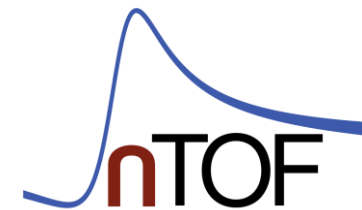


$^{50,53}\text{Cr}(n,\gamma)$ cross section measurement at n_TOF

P. PÉREZ-MAROTO, C. GUERRERO, A. CASANOVAS, M.E. STAMATI,
B. FERNÁNDEZ, N. PATRONIS & THE N_TOF COLLABORATION.

WONDER 2023, AIX-EN-PROVENCE. 05/06/2023



Motivation: nuclear data for criticality safety

NEA Nuclear Data High Priority Request List, HPRL

HPRL Main	High Priority Requests (HPR)	General Requests (GR)	Special Purpose Quantities (SPQ)		New Request	EG-HPRL (SG-C)
			Standard	Dosimetry		

Request ID	Request			Type of the request	High Priority request	
Target	Reaction and process	Incident Energy	Secondary energy or angle	Target uncertainty	Covariance	
24-CR-53	(n,g) SIG	1 keV-100 keV		8-10	Y	
Field	Subfield	Created date	Accepted date	Ongoing action	Archived Date	
Fission		20-JAN-18	05-FEB-18	Y		

Send a comment on this request to NEA.

Requester: Dr Roberto CAPOTE NOY at IAEA, AUT
Email: roberto.capotenoy@iaea.org

Project (context):

Impact:

Neutron absorption in the Cr isotopes of structural materials affects the criticality of fast reactor assemblies [Koscheev2017]. These cross sections are also of interest for stellar nucleosynthesis [Kadonis10].

Accuracy:

8-10% in average cross-sections and calculated MACS at 10, 30, 100 keV.

Selected criticality benchmarks with large amounts of Cr (e.g., PU-MET-INTER-002, and HEU-COMP-INTER-005/4=KBR-15/Cr) show large criticality changes of the order of 1000 pcm due to 30% change in Cr-53 capture in the region from 1 keV up to 100 keV [Trkov2018]. On the other side different evaluations (e.g., BROND-3.1, ENDF/B-VII.1, ENDF/B-VIII.0 and JEFF-3.3) for Cr-53(n,g) are discrepant by 30% in the same energy region. For Cr-50, evaluated files show better agreement at those energies but they are lower than Mughabghab evaluation of the resonance integral by 35%. These discrepancies are not reflected in estimated uncertainty of the evaluated files (e.g., JEFF-3.3 uncertainty is around 10% which is inconsistent with the observed spread in evaluations). Due to these differences we request new capture data with 8-10% uncertainty to discriminate between different evaluations and improve the C/E for benchmarks containing Chromium and/or SS.

Justification document:

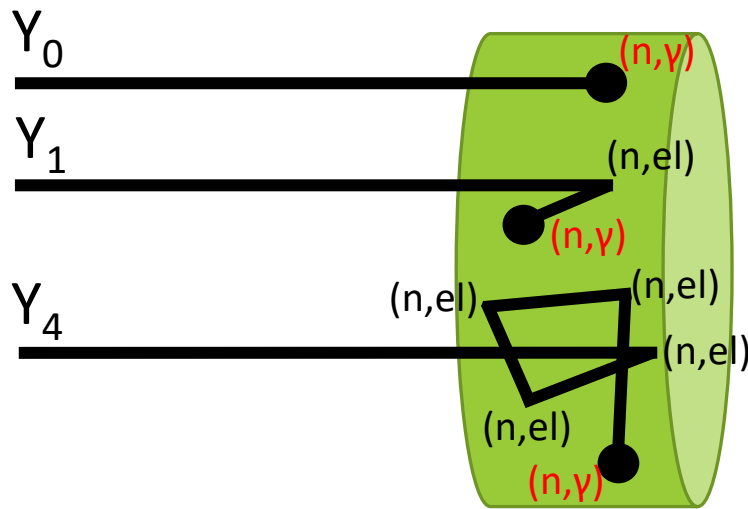
Criticality benchmarks can test different components of stainless steel (SS), including Cr which is a large component of some SS. Currently, a large part of the uncertainty in SS capture seems to be driven by uncertainty in Cr capture [Koscheev2017]. Indeed, some benchmarks highly sensitive to Cr (as a component of SS) indicate a need for much higher capture in Cr for both Pu and U fueled critical assemblies (e.g., HEU-COMP-INTER-005/4=KBR-15/Cr and PU-MET-INTER-002=ZPR-6/10).



- Stainless Steel is often used as a **structural material in nuclear reactors** and contains between **11-26% of chromium**.
- There are **serious discrepancies (~30%)** between the different evaluated data of **^{50}Cr and ^{53}Cr capture cross section**, which is not present in the corresponding estimated uncertainties.
- **OECD NEA-HPRL (High Priority Request List)**
 → **$^{50,53}\text{Cr}(n,\gamma)$ within 8-10% at 1 to 100 keV.**

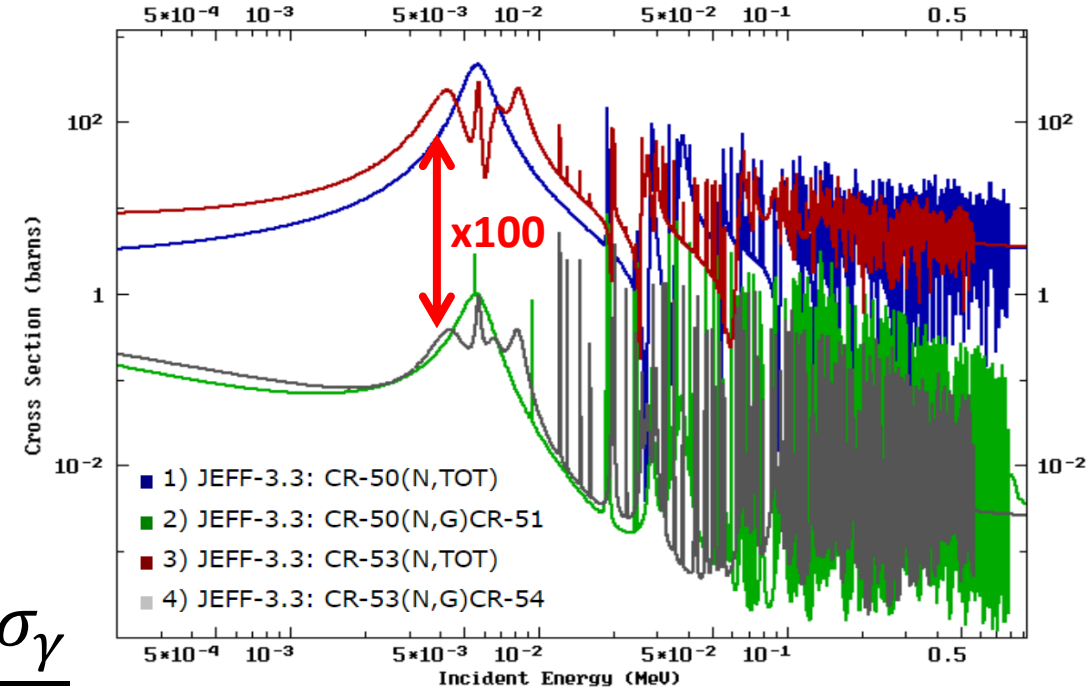
Why the discrepancies?

- The main problem for measuring $Cr(n,\gamma)$ is the large neutron multiple-scattering effects
- In the previous measurements thick samples were used, aiming for good statistics in a very wide energy range



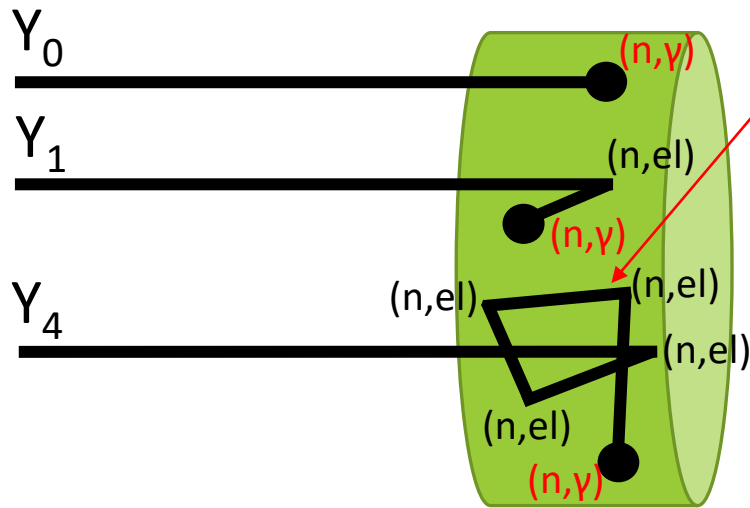
$$Y_0 = (1 - e^{-n\sigma_t}) \frac{\sigma_\gamma}{\sigma_t}$$

Capture yield (captures/neutron) $\rightarrow Y = \underbrace{Y_0}_{\text{Analytical (accurate)}} + \underbrace{Y_1 + Y_2 + Y_3 \dots}_{\text{Numerical (approximate)}}$



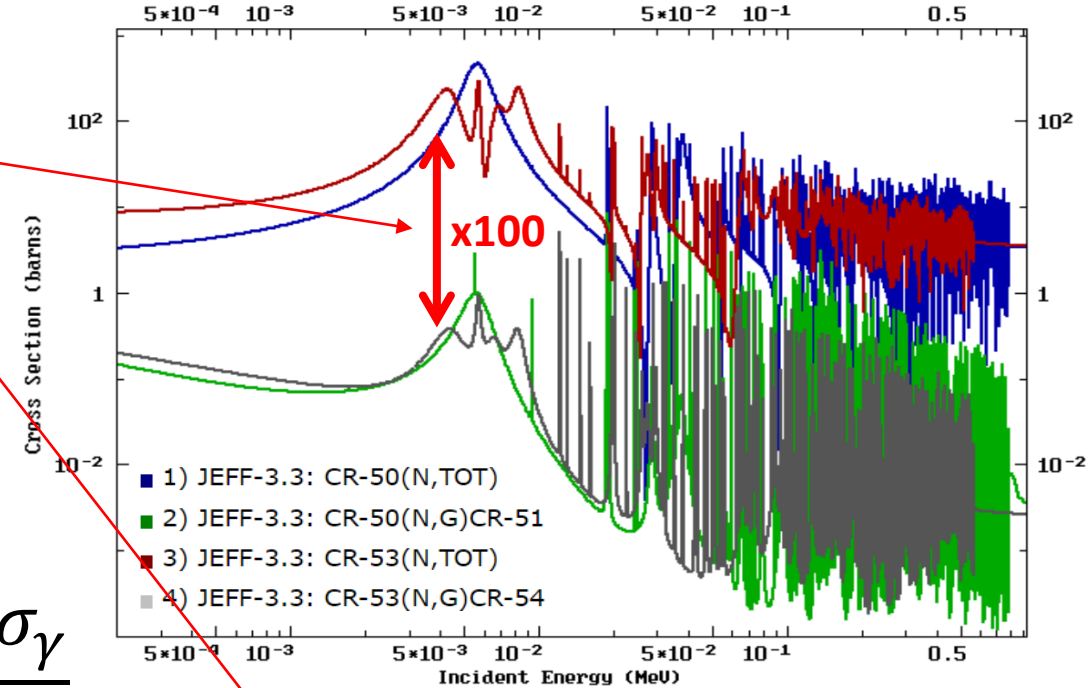
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How to improve $\sigma(n,\gamma)$ down to a few %?

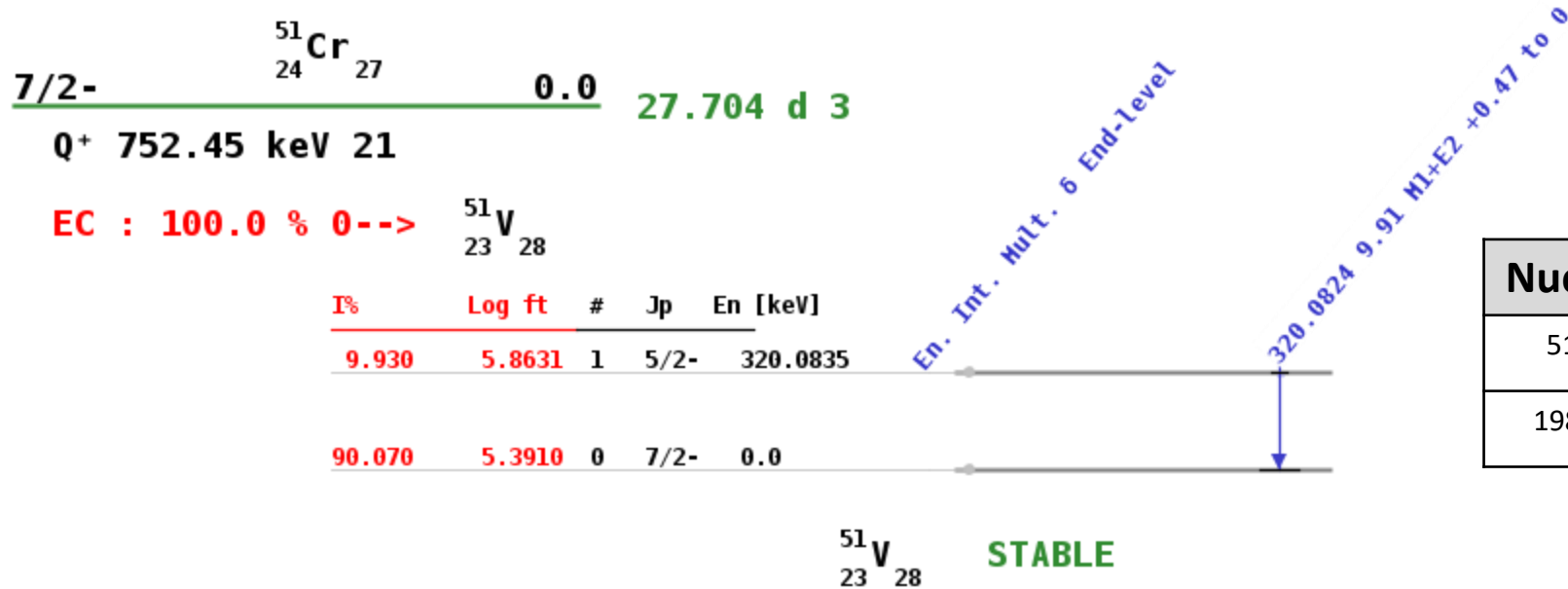
- Enriched (expensive and scarce) material with high purity \rightarrow 94,6% ^{50}Cr & 97,7% ^{53}Cr
- Controlling multiple-scattering effects:
 - Very thin/thin sample approach
 - C_6D_6 detectors (low sensitivity to scattered neutrons)

Experiment	Beer (1975)	Stieglitz (1971)	Brusegan (1986)	Kenny (1977)	Guber (2011)	This work (2022)
Facility	FZK	RPI	GELINA	ORELA	ORELA	n_TOF
L (m)	0,7	27	60	40	40	185
Energy (keV)	1-300	1-200	1-200	1-200	0,01-600	1-100
<u>Density ^{50}Cr</u> <u>(10^{-3} at/barns)</u>	<u>18</u>	<u>8</u>	<u>7</u>	<u>5/8</u>	-	0,6/1,9
<u>Density ^{53}Cr</u> <u>(10^{-3} at/barns)</u>	<u>14</u>	<u>14</u>	<u>12/60</u>	<u>8/12</u>	14	1,2/6

**Our “thicks” are thinner than all previous
 \rightarrow lower multiple interaction corrections**

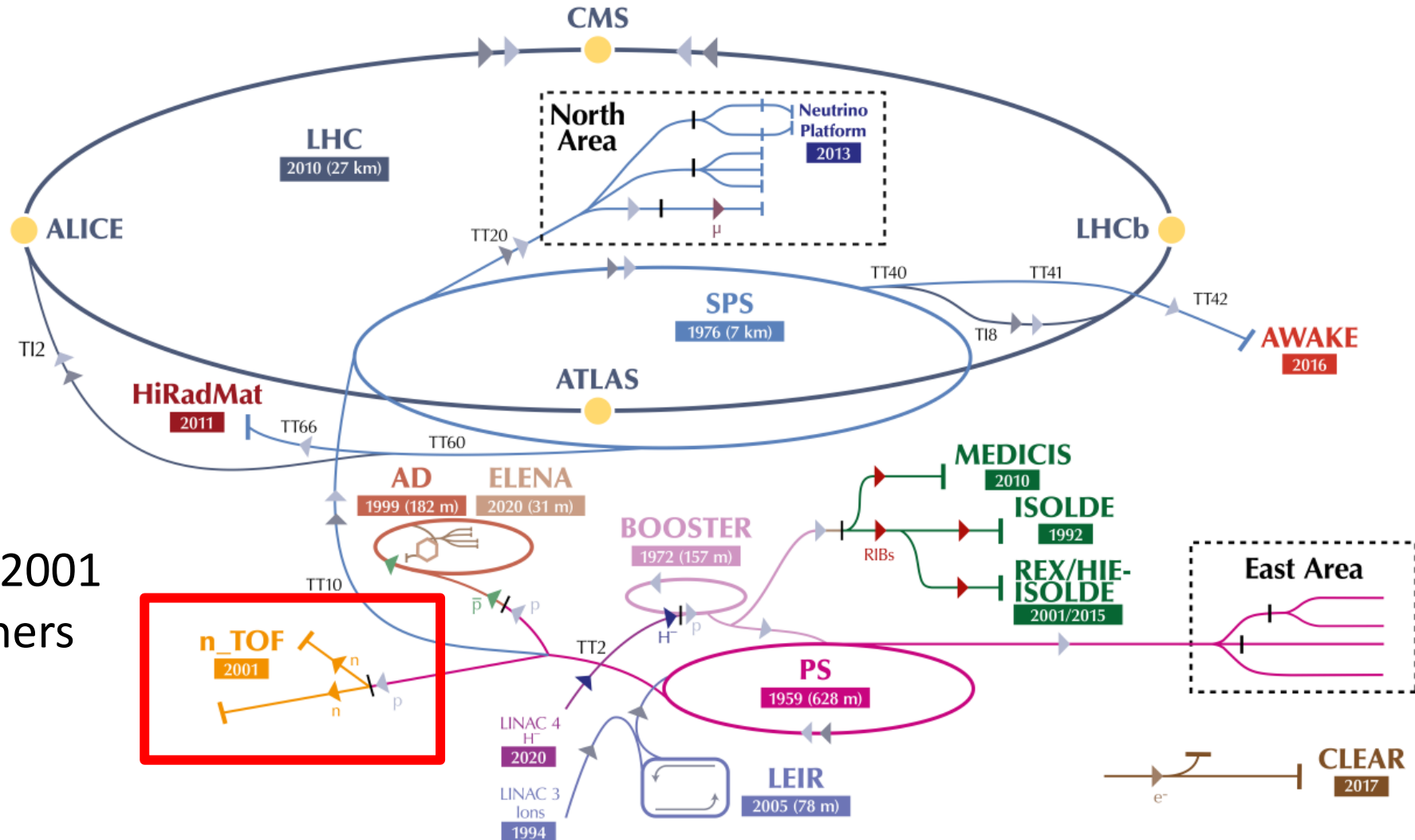
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- Enriched (expensive and scarce) material with high purity \rightarrow 94,6% ^{50}Cr & 97,7% ^{53}Cr
- Controlling multiple-scattering effects:
 - Very thin/thin sample approach
 - C_6D_6 detectors (low sensitivity to scattered neutrons)
- Complementing with ^{50}Cr activation measurement \rightarrow HiSPANoS@CNA (Seville, Spain)



Nucleus	$T_{1/2}$ (days)	E_γ (keV)	I_γ (%)
^{51}Cr	27,7	320,1	9,9
^{198}Au	2,69	411,8	95,5

The neutron Time-Of-Flight facility at CERN

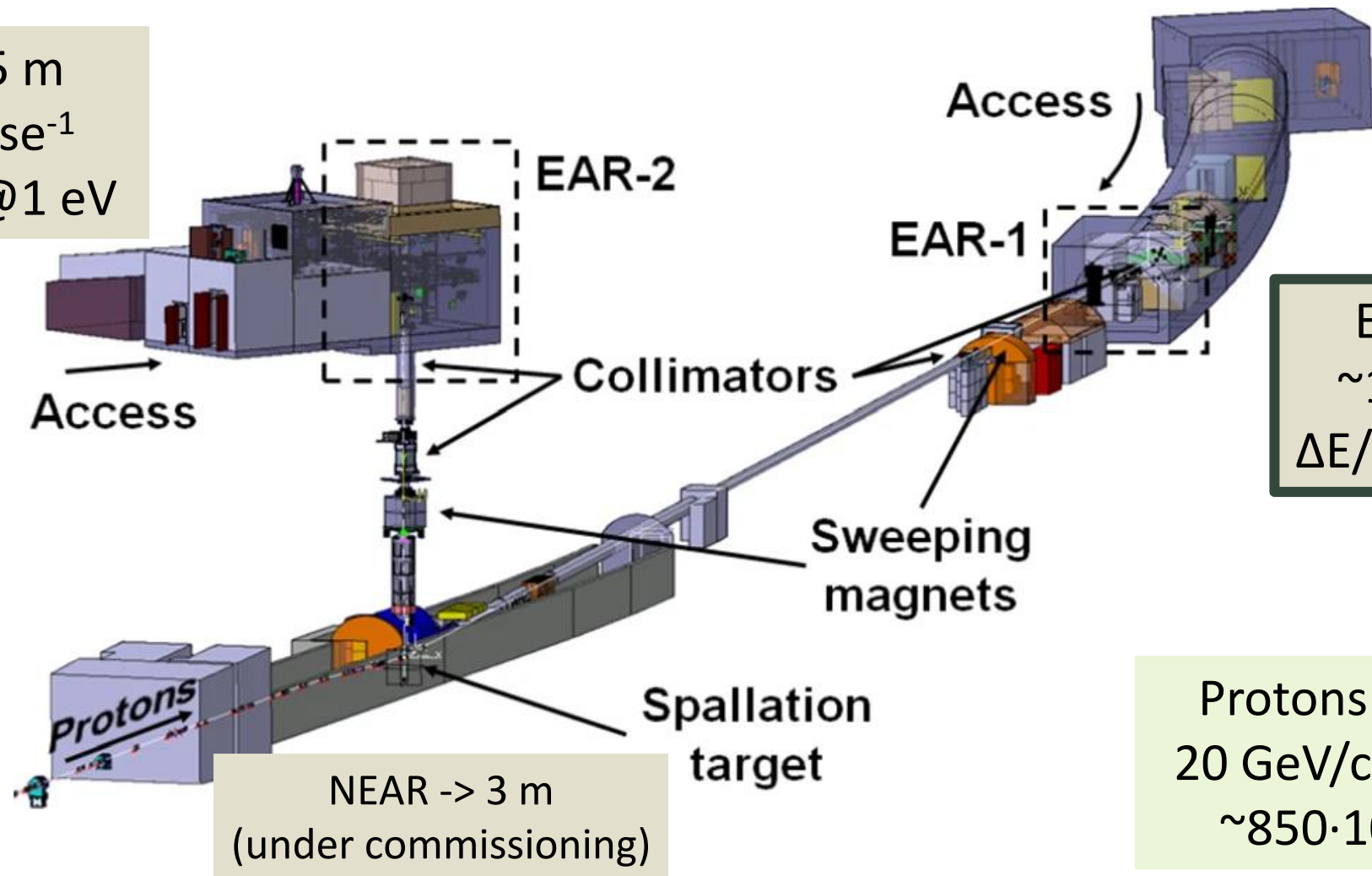


n_TOF@CERN

In operation since 2001
 2022: 131 researchers
 from 37 institutes

The neutron Time-Of-Flight facility at CERN

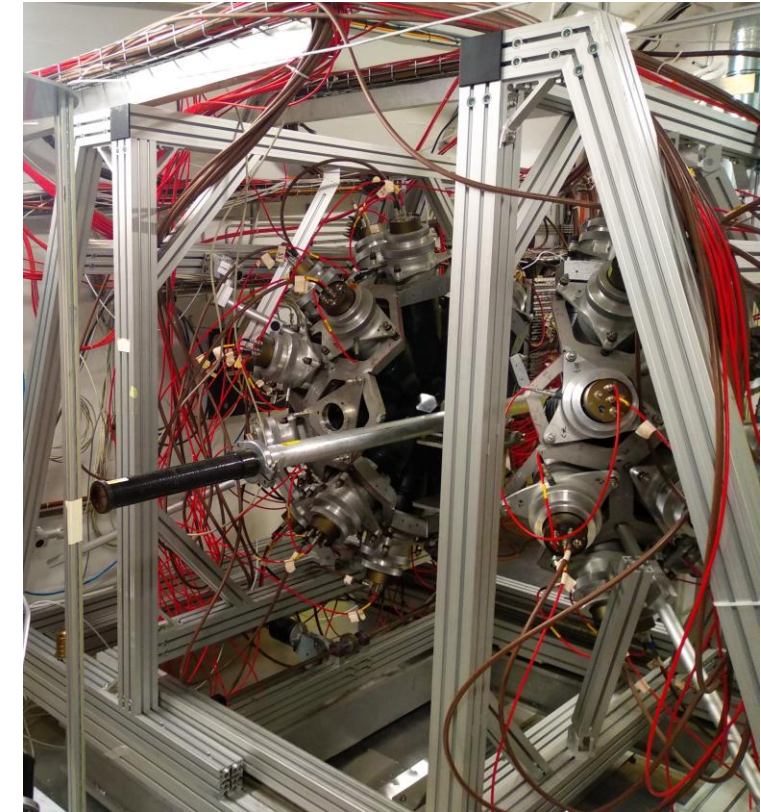
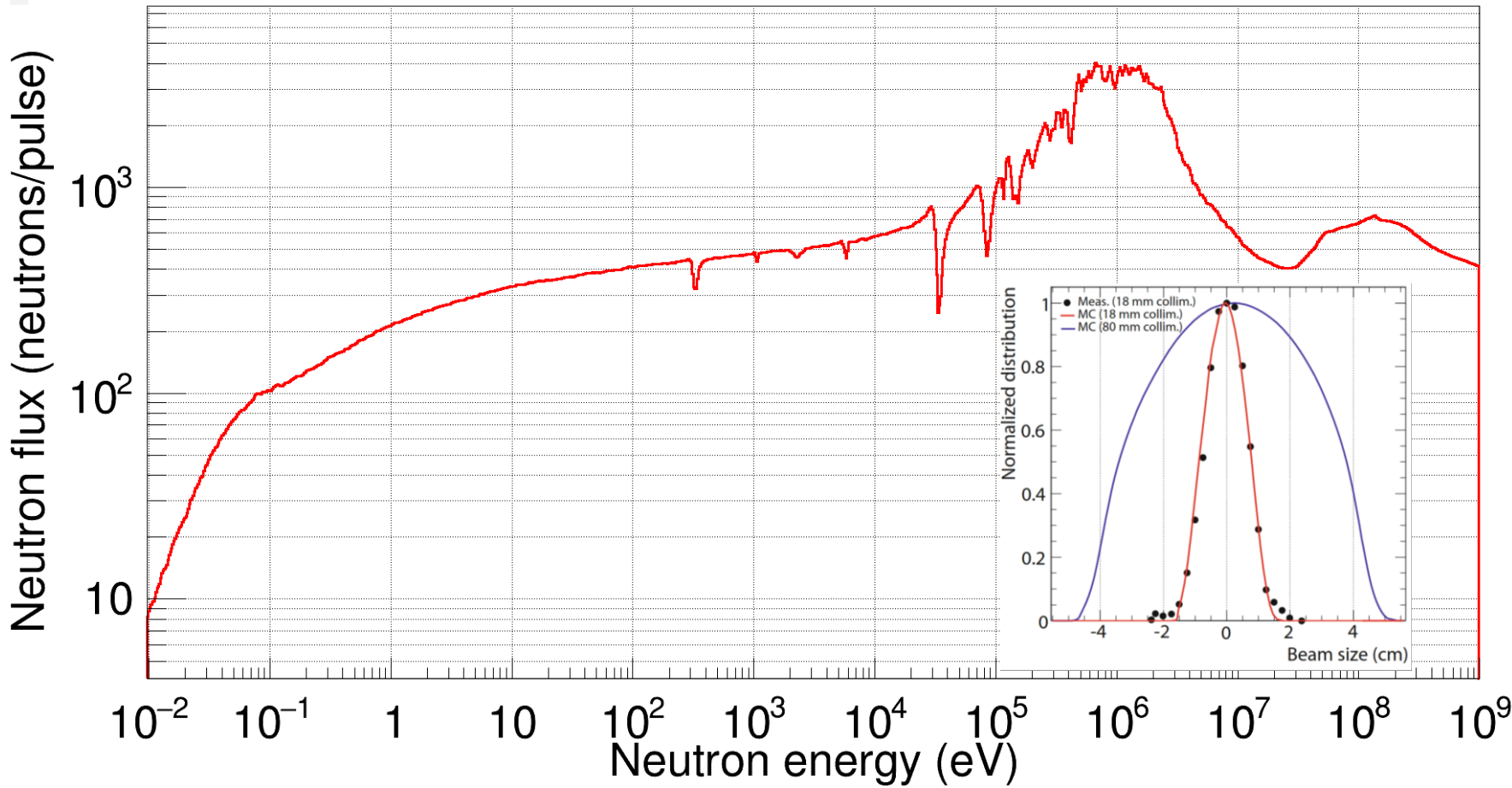
EAR2 -> 18,5 m
 $\sim 10^6 \text{ cm}^{-2} \text{ pulse}^{-1}$
 $\Delta E/E \sim 4 \cdot 10^{-3} @ 1 \text{ eV}$



EAR1 -> 185 m
 $\sim 10^4 \text{ cm}^{-2} \text{ pulse}^{-1}$
 $\Delta E/E \sim 5 \cdot 10^{-4} @ 1 \text{ eV}$

Protons from PS
20 GeV/c at 0,8 Hz
 $\sim 850 \cdot 10^{10} \text{ ppp}$

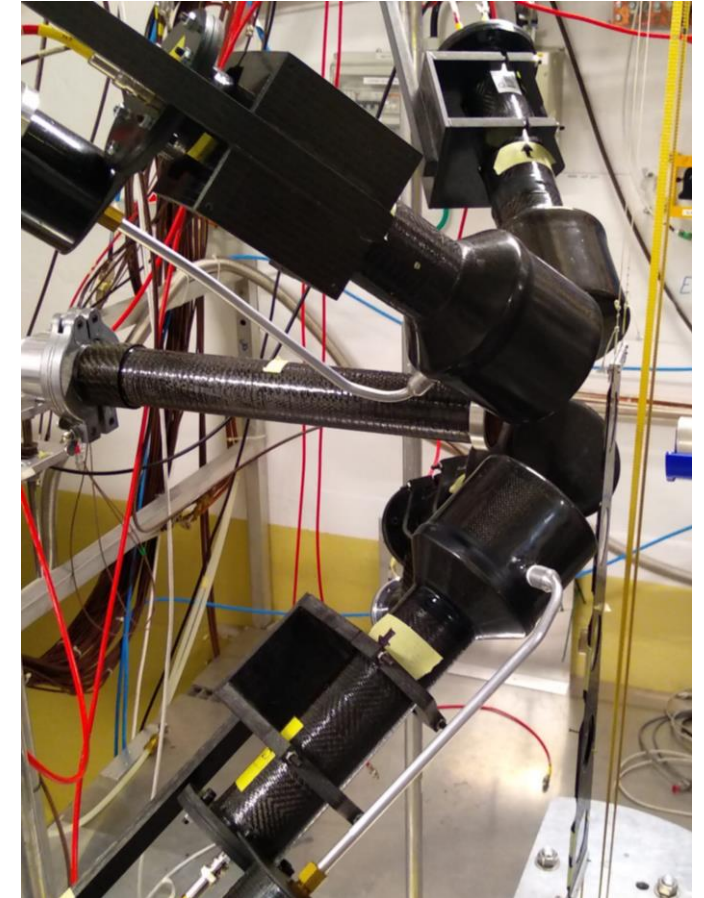
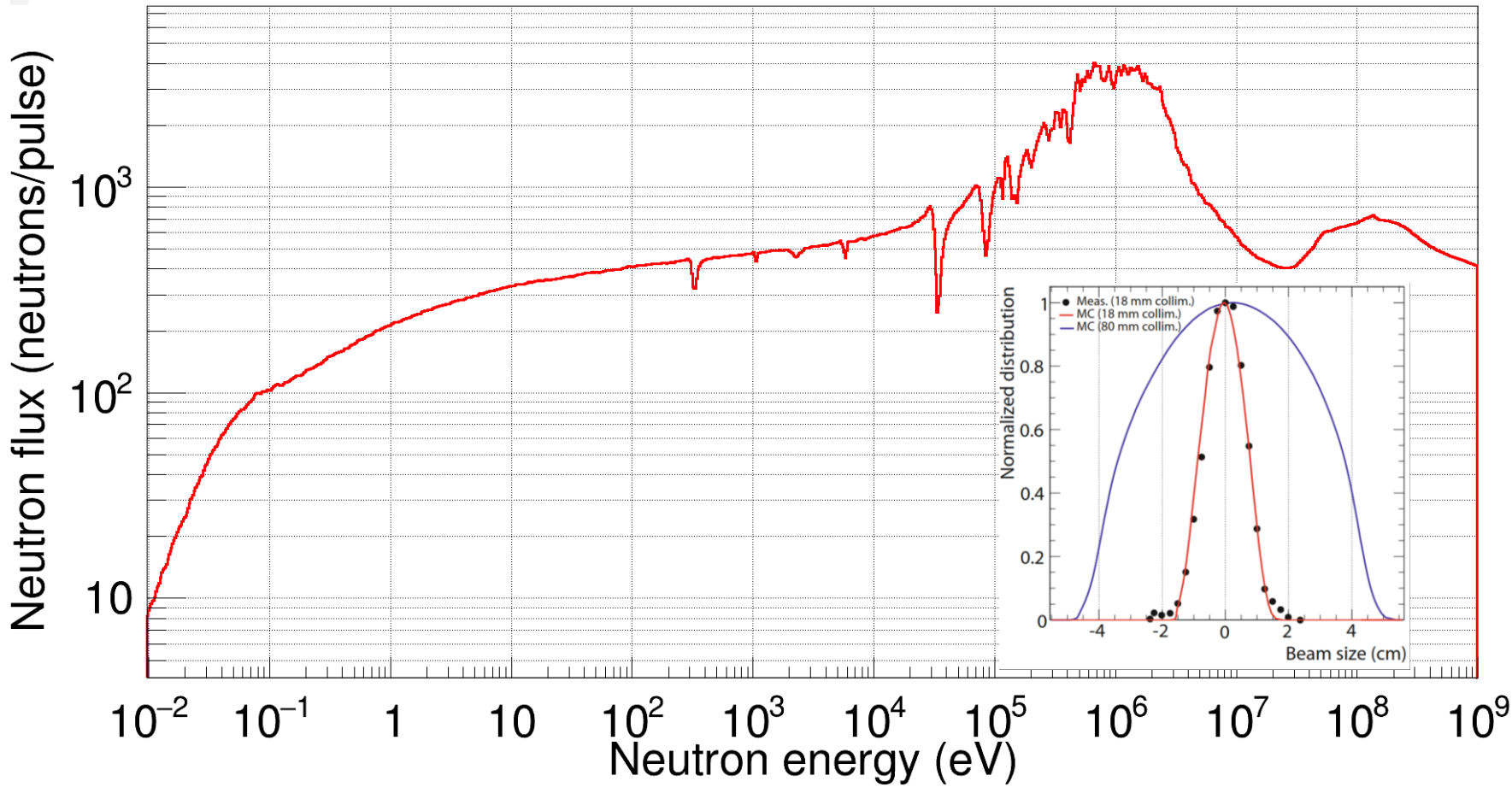
n_TOF – EAR1. Detection techniques



- Total Absorption technique
- Full γ -ray cascade detected
- High neutron sensitivity



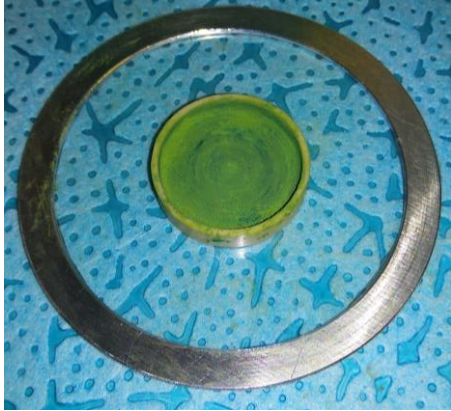
n_TOF – EAR1. Detection techniques



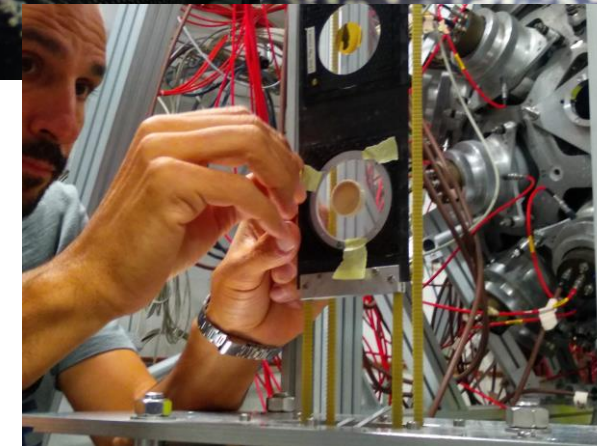
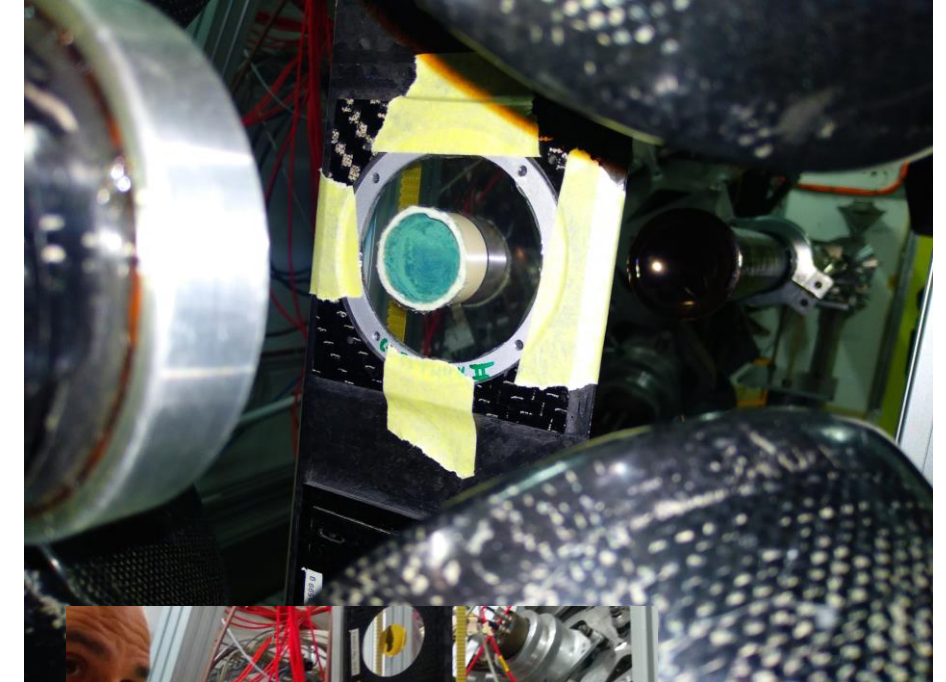
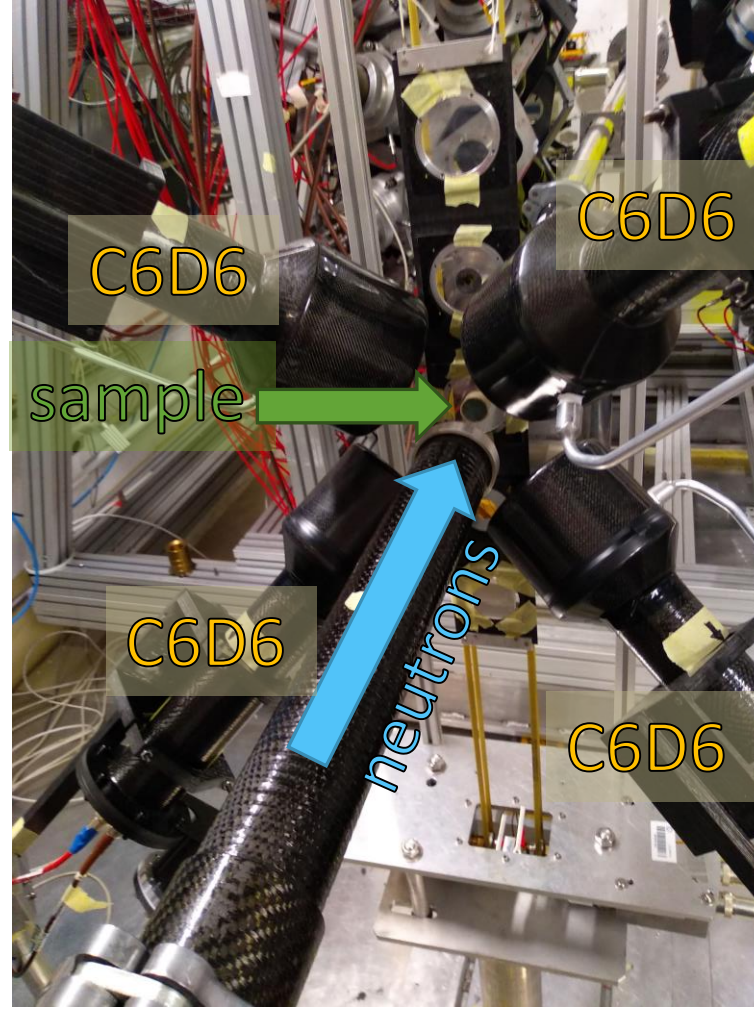
- Total Energy Detection
- Only one γ -ray detected
- Low neutron sensitivity



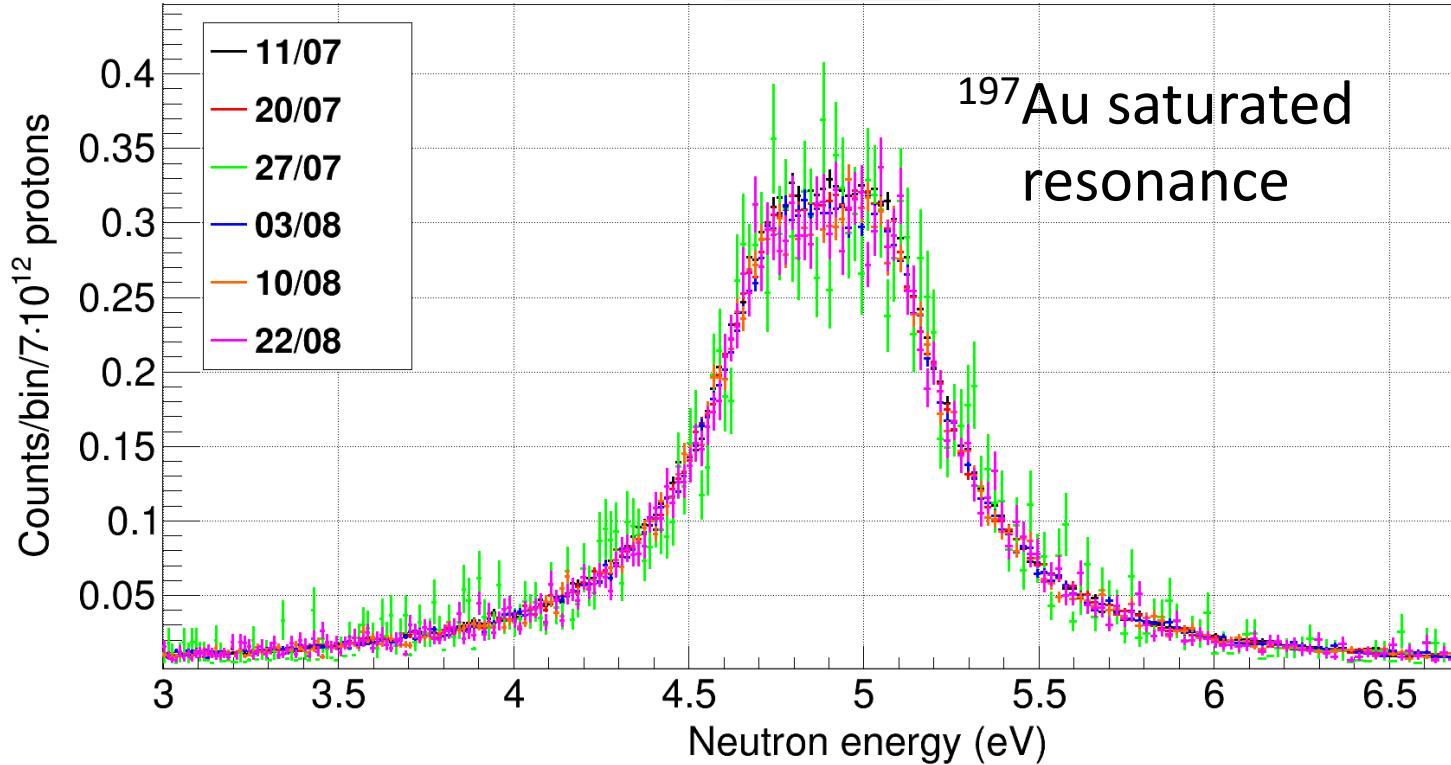
Samples and detector set-up (EAR1)



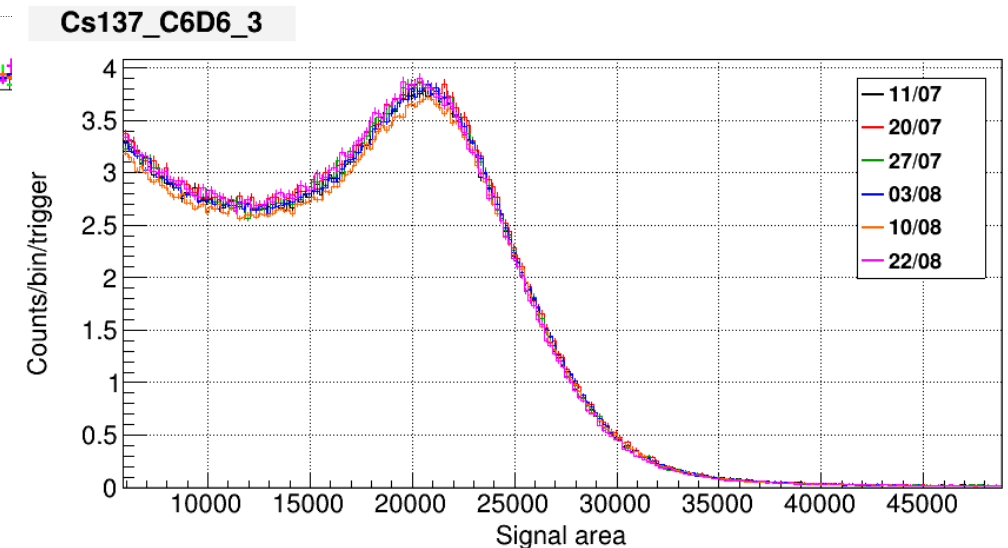
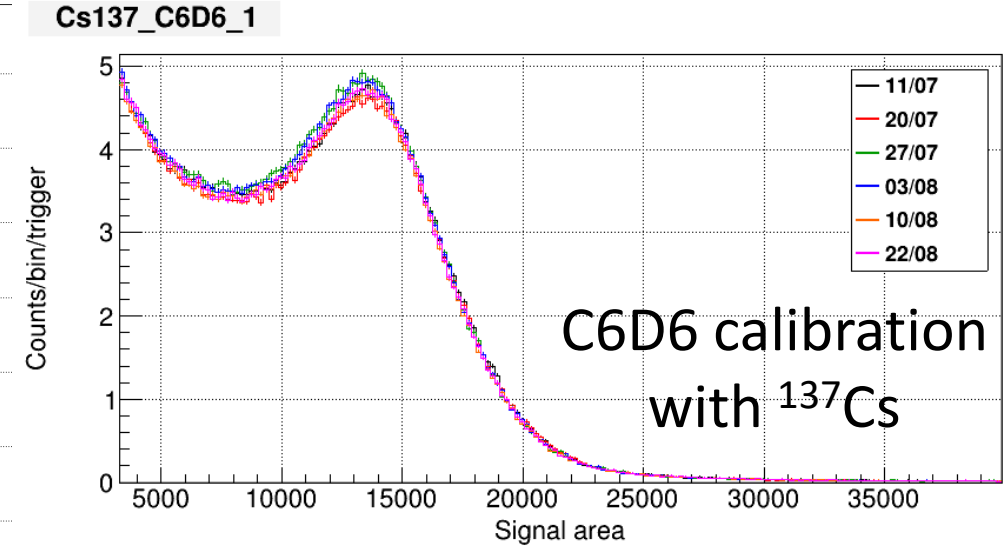
Cr_2O_3 powder pressed in a PEEK capsule & Al holder



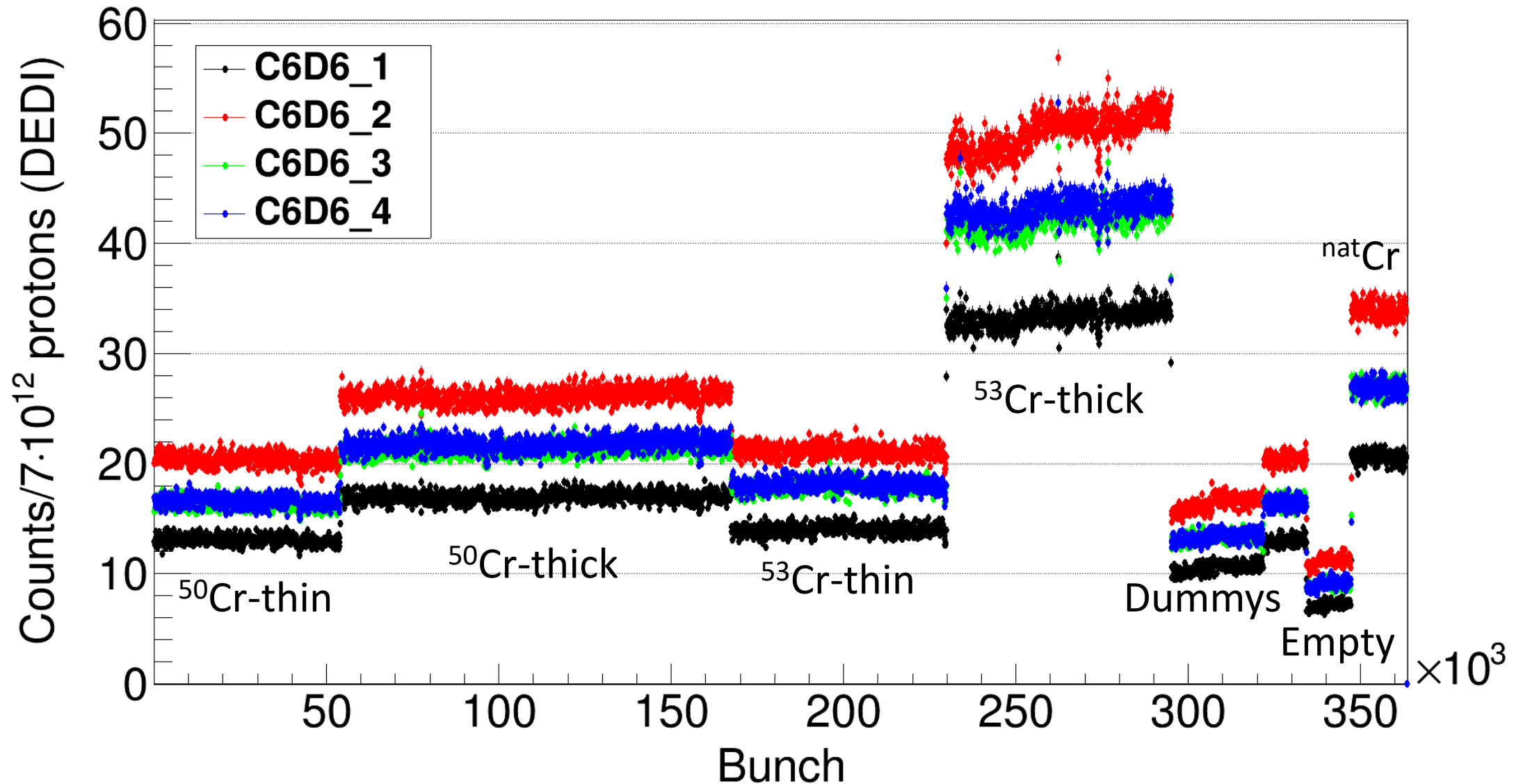
Monitoring the measurement



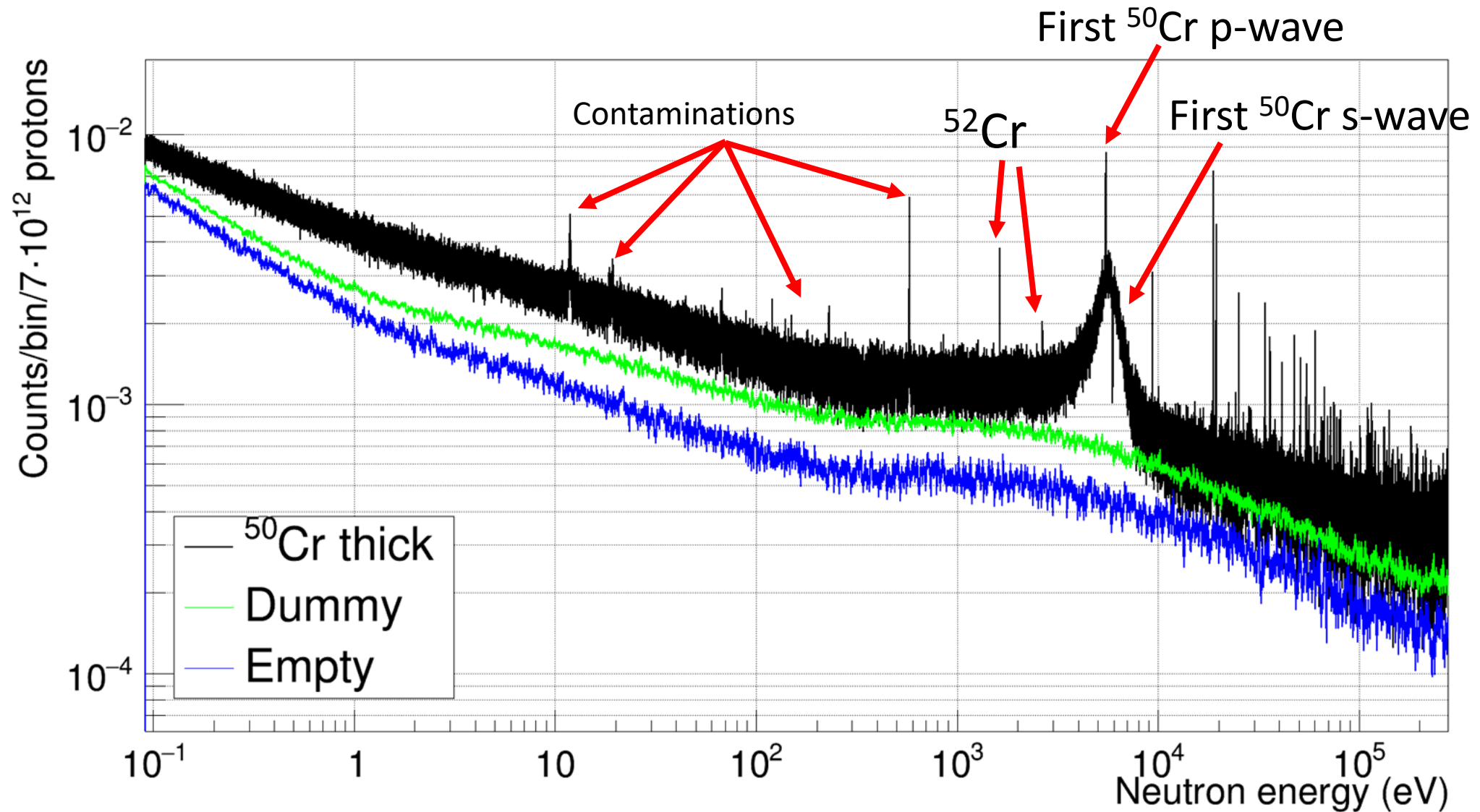
- Several monitors and stability checks regarding proton beam, neutron flux, detector gain-shifts...
- Very good behavior during the whole campaign.
- Dummy, empty, ^{nat}Cr , filters,...



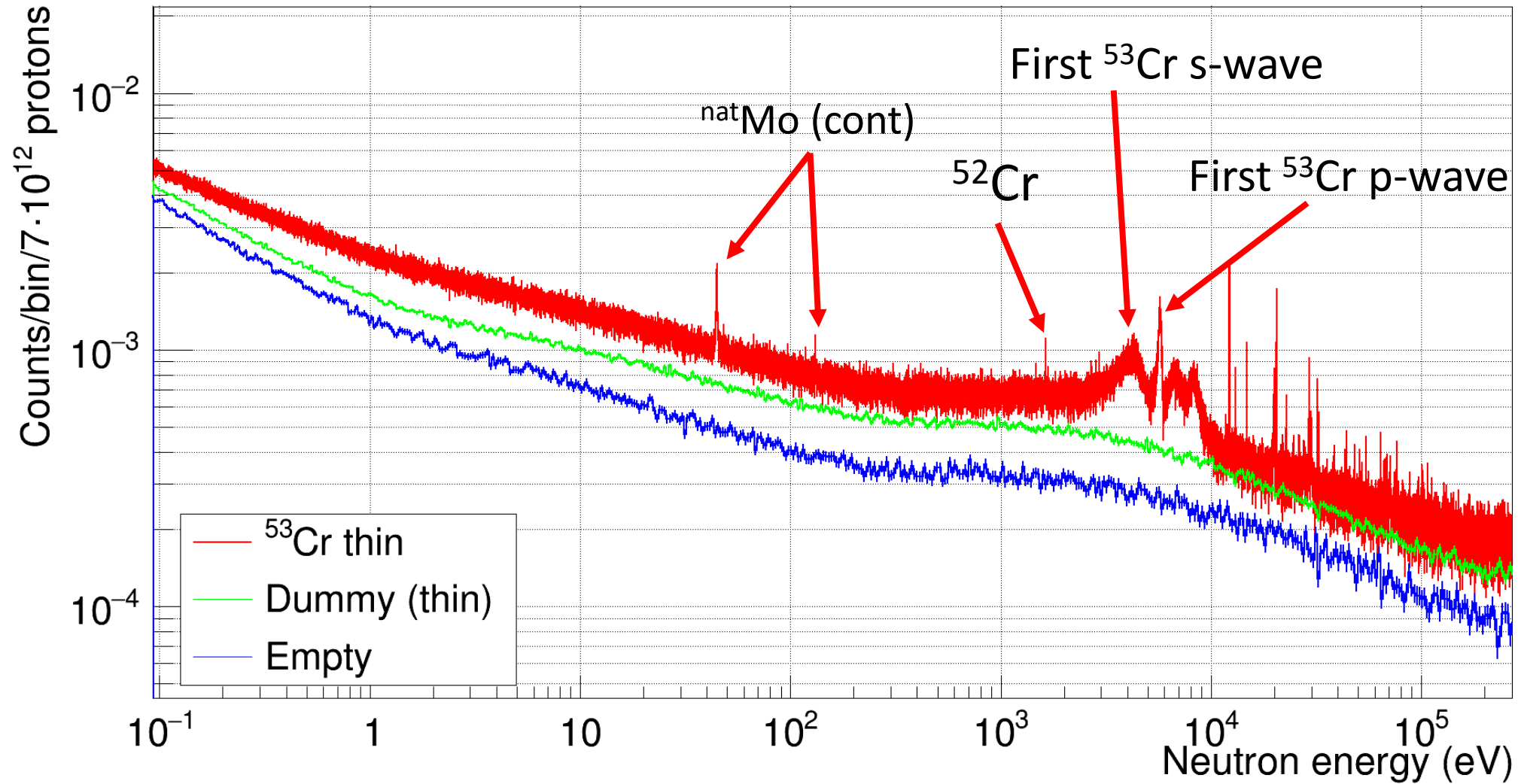
Monitoring the measurement



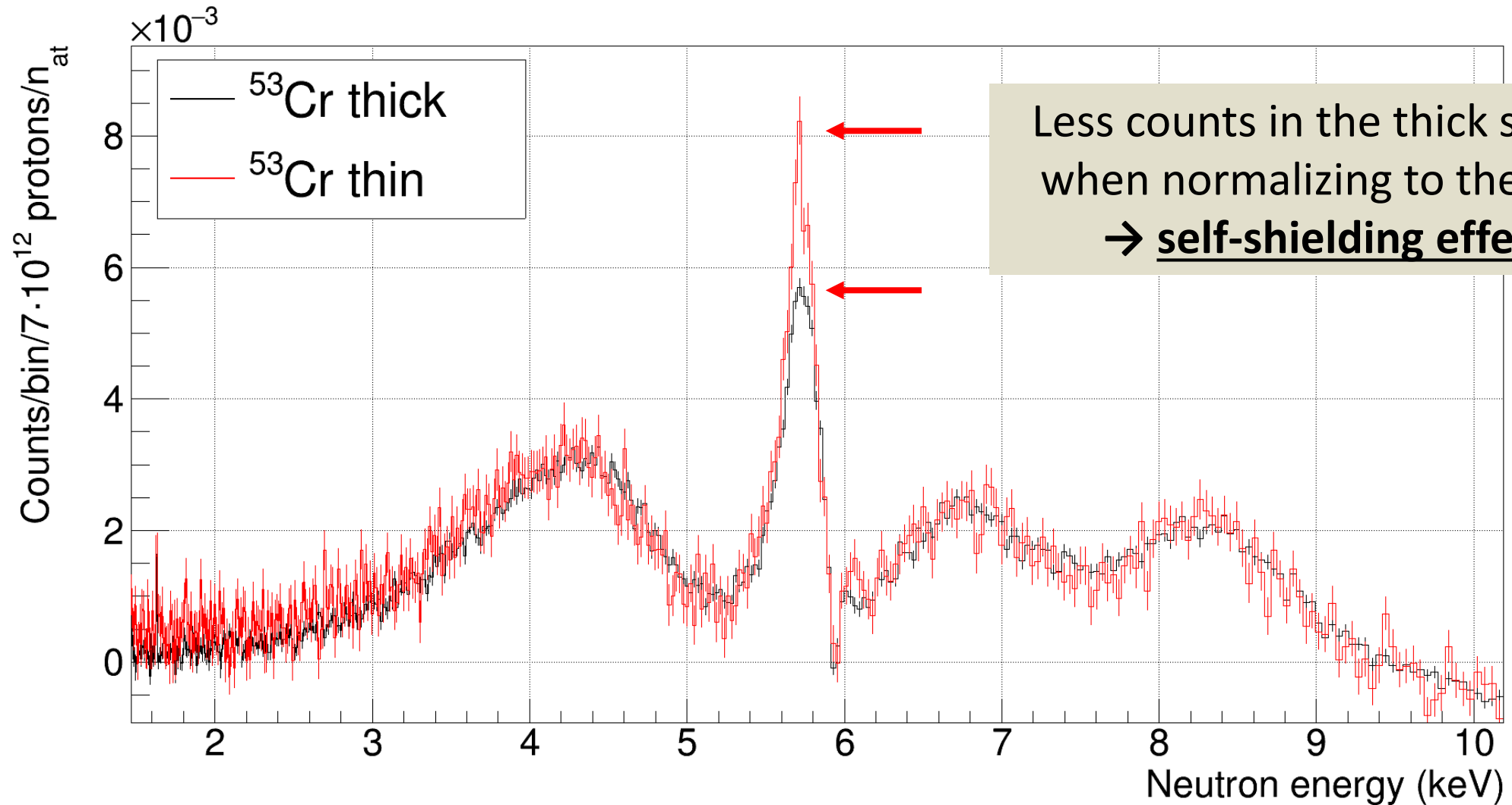
Preliminary results (^{50}Cr -thick)



Preliminary results (^{53}Cr -thin)



Preliminary results (^{53}Cr : thin vs. thick)

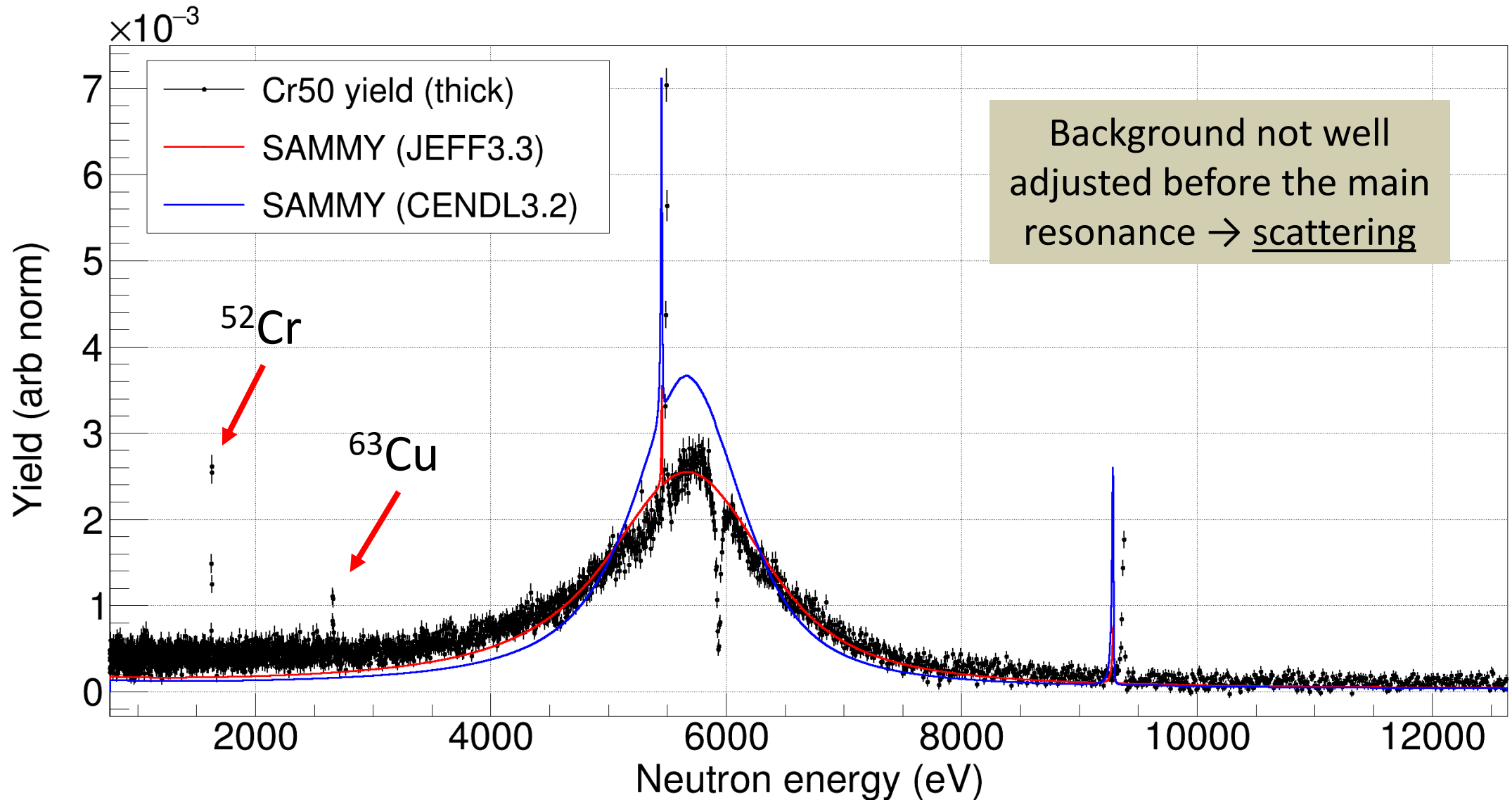


Preliminary yield (^{50}Cr)

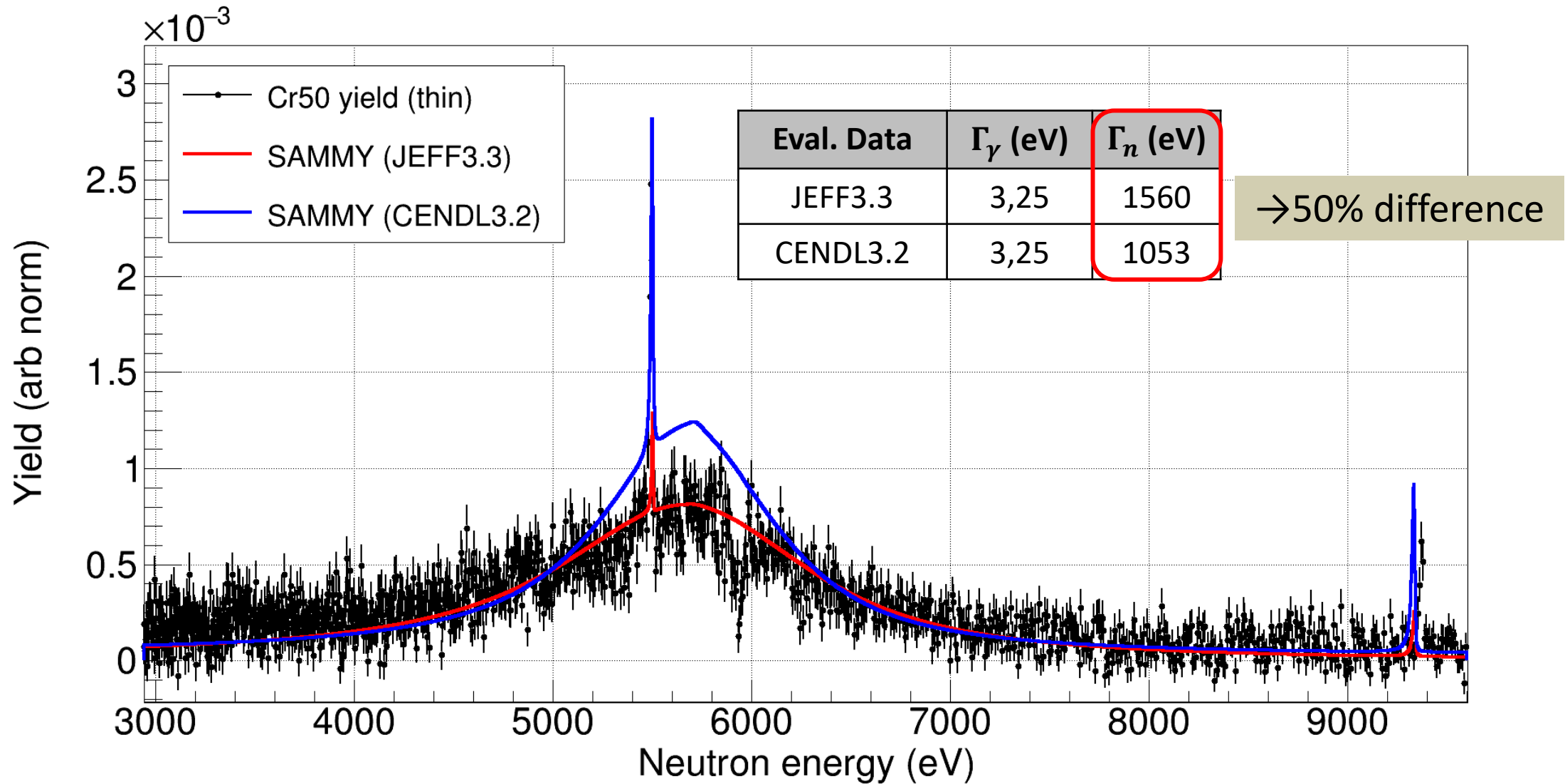
- We have obtained a preliminary “yield” to check our experimental data.
- To do so:
 1. Only background from dummy considered
 2. Normalize to the main resonance
 3. n_TOF Resolution Function to be included
 4. Compared with SAMMY + JEFF3.3 & CENDL3.2

$$Y(E_n) = F \frac{C(E_n) - B(E_n)}{\Phi(E_n)\varepsilon_\gamma}$$

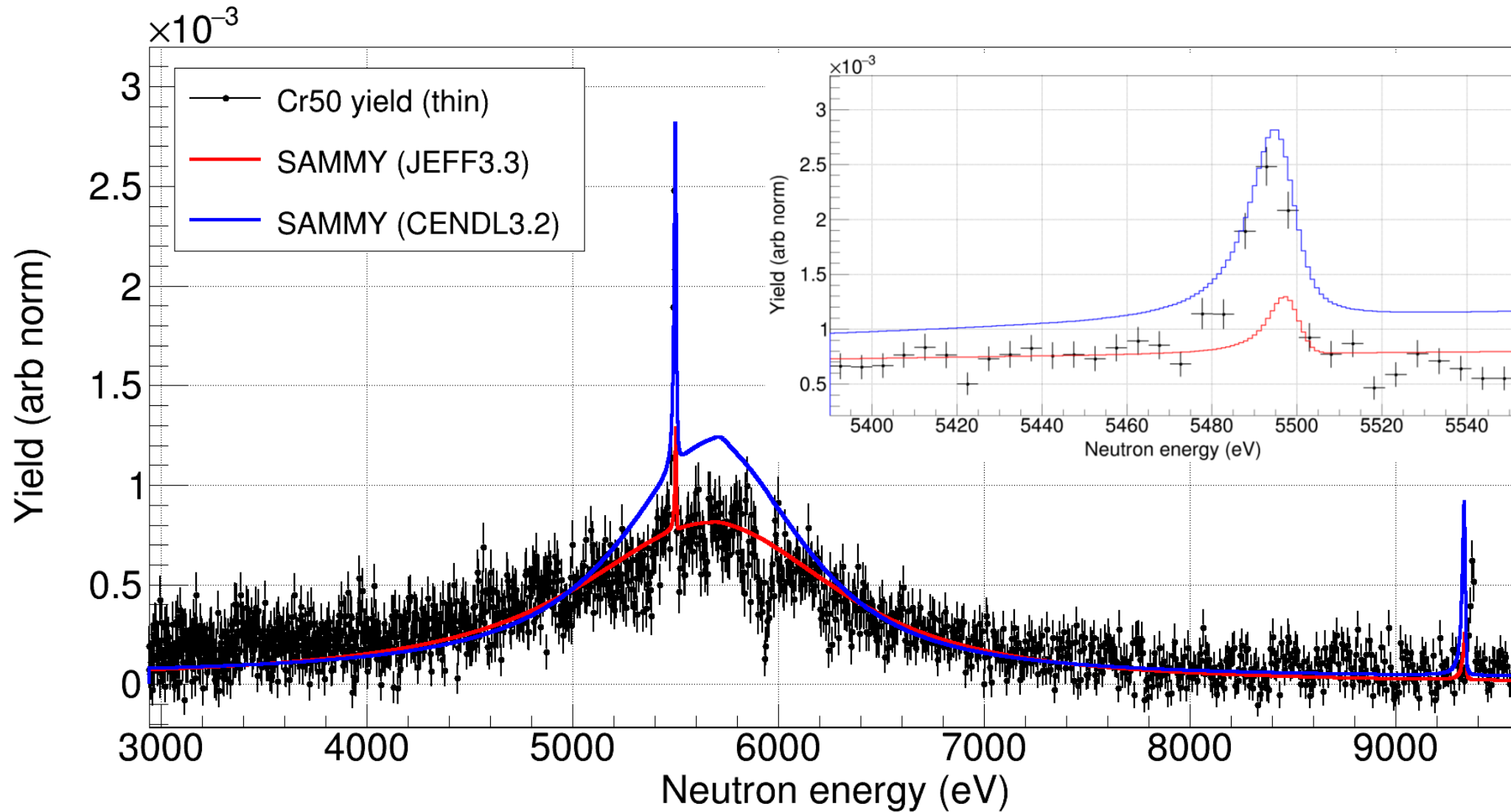
Preliminary yield (^{50}Cr)



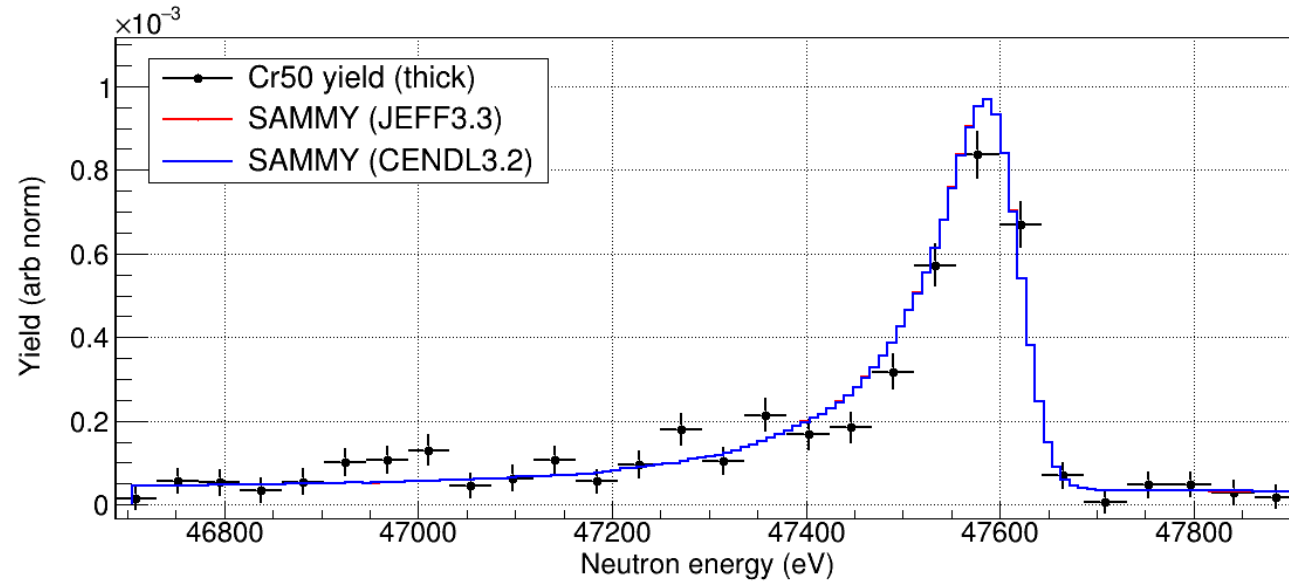
Preliminary yield (^{50}Cr)



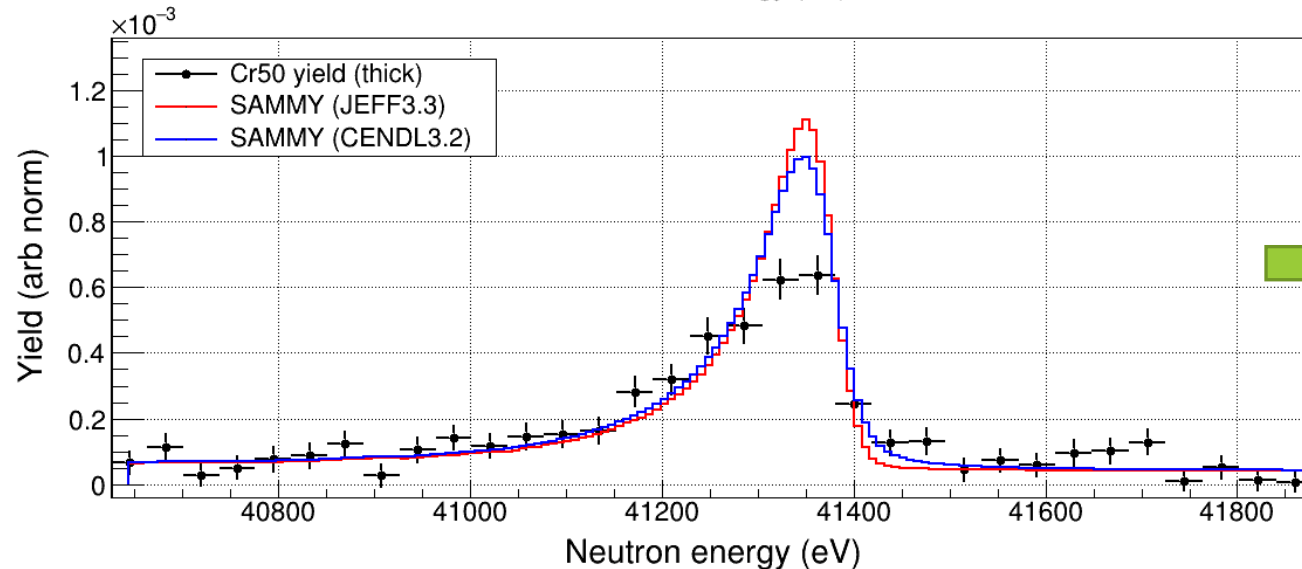
Preliminary yield (^{50}Cr)



Preliminary yield (^{50}Cr)

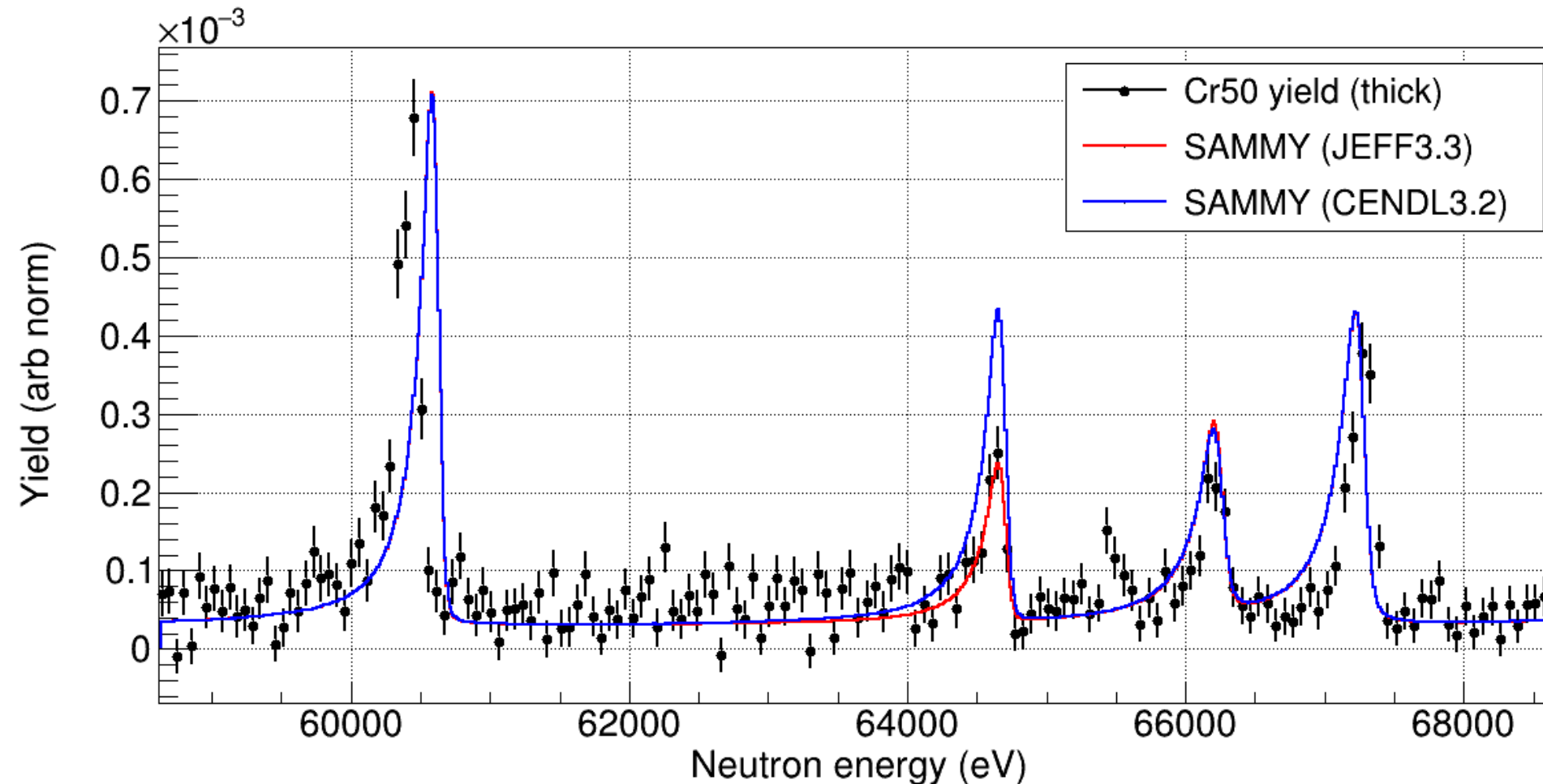


- The shape of the yield matches the one obtained with SAMMY.
- This is the case for most of the resonances, for a few of them there are differences \rightarrow bigger scattering contribution?



Eval. Data	Γ_γ (eV)	Γ_n (eV)
JEFF3.3	0,43	3,5
CENDL3.2	0,43	18,1

Preliminary yield (^{50}Cr)

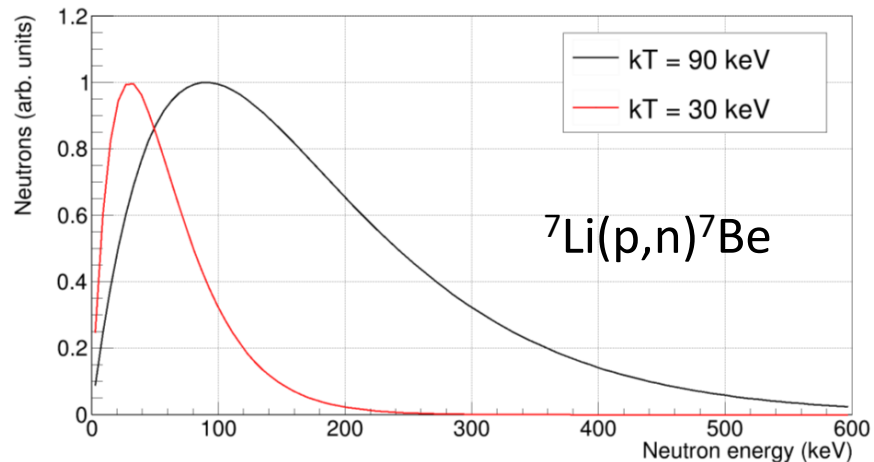


- In general, if there are discrepancies between evaluations, we agree with JEFF3.3
- A new measurement to solve the problem was indeed necessary.
- Let's wait for the results of our data!

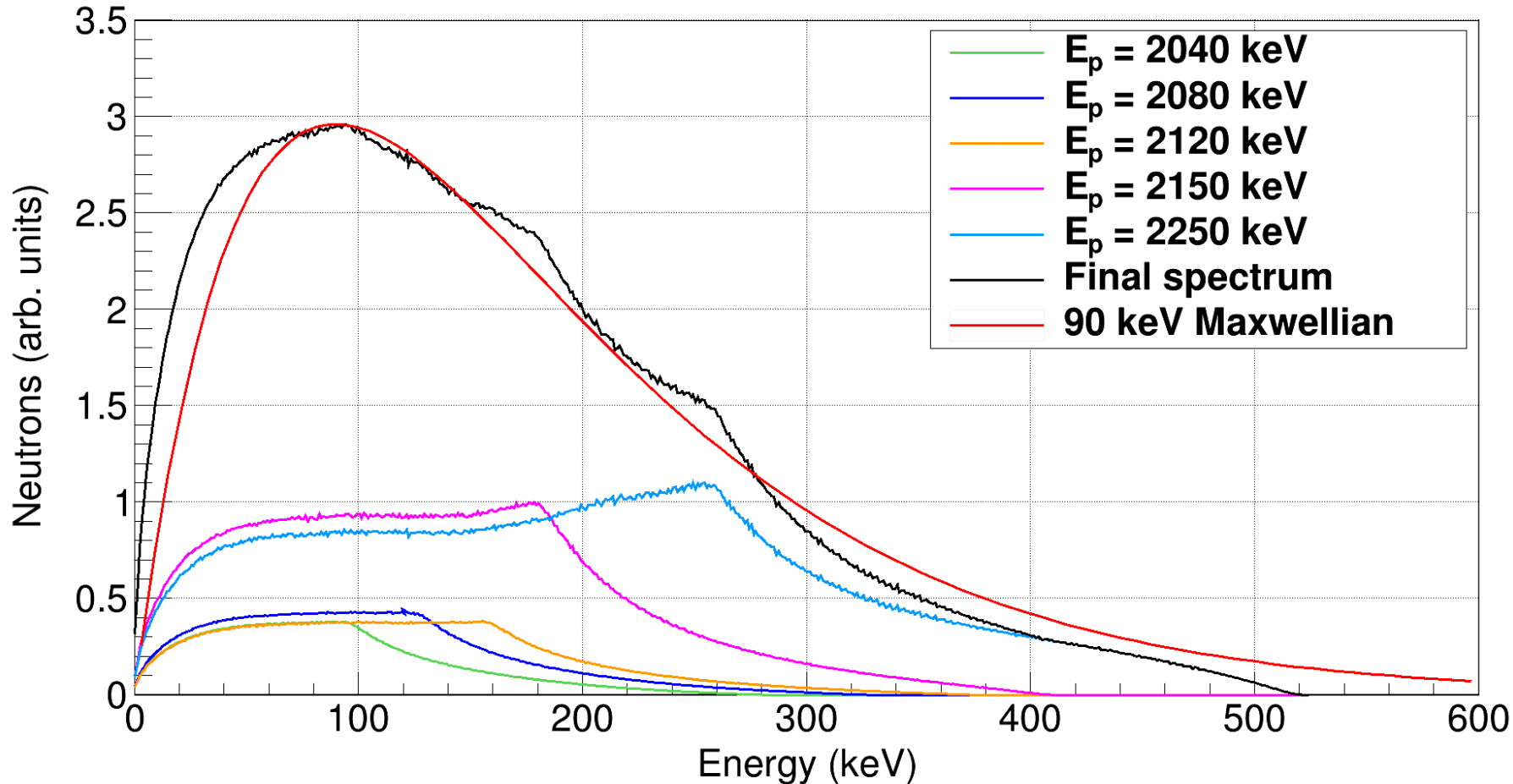
^{50}Cr MACS measurement at HiSPANoS@CNA

	Time of flight technique	Neutron activation
Energy and resonance shape	Very well defined	Limited “resolution” (MB distribution)
Absolute value	Susceptible to systematic effects	Very accurate (“easily” ~5%)

- An integral measurement can be very helpful with the analysis.
- ***“Development of a 90 keV Maxwellian neutron spectrum and measurement of the 30 & 90 keV ^{50}Cr MACS for criticality safety” (H2020-ARIEL Transnational Access).***



^{50}Cr activation: 90 keV MB neutron distribution

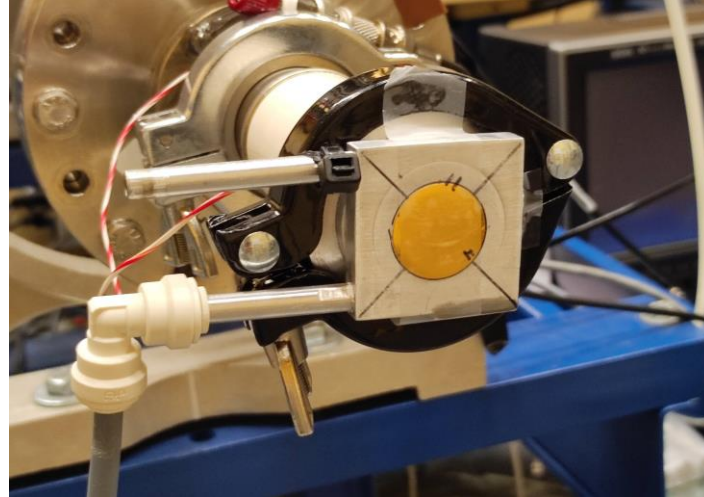


- A 30 keV spectrum can be produced with $E_p = 1912$ keV.
- For the 90 keV spectrum we need a linear combination of different proton energies.
- Maybe subtracting 1912 keV fits better a 90keV MB.

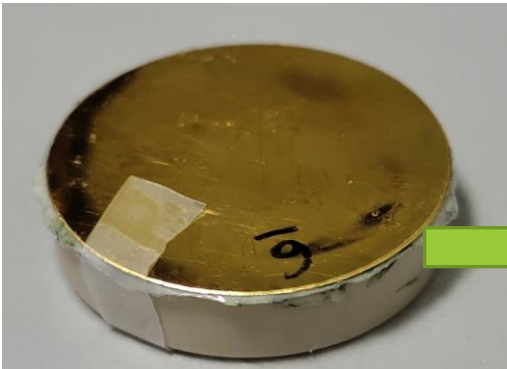
^{50}Cr activation: set-up



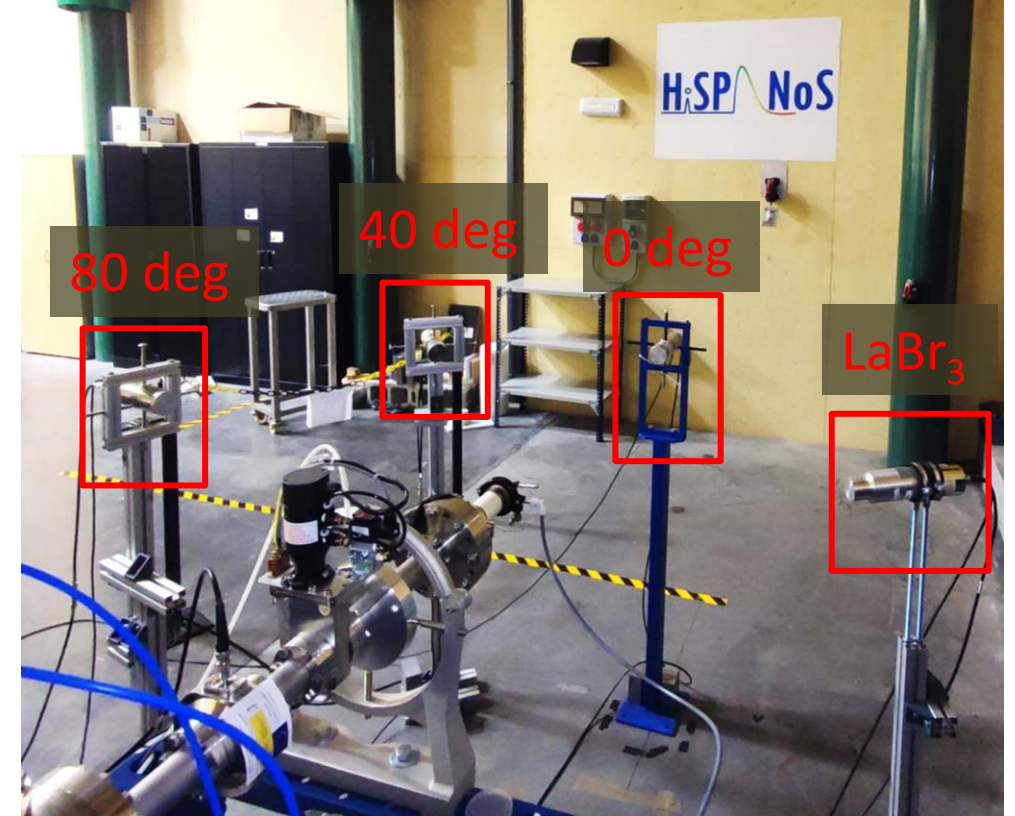
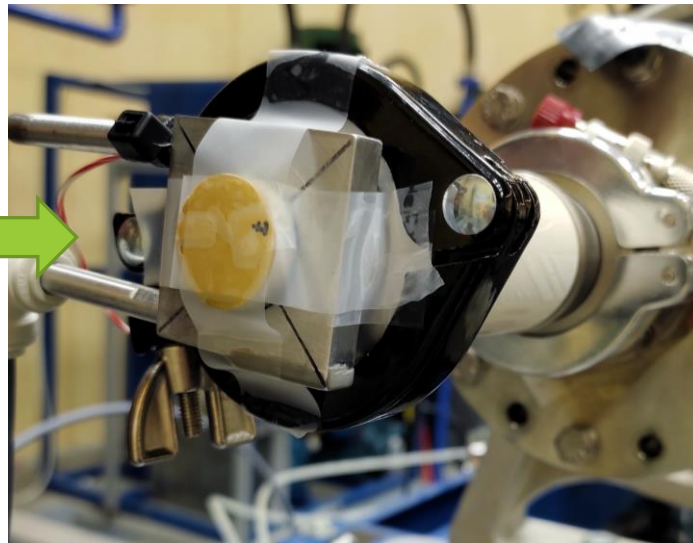
Metallic Li for higher production \rightarrow cooled target



^{197}Au irradiation for activation checks

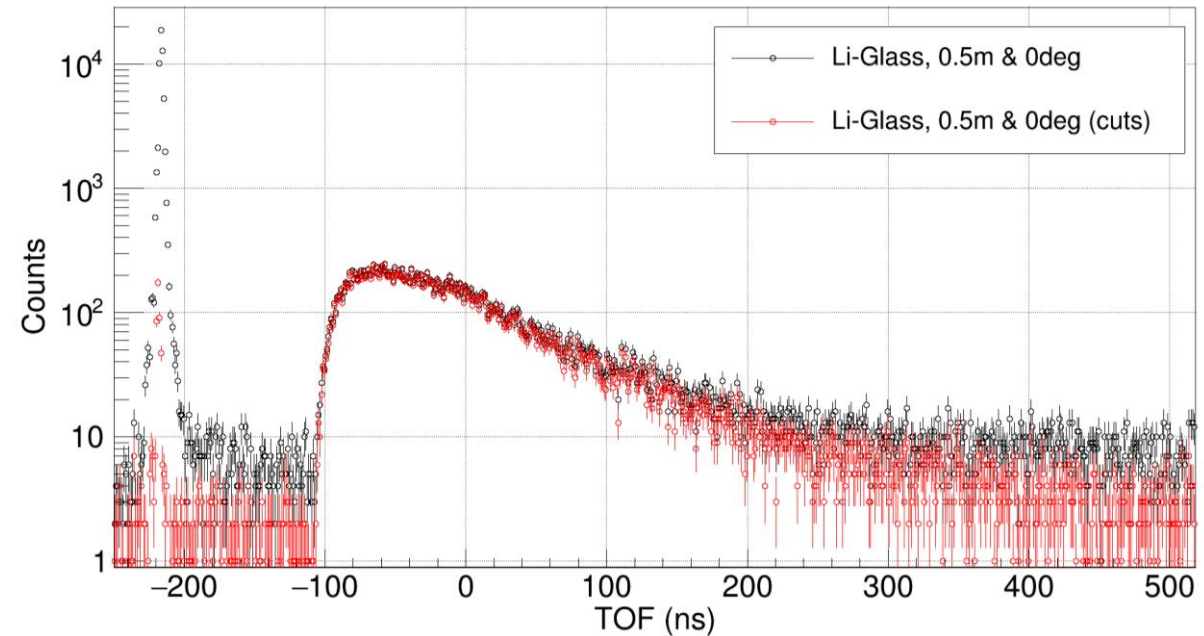
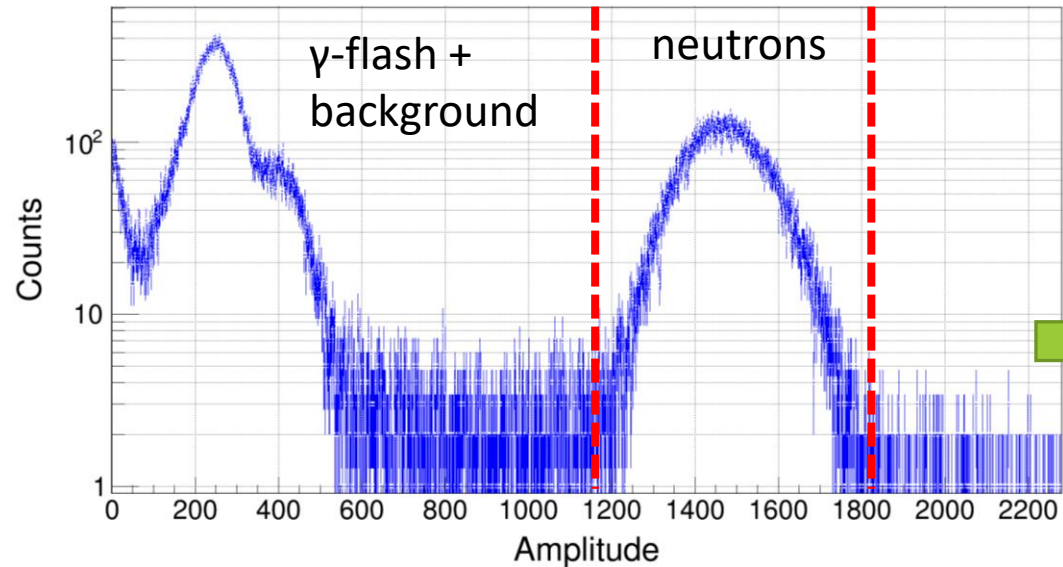


$^{197}\text{Au} + ^{50}\text{Cr} + ^{197}\text{Au}$ sample



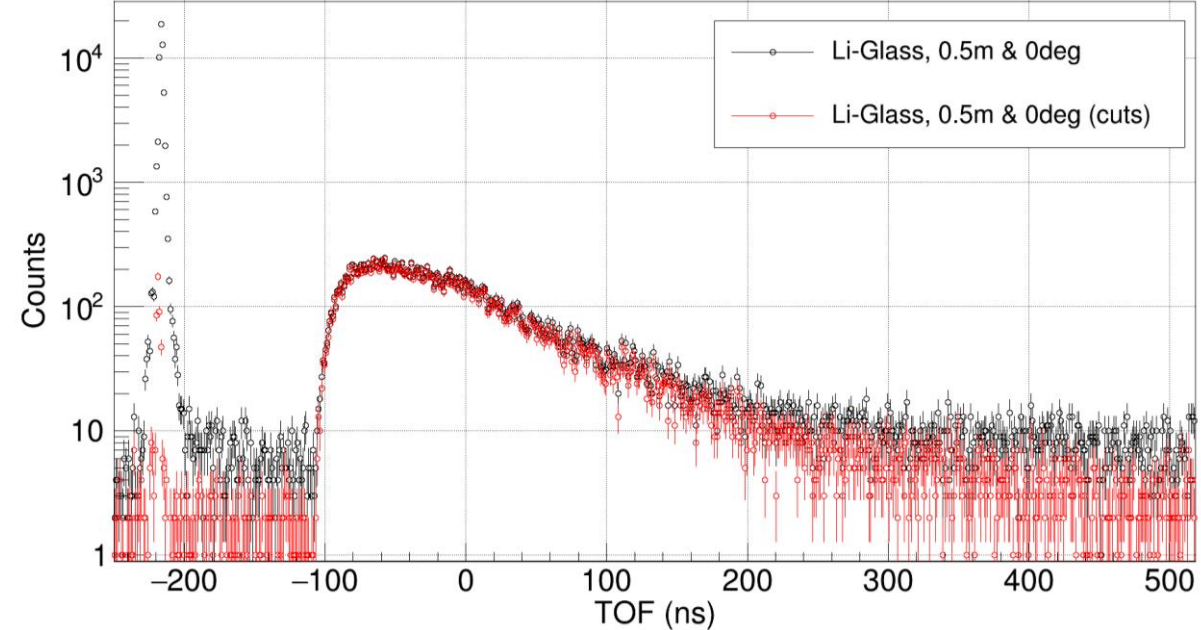
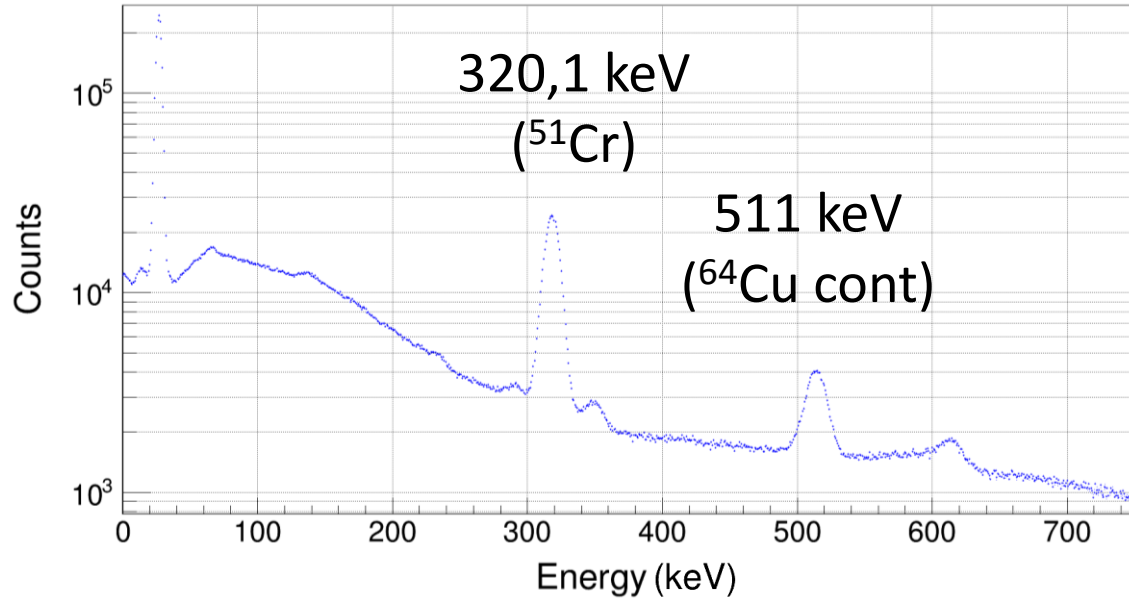
- 3 Lithium-glass neutron monitors
- 1 LaBr_3 for ^7Be decay
- 1 LaBr_3 for ^{198}Au and ^{51}Cr decay

^{50}Cr activation: preliminary results (1912 keV)



- Every neutron flux measured with Lithium-glass detectors at 3 angles.
- Flight paths of 0,5m ($E_p = 1912$ & 2080 keV) and 1m (the rest).
- With cuts in deposited energy we remove the gamma flash.

^{50}Cr activation: preliminary results (1912 keV)



- Every neutron flux measured with Lithium-glass detectors at 3 angles.
- Flight paths of 0,5m ($E_p = 1912$ & 2080 keV) and 1m (the rest).
- With cuts in deposited energy we remove the gamma flash.
- A lot of work ahead!

$$SACS = \frac{N_{198Au}}{N_{7Be} n_{at}}$$

$$MACS = \frac{2}{\sqrt{\pi}} \frac{\langle \sigma_{MB} \rangle}{\langle \sigma_{\Phi} \rangle} SACS$$

HiSPANoS SACS	584 mb*
HiSPANoS MACS	631 mb*
KADONIS MACS	612(6) mb
IAEA MACS	620(11) mb

*(Uncertainties not yet estimated)

Summary & Outlook

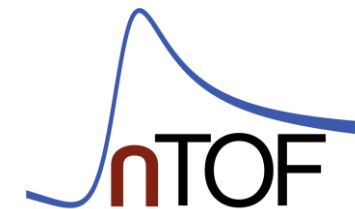
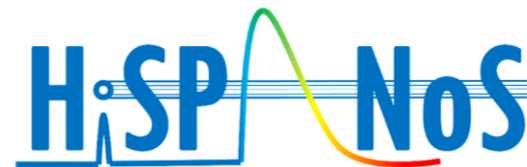
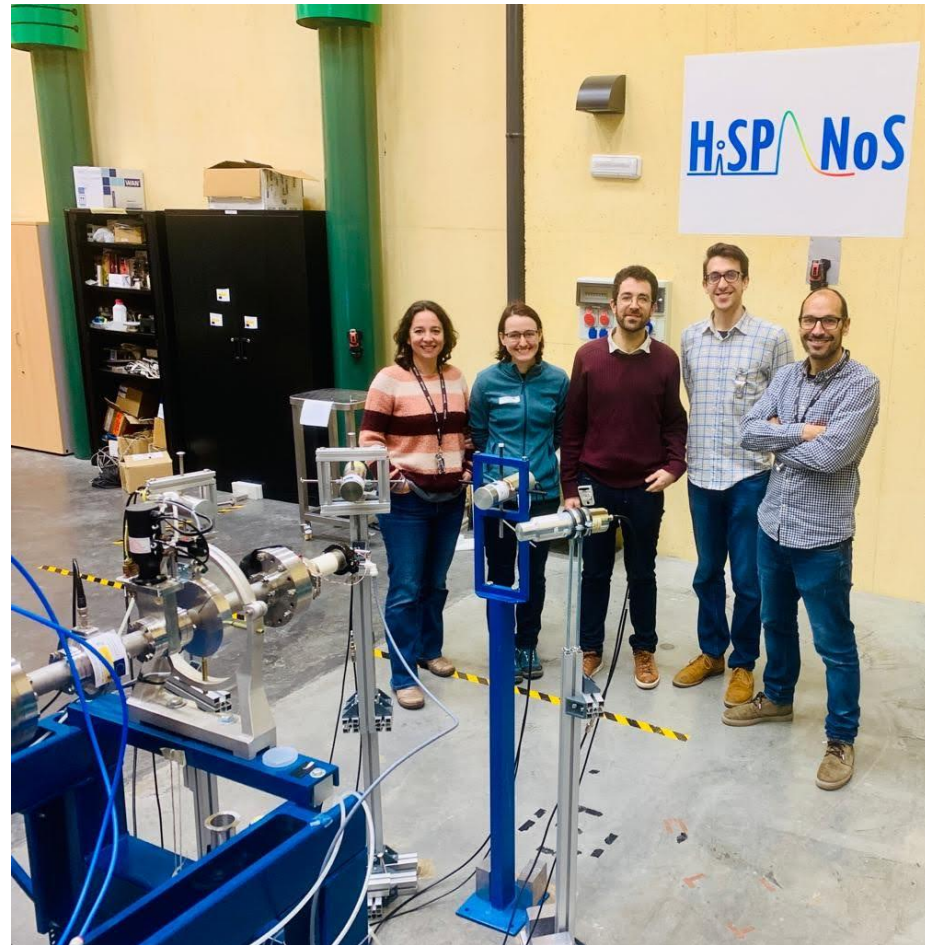
- The goal is to improve the $^{50,53}\text{Cr}(n,\gamma)$ cross section to 8-10% accuracy at 1-100 keV
- Two experiments:
 - n_TOF@CERN, Summer'22 (H2020-Ariel Scientific Visit).
 - HiSPANoS@CNA, March'23 (H2020-Ariel Transnational Access).
- Preliminary results show high quality data.
- Next steps:
 - Identify (and correct?) systematic effects
 - Counts/pulse \rightarrow Yield (capture/neutrons)
 - Resonance analysis with SAMMY
 - Activation @CNA data analysis (2023)



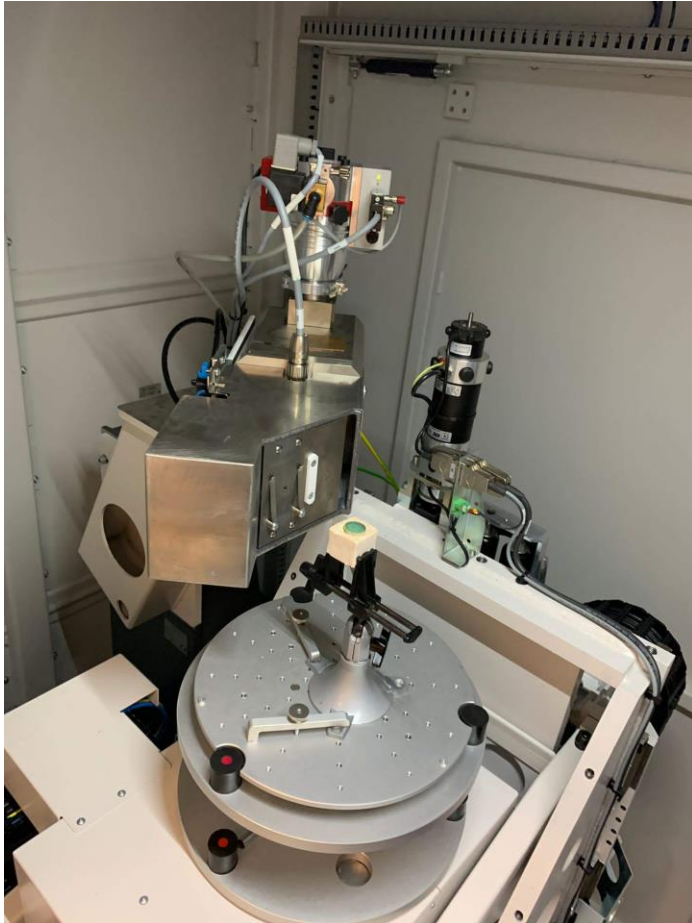
n_TOF experiment
data analysis (2023)

Thank you!

Pablo Pérez Maroto
ppmaroto@us.es



Backup. Tomography for homogeneity check



- Tomography of the samples → **MME Group (CERN)**
- Very helpful for determining the density and thickness of the samples.
- With the thinnest sample (^{50}Cr thin, ~ 240 mg) serious imperfections were observed → we were able to redo this sample.
- With simulations we will estimate **self-shielding** and **multiple scattering effects** and the cross section **uncertainty**.

Backup. Samples and detector set-up (EAR1)

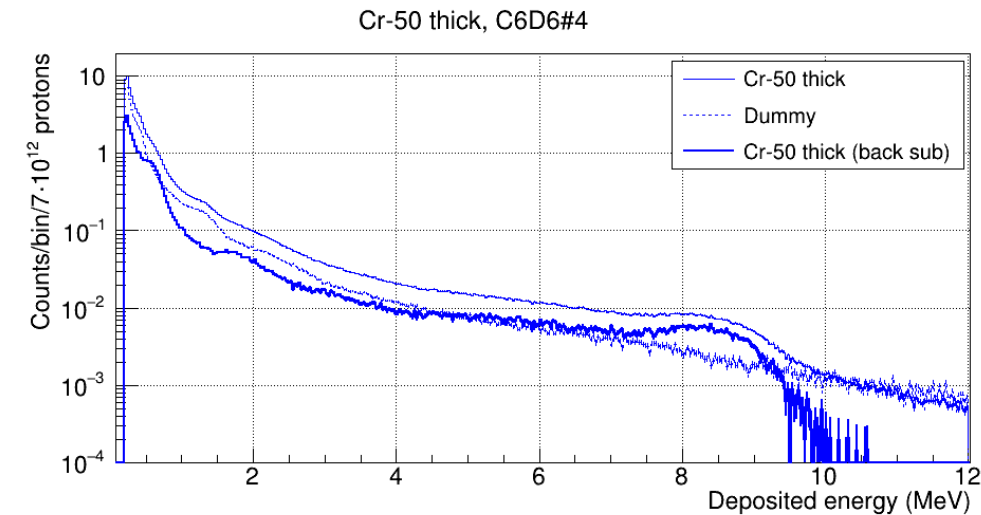
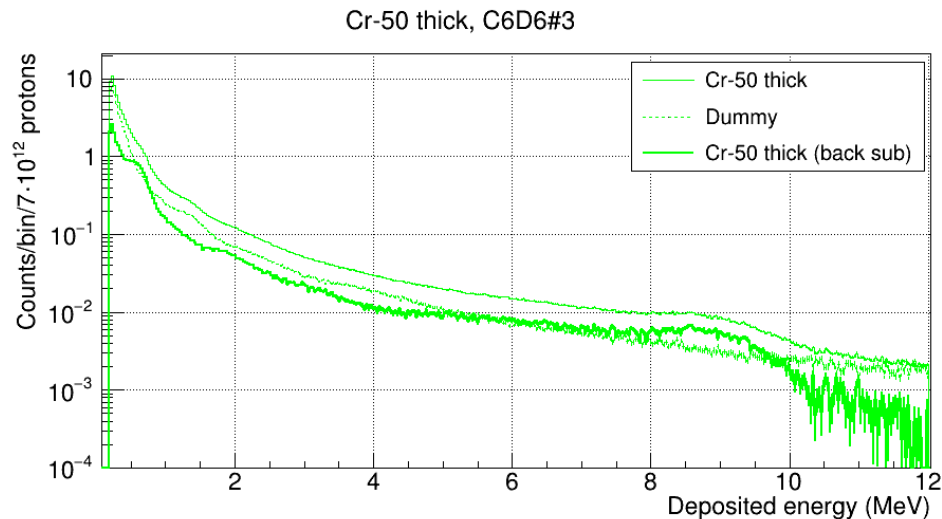
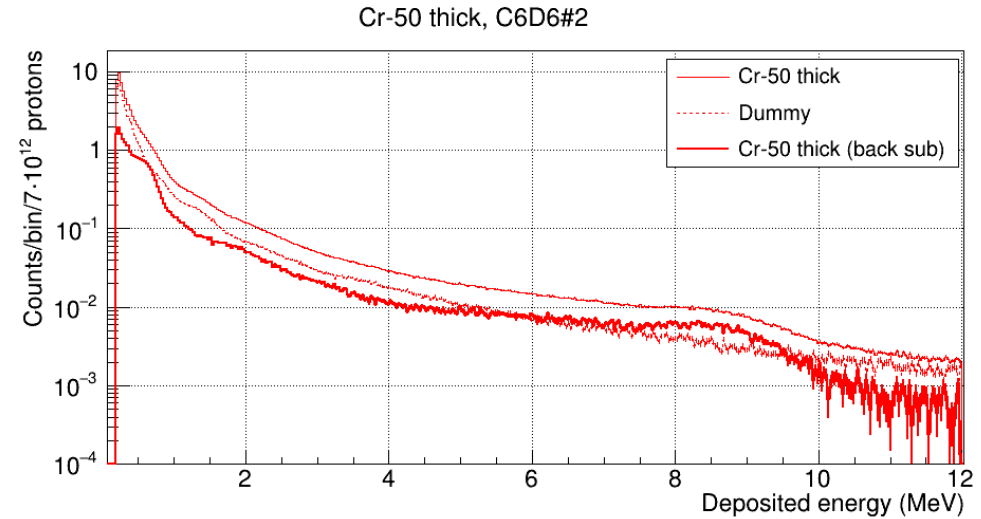
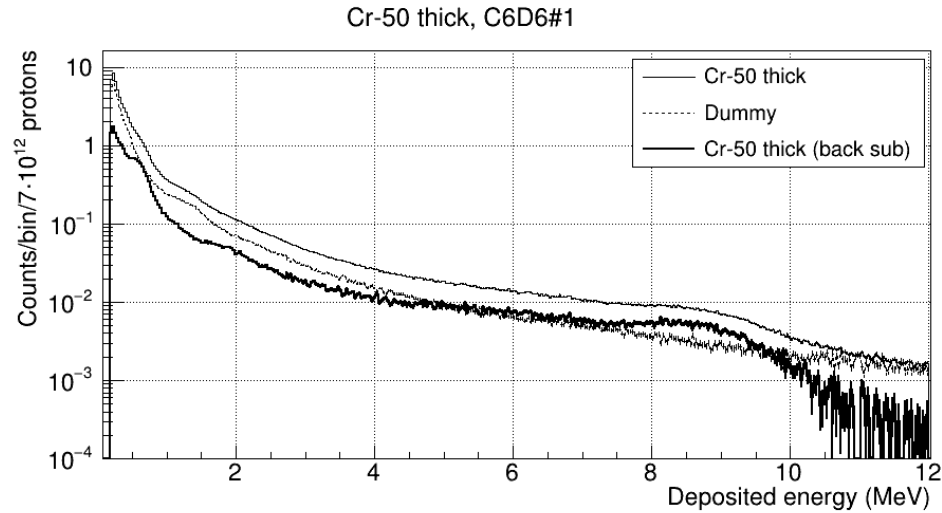
- 12 weeks of experiment (Summer '22)
- Stay funded by the H2020-ARIEL project
- 42 days of beam: 11/07 → 22/08
- $4,6 \cdot 10^{18}$ protons in total

Sample	Protons·10 ¹⁷ (meas.)	Protons·10 ¹⁷ (proposal)
⁵⁰ Cr – thin	5,5	5 (110%)
⁵⁰ Cr – thick	14,7	8 (184%)
⁵³ Cr – thin	6,8	5 (136%)
⁵³ Cr – thick	6,9	17 (40%)
Back. & norm.	13,0	5 (260%)
Total	45,9	40 (115%)

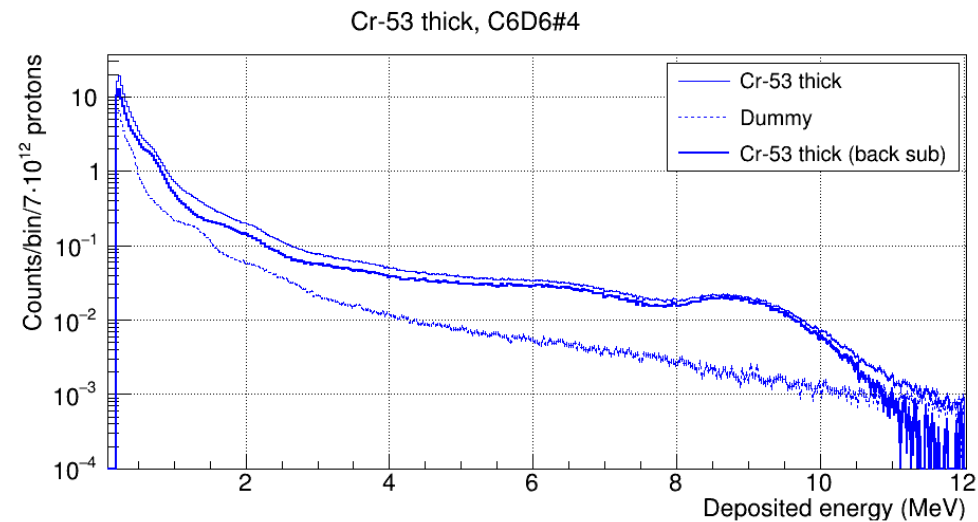
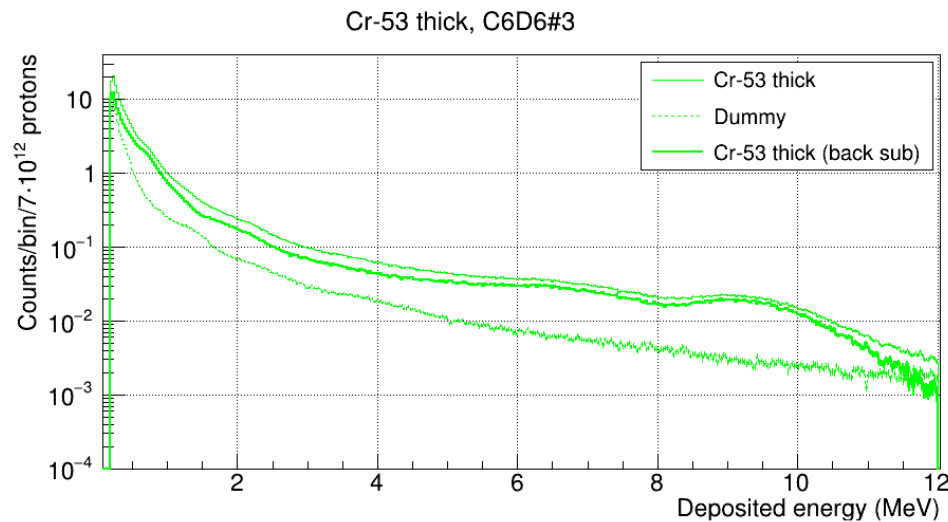
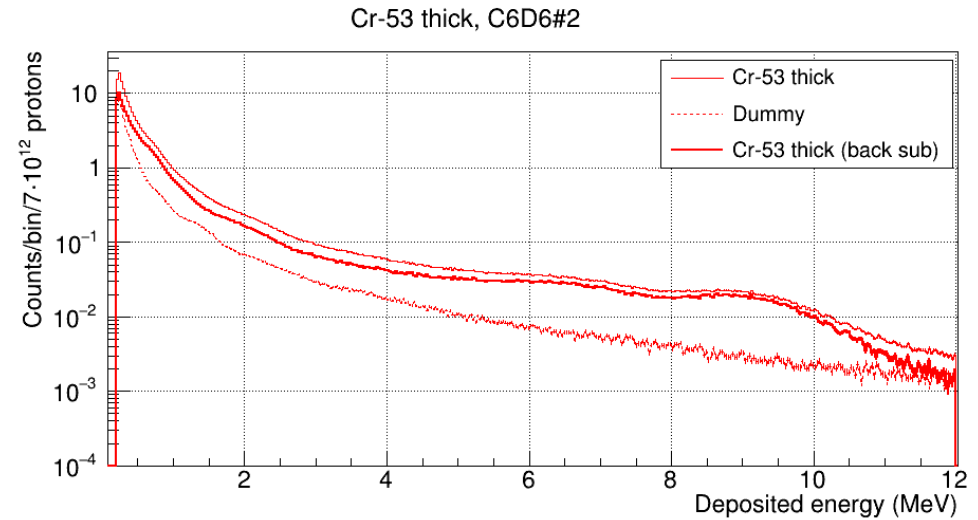
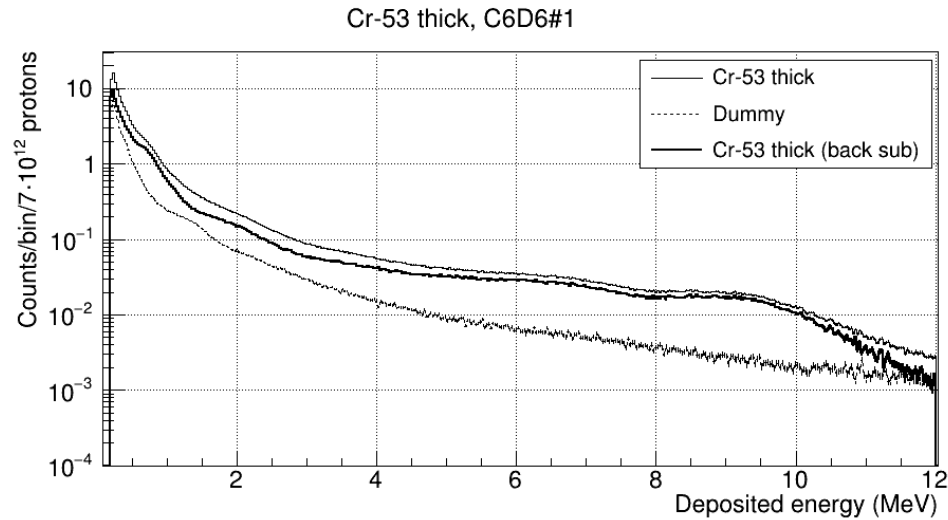
Complementary measurements

natCr	Resonance identification
¹⁹⁷ Au	Normalization
²⁷ Al	High E _γ calibration
Empty	Background
Dummy (x2)	Background
^{50,53} Cr & Dummy with filters	²⁰⁶ Bi and ²⁷ Al filters Background

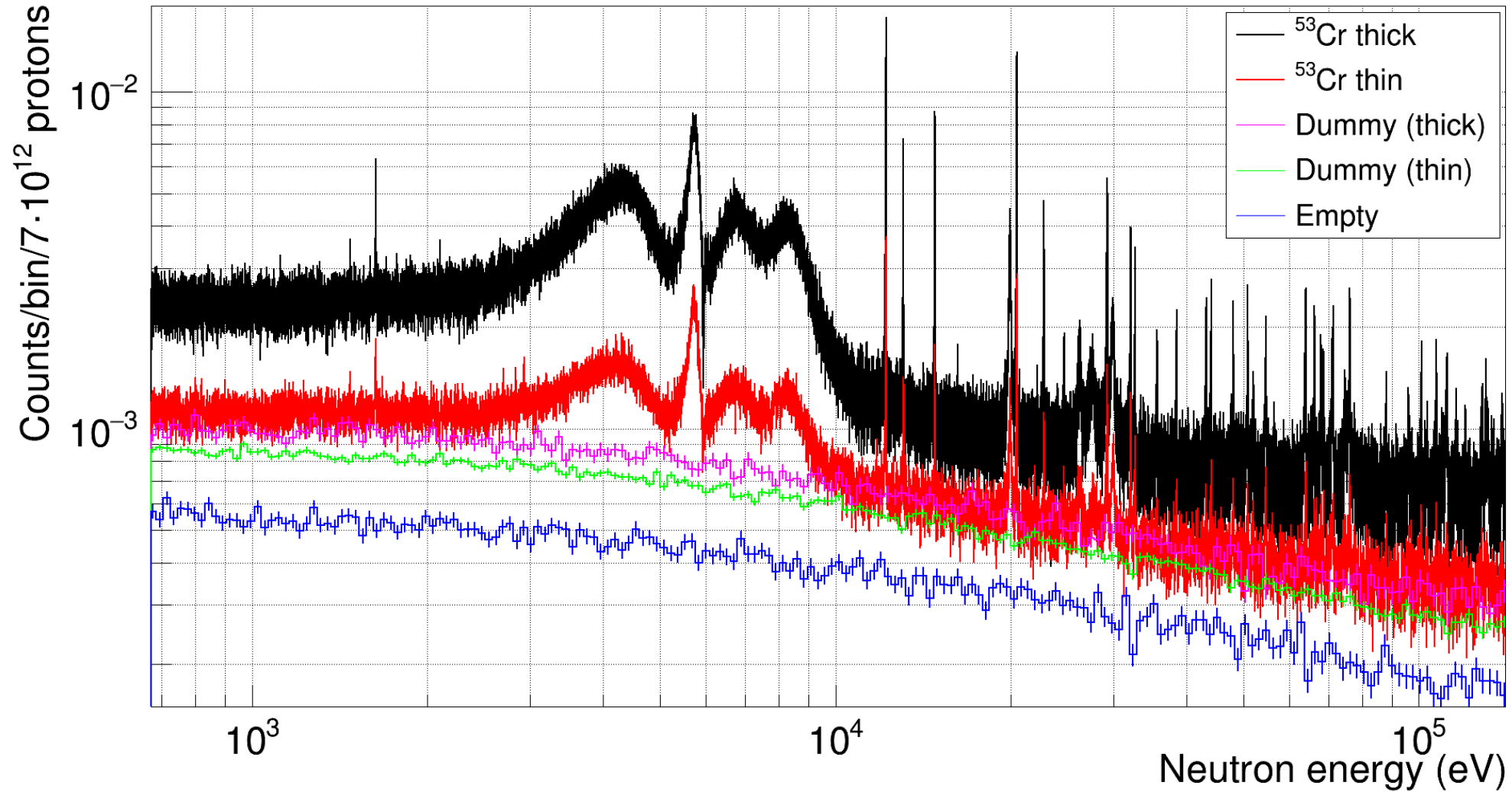
Backup. ^{50}Cr cascades ($S_n=9,3$ MeV)



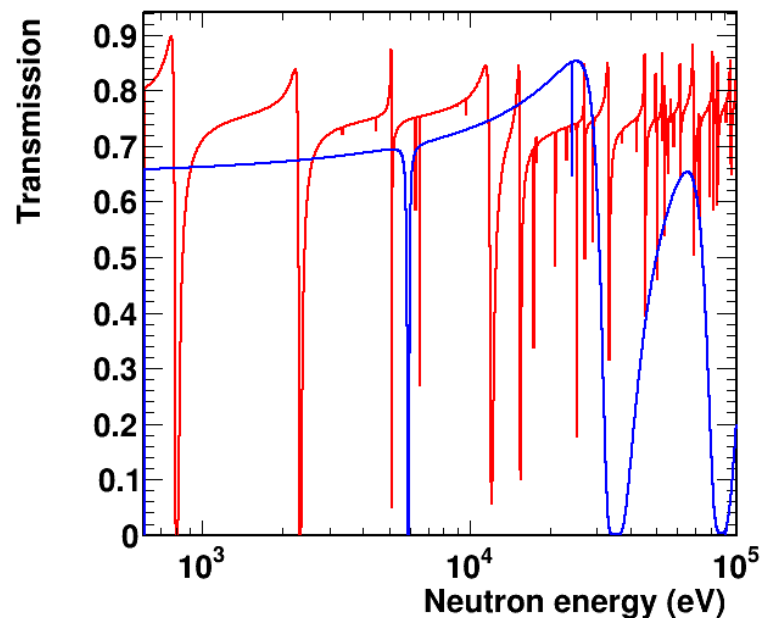
Backup. ^{53}Cr cascades ($S_n=9,7$ MeV)



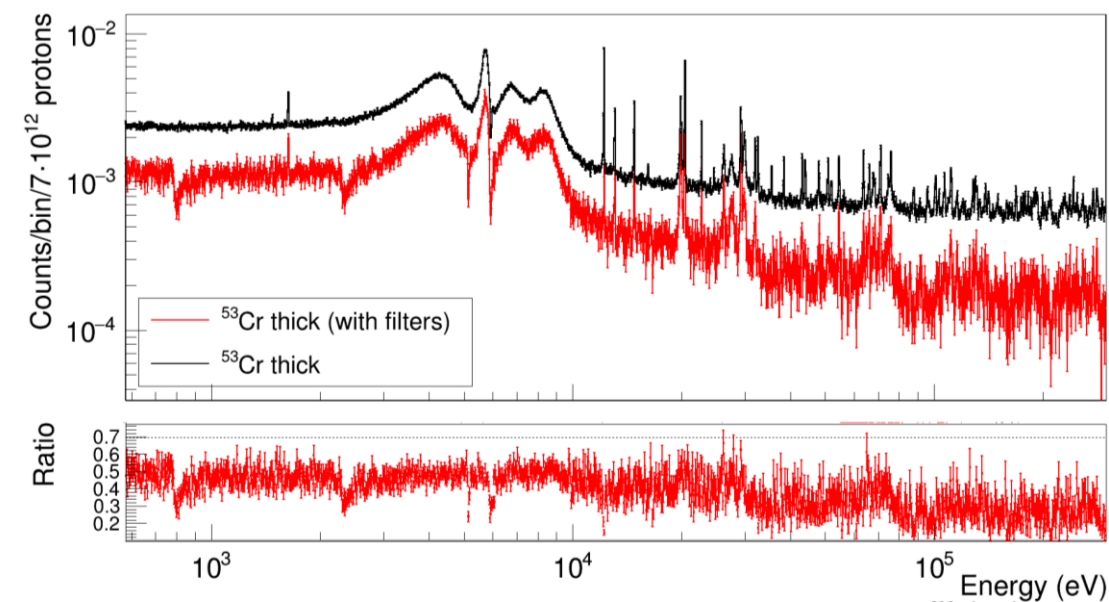
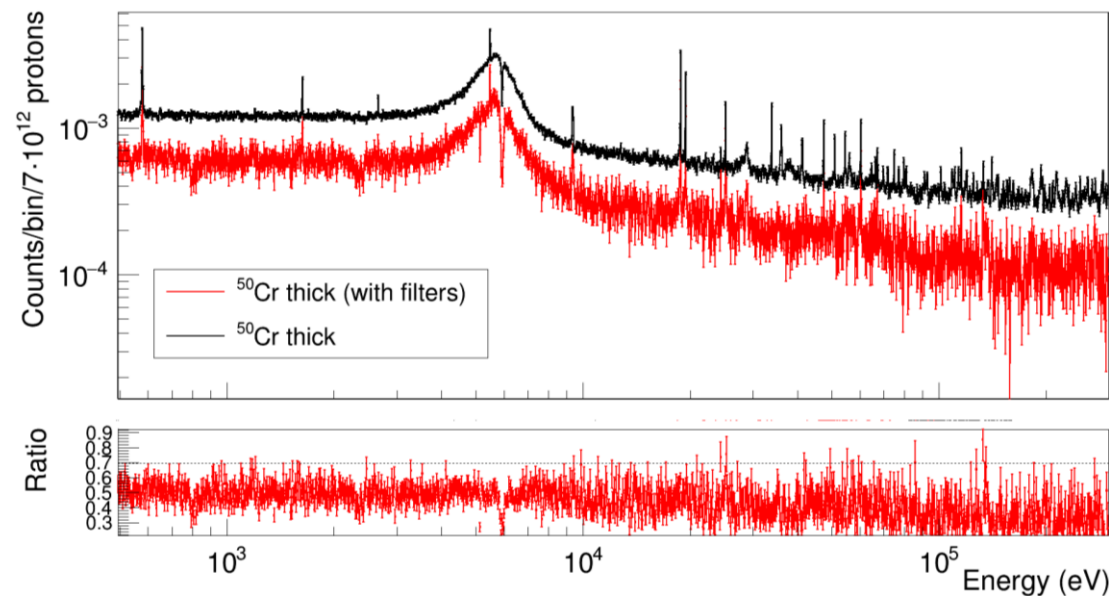
Backup. Preliminary results (^{53}Cr : thin vs. thick)



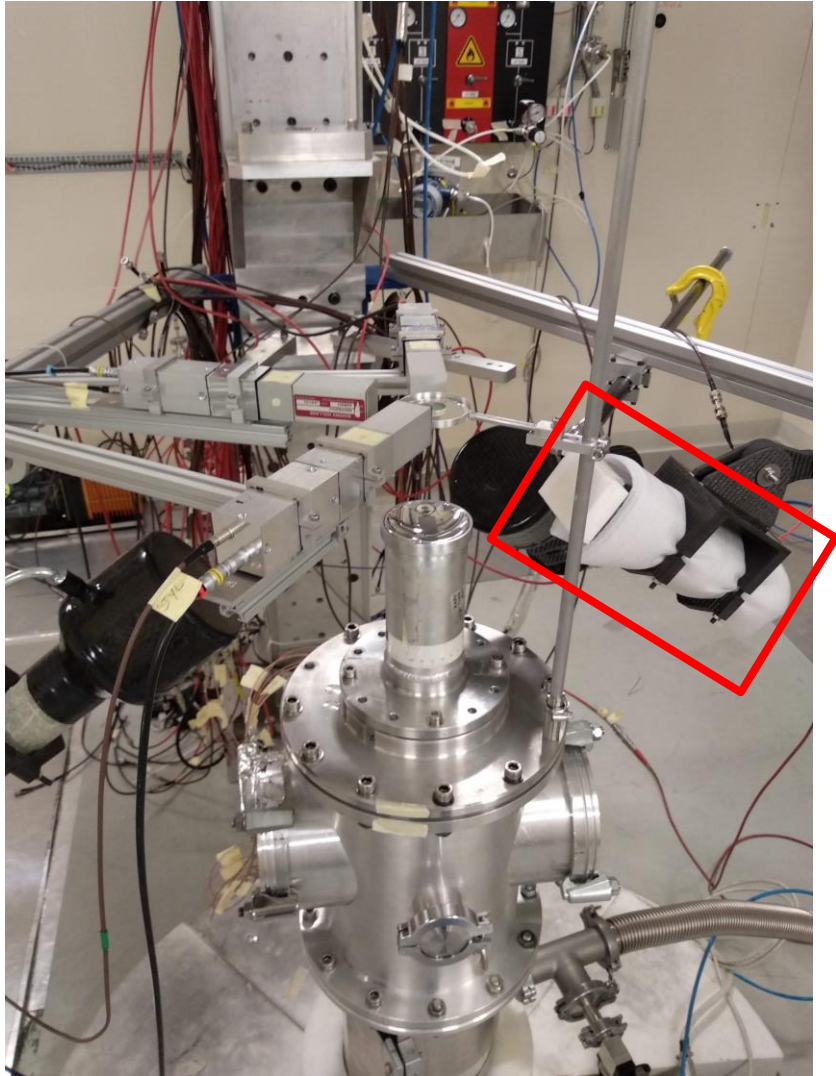
Backup. Filters



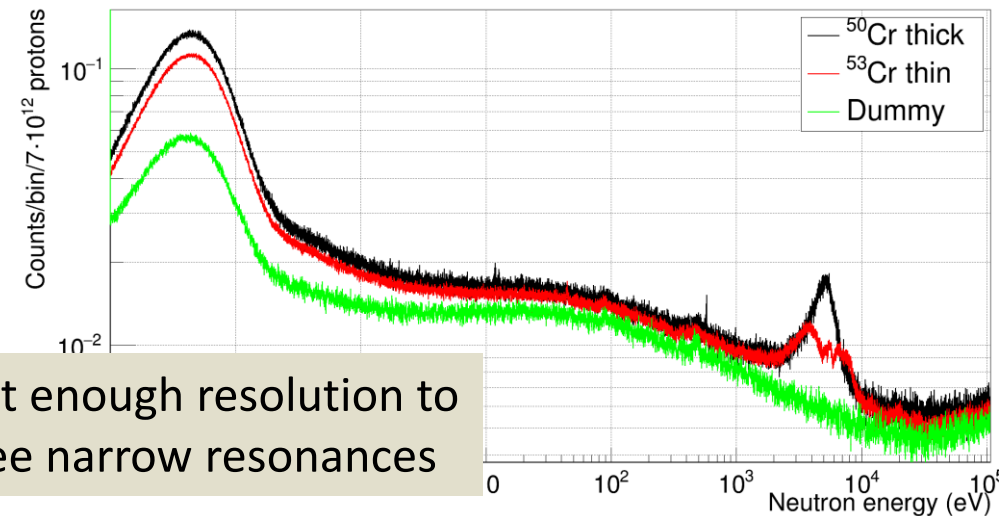
- For neutron scattering background
- 10 mm ^{209}Bi & 50mm ^{27}Al filters
- Dummy and thick samples



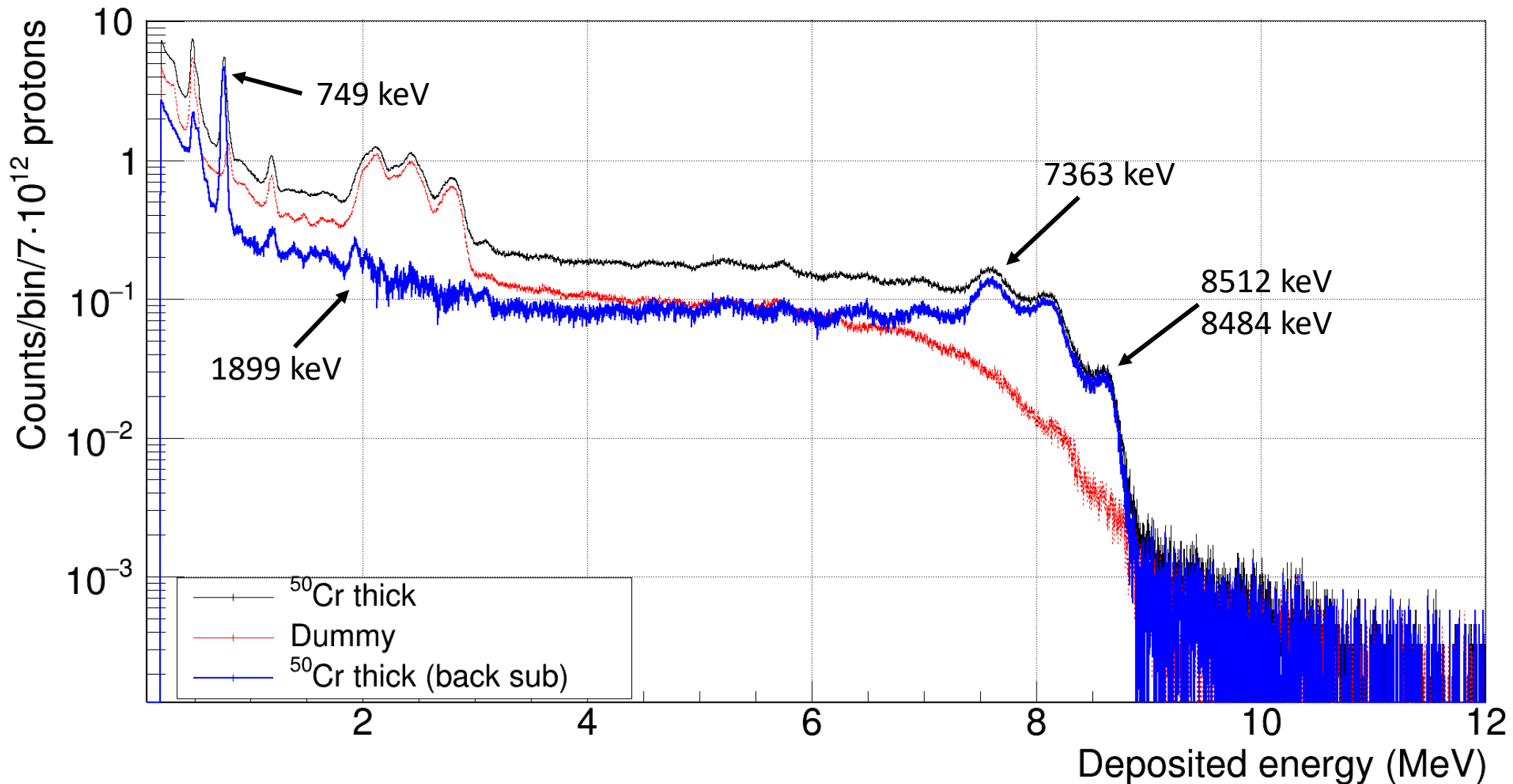
Backup. Cascades at EAR2



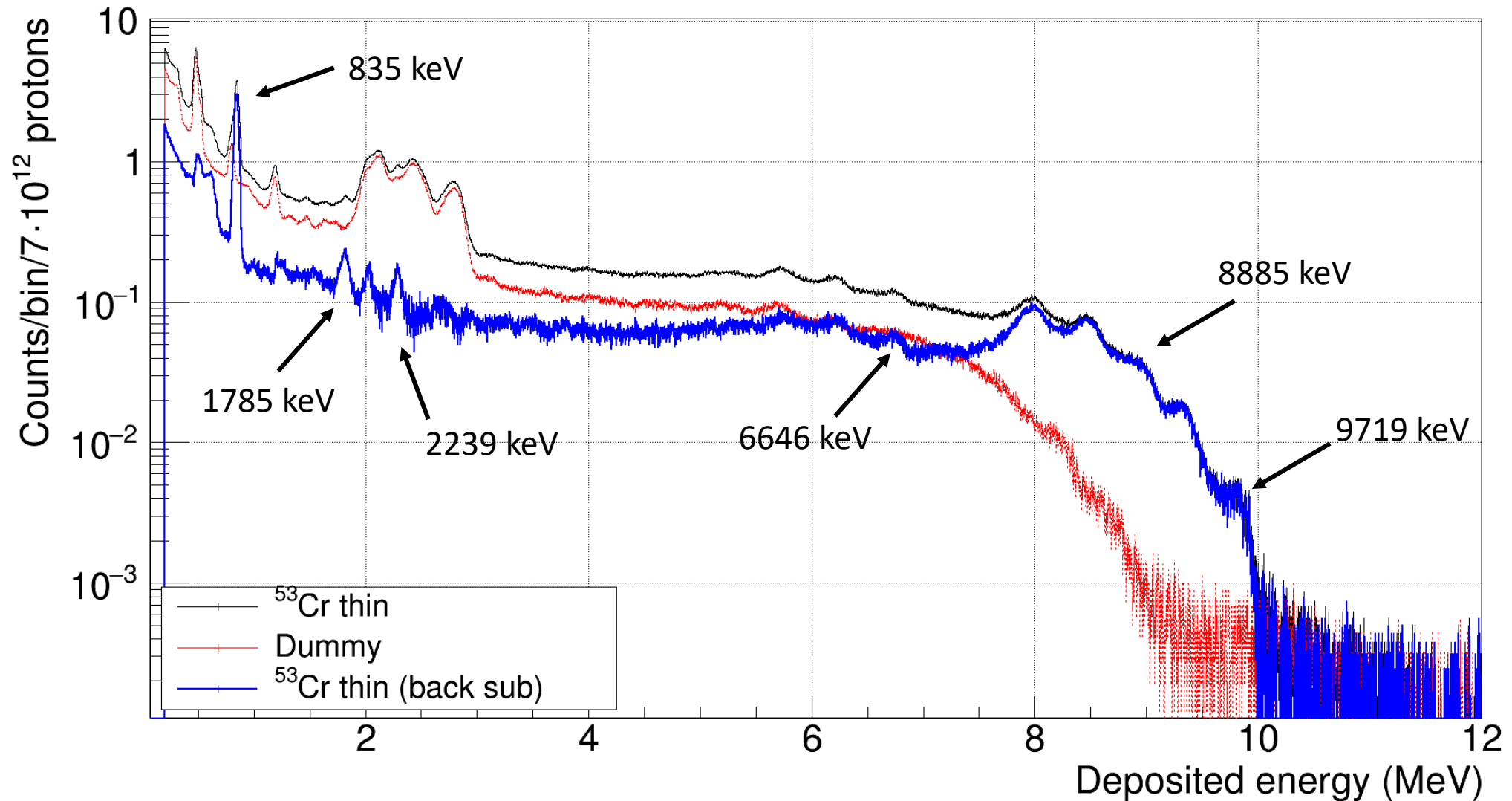
- Goal → measure the cascades with high resolution to validate future simulations.
- 3 sTED's & 2 C6D6's for monitoring, **1 LaCl₃** for the cascades.
- Gain shift depending on counting rate (malfunctioning PMT?) → thin samples, low voltage and not too close to the beam.



Backup. $^{50}\text{Cr}(n,\gamma)$ cascades at EAR2 (preliminary)



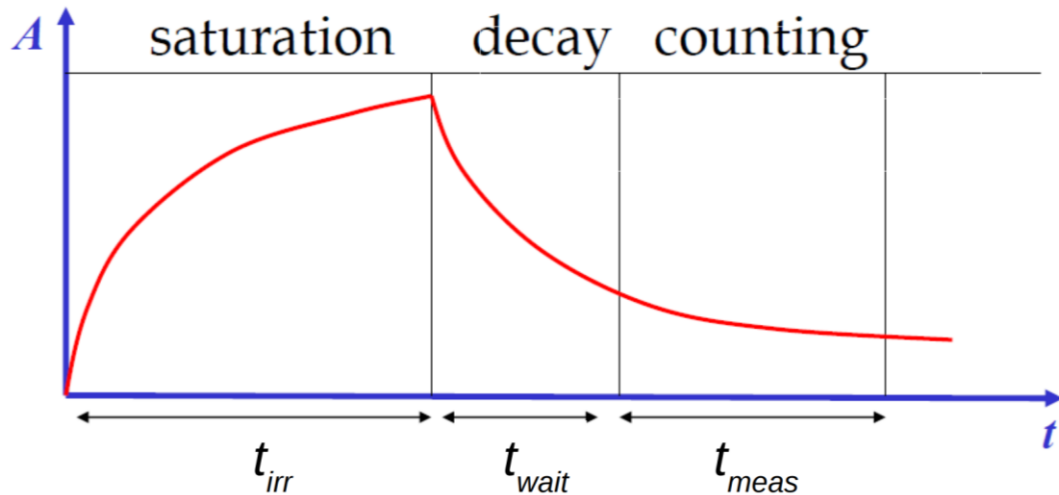
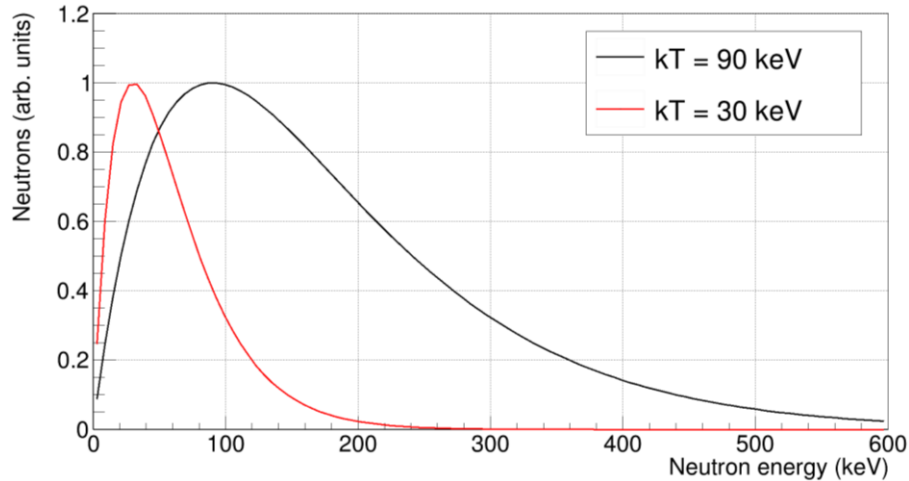
Backup. $^{53}\text{Cr}(n,\gamma)$ cascades at EAR2 (preliminary)



Backup. ^{54}Cr gamma emission

E_γ^\dagger	I_γ^\ddagger	$E_i(\text{level})$	J_i^π	E_f	J_f^π	Mult. [@]	$\delta^@$						
205.62 20	0.05 1	3925.59	2 ⁺	3719.99	2 ⁺			3393.35 7	0.67 6	3393.42	2 ⁺	0.0	0 ⁺
745.37 16	0.06 1	4872.36	2 ⁺	4127.08	3 ⁻			3403.55 9	0.17 7	(9720.18)	(1 ⁻)	6316.42	
^x 789.22 2	0.07 1							^x 3509.86 17	0.21 2				
817.20 7	0.07 1	3436.88	2 ⁺	2619.69	2 ⁺			3545.92 13	0.32 4	4380.74	2 ⁻	834.879	2 ⁺
834.87 2	79.0 [#] 2	834.879	2 ⁺	0.0	0 ⁺			3576.08 9	0.20 4	(9720.18)	(1 ⁻)	6143.59	
843.57 12	0.17 2	5226.22	2 ⁻	4380.74	2 ⁻			3719.84 7	3.69 [#] 2	3719.99	2 ⁺	0.0	0 ⁺
847.90 17	0.08 1	6143.59		5294.47	1 ⁺ ,(2 ⁺)			3863.64 11	0.39 5	(9720.18)	(1 ⁻)	5856.39	
890.41 2	0.43 3	3719.99	2 ⁺	2829.56	0 ⁺			3898.51 14	0.11 2	(9720.18)	(1 ⁻)	5821.49	
944.57 19	0.03 1	4872.36	2 ⁺	3927.70	2 ⁺			3927.57 9	0.52 7	3927.70	2 ⁺	0.0	0 ⁺
946.80 15	0.05 1	4872.36	2 ⁺	3925.59	2 ⁺			4133.15 8	0.48 5	(9720.18)	(1 ⁻)	5586.92	1 ⁺ ,2 ⁺
989.08 2	0.76 5	1823.96	4 ⁺	834.879	2 ⁺	E2		^x 4168.1 6	0.12 4				
1100.38 6	0.64 4	3719.99	2 ⁺	2619.69	2 ⁺			^x 4229.9 3	0.10 4				
1106.38 10	0.02 1	5189.62	2 ⁺	4083.24	3 ⁺			^x 4393.28 9	0.06 4				
^x 1205.33 10	0.05 1							4425.63 16	0.50 6	(9720.18)	(1 ⁻)	5294.47	1 ⁺ ,(2 ⁺)
1241.36 7	0.78 5	3861.02	2 ⁺	2619.69	2 ⁺			4433.43 21	0.20 3	5268.47	2 ⁺	834.879	2 ⁺
1335.26 6	0.06 1	3159.21	4 ⁺	1823.96	4 ⁺			4451.47 18	0.45 5	(9720.18)	(1 ⁻)	5268.47	2 ⁺
1340.81 10	0.12 2	5268.47	2 ⁺	3927.70	2 ⁺			4459.28 21	0.38 5	5294.47	1 ⁺ ,(2 ⁺)	834.879	2 ⁺
1435.49 18	0.23 2	4872.36	2 ⁺	3436.88	2 ⁺			4494.00 14	0.13 5	(9720.18)	(1 ⁻)	5226.22	2 ⁺
1460.10 14	0.04 2	5586.92	1 ⁺ ,2 ⁺	4127.08	3 ⁻			4530.38 21	0.19 5	(9720.18)	(1 ⁻)	5189.62	2 ⁺
1463.33 14	0.07 2	4083.24	3 ⁺	2619.69	2 ⁺			4751.83 10	0.18 4	5586.92	1 ⁺ ,2 ⁺	834.879	2 ⁺
1503.62 9	0.06 2	5586.92	1 ⁺ ,2 ⁺	4083.24	3 ⁺			4847.54 11	1.96 7	(9720.18)	(1 ⁻)	4872.36	2 ⁺
1508.24 25	0.06 2	4127.08	3 ⁻	2619.69	2 ⁺			4872.27 10	1.06 8	4872.36	2 ⁺	0.0	0 ⁺
1597.72 4	0.03 2	4217.56	(2 ⁺),3 ⁺	2619.69	2 ⁺			5021.29 34	0.16 6	5856.39		834.879	2 ⁺
^x 1619.17 7	0.09 2							5086.36 12	0.23 6	(9720.18)	(1 ⁻)	4633.57	2 ⁺
1784.69 5	10.14 [#] 4	2619.69	2 ⁺	834.879	2 ⁺	M1+E2	-0.53 18	5339.27 18	0.29 4	(9720.18)	(1 ⁻)	4380.74	2 ⁻
1798.22 5	0.25 2	4872.36	2 ⁺	3074.06	2 ⁺			5501.78 26	0.13 2	(9720.18)	(1 ⁻)	4217.56	(2 ⁺),3 ⁺
1804.00 14	0.24 2	4633.57	2 ⁺	2829.56	0 ⁺			5636.90 42	0.13 3	(9720.18)	(1 ⁻)	4083.24	3 ⁺
1831.34 17	0.03 2	5268.47	2 ⁺	3436.88	2 ⁺			5707.09 12	1.35 11	(9720.18)	(1 ⁻)	4012.87	0 ⁺
1994.56 5	2.93 15	2829.56	0 ⁺	834.879	2 ⁺	E2		5792.2 6	0.46 7	(9720.18)	(1 ⁻)	3927.70	2 ⁺
2066.99 7	0.04 2	5226.22	2 ⁺	3159.21	4 ⁺			5794.3 4	0.17 5	(9720.18)	(1 ⁻)	3925.59	2 ⁺
2101.43 12	0.10 2	5821.49		3719.99	2 ⁺			5858.98 14	1.21 8	(9720.18)	(1 ⁻)	3861.02	2 ⁺
2233.09 6	0.07 3	6316.42		4083.24	3 ⁺			5999.95 13	4.49 [#] 4	(9720.18)	(1 ⁻)	3719.99	2 ⁺
2239.07 5	10.70 [#] 5	3074.06	2 ⁺	834.879	2 ⁺	M1+E2	0.02 5	6283.02 14	2.03 14	(9720.18)	(1 ⁻)	3436.88	2 ⁺
2259.22 5	0.21 2	4083.24	3 ⁺	1823.96	4 ⁺			6326.11 14	1.19 12	(9720.18)	(1 ⁻)	3393.42	2 ⁺
2393.70 7	0.10 2	4217.56	(2 ⁺),3 ⁺	1823.96	4 ⁺			6645.64 13	9.71 [#] 8	(9720.18)	(1 ⁻)	3074.06	2 ⁺
2464.23 19	0.09 3	5294.47	1 ⁺ ,(2 ⁺)	2829.56	0 ⁺			6890.16 15	2.35 16	(9720.18)	(1 ⁻)	2829.56	0 ⁺
2558.45 5	1.15 7	3393.42	2 ⁺	834.879	2 ⁺			7100.11 14	7.61 [#] 7	(9720.18)	(1 ⁻)	2619.69	2 ⁺
2601.91 8	2.31 13	3436.88	2 ⁺	834.879	2 ⁺	M1+E2	-0.11 +12-16	8884.81 18	44.4 [#] 6	(9720.18)	(1 ⁻)	834.879	2 ⁺
2619.57 9	0.42 3	2619.69	2 ⁺	0.0	0 ⁺			9718.79 19	15.8 [#] 2	(9720.18)	(1 ⁻)	0.0	0 ⁺

Backup. Averaged cross section equations



$$N_{act} = \frac{A_{EOI}}{\lambda} = \frac{\dot{C} t_{meas} e^{\lambda t_{wait}}}{I_{\gamma} \varepsilon (1 - e^{-\lambda t_{meas}})}$$

$$\varepsilon = \varepsilon_{\lambda} K_{\Omega} K_{\gamma}$$

$$\sigma_{exp} = \frac{1}{n_{at}} \frac{N_{act}}{N_{neutrons}} = \frac{1}{n_{at}} \frac{N_{act}}{N_{act} {}^7Be}$$

$$\langle \sigma \rangle = \frac{\int \sigma(E_n) \Phi(E_n) dE_n}{\int \Phi(E_n) dE_n}$$

Backup. ^{50}Cr activation: preliminary results

