

The Periodic Table of Elements

Elements at the limits of nuclear stability

- Why do SHE exist at all ?
- What is nuclear structure: what is their shape and size ?
- How are they best produced in the lab?
- How do their atomic and chemical properties compare to known (lighter) elements ?



Superheavy Elements = transactinides



Chart of Nuclides



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Superheavy Elements – Present Status



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Island of Stability – Status 2023



Properties of the heaviest elements

Laser spectroscopy for fundamental properties

- Understanding of atomic structure & chemical behaviour •
- What is nuclear structure: what is their shape and size? •



La	⁵⁸ Ce	⁵⁹ Pr	Nd	Pm	Sm	Eu	Gd	⁵⁵ Tb	⁶⁶ Dy	Ho	Er	[®] Tm	Yb	Lu
Åc	^⁰ Th	Pa	⁹² U	⁹³ Np	P4 Pu	⁹⁵ Am	[%] Cm	⁹⁷ Bk	⁹⁸ Cf	[»] Es	Fm	Md	¹⁰² No	Lr

Electron shell

atomic structure chemical properties \rightarrow defines the element



Nucleus

nuclear structure stability of elements

Atomic structure is linked to light



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Overview on atomic levels reported for the heavy actinides



Block, M. et al, Progress in Particle and Nuclear Physics (2020): 103834.

S. Raeder – 29.05.2024 – ISOL France Workshop Strasbourg

• Atomic structure

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- Sparse for heavier element
- For Z ≥ 100 only calculations are available

Blaise 1992Experimental levelsTheory – calculated levels

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⁹⁹Es – analysis of fluorescence light



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- magnetic moment of ²⁵³Es from HFS
- too little material for Zeemann splitting

[Wor74] Worden E.F., et al. "Hyperfine structure in the 253Es emission spectrum, III: Extension of the line list, levels of Es i and Es ii, nuclear magnetic-dipole and quadrupole moments* " *J.Opt. Soc.Am.* 64.1 (1974): 77.

¹⁰⁰Fm – only possible with laser spectroscopy

2003: First atomic information on Fm Scan Regior Counts Mainz: Institut für Kernphysik, Institut für Kernchemie 25000 26000 Channeltron breeding of ²⁵⁵Es **Buffer Gas Cell** Detector Ion Optics 35 mbar Ar at Oak Ridge, USA Nozzle Segmented OPIG QMS 27395 v [cm⁻¹] $\Phi_{\rm n} = 2.6 \cdot 10^{15} / {\rm cm}^2 \cdot {\rm s}$ ²⁵⁵Fm T = 1a90° Filament Deflector 8.10⁻³ mbar 2.10-4 mbar 2.10-5 mba 2.10-6 mbar 4 ng 255 Fm (t_{1/2}= 20 h) TMP TMP TMP 0 5cm Laser (10^{12} atoms) 700 l/s 360 l/s 330 l/s 230 l/s Beams

Scan Region Scan

λ [nm] 354 353

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7 atomic transitions



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Laser system: 100 Hz, Excimer pumped Dye laser + 50 Hz OPO

[Sew03] Sewtz, M., et al. "First observation of atomic levels for the element fermium (Z= 100)." *Phys. Rev. Lett.* 90.16 (2003): 163002. [Bac06] Backe, H., et al. "Laser spectroscopic investigation of the element fermium (Z= 100)." Laser 2004. (2006). 3-14.

Resonance Ionization Spectroscopy



- Resonant laser excitation selective & efficient
- Efficient excitation schemes generally exist for every element

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• Atomic spectra incomplete – development often necessary

Motivation - Atomic Structure



- $Z\alpha \rightarrow 1$: large QED contribution
 - & relativistic effects

in the electronic structure

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 $6s_{1/2}$ $7s_{1/2}$ 8s1/2 118 6**p**3/2 5p3/7 $5p_{1/2}$ $7p_{1/2}$ 7s $5s_{1/2}$ $6s_{1/2}$ (s_{1/2} $5d_{5/2}$ $4d_{5/2}$ NR R NR SR R R

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

6 (MCDF): P.Indelicato et al., Phys. Rev. A 90 (2014) 012504 Eur. Phys. J. D 45 (2007) 155 5 (MCDF): Y.Liu et al., 3 (IHFSCC): A.Borschevsky et al., 7 (extrapolation): J.Sugar, Phys. Rev. A 76 (2007) 062503 J. Chem. Phys. 60 (1974) 4103 Phys. Rev. A 75 (2007) 042514



Motivation - Atomic Structure



- Zα → 1: large QED contribution & relativistic effects in the electronic structure
- Strong electron correlations
- Benchmark predictive power of atomic theory
- Ionization potential (IP) \rightarrow chemical properties
- Determination of nuclear properties

 Model
 1, 2 (MCDF): S.Fritzsche, Eur. Phys. J. D 33 (2005) 15

 calculations
 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



Motivation – Nuclear Properties



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Nuclear Information from Laser Spectroscopy



Orbital energy changes with deformation – Nilsson model

Hyperfine structure:

• Deformation (Q, β_2) from HFS



• Magnetic moments $\mu = g \cdot I$

Provides g-factors of unpaired nucleon characteristic for the wavefuctions

• Assignment of nuclear Spin I

Charge radii:

Change in deformation



Optical Investigation of Heavy Elements

Laser spectroscopy for fundamental properties

- Understanding of atomic structure & chemical behaviour
- Nuclear structure: what is their shape and size ?



Production of SHE at GSI



Production through fusion-evaporation reactions



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Production of SHE at GSI



Production of SHE at GSI



Superheavy Experiments at GSI



Production: Velocity Filter SHIP



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Radiation Detected Resonance Ionization Spectroscopy

RADRIS Method:

- Thermalizing of incoming fusion products
- Collecting onto thin tantalum wire
- Evaporation and two-step photoionization process
- Transport to detector and detection of alpha decay
- High power 100 Hz Laser system

H. Backe et al., Eur. Phys. J. D 45, 99 (2007) F. Lautenschläger et al., NIMB 383, 115 (2016)





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Level Search in ²⁵⁴No



Nobelium: The Ground-State Transition

Observed strong atomic ground state transition

- **Resolution 5 GHz**
- A total efficiency of 6.4(10) % for ²⁵⁴No
- Less than 30 000 atoms were delivered to the cell
- Saturates at low photon fluxes

[1] M. Laatiaoui et al., Nature 538 (2016) 7626

[3] P. Indelicato et al., Eur. Phys. J. D 45, (2007) 155

	v ₁ (cm ⁻¹)	A _{ki} (s ⁻¹) x 10 ⁸
Experiment [1]	29,961.457(7) _{stat}	4.2 (2.6) _{stat}
IHFSCC [2]	30,100(800)	5.0
MCDF [3]	30,650(800)	2.7

Agrees with predicted ${}^{1}S_{0} \rightarrow {}^{1}P_{1}$ transition



Isotope Shift of ²⁵²⁻²⁵⁴No & HFS in ^{253,255}No



- Isotope shift for ²⁵²⁻²⁵⁴No measured
- Change in charge radii: Input from atomic theory
 - Mass-shift constant: 1044 GHz u
 - Field-shift parameter: -95.8(7.0) GHz/fm

(R. Beerwerth & S. Fritzsche (MCDF), V. Dzuba, M. Safranove (CI), A. Borschevsky (RCC))



experiment

Note: requires high precision atomic calculations !



Isotope Shift of ²⁵²⁻²⁵⁴No & HFS in ^{253,255}No



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On-line Laser Spectroscopy of Fm Isotopes



Accessible Fm isotopes

More Fm isotopes in reach from different No isotopes



Es – Fm Sample available from Oak Ridge



2019: contact from Florida: Fm sample available

"waste" in the ²⁵²Cf production process



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Radiochemical separation required \rightarrow JGU - nuclear chemistry



Hot Cavity Resonance Ionization Spectroscopy



- Used for production of radioactive ion beams
- Laser spectroscopy with high efficiency
- Background from surface ionization
- Resolution limited by

source temperature and laser bandwidth

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lasers

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magnet

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10 kHz Laser system

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Ti:sapphire laser system



Three laser types for different applications

	Standard	Grating	Injection-locked		
Repetition rate		10 kHz			
Pulse width	40 to 60 ns				
Avg. Power	3 – 5 W	1 – 2 W	2-3 W		
Output range	700 – 1	1020 nm	seed source		
Tuning range	100 GHz	700 – 1020 nm	typ. 20 GHz		
Spectral bandwidth	1 to 10 GHz	1 to 3 GHz	20 MHz		
Application	Laser ion source	BB spectroscopy	HR spectroscopy		

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Extension of output range by SHG, frequency mixing possible



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Sample Analysis with Laser Ionization

Lasers tuned to resonantly ionize different actinide elements



Stated sample composition

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.

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.



Es Laser Ionization Spectroscopy

Idendification of ionization schemes: Investigation of atomic struture in Einsteinium



Characterization of 10 Levels using only 10¹⁰ atoms (10 pg), revised ionization potential

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. *F. Weber et al., Phys. Rev. Res. 4, 043053 (2022)*

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Hyperfine Structure of Es Isotopes



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Nuclear Properties of Cf Isotopes

PI-LIST ensures higher resolution

Cf samples as low as 5•10⁷ atoms !

Hyperfine structure measurements

- Fixed ionization scheme
- Nuclear moments deduced

\rightarrow input from atomic theory

(V. Dzuba et al., CIPT)

Isotope	Ι	$\mu_I \ (\mu_{ m N})$	$\mu_{I,\mathrm{HFB}} \ (\mu_{\mathrm{N}})$	$Q_{ m S}\ (e{ m b})$	$Q_{ m S,HFB}\ (e{ m b})$
²⁴⁹ Cf	9/2	$-0.395(18)^{\dagger}$	-0.731	6.27(31)	6.86
^{251}Cf ^{253}Cf	$\frac{1/2}{7/2}$	-0.571(26) -0.731(33)	-1.015 -0.755	5.53(28)	5.40

[†] Literature value: 0.28(6) [2]



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[1] F. Weber et al., Phys. Rev. C 107, 034313 (2023) [2] N. Edelstein, D. G. Karraker, J. Chem. Phys. 62, 938 (1975) S. Raeder – 29.05.2024 – ISOL France Workshop Strasbourg

Deformation of the Actinides



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N.J. Stone , IAEA INDC(NDS)-0594 (2015)

Deformation of the Actinides



N.J. Stone - B. Pritychenko et al , Atom. Dat.Nucl.Dat.Tab.107, 1-139 (2016) (2015)

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What about Fermium?

• Preparation: Chemical separation to increase the fermium content





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.

\rightarrow first laser spectroscopy of ²⁵⁷Fm

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.

[Sew03] Sewtz, M., et al. Phys. Rev. Lett. 90.16 (2003): 163002. -- [Bac06] Backe, H., et al. " Laser 2004. (2006). 3-14.



Laser spectroscopy of Fm-257

• Preparation: Chemical separation to increase the fermium content



→ first laser spectroscopy of $^{257}\mathrm{Fm}$

despite a much lower sample size then expected



[Sew03] Sewtz, M., et al. Phys. Rev. Lett. 90.16 (2003): 163002. -- [Bac06] Backe, H., et al. "Laser 2004. (2006). 3-14.

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5x10⁷ atoms ²⁵⁷Fm

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Fm-255 – boostered sample from ILL



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Fm isotopic chain – change in rms charge radii



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RADRIS – Perspectives

Achievements in Laser spectroscopy:

- ✓ a sensitive & versatile tool for investigating atomic & nuclear properties
- ✓ Many atomic levels observed in ²⁵⁴No
- ✓ Differential mean square charge radii extracted for ²⁵²⁻²⁵⁵No & ^{248-250,254,255,257}Fm
- Complementary off-line laser spectroscopy
- ✓ Exotic Cf isotopes accessible online
- Limitations of the RADRIS technique
 - Collection cycle limits short lifetimes
 - Radiation detection difficult for long-lived nuclides
 - Gas environment limits the spectral resolution



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Gas jet – High Resolution & High Efficiency



• Low temperature & low density supersonic gas jets are ideal

environments for laser spectroscopy experiments of (trans)actinides



[1] R. Ferrer, et al., Nature Commun., 8, 2017.

In-gas-jet laser spectroscopy on ²⁵⁴No at GSI

Combination of high-efficiency RADRIS with high resolution in-jet methods



Beamtime 2022

• First in-gas-jet laser spectroscopy on ²⁵⁴No with improved resolution

S. Raeder et al., NIM B 463 (2020)(2019) 272 M. Laatiaoui et al., Nature (London) 538, 495 (2016),

J. Lantis et al, Pys Rev Rese. Accepted



In-gas-jet laser spectroscopy on ^{253,254m,255}No at GSI

Combination of high-efficiency RADRIS with high resolution in-jet methods



 In-gas-jet laser spectroscopy on ^{253,255}No will enable improved precision in nuclear deformation to assign Nilsson orbital→ validation nuclear models

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

 S. Raeder et al., NIM B 463 (2020)(2019) 272
 R.-D. Herzberg, et al., Nature, 442(7105) 896, 2006



In-gas-jet laser spectroscopy on ^{253,254m,255}No at GSI

Combination of high-efficiency RADRIS with high resolution in-jet methods



• For ²⁵⁴No in-jet-spectroscopy will enable to study the nature of the low lying 8⁻ *K*-isomer

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

 S. Raeder et al., NIM B 463 (2020)(2019) 272
 R.-D. Herzberg, et al., Nature, 442(7105) 896, 2006



Summary

Laser spectroscopy on heavy elements is important for the understanding of atomic/chemical character and for investigations of the nuclei

- RADRIS: First optical transistion in a transfermium element
 - ✓ Differential mean square charge radii extracted (²⁵²⁻²⁵⁴No)
 - ✓ First on-line measurements of Fm isotopes
- Insoure laser spectroscopy of actinides
 - ✓ Laser spectroscopy with Es (10¹⁰ atoms) and ^{255,257}Fm (10⁷ atoms)

Competitive laser spectroscopy program will continue

- RADRIS & gas jet beamtime running
- ✤ Gas jet: development for improved resolution
- LRC: new approach to extend the realm of laser spectroscopy



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