



JYVÄSKYLÄN YLIOPISTO
UNIVERSITY OF JYVÄSKYLÄ

Accessing the immediate vicinity of tin-100 with a hot cavity laser ion source

INPC 2025 Daejeon, Korea

Mikael Reponen

Outline

- Motivation
- IGISOL and Hot cavity catcher laser ion source
- Results:
 - Evolution of charge radii and masses across $N = 50$ in silver
 - Energetics of the two-proton decay of ^{94}Ag
 - View over the $N=Z$ line from ^{92}Pd
 - Laser spectroscopy at the $N=Z$ line
- Outlook



Motivation: Region of interest



$Z=50$				Sn 99	Sn 100 1.16 s	Sn 101 1.97 s	Sn 102 3.8 s	Sn 103 7.0 s	Sn 104 20.8 s	Sn 105 34 s	
		In 96	In 97	In 98 37 ms	In 99 3.1 s	In 100 5.83 s	In 101 15.1 s	In 102 23.3 s	In 103 60 s	In 104 1.80 m	
		Cd 94	Cd 95 32 ms	Cd 96 880 ms	Cd 97 1.10 s	Cd 98 9.2 s	Cd 99 16 s	Cd 100 49.1 s	Cd 101 1.36 m	Cd 102 5.5 m	Cd 103 7.3 m
Ag 92	Ag 93	Ag 94 26 ms	Ag 95 1.76 s	Ag 96 4.44 s	Ag 97 25.5 s	Ag 98 47.5 s	Ag 99 2.07 m	Ag 100 2.01 m	Ag 101 11.1 m	Ag 102 12.9 m	
Pd 91 30 ms	Pd 92 1.0 s	Pd 93 1.15 s	Pd 94 9.0 s	Pd 95 7.5 s	Pd 96 122 s	Pd 97 3.10 m	Pd 98 17.7 m	Pd 99 21.4 m	Pd 100 3.63 d	Pd 101 8.47 h	
Rh 90 29 ms	Rh 91 1.5 s	Rh 92 4.7 s	Rh 93 12.2 s	Rh 94 70.6 s	Rh 95 5.02 min	Rh 96 9.9 m	Rh 97 30.7 m	Rh 98 8.72 m	Rh 99 16.1 d	Rh 100 20.5 h	
$N=50$											

$N=50$

$N=50$

Z=48	94Cd	95Cd	96Cd	97Cd	98Cd	99Cd	100Cd	101Cd	102Cd	103Cd	104Cd	105Cd	106Cd	107Cd	108Cd	109Cd	110Cd	111Cd	112Cd	113Cd	114Cd	115Cd	116Cd	117Cd	118Cd	119Cd	120Cd	121Cd	122Cd	123Cd	124Cd	125Cd	126Cd	127Cd	128Cd	129Cd	130Cd		
47	92Ag	93Ag	94Ag	95Ag	96Ag	97Ag	98Ag	99Ag	100Ag	101Ag	102Ag	103Ag	104Ag	105Ag	106Ag	107Ag	108Ag	109Ag	110Ag	111Ag	112Ag	113Ag	114Ag	115Ag	116Ag	117Ag	118Ag	119Ag	120Ag	121Ag	122Ag	123Ag	124Ag	125Ag	126Ag	127Ag	128Ag	129Ag	
	90Pd	91Pd	92Pd	93Pd	94Pd	95Pd	96Pd	97Pd	98Pd	99Pd	100Pd	101Pd	102Pd	103Pd	104Pd	105Pd	106Pd	107Pd	108Pd	109Pd	110Pd	111Pd	112Pd	113Pd	114Pd	115Pd	116Pd	117Pd	118Pd	119Pd	120Pd	121Pd	122Pd	123Pd	124Pd	125Pd	126Pd	127Pd	128Pd

Heavy-ion fusion-evaporation reactions

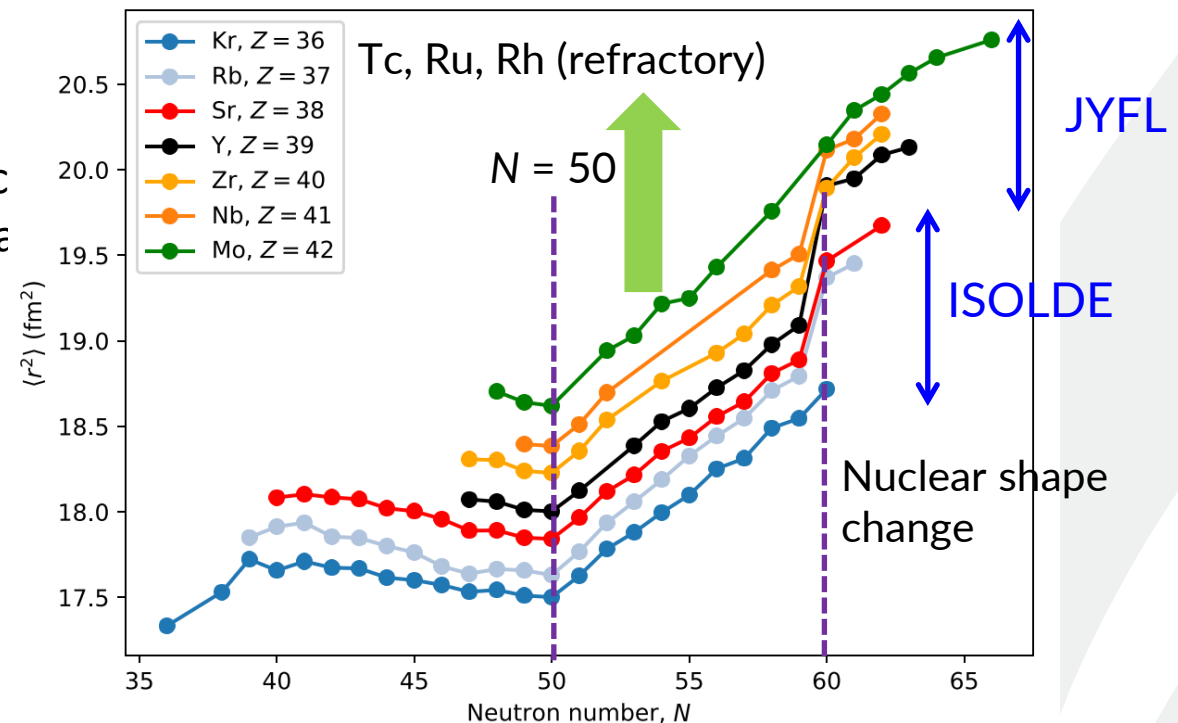
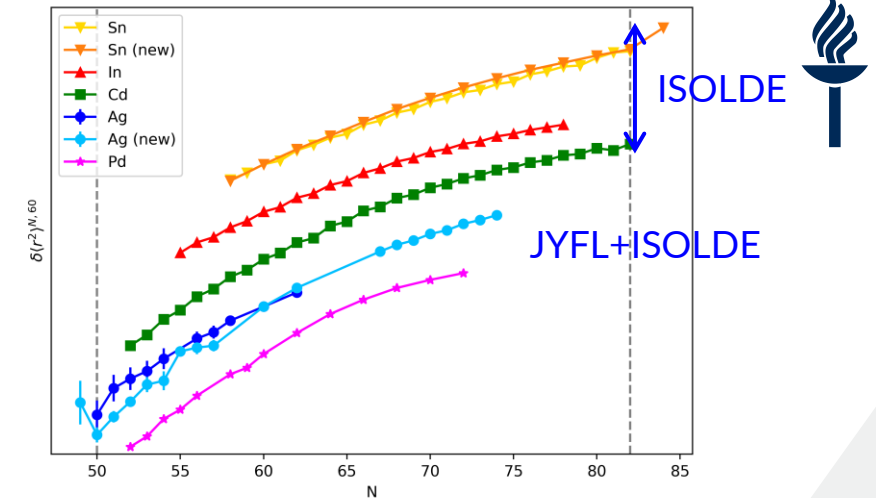
- $N=50$
 - Charge radii, moments
 - Pd and Rh accessible in high-resolution
- $N=Z$
 - Masses
 - Tin-100 accessible with Penning trap.
 - Charge radii, moments
 - ^{94}Ag , ^{100}Sn
- Beyond $N=Z$
 - Masses

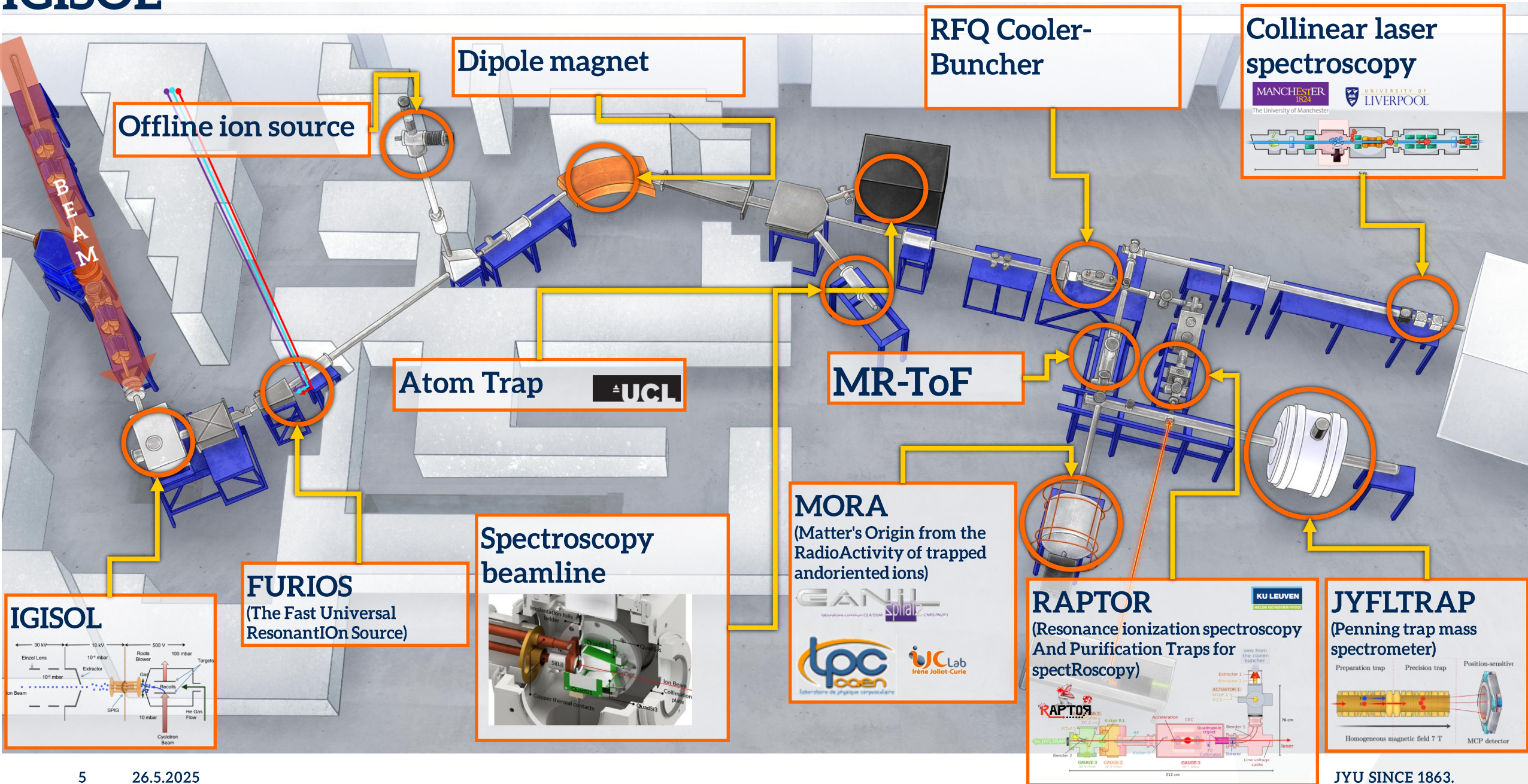
$N=82$

Motivation: Nuclear structure trends

- Electromagnetic moments, charge radii and binding energies are essential* benchmarks of theory
- Region below Sn ($Z = 50$) is of high interest Data on neutron-rich odd-odd Ag isotopes is absent or incomplete
- Spin assignments are often tentative and in conflict to experiments lacking isomeric or isobaric purification
- High precision mass and laser spectroscopy measurements can be combined with decay spectroscopy data

*B. van den Borne, Phys. Rev. C 111, 014329





Inductively Heated Hot Cavity Catheter Laser Ion Source



- Implantation of reaction products to as solid material
- The high temperature => fast diffusion to cavity volume
- Effusing atoms selectively ionized with lasers
- Based on GSI catcher ion source systems

R. Kirchner et al., NIM 186, 1-2, (1981)

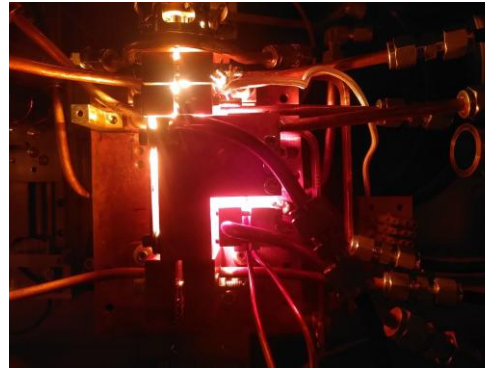
^{101}Sn : Z Janas et al 1995 Phys. Scr. **262** (1995)

^{100}In : W. Kurcewicz, Z. Phys. A - Atoms and Nuclei **308**, 21-31 (1982)

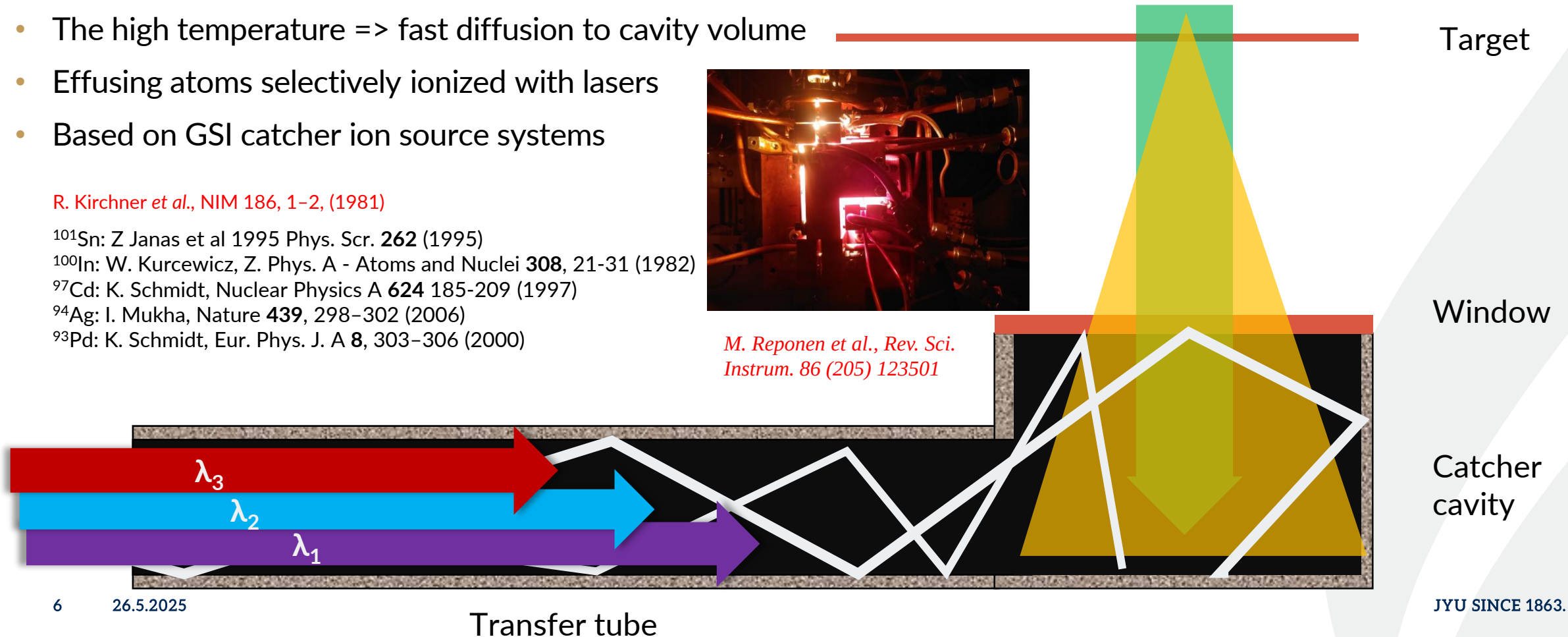
^{97}Cd : K. Schmidt, Nuclear Physics A **624** 185-209 (1997)

^{94}Ag : I. Mukha, Nature **439**, 298-302 (2006)

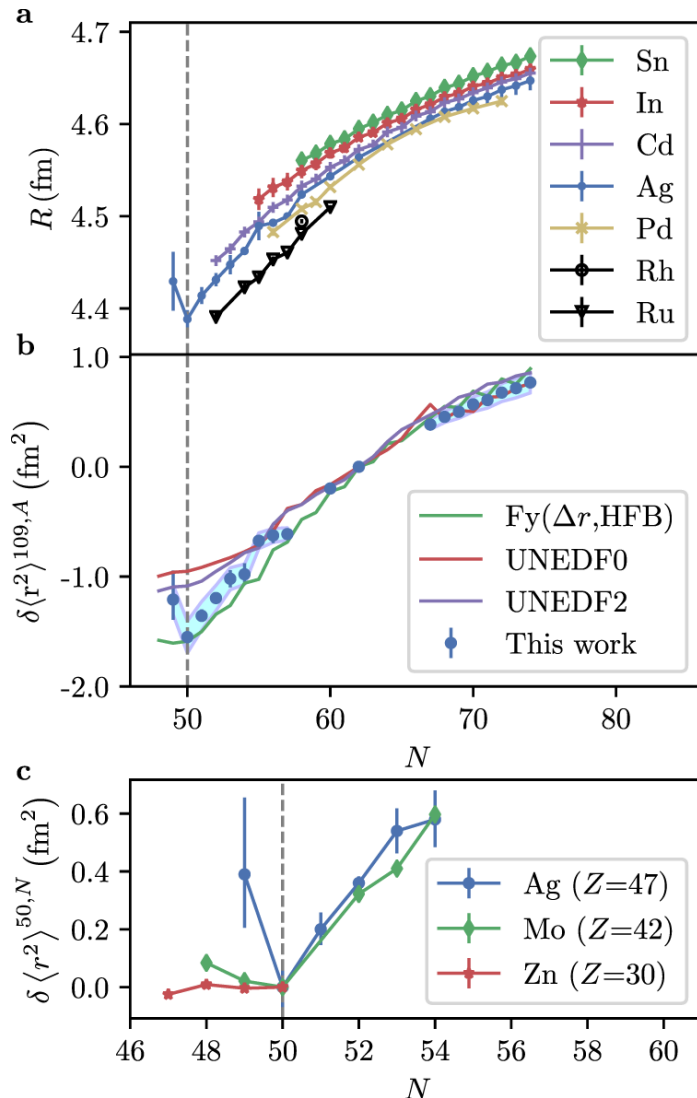
^{93}Pd : K. Schmidt, Eur. Phys. J. A **8**, 303-306 (2000)



M. Reponen et al., Rev. Sci. Instrum. 86 (205) 123501



Evolution of charge radii across $N = 50$



- Measurements cross $N=50$ shell closure in the region of ^{100}Sn for the first time.
- UNEDF functionals predict a rather smooth behaviour; Fayans EDF better reproduces local variations
- None of the models reproduces the remarkably sharp increase in crossing $N=50$

$$\delta \nu_i^{A,A'} = M_i \frac{A' - A}{AA'} + F_i \delta \langle r^2 \rangle^{A,A'}$$

Atomic factors F and M from empirical estimation:

$$F = -4300(300) \text{ MHz fm}^{-2}, M = 1956(360) \text{ GHz u}$$

More recent *ab initio* calculations:

$$F = -3557(49) \text{ MHz fm}^{-2}, M = 1479(14) \text{ GHz u}$$

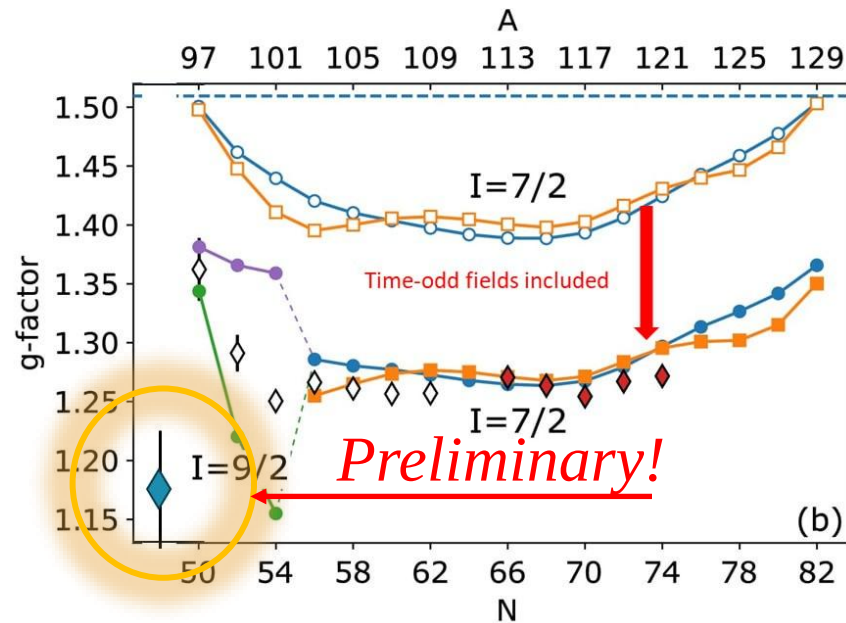
B. Ohayon et al., Phys. Rev. Res. 6 (2024) 033040

M. Reponen et al., Nature Comm. 12 (2021) 4596

Towards the $N=Z$ line

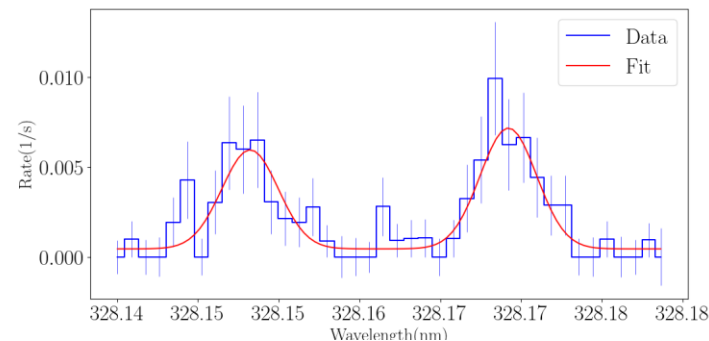
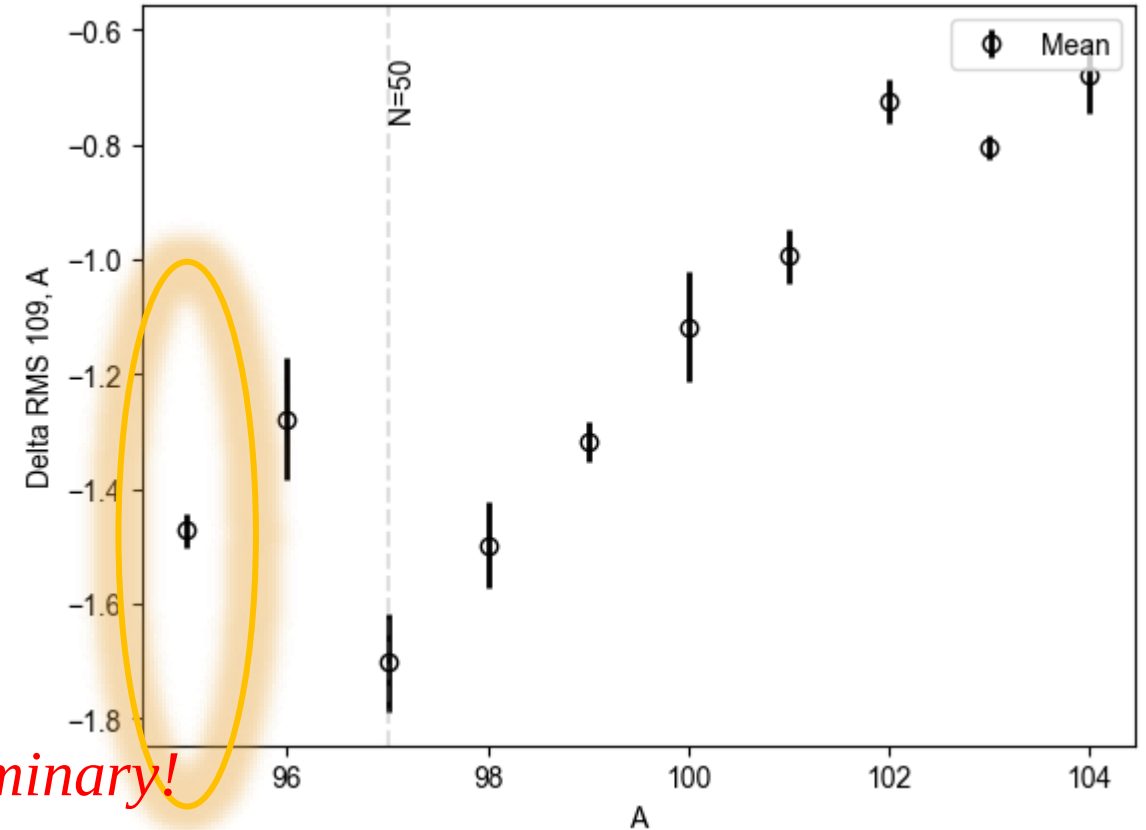


- New data for $^{97,96,95}\text{Ag}$ using $^{40}\text{Ca}(^{58/60}\text{Ni}, \text{pxn})$
 - confirms the large kink at $N=50$.
- The g -factor of ^{95}Ag exhibits possibly a large jump after $N=50$

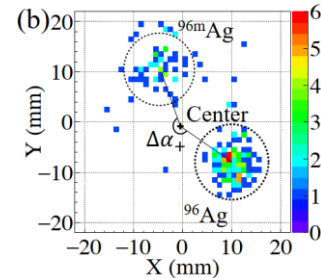


R.P. de Groote et al., Phys. Lett. B 848 (2024) 138352

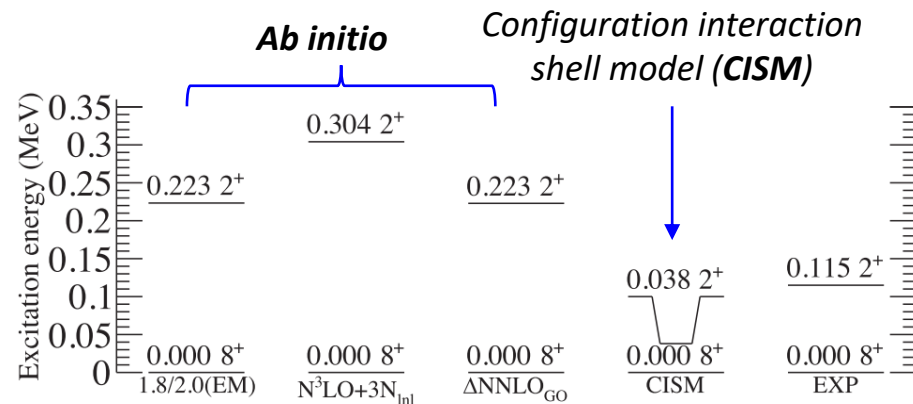
Preliminary!



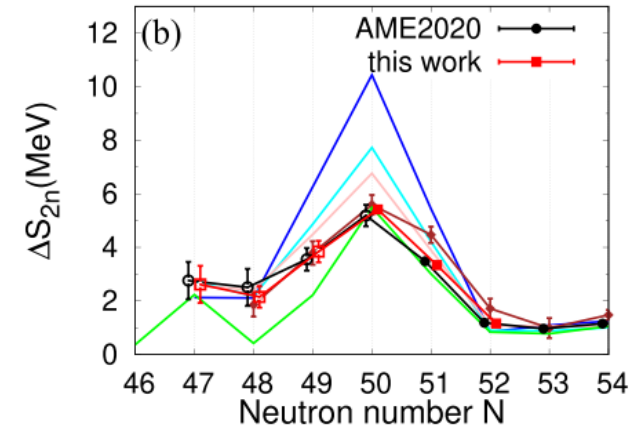
Mass measurements beyond $N = 50$



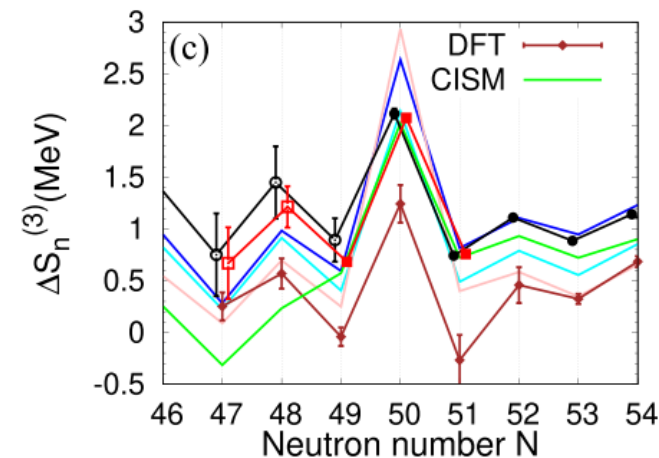
- Probing the robustness of the $N = 50$ shell closure
- Atomic masses of β -decaying 2^+ and 8^+ states identified in ^{96}Ag and measured for the first time
- Excitation energy of $^{96\text{m}}\text{Ag} (2^+) = 115.0(17)$ keV
- Atomic mass of ^{95}Ag measured for the first time (precision of 1.4 keV)



Z. Ge, M. Reponen et al., Phys. Rev. Lett. 133 (2024) 132503



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

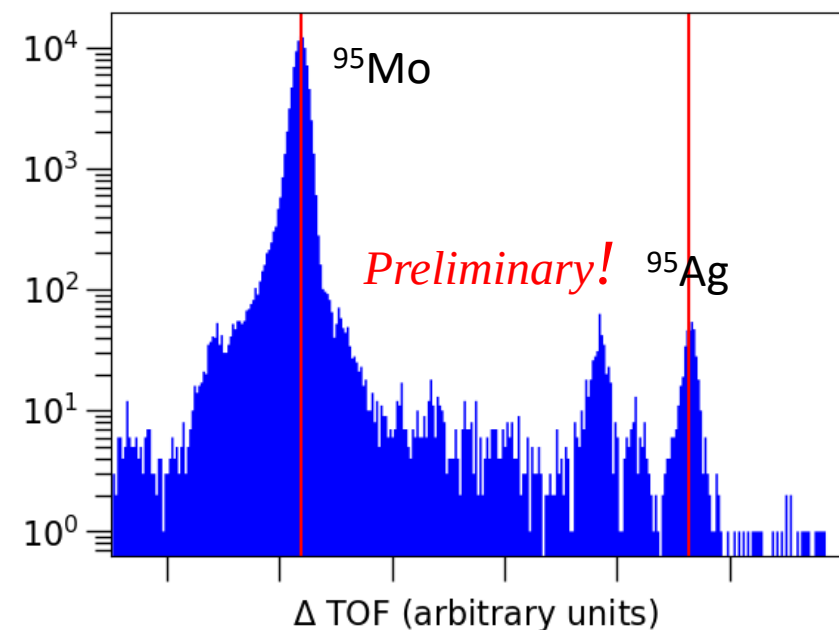


$$\Delta S_n^{(3)} = (-1)^N [S_n(Z, N) - S_n(Z, N + 1)]/2$$

MR-TOF Mass Spectrometry of $N = Z$ ^{94}Ag

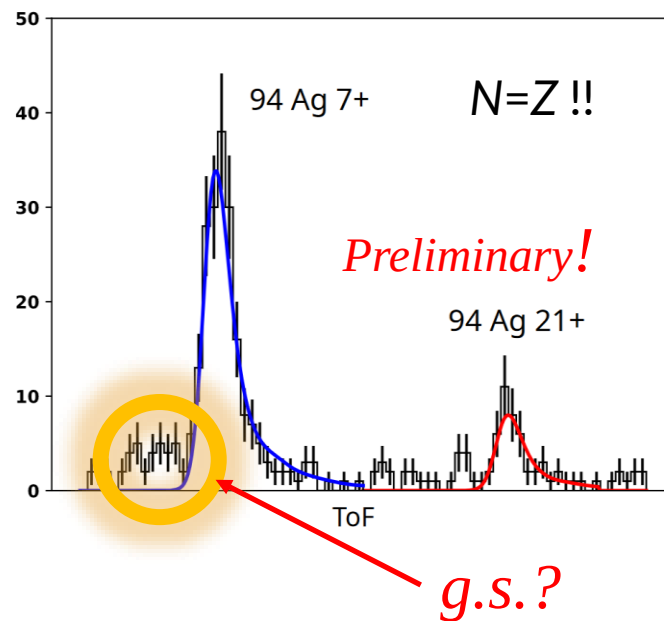


- The reliability of the MR-TOF method was verified with ^{94}Zr and ^{95}Ag (the latter measured at JYFLTRAP to 1.4 keV)
- $^{58}\text{Ni}(^{40}\text{Ca}, \text{xpxn})^{95,94}\text{Ag}/^{93,92}\text{Pd}$

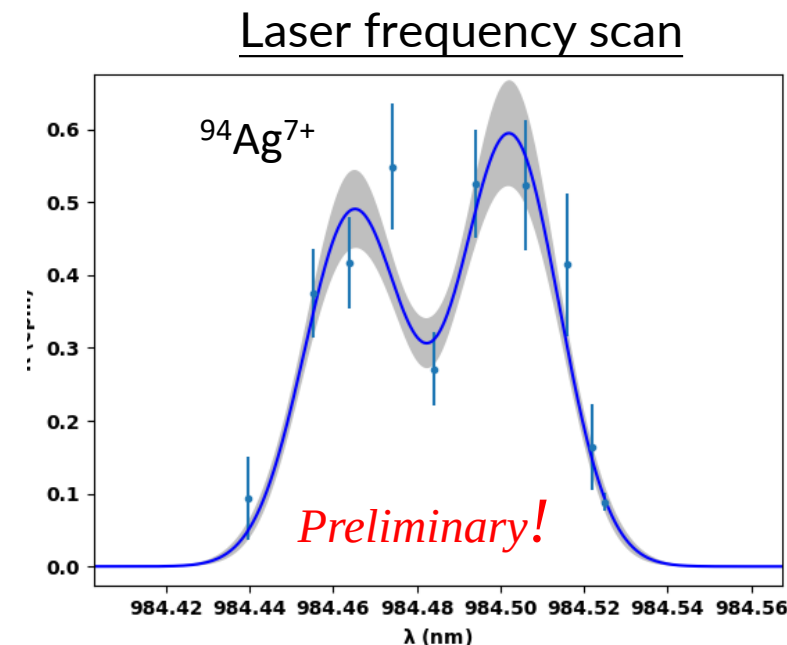


Mass precision of ^{95}Ag ~ 10 keV

V. Virtanen, M. Reponen et al., to be submitted



Mass precision of $^{94}\text{Ag}^{21+}$ ~ 90 keV



Ion rates

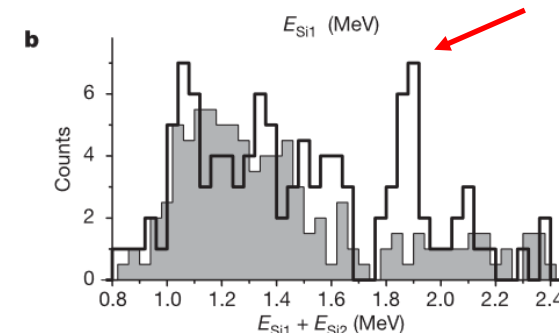
- $(7^+) = 0.55$ ions/min
- $(21^+) = 0.15$ ions/min

Energetics of the (21+) isomer in ^{94}Ag

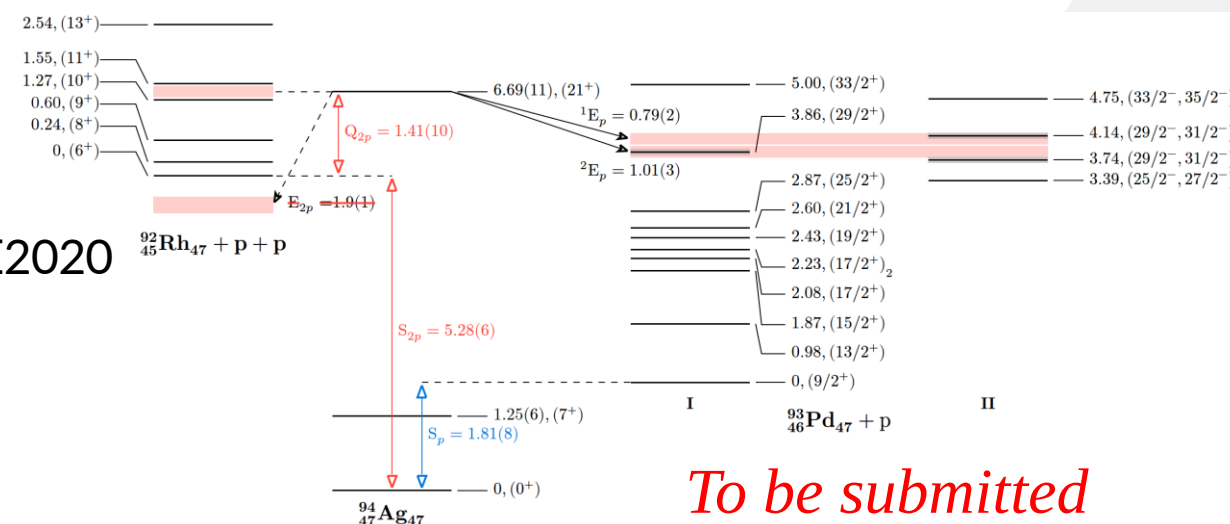


$$E_{2p} = 1900(100) \text{ keV}$$

- ME of the (21⁺) state measured with ~90 keV uncertainty
- Combined with the ^{92}Rh ground state ME (JYFLTRAP) =>
 $Q_{2p} \sim 1.4 \text{ MeV}$ (to the ground state),
 - Enforces an angular momentum difference >10.
- Candidate peak for $^{94}\text{Ag}^{(0+)}$
 - ME in line with Ft-based Q_{EC} estimates and SM calculations, but to be confirmed!
 - About 1.3 MeV more bound compared to AME2020



*I. Mukha et al.,
Nature 439 (2006)
298*



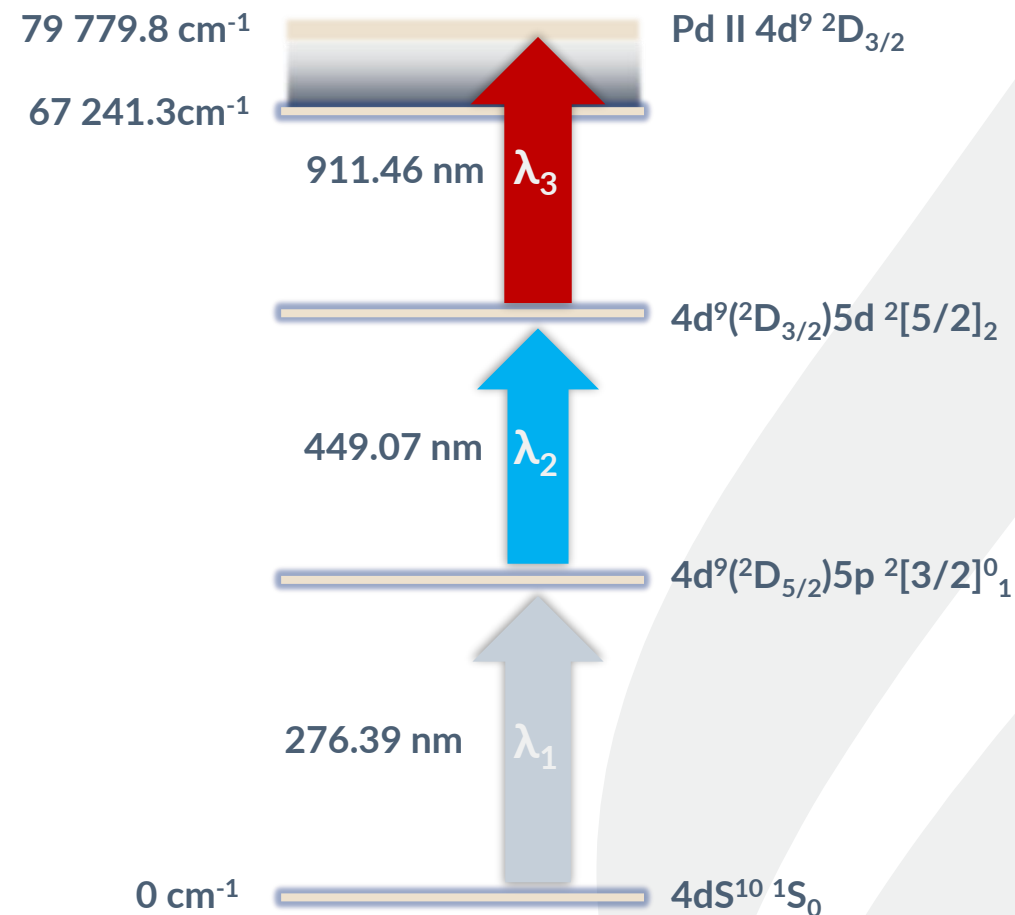
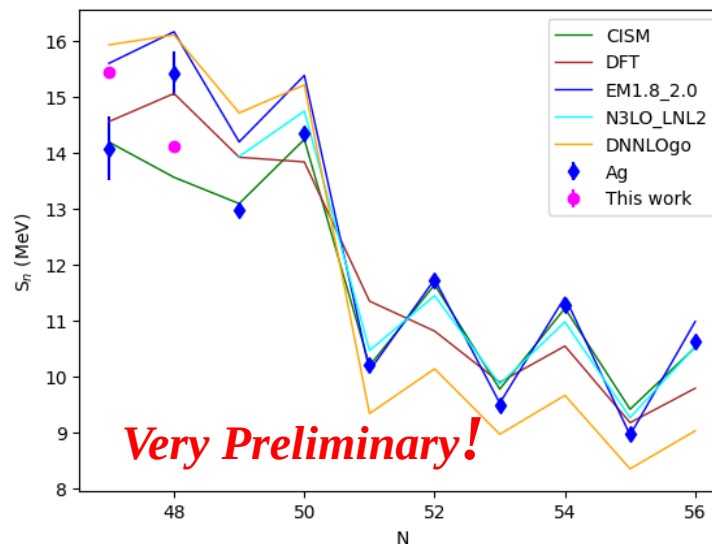
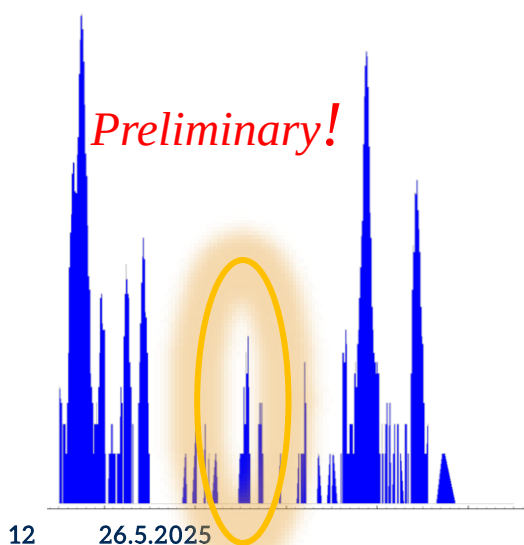
To be submitted

*Also measured with the FRS Ion Catcher at GSI (G. Kriepko-Koncz, submitted)

MR-TOF Mass Spectrometry of $N = Z$ ^{92}Pd



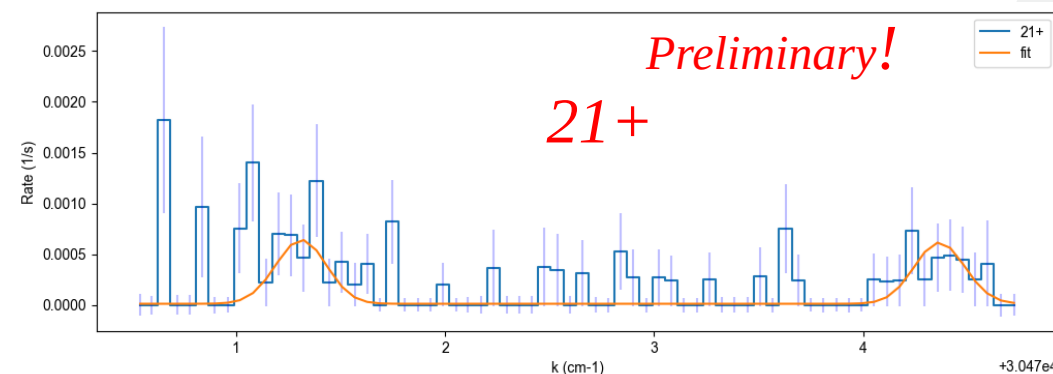
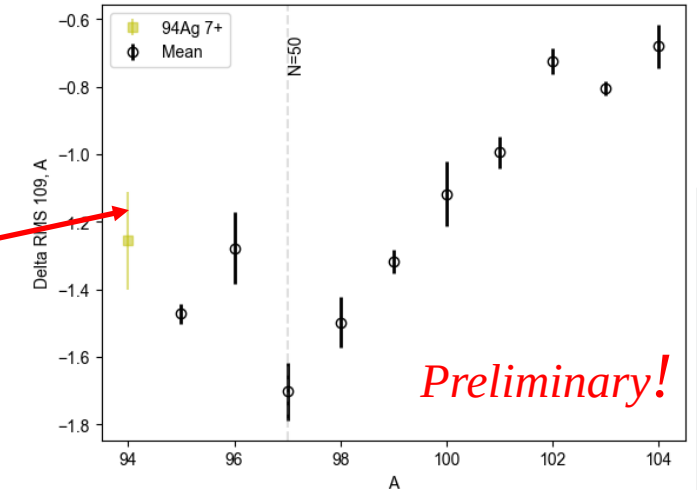
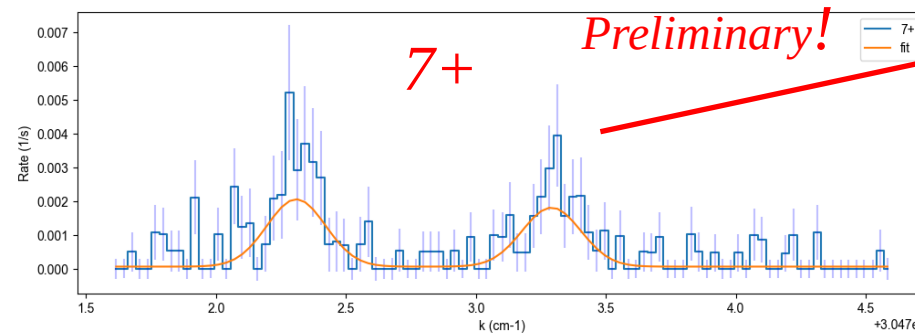
- $^{58}\text{Ni}(^{40}\text{Ca}, 4\text{p}2\text{n}) ^{92}\text{Pd}$
- ^{92}Pd ME close to the AME2020 extrapolation
- Extends silver masses beyond the $N=Z$ line*
- Benchmark for SM** and DFT calculations***
 - More theory work under way!



Laser spectroscopy at the $N=Z$ line



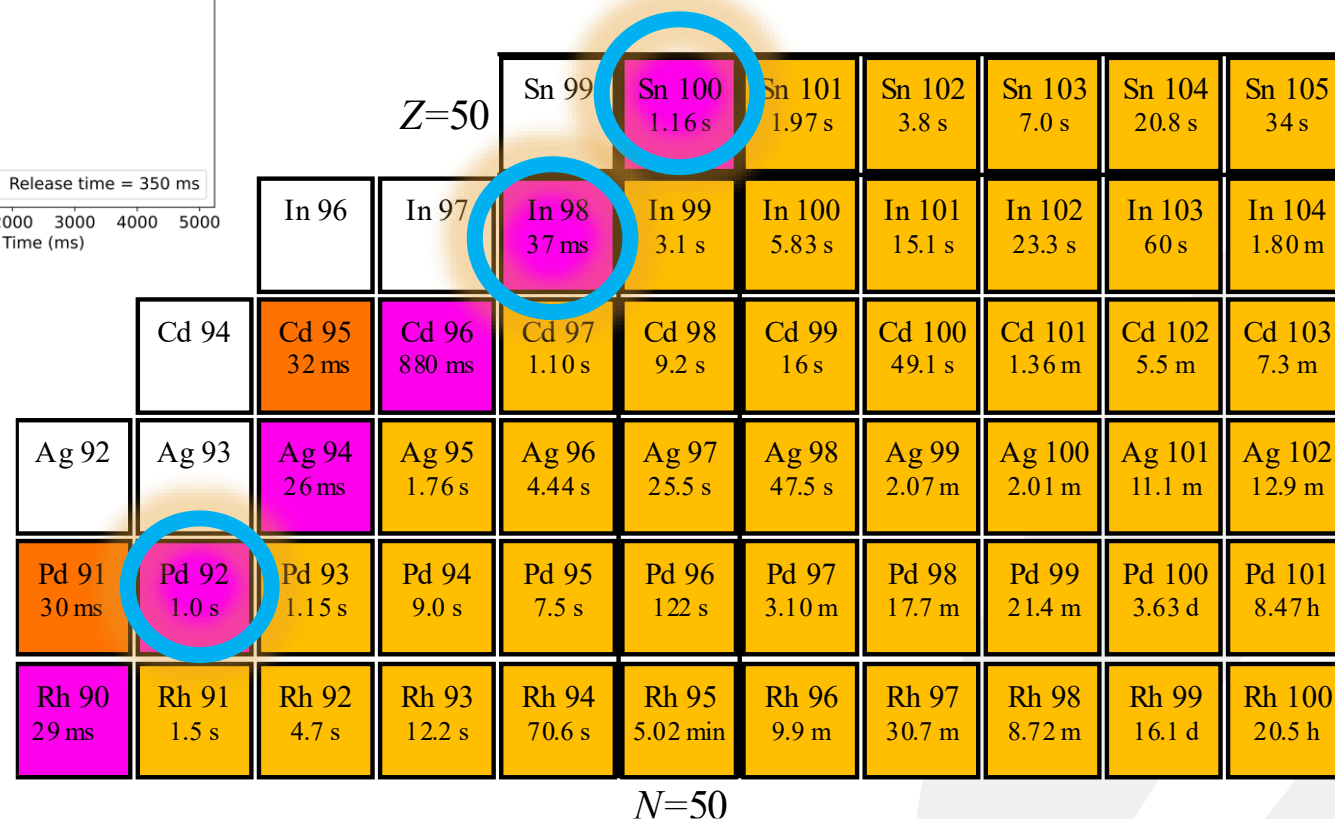
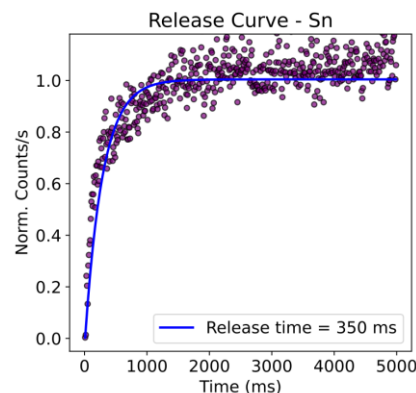
- MR-ToF assisted in-source RIS of ^{94}Ag .
 - $7+$ and $21+$ states observed
- Analysis under way
 - Charge radii
 - Magnetic moment
- Statistics to be improved in near future
 - **However, this already marks the first laser spectroscopy of a proton emitter!**



Outlook:



- Pd: In principle feasible
 - RIS scheme insensitive*.
- Sn: Release time enough for tin-100**, estimated yield comparable to $^{94}\text{Ag}^{(21+)}$.
 - JYFLTRAP measurement feasible.
 - RIS sensitivity for even cases.
- Release test for indium to be done
 - Targeting ^{98}In .
- Beyond $N=Z$
 - Development work ongoing for ^{91}Pd and ^{95}Cd



** S. Chinthakayala thesis work

*A. Ortiz Cortes, JYU Dissertations 2023





Acknowledgements

The IGISOL team:

Particularly:

- Ville Virtanen (MR-TOF)
- Zhuang Ge (PI-ICR)
- Sonja Kujanpää (Lasers)
- Saikumar Chinthakayala (Lasers)

Collaborators:

- R. P. de Groote (KU Leuven)
- M. Mukai (KEK)



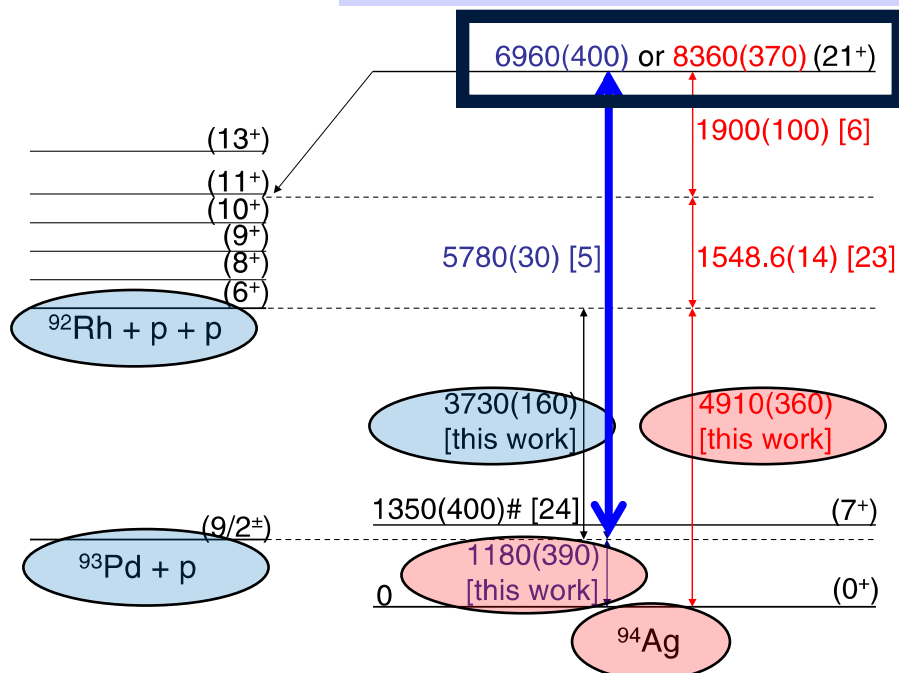
Motivation: two-proton radioactivity in $N = Z$ ^{94}Ag ?



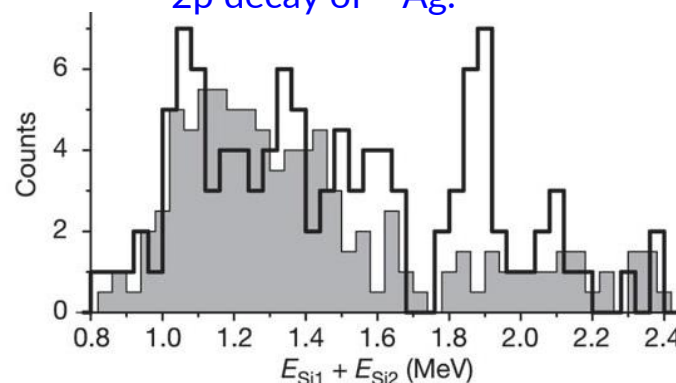
The high-spin isomer (21^+) in ^{94}Ag ($T_{1/2} = 0.39(4)\text{s}$) has been observed to decay via:

- β -decay *C. Plettner et al., Nucl. Phys. A 733 (2004) 20*
- β -delayed proton emission *I. Mukha et al., Phys. Rev. C 70 (2004) 044311*
- direct 1p-decay (to ^{93}Pd) *I. Mukha et al., Phys. Rev. Lett. 95 (2005) 022501*
- direct 2p-decay (to ^{92}Rh) *I. Mukha et al., Nature 439 (2006) 298*

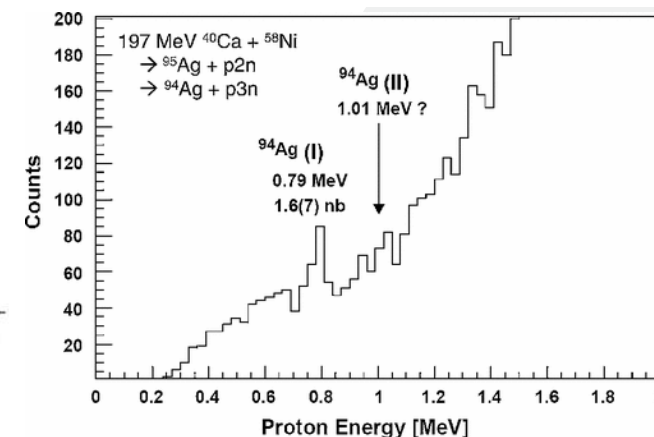
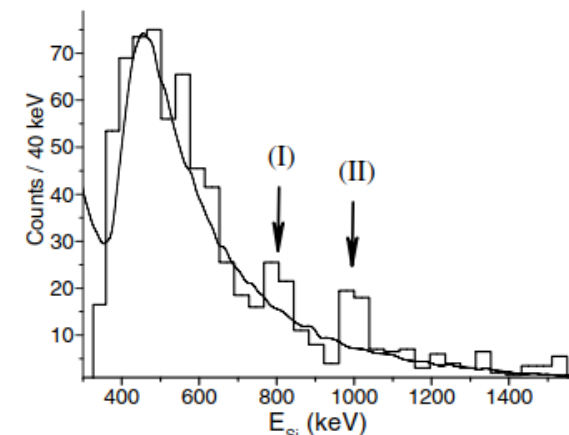
INTENSE SCIENTIFIC DISCUSSIONS!!



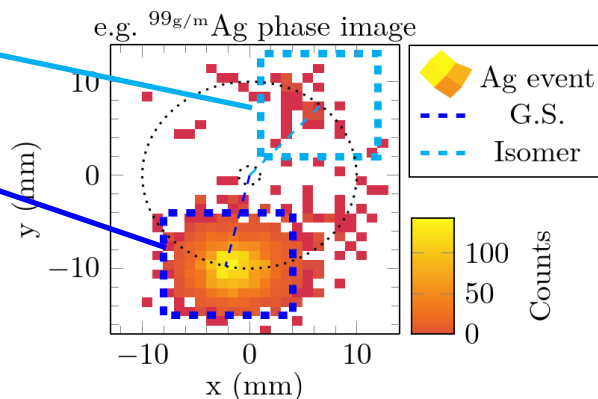
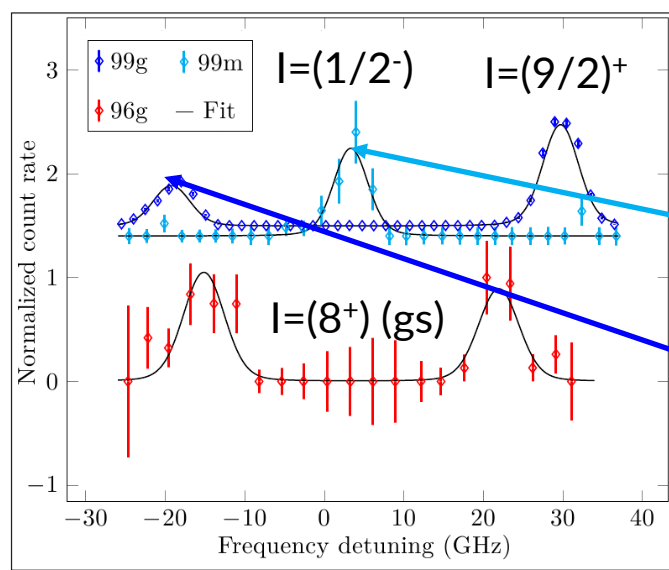
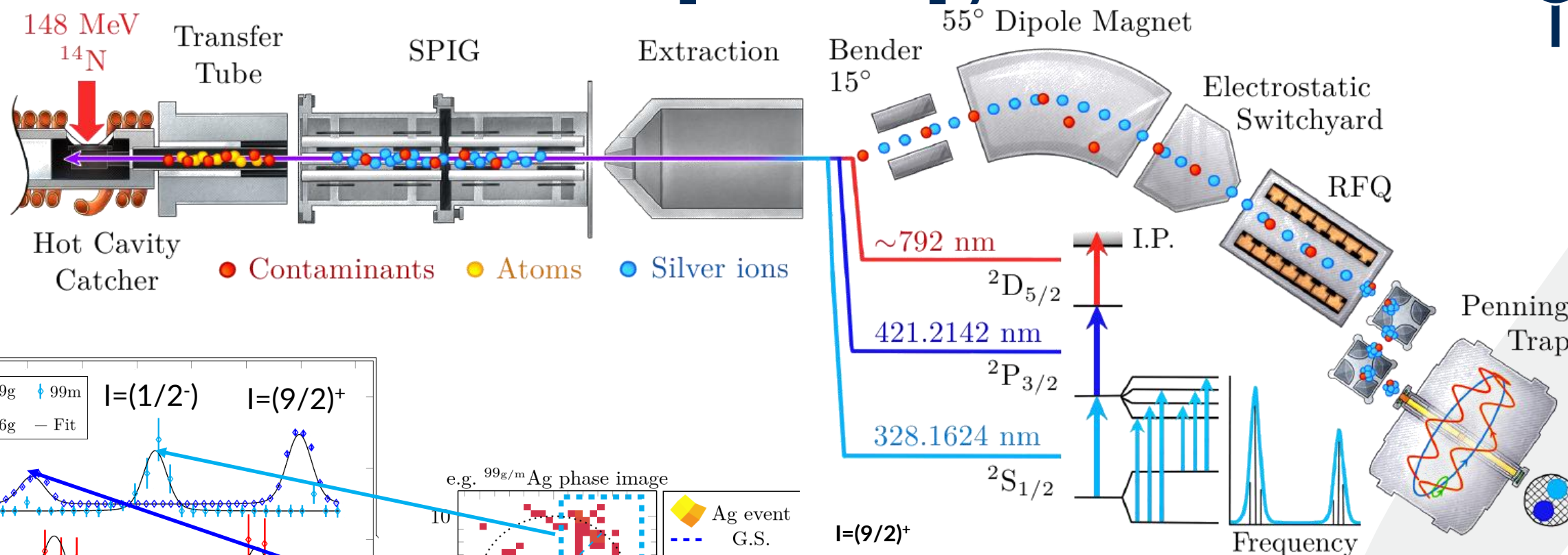
Si_1 - Si_2 - γ - γ coincidence, with double gates on ^{92}Rh . Peak at 1.9 MeV interpreted as 2p decay of ^{94}Ag .



Peaks (I) and (II) indicate direct p-decay into ^{93}Pd . Using Si- γ - γ coincidence)



PI-ICR assisted in-source spectroscopy

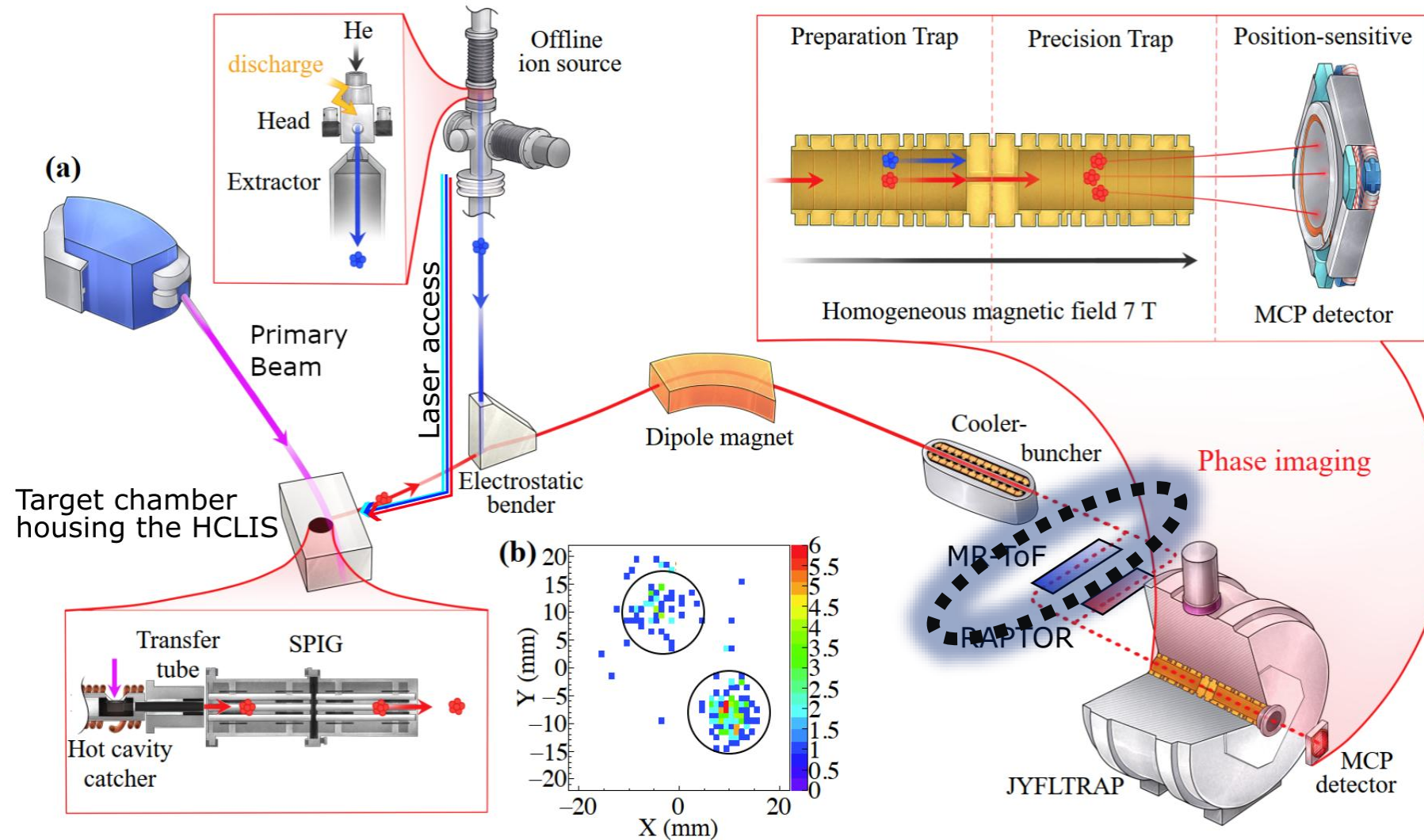


$I=(9/2^+)$

$I=(1/2^-)$

- Count ions as a function of laser frequency
- Different gates on detector select different nuclear state
- ^{96}Ag "on resonance" 1 ion per 5 mins.
- Further improvement of ^{96}Ag yield to 1 ion per 25 seconds

Probing the $N=Z$ line



JYFL-ACCLAB



C-Linac

MCC30
E_{max}=30 MeV

K130:
E_{max}= 130 (q^2/A) MeV
Beams from p to Au
Annual use: 6500-7000 h/year

IGISOL

- Optical spectroscopy
- Atomic masses
- Decay spectroscopy

RADEF

- Radiation effects
- Microfilters

LSC

- Nuclear reactions

RITU/MARA/ MARA-LEB

- Nuclear spectroscopy
- (Optical spectroscopy)

Ion sources

ECR: (6.5 GHz, 14 GHz, 18 GHz)
Multicusp: (H-,D-)

Pelletron

E_{MAX}=1.7 MeV

Accelerator based material physics

- ToF-ERDA/RBS/PIXE
- Ion source development (Lasers + Cs sputtering)

A nuclear fingerprint on the atomic levels

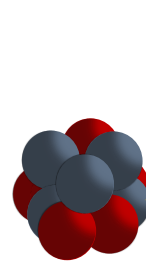
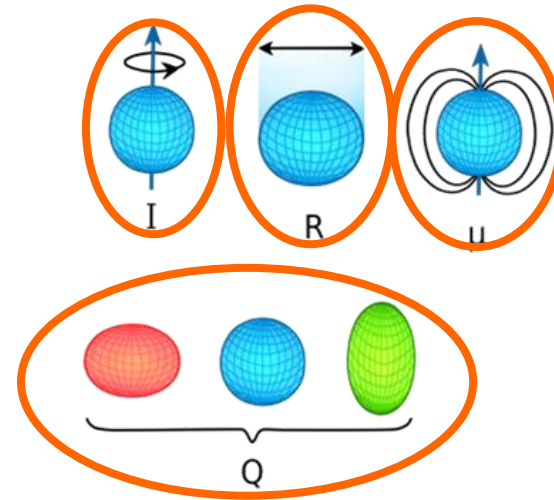


- Nuclear properties are mapped to the atomic structure.
 - Nuclear spin I
 - Changes in the mean square charge radii $\delta\langle r^2 \rangle$
 - Magnetic dipole moment μ
 - Electrical quadrupole moment Q

$$\delta\nu^{AA'} = M \frac{m_{A'} - m_A}{m_A m_{A'}} + F \delta\langle r^2 \rangle^{AA'}$$

$$g = \mu/I$$

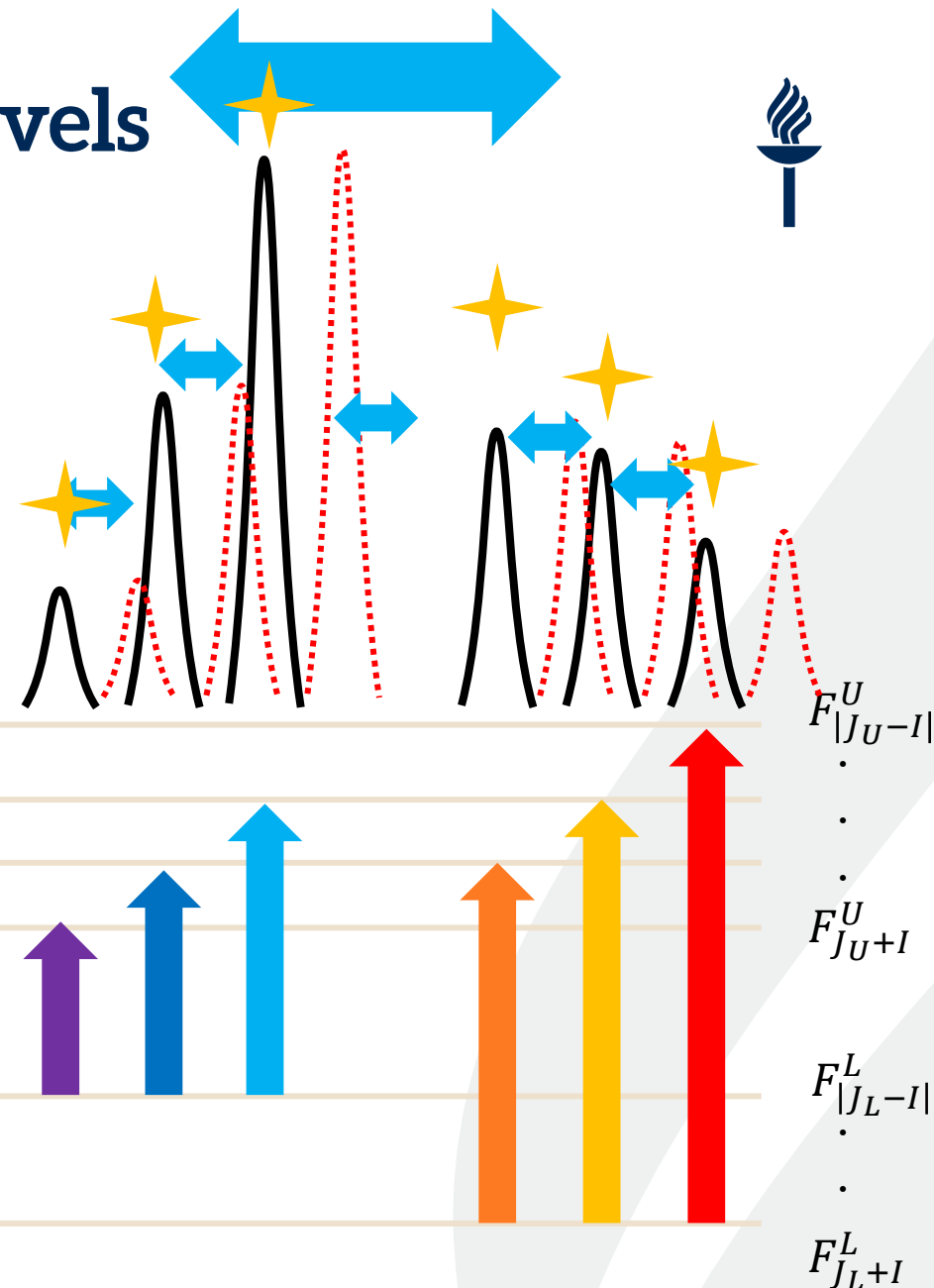
Information on the configuration.



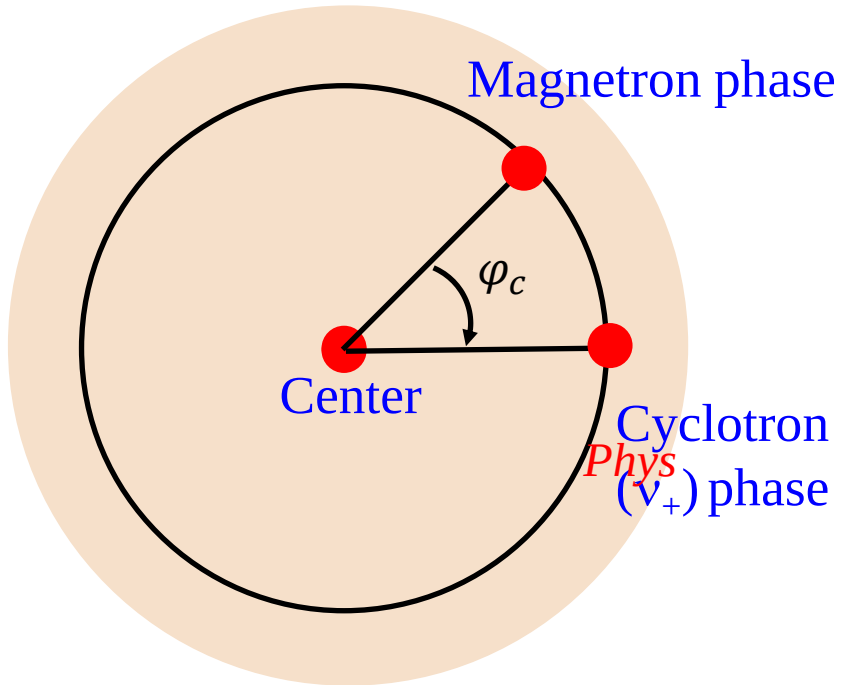
I

$|J_U\rangle$

$|J_L\rangle$

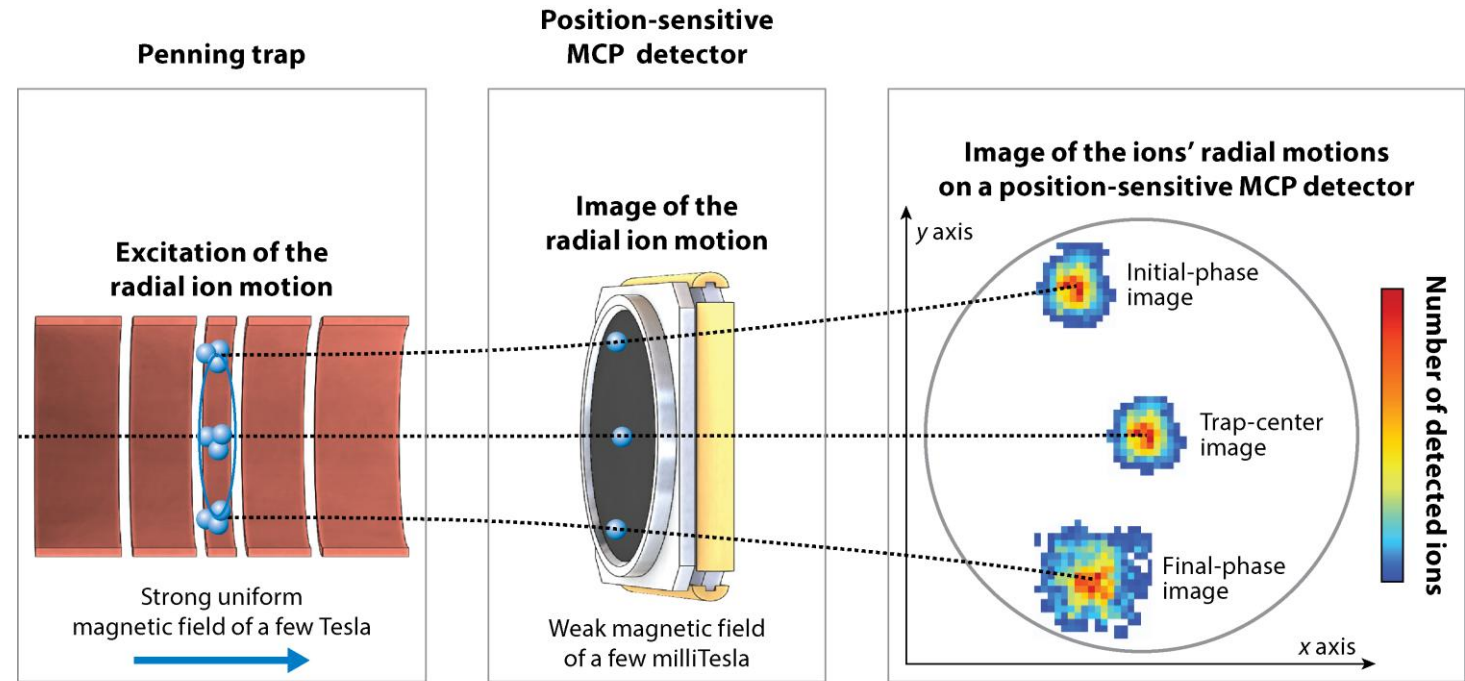


Phase-Imaging Ion Cyclotron Resonance method



$$\nu_c = \frac{\varphi_c + 2\pi n_c}{2\pi t}$$

T. Eronen et al., Eur. Phys. J. A 48 (2012) 46
S. Eliseev et al., Appl. Phys. B 114 (2014) 107
D.A. Nesterenko et al., Eur. Phys. J. A 54 (2018) 154



Dilling J, et al. 2018.
Annu. Rev. Nucl. Part. Sci. 68:45–74

J. Dilling et al., Annu. Rev. Nucl. Part. Sci. 68 (2018) 45

$n_{\pm}$ = number of turns
 $\phi_{\pm}$ = phase of the radial frequency $\nu_{\pm}$
 t = phase accumulation time

Offers a 5-fold gain in precision of the cyclotron-frequency determination, and 40-fold gain in resolving power.

Summary

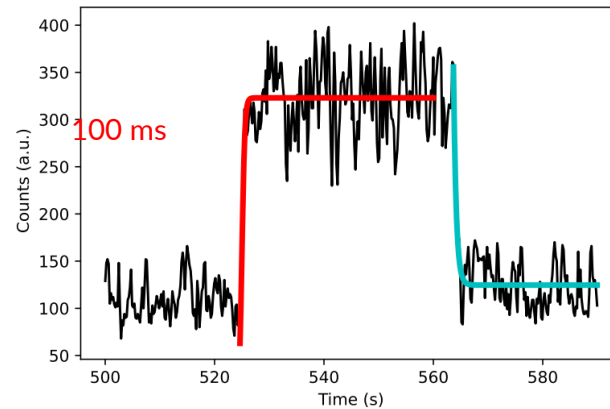


- The charge radii trend has crossed the $N = 50$ shell closure in the vicinity of ^{100}Sn for the first time, challenging modern nuclear energy density functionals.
- High-precision mass measurements revealed the robust nature of the $N = 50$ shell.
- The new measurement techniques have finally revealed the energetics of the isomeric states in $N = Z$ ^{94}Ag .
- The mass excess of $N = Z$ ^{92}Pd has been determined.
 - The charge radius of the isomeric states in ^{94}Ag will be run soon (Another run coming up in near future).
 - Feasibility for probing the charge radii of tin-100 demonstrated.
- A favourable outlook on advancing beyond $N=Z$ with a possibility of enabling a new research opportunities in the lanthanide region.

Outlook: Hot cavity development: Beyond $N=Z$ and towards proton emitters.

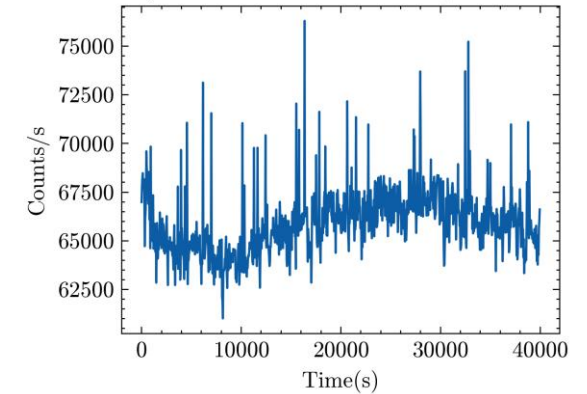


- The current HCLIS iteration maybe too slow for other $N=Z$ (and beyond).
 - Yields are reasonable though!
- Definitely too slow for Lanthanides!
 - E.g. ^{146}Tm proton emitter, release studied with implanted stable tm beam
- Current material (combinations) do not allow higher temperatures
- New material needed: DLC windows, All tungsten constructs, Nb catchers:
 - Towards 2800 K



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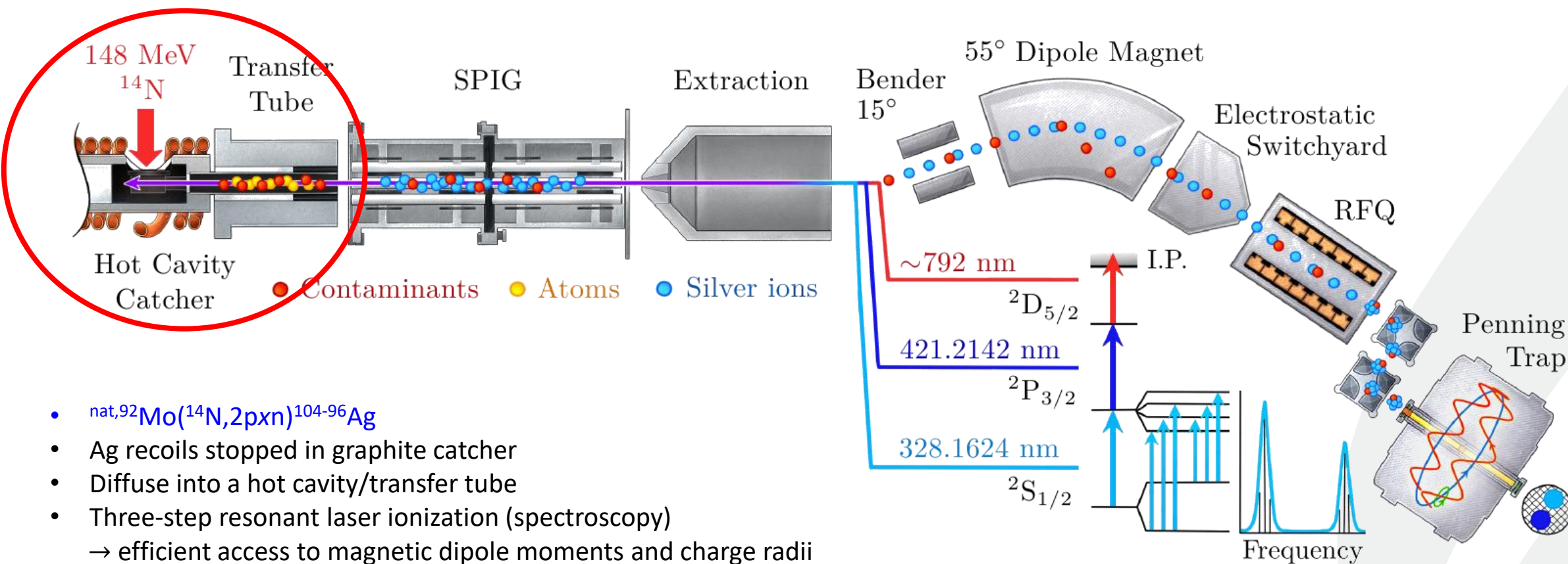
Tm TDC 40s Cycle @ 1467W C-Catcher, Ta lined transfer tube



PI-ICR assisted in-source spectroscopy



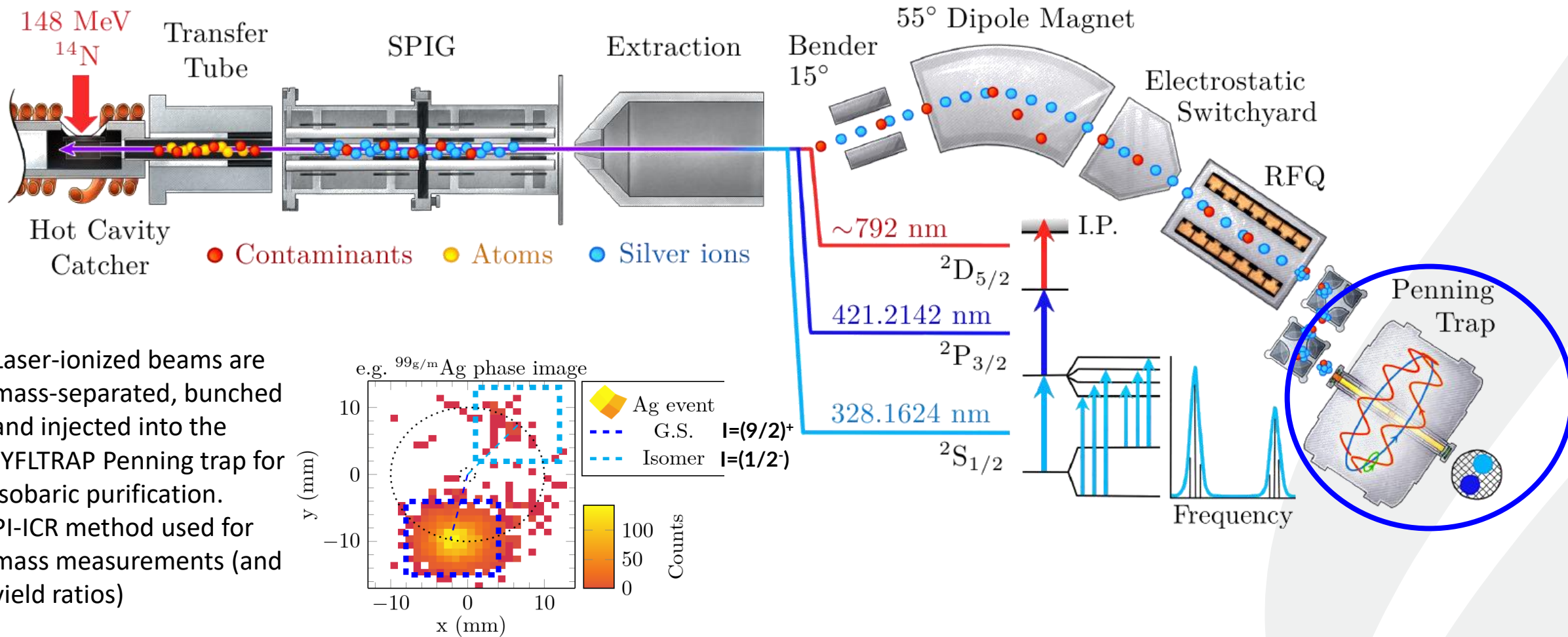
Recent efforts to combining methods from ion manipulation and trapping techniques with laser resonance ionization have enabled almost a complete suppression of background, opening a window to the most exotic nuclei.



- $^{nat,92}\text{Mo}(^{14}\text{N}, 2\text{pxn})^{104-96}\text{Ag}$
- Ag recoils stopped in graphite catcher
- Diffuse into a hot cavity/transfer tube
- Three-step resonant laser ionization (spectroscopy)
→ efficient access to magnetic dipole moments and charge radii

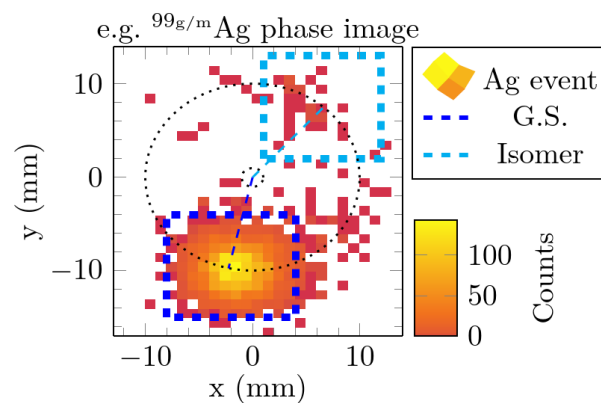
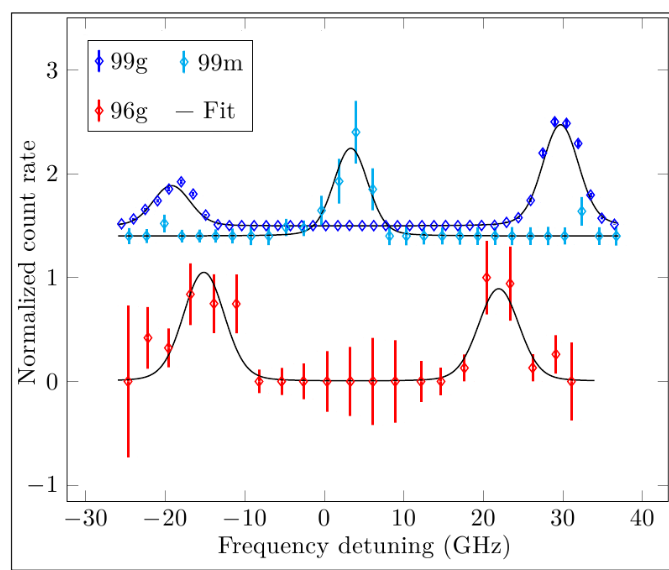
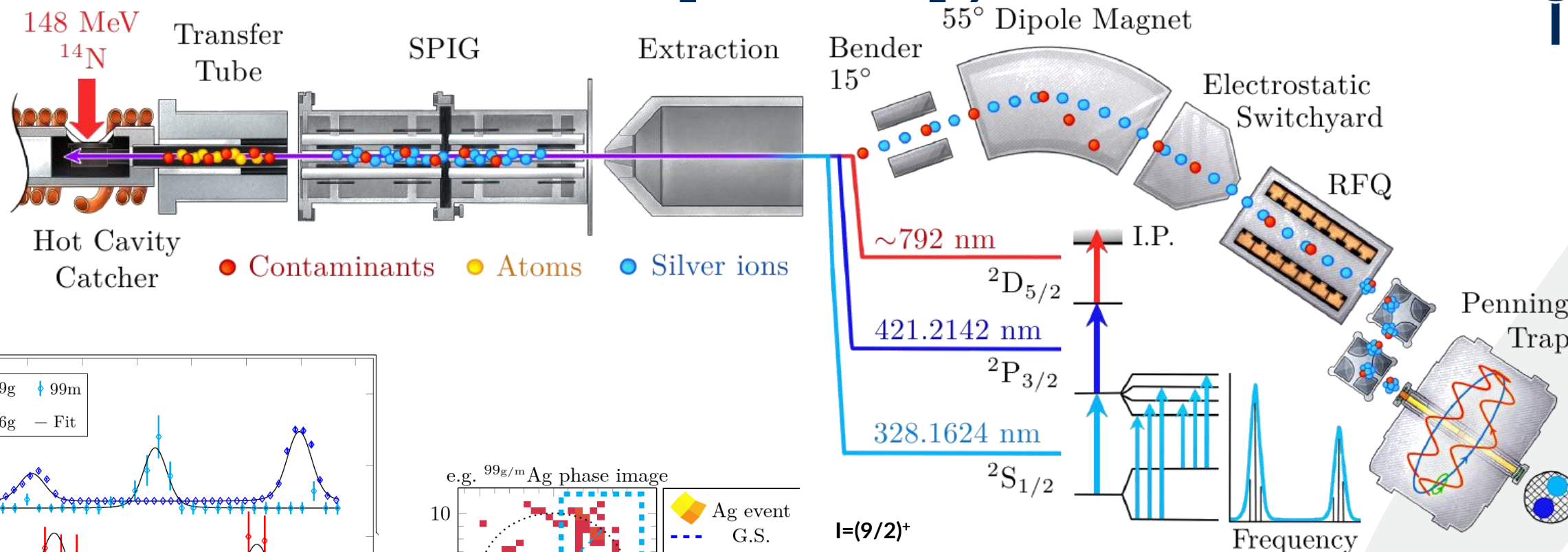
T. Eronen et al., Eur. Phys. J. A 48 (2012) 46
S. Eliseev et al., Appl. Phys. B 114 (2014) 107
D.A. Nesterenko et al., Eur. Phys. J. A 54 (2018) 154

PI-ICR assisted in-source spectroscopy



- Laser-ionized beams are mass-separated, bunched and injected into the JYFLTRAP Penning trap for isobaric purification.
- PI-ICR method used for mass measurements (and yield ratios)

PI-ICR assisted in-source spectroscopy



$I=(9/2)^+$

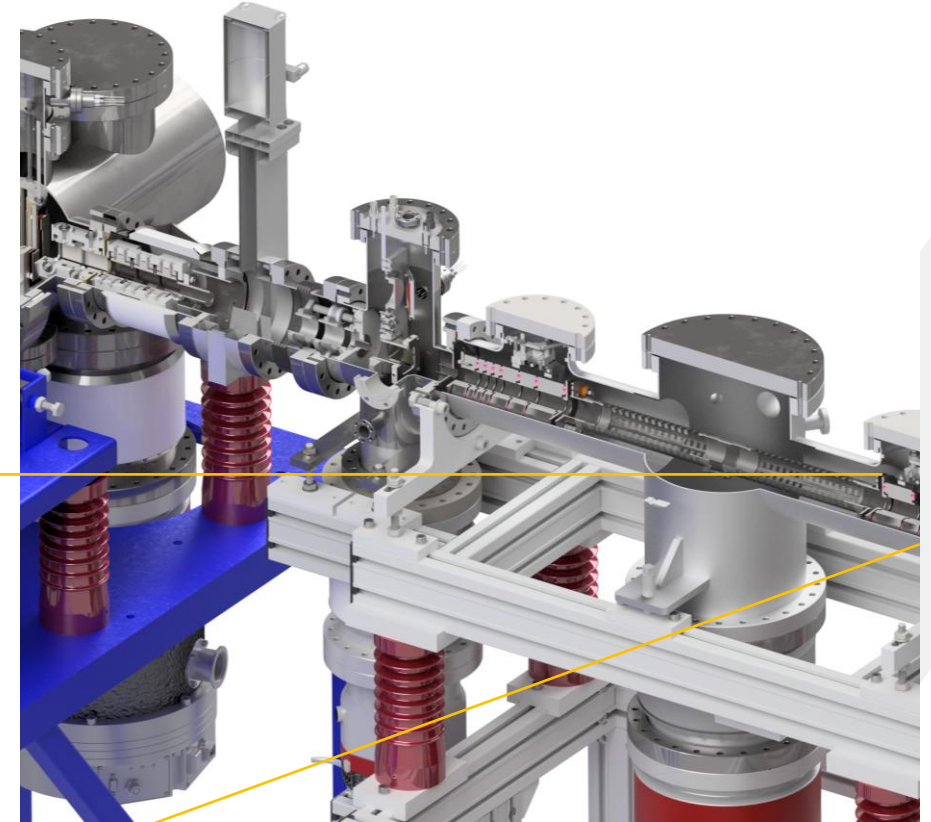
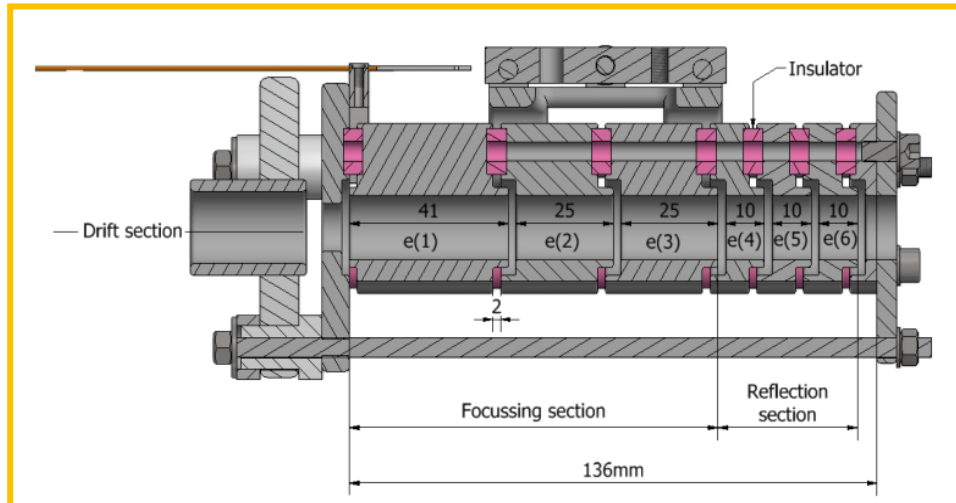
$I=(1/2)^-$

- Count ions as a function of laser frequency
- Different gates on detector select different nuclear state
- ^{96}Ag "on resonance" 1 ion per 5 mins.
- Further improvement of ^{96}Ag yield to 1 ion per 25 seconds

Multi-Reflection Time-of-Flight Mass Spectrometer



- Works as a separator and as a spectrometer
- Non-scanning – measures full mass spectrum simultaneously
- Fast -> exotic radioactive nuclei
- Ions trapped with the same energy -> $t = a \sqrt{\frac{m}{q}} + b$



$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t} \sim 200\,000$$