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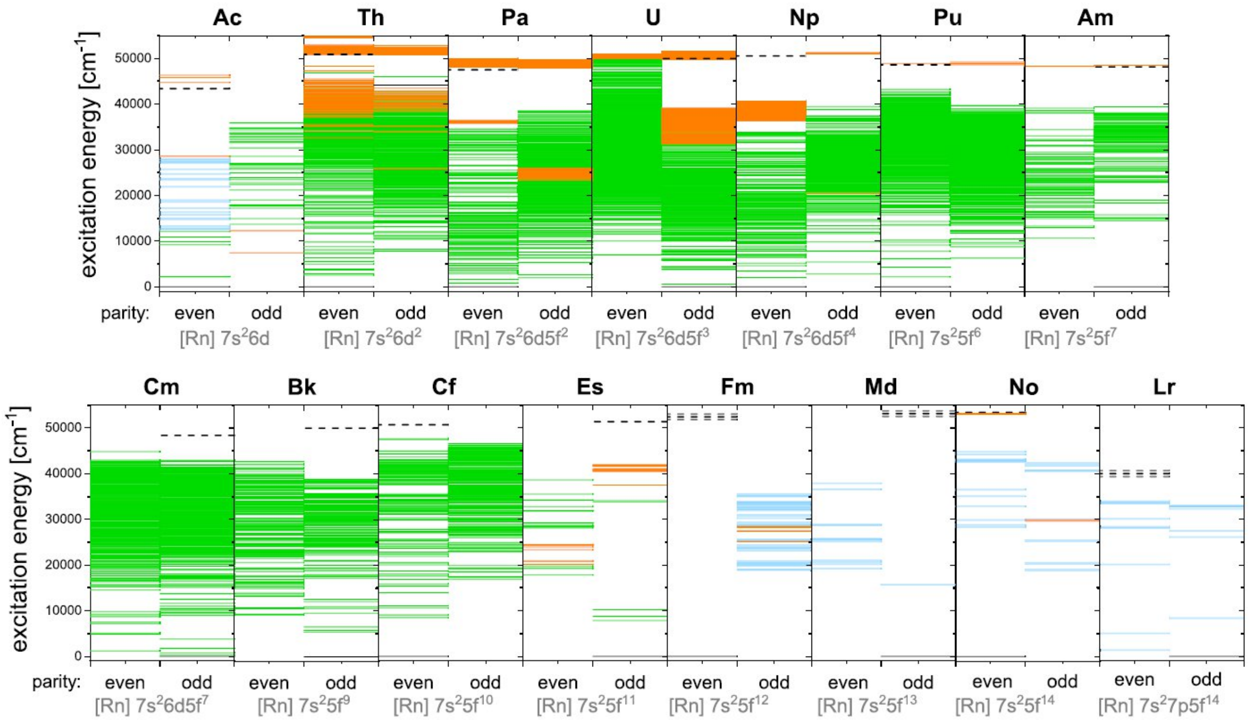
Laser spectroscopy of fermium isotopes across the N =152 shell gap

Dominik Studer for the Fermium collaboration

Helmholtz Institute Mainz
GSI Darmstadt

TASCA workshop 2025

Atomic levels of the actinides



[1]

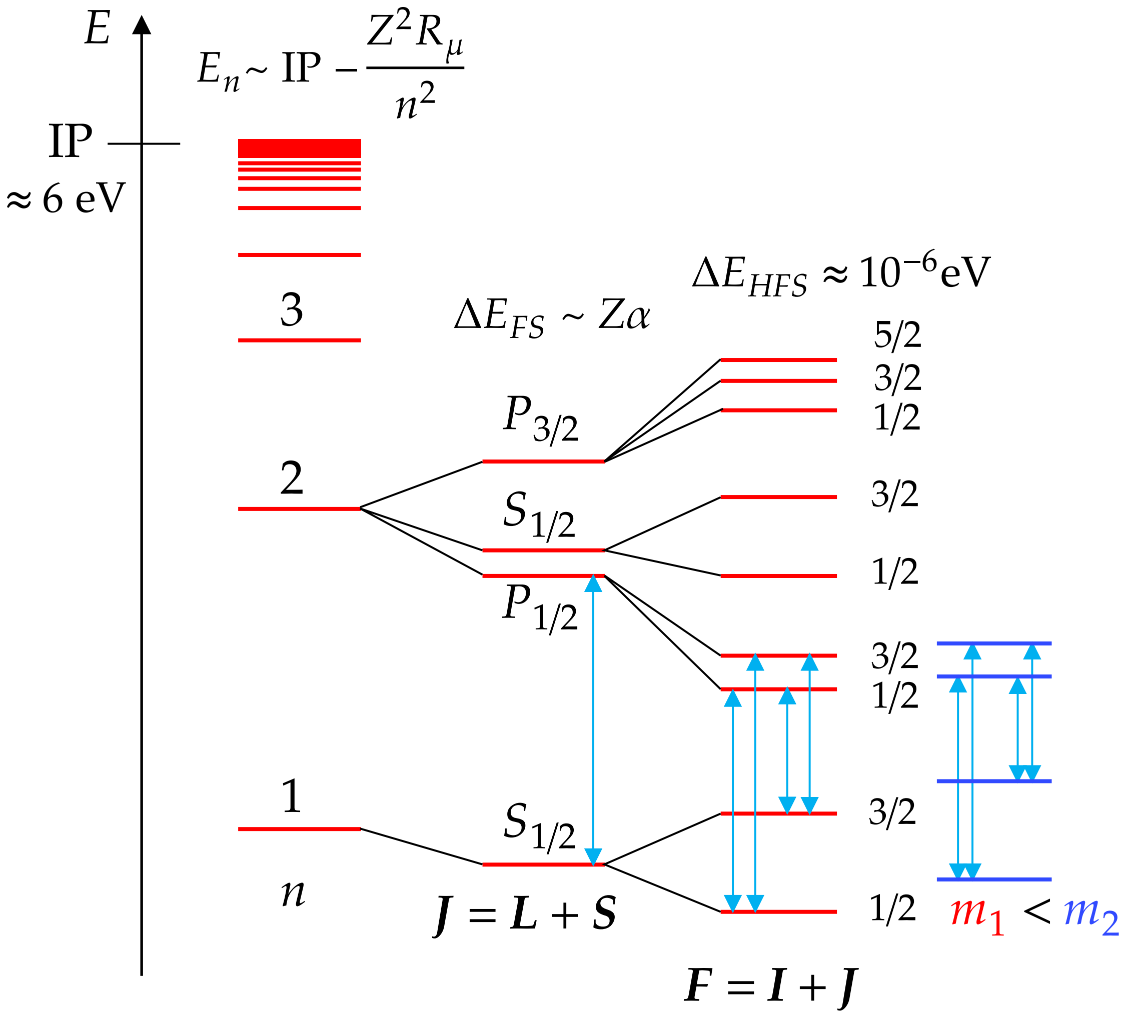
- Blaise, Wyart 1992 [2]
- Levels from recent experiments
- Theory – calculated levels

- Actinide atoms exhibit extremely complex spectra
  - Configuration interactions
  - Relativistic corrections
- Theoretical predictions become increasingly challenging
- Spectral data sparse due to limited availability or low production rates

High-efficiency spectroscopic experiments required!

[1] M. Block, M. Laatiaoui, S. Raeder, Prog. Part. Nucl. Phys. **116**, 103834 (2021)
[2] J. Blaise, J. F. Wyart, *Selected constants energy levels and atomic spectra of actinides*, Paris 1992

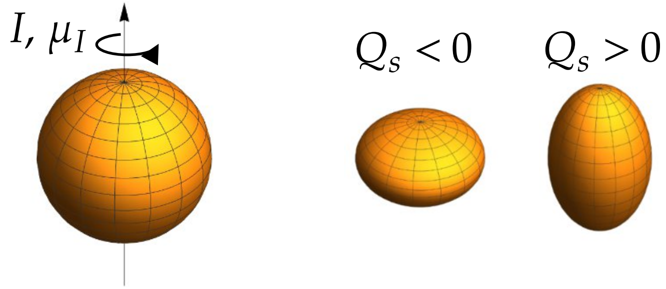
Probing atomic and nuclear structure



- Atomic level structure through optical transitions
- Ionization limit through Rydberg convergences
- Nuclear spins and electromagnetic moments through hyperfine interaction

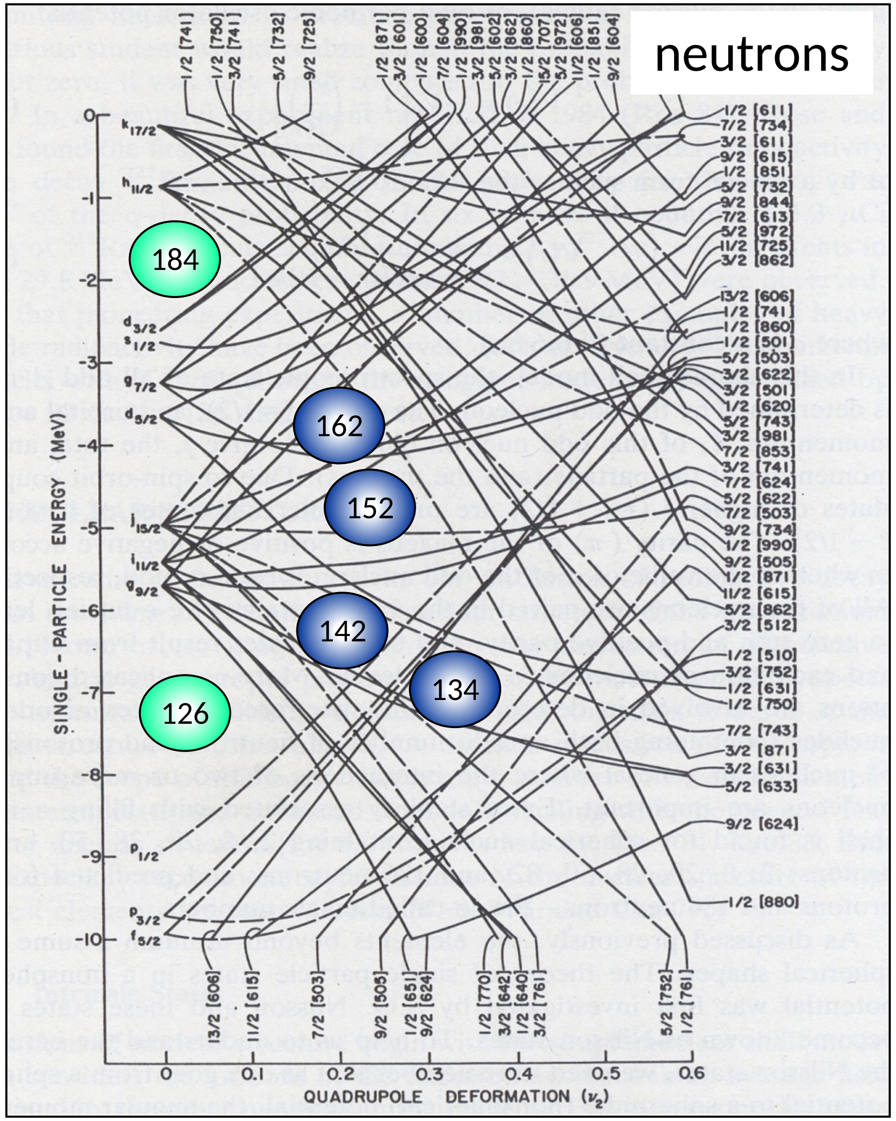
$$A = \frac{\mu_I \overline{H(0)}}{IJ} \qquad B = eQ_s \left\langle \frac{\partial^2 \phi}{\partial z^2} \right\rangle_{r=0}$$

Atomic shell
Nucleus



- Relative charge radii through isotope shifts

Probing atomic and nuclear structure



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$$A = \frac{\mu_I \overline{H(0)}}{IJ} \qquad B = eQ_s \left\langle \frac{\partial^2 \phi}{\partial z^2} \right\rangle_{r=0}$$

Atomic shell
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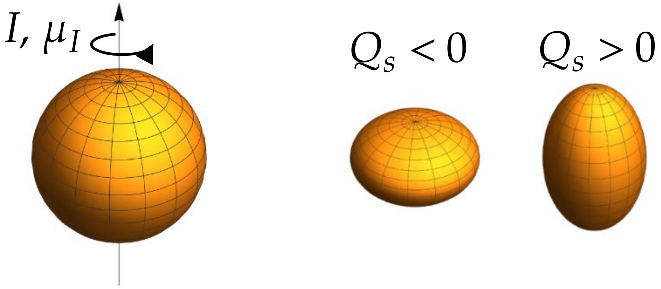
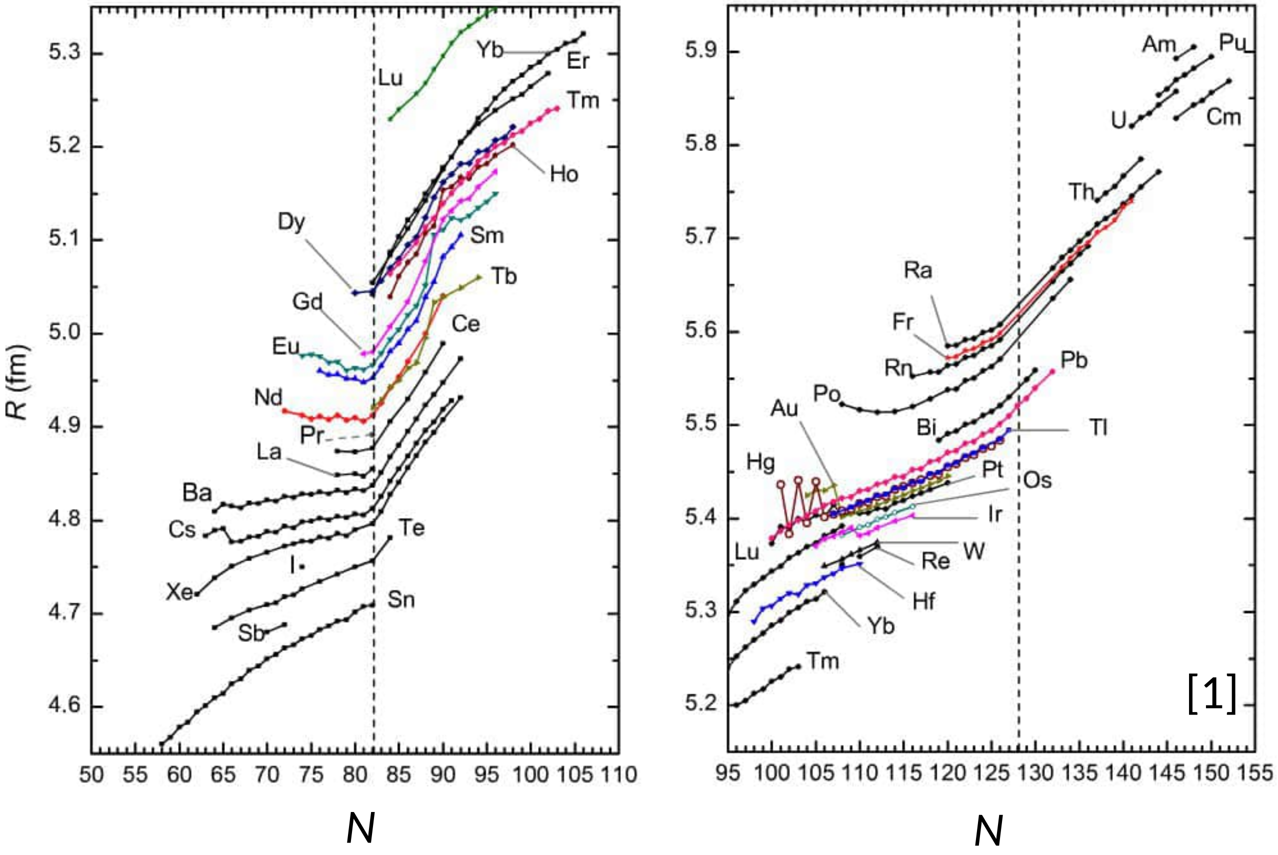


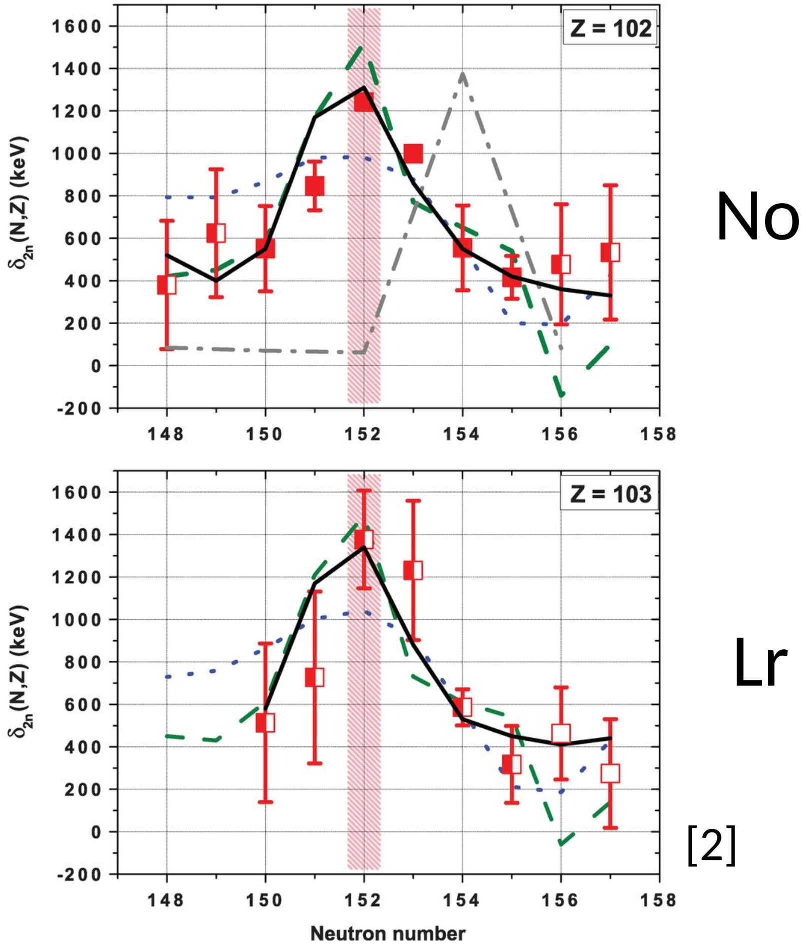
- Relative charge radii through isotope shifts

Nuclear shell gap signatures

Trends in nuclear charge radii
→ Laser spectroscopy



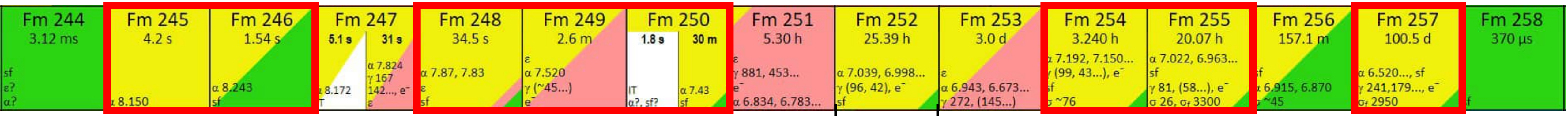
Binding energies
→ Mass measurements



*[1] I. Angeli, K.P. Marinova, Atomic Data and Nuclear Data Tabes **99**, 69 (2013)*
*[2] E. Minaya Ramirez et al., Science **337**,1207 (2012)*

Fermium laser spectroscopy campaigns

- 2000 Mainz off-line Fm-255
- 2019 Mainz off-line Fm-257
- 2019 GSI on-line Fm-248-250
- 2020 GSI on-line Fm-249,254
- 2021 Mainz off-line Fm-255
- 2021 GSI on-line Fm-245,246
- 2023 Mainz off-line Fm-255
- 202x GSI on-line Fm-252

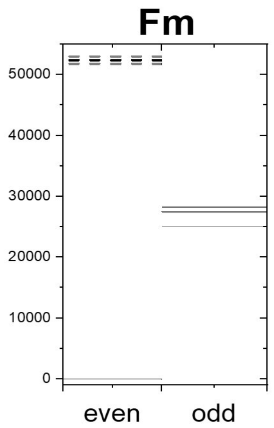


N=152

Results of the Fm campaign in 2000

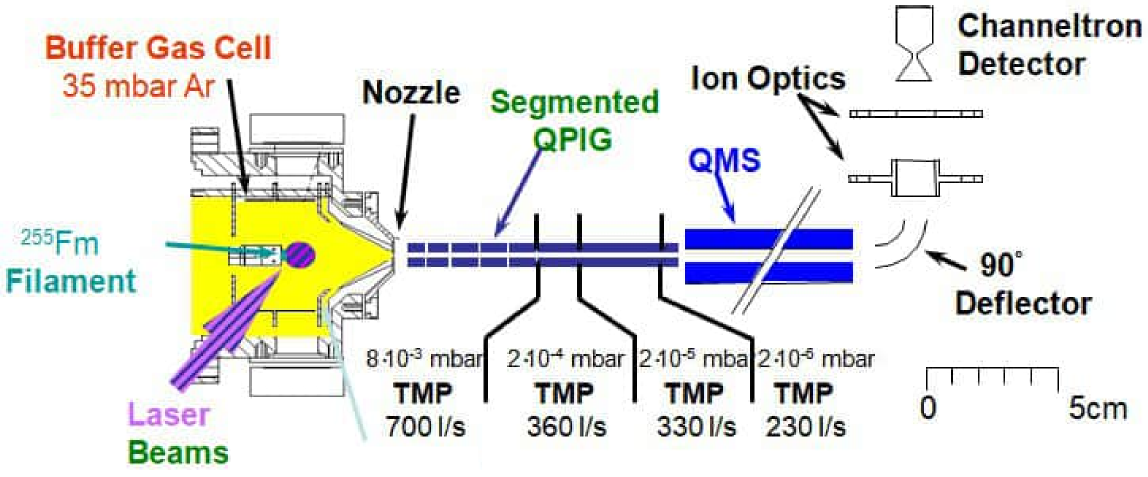
Atomic spectroscopy of ^{255}Fm [1,2]

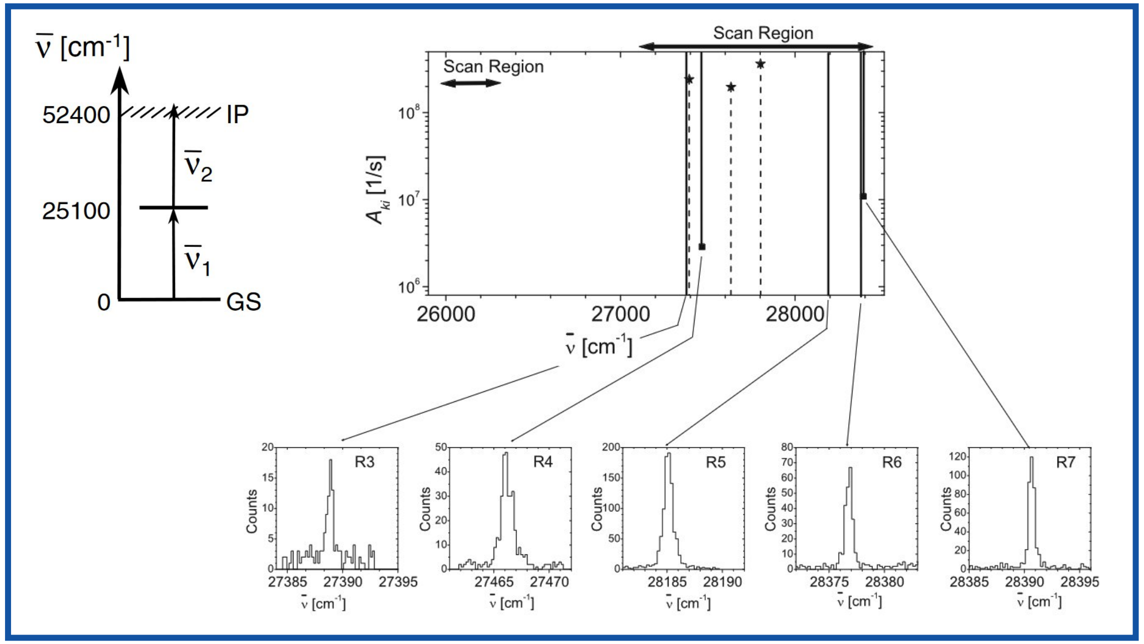
- Evaporation from hot filament
- Spectroscopy in 35 mbar Ar gas
- 200 Hz rep. rate dye laser system



Ionization energy with large uncertainty [3]

7 known atomic levels in Fm [1,2]





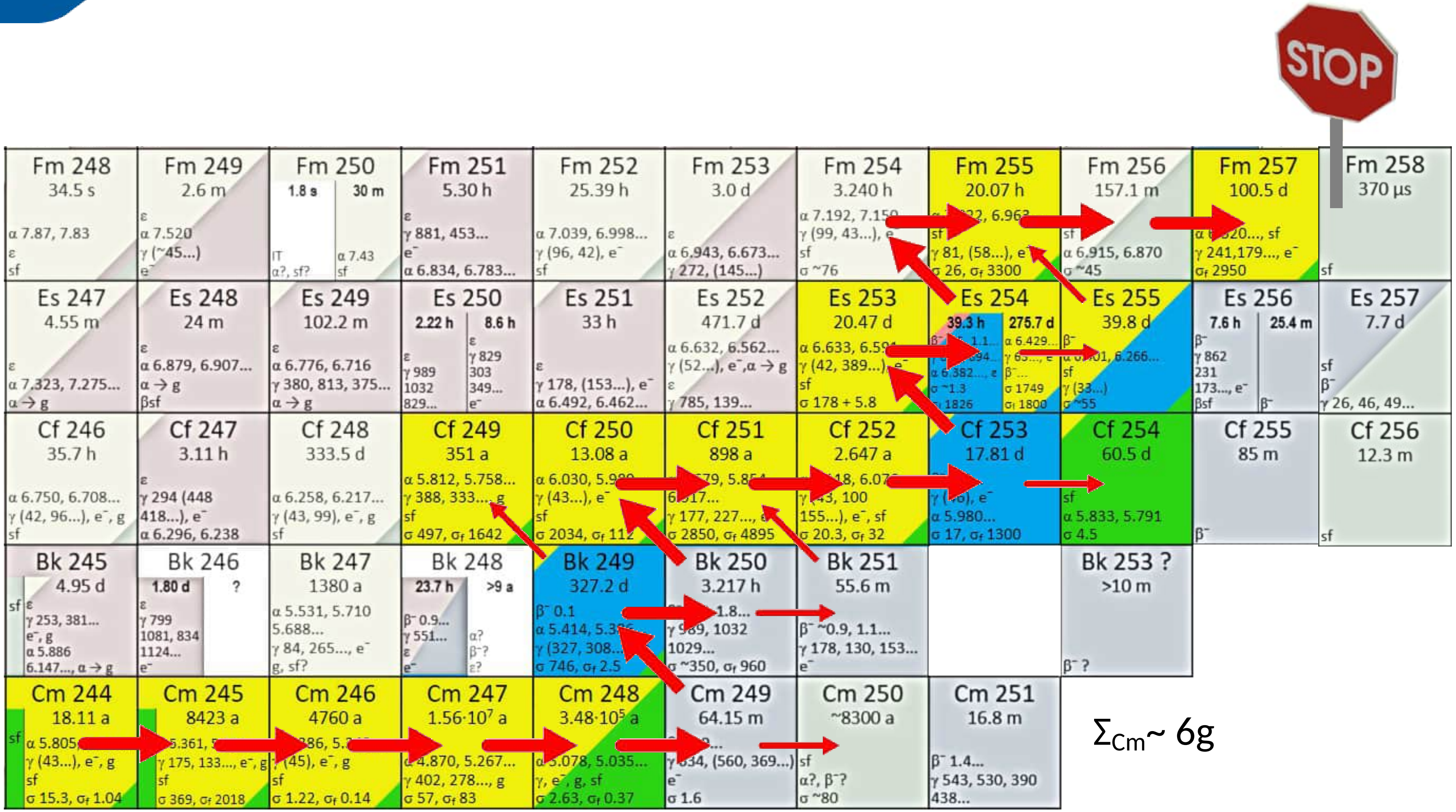
[1] M. Sewtz et al., Phys. Rev. Lett. **90**, 163002 (2003)
[2] H. Backe et al., Hyperfine Interact. **162**, 3 (2006)
[3] T. K. Sato et al., J. Am. Chem. Soc. **140**, 14609 (2018)

Production of heavy actinides for off-line studies

High Flux Isotope Reactor (HFIR) at ORNL

U.S. DEPARTMENT OF ENERGY | Office of Science

Oak Ridge National Laboratory

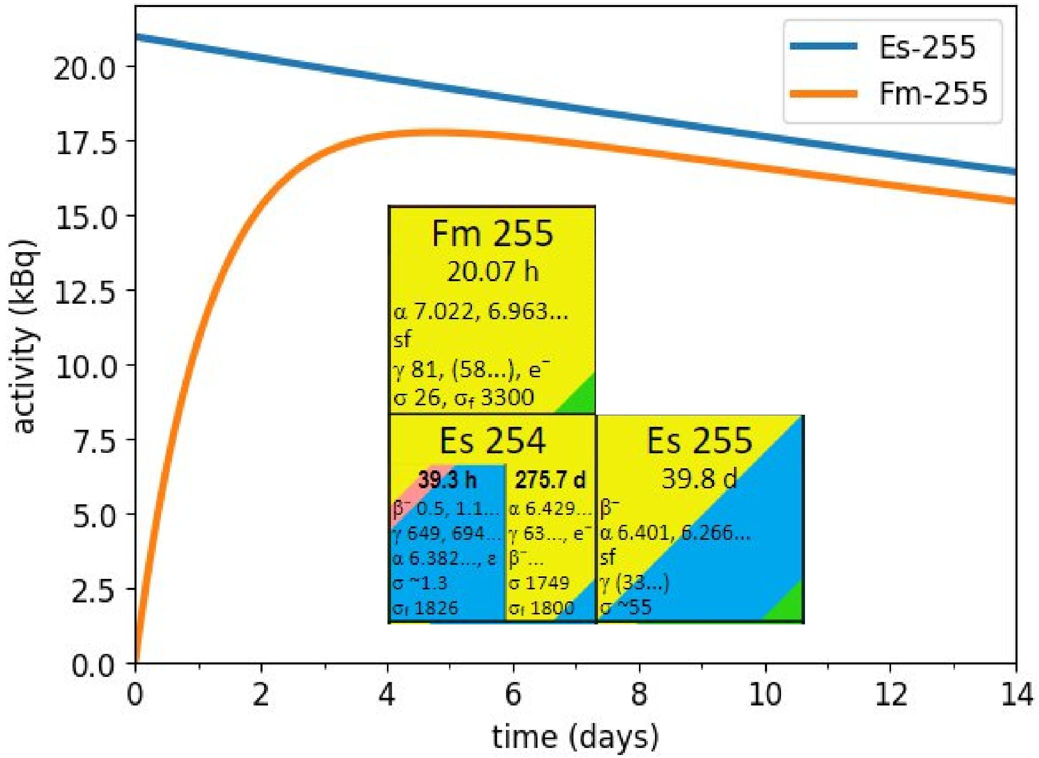


Σ_{Cm}~ 6g

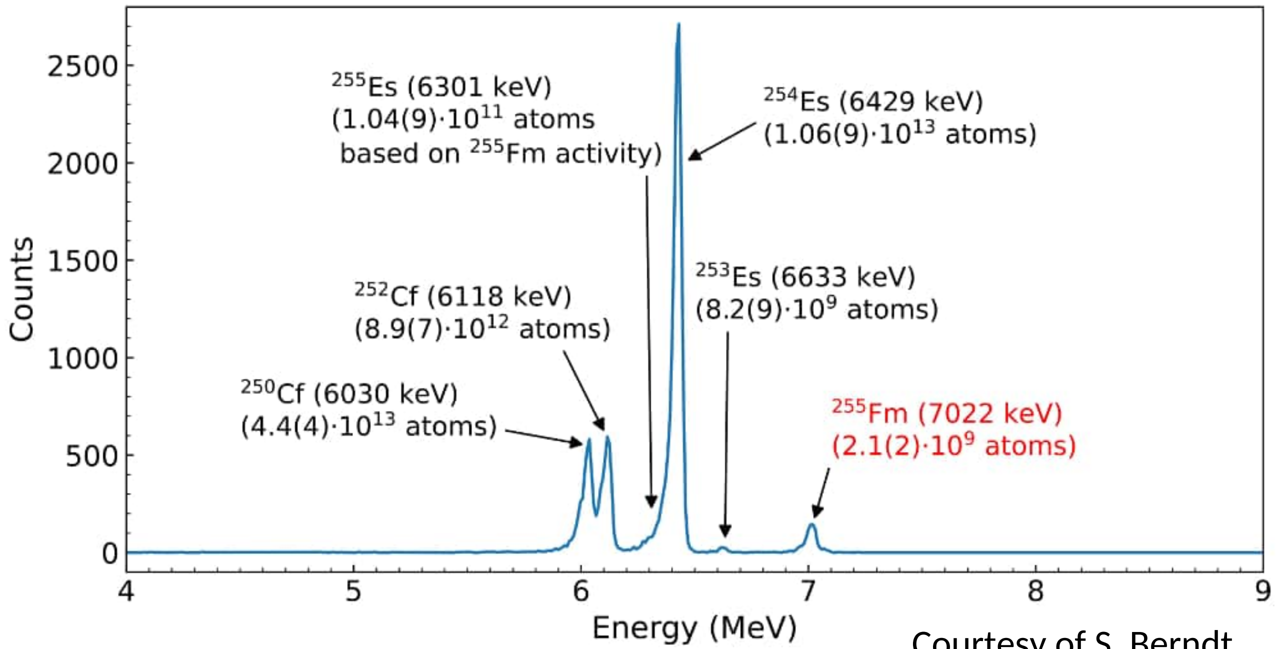
The isotopes used in this research were supplied by the U.S. Department of Energy, Office of Science, by the Isotope Program in the Office of Nuclear Physics. The ^{253,254,255}Es and ^{255,257}Fm were provided to Florida State University and the University of Mainz via the Isotope Development and Production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

^{255}Es/^{255}Fm generator system

1) Production of Es-255 by neutron irradiation of the “long-lived” Es-254 at ILL
2) Es-255 decays with 92% by β^- to Fm-255
3) Upon approaching equilibrium (few days) the Fm is extracted with an ion exchange column

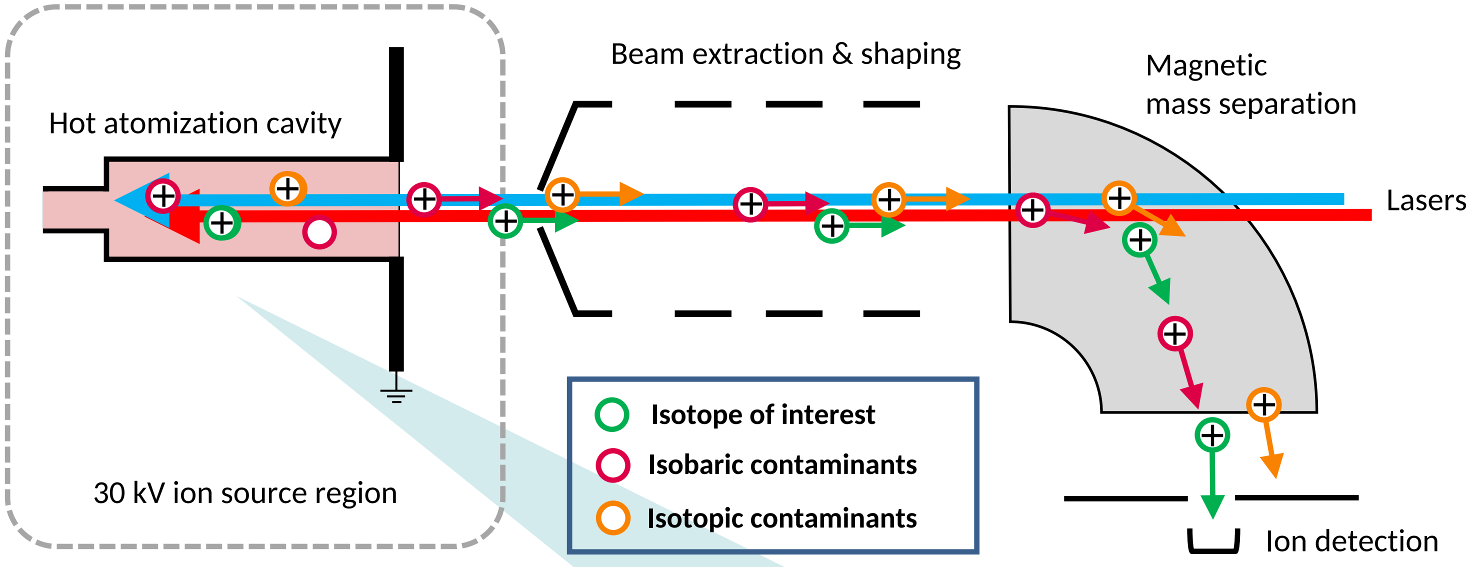


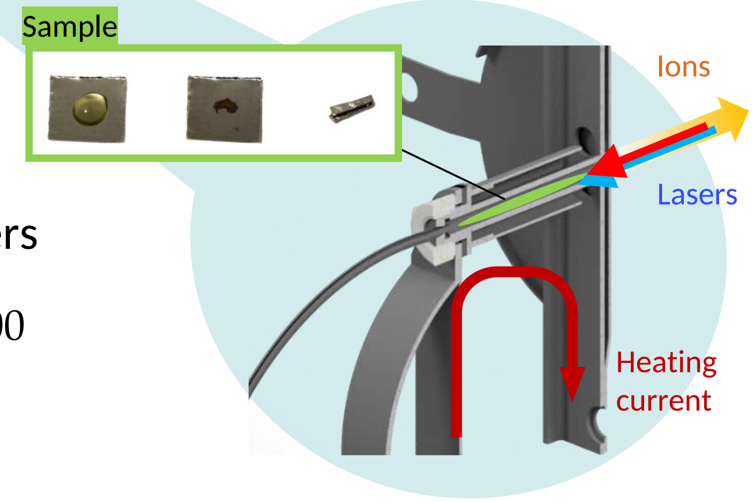
Alpha spectrum of an aliquot

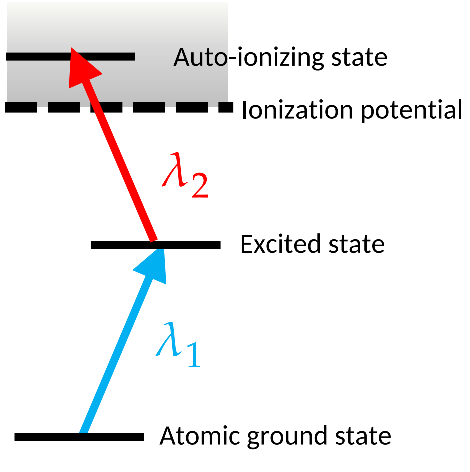


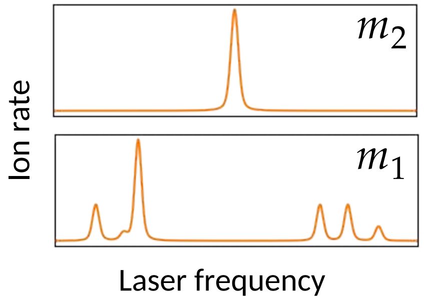
Courtesy of S. Berndt

RISIKO setup at Mainz









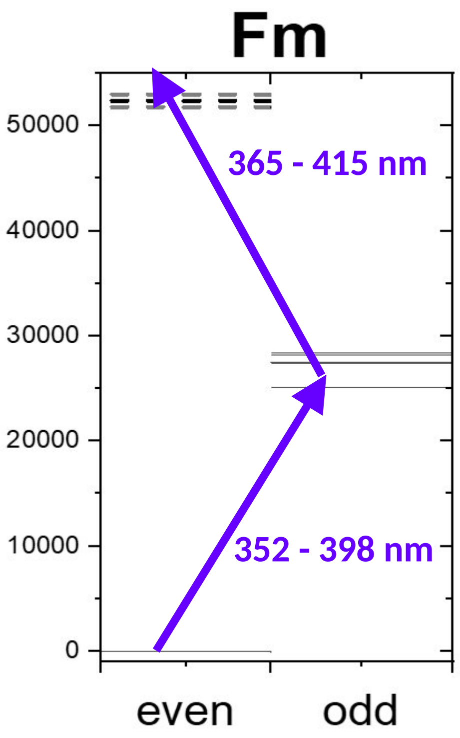
- Chemical sample preparation
- Sample evaporation in hot cavity
- Multi-step photoionization by pulsed lasers
- Mass separation in dipole magnet $\frac{m}{\Delta m} \approx 800$
- Single ion detection

RISIKO

Fermium ionization schemes

M. Sewtz et al., Phys. Rev. Lett. **90**, 163002 (2003)
H. Backe et al., Hyperfine Interact. **162**, 3 (2006)

2019, 2021, 2023 off-line measurements

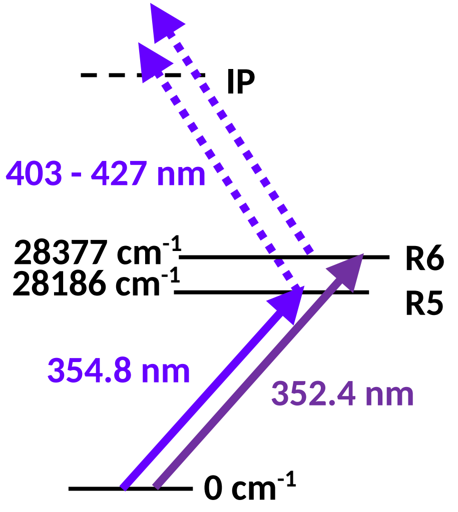


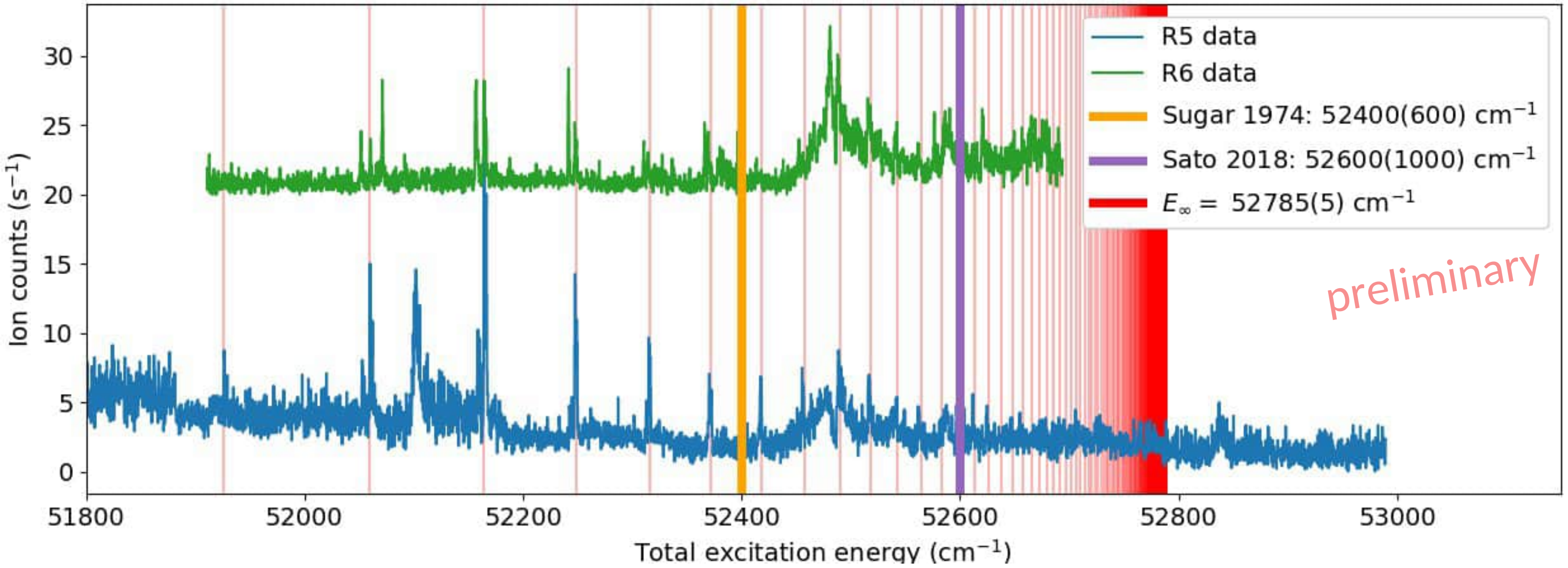
No.	Wavenumber (cm^{-1})	A_{ki} (Hz)	Observed (cm^{-1})	AI State (cm^{-1})	τ (ns)	Hyperfine A,B (MHz)	Ryd.
R1	25 099.8 (2)	3.4(8) x10^6	25 099.8	52101.4	440(20)	*A* = -310(10) *B* = -12800(100)	(✓)
R2	25 111.8 (2)	3.54(7) x10^6	25 111.8	52 071.6	460(20)	*A* = -10(10) *B* = -13500(100)	(✓)
R3	27 389 (1.5)		✗				
R4	27 466 (1.5)	≥ 2.9 x10^6	✗				
R5	28 185 (1.5)		28 186.9	52 164.4	< 50		✓
R6	28 377 (1.5)		28 379.8	52 481.0	< 50		✓
R7	28 391 (1.5)	≥ 1.1 x10^7	28 389.8		< 50		

preliminary

Rydberg convergences in fermium

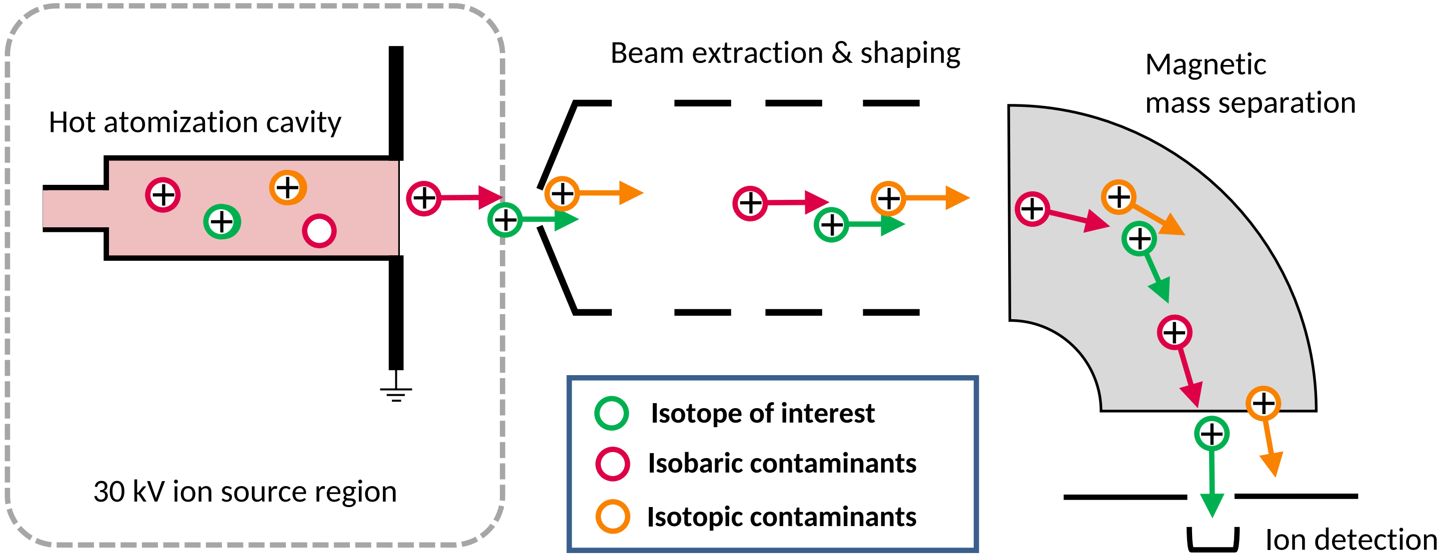
- Ionization potentials from literature have large uncertainties
  - Interpolation of actinide IPs: IP = 52400(600) cm^{-1} [1]
  - Surface ionization: IP = 52600(1000) cm^{-1} [2]
- Convergence limit of Rydberg series is precise, but configuration unclear
  - Collaboration with atomic theory (MCDHF, J. Andrews) to calculate first excited state in Fm II





[1] J. Sugar et al., J. Chem. Phys. **60**, 4103 (1974)
[2] T. K. Sato et al., J. Am. Chem. Soc. **140**, 14609 (2018)

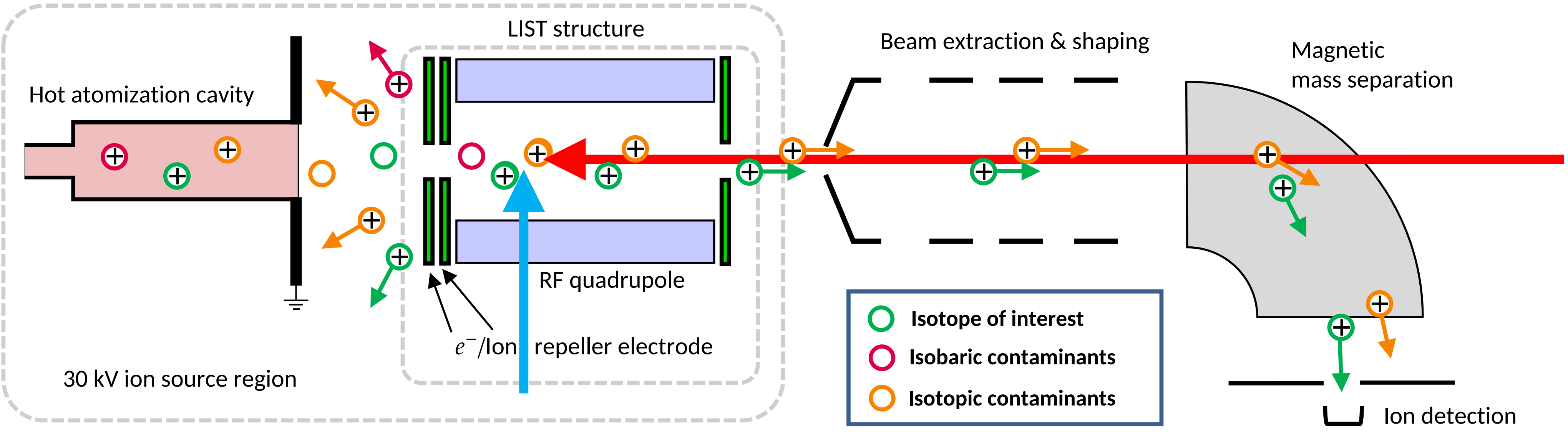
The PI-LIST ion source



Standard Laser Ion Source

- No selectivity against surface ions
- Spectral Doppler broadening from thermal velocity distribution (few GHz)
- High efficiency in the order of 10%

The PI-LIST ion source



Standard Laser Ion Source

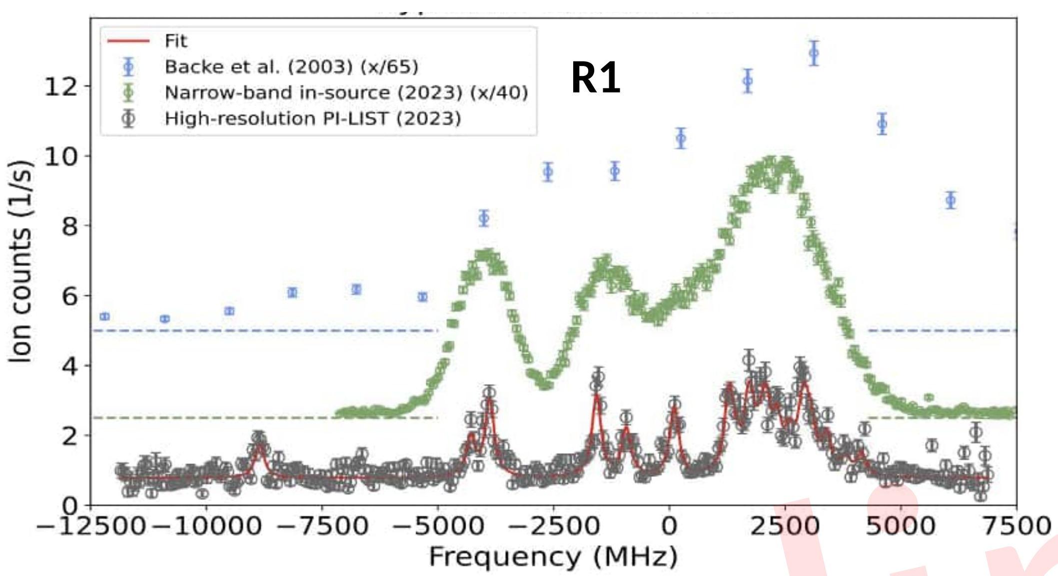
- No selectivity against surface ions
- Spectral Doppler broadening from thermal velocity distribution (few GHz)
- High efficiency in the order of 10%

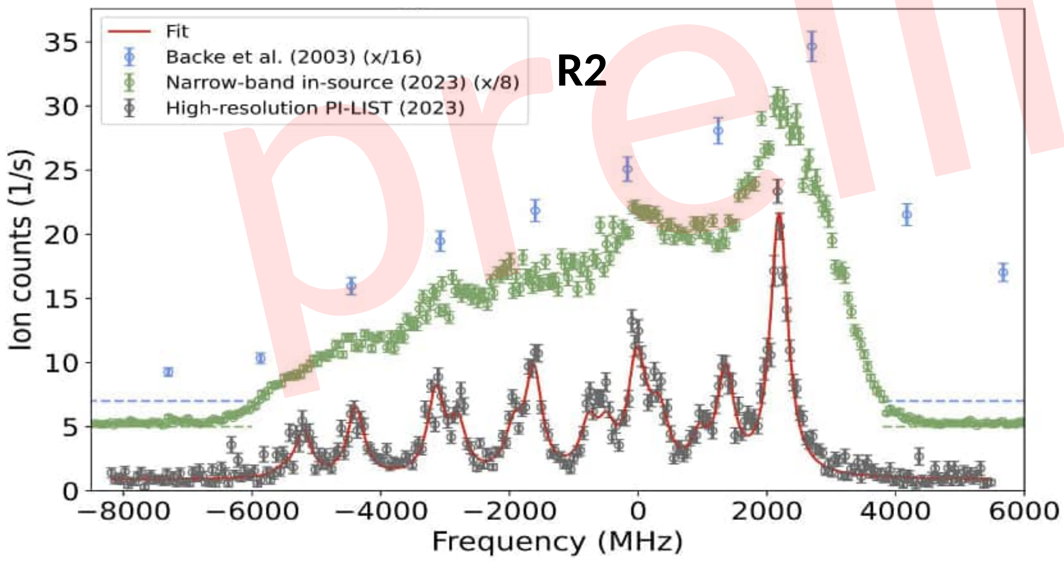
Perpendicularly Illuminated Laser Ion Source & Trap (PI-LIST)

- Suppression of surface ions, electrons
- Reduced Doppler broadening (<100 MHz)
- Loss factor 10^2 to 10^4 compared to Standard LIS

R. Heinke et al., Hyperfine Interact. **238**, 6 (2017)

Nuclear moments of Fm-255





Hyperfine coupling constants (ground state)
Experiment & atomic theory

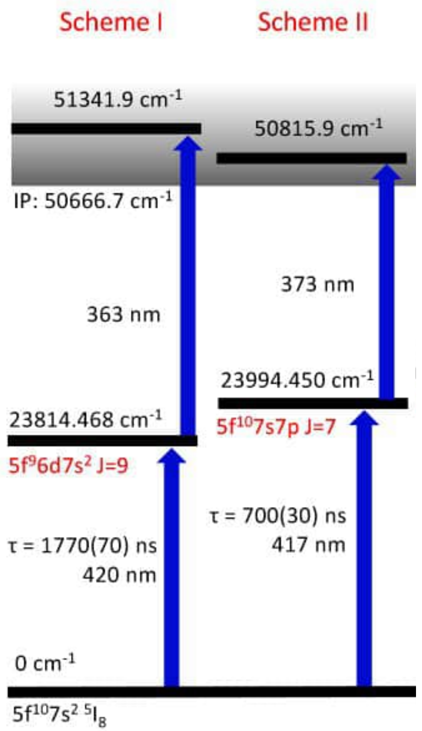
A (MHz)	*B* (MHz)	Method
-320	-22000	Experiment, Backe et al. 2000 [1]
-150	-10450	Experiment, this work
206/μ_N	-1782/Q	MCDHF, Beerwerth et al. 2019
187/μ_N	-1750/Q	CIPT, Allehabi et al. 2020 [2]
194/μ_N	-1789/Q	MCDHF, Andrews et al. 2025

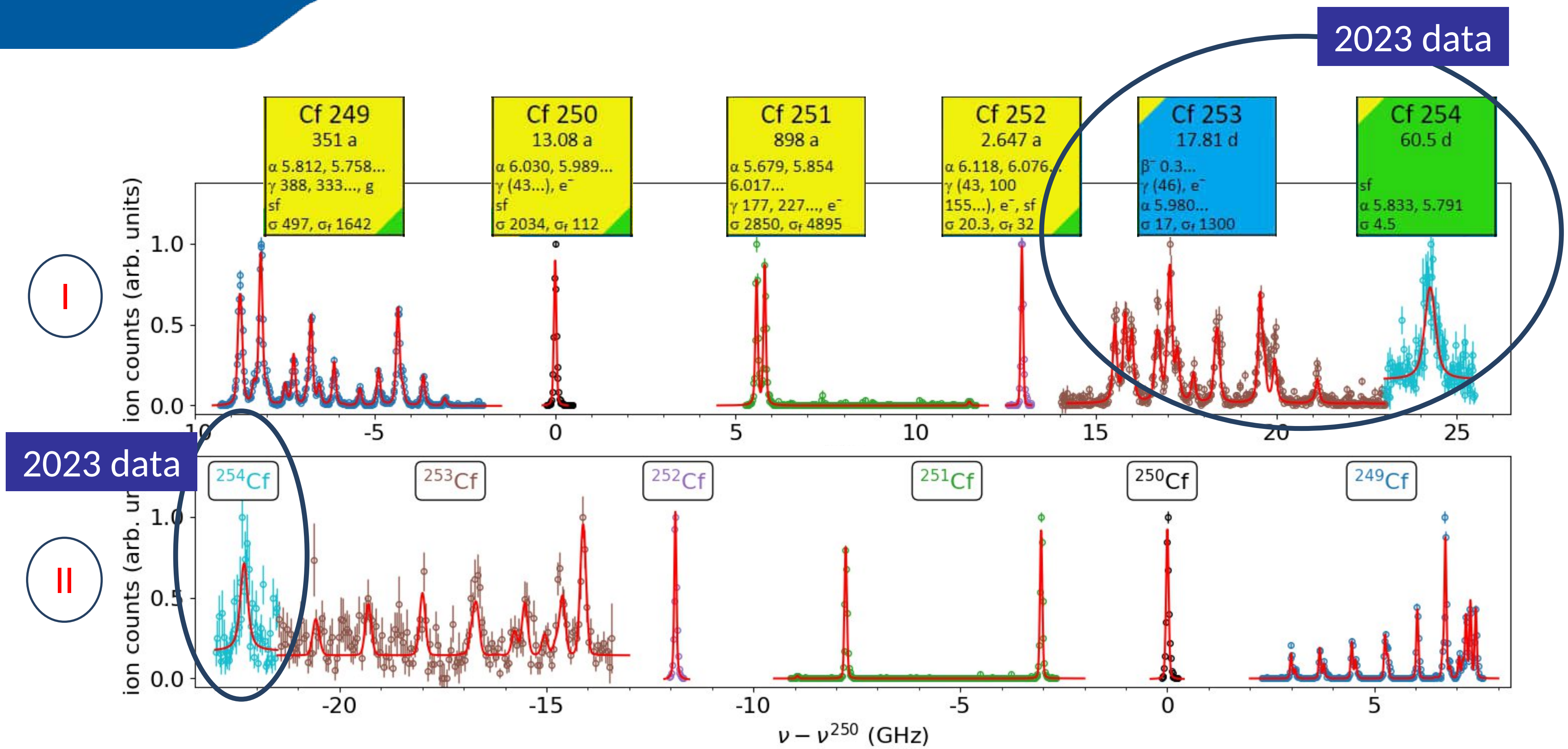
Nuclear moments
Nuclear theory & Experiment + atomic theory

μ (μ_N)	*Q* (*eb*)	Method
-0.77	+5.84	Exp. + MCDHF (this work)
-0.77	+5.78	HFB, Hilaire et al. 2025
-1.0	+6.2	DFT, Bally et al. 2025

[1] H. Backe et al., Hyperfine Interact. **162**, 3 (2006)
[2] S. O. Allehabi et al., JQSRT **253**, 107137 (2020)

Californium spectroscopy





Isotope	No. of atoms	Method
Cf-252	8.9(7)e12	Alpha
Cf-253	5.4(5)e9	RIMS; Ref. 252
Cf-254	<4(2)e7	RIMS; Ref. 252

F. Weber et al., Phys. Rev. C **107**, 034313 (2023)

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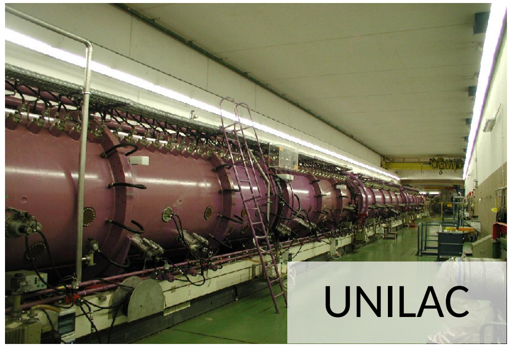
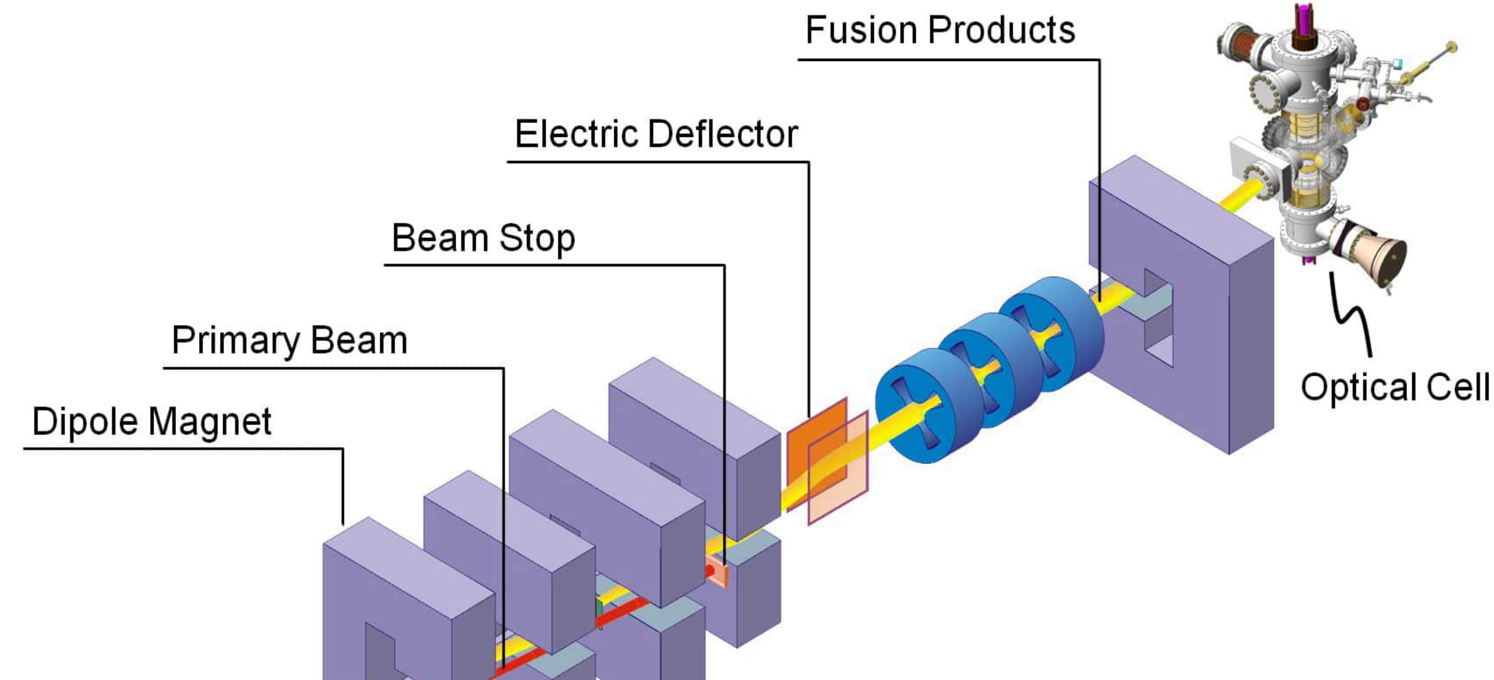
On-line laser spectroscopy at GSI

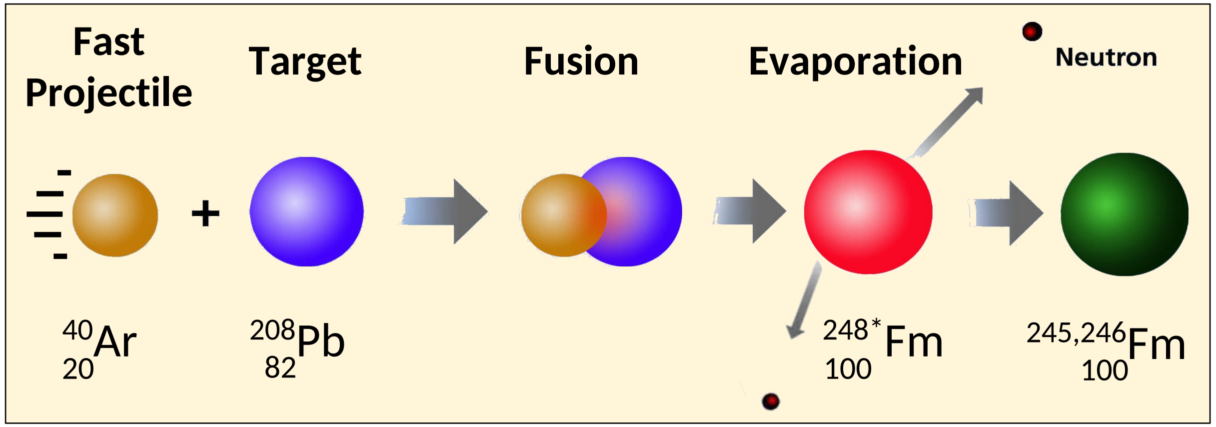
On-line production of Fm: Velocity Filter SHIP



SHIP target wheel

^{48}Ca
4.55 MeV/u
1 pμA

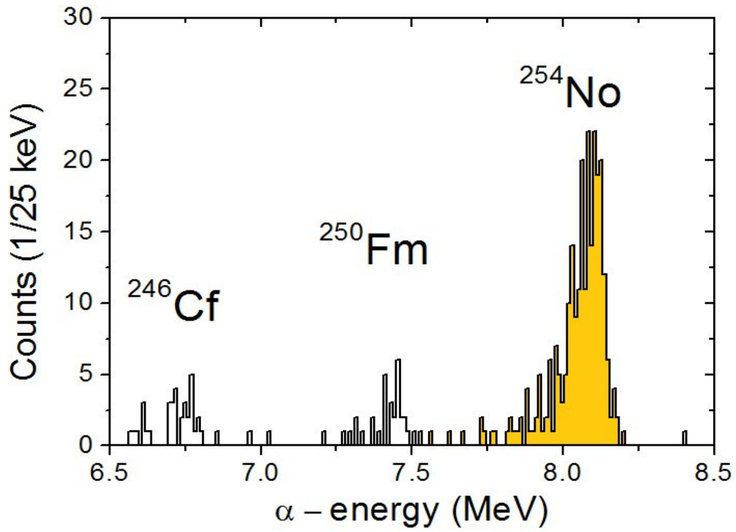


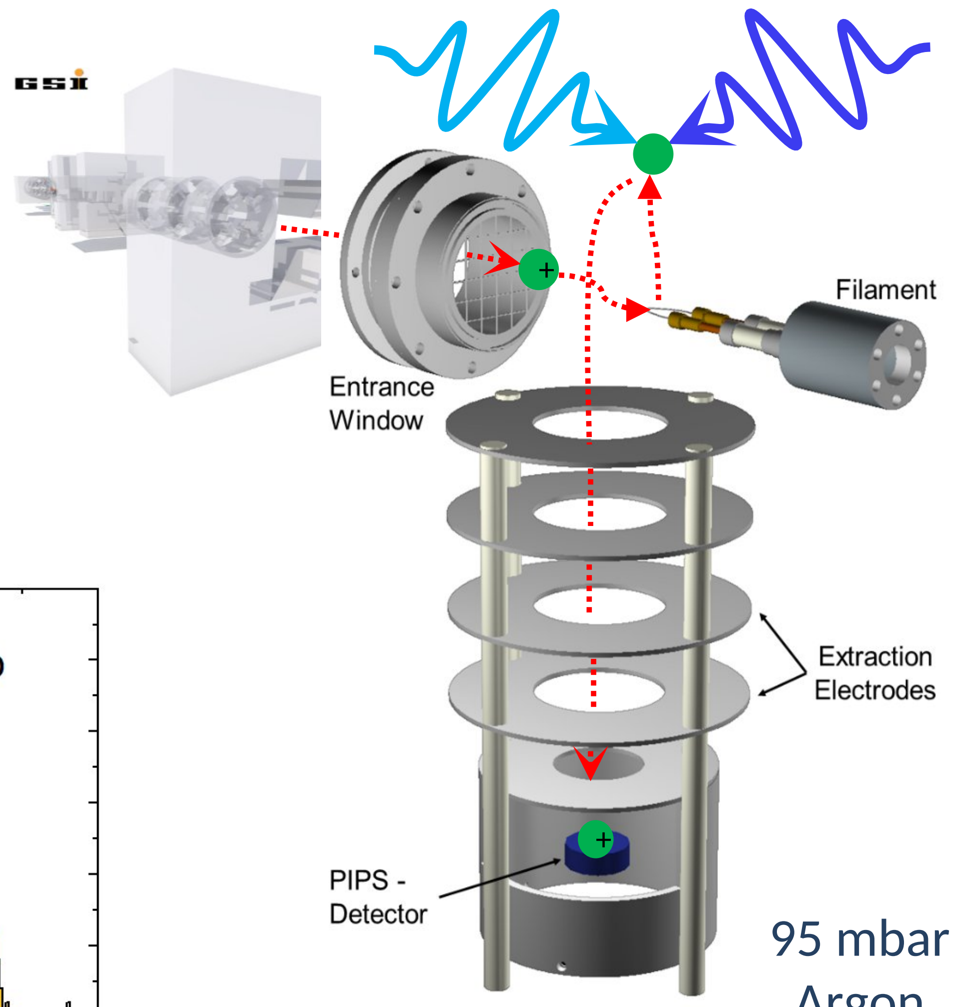


Radiation Detected Resonance Ionization Spectroscopy

RADRIS Method:

1) Thermalizing of incoming fusion products in Ar gas
2) Collecting ions onto thin hafnium filament
3) Evaporation
4) Two-step photoionization process with high power 100 Hz laser system
5) Transport to detector and detection of alpha decay



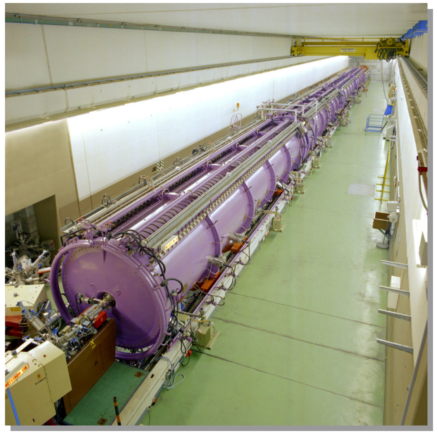


95 mbar Argon

H. Backe et al., Eur. Phys. J. D **45**, 99 (2007)
F. Lautenschläger et al., NIMB **383**, 115 (2016)
J. Warbinek et al., Atoms **10**, 41 (2022)

On-line production of fermium isotopes

GSI UNILAC linear accelerator



$^{208}Pb(^{40}Ar,2n)\,^{246}Fm$

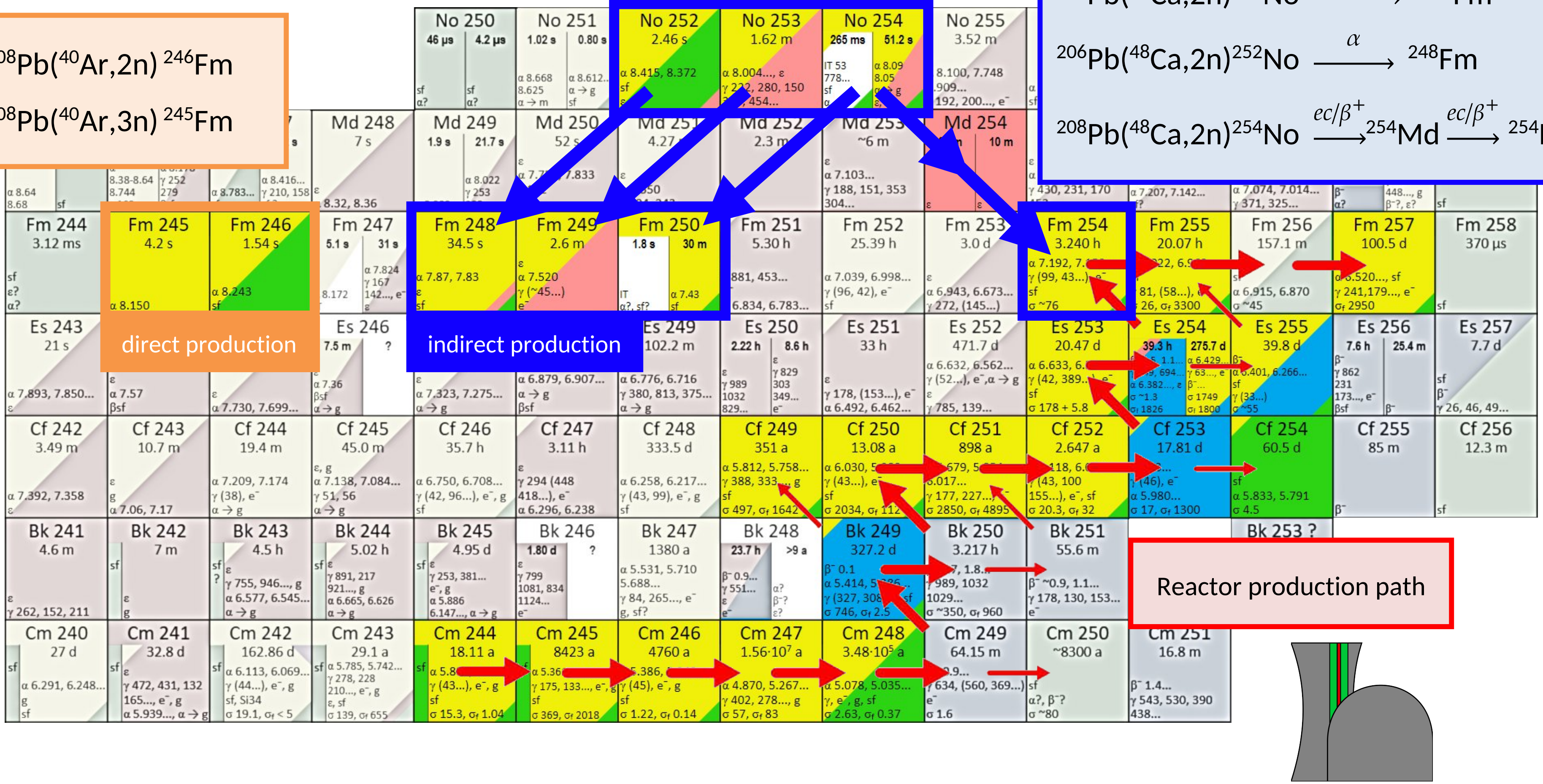
$^{208}Pb(^{40}Ar,3n)\,^{245}Fm$

$^{208}Pb(^{48}Ca,2n)^{254}No \xrightarrow{\alpha} {}^{250}Fm$

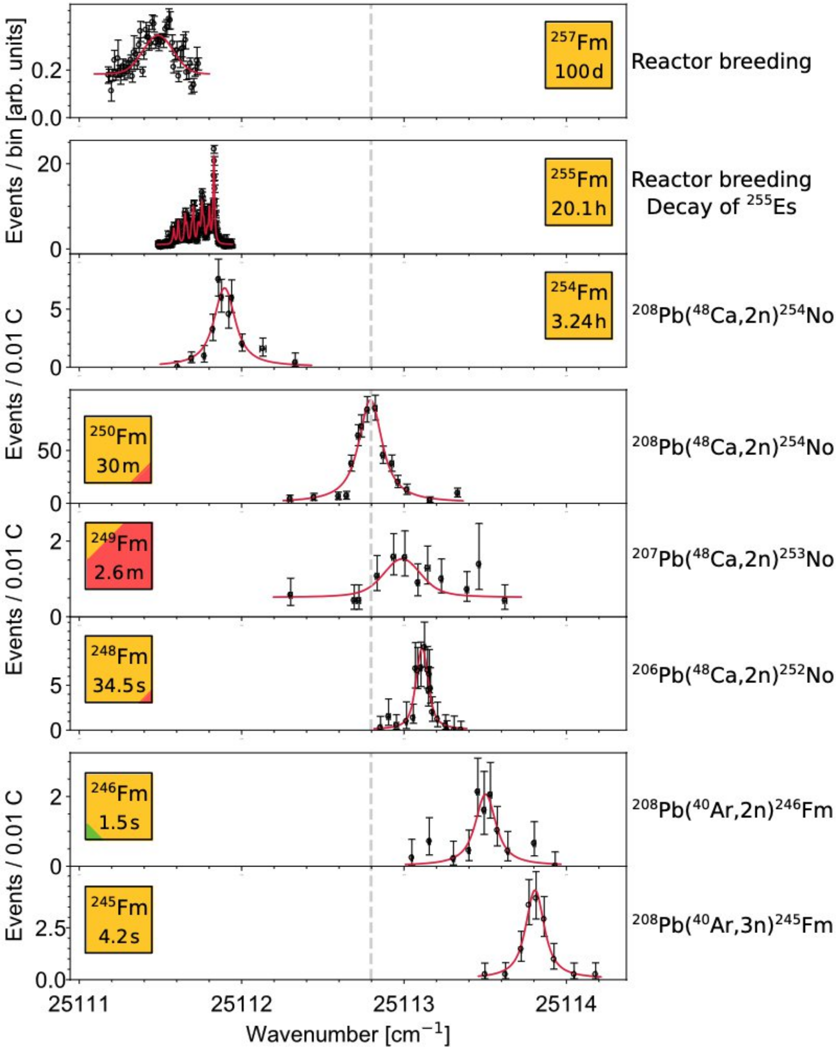
$^{207}Pb(^{48}Ca,2n)^{253}No \xrightarrow{\alpha} {}^{249}Fm$

$^{206}Pb(^{48}Ca,2n)^{252}No \xrightarrow{\alpha} {}^{248}Fm$

$^{208}Pb(^{48}Ca,2n)^{254}No \xrightarrow{ec/\beta^+} {}^{254}Md \xrightarrow{ec/\beta^+} {}^{254}Fm$



Relative charge radii across *N*=152



^{255,257}Fm

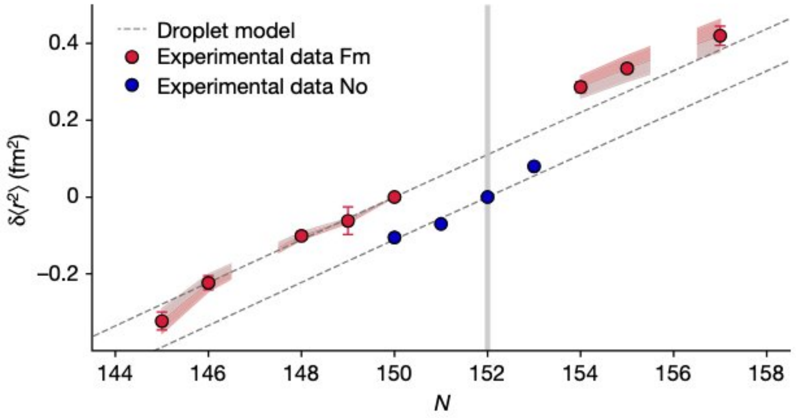
studied off-line at RISIKO

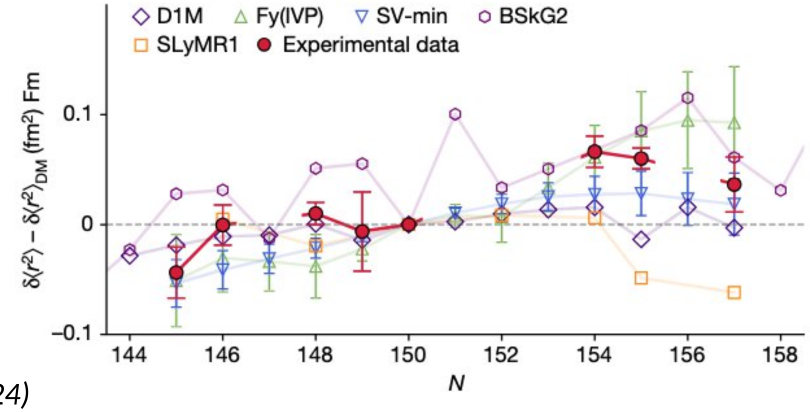
$^{245,246,248-250,254}$Fm

studied on-line at SHIP

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

Experiment Theory



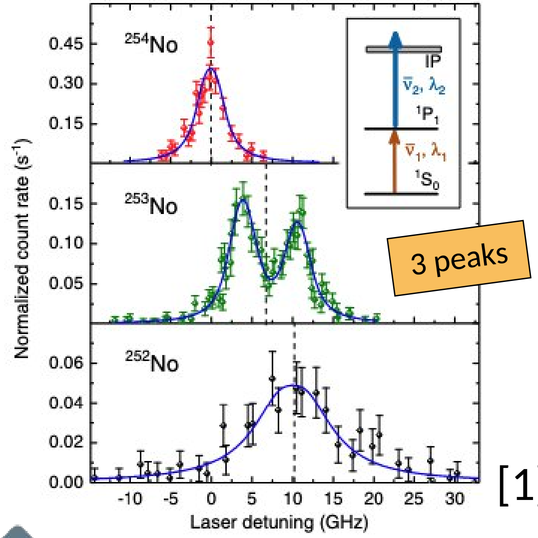


J. Warbinek, E. Rickert et al., Nature ***634****, 1075 (2024)*

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.

Limitations of the RADRIS method

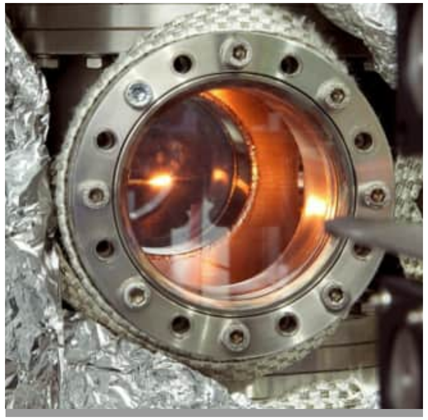
1) Spectral resolution due to Doppler and pressure broadening
   - ➢ Limited precision in isotope shift measurements
   - ➢ No determination of nuclear spins/moments



[1]

2) Measurement of long-half-life-isotopes (>hours) with alpha-detection requires long cycles (and is limited to alpha-emitters)

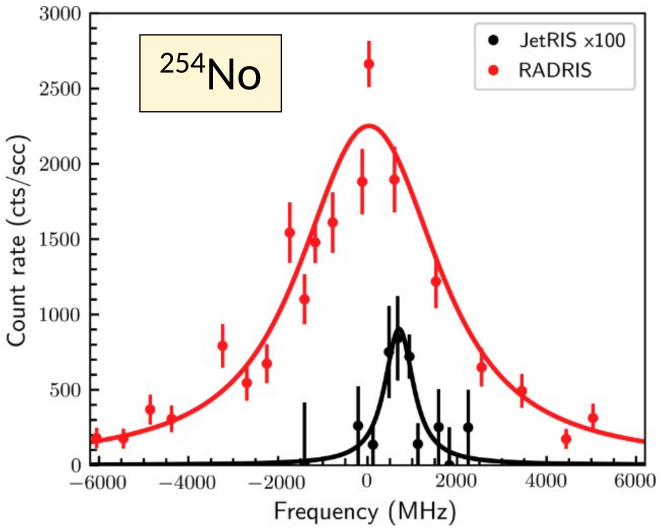
3) Desorption of some elements from hot filament is challenging (e.g. Lr)

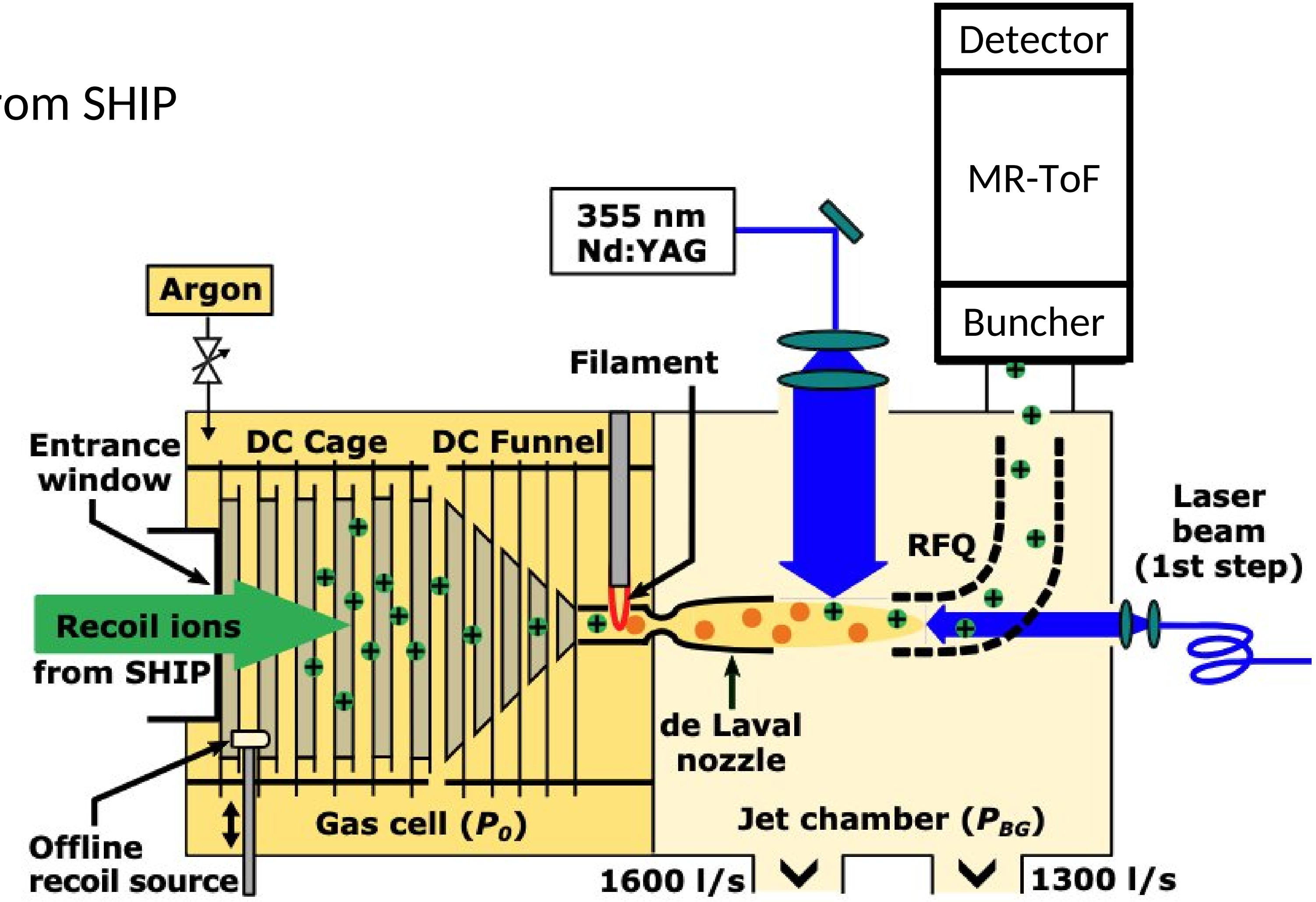


*[1] S. Raeder et al., PRL **120**, 232503 (2020)*

Working principle of JetRIS

1) Thermalization of the recoil ions from SHIP
2) Ion transport to the filament
3) Neutralization
4) Formation of a gas jet
5) 10 kHz RIS in the gas jet
6) Transport and detection



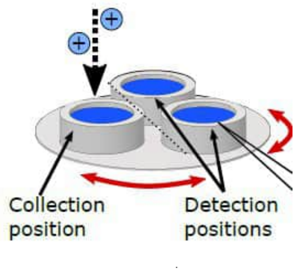


J. Lantis et. al., Phys. Rev. Res. **6**, 023318 (2024)

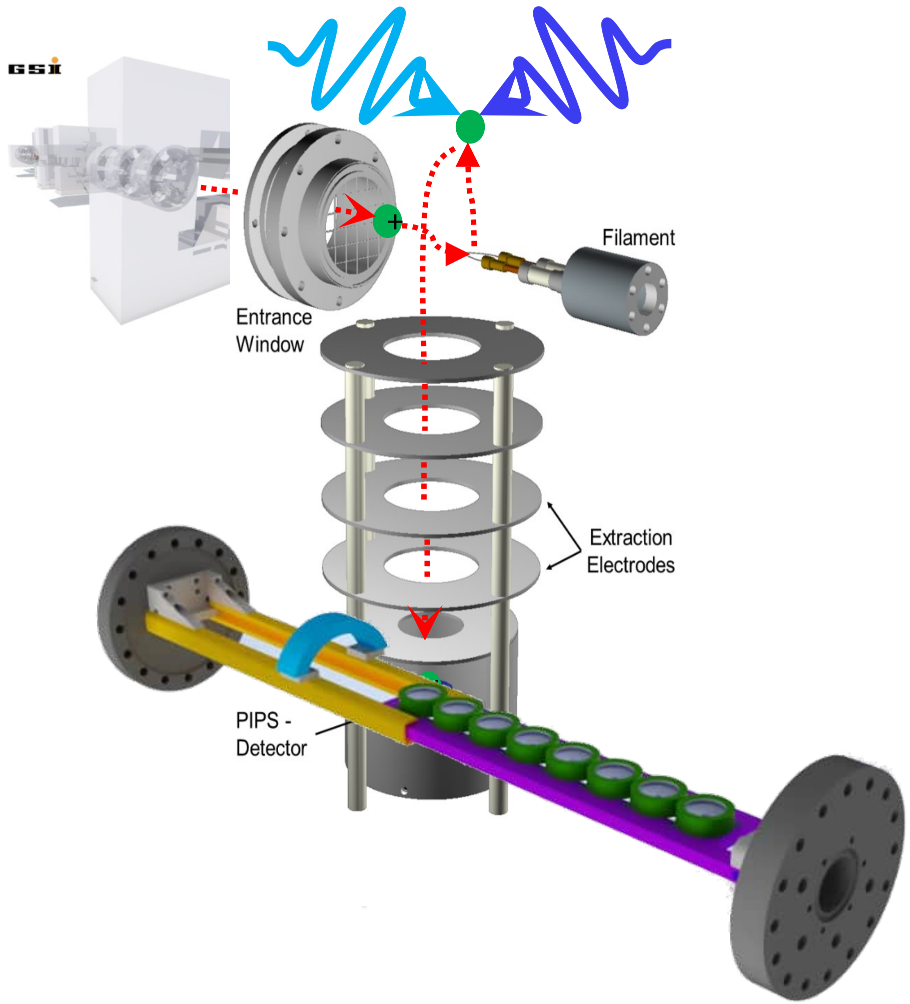
RADRIS 8-detector upgrade

- Measurement of long-lived species with RADRIS

- ^{254}Fm ($T_{1/2}$=3h) was measured with 3-fold detector

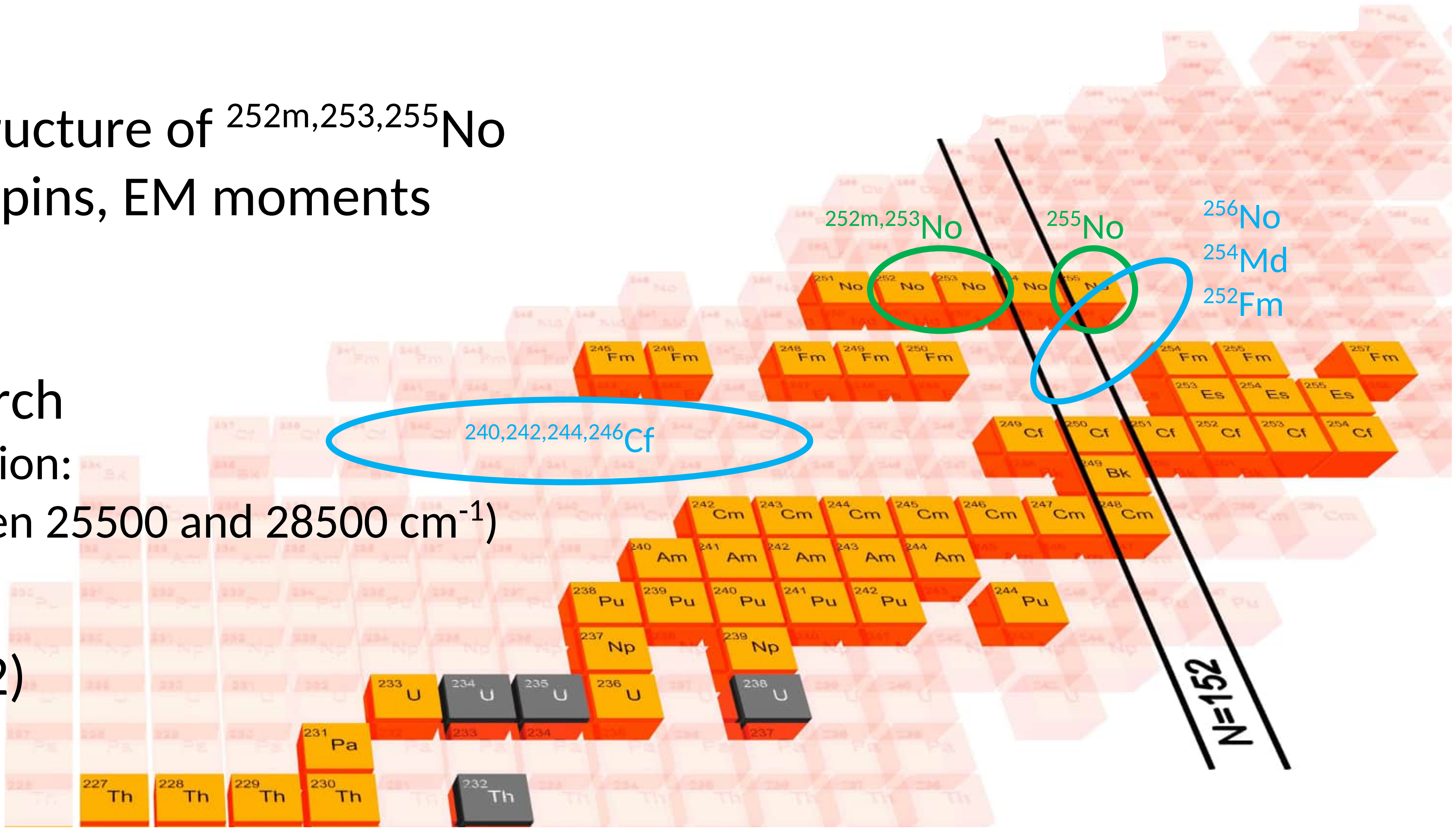


- ^{252}Fm ($T_{1/2}$=20h) to be measured with upgraded 8-fold detector design

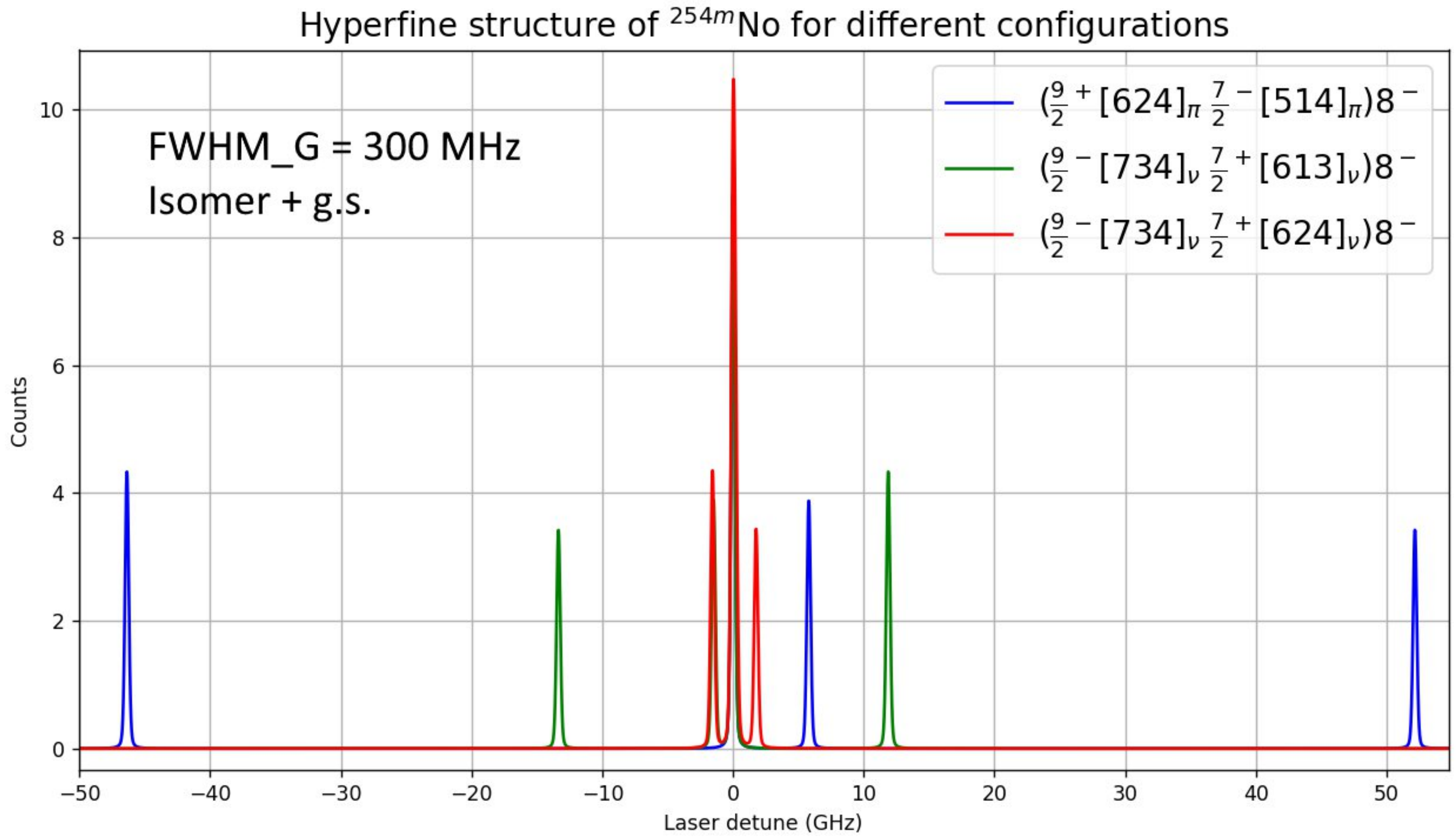


SHIP Beam Time 2025 and future plans

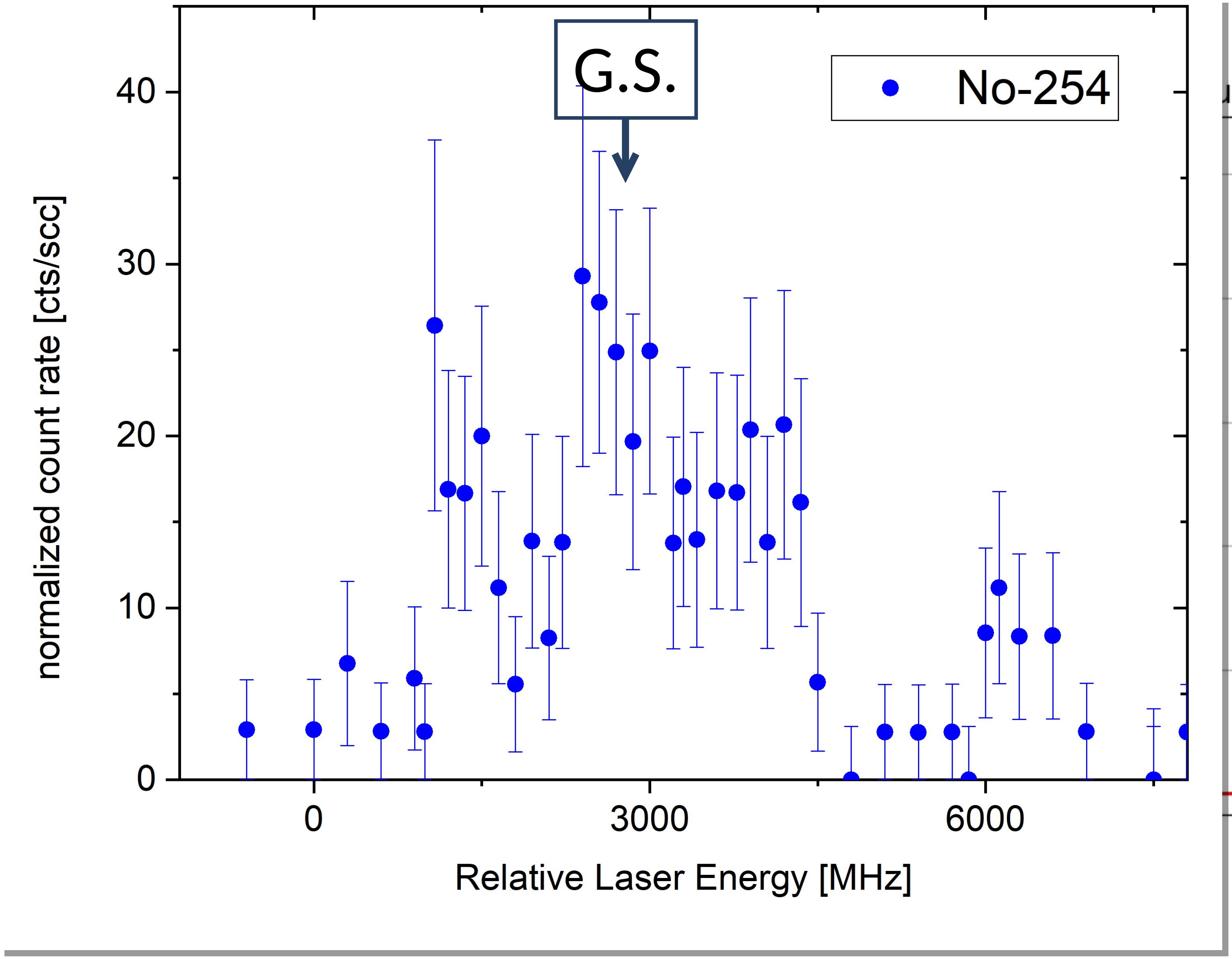
- JetRIS
  - Hyperfine structure of ^{252m,253,255}No
    —> nuclear spins, EM moments
- RADRIS
  - Md level search
    (theory prediction:
    5 levels between 25500 and 28500 cm^{-1})
  - ^{240,242,244,246}Cf
  - ^{252}Fm (N=152)
  - ^{256}No

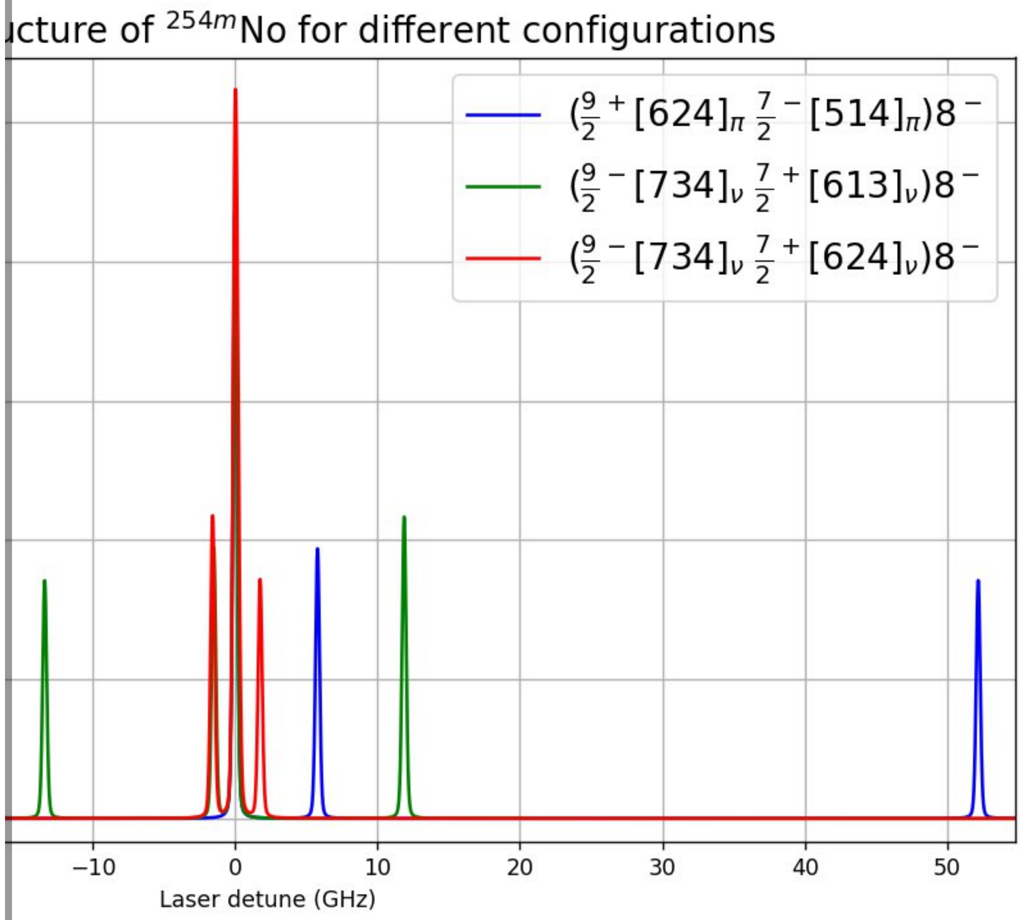


Hyperfine structure of No-254 and No-254m



Hyperfine structure of No-254 and No-254m





Summary

- Measurements of reactor-produced samples on the picogram to femtogram level are feasible!

- Laser spectroscopy of Fm
  - Confirmed previously reported ground-state transitions
  - Hyperfine structures using the PI-LIST technique
  - Rydberg convergences for IP determination
  - Isotope shifts of 8 Fm isotopes across the *N*=152 shell gap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Smooth trends in fermium charge radii and the impact of shell effects
Jessica Warbinek, Elisabeth Rickert, Sebastian Raeder, Thomas Albrecht-Schönzart, Brankica Andelic, Julian Auler, Benjamin Bally, Michael Bender, Sebastian Berndt, Michael Block, Alexandre Brizard, Pierre Chauveau, Bradley Cheal, Premaditya Chhetri, Arno Claessens, Antoine de Roubin, Charlie Devlin, Holger Dorrer, Christoph E. Düllmann, Julie Ezold, Rafael Ferrer, Vadim Gadelshin, Alyssa Gaiser, Francesca Giacoppo, ... Klaus Wendt + Show authors
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- The N=152 shell gap is not apparent in nuclear charge radii, confirming the collective nature of the strongly deformed heavy actinides

The fermium collaboration

Universität Mainz
Julian Auler
Sebastian Berndt
Holger Dorrer
Christoph E. Düllmann
Vadim Gadelshin
Raphael Hasse
Magdalena A. Kaja
Nina Kneip
Mustapha Laatiaoui
Andrea T. Loria Basto
Christoph Mokry
Thorben Niemeyer
Dennis Renisch
Jörg Runke
Matou Stemmler
Petra Thörle
Norbert Trautmann
Felix Weber
Klaus Wendt

GSI Darmstadt
Michael Block
Francesca Giacoppo
Manuel Gutiérrez-Torres
Sebastian Raeder
Kenneth van Beek
Jessica Warbinek

ILL Grenoble
Ulli Köster

FSU Tallahassee
Thomas Albrecht-Schönzart
Alyssa Gaiser
Joseph Sperling

HÜBNER Photonics
Korbinian Hens
Volker Sonnenschein
Mitzi Urquiza-González

HIM Mainz
Premaditya Chhetri
Tom Kieck
Jeremy Lantis
Danny Münzberg
Steven Nothhelfer
Elisabeth Rickert
Dominik Studer

GANIL
Alexandre Brizard
Nathalie Lecesne

University of Gothenburg
Dag Hanstorp

Oak Ridge National Laboratory
Julie Ezold
Ashley Harvey
Kristian Mhyre
Samantha Schrell
Shelley Van Cleve

TU Darmstadt
Thomas Walther

Nagoya University
Hideki Tomita

CERN
Reinhard Heinke

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Universität Mainz
Mechanical workshop at TRIGA Mainz
R. Jera, Glassblower at JGU Chemistry
Radiation protection staff at TRIGA
S. Karpuk

ILL Grenoble
ILL reactor team and health physics

Oak Ridge National Laboratory
Nate Sims, Radioisotope Laboratory Technician
Nonreactor Nuclear Facilities Division Hot Cell Staff

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Thank you for your attention