



Developments for actinide molecular ion beams at ISOLDE

Mia Au

ISOLDE Workshop and Users Meeting 2022 Geneva, Switzerland



Agenda

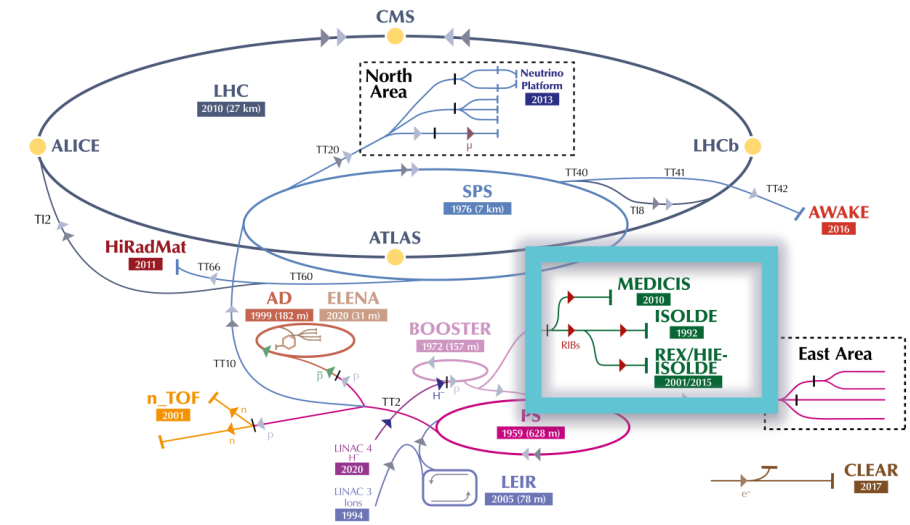
1. ISOLDE beams

2. Actinides at ISOLDE

3. Molecular beams at ISOLDE

4. Actinide molecules

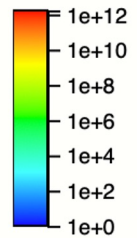
5. Next steps



▶ H^- (hydrogen anions) ▶ p (protons) ▶ ions ▶ RIBs (Radioactive Ion Beams) ▶ n (neutrons) ▶ \bar{p} (antiprotons) ▶ e^- (electrons) ▶ μ (muons)

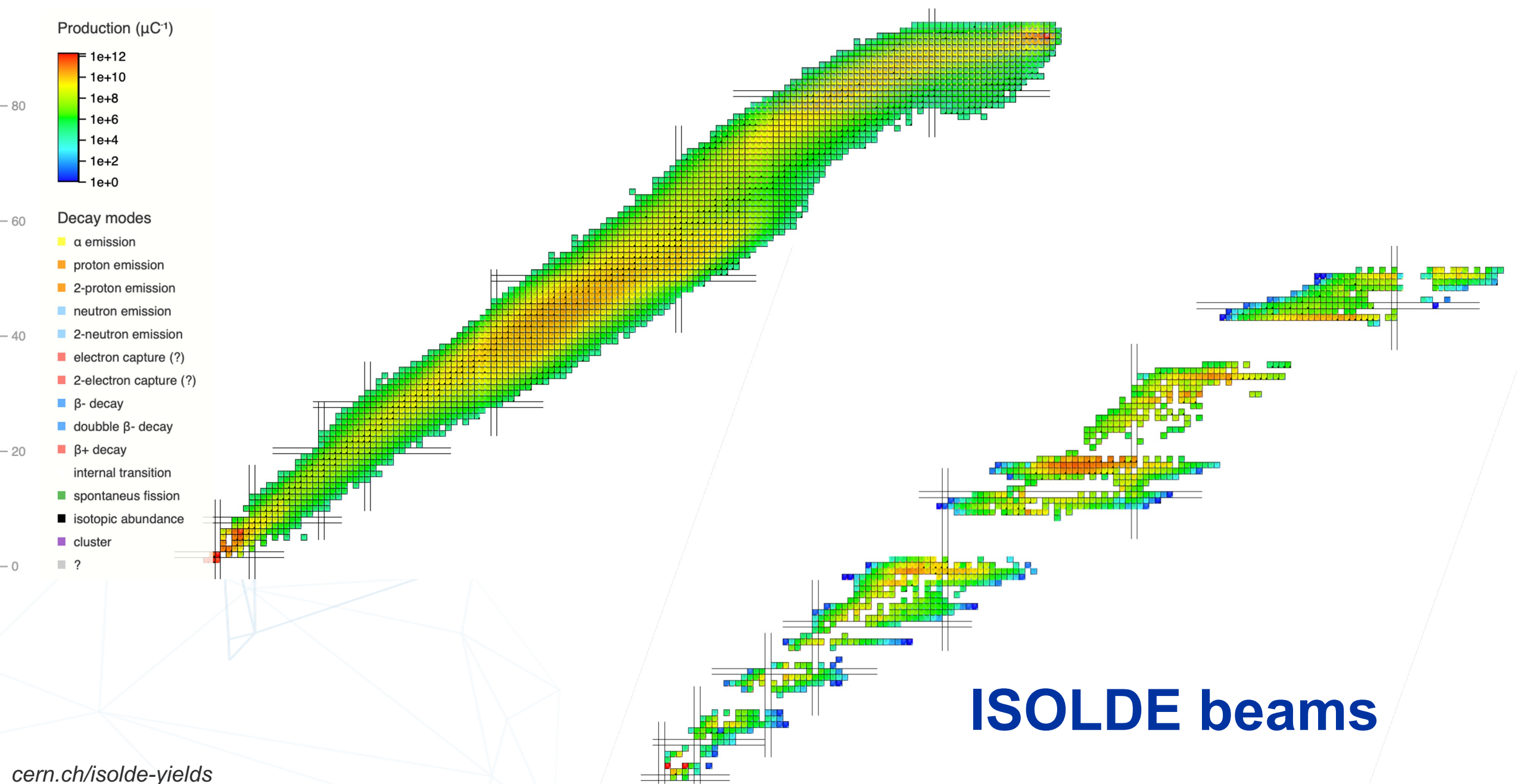
LHC - Large Hadron Collider // SPS - Super Proton Synchrotron // PS - Proton Synchrotron // AD - Antiproton Decelerator // CLEAR - CERN Linear Electron Accelerator for Research // AWAKE - Advanced WAKEfield Experiment // ISOLDE - Isotope Separator OnLine // REX/HIE-ISOLDE - Radioactive Experiment/High Intensity and Energy ISOLDE // MEDICIS // LEIR - Low Energy Ion Ring // LINAC - LINeAR Accelerator // n_TOF - Neutrons Time Of Flight // HiRadMat - High-Radiation to Materials // Neutrino Platform

Production (μC^{-1})



Decay modes

- α emission
- proton emission
- 2-proton emission
- neutron emission
- 2-neutron emission
- electron capture (?)
- 2-electron capture (?)
- β- decay
- double β- decay
- β+ decay
- internal transition
- spontaneous fission
- isotopic abundance
- cluster
- ?



ISOLDE beams

cern.ch/isolde-yields



SY
Accelerator Systems

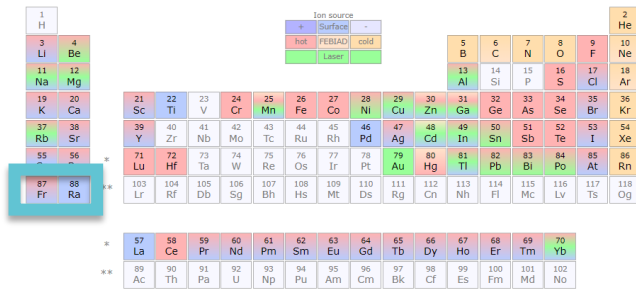


01/12/2022

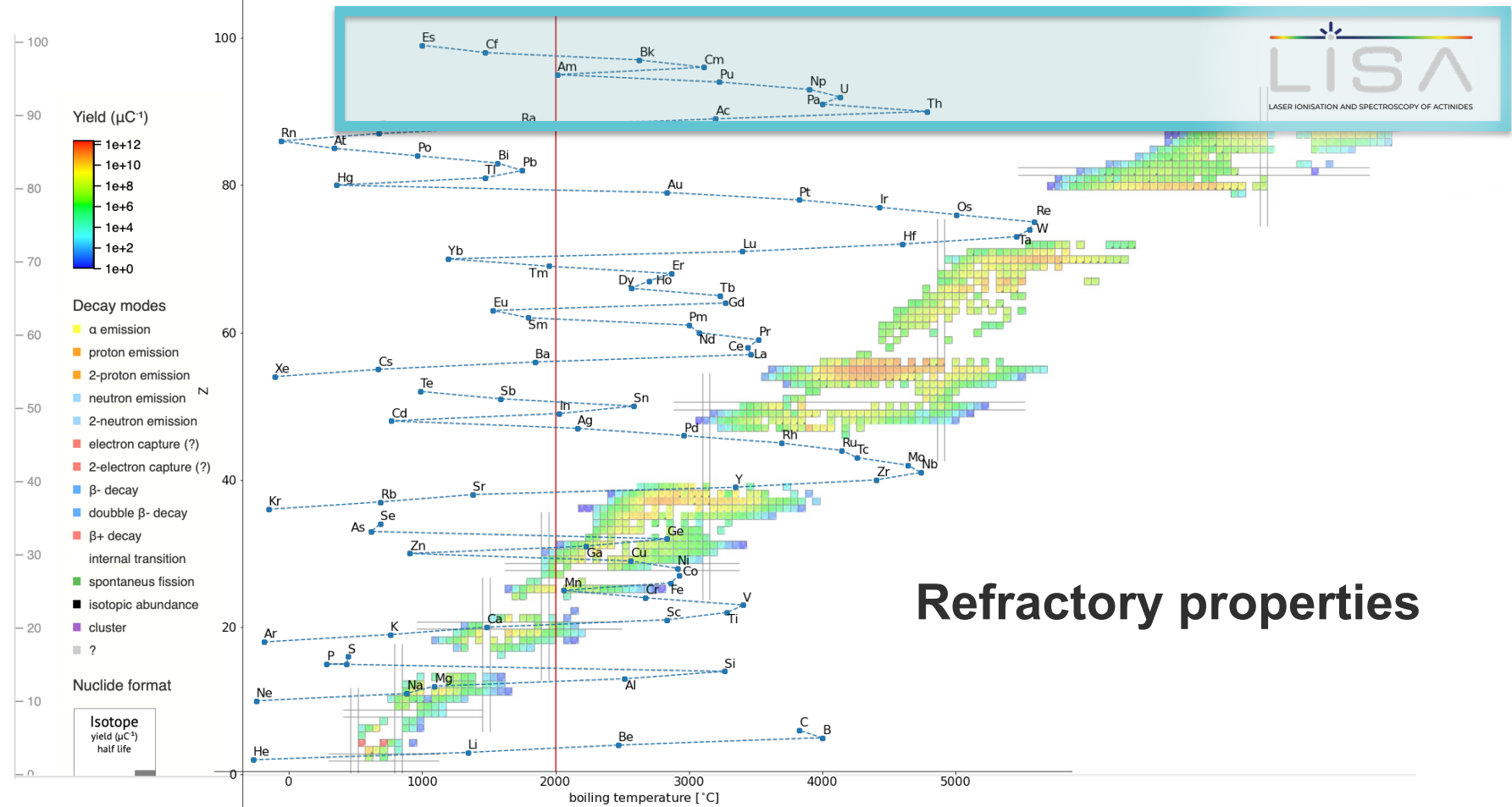
ISOLDE Workshop 2022 | M. Au

Challenges

Contaminants

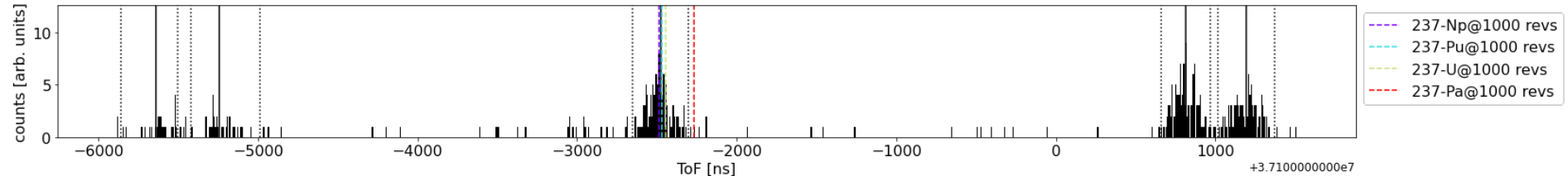


cern.ch/isolde-yields

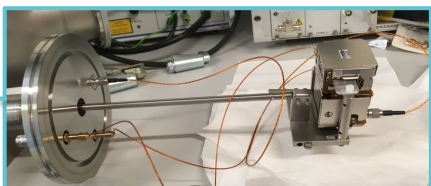
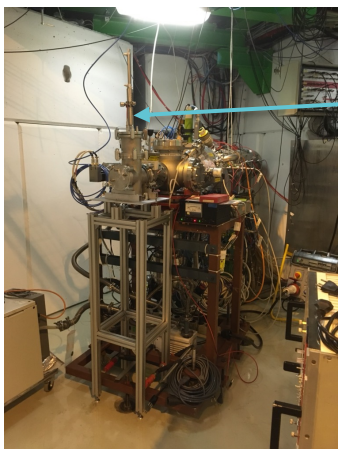


Mass resolving power

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t}$$



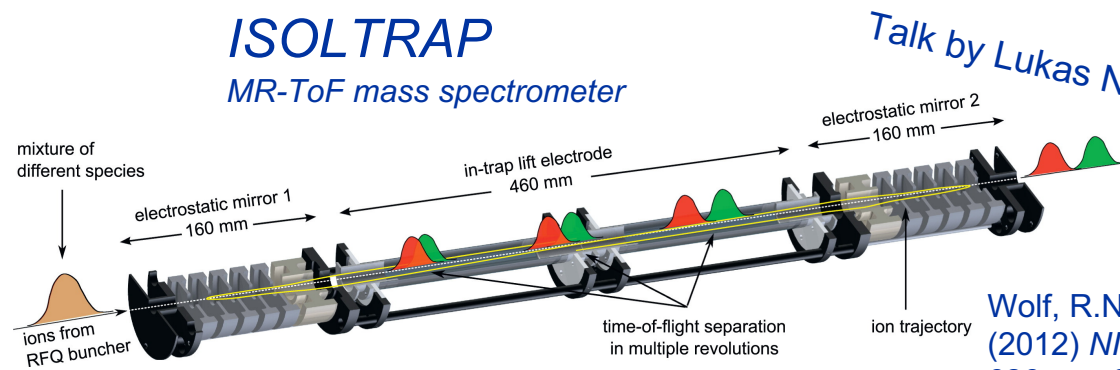
Actinide atomic beams



TISD chamber

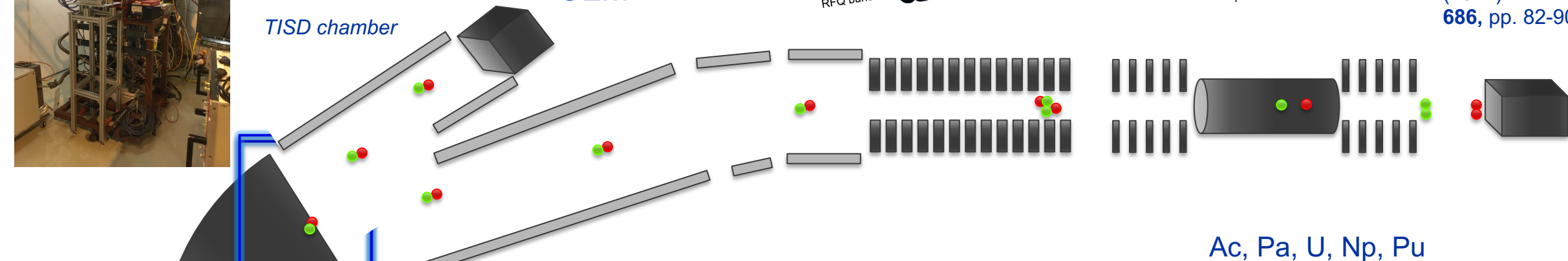
ISOLDE
GLM

ISOLTRAP
MR-ToF mass spectrometer



Talk by Lukas Nies

Wolf, R.N. et al.
(2012) NIM A
686, pp. 82-90.

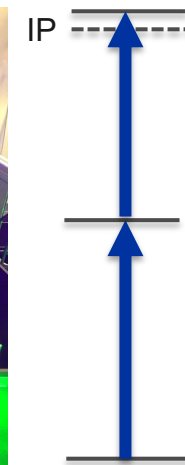
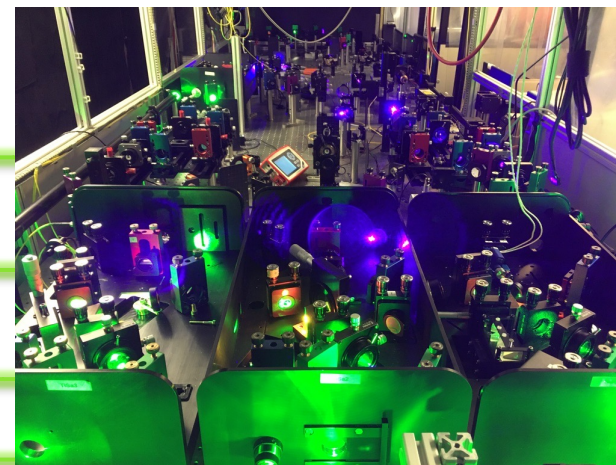
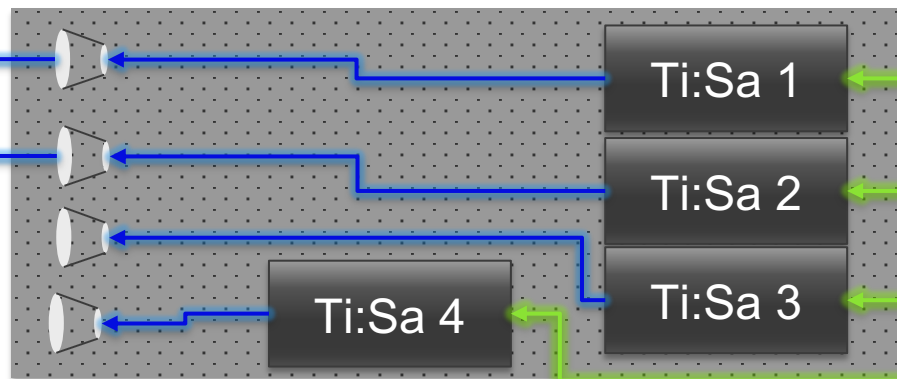


Ac, Pa, U, Np, Pu

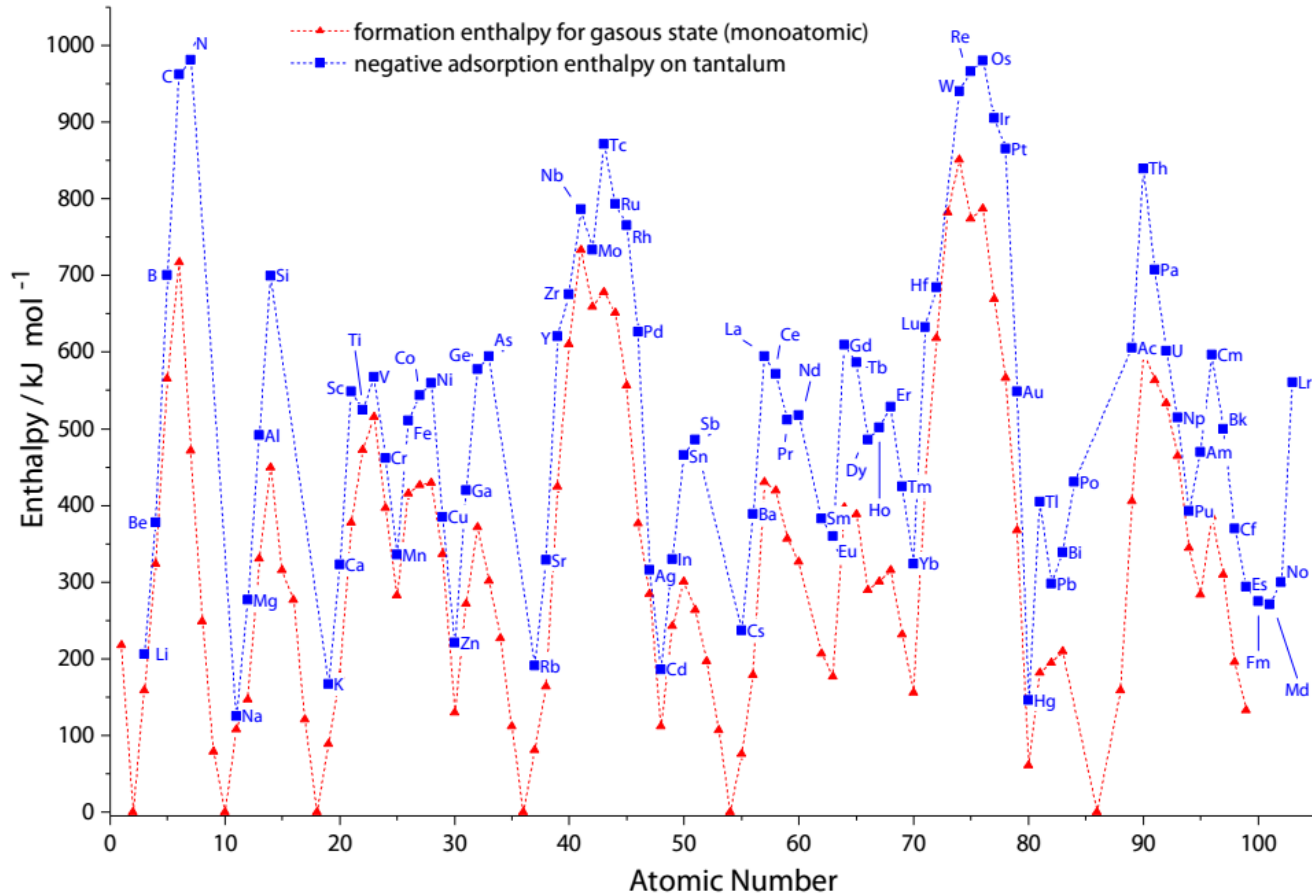
ISOLDE
GPS



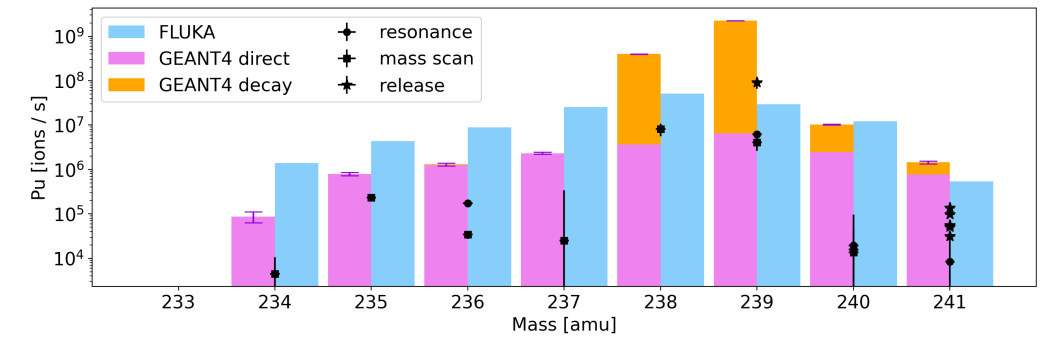
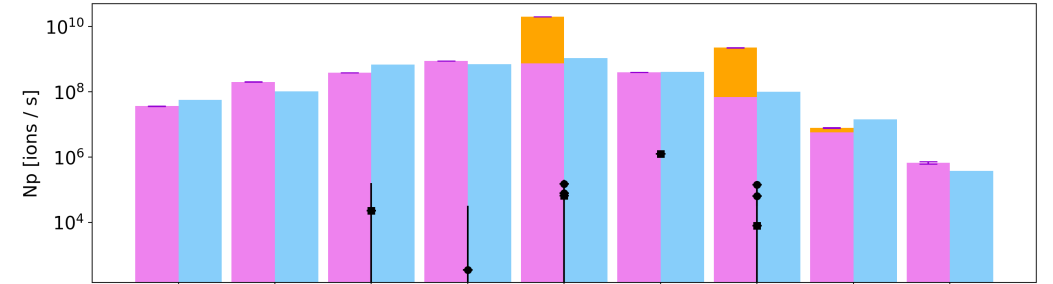
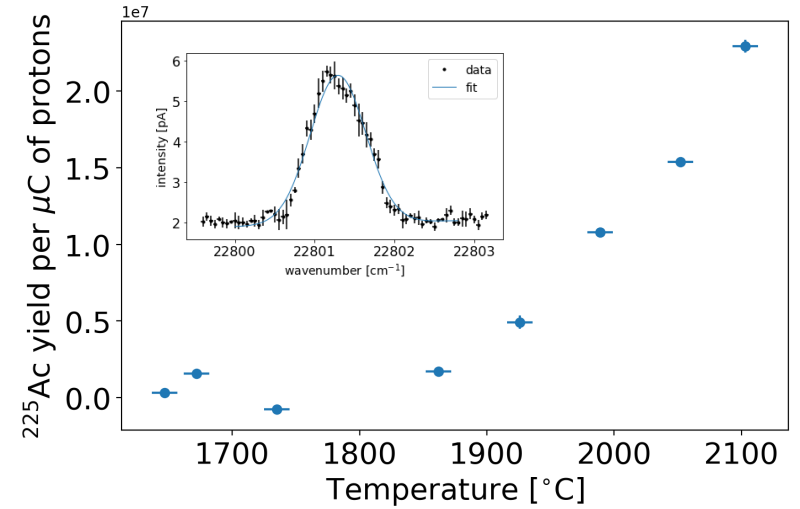
RILIS



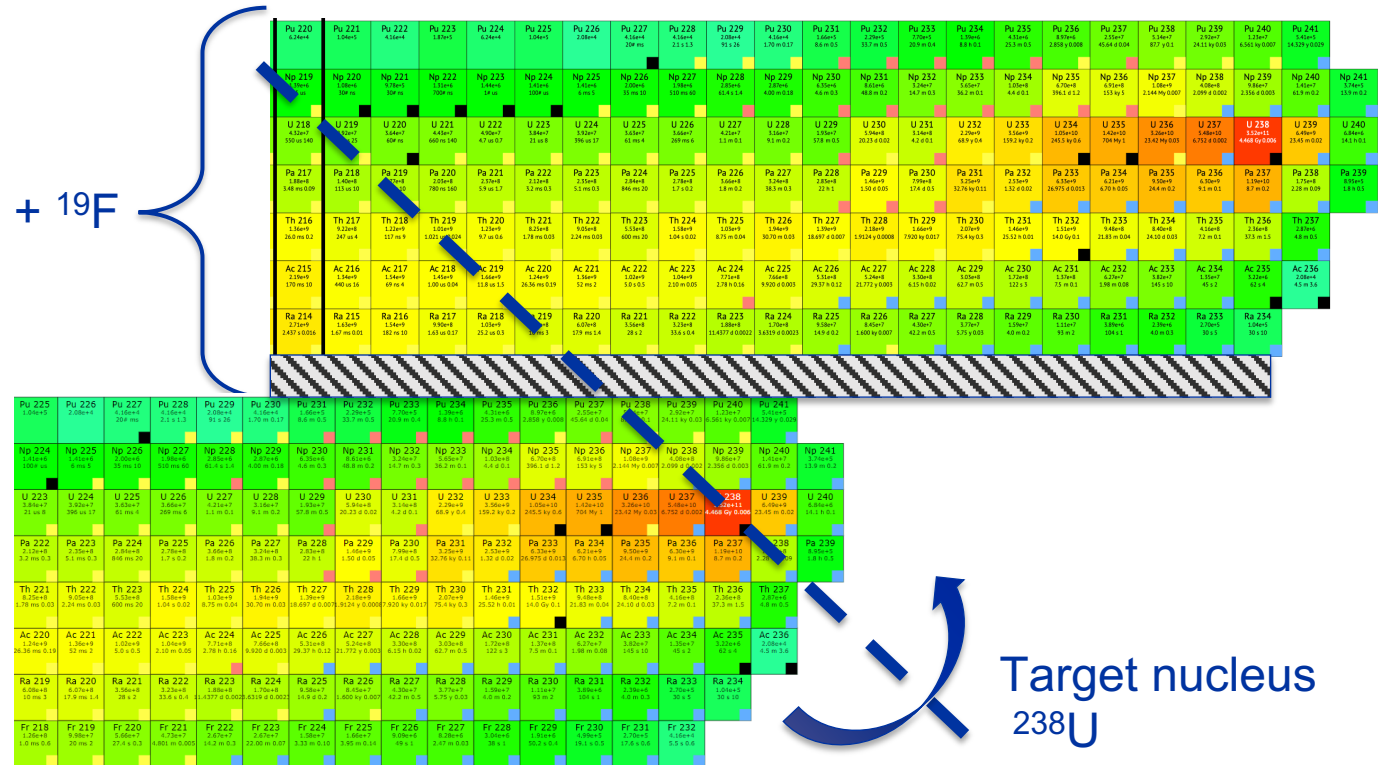
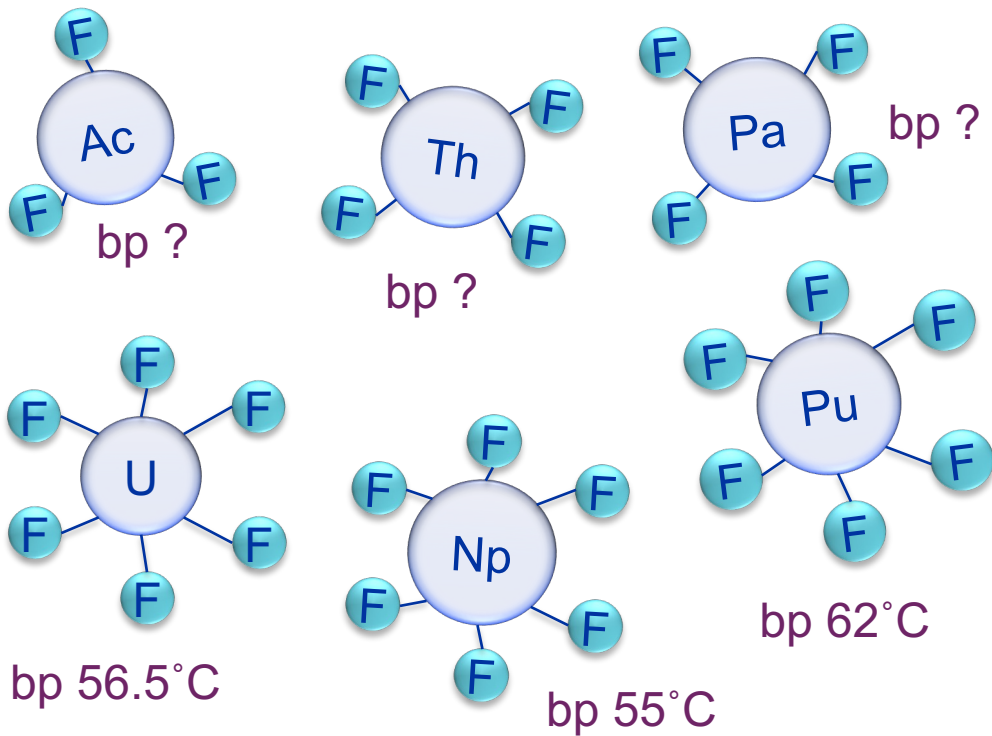
Ac, Np, Pu



Balof, J. (2021) Radioactive Molecular Beams at CERN-ISOLDE

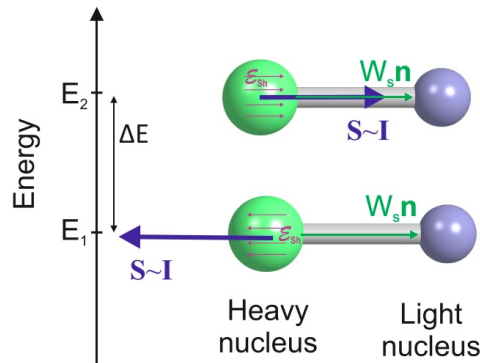


Au, M et al., (2022) In preparation



Molecular beams

1. Volatilization
2. Sideband extraction
3. Research opportunities

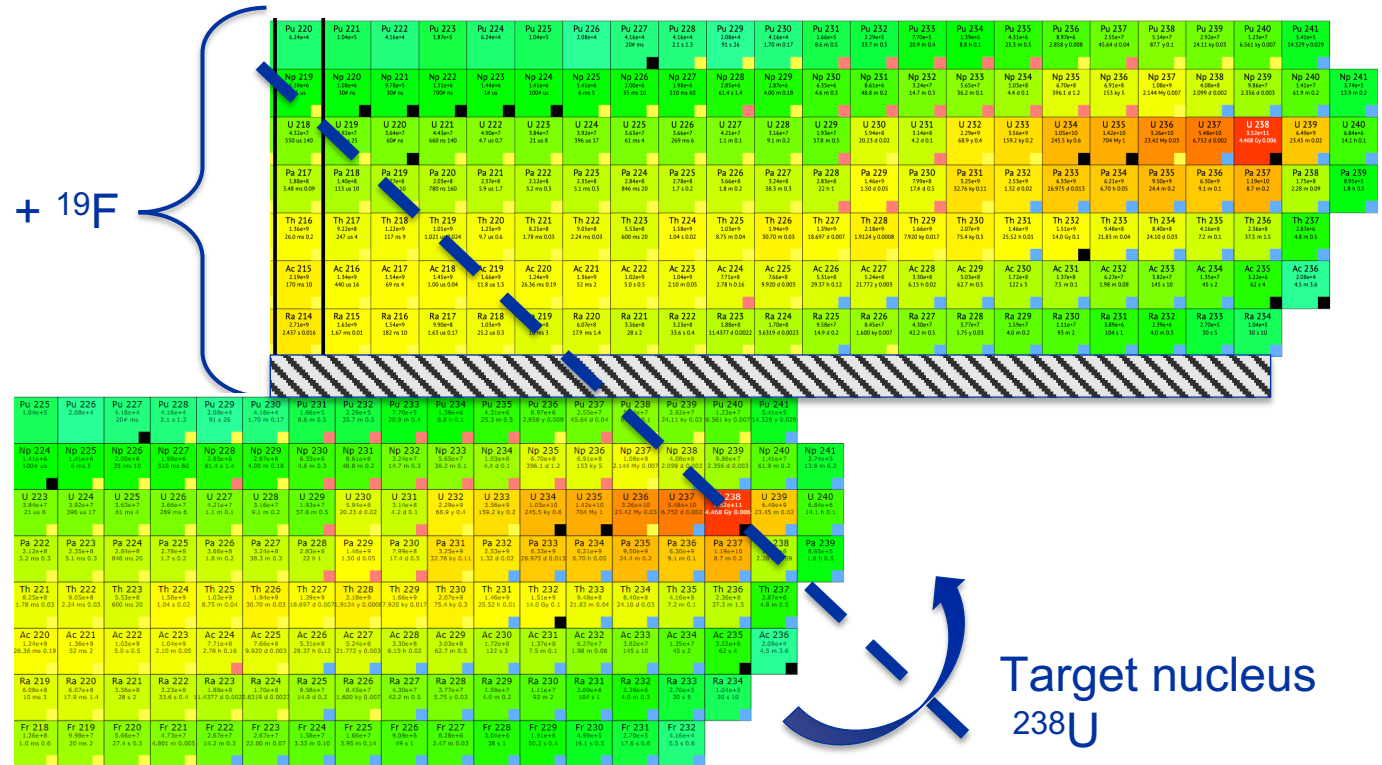
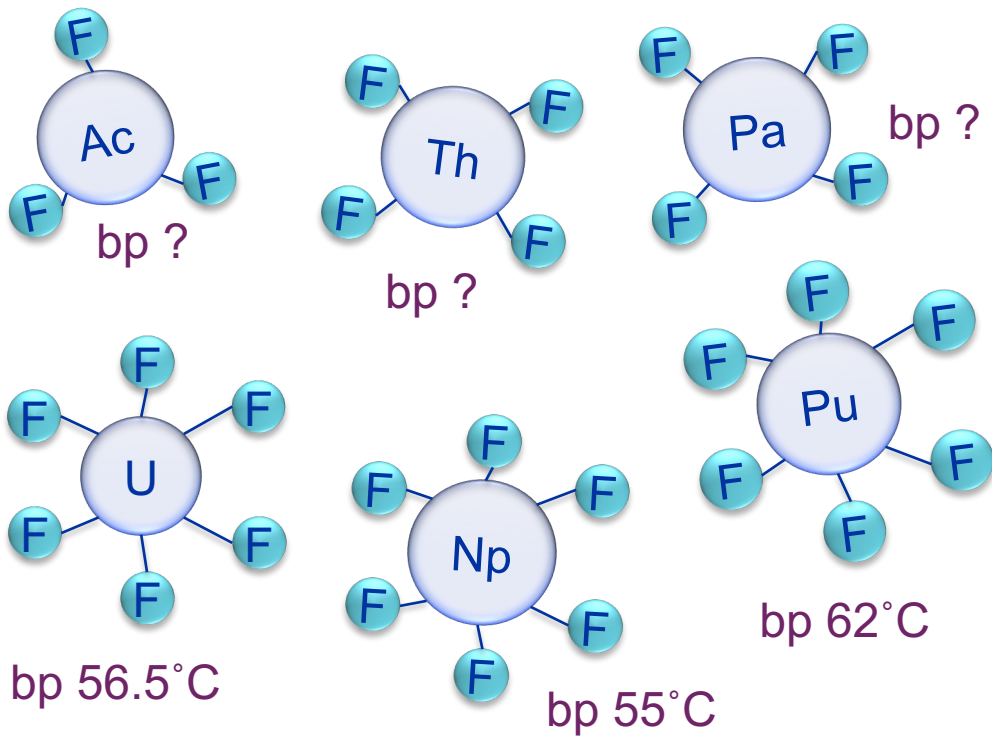


New results from the laser spectroscopy of RaF at CRIS towards searches for new physics
Michail Athanasakis-Kaklamanakis

Effect of temperature distribution on the spectra of RaF
503/1-001 - Council Chamber, CERN
Carlos Mario Fajardo Zambrano et al.
17:48 - 17:50

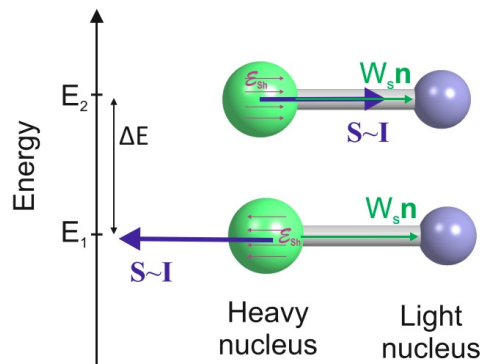
Molecular Ions as Powerful Low-Energy Probes for Fundamental Physics
503/1-001 - Council Chamber, CERN
Carsten Zuelch
18:02 - 18:04

Garcia Ruiz et al. (2020) Nature 581. 396
Safronova, Budker, DeMille et al. (2018) Rev. Mod. Phys. 90. 2,
Skripnikov et al. (2020) Phys. Chem. Chem. Phys. 22. 33, 18374



Molecular beams

1. Formation
2. Detection
3. Identification



New results from the laser spectroscopy of RaF at CRIS towards searches for new physics
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Effect of temperature distribution on the spectra of RaF
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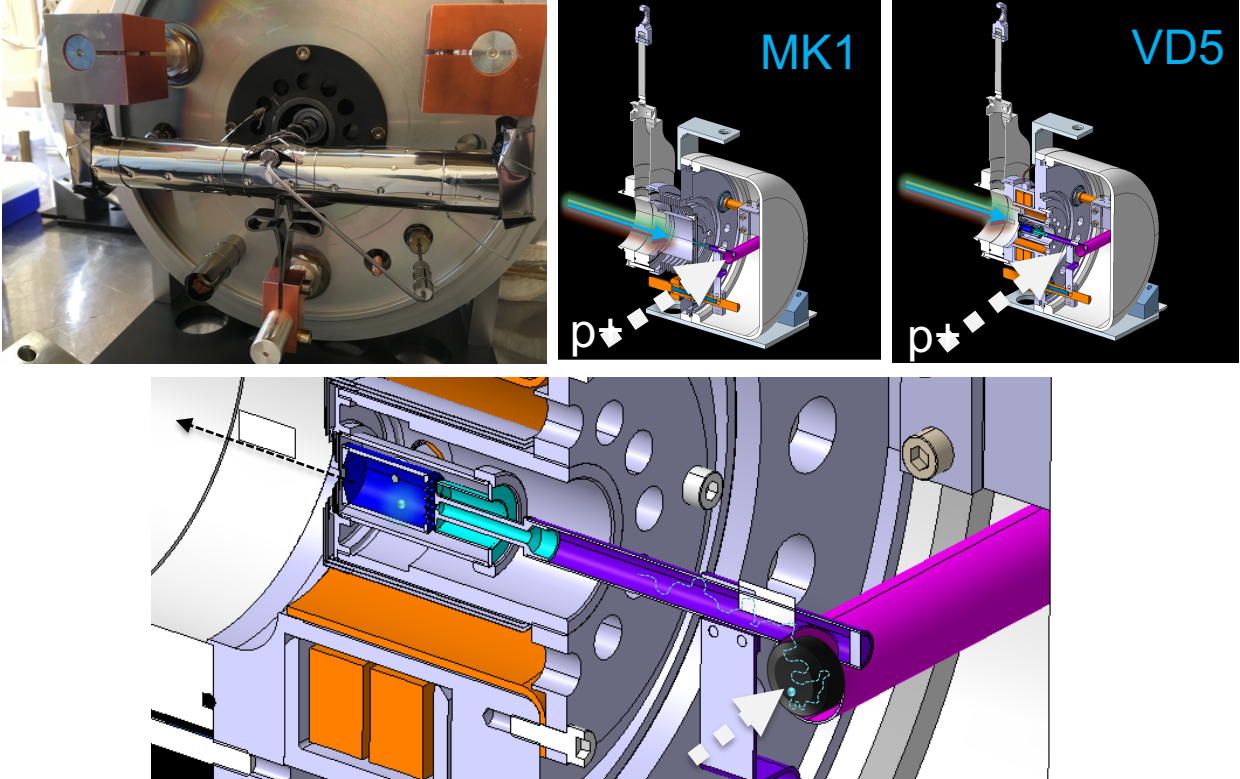
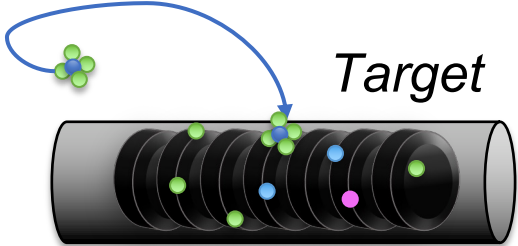
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 Safronova, Budker, DeMille et al. (2018) Rev. Mod. Phys. 90. 2,
 Skripnikov et al. (2020) Phys. Chem. Chem. Phys. 22. 33, 18374

Formation: Making the molecules

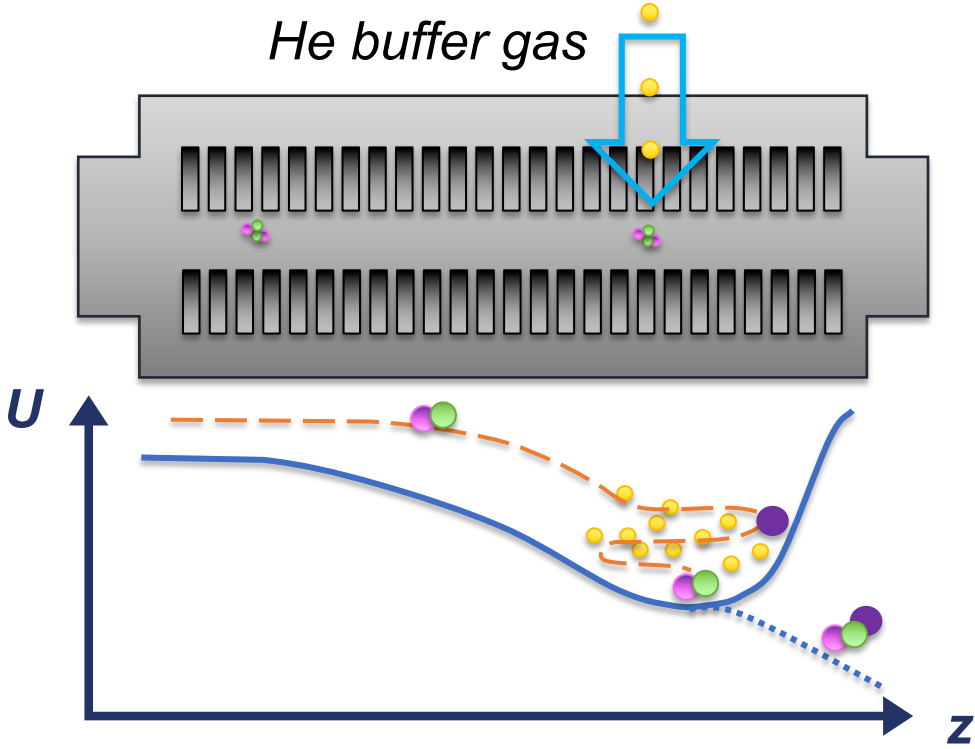
In-target

Reactive gas:
 CF_4 , NF_3 , SF_6



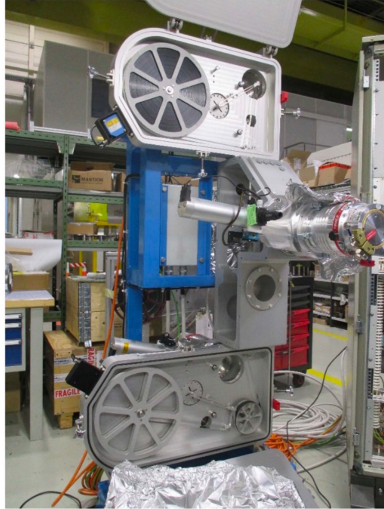
In-trap

Radio-frequency quadrupole cooler-buncher (RFQ-cb)

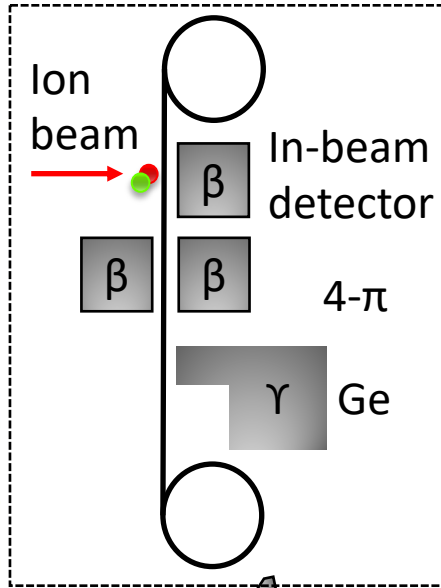


Detection and identification

- Contaminant ions
- Contaminant neutrals
- Isotope of interest
- Neutral molecule of interest
- Molecular ion of interest
- Reactive gas
- Buffer gas

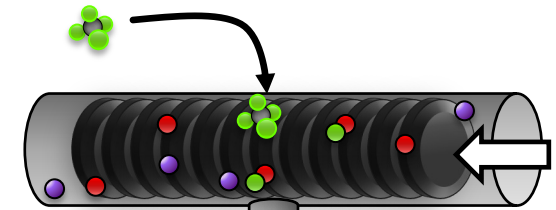


Catherall, R. et al. (2017)
J. Phys. G. 44, 9



ISOLDE
FTS

UCx target

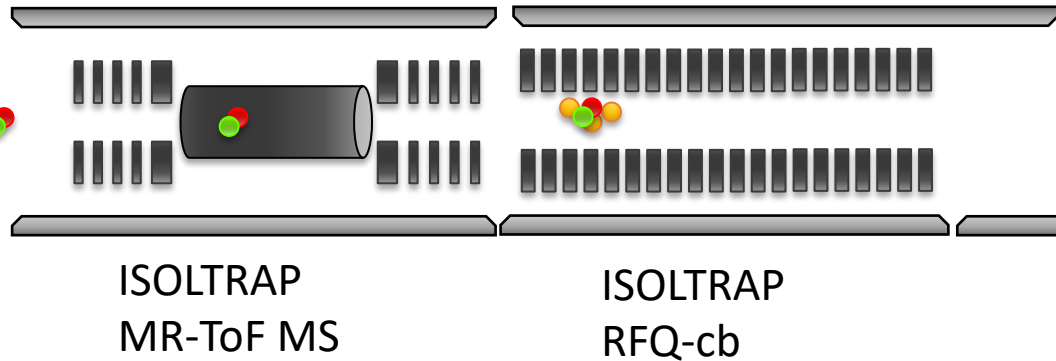
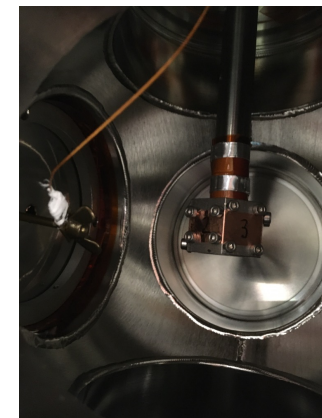


ISOLDE
HRS

Mass
separator

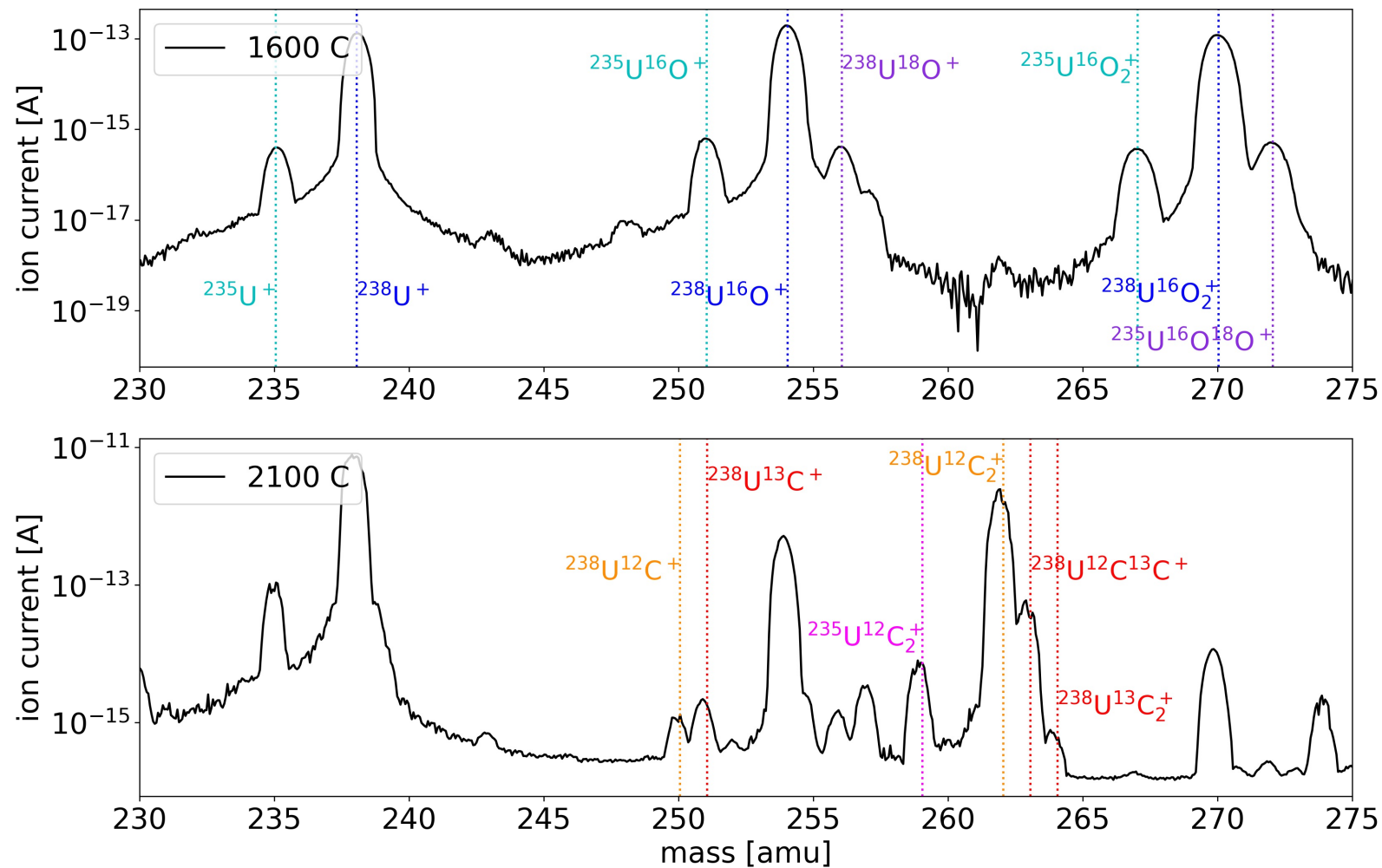
ISOLDE
LA1

collections chamber



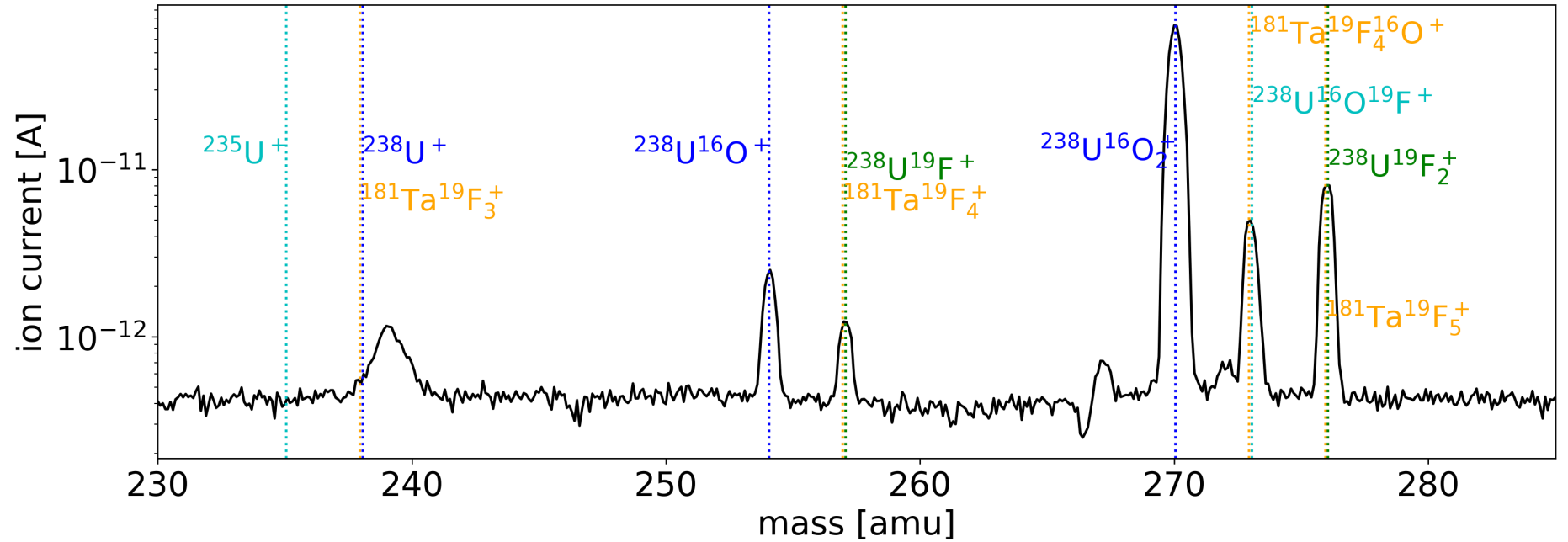
Surface ionized target molecules

Species	Rate [ions/s]
$^{235}\text{U}^+$	7E5
$^{238}\text{U}^+$	3.7E7
$^{238}\text{U}^{12}\text{C}^+$	7.5E3
$^{235}\text{U}^{16}\text{O}^+$	1.3E4
$^{238}\text{U}^{16}\text{O}^+$	4.2E6
$^{238}\text{U}^{18}\text{O}^+$	9E3
$^{235}\text{U}^{12}\text{C}_2^+$	6.5E4
$^{238}\text{U}^{12}\text{C}_2^+$	1.1E7
$^{238}\text{U}^{12}\text{C}^{13}\text{C}^+$	4.8E5
$^{238}\text{U}^{13}\text{C}_2^+$	1.65E3
$^{235}\text{U}^{16}\text{O}_2^+$	2E3
$^{238}\text{U}^{16}\text{O}_2^+$	7E5
$^{235}\text{U}^{16}\text{O}^{18}\text{O}^+$	2.5E3



Fluorination

Species	Rate [ions/s]
$^{238}\text{U}^+$?
$^{235}\text{U}^{16}\text{O}^+$	1.3E4
$^{238}\text{U}^{16}\text{O}^+$	4.2E6
$^{238}\text{U}^{19}\text{F}^+$?
$^{181}\text{Ta}^{19}\text{F}_5^+$?
$^{238}\text{U}^{16}\text{O}_2^+$	7E5
$^{181}\text{Ta}^{19}\text{F}_4^{16}\text{O}^+$?
O^+	
$^{238}\text{U}^{19}\text{F}^{16}\text{O}^+$?
$^{238}\text{U}^{19}\text{F}_2^+$?
$^{181}\text{Ta}^{19}\text{F}_5^+$?



Metastable molecular ions

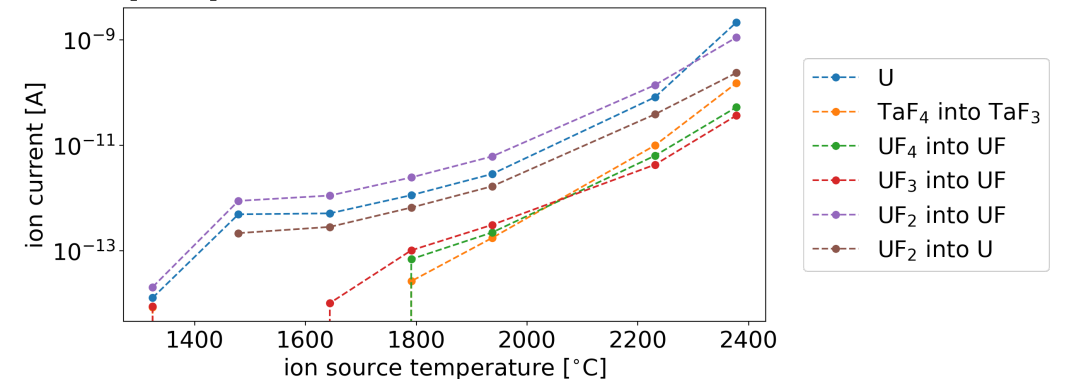
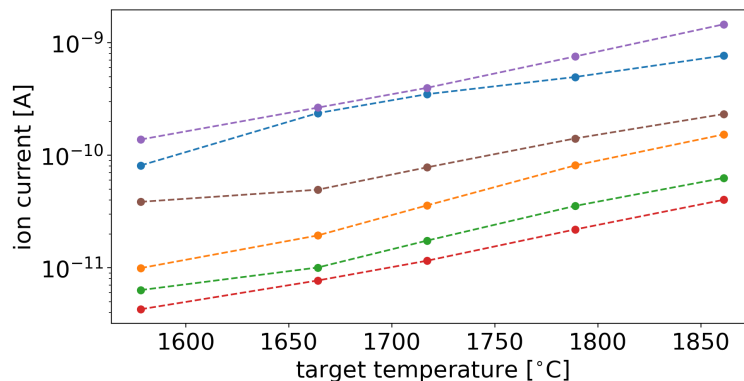
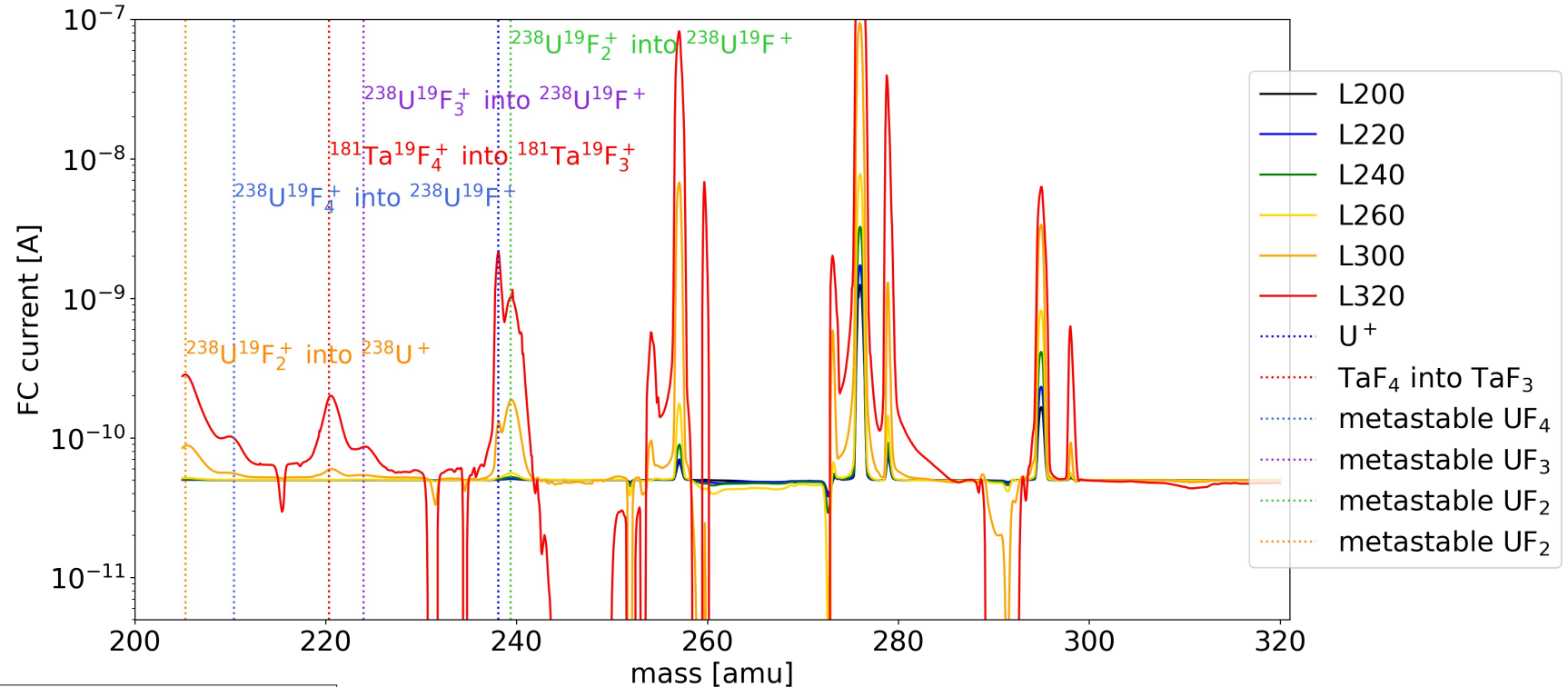
Dissociation **after** acceleration,
before mass separation.

apparent mass: $m^* = \frac{m_f^2}{m_p}$

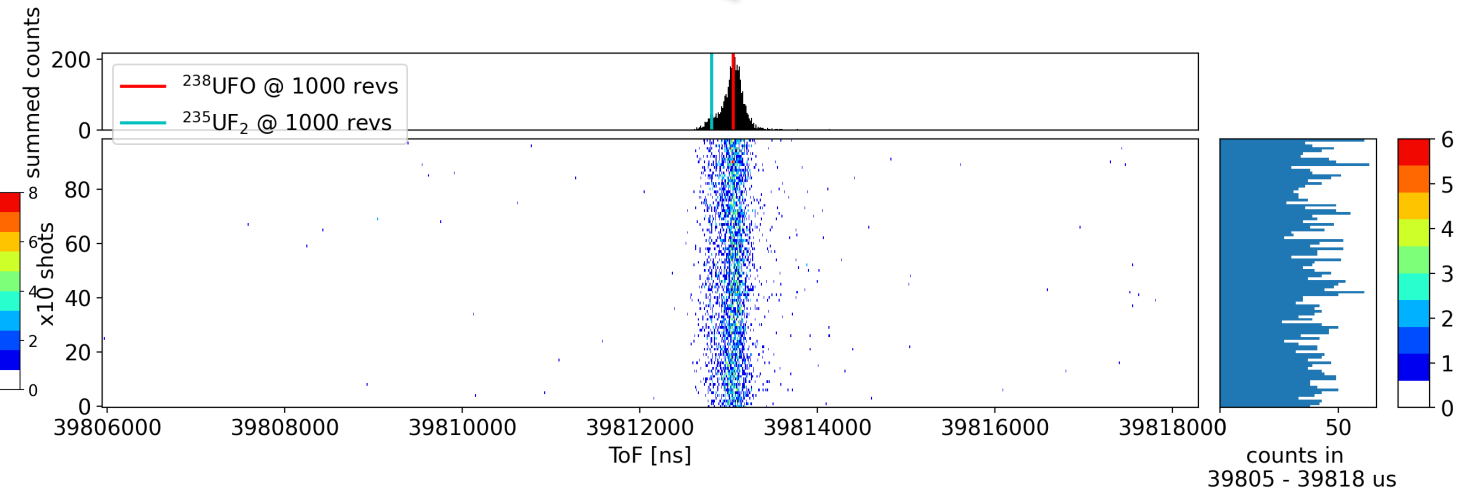
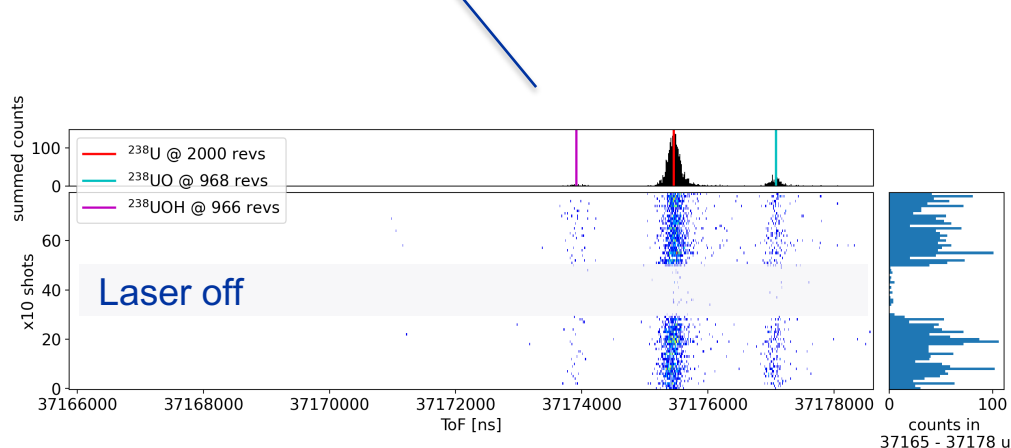
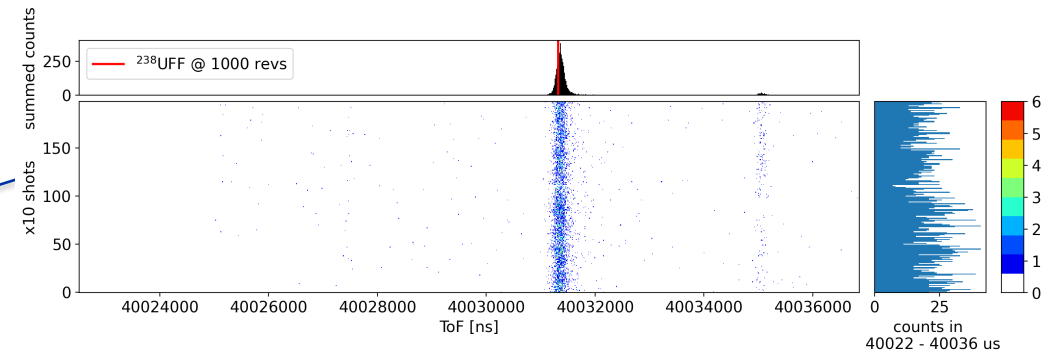
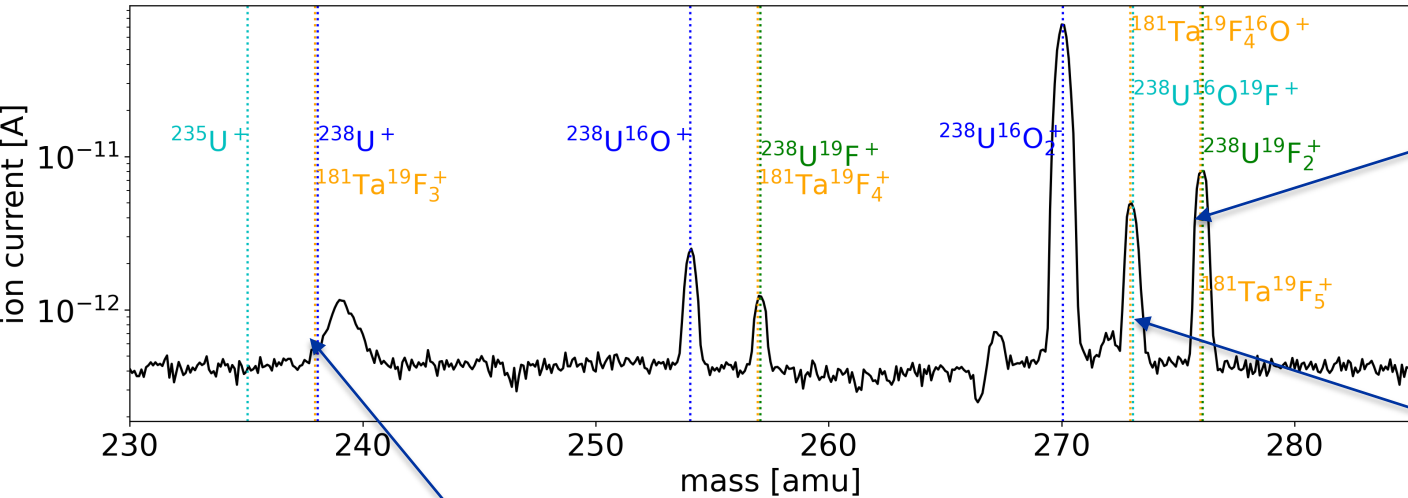
- m_f : mass of fragment ion
- m_p : mass of precursor ion

Ex: $\text{UF}_2^+ \text{ into } \text{UF}^+ + \text{F}$

$$m^* = \frac{(238 + 19)^2}{238 + 2(19)} = 239.31$$

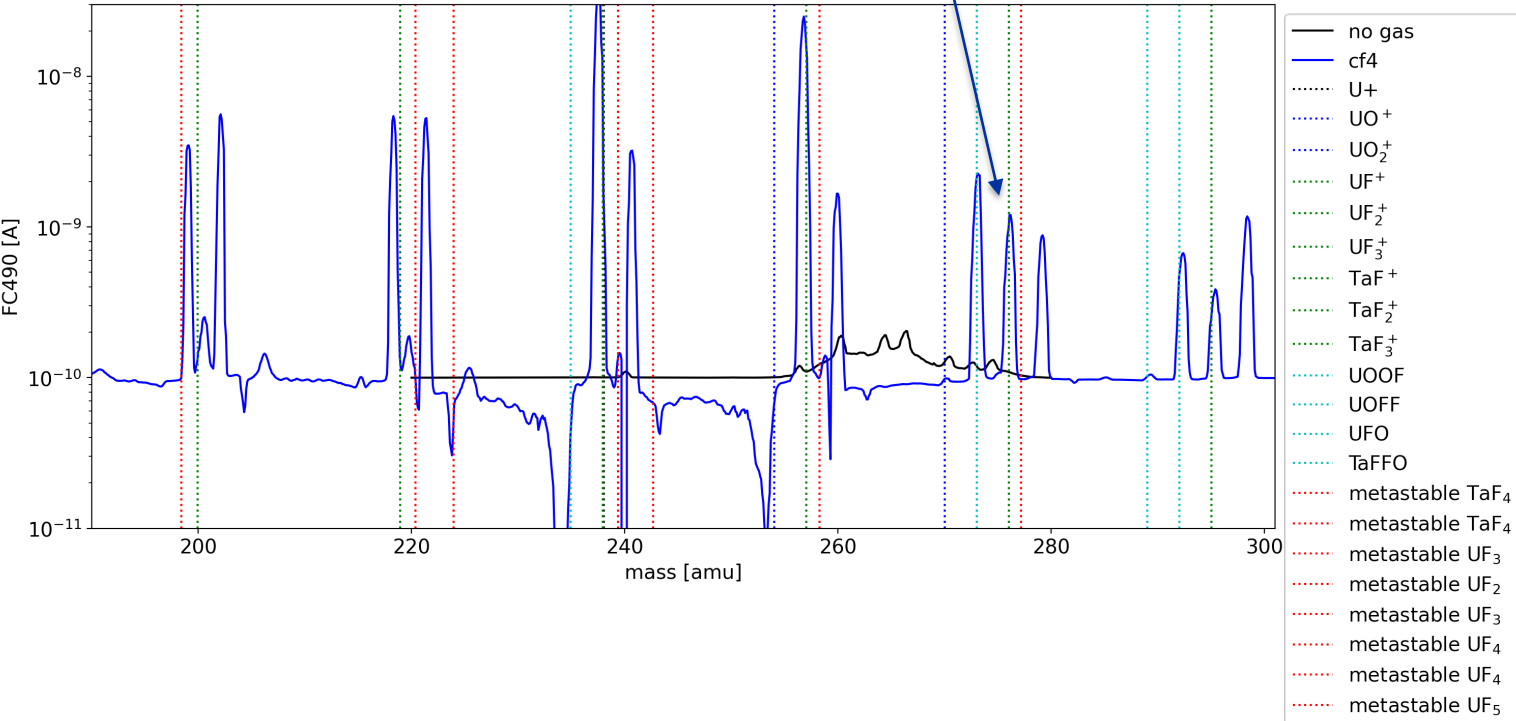
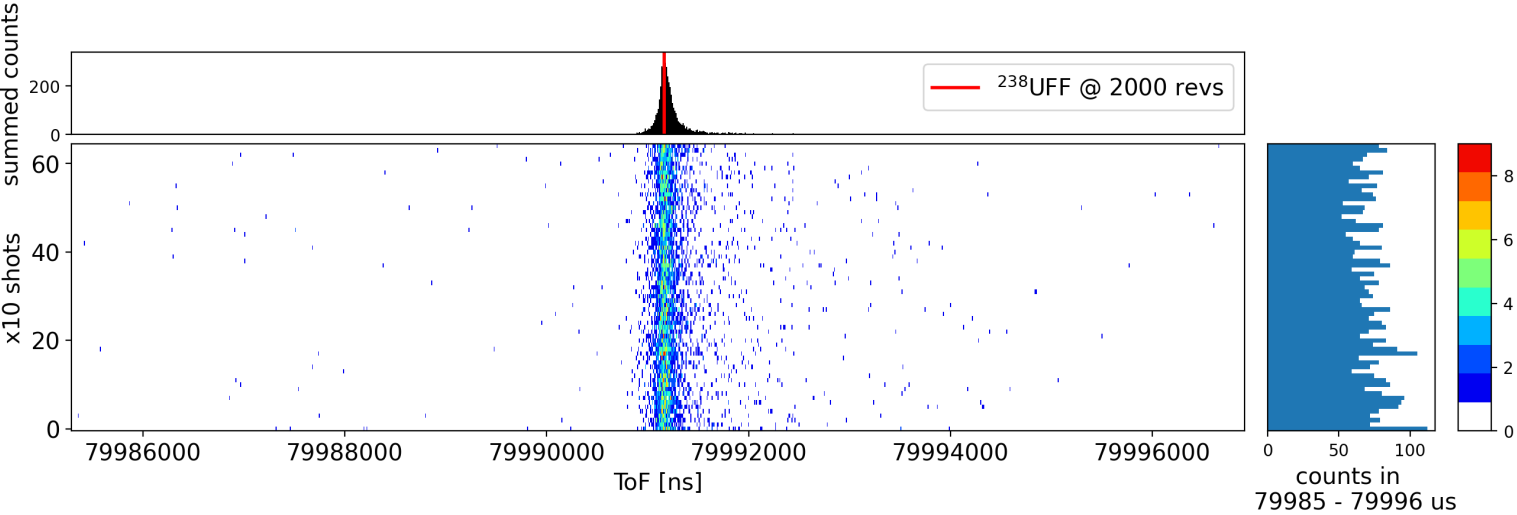


Decomposing mass spectra



Plasma ionization

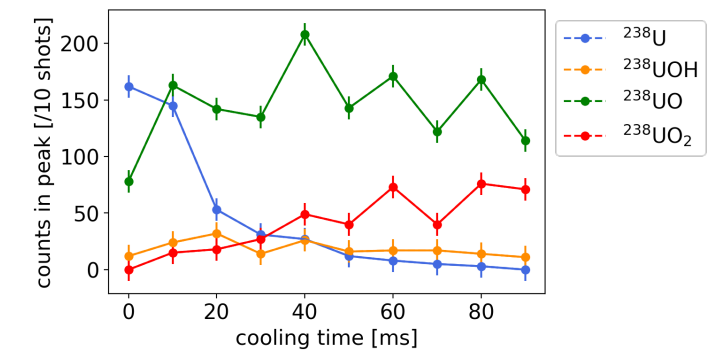
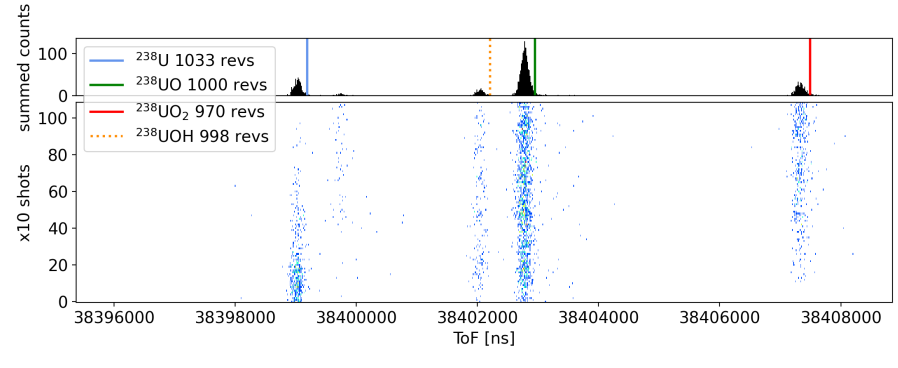
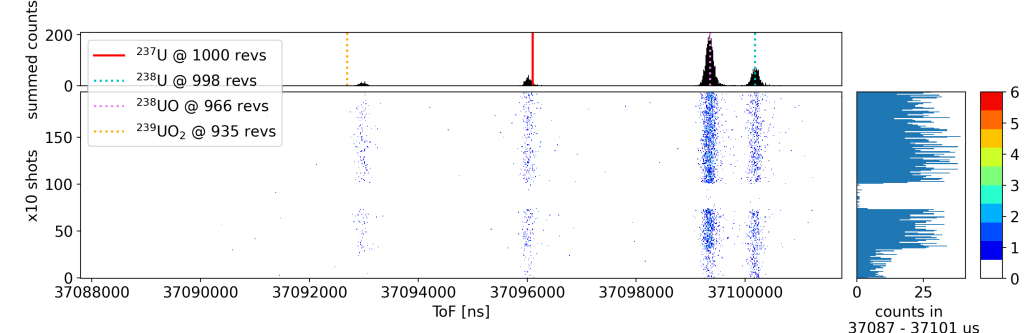
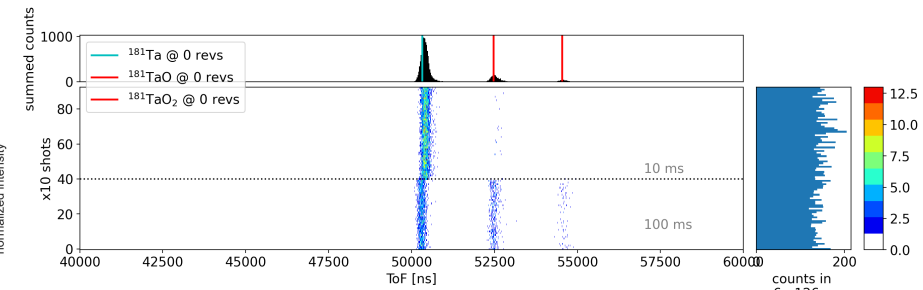
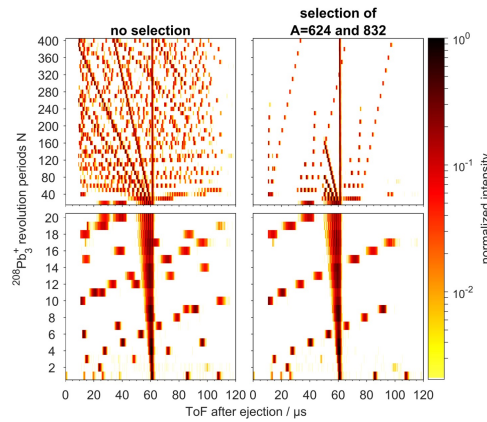
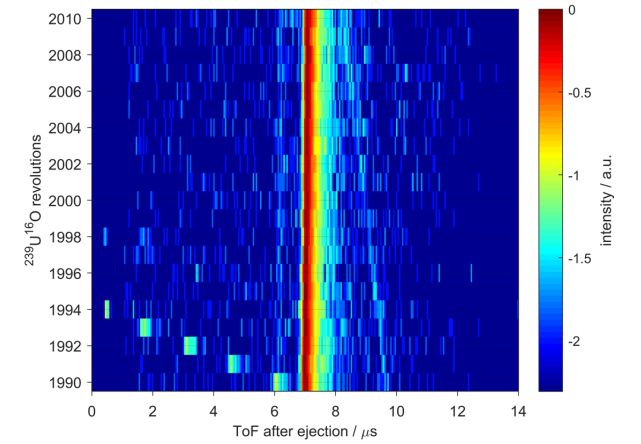
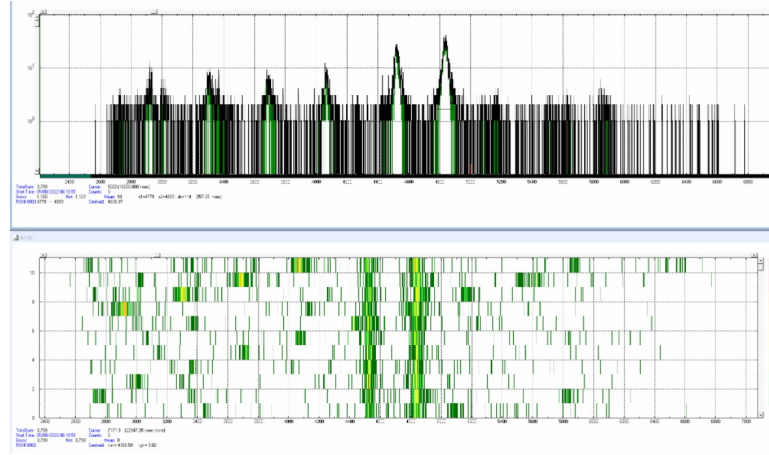
- High efficiency
- Low selectivity
- Dissociation



In-trap formation

UO_x, TaO_x

- ID by ToF and revs vs ToF
- Sideband populations



Fischer,
Schweikhard (2020)
Rev. Sci. Inst. **91**,
023201, DOI
10.1063/1.5131582

In-target formation

AcF_x

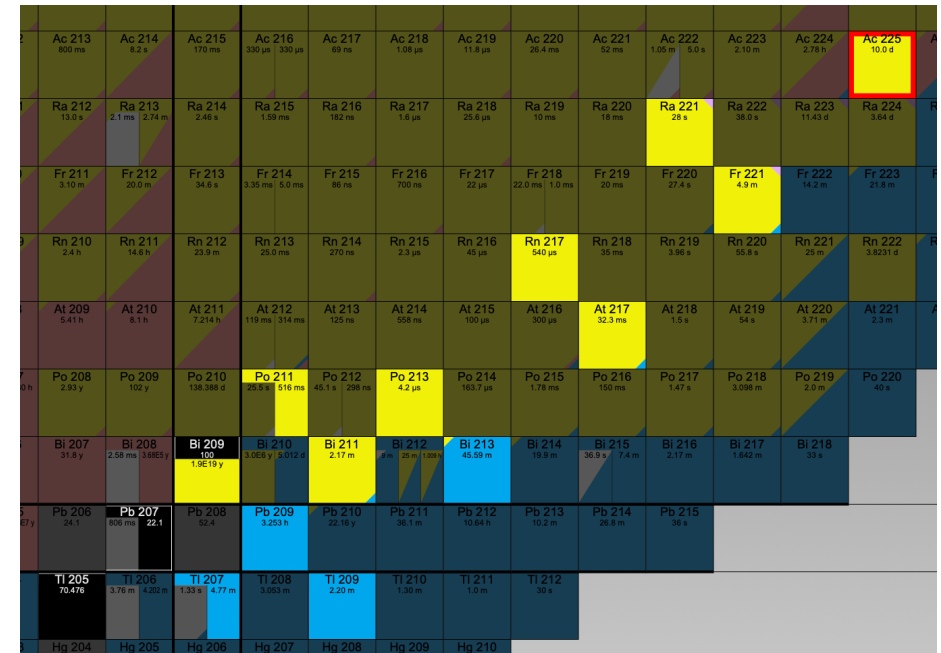
- Ac
 - Enhanced extraction
- AcF
 - Laser spectroscopy
- AcF_x
 - Coordination chemistry
- ²²⁵Ac
 - Radiopharmaceutical “theranostic”
 - Optimized production

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH
 Proposal to the ISOLDE and Neutron Time-of-Flight Committee

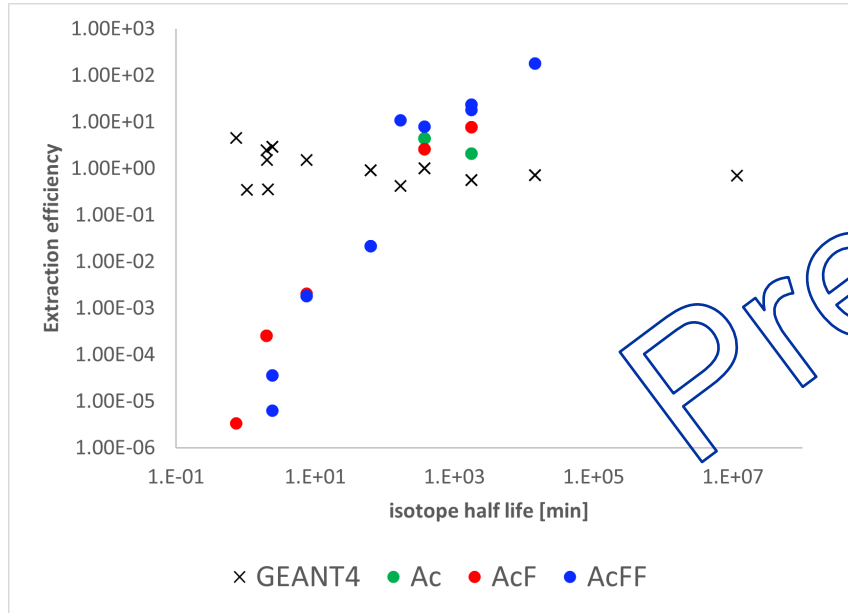
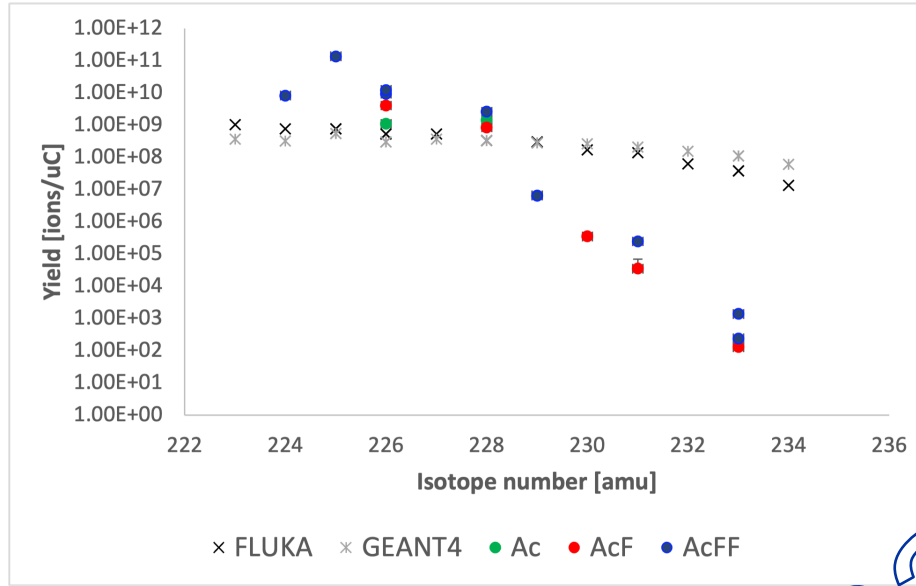
Laser ionization spectroscopy of AcF

September 28, 2021

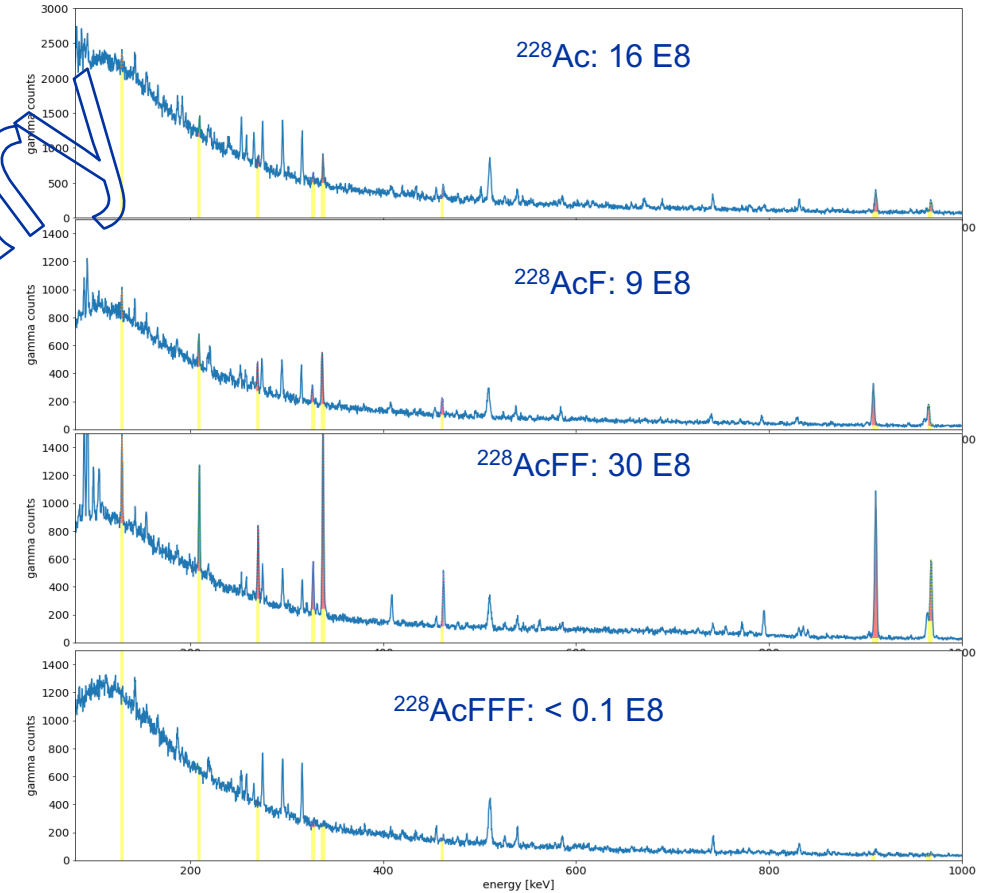
M. Athanasakis-Kaklamanakis^{1,2}, S.G. Wilkins³, M. Au^{4,5}, R. Berger⁶, A. Borschevsky⁷,
 K. Chrysalidis⁸, T.E. Cocolios², R.P. de Groote², Ch.E. Düllmann^{5,9,10},
 K.T. Flanagan^{11,12}, R.F. Garcia Ruiz³, S. Geldhof², R. Heinke⁸, T.A. Isaev¹³,
 J. Johnson², A. Kiuberis⁷, Á. Koszorús¹, L. Lalanne², M. Mougeot¹, G. Neyens²,
 L. Nies^{1,14}, J. Reilly¹¹, S. Rothe⁴, L. Schweikhard¹⁴, A.R. Vernon³, X.F. Yang¹⁵



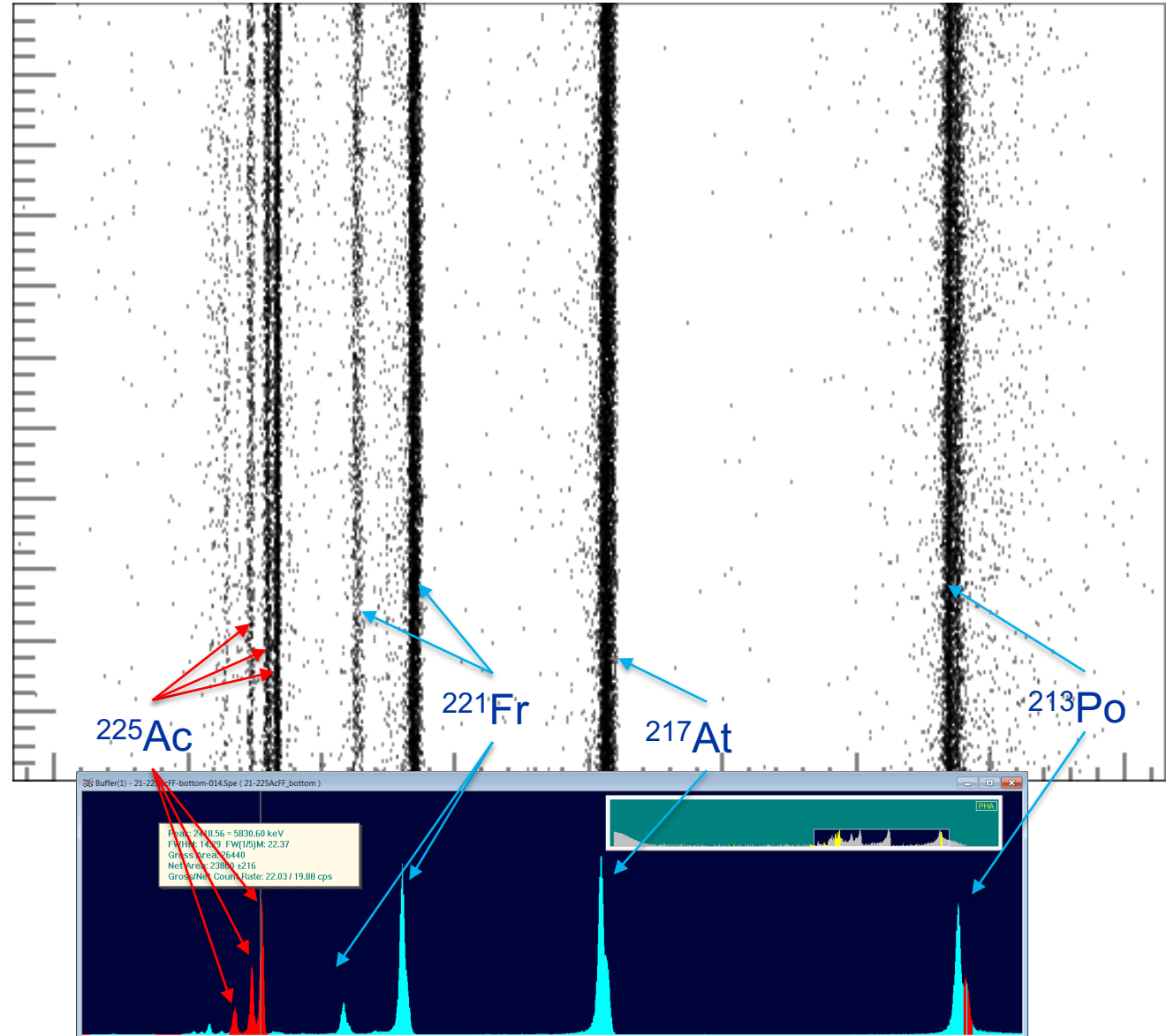
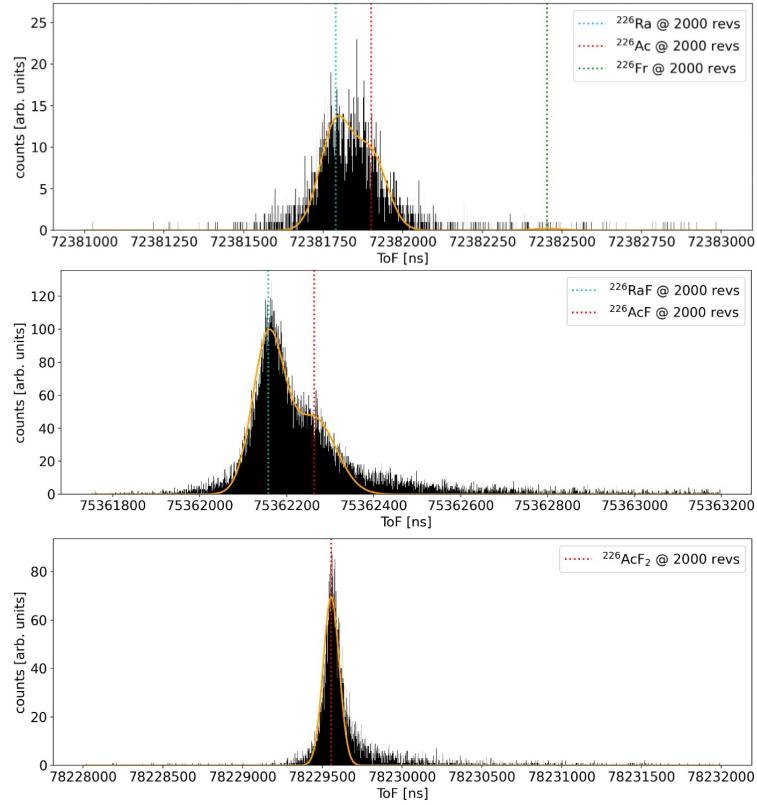
AcF_x



U 225 61 m	U 226 350 m	U 227 1.1 m	U 228 9.1 m	U 229 58 m	U 230 20.8 d	U 231 4.2 d	U 232 69.8 y	U 233 1.59265 y	U 234 2.44525 y	U 235 7.04e8 y	U 236 2.37e7 y	U 237 6.75 d	U 238 4.468e9 y
Pa 224 664 m	Pa 225 1.7 s	Pa 226 1.8 m	Pa 227 38.3 m	Pa 228 22 h	Pa 229 1.52 d	Pa 230 17.4 d	Pa 231 3.27054 y	Pa 232 1.31 d	Pa 233 27.0 d	Pa 234 117 m	Pa 235 24.3 m	Pa 236 9.1 m	Pa 237 8.7 m
Th 223 600 m	Th 224 1.05 s	Th 225 8.72 m	Th 226 30.57 m	Th 227 18.718 d	Th 228 1.9127 y	Th 229 29 d	Th 230 7.5464 y	Th 231 1.0633 d	Th 232 1.405e10 y	Th 233 22.3 m	Th 234 24.09 d	Th 235 6.9 m	Th 236 37.5 m
Ac 222 1.05 m	Ac 223 2.10 m	Ac 224 2.78 h	Ac 225 10.0 d	Ac 226 1.224 d	Ac 227 21.773 y	Ac 228 6.15 h	Ac 229 1.046 h	Ac 230 2.63 m	Ac 231 7.5 m	Ac 232 1.98 m	Ac 233 2.42 m	Ac 234 44 s	Ac 235 40 s
Ra 221 28 s	Ra 222 38.0 s	Ra 223 11.43 d	Ra 224 3.64 d	Ra 225 14.8 d	Ra 226 1.600e3 y	Ra 227 42.2 m	Ra 228 5.75 y	Ra 229 4.0 m	Ra 230 1.55 h	Ra 231 1.72 m	Ra 232 2.42 m	Ra 233 30 s	Ra 234 30 s
Fr 220 27.4 s	Fr 221 4.9 m	Fr 222 14.2 m	Fr 223 21.8 m	Fr 224 3.33 m	Fr 225 4.0 m	Fr 226 49 s	Fr 227 2.47 m	Fr 228 38 s	Fr 229 50.2 s	Fr 230 19.1 s	Fr 231 17.6 s	Fr 232 5 s	

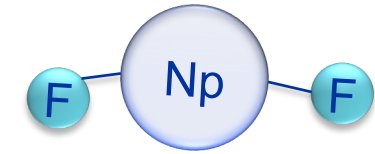


AcF_x



	Yield [ions / uC p ⁺]	Efficiency [%]
sample: $^{229}\text{AcFF}$	1.6 (1) E5	0.05 (2)
sample: $^{225}\text{AcFF}$	1.93 (0.05) E7	2.5 (0.04)

Outlook: Other actinides?

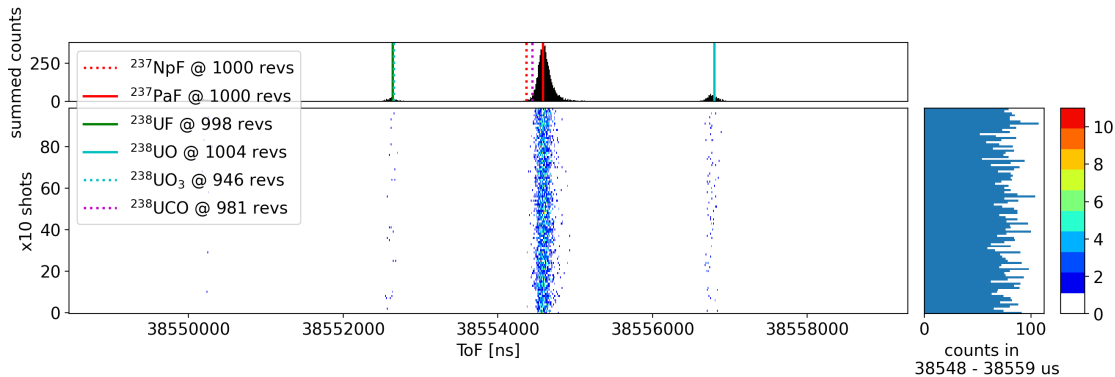


Actinide fluorides

- NpF_x
- PuF_x

Next up

- Improving statistics



	Yield [ions / micro C p ⁺]	Efficiency [%]
sample 12: $^{238}\text{NpFF}$	1.6 (0.2) E3	0.039 (5)
sample 13: ^{234}Np	< 3.5	
sample 24: ^{234}NpF	1.4 E2	0.00013
sample 14: $^{234}\text{NpFF}$	3.8 (1) E2	0.037 (10)
sample 23: $^{234}\text{NpFFF}$	< 1.2 E2	< 0.0001

Conclusion

1. Surface, plasma beam compositions for actinide mass region
2. In-trap formation (UO_x , TaO_x)
3. In-target formation : AcF_x production, ID and measurement
4. Molecular spectroscopy of AcF : ongoing!
5. Outlook: AnF_x

Acknowledgements

IS706 experimental team

Morning	Evening	Night
Agi	Michail	Shane
Mia	Jordan	Louis
Julius	Carlos	Kara
Kieran	Abi	Fabian
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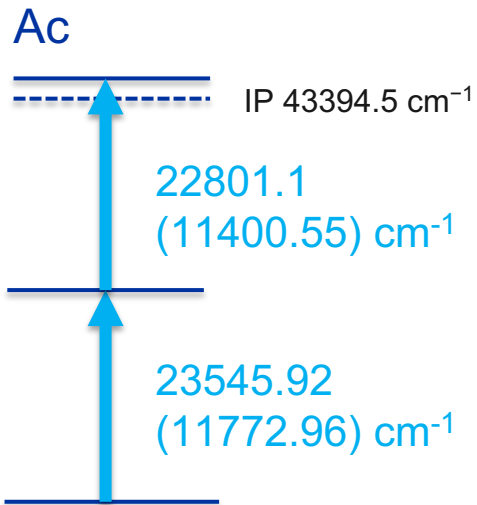




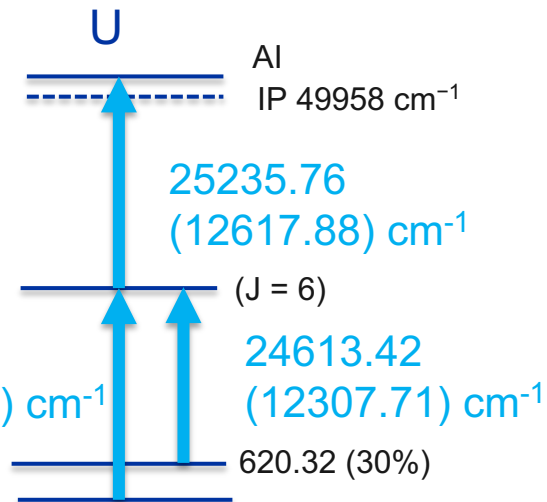
Thank you!

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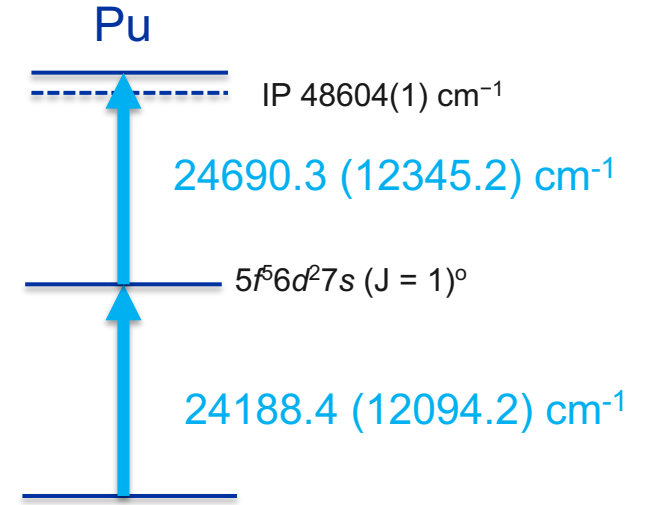
Laser ionization schemes used



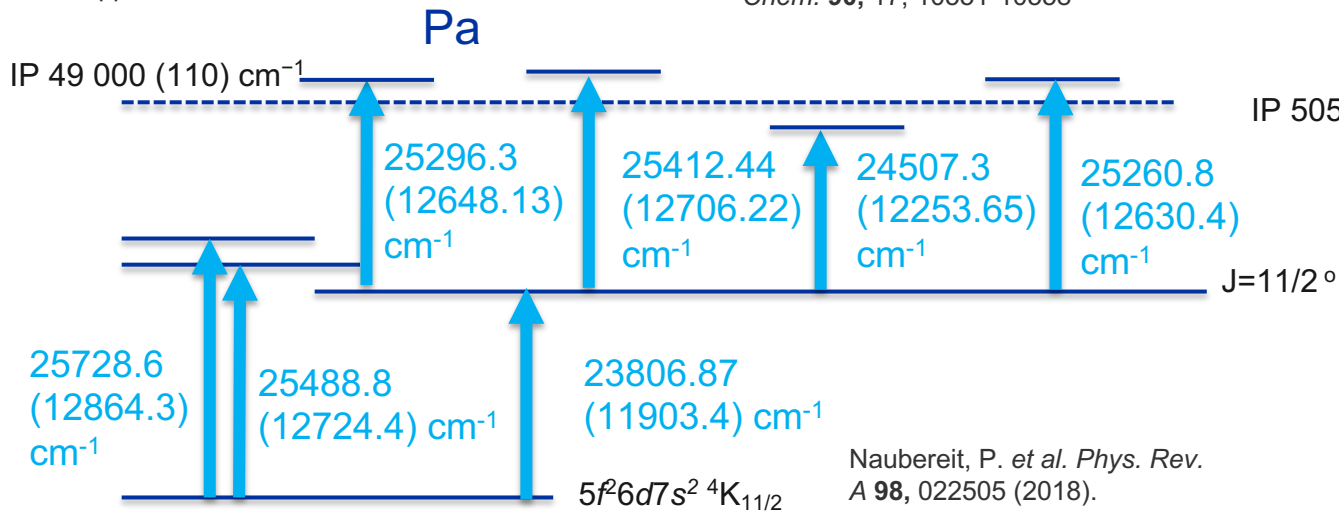
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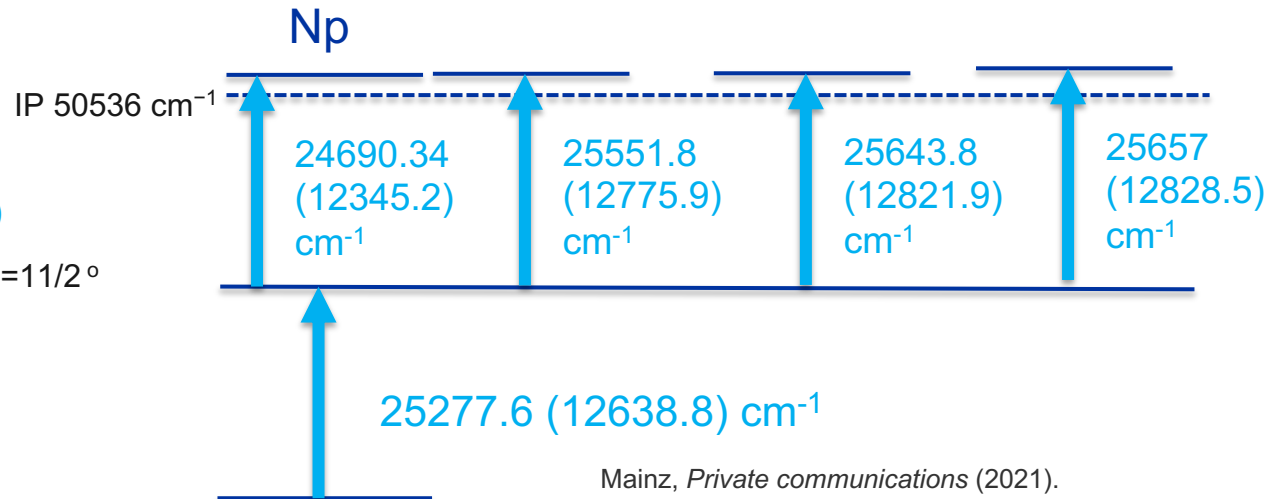
Savina, M. *et al.* (2018) *Anal. Chem.* **90**, 17, 10551-10558



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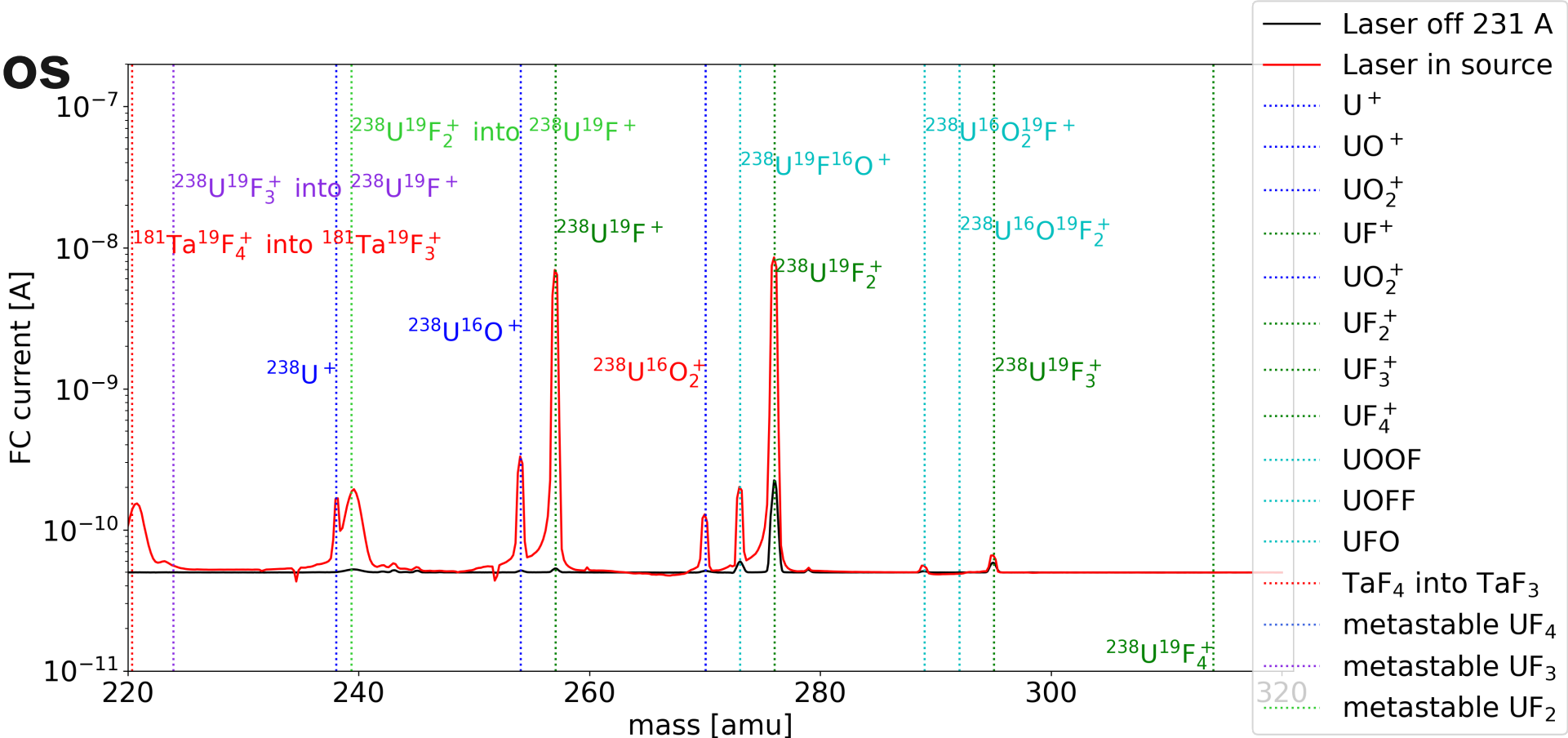


Mainz, *Private communications* (2021).

Surface ionization vs laser enhancement

Outgassing over time

Sideband ratios



AcF_x

Ac⁺, AcF⁺, AcFF⁺, AcFO⁺

