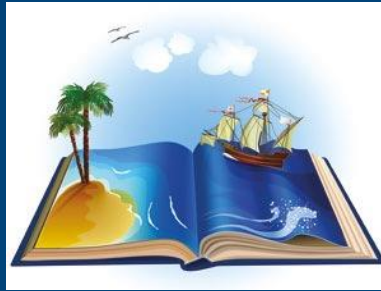


Ground-State Properties from Laser Spectroscopy

Part III: Examples from Recent Experiments



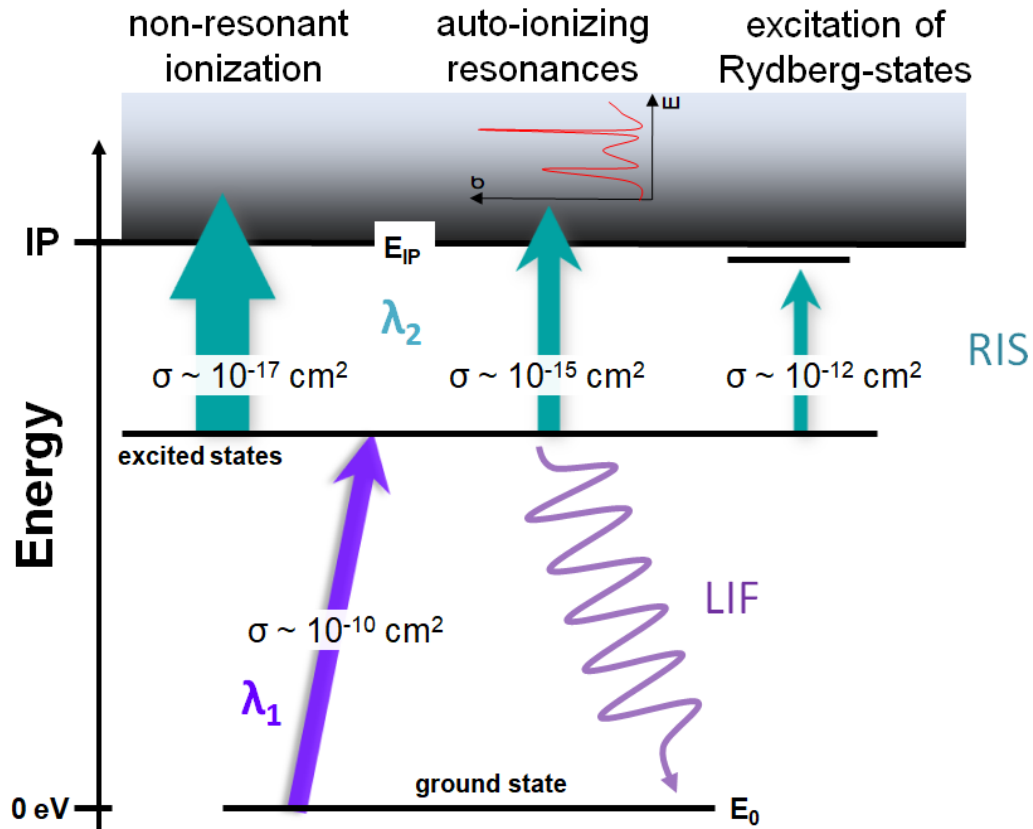
Michael Block

GSI Darmstadt, Germany
Helmholtz Institut Mainz (HIM), Germany
Department Chemie, Universität Mainz, Germany

Part II – Laser Spectroscopy for Nuclear Physics

- laser spectroscopy of fermium, nobelium, and lawrencium at GSI
- laser spectroscopy of actinides at RISIKO Mainz
- comparison of different techniques

Principles of Resonant Ionization Laser Spectroscopy



- 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

Ionization Probability vs. Excess Energy in RIS

Near threshold ($E \gtrsim IP$)

- Photoionization cross section follows Wigner threshold law:

$$\sigma \propto (E - IP)^{(l+1/2)} \quad (\approx \sqrt{E-IP} \text{ for s-wave electrons})$$

- Ionization probability initially rises slowly

Intermediate excess energy

- Cross section typically reaches a maximum
- $P = 1 - \exp(-\sigma F)$; saturation when $\sigma F \gg 1$.

Far above threshold

- Cross section generally decreases ($\sim E^{-3}$).
- More excess energy does not necessarily increase efficiency.

Availability of Superheavy Elements

1 H	2 He																			18 He
3 Li	4 Be																			2 Ne
11 Na	12 Mg																			10 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			18 Xe
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			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			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 Ts	118 Og			118 Og
11 9	12 0																			

Accelerated heavy-ion beam

1 atom min⁻¹

3 atoms day⁻¹

< 1 s

< 1 s

< 1 s

57 La	Targets on thin foils										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	

tons

mg / µg / pg

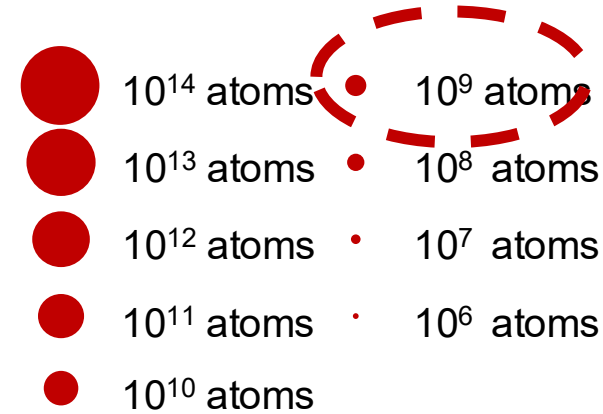
50 atoms min⁻¹

~1 h

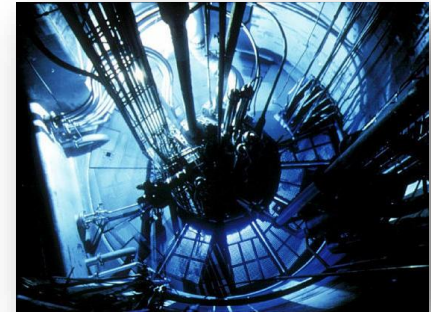
Production of Heavy Actinides for Off-line Studies

First analytics of the unseparated Fm sample 7 days after irradiation at ILL

Fm 248 34.5 s α 7.87, 7.83 sf	Fm 249 2.6 m ε 7.520 γ (~45...) e	Fm 250 1.8 s 30 m IT α 7.43 sf	Fm 251 5.30 h ε 881, 453... γ 96, 42, e ⁻ α 6.834, 6.783... sf	Fm 252 25.39 h α 7.039, 6.998... γ (96, 42), e ⁻ α 6.943, 6.673... γ 272, (145...) sf	Fm 253 3.0 d α 7.192, 7.150... γ (99, 43...), e ⁻ sf α ~76	Fm 254 3.240 h α 7.022, 6.963... γ 81, (58...), e ⁻ sf α 6.915, 6.870 α ~45	Fm 255 20.07 h α 6.520..., sf γ 241, 179..., e ⁻ α 2950 sf	Fm 256 157.1 m α 6.520..., sf γ 241, 179..., e ⁻ α 2950 sf	Fm 257 100.5 d α 6.520..., sf γ 241, 179..., e ⁻ α 2950 sf	Fm 258 370 μs α 6.520..., sf γ 241, 179..., e ⁻ α 2950 sf
Es 247 4.55 m e α 7.323, 7.275... α → g	Es 248 24 m α 6.879, 6.907... α → g βsf	Es 249 102.2 m α 6.776, 6.716 γ 380, 813, 375... α → g	Es 250 2.22 h 8.8 h γ 829, 303 1032, 349... e ⁻ α 6.492, 6.462... e ⁻	Es 251 33 h α 6.632, 6.562... γ (52...), e ⁻ , α → g sf γ 178, (153...), e ⁻ α 6.492, 6.462... e ⁻	Es 252 471.7 d α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8	Es 253 20.47 d α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8	Es 254 39.3 s 75.7 d α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8	Es 255 39.8 d α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8	Es 256 7.8 h 28.4 m α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8	Es 257 7.7 d α 6.633, 6.591... γ (42, 5...), e ⁻ sf α 178 + 5.8
Cf 246 35.7 h α 6.750, 6.708... γ (42, 96...), e ⁻ , g sf	Cf 247 3.11 h α 6.750, 6.708... γ (42, 96...), e ⁻ , g sf	Cf 248 333.5 d α 6.258, 6.217... γ (43, 99), e ⁻ , g sf	Cf 249 351 a α 5.81, 5.758... γ 388, 813, 375... α 497, α 1642 sf	Cf 250 13.08 a α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 251 898 a α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 252 2.647 a α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 253 17.81 d α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 254 60.5 d α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 255 85 m α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf	Cf 256 12.3 m α 5.81, 5.758... γ 388, 813, 375... α 2034, α 112 sf
Bk 245 4.95 d sf γ 253, 381... e ⁻ , g α 5.886 6.147... α → g	Bk 246 1.80 d ? e γ 799 1081, 834 1124... e ⁻	Bk 247 1380 a α 5.531, 5.710 5.688... γ 84, 265..., e ⁻ g, sf?	Bk 248 23.7 h 9 a β ⁻ 0.9... α 5.514, 5.386... γ (327, 308...), sf α 746, α 2.5	Bk 249 327.2 d β ⁻ 0.1 α 5.514, 5.386... γ (327, 308...), sf α 746, α 2.5	Bk 250 3.217 h β ⁻ 0.7, 1.8... γ 989, 1032 1029... α ~350, α 960 e ⁻	Bk 251 55.6 m β ⁻ ~0.9, 1.1... γ 178, 130, 153... e ⁻	Bk 252 ? >10 m β ⁻ ?	Bk 253 ? >10 m β ⁻ ?	Bk 254 ? >10 m β ⁻ ?	Bk 255 ? >10 m β ⁻ ?
Cm 244 18.11 a sf α 5.805, 5.763... γ (43...), e ⁻ , g sf α 15.3, α 1.04	Cm 245 8423 a sf α 5.361, 5.304... γ 175, 133..., e ⁻ , g sf α 369, α 2018	Cm 246 4760 a α 5.386, 5.343... γ (45), e ⁻ , g sf α 1.22, α 0.14	Cm 247 1.56 · 10 ⁷ a α 4.870, 5.267... γ 402, 278..., g α 57, α 83	Cm 248 3.48 · 10 ⁵ a α 5.078, 5.035... γ, e ⁻ , g, sf α 2.63, α 0.37	Cm 249 64.15 m β ⁻ 0.9... γ 634, (560, 369...) e ⁻ α 7, β ⁻ ? α ~80	Cm 250 ~8300 a sf α 7, β ⁻ ? α ~80	Cm 251 16.8 m β ⁻ 1.4... γ 543, 530, 390 438...	Cm 252 16.8 m β ⁻ 1.4... γ 543, 530, 390 438...	Cm 253 16.8 m β ⁻ 1.4... γ 543, 530, 390 438...	Cm 254 16.8 m β ⁻ 1.4... γ 543, 530, 390 438...



High Flux Reactor at ILL



ILL
NEUTRONS
FOR SOCIETY

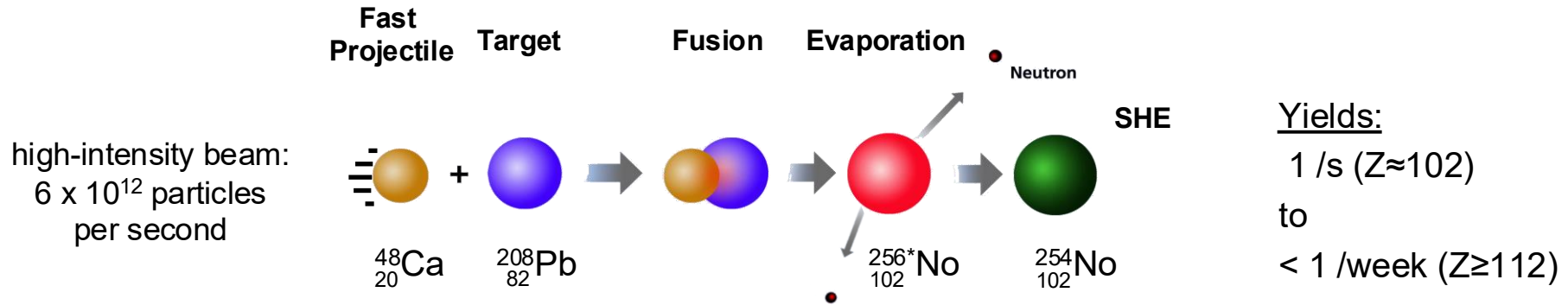
Courtesy S. Berndt

HIM
HELMHOLTZ
Helmholtz Institute Mainz

GSI

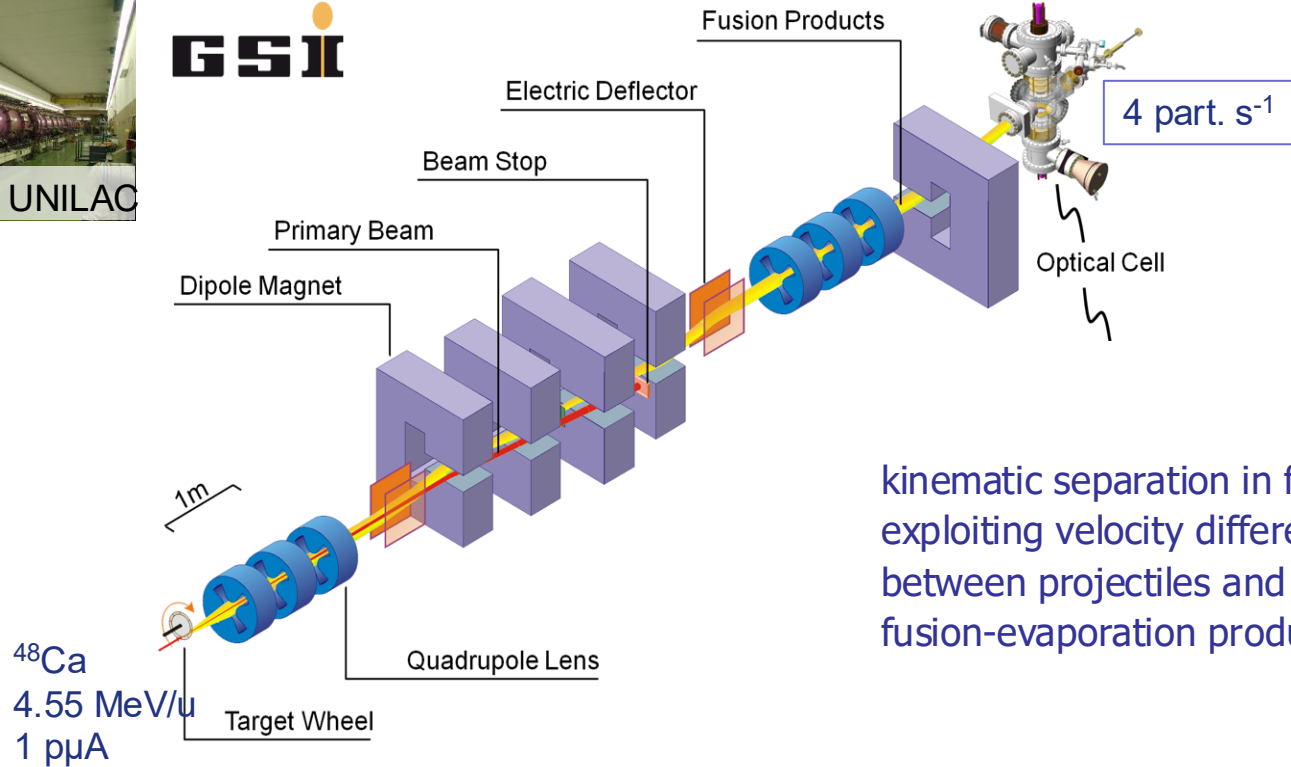
JG|U
JOHANNES GUTENBERG
UNIVERSITÄT MAINZ

Production in Fusion-Evaporation Reactions



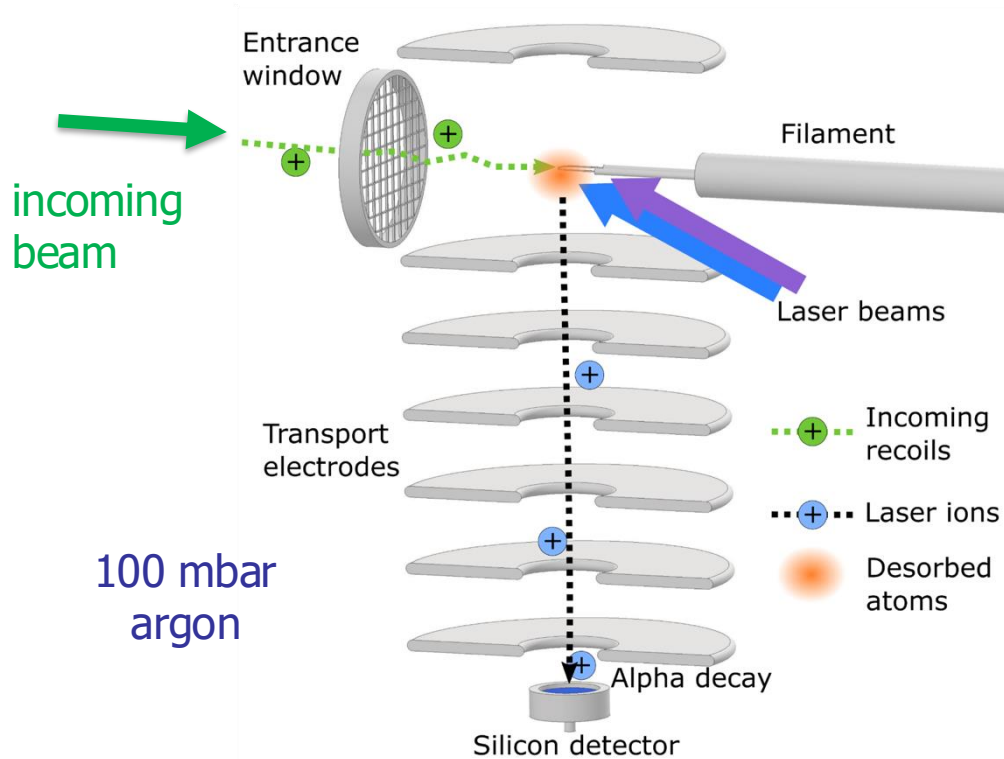
- production in heavy-ion induced fusion-evaporation reactions
- besides low production rates, high energy of reaction products poses challenges for mass spectrometry and laser spectroscopy
- utilizing buffer-gas stopping cells for slowing down and thermalizing ion beams

Production and Separation with Velocity Filter SHIP



kinematic separation in flight
exploiting velocity difference
between projectiles and
fusion-evaporation products

Radiation Detected Resonance Ionization Spectroscopy



Pioneered at Mainz/GSI

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

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

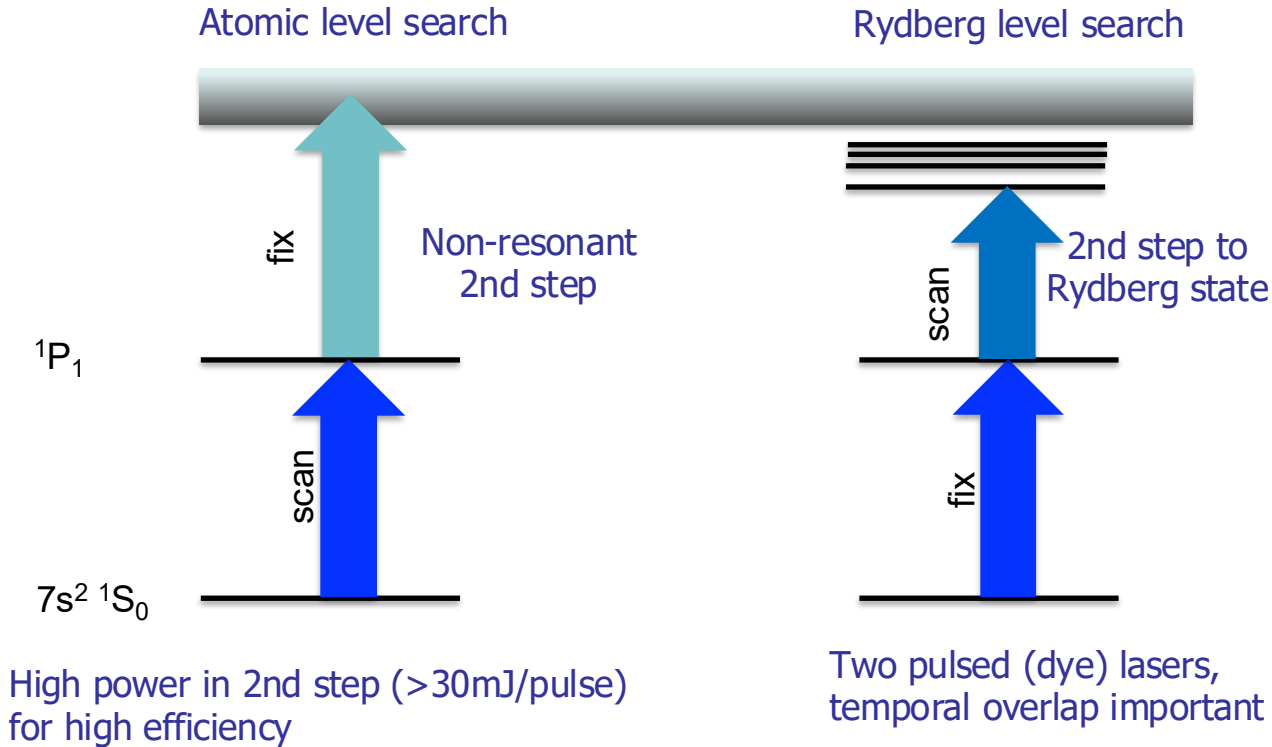
H. Backe et al. Eur. Phys. J. D, 45 (1) (2007), 99

F. Lautenschläger et al. Nucl. Instrum. Meth.

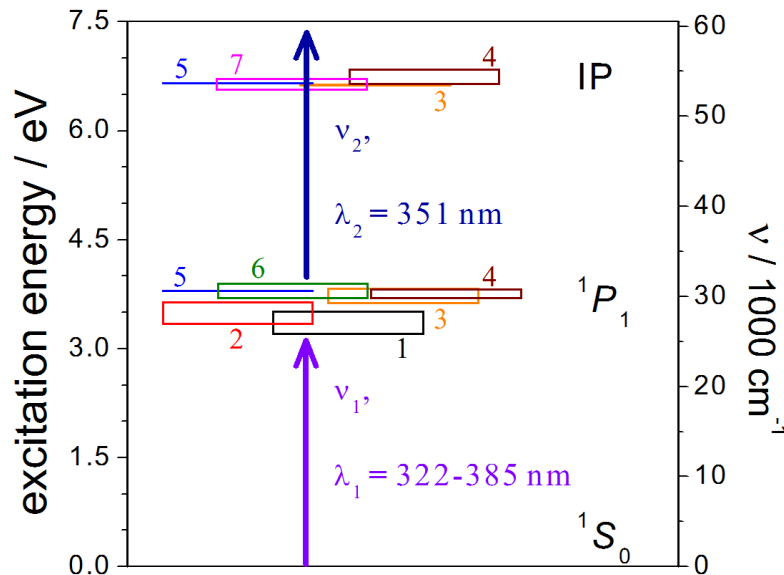
B, 383 (2016), 115

J. Warbinek et al. Atoms (2022)

Resonant Laser Ionization – Nobelium Excitation Schemes



Predictions for Atomic Transitions in Nobelium



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

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

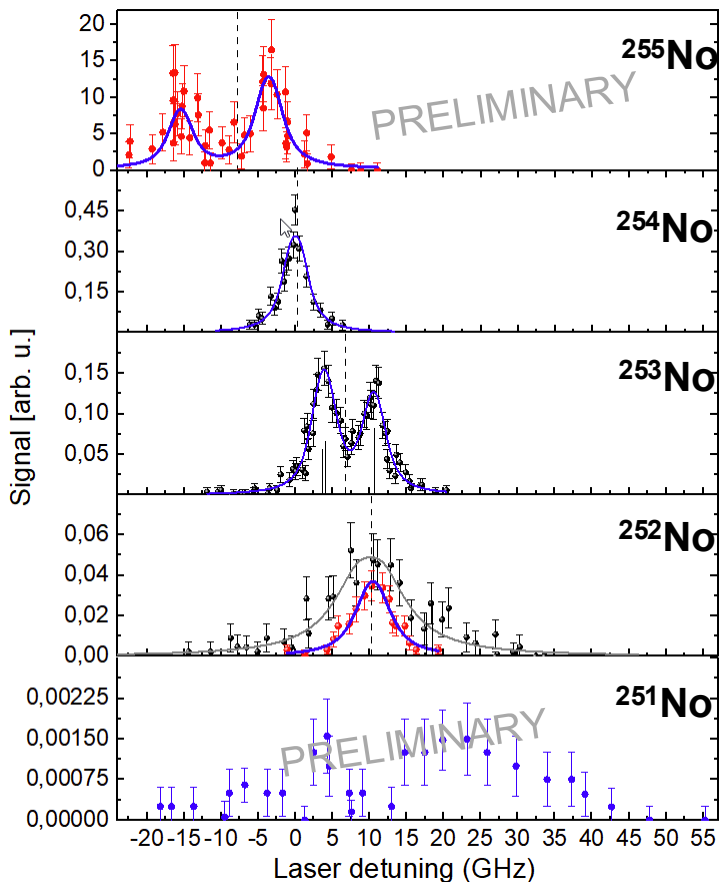
5 (MCDF): Y.Liu et al., Phys. Rev. A 76 (2007) 062503

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**
- **depending on atomic properties, one step may take several minutes**

Laser Spectroscopy of Nobelium (Z=102) Isotopes



Experiment: S. Raeder *et al.*

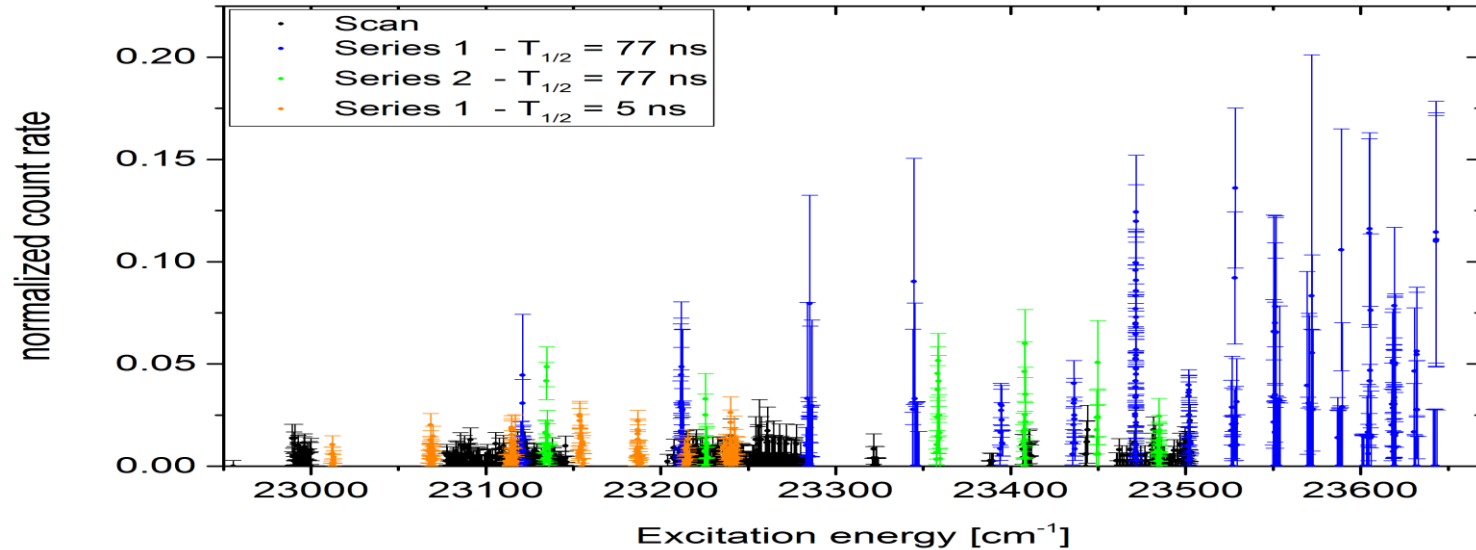
Theory: A. Borschevsky V. Dzuba, S. Fritzsche,
B. Schütrumpf, W. Nazarewicz *et al.*

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



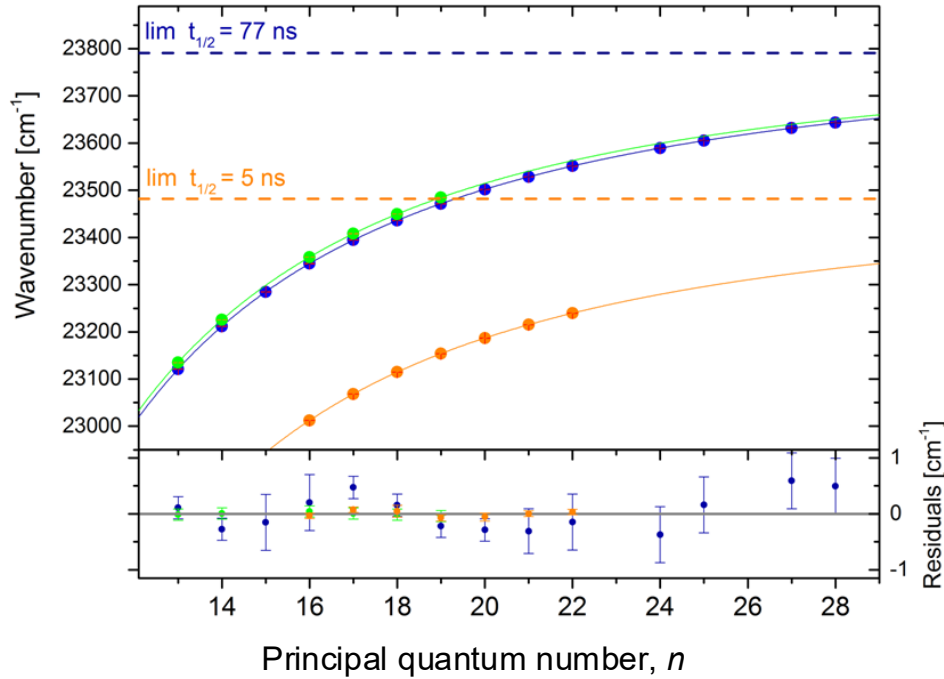
M. Laatiaoui *et al.*, Nature 538, 495 (2016)
S. Raeder *et al.*, Phys. Rev. Lett. 120 (2018) 232503

Search for Rydberg States in ^{254}No

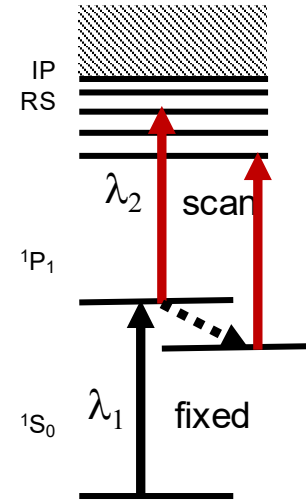


- Rydberg series starting from different states observed and identified
- IP and energy difference of D state can be determined accurately

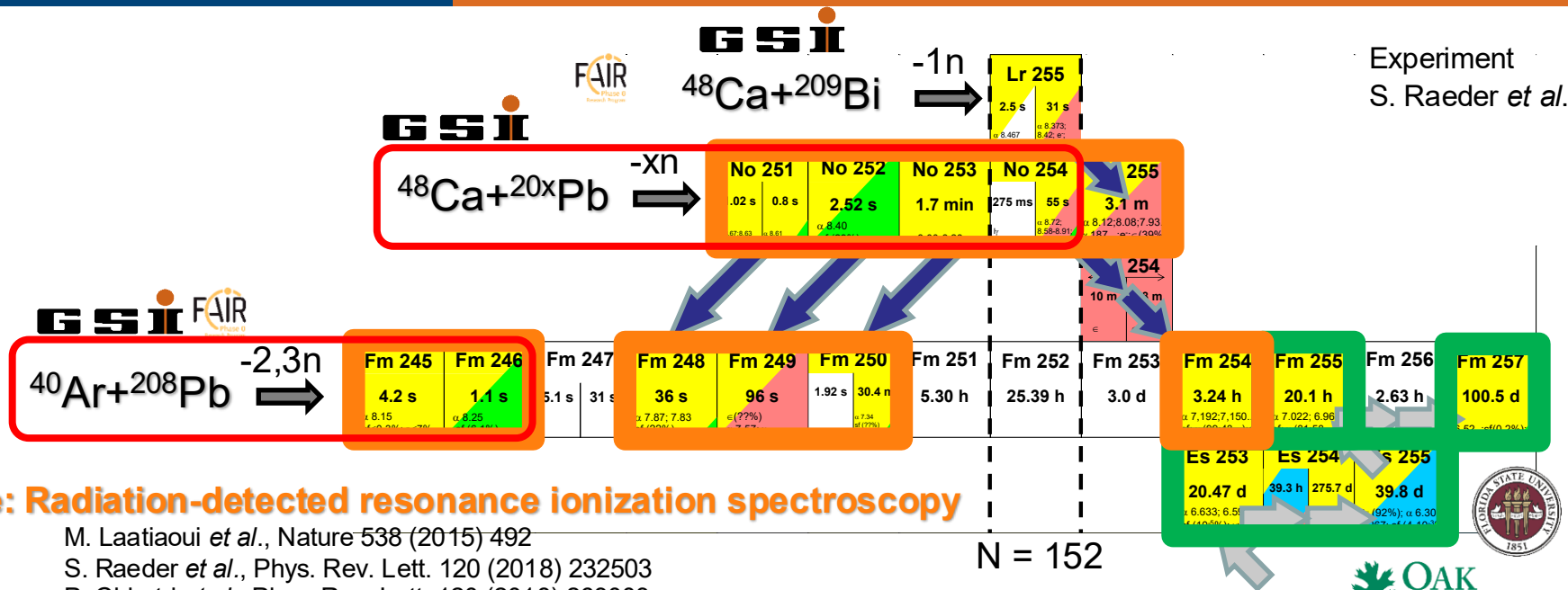
Ionization limits



	IP (eV)
This work	6.6261(3)
Theory (Borschevsky et al.)	6.632
Extrapolation (Sugar)	6.65(7)



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



Experiment
S. Raeder *et al.*

Online: Radiation-detected resonance ionization spectroscopy

M. Laatiaoui *et al.*, Nature 538 (2015) 492
S. Raeder *et al.*, Phys. Rev. Lett. 120 (2018) 232503
P. Chhetri *et al.*, Phys. Rev. Lett. 120 (2018) 263003

Offline: Resonance ionization spectroscopy of Es

S. Nothhelfer *et al.*, PRC 105 (2022) L021302
F. Weber *et al.*, PRR 4 (2022) 043053

From HFIR

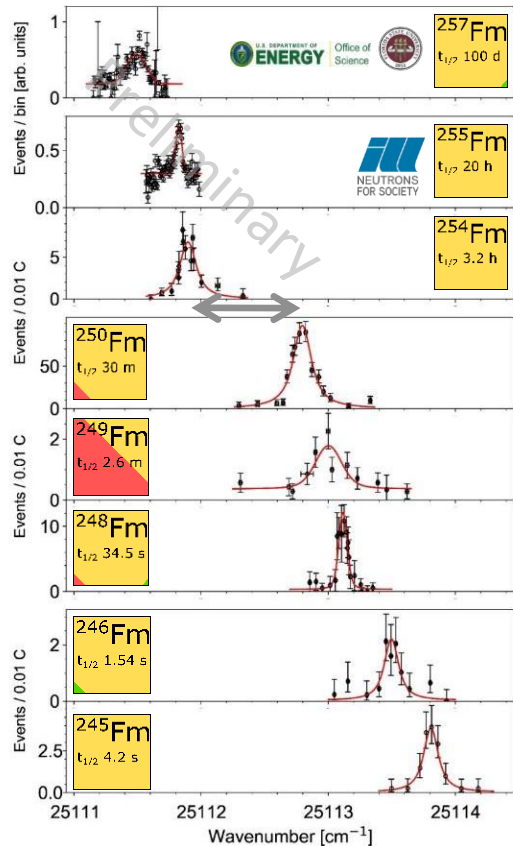
OAK RIDGE
National Laboratory

Es reirradiated at

Laser Spectroscopy of Fm (Z=100) Isotopes

Smooth trends in fermium charge radii and the impact of shell effects

J. Warbinek et al., Nature 634, 1075 (2024) **nature**

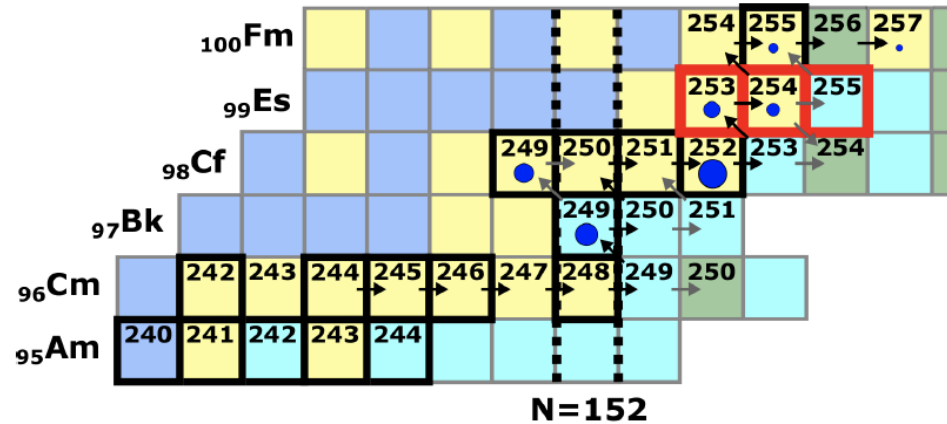


- Determination of change in nuclear charge radii from isotope shifts for a long chain of Fm isotopes produced in different reactions
- comparison to several nuclear EDF calculations

Es – Fm Sample available from Oak Ridge

- Small amount of Es/Fm as „leftovers“ in the ^{252}Cf production process
- 2019: Fm sample available from Florida
- Radiochemical separation of Es/Fm

**High Flux Isotope Reactor
Oak Ridge National Laboratory**



REDC-2606-B		
Cf-249	5.10E-03	µg.
Es-253	2.29E-03	µg.
Es-254	4.02E-03	µg.
Fm-257	1.38E-06	µg.

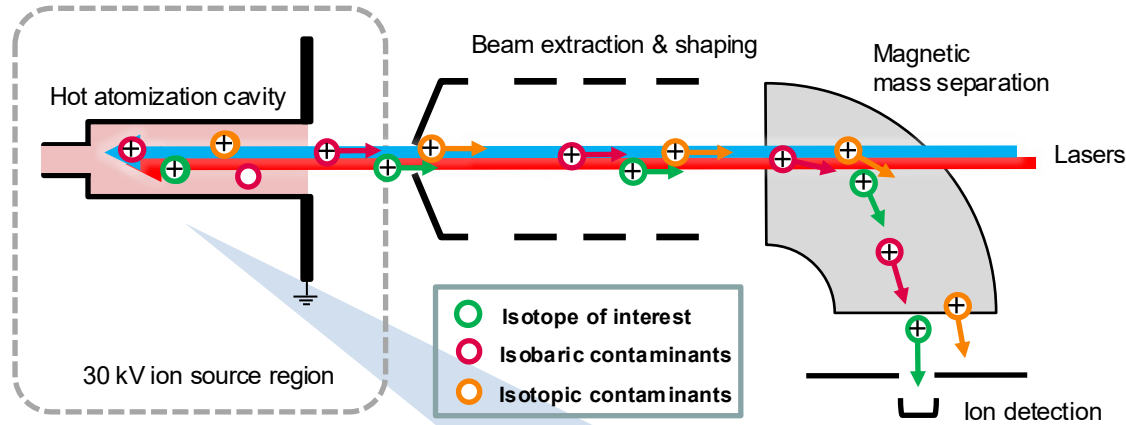
10^9 atoms



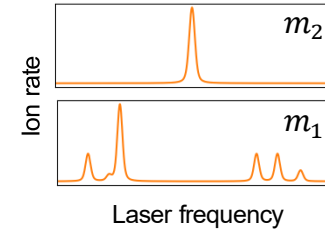
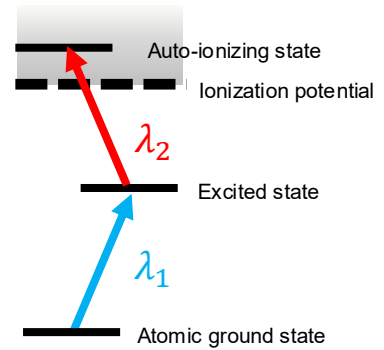
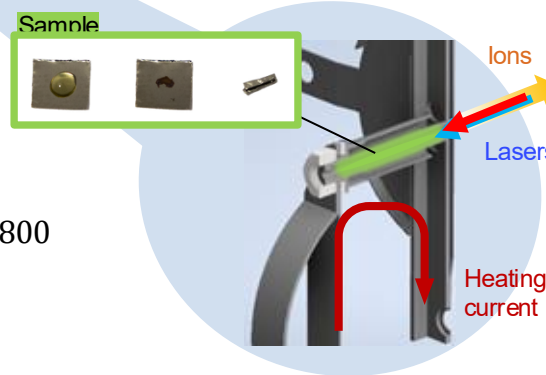
U.S. DEPARTMENT OF
ENERGY

Office of
ScienceJOHANNES GUTENBERG
UNIVERSITÄT MAINZ

RISIKO Setup at Mainz University



- Chemical sample preparation
- Sample evaporation in hot cavity
- Multi-step insource photoionization by pulsed lasers
- Mass separation in dipole magnet $\frac{m}{\Delta m} \approx 800$
- Single ion detection

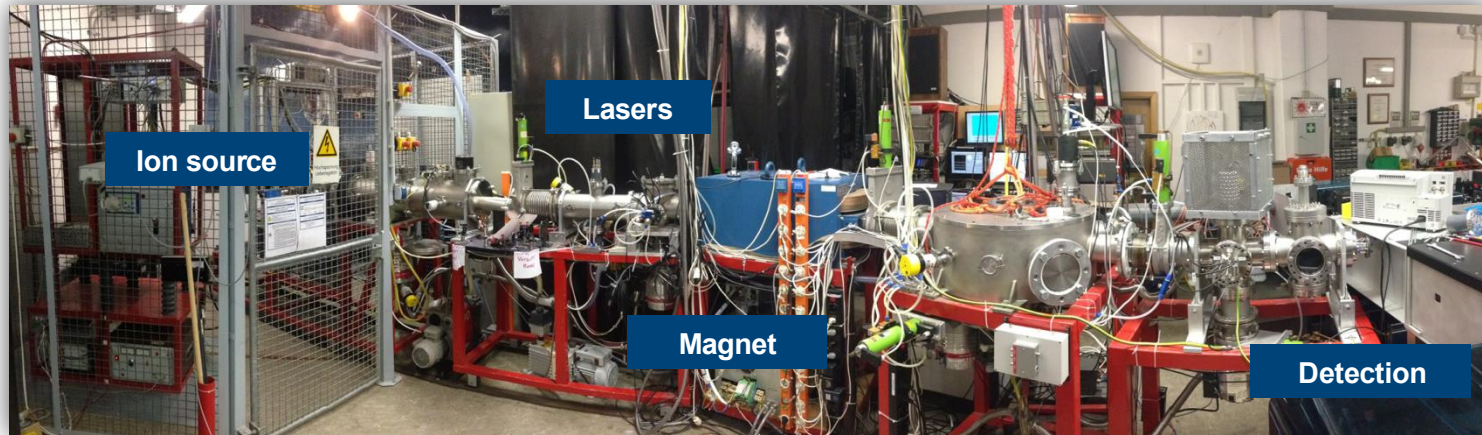
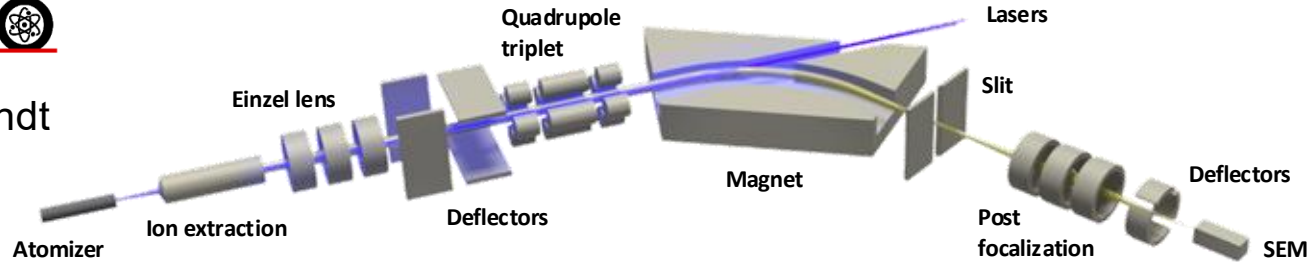


RISIKO

RISIKO Setup at Mainz University



Prof. K. Wendt

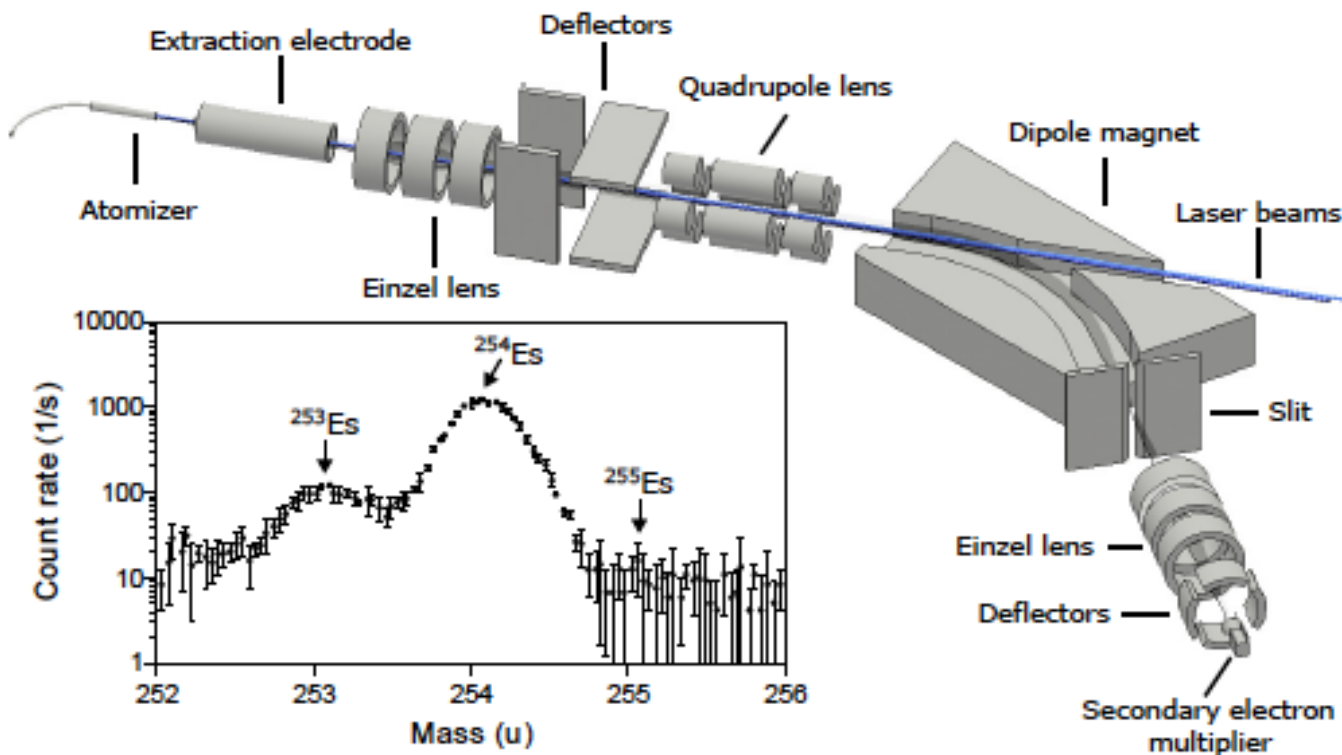


T. Kieck et al., NIM A 945, 162602 (2019).

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

Hyperfine Spectroscopy of Es at RISIKO/Mainz

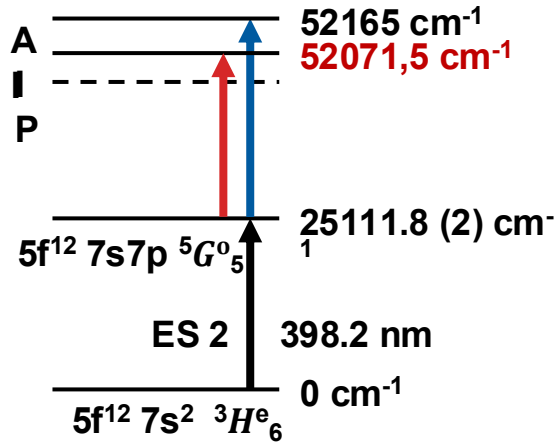
RISIKO separator
K. Wendt et al.



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

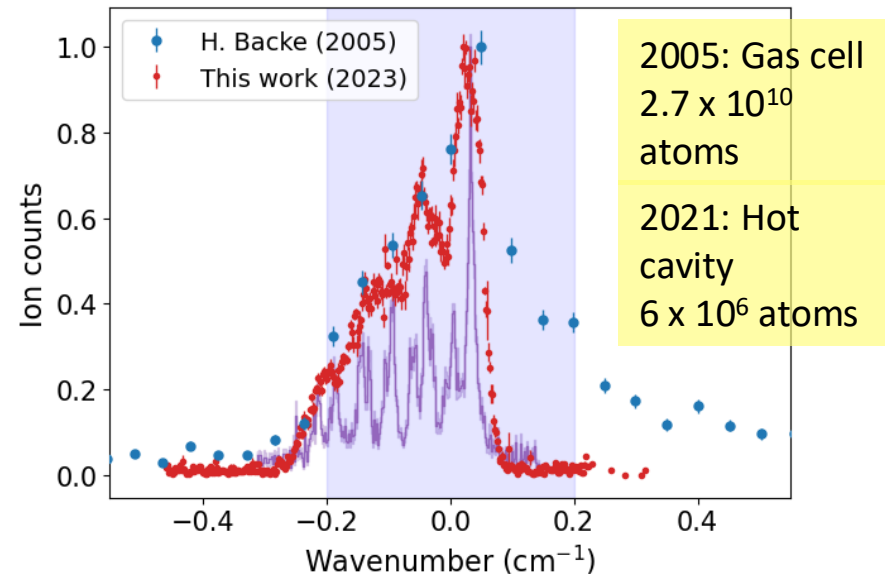
Laser Spectroscopy of Fm Isotopes

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



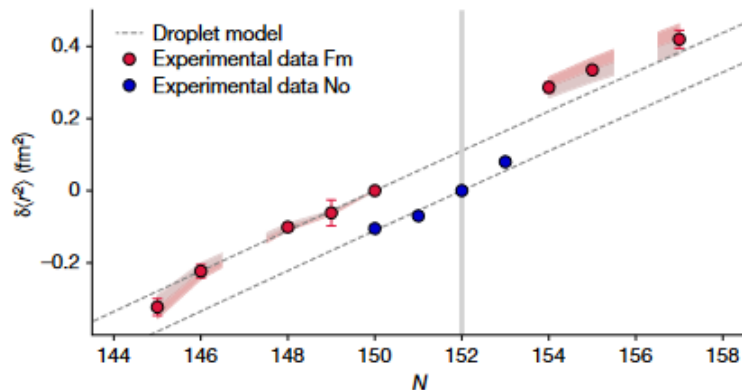
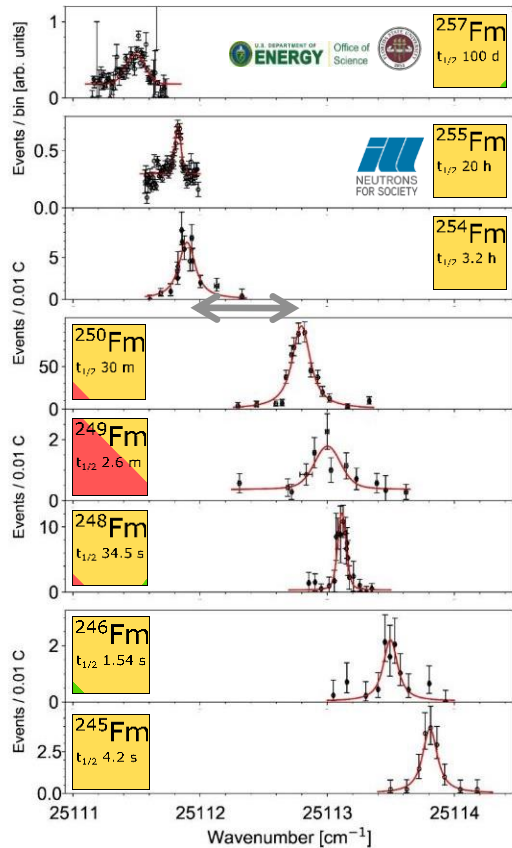
RISIKO mass separator in Mainz

- laser spectroscopy on Fm samples with only 10^7 atoms



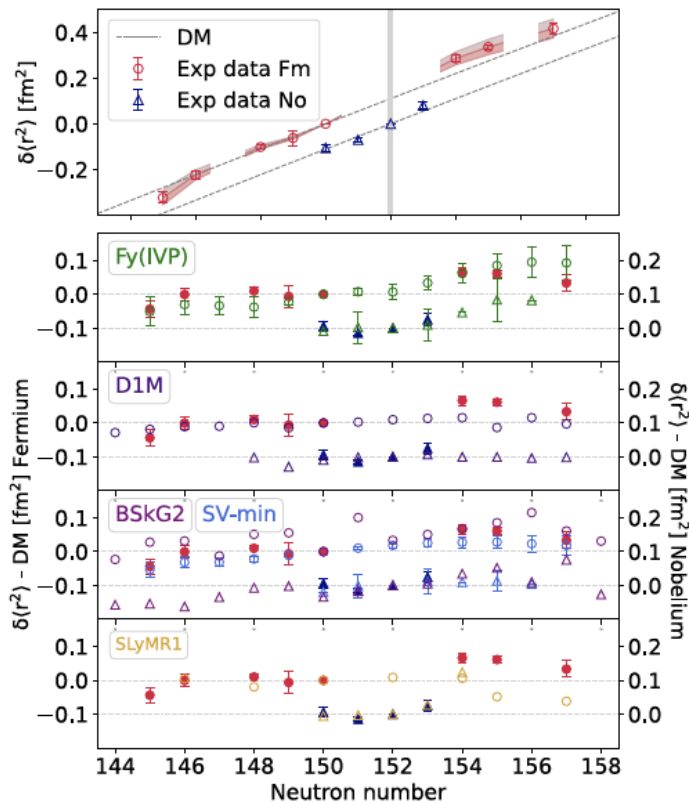
Laser Spectroscopy of Fm (Z=100) Isotopes

- isotope shifts in Fm isotope chain produced in different direct and indirect schemes
- determined change in nuclear charge radii from isotope shift with F factor from atomic theory



J. Warbinek et al., Nature 634, 1075 (2024) and PhD thesis University of Mainz

Results – Comparison with Theoretical Calculations



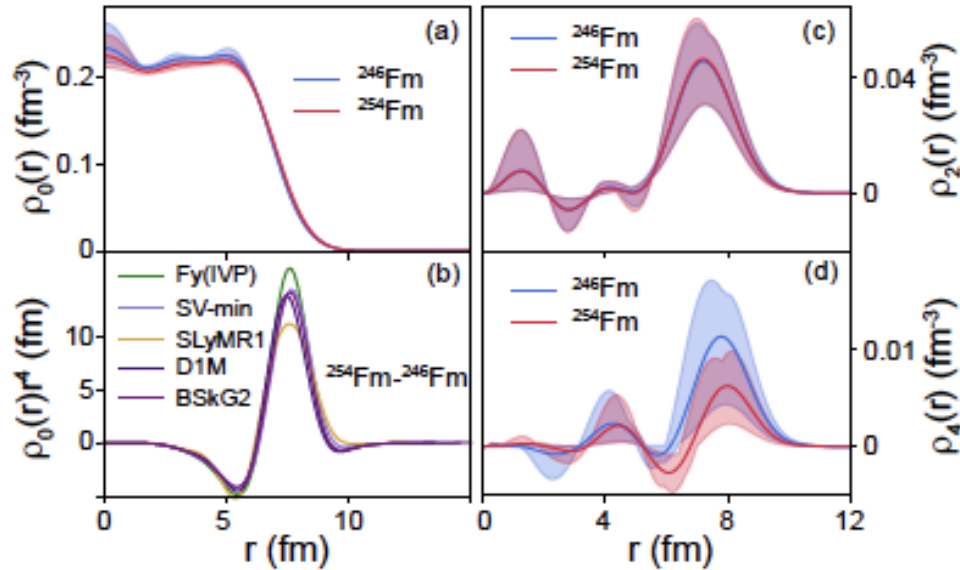
calculations of charge radii using different nuclear energy density functionals by:

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

J. Warbinek et al., Nature 634, 1075 (2024)

Theoretical calculations

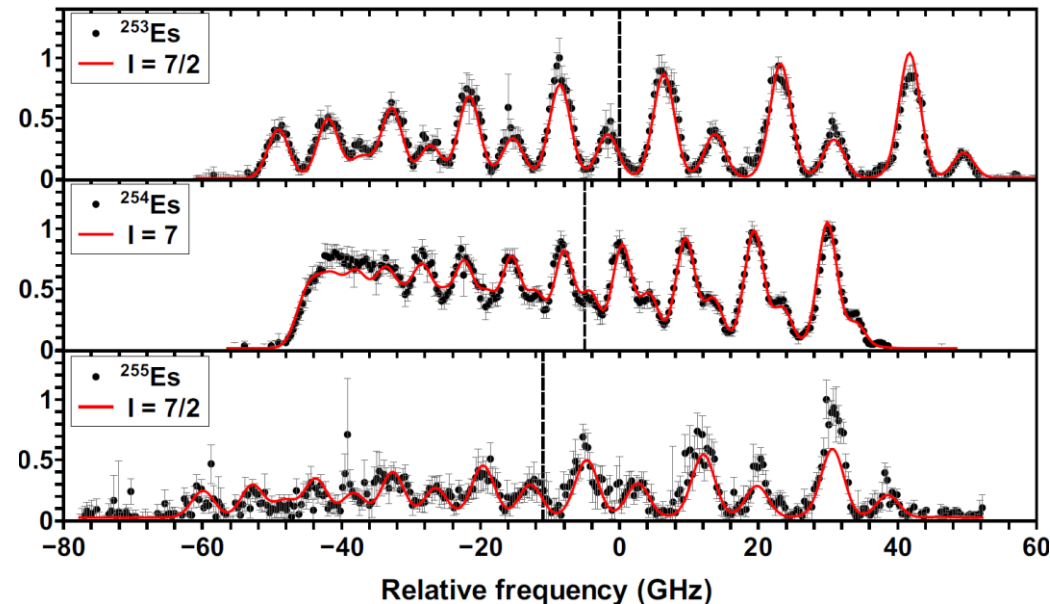


B. Bally, M. Bender, S. Goriely, S. Hilaire, W. Nazarewicz, S. Peru, P.G. Reinhard, W. Ryssens

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

J. Warbinek et al., Nature 634, 1075 (2024)

Hyperfine Spectroscopy of Es at RISIKO/Mainz



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



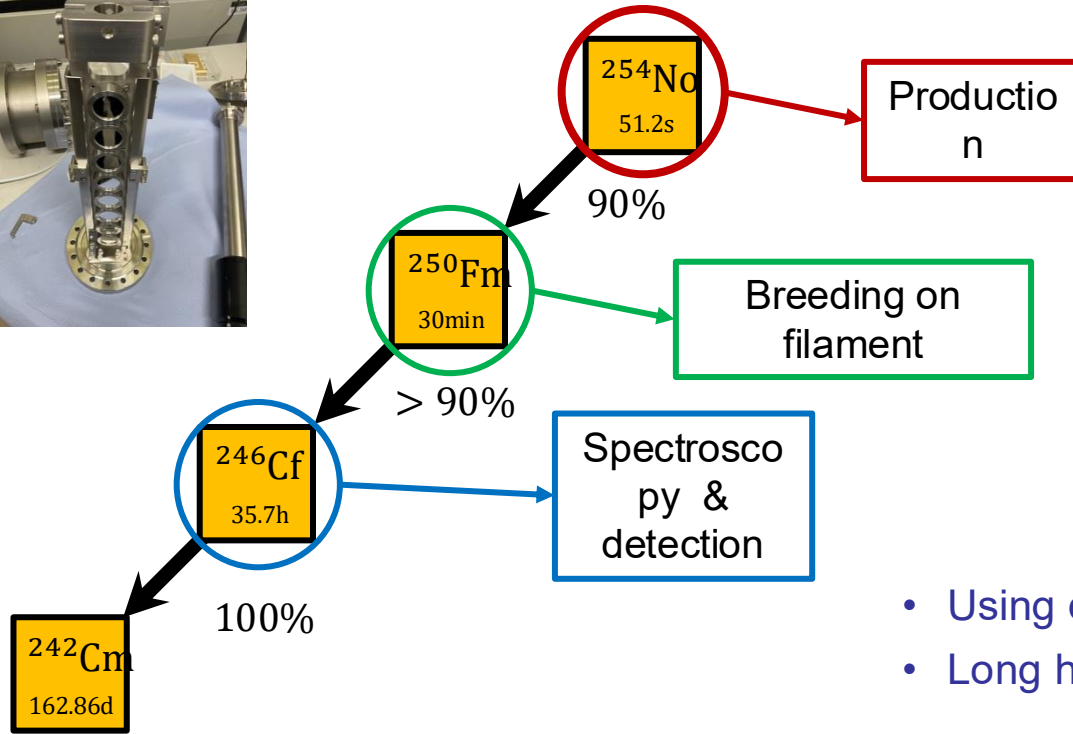
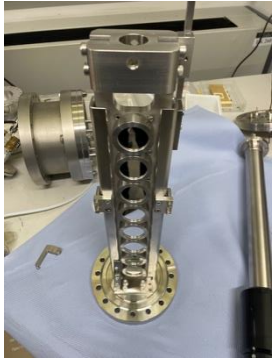
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Laser Spectroscopy of Californium Isotopes

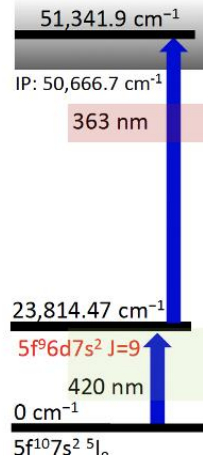


Isotope	Reaction
^{240}Cf	$^{195}\text{Pt}(^{48}\text{Ca}, 3n)$
^{241}Cf	$^{196}\text{Pt}(^{48}\text{Ca}, 3n)$
^{242}Cf	$^{196}\text{Pt}(^{48}\text{Ca}, 2n)$
^{244}Cf	$^{198}\text{Pt}(^{48}\text{Ca}, 2n)$
^{246}Cf	$^{208}\text{Pb}(^{48}\text{Ca}, 2n)$ 90% α , >90% α

- Using direct and indirect production schemes
- Long half-life of ^{246}Cf needs tailored detector

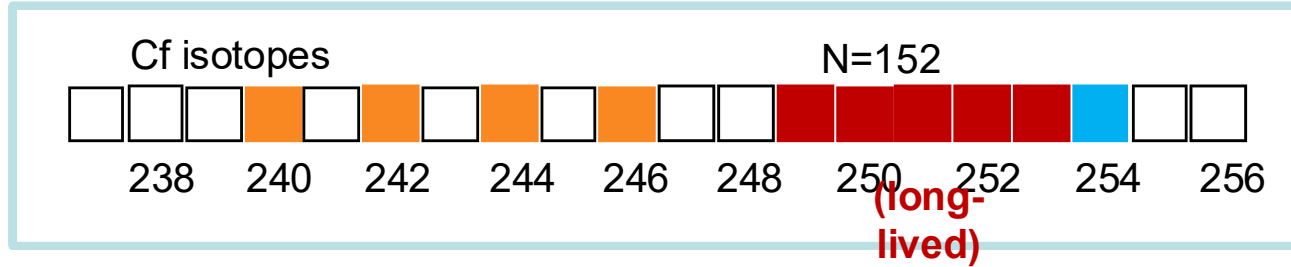
K. van Beek (PhD thesis), S. Raeder et al.

Laser Spectroscopy of Californium Isotopes



Goal: investigate the evolution of size and shape in the heavy elements

- extend systematic studies of radii and nuclear moments in Cf, Fm, and No isotopic chains around $N = 152$
- compare to state-of-the-art theoretical models



Nuclear structure
Nuclear Astrophysics (Kilonova)

