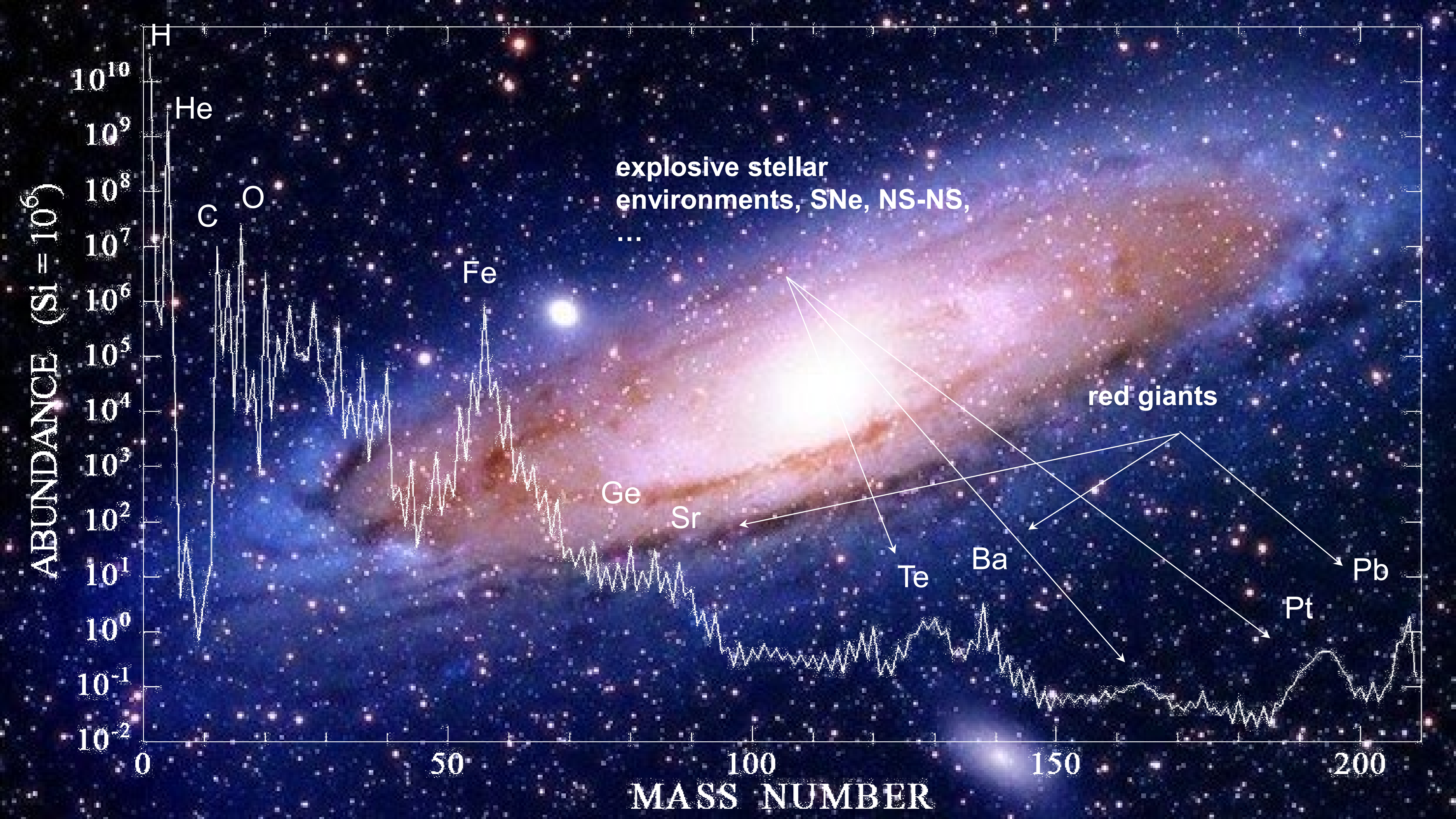
Recent results on neutron capture reactions at n_TOF

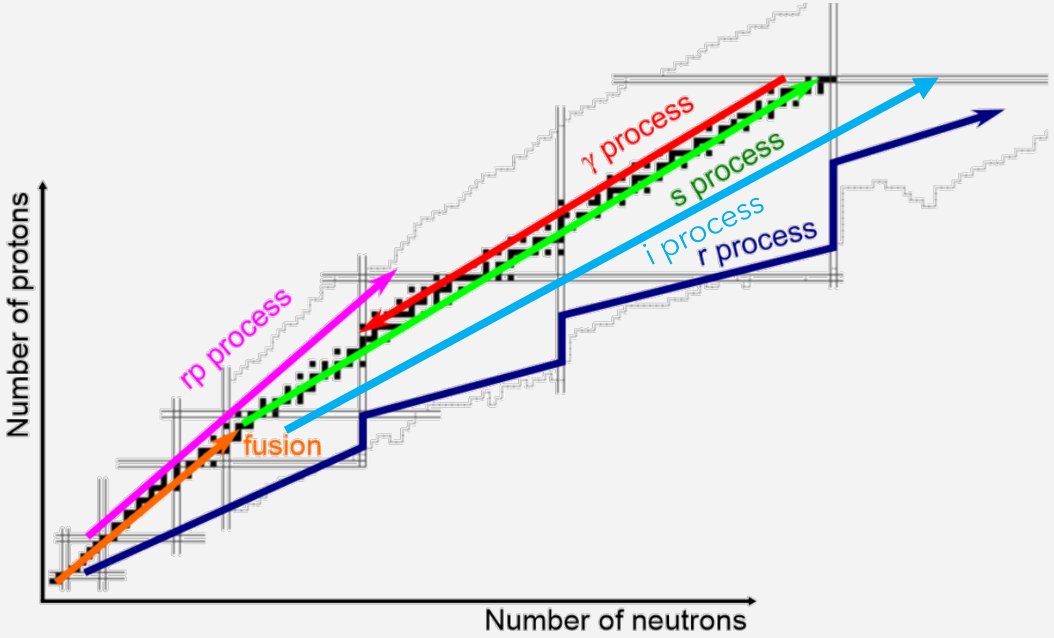
Riccardo Mucciola

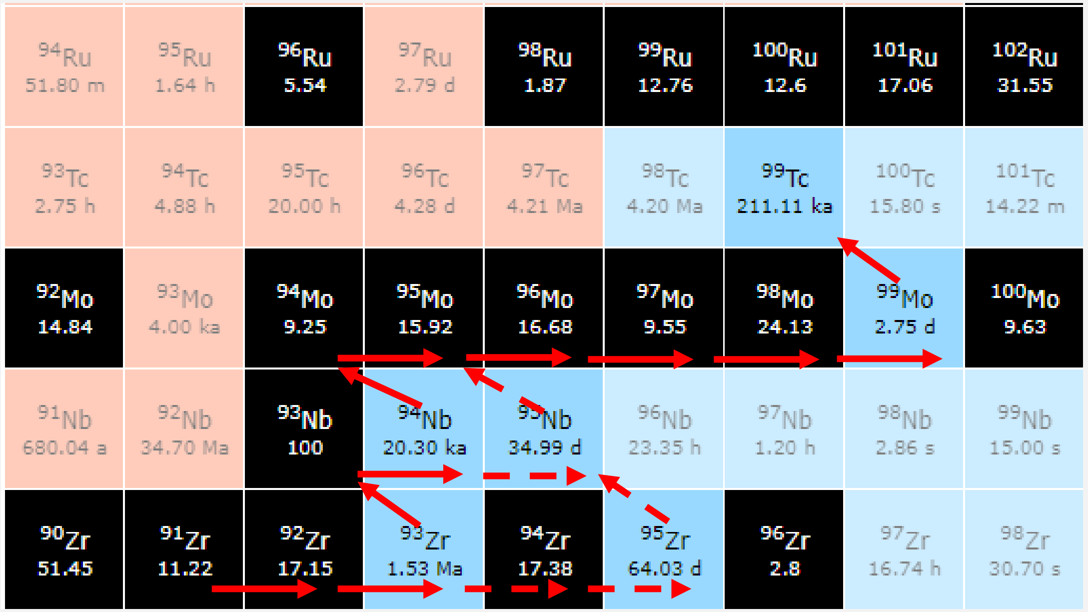


H
He
C
O
Fe
Ge
Sr
Te
Ba
Pt
Pb
explosive stellar environments, SNe, NS-NS, ...
red giants
ABUNDANCE (Si = 10^6)
MASS NUMBER
10^{10}
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
0
50
100
150
200

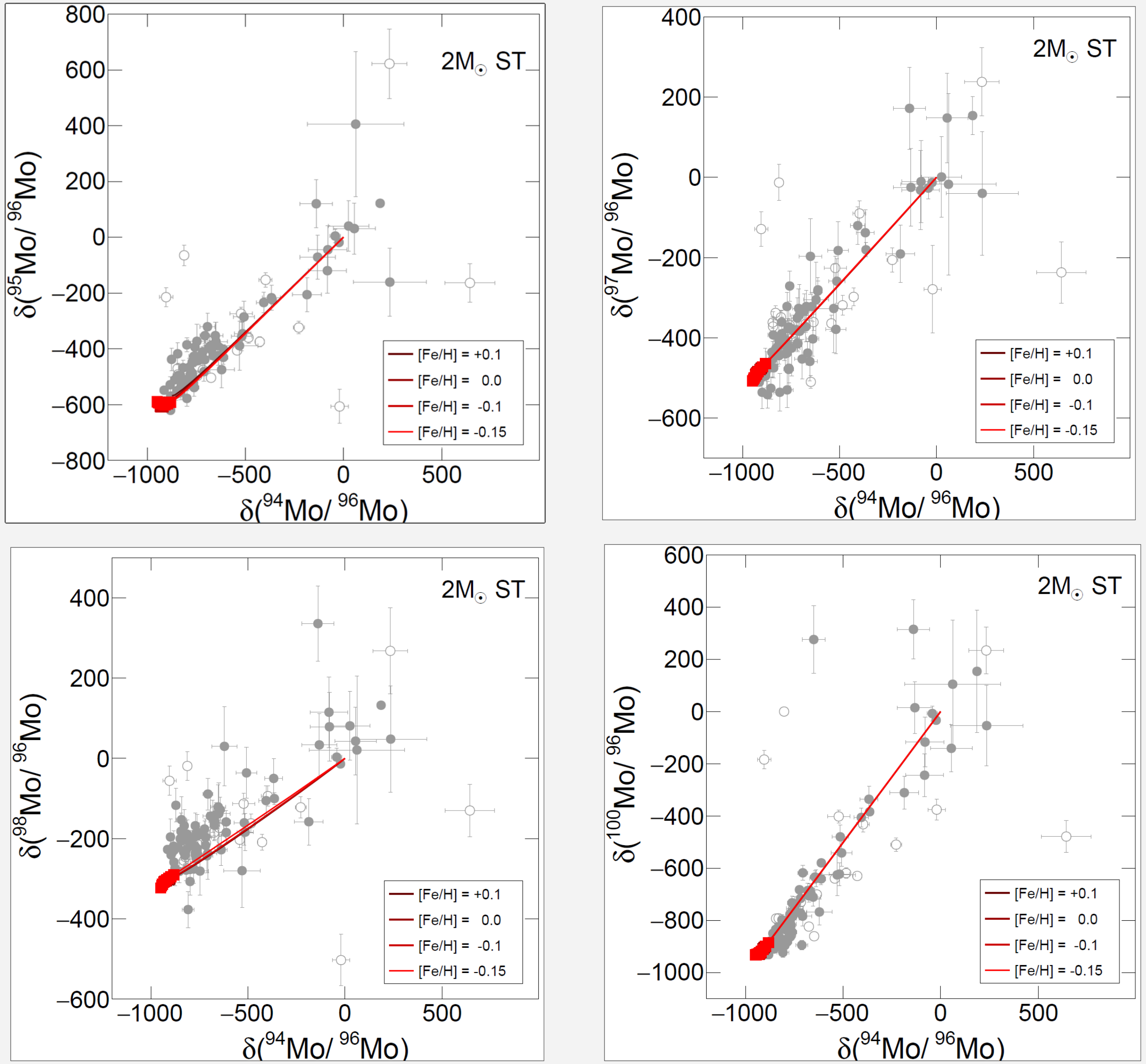
Stellar nucleosynthesis

- Four main nucleosynthesis processes for elements heavier than iron: s-process, r-process, i-process, and p-process;
- Some isotopes, due to position in the chart of nuclides, can be synthetized only by one process;
- Possible to set constraints on intensity of the processes.





Presolar grain composition



- Silicon carbide (SiC) grains help to study abundance in presolar AGB;
- Abundances of elements in grains are expressed as:

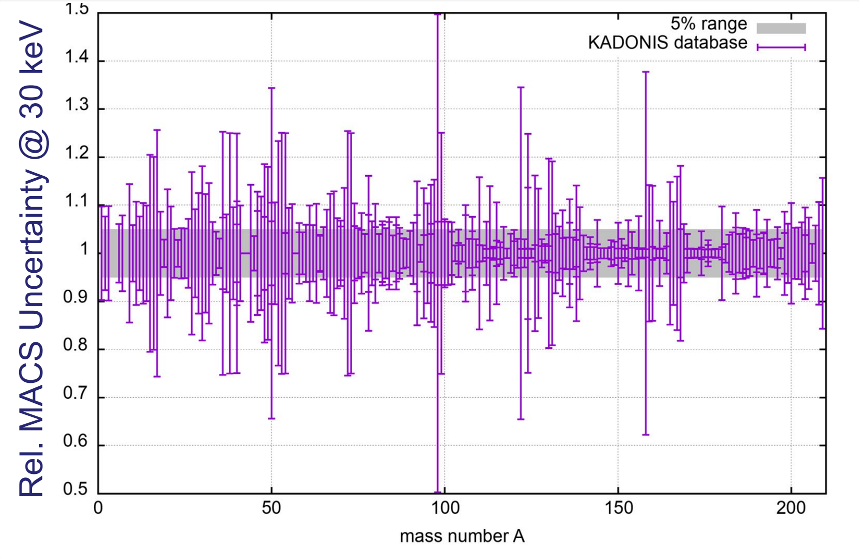
$$\delta\left(\frac{^{95}Mo}{^{96}Mo}\right) = 10^3 \times \left[\left(\frac{^{95}Mo}{^{96}Mo}\right) \Big/ \left(\frac{^{95}Mo}{^{96}Mo}\right)_{\odot} - 1 \right]$$

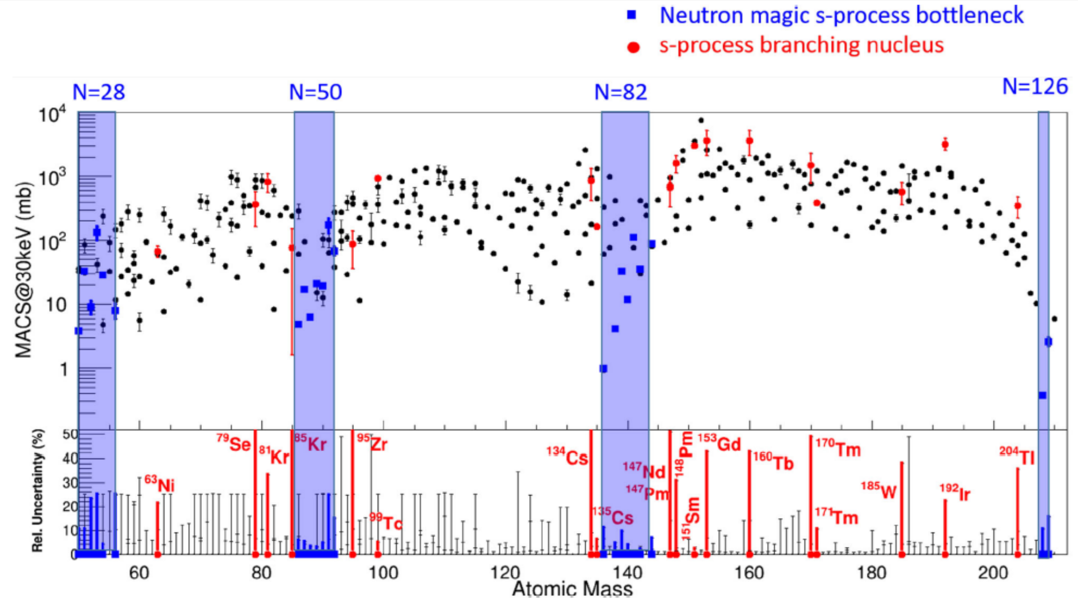
- Comparison of abundances predicted by models with observation.

Status of data in literature

- MACS uncertainty still far from 5% target for many isotopes,
- Many important isotopes for s-process still reported with significant uncertainties,
- Majority of long-lived unstable isotopes never measured,

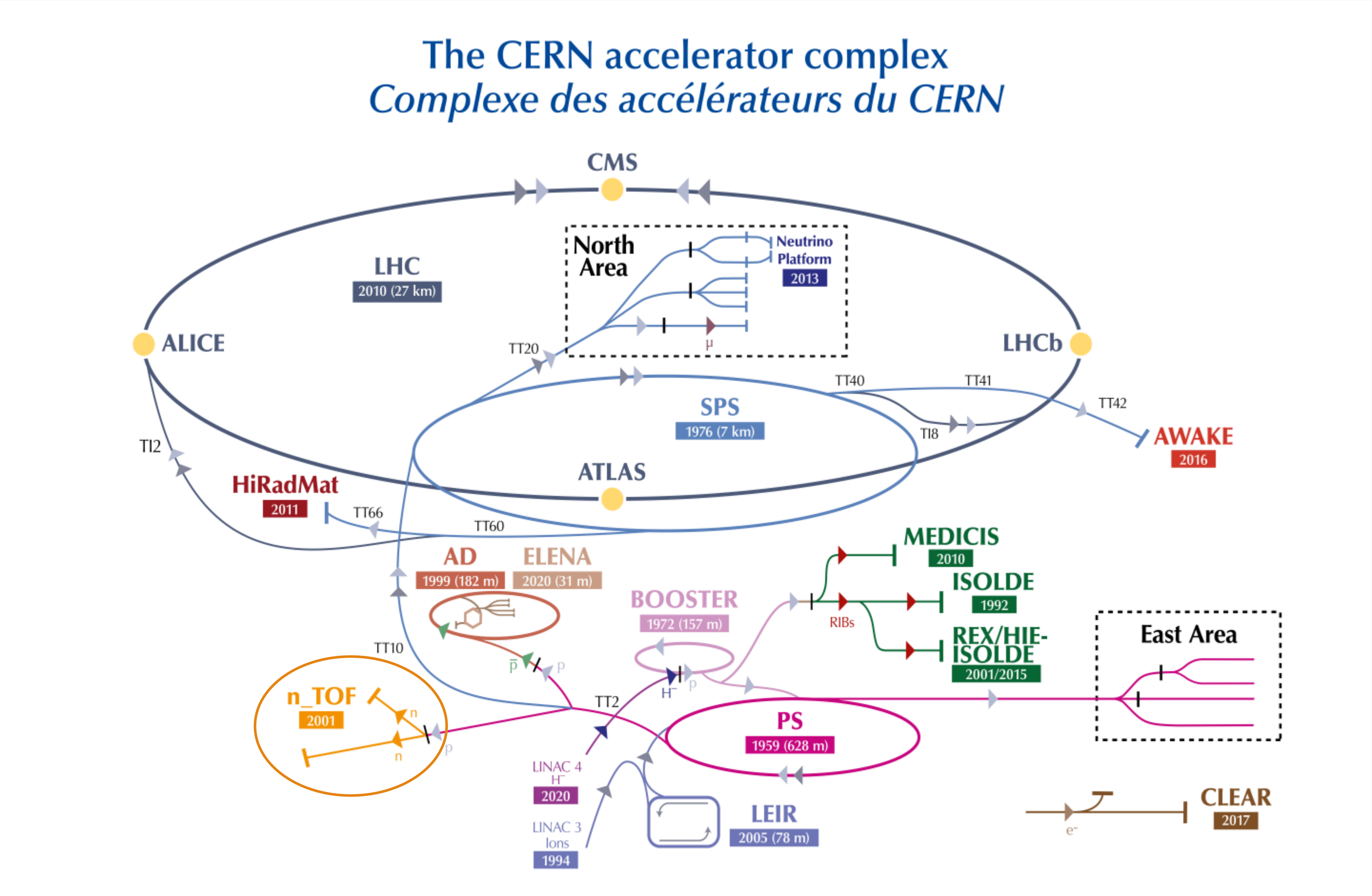
Nuclear data still necessary today!



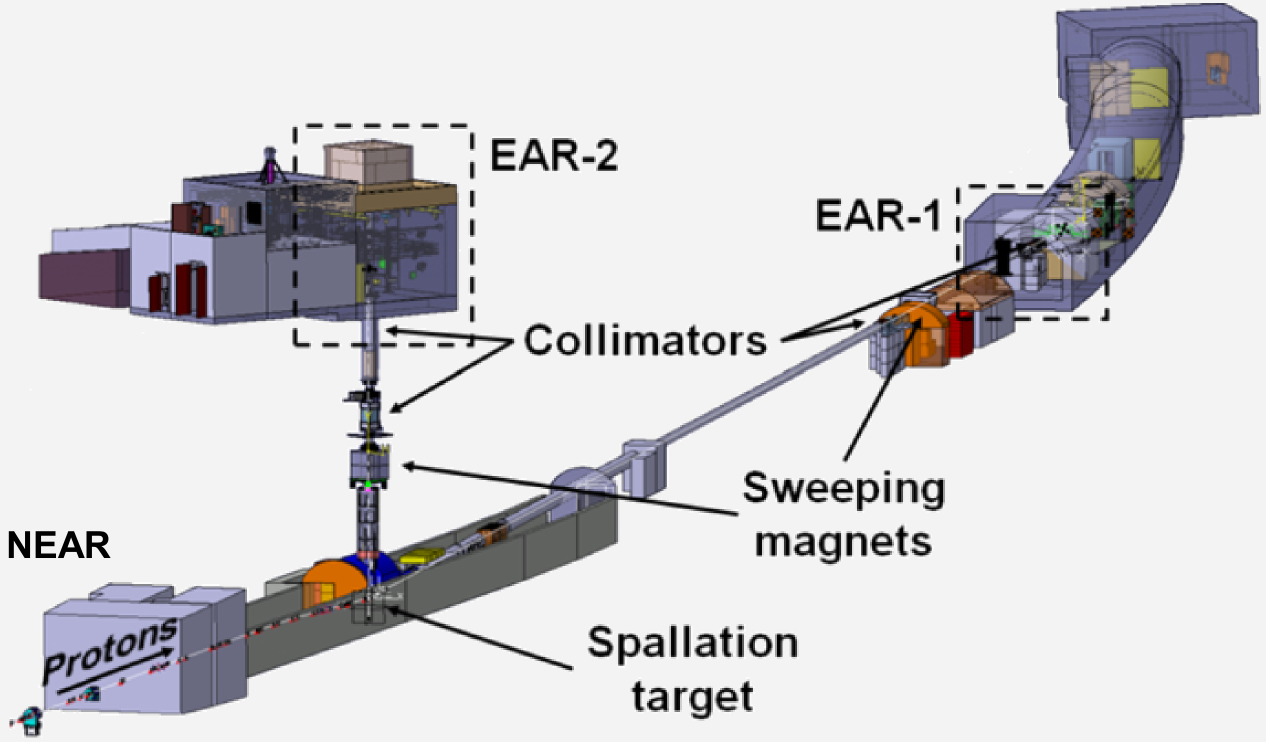


n_TOF facility

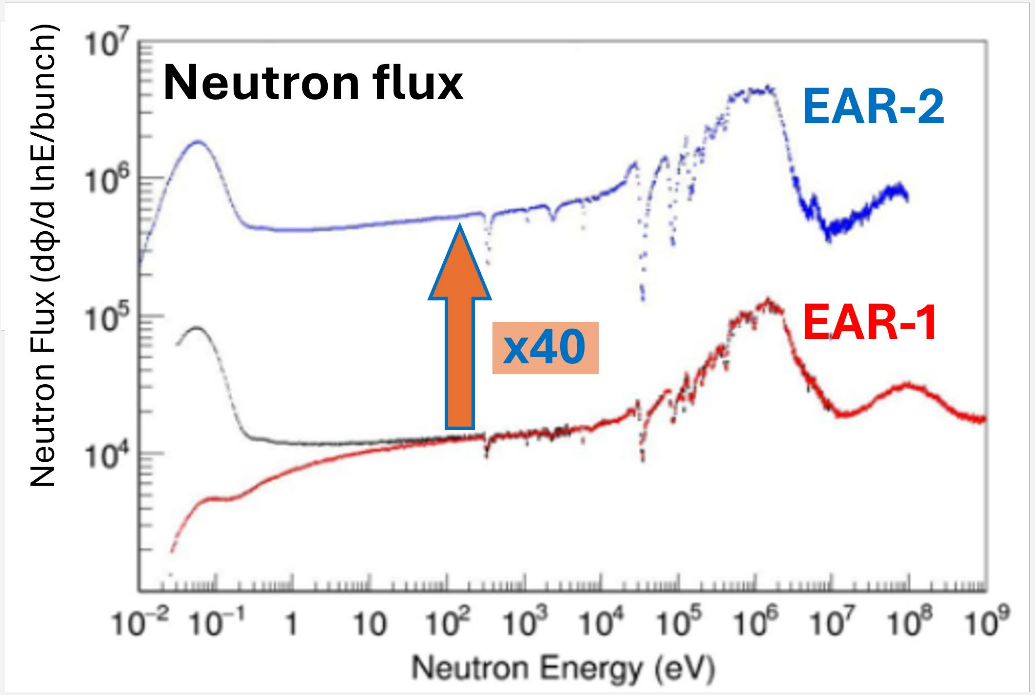
n_TOF @ CERN


The CERN accelerator complex
Complexe des accélérateurs du CERN
CMS
North Area
Neutrino Platform
2013
LHC
2010 (27 km)
ALICE
TT20
LHCb
TT40
TT41
TT42
SPS
1976 (7 km)
TI8
AWAKE
2016
TI2
ATLAS
HiRadMat
2011
TT66
TT60
MEDICIS
2010
AD
1999 (182 m)
ELENA
2020 (31 m)
ISOLDE
1992
BOOSTER
1972 (157 m)
RIBs
REX/HIE-ISOLDE
2001/2015
East Area
TT10
n_TOF
2001
TT2
PS
1959 (628 m)
LINAC 4
2020
LINAC 3
Ions
1994
LEIR
2005 (78 m)
CLEAR
2017

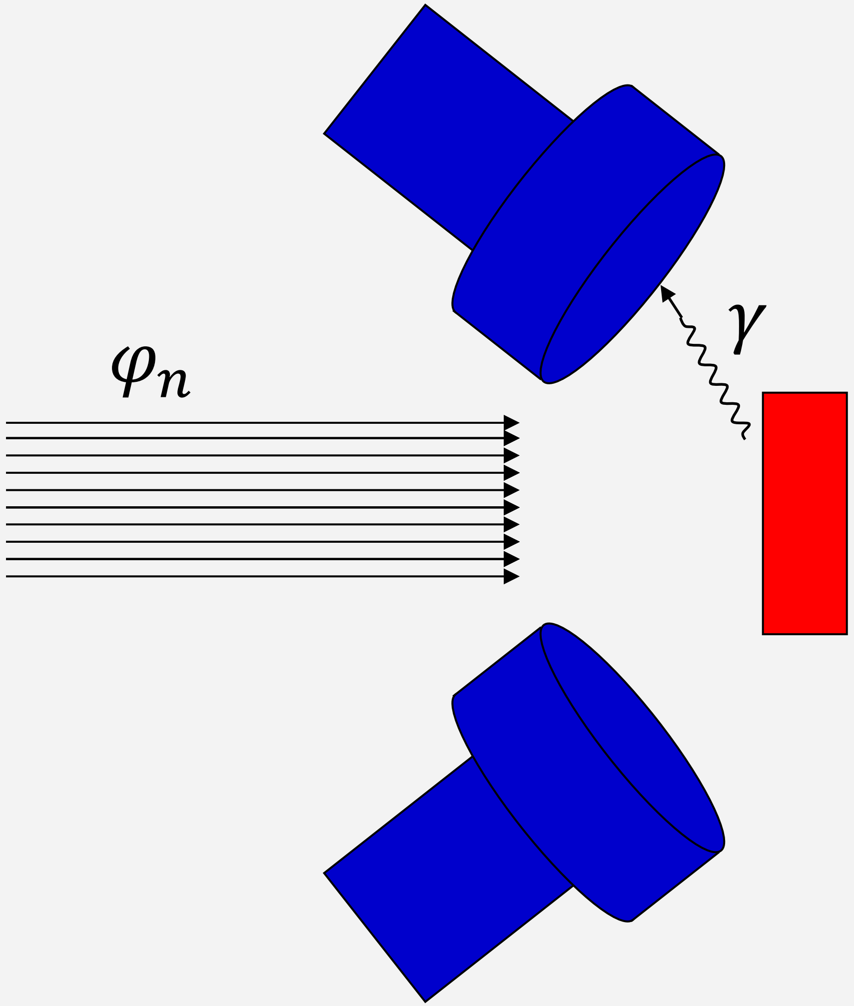
n_TOF



- Located at CERN;
- Neutron beam produced using PS proton on lead target;
- Production of neutrons via spallation;
- Pulsed neutron source (10 meV < E < 1 GeV);
- Three experimental areas (EAR1, EAR2 and NEAR).



Radiative capture



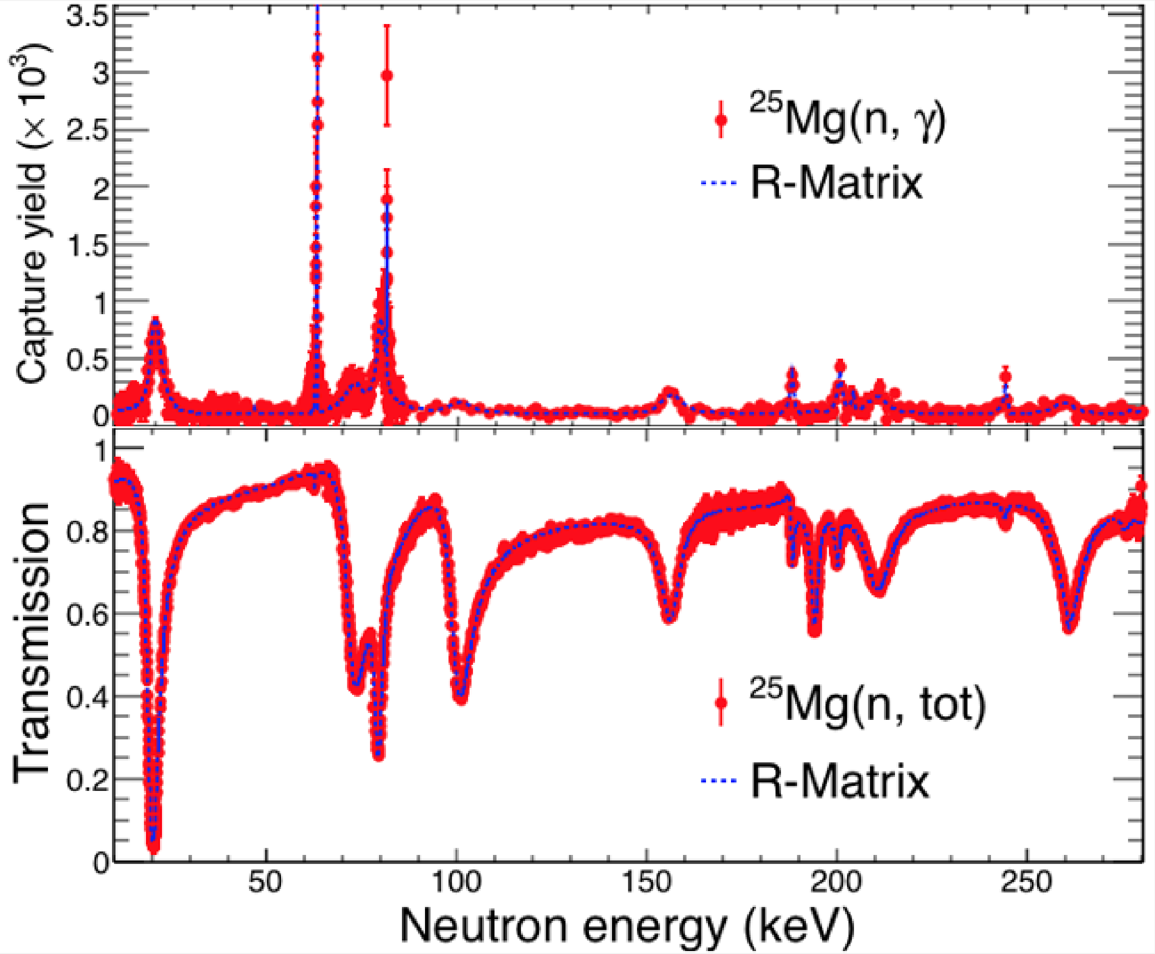
Experimental observable is capture yield

- Percentage of neutrons that undergoes capture reaction in the sample
- Related to capture cross section via:

$$Y_{exp} = N \frac{C_\gamma(t) - B_\gamma(t)}{C_\varphi(t) - B_\varphi(t)} Y_\varphi = (1 - T) \frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}}$$

- Normalization factor energy and nuclide independent, obtained with Au measurement
- Background obtained with additional measurement (empty, lead)

Resonance Shape Analysis



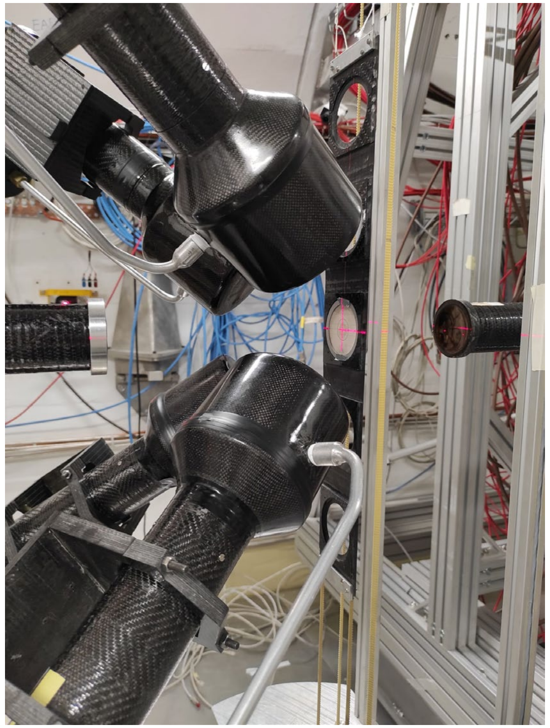
- Determination of the resonance parameter E_0, Γ_γ, Γ_n
- Simultaneous fit of transmission and capture data
- Fit performed using theoretical parametrization

Parametrization of cross section using resonance parameters

C. Massimi et al., Phys. Rev. C 85, 044615 (2012)

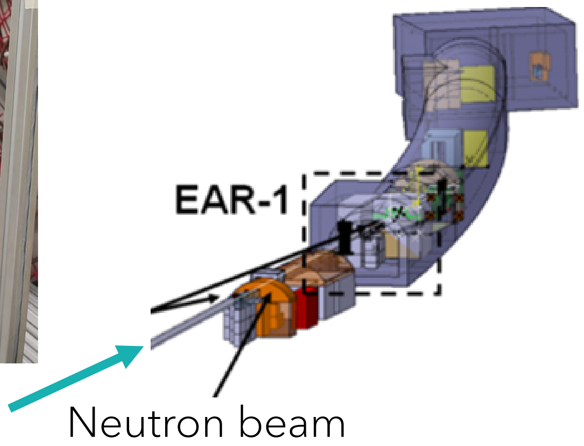
n_TOF capture measurements setups

EAR1

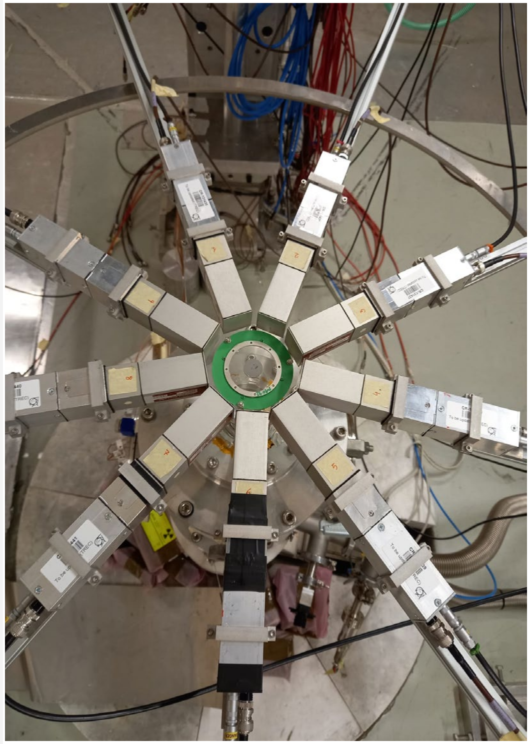


Setup:

- 4 C6D6

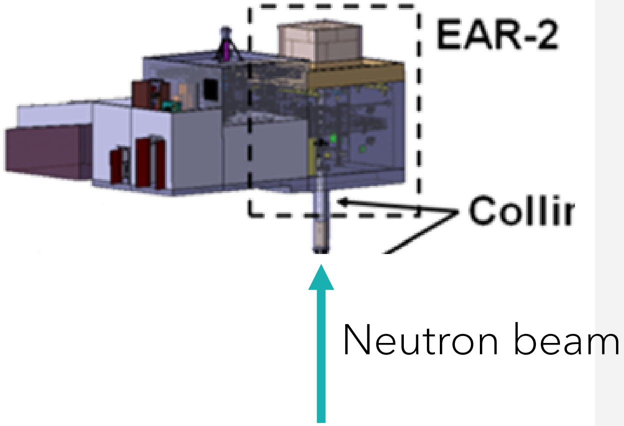


EAR2

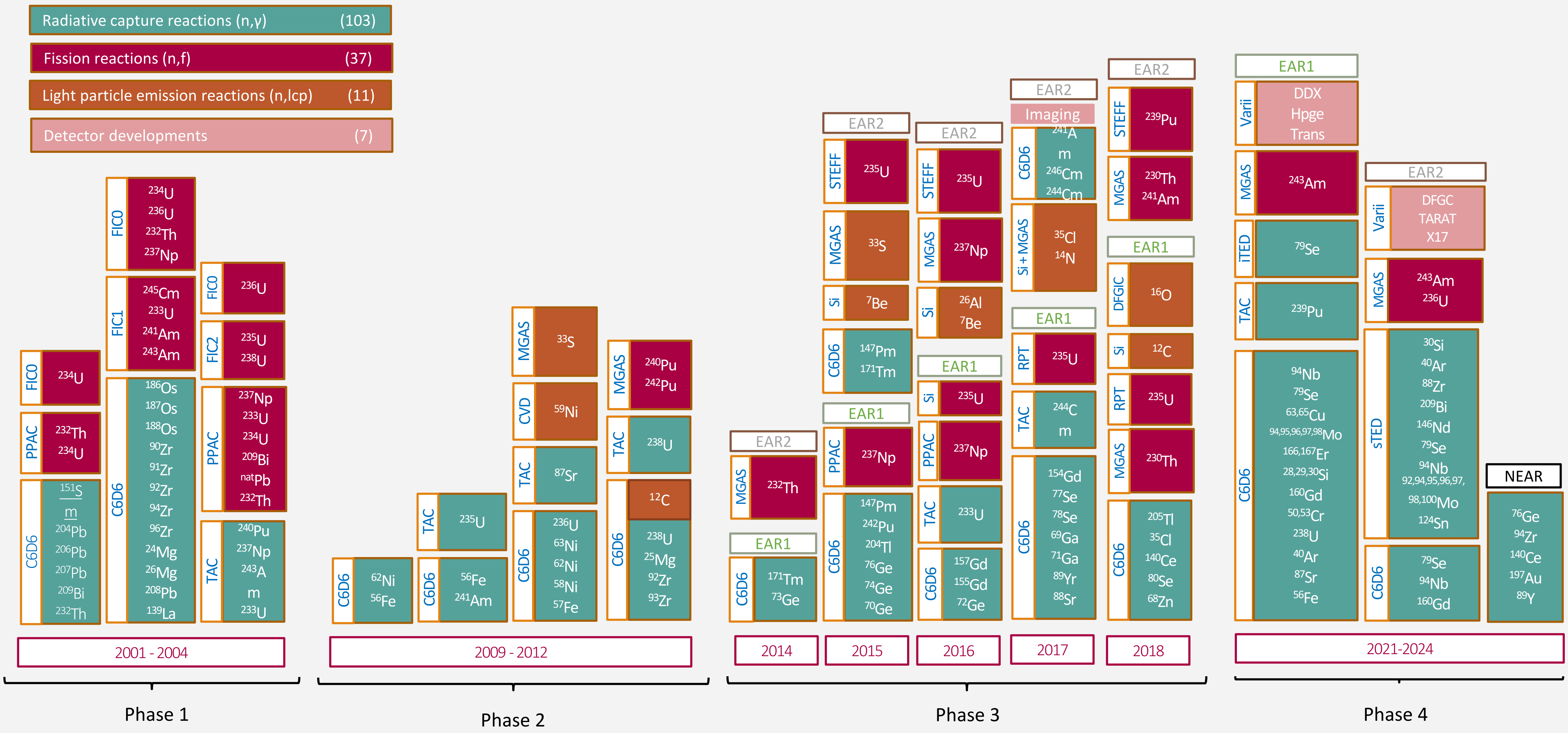


Setup:

- 8 sTED,
- 2 C6D6,



Done at n_TOF

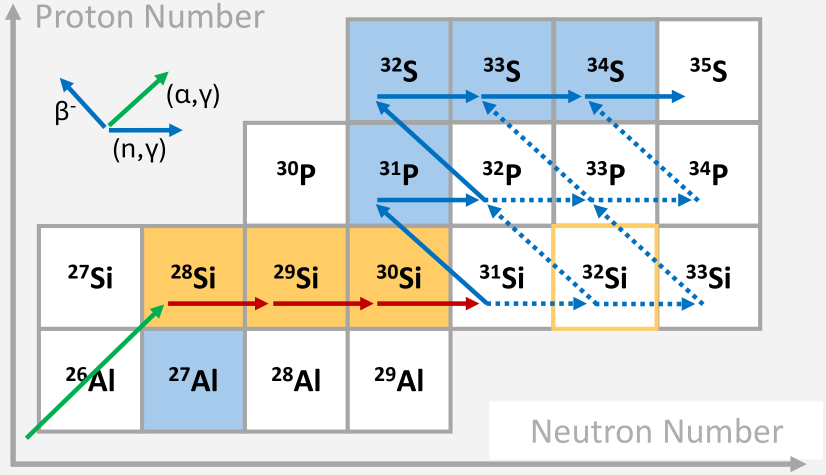


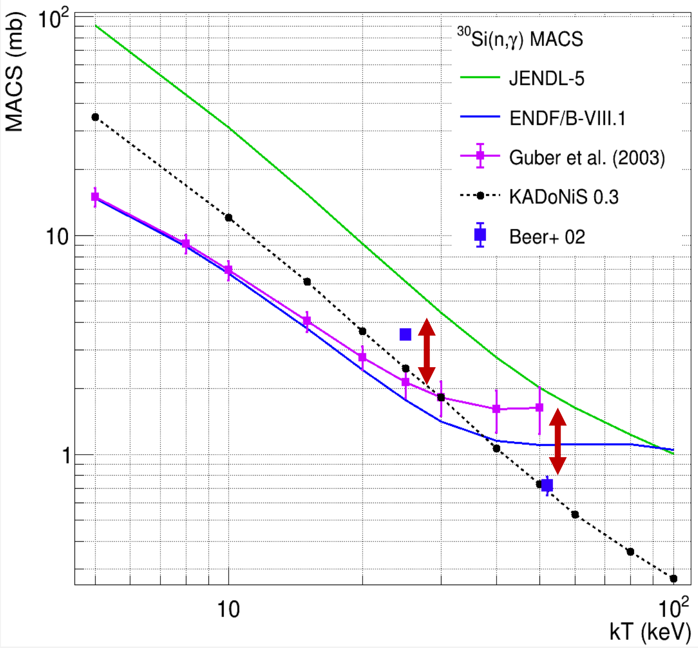
Radiative capture reactions (n,γ) (103)
Fission reactions (n,f) (37)
Light particle emission reactions (n,lcp) (11)
Detector developments (7)
FIC0 ^{234}U ^{236}U ^{232}Th ^{237}Np
FIC1 ^{245}Cm ^{233}U ^{241}Am ^{243}Am
FIC0 ^{234}U
PPAC ^{232}Th ^{234}U
C6D6 ^{151}Sm ^{204}Pb ^{206}Pb ^{207}Pb ^{209}Bi ^{232}Th
C6D6 ^{186}Os ^{187}Os ^{188}Os ^{90}Zr ^{91}Zr ^{92}Zr ^{94}Zr ^{96}Zr ^{24}Mg ^{26}Mg ^{208}Pb ^{139}La
FIC0 ^{236}U
FIC2 ^{235}U ^{238}U
PPAC ^{237}Np ^{233}U ^{234}U ^{209}Bi ^{nat}Pb ^{232}Th
TAC ^{240}Pu ^{237}Np ^{243}Am ^{233}U
2001 - 2004
Phase 1
MGAS ^{33}S
CVD ^{59}Ni
TAC ^{87}Sr
C6D6 ^{236}U ^{63}Ni ^{62}Ni ^{58}Ni ^{57}Fe
TAC ^{235}U
C6D6 ^{56}Fe ^{241}Am
C6D6 ^{62}Ni ^{56}Fe
MGAS ^{240}Pu ^{242}Pu
TAC ^{238}U
C6D6 ^{12}C ^{238}U ^{25}Mg ^{92}Zr ^{93}Zr
2009 - 2012
Phase 2
EAR2
MGAS ^{232}Th
EAR1
C6D6 ^{171}Tm ^{73}Ge
2014
EAR2
STEFF ^{235}U
MGAS ^{33}S
Si ^{7}Be
C6D6 ^{147}Pm ^{171}Tm
EAR1
PPAC ^{237}Np
C6D6 ^{147}Pm ^{242}Pu ^{204}Tl ^{76}Ge ^{74}Ge ^{70}Ge
2015
EAR2
STEFF ^{235}U
MGAS ^{237}Np
Si ^{26}Al ^{7}Be
EAR1
Si ^{235}U
PPAC ^{237}Np
TAC ^{233}U
C6D6 ^{157}Gd ^{155}Gd ^{72}Ge
2016
EAR2
Imaging
C6D6 ^{241}Am ^{246}Cm ^{244}Cm
Si + MGAS ^{35}Cl ^{14}N
EAR1
RPT ^{235}U
TAC ^{244}Cm
C6D6 ^{154}Gd ^{77}Se ^{78}Se ^{69}Ga ^{71}Ga ^{89}Yr ^{88}Sr
2017
EAR2
STEFF ^{239}Pu
MGAS ^{230}Th ^{241}Am
EAR1
DFGIC ^{16}O
Si ^{12}C
RPT ^{235}U
MGAS ^{230}Th
C6D6 ^{205}Tl ^{35}Cl ^{140}Ce ^{80}Se ^{68}Zn
2018
Phase 3
EAR1
Varii DDX Hpge Trans
MGAS ^{243}Am
iTED ^{79}Se
TAC ^{239}Pu
C6D6 ^{94}Nb ^{79}Se ^{63,65}Cu ^{94,95,96,97,98}Mo ^{166,167}Er ^{28,29,30}Si ^{160}Gd ^{50,53}Cr ^{238}U ^{40}Ar ^{87}Sr ^{56}Fe
EAR2
Varii DFGC TARAT X17
MGAS ^{243}Am ^{236}U
sTED ^{30}Si ^{40}Ar ^{88}Zr ^{209}Bi ^{146}Nd ^{79}Se ^{94}Nb ^{92,94,95,96,97,98,100}Mo ^{124}Sn
C6D6 ^{79}Se ^{94}Nb ^{160}Gd
NEAR ^{76}Ge ^{94}Zr ^{140}Ce ^{197}Au ^{89}Y
2021-2024
Phase 4

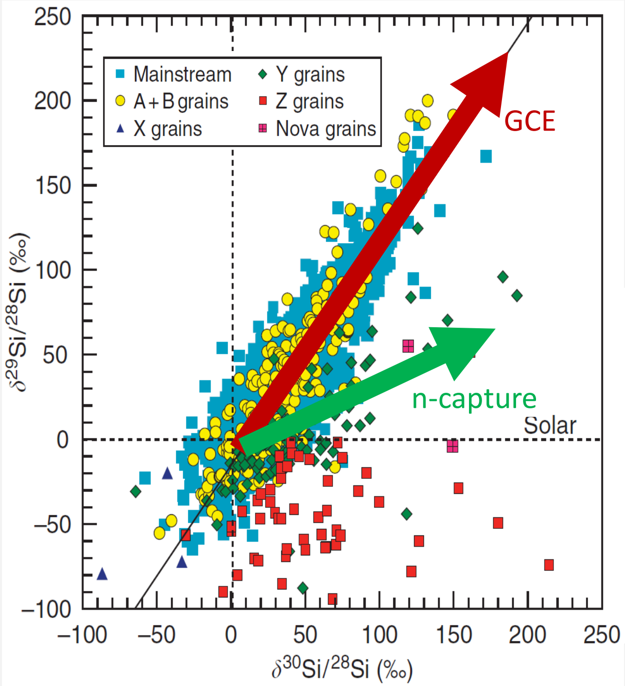
Latest measurement: a review

$^{30}Si(n,\gamma)$

- Crucial for modeling nucleosynthesis of Si (P, S);
- Understanding presolar SiC composition, mixing of GCE and n-capture;
- Produced in convective carbon-shell of massive star;
- Inconsistencies in literature data.



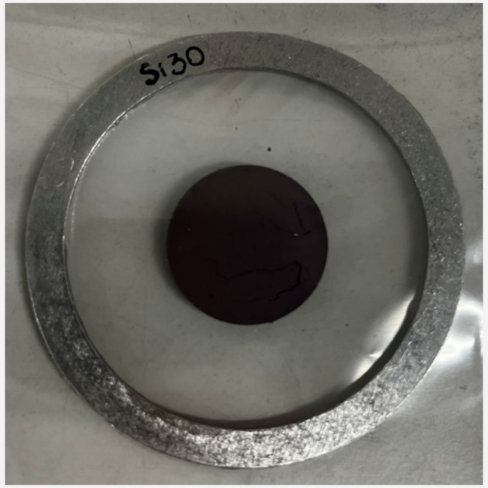


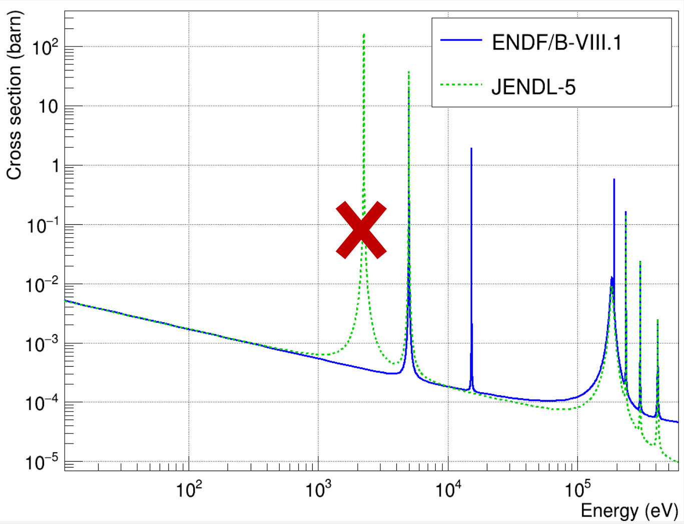


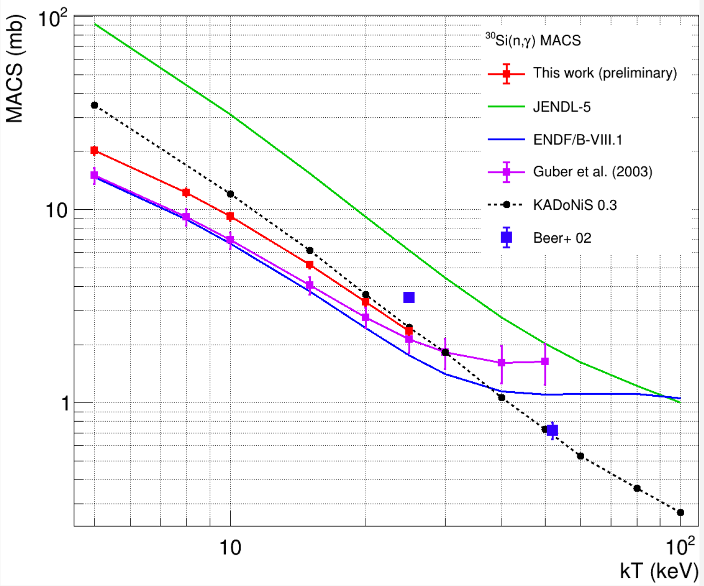
Courtesy: M. Spelta

$^{30}Si(n,\gamma)$

- First isotopically enriches sample used in capture!
- Absence of big resonance at 14 keV included in evaluations;
- Larger kernel than in previous measurements;
- Preliminary value of MACS intermediate between previous time-of-flight and activation measurements.



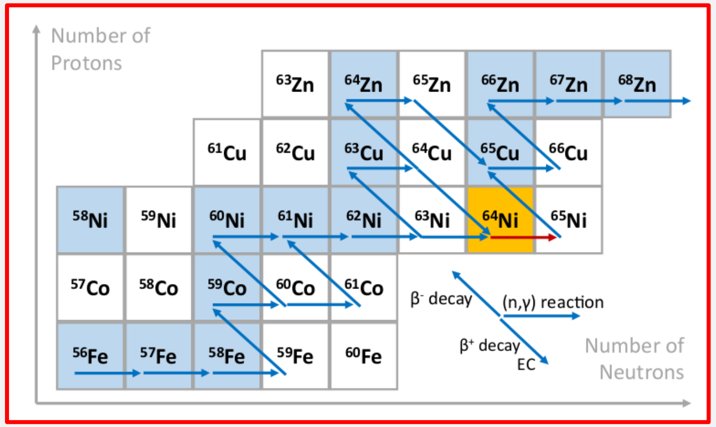


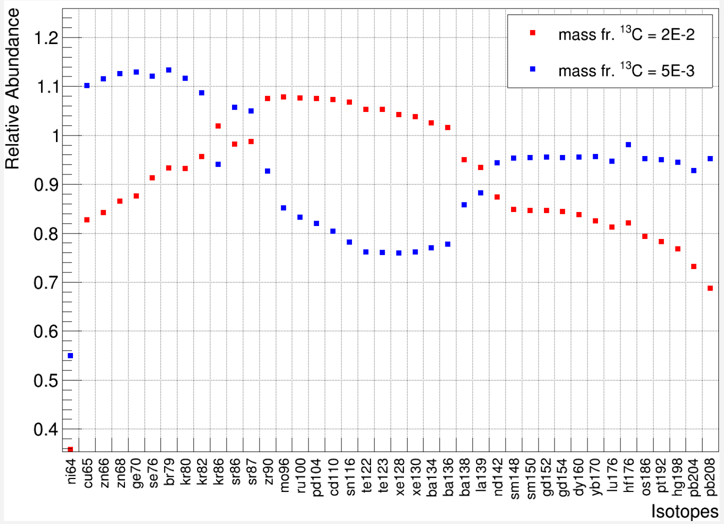


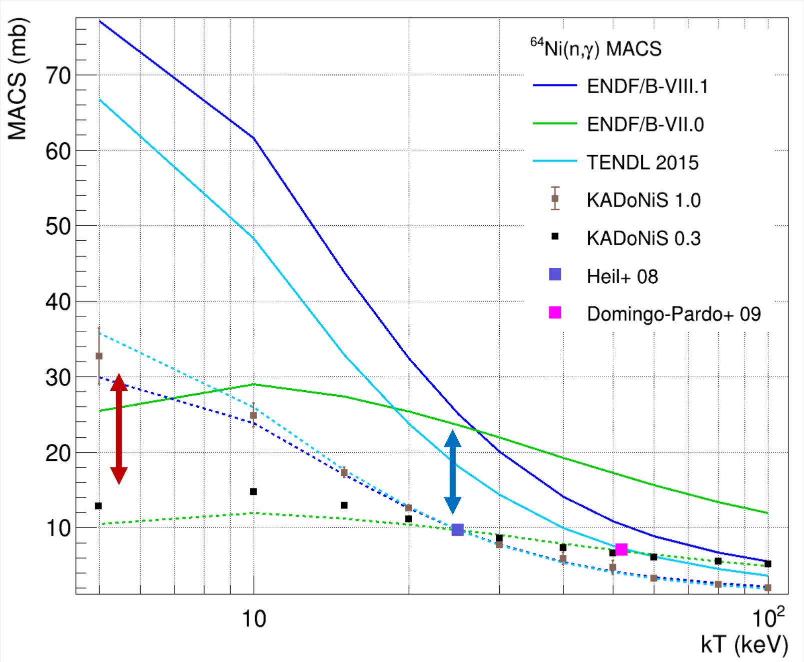
Courtesy: M. Spelta

^{64}Ni(n,γ)

- Seed of the s-process;
- The MACS has an effect on heavier isotope production in main s-process;
- Discrepancies in MACS from activation and libraires;
- Inconsistencies in date in literature.



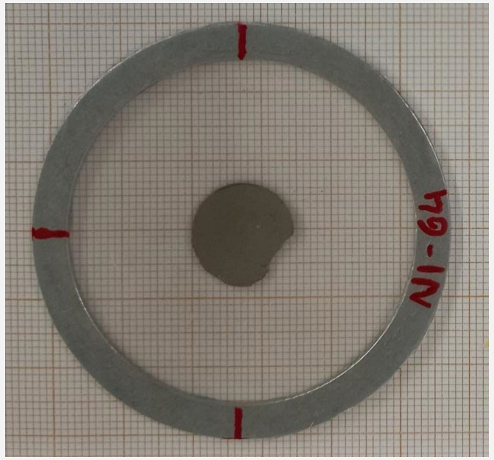


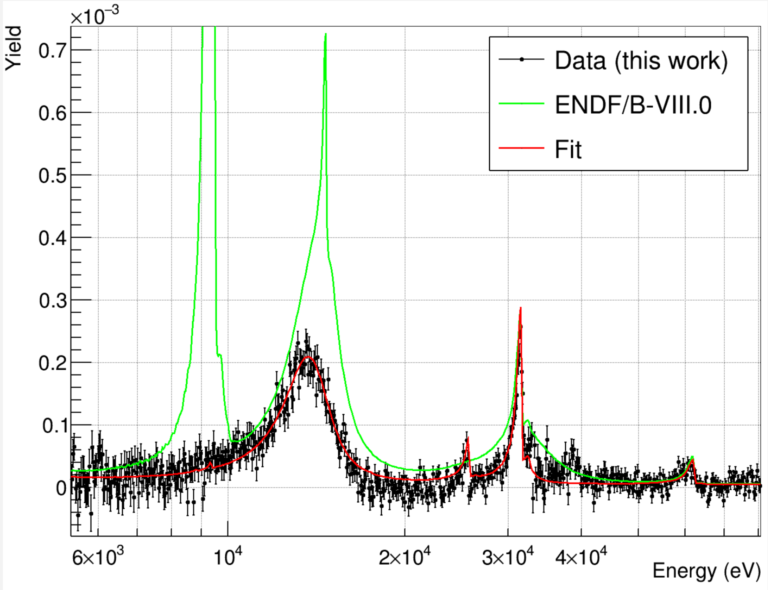


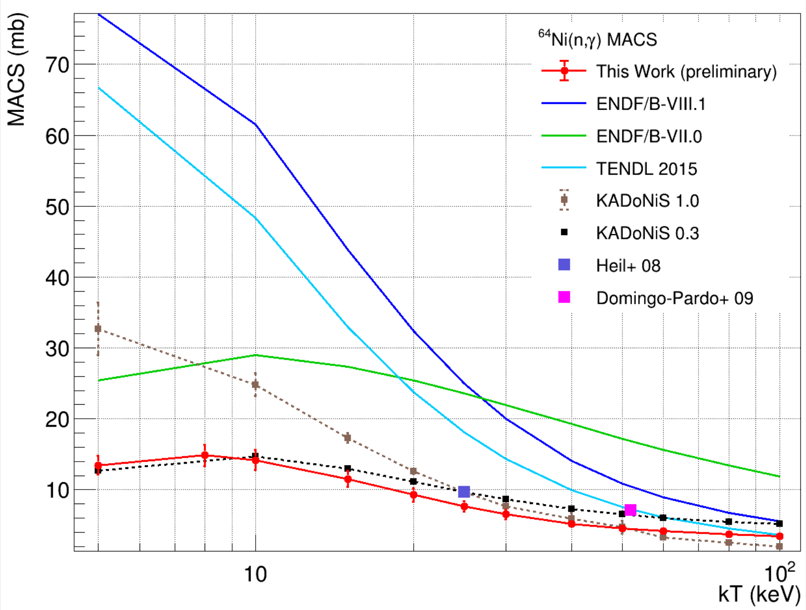
Courtesy: M. Spelta

$^{64}Ni(n,\gamma)$

- Sample of only 427mg;
- Clear absence of 9,5 keV and 14,8 keV resonances reported in libraries;
- Reduction of MACS, better agreement with activation data!







Courtesy: M. Spelta

^{63,65}Cu(n,γ)...

- Produced in explosive environments like Type Ia supernovae;
- Accurate knowledge of MACS important for nucleosynthesis in massive stars (weak s-process);
- Additional application for nuclear technologies (research reactor TAPIRO and fusion)

^{62}Ga 116.00 ms β+	^{63}Ga 32.40 s β+	^{64}Ga 2.63 m β+	^{65}Ga 15.20 m β+	^{66}Ga 9.49 h β+	^{67}Ga 3.26 d β+	^{68}Ga 1.13 h β+
^{61}Zn 1.48 m β+	^{62}Zn 9.19 h β+	^{63}Zn 38.47 m β+	^{64}Zn 48.63 59 mb	^{65}Zn 243.63 d 162 mb, β+	^{66}Zn 27.9 35 mb	^{67}Zn 4.1 153 mb
^{60}Cu 23.70 m β+	^{61}Cu 3.33 h β+	^{62}Cu 9.67 m β+	^{63}Cu 69.17 94 mb	^{64}Cu 12.70 h β+	^{65}Cu 30.83 41 mb	^{66}Cu 5.12 m β-
^{59}Ni 75.99 ka 87 mb, β+	^{60}Ni 26.223 30 mb	^{61}Ni 1.14 82 mb	^{62}Ni 3.634 22.3 mb	^{63}Ni 100.11 a 31 mb, β-	^{64}Ni 0.926 8.7 mb	^{65}Ni 2.52 h β-
^{58}Co 70.86 d β+	^{59}Co 100 38 mb	^{60}Co 5.27 a β-	^{61}Co 1.65 h β-	^{62}Co 1.50 m β-	^{63}Co 27.40 s β-	^{64}Co 300.00 ms β-
^{57}Fe 2.119 40 mb	^{58}Fe 0.282 12.1 mb	^{59}Fe 44.50 d β-	^{60}Fe 1.50 Ma β-	^{61}Fe 5.98 m β-	^{62}Fe 1.13 m β-	^{63}Fe 6.01 s β-

Recommended MACS30 (Maxwellian Averaged Cross Section @ 30keV)

^{63}Cu (n,γ) ^{64}Cu

Total MACS at 30keV: 60.1 ± 6.2 mb

Cross sections do not include stellar enhancement factors!

History

Version	Total MACS [mb]	Partial to gs [mb]	Partial to isomer [mb]
1.0	60.1 ± 6.2	-	-
0.3	55.6 ± 2.2	-	-
0.0	94 ± 10	-	-

(Version 0.0 corresponds to Bao et al.)

Comment

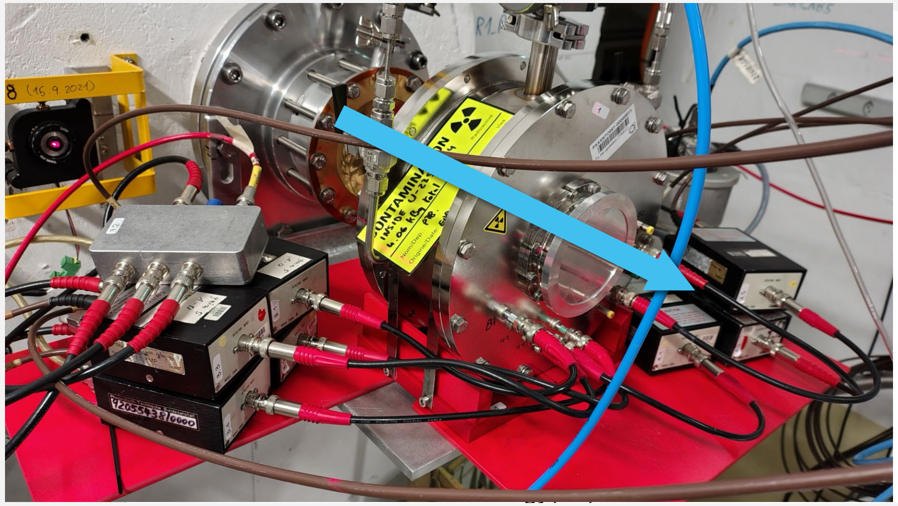
New rec. value is from HKU08, renormalized by 632 mb/586 mb = 1.0785, and recalculated with normalized energy dependencies of tendl15,endfb71,jendl40. Uncertainty is the deviation between different evaluations plus 4% exp. uncertainty from HKU08. **Note the large deviation between the activation measurement and the TOF measurements. More investigation needed!**
Last review: April 2017

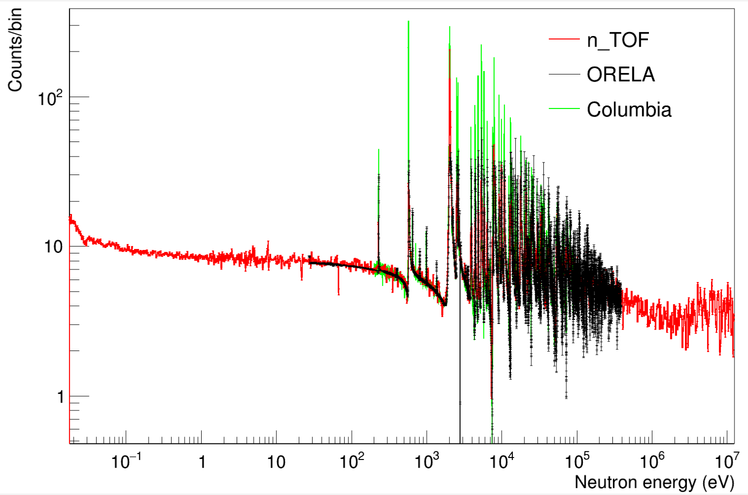
Courtesy: N. Pieretti

^{63,65}Cu(n,γ)+(n,tot)

- Capture measurement performed in 2024 in EAR1 with enriched samples;
- First transmission campaign at n_TOF!
- Total cross section compatible with previous experiment, but possible to extend range up to several MeV!

Experiment	Sample	Protons	Comments
Capture	^{63}Cu	2.0×10^{18}	
Capture	^{65}Cu	2.0×10^{18}	
Capture	^{nat}Cu	0.3×10^{18}	EAR1 or EAR2
Capture	Empty-sample	0.2×10^{18}	background study
Capture	Pb	0.2×10^{18}	background study
Capture	C	0.2×10^{18}	background study
Capture	^{197}Au	0.1×10^{18}	normalization
Transmission	^{63}Cu	1.0×10^{18}	"Sample-in"
Transmission	^{65}Cu	1.0×10^{18}	"Sample-in"
Transmission	Empty-sample	1.0×10^{18}	"Sample-out"
		8.0×10^{18}	

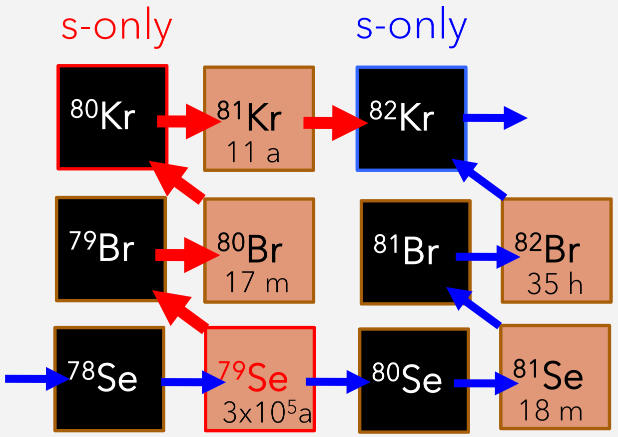


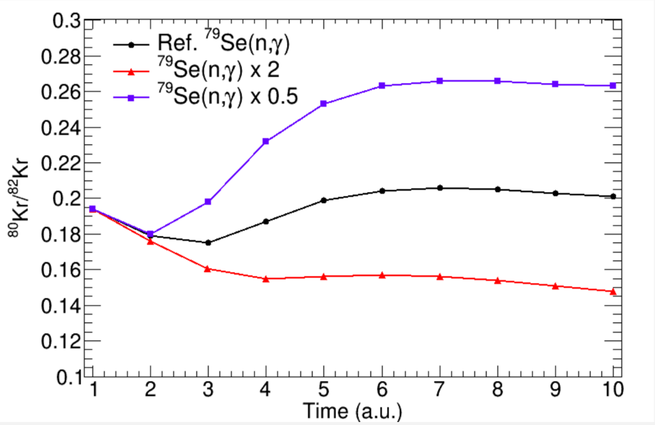


Courtesy: N. Pieretti

$^{79}Se(n,\gamma)$

- Branching point with direct impact on s-only isotopes $^{80,82}Kr$;
- MACS only known from average parameters;
- One of the **21 key s-nuclei** listed in: Kaeppeler, *Rev. Mod. Phys* 83, 157 (2011)
- Listed as **1st-level priority** (two times) in the sensitivity study: Cescutti et al., *MNRS* 478 (2018)

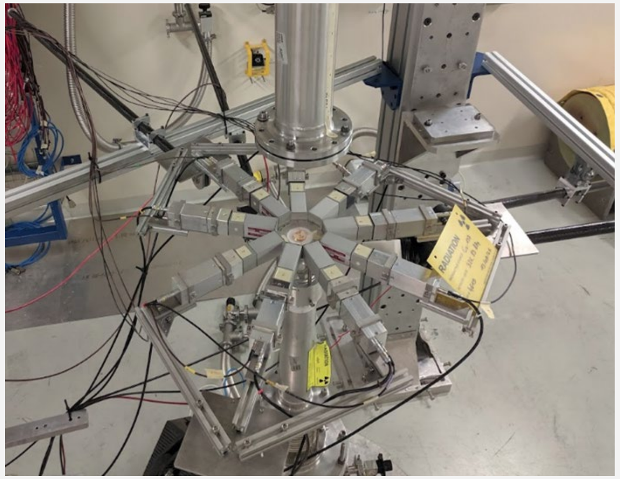


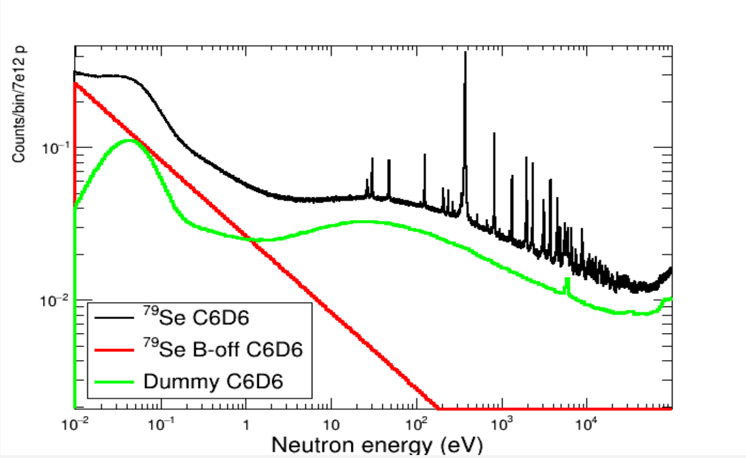


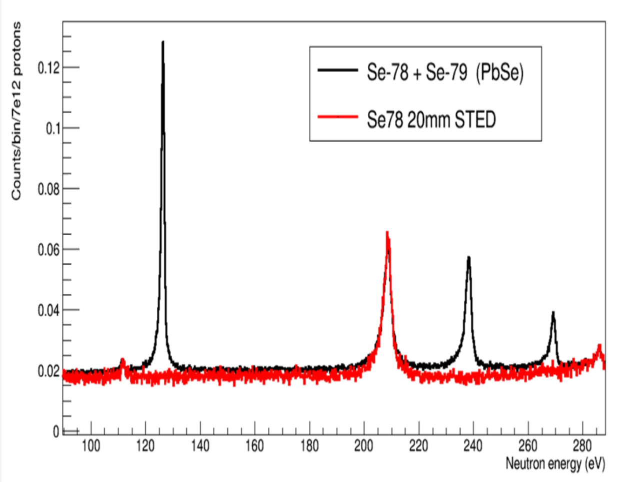
Courtesy: J. Lerendegui

$^{79}Se(n,\gamma)$

- Production of 3mg of ^{79}Se from irradiation of ^{78}Se at ILL;
- Production of Pb-Se at PSI
- 6MBq of activity!
- Measurement at EAR2 show good signal-background ratio
- First ever directly observed resonances of ^{79}Se!





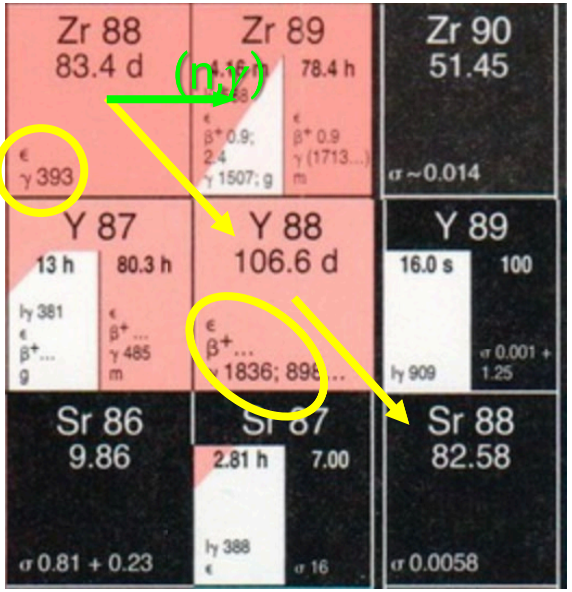


Courtesy: J. Lerendegui

$^{88}Zr(n,\gamma)$

- Very high cross section (800kb) observed at previous transmission measurement at LANL;
- Highly radioactive isotope (83d half life);
- Material produced at Los Alamos $^{89}Y(p,2n)$ and prepared at PSI;
- Very low mass sample!

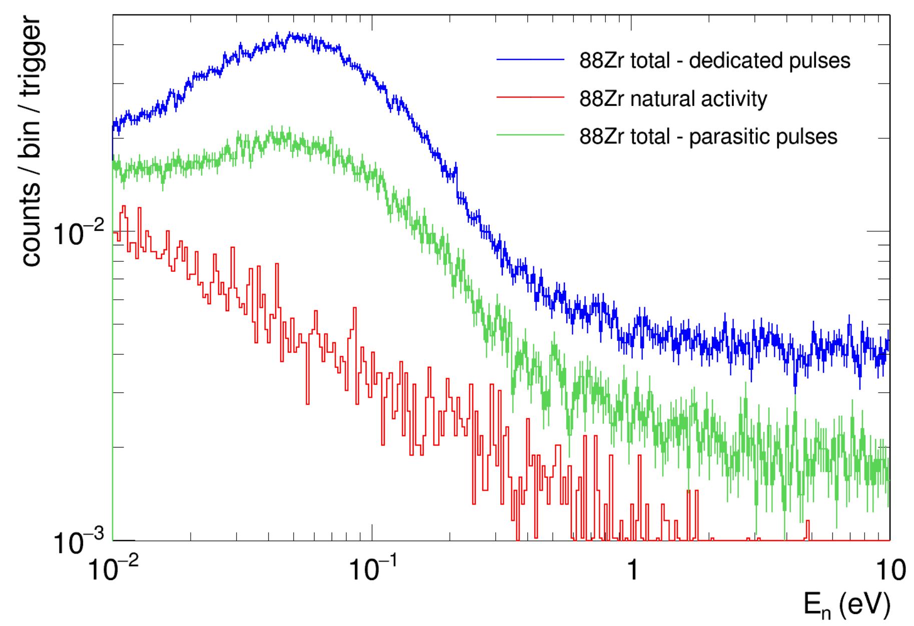
Atoms : 1.15e16
Mass : 1.68 μg
Activity : 1.1 GBq

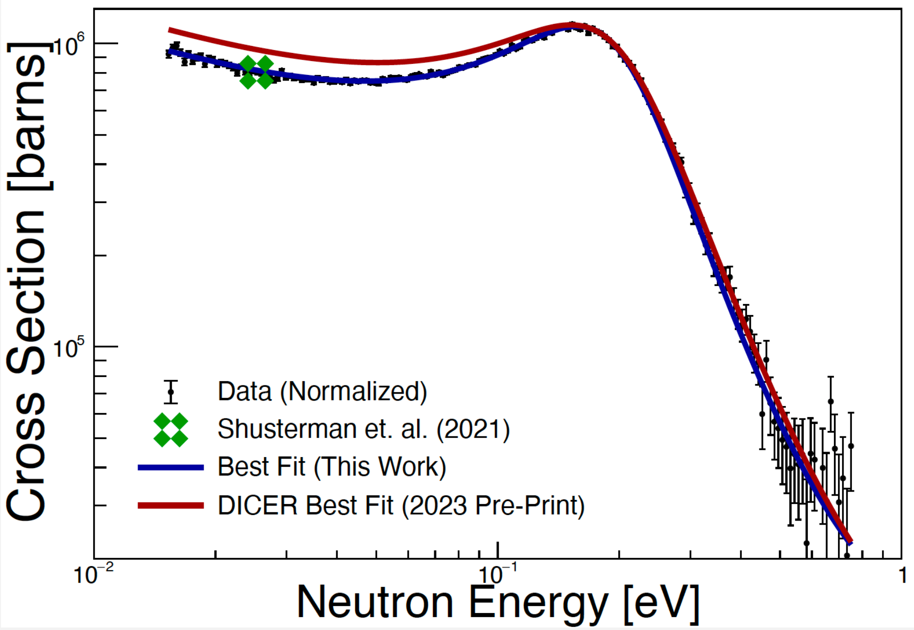


Courtesy: W. Flanagan

$^{88}Zr(n,\gamma)$

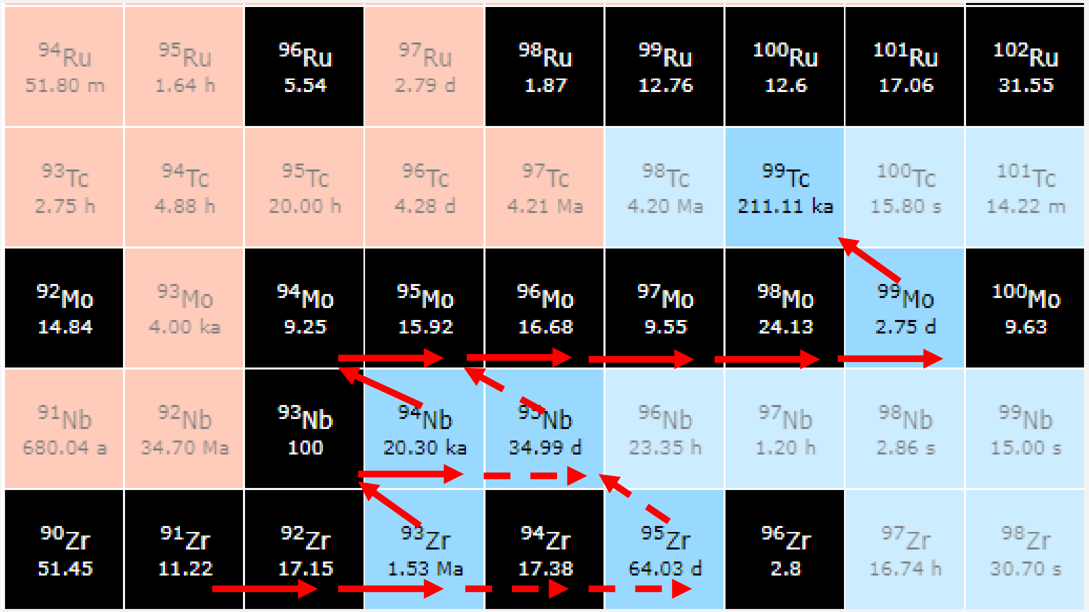
- Radioactivity from the sample (^{88}Zr + ^{88}Y from decay) not overwhelming;
- Signals from capture events clearly seen;
- Preliminary estimate of cross section shows discrepancy of resonance shape respect to DICER.



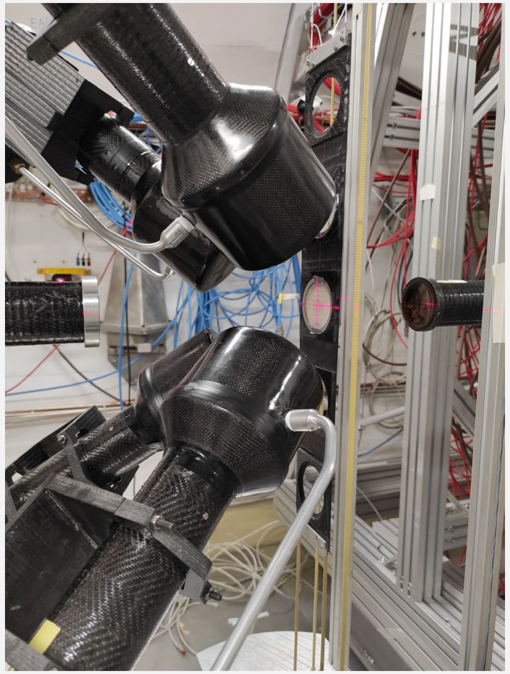


Courtesy: W. Flanagan

Mo(n,γ)

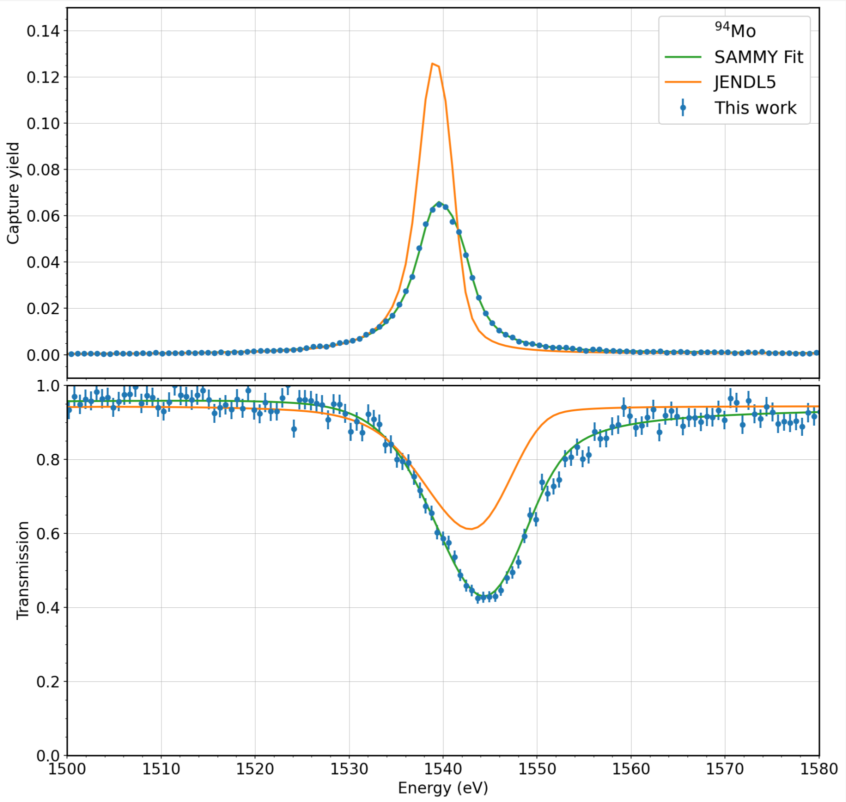


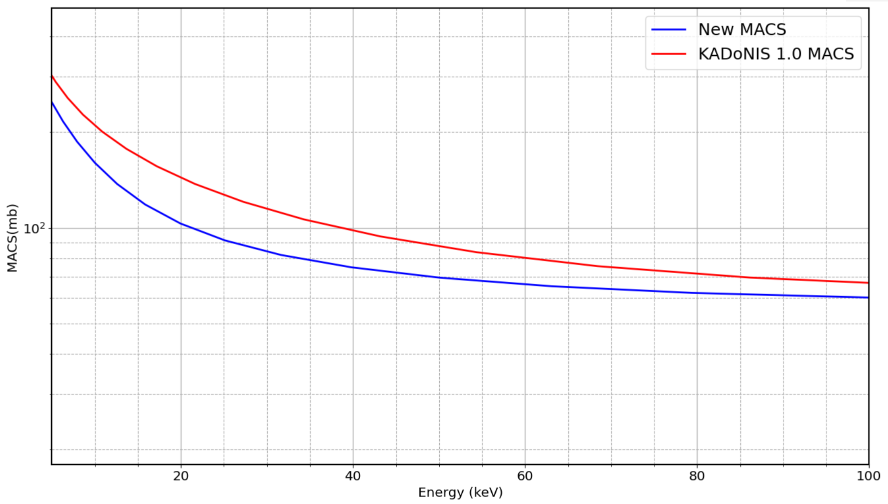
- Discrepancies in SiC grain composition for different isotopes;
- **All stable isotopes** measured at n_TOF with additional transmission @ GELINA for ^{94,95,96}Mo;
- Measurements at both areas to cover energy region from thermal up to hundreds of keV;
- Samples prepared at n_TOF.



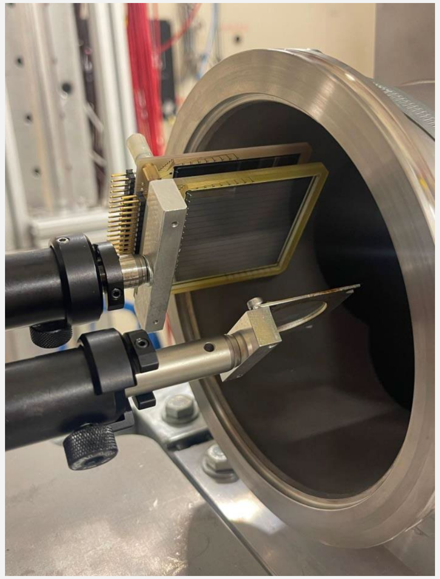
Mo(n,γ)

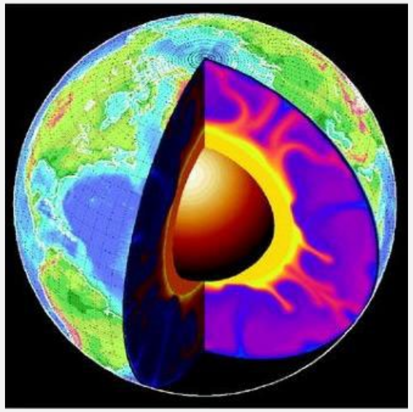
- Discrepancies observed from literature for all isotopes;
- Good agreement between capture and transmission data;
- Reduction of MACS observed for ^{94}Mo.

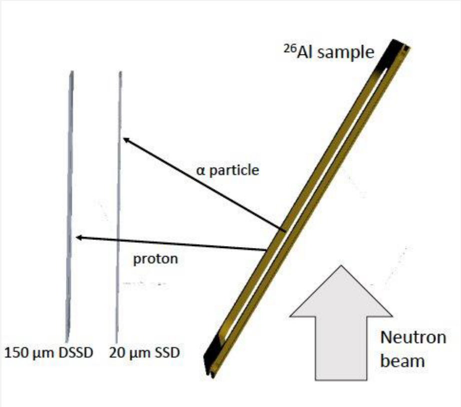




...More than just (n,γ)







^{40}K(n,cp)

- Radiogenic heating of young exoplanets;
- Crucial for biosphere and habitability;
- Produced in massive stars core and in s-process;
- Only one direct measurement in literature!

^{41}Ca(n,α)

- Short half lived isotope ($t_{1/2}$~100k years);
- Produced in massive stars and ejected in ESS material from explosions or winds;
- Produced in He and C burning phases;
- ^{41}Ca(n,α) main destruction channel.

Conclusions

- Latest updates on the facility, detection systems and sample production enabled previously very challenging measurements at n_TOF (^{79}Se, ^{88}Zr, ^{64}Ni);

- Many neutron capture cross section are still known with accuracy far from the 5% target, the use on state-of-the-art facility and detector setups can help reaching very low levbel of uncertainty (Mo, ^{30}Si,$^{63,65}Cu$);

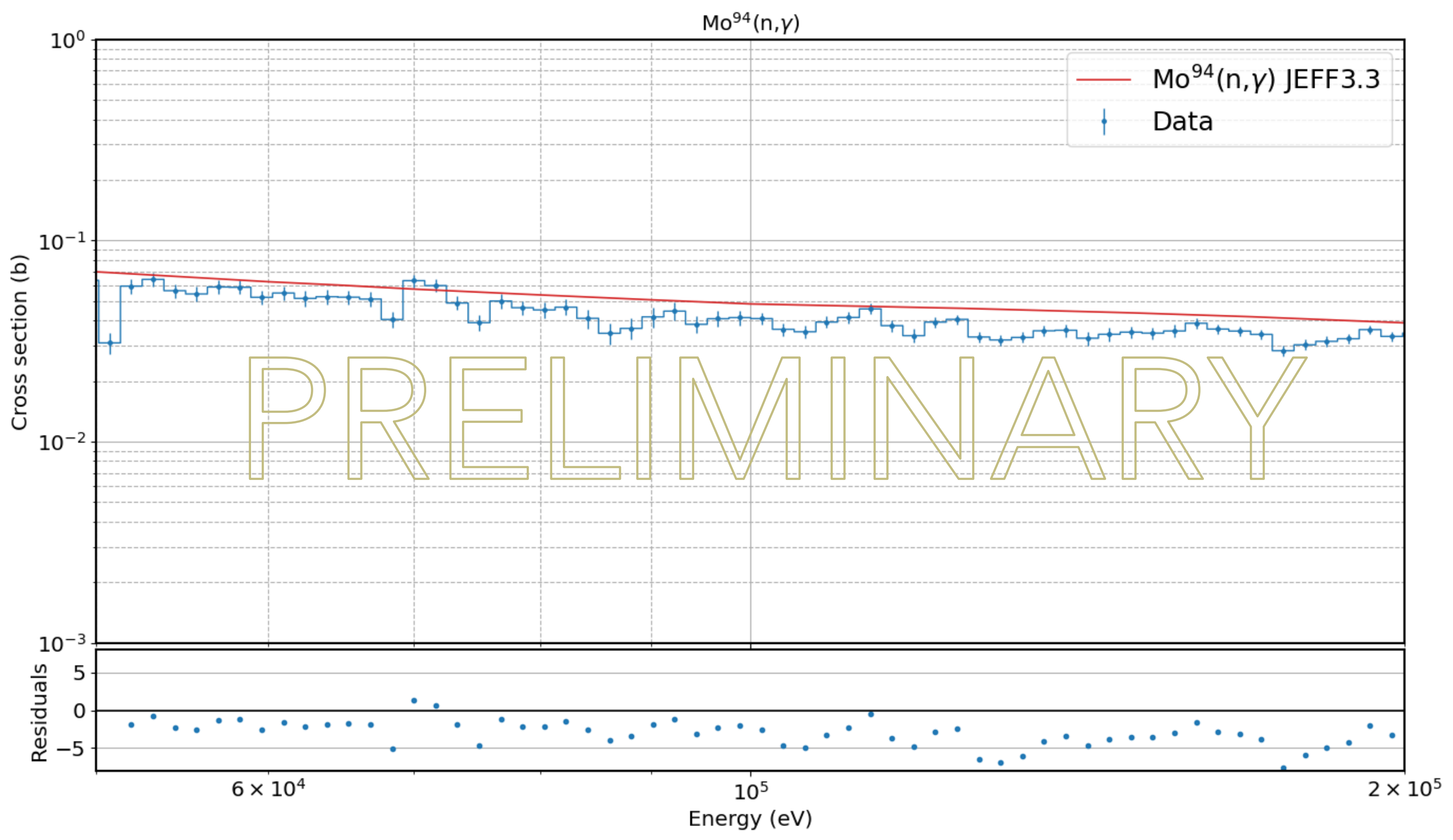
- Combination with activation measurements, like the one performed at NEAR, can further constrain the MACS;

- Additional reaction channels crucial for astrophysics are also being explored at n_TOF for radioactive samples, exploiting the high flux of EAR2;

Thank you for your attention!

Backup

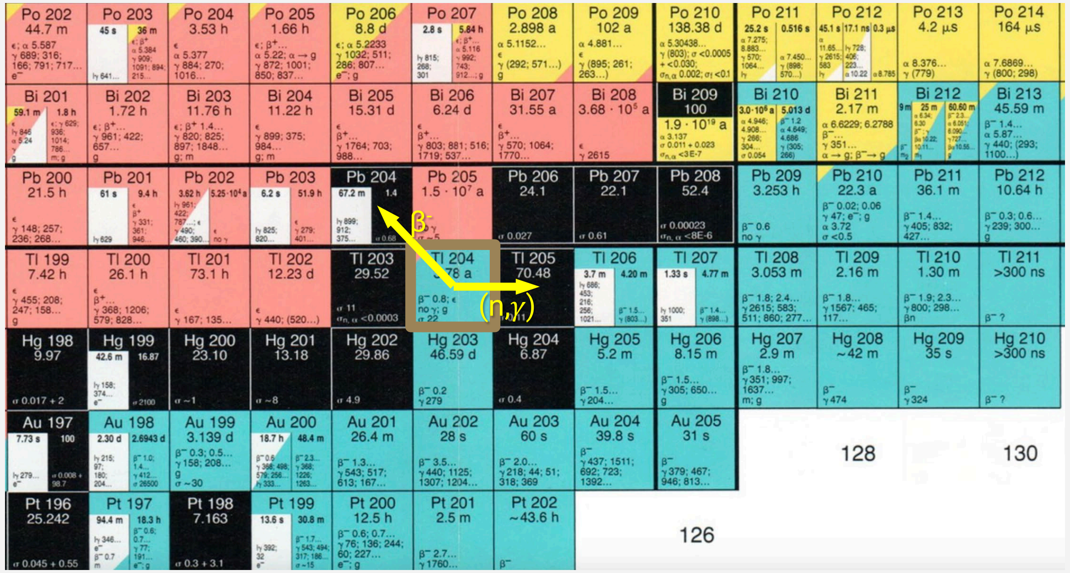
^{94}Mo Unresolved Resonance Region

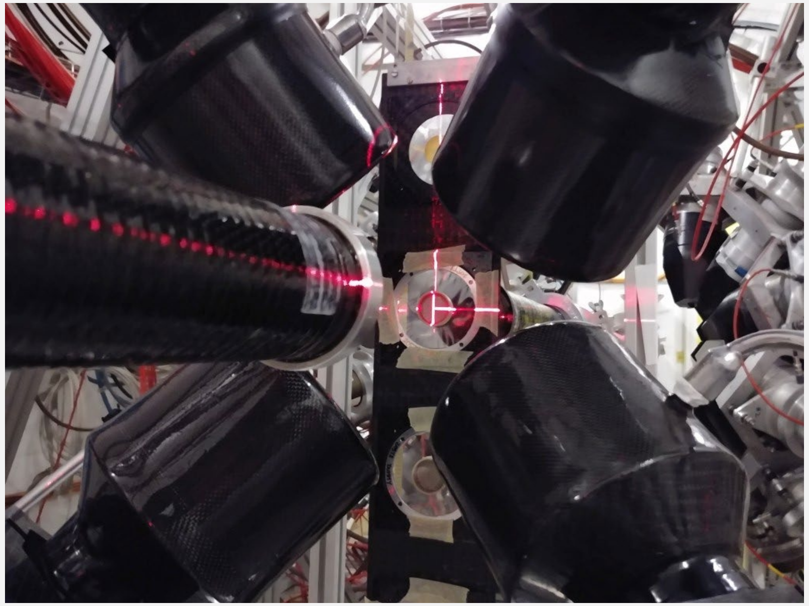


- Calculation of the preliminary cross section in the URR (>75keV);
- Comparison with JEFF 3.3 cross section data;
- This comparison shows a reduction of 10-20% in the cross section of ^{94}Mo.

^{204}Tl(n,γ)

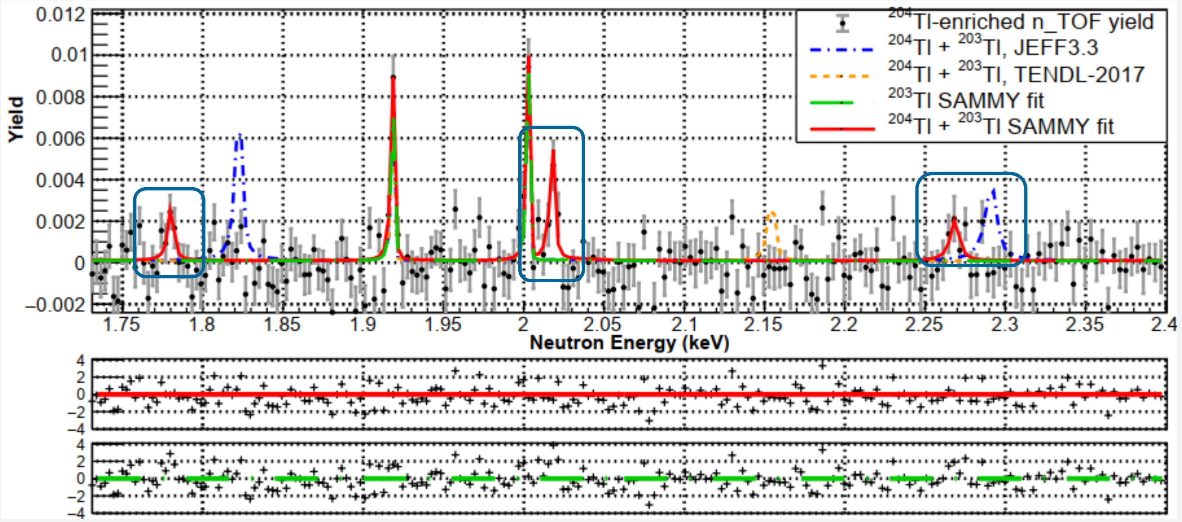
- Branching point crucial for production of ^{204}Pb;
- Sample of ^{203}Tl produced at PSI, successively irradiated at ILL: 9mg of ^{204}Tl;
- 180 GBq of activity!

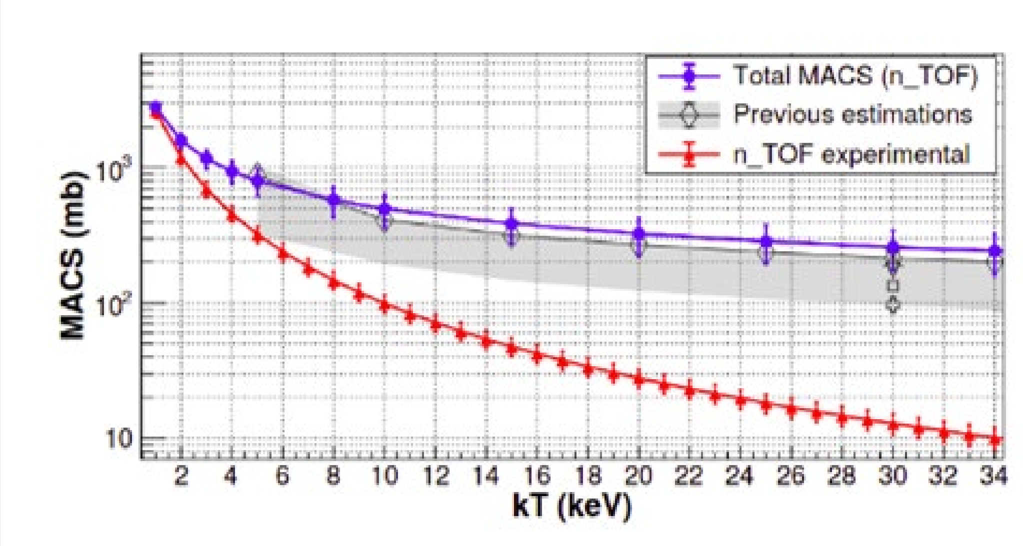




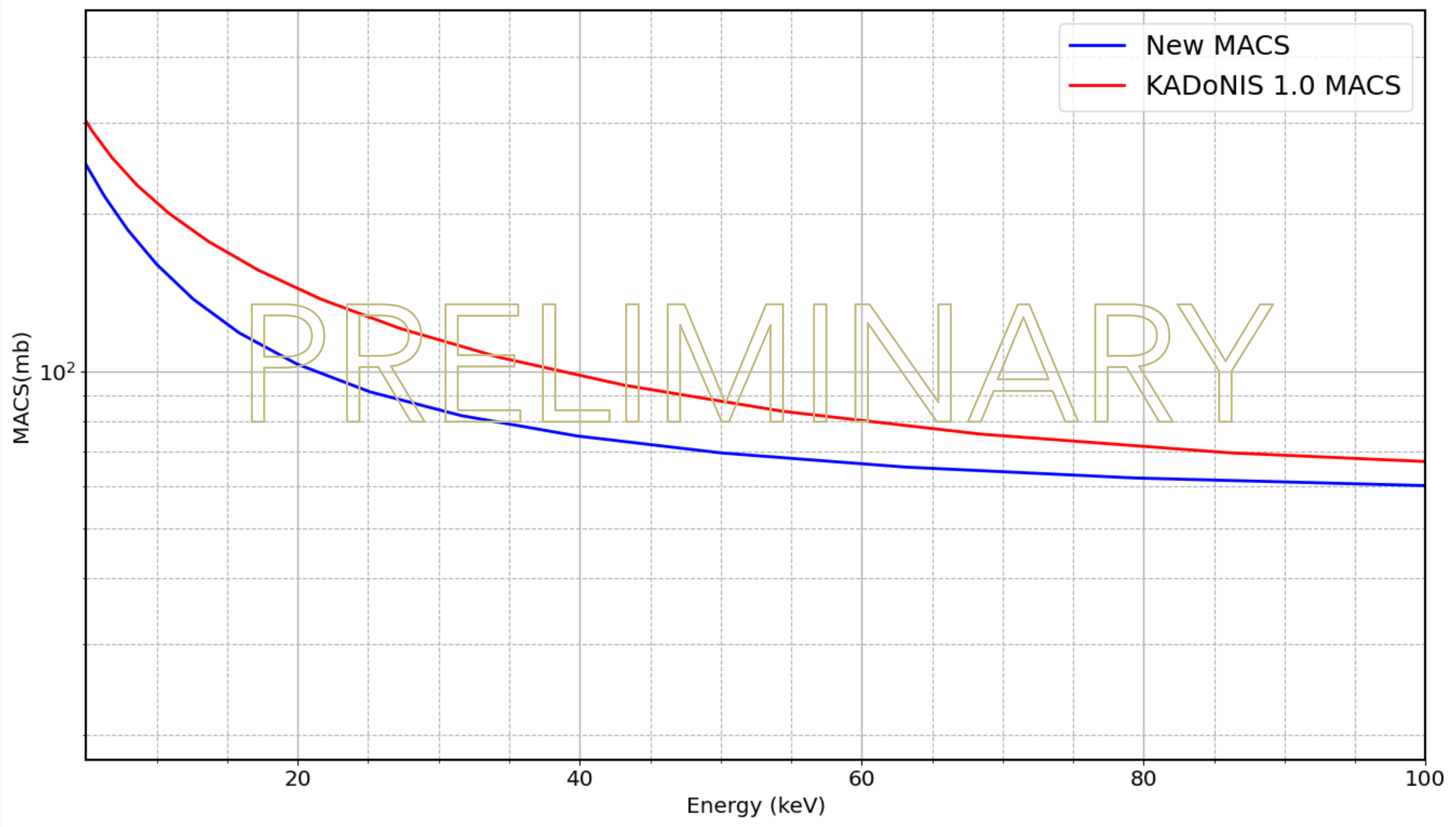
^{204}Tl(n,γ)

- First measurement of ^{204}Tl(n,γ) cross section at n_TOF!
- Uncertainty on the *s*-process abundance of ^{204}Pb has been reduced from ~30% down to +8%/−6%
- Calculations are in agreement with the latest solar system abundance of ^{204}Pb reported by K. Lodders in 2021.





Preliminary MACS of ^{94}Mo



- Preliminary values of the Maxwellian Averaged Cross Section (MACS) have been evaluated for ^{94}Mo,
- The new values of the MACS show a reduction between 10% and 30%.

Experimental measurements

Transmission

Percentage of neutrons that traverses a samples without interacting with it

- Related to total cross section:

$$T = \mathrm{N}\frac{C_{in}(t)-KB_{in}(t)}{C_{out}(t)-\mathrm{K}B_{out}(t)} = \frac{\varphi_n e^{-n\sigma_{tot}}}{\varphi_n} = e^{-n\sigma_{tot}}$$

Radiative capture (capture yield)

Percentage of neutrons that undergoes capture reaction in the sample

- Related to capture cross section via:

$$\mathrm{Y}_{exp} = \mathrm{N}\frac{C_\gamma(t) - B_\gamma(t)}{C_\varphi(t) - B_\varphi(t)}\mathrm{Y}_\varphi = (1-T)\frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}}$$

Time-of-flight technique

$$E_n = mc^2(\gamma - 1) \approx \frac{1}{2}mv^2$$

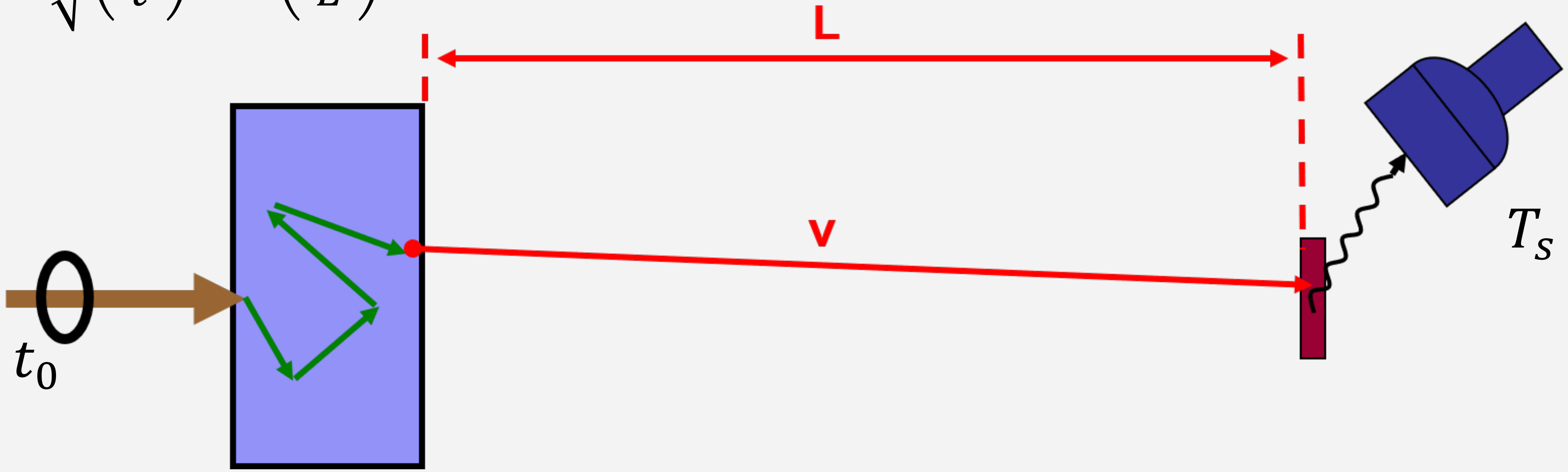
$$v = \frac{L}{t}$$

L → Flight path

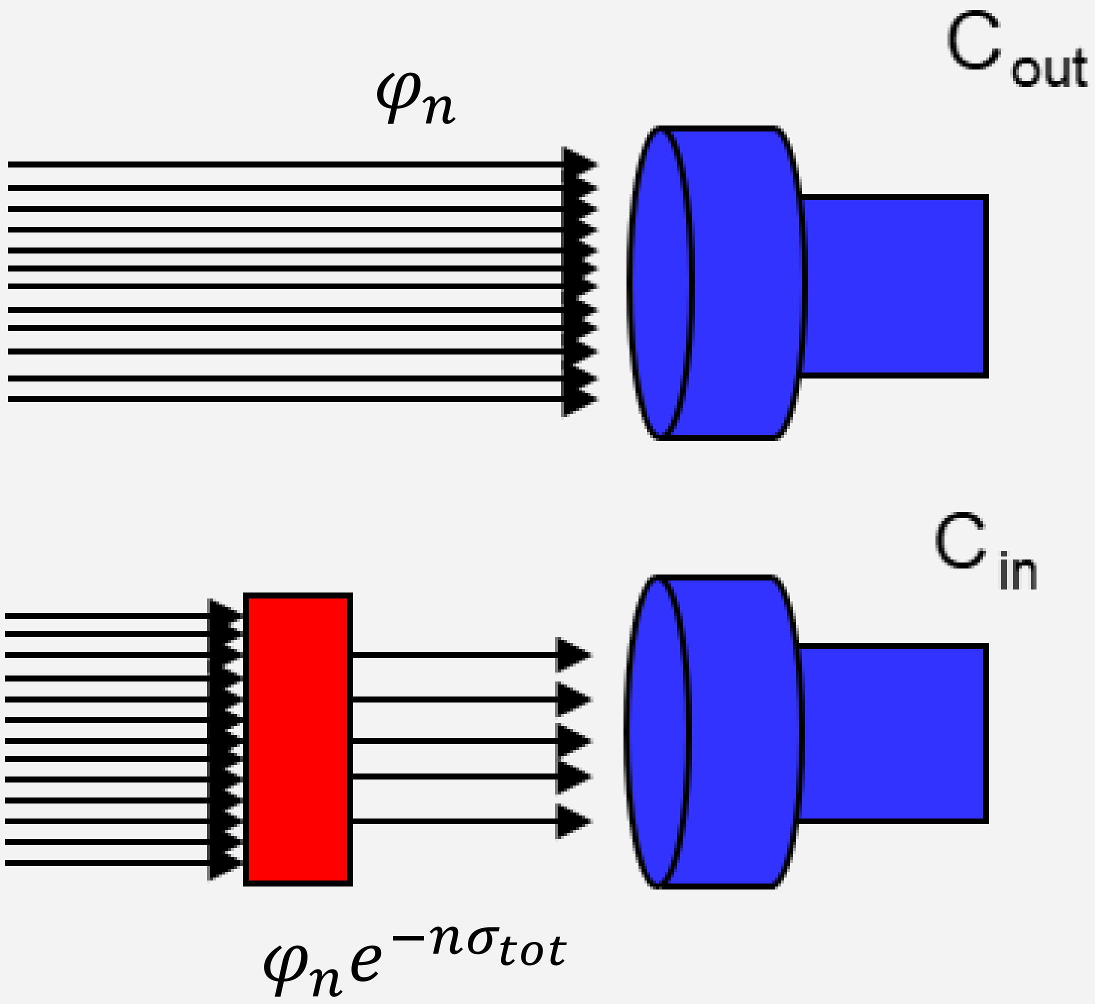
t → Time-of-flight

$$t = (T_s - t_0) - (t_\gamma - L/c)$$

$$\frac{\Delta E}{E} = (1+\gamma)\gamma\frac{\Delta v}{v} \approx 2\frac{\Delta v}{v} = 2\sqrt{\left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta L}{L}\right)^2}$$



Transmission



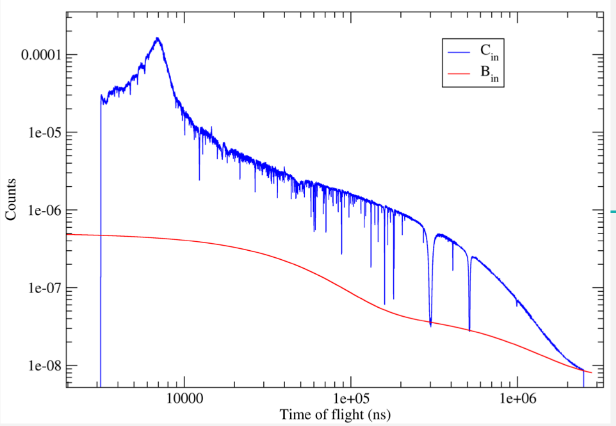
Percentage of neutrons that traverses a sample without interacting with it

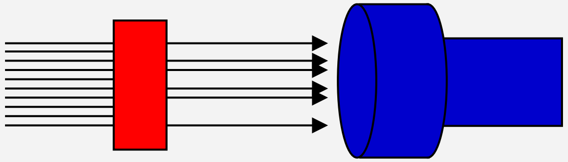
$$T = \mathrm{N}\frac{C_{in}(t) - KB_{in}(t)}{C_{out}(t) - KB_{out}(t)} = \frac{\varphi_n e^{-n\sigma_{tot}}}{\varphi_n} = e^{-n\sigma_{tot}}$$

- Sample-in and sample-out measurement divided in many short cycles
- Estimation of background using black resonance filters *(see later)*
- N normalization factor (1,0000 ± 0,0025)
- K correlated uncertainty component (1,00 ± 0,04)

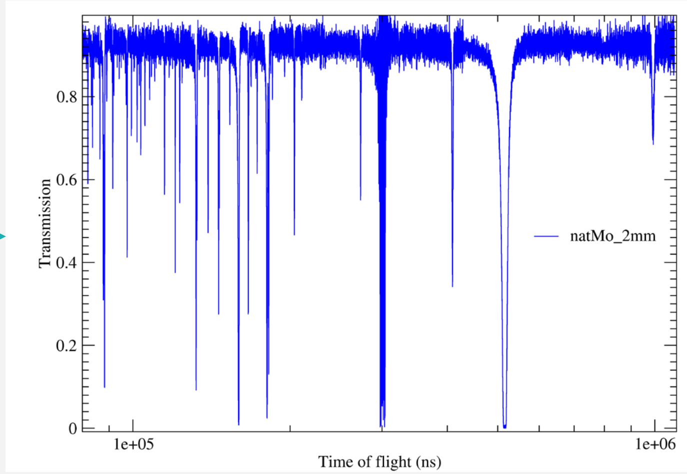
Transmission spectrum

$$C_{in} = \varepsilon T \varphi_n$$

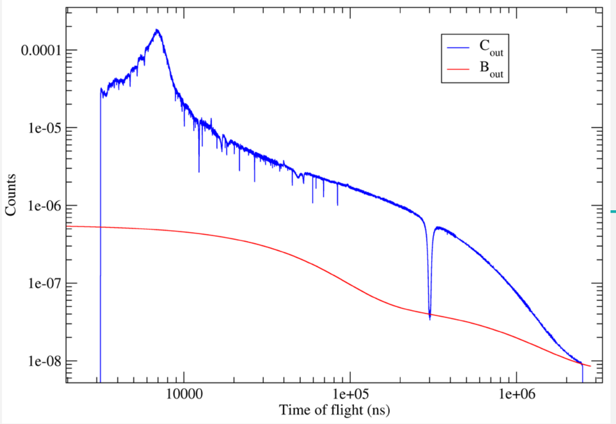


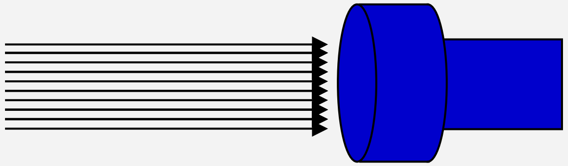


$$T_{exp} = N \frac{C_{in} - KB_{in}}{C_{out} - KB_{out}}$$



$$C_{out} = \varepsilon \varphi_n$$





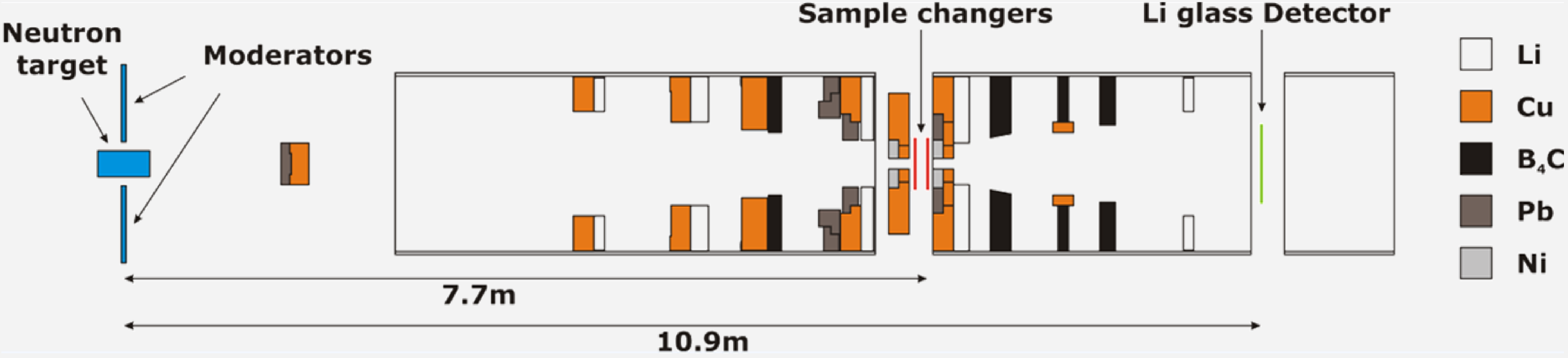
Transmission measurements

$$T_{exp} = \frac{C_{in}}{C_{out}} \propto e^{-n\sigma_{tot}}$$

1) All detected neutrons passed through the sample
2) Neutrons scattered in the sample do not reach detector
3) Sample perpendicular to parallel neutron beam

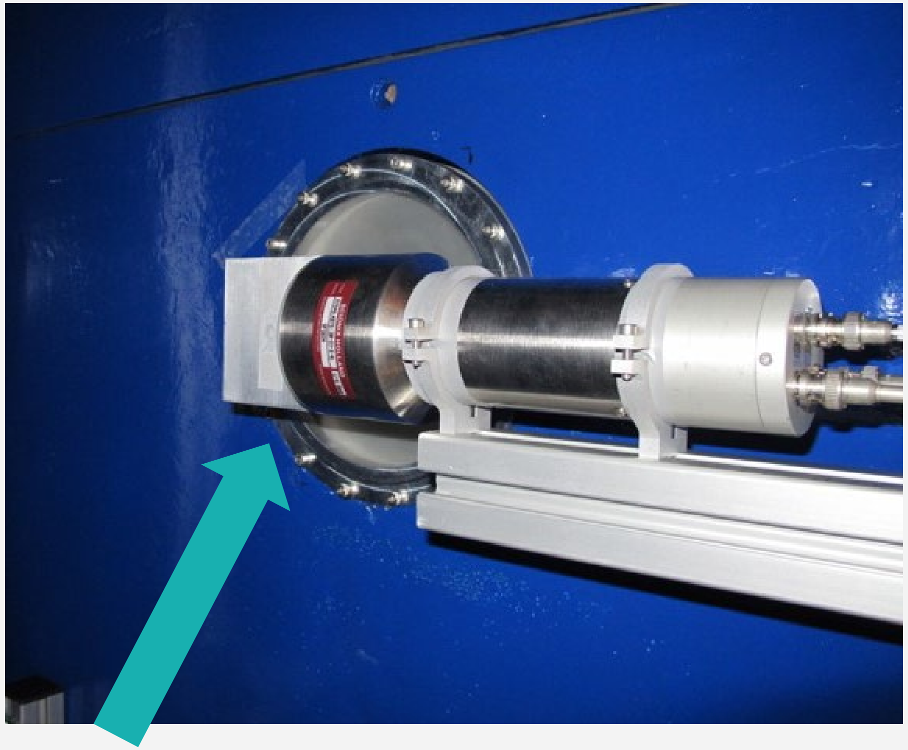
$\Rightarrow$ Good transmission geometry (collimation)

4) Homogeneous sample:
 - no spatial distribution
 - no holes



Detection system

- Li glass scintillators
- Enriched to 95% in ^{6}Li
- Placed inside metallic "castle" to reduce background
- Amplitude and time signals
- Time resolution 4,21 ns



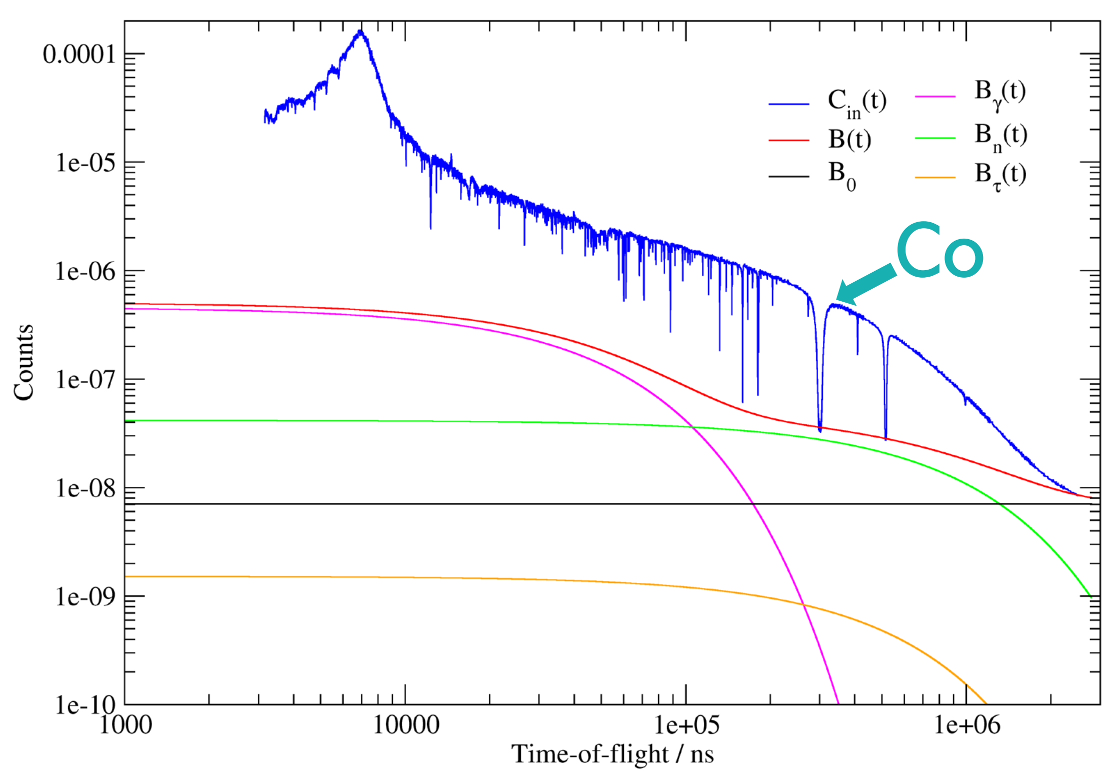
Scintillator

Castle

Background

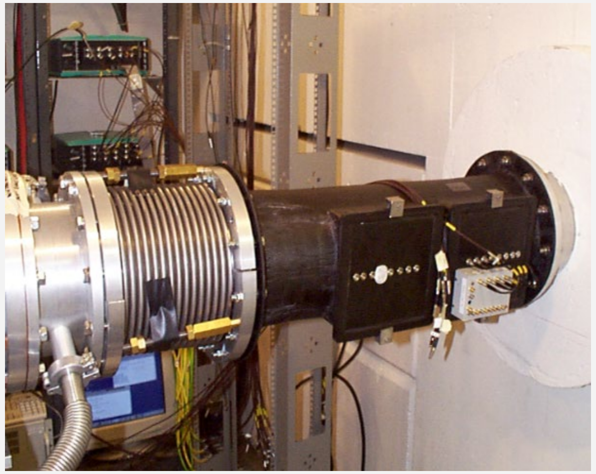
$$B(t) = b_0 + b_1 e^{-\lambda_1 t} + b_2 e^{-\lambda_2 t} + b_3 e^{-\lambda_3 (t+\tau_0)}$$

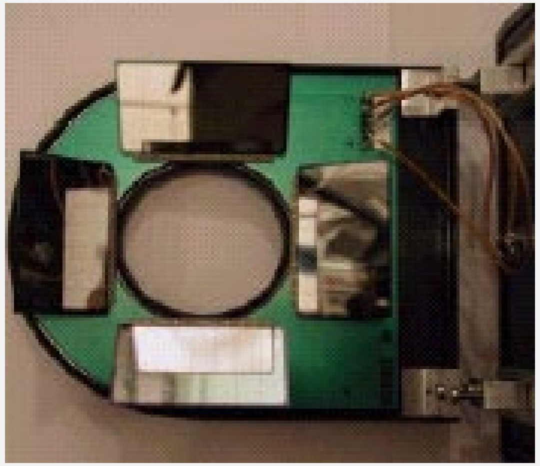
- b_0 time independent background
- $b_1 e^{-\lambda_1 t}$ neutron capture in hydrogen of moderator
- $b_2 e^{-\lambda_2 t}$ neutrons scattered inside the detector station
- $b_3 e^{-\lambda_3 (t+\tau_0)}$ neutron from previous cycle ($\tau_0 = 1/f$)

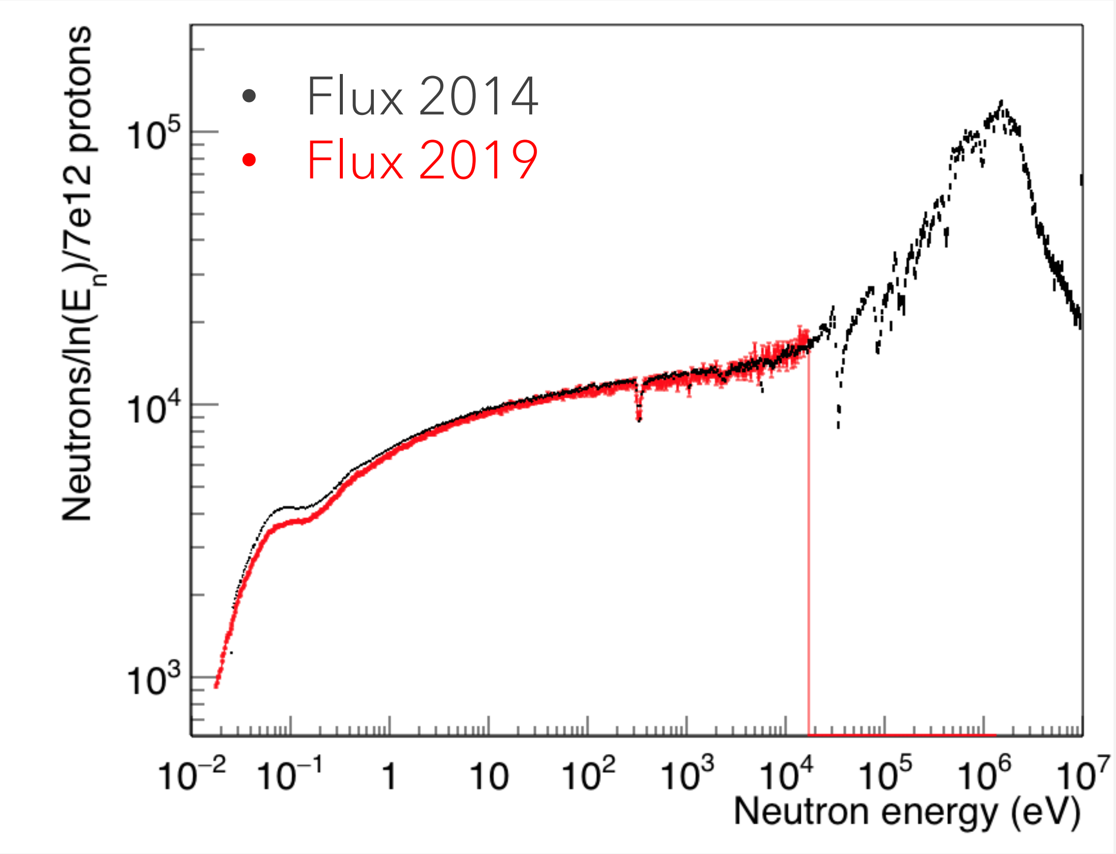


Neutron flux monitor

- Neutron flux continuously monitored;
- SiMON (Silicon MONitor) in beam;
- Silicon detectors facing mylar foil coated in lithium;
- Minimal reduction of neutron flux.



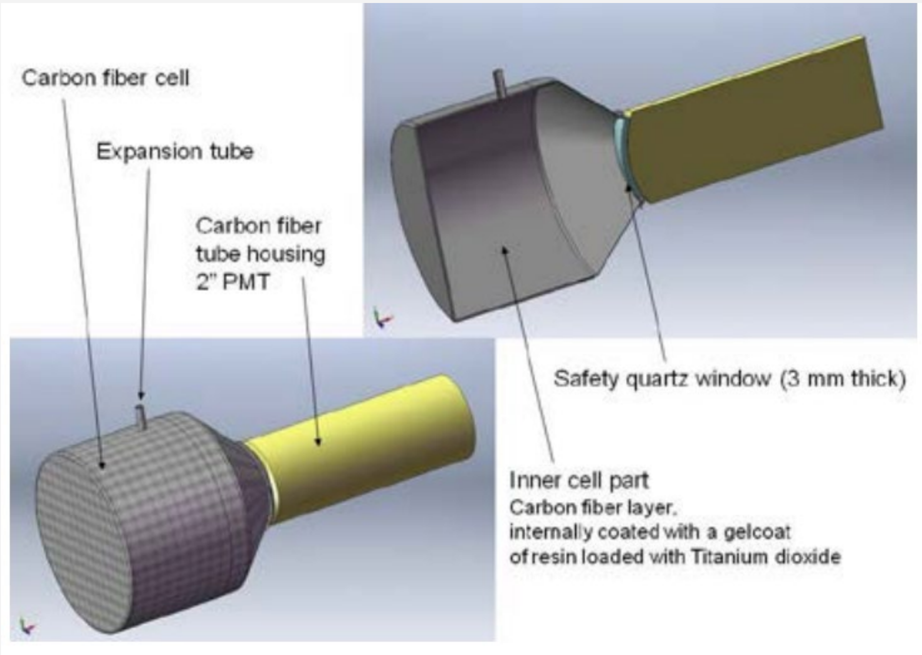


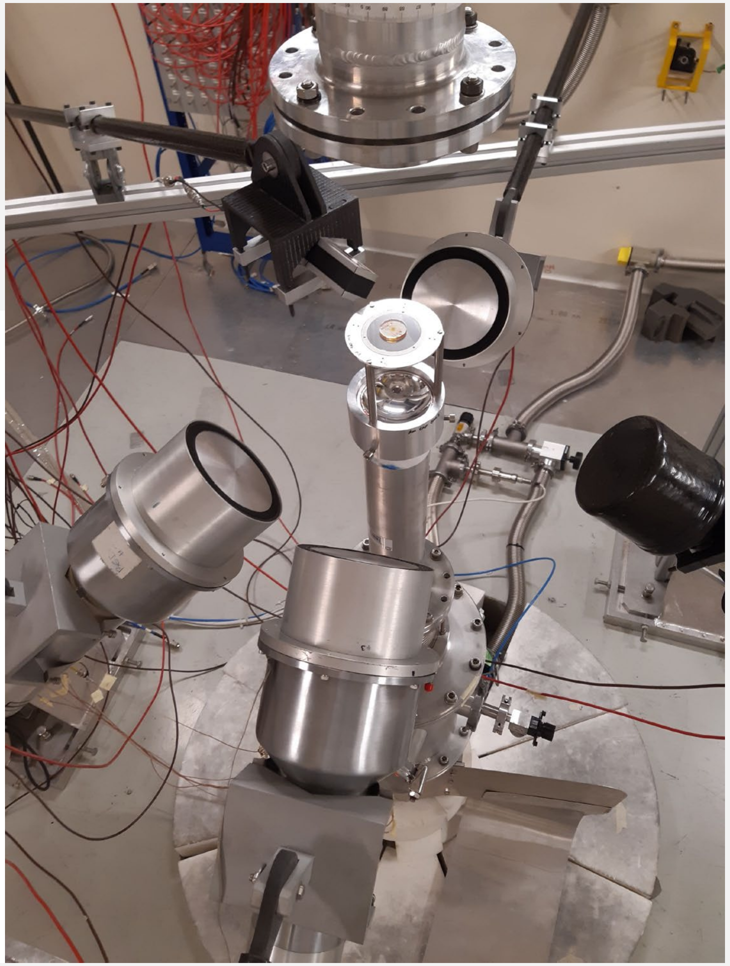


Capture detectors

C6D6 detectors

- Low sensitivity to scattered neutrons;
- Fast recovery from gamma flash;
- Small gamma detection efficiency.



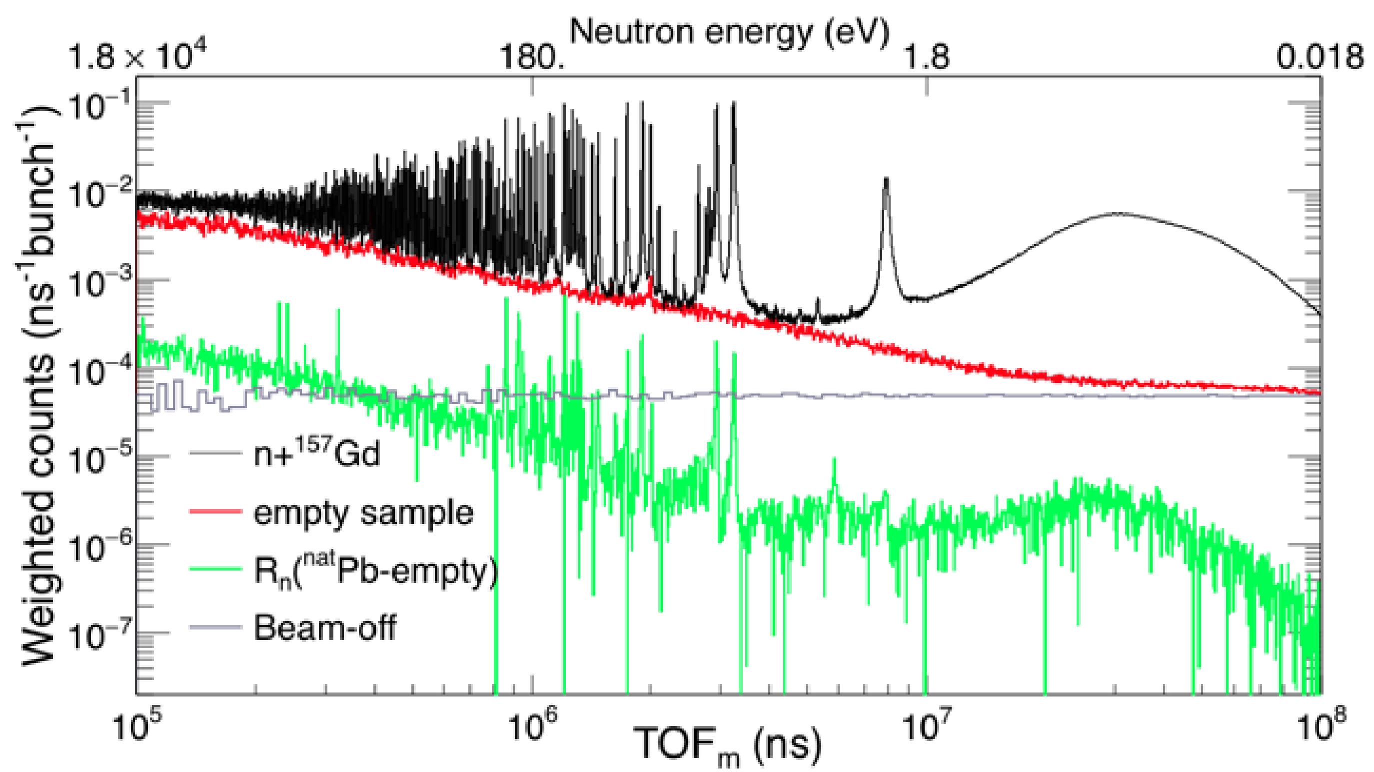


Background

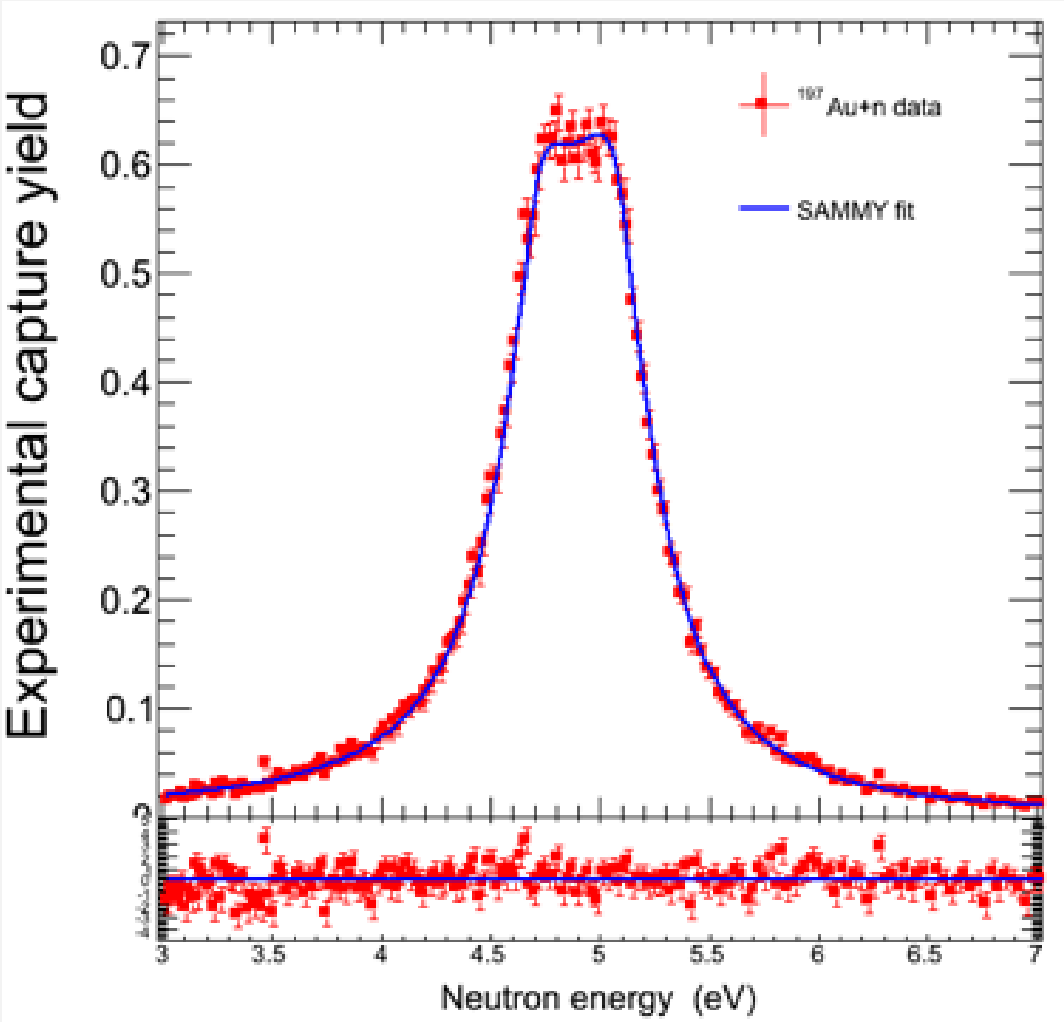
$$B(t) = a_0 + a_1 C_{OB} + a_2 R_n (C_{Pb} - C_{OB})$$

Measurements with open beam, Pb samples and beam off

- a_0 time independent background
- $a_1 C_{OB}$ sample independent, open beam measurement
- $a_2 R_n (C_{Pb} - C_{OB})$ neutrons scattered by the sample, obtained from Pb measurement



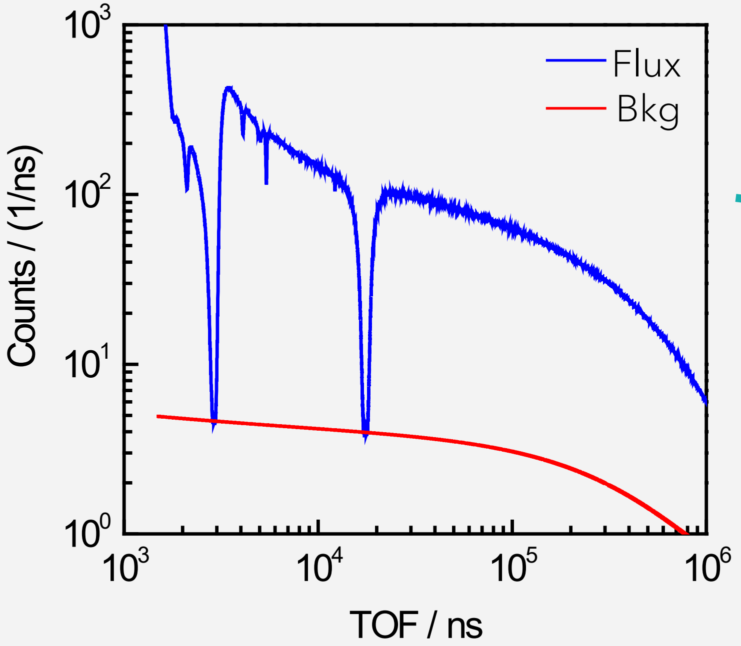
Normalization



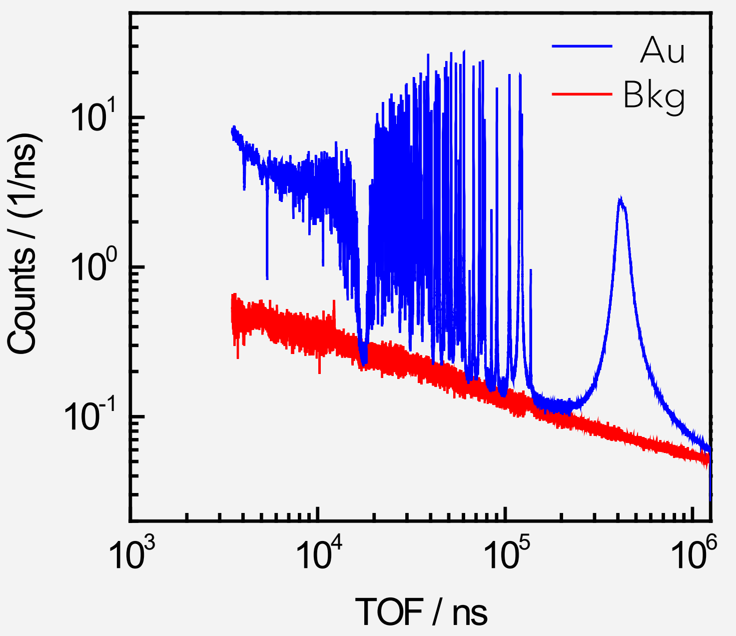
- For a capture resonance with $\Gamma_\gamma \gg \Gamma_n$ the capture cross section is approximately equal to the total cross section $\frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}} \approx 1$
- A saturated resonance ($n\sigma_{tot} \gg 1$) absorbs all the impinging neutrons T≈ 0
- When both conditions are met the capture yield is equal to 1

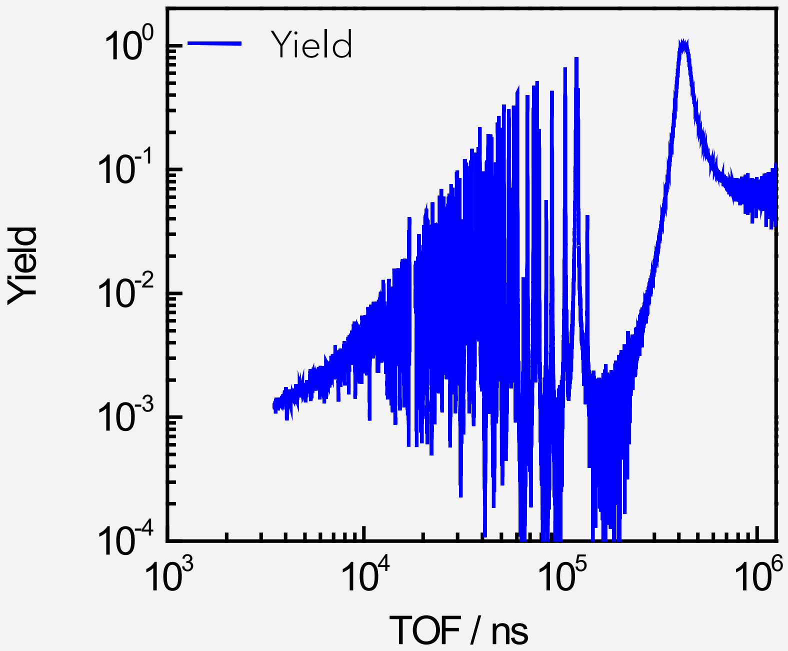
Extract normalization factor from ^{197}Au saturated resonance

Capture yield



$$Y_{\mathrm{exp}} = N \frac{C_\gamma(t) - B_\gamma(t)}{C_\varphi(t) - B_\varphi(t)} Y_\varphi$$





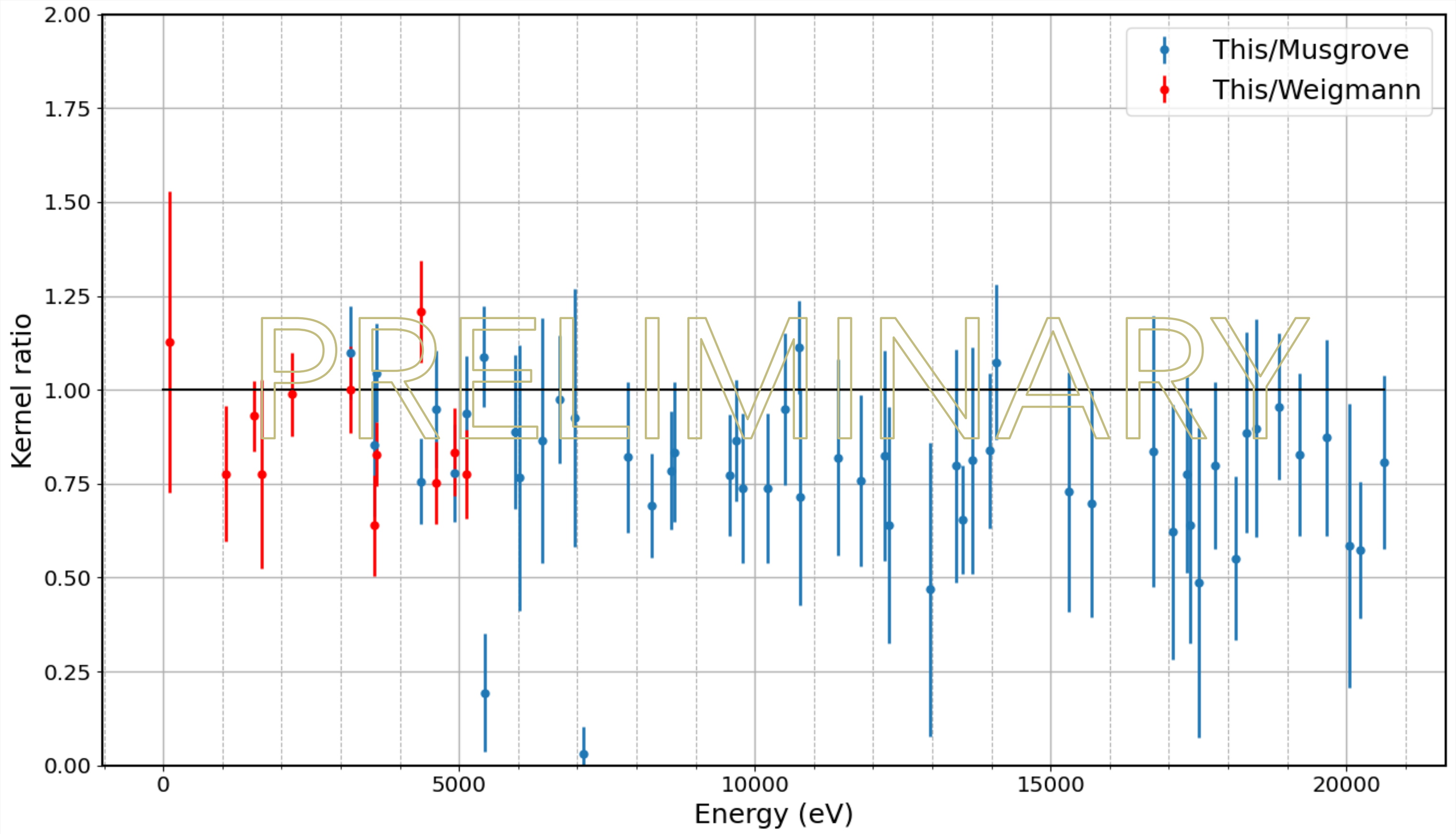
Pulse Height Weighting Technique (PHWT)

Use of detectors with small detection efficiency proportional to γ-ray energy → Capture efficiency proportional to cascade energy → Efficiency almost independent of neutron energy at low energies

↓

Use of Weighting Function (WF) calculated with Monte Carlo simulations to make efficiency proportional to γ-ray energy

Kernel ratio with literature ^{94}Mo



- The preliminary kernels obtained with SAMMY were compared to the ones in literature (Weigmann and Musgrove capture measurements);
- Main measurements used in libraries.