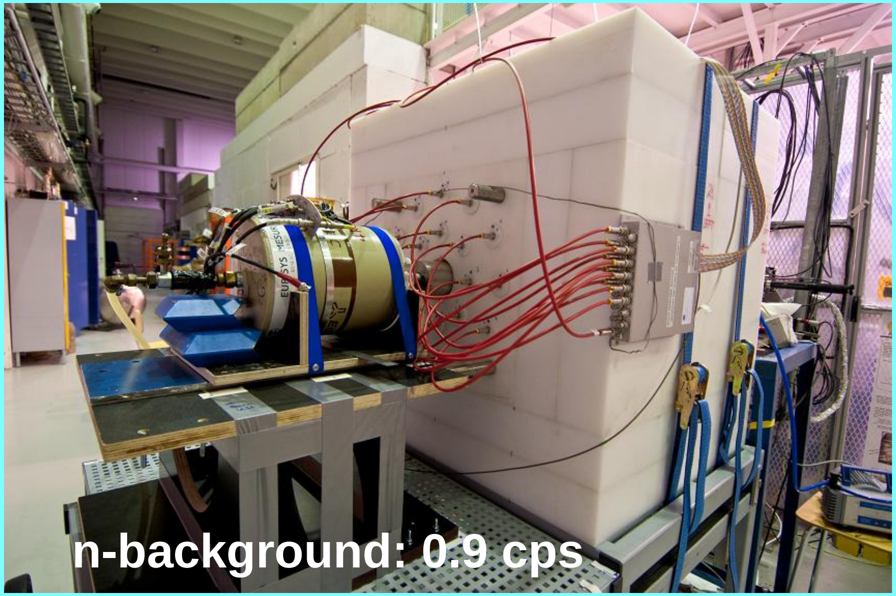
Experimental setup:

BELEN-20 detector

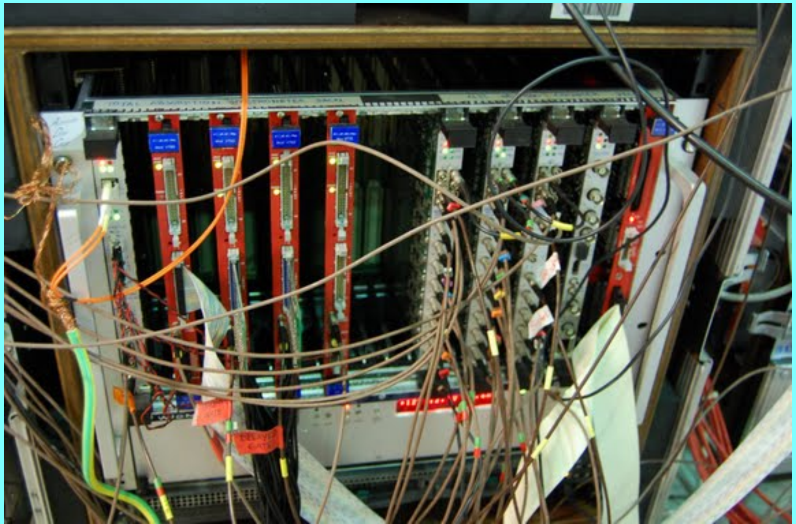
20 ⊘2.5cm×60cm 3He tubes @20atm

- 30keV beam implanted on tape
- Si or plastic detector for β detection
- HPGe detector for γ detection





Neutron background shield: 20cm PE



Self triggered DACQ:
-Time-energy pairs for every neutron or β
-Clean noise separation
-Minimum dead time:<0.5%

Data analysis

- To obtain P_n we need to count the total number of decays and the number of decays followed by n emission
- For this we count β and n or βn coincidences

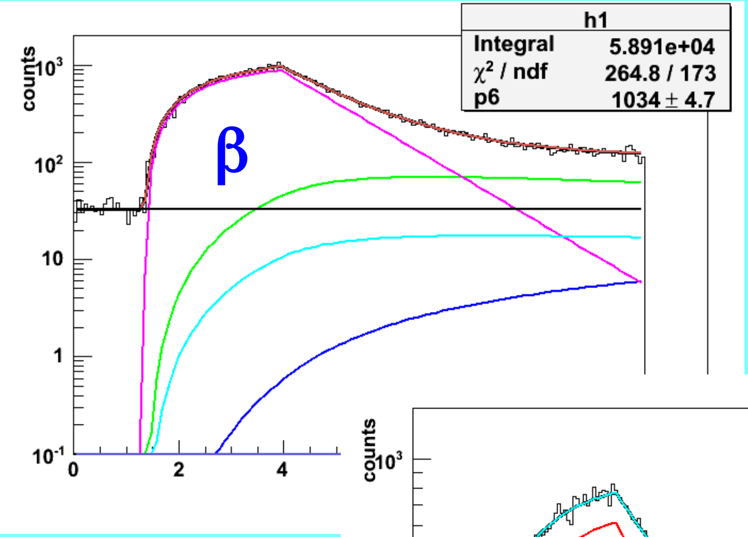
$$N_\beta = \bar{\varepsilon}_\beta N_{dec}$$
$$N_n = \bar{\varepsilon}_n P_n N_{dec}$$
$$N_{\beta n} = \bar{\varepsilon}'_\beta \bar{\varepsilon}_n P_n N_{dec}$$

$$P_n = \frac{\bar{\varepsilon}_\beta}{\bar{\varepsilon}_n} \frac{N_n}{N_\beta}$$

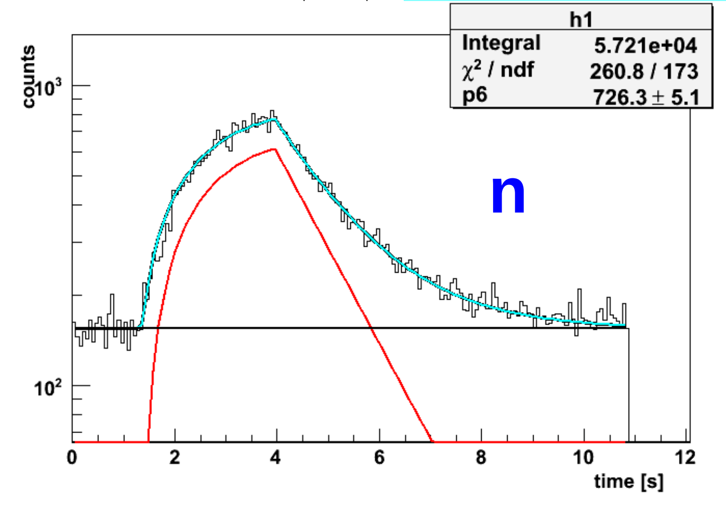
$$P_n = \frac{\bar{\varepsilon}_\beta}{\bar{\varepsilon}'_\beta \bar{\varepsilon}_n} \frac{N_{\beta n}}{N_\beta}$$

NOTE: average efficiencies over all transitions

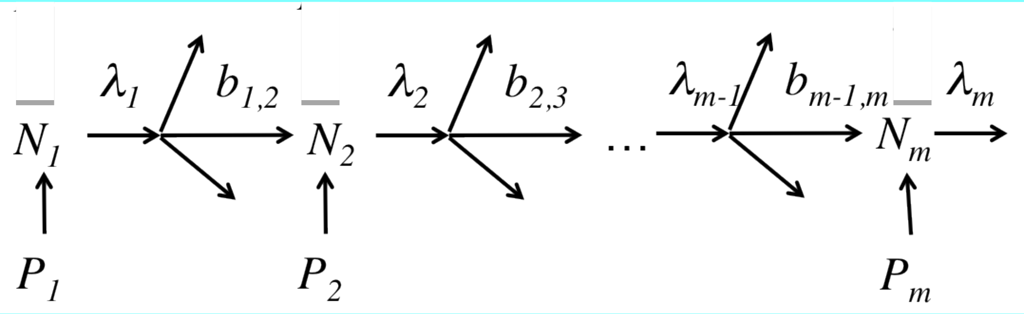
- We need to disentangle the counts from the nucleus of interest from other nuclei
- For this we measure grow and/or decay curves of the activity and fit with appropriate solutions of the Bateman equations



^{86}As



Solution of Bateman equations from Skrable et al., Health. Phys. 27 (1974) 155)



Equations:

$$\frac{dN_1}{dt} = P_1 - \lambda_1 N_1$$

$$\frac{dN_2}{dt} = P_2 + \lambda_1 b_{1,2} N_1 - \lambda_2 N_2$$

$$\ldots$$

$$\frac{dN_m}{dt} = P_m + \lambda_{m-1} b_{m-1,m} N_{m-1} - \lambda_m N_m$$

Measured activity:

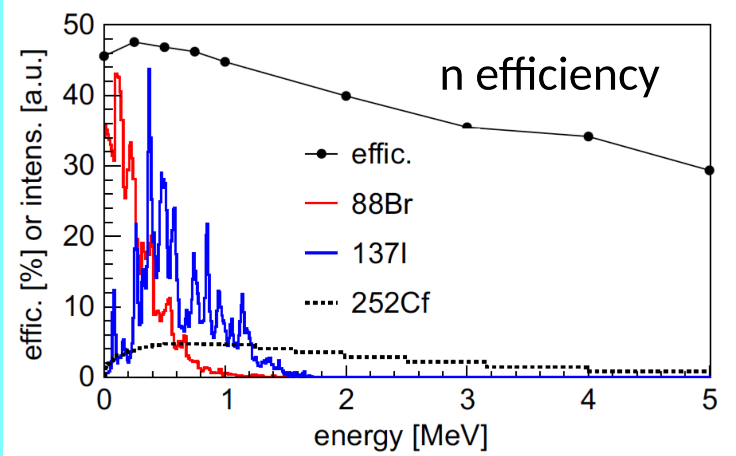
$$A_m^{\beta}(t) = \bar{\varepsilon}_{\beta} \lambda_m N_m(t)$$

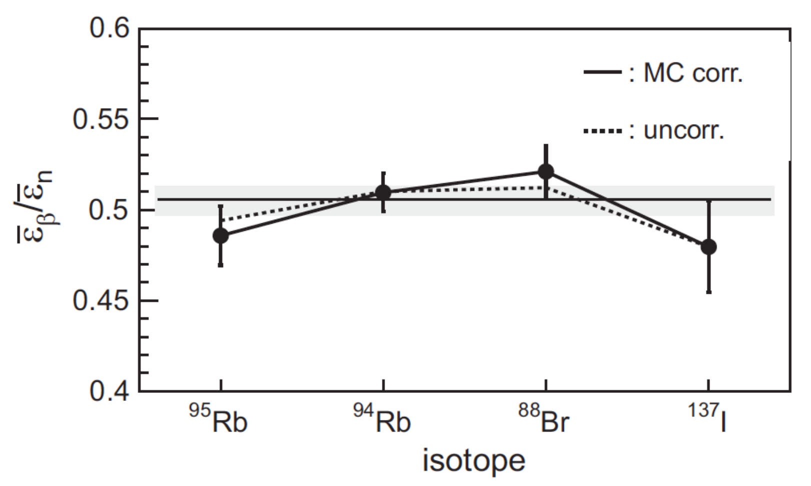
$$A_m^{n}(t) = \bar{\varepsilon}_{n} P_n \lambda_m N_m(t)$$

Solution (number of nuclei as a function of time):

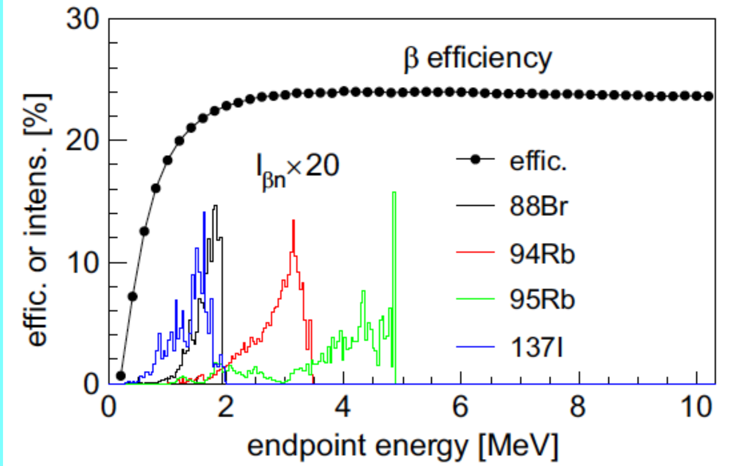
$$N_m(t) = \sum_{i=1}^{m} \left[\left(\prod_{j=i}^{m-1} \lambda_j b_{j,j+1} \right) \times \sum_{j=i}^{m} \left(\frac{N_i^0 e^{-\lambda_j t}}{\prod_{k=i,k\neq j}^{n} (\lambda_k - \lambda_j)} + \frac{P_i \left(1 - e^{-\lambda_j t}\right)}{\lambda_j \prod_{k=i,k\neq j}^{n} (\lambda_k - \lambda_j)} \right) \right]$$

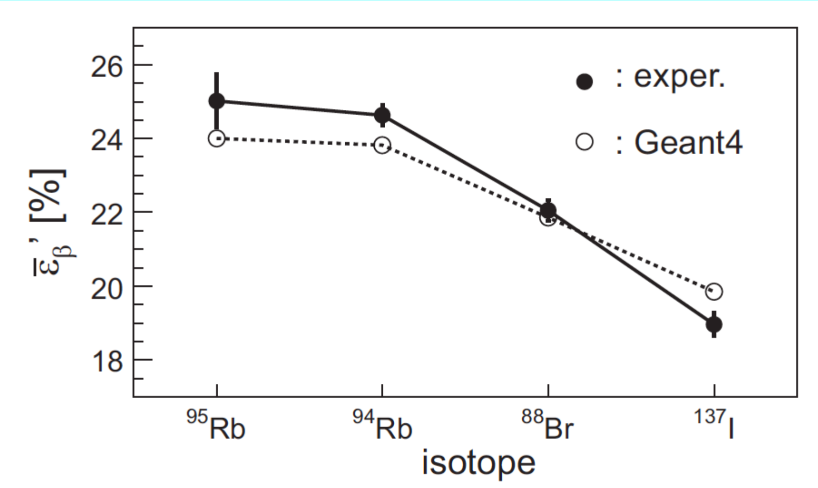
Determination of average efficiencies (nucleus dependent): source of systematic errors



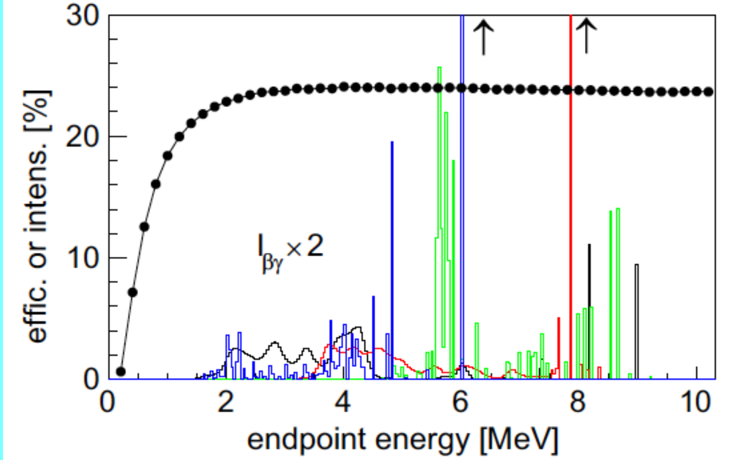


$$\frac{\overline{\varepsilon}_\beta}{\overline{\varepsilon}_n} = P_n \frac{N_\beta}{N_n}$$





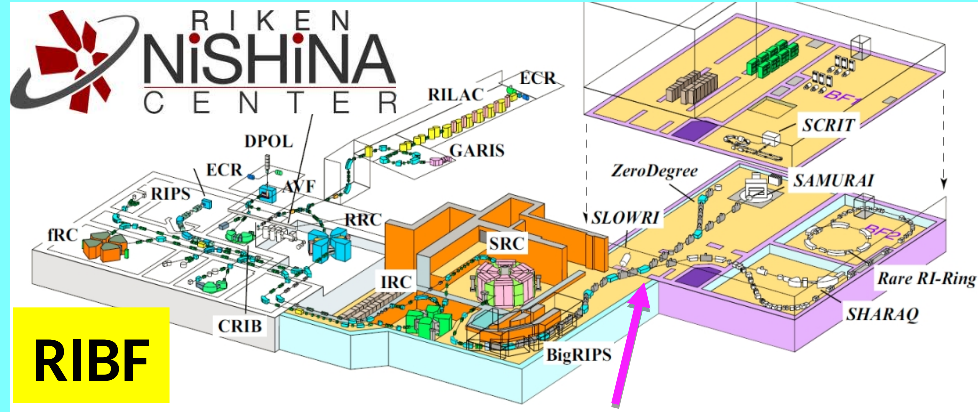
$$\overline{\varepsilon}'_\beta = \frac{N_{\beta n}}{N_n}$$



Agramunt+, NIMA807 (2016) 69

Experiment at fragmentation facility: production and selection of isotopes

345 MeV/u ^{238}U + ^{9}Be(4mm): fragmentation/fission



ΔE-TOF-Bρ method with track reconstruction → Improve Bρ and TOF resolution

Measure ΔE, TOF, Bρ @ 2nd stage

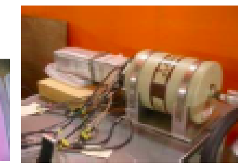
+ isomeric γ-ray

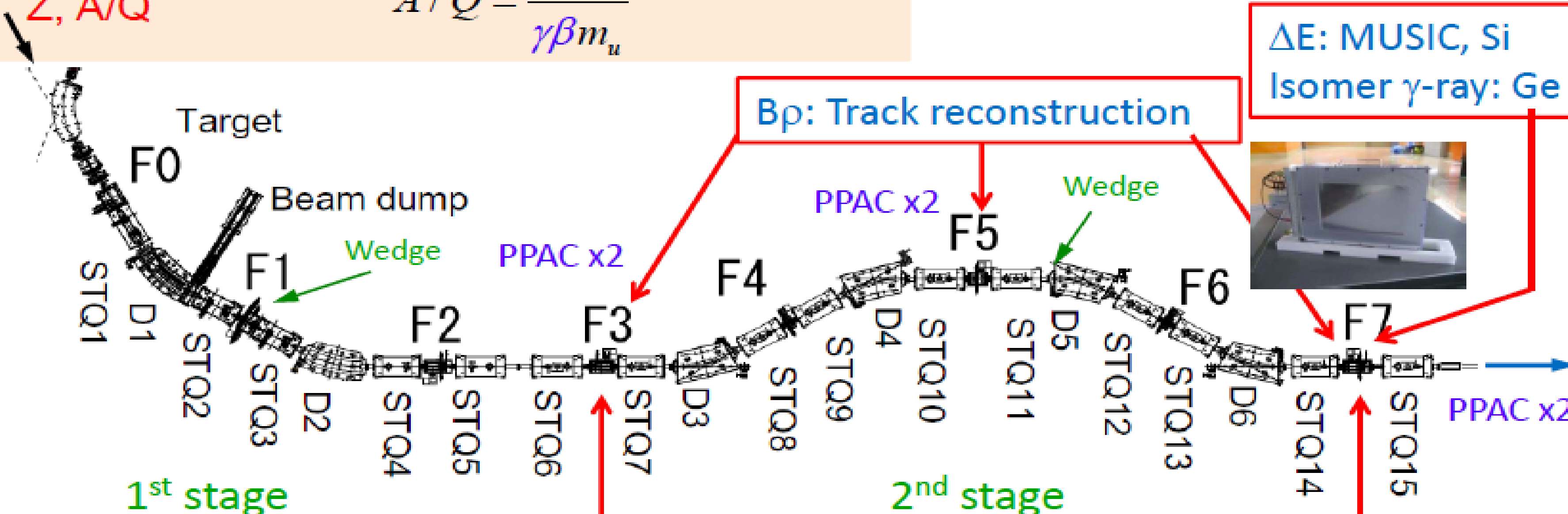
Z, A/Q

$$Z \leftarrow -dE/dx = f(Z,\beta)$$

$$A/Q = \frac{B\rho}{\gamma\beta m_u}$$







Bρ: Track reconstruction

ΔE: MUSIC, Si
Isomer γ-ray: Ge

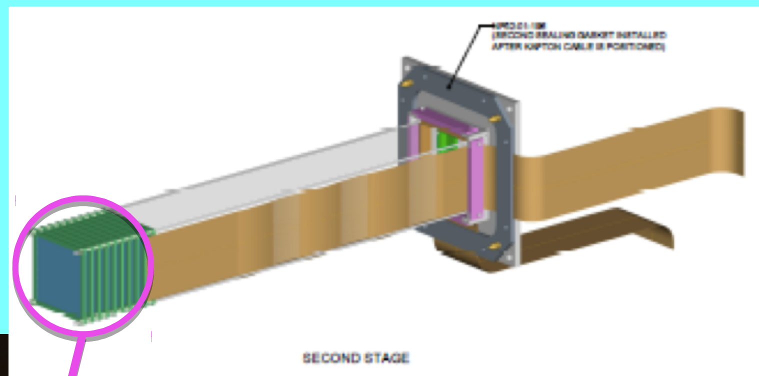
β: TOF

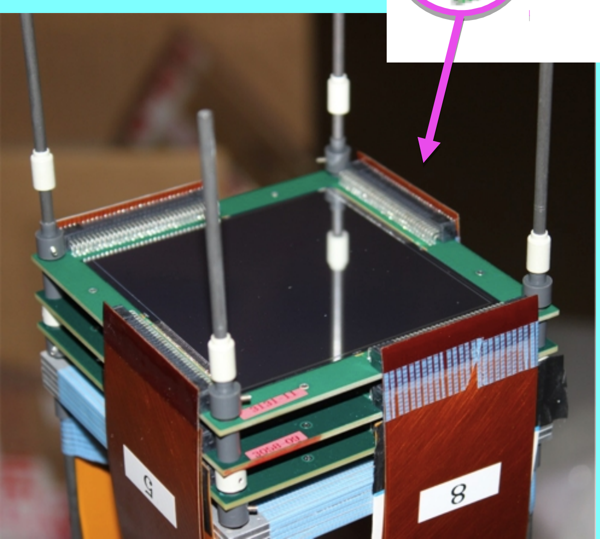
Plastic scintillator

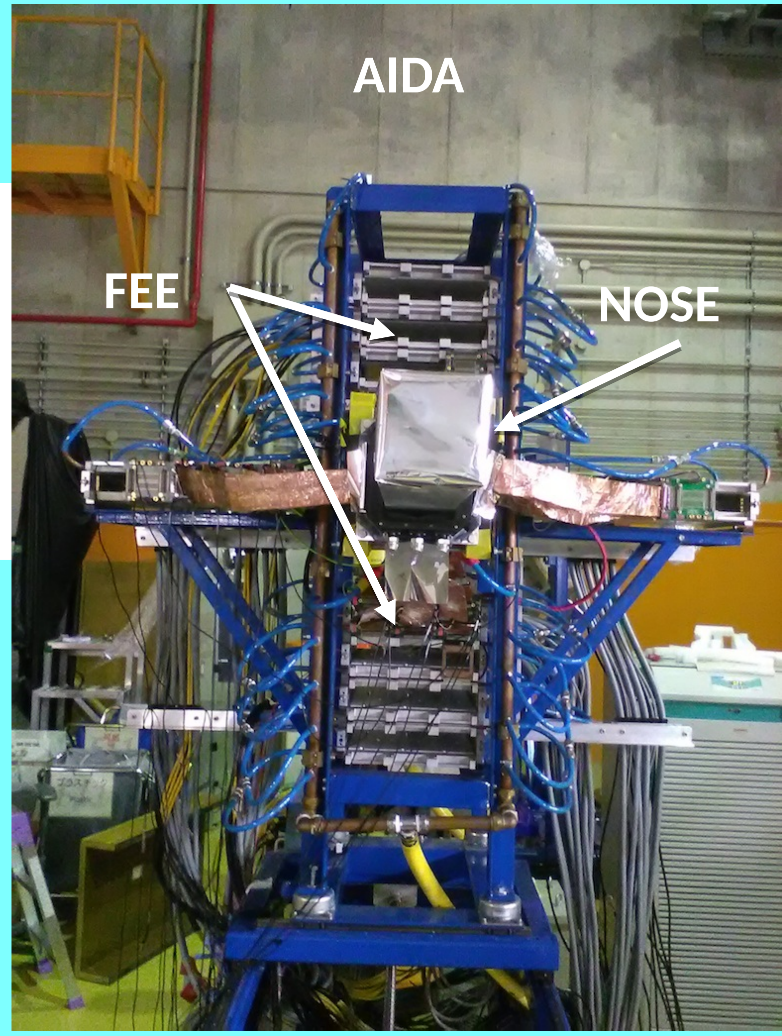
BigRIPS spectrometer

Advanced Implantation Detector Array (AIDA)

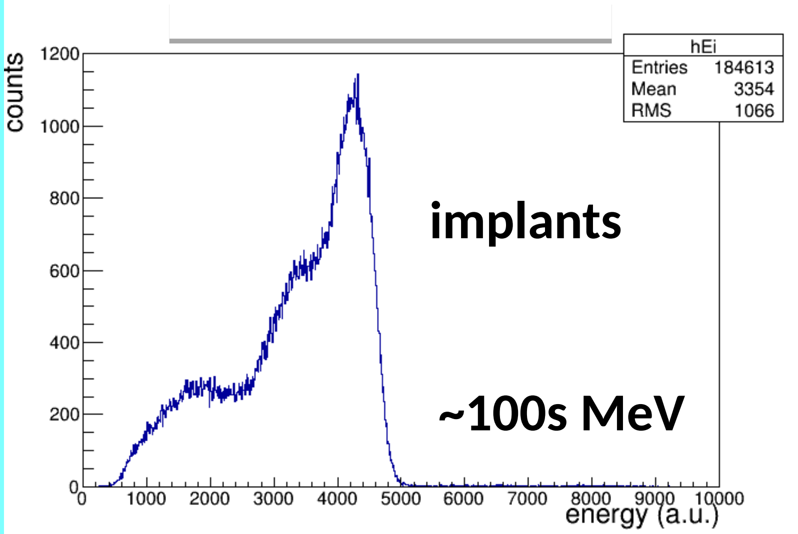
- Stack of six Si DSSD
- Size: 1mm×72mm×72mm
- Granularity: 128×128 pixels (0.51mm strip)
- Low gain (implant) and high gain (betas) preamplifiers
- Total data readout DACQ (1536 ch)

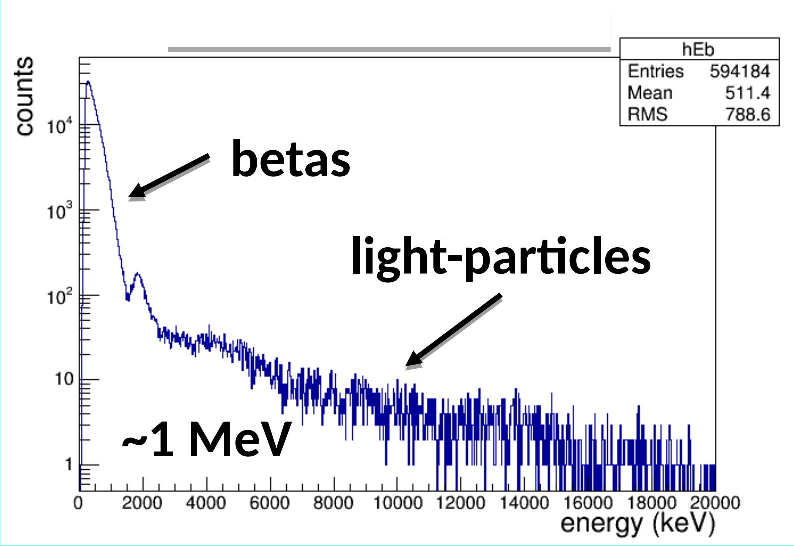




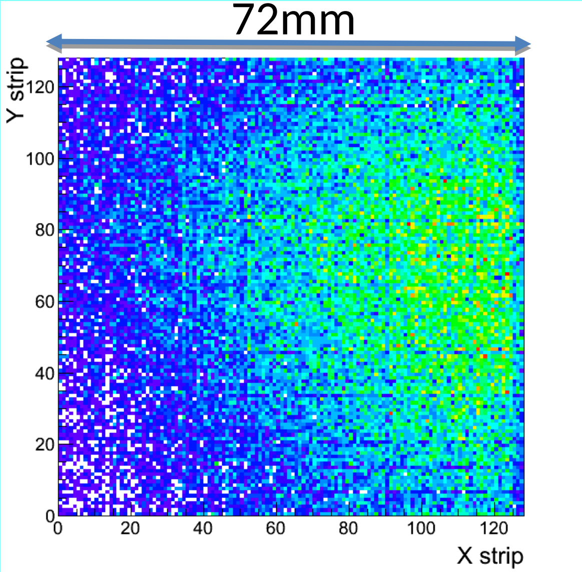


- Implants and betas are distinguished by the energy released in the detector
- Betas corresponding to each implanted ion are associated by spatial correlations

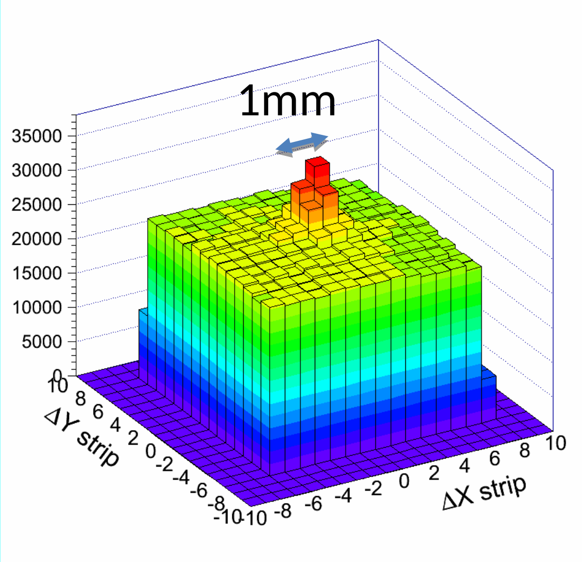




Implant position distribution

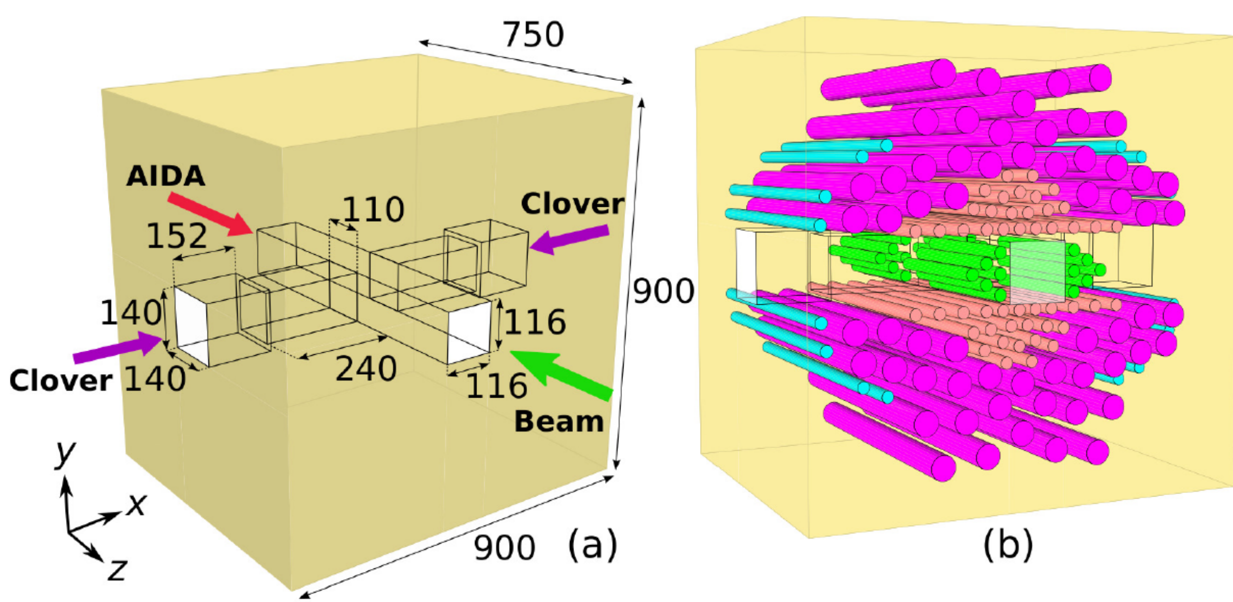


Implant-beta spatial correlation

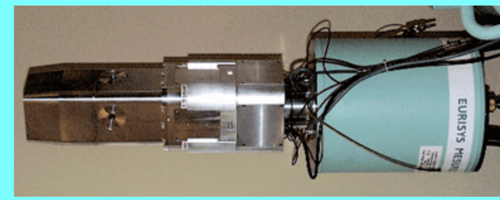


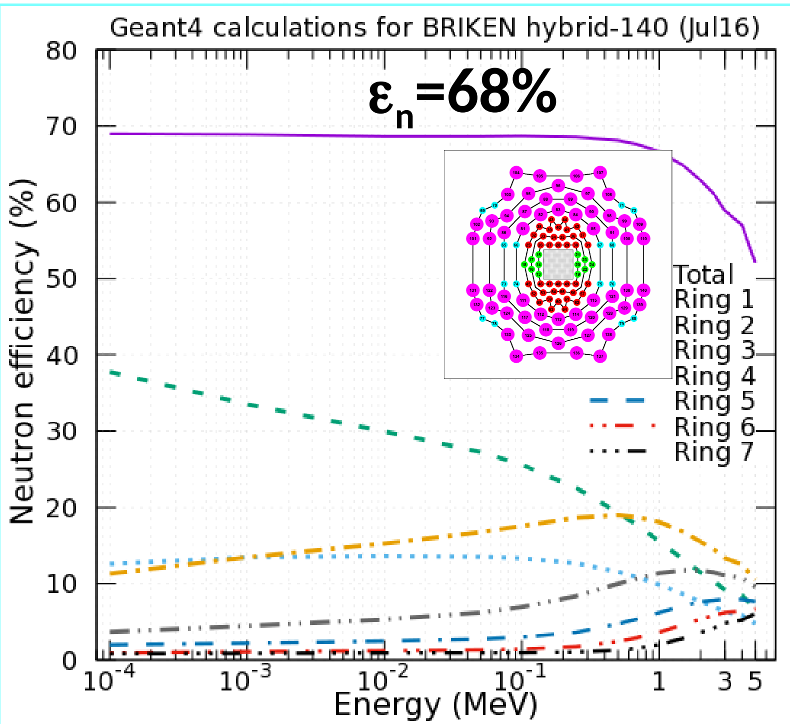
BRIKEN neutron counter

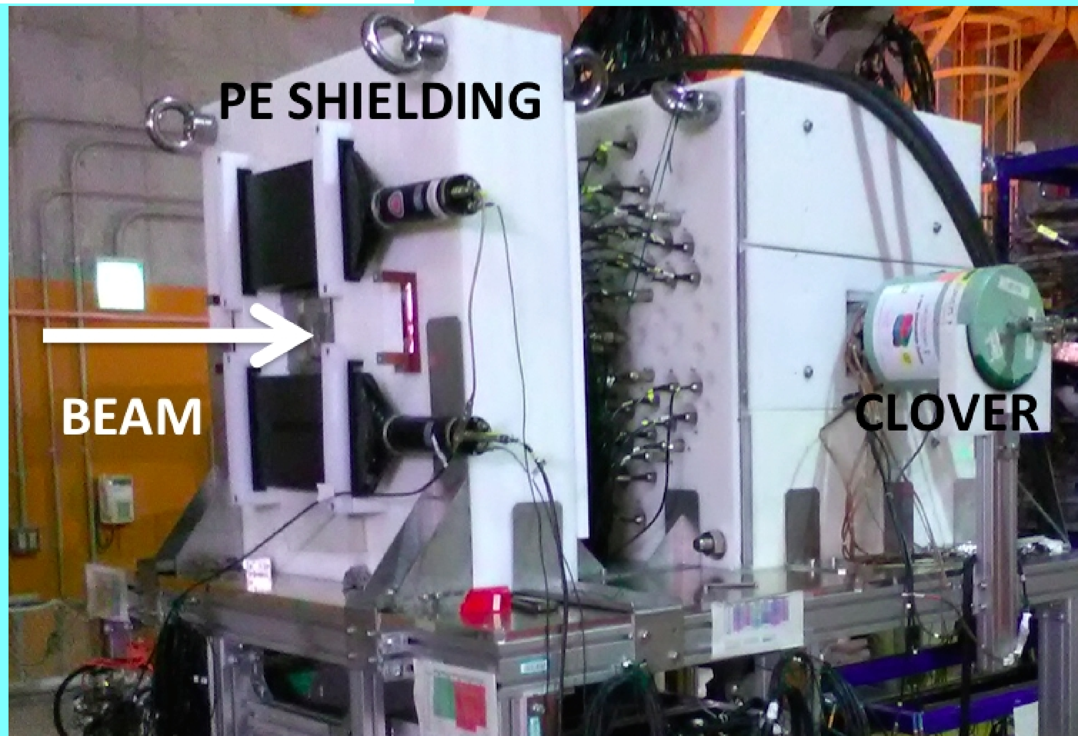
Tarifeño+, JInstr1(2017)P04006



Hybrid setup:
- 140 ^{3}He tubes (4 types)
- 2 CLOVER HPGe

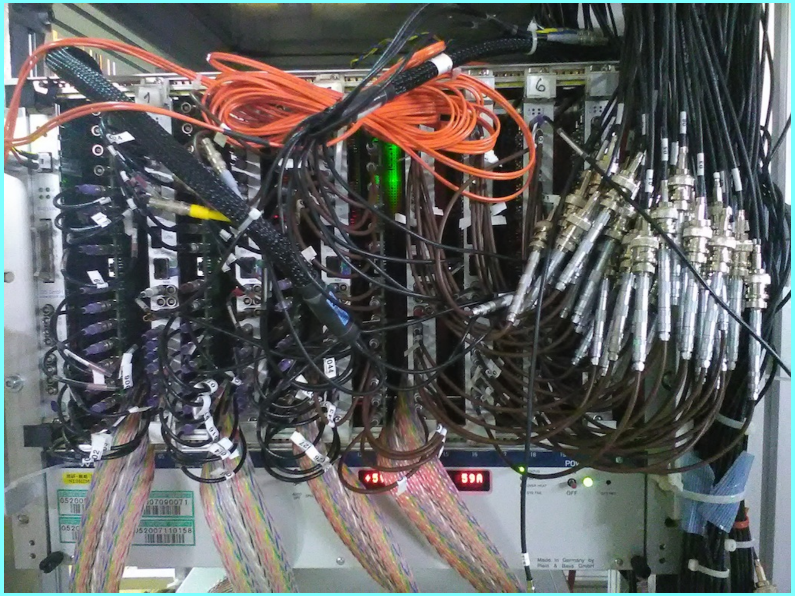






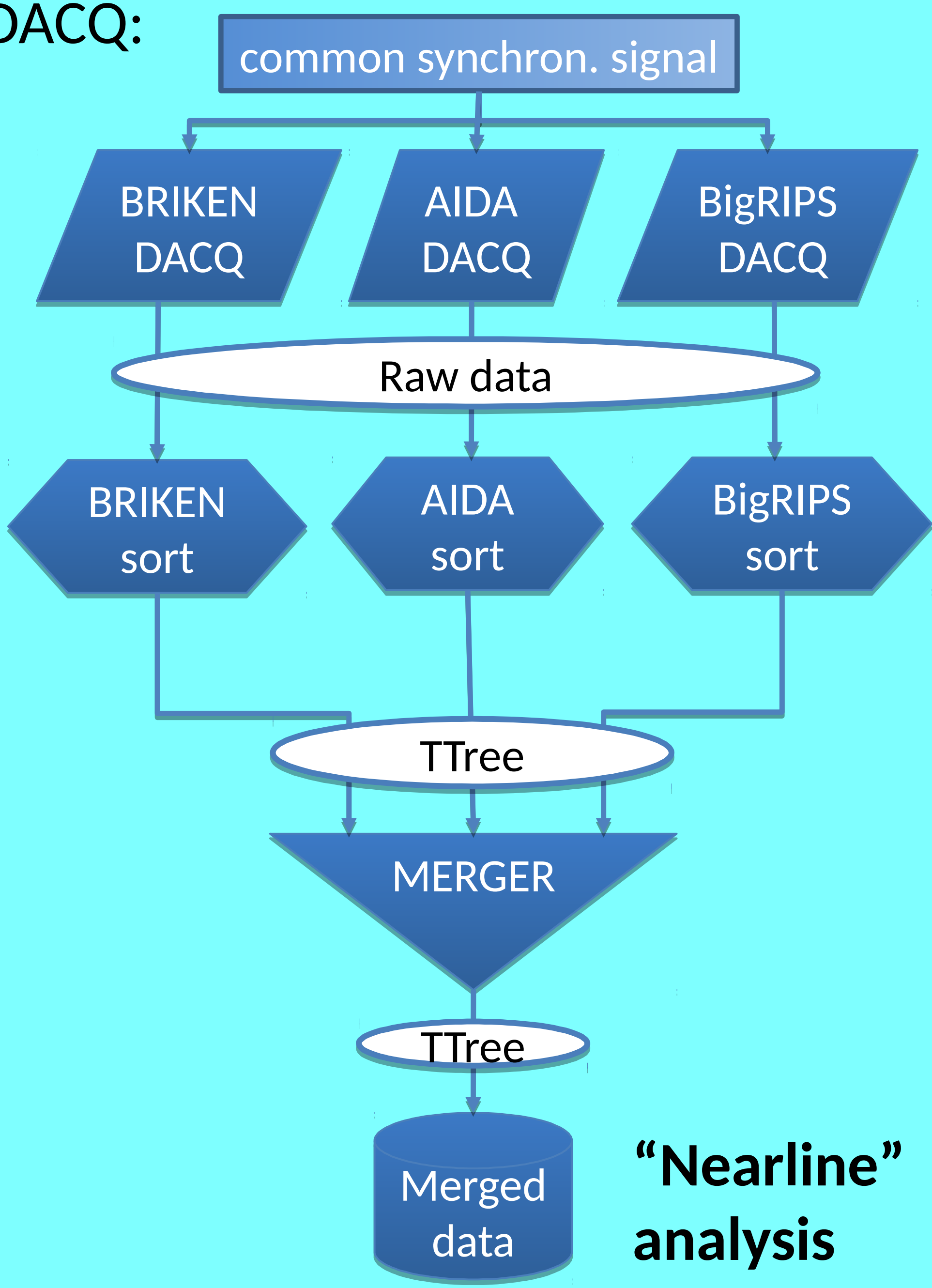
BRIKEN Gasific70 DACQ:

- ^{3}He tubes, CLOVER, ancillaries
- SIS3316 and SIS3302 digitizers
- Self triggered, common clock



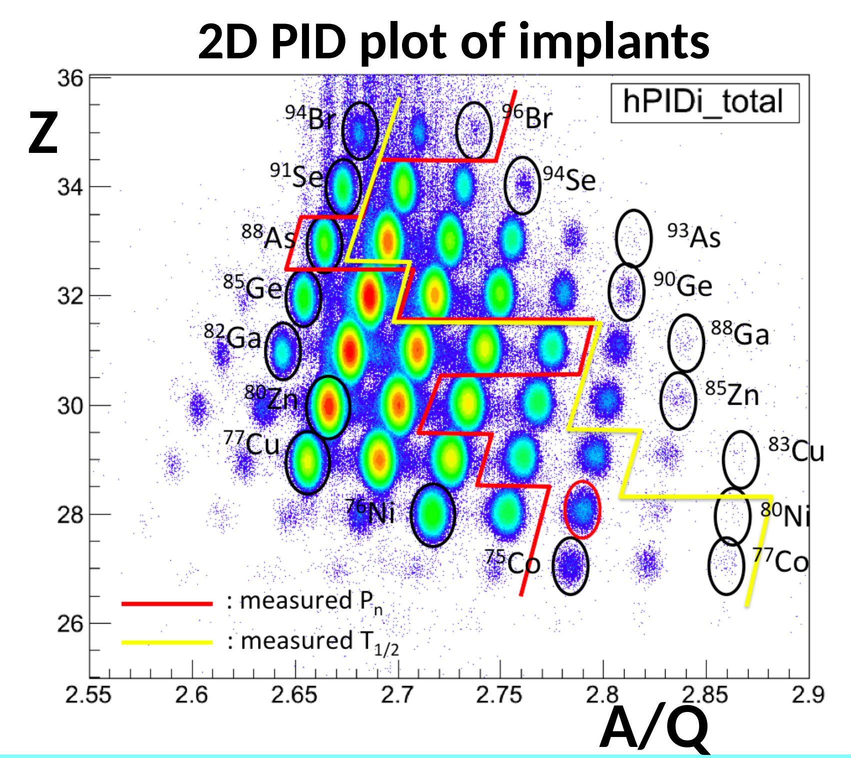
Agramunt+, NIMA807(2016)69

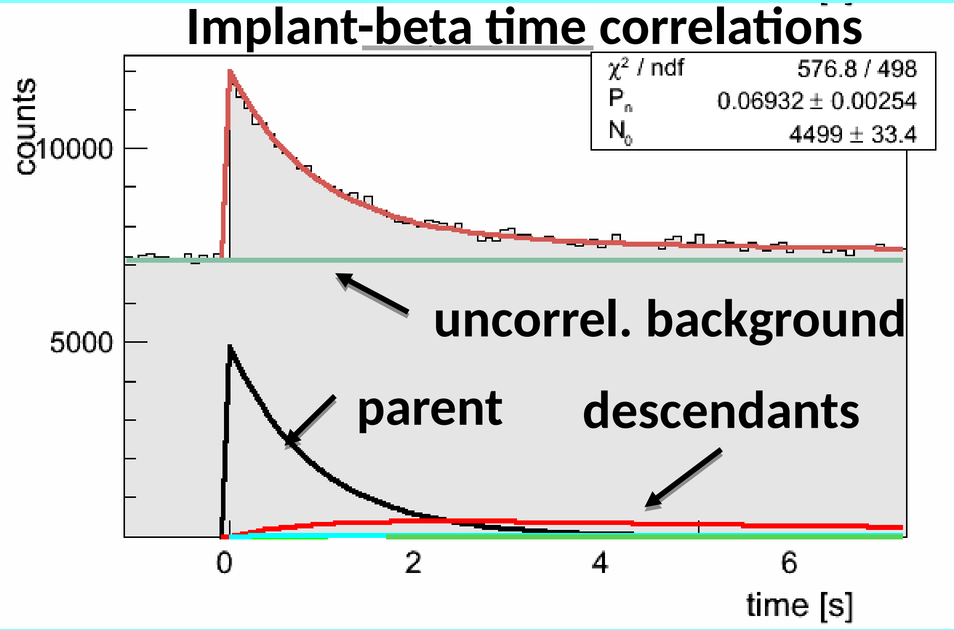
Merging of data from 3 independent DACQ:



Data analysis:

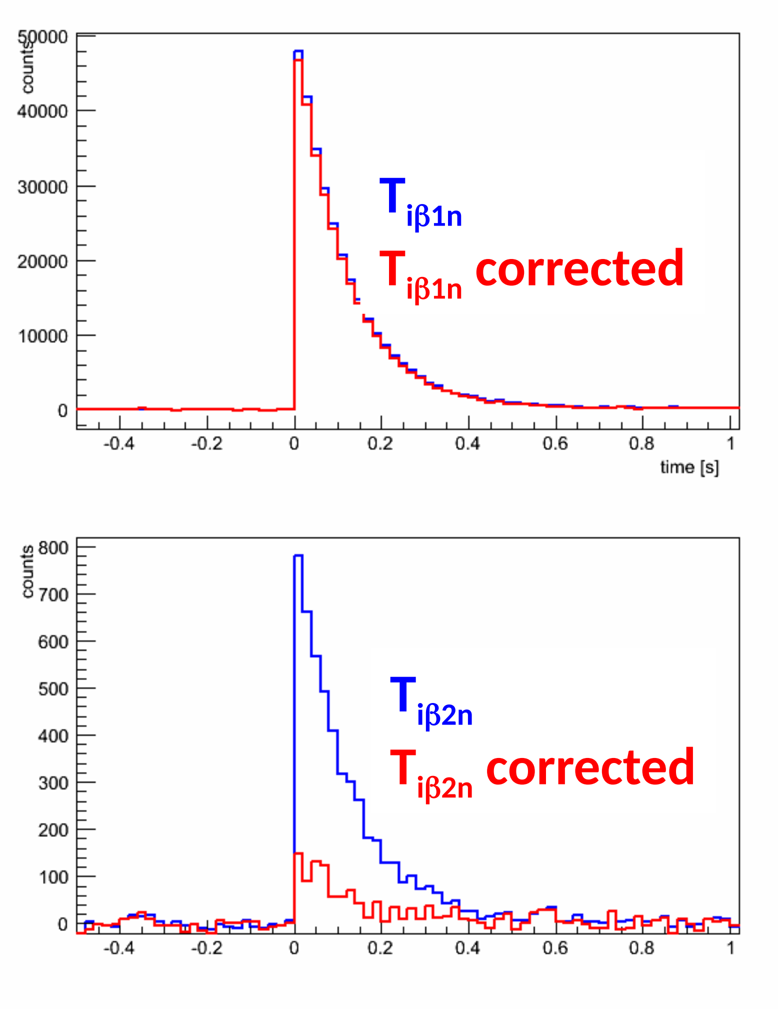
- Each implanted ion in AIDA is identified using the information from BigRIPS in prompt coincidence
- The associated β decay is assigned to the identified ion on a statistical basis from implant-β space-time correlations (delayed coincidence)
- Random coincidences are quantified from the backwards in time correlations
- Fitting with appropriate solutions of the Bateman equations serves to separate parent from descendant β signals





Tolosa+, NIMA925(2019)133

- Adding the condition that 1, 2, … neutrons come within ~200μs of the β we obtain the implant-β1n, implant-β2n, … time correlations
- Random 1n, 2n, … events contribute to the implant-βxn correlated background and must be corrected



- β2n decay contributes to the counts observed in β1n correlations and should be corrected

$$N_{1n}(t) = \varepsilon_n P_{1n} N_{dec} + 2\varepsilon_n (1-\varepsilon_n) P_{2n} N_{dec}$$

$$N_{2n}(t) = (\varepsilon_n)^2 P_{2n} N_{dec}$$

$$N_{1n(2n)}(t) = 2\frac{1-\varepsilon_n}{\varepsilon_n} N_{2n}(t)$$

- To disentangle parent and descendant contributions we fit the time spectra with appropriate solutions of Bateman equations

Tolosa+, NIMA925 (2019) 133

Fit functions:

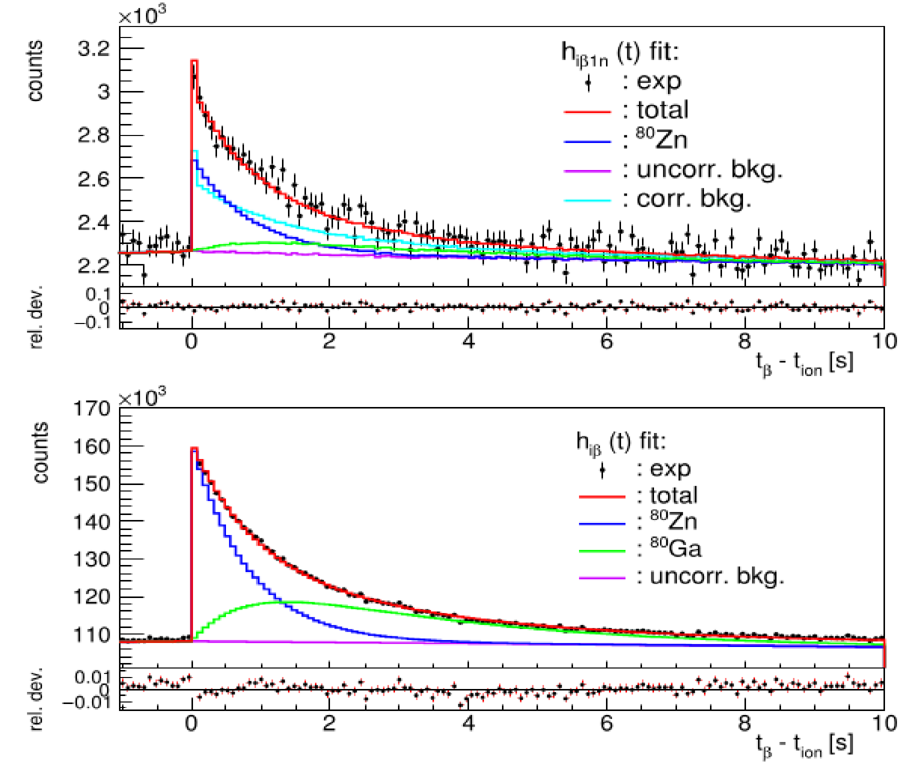
$$f_{\beta}(t) = \sum_{i \in \beta} \bar{\varepsilon}_{\beta}^{i} \lambda_i N_i(t)$$

$$f_{\beta 1n}(t) = \sum_{j \in \beta 1n} \bar{\varepsilon}_{\beta}^{j} \bar{\varepsilon}_{n}^{j} P_{1n}^{j} \lambda_j N_j(t)$$

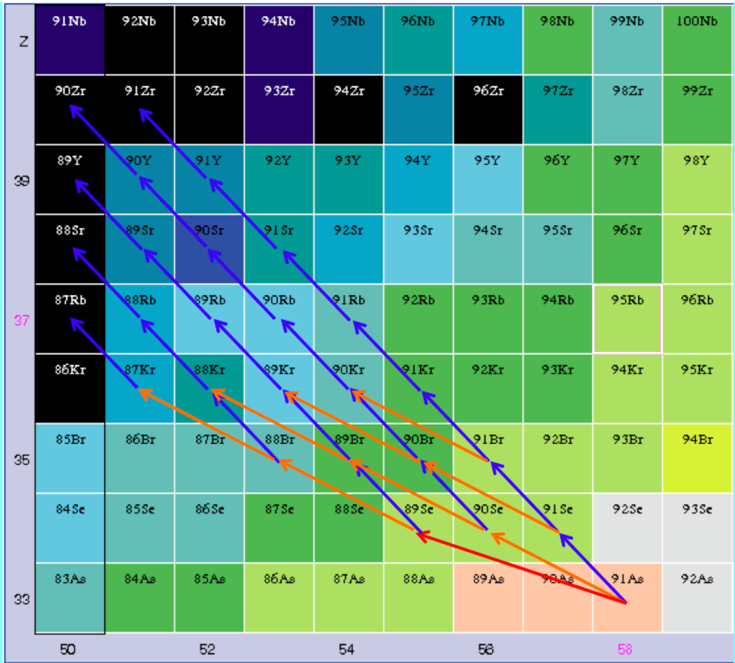
$$f_{\beta 2n}(t) = \sum_{k \in \beta 2n} \bar{\varepsilon}_{\beta}^{k} \left(\bar{\varepsilon}_{n}^{k}\right)^2 P_{2n}^{k} \lambda_k N_k(t)$$

$$N_k(t) = N_1 \prod_{i=1}^{k-1} (b_{i,i+1} \lambda_i) \times \sum_{i=1}^{k} \frac{e^{-\lambda_i t}}{\prod_{j=1 \neq i}^{k} (\lambda_j - \lambda_i)}$$

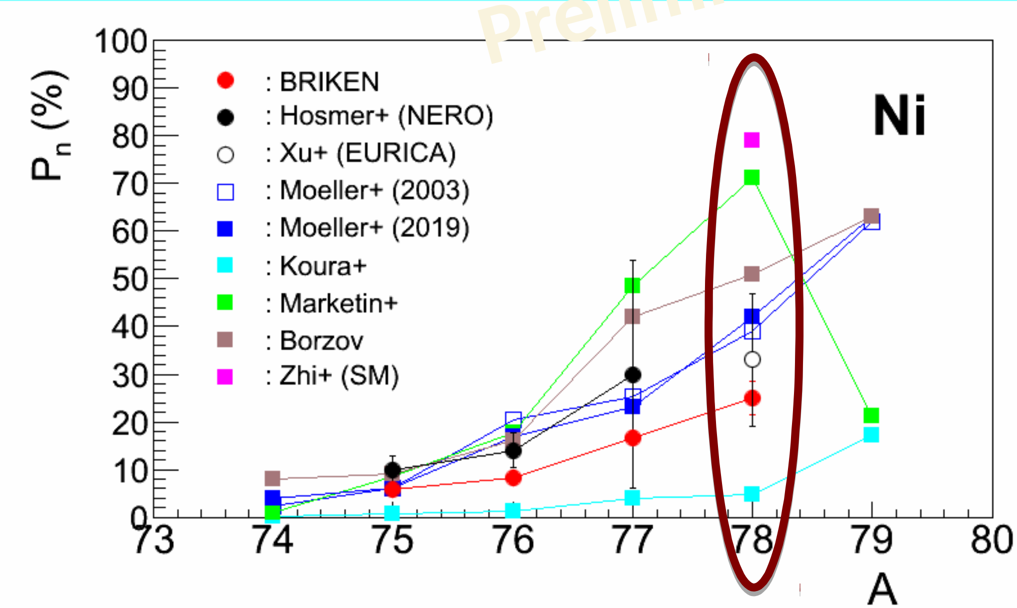
$$b_{i,i+1} = P_{1n}^{i},\ P_{2n}^{i} \text{ or } 1 - P_{1n}^{i} - P_{2n}^{i}$$



Decay pattern can be quite complex far from stability



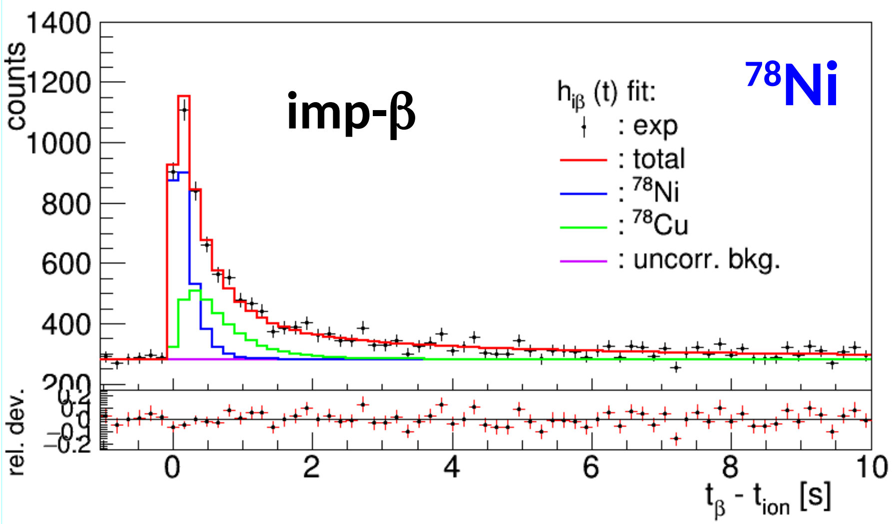
Ni isotopes: P_{1n} compared with theory and previous data

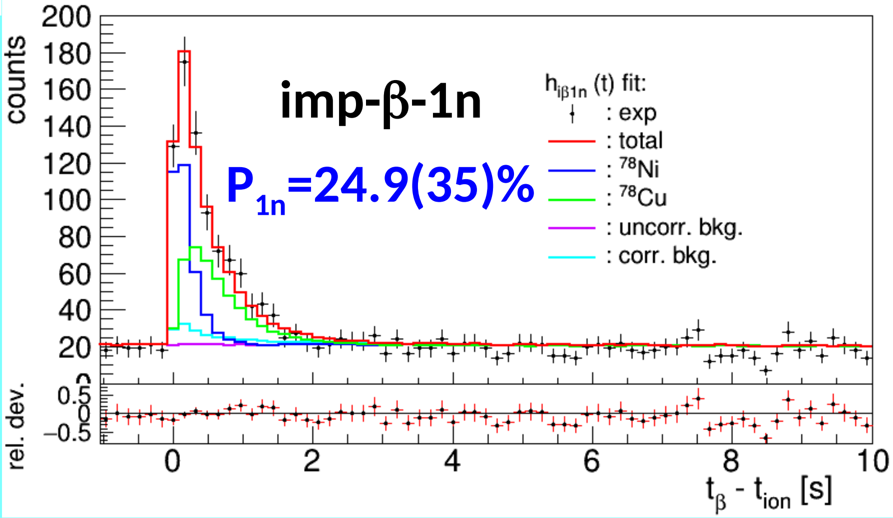


Hosmer+, PRC82(2010)025806
Z.Y.Xu, PhD Thesis (2014)
Borzov, PRC71(2005)065801
Zhi+, PRC87(2013)025803

- **At shell closure: large spread of theoretical estimates, all off the experiment**

First detection of n from ^{78}Ni



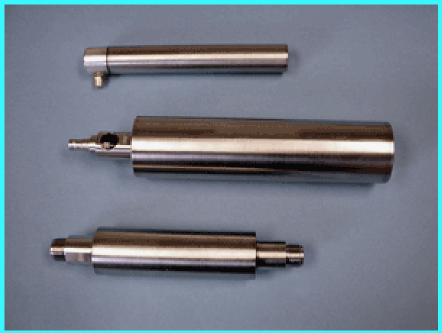


(from Alvaro Tolosa PhD Thesis)

Appendix: Measurement of β-delayed neutron spectra

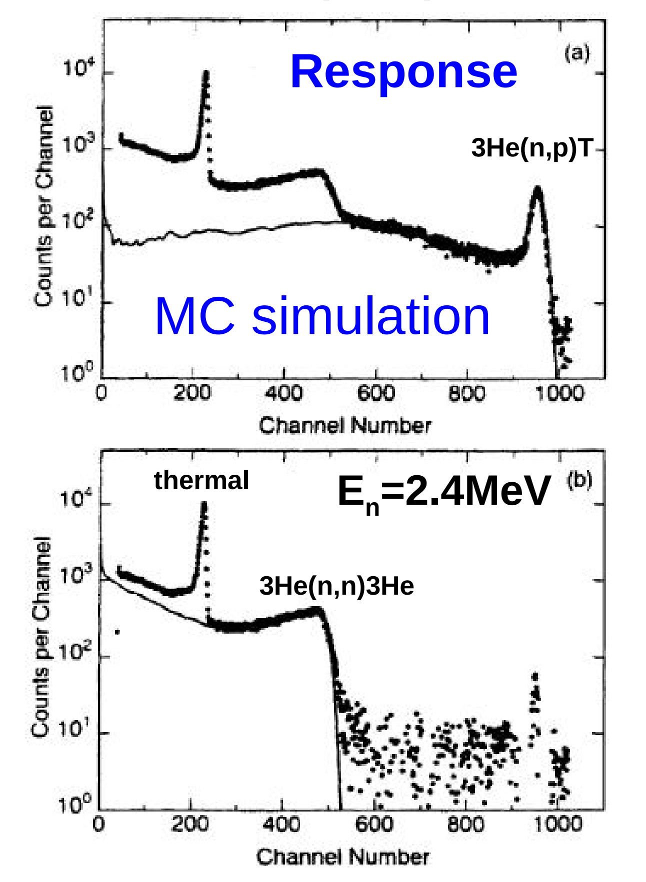
^{3}He detectors

Cutler-Shalev ionization chamber



$$n + {}^3He \rightarrow p + {}^3H + 764\,keV + E_n$$

NIMA422 (1999) 69



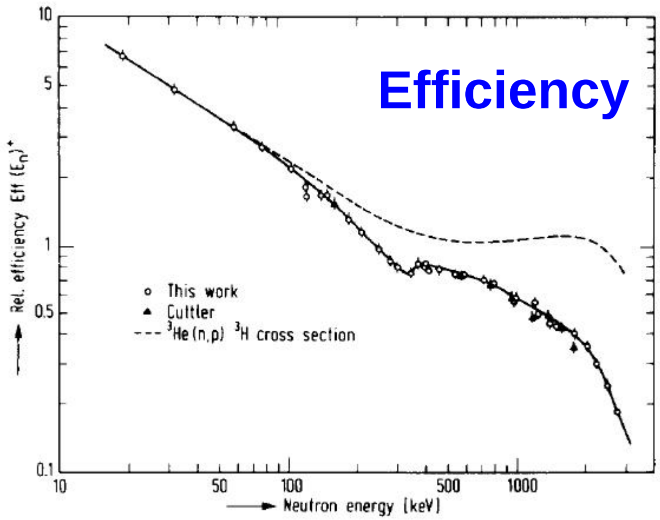


Fig. 7. Relative efficiency of the ^{3}He fast neutron spectrometer, normalized to previous measurements[15]), along with the ^{3}He(n,p)^{3}H cross section.

NIM144 (1977) 253

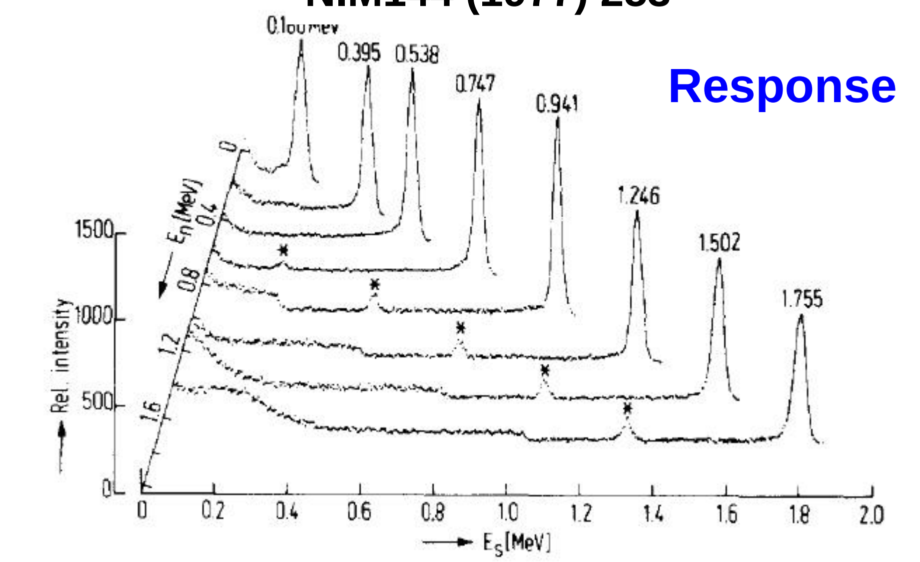
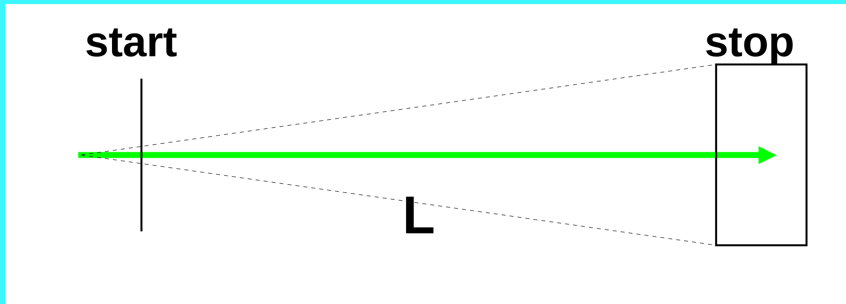


Fig. 2. Response of the ^{3}He ionization chamber to monoenergetic neutrons produced by the ^{7}Li(p,n)^{7}Be reaction. E_n is the

- ✗ Limited efficiency
- ✗ Integral measurement
- ✔ Very good energy resolution
- ✔ Linear energy response

Time of Flight Spectrometer

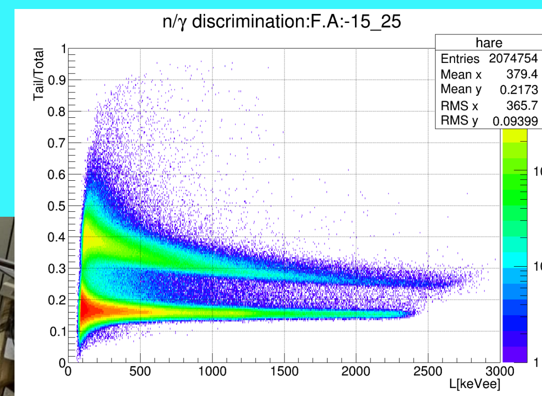


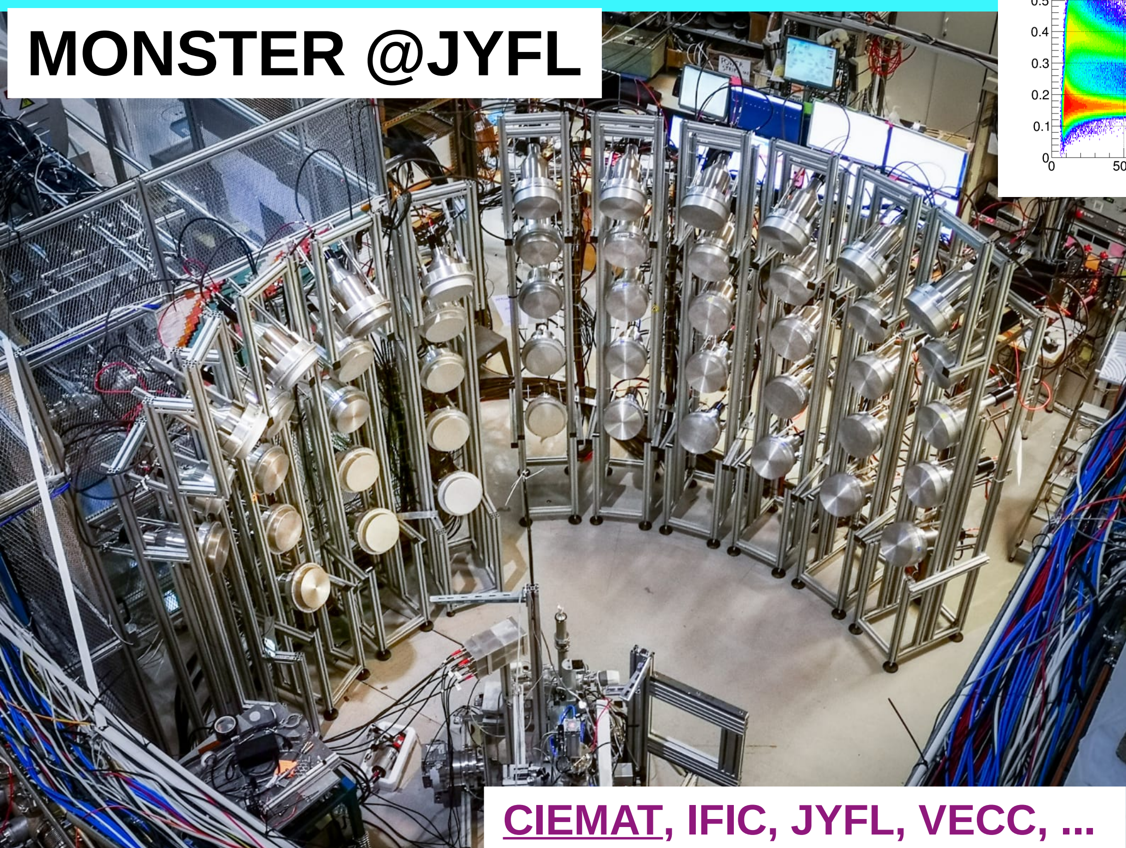
$$E_n = \frac{1}{2} m_n \frac{L^2}{t^2}$$

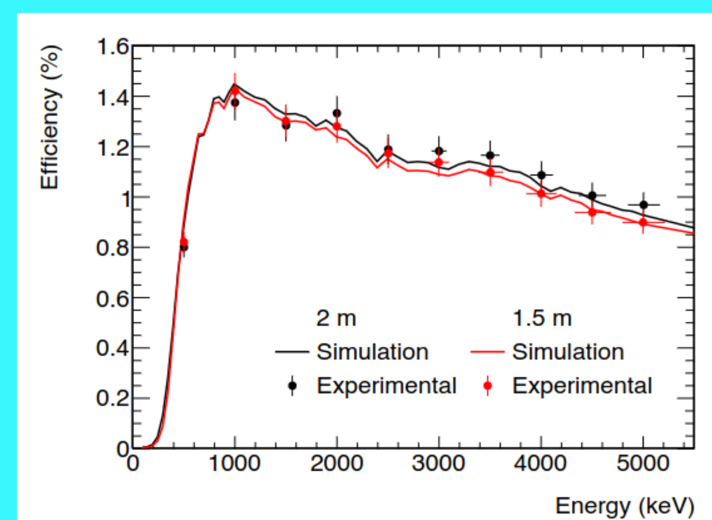
$$\frac{\Delta E}{E} = 2\sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}$$

- ✔Event-by-event measurement
- ✔Good intrinsic efficiency
- ✗ Poor energy resolution
- ✗ Non-linear energy response

<u>Start</u>: n-generation related signal
<u>Stop detector</u>: organic scintillator (liquid for γ/n discrimination)



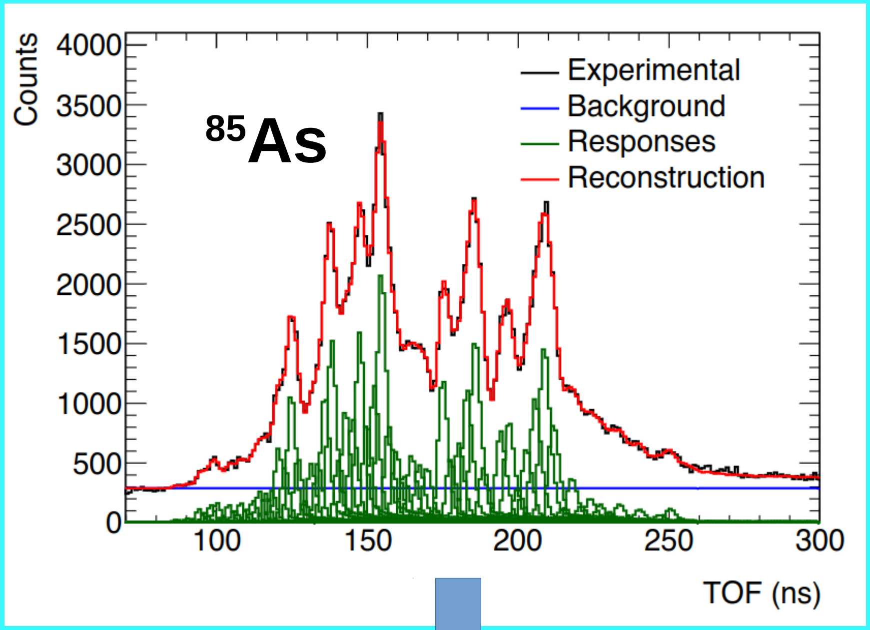




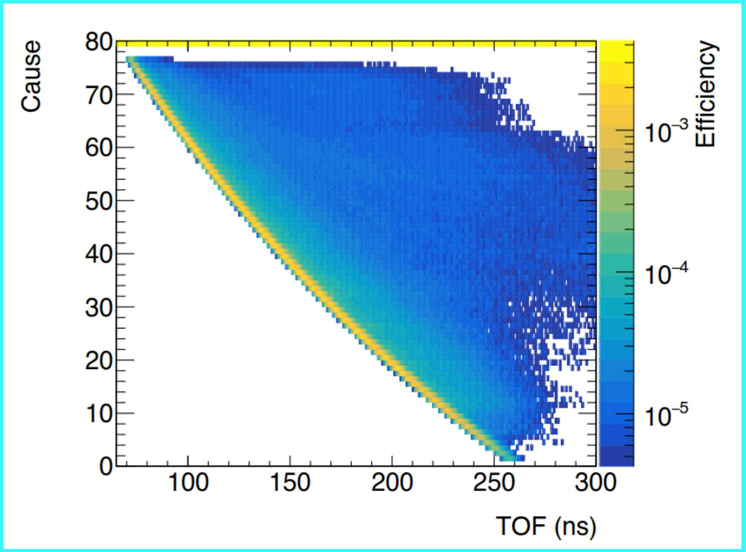
A.R. Garcia+, JINST/(2012)C05012

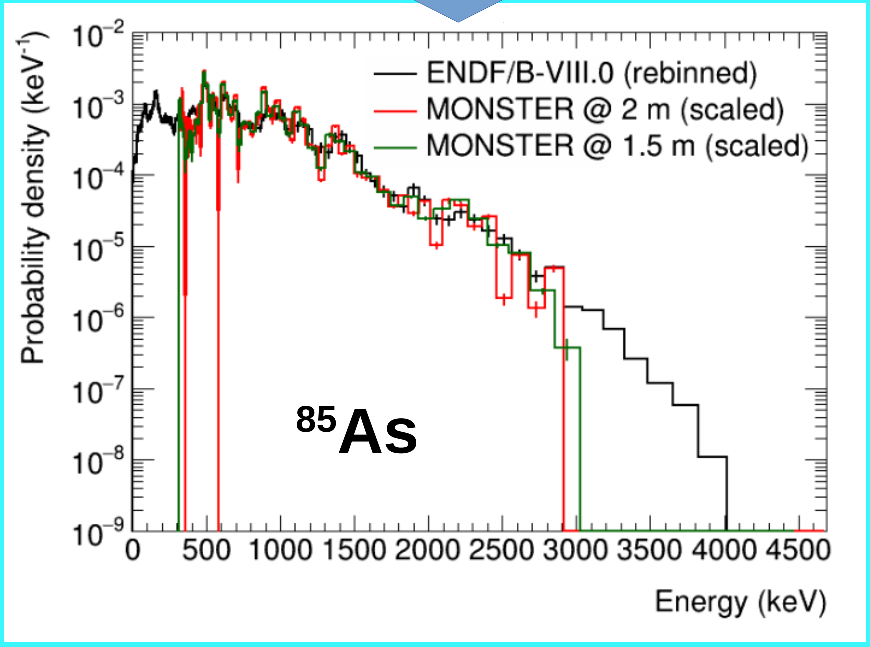
A. Perez-Rada, PhD Thesis, 2023

ToF spectrum deconvolution using EM algorithm (Bayesian iterative)

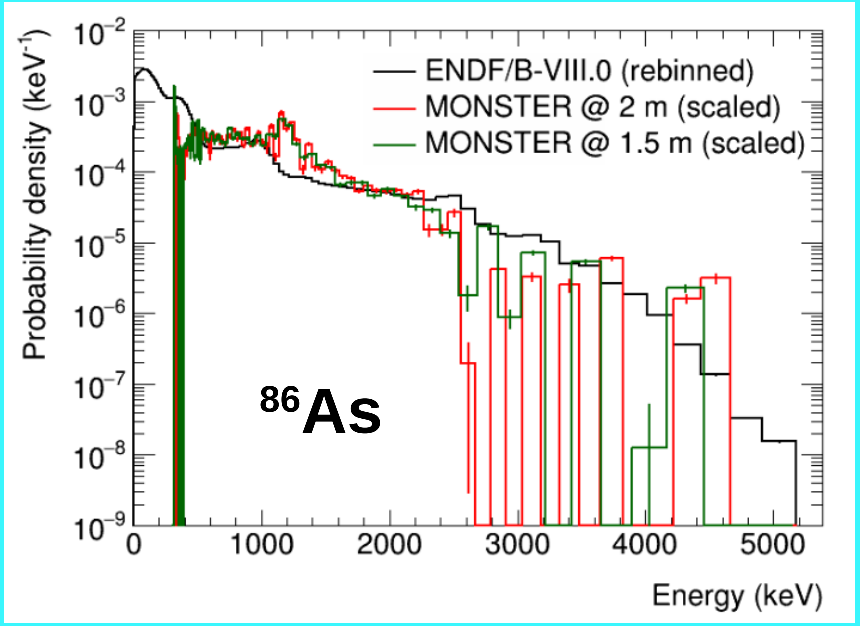


Geant4 simulated Energy vs. ToF response matrix





Comparison with previous measurement



First-ever measurement of ^{86}As βDN spectrum