Physics and chemistry of the heaviest elements

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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
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 $Z \geq 100$ only calculations are available

Blaise 1992

Experimental levels

Theory – calculated levels

Light actinides: analysis of fluorescence light

Spectrometry of light from discharge source

Macroscopic amount (mg…µg)

→ recording of images

Light actinides: analysis of fluorescence light

Table of transitions

+ strengths

+ magnetic field

⇒ Zeemann splitting

$J$-information

+ analysis of differences

⇒ level scheme
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

100Fm – 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

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

$T = 1 \text{ a}$

4 ng $^{255}$Fm ($t_{1/2} = 20 \text{ h}$) 

$(10^{12} \text{ atoms})$

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


Spectroscopic basis of RIS – atomic structure

- **Energy**
  - **Ground state** $E_0$
  - **First excited state** $E_1$
  - **Higher excited states** $E_2$

- **Ionization potential** $E_{IP}$

- **First excited state** $E_1$
  - Ionization potential $E_{IP}$
  - $10^{-17}$ cm$^2$

- **Higher excited states** $E_2$
  - $10^{-10}$ cm$^2$
  - $10^{-13}$ cm$^2$
  - $10^{-12}$ cm$^2$

- **Ionization via**
  - Black Body Rad.
  - IR Lasers
  - Collisions
  - Electrical Field

- **Energy levels**
  - 0 eV
  - ~6 eV (5-9 eV)
ionization potential ~6 eV

(5-9 eV)

higher excited states

~ 10^-17 cm^2

E_1

0 eV

ground state

e^-

E_0

~ 10^-10 cm^2

first excited state

~ 10^-10 cm^2

E_1

E_2

~ 10^-13 cm^2

Excitation of Rydberg-states

ionization via
- Black Body Rad.
- IR Lasers
- Collisions
- Electrical Field

ground state

+ Mass separation \(\rightarrow\) isotope selectivity

E_0

E_2

non-resonant ionization

auto-ionizing resonances

excitation of Rydberg-states

ionization potential

Energy

Resonant Laser Ionization

Mass separation

Ionization via

- Black Body Rad.
- IR Lasers
- Collisions
- Electrical Field

S. Raeder – 08.10.2021 – Lecture 2 - Joliot-Curie School – Isle d’Oleron
Sample Analysis with Laser Ionization

- Sample with actinide mixture and limited information

→ Lasers tuned to resonantly ionize different actinide elements

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Characterization of sample from ONRL

→ Trace analysis applications
Which laser system to choose?

Efficiency is important and requires

1. \( \sigma_{\text{Ion}} \cdot F \gg \beta \) → ionization rate \( \gg \) loss rate

2. \( \sigma_{\text{Ion}} \cdot \varphi \gg 1 \) → number of ionized atoms per laser interaction time (pulse)

\( \sigma_{\text{Ion}} \) ionization cross section (non-resonant) (cm\(^2\))

\( \beta \) loss rates to (metastable) states, state dependent

\( F \) photon flux (cm\(^{-2}\) s\(^{-1}\))

\( \varphi \) photon fluence (= photon flux \( \times \) laser interaction time)
Which laser system to choose?

### Efficiency is important and requires

1. \( \sigma_{\text{ion}} \cdot F \gg \beta \) → ionization rate » loss rate

2. \( \sigma_{\text{ion}} \cdot \varphi \gg 1 \) → number of ionized atoms per laser interaction time (pulse)

### Typical values:

- \( \sigma_{\text{ion}} \rightarrow 10^{-17} \text{ cm}^2 \)
- \( \beta \rightarrow 10^6 \text{ s}^{-1} \)

**Assumption:**
- laser beam area of 1 mm\(^2\)
- and photon energy of 3 eV.

### Flux

- From (1): \( \text{Flux } F \gg 10^{23} \text{ cm}^{-2}\text{s}^{-1} \)
  → # photons required >> 10\(^{21}\) /s
  → >> 500 W
  → Impossible

### Continuous Laser:

- \( \text{Continous Laser:} \)

### Pulsed Laser:

- \( \text{Pulsed Laser: (10 ns)} \)

- With a pulsed laser system: \( \text{(1)} \)
  → \( \gg 5 \mu\text{J/pulse} \)
  → No problem !!

- Lets add in the Fluence \( \text{(2)} \)
  → \( \gg 0.5 \text{ mJ/pulse} \)
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

https://www.larissa.physik.uni-mainz.de/
Ionization Potential of Ac

Rydberg Ritz formula

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

Weighted average value

\[ E_{IP} = 43\ 394.45(19) \text{ cm}^{-1} \]
Ionization potentials of the actinides

Electron affinity is also under study pushing towards actinide elements

Ionization potentials of the actinides

Heavy actinides: Surface ionization efficiency in a hot cavity

Ionization potentials of the actinides

Surface ionization efficiency in a hot cavity

\[
I_{\text{eff}} = \frac{N_{\exp} \left( \phi - \text{IP}_1^* \right)}{kT} \left[ 1 + N_{\exp} \frac{\phi - \text{IP}_1^*}{kT} \right]
\]

\[
\text{IP}_1^* = \text{IP}_1 - kT \ln \left( \frac{Q_i}{Q_0} \right)
\]

Access to effective IP

→ atomic theory input required

Fm-Lr (± 600 cm\(^{-1}\))


Surface ionization – Tokai
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

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

Model calculations

Production: Velocity Filter SHIP

Isotope of Interest:

$^{254}\text{No} \ (T_{1/2} = 55 \text{ s}; I=0)$

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

$\sigma = 2050 \text{ nb}$

$\rightarrow 17 \text{ s}^{-1}@\text{Target}$

$^{48}\text{Ca}
4.55 \text{ MeV/u}
1 \text{ p\mu A}$
(a) Stopping of the incoming fusion products
Radiation Detected Resonance Ionization Spectroscopy

(a) Stopping of the incoming fusion products
(b) Collecting onto thin tantalum wire
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
(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

F. Lautenschläger et al., NIMB 383, 115 (2016)
Lab impressions

Laser system

Gas cell

Dye lasers

Excimer lasers

SHIP

Glass fibre

Interaction volume

Filament

Bilder: GSI Helmholtzzentrum für Schwerionenforschung
Level Search in $^{254}$No

<table>
<thead>
<tr>
<th>Year</th>
<th>2007</th>
<th>2014</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scan range (cm$^{-1}$)</td>
<td>25920 – 31001</td>
<td>28887 – 33191</td>
</tr>
<tr>
<td>Net scan time (h)</td>
<td>39</td>
<td>67</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th></th>
<th>$\nu_1$ (cm$^{-1}$)</th>
<th>$A_{ki}$ (s$^{-1}$) $\times 10^8$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiment [1]</td>
<td>29,961.457(7) stat</td>
<td>4.2 (2.6) stat</td>
</tr>
<tr>
<td>IHFSCC [2]</td>
<td>30,100(800)</td>
<td>5.0</td>
</tr>
<tr>
<td>MCDF [3]</td>
<td>30,650(800)</td>
<td>2.7</td>
</tr>
</tbody>
</table>

Agrees with predicted $^1S_0 \rightarrow ^1P_1$ transition

Independent lifetime determination by delayed ionization

No

\[ T_{\frac{1}{2}} > 50 \text{ ns} \] – suggest much weaker optical transition

Yb

\[ T_{\frac{1}{2}} > 50 \text{ ns} \] – suggest much weaker optical transition

\[ \lambda_1 = 333.8 \text{ nm} \]

\[ \lambda_1 = 351 \text{ nm} \]

\[ t_d \]

\[ \text{IP} \]

\[ ^1P_1 \]

\[ ^3S_0 \]

\[ 5 \text{ mbar} \]

\[ 100 \text{ mbar} \]
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?

$\frac{1}{2} A_{kl} S(\omega_L, \omega_{12})$
Gas induced quenching

Excitation to higher lying levels $\rightarrow$ Rydberg states

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


Dependence of the extracted IP from the gas pressure
Observation of Rydberg states with two-step resonance ionization

- Identification 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:

\[ E_n = E_{1p} - \frac{R_\mu}{[n - \delta(n)]^2} \]

<table>
<thead>
<tr>
<th>IP (eV)</th>
<th>Experiment</th>
<th>IHFSCC [1]</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.6261(3)</td>
<td></td>
<td>6.632</td>
</tr>
</tbody>
</table>

Hyperfine Structure Studies in $^{253}\text{No}$

Hyperfine structure partly resolved

Energy splitting

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

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

**Feedback from atomic theory** for nuclear moments

<table>
<thead>
<tr>
<th>Theory</th>
<th>Ref.</th>
<th>$A/\mu_N$ (GHZ)</th>
<th>$B/\text{eb}$ (GHZ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CI [1]</td>
<td>-6.3(0.9)</td>
<td>0.486 (70)</td>
<td></td>
</tr>
<tr>
<td>RCC [2]</td>
<td>0.465(70)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>MCDHF [3]</td>
<td>-4.1(1.8)</td>
<td>0.444(75)</td>
<td></td>
</tr>
</tbody>
</table>

Hyperfine Structure Studies in $^{253}\text{No}$

The hyperfine structure partly resolved.

Energy splitting

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

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

<table>
<thead>
<tr>
<th>Laser spec. (this work)</th>
<th>$\mu$ ($\mu_N$)</th>
<th>$Q_s$ (eb)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>-0.527(33)[75]</td>
<td>5.9(14)[8]</td>
</tr>
<tr>
<td>(calculated value)</td>
<td>-0.593</td>
<td>7.145 eb</td>
</tr>
</tbody>
</table>

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

$B = eQ_s \left[ \frac{\delta^2 V}{\delta z^2} \right]_{z=0}$

atomic
nuclear
properties

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

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

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

\begin{align*}
\text{252}\text{No} & \quad T_{1/2} = 2.4 \text{ s} \\
\text{253}\text{No} & \quad T_{1/2} = 102 \text{ s} \\
\text{254}\text{No} & \quad T_{1/2} = 55 \text{ s}
\end{align*}
Isotope Shift of $^{252-254}$No & HFS in $^{253,255}$No

- Isotope shift for $^{252-254}$No measured
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(R. Beenwerth & S. Fritzsche (MCDF), V. Dzuba, M. Safranove (CI), A. Borschevsky (RCC))

Agrees well with nuclear DFT calculations

Shape from DFT calculation

$^{254}$No
$T_{1/2} = 55$ s

$^{253}$No
$T_{1/2} = 102$ s

$^{252}$No
$T_{1/2} = 2.4$ s

normalized count rate (s$^{-1}$)

Laser detuning (GHz)

Normalized Counts

Optical spectroscopy

Laser spectroscopy advanced to transfermium elements

Advancements:

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

P. Campbell et al., Prog. Part Nucl. Phys 86 (2016) 127
(c) **Evaporation** and two-step photoionization process

Desorption on-line

![Graph showing desorption measurements](image)

- **$^{254}$No**
- **51 s**
- **$\alpha$: 90 %**
- **$\epsilon$: 10 %**

Desorption measurement but with ~1000 atoms per datapoint

- **Ar**
- **95 mbar**

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