Isomeric yield ratio measurements for fission dynamics

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UNIVERSITY OF JYVÄSKYLÄ

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Outline

Motivation and background

Isomeric yield ratios (IYRs) for studying

angular momenta of fission fragments

Experimental approach and ...

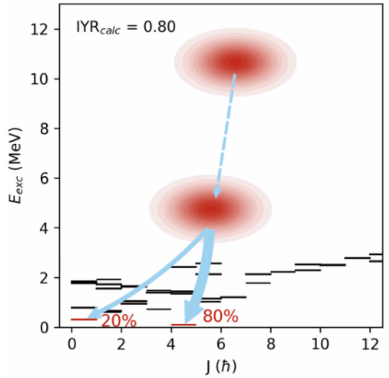
… results from IGISOL for ^{232}Th(α,f)

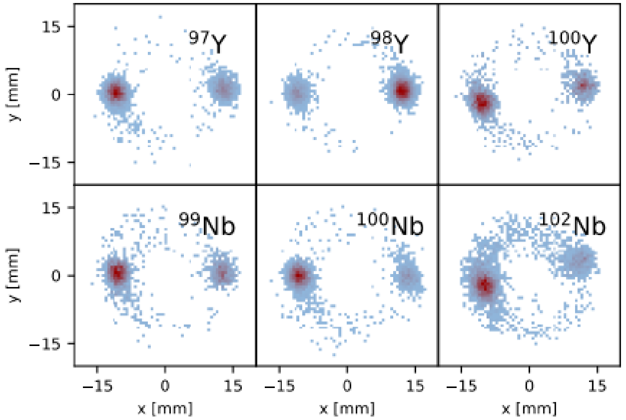
highlighting the impact of CN angular momentum

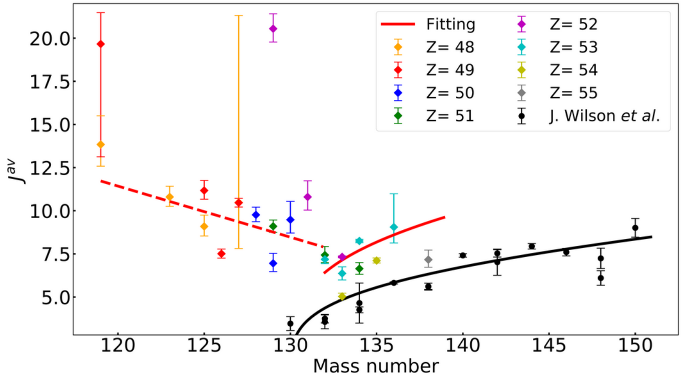
… with special attention to the ^{98}Y region

Linking isomeric yields to fission fragment angular momenta

Summary and outlook







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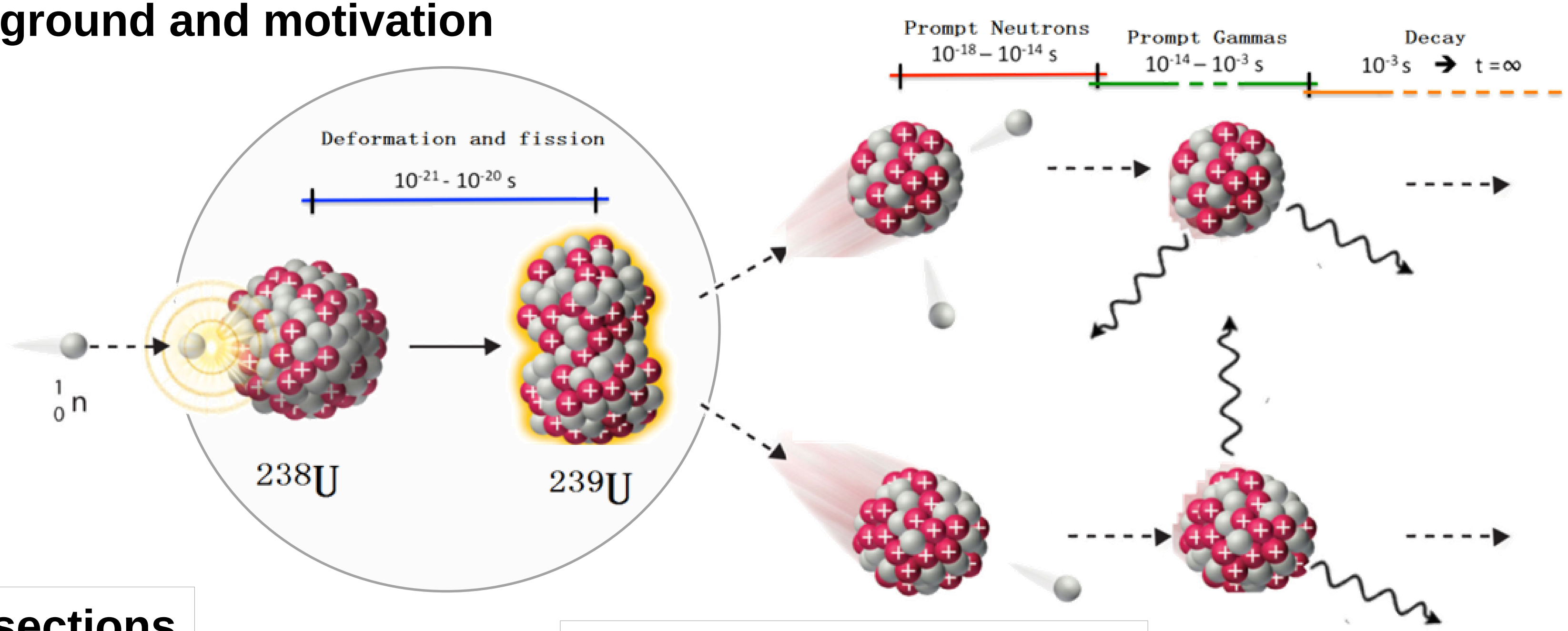
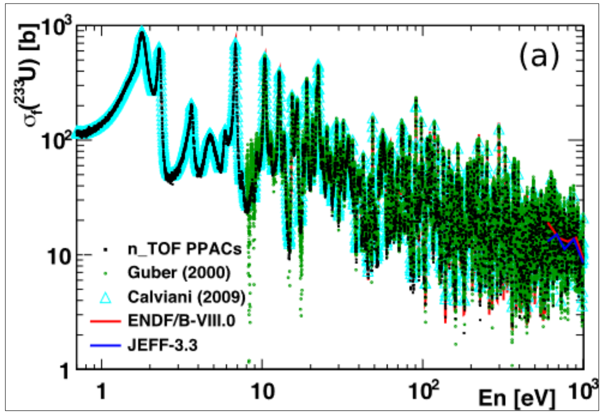
Background and motivation

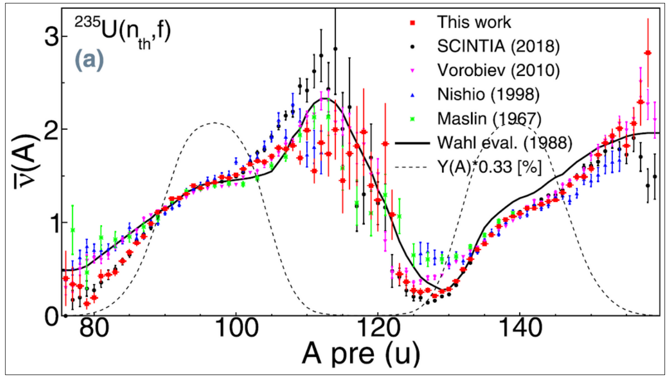


cross sections



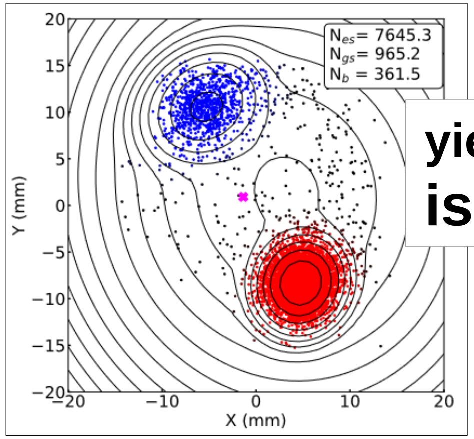
D. Tarrío et al, Phys. Rev. C **107**, 044616 (2023)

prompt neutron emission



A. Al-Adili et al, Phys. Rev. C **102**, 064610 (2021)

yields and isomeric ratios



Z. Gao et al, Phys. Rev. C **108**, 054613 (2023)

Angular momentum of fission fragments

PHYSICAL REVIEW C VOLUME 5, NUMBER 6 JUNE 1972

Angular Momentum of Primary Products Formed in the Spontaneous Fission of ^{252}Cf†

J. B. Wilhelmy,* E. Cheifetz,‡ R. C. Jared, S. G. Thompson, and H. R. Bowman
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

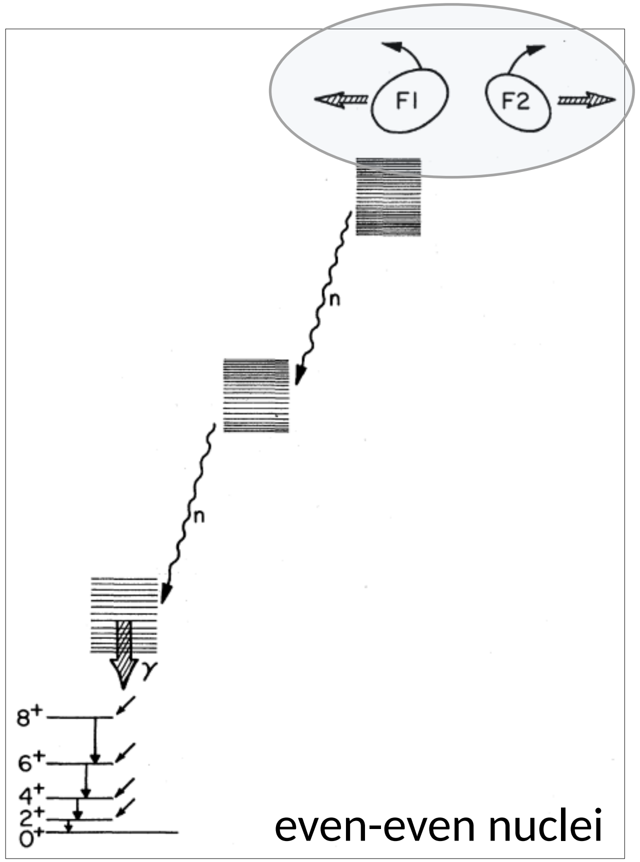
and

J. O. Rasmussen
Chemistry Department, Yale University, New Haven, Connecticut 06520

(Received 22 November 1971)

Wilhelmy et al (1972):
Fission fragments from ^{252}Cf(sf)
have **angular momenta of about 7 ħ**.

Somehow the fission fragments are tumbling apart after scission. What makes them do so?



Already before the experimental evidence for these surprisingly large angular momenta there where theories suggesting

- **pre-fission** angular momentum generation;
  collective vibrations; wriggling, bending, twisting;
  see e.g. J. Randrup and R. Vogt and the FREYA code

- **post-scission** models
  Coulomb forces; microscopic theories (e.g. Bertsch et al. 2019)

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Angular momentum of fission fragments

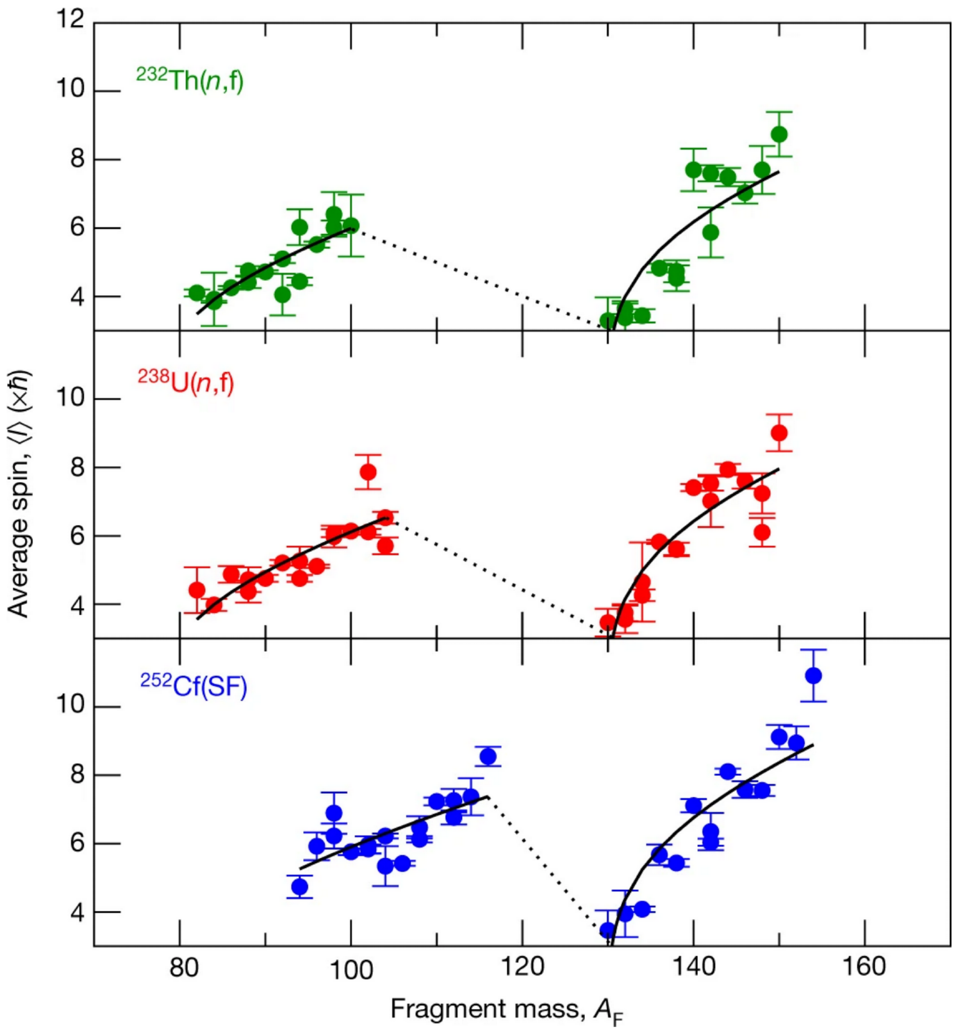
The debate got fresh fuel with the results
from J. Wilson et al., Nature **590**, 566 (2021).

All three studied systems showed
the same sawtooth-like behaviour*.

Furthermore:

"*Here we show that there is* ***no significant correlation*** *between the spins of the fragment partners, which leads us to conclude that angular momentum in fission is actually* ***generated after the nucleus splits (post-scission)***."

Several subsequent papers challenged this conclusion,
e.g., Randrup&Vogt, PRL 127, 062502 (2021).



* Based on gamma-spectrometry of side-feeding in 0^+–nuclei.

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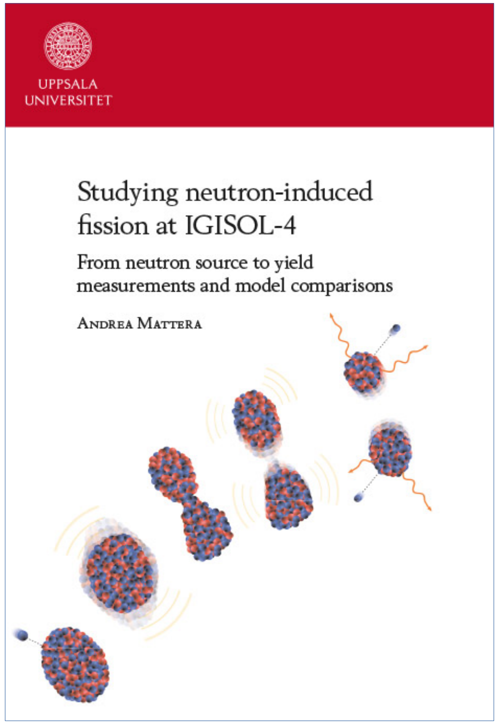
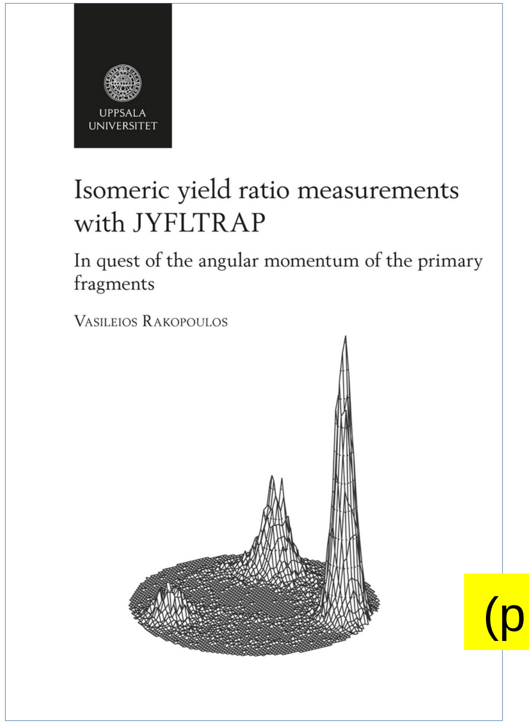
We study <u>isomeric yield ratios</u> from fission to address this quest of angular momentum generation



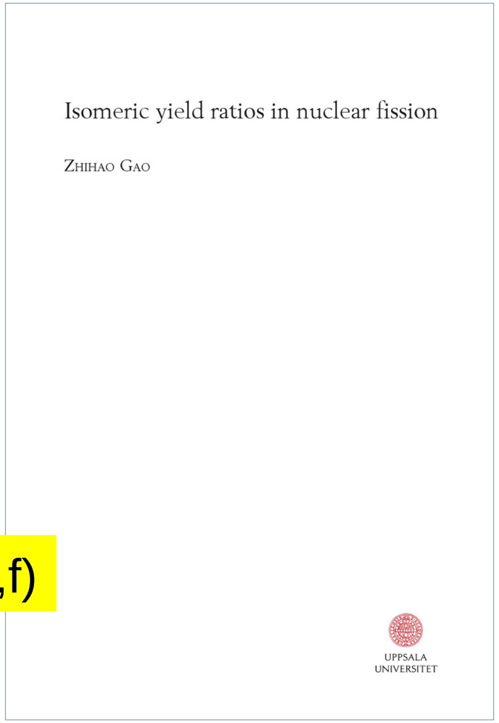
EPJ A (2017) **53**: 173
EPJ A (2018) **54**: 33

(n,f)

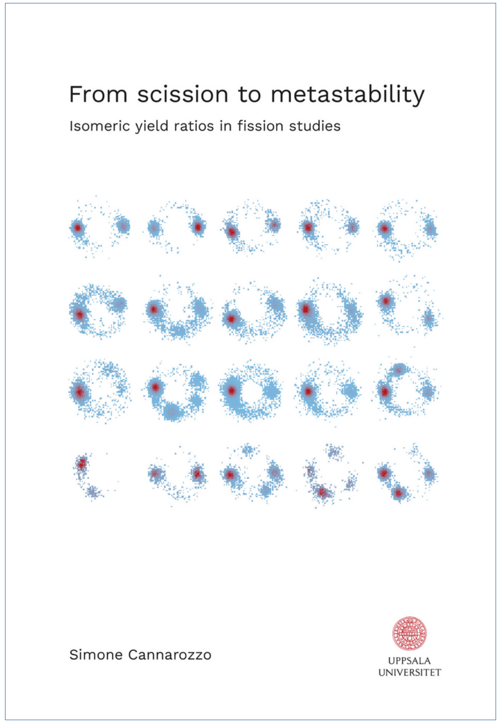


PRC **98** 024612 (2018)
PRC **99** 014617(2019)

(p,f)



EPJ A (2022) **58**: 27
EPJ A (2023) **59**: 169
PRC **108** 054613 (2023)
PRC **109** 064626 (2024)



EPJ A (2023) **59**: 295
PRC **111** L031601 (2025)
Manuscript submitted to PLB

(α,f)

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Isomers from fission

The existence of nuclear isomers
Was suggested already 1917
by Frederick Soddy
as ”a finer degree of isotopy”.

They were experimentally
discovered by Otto Hahn
104 years ago.

Today about 2500 isomers
with half-lives
larger than **10 ns** are known.

Falls UX_1 die Muttersubstanz ist, dann muß die Ausbeute an Z aus UX-Präparaten mit der Halbwertszeit des UX_1, also rund 24 Tagen, abnehmen. Ob dies tatsächlich der Fall ist, konnte noch nicht genau festgestellt werden. Versuche darüber sind im Gange. Falls sich diese genetische Beziehung bestätigt, so muß man für das UX_1 einen dualen Zerfall annehmen, wie ihn das folgende Schema veranschaulicht:

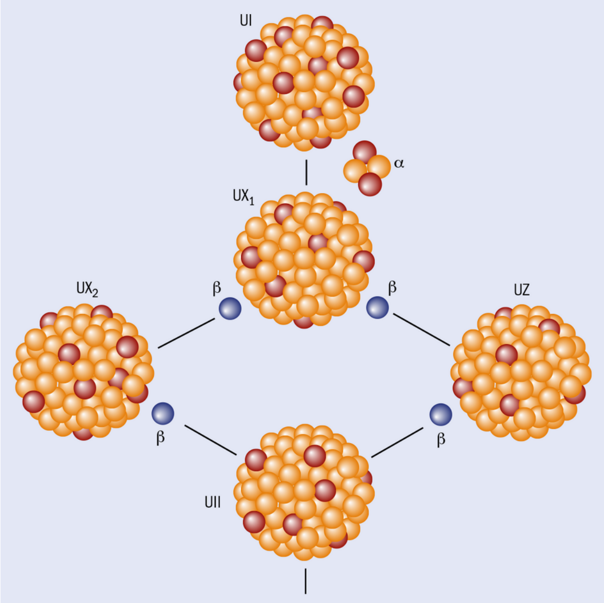
U I
| α
UX_1
β ↙ ↘ β
UX_2 Z
β ↘ ↙ β
U II
| α

Sollte es sich dagegen herausstellen, daß UX_1 nicht die Muttersubstanz von Z ist, so hätte man ein mit UX_1 isotopes Produkt anzunehmen, für dessen Ursprung dann nur ein neues Uranisotop UIII in Frage käme. Dieses wäre dann vermutlich die Ausgangssubstanz für eine neue Reihe radioaktiver Zerfallsprodukte, deren einzelne Glieder sich unter die entsprechenden isotopen Glieder der Uran-Radium-Reihe mischten, und die man bei ihrer geringen Intensität bisher sehr wohl hätte übersehen können. Auf die mancherlei sich hieraus ergebenden Folgerungen sei hier vorerst nicht eingegangen, weil ja das Experiment in nicht zu langer Zeit entscheiden wird, welche der beiden Entstehungsmöglichkeiten von Z den Tatsachen entspricht.

Berlin-Dahlem, den 21. Januar 1921.

Otto Hahn,
Kaiser-Wilhelm-Institut für Chemie.

Hahn, Otto: „Über ein neues radioaktives Zerfallsprodukt im Uran“,
Die Naturwissenschaften **9**, 84 (1921) doi:10.1007/BF01491321



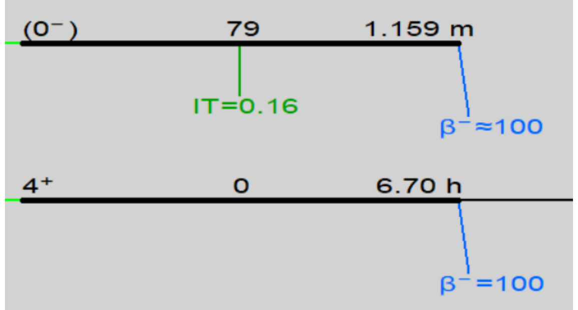
^{238}U

^{234}Th

^{234}Pa

^{234}U

Ph. Walker and Zsolt Podolyák:
”Celebrating a centrury of nuclear isomers”,
Physics World, April 2021.



^{234}Pa

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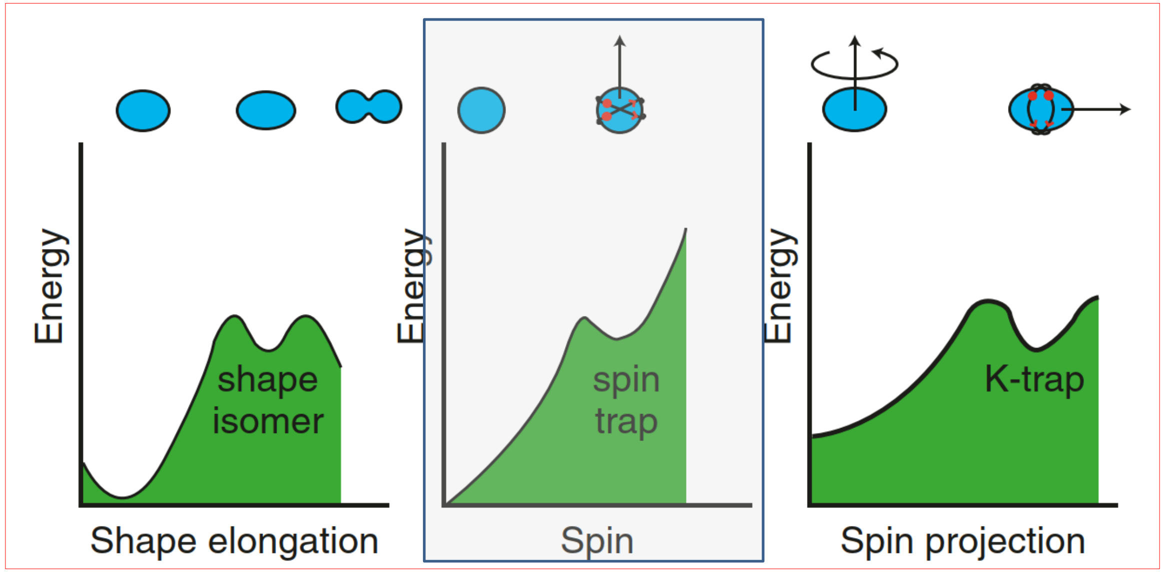
Isomers from fission and fragment angular momenta

Relative population of different spin states,
reflects the angular momentum of a fission fragment
decaying into either of these states.



Walker & Dracoulis,
”Energy traps in atomic nuclei”, Nature **399**, 35 (1999)
https://doi.org/10.1038/19911

PHYSICAL REVIEW VOLUME 120, NUMBER 4 NOVEMBER 15, 1960

Interpretation of Isomeric Cross-Section Ratios for (n,γ) and (γ,n) Reactions*

J. R. HUIZENGA AND R. VANDENBOSCH
Argonne National Laboratory, Argonne, Illinois

(Received July 11, 1960)

The relative probability of forming each member of a pair of nuclear isomeric states has been compared with theoretical predictions in order to learn which nuclear parameters can be determined from these data. For thermal neutron capture reactions, the observed ratios do not give much information about the dependence of the nuclear level density on spin, but they are consistent with a spin cutoff factor, $\exp[-(J+\frac{1}{2})^2/2\sigma^2]$, where $\sigma \leq 5$. The calculations are sufficiently consistent with experiment to make their predictions usable as a guide for assigning spins to the compound states formed in thermal or resonant energy neutron capture. For (γ,n) reactions, the calculations reproduce the energy dependence of the experimentally observed isomeric cross-section ratios. In order to obtain quantitative information about the spin dependence of the nuclear level density, it is necessary to consider reactions where particles are emitted which can carry off enough angular momentum to reach many spin states of the residual nucleus.

1. INTRODUCTION

THE relative probability of forming each state of an isomeric pair seems to be governed mainly by the spin differences between the states which decay to the isomers and the isomer spins themselves. In the many cases (encountered in radioactivity) in which a third low-lying state can decay to either of the isomers, the well-known preference of the photon transition of

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

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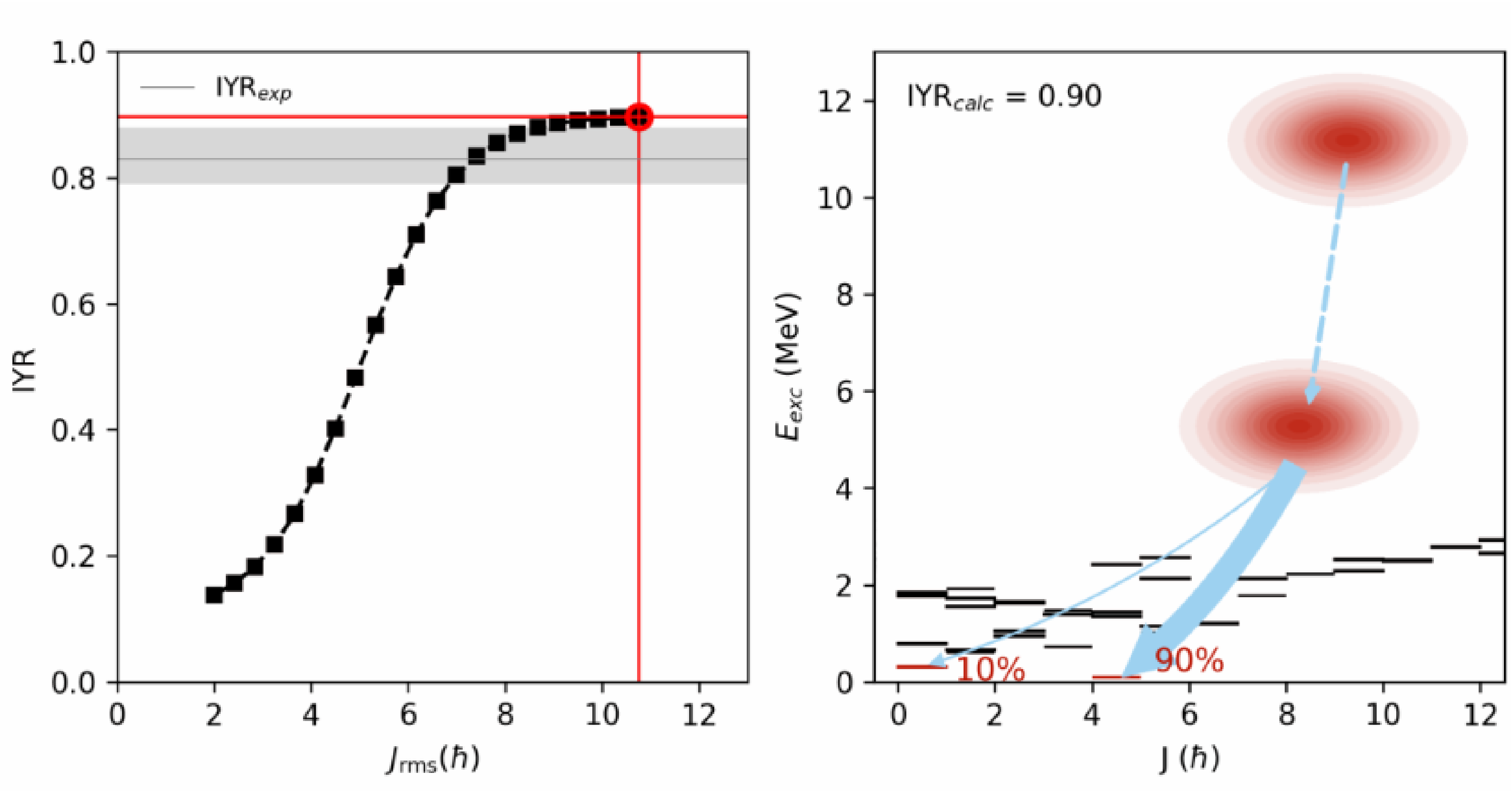
Isomers from fission and fragment angular momenta

1) Measure IYR, i.e., relative population of (long-lived) isomers.

2) Develop methods to “back-track” and determine J_{rms} of fragments.

$$IYR = \frac{Y_{high\ spin}}{Y_{high\ spin} + Y_{low\ spin}}$$



Fission fragment

Prompt neutron emission

Fission product

Prompt gamma emission

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Experimental approach

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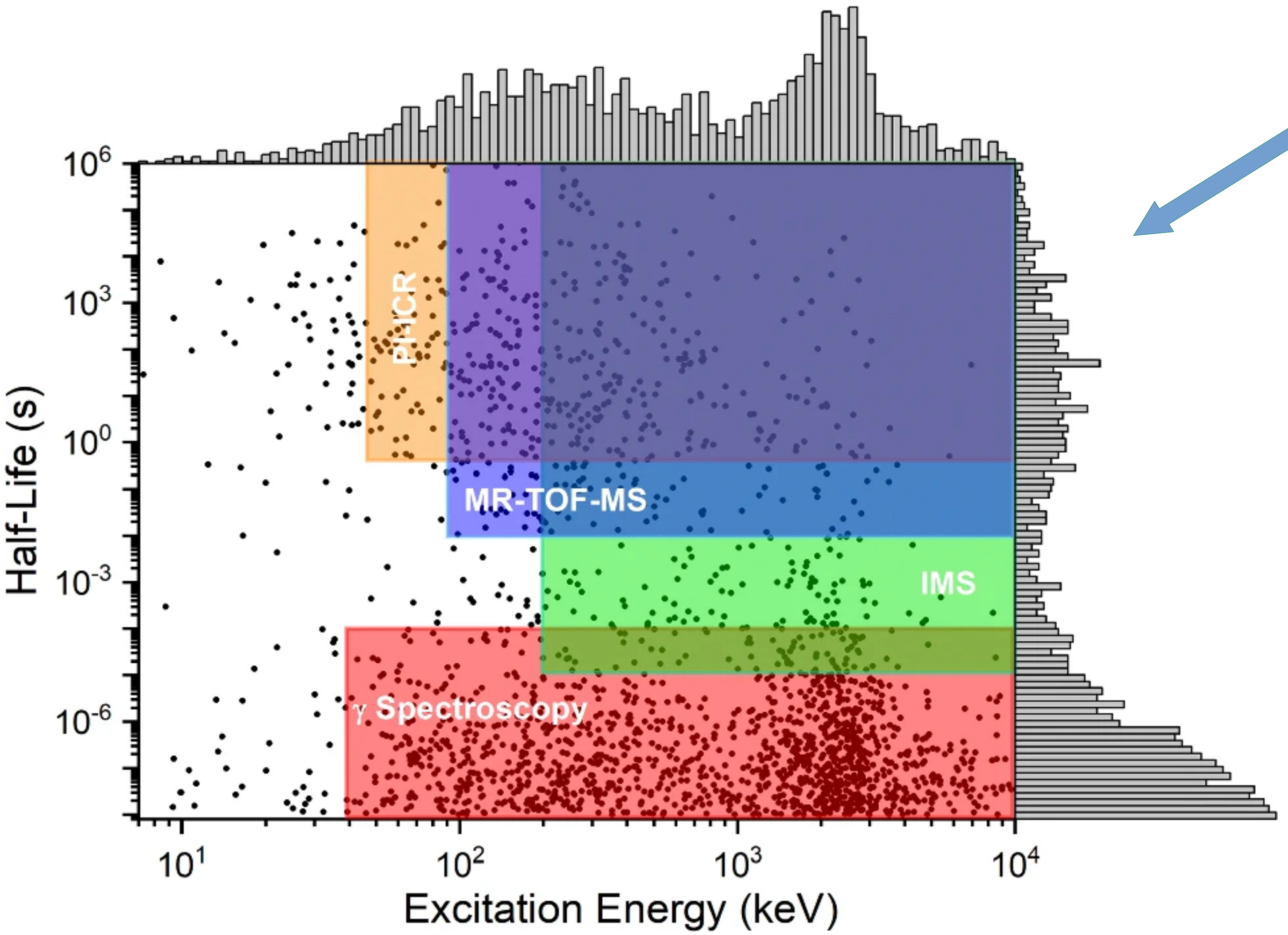
Possible measurement techniques for IYR



Dickel, T., Mollaebrahimi, A., *Eur. Phys. J. Spec. Top.* **233**, 1181–1190 (2024)

We use direct ion counting,
i.e., mass measurement techniques like phase-imaging ion-cyclotron-resonance (PI-ICR) and multi-reflection time-of-flight (MR-TOF)

These techniques offer a significant advantage compared to techniques using γ-ray spectroscopy, since **we do not rely on available knowledge of the nuclear level schemes.**

However, these techniques require relatively "long" half-lives.

This is pioneered by the Uppsala group in collaboration with the IGISOL group at the University of Jyväskylä, Finland.

See, e.g., Rakopoulos et al PRC 2018 and 2019, and R. Korkiamäki, MSc thesis, University of Jyväskylä (2024)

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Let’s go to Jyväskylä in Finland! [Mennään Jyväskylään Suomessa!]

IGISOL + Penning Trap to measure IYR from proton-and alpha-induced fission

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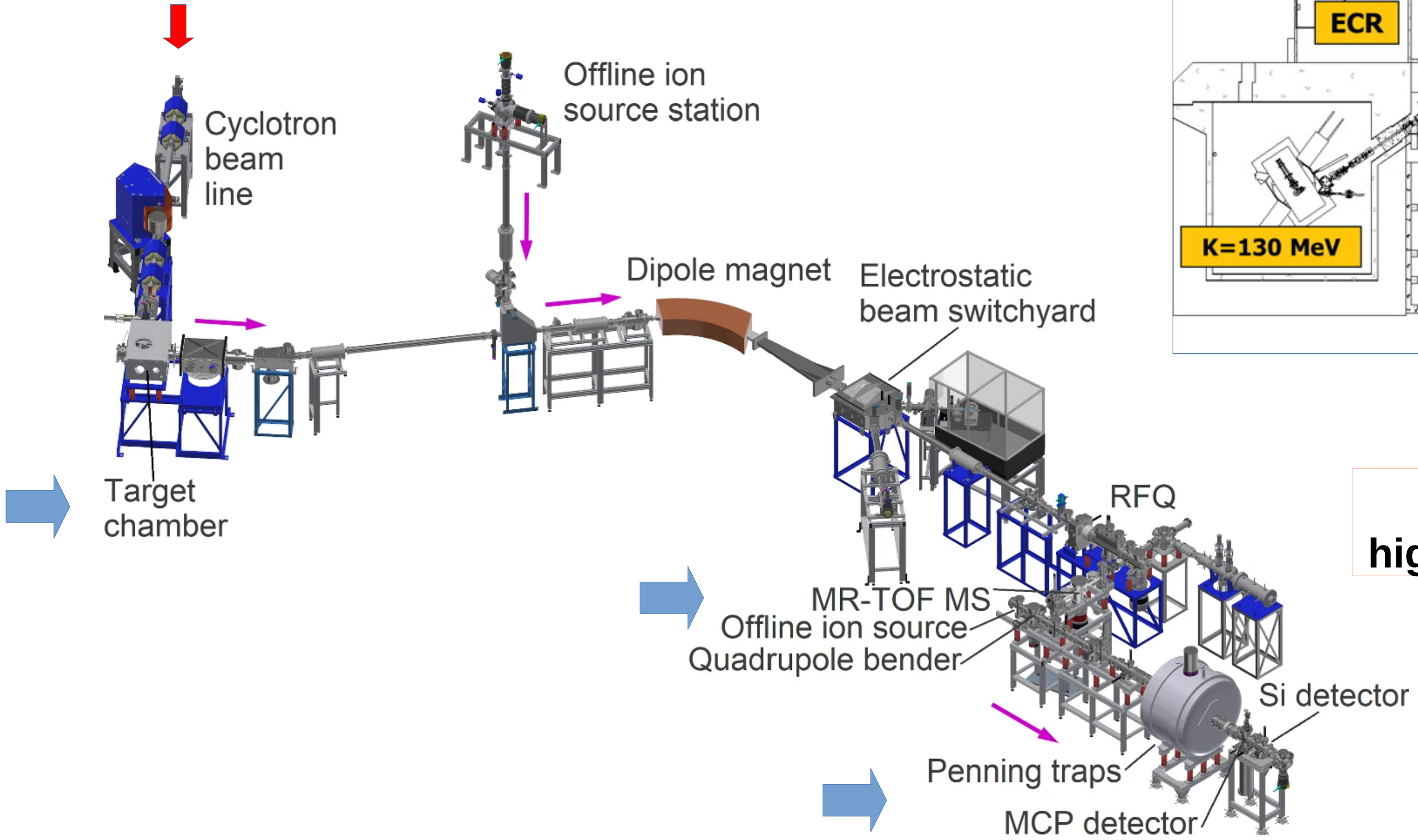
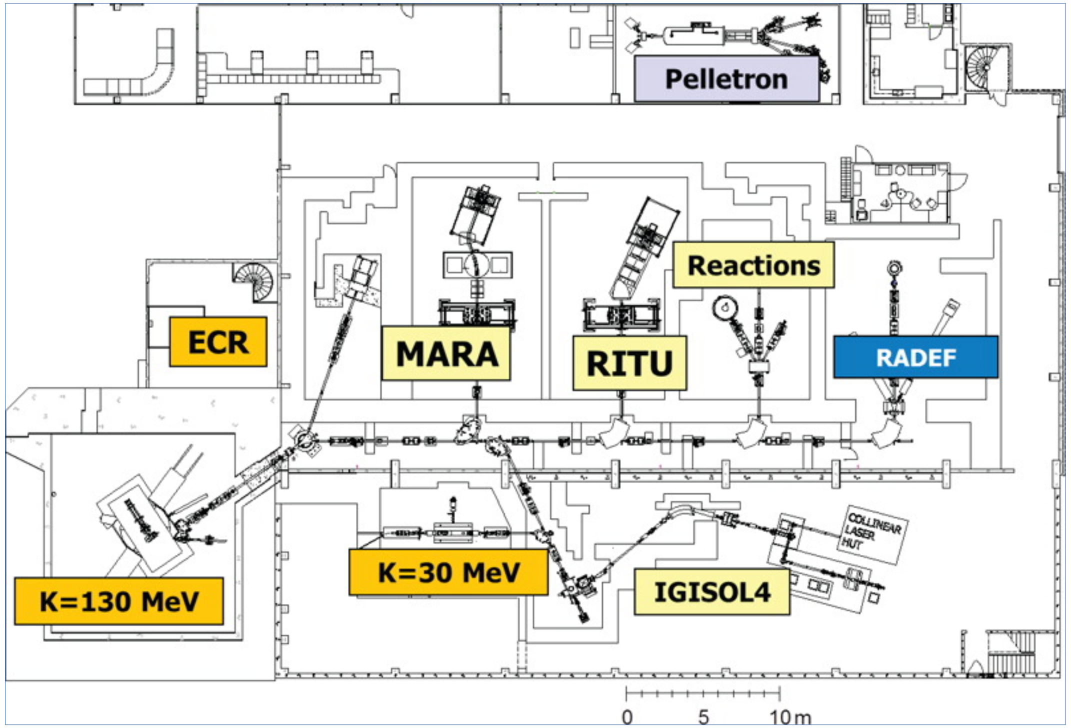
IGISOL with MR-TOF and JYFLTRAP

Beam from K-130 cyclotron





Ari Jokinen, Nuclear Physics News 24(4), 2014
https://doi.org/10.1080/10619127.2014.972165

The lab is well-known for, e.g.,
high-precision mass measurements

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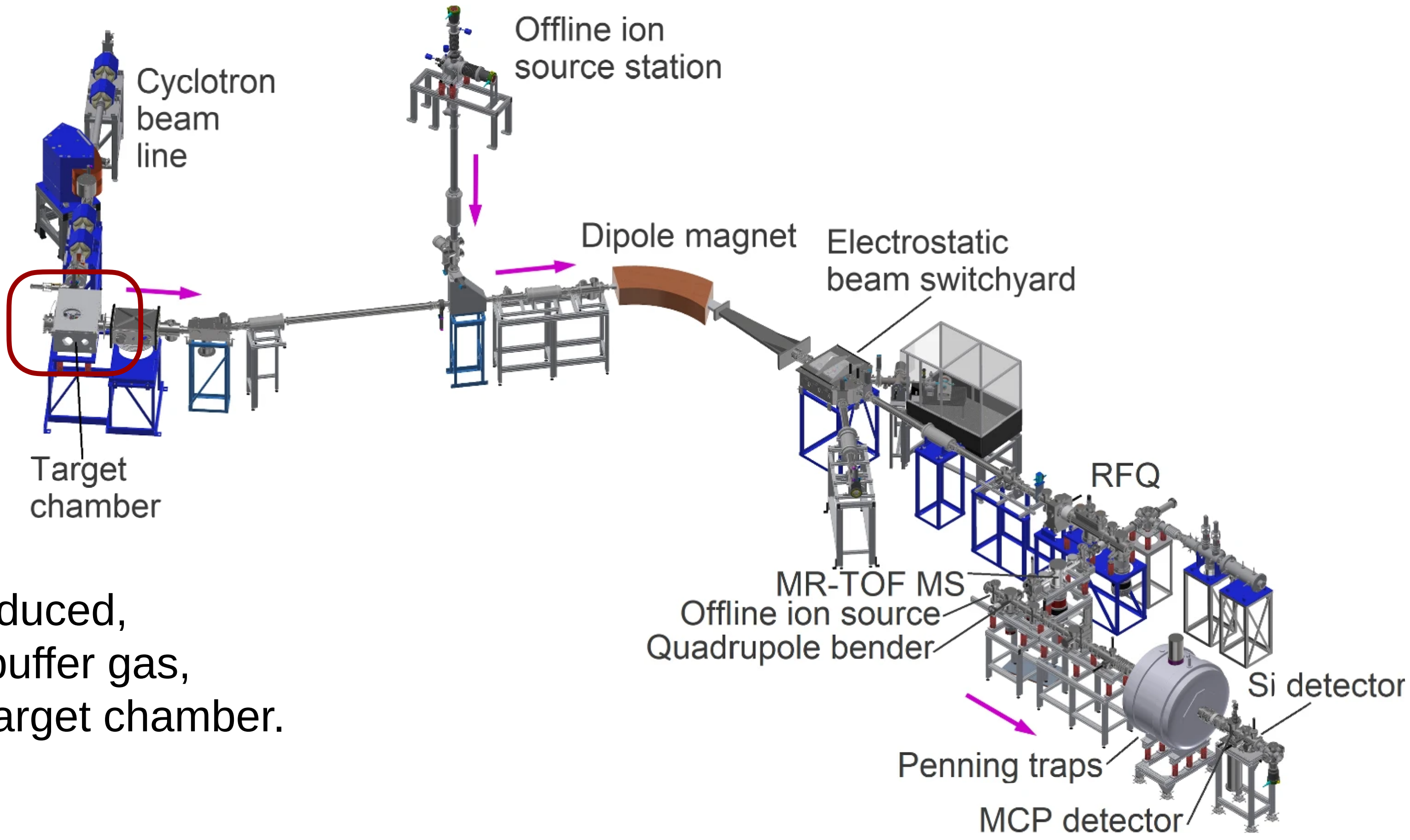
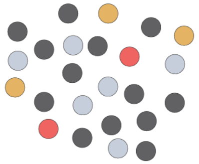
IGISOL with MR-TOF and JYFLTRAP





Fission products produced,
slowed down in He-buffer gas,
and extracted from target chamber.

Nesterenko, D.A., Eronen, T., Ge, Z. et al. “Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer.” Eur. Phys. J. A 57, 302 (2021)

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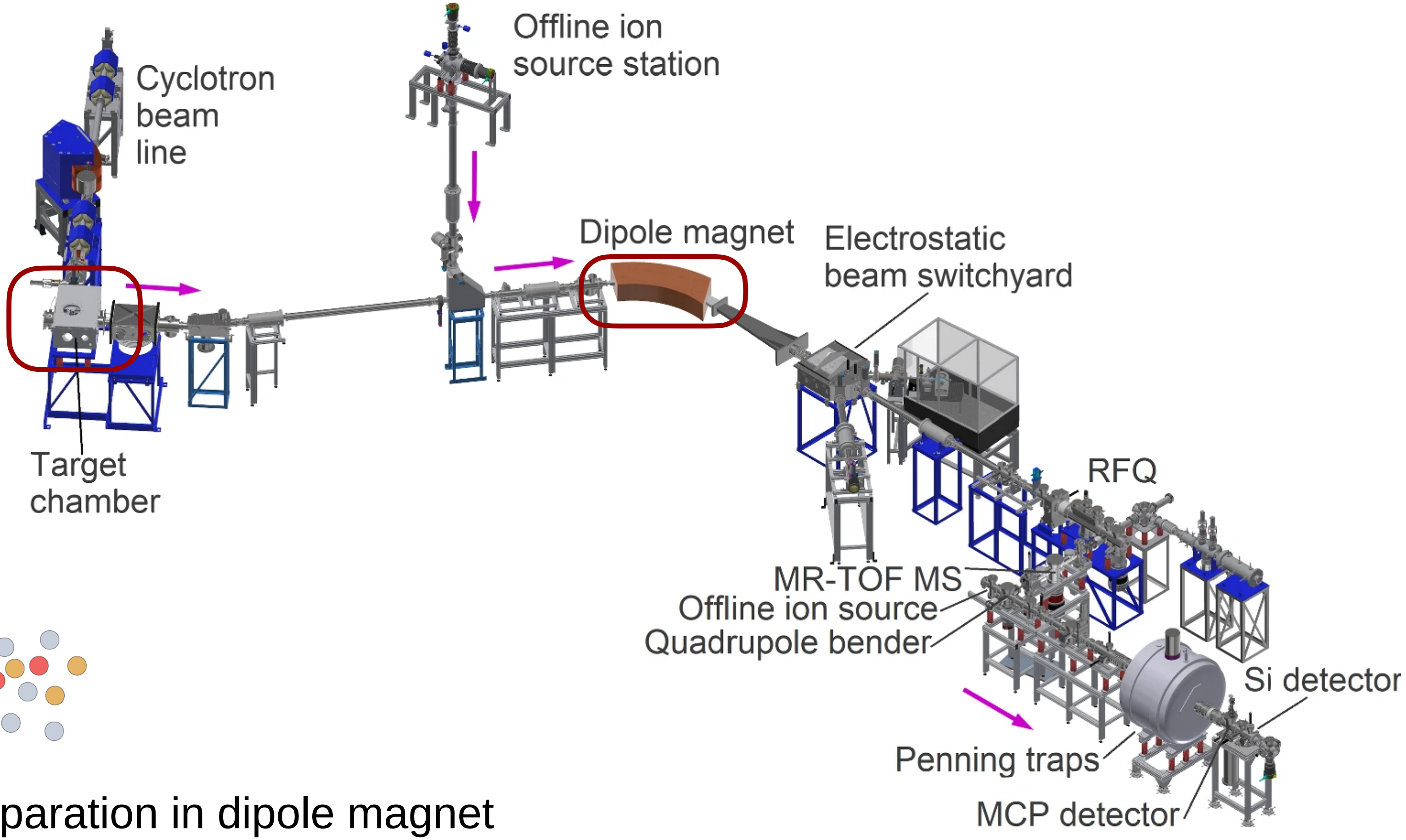
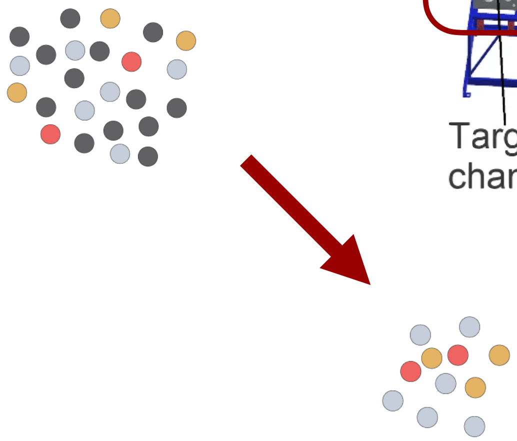
IGISOL with MR-TOF and JYFLTRAP





First mass separation in dipole magnet selects one isobar (usually in 1^+ state).

Nesterenko, D.A., Eronen, T., Ge, Z. et al. “Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer.” Eur. Phys. J. A 57, 302 (2021)

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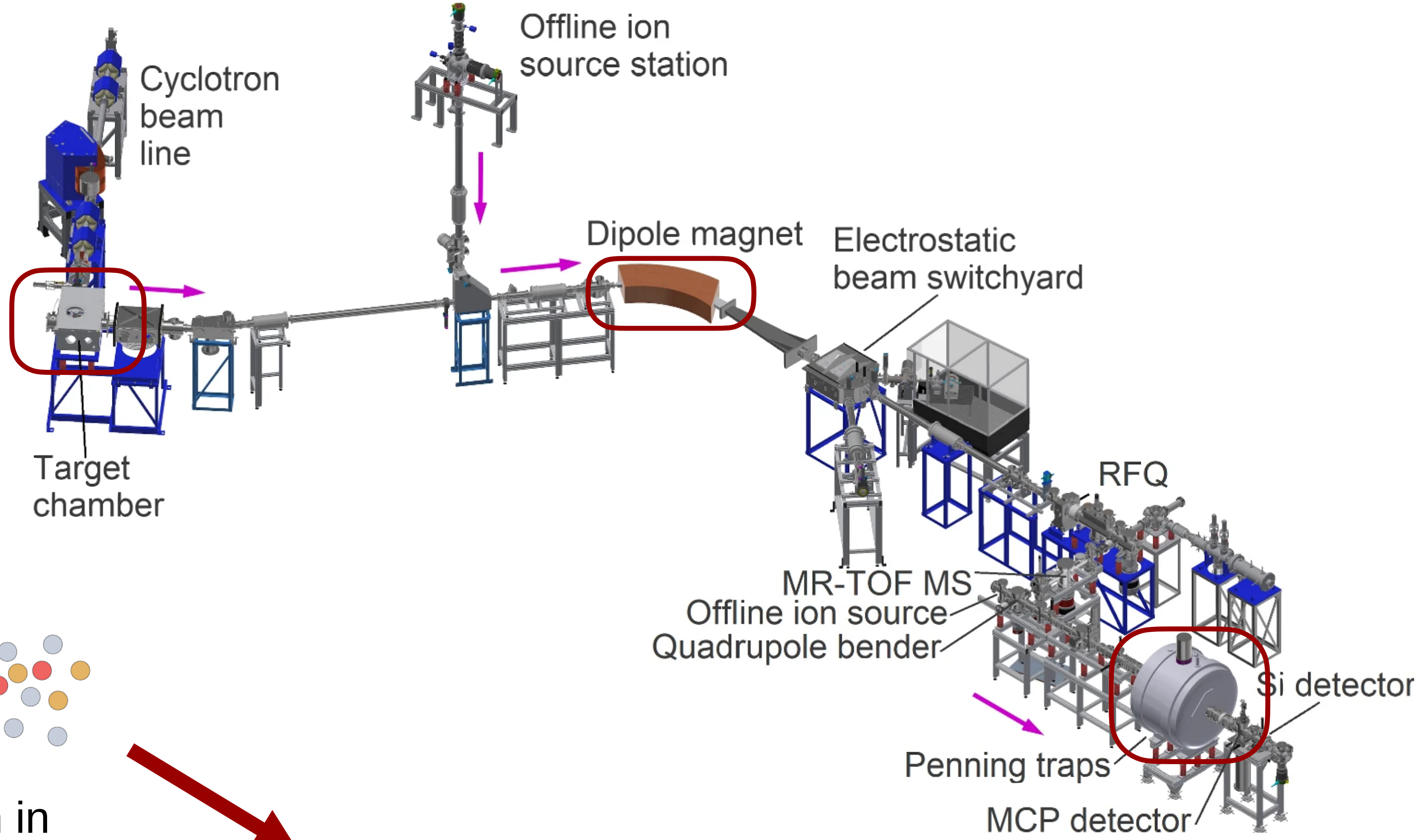
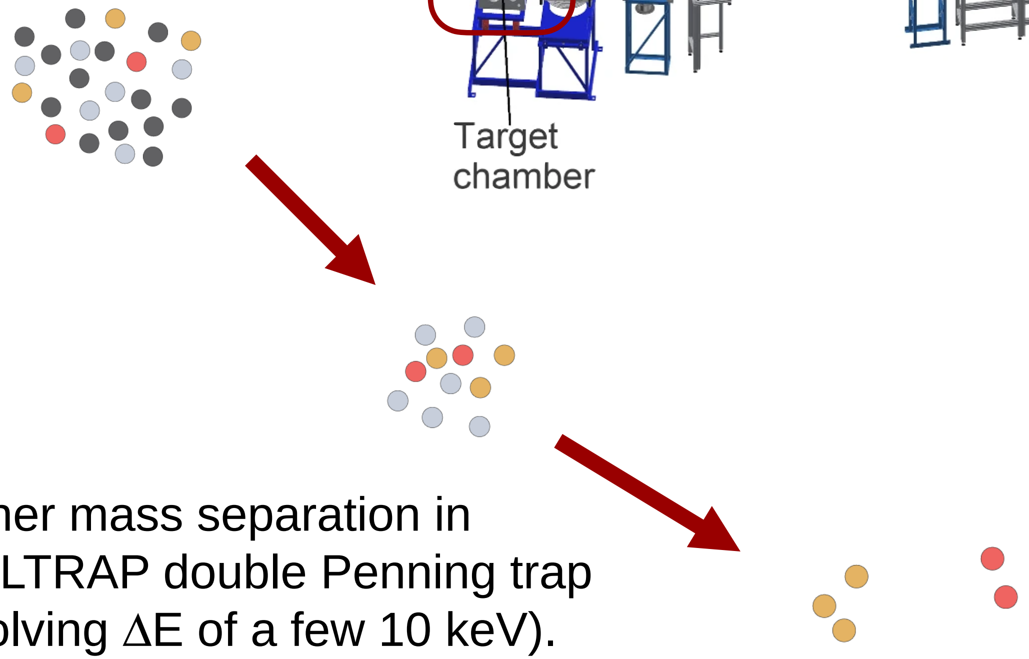
IGISOL with MR-TOF and JYFLTRAP





Further mass separation in
JYFLTRAP double Penning trap
(resolving ΔE of a few 10 keV).

Nesterenko, D.A., Eronen, T., Ge, Z. et al. “Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer.” Eur. Phys. J. A 57, 302 (2021)

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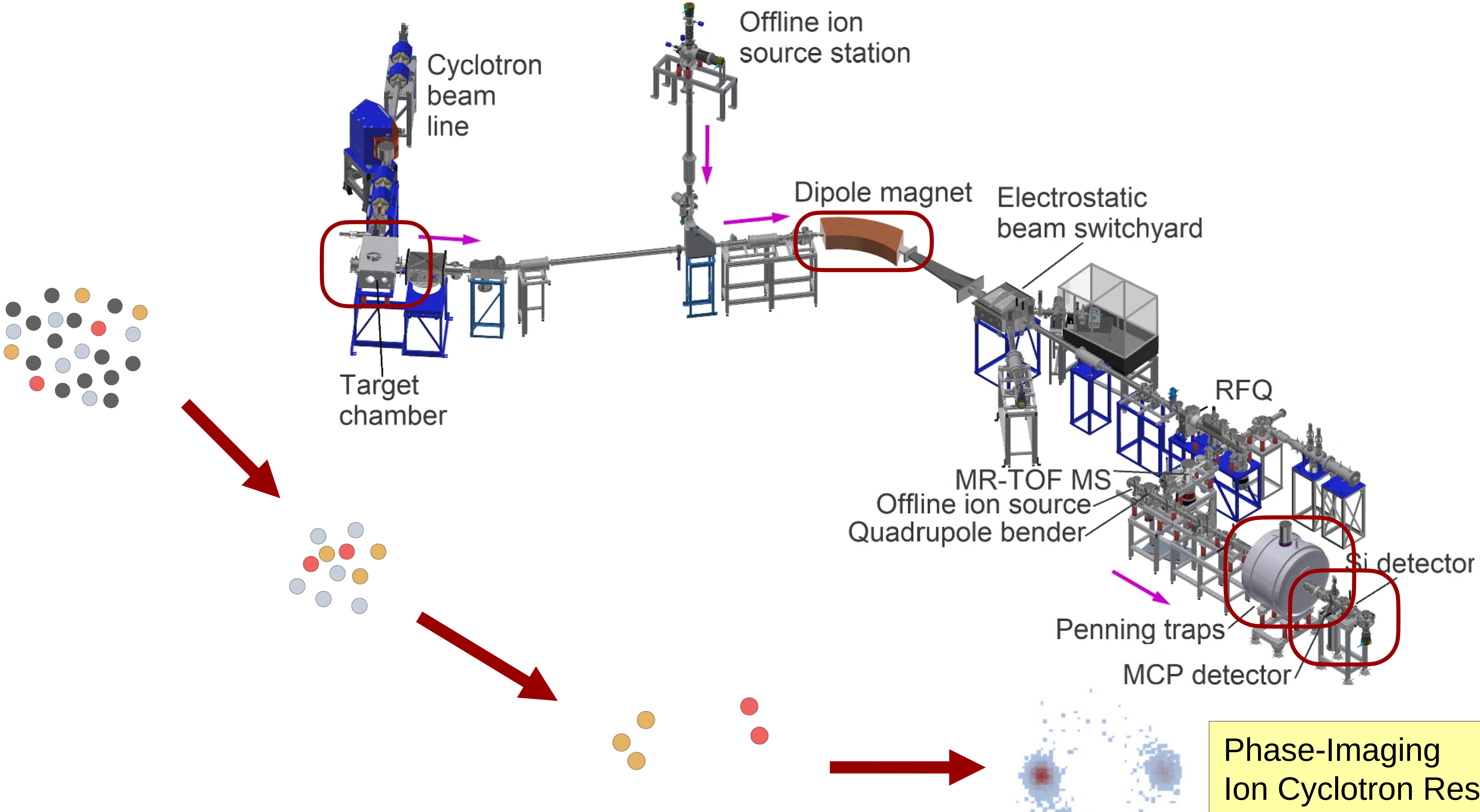
IGISOL with MR-TOF and JYFLTRAP



Phase-Imaging
Ion Cyclotron Resonance
(PI-ICR)

Nesterenko, D.A., Eronen, T., Ge, Z. et al. “Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer.” Eur. Phys. J. A 57, 302 (2021)

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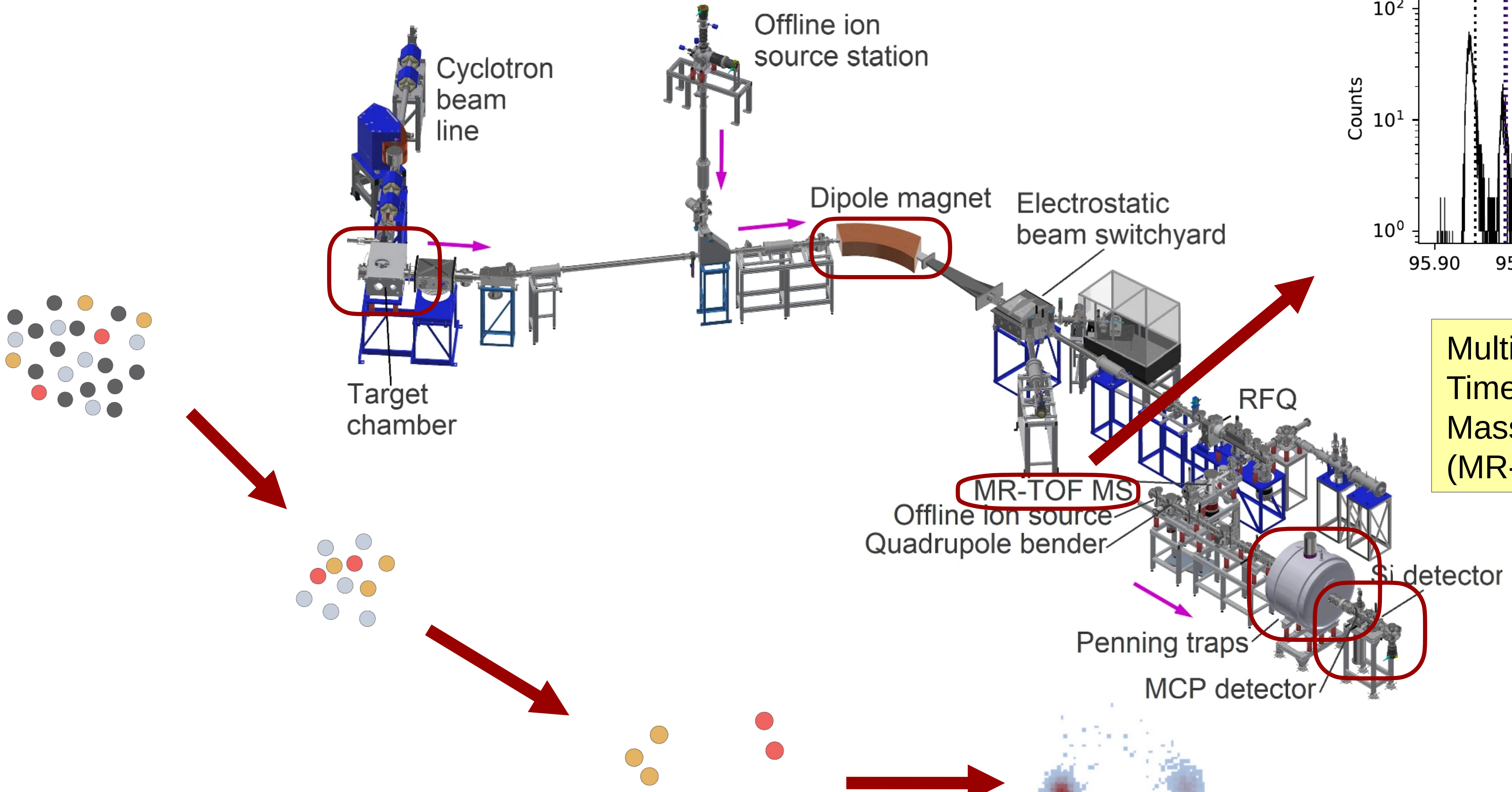
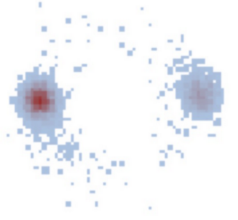
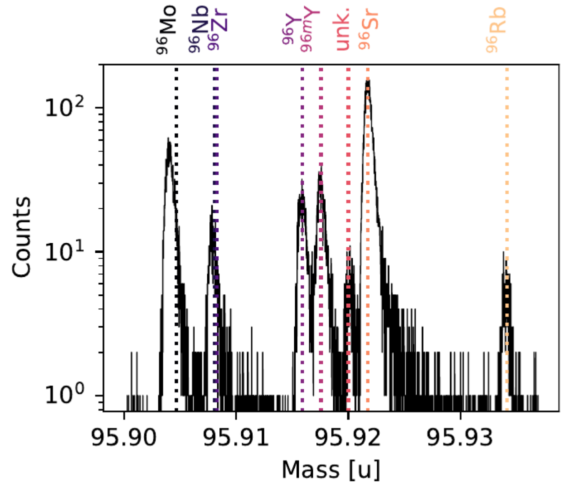
IGISOL with MR-TOF and JYFLTRAP







Multi-Reflection Time-Of-Flight Mass Spectrometer (MR-TOF-MS)

Nesterenko, D.A., Eronen, T., Ge, Z. et al. “Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer.” Eur. Phys. J. A 57, 302 (2021)

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The phase-imaging ion-cyclotron-resonance technique (PI-ICR) is powerful!

PHYSICAL REVIEW C **99**, 014617 (2019)

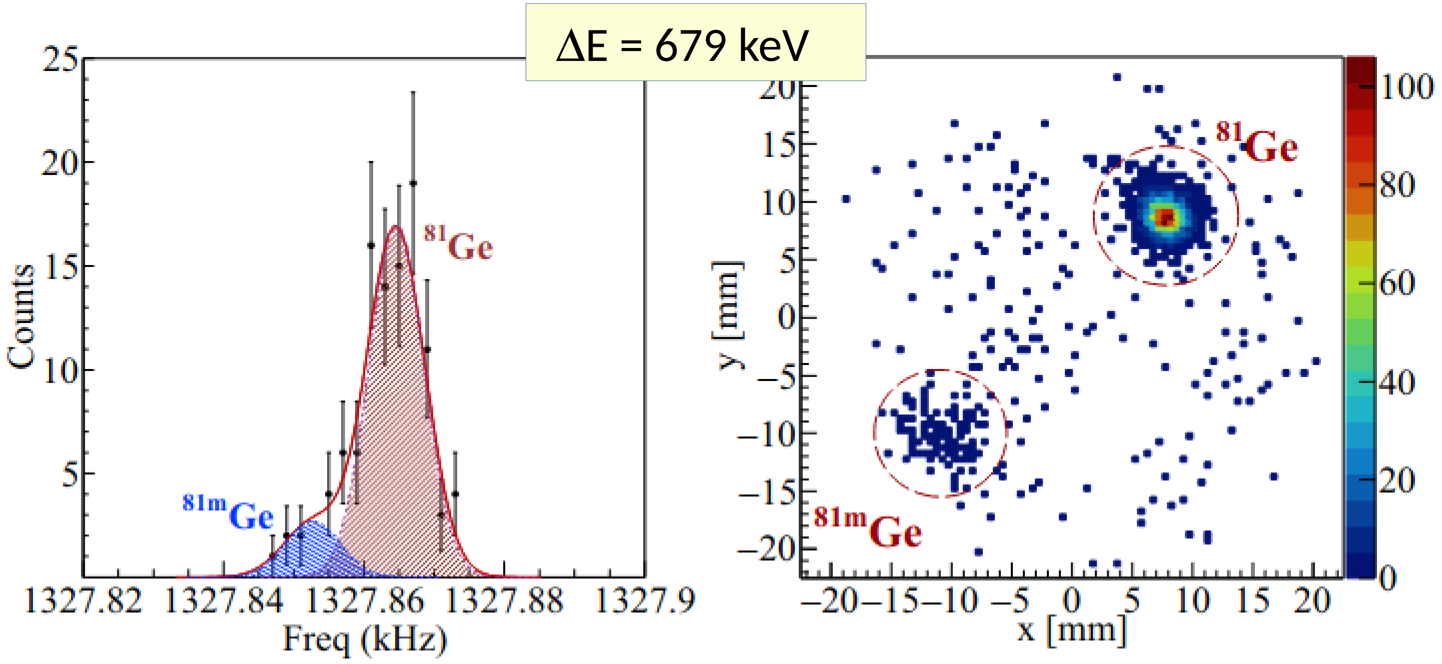

FIG. 1. The case of ^{81}Ge, observed with the sideband cooling technique (left) and with the PI-ICR technique (right). The dashed circles are used to guide the eye.

By now we have a MRP of about 10^7 and can resolve ^{129}Sn (35 keV)

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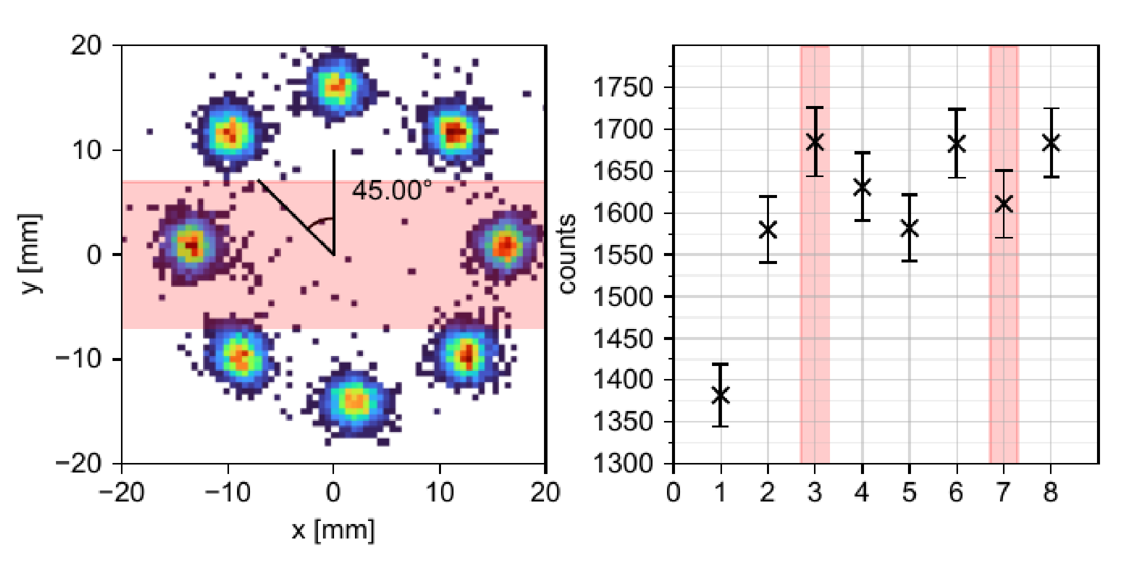
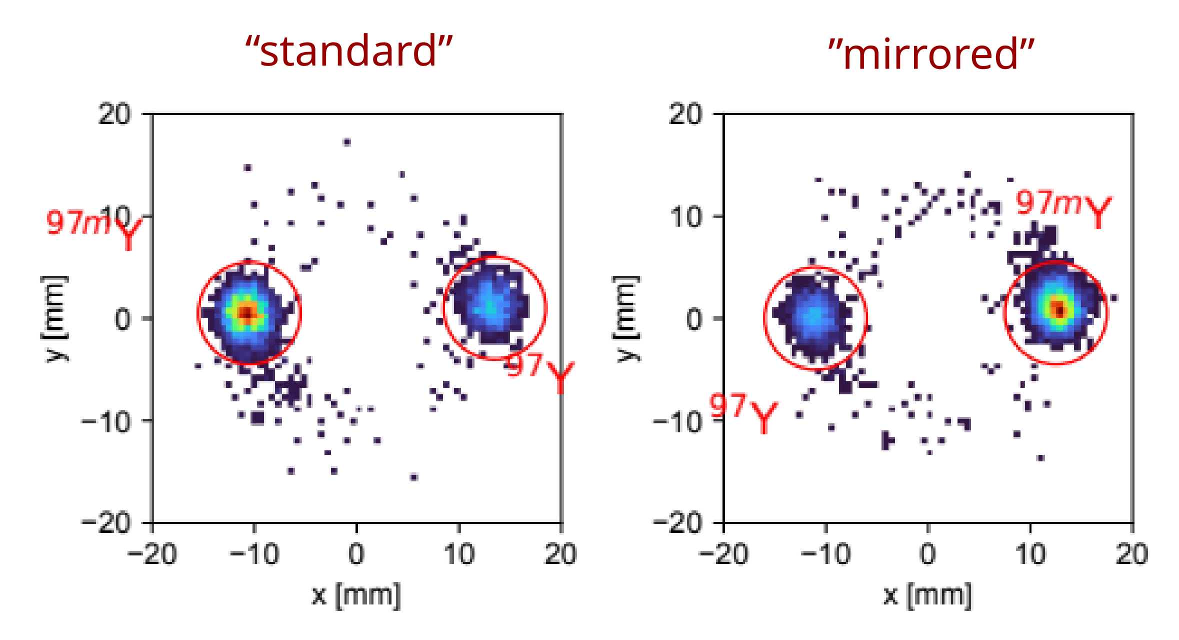
Main corrections: efficiency of the multi-channel plate (MCP) detector

Measurement of rel. detection efficiency



Stable offline ion beam impinges the MCP
(for same amount of time) at different positions.



Two positions are selected and measurements
Are repeated with flipped positions of the ion spots
To cancel out efficiency differences as much as possible.

S. Cannarozzo et al., EPJ Web Conf. **322**, 08007 (2025) (Proceedings of CNR*24)

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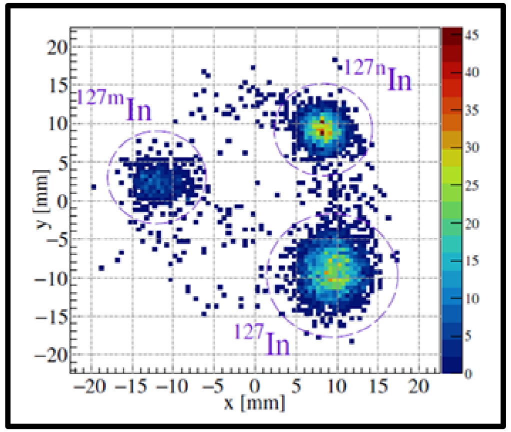
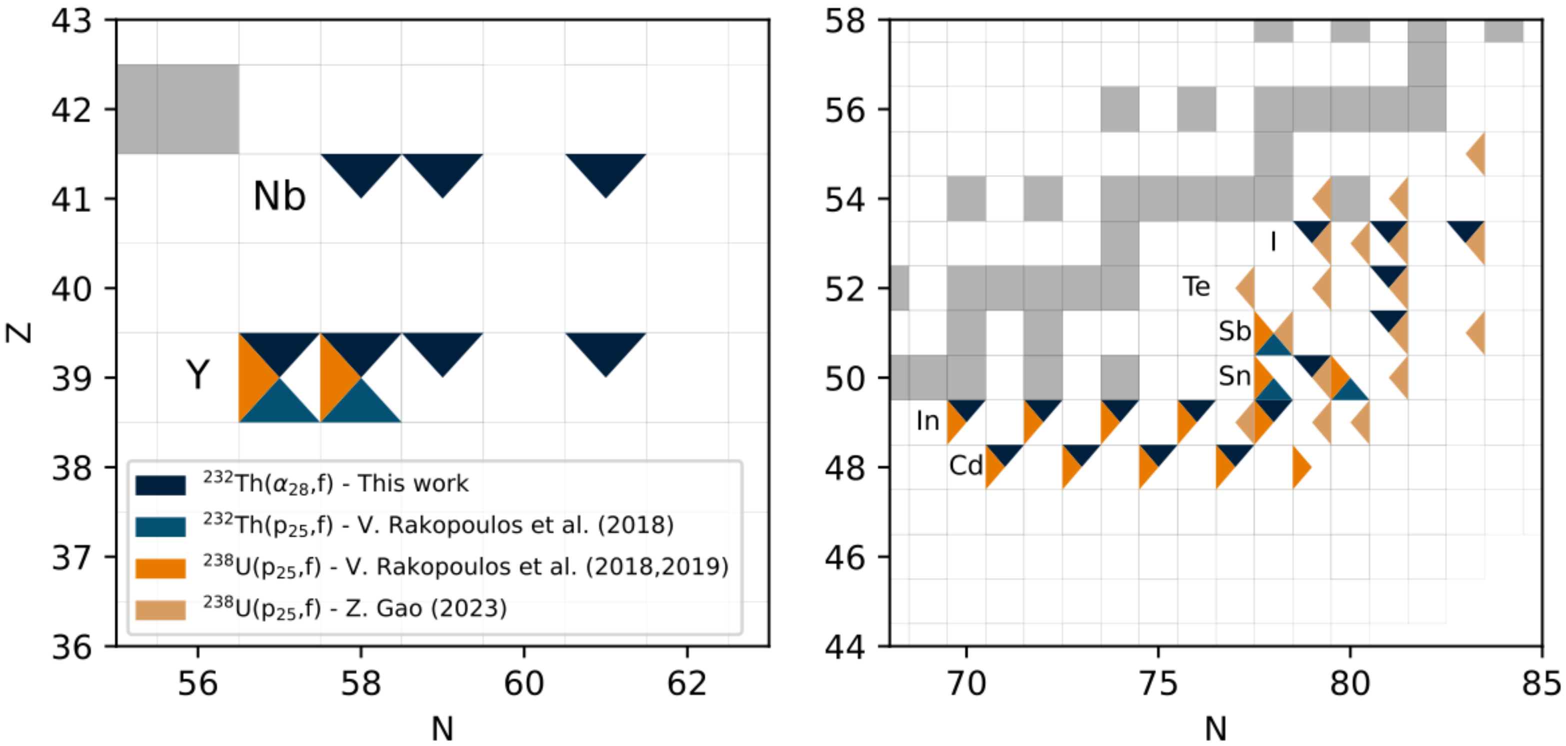
Nuclei with IYR data from IGISOL

So far 37 IYR measured; several for multiple fissioning systems:





(and ^{81}Ge)

Fig. from thesis of S. Cannarozzo, UU 2025

Several campaigns over the past years:

^{232}Th(p,f) and ^{238}U(p,f) at 25 MeV
(V. Rakopoulos, Z. Gao)

^{232}Th(α,f) at 28 MeV (S. Cannarozzo)

also some data from

^{238}U(n,f) using γ-spectrometry (A. Mattera)

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Experimental results

1) IYR from $^{232}Th(\alpha,f)$ at 28 MeV and
comparison to literature data for IYR from $^{233}U(n,f)$ and $^{235}U(n,f)$.

2) IYR from $^{232}Th(\alpha,f)$ at 28 MeV in the ^{98}Y region.

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IYR from fission using a Penning trap

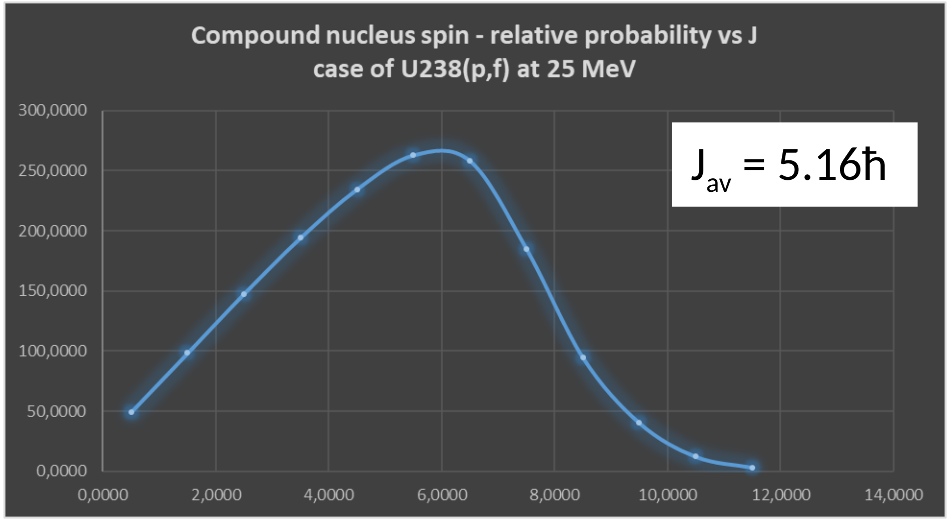
We measured many IYR ^{238}U(p,f), mainly in the ^{132}Sn region.
(incident proton energy: 25 MeV)

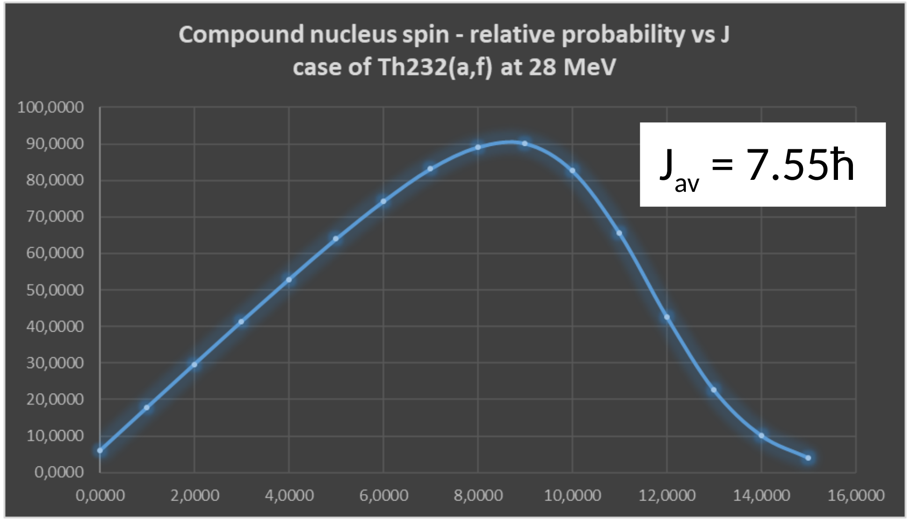
Efforts to obtain IYR from (n,f) ongoing …   (development of new ion guide ongoing)

Q: What is the impact of the angular momentum of the compound nucleus?

If we can't go down, let's go up and study ^{232}Th(α,f) and compare to IYR from ^{233,235}U(n_{th},f) !

Calculations with TALYS 1.96





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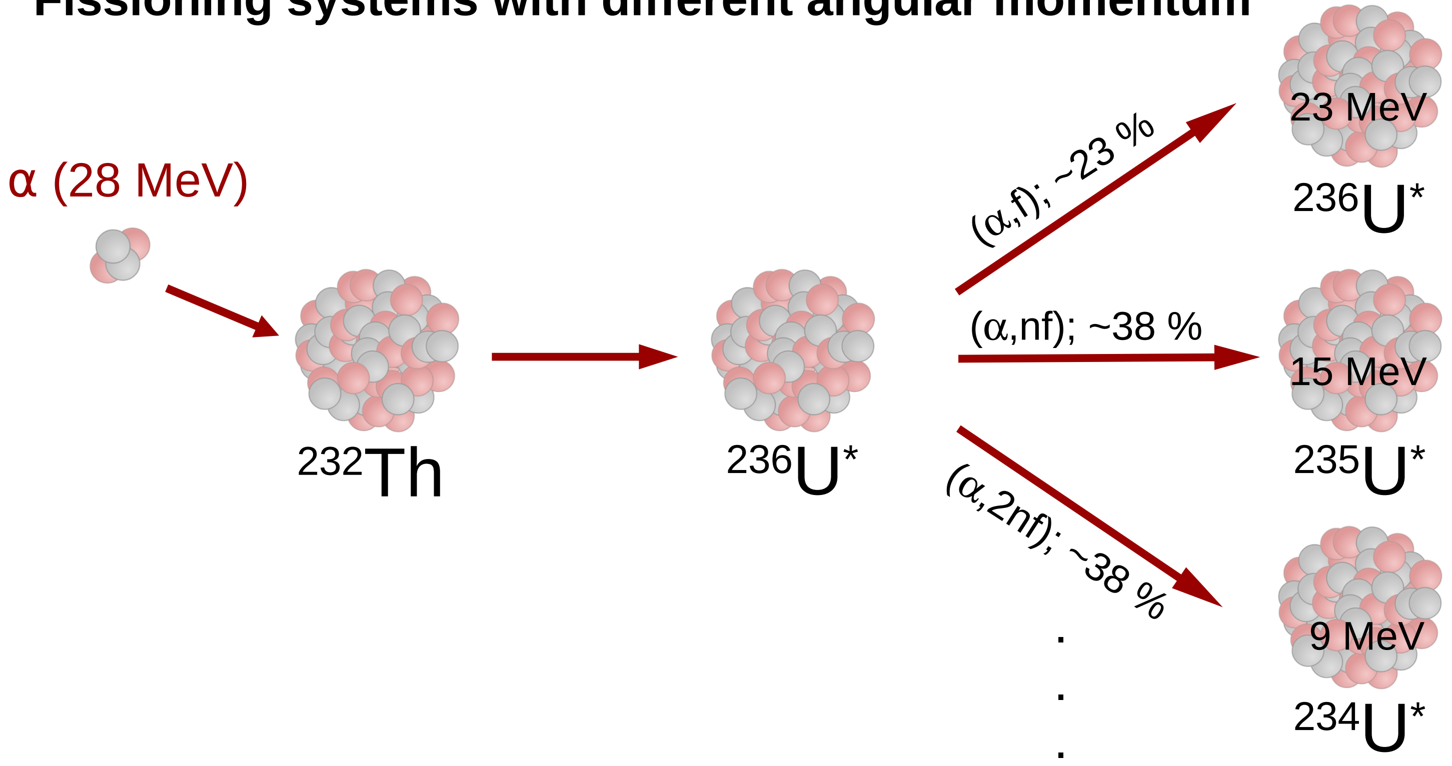
Fissioning systems with different angular momentum



Measured fissioning system

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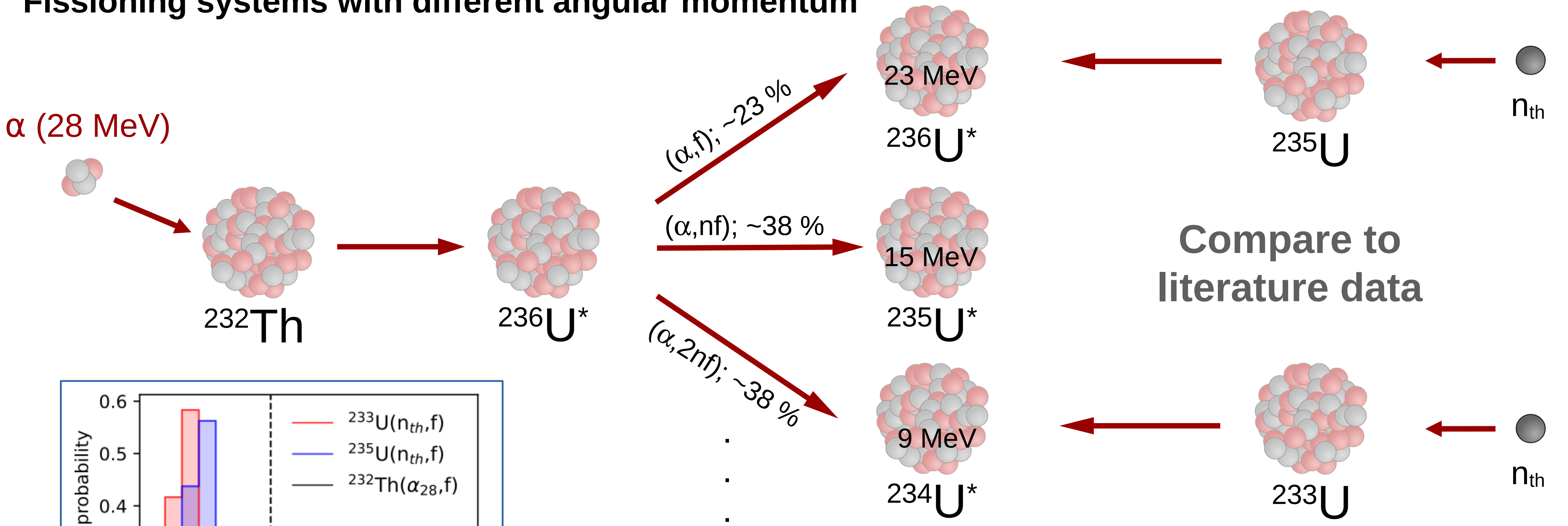
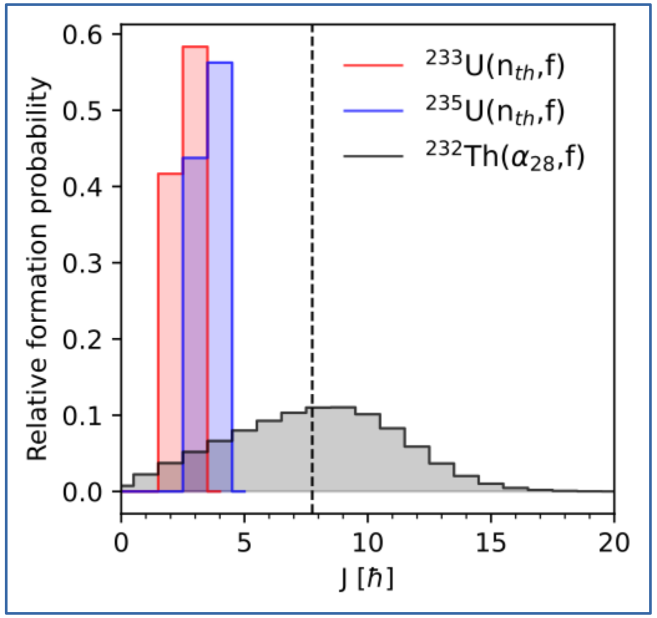
Fissioning systems with different angular momentum





Measured fissioning system

Phys. Rev. C 111, L031601 (2025)

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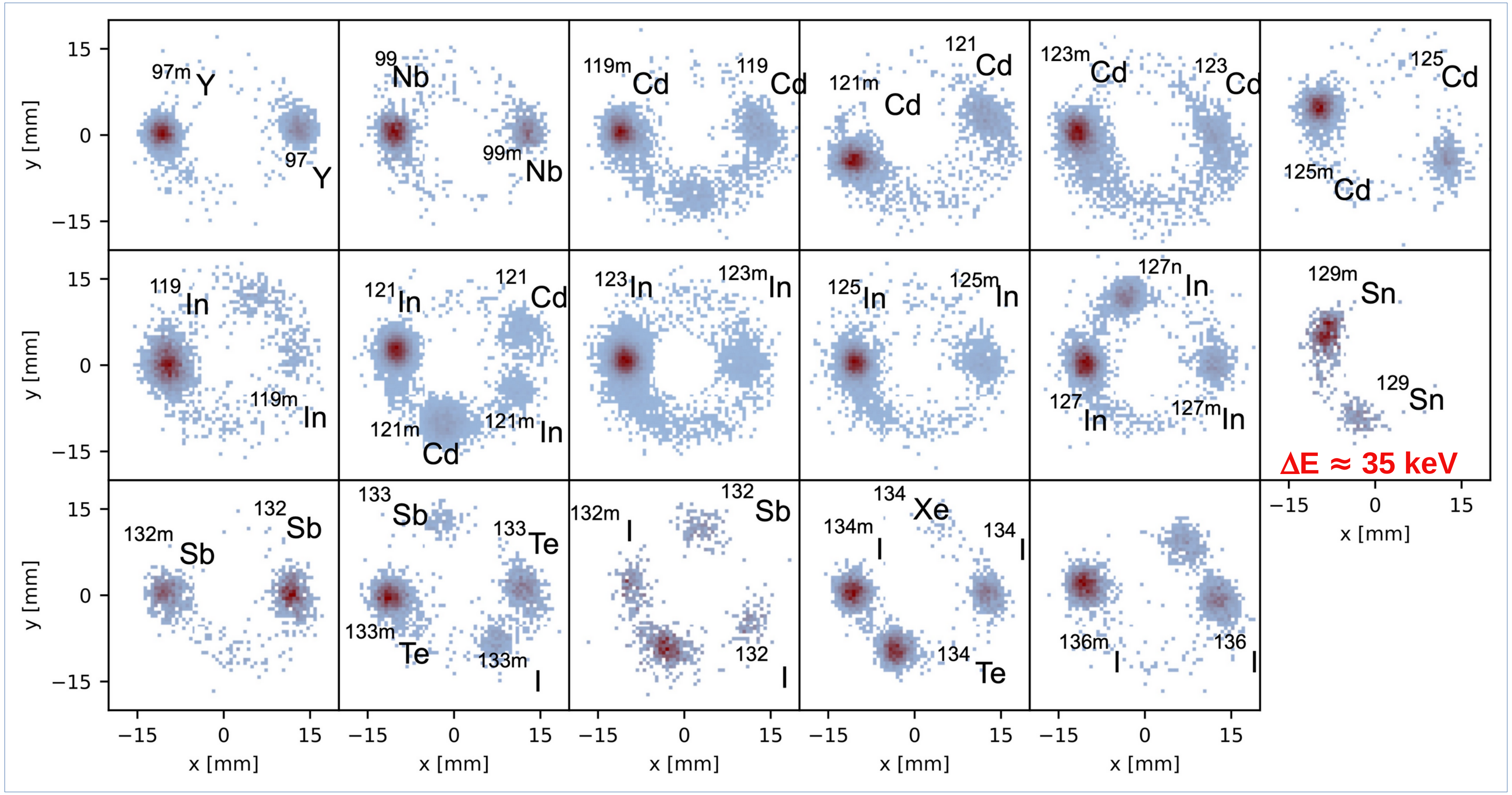
Results (1) IYR from ^{232}Th(α,f) at 28 MeV



Measured twice
at mirrored MCP positions.

Decay corrections applied.

Obtain number of counts in ground and excited isomeric state.

Calculate the IYR.
(“high-spin state over total”)

S. Cannarozzo et al., PRC **111** L031601 (2025)

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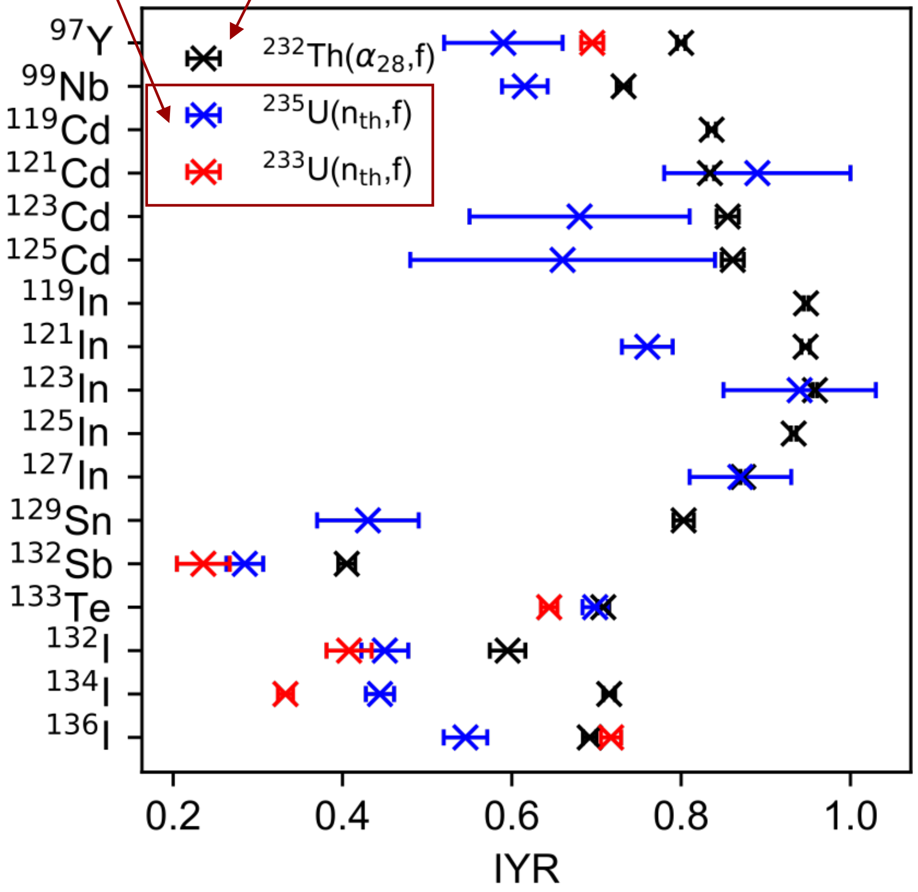
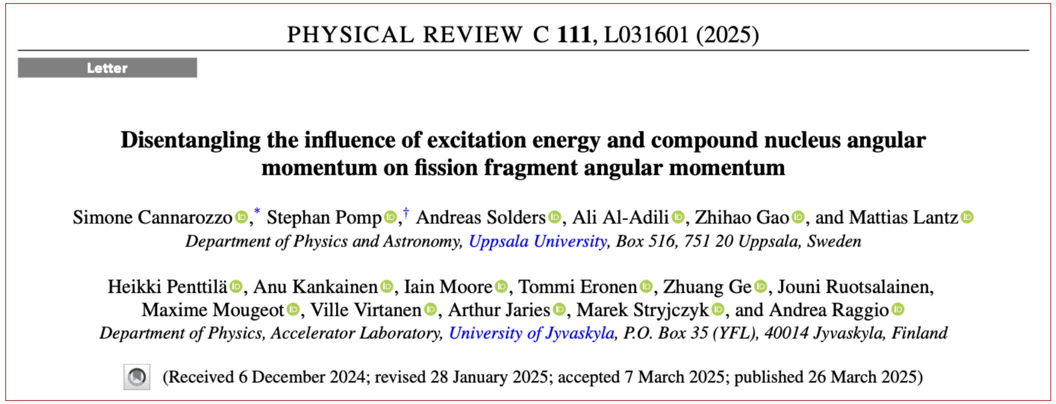
Results (1) IYR from ^{232}Th(α,f) at 28 MeV

Average of **literature values**

Our IYR data from IGISOL





PHYSICAL REVIEW C **111**, L031601 (2025)

Letter

Disentangling the influence of excitation energy and compound nucleus angular momentum on fission fragment angular momentum

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Heikki Penttilä, Anu Kankainen, Iain Moore, Tommi Eronen, Zhuang Ge, Jouni Ruotsalainen, Maxime Mougeot, Ville Virtanen, Arthur Jaries, Marek Stryjczyk, and Andrea Raggio
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(Received 6 December 2024; revised 28 January 2025; accepted 7 March 2025; published 26 March 2025)

Compound nucleus angular momentum is about 7.5 ħ.

Impact on IYR?

Compare to ^{233,235}U(n_{th},f) (with ~3 ħ). Average increased IYR.

We conclude that a significant part (at least 40%) of the extra CN spin goes to the FF.

(remaining part likely goes to orbital ang. momentum)

Could this not be due to the excitation energy?
We think not ...

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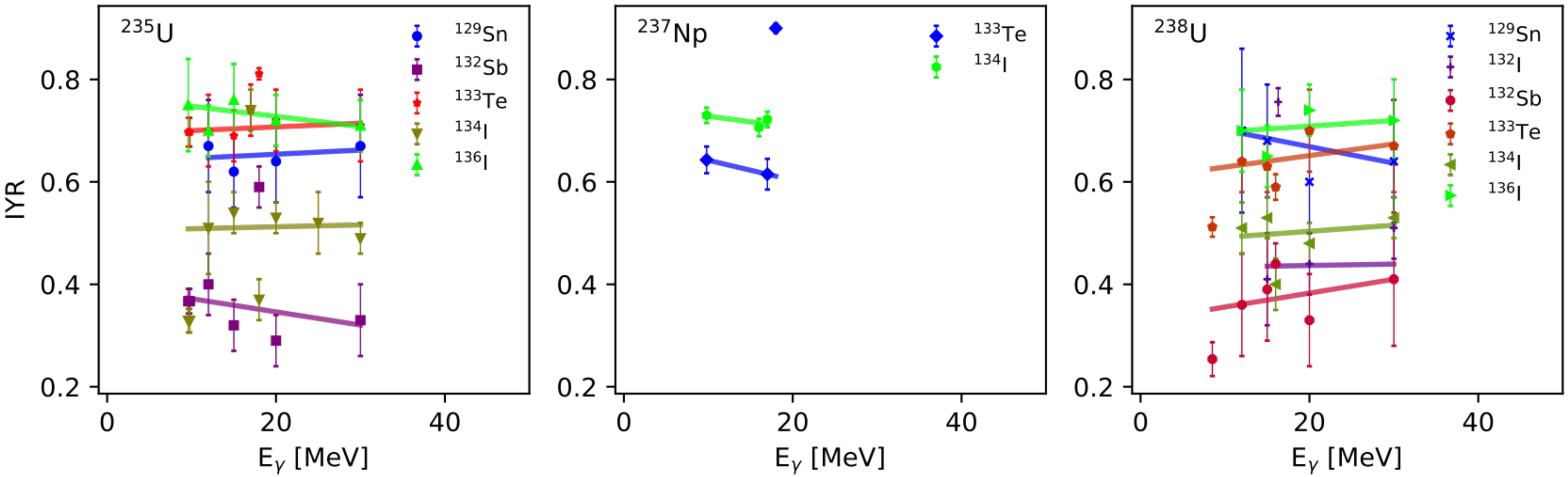
Results (1)   Impact of excitation energy?

- Photo-fission literature data was used to study influence of compound nucleus excitation energy on IYRs.



- $\Delta IYR/\Delta E = 0.0001(5)\ MeV^{-1}$ → No significant dependence of the IYRs on the incident γ-ray energy.

S. Cannarozzo et al., PRC **111** L031601 (2025)

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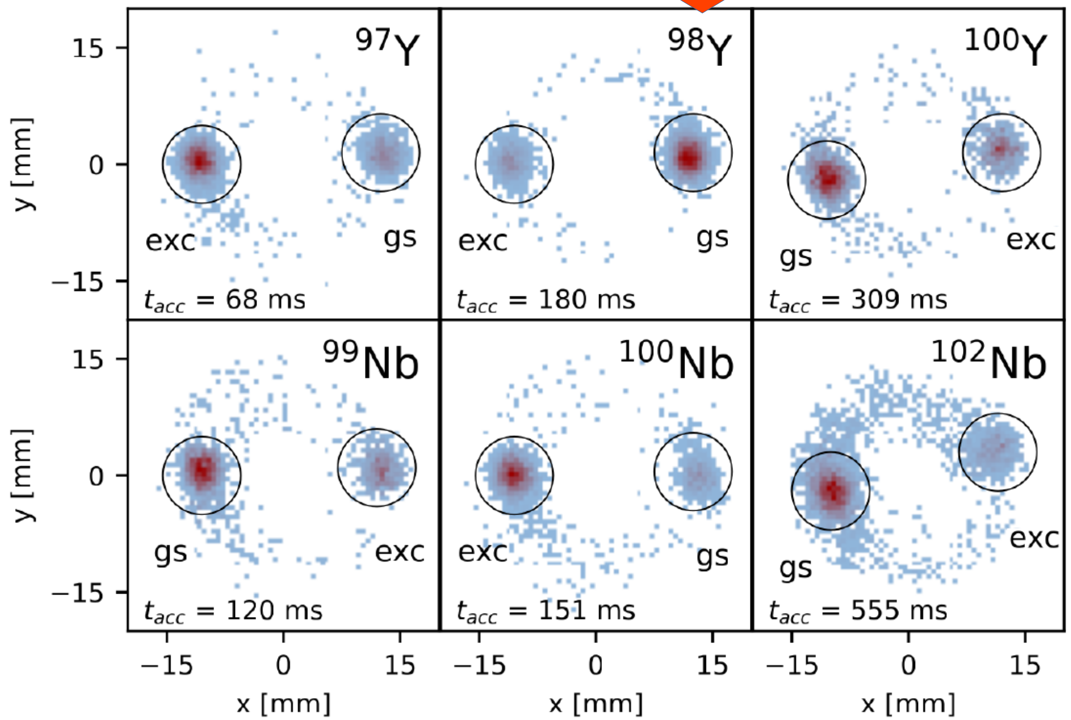
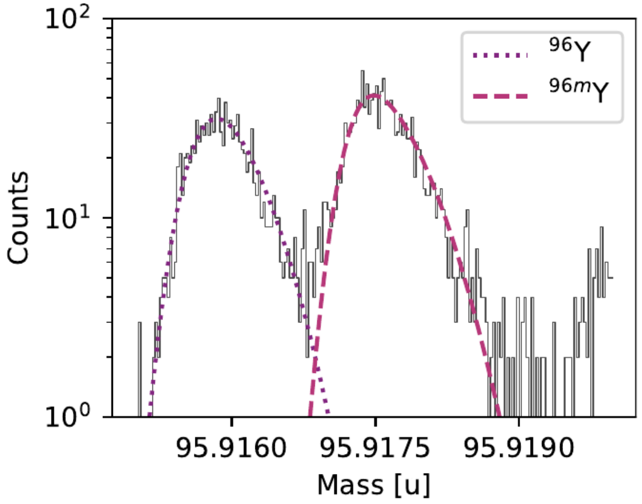
Results (2)

IYR from $^{232}Th(\alpha,f)$ at 28 MeV
The case of ^{98}Y

High spin state always on the left side in the figure



^{98}Y shows a remarkably low IYR.



Nuclide	Ground state $t_{1/2}$ (s)	Ground state I^π	Excited state $t_{1/2}$ (s)	Excited state I^π	Excited state E_x (keV)	IYR
^{96}Y	5.34(5)	0^-	9.6(2)	8^+	1540.5(4)	0.58(2)
^{97}Y (*)	3.75(3)	$1/2^-$	1.17(3)	$9/2^+$	667.52(23)	0.800(5)
^{98}Y	0.548(2)	0^-	2.32(8)	$(6^+, 7^+)$	465.7(7)	0.129(4)
^{100}Y	0.94(3)	4^+	0.727(6)	1^+	144(16)	0.727(9)
^{99}Nb (*)	15.0(2)	$9/2^+$	150(12)	$1/2^-$	365.27(8)	0.732(9)
^{100}Nb	1.5(2)	1^+	2.99(11)	(5^+)	313(8)	0.80(1)
^{102}Nb	4.3(4)	(4^+)	1.31(16)	(1^+)	94(7)	0.74(1)

State ordering of ^{98}Y, ^{100}Y, ^{100}Nb, ^{102}Nb verified.

Confirms NUBASE2020; i.e. reversed order of ^{100}Y in ENSDF.

S. Cannarozzo et al., submitted to PLB; 2504.11274v3

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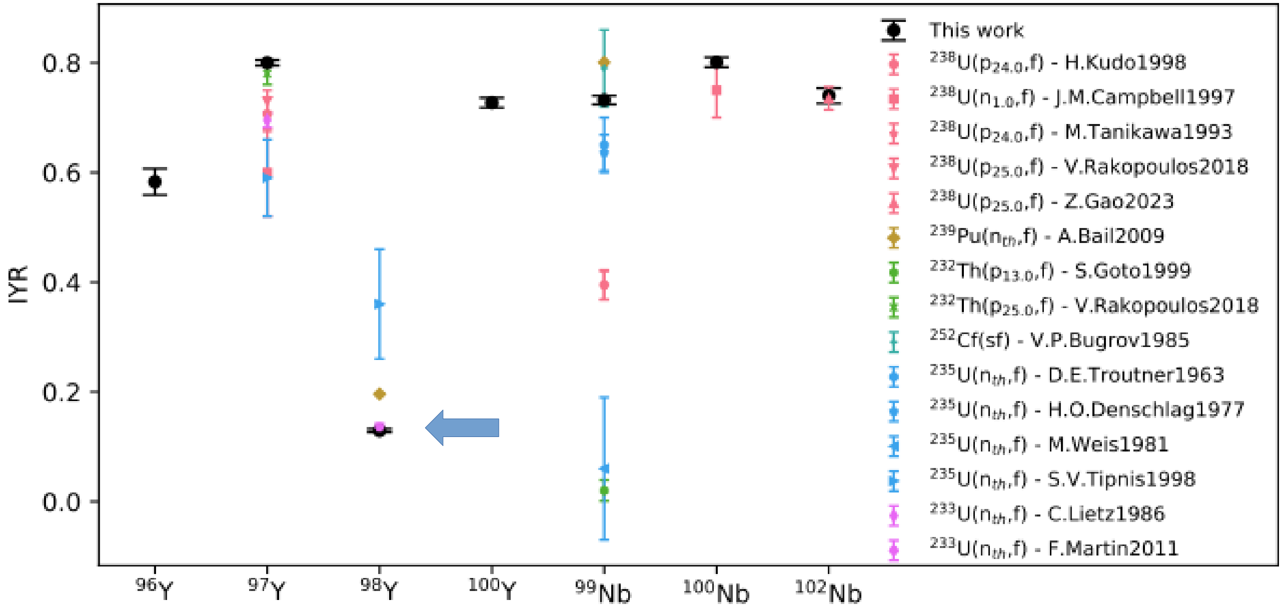
Results (2)

IYR from ^{232}Th(α,f) at 28 MeV
The case of ^{98}Y



^{98}Y shows a remarkably low IYR.

Outlier reason?
^{98}Y known for large isomeric shift
But how to explain?
State ordering experimentally verified!

	Ground state		Excited state			
Nuclide	$t_{1/2}$ (s)	I^π	$t_{1/2}$ (s)	I^π	E_x (keV)	IYR
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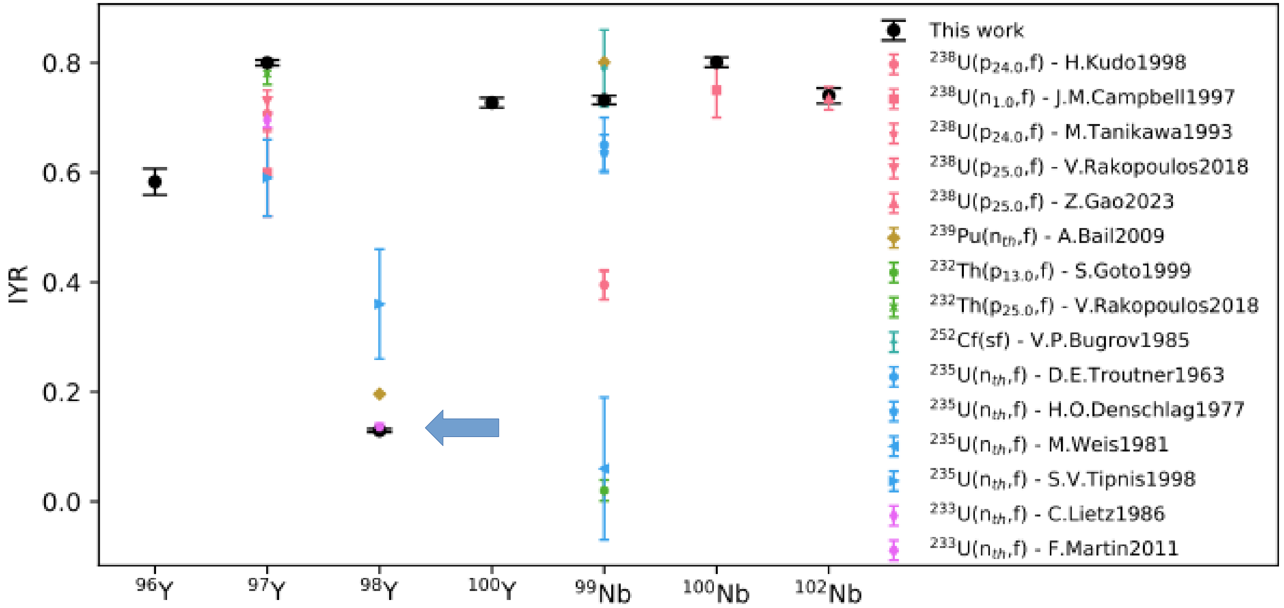
Results (2) IYR from ^{232}Th(α,f) at 28 MeV
The case of ^{98}Y



No other cases of such low IYR known.

The earlier reported low IYR for ^{114}Rh was due to wrong spin ordering.

M. Stryjczyk et al.,
“Discovery of a new long-lived isomer in ^{114}Rh via Penning-trap mass spectrometry”
Phys. Lett. B **862**, 139359 (2025).

One case with IYR ≈ 0.27 is ^{133}I, but spins of the isomeric pairs are 7/2 and 19/2.

S. Cannarozzo et al., submitted to PLB; 2504.11274v3

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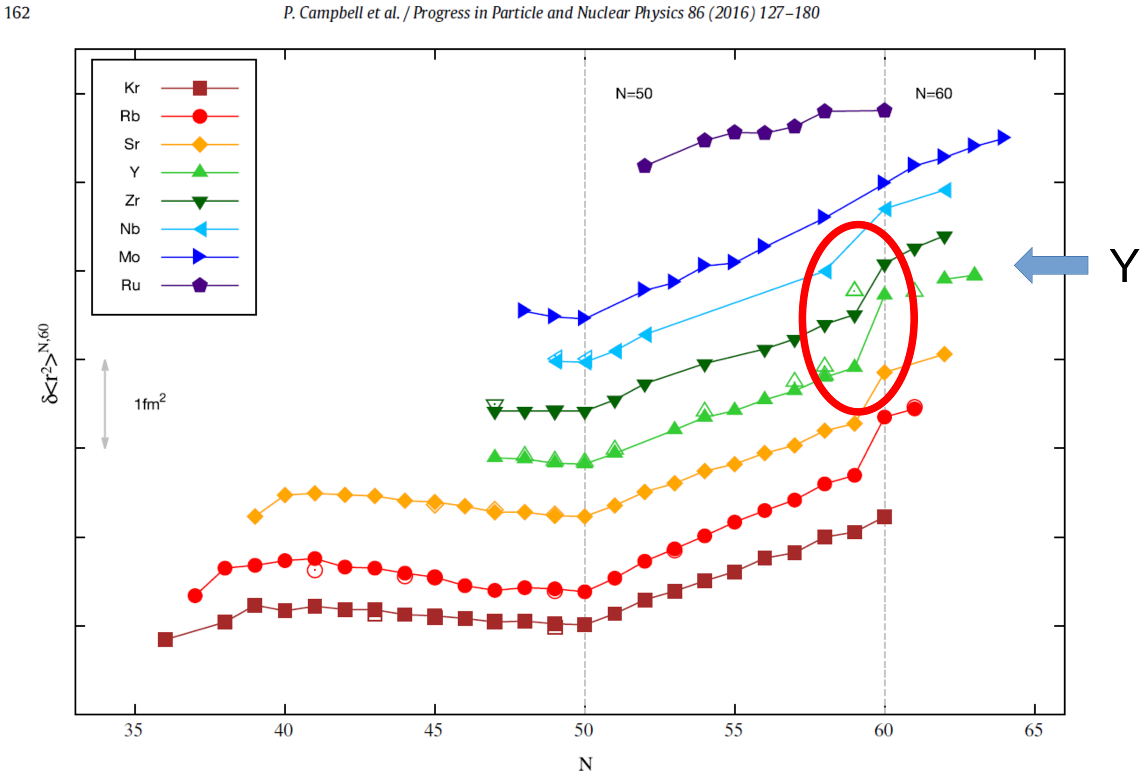
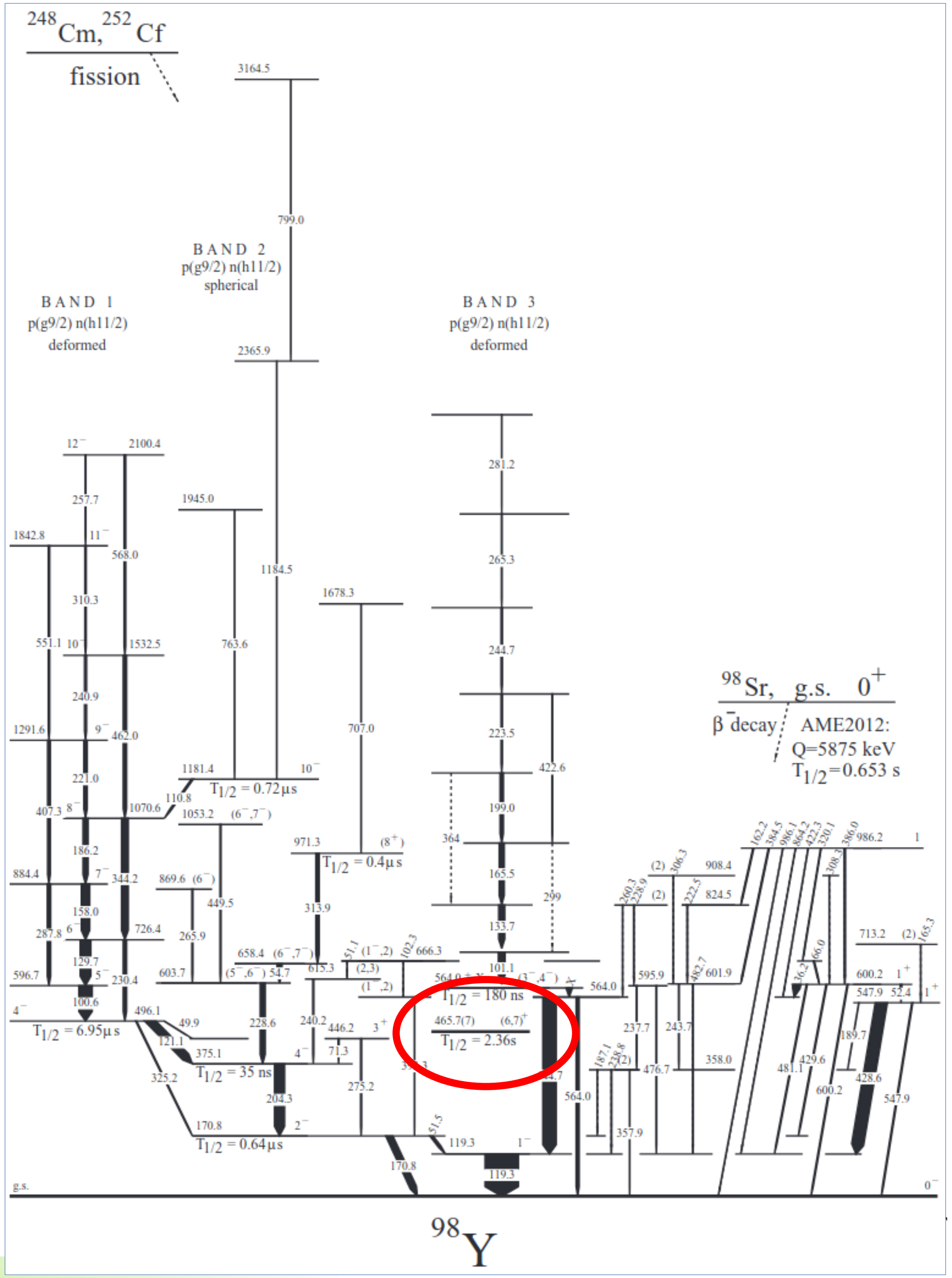
Results (2) IYR from $^{232}Th(\alpha,f)$ at 28 MeV
The case of ^{98}Y



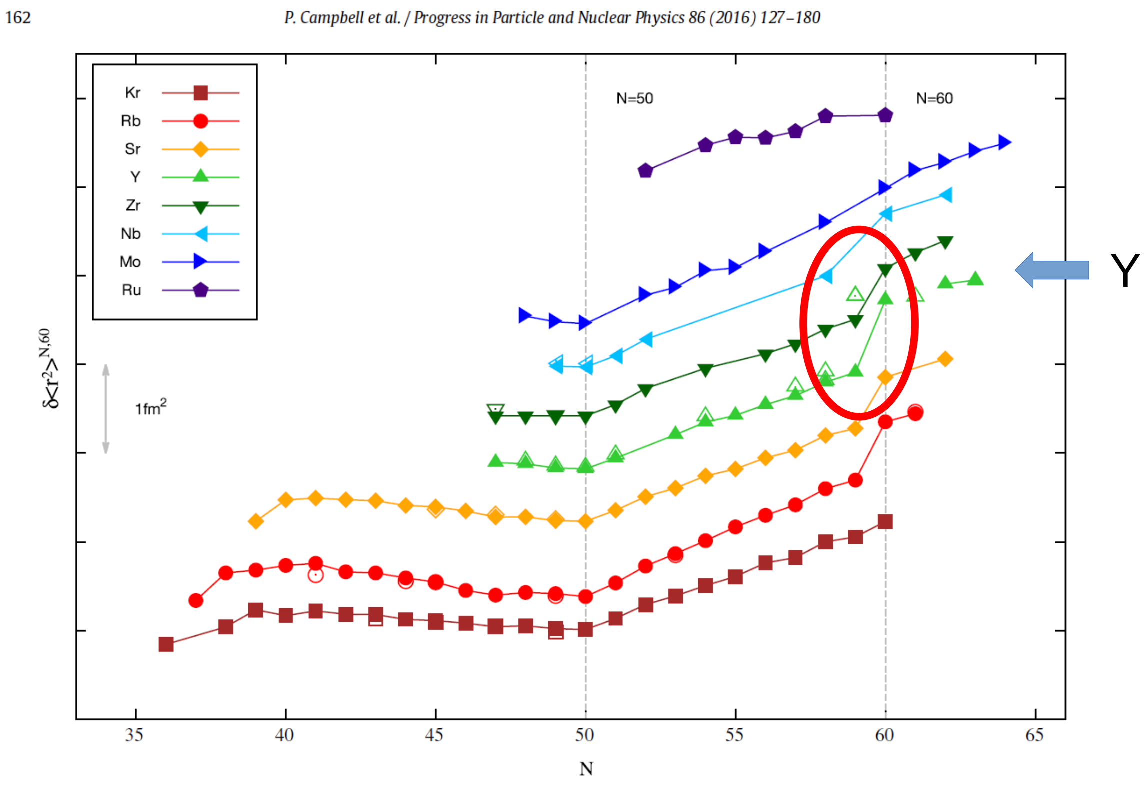
- ^{98}Y shows a large isomeric shift (also ^{79}Zn)
- Shape co-existence
- Isomer rarely populated in fission

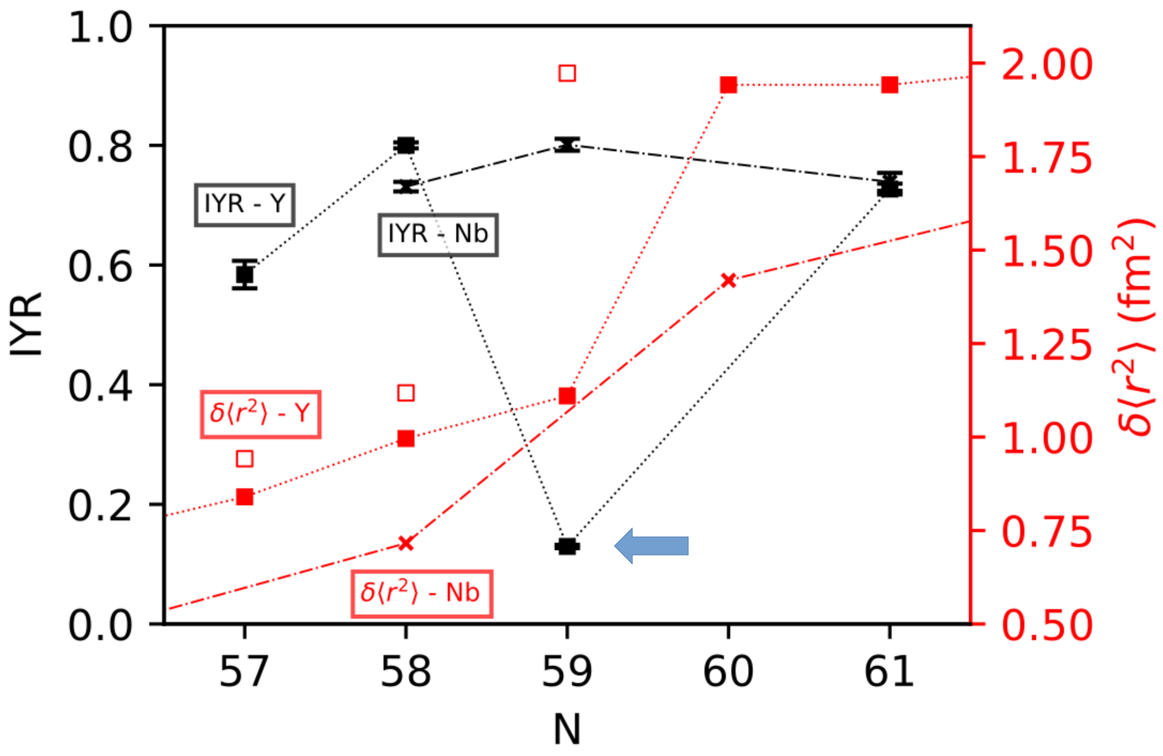


W. Urban et al., Phys. Rev. C **96**, 044333 (2017)

Results (2)

IYR from $^{232}Th(\alpha,f)$ at 28 MeV
The case of ^{98}Y





S. Cannarozzo et al., submitted to PLB

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Fission and isomers – relevance for other field

PHYSICAL REVIEW C **111**, L061601 (2025)

Letter | Editors' Suggestion

Isomeric yield ratios of fission products: A missing piece in reactor antineutrino summation calculations

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[1] *National Nuclear Data Center, Brookhaven National Laboratory Bldg. 817, P.O. Box 5000, Upton, New York 11973-5000, USA*

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(Received 7 August 2024; revised 18 December 2024; accepted 7 May 2025; published 11 June 2025)

"The **sensitivity study identified ^{98}Y**, shown in Fig. 4(b), as the fission product whose newly evaluated IYR has the greatest impact on the ν_e spectrum, accounting for an increase of over 20% at 8 MeV for ^{235}U thermal fission. This can be explained as the **experimental IYR [13.9(6)%]** recommended in Ref. [48] corresponding to a substantial inversion of the population of isomer and ground state compared to the value used in evaluated libraries and based on the **Madland & England model, which predicts a IYR of 81%.**" (my emphasis)

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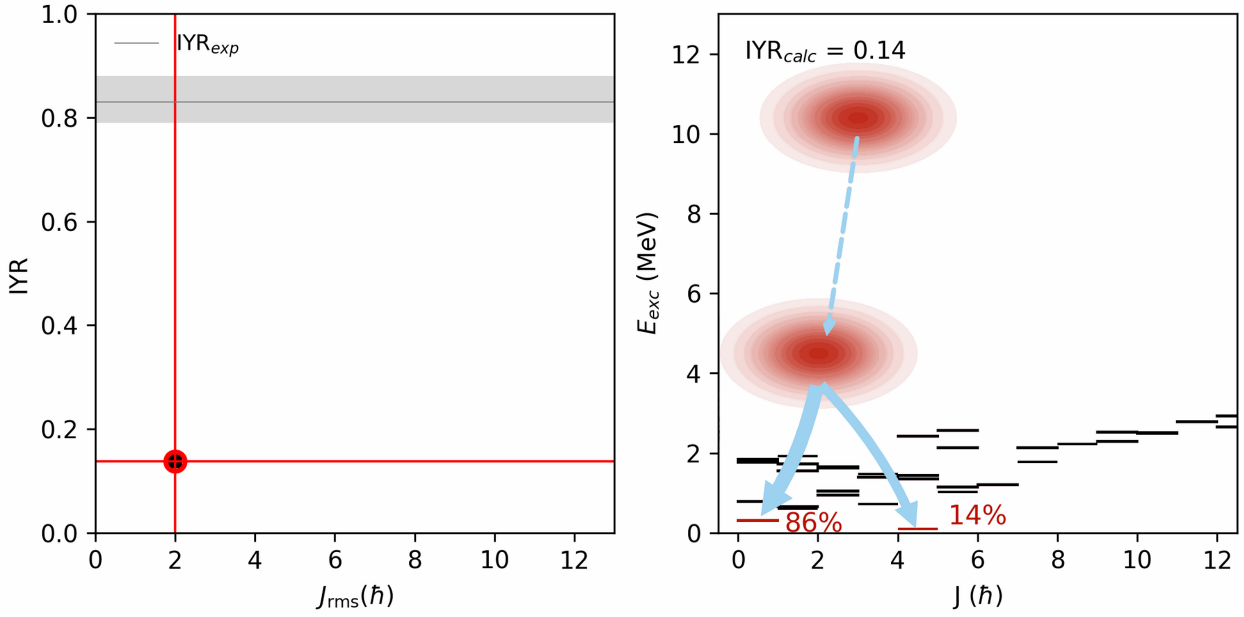
From IYR to fission fragment angular momenta (VERY brief)

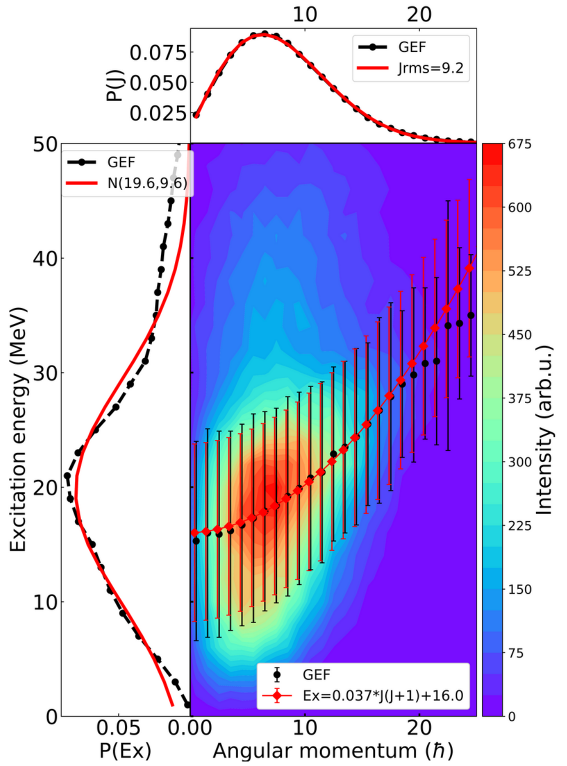


Animation by Simone Cannarozzo

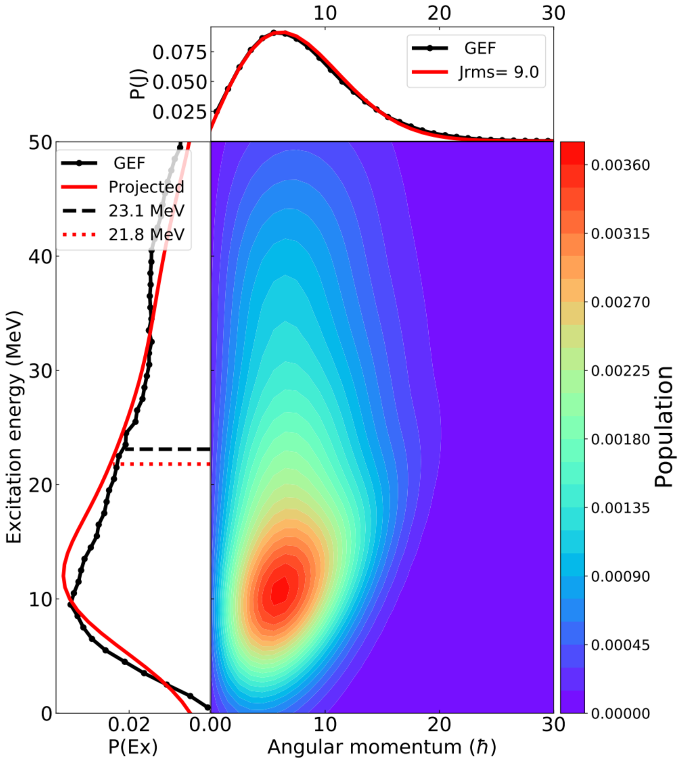
Angular momentum estimation – Example of ^{133}Te

Just to give you a rough idea.
(details would need another talk ...)

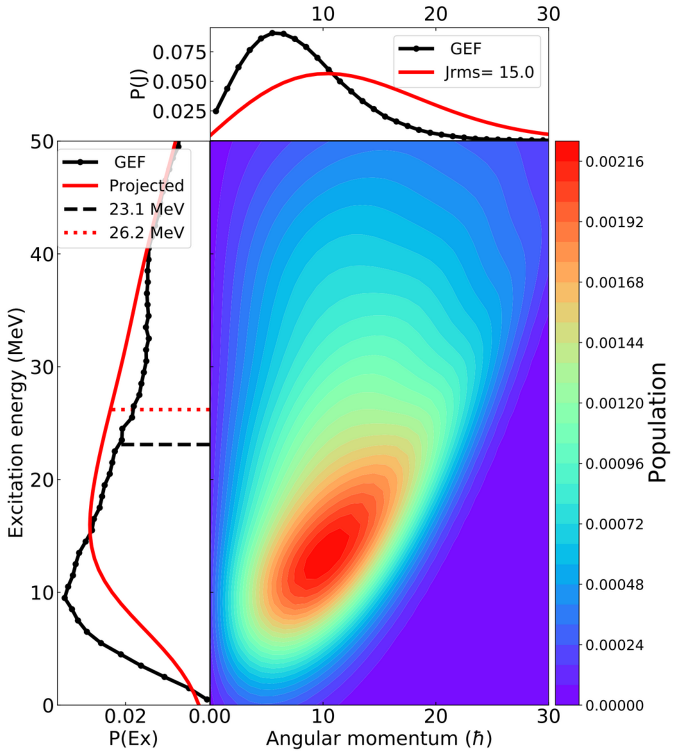
Fit to GEF data for each fission channel.



Weighted sum of matrices for 'all' fission channels.



Creation of new matrices for any J_{rms}



Z. Gao et al., PRC **109**, 064626 (2024)

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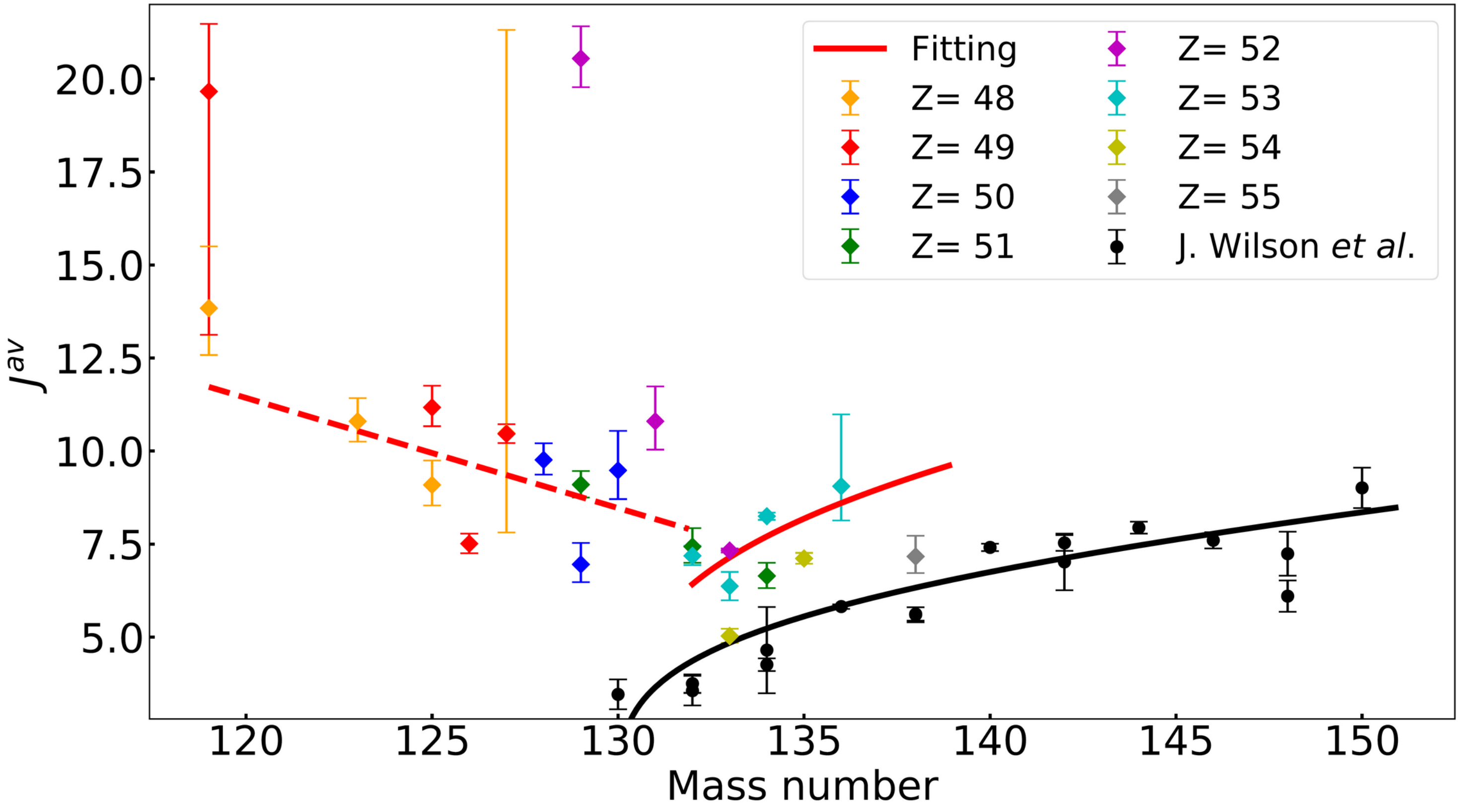
Comparing derived Jrms from ^{238}U(p,f) at 25 MeV with the data from Wilson et al.

- Comparison to Wilson data for $^{238}U(n_{th},f)$.

  J. Wilson et al., Nature **590**, 566 (2021)

- Overall larger average angular momenta. ($Supports our ^{232}Th($\alpha$,f) study)

- Minimum angular momentum: A ≈ 130.



Gao et al., Phys. Rev. C 109, 064626 (2024)

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Summary and outlook

Over the past years **we measured IYR for 37 nuclei at IGISOL**.

Recently we studied **IYR from ^{232}Th(α,f)** and show that:

1) Increased **CN angular momentum** → **larger IYR**
i.e. added CN angular momentum leads to higher fission fragment angular momentum

2) Using literature data, we find **no significant dependence on excitation energy**.

3) We highlight the **unusual IYR for ^{98}Y** (and establish/verify state ordering of 4 Y and Nb isotopes).

Furthermore:

4) The IYR can be used to **derive J_{rms}** of the fission fragment (model dependent).

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Summary and outlook

Plans:

A) Additional IYR measurements from **^{232}Th(α,f)** at IGISOL in the **symmetry and low mass** region.

B) IYR from spontaneous fission of the 0^+ nuclei **^{248}Cm and ^{252}Cf** FRS-CSC at GSI, Germany.

C) Continue development of **methods to derive fragment angular momenta** from the IYR.

D) Develop a **new ion guide** with increased efficiency to allow for measuring **(n,f)** at IGISOL.

In this context the output from the new **IAEA CRP on nuclear level densities** will be valuable.

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Ackowledgements

Results presented here are based on the PhD theses of
Andreas Mattera (2017), **Vasileios Rakopoulos** (2018), **Zhihao Gao** (2023), **Simone Cannarozzo** (June 10, 2025)

All work is done in close collaboration with the **IGISOL group** of the University of Jyväskylä.
We acknowledge the staff of the Accelerator Laboratory of University of Jyväskylä (JYFL-ACCLAB) for providing excellent beams.

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JYVÄSKYLÄN YLIOPISTO
UNIVERSITY OF JYVÄSKYLÄ

Thank you!

Vetenskapsrådet

Suomen Akatemia

Strålsäkerhets myndigheten
Swedish Radiation Safety Authority

SKC
Swedish Centre for Nuclear Technology

SANDA
Supplying Accurate Nuclear Data for energy and non-energy Applications

HORIZON2020

Accelerator and Research reactor Infrastructures for Education and Learning
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