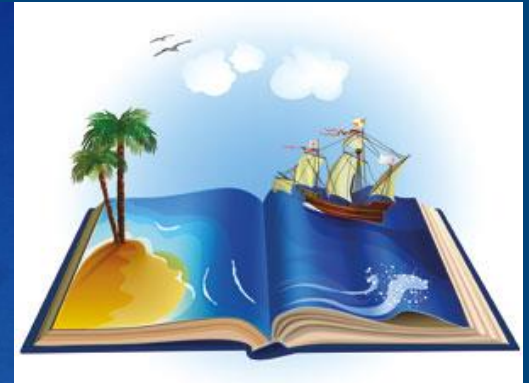


GSI Colloquium 19.05.2015

# Investigating the Atomic and Nuclear Properties of the Heaviest Elements



**Michael Block**

**GSI Darmstadt**

**Helmholtzinstitut Mainz**

**Institut für Kernchemie der Universität Mainz**

# Outline

- Status of superheavy element (SHE) research
- Basics of Penning trap mass spectrometry (PTMS)
- Direct mass measurements of nobelium and lawrencium isotopes
- New developments and selected results related to neutrino physics
- Basics of resonance ionization laser spectroscopy (RIS)
- Experimental efforts towards RIS of  $^{254}\text{No}$  at GSI
- Summary and conclusions

# SHIPTRAP Collaborators



D. Ackermann, K. Blaum, S. Chenmarev, C. Droese, Ch. Duellmann,  
M. Eibach, S. Eliseev, P. Filanin, F. Giacoppo, M. Goncharov, E. Haettner,  
F. Herfurth, F. P. Heßberger, O. Kaleja, M. Laatiaoui, G. Marx,  
D. Nesterenko, Yu. Novikov, W. R. Plaß, S. Raeder, D. Rodríguez,  
D. Rudolph, C. Scheidenberger, S. Schmidt, L. Schweikhard,  
P. Thirolf, G. Vorobjev, C. Weber, ...



# Laser Spectroscopy Collaborators



D. Ackermann, M. Block,  
F.P. Heßberger



H. Backe, W. Lauth



F. Lautenschläger, P. Chhetri,  
Th. Walther



M. Laatiaoui, S. Raeder



P. Kunz



R. Ferrer, P. Van Duppen



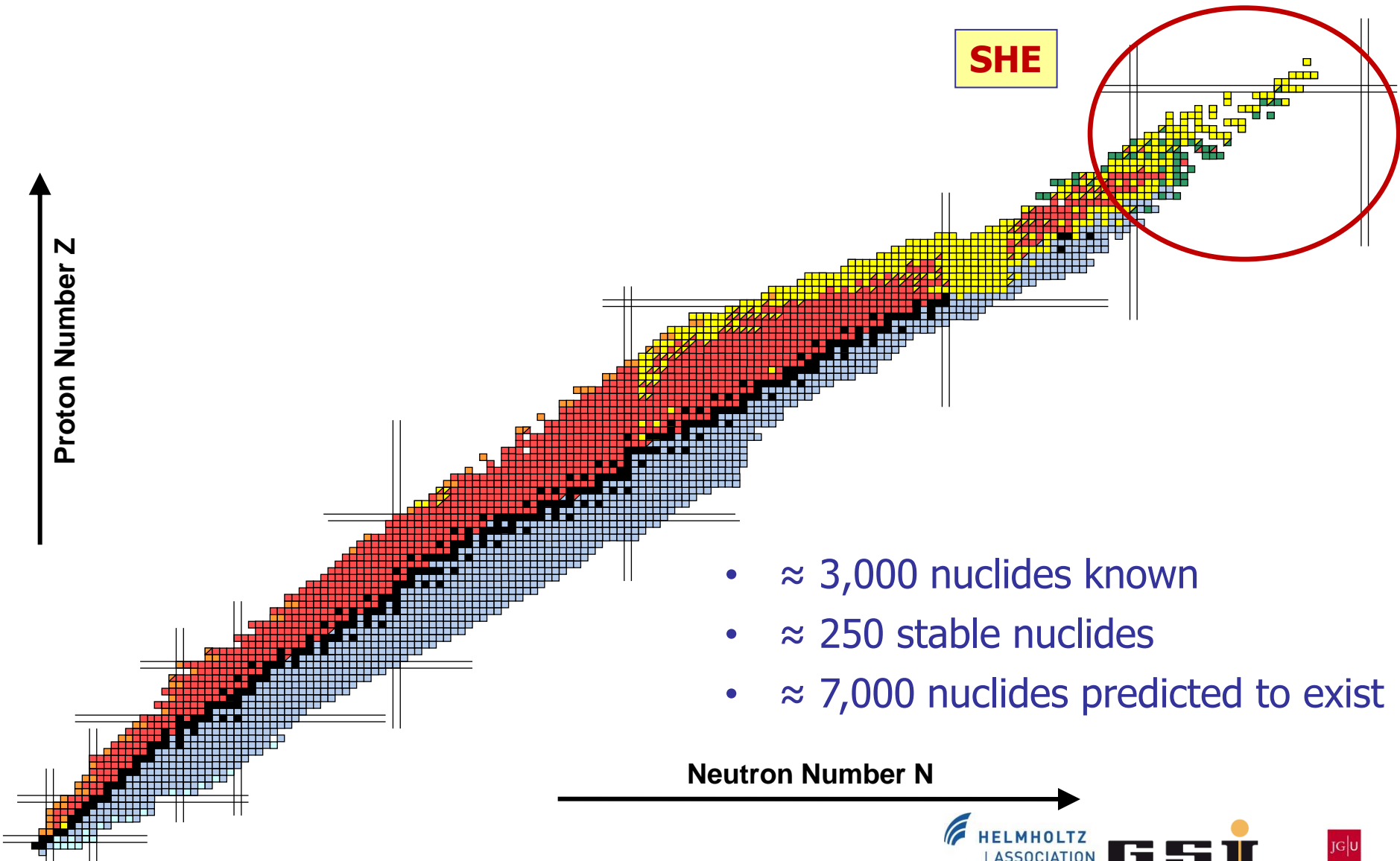
B. Cheal, C. Wraith

## ***Former members:***

*E. Minaya Ramirez, J. Even, Ch. Droese*

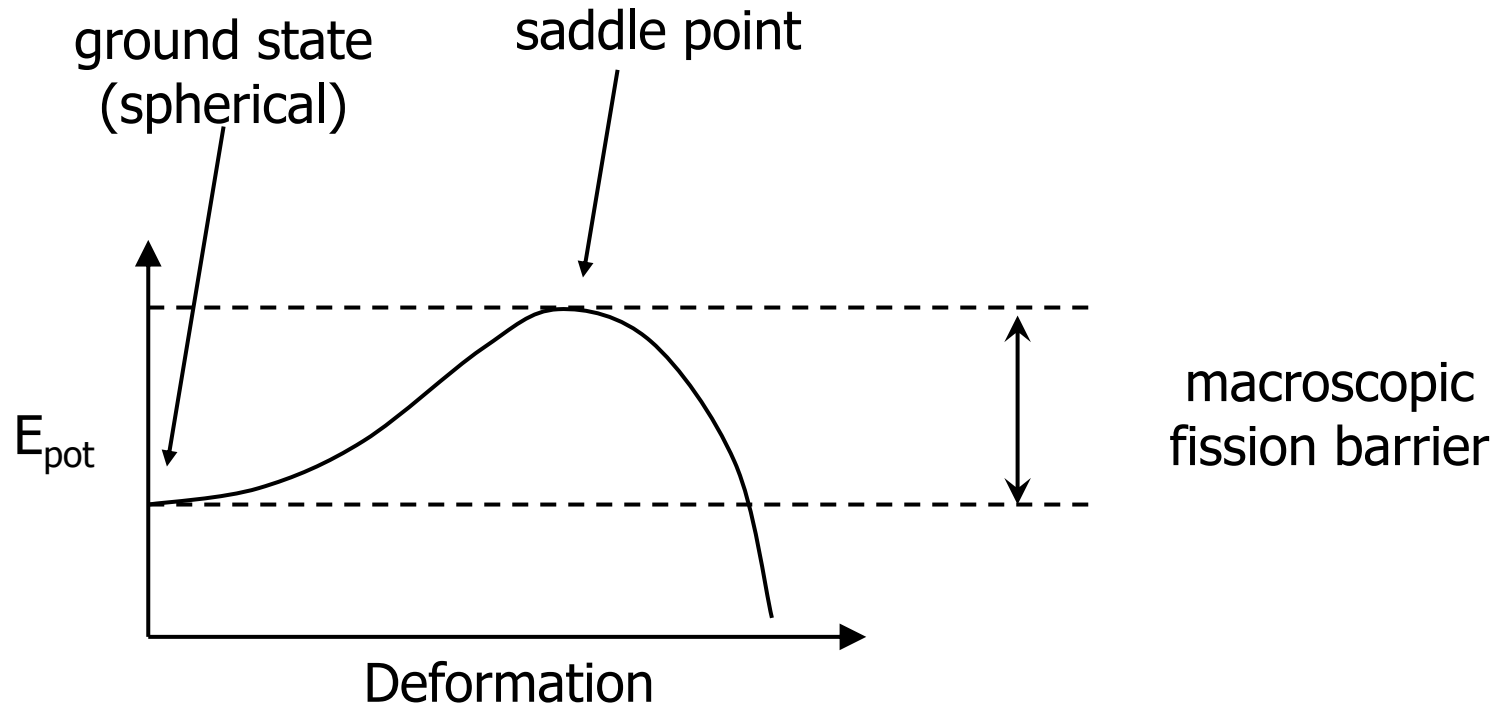
# Superheavy Elements – Present Status and Key Questions

# Nuclear Chart



- $\approx 3,000$  nuclides known
- $\approx 250$  stable nuclides
- $\approx 7,000$  nuclides predicted to exist

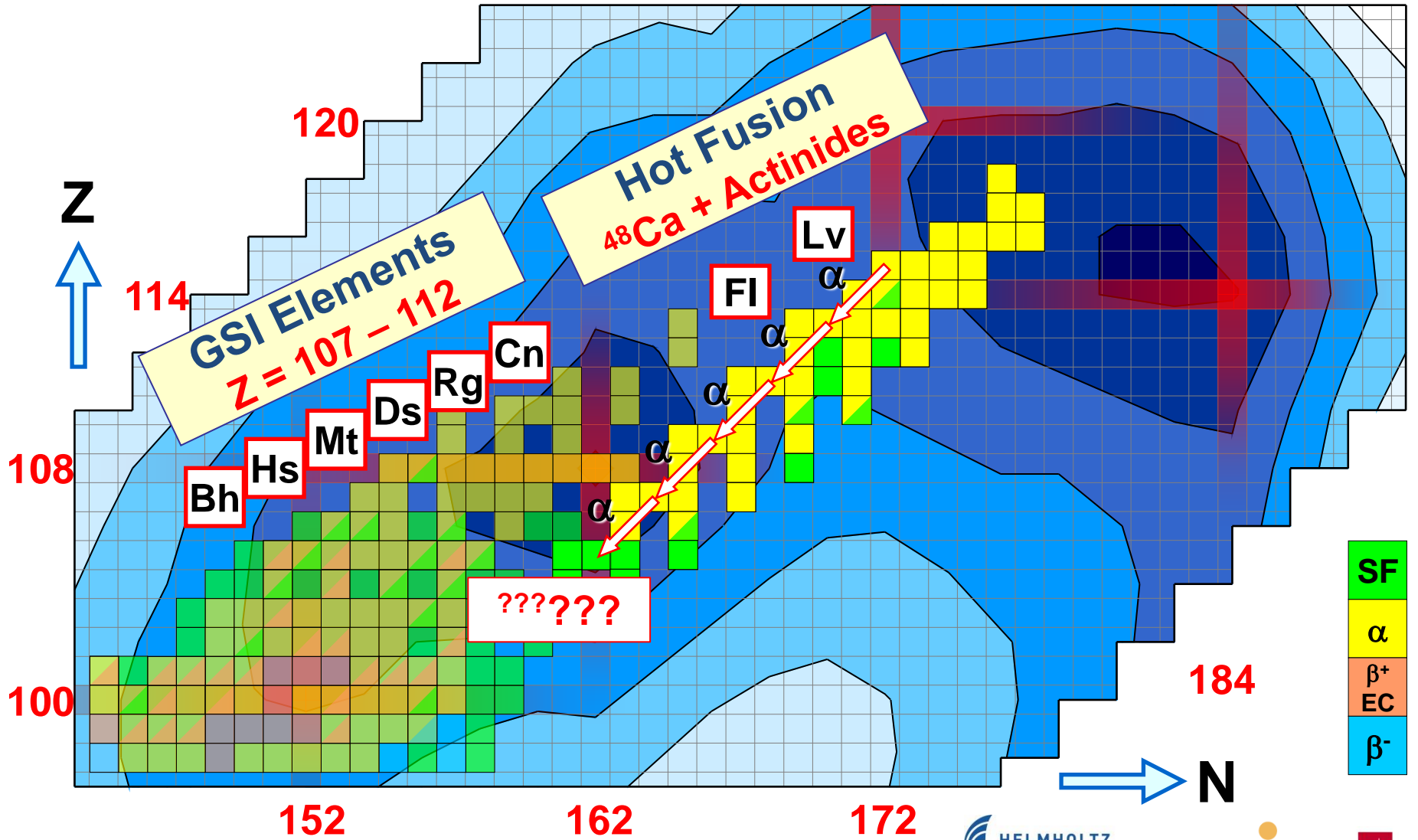
# Superheavy Nuclei (SHN)



- fission barrier in liquid drop model vanishes for  $Z \approx 106$
- stabilization against spontaneous fission by nuclear shell effects

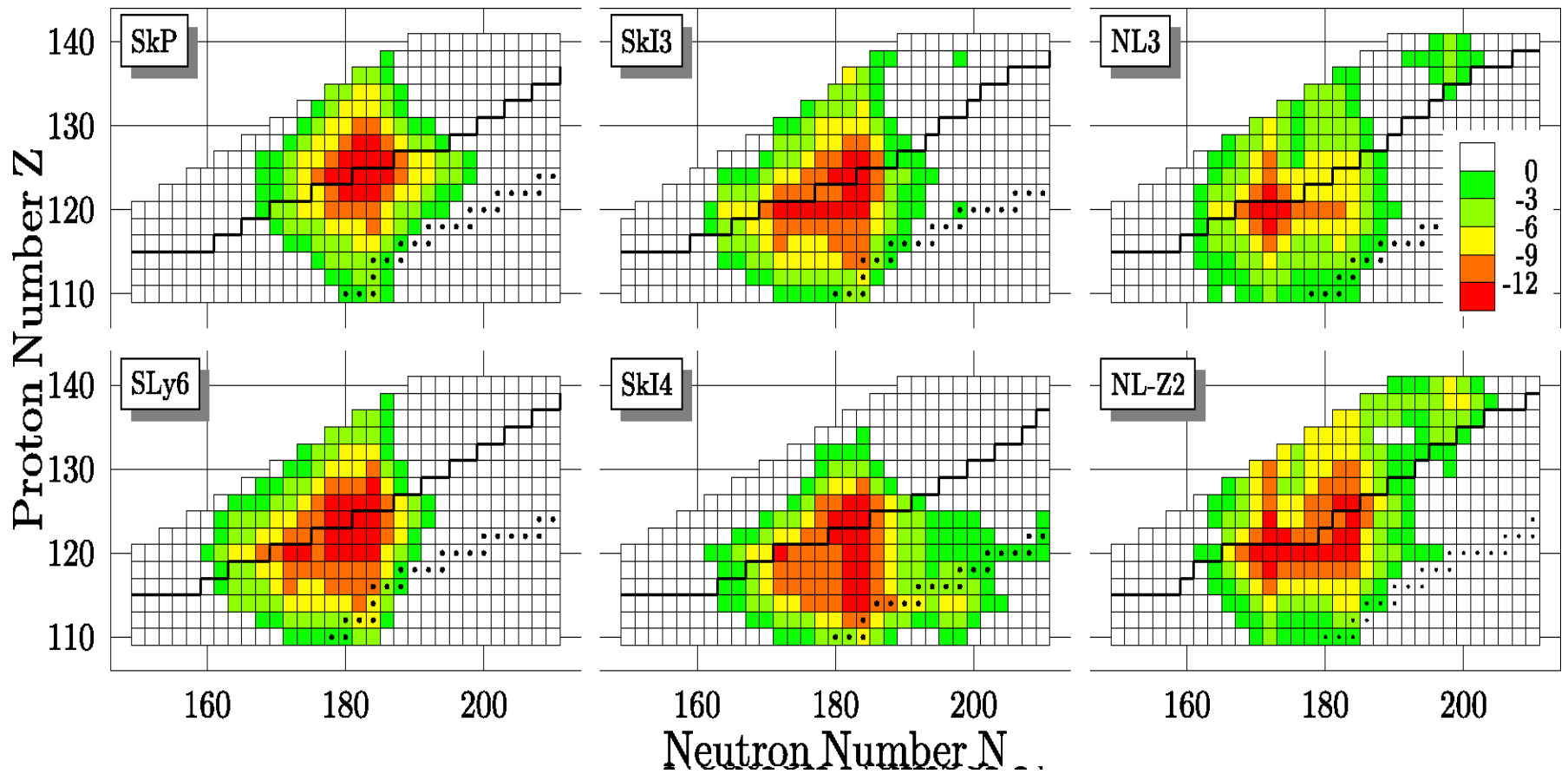
**superheavy nuclei owe their very existence to shell effects**

# Superheavy Nuclides – Current Landscape

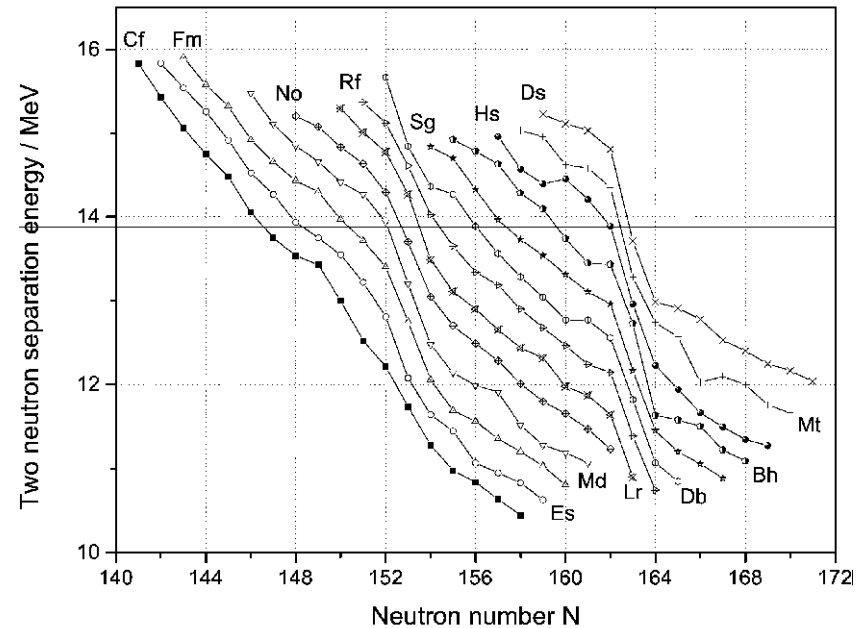
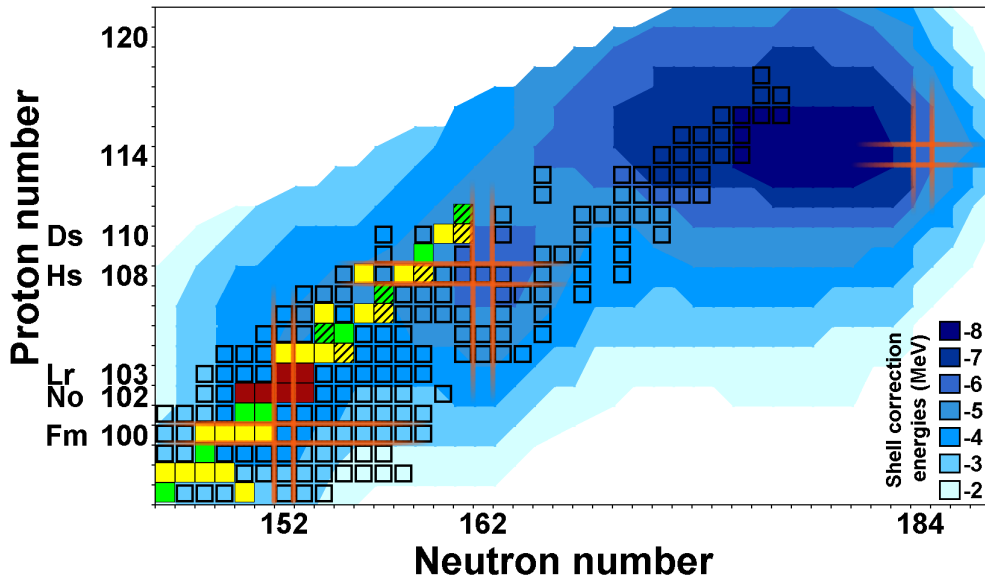




# Nuclear Shells: Magic Numbers in SHE?



# Importance of Masses for $Z > 100$



high-precision mass measurements provide

- accurate absolute binding energies to map nuclear shell effects
- anchor points to fix decay chains

➔ Studies the nuclear structure evolution

➔ Benchmark theoretical nuclear models

$$= N \cdot \text{green circle} + Z \cdot \text{blue circle} + Z \cdot \text{red circle} - \text{binding energy}$$

# Atomic Physics Studies of the Heaviest Elements

1																	18		
1 H	2											13	14	15	16	17	2 He		
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne		
11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar		
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr		
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe		
55 Cs	56 Ba	57+ <sup>*</sup> La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn		
87 Fr	88 Ra	89+ <sup>"</sup> Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs				112 Cn			114 Fl					
								109 Mt	110 Ds	111 Rg			113 Nh			115 Nh	116 Lv	117 Ts	118 Og

chemistry with single atoms

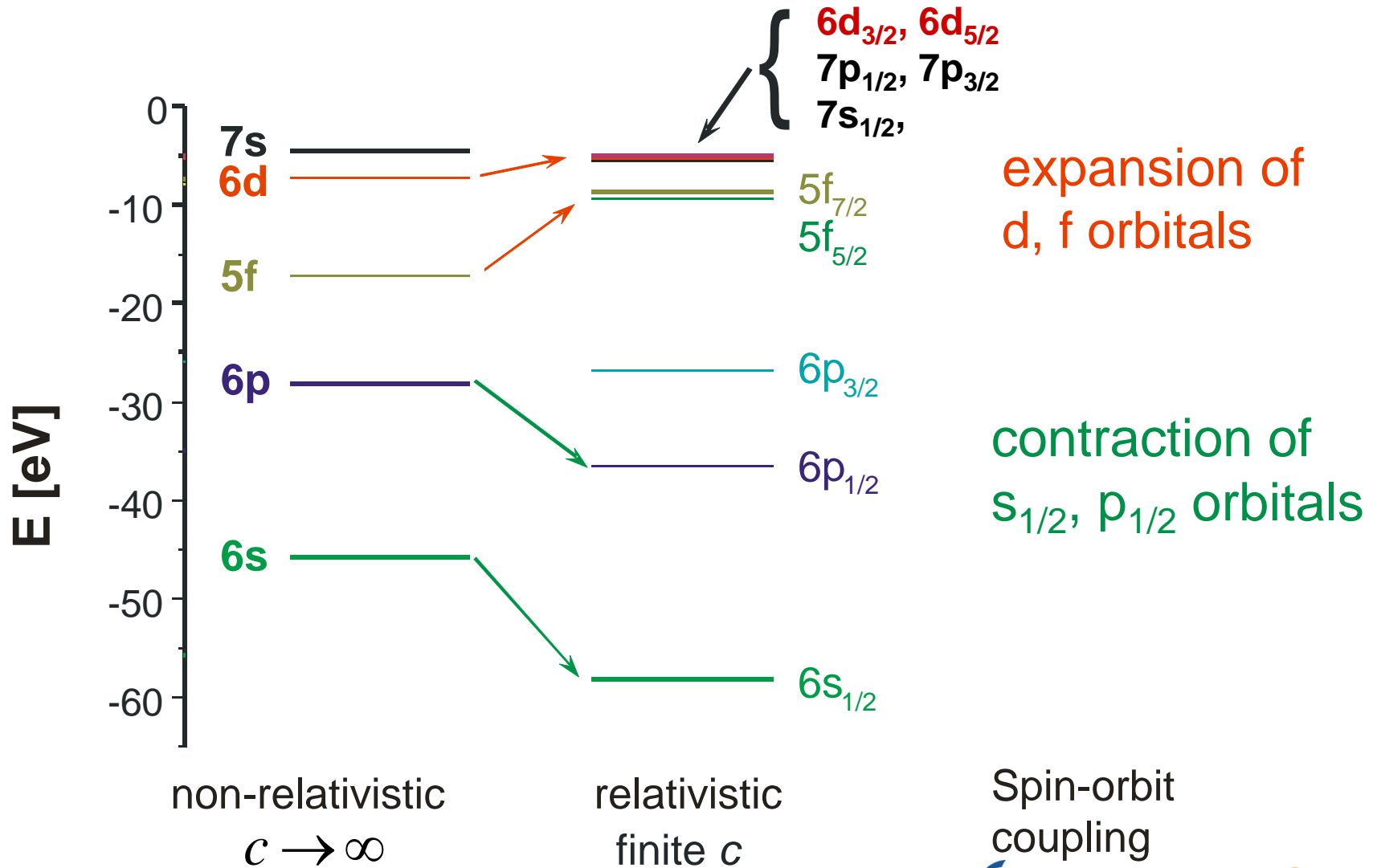
Lanthanides

*	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
"	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Actinides

- study atomic structure and architecture of periodic table
- affected by strong relativistic effects and QED
- benchmark theoretical calculations

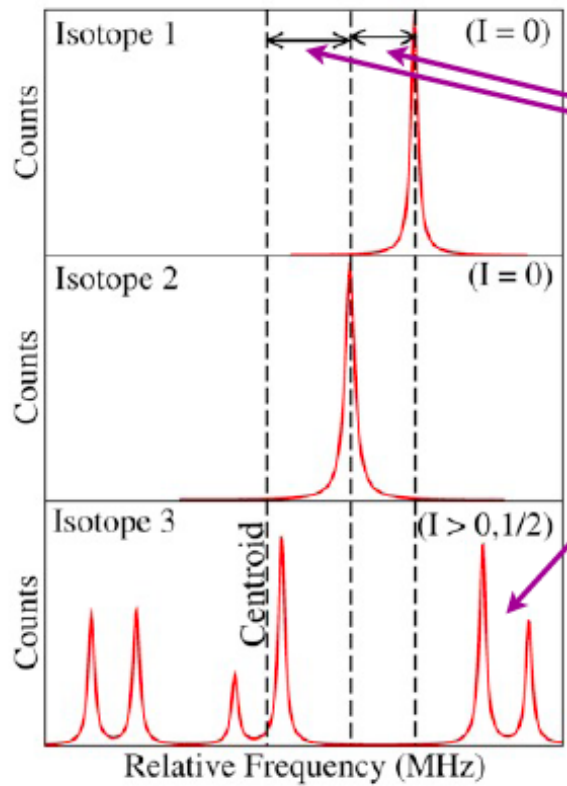
# Relativistic Effects in Uranium



Spin-orbit  
coupling

# Laser Spectroscopy of the Heaviest Elements

<b>Methods:</b>	Search for atomic levels	hyperfine spectroscopy	Measurement of isotopic shifts
<b>Motivation:</b>	<b>relativistic and QED effects</b>	<b>Nuclear moments &amp; spins</b>	<b>changes in mean square charge radii</b>



**Isotope Shifts**  
 $\rightarrow \delta \langle r^2 \rangle$

**Hyperfine Structure**  
 $\rightarrow \mu$   
 $\rightarrow Q_s \rightarrow \langle \beta_2 \rangle$   
 $\rightarrow$  Nuclear spin

# Laser spectroscopy – Status of Measurements

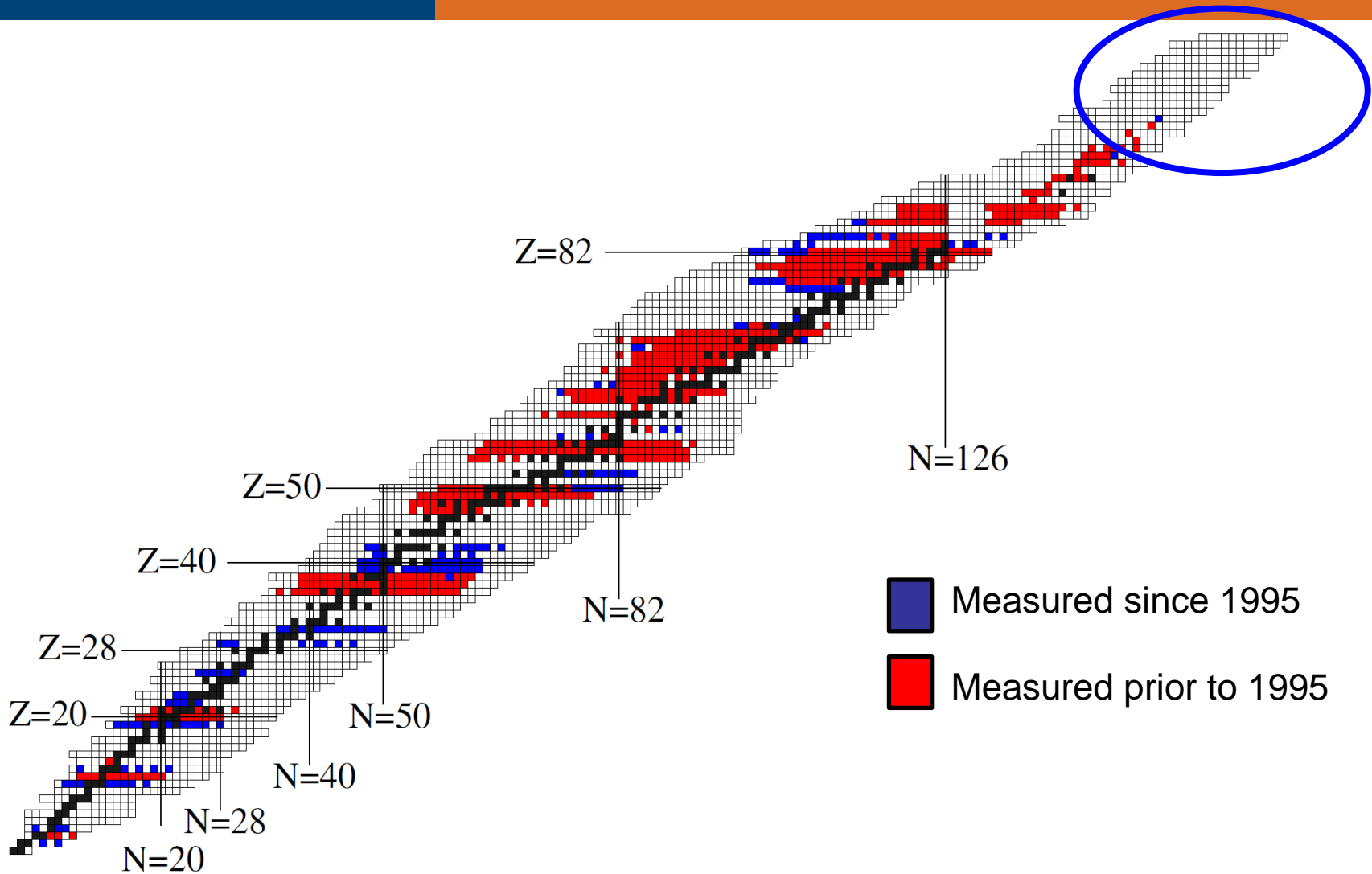


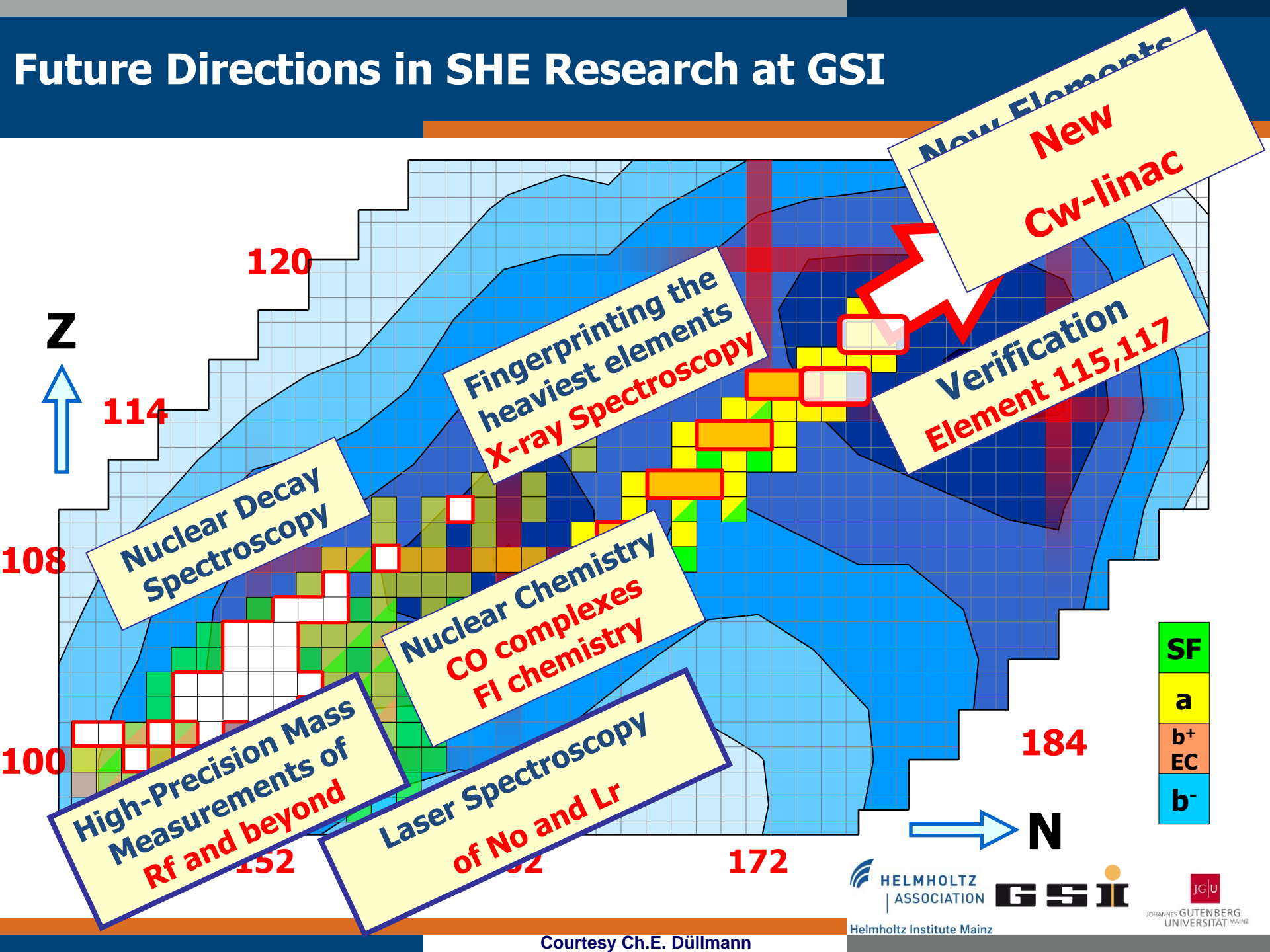
Figure from B. Cheal and K.T. Flanagan, J. Phys. G. **37** (2010) 113101

# Superheavy Elements – Key Questions

- Where is the end of the periodic table in atomic number and mass?
- What is the heaviest element that we can synthesize?
- What are the properties and boundaries of the predicted “island of stability” of superheavy elements?
- What are the details of the fission process and competing decay modes?
- Are there remnants of long-lived superheavy elements on earth?
- How do relativistic effects affect the architecture of the periodic table?

**SHE research at GSI/HIM follows a comprehensive approach investigating atomic, chemical, and nuclear properties of SHE**

# Future Directions in SHE Research at GSI





# Production of the Heaviest Elements

# Requirements – Some Facts and Figures

## Beam intensity:

- present:  $6 \times 10^{12}$  pps ( $1\mu\text{A}_p$ ) for typical beams  $^{48}\text{Ca}$ ,  $^{50}\text{Ti}$ , ...
- future:  $\geq 6 \times 10^{13}$  pps ( $10\mu\text{A}_p$ ) feasible
  - need for high-power targets

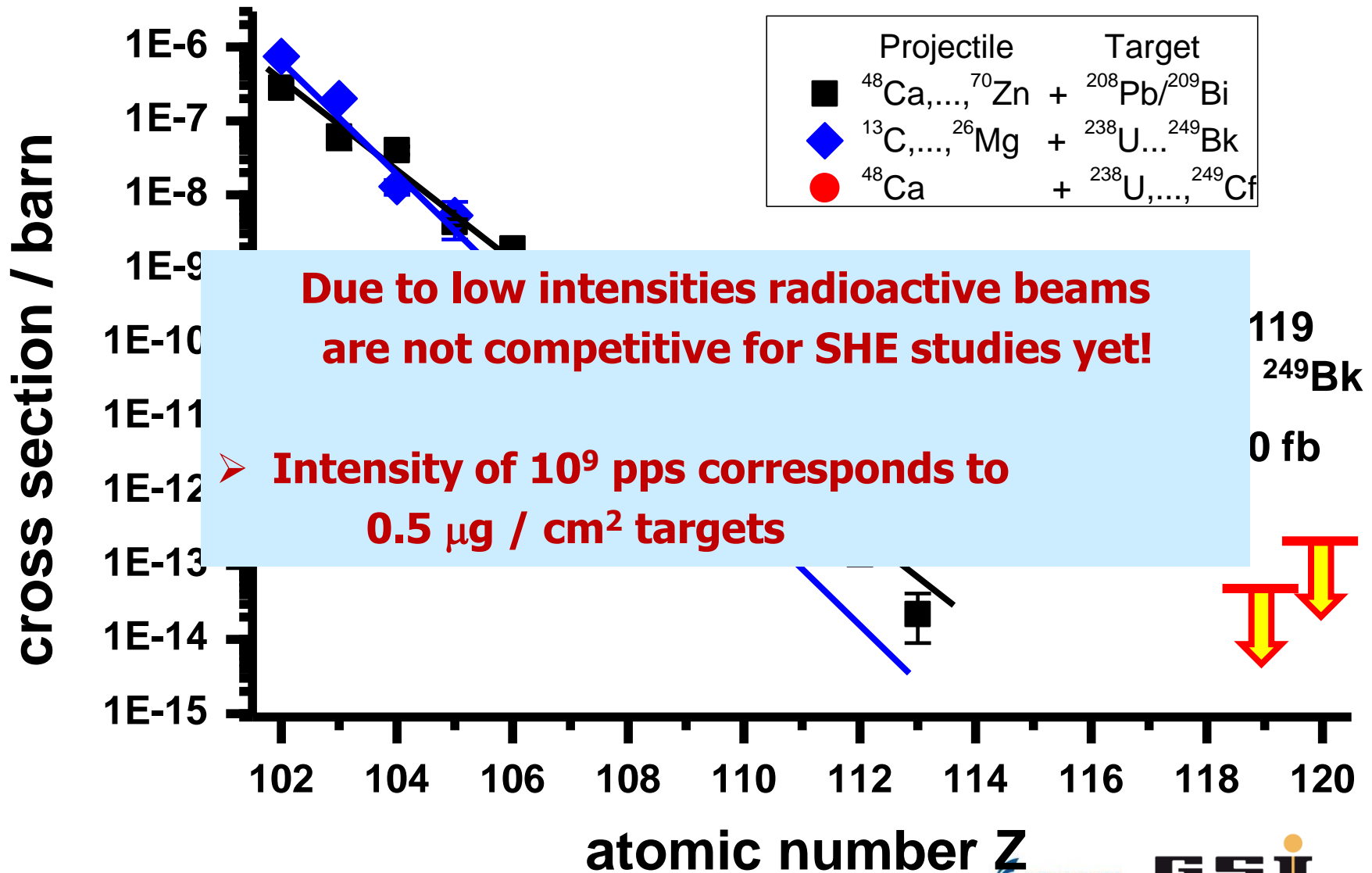
## Targets:

- 0.5-1.0 mg/cm<sup>2</sup> thickness
- about 10 mg of material needed for typical target wheel geometries
  - limited availability of actinide material

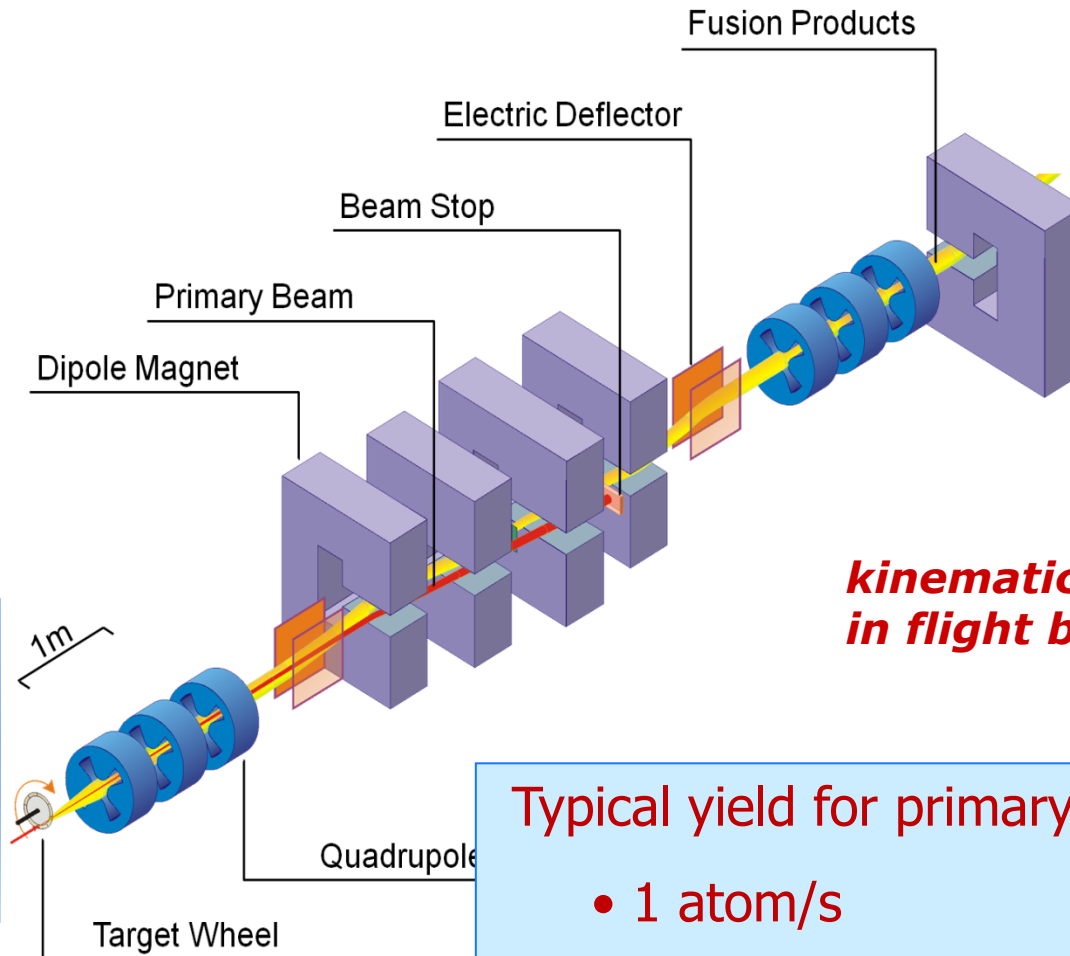
## Recoil separator

- High transmission, short separation time
- low background (beam suppression, low n,  $\gamma$  background)

# Cross Sections for SHE Production



# Synthesis and Separation by SHIP



***kinematic separation  
in flight by velocity filter***

Typical yield for primary beam  $\approx 6 \times 10^{12} / s$

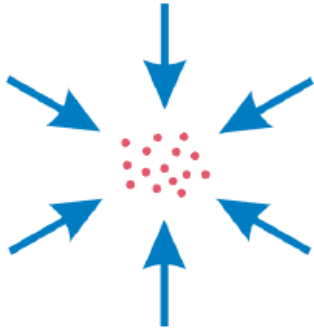
- 1 atom/s @  $Z \approx 102$  ( $\sigma \approx 1 \mu b$ )
- 1 atom/week @  $Z = 112$  ( $\sigma \approx 1 pb$ )

# Basics of Penning Trap Mass Spectrometry

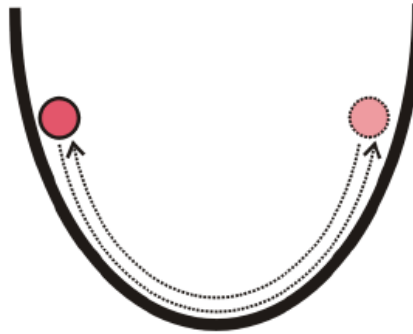
# Basic Idea of a Particle Trap

restoring force 3D

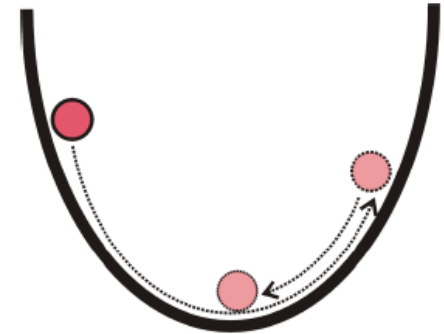
$$\vec{F} \propto -\vec{r}$$



harmonic oscillation



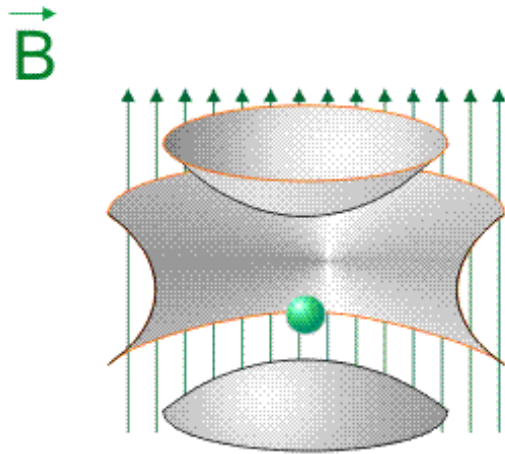
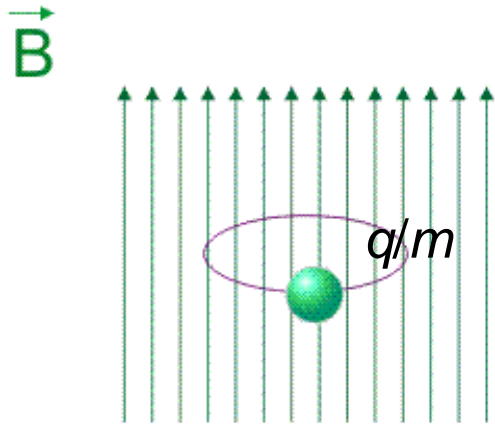
reducing the kinetic energy by cooling



- confine single particles (nearly) at rest
- minimize perturbations (collisions, field imperfections, ...)
- long observation / measurement times

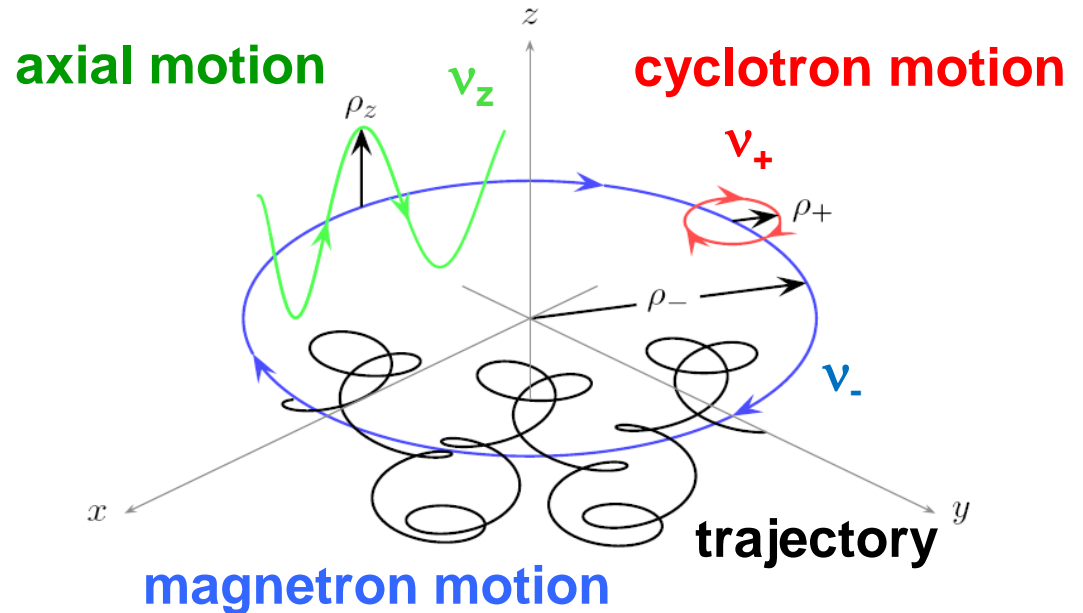
# Principle of Penning Traps

## PENNING trap

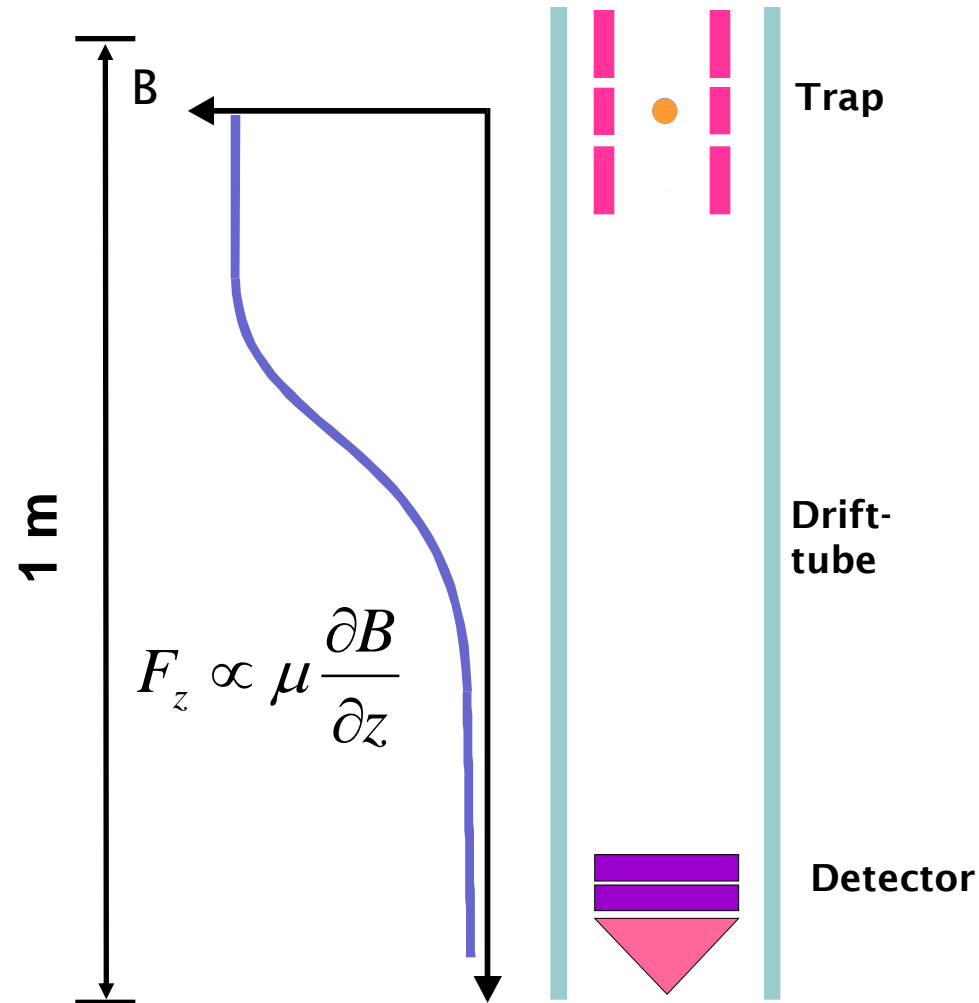
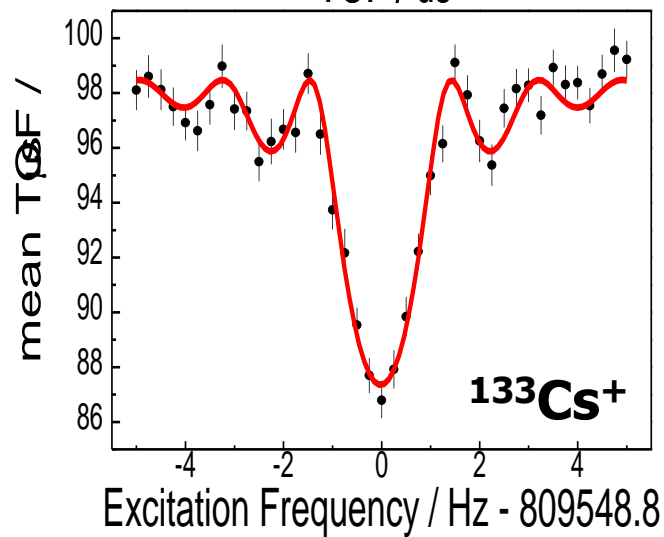
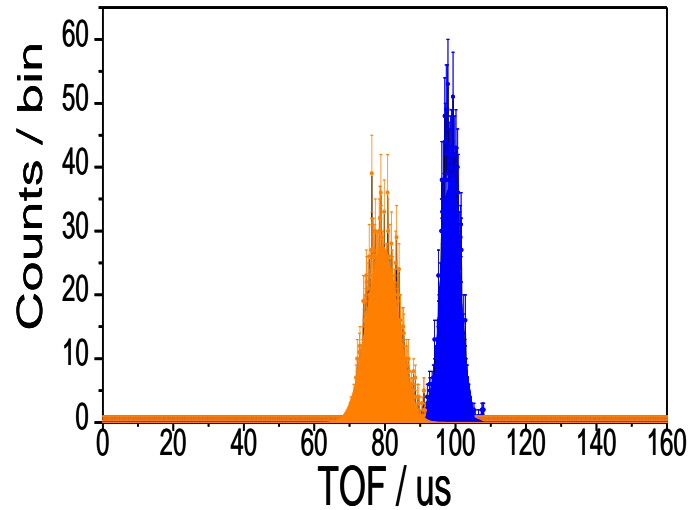


- Strong homogeneous magnetic field
- Weak electric 3D quadrupolar field

**Cyclotron frequency:** 
$$f_c = \frac{1}{2\pi} \cdot \frac{q}{m} \cdot B$$



# Cyclotron frequency measurement



Time-of-flight resonance technique

M. König et al., Int. J. Mass Spec. Ion Process. 142 (1995) 95



# Penning Trap Mass Spectrometry

determine mass via cyclotron frequency measurement

$$\nu_c = \frac{1}{2\pi} \frac{qB}{m}$$

magnetic field calibration

$$\nu_{ref} = \frac{1}{2\pi} \frac{q_{ref} \cdot B}{m_{ref}}$$



$$r = \frac{\nu_{Ref}}{\nu_c} = \frac{m}{m_{ref}}$$

⇒ atomic mass

$$m = \frac{q}{q_{ref}} \left( m_{ref} - q_{ref} \cdot m_e \right) \frac{\nu_{ref}}{\nu_c} + q \cdot m_e$$

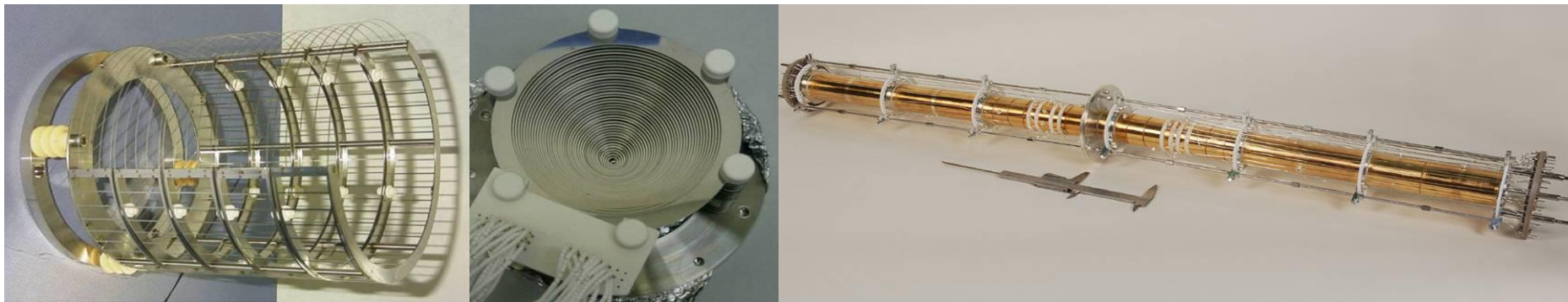
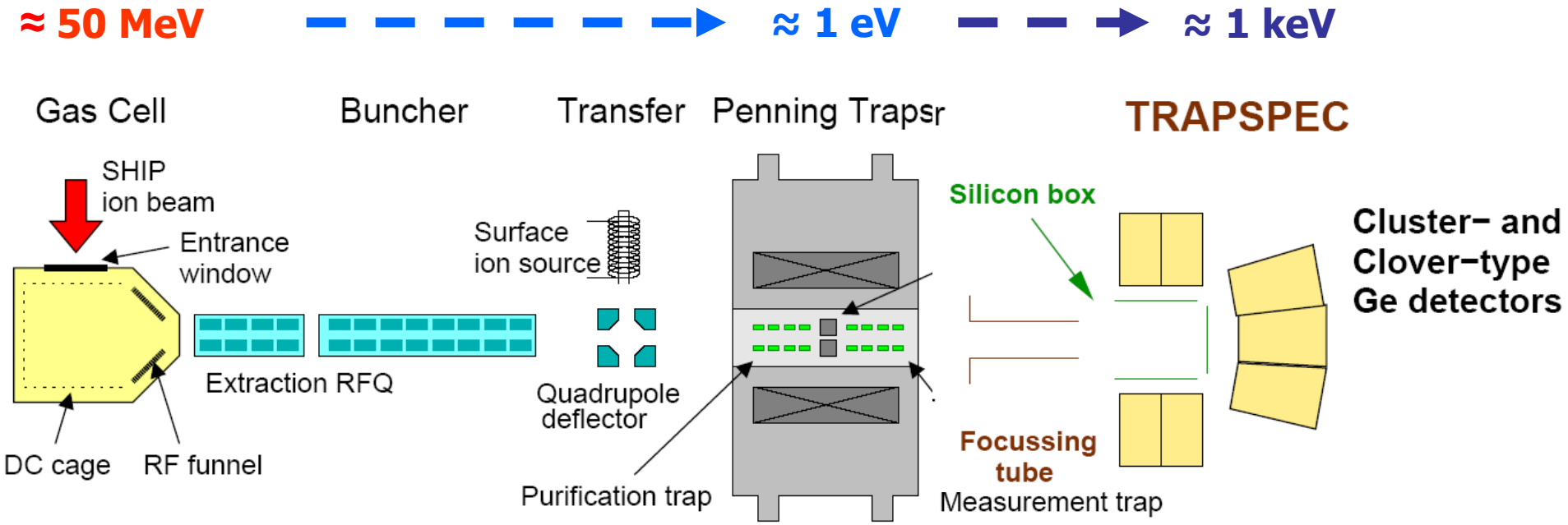
statistical uncertainty

$$s(\nu) \propto \frac{1}{\nu_c} \frac{1}{T_{RF} \sqrt{N_{tot}}}$$

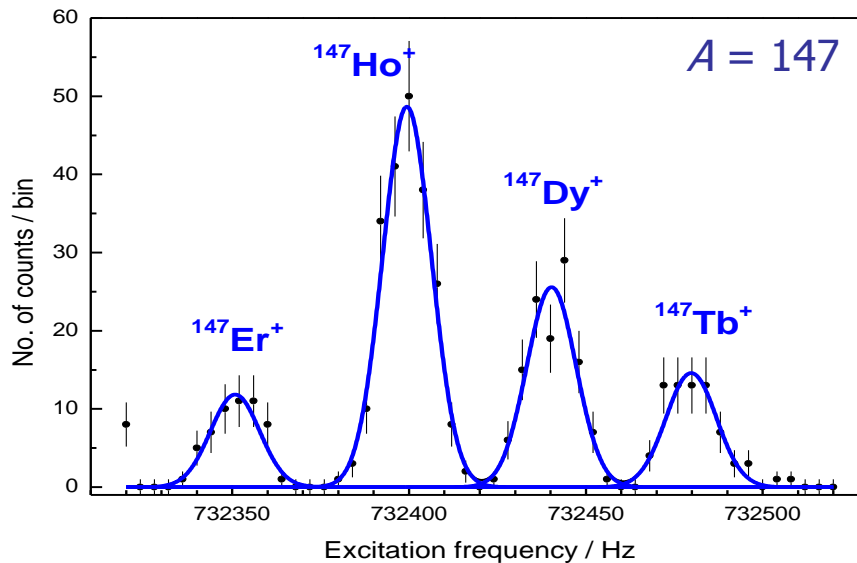
$T_{RF}$  observation time  
 $N_{tot}$  number of det. ions

# Direct Mass Measurements of Nobelium and Lawrencium Isotopes with SHIPTRAP

# SHIPTRAP Setup

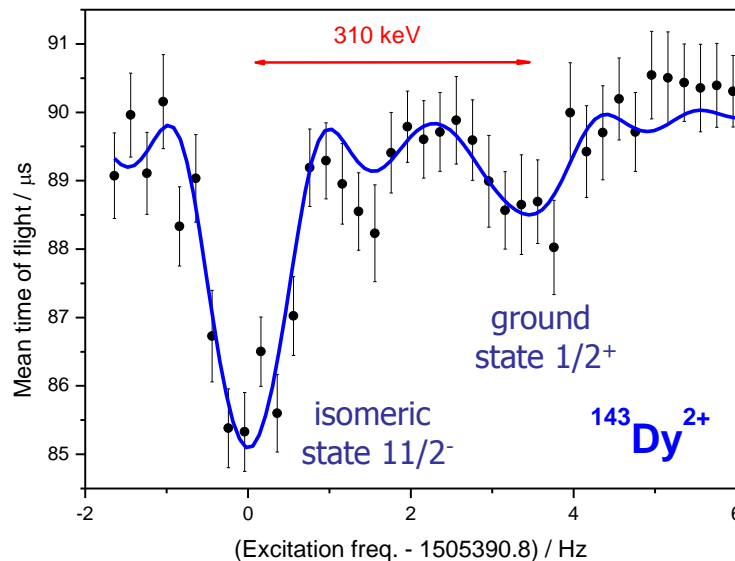


# SHIPTRAP Performance



Mass resolving power of  
 $m/\delta m \approx 100,000$   
in purification trap:

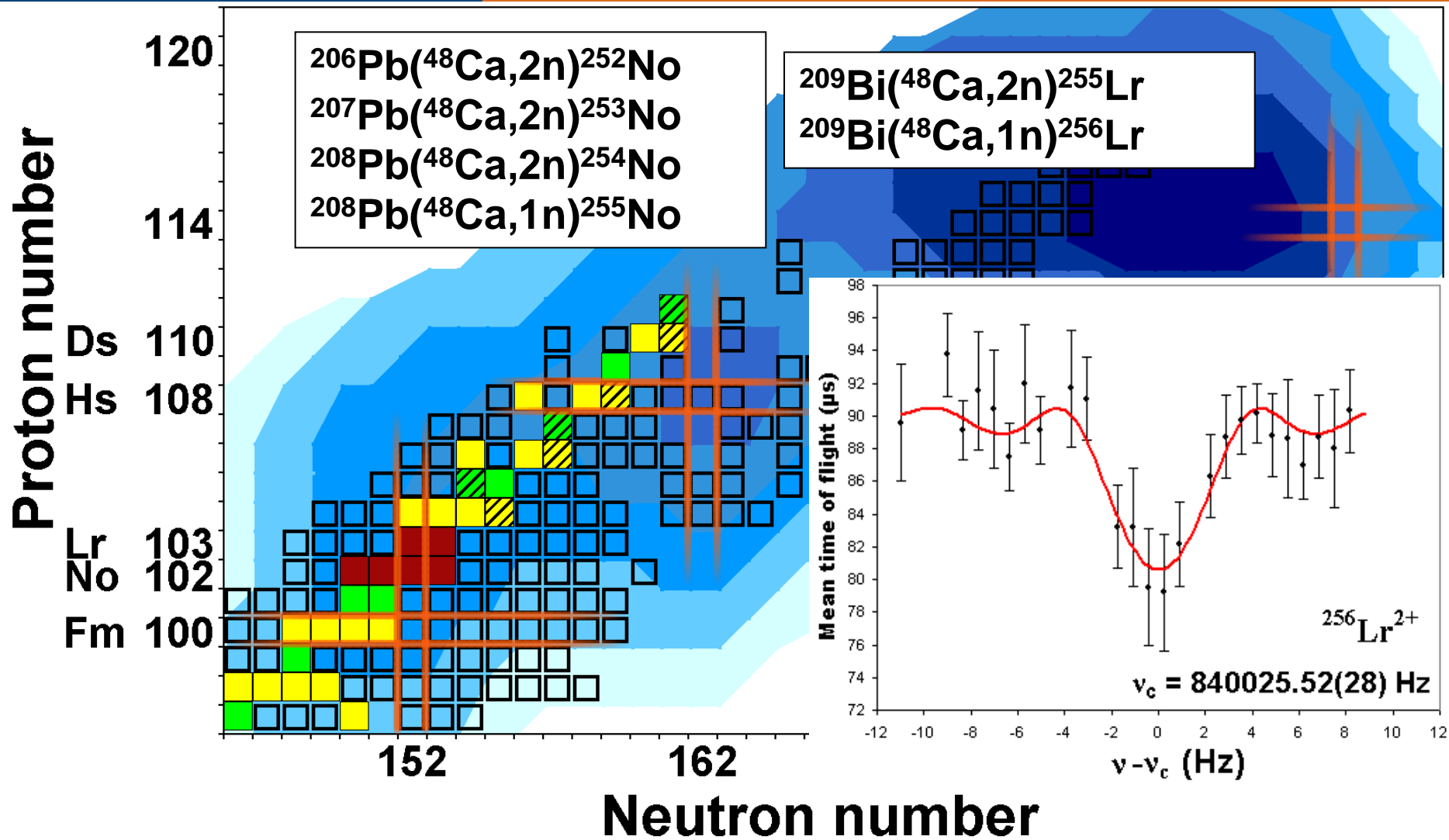
⇒ separation of isobars



Mass resolving power of  
 $m/\delta m \approx 1,000,000$   
in measurement trap:

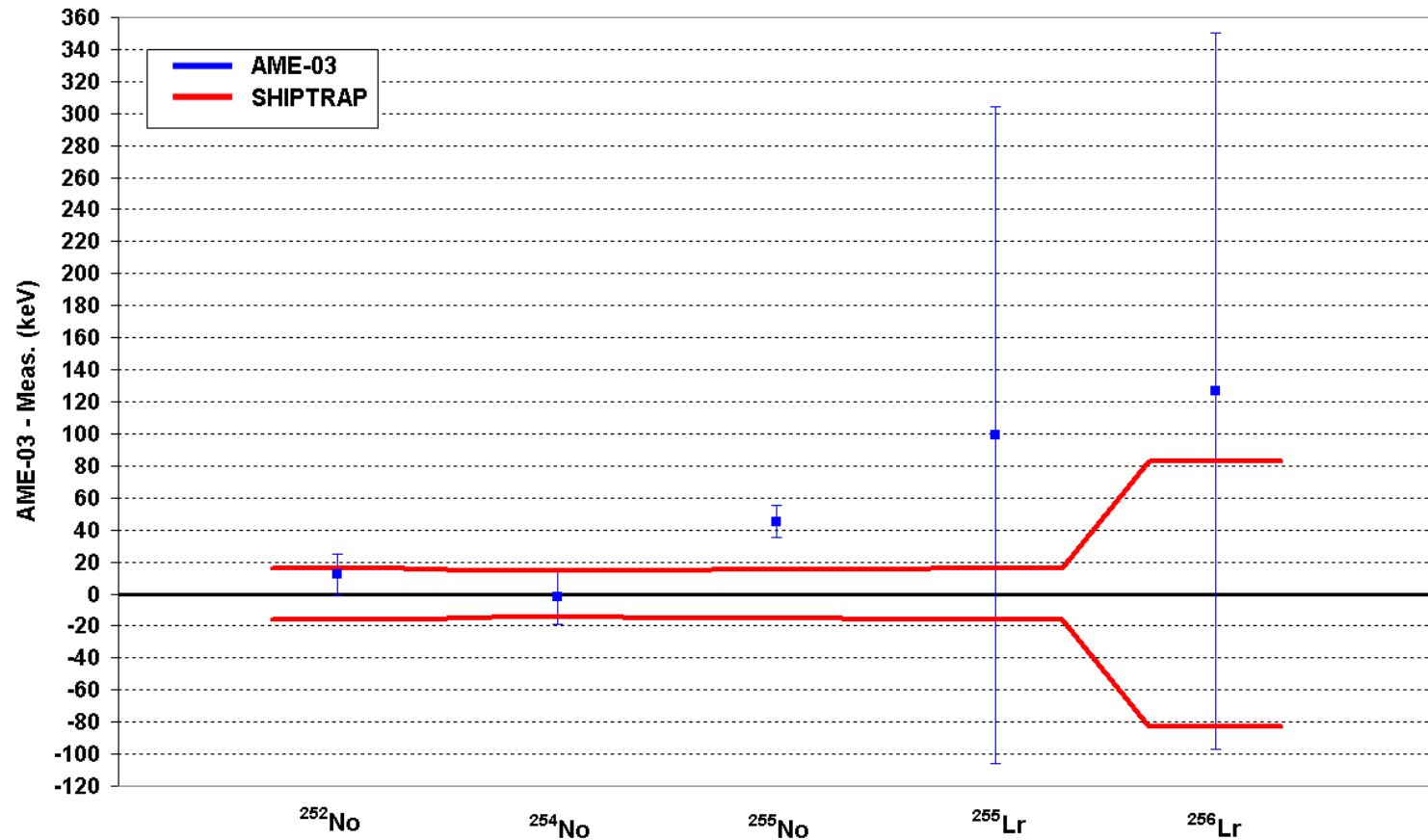
⇒ separation of isomers

# Direct mass measurements with SHIPTRAP



M. Block et al., Nature 463, 785 (2010), M. Dworschak et al., Phys. Rev. C 81, 064312 (2010)  
 E. Minaya Ramirez et al., Science 337, 1183 (2012)

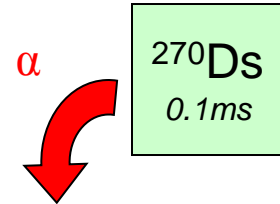
# SHIPTRAP Results vs. Atomic Mass Evaluation



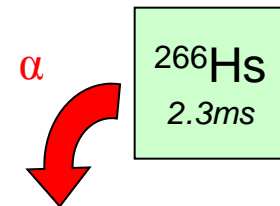
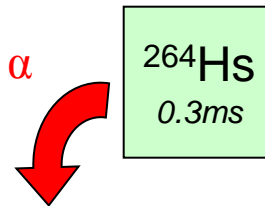
# Pinning Down $\alpha$ -Decay Chains

Z = 110

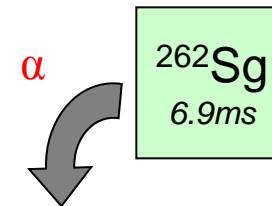
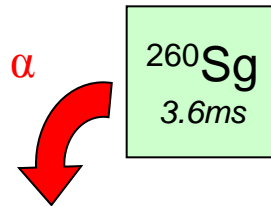
$^{270}\text{Ds}$  mass can be fixed with  
about 40 keV uncertainty now



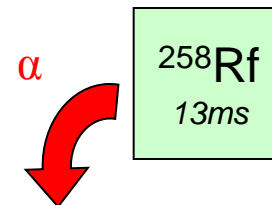
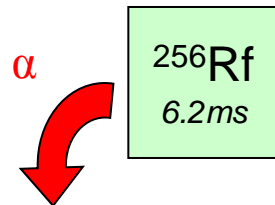
Z = 108



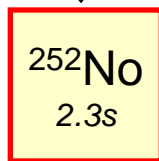
Z = 106



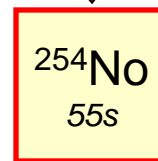
Z = 104



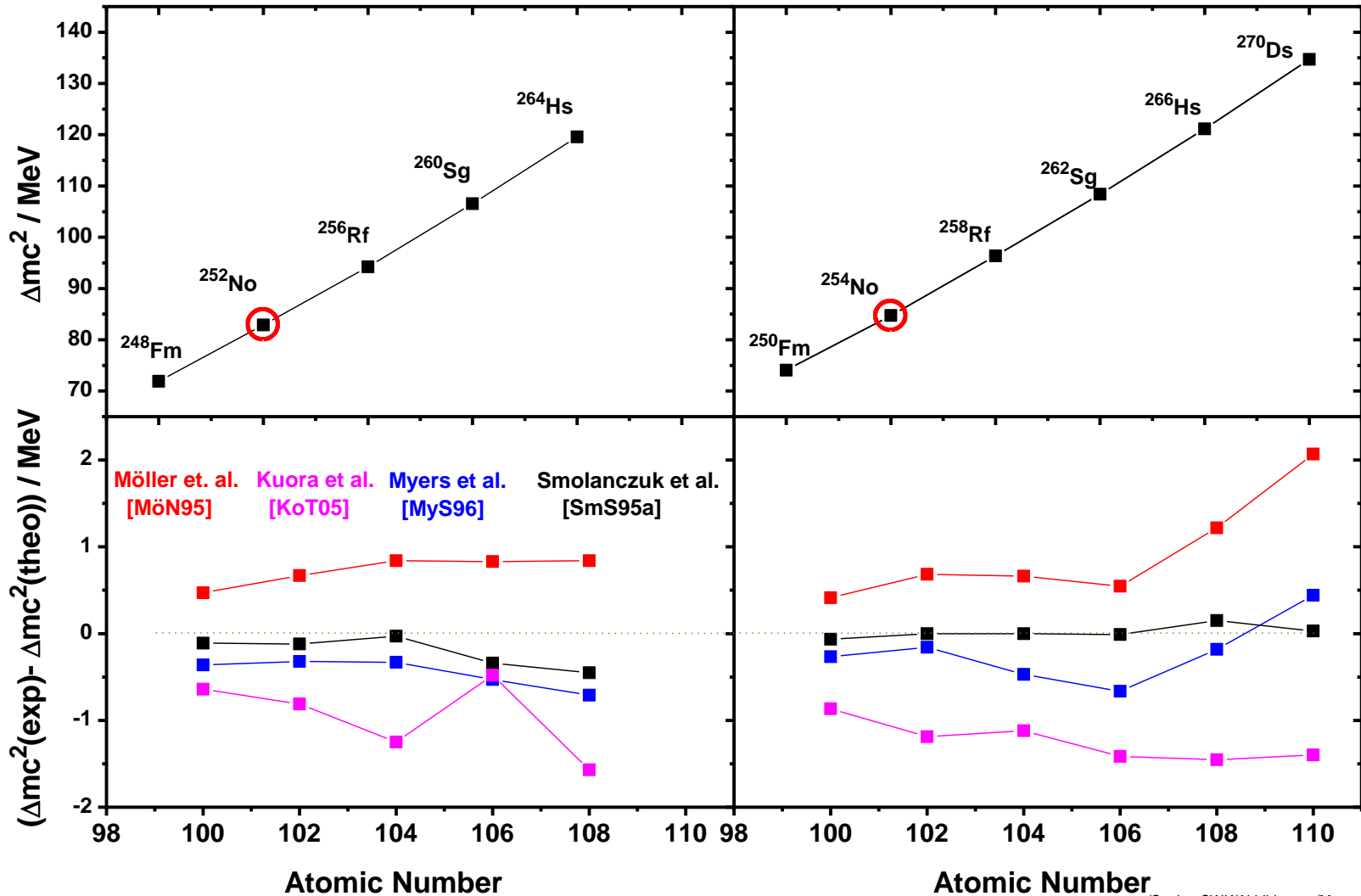
Z = 102



Anchor points



# Masses of even-even $N-Z = 48$ and $N-Z = 50$ Nuclei



/StrukturSWK/Abbildungen/Massen,  
F.P. Heßberger, 3.9.2013



Helmholtz Institute Mainz



courtesy F. P. Hessberger

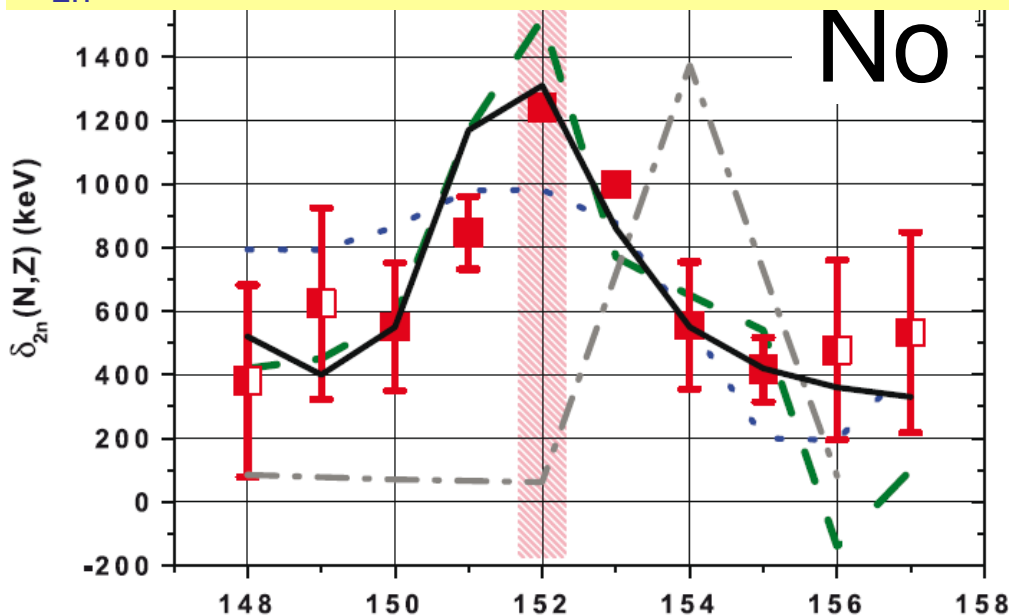


# SHIPTRAP: Probing the Strength of Shell Effects

## Direct Mapping of Nuclear Shell Effects in the Heaviest Elements

E. Minaya Ramirez,<sup>1,2</sup> D. Ackermann,<sup>2</sup> K. Blaum,<sup>3,4</sup> M. Block,<sup>2\*</sup> C. Droese,<sup>5</sup> Ch. E. Düllmann,<sup>6,2,1</sup>  
 M. Dworschak,<sup>2</sup> M. Eibach,<sup>4,6</sup> S. Eliseev,<sup>3</sup> E. Haettner,<sup>2,7</sup> F. Herfurth,<sup>2</sup> F. P. Heßberger,<sup>2,1</sup>  
 S. Hofmann,<sup>2</sup> J. Ketelaer,<sup>3</sup> G. Marx,<sup>5</sup> M. Mazzocco,<sup>8</sup> D. Nesterenko,<sup>9</sup> Yu. N. Novikov,<sup>9</sup> W. R. Plaß,<sup>2,7</sup>  
 D. Rodríguez,<sup>10</sup> C. Scheidenberger,<sup>2,7</sup> L. Schweikhard,<sup>5</sup> P. G. Thirolf,<sup>11</sup> C. Weber<sup>11</sup>

$$\delta_{2n}(N,Z) = 2B(N,Z) - B(N-2,Z) - B(N+2,Z)$$

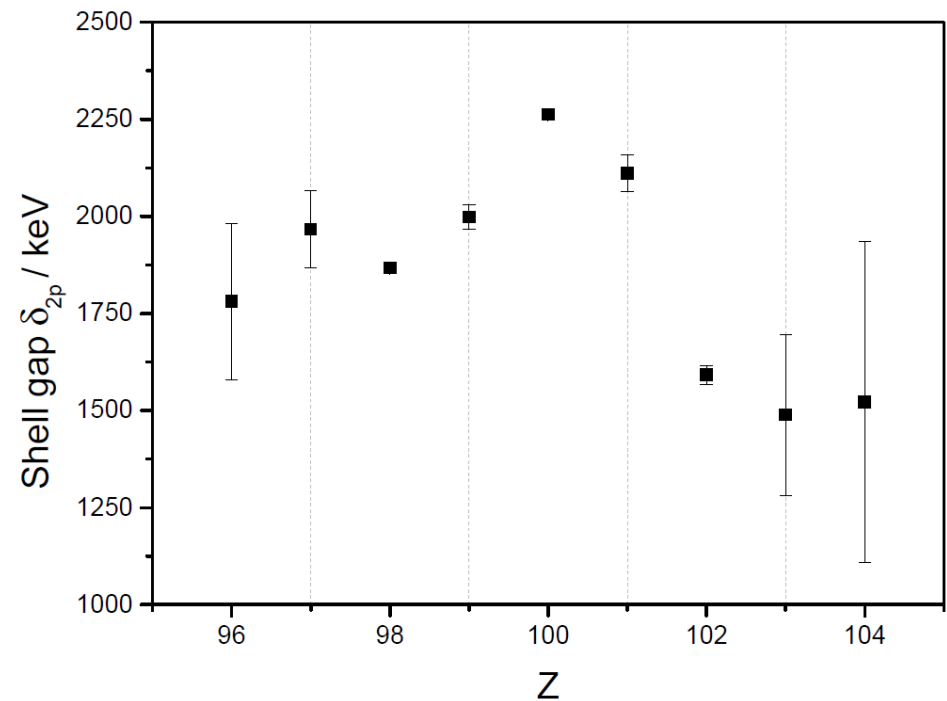
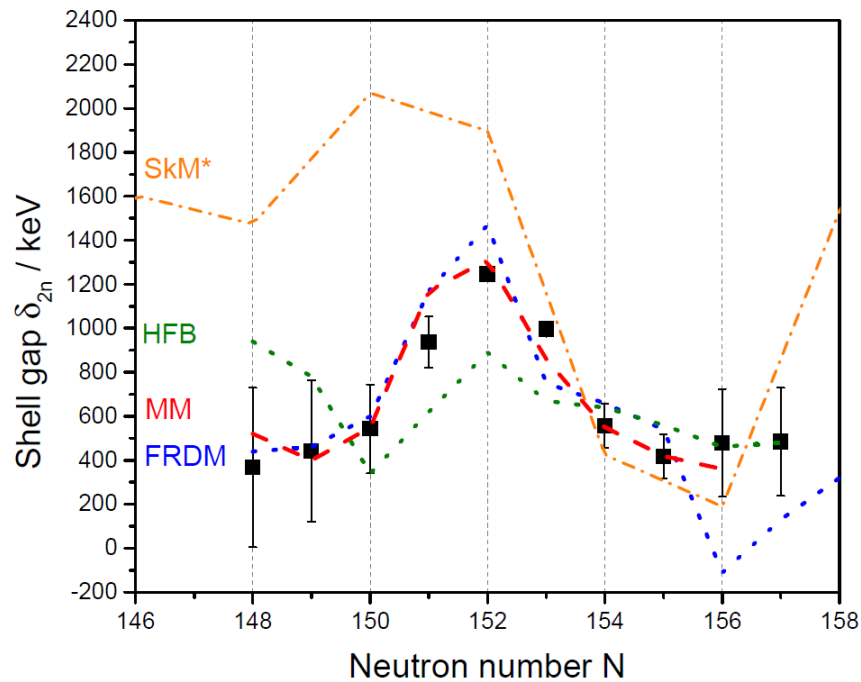


- Experimental
- Muntian (mic-mac)  
Z=114 N=184
- Möller FRDM  
Z=114 N=184
- TW-99  
Z=120 N=172
- SkM\*  
Z=126 N=184

**Science** 337 (2012) 1207

# SHIPTRAP: Probing the Strength of Shell Effects

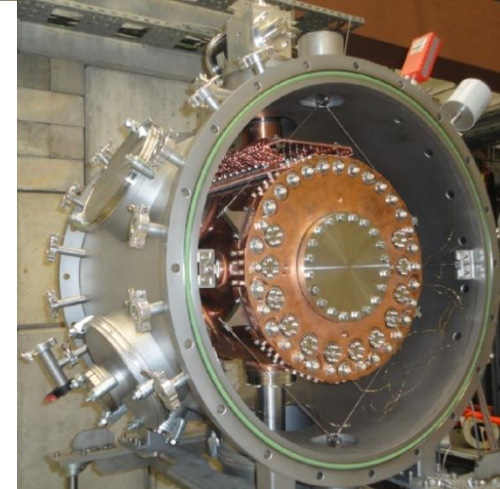
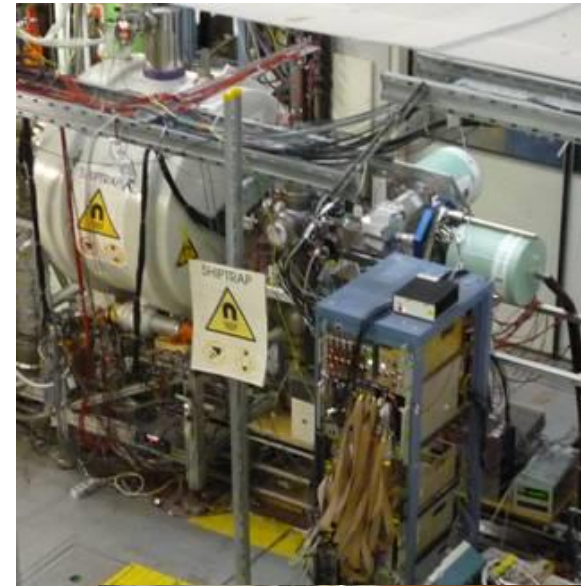
$$\delta_{2n}(N,Z) = 2B(N,Z) - B(N-2,Z) - B(N+2,Z)$$



# Upgrades and Improvements

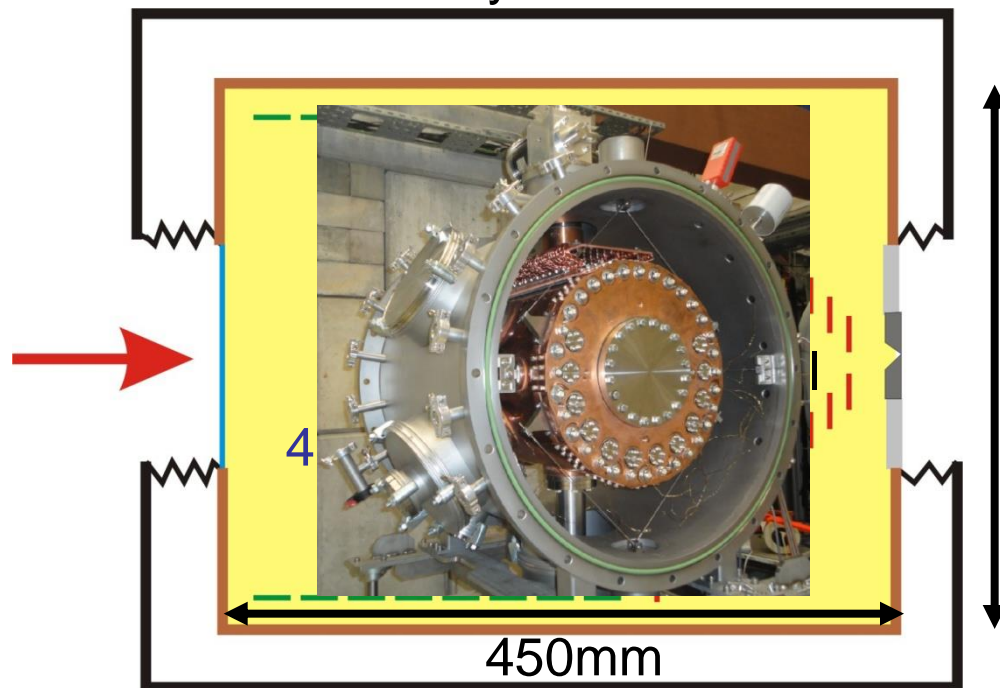
# Upgrades and Combinations

- Novel experiments
  - trap-assisted nuclear decay spectroscopy
  - laser spectroscopy (in gas cell / gas jet)
  - gas-phase chemistry (in gas cell / ion trap)
- Increase efficiency and sensitivity
  - cryogenic gas cell
  - novel measurement schemes (PI-ICR)
  - single-ion mass measurements (FT-ICR)  
(→ TRIGA-TRAP, TRAPSENSOR)



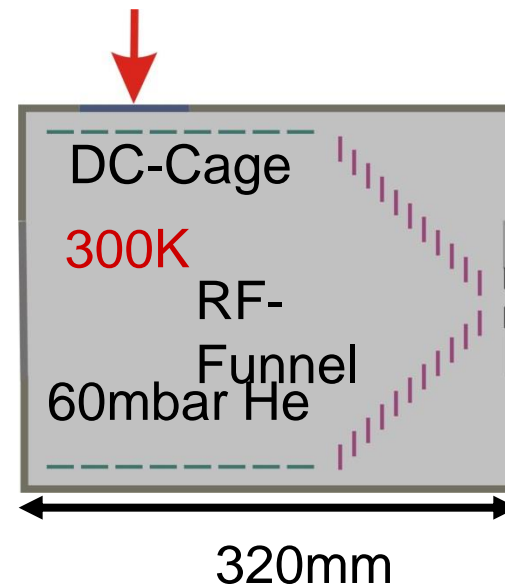
# Cryogenic Gas Stopping Cell

Cryo Cell



Gas Cell

400mm

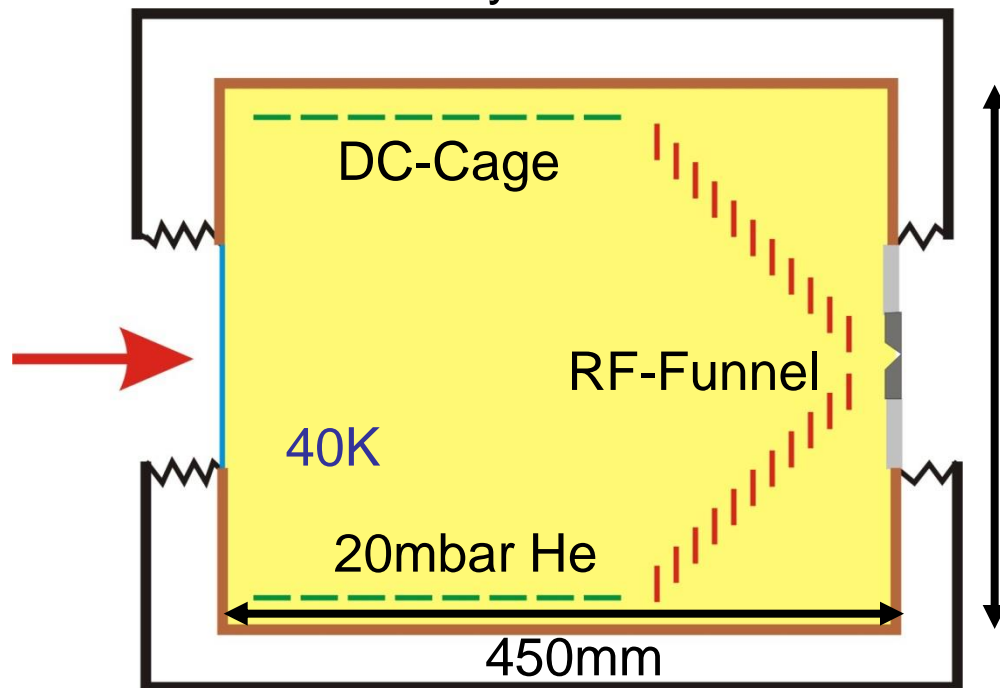


## Advantages compared to 1st generation gas cell:

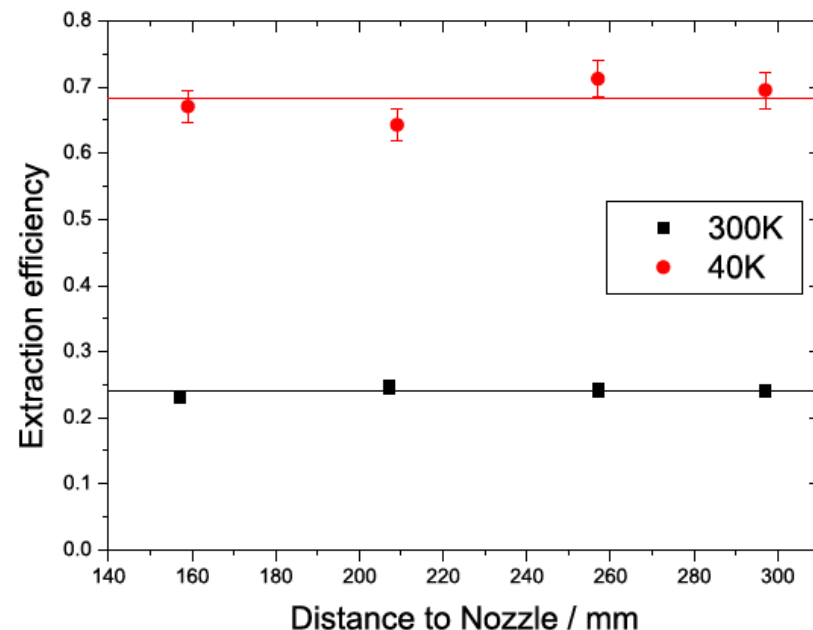
- Larger stopping volume and Coaxial injection of reaction products
- Higher cleanliness due to cryogenic operation
- Larger gas density at a lower absolute pressure

# Cryogenic Gas Stopping Cell

Cryo Cell



Gas Cell



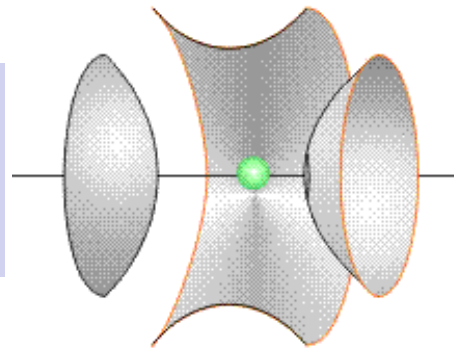
## Advantages compared to 1st generation gas cell:

- Larger stopping volume and Coaxial injection of reaction products
- Higher cleanliness due to cryogenic operation
- Larger gas density at a lower absolute pressure

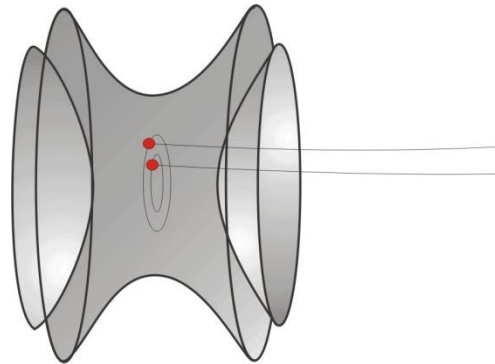
# Phase-Imaging Ion-Cyclotron-Resonance Technique

# Recent Breakthrough

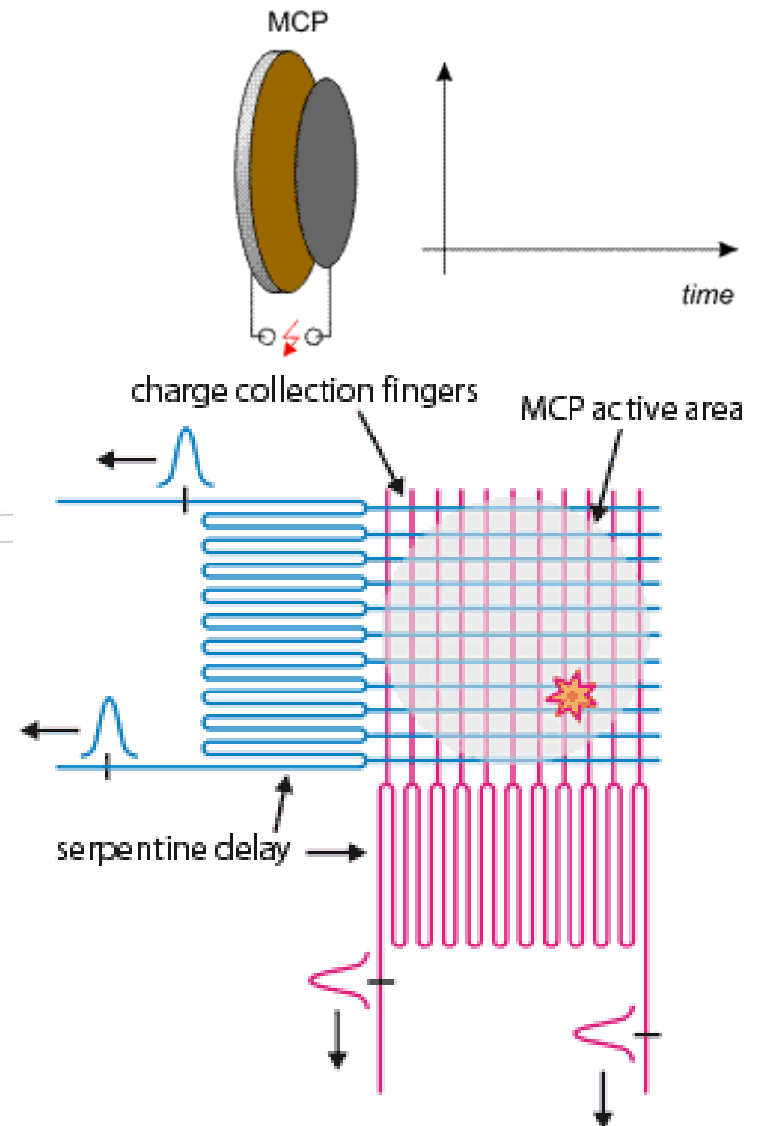
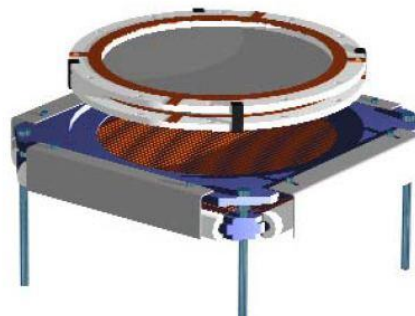
*Destructive  
time-of-flight  
detection*



*Spatially  
resolved  
detection*

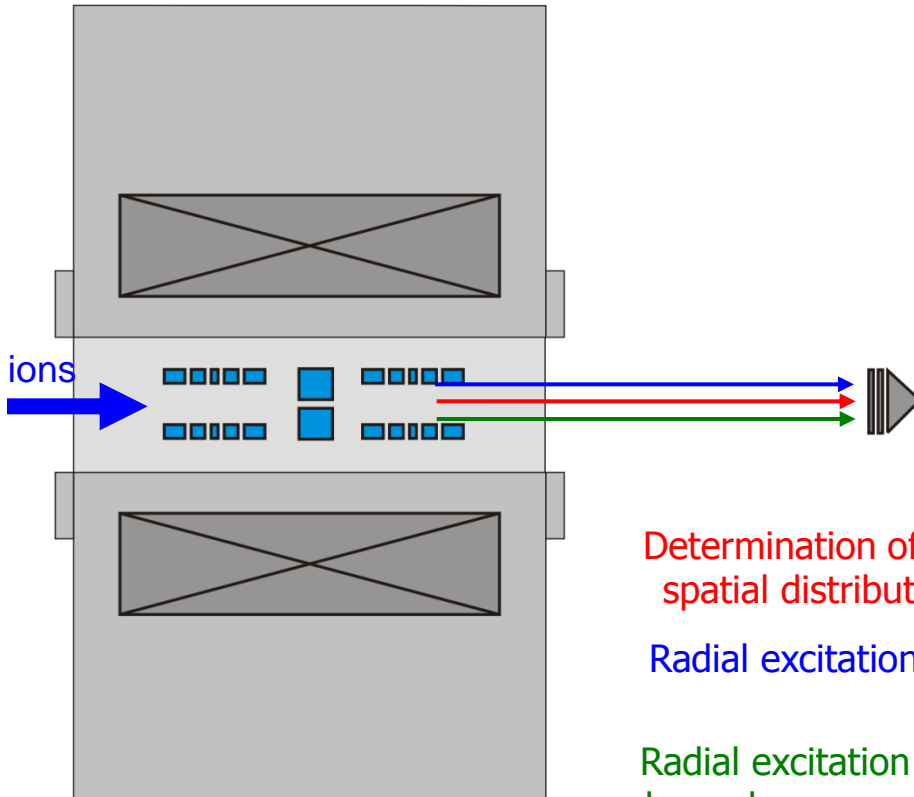


*Delay-line  
detector*

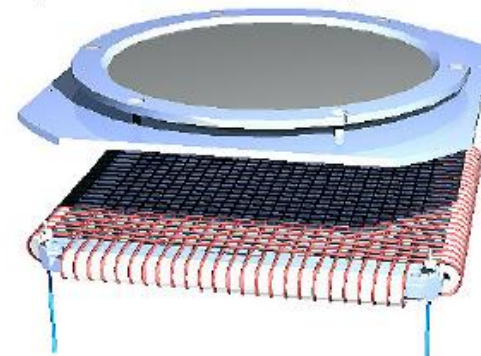




# Phase-Imaging Ion-Cyclotron-Resonance Method



Delay-Line Detector by Roentdek

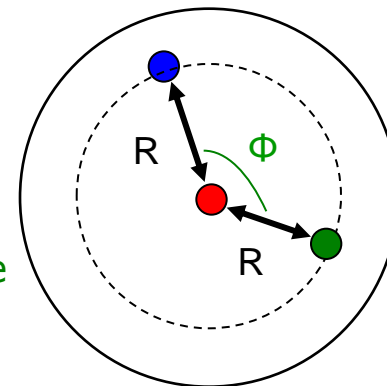


Independent Measurements of Eigenfrequencies  $\nu_+$  and  $\nu_-$

Determination of the spatial distribution

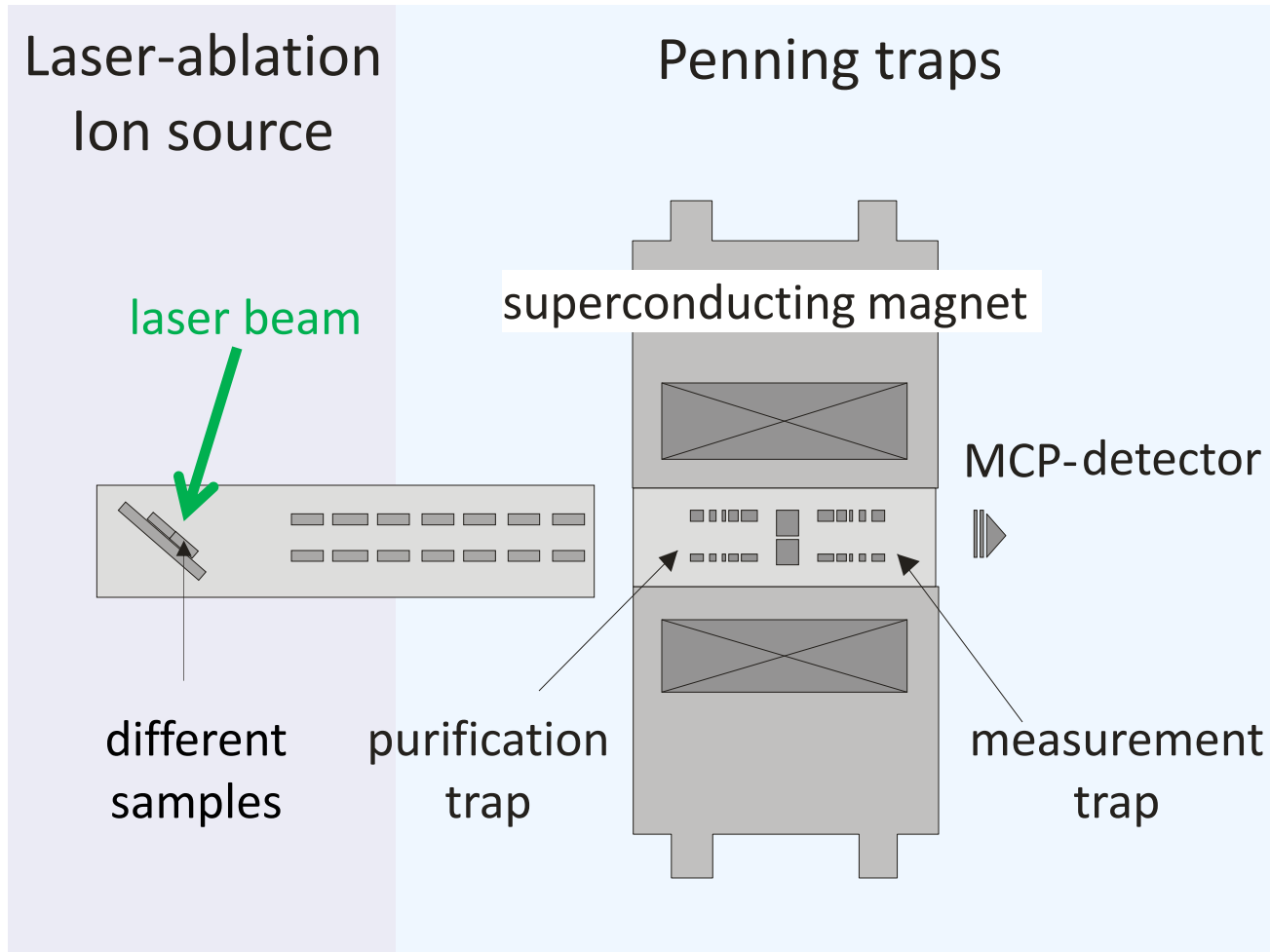
Radial excitation

Radial excitation followed by a phase accumulation time



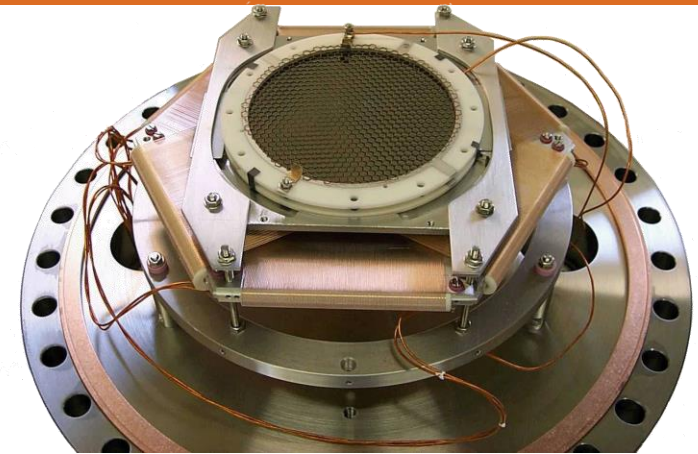
$$\phi + 2\pi n = 2\pi \nu t \quad \Delta \nu = \frac{\Delta \phi}{2\pi} = \frac{\Delta R}{\pi t R}$$

# SHIPTRAP Off-line Setup



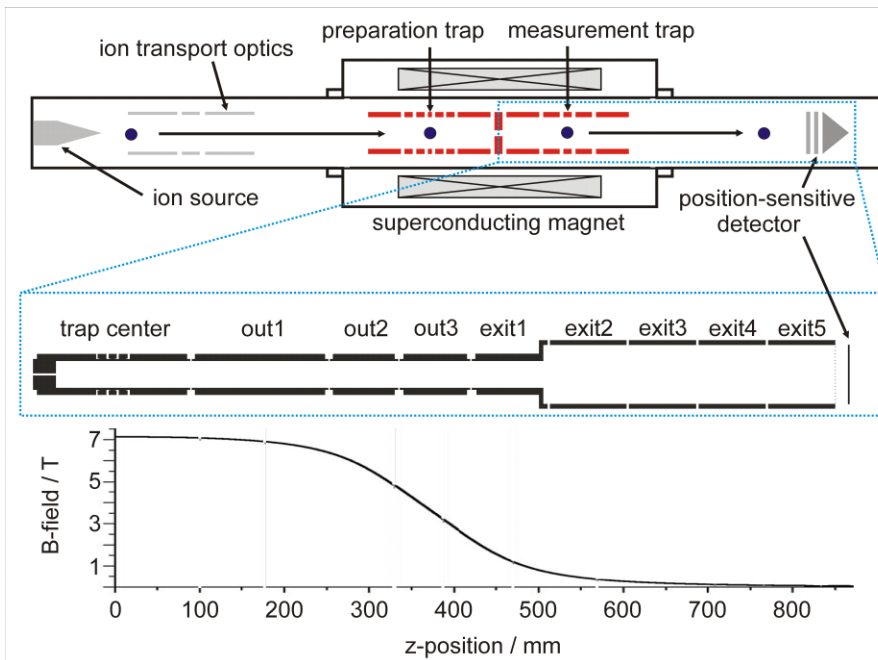
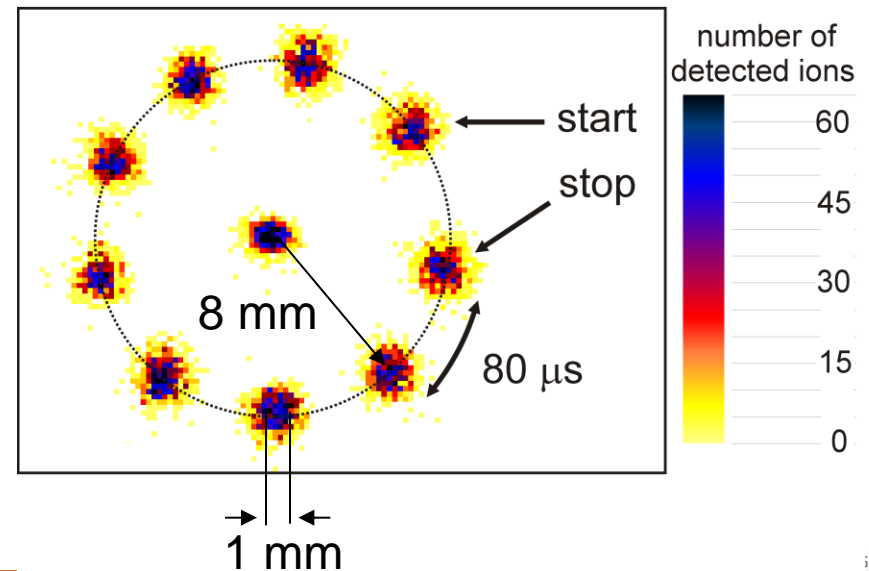
# Position-sensitive Delayline Detector

Active diameter	42 mm
Channel diameter	25 $\mu\text{m}$
Open area ratio	>50 %
<b>Position resolution</b>	<b>70 <math>\mu\text{m}</math></b>
<b>Max. B-field</b>	<b>a few mT</b>
Time resolution	$\sim 10$ ns



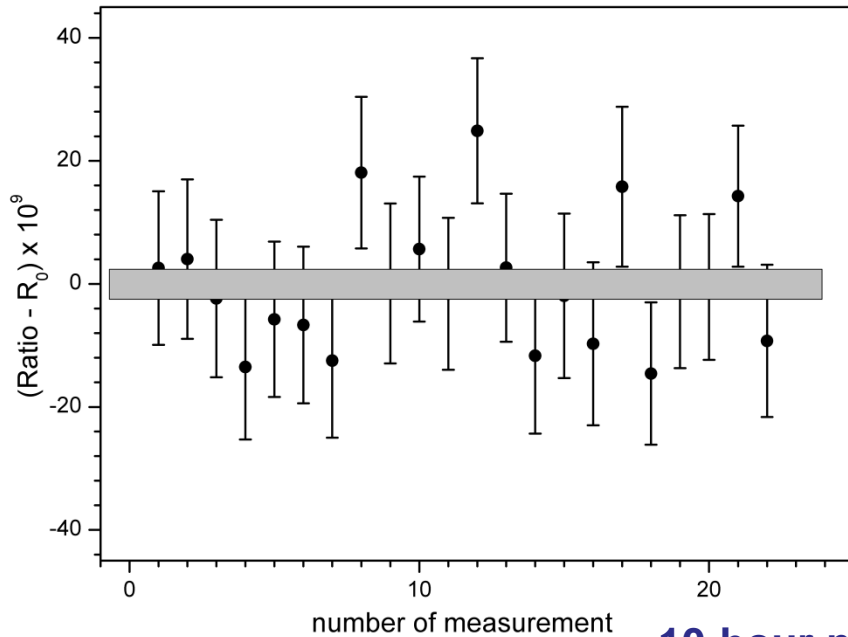
(RoentDek GmbH DLD40)

image of magnetron motion ( $G \approx 20$ )

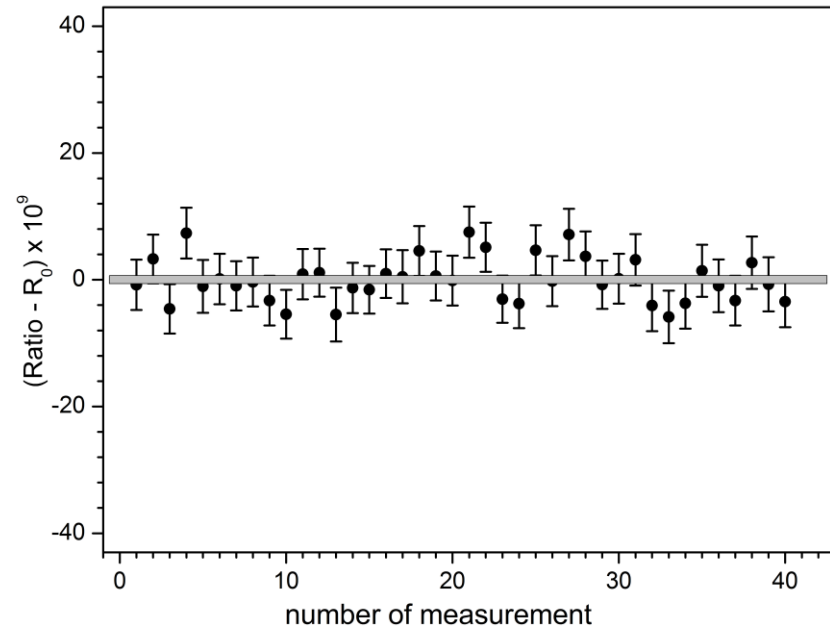


# Increased Precision with Phase Method

## ToF-ICR (Ramsey)



## PI-ICR



10-hour measurements

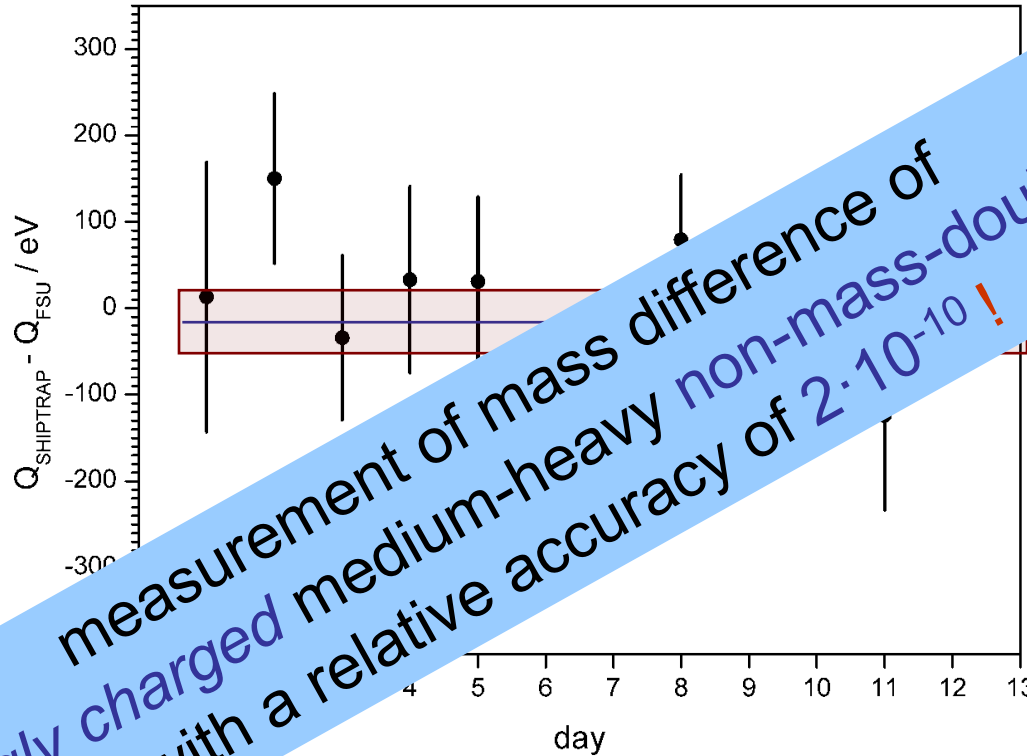
$$\delta[M(^{124}\text{Xe}) - M(^{124}\text{Te})] \sim 300 \text{ eV}$$

$$\delta[M(^{132}\text{Xe}) - M(^{131}\text{Xe})] \sim 70 \text{ eV} !!!$$

**Gain in Precision  $\approx 4.5$  !!!**

# Verifying the Accuracy of PI-ICR

$$\Delta M = M(^{132}\text{Xe}) - M(^{131}\text{Xe})$$



$$\sigma(\Delta M)_{\text{SHIPTRAP}} = (30_{\text{stat}})(12_{\text{sys}}) \text{ eV}$$

$$\Delta M_{\text{SHIPTRAP}} - \Delta M_{\text{FSU}} = (8 \pm 35) \text{ eV}$$

# Selected SHIPTRAP Results with the PI-ICR Technique

# Neutrino Mass Determination

- absolute mass and mass hierarchy of neutrinos still an open question
- present limits:  $m(\bar{\nu}_e) < 2 \text{ eV} / m(\nu_e) < 225 \text{ eV}$

[K.A. Olive \*et al.\* \(Particle Data Group\), \*Chin. Phys. C\*, \*\*38\*\*, 090001 \(2014\).](#)

## Different experimental approaches:

- search for neutrinoless double beta transformation processes
- cosmology
- direct (anti)neutrino mass determination aiming at sub-eV uncertainty
  - spectrometry (KATRIN:  ${}^3\text{H}$   $\beta$  decay )
  - calorimetry (MARE:  ${}^{187}\text{Re}$   $\beta$  decay; ECHO ,HOLMES:  ${}^{163}\text{Ho}$  EC)

**required: independent measurement of  $Q$  - value (mass difference)  
with accuracy on the order of eV**

# $\beta^-$ decay Spectrum Measurement

$\beta^-$ -decay of  ${}^3\text{H}$ ;       $Q$ -value  $\approx 18.6$  keV  
 $\beta^-$ -decay of  ${}^{187}\text{Re}$ ;     $Q$ -value  $\approx 2.47$  keV

**KATRIN**  
**MARE**

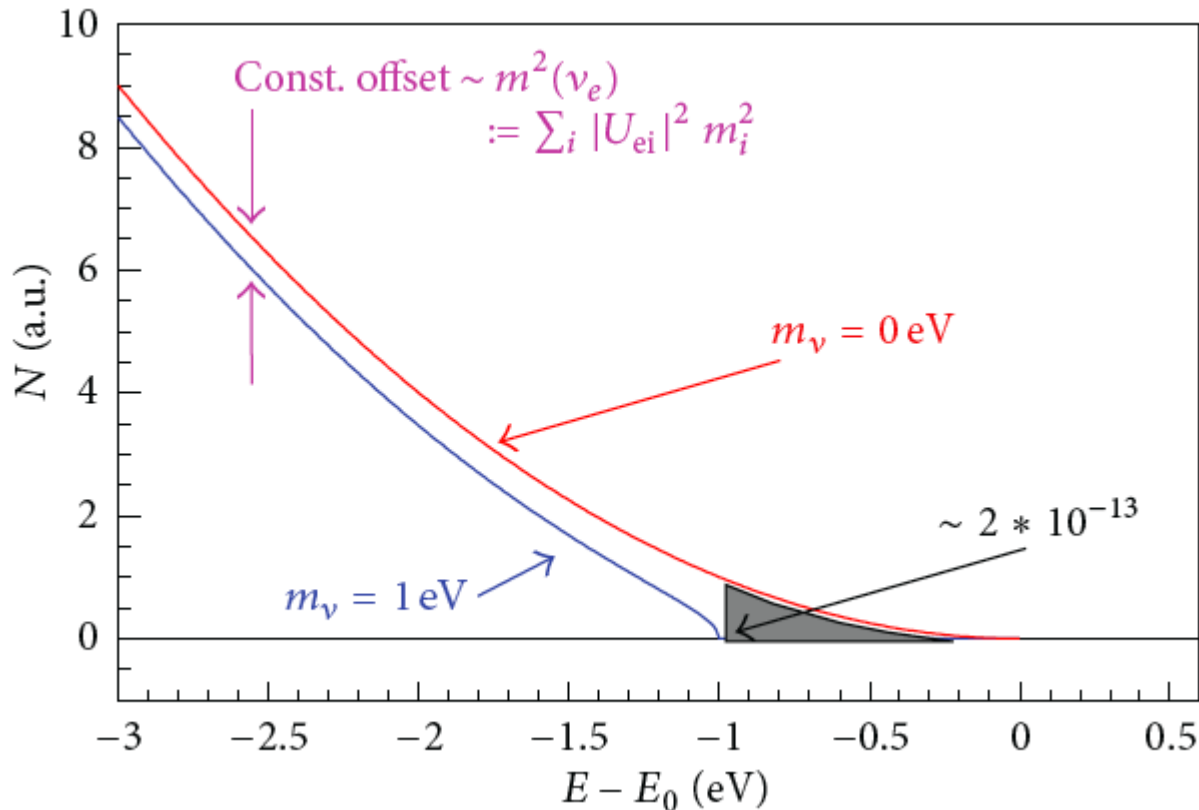
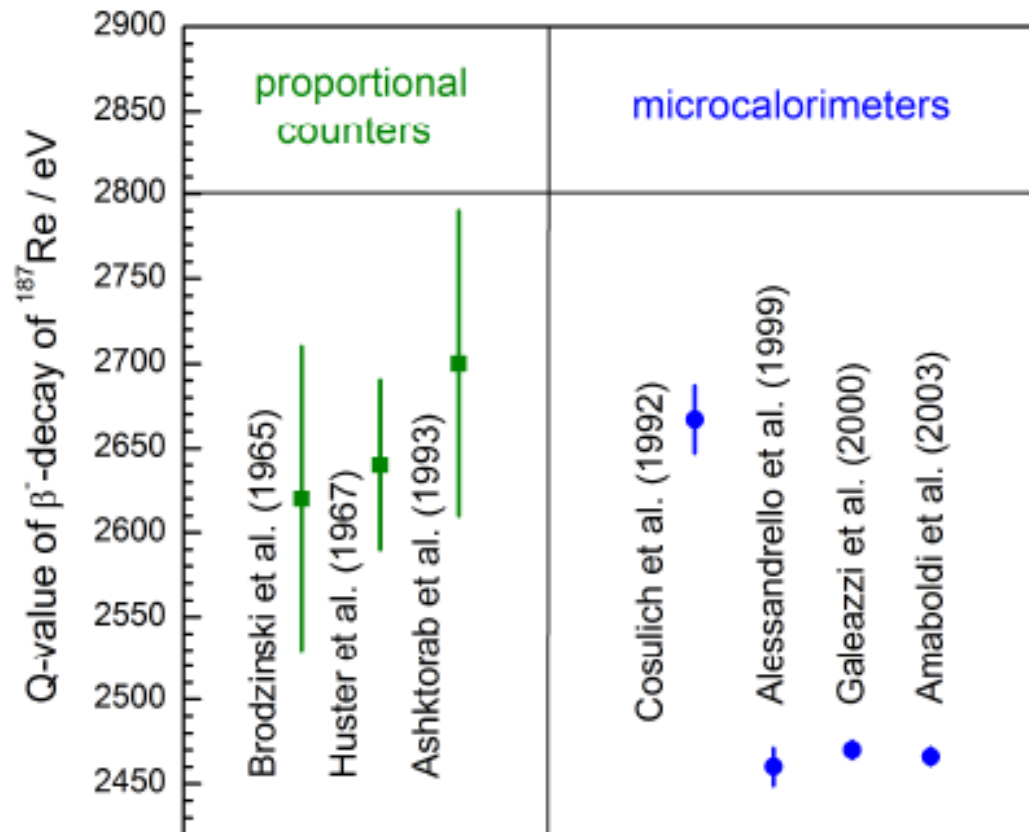


Fig. from G. Drexlin, V. Hannen, S. Mertens, and C. Weinheimer  
Advances in High Energy Physics Volume 2013 (2013)

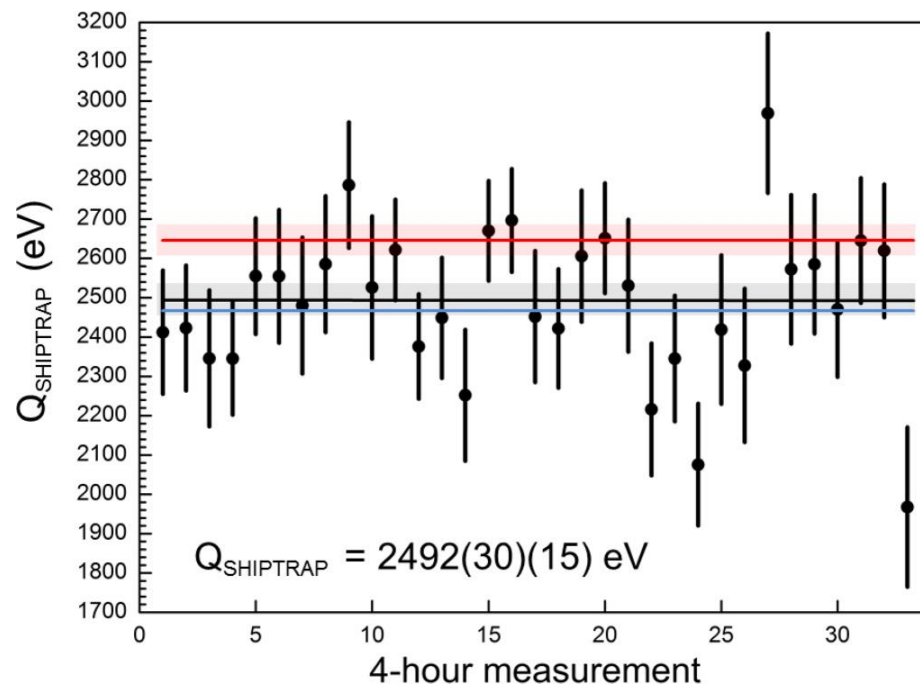
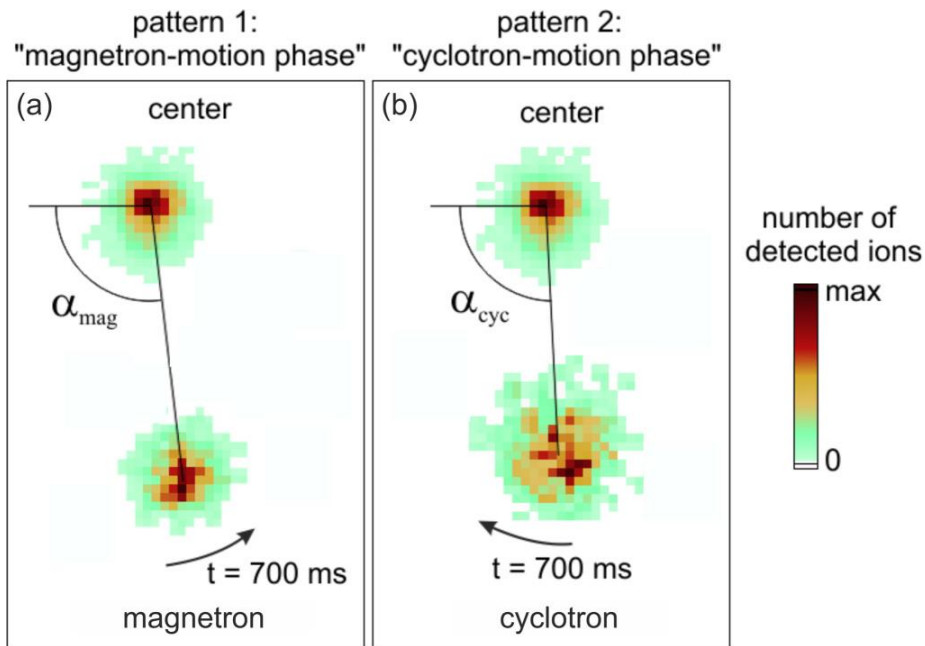


# $^{187}\text{Re}$ $\beta$ Decay $Q$ - Value



$\beta^-$ -decay of  $^{187}\text{Re}$ ;  $Q$ -value  $\approx 2.47$  keV

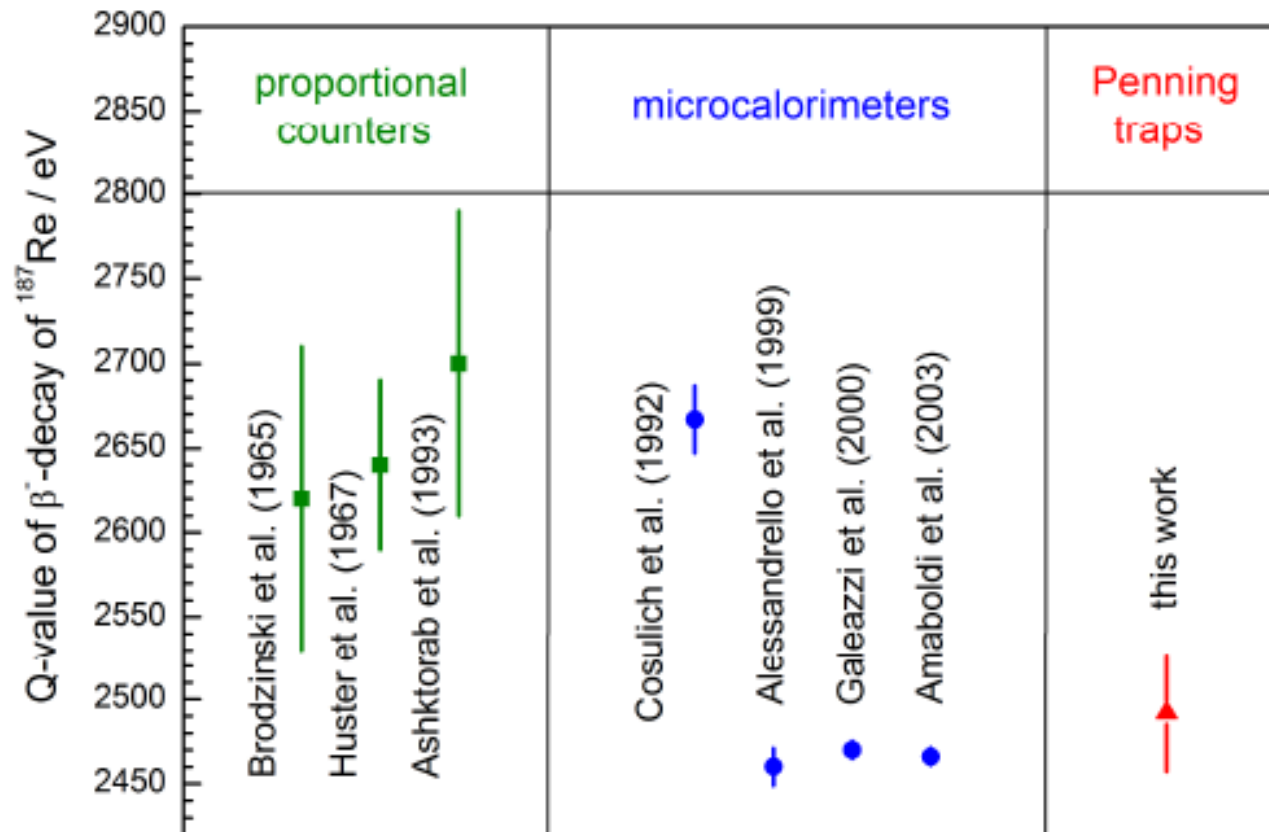
# SHIPTRAP results $^{187}\text{Re}/^{187}\text{Os}$ mass difference



**SHIPTRAP:**  $^{187}\text{Re}$   $\beta^-$ -decay

**$Q$  - value = 2492(30)(15) eV**

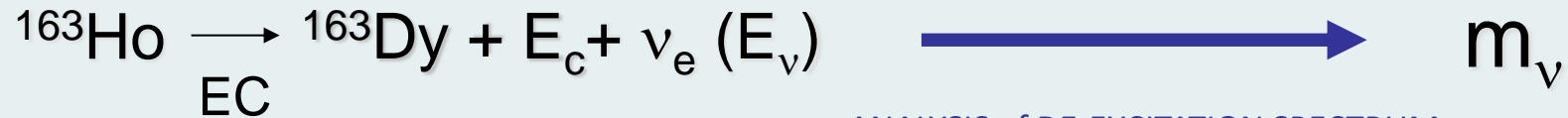
# $^{187}\text{Re}$ $\beta$ Decay $Q$ - Value



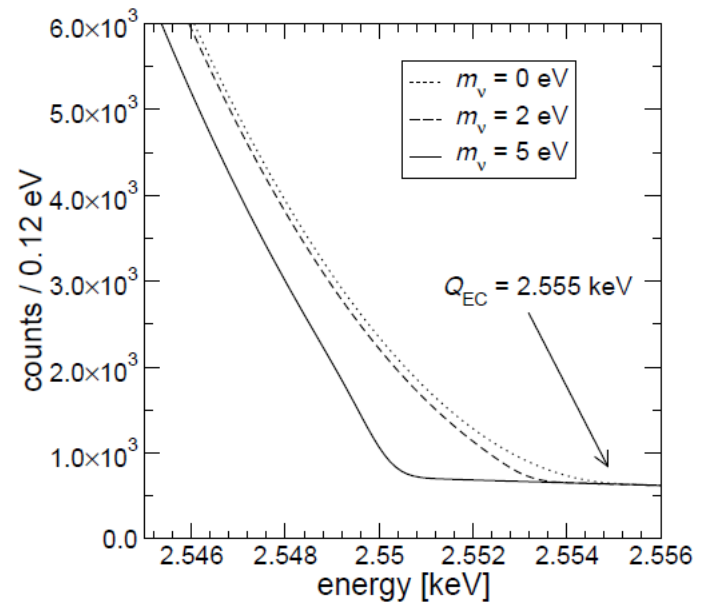
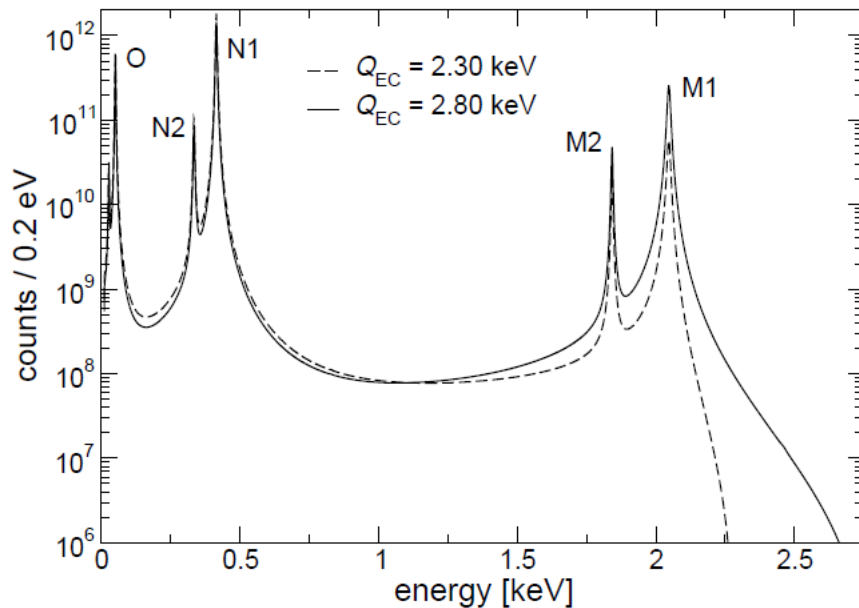
**SHIPTRAP result confirms latest micro-calorimeter results**

# Electron Capture in $^{163}\text{Ho}$

EC in  $^{163}\text{Ho}$ ; Q-value  $\approx 2.55$  keV (AME 2012)

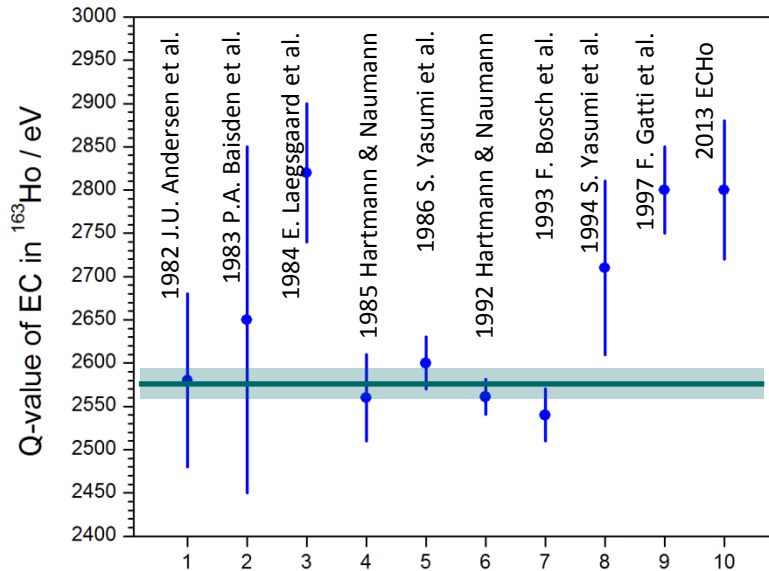


ANALYSIS of DE-EXCITATION SPECTRUM

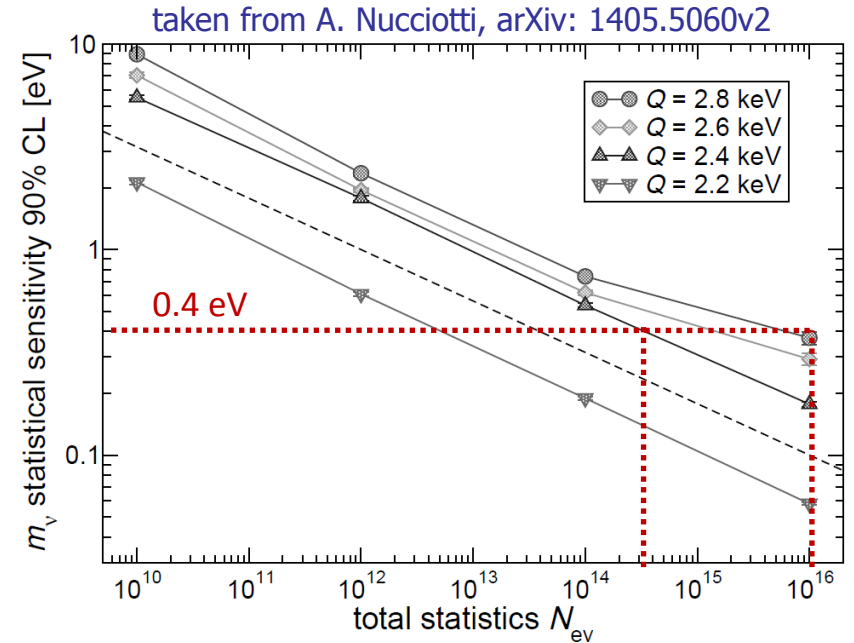


# $^{163}\text{Ho}$ EC Decay $Q$ - Value

## $Q$ -value of EC in $^{163}\text{Ho}$



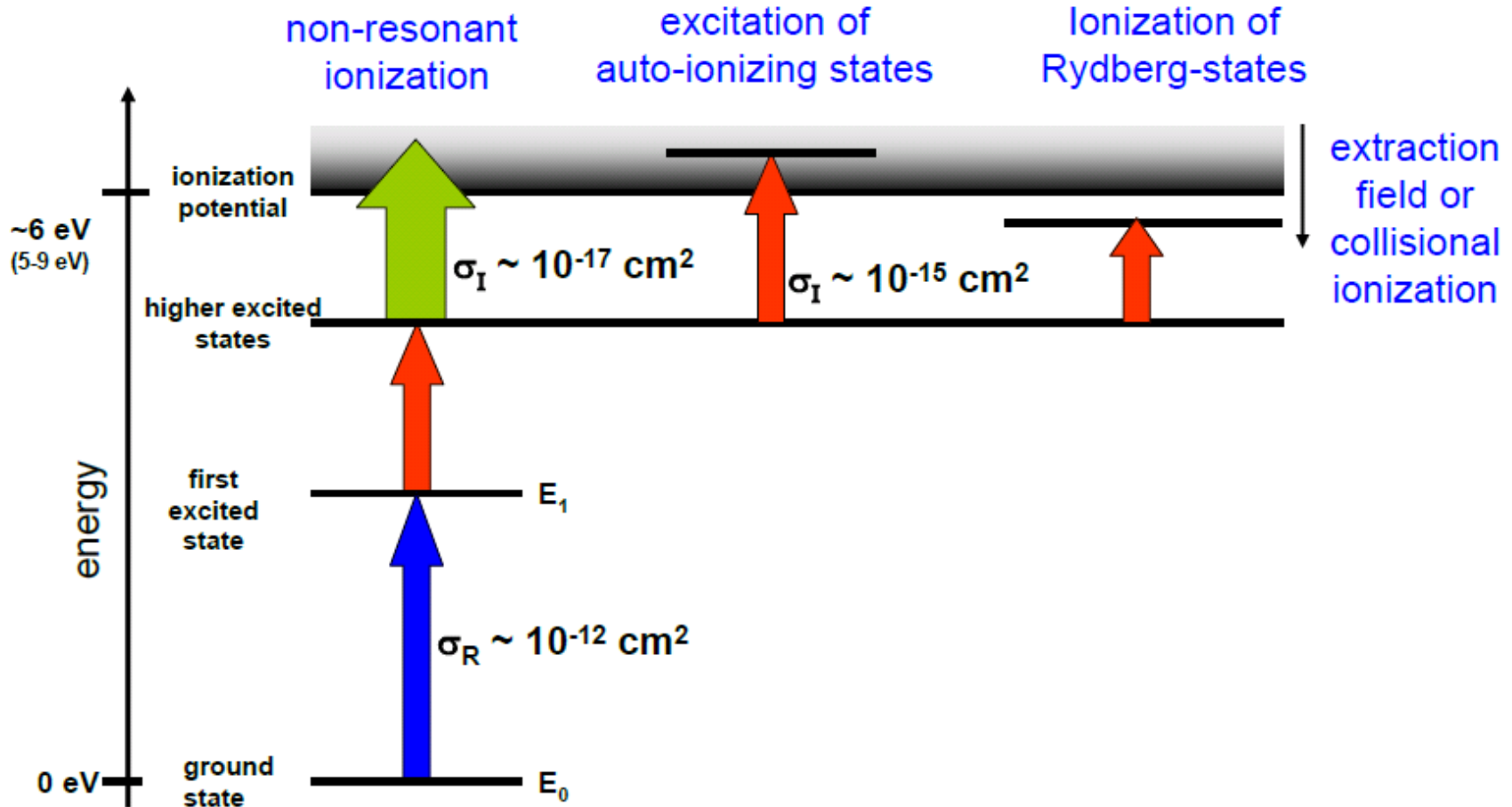
## Statistical sensitivity to $m_\nu$



direct measurement of mass difference  
 $^{163}\text{Ho}$ - $^{163}\text{Dy}$  can clarify situation

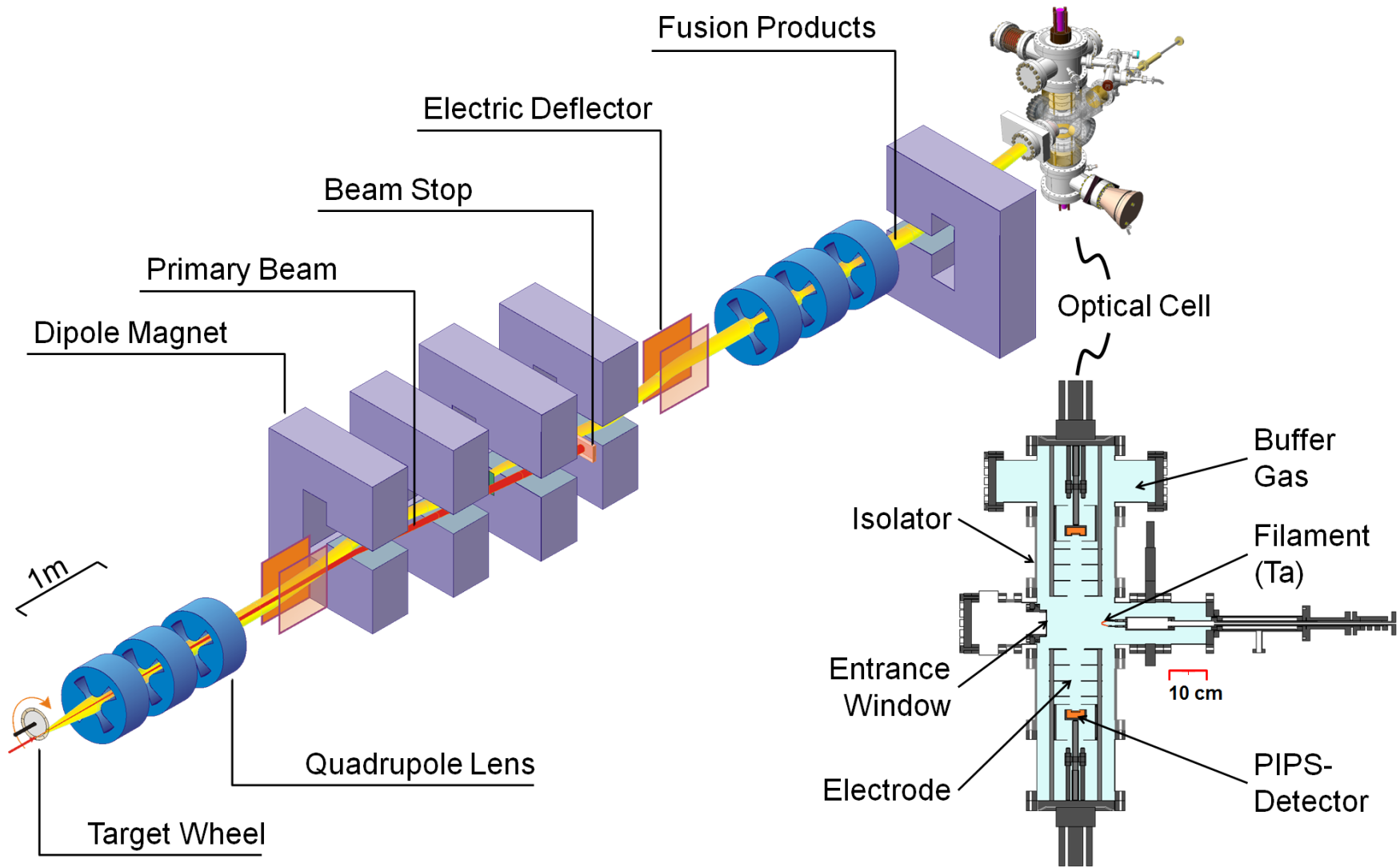
# Laser Spectroscopy of the Heaviest Elements

# Principles of Resonant Laser Ionization



**Efficiency  $\times$  Selectivity**

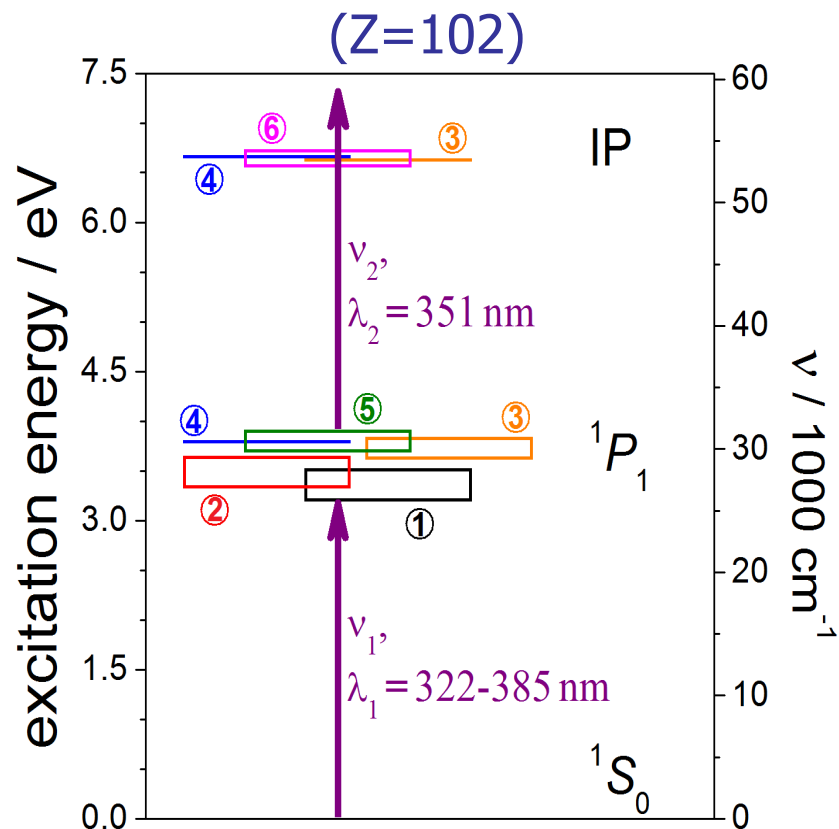
# Radiation Detected Resonance Ionization Spectroscopy





# Search for Atomic Transitions in Nobelium

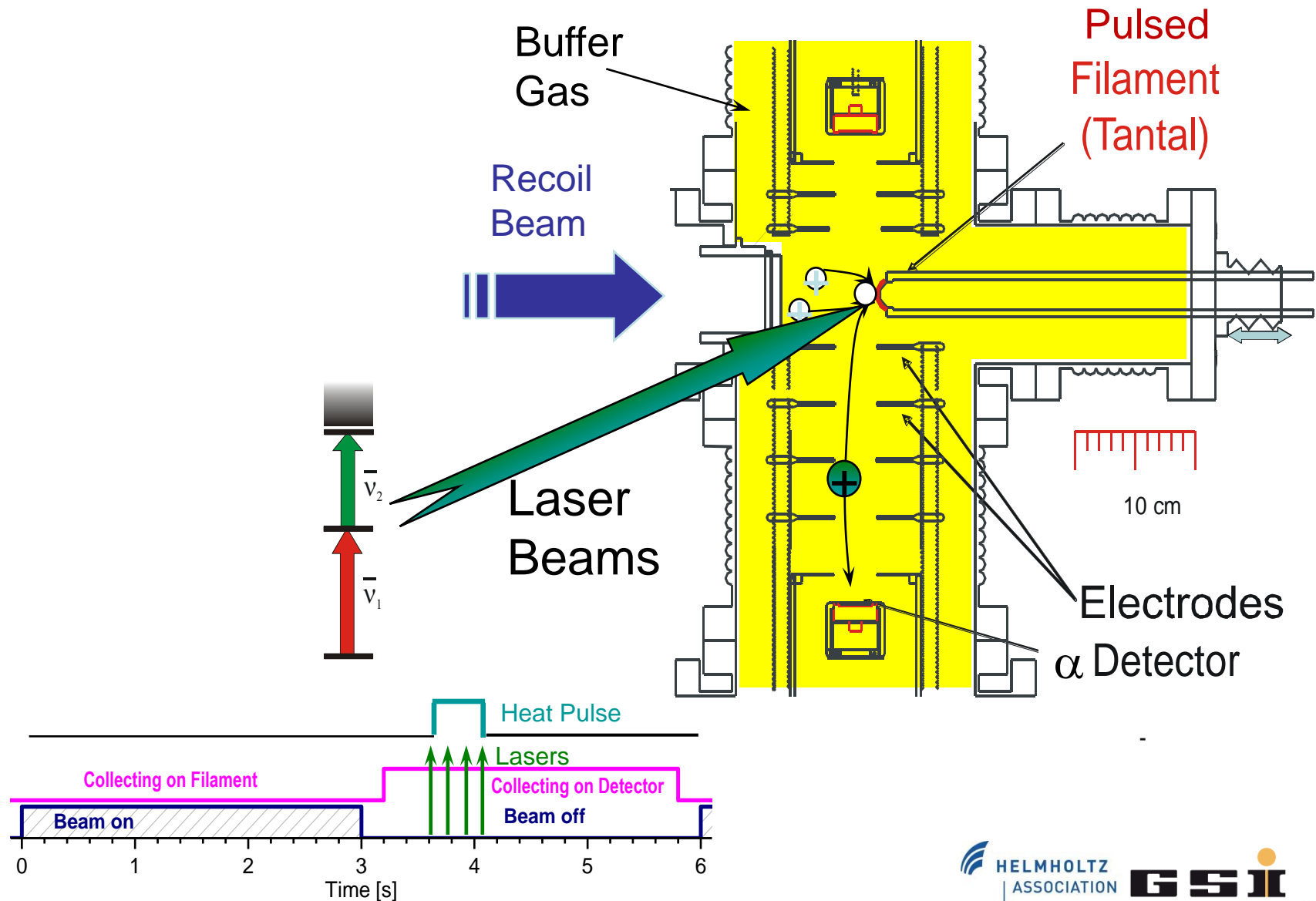
## Theoretical predictions for the $^1S_0$ - $^1P_1$ - transition in the element nobelium



- RIS with two step excitation and non-resonant second step
- search for  $^1P_1$  level in range predicted by different theories
- determine IP via Rydberg series
- Measure isotope shift of  $^1P_1$ - $^1S_0$  transition

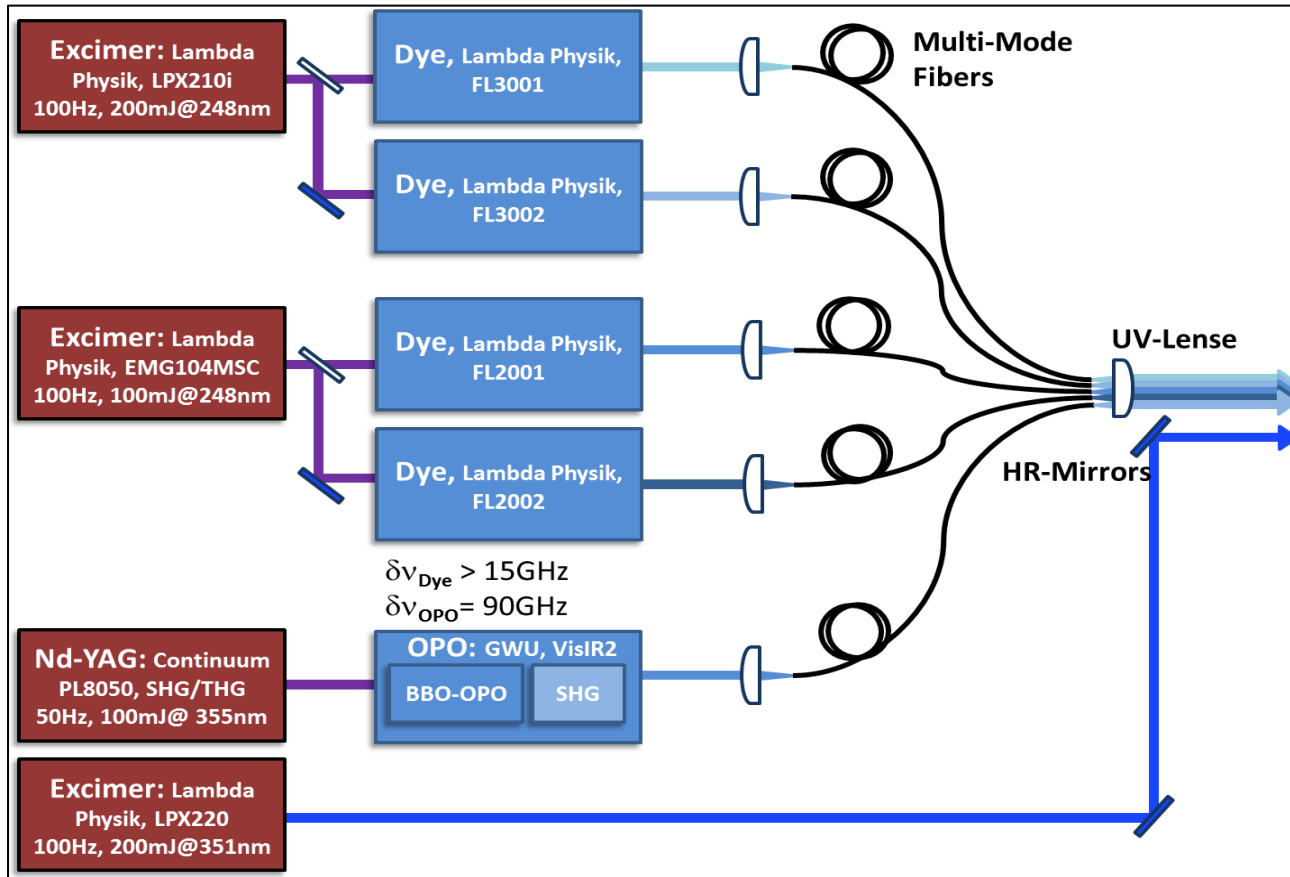
- [1],[2]: S. Fritzsche, Eur. Phys. J. D 33 (2005) 15  
[3]: A. Borschevsky et al., Phys. Rev. A 75 (2007) 042514  
[4]: Y. Liu et al., Phys. Rev. A 76 (2007) 062503  
[5]: P. Indelicato et al., Eur. Phys. J. D 45 (2007) 155  
[6]: J. Sugar, J. Chem. Phys. 60 (1974) 4103

# Resonant Ionization Laser Spectroscopy of Nobelium

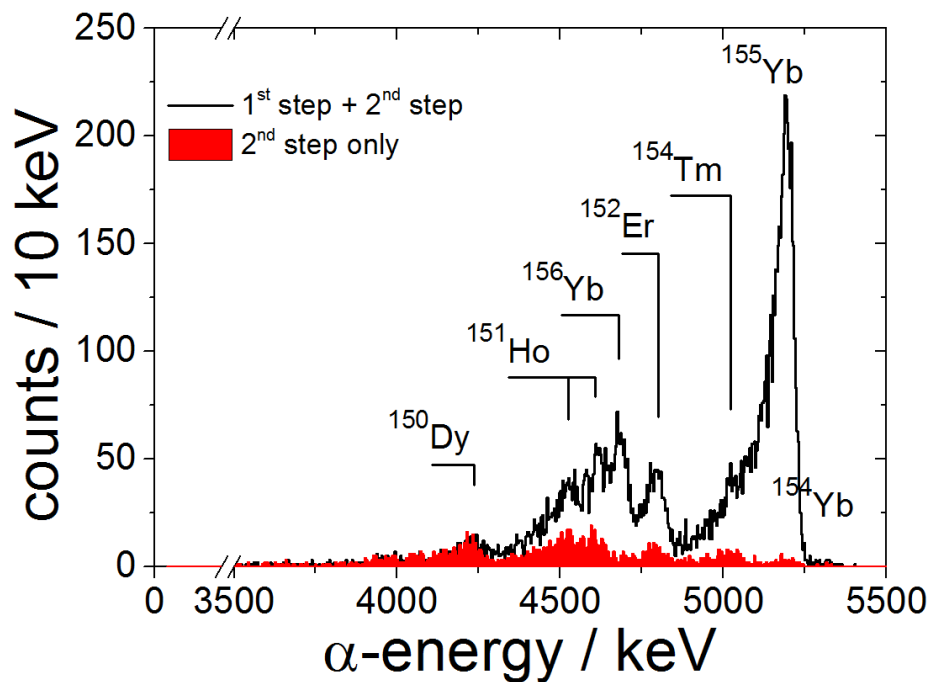


# Laser System

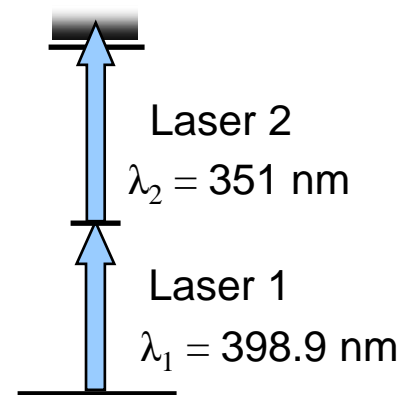
## Laser Systems



# RADRIS-optimization: on-line experiments ( $^{155}\text{Yb}$ )



2-step excitation  $^{155}\text{Yb}$



**overall efficiency 1% achieved**

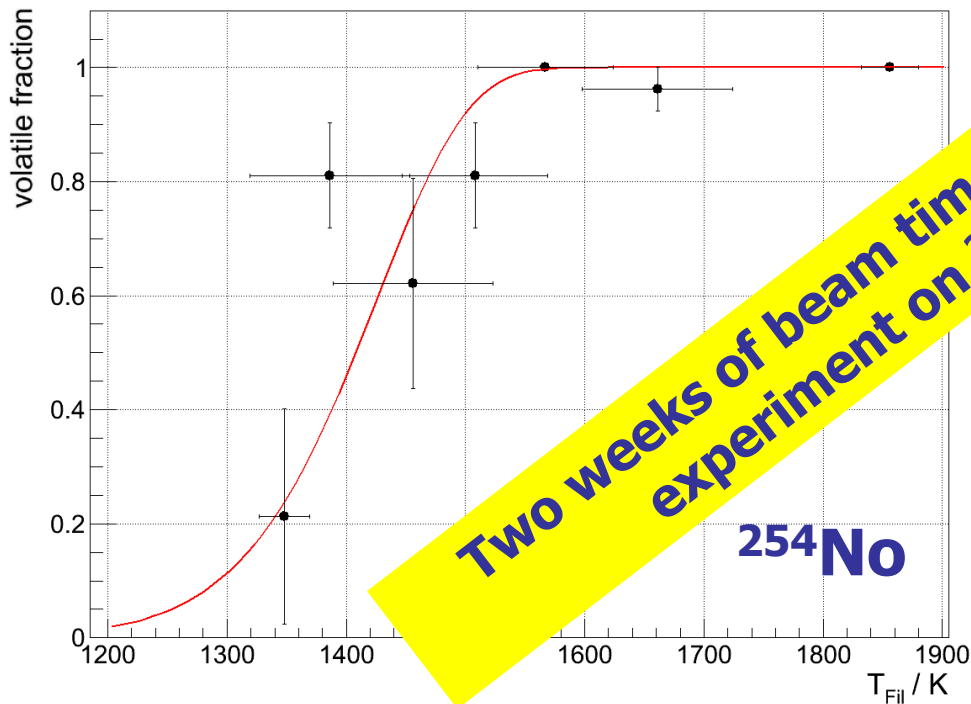
experiments on the chemical homolog Yb allow:

- optimization of full setup
- localizing the atom cloud
- monitoring of overall efficiency during level-search in nobelium

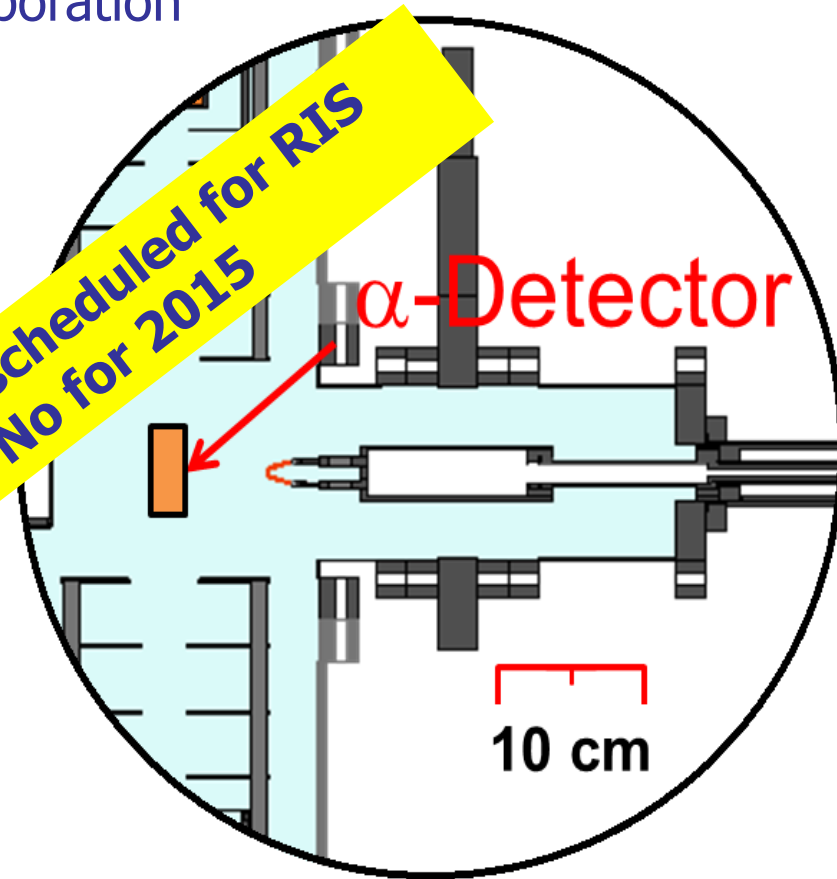
# Evaporation of $^{254}\text{No}$ from Ta Filament

- measure residual activity on filament
- determine filament temperature for evaporation
- Desorption enthalpy of No from Ta:

$246 \pm 24 \text{ kJ/mol}$



Two weeks of beam time scheduled for RIS experiment on  $^{254}\text{No}$  for 2015



# Future Perspectives



# Superheavy Elements Subcollaboration of NUSTAR @ FAIR

Proposal to integrate new "Superheavy Element" subcollaboration in NUSTAR @ FAIR submitted to Board of Representatives (Summer '14)

**Focus: synthesis, nuclear structure, atomic physics, nuclear chemistry experiments in region  $Z \geq 100$**

Existing facilities: SHIP, TASCA, SHIPTRAP, Chemistry beamline  
Developments for high-intensity cw-Linac ongoing (HIM, GSI, U Frankfurt)



Complementary to existing NUSTAR activities at Super-FRS

Organizational Structure:

Spokesperson: R.-D. Herzberg (Univ. Liverpool)  
Deputy: M. Block (GSI/HIM/JGU)  
Technical Director: A. Yakushev (GSI)

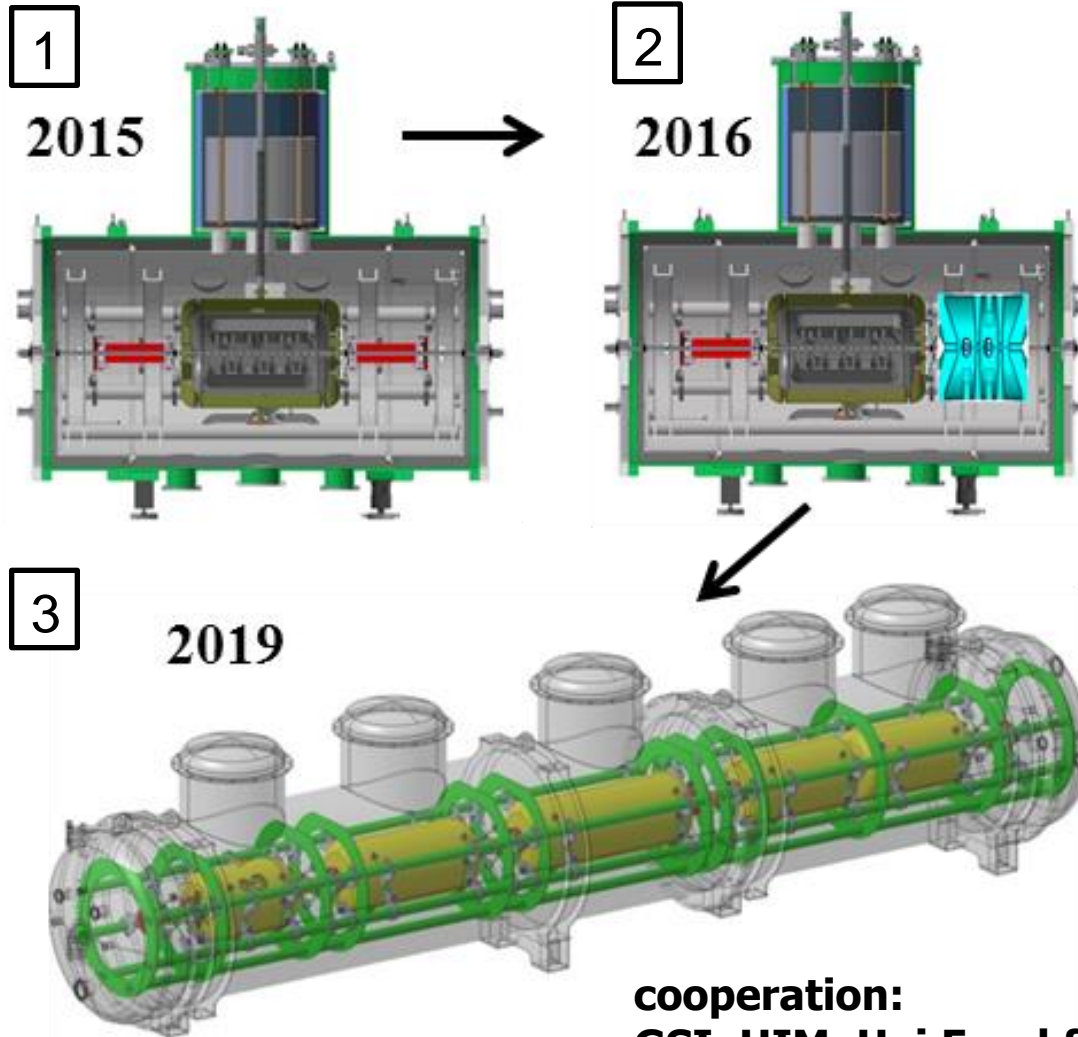
Currently includes 9 German and 17 international institutes

*Endorsed by NUSTAR Collaboration Committee:  
submission to FAIR management:*

*Sept. 25, 2014  
summer 2015*



# Staged Approach towards cw linac for SHE



## 1. Full performance test of sc cw LINAC Demonstrator

- @GSI HLI
- proof of principle

## 2. Full performance test of a shorter sc cavity

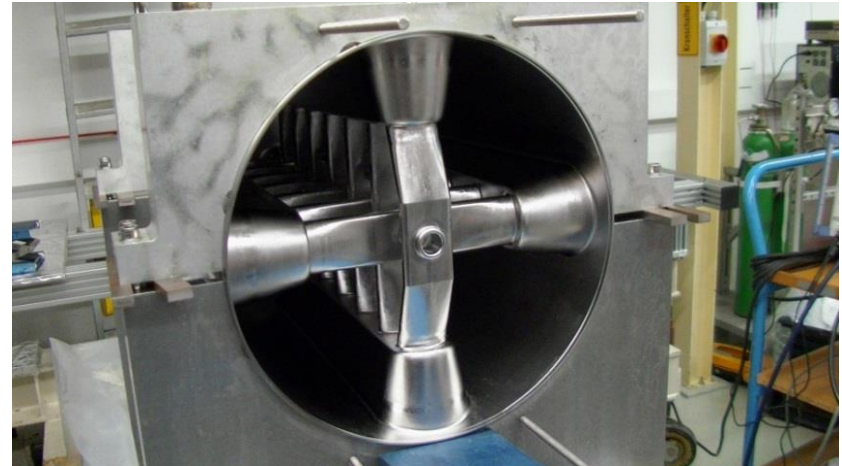
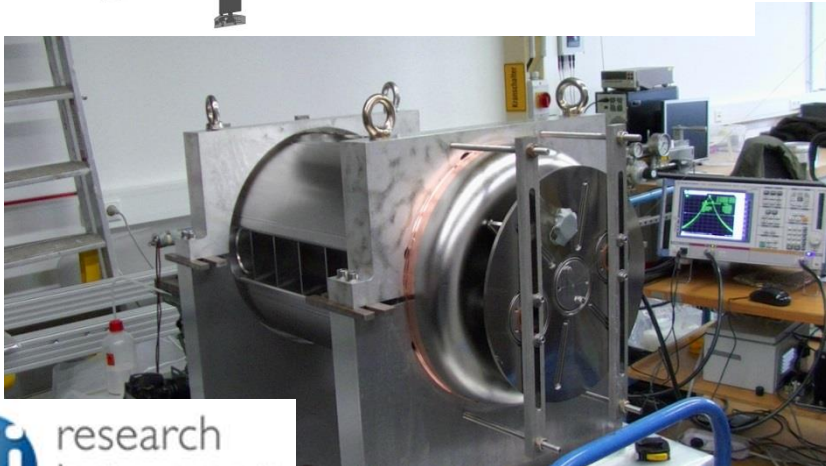
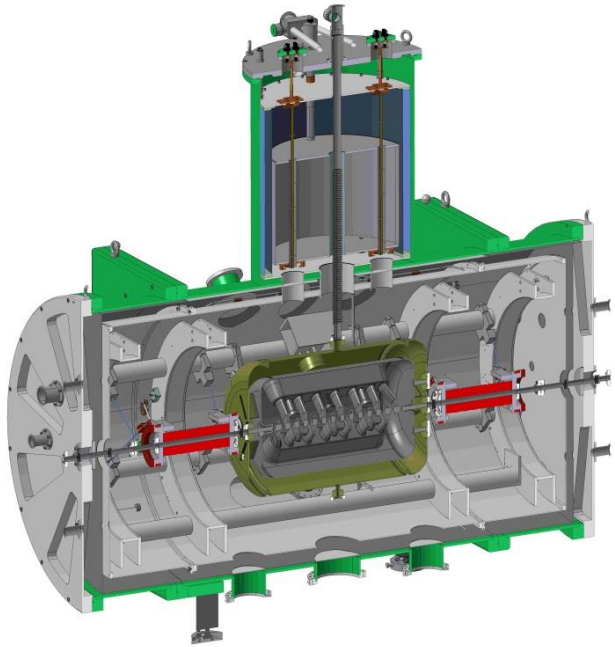
- energy variation (by Ampl & Phase)
- 8 gaps
- simpler design
- easier to fabricate

## 3. Advanced Demonstrator

- up to 4.61 MeV/u @  $A/Q = 6$
- 5× sc CH-Cavity, 5× sc Solenoid
- possible to place in HLI@GSI

cooperation:  
GSI, HIM, Uni Frankfurt

# First components – October 2014



# Summary and Conclusions

- Direct high-precision mass measurements provide complementary tools to map nuclear structure effects in the heaviest elements
- Increased resolving power and higher precision of novel PI-ICR method opens the door for applications in fundamental physics
- Laser spectroscopy for  $Z > 100$  allows studying the impact of relativistic effects on the atomic structure
- Laser spectroscopy will also provide information of nuclear properties such as spins, moments, and changes in charge radii (model independent)
- stepwise approach for new cw-linac underway -> GSI will maintain leading position in SHE research

**Thank you for your attention !**