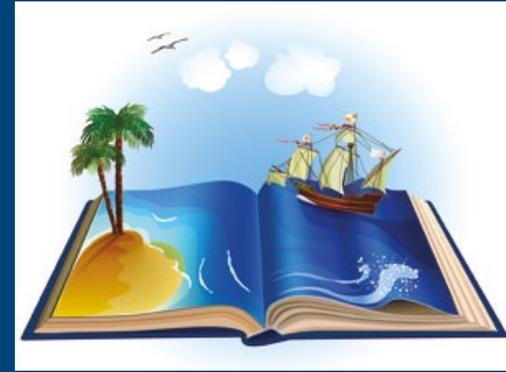
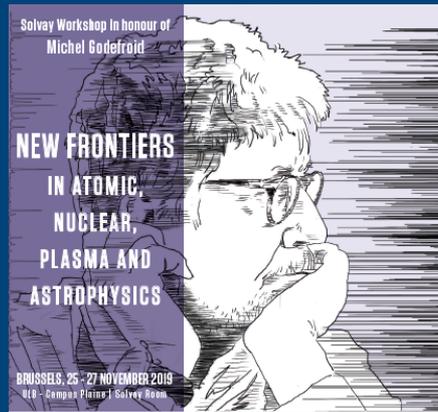
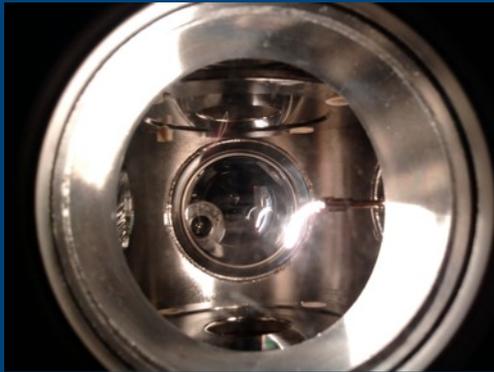




Laser spectroscopy of heavy and superheavy elements



Michael Block
GSI Darmstadt
Helmholtzinstitut Mainz
Institut für Kernchemie der Universität Mainz

50
YEARS
GSI

10
JAHRE
HIM

Outline

- Brief introduction to superheavy elements
- challenges for laser spectroscopy in heavy and superheavy elements
- atomic and nuclear properties revealed by laser spectroscopy
- recent results obtained at GSI Darmstadt and JGU Mainz
- summary and outlook

Superheavy Element Research – Key Questions

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

S.A. Giuliani et al., Rev. Mod. Phys. 91, 011001 (2019)

SHIP Laser Spectroscopy Collaboration



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KU LEUVEN



UNIVERSITY OF
LIVERPOOL



TRIUMF

GANIL
laboratoire commun CEA/DRF

cea

HELMHOLTZ
Helmholtz-Institut Mainz

GSI

JGU
JOHANNES GUTENBERG
UNIVERSITÄT MAINZ

M. Block

The Es/Fm Collaboration

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.



Groups of

- T. Albrecht-Schmitt, FSU
- M. Block, Mainz, GSI
- Ch. Duellmann, Mainz
- K. Wendt, Mainz

Activities led by

S. Raeder, GSI



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Laser Spectroscopy - Current Status

Venturing towards uncharted territory...

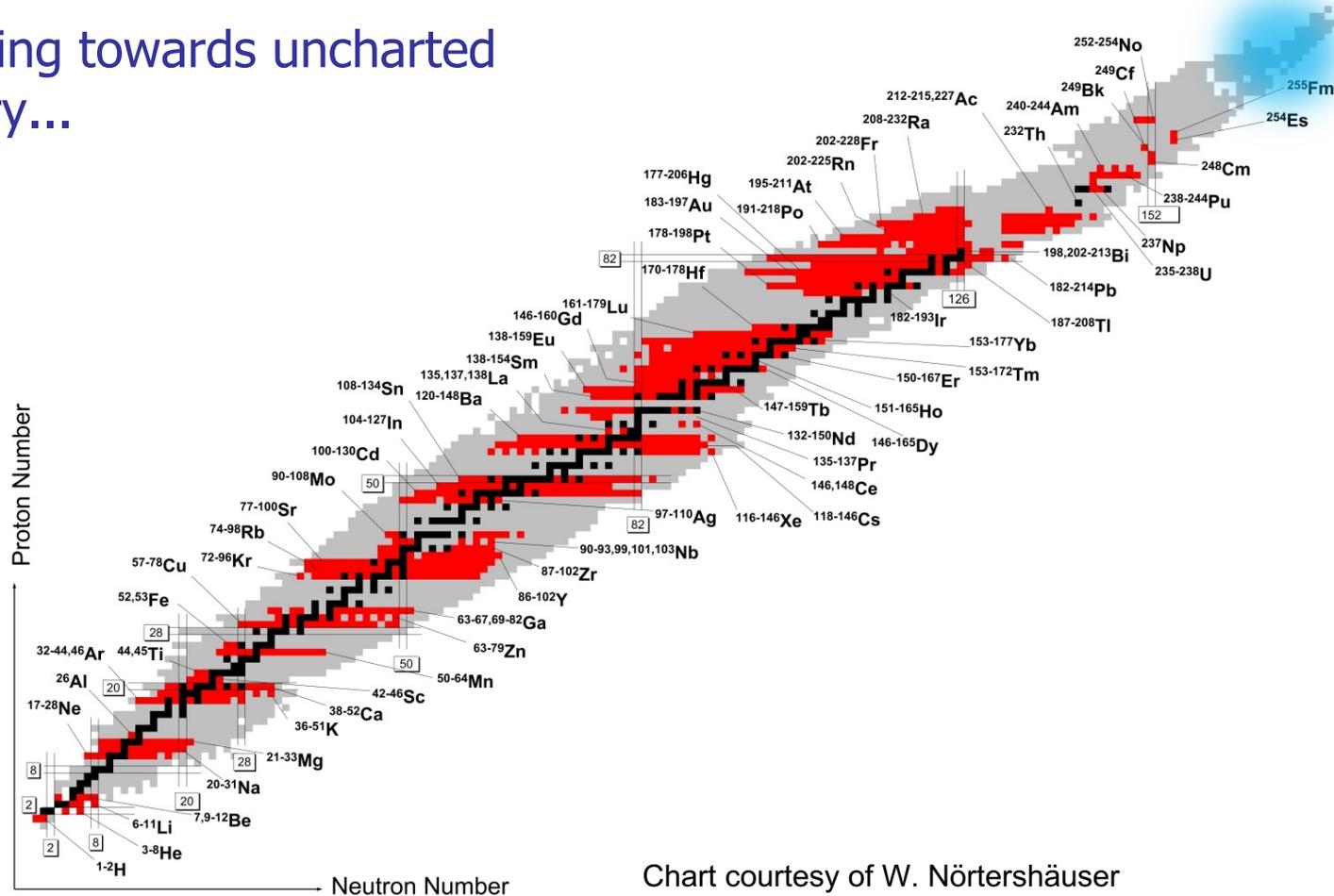


Chart courtesy of W. Nörtershäuser

Present Knowledge of Ionization Potentials



United Nations
Educational, Scientific and
Cultural Organization



International Year
of the Periodic Table
of Chemical Elements

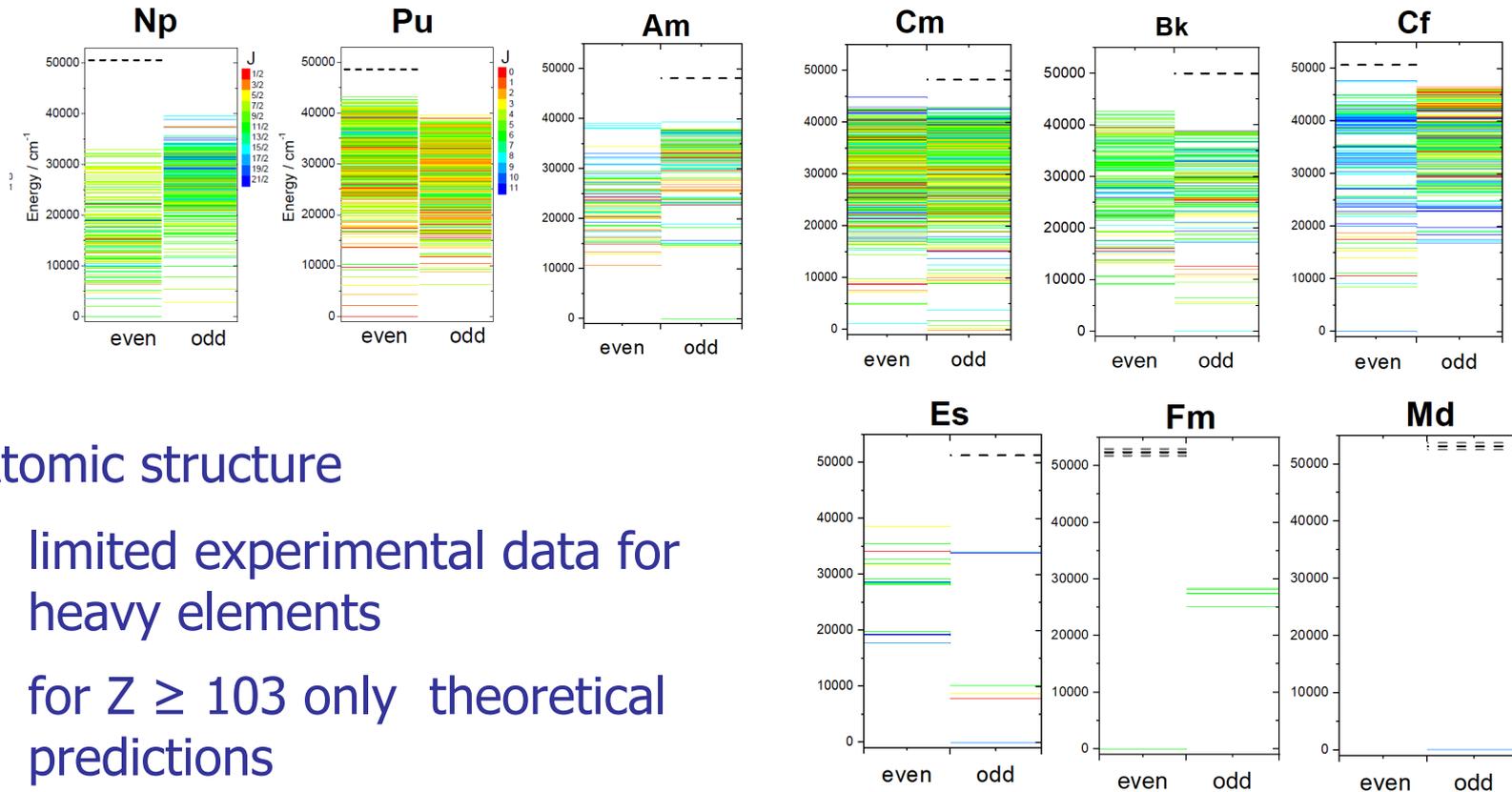
		<table border="0"> <tr> <td style="background-color: #006400;">Uncertainty < 0.1 μeV</td> </tr> <tr> <td style="background-color: #00FF00;">Uncertainty 0.1 - 1.0 μeV</td> </tr> <tr> <td style="background-color: #FFFF00;">Uncertainty 1 - 10 μeV</td> </tr> <tr> <td style="background-color: #FFA500;">Uncertainty 10 - 100 μeV</td> </tr> <tr> <td style="background-color: #FF0000;">Uncertainty 0.1 - 1 meV</td> </tr> <tr> <td style="background-color: #FFFFFF;">Uncertainty 10 - 200 meV</td> </tr> <tr> <td style="color: red;">No experimental value</td> </tr> </table>										Uncertainty < 0.1 μeV	Uncertainty 0.1 - 1.0 μeV	Uncertainty 1 - 10 μeV	Uncertainty 10 - 100 μeV	Uncertainty 0.1 - 1 meV	Uncertainty 10 - 200 meV	No experimental value										
Uncertainty < 0.1 μeV																												
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Uncertainty 10 - 200 meV																												
No experimental value																												
1 H	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	57 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 Ac	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 Ts	118 Og											

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

Courtesy U. Koester, D. Studer

Experimental Data atomarer Niveaus in Actiniden

Overview of known atomic levels in the heavy actinides



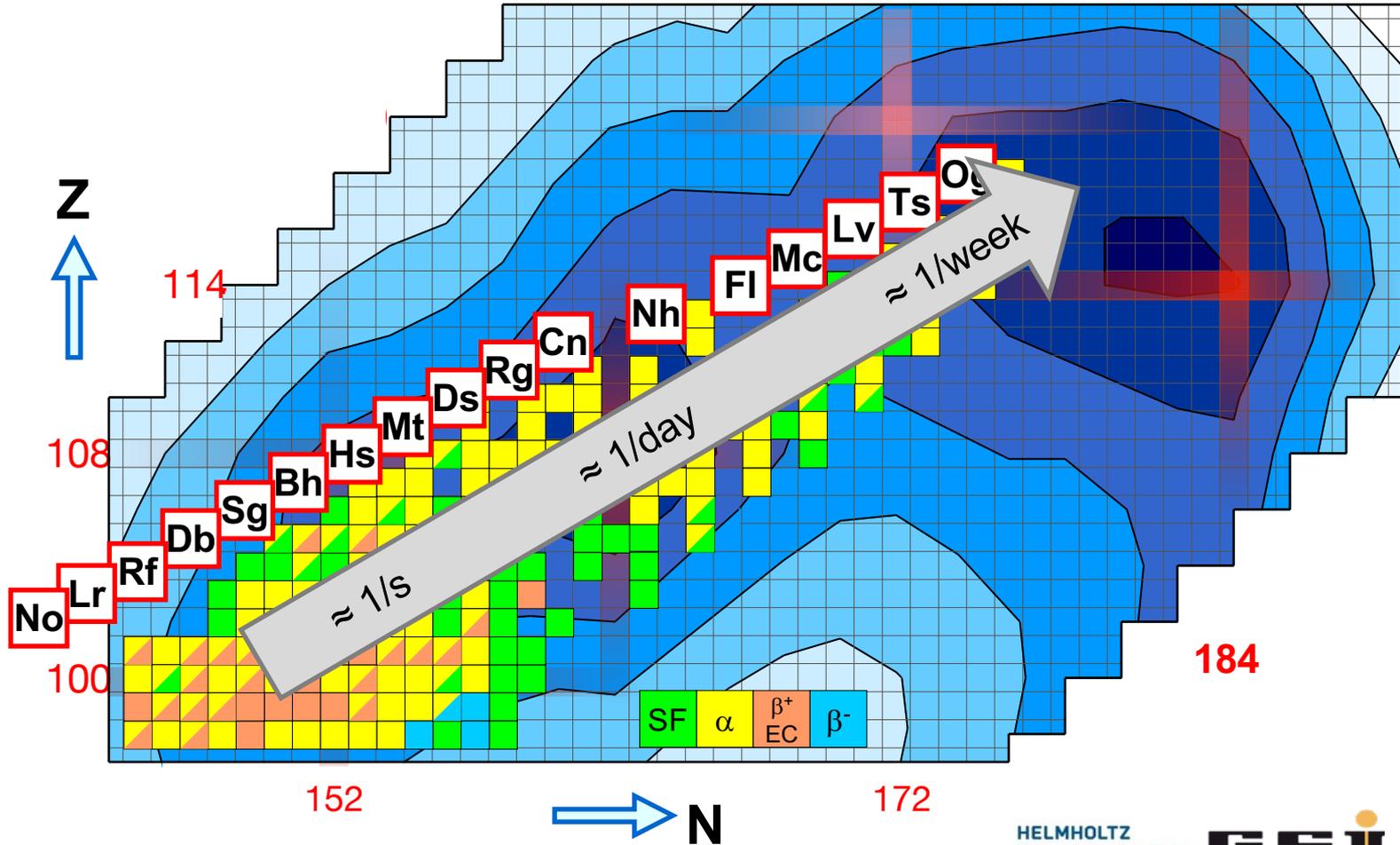
Atomic structure

- limited experimental data for heavy elements
- for $Z \geq 103$ only theoretical predictions

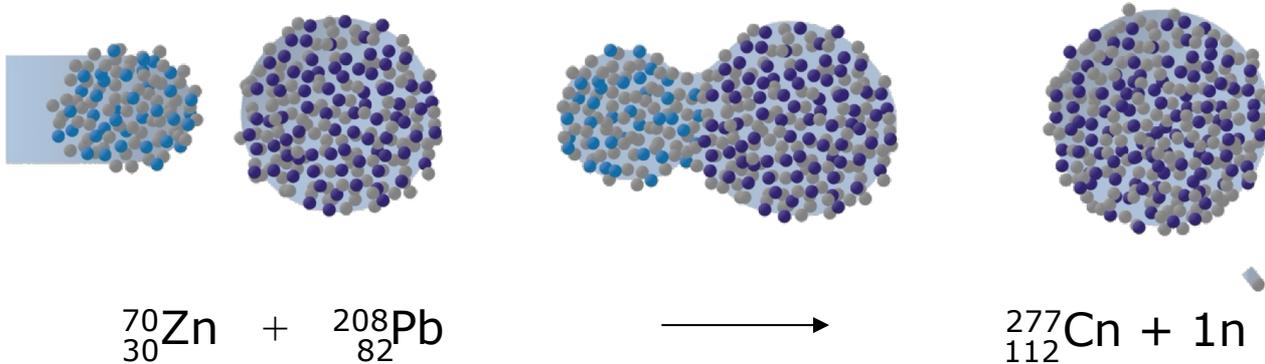
Challenges for Laser Spectroscopy of Heaviest Nuclides

- very low production rates / low amount of atoms
 - production at high energy (tens of MeV) for laser spectroscopy
 - no stable or long-lived isotopes for $Z > 100$ available
- **Sensitive and efficient methods required**
- atomic theory crucial to guide and interpret experiments
 - high spectral resolution important to obtain nuclear properties

Superheavy Element Research – Yield



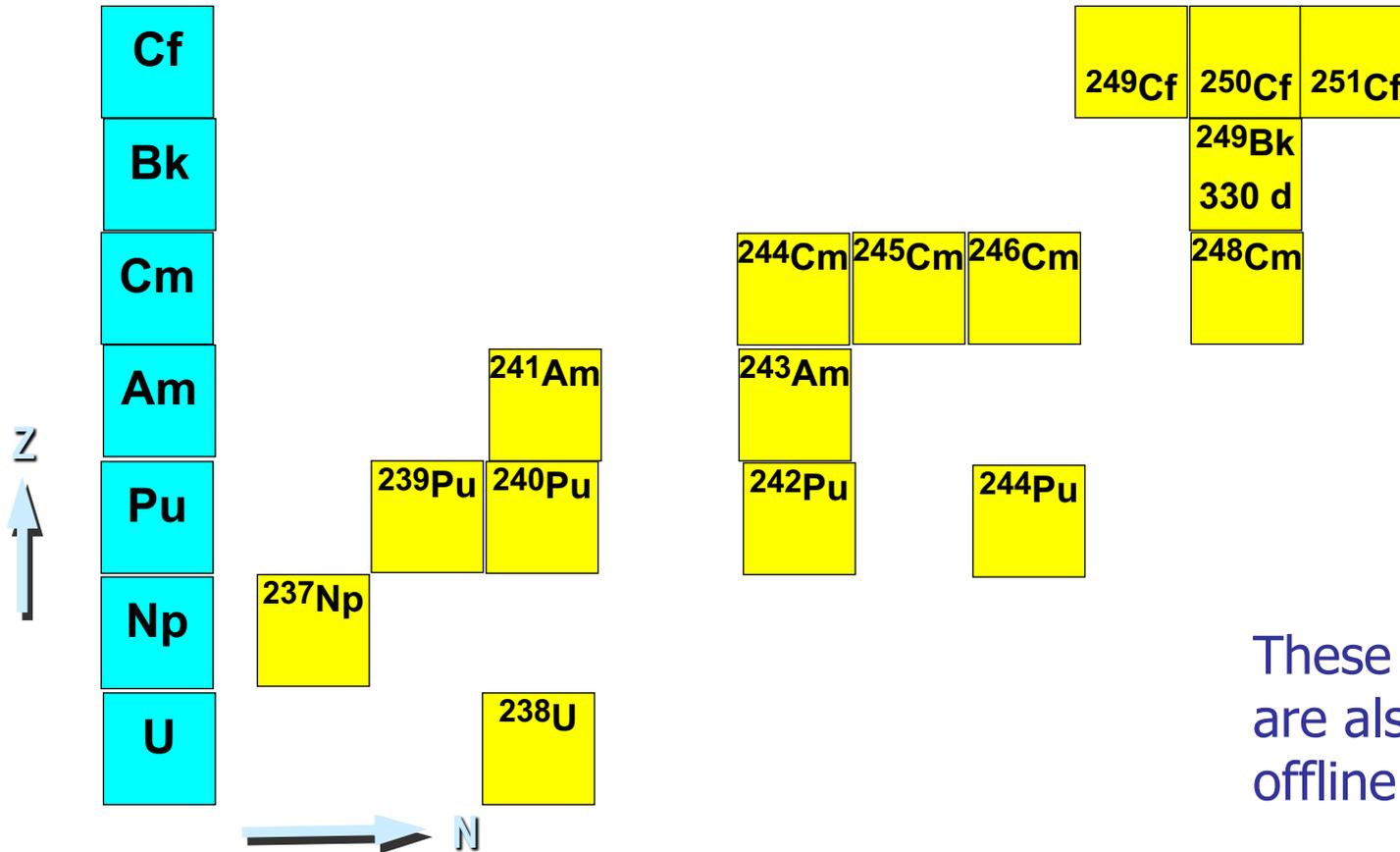
Produktion in Schwerionen-Fusionsreaktionen



Primary beam intensity:

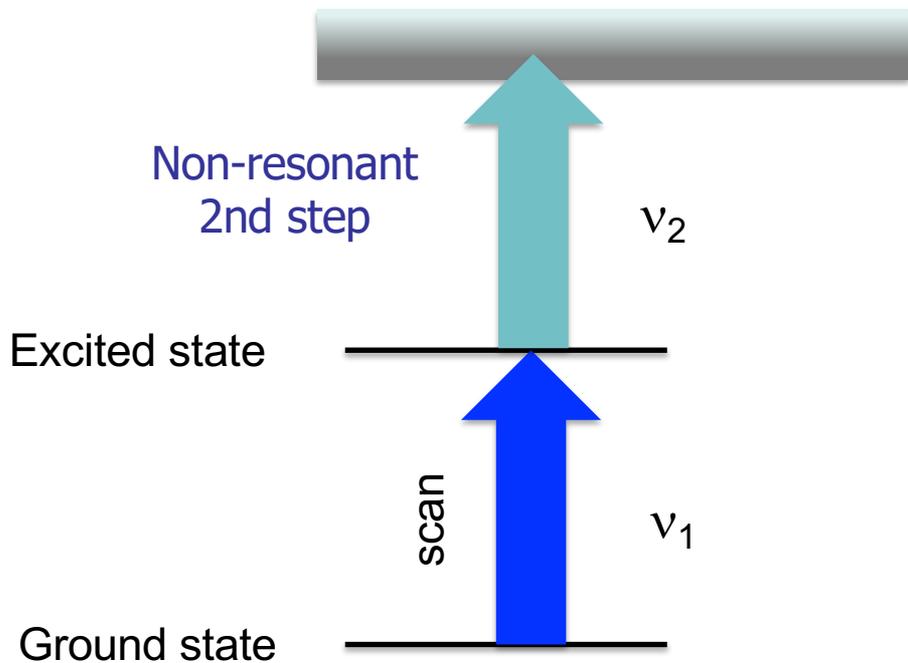
- about 6×10^{12} per second for typical beams ${}^{48}\text{Ca}$, ${}^{50}\text{Ti}$, ..., ${}^{70}\text{Zn}$
- Beam energy 4.5-6.5 MeV/u

Available Actinide Isotopes for Target Production



These long-lived nuclides are also interesting for offline measurements

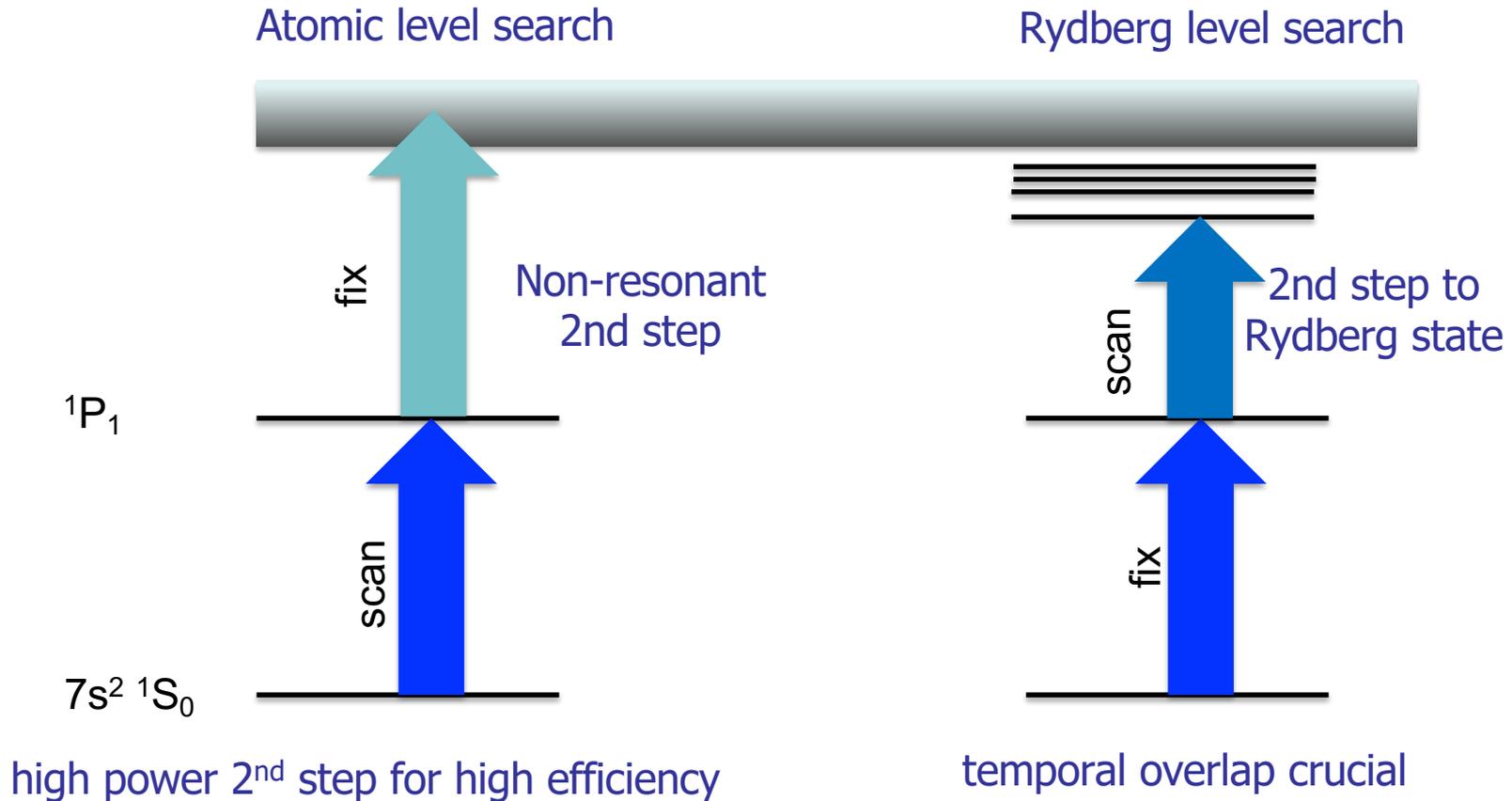
Resonant Laser Ionization – Excitation Schemes



$$h (\nu_1 + \nu_2) > E_{IP}$$

- ion detection more efficient than fluorescence detection
- detection of radioactive decay increases sensitivity
- 2-step ionization for initial level search if no data known

Resonant Laser Ionization – Excitation Schemes

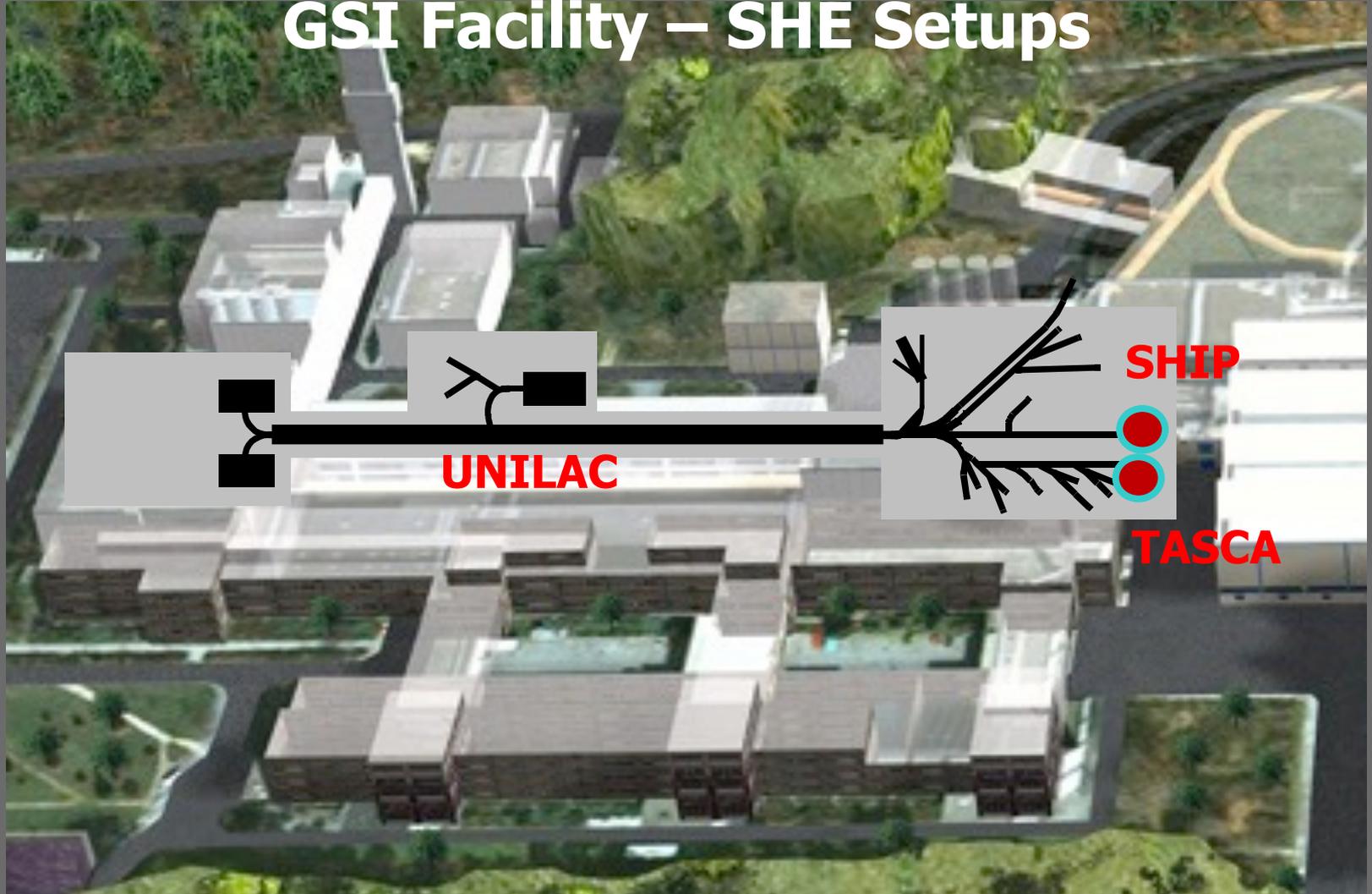


Online experiments on radionuclides produced in fusion evaporation reactions

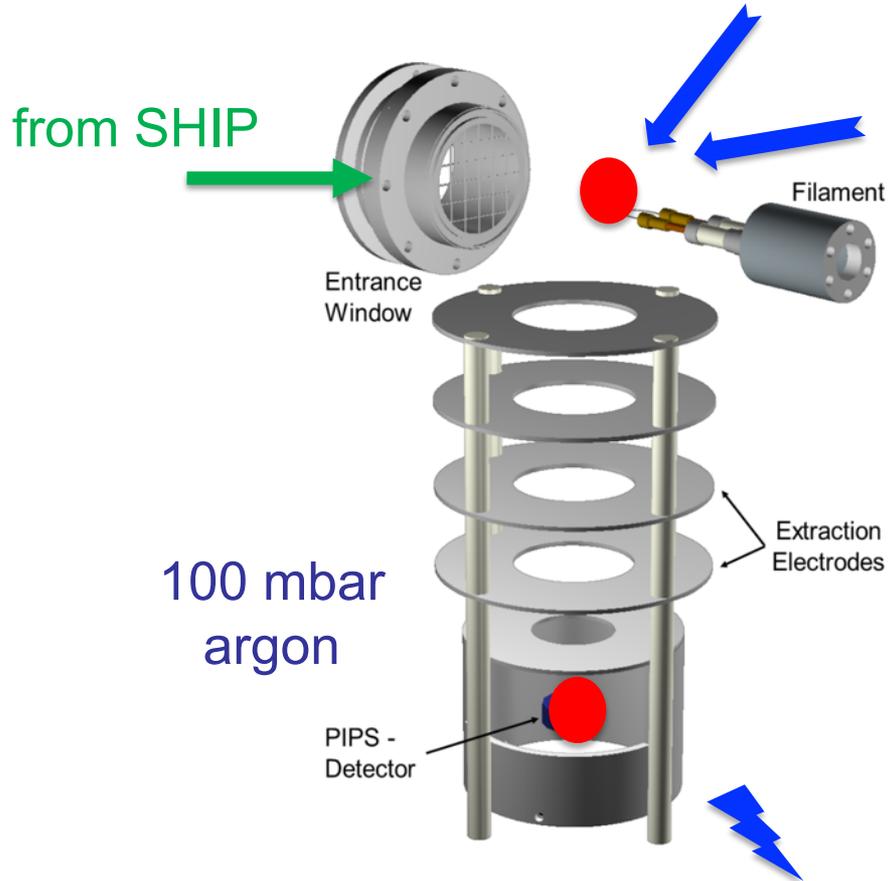
GSI / FAIR Facility



GSI Facility – SHE Setups



Radiation Detected Resonance Ionization Spectroscopy Method

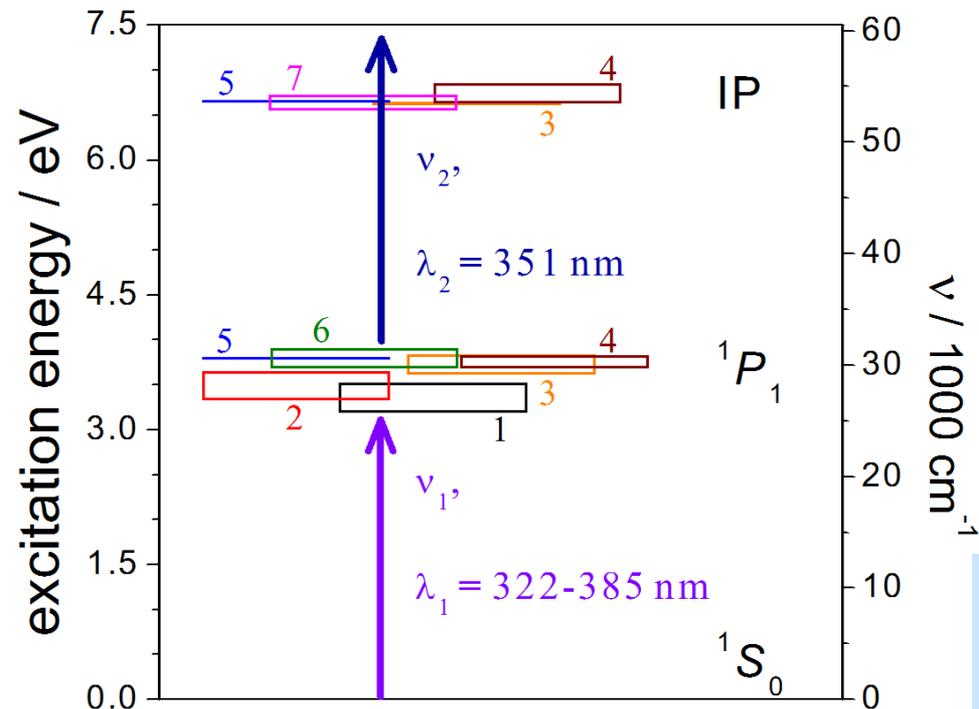


- separate from primary beam
- slow down and neutralize
- evaporate atoms
- two-step photo-ionization
- transport to detector
- register radioactive decay

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

M. Block

Predictions for Atomic Transitions in Nobelium



1 (MCDF): S.Fritzsche, Eur. Phys. J. D 33 (2005) 15

2 (MCDF): S.Fritzsche, Eur. Phys. J. D 33 (2005) 15

3 (IHFSCC): A.Borschevsky et al., Phys. Rev. A 75 (2007) 042514

4 (RCC): V.A.Dzuba et al., Phys. Rev. A 90 (2014) 012504

5 (MCDF): Y.Liu et al., Phys. Rev. A 76 (2007) 062503

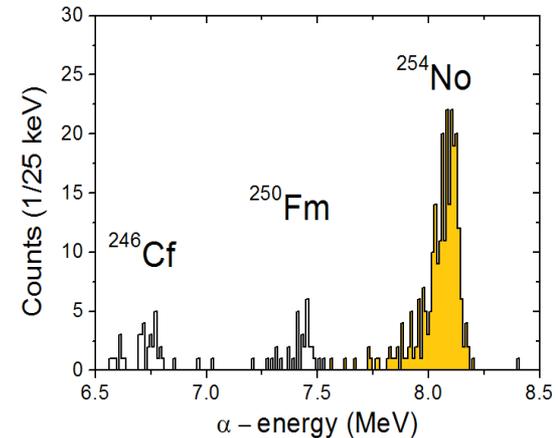
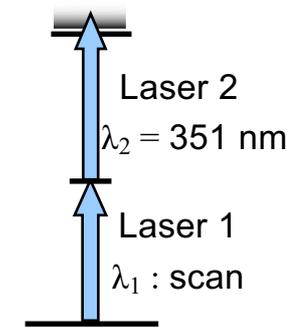
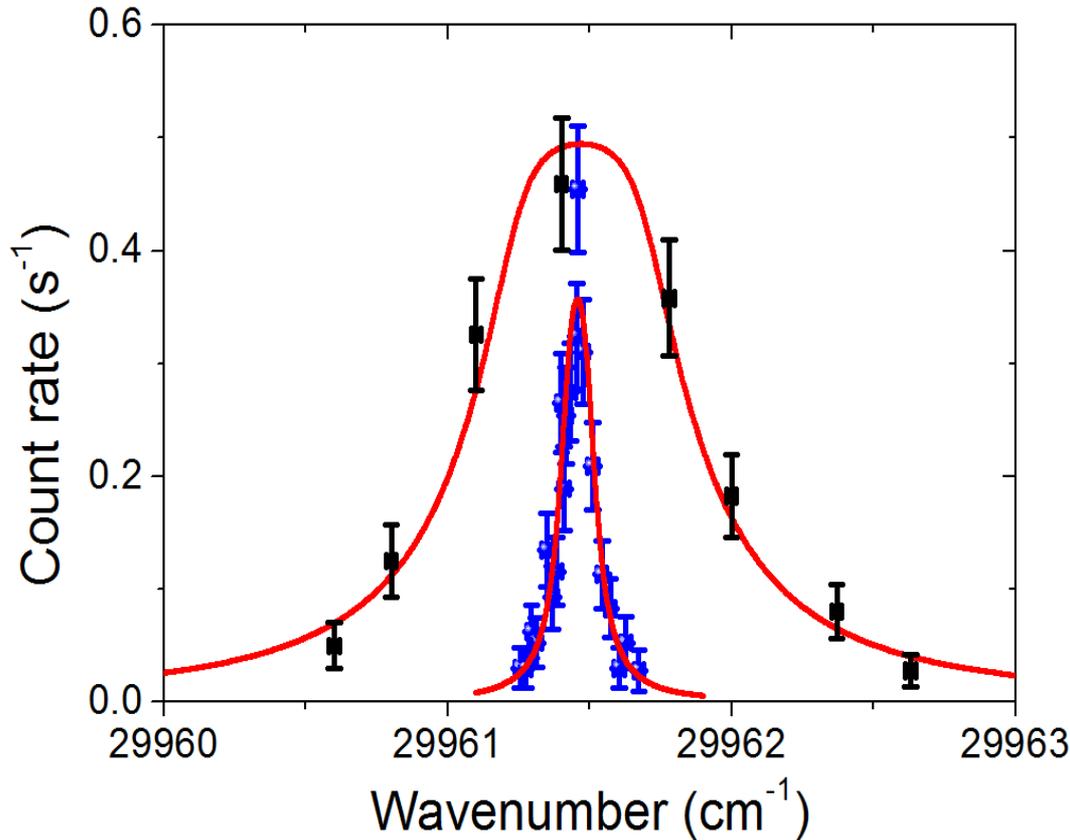
6 (MCDF): P.Indelicato et al., Eur. Phys. J. D 45 (2007) 155

7 (extrapolation): J.Sugar, J. Chem. Phys. 60 (1974) 4103

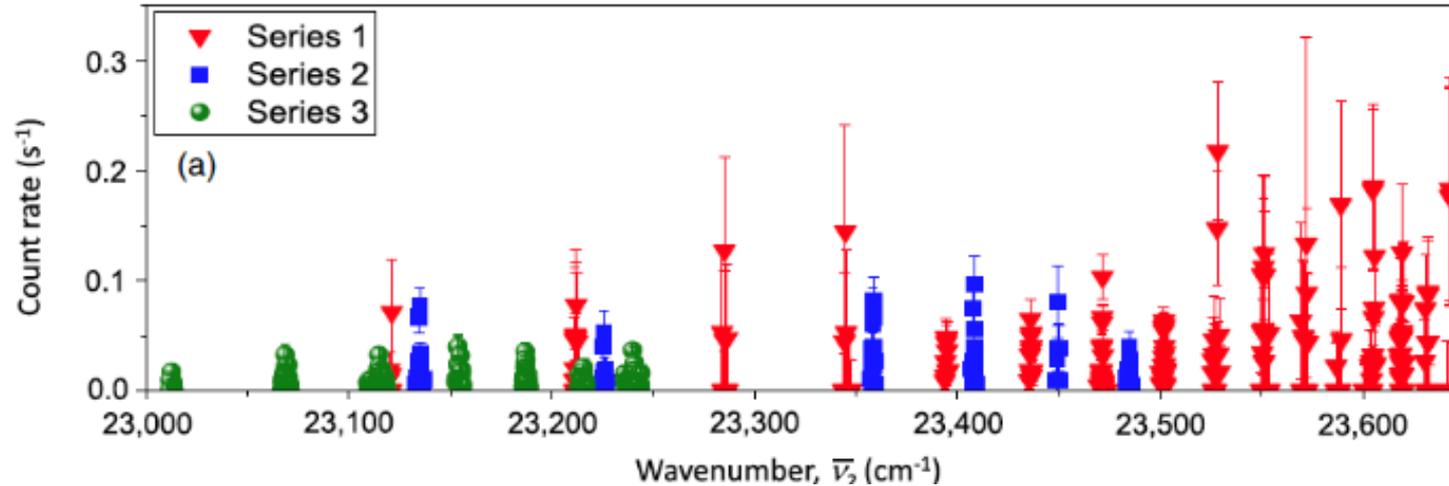
theoretical uncertainty corresponds to a scan range of about 5,000 steps - one step takes about 5 minutes

Z=102 atomic ground state: $[\text{Rn}]5f^{14}7s^2 1S_0$

Breakthrough in 2015 – First Resonance



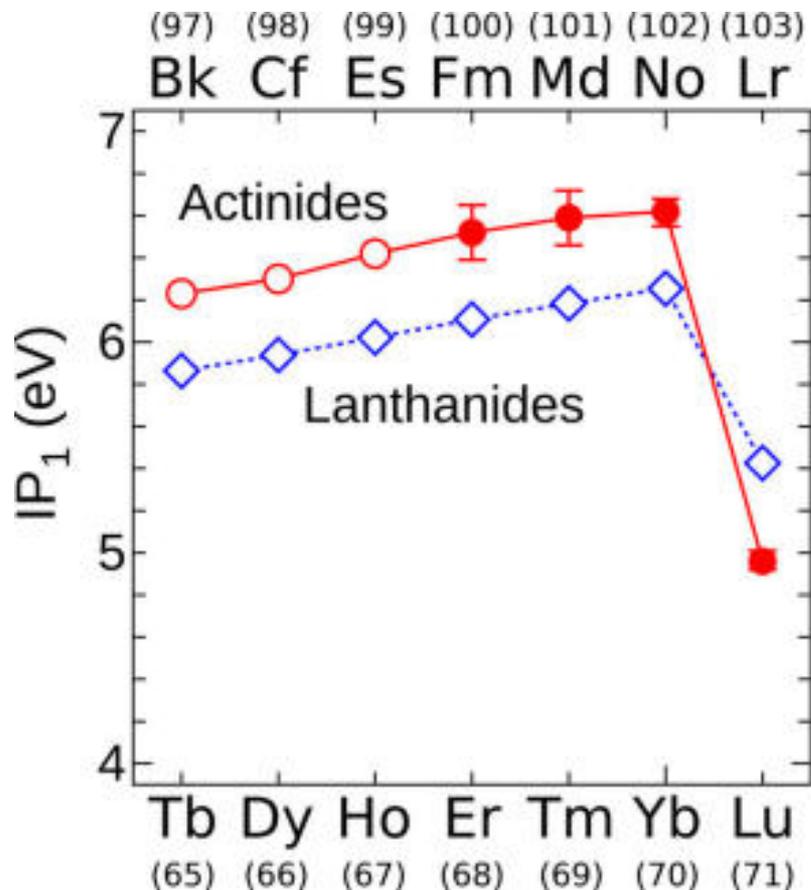
Nobelium Ionization Potential from Rydberg Series



Method	IP (cm ⁻¹)	³ D ₃ (cm ⁻¹)
Experiment (this work)	53 444.0 ± 0.4	29 652 ⁺⁸ ₋₁
IHFSCC [4]	53 489 ± 800	29 897 ± 800
CI+ all orders [5]	54 390 ± 1100	30 183 ± 1100
MCDF [6]	53 701 ± 1100	
Extrapolation [30]	53 600 ± 600	

- 35 atomic states observed
- good agreement with theory predictions

Ionization Potential of Actinides and Transactinides



	IP (eV)
Laser spectroscopy At SHIP / GSI	6.62621(5)
Theory (Borschevsky et al., RCC)	6.632
Extrapolation (Sugar)	6.65(7)

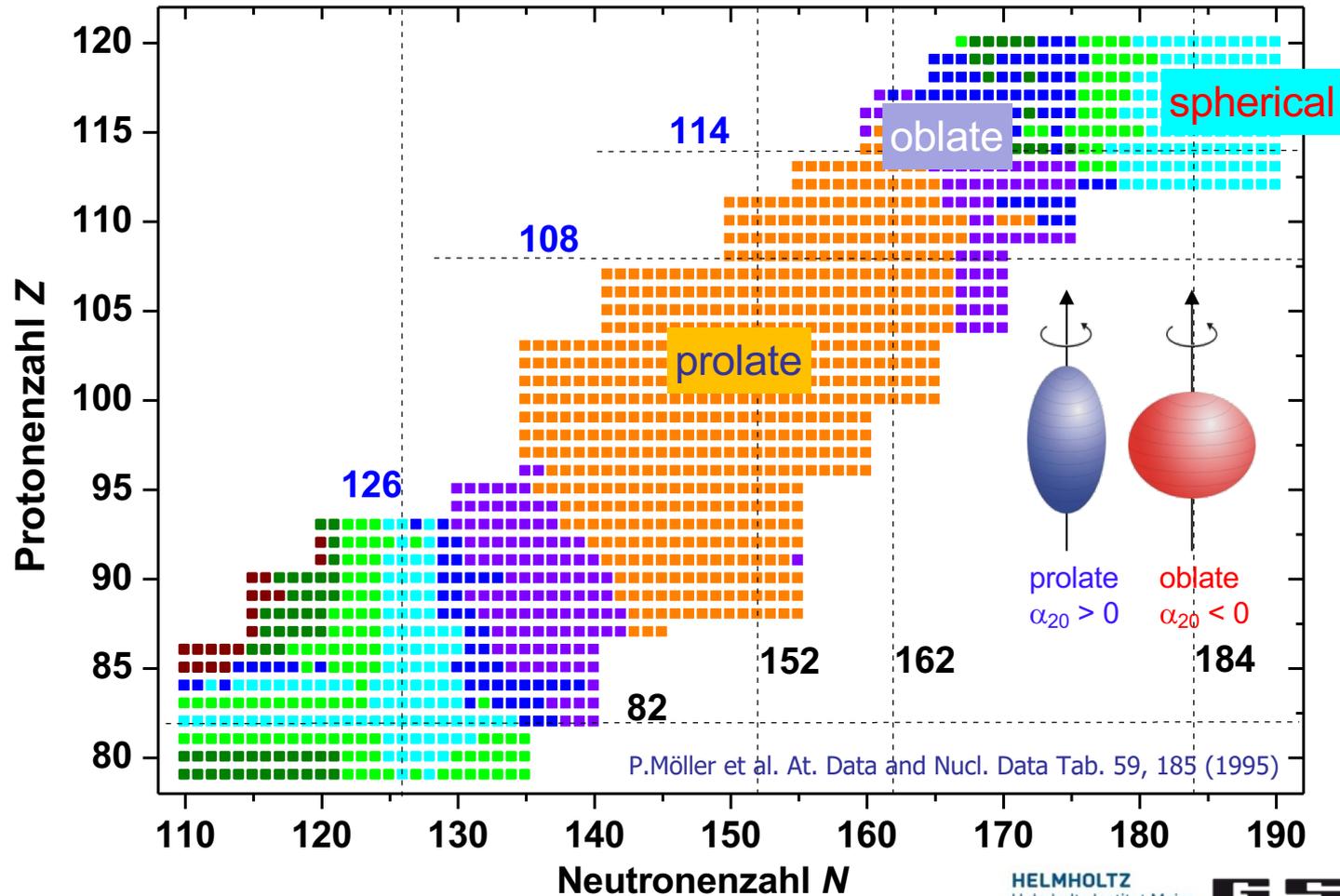
T. K. Sato *et al.*
Nature **520** (Apr.9) (2015) 209-211.

T. Sato et al. JACS 2018, 140, 14609

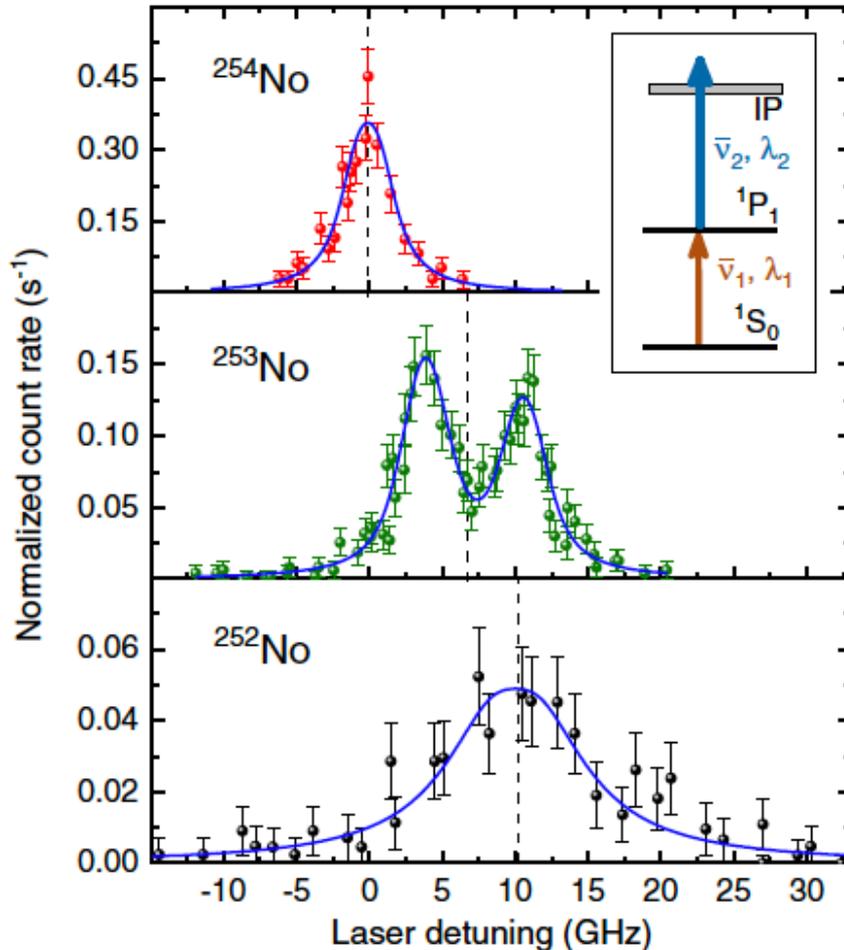
No: $IP_1(\text{No}) = 6.63 \pm 0.08 \text{ eV}$

Lr: $IP_1(\text{Lr}) = 4.96 \pm 0.08 \text{ eV}$
 $\rightarrow \text{Lr: } [\text{Rn}]5f^{14}7s^27p_{1/2}$

Deformation in the Region of Superheavy Nuclides



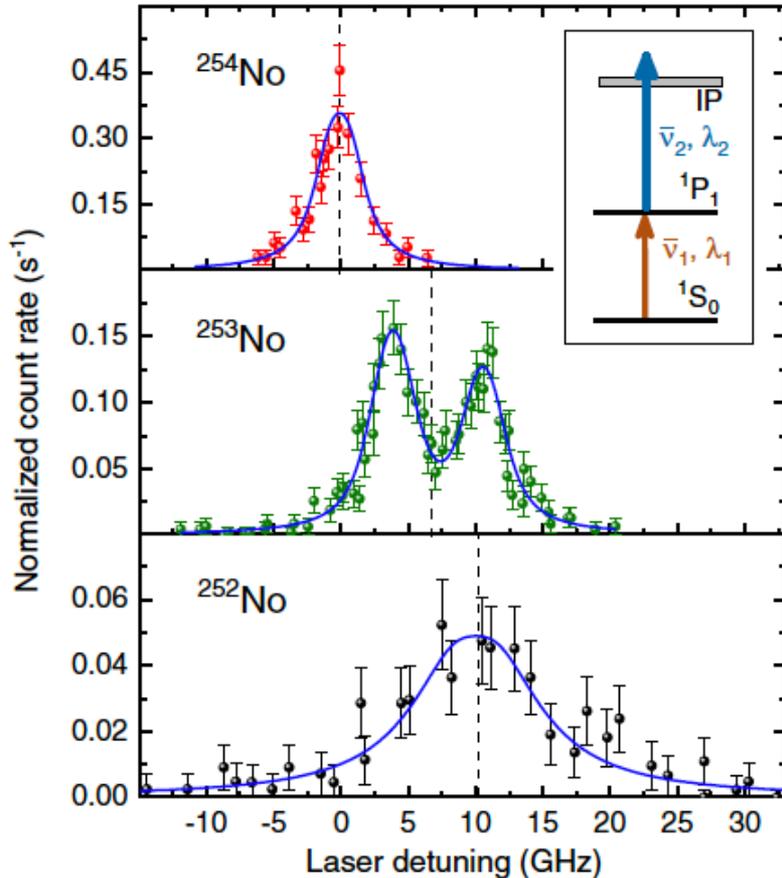
Laser Spectroscopy of Nobelium Atoms



- First optical spectroscopy beyond $Z=100$ despite low yields on the atom-at-a-time scale
- Half-life range 2.4 s – 55 s
- Several atomic and nuclear properties determined

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

Hyperfine Structure in ^{253}No



experimental results

$A = 734(46)$ MHz; $B = 2815(686)$ MHz

and input from atomic theory provide
magnetic moment and quadrupole moment

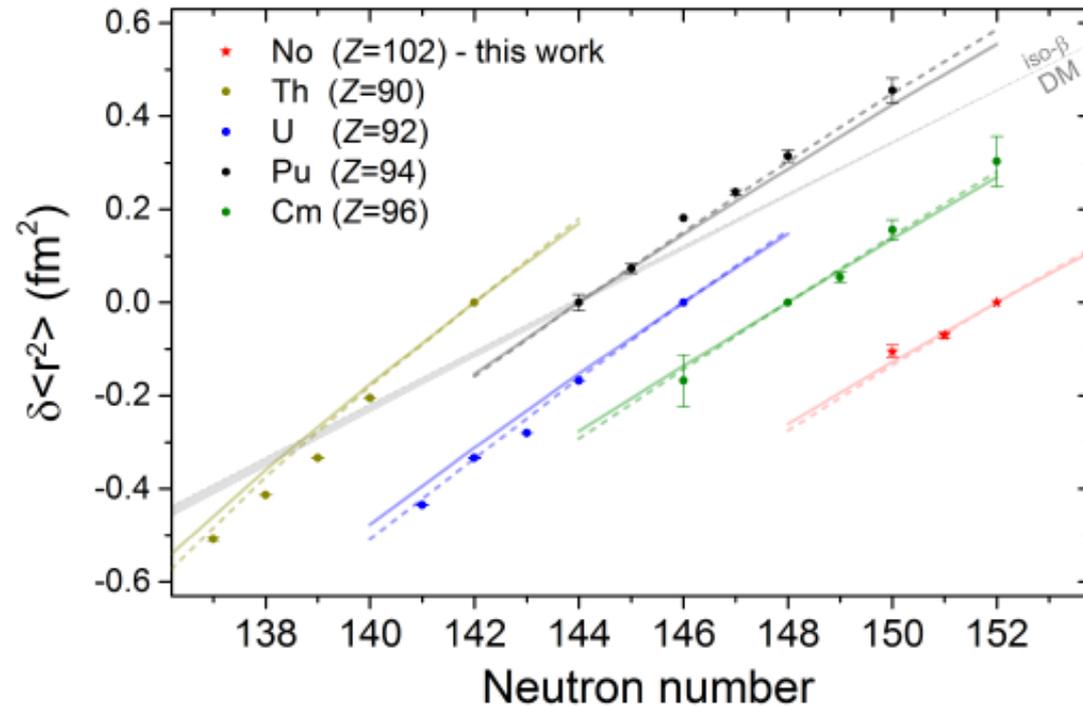
	Ref.	μ (μ_N)	Q_s (b)
Laser spec. this work	[1]	-0.527 ± 0.034	5.79 ± 1.42
	[2]	-0.549 ± 0.034	
Nucl. spec.	[3]	-0.593	7.145

[1] V.A. Dzuba et al. (RCC)

[2] A. Borschevsky et al. (RCC)

[3] R.D. Herzberg et al., *Eur. Phys. J. A* **42**, 333-337 (2009)

Charge Radii in Actinides



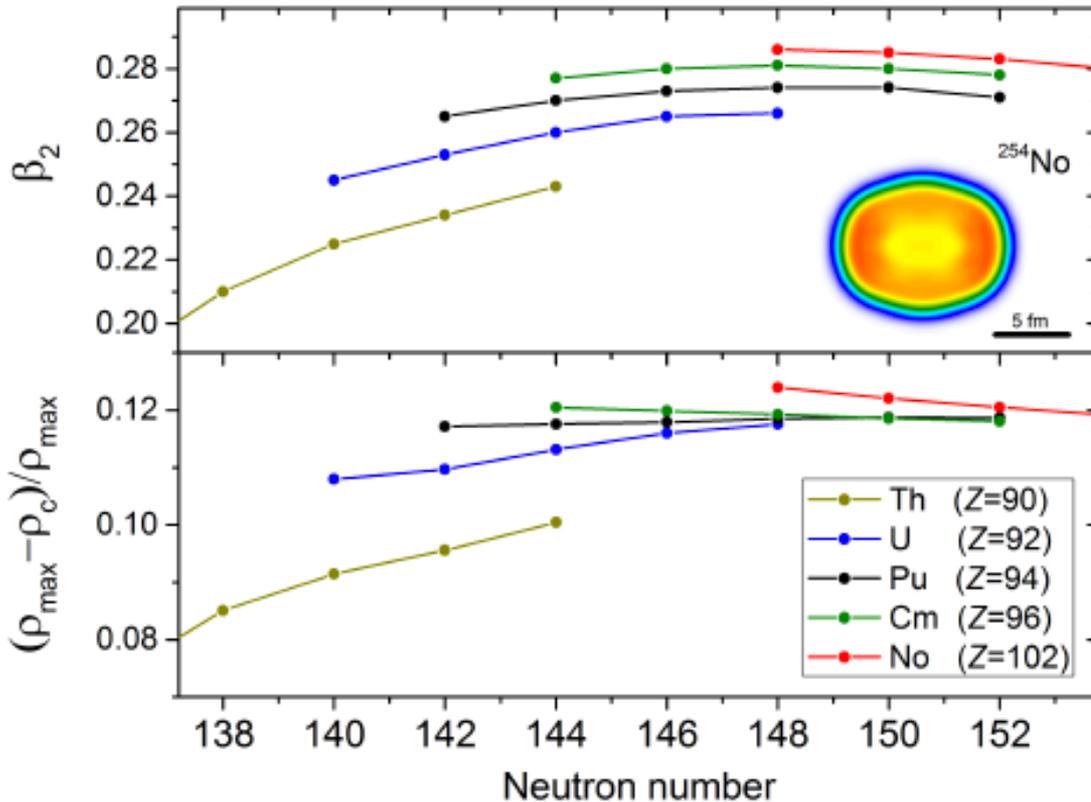
good agreement between
experimental data for actinides
from laser spectroscopy and
theoretical calculations
using density functional theory

J. Maruhn et al. Phys. Commun. 185 2195 (2014)

P.-G. Reinhard, W. Nazarewicz, Phys. Rev. C 95 064328 (2017)

Exp. Date for No: S. Raeder et al., Phys. Rev. Lett. 120 (2018) 232503

Deformation in Nobelium



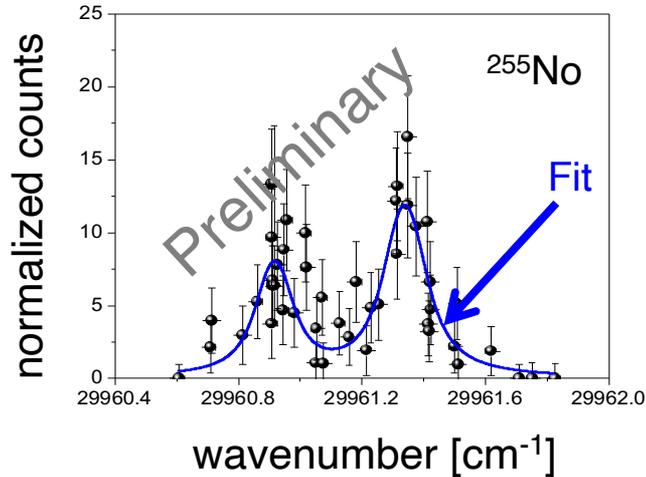
- Theoretical calculations using density functional theory predict:
- maximum deformation around $N = 152$
- central depression in proton density already for nobelium

Calculations B. Schuetrumpf, W. Nazarewicz et al.

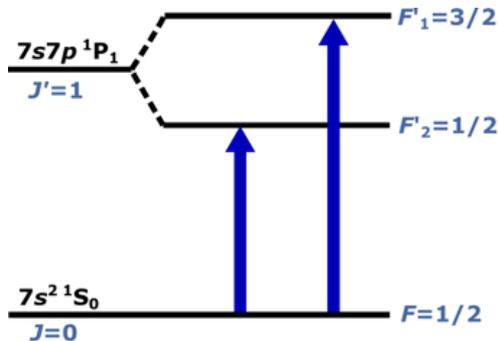
Extending laser spectroscopy at SHIP

Production $^{209}\text{Bi}(^{48}\text{Ca}, 2n)^{255}\text{Lr} \xrightarrow{\text{EC (15\%)}} ^{255}\text{No}$

2019 beamtime



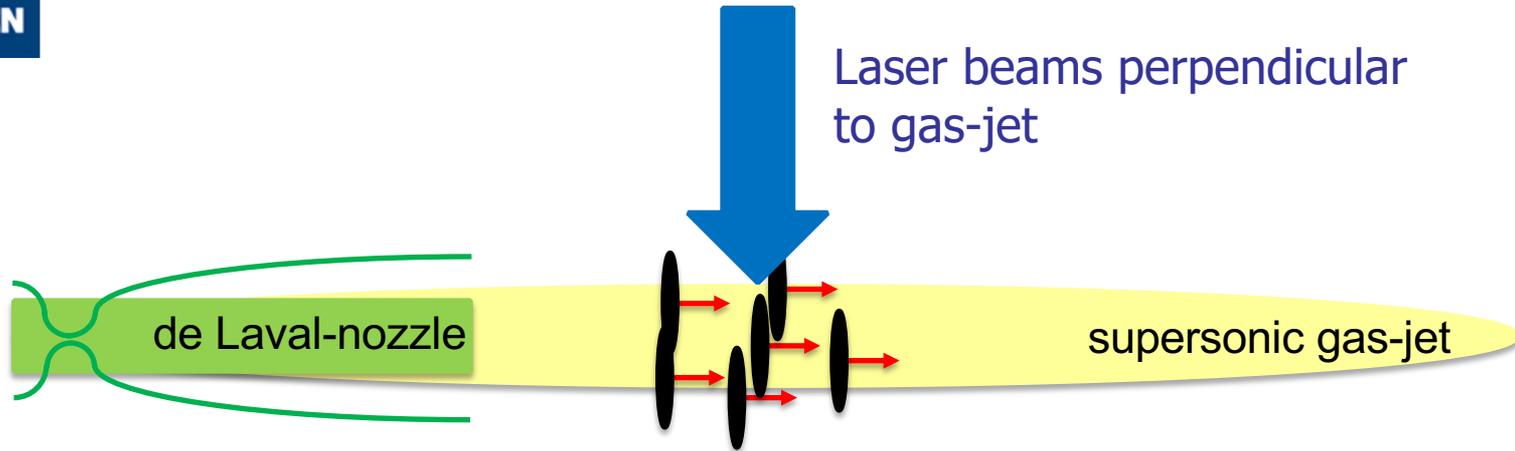
- yield: about 0.05 atoms / second
- preliminary results confirm nuclear spin $I=1/2$
- ongoing analysis will provide magnetic moment



- preparatory experiments for Lr ($Z=103$) completed – ready for level search in 2020

Improving Spectral Resolution: In-Gas-Jet Spectroscopy

KU LEUVEN

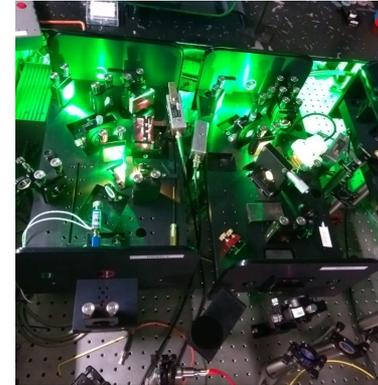
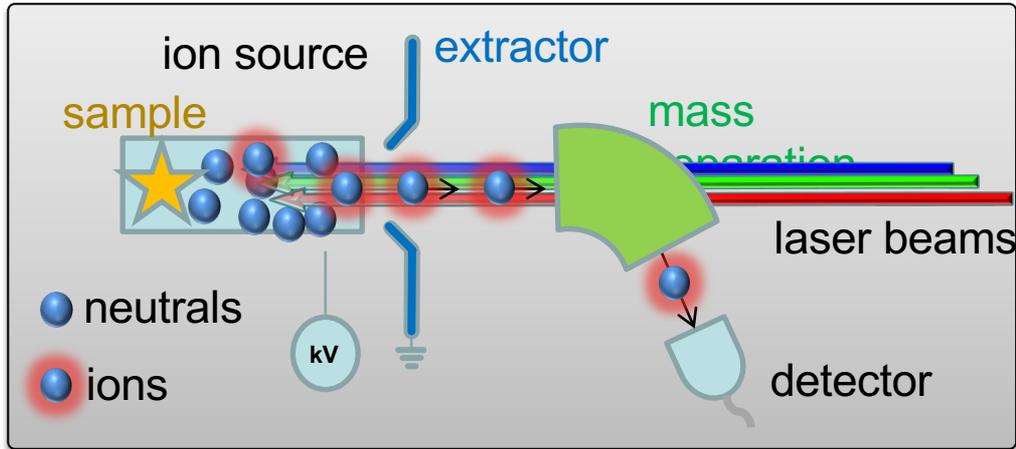


- directed movement of atoms in gas-jet, perpendicular ionization
 - high Mach-number for low pressure and low temperature
- reduction of Doppler-broadening for higher spectral resolution

For more details see contribution by R. Ferrer

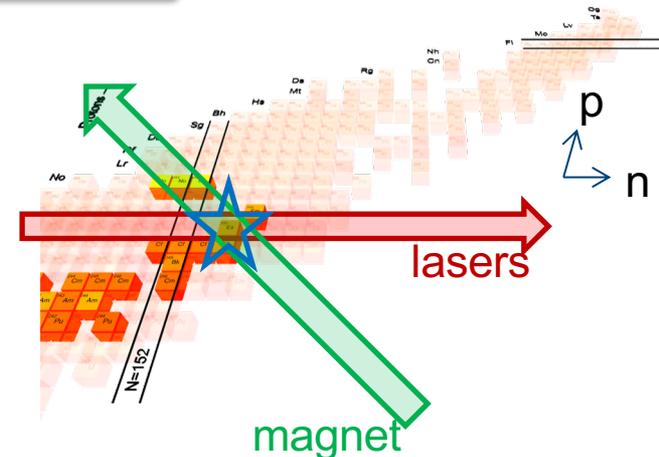
Offline Experiments on Long-Lived Actinide Isotopes At the RISIKO Separator at JGU Mainz

Hot Cavity Resonance Ionization Spectroscopy

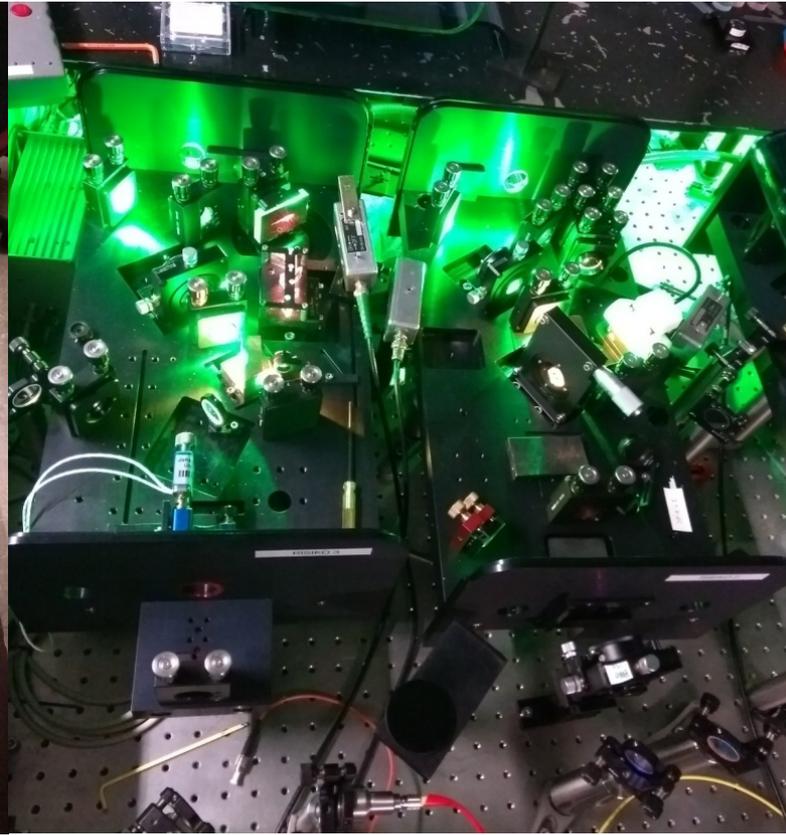
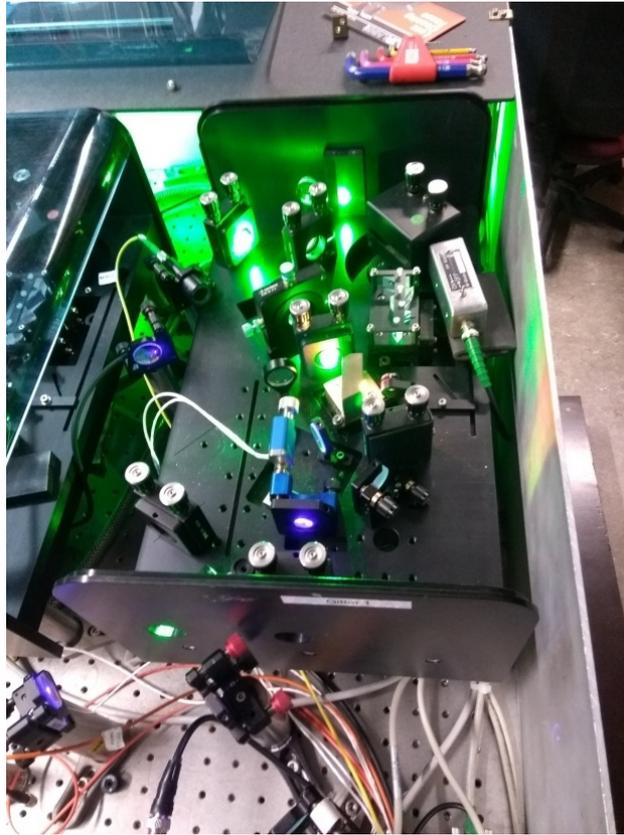


10 kHz Laser system

- laser spectroscopy with high efficiency
- used for production of radioactive ion beams
- background from surface ionization
- Resolution limited by source temperature and laser bandwidth



Laser Systems and the Sample



Courtesy of S. Raeder



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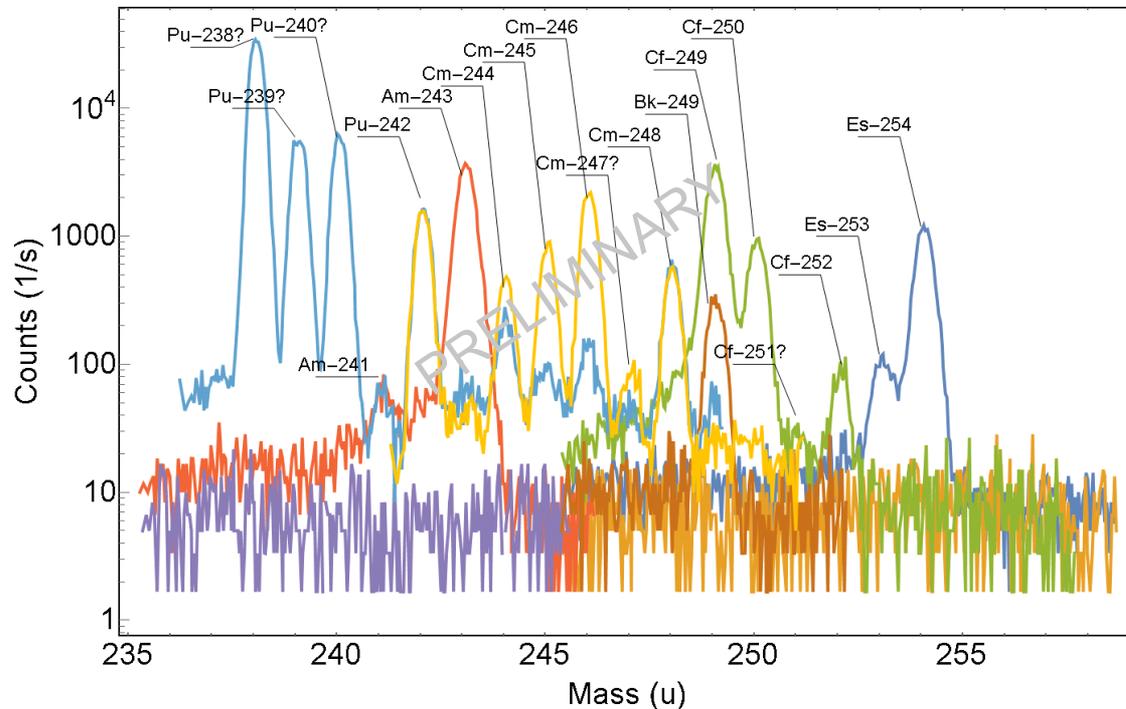


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Sample Analysis with Laser Ionization



- Radiochemical separation of actinide samples challenging
- Lasers tuned to resonantly ionize different actinide elements
- Multiple nuclides in one sample

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.

F. Weber, K. Wendt et al.

Courtesy of S. Raeder



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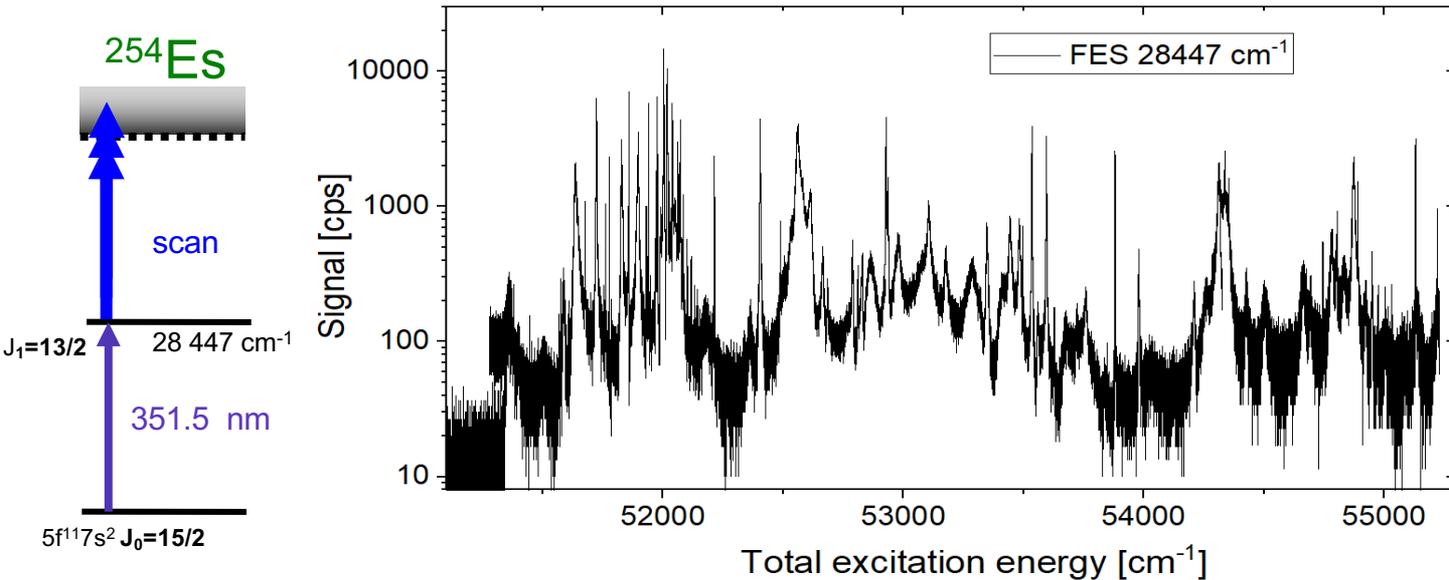
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Laser Spectroscopy on Es

- Es sample from Oak Ridge / Florida State University
- Mainz: RISIKO separator filled with one aliquot



-10^{10} atoms ^{254}Es
($T_{1/2} = 276$ d)

-10^9 atoms ^{253}Es
($T_{1/2} = 20.5$ d)

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.

Courtesy of S. Raeder

Results for Fermium

- Investigated 3 atomic transitions in ^{257}Fm ($T_{1/2} = 30$ d)
- out of 7 transitions reported in ^{255}Fm by Sewtz et al. 2003 / Backe et al. 2006

No.	WN (cm-1)	FWHM (cm-1)	A_ki (1/s)	WN fund. (cm-1)	WL fund. (nm)	WN to IP (cm-1)	WN to IP fund. (cm-1)	WL to IP fund. (nm)
R1	25,099.80 pm 0.2	-	3.4 pm 0.8 E6	12,549.90	796.8	27,300.2	13650.1	732.6
R2	25,111.80 pm 0.2	-	3.5 pm 0.7 E6	12,555.90	796.4	27,288.2	13644.1	732.9
R3	27,389 pm 1.5	0.85 pm 0.16	-	13,694.5	730.2	25,011	12505.5	799.6
R4	27,466 pm 1.5	1.34 pm 0.09	≥ 2.9 E6	13,733	728.2	24,934	12467	802.1
R5	28,185 pm 1.5	1.08 pm 0.05	-	14,092.5	709.6	24,215	12107.5	825.9
R6	28,377 pm 1.5	0.75 pm 0.05	-	14,188.5	704.8	24,023	12011.5	832.5
R7	28,391 pm 1.5	0.61 pm 0.03	≥ 1.1 E7	14,195.5	704.4	24,009	12004.5	833.0

Sewtz, M., et al. "First observation of atomic levels for the element fermium ($Z= 100$)." *Phys. Rev. Lett.* 90.16 (2003): 163002.
 Backe, H., et al. "Laser spectroscopic investigation of the element fermium ($Z= 100$)." *Laser 2004.* (2006). 3-14.

Summary

- precision measurements of atomic properties by laser spectroscopy extended to heavier elements
- new opportunities to track the nuclear structure evolution in the heaviest nuclei providing information on shape, size and electromagnetic properties
- Online work complemented by “classical” laser spectroscopy on long-lived actinide isotopes
- all recent data in good agreement with theoretical calculations
- technical and methodical developments for extension to heavier elements under way

Thank you for your attention !