A Tale of Two Isotopes: ^{88}Zr and ^{135}Xe

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A Tale of Two Isotopes: ^{88}Zr and ^{135}Xe

^{88}Zr

- Second largest thermal neutron capture cross section

- Not a fission product
- Neutron-Poor
- Large neutron absorption cross section discovered in 2019
- Radioactive (83.4 days)

- Let's start our journey here

^{135}Xe

- Largest thermal neutron capture cross section
  - Or is it…

- Fission product
- Neutron-Rich
- Large neutron absorption cross section discovered in 1944
- Radioactive (9.14 hours)

- Also, 2026 ILL FIPPS campaign on other xenon neutron capture prompt gammas

^{88}Zr(n,γ) - Background

- In 2019, ^{88}Zr was discovered to have a thermal neutron absorption cross section of ~800,000 barns (measured) rather than 10 barns (“expected”).
  - Larger than ^{157}Gd, ^{10}B, ^{6}Li, ^{3}He
  - Smaller than ^{135}Xe

Perhaps more surprising, many argue that thermal neutron cross sections are not able to be predicted!!!
S. Heinitz, U. Köster Nature Physics volume 15, 208–209 (2019)

- ^{88}Zr is the only neutron absorber above 10,000 barns with even neutron number

Zr 88	Zr 89	Zr 90	Zr 91	Zr 92	Zr 93	Zr 94	Zr 95	Zr 96
83.4 days	3.27 days	51.45%	11.22%	17.15%	1.6×10^6 years	17.38%	64.0 days	2.8%
								2×10^{19} years
σ: 861,000 b		σ: 0.01 b	σ: 1.3 b	σ: 0.13 b	σ: 0.7 b	σ: 0.05 b		σ: 0.02 b

https://doi.org/10.1038/s41586-018-0838-z

LETTER

The surprisingly large neutron capture cross-section of ^{88}Zr

Jennifer A. Shusterman[1,2,3]*, Nicholas D. Scielzo[1], Keenan J. Thomas[1], Eric B. Norman[4], Suzanne E. Lapi[5], C. Shaun Loveless[5], Nickie J. Peters[6], J. David Robertson[6], Dawn A. Shaughnessy[1] & Anton P. Tonchev[1]

^{88}Zr is a neutron sponge – Who cares?

- Nuclear physicists
  - How are we wrong about basic properties by 4 orders of magnitude?

- High energy zirconium applications
  - Accelerator and spallation sources

Is this massive cross section relating to new fundamental nuclear physics?

Does ^{88}Zr 'feel a tug' towards the magic number of 50 neutrons (^{90}Zr)?

Or was there simply an unexpected resonance at just the right (thermal) energy?

^{88}Zr(

- Fit ratio of most prominent ^{89m}Zr
- Shape is predominantly ^{89m}Zr 4.2 … ıdance, except that ^{89m}Zr decays also fee
- Vary isomeric yield ratio as well a
- Best fit at χ^2 minimum
  - 7 penalty terms associated
- 68% C.L. at χ^2 increase of 1

$$\chi^2 = \chi^2_{min} + \Delta\chi^2$$

Parameter	
Cross section × flux	3.52 ± 0.
^{88}Zr half-life	83.4
^{89}Zr half-life	78.361
^{89m}Zr half-life	4.161 ±
^{89m}Y half-life	15.663
Rel. Eff. × ICC	1.44671
^{89m}Zr branching ratio	0.9377

PHYSICAL REVIEW C **111**, L021601 (2025)

Letter

Measurement of the isomeric yield ratio for ^{88}Zr thermal neutron absorption

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In light of the recently observed 8×10^5 barn thermal neutron absorption cross section of zirconium-88, this work investigates the rate (isomeric yield ratio) of metastable zirconium-89 production and resulting implications for ongoing measurements around zirconium-88 neutron absorption. The metastable state of zirconium-89 resides at 588 keV above the ground state with a half-life of 4.2 min. A 5 μCi zirconium-88 sample was irradiated for 10 min in the core of a TRIGA Mark II nuclear research reactor and measured with a high purity germanium detector starting 3 min after irradiation. The isomeric yield ratio was measured to be 74.9 ± 0.6%.

DOI: 10.1103/PhysRevC.111.L021601

Introduction. The thermal neutron absorption cross section of zirconium-88 (^{88}Zr) was measured to be $8.61 \times 10^5 \pm 6.9 \times 10^4$ barns in 2019 when a 10 barn cross section was expected [1]. This value was measured again as $8.04 \times 10^5 \pm 6.3 \times 10^4$ barns in a 2021 measurement by the same group [2] and remains an intriguing result to the nuclear physics community. An energy-resolved transmission-based measurement has reported a $7.71 \times 10^5 \pm 3.1 \times 10^4$ barn thermal cross section with a resonance at 0.171 eV [3]. An energy-resolved direct capture-based measurement is being performed by the authors of this study and additional collaborators at the CERN n_TOF facility [4].

A traditional neutron capture measurement uses γ-ray spectroscopy to measure the absolute quantities of reaction products and reactants before and after irradiation. The capture products and related decay chains must be carefully defined to maximize precision. ^{88}Zr in neutron flux undergoes the reaction ^{88}Zr$(n,\gamma)^{89}$Zr at a high rate due to the large cross section. The product ^{89}Zr compound nucleus has significant excess energy (9.3 MeV) which is dissipated in a cascade through nuclear energy levels to the ground state, ^{89g}Zr. Some fraction of these nuclei are left in a metastable state, ^{89m}Zr, at a rate defined as the isomeric yield ratio: $\mathrm{IYR} \equiv \frac{^{88}Zr(n,\gamma)^{89m}Zr}{^{88}Zr(n,\gamma)^{89(m,g)}Zr}$. ^{89m}Zr emits a characteristic 588 keV γ ray upon decay to ground state; its short half-life (4.2 min) motivates counting promptly after exposure. The decay of the ground state ^{89g}Zr in the sample is measured simultaneously by counting a 909 keV γ. The relevant absorption and decay paths are summarized in Fig. 1.

While the experimental and theory communities evaluate and explain the large ^{88}Zr neutron absorption cross section, a measurement of the isomeric yield ratio is of interest for a few reasons. First, the IYR of the ^{88}Zr(n,γ) reaction impacts previous measurements of the cross section when the amount of ^{89}Zr is measured after a known fluence. Rather than an isomeric transition to the ^{89}Zr ground state, 6.2% of ^{89m}Zr decays to ^{89}Y [5]. Therefore the amount of ^{89}Zr formed by ^{88}Zr(n,γ) depends on the fraction of reactions with ^{89m}Zr as an intermediate state. Second, the IYR directly impacts the amount of prompt γ energy released during such capture reactions (8.7 vs 9.3 MeV). Finally, the IYR of ^{89m}Zr during ^{88}Zr neutron capture is not previously measured and such a measurement allows tuning of gamma cascade generators for ^{88}Zr(n,γ) direct capture measurements [6].

Background. The isomeric yield ratio of ^{89}Zr after other reactions has been observed previously. Mangal and Gill measured the IYR of ^{90}Zr$(n,2n)^{89}$Zr with 14 MeV incident neutrons to be 0.72 ± 0.08 [7]. Katz, Baker, and Montalbetti measured the IYR of ^{90}Zr$(\gamma,n)^{89}$Zr to have an energy dependence but remain roughly constant at 0.56 ± 0.03 above an incident γ energy of 16 MeV [8]. Satheesh, Musthafa, Singh, and Prasad found the IYR of ^{89}Y$(p,n)^{89}$Zr to vary between 0.2 and 0.4 at an energy range between 6 MeV and 16 MeV [9]. These disparate values are expected as the various reactions correspond to a range of compound nuclear energies and initial nuclear spin configurations, and none are directly applicable to the ^{88}Zr$(n,\gamma)^{89}$Zr reaction which we have explored. Though no previous such measurements are available, the nuclear reaction model code TALYS [10] with ^{89}Zr energy levels given by the RIPL-3 database [11] give an IYR of 0.89. The metastable state is preferred given that ^{88}Zr is 0+ and capture of a thermal neutron should be s-wave and hence lead to a dominant production of the 1/2− (metastable) isomer.

Experimental setup. A sample of ^{88}Zr was produced at Los Alamos National Laboratory Isotope Production Facility through proton irradiation of a yttrium target ^{89}Y$(p,2n)^{88}$Zr and was delivered to the Nuclear Engineering Teaching Laboratory (NETL) at the University of Texas at Austin in 2N HCl solution. ^{88}Zr electron capture decays to ^{88}Y with a 83.4 day half-life which further decays through electron capture to stable ^{88}Sr with a 106.6 d half-life. As the sample was over a year past initial separation, the ^{88}Y activity exceeded the ^{88}Zr activity. Furthermore, ^{88}Y γs, predominately 898 keV (93% branching ratio) and 1.836 MeV (99% branching ratio),

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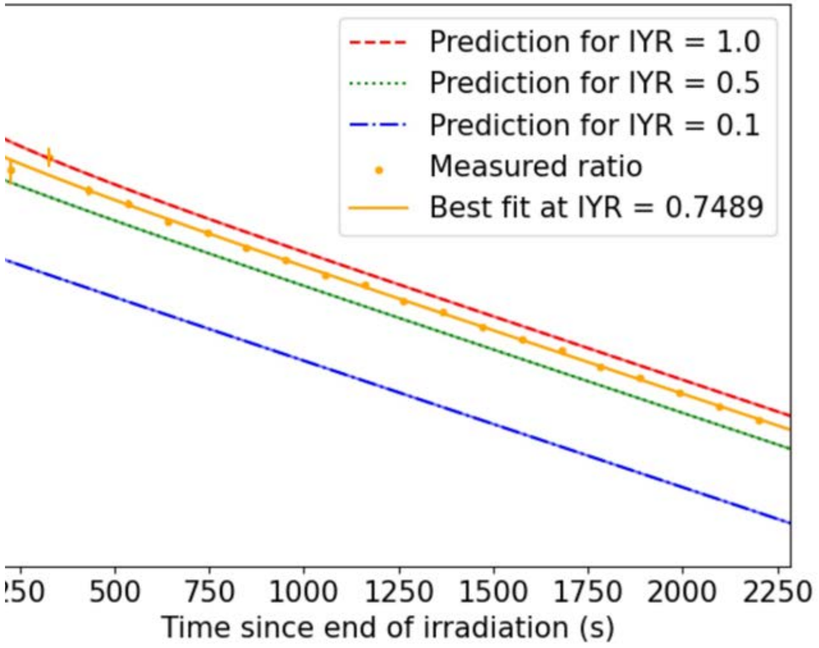
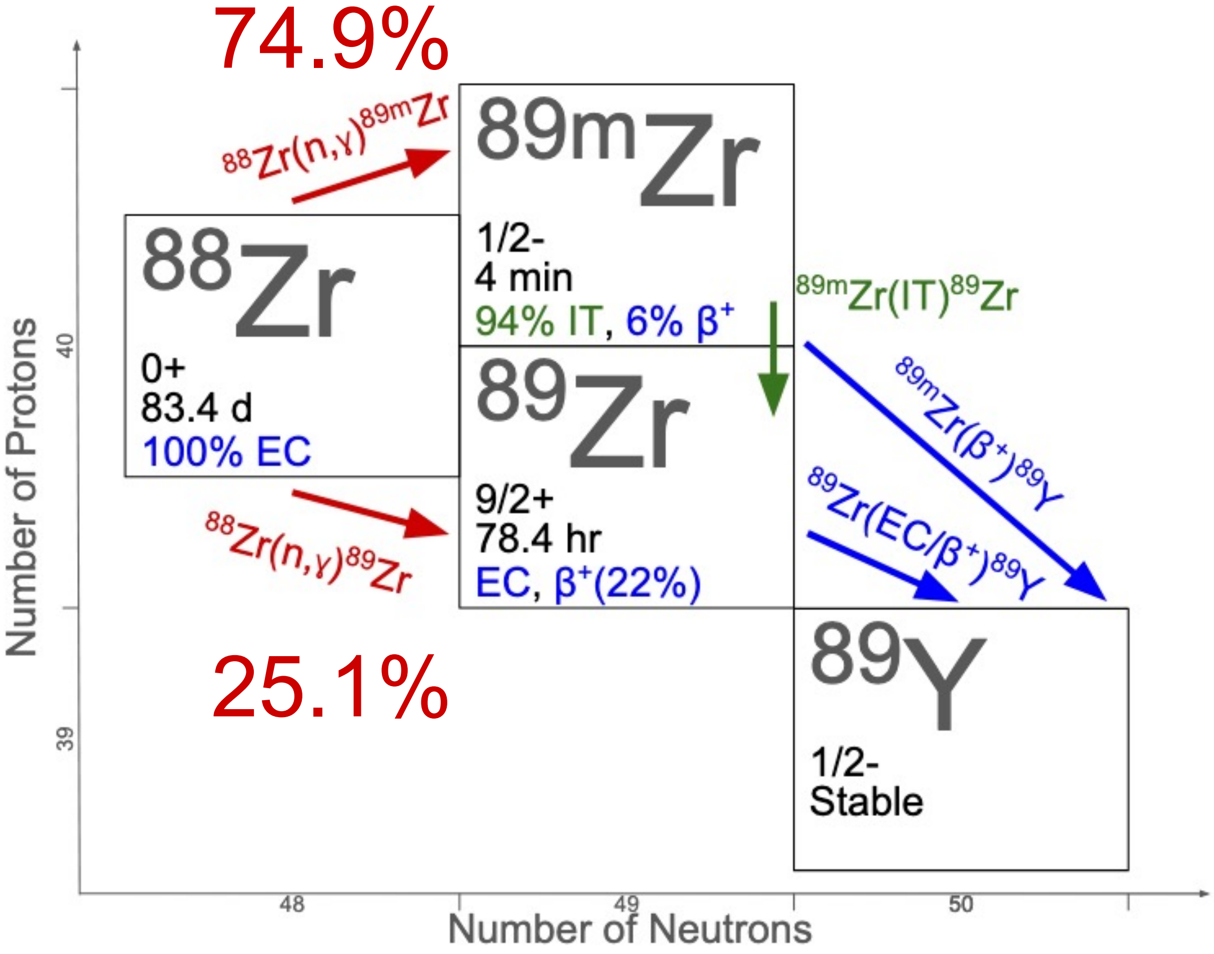


^{89m}Zr is populated 74.9±0.6% of the time
Published in Phys. Rev. C last month

Relevance to Other Measurements

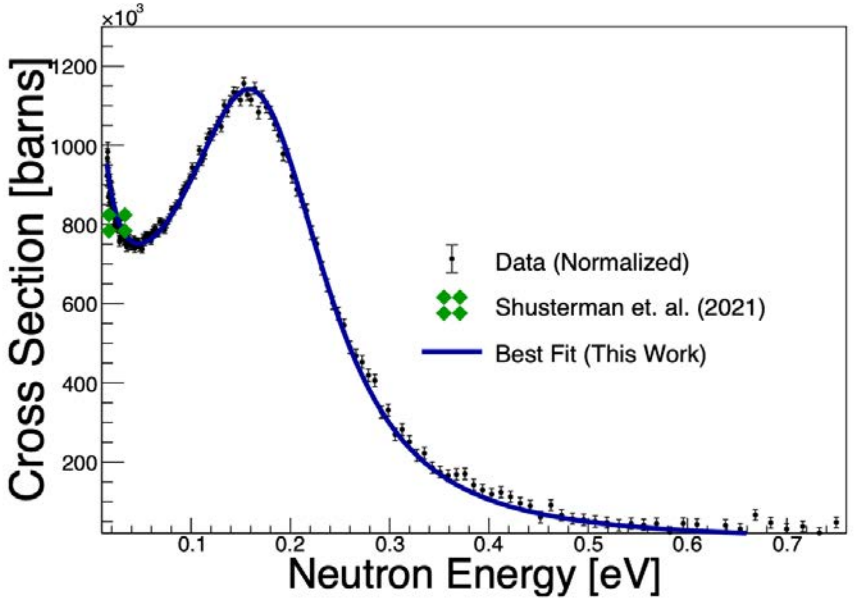
- Our isomeric yield ratio: **74.9±0.6%**.
- TALYS with ^{89}Zr energy levels given by RIPL-3, predicts **89%**.
- 2019 discovery paper (Shusterman et al) assumed ^{88}Zr(n,γ) *always* produced ^{89m}Zr
- And they measured the ^{88}Zr(n,γ) cross section by accounting for the amount of ^{88}Zr destroyed and ^{89g}Zr produced.
  - They assumed 6% of ^{88}Zr(n,γ) neutron captures didn't produce ^{89g}Zr due to ^{89m}Zr's 6% β^+ decay to ^{89}Y
  - But we show that 25.1% of ^{88}Zr(n,γ) produces ^{89g}Zr directly
  - And only 74.9% of ^{88}Zr(n,γ) produces ^{89m}Zr
    - So only 6% × 74.9% = 4.5% of reactions create ^{89}Y instead of ^{89g}Zr
- 1.5% difference in amount of ^{89}Y predicted.

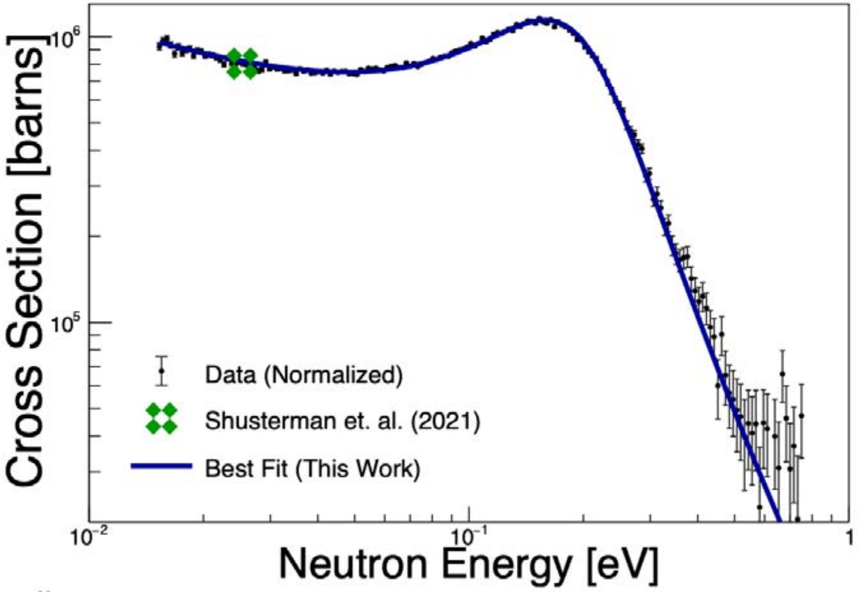


^{88}Zr – CERN n_TOF

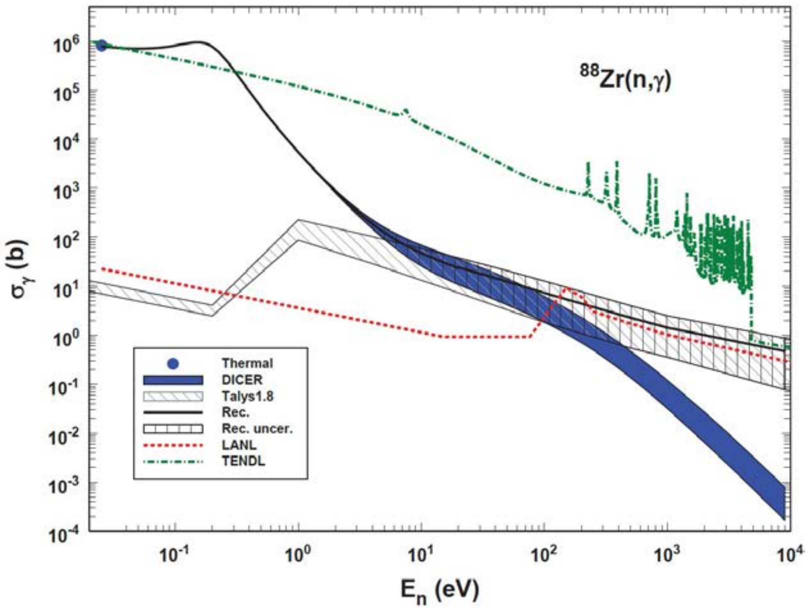
- **We confirm the resonance suggested by LANL DICER (transmission) at 0.17 eV!**
  - **Single Level Breit Wigner fit parameters within 1σ**
- But our normalization is difficult…
  - <u>**We don't know our gamma cascades (and efficiencies of gamma energy cuts)**</u>
- **Normalize based on LLNL thermal measurement**

Capture (CERN n_TOF)



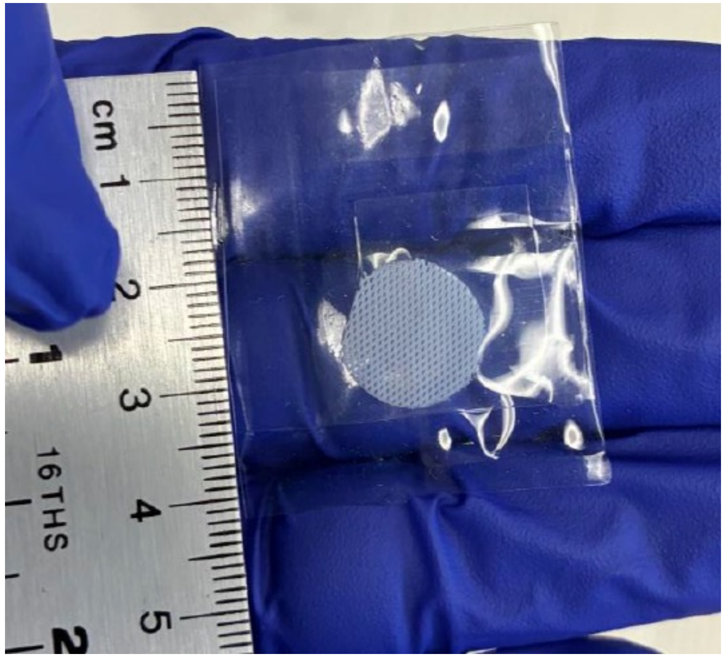


Transmission (LANL DICER)



^{88}Zr – ILL FIPPS

- Next step - **prompt gamma cascades at ILL FIPPS** (thermal, 10^8 neutrons/s/cm^2, 64 HPGe)
  - FIPPS has successful record with radioactive samples (2MBq calcium-41 and 2GBq nickel-63, for example)
  - **August – October 2025 campaign**

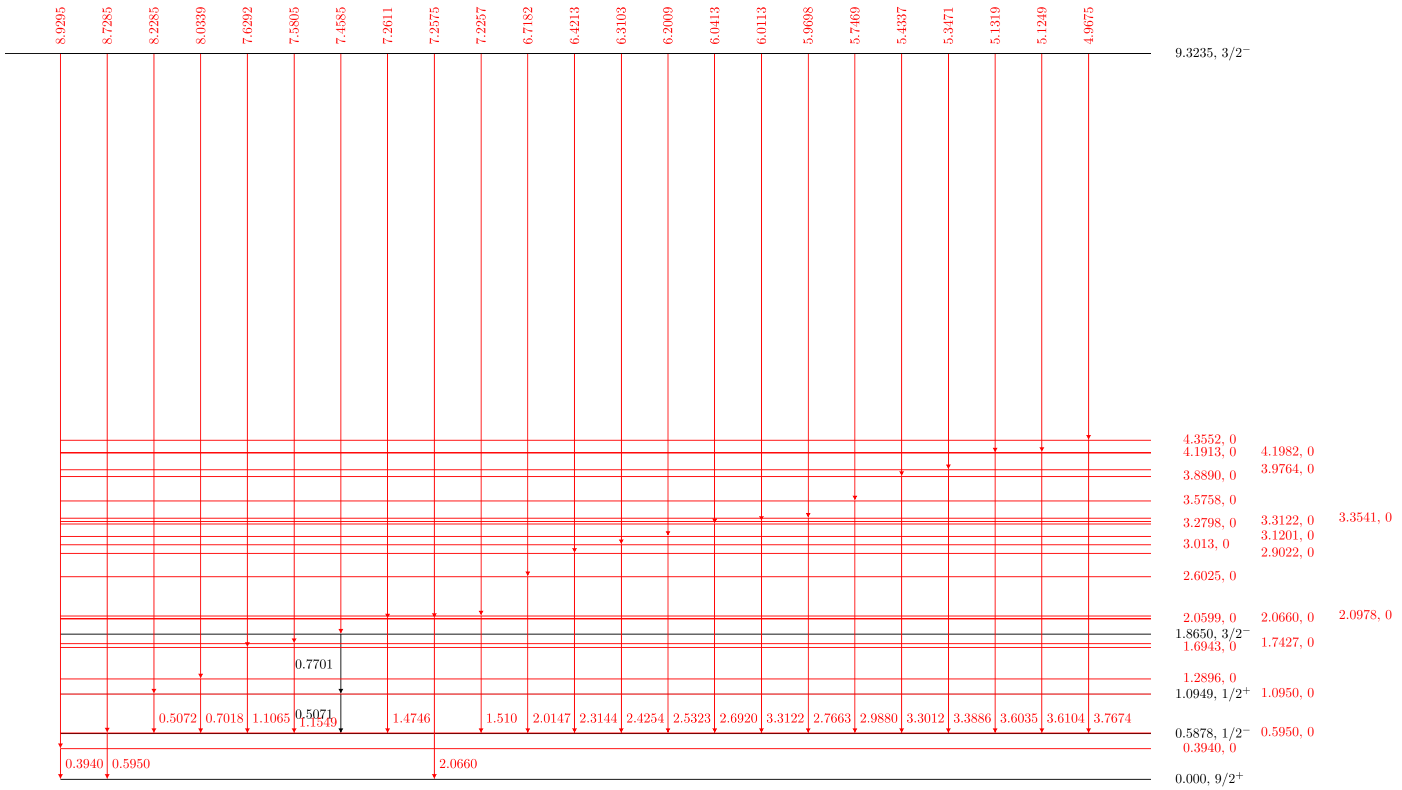


^{88}Zr – ILL FIPPS

WHAT STARTS HERE CHANGES THE WORLD

- Promising results so far!
  - 23 primaries are measured for the first time and 21 of the 26 nuclear states are measured for the first time.
  - In level scheme, red is new

  - PRELIMINARY!!!



- Also a chance to re-measure the isomeric yield ratio

^{88}Zr – Conclusions

- **The ^{88}Zr(n,γ) thermal cross section is anomalously large because of a sub-eV resonance.**
  - We confirm the (transmission-based) DICER 0.17 eV resonance.

- CERN INTC approval to measurement was less than 1 year.
  - **Rapid progress is possible when partners like LANL, PSI, and CERN work together!**

- **And infusion of ILL FIPPS data helps to complete the story!**
  - Improved understanding of prompt gamma energy deposited in scintillators

  - **Relative → Absolute cross section at n_TOF**

A Tale of Two Isotopes: ^{88}Zr and ^{135}Xe

^{135}Xe

- Largest thermal neutron capture cross section
  - Or is it…

- Fission product
- Neutron-Rich
- Large neutron absorption cross section discovered in 1944
- Radioactive (9.14 hours)

Two interesting opportunities

- ^{135}Xe has a 15-minute metastable state (^{135m}Xe)
- ^{135m}Xe cross section is predicted to be **4x** ^{135}Xe cross section
- Since molten salt reactors actively extract xenon (~15m), **^{135m}Xe (15m) will be a more sizable fraction of ^{135}Xe (9hr)**.

- Is the ^{135m}Xe cross section is truly 10.1 Mb?

- Prompt gammas not known
  - Important to gamma heating within reactors

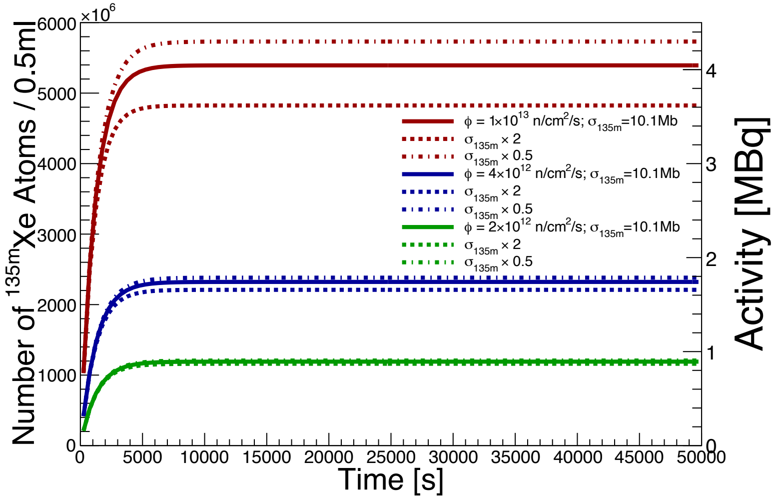
^{135m}Xe scientific merit

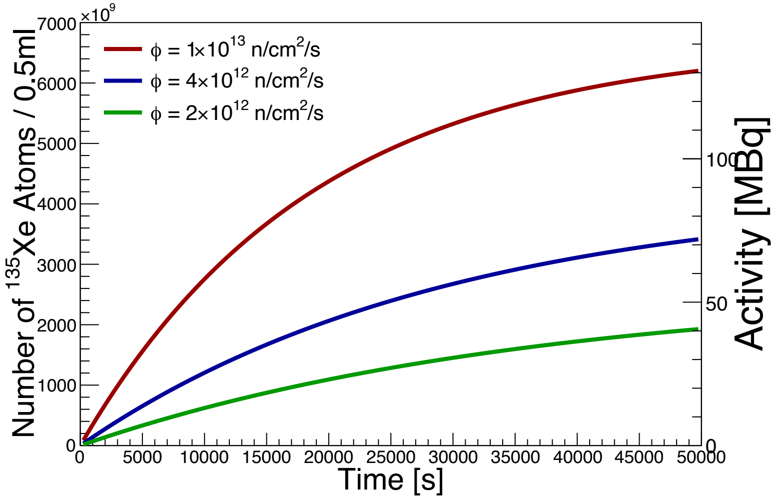
- Two interesting papers on ^{135m}Xe in nuclear engineering
  - J. Kim and Y. Kim, **Impacts of Xe-135m on Xenon Reactivity in Thermal Reactors**, Transactions of the Korean Nuclear Society conference proceeding (2013)
  - M. Eades, et. al. **The influence of Xe-135m on steady-state xenon worth in thermal molten salt reactors**, Progress in Nuclear Energy **93** 397-405 (2016)

- Since MSRs will do xenon stripping, the 15-minute Xe-135m steady state ratio to 9-hour Xe-135 will be almost two orders of magnitude larger than current reactors.

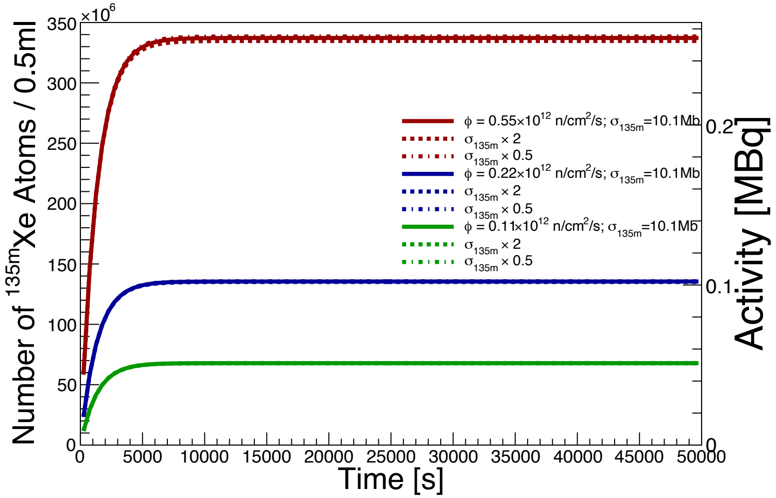
$^{135m}Xe(n,\gamma)$ cross section

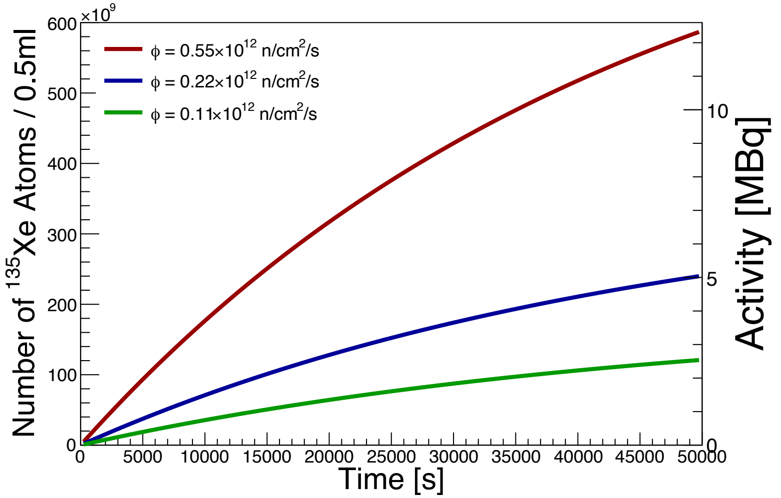
At full power, the transient equilibrium concentration of ^{135m}Xe varies by ~10% for factors of two variation in $^{135m}Xe(n,\gamma)$ cross section at CT, but less so at 3EL and RSR.





At 50kW, the concentration of ^{135m}Xe is not impacted by $^{135m}Xe(n,\gamma)$ cross section. Rather, the 15-minute half-life dominates.



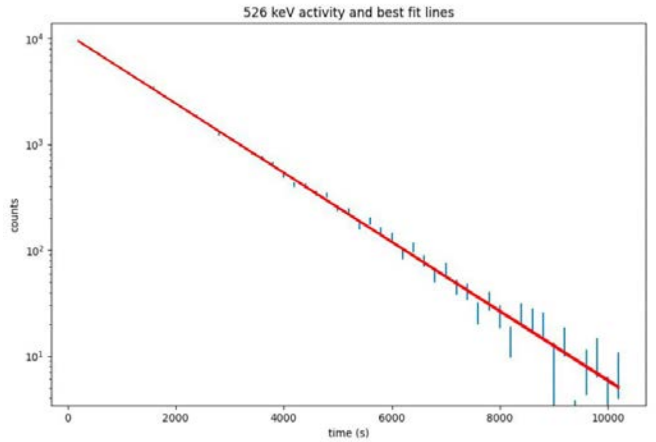


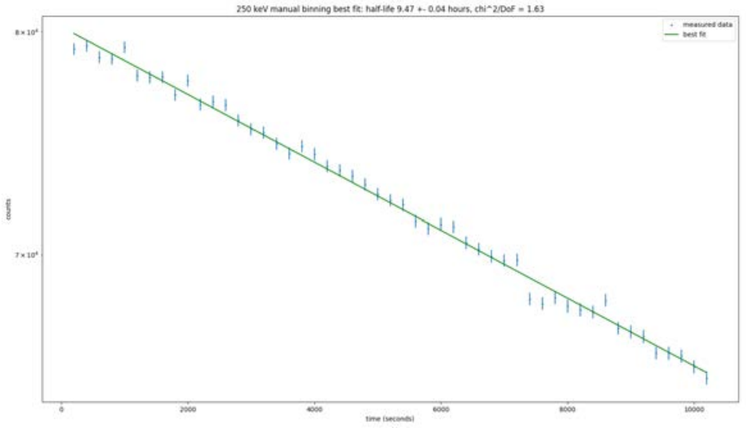
- But you need to know how often $^{134}Xe(n,\gamma)$ makes ^{135g}Xe vs ^{135m}Xe! … (isomeric yield ratio not well known)

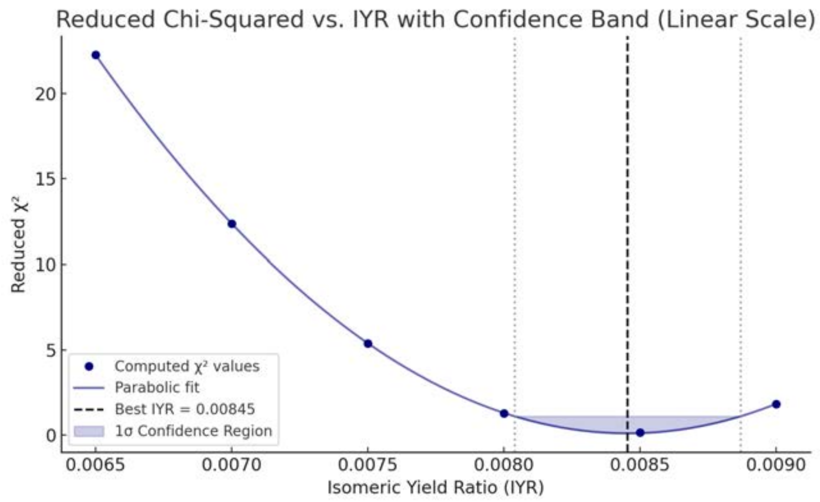
$^{134}Xe(n,\gamma)$ isomeric yield ratio

- Data taken for $^{134}Xe(n,\gamma)$ isomeric yield ratio on July 3 and 25
  - July 3 – NETL 3EL – 90s at 100kW – 15mL in PFA, transferred to fresh PTFE cylinder
    - >60k observed 526 $^{135m}Xe(IT)^{135g}Xe$ gammas
    - Simulated $^{152/154}Eu$ gas to account for large volume
  - July 25 – NETL CT – 1hr at 100kW – 1mL in quartz, counted in fresh quartz ampoule
    - >40k observed 526 $^{135m}Xe(IT)^{135g}Xe$ gammas

  - This data should be sufficient except that we have a hardware issue with spectral line instability
    - HPGe lines 'broaden' in a way that affects counting rate.
    - First noticed when checking half-lives:
      - Us: ^{135m}Xe $t_{1/2}$: 15.35±0.045m (stat), ENDF: 15.29±0.05m (Good!)
      - Us: ^{135g}Xe $t_{1/2}$: 9.47±0.05h, ENDF: 9.14±.05h (Bad!)
  - We are investigating…



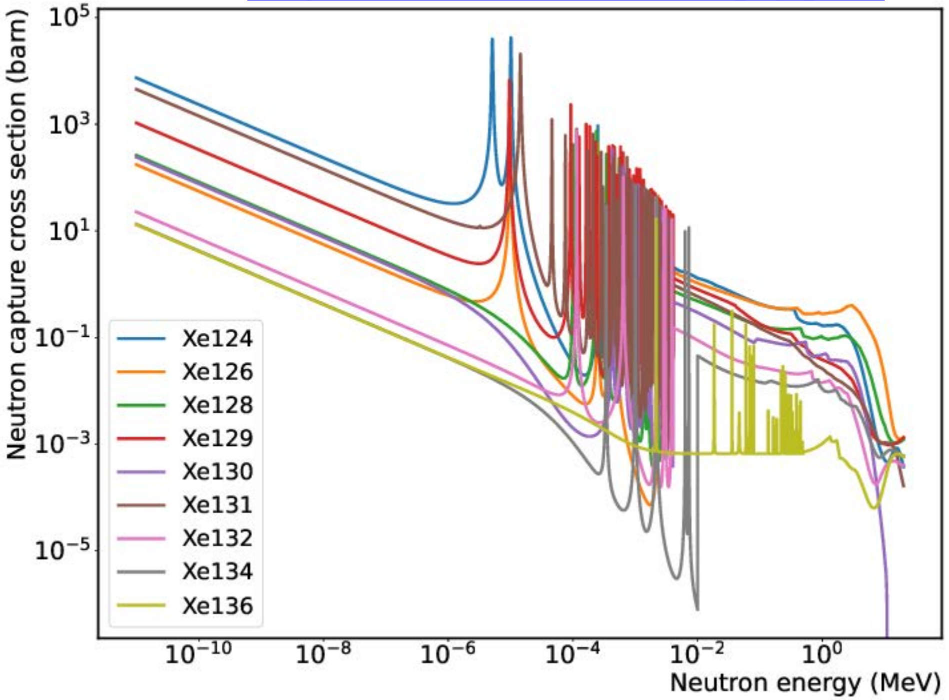




^{134}Xe Prompt Gamma scientific merit

TEXAS
The University of Texas at Austin
WHAT STARTS HERE CHANGES THE WORLD

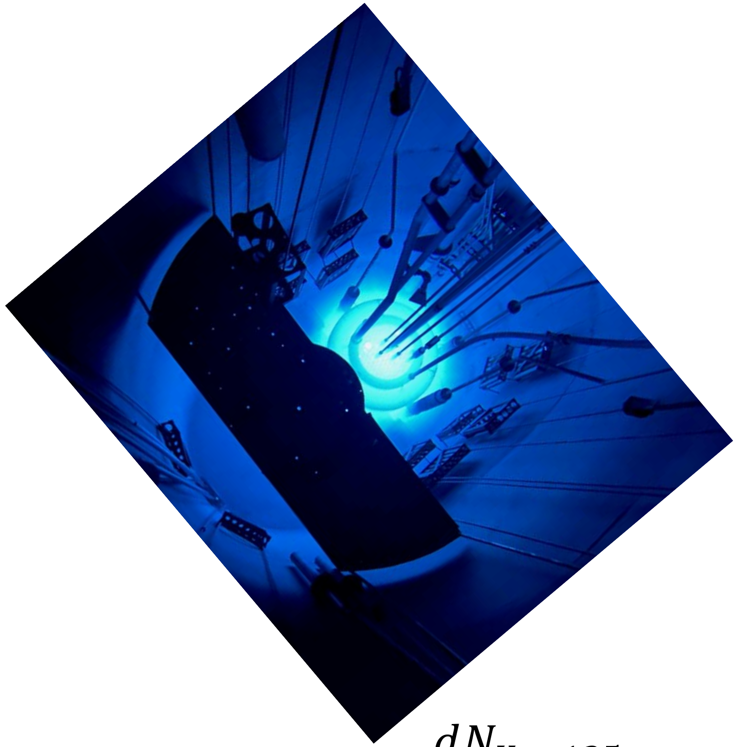
- Despite being stable, the prompt gammas of ^{126}Xe, ^{132}Xe and ^{134}Xe are not published.
  - For instance, IAEA PGAA database: https://www-nds.iaea.org/pgaa/

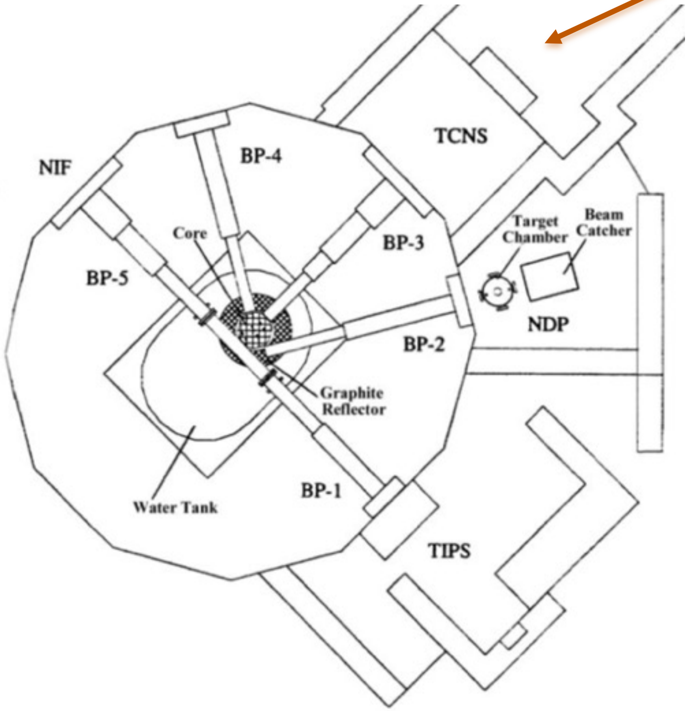


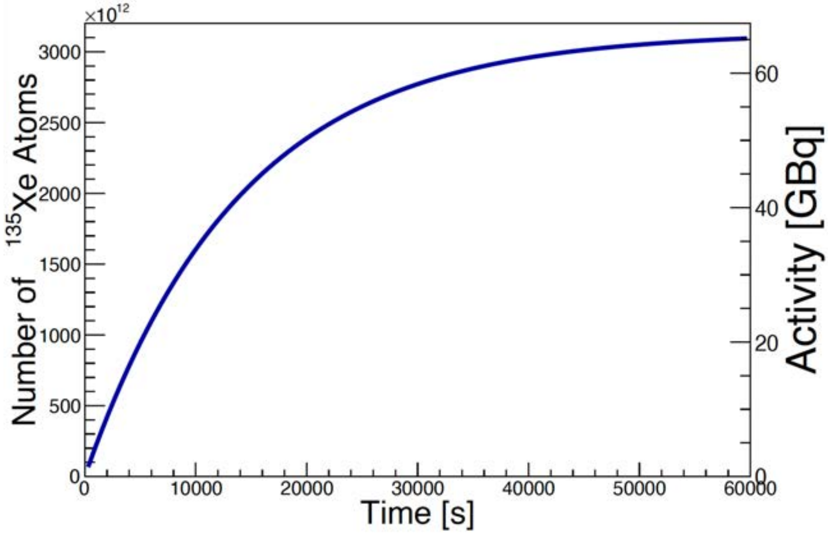
- Furthermore, the ^{135}Xe prompt gammas are not known despite this reaction dominating neutronics
- A.J. Koning and D. Rochman, Data Sheets, 113 2841–2934 (2012)
  - '**It is difficult to calculate the amount of gamma heating in an experiment**, even though the required accuracy is only 10%. This is because **photon production has never received the same sort of attention** as neutron reactions typically have: fission product decay photons are not part of neutron transport data in JEFF-3.1.2, JENDL-4.0 or ENDF/B-VII.1, and there are photon production data for almost none of the fission products (**even for the most notorious neutron capturing isotopes, ^{135}Xe**, ^{149}Sm, or ^{151}Sm, there is no photon production).' quote from Section VI. D, "TENDL Application for Heating Calculation"

^{135}Xe Prompt Gammas - ^{134}Xe

- Produce ^{135}Xe through in-core irradiation of ^{134}Xe then move to Texas Cold Neutron Source / "Beam Port 3" during same day
  - 28K/0.0024eV, 2×10^7 neutrons/s/cm^2 equivalent, 2 HPGe
    - ^{135}Xe(n,γ) reaction rate to ^{134}Xe(n,γ) is 0.6 and falling…







$$\frac{dN_{Xe-135}}{dt} = +\phi\sigma_{Xe-134(n,\gamma)}N_{Xe-134} - \left[\lambda_{Xe-135} + \phi\sigma_{Xe-135(n,\gamma)}\right]N_{Xe-135} = 0$$

$$\therefore \frac{N_{Xe-135}}{N_{Xe-134}} = \frac{\phi\sigma_{Xe-134(n,\gamma)}}{\lambda_{Xe-135} + \phi\sigma_{Xe-135(n,\gamma)}} \rightarrow \frac{N_{Xe-135}\cdot\sigma_{Xe-135(n,\gamma)}}{N_{Xe-134}\cdot\sigma_{Xe-134(n,\gamma)}} = \frac{\phi}{\frac{\lambda_{Xe-135}}{\sigma_{Xe-135(n,\gamma)}} + \phi}$$

Xenon Prompt Gammas

Neutron Capture Reaction	Target Lifetime	Cross Section [b]	Known Prompt Gammas*	Range of Known Prompt Gammas*	Capture Q-value [keV]	2026 Campaign Priority
$^{124}Xe(n,\gamma)^{125}Xe$	**1.1×10^{22}y**	**150.2**	**2**	**223.7-335.46**	**7603.3**	**2**
$^{125}Xe(n,\gamma)^{126}Xe$	16.87h		0		10018.3	
$^{126}Xe(n,\gamma)^{127}Xe$	**stable**	**3.487**	**0**		**7246**	**4**
$^{127}Xe(n,\gamma)^{128}Xe$	36.342d		0		9611	
$^{128}Xe(n,\gamma)^{129}Xe$	**stable**	**5.192**	**5**	**278.56-403.1**	**6906.854**	**3**
$^{129}Xe(n,\gamma)^{130}Xe$	stable	21.00	59	470.09-9255.21	9255.722	5
$^{130}Xe(n,\gamma)^{131}Xe$	**stable**	**4.778**	**13** → **37**	**295.30-723.29** → **80.185-6524.2**	**6604.419**	**3**
$^{131}Xe(n,\gamma)^{132}Xe$	stable	90.03	72	183.32-6467.09	8936.718	5
$^{132}Xe(n,\gamma)^{133}Xe$	**stable**	**0.4506**	**0**		**6435.9**	**4**
$^{133}Xe(n,\gamma)^{134}Xe$	5.2474d	190.0	0		8553.6	
$^{134}Xe(n,\gamma)^{135}Xe$	**stable**	**0.2649**	**0**		**6359**	**1**
$^{135}Xe(n,\gamma)^{136}Xe$	9.14h	2665000	0		8087	
$^{136}Xe(n,\gamma)^{137}Xe$	2.18×10^{21}y	0.2607	83	268.34-3424.49	4025.56	5

Radioactive

Previously Studied

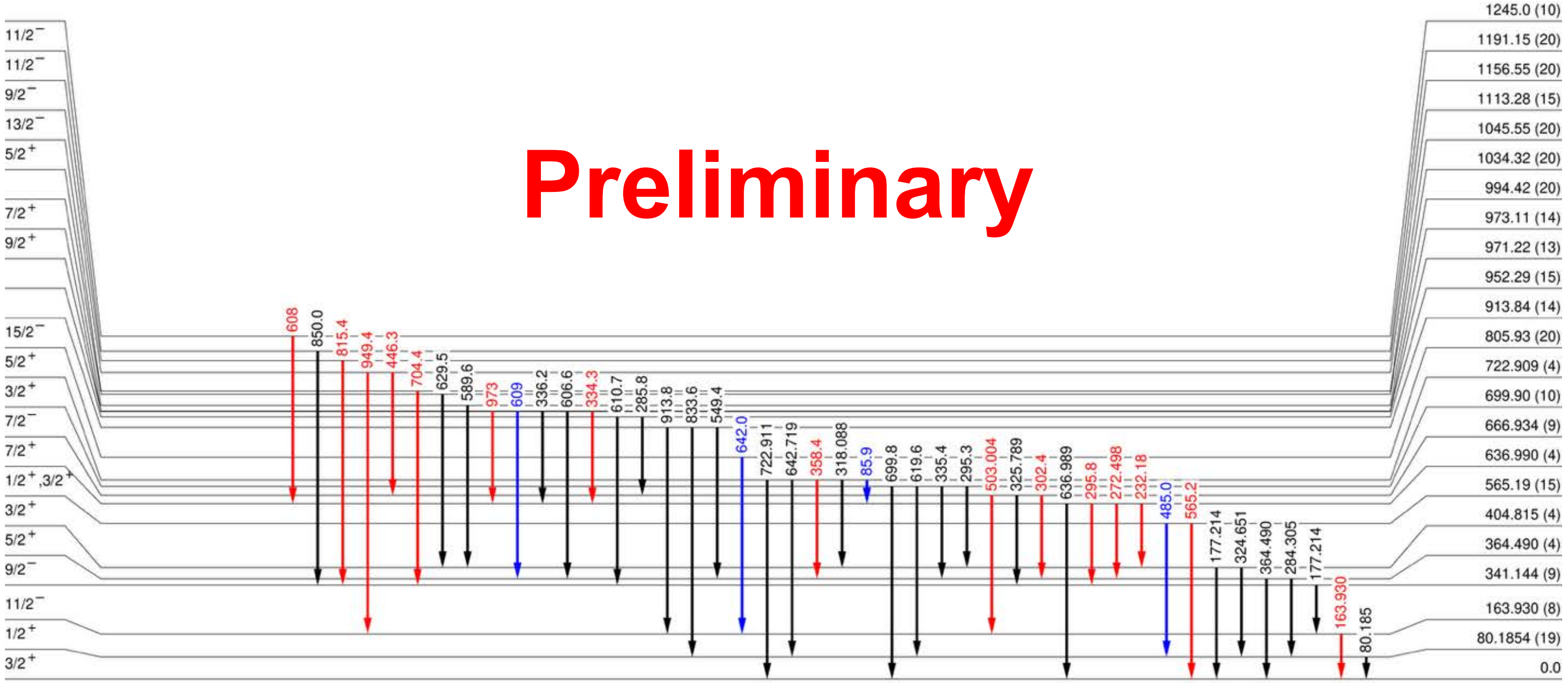
FIPPS 2025 Preliminary Data

FIPPS 2026 Campaign

*Outside $^{129/131/136}Xe(n,\gamma)$, previously known prompt gammas rely on $^{nat}XeF_2$ data from Budapest, interpreted by Rick Firestone (LBNL)

$^{130}Xe(n,\gamma)^{131}Xe$ Preliminary Data

- **Of 13 prompt gammas in IAEA database, we confirm 12**. One line has interferences.
- **Discovered 7 primary gamma rays** (those radiating from the separation energy) **in coincidence with known low-lying states**.
- **Evidence for 5 new ^{131}Xe candidate nuclear states**
- **Measure 17 gamma rays from low-lying states which had not previously been measured via neutron capture** but had been measured in reactions such as $^{131}I(\beta-)^{131}Xe$ and $^{131}Cs(\varepsilon)^{131}Xe$



- A level scheme of all known nuclear states of ^{131}Xe **below 1250 keV** with **black denoting gamma rays seen in our preliminary $^{130}Xe(n,\gamma)^{131}Xe$ FIPPS data**, **blue denoting possible interreferences**, and **red denoting lines which are not clearly present**. **This level scheme does not include five new candidate states** from our preliminary data.

^{135}Xe Ongoing Progress

- To Summarize:

- ^{135m}Xe irradiated in-core, counted within 30 minutes at UT NETL.
  - More than 100k ^{135m}Xe(IT)^{135g}Xe observed in July.

  - Preparing for measurement of ^{135m}Xe cross section via ^{135m}Xe burnup at high power (50 kW vs 1 MW)
    - Test 10.1 Mbarn ^{135m}Xe(n,γ) cross section
  - But first finalizing ^{134}Xe(n,γ) isomeric yield ratio…
    - Resolving issue with line broadening during data collection.

- We are beginning a campaign of xenon prompt gammas
  - At FIPPS this summer!!
  - Ultimate goal is ^{135}Xe(n,γ)
    - Gamma heating, most common neutron capture in many nuclear applications
  - ^{134}Xe(n,γ) is an intermediate step
    - Not known despite being a stable isotope
  - Verified ability to measure known isotopes such as ^{124}Xe and ^{130}Xe

Questions?

^{88}Zr

- Second largest thermal neutron capture cross section

- Not a fission product
- Neutron-Poor
- Large neutron absorption cross section discovered in 2019
- Radioactive (83.4 days)

- Recent FIPPS data:
  - Gain deeper understanding of nuclear structure
  - help CERN n_TOF analysis (gamma efficiencies)

^{135}Xe

- Largest thermal neutron capture cross section
  - Or is it… ^{135m}Xe

- Fission product
- Neutron-Rich
- Large neutron absorption cross section discovered in 1944
- Radioactive (9.14 hours)

- Huge ^{135m}Xe cross section will be important for Molten Salt Reactors. Can we verify it?

- Use FIPPS to work towards ^{135}Xe(n,γ) prompt gammas?

Many thanks to brilliant collaborators and students!

UT NETL reactor, radiochemistry – Don Nolting, Joseph Lapka, Bill Charlton, Tracy Tipping, Rodrigo Viveros

^{89m}Zr – Isaac Kelly (undergraduate → PhD student at Notre Dame), Jacob Moldenhauer

CERN n_TOF – Genevieve Alpar (undergraduate → Master at AFIT), Albert Parmenter (undergraduate), Beth Jacoby (undergraduate), Michi Bacak, Javi Balibrea, Claudia Lederer-Woods, Jorge Lerendegui-Marco, Emilio Maugeri, Alberto Mengoni

ILL FIPPS – Caterina Michelagnoli, Marek Stryjczyk and many from n_TOF including Matt Birch, Toby Wright, Fran Garcia-Infantes, Javi Balibrea, Jorge Lerendegui-Marco, Michi Bacak, Andrew Rosen (undergraduate)

^{135m}Xe – Ally Leonesio (undergraduate), Gabe Garrison (undergraduate), Grace Martin (undergraduate), Zach Boor (undergraduate)

Backup

^{88}Zr(n,γ) – Before CERN n_TOF

- 2019 LLNL – thermal neutron capture σ_T= 861,000±69,000 barns
- 2021 LLNL – thermal neutron capture σ_T = 804,000±63,000 barns and resonance integral I = 2,530,000±280,000 barns
- 2023 LANL – thermal neutron capture σ_T = 771,000±31,000 barns and resonance integral I = 15,210±670 barns
  - LANL DICER uses neutron ToF with transmission for energy-resolved measurement
  - **Resonance measured at 0.171 eV**
    - A. Stamatopoulos *et al.*, “Origin of the enormous ^{88}Zr neutron-capture cross section and quantifying its impact on applications,” Phys. Rev. Lett. **134**, 112702 (2025)

Aqueous harvesting of ^{88}Zr at a radioactive-ion-beam facility for cross-section measurements

Jennifer A. Shusterman, Nicholas D. Scielzo, E. Paige Abel, Hannah K. Clause, Nicolas D. Dronchi, Wesley D. Frey, Narek Gharibyan, Jason A. Hart, C. Shaun Loveless, Sean R. McGuinness, Logan T. Sutherlin, Keenan J. Thomas, Suzanne E. Lapi, J. David Robertson, Mark A. Stoyer, Eric B. Norman, Graham F. Peaslee, Gregory W. Severin, and Dawn A. Shaughnessy

Phys. Rev. C **103**, 024614 – Published 26 February 2021

Research Square

Preprints are preliminary reports that have not undergone peer review. They should not be considered conclusive, used to inform clinical practice, or referenced by the media as validated information.

Discovery of the origin of the enormous 88Zr neutron-capture cross section and quantifying its impact on applications

Athanasios Stamatopoulos (thanos@lanl.gov)
Los Alamos National Laboratory https://orcid.org/0000-0001-9607-1185

www.nature.com/scientificreports

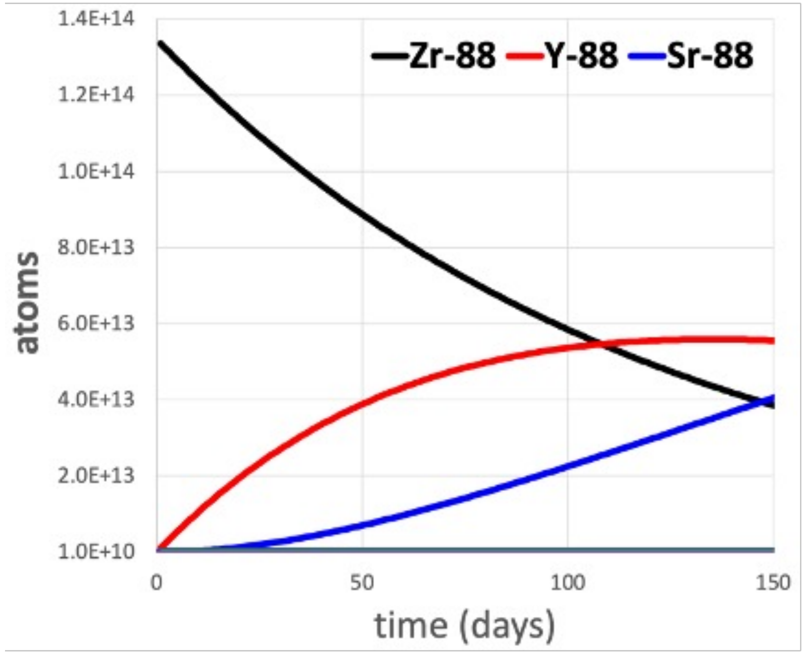
scientific reports

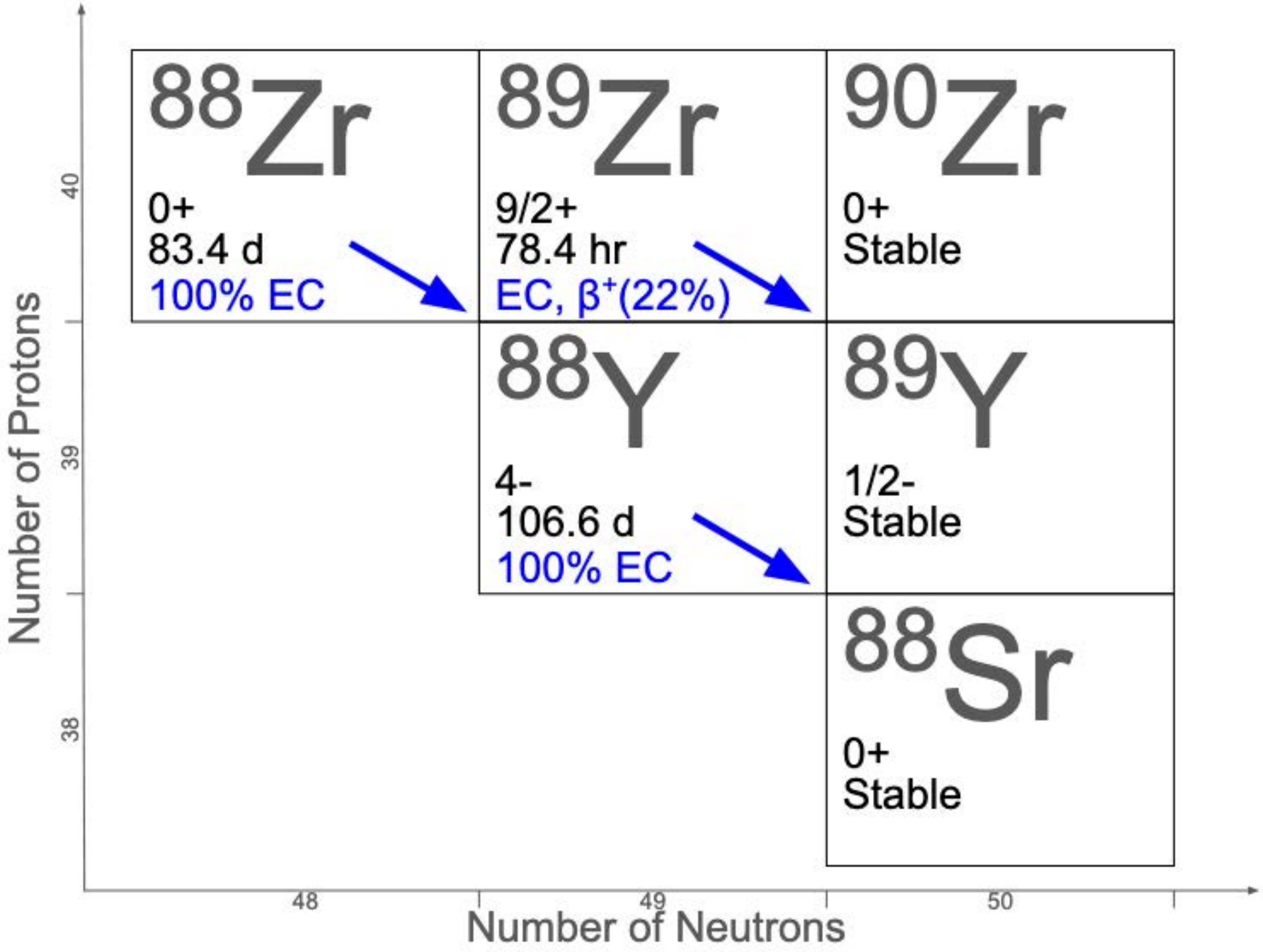
OPEN **Production of zirconium-88 via proton irradiation of metallic yttrium and preparation of target for neutron transmission measurements at DICER**

Artem V. Matyskin, Athanasios Stamatopoulos, Ellen M. O'Brien, Brad J. DiGiovine, Veronika Mocko, Michael E. Fassbender, C. Etienne Vermeulen & Paul E. Koehler

^{88}Zr(n,γ) – Measurement Challenges

- ^{88}Zr Produced from protons on yttrium at LANL IPF.
  - ^{89}Y(p,2n)^{88}Zr
- ^{88}Zr is radioactive with an 83.4 day half life.
- Decays to ^{88}Y with a 106.6 day half life





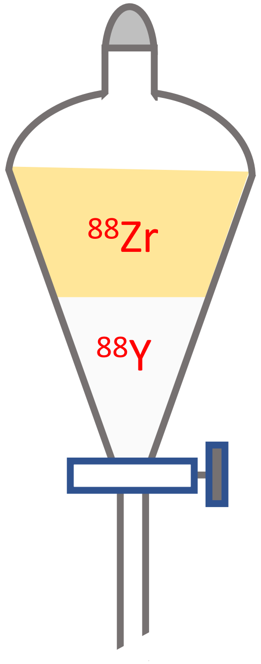
^{88}Zr(n,γ) – Measurement Challenges

WHAT STARTS HERE CHANGES THE WORLD

- **^{88}Y buildup creates a major background**
- ^{88}Zr electron capture produces **393 keV** gammas (97% branching ratio)
- But ^{88}Y electron capture produces **898 keV** (93%) and **1.836 MeV** (99%) gamma rays.

Organic layer *dodecane*
ρ=0.73 g/ml

Aqueous layer H_2O
ρ=1 g/ml

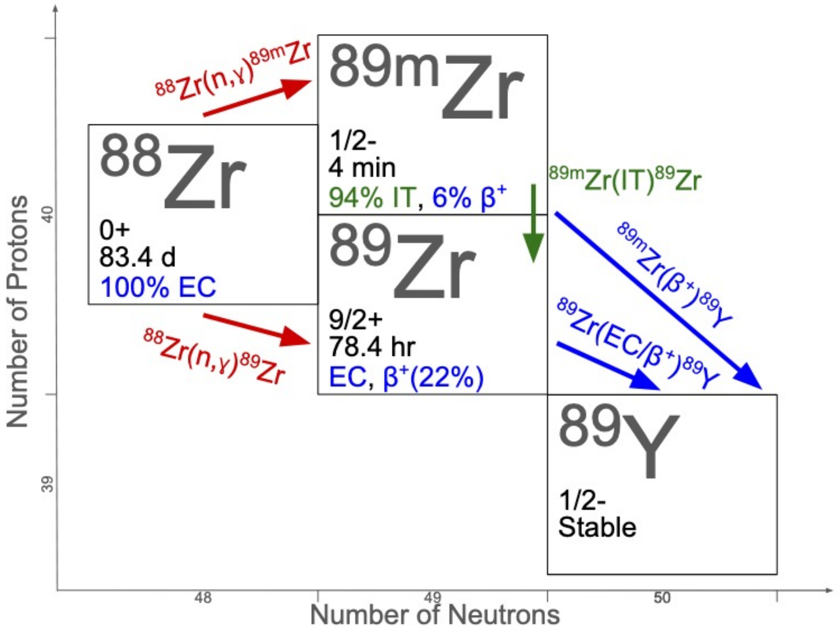


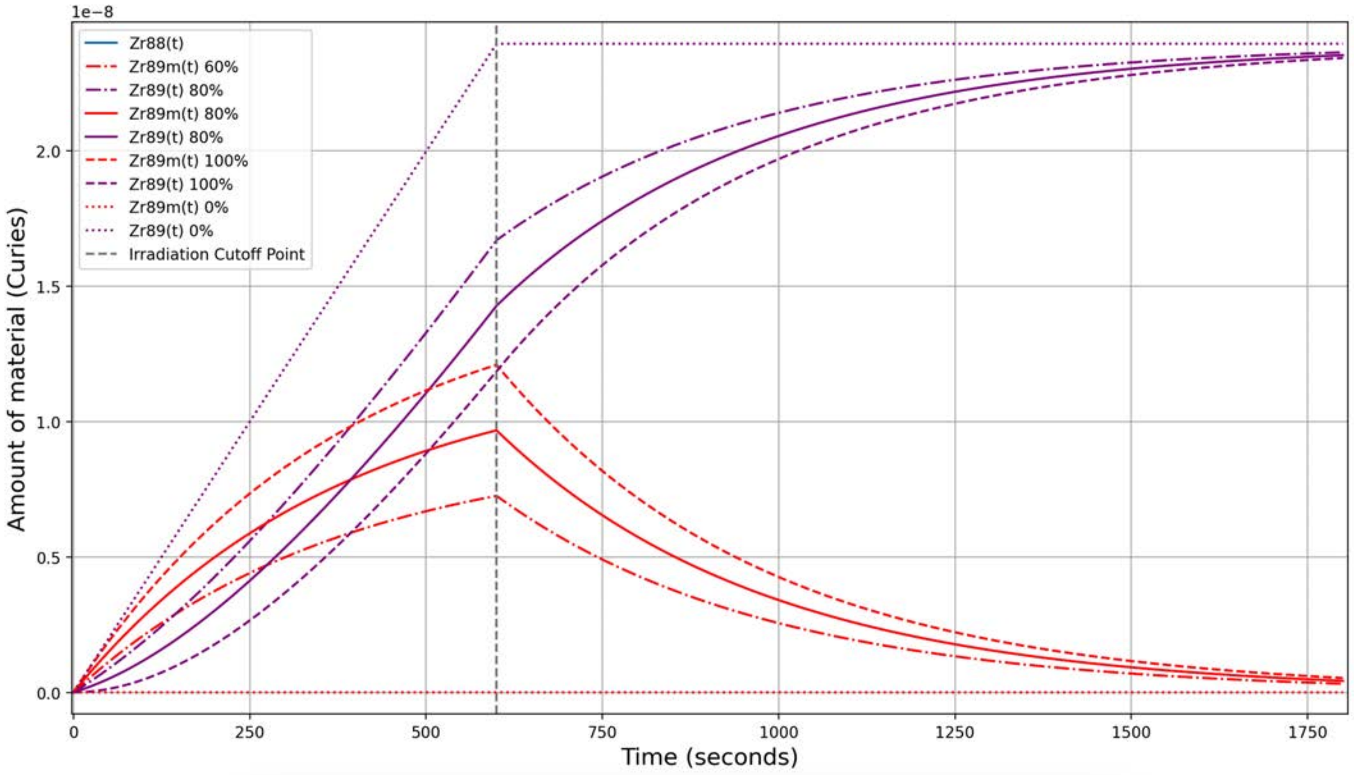
- ^{88}Zr arrives from LANL in 2M HCl
- Developed ^{88}Zr/^{88}Y chemical separation
- ^{88}Zr metal cation complexes with $(HDEHP)_2$ and dissolves in the organic layer
  - Then digest dodecane with oxalic acid, nitric acid, peroxide

$^{88}Zr(n,\gamma)$ – Isomeric Yield Ratio

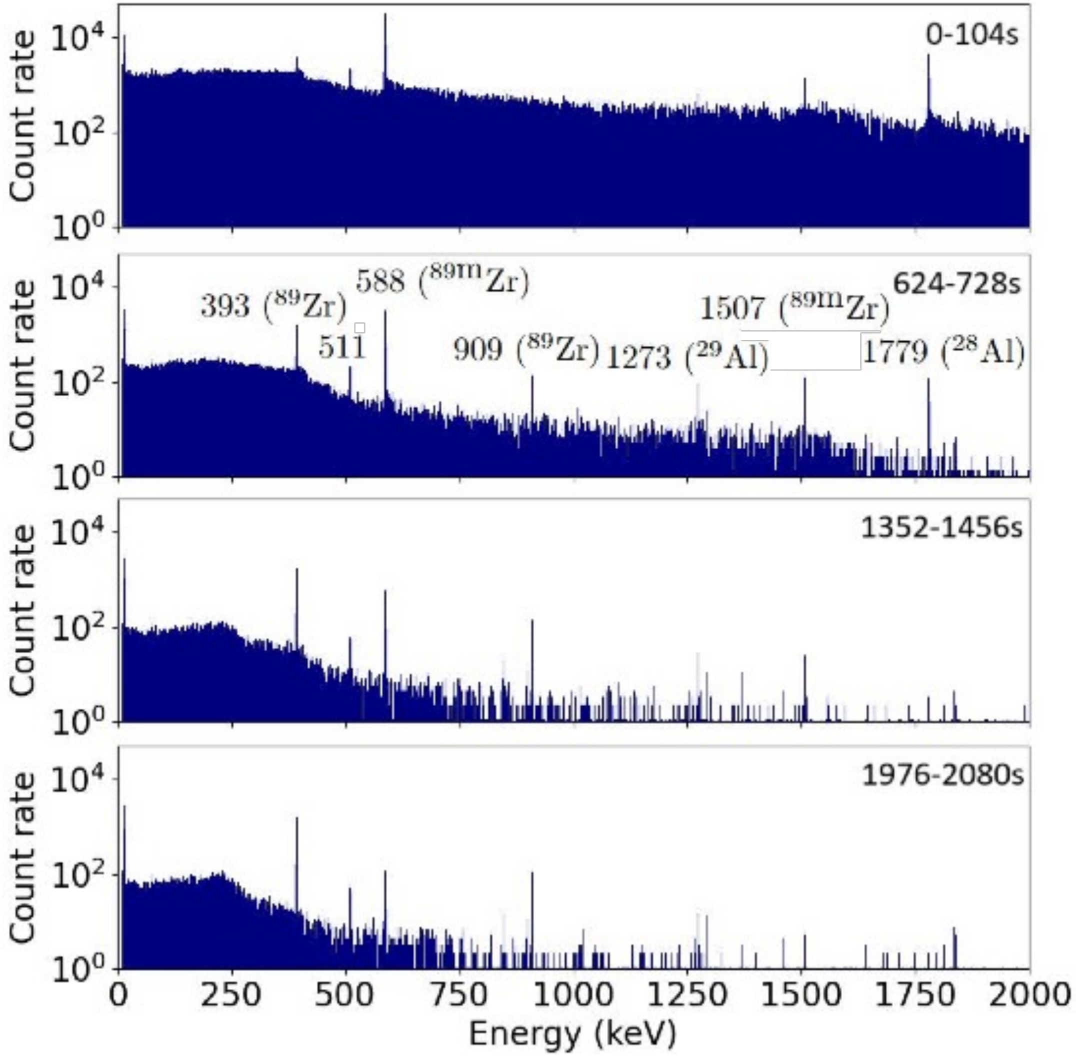
- **How often is compound nucleus "trapped" in metastable state after capture?**
  - Isomeric Yield Ratio
  - ^{89m}Zr 588 keV above ground state ^{89g}Zr
  - $^{88}Zr(n,\gamma)^{89g}Zr$ - 9.3 MeV of prompt gammas
  - $^{88}Zr(n,\gamma)^{89m}Zr$ – 8.7 MeV of prompt gammas

- 10-minute irradiation at UT NETL (TRIGA) reactor
  - Reactor core 10^{13} n/cm^2/s
- Pneumatic transfer to HPGe counting room
- Data collection begins 3 minutes after irradiation





$^{88}Zr(n,\gamma)^{89m/89g}Zr$ – HPGe Spectra



- Prominent lines for ^{89m}Zr:
  - **588 keV** (IT) and **1507 keV** (in conjunction with β^+ emission)
  - Confirmed previous measurements of ^{89m}Zr branching ratios
    - D. Van Patter and S. Shafroth, Decay of 78.4 h ^{89}Zr and 4.18 min ^{89m}Zr, Nucl. Phys. 50, 113 (1964)
- Prominent line for ^{89g}Zr:
  - 393 keV (EC to ^{89}Y)
- Other prominent lines:
  - 511 keV
  - 1273 keV (^{29}Al)
  - 1779 keV (^{28}Al)
    - ^{88}Zr evaporated on Kapton tape substrate

$^{88}Zr(n,\gamma)$ – Isomeric Yield Ratio

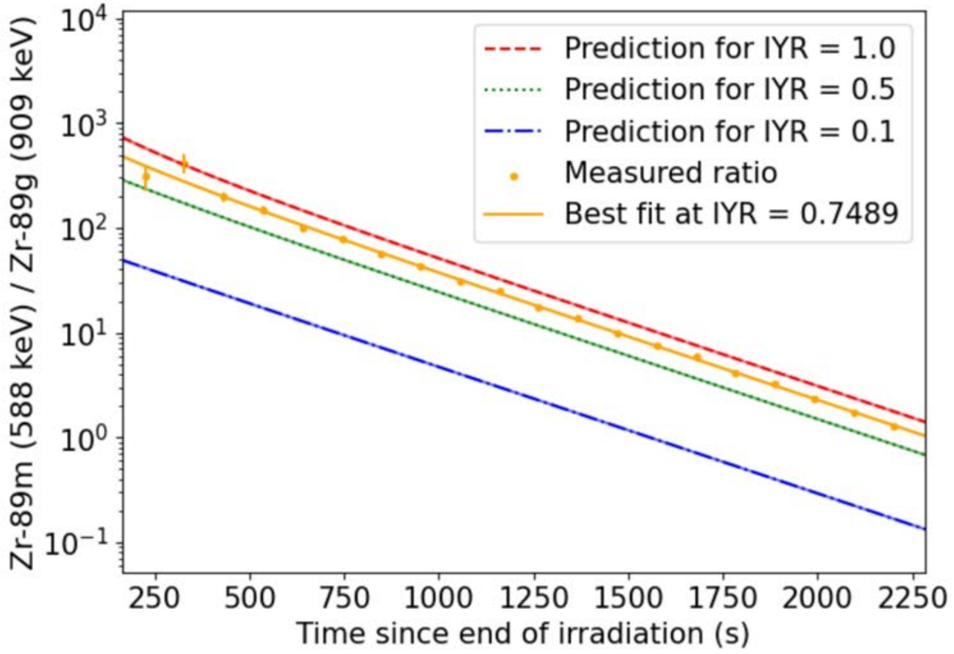
- Fit ratio of most prominent ^{89m}Zr and ^{89g}Zr lines over time.
- Shape is predominantly ^{89m}Zr 4.2min half life over (fairly constant, 78.4hr) ^{89g}Zr abundance, except that ^{89m}Zr decays also feed ^{89g}Zr.

- Vary isomeric yield ratio as well as 7 sources of systematic uncertainty
- Best fit at χ^2 minimum
  - 7 penalty terms associated with systematics
- 68% C.L. at χ^2 increase of 1

$$\chi^2 = \chi^2_{\min} + \Delta\chi^2$$

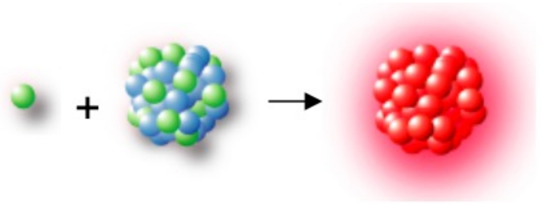
$$\chi^2 = (\chi^2)_{\text{stat}} + \sum_{n=1} \frac{\Delta\alpha_i}{\sigma_{\alpha_i}}$$

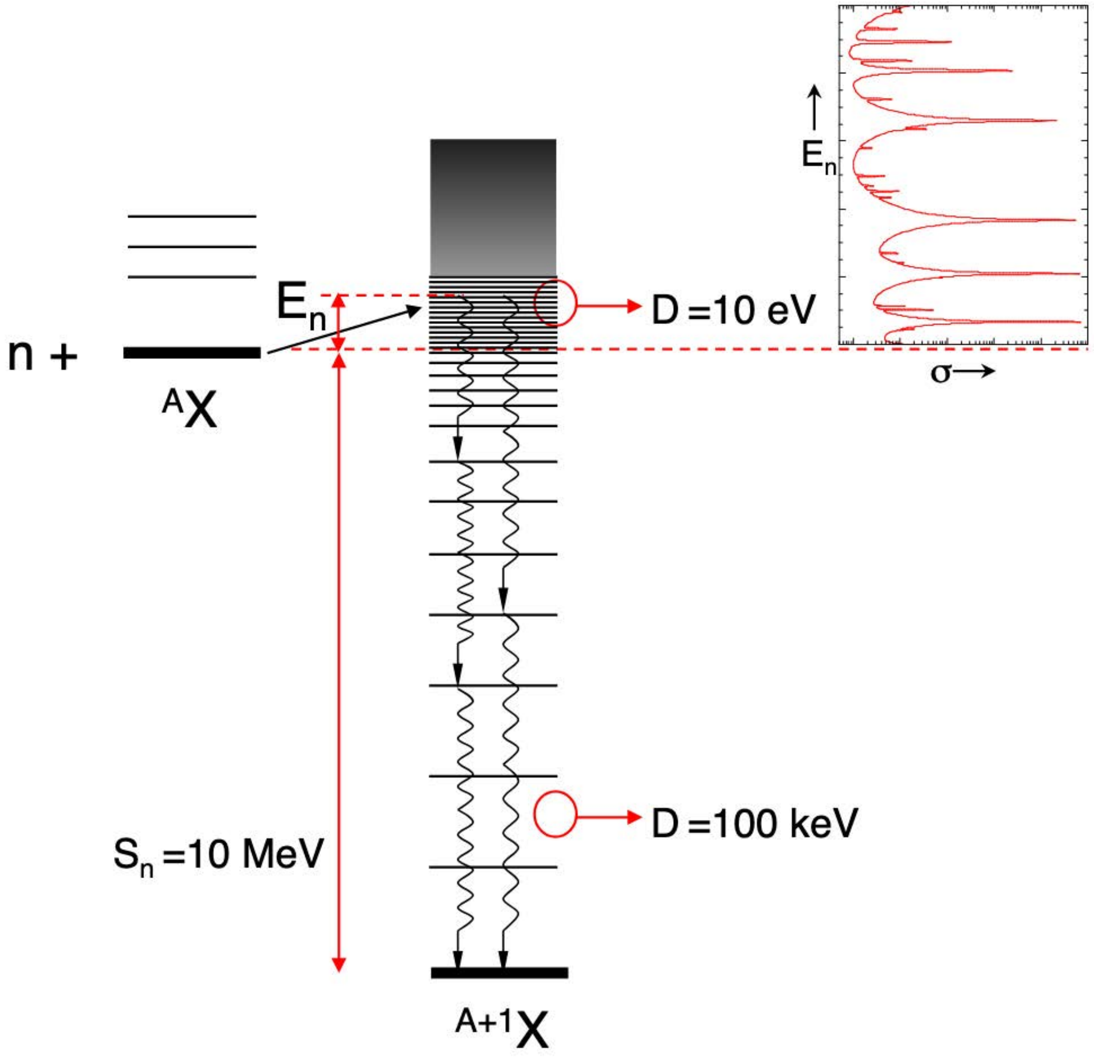
Parameter	Value	Deviation at best-fit (σ)
Cross section × flux	$3.52 \pm 0.33 \times 10^{-6}$ n/s [2]	$<10^{-3}$
^{88}Zr half-life	83.4 ± 0.3 d [14]	$<10^{-3}$
^{89}Zr half-life	78.361 ± 0.025 h [15]	$<10^{-3}$
^{89m}Zr half-life	4.161 ± 0.010 min [16]	0.33
^{89m}Y half-life	15.663 ± 0.005 s [18]	$<10^{-3}$
Rel. Eff. × ICC	1.44671 ± 0.00126 [15]	0.002
^{89m}Zr branching ratio	0.9377 ± 0.0012 [18]	$<10^{-3}$



Moving Forward - Neutron Capture

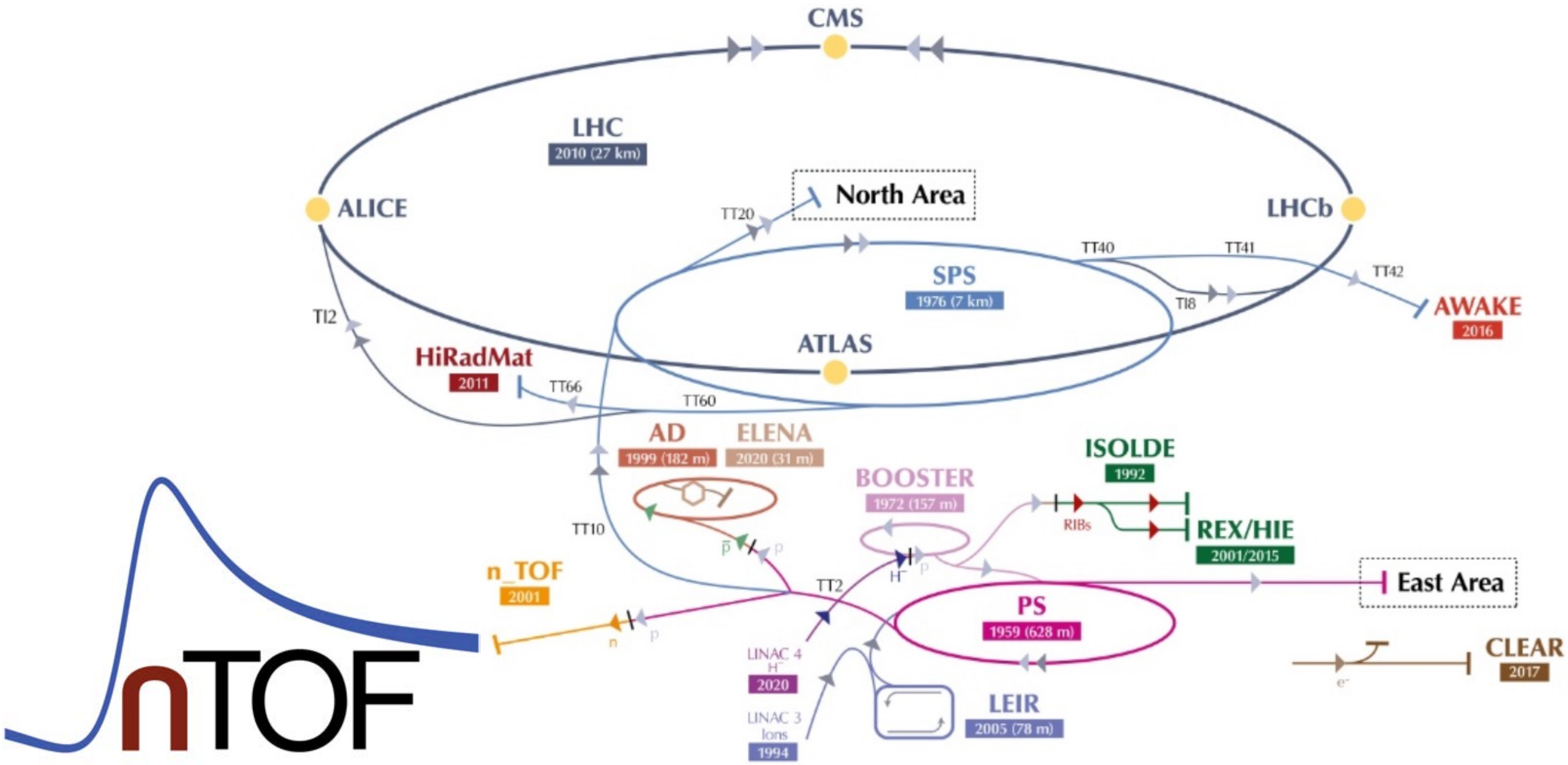
- Neutron capture creates **compound nucleus**
  - Excess energy creates **prompt gamma rays**





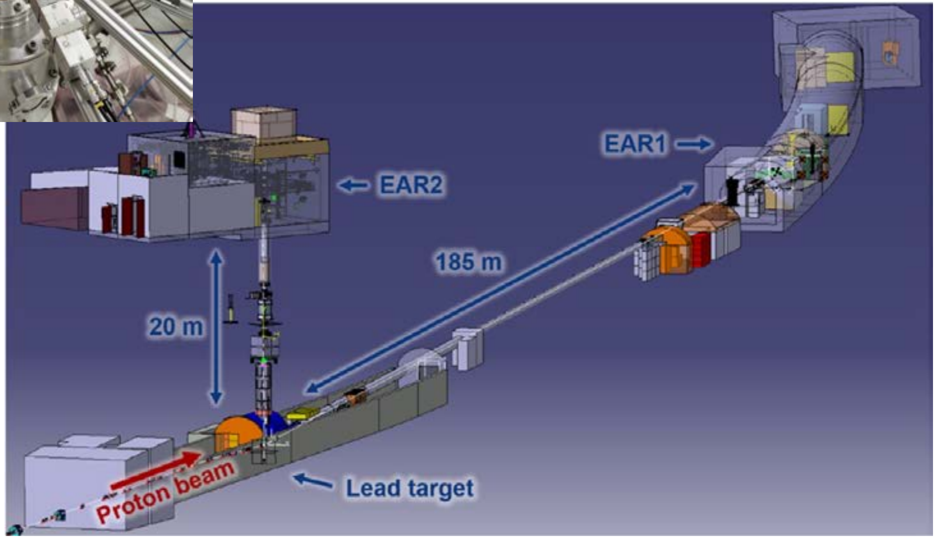
Neutron Time of Flight at CERN

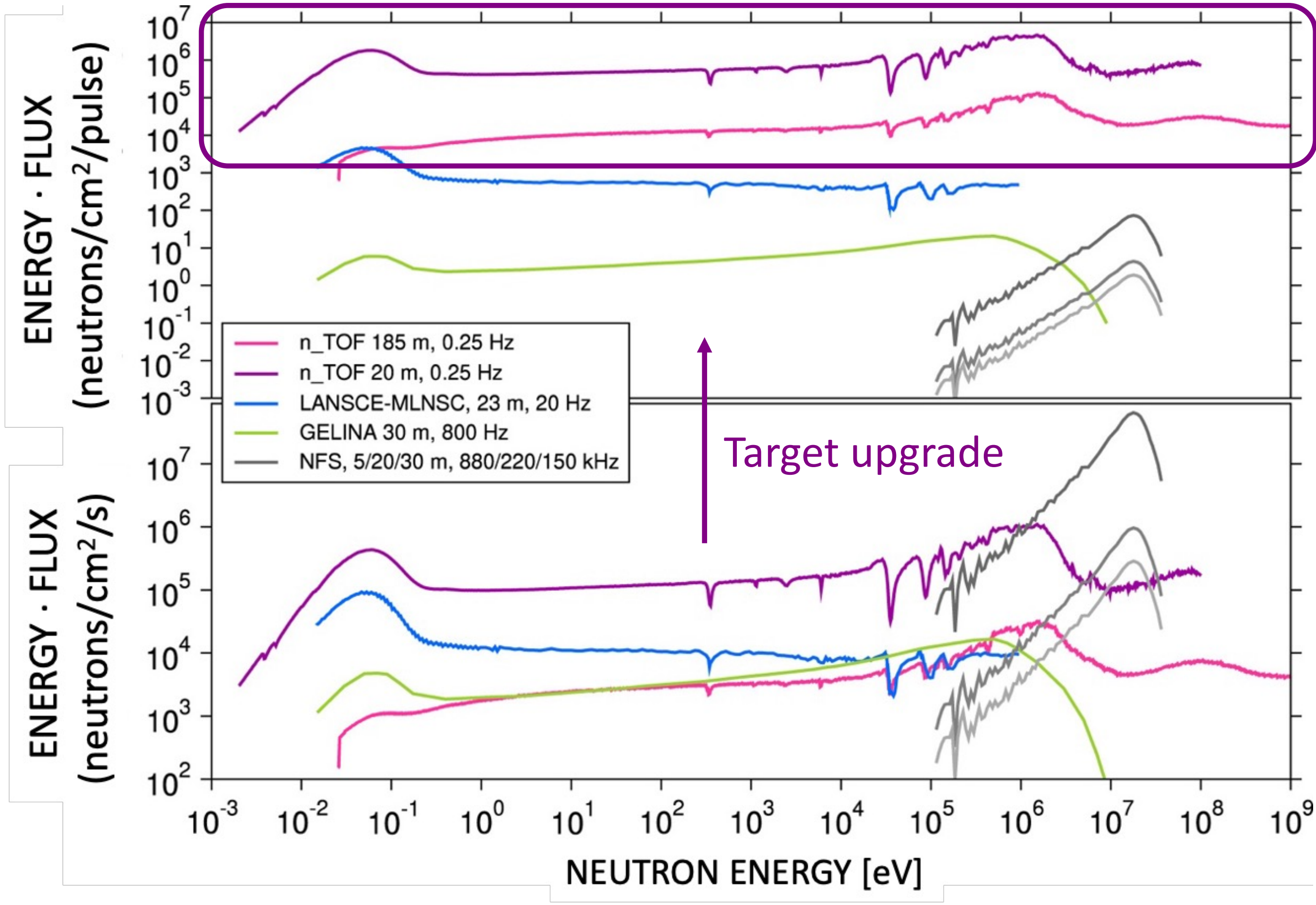
- LINAC4 H^- → 160 MeV ; Booster H^+ → 2 GeV ; Proton Synchrotron H^+ → 25 GeV
- 7ns proton bunch width, lead spallation target



Neutron Time of Flight at CERN

- Experimental Area 2 (EAR2) utilized due to **higher instantaneous flux**, **fast segmented Total Energy Detectors** (C_6D_6 sTEDs), and **“almost dead-time free” DAQ**.

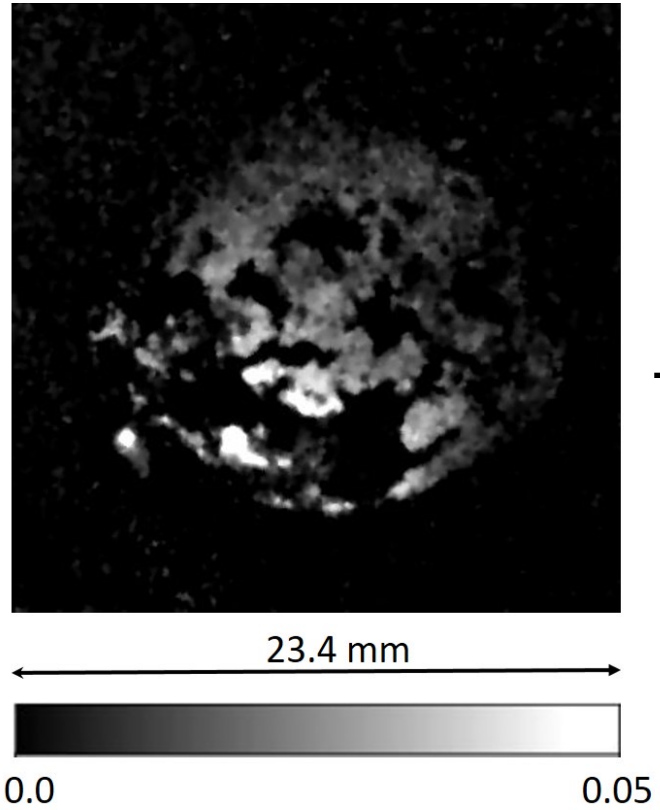


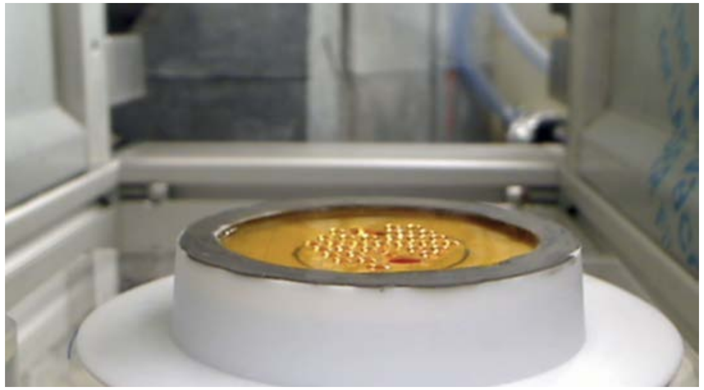


Neutron Time of Flight at CERN

- **Evaporation of salt not uniform!**
  - Beam interception factor

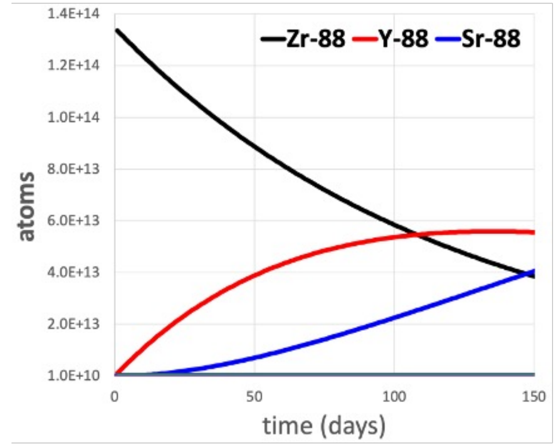
- Neutron radiograph from NEUTRA at Paul Scherrer Institute.
  - March 2024 (test) deposition
  - July 2024 image
- **Lesson learned for (main) August 2024 sample!**



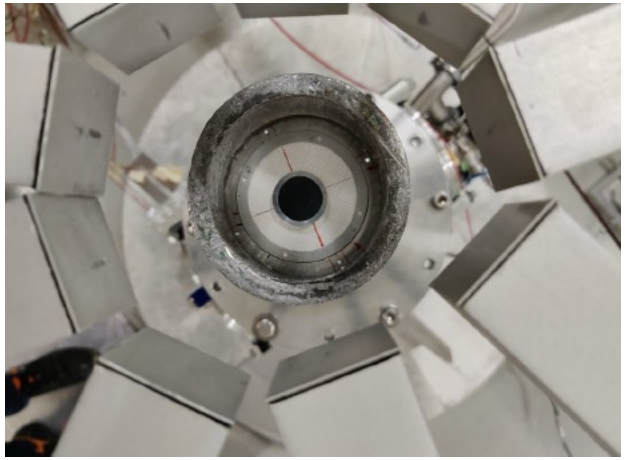


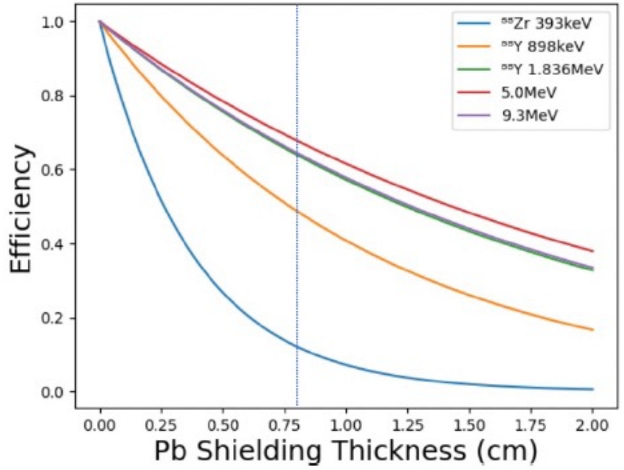
Minimize Detector Rate

- Obtained 1mCi ^{88}Zr source from LANL IPF for March **test exposure** at CERN n_TOF
  - Allowed us to test shipping logistics to Europe
- Obtained 30mCi ^{88}Zr source from LANL IPF for August-September beam time.
  - 5 AUG 24 LANL separation
  - 12 AUG 24 receipt at PSI
  - 16 AUG 24 begin irradiation at CERN



- Lead shielding to attenuate 85% of ^{88}Zr activity while maintaining roughly half of prompt gammas.

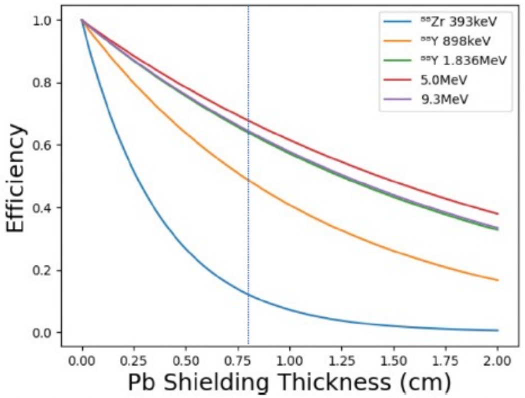


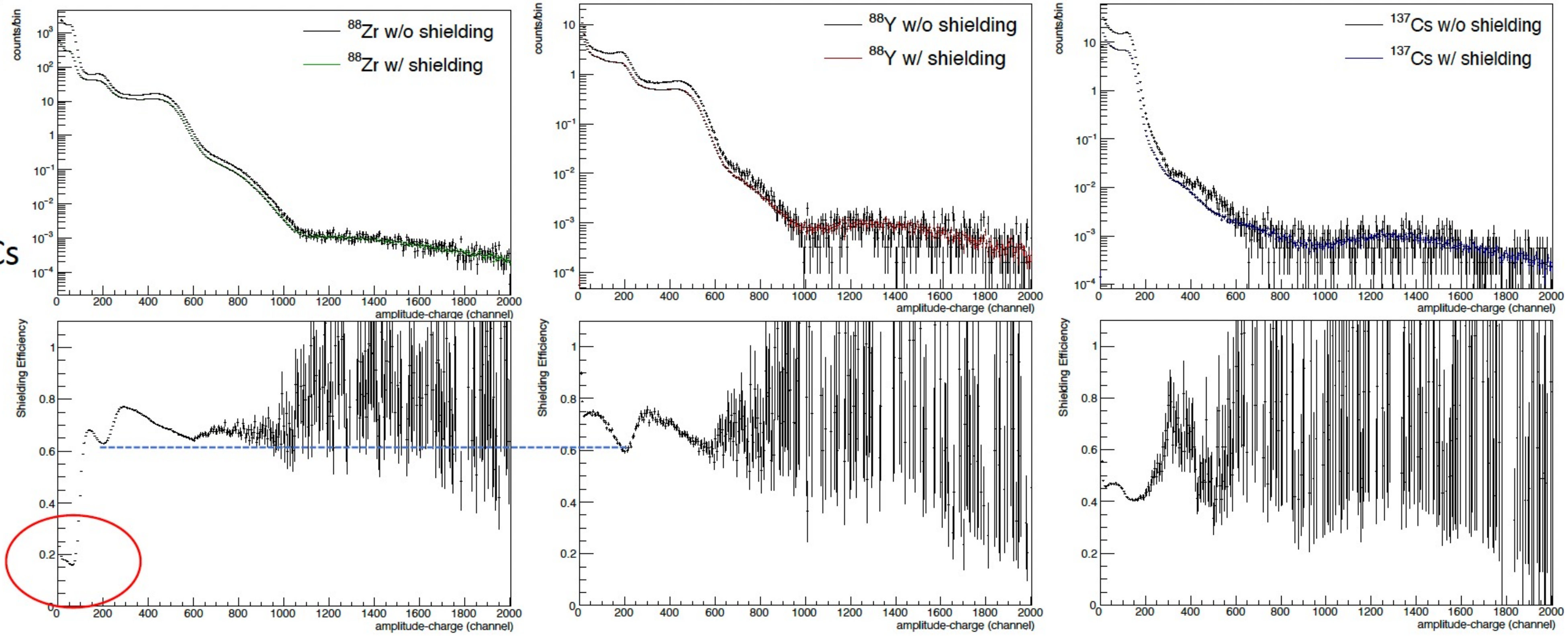


Shielding Validation (March 2024)

Shielding efficiency:

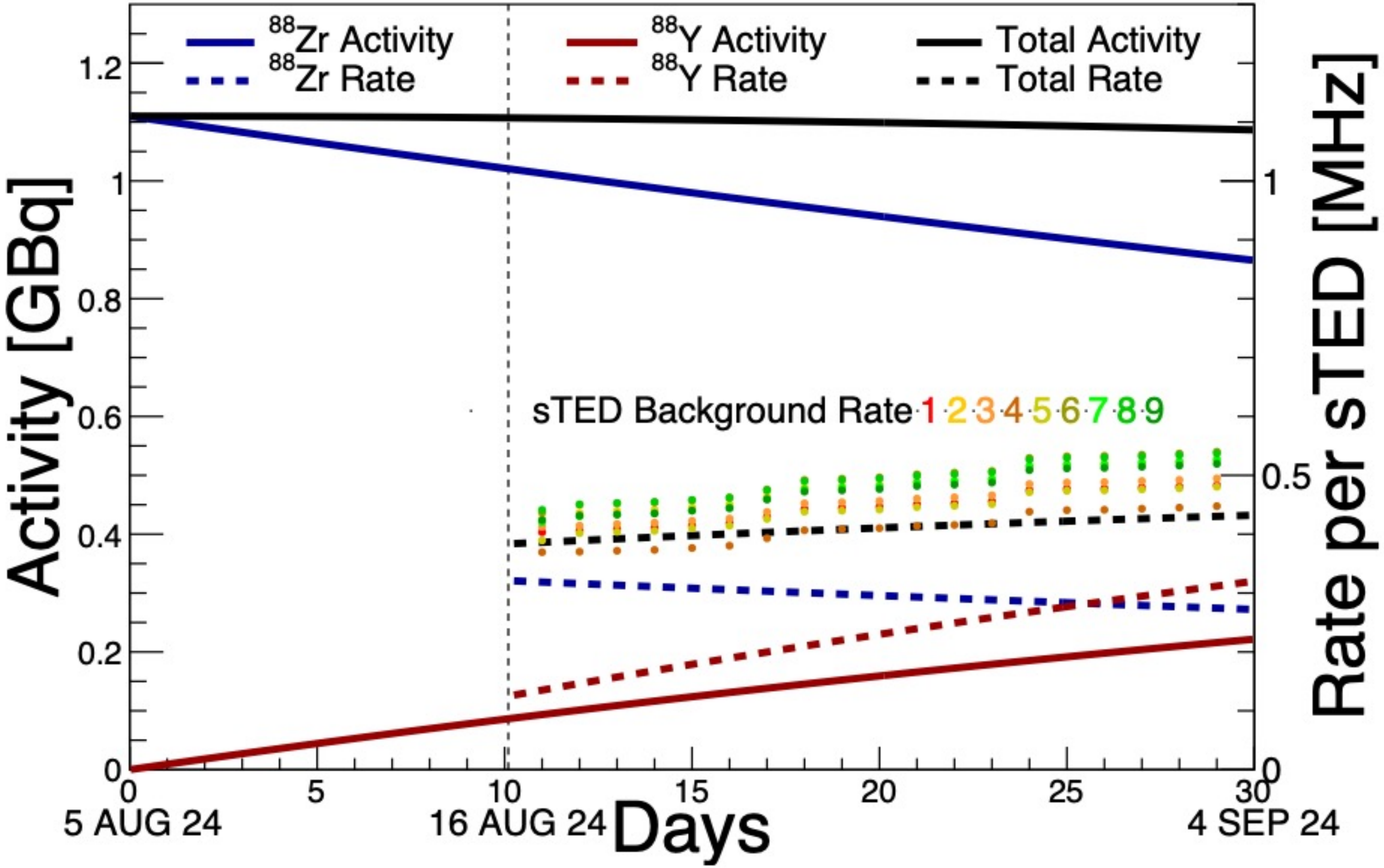
- Efficiency of 393keV ^{88}Zr 15-20%. ✓
- Efficiency of ^{88}Y contamination in ^{88}Zr sample same as ^{88}Y calibration data. ✓
- Efficiency of 662keV ^{137}Cs 40-45%. ✓
- Rates are as expected.





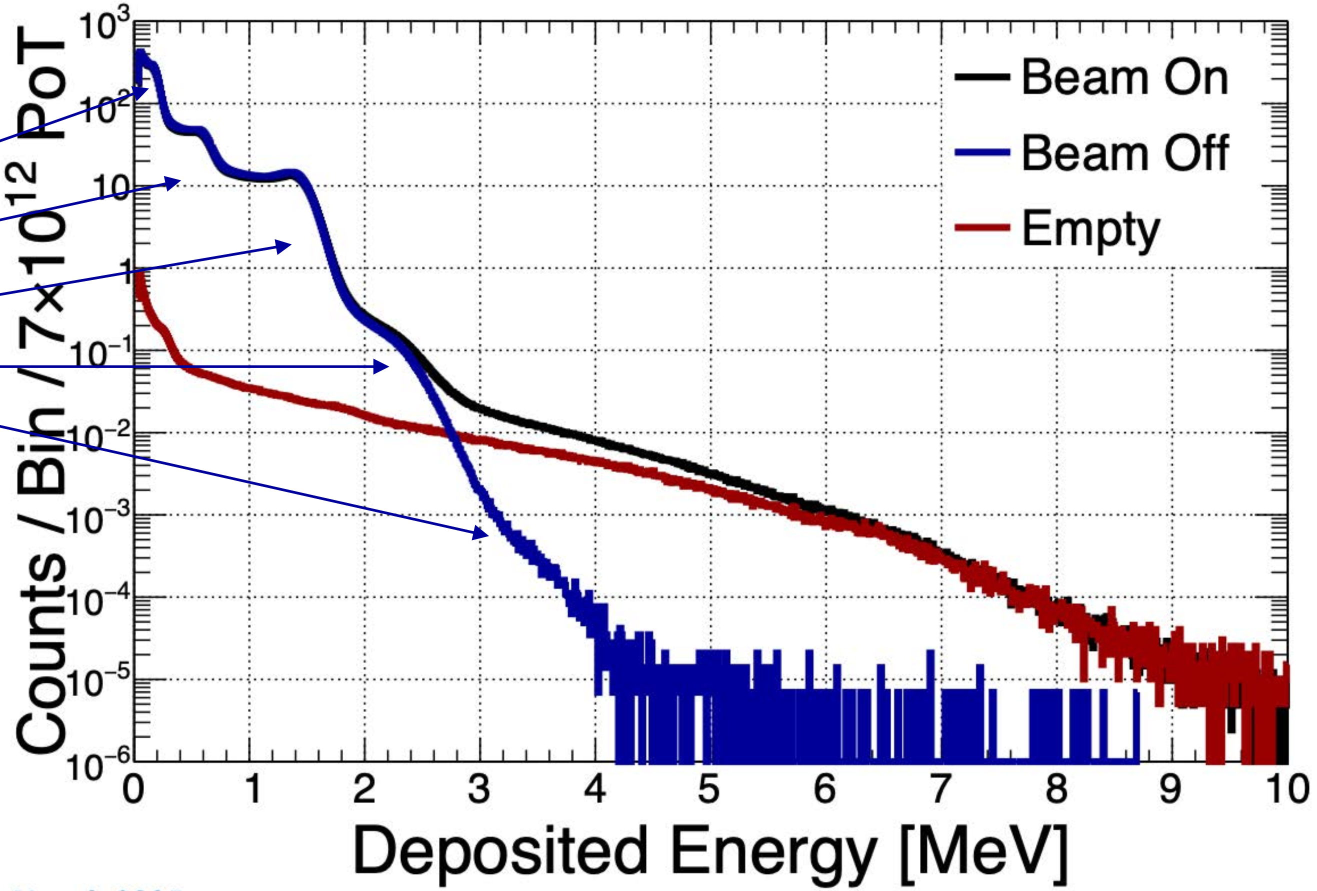
Detector Rates (August 2024)

- Maintained a detector rate below 1 MHz to assure detector performance
  - Continuous calibrations with ^{88}Y



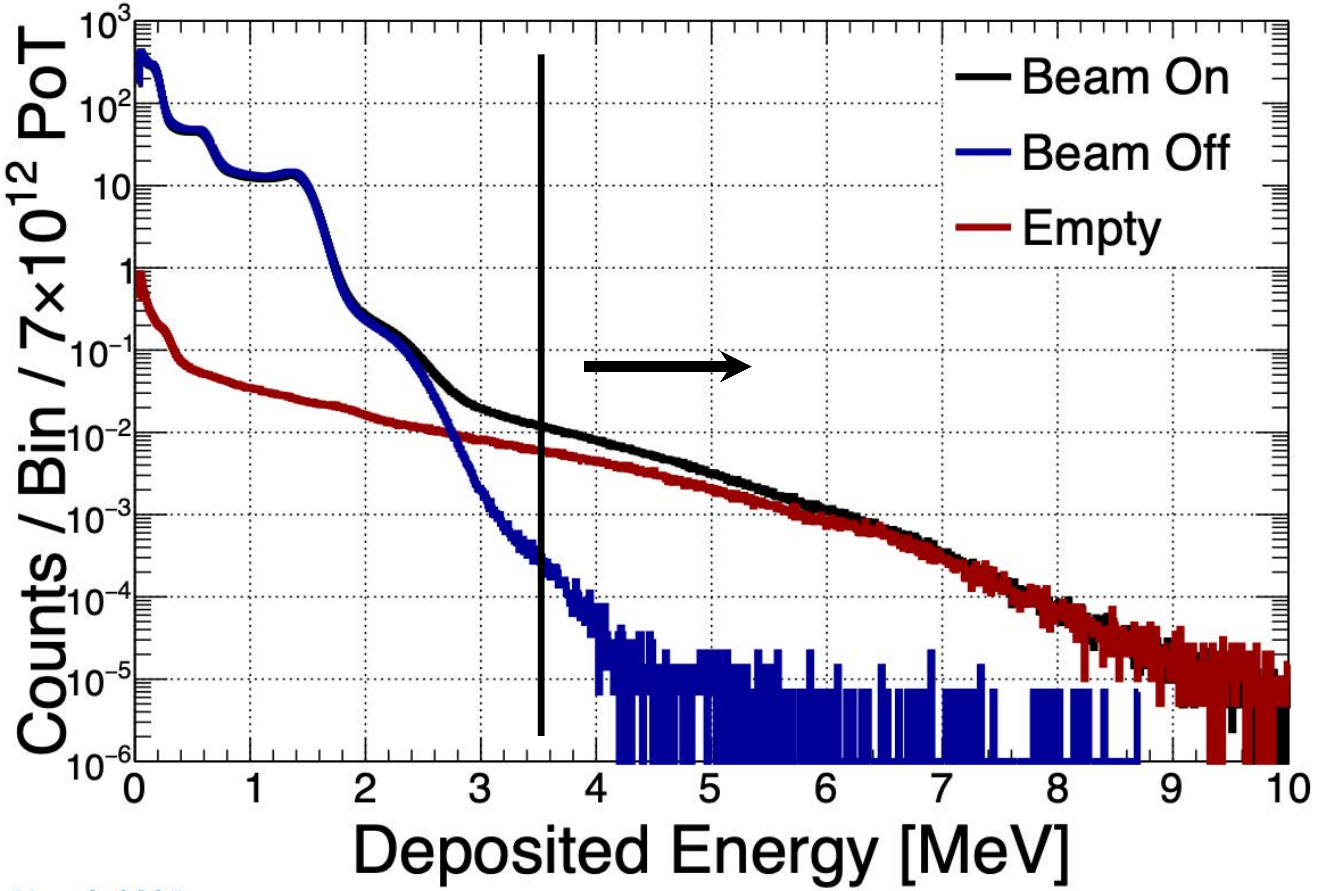
^{88}Zr – Deposited Energy (Gammas)

Compton shoulders:
^{88}Zr 393 keV (97%)
^{88}Y 898 keV (94%)
^{88}Y 1.836 MeV (99%)
^{88}Y 2.734 MeV (0.6%)
^{88}Y 3.218 MeV (0.007%)



^{88}Zr – Background Subtraction

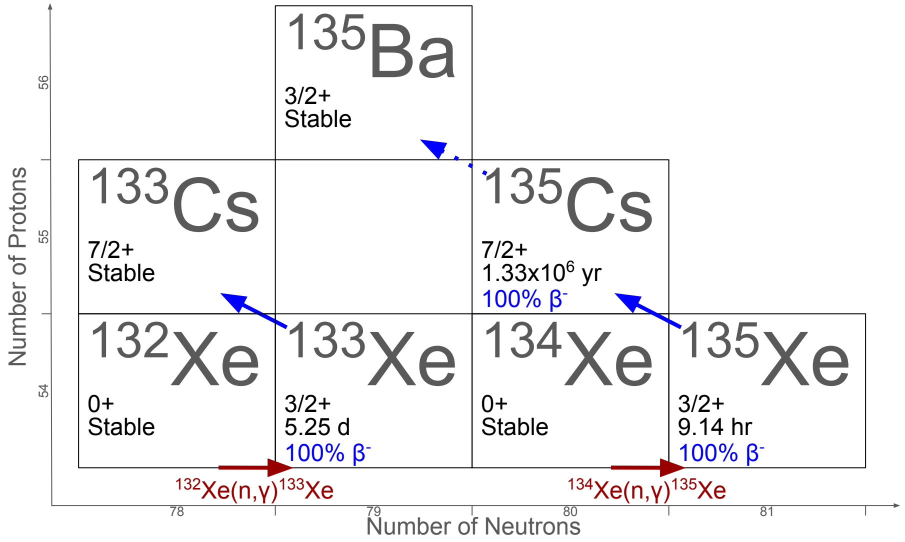
- Require 3.5 MeV of energy to be deposited in scintillators
  - To eliminate ^{88}Zr and ^{88}Y EC gammas

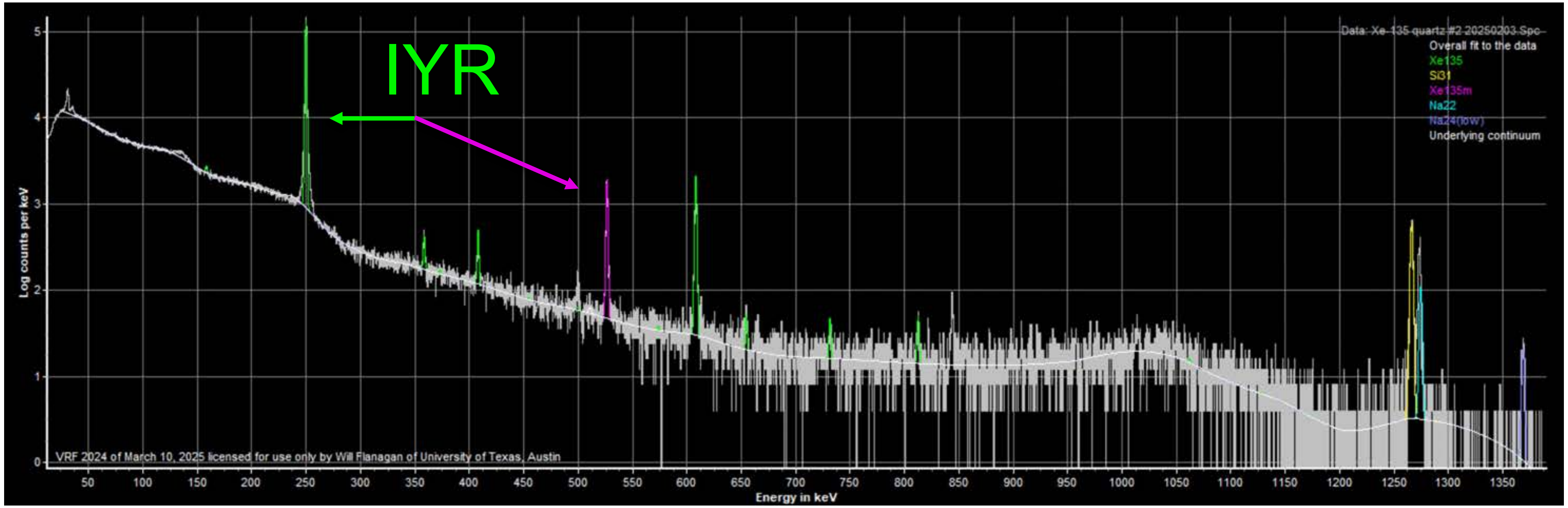


$^{134}Xe(n,\gamma)$ isomeric yield ratio

- Below is data from an irradiated high-purity quartz vial containing ^{134}Xe

- 526 keV $^{135m}Xe(IT)^{135g}Xe$
- 250 keV $^{135g}Xe(\beta^-)^{135}Cs$

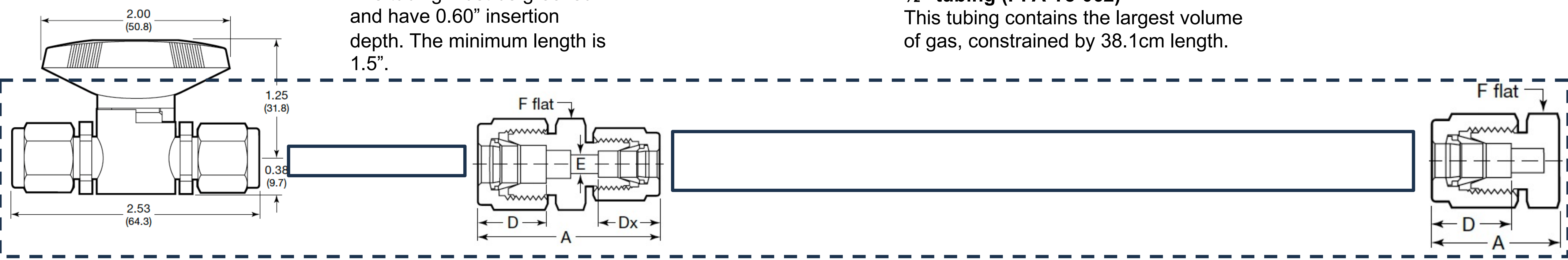




3EL Irradiation Geometry

¼” tubing (PFA-T4-062)
The tubing must be grooved and have 0.60” insertion depth. The minimum length is 1.5”.

½” tubing (PFA-T8-062)
This tubing contains the largest volume of gas, constrained by 38.1cm length.



PFA-43S4
This plug valve is frequently used for 0.56ml gas irradiations. Handle is taken off during irradiation due to diameter constraint.

Reducing Union (¼” to ½”, PFA-820-6-4)

½” cap (PFA-820-C)

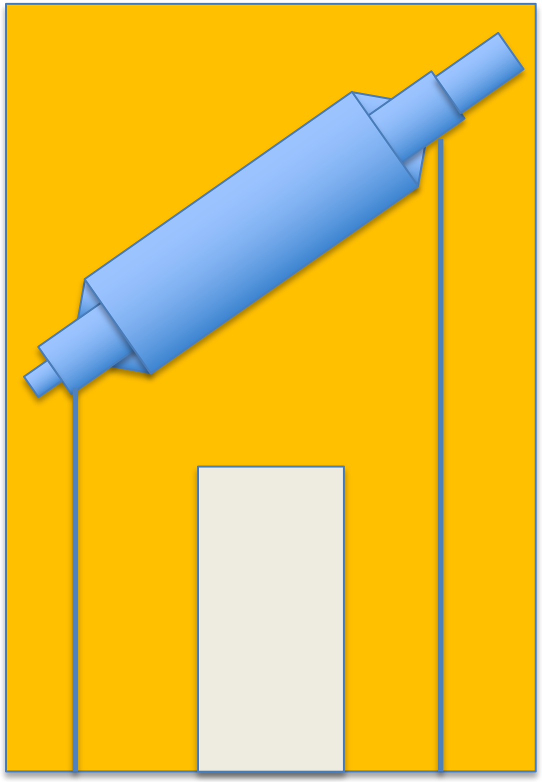
***Dashed lines represent 15” (38.1cm) length and 1.5” (3.81cm) diameter constraints**

- Based off Swagelok PFA plug valve (~0.5ml) which is regularly used for ^{135}Xe production at NETL
- → ¼” tubing→ ½” tubing for increased volume.
  - Approximately 15ml total volume

HPGe Counting Geometry

- In-core irradiation (90s) within 15mL PFA container, transfer to 60mL PTFE container (20 min)

- Irradiated $^{151/153}Eu$ to make $^{152/154}Eu$ for calibration
- Simulated gas using polystyrene balls to provide low density inside counting cylinder

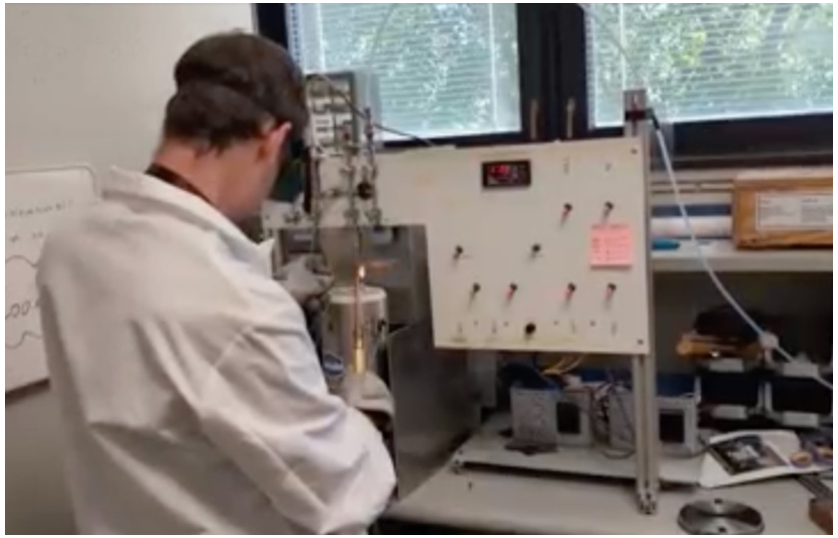


*Roughly To Scale, inside Clamshell

High-Purity Quartz Encapsulation

- High-purity quartz encapsulation allows in-core irradiation and rapid transfer
- Liquid nitrogen bath allows low-loss transfer

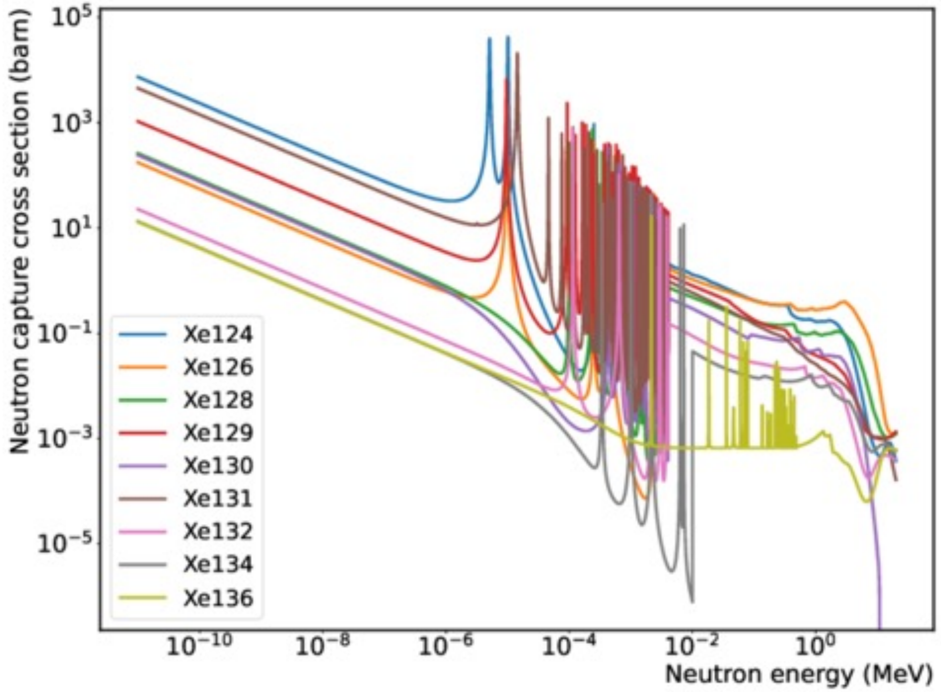
- Isotopically enriched xenon gas (^{124}Xe up to ^{136}Xe) available at UT NETL from long history with radioxenon!

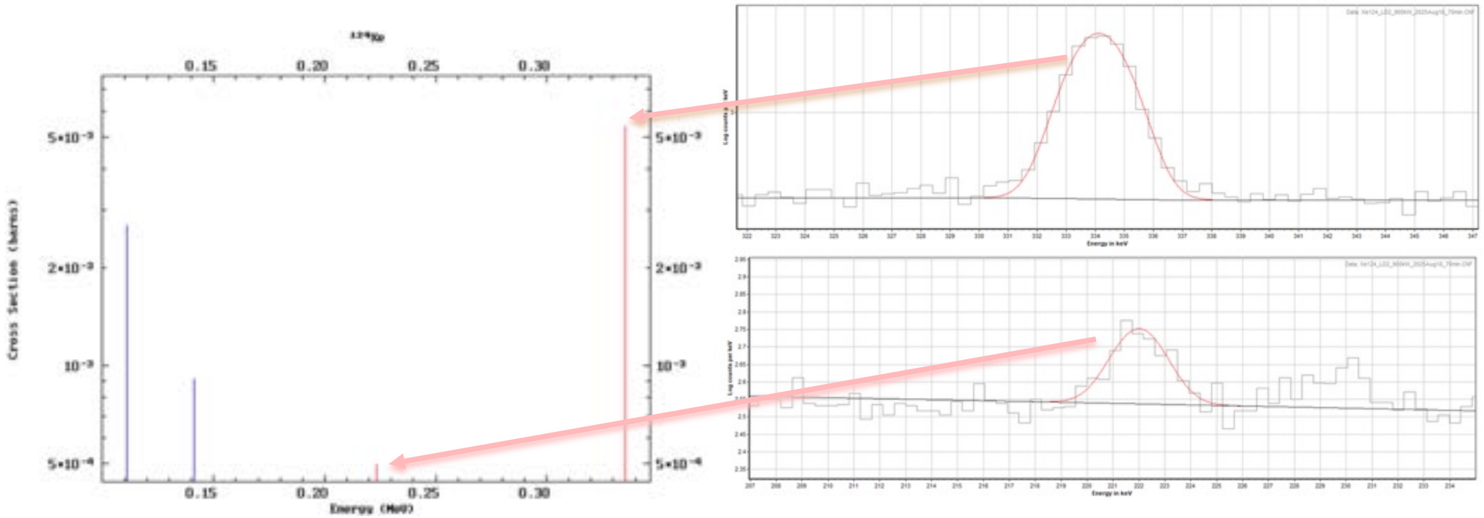


Xe Prompt Gamma Progress So Far

TEXAS
The University of Texas at Austin
WHAT STARTS HERE CHANGES THE WORLD

- ^{124}Xe prompt gammas confirmed in last two weeks as well as various natural xenon lines.
  - ^{124}Xe has large cross section and simple structure.
    - 335 keV (top) and 224 keV (middle) prompt gammas
    - Validates form factor, methodology … even without the beautiful coincidence of FIPPS





IAEA database

July 18, 1hr of data

Additional prompt gammas of ^{nat}Xe