MAINZ PHYSICS ACADEMY (MPA) Retreat 2024
Höchst, 02.10.2024

Tailored (Molecular) Actinide Ions for Fundamental Physics Experiments around the World

J. Stricker[1,2,3], P. Fischer[4], J. Velten[1], D. Renisch[1,2,3], L. Schweikhard[4], Ch. E. Düllmann[1,2,3,5]
and the TACTICa Collaboration[1,2,3,5]

[1] Department of chemistry – TRIGA site, JGU Mainz, 55099 Mainz, Germany;
[2] Helmholtz Institute Mainz, 55099 Mainz, Germany;
[3] PRISMA+ Cluster of Excellence, JGU Mainz, 55099 Mainz, Germany;
[4]Institute for Physics, university of Greifswald, 17487 Greifswald, Germany;
[5] GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany.

Author contact: jostrick@uni-mainz.de

DFG Deutsche Forschungsgemeinschaft

MPA MAINZ PHYSICS ACADEMY

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JG|U

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TACTICa

European Commission

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EURO-LABS EUROPEAN LABORATORIES FOR ACCELERATOR BASED SCIENCES

D. Budker, F. Schmidt-Kaler and L. von der Wense

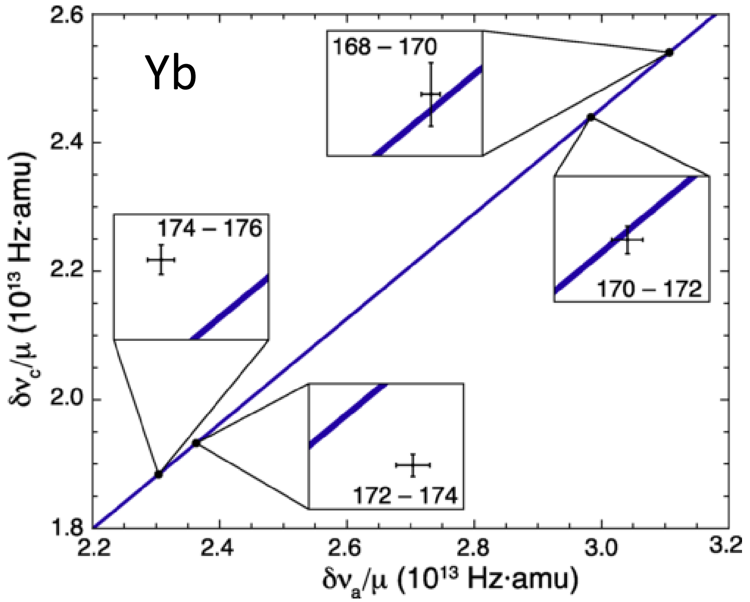
TACTICa Project

Trapping **A**nd **C**ooling **T**horium **I**on spectroscopy via **Ca**lcium

Th 228	Th 229	Th 230	Th 231	Th 232	Th 234
1.91 a	7µs | 7920 a	$7.54 \cdot 10^4$ a	25.5 h	$1.40 \cdot 10^{10}$ a	24.10 d

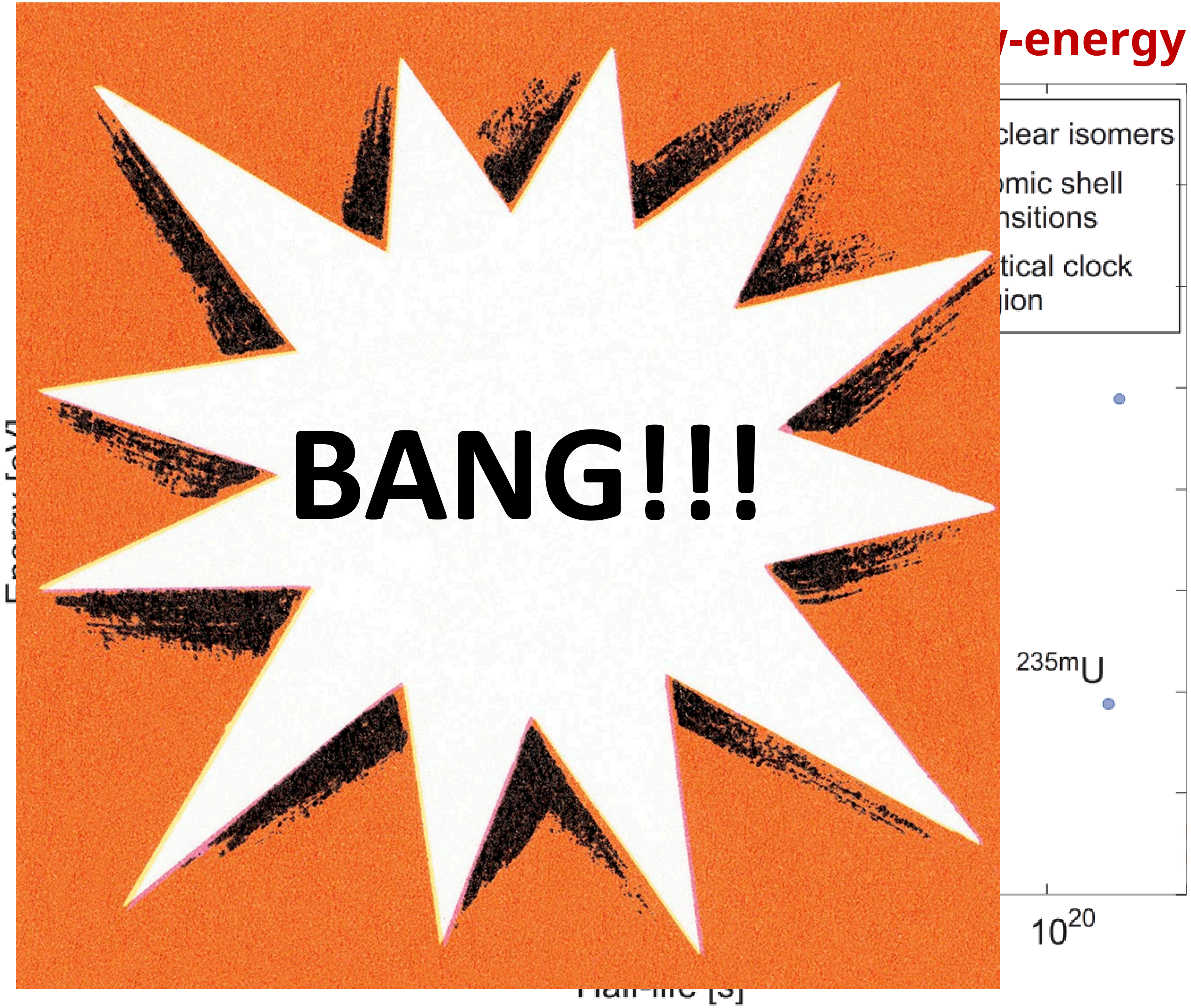
On the hunt of beyond standard model physics:

- Precise hyperfine structure measurements
  → ions near ground state
- King plot of the isotopic chain
  → deviation in the linearity of the isotopic shift?



N. L. Figueroa et al. *Phys. Rev. Lett.* **2022** 128, 073001.

-energy frontier



[1] L. von der Wense et al, *Nature* **2016**, 132, 182501.
[2] J. Tideau et al, *Phys. Rev. Lett.* **2024**, 132, 182501.
[3] V. V. Flambaum, *Phys. Rev. C.* **2019**, 99, 035501.

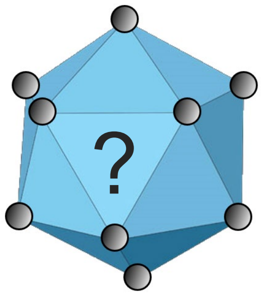
Interdisciplinary use of molecular Th ions

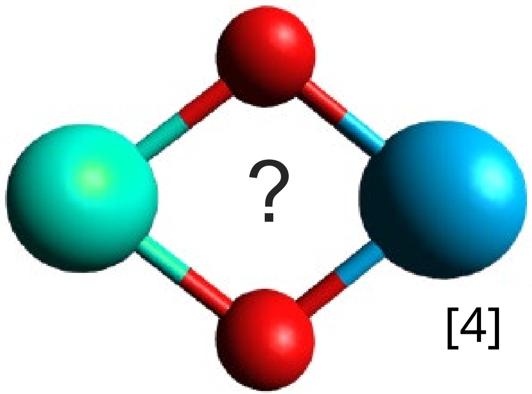
Th 228	Th 229	Th 230	Th 231	Th 232	Th 234
1.91 a	7μs | 7920 a	$7.54 \cdot 10^4$ a	25.5 h	$1.40 \cdot 10^{10}$ a	24.10 d

+ ThF ThO ThOH [3]

Chemistry:

- Enhance the understanding of laser ablation of thorium and actinides
- Enhance the understanding of thorium gas phase chemistry
- Research on gas phase actinide – actinide bonding

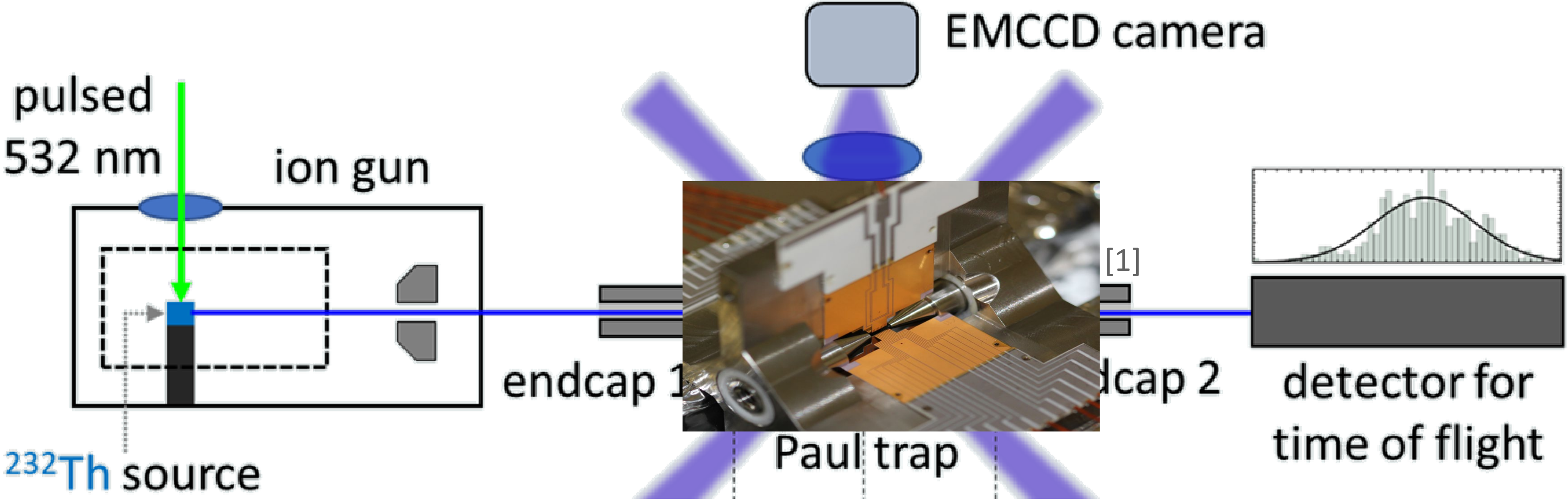




[4]

[1] L. von der Wense et al, *Nature* **2016**, 132, 182501.
[2] J. Tideau et al, *Phys. Rev. Lett.* **2024**, 132, 182501.
[3] V. V. Flambaum, *Phys. Rev. C.* **2019**, 99, 035501.
[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Experimental setup: Paul trap

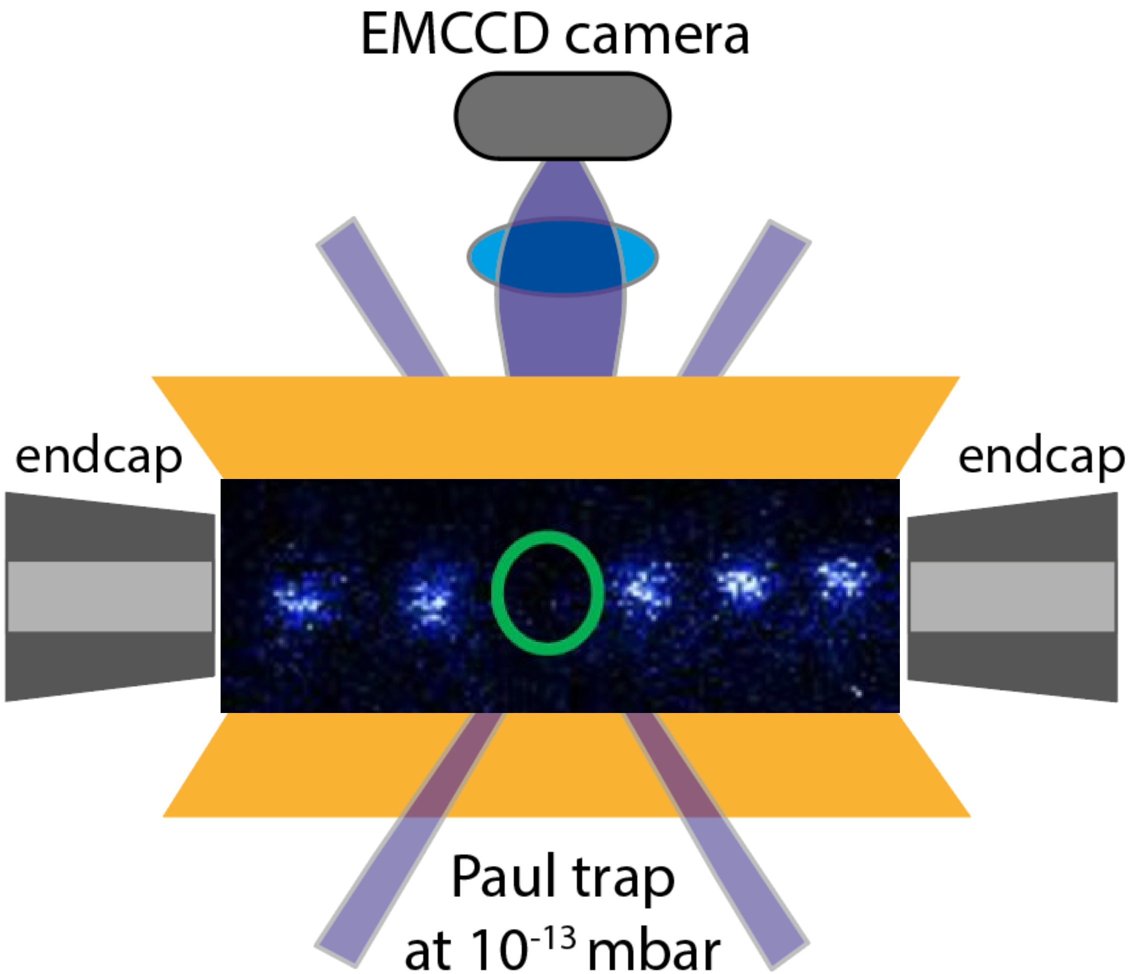


- Laser ablation of $^{232}Th^{+}$ in modified ion gun [SPECS IQE 12/38]
- Trapping, cooling and visualization of Th through $^{40}Ca^{+}$ ion crystal
- One laser system for quantum logic spectroscopy of every Th isotope
- Identification via time of flight (ToF) and optical spectroscopy [2; 3]

[1] G. Jacob, *Dissertation* **2016**, JGU Mainz.
[2] K. Groot-Berning et al., *Phys. Rev. A.* **2019**, 99, 023420.
[3] F. Stopp et al., *Hyp. Int.* **2019**, 240, 33.

Quantum logic spectroscopy of Th ions

- $^{232}Th^{+}$ in linear Paul trap [2]
- Th is visible through void in Ca^{+} crystal
- Permanent ultra cold crystal (µK regime)
    - → Suppresses thermic sideband broadening
    - → Hyperfine (sideband) spectroscopy
- Excite thorium → read out crystal
    - → Quantum logic spectroscopy

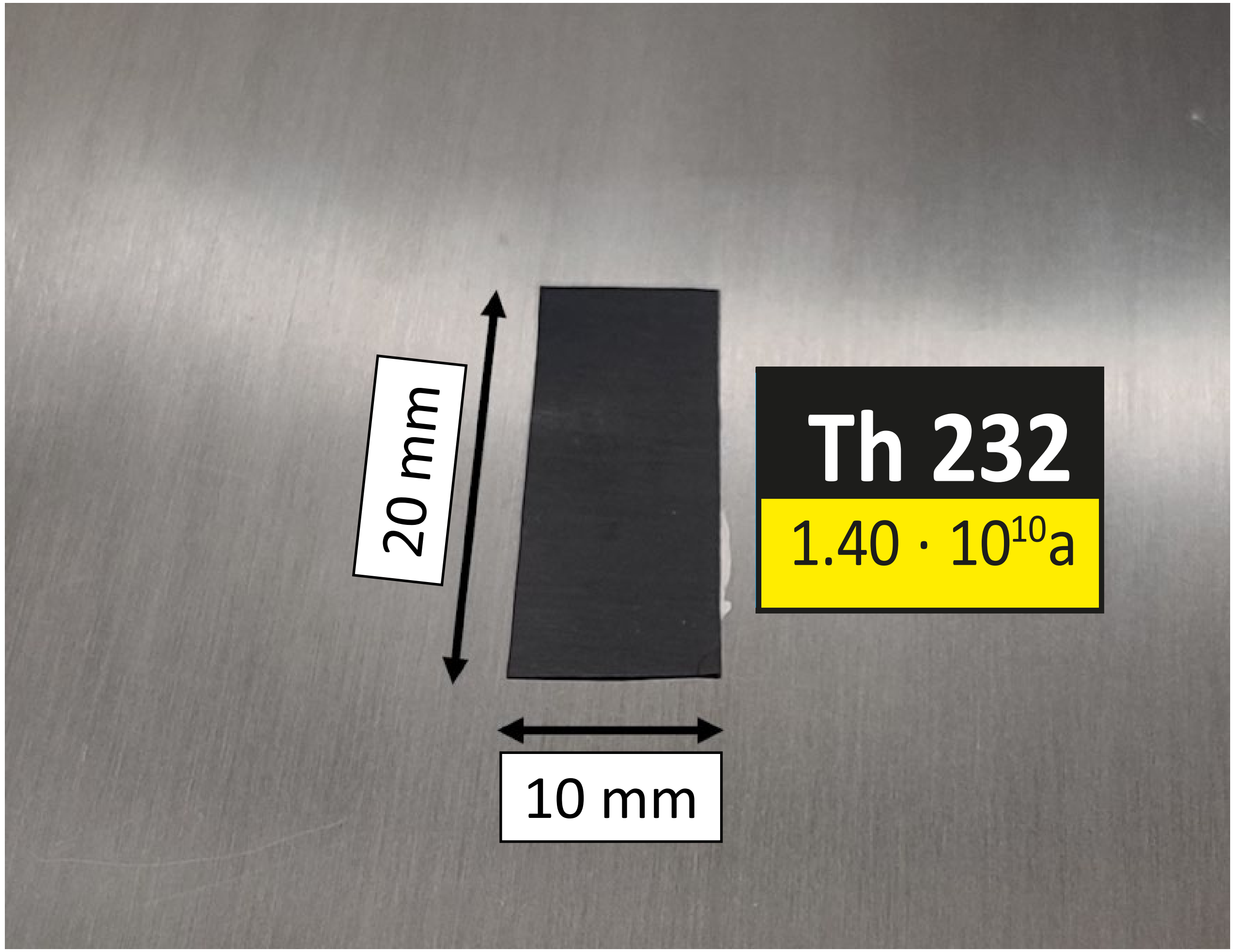


[2] K. Groot-Berning et al., *Phys. Rev. A.* **2019**, 99, 023420.

2. Ablation from a legacy metallic Th-foil

Origin:
- Produced in USA
- Shipped to CERN in 1987
- Shipped to univ. Mainz in 1988
- Targets shipped for various physics experiments all over the world
- Shipped to univ. Greifswald in 2023

Laser-ablation target:
- 89-micrometer ^{232}Th foil
- Heavily oxidized
- Glued to target holder
- 250 mg = 1 kBq



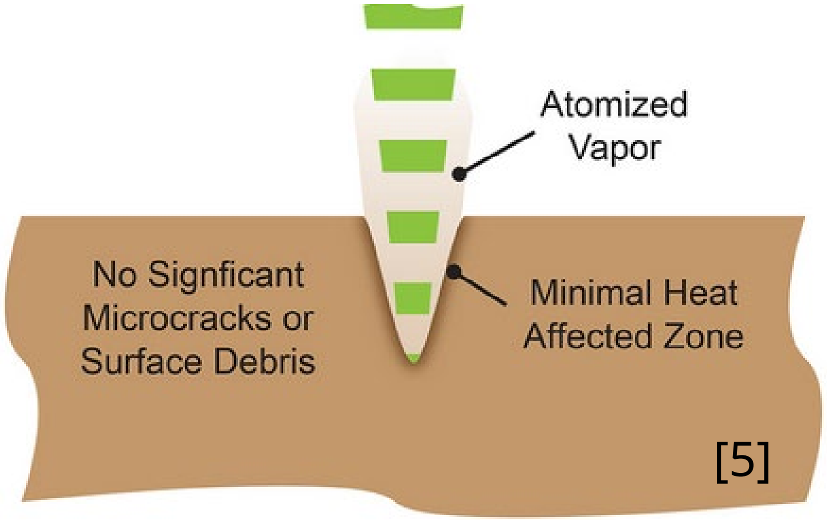
[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Production of tailored (molecular) ions

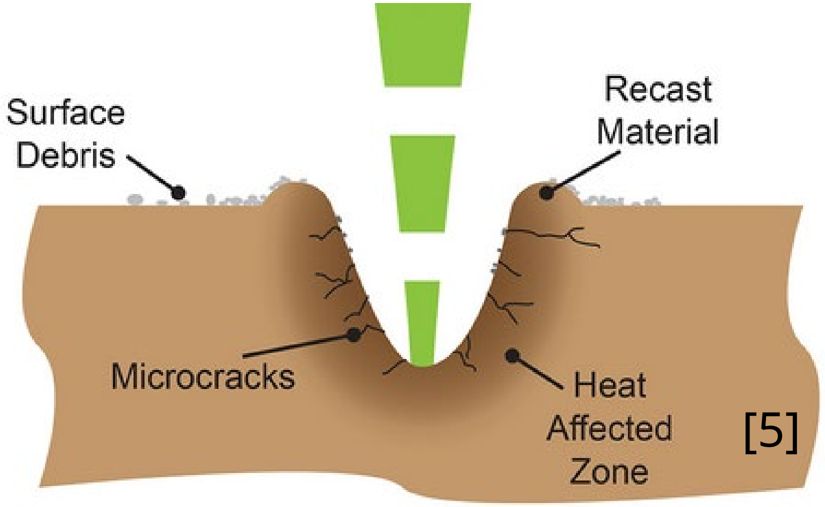
Direct production of (molecular) ions from metallic samples:

- Laser ablation [4] = evaporating (and ionizing) of a material with laser beam

Athermic ablation
(Pulse duration < 10 ns)



Thermic ablation
(Pulse duration > 10 ns)



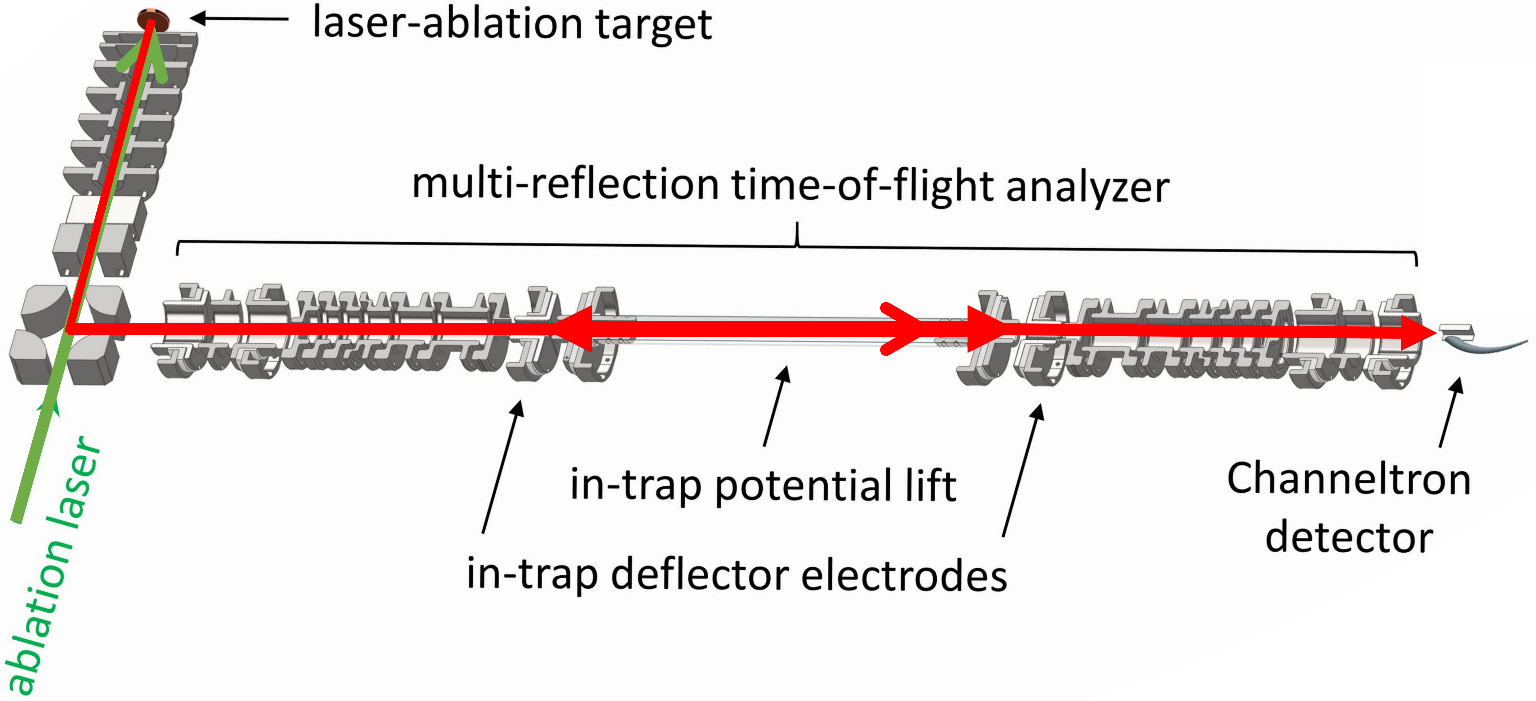
Production of **molecular ions in-flight**:

- Laser ablation coupled with gas guidance [5]
  → Reaction of ablated ions inside of buffer- and reaction-gas mixture
  → Only charge states of < 3 possible (only doable with ion – ion collisions)

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.
[5] Coherent; https://www.coherent.com/de/news/glossary/laser-ablation 2024.
[6] J.T. Stewart et al., *J. Mol. Spectrosc.* **2016** 322, 18–21.

The Greifswald multi reflection-ToF MS

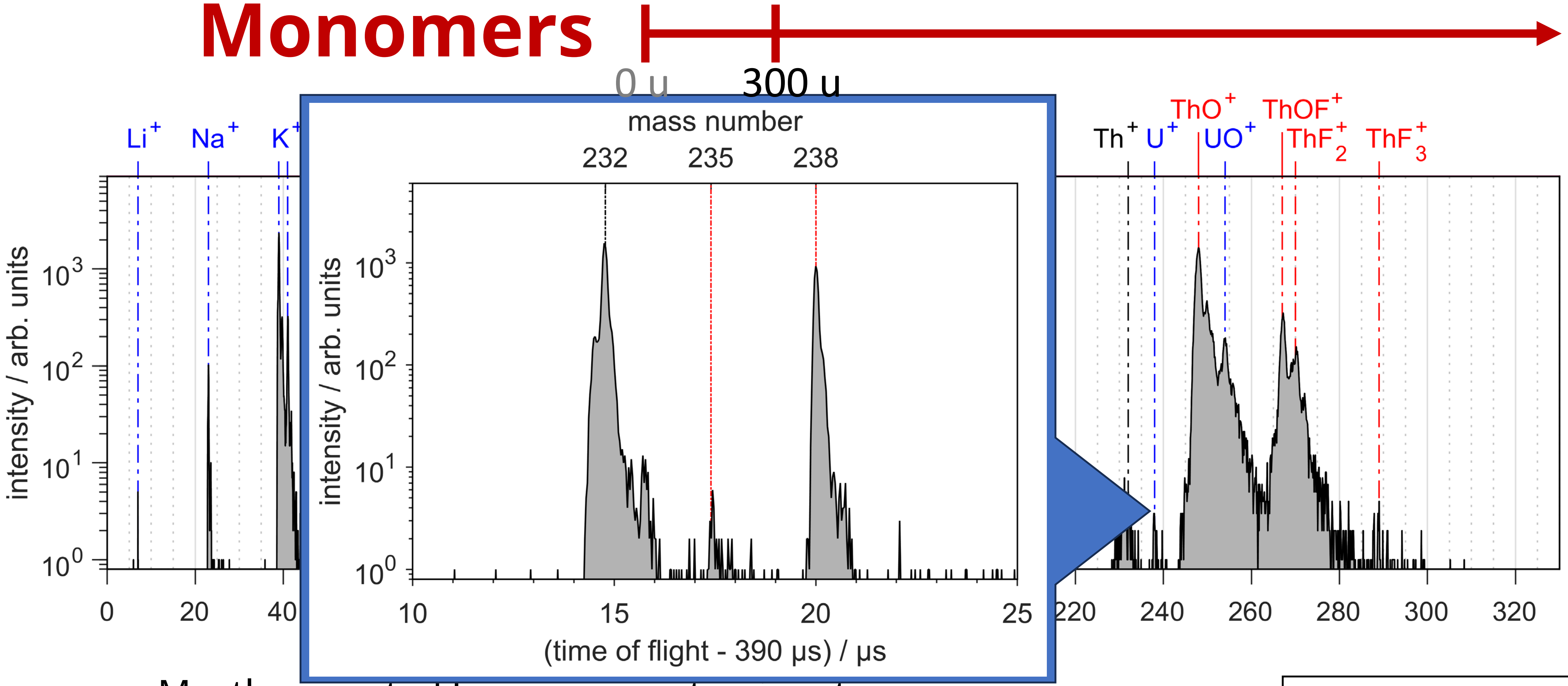
- High-vacuum laser-ablation source: 532 nm, 10 ns, Ø2 mm on target, < 2 mJ/pulse
- Biased with an electrostatic potential for acceleration of anionic or cationic ions
- Multi reflection of ions through ion lenses for longer flight path
- Flight paths of kilometers in meter long apparatus (m/Δm > 100.000; Th-232: 232.038(2) u)



overview of full setup:
P. F. Giesel et al., *Rev. Sci. Instrum.* 95:023201 (2024)

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Monomers



- Mostly expected low-mass contaminants
- Atomic thorium ions found as well as thorium oxides, fluorides and oxyfluorides found
- Found amounts of natural uranium → Foil produced 1950s?

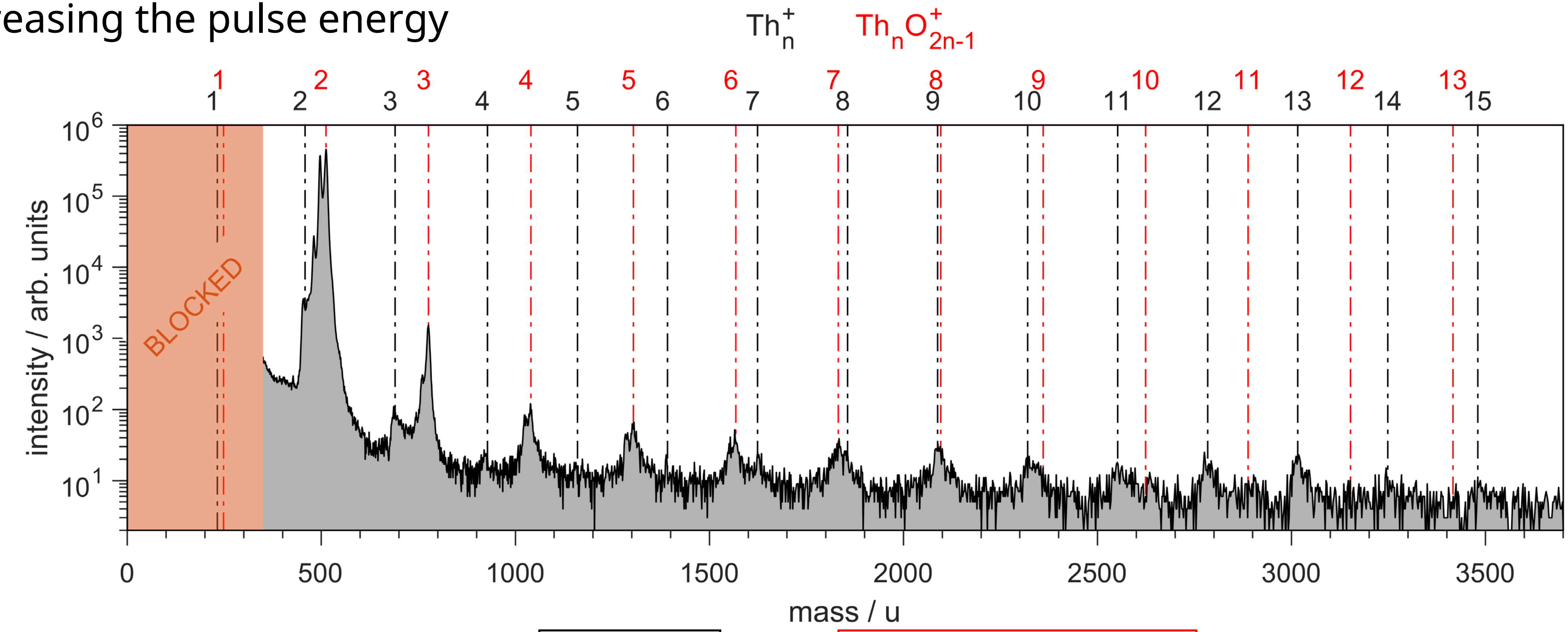
labeled species are confirmed by precision mass measurements

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

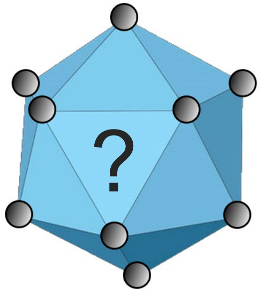
Clusters

0 u — 300 u — 520 u — 780 u — – 3500 u

- Increasing the pulse energy



- Description requires two series: pure Th_n^+ and oxidized $Th_nO_{2n-1}^+$
- Smaller species ($n \lesssim 8$) are predominantly found as oxides, larger as pure clusters
- Production drops above Th_{13}^+ → magic cluster size ?



[7] P. Fischer, J. Stricker et al., in preparation **2024**.

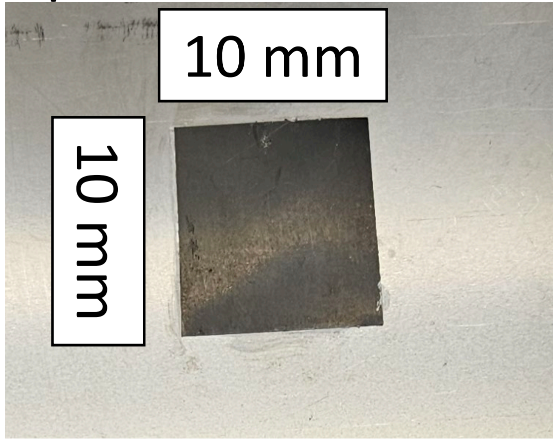
Uranium

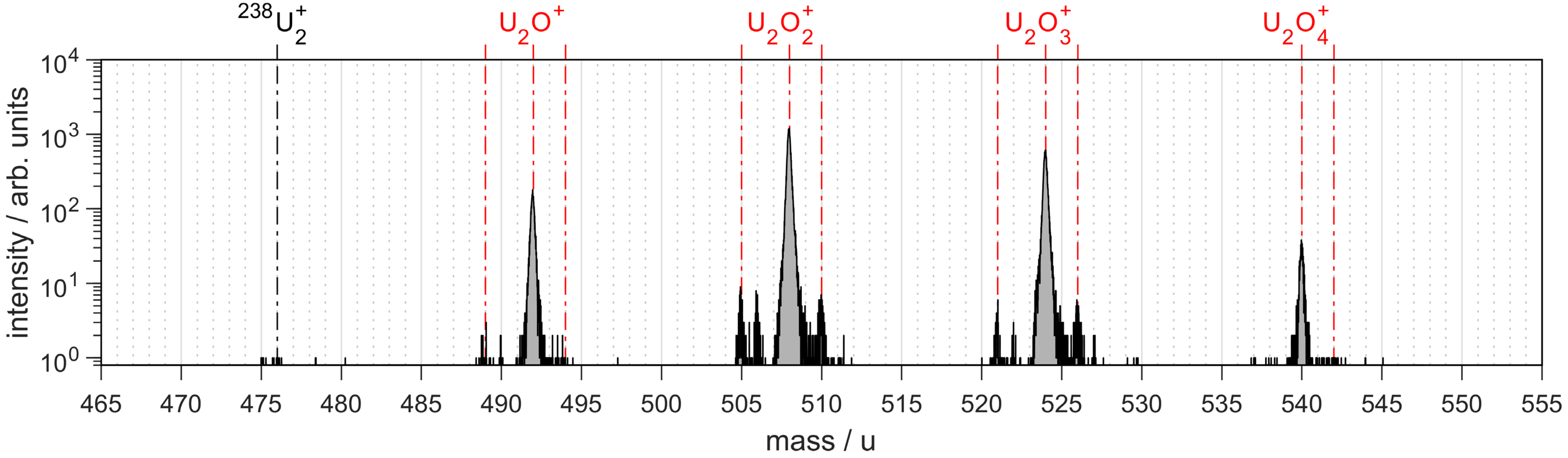
- U-238 (760 mg = 9.5 kBq) → no evidence of clusters in gas phase

Next Steps:

U 238

4.47 $\cdot 10^9$ a





[10] J. Stricker, P. Fischer et al., in preparation **2025**.

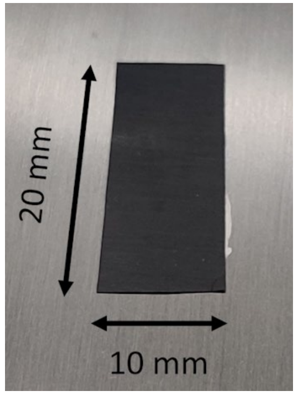
3. Research on laser ablation of Th salts

- Research on laser ablation behavior of thorium salts with ToF-MS [4]
- Th isotopes other than Th-232 only available as salts
- Parameter studies for different spot size and laser power
- Research on interplay of laser ablation and chemically different Th salts

Pulsed solid state diode laser:

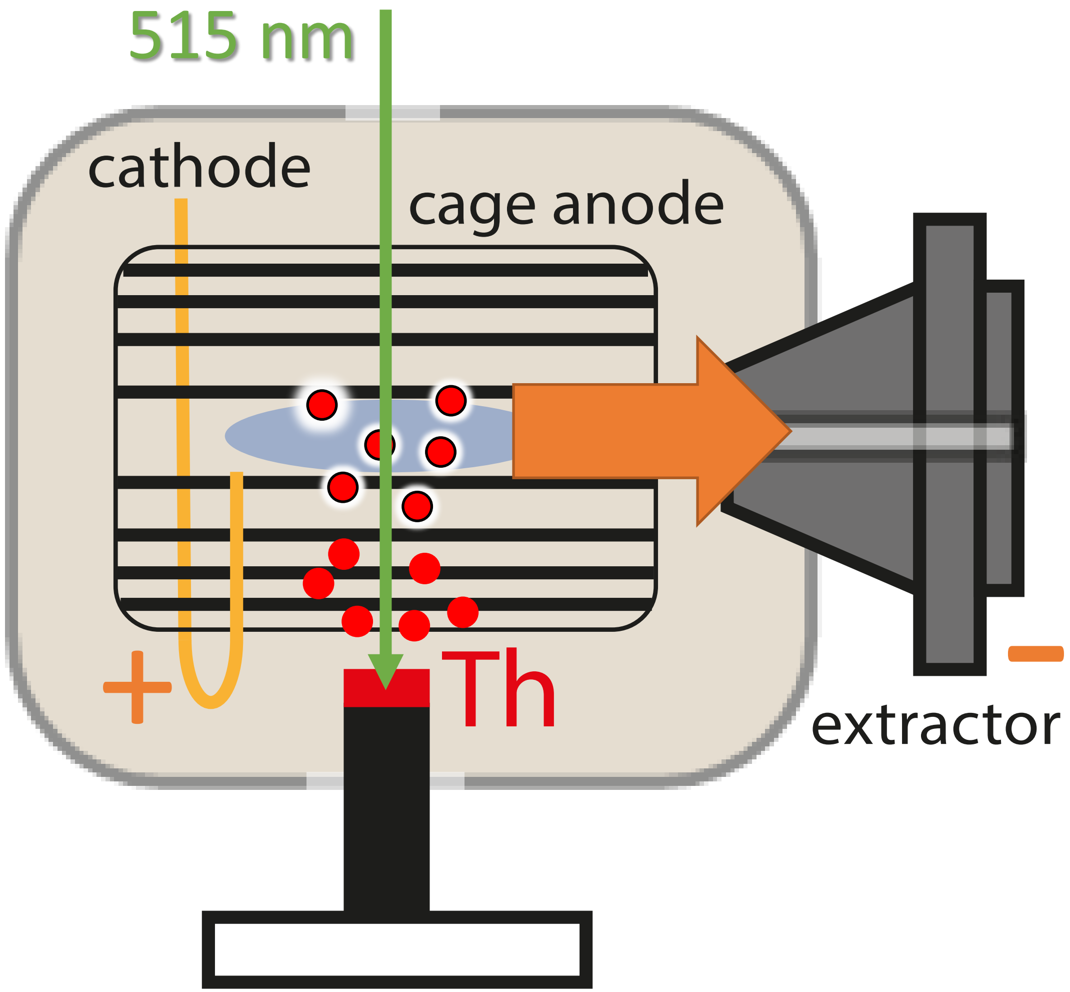
- Coherent Flare NX
- Wavelength λ = 515 nm
- Pulse energy E = 50 – 330 µJ
- Min. spot size S = 70 µm
- Pulse width t = 1.42 ns

Th 228	Th 229	Th 230	Th 232
1.91 a	7µs / 7920 a	$7.54 \cdot 10^4$a	$1.40 \cdot 10^{10}$a



[8] J. Stricker, *Master's thesis* **2023**, JGU Mainz.

Ionization of thorium



New ionization method:

1. Electron cloud inside cage anode
2. Ablation of neutral atoms
3. Ionization by electron impact
4. Acceleration by extraction voltage

Results in:

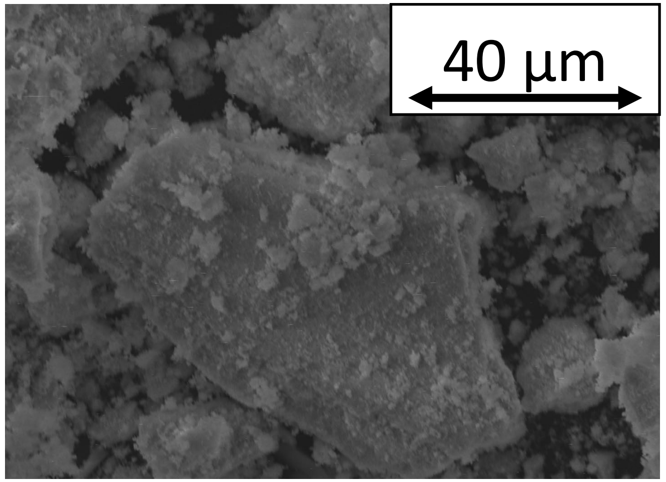
→ Ablation of metallic and salt-based samples

→ Tunable kin. energies 0.2 – 5 keV

→ Ultra high vacuum down to 10^{-10} mbar

[8] J. Stricker, *Master's thesis* **2023**, JGU Mainz.

Micro synthesis of thorium fluoride



Synthesis of $^{232}ThF_4$ in microgram scale:

- Reaction of 10 µg $Th(NO_3)_4$ with hydrofluoric acid:

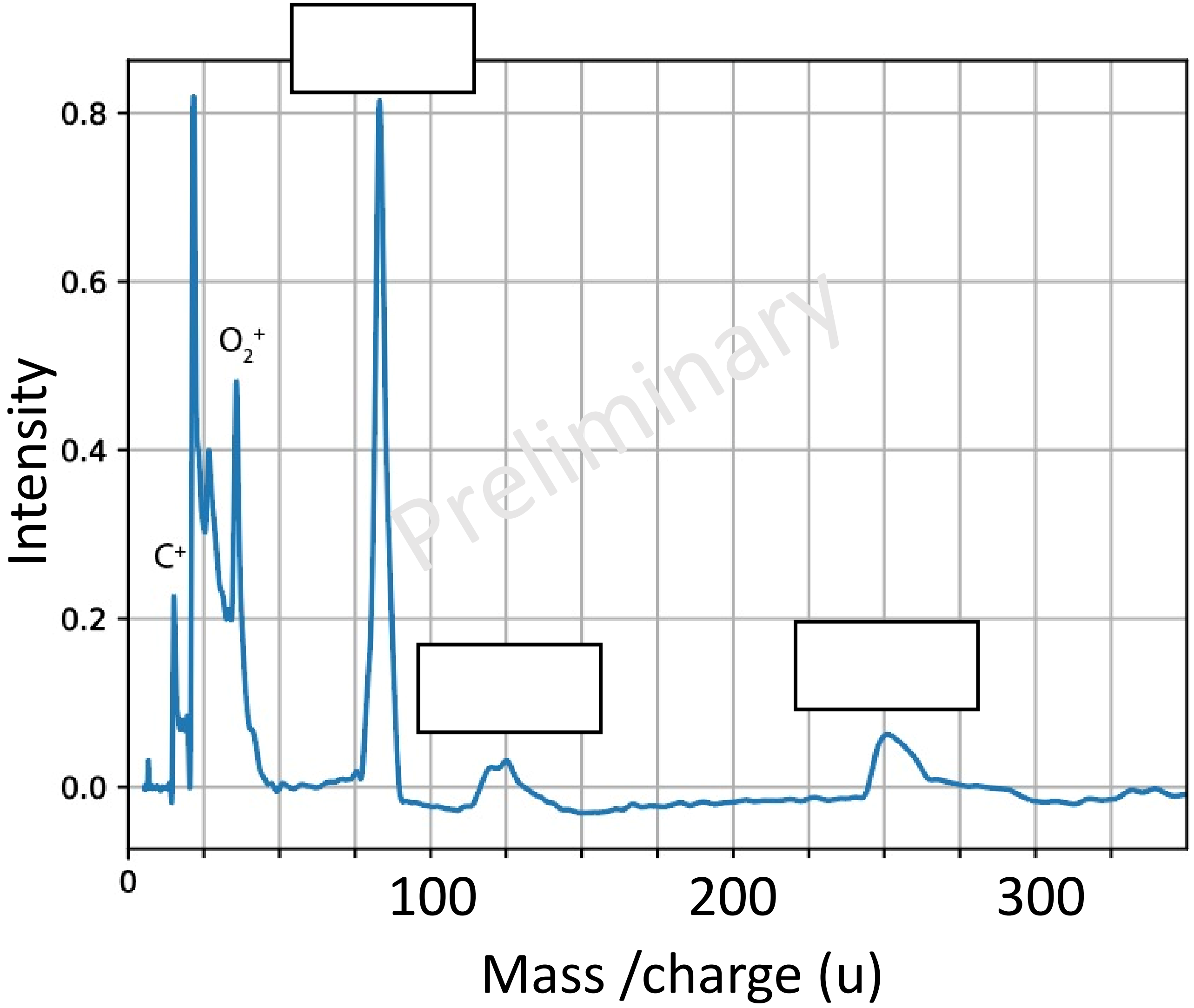
$$Th(NO_3)_4(s) + 4\ HF(aq.) \xrightarrow[10\ min]{\Delta} ThF_4(s) + 4\ HNO_3(g)\uparrow$$

- Direct synthesis on target holder
  → Thorium fluoride is inert
  → Reduce loss of product
- Quantitative reaction and oxygen free product
  → Analyzed by:
  - scanning electron microscopy
  - energy dispersive X-ray spectroscopy
  - X-ray diffraction (by O. Walter @ JRC Karlsruhe)

[10] J. Stricker et al., in preparation **2024**.

Ablation of thorium fluoride

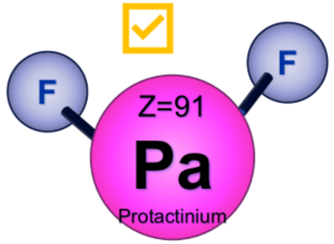
- Measured ThF_4
- Direct production of Th molecules
- Th molecular ions in **higher charge states**!
  → **ThF^{2+}**
- New method to produce **triply-charged cations**
  → **ThF^{3+}**
- No target or gas phase reaction
  → **Higher charge state retains after ablation**



[10] J. Stricker et al., in preparation **2024**.

4. Summary

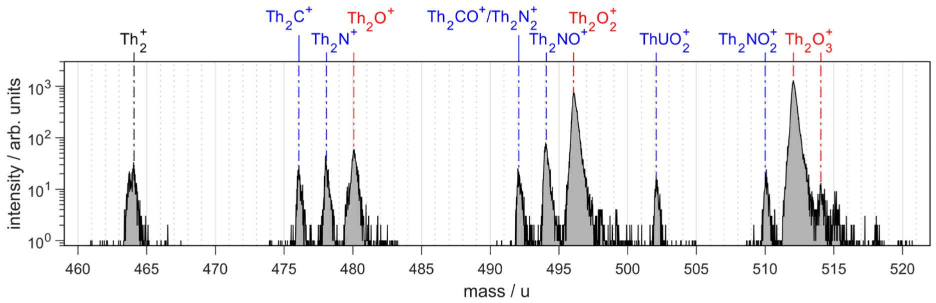
- Production of protactinium ion beam at ISOLDE CERN
- Laser ablation of metallic thorium and uranium in Greifswald

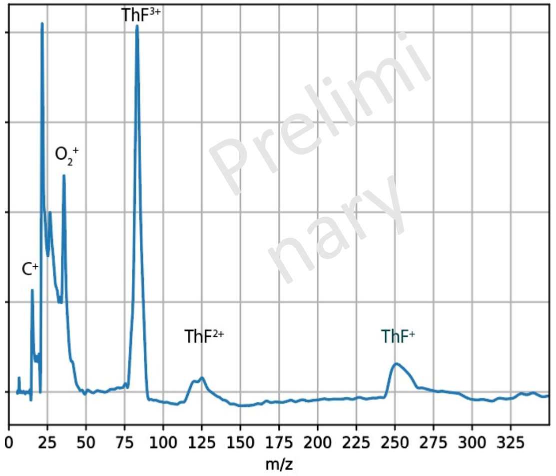


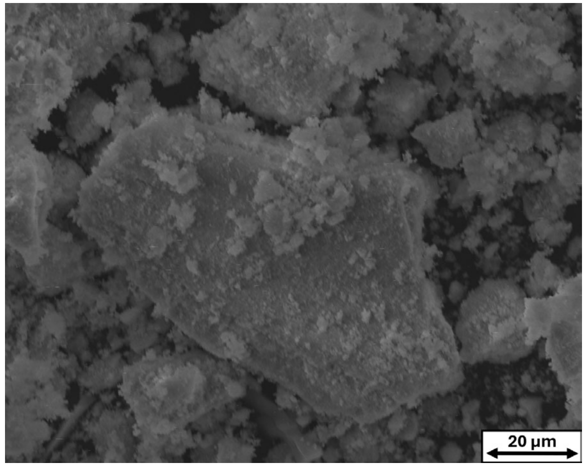
(from offline Pa source)

- Micro syntheses and ablation of thorium ions from salt targets in Mainz
- Molecules in higher charge states due to new ionization method

→ **Production of tailored molecular Th ions for testing the standard model**







Thank you for your attention!

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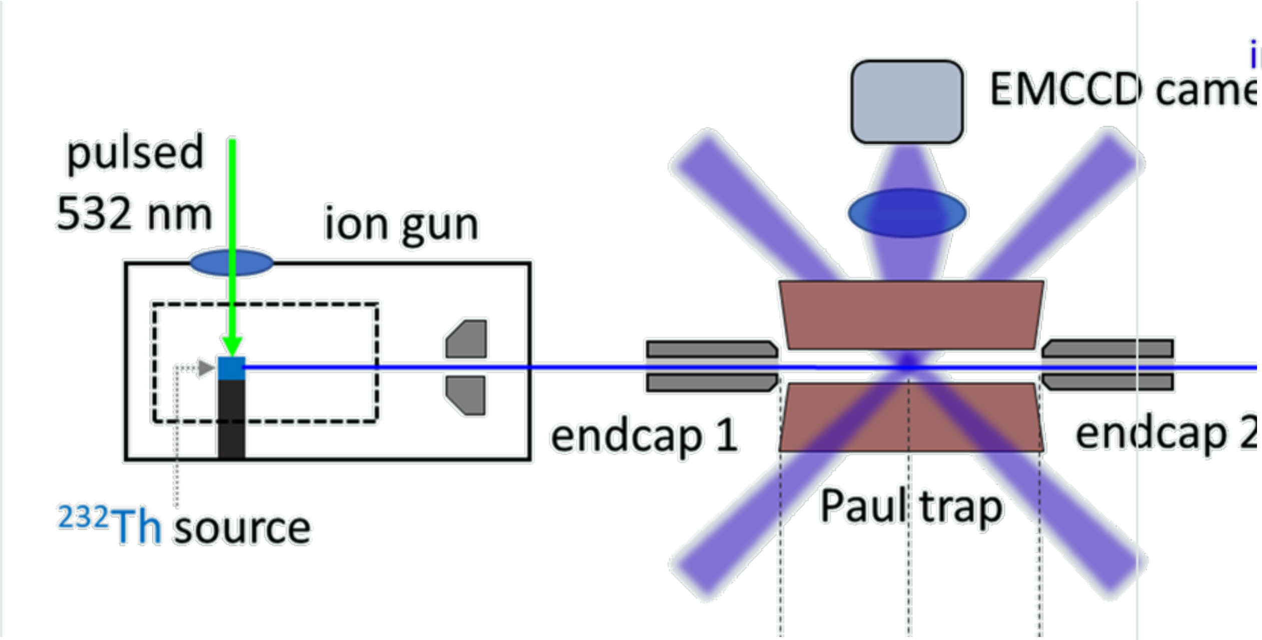
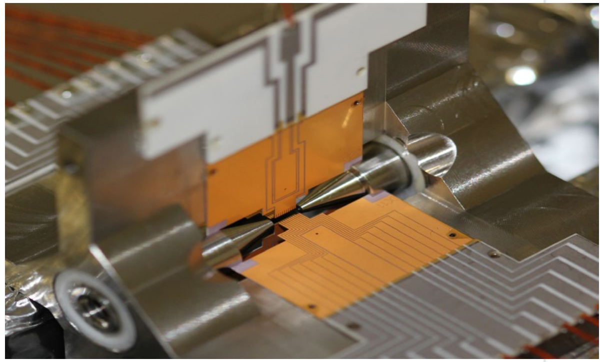
https://www.hi-mainz.de/research/helmholtz-excellence-networks/tactica

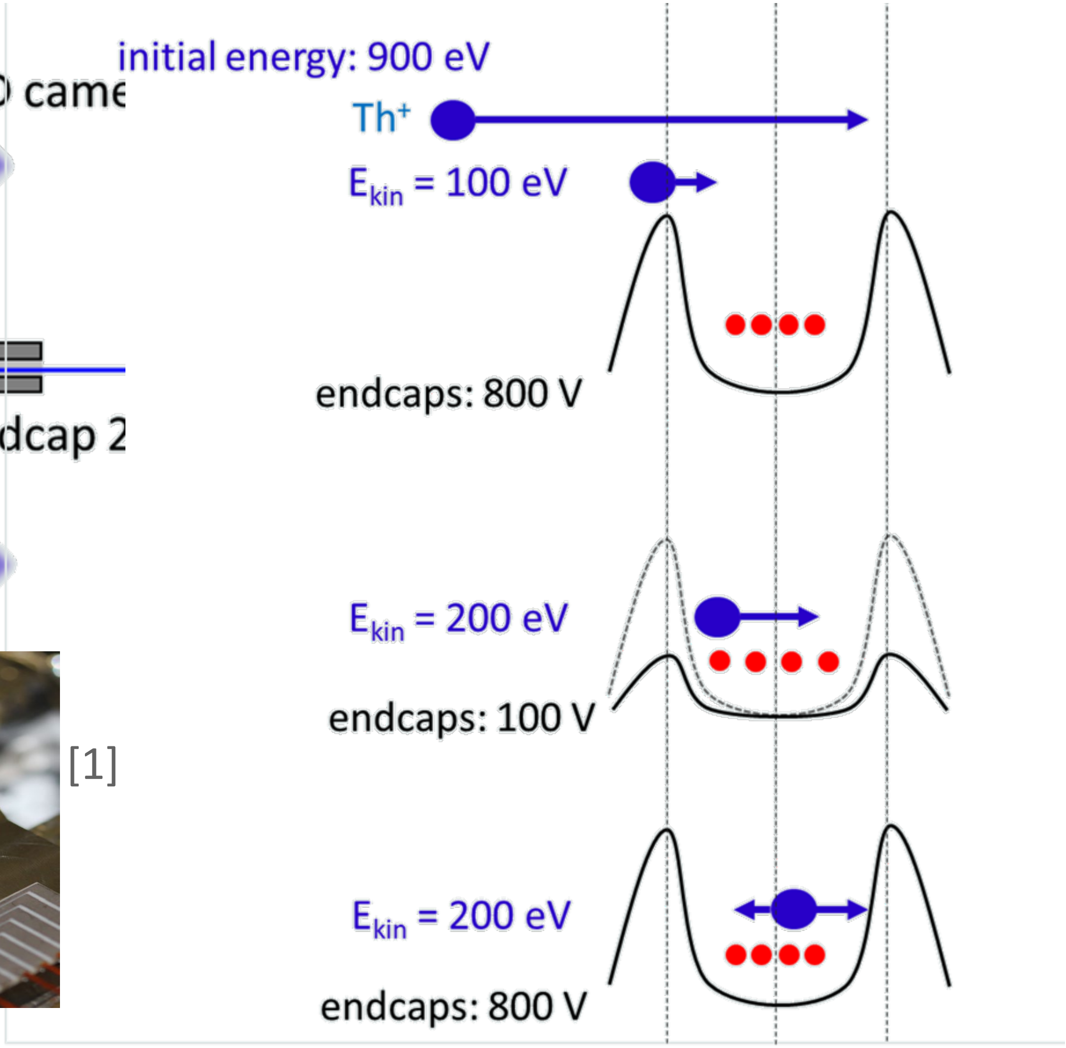
 Author contact: jostrick@uni-mainz.de

Trapping mixed ion crystals





[1]

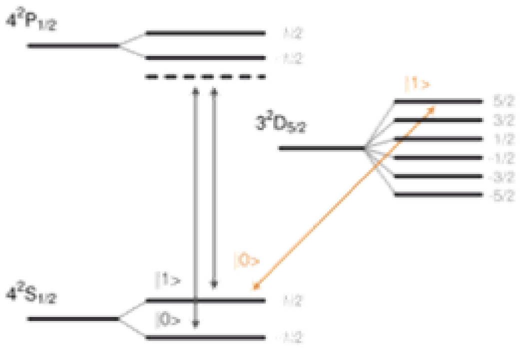


K. Groot-Berning et. al., *Phys. Rev. A.* **2019**, 99, 023420.
F. Stopp et. al., *Hyp. Int.* **2019**, 240, 33.
W. Li et. al., *New J. Phys* **2022** 24, 043028.

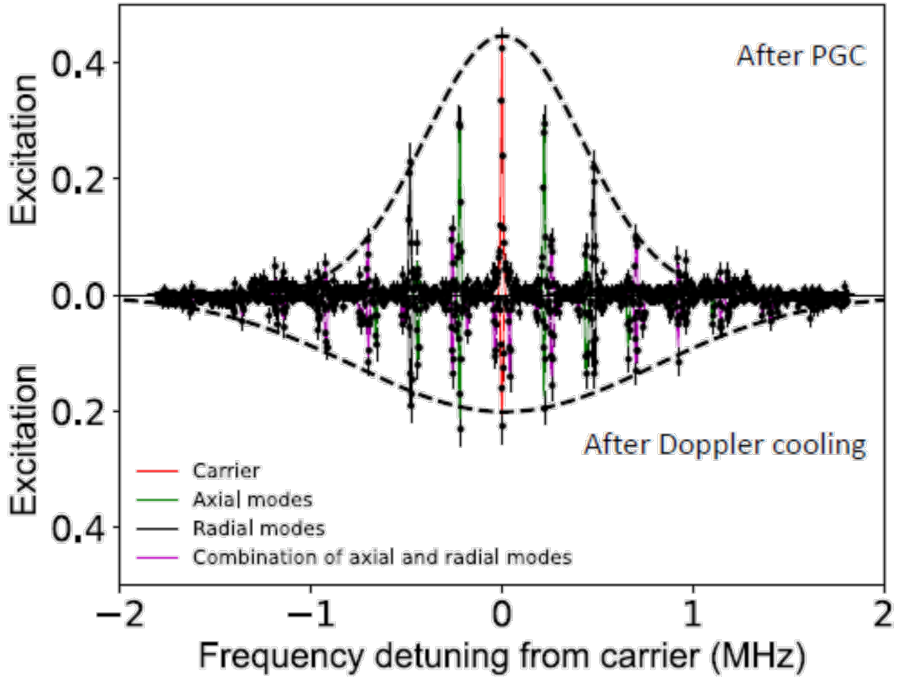
SIDEBAND SPECTROSCOPY

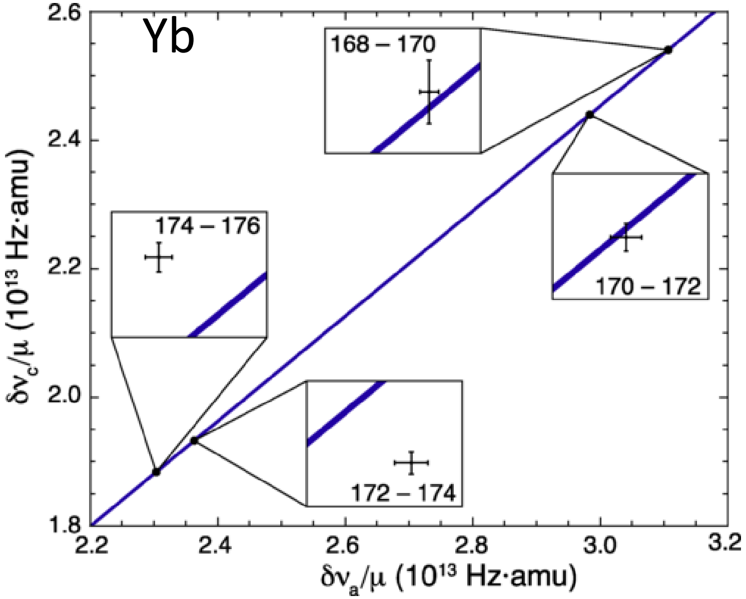
Temperature of trapped $^{232}Th^{+}$: Sideband spectroscopy on Ca^{+} crystal

For temperature measurements: sideband spectroscopy on the $|S_{1/2}, m=1/2\rangle \rightarrow |D_{5/2}, m=5/2\rangle$ transition of $^{40}Ca^{+}$

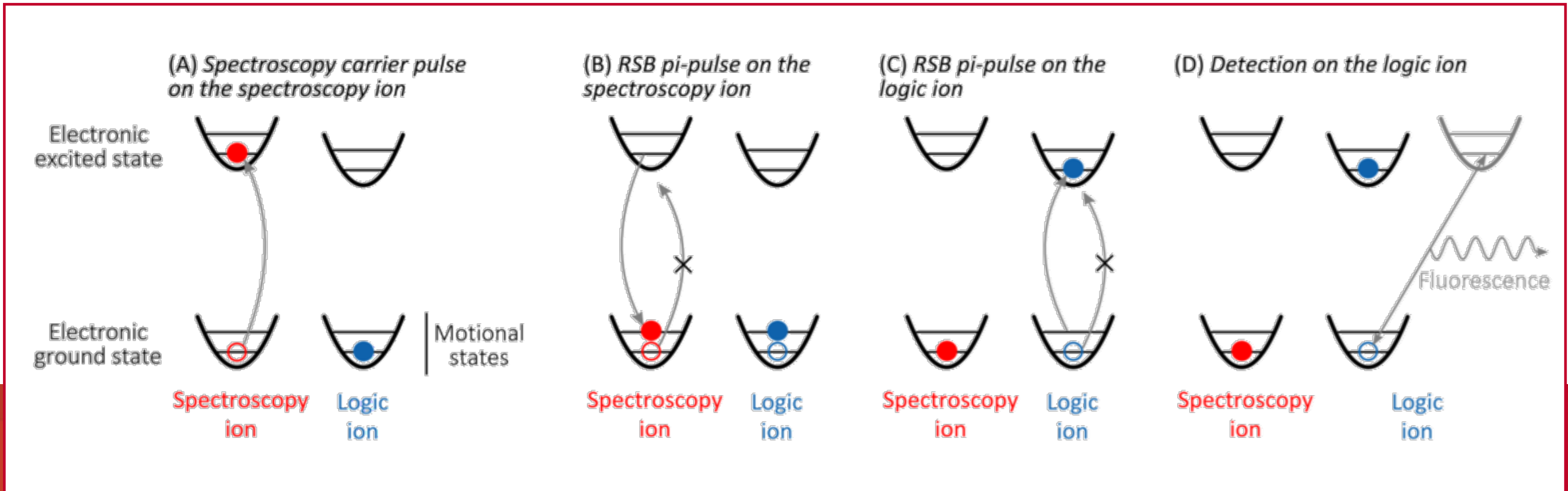


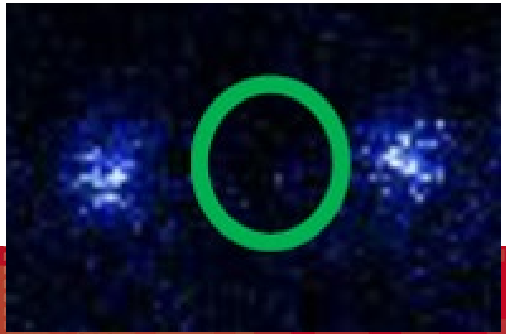
Ca^{+} level scheme with Zeeman sublevels





N. L. Figueroa et al. *Phys. Rev. Lett.* **2022** 128, 073001.

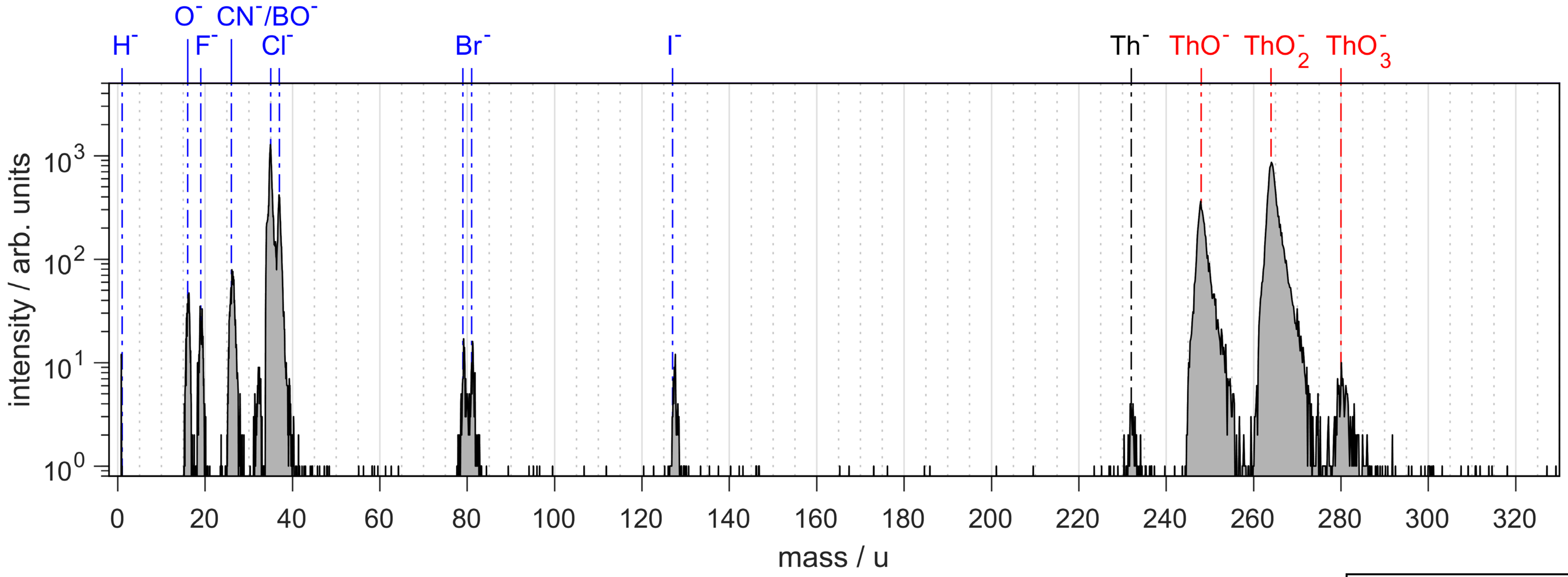




JG|U

Laser ablation of thorium – anions

anions up to 300 u



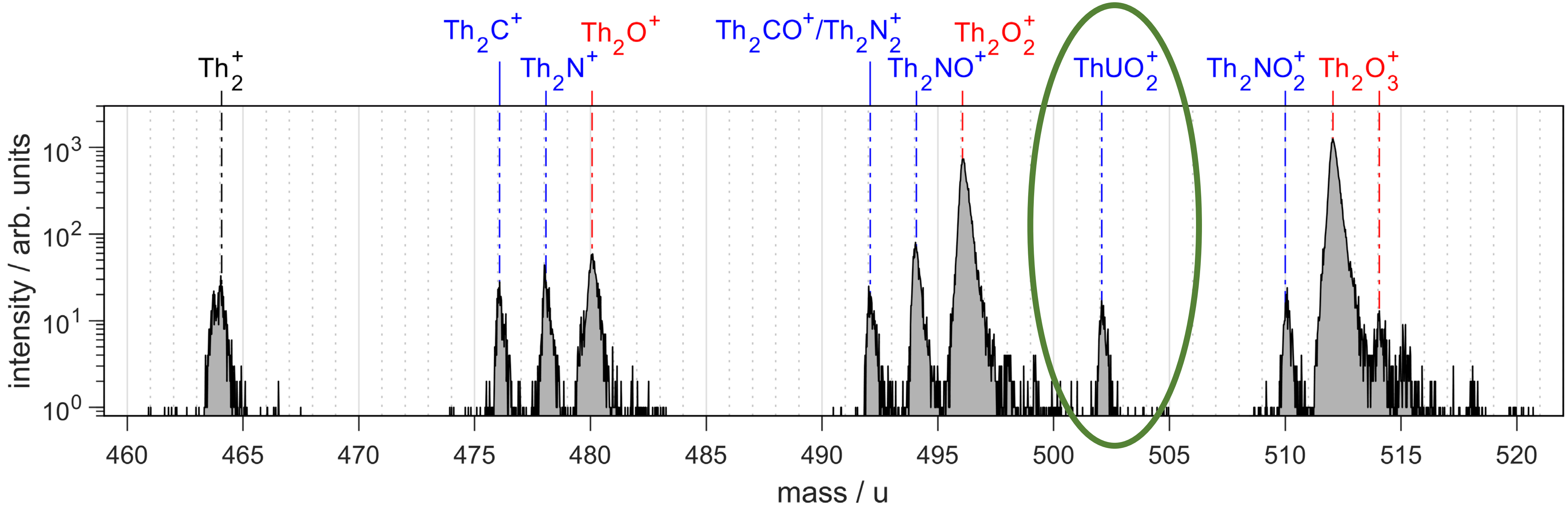
- Mostly expected low-mass contaminants
- Atomic thorium anions and thorium oxides found

labeled species are confirmed by precision mass measurements

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Dimers

0 u — 300 u — 460 – 520 u

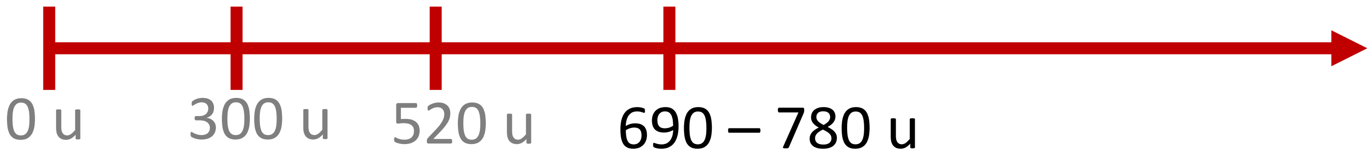


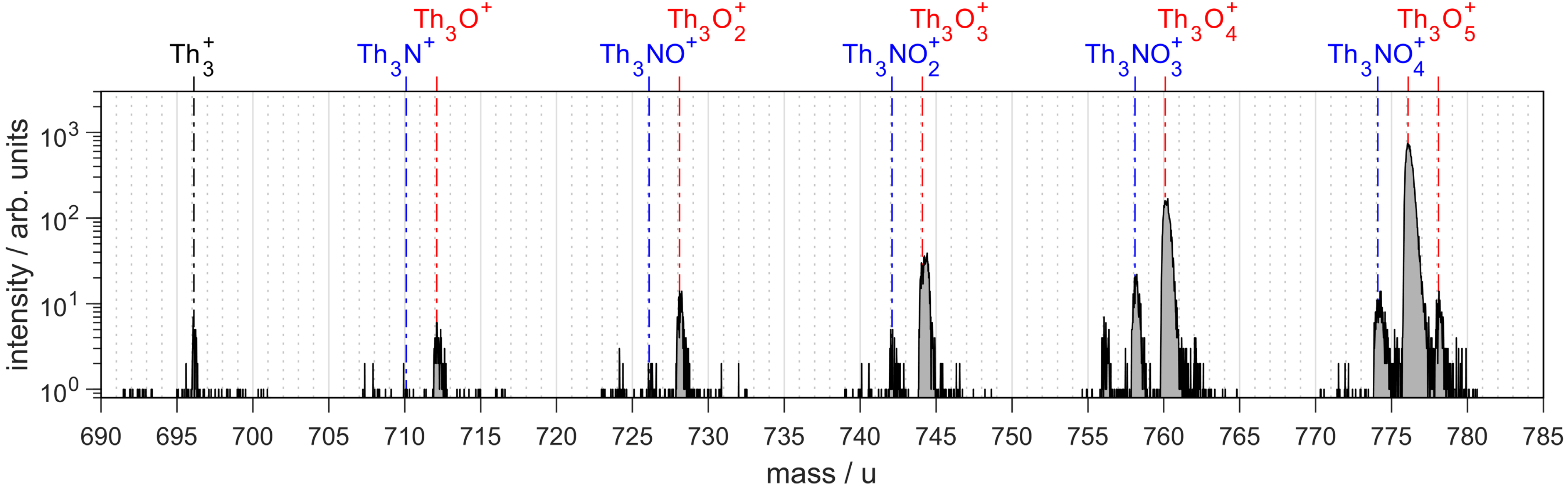
- Atomic thorium dimer as well as di thorium molecules found
- Th containing C, N, O

- $ThUO_2^+$: the only observed thorium-uranium mixed compound species

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Trimers



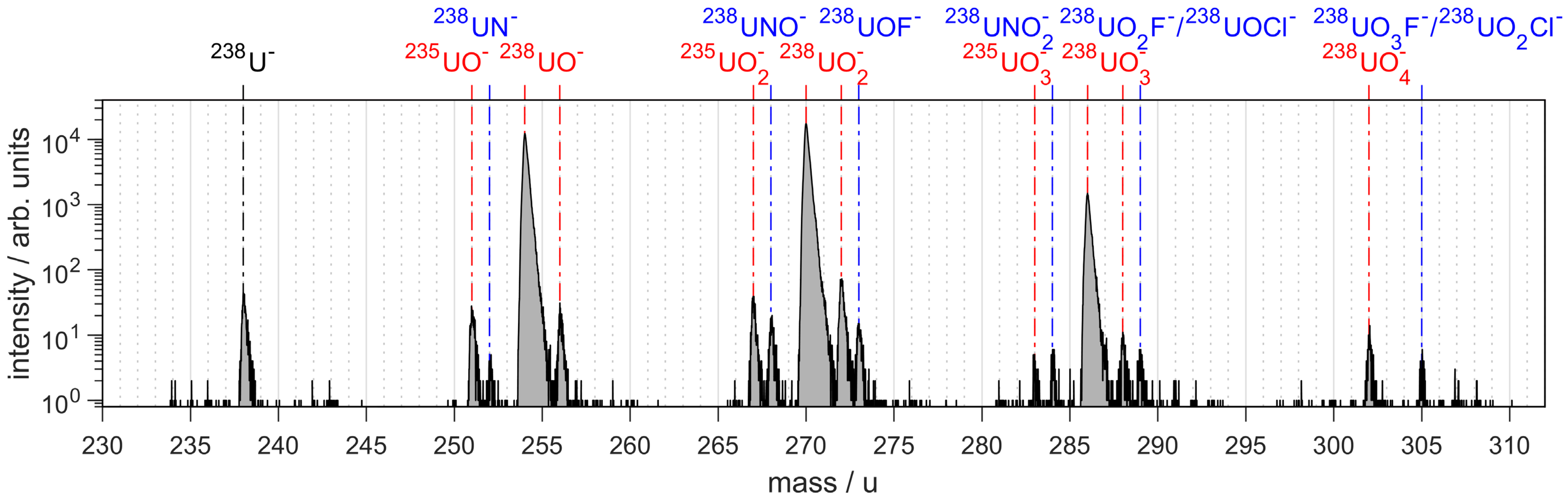


- Atomic thorium trimer as well as tri thorium molecules found
- Th containing N, O; no C species
- No thorium-uranium mixed compound species
- No trimers of U found while analyzing depleted U foil

[4] P. Fischer, J. Stricker et al., submitted to PRR **2024**.

Uranium

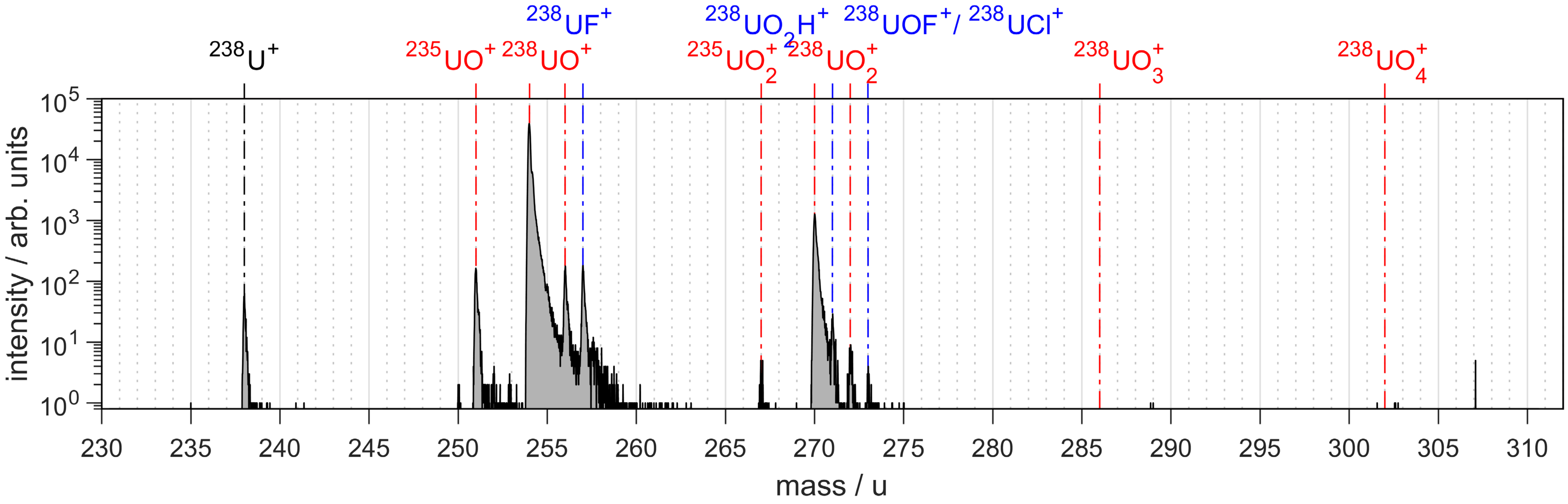
- Depleted uranium was ablated (0.2 % U-235)
- Produced in Oak Ridge; Analyzation in 2023
- Shipped to Mainz in 2024
- Shipped to Greifswald in 2024
- O, N, F, Cl contaminations found



[10] J. Stricker, P. Fischer et al., in preparation **2024**.

Uranium

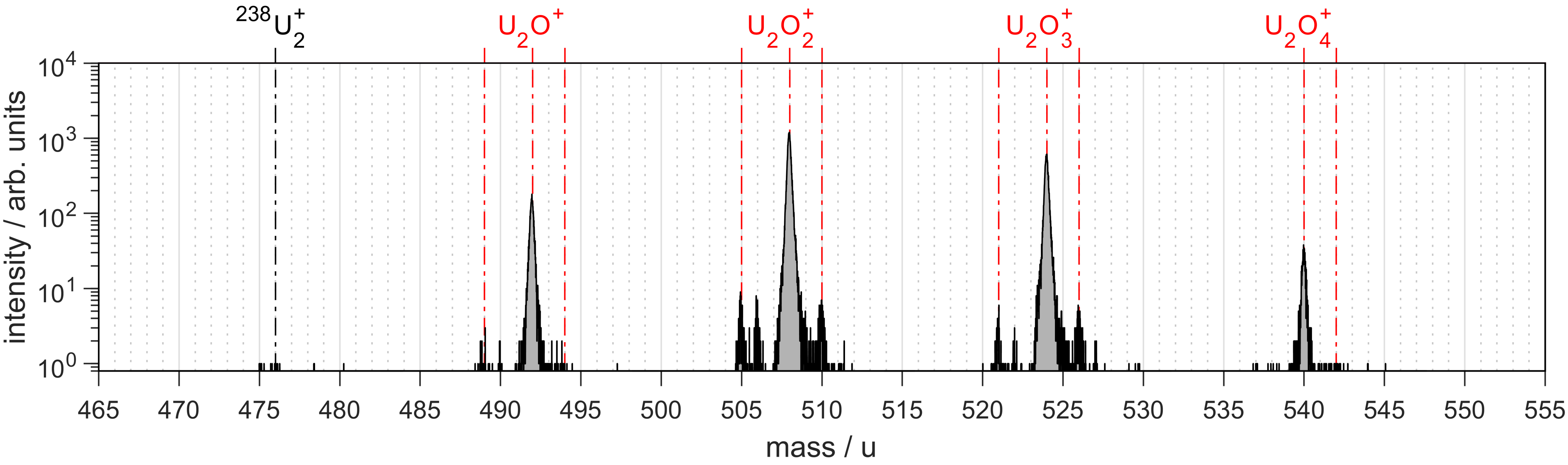
- No Th contamination found
- Better separation during production of foil
  → produced this century



[10] J. Stricker, P. Fischer et al., in preparation **2024**.

Uranium

- No trimers found → no evidence of clusters in gas phase



[10] J. Stricker, P. Fischer et al., in preparation **2024**.