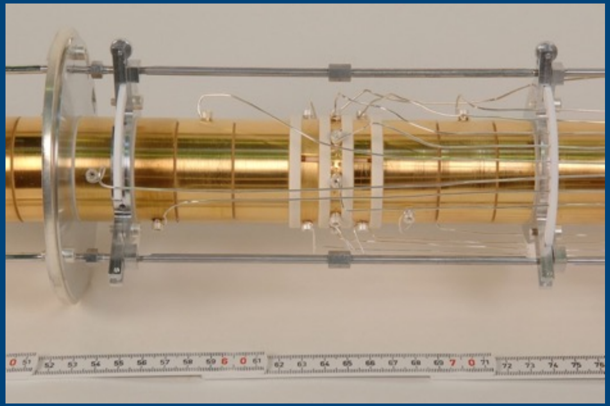
Recent Results from Penning-Trap Mass Measurements of Radionuclides



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GEFÖRDERT VOM Bundesministerium für Bildung und Forschung

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by the SHIPTRAP collaboration, and by the GSI accelerator

and technical departments are gratefully acknowledged!

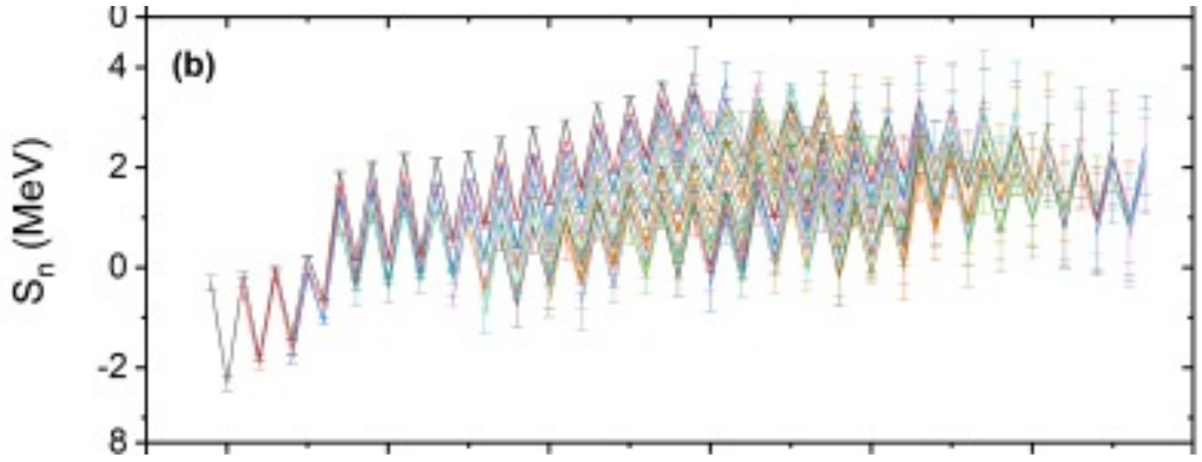
Outline

- motivation
- techniques in Penning-trap mass spectrometry
- select results from recent mass measurements at different facilities
  - In mass measurements at ISOLTRAP
  - Zr mass measurements at LEBIT
  - mass spectrometry of heavy nuclei with SHIPTRAP

Mass differences reveal nuclear structure

nucleon-separation energies reflect changes in binding energy due to structure

- neutron-separation energy $S_n(N,Z) = M(N,Z) – M(N\text{-}1,Z)$
- proton-separation energy $S_p(N,Z) = M(N,Z) – M(N,Z\text{-}1)$

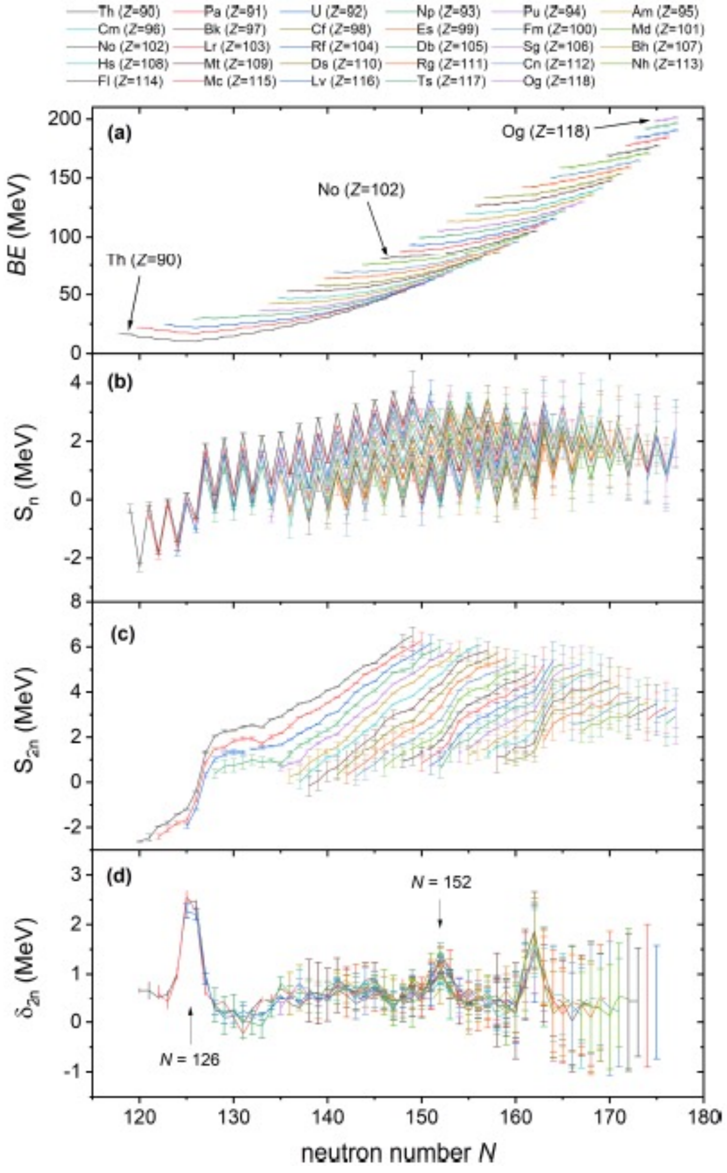


120 150 180

Neutron number

odd-even staggering reflects pairing

Mass differences reveal nuclear structure



- nuclear mass differences such as one/two-nucleon-separation energies reflect nuclear structure effects
- signatures of shell closures, pairing, and the onset of deformation
- comparison of data to nuclear models provides valuable information

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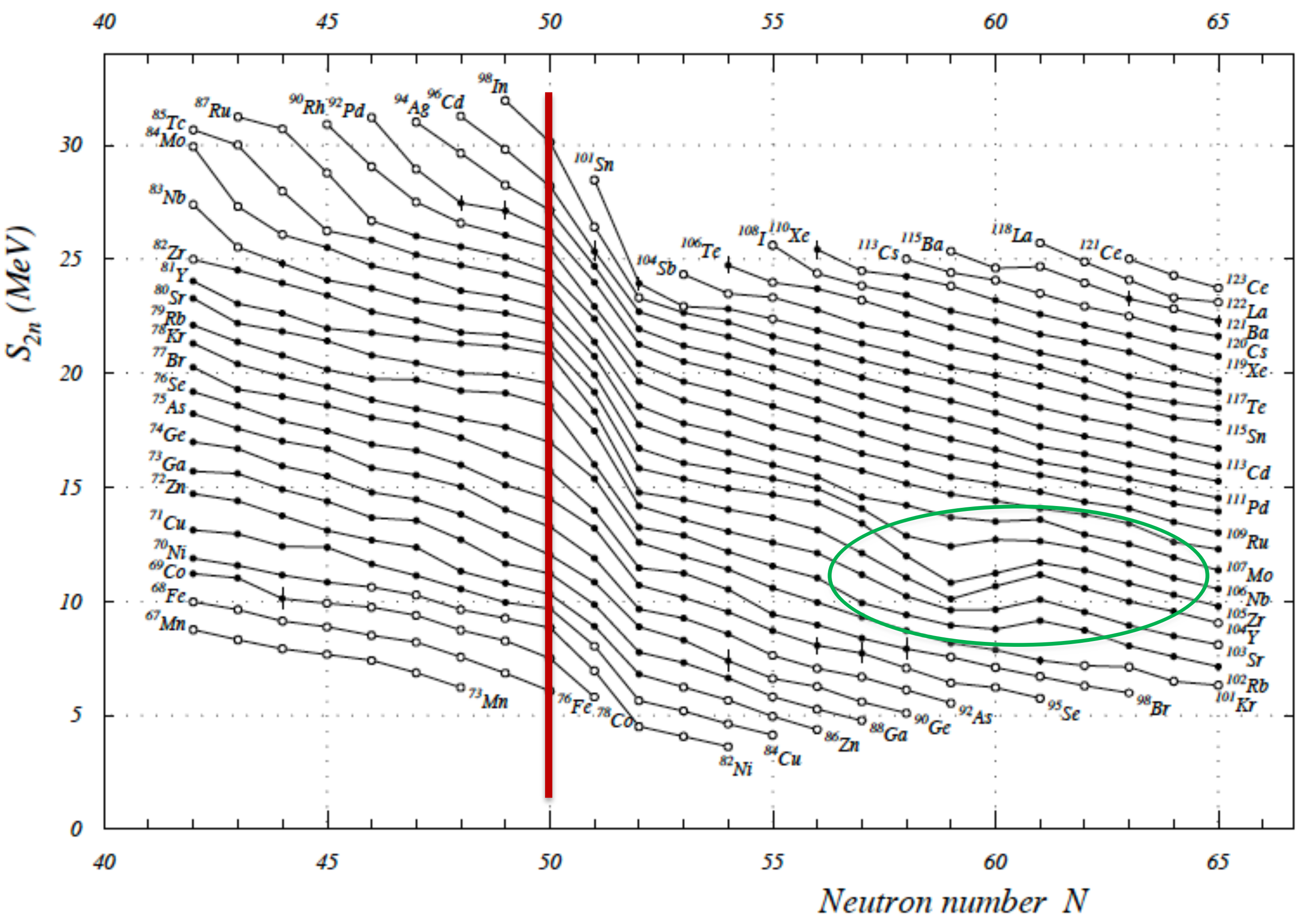
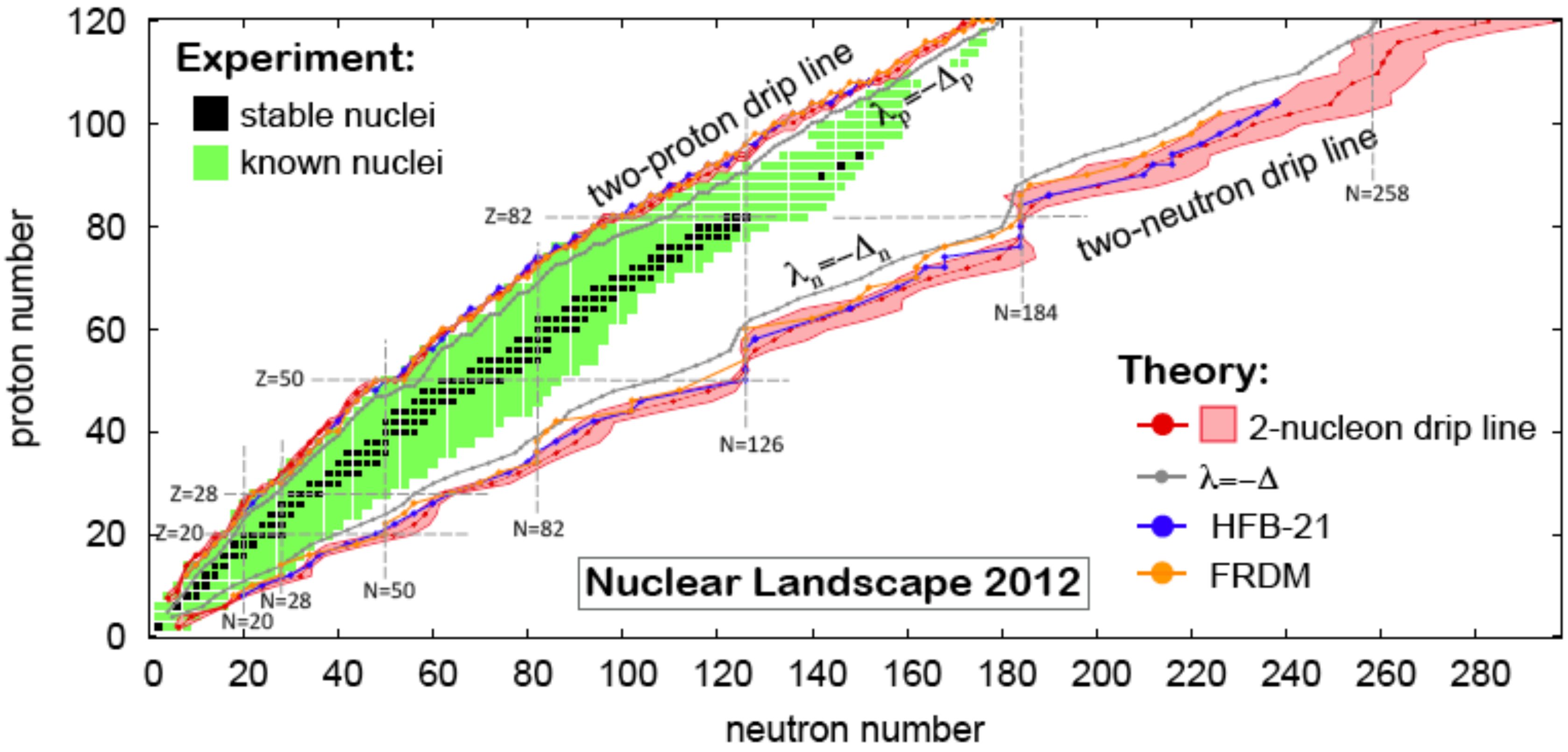
Nucleon-Separation energies and nuclear structure



Example (Data from AME 2020):

- two-neutron separation energies for various elements for N=40-65
- shell closure at N=50
- region of deformed nuclei around N=60

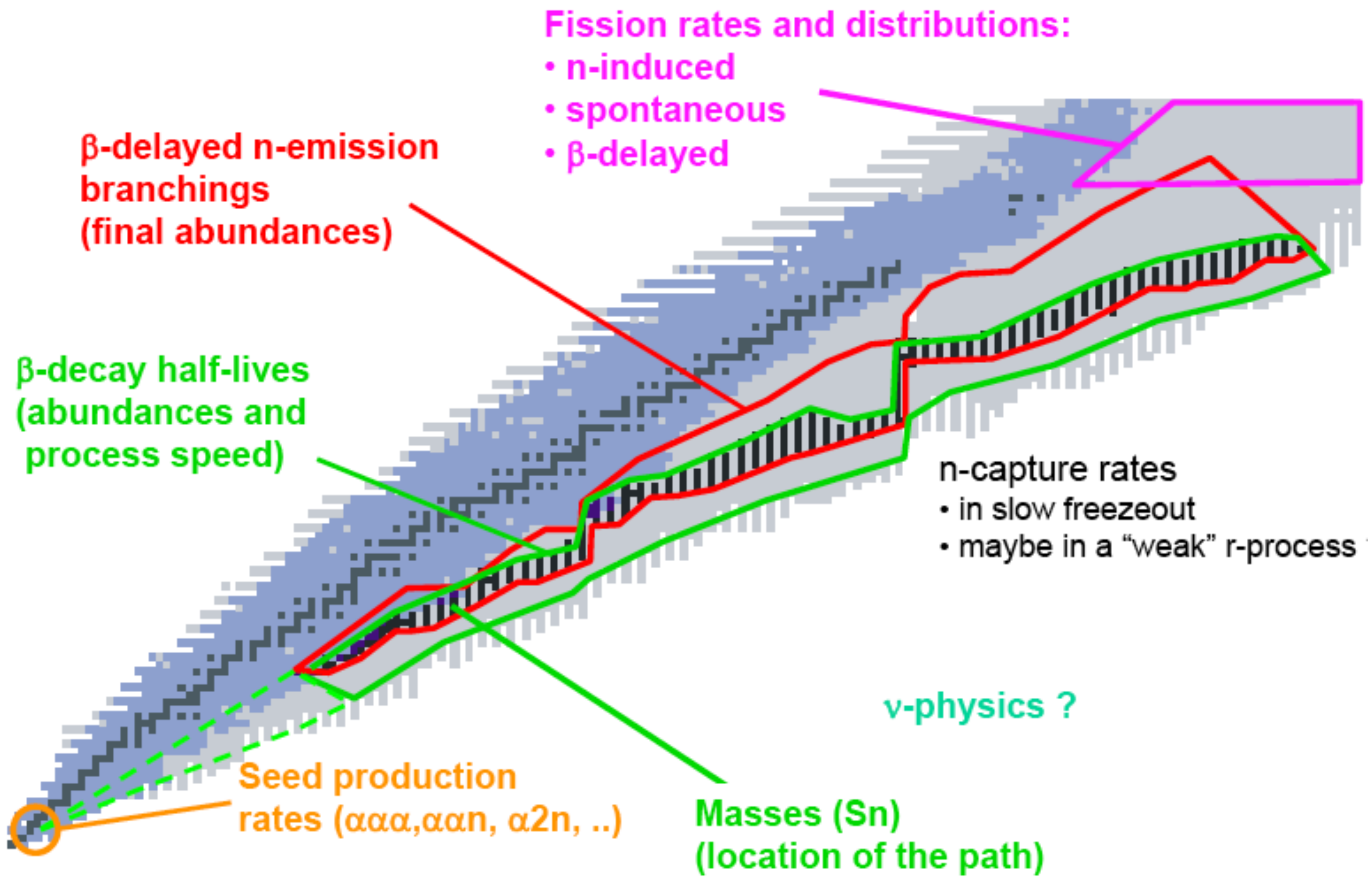
Exploring the Limits of the Nuclear Chart



- Prediction:
  about 7000 nuclides exist
- nucleon separation energies (masses) define drip lines
- Nuclear chart is shaped by nature of underlying forces

J. Erler et al., Nature 486, 509 (2012)

Nuclear Astrophysics



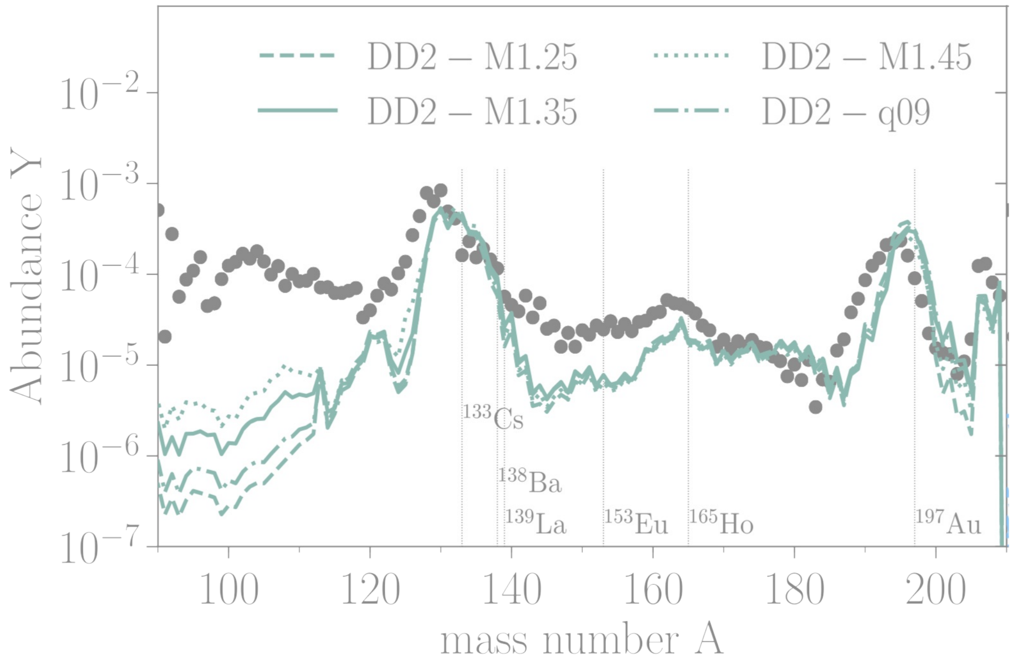
Production of SHE?

Courtesy H. Schatz

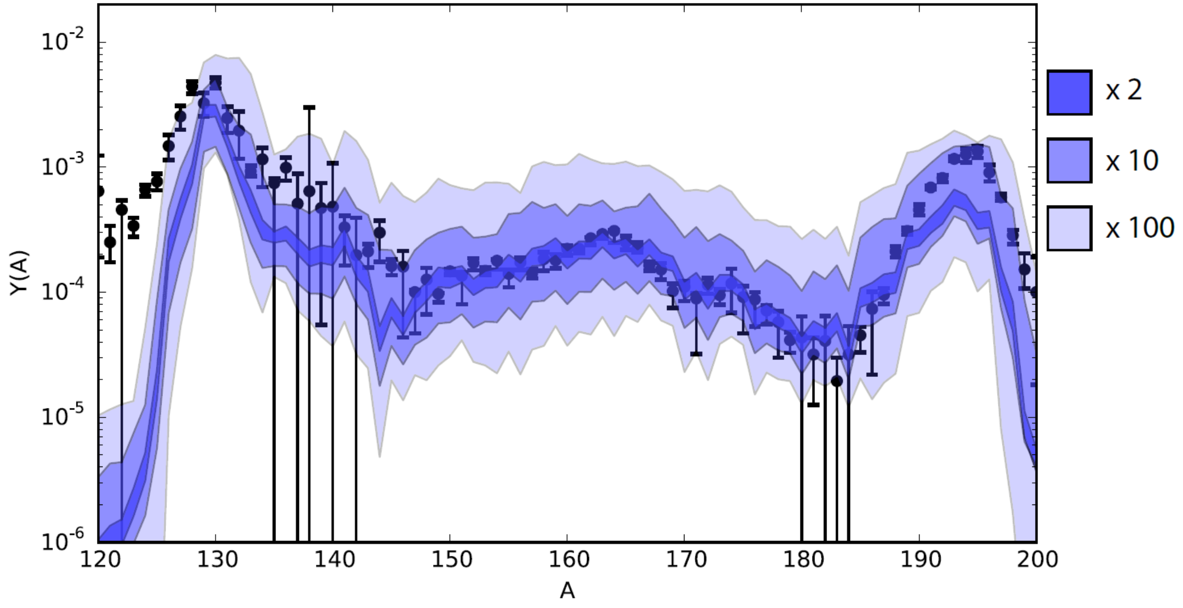
Einfluss Neutron-induzierter Reaktionen im r-Prozess

Impact of masses and (n,γ)-rates on the elemental abundances in r process can be studied by model predictions

Masses



(n,γ)-Rates



Nikas et al (2020) PRC, submitted, arxiv

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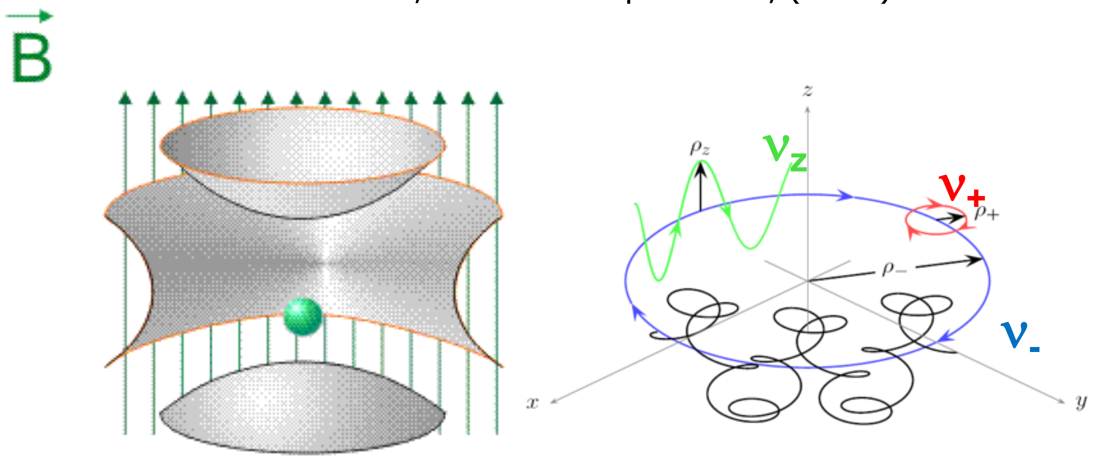
Courtesy R.Reifarth

Direct Mass Measurement Techniques

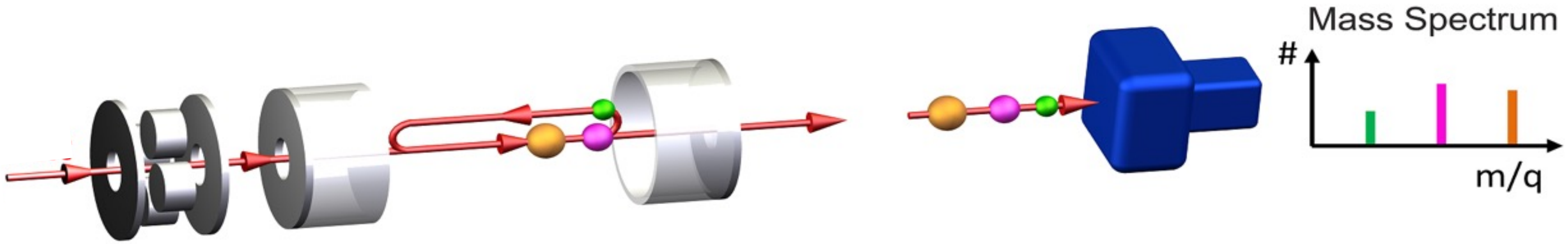
Frequency-based mass measurements,
mainly in Penning traps

cyclotron motion $\omega_c = qB/m$

L. S. Brown and G. Gabrielse, Rev. Mod. Phys. 58 (1986) 233
G. Gabrielse, Int. J. Mass Spectr. 279, (2009) 107



Time-of-flight mass spectrometry,
nowadays with MR-ToF



H. Wollnik et al., Int. J. Mass Spectrom. Ion Processes 96 (1990) 267

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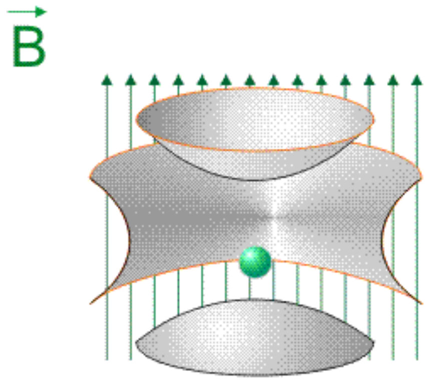
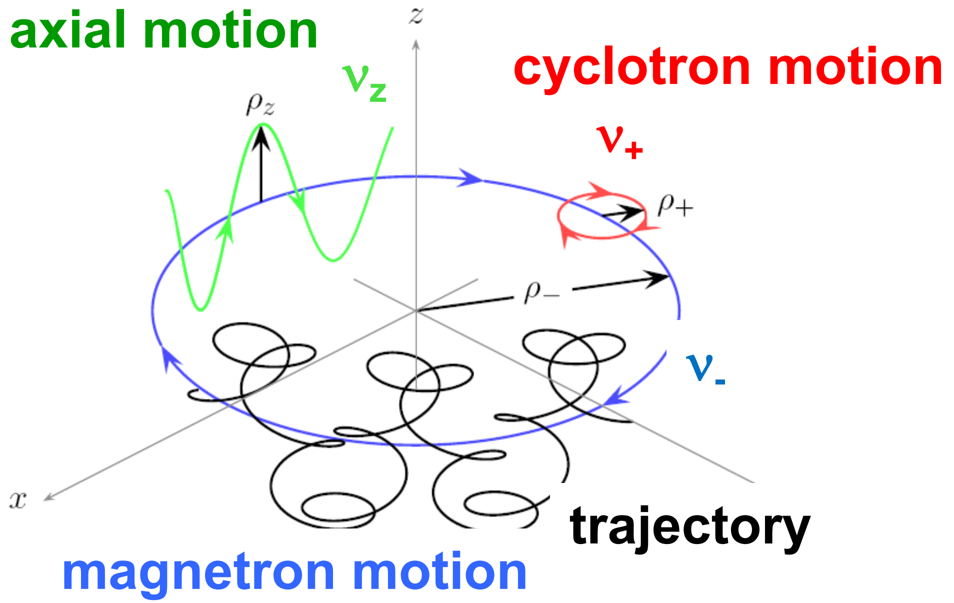
Penning Trap Mass Spectrometry





$$f_c = \frac{1}{2\pi} \cdot \frac{q}{m} \cdot B$$

- confine ion with mass *m* and charge *q* by combination of homogenous *B* field and electrostatic quadrupole field in vacuum
- measurement of cyclotron frequency yields mass value
- magnetic field calibration by reference ions with well-known mass

Complementarity of Penning Traps

Production	ISOL TRAP	SHIP TRAP	JYFL TRAP	CPT	LEBIT	TITAN	TRIGA TRAP	MATS	MLL TRAP
ISOL	X					X			
Fusion-Evaporation		X	X	X					X
Projectile Fragmentation					X			X	
induced fission			X				X		
spontaneous fission				X					X
Highly-charged ions						X			

Schematic of a Mass Spectrometer for In-flight Production

≈ 10-100 MeV → ≈ 1 eV

	Slowing down	Cooling, bunching, stacking	Measurement	Follow-up exp Decay Spec
	Buffer Gas Stopper	RFQ cooler buncher	Penning trap MR-ToF	
Efficiency	≈ 50 %	≈ 80 %	≈ 50 %	
Time	10-100 ms	10 ms	10 ms – 1 s	

Typical Performance and Reach for Rare Isotopes

- required yield: ≈ few particles per hour (SHIPTRAP 1 / day)
- accessible half-life ≈10 ms - stable
- relative mass uncertainty ≈ 10^{-8}

  for mass doublets and certain cases ≈ 10^{-9}
- required number of ions for a measurement 5-10

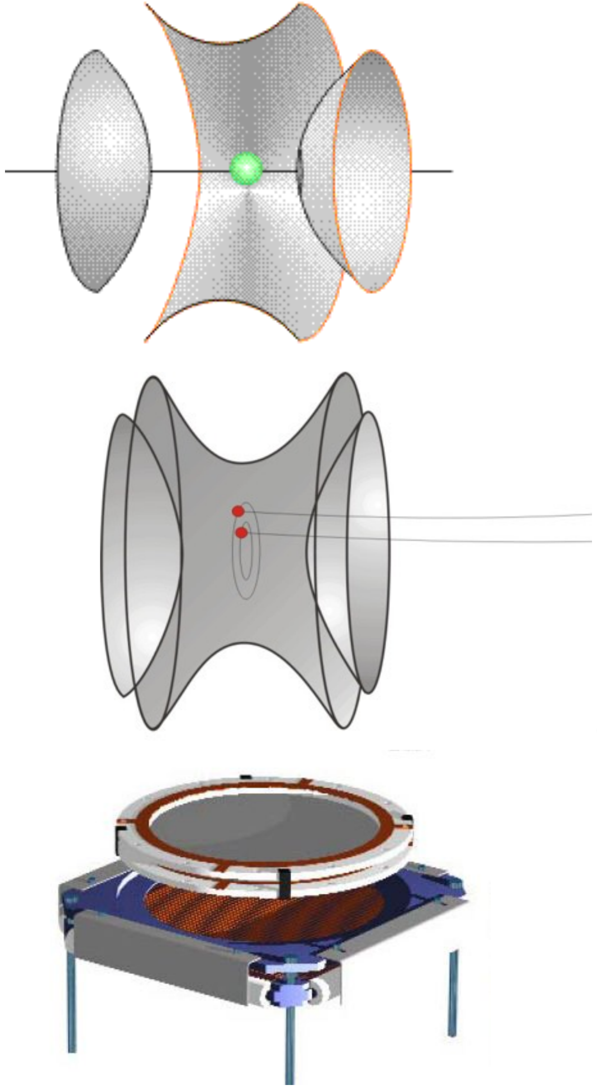
Yield often not the limiting factor but contaminants / background

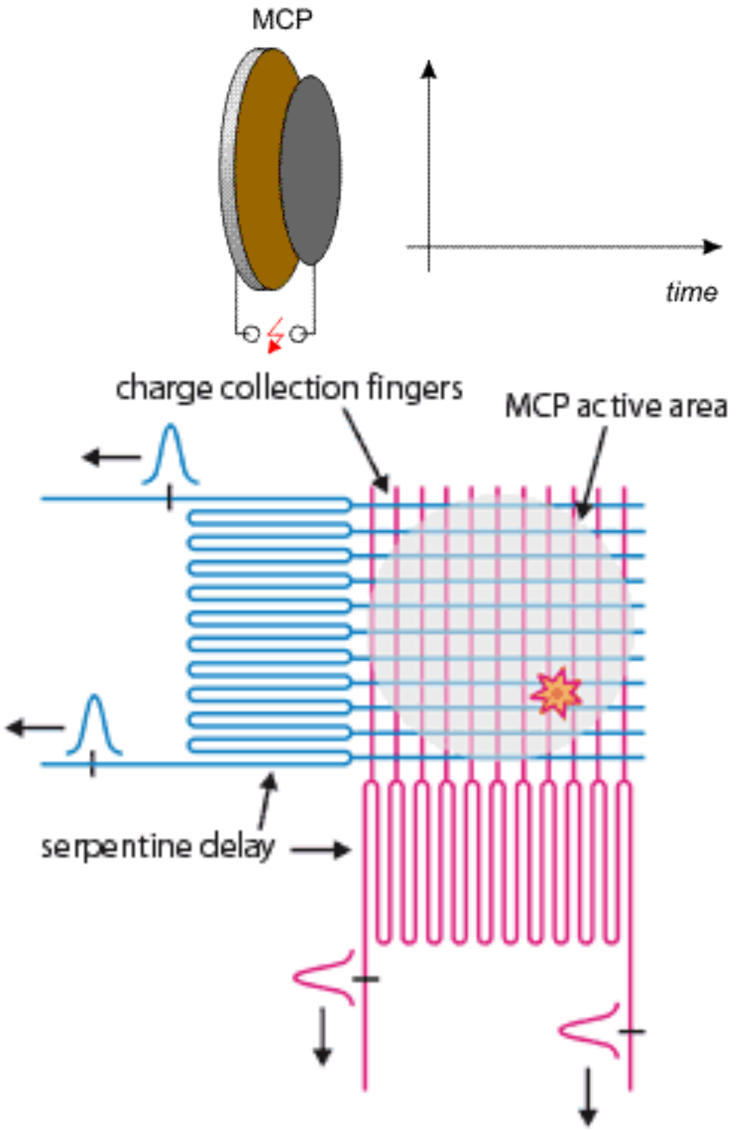
New Gold Standard in Penning-Trap Mass Spectrometry

Destructive time-of-flight detection

Spatially resolved detection

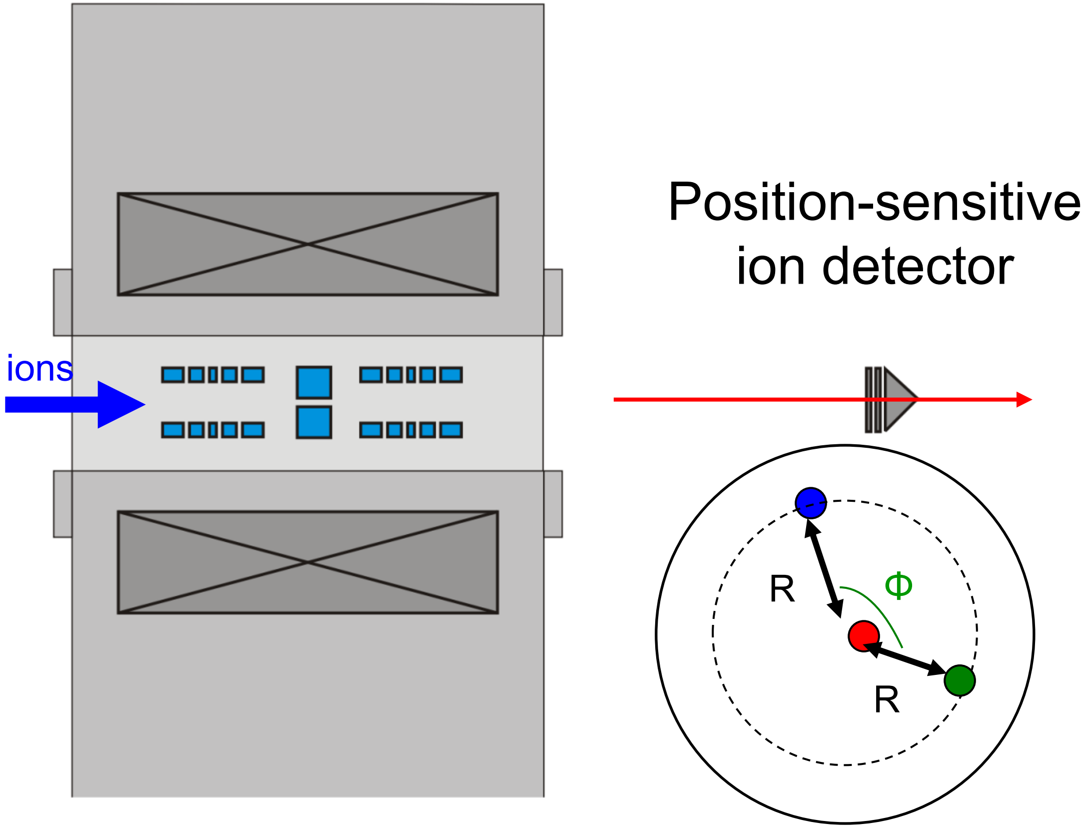
Delay-line detector





S. Eliseev et al., Appl. Phys. B114, 107 (2014)

Phase-Imaging Ion-Cyclotron-Resonance Method (PI-ICR)



pioneered at SHIPTRAP

Now in use at JYFLTRAP, ISOLTRAP, LEBIT, CPT

S. Eliseev et al., Phys. Rev. Lett. 110, 082501 (2013)

high-precision measurement of ion's (radial) frequency via phase measurement

- cool ions motions and remove unwanted by-products, e.g., nuclear isobars
- determine trap center
- prepare ions in radial motion at defined radius and let them orbit for defined time
- determine ion phase on detector

$$\phi + 2\pi n = 2\pi \nu t \qquad \Delta\nu = \frac{\Delta\phi}{2\pi t} = \frac{\Delta R}{\pi t R}$$

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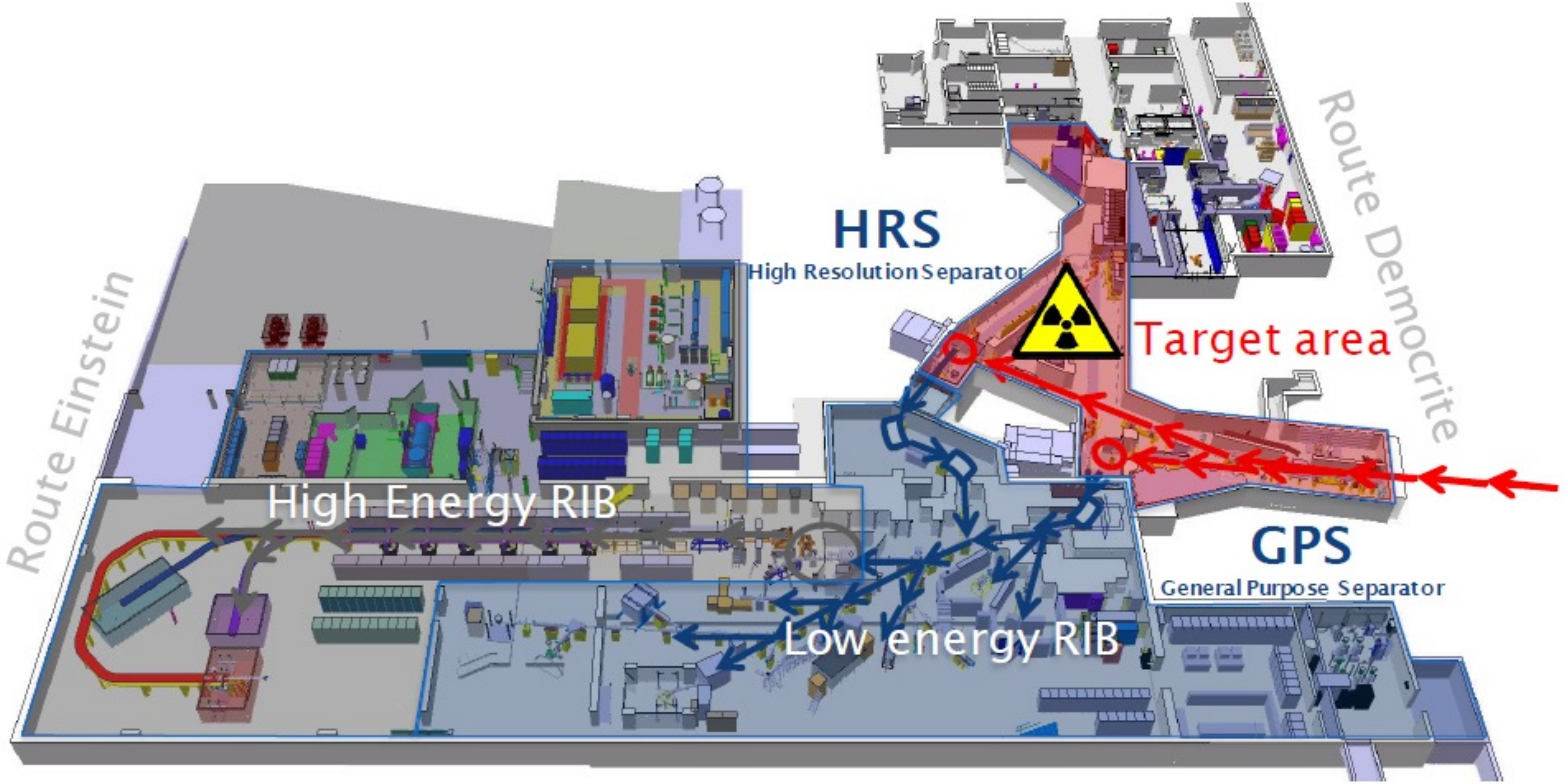
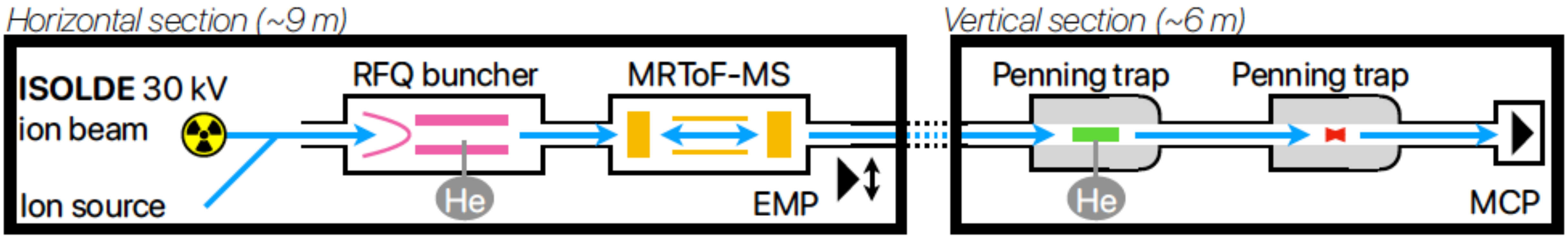
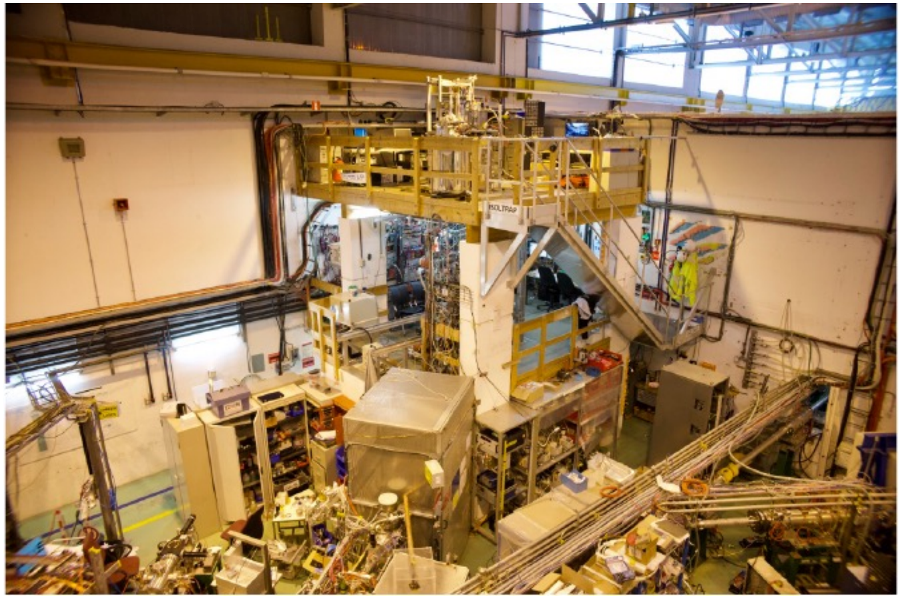
ISOLTRAP at ISOLDE/CERN



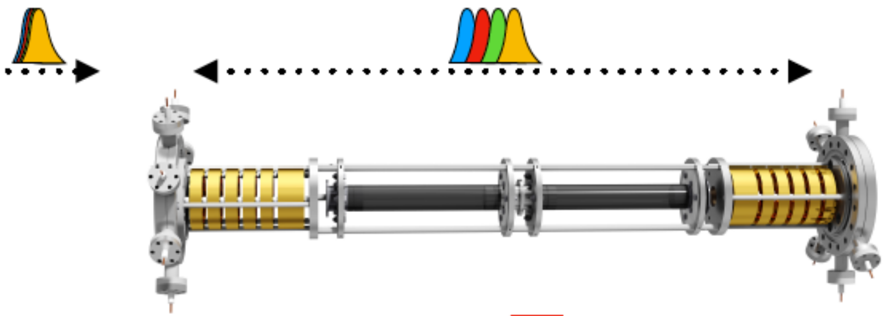
ISOLTRAP Setup





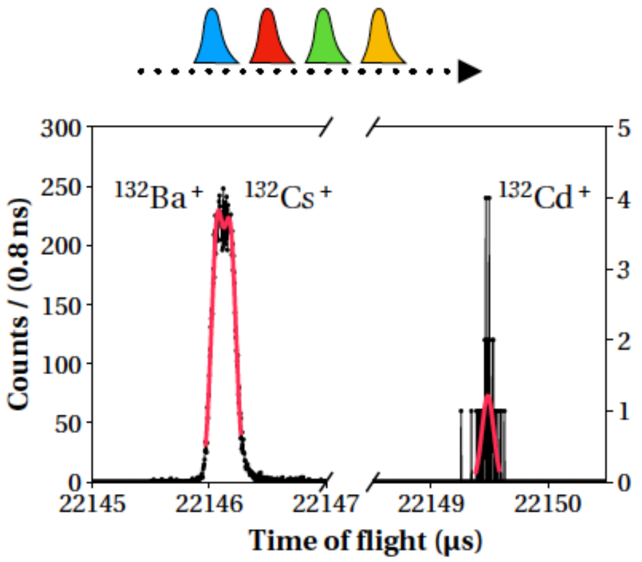
- First Penning trap at accelerator facility (Kluge et al.)
- Use wide range of beams provided by ISOLDE
- Major additions over time:
- RFQ cooler buncher to increase efficiency (R. Moore et al.)
- MR-ToF to remove isobars and measure short-lived nuclides not accessible by Penning trap

ISOLTRAP MR-ToF



$$t = \alpha \cdot \sqrt{m} + \beta$$

- t_{meas} ~ 5 - 20 ms
- $^{m}/_{\Delta m}$ ~ 10^5
- $R_{contamination} \geqq 10^4$

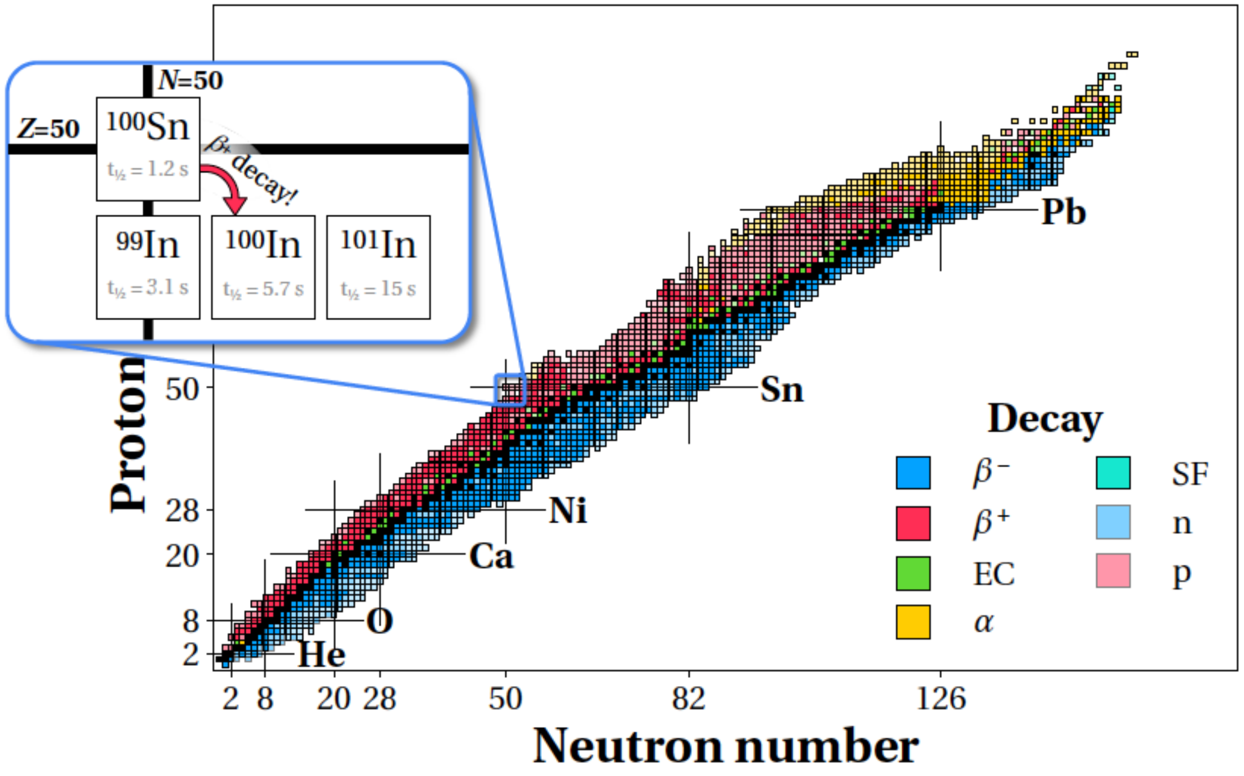


For details on MR-ToF see e.g., R. Wolf *et al.*, IJMS 313, 8 (2012)

MR-ToFs in use at various facilities, GSI/FRS, RIKEN, CPT, JYFL, ...

- broadband time-of-flight mass spectra
- fast cycles (on the order of 10 ms)
- mass measurements with only few detected ions
- efficient isobar separator

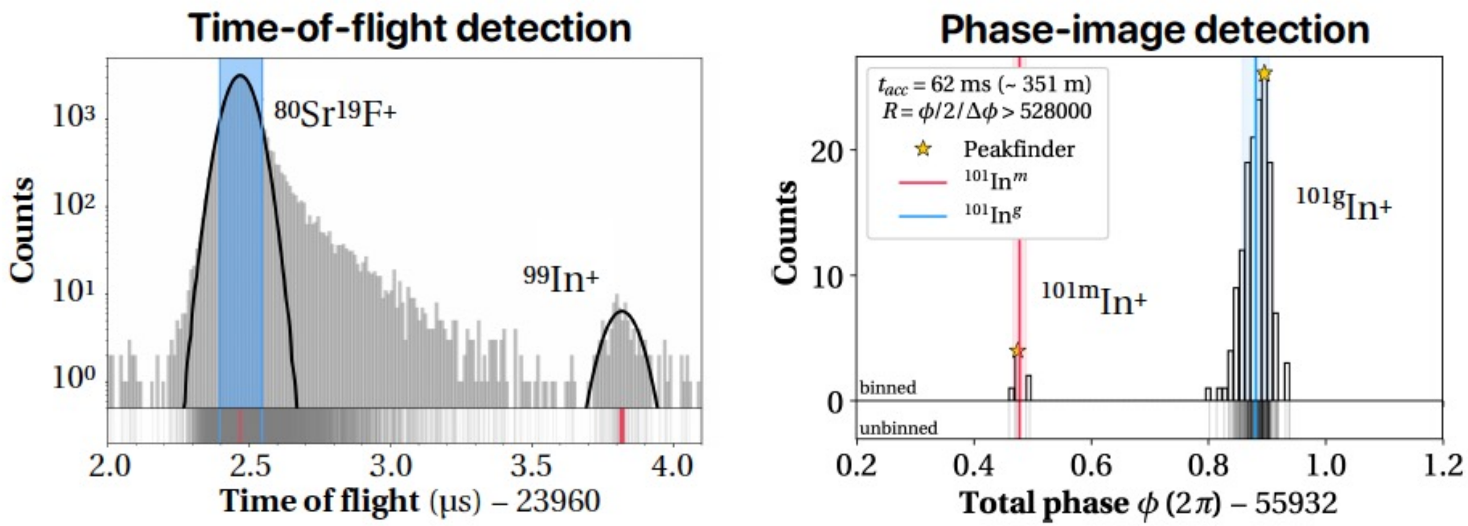
Nuclear Structure Studies of In Isotopes



- study short-range proton-neutron pairing near ^{100}Sn
- heaviest self-conjugate nucleus
- strongest beta decay
- Previously beta-decay studies, mostly at fragmentation facilities (GSI, RIKEN)
- mass measurements on In isotopes ^{99,100,101g,101m}In with ISOLTRAP

Production using LaC target
and two-step laser ionization

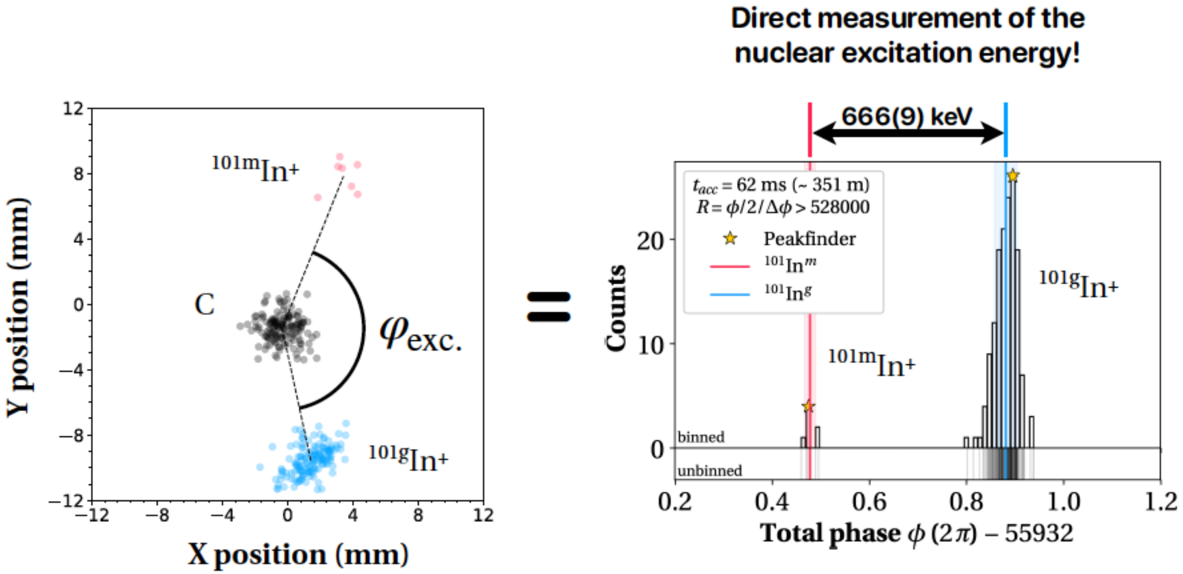
Mass measurements using two techniques



- ISOLDE beam dominated by contaminants SrF
- Using MR-ToF for isobar separations and mass measurement of ^{99}In
- Using Penning trap for cases where high-resolving power is required, e.g., to resolve isomers

M. Mougeot et al., Nature Phys. 17, 1099 (2021)

Determining the Isomer’s Excitation Energy



- ISOLTRAP results agree with results from CSRe and GSI/FRS ion catcher
- Precision improved by > factor 5

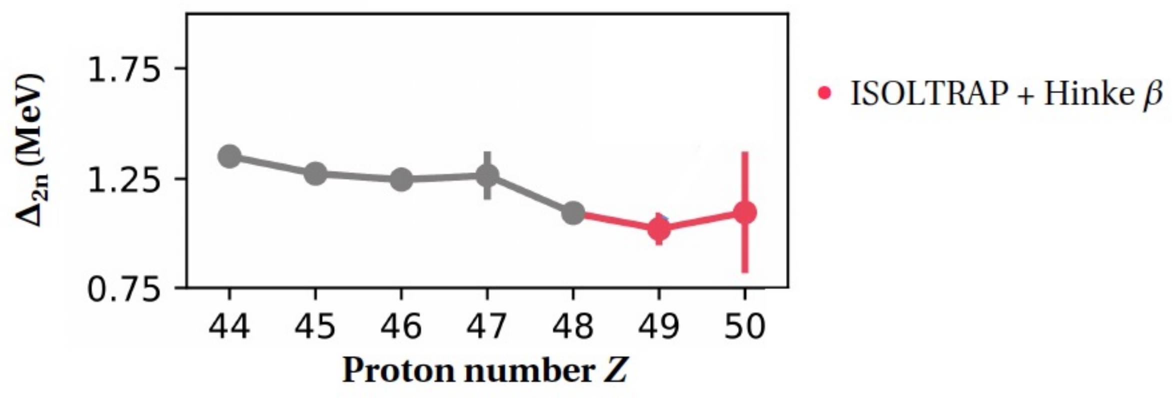
in even-N neutron-deficient indium isotopes:

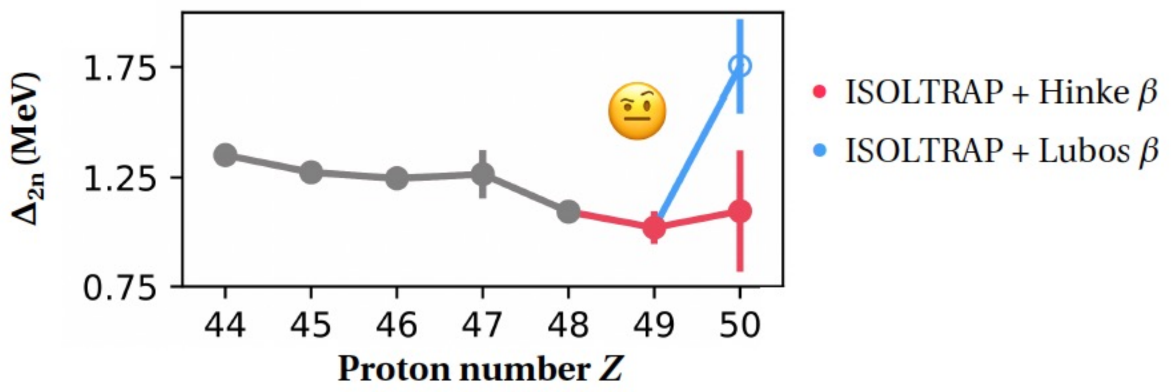
close energy proximity between the πg9/2 and πp1/2 states with large spin difference give rise to long-lived isomeric states lying a few hundred keV above the ground state

➢ requires high mass resolving power

M. Mougeot et al., Nature Phys. 17, 1099 (2021)

Combining ISOLTRAP Mass Values with Decay Data

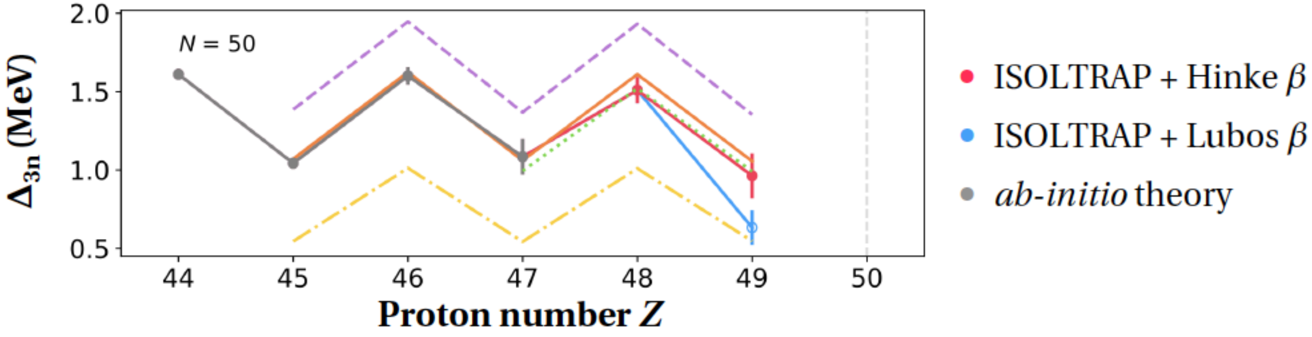


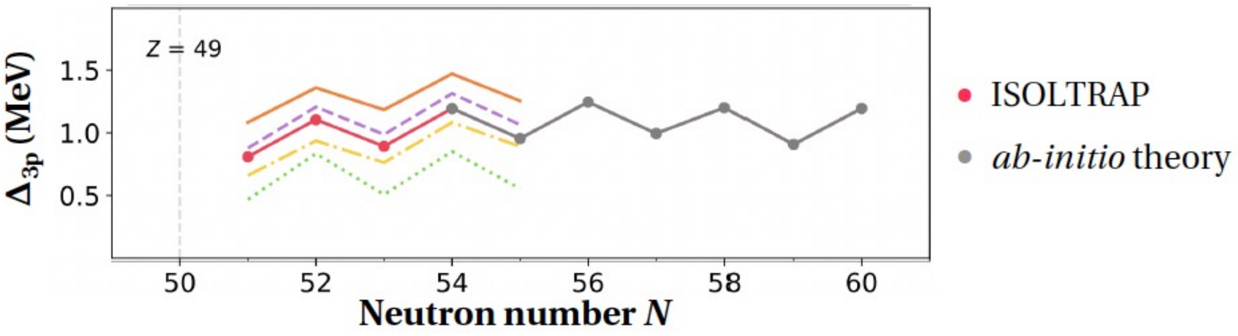


- New mass value from ^{100}Sn derived from ISOLTRAP data **130 keV** more bound
- Downward trend in Δ_{2n} supports the doubly-magic character of 100Sn

Lubos, D. et al. Phys. Rev. Lett. 122, 222502 (2019).
Hinke, C. B. et al. Nature 486, 341(2012).
M. Mougeot et al., Nature Phys. 17, 1099 (2021)

Ab-Inito Theory

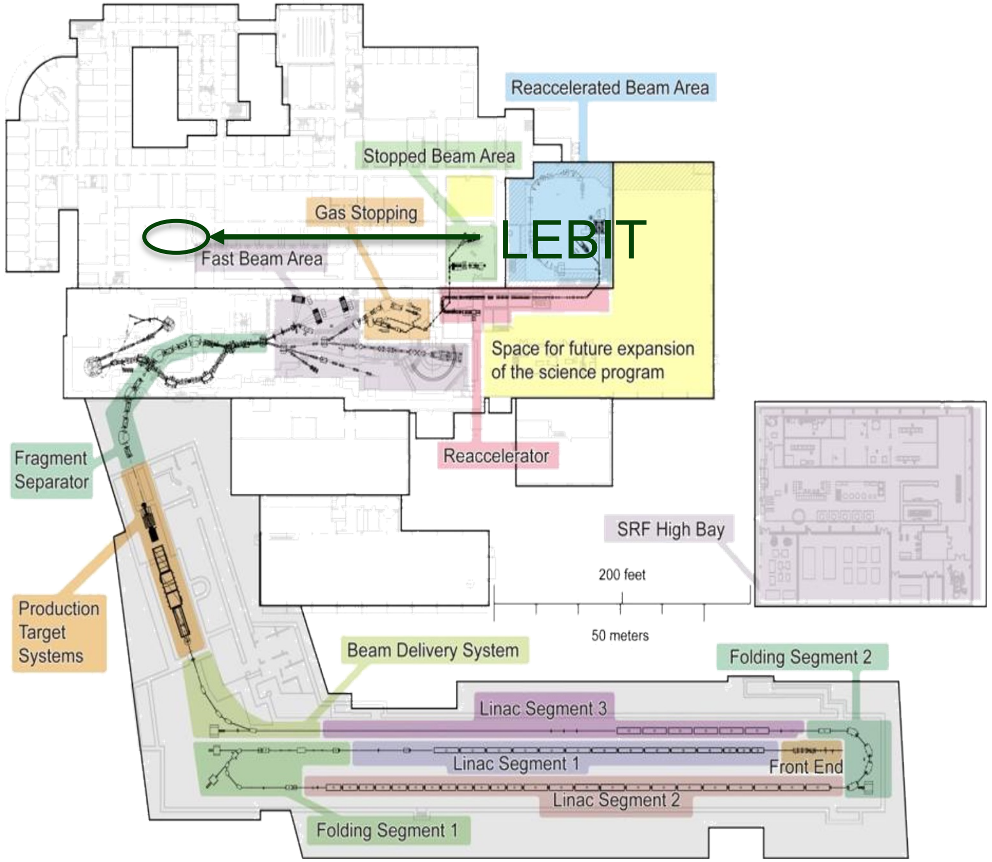




- ISOLTRAP data combined with data from Lubos et al. does not agree with expected trends for doubly-magic nuclei
- Ab-initio theory reproduces well the trends in indium and tin based on data from ISOLTRAP and Hinke et al.

M. Mougeot et al., Nature Phys. 17, 1099 (2021)

FRIB – Facility for Rare Isotope Beams
World-Leading Next-generation Rare Isotope Beam Facility



Most recently, 35% of experiments used beams requiring gas stopping.

- FRIB will produce ~1000 NEW isotopes at useful rates (4500 available for study)
  - Higher-energy primary beams (200 MeV/ u for uranium)
  - Highest intensity rare isotope beams available anywhere
- Fast (~ 200 MeV/u), stopped (~ 30 keV), and re-accelerated (~ 6 MeV/u) beams available (requires gas stopping).

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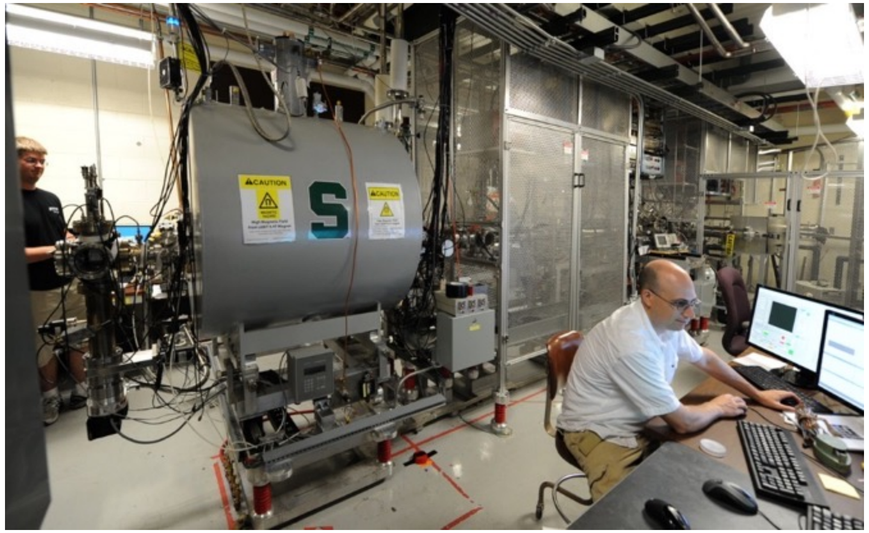
Courtesy R. Ringle

LEBIT Facility at FRIB

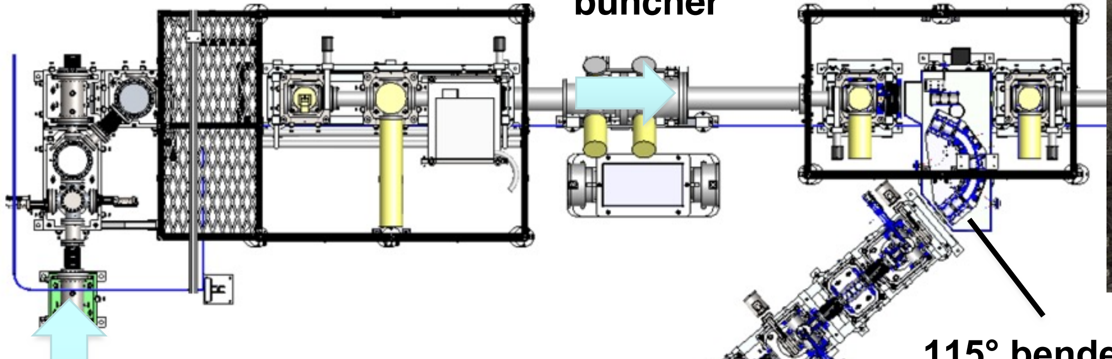


LEBIT facility

Schwarz et al. NIM A 816, 131 (2016)

Off-line Ion sources

RFQ cooler buncher

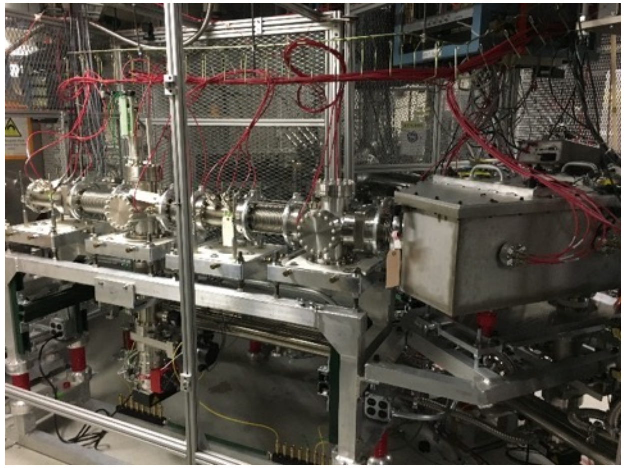


Rare isotope beam (On-line source)

Ringle et al. NIM A 604, 536 (2009)

9.4 T Penning Trap

115° bender + electrostatic quadrupoles



7 T SIPT

Hamaker et al. Hyp. Int. 240, 34 (2019)

Ringle et al., Int. J. Mass Spectrom. 349, 87 (2013)

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Courtesy R. Ringle

High-Precision Penning Trap Mass Measurements with LEBIT

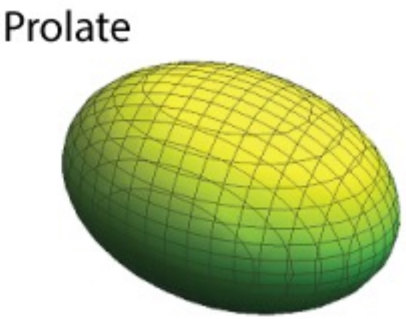
- **Program Goals**
  - Measurements for nuclear structure, nuclear astrophysics, fundamental interactions and symmetry tests
- Recent Publications:
  - **Mass measurement of doubly-magic ^{80}Zr**
    - Nat. Phys. (2021)
  - **Mass measurements of neutron-rich Sc refining N=32,34 shell closures**
    - Phys. Rev. Lett 126 (2021) 042501
  - **Mass measurements of ^{36}Ca, ^{44}V, and ^{66}As for fundamental interactions**
    - Phys. Rev. C 103 (2021) 014323
    - Phys. Rev. C 101 (2020) 064309
    - Nuc. Phys. A 989 (2019) 201
  - **Mass measurement of ^{61}Zn to refine urca neutrino luminosities in accreted neutron stars**
    - Phys. Rev. C 105 (2022) 025804
  - **Offline Q-value measurements of exotic decays of ^{89}Sr, ^{139}Ba, and ^{138}La**
    - Phys. Rev. C 100 (2019) 024309
    - Phys. Rev. C 100 (2019) 014308

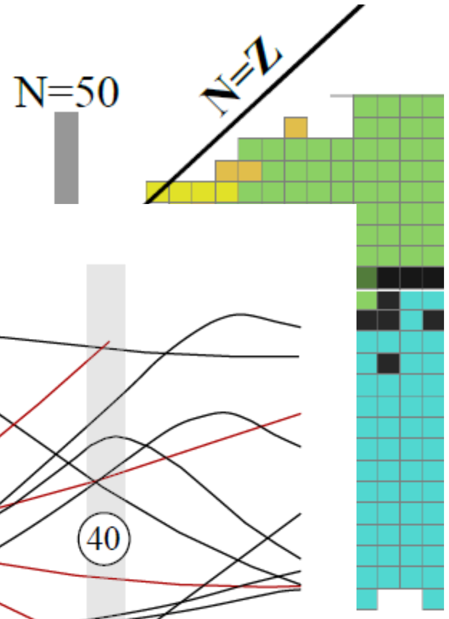
Courtesy R. Ringle

Nuclear Structure Studies Around N=Z=40

- N=Z line is a rich lab for p-n investigations
  - Protons and neutrons occupy same orbitals
  - Enhanced binding due to Wigner energy

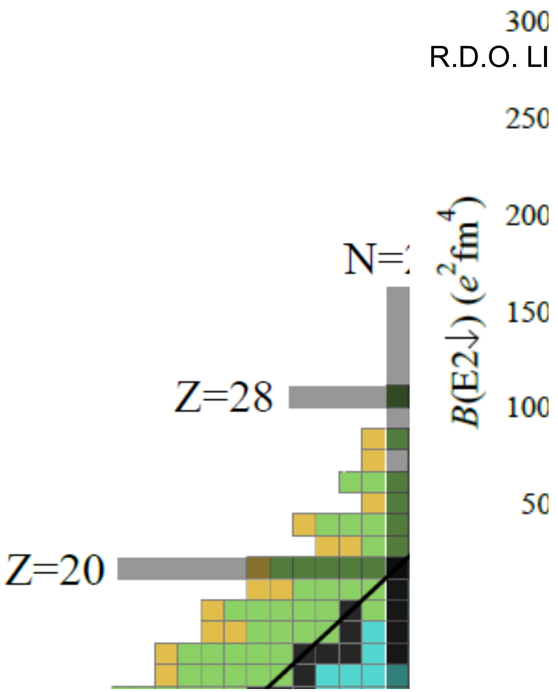
Bentley & Frauendorf Phys. Rev. C 88, 014322 (2013)

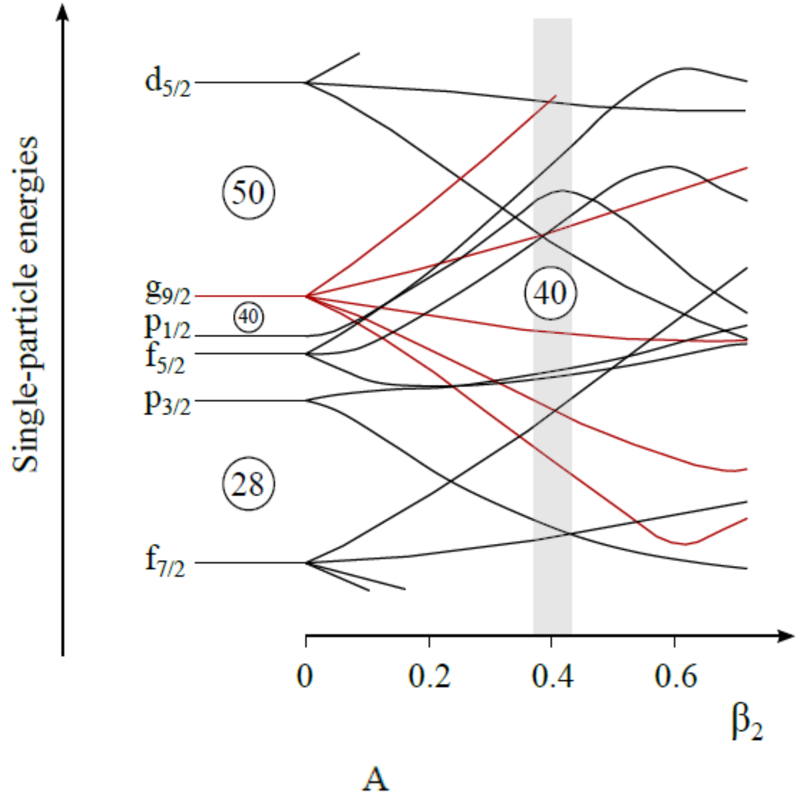




- A region of very strong nuclear deformation
  - Among the most deformed ground-states in the nuclear chart
  - $\beta_2 \approx 0.4$ Lister PRL 59, 12 (1987), Llewellyn PRL 124, 152501 (2020)
  - Possible deformed shell closure predicted at nucleon number 40

Nazarewicz et al. Nucl. Phys. A 435, 397 (1985)





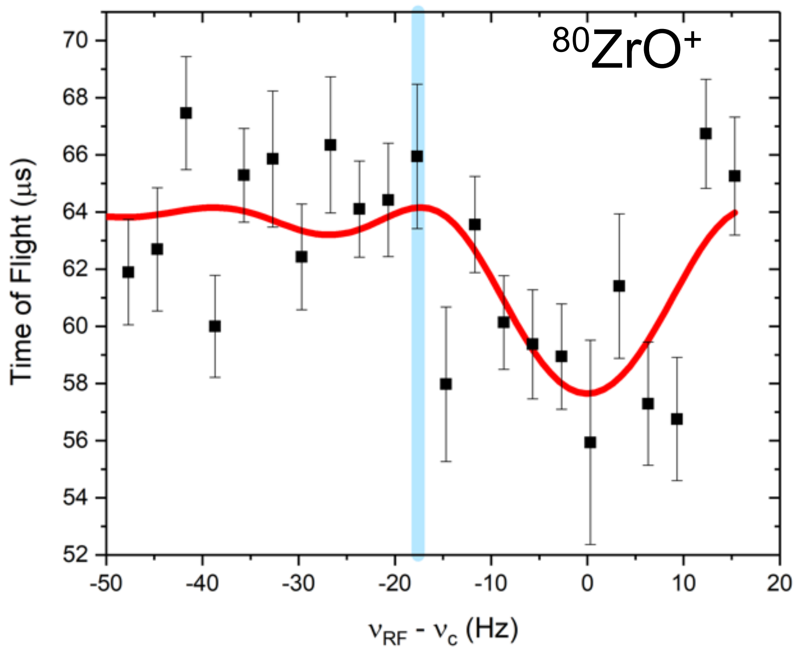
- ^{80}Zr mass in AME20 is extrapolated
  - Experimental values not considered

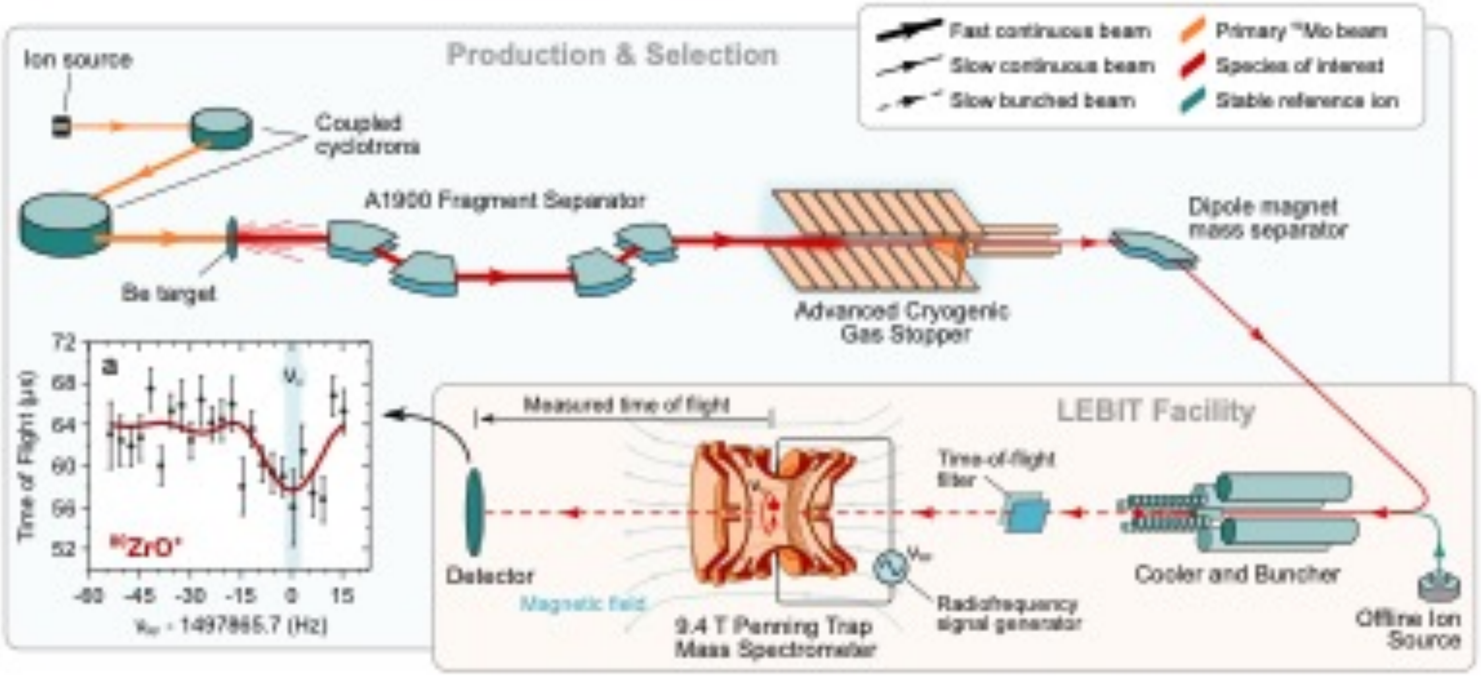
Huang et al. Chin. Phys. C 45, 030002 (2021)

Courtesy R. Ringle

Performed Mass Measurement of ^{80}Zr with LEBIT

Center of scan from AME16*



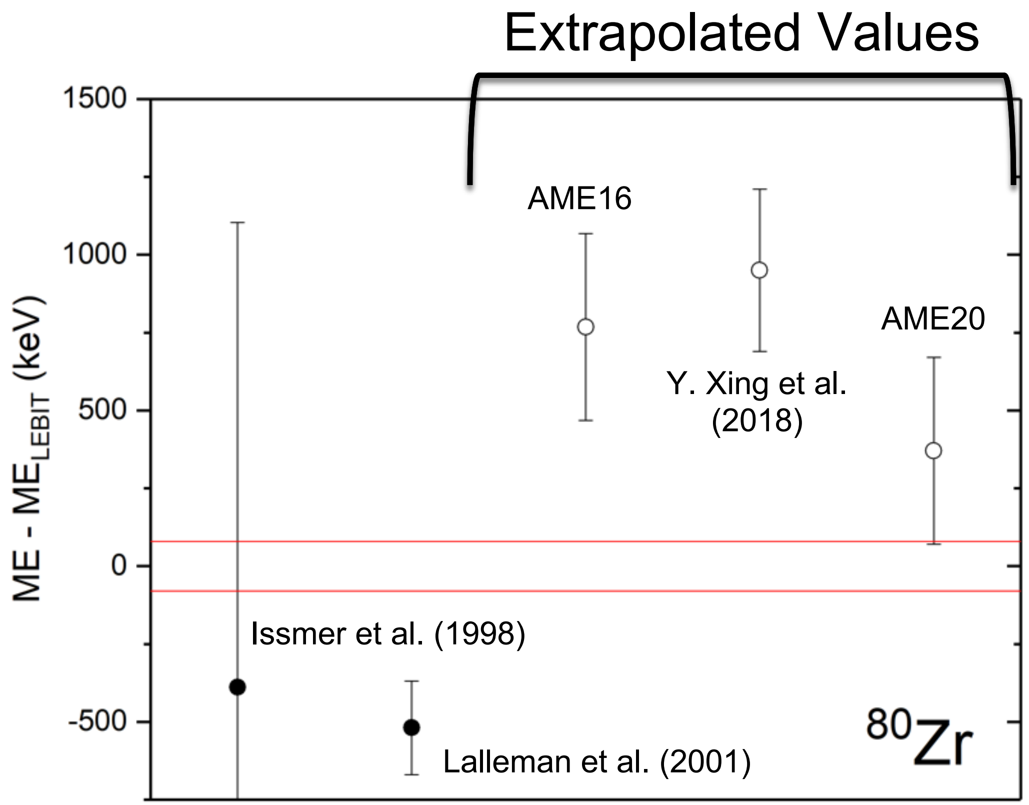


- Measured Mass Uncertainty for ^{80}Zr: 80 keV
- ^{80}ZrO^{+} rate at LEBIT ≤ 1 ion per minute

S. Hamaker et al. Nat. Phys. (2021)

Courtesy R. Ringle

Performed Mass Measurement of ^{80}Zr with LEBIT



Isotope	Ion	Ion Ref.	$\bar{R}$	Mass Excess	AME20[23]	Difference
^{80}Zr	$^{80}Zr\,^{16}O^{+}$	$^{85}Rb^{+}$	1.129 829 01 (99)	-55 128 (80)	-55 517 (1 500)[a]	389 (1 500)
^{81}Zr	$^{81}Zr^{2+}$	$^{41}K^{+}$	0.987 971 08 (13)	-57 556 (10)	-57 524 (92)	-32 (93)
^{82}Zr	$^{82}Zr\,^{16}O^{+}$	$^{87}Rb^{+}$	1.126 770 338 (31)	-63 618.6 (2.5)	-63 614.1 (1.6)	-4.5 (3.0)
^{83}Zr	$^{83}Zr^{2+}$	$^{41}K^{+}$	1.012 274 829 7 (85)	-65 916.33 (65)	-65 911.7 (6.4)	-4.7 (6.5)

[a] Experimental result based on one ^{80}Zr event[24], not included in the AME20.

- LEBIT mass value for ^{80}Zr – significantly more bound
- Closer to some old data than to AME 2020
- Mass uncertainty could be further improved

A. Hamaker et al. Nat. Phys. (2021)

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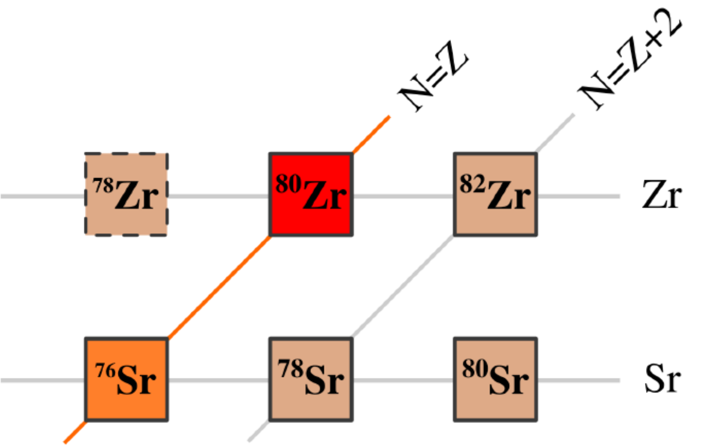
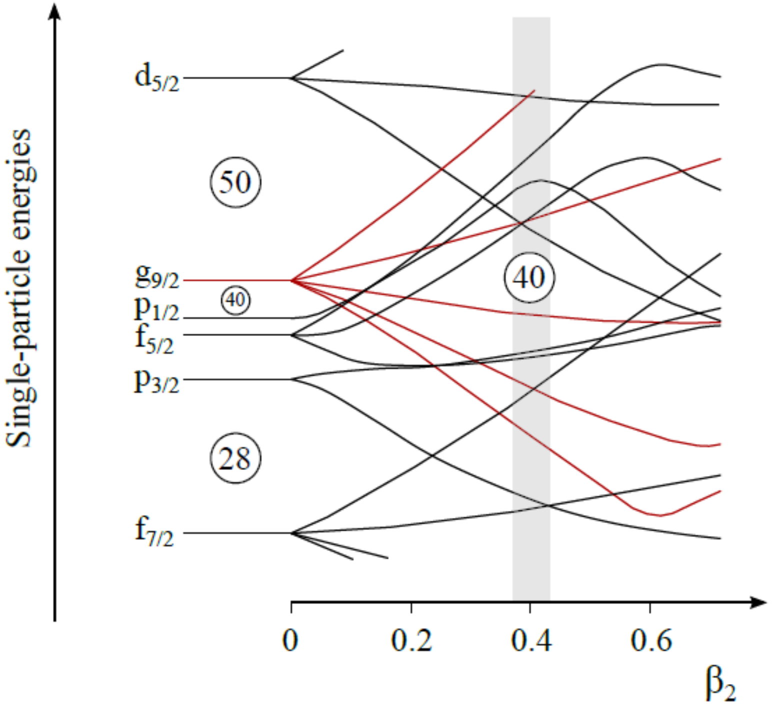
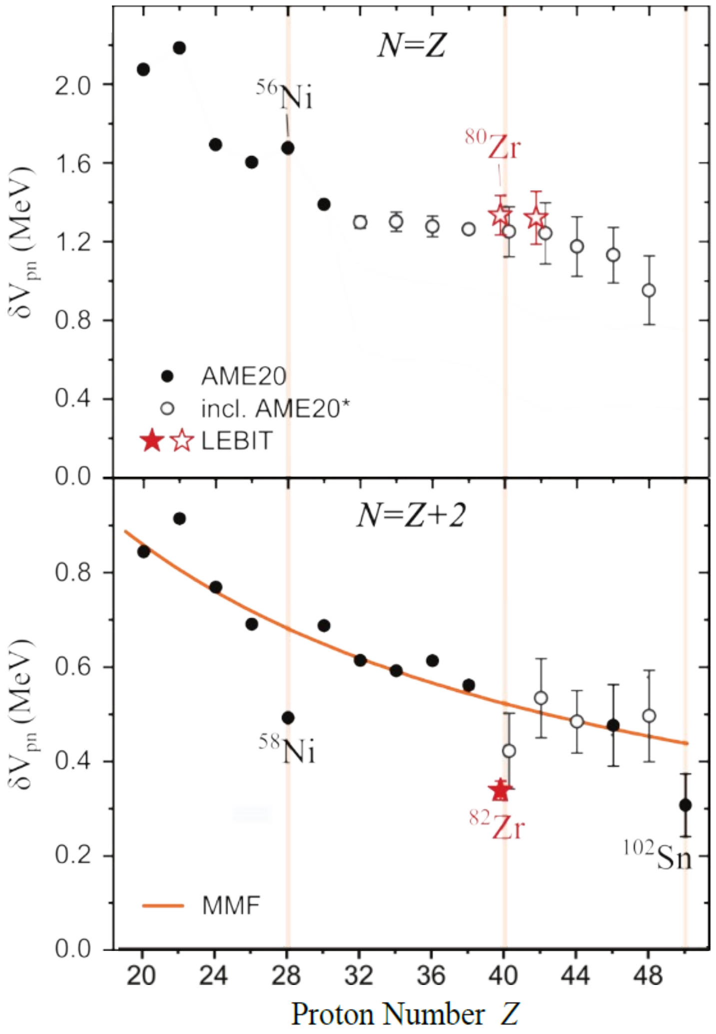
Mass Anomaly Discovered in ^{80}Zr

- δV_{pn} normally used to study p-n interactions
  - ^{82}Zr value no longer relies on e
  - ^{80}Zr behaving in a similar way t
    N=Z nuclei such as ^{56}Ni and ^{100}

Deformed shell closure at N=Z=40







$$\delta V_{pn}(^{80}\mathrm{Zr}) = [(\mathrm{B}(^{80}\mathrm{Zr}) + \mathrm{B}(^{76}\mathrm{Sr})) -$$

$$\delta V_{pn}(^{82}\mathrm{Zr}) = [(\mathrm{B}(^{82}\mathrm{Zr}) + \mathrm{B}(^{78}\mathrm{Sr})) - (\mathrm{B}(^{80}\mathrm{Zr}) + \mathrm{B}(^{80}\mathrm{Sr}))]/4$$

Yang et al. PLB 227, 1 (1989)

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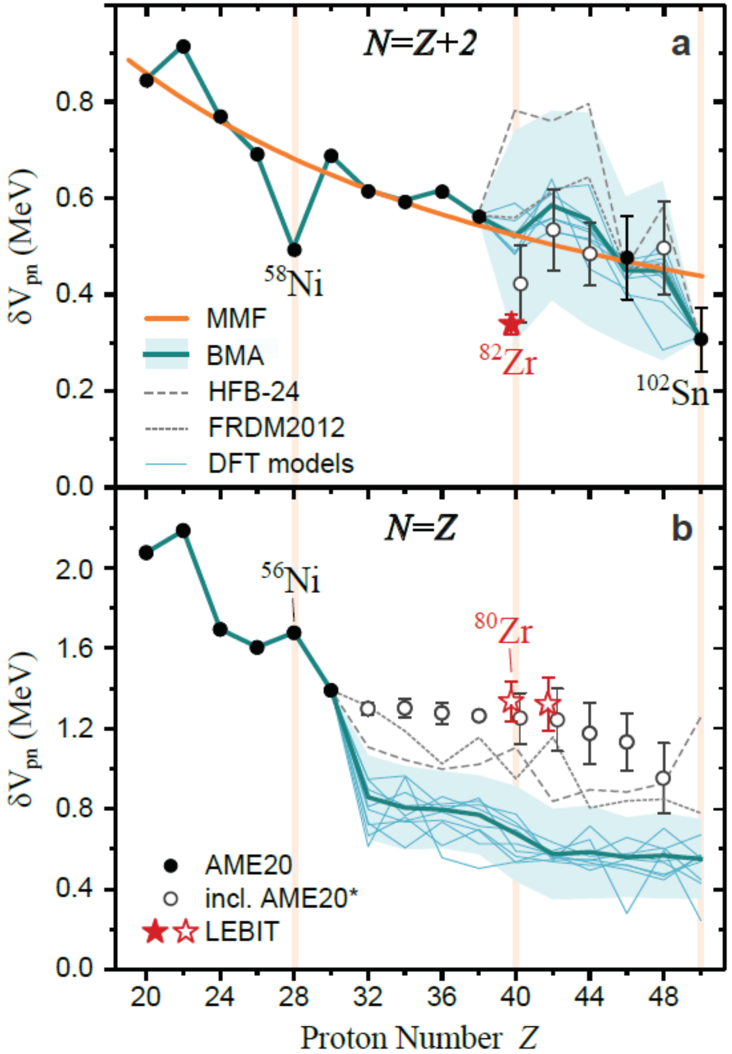
Courtesy R. Ringle

Theory Weighs in on ^{80}Zr Mass

- Can nuclear theories predict the mass anomaly?
- Bayesian Model Averaging
  - Theory and statistics team: R. Jain, S. A. Giuliani, W. Nazarewicz, L. Neufcourt
  - Statistical analysis of 11 global mass models
    - » 9 DFT models: SkM*, SkP, SLy4, SV-min, UNEDF0, UNEDF1, UNEDF2, D1M, and BCPM
    - » HFB-24, FRDM2012
  - Mass models are constrained by experimental results, and used to make predictions and quantify uncertainties at data-scarce region around N=Z

Competition between deformation and p-n pairing must be explored further



Neufcourt et al. PRC 101, 044307 (2020)
Neufcourt et al. PRC 101, 014319 (2020)
Hamaker et al. Nat. Phys. (2021)

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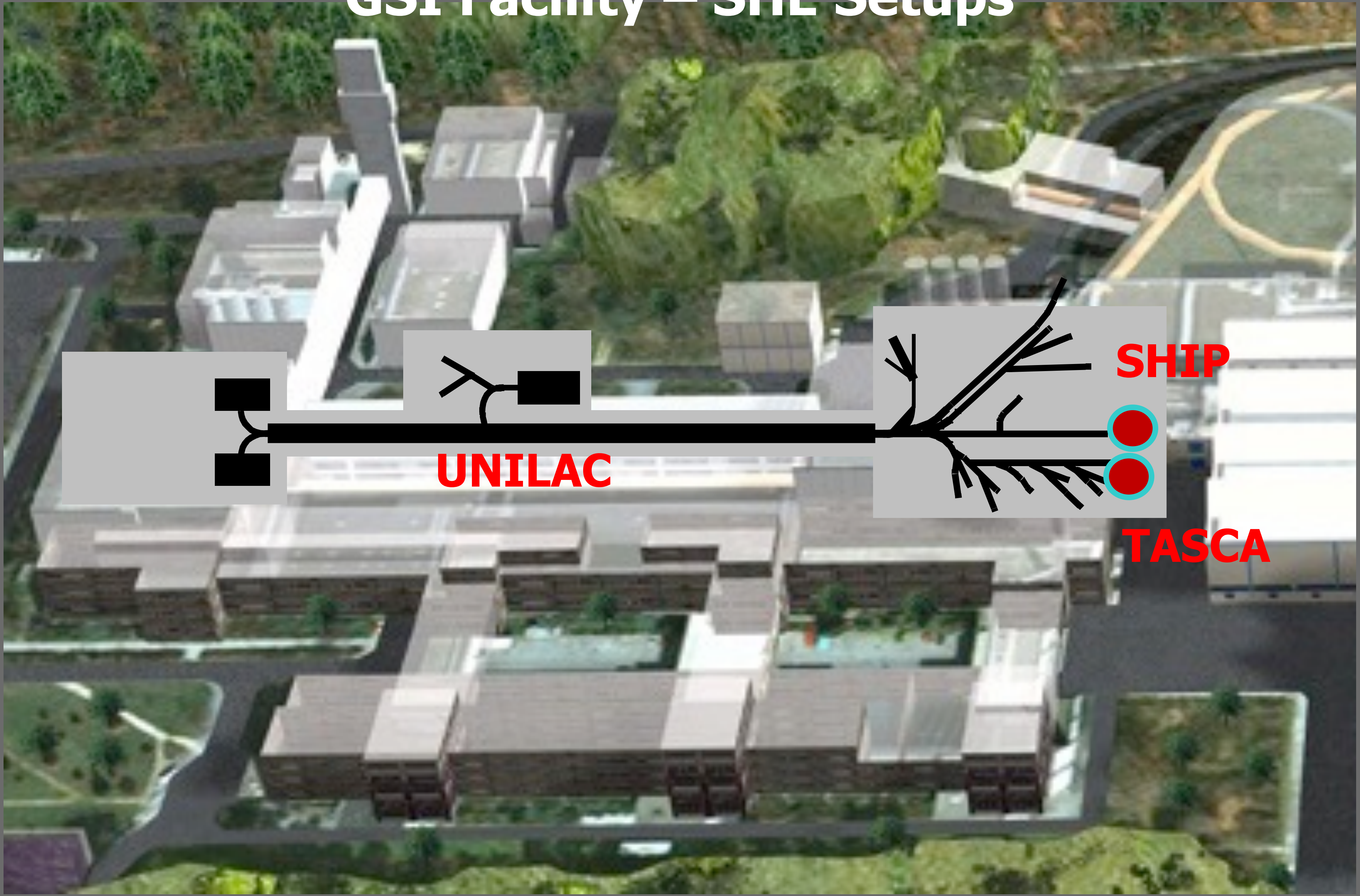
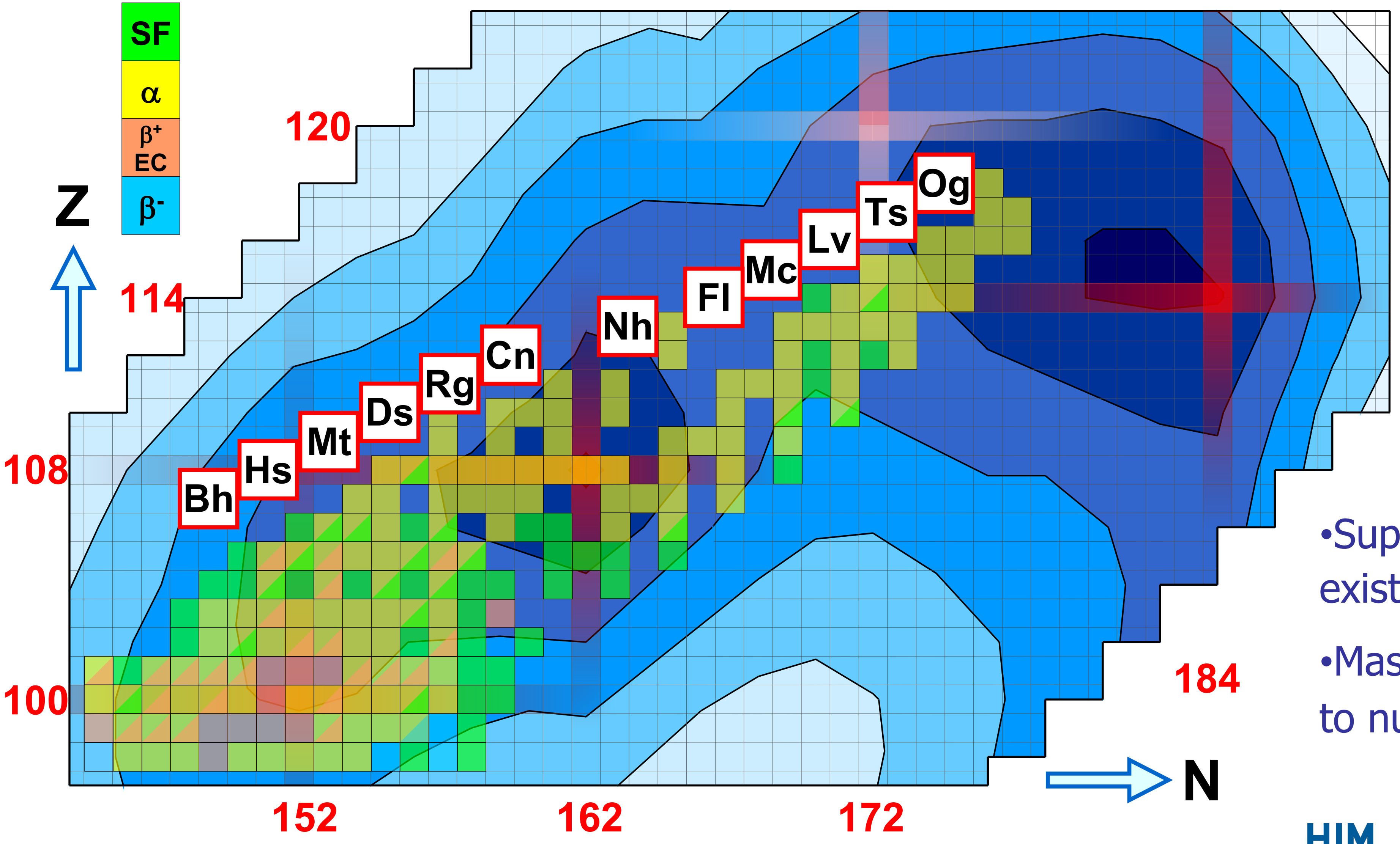
Courtesy R. Ringle

GSI Facility – SHE Setups

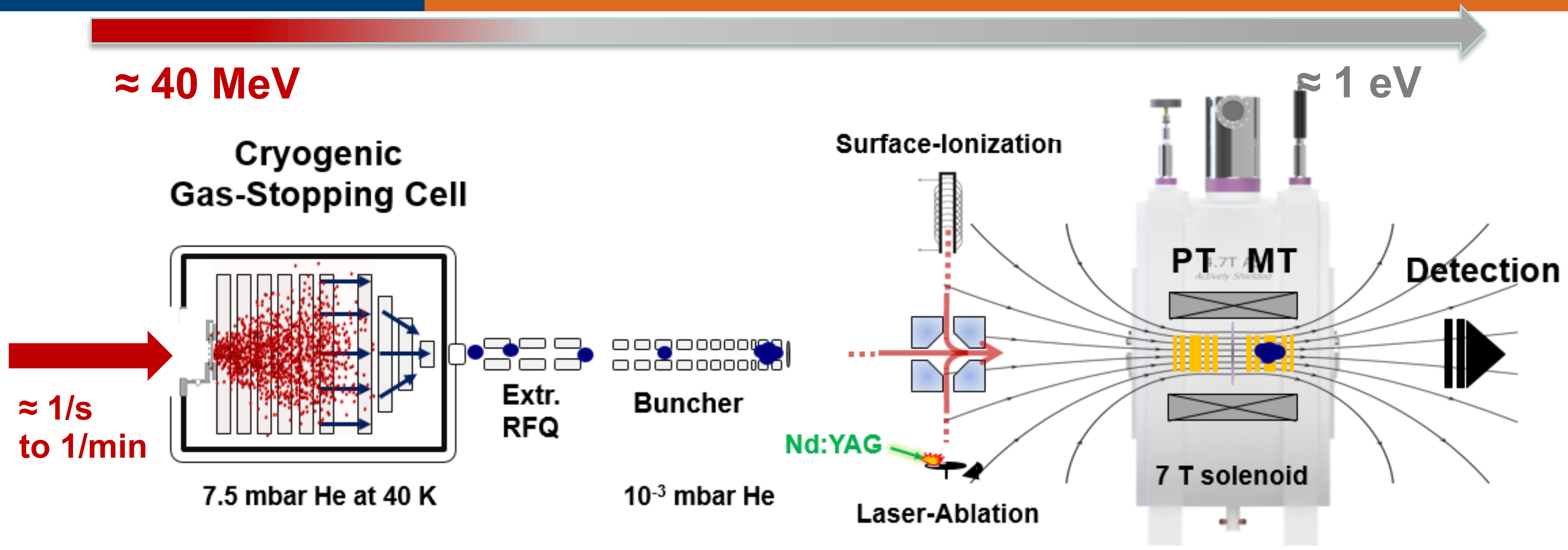


Superhevy Element Landscape



- Superheavy elements owe existence to nuclear shell effects
- Mass measurements give access to nuclear shell structure

SHIPTRAP Setup at GSI Darmstadt



Stopping

Tansport & Bunching

Measurement

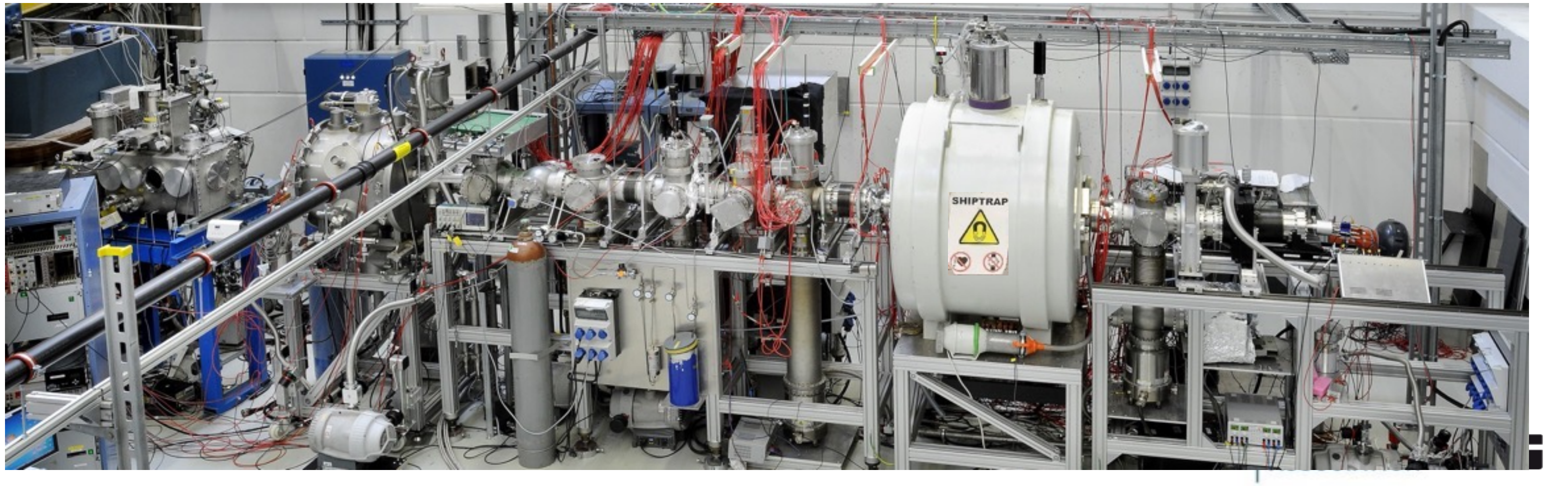
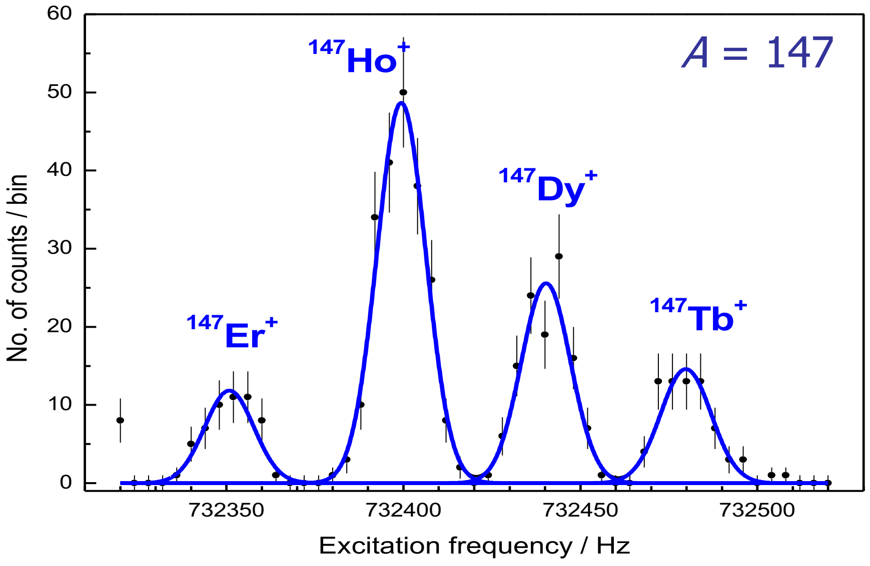


Photo: G. Otto, GSI

SHIPTRAP Performance

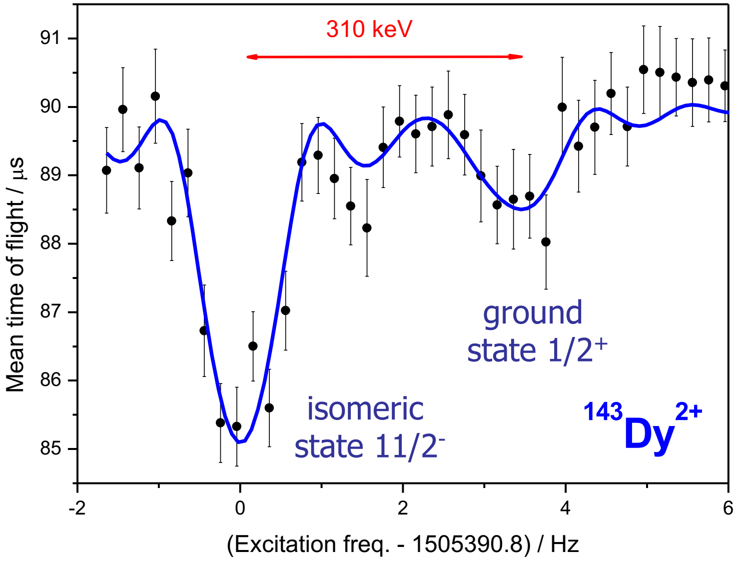


Mass resolving power of

m/δm ≈ 100,000

in purification trap:

⇒ separation of isobars



Mass resolving power of

m/δm ≈ 1,000,000

in measurement trap:

⇒ separation of isomers

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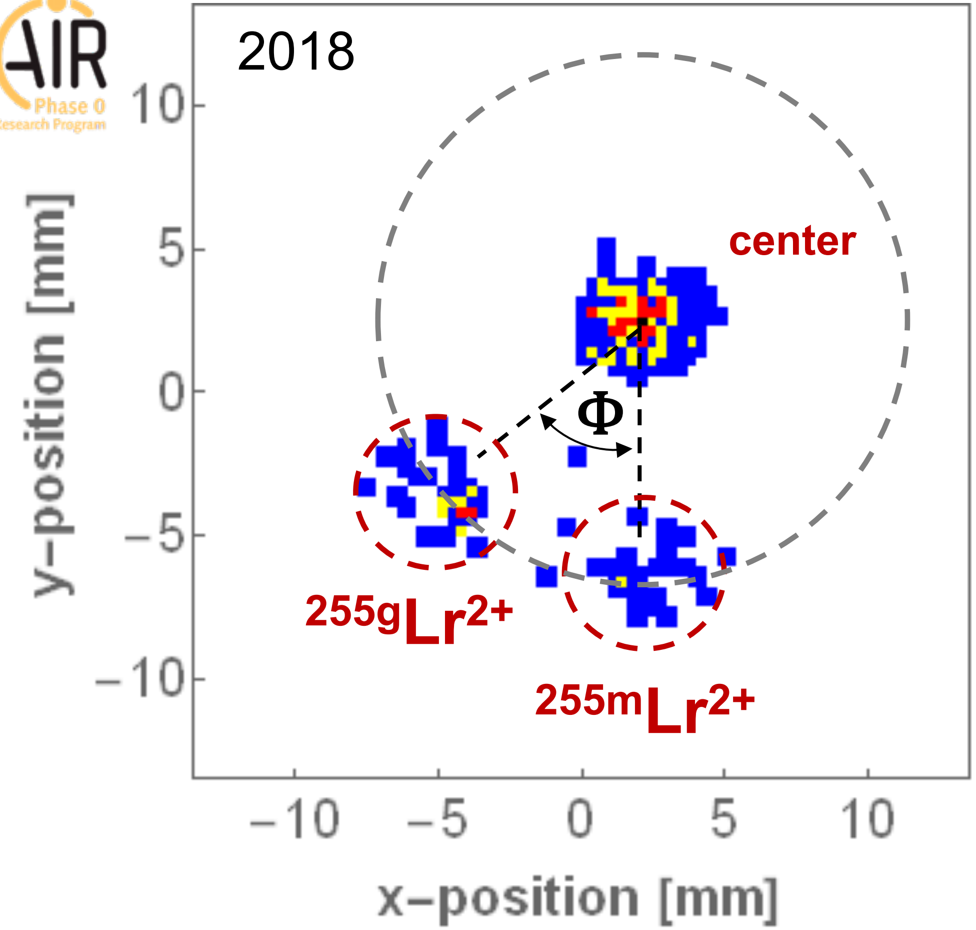
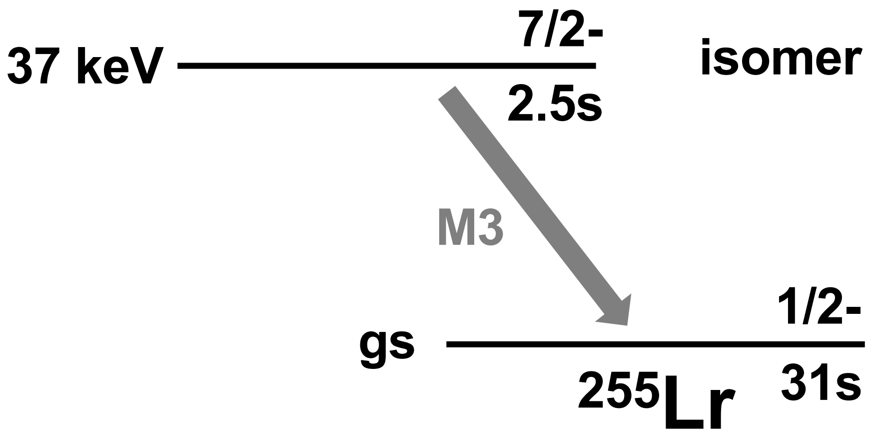
SHIPTRAP Results - Example $^{255(m)}Lr^{2+}$



- **mass resolving power m/Δm ≈ 10^7**
- long-lived isomer with E* ≈ 37 keV was resolved from ground state



Z=103, N=152

- figure shows part of data taken in 10 hours
- 1200 ms phase-evolution time

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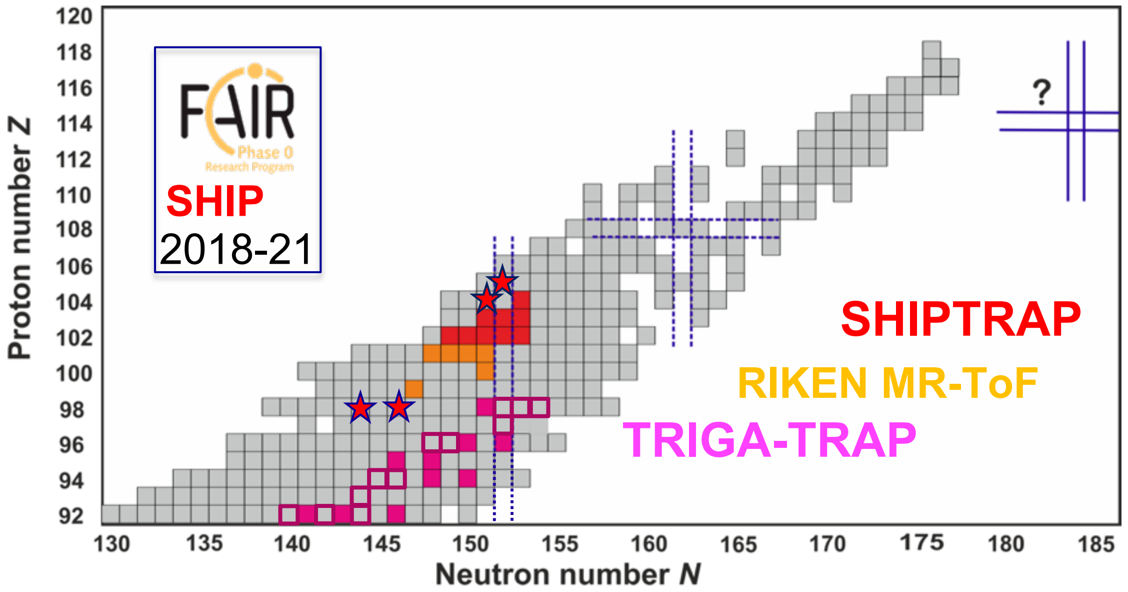
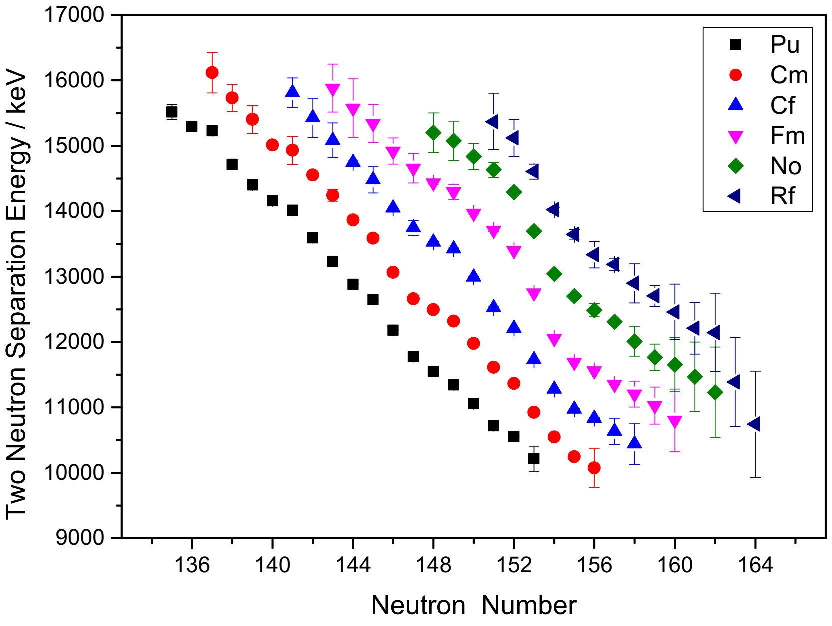
Mass measurements of Actinides and Transactinides

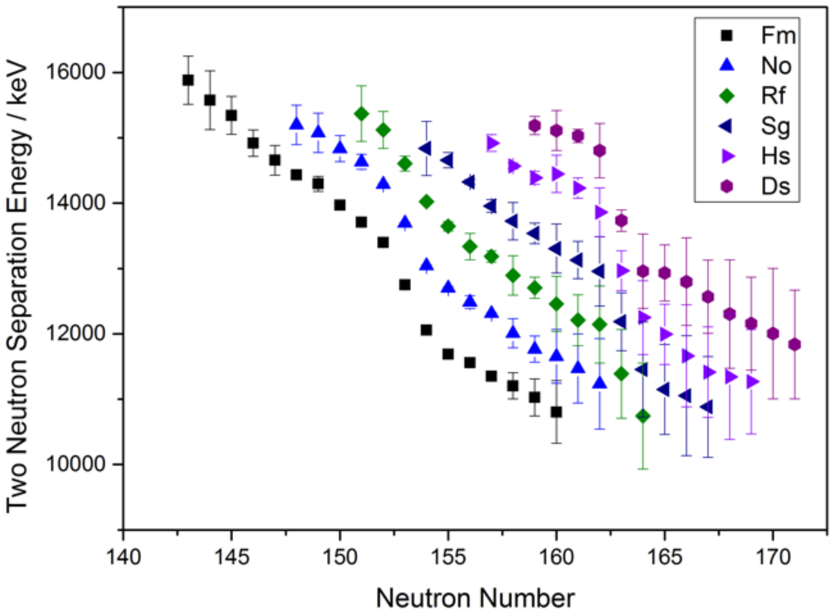


- first (direct) mass spectrometry beyond Z = 100
- measurements with rates of ≈ 0.00002/s and 5 detected ions in total
- rel. statistical mass uncertainty a few 10^{-9}
- Mass resolving power up to m/Δm =11,000,000
- unambiguous identification of low-lying isomers with tens of keV

- O. Kaleja, F. Giacoppo et al., in preparation
- O. Kaleja, PhD thesis Uni Mainz 2020
- B. Andelic, PhD thesis Uni Groningen 2021
- E. Minaya Ramirez et al. Science 337, 1207 (2012)
- M. Block et al., Nature 463, 785 (2010)
- Y. Ito et al., Phys. Rev. Lett. 120, 152501 (2018)
- J. van de Laar et al., in preparation
- M. Eibach et al., Phys. Rev. C 89, 064318 (2014)

Nuclear Shell effects from Separation Energies





neutron separation energies derived from masses and Q_{α}-values show signatures of nuclear shell effects at $N = 152$ and $N = 162$

MB. Radiochimica Acta 703, 603 (2019)

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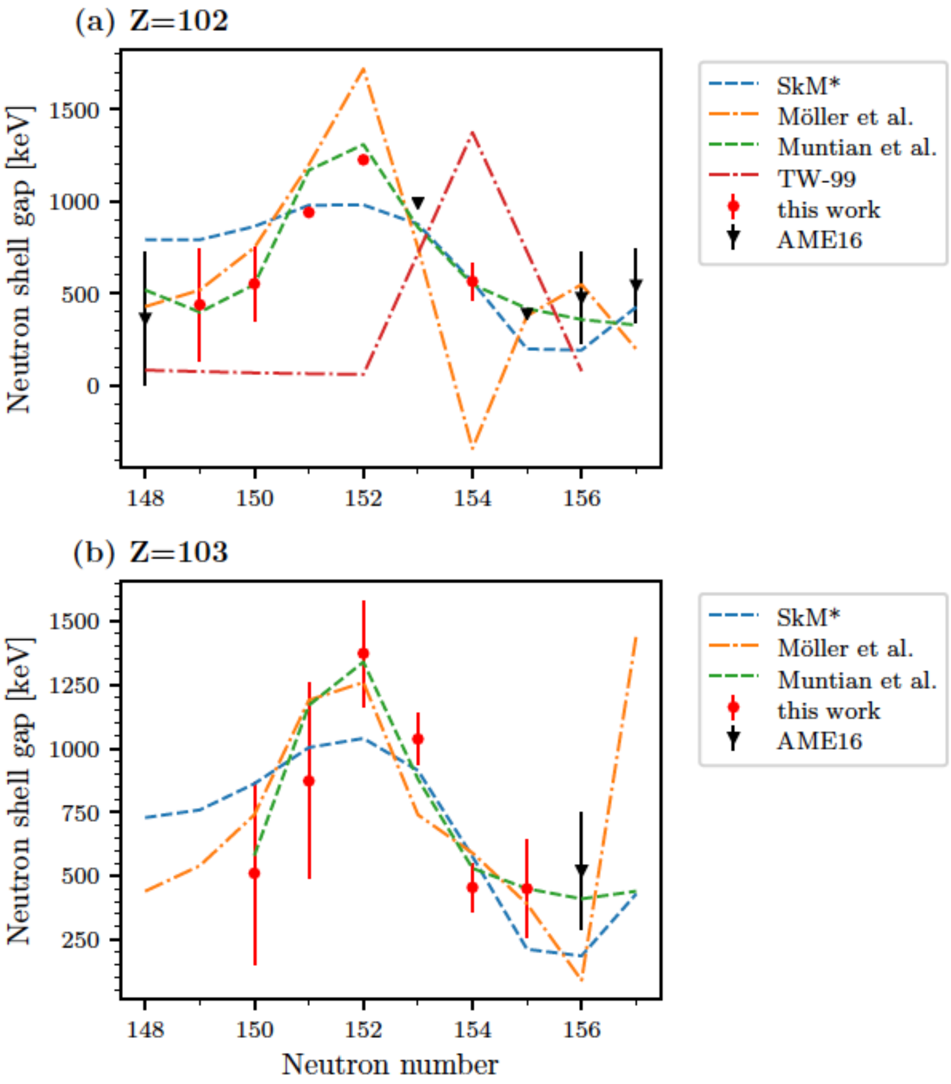
Comparison of Results to Nuclear Models

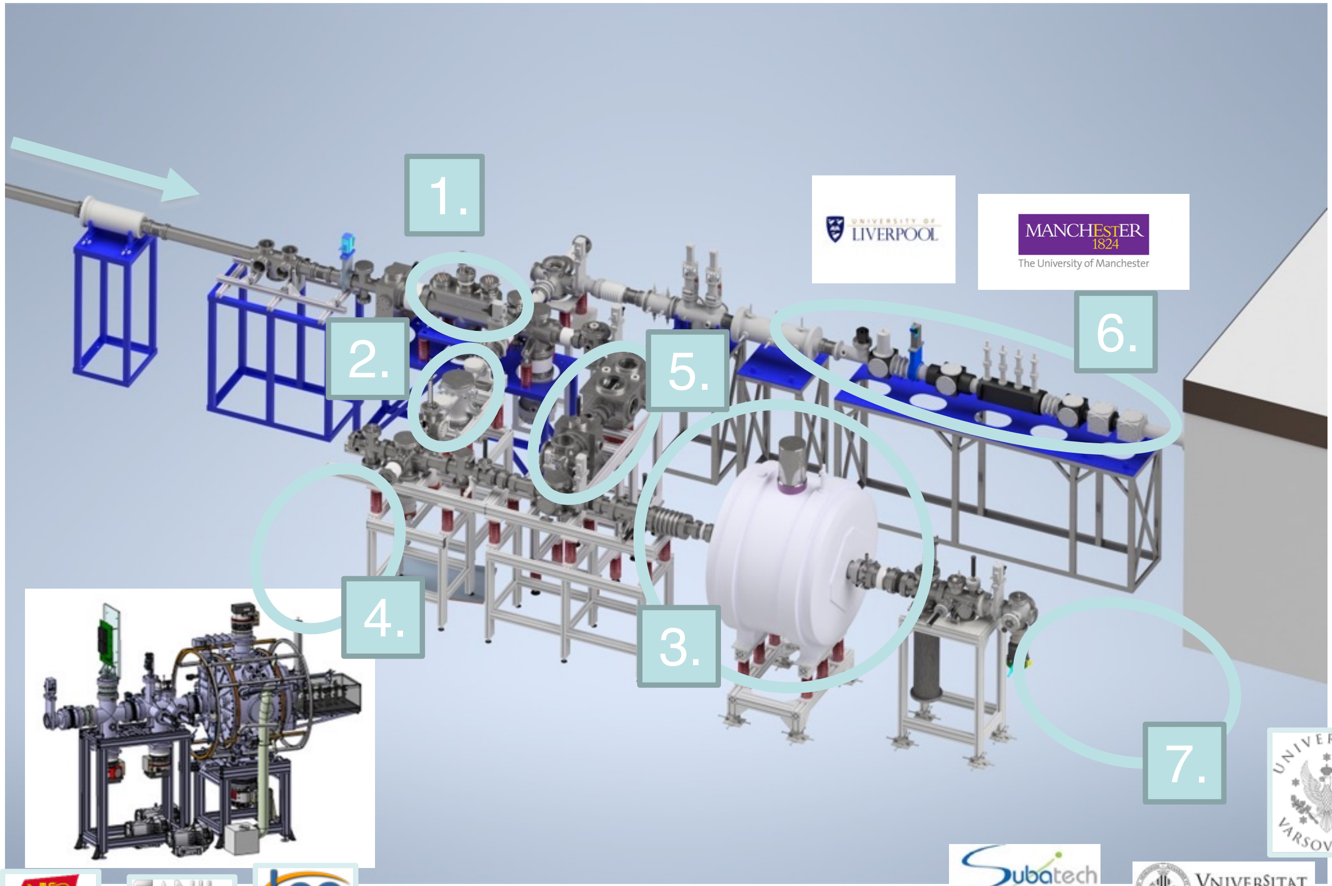


- nuclear models reproduce the general trend relatively well
- absolute masses are sometimes off by up to 1 MeV
- mass differences may be described properly

Experiments with cooled and bunched ions at IGISOL

1. Cooler-buncher
2. MR-TOF
   – Mass separation + spectrometry
3. JYFLTRAP
   – Mass separation + spectrometry
4. MORA
   – ^{23}Mg beta decay, beyond SM physics
5. RAPTOR
   – Hi-res laser re-ionization
6. Collinear laser spectroscopy
7. Post-trap decay spectroscopy

KU LEUVEN

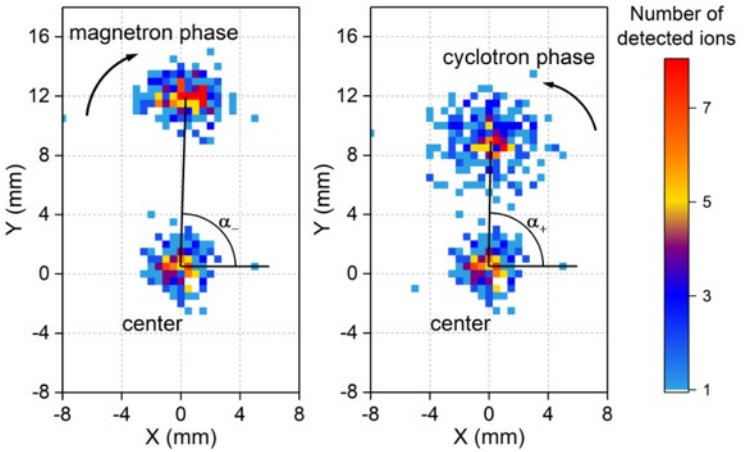


Courtesy T. Eronen

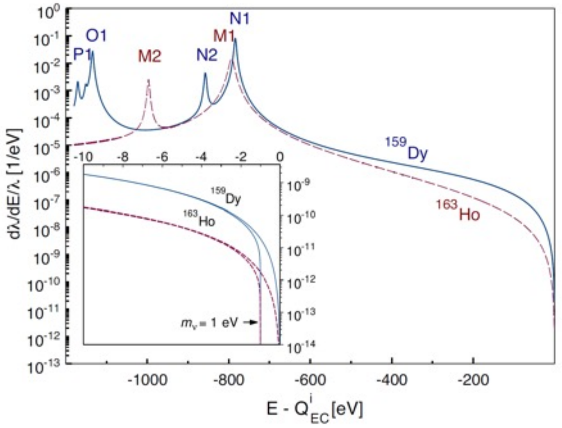
Recent mass measurements at JYFLTRAP

- Utilizing mostly the PI-ICR technique

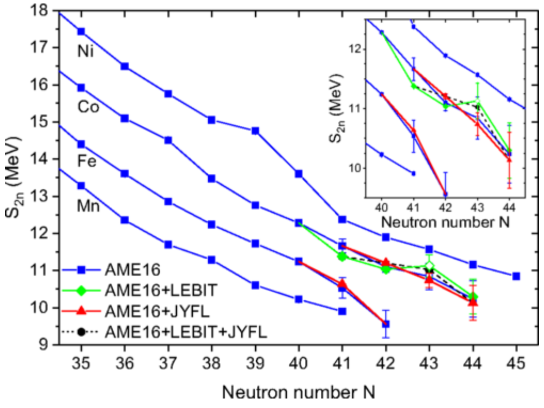
- Fundamental physics
- Neutrino physics
- Astrophysics
- Nuclear structure



$2\beta\beta$ Q-value of ^{98}Mo
D. Nesterenko, L. Jokiniemi et al.
Eur. Phys. J. A (2022) in press



^{159}Dy Q-value measurement
Z. Ge et al.
PRL **127**, 272301 (2021)

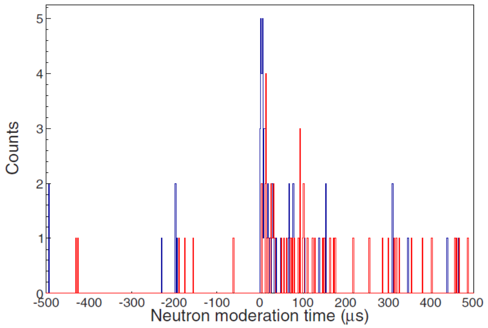


^{67}Fe and ^{69,70}Co masses, nuclear structure at N=70 and impact on r-process reaction rates
L. Canete et al.
Phys. Rev. C **101**, 041304(R) (2020)

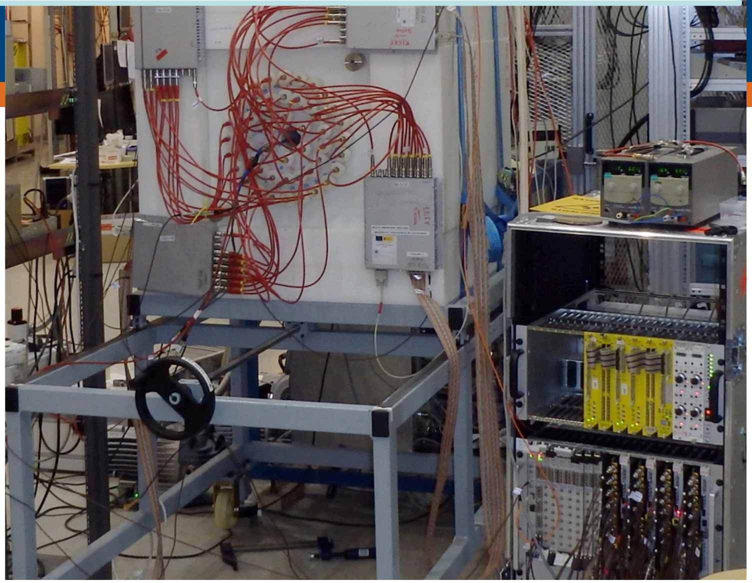
Courtesy T. Eronen

Post-trap decay spectroscopy

- Trap-purified beams with mass resolving power > 10^5
- Mostly of n-rich nuclei



Beta dELayEd Neutron (BELEN) detecto



^{136}Sb, one of the heaviest $\beta 2n$ emitters studied
R. Caballero-Folch et al., PRC 98, 034310 (2018)

Decay spectroscopy of the ^{81}Ge ground state

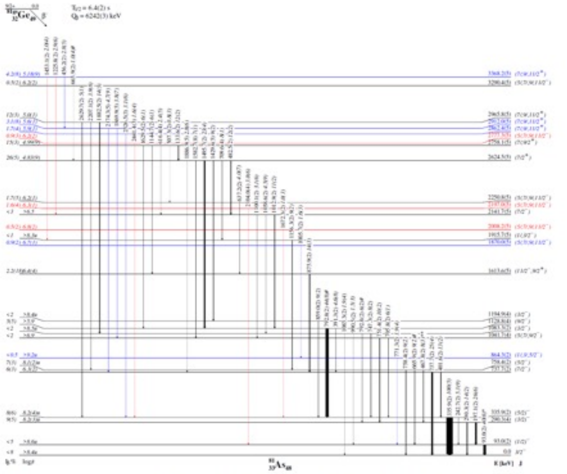
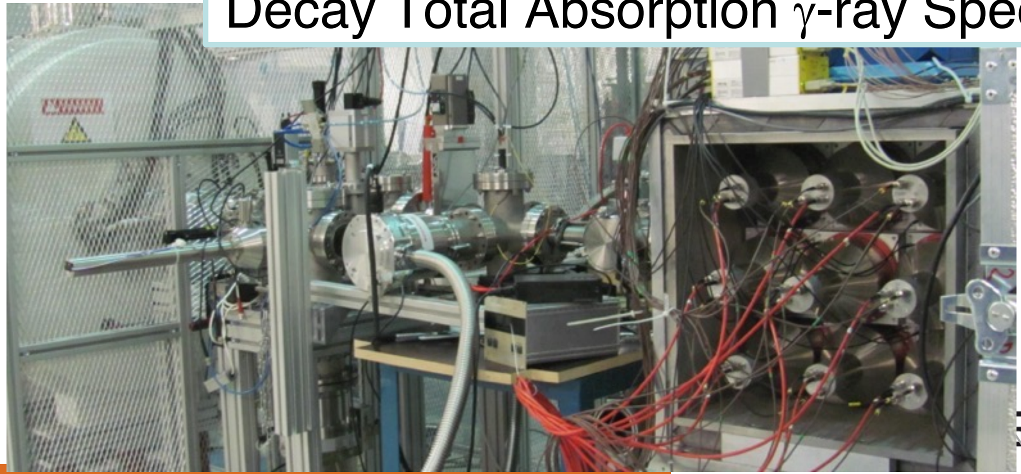


Fig. 5. ^{81g,m}Ge β-decay scheme. Levels and transitions shown in black (blue) were already attributed to the ^{81}Ge ground state (isomer) decay in Ref. [14], respectively. Levels and transitions shown in red are those newly proposed from the present work. * indicates that intensity has been corrected by conversion coefficient assuming M1 multipolarity. ** indicates that intensity has been corrected taking into account the ^{81}Se transition. # are indicating intensities estimated using $\gamma-\gamma$ coincidences for the doublets. Spin-parity notation $(1/,3/,5/2^-)$ stands for $(1/2^-)$, $(3/2^-)$ or $(5/2^-)$.

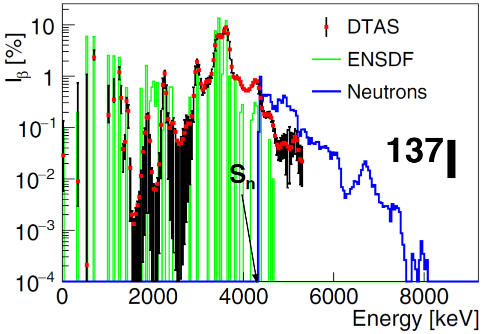
C. Delafosse et al.
EPJA (2022) in print

Decay Total Absorption γ-ray Spectrometer (DTAS)



Courtesy T. Eronen

Guadilla et al., PRC 100, 044305 (2019)



Summary and Conclusions

- New techniques in Penning-trap mass spectrometry have
  - extended the reach of towards more exotic nuclei
  - boosted the precision to the 10^{-9} level
  - allowed the resolution of low-lying isomers
- complementary setups exploit different nuclear reactions so essentially radioisotopes from all elements are accessible
- variety of mass measurements for nuclear structure studies, nuclear astrophysics, and fundamental physics performed recently

Directions for future imporvements

- exploit next-generation RIB facilities to extend the reach toward more exotic nuclides
- extending mass measurements to rarer nuclides goes hand in hand with technical and methodological developments to

➢ improve sensitivity

➢ provide higher efficiency

➢ obtain higher resolution that leads to higher precision

THANK YOU!

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