

Nuclear Structure of Heavy Nuclei studied by Laser Spectroscopy and Mass Spectrometry at GSI/SHIP

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Acknowledgements

Contributions by the SHE physics groups @ GSI / HIM, the SHIPTRAP, RADRIS, Gas-Jet, and Fm- Collaborations as well as the GSI accelerator department and the GSI target lab are gratefully acknowledged!



Fermium Collaboration

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CNRS/IN2P3



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Th. Albrecht-Schönzart, A. Gaiser, J. Sperling

Institut Laue Langevin

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This Marie Skłodowska-Curie Action (MSCA) Innovative Training Network (ITN) receives funding from the European Union H2020 Framework Programme under grant agreement no. 861198. LISA will run from November 2019 to October 2023. This research is supported by the U.S. DOE, Office of Science, BES Heavy Element Chemistry program. The isotopes used in this research were supplied by the U.S. DOE Isotope Program, managed by the Office of Science for Nuclear Physics.



References

Review Articles:

- M. Block, F. Giacoppo, F.P. Hessberger, S. Raeder, Riv. Nuovo Cim. 45, 279–323 (2022)
- Ch. Düllmann, M. Block, *et al.*, Radiochim. Acta, 110, 417 (2022)
- M. Block, M. Laatiaoui, S. Raeder, Prog. Nucl. Part. Phys. 116 (2021) 103834

Select Recent Publications on Laser Spectroscopy and Mass Measurements:

- J. Warbinek *et al.*, *Atoms* 10, 41 (2022)
- O. Kaleja *et al.*, PRC 120.23 (2022): 054325
- S. Raeder *et al.*, *NIM B* 463 (2020): 272-276
- S. Raeder *et al.*, PRL 120.23 (2018): 232503
- P. Chhetri *et al.*, PRL 120.26 (2018): 263003.
- M. Laatiaoui *et al.*, Nature 538.7626 (2016): 495-498.

See also: <https://orcid.org/0000-0001-9282-8347>

Outline

- Open Questions in Superheavy Element Research
- Employed Methods and Techniques:
 - Mass Spectrometry with Penning Traps
 - Laser spectroscopy with Gas Cells
- Shell Structure around $N = 152$ from Mass Measurements
- Changes in Charge Radii of Fm and No Isotopes around $N = 152$
- Summary and Conclusions

The Periodic Table of Elements 2022



United Nations
Educational, Scientific and
Cultural Organization



2019
International Year
of the Periodic Table
of Chemical Elements

1 H																	2 He	
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne	
11 Na	12 Mg											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			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			104 Rf 1964	105 Db 1968	106 Sg 1974	107 Bh 1981	108 Hs 1984	109 Mt 1982	110 Ds 1994	111 Rg 1994	112 Cn 1996	113 Nh 2004	114 Fl 1999	115 Mc 2004	116 Lv 2000	117 Ts 2010	118 Og 2006
119 ---	120 ---																	

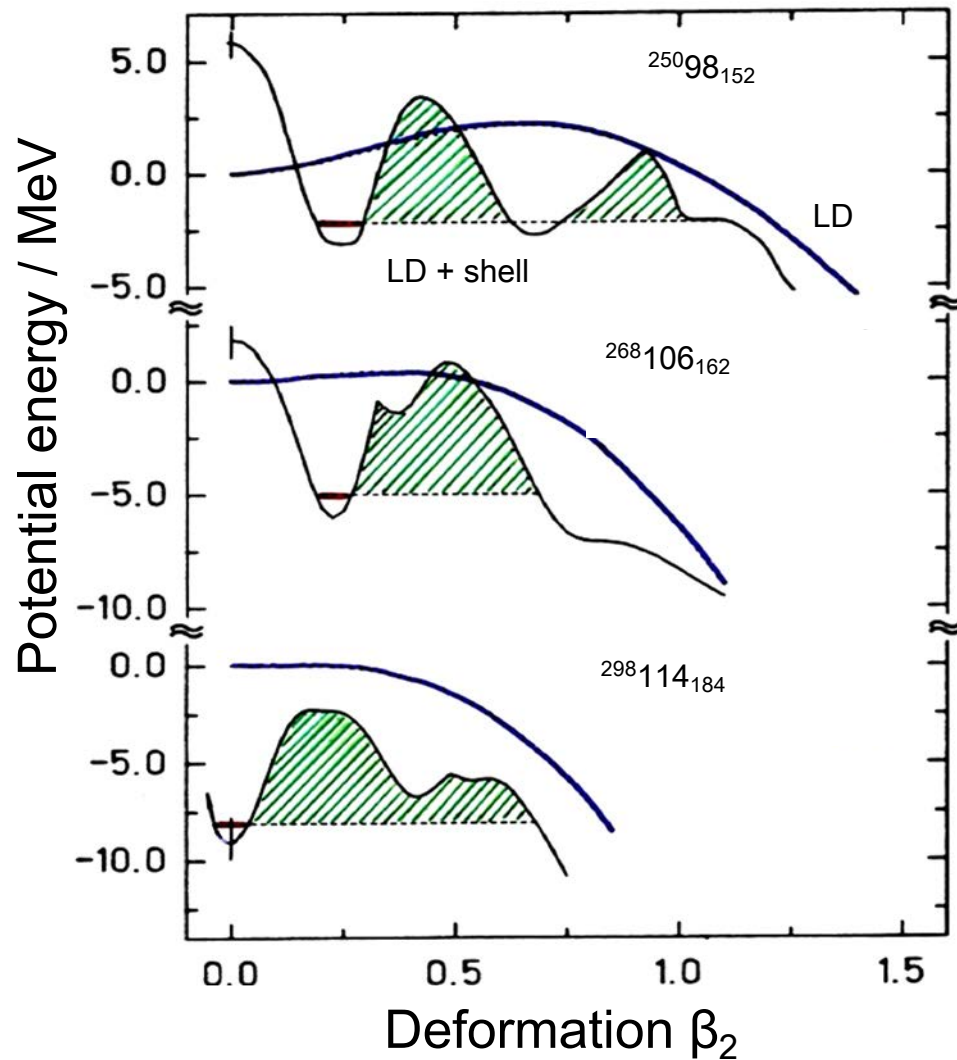
57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

- elements beyond uranium can be produced in nuclear reactors and with accelerators
- elements **Bh, Hs, Mt, Ds, Rg, Cn** (Z=107-112) were discovered at SHIP / GSI Darmstadt

Superheavy Elements
= Transactinide Elements

Actinide Elements

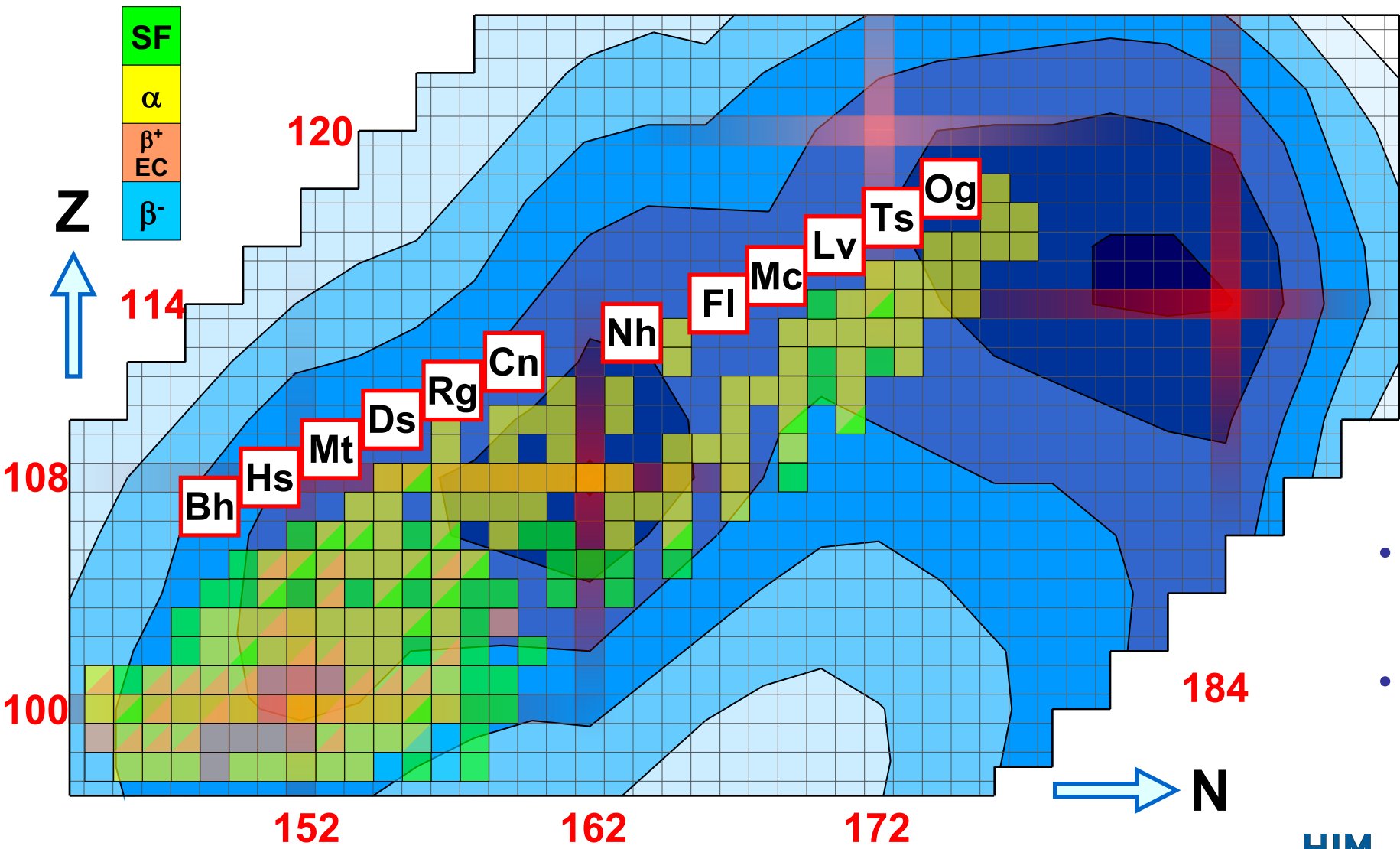
Fission Barriers in Superheavy Nuclei



- fission barrier decreases with increasing Z
- liquid drop barrier vanishes around $Z = 106$
- superheavy nuclei (SHN) gain up to 10 MeV in binding energy by nuclear shell effects
- leads to finite fission barrier in SHN with $Z > 106$

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

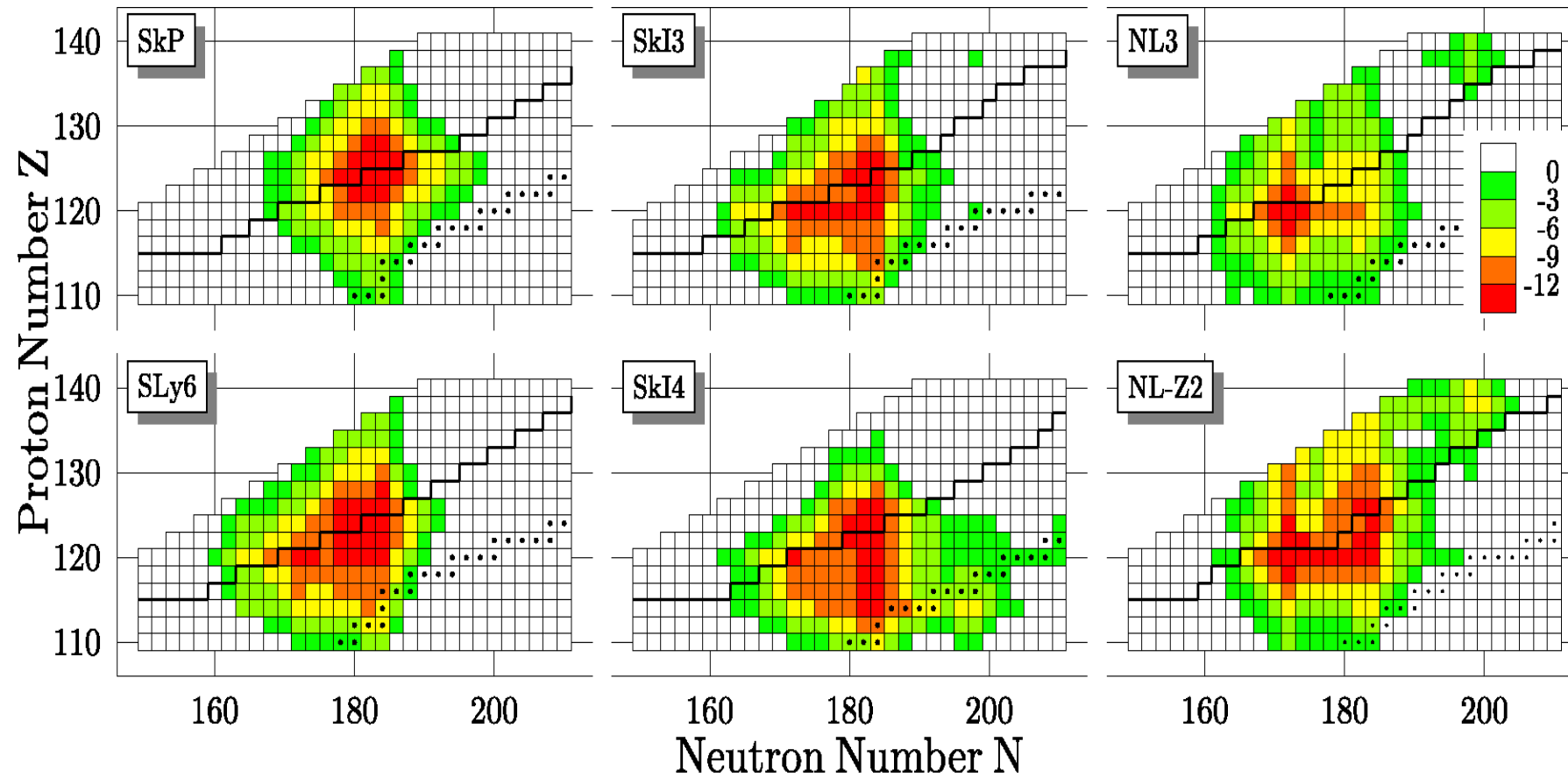
Superheavy Nuclei – Landscape



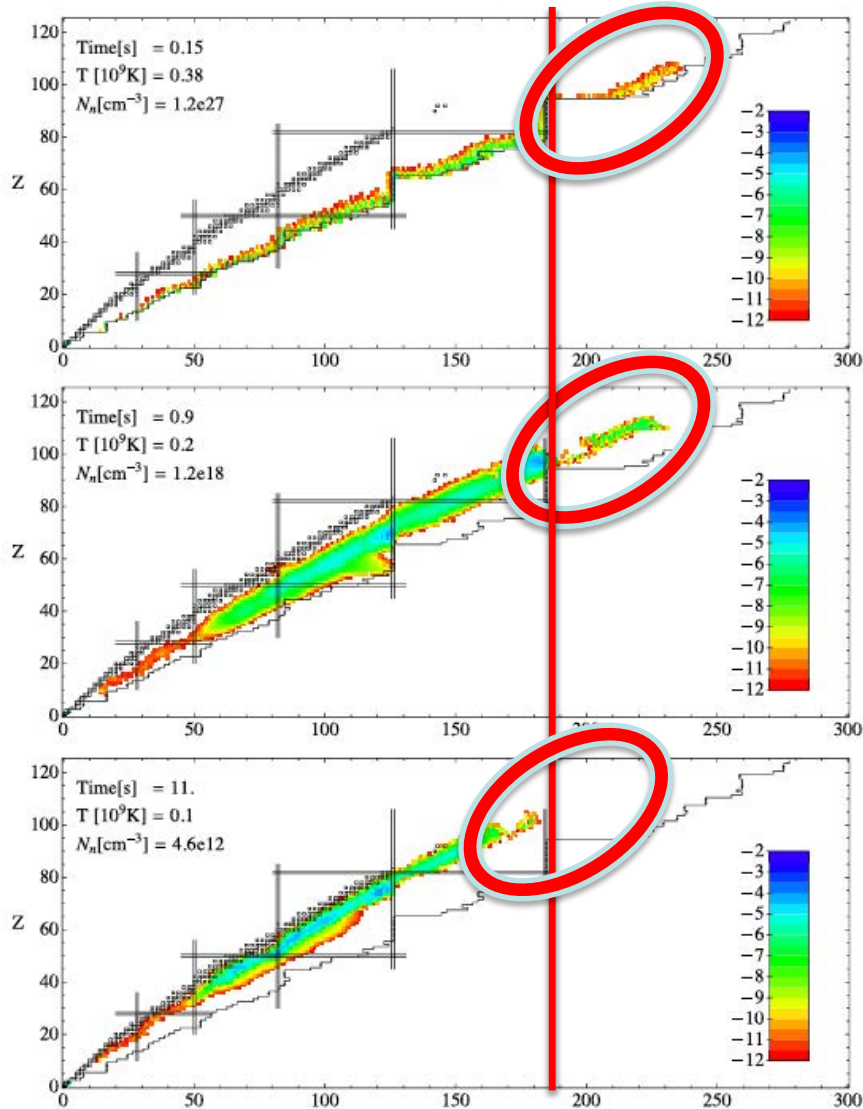
- presently 118 elements known up Og
- rather neutron-deficient and short-lived isotopes

Figure courtesy Ch. Düllmann

Nuclear Shells: Magic Numbers in SHE?



Are superheavy elements produced in the r process?



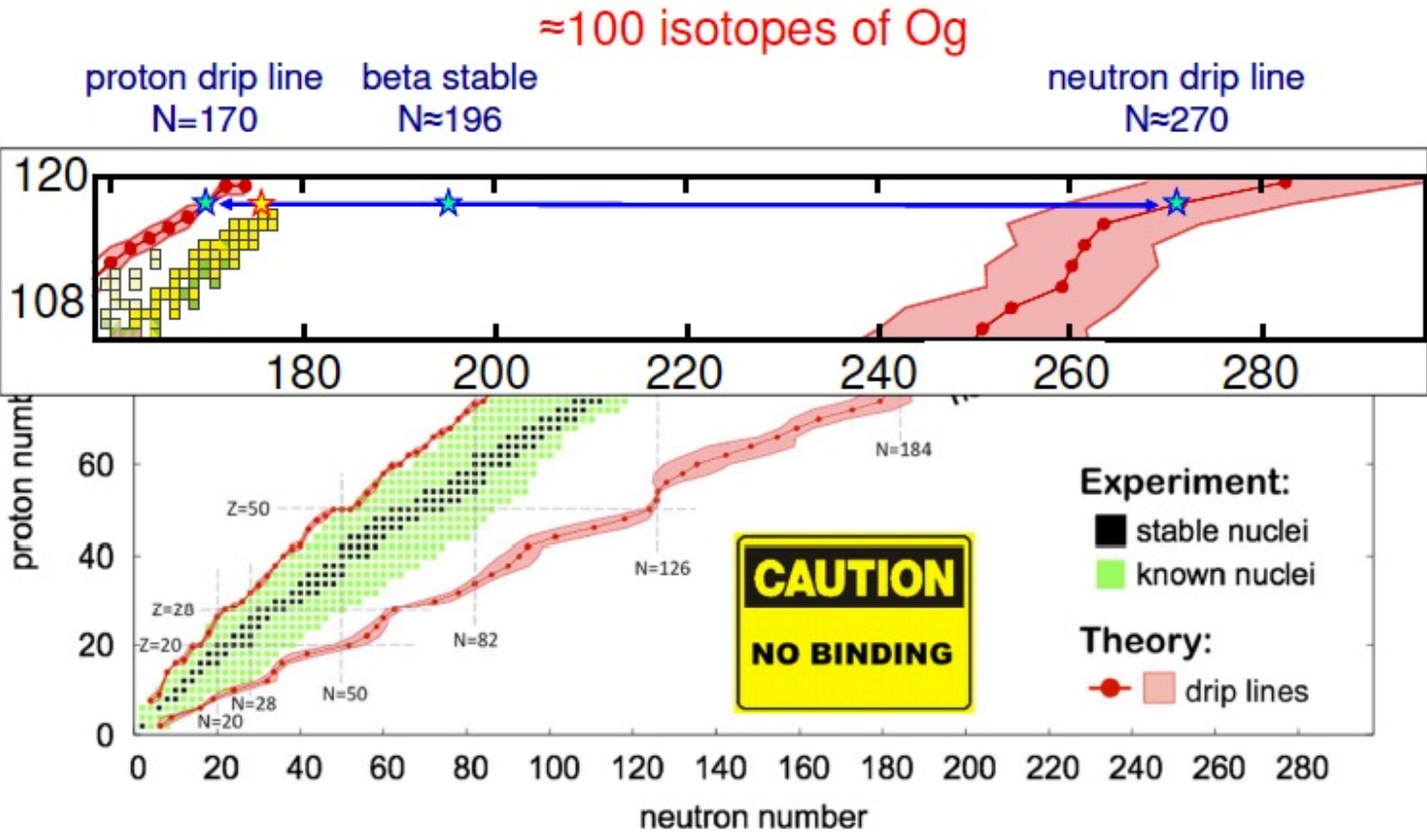
- r process path towards heaviest elements terminated by fission (fission recycling)
- fission barrier heights strongly model dependent, thus accurate description of fission is crucial
- impact of shell structure, e.g., $N = 184$
- isomers play a role
- limited experimental data for relevant nuclei

Superheavy Element Research – Key Questions

- Where is the end of the periodic table in atomic number and mass?
- What are the boundaries of the *island of stability* and what are the properties of nuclei there?
- Are there remnants of long-lived superheavy elements on earth?
- How do relativistic effects affect the architecture of the periodic table?

See e.g. recent review by S.A. Giuliani et al., Rev. Mod. Phys. 91, 011001 (2019) and special issue on SHE in Nucl. Phys. A 944 (2015)

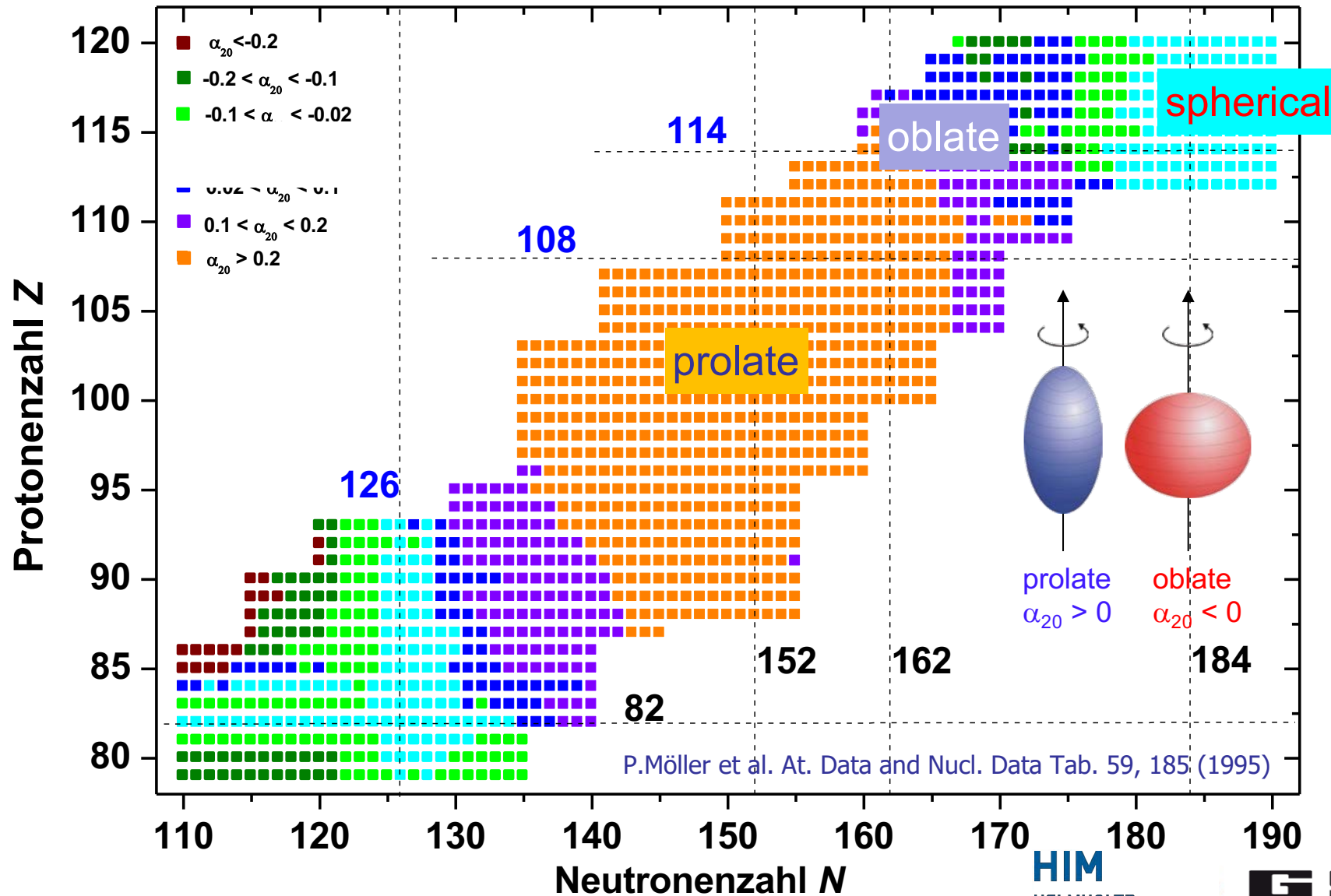
Superheavy Element Territory



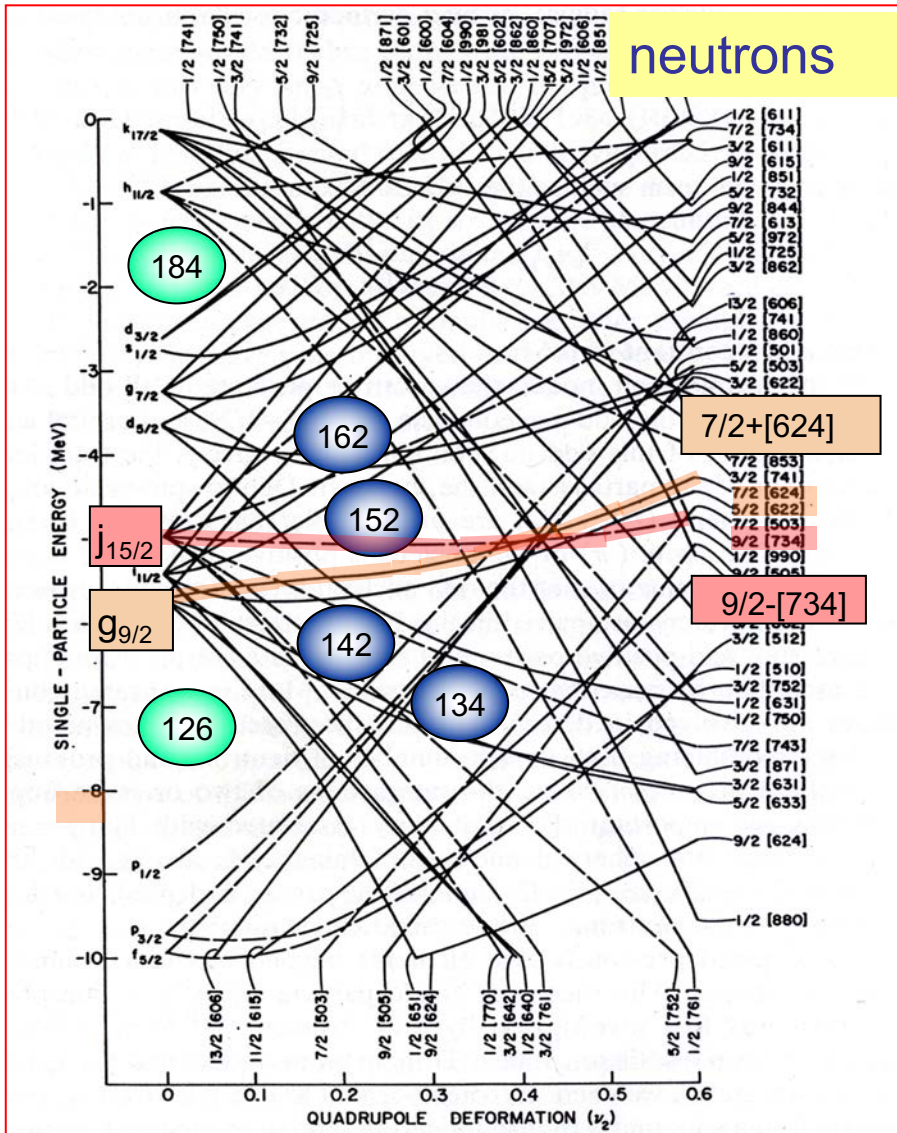
- nuclear models predict existence of many more superheavy nuclides
- maybe not too many new elements, but many more neutron-rich isotopes
- difficult to produce
 - radioactive beams?
 - other nuclear reaction mechanisms?

figure courtesy of W. Nazarewicz, MSU/FRIB

Deformation in the Region of Superheavy Nuclides

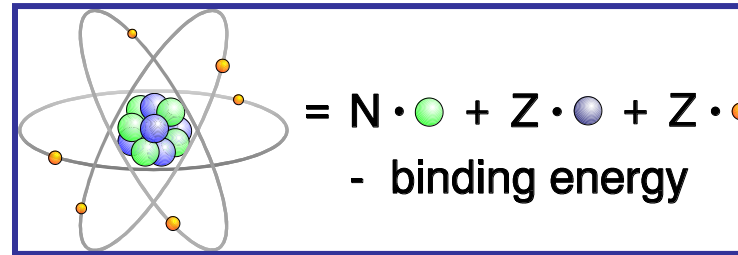


Nuclear Shell Structure and Deformation



- single-particle energies change with nuclear deformation – Nilsson model
- “new“ energy gaps appear for specific values of nuclear deformation , e.g., $N = 152, 162$
- shell effects are linked to stability and hence, existence of superheavy nuclei
- mass measurement gives access to shell effects via binding energy
- laser spectroscopy provides information on shape and size of nuclei

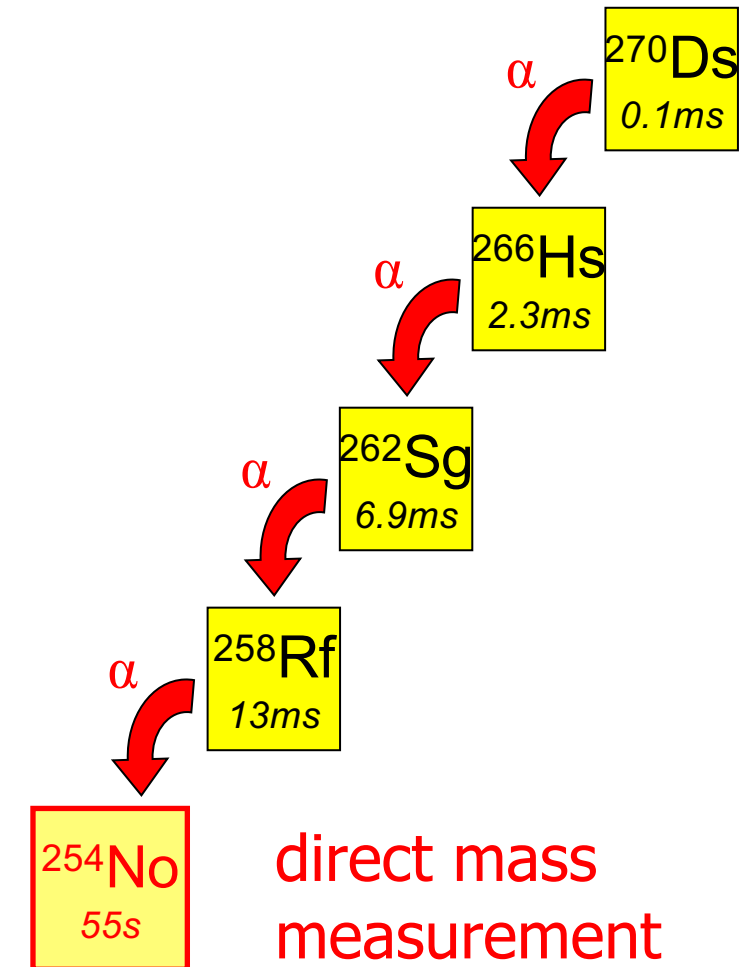
Direct Mass Measurements of Heavy and Superheavy Nuclei



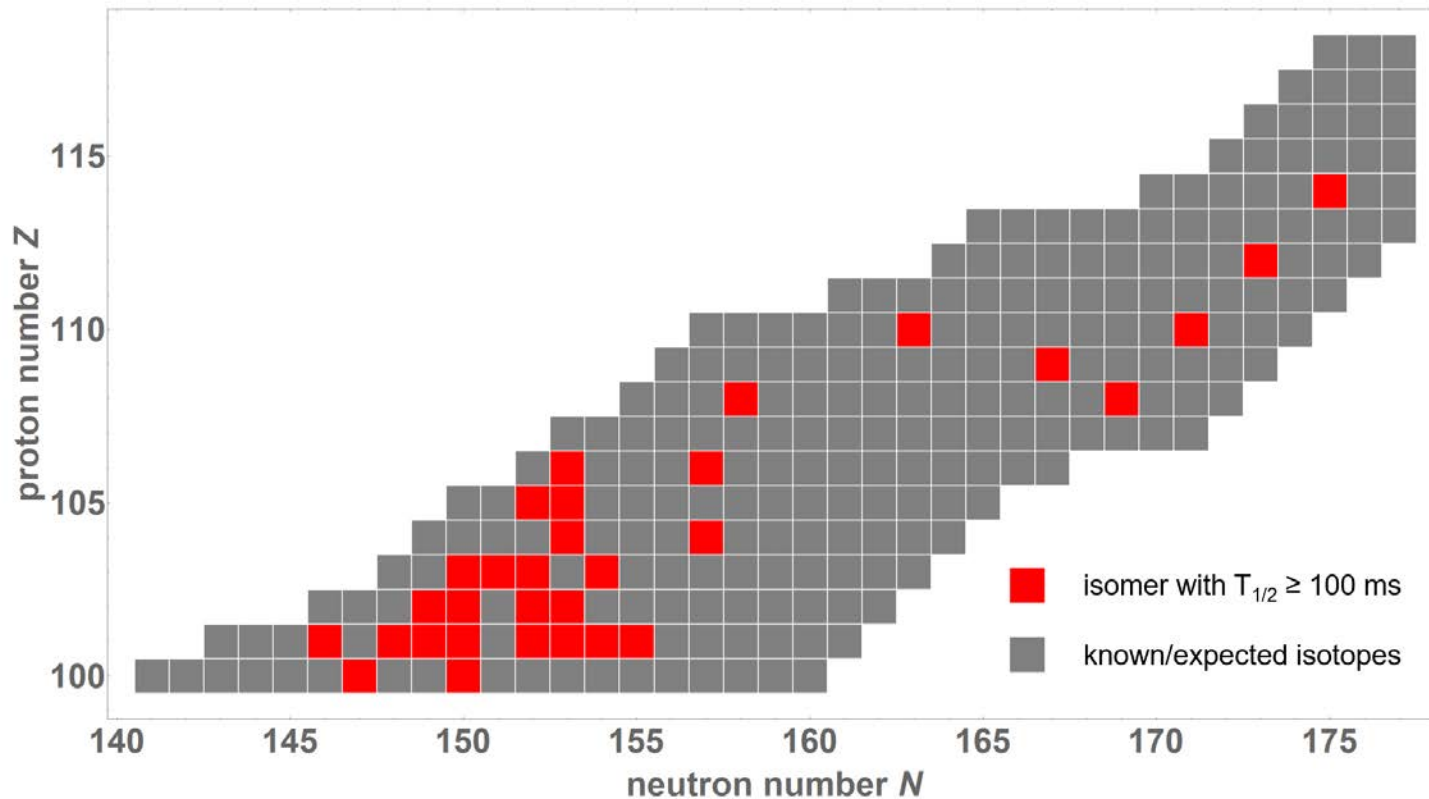
high-precision mass measurements provide

- accurate absolute binding energies
- anchor points to pin down decay chains
- options to identify (long-lived) isomers
- possible A/Q identification

- **study nuclear structure evolution far from stability**
- **benchmark and improve nuclear models**

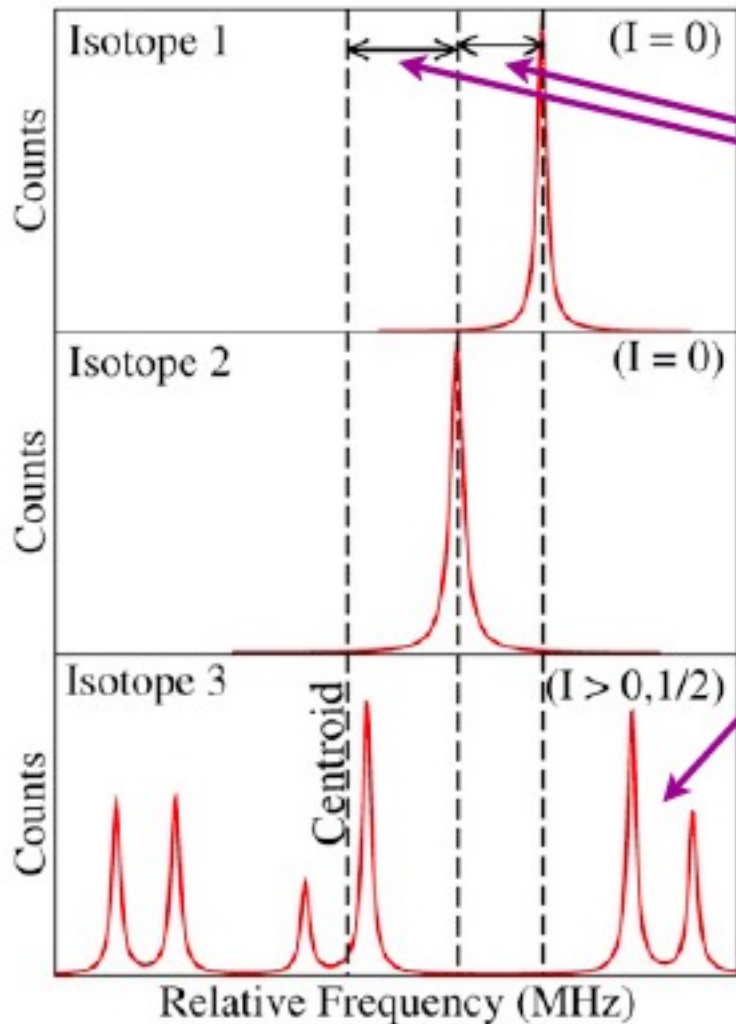


Long-Lived Isomers in the Heaviest Elements



- several (long-lived) isomeric states known, further may exist
- many of these are difficult to observe experimentally
- experiments also suffer from low yield
- Penning-trap mass spectrometry well suited to locate isomers that are low in energy and relatively long-lived

Obtaining Nuclear Properties



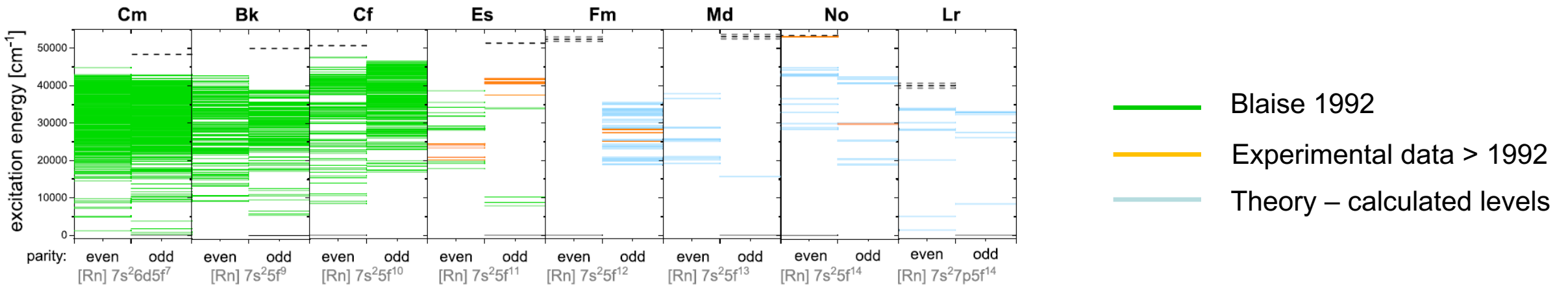
Isotope shift yields information on changes in mean-square charge radii from which we infer nuclear size

$$\delta \langle r^2 \rangle^{AA'} = \left(\underbrace{\Delta v^{AA'}}_{\text{Experiment}} - \frac{A - A'}{AA'} \underbrace{M}_{\text{Theory}} \right) \frac{1}{F}$$

hyperfine spectroscopy yields parameters linked to magnetic dipole moment and spectroscopic quadrupole moment

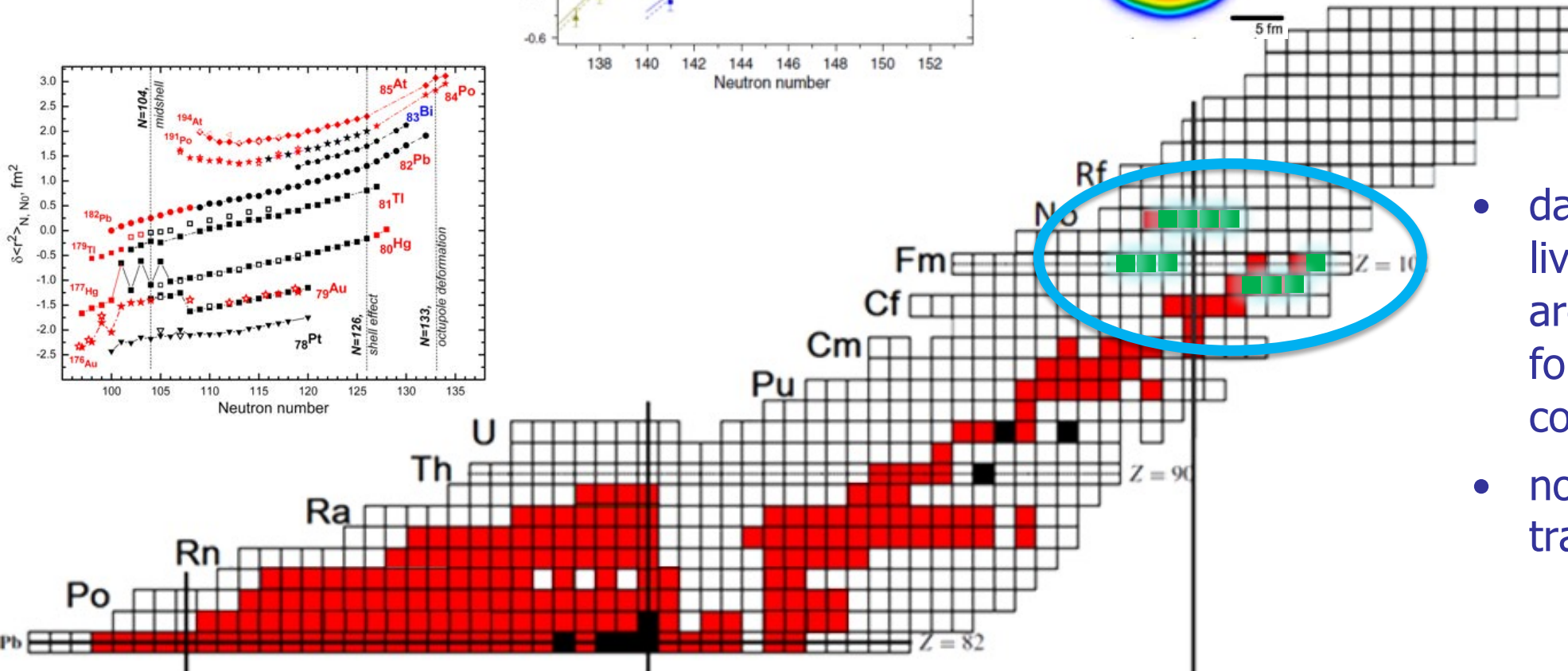
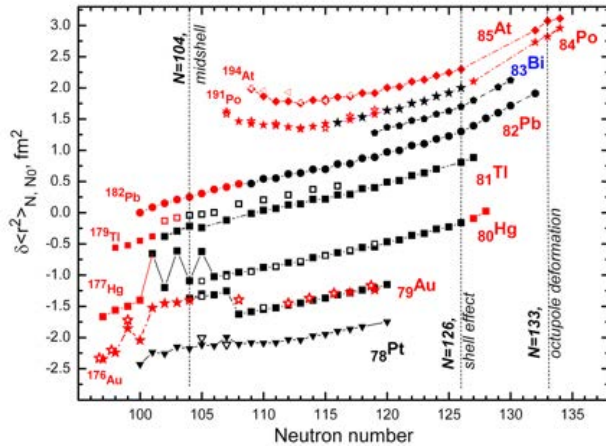
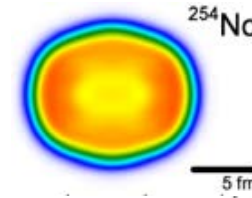
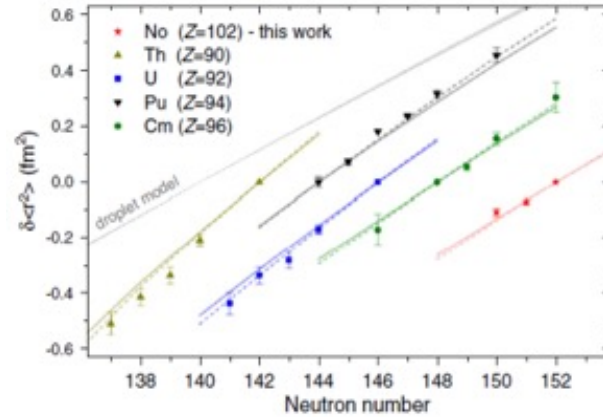
$$A = \underbrace{\mu}_{\text{Nuclear}} \frac{B_e(0)}{IJ} \quad B = eQ_s \underbrace{\left(\frac{\delta^2 V}{\delta z^2} \right)}_{\text{Atomic}}$$

Overview on Atomic Levels reported for Actinides



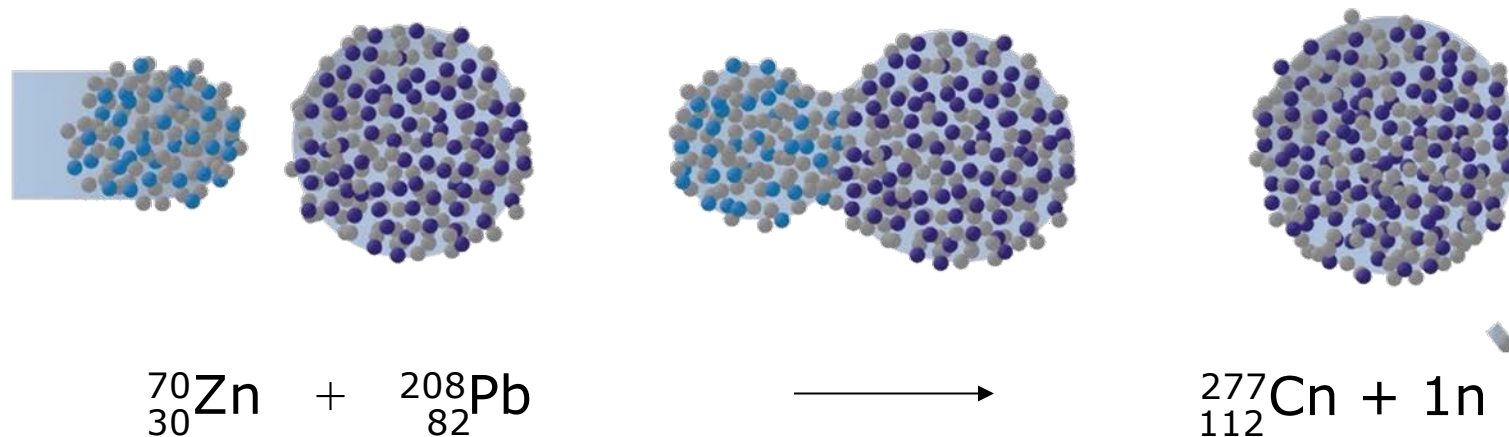
- complex atomic structure
- limited data for heavier actinide elements
- for heaviest elements often only calculations available

Status of Laser Spectroscopy in Heavy Elements



- data exists for some long-lived actinide isotopes that are available in quantities for offline studies with conventional methods
- no experimental data for transactinides

Production in Fusion-Evaporation Reactions



- presently only viable way to produce superheavy nuclides is by heavy-ion induced fusion-evaporation reactions
- requires high-intensity heavy-ion beams at Coulomb-barrier energies

Production of SHN by Fusion-Evaporation Reactions

Typical primary beams:

- ^{22}Ne , ..., ^{48}Ca , ^{50}Ti , ..., ^{70}Zn with intensities of 6×10^{12} projectiles/second

Common Targets:

- $^{204,206,207,208}\text{Pb}$, ^{209}Bi , ^{238}U , $^{242,244}\text{Pu}$, ^{243}Am , ^{248}Cm , ^{249}Bk , ^{249}Cf
- wheels rotating synchronously with beam-pulse structure
- Comprise several segments with about 0.5 mg/cm^2

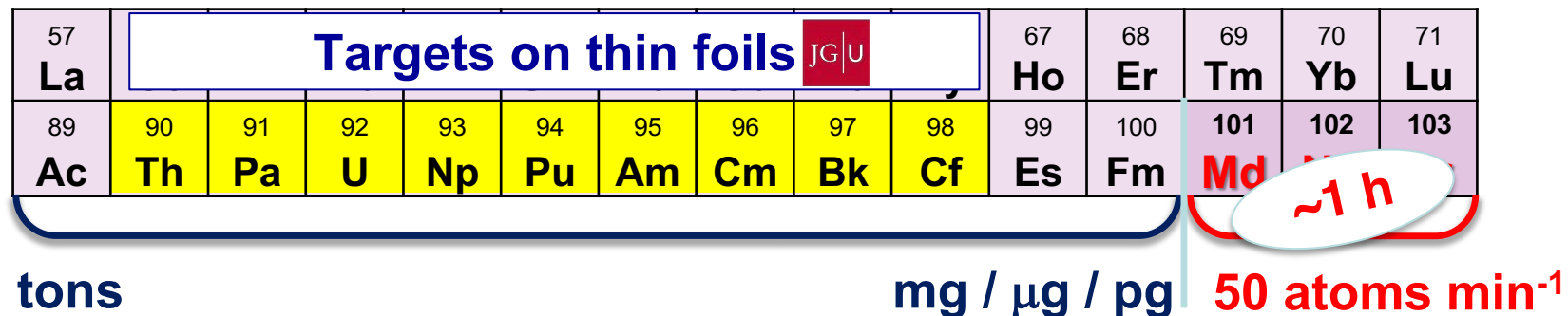
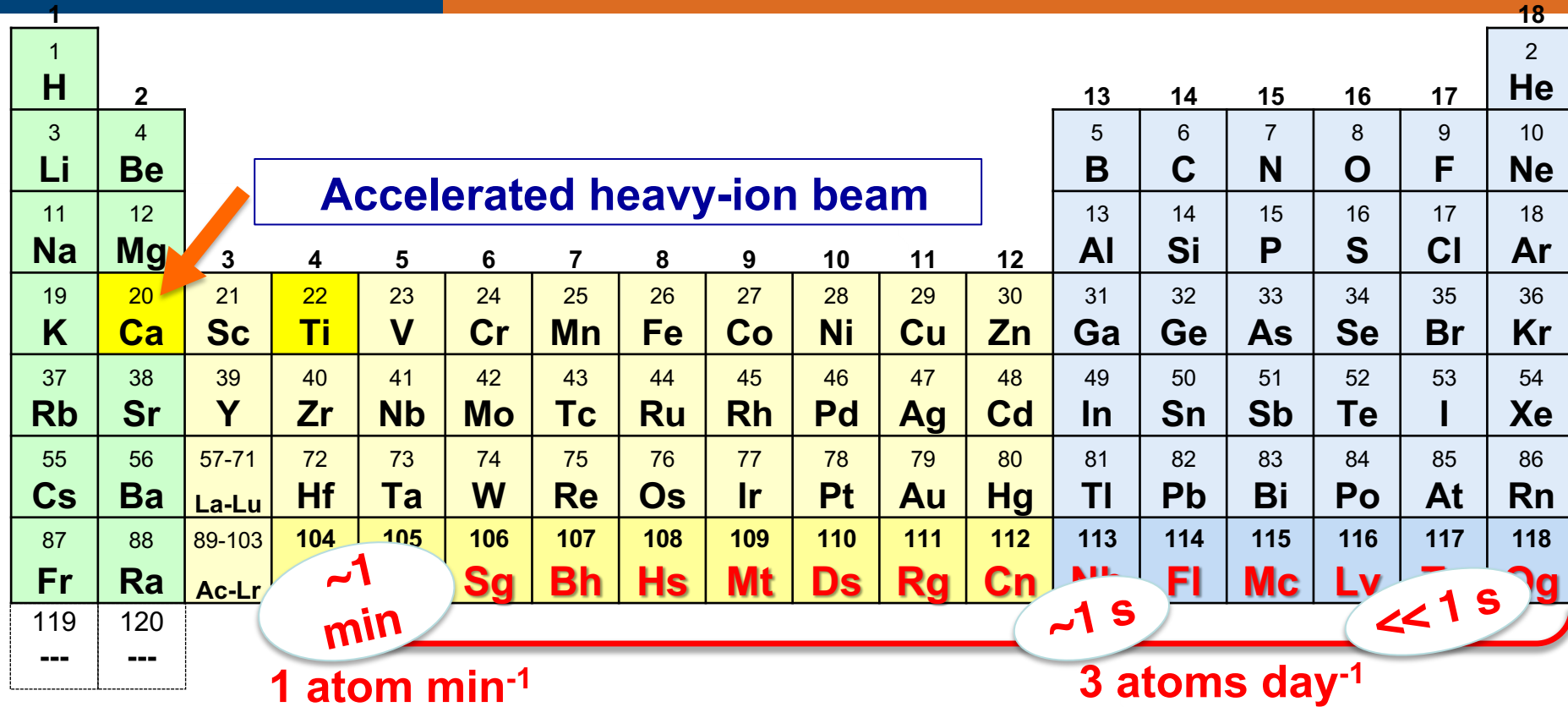
Challenges:

- long irradiation with high beam intensity damages targets
- limited availability of relevant actinide isotopes
- radioactive targets



TASCA ^{249}Bk target
produced at Mainz
Ch. Düllmann et al.

Production and study of superheavy elements



GSI / FAIR – The Universe in the Lab

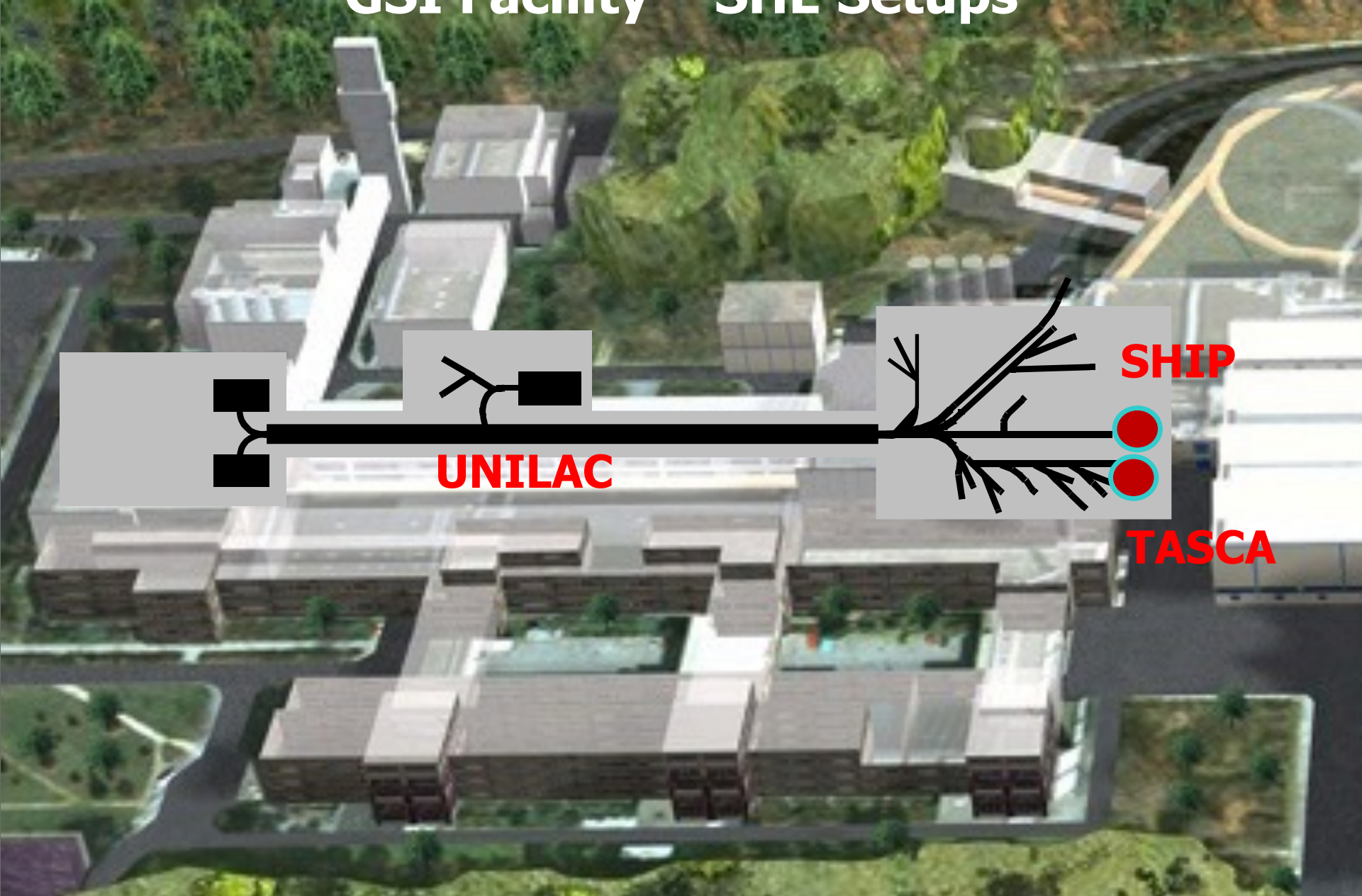
April 2021



June 2021



GSI Facility – SHE Setups



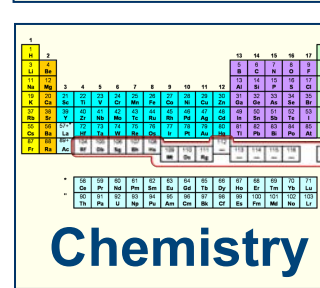
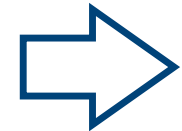
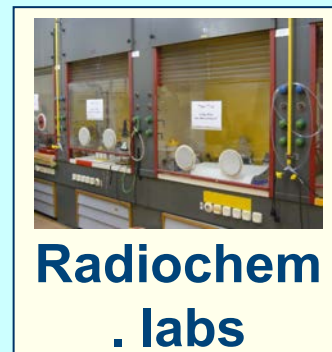
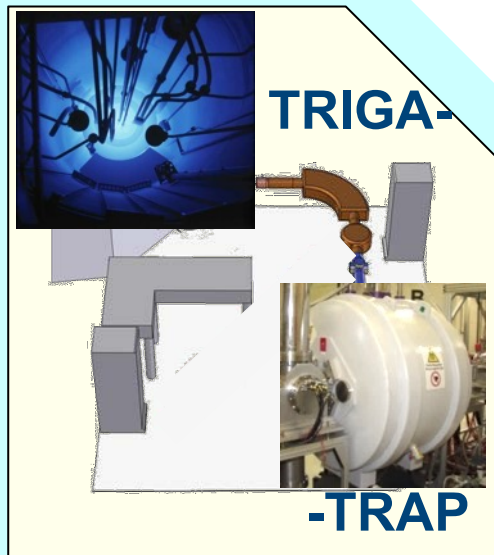
Unique Combination of Experimental Setups

GSII

HIM

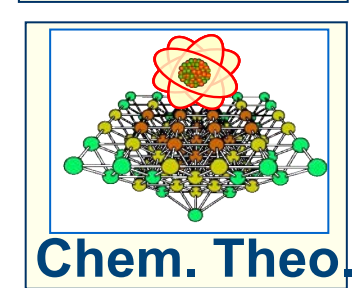
JG|U

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**Target
Lab**

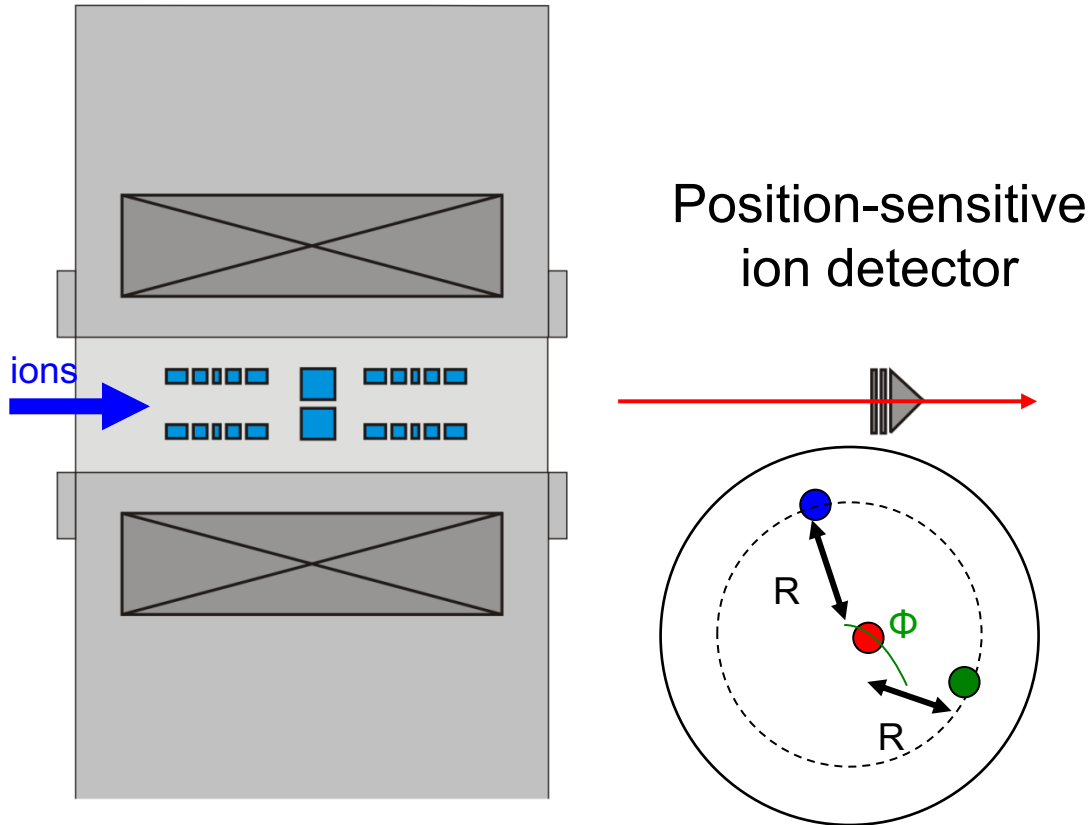
**Experiment
Electronics**



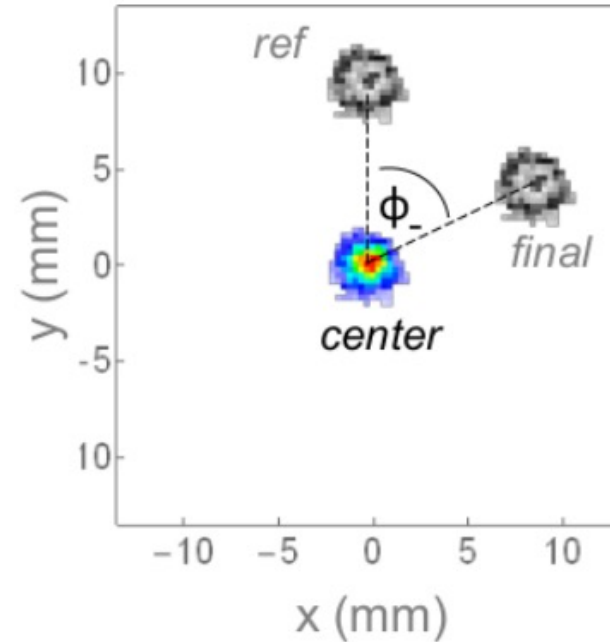
**Detector
Lab**



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



**pioneered at SHIPTRAP
- now used worldwide**



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

$$\phi + 2\pi n = 2\pi \nu t$$

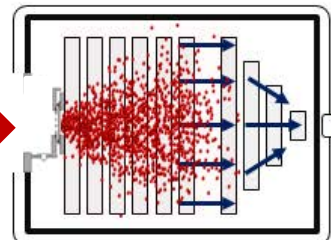
- determine phases of (radial) eigen motions to obtain cyclotron frequency
- improved resolving power / precision

S. Eliseev et al., Phys. Rev. Lett. 110, 082501 (2013)
S. Eliseev et al., Appl. Phys. B 114, 107 (2014)

SHIPTRAP Setup at GSI Darmstadt

$\approx 40 \text{ MeV}$

Cryogenic Gas-Stopping Cell



7.5 mbar He at 40 K

$\approx 1/s$
to 1/min

Extr. RFQ

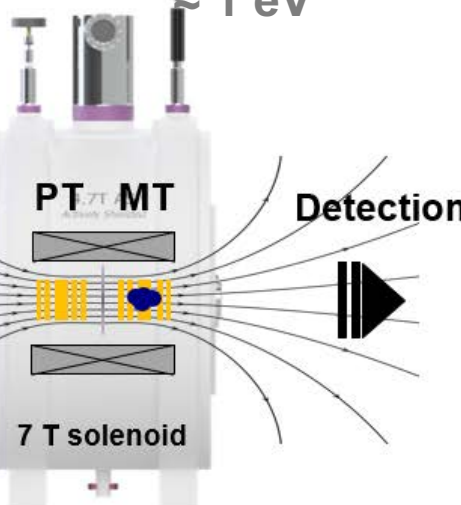
Buncher

10^{-3} mbar He

Surface-Ionization

Nd:YAG
Laser-Ablation

$\approx 1 \text{ eV}$



Detection

Stopping

Transport & Bunching

Measurement

magnetron phase image

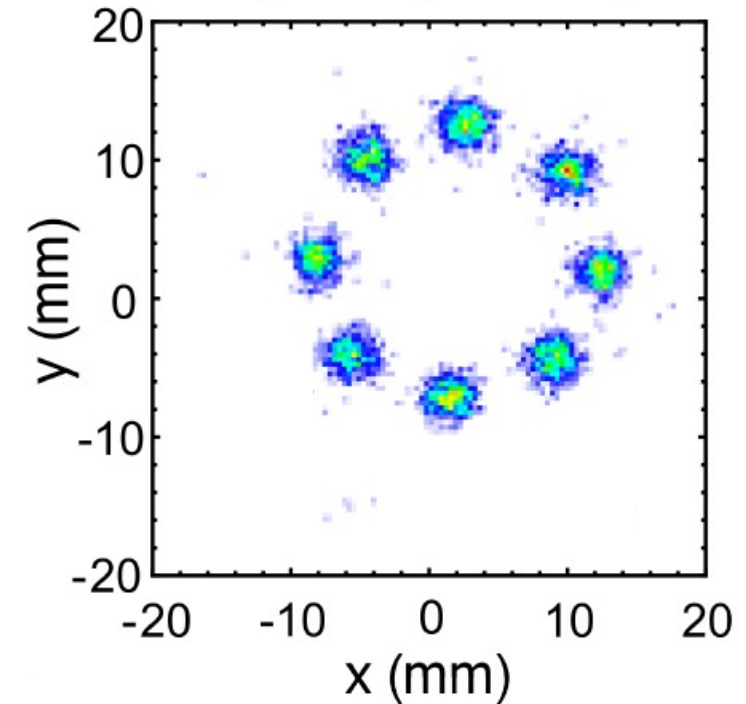
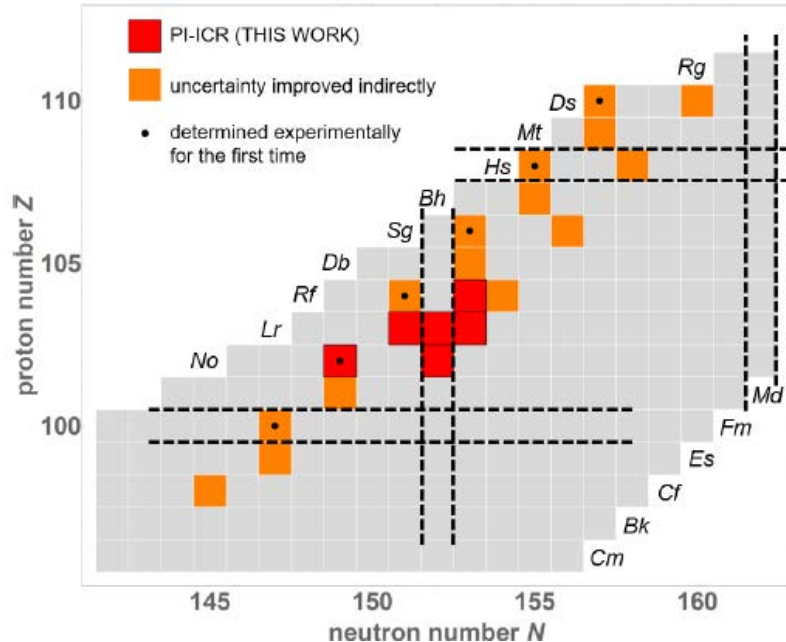


Photo: G. Otto, GSI



Nuclide Masses Obtained with SHIPTRAP

Isotope	Ions	Ratios	R_{mean}	rel. unc.	$R_{\text{ToF-ICR}}$	rel. unc.	$ME(\text{keV}/c^2)$	$ME_{\text{lit}}(\text{keV}/c^2)$	$ME_{\text{new}}(\text{keV}/c^2)$
^{251}No	39	9	0.944614687(9)	9.5E-9	-	-	82851.3(23)	82849(181)	82851.1(21)
^{254}No	2448	24	0.955908554(6)	6.3E-9	0.955908520(60) [22] 0.955908550(40) [23]	6.3E-8 4.2E-8	84733.5(15)	84723.3(97)	84733.3(15)
^{254}Lr	156	14	0.955928750(27)	2.8E-8	-	-	89734.0(67)	89645.9(913)	89733.9(64)
^{255}Lr	278	6	0.959691642(7)	7.3E-9	0.959691740(60) [23]	6.3E-8	89933.0(17)	89947.3(177)	89932.6(17)
^{256}Lr	124	11	0.963461017(23)	2.4E-8	0.9634610(3) [23]	3.1E-7	91737.2(57)	91746.6(829)	91737.2(57)
^{257}Rf	5	2	0.967240149(670)	6.9E-7	-	-	95960(170)	95866.4(108)	95866.4(108)



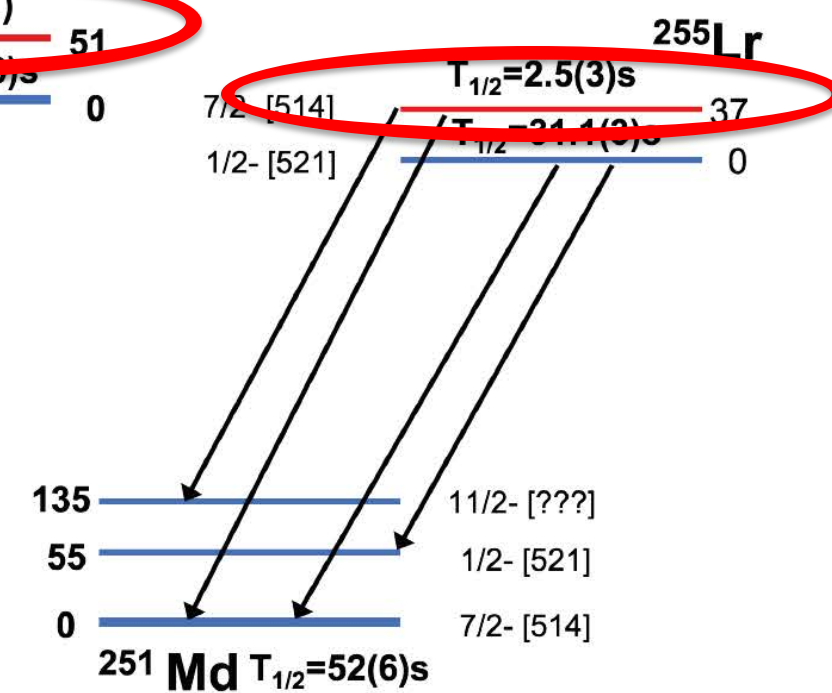
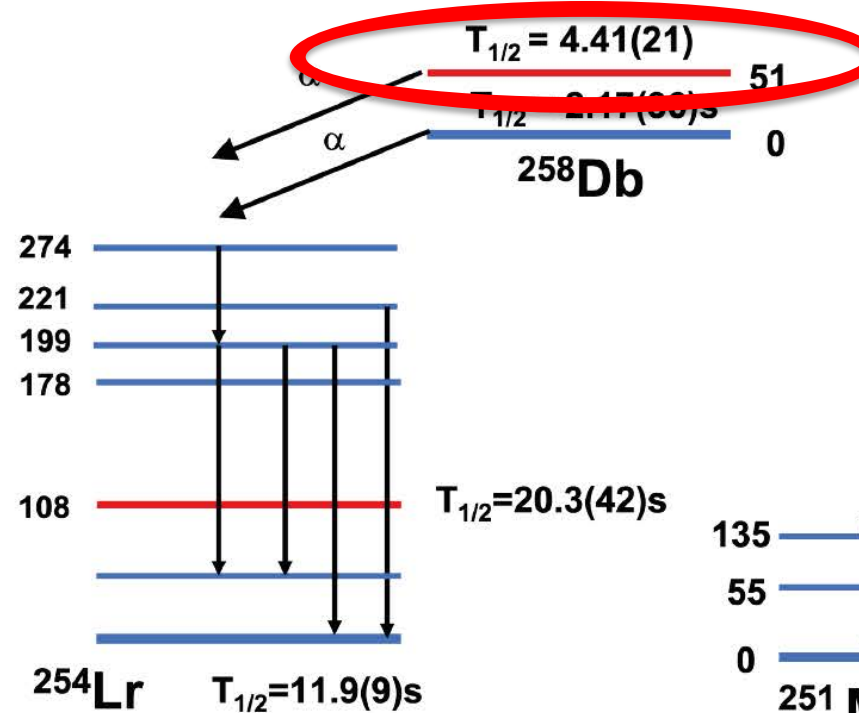
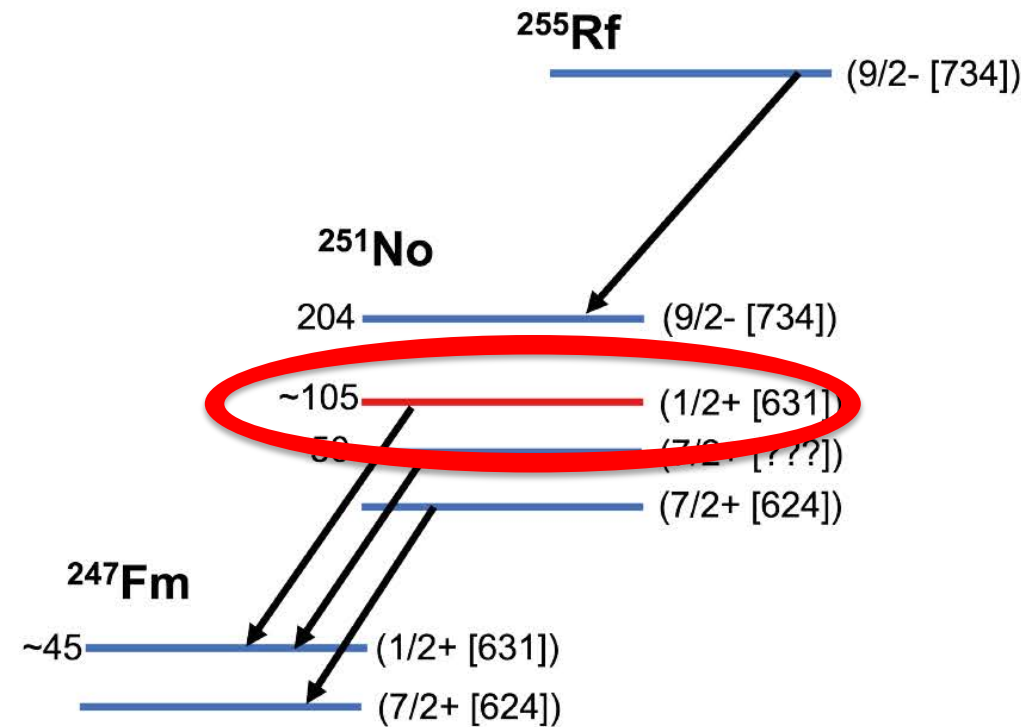
- data were implemented in AME network and showed good consistency
- many mass values improved directly and additional ones indirectly via the AME network's links

Information on Isomers from Decay Studies

^{251}No : Z=102, N=149

^{254}Lr : Z=103, N=151

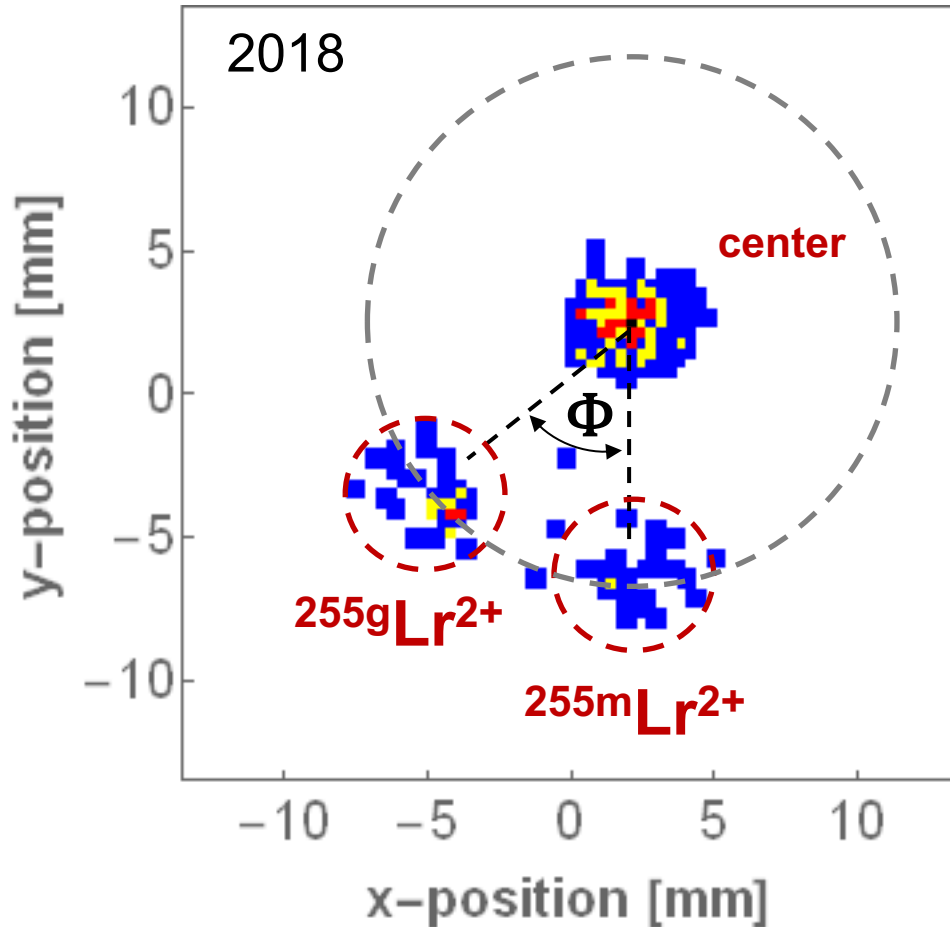
^{255}Lr : Z=103, N=152



Investigated with SHIPTRAP

- A. Chatillon *et al.*, *Eur. Phys. J. A* 30, 397 (2006)
- F.P. Hessberger *et al.*, *Eur. Phys. J. A* 30, 561 (2006)
- S. Antalic *et al.*, *Eur. Phys. J. A* 38, 219 (2008)

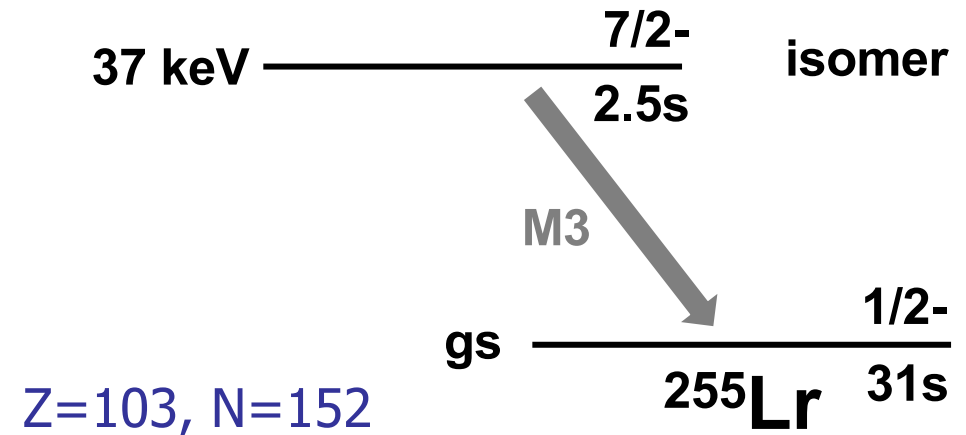
SHIPTRAP Results - Example $^{255(m)}\text{Lr}$



Work by decay spectroscopy

A. Chatillon *et al.*, *Eur. Phys. J. A* 30, 397–411 (2006)

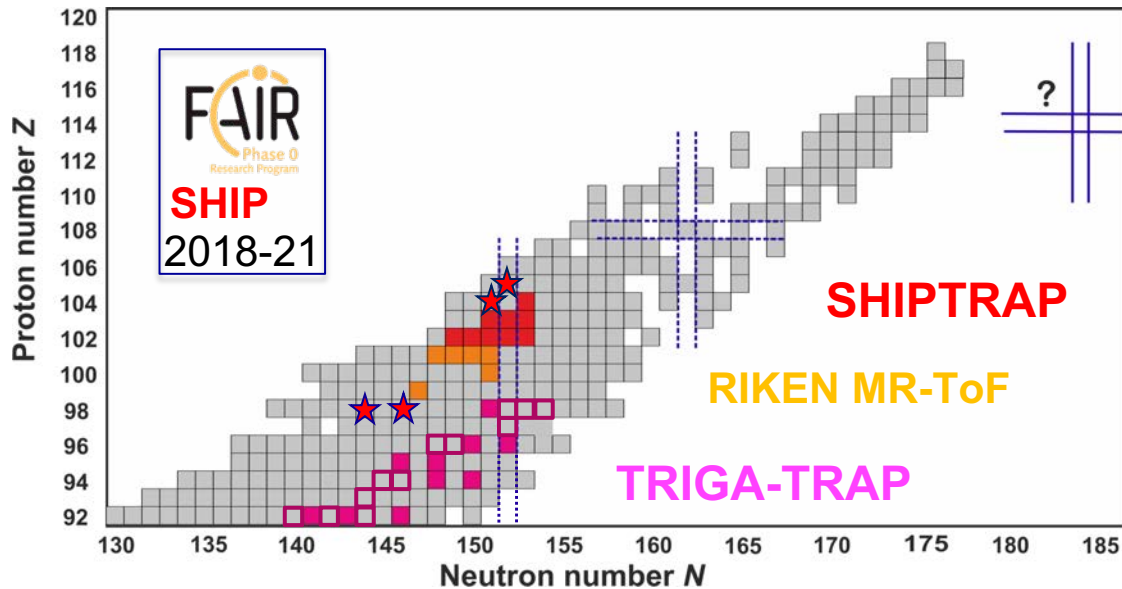
S. Antalic *et al.*, *Eur. Phys. J. A* 38, 219–226 (2008)



- Isomer known from decay spec. via difference of α energies
- SHIPTRAP resolved isomer with mass resolving power $m/\Delta m \approx 10^7$

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

Mass measurements of Actinides and Transactinides

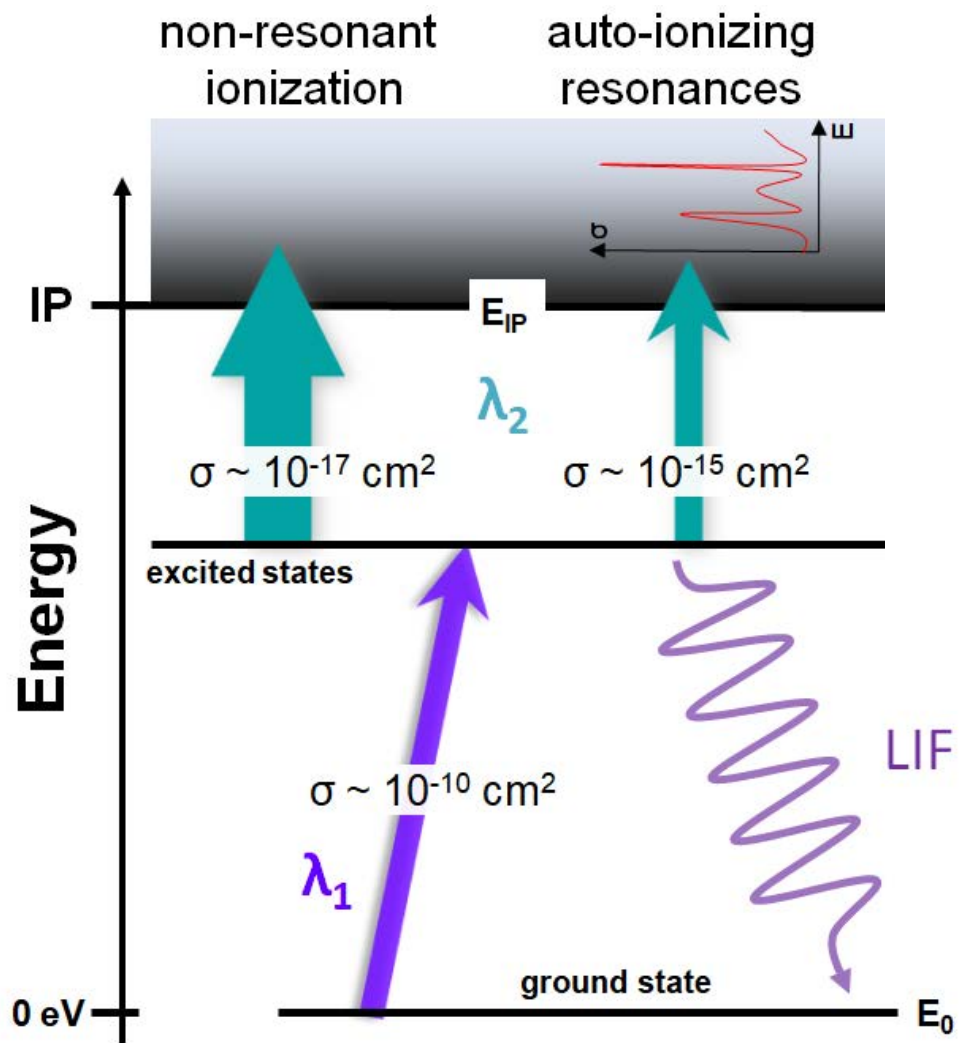


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- O. Kaleja, PhD thesis Uni Mainz 2020
- B. Andelic, PhD thesis Uni Groningen 2021
- E. Minaya Ramirez et al. Science 337, 1207 (2012)
- M. Block et al., Nature 463, 785 (2010)
- Y. Ito et al., Phys. Rev. Lett. 120, 152501 (2018)
- M. Eibach et al.,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

- SHIPTRAP performed first (direct) mass spectrometry beyond $Z = 100$ in 2008
- Recent mass measurements with rates of $\approx 0.00002/s$ and 5 detected ions in total
- rel. mass uncertainty down to a few 10^{-9}
- $m/\Delta m = 11,000,000$ for unambiguous identification of long-lived low-lying isomers
- extended measurements to ^{258}Db ($Z=105$)

Principles of Resonant Ionization Laser Spectroscopy



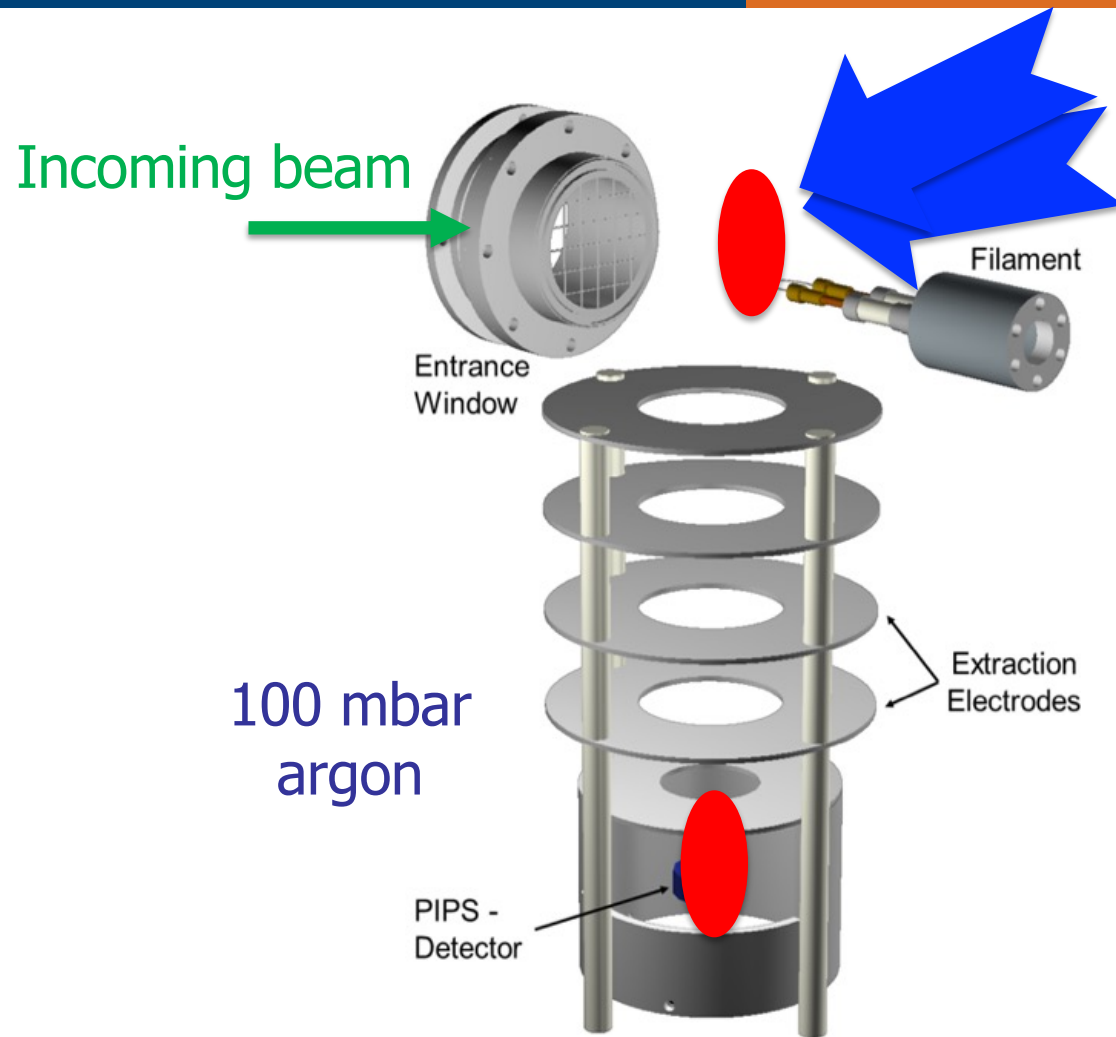
method of choice is for studies of rare isotopes

- ion detection more efficient than fluorescence photon detection
- low-background conditions, particularly for detection via decay
- sensitive method applied in ultra-trace analysis and in laser-ion sources

Challenge for the heaviest elements:

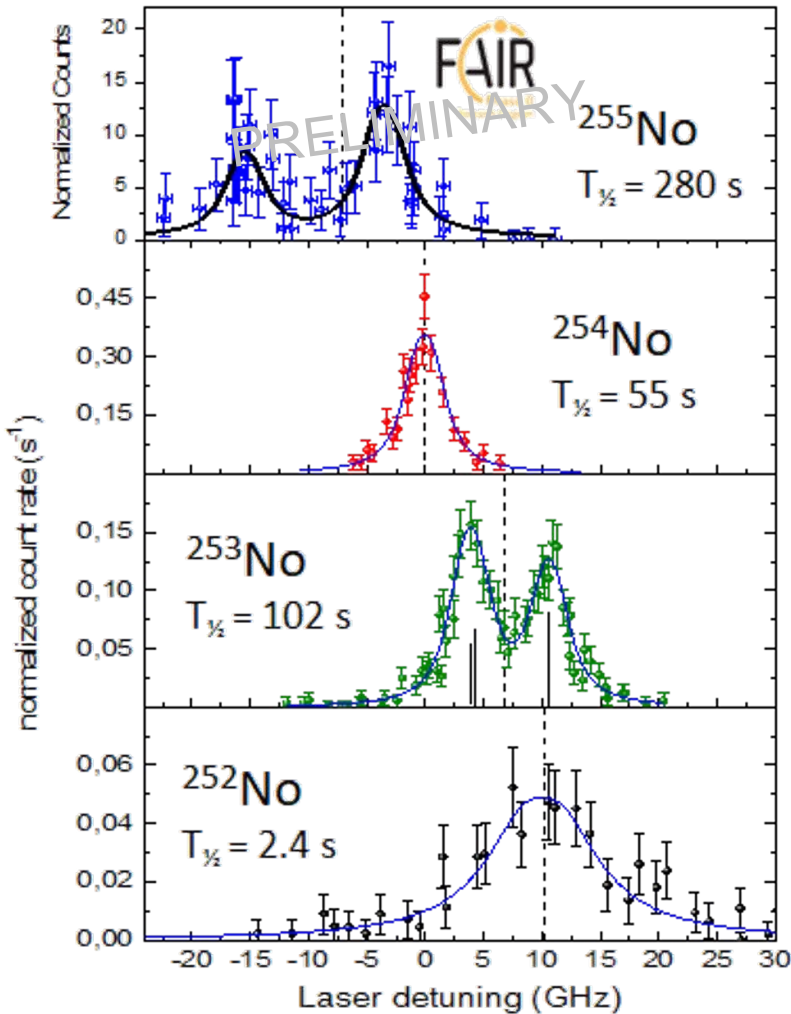
- finding states for excitation schemes
- low yield

Radiation Detected Resonance Ionization Spectroscopy (RADRIS)



- RADRIS method tailored to actinides produced by fusion with lowest rates
- slow down and neutralize in Ar gas
- evaporate atoms
- two-step photo-ionization
- transport to detector
- register radioactive decay

Laser Spectroscopy of Nobelium (Z=102) Isotopes



- first laser spectroscopy spectroscopy beyond Z=100
- yield as low as 0.05 atoms / second
- isotope shift allowed determining changes in mean-square charge radii around $N = 152$
- magnetic dipole and electric quadrupole moment of ^{253,255}No obtained from hyperfine splitting

Experiment: S. Raeder, M. Laatiaoui *et al.*

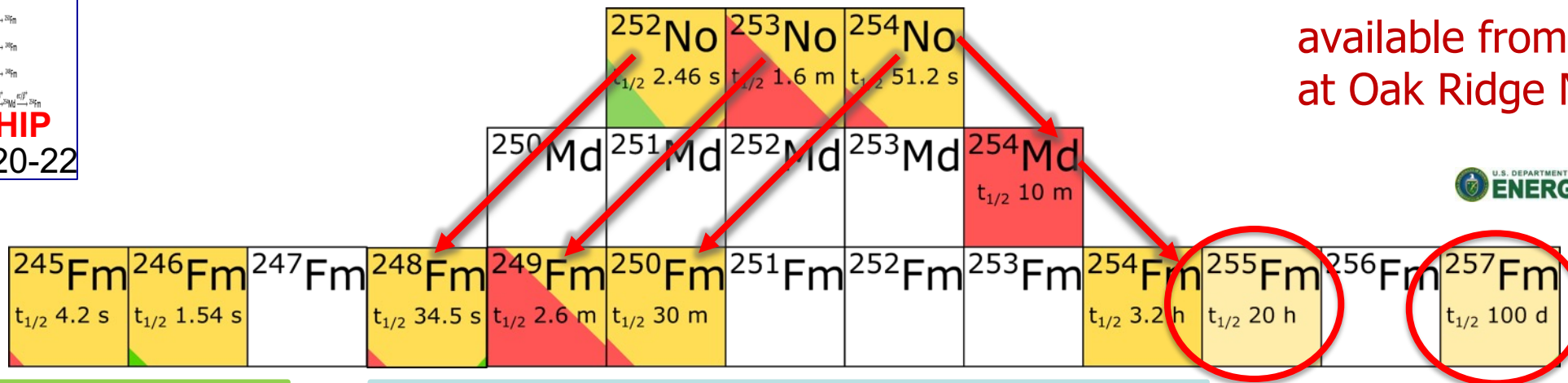
Theory: A. Borschevsky V. Dzuba, S. Fritzsche,
B. Schütrumpf, W. Nazarewicz *et al.*

M. Laatiaoui *et al.*, Nature 538, 495 (2016)

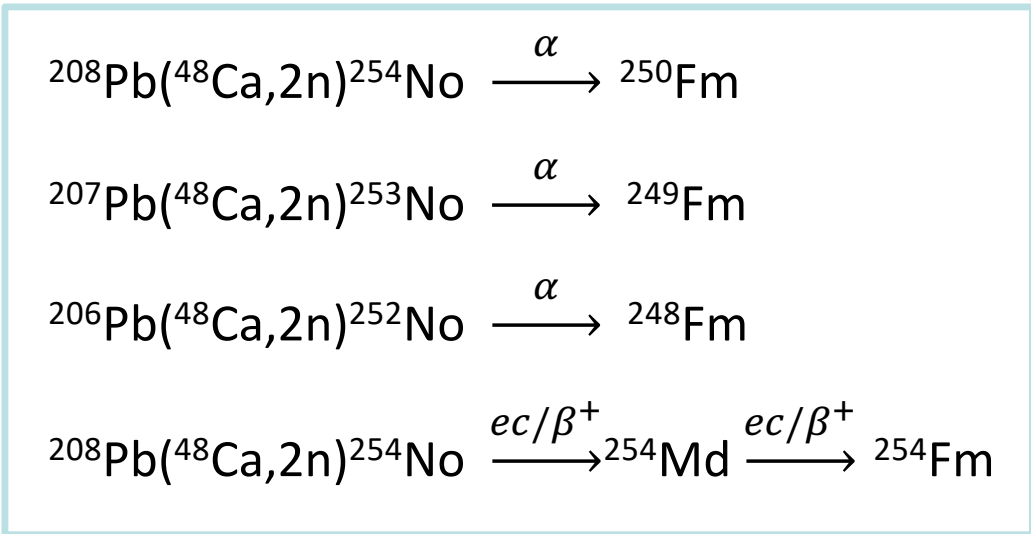
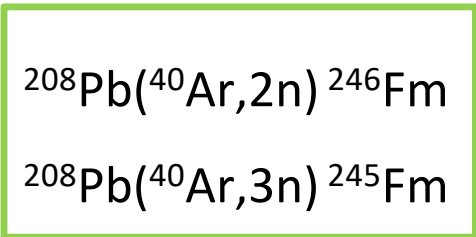
S. Raeder *et al.*, Phys. Rev. Lett. 120 (2018) 232503

Production of Fermium (Z=100) Isotopes

SHIP
20-22



available from HFIR reactor
at Oak Ridge National Lab



- direct production of certain Fm isotopes difficult
- production of Fm isotopes via decay of No isotopes on filament
- challenge: long half-lives

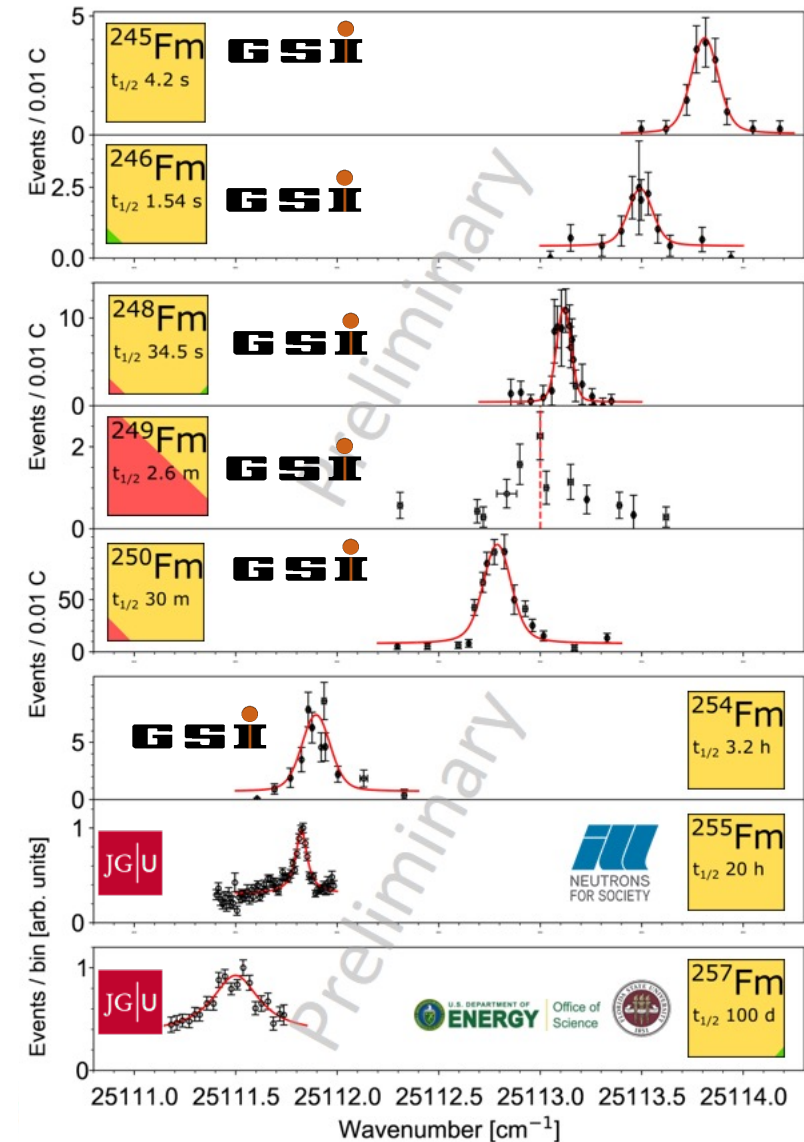
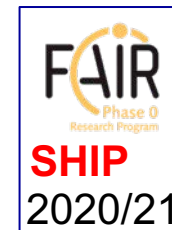
Laser Spectroscopy of Fm Isotopes

- short-lived Fm isotopes measured online at GSI
- produced via decay of directly produced No isotopes
- long-lived isotope $^{255,257}\text{Fm}$ from ORNL / ILL measured at RISIKO after radiochemical separation by Mainz nuclear chemistry (Ch. Düllmann et al.)
- isotope shift measured in Fm isotope chain allows determination of changes in mean-square charge radii around $N = 152$

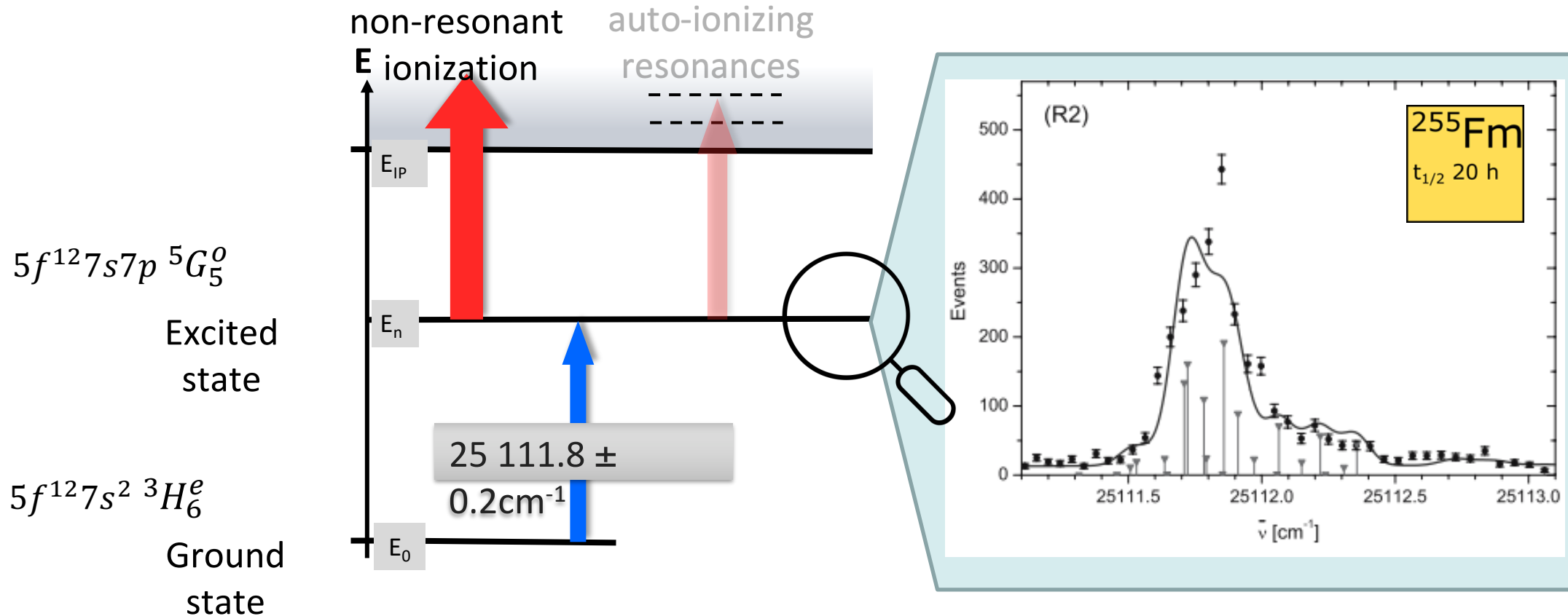
Experiment S. Raeder (GSI) *et al.*

Data analysis: S. Raeder, J. Warbinek, E. Rickert

The isotopes used in this research were supplied by the U.S. Department of Energy, Office of Science, by the Isotope Program in the Office of Nuclear Physics. The $^{253,254,255}\text{Es}$ and $^{255,257}\text{Fm}$ were provided to Florida State University and the University of Mainz via the Isotope Development and Production for Research and Applications Program through the Radiochemical Engineering and Development Center at Oak Ridge National Laboratory.



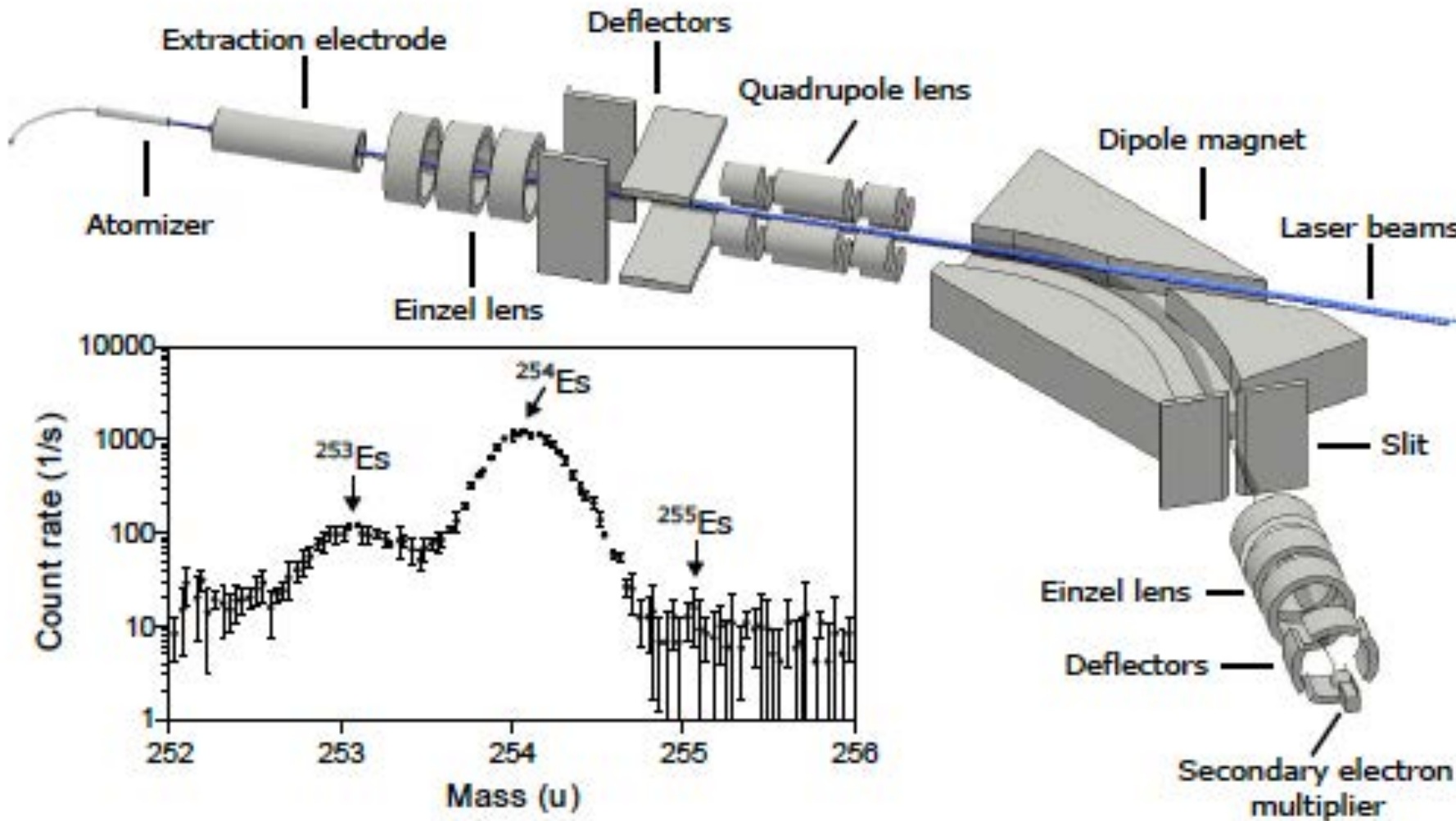
Atomic Levels in Fermium – Previous Work



M. Sewtz et al., *Physical Review Letters* 90 (2003).
 H. Backe et al., *Hyperfine Interactions* 162 (2005).

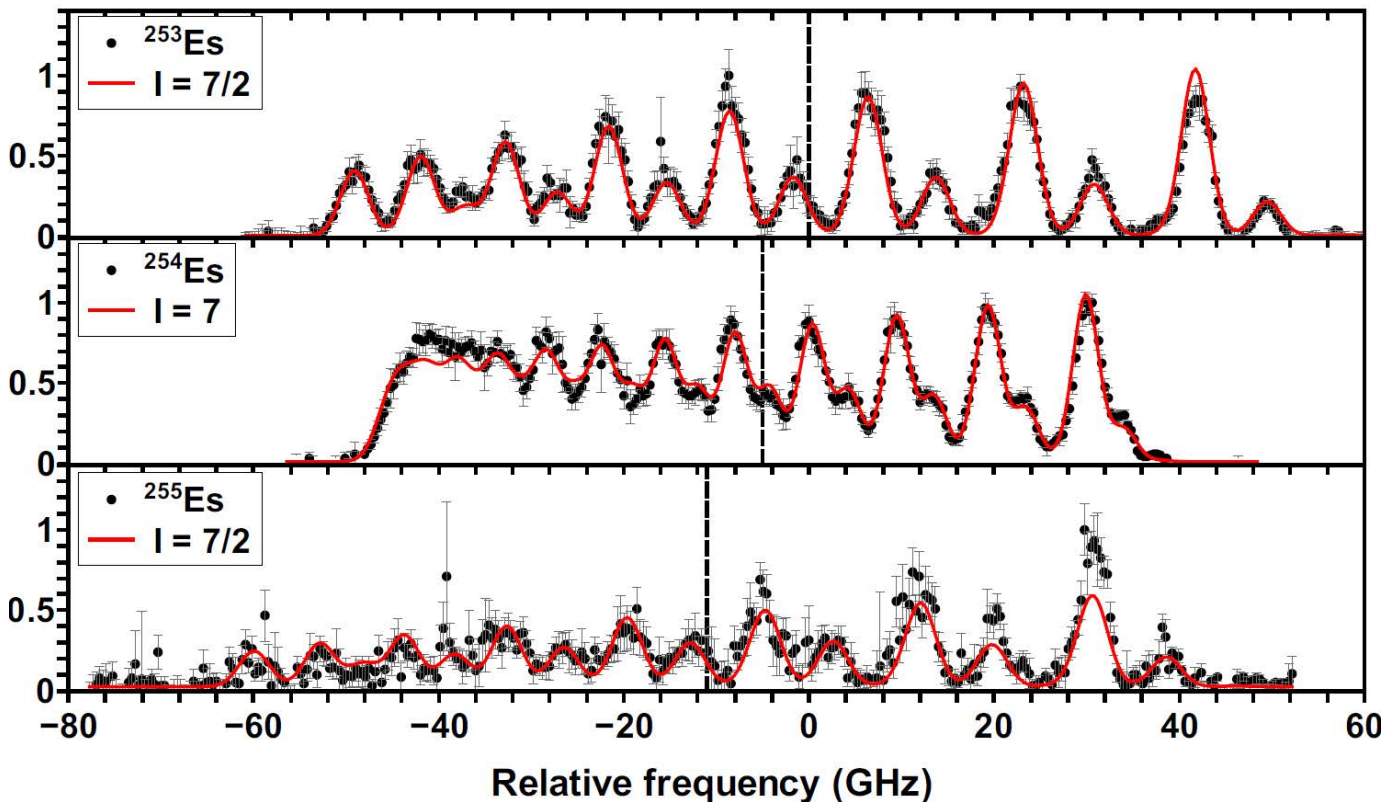
Hyperfine Spectroscopy of Es at RISIKO/Mainz

RISIKO separator
K. Wendt et al.



- Laser ionization
In hot cavity
- mass-separated
ion detection

Hyperfine Spectroscopy of Es at RISIKO/Mainz



S. Nothhelfer Phys. Rev. C 105, L021302 (2022)

- Hyperfine measurements with picogram to femtogram samples
- complex hyperfine structure investigated for several transitions in $^{253-255}\text{Es}$
- spins and electromagnetic moments can be obtained from hyperfine parameters

The isotopes used in this research were supplied by the U.S. Department of Energy, Office of Science, by the Isotope Program in the Office of Nuclear Physics. The $^{253,254,255}\text{Es}$ and $^{255,257}\text{Fm}$ were provided to Florida State University and the University of Mainz via the Isotope Development and Production for Research and Applications Program through the Radiochemical Engineering and Development Center at Oak Ridge National Laboratory.

S. Raeder, K. Wendt, Ch. Düllmann,
T. Albrecht-Schönzart, MB et al.



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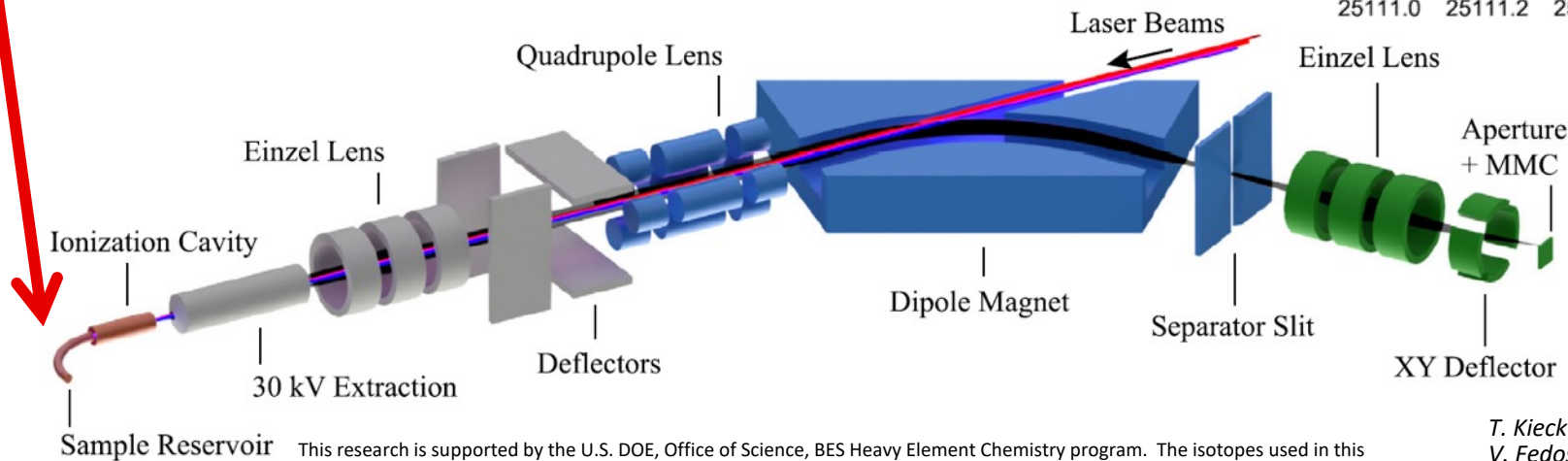
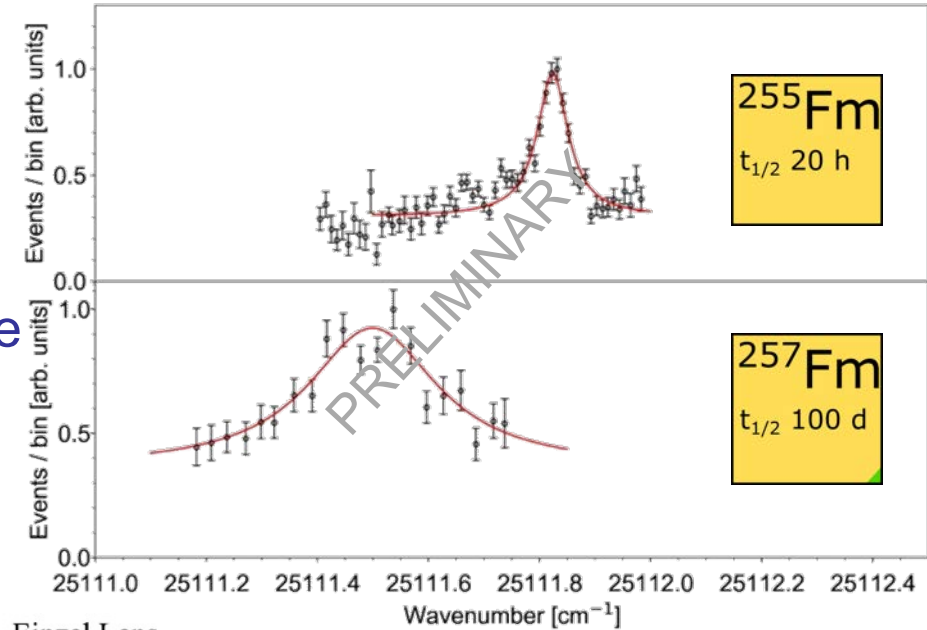


Laser Spectroscopy of Fm at RISIKO/Mainz



RISIKO mass separator in Mainz

- Production of radioactive ion beams
- Laser spectroscopy with high efficiency
- Resolution limited by source temperature and laser bandwidth



This research is supported by the U.S. DOE, Office of Science, BES Heavy Element Chemistry program. The isotopes used in this research were supplied by the U.S. DOE Isotope Program, managed by the Office of Science for Nuclear Physics.

T. Kieck et al., *NIM A* 945, 162602 (2019).

V. Fedosseev et al., *J. Phys. G Nucl. Part. Phys.* 44, 084006 (2017).

Summary and Conclusions

- novel techniques and tailored variants of laser spectroscopy have extended the reach to heavy nuclides with $Z > 99$ despite lowest yields
- laser spectroscopy of several Es, Fm, and No isotopes provided new data to study the impact of the nuclear shell at $N = 152$ on the nuclear structure
- Mass measurements up to ^{258}Db allowed us to perform a complementary study via the binding energies
- High-resolution mass spectrometry with Penning traps was used to investigate low-lying long-lived isomers in very heavy nuclei
- paved the way for comprehensive studies of nuclear structure evolution and atomic physics in heavy elements

THANK YOU FOR YOUR ATTENTION!