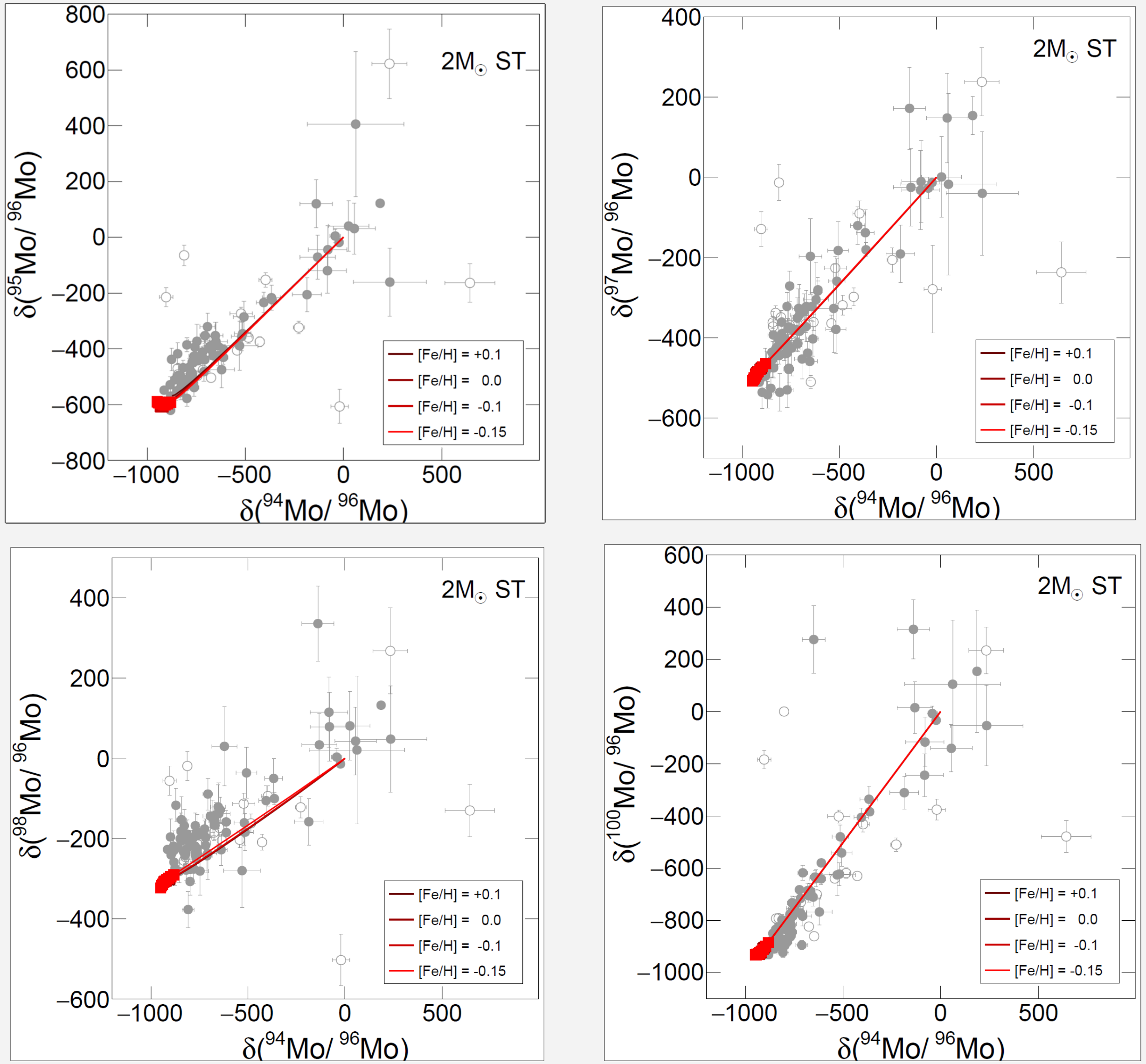
Neutron capture and total cross section measurements on Mo isotopes at n_TOF and GELINA

Riccardo Mucciola

Importance of molybdenum

- Fission product in nuclear power plants;
- Transport casks, irradiated fuel storage;
- Research reactors and Accident Tolerant Fuels;
- Structural material in fusion reactors;
- Stellar nucleosynthesis;
- Production of ^{99}Tc.

Presolar grain composition



- Comparison of SiC grains composition versus stellar model (FRANEC) using delta notation:

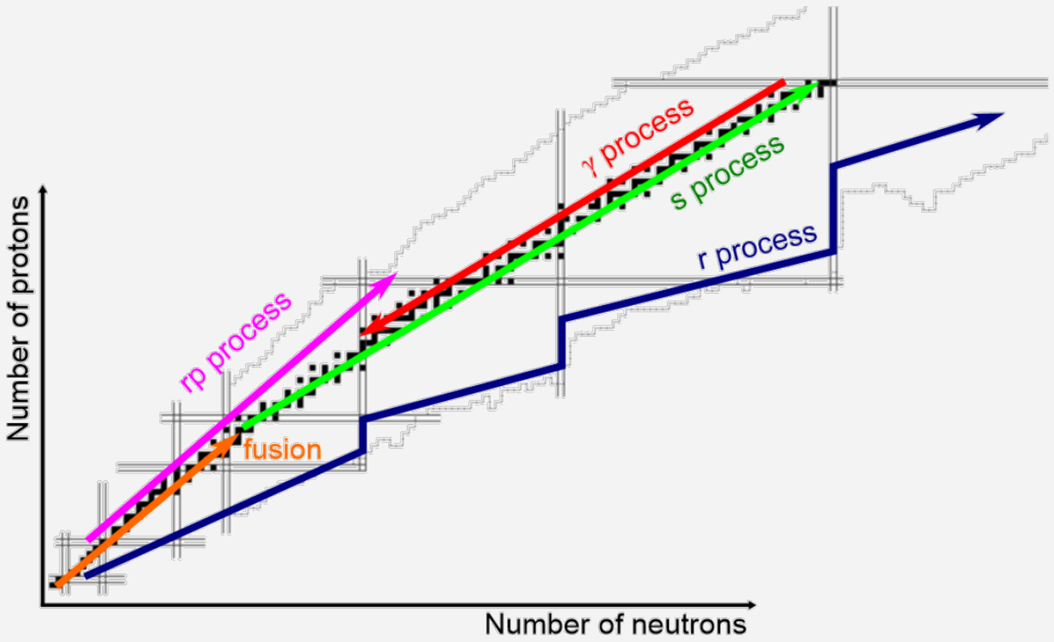
$$\delta\left(\frac{^{95}Mo}{^{96}Mo}\right) = 10^3 \times \left[\left(\frac{^{95}Mo}{^{96}Mo}\right) \Big/ \left(\frac{^{95}Mo}{^{96}Mo}\right)_{\odot} - 1 \right]$$

- MACS from KADoNiS v1.0 database,
- Slight discrepancies between model and isotopic composition,
- Possible overestimation of MACS in KADoNiS.

S. Palmerini et al., ApJ 921 7 (2021)

Stellar nucleosynthesis

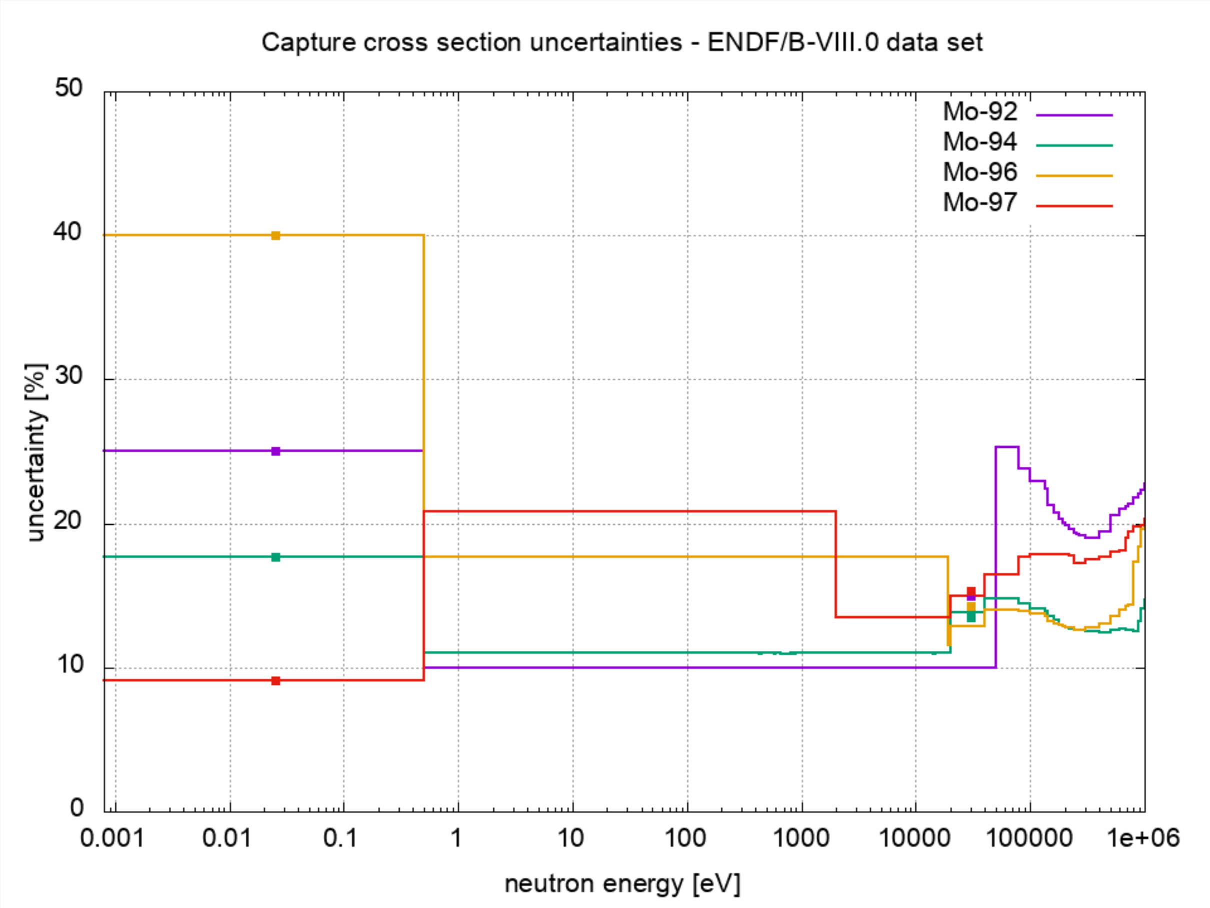
- Four main nucleosynthesis processes for elements heavier than iron: s-process, r-process, i-process, and p-process;
- Some isotopes can be synthetized only by one process (e.g., ^{96}Mo by s-process);
- Possible to set constraints on intensity of the processes.

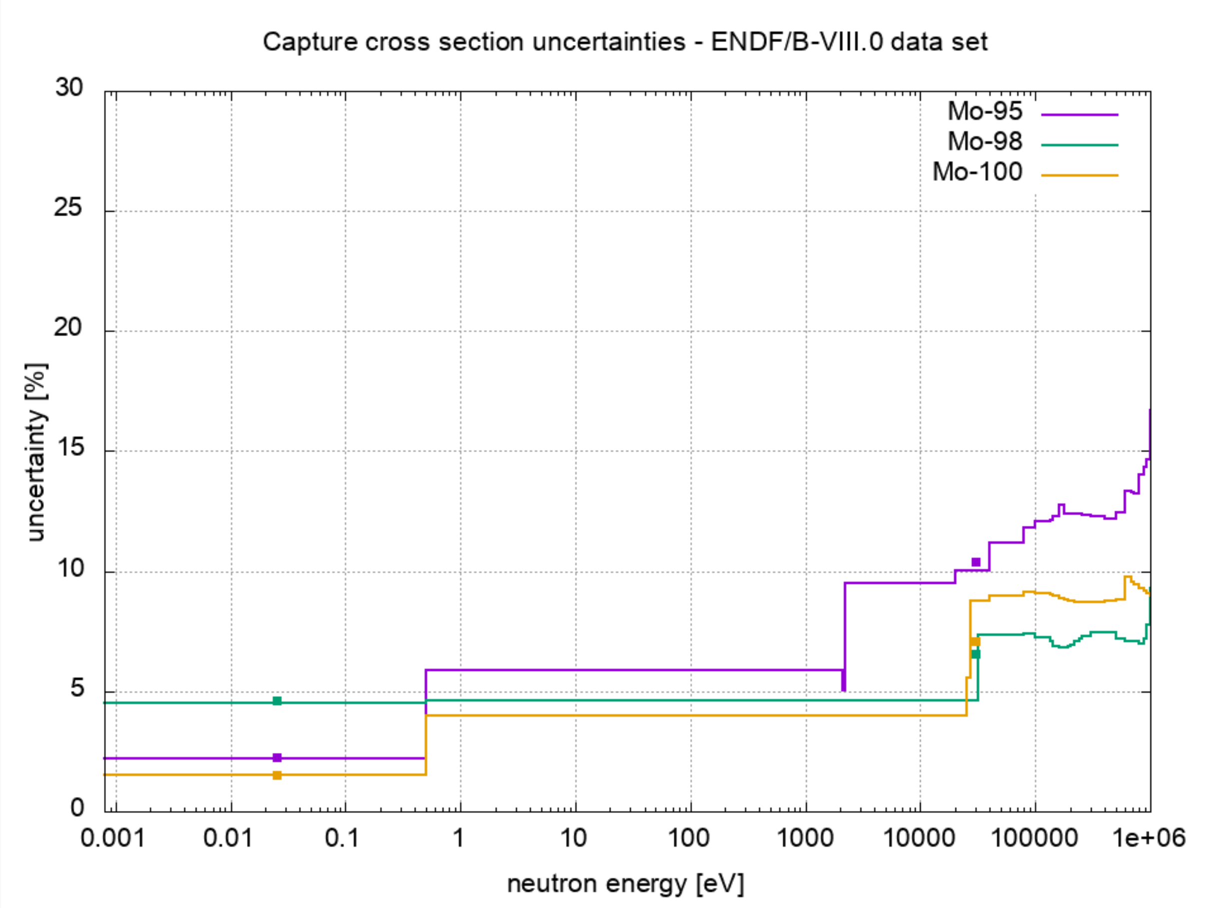


s-process path
around molybdenum

^{94}Ru 51.80 m	^{95}Ru 1.64 h	^{96}Ru 5.54	^{97}Ru 2.79 d	^{98}Ru 1.87	^{99}Ru 12.76	^{100}Ru 12.6	^{101}Ru 17.06	^{102}Ru 31.55
^{93}Tc 2.75 h	^{94}Tc 4.88 h	^{95}Tc 20.00 h	^{96}Tc 4.28 d	^{97}Tc 4.21 Ma	^{98}Tc 4.20 Ma	^{99}Tc 211.11 ka	^{100}Tc 15.80 s	^{101}Tc 14.22 m
^{92}Mo 14.84	^{93}Mo 4.00 ka	^{94}Mo 9.25	^{95}Mo 15.92	^{96}Mo 16.68	^{97}Mo 9.55	^{98}Mo 24.13	^{99}Mo 2.75 d	^{100}Mo 9.63
^{91}Nb 680.04 a	^{92}Nb 34.70 Ma	^{93}Nb 100	^{94}Nb 20.30 ka	^{95}Nb 34.99 d	^{96}Nb 23.35 h	^{97}Nb 1.20 h	^{98}Nb 2.86 s	^{99}Nb 15.00 s
^{90}Zr 51.45	^{91}Zr 11.22	^{92}Zr 17.15	^{93}Zr 1.53 Ma	^{94}Zr 17.38	^{95}Zr 64.03 d	^{96}Zr 2.8	^{97}Zr 16.74 h	^{98}Zr 30.70 s

Cross section uncertainties in ENDF/B-VIII

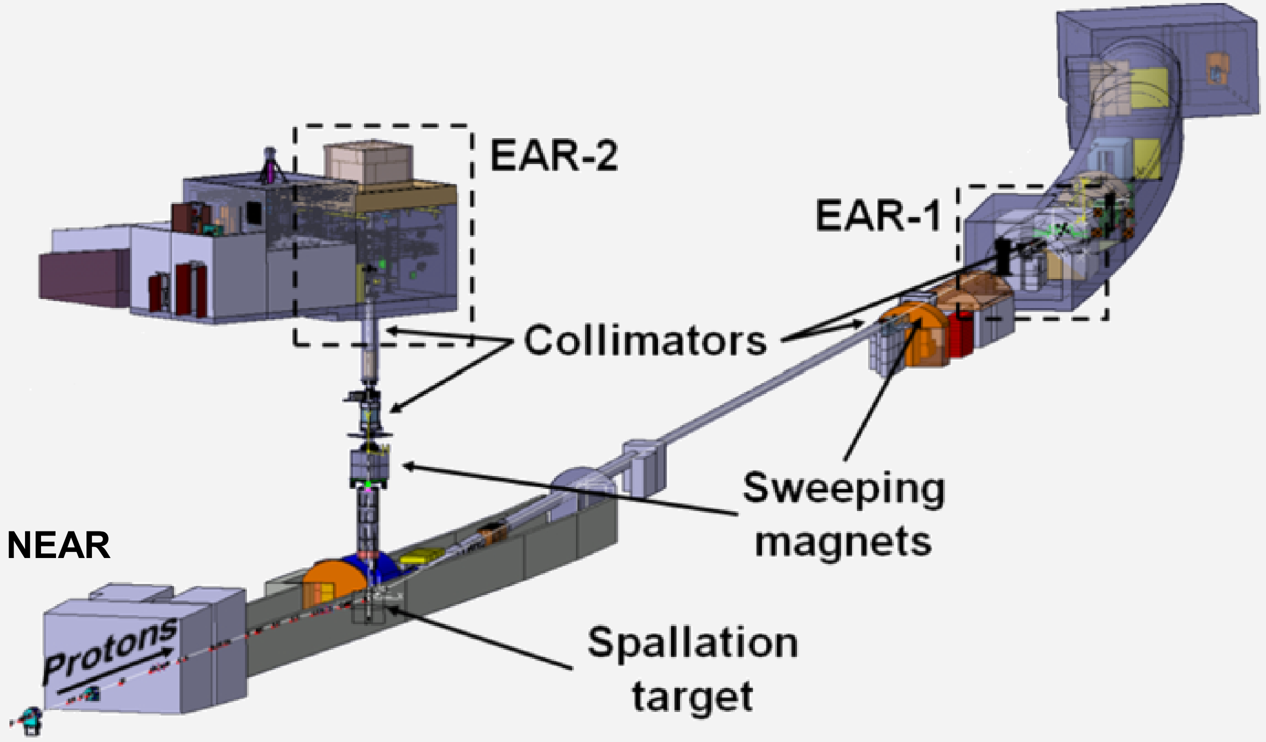




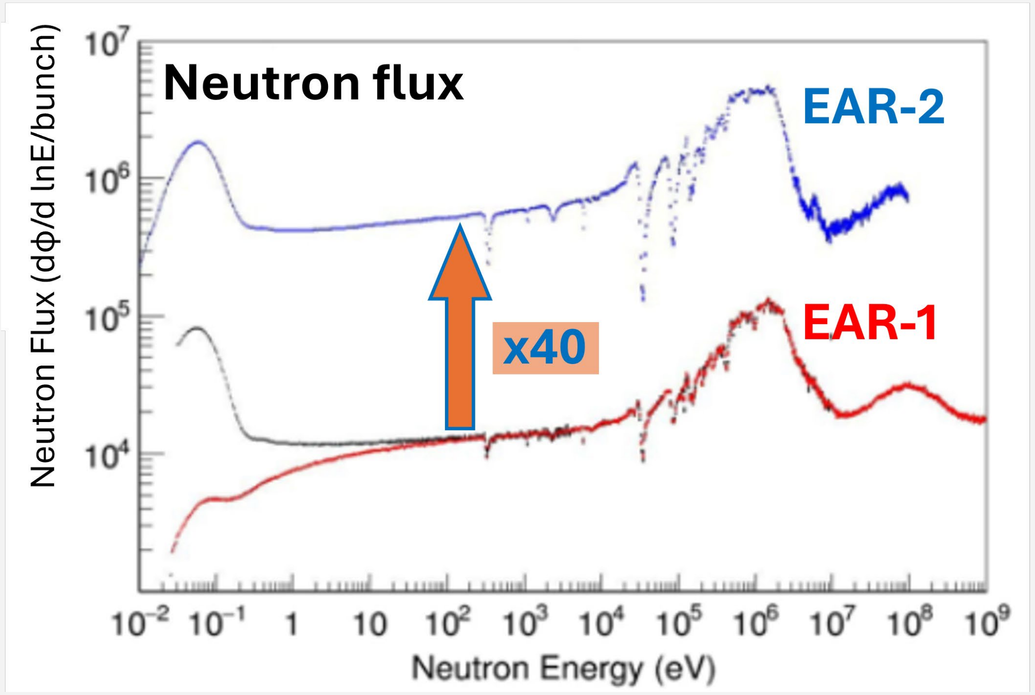
ENDF/B-VIII: D. Brown et al., Nucl. Data. Sheets 148 (2012)

Experimental campaings

n_TOF

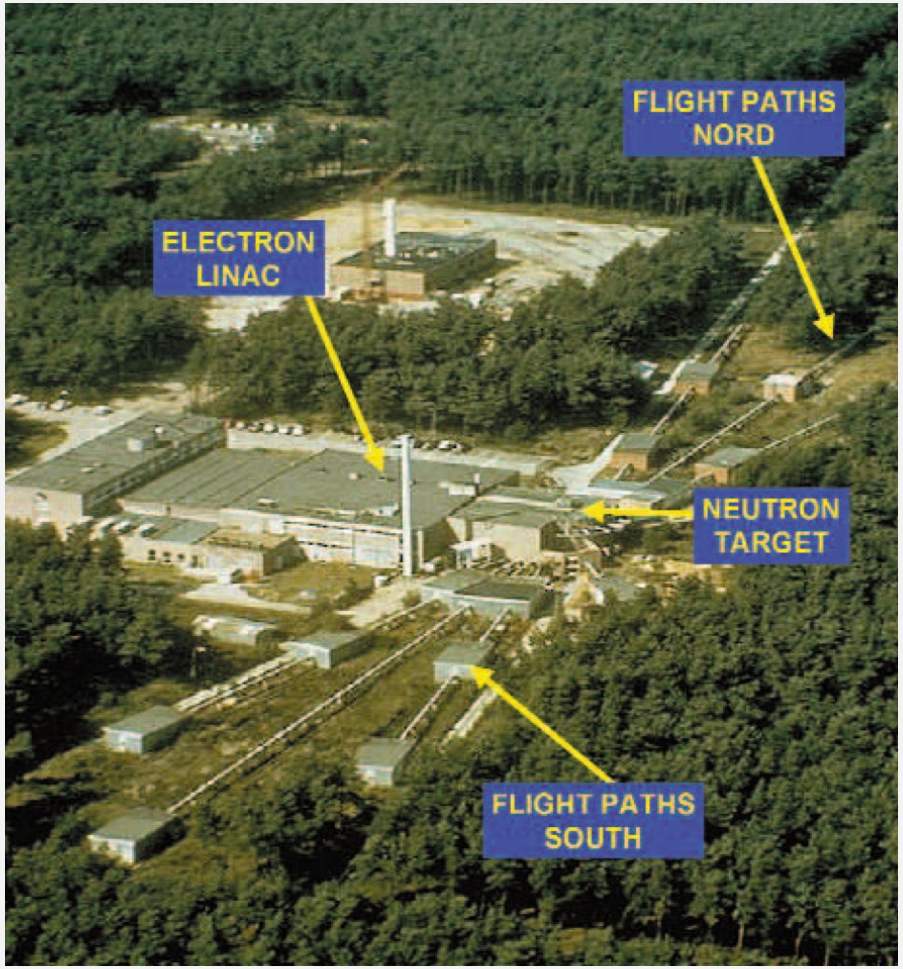


- Located at CERN;
- Neutron beam produced using PS proton on lead target;
- Production of neutrons via spallation;
- Pulsed neutron source (10 meV < E < 1 GeV);
- Three experimental areas (EAR1, EAR2 and NEAR).



GELINA

- Located at JRC-Geel
- Multi-user time-of-flight facility
- Electron beam produced by LINAC (E = 140 MeV)
- Rotating uranium target
- Production of neutrons via (γ,n) or (γ,f)
- Pulsed neutron source (10 meV < E < 20 MeV)
- Water moderators



Enriched pellets preparation

To avoid the background coming from capsules, self-sustaining pellets were prepared using enriched metallic powder:

- Pellets prepared at JRC-Geel and CERN;
- Self sustaining pellets of ~ 2g;
- Additional ^{nat}Mo samples prepared using powder with different grain sizes;

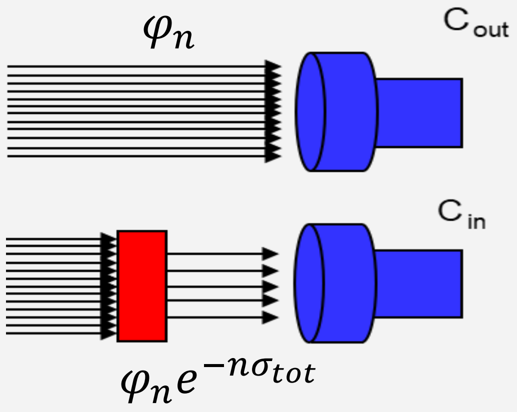
Mo pellet samples

Atomic %	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
^{92}Mo	99.80%	0.114%	0.053%	0.018%	0.0047%	0.01%	0.003%
^{94}Mo	0,63%	98,97%	0,36%	0,01%	0,01%	0,01%	0,01%
^{95}Mo	0,31%	0,69%	95,40%	2,24%	0,51%	0,65%	0,20%
^{96}Mo	0,28%	0,24%	1,01%	95,90%	1,00%	1,32%	0,25%
^{97}Mo	0.05%	0.16%	0.28%	0.55%	98.2%	0.56%	0.2%
^{98}Mo	0.005%	0.005%	0.005%	0.005%	1.15%	98.67%	0.16%
^{100}Mo	0.002%	0.002%	0.002%	0.002%	0.002%	0.18%	99,81%

Experimental campaigns

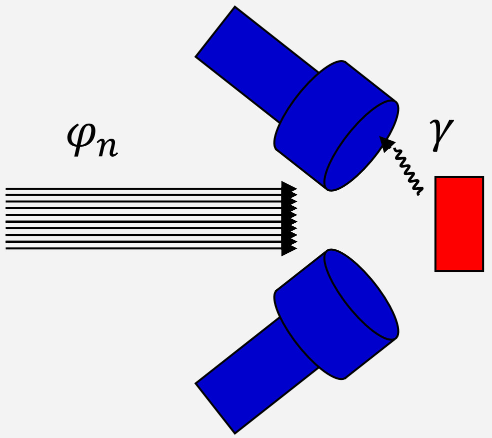
Transmission measurements

- Carried out at GELINA
- Total cross section measurement
- Natural and enriched samples
- 10 m and 50 m flight path



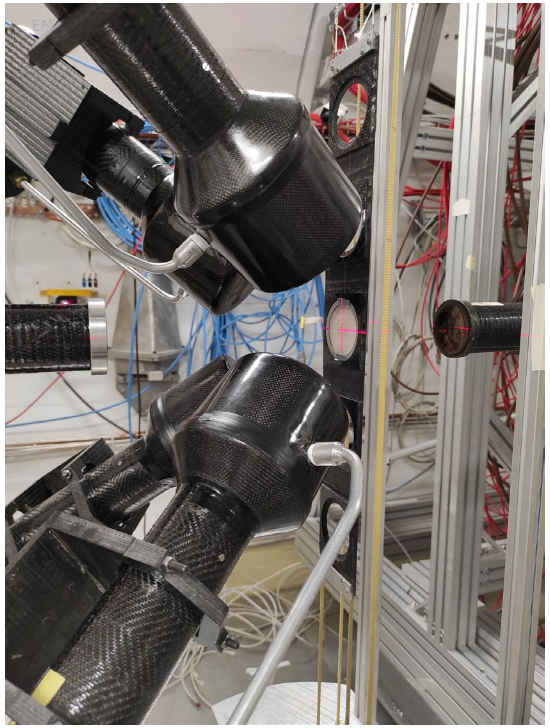
Radiative capture measurements

- Carried out at n_TOF
- Neutron capture cross section
- Both experimental areas of n_TOF (EAR1 and EAR2)



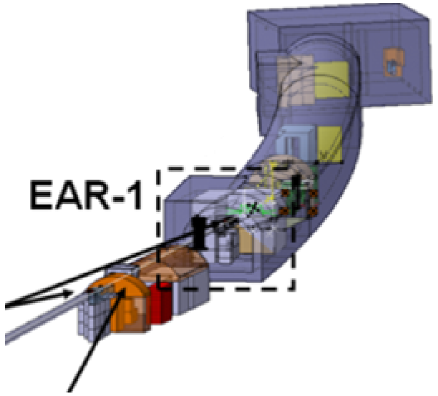
n_TOF measurements setup

EAR1



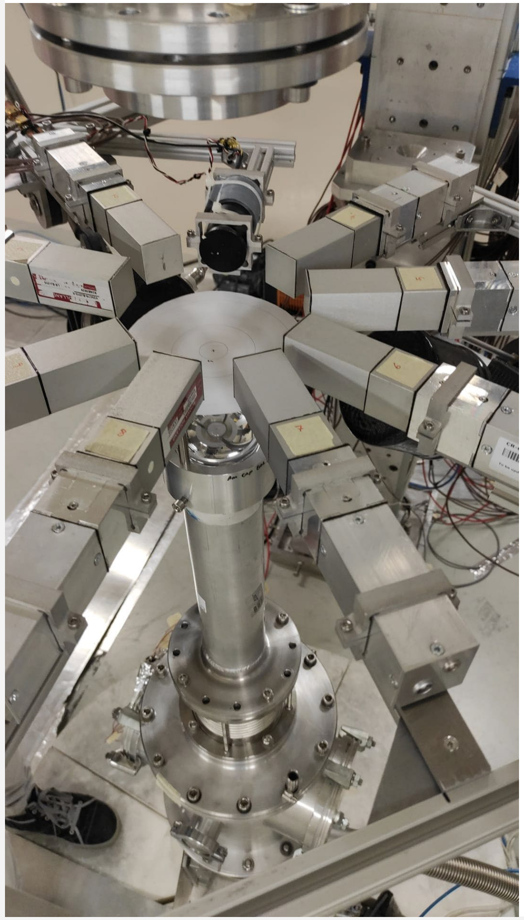
Setup:

- 4 C6D6



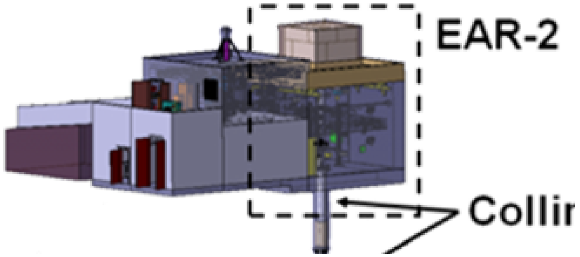
Neutron beam

EAR2



Setup:

- 8 sTED,
- 2 C6D6,
- 1 Stilbene.

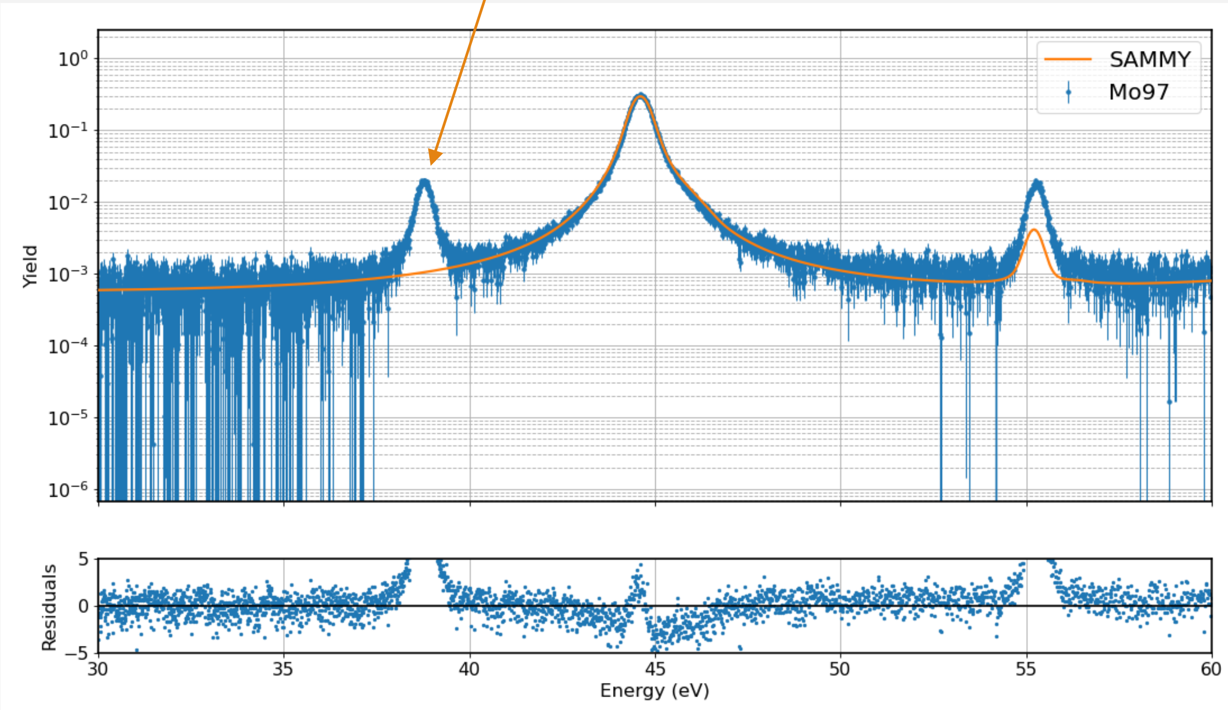


Neutron beam

Preliminary results

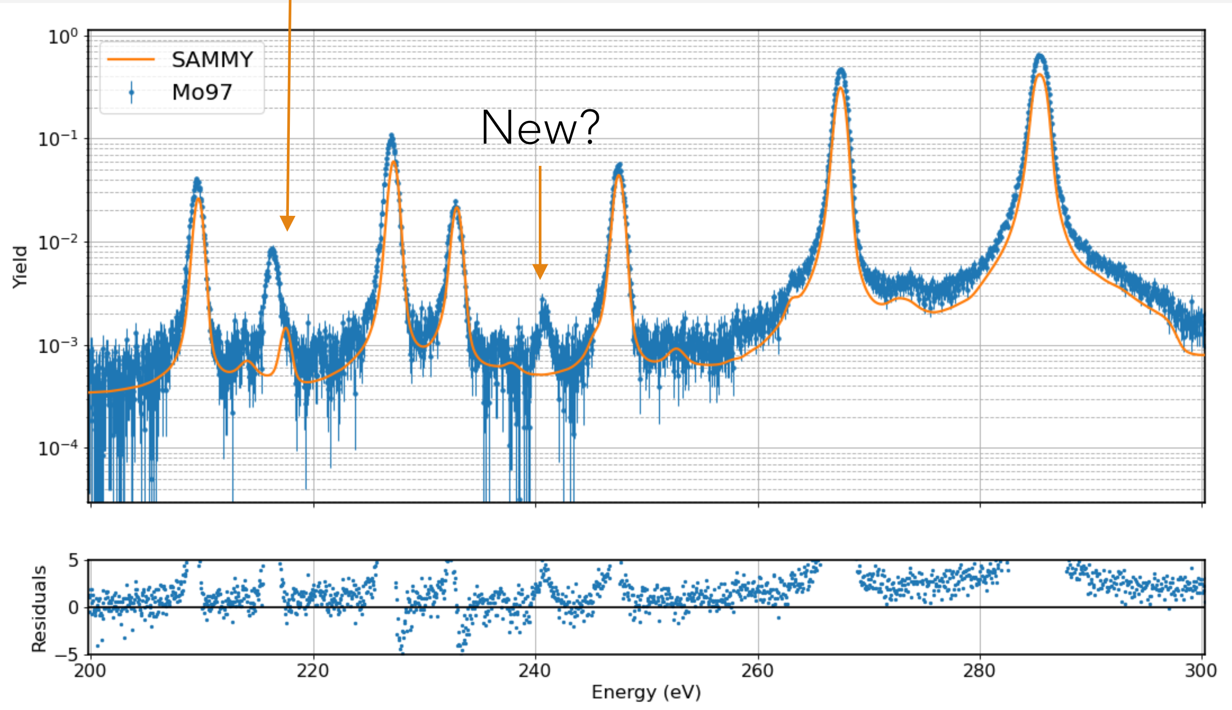
Preliminary ^{97}Mo resonances

Observed at DANCE [1]



Observed at DANCE [1]

New?



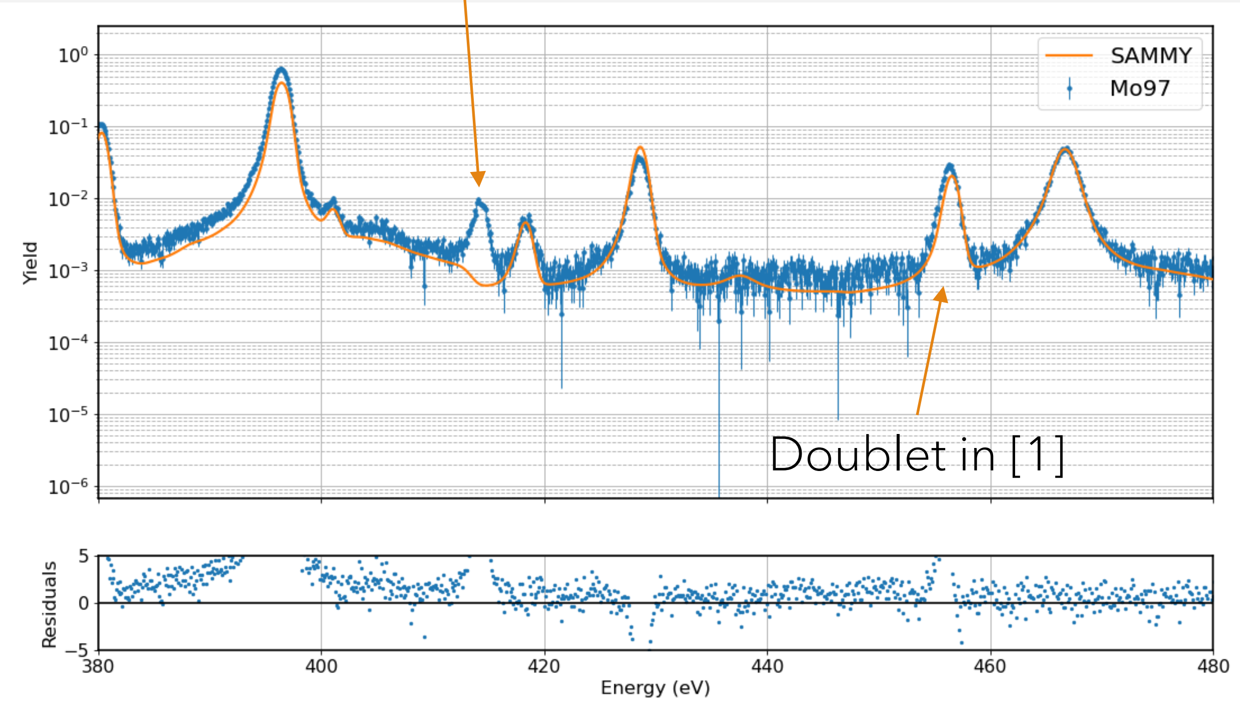
[1] Walker et al., PRC 92,014324 (2015)

Preliminary ^{97}Mo resonances

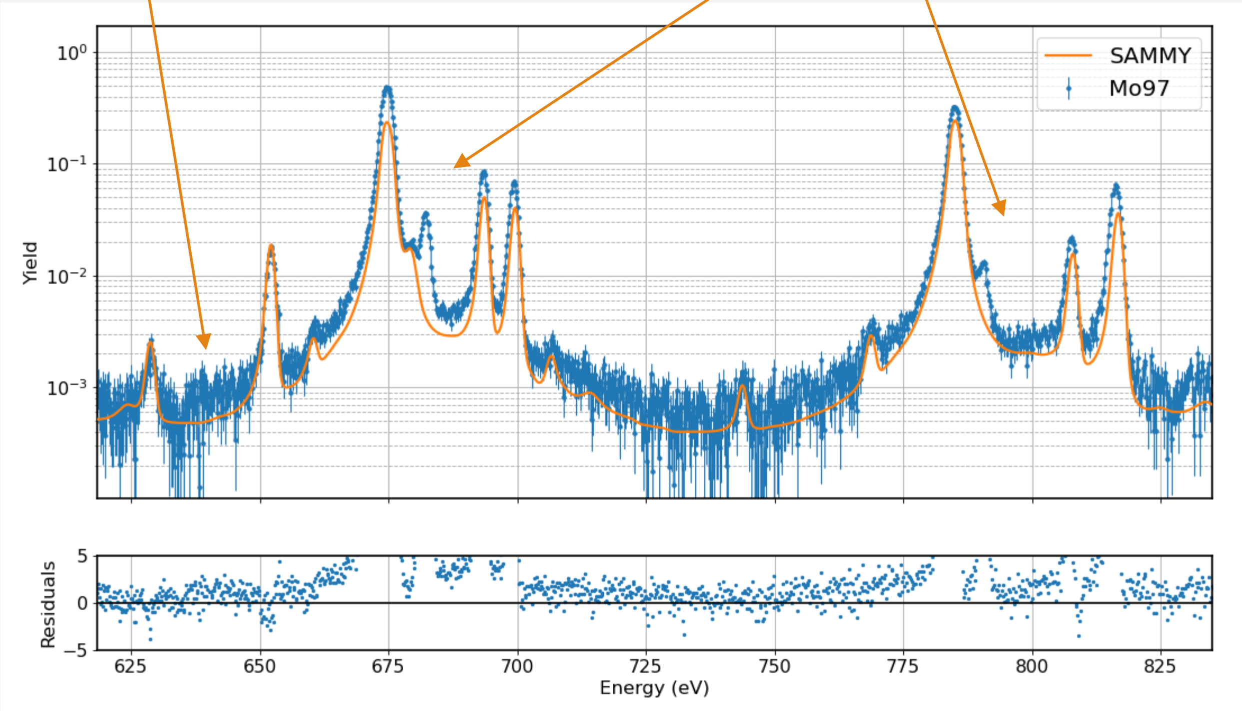
Observed at DANCE [1]

Missing resonance reported in [1]

New?



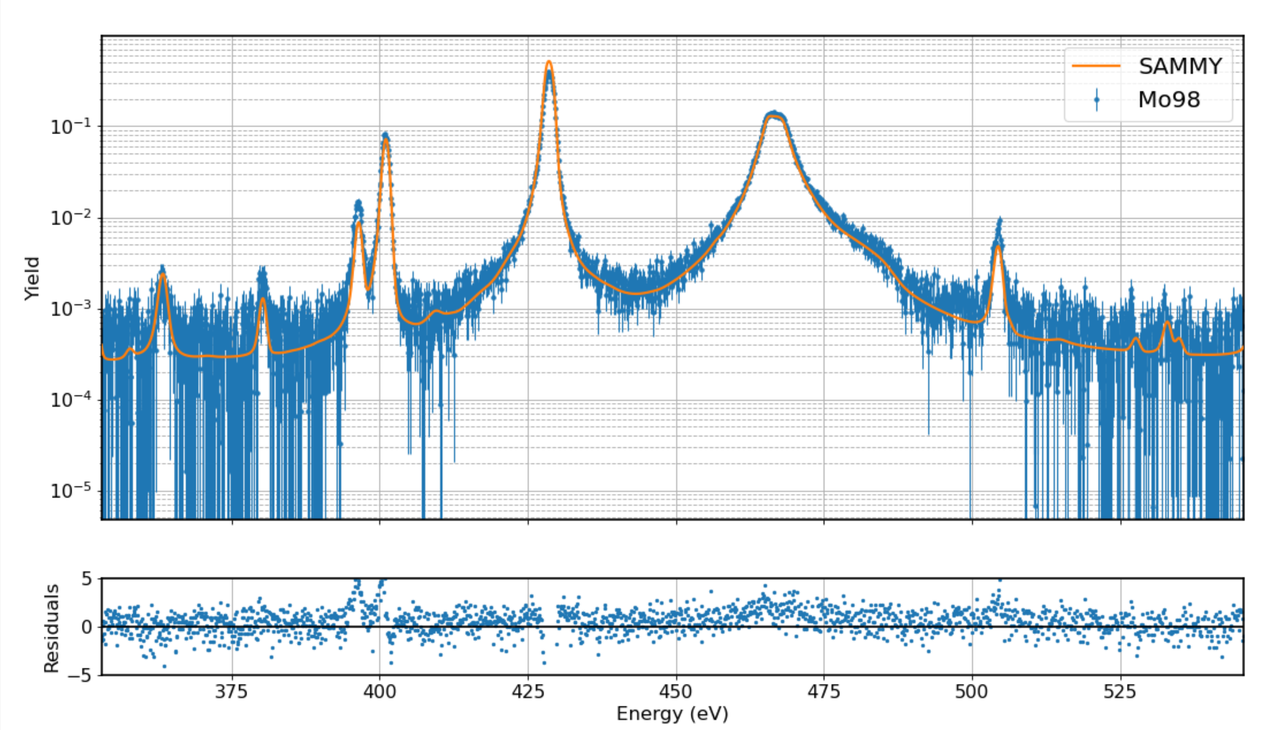
Doublet in [1]

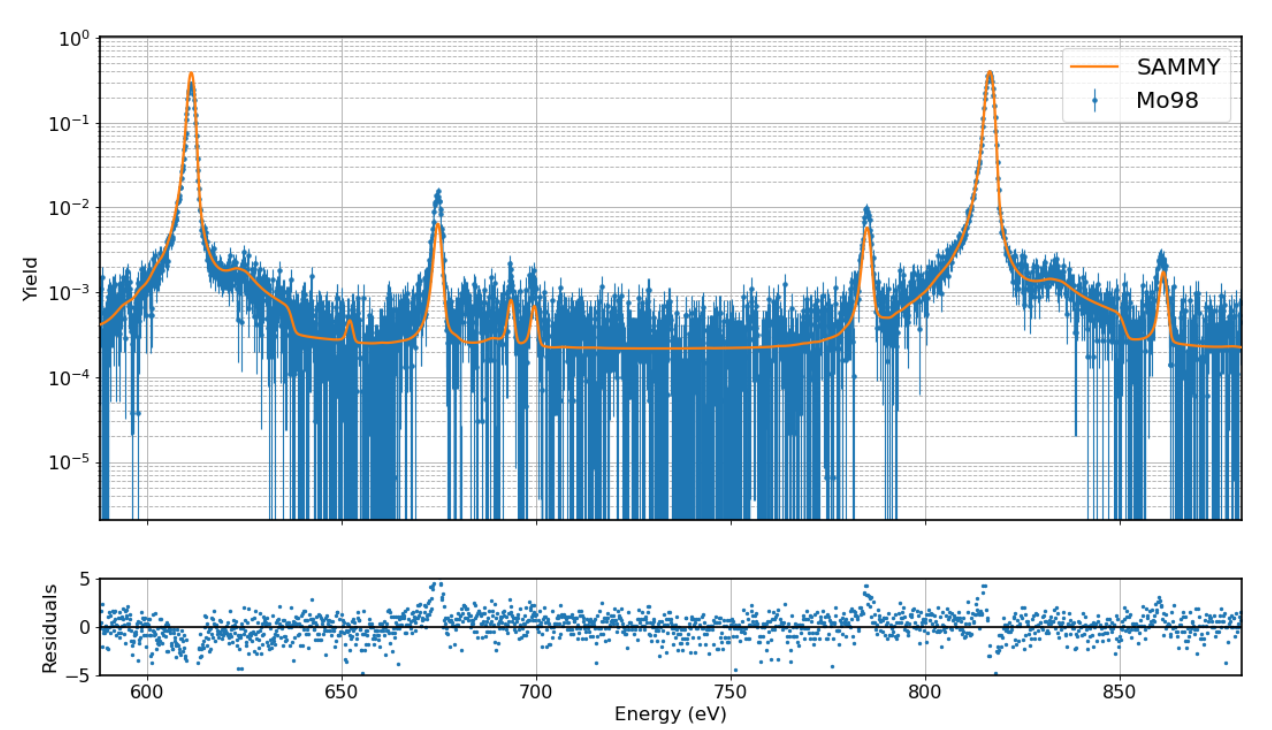


[1] Walker et al., PRC 92,014324 (2015)

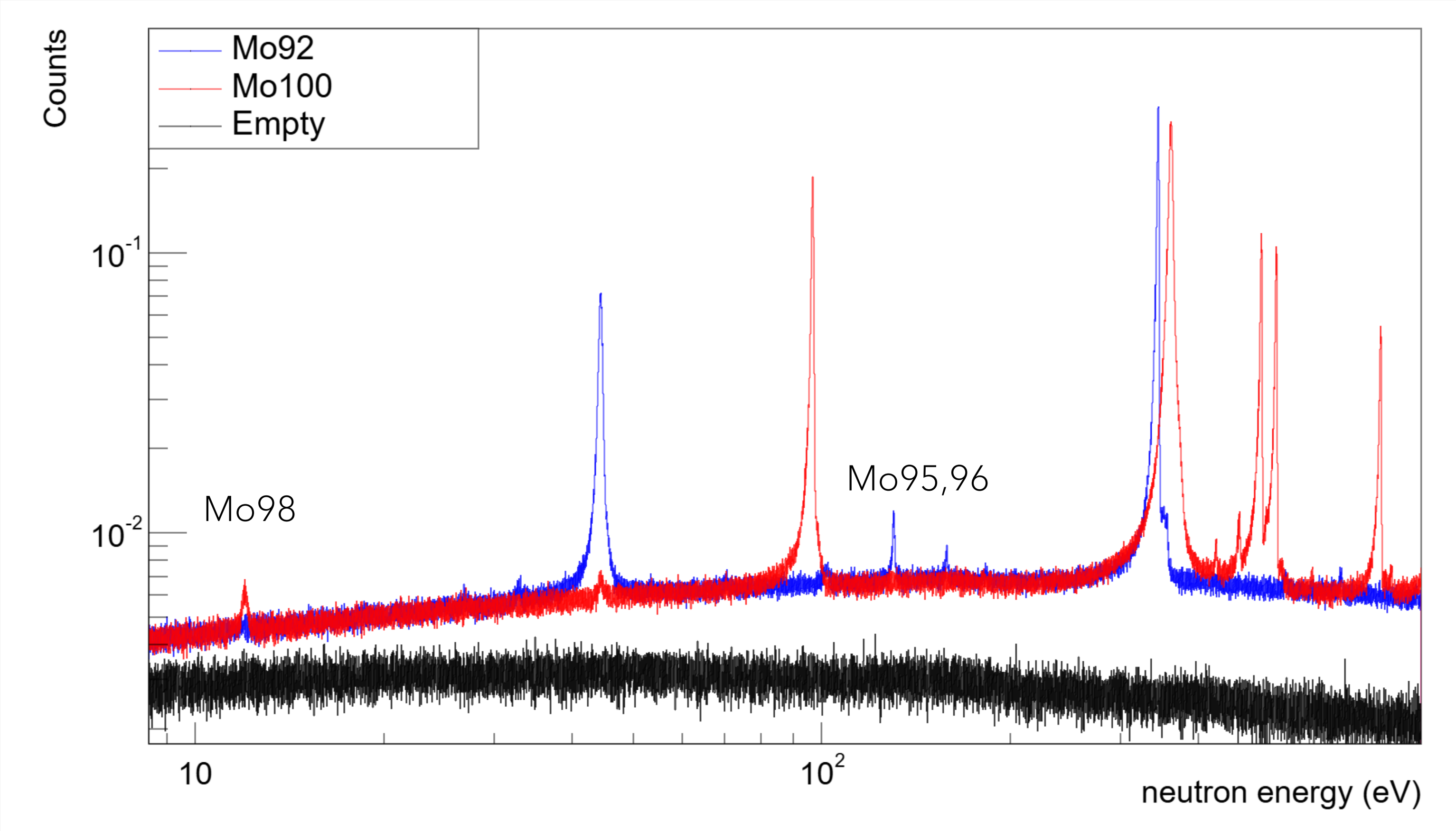
Preliminary ^{98}Mo resonances

Some small deviation from libraries, overall good agreement!





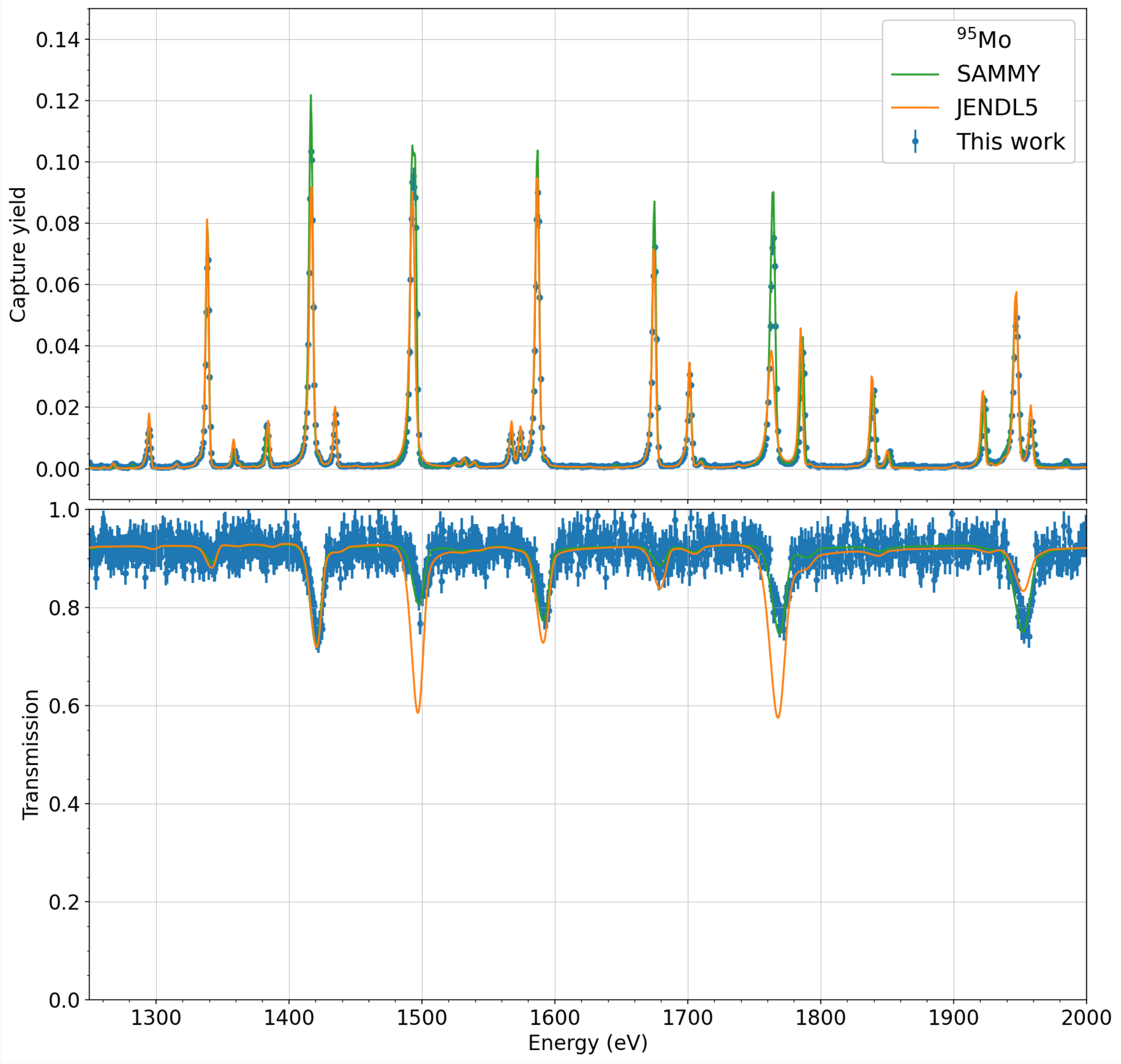
First look at ^{92,100}Mo in EAR2

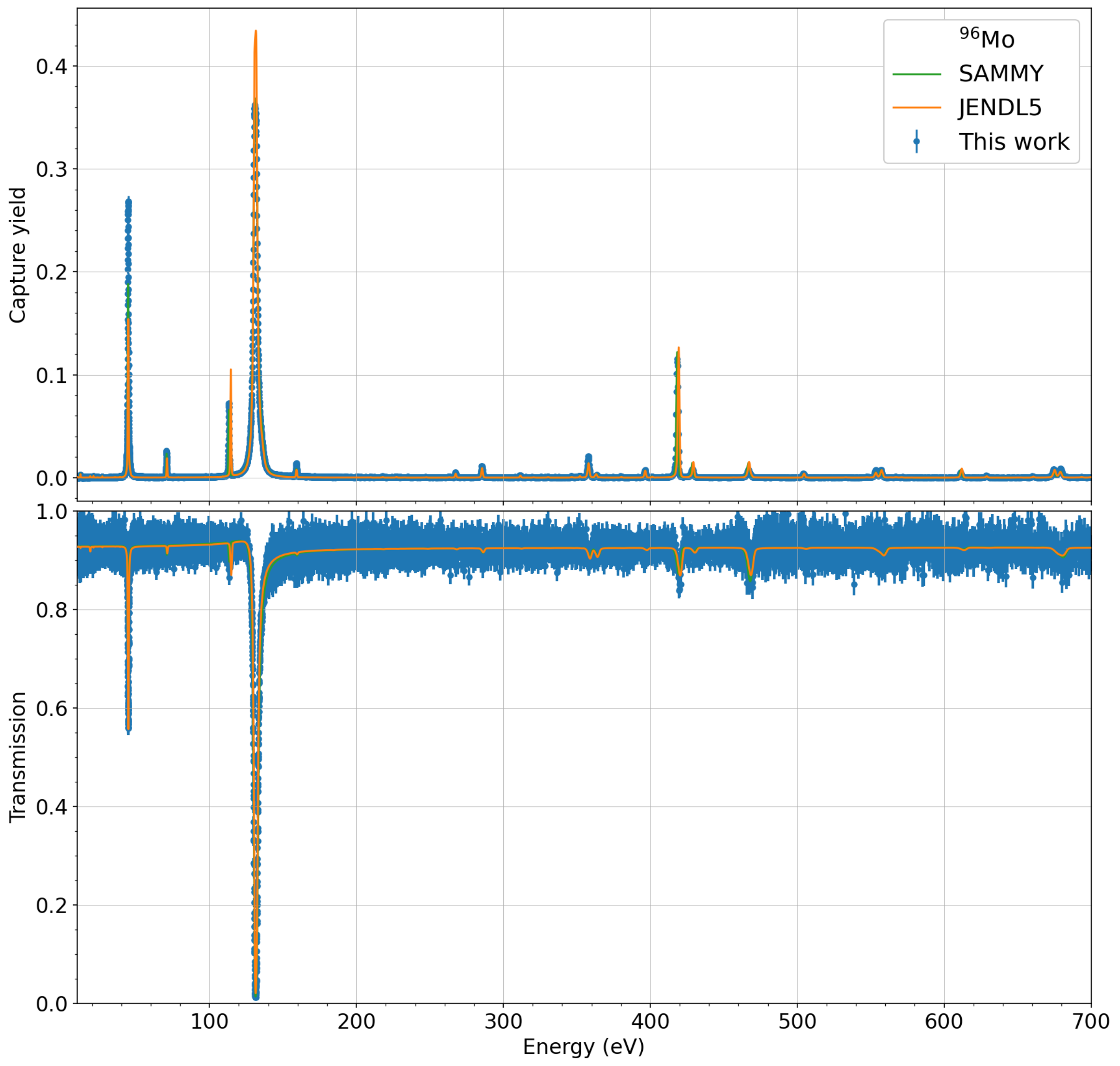


- First TOF spectra of latest measurements with ^{92,100}Mo at EAR2,
- Very small amount of contamination in samples!

Experimental spectra of $^{95,96}Mo$

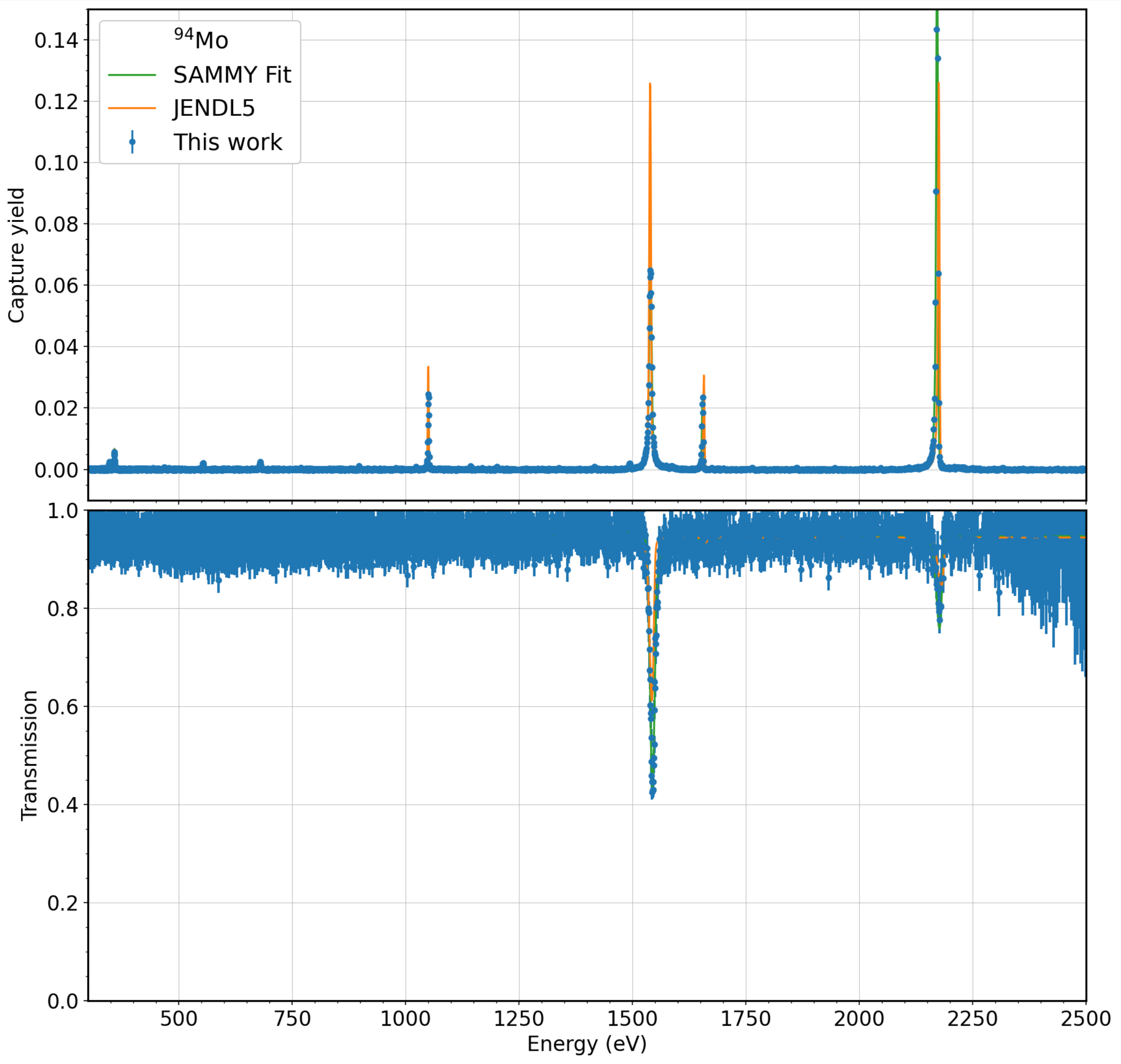
Some deviation from libraries. Good agreement between capture and transmission data!

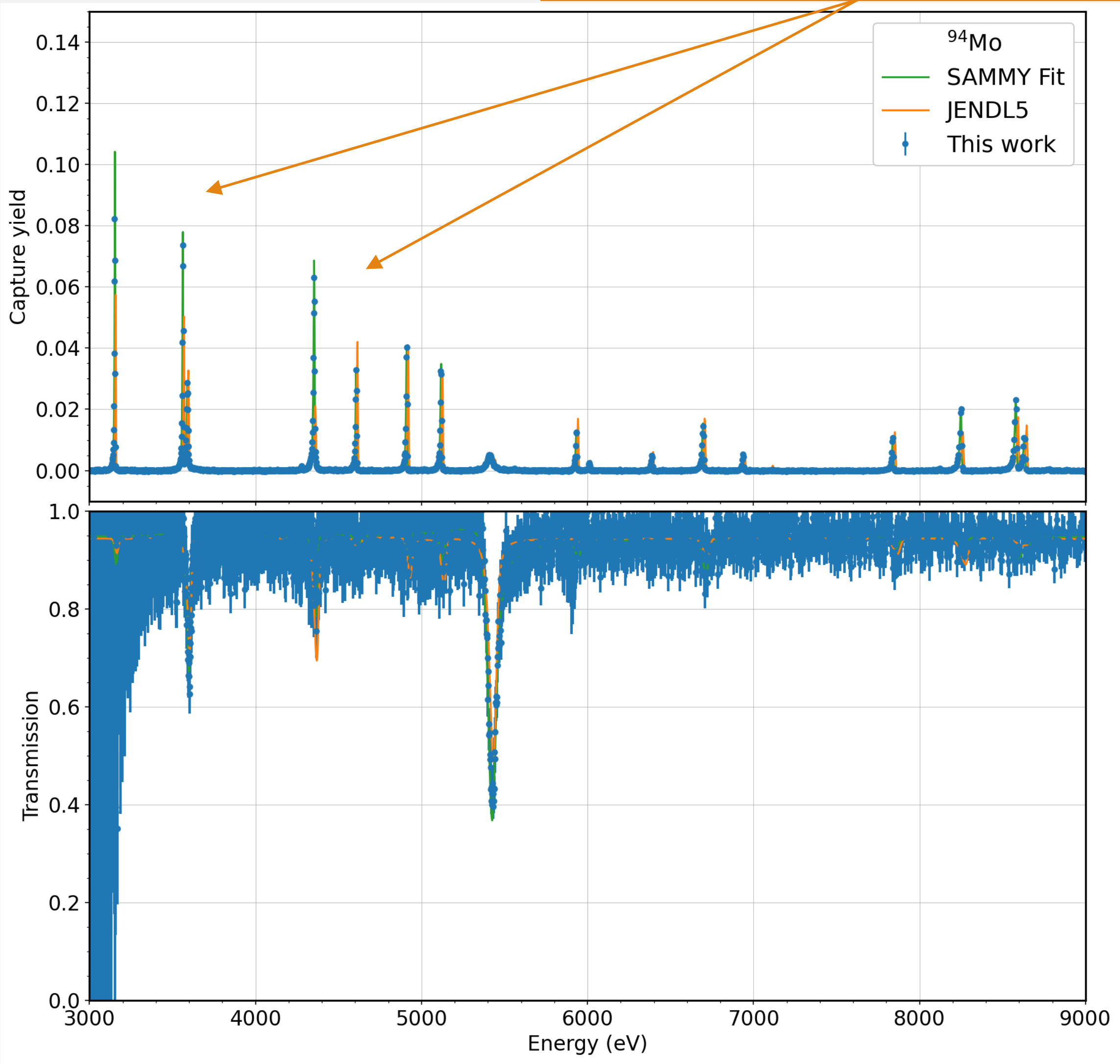




Preliminary resonance parameters ^{94}Mo

JENDL library doesn't reproduce the data accurately

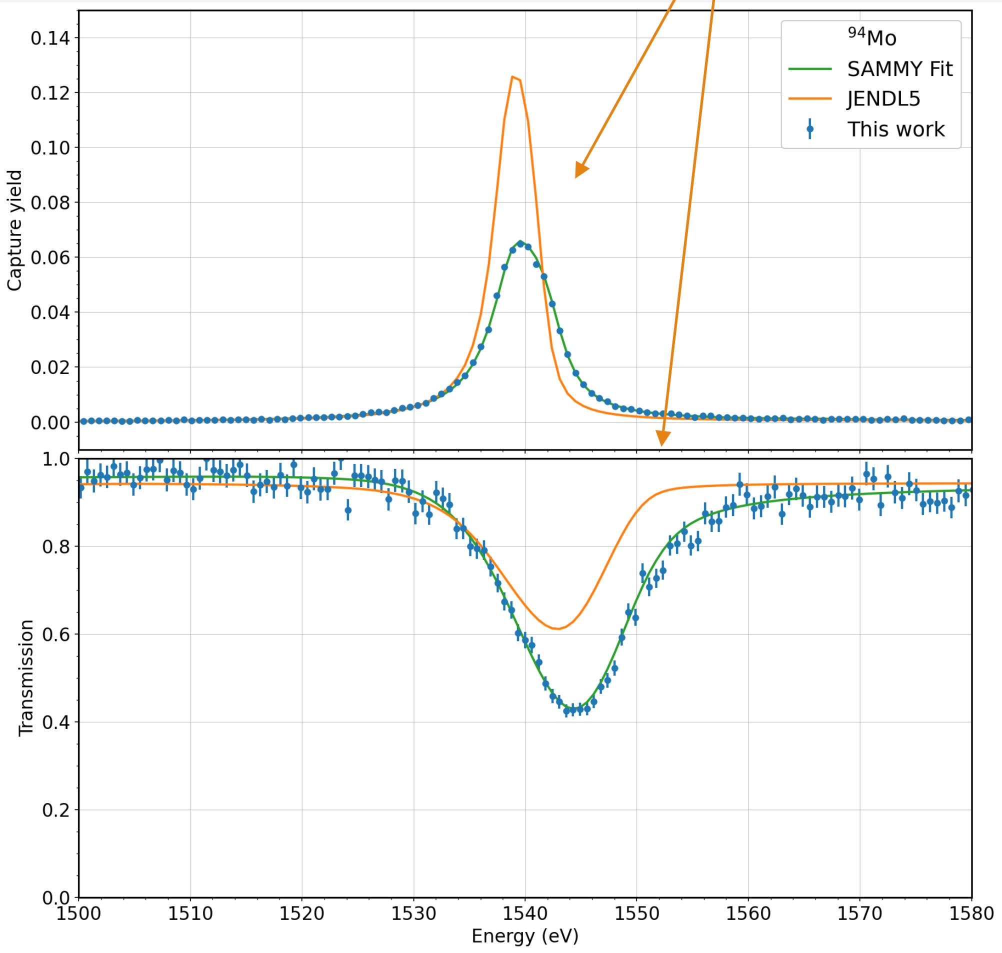




Preliminary resonance parameters ^{94}Mo

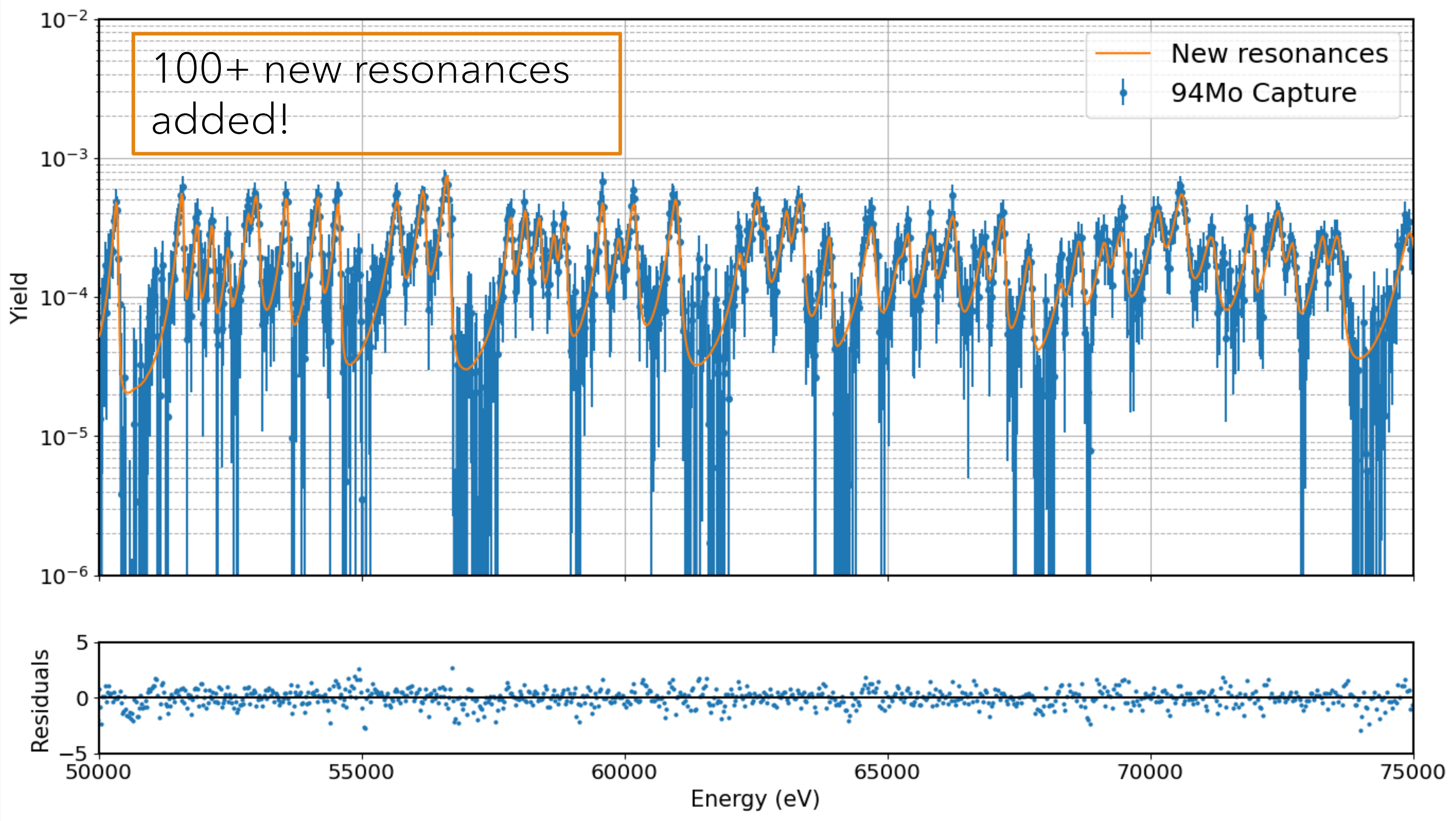
JENDL library doesn't reproduce the data accurately

- Resonance parameters have been adjusted in all the resolved resonance region (<21 keV);
- Extended resolved resonance region up to 75 keV;
- Example of fit showed here compared to the calculation performed with JENDL5 parameters;
- Good agreement between transmission and capture data with enriched samples.

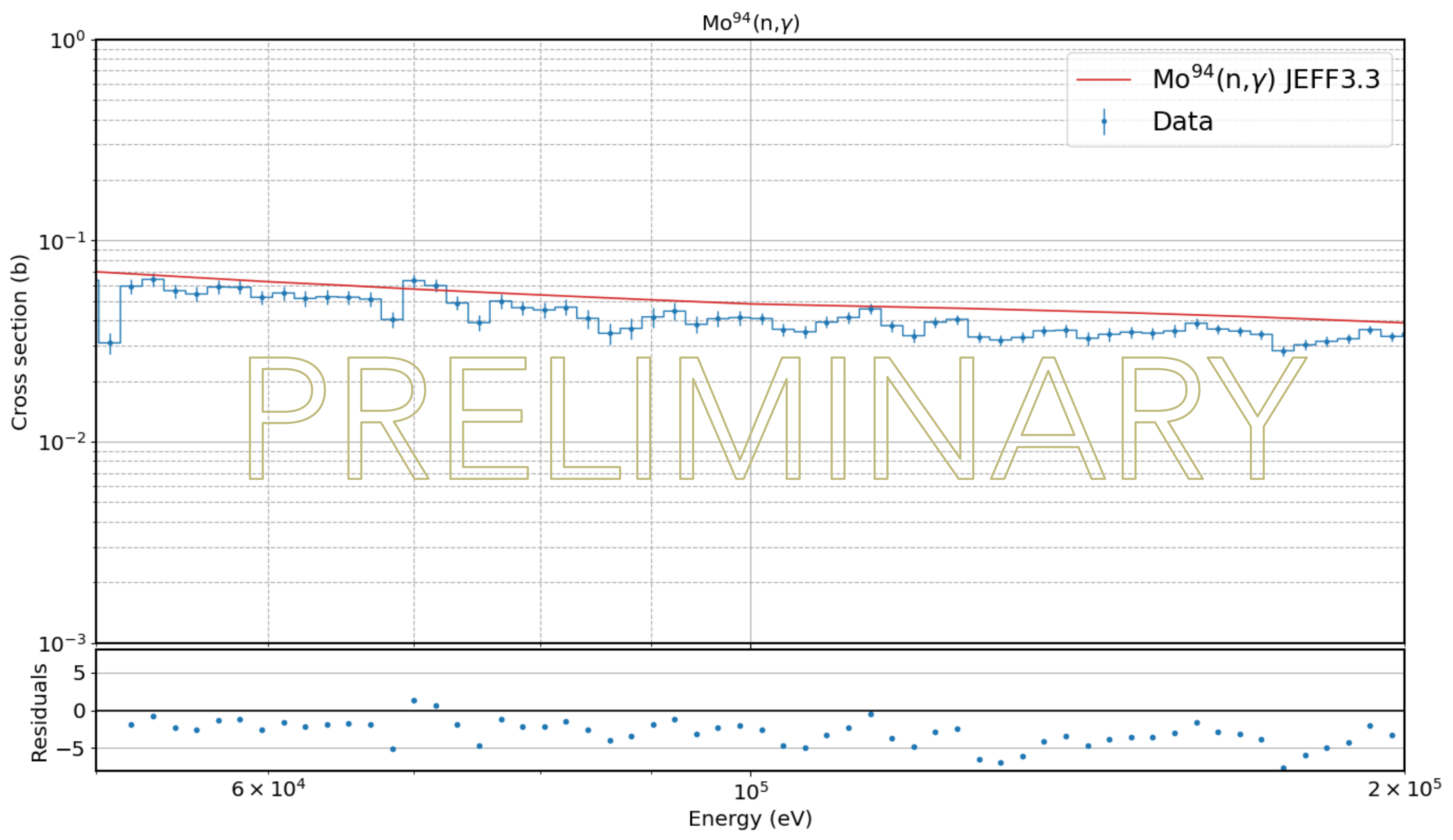


Preliminary resonance parameters ^{94}Mo

- Extended resolved resonance region up to 75 keV using data from capture measurements,
- New resonances not present in literature.

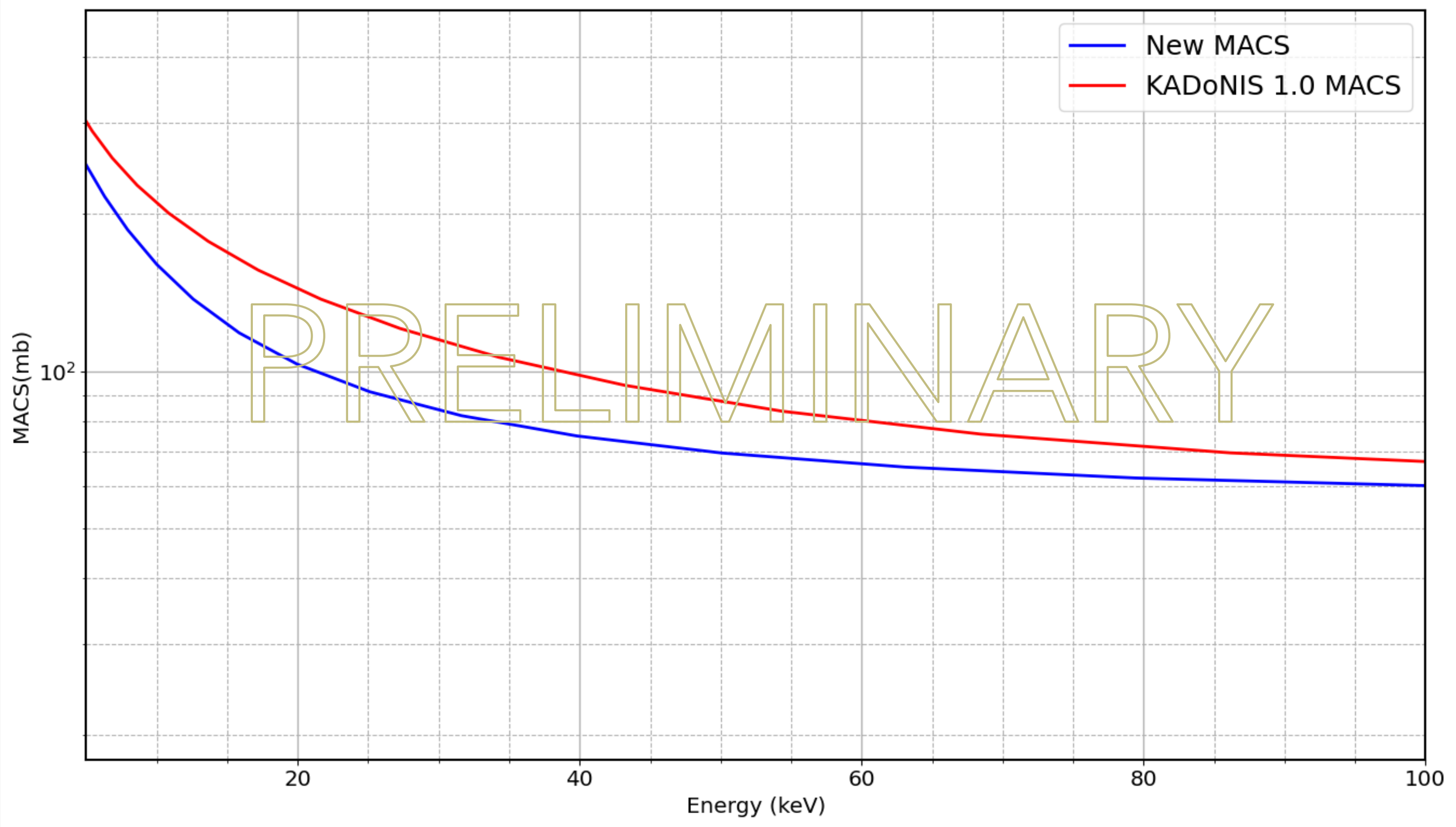


^{94}Mo Unresolved Resonance Region



- 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.

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%.

Conclusions

- Transmission and capture measurements were performed using highly isotopically enriched samples in $^{92,94,95,96,97,98,100}Mo$,
- Capture measurements performed at n_TOF shows good energy resolution up to tens of keV, with the possibility of extending the resolved resonance region,
- Preliminary results of resonance parameters for Mo shows some deviations with respect to data libraries (e.g. JENDL) but a general agreement with literature data,
- Neutron capture spectra of ^{97}Mo shows many resonances not present in latest evaluations,
- Preliminary resonance parameters on ^{94}Mo shows a reduction in the resonance kernels with respect to measurements in literature is observed,
- Preliminary MACS of ^{94}Mo shows a reduction with respect to KADoNIS 1.0.

Thank you for your attention!

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 847594 (ARIEL).

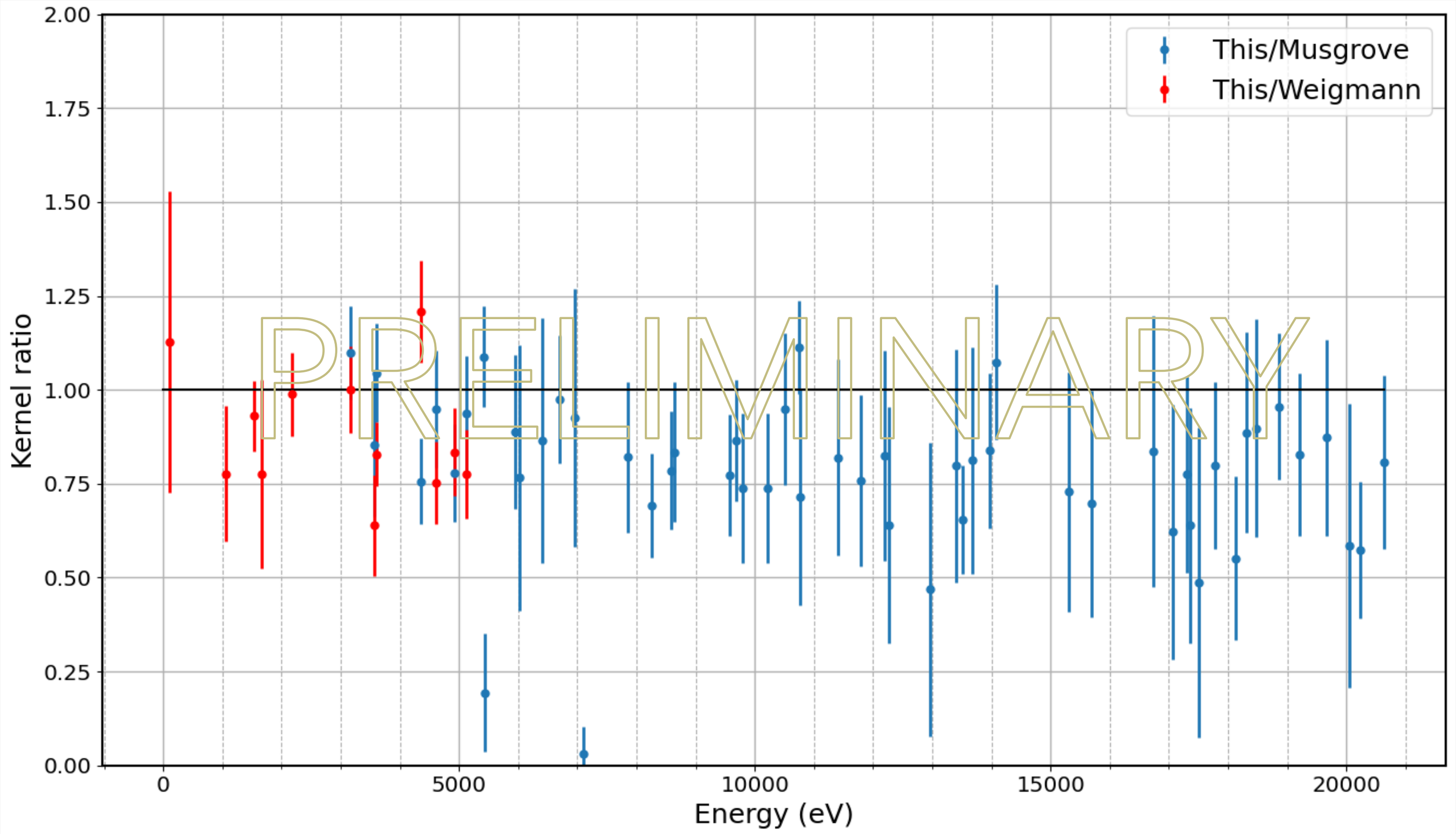
Backup

Mo pellet samples

Atomic %	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
^{94}Mo	0,63%	98,97%	0,36%	0,01%	0,01%	0,01%	0,01%
^{95}Mo	0,31%	0,69%	95,40%	2,24%	0,51%	0,65%	0,20%
^{96}Mo	0,28%	0,24%	1,01%	95,90%	1,00%	1,32%	0,25%

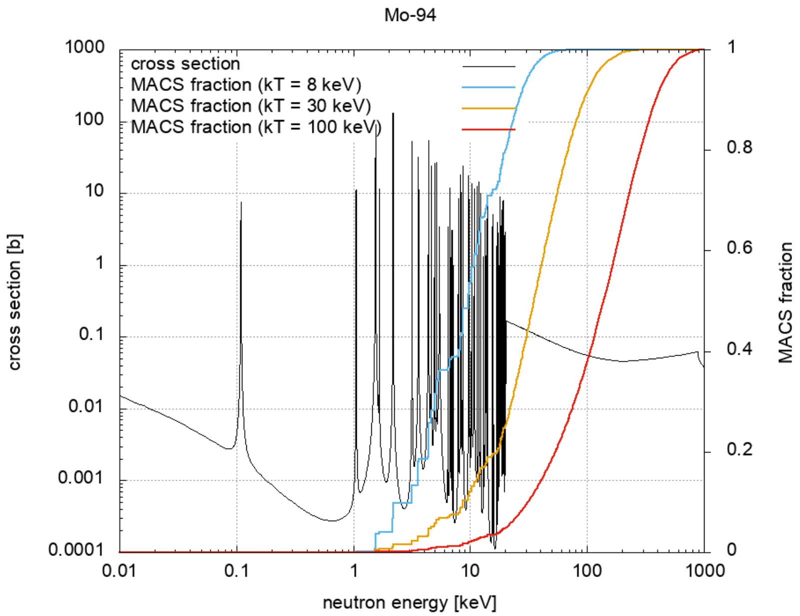
Isotope	Mass (g)	Areal density (atoms/b)
^{94}Mo	1,9526	3,9592E-03
^{95}Mo	1,9745	3,9558E-03
^{96}Mo	1,9175	3,8064E-03
^{nat}Mo-5 μm	2,014	4,0059E-03
^{nat}Mo-350 μm	1,989	3,9584E-03

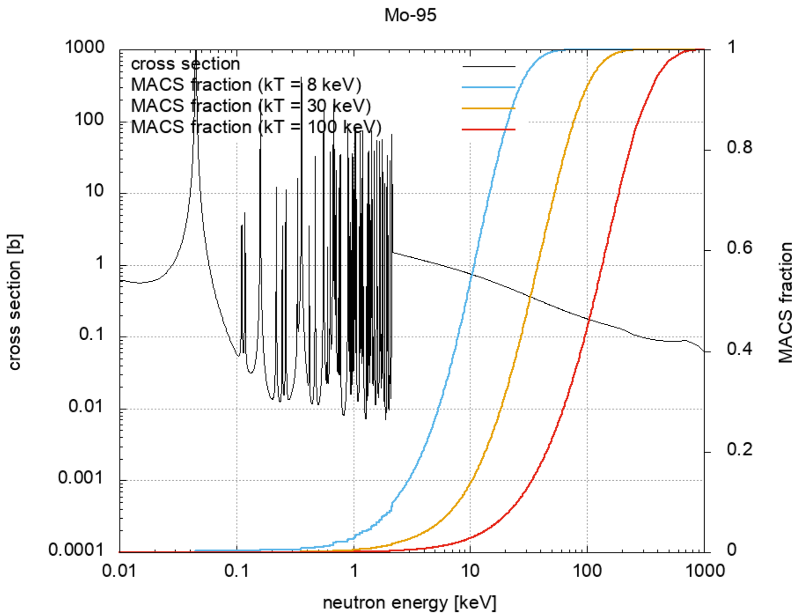
Kernel ratio with literature ^{94}Mo

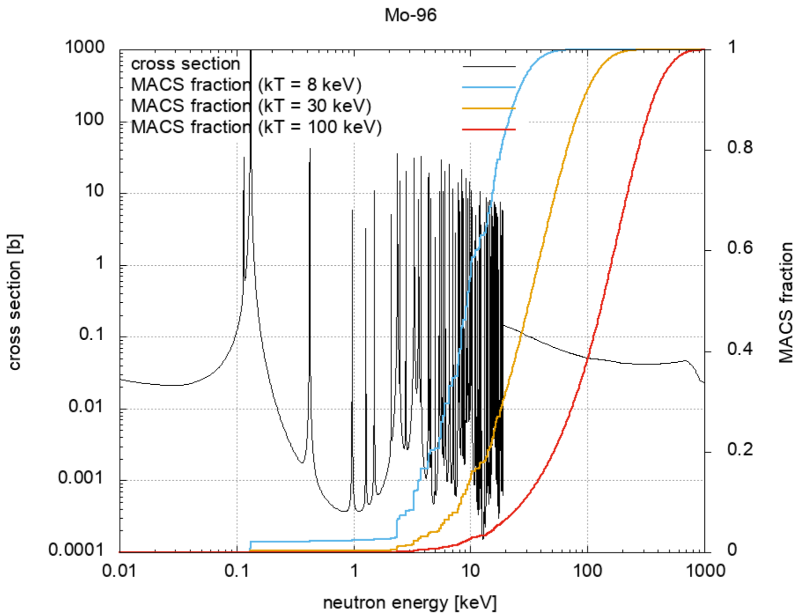


- The preliminary kernels obtained with SAMMY were compared to the ones in literature (Weigmann and Musgrove capture measurements);
- Main measurements used in libraries.

MACS fractions







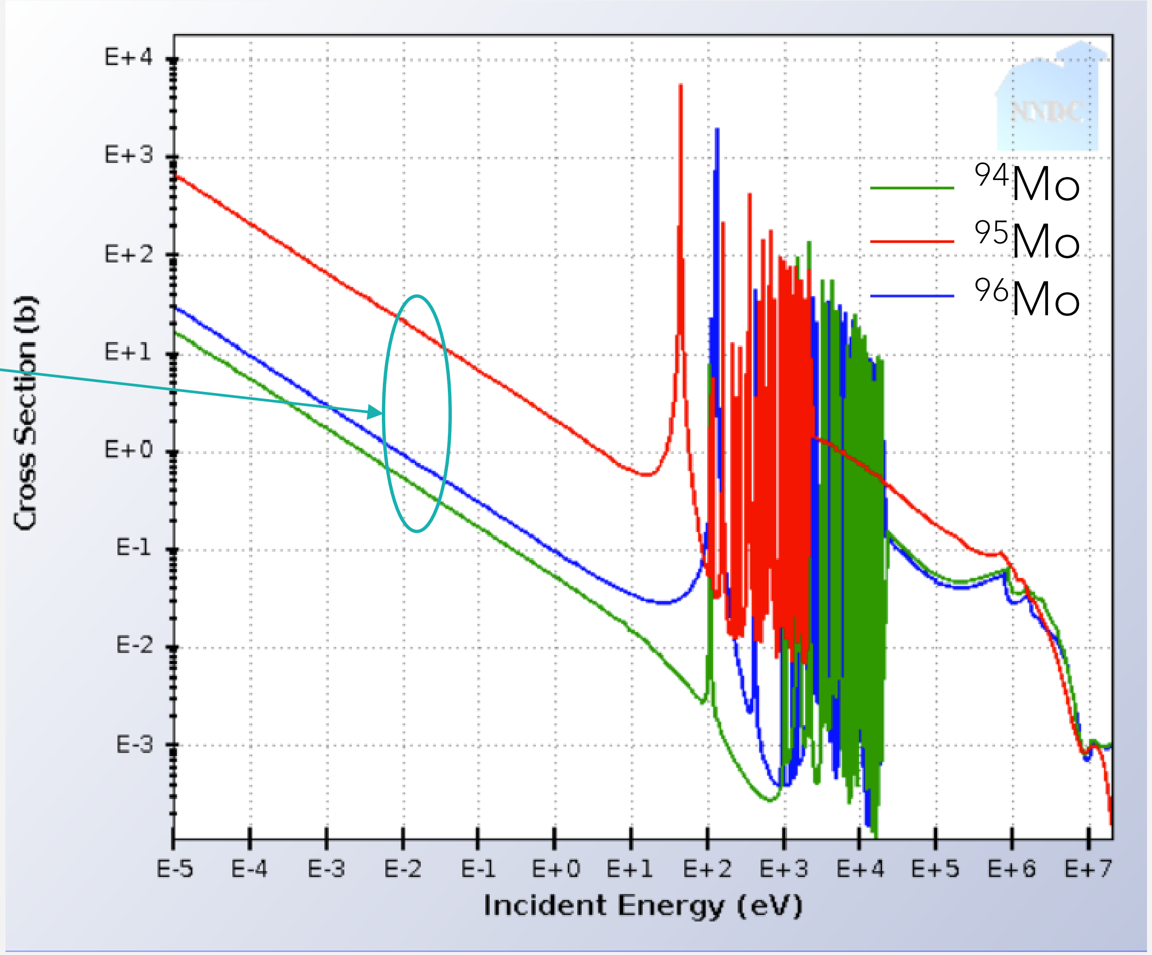
Objective of experiments

Improve capture cross section accuracy for neutron energies from thermal (10 meV) to hundreds keV

Submit results to EXFOR to improve nuclear data libraires (ENDF, JEFF, JENDL ecc.)

Capture cross section ENDF/B-VIII

Thermal cross section:
^{94}Mo ~ 350mb
^{95}Mo ~ 13b
^{96}Mo ~ 620mb



Experimental measurements

EAR2_2021	EAR1_2022	EAR2_2022
1.7 10^{18} protons	6.0 10^{18} protons	1.7 10^{18} protons
3 B6D6, 1 L6D6, 1 STED	4 C6D6	8 STED, 2 L6D6, 1 DSTI
Powder sample in aluminum canning	Pressed pellets in plastic bags	Pressed pellets in plastic bags

+ additional transmission measurement with enriched pellets at 10m station of GELINA
+transmission measurements with natural samples at 50m station of GELINA

Time-of-flight technique

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

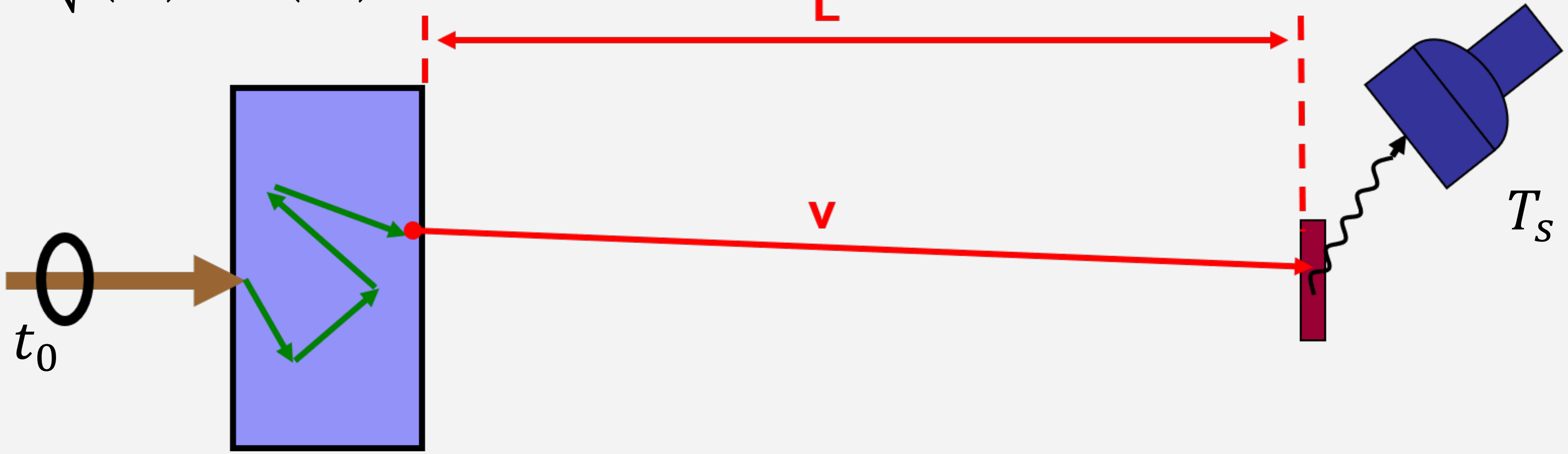
$$v = \frac{L}{t}$$

Flight path

Time-of-flight

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

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



Experimental measurements

Transmission

Percentage of neutrons that traverses a samples without interacting with it

- Related to total cross section:

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

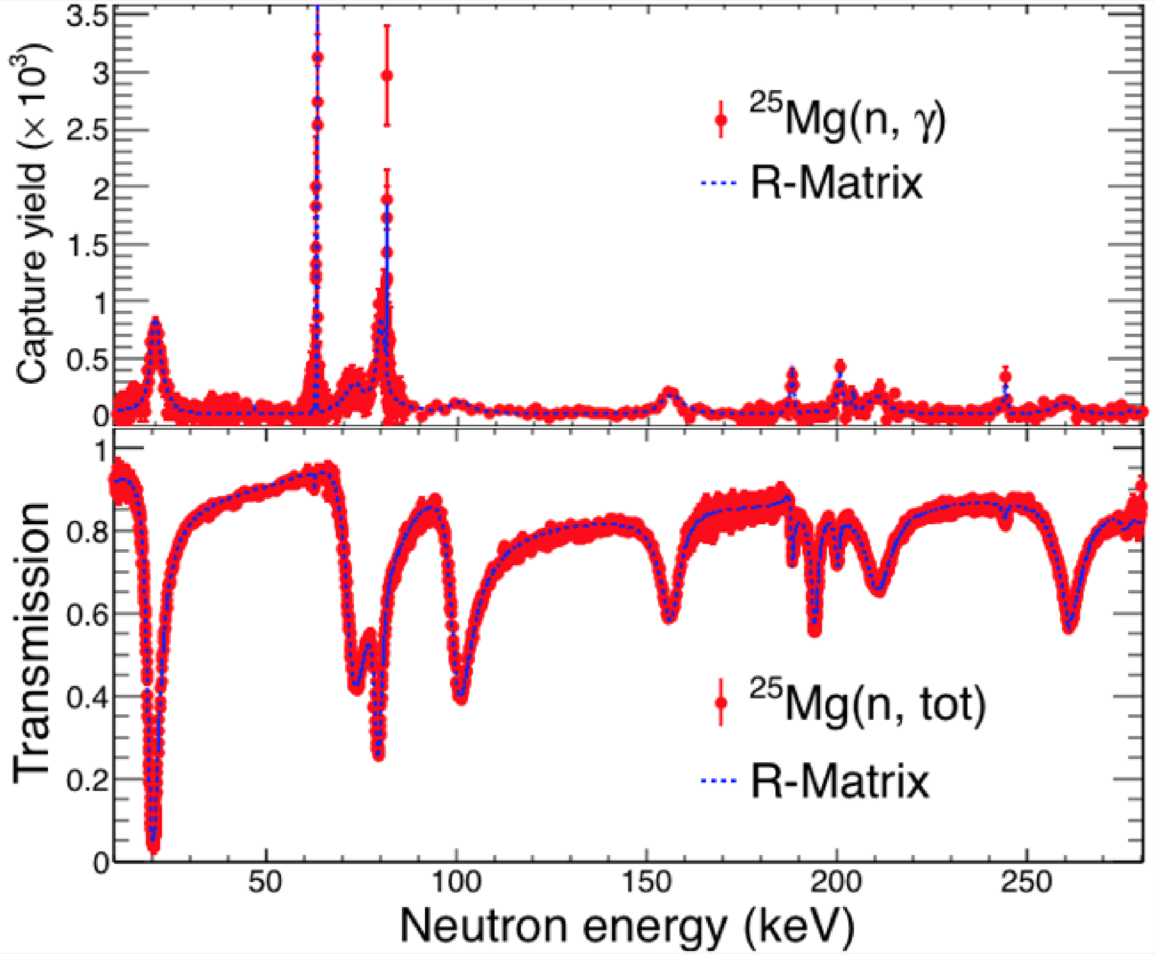
Radiative capture (capture yield)

Percentage of neutrons that undergoes capture reaction in the sample

- Related to capture cross section via:

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

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)

^{94}Mo preliminary resonance parameters

J	L	Energy (eV)	Unc_E	Γγ(meV)	Unc_Cap	Γn (meV)	Unc_ n
-0.5	1	108.7365	2.29E-03	158.837	4.69049	0.180556	1.22E-03
-1.5	1	1051.963	1.48E-02	237.578	25.6533	2.35311	3.02E-02
0.5	0	1542.773	1.16E-02	124.952	0.568967	1673.86	8.59281
-1.5	1	1657.322	2.08E-02	169.781	30.3225	4.65519	6.62E-02
-1.5	1	2175.49	1.01E-02	159.592	1.06928	340.652	4.81211

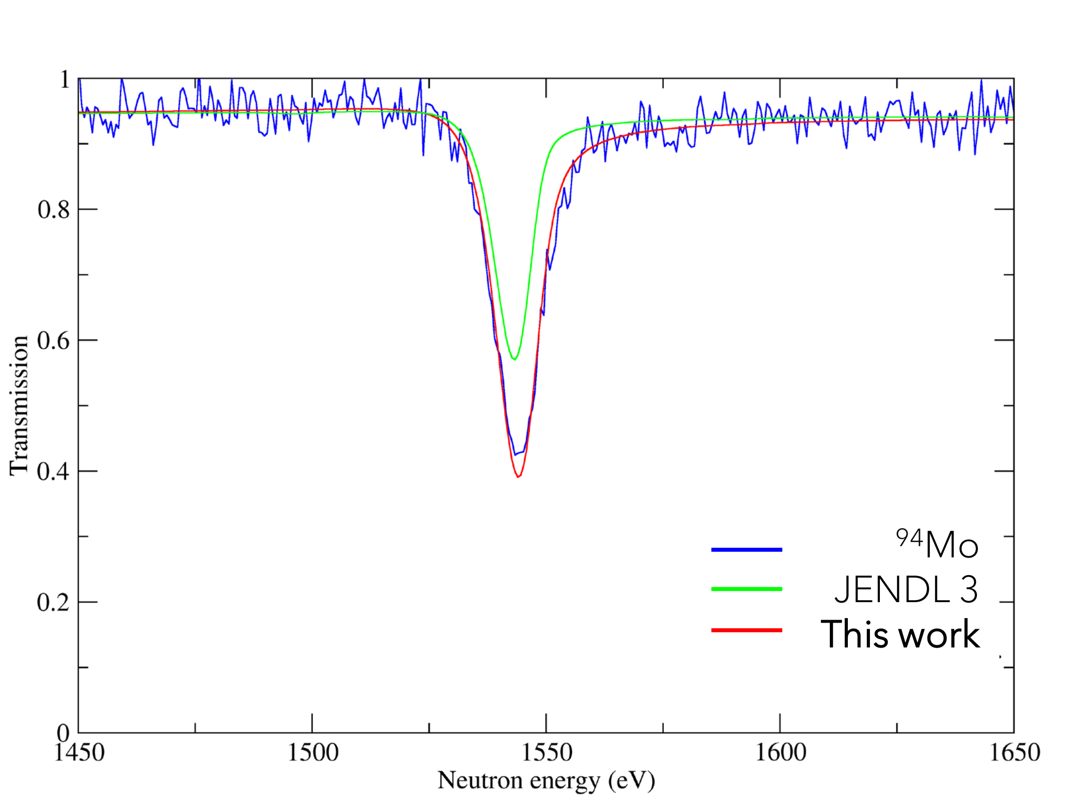
⋮

-1.5	1	9576.481	0.109357	122.857	2.46143	673.324	68.231
0.5	0	9689.416	0.184379	98.0503	2.40078	2383.27	162.983
-1.5	1	9797.066	0.132802	95.4524	7.68889	230.418	44.3515

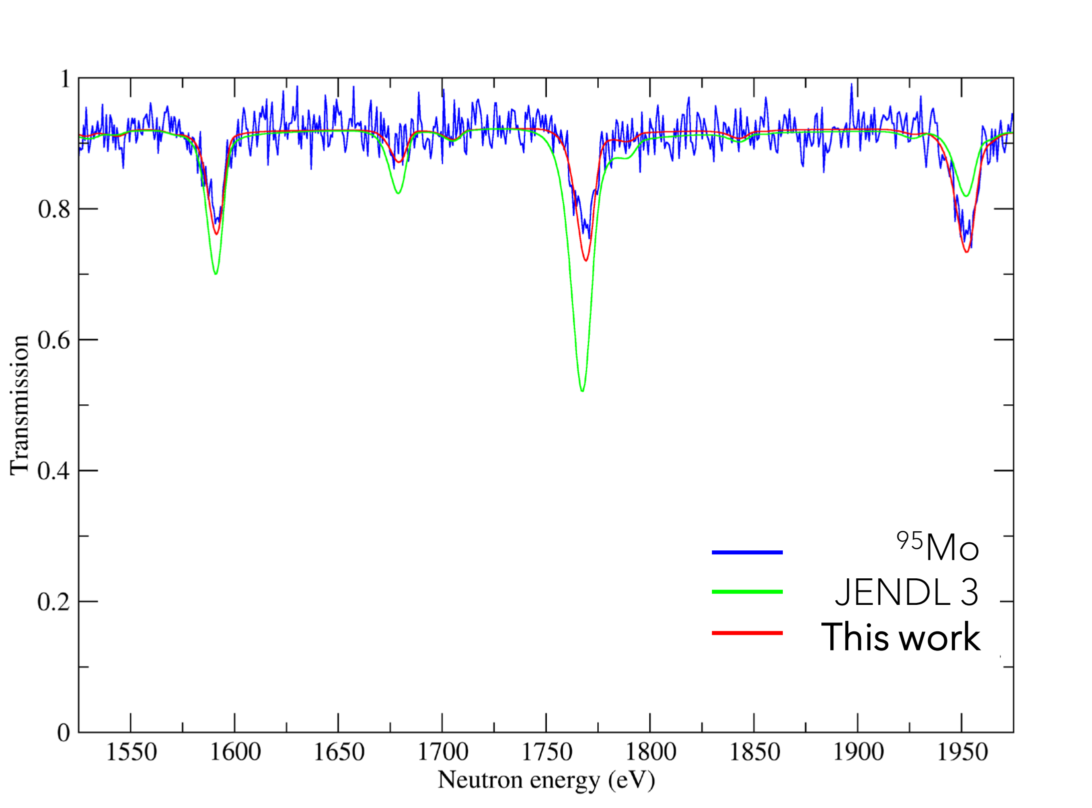
Transmission with enriched Mo

Transmission at 10 m station of GELINA

^{94}Mo transmission

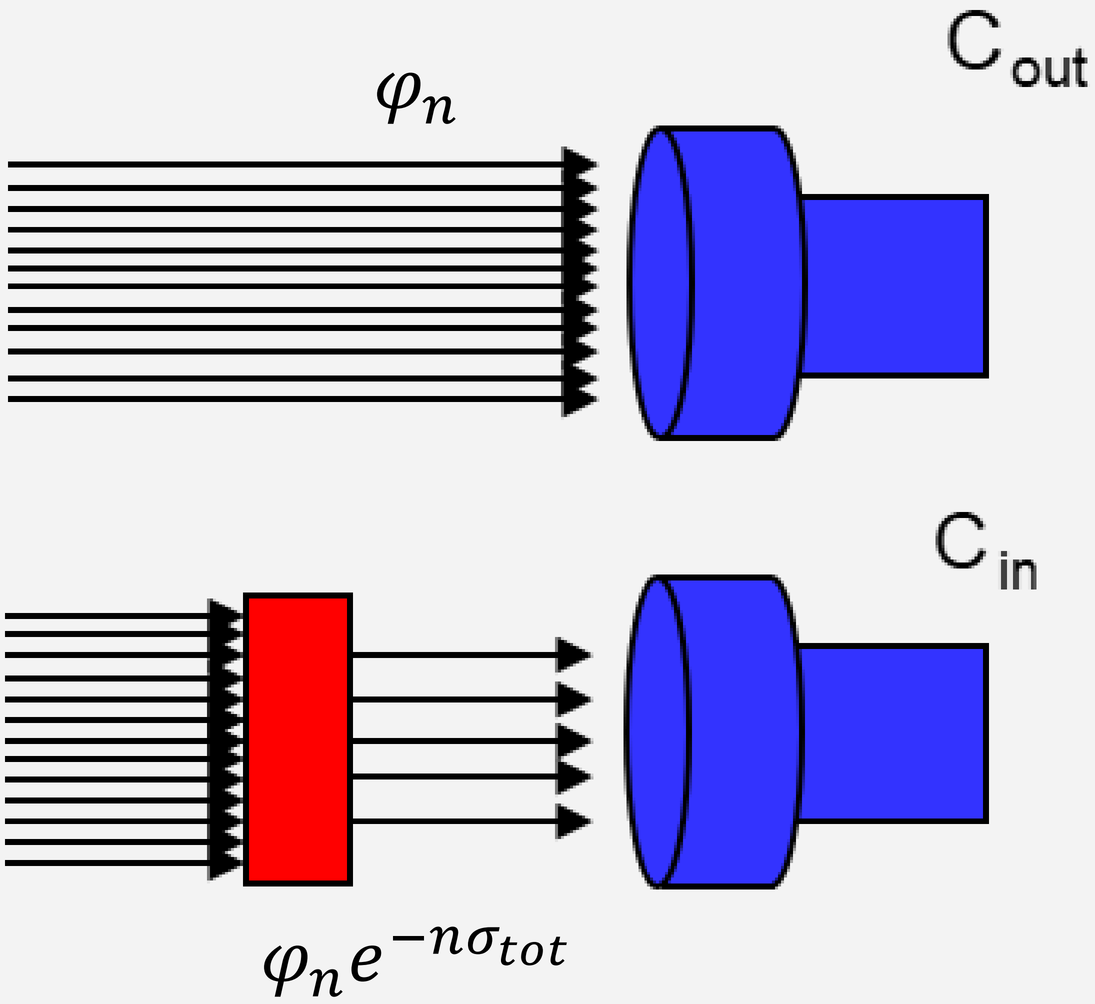


^{95}Mo transmission



Transmission with enriched samples confirm RP file!

Transmission



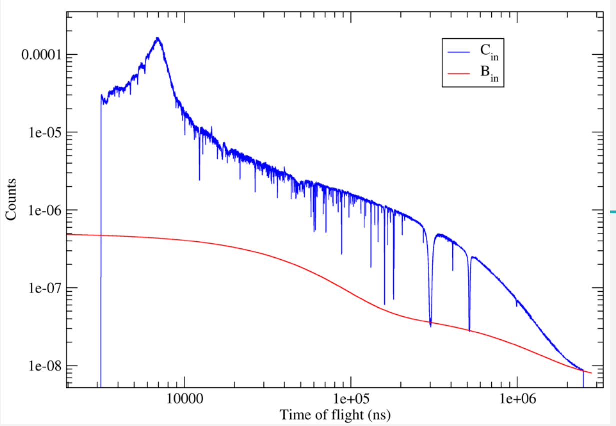
Percentage of neutrons that traverses a sample without interacting with it

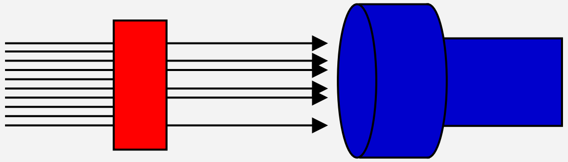
$$T = \mathrm{N}\frac{C_{in}(t) - KB_{in}(t)}{C_{out}(t) - KB_{out}(t)} = \frac{\varphi_n e^{-n\sigma_{tot}}}{\varphi_n} = e^{-n\sigma_{tot}}$$

- 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)

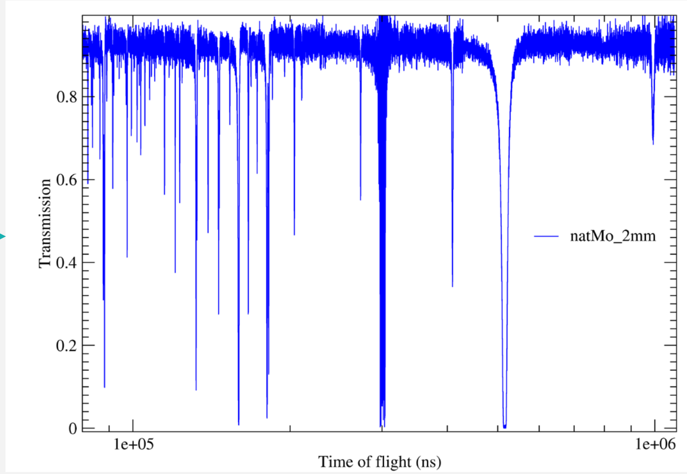
Transmission spectrum

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

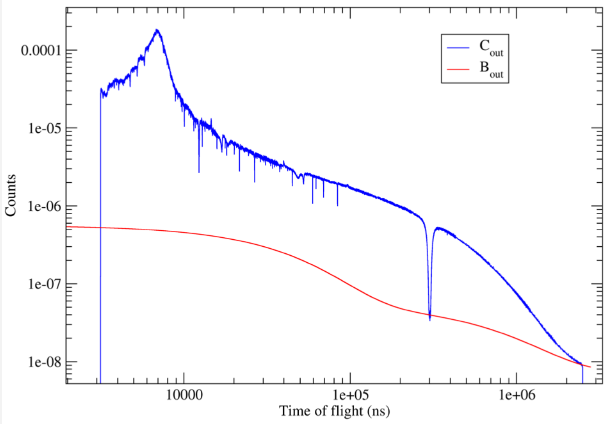


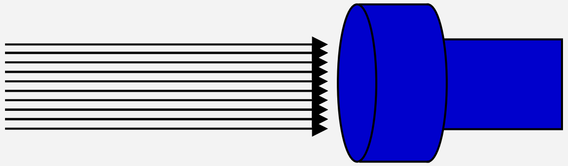


$$T_{exp} = N \frac{C_{in} - KB_{in}}{C_{out} - KB_{out}}$$



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





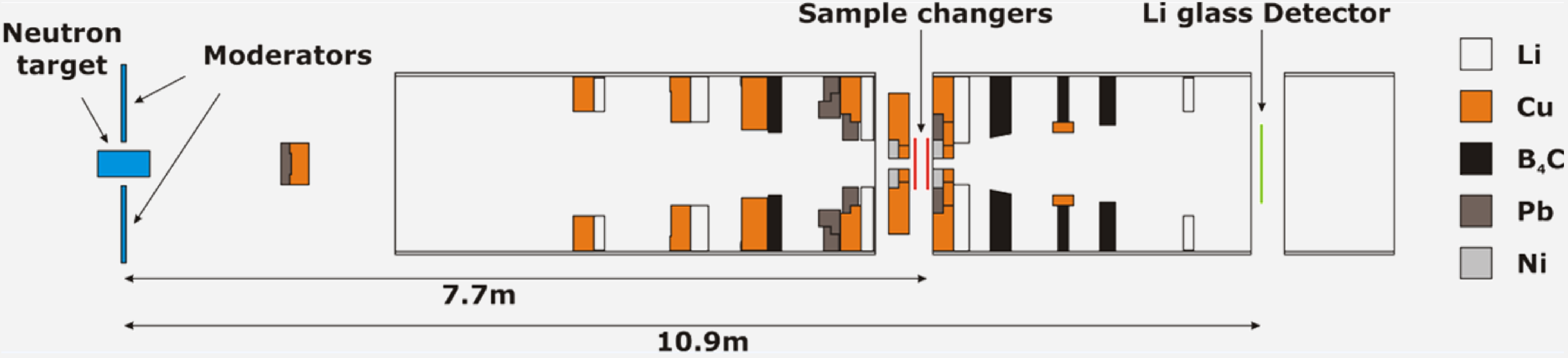
Transmission measurements

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

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

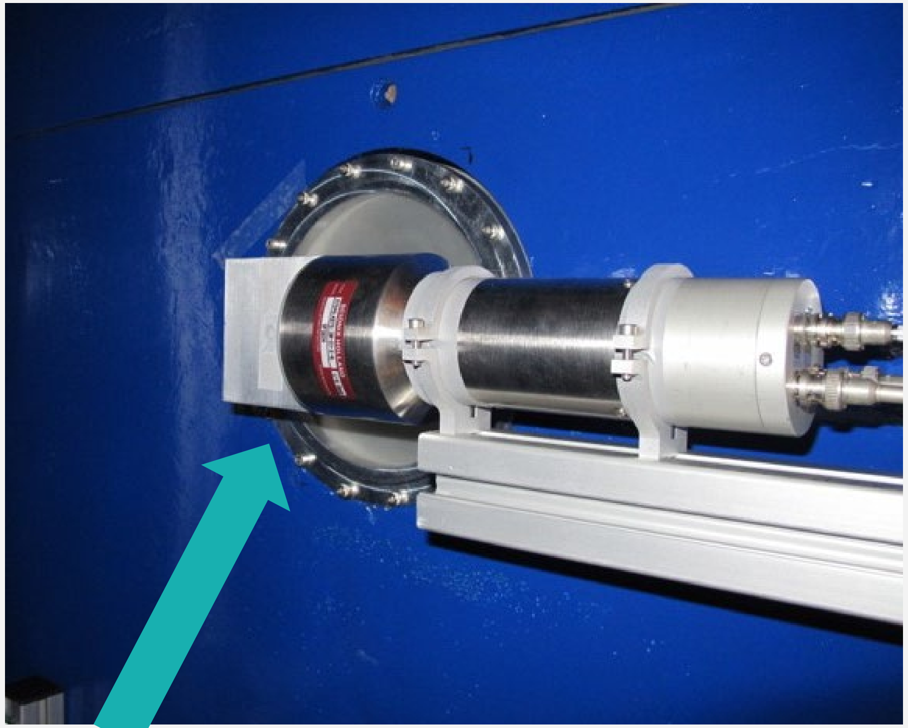
$\Rightarrow$ Good transmission geometry (collimation)

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



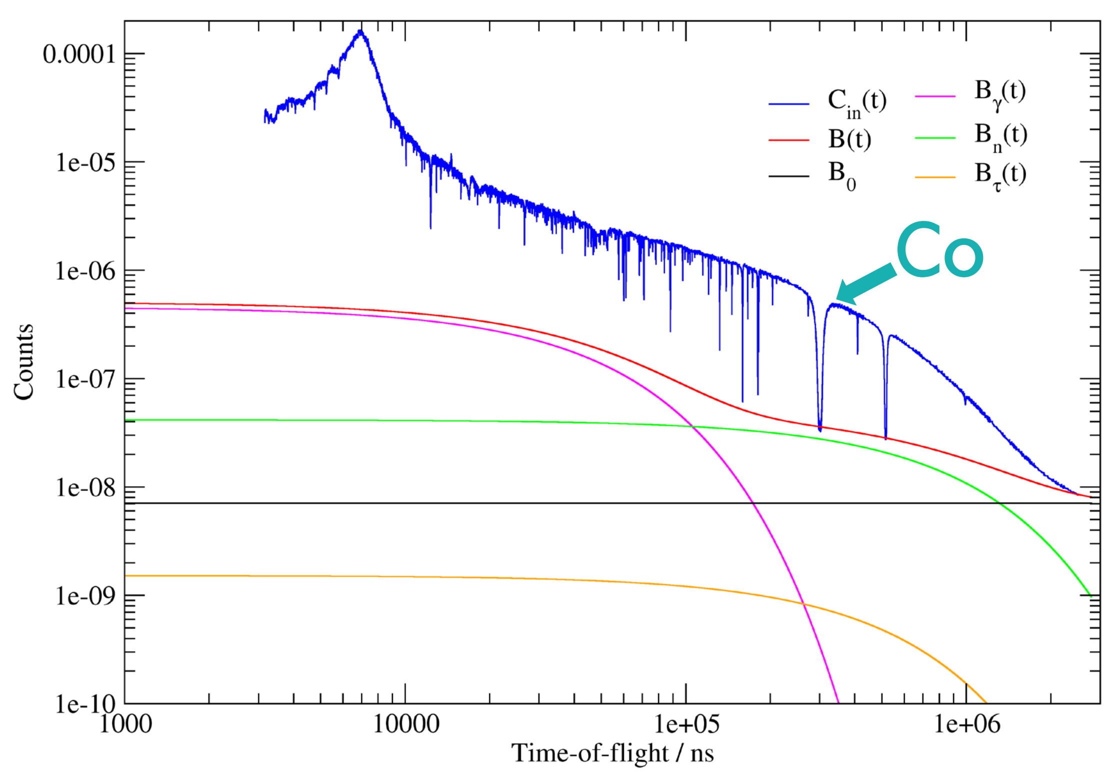
Scintillator

Castle

Background

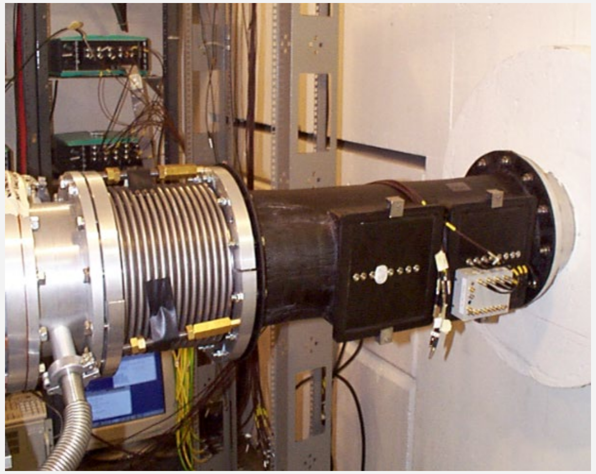
$$B(t) = b_0 + b_1 e^{-\lambda_1 t} + b_2 e^{-\lambda_2 t} + b_3 e^{-\lambda_3 (t+\tau_0)}$$

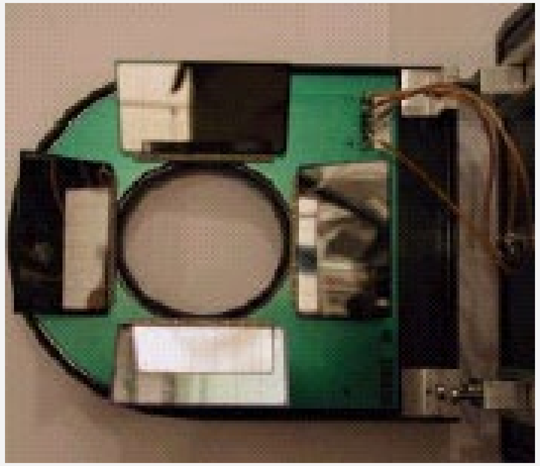
- b_0 time independent background
- $b_1 e^{-\lambda_1 t}$ neutron capture in hydrogen of moderator
- $b_2 e^{-\lambda_2 t}$ neutrons scattered inside the detector station
- $b_3 e^{-\lambda_3 (t+\tau_0)}$ neutron from previous cycle ($\tau_0 = 1/f$)

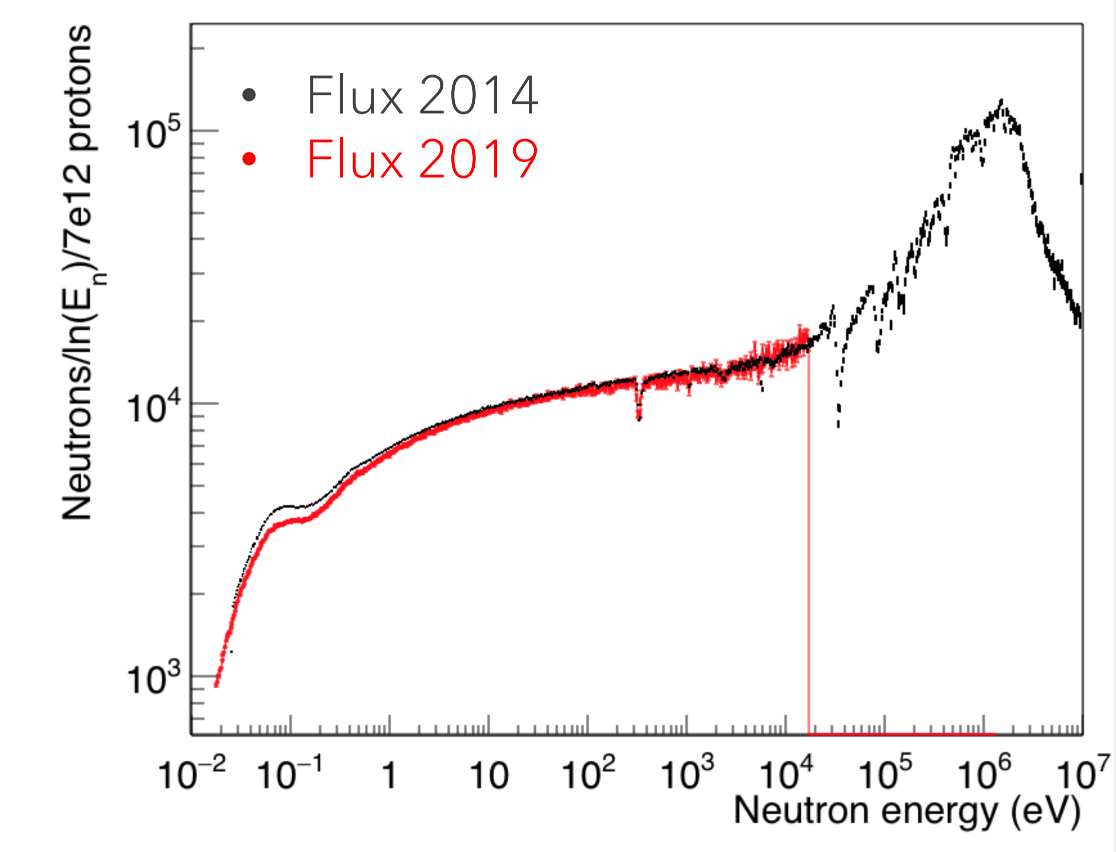


Neutron flux monitor

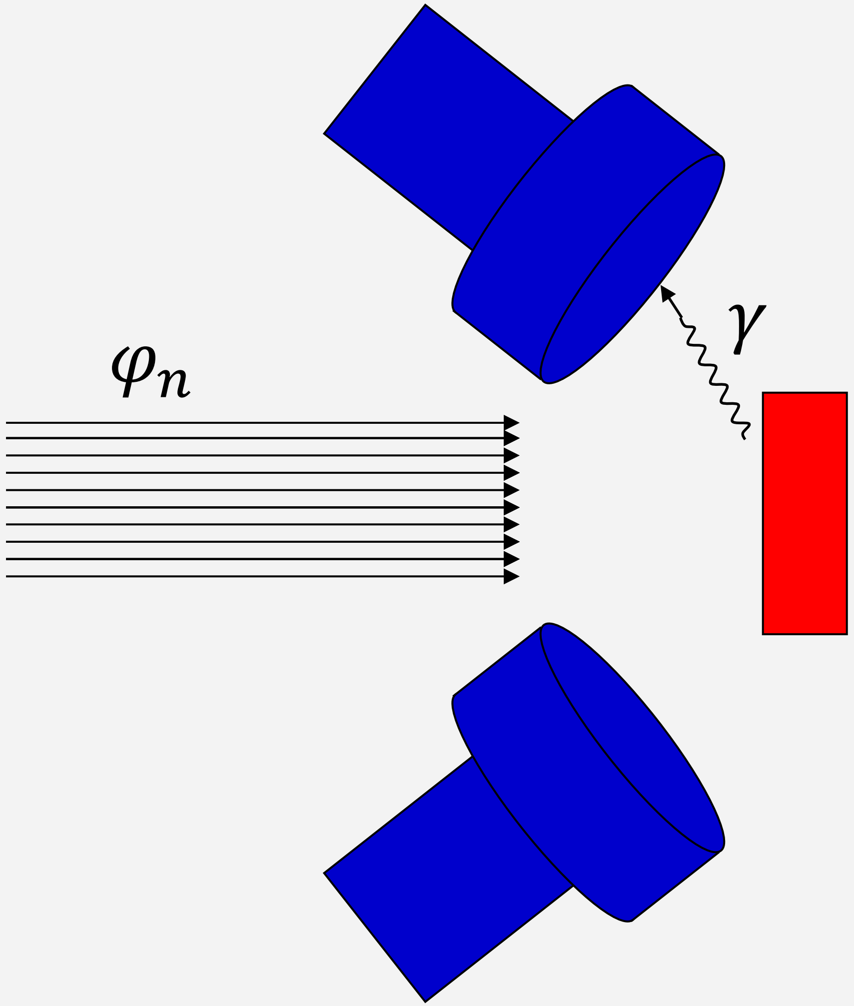
- Neutron flux continuously monitored;
- SiMON (Silicon MONitor) in beam;
- Silicon detectors facing mylar foil coated in lithium;
- Minimal reduction of neutron flux.







Radiative capture



Experimental observable is capture yield

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

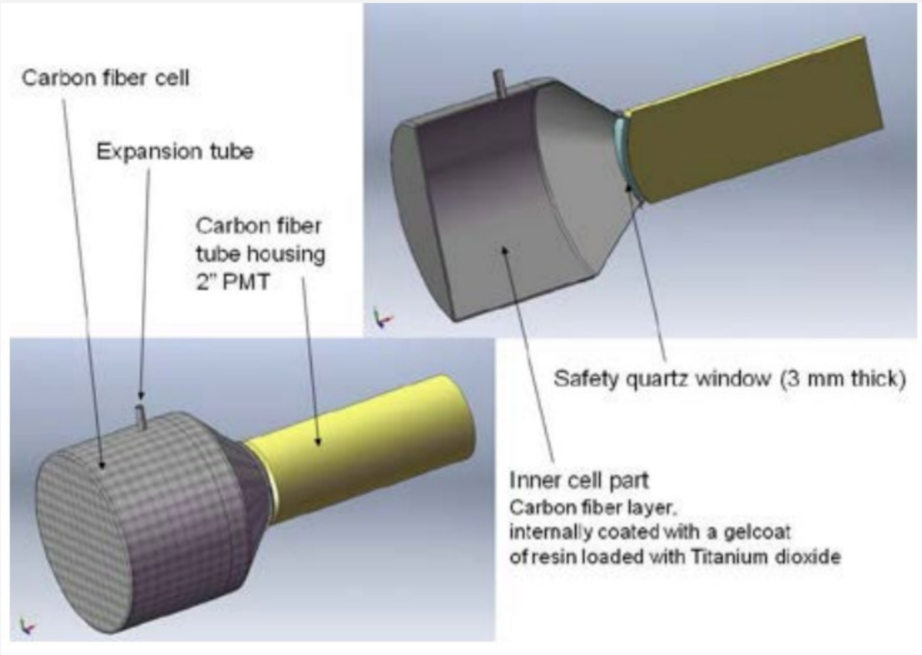
$$\mathrm{Y}_{exp} = \mathrm{N}\frac{C_\gamma(t) - B_\gamma(t)}{C_\varphi(t) - B_\varphi(t)}\mathrm{Y}_\varphi = (1-T)\frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}}$$

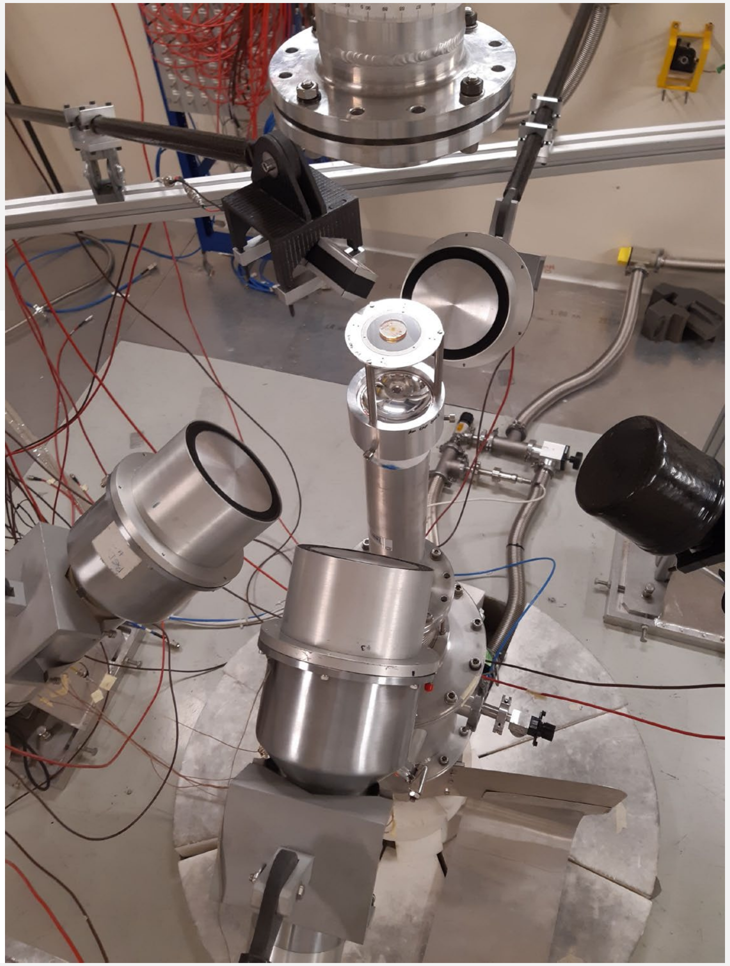
- Normalization factor energy and nuclide independent, obtained with Au measurement
- Background obtained with additional measurement (empty, lead)

Capture detectors

C6D6 detectors

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



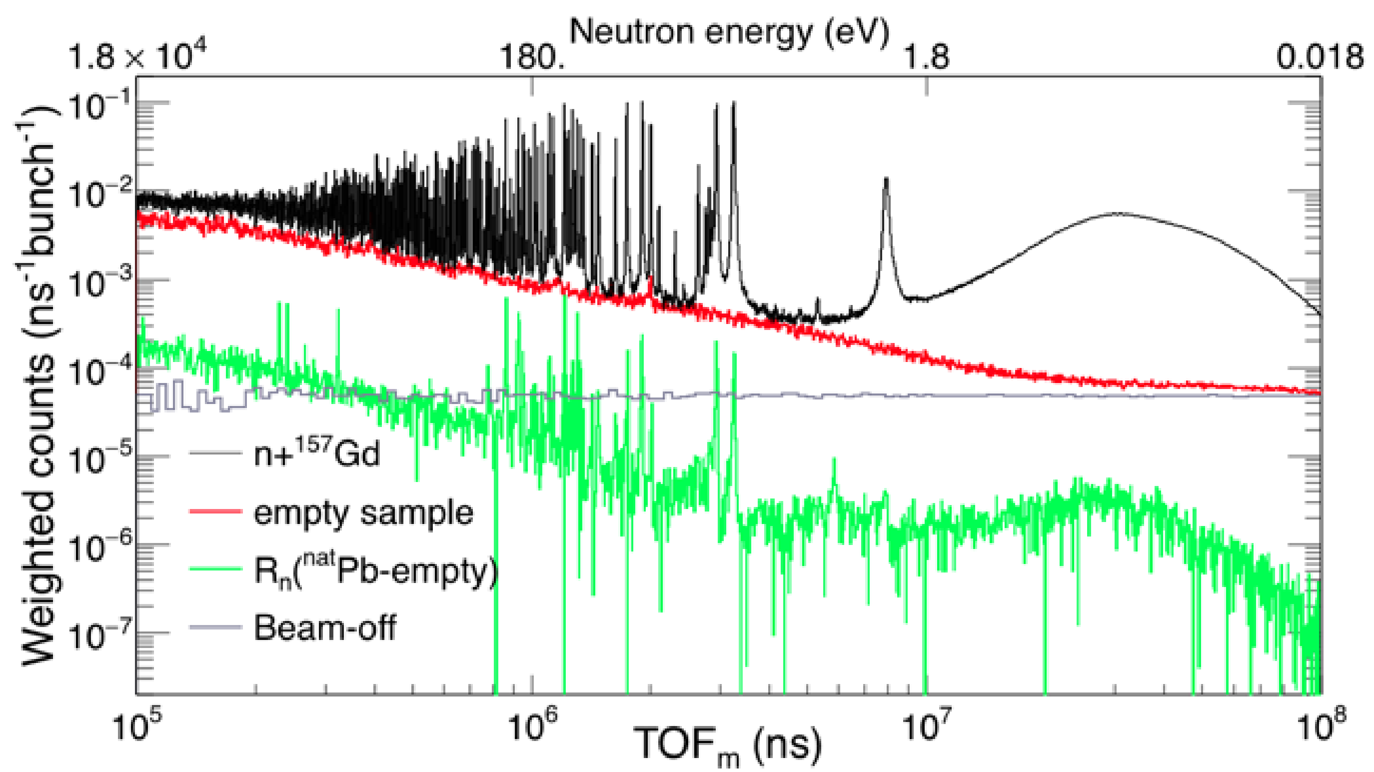


Background

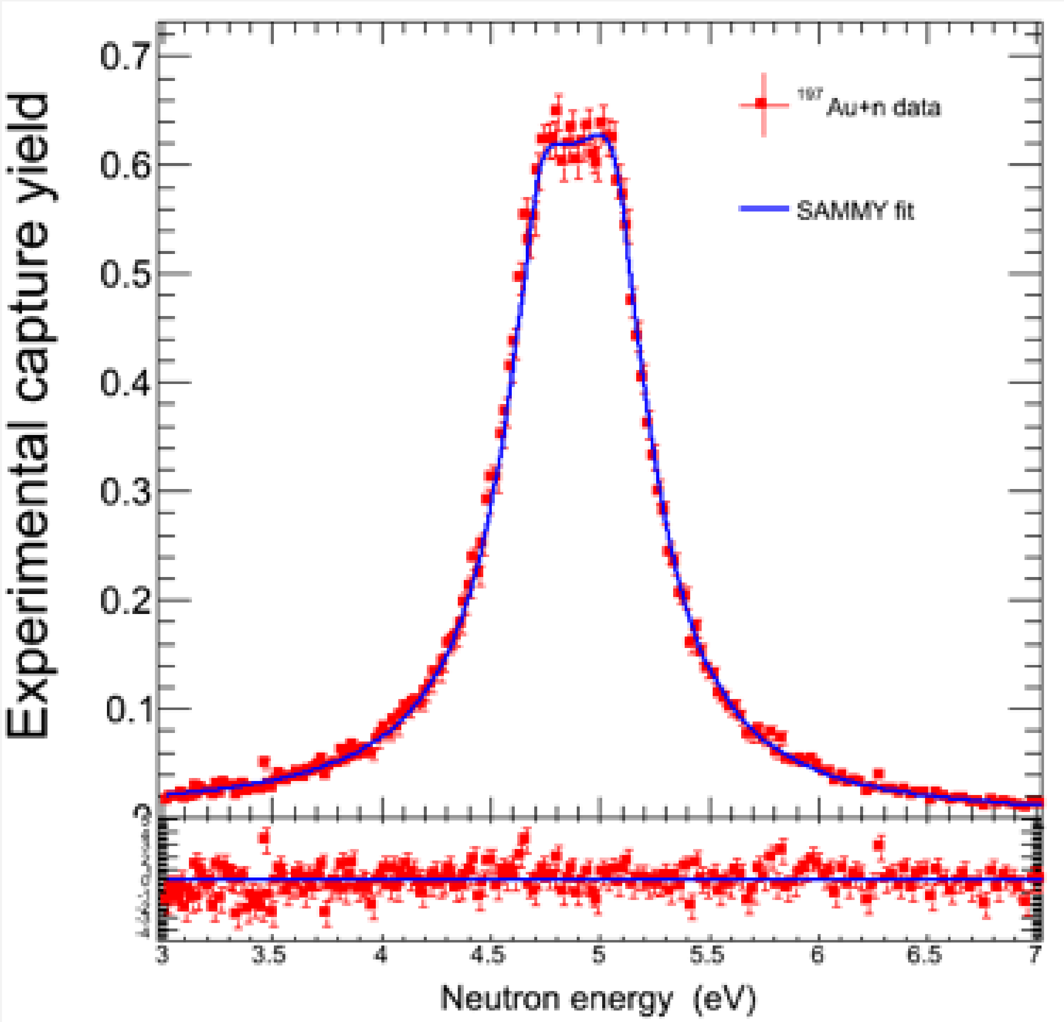
$$B(t) = a_0 + a_1 C_{OB} + a_2 R_n (C_{Pb} - C_{OB})$$

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



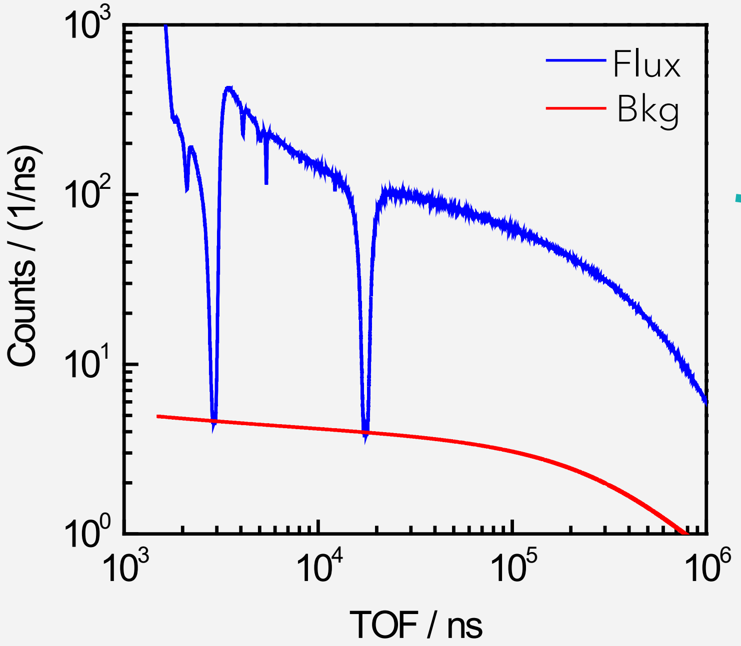
Normalization



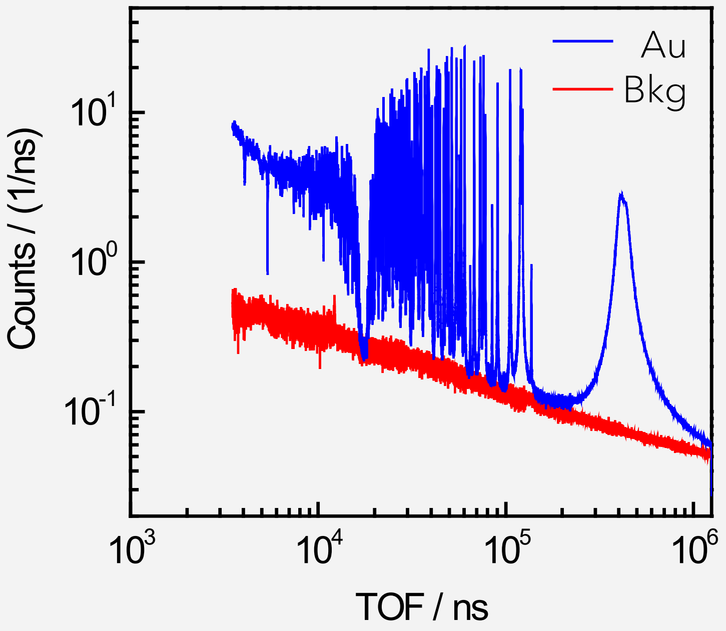
- For a capture resonance with $\Gamma_\gamma \gg \Gamma_n$ the capture cross section is approximately equal to the total cross section $\frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}} \approx 1$
- A saturated resonance $(n\sigma_{tot} \gg 1)$ absorbs all the impinging neutrons T≈ 0
- When both conditions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

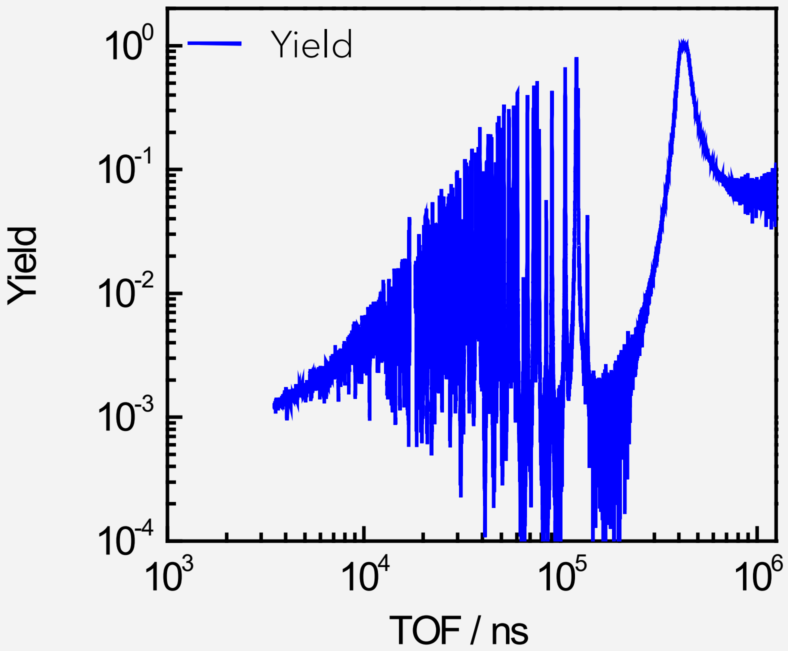
Extract normalization factor from ^{197}Au saturated resonance

Capture yield



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





RP compilation from literature

1) Define consistent energy scale:
   Weigmann et al. (1971) (capture experiments at GELINA)

2) Select $g\Gamma_n$ reference:
   E < 2keV: Leinweber et al. (2010)
   E > 2keV: Whynchank et al (1968)

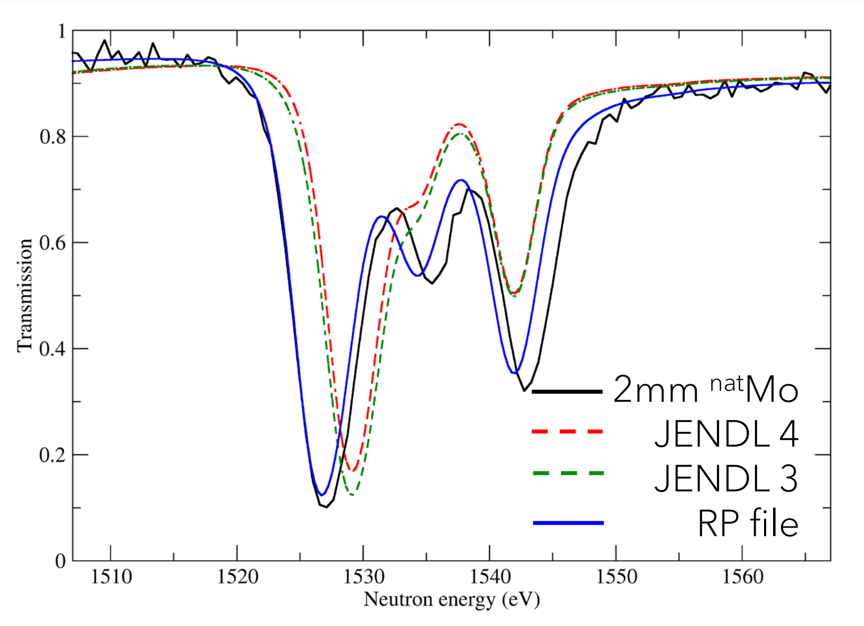
3) Select $\frac{g\Gamma_\gamma\Gamma_n}{\Gamma}$ reference:
   Weigmann et al. (1971)
   Musgrove et al. (1976) for odd isotopes and E>3keV

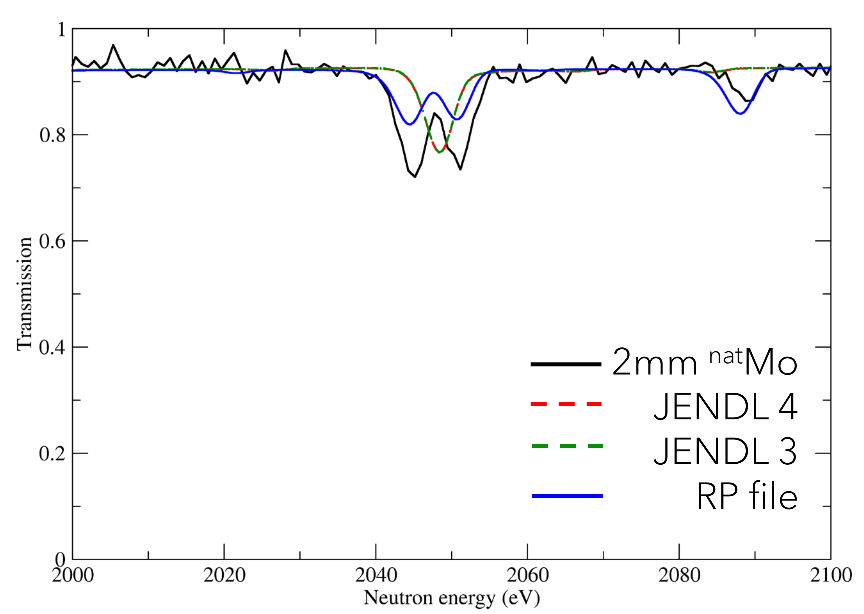
All isotopes up to 25 keV

Consistent parameters

- Compilation of RP file from literature data
- ^{nat}Mo transmission measurements at GELINA to validate and improve RP file

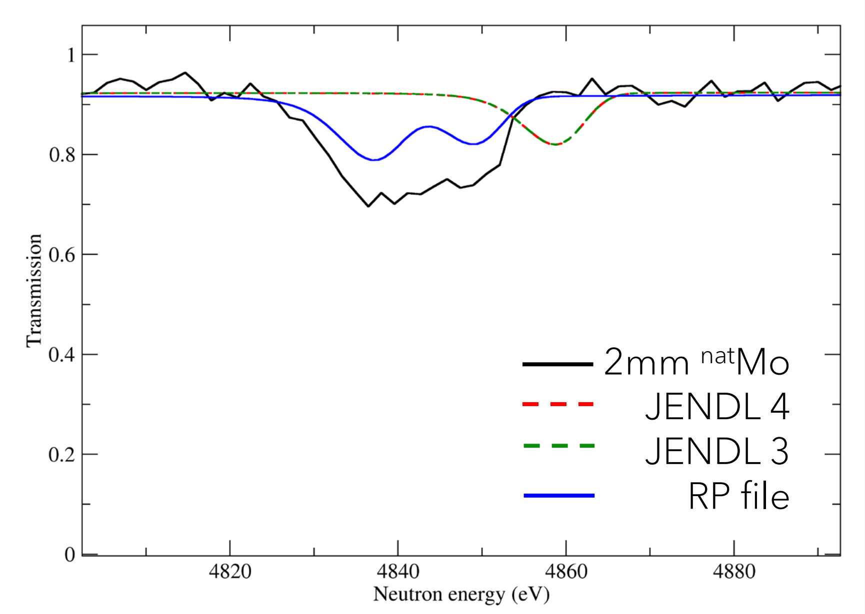
Validation of compiled RP file





- RP file verified by transmission data (50 m) of 2mm and 5mm thick ^{nat}Mo samples
- Missing resonances in libraries reported in literature data
- Literature parameters more consistent with transmission data

New RP file improve data description.



Pulse Height Weighting Technique (PHWT)

Use of detectors with small detection efficiency proportional to γ-ray energy

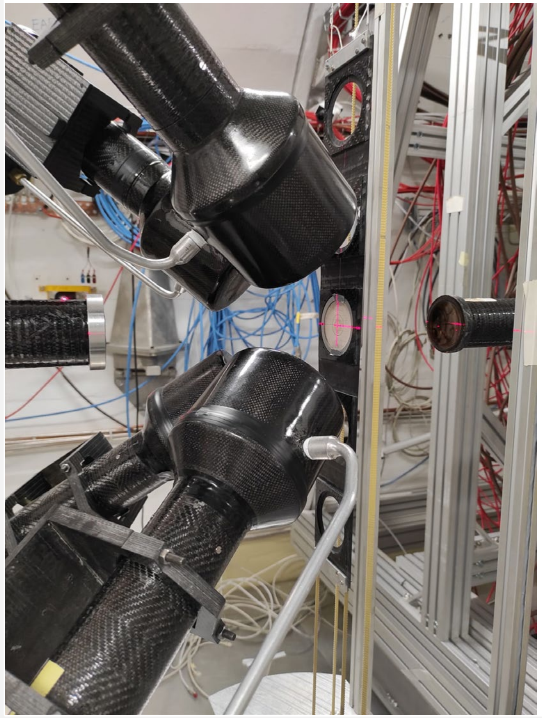
Capture efficiency proportional to cascade energy

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

Experimental conditions @ EAR1

DETECTION SETUP



Setup:

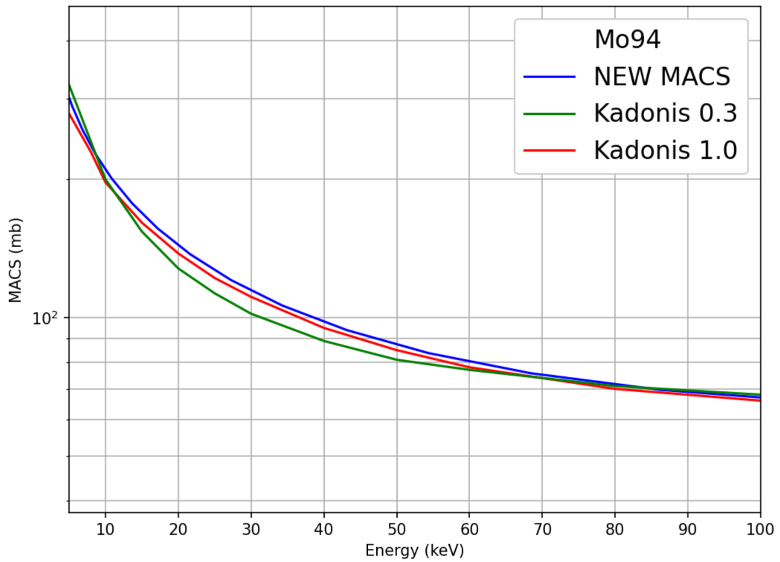
- 4 C6D6,
- 8 cm from sample.

SAMPLES

Samples:

- Pressed pellets in thin plastic bags,
- Samples mounted in sample exchanger.

Updated MACS



New MACS for all Mo isotopes using parameters from this thesis