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Exploring the structure of the heaviest nuclei through laser spectroscopy and mass spectrometry

Michael Block

GSI Darmstadt, Helmholtz Institute Mainz, University Mainz

DPG Frühjahrstagung Freiburg – March 2024

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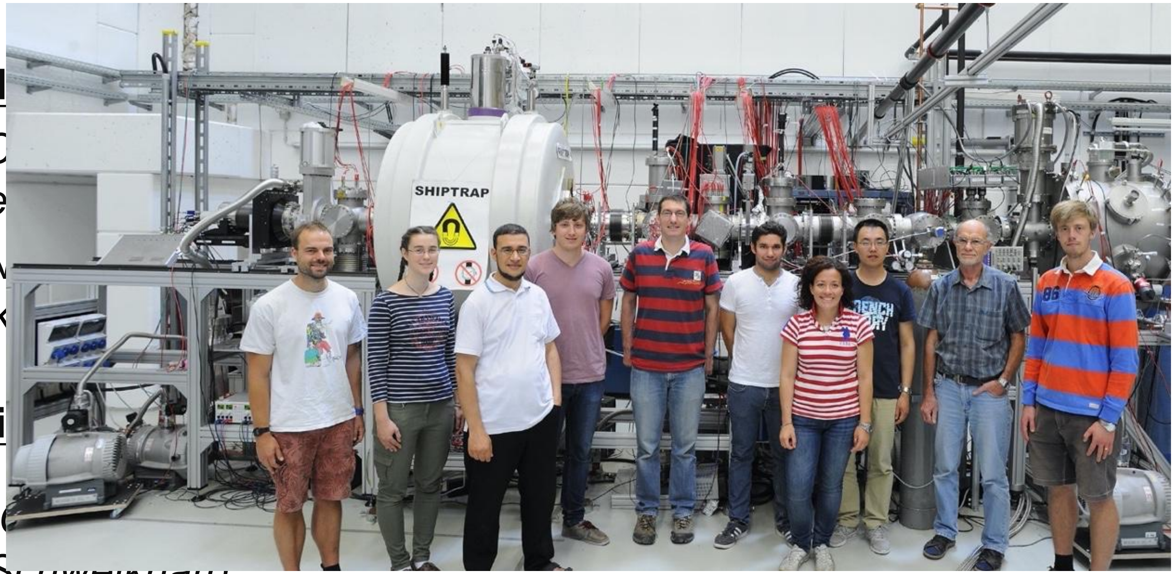

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FAIR Phase 0 Research Program

LMU

IJCLab Irène Joliot-Curie Laboratoire de Physique des 2 Infinis

Bundesministerium für Bildung und Forschung

EL EM EN TS

The Fermium Collaboration

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U.S. DEPARTMENT OF ENERGY

RADRIS & JetRIS Collaboration

GSI Darmstadt

B. Andelic, M. Block, F. Giacoppo, F.P. Heßberger, O. Kaleja, T. Kieck, A. Mistry, S. Raeder, J. Warbinek, A.Yakushev, M. Guiterrez-Torres

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P. Kunz

University of Liverpool

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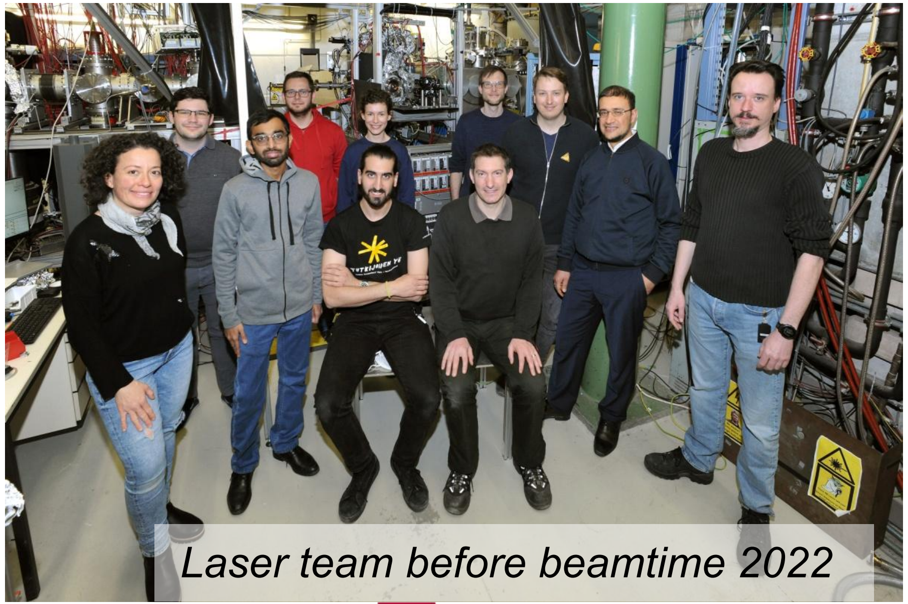

Laser team before beamtime 2022

Collaboration meeting 2022

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Outline

- Open questions in superheavy element research
- Experimental challenges studying (super)heavy nuclei
- Laser spectroscopy and mass measurements of (super)heavy nuclides at GSI Darmstadt and JGU Mainz
- Summary and conclusions

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What are superheavy elements?

Periodic Table around 1939

PERIODIC TABLE - BEFORE WORLD WAR II

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac	Th	Pa	U	93	94	95	96	97	98	99	100				
		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	

G. T. Seaborg J. Chem. Educ. (1968)

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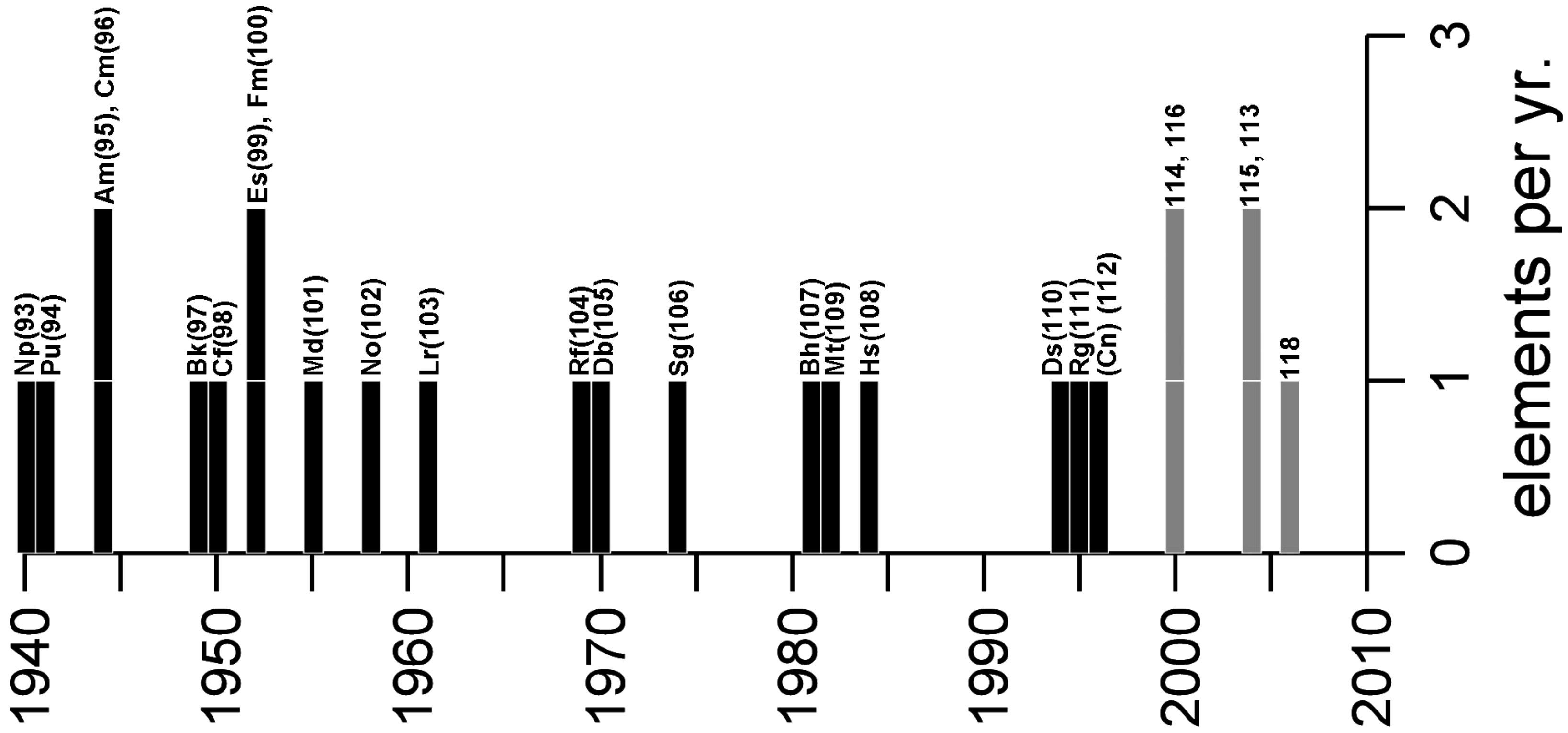
Discovery of Transuranium Elements



Year of Discovery

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The Periodic Table of Elements 2024

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

- established by Mendeleev, Meyer, and others about 150 years ago
- quantum mechanical description of atoms explains periodicity
- some deviations due to relativity

Transactinide Elements
= Superheavy Elements

Actinide Elements

elements **Bh, Hs, Mt, Ds, Rg, Cn** with Z=107-112 discovered GSI Darmstadt (1981-1996)

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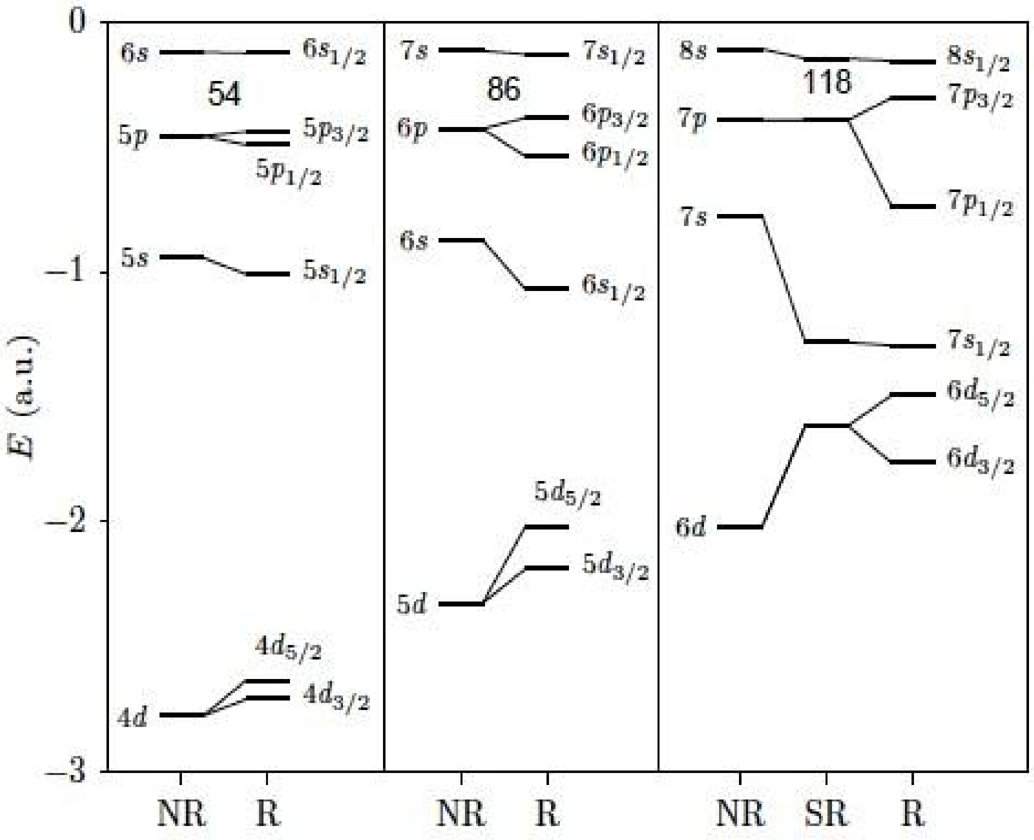
Atomic Structure of Heaviest Elements



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

- atomic structure of heavy elements is strongly influenced by relativistic effects
- Zα ≈ 1: quantum electrodynamics (non perturbative regime) plays role
- many-body problem: significant influence of electron correlations
- accurate theoretical descriptions challenging

➢ benchmark by experiments

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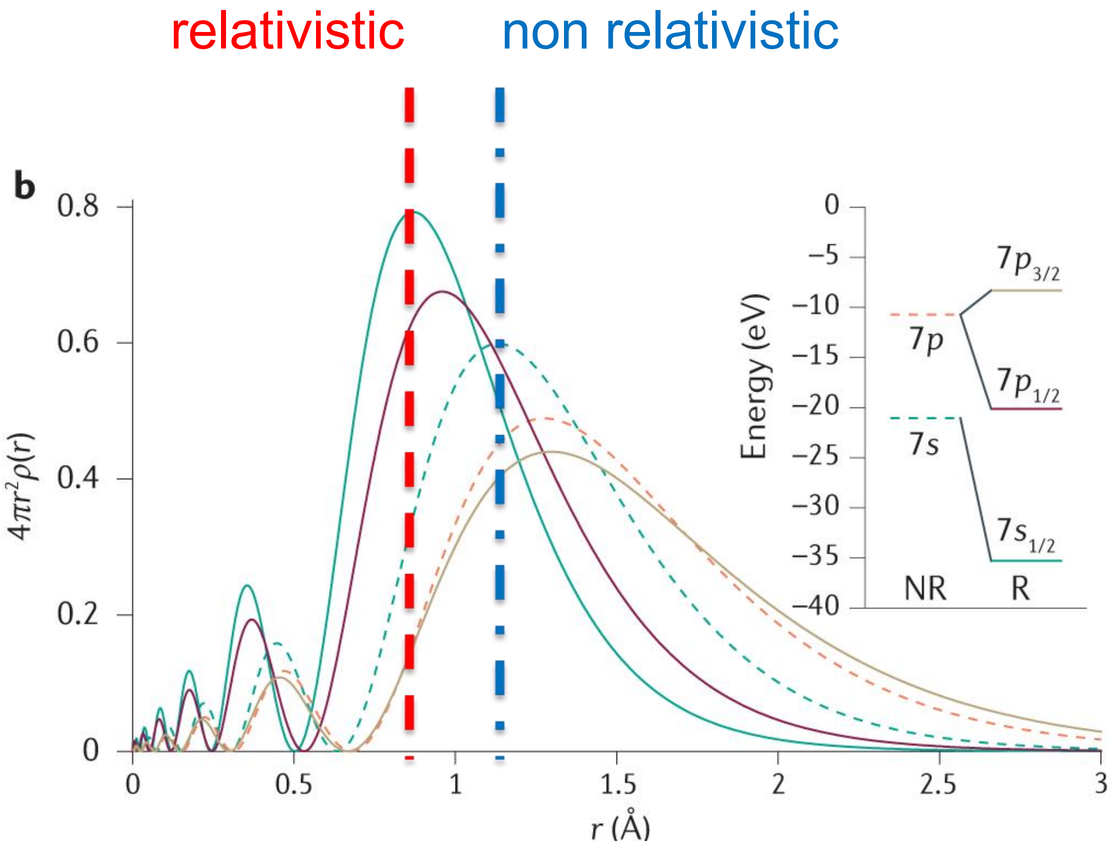
Atomic Structure of Heaviest Elements



O. Smits et al,. Phys. Rep. 1035 (2023) 1

- atomic structure of heavy elements is strongly influenced by relativistic effects
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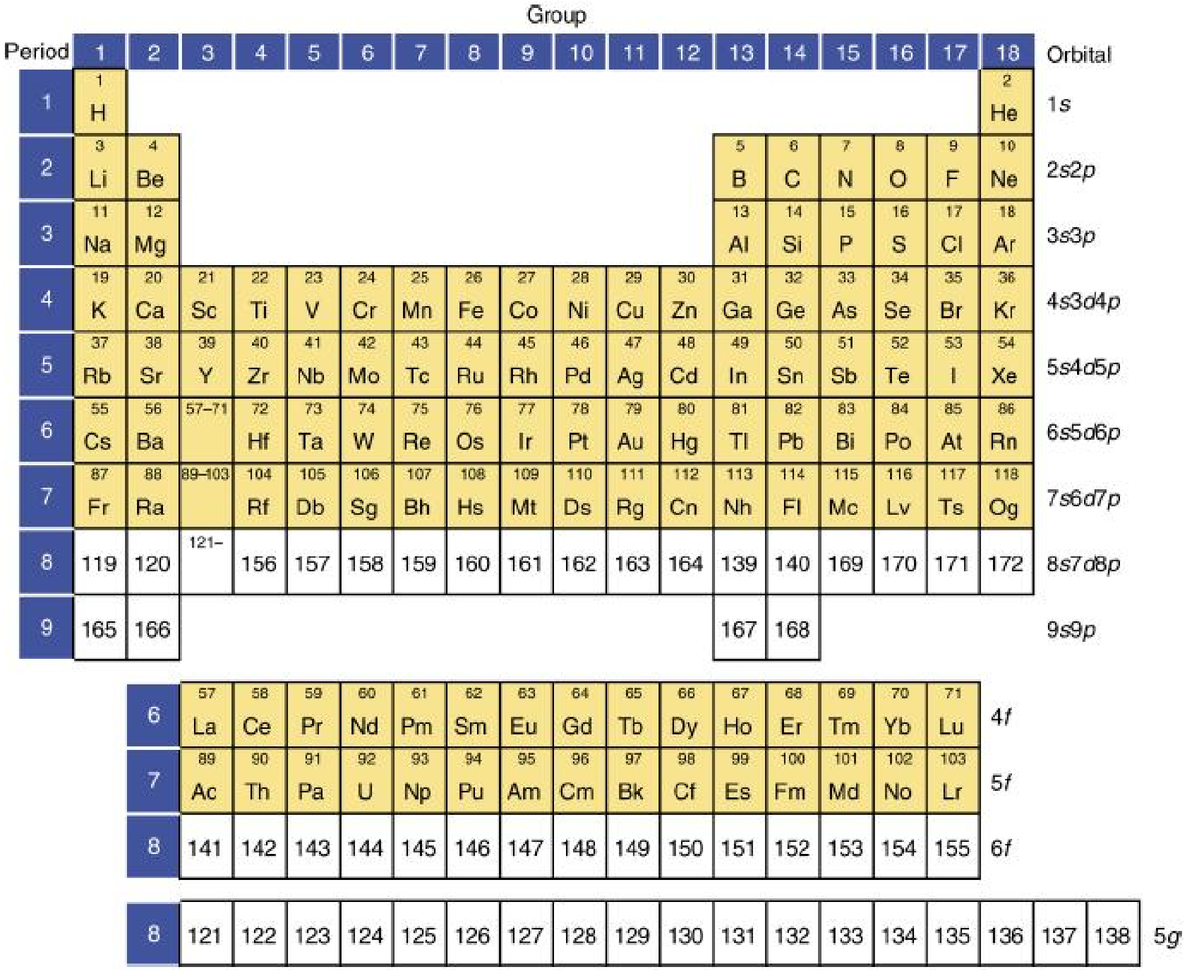
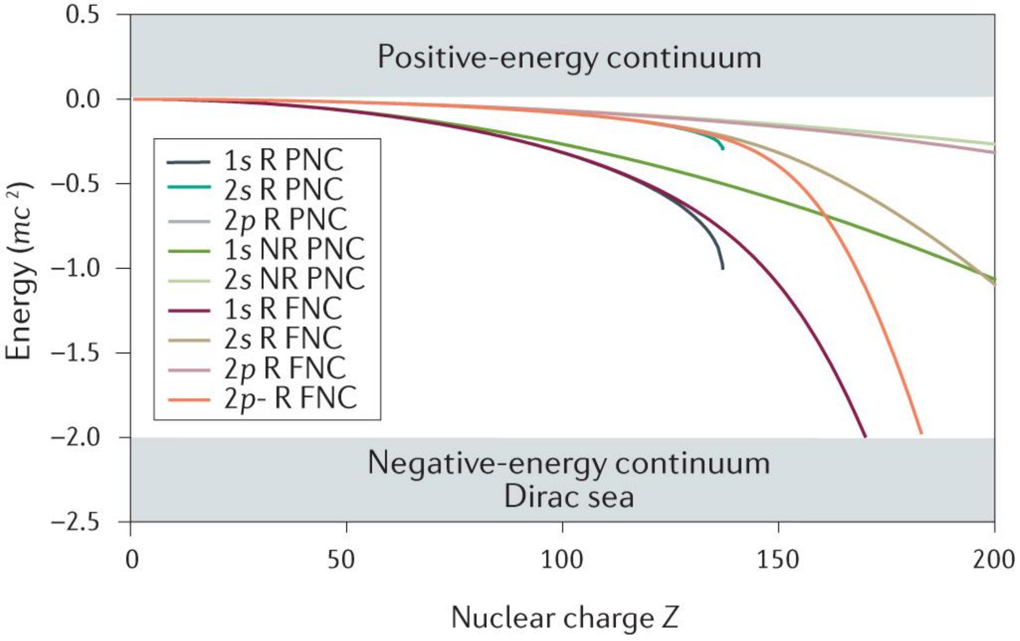
The Limits of the Periodic Table



P. Pyykkö, Physical Chemistry Chemical Physics 13(1), 161 (2011)



Limits of the periodic table:

- 1s dives into the Dirac sea around Z ≈ 172 for finite-size nuclei (FNC)
- different predictions for limits of Periodic Table

O. Smits et al,. Phys. Rep. 1035 (2023) 1

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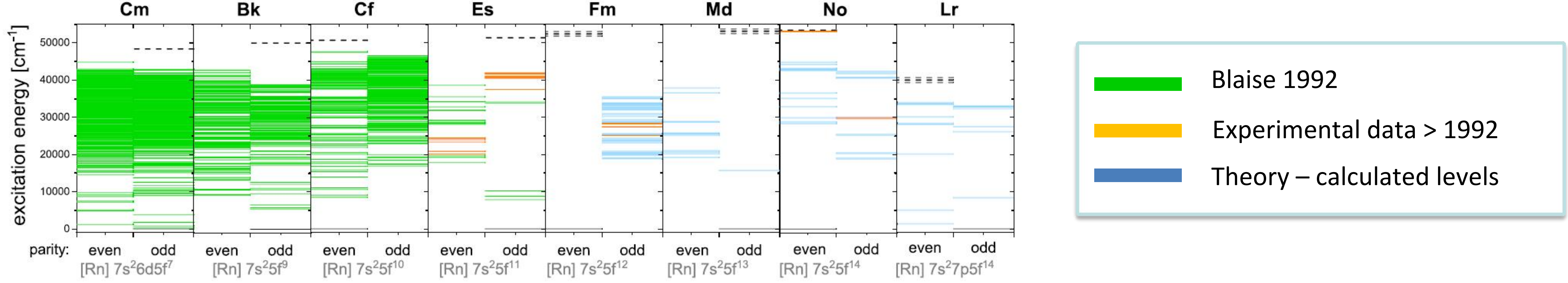
Atomic Structure of Actinides



- complex atomic structure with many close-lying levels
- limited experiment data for many actinide elements
- mainly theoretical predictions for heaviest actinides
- theoretical uncertainties ≈ 300 cm^{-1}

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

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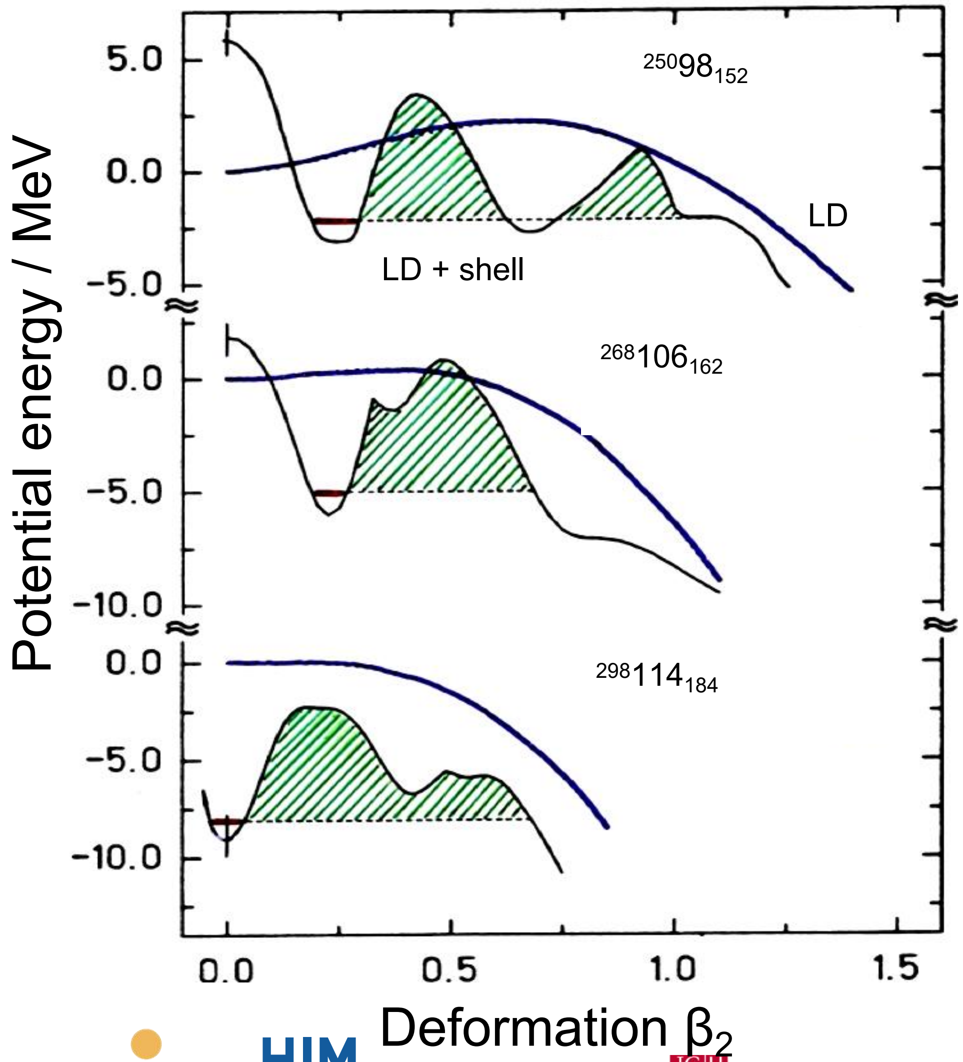
What is special about superheavy nuclei?

Fission in Superheavy Nuclei

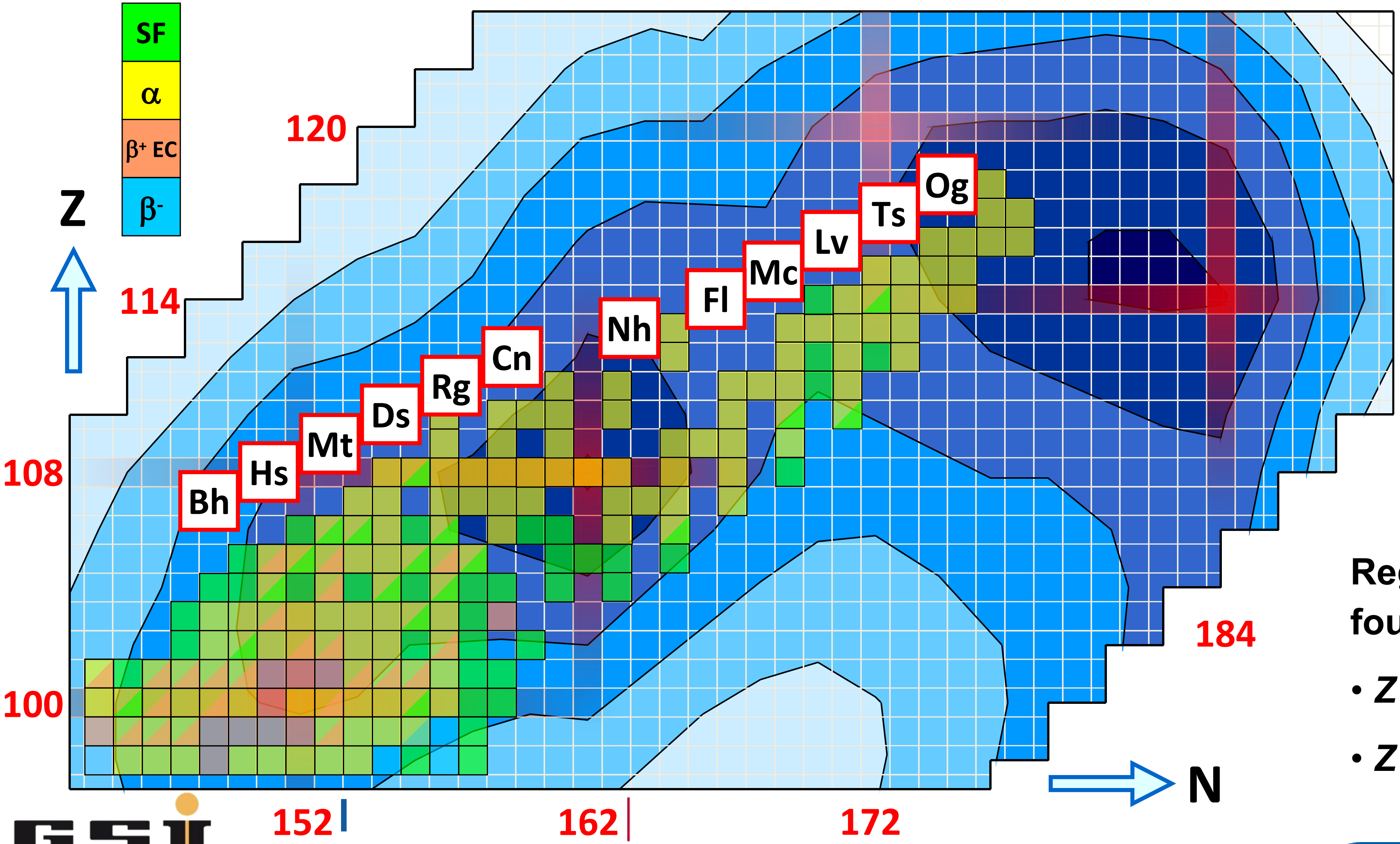


- Existence of heavy nuclei depends on interplay of repulsive Coulomb and attractive nuclear force
- Heavy nuclei often deformed with fission barrier decreasing with increasing *Z*
- Nuclear shell effects stabilize superheavy nuclei (binding energy gain up to 10 MeV)

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

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

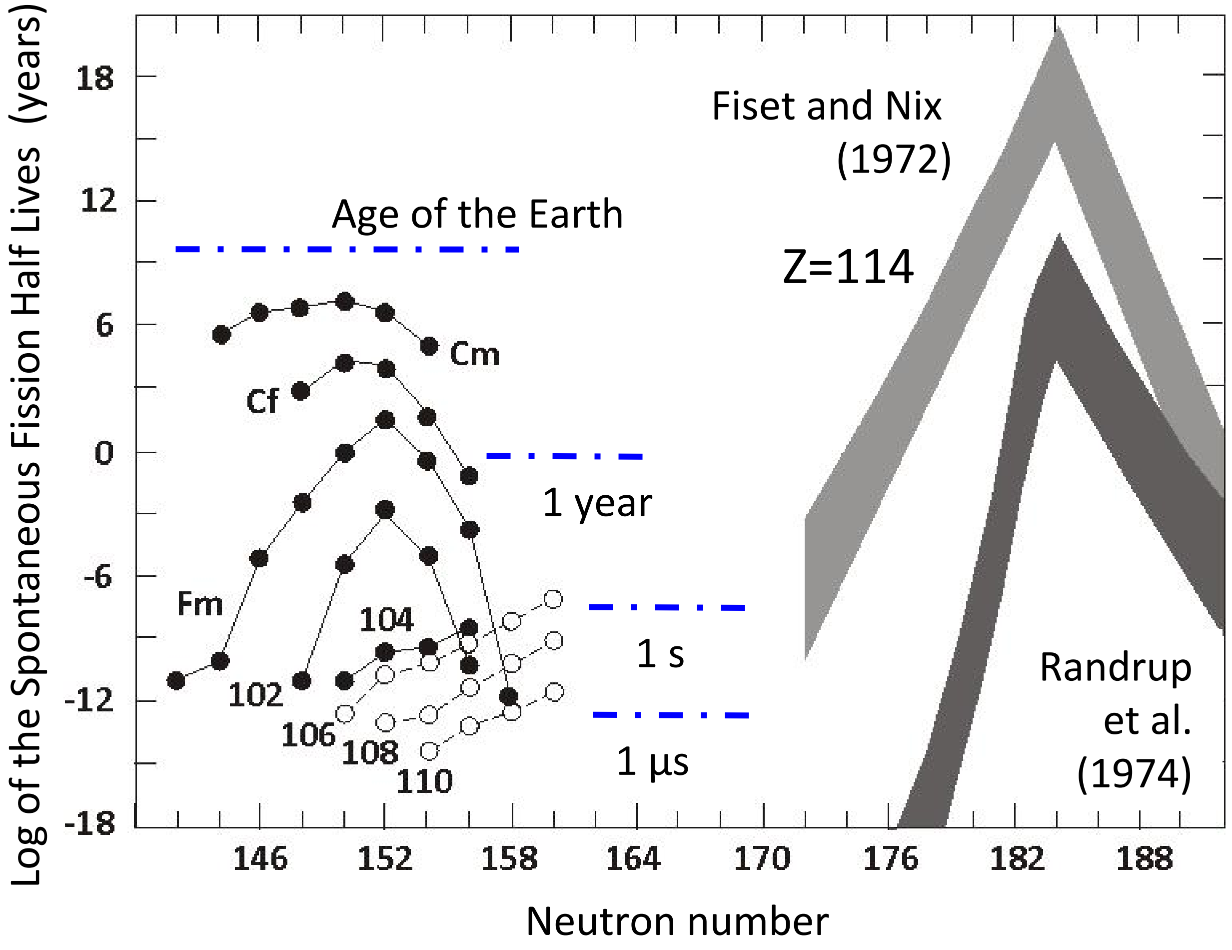
Landscape of Superheavy Nuclei



Regions of enhanced stability found for deformed nuclei with:

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

Predictions in 1970s: Island of Stability



- superheavy nuclei with Z ≈ 114 and N ≈ 184 predicted to form **„Island of Stability“**
- theoretical models predict:
  $T_{1/2}(SF) > 10^9$ years
- initiate search for existence in nature
  – until now no evidence

Figure Courtesy Yuri Oganessian

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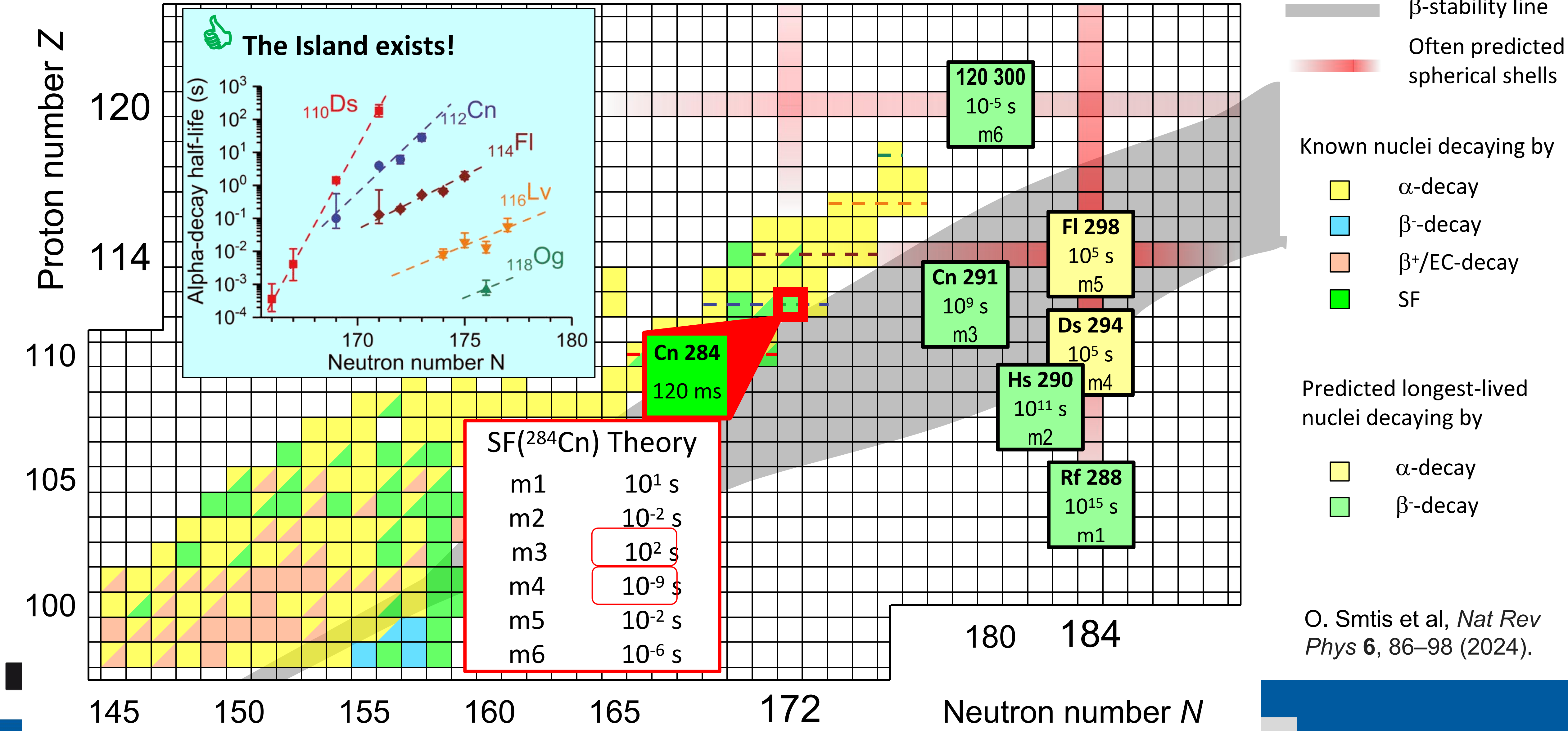
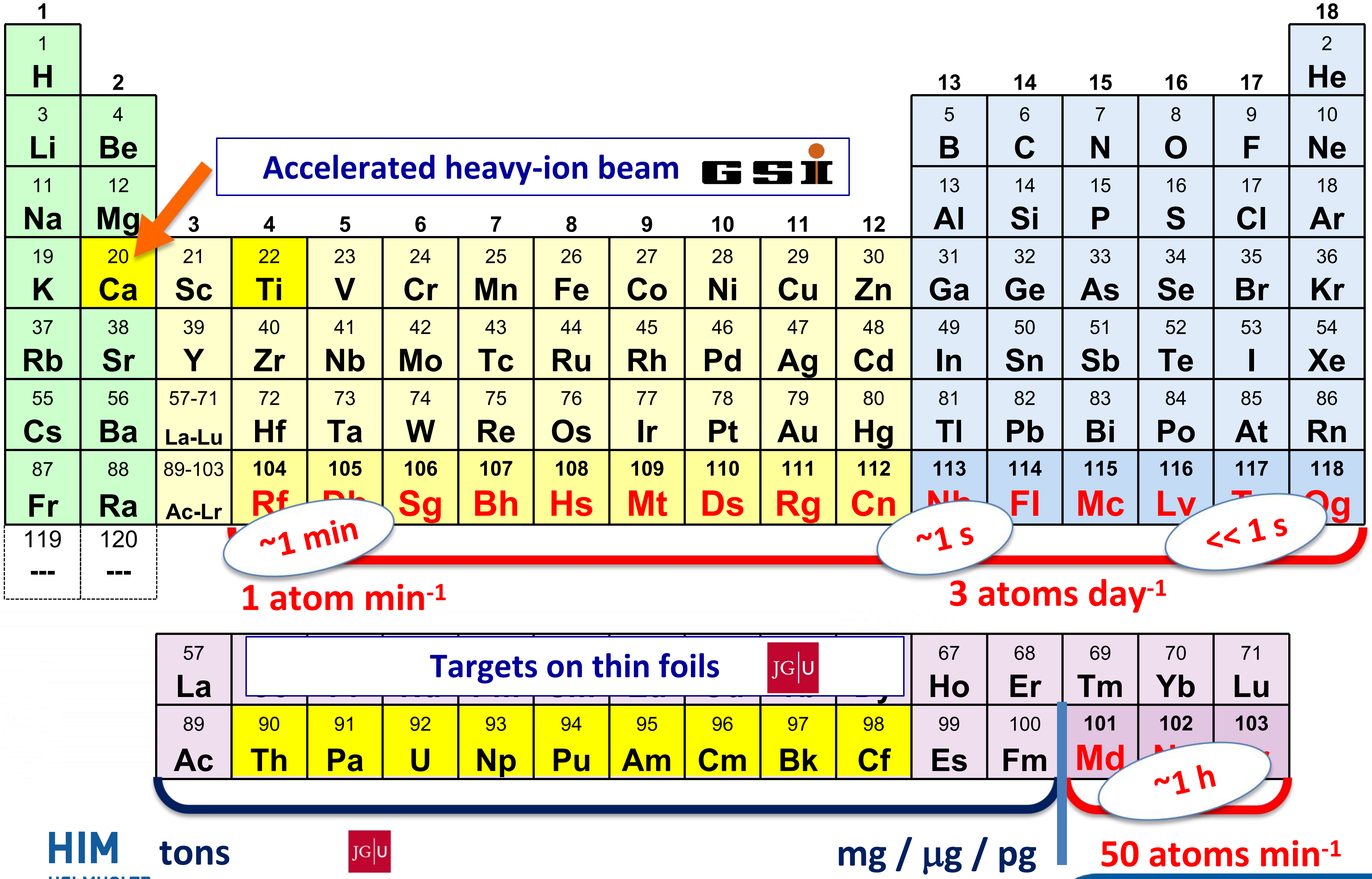
Island of Stability – Status Today



O. Smtis et al, *Nat Rev Phys* **6**, 86–98 (2024).

Experimental Challenges

Availability of Superheavy Elements



Courtesy
Ch. Düllmann

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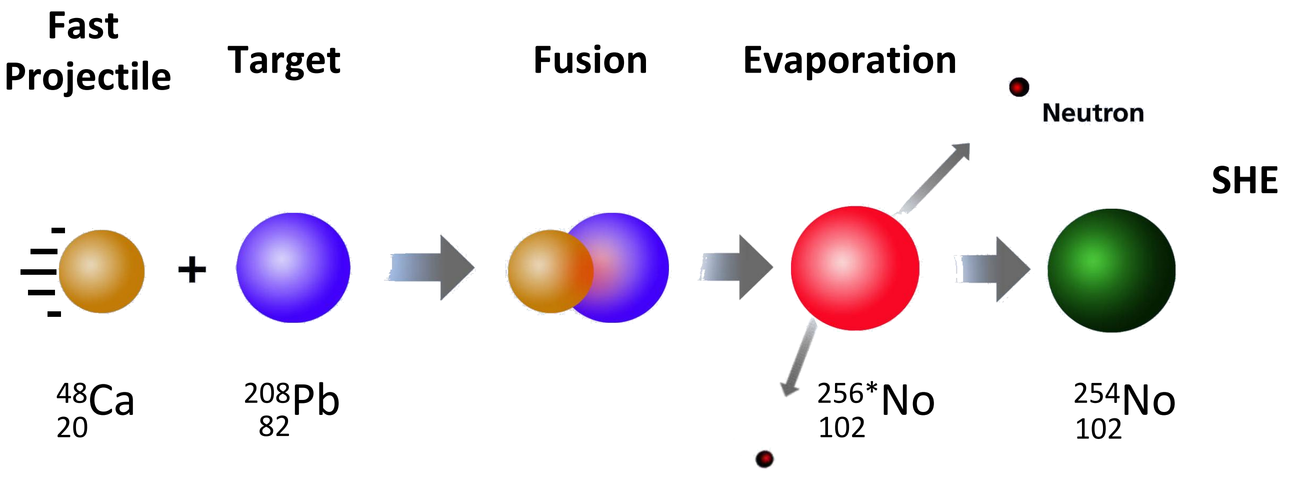
Production in Fusion-Evaporation Reactions

high-intensity beam:
6 x 10^{12} particles
per second



Yields:
1 /s (Z≈102)
to
< 1 /week (Z≥112)

- production in heavy-ion induced fusion-evaporation reactions
- besides low production rates, high energy of reaction products poses challenges for mass spectrometry and laser spectroscopy
- utilizing buffer-gas cells for slowing down and thermalizing

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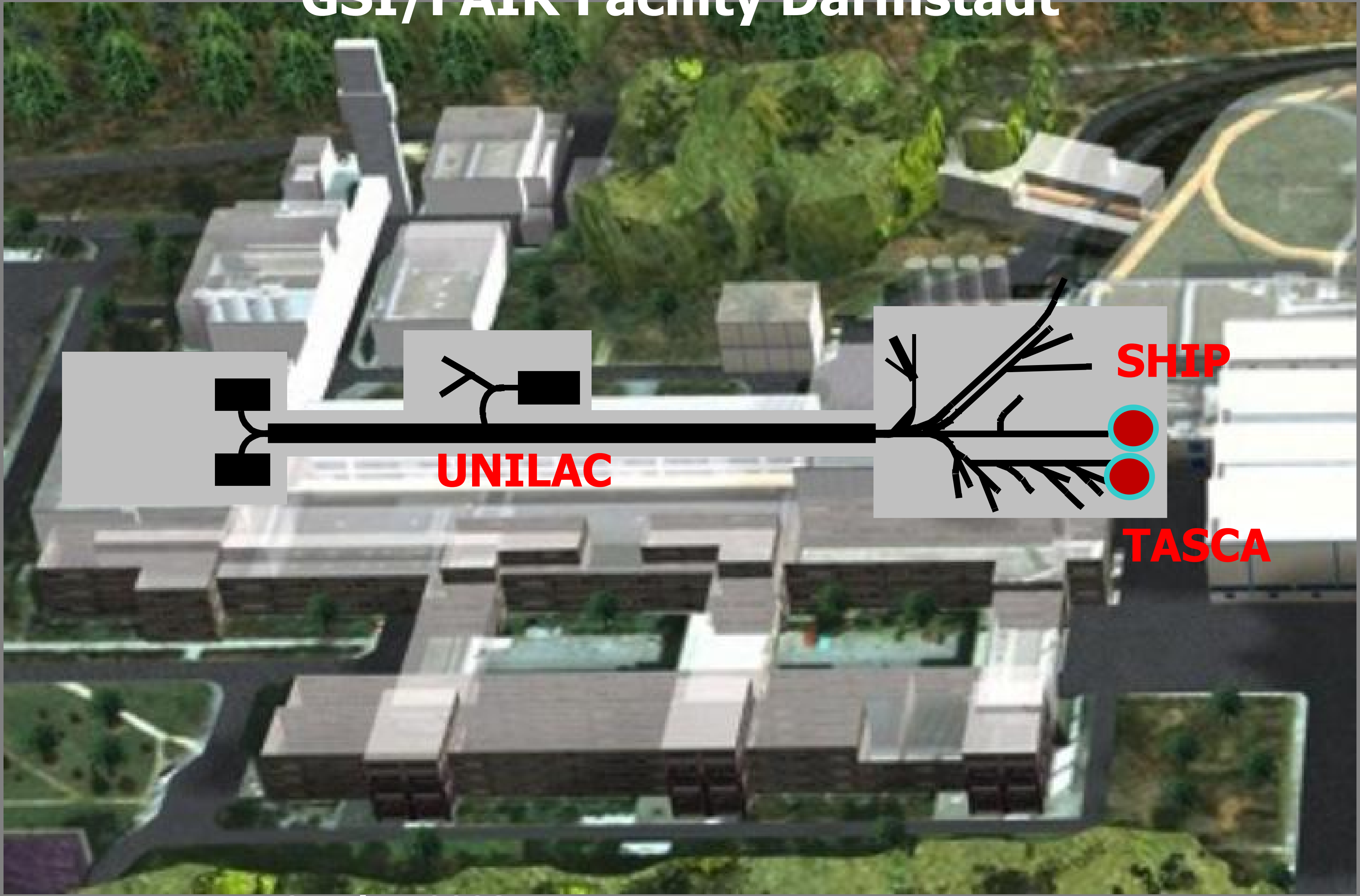
GSI/FAIR Facility Darmstadt



October 2023

Mass spectrometry of the heaviest elements

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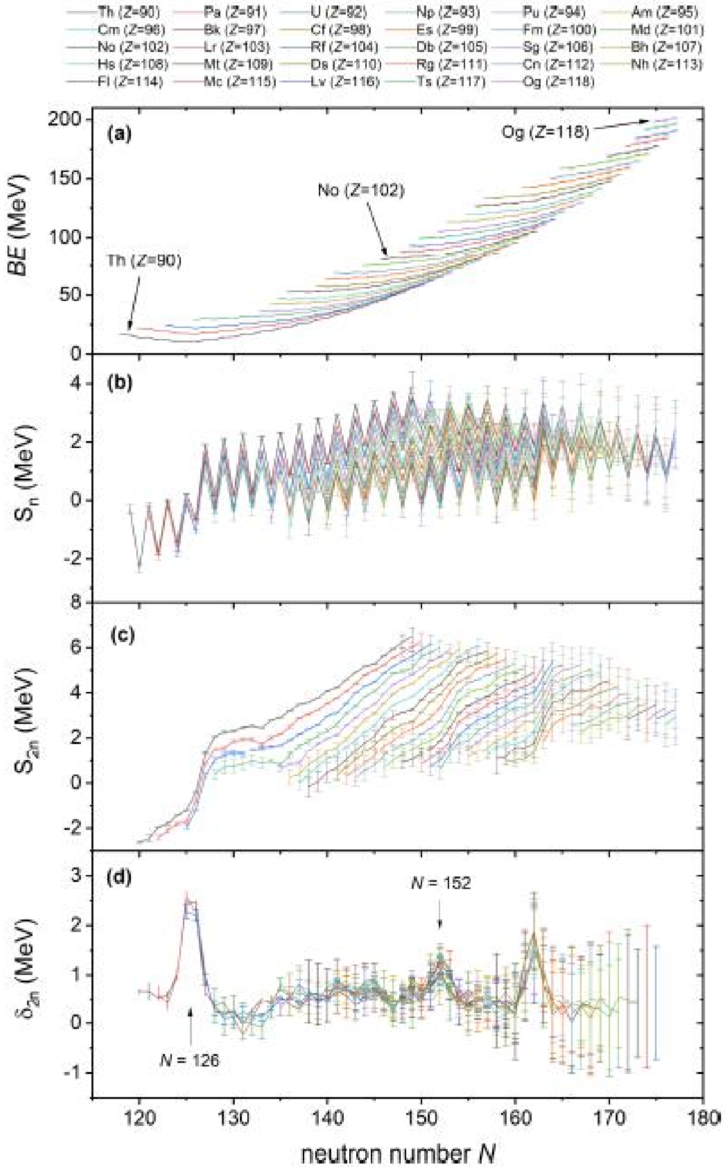
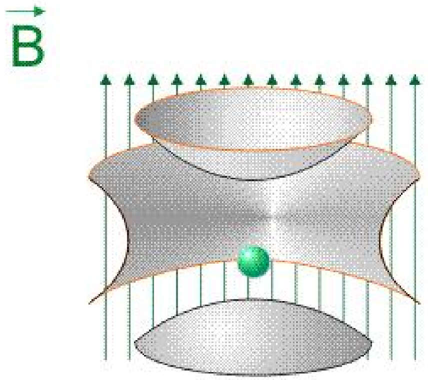
Masses and Nuclear Shell Structure

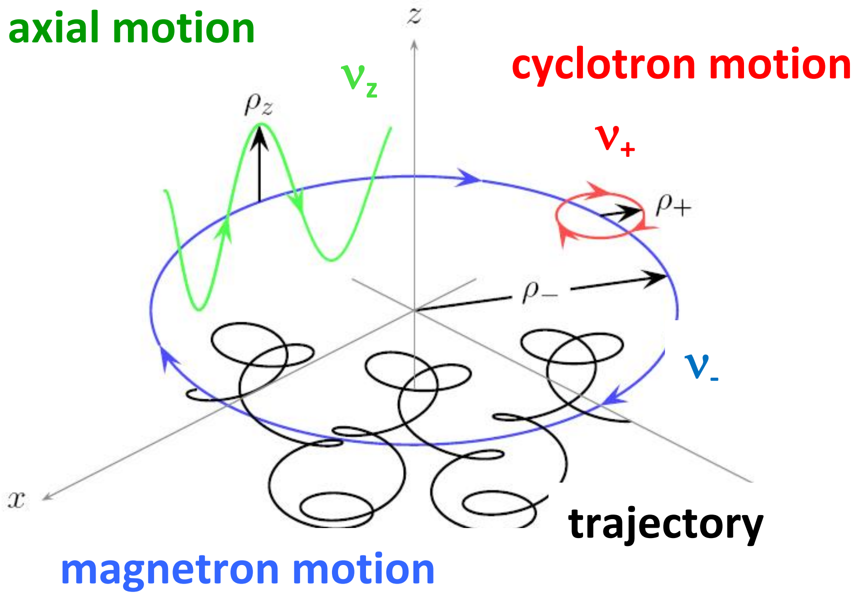


- nuclear masses and mass differences reflect nuclear shell structure
- signatures of shell closures, nuclear pairing, and the onset of deformation can be observed
- precision of experimental masses few keV or better even for many exotic nuclei
- mass data show deformed shell gaps in SHN at N = 152 and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

Figure courtesy O. Kaleja

Penning Trap Mass Spectrometry



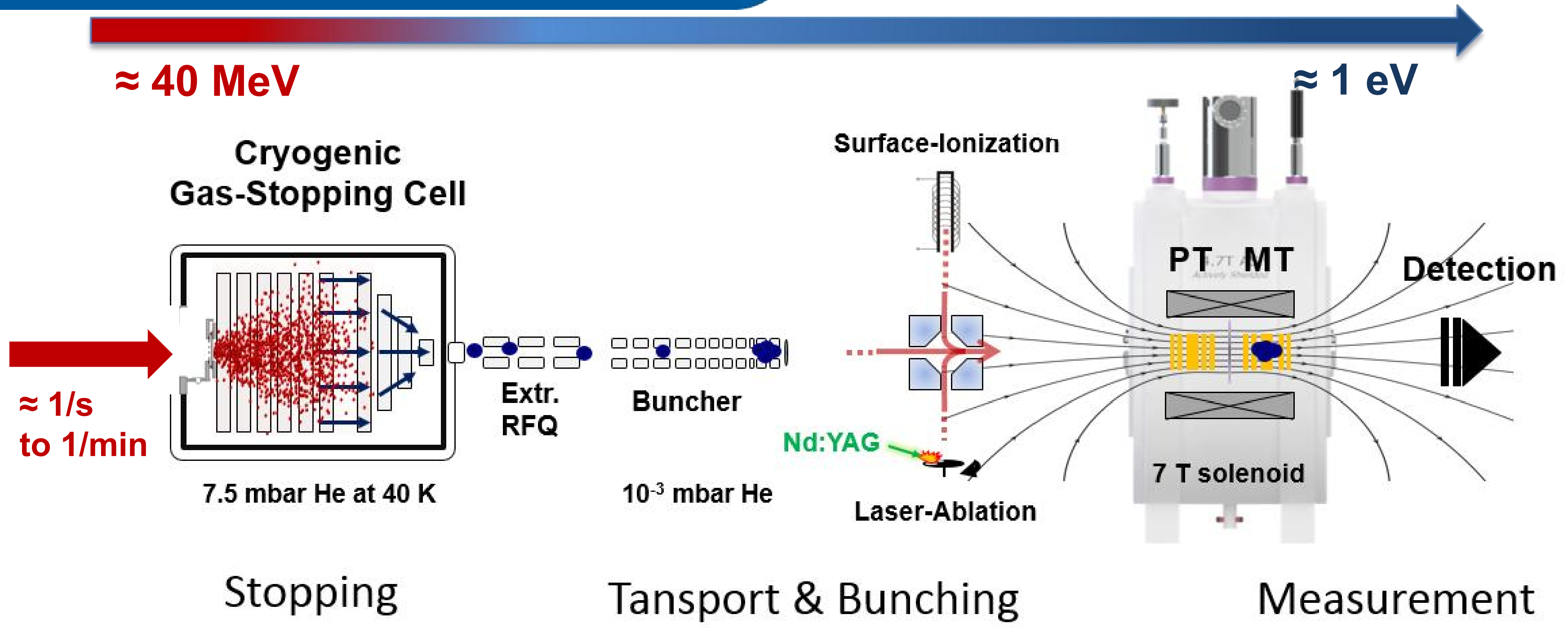


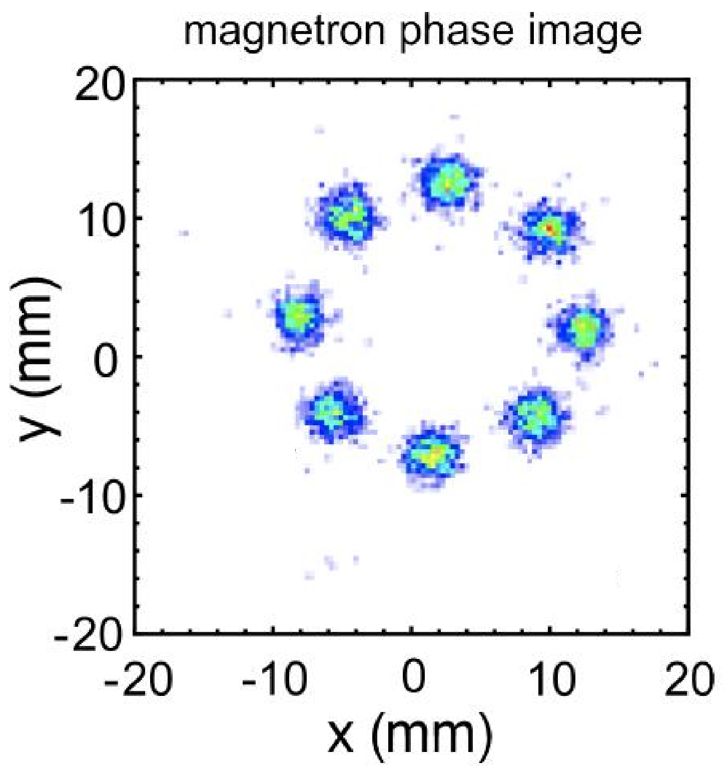
$$f_c = \frac{1}{2\pi} \cdot \frac{q}{m} \cdot B$$

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

- confine ion with mass *m* and charge *q* by homogenous *B* field and electrostatic quadrupole field
- magnetic field calibration by reference ions with well-known mass
- measurement of cyclotron frequency yields mass value

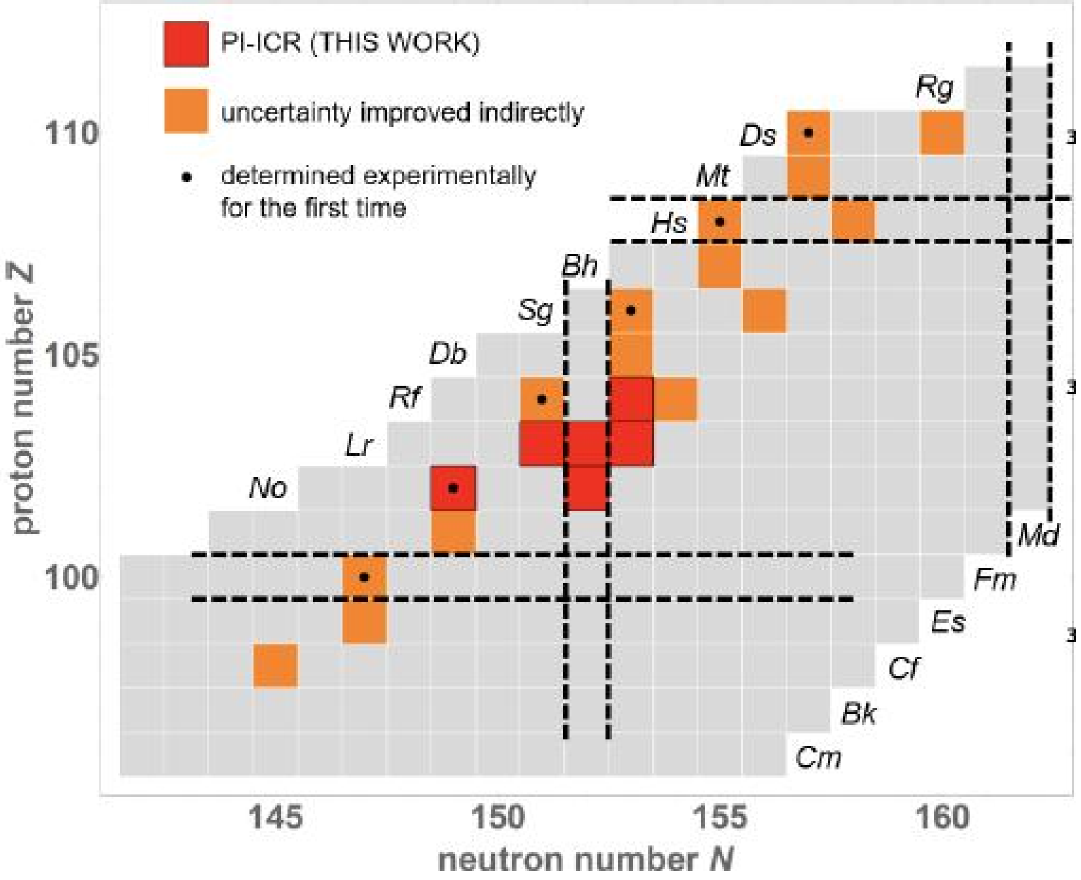
SHIPTRAP Setup at GSI Darmstadt





SHIPTRAP Coordination:
Francesca Giacoppo (GSI)

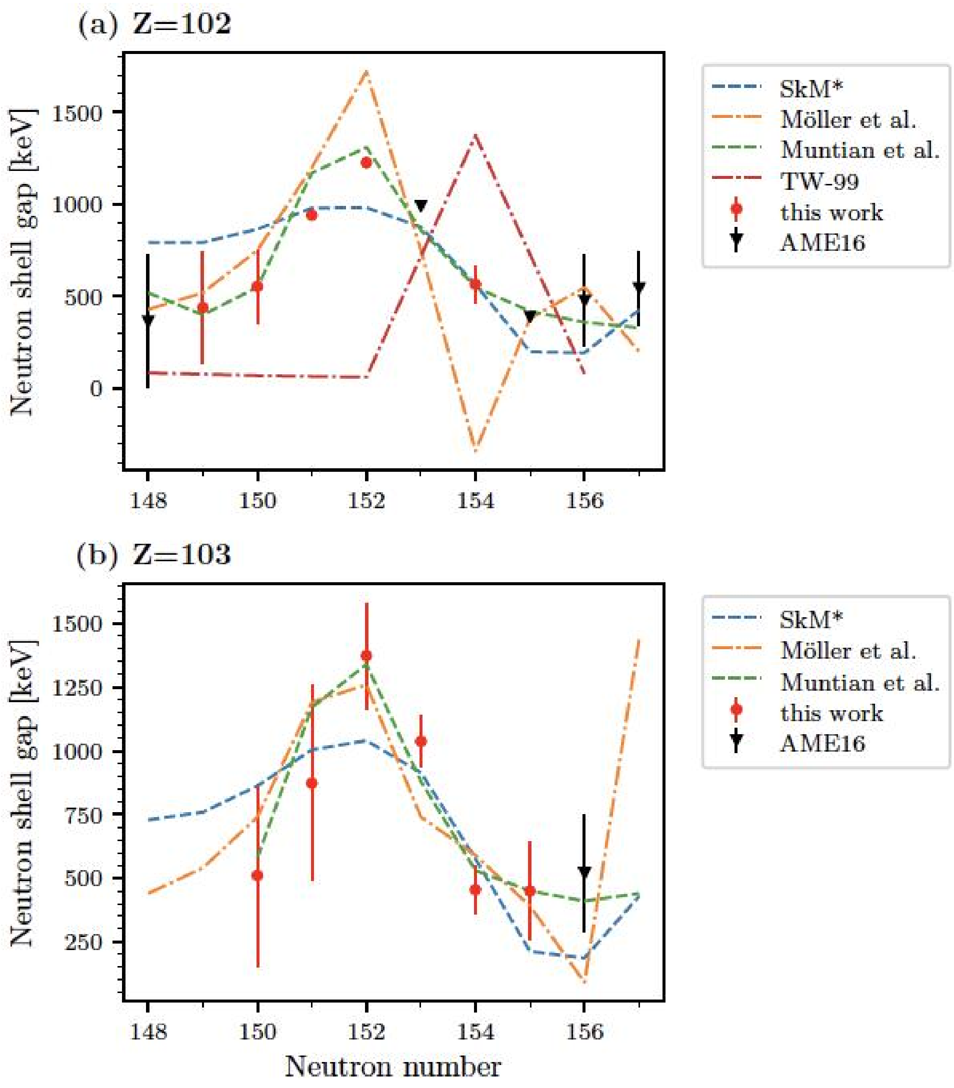
Recent SHIPTRAP Results



- masses of nobelium, lawrencium, and rutherfordium isotopes measured with keV-precision
- rel. mass uncertainty of 10^{-8} and better
- data were implemented in Atomic Mass Evaluation (AME) and showed good consistency
- mass values of several additional nuclides improved indirectly via the AME network's links

O. Kaleja, Phys. Rev. C (2022) 054325 and PhD thesis JGU Mainz

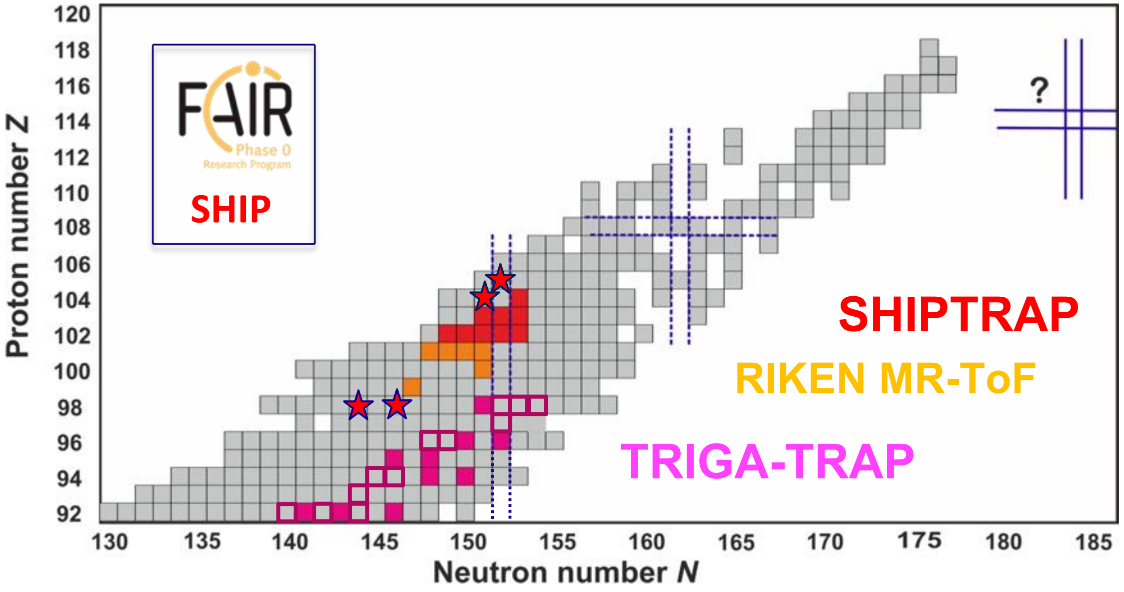
Comparison of Results to Nuclear Models



- experimental data form SHIPTRAP show deformed shell gap at N=152
- nuclear models reproduce the general trends relatively well
- absolute masses are sometimes off by up to 1 MeV
- mass differences may still be described with reasonable precision

E. Minaya Ramirez et al. Science 337, 1207 (2012)
M. Block et al., Nature 463, 785 (2010)

Masses of Heavy Nuclei - Status



- direct mass spectrometry $Z > 100$ established: SHIPTRAP/GSI 2008
- mass measurements to study nuclear shell structure around $Z = 100$, $N = 152$
- measurements performed with detected rates of ≈ 0.00002/s and 5 detected ions
- high mass resolving power of SHIPTRAP allows identifying nuclear isomers

TRIGA-TRAP, JGU Mainz:

- M. Eibach et al., Phys. Rev. C 89, 064318 (2014)

RIKEN/KEK (Japan):

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

SHIPTRAP measurements, GSI Darmstadt:

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

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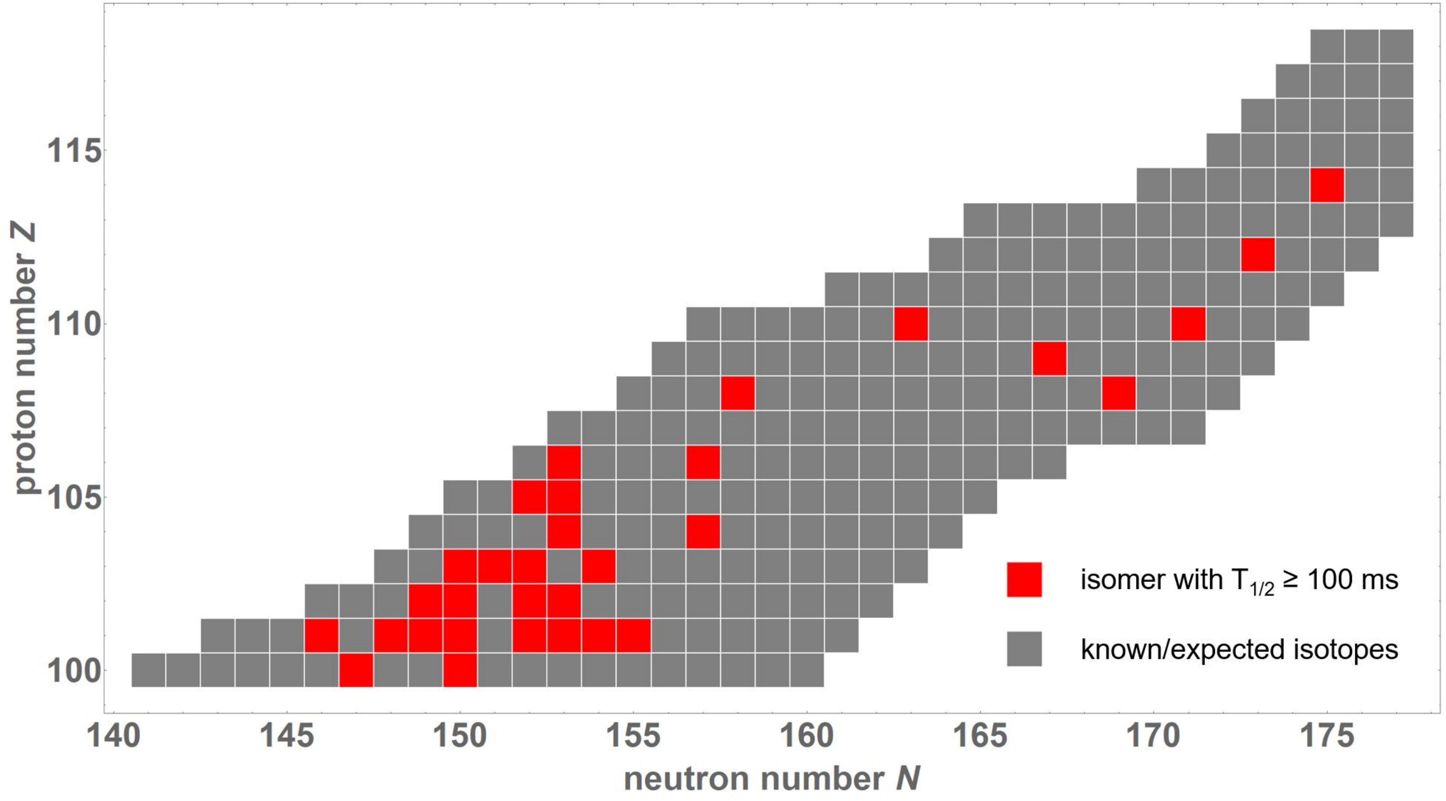
Long-Lived Isomers in the Heaviest Elements



- several (long-lived) isomeric states known, further may exist
- many of these are difficult to observe experimentally (low yield)
- Penning-trap mass spectrometry well suited to locate such isomers due to high mass resolving power

Figure courtesy O. Kaleja

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Long-Lived Isomers in the Heaviest Elements

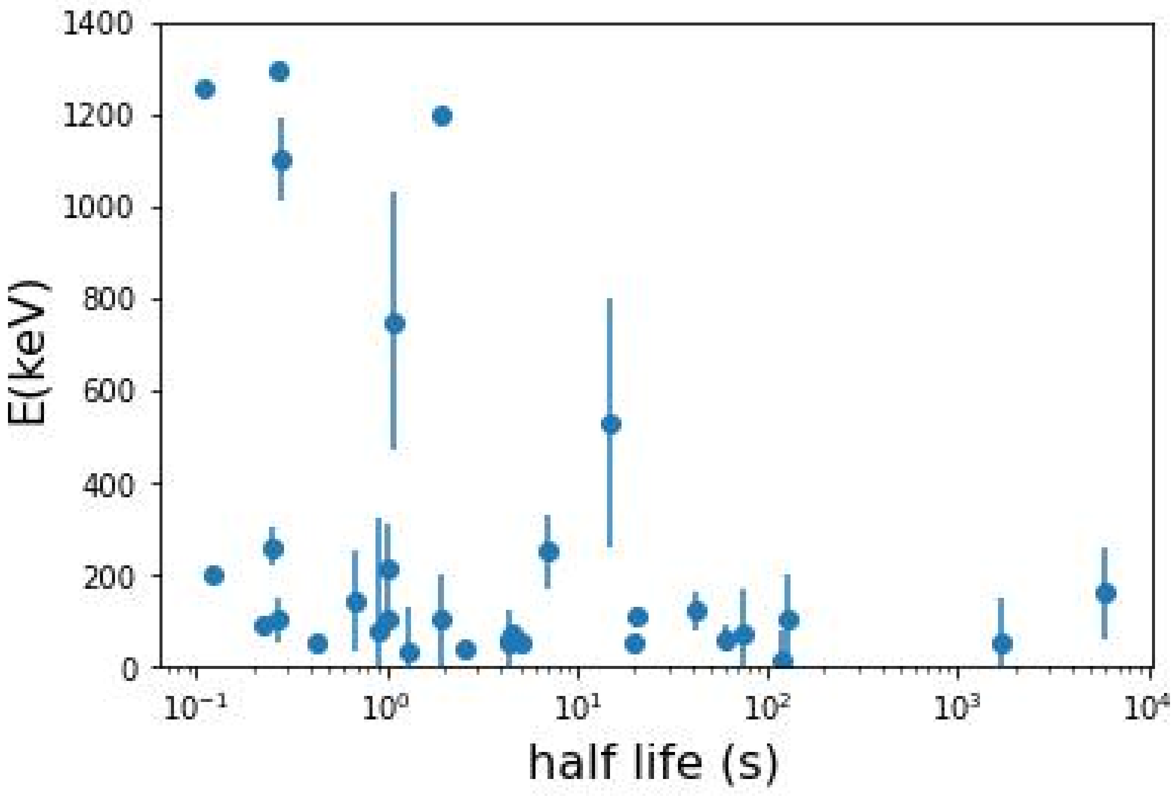

Figure courtesy O. Kaleja

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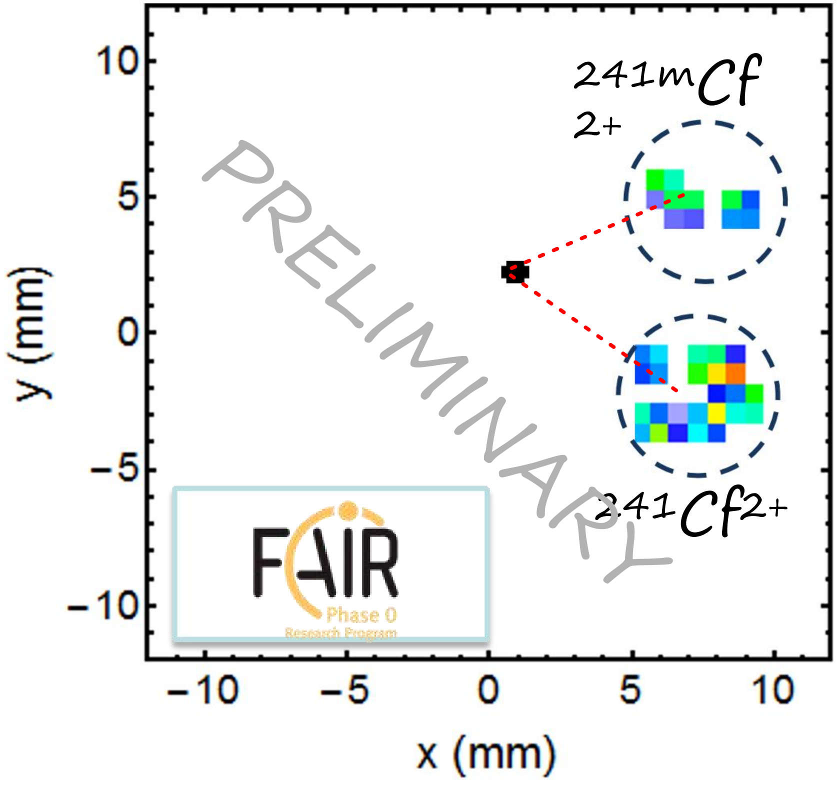
Long-Lived Isomer in ^{241}Cf (Z=98)

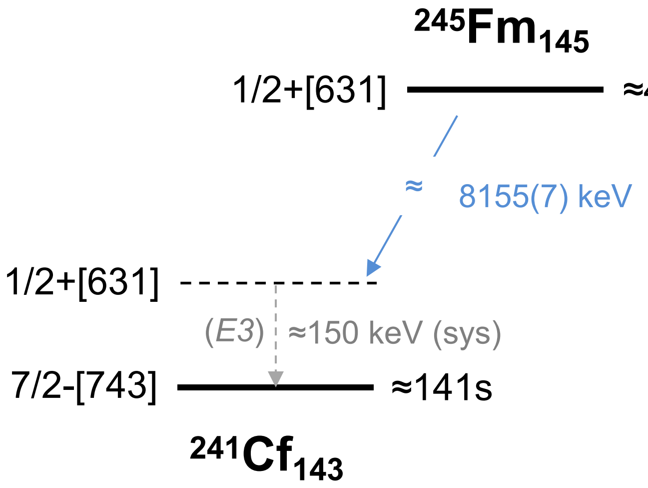
SHIPTRAP beamtime 2021



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

nuclear decay spectroscopy at SHIP
(J. Khuyagbaatar *et al.*, PRC (2020) **102,** 044312):

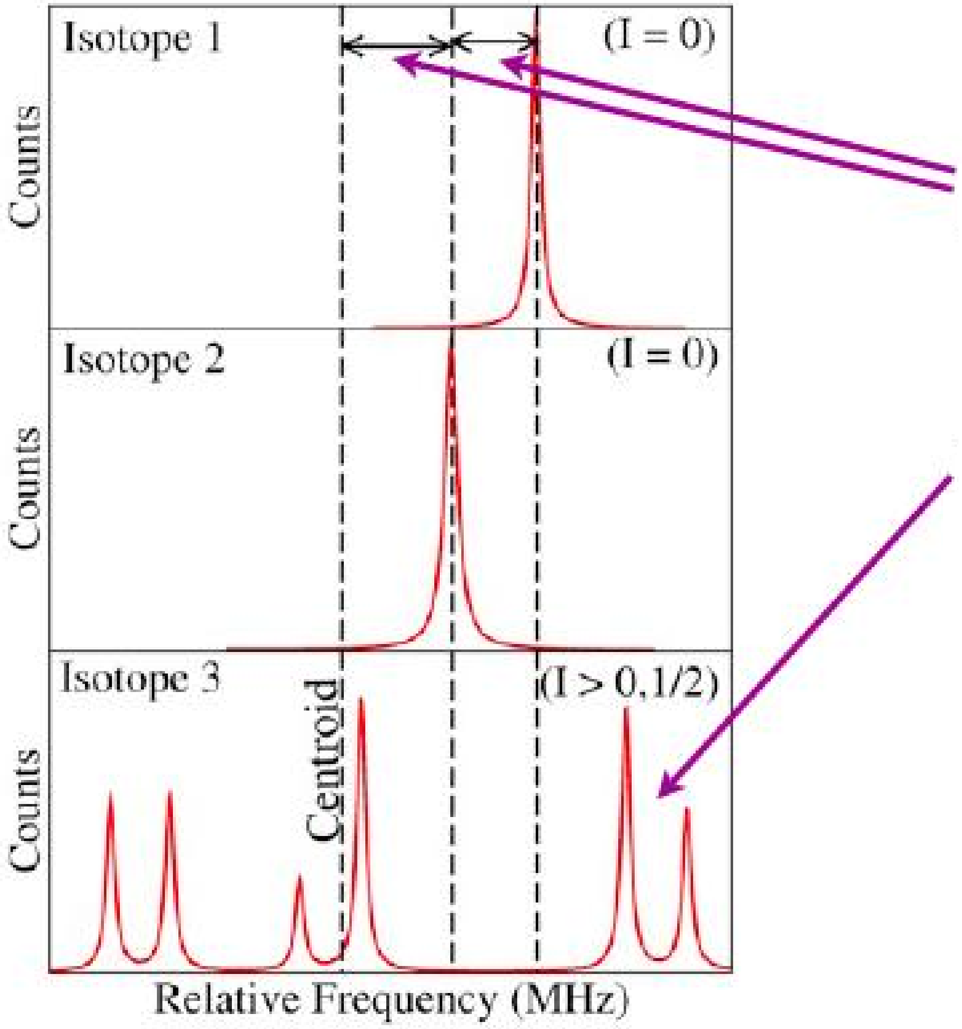
- systematics of lighter N = 141,143 isotones suggest existence of isomeric state in ^{241}Cf at ≈ 150 keV



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

Laser spectroscopy of the heaviest elements

Laser Spectroscopy of Radionuclides

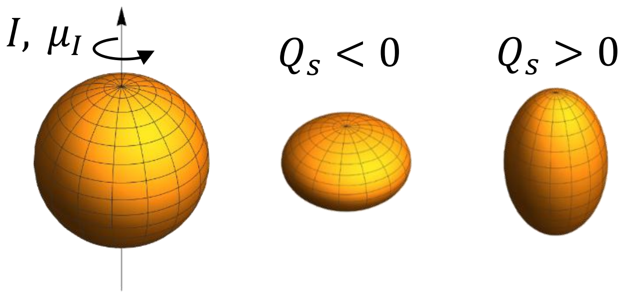


Isotope shift yields information on changes in mean-square charge radii from which we infer nuclear size

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

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

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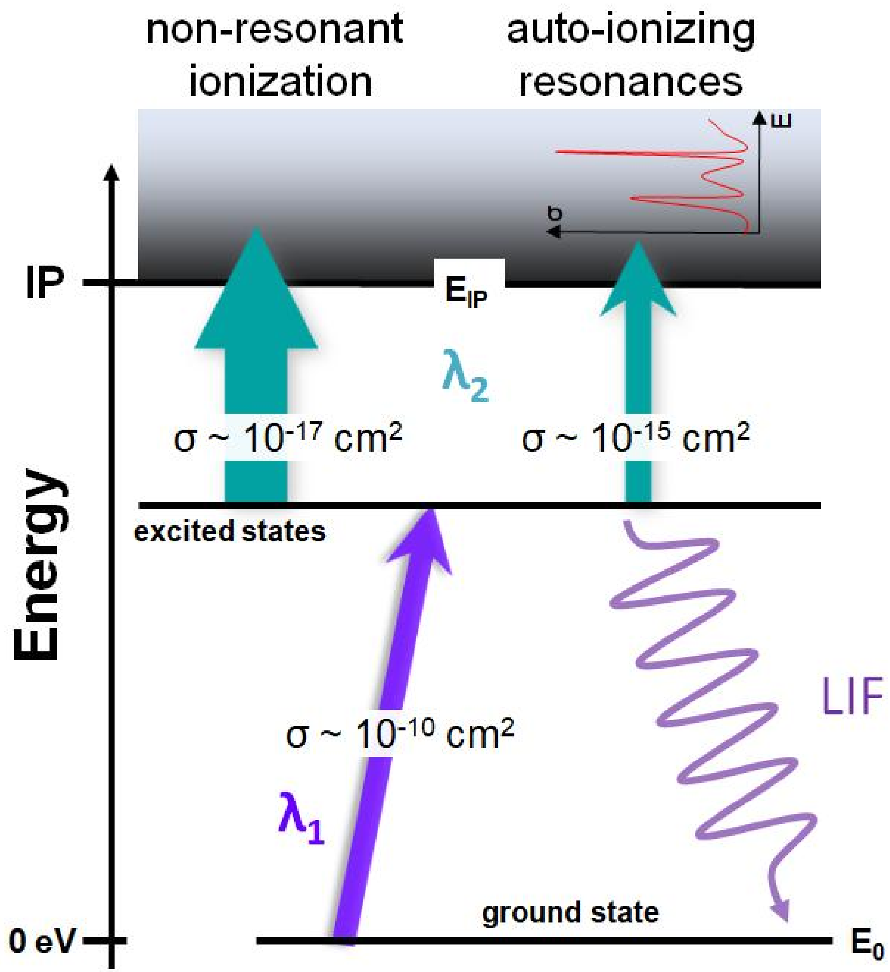
Resonance Ionization Laser Spectroscopy (RIS)



RIS method features:

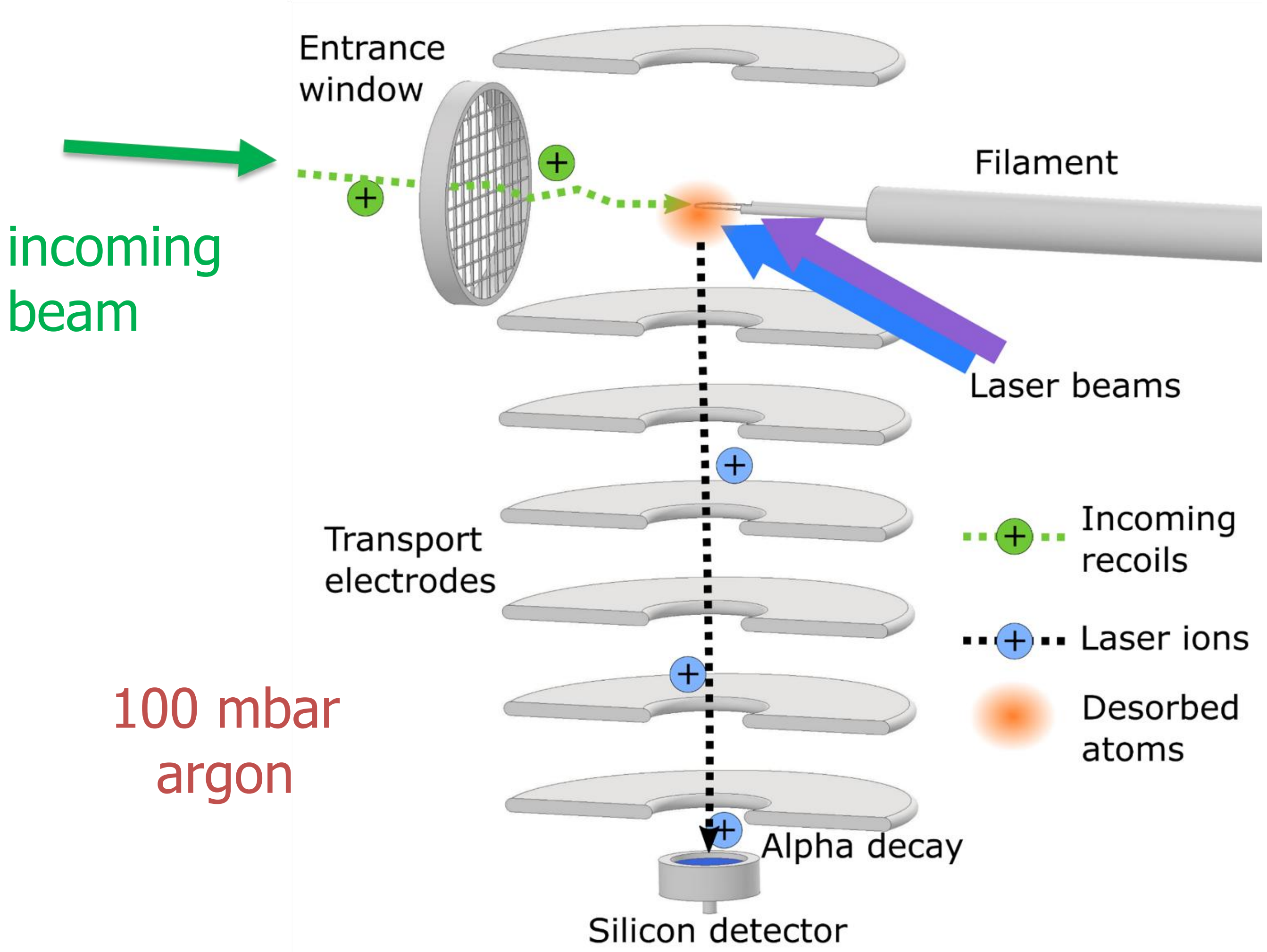
- high sensitivity
- high selectivity
- high efficiency

challenges for heaviest elements:

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

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

Radiation Detected Resonance Ionization Spectroscopy



tailored method for measurements of actinide isotopes produced by fusion reactions with lowest rates:

- slow down in Ar gas
- Neutralize on filament
- re-evaporate atoms
- two-step photo-ionization
- transport laser ions to detector and 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)

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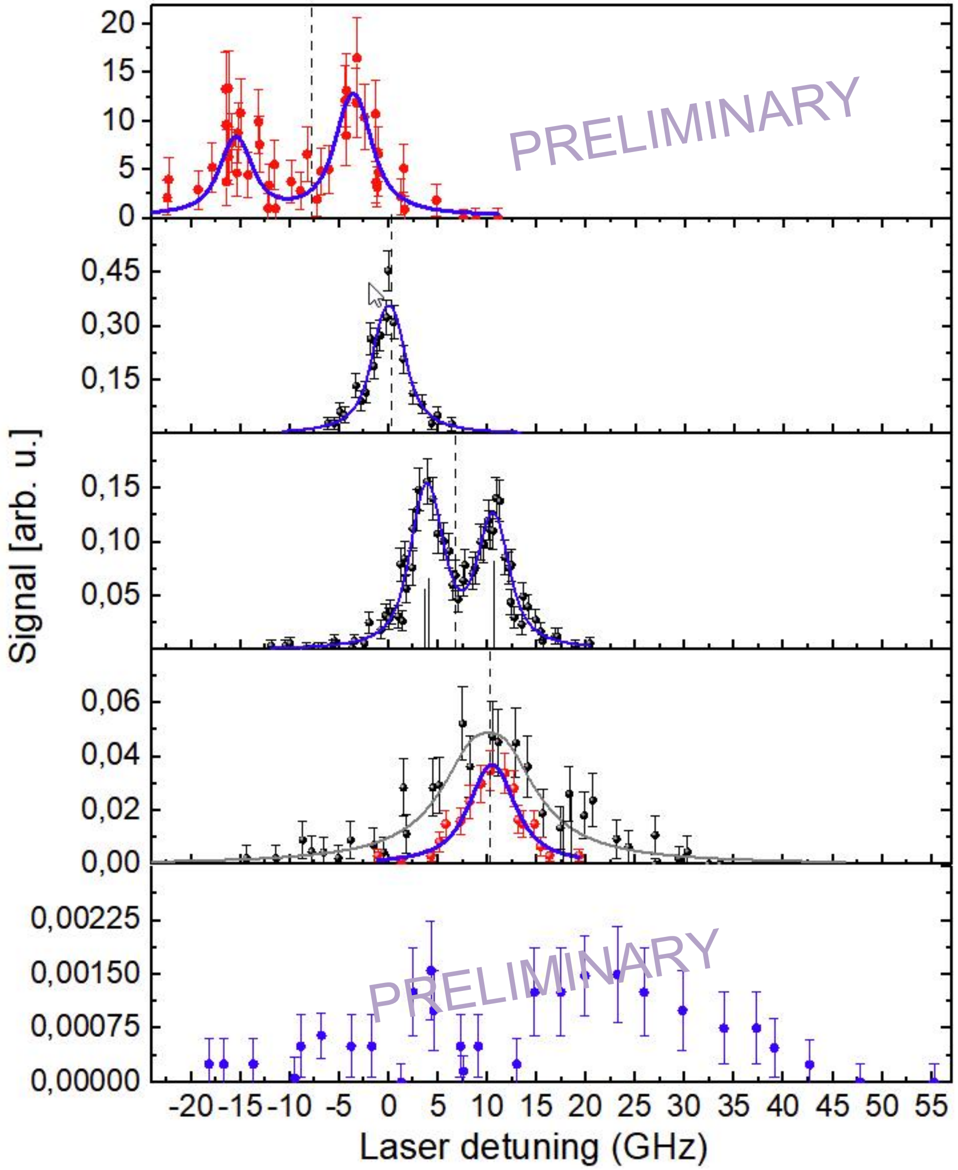
Laser spectroscopy

of fermium and nobelium

Laser Spectroscopy of Nobelium (Z=102) Isotopes



Experiment: S. Raeder *et al.*

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

FAIR Phase 0 Research Program

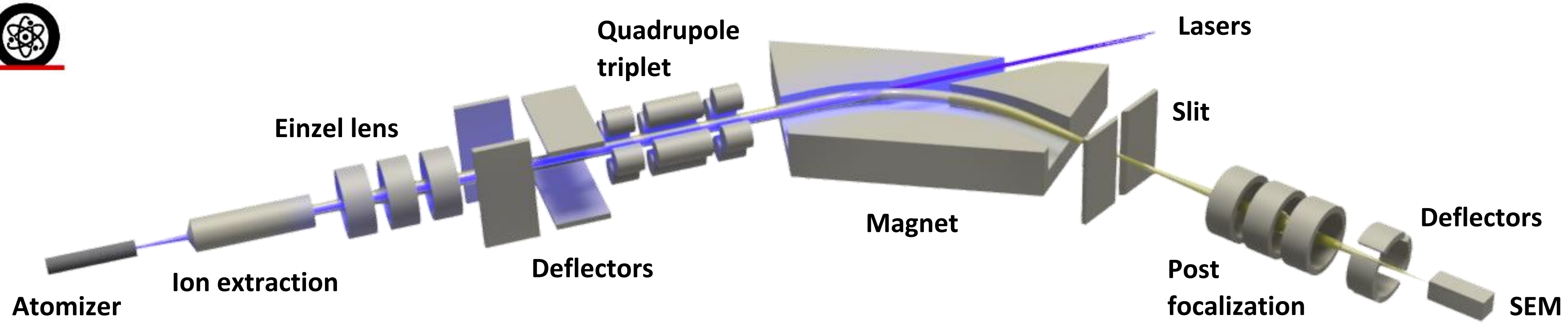
SHIP

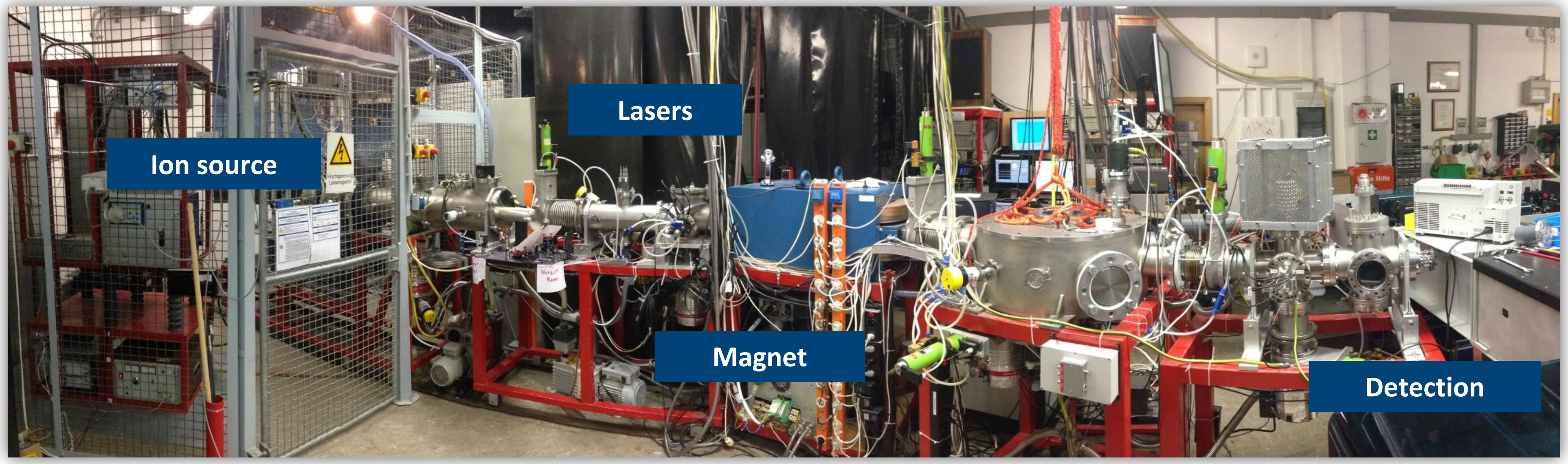
- first laser spectroscopy spectroscopy beyond Z=100
- yield as low as 0.05 atoms / second
- isotope shift allowed determining changes in mean-square charge radii around *N* = 152
- magnetic dipole and electric quadrupole moment of ^{253,255}No obtained from hyperfine splitting

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

RISIKO setup at Mainz University

Prof. K. Wendt





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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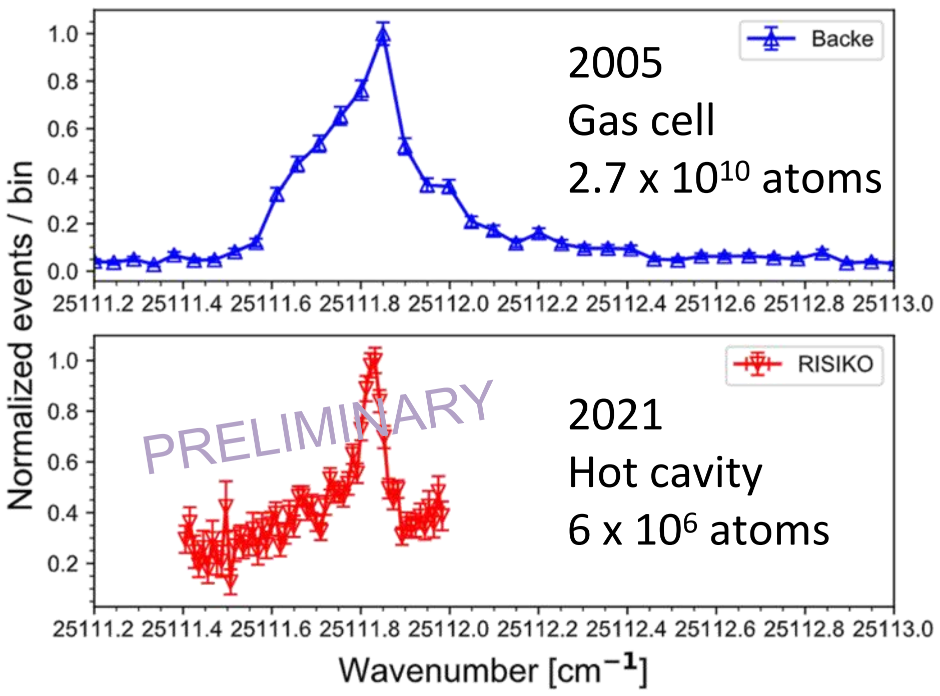
Laser Spectroscopy of Fm Isotopes



- first studies of ^{255}Fm ($t_{1/2}$=20h) by M. Sewtz, H. Backe et al. with variant of RADRIS method in gas cell in 2003
- 8 atomic level in Fm identified

RISIKO mass separator in Mainz

- production of radioactive ion beams in hot cavity with laser ionization
- laser spectroscopy om Fm samples with only 10^7 atoms
- 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.

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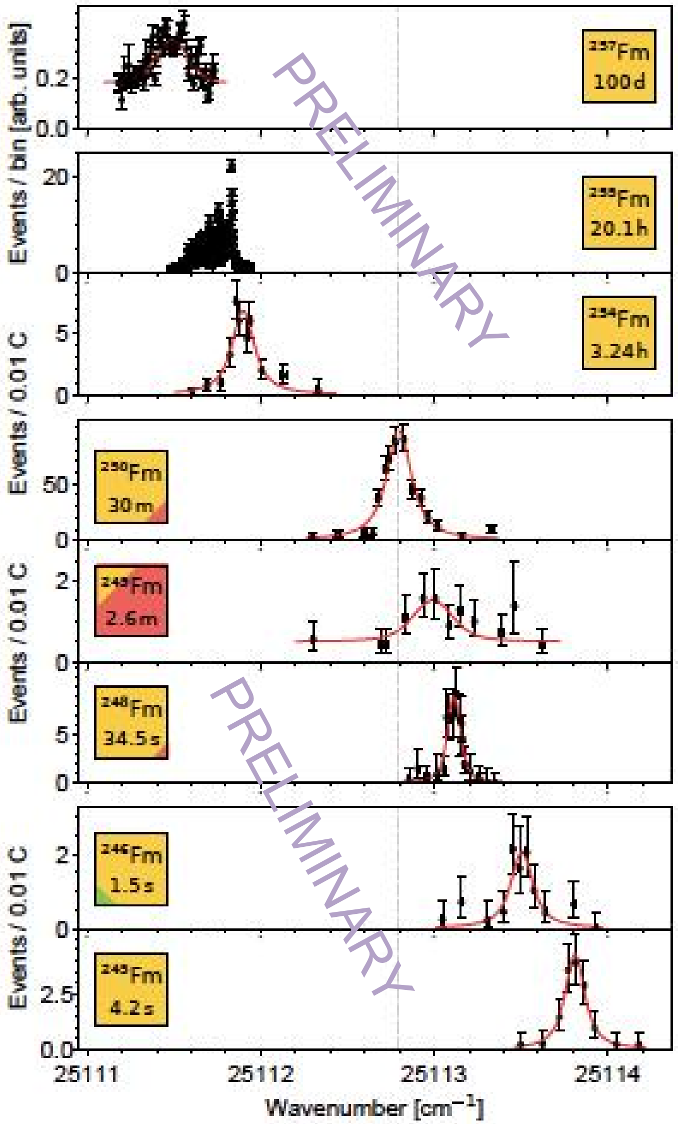
Laser Spectroscopy of Fm (Z=100) Isotopes



- short-lived fermium isotopes measured *online* at GSI
- some isotopes produced via decay of directly produced nobelium isotopes
- long-lived ^{255,257}Fm from ORNL / ILL measured *offline* at RISIKO Mainz (K. Wendtet al.) after radiochemical separation (Ch. Düllmann et al.)
- measured isotope shift in long fermium isotope chain

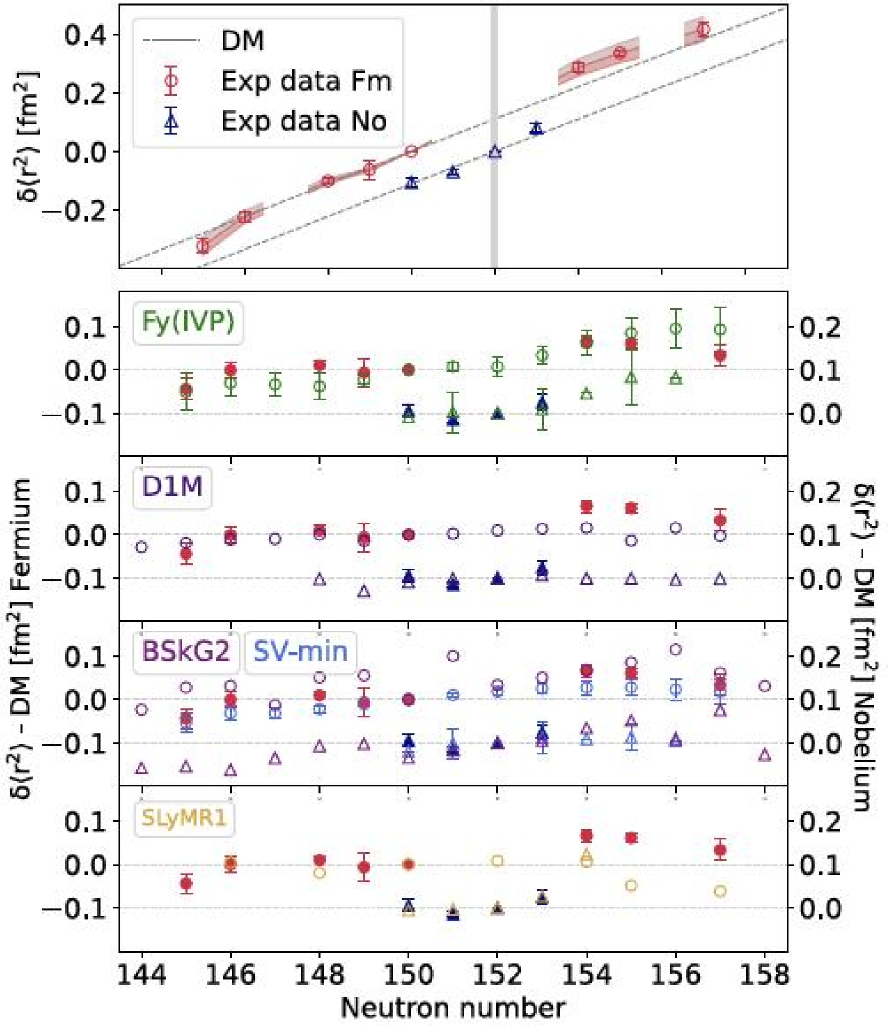
SHIP

Experiment S. Raeder *et al*.

Data analysis: S. Raeder, J. Warbinek (PhD thesis), 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.

Results – Comparison with Theoretical Calculations



calculations of charge radii using different nuclear energy density functionals by:

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

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

Summary

- Superheavy elements up to Z=118 synthesized
- Evidence for existence of *Island of Enhanced Stability*
- Limited experimental knowledge of several atomic and nuclear properties
- Pioneering experiments extended reach of mass measurements and laser spectroscopy to heavy actinides and beyond
- Technical and methodological developments crucial in this endeavor

THANK YOU FOR YOUR ATTENTION!

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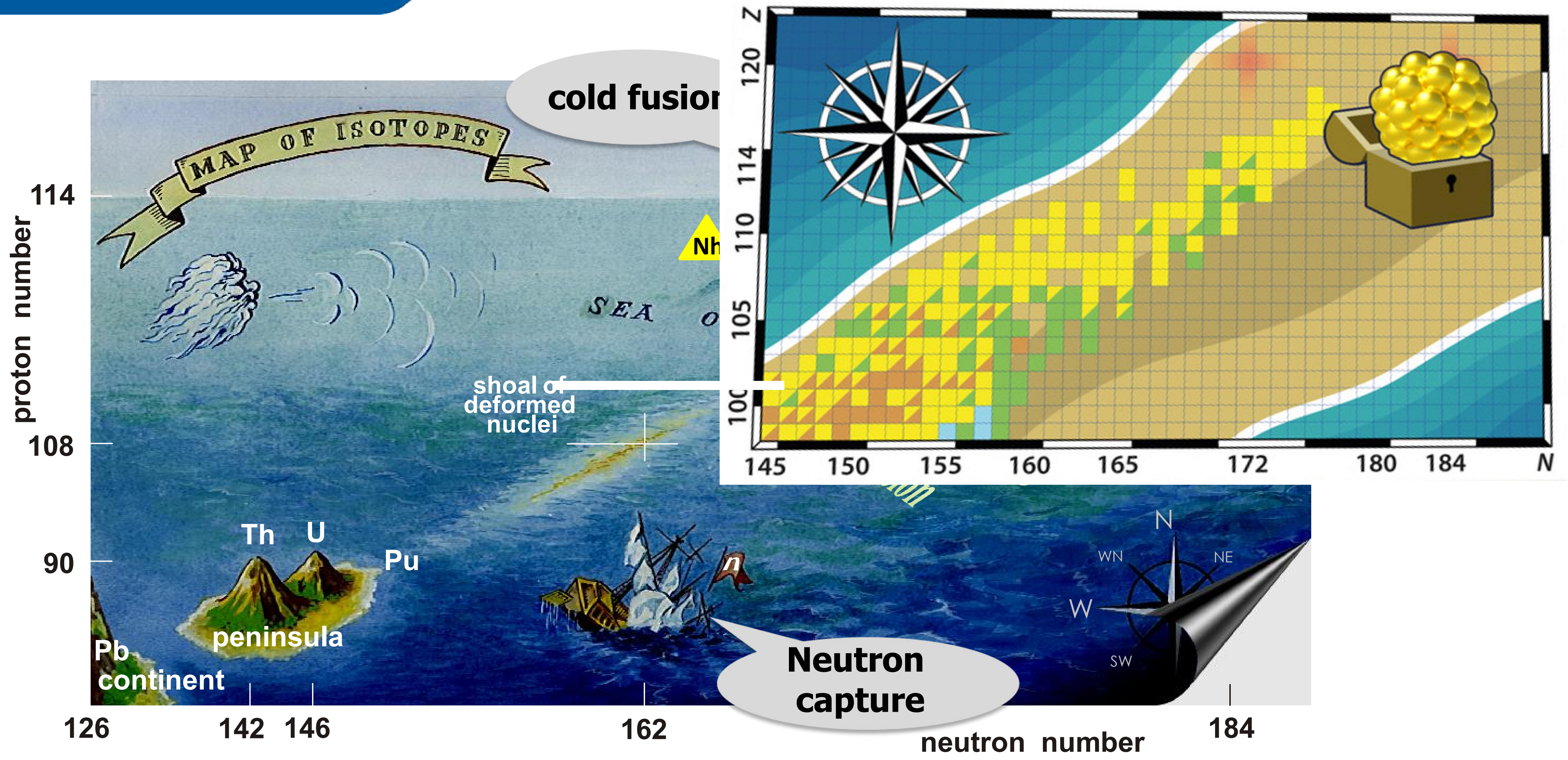
Superheavy Element Research – Open Questions

- 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

Island of Stability



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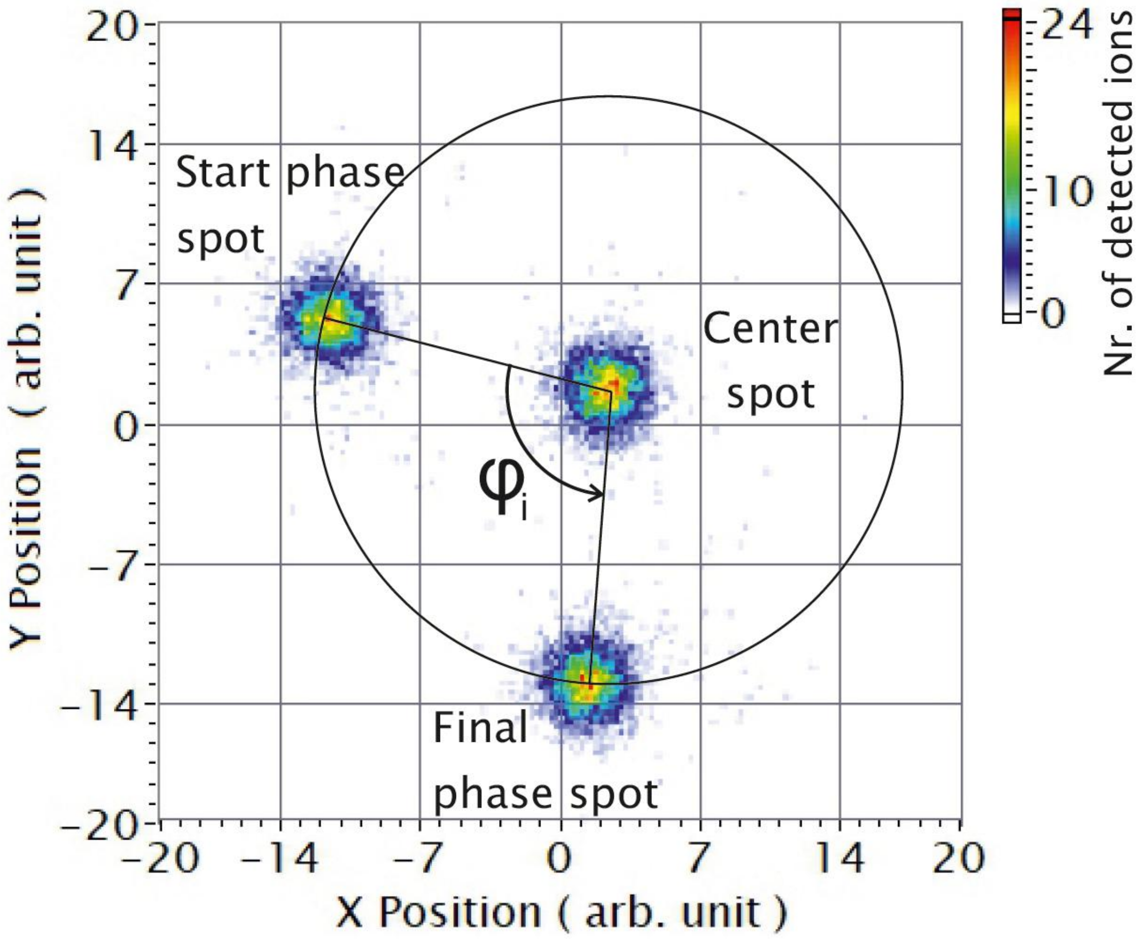
Measurement Method „PI-ICR“



- Phase-imaging method developed at SHIPTRAP/GSI around 2013
- new standard at RIB facilities world-wide
- requires fewer ions for measurement
- typical accuracy for radionuclides 5 x 10^{-9}
- very high mass resolving power enables identification of isomers

S. Eliseev et al., Phys. Rev. Lett. 110, 082501 (2013)

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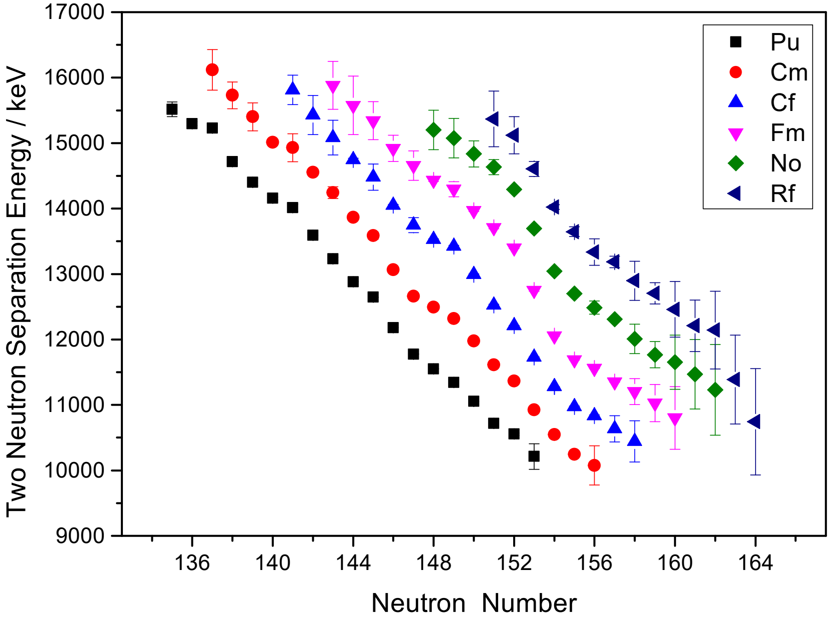
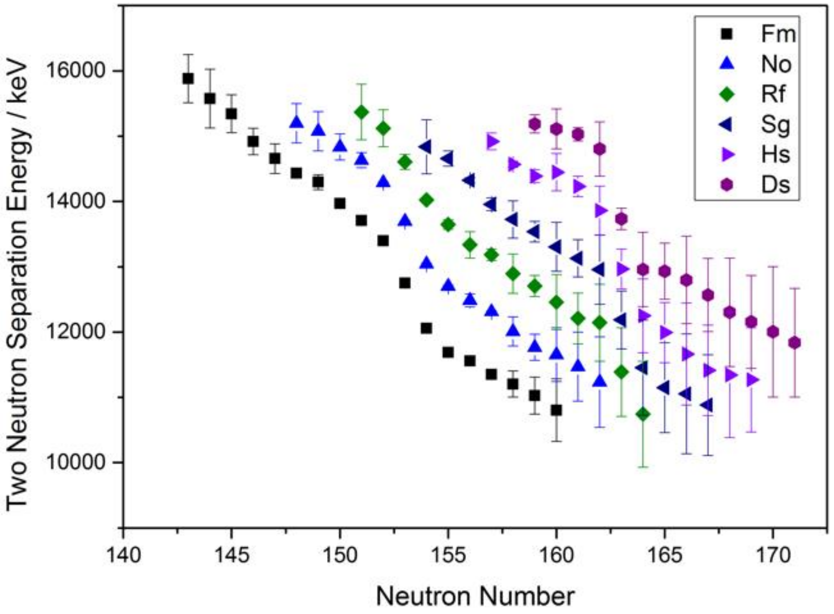
Nuclear Shell effects from Separation Energies



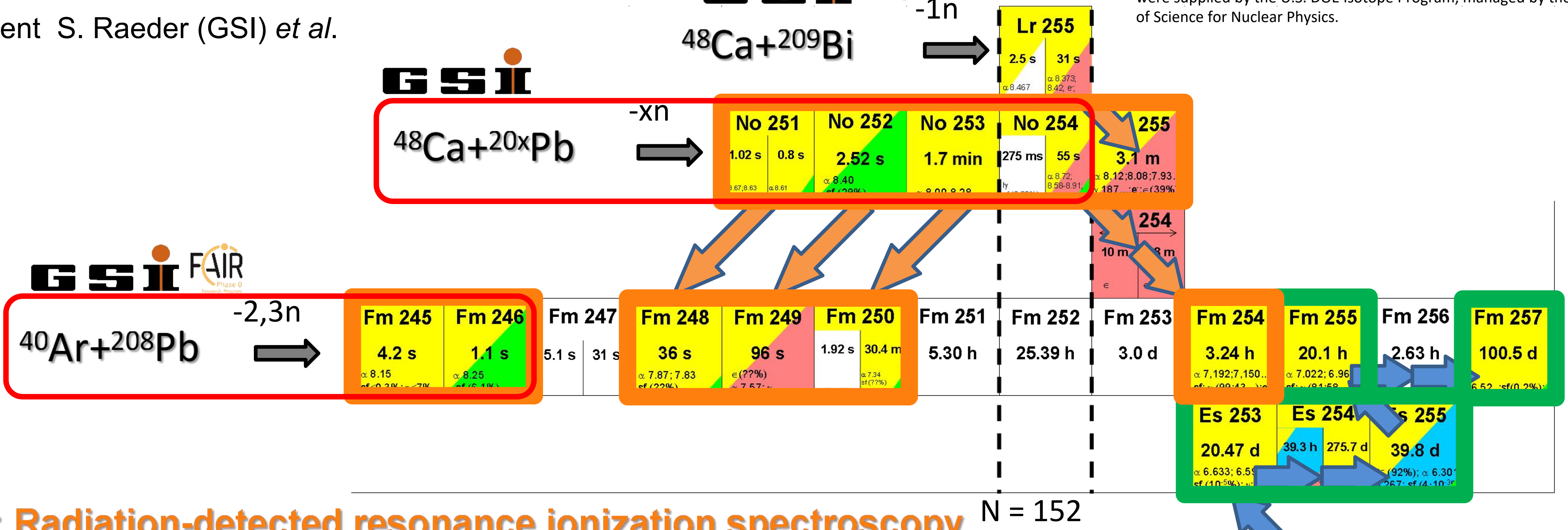


neutron separation energies derived from masses and Q_a-values show signatures of nuclear shell effects at N = 152 and N = 162

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

From HFIR

Es reirradiated at