



M2 PSA Internship Defense 2024

Exploration of the Island of Stability: Spectroscopic Studies of Superheavy Elements and Synthesis of New Elements Beyond $Z = 118$

Under the supervision of Pr. Olivier Dorvaux

Linda Müller

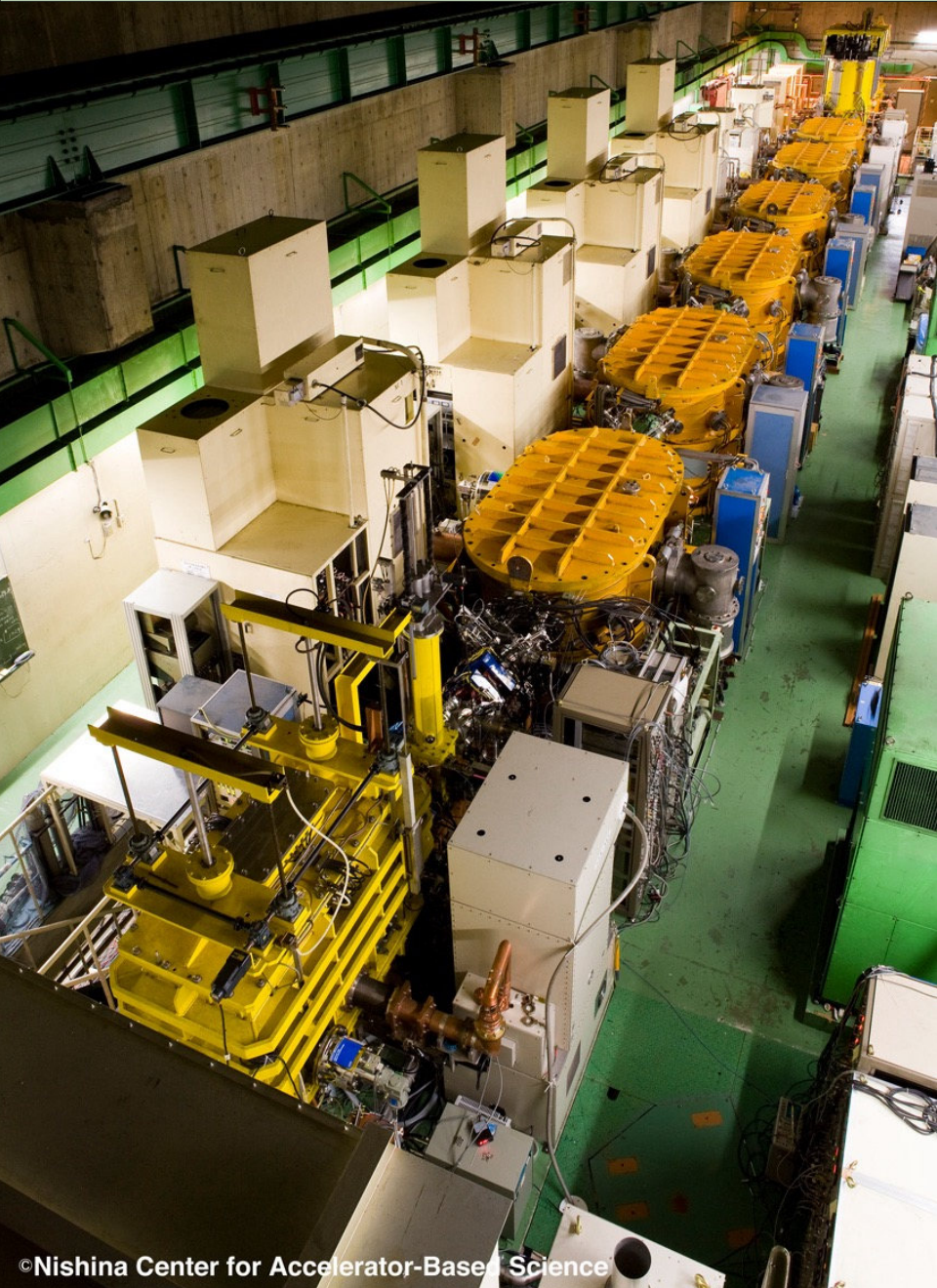
M2 Physique Subatomique et Astroparticules

University of Strasbourg



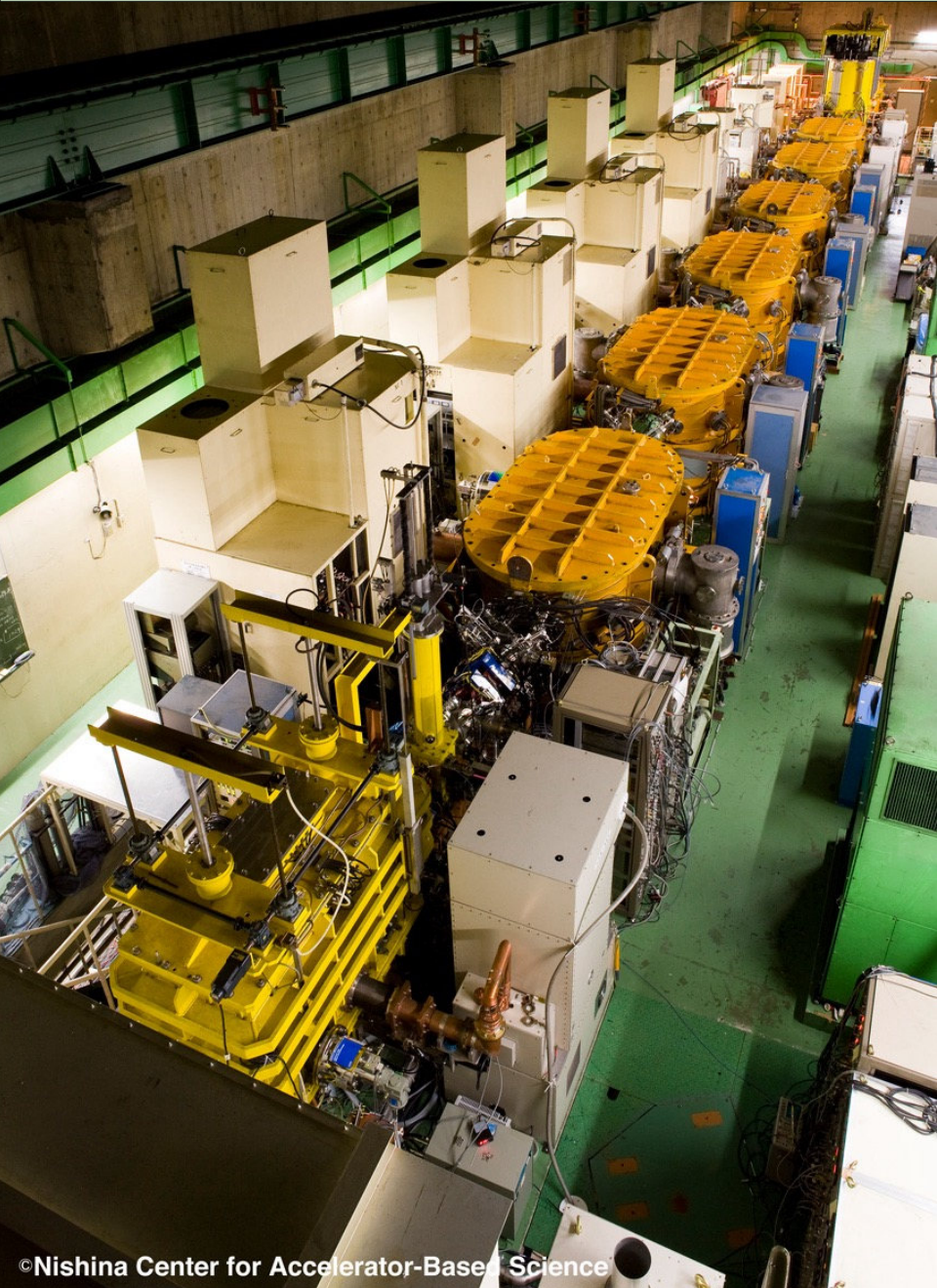
Photograph: LINAC at the Nishina Center for Accelerator Based Science ([link](#))

Presentation Outline



1. Definition and motivation
2. Synthesizing Superheavy Elements: Challenges & Experimental Setup
3. Data Selection
4. Results
 - Characterizing an Ion Separator with an Excitation Function
 - Identifying Rare Event Decay Chains
5. Conclusion and Further Perspectives
6. Figure References and Links

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What are Superheavy Elements?

Chart of Known Nuclides

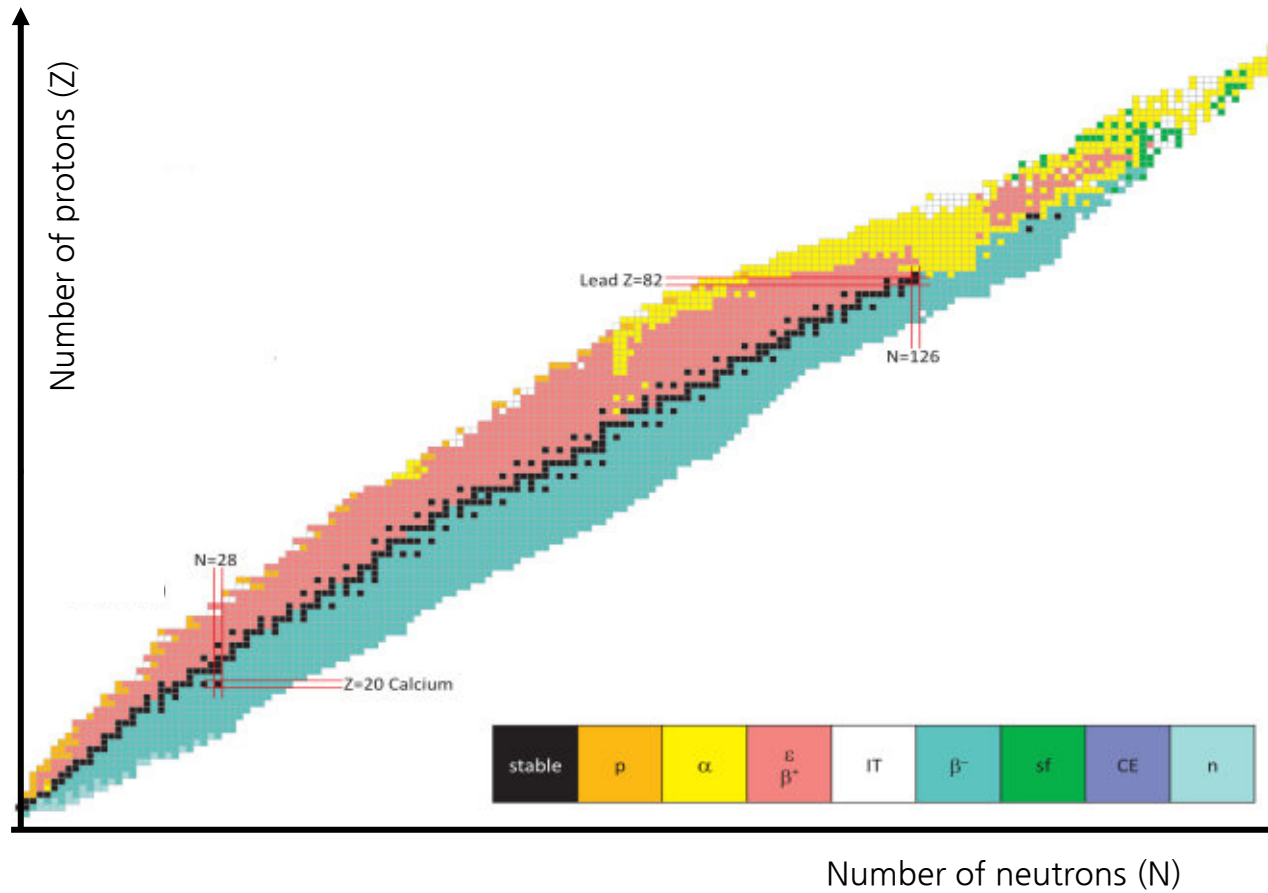


Diagram adapted from Karlsruhe Nuclide Chart – New 10th edition 2018 [1]

What are Superheavy Elements?

Chart of Known Nuclides

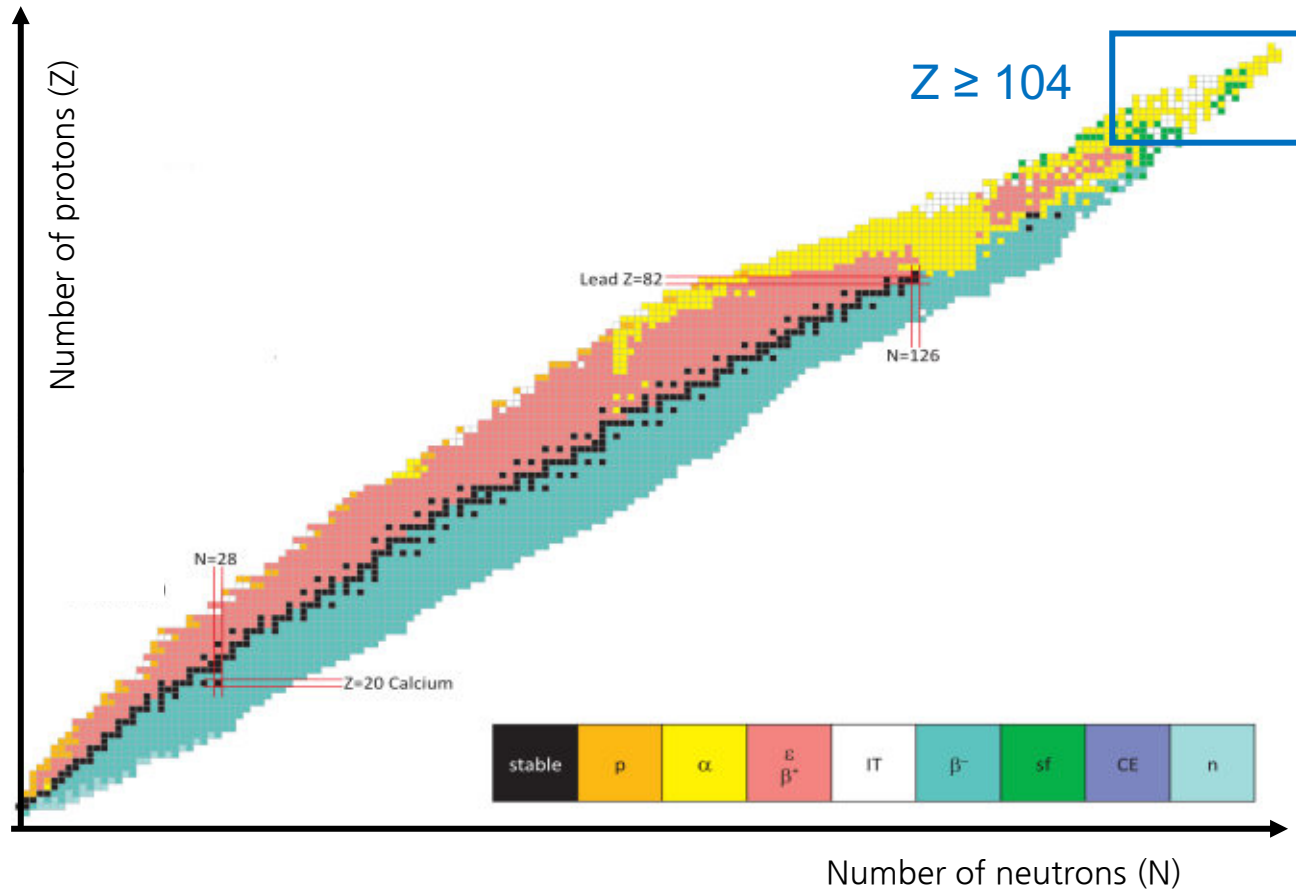
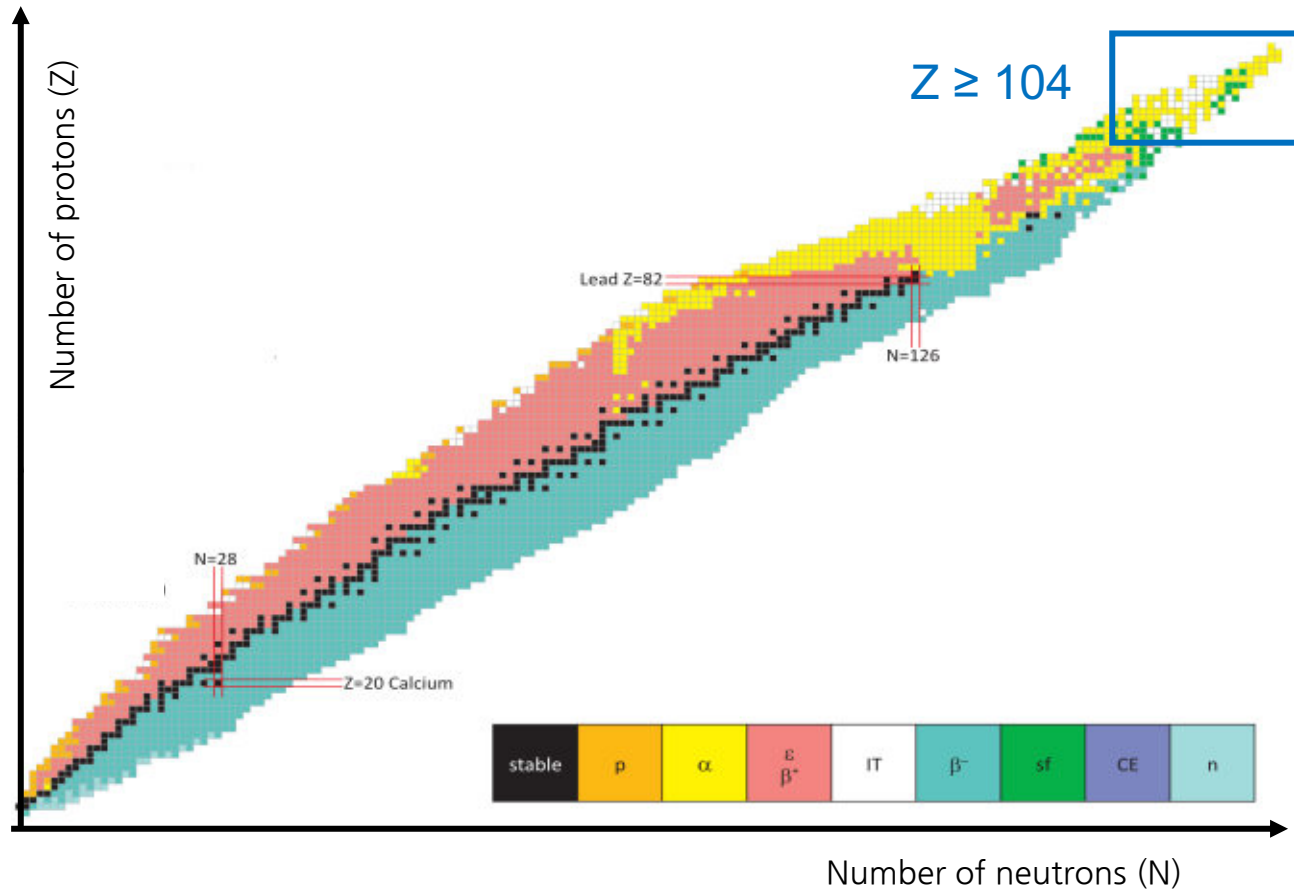


Diagram adapted from Karlsruhe Nuclide Chart – New 10th edition 2018 [1]

What are Superheavy Elements?

Chart of Known Nuclides



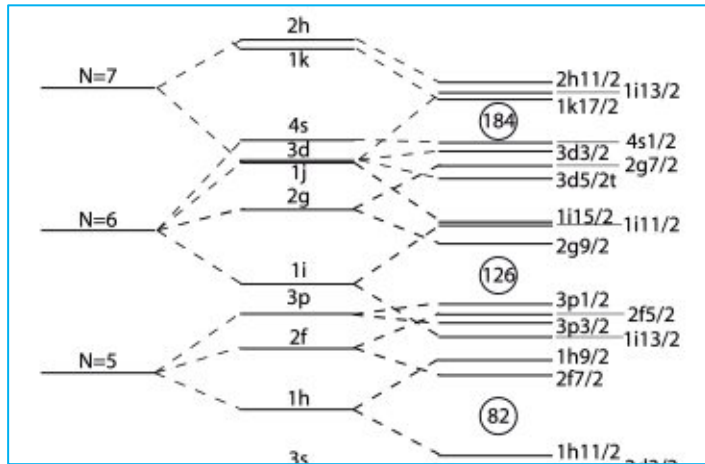
Immense Coulomb repulsion
↓
Stability explainable through
quantum effects only!

Diagram adapted from Karlsruhe Nuclide Chart – New 10th edition 2018 [1]

Why Study Superheavy Elements?

Reason 1: To refine our understanding of **nuclear** structure by the finding the next **shell gaps**!

Diagram adapted from Nilsson et al. (1969) [2]

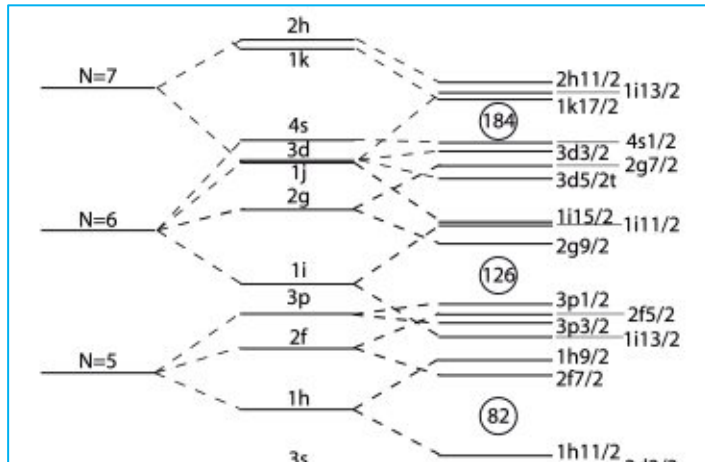


Shell model → spherical gaps

Why Study Superheavy Elements?

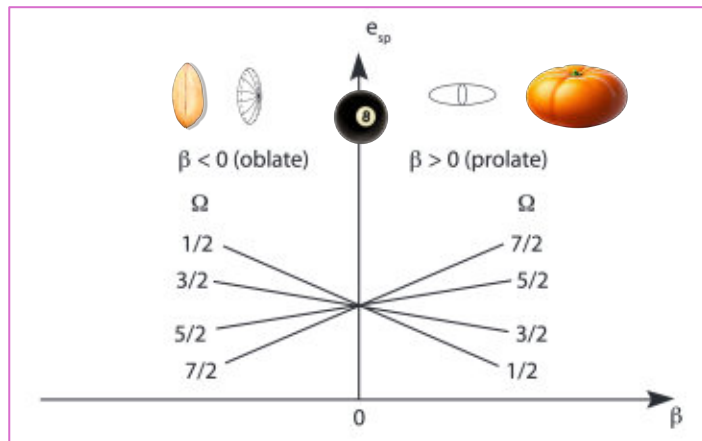
Reason 1: To refine our understanding of **nuclear** structure by the finding the next **shell gaps**!

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Shell model → spherical gaps

Diagram adapted from Piot-Julien, Thesis (2010) [3]

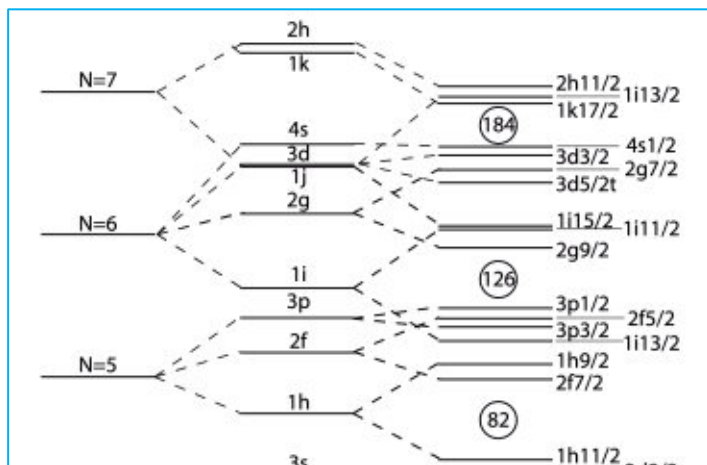


Nuclear deformation → deformed gaps

Why Study Superheavy Elements?

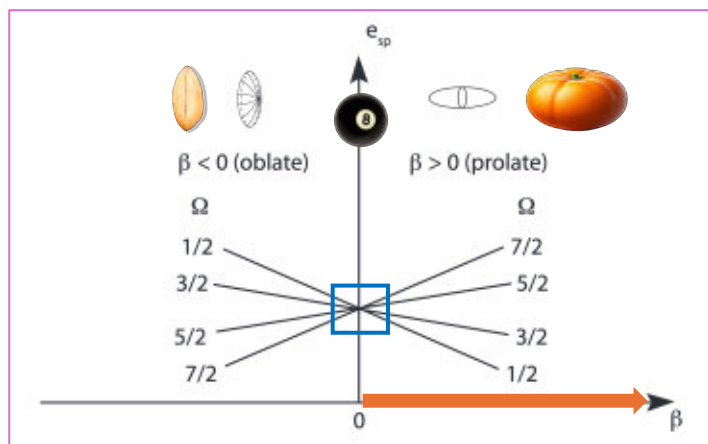
Reason 1: To refine our understanding of **nuclear** structure by the finding the next **shell gaps**!

Diagram adapted from Nilsson et al. (1969) [2]



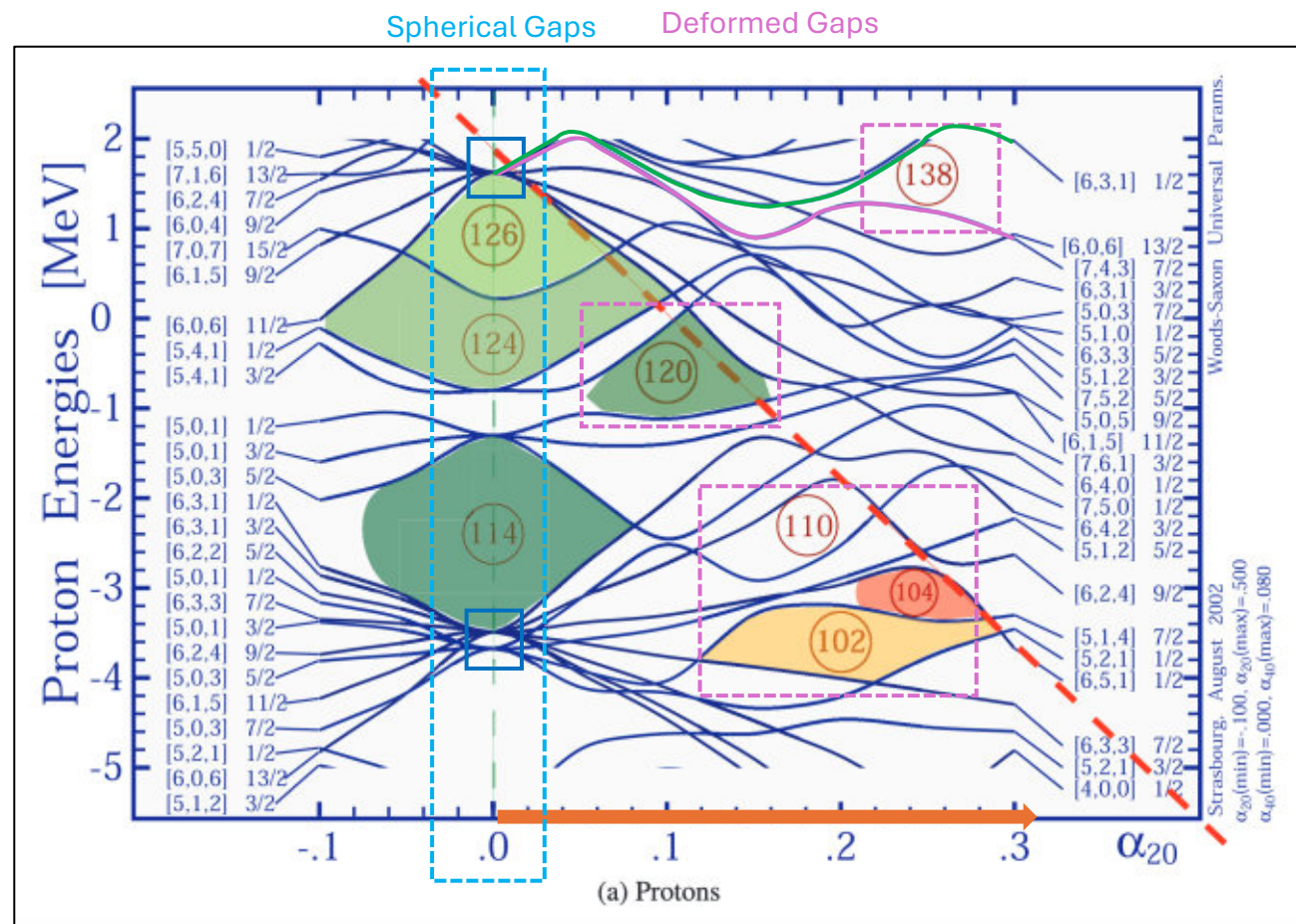
Shell model → spherical gaps

Diagram adapted from Plot Julien, Thesis (2010) [3]



Nuclear deformation → deformed gaps

Diagram adapted from J. Dudek, private communication.




By extrapolating the data measured for deformed gaps, we can find the next spherical gaps.

Why Study Superheavy Elements? Another Motivation

Reason 2: To refine our understanding of **atomic** structure by studying the **chemical properties** of new elements!

Z = 118 complete the 7th period of the periodic table of elements and fits into the noble gas column. However...



Communications

Angewandte
International Edition
Chemie

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International Edition: doi.org/10.1002/anie.202011976
German Edition: doi.org/10.1002/ange.202011976

Oganesson

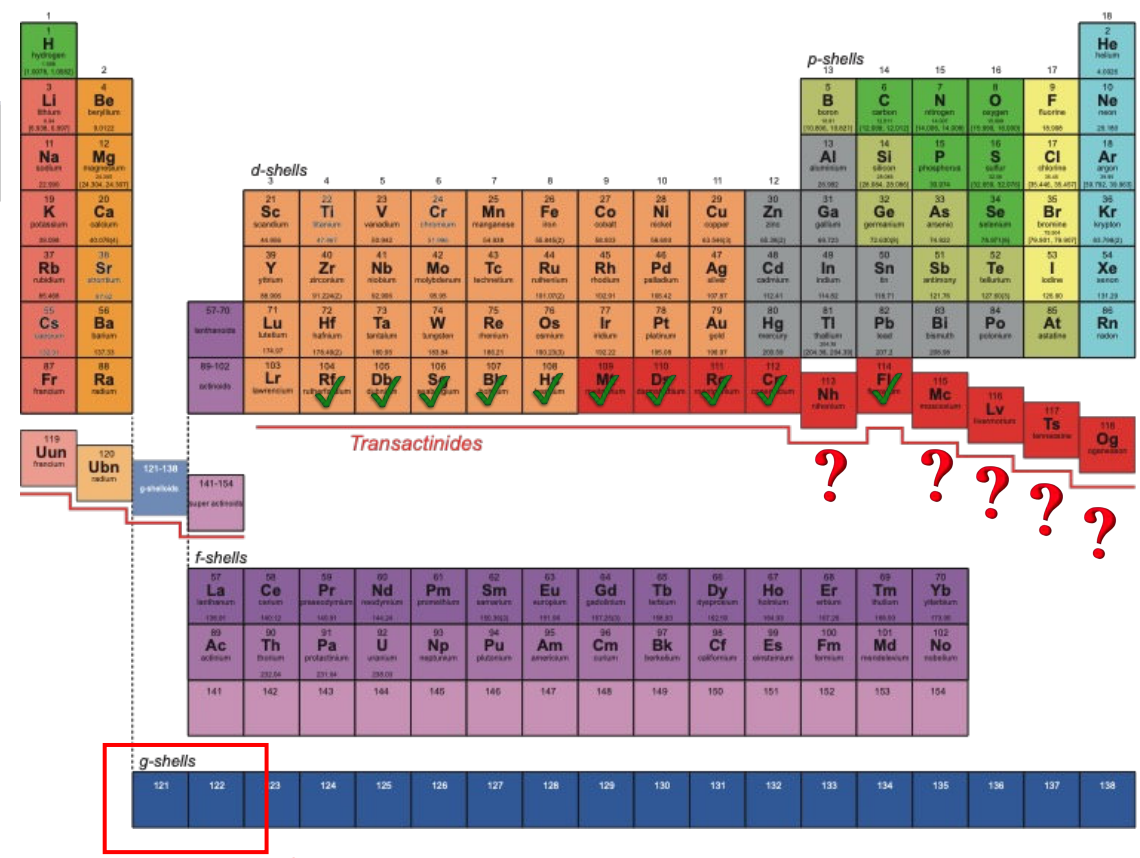
Oganesson: A Noble Gas Element That Is Neither Noble Nor a Gas

Odile R. Smits,* Jan-Michael Mewes,* Paul Jerabek,* and Peter Schwerdtfeger*

Abstract: Oganesson (Og) is the last entry into the Periodic Table completing the seventh period of elements and group 18 of the noble gases. Only five atoms of Og have been successfully produced in nuclear collision experiments, with an estimate half-life for ²⁹⁴Og of 0.69^{+0.64}_{-0.22} ms.^[1] With such a short lifetime, chemical and physical properties inevitably have to come from accurate relativistic quantum theory. Here, we employ two complementary computational approaches, namely parallel tempering Monte-Carlo (PTMC) simulations and first-principles thermodynamic integration (TI), both calibrated against a highly accurate coupled-cluster reference to pin-down the melting and boiling points of this super-heavy element. In excellent agreement, these approaches show Og to be a solid at ambient conditions with a melting point of ≈325 K. In contrast, calculations in the nonrelativistic limit reveal a melting point for Og of 220 K, suggesting a gaseous state as expected for a typical noble gas element. Accordingly, relativistic effects shift the solid-to-liquid phase transition by about 100 K.

due to strong relativistic effects.^[8–10] For example Cn and Fl are predicted to be chemically inert^[7,11,12] due to the relativistic 7s shell contraction for Cn and the large spin-orbit splitting of the 7p shell, resulting in a closed 7p_{1/2} shell for Fl.

In contrast to all other noble-gas solids, Og was recently predicted to be a semiconductor.^[13] Further, the electron localization function for the Og atom shows a uniform electron-gas-like behavior in the valence region, accompanied by a large dipole polarizability.^[8] These findings indicate that for the interaction between Og atoms, 3-body effects might become more important than for the lighter noble gases. Indeed, this was recently confirmed by calculations, which also revealed a stark increase in the many-body interaction due to relativistic effects.^[14] Based on such a many body expansions derived rigorously from relativistic coupled cluster theory, the melting temperature of the noble gases from Ne to Rn were obtained through parallel tempering Monte Carlo (PTMC), resulting in deviations of not more

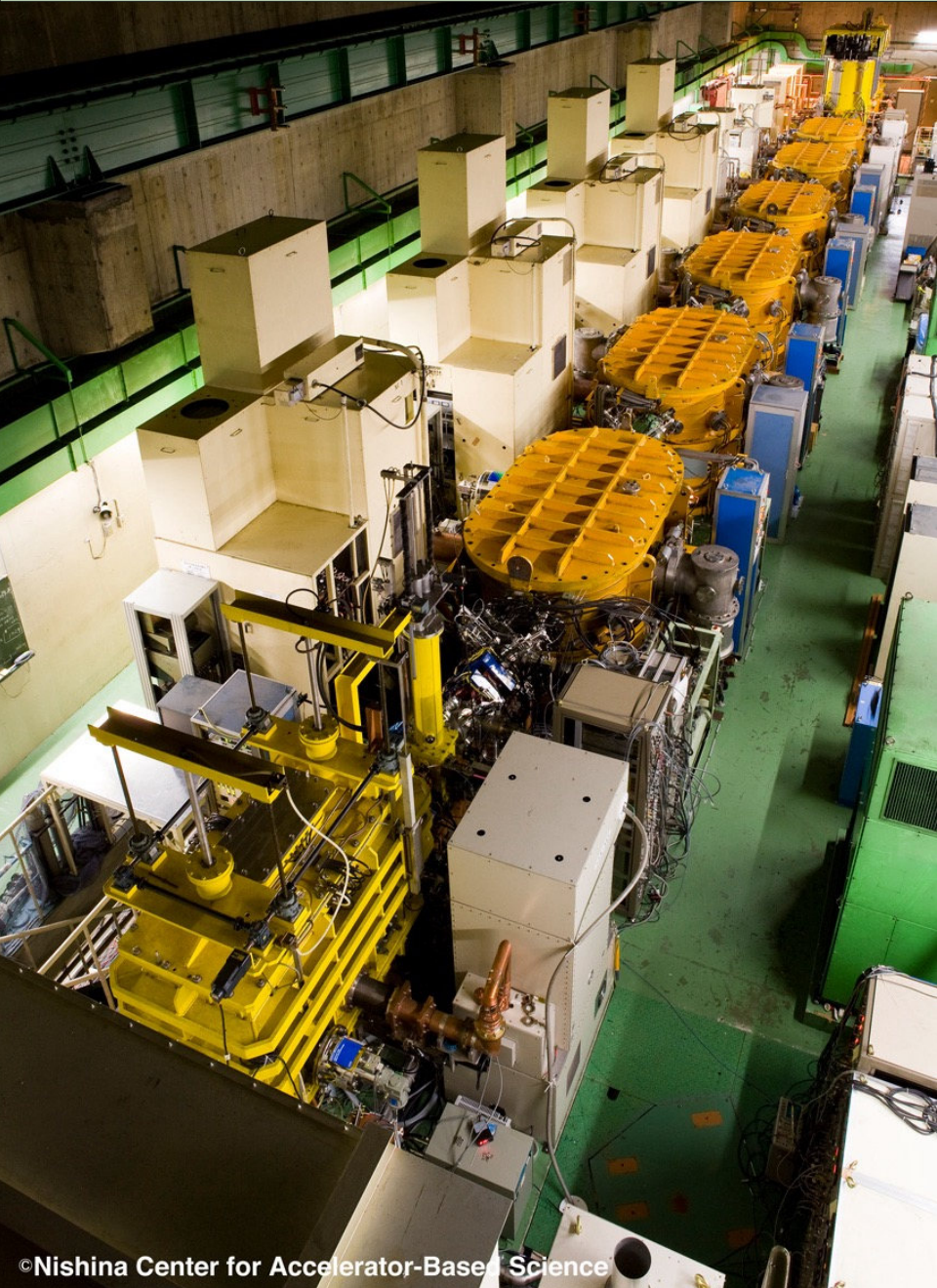


Article from Smits et al. (2020) [5]

Figure from Benoît Gall, private communication.

New electron shell

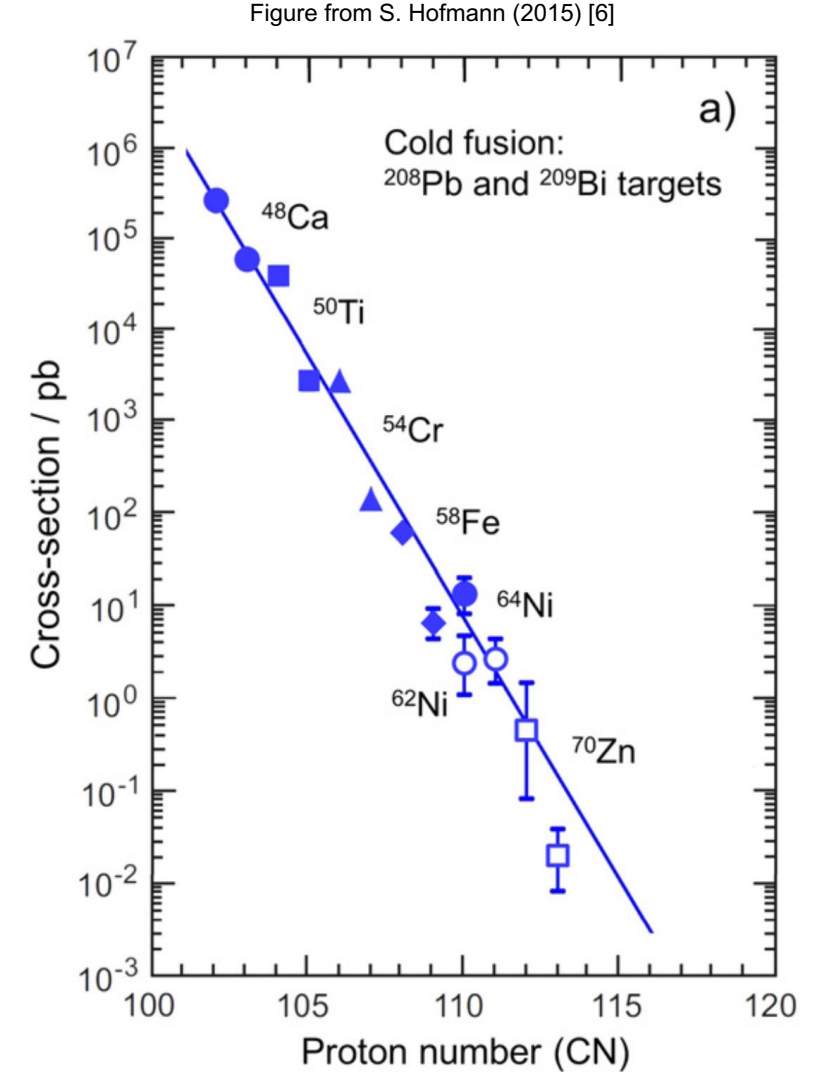
Presentation Outline



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Experimental Challenges for Synthesizing SHE

Challenge n°1 - Exceedingly small cross-sections



Experimental Challenges for Synthesizing SHE

Challenge n°1 - Exceedingly small cross-sections

Example: $Z = 113$ (Nihonium)

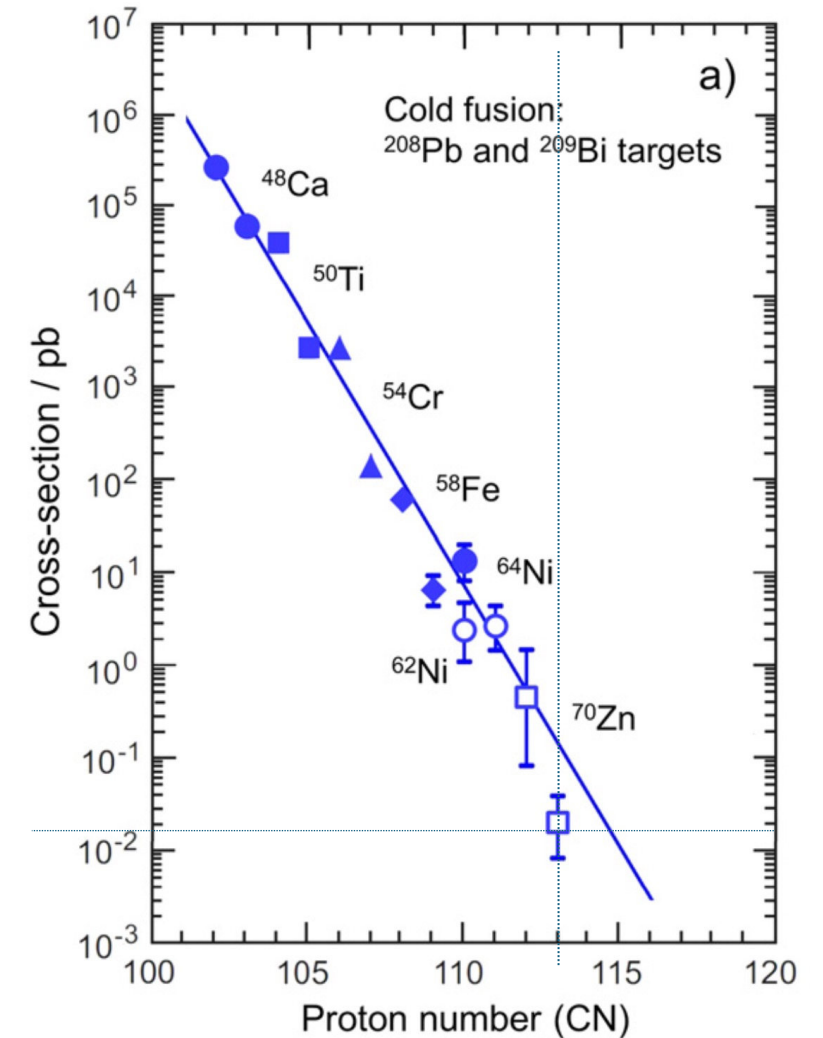
Cross-Section ~ 20 fb

Atoms produced: 3

Cumulated Beam Time ~ 3 years

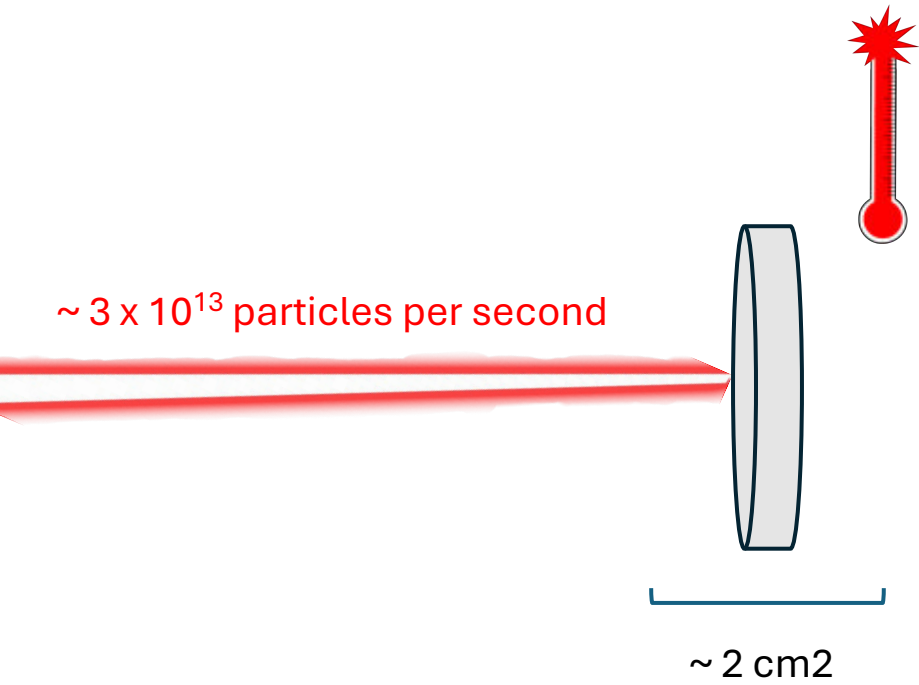
Cumulated Dose $\sim 10^{19}$ particles

Figure from S. Hofmann (2015) [6]



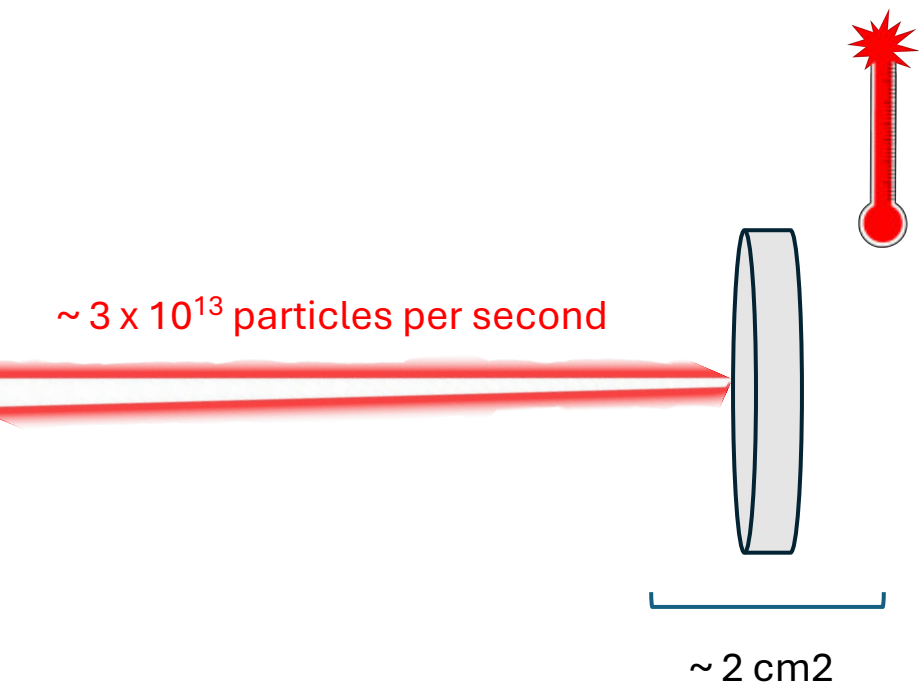
~ 20 fb

Challenge n°2 - Targets



Experimental Challenges for Synthesizing SHE

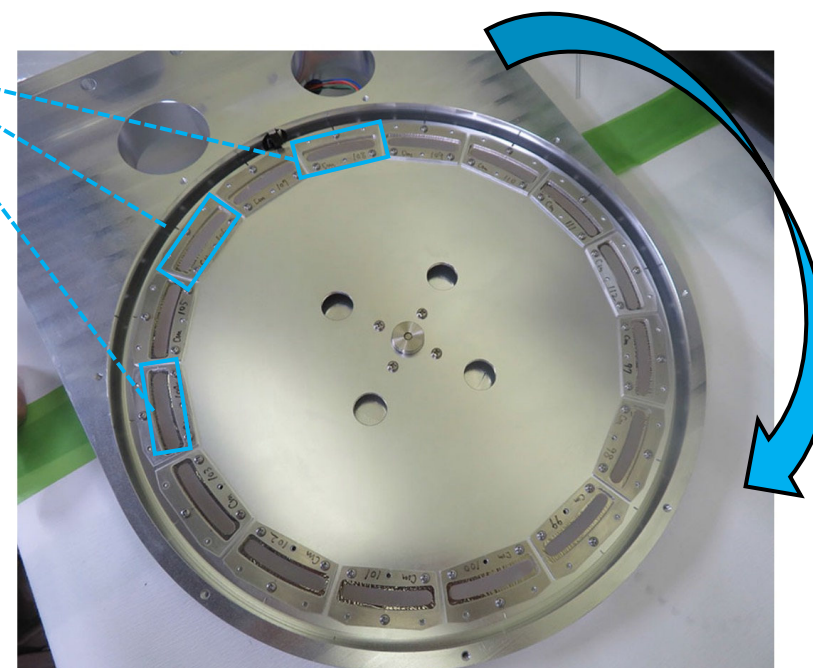
Challenge n°2 - Targets



Melting from extreme temperatures!

Solution: rotating wheel inside a gas to help with heat dissipation

Sector targets



~ 2000 rpm
(rotations per minute)

Fig. 17 Photograph of the rotating wheel with sixteen ^{248}Cm sector targets in the semiclosed inner-target box. The cover plate of the inner-target box is removed to display the interior

Photograph from Sakai et al. (2022) [7]

RIKEN Experimental Setup for Synthesizing SHE

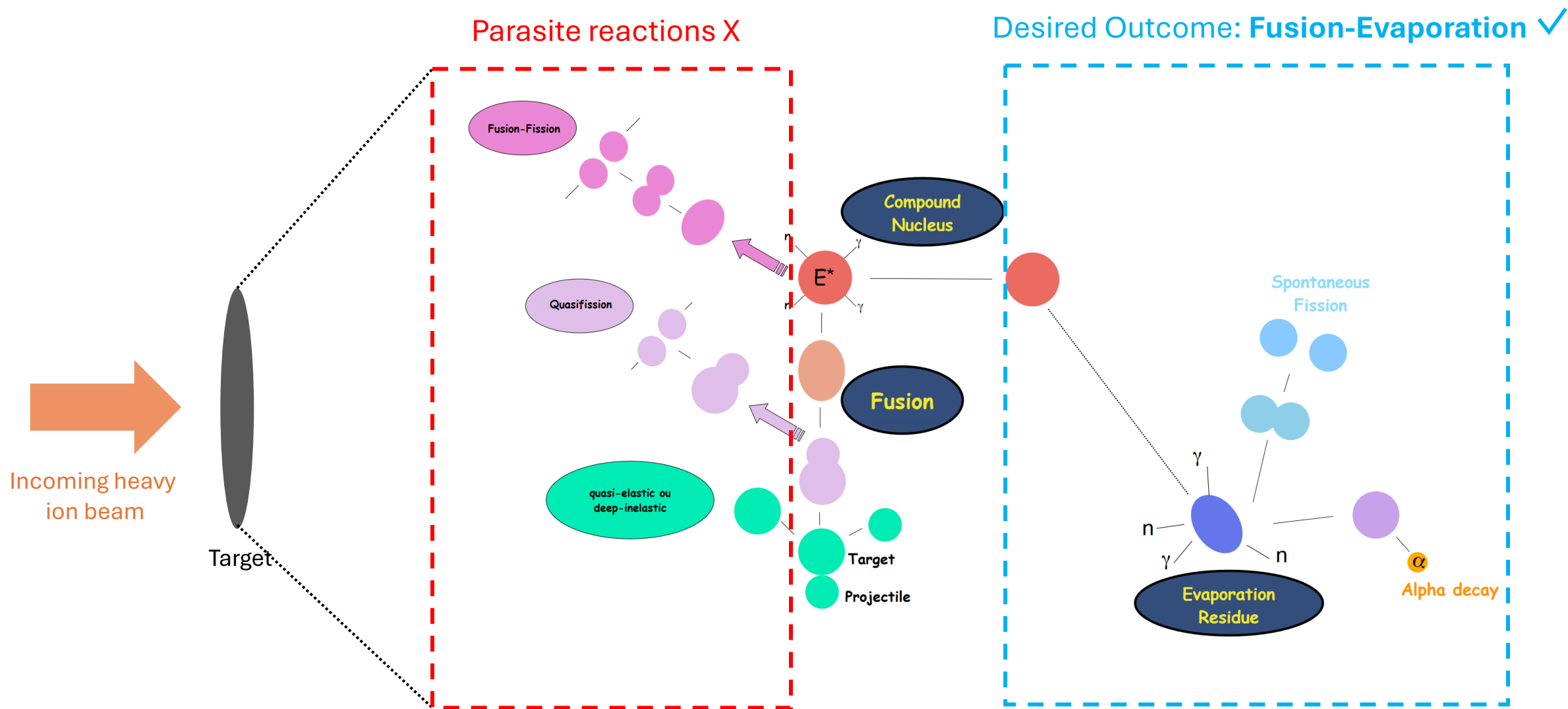


Figure from Olivier Dorvaux, private communication.

RIKEN Experimental Setup for Synthesizing SHE

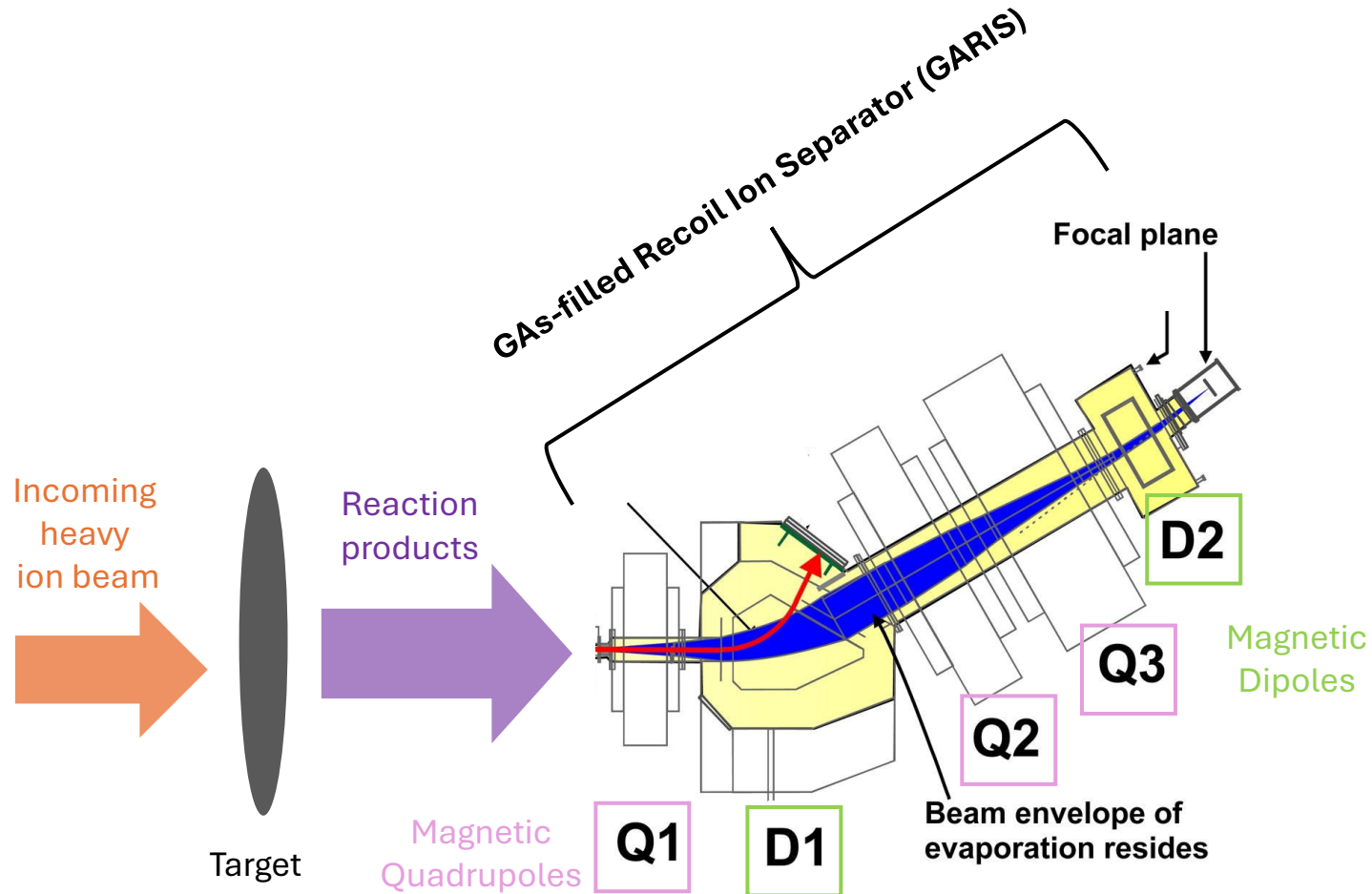
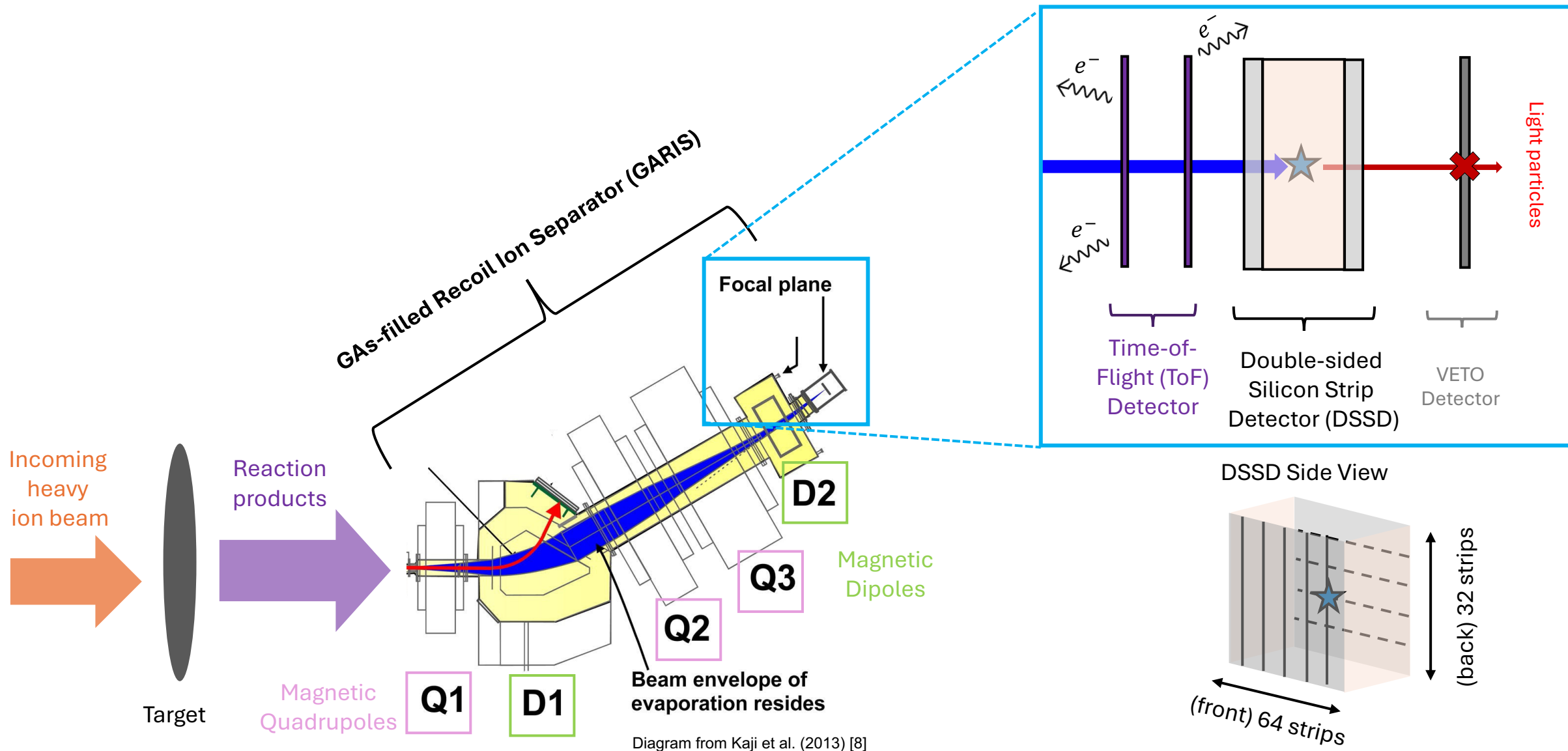


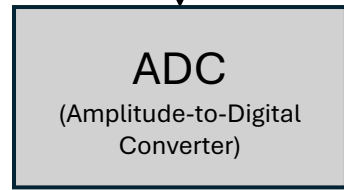
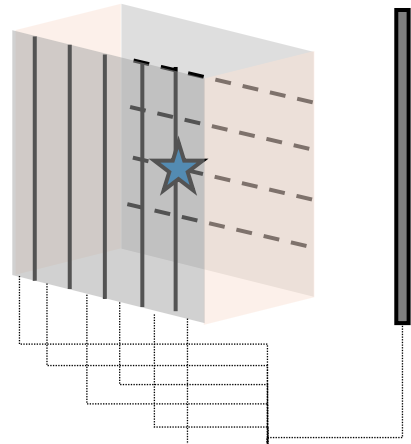
Diagram from Kaji et al. (2013) [8]

RIKEN Experimental Setup for Synthesizing SHE



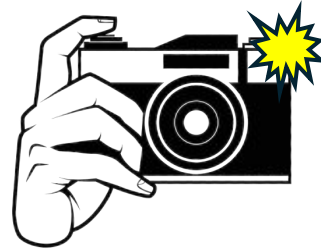
RIKEN - Data Acquisition Chain

DSSD VETO Detector



95 Signal Amplitudes
(62 + 32 Strips & 1 Veto)

DSSD Implantation = Trigger

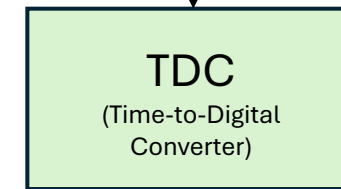
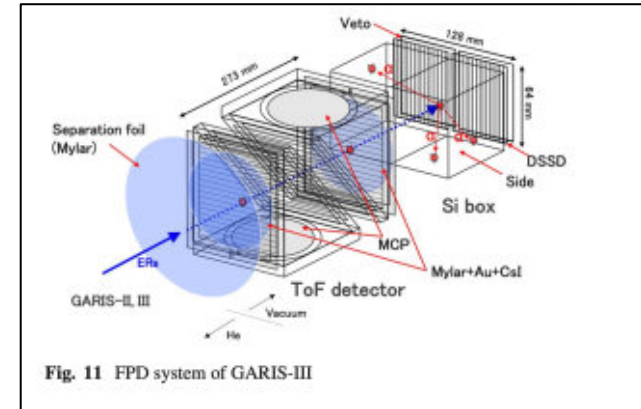


Clock

Timestamp

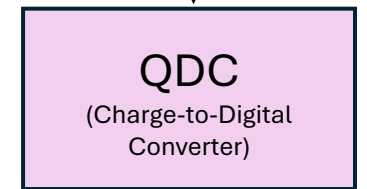
Available Data for 1 Event

Time-of-Flight Detector



TDC1

TDC2



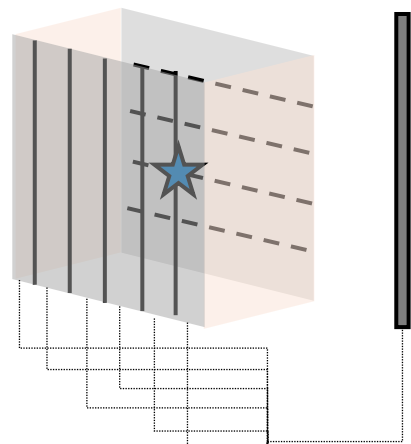
QDC1

QDC2

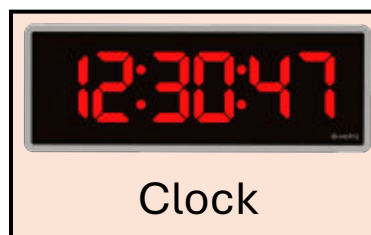
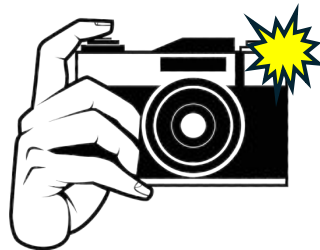
Time-of-Flight

RIKEN - Data Acquisition Chain

DSSD VETO Detector



DSSD Implantation = Trigger



Clock

Timestamp

ADC
(Amplitude-to-Digital Converter)

95 Signal Amplitudes
(62 + 32 Strips & 1 Veto)

1 strip = 1 calibration

Available Data for 1 Event

Time-of-Flight Detector

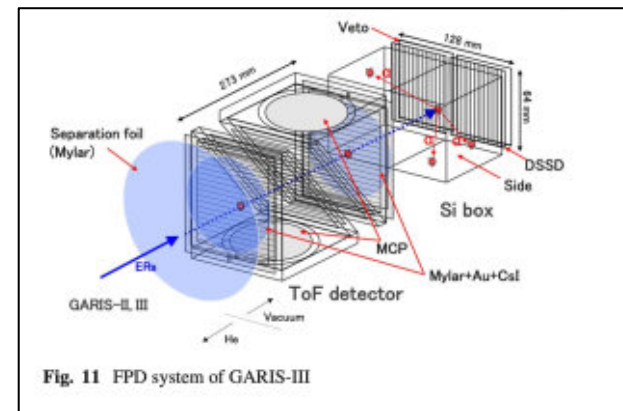


Diagram from Sakai et al. (2022) [7]

TDC
(Time-to-Digital Converter)

TDC1

TDC2

Time-of-Flight

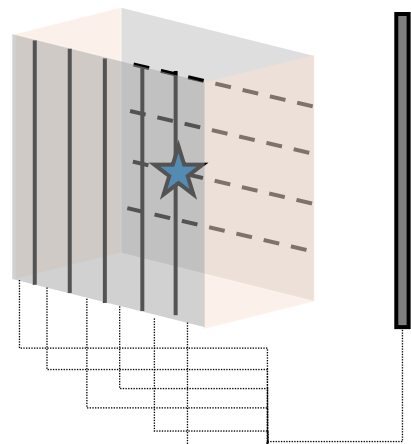
QDC
(Charge-to-Digital Converter)

QDC1

QDC2

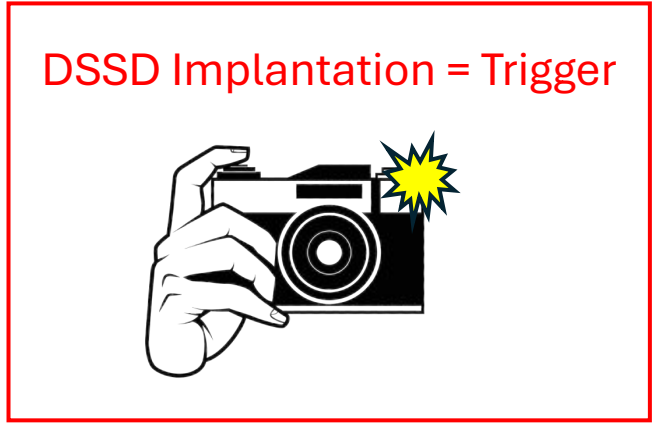
RIKEN - Data Acquisition Chain

DSSD VETO Detector



95 Signal Amplitudes
(62 + 32 Strips & 1 Veto)

(1 strip = 1 calibration) x 2 data sets



Timestamp

Available Data for 1 Event

Time-of-Flight Detector

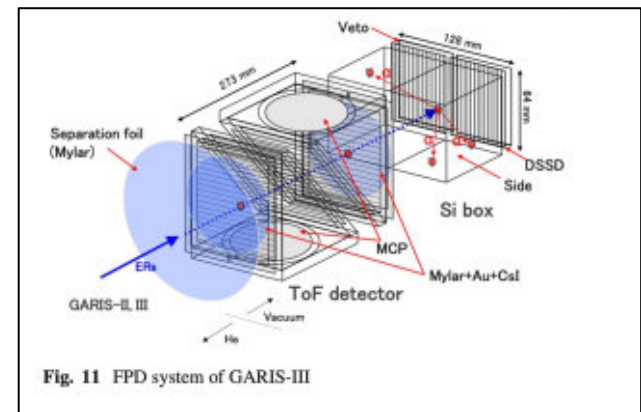
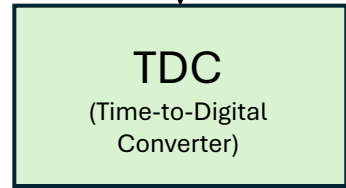
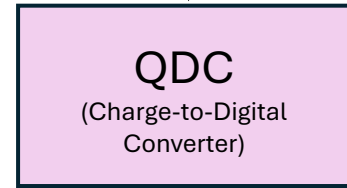


Diagram from Sakai et al. (2022) [7]



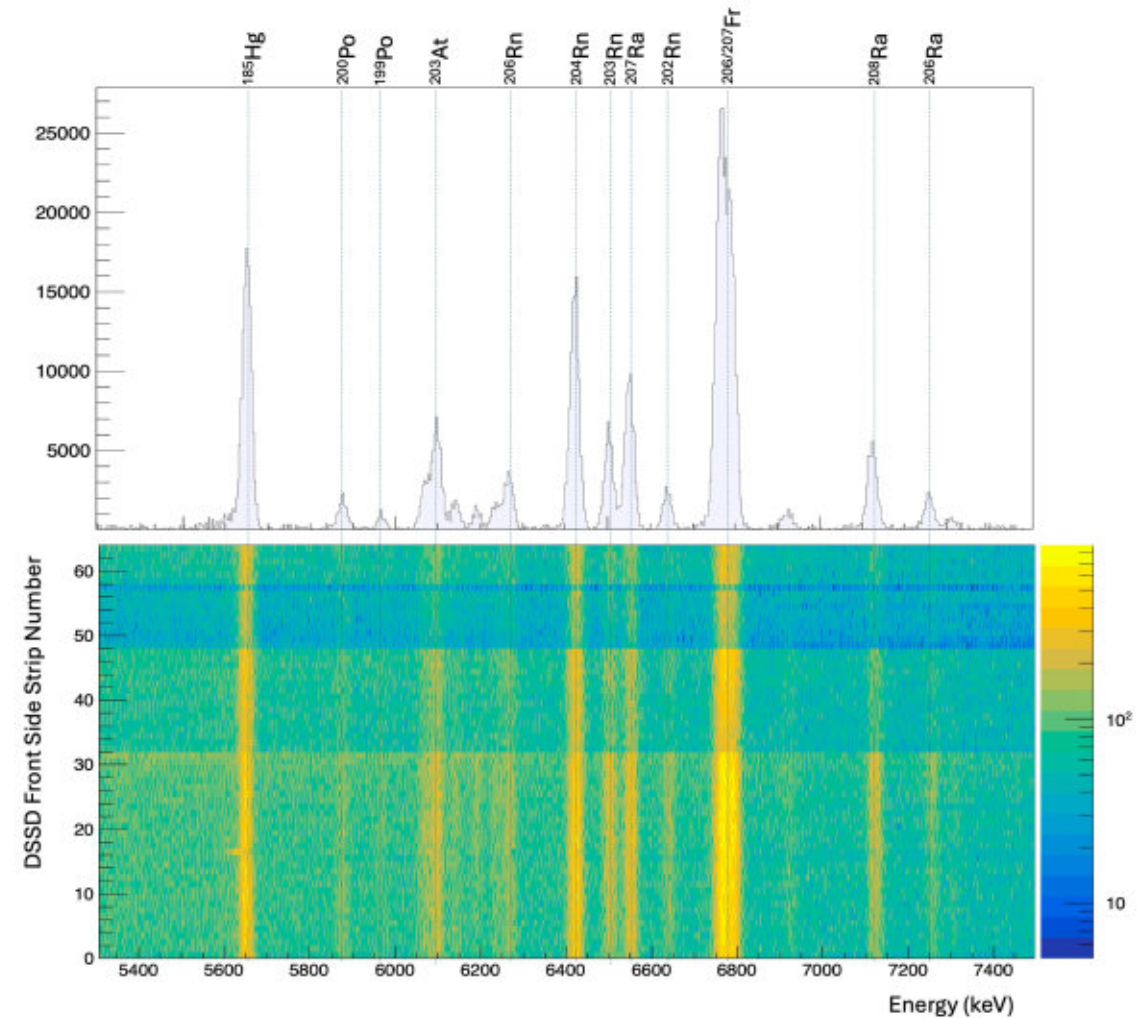
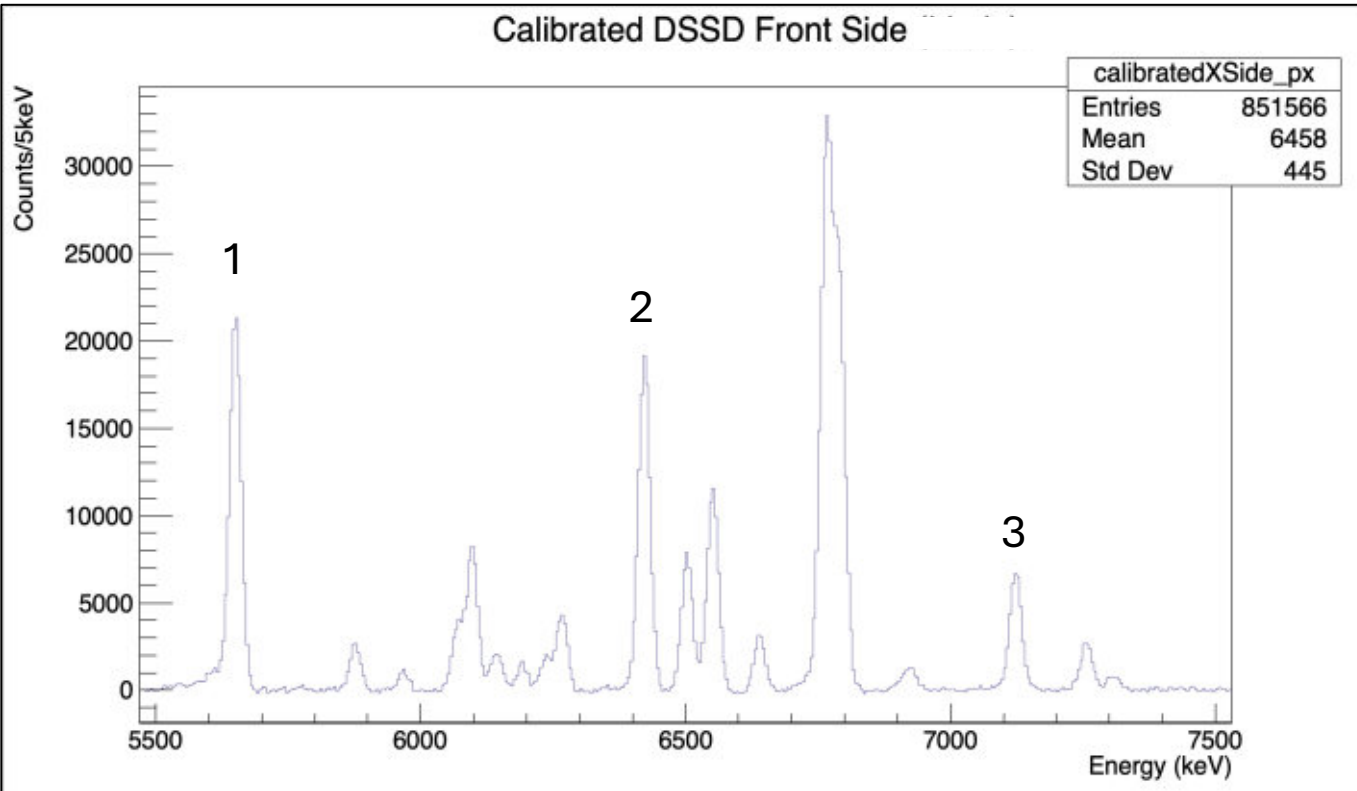
TDC1 TDC2



QDC1 QDC2

Time-of-Flight

Calibration Results



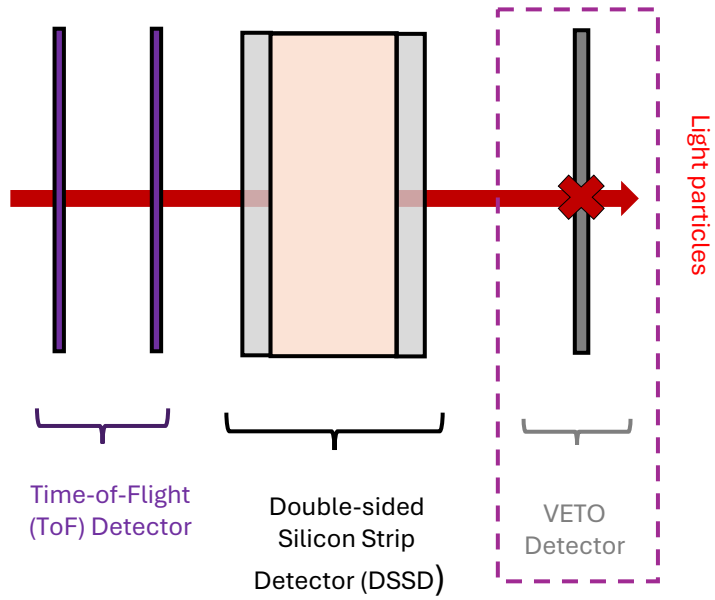
Peak Number	Peak Center (keV)	Uncertainty	Resolution (keV)	Uncertainty
1	5650,17	0,03	26,12	0,06
2	6421,94	0,03	25,93	0,06
3	7122,95	0,06	27,92	0,13

Presentation Outline

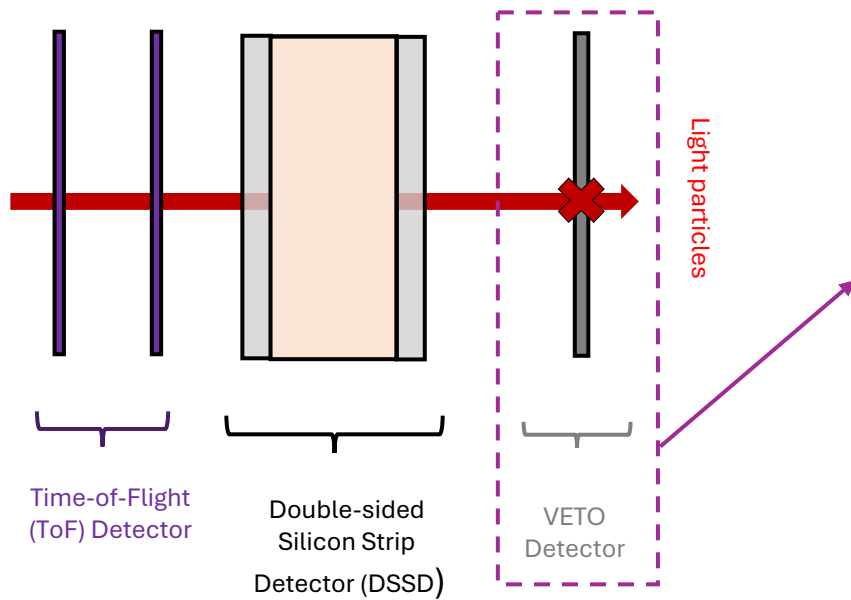


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Data Selection - Veto Detector

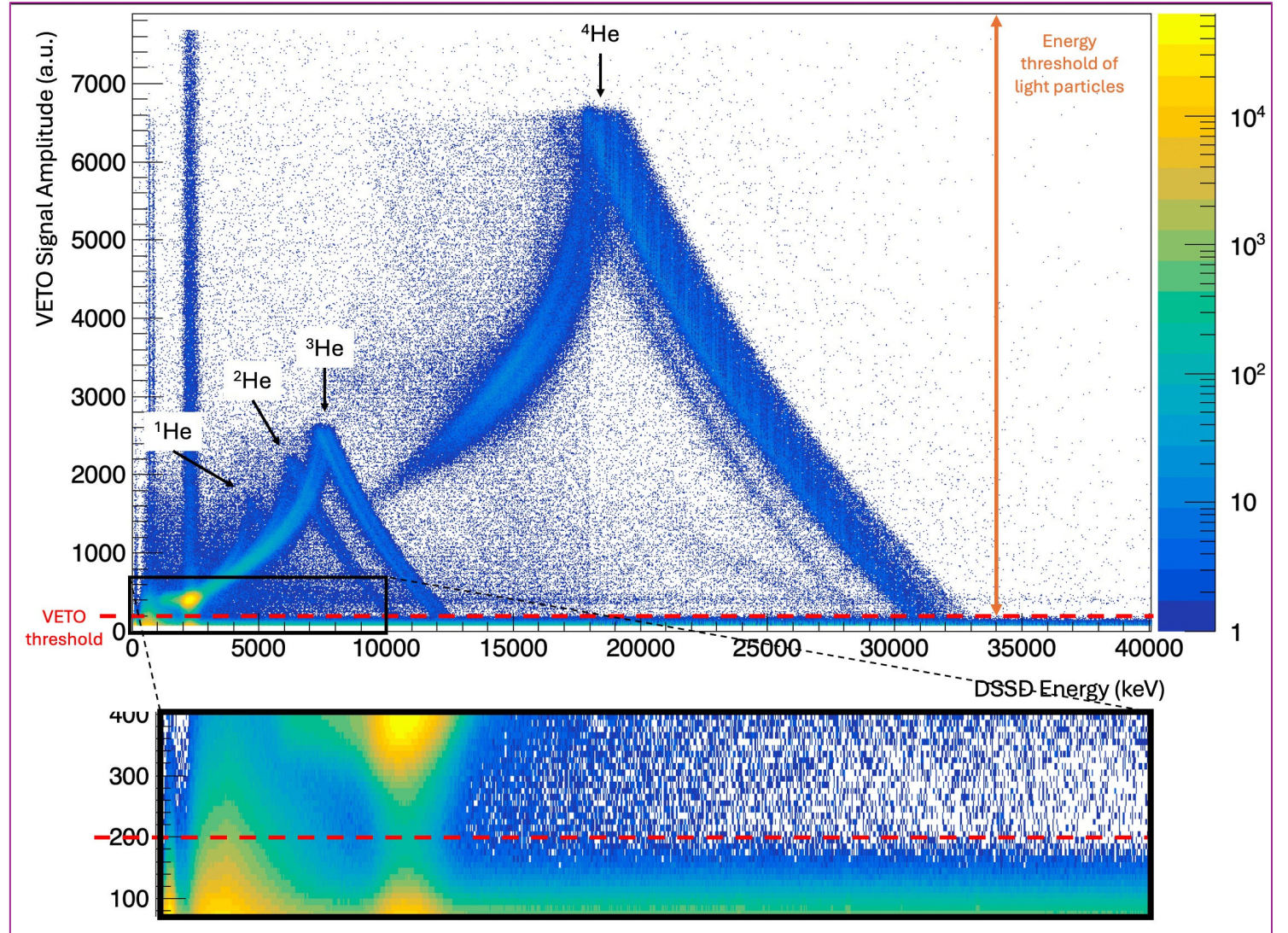


Data Selection - Veto Detector



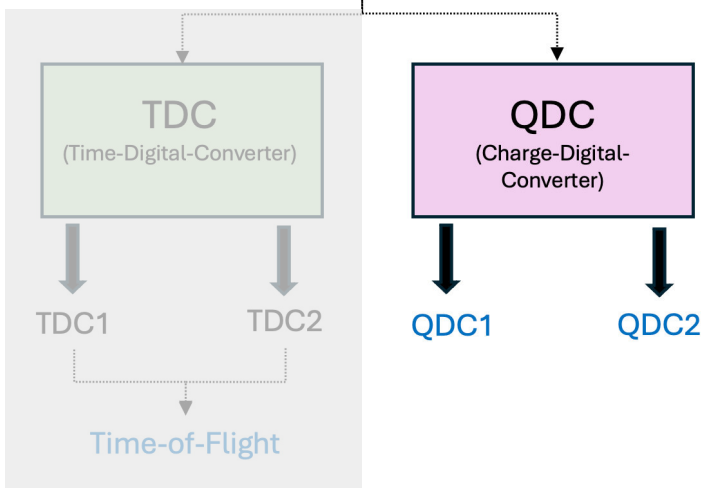
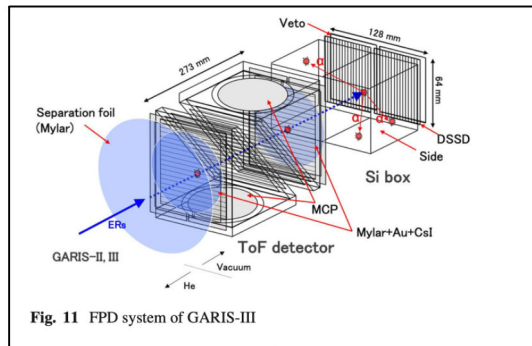
The VETO detector gives us insight about two interesting phenomena:

1. An electronic noise band \sim (0 to 200 a.u.)
2. The Bragg peaks of the light particles traversing the DSSD



Data Selection - QDC (Charge-to-Digital Converter)

Time-of-Flight Detector



Data Selection - QDC (Charge-to-Digital Converter)

Time-of-Flight Detector

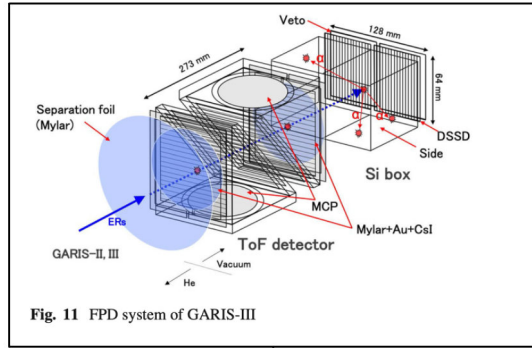
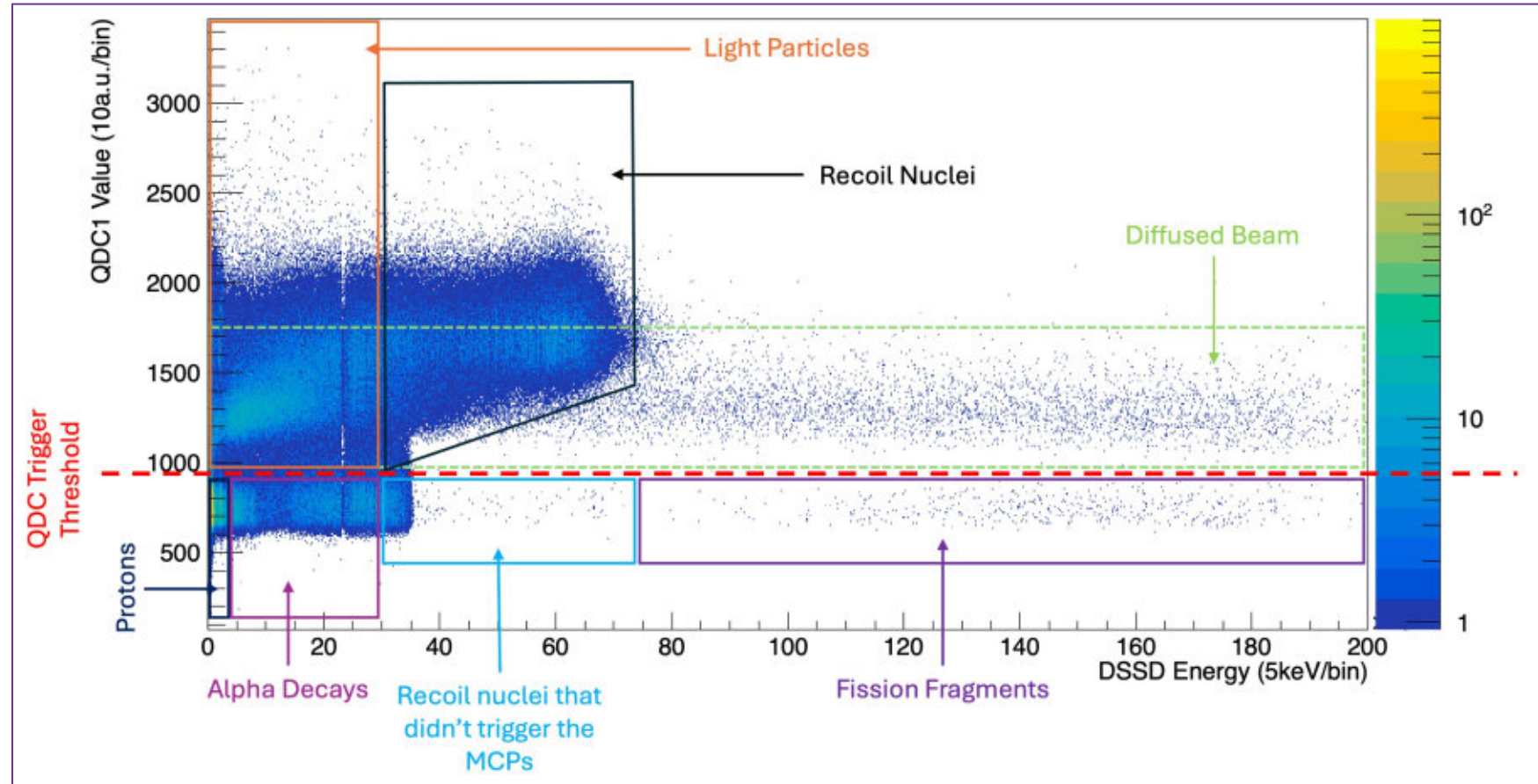
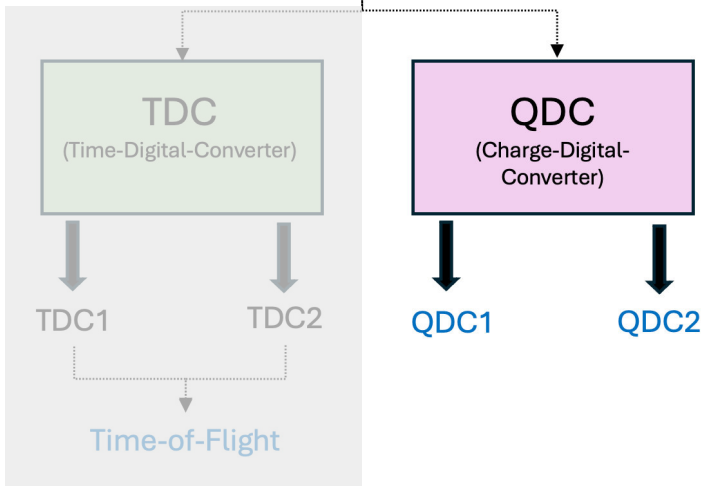


Diagram from Sakai et al. (2022) [7]



The QDC gives us insights about **many different physical phenomena** that occur inside the DSSD.

Data Selection - ToF (Time-to-Flight)

Time-of-Flight Detector

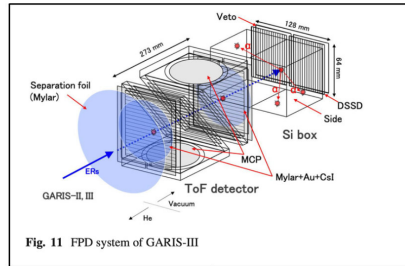
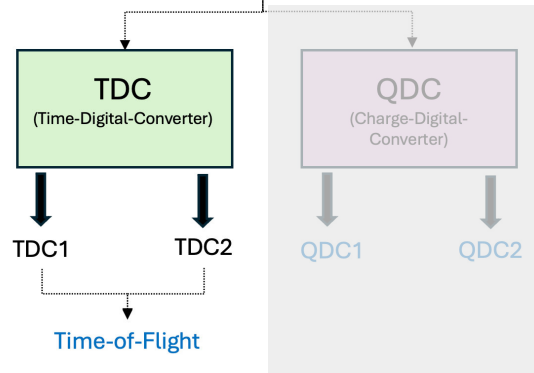


Diagram from Sakai et al. (2022) [7]



Data Selection - ToF (Time-to-Flight)

Time-of-Flight Detector

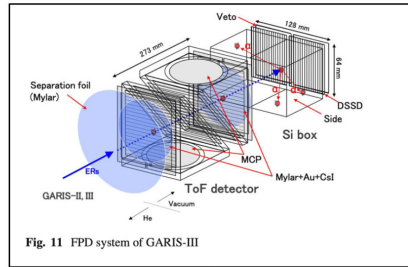
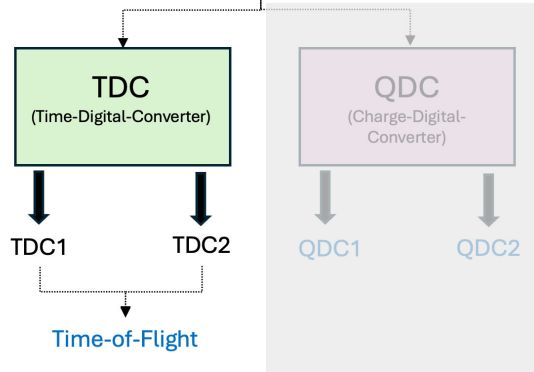


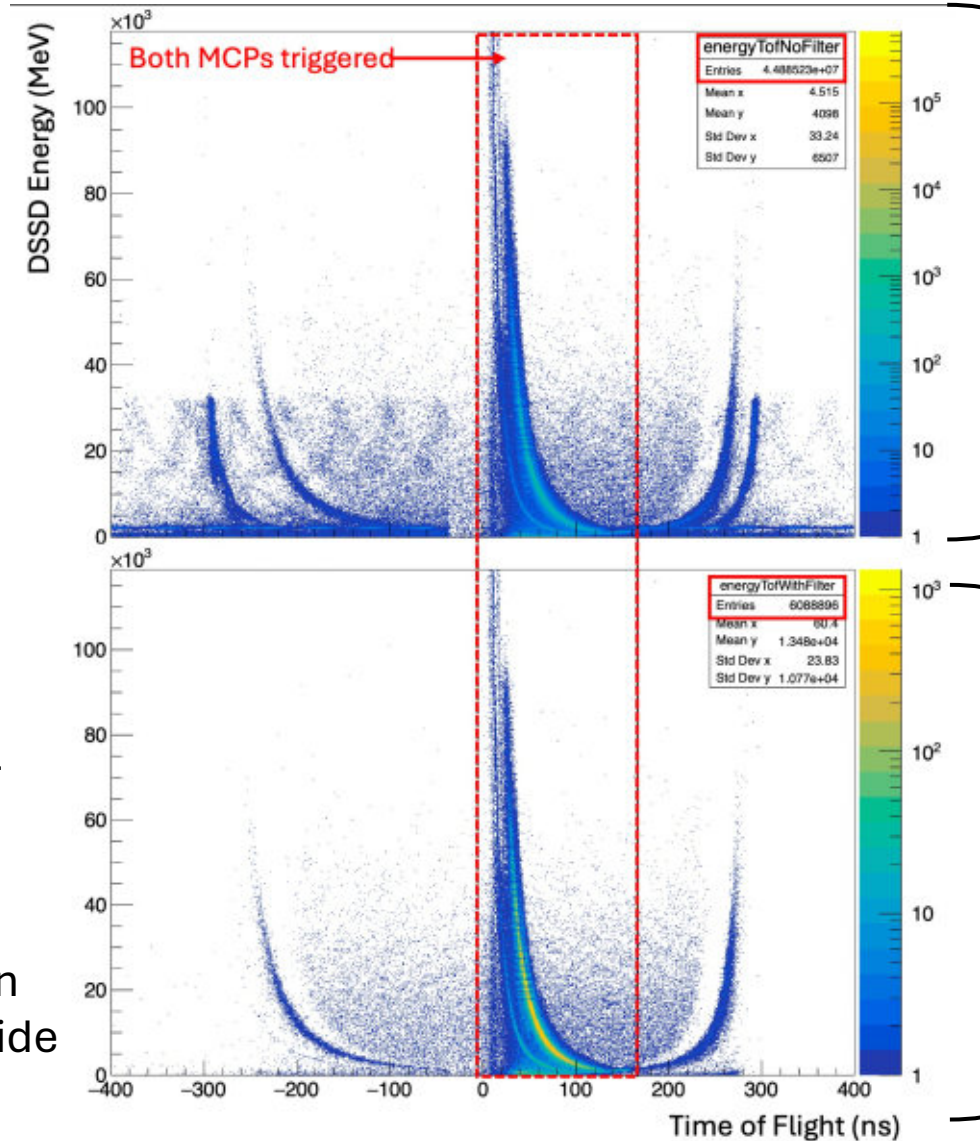
Diagram from Sakai et al. (2022) [7]



Notice the difference before and after applying Veto and QDC selection!



~86% of the original events have been rejected with only an 8% difference inside the dashed lines.



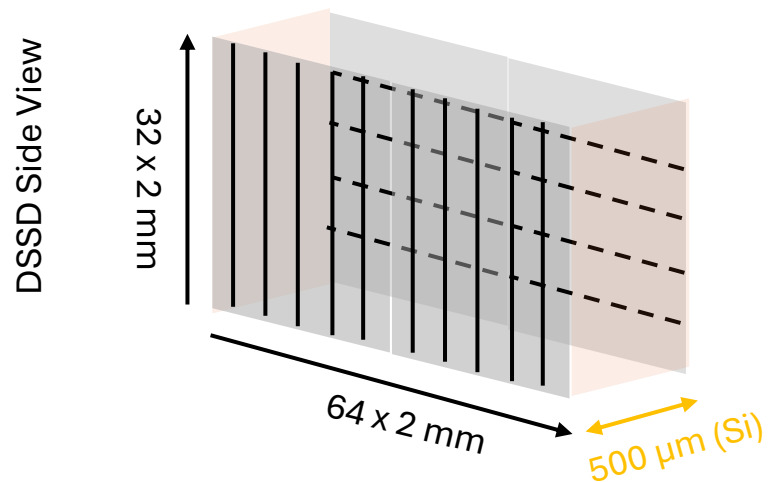
All events (no selection):

- 44,885,230 entries total
- 6,534,106 entries inside []

Events after Veto & QDC cuts:

- 6,088,896 entries total
- 5,990,440 entries inside []

How do I identify an implanted nucleus?



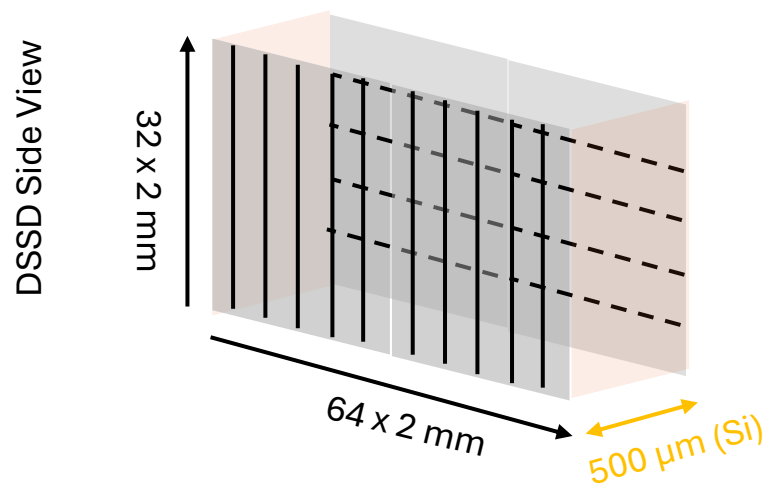
Event Type	VETO	QDC	Energy-vs-ToF
Recoil Events	Untriggered	Above threshold	Inside red dashed lines
Decay Events	Untriggered	Underneath threshold	Not applicable
Parasite Events	Electronic noise band Bragg peaks	Light particles Diffused beam	Outside red dashed lines

Table 4.1: Summary of event identification criteria using VETO, QDC, and energy-vs-ToF filters.

How do I identify an implanted nucleus?

Method:

1) Select an event fulfilling recoil criteria



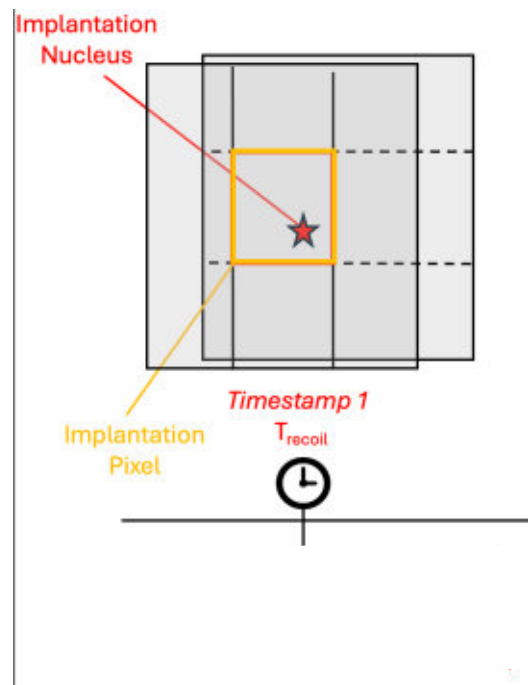
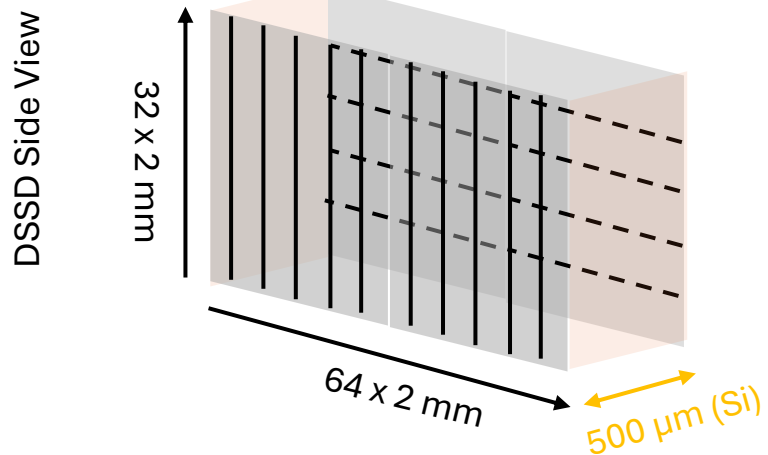
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Table 4.1: Summary of event identification criteria using VETO, QDC, and energy-vs-ToF filters.

How do I identify an implanted nucleus?

Method:

- 1) Select an event fulfilling recoil criteria
- 2) Locate Pixel



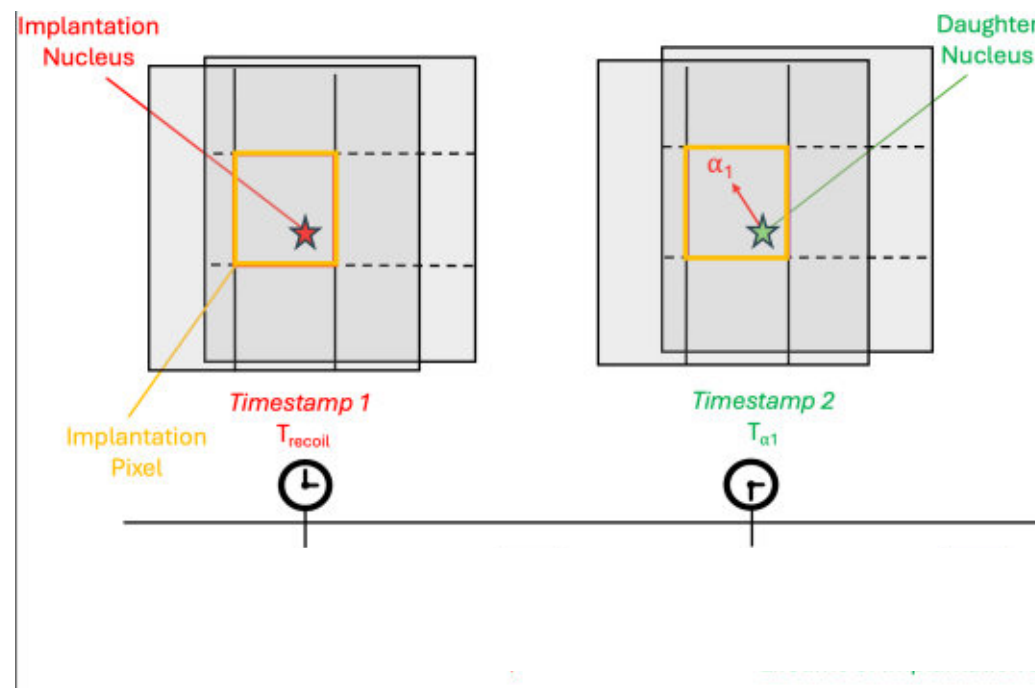
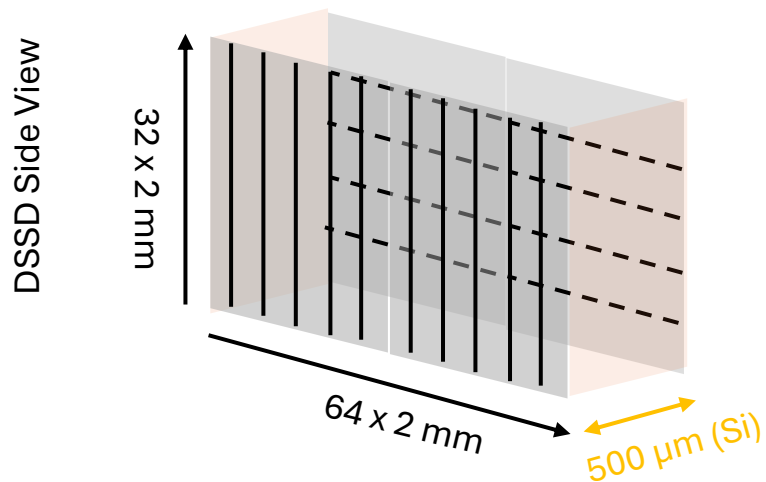
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Recoil Events	Untriggered	Above threshold	Inside red dashed lines
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Table 4.1: Summary of event identification criteria using VETO, QDC, and energy-vs-ToF filters.

How do I identify an implanted nucleus?

Method:

- 1) Select an event fulfilling recoil criteria
- 2) Locate Pixel
- 3) Wait for an alpha decay inside the same pixel



Event Type	VETO	QDC	Energy-vs-ToF
Recoil Events	Untriggered	Above threshold	Inside red dashed lines
Decay Events	Untriggered	Underneath threshold	Not applicable
Parasite Events	Electronic noise band Bragg peaks	Light particles Diffused beam	Outside red dashed lines

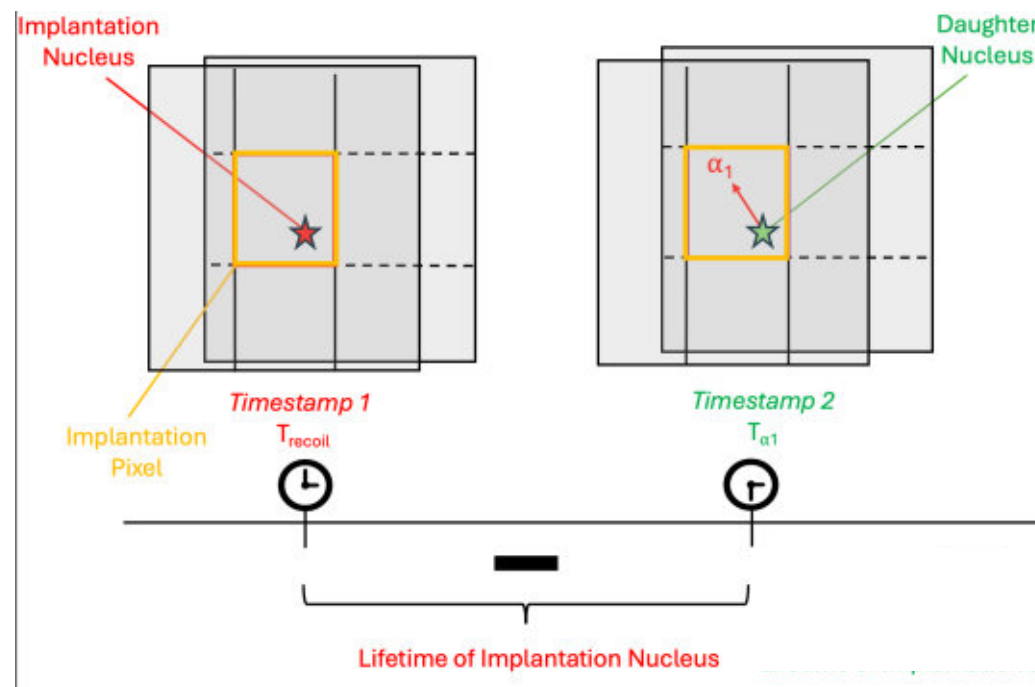
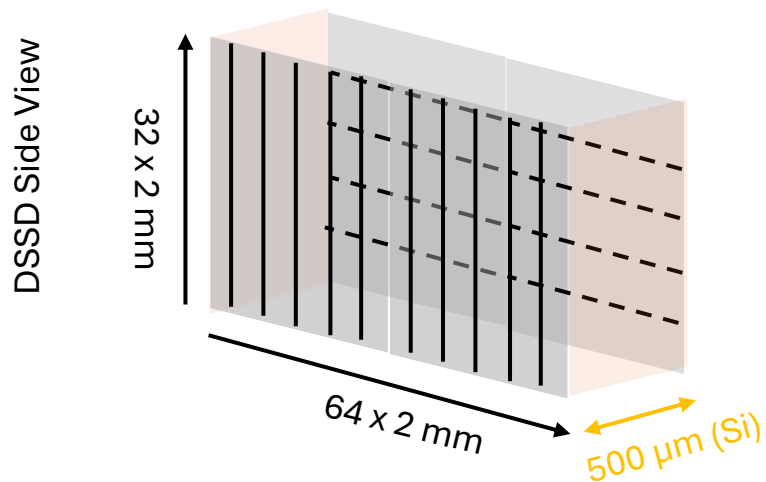
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How do I identify an implanted nucleus?

Method:

- 1) Select an event fulfilling recoil criteria
- 2) Locate Pixel
- 3) Wait for an alpha decay inside the same pixel
- 4) Subtract decay event and recoil event timestamps

$$T_{\alpha_1} - T_{recoil}$$



Event Type	VETO	QDC	Energy-vs-ToF
Recoil Events	Untriggered	Above threshold	Inside red dashed lines
Decay Events	Untriggered	Underneath threshold	Not applicable
Parasite Events	Electronic noise band Bragg peaks	Light particles Diffused beam	Outside red dashed lines

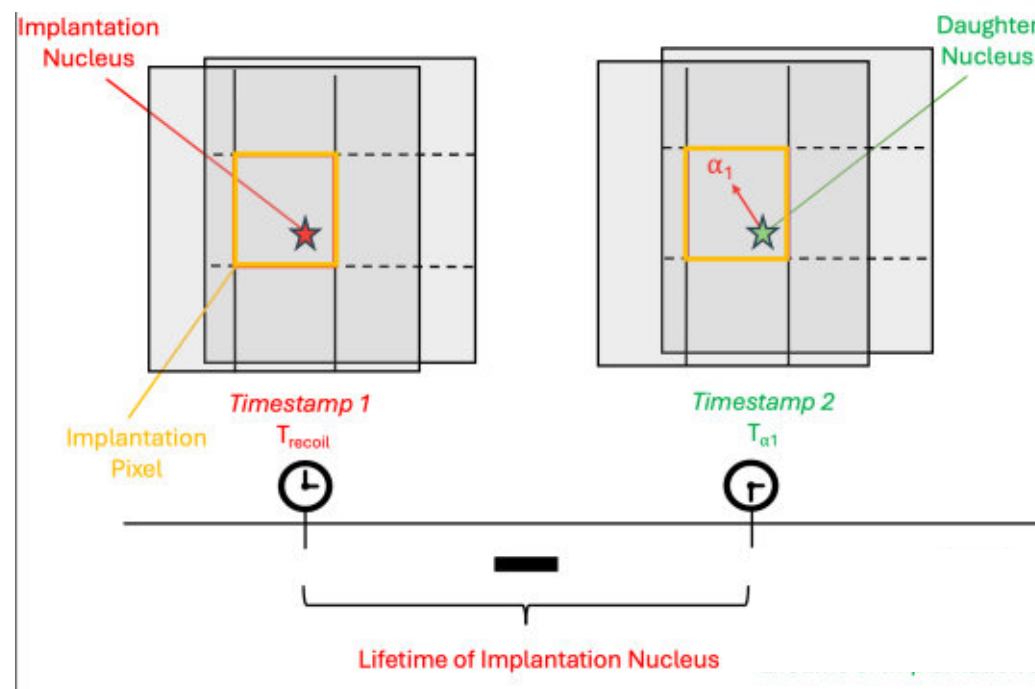
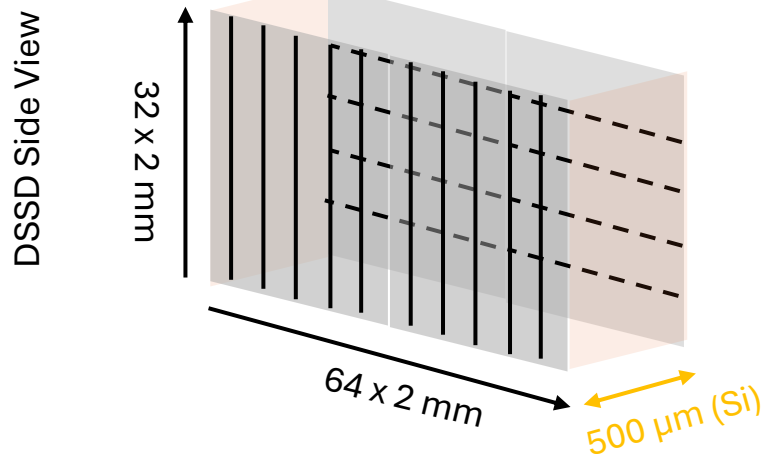
Table 4.1: Summary of event identification criteria using VETO, QDC, and energy-vs-ToF filters.

How do I identify an implanted nucleus?

Method:

- 1) Select an event fulfilling recoil criteria
- 2) Locate Pixel
- 3) Wait for an alpha decay inside the same pixel
- 4) Subtract decay event and recoil event timestamps

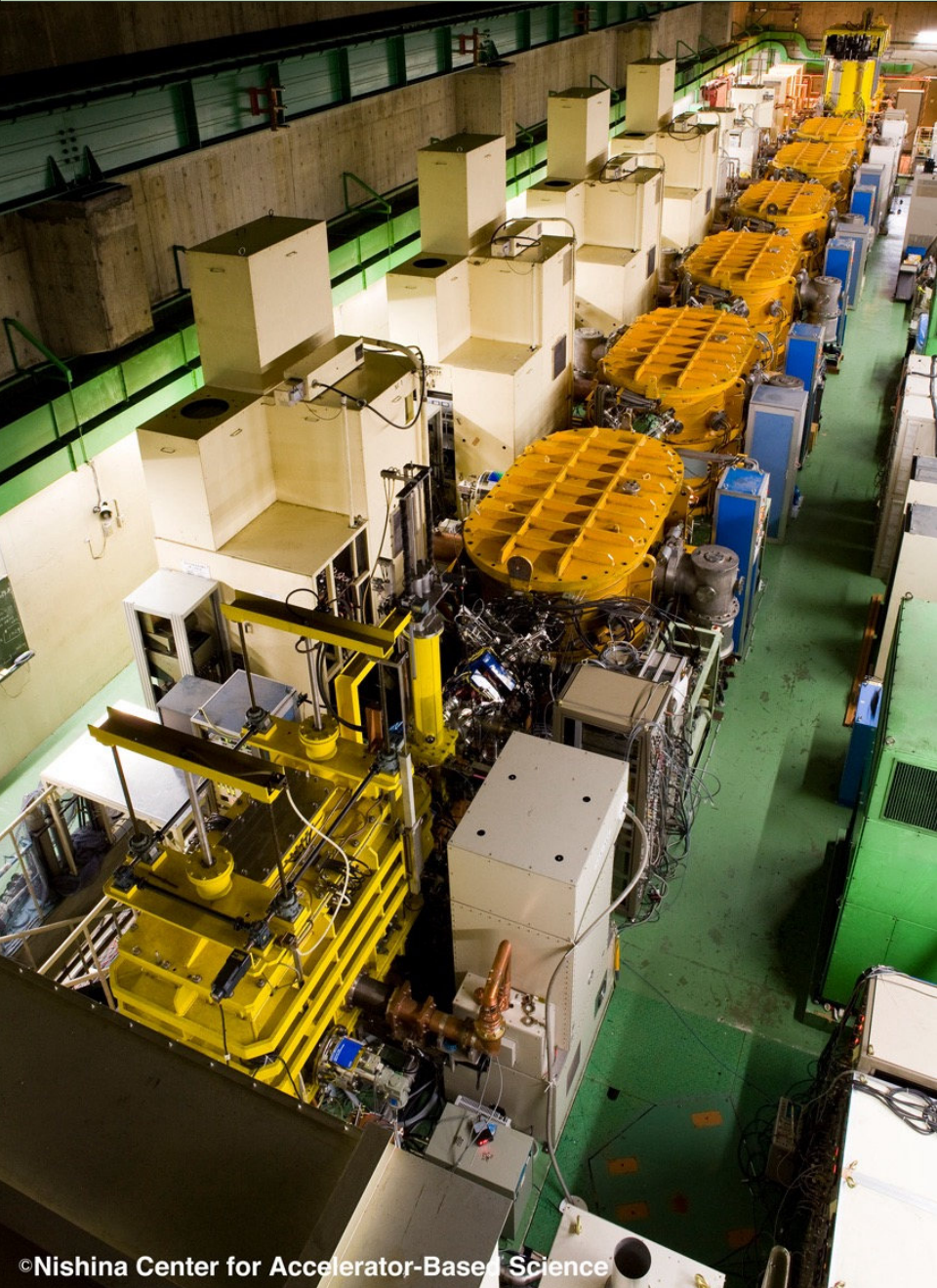
$$T_{\alpha 1} - T_{recoil}$$
- 5) Plot $\text{Log}_2(T_{\alpha 1} - T_{recoil})$ vs E_{α} to identify nucleus!



Event Type	VETO	QDC	Energy-vs-ToF
Recoil Events	Untriggered	Above threshold	Inside red dashed lines
Decay Events	Untriggered	Underneath threshold	Not applicable
Parasite Events	Electronic noise band Bragg peaks	Light particles Diffused beam	Outside red dashed lines

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Plotting an Excitation Function

The reaction I studied was ${}_{18}^{40}\text{Ar} + {}_{82}^{208}\text{Pb} \rightarrow {}_{100}^{248}\text{Fm}^* \rightarrow {}_{100}^{248-x}\text{Fm} + xn$ for six different beam energies.

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Channel	Isotope	E_{α_1}	E_{α_2}	Fission Fragments	Half-Life	$\text{Log}_2(\text{Half-Life})$
4n	${}^{244}\text{Fm}$	X	X	Yes	3.12 ms	18.25
3n	${}^{245}\text{Fm}$	8.150 MeV	X	No	4.2 s	28.65
2n	${}^{246}\text{Fm}$	8.198 MeV	8.242 MeV	Yes	1.54 s	27.20

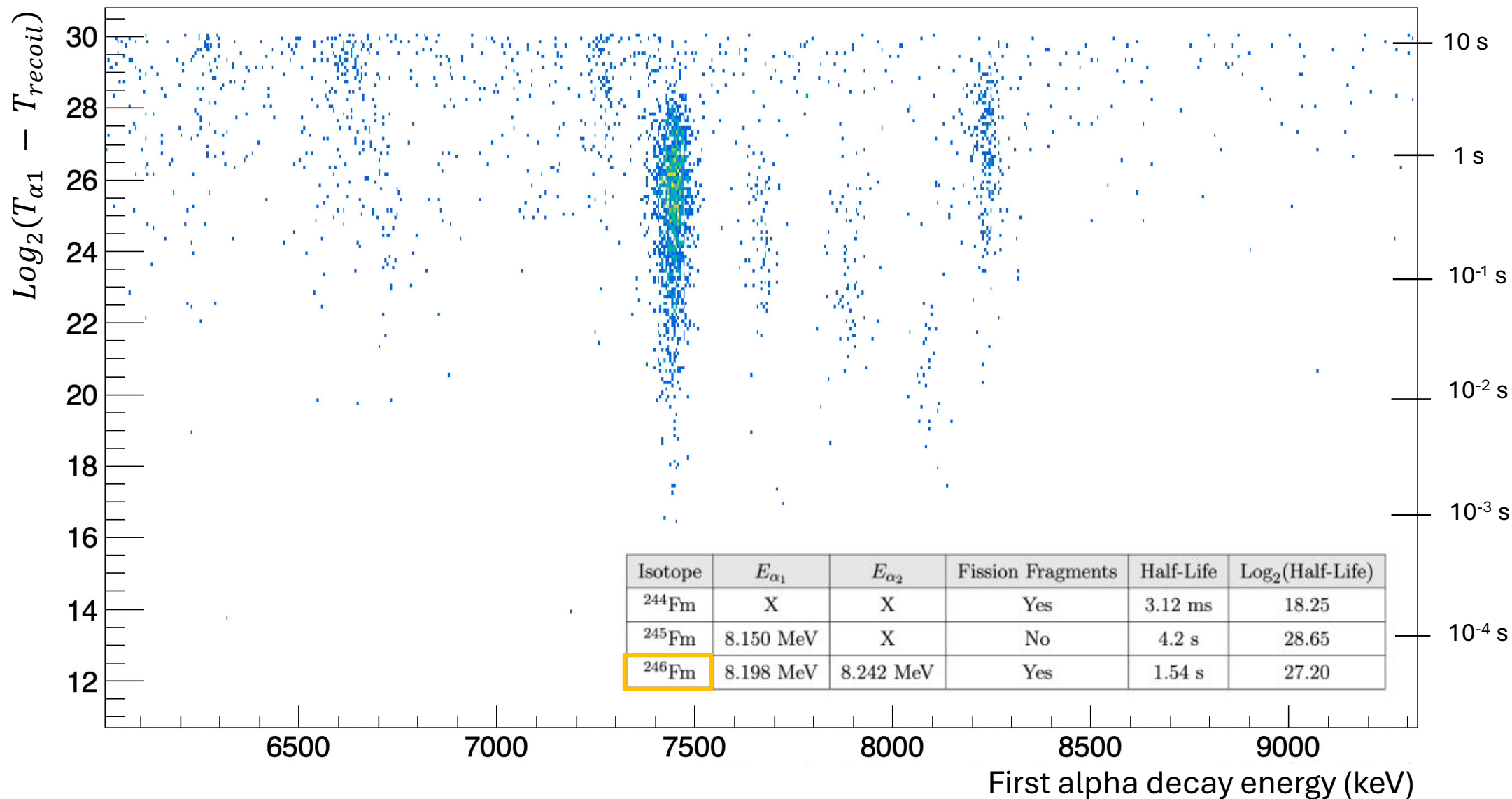


What I am looking for

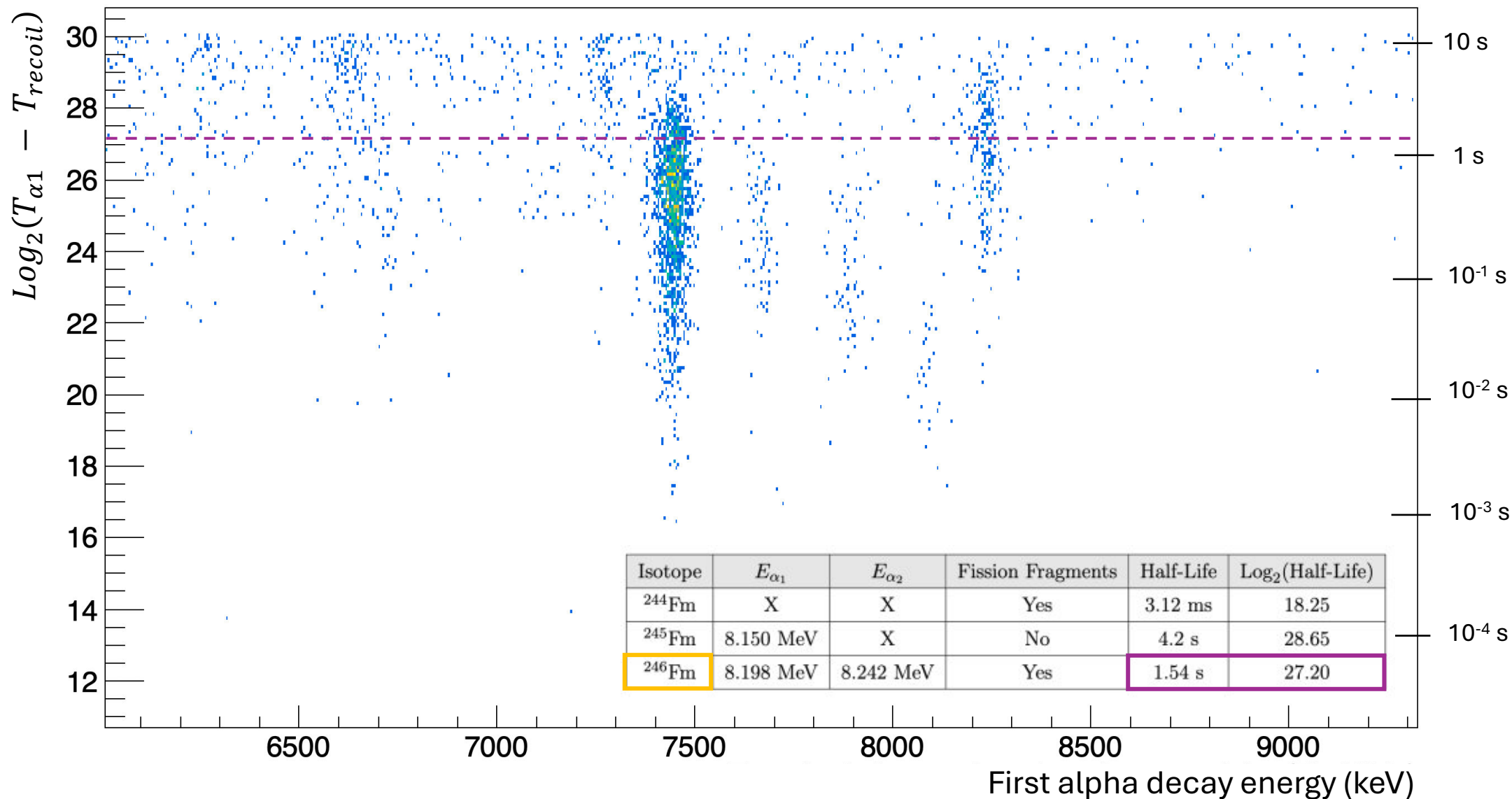


What I use to find it

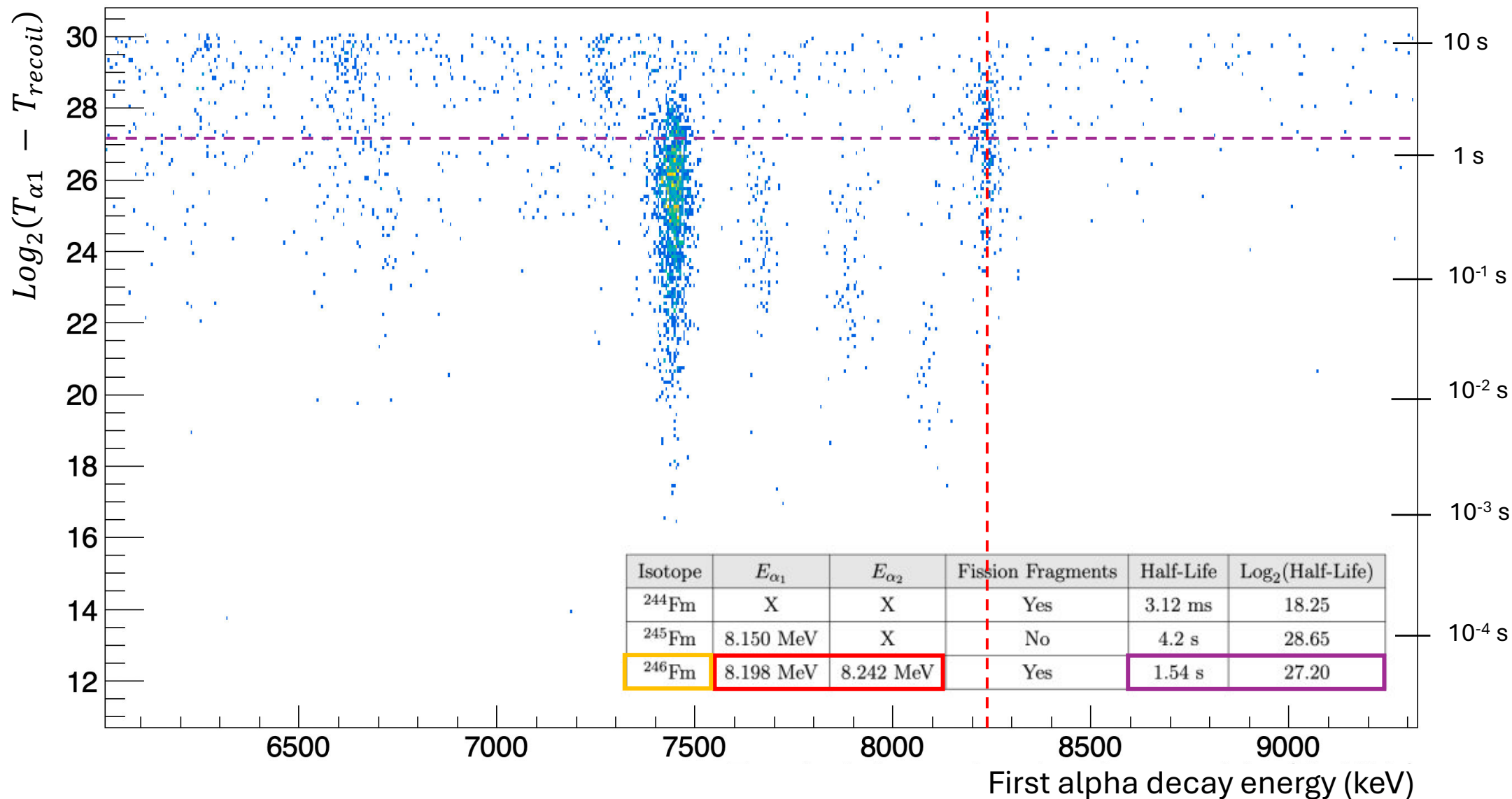
Plotting an Excitation Function: (1) Identifying ^{246}Fm α decays



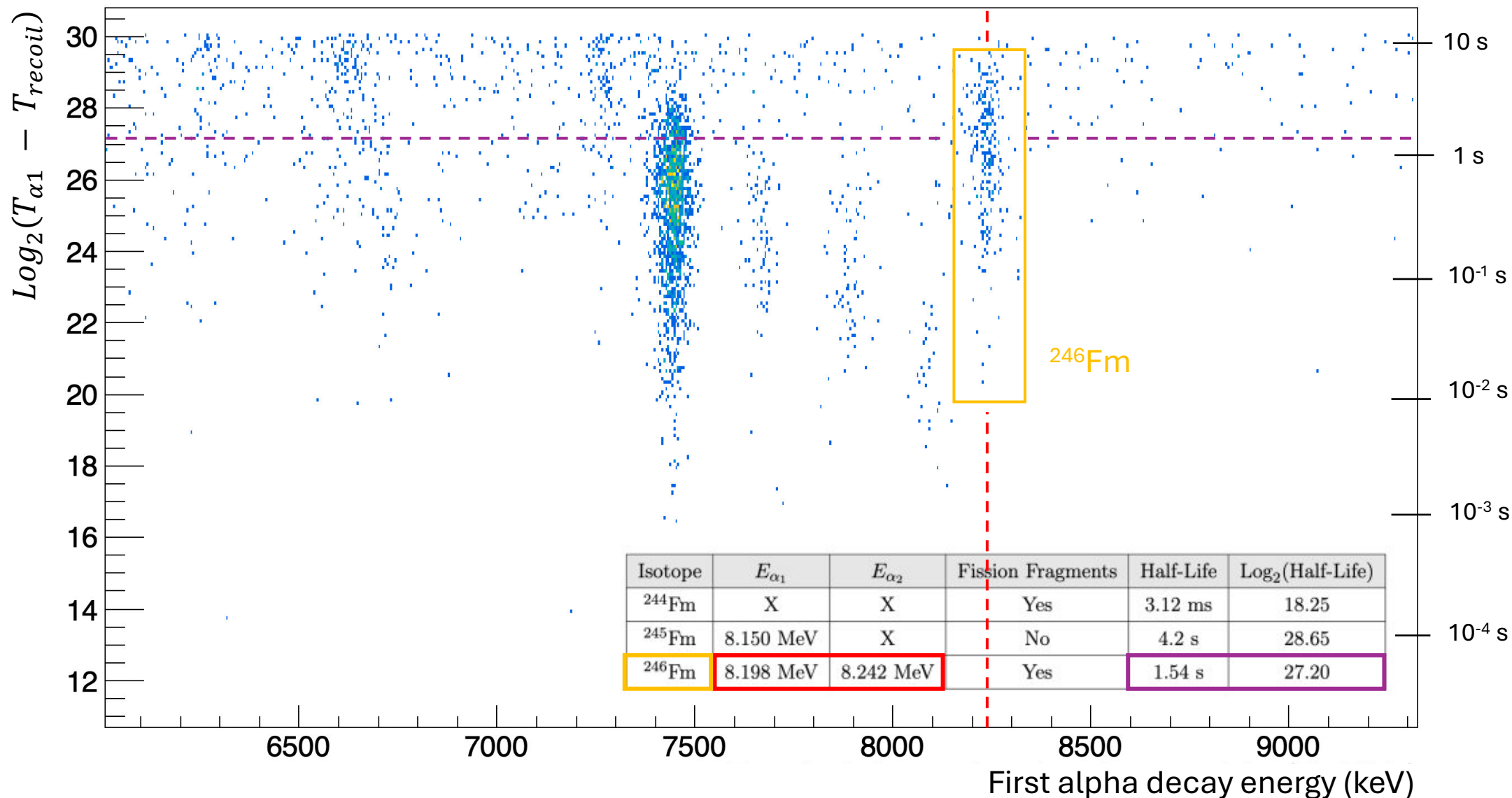
Plotting an Excitation Function: (1) Identifying ^{246}Fm α decays



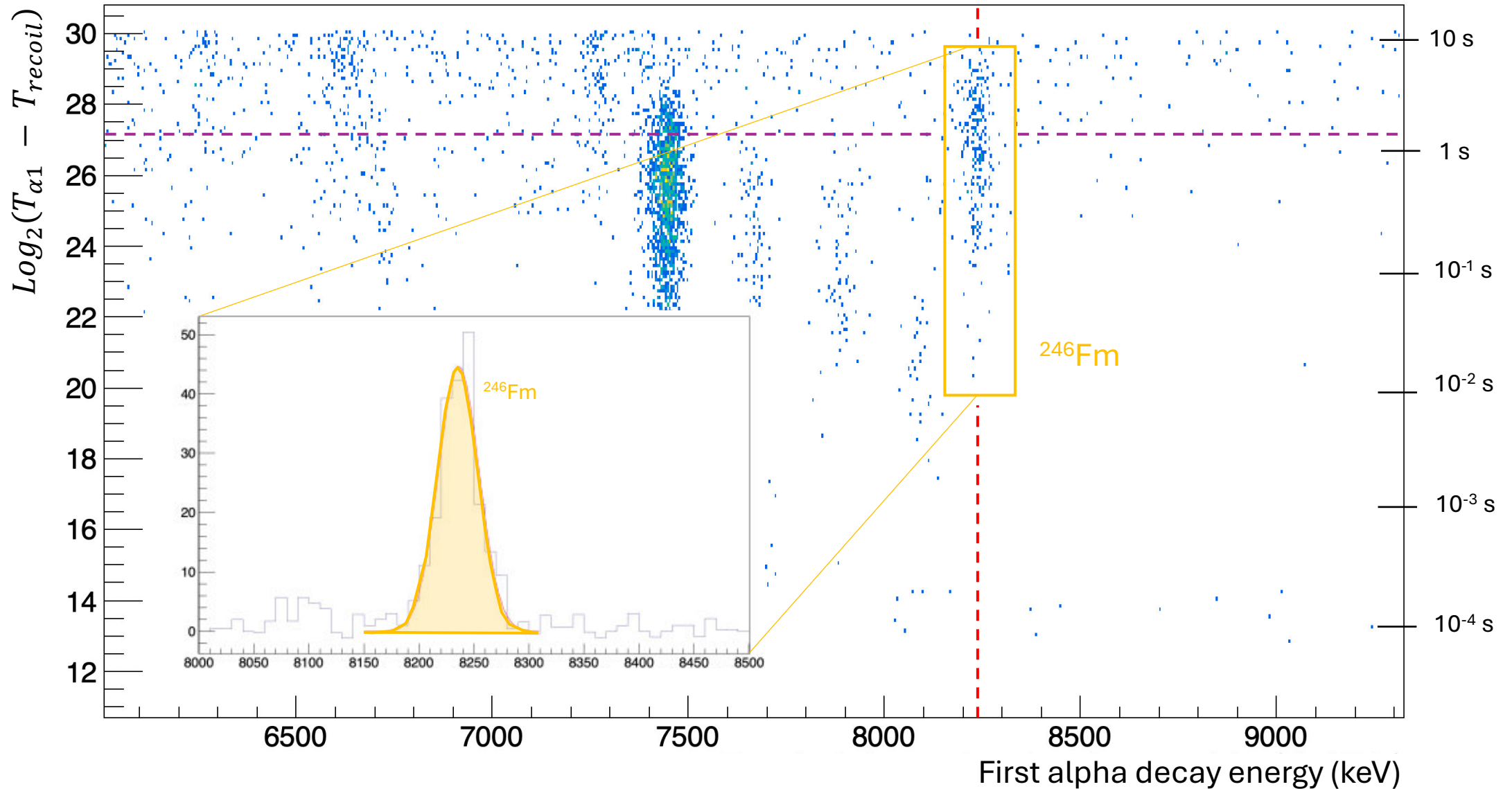
Plotting an Excitation Function: (1) Identifying ^{246}Fm α decays



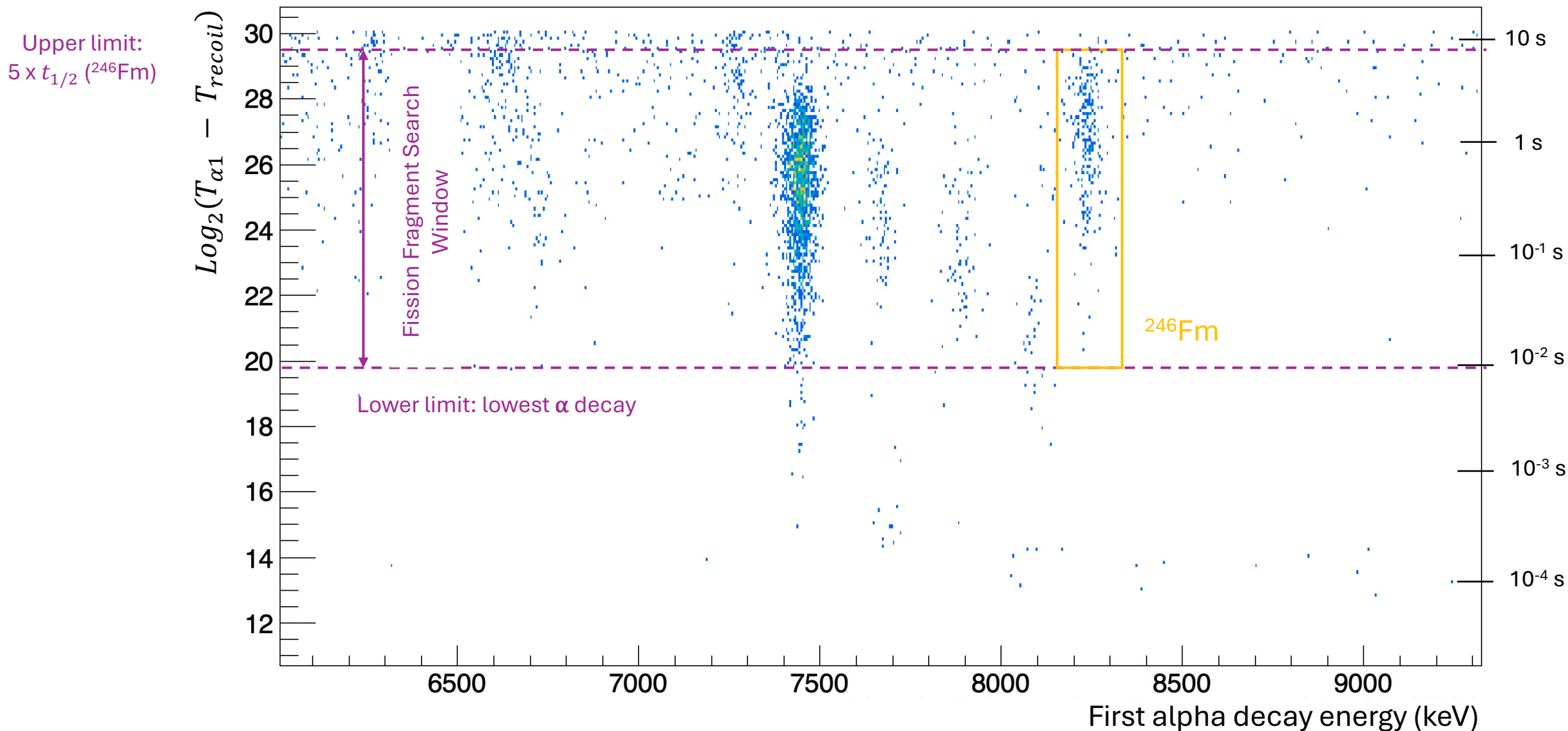
Plotting an Excitation Function: (1) Identifying ^{246}Fm α decays



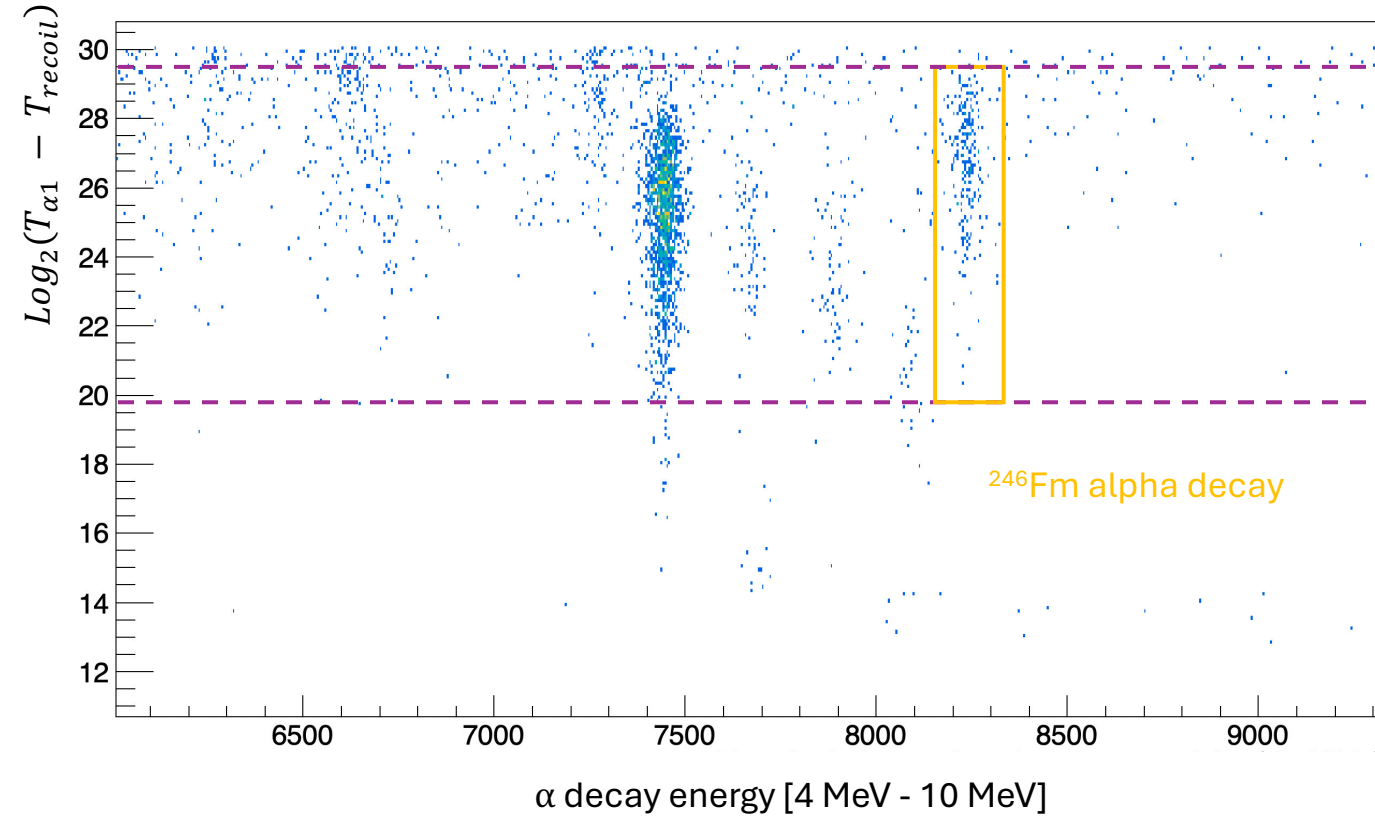
Plotting an Excitation Function: (1) Identifying ^{246}Fm α decays



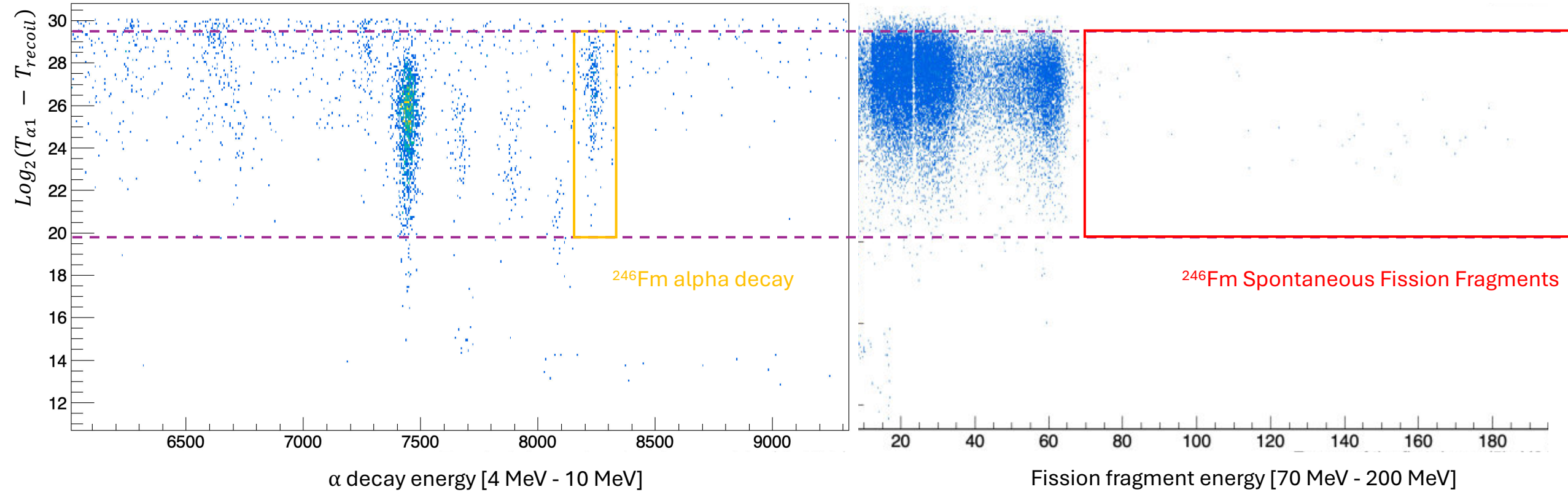
Plotting an Excitation Function: (2) Identifying ^{246}Fm Fission Fragments



Plotting an Excitation Function: Example for ^{246}Fm



Plotting an Excitation Function: Example for ^{246}Fm



Plotting an Excitation Function: Computing the Experimental Cross-Section

Results for all beam energies and all evaporation channels

$E_{\text{beam}}(\text{MeV})$	E^* (MeV)	^{244}Fm		^{245}Fm		^{246}Fm			
		N_{FF}	ΔN_{FF}	N_{α}	ΔN_{α}	N_{FF}	ΔN_{FF}	N_{α}	ΔN_{α}
197	34.71	38	6.16	1133.75	33.67	1	1	24	4.90
200	37.23	54	7.35	86	9.27	1	1	12	3.46
203	39.75	297	17.23	106.31	10.31	1	1	10	3.16
194	32.20	2	1.41	1352.81	36.78	6	2.45	38	6.16
191	29.67	1	1	655.15	25.60	23	4.80	149.66	12.23
185	24.65	1	1	48	6.93	23	4.80	218.8	14.80

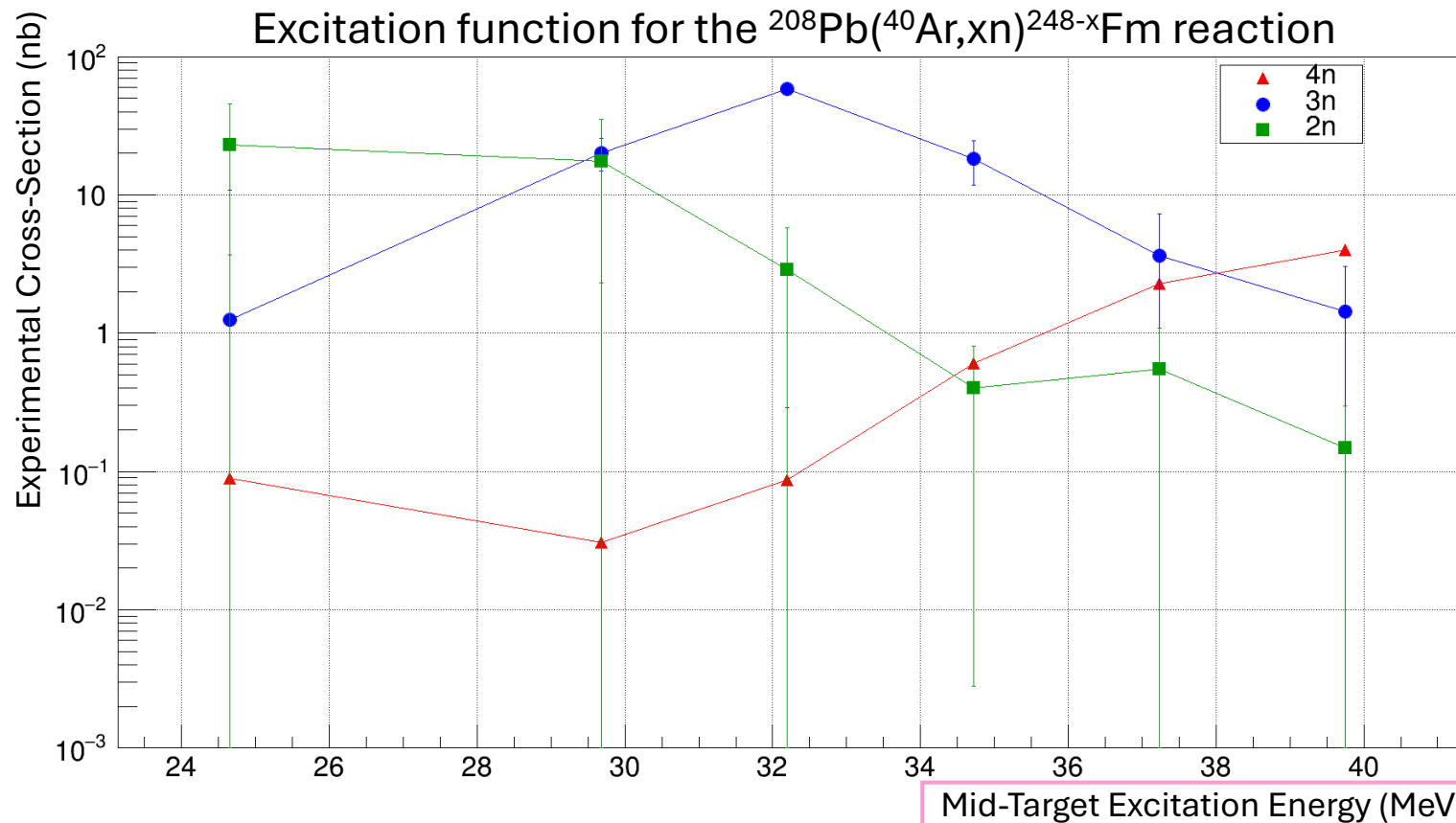
Table 7: Detected Isotopes

(cross-section) $\sigma = \frac{N_{\text{detected}}}{\text{dose} \times e \times \varepsilon \times t}$ (detection efficiency)

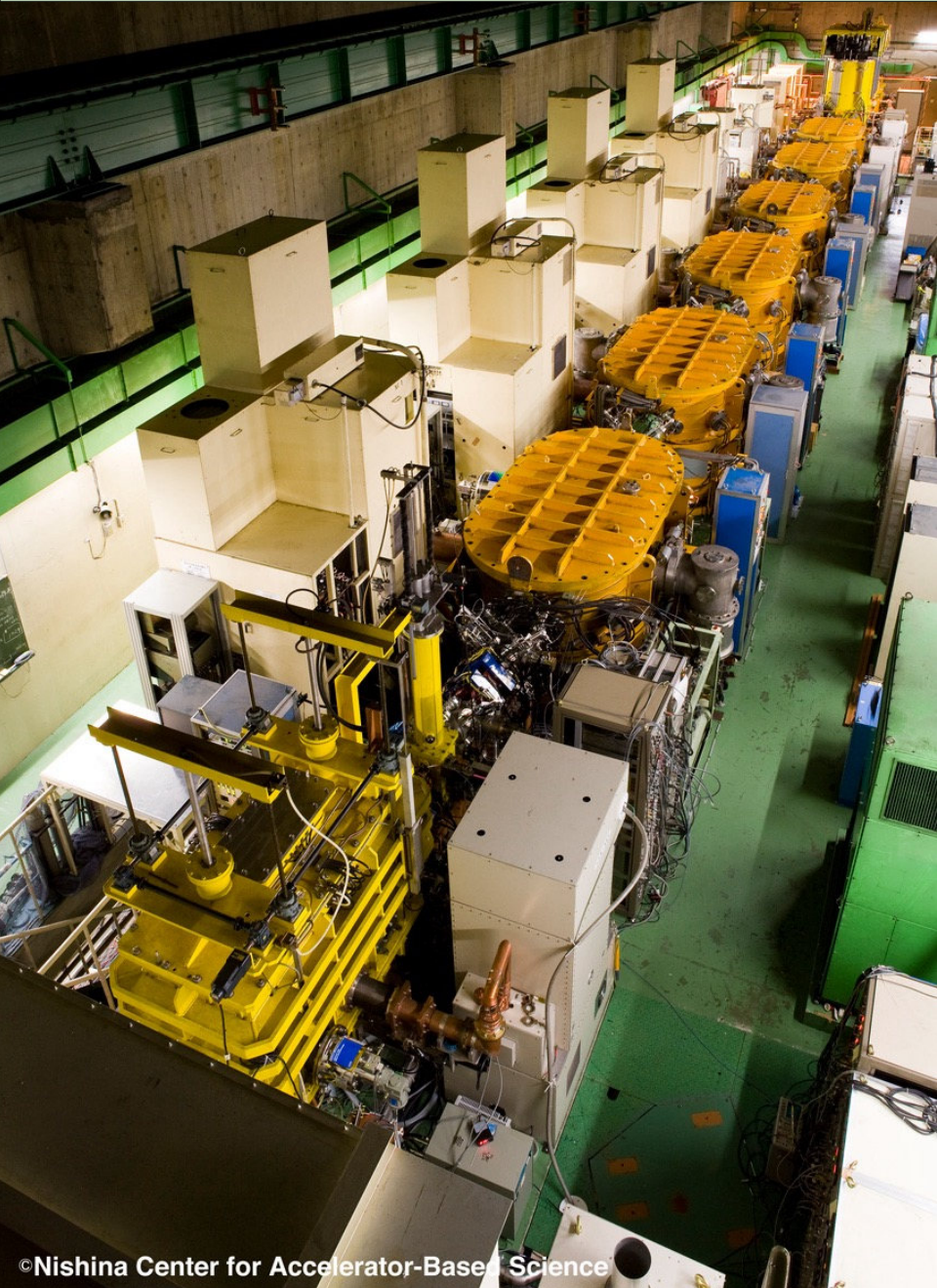
(target thickness)

Plotting an Excitation Function: Final Result

$E_{\text{beam}}(\text{MeV})$	$E^*(\text{MeV})$	^{244}Fm		^{245}Fm		^{246}Fm			
		N_{FF}	ΔN_{FF}	N_{α}	ΔN_{α}	N_{FF}	ΔN_{FF}	N_{α}	ΔN_{α}
197	34.71	38	6.16	1133.75	33.67	1	1	24	4.90
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Identifying Rare Event Decay Chains

The reaction I studied was ${}_{23}^{51}V + {}_{82}^{208}Pb \longrightarrow {}_{105}^{259}Db^* \longrightarrow {}_{105}^{259-x}Db + xn$ (for a single beam energy)

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Why study this reaction?

This reaction acts as a **surrogate** for the reaction used to synthesize element 119 for which the data is **confidential**. It uses the **same beam** on a different target and requires the **same type of search algorithm**.

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Channel	Isotope	E_{α} [MeV]	Half-Life	$\text{Log}_2(\text{Half-Life})$
2n	${}^{257}\text{Db}$	8.874	2.3 s	27.78
		8.965		
		9.066		
		9.155		
1n	${}^{258}\text{Db}$	9.014	4.3 s	28.68
		9.089		
		9.109		
		9.134		
		9.166		
		9.196		
		9.280		
9.353				

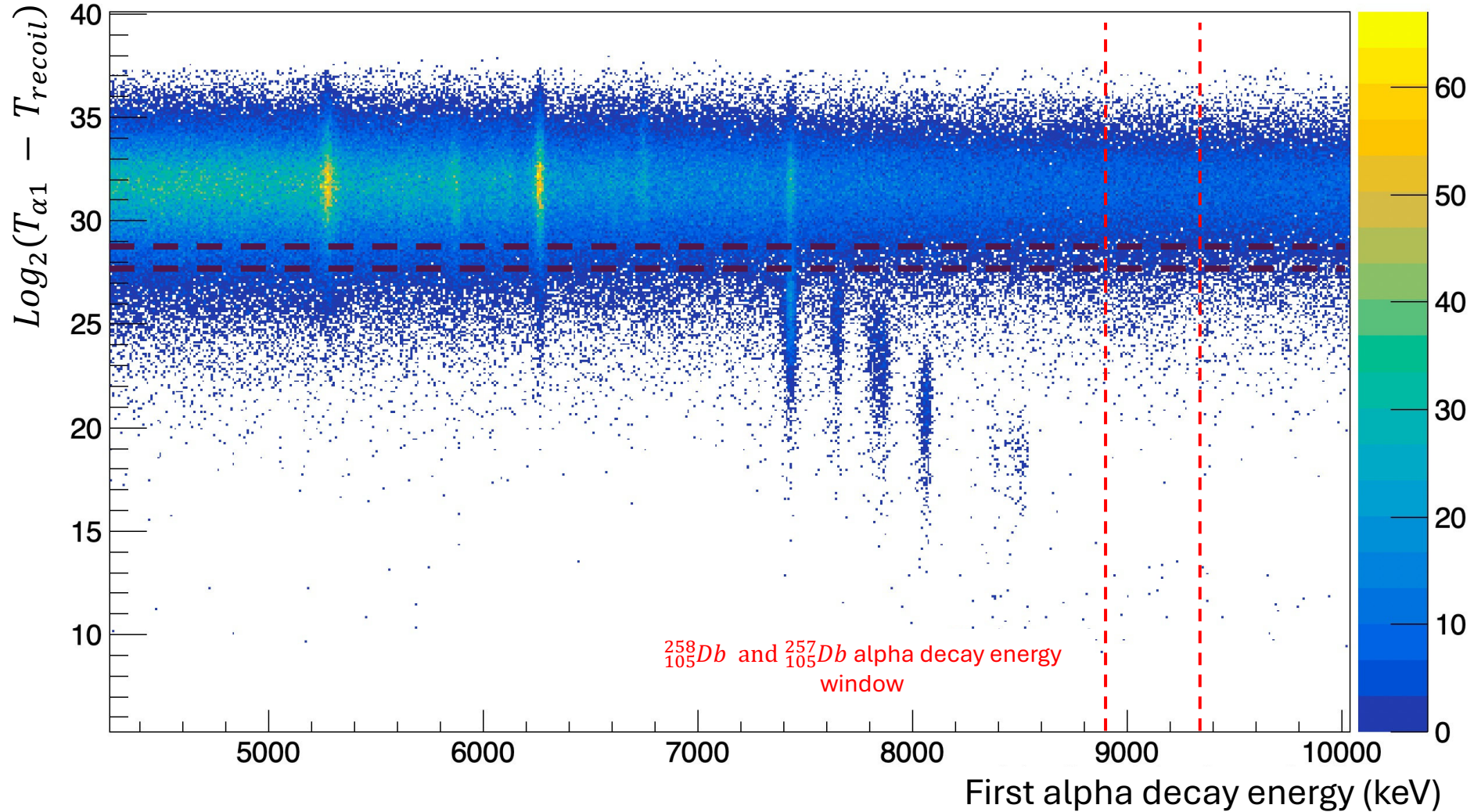
What I am looking for

What I use to find it (?)

Identifying Rare Event Decay Chains

Energy of the First Emitted Alpha vs Recoil Decay Time

Channel	Isotope	E_α [MeV]	Half-Life	$\text{Log}_2(\text{Half-Life})$
2n	^{257}Db	8.874	2.3 s	27.78
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		9.109		
		9.134		
		9.166		
		9.196		
		9.280		
9.353				



The isotopes we are looking for are drowned in the background...

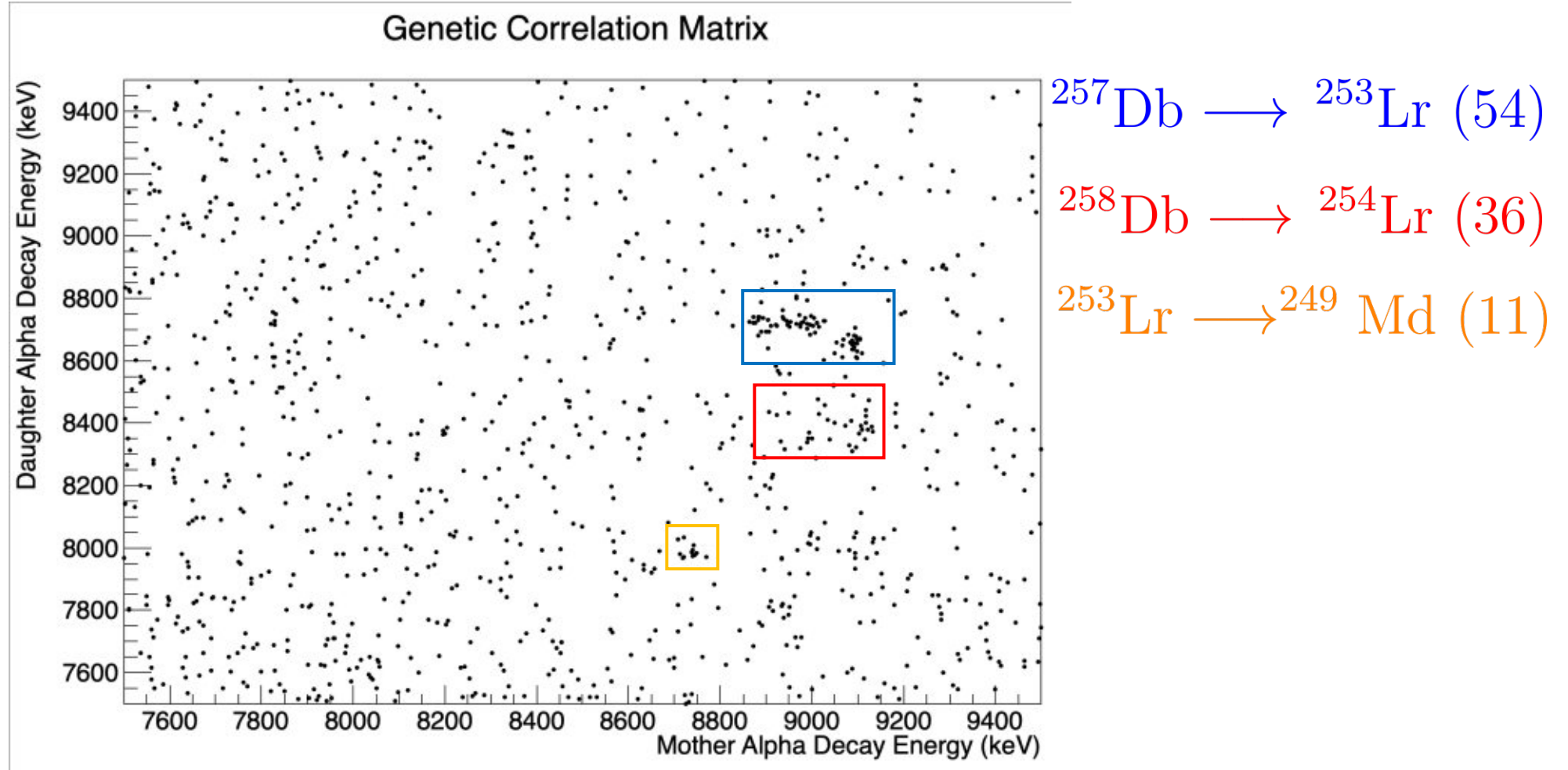
Identifying Rare Event Decay Chains : Genetic Correlations

Isotope	E_α [MeV]	Half-Life
^{257}Db	8.874	2.3 s
	8.965	
	9.066	
	9.155	
^{253}Lr	8.719	1.42 s
^{249}Md	8.026	24.8 s
^{258}Db	9.014	4.3 s
	9.089	
	9.109	
	9.134	
	9.166	
	9.196	
	9.280	
9.353		
^{254}Lr	8.408	13 s
	8.460	

Decay Chain 1 (rows 1-4)

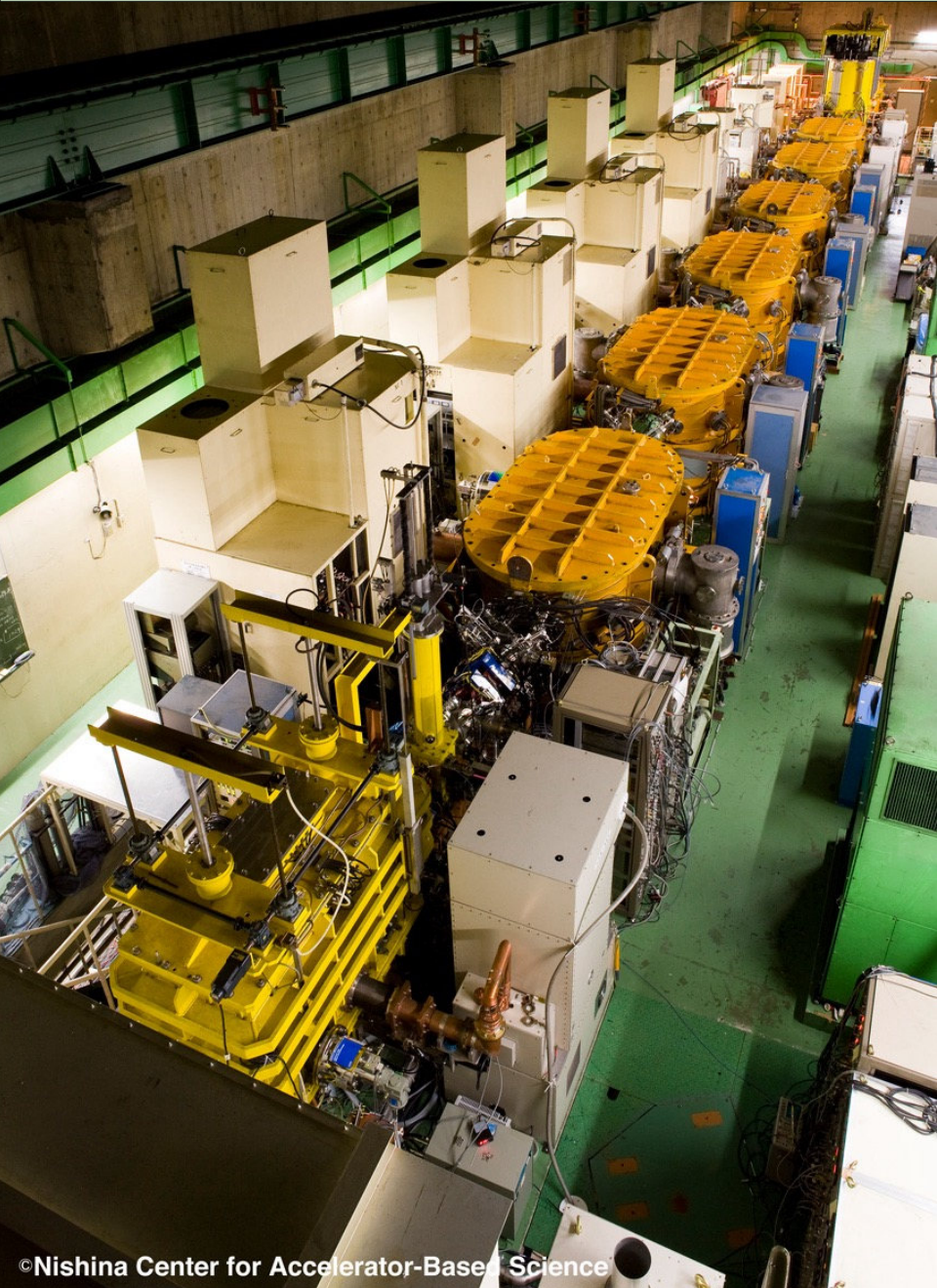
Decay Chain 2 (rows 5-10)

Identifying Rare Event Decay Chains : Genetic Correlations



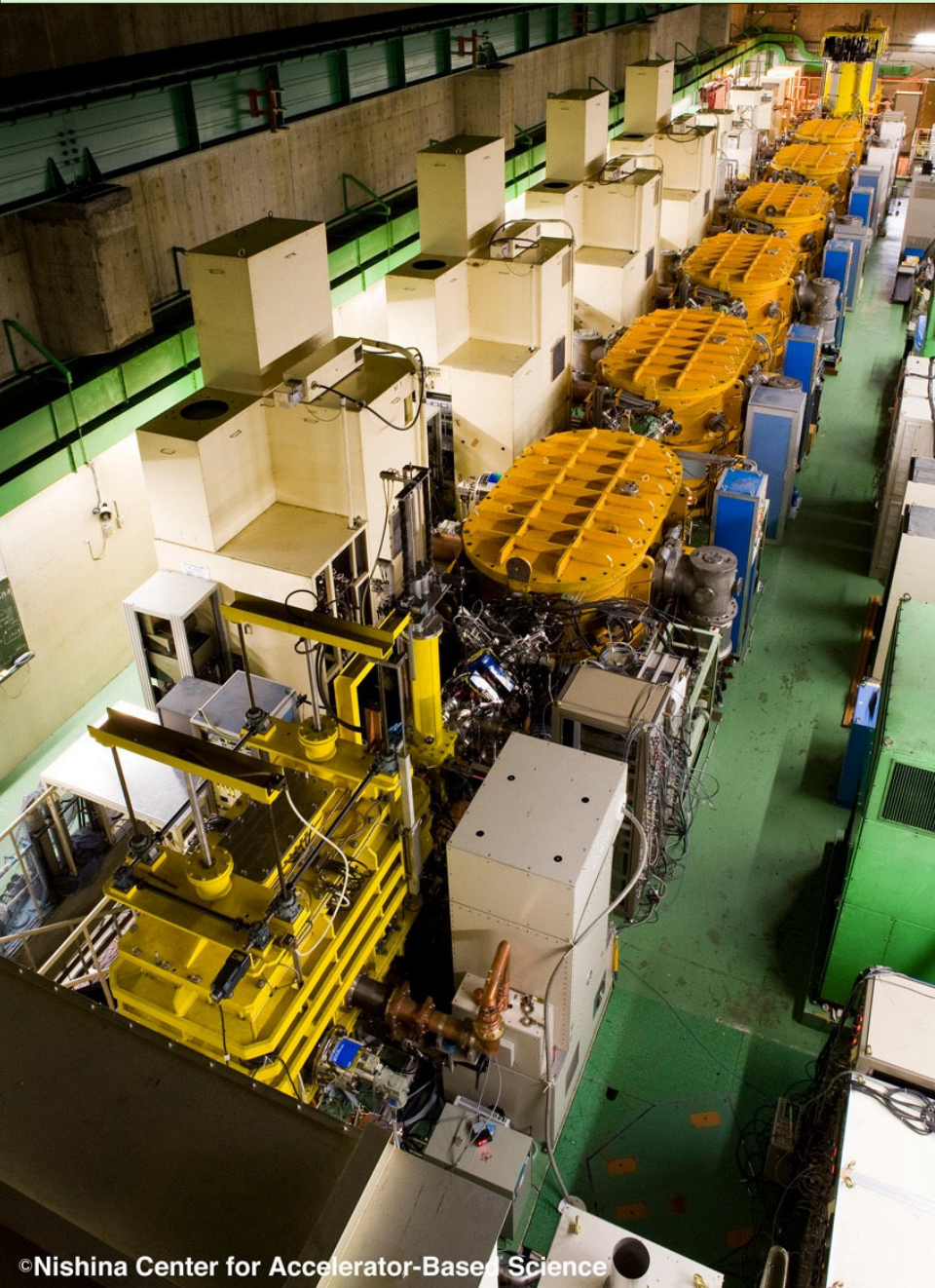
➔ A genetic correlation matrix is a powerful tool extract rare events from an overwhelming background.

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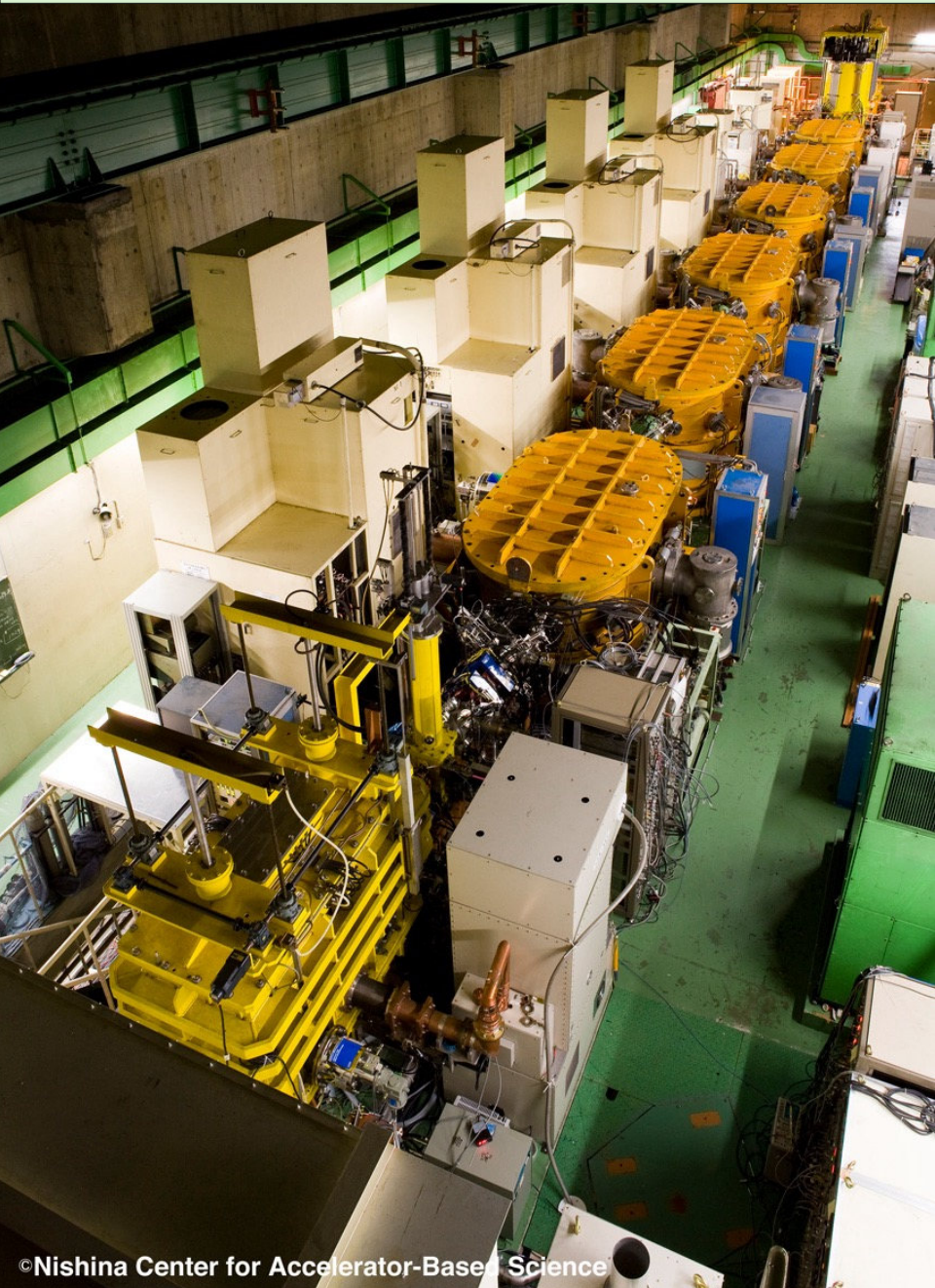
Conclusion & The Future of SHE Physics



The theoretical knowledge I acquired:

- Nuclear reaction mechanisms
- Nuclear structure and stability

Conclusion & The Future of SHE Physics



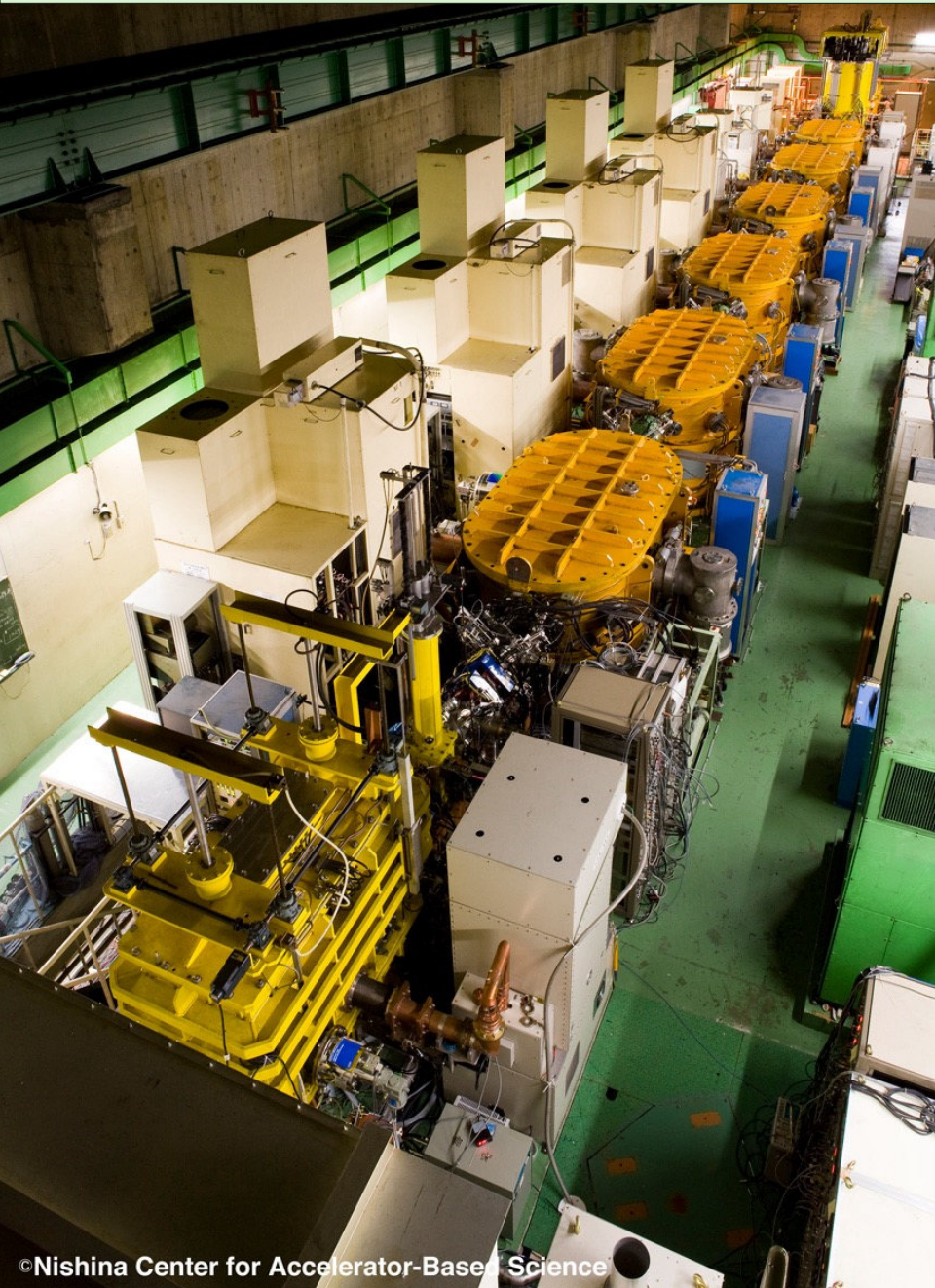
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What I accomplished:

- I computed the excitation function of ${}^{40}_{18}\text{Ar} + {}^{208}_{82}\text{Pb}$ and determined which beam energies favored which evaporation channels.
- I successfully extracted the decay chains of ${}^{257}\text{Db}$ from an overwhelming amount of background events.

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- I successfully extracted the decay chains of ${}^{257}\text{Db}$ from an overwhelming amount of background events.

The future of SHE physics:

- Elements 119 and 120 are being synthesized at RIKEN (Japan) and Berkeley (USA) laboratories
- Ongoing programs for SHE spectroscopy worldwide (France, Finland, USA, Russia)
- Metallic Beams & target backings are being researched by our colleagues at IPHC!

List of Figure References

- [1] Soti, Z., Magill, J., & Dreher, R. (2019). Karlsruhe Nuclide Chart – New 10th Edition 2018. *EPJ Nuclear Sciences & Technologies*, 5(6). ([link](#))
- [2] Nilsson, S. G., Tsang, C. F., Sobiczewski, A., Szymanski, Z., Wycech, S., Gustafson, C., Lamm, I.-L., Möller, P., & Nilsson, B. (1969). On the Nuclear Structure and Stability of Heavy and Superheavy Elements. *Nuclear Physics, Section A*, 131, 1-66. ([link](#))
- [3] Piot, J. (2010). Sur la route de l'Îlot de Stabilité Superlourd : Spectroscopie Prompte des Noyaux ^{246}Fm et ^{256}Rf . *Thesis, University of Strasbourg*. ([link](#))
- [4] Schädel, M. (2012). Chemistry of Superheavy Elements. *Radiochimica Acta*, 100(8-9), 579-604. ([link](#))
- [5] Smits, et al. (2020). Oganesson: A Noble Gas Element That Is Neither Noble Nor a Gas. *Angewandte Chemie*, 59(52). ([link](#))
- [6] Hofmann, S. (2015). Super Heavy Nuclei. *Journal of Physics G: Nuclear and Particle Physics*, 42, 114001. ([link](#))
- [7] Sakai, H., Haba, H., Morimoto, K. et al. (2022). Facility Upgrade for Superheavy-Element Research at RIKEN. *The European Physical Journal A*, 58, 238. ([link](#))
- [8] Kaji, D., Morimoto, K., Sato, N., Yoneda, A., & Morita, K. (2013). Gas-filled Recoil Ion Separator GARIS-II. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 317(Part B), 311-314. ([link](#))

Backup 1 - “Semibubble” Structure Prediction

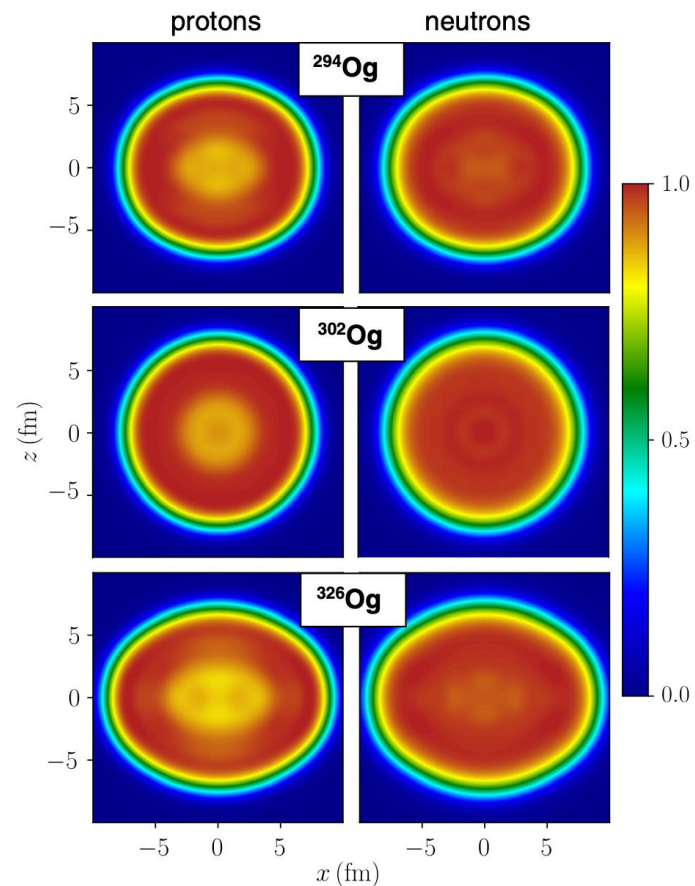
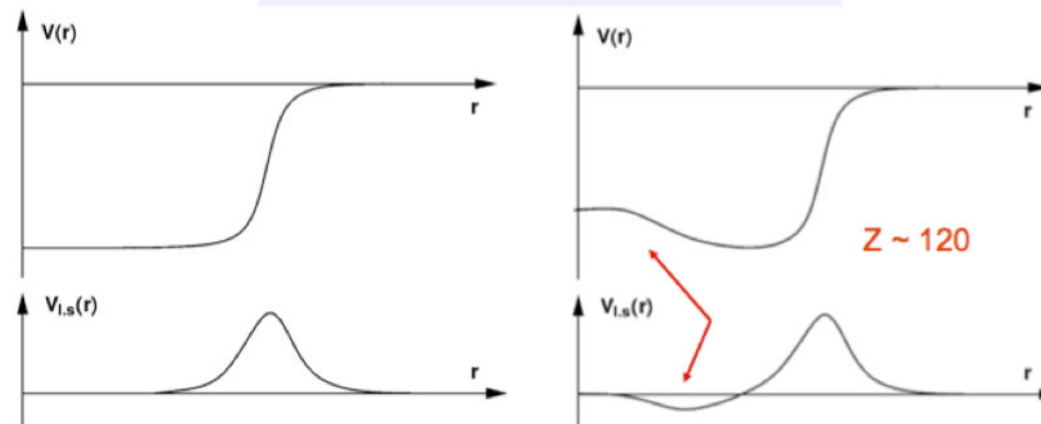


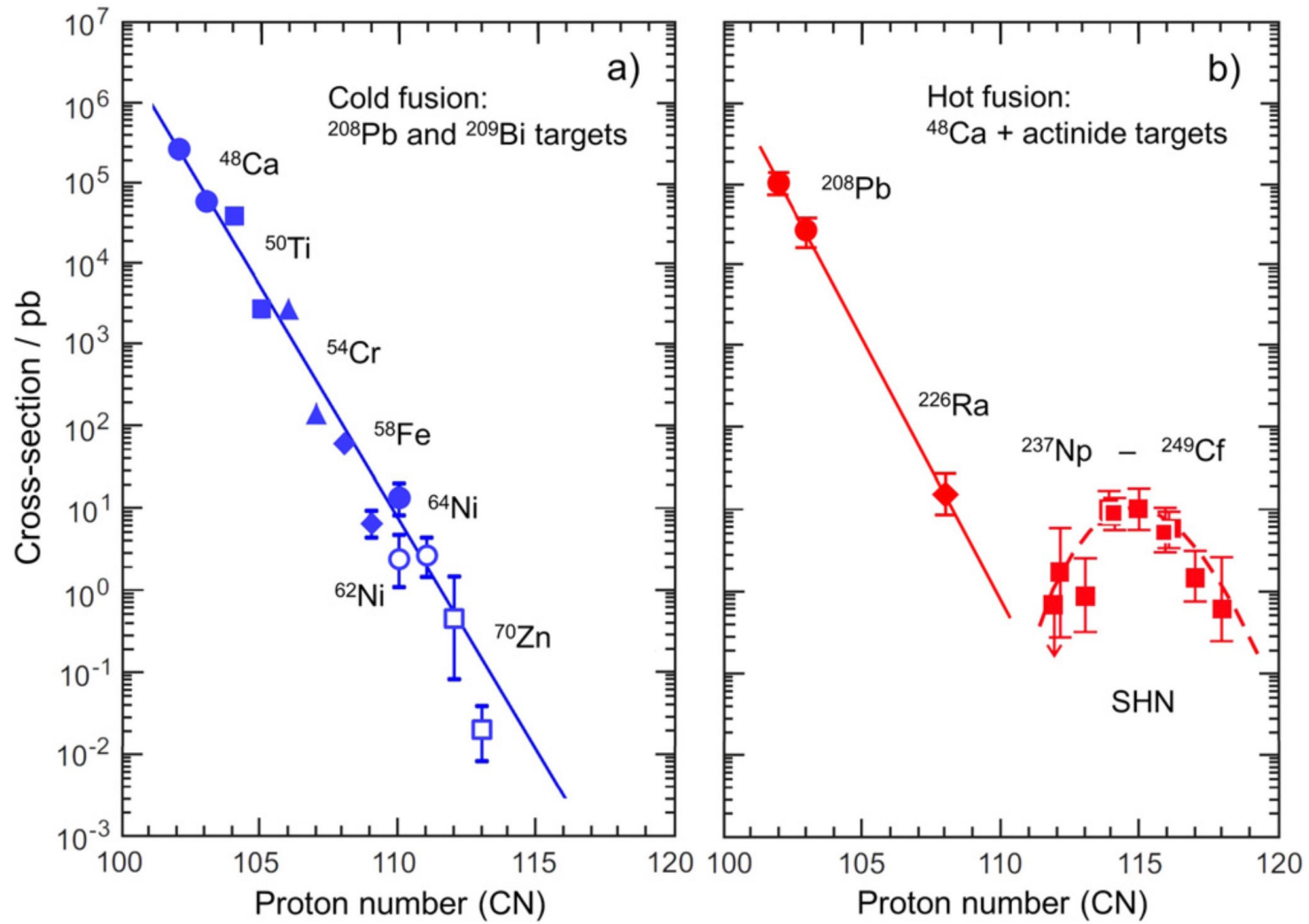
FIG. 3. Proton (left) and neutron (right) densities of ^{294}Og (top), ^{302}Og (middle), and ^{326}Og (bottom) calculated with SV-min in the (x, z) plane at $y = 0$. The densities are normalized to the maximum density. The central depression of the proton density (semibubble structures) is clearly seen in all three cases. From Schuetrumpf, Nazarewicz, and Reinhard, 2017.

$$V_{l.s}(r) = -\frac{1}{r} \frac{\partial V(r)}{\partial r}$$



Total spin-orbit splitting depends on the location of the radial wavefunctions

Backup 1 - Cross-Section



Backup 2 - Range-Energy Relation for Alpha Particles in Silicon

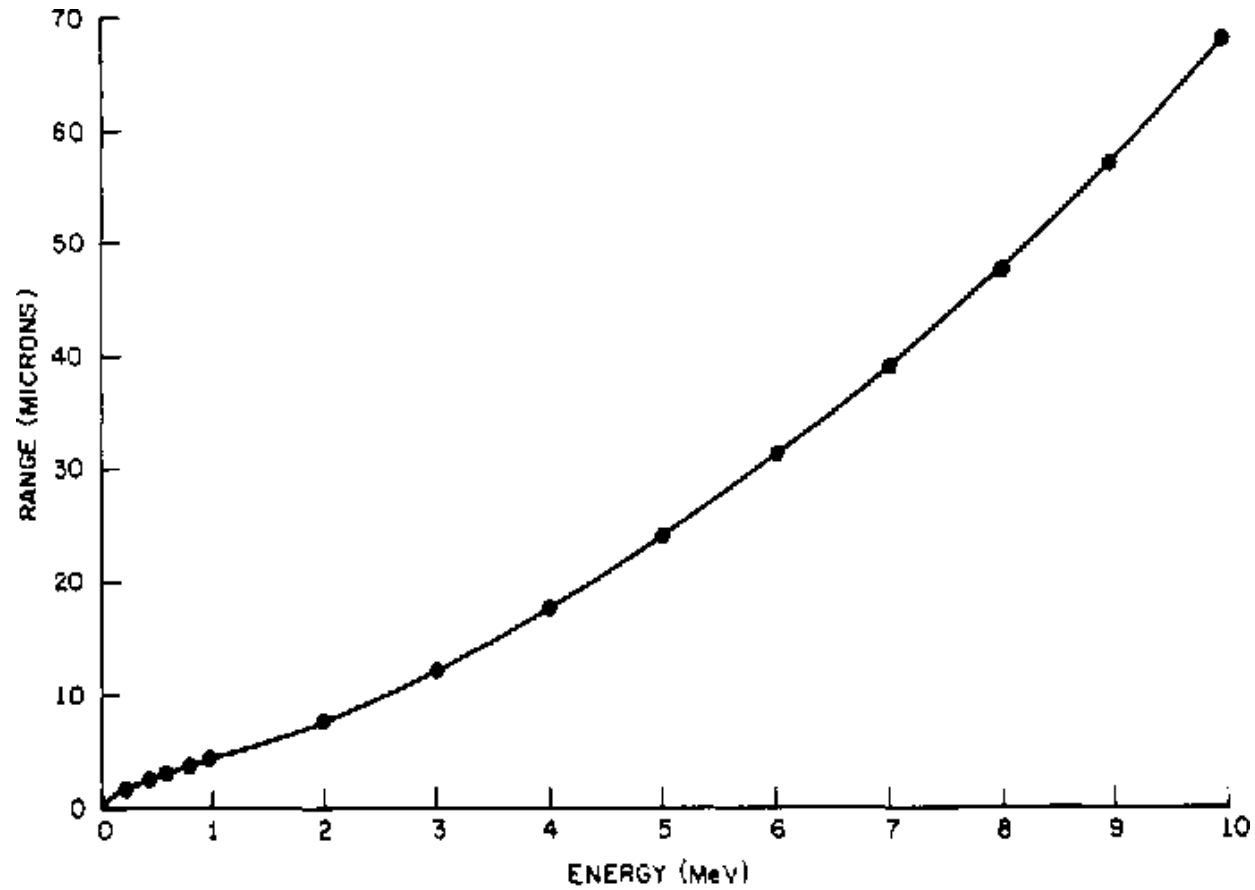


Fig. 1. Range-energy relation for alpha particles in silicon.

Backup 3 - Genetic Correlations for Z = 113



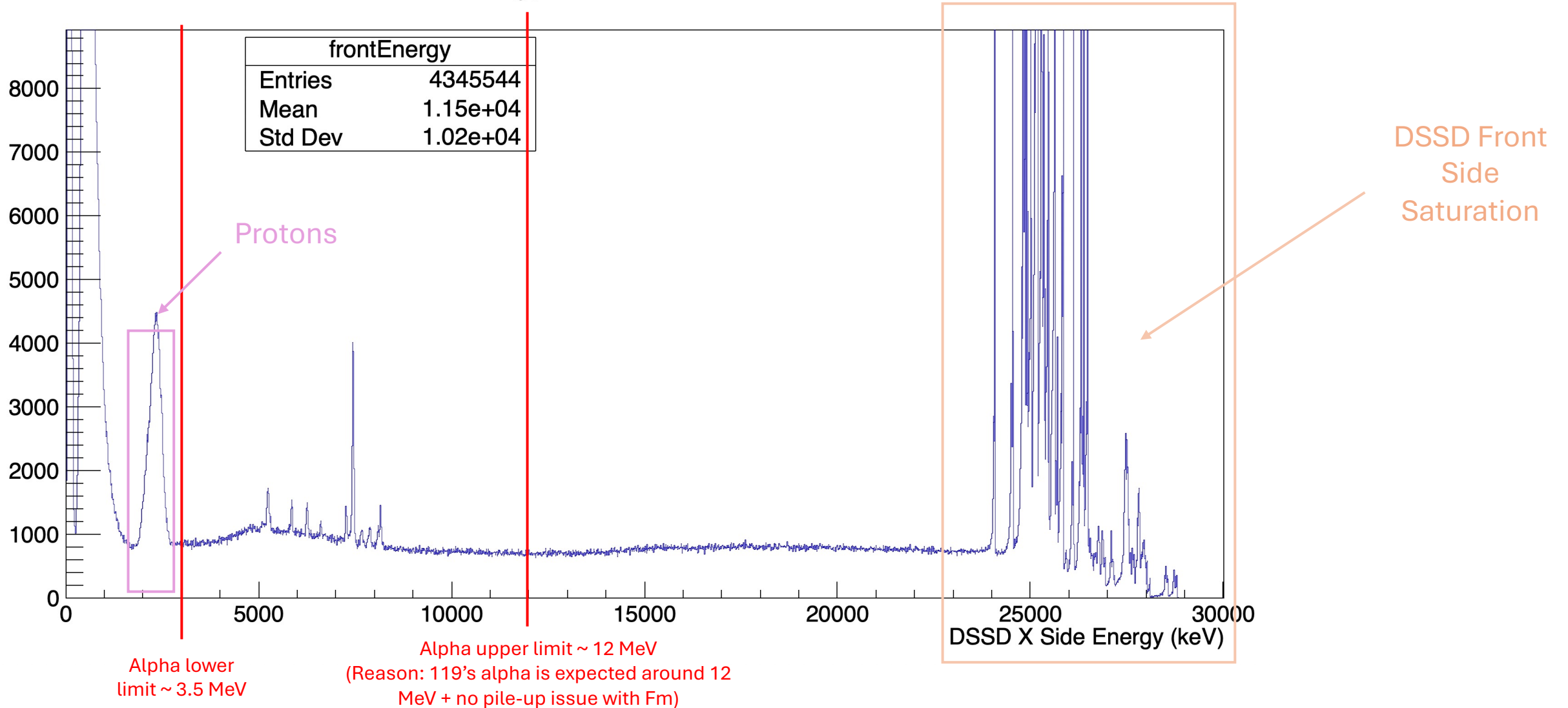
Fig. 17: Decay chains attributed to start from ²⁷⁸Nh [135]

Isotope	chain 1		chain2		chain 3	
	E _α /MeV	T _{1/2}	E _α /MeV	T _{1/2}	E _α /MeV	T _{1/2}
²⁷⁸ 113	11.68±0.04	0.344 ms	11.52±0.04	4.93 ms	11.82±0.06	0.667 ms
²⁷⁴ Rg	11.15±0.07	9.26 ms	11.31±0.07	34.3 ms	10.65±0.06	9.97 ms
²⁷⁰ Mt	10.03±0.07	7.16 ms	2.32 (esc)	1.63 s	10.26±0.07	444 ms
²⁶⁶ Bh	9.08±0.04	2.47 s	9.77±0.04	1.31 s	9.39±0.06	5.26 s
²⁶² Db	sf	40.9 s	sf	0.787 s	8.63±0.06	126 s
²⁵⁸ Lr					8.66±0.06	3.78 s

Tab. 5: Decay chains observed at GARIS, RIKEN in the reaction ⁷⁰Zn + ²⁰⁹Bi and interpreted to start from ²⁷⁸113 [135]. 'esc' denotes that the α particle escaped the 'stop' detector and only an energy loss signal was recorded.

Backup 4 - DSSD Front Side Saturation

Energy Read on the X Side



Backup 4 - Alpha Decay Spectrum

DSSD Energy on the X Side for Internal Events

