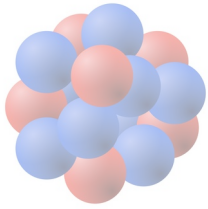




The success and future perspectives for high-precision atomic mass measurements using MRTOF-MS at RIBF

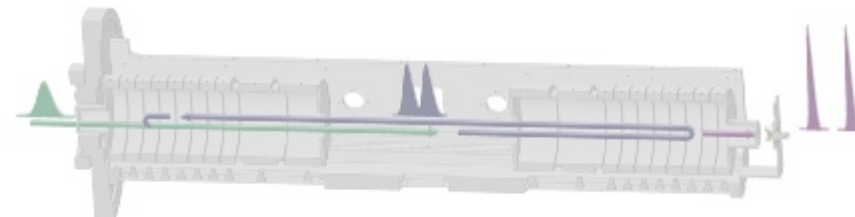
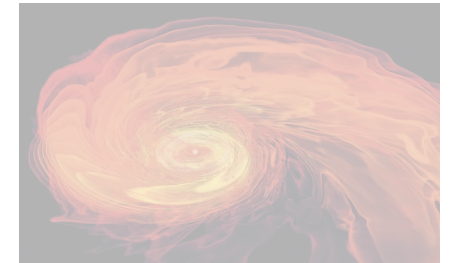
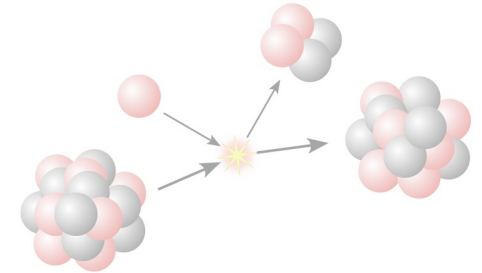


Marco Rosenbusch for RIKEN/SLOWRI and the WNSC-IPNS-KEK collaboration

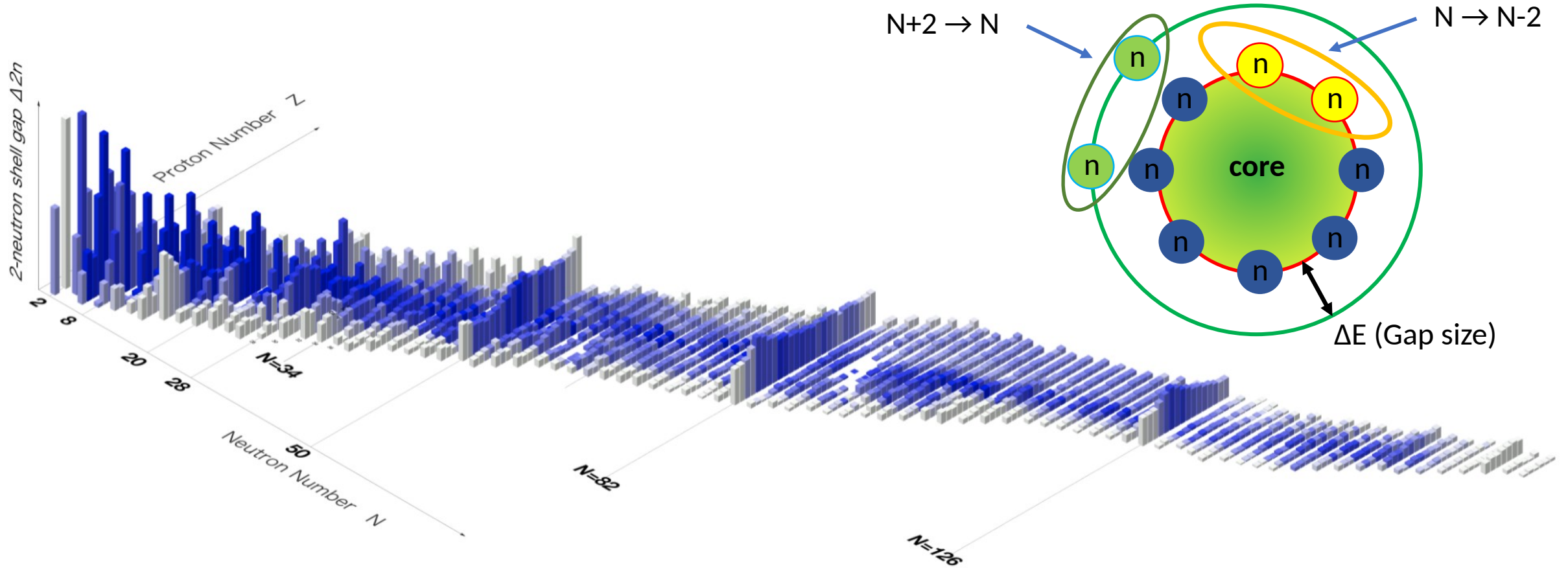
RIKEN Nishina Center for Accelerator-based Science

2-1 Hirosawa, Wako, Saitama 351-0198, Japan

- **Little starter: a short overview of MRTOF setups at the RIBF facility**
- **MRTOF-MS and the new setup at BigRIPS/SLOWRI**
- **Essential tools to save our bread**
- **Timeline of the ZD MRTOF experiment**
- **Great recent results!**
- **How about future developments?**



Masses for nuclear structure and astrophysics



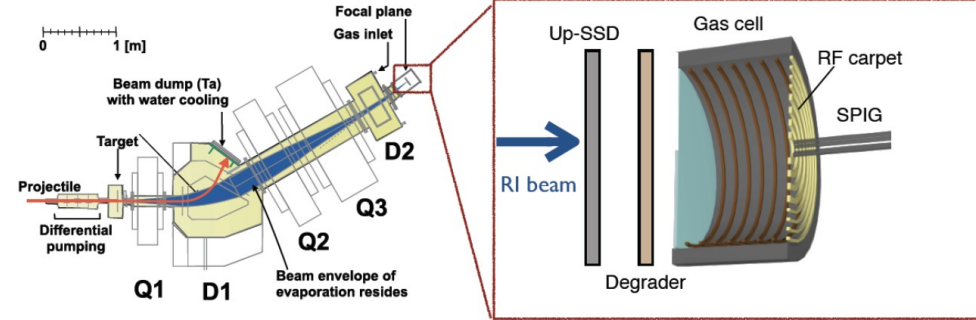
**Nuclear structure
by finite difference formulae**

$$S_{2n}(N, Z) = m(N-2, Z) - m(N, Z) + 2m_n$$

$$\Delta_{2n}(N, Z) = S_{2n}(Z, N) - S_{2n}(Z, N+2)$$

Current MRTOF facilities at RIBF

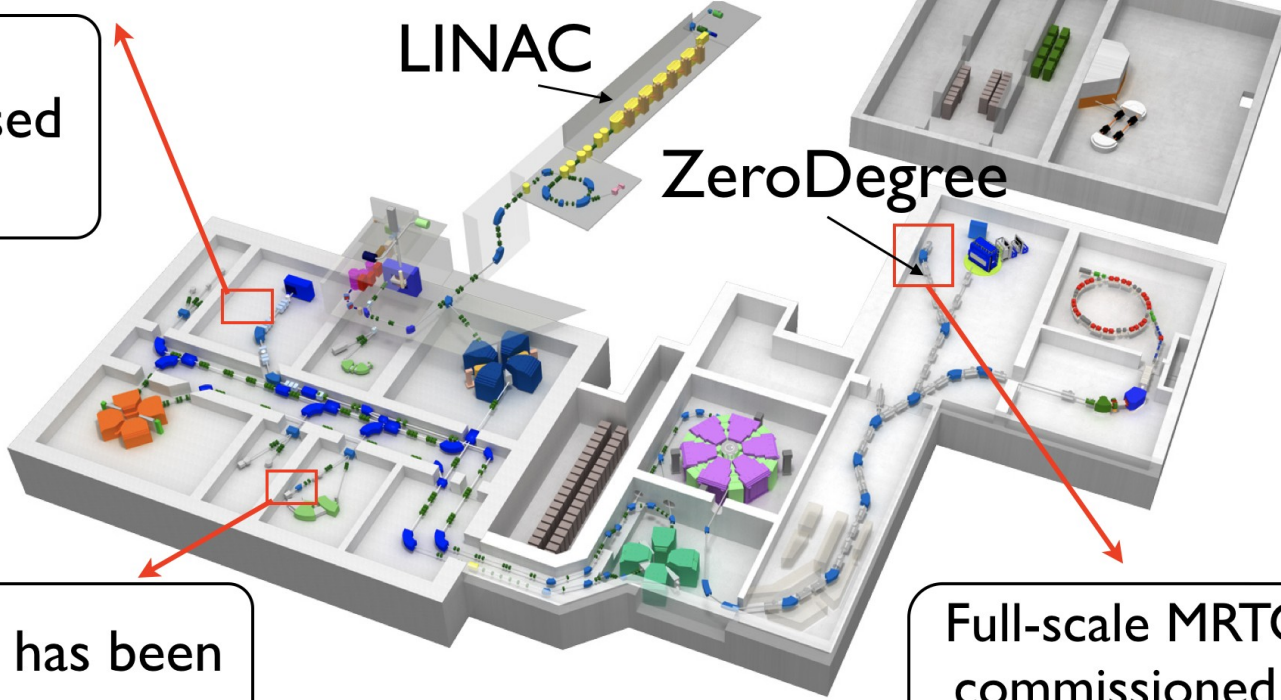
ガス充填型反跳核分離装置: GARIS-II (by GARIS team)



GARIS-III MRTOF coming soon



Original full-scale MRTOF primarily used for SHE studies



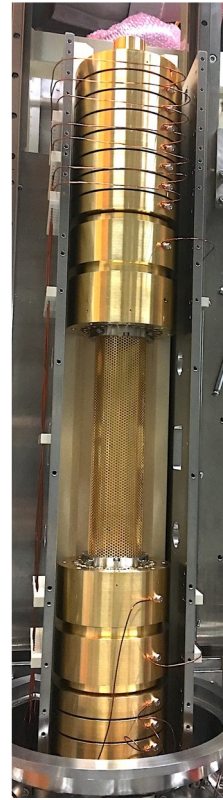
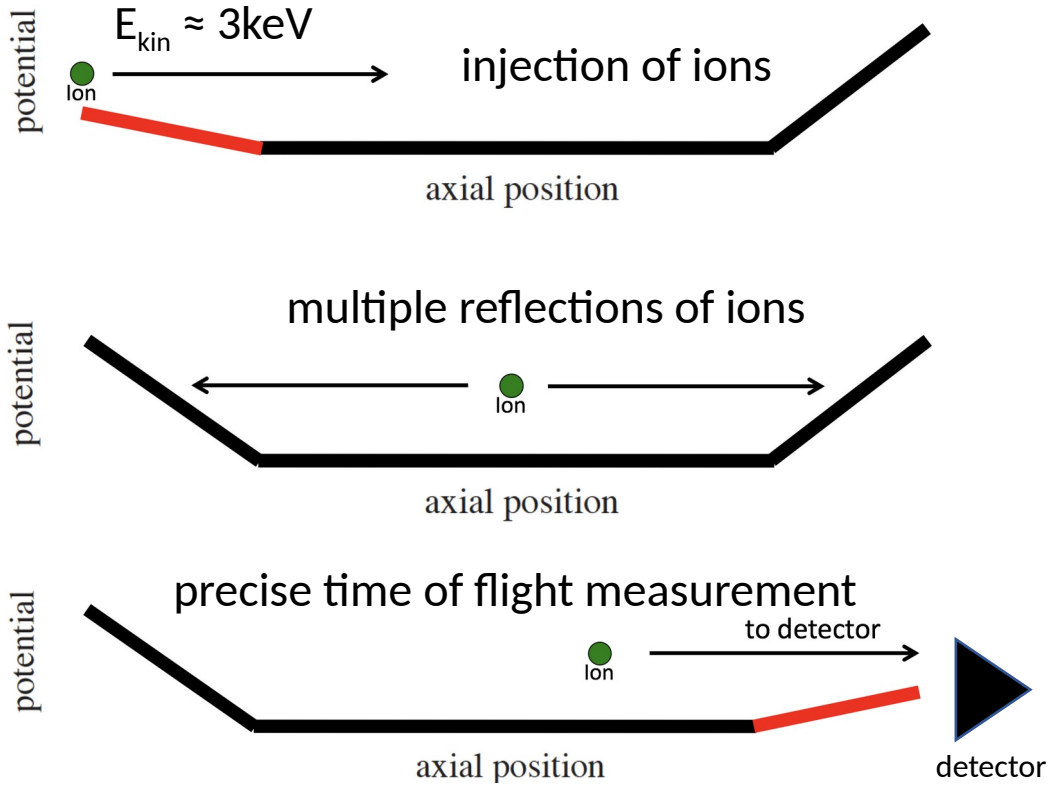
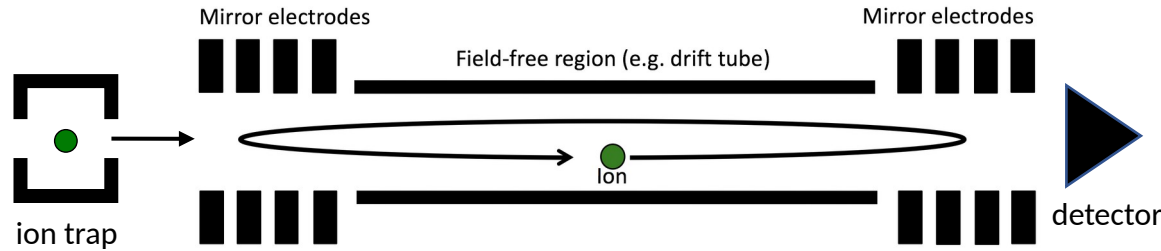
Half-scale MRTOF has been useful in mass-spectroscopy-assisted laser spectroscopy

Full-scale MRTOF has been commissioned commensal to in-beam gamma-ray spectroscopy

- Full scale = 1m
- Half-scale = 50cm

MRTOF-MS and the new setup at BigRIPS/SLOWRI

H.Wollnik and M. Przewloka, Int. J. Mass Spectrom. Ion Proc. 96, 267 (1990)



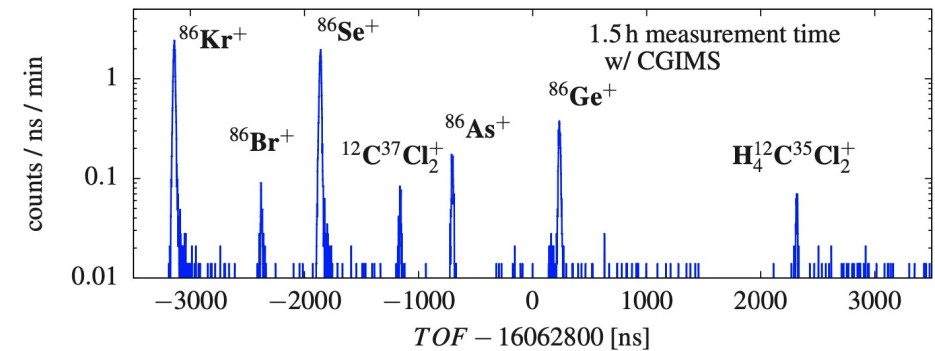
Total time of flight predominantly determined by the **electrostatic term** of the system

$$\overline{t(q, m)} = A \cdot \sqrt{\frac{m}{q}}$$

average from ion distribution

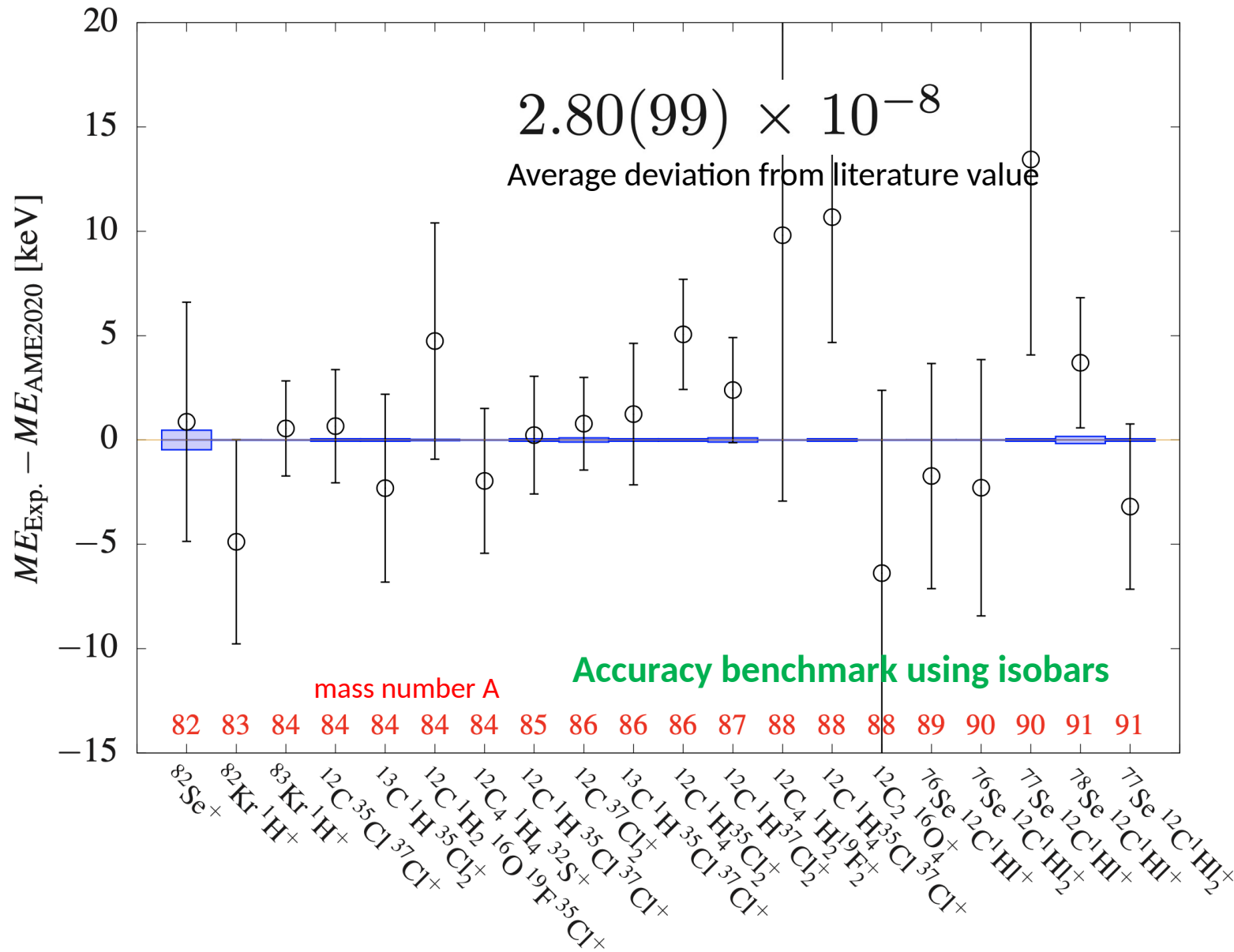
device constant

electrostatic contribution



- Flight path of a few kilometers possible
- Short measurement time $\approx 10\text{-}20$ ms
- Mass resolving power $R_m > 100,000$
- Relative mass precision $\delta m/m < 10^{-7}$
- High accuracy (acceptable syst. effects)

Mass accuracy study (isobaric referencing):



Wideband mass accuracy?

- Yes, possible and formerly used with dedicated t_0 calibration
- However: we must be aware, MRTOF-MS is not fully electrostatic

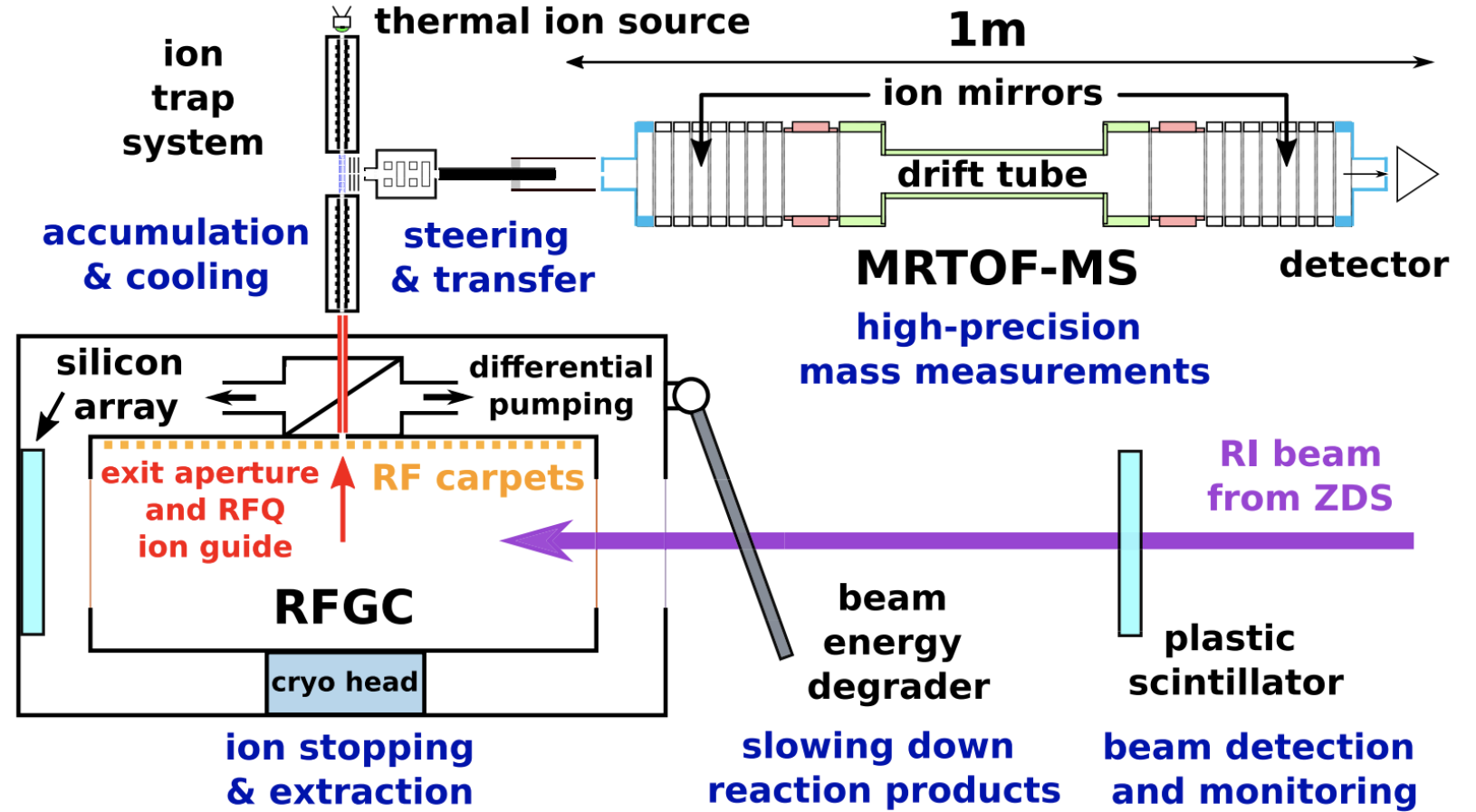
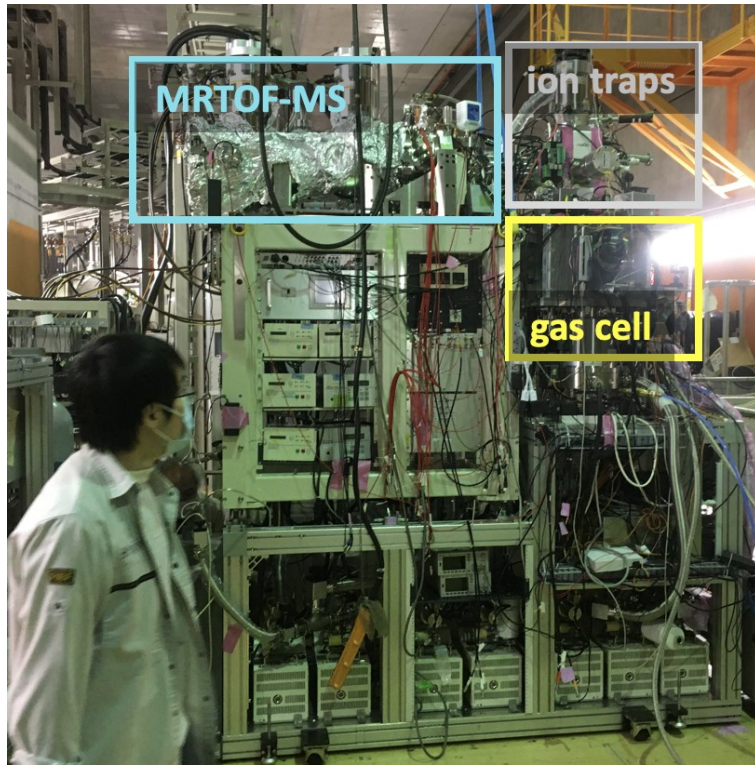
$$\overline{t(q, m)} = \underbrace{A \cdot \sqrt{\frac{m}{q}}}_{\text{electrostatic contribution}} + \underbrace{t_0}_{\text{offset time}} + \underbrace{t_{NS}(q, m)}_{\text{non-static fields}} + \underbrace{t_{\text{other}}(q, m)}_{\text{ion-trap physics and ToF detect.}}$$

device constant

average from ion distribution

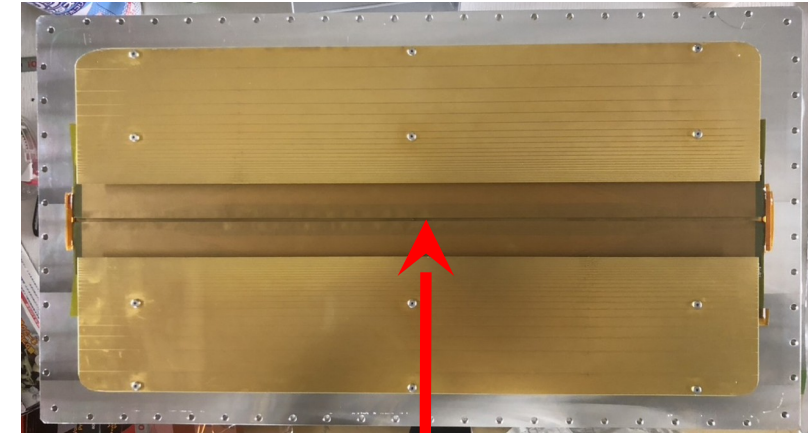
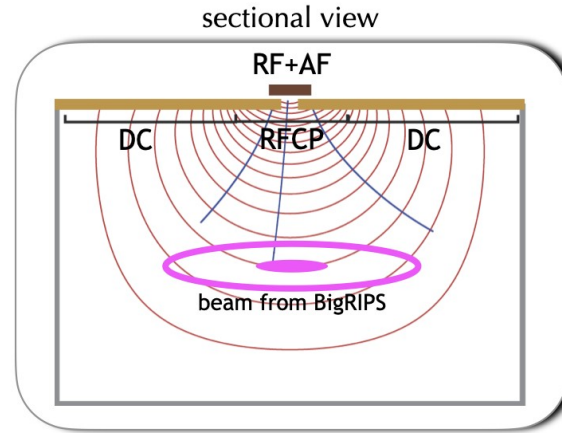
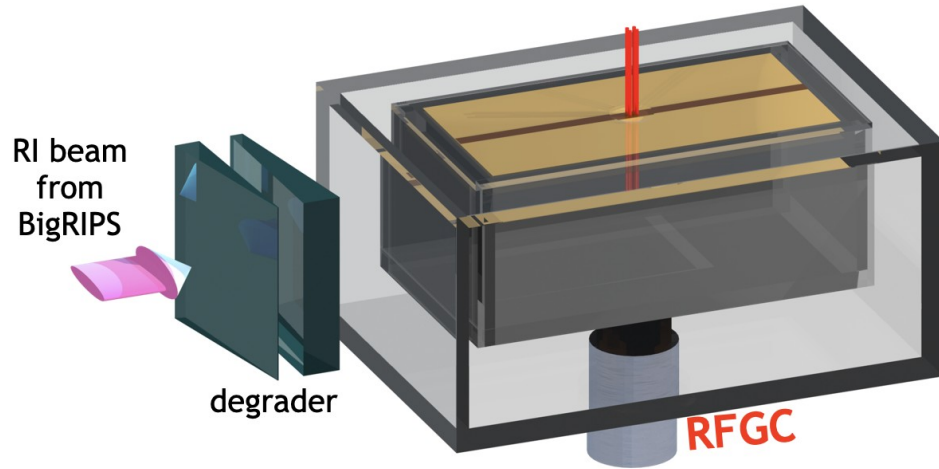
MRTOF-MS and the new setup at BigRIPS/SLOWRI

ZD MRTOF-MS overview

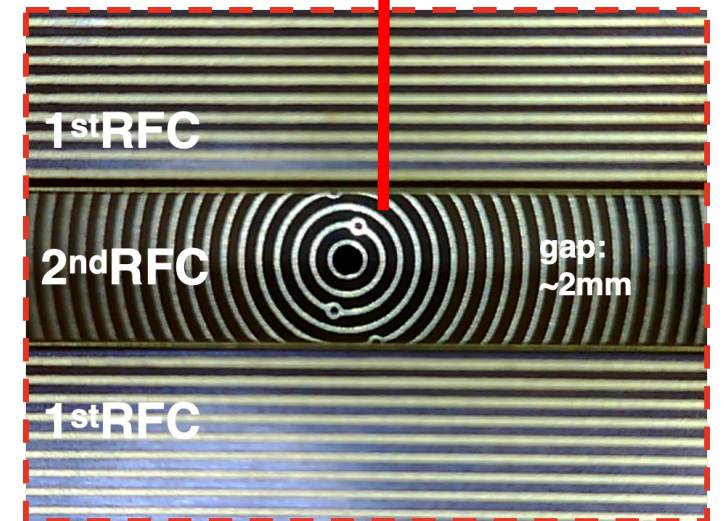
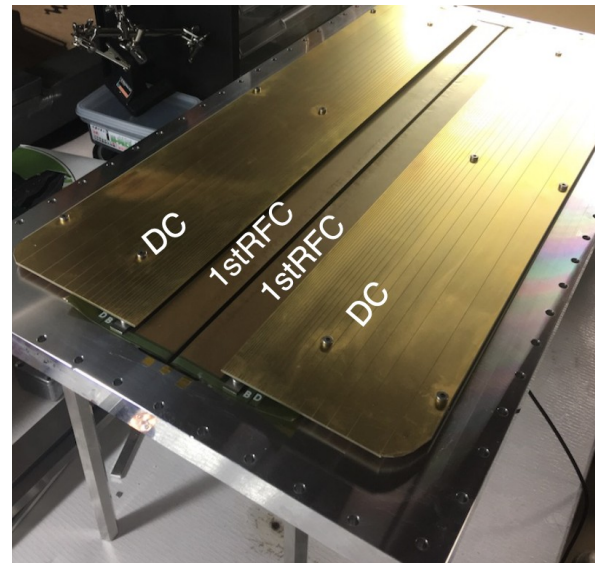


MRTOF-MS and the new setup at BigRIPS/SLOWRI

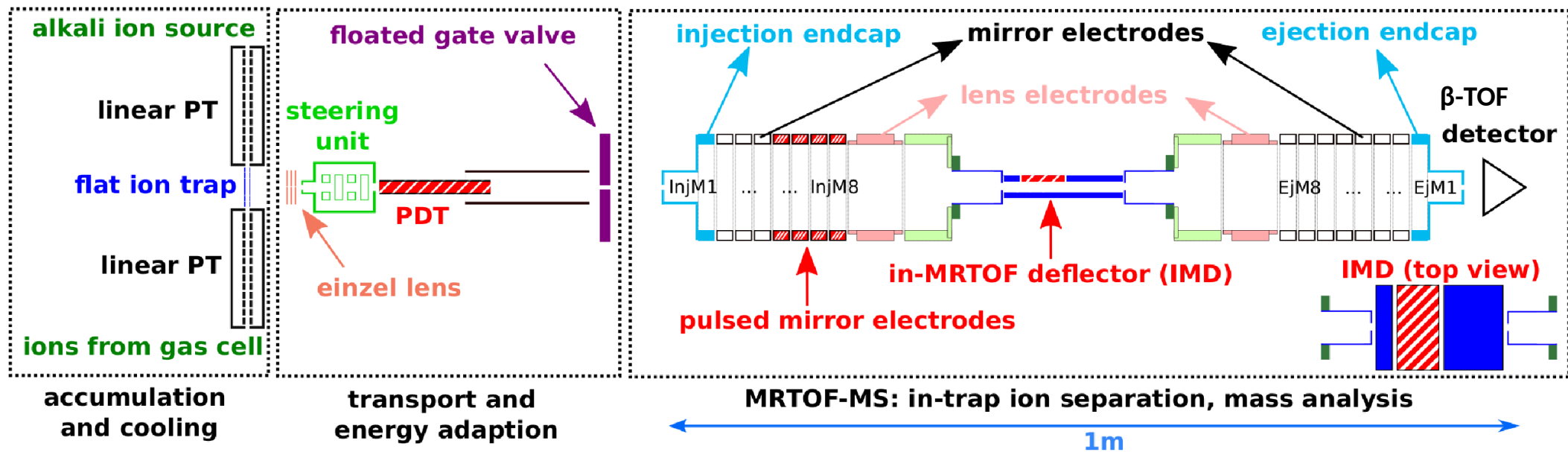
New He gas-filled ion catcher (or also “radiofrequency gas cell” RFGC)



- Beam-energy degrader is used to slow down the beam
- If energy matches the stopping power of the gas, the ions will stop inside the volume

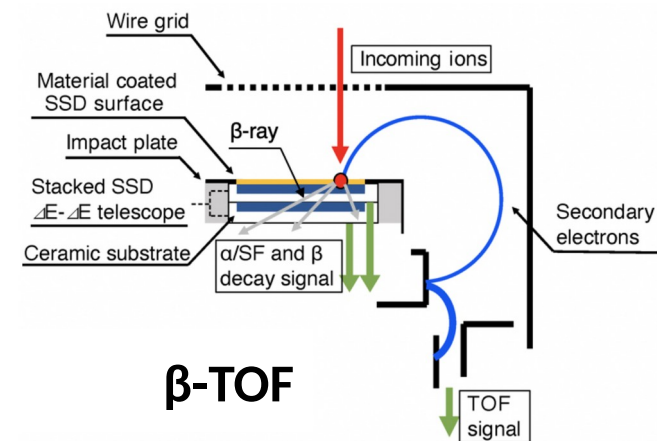
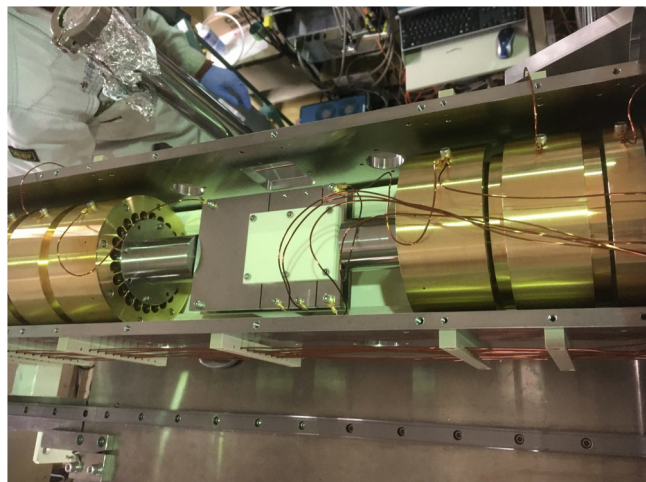


MRTOF-MS and the new setup at BigRIPS/SLOWRI



Essential tools

- Energy adaption available by pulsed drift tube
- In-trap ion separation available (two methods tested)
- Beta-TOF detection available

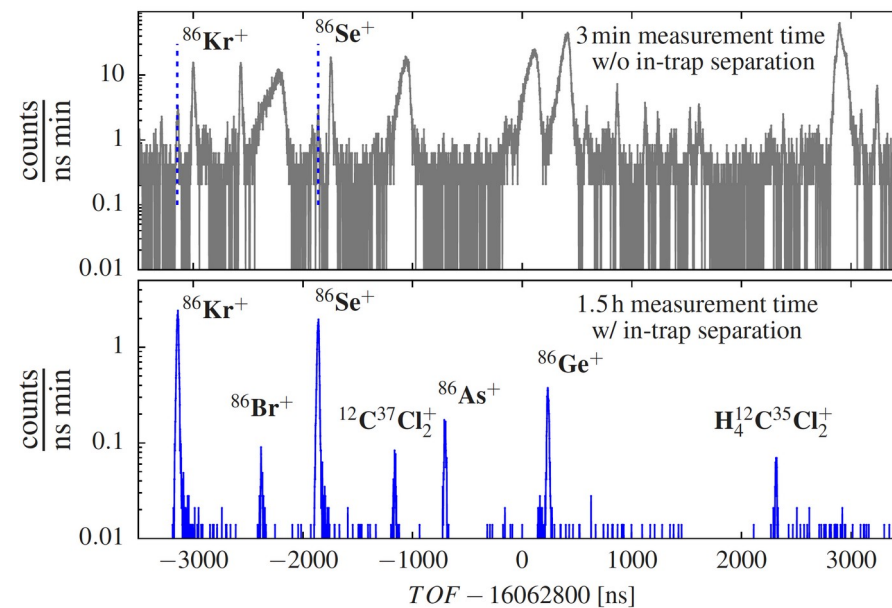
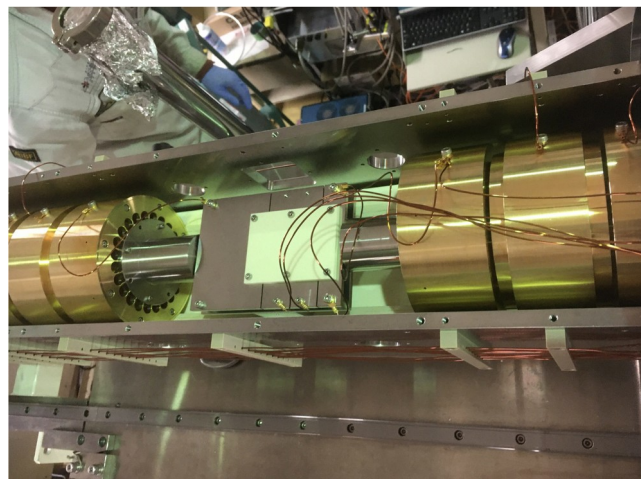
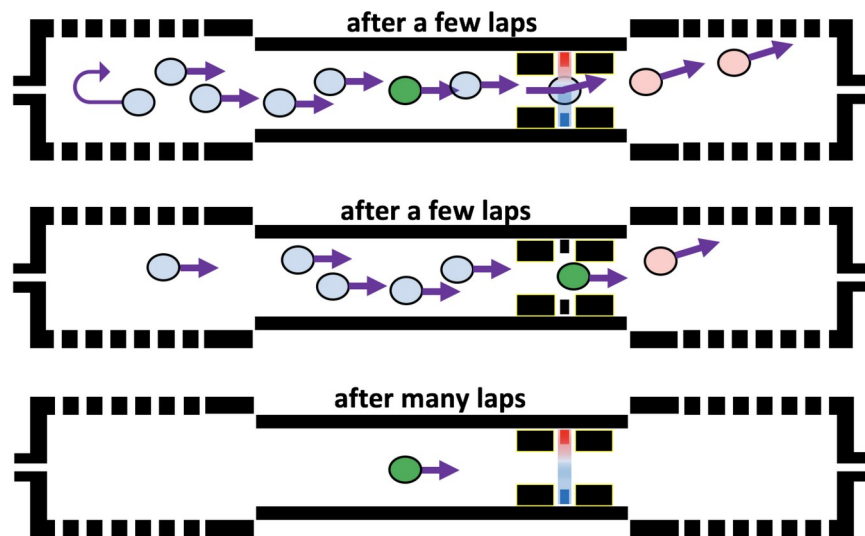


M. Rosenbusch *et al.*, Nucl. Instr. Meth. A 1047, 167824 (2023)

Essential tools: In-MRTOF deflector

- Selective kick-out possible
- **Selective protection of several masses now possible**
- Proper design: Kick out of unwanted ions with weak (20 V) pulse

● wanted mass ○ kicked out
● unwanted mass



First MRTOF in-trap cleaning with deflector:
Y. Toker et al., J. Instrum. 4, P09001 (2009).

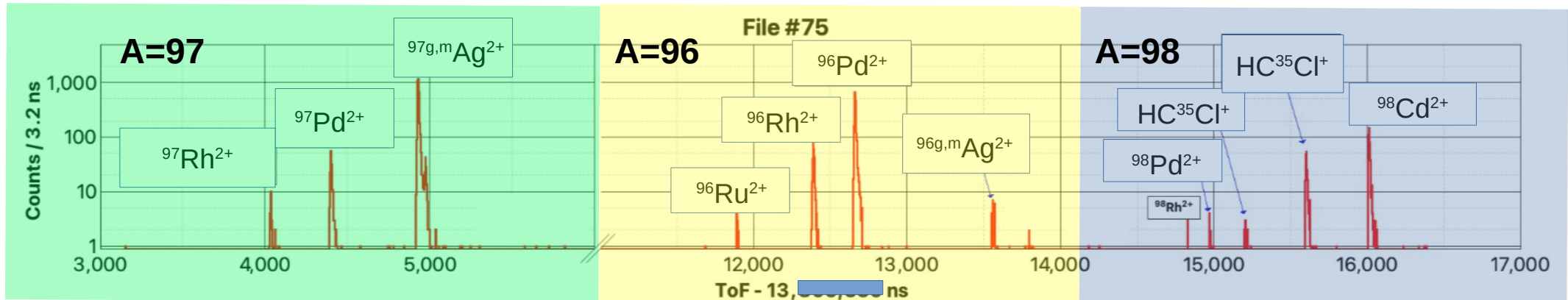
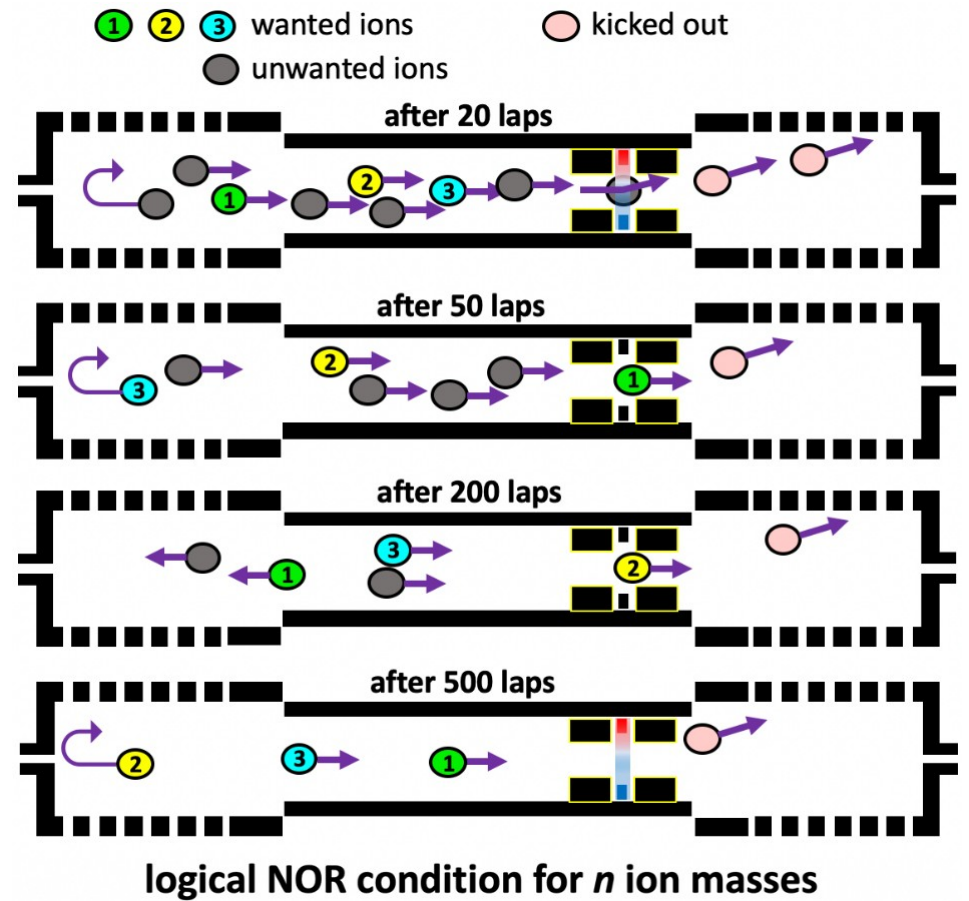
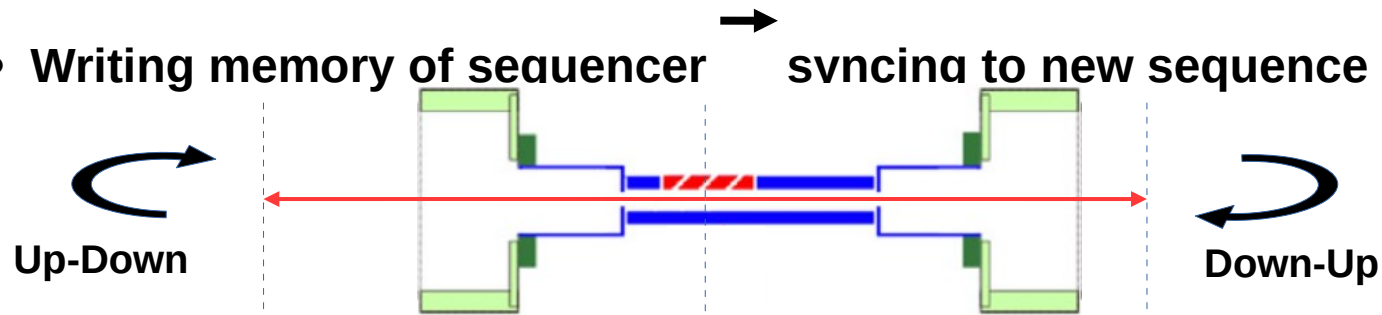
Development of deflectors:
T. Dickel et al., Nucl. Instrum. Meth. 777, 172 (2015).
P. Fischer et al., Rev. Sci. Instrum. 89, 015114 (2018).

Usage of mirror endcaps:
J. T. Johnson et al., Anal. Chem. 91, 8789 (2019).

- Selecting various isobar groups at the same time
- Selecting the moments and number of pulses
- Calculating crossing time for all selected isotopes
- Defining “safety region” (distance from IMD), including asymmetry of motion (Down-Up / Up-Down)

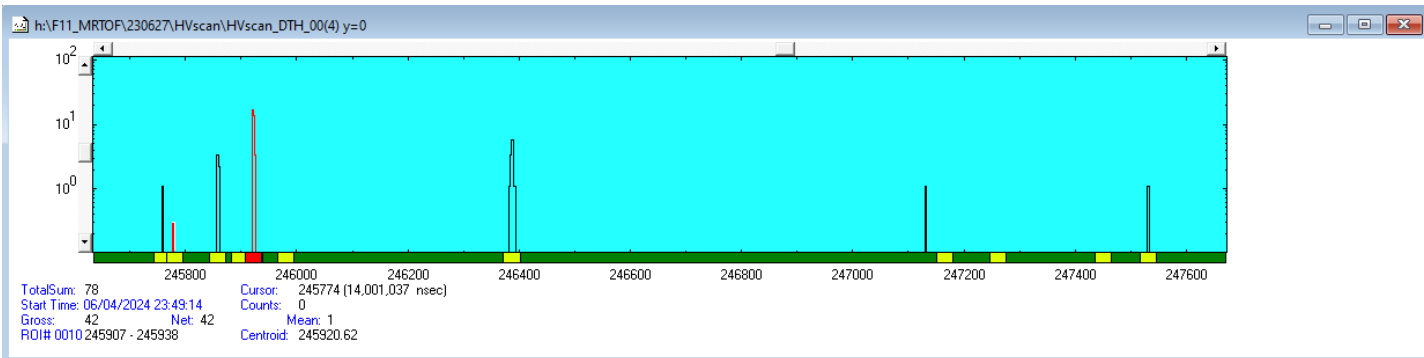
- Generating pulse-train sequence

- Writing memory of sequencer → svncina to new sequence



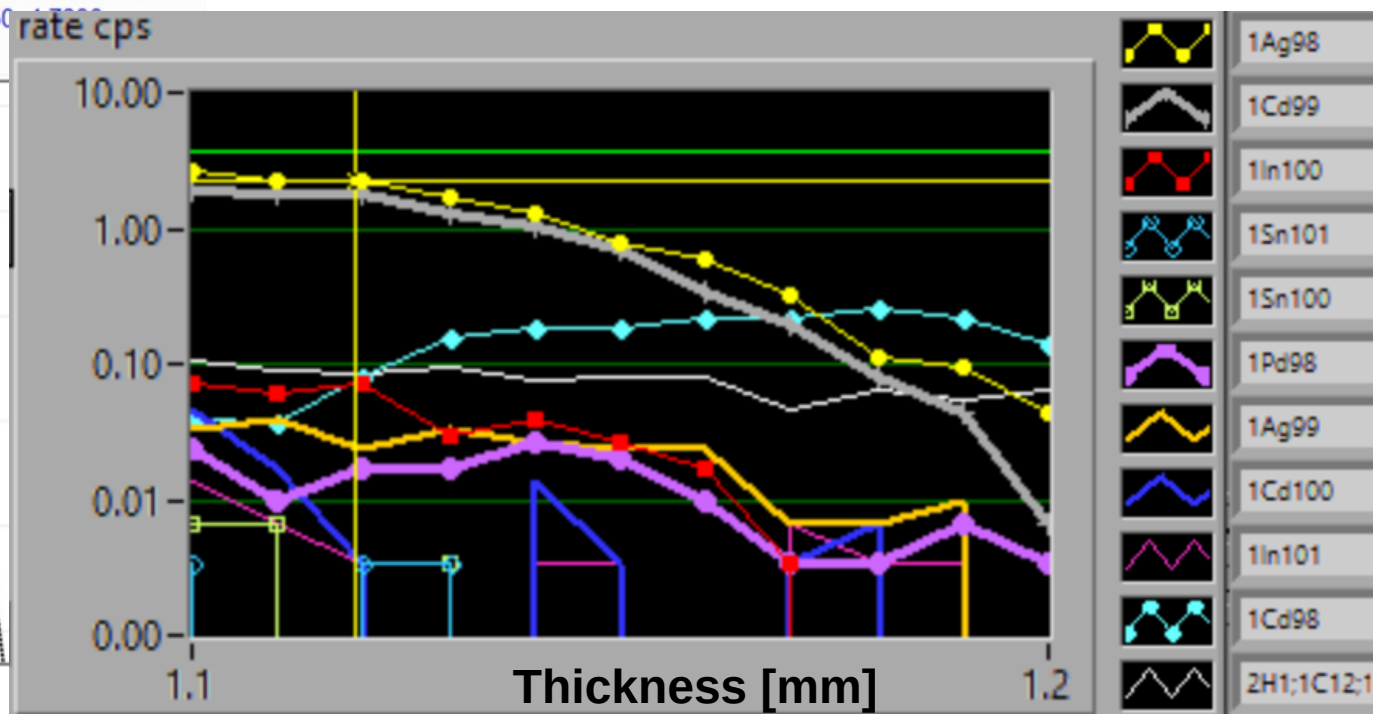
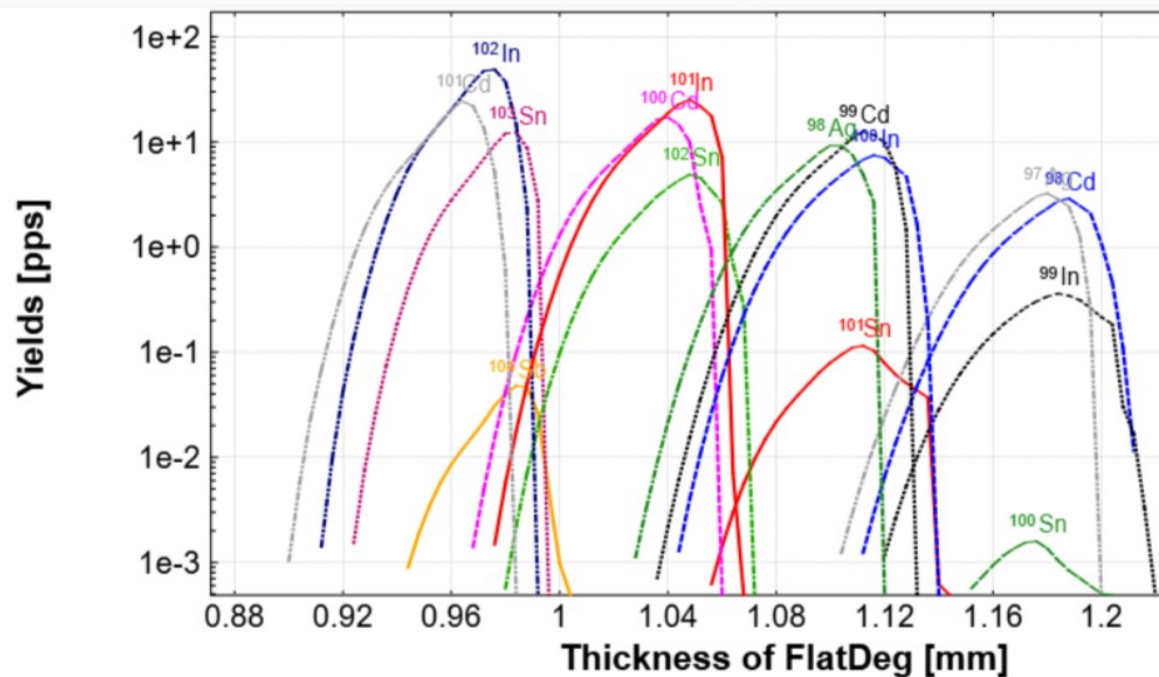
Essential tools: In-MRTOF deflector

- Easy-to-go degrader scanning
- Focusing on several isotopes simultaneously
- Background-free measurement



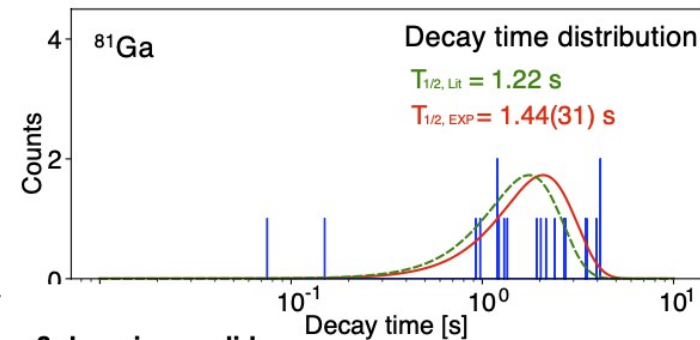
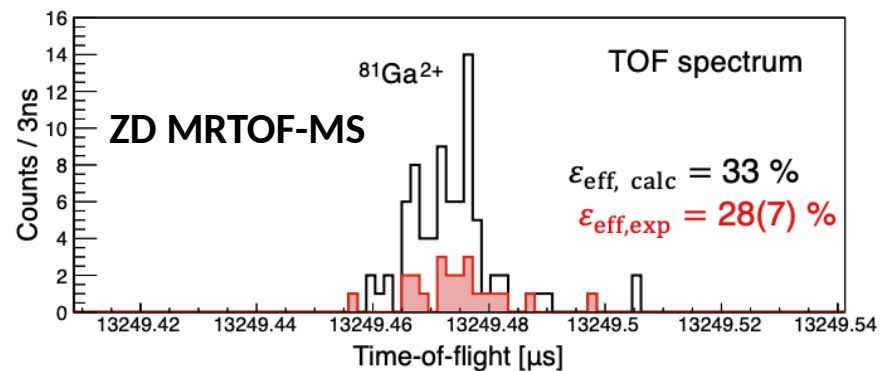
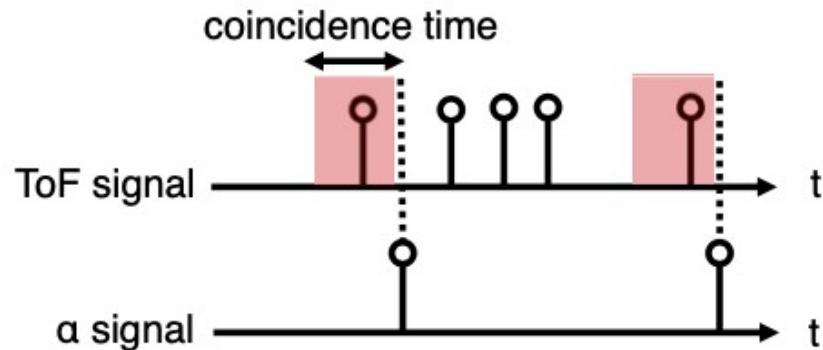
Range 1D-Optimizer: Isotopes stopped in gascell

^{124}Xe (345 MeV/u) + Be (3 mm); Settings on ^{101}Sn ; Config: D^{bbb} D^{bbb} DD^m DD^m dp/p=3.36%; Wedge(s): Al (2 mm), Al (1 mm), Al (0.43 mm); Bp (Tm): 5.4335, 5.0342, 4.9850, 4.9850
Yield_{max} = 1.13e-01 pps @ 1.11 mm

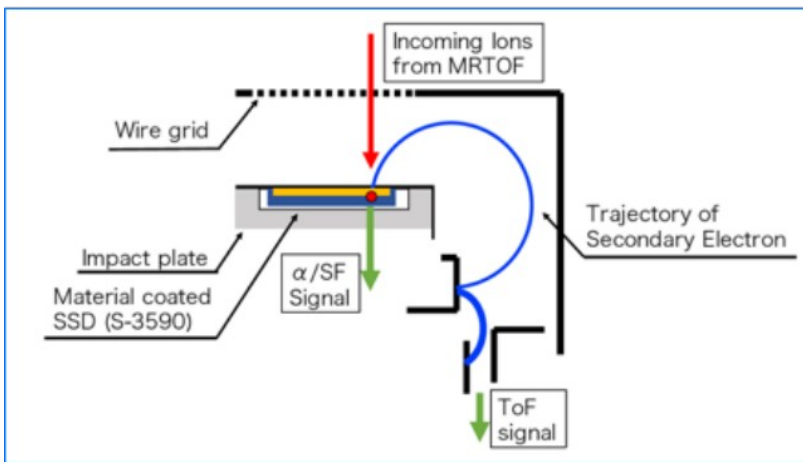


Essential tools: α/β - TOF detector

ToF-decay correlated mass spectrometry

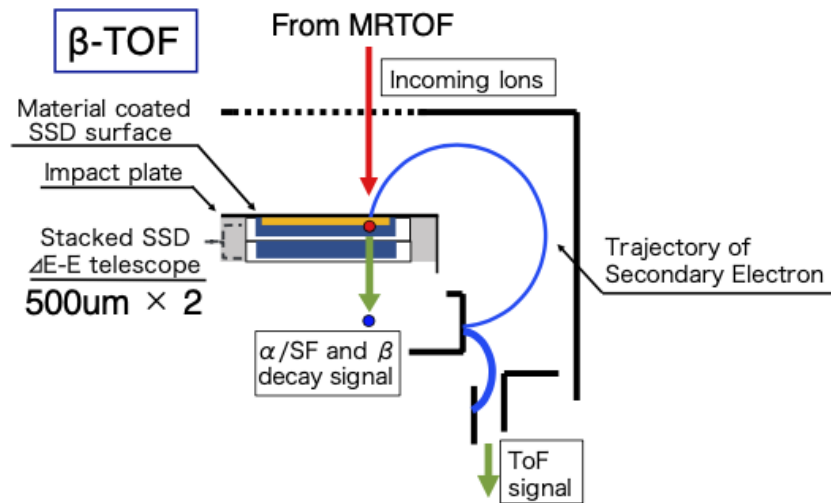


Alpha ToF



Coated material : Au +MgO (or Al_2O_3)
 100Å + 100Å

β -TOF

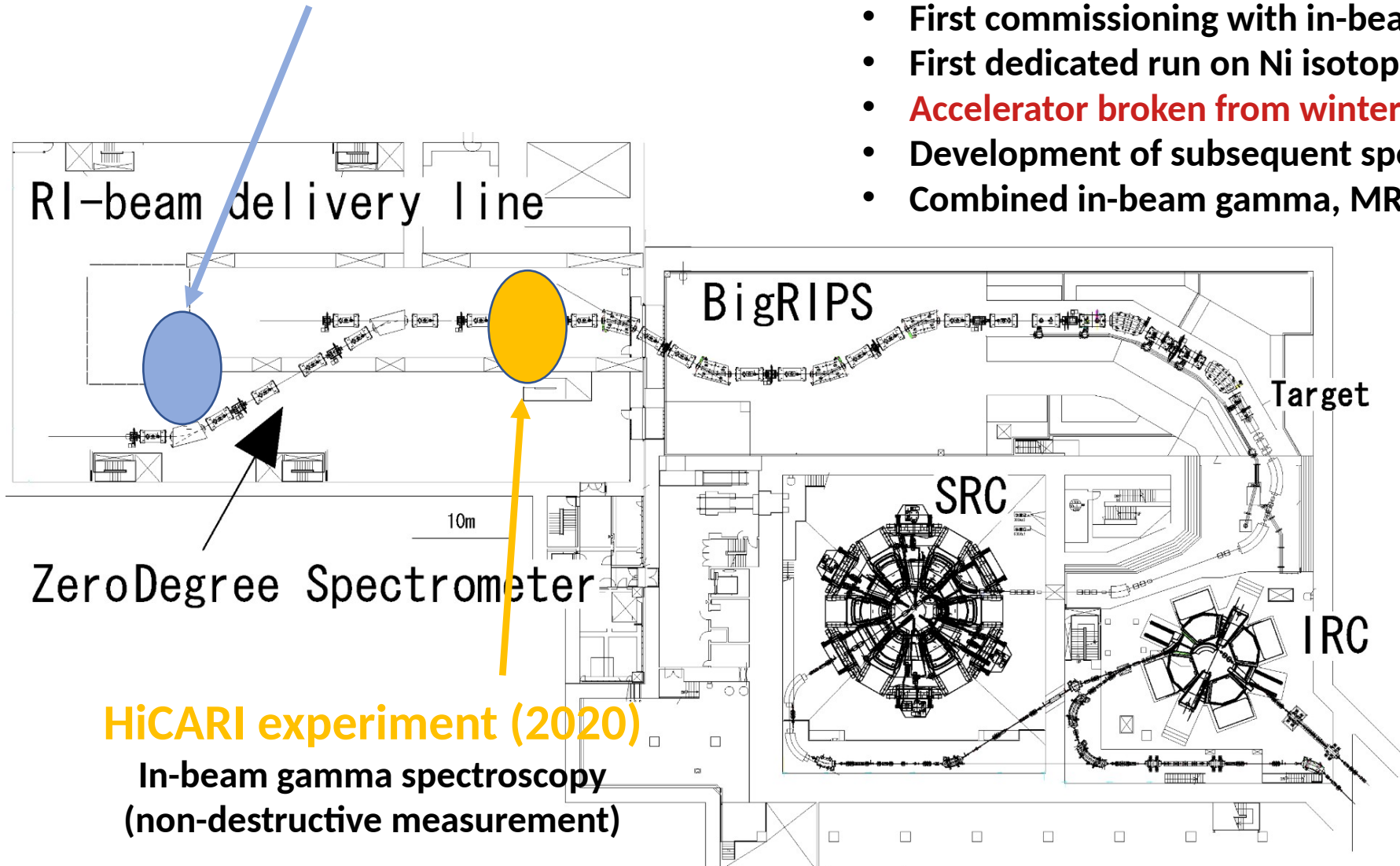


Correlation with TOF and β -signal
 (The β -signal is the one passing through the double layered Si.)



Timeline of the ZD MRTOF experiment

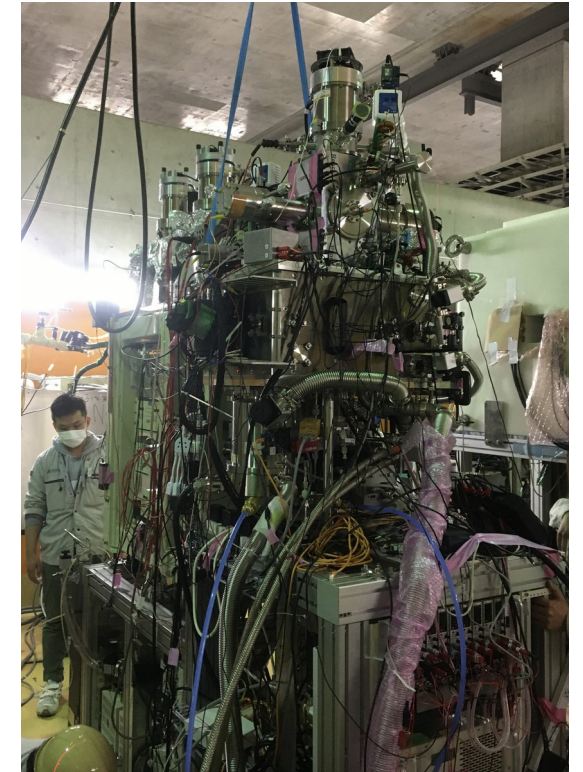
MRTOF-MS setup



HiCARI experiment (2020)

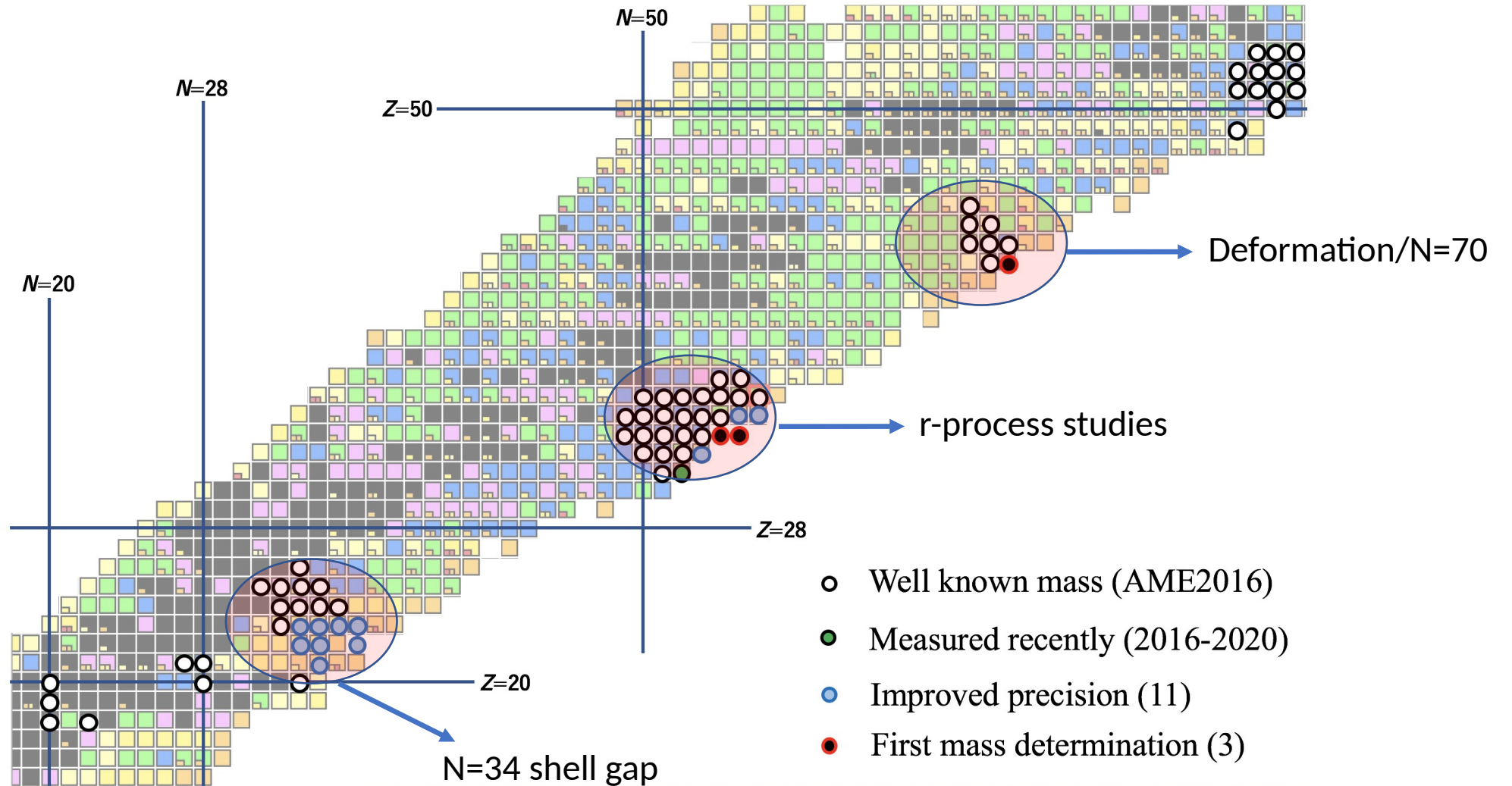
In-beam gamma spectroscopy
(non-destructive measurement)

- Basic assembly from 2018
- Starting offline GC development in 2019
- GC coupled to MRTOF-MS spring/summer 2020
- Transport to ZD spectrometer fall 2020
- First commissioning with in-beam gamma in winter 2020
- First dedicated run on Ni isotopes winter 2021 / spring 2022
- **Accelerator broken from winter 2022**
- Development of subsequent spectroscopy during 2023
- Combined in-beam gamma, MRTOF-MS, and decay spec. 2024



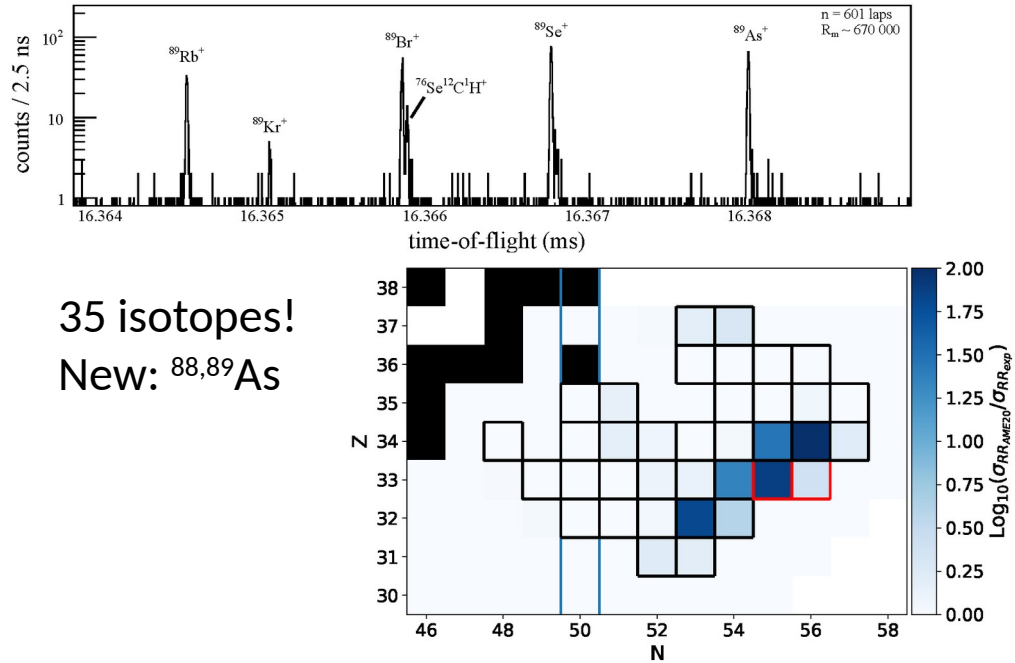
Timeline of the ZD MRTOF experiment

- ~ 70 mass measurements in different regions of the nuclide chart
- Three nuclear masses measured for the first time
- Eleven nuclear masses improved in precision
- Total system efficiency measured (0.3% - 1.5%)



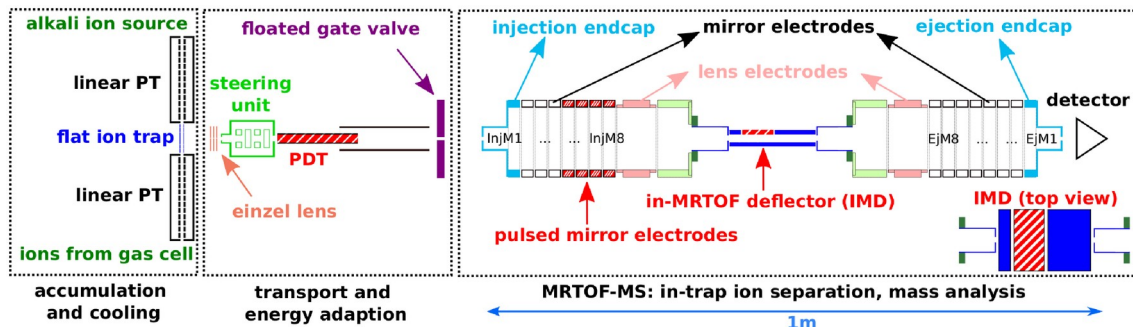
Timeline of the ZD MRTOF experiment

W. Xian, S. Chen et al., Phys. Rev. C 109, 035804 (2024)

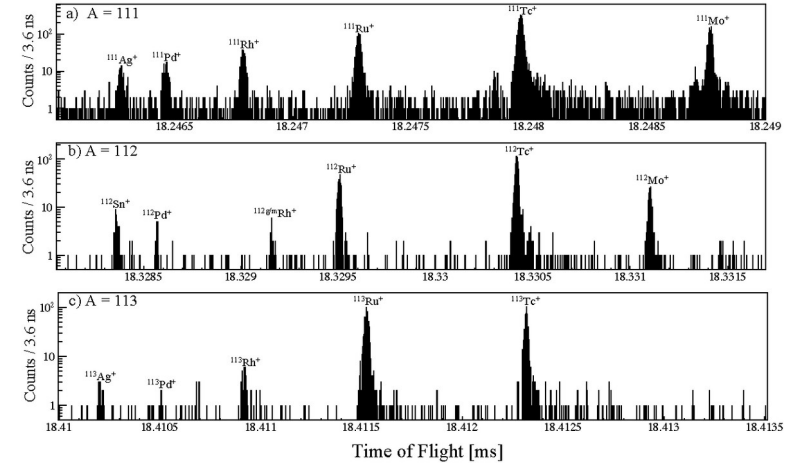


35 isotopes!
New: $^{88,89}\text{As}$

M. Rosenbusch et al., Nucl. Instr. Meth. A 1047, 167824 (2023)

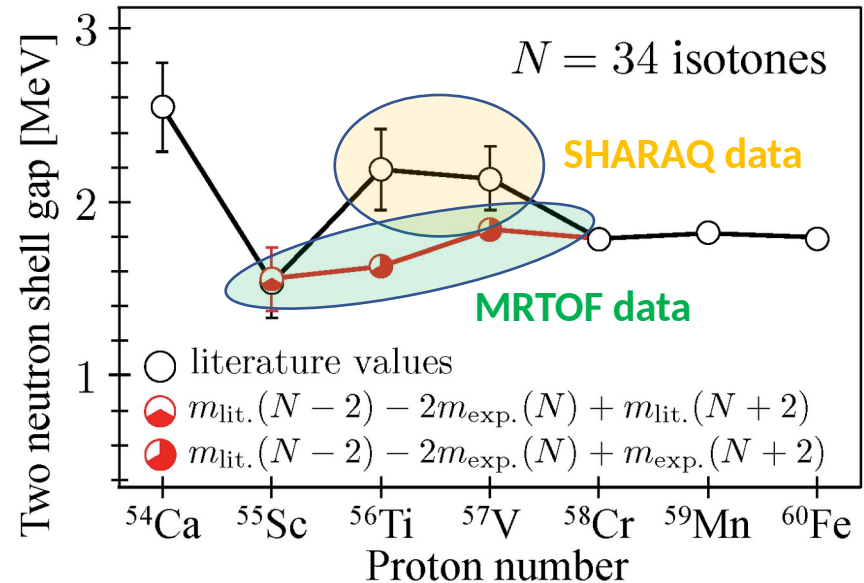


D.S. Hou, A. Takamine et al., Phys. Rev. C 108, 054312 (2023)



14 isotopes!
New: ^{112}Mo

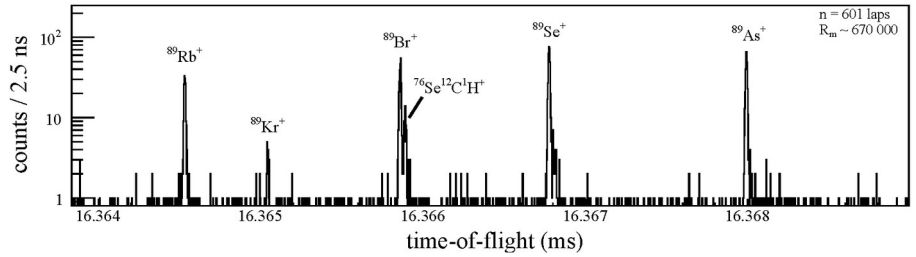
S. Iimura, M. Rosenbusch et al., PRL 130, 012501 (2023)



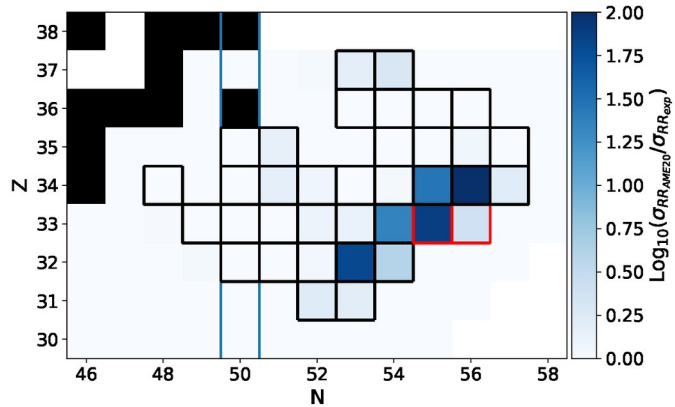
19 isotopes!
New: $N=34$ shell gap

Timeline of the ZD MRTOF experiment

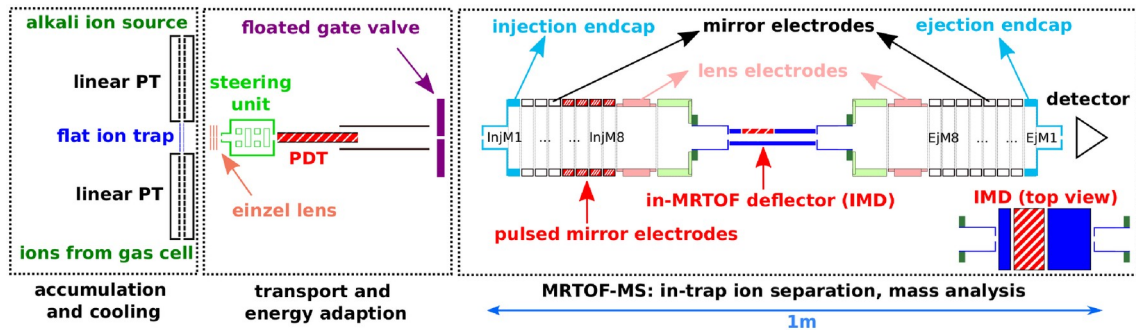
W. Xian, S. Chen et al., Phys. Rev. C 109, 035804 (2024)



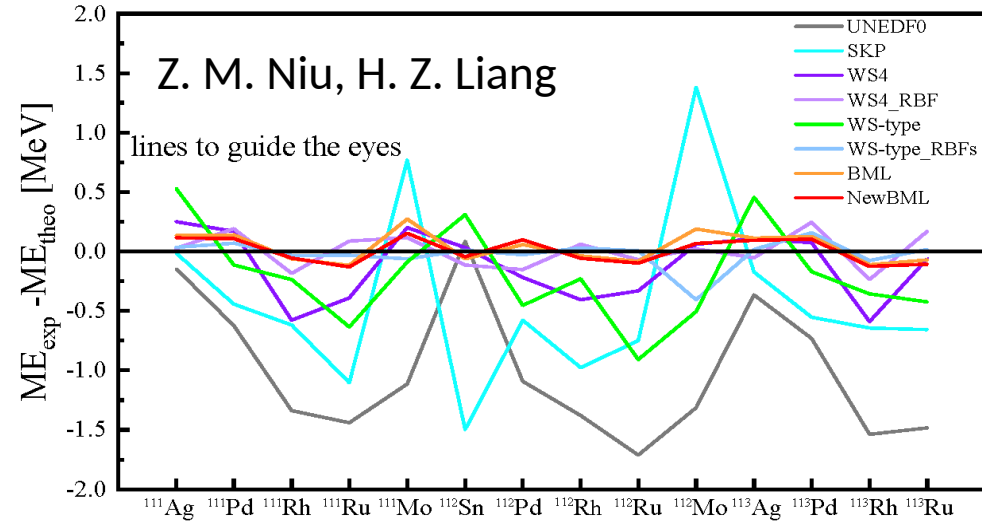
35 isotopes!
New: $^{88,89}\text{As}$



M. Rosenbusch et al., Nucl. Instr. Meth. A 1047, 167824 (2023)

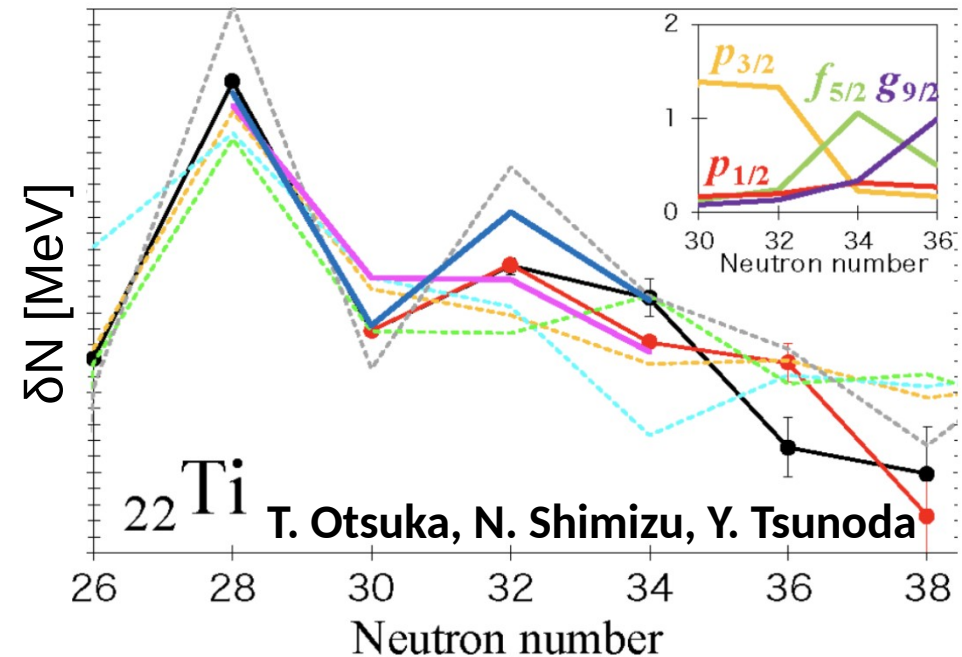


D.S. Hou, A. Takamine et al., Phys. Rev. C 108, 054312 (2023)



14 isotopes!
New: ^{112}Mo

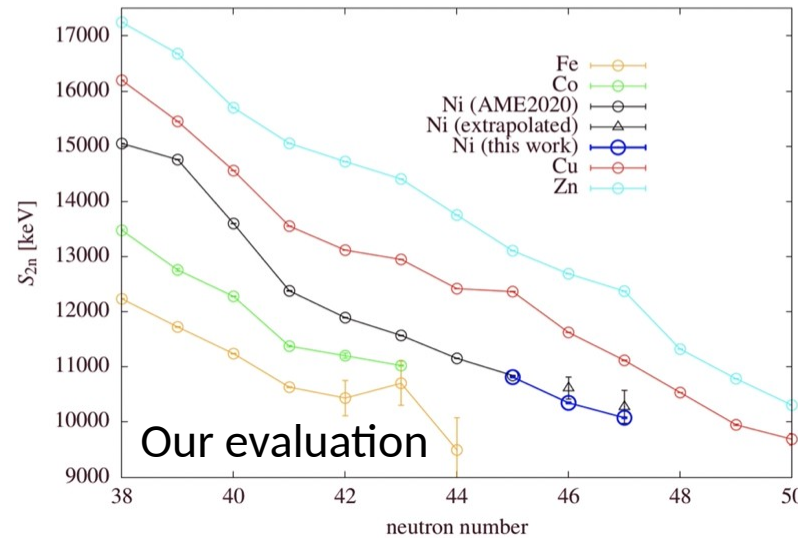
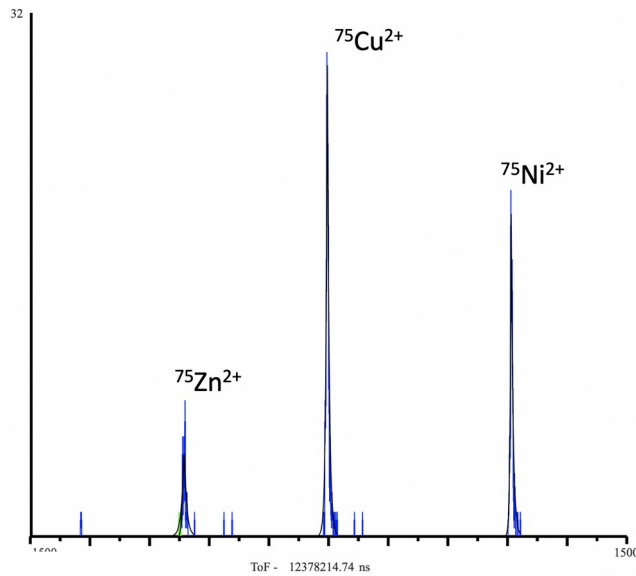
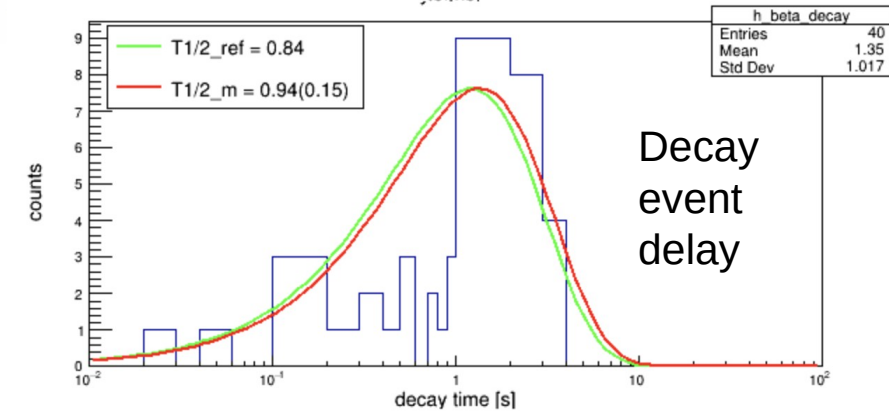
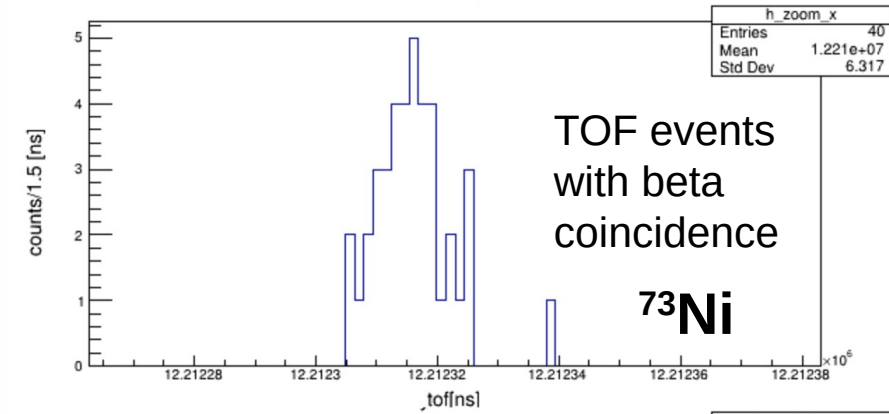
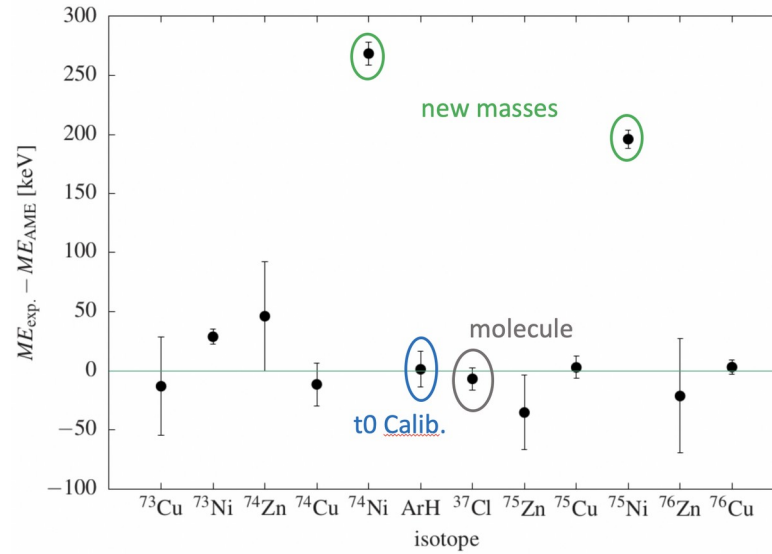
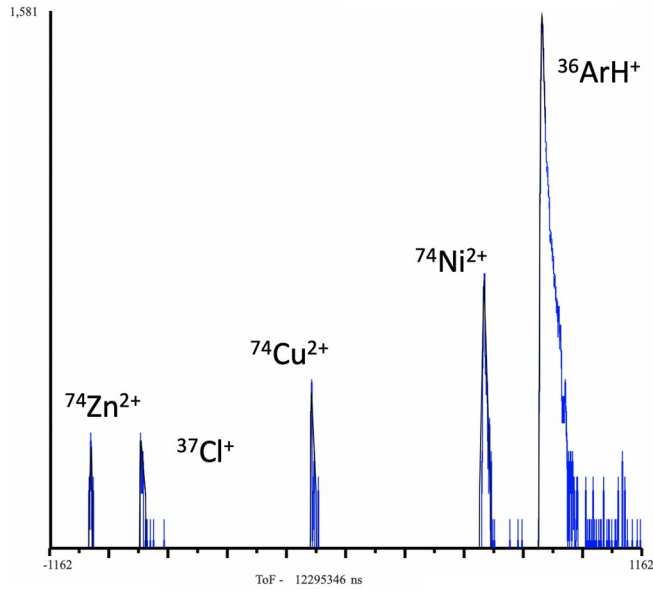
S. Iimura, M. Rosenbusch et al., PRL 130, 012501 (2023)



19 isotopes!
New: N=34
shell gap

Timeline of the ZD MRTOF experiment

Mass measurements of $^{73-75}\text{Ni}$, and beta-TOF POP measurements

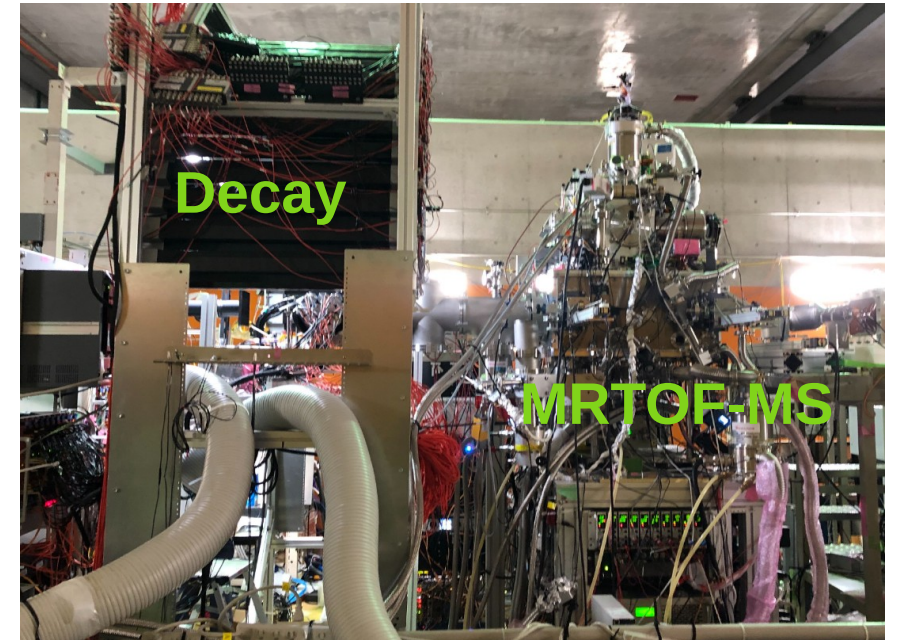
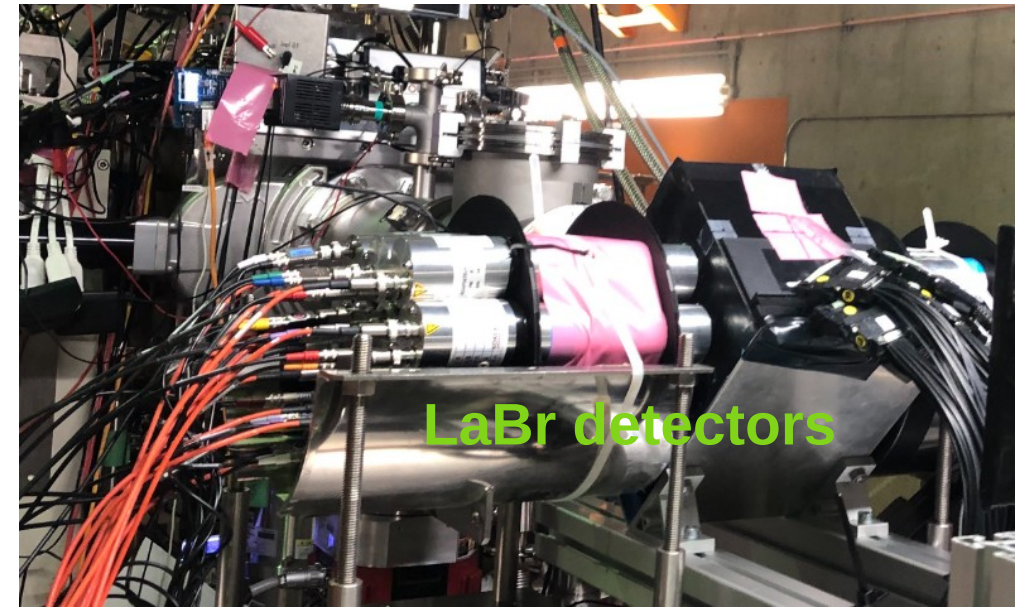
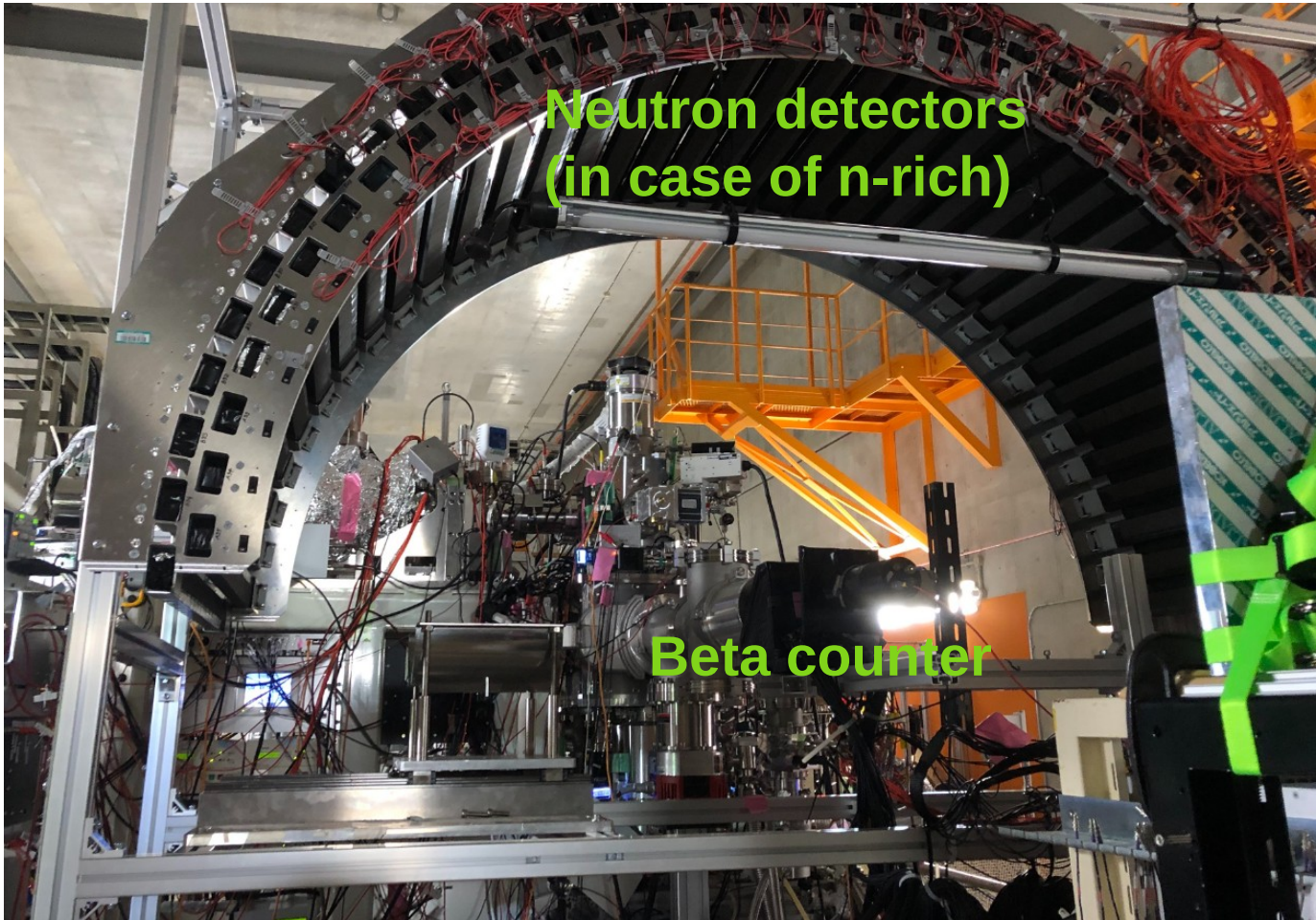


$^{74,75}\text{Ni}$ also recently published by JYFLTRAP: S. Giraud *et al.*, Phys. Lett. B 833, 137309 (2022)

Timeline of the ZD MRTOF experiment

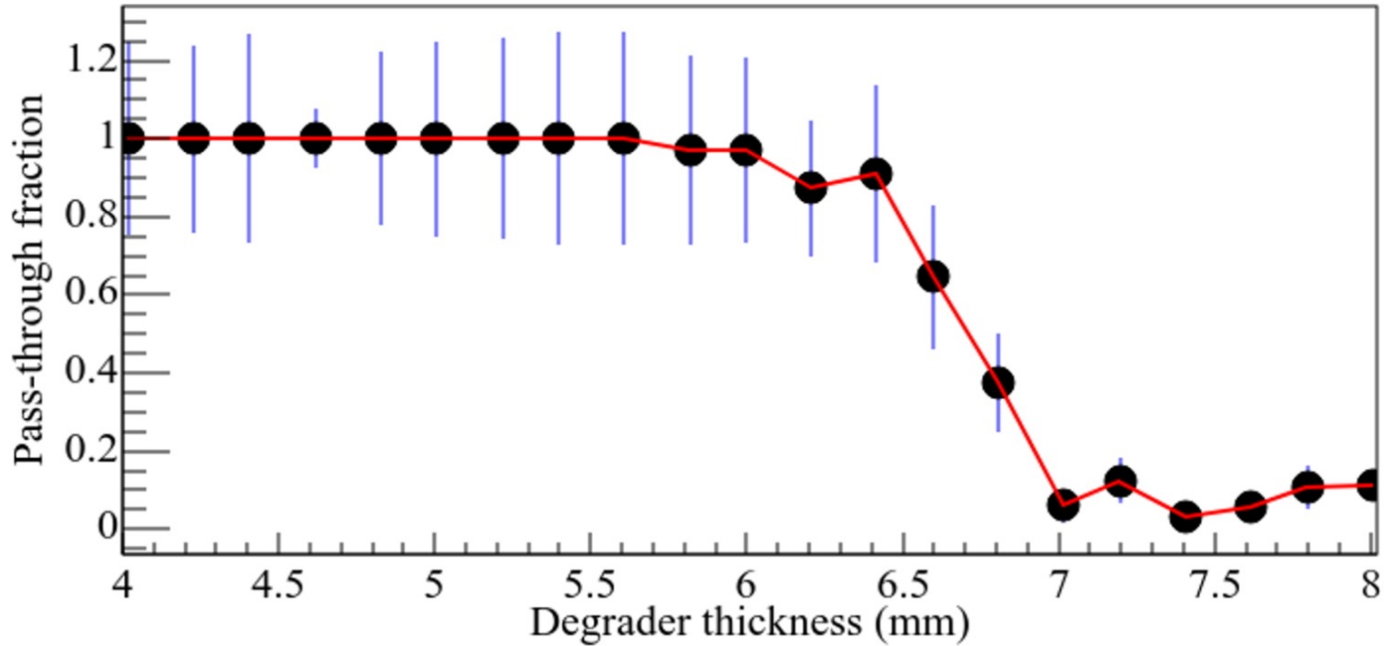
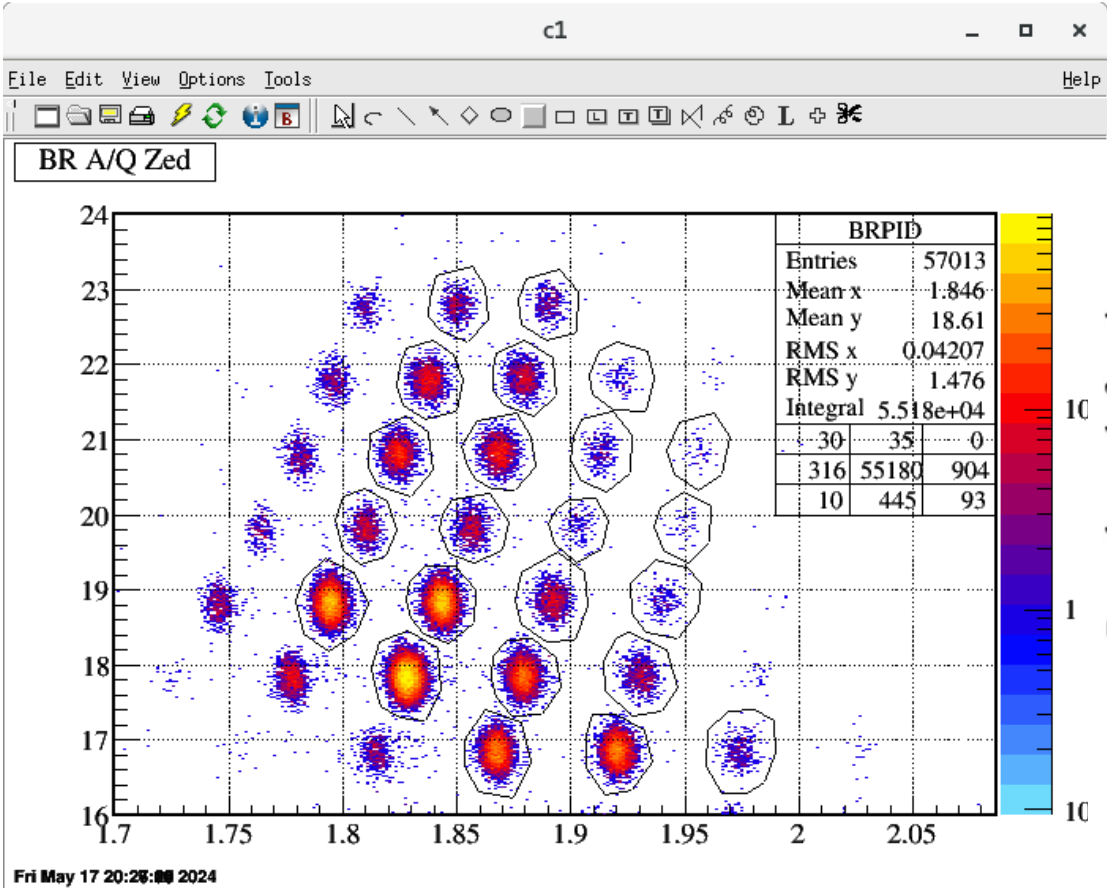
2023-2024

Development of a vacuum beam line
for subsequent decay spectroscopy



TOF and beta-TOF signal added to same DAQ (CAEN waveform recorders)

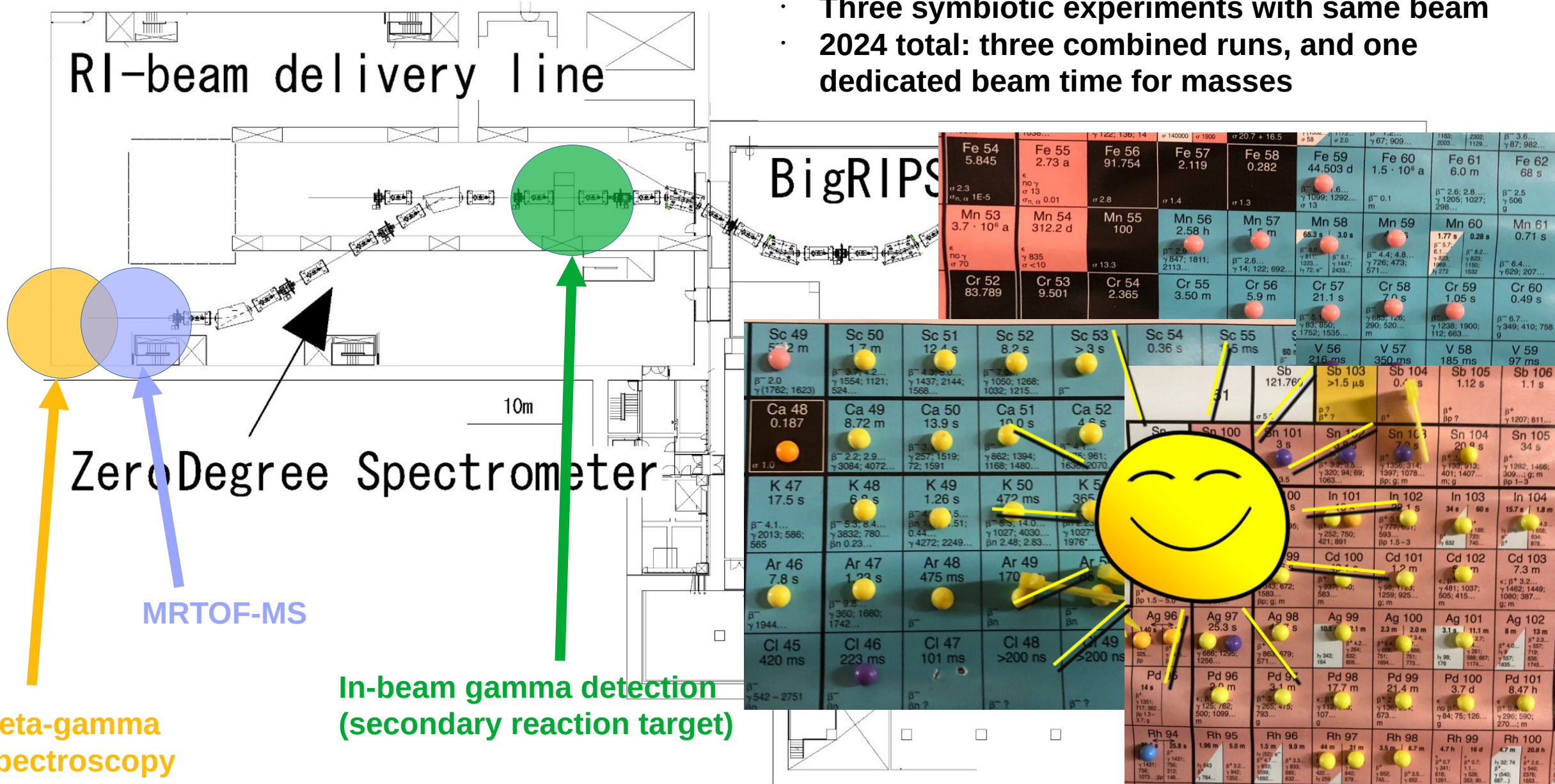
Timeline of the ZD MRTOF experiment



Great recent results!

And it works!

- Three symbiotic experiments with same beam
- 2024 total: three combined runs, and one dedicated beam time for masses



Great recent results!

Little disclaimer:

Our pins mean: "Candidates"! They don't necessarily tell that it is already a proper (publishable) measurement. ;-)

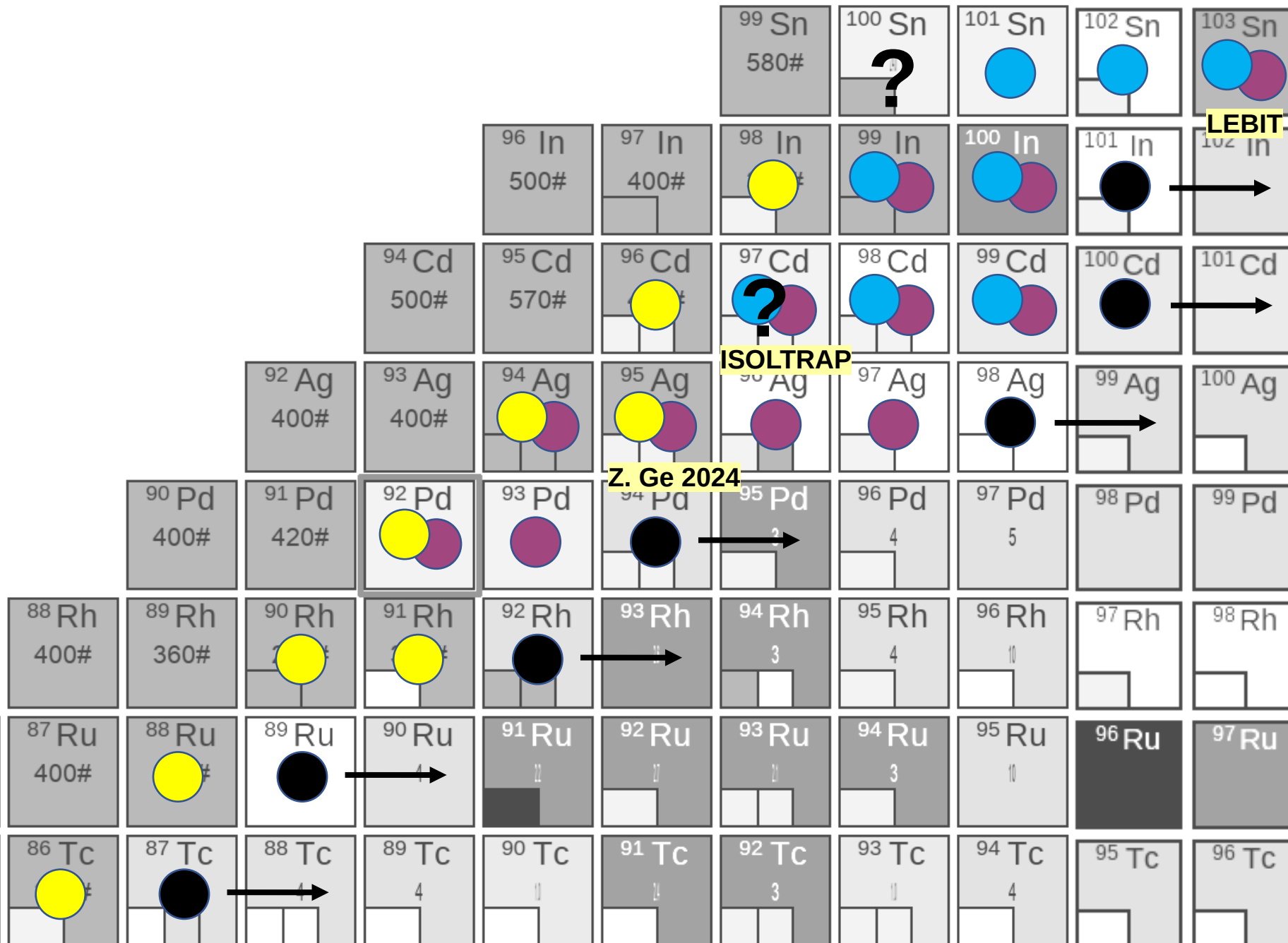


		Sb 121.760	Sb 103 >1.5 μs	Sb 104 0.1 s	Sb 105 1.12 s	Sb 106 1.1 s
51		σ 5.2	$p?$ $\beta^+?$	β^+	β^+ $\beta p?$	β^+ γ 1207; 811...
Sn 118.710	Sn 100 1.1 s	Sn 101 3 s	Sn 102 3.8 s	Sn 103 7.9 s	Sn 104 20.9 s	Sn 105 34 s
γ 0.61	β^+ 3.4 γ	β^+ βp 2-3.5	β^+ 3.2; 3.5... γ 320; 94; 69; 1063...	β^+ γ 1356; 314; 1397; 1078...	β^+ 2.3 γ 133; 913; 401; 1407...	β^+ γ 1282; 1466; 309...; g; m βp 1-3
In 98 1.7 s 45 ms	In 99 3.1 s	In 100 5.9 s	In 101 16 s	In 102 22.1 s	In 103 34 s 60 s	In 104 15.7 s 1.8 m
β^+ $\beta p?$	β^+ $\beta p?$	β^+ γ 106...; 795; 297...	β^+ γ 252; 750; 421; 891	β^+ 3.5 γ 777; 601; 593...	β^+ 2.3 γ 180; 720; βp 632 740...	β^+ 4.3... γ 656; 834; β^+ 878...
Cd 97 1.5 s	Cd 98 9.2 s	Cd 99 16 s	Cd 100 49.1 s	Cd 101 1.2 m	Cd 102 5.1 m	Cd 103 7.3 m
β^+ βp 1.5-5.0	β^+ γ 347; 1176; 107; 61...	β^+ γ 343; 672; 1583...	β^+ γ 937; 140; 583...	β^+ γ 98; 1723; 1259; 925...	ϵ ; β^+ γ 481; 1037; 505; 415...	ϵ ; β^+ 3.2... γ 1462; 1449; 1080; 387... g; m
Ag 96 1.40 s	Ag 97 25.3 s	Ag 98 4.7 s	Ag 99 10.5 s 2.1 m	Ag 100 2.3 m 2.0 m	Ag 101 3.1 s 11.1 m	Ag 102 8 m 13 m
β^+ βp 1.3-3.7; g	β^+ γ 686; 1295; 1256...	β^+ γ 863; 679; 571...	β^+ 4.2... γ 264; 832; βp 343; 164	β^+ 3.4; γ 666; 751; 751; 1694... 773...	β^+ 2.7; γ 251; 588; 667; 1174...	β^+ 2.3... γ 557; 719; 836; 1745...
Pd 95 14 s	Pd 96 2.0 m	Pd 97 3.1 m	Pd 98 17.7 m	Pd 99 21.4 m	Pd 100 3.7 d	Pd 101 8.47 h
β^+ γ 1951; 717; 382...	ϵ ; β^+ γ 125; 762; 500; 1099...	β^+ 3.1 γ 265; 475; 793...	ϵ ; β^+ γ 112...; 107...	β^+ 2.3 γ 136...; 673...	ϵ no β^+ γ 84; 75; 126...	β^+ 3.3... γ 296; 590; 270...; m
Rh 94 70.6 s 25.8 s	Rh 95 1.96 m 5.0 m	Rh 96 1.5 m 9.9 m	Rh 97 44 m 31 m	Rh 98 3.5 m 8.7 m	Rh 99 4.7 h 16 d	Rh 100 4.7 m 20.8 h
β^+ γ 1431; 756; 312; 1073...; βp 146	β^+ 3.2... β^+ γ 543 784... 1352...	β^+ 3.3... β^+ 4.7... γ 833; 1099; 1692...	β^+ 3.3... β^+ 3.3... γ 833; 685; 632...	β^+ γ 652; 745...	ϵ 0.7 γ 341; 618; 1261...	ϵ 0.7; 1.1... β^+ 0.7; β^+ 0.7; γ 526; 353; 90... 687...)

Overview:

High-precision mass measurements of N=Z around ^{100}Sn

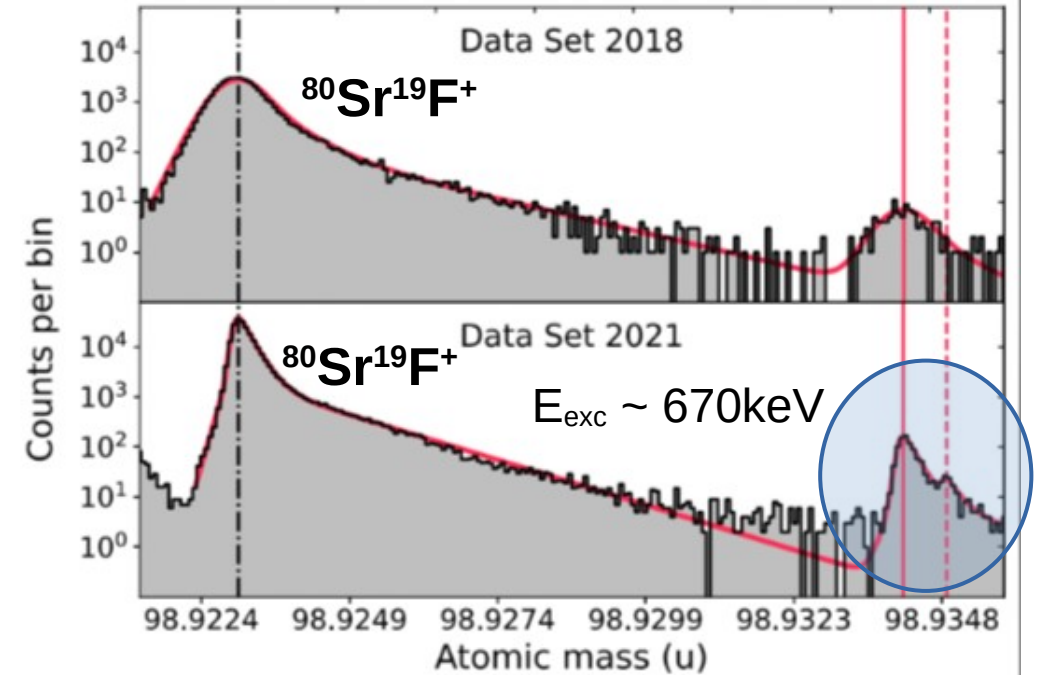
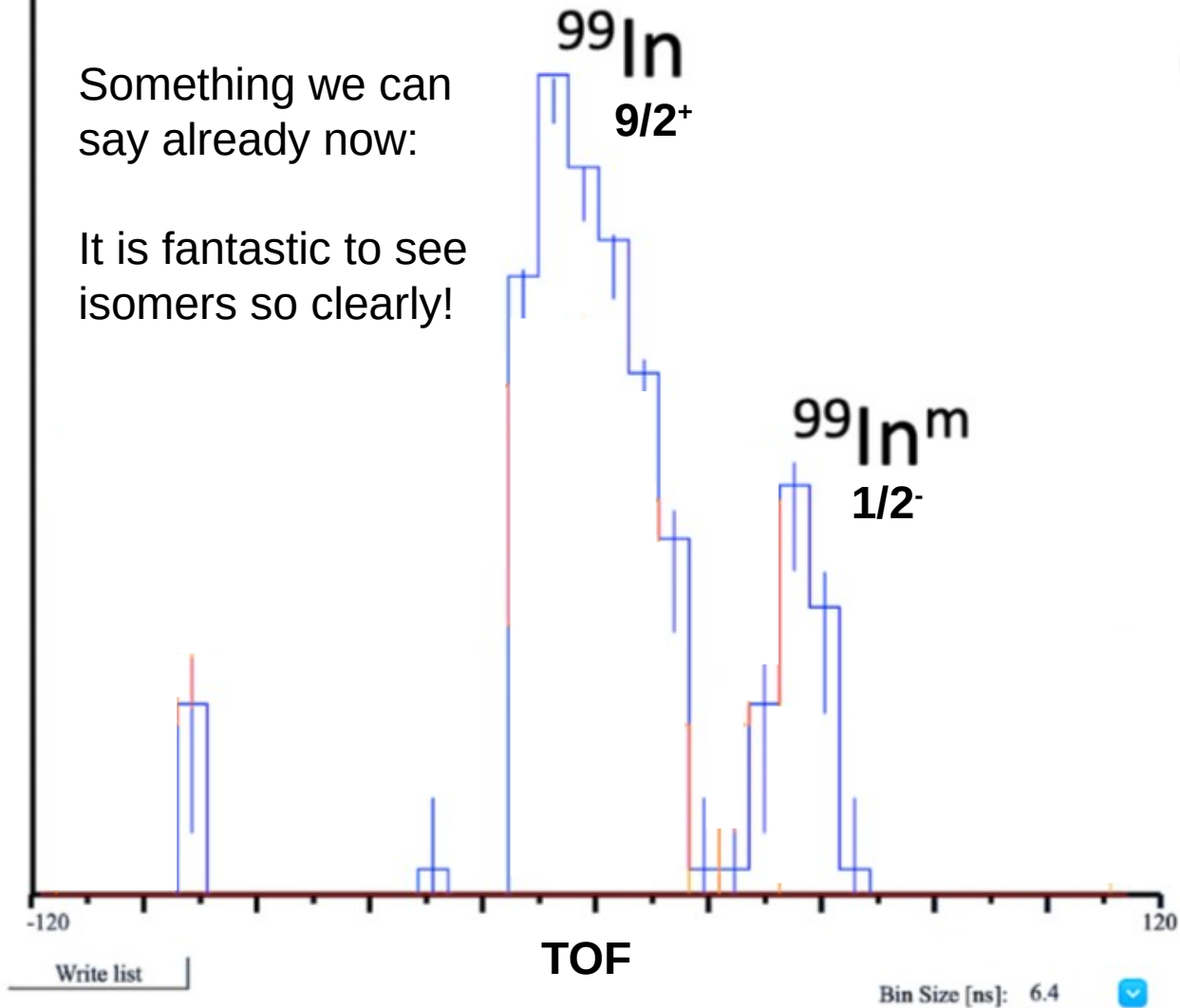
-  Accomplished (new or improved)
-  Still to do
-  Known by other exp.
-  Well known



Great recent results!

Something we can say already now:

It is fantastic to see isomers so clearly!

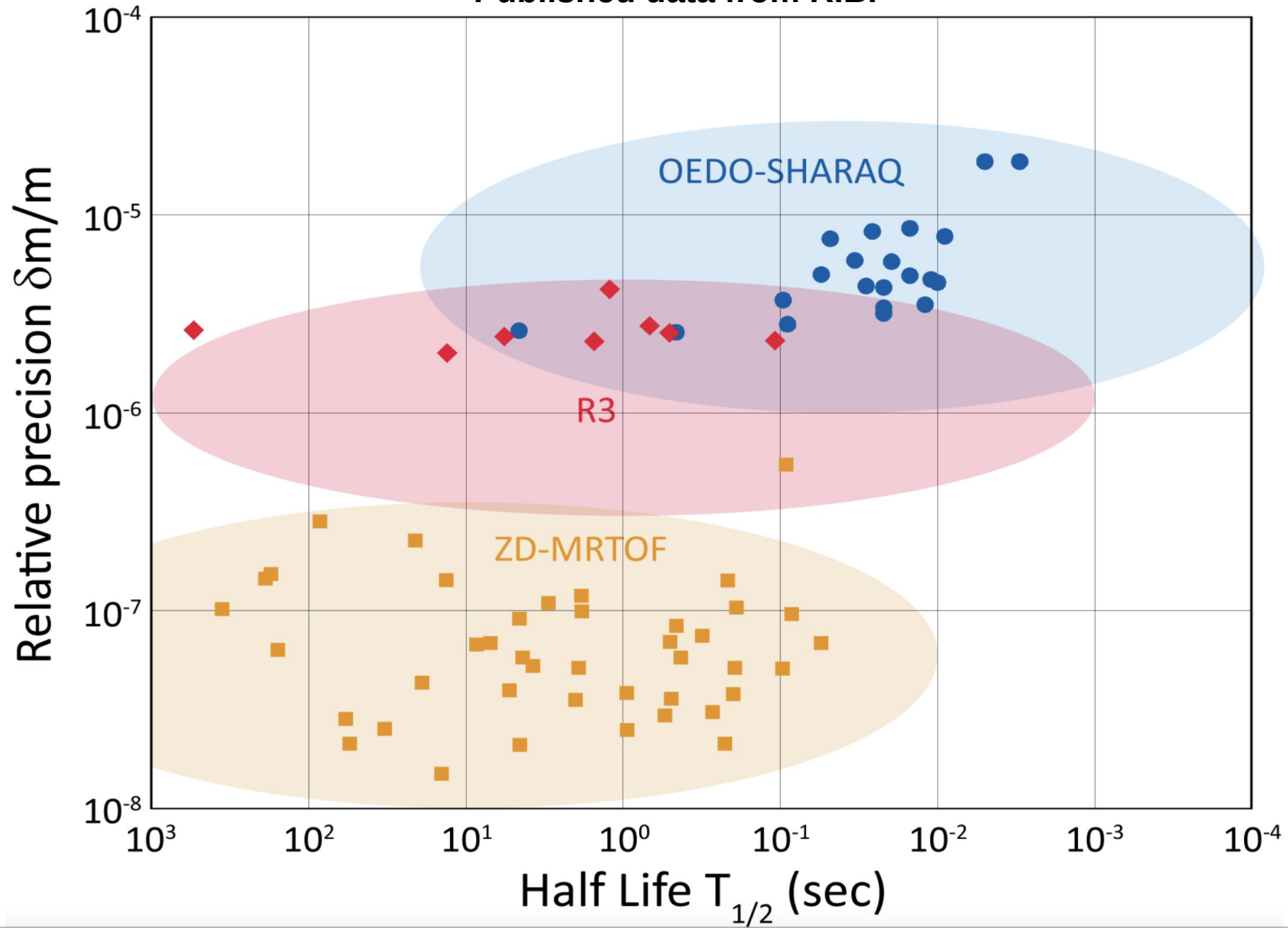


Spectrum from ISOLTRAP MRTOF

L. Nies et al., Phys. Rev. Lett.
131, 022502 (2023)

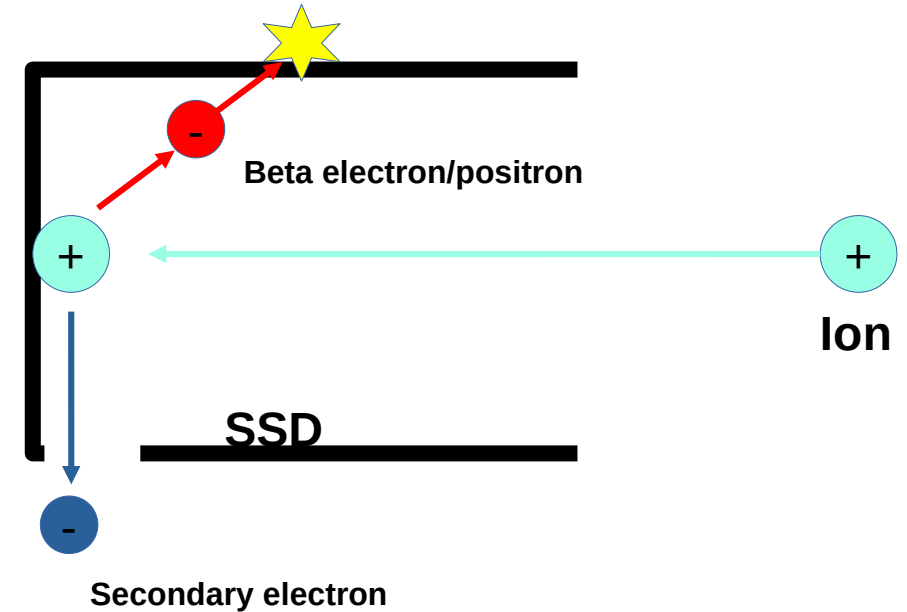
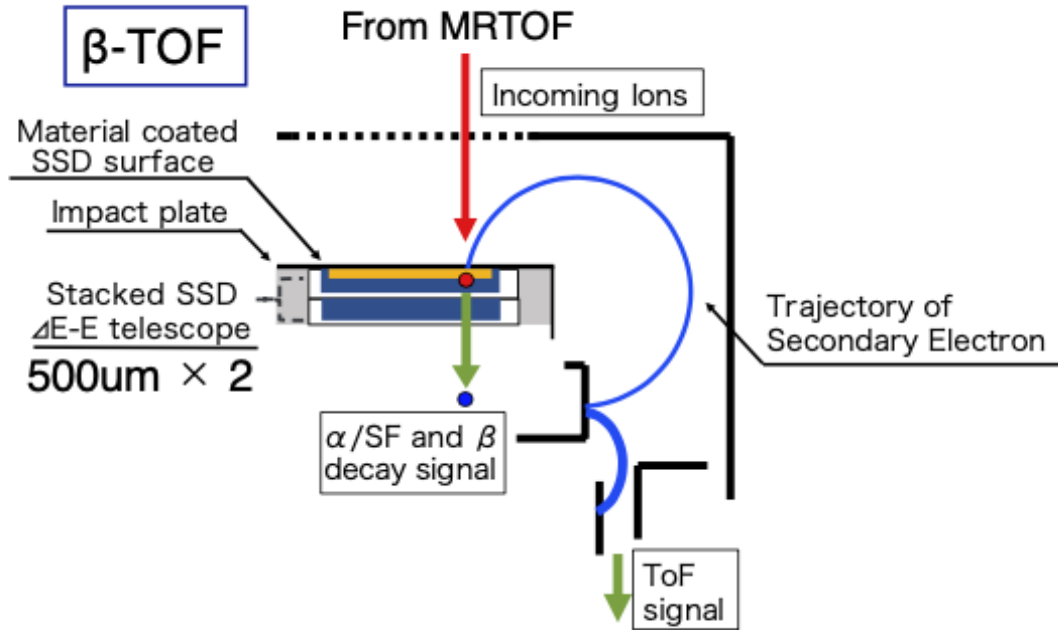


Published data from RIBF



How about future developments?

We need to increase the efficiency and quality of beta-TOF detection



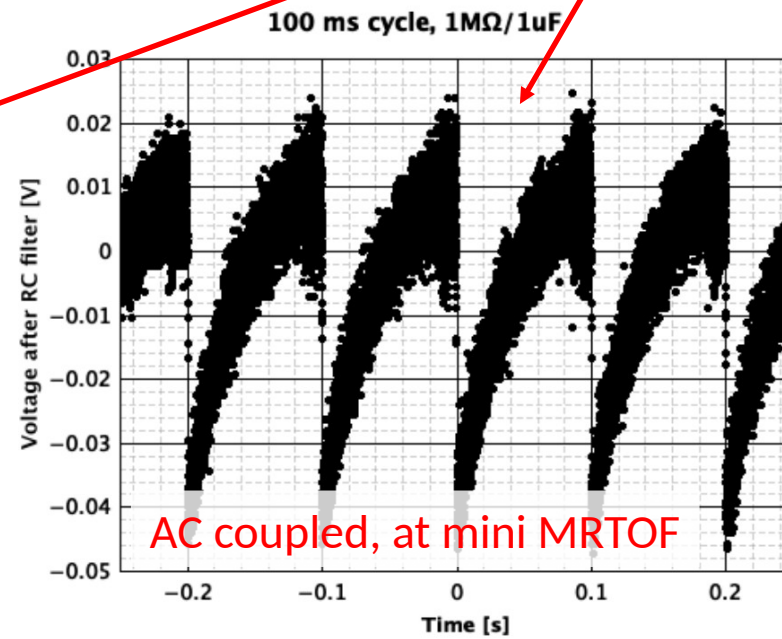
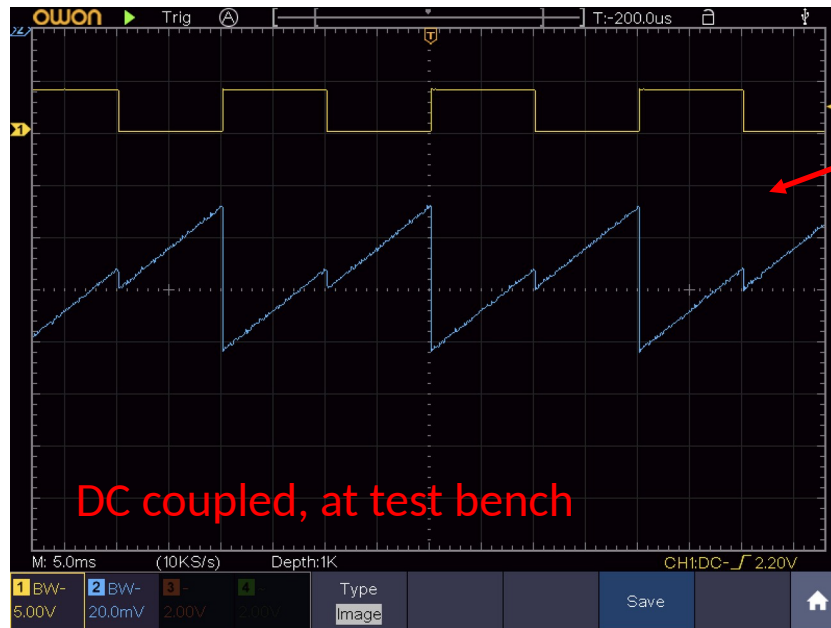
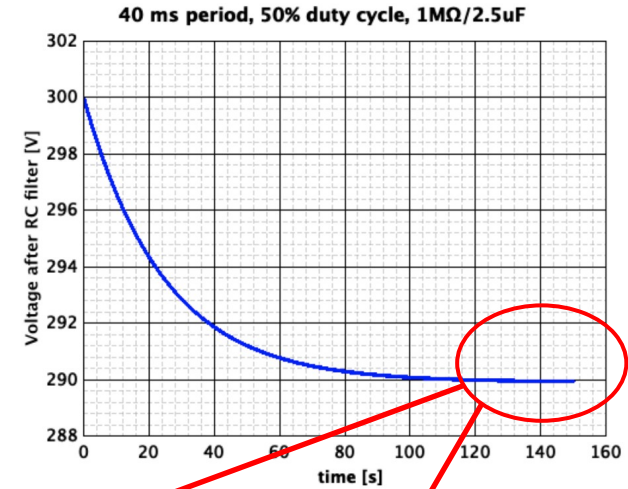
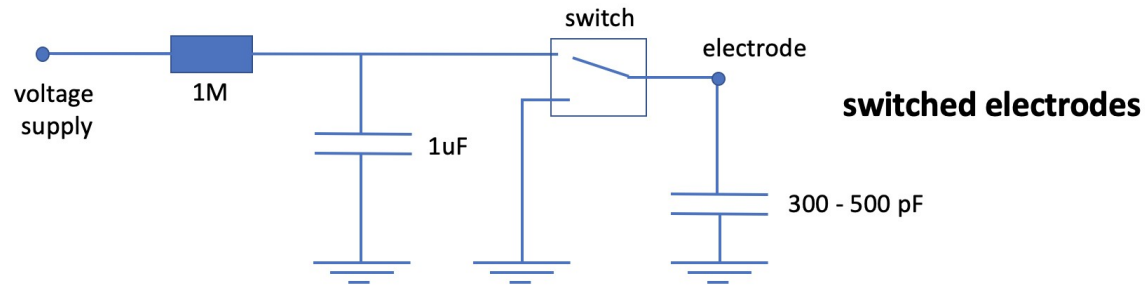
Correlation with TOF and β -signal
(The β -signal is the one passing through the double layered Si.)

Finding ways for more fruitful decay spectroscopy tagged on masses

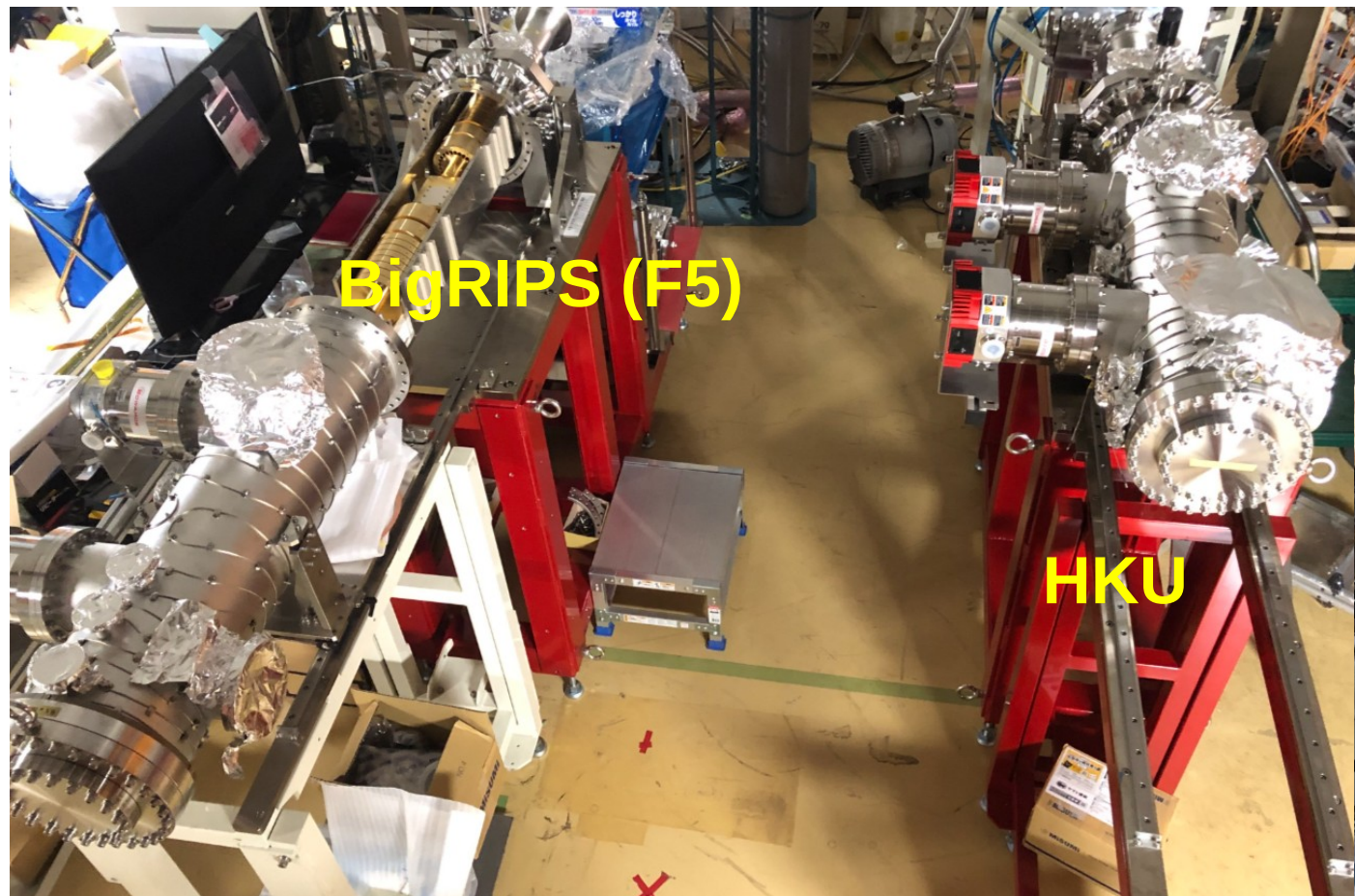
How about future developments?

Making the electrostatic system more electrostatic

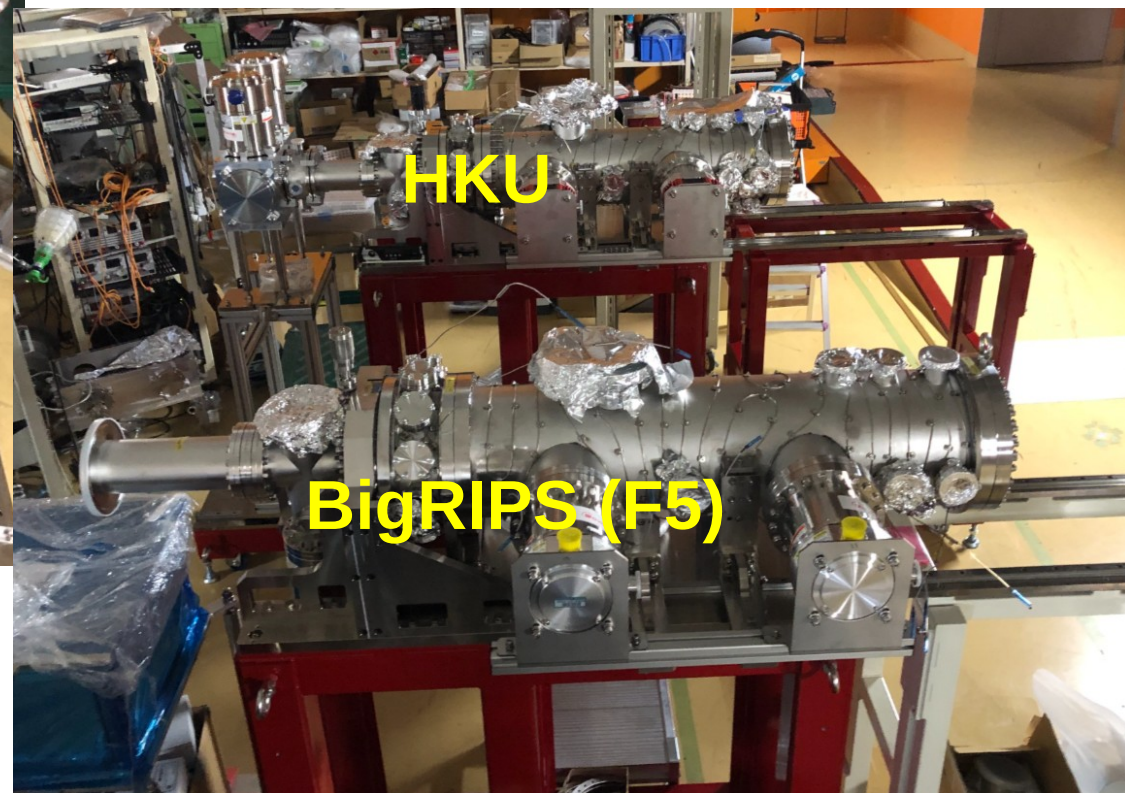
Micro-ramping at the MRTOF mirrors



How about future developments?



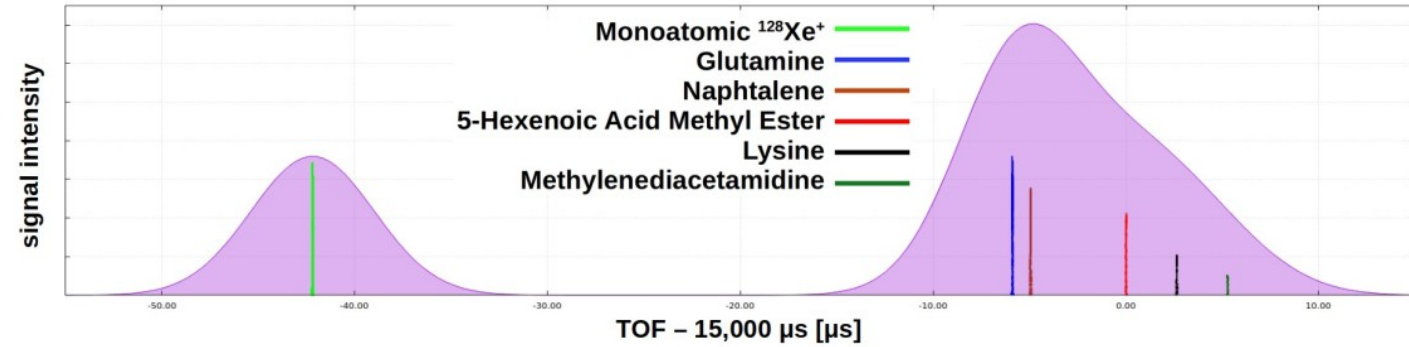
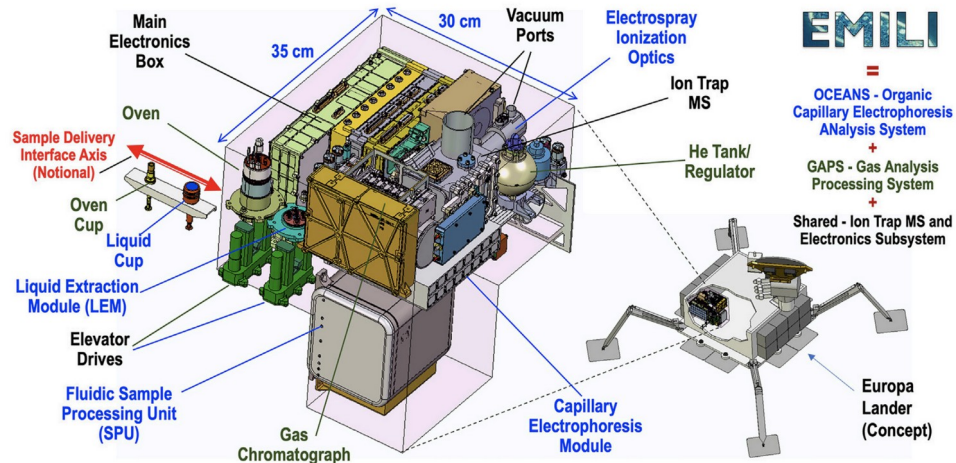
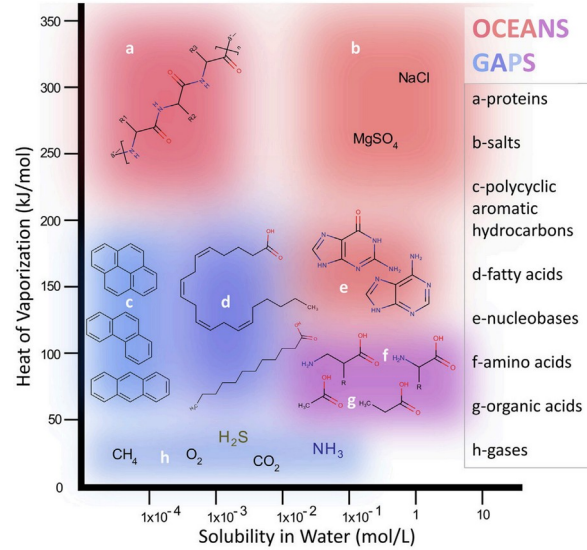
Building new MRTOF spectrometers!



How about future developments?

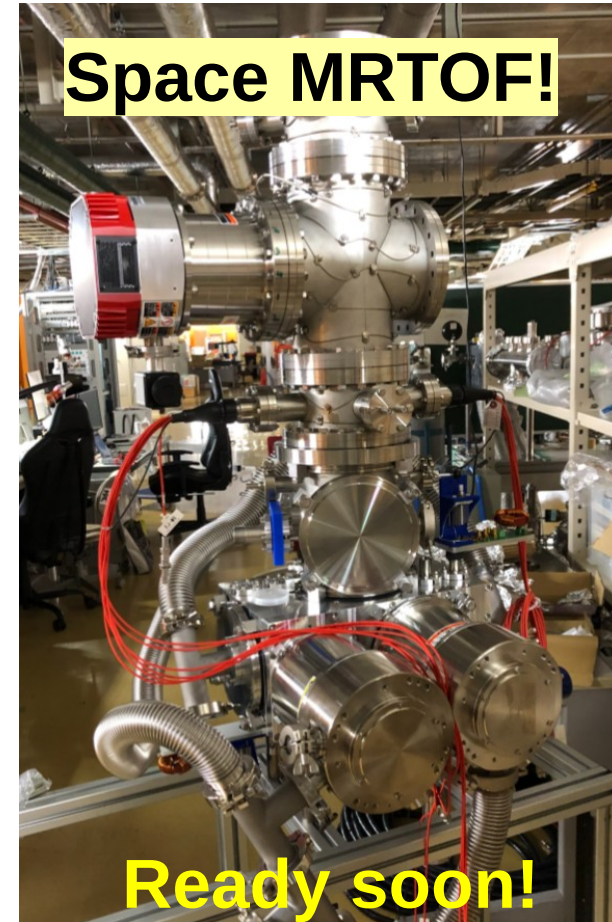
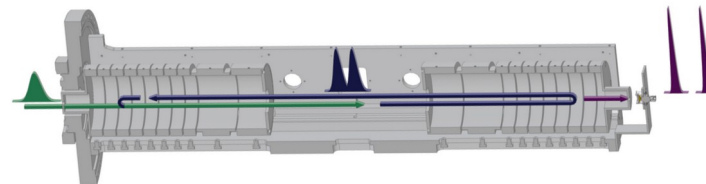
Outreach: Mass spectrometry in space with NASA

William B Brinckerhoff



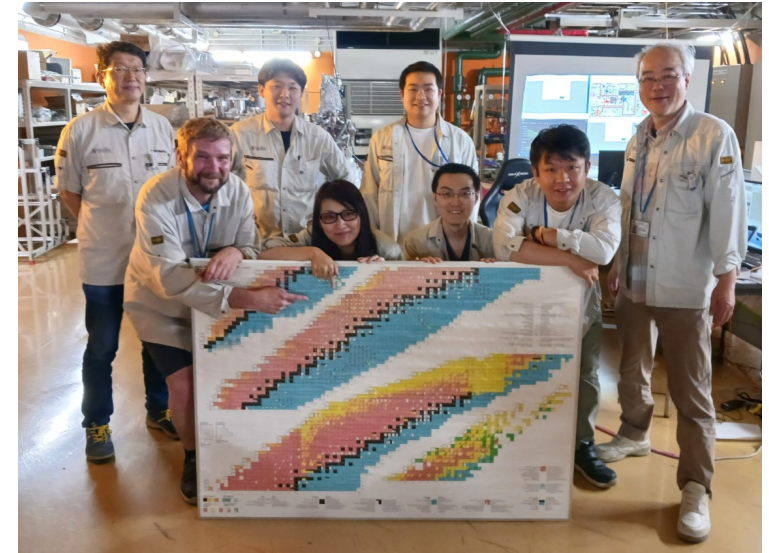
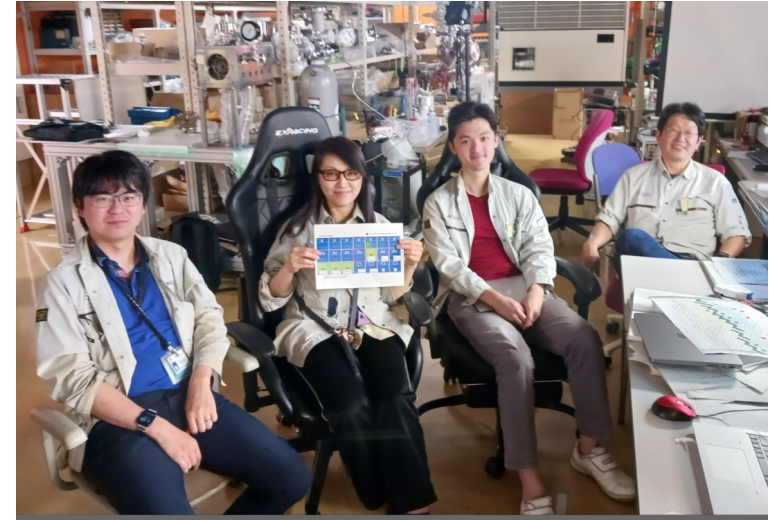
Search for signs of former life:

- Certain molecules can only be formed by living beings
- Identification by mass spectrometers
- MRTOF-MS technology is robust and able to survive in space environments



Summary

- The ZD MRTOF-MS became one of the most successful projects for mass measurements of the recent years!
→ 68 isotopes published!
- Background problems in case of low production rates are tackled by efficient use of an IMD system, and beta-correlated mass spectroscopy
- From test setup to ready-to-use device in about four years of time line
- Development of combined runs with in-beam gamma campaigns
→ powerful proposals
- Development of through-beam experiments for simultaneous decay spectroscopy





香港大學

THE UNIVERSITY OF HONG KONG



2020 first ZD MRTOF commissioning



Thanks to:

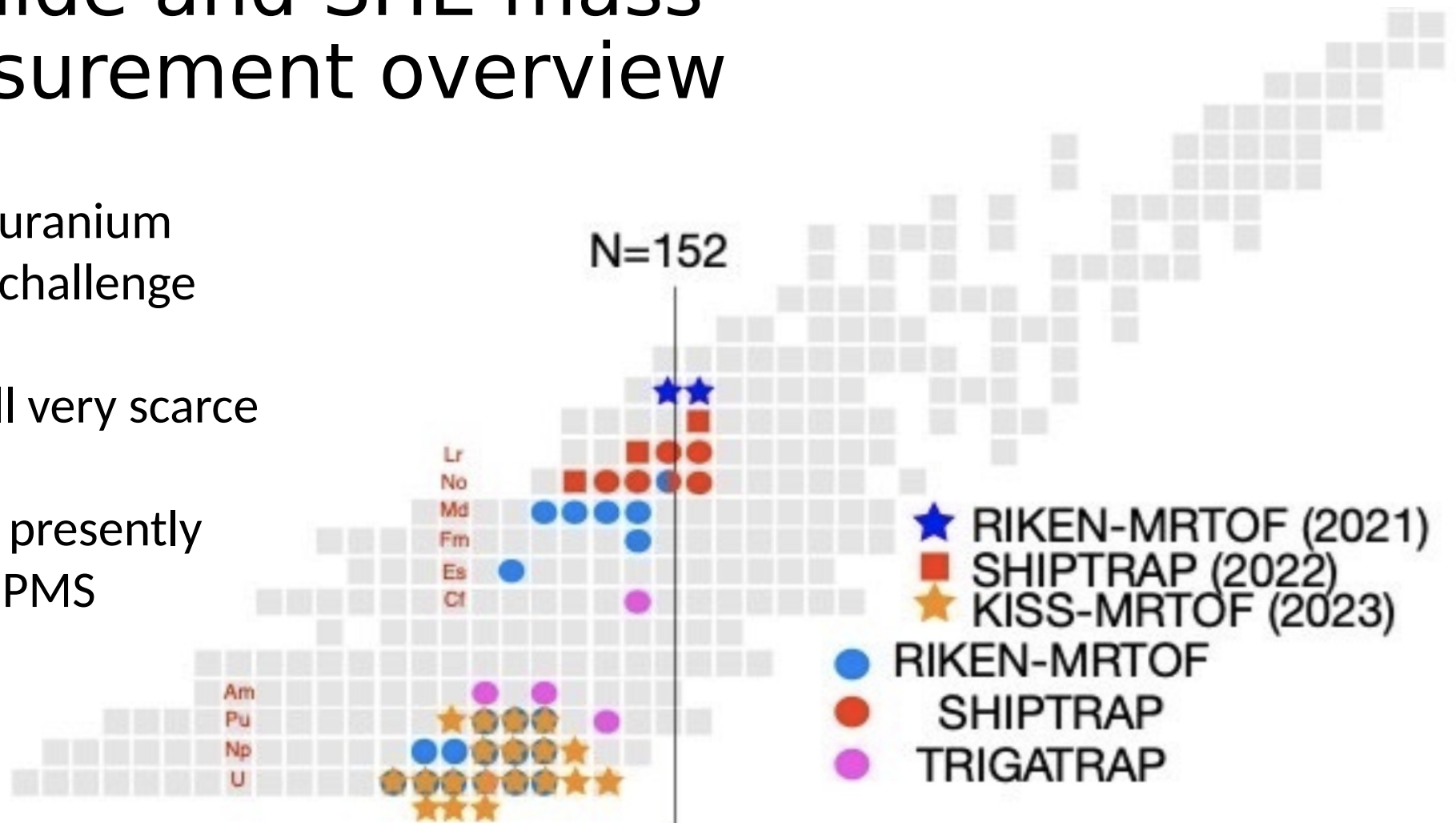
- KISS collaboration
- SHE mass collaboration
- ZD MRTOF collaboration



The SHE Mass facility and status of $^{257,258}\text{Db}$ mass measurements

Actinide and SHE mass measurement overview

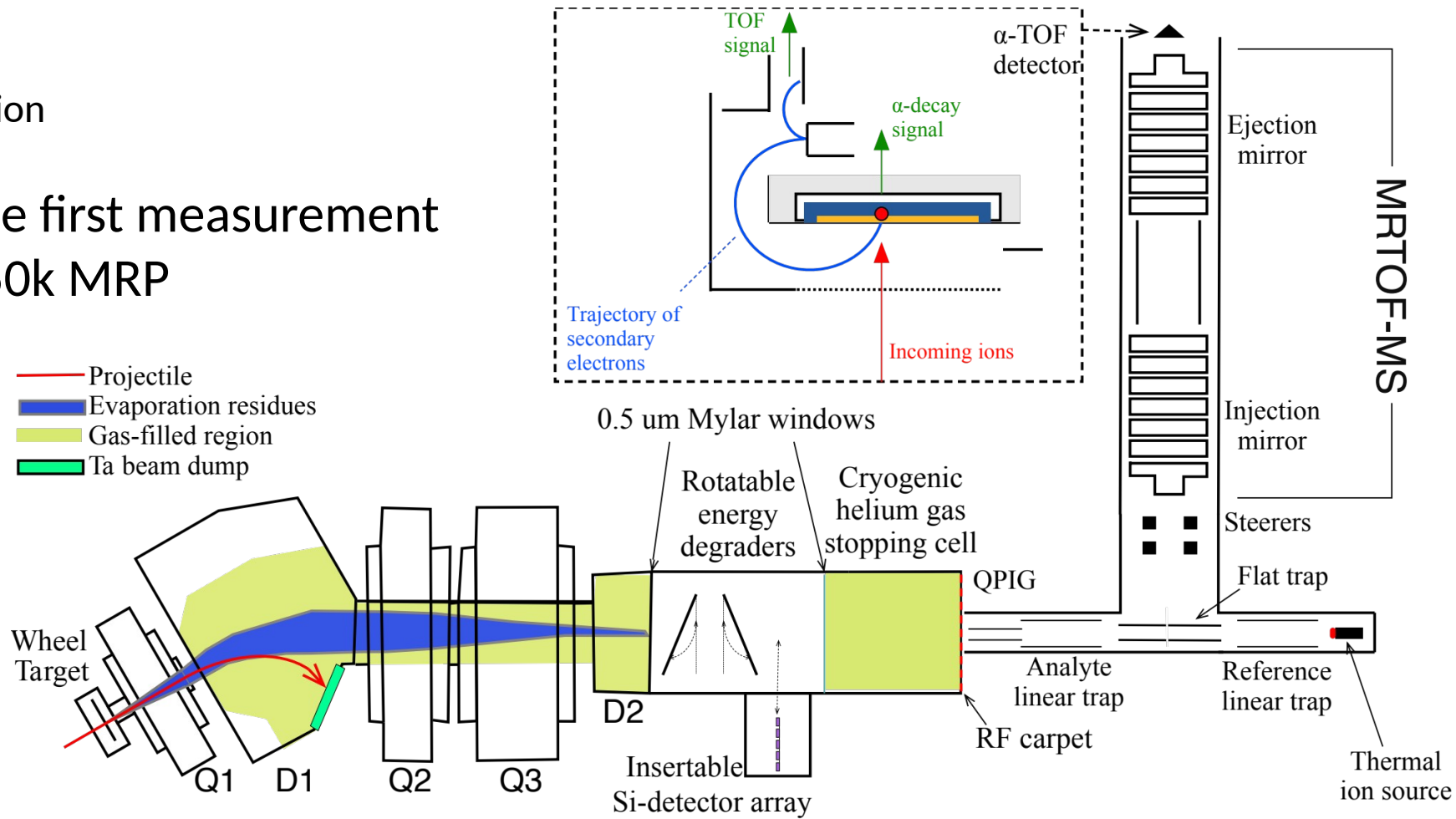
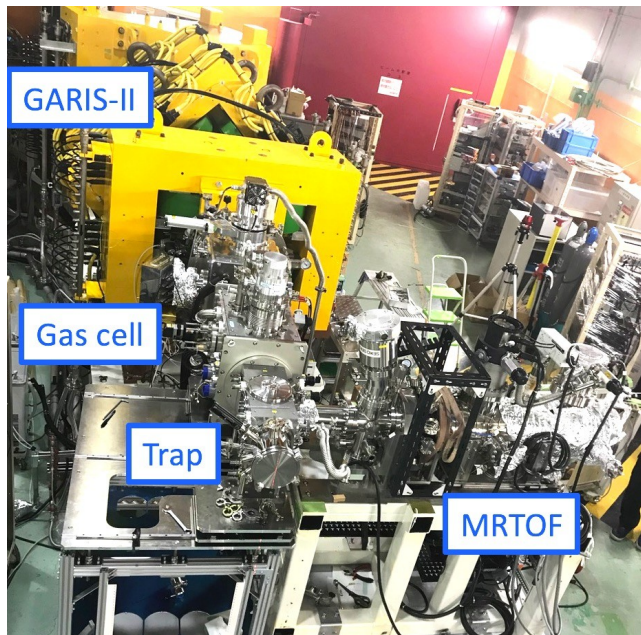
- Production of transuranium isotopes is a major challenge
- Available data is still very scarce
- Only a few projects presently join the efforts of HPMS



The SHE Mass facility and status of $^{257,258}\text{Db}$ mass measurements

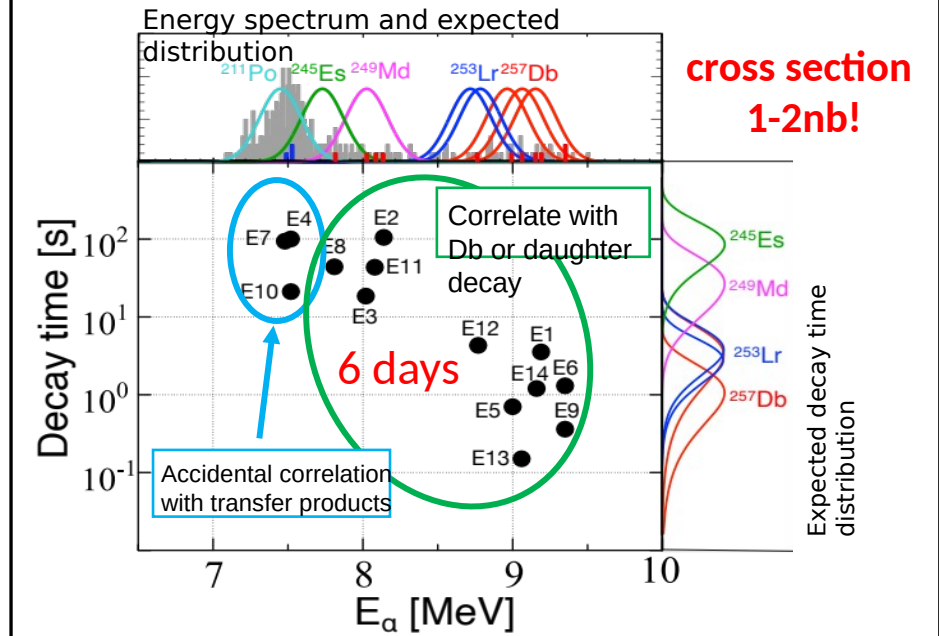
SHE-Mass facility overview

- System efficiency $\sim 5\%$
 - 25%~30% stopping+extraction
 - $\sim 20\%$ transport
- MRTOF $\sim 300\text{k}$ MRP in the first measurement
- Recently increased to 750k MRP



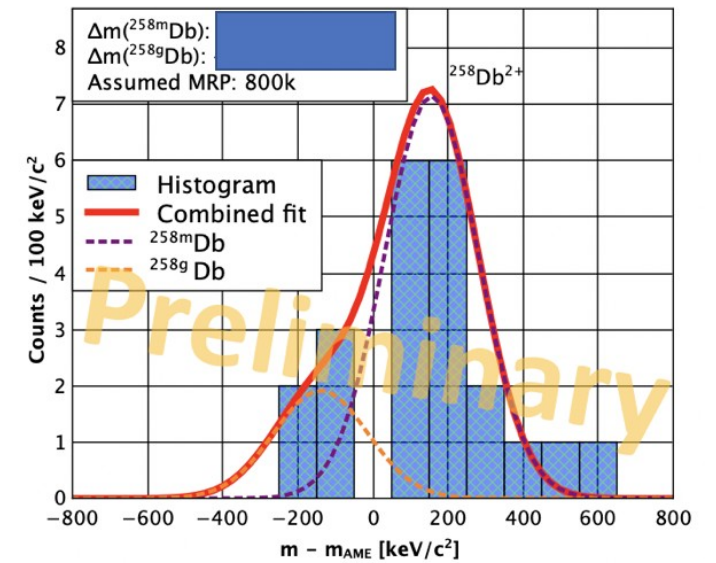
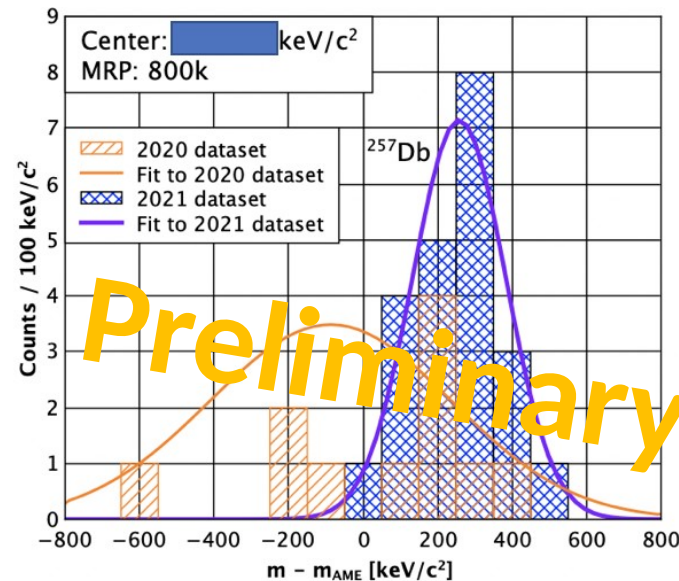
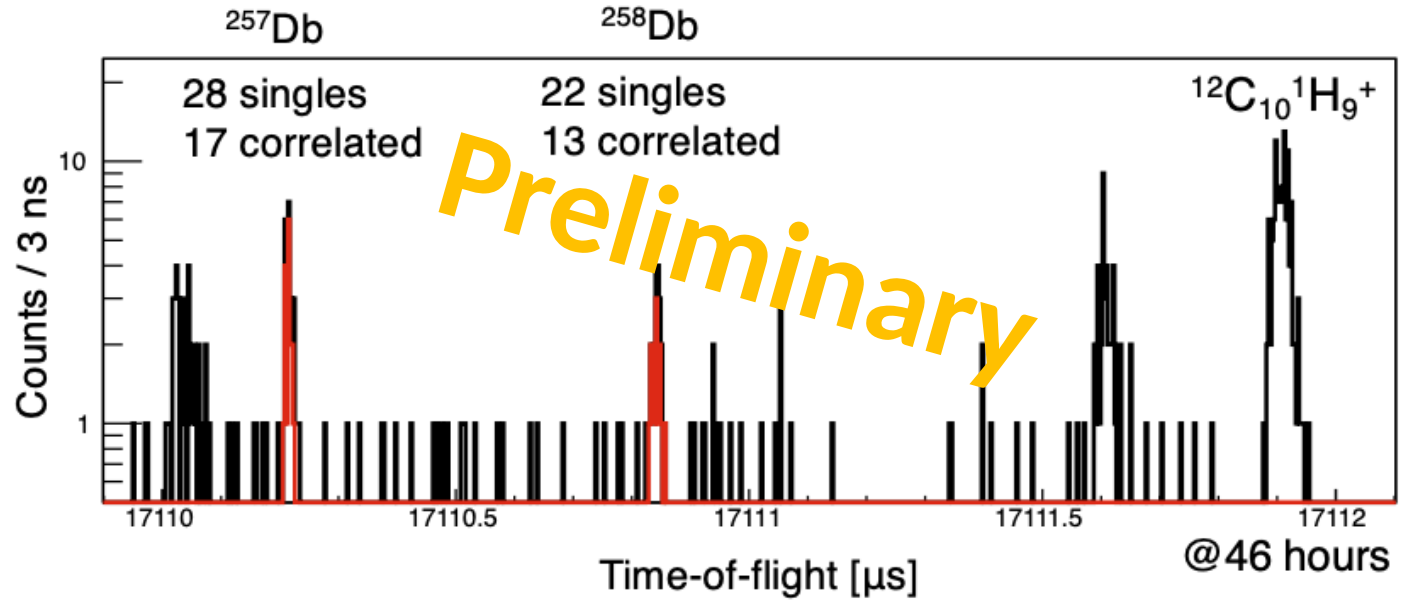
The SHE Mass facility and status of $^{257,258}\text{Db}$ mass measurements

P. Schury et al, PRC104, L021304 (2021)



First effort: metallic Pb targets
 Primary beam of 500 pA ^{51}V on nat. Pb, 6MeV/u
 11 ToF events correlated with α -decays
 Mass resolving power limited to $m/\Delta m \approx 300k$

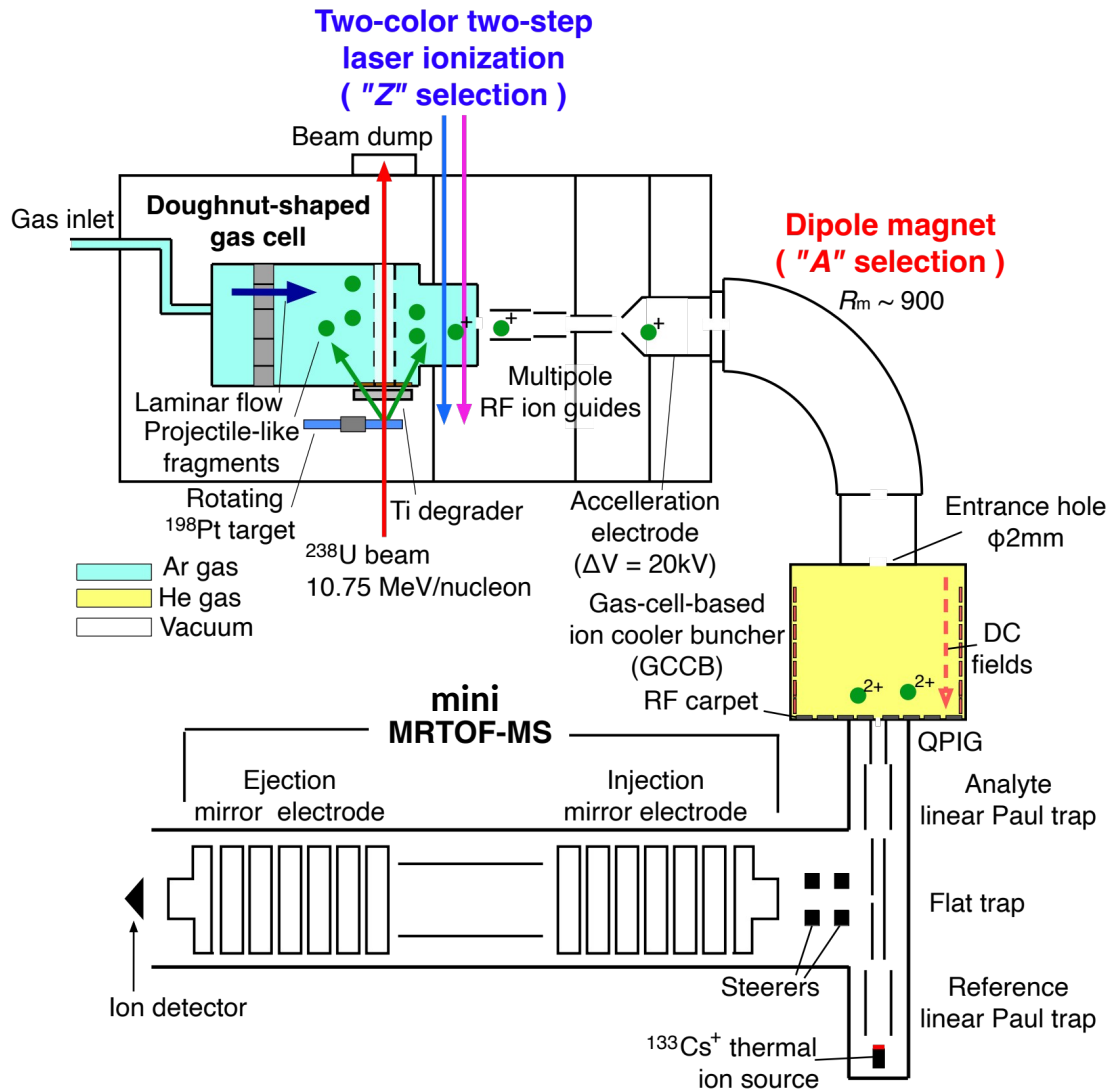
Second effort with **PbS** targets
 Primary beam intensity up to 2 pA, 6MeV/u possible
 Many more correlated events
 Mass resolving power reached $m/\Delta m \approx 750k$
 Could exclude $E_{ex} (^{257m}\text{Db}) \geq 300$ keV



The KISS facility and the discovery of a new uranium isotope

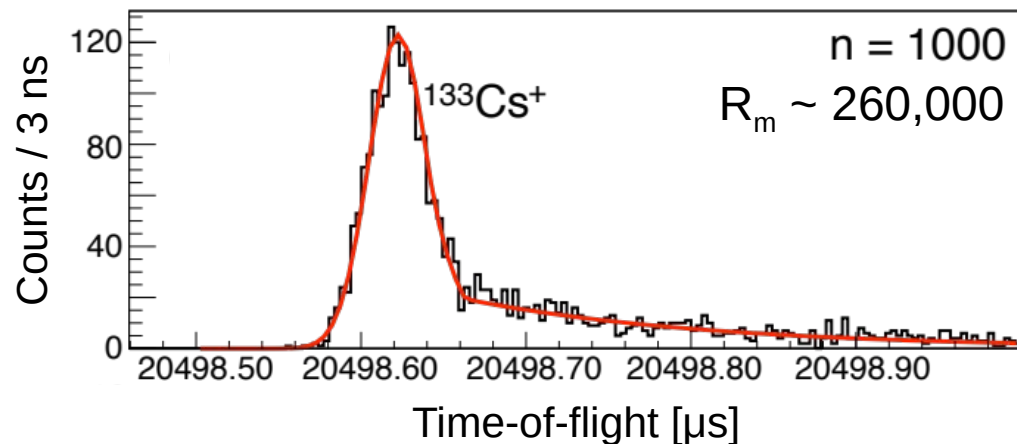
Schematic view of KISS-MRTOF

- Beam : ^{238}U 10.75 MeV/nucleon $\sim 30\text{pnA}$
- Target : ^{198}Pt target (enriched to 91.63%) $\sim 30\text{pnA}$



- ① Thermalization of MNT products in Ar gas
- ② Selection of **atomic number** by **laser-ionization** technique
- ③ Selection of **mass number** by **dipole magnet**
- ④ Spectroscopy with pure-beam ($\beta\gamma$, half-life, mass)

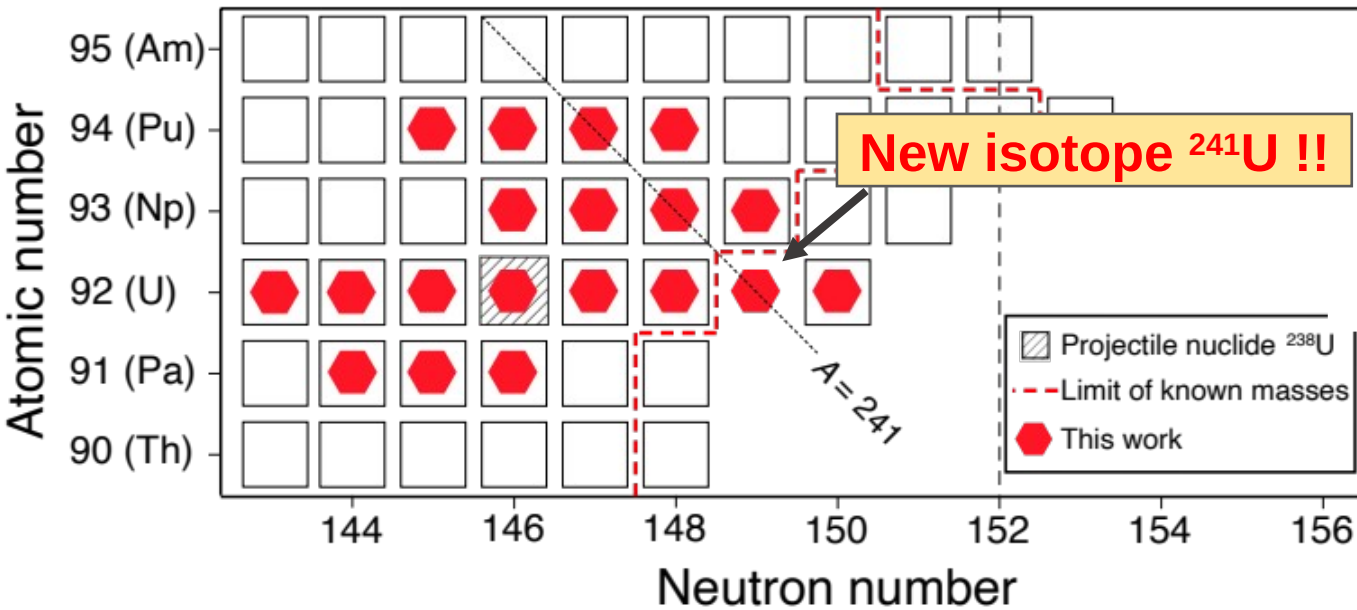
Mass measurement by MRTOF-MS



The KISS facility and the discovery of a new uranium isotope

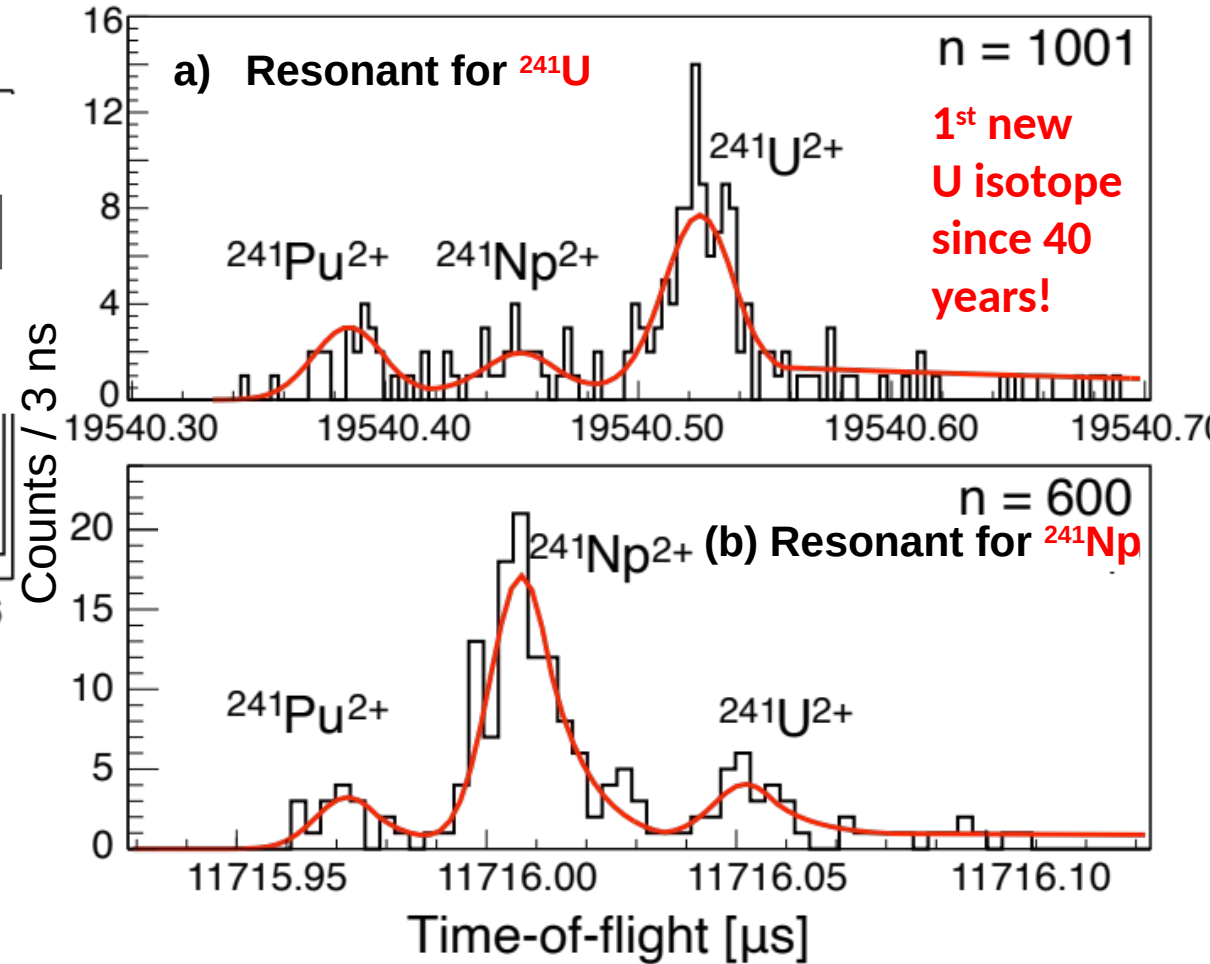
Totally 19 nuclides were measured

$^{235-242}\text{U}$, $^{235-237}\text{Pa}$, $^{239-242}\text{Pu}$, $^{239-242}\text{Np}$



- Beam : ^{238}U 10.75 MeV/nucleon $\sim 30\text{pnA}$
- Target : ^{198}Pt target (enriched to 91.63%)

TOF spectrum of $A = 241$ species



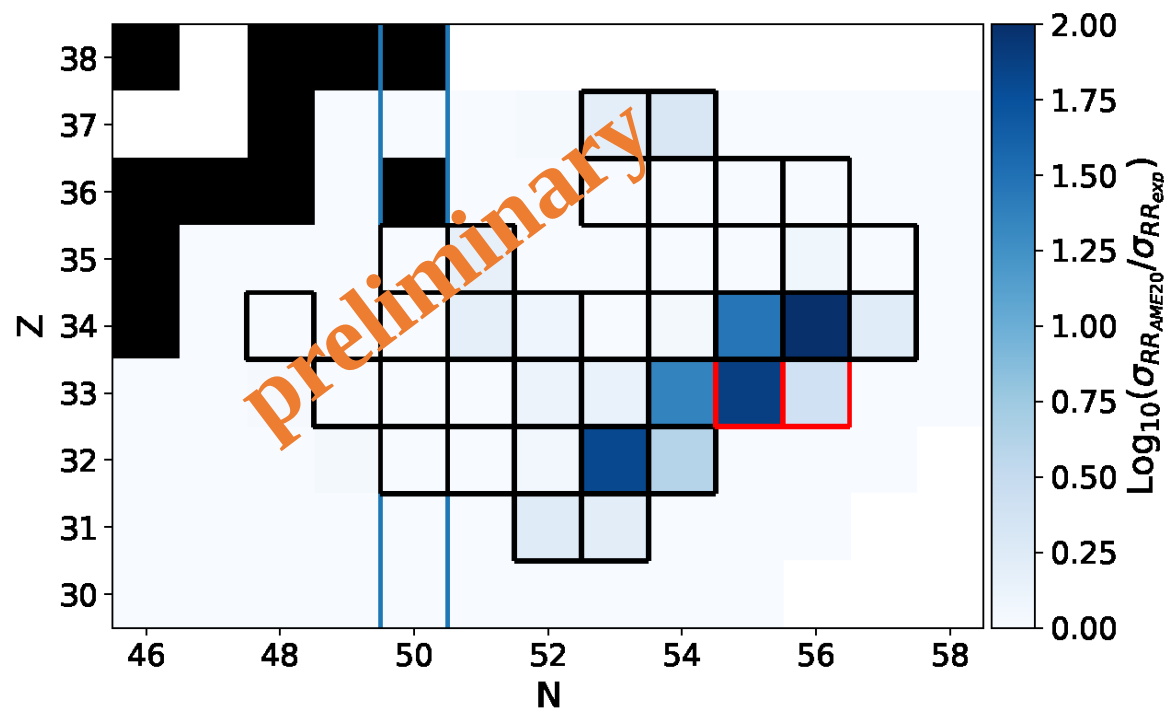
Clearly identified as new isotope ^{241}U

T. Niwase *et al.*, Phys. Rev. Lett. 130, 132502 (2023).

Editors suggestion!

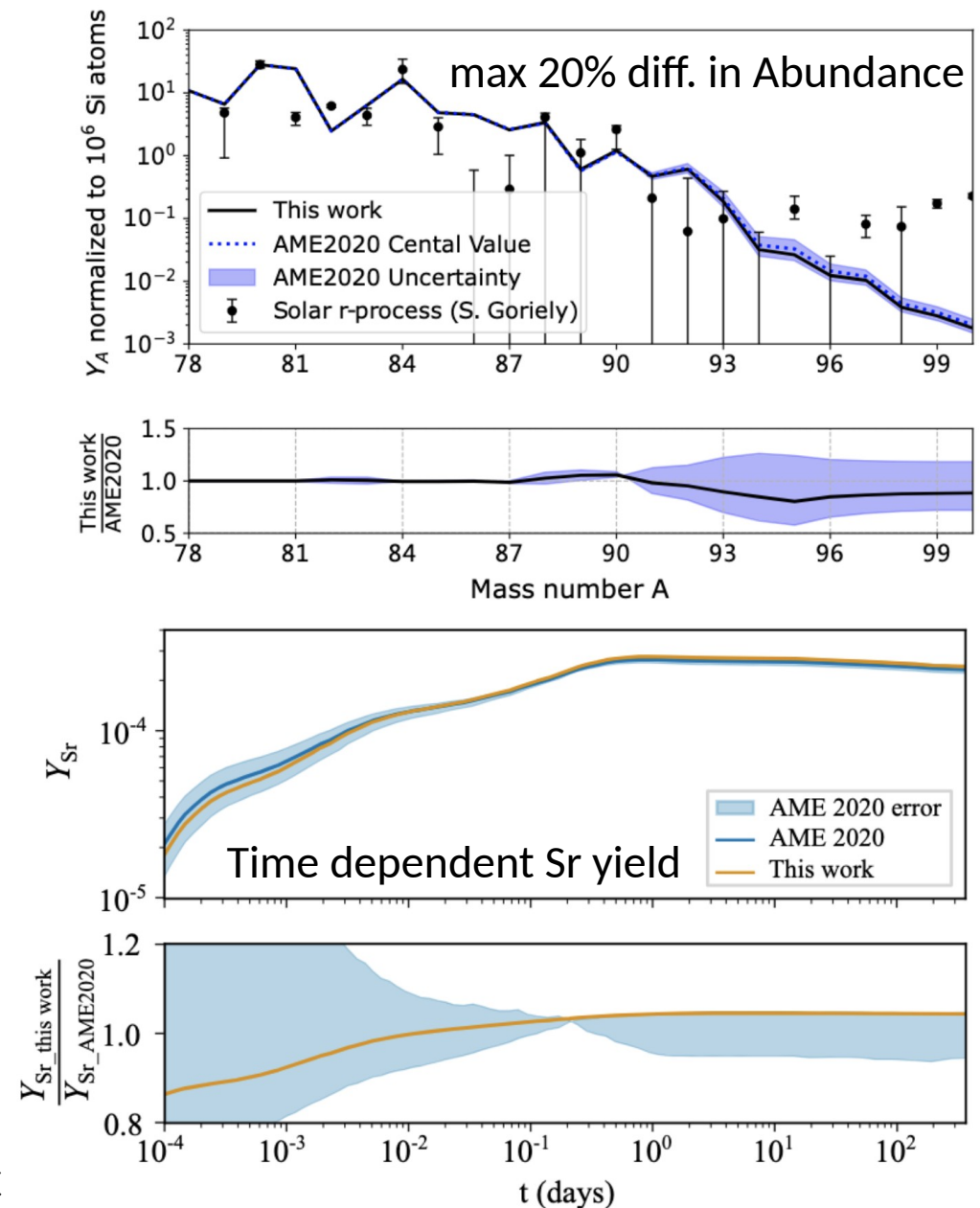
High-precision mass measurements of n-rich Ga-Br isotopes

Astrophysical considerations



- Theoretical work by S. Nikas
- Pinning down reaction rates for neutron capture
- Comparing r-process abundance using new data
- Dynamic evolution of Sr yield motivated by GW170817

W. Xian, S. Chen *et al.*, under review at PRC



High-precision mass measurements of n-rich Mo isotopes

Parasitic experiment of HiCARI (in-beam gamma) Campaign: W. Korten

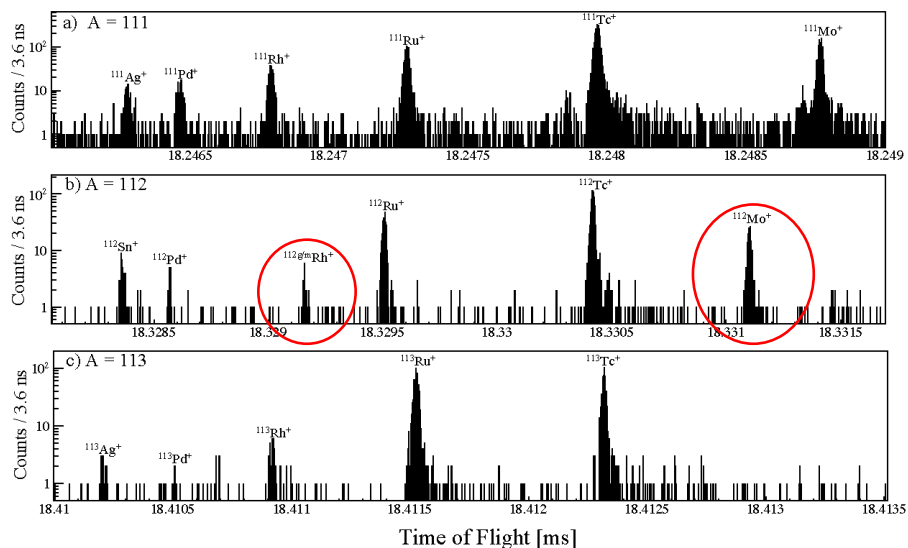
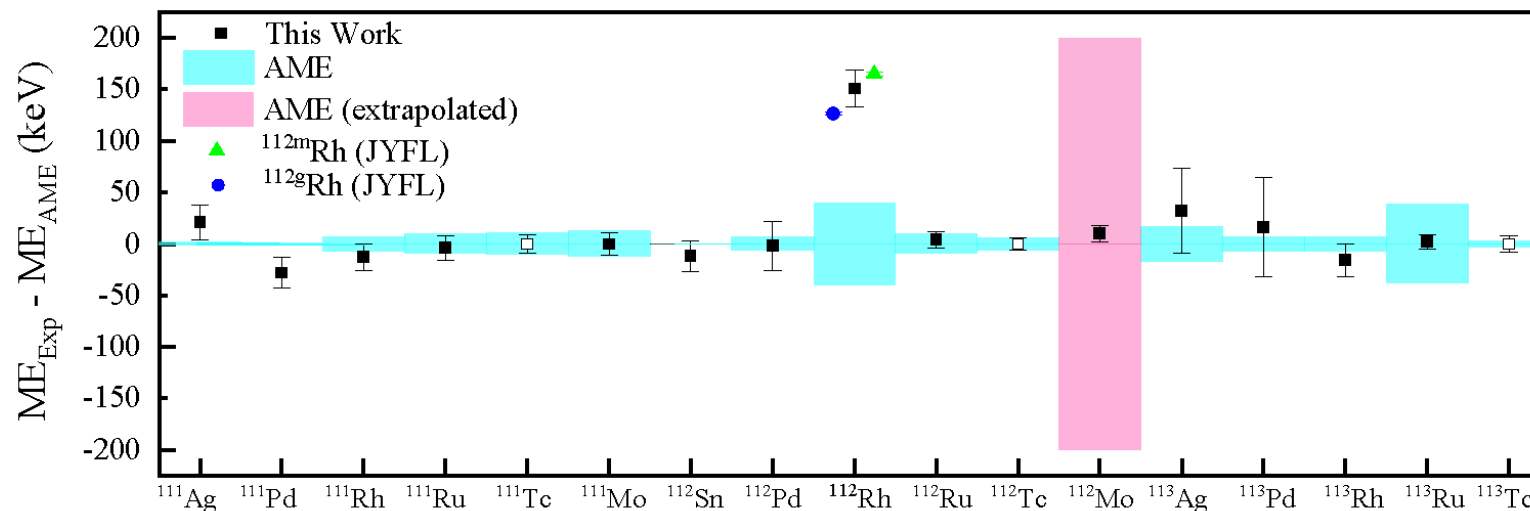


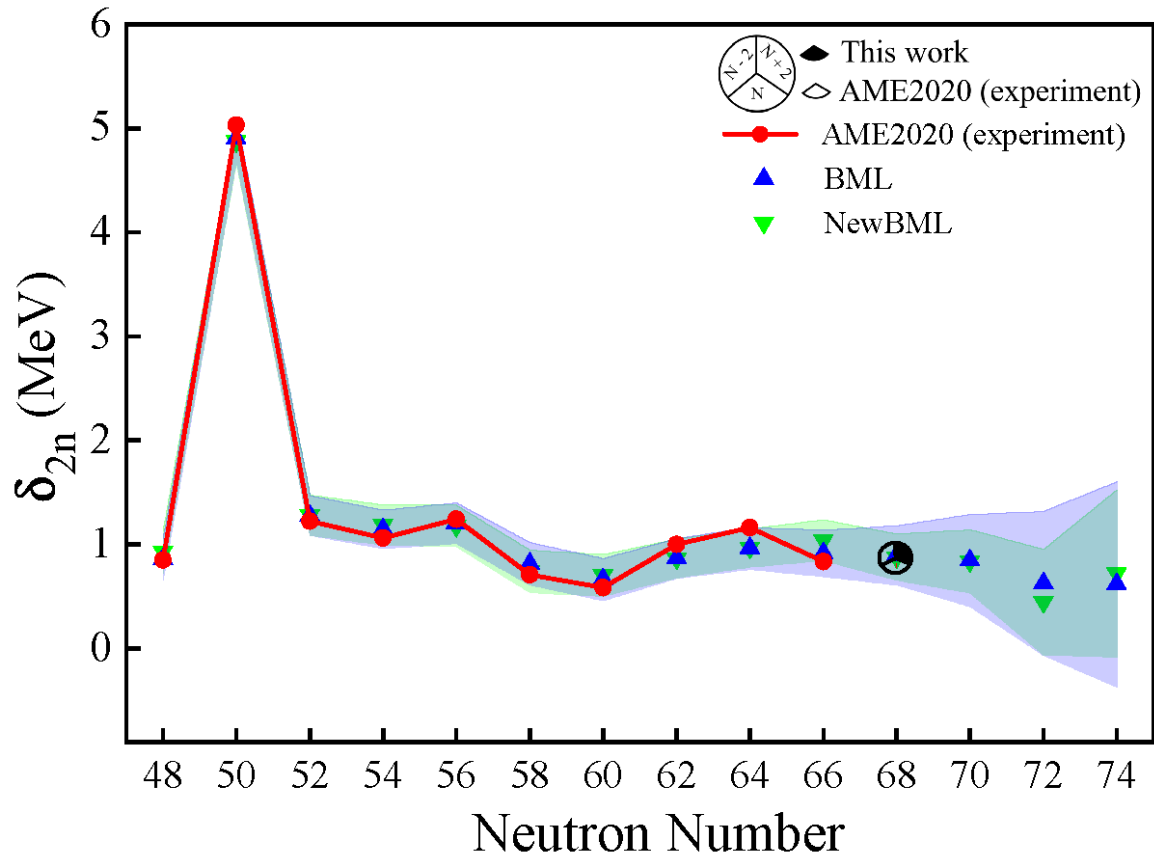
TABLE I. Measured radioactive isotopes, the reference ion, measured mass excess ME_{MRTOF} , mass excess from literature AME2020 ME_{AME2020} , mass deviation calculated as $\Delta m = ME_{\text{MRTOF}} - ME_{\text{AME2020}}$. Extrapolated values of AME2020 are denoted by #, and the total number of the detected ions N_{ion} in this work.

Species	Ref. ion	ME_{MRTOF} (keV/c ²)	ME_{AME2020} (keV/c ²)	Δm (keV/c ²)	N_{ion} (Counts)
¹¹¹ Ag	¹¹¹ Tc	-88195.22(15.40)	-88215.48(1.46)	20.23(15.47)	115
¹¹¹ Pd	¹¹¹ Tc	-86010.80(14.17)	-85985.89(0.73)	-24.91(14.19)	125
¹¹¹ Rh	¹¹¹ Tc	-82318.53(11.28)	-82303.87(6.85)	-14.66(13.19)	262
¹¹¹ Ru	¹¹¹ Tc	-76791.50(9.16)	-76785.33(9.67)	-6.17(13.33)	743
¹¹¹ Mo	¹¹¹ Tc	-59939.88(8.47)	-59939.51(12.58)	-0.07(15.16)	1300
¹¹² Sn	¹¹² Tc	-88667.21(14.43)	-88655.5(0.29)	-12.16(14.44)	83
¹¹² Pd	¹¹² Tc	-86323.23(23.68)	-86321.04(6.55)	-2.19(24.57)	34
¹¹² Ru	¹¹² Tc	-75626.83(6.55)	-75630.87(9.60)	4.05(11.62)	493
¹¹² Mo	¹¹² Tc	-57469.80(7.51)	-57480.00(200.00)#	10.21(200.14)	283
¹¹³ Ag	¹¹³ Tc	-86994.56(40.58)	-87026.83(16.64)	32.26(43.84)	16
¹¹³ Pd	¹¹³ Tc	-83574.42(47.75)	-83590.50(6.95)	16.08(48.26)	18
¹¹³ Rh	¹¹³ Tc	-78783.08(15.9)	-78766.94(7.13)	-16.13(17.45)	80
¹¹³ Ru	¹¹³ Tc	-71866.53(6.33)	-71867.82(38.28)	1.29(38.80)	1051

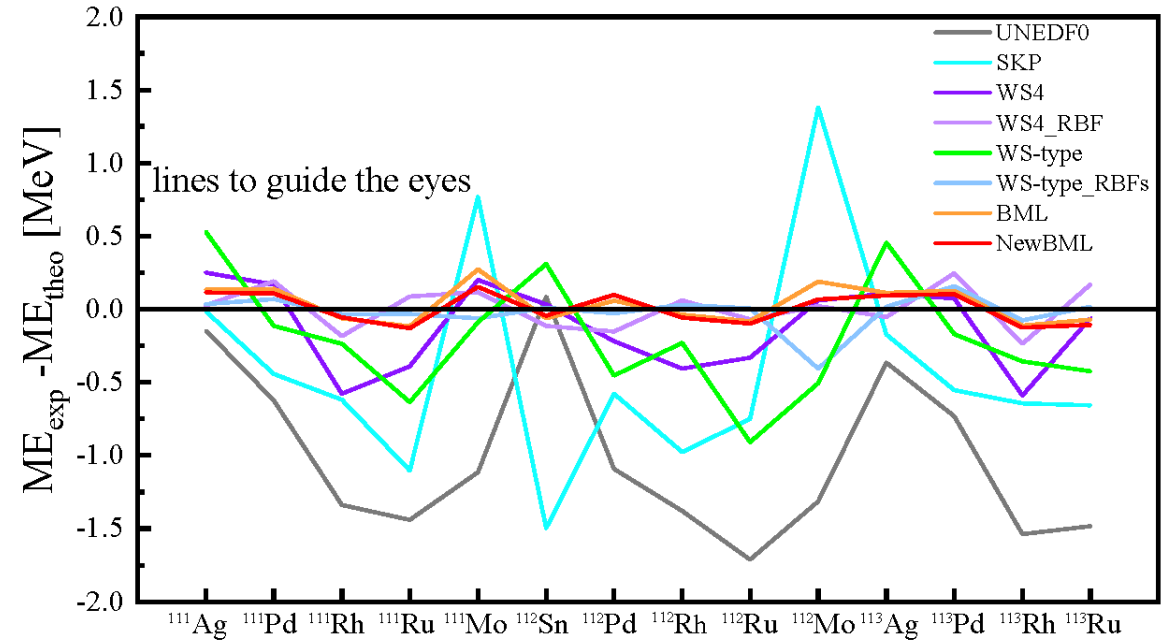
D. Hou *et al.*, now accepted by PRC!!!

moderate efficiency: 0.5%

High-precision mass measurements of n-rich Mo isotopes



Learning how to extrapolate...



Co-work with University of Tokyo and IMP:

- BML mass model using machine learning
- New data included into BML code in this study