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Exploring the heaviest elements through laser spectroscopy and mass spectrometry

Michael Block

GSI Darmstadt, Helmholtz Institute Mainz, University Mainz

60th Bormio Conference – January 2024

Acknowledgements

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

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U.S. DEPARTMENT OF ENERGY | Office of Science

FLORIDA STATE UNIVERSITY 1851

EL EM EN TS

LISA
LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

GEFÖRDERT VOM
Bundesministerium
für Bildung
und Forschung

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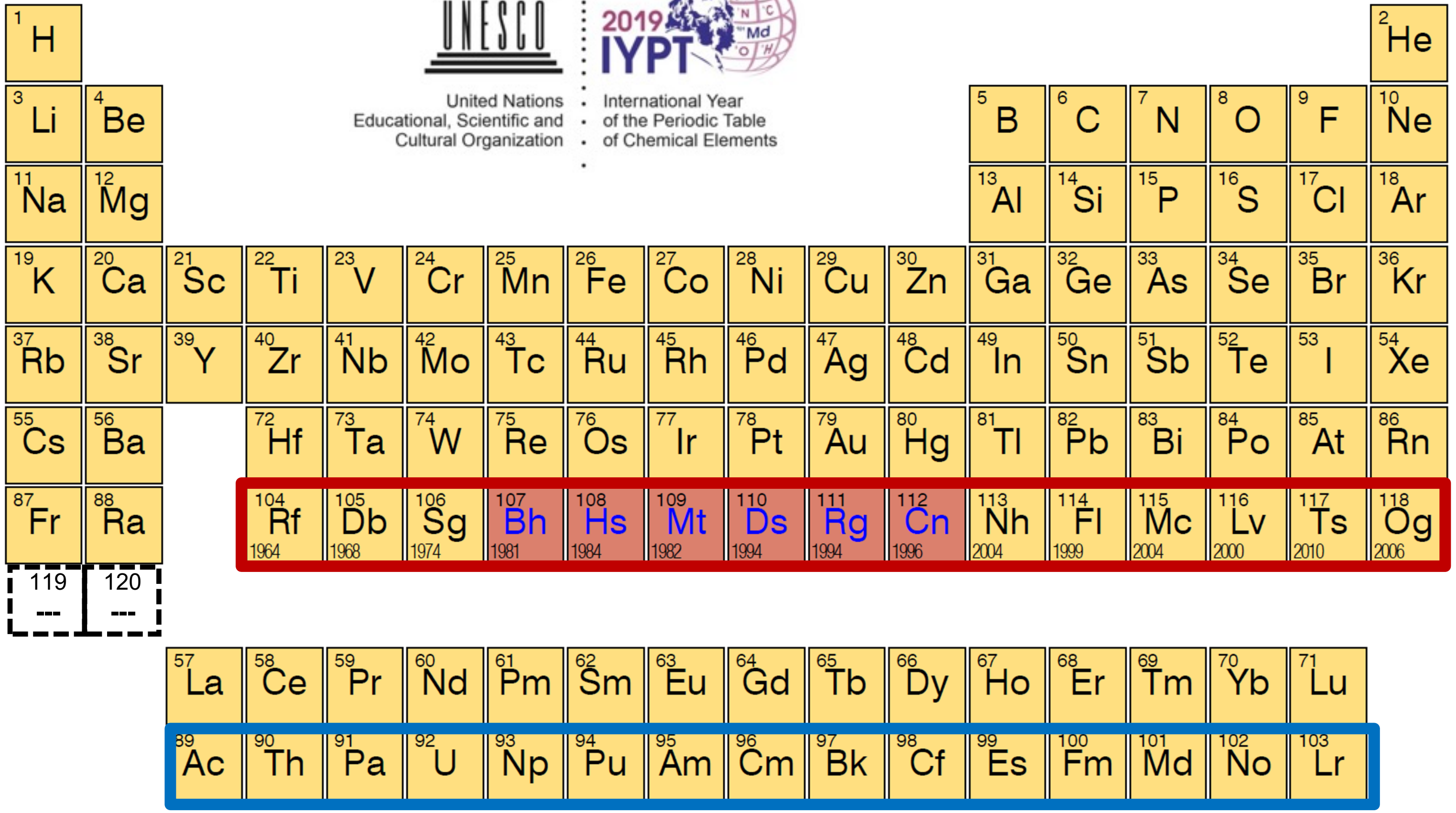
FAIR
Phase 0
Research Program

Outline

- Status and open questions in superheavy element research
- Production of (super)heavy nuclei
- Differential charge radii of fermium and nobelium isotopes from laser spectroscopy at GSI and influence of nuclear shell structure
- Mass measurements of superheavy nuclides with SHIPTRAP/GSI
- Summary and conclusions

The Periodic Table of 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	

United Nations
Educational, Scientific and
Cultural Organization

2019 IYPT
International Year
of the Periodic Table
of Chemical Elements

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

Superheavy Elements
= Transactinide Elements

Actinide Elements

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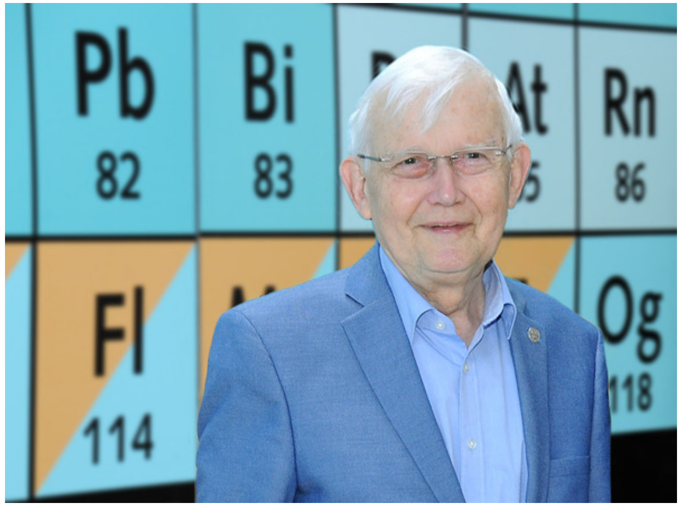
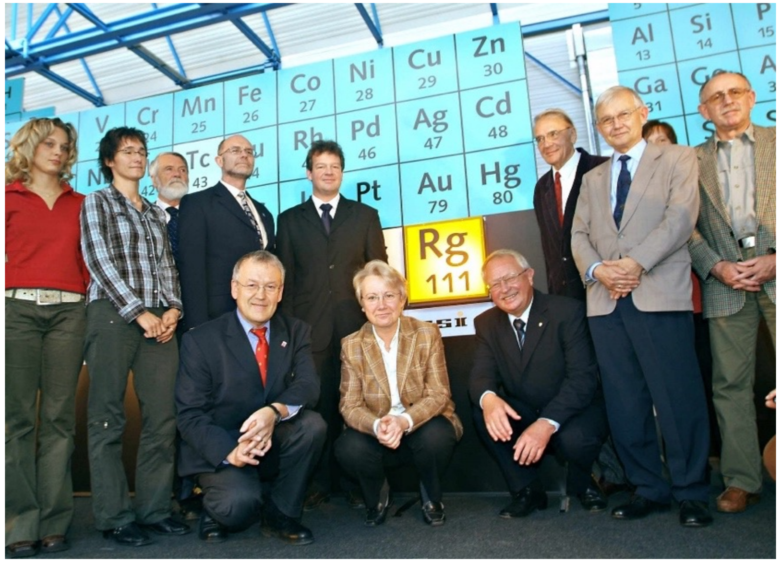
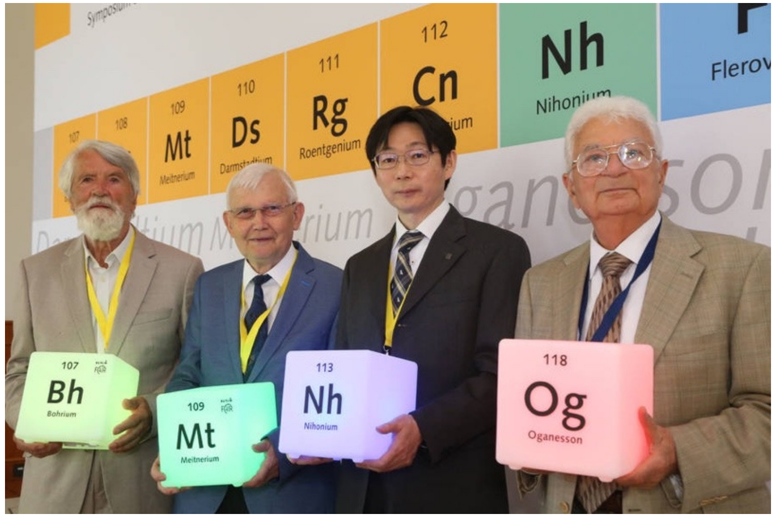
Gottfried Münzenberg (1940 - 2024)







- PhD at Giessen University with Heinz Ewald
- 1996: Professor at University of Mainz
- Department leader at GSI
- Important role in construction of SHIP and FRS
- (Co-)discoverer of new elements at GSI/SHIP
- (Co-)discoverer of many new isotopes at GSI/FRS

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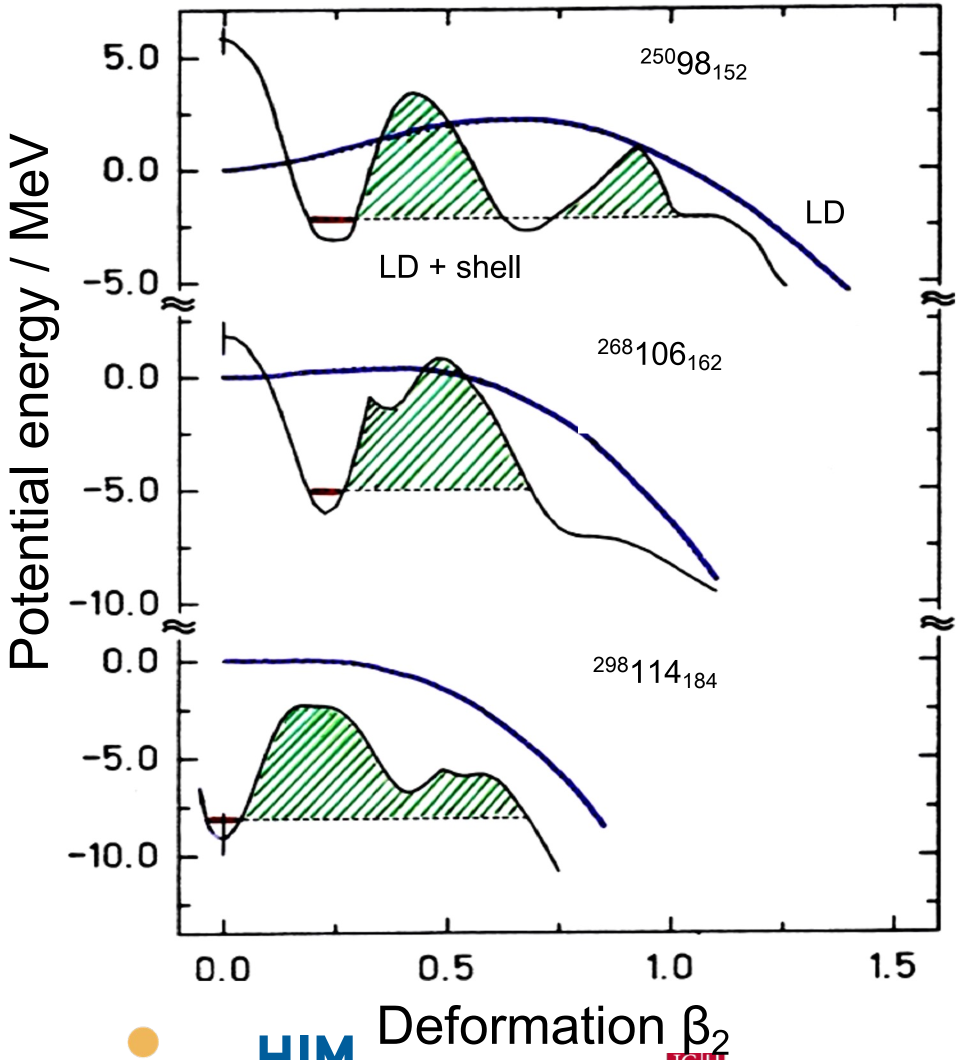
Fission Barriers in Superheavy Nuclei

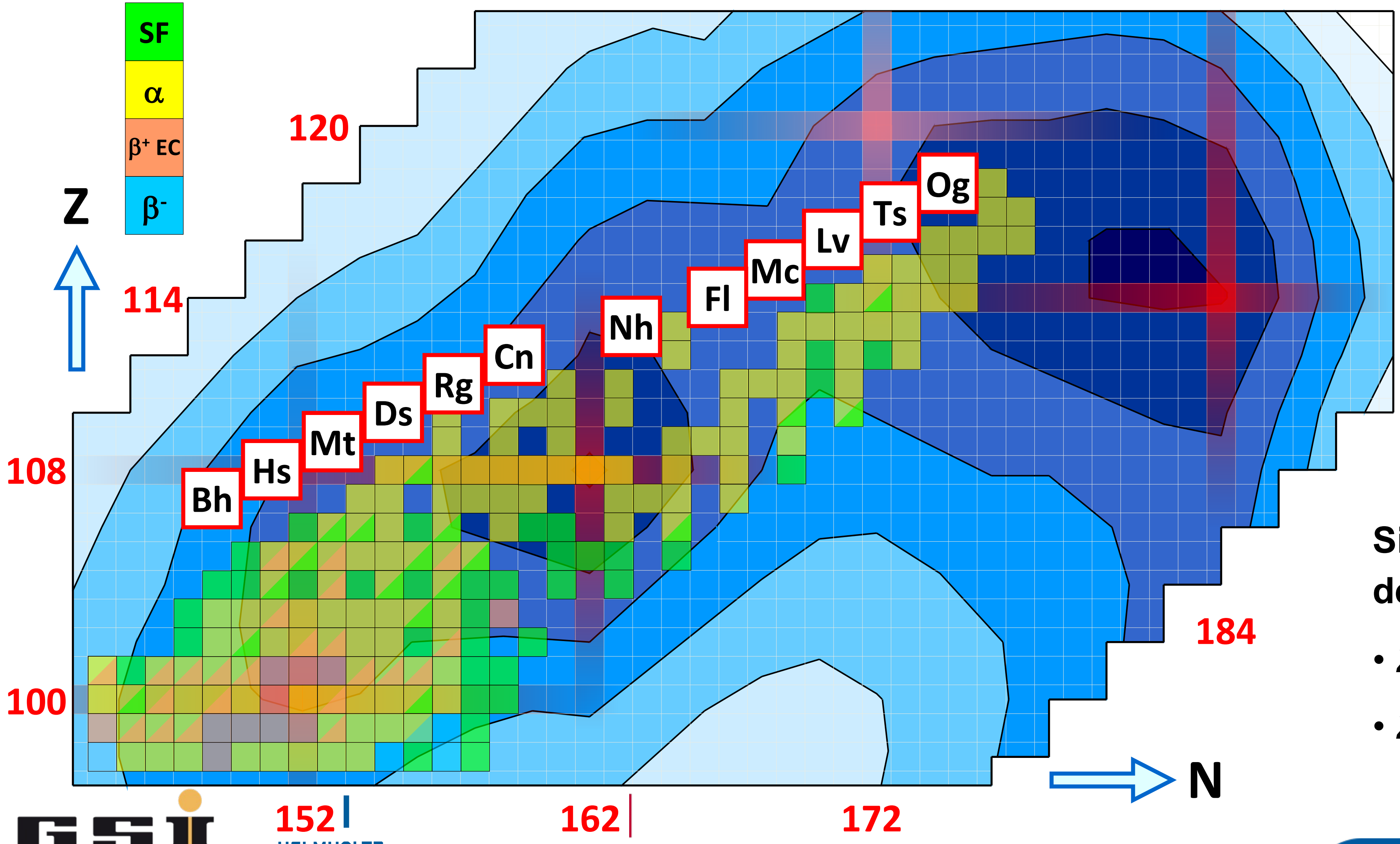


- fission barrier decreases with increasing Z
- liquid drop barrier vanishes around $Z = 106$
- superheavy nuclei (SHN) gain up to 10 MeV in binding energy by nuclear shell effects
- leads to finite fission barrier in SHN with $Z > 106$

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

Calculations by A. Sobiczewski, figure courtesy of S. Hofmann

Superheavy Nuclei – Shell Correction Energies



Sizeable shell gaps found for deformed nuclei around:

- $Z = 100$ and $N = 152$
- $Z = 108$ and $N = 162$

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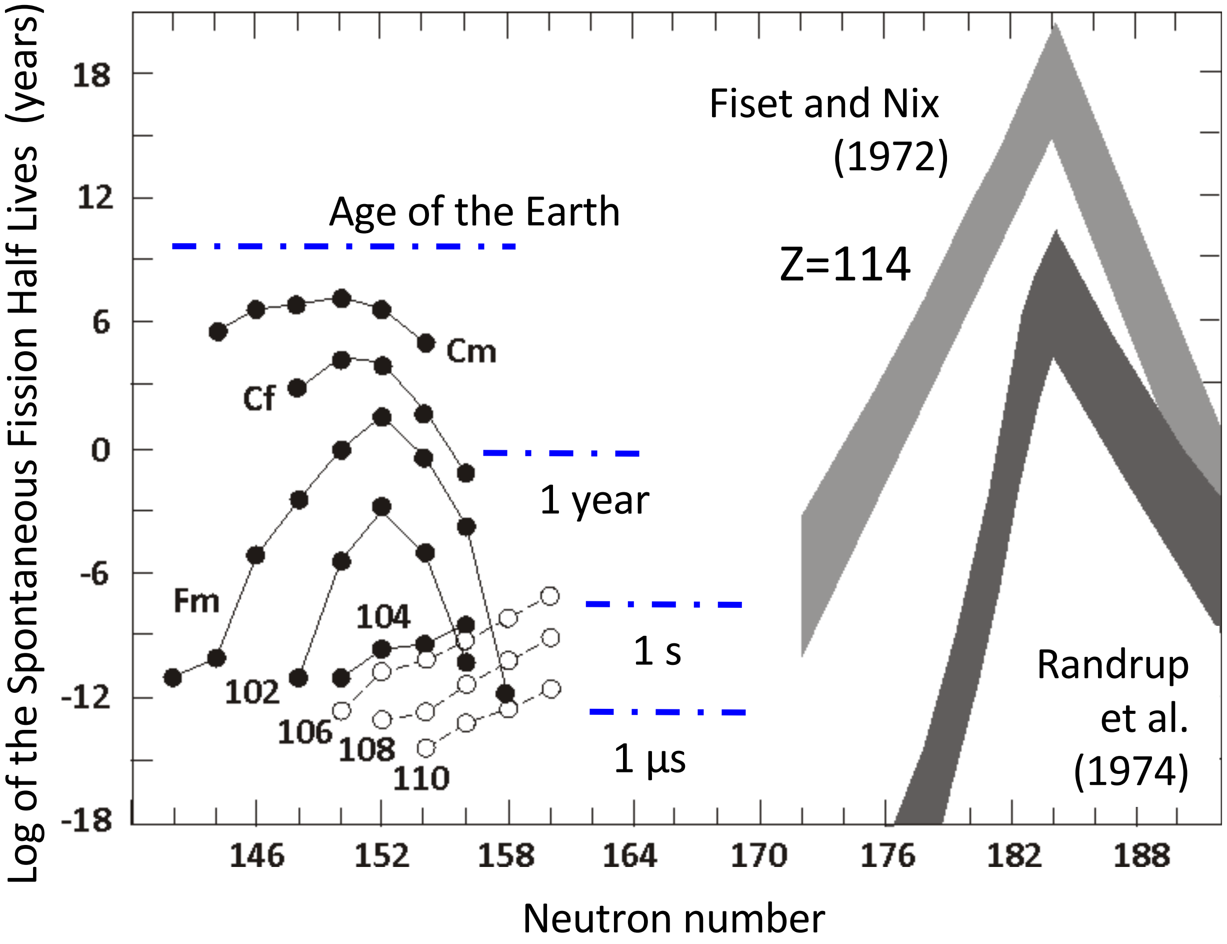
Superheavy Element Research – Key Questions

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

See e.g. recent reviews by
O. Smits et al. Nat. Rev. Phys. (2024), https://doi.org/10.1038/s42254-023-00668-y
S.A. Giuliani et al., Rev. Mod. Phys. 91, 011001 (2019)
and special/topical issues on SHE in Nucl. Phys. A 944 (2015) and Eur. Phys. J. A

Superheavy Nuclei – Island of Stability



- Superheavy nuclei with $Z \approx 114$ and $N \approx 184$ form

  „Island of Stability“

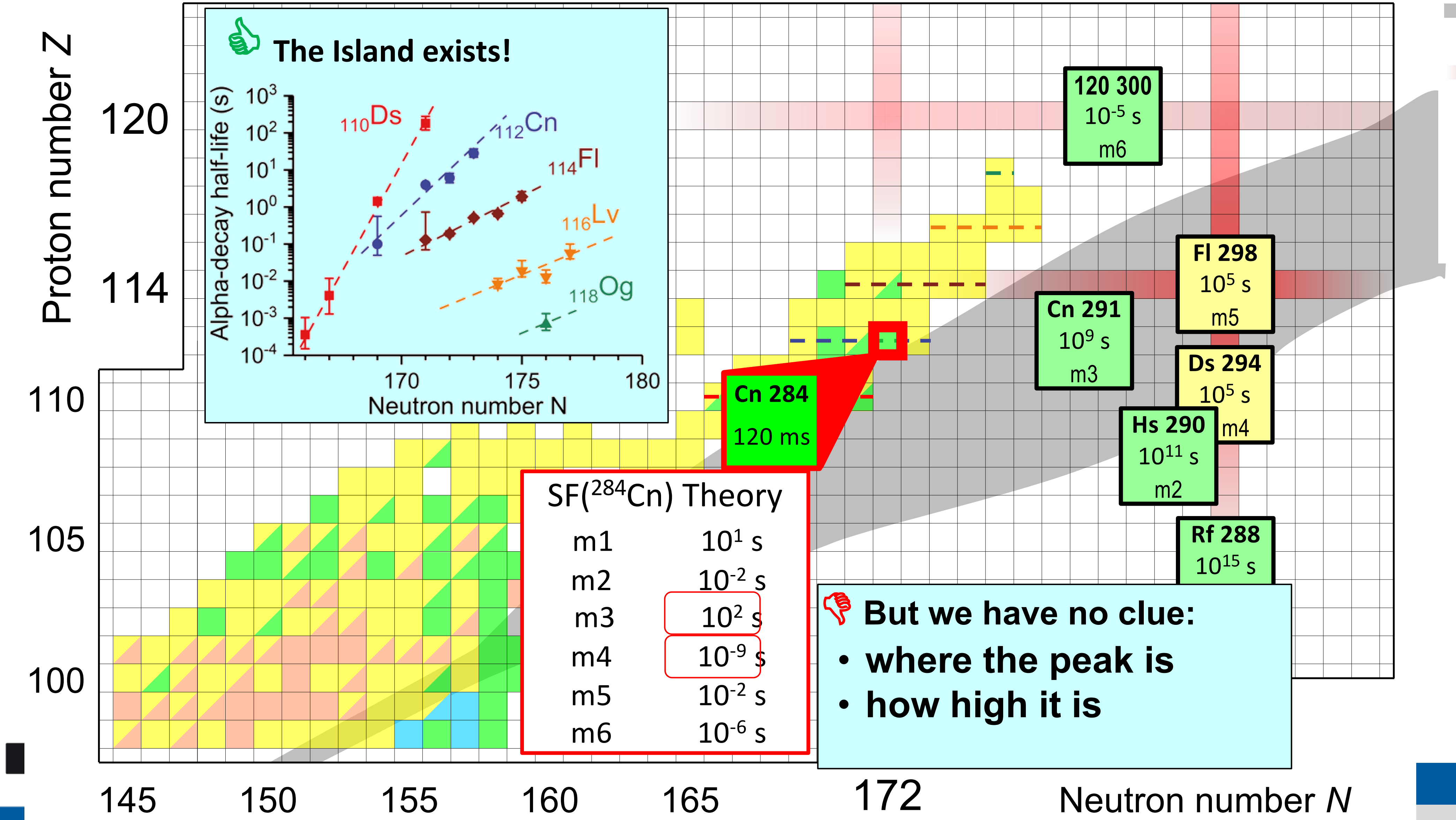
- in 1970s predicitons:

  $T_{1/2}(SF) > 10^9$ years

- initiate search for SHN in nature – until now no evidence

Figure Courtesy Yuri Oganessian

Island of Stability – Status 2023



The Island exists!

SF(^{284}Cn) Theory

m1	10^{1} s
m2	10^{-2} s
m3	10^{2} s
m4	10^{-9} s
m5	10^{-2} s
m6	10^{-6} s

But we have no clue:

- **where the peak is**
- **how high it is**

β-stability line

Often predicted spherical shells

Known nuclei decaying by

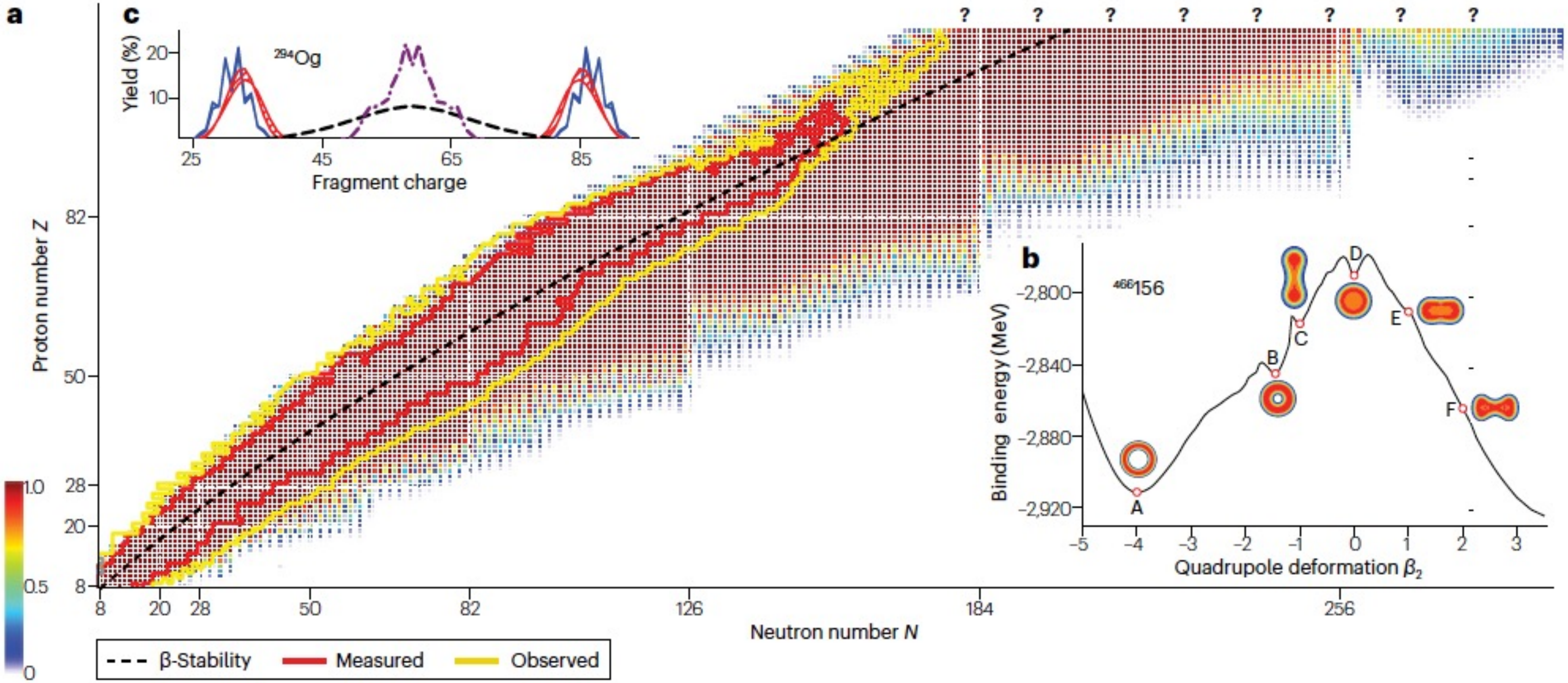
🟨	α-decay
🟦	β⁻-decay
🟧	β⁺/EC-decay
🟩	SF

Predicted longest-lived nuclei decaying by

🟨	α-decay
🟩	β⁻-decay

O. Smis et al,
Nat. Rev. Phys.
(2023)

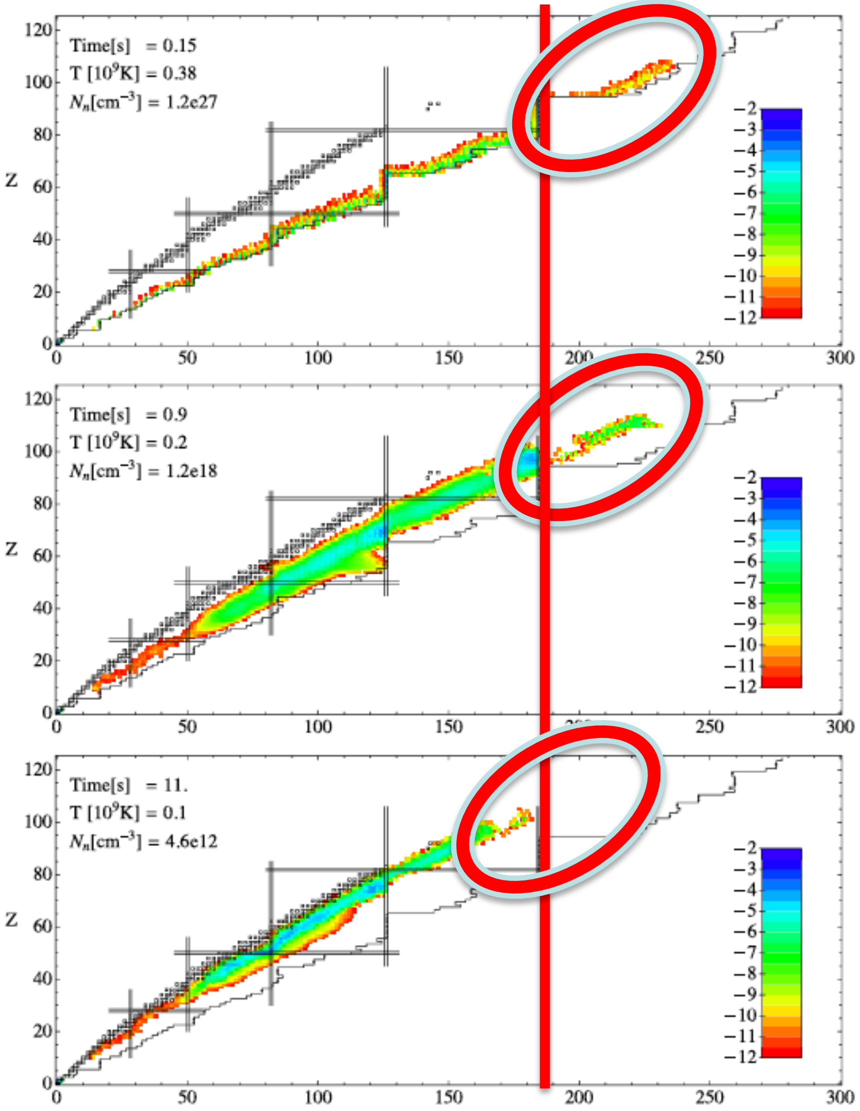
Nuclear Chart – Limits of Nuclear Stability



- Many more superheavy nuclei expected to exist
- Exotic topologies expected to appear

O. Smis et al, Nat. Rev. Phys. (2023)

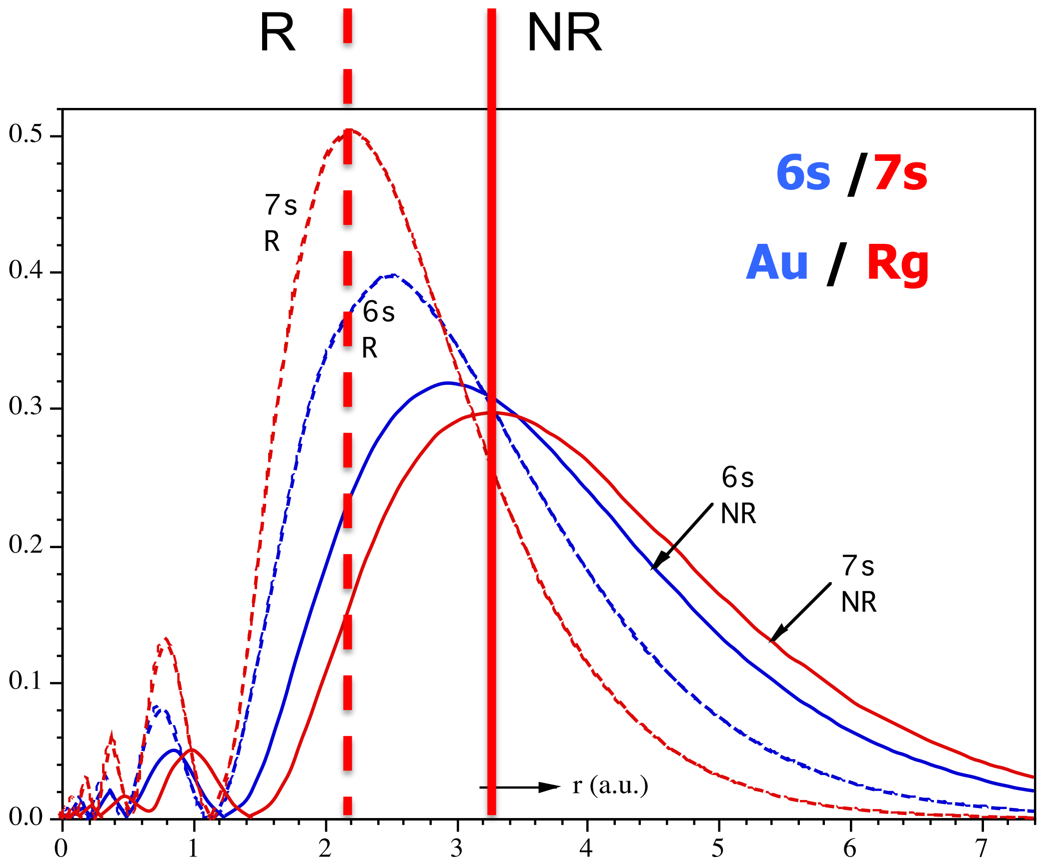
Are Superheavy Elements produced in R Process?



- r process path towards heaviest elements terminated by fission (“fission recycling”)
- limited experimental data for relevant nuclei, hence uncertainties of models influence predictions
- fission barrier heights strongly model dependent, thus, accurate description of fission is crucial
- impact of shell structure, e.g., N = 184
- Isomers (may) play a role

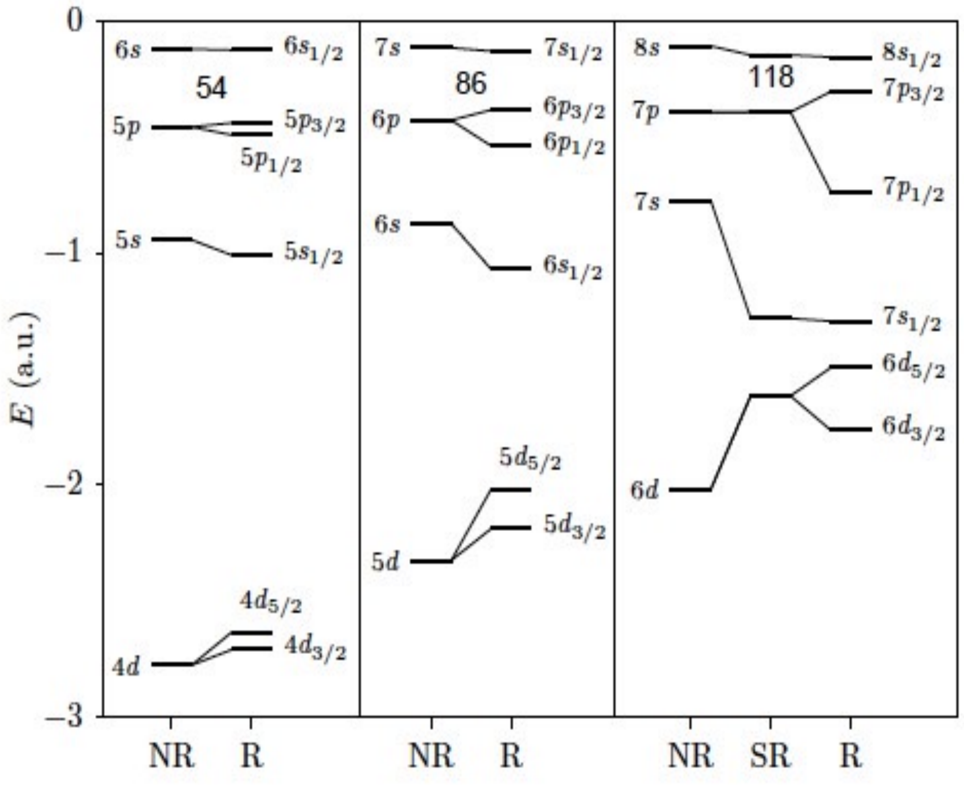
S. Goriely, G. Martinez Pinedo, Nucl. Phys. A 944 (2015) 158

Atomic Structure



- atomic structure of heavy elements is strongly influenced by relativistic effects
- Zα = 1: QED contributions significant
- strong impact of electron correlations
- large fine structure splitting
- many close-lying levels

Atomic Structure



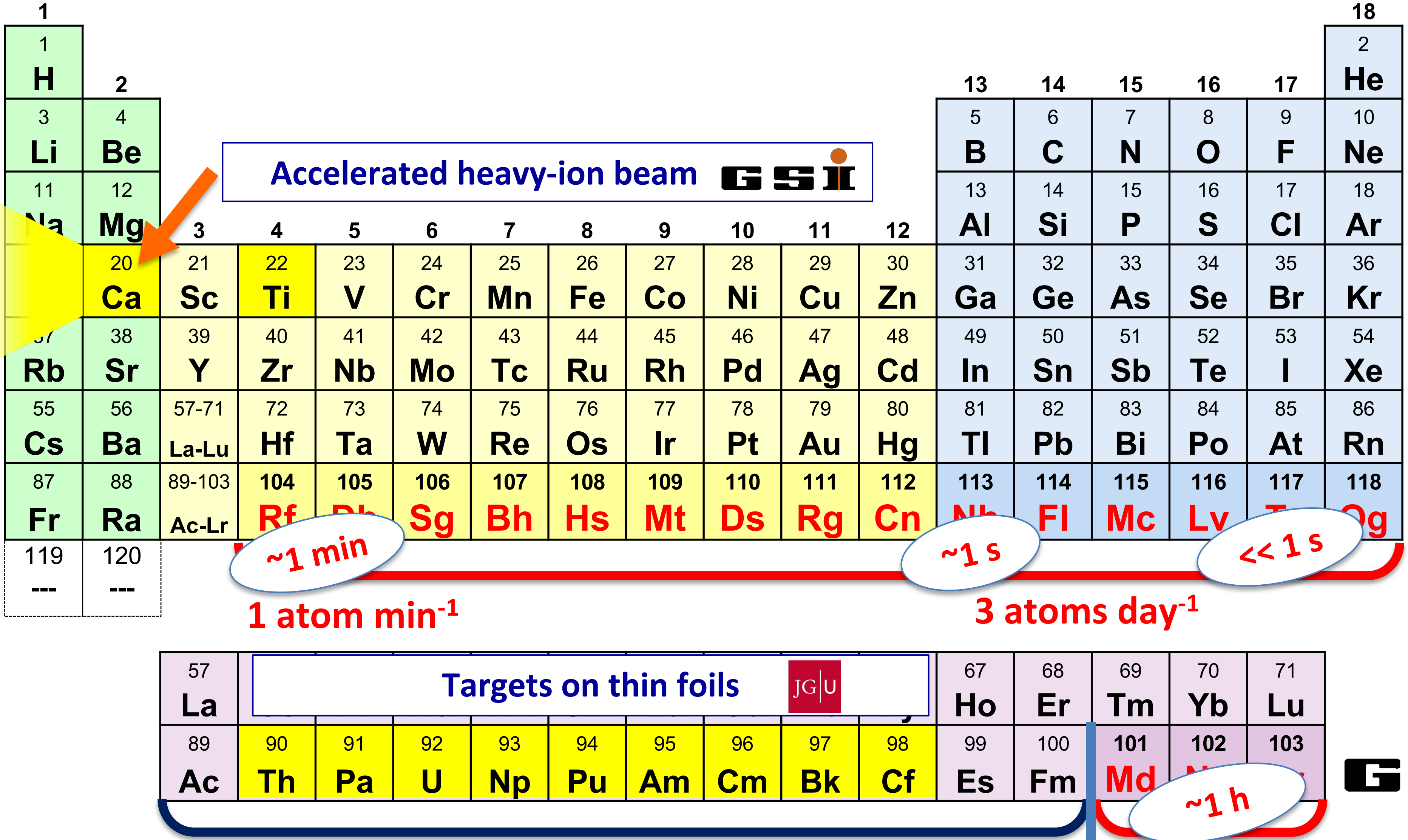
P. Jerabek et al., PRL **120,** 053001 *(2018)*

- atomic structure of heavy elements is strongly influenced by relativistic effects
- Zα = 1: QED contributions significant
- strong impact of electron correlations
- large fine structure splitting
- many close-lying levels

Production and study of superheavy elements

Exploration of mitigation of ^{48}Ca supply chain disruption started

Accelerated heavy-ion beam



1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
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
	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	57-71 La-Lu	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	89-103 Ac-Lr	**104** **Rf**	**105** **Db**	**106** **Sg**	**107** **Bh**	**108** **Hs**	**109** **Mt**	**110** **Ds**	**111** **Rg**	**112** **Cn**	**113** **Nh**	**114** **Fl**	**115** **Mc**	**116** **Lv**	**117**	**118** **Og**
119 ---	120 ---																

~1 min

~1 s

<< 1 s

1 atom min^{-1}

3 atoms day^{-1}

Targets on thin foils

57 La										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**	**103**

~1 h

tons

mg / µg / pg

50 atoms min^{-1}

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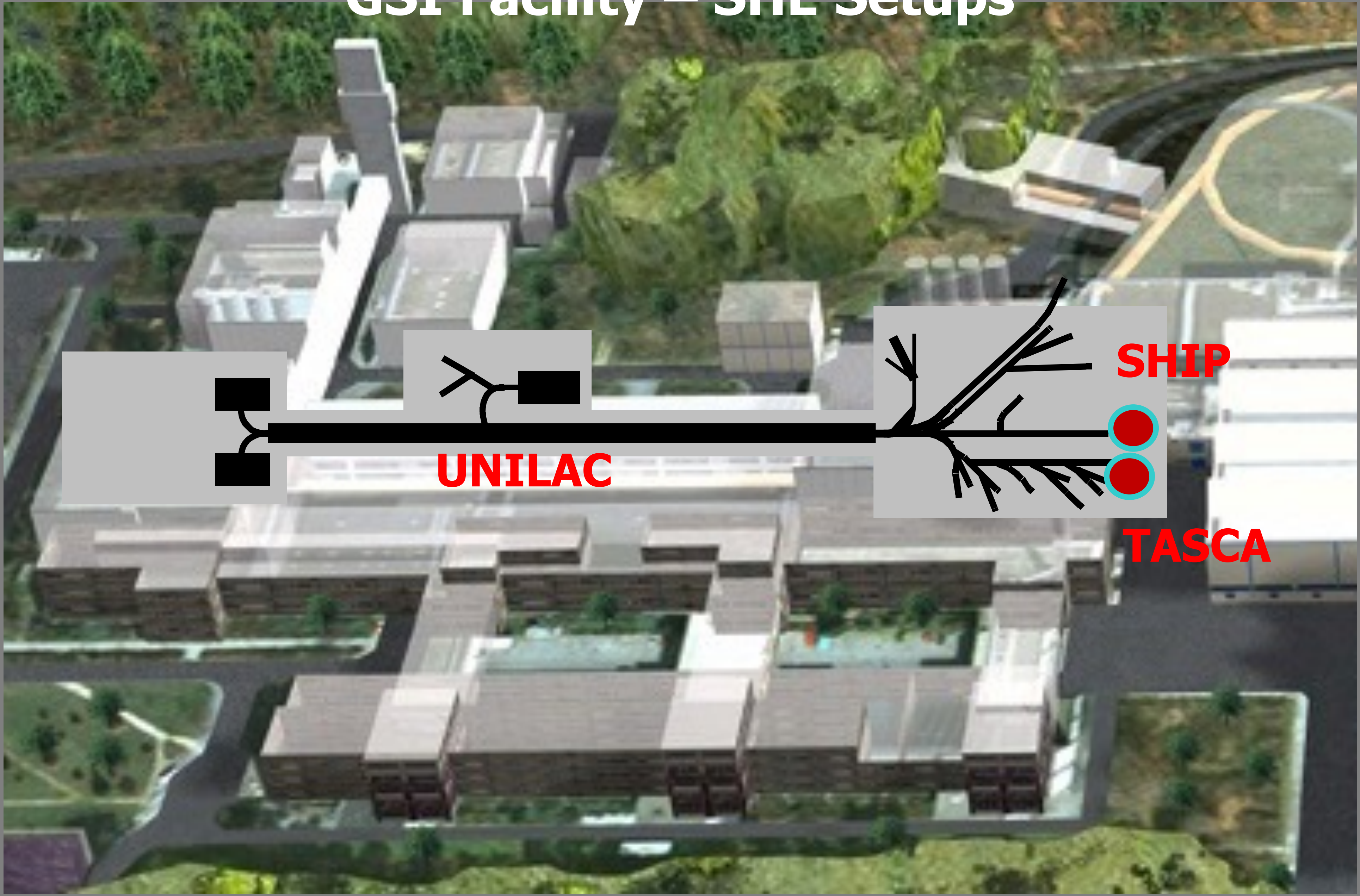
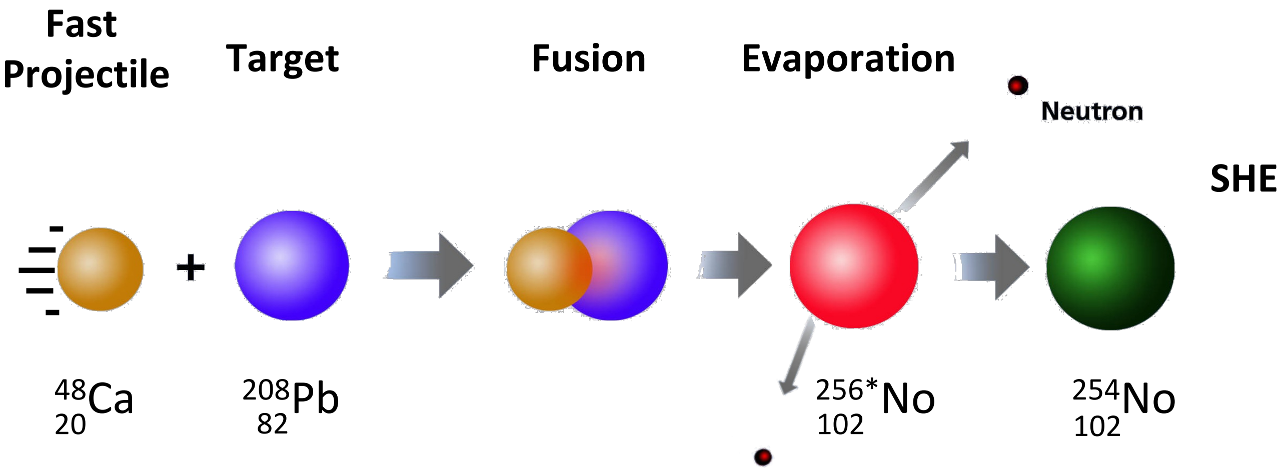
GSI Facility – SHE Setups



Production in Fusion-Evaporation Reactions



Yields

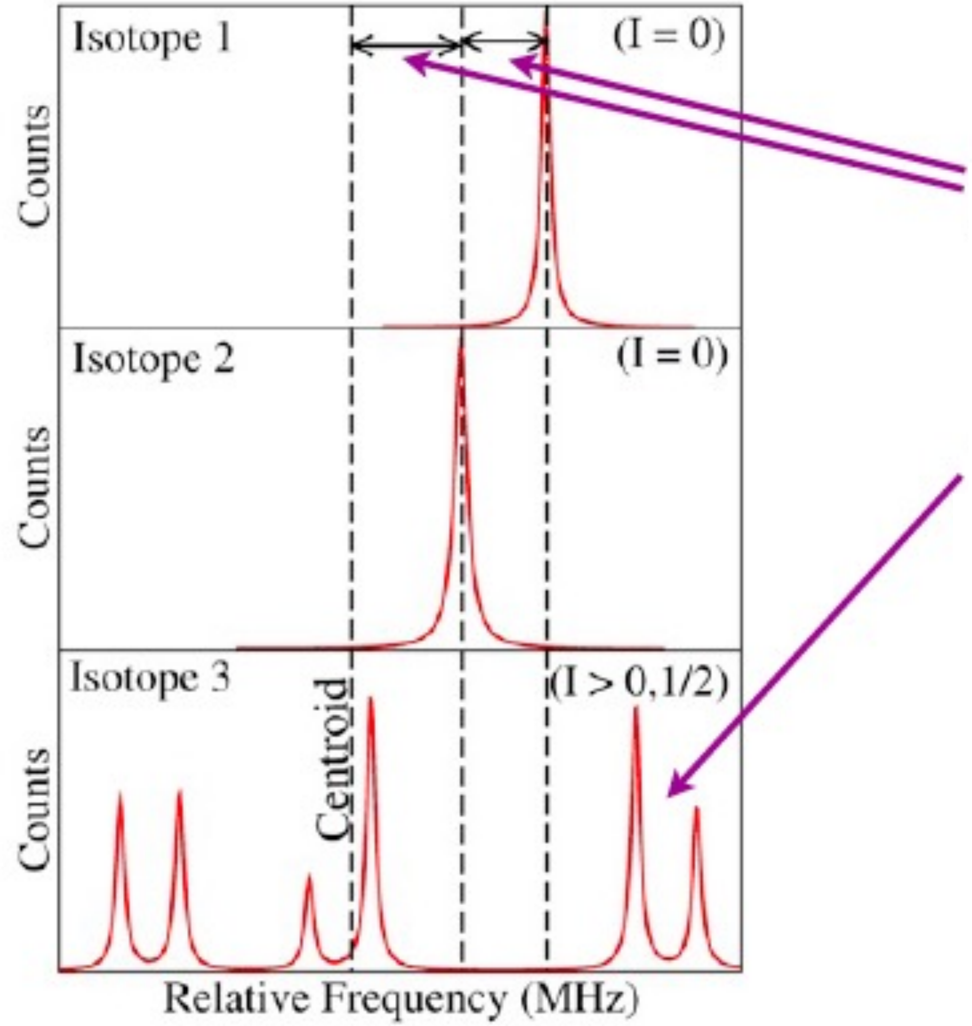
1 /s (Z≈102)

to

< 1 /week (Z≥112)

- produce heaviest nuclides through heavy-ion induced fusion-evaporation reactions
- requires high-intensity heavy-ion beams at few MeV/u
- high energy of reaction products calls for tailored laser spectroscopy techniques using gas cells for thermalization

Laser Spectroscopy of Radionuclides



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

$$\langle r^2\rangle^{AA'} = \left(\Delta\nu^{AA'} - \frac{A-A'}{AA'}M\right)\frac{1}{F}$$

Experiment Theory

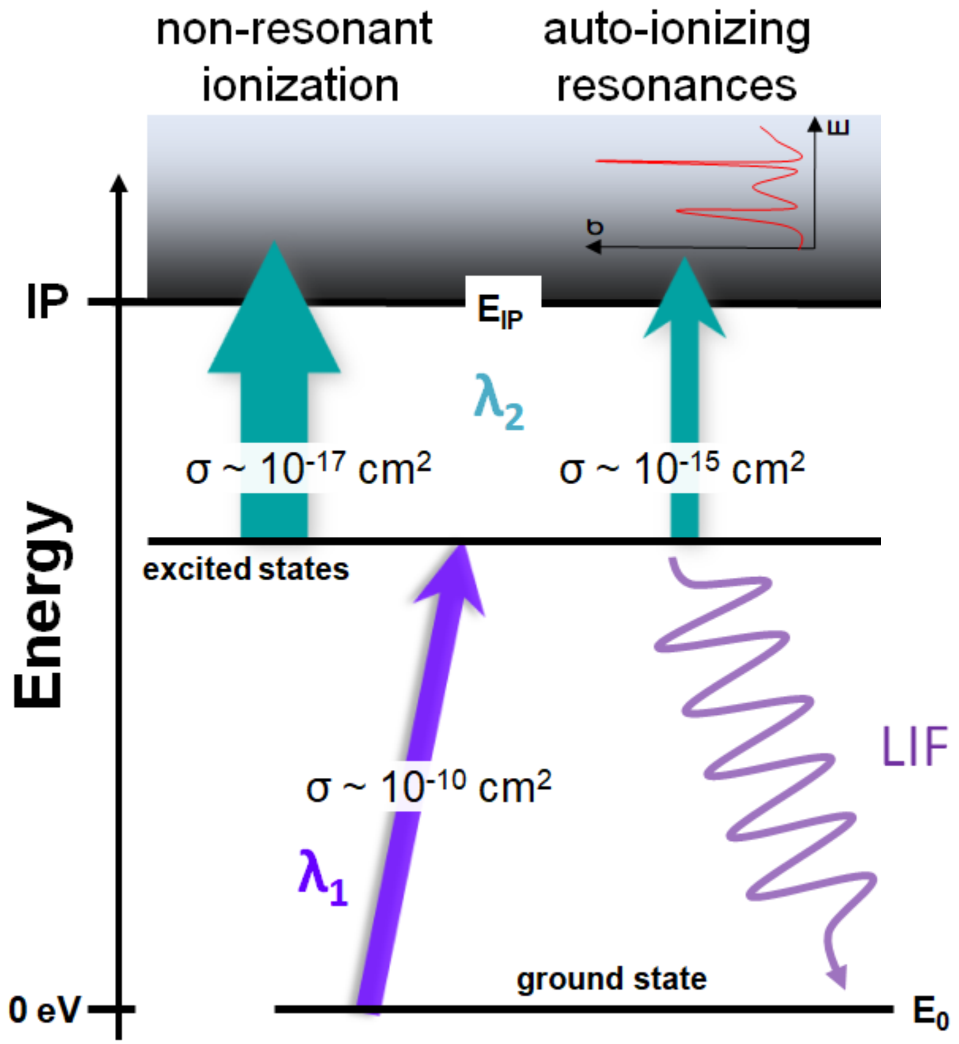
hyperfine spectroscopy yields parameters linked to magnetic dipole moment and spectroscopic quadrupole moment

$$A = \mu\frac{B_e(0)}{IJ} \qquad B = eQ_s\left\langle\frac{\delta^2 V}{\delta z^2}\right\rangle$$

Nuclear Atomic

Resonance Ionization Laser Spectroscopy

MB, M. Laatiaoui, S. Raeder, Prog. Nucl. Part. Phys. 116 (2011) 103834

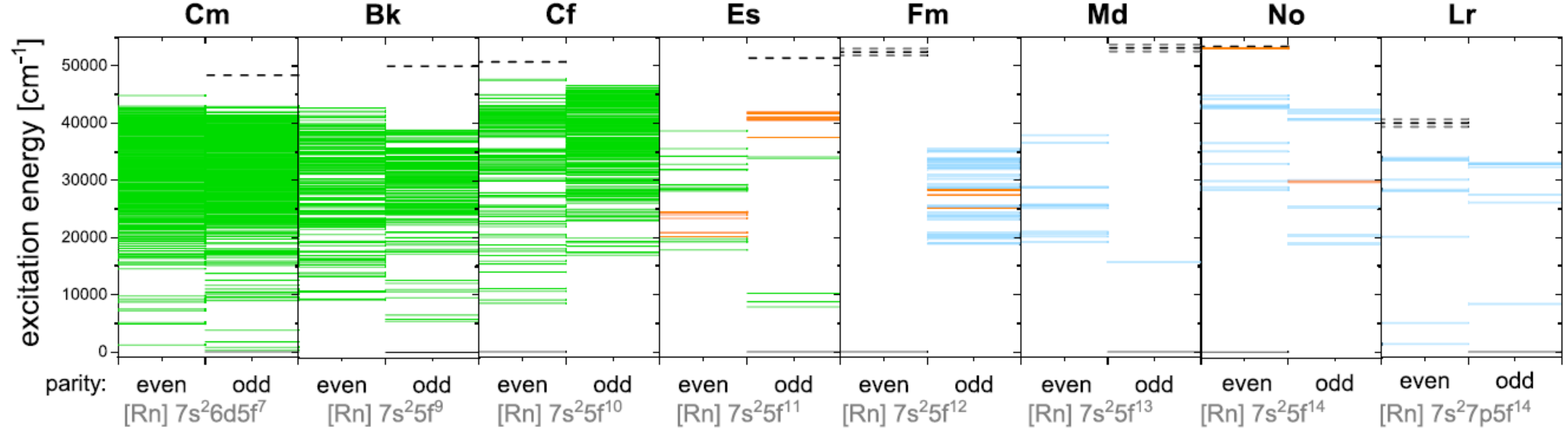


- ion detection more efficient than fluorescence photon detection
- low-background conditions when utilizing radioactive decay
- sensitive method applied in ultra-trace analysis and in laser-ion sources

Challenges for heaviest elements:

- no stable (long-lived) reference nuclides
- low yield and often short half-life

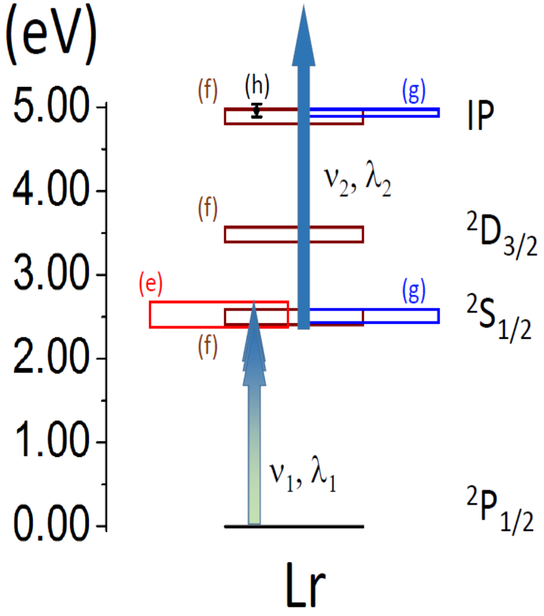
Atomic Structure of Actinides



🟩 Blaise 1992

🟨 Experimental data > 1992

🟦 Theory – calculated levels



- complex atomic structure with many close-lying levels
- limited experiment data for many actinide elements
- only theoretical predictions for heaviest ones

➢ obtain data on level energies, lifetimes, …

MB, M. Laatiaoui, S. Raeder, Prog. Nucl. Part. Phys. 116 (2021) 103834

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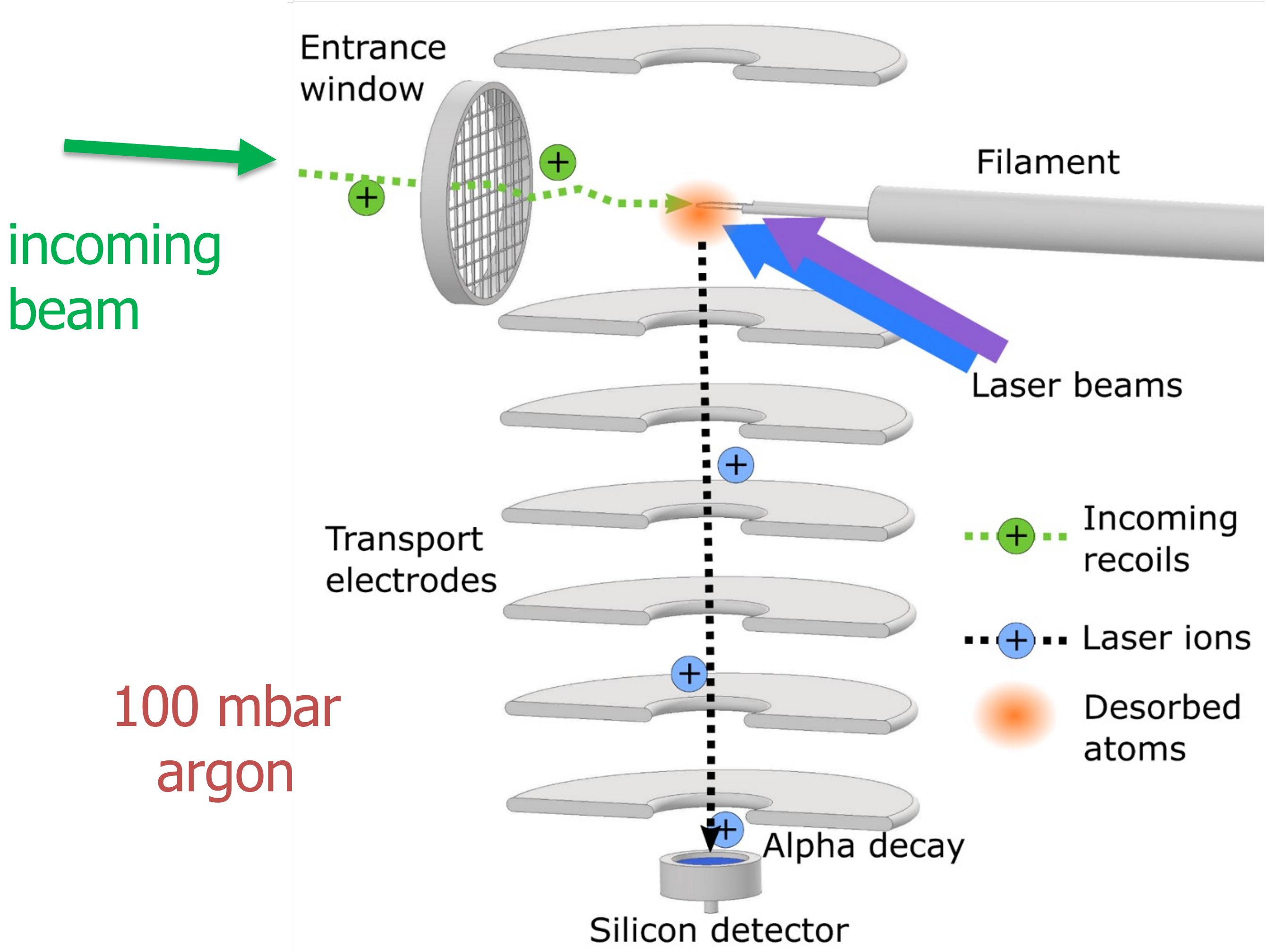
Radiation Detected Resonance Ionization Spectroscopy (RADRIS)

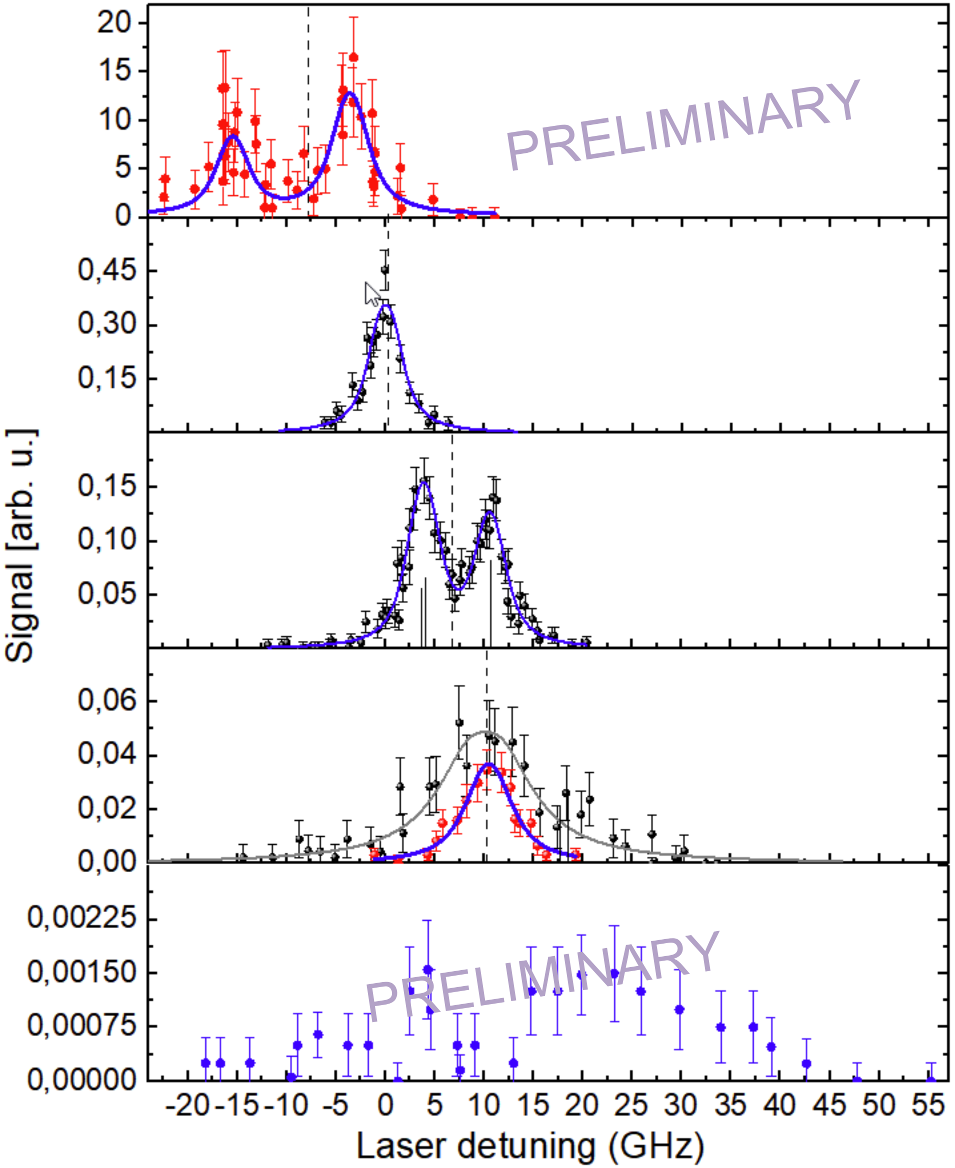


- RADRIS method tailored to measurements of actinide isotopes produced by fusion reaction with lowest rates

- slow down and neutralize in Ar gas
- evaporate atoms
- two-step photo-ionization
- transport to detector
- register radioactive decay

H. Backe et al. Eur. Phys. J. D, 45 (1) (2007), 99
F. Lautenschläger et al. Nucl. Instrum. Meth. B, 383 (2016),115
J. Warbinek et al., Atoms (2022)

Laser Spectroscopy of Nobelium (Z=102) Isotopes



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

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

FAIR Phase 0 Research Program
SHIP

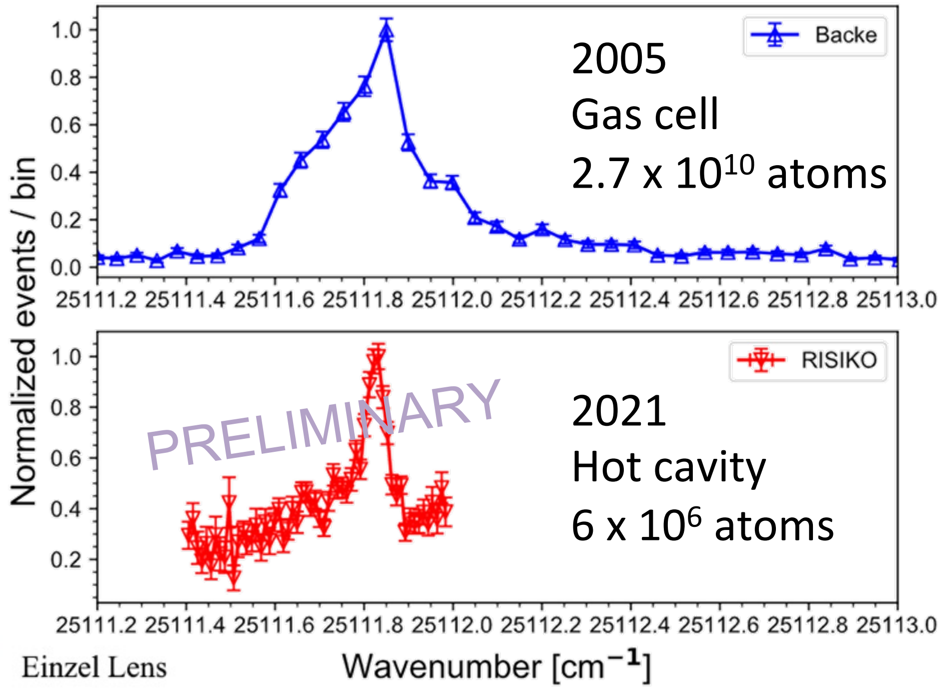
- first laser spectroscopy spectroscopy beyond Z=100
- yield as low as 0.05 atoms / second
- isotope shift allowed determining changes in mean-square charge radii around *N* = 152
- magnetic dipole and electric quadrupole moment of ^{253,255}No obtained from hyperfine splitting

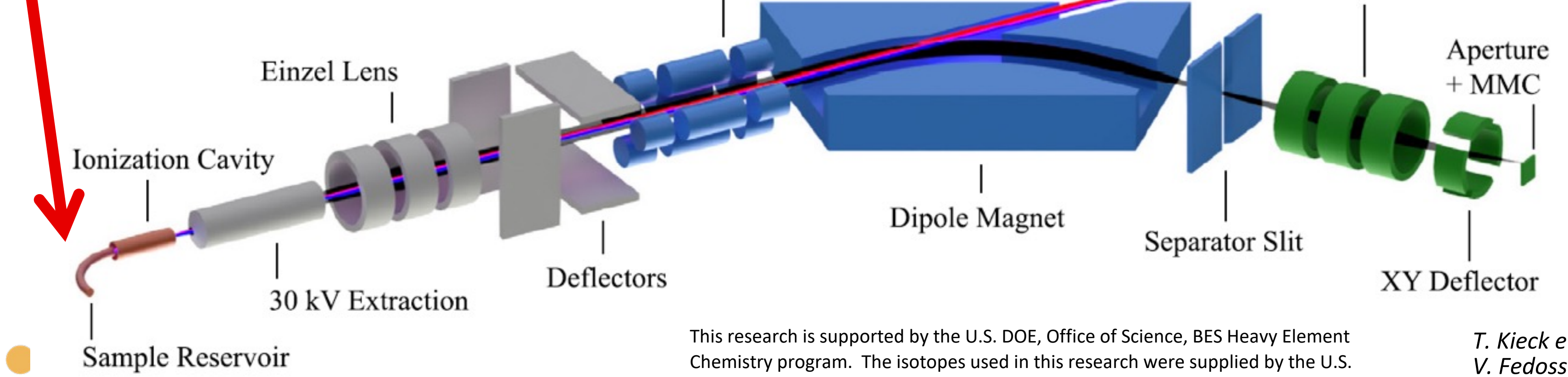
M. Laatiaoui *et al.*, Nature 538, 495 (2016)
S. Raeder *et al.*, Phys. Rev. Lett. 120 (2018) 232503

Laser Spectroscopy of Fm Isotopes in Hot Cavity

RISIKO mass separator in Mainz

- Production of radioactive ion beams
- Laser spectroscopy with high efficiency
- Resolution limited by temperature in the source and the 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).

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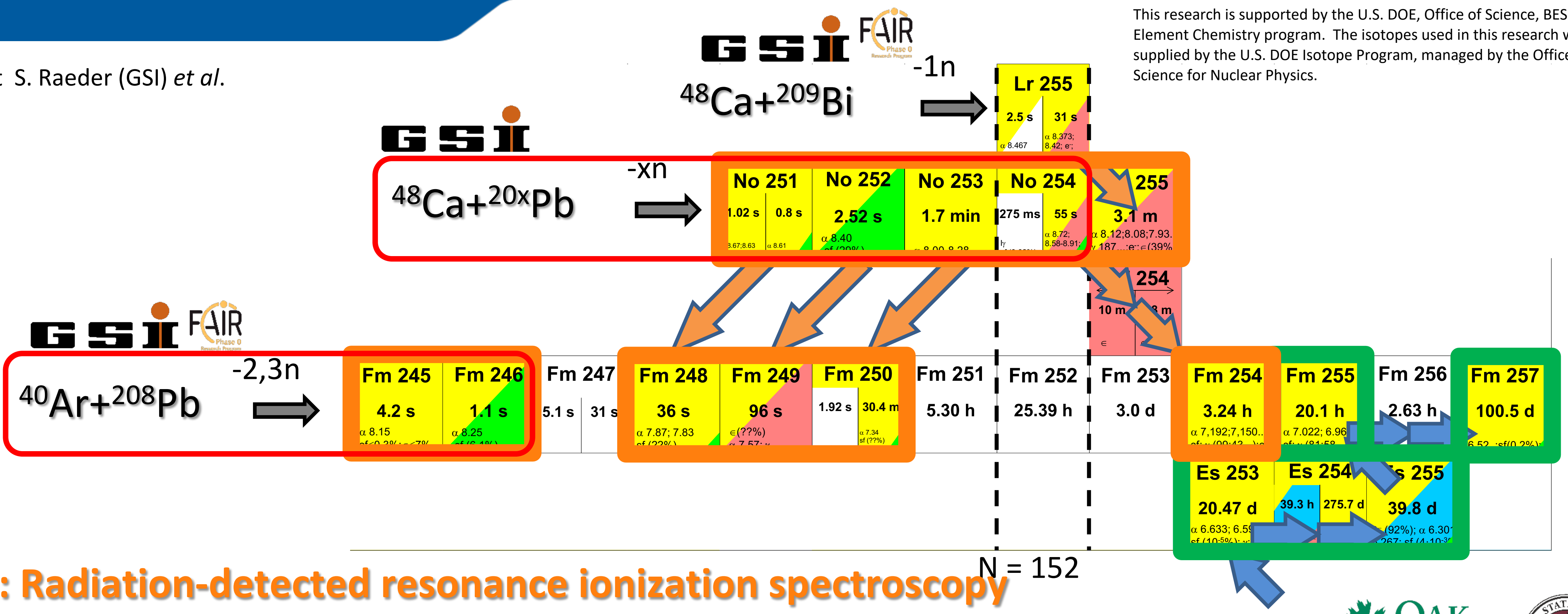
Production of $_{99}$Es, $_{100}$Fm, and $_{102}$No nuclei

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.

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



Online: Radiation-detected resonance ionization spectroscopy

M. Laatiaoui *et al.*, Nature 538 (2015) 492

Offline: Resonance ionization spectroscopy of Es

S. Nothhelfer *et al.*, PRC 105 (2022) L021302

Es reirradiated at

OAK RIDGE National Laboratory

NEUTRONS FOR SOCIETY

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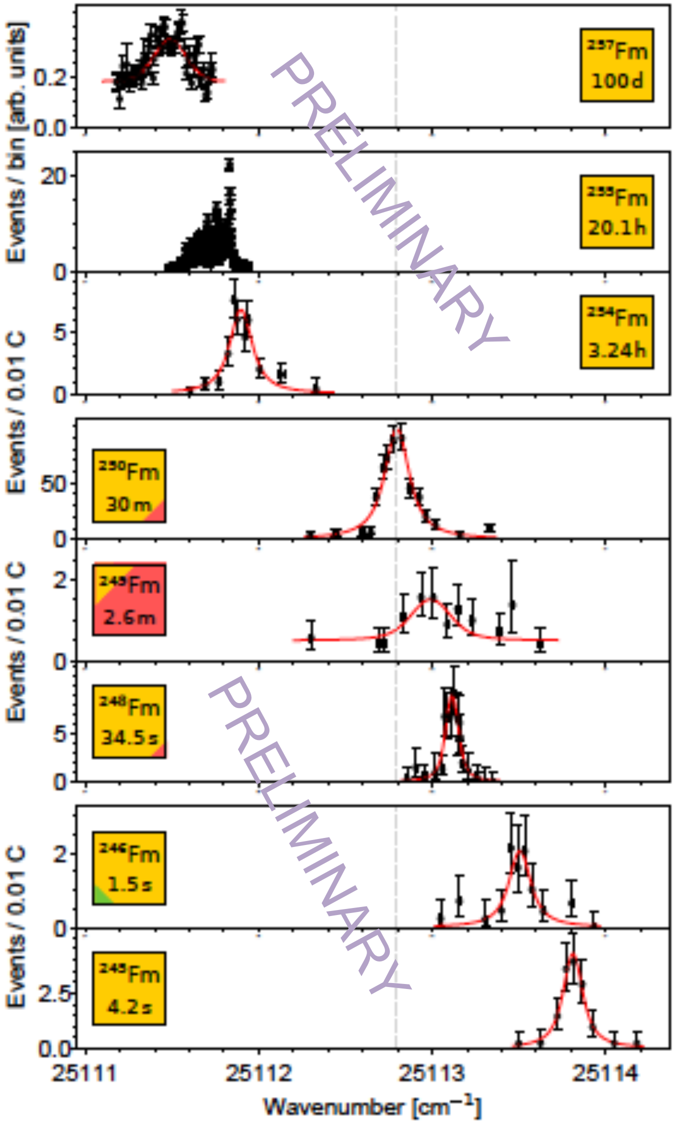
Laser Spectroscopy of Fm (Z=100) Isotopes



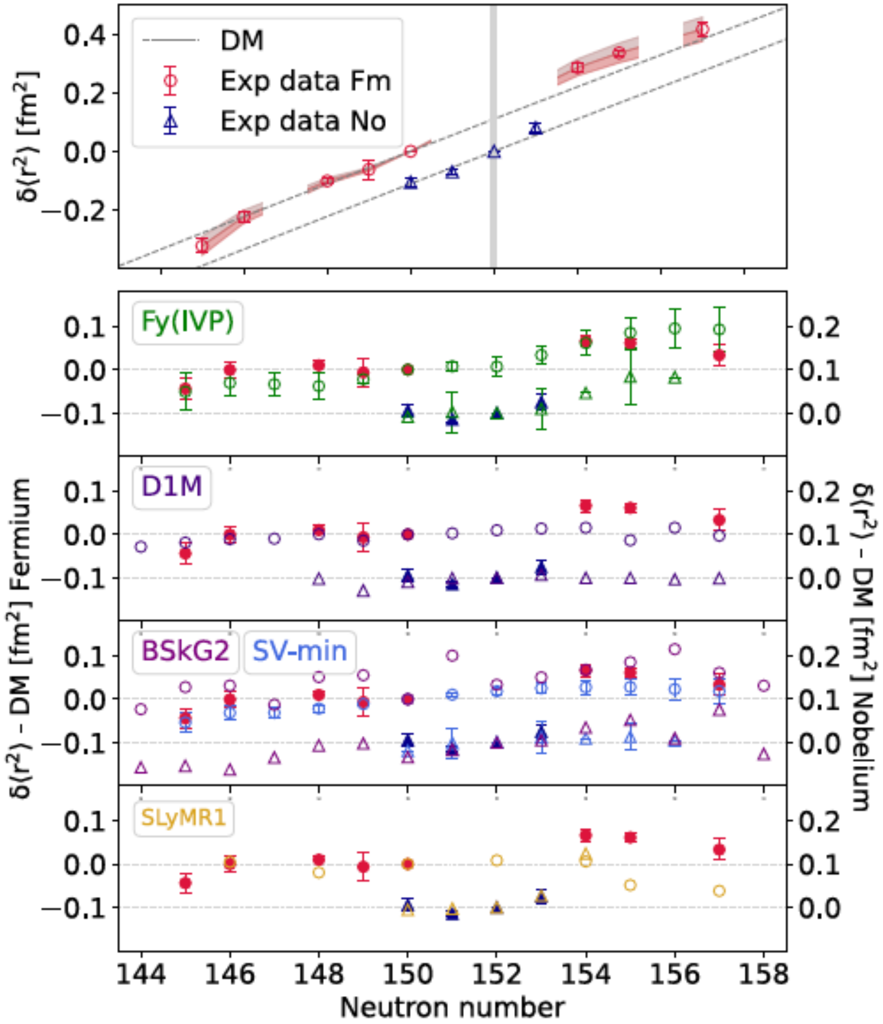
- short-lived Fm isotopes measured online at GSI
- some isotopes were produced indirectly via decay of directly produced No isotopes
- long-lived isotope ^{255,257}Fm from ORNL / ILL measured at RISIKO/Mainz after radiochemical separation by Mainz nuclear chemistry (Ch. Düllmann et al.)
- measured isotope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

Experiment S. Raeder, J. Warbinek (PhD thesis) *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}Es and ^{255,257}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.

Theoretical calculations



Calculations using different energy density functionals were carried out by W. Nazarewicz, P. G. Reinhard, S. Goriely, S. Hilaire, S. Peru, M. Bender, B. Bally

- models agree well with each other and with experimental data
- charge radii show no significant signature
- of single-particle structure
- deformed shell gap at *N*=152 established in masses, SF half-lives

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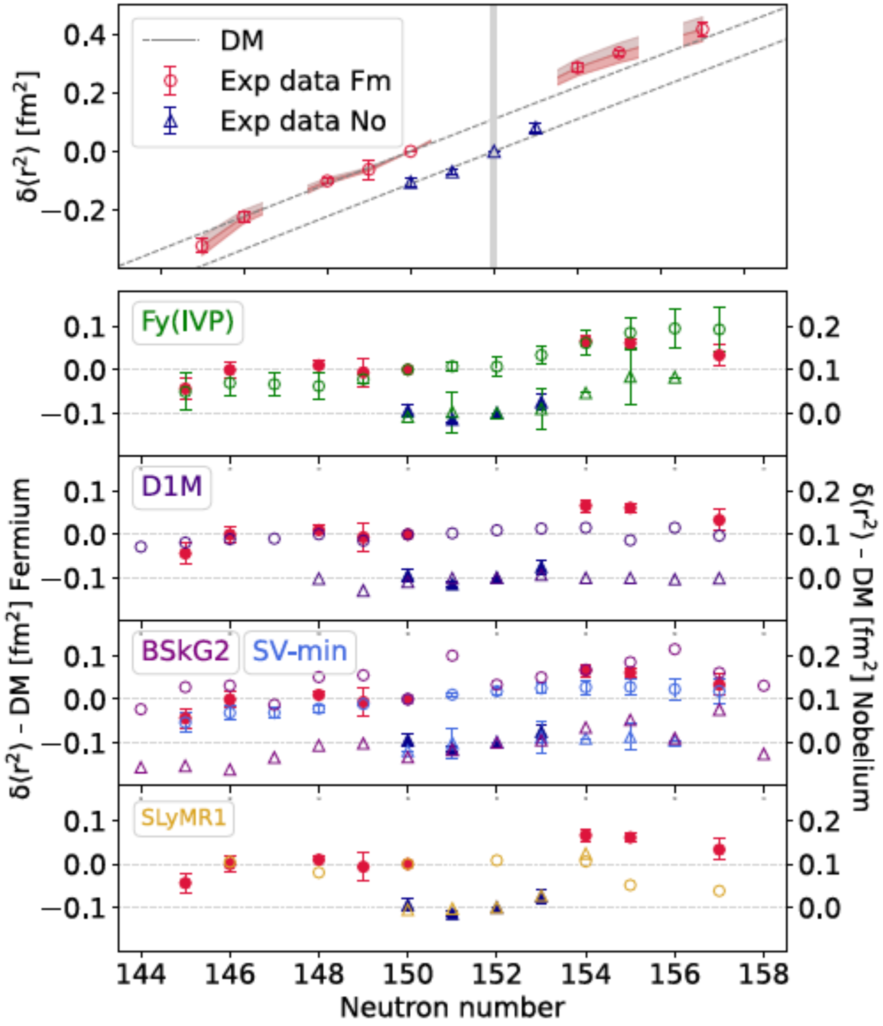
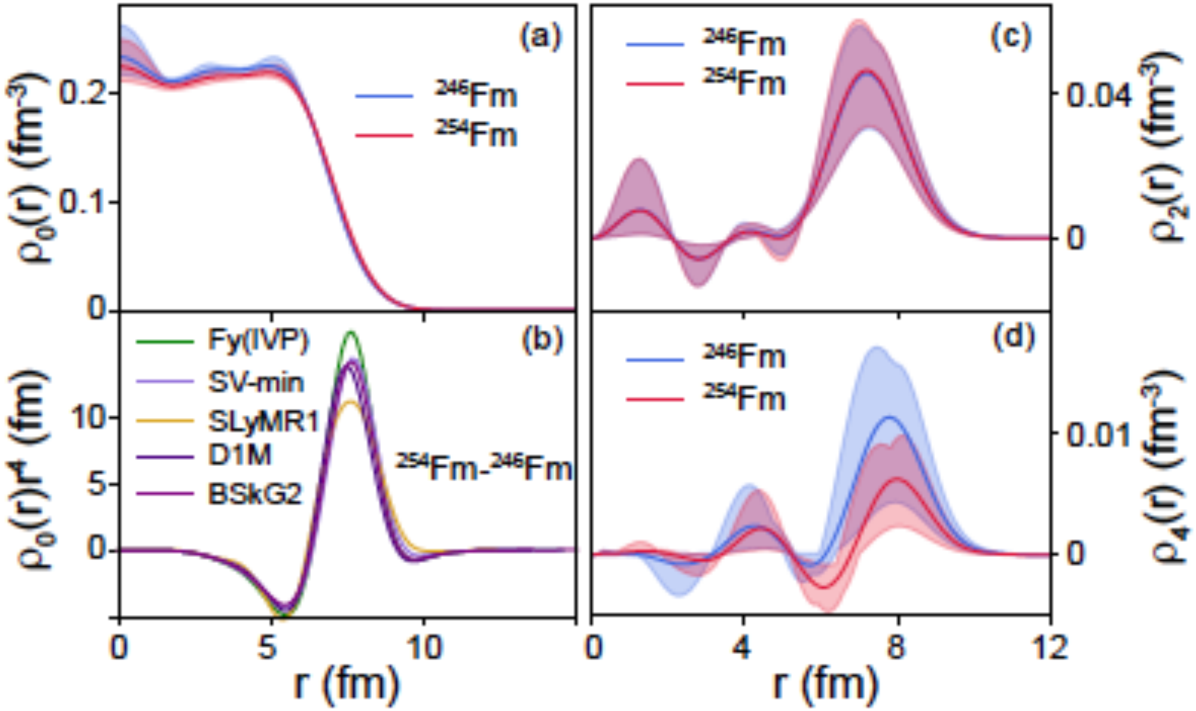
Theoretical calculations



- models agree well with each other and with data
- charge radii show no significant signature of single-particle structure

Theoretical calculations

B. Bally, M. Bender, S. Goriely, S. Hilaire, W. Nazarewicz, S. Peru, P.G. Reinhard, W. Ryssens



- Different nuclear models agree well with each other and with experimental data
- charge radii show no significant signature of single-particle structure

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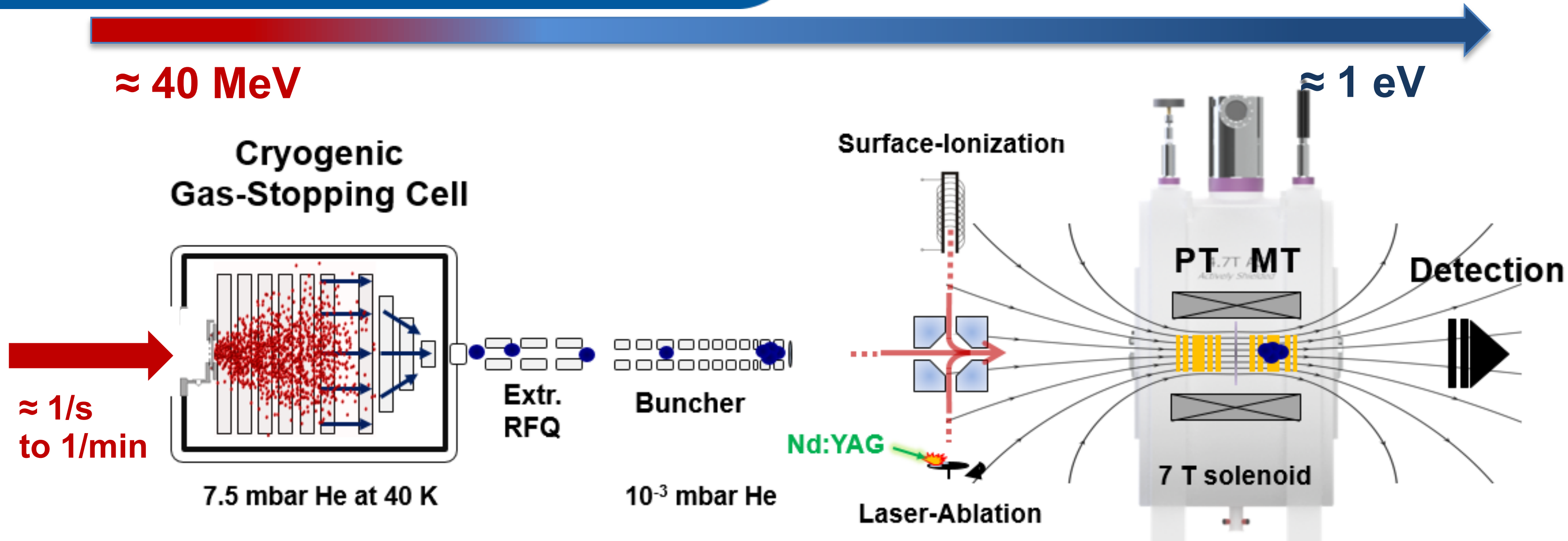
SHIPTRAP Setup at GSI Darmstadt



≈ 40 MeV

≈ 1 eV

≈ 1/s
to 1/min

Cryogenic Gas-Stopping Cell

7.5 mbar He at 40 K

Extr. RFQ

Buncher

10^{-3} mbar He

Surface-Ionization

Nd:YAG

Laser-Ablation

PT **MT**

7 T solenoid

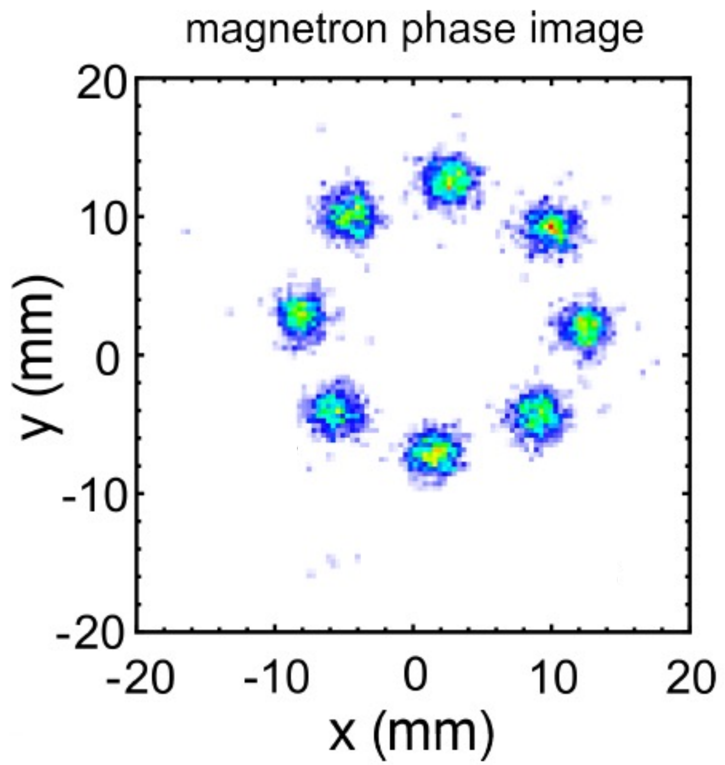
Detection

Stopping

Tansport & Bunching

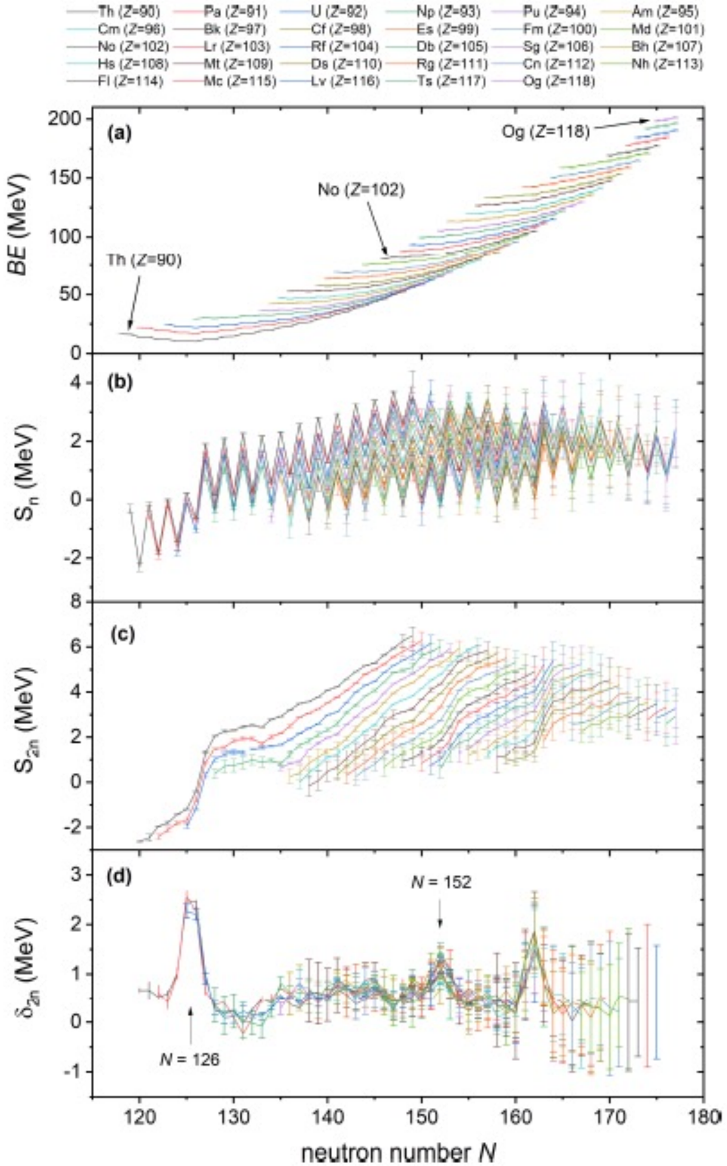
Measurement

Photo: G. Otto, GSI



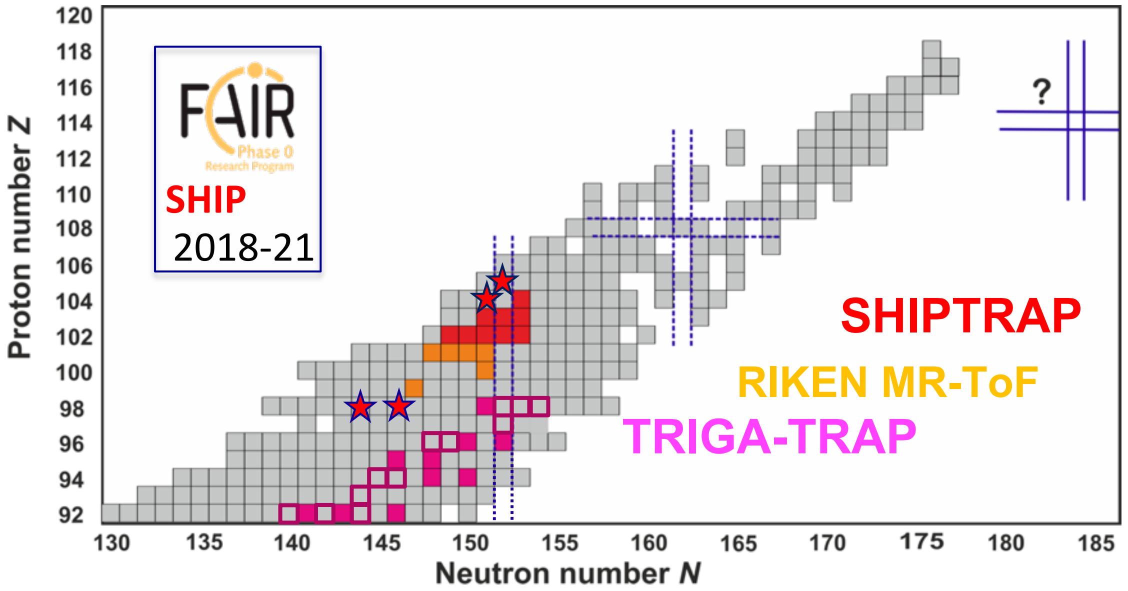
Experiment F Giacoppo (GSI) *et al*.

Masses and Nuclear Shell Structure



- nuclear masses and mass differences reflect nuclear shell structure: signatures of shell closures, pairing, and the onset of deformation can be observed
- precision of experimental data nowadays on the order of few keV or better even for many exotic nuclei
- mass data show deformed shell gaps in SHN at *N* = 152 and *N* =*162*

Masses of Heavy Nuclei - Status



- O. Kaleja, Phys. Rev. C (2022) 054325
- M. Eibach et al., Phys. Rev. C 89, 064318 (2014)
- E. Minaya Ramirez et al. Science 337, 1207 (2012)
- M. Block et al., Nature 463, 785 (2010)

RIKEN/KEK:

- P. Schury et al., Phys. Rev. C 104, L021304 (2022)
- Y. Ito et al., Phys. Rev. Lett. 120, 152501 (2018)

- direct mass spectrometry Z > 100 established with SHIPTRAP/GSI in 2008
- Measurements performed with rates of ≈ 0.00002/s and 5 detected ions in total
- rel. mass uncertainty of 10^{-8} and better
- high mass resolving power of Penning traps allows identification / study of (long-lived) isomers
- mass measurements investigate shell structure around Z=100, N=152

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Long-Lived Isomers in the Heaviest Elements

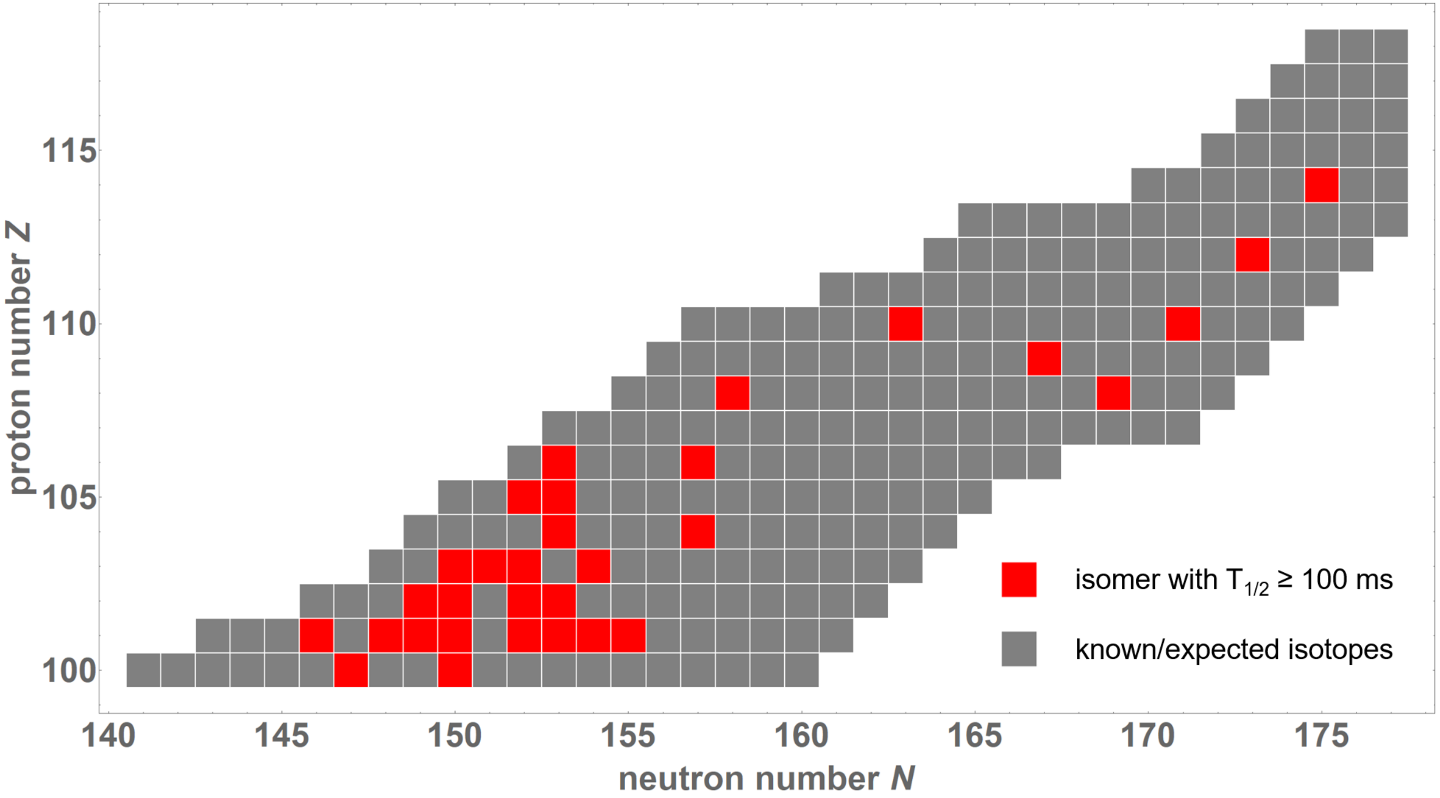

Figure courtesy O. Kaleja

- 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

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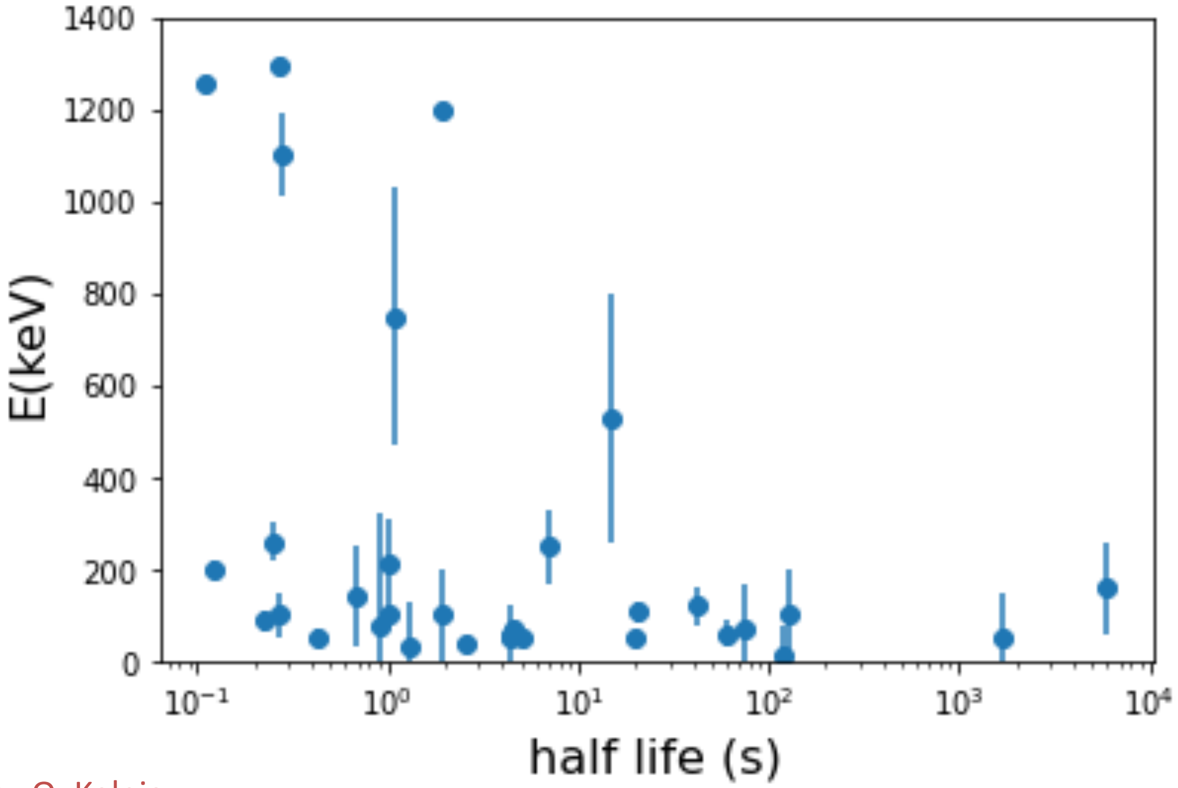

Figure courtesy O. Kaleja

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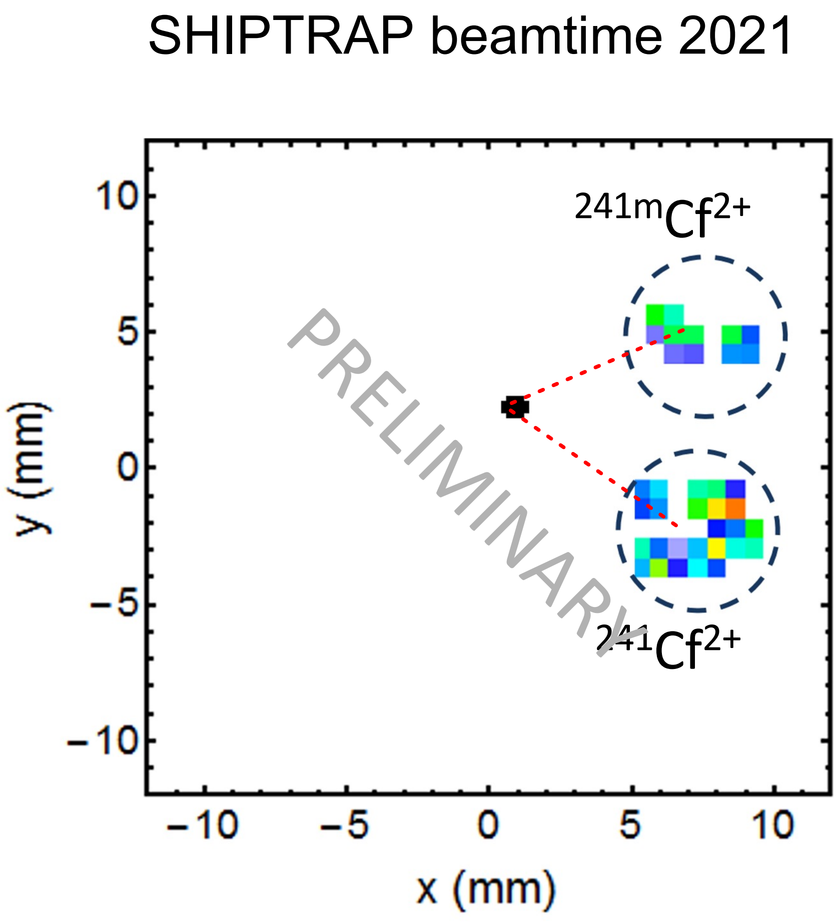
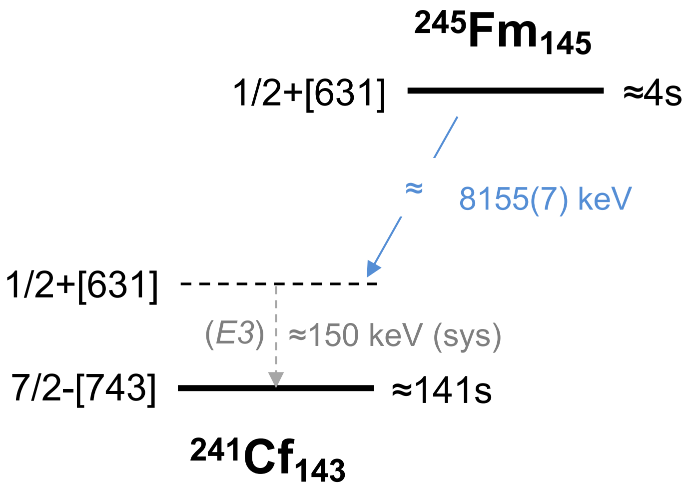
Long-Lived Isomer in ^{241}Cf


SHIPTRAP beamtime 2021

- decay spectroscopy at SHIP (J. Khuyagbaatar *et al.*, PRC (2020) **102,** 044312): systematics of lighter N = 141,143 isotones suggest existence of isomeric state in ^{241}Cf at ≈ 150 keV


^{245}Fm$_{145}$

1/2+[631] ≈4s

≈ 8155(7) keV

1/2+[631]

(*E*3) ≈150 keV (sys)

7/2-[743] ≈141s

^{241}Cf$_{143}$

- Isomer in ^{241m}Cf eventually detected with SHIPTRAP in direct mass measurement of ^{241}Cf with $T_{1/2}$ >100ms

Experiment and data analysis F. Giacoppo, M.J Gutierrez et al.

Summary

- Many isotopes of superheavy elements up to Z=118 synthesized in last decades, most of them neutron-deficient compared to N=184
- Evidence for region of enhanced (alpha-decay) lifetimes but no island of stability expected
- mass measurements and laser spectroscopy extended to ever-heavier elements to open the door for more comprehensive investigation of superheavy nuclides
- Technical and methodological developments crucial to access rarest nuclides

THANK YOU FOR YOUR ATTENTION!