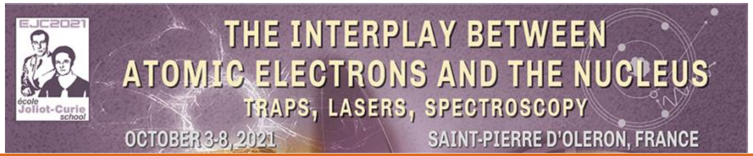
Physics and chemistry of the heaviest elements

S. Raeder

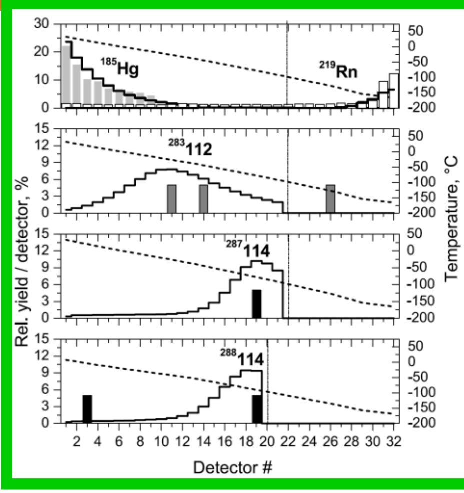
GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany

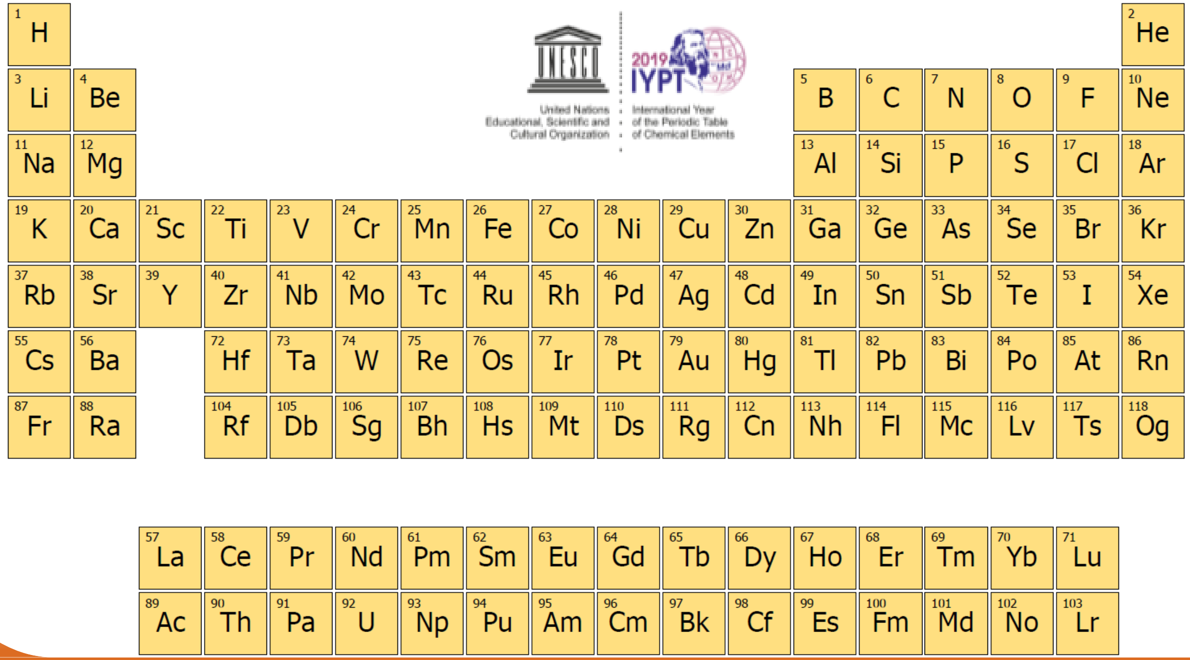


Heaviest Elements

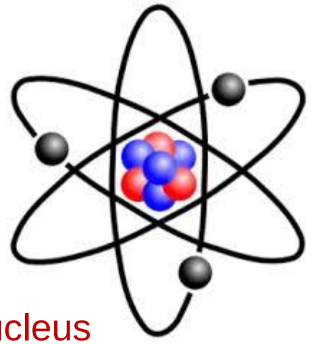
Elements at the limits of nuclear stability

- Why do SHE exist at all ? → **Shell effects**
- How are they best produced in the lab? → **For now: Fusion-evaporation**
- What is nuclear structure: binding energies, excitations, shape and sizes
- How do their atomic and chemical properties compare to known (lighter) elements?

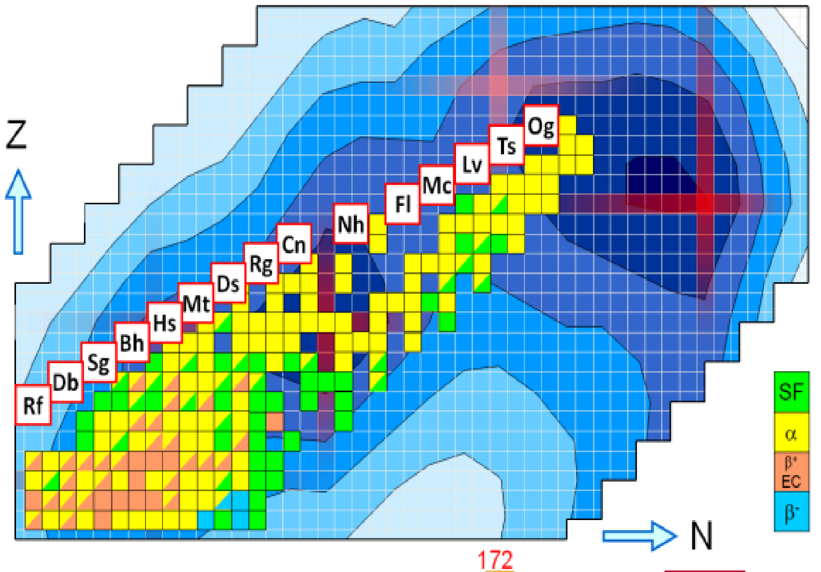




Electron shell
atomic structure
chemical properties
→ defines the element



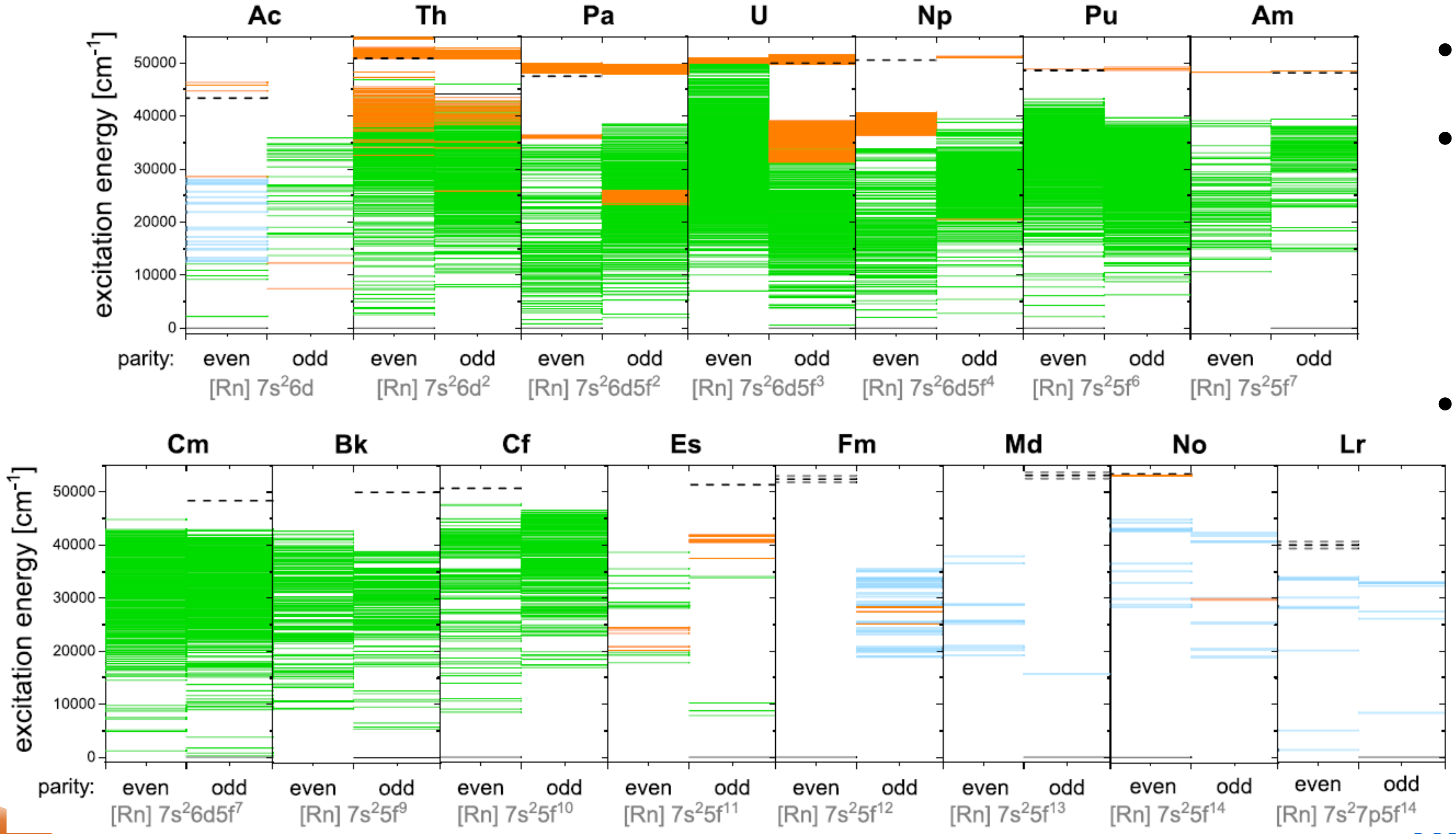
Nucleus
nuclear structure
stability of elements



HIM HELMHOLTZ Helmholtz-Institut Mainz
GSI RESEARCH FOR GRAND CHALLENGES
JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Density of Atomic Levels in the Actinides

Overview on atomic levels reported for the heavy actinides



- Atomic structure
- Sparse for heavier element

  (remember production)

- For Z ≥ 100 only calculations are available

Blaise 1992

Experimental levels

Theory – calculated levels

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

HIM HELMHOLTZ Helmholtz-Institut Mainz

GSI RESEARCH FOR GRAND CHALLENGES

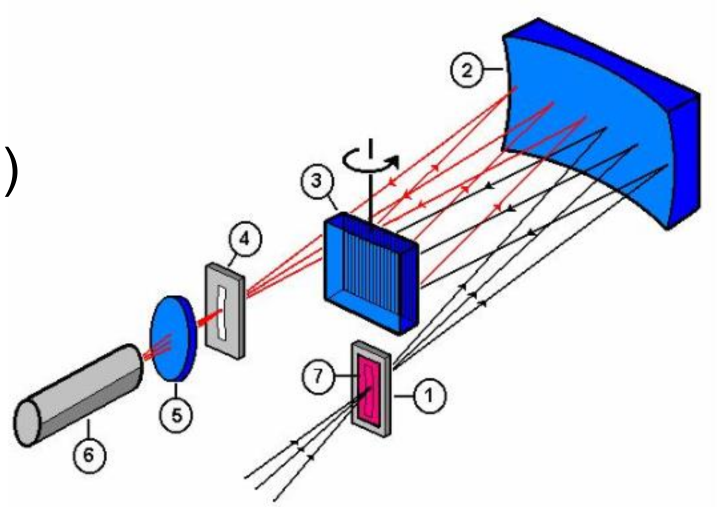
JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

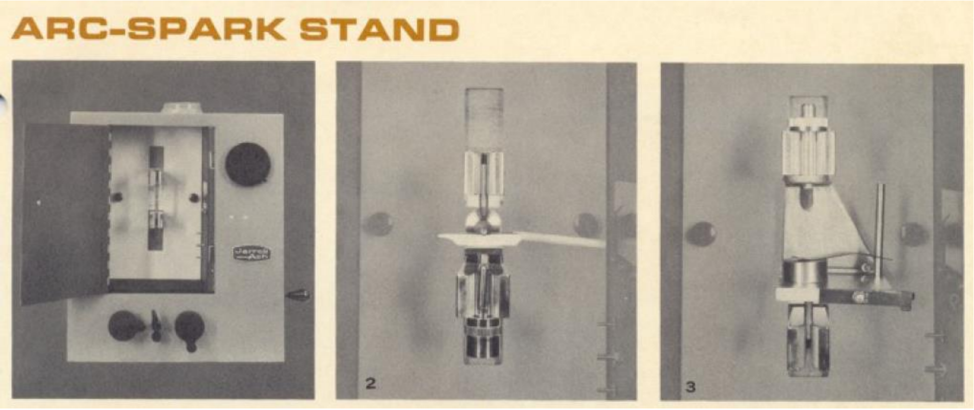
Light actinides: analysis of fluorescence light

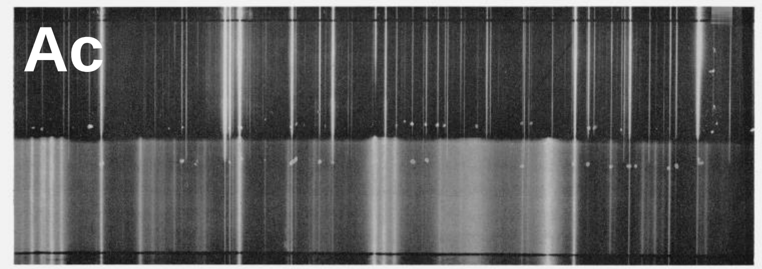
Spectrometry of light from discharge source

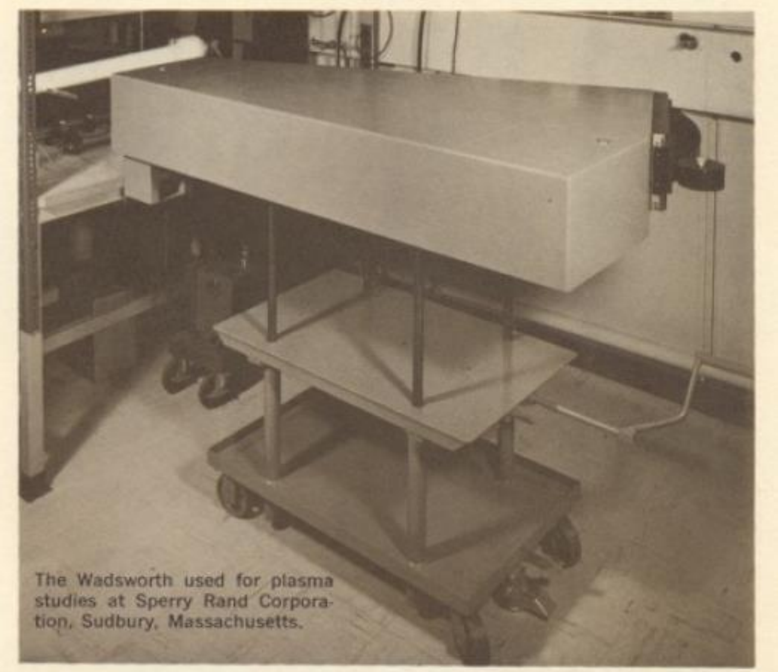
Macroscopic amount (mg…μg)

→ recording of images





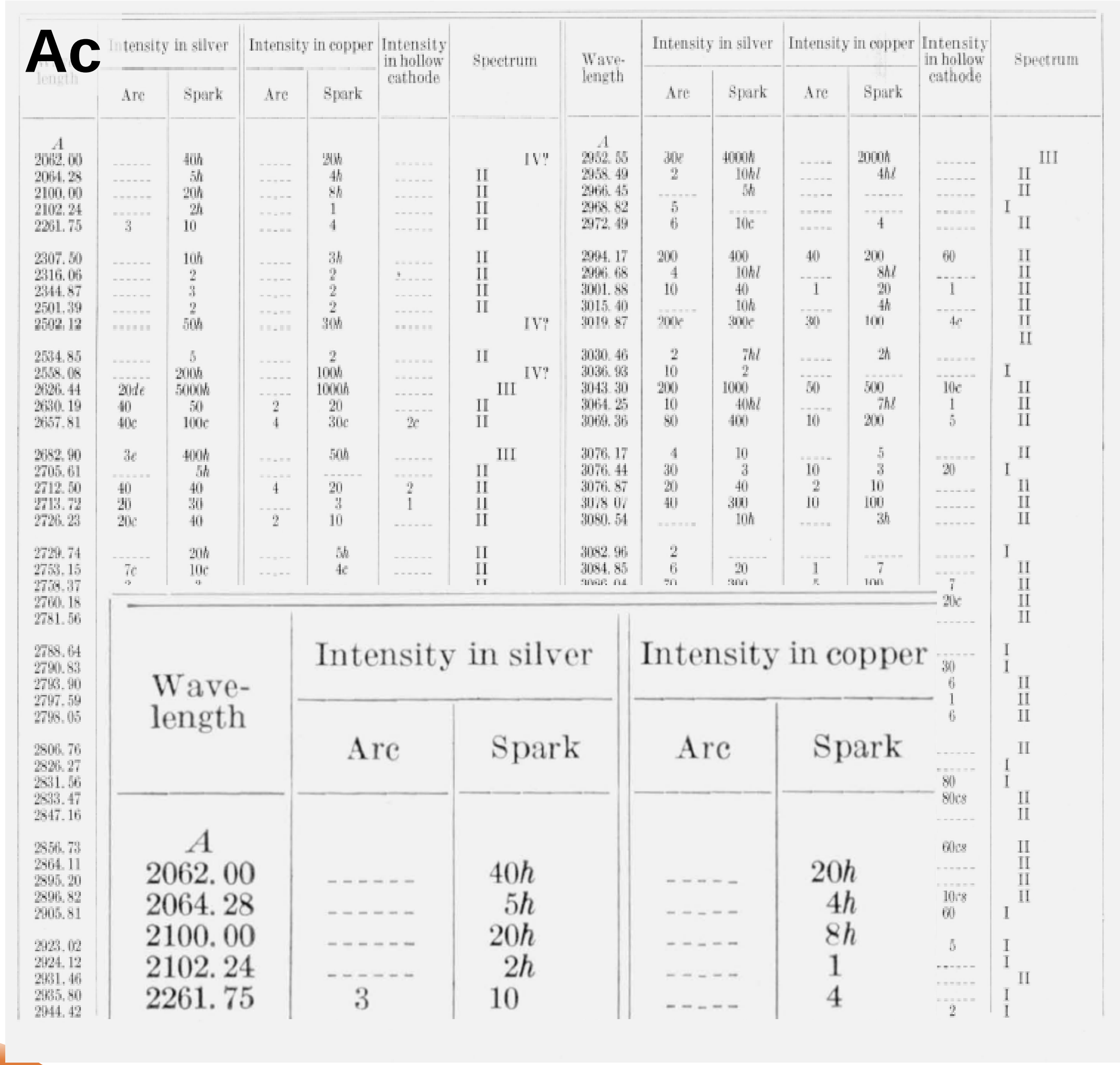




W.F. Meggers Spectrochim. Acta 10 , 2 (1957) 195

HIM HELMHOLTZ Helmholtz-Institut Mainz
GSI RESEARCH FOR GRAND CHALLENGES
JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Light actinides: analysis of fluorescence light



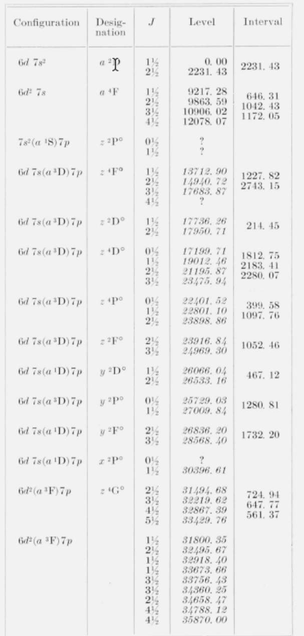
Wave-length	Intensity in silver		Intensity in copper	
	Arc	Spark	Arc	Spark
A				
2062.00	------	40h	-----	20h
2064.28	------	5h	-----	4h
2100.00	------	20h	-----	8h
2102.24	------	2h	-----	1
2261.75	3	10	-----	4

Table of transitions

+ strengths

+ magnetic field
→ Zeemann splitting
J-information

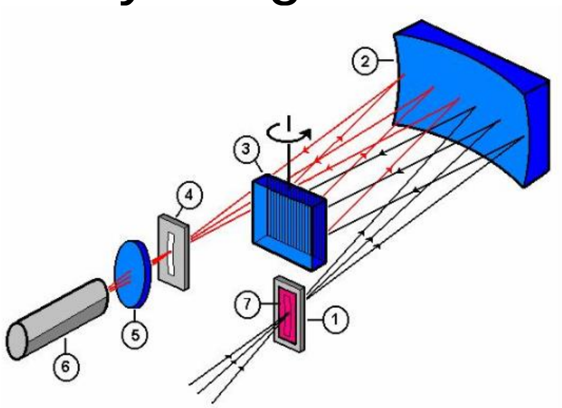
+ analysis of differences
→ level scheme

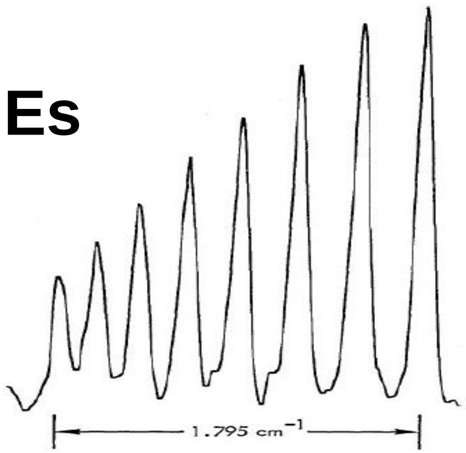


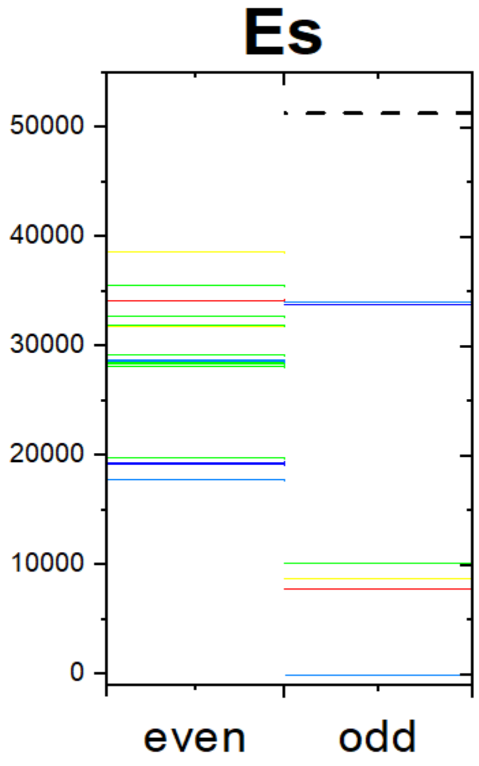
W.F. Meggers Spectrochim. Acta 10 , 2 (1957) 195

^{99}Es – analysis of fluorescence light

Spectrometry of light from discharge







- 0.6 µg – 48 µg ^{253}Es (10^{16} atoms) – 1970's
  - report of ~300 optical lines (Es I & Es II)
  - level assignment from analysis
  - magnetic moment of ^{253}Es from HFS
  - too little material for Zeemann splitting

Worden E.F., et al. *J.Opt. Soc.Am.* 64.1 (1974): 77. *Worden, E.F. et al.*, et al. *J.Opt. Soc.Am. B* 12.7 (1995): 1186.

^{100}Fm – only possible with laser spectroscopy

2003: First atomic information on Fm

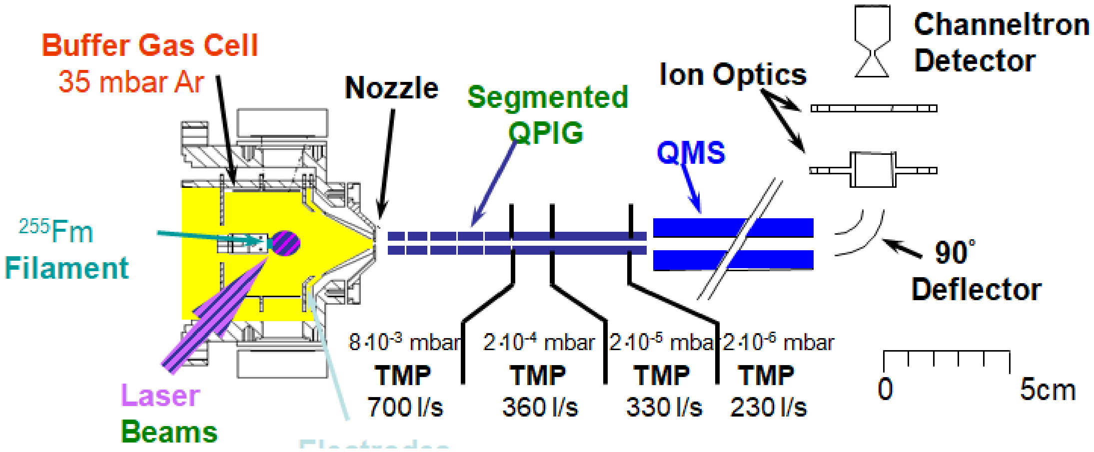
Mainz: Institut für Kernphysik, Institut für Kernchemie

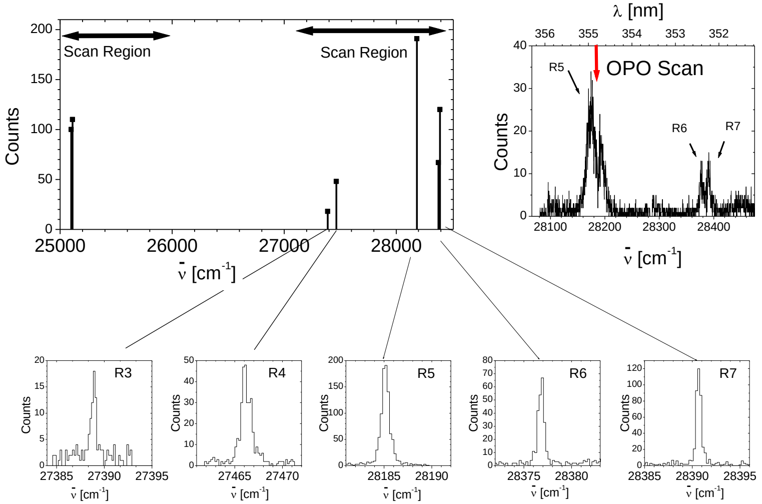
breeding of ^{255}Es
at Oak Ridge, USA

$\Phi_n = 2.6 \cdot 10^{15}/cm^2 \cdot s$
T = 1 a

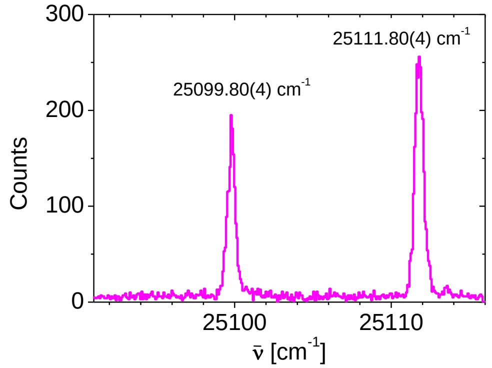
4 ng ^{255}Fm ($t_{1/2}$= 20 h)
(10^{12} atoms)

(1pg ^{257}Fm ($t_{1/2}$= 100 a))





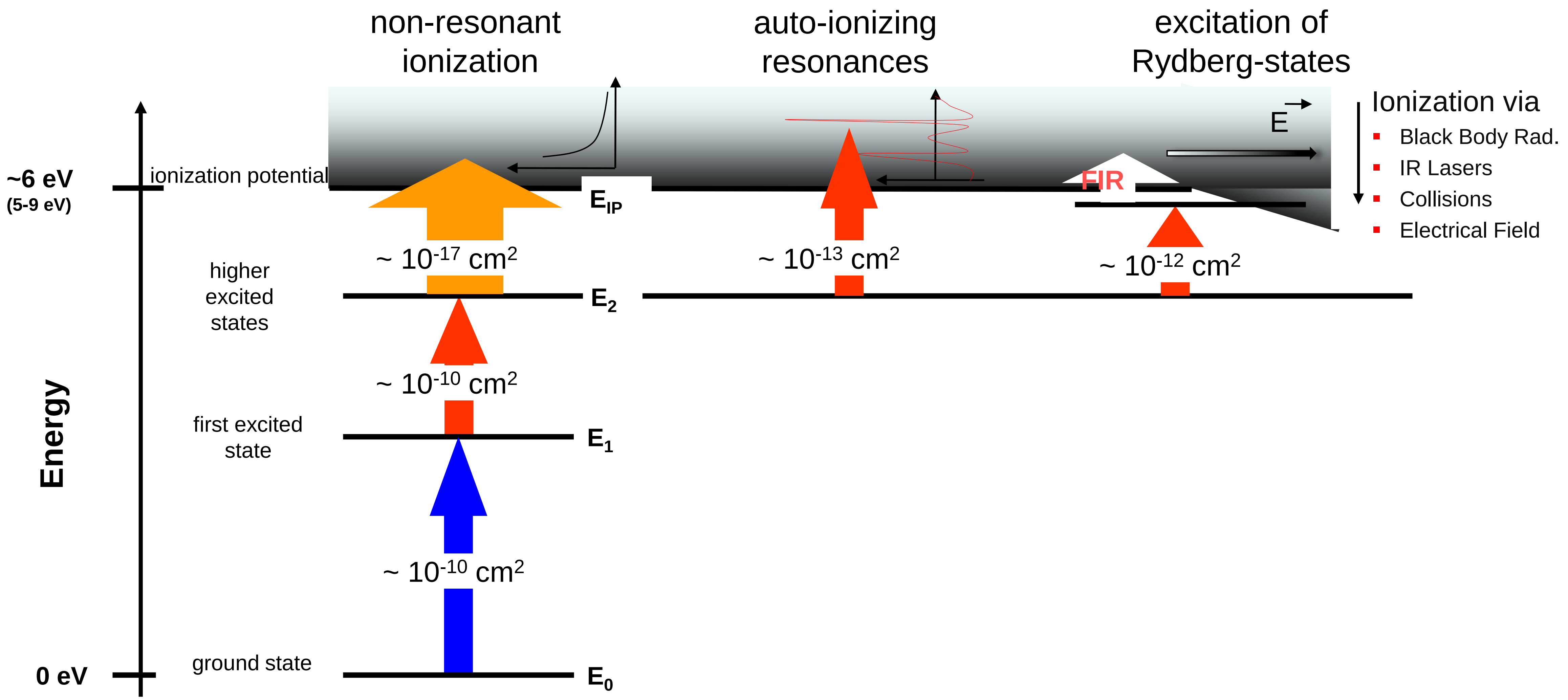
7 atomic transitions



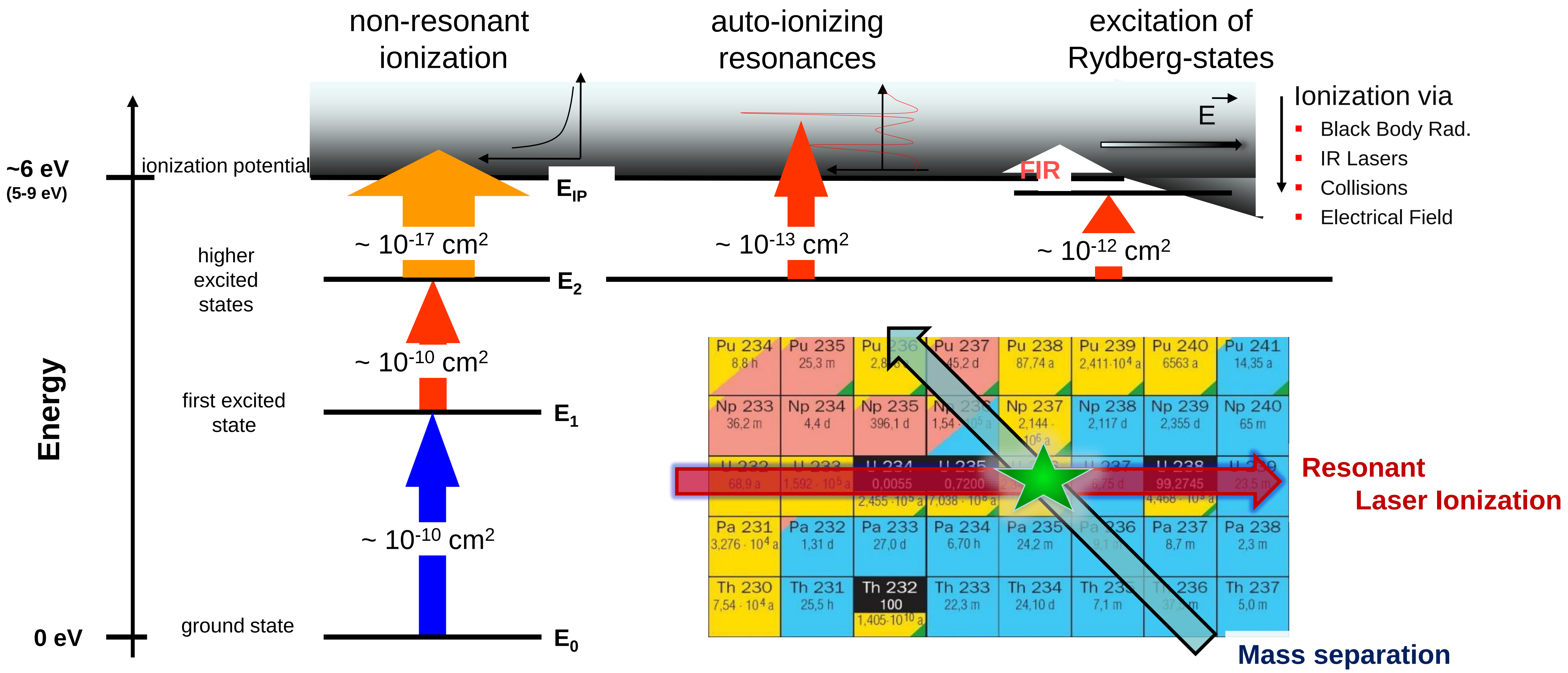
Laser system: 100 Hz, Excimer pumped Dye laser + 50 Hz OPO

[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.

Spectroscopic basis of RIS – atomic structure



Spectroscopic basis of RIS – atomic structure

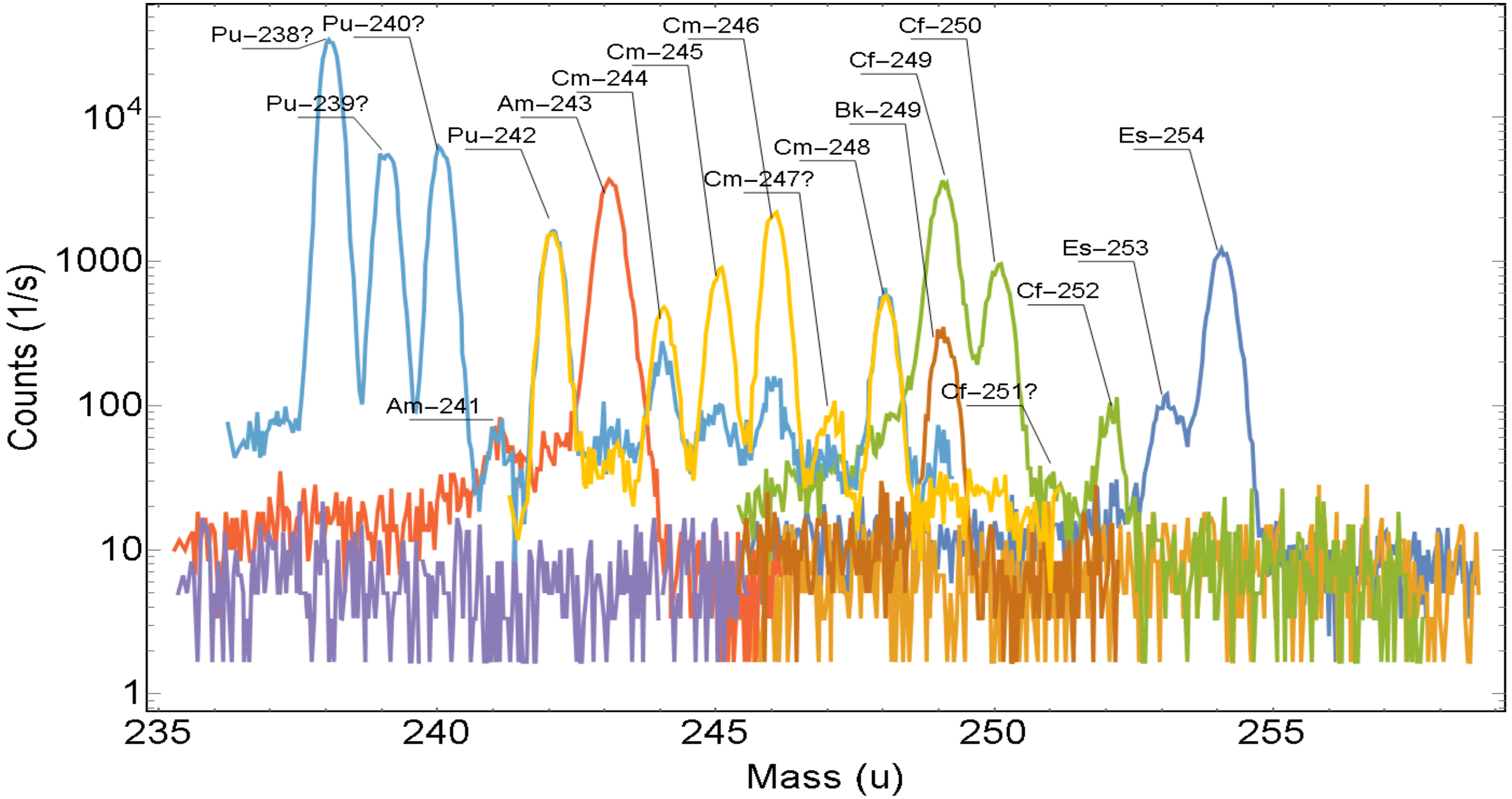


+ Mass separation → isotope selectivity

Sample Analysis with Laser Ionization

- Sample with actinide mixture and limited information

→ Lasers tuned to resonantly ionize different actinide elements



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.

Characterization of sample from ONRL

→ Trace analysis applications

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.

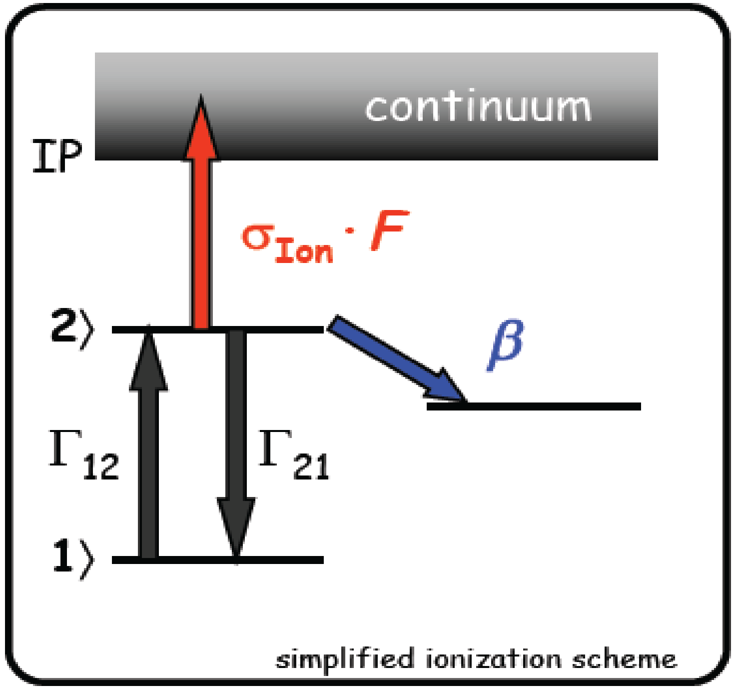
Which laser system to choose?

Efficiency is important and requires

① $\sigma_{Ion} \cdot F \gg \beta$ ⟶ ionization rate » loss rate

② $\sigma_{Ion} \cdot \varphi > 1$ ⟶ number of ionized atoms per laser interaction time (pulse)

σ_{Ion} ionization cross section (non-resonant) (cm^2)
β loss rates to (metastable) states, state dependent
F photon flux ($cm^{-2}\ s^{-1}$)
φ photon fluence (=photon flux · laser interaction time)



Which laser system to choose?

Efficiency is important and requires

1	$\sigma_{Ion} \cdot F \gg \beta$ →	ionization rate » loss rate
2	$\sigma_{Ion} \cdot \varphi > 1$ →	number of ionized atoms per laser interaction time (pulse)

Typical values:

σ_{ion} → 10^{-17} cm^2

β → 10^6 s^{-1}

Assumption:
laser beam area of 1 mm^2
and photon energy of 3 eV.

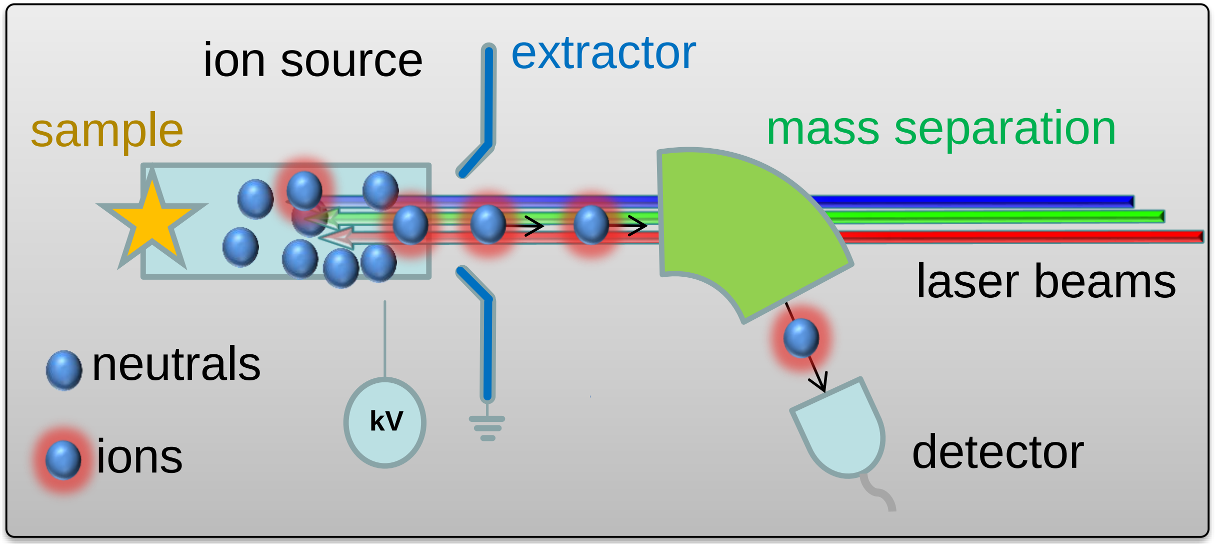
~~Continous Laser:~~ From (1.) Flux $F \gg 10^{23}$ $cm^{-2}s^{-1}$ → # photons required $\gg 10^{21}$ /s → » 500 W Impossible

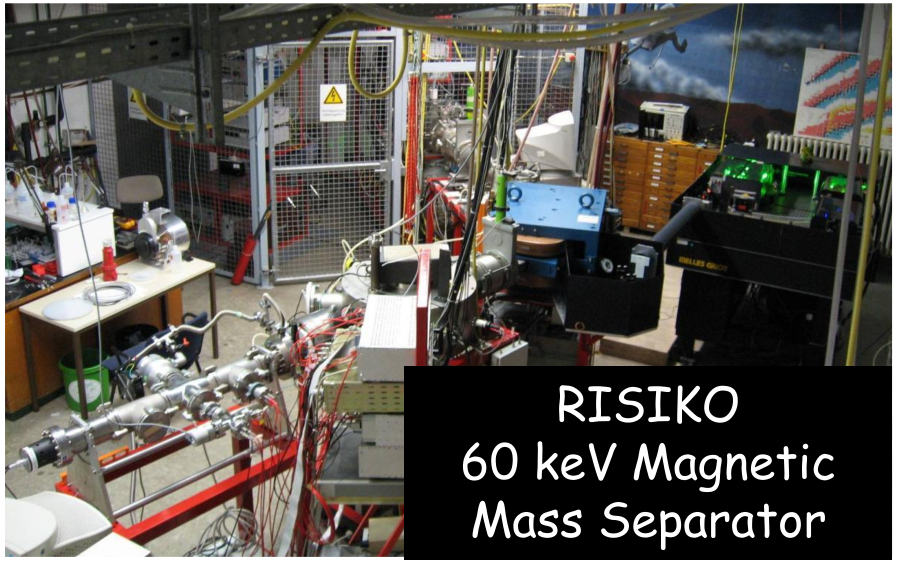
Pulsed Laser: (10 ns)

With a pulsed laser system: (1.) → » 5 µJ/pulse No problem !!

Lets add in the **Fluence** (2.) condition → > 0.5 mJ/pulse

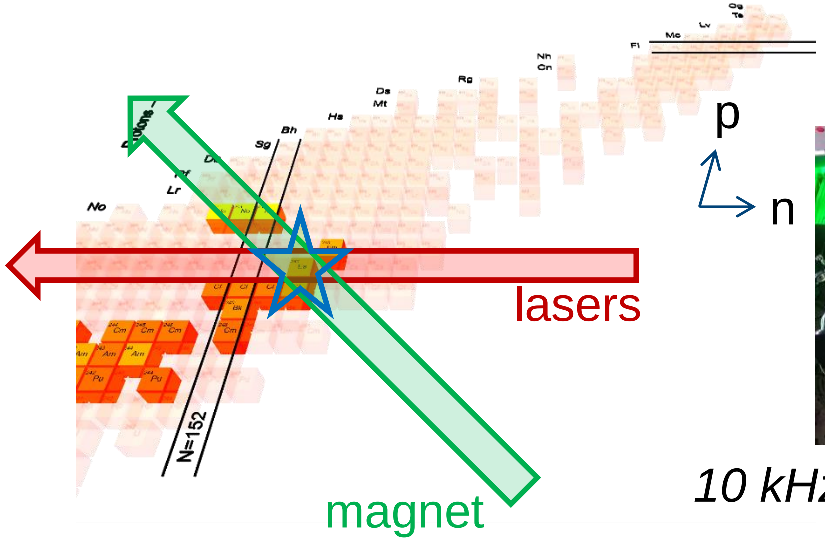
Hot Cavity Resonance Ionization Spectroscopy

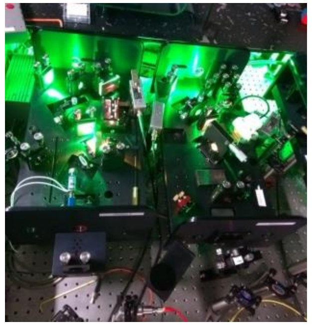




- Used for production of radioactive ion beams
- Laser spectroscopy with high efficiency
- Background from surface ionization
- Resolution limited by

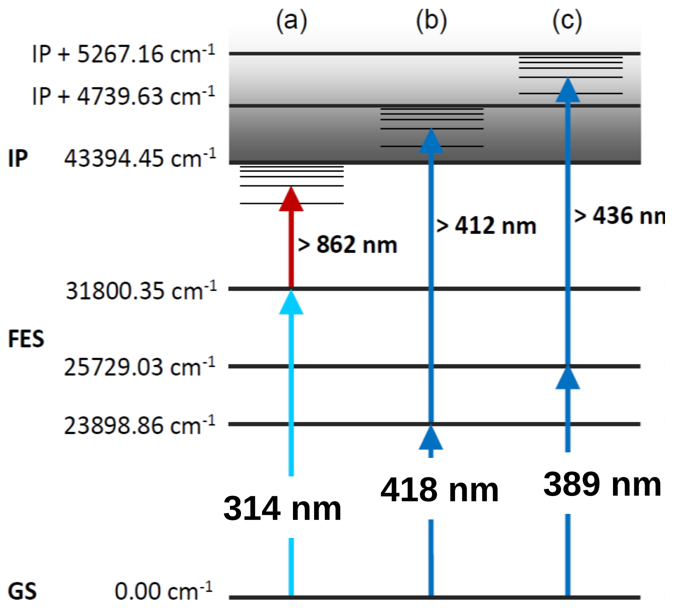
  source temperature and laser bandwidth

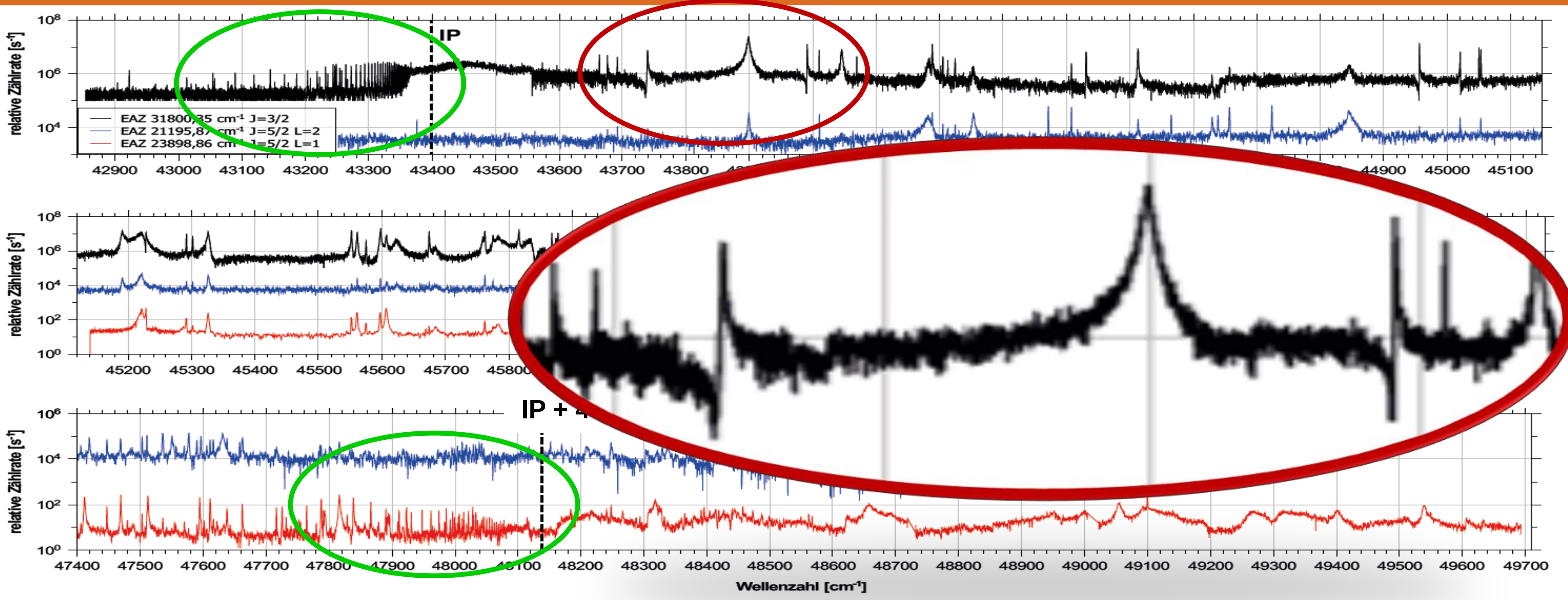


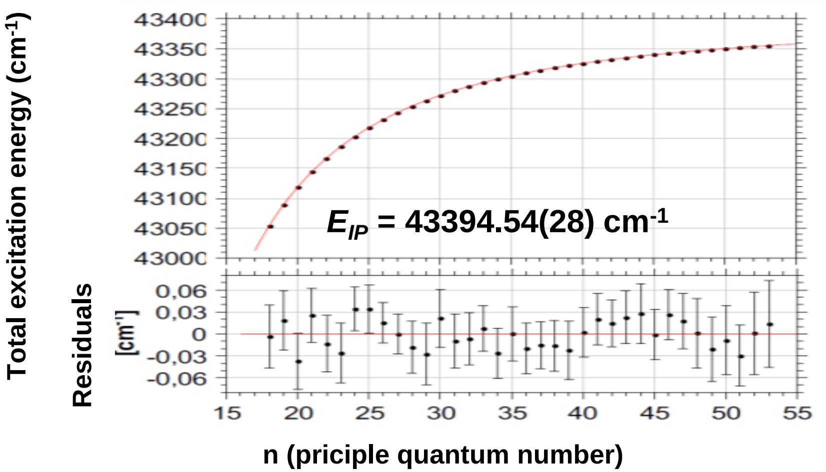


10 kHz Laser system

Ionization Potential of Ac







Rydberg Ritz formula

$$E_n = E_{IP} - \frac{R_M}{(n-\delta(n))^2}$$

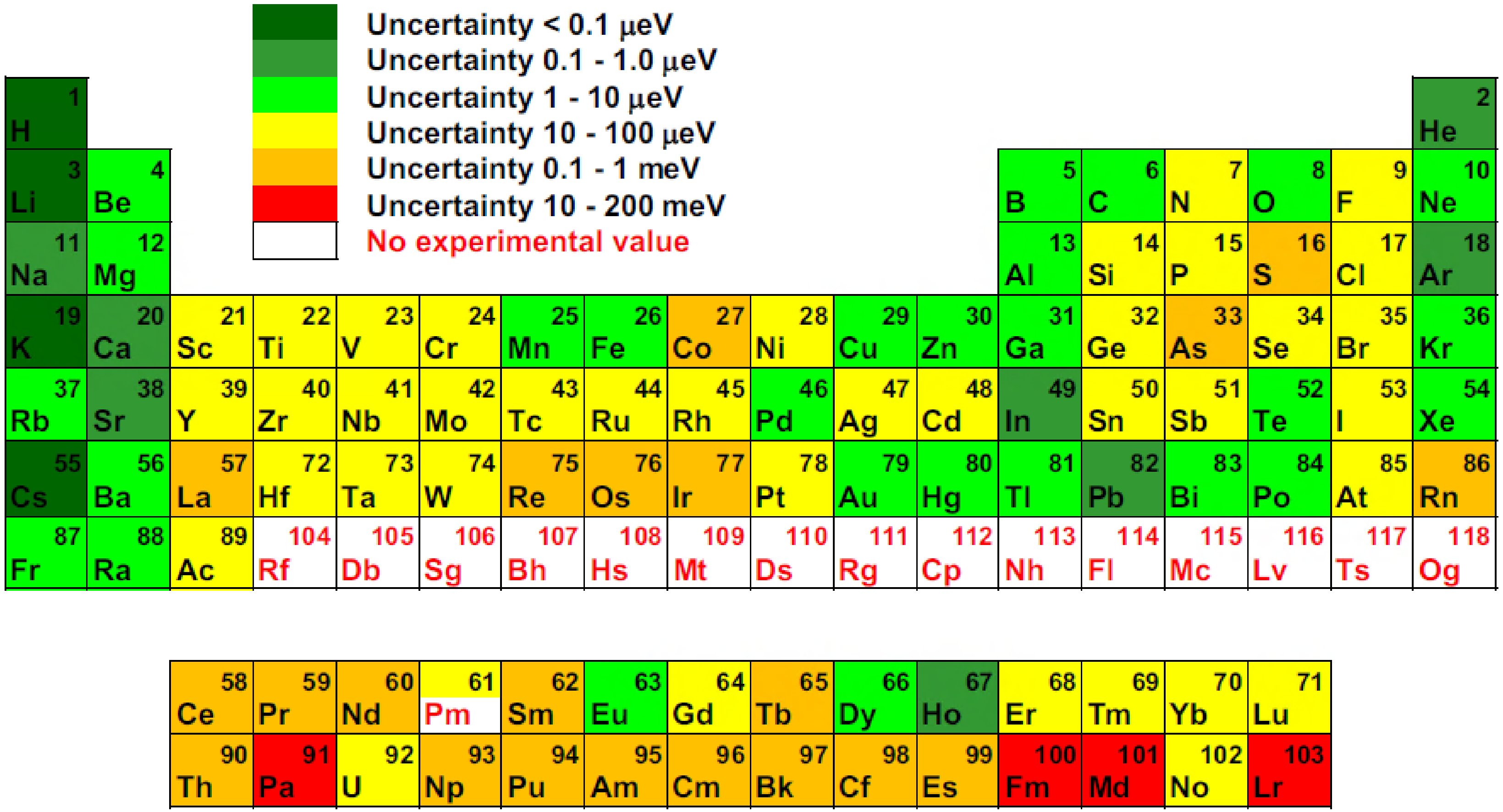
Weighted average value

E_{IP} = **43 394.45(19) cm^{-1}**

$$R_M = \frac{M}{M+m} R_\infty \qquad \delta(n) = A + \frac{B}{(n-A)^2}$$

J. Rossnagel *et al.*, Phys. Rev. A **85**, 012525 (2012)

Ionization potentials of the actinides



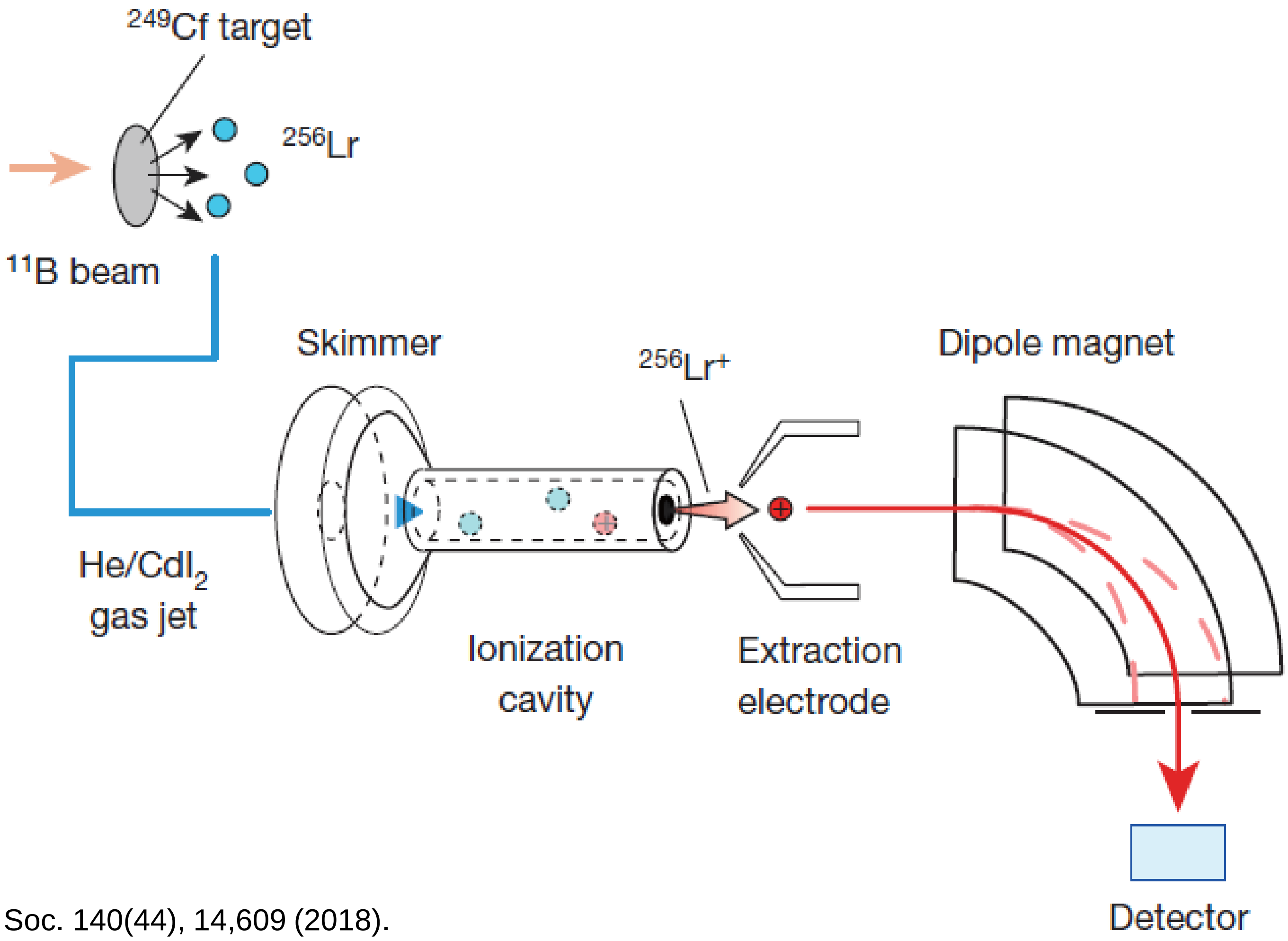
Electron affinity is also under study pushing towards actinide elements

Studer, Dominik, et al., *Physical Review A* 99.6 (2019): 062513.
Wendt, K., et al., *Hyperfine Interactions* 227.1 (2014): 55-67.

Ionization potentials of the actinides

Heavy actinides: Surface ionization efficiencv in a hot cavitv

Japan Atomic Energy Agency

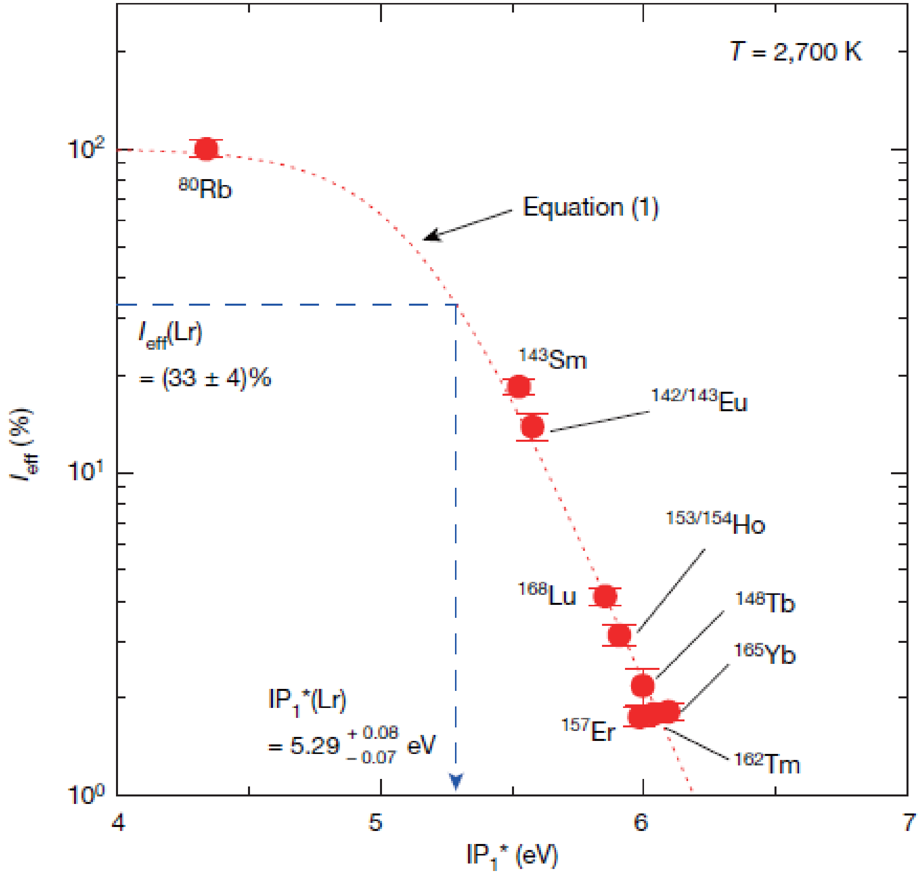


Sato, T.K. et al., J. Am. Chem. Soc. 140(44), 14,609 (2018).
Sato, T. K., et al., *Nature* 520.7546 (2015): 209-211.

Ionization potentials of the actinides

Surface ionization efficiency in a hot cavity

Japan Atomic Energy Agency

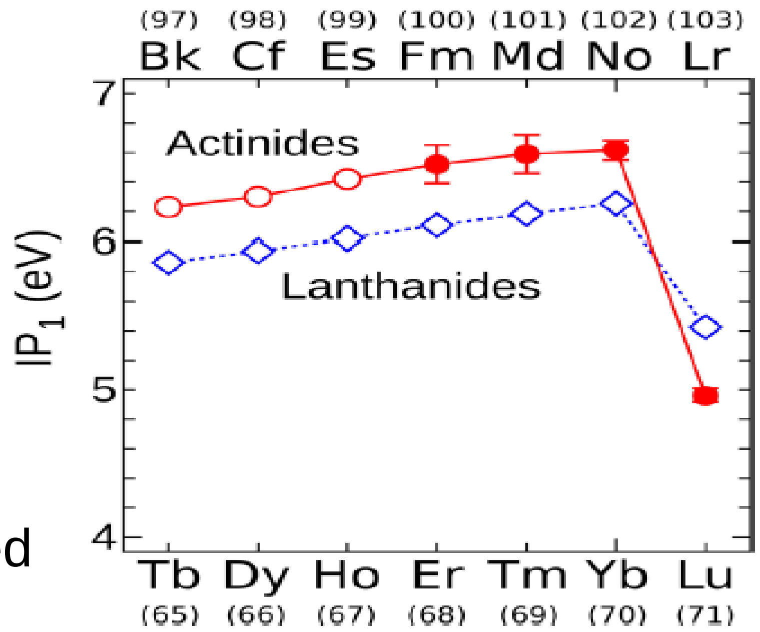


$$I_{\mathrm{eff}} = \frac{N\exp\left(\frac{\phi - \mathrm{IP}_1^*}{kT}\right)}{1 + N\exp\left(\frac{\phi - \mathrm{IP}_1^*}{kT}\right)}$$

$$\mathrm{IP}_1^* = \mathrm{IP}_1 - kT\ln\left(\frac{Q_i}{Q_0}\right)$$

Access to effective IP

→ atomic theory input required



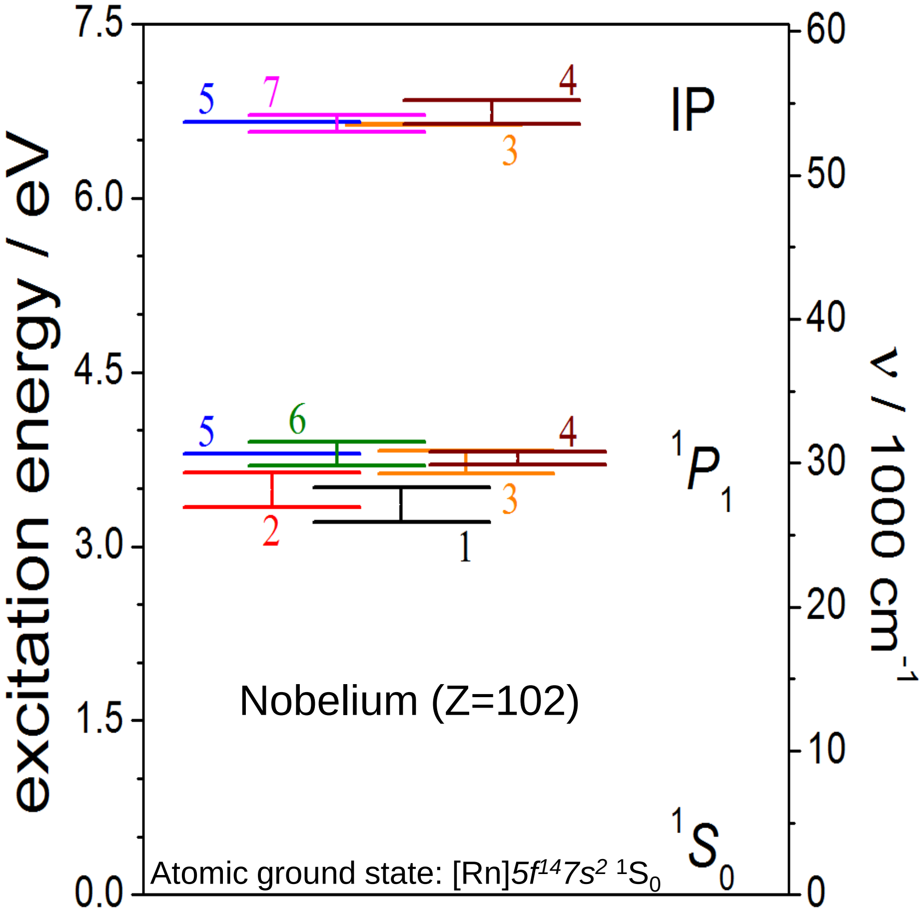
Fm-Lr (± 600 cm^{-1})

Sato, T.K. et al., J. Am. Chem. Soc. 140(44), 14,609 (2018).
Sato, T. K., et al., *Nature* 520.7546 (2015): 209-211.

Surface ionization – Tokai

HIM HELMHOLTZ Helmholtz-Institut Mainz
GSI RESEARCH FOR GRAND CHALLENGES
JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Laser spectroscopy of nobelium



- Element of interest: No (Z=102)
  - "simple" atomic structure GS: [Rn]$5f^{14}7s^2$ $^1S_0$
  - Relatively high production cross sections

Isotope	I^P	$T_{1/2}$ (s)	Nuclear reaction	Max. production on target (1/s)	Alpha energy (MeV)
^{251}No	0	0.8	^{206}Pb(^{48}Ca,3n)^{251}No	0.2	8.61
^{252}No	0	2.4	^{206}Pb(^{48}Ca,2n)^{252}No	4	8.42
^{253}No	(9/2$^-$)	102	^{207}Pb(^{48}Ca,2n)^{253}No	11	8.01
^{254}No	**0**	**51**	**^{208}Pb(^{48}Ca,2n)^{254}No**	**17**	**8.10**
^{255}No	(1/2$^+$)	186	^{208}Pb(^{48}Ca,1n)^{255}No	2	8.12
^{255}No	(1/2$^+$)	186	^{209}Bi(^{48}Ca,2n)^{255}Lr → EC	1	8.12
^{255}Lr	**(1/2$^-$)**	**31.1**	**^{209}Bi(^{48}Ca,2n)^{255}Lr**	**3.4**	**8.37**

Model calculations

1, 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

Production: Velocity Filter SHIP

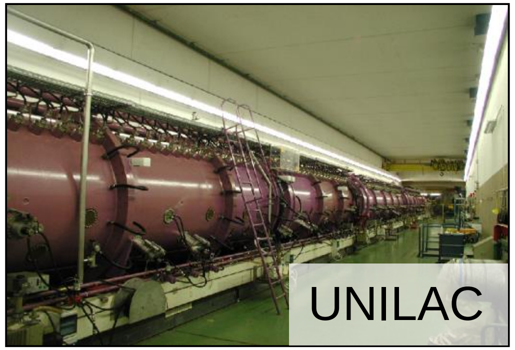
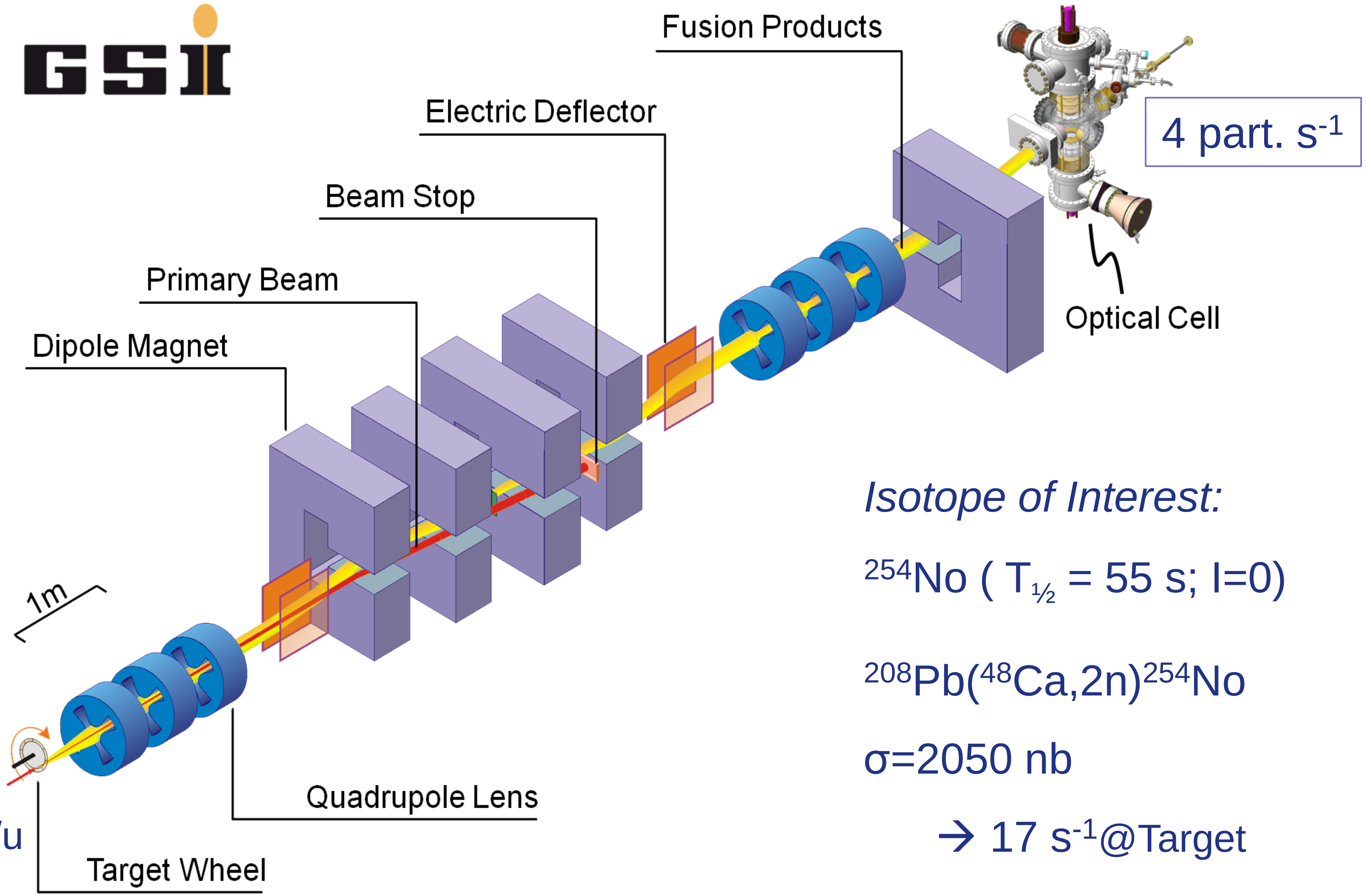


Bild: GSI Helmholtzzentrum für Schwerionenforschung



^{48}Ca
4.55 MeV/u
1 pμA

Isotope of Interest:

^{254}No ($T_{½}$ = 55 s; I=0)

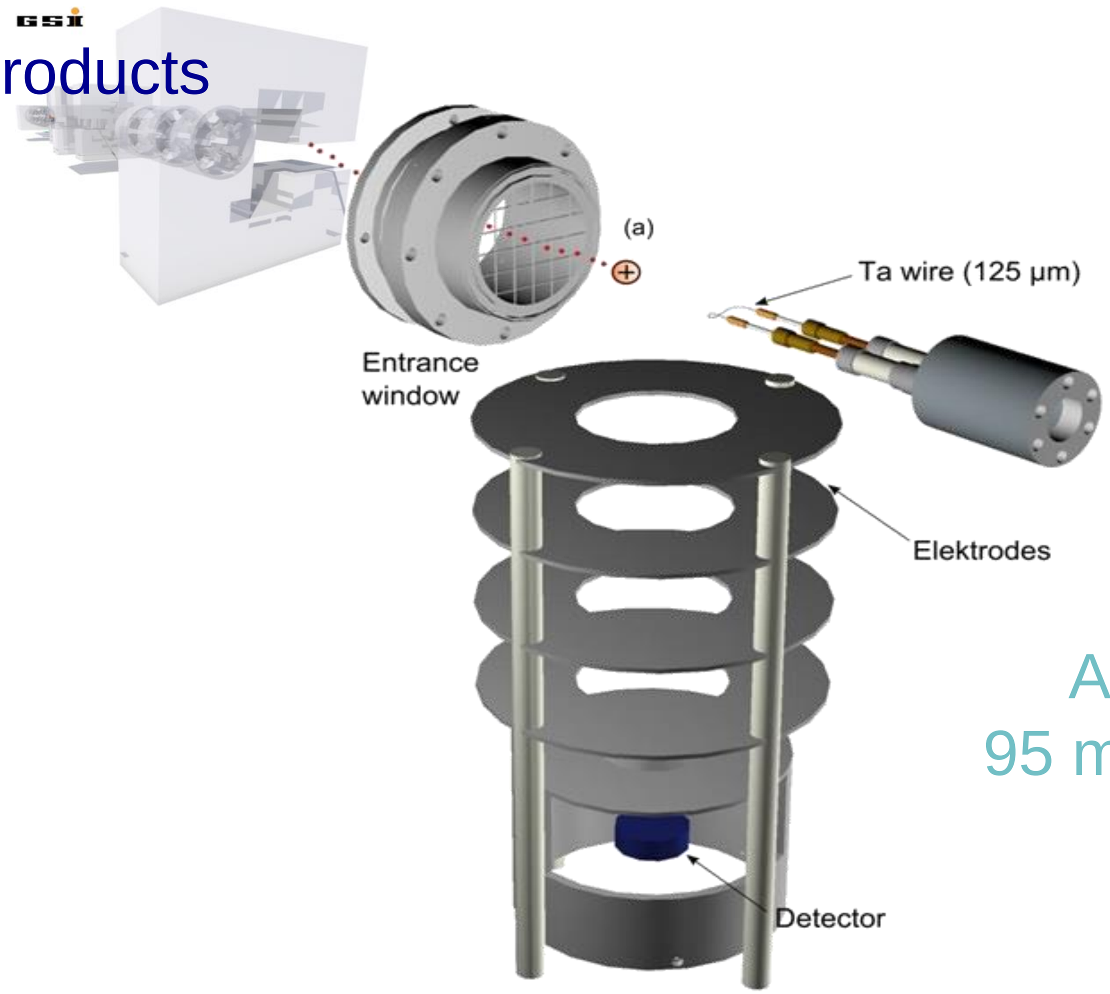
^{208}Pb(^{48}Ca,2n)^{254}No

σ=2050 nb

→ 17 s^{-1}@Target

Radiation Detected Resonance Ionization Spectroscopy

(a) Stopping of the incoming fusion products

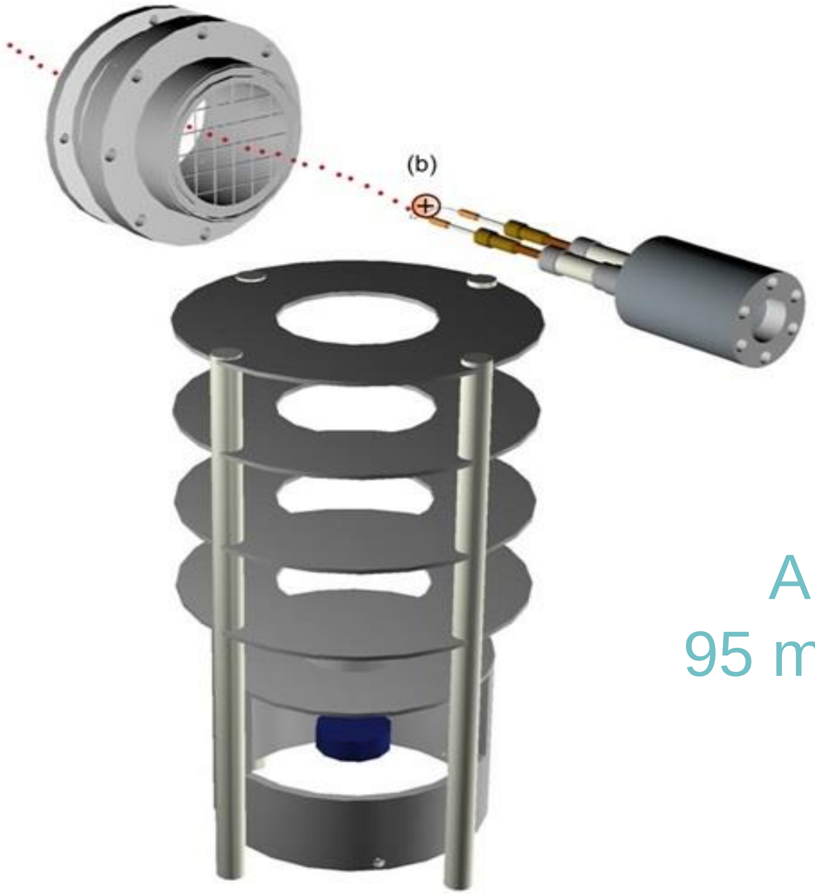


Ar
95 mbar

Radiation Detected Resonance Ionization Spectroscopy

(a) Stopping of the incoming fusion products

(b) Collecting onto thin tantalum wire



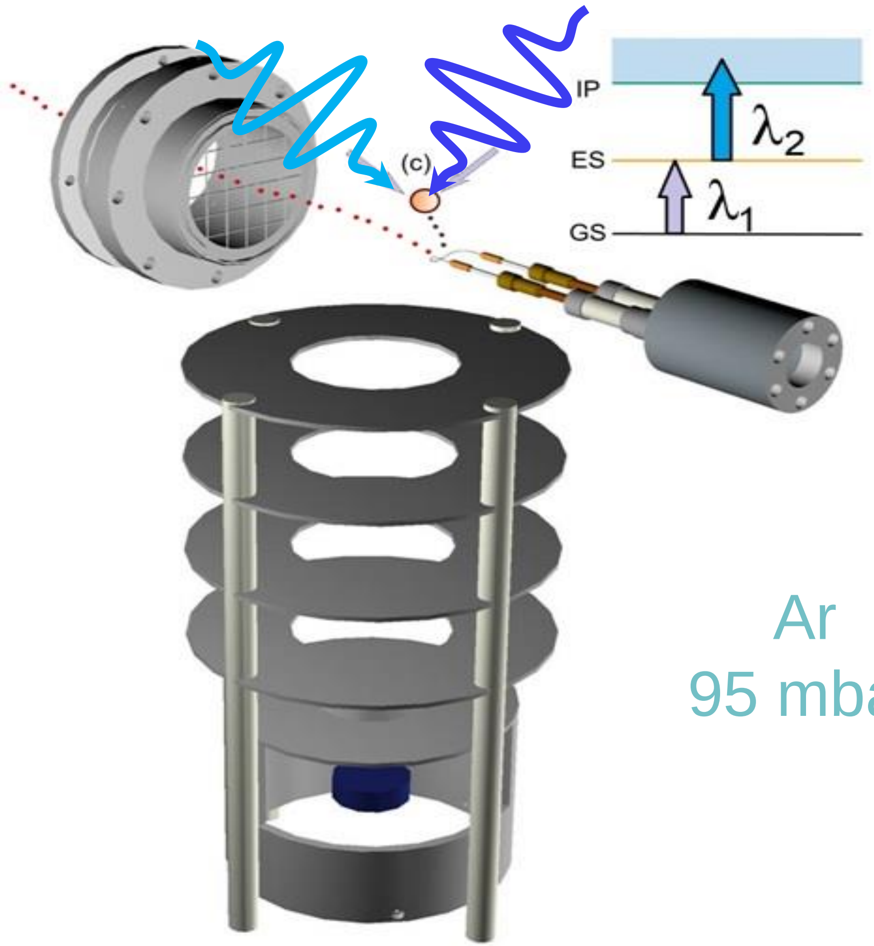
Ar
95 mbar

Radiation Detected Resonance Ionization Spectroscopy

(a) Stopping of the incoming fusion products

(b) Collecting onto thin tantalum wire

(c) Evaporation and two-step photoionization process



Ar
95 mbar

Radiation Detected Resonance Ionization Spectroscopy

(a) Stopping of the incoming fusion products

(b) Collecting onto thin tantalum wire

(c) Evaporation and two-step photoionization
process

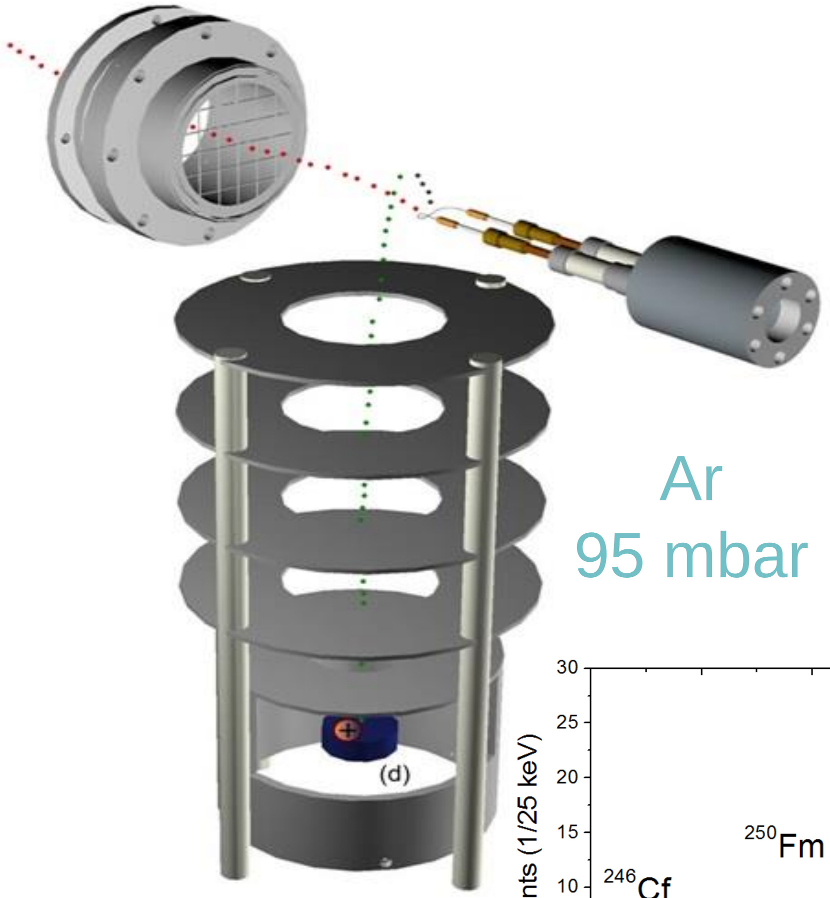
(d) Transport to detector and detection
of alpha decay

RADRIS

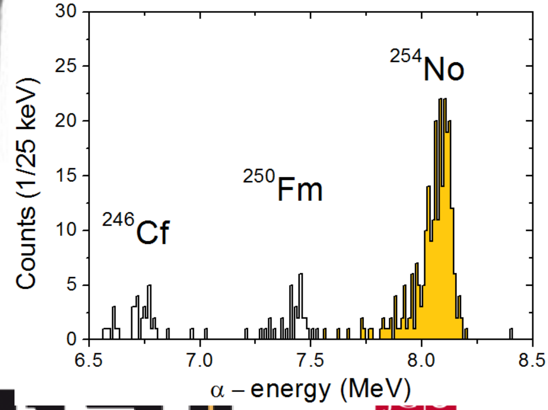
Radiation Detected Resonance
Ionization Spectroscopy

Short-lived alpha emitters ($t_{1/2} \leq 4$ min)

Low production rates down to 1/10s



Ar
95 mbar

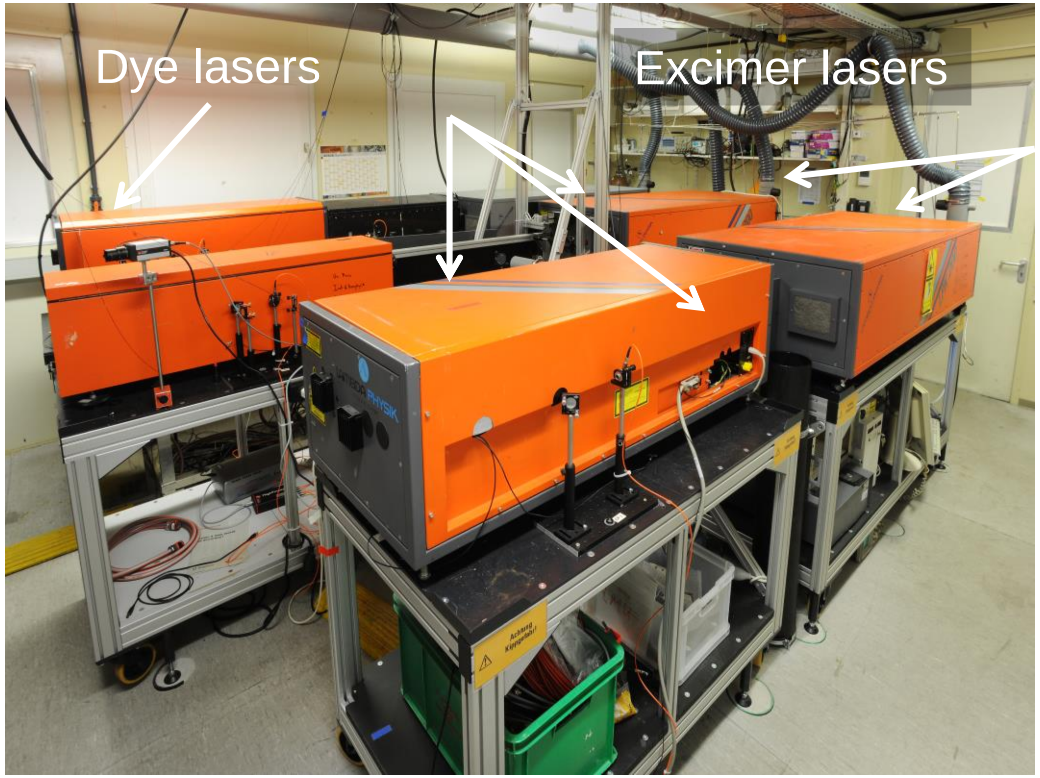


F. Lautenschläger et al., NIMB ***383****, 115 (2016)*   *H. Backe et al., Nucl Phys. A* ***944****, 492 (2015)*

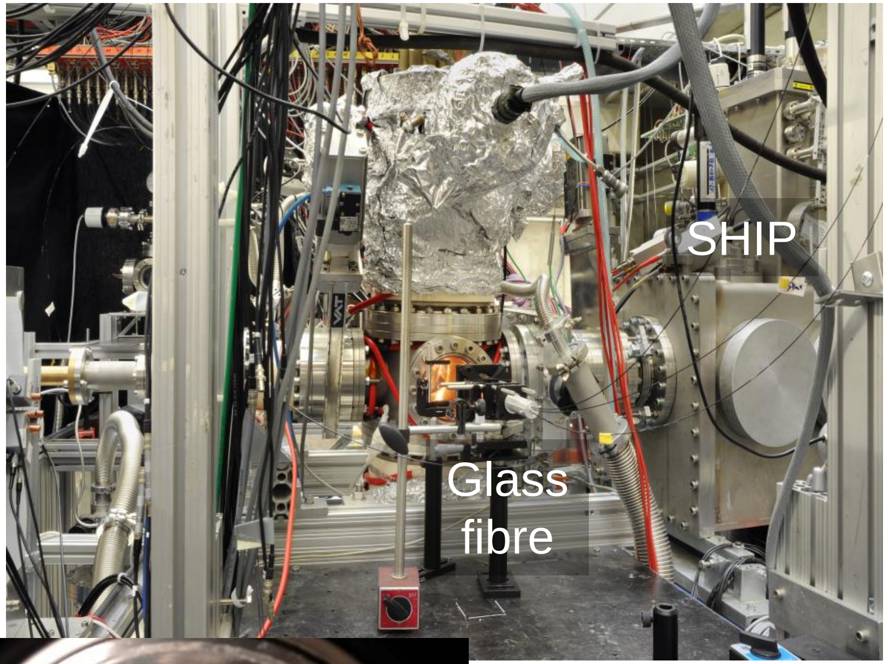
HIM HELMHOLTZ Helmholtz-Institut Mainz
RESEARCH FOR GRAND CHALLENGES
JOHANNES GUTENBERG UNIVERSITÄT MAINZ

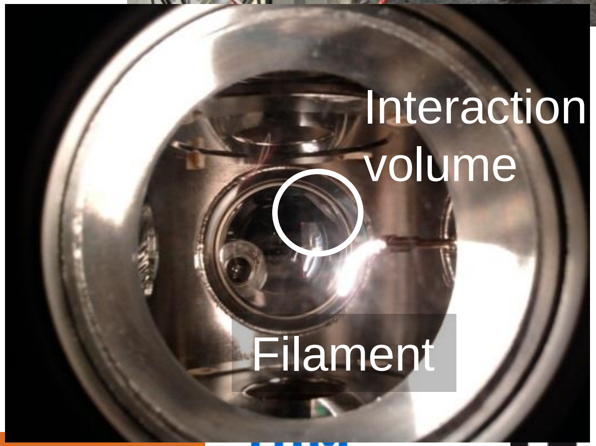
Lab impressions

Laser system



Gas cell



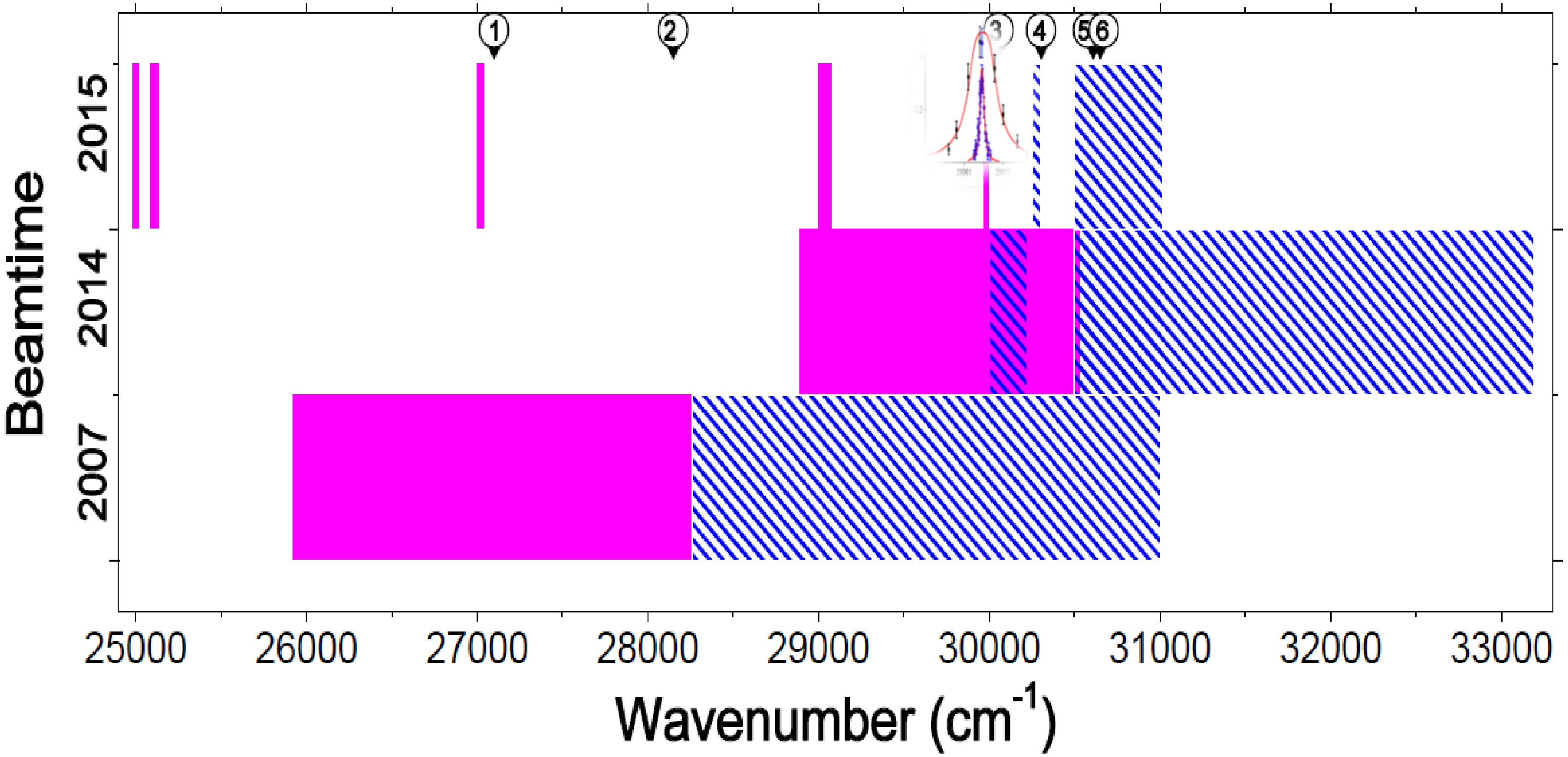


Bilder: GSI Helmholtzzentrum für Schwerionenforschung

HELMHOLTZ Helmholtz-Institut Mainz · RESEARCH FOR GRAND CHALLENGES · JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Level Search in ^{254}No

Year	2007	2014
Scan range (cm^{-1})	25920 – 31001	28887 – 33191
Net scan time (h)	39	67



1: MCDF (2005), 2: MCDF (2005), 3: IHFSCC (2007), 4: RCC (2014), 5: MCDF (2007), 6: MCDF (2007)

HIM HELMHOLTZ Helmholtz-Institut Mainz

GSI RESEARCH FOR GRAND CHALLENGES

JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

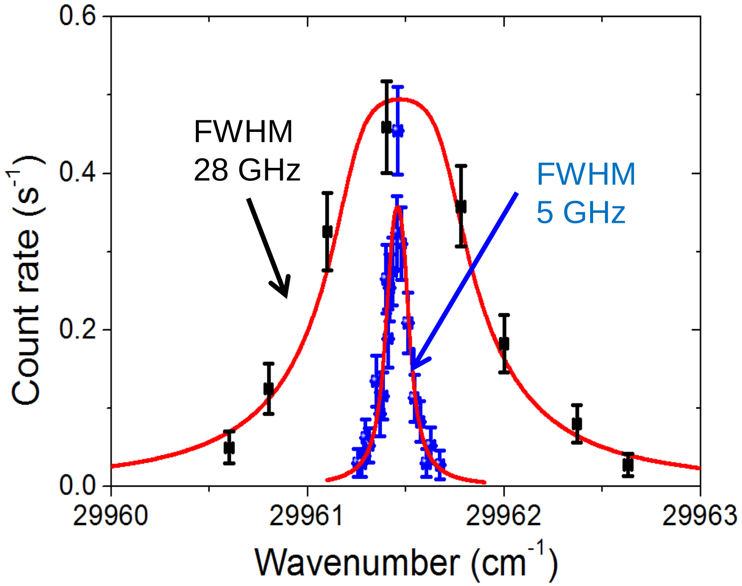
The Ground-State Transition

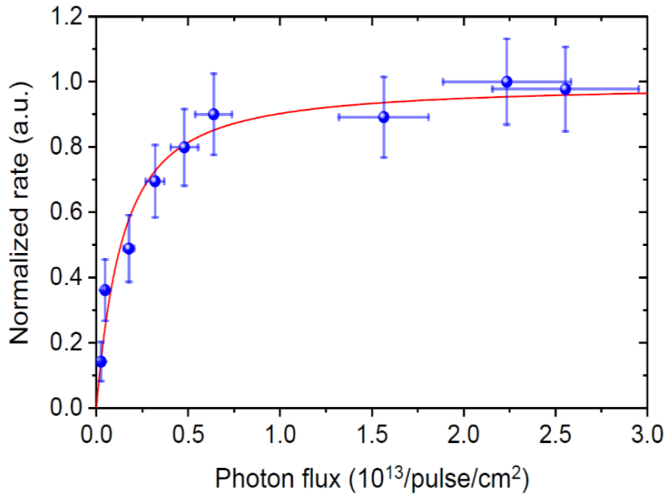
Observed strong atomic ground state transition

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

	ν_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 $^1S_0 \rightarrow {}^1P_1$ transition





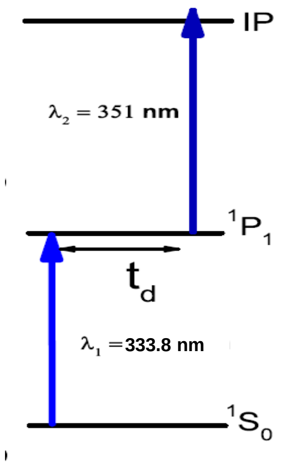
[1] *M. Laatiaoui et al., Nature* 538 (2016) 7626 *[2] A. Borschevsky et al., Phys. Rev. A* ***75*** *(2007) 042514*
[3] P. Indelicato et al., Eur. Phys. J. D ***45****, (2007) 155*

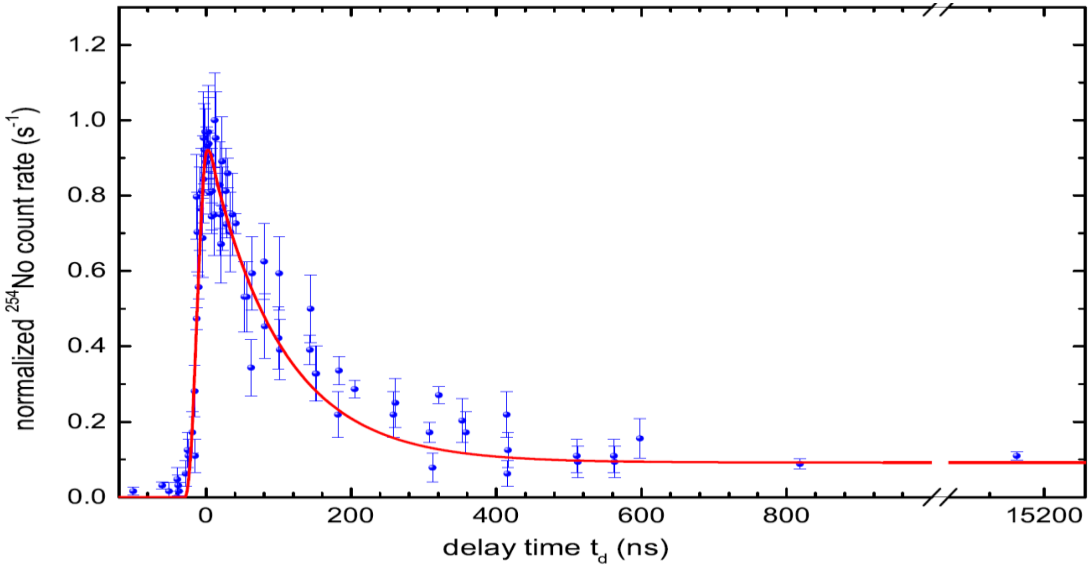
HIM HELMHOLTZ Helmholtz-Institut Mainz GSI RESEARCH FOR GRAND CHALLENGES JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Delayed ionization

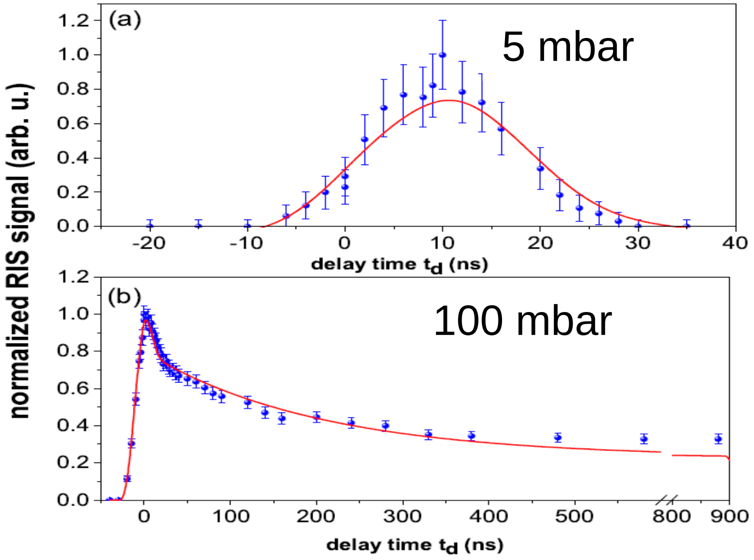
Independent lifetime determination by delayed ionization

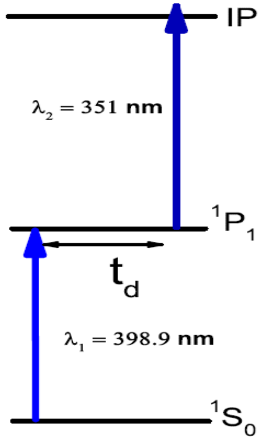
No





Yb



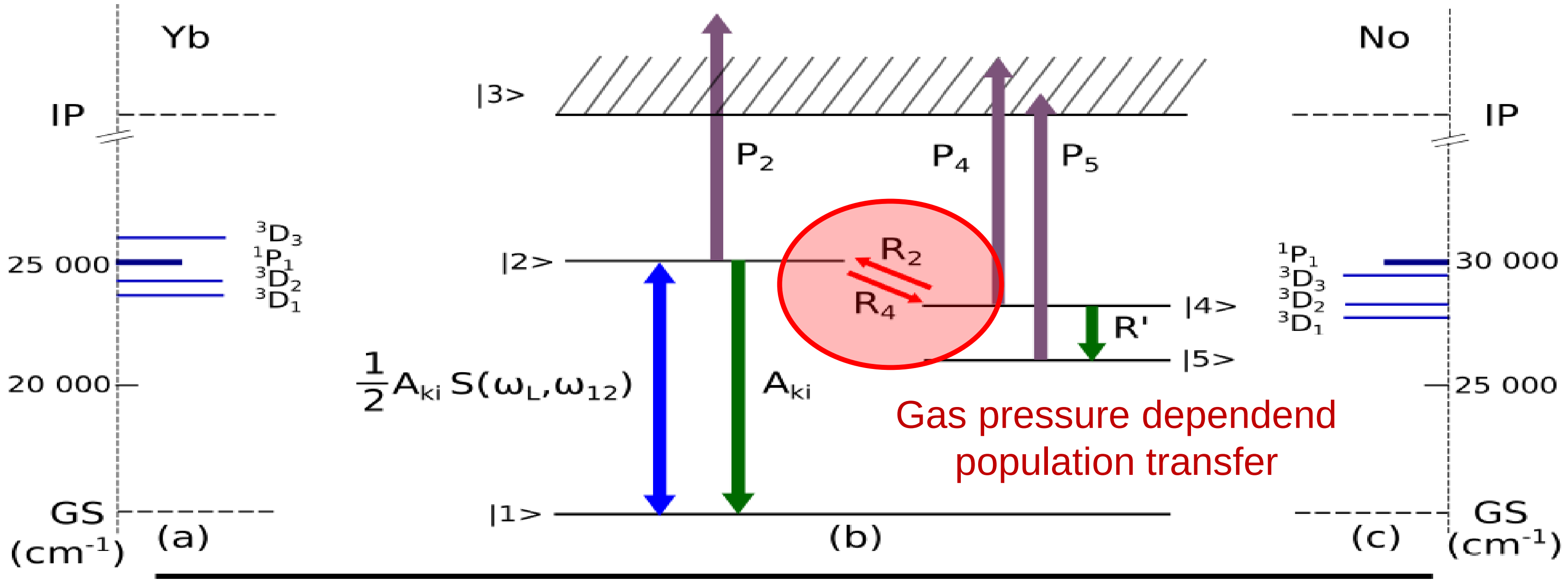


$T_{½}$ > 50 ns – suggest much weaker optical transition

Gas induced quenching

Population transfer to close lying levels

needs to be energetically lower but closeby

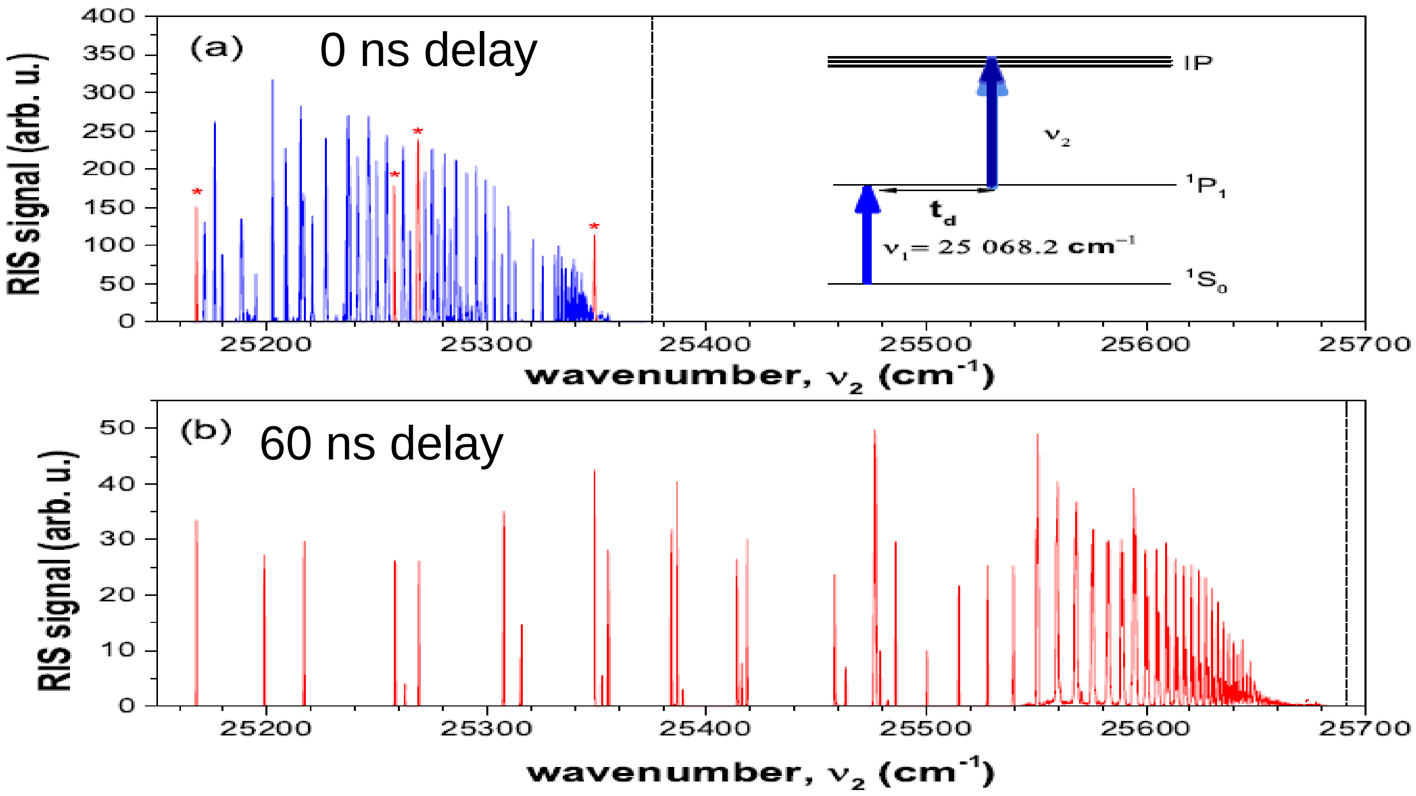


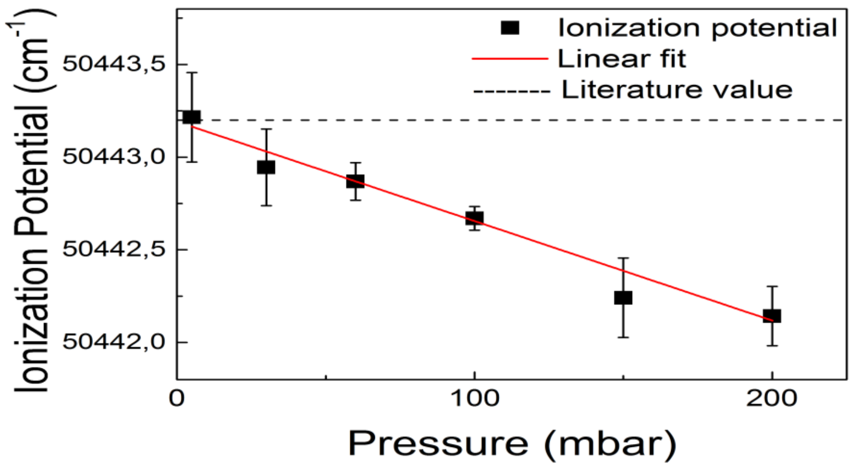
- Collisions with buffer gas atoms– quenching of $^1P_1$ State
- Population of metastable D-states

How to probe this?

Gas induced quenching

Excitation to higher lying levels → Rydberg states





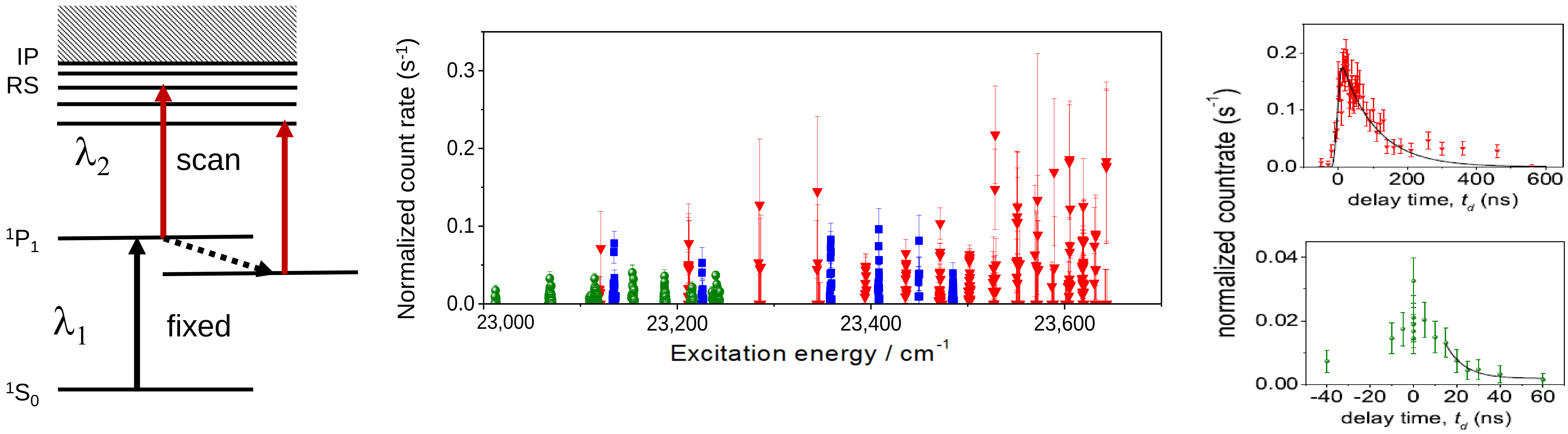
Dependence of the extracted IP from the gas pressure

Ionization via Rydberg state yields the correct lifetime in the gas environment

*P. Chhetri al., Eur. Phys. J. D **71** (2017) 195*

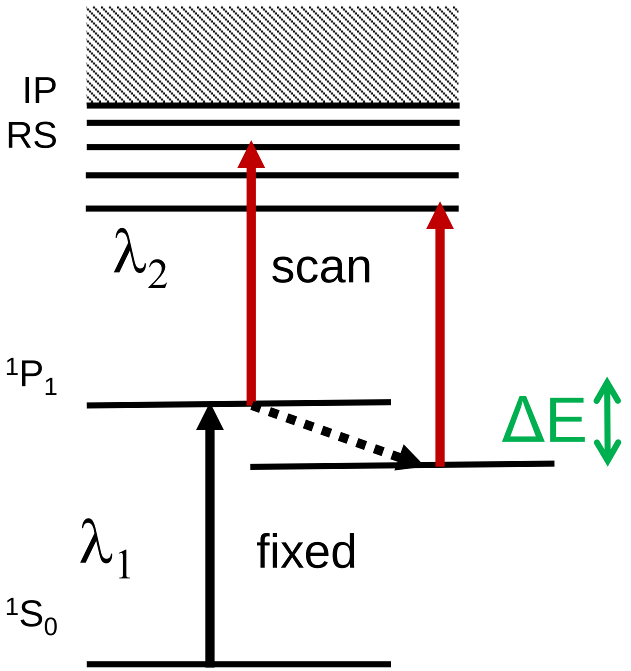
First Ionization Potential of No

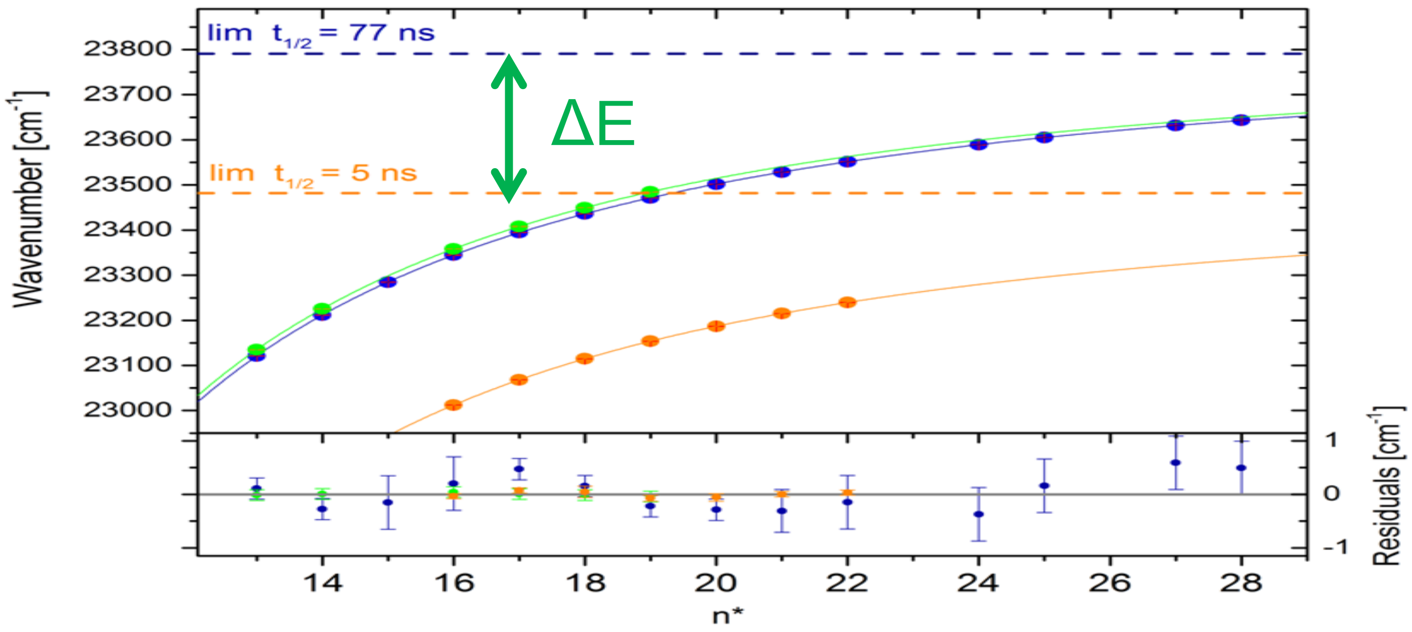
Observation of Rydberg states with two-step resonance ionization



- Idendification of 30 Rydberg levels in ^{254}No
- Quenching in buffergas, signature of different life time

Ionization Limits & Ionization Potential





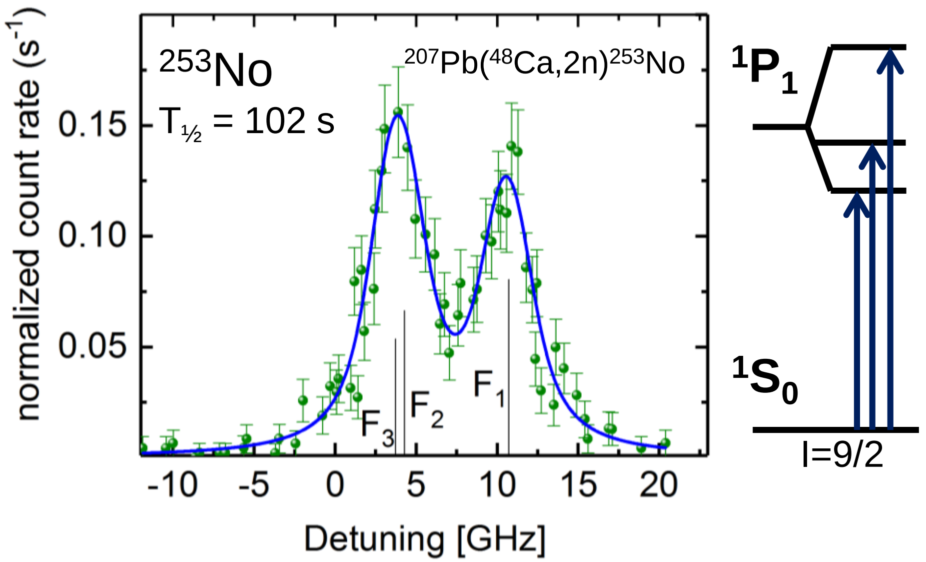
- Series fitted with Rydberg-Ritz formula:

$$E_n = E_{\mathrm{IP}} - \frac{R_\mu}{[n - \delta(n)]^2}.$$

	IP (eV)
Experiment	6.6261(3)
IHFSCC [1]	6.632

[1] *A. Borschevsky et al., Phys. Rev. A* ***75*** *(2007) 042514*

Hyperfine Structure Studies in ^{253}No



Hyperfine structure partly resolved

Energy splitting

$$\Delta E_{HFS} = A\cdot\frac{C}{2} + B\cdot\frac{3/4C(C+1)-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

$$C = F(F+1) - I(I-1) - J(J-1)$$

nuclear atomic properties

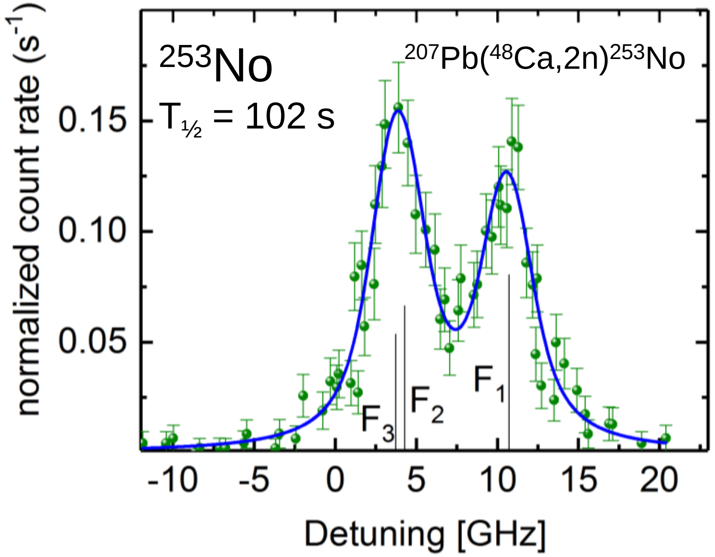
$$A = \mu\cdot\frac{B_e(0)}{IJ}$$

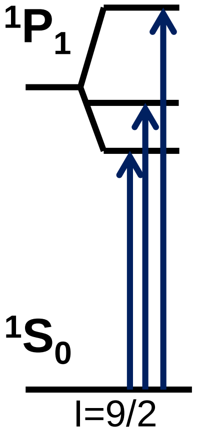
$$B = eQ_s\left\langle\frac{\delta^2 V}{\delta z^2}\right\rangle_{z=0}$$

Feedback from **atomic theory** for nuclear moments

	Ref.	A/μ_N (GHZ)	B/eb (GHZ)
Theory	CI [1]	-6.3(0.9)	0.486 (70)
	RCC [2]		0.465(70)
	MCDHF [3]	-4.1(1.8)	0.444(75)

S. Raeder et al., Phys.Rev.Lett. 120, 232503 (2018)





Hyperfine structure partly resolved

Energy splitting

$$\Delta E_{HFS} = A \cdot \frac{C}{2} + B \cdot \frac{3/4C(C+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

$$C = F(F+1) - I(I-1) - J(J-1)$$

nuclear atomic properties

$$A = \mu \cdot \frac{B_e(0)}{IJ}$$

$$B = eQ_s \left\langle \frac{\delta^2 V}{\delta z^2} \right\rangle_{z=0}$$

experiment

	μ (μ_N)	Q_s (eb)
Laser spec. (this work)	-0.527(33)[75]	5.9(14)[8]

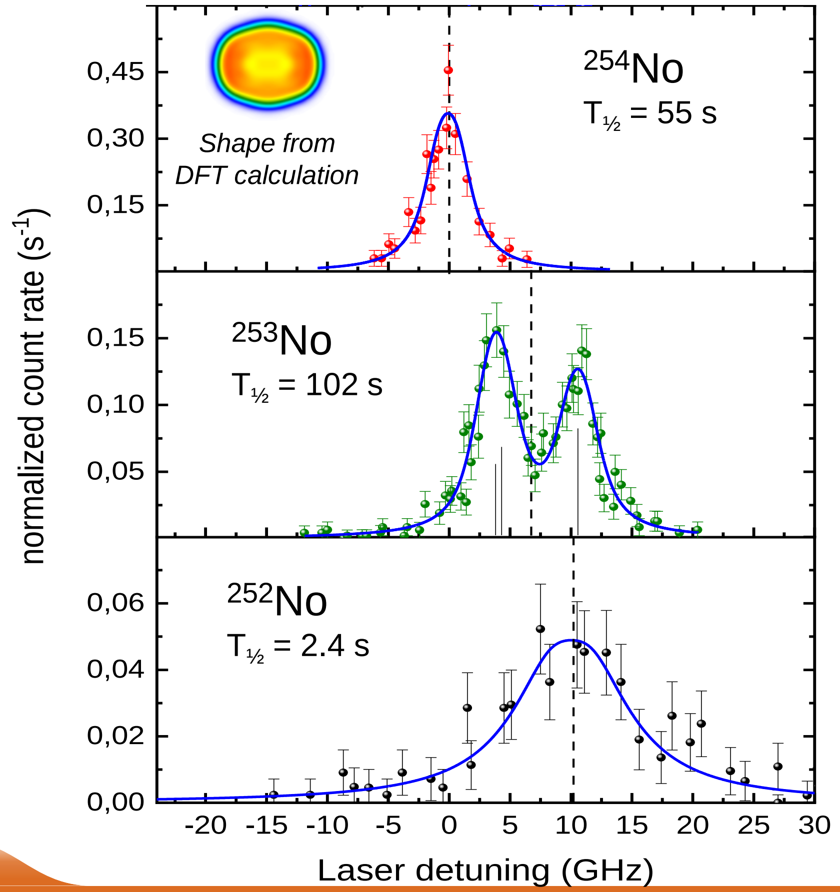
-0.593 (calculated value)

7.145 eb from ^{254}No

Isotope Shift of $^{252\text{-}254}$No & HFS in ^{253,255}No

- Isotope shift for $^{252\text{-}254}$No 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))



nuclear | atomic | properties

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

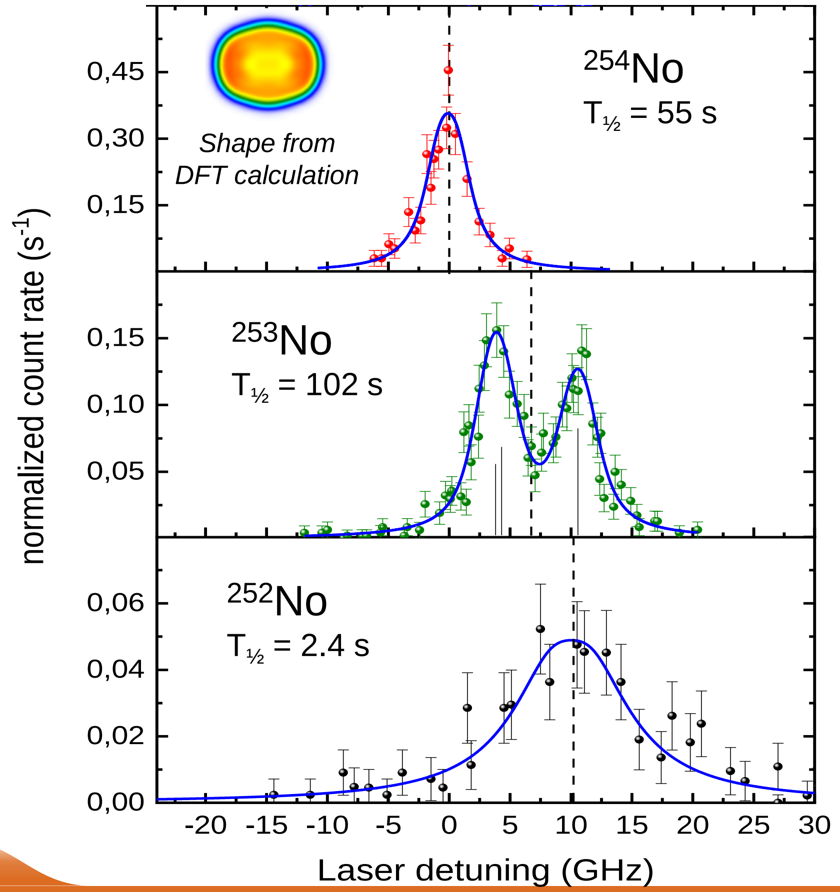
experiment

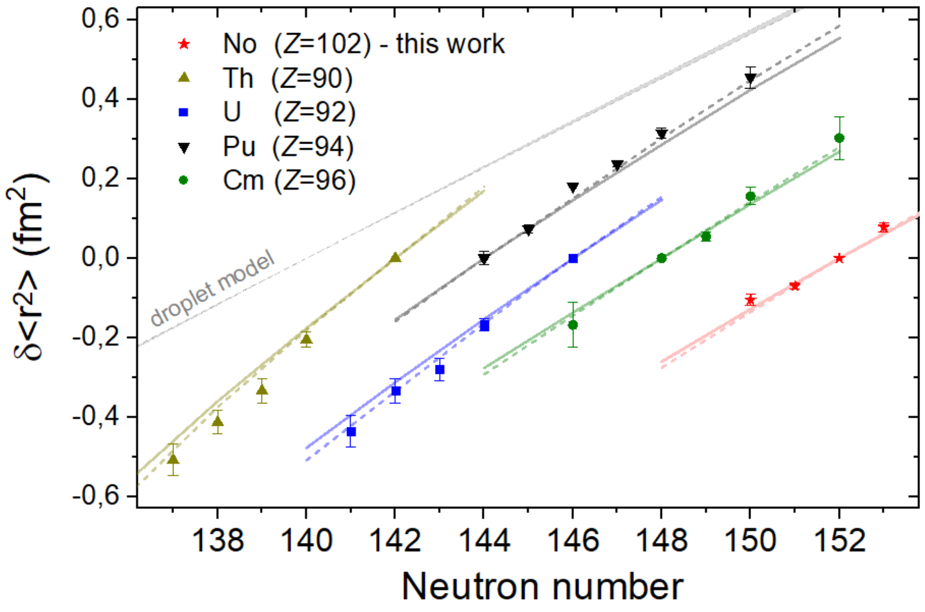
*S. Raeder et al., Phys. Rev. Lett. **120** (2018) 232503*

Isotope Shift of $^{252\text{-}254}No$ & HFS in $^{253,255}No$

- Isotope shift for $^{252\text{-}254}No$ 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))





Agrees well with nuclear DFT calculations

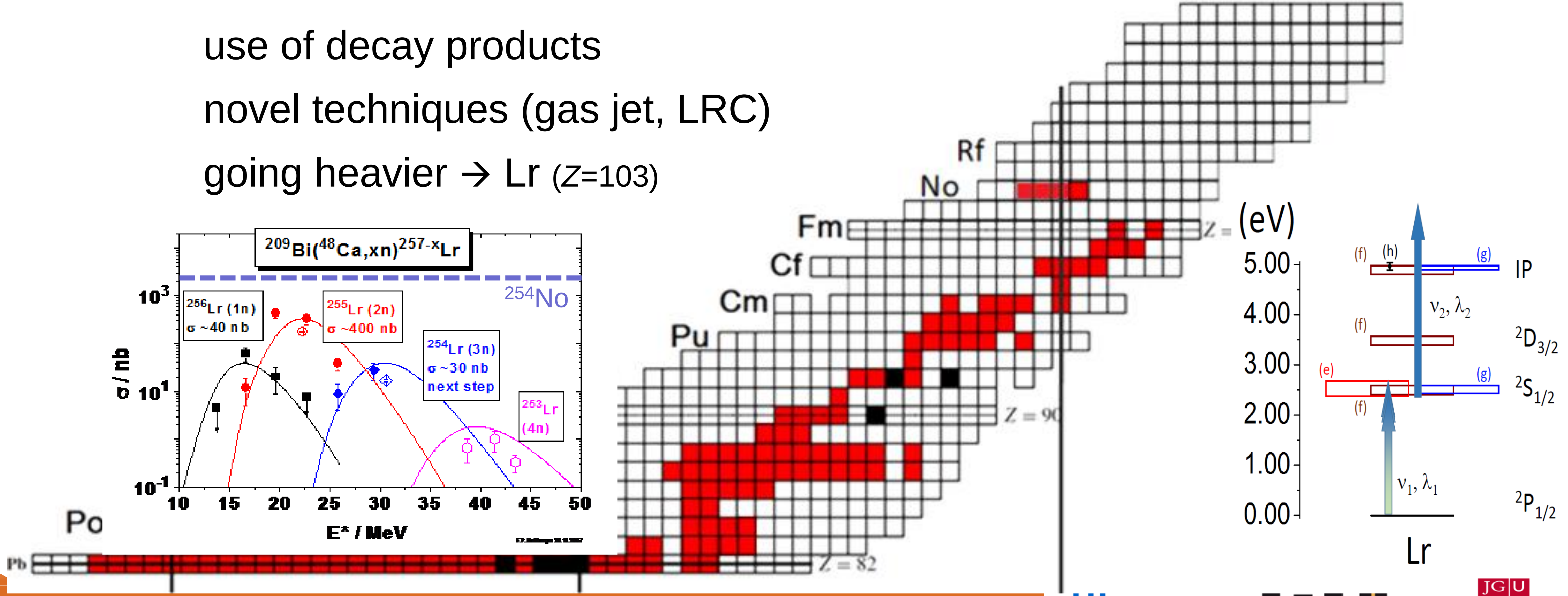
*S. Raeder et al., Phys. Rev. Lett. **120** (2018) 232503*

Optical spectroscopy

Laser spectroscopy advanced to transfermium elements

Advancements:

- use of decay products
- novel techniques (gas jet, LRC)
- going heavier → Lr (*Z*=103)

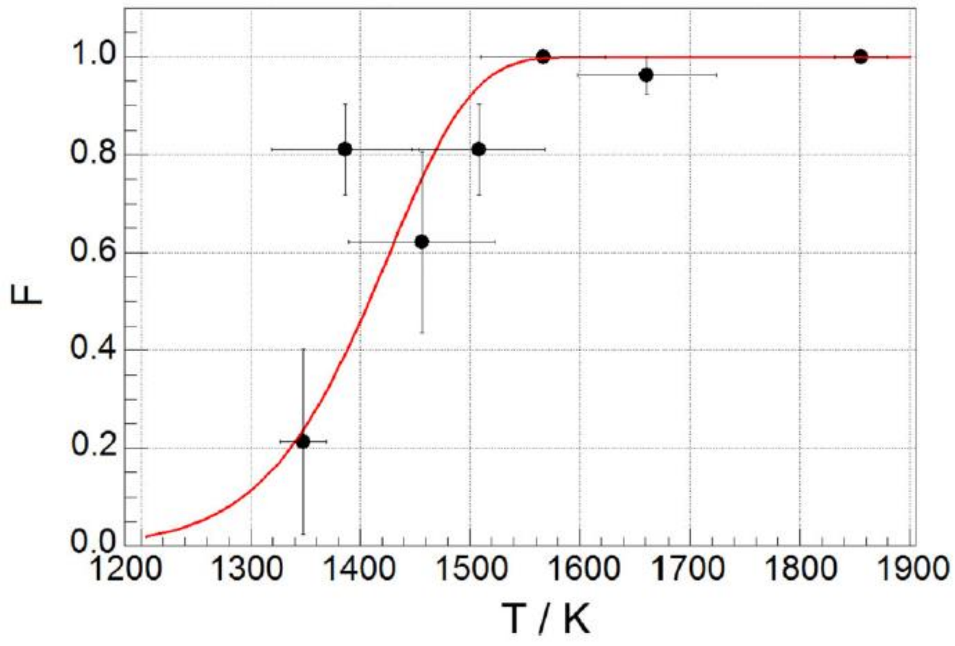


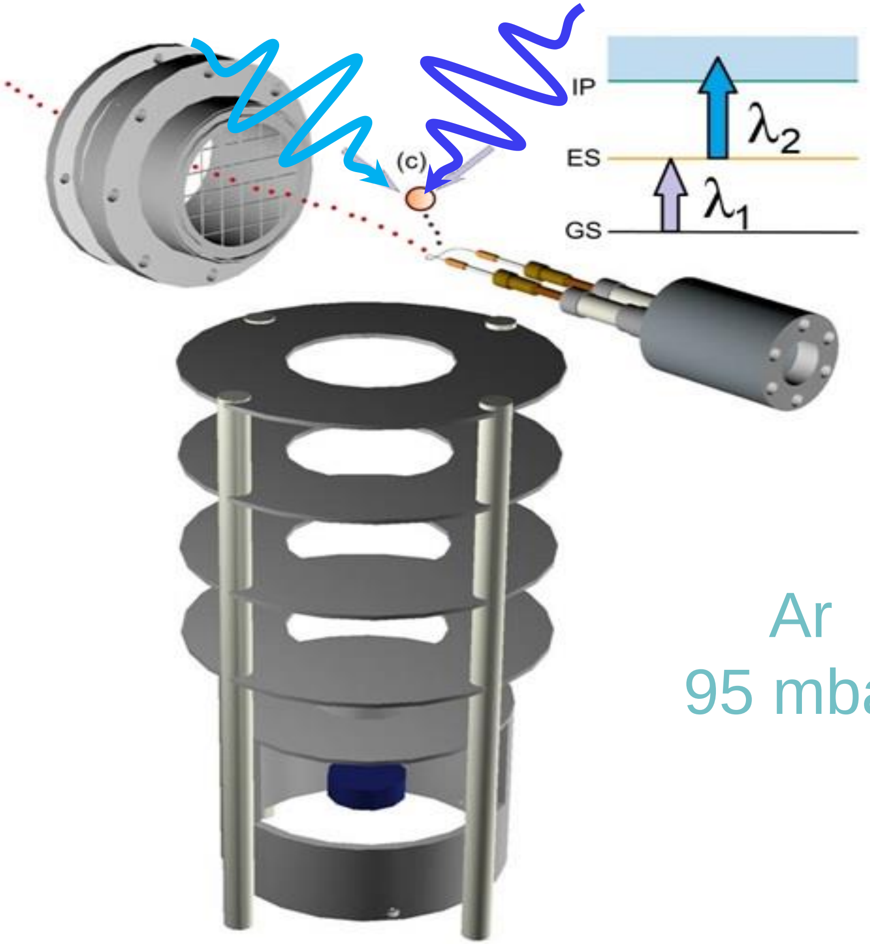
Radiation Detected Resonance Ionization Spectroscopy

(c) **Evaporation** and two-step photoionization process

^{254}No
51 s
α: 90 %
ε: 10 %

Desorption on-line





Ar
95 mbar

Desorption measurement but with ~1000 atoms per datapoint

Laatiaoui, M. et al. *Europ. Phys. J. D* 68.3 (2014): 1-6.

Physics and chemistry of the heaviest elements

Neutrons →

HIM HELMHOLTZ Helmholtz-Institut Mainz
GSI RESEARCH FOR GRAND CHALLENGES
JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ