

Production of actinide atomic and molecular ion beams at CERN-ISOLDE

LISA conference 2024

M. Au | CERN SY-STI-RBS



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Outline: Actinide beams at CERN-ISOLDE

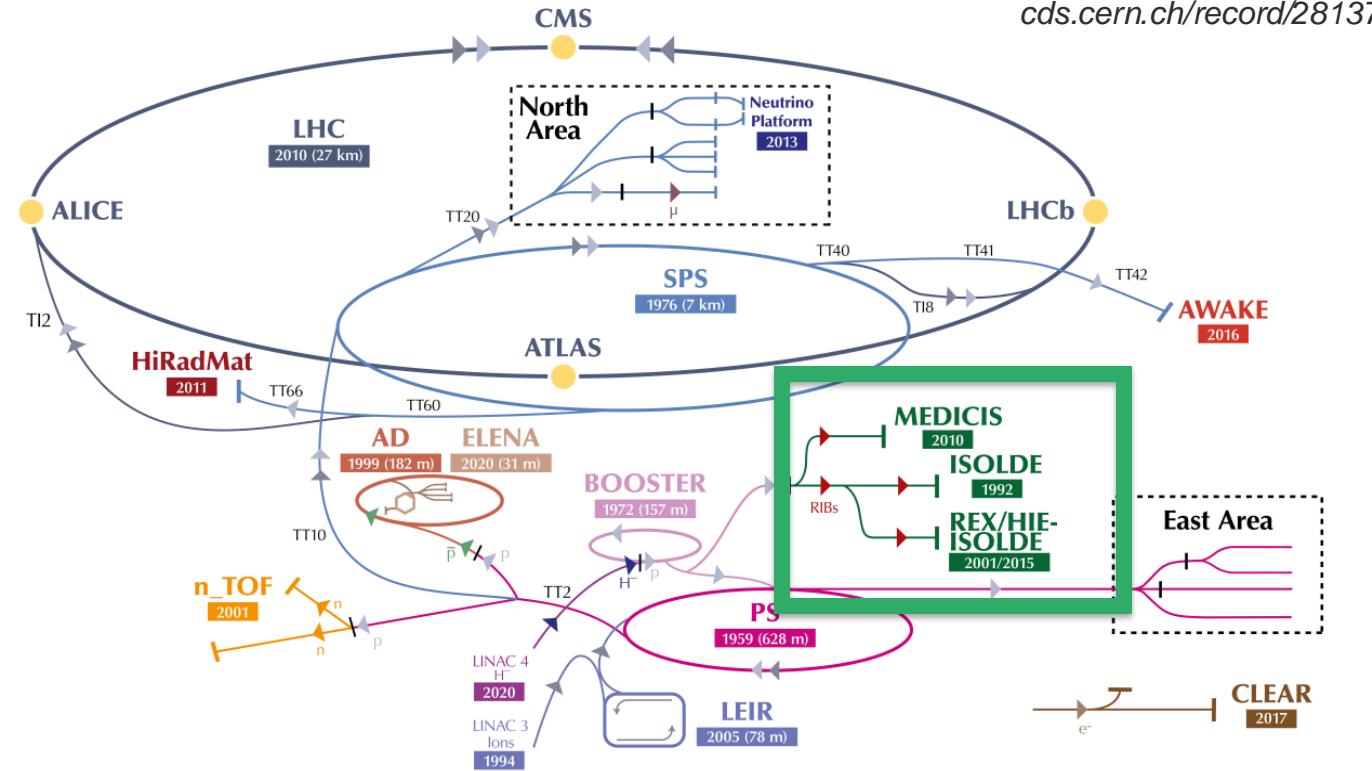
CERN-PHOTO-202206-116
cds.cern.ch/record/2813716

1 CERN-ISOLDE and ISOL

2 Atomic actinide beams

3 Radioactive molecules

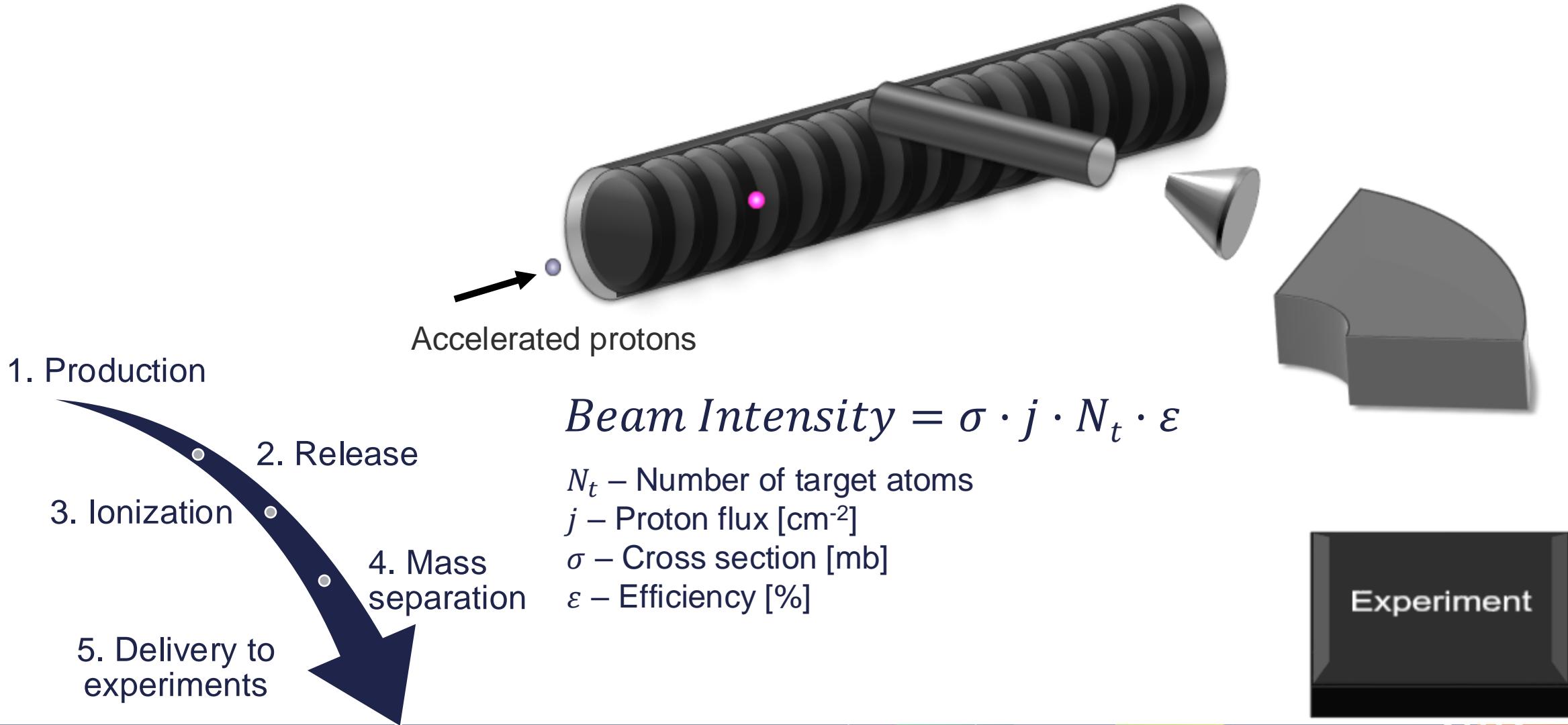
4 Offline developments



► 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

The Isotope Separation On-Line (ISOL) method



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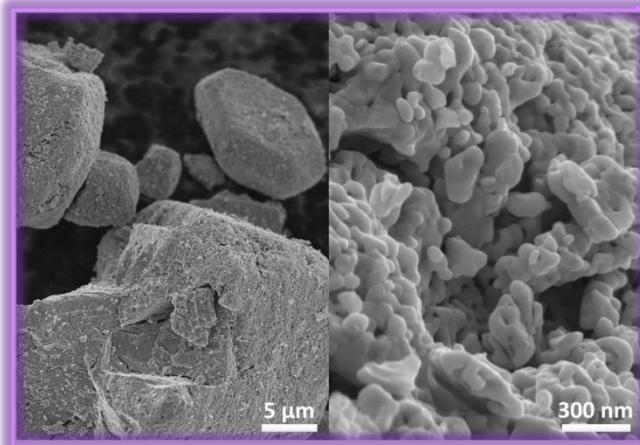
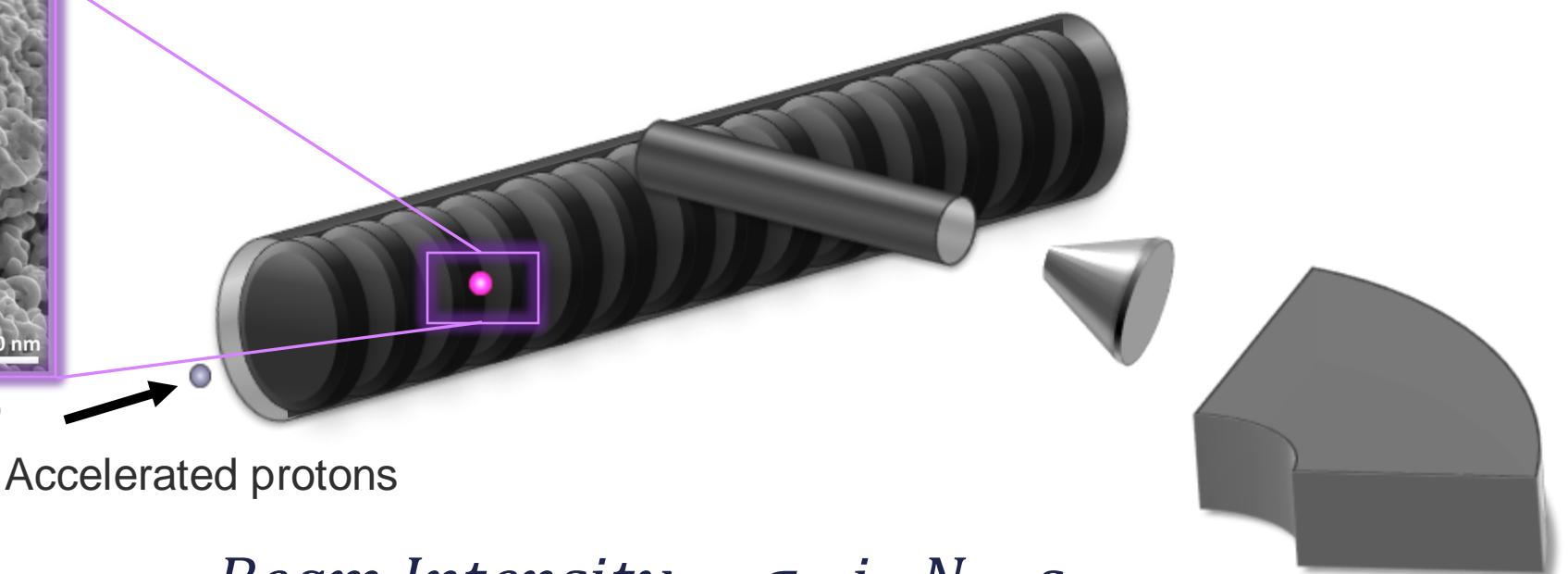


Figure published in Ramos et al., (2020)
NIM B 463, 201

1. Production
2. Release
3. Ionization
4. Mass separation
5. Delivery to experiments



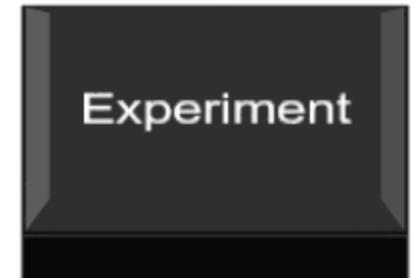
$$\text{Beam Intensity} = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms

j – Proton flux [cm^{-2}]

σ – Cross section [mb]

ε – Efficiency [%]



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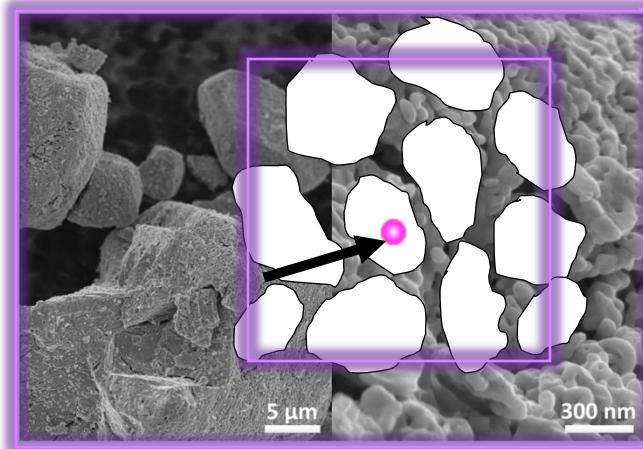
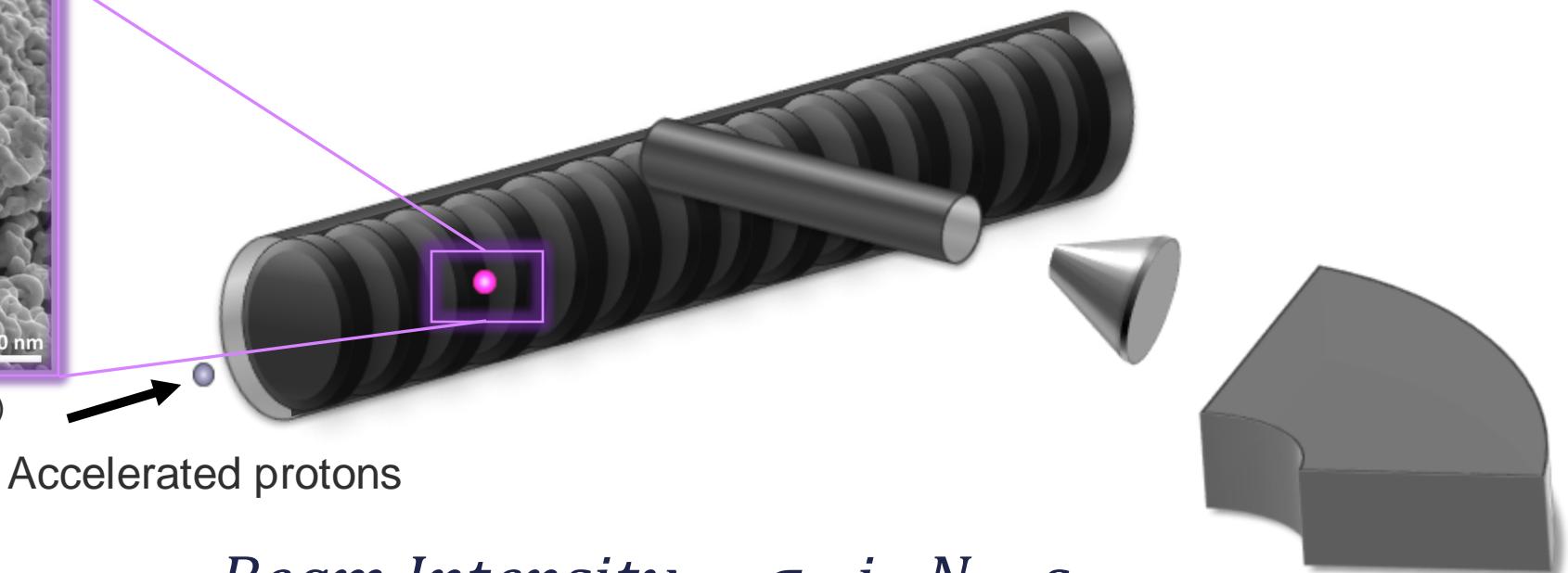


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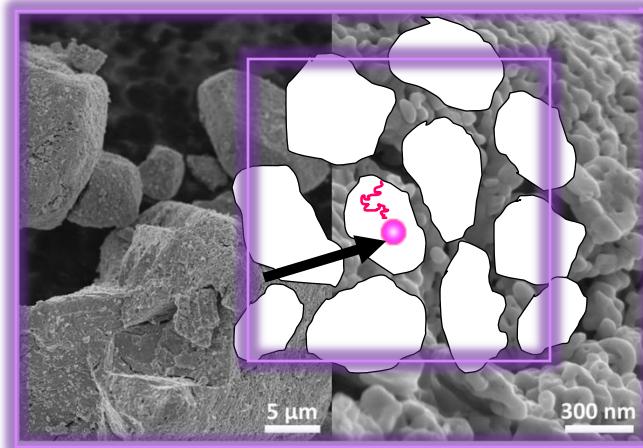


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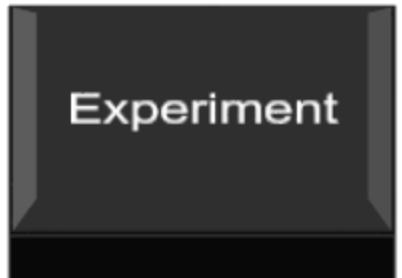
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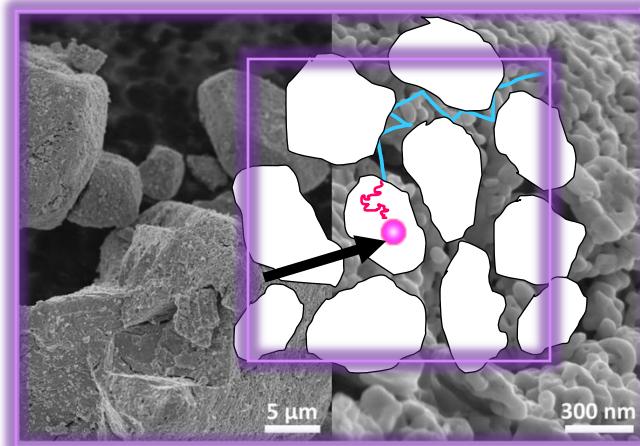
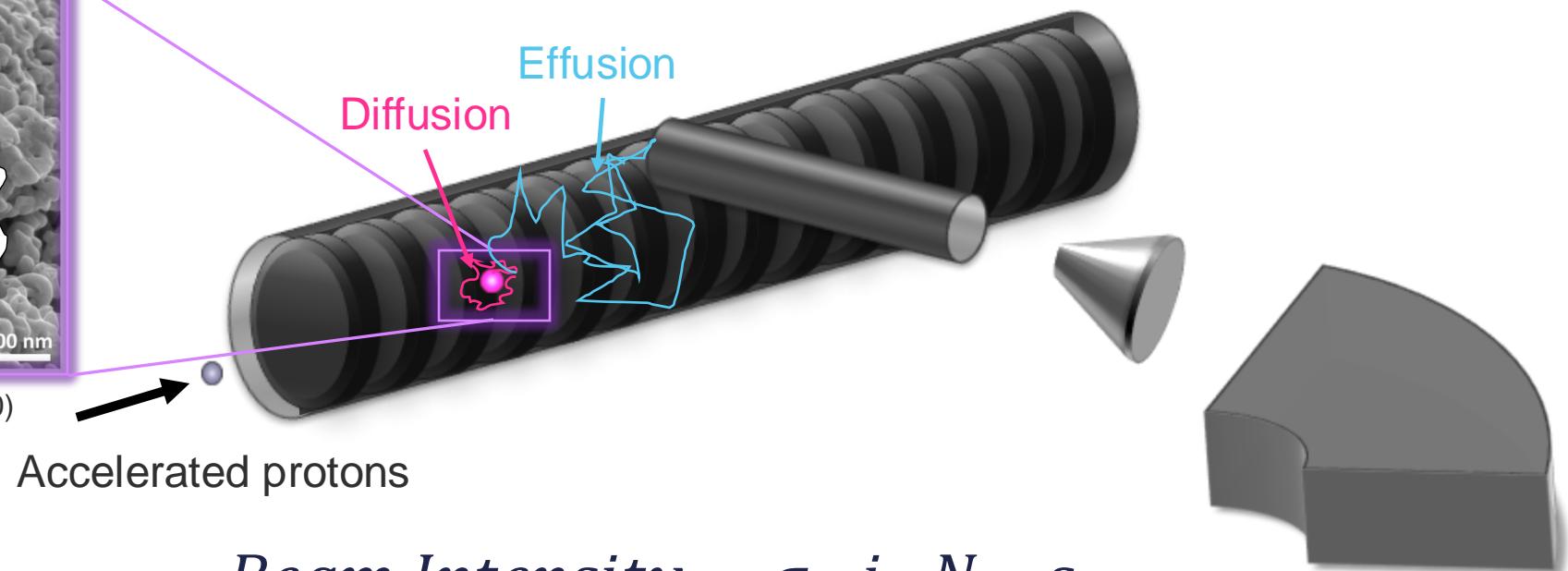


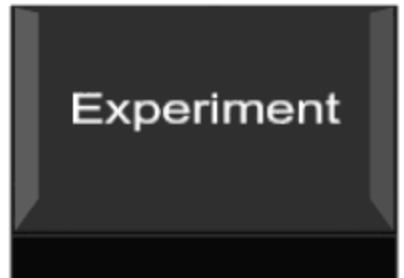
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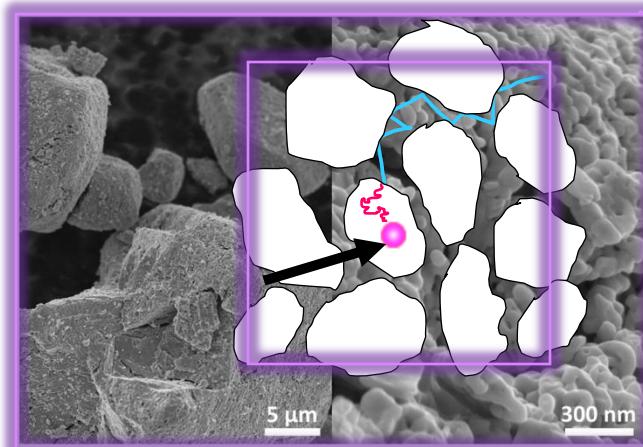


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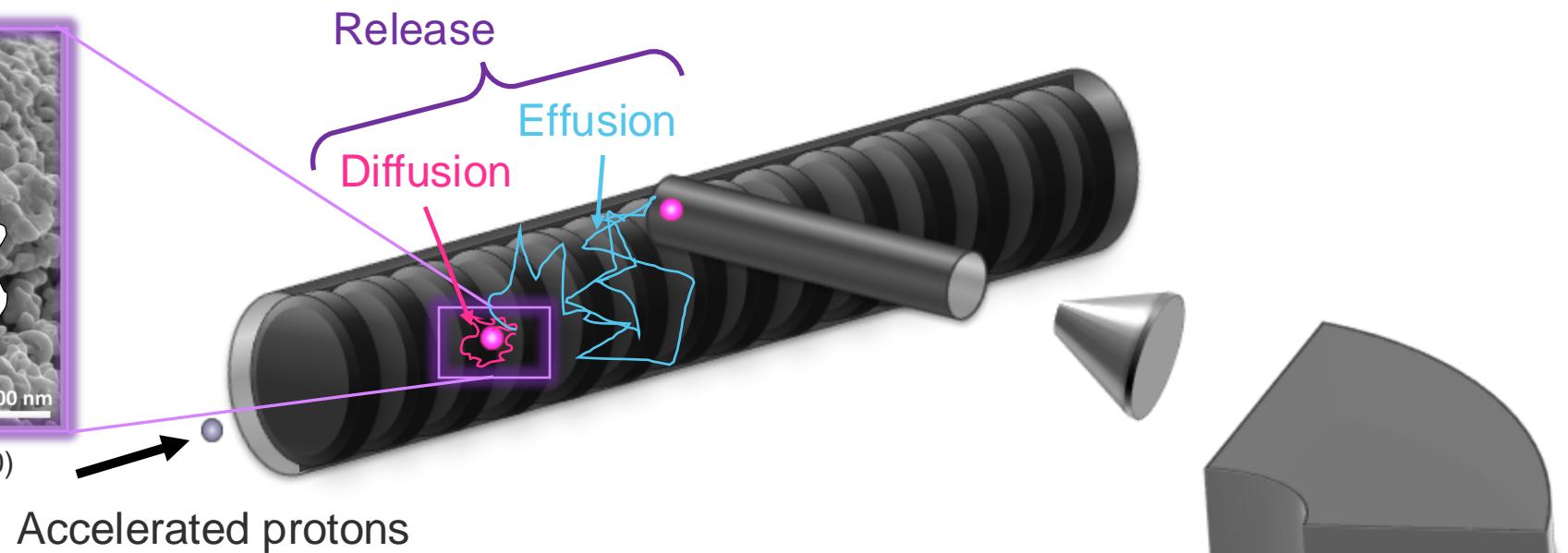
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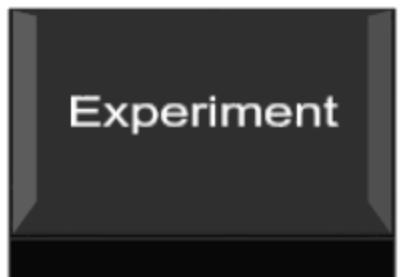
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$$\varepsilon = \varepsilon_{\text{diff}} \varepsilon_{\text{eff}}$$



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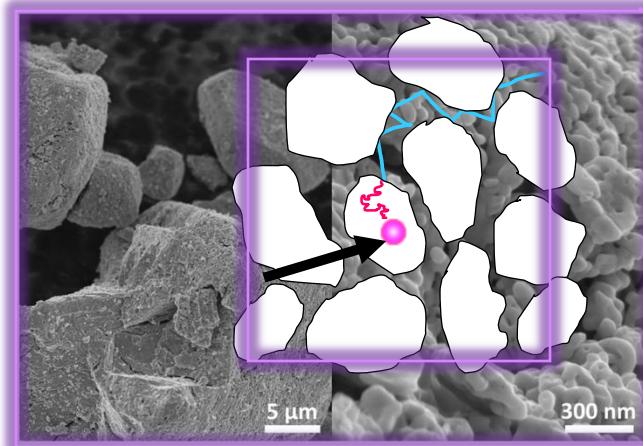


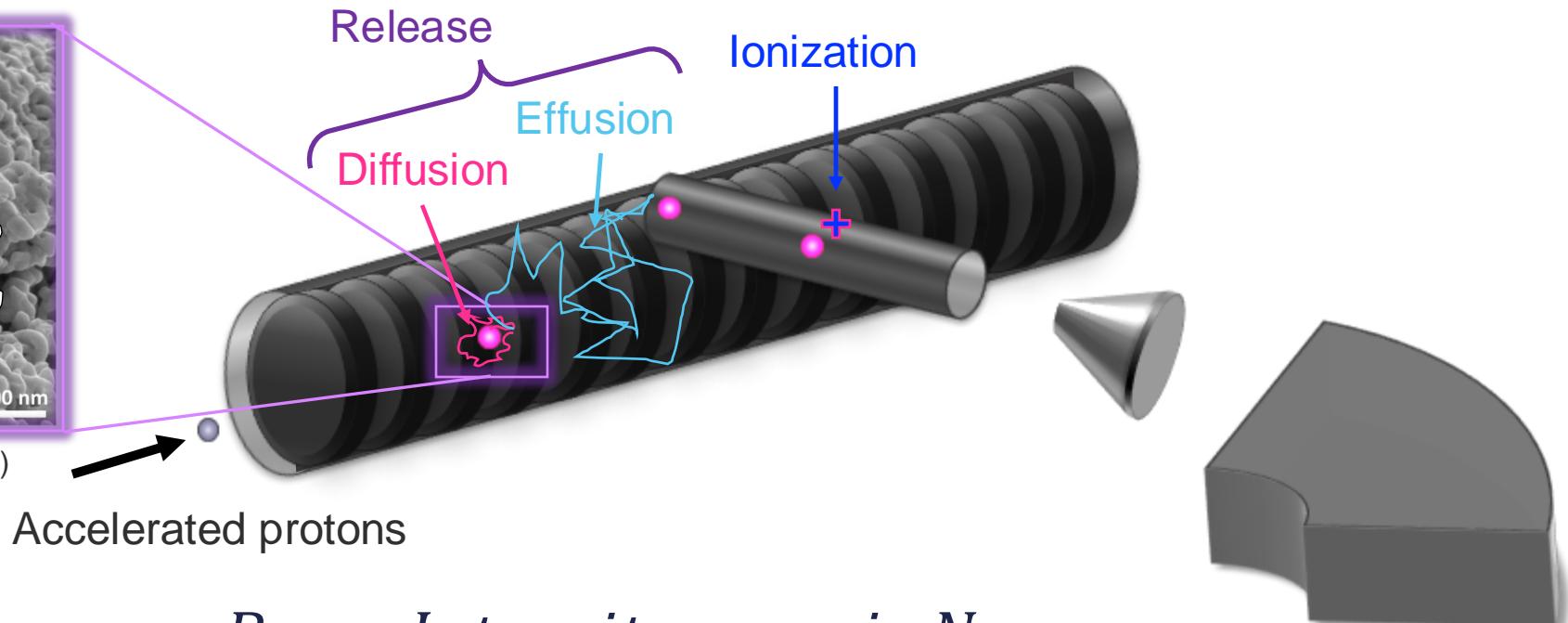
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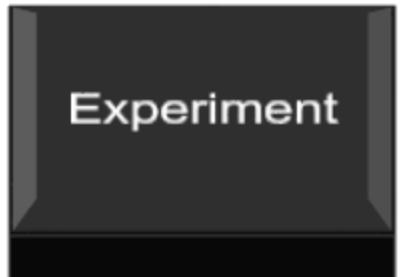
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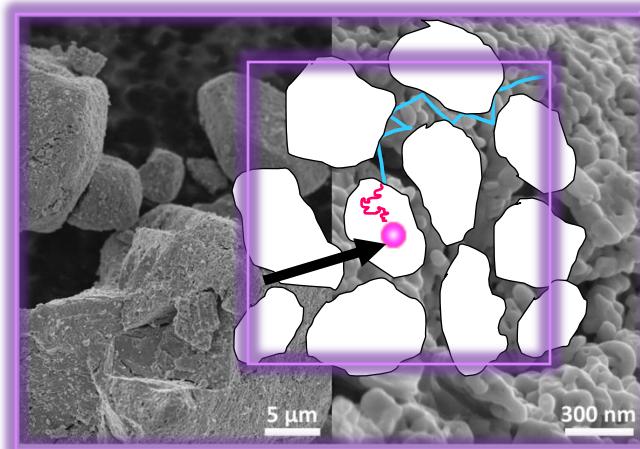
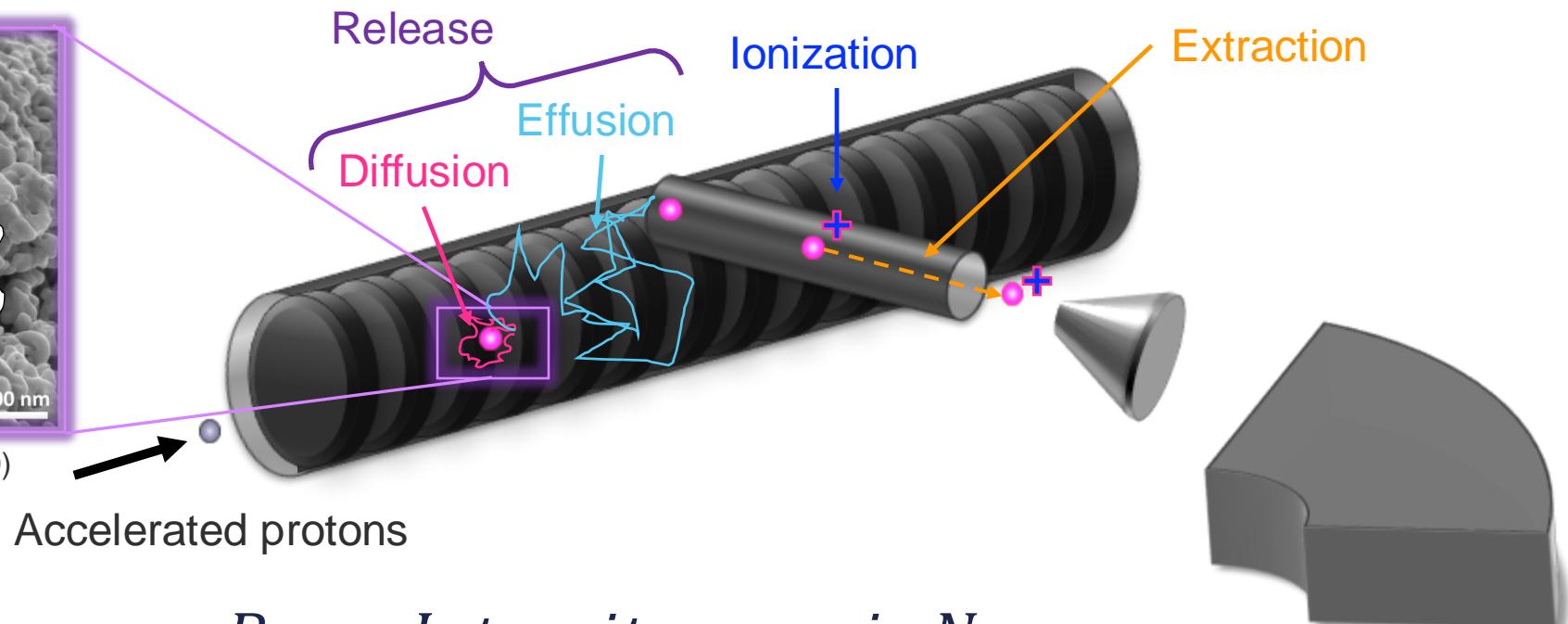


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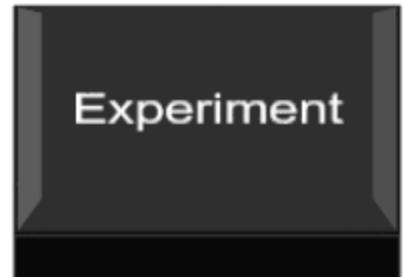
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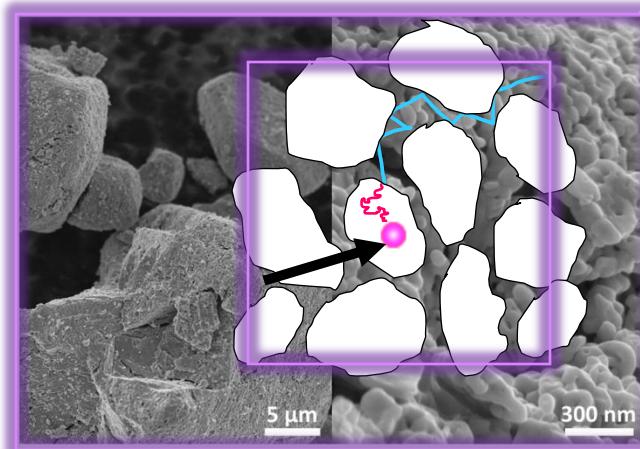


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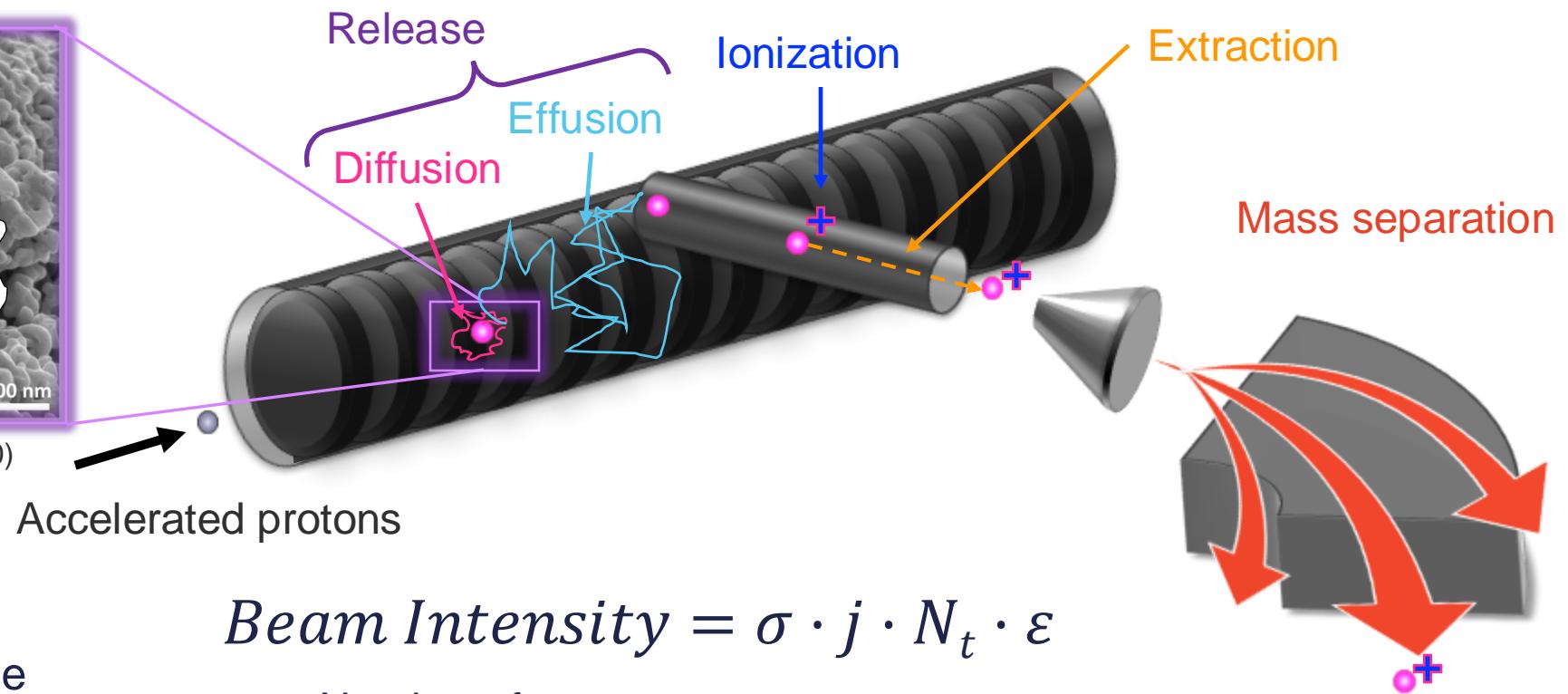
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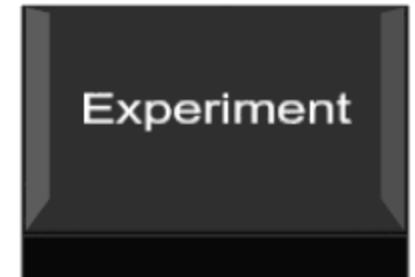
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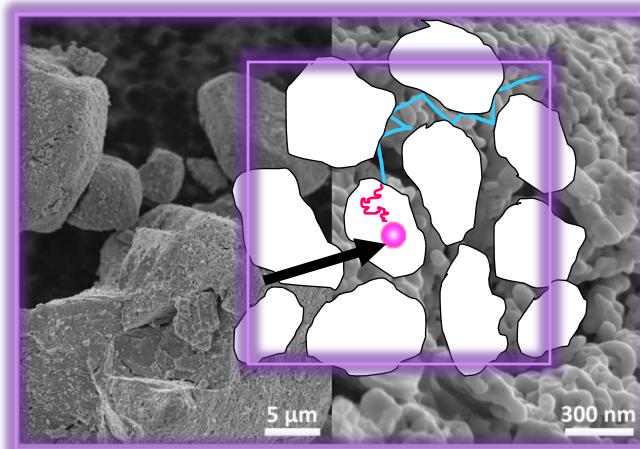


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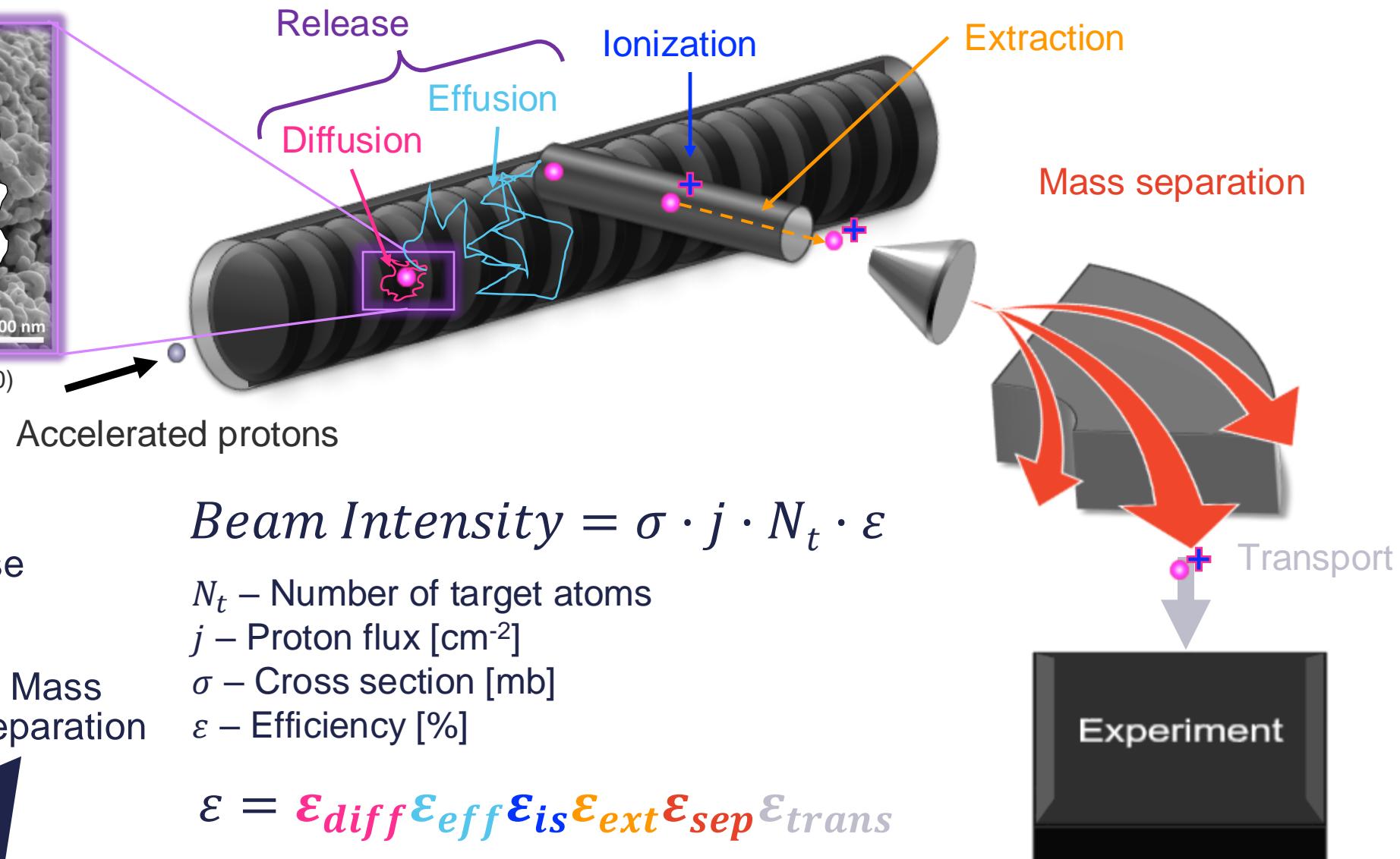
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ISOL step 1: Production

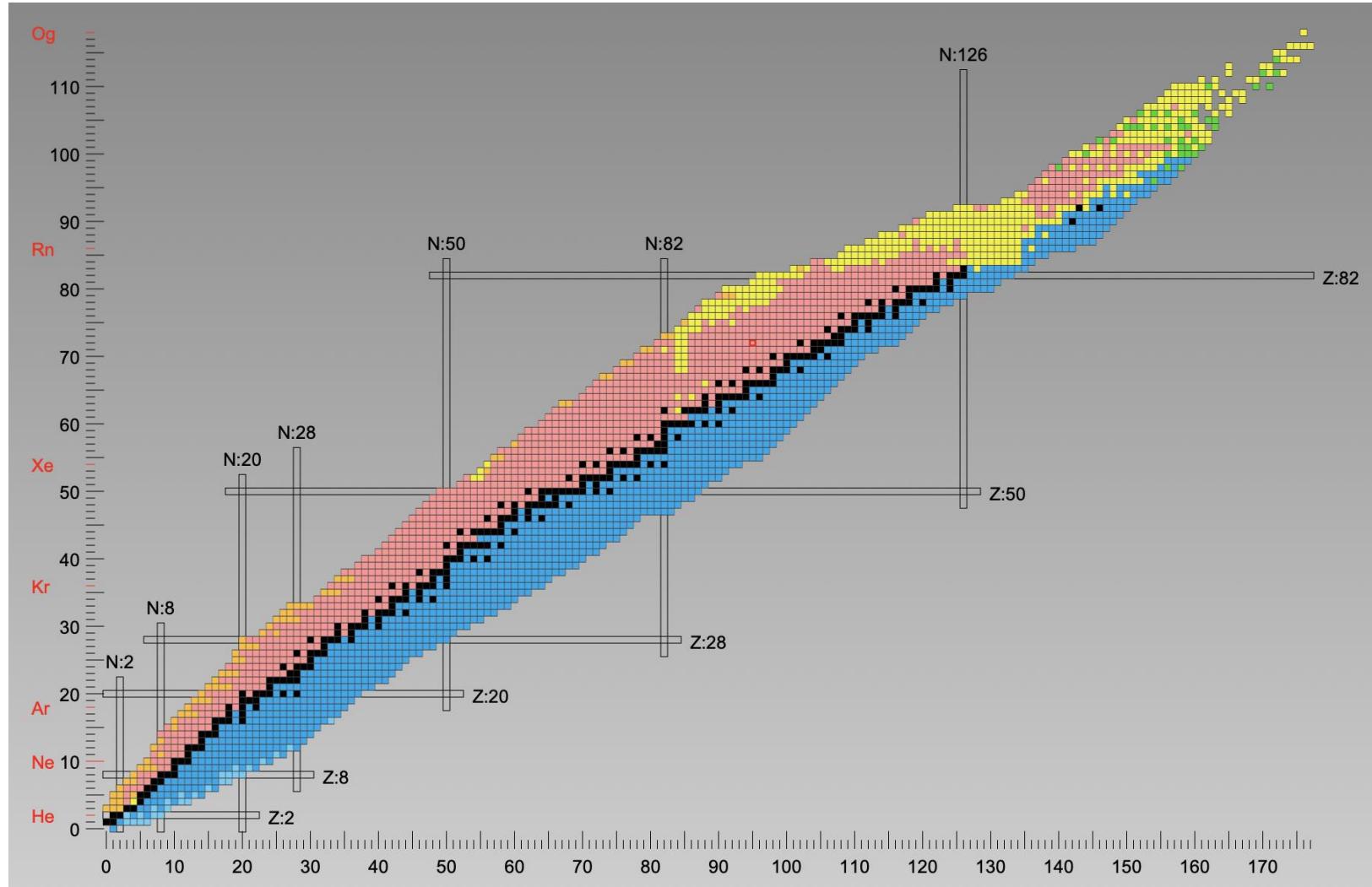
Target selection

- Cross sections
- Bulk
- Half-lives



At ISOLDE

- 1.4-GeV p
- ^{232}Th , ^{238}U



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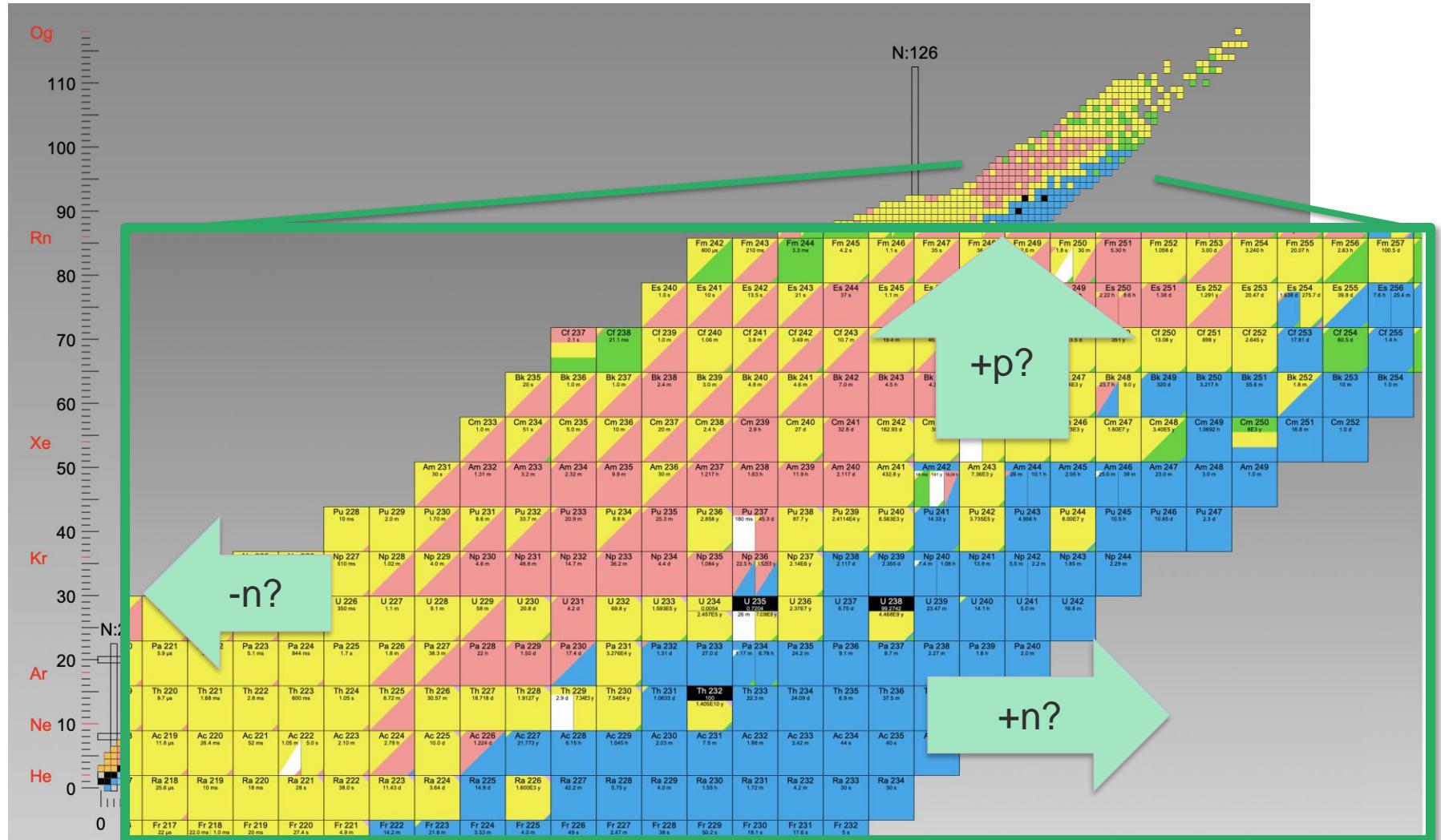
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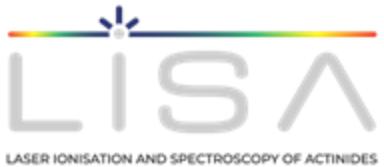


ISOL step 1: Production

Ballof et al. (2020) NIM B 463, 211-215
cern.ch/isolde-yields

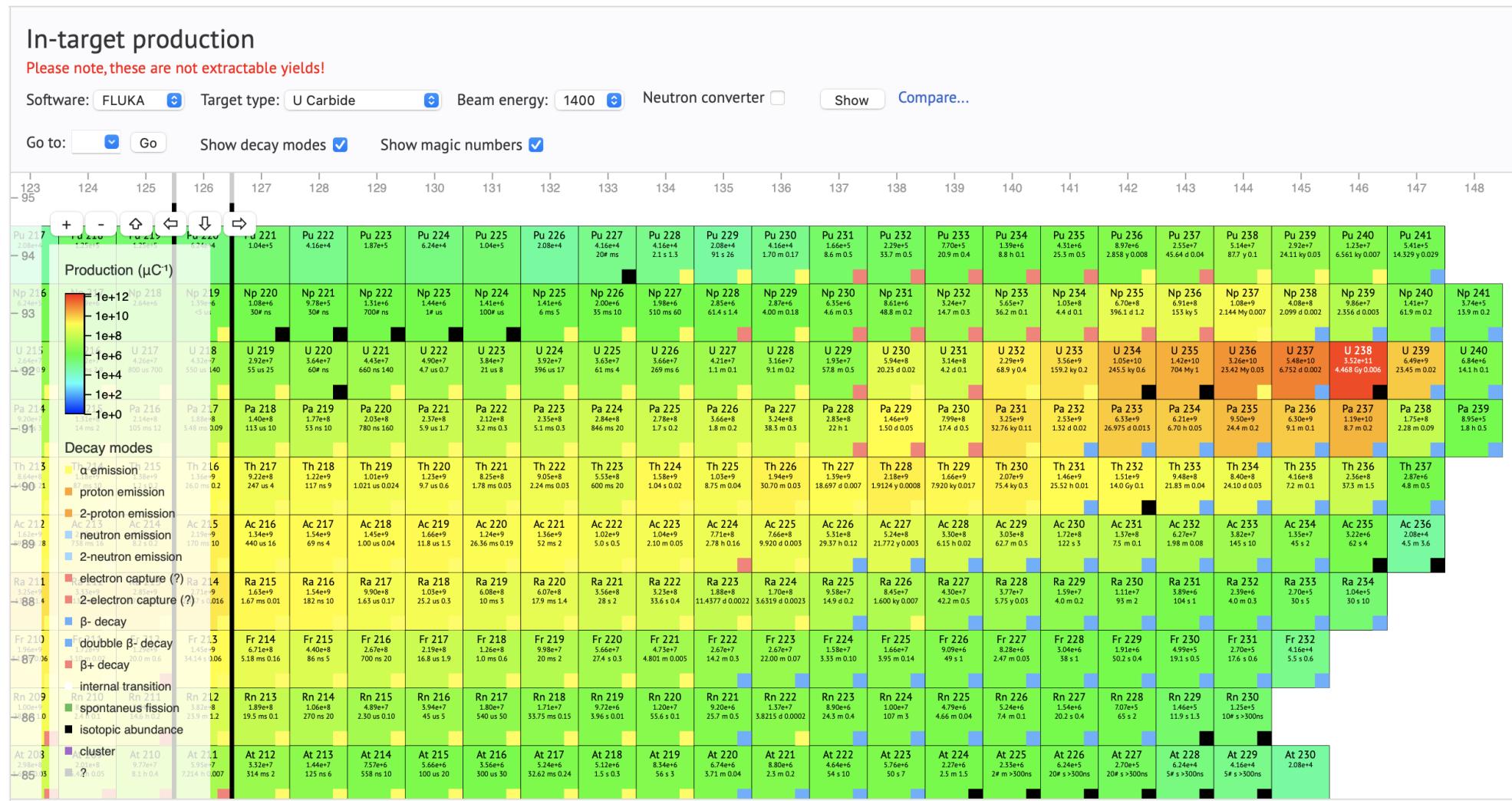
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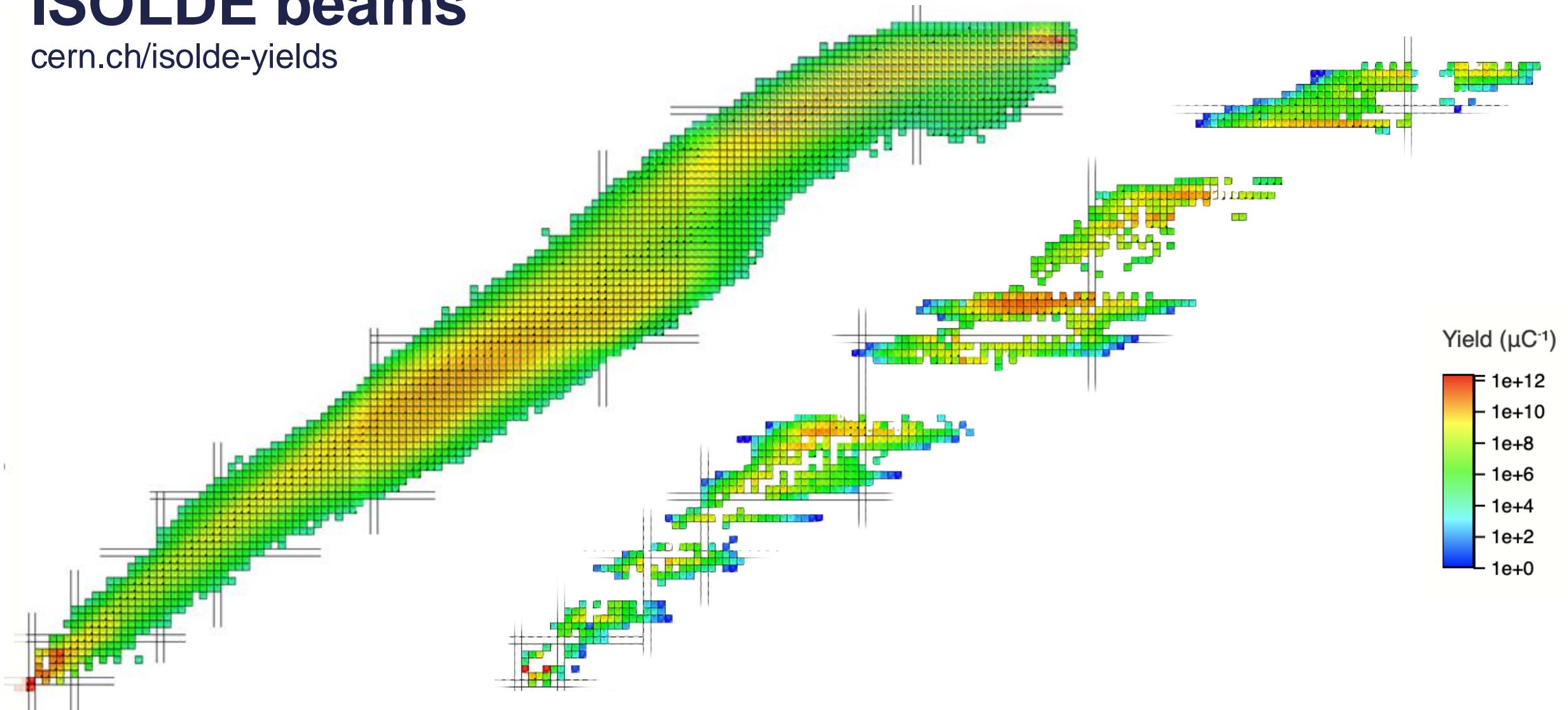
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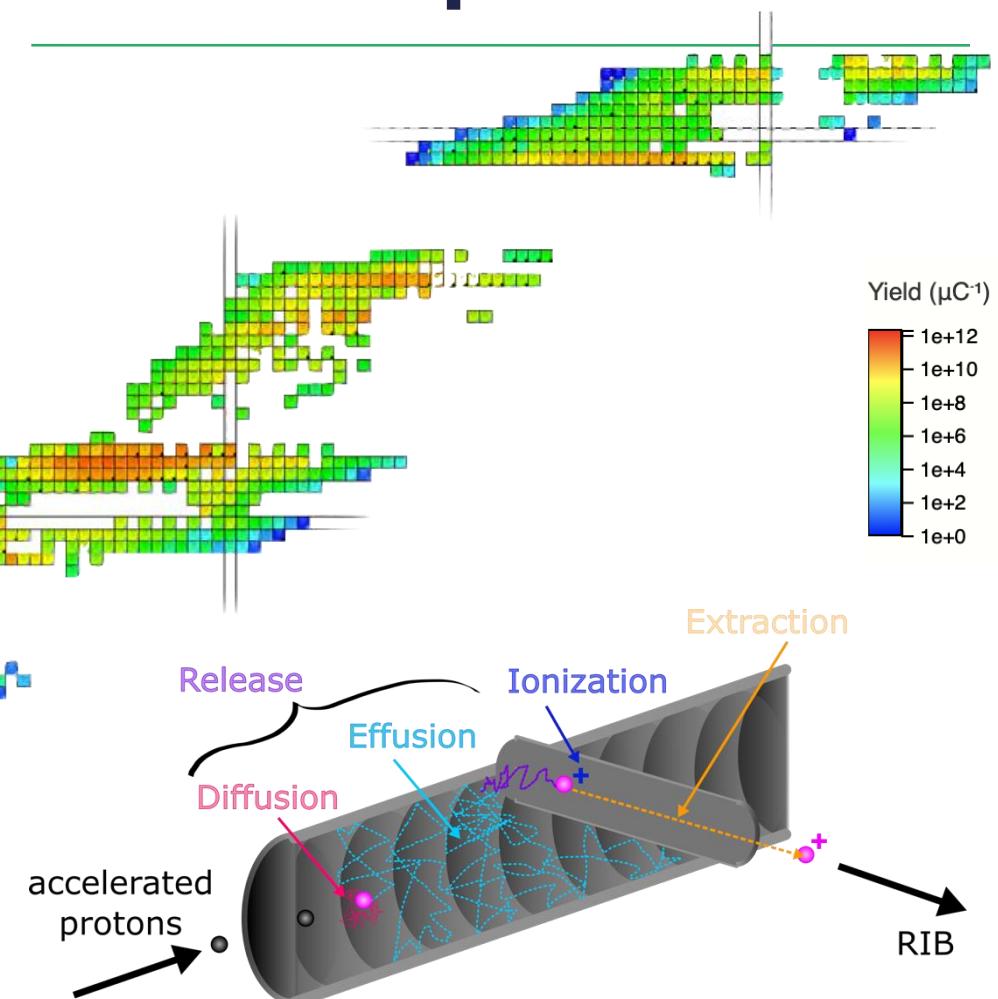
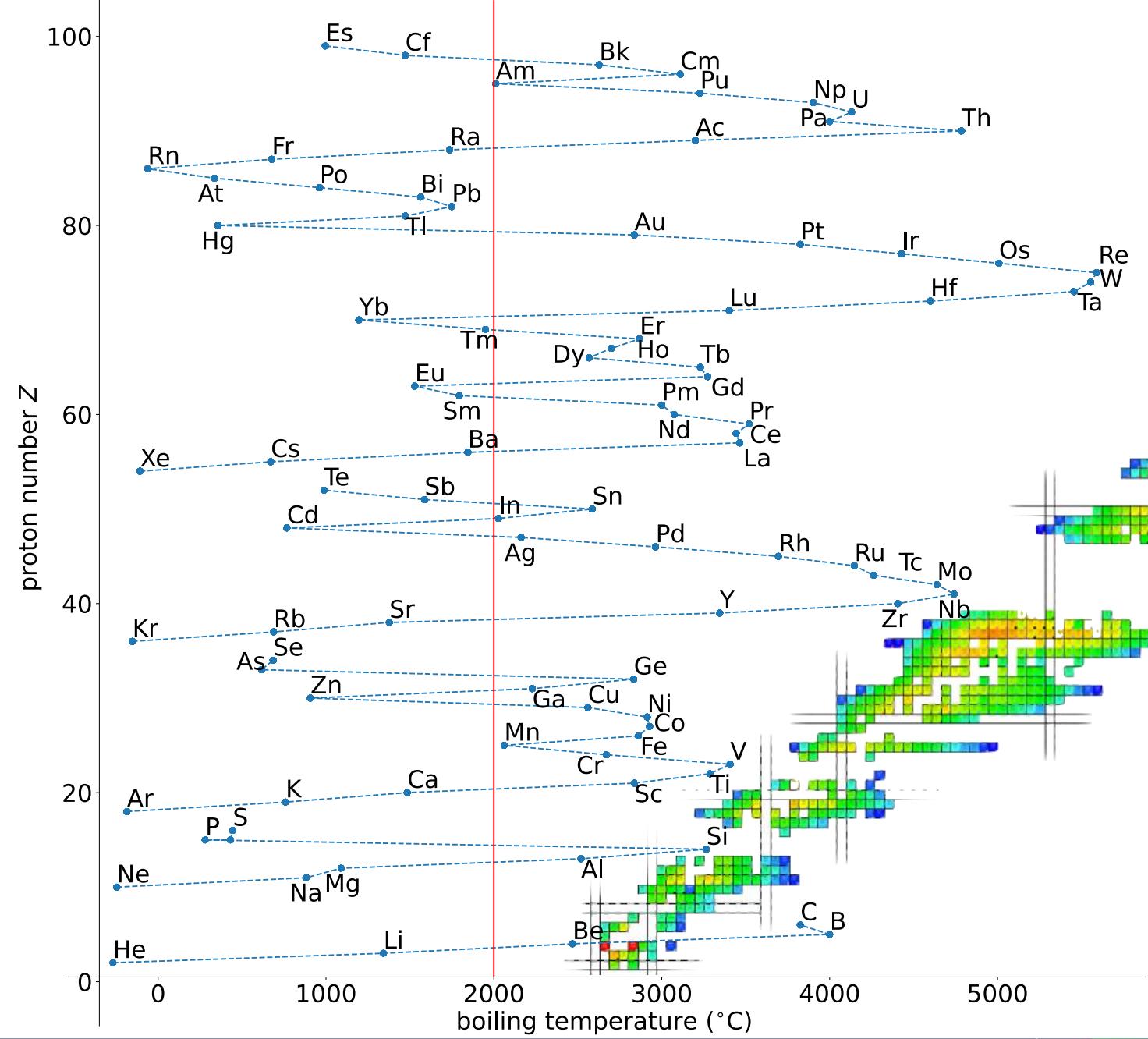


ISOLDE beams

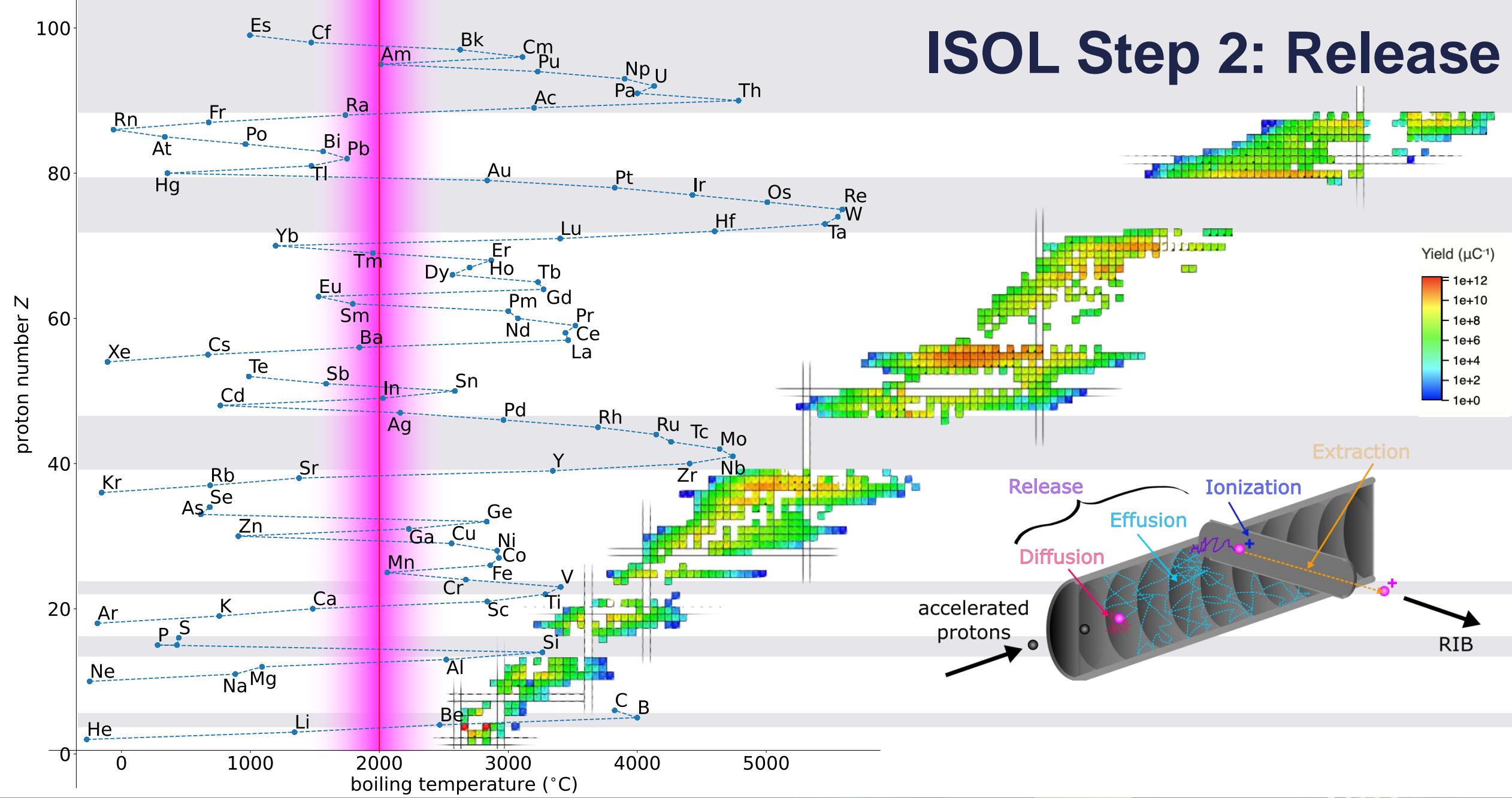
cern.ch/isolde-yields



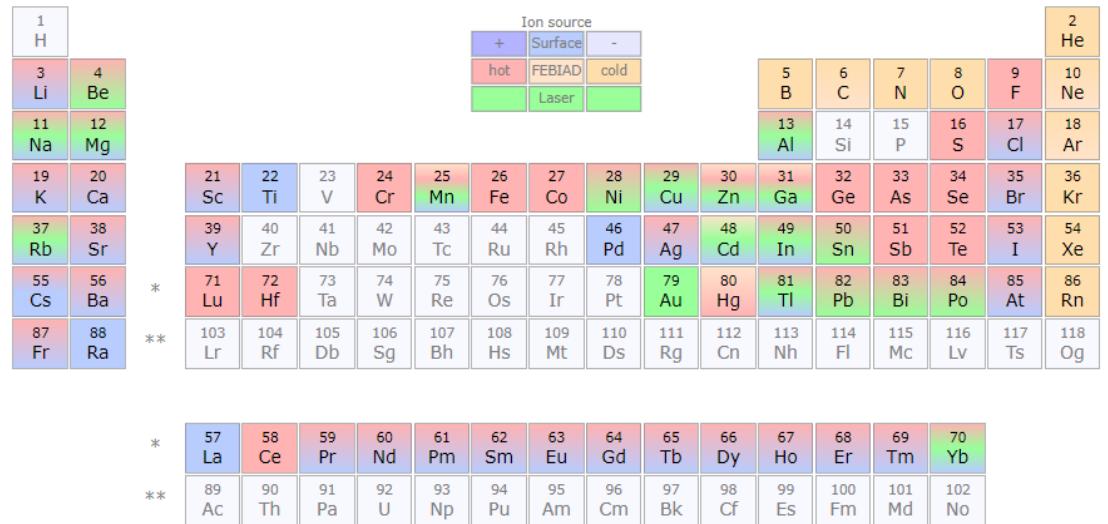
ISOL Step 2: Release



ISOL Step 2: Release



ISOL Step 3: Ionization

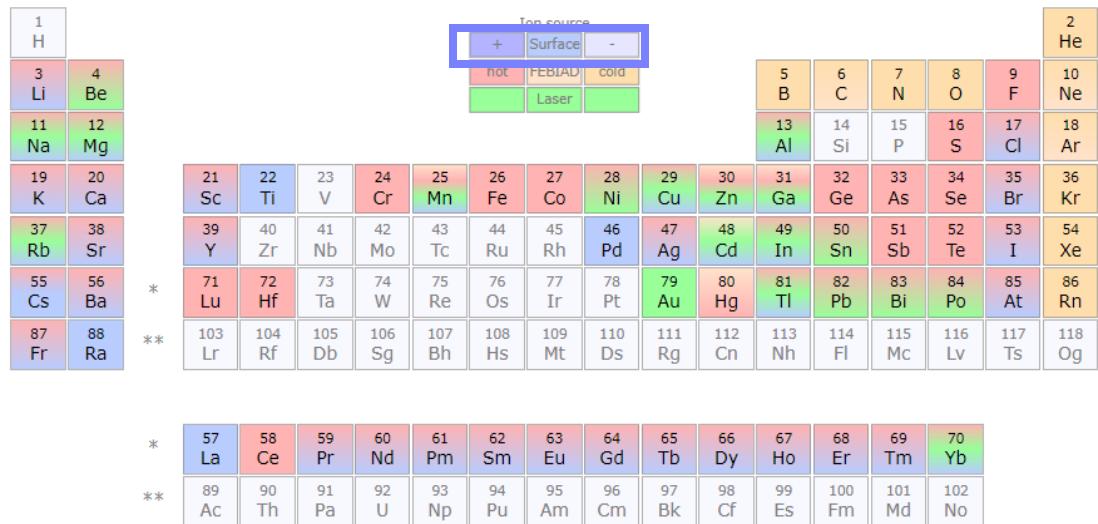


cern.ch/isolde-yields

Ion sources

- Surface ionization
- Plasma / electron impact ionization
- Resonance laser ionization

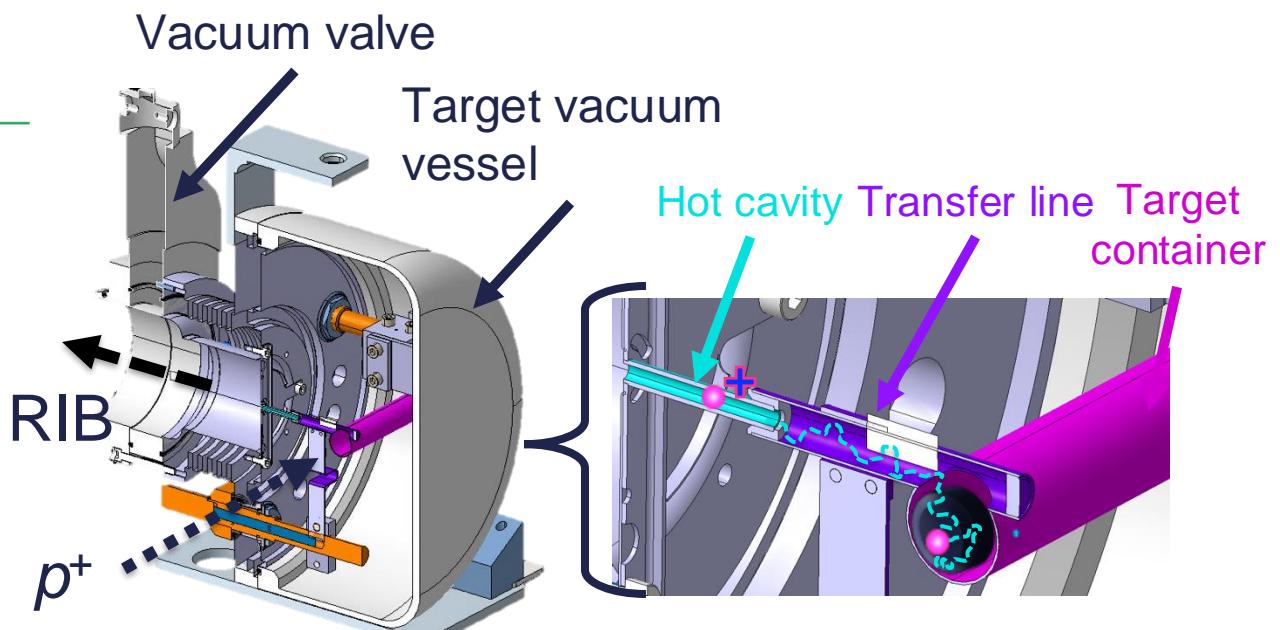
ISOL Step 3: Ionization



cern.ch/isolde-yields

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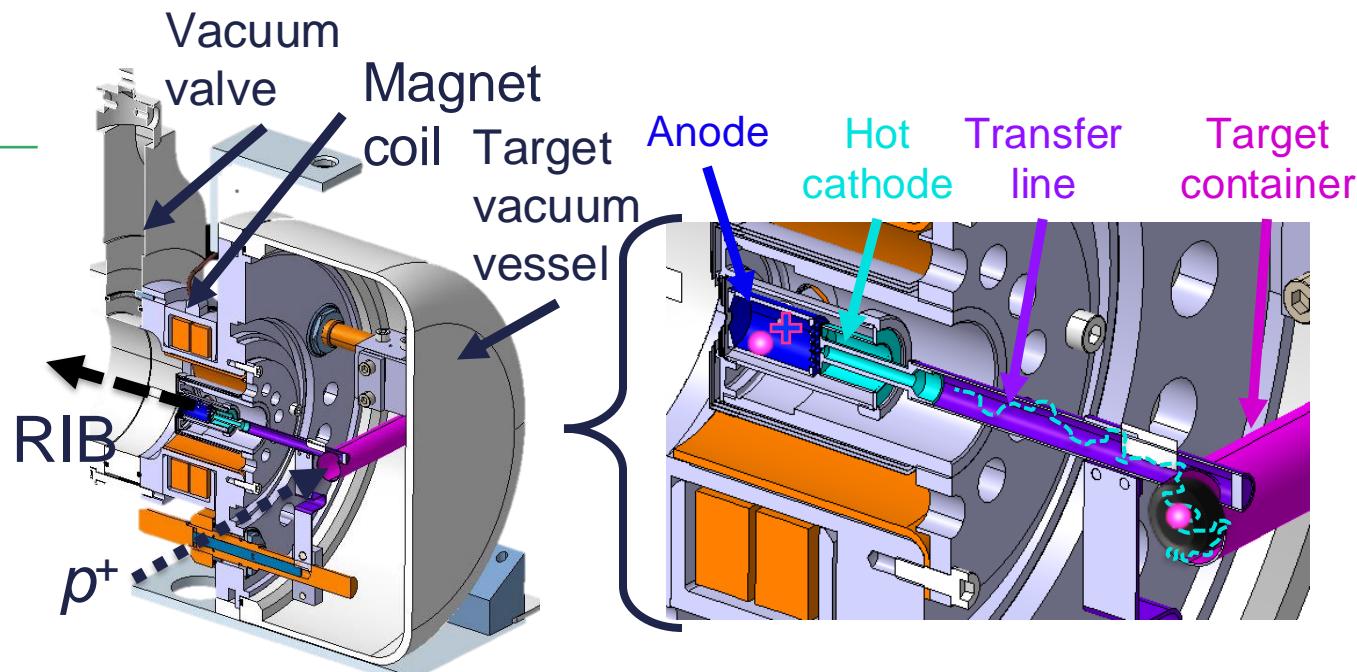


ISOL Step 3: Ionization

1	H
3	Li
4	Be
11	Na
12	Mg
19	K
20	Ca
37	Rb
38	Sr
55	Cs
56	Ba
87	Fr
88	Ra

Ion source											
+		Surface				-					
hot	FEBIAD	cold	Laser								
21	Sc	22	Ti	23	V	24	Cr	25	Mn	26	Fe
39	Y	40	Zr	41	Nb	42	Mo	43	Tc	44	Ru
71	Lu	72	Hf	73	Ta	74	W	75	Re	76	Os
*											
103	Lr	104	Rf	105	Db	106	Sg	107	Bh	108	Hs
**											
57	La	58	Ce	59	Pr	60	Nd	61	Pm	62	Sm
89	Ac	90	Th	91	Pa	92	U	93	Np	94	Pu
95	Am	96	Cm	97	Bk	98	Cf	99	Es	100	Fm
101	Md	102	No								

cern.ch/isolde-yields



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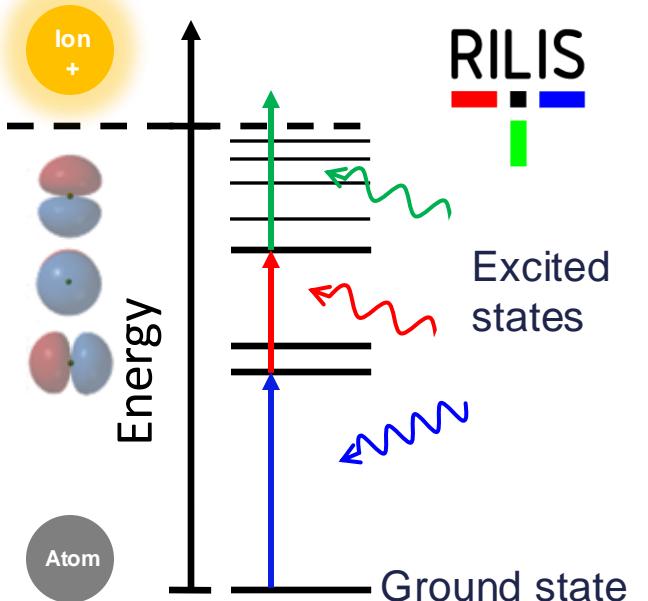
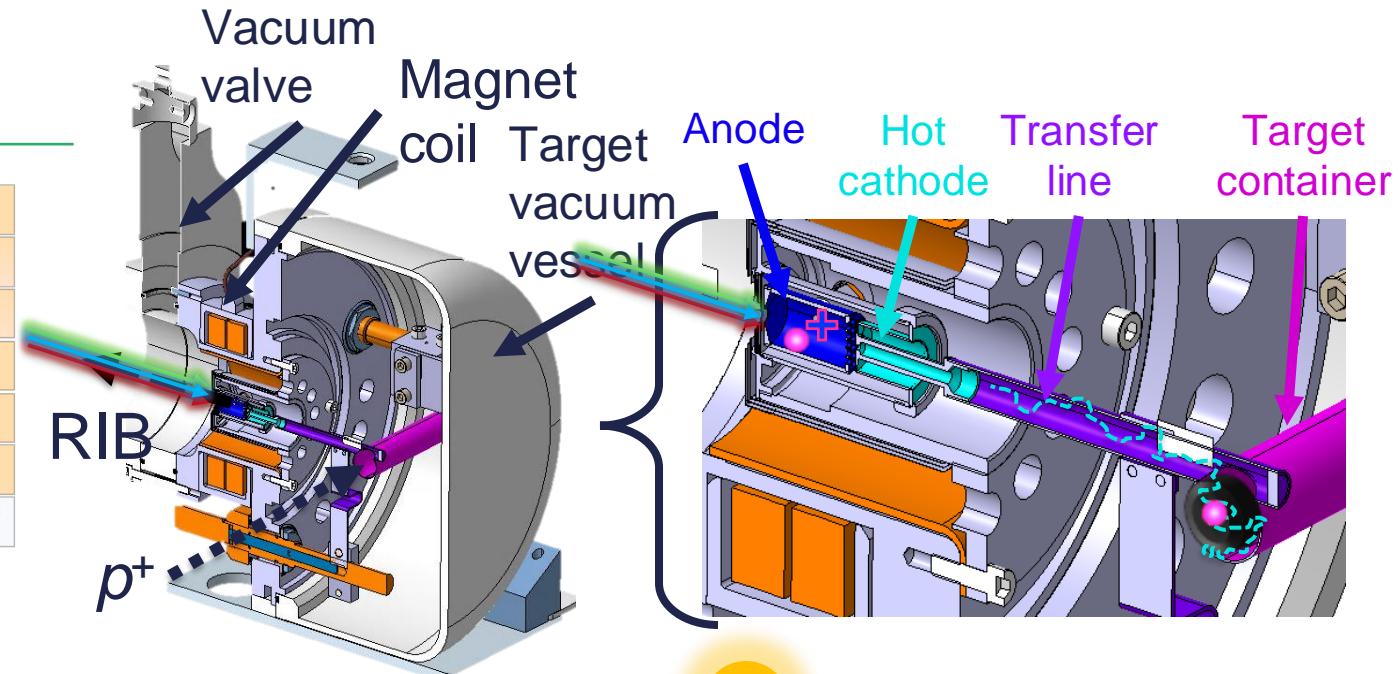
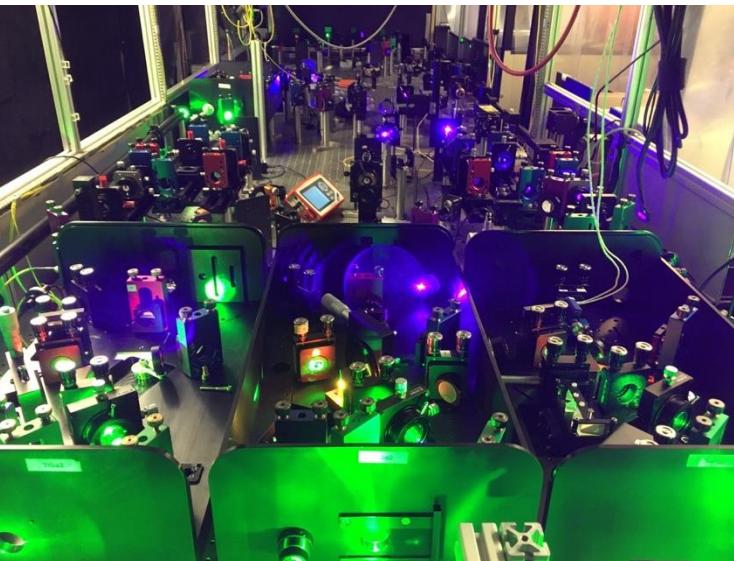
ISOL Step 3: Ionization

Ion source											
+			Surface			-					
hot	FEBIAD	cold	Laser	Laser	Laser						
1 H						2 He					
3 Li		4 Be				5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na		12 Mg				13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca					30 Cu	31 Zn	32 Ga	33 As	34 Se	35 Br
37 Rb	38 Sr					40 Ti	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh
55 Cs	56 Ba	*				46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb
87 Fr	88 Ra	**				52 Te	53 I	54 Xe	55 At	56 Rn	57 Og
103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl
57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm
101 Md	102 No										

<cern.ch/isolde-yields>

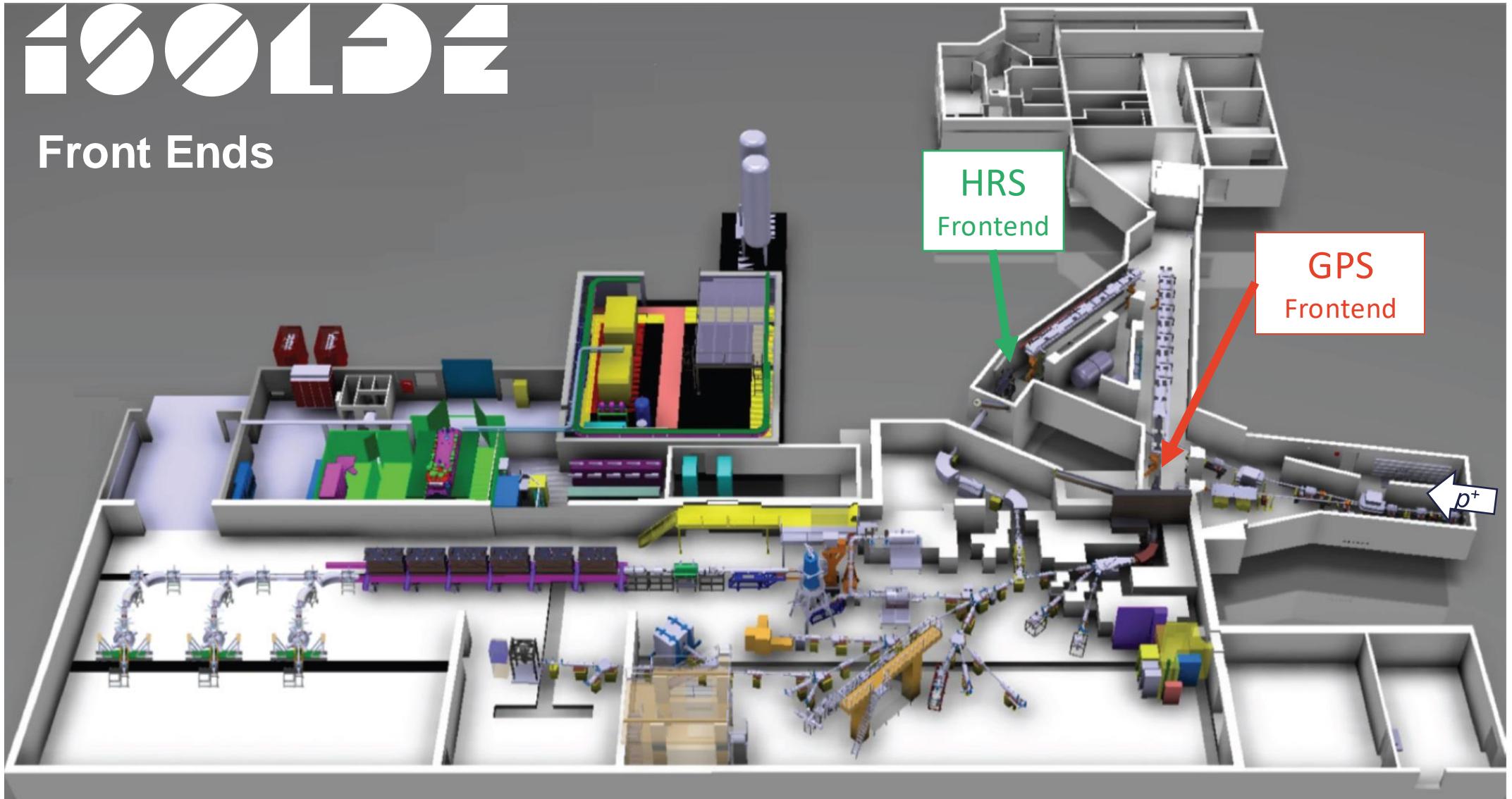
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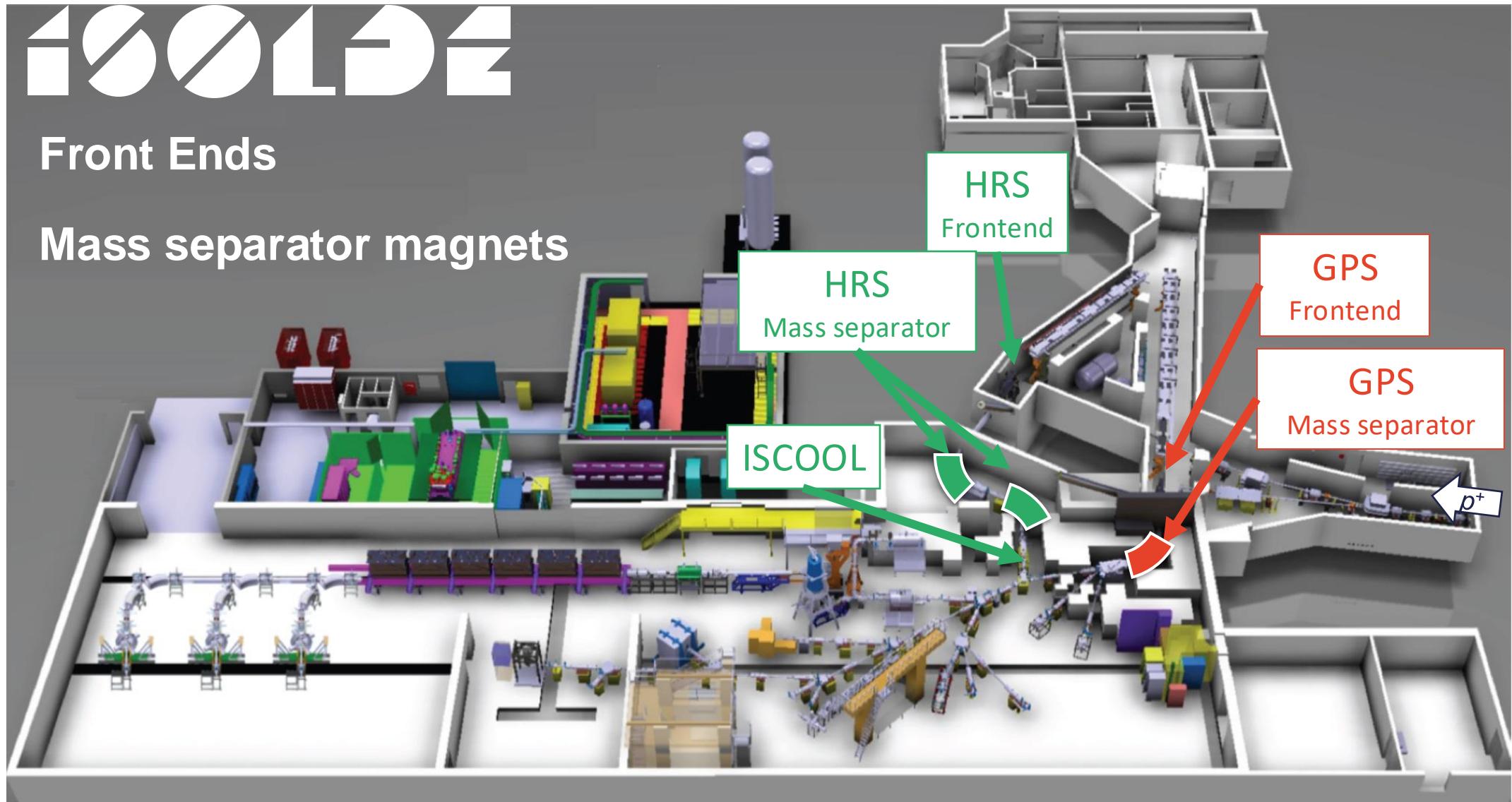
ISOL Step 4: Mass separation

Catherall et al. (2017) *J. Phys G* 44, 094002
isolde.web.cern.ch



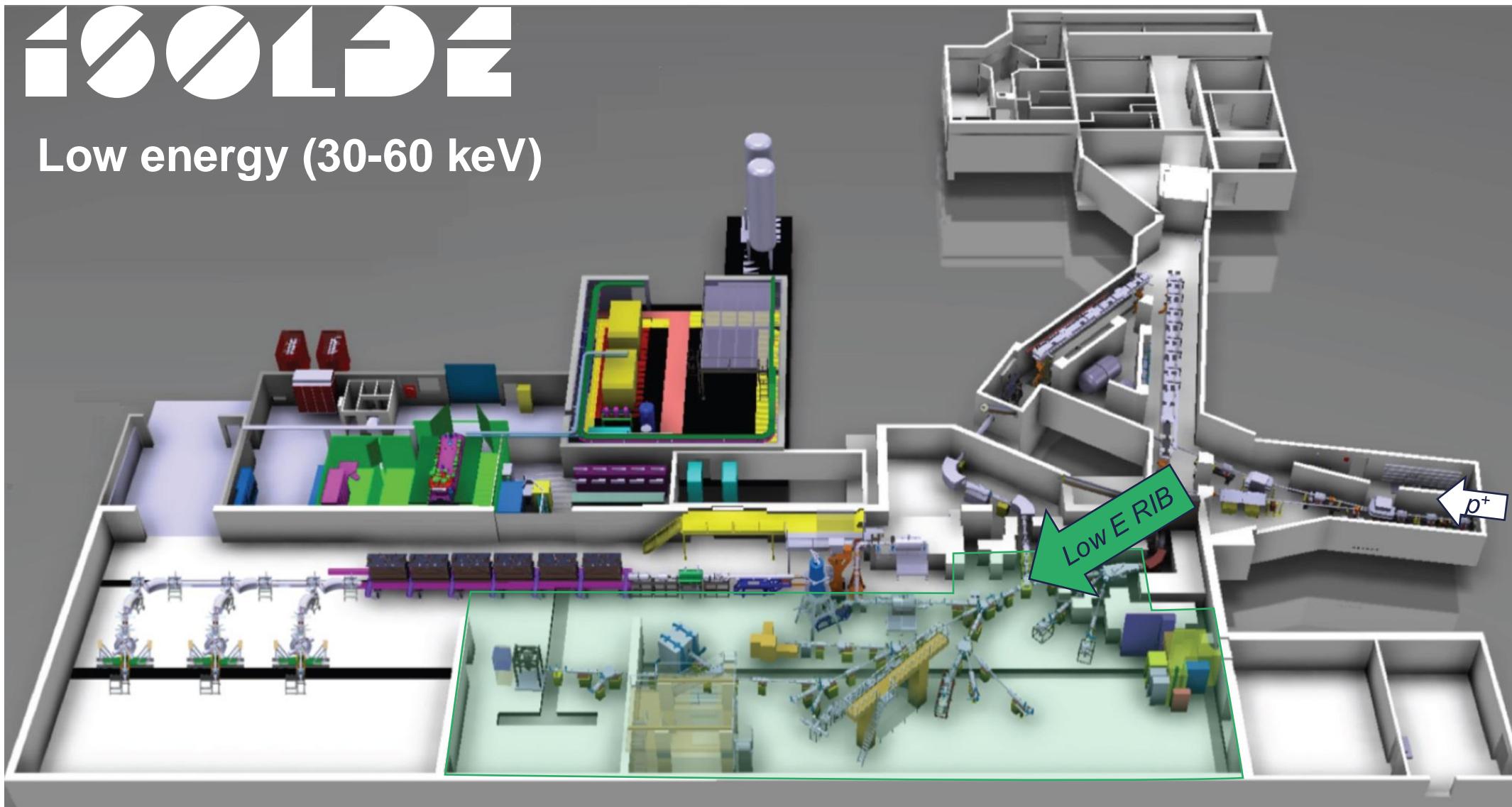
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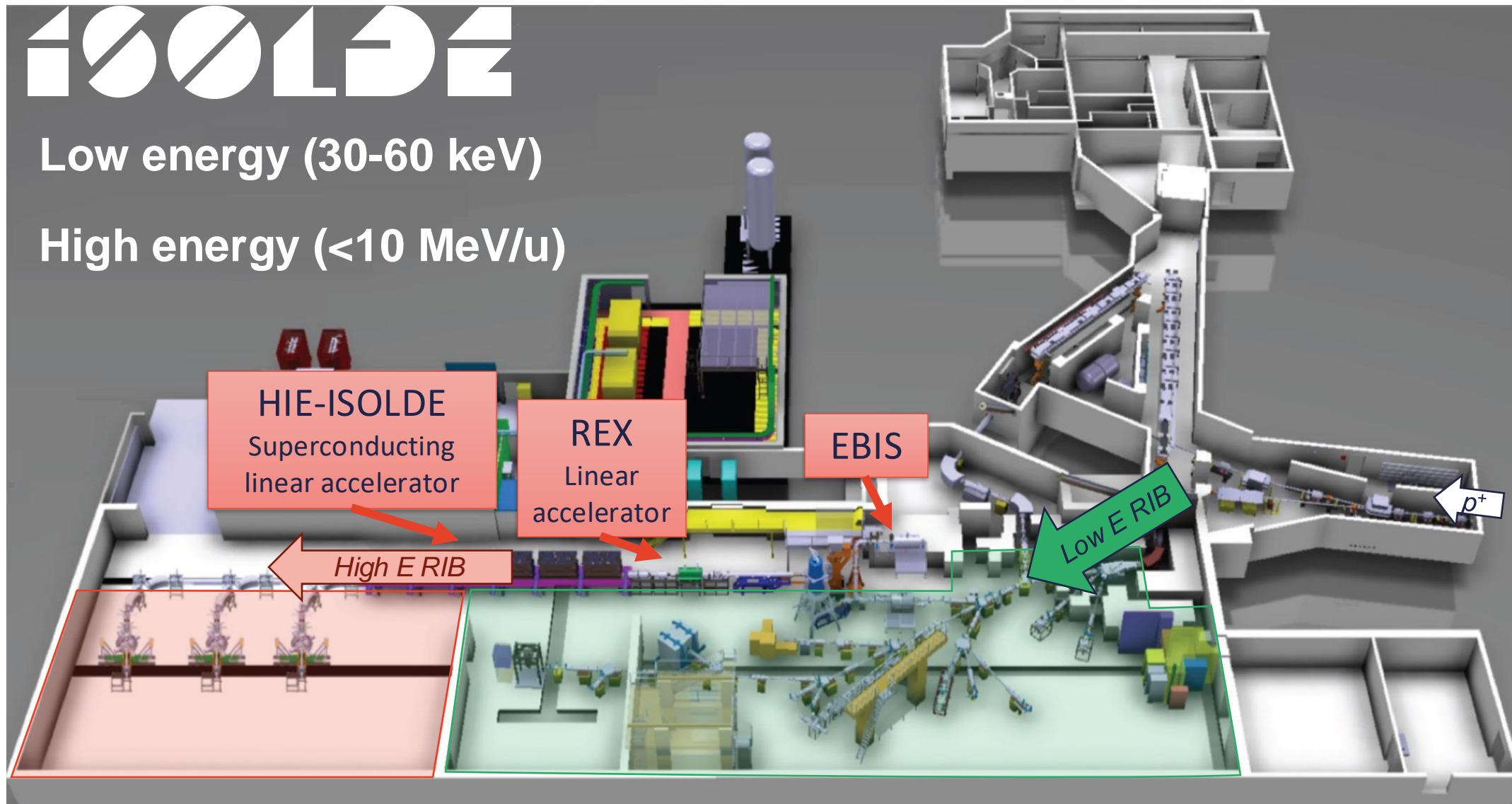
ISOL Step 5: Delivery to Experiments

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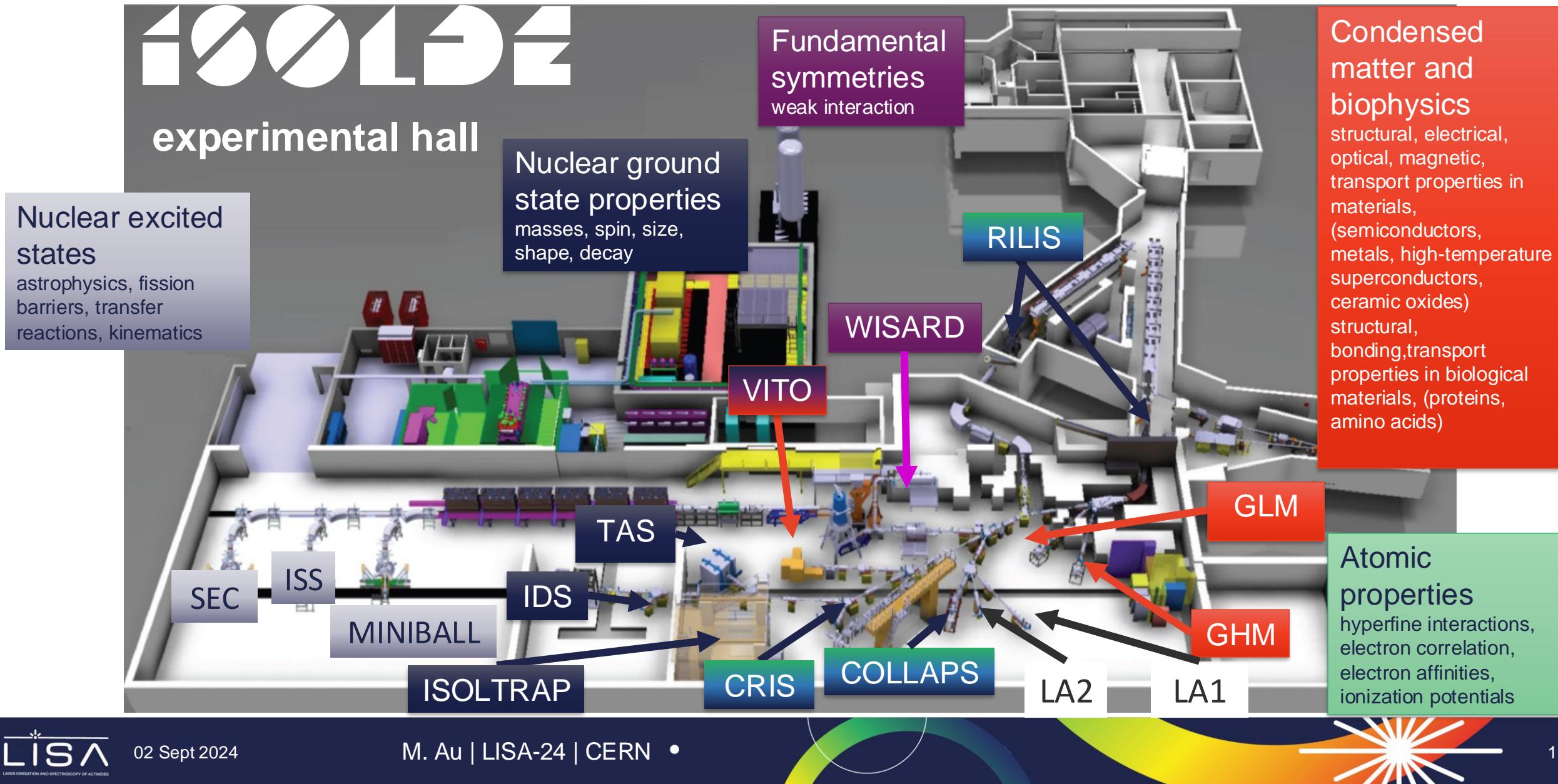
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CERN-ISOLDE

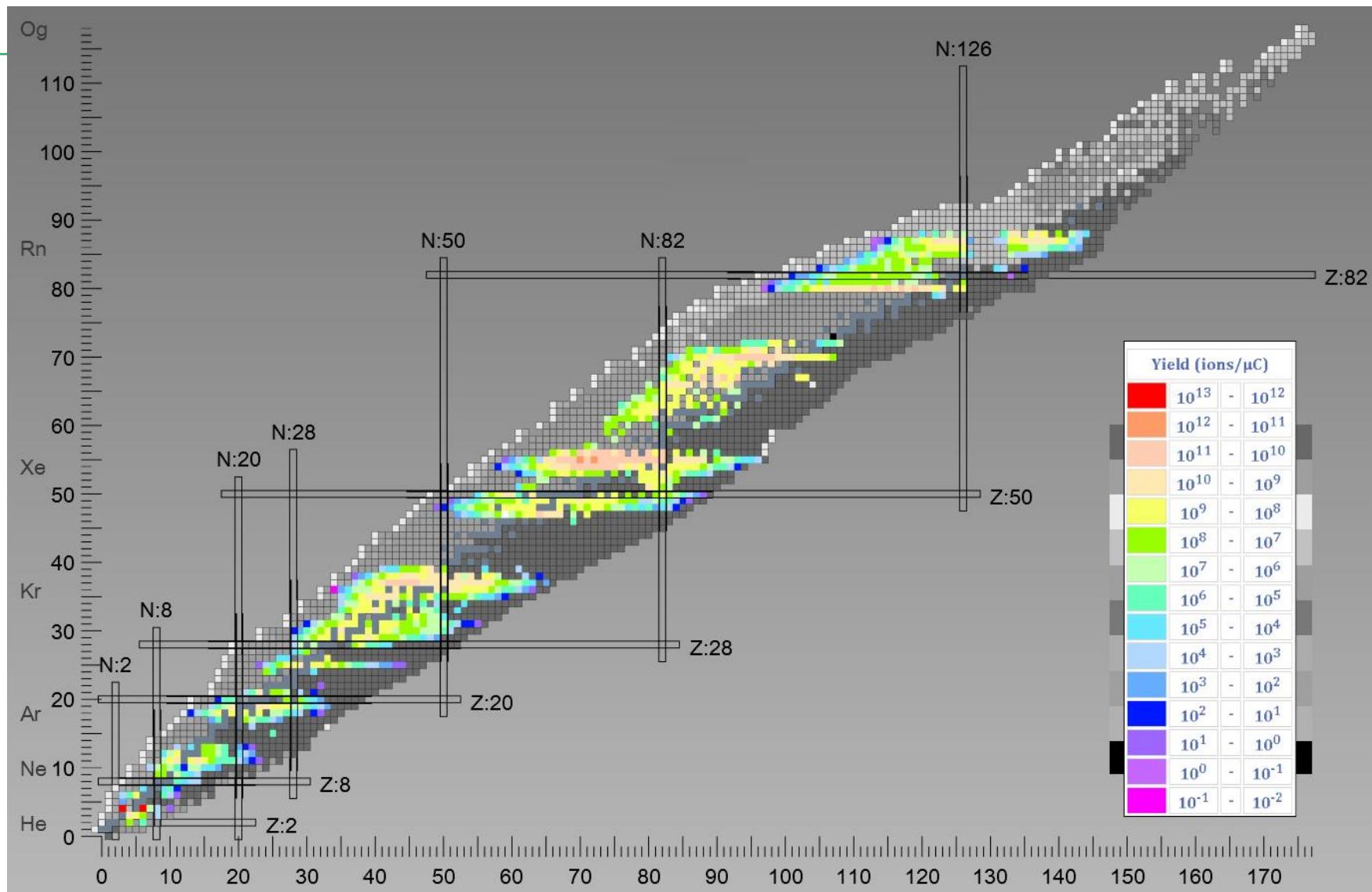
>1000 isotopes and isomers

74 elements

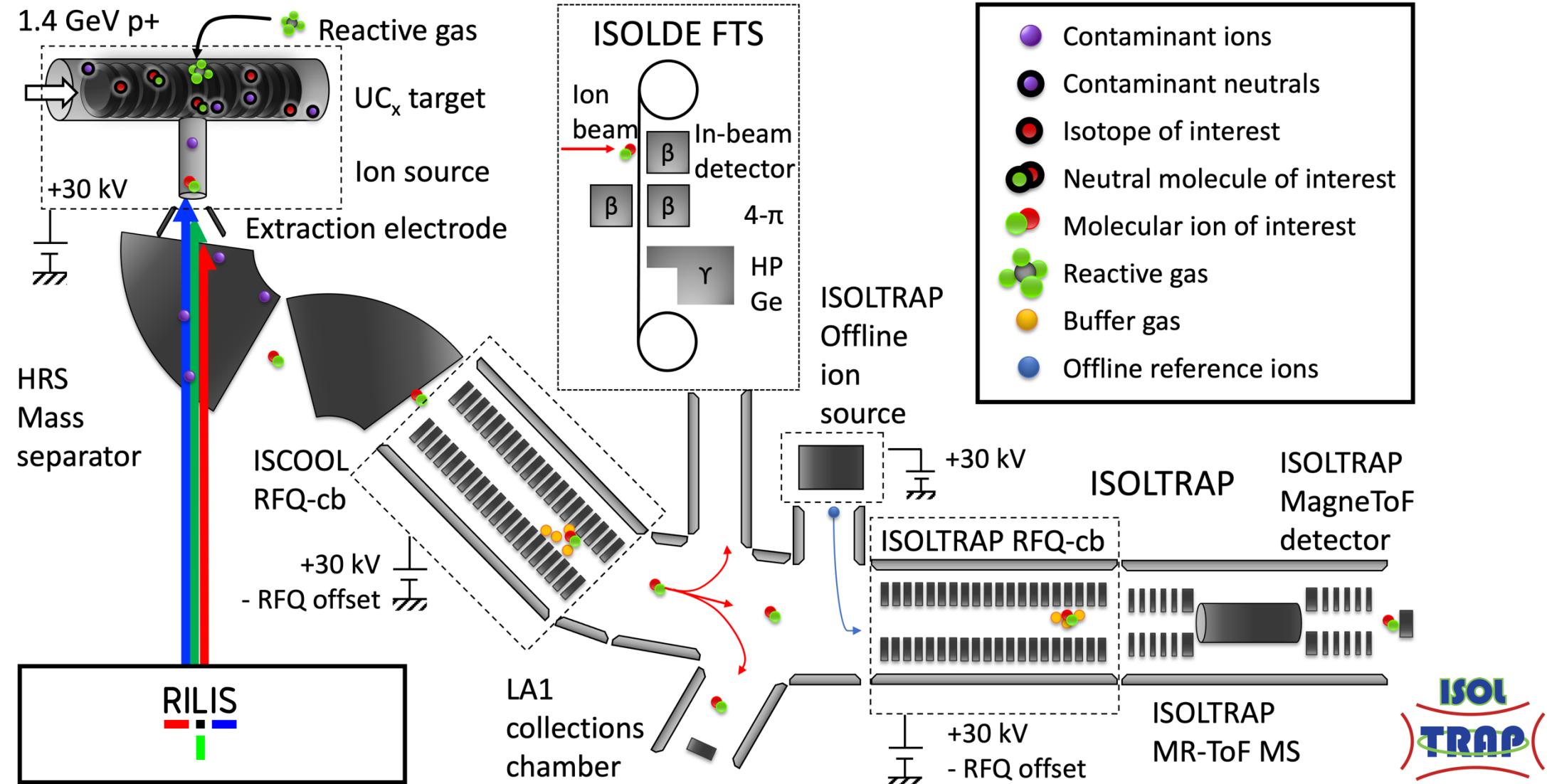
Balof et.al, (2020) NIM B 463, 211-215
cern.ch/isolde-yields

www.nucleonica.com

Dataset: JEFF-3.1 Nuclear Data Library, NEA (2023)



Beam development at ISOLDE



Atomic actinides

Up to 2020

Actinium

- Delivered as Ra/Fr: ^{225}Ac [1]
- Laser-ionized: IS637 $^{222-231}\text{Ac}$ [2]



Actinium

- Laser-ionized: $^{225,227,228}\text{Ac}$ [3]

Thorium

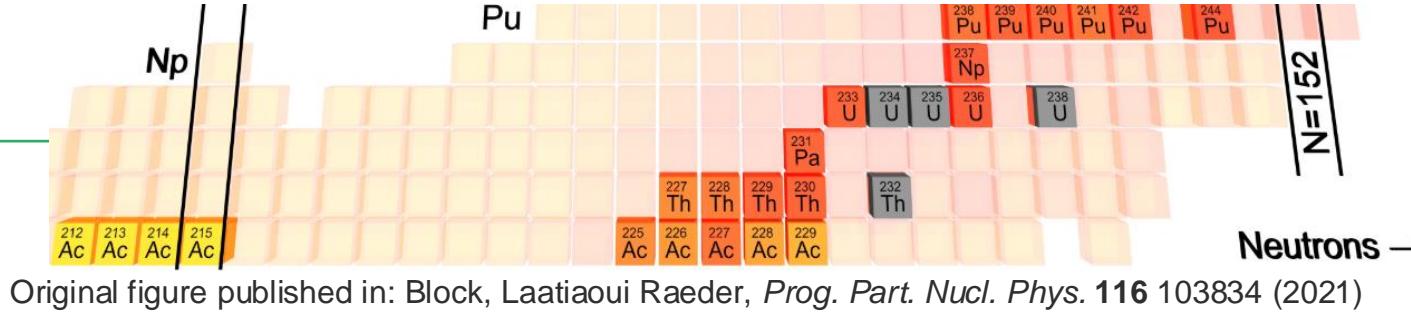
- From Ac/Ra/Fr: ^{229}Th [6]

Protactinium

- Laser-ionized: tried, failed

Uranium

- $^{234,235,238}\text{U}$ [7]



Neptunium

- Laser-ionized: $^{235-241}\text{Np}$

Plutonium

- Laser-ionized: $^{234-241}\text{Pu}$

[1] Guglielmetti et al, EPJA 12, 383-386 (2002)

[2] Jajčišinová, E., et al. Sci Rep 14, 11033 (2024).

[3] Andreyev et al, INTC-LOI-216 (2020)

[4] Heinke et al, NIM B 541, 8-12 (2023)

[5] Heinke, Jaradat, Zenodo, 7824897 (2022)

[6] Kraemer et al, Nature 617, 706-710 (2022)

[7] Au et al, NIM B 541, 375-379 (2023)

[8] Kaja, *PhD thesis* (2024)

[9] Au et al, INTC-LOI-243 (2022)

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Actinium

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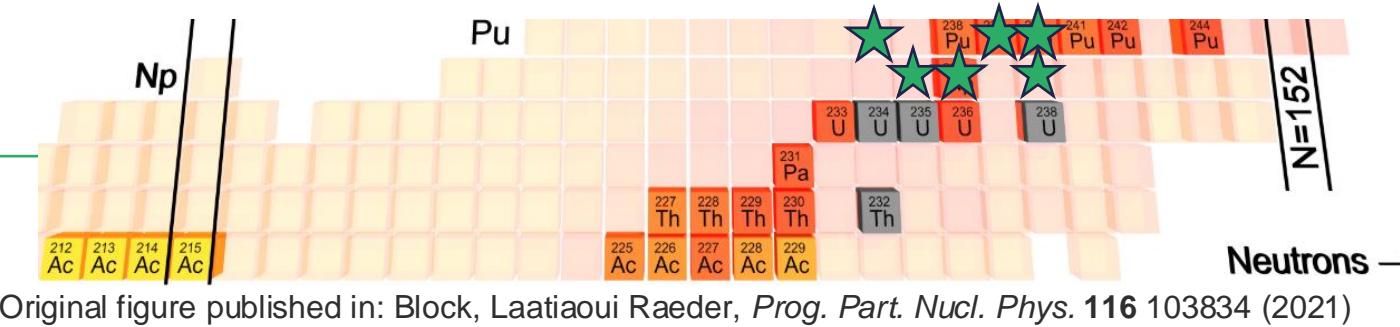
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Original figure published in: Block, Laatiaoui Raeder, *Prog. Part. Nucl. Phys.* **116** 103834 (2021)

Neptunium

- Laser-ionized: $^{235-241}\text{Np}$

Plutonium

- Laser-ionized: $^{234-241}\text{Pu}$

PHYSICAL REVIEW C **107**, 064604 (2023)

Production of neptunium and plutonium nuclides from uranium carbide using 1.4-GeV protons

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(Received 11 March 2023; accepted 8 May 2023; published 8 June 2023; corrected 7 August 2023)

[1] Guglielmetti et al, EPJA 12, 383-386 (2002)

[2] Jajčišinová, E., et al. Sci Rep 14, 11033 (2024).

[3] Andreyev et al, INTC-LOI-216 (2020)

[4] Heinke et al, NIM B 541, 8-12 (2023)

[5] Heinke, Jaradat, Zenodo, 7824897 (2022)

[6] Kraemer et al, Nature 617, 706-710 (2022)

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[8] Kaja, *PhD thesis* (2024)

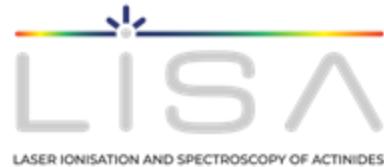
[9] Au et al, INTC-LOI-243 (2022)

Atomic actinides

Up to 2020

Actinium

- Delivered as Ra/Fr: ^{225}Ac [1]
- Laser-ionized: IS637 $^{222-231}\text{Ac}$ [2]



Actinium

- Laser-ionized: $^{225,227,228}\text{Ac}$ [3]
- In-source spectroscopy: $^{224-231}\text{Ac}$ [4,5]

Thorium

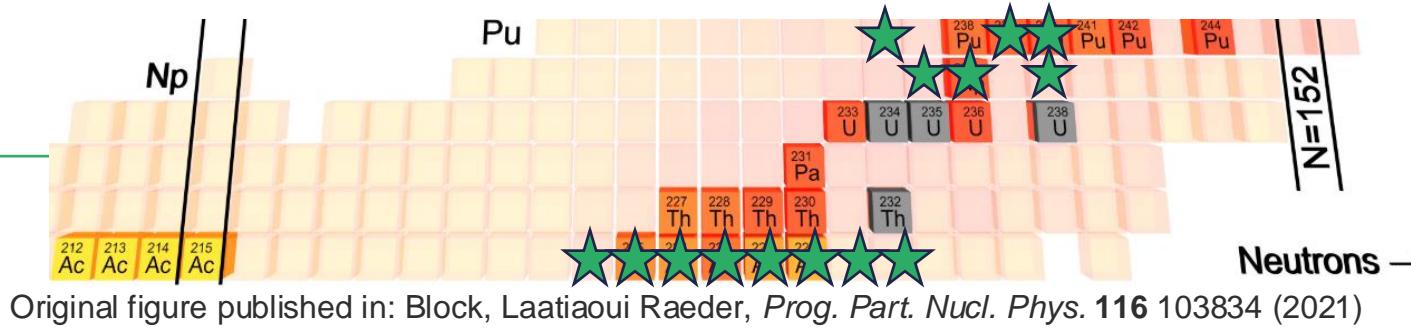
- From Ac/Ra/Fr: ^{229}Th [6]

Protactinium

- Laser-ionized: tried, failed

Uranium

- $^{234,235,238}\text{U}$ [7]



Original figure published in: Block, Laatiaoui Raeder, *Prog. Part. Nucl. Phys.* **116** 103834 (2021)

Neptunium

- Laser-ionized: $^{235-241}\text{Np}$

Plutonium

- Laser-ionized: $^{234-241}\text{Pu}$



First on-line application of the high-resolution spectroscopy laser ion source PI-LIST at ISOLDE

Reinhard Heinke ^{a,*}, Mia Au ^{a,b}, Cyril Bernerd ^{a,c}, Katerina Chrysalidis ^b, Thomas E. Cocolios ^c, Valentin N. Fedossev ^a, Isabel Hendriks ^{a,d}, Asar A.H. Jaradat ^a, Magdalena Kaja ^e, Tom Kieck ^{f,g}, Tobias Kron ^e, Ralitsa Mancheva ^{b,h}, Bruce A. Marsh ^a, Stefano Marzari ^a, Sebastian Raeder ^{f,g}, Sebastian Rothe ^a, Dominik Studer ^{f,g}, Felix Weber ^e, Klaus Wendt ^e

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^b Chemistry department, Johannes Gutenberg University Mainz, Germany

^c Institute for Nuclear and Radiation Physics, KU Leuven, Belgium

^d Lund University, Sweden

^e Institute of Physics, Johannes Gutenberg University Mainz, Germany

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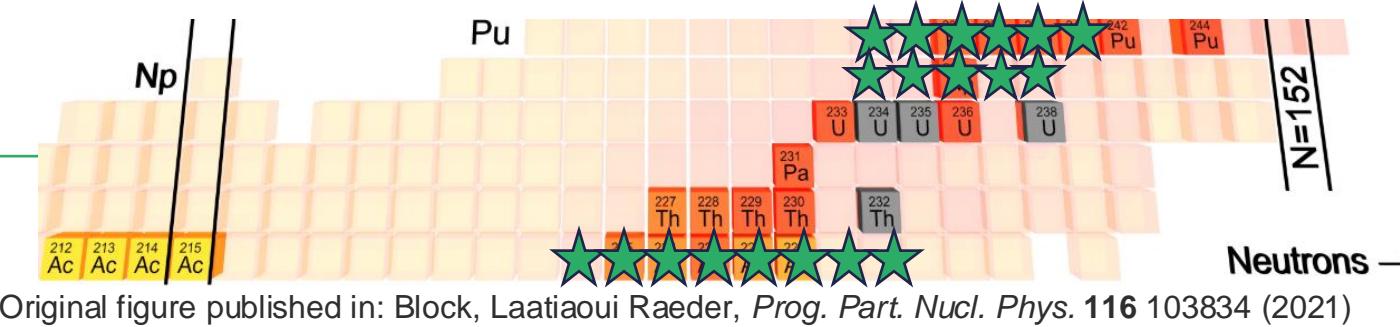
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Neptunium

- Laser-ionized: $^{235-241}\text{Np}$
- In-source spectroscopy, analysis [8,9]

Magdalena Anna Kaja

14:45 - 15:15

Plutonium

- Laser-ionized: $^{234-241}\text{Pu}$
- In-source spectroscopy, analysis [9]



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



In-source laser resonance ionization spectroscopy of neptunium and plutonium

May 13, 2022

Mia Au^{1,2}, Anastasia Borschevsky³, Katerina Chrysalidis¹, Raphaël Crosa-Rossa³, Christoph Düllmann^{2,4,5}, Reinhard Heinke¹, Asar Jaradat¹, Magdalena Kaja², Bruce Marsh¹, Iain Moore⁶, Andrea Raggio⁶, Sebastian Rothe¹, Simon Stegemann¹, Darcy van Eerten⁷, Clemens Walther⁷

¹SY-STI, CERN, Switzerland

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³Rijksuniversiteit Groningen, Groningen, Netherlands

⁴Institute for Nuclear and Radiation Physics

⁵Lund University, Sweden

⁶Institute of Physics, Johannes Gutenberg

⁷GSI Helmholtzzentrum für Schwerionenforschung, Germany

⁸Helmholtz Institute Mainz, Germany

⁹University of Jyväskylä, Finland

¹⁰IRS, Leibniz Universität Hannover, Germany

Spokesperson: Mia Au mia.au@cern.ch, Magdalena Kaja mkaja@uni-mainz.de
Contact person: Mia Au mia.au@cern.ch

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[1] Guglielmetti et al, EPJA 12, 383-386 (2002)

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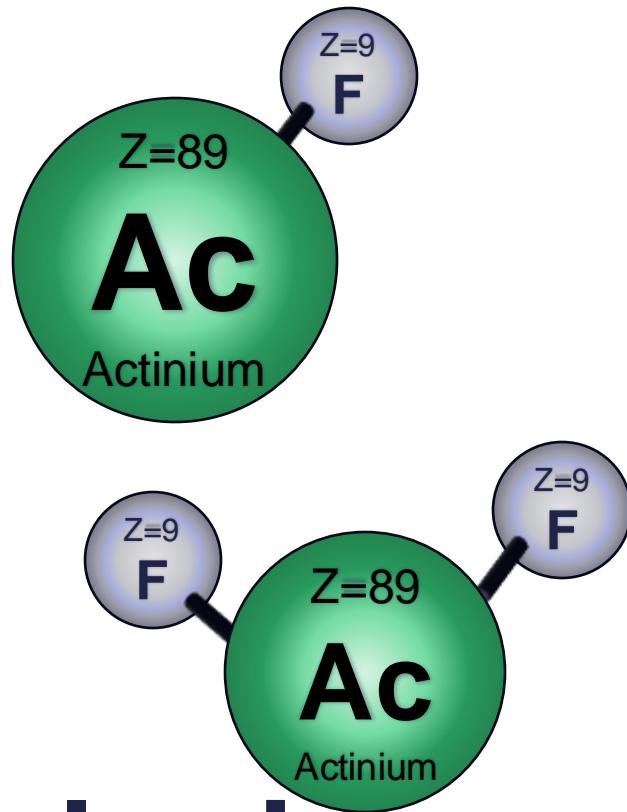
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Image published in EP
Newsletter, CERN (2020)



Molecular beams



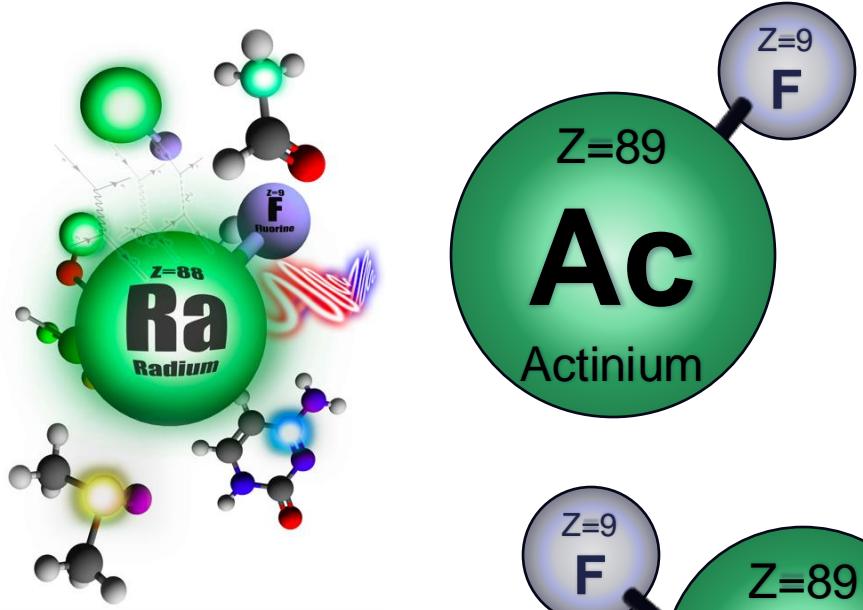
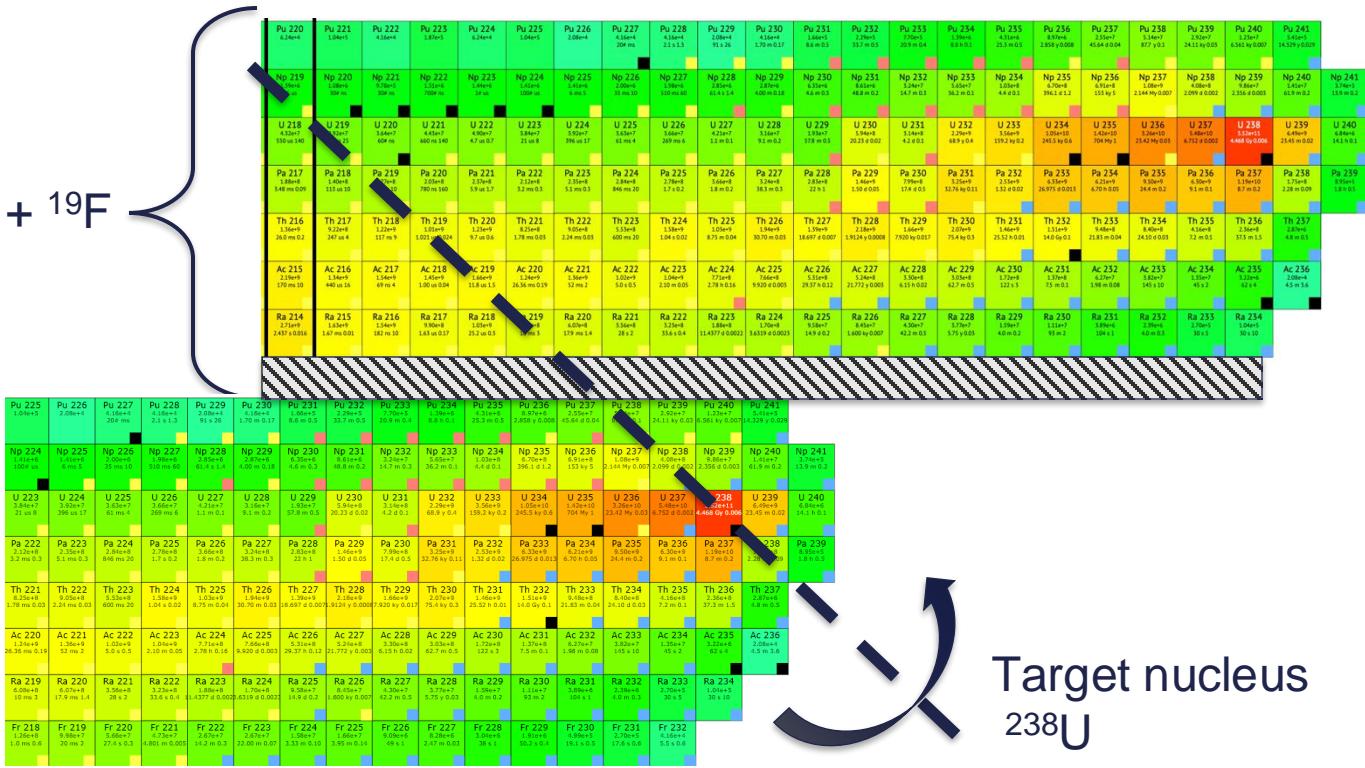


Image published in EP
Newsletter, CERN (2020)

Molecular beams

Sideband extraction



Target nucleus
238U

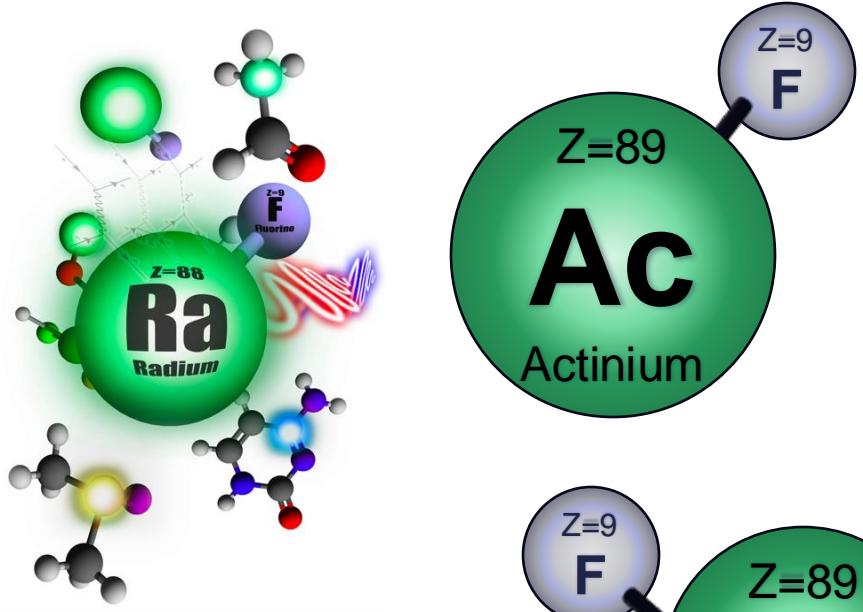
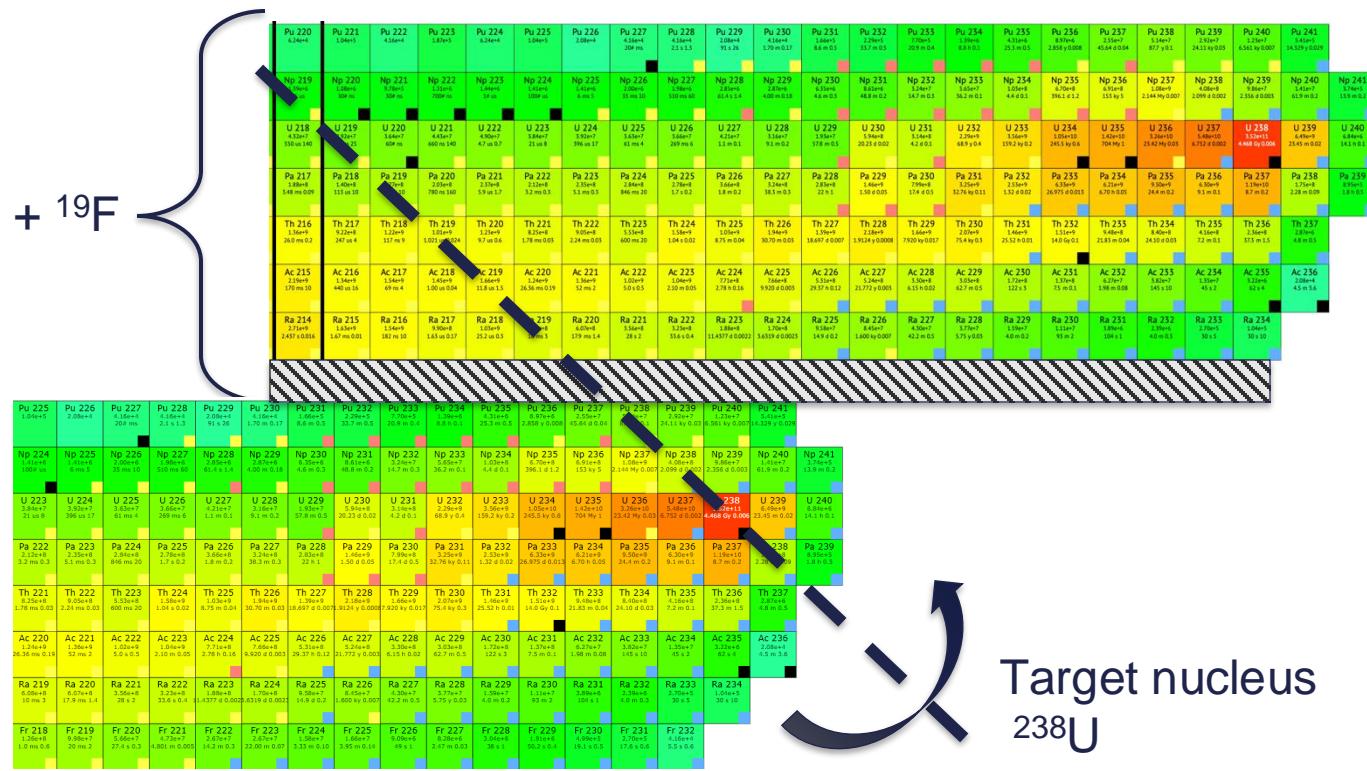


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Molecular beams

Sideband extraction

Volatilization



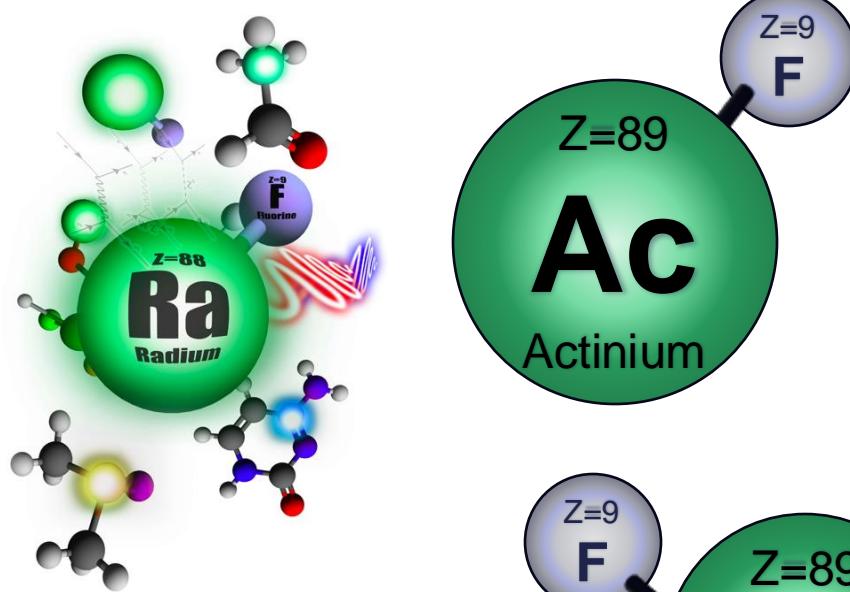


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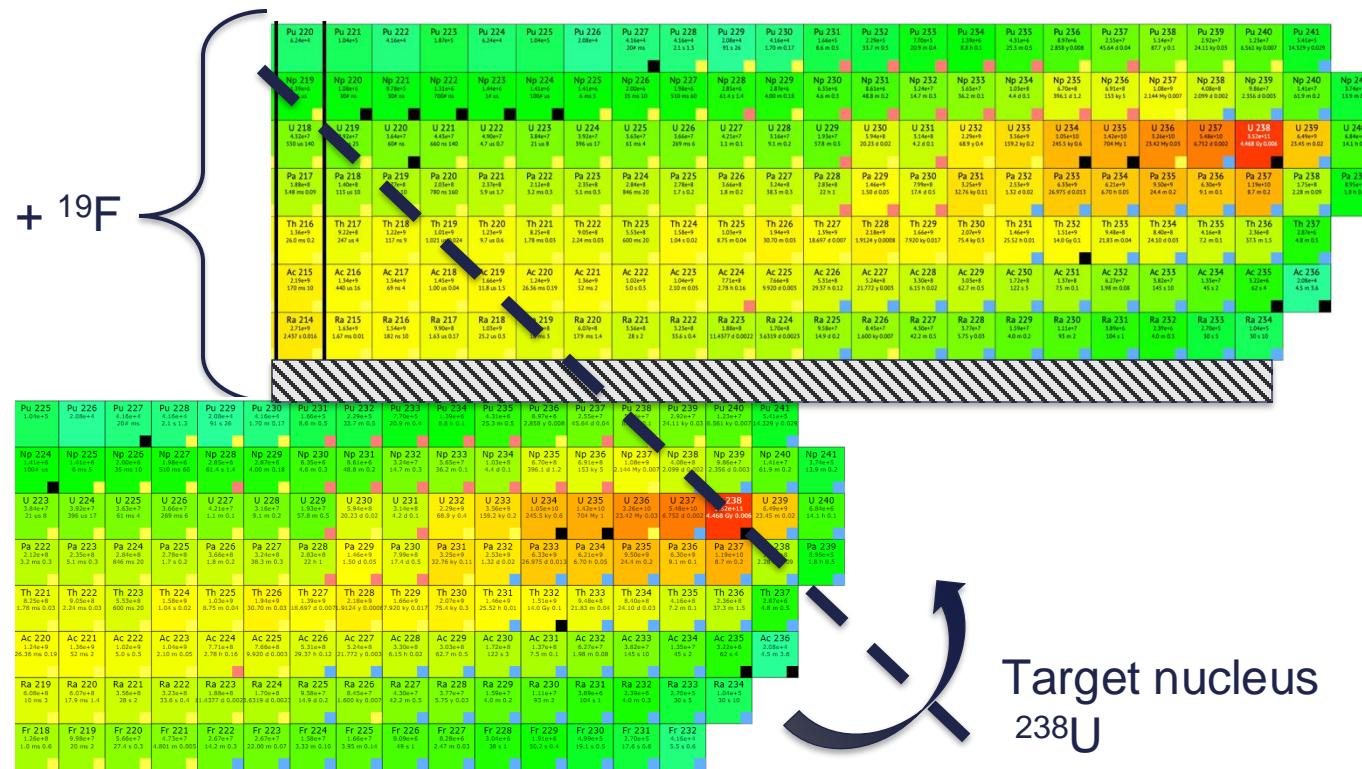
Molecular beams

Sideband extraction

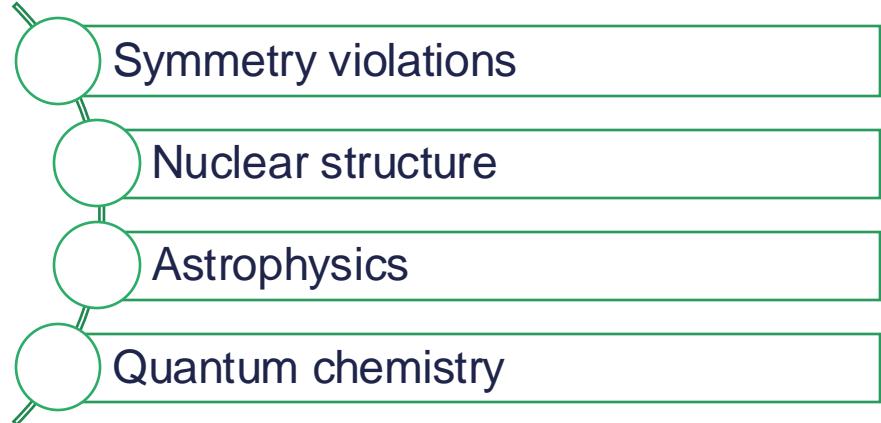
Volatilization

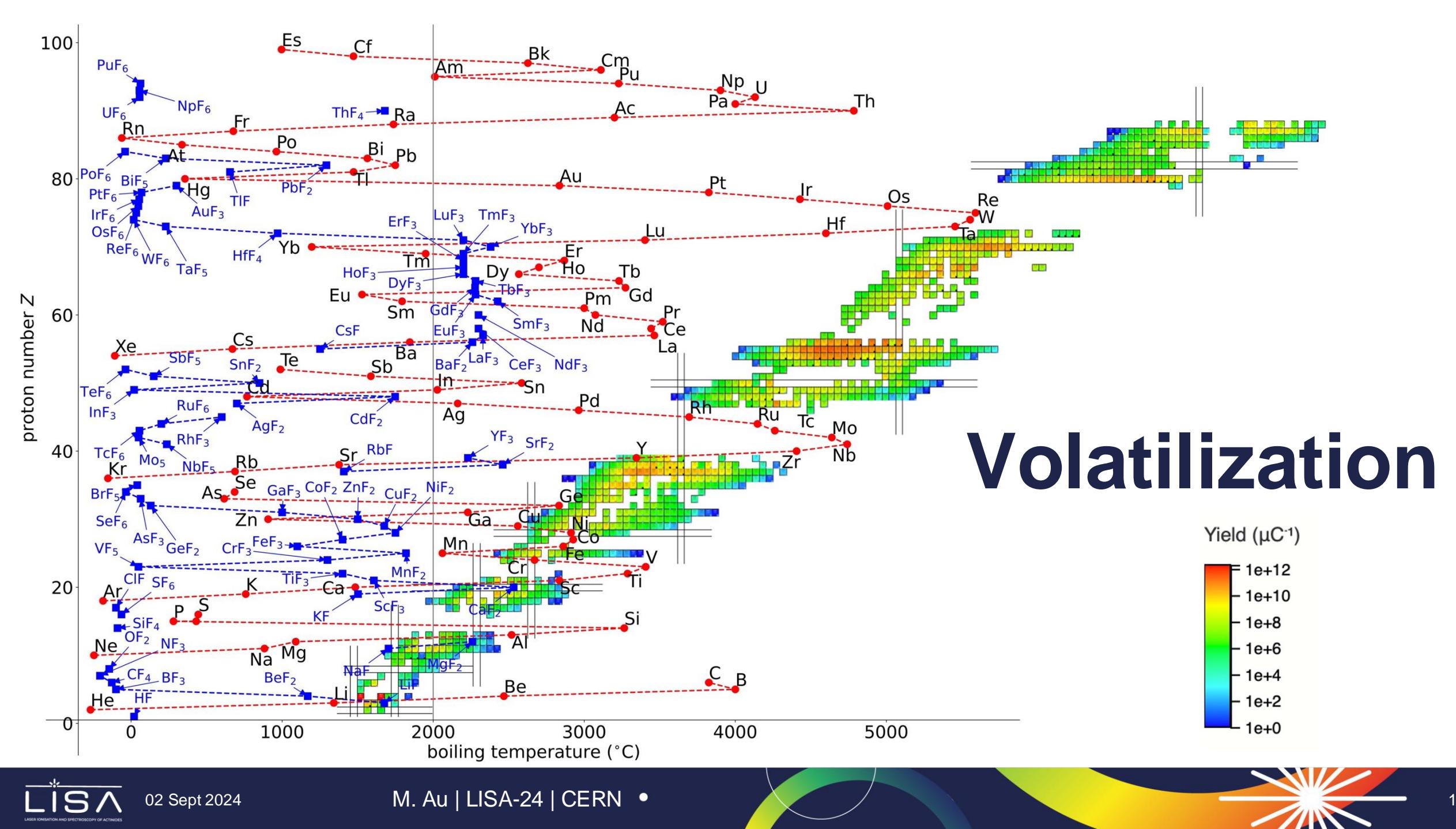
Research opportunities

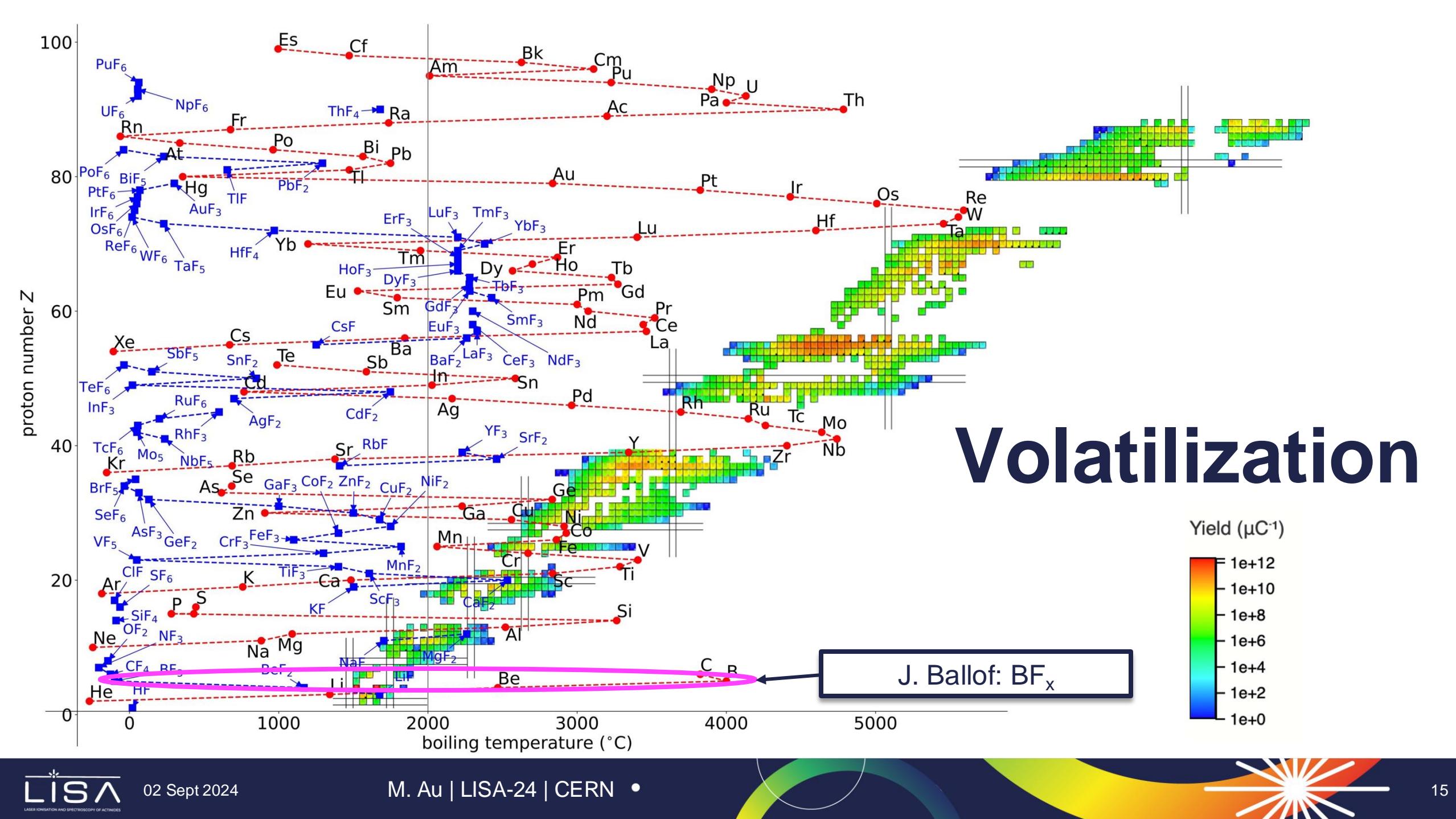
Opportunities for Fundamental Physics Research
with Radioactive Molecules, PMID: 38215499
DOI: 10.1088/1361-6633/ad1e39 (2024)

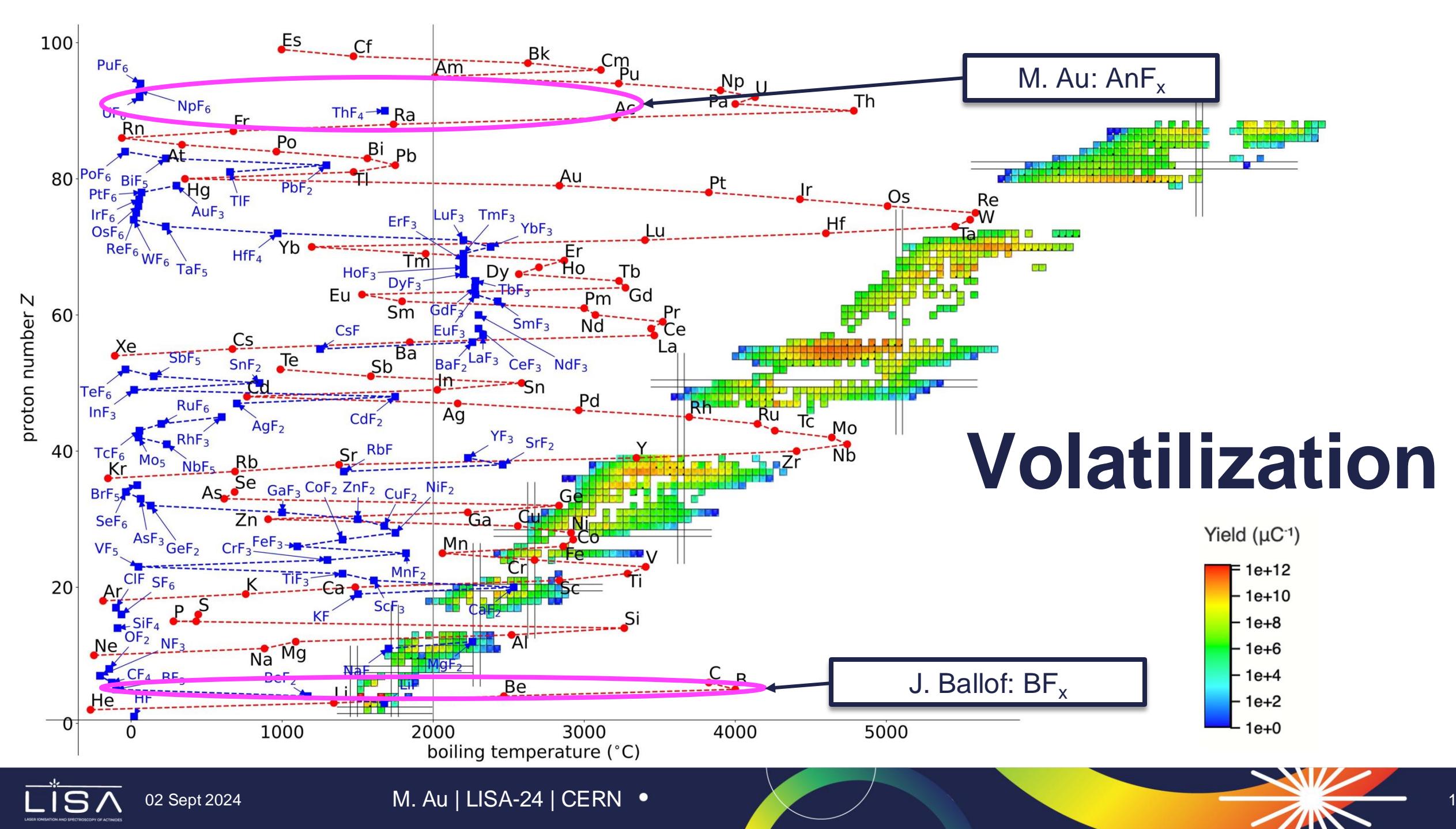


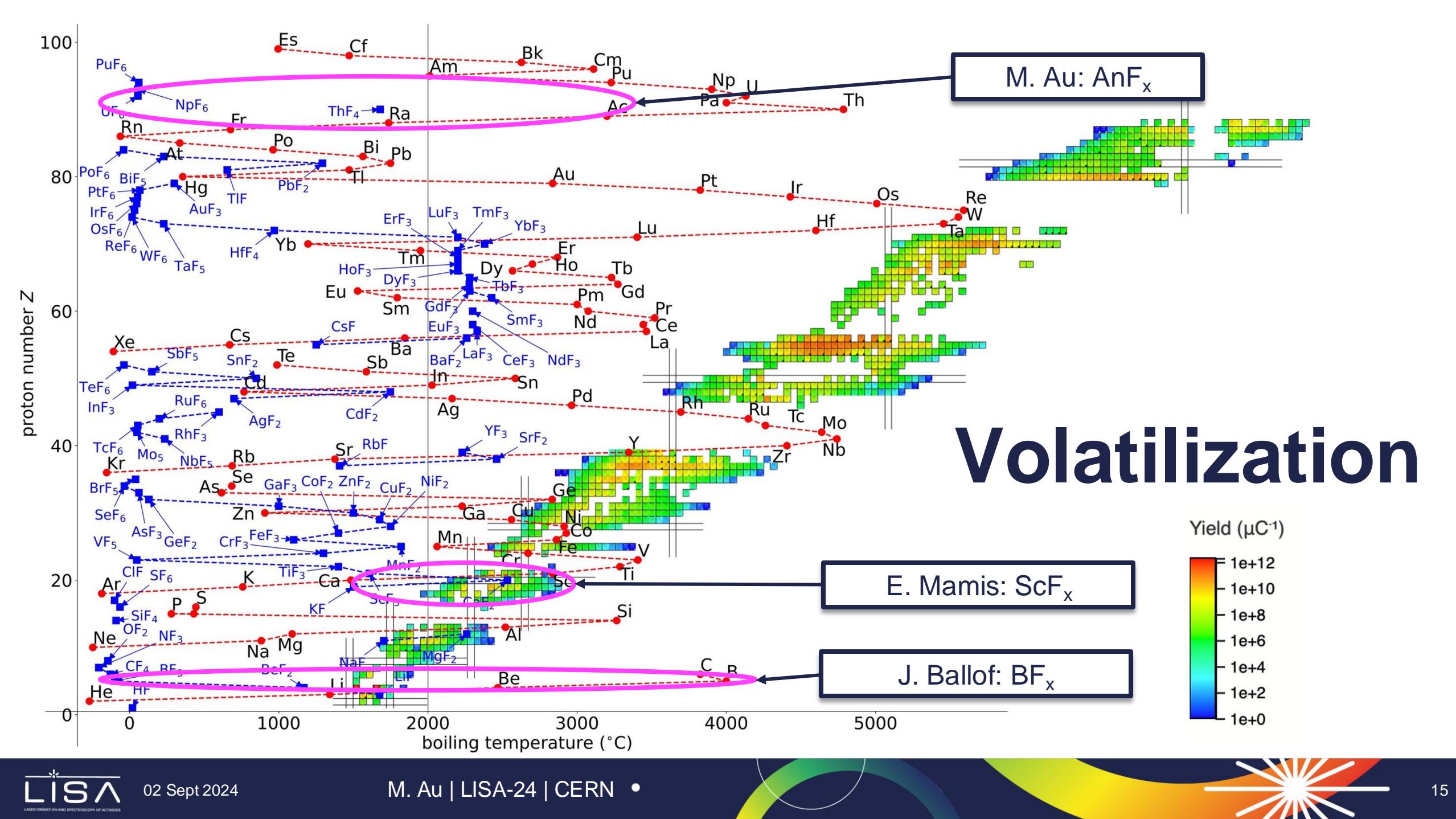
Target nucleus
 ^{238}U

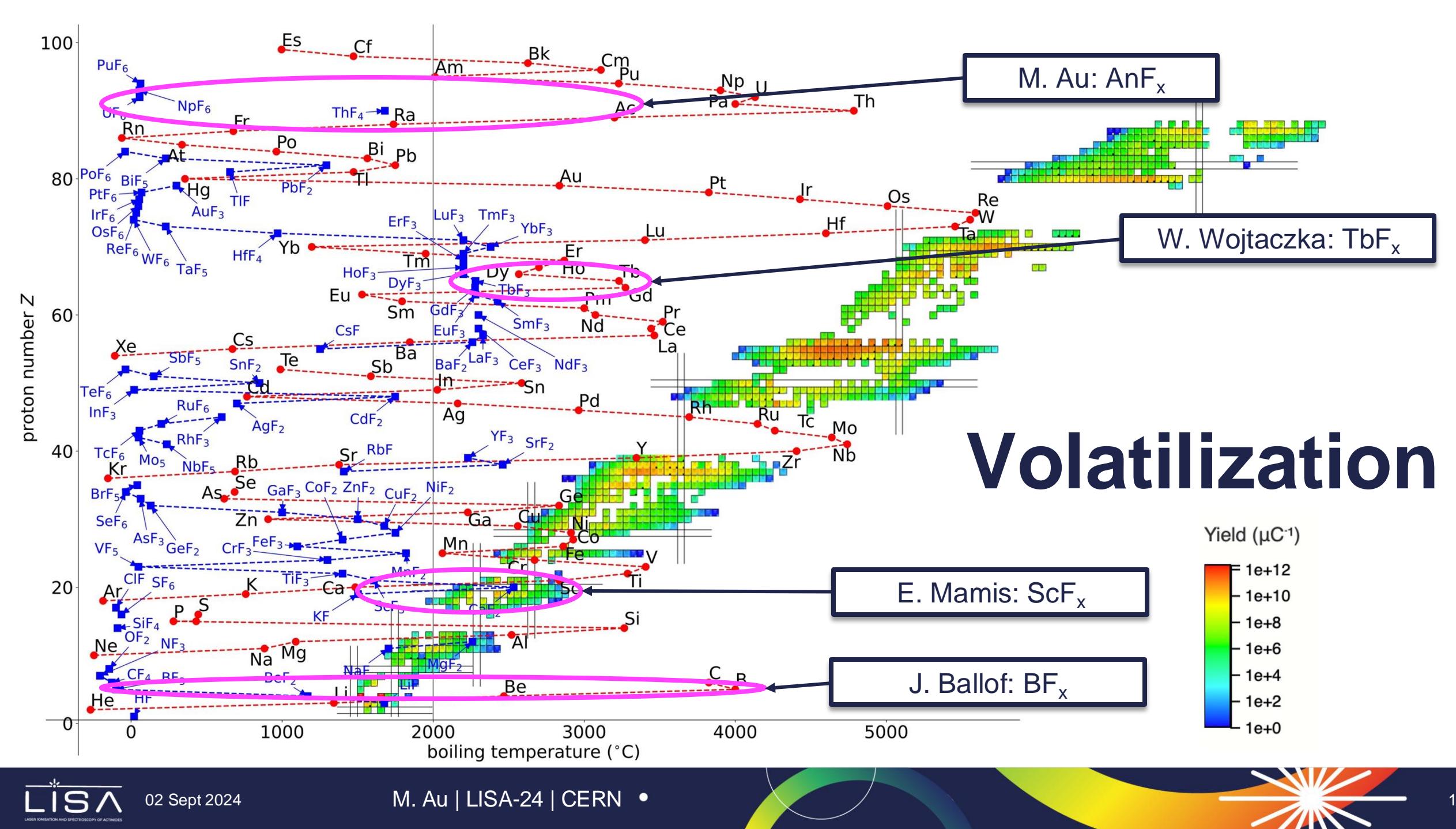












Research opportunities: a small detour

Matter-antimatter asymmetry

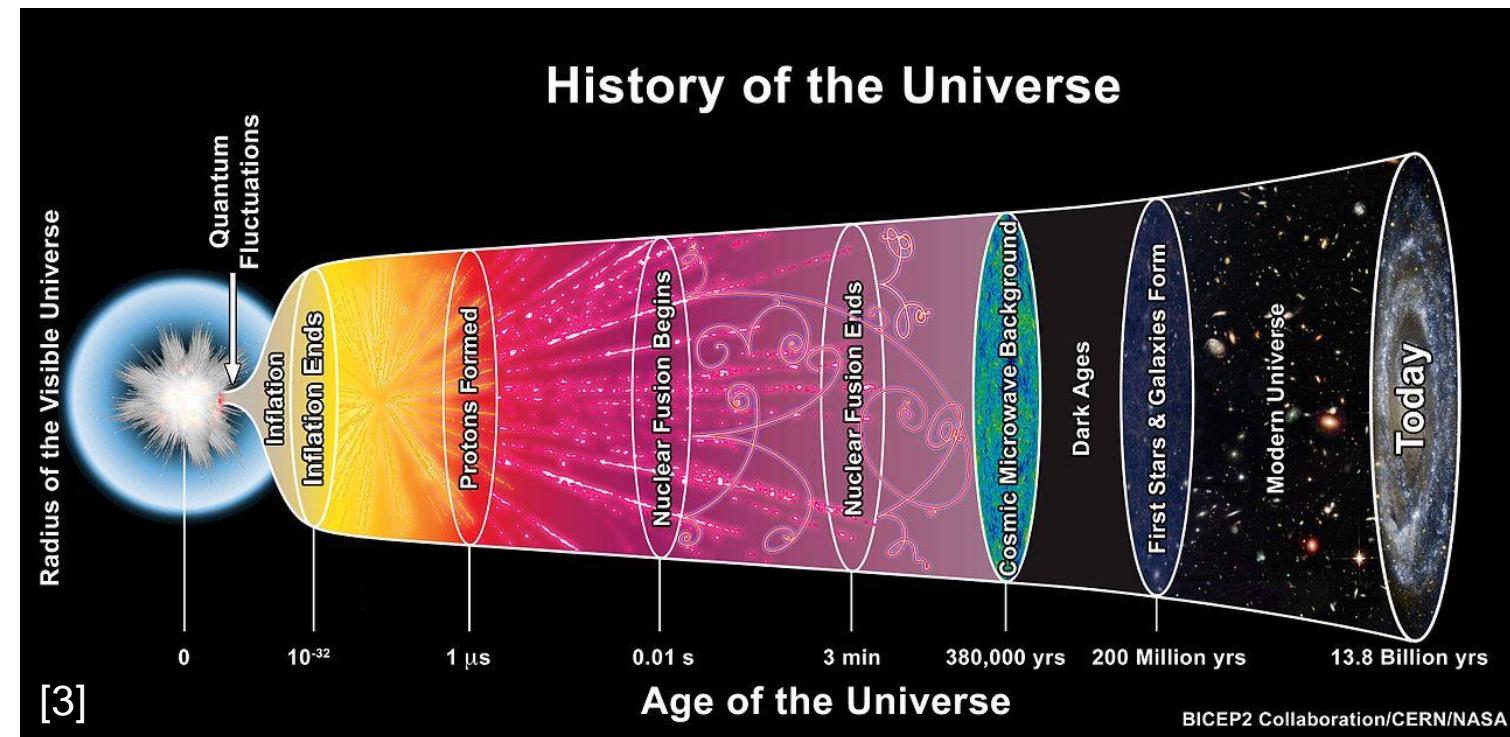
- Initial conditions of expanding universe

Sakharov's conditions [1]

- CP violation for baryogenesis

CPV in the Standard Model

- CKM phase δ_{CKM} (order 1)
- Strong CP-phase $\bar{\theta}$ (tiny)
- Unconfirmed: PMNS matrix, Majorana neutrinos, other operator?
- Required: Evaluation from complementary sources [2]*



“... the occurrence of C asymmetry is the consequence of violation of *CP* invariance in the nonstationary expansion of the hot universe during the superdense stage, as manifest in the difference between the partial probabilities of the charge-conjugate reactions. This effect has not yet been observed experimentally, but its existence is theoretically undisputed”
– [1]

[1] Sakharov (1991) Sov. Phys. Usp. 34 392

[2] Alarcon et al., (2022) arXiv 2203.08103

[3] The BICEP2 Collaboration CERN-NASA (1991) <https://home.cern/news/series/lhc-physics-ten/recreating-big-bang-matter-earth>

Research opportunities

- Low energy observables for physics beyond the Standard Model

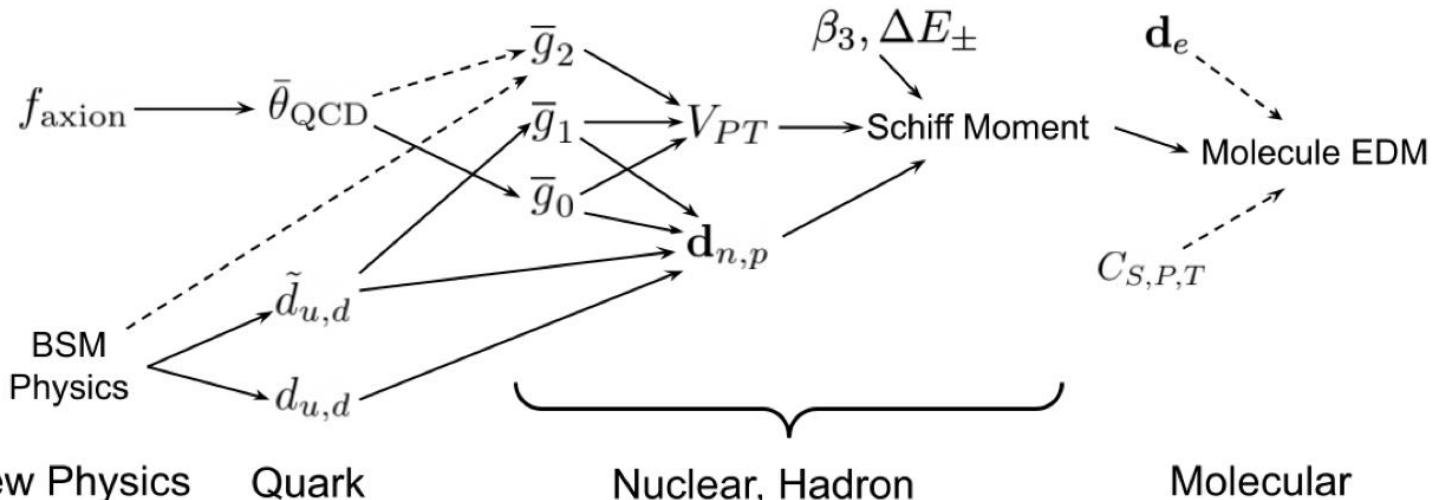
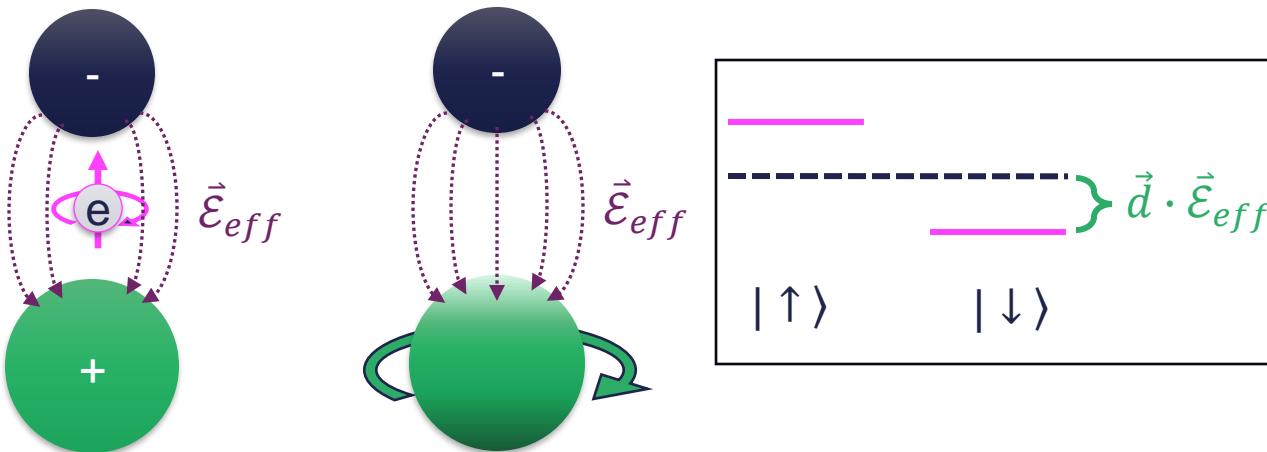
eEDM



- Sensitive to electron-coupled CPV

Hadronic CPV

- Enhanced in heavy, deformed nuclei



[1] Contribution to Snowmass 2021: *EDMs and the search for new physics*, arXiv 2203.08103 (2021)
[2] Safronova et al. (2018) *Rev. Mod. Phys.* 90, 2

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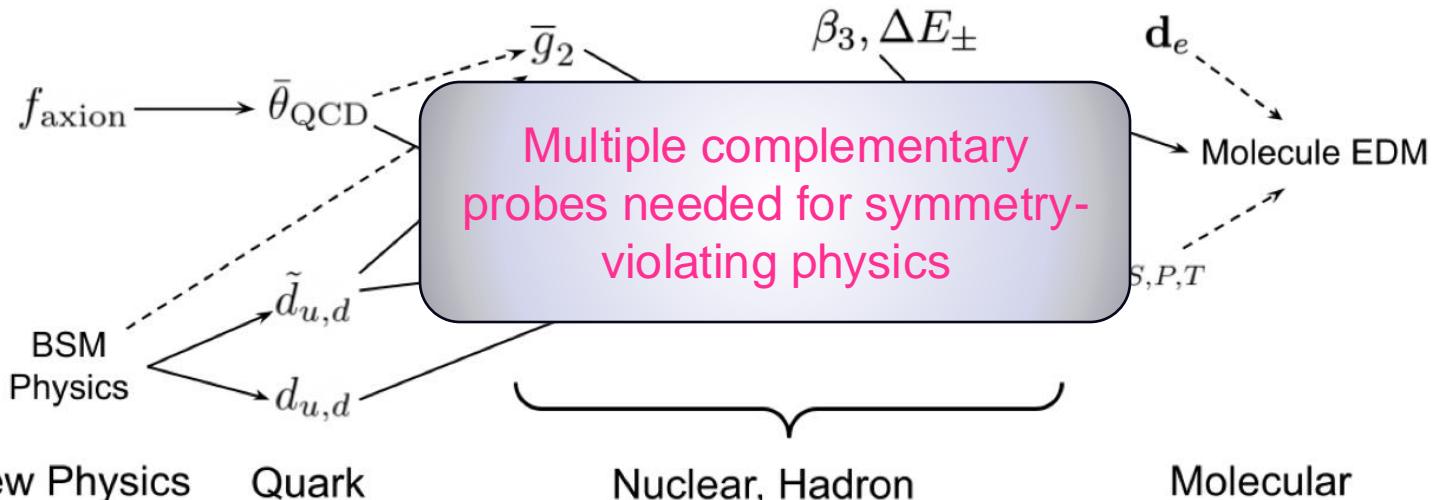
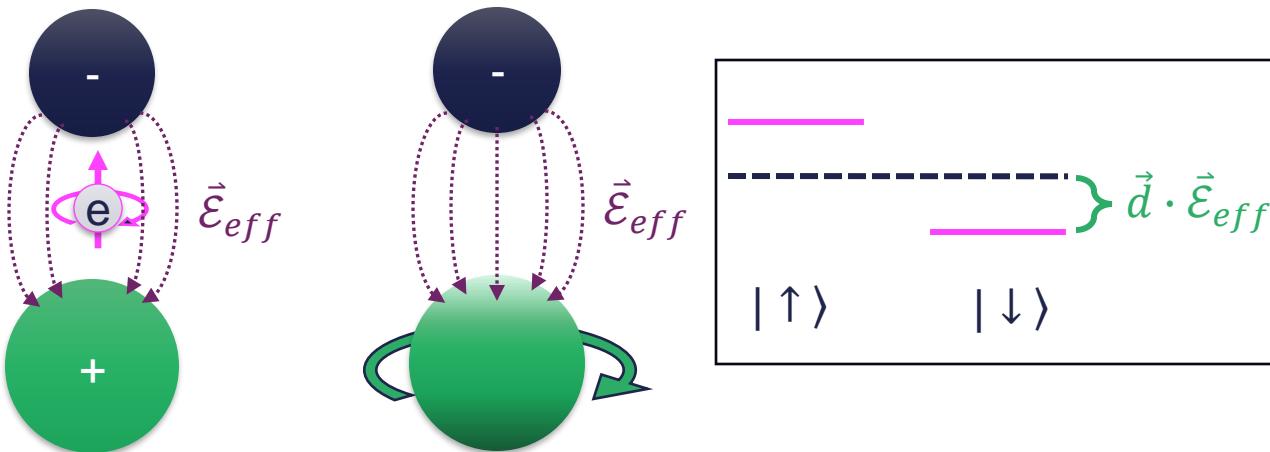
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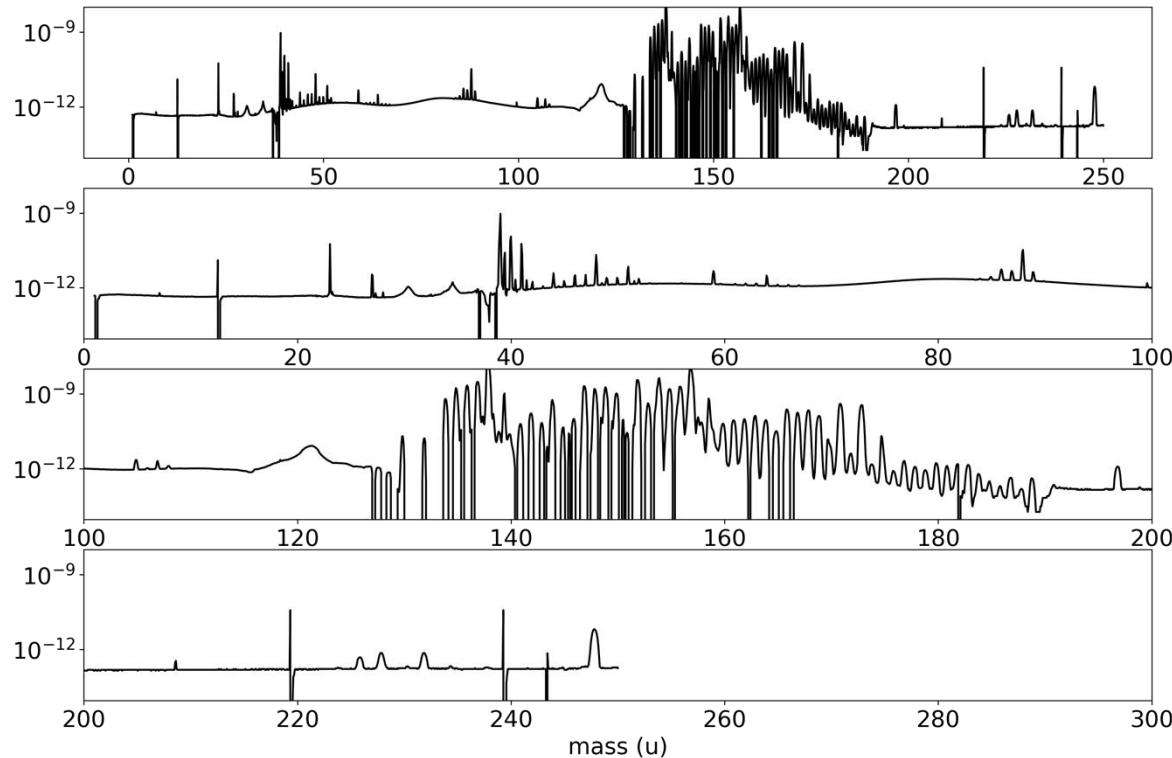
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Molecular beam production: Ion sources and effects

Surface

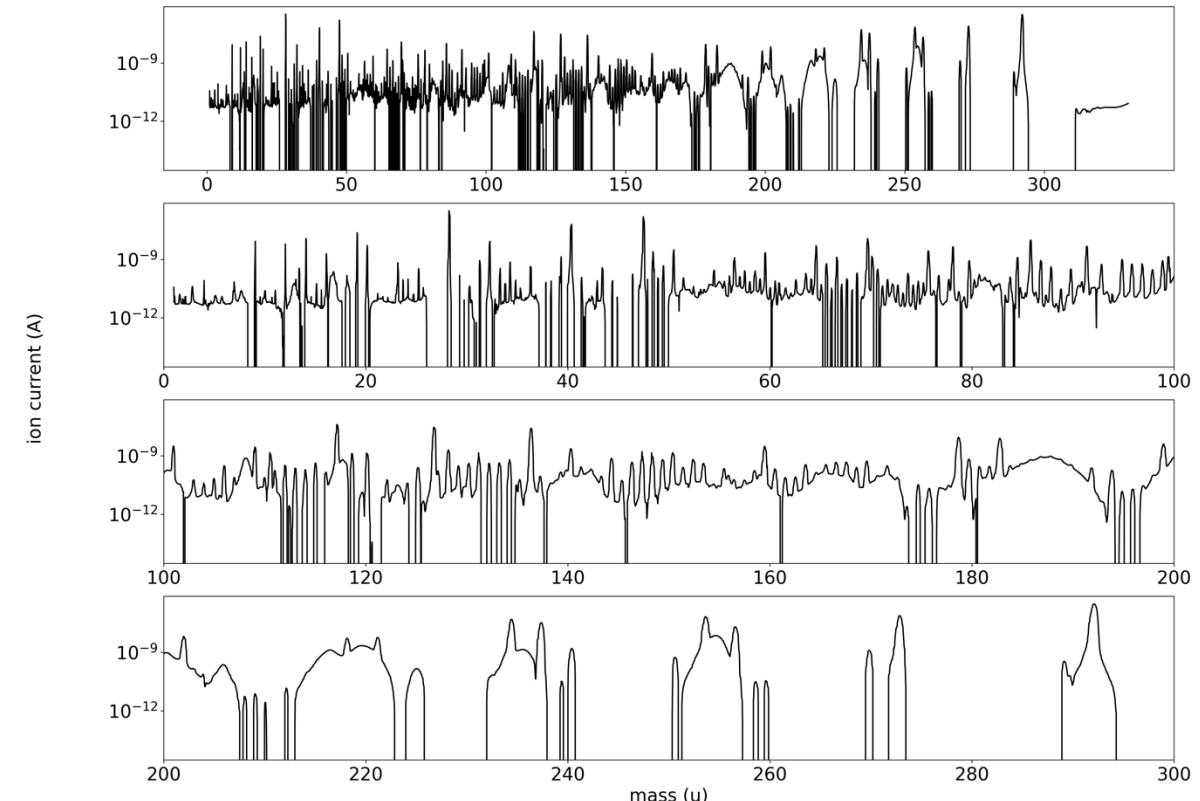
- Low IPs
- Surface ionization efficiency



[1] Wilkins et al., arXiv:2408.14673 (2024)

FEBIAD

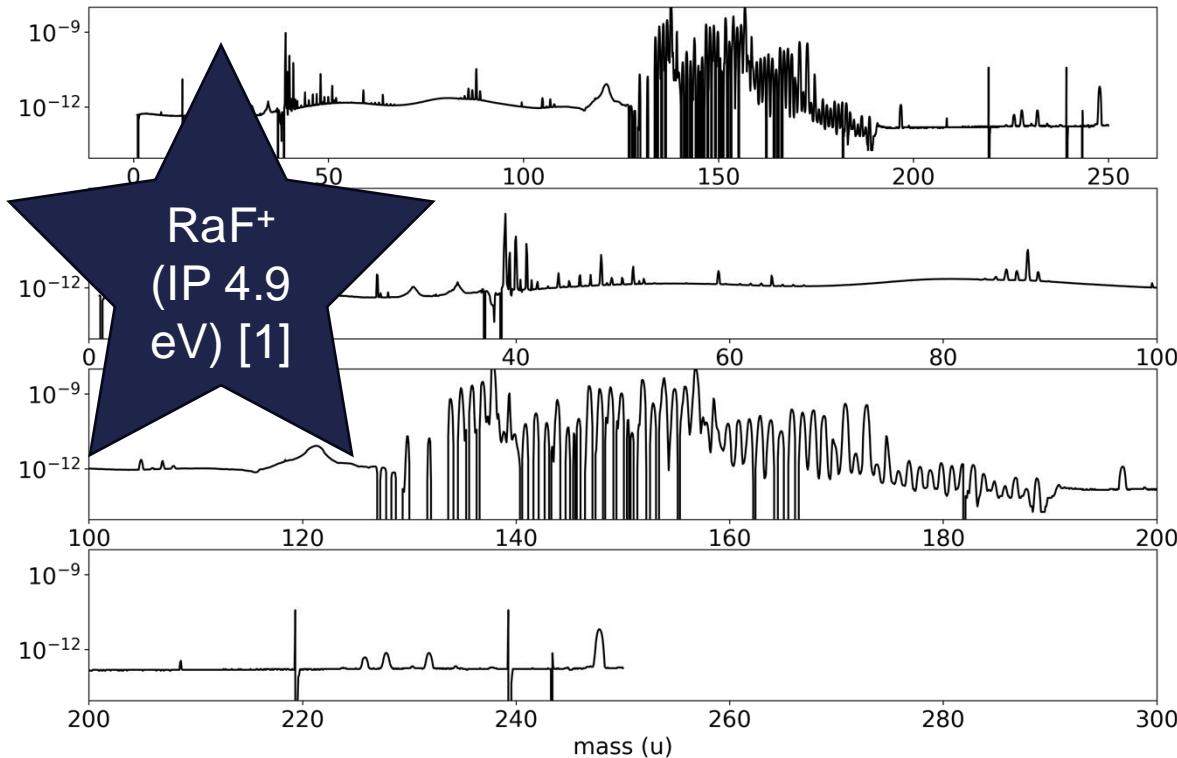
- High/unknown IPs
- High efficiency



Molecular beam production: Ion sources and effects

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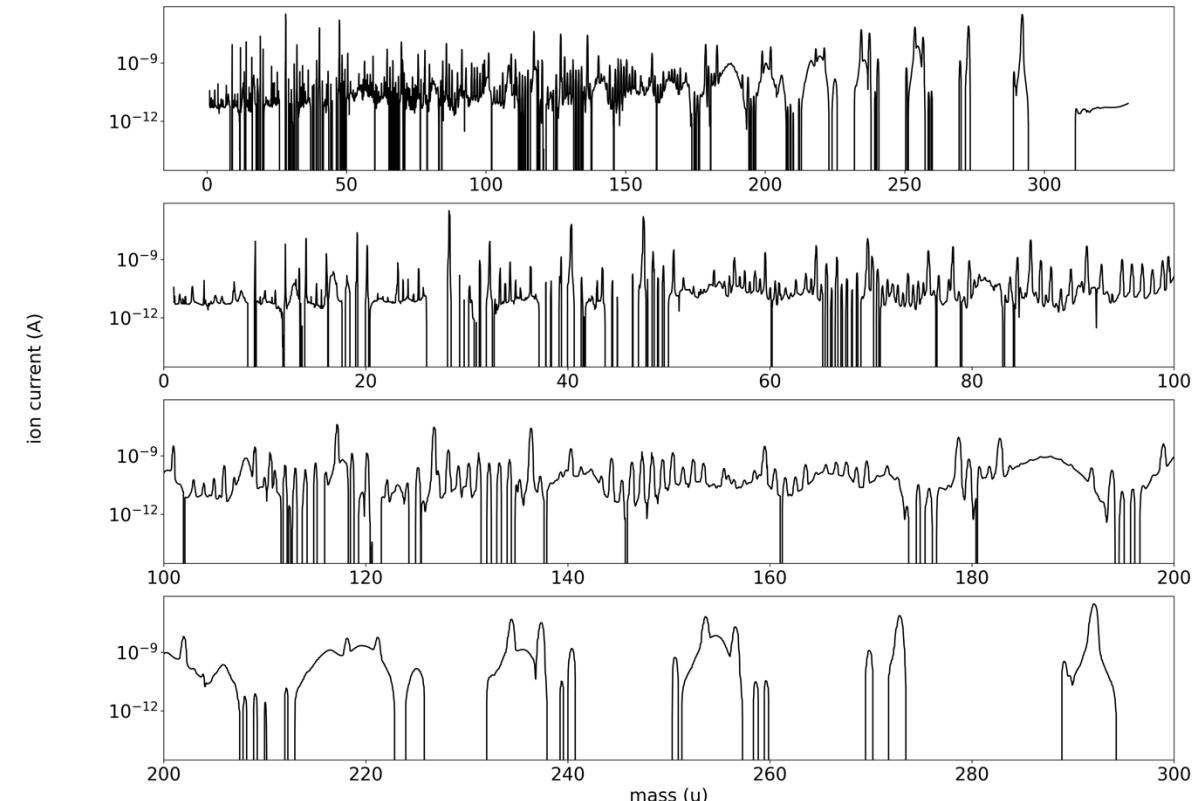
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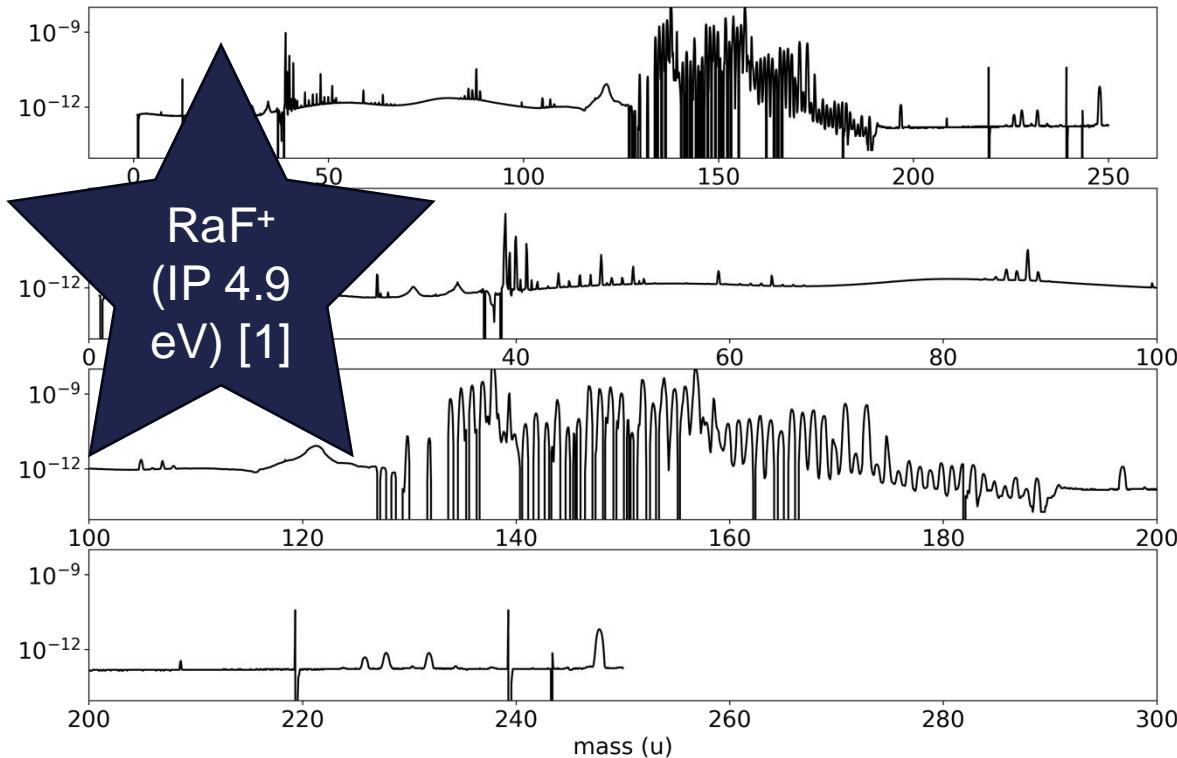
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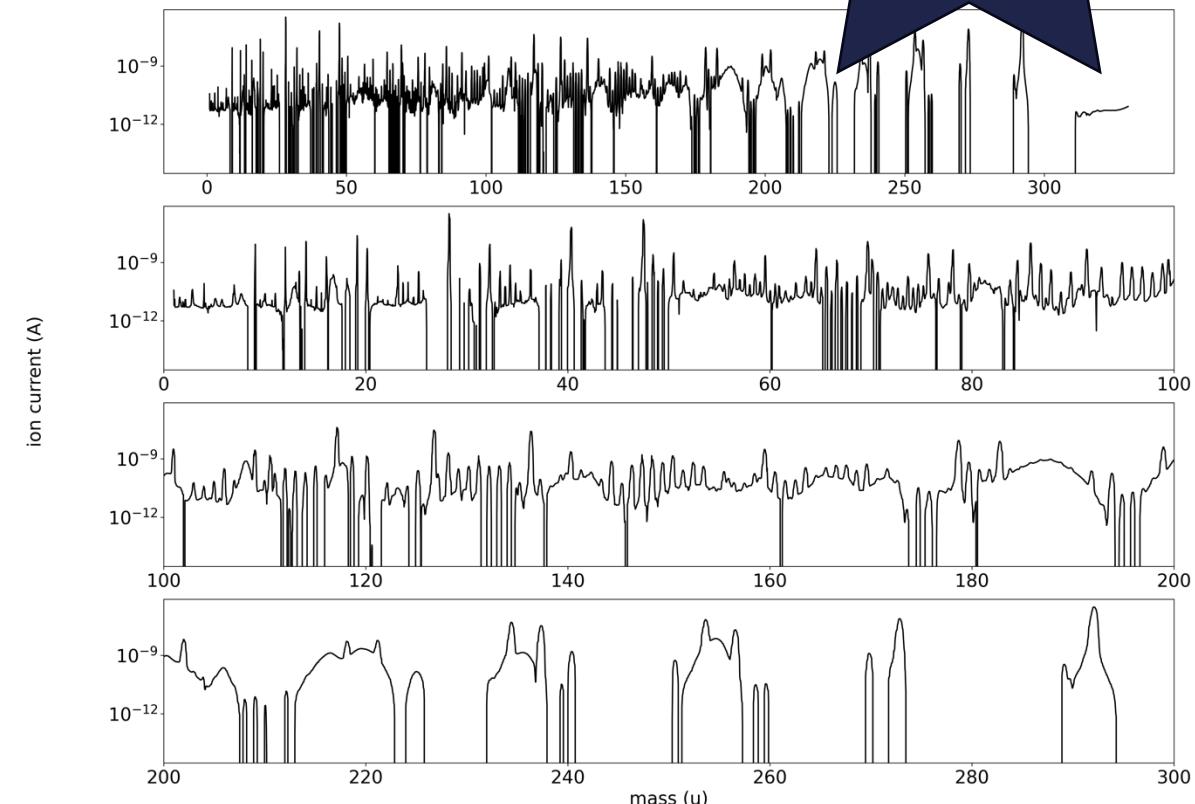
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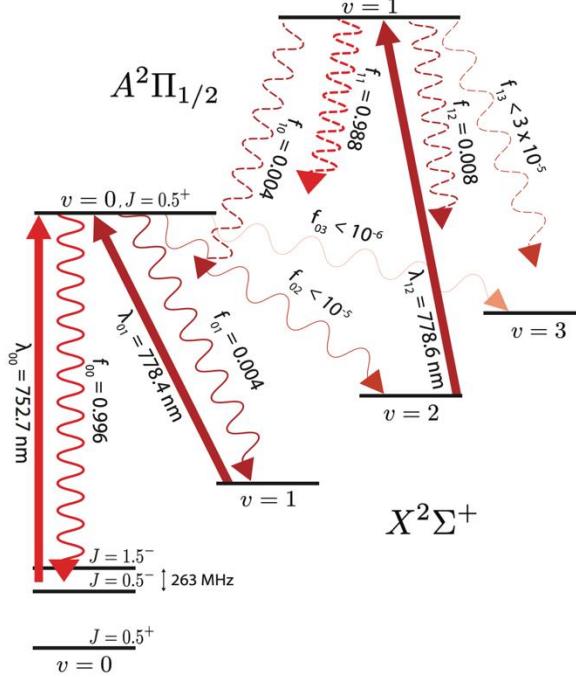
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Production of RaF



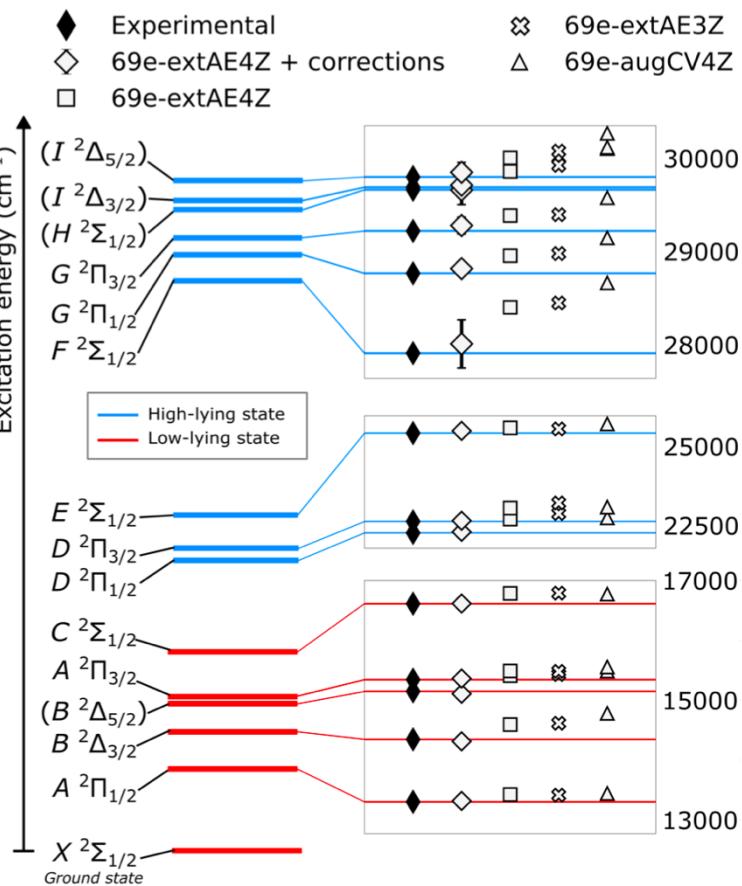
Laser cooling [1]



- [1] Udrescu *et al.*, Nat. Phys. 20, 202 (2024)
- [2] Athanasakis-Kaklamanakis *et al.*, arXiv 2308.14862 submitted (2024)
- [3] Athanasakis-Kaklamanakis *et al.*, PRA 110, L010802 (2024)
- [4] Wilkins *et al.*, arXiv 2311.04121 in review (2024)

Excited states [2]

- agreement $\geq 99.64\%$ (~ 12 meV)

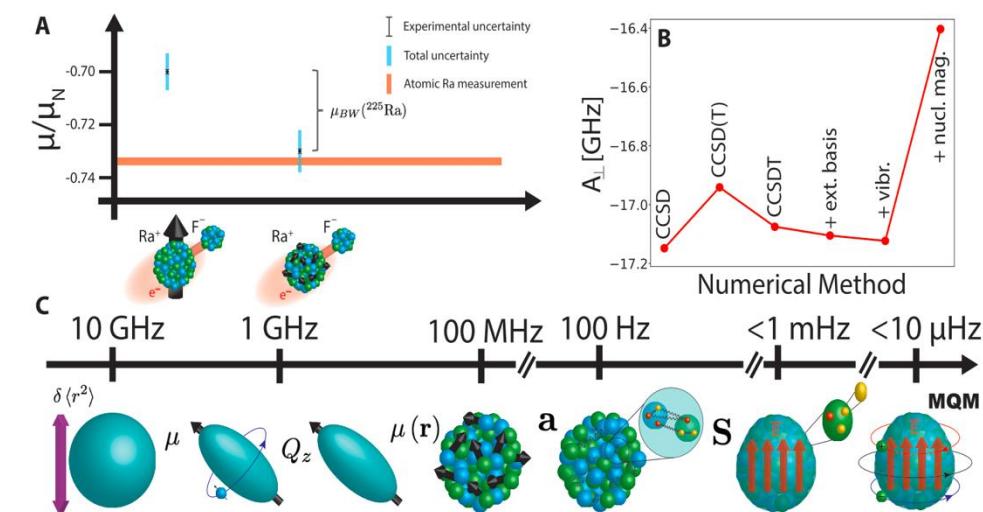


State lifetimes [3]

- Radiative lifetime of A ^2\Pi_{1/2} state

Nuclear magnetization effect [4]

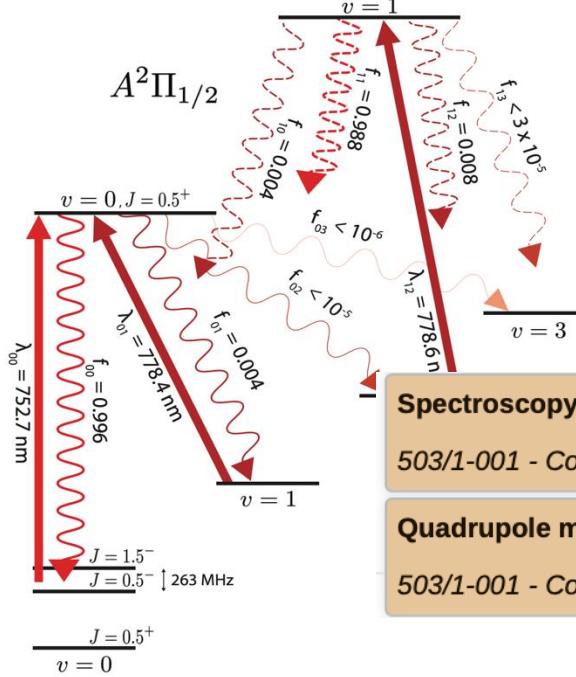
- $\mu(^{225}\text{Ra})$



Production of RaF

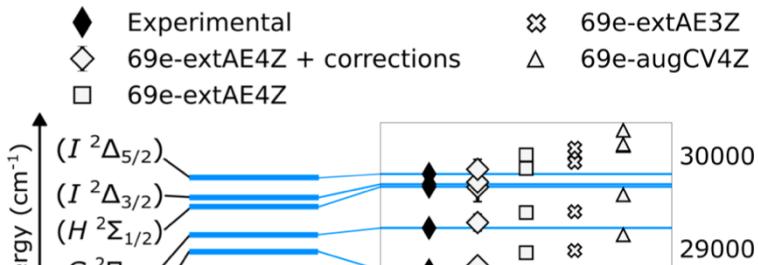


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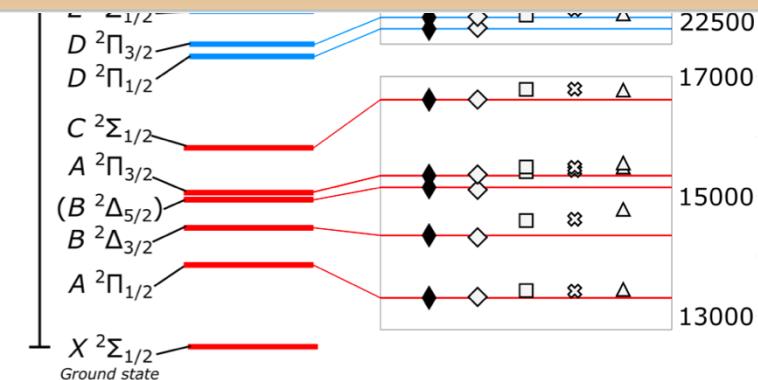


Spectroscopy of a radium containing diatomic molecule

503/1-001 - Council Chamber, CERN

Quadrupole moment of ^{223}Ra from molecular spectroscopy on ^{223}RaF

503/1-001 - Council Chamber, CERN



[1] Udrescu *et al.*, Nat. Phys. 20, 202 (2024)

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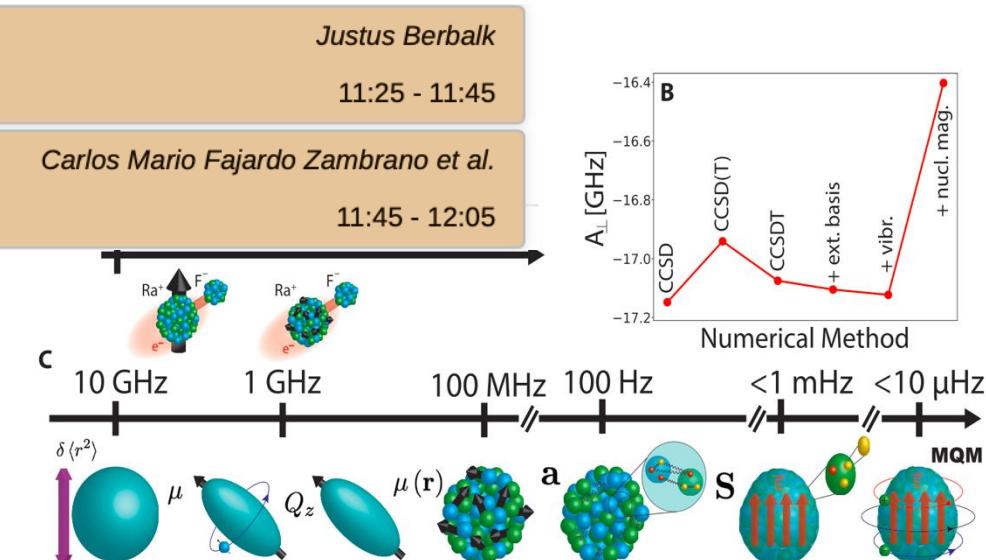
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- Radiative lifetime of $A\ ^2\Pi_{1/2}$ state

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Motivation: Actinium

[4]

Ac: Nuclear properties

- Octupole deformation
- Low-lying opposite parity states
- Schiff moment enhancement

TABLE IV. Schiff moments (S) and EDMs (d_A) of some atoms in terms of the QCD θ -term constant $\bar{\theta}$. We remind the reader that the current experimental limit is $|\bar{\theta}| < 10^{-10}$.

Z	Atom	S [$e \text{ fm}^3 \bar{\theta}$]	$10^{-17} S$ [$e \text{ fm}^3$]	d_A [$e \text{ cm}$] $10^{-17} \bar{\theta}$
63	^{153}Eu	-3.7	-1.63	6
63	$^{153}\text{Eu}^{3+}$	-3.7	0.33	-1.2
66	^{161}Dy	$\lesssim 4$	-2.23	$\lesssim 9$
80	^{199}Hg	0.005	-2.50	-0.013
81	$^{205,203}\text{Tl}^+$	0.02	-2.79	-0.06
82	$^{207}\text{Pb}^{2+}$	0.005	-2.99	-0.015
86	^{223}Rn	-3	3.3	-10
87	$^{223}\text{Fr}^+$	1.6	2.87	4.6
88	^{225}Ra	-1	-8.25	8
89	^{227}Ac	-6	-10.1	60
89	$^{227}\text{Ac}^+$	-6	-9.8	60
90	$^{229}\text{Th}^{2+}$	$\lesssim 2$	-6.93	$\lesssim 14$
91	$^{229}\text{Pa}^{\text{a}}$	-40	-11.4	460
92	^{233}U	$\lesssim 2$	-12.1	$\lesssim 20$
93	^{237}Np	-4	-7.5	30
94	^{239}Pu	$\lesssim 0.1$	-9.2	$\lesssim 1$

^aEstimates for ^{229}Pa are presented assuming that the existence of a very close nuclear doublet level will be confirmed.

- [1] Verstraelen et al., Phys. Rev. C. 100, 044321 (2019)
- [2] Heinke et al., CERN-INTC-2020-029, INTC-P-556 (2020)
- [3] Flambaum, Feldmeier, Phys. Rev. C. 101, 015502 (2020)
- [4] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

Motivation: Actinium Fluoride [4]

Ac: Nuclear properties

- Octupole deformation
- Low-lying opposite parity states
- Schiff moment enhancement

AcF: molecular enhancement

- Enhanced sensitivity to CP-violating observables?

TABLE IV. Schiff moments (S) and EDMs (d_A) of some atoms in terms of the QCD θ -term constant $\bar{\theta}$. We remind the reader that the current experimental limit is $|\bar{\theta}| < 10^{-10}$.

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^aEstimates for ^{229}Pa are presented assuming that the existence of a very close nuclear doublet level will be confirmed.

The interaction constant W_S for the effective T,P-violating interaction in ^{227}Ac -containing molecules is

$$W_S \approx 46000 \text{ a.u.} \quad (55)$$

The energy shift is

$$2W_S S = 5 \times 10^7 \bar{\theta} \text{ h Hz.} \quad (56)$$

- [1] Verstraelen et al., Phys. Rev. C. 100, 044321 (2019)
[2] Heinke et al., CERN-INTC-2020-029, INTC-P-556 (2020)
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[4] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

[3]

Motivation: Actinium Fluoride [4]

Ac: Nuclear properties

- Octupole deformation
- Low-lying opposite parity states
- Schiff moment enhancement

AcF: molecular enhancement

- Enhanced sensitivity to CP-violating observables?

Production

- IP: ? D_e: ?

[3]

TABLE IV. Schiff moments (S) and EDMs (d_A) of some atoms in terms of the QCD θ -term constant $\bar{\theta}$. We remind the reader that the current experimental limit is $|\bar{\theta}| < 10^{-10}$.

Z	Atom	S [$e \text{ fm}^3 \bar{\theta}$]	$10^{-17} S$ [$e \text{ fm}^3$]	d_A [$e \text{ cm}$] $10^{-17} \bar{\theta}$
63	¹⁵³ Eu	-3.7	-1.63	6
63	¹⁵³ Eu ³⁺	-3.7	0.33	-1.2
66	¹⁶¹ Dy	$\lesssim 4$	-2.23	$\lesssim 9$
80	¹⁹⁹ Hg	0.005	-2.50	-0.013
81	^{205,203} Tl ⁺	0.02	-2.79	-0.06
82	²⁰⁷ Pb ²⁺	0.005	-2.99	-0.015
86	²²³ Rn	-3	3.3	-10
87	²²³ R _r ⁺	1.6	2.87	4.6
88	²²⁵ Ra	-1	-8.25	8
89	²²⁷ Ac	-6	-10.1	60
89	²²⁷ Ac ⁺	-6	-9.8	60
90	²²⁹ Th ²⁺	$\lesssim 2$	-6.93	$\lesssim 14$
91	²²⁹ Pa ^a	-40	-11.4	460
92	²³³ U	$\lesssim 2$	-12.1	$\lesssim 20$
93	²³⁷ Np	-4	-7.5	30
94	²³⁹ Pu	$\lesssim 0.1$	-9.2	$\lesssim 1$

^aEstimates for ²²⁹Pa are presented assuming that the existence of a very close nuclear doublet level will be confirmed.

The interaction constant W_S for the effective T,P-violating interaction in ²²⁷Ac-containing molecules is

$$W_S \approx 46000 \text{ a.u.} \quad (55)$$

The energy shift is

$$2W_S S = 5 \times 10^7 \bar{\theta} \text{ h Hz.} \quad (56)$$

- [1] Verstraelen et al., Phys. Rev. C. 100, 044321 (2019)
- [2] Heinke et al., CERN-INTC-2020-029, INTC-P-556 (2020)
- [3] Flambaum, Feldmeier, Phys. Rev. C. 101, 015502 (2020)
- [4] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

Motivation: Actinium-225

T- α T (targeted alpha therapy)

- Damage to cancer cells
 - DNA double strand breaks, membrane, mRNA damage
 - Ionization through free radicals
- High linear energy transfer

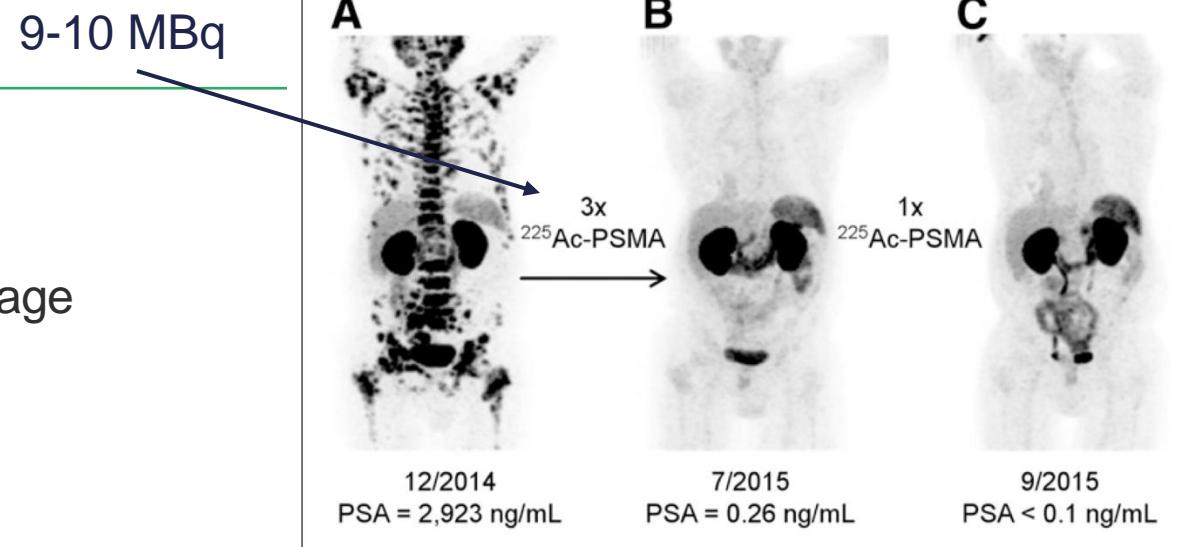


Figure published in: Kratchowil *et al.* (2016) J. Nucl. Med. 57 1941-1944

Production routes

- ^{226}Ra
 - $^{226}\text{Ra}(\text{p},2\text{n})^{225}\text{Ac}$
- **^{225}Ra (generator)**
 - $^{226}\text{Ra}(\gamma,\text{n})^{225}\text{Ra} \rightarrow ^{225}\text{Ac}$
 - $^{226}\text{Ra}(\text{n},2\text{n})^{225}\text{Ra} \rightarrow ^{225}\text{Ac}$

Actinide

- $^{\text{nat}}\text{Th}(\text{p},\text{x})^{225}\text{Ac}$, ^{225}Ra ($^{\text{nat}}\text{Th}(\text{p},\text{x})^{227}\text{Ac}$, ^{227}Ra)
- $^{\text{nat}}/\text{dep}\text{U}(\text{p},\text{x})^{225}\text{Ac}$, ^{225}Ra ($^{\text{Nat}}/\text{dep}\text{U}(\text{p},\text{x})^{227}\text{Ac}$, ^{227}Ra)

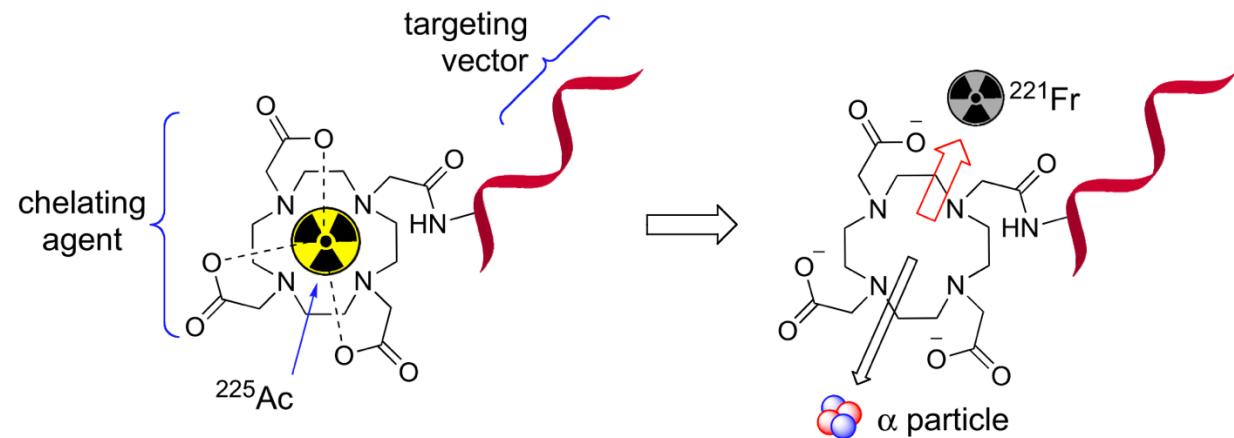


Figure published in: Robertson *et al.* (2018) Current Radiopharmaceuticals. 11 156-172

Motivation: Actinium-225

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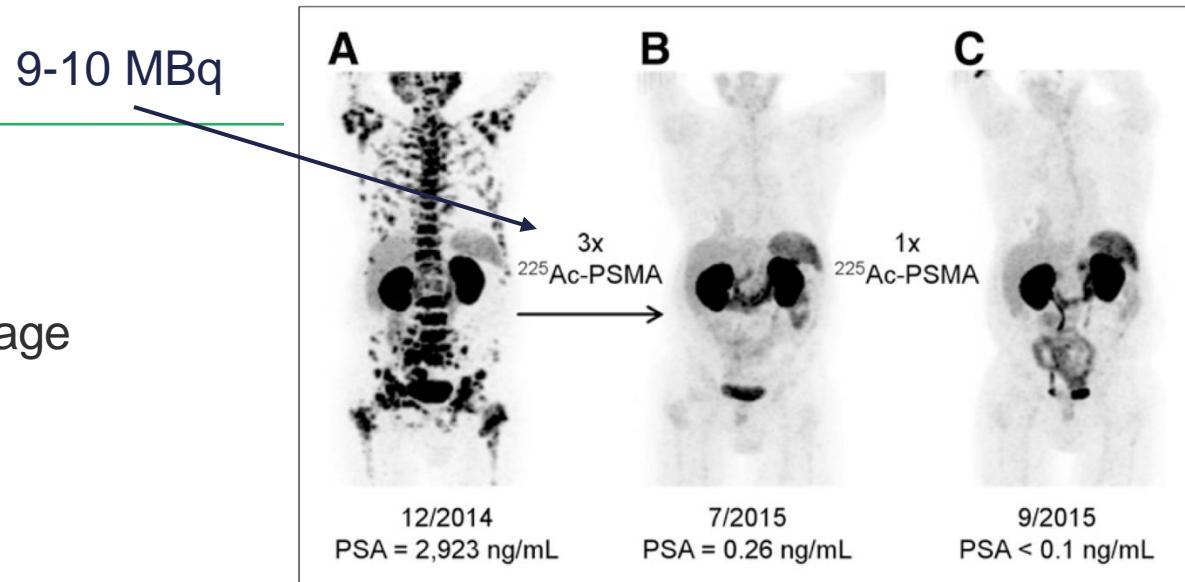


Figure published in: Kratchowil *et al.* (2016) J. Nucl. Med. 57 1941-1944

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Jake David Johnson *et al.*

11:35 - 12:05

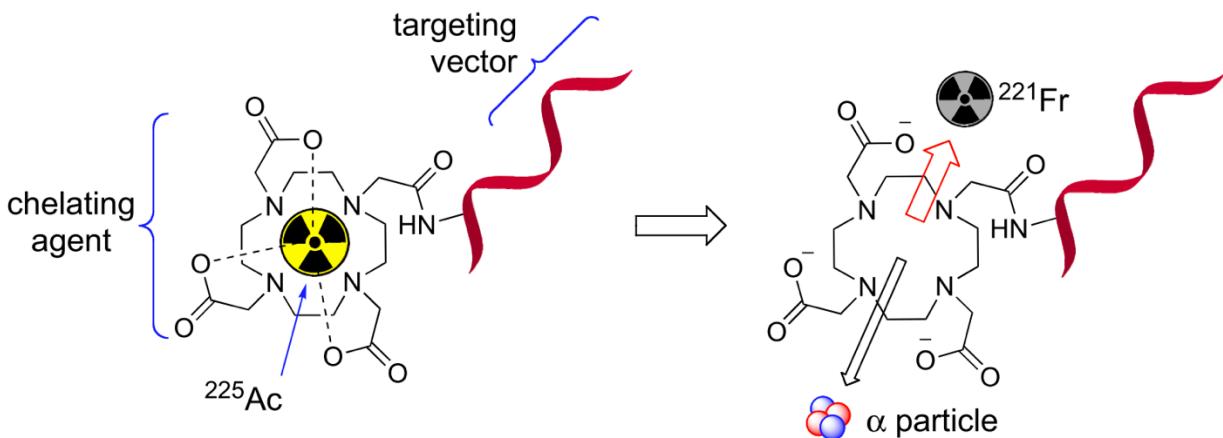


Figure published in: Robertson *et al.* (2018) Current Radiopharmaceuticals. 11 156-172

Motivation: Actinium-225

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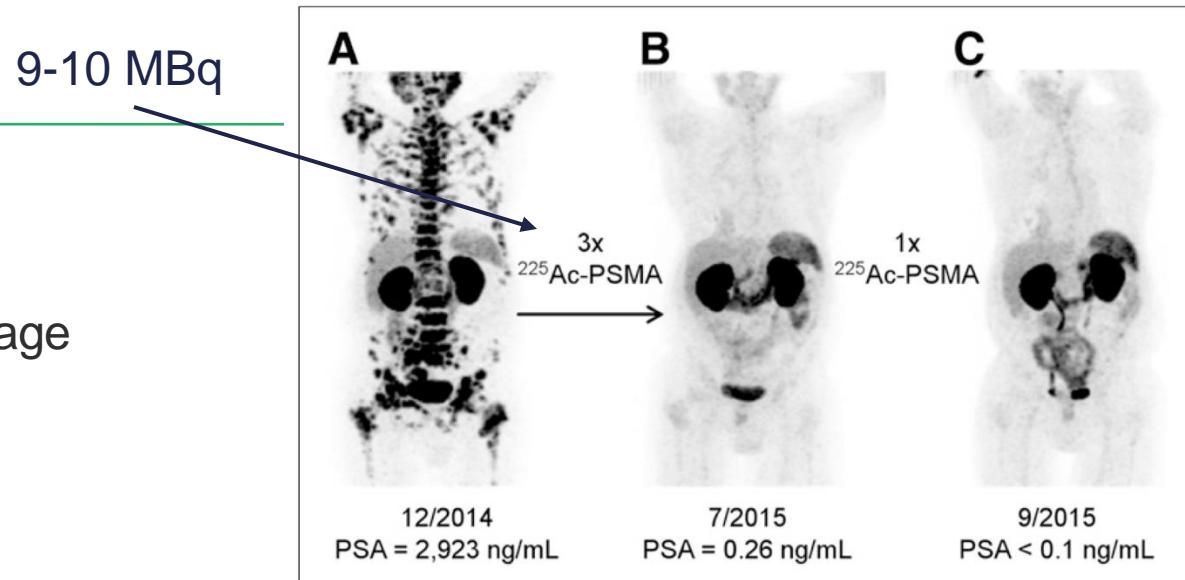


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11:35 - 12:05

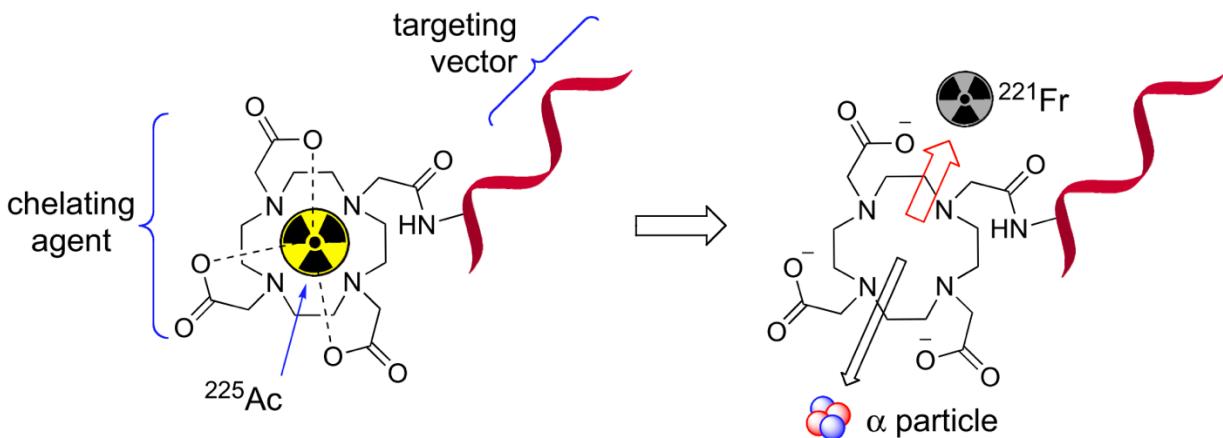
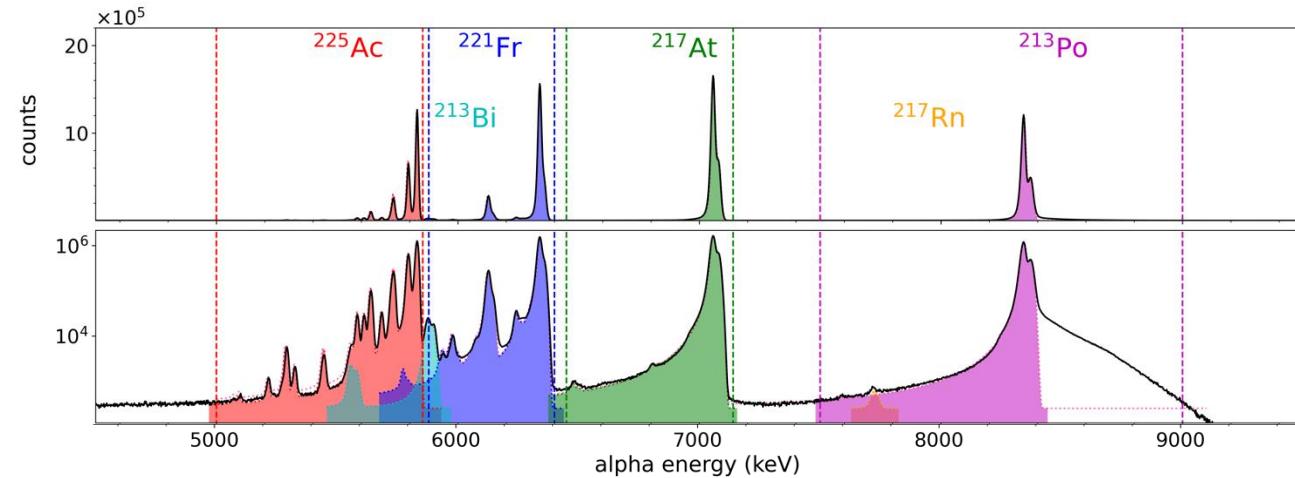
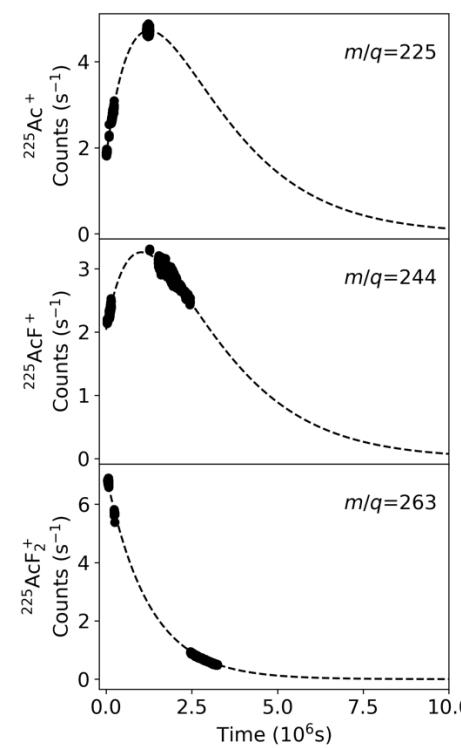
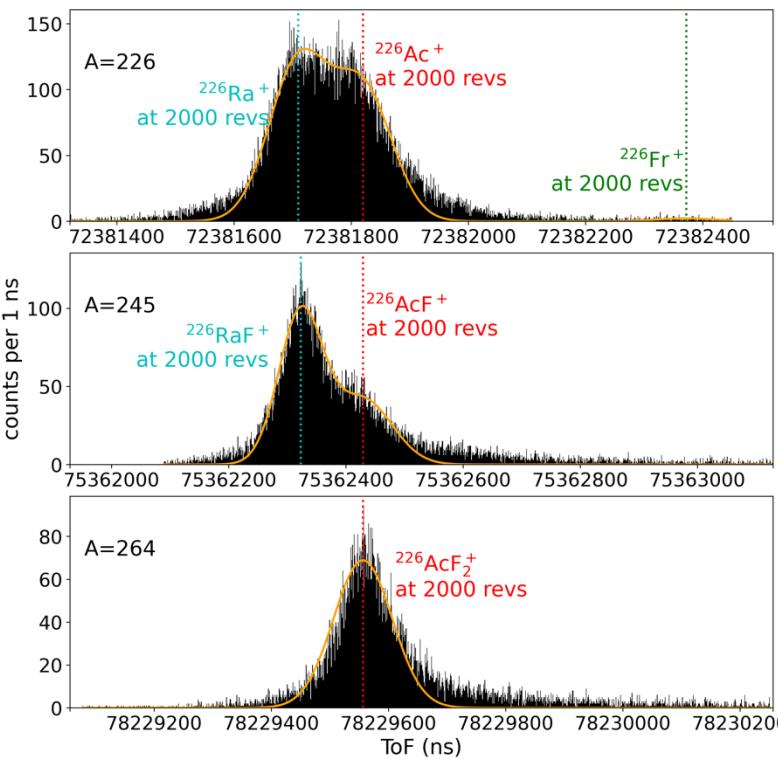


Figure published in: Robertson *et al.* (2018) Current Radiopharmaceuticals. 11 156-172

Production of AcF_x^+

- AcF spectroscopy - characterization
- ^{225}Ac : Targeted-alpha therapy
- Ac: enhanced extraction



[1] Au, *PhD thesis* (2023)

[2] Au et al., *in review* (2024)



AcF spectroscopy: characterization

ISOLDE

Proposal [1]

Production

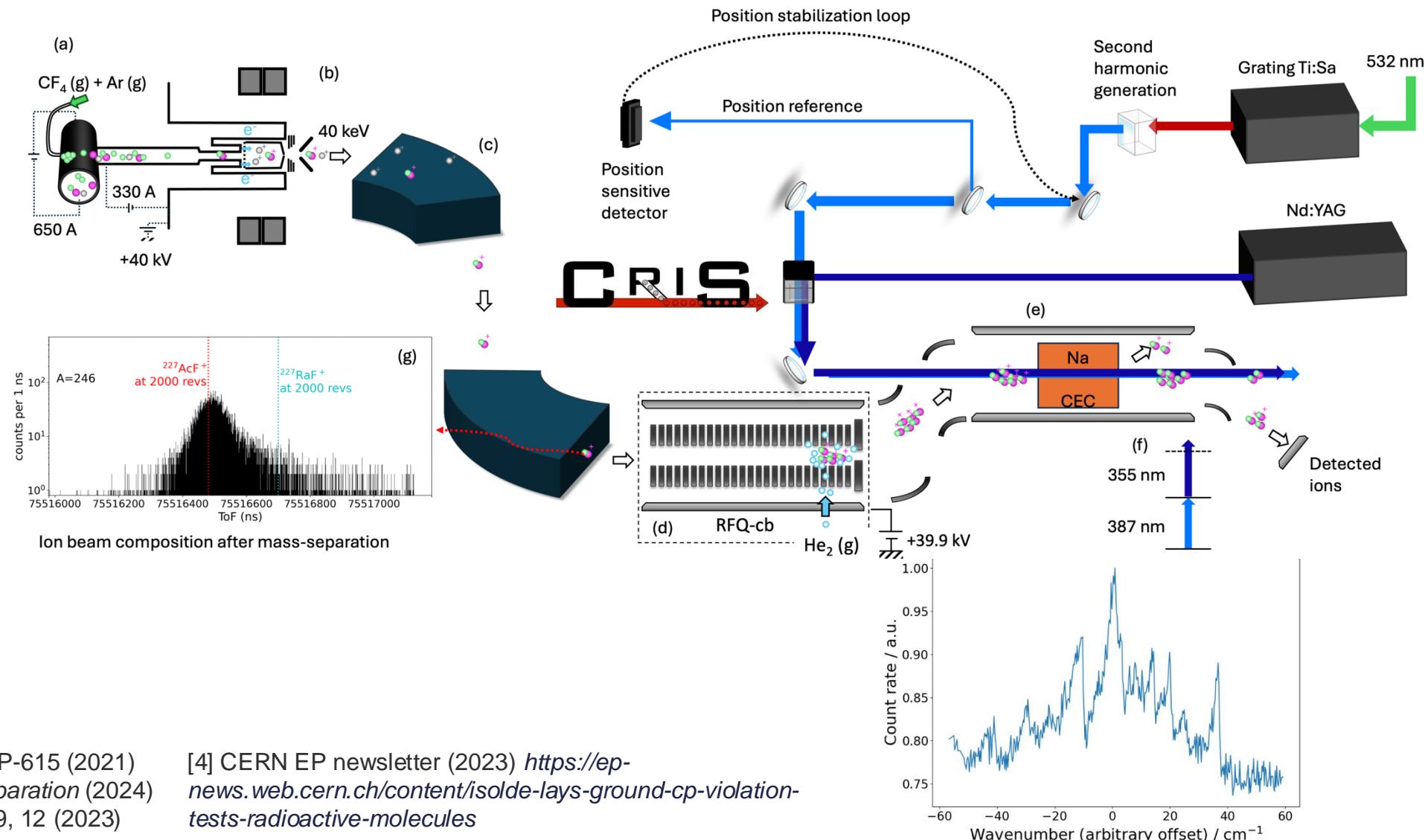
- 1 year proposal to experiment

Experiment

- CRIS

Molecular theory

- IH-FS-RCCSD [3]
- IP = $48,866 \text{ cm}^{-1}$
- $D_e = 57,214 \text{ cm}^{-1}$



- [1] Athanasakis-Kaklamanakis et al, INTC-P-615 (2021)
[2] Athanasakis-Kaklamanakis et al, *in preparation* (2024)
[3] Oleynichenko et al., *J. Chem. Phys.* 159, 12 (2023)

- [4] CERN EP newsletter (2023) <https://ep-news.web.cern.ch/content/isolde-lays-ground-cp-violation-tests-radioactive-molecules>

The challenging case: protactinium

Protactinium

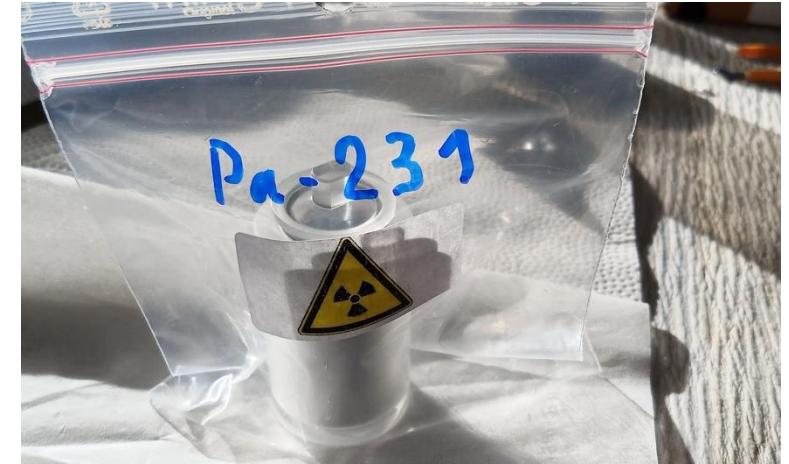
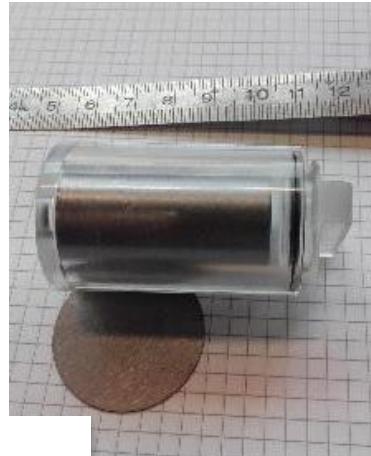
- Laser-ionized: tried, failed

Protactinium chemistry at ISOLDE from external sources

September 22, 2023

M. Au^{1,2}, M. Athanasakis-Kaklamakanis^{1,3}, L. Nies¹, K. Blaum⁴ C. Duchemin¹, Ch.E. Düllmann^{2,5,6}, C. M. Fajardo-Zambrano³, P. F. Giesel⁷, M. Heaven⁸, L. Lambert¹, D. Lange⁴, U. Köster⁹, G. Neyens³, D. Renisch^{2,6}, S. Rothe¹, Ch. Schweiger⁴, L. Schweikhardt⁷, J. Stricker^{2,6}, W. Wojtaczka³

950 Bq 231-Pa (1.417e+15 atoms)



Protactinium results: preliminary

Pa molecules

- $^{231}\text{Pa}^+$, $^{231}\text{PaO}^+$, $^{231}\text{PaO}_2^+$, $^{231}\text{PaF}^+$, $^{231}\text{PaF}_2^+$, $^{231}\text{PaF}_3^+$, $^{231}\text{PaFO}^+$, $^{231}\text{PaF}_2\text{O}^+$

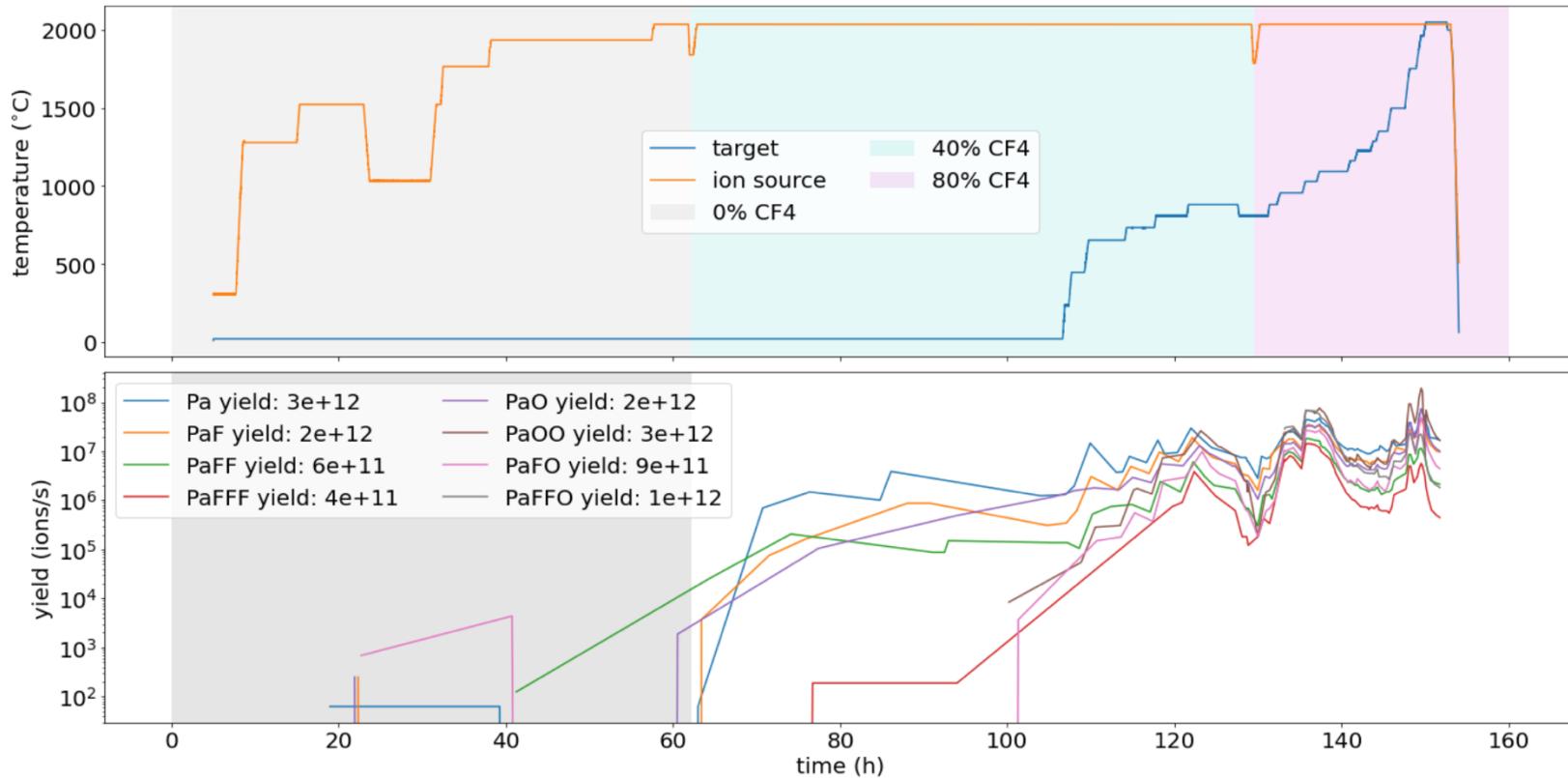
Formed in RFQ:

- From $^{231}\text{Pa}^+$: $^{231}\text{PaO}^+$, $^{231}\text{PaO}_2^+$
- From $^{231}\text{PaF}^+$: $^{231}\text{PaFO}^+$,
 $^{231}\text{PaFOH}_2^+$
- From $^{231}\text{PaF}_2^+$: $^{231}\text{PaF}_2\text{O}^+$,
- From $^{231}\text{PaO}^+$: $^{231}\text{PaO}_2^+$,

Checked but not seen:

- PaN^+ , PaC^+ , PaFO_2^+ , PaNO^+

Preliminary yield across all measured sidebands: $1.2\text{e}+13 / 1.417\text{e}+15 = 0.8\%$



Fluoride beams current status

- RaF_x : developed, available*
- AcF_x : developed, available*
- NpF_x , PuF_x : observed
- PaF_x , PaO_x : external sources, development
- ThF_x , UF_x : not observed
- ScF_x , TbF_x : ongoing development
- VF_x : requested

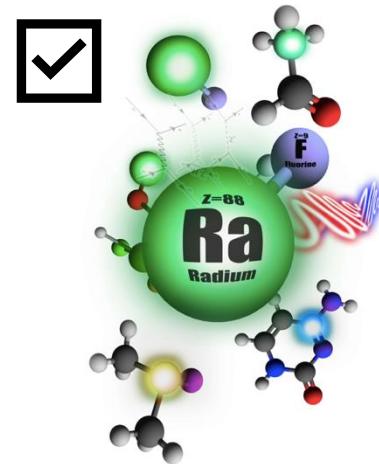
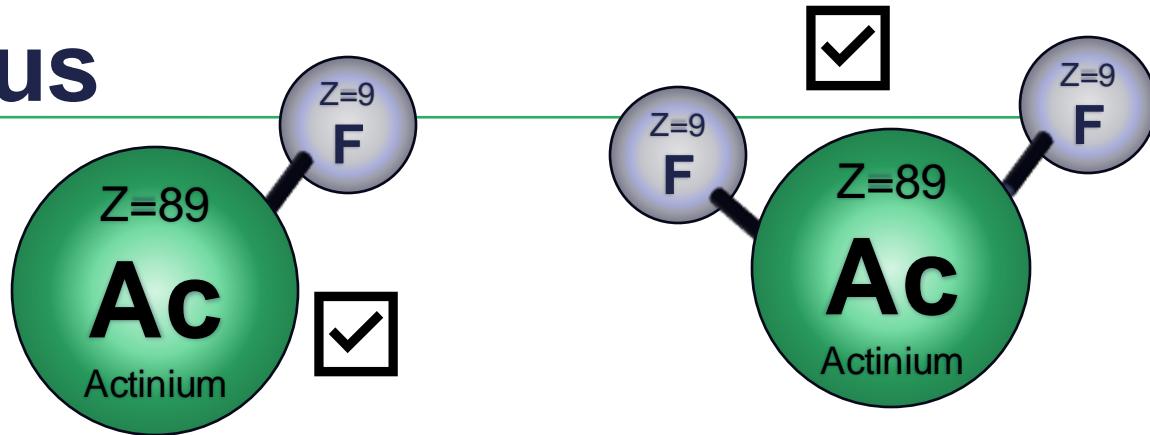


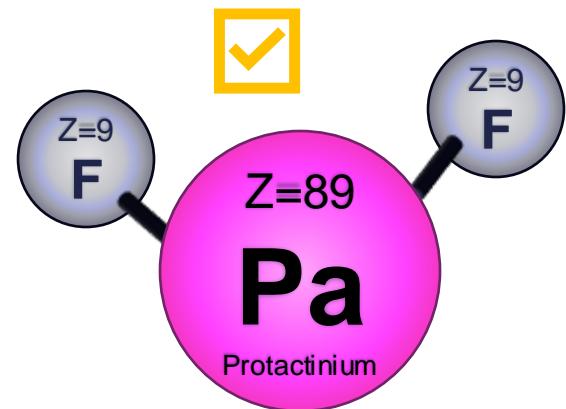
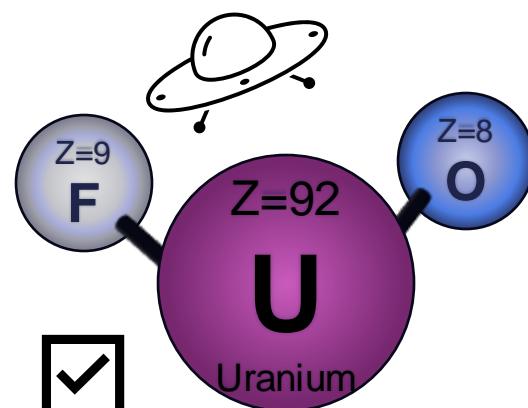
Image published in EP
Newsletter, CERN (2020)

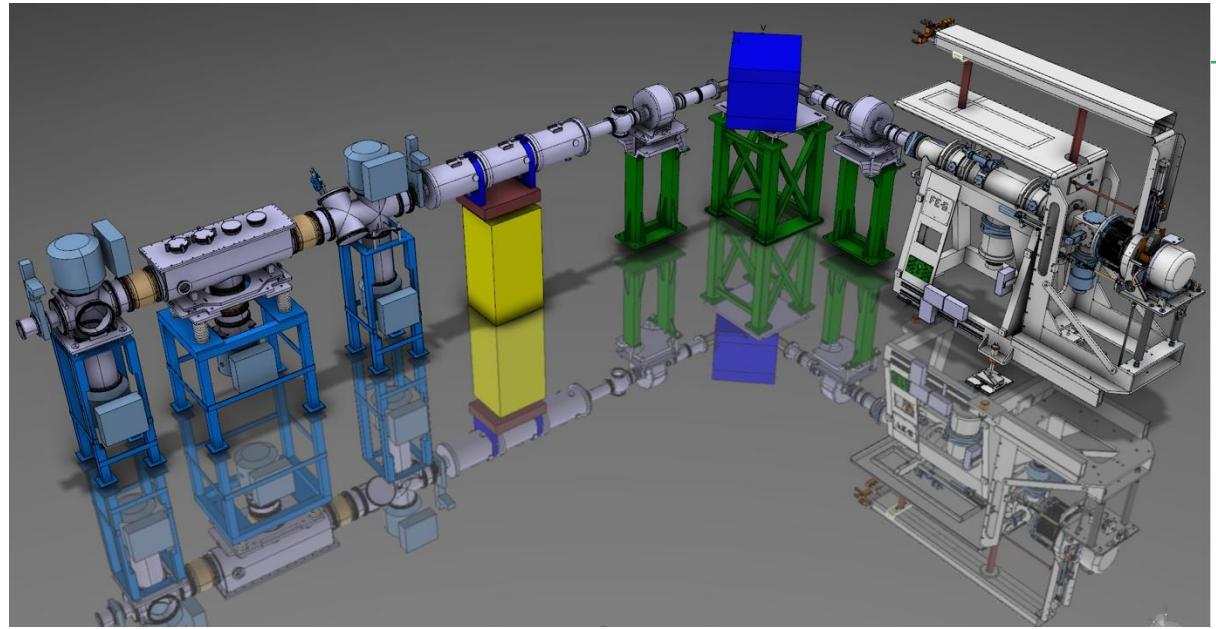


Next steps

- Characterization
- Target developments

*Yield publication in progress

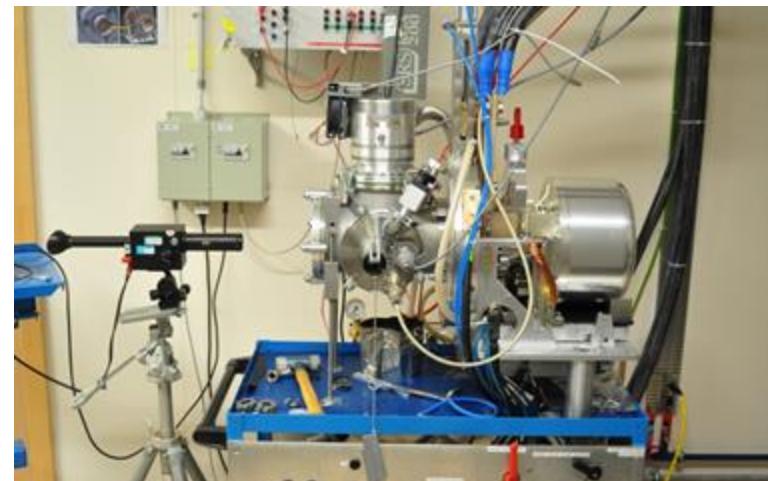




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ISOLDE OFFLINE 2
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ISOLDE
PUMP STAND
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Behind the scenes: Offline developments



Next steps

Molecular formation

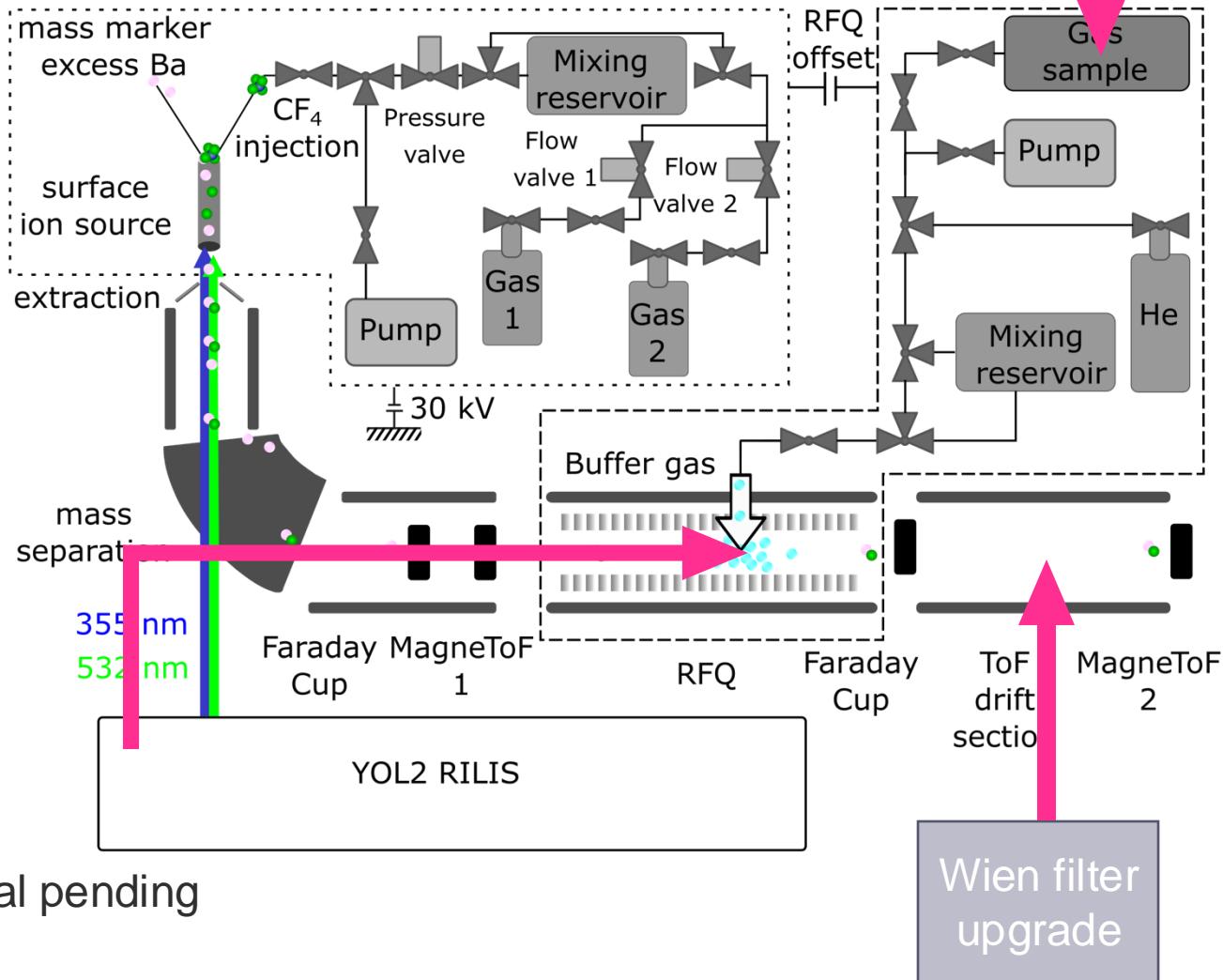
- Ion source developments
- RFQ gas mixing and injection tests
- Implantation/ablation

Laser setup

- New end of beamline
- Laser path to RFQ

Characterization / spectroscopy

- In-source: Offline LIST – fluorides, oxides
- In-trap: molecular formation studies – polyatomics
- Towards high precision radioactive studies: proposal pending



Acknowledgements



-

Michail Athanasakis-Kaklamakanis, Jochen Ballof, Ermanno Barbero, Mathieu Bovigny, Katerina Chrysalidis, Bernard Crepieux, Charlotte Duchemin, Christoph Düllmann, Paul Florian Giesel, Paul Fischer, Simone Gilardoni, Reinhard Heinke, Jake Johnson, Ulli Köster, Laura Lambert, Daniel Lange, Bruce Marsh, Maxime Mugeot, Lukas Nies, Bianca Reich, Jordan Reilly, Edgar Reis, Sebastian Rothe, Moritz Schlaich, Christoph Schweiger, Simon Stegemann, Jonas Stricker, Yago Nel Vila Gracia, Julius Wessolek, Frank Wienholtz, Shane Wilkins, Wiktoria Wojtaczka,

ISOLDE operations team, ISOLDE RILIS, CRIS, and ISOLTRAP teams
ISOLDE targets and ion sources team

This project has received funding from the European's Union Horizon 2020 Research and Innovation Programme under grant agreement number 861198 project 'LISA' (Laser Ionization and Spectroscopy of Actinides) Marie Skłodowska-Curie Innovative Training Network (ITN)

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TECHNISCHE
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DARMSTADT

TISD 2023

 =

GPS Schedule 2023																																
WK	April		May		June		July		August		September		October		November																	
	15	16	17	18	19	20	21	22	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46		
MO																																
TU																																
WE																																
TH																																
FR																																
SA																																
SU																																
	RILIS REs	RILIS In	RILIS In	RILIS Dy		RILIS Cd	RILIS Hg		111Cd	Noblegases		RILIS : In		RILIS : Ac	RILIS Hg	RILIS Dy	RILIS : Ca	RILIS : Ca	RILIS : Mg		RILIS : Zn	RILIS : Zn	RILIS : Mn	111Cd	RILIS : Ca	RILIS : Pb		RILIS : Sb/In	RILIS : In	RILIS : Be	RILIS : Ag	Ra/Rn coll.

 =

HRS schedule 2023																														
WK	April		May		June		July		August		September		October		November															
	15	16	17	18	19	20	21	22	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46
MO																														
TU																														
WE																														
TH																														
FR																														
SA																														
SU																														



= Yield measurements, proton scans, setup



23 : U, Np, Pu, Dy, Tm, Pm, Er, Gd, Yb, In, Cd, Hg, Al, Cr, Ac, Ca, Mg, Zn, Mn, Pb, Sb, Be, Ag

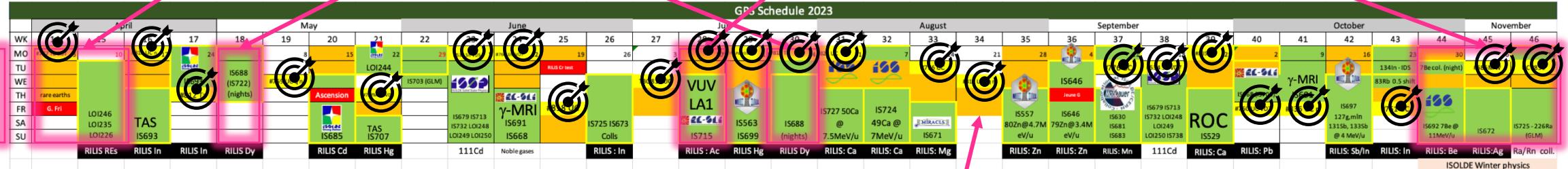
TISD 2023

Back-of-line heating:

Dy collections

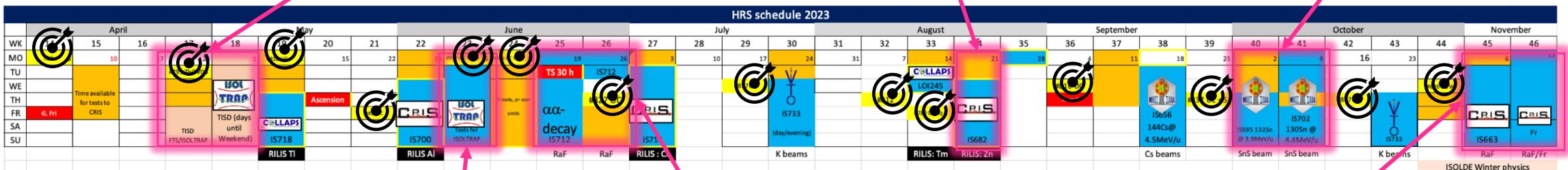
Batch mode: Ra → Ac

LIST: lanthanides



LIST: actinides

ThC_x VD5+CF₄



Prototype target +
prototype ion source

Molecular
beams:
RaF



=

Yield measurements, proton scans, setup

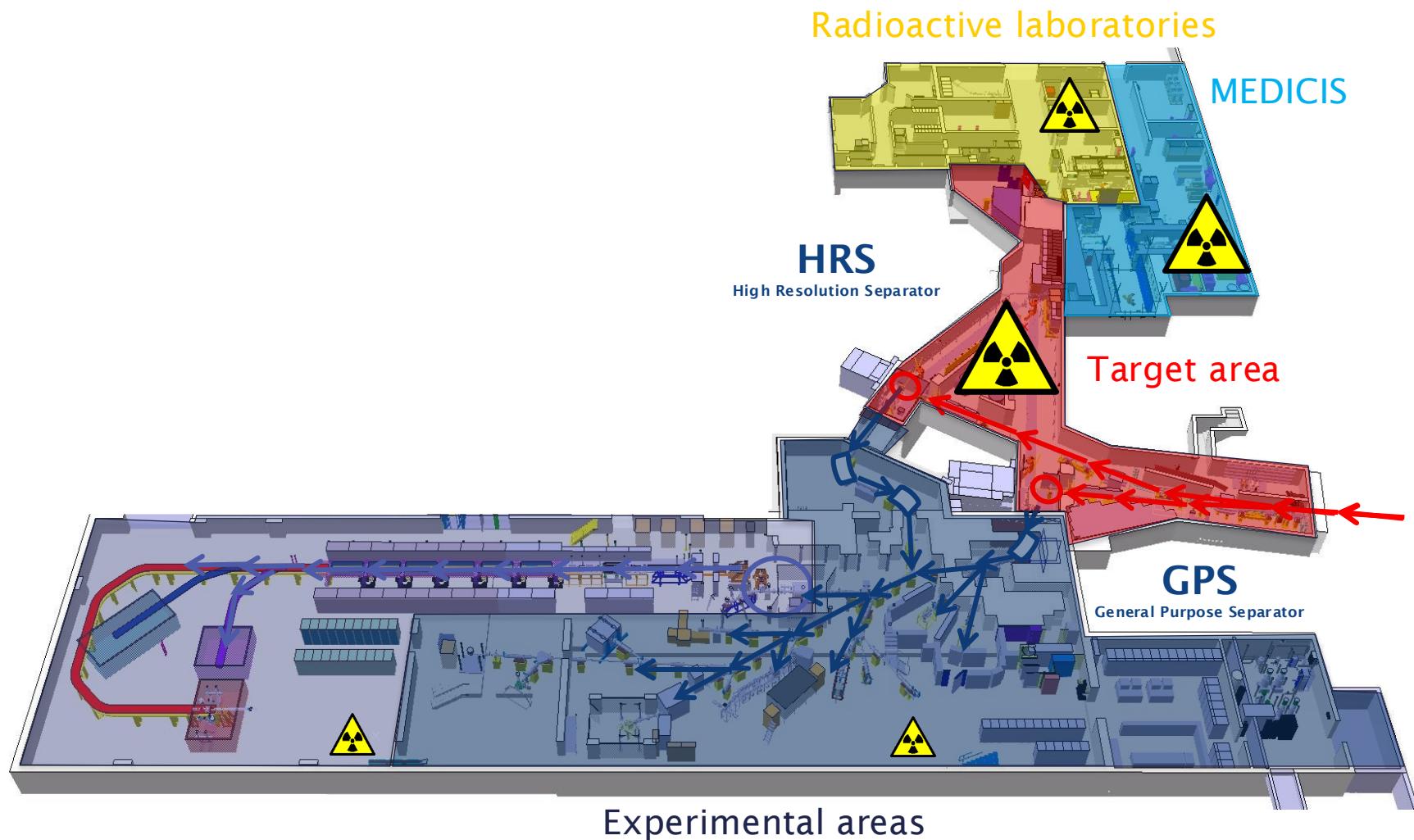
23 : U, Np, Pu, Dy, Tm, Pm, Er, Gd, Yb, In, Cd, Hg, Al, Cr, Ac, Ca, Mg, Zn, Mn, Pb, Sb, Be, Ag



ISOL

“On-Line”:

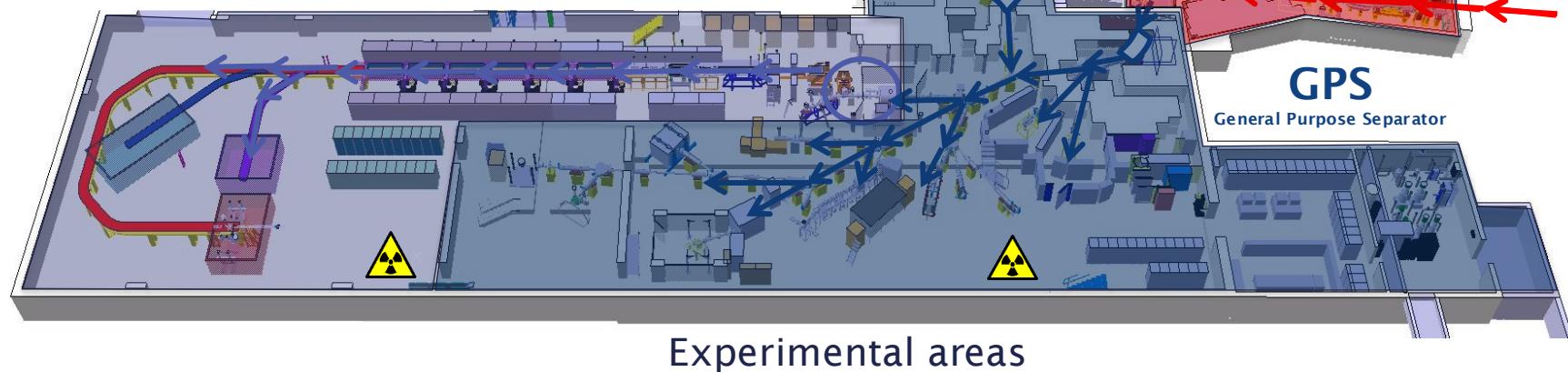
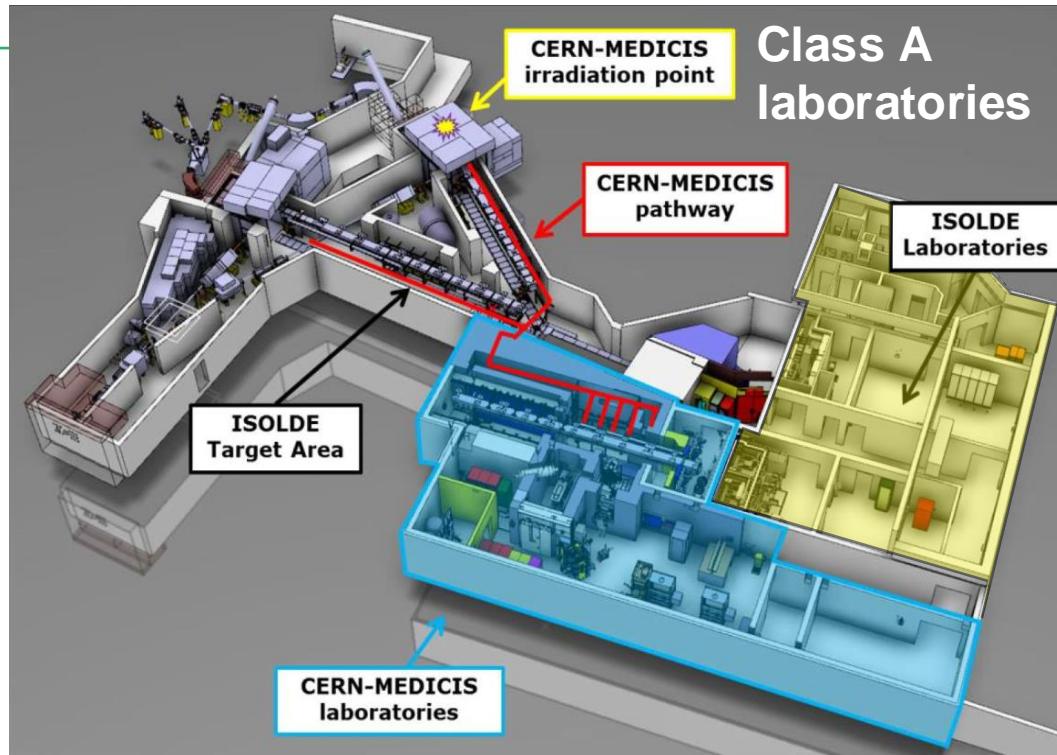
- Production
- Release
- Ionization
- Extraction



ISOL

“On-Line”:

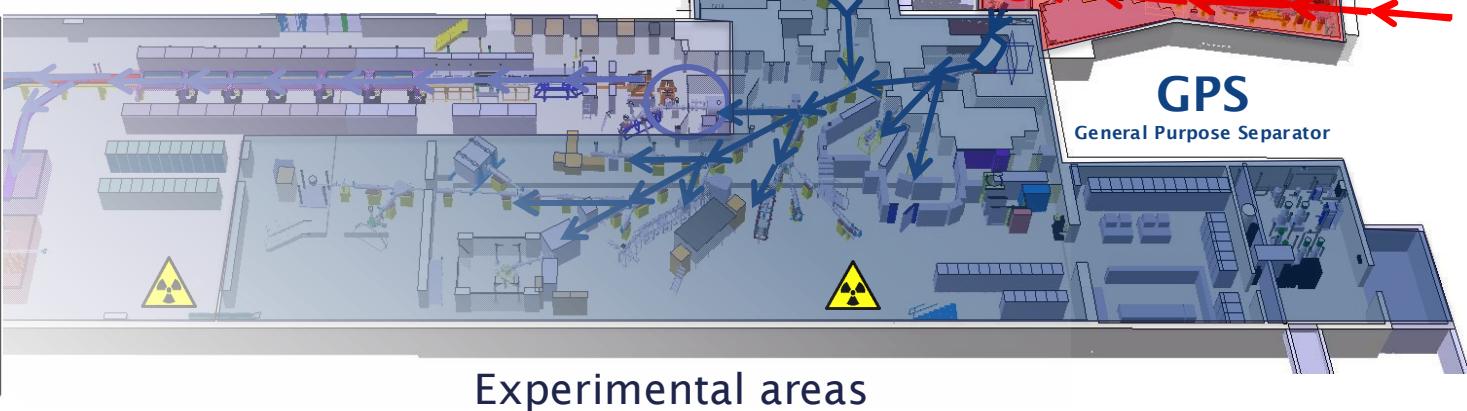
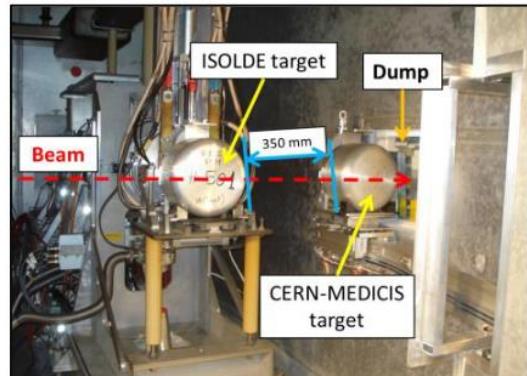
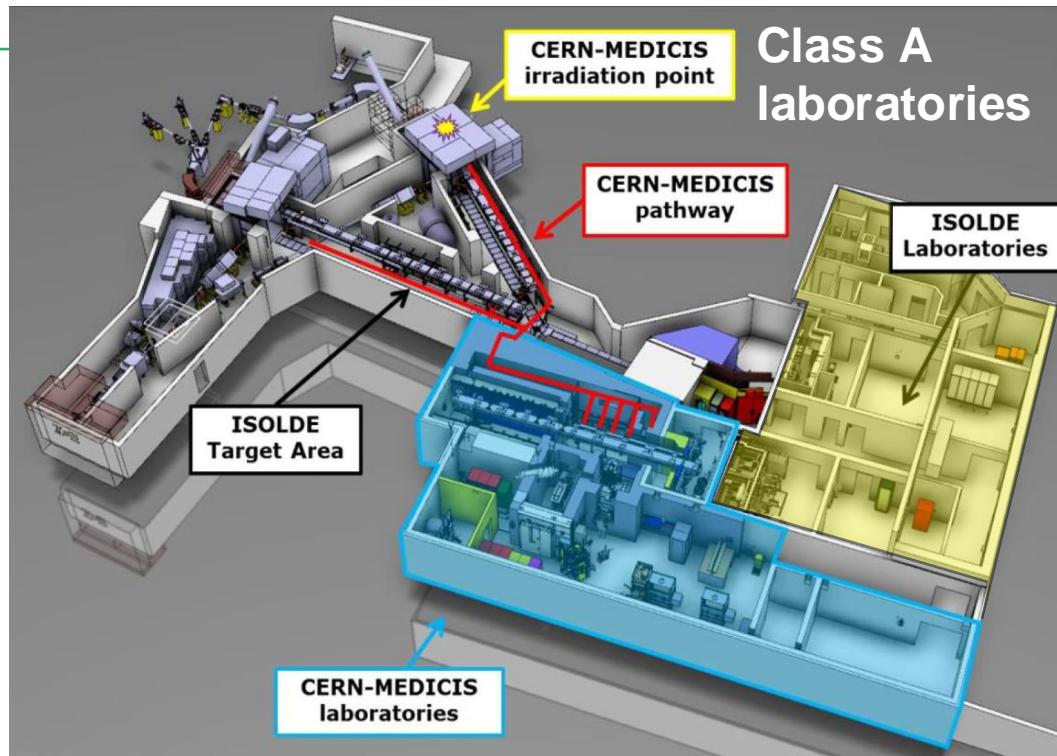
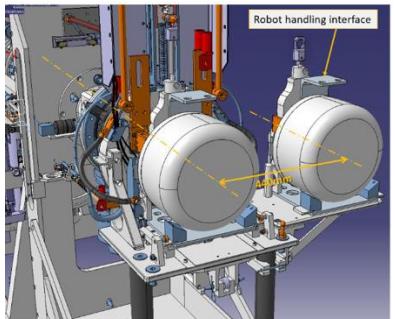
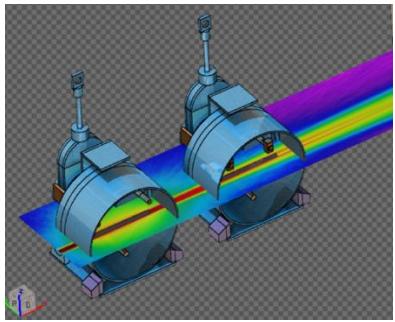
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ISOL

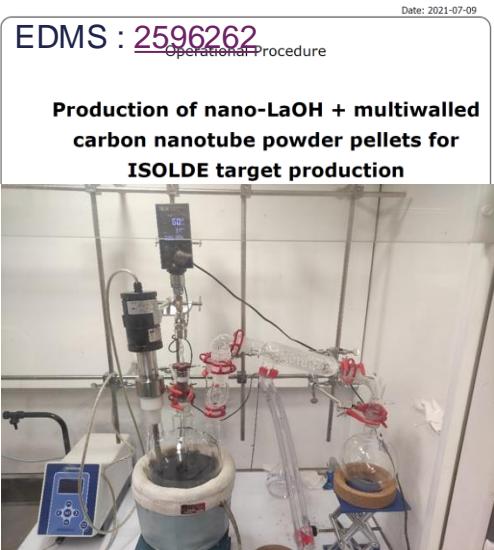
“On-Line”:

- Production
- Release
- Ionization
- Extraction



Non-actinide development and characterization lab

Planetary ball mill – Powder particle size reduction



Reactor setup

Photos courtesy of V. Berlin, E. Reis, L. Lambert, S. Rothe

Laser diffraction particle size analyzer



Gas sorption – Pore size distribution (BET)



Carburization pumpstand
Target development,
sintering studies

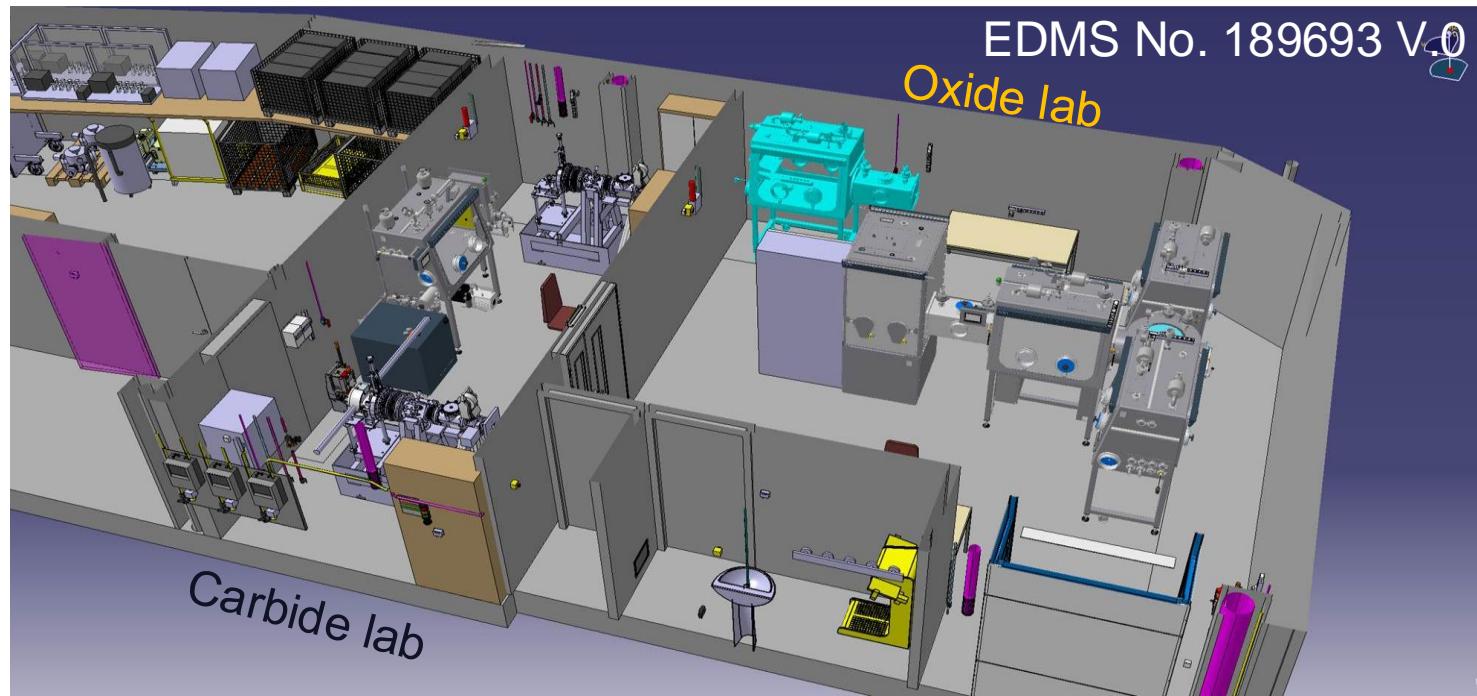


Gas pycnometry
Apparent density
determination



TGA-MS – Reaction kinetics

The Nanolab: Production and Research



5 Glove boxes

- 4 connected in T shape: non-pyrophoric
- 1 inert atmosphere: carbides

Production alternating with development



Storage capsules

Carbide lab



Oxide lab



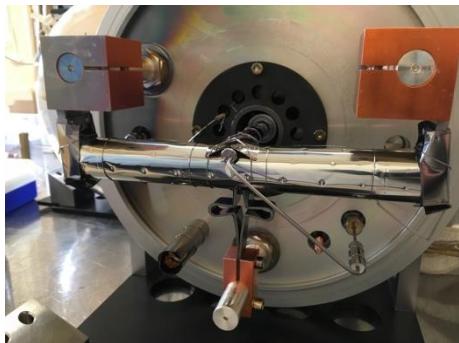
Photos courtesy of L. Lambert



Material developments

Gas injection

- Reactive/corrosive gases



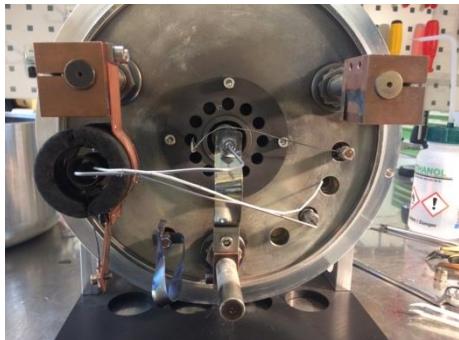
Reactants

Mass markers

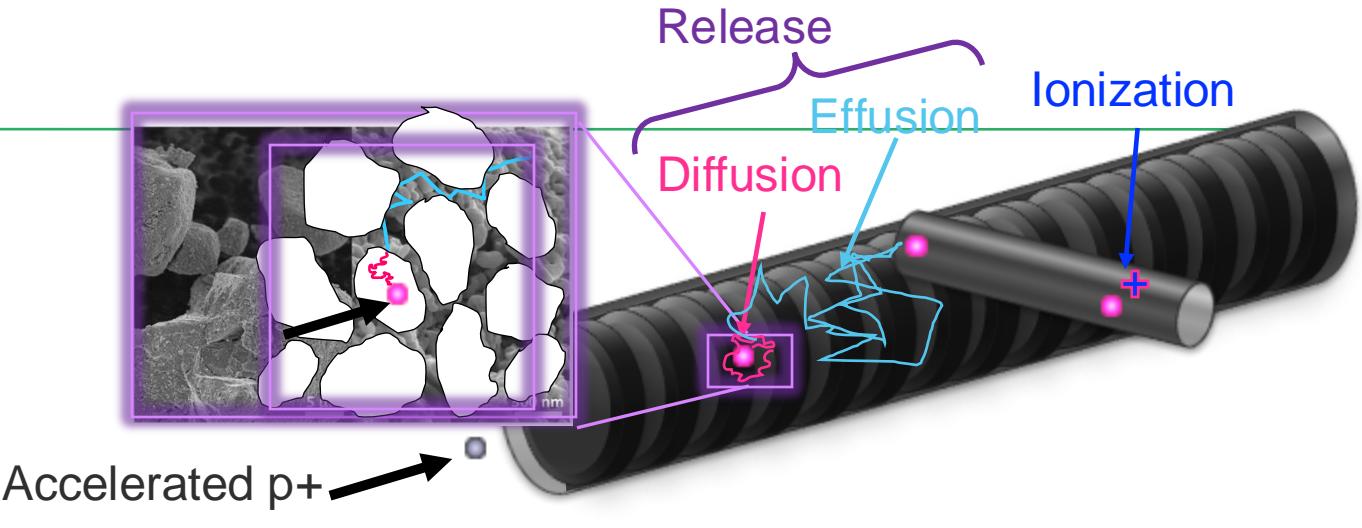
Target materials

Particle size

Open porosity



Adapted from:
J.P. Ramos. EMIS XIII, CERN, Geneva, 2018.



$$\text{Beam Intensity} = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms

j – Proton flux [cm^{-2}]

σ – Cross section [mb]

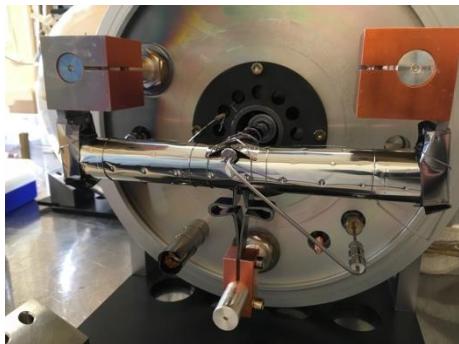
ε – Efficiency [%]

$$\varepsilon = \varepsilon_{\text{diff}} \varepsilon_{\text{eff}} \varepsilon_{\text{is}} \varepsilon_{\text{ext}} \varepsilon_{\text{sep}} \varepsilon_{\text{trans}}$$

Material developments

Gas injection

- Reactive/corrosive gases



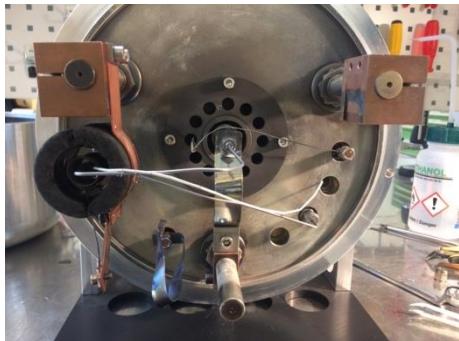
Reactants

Mass markers

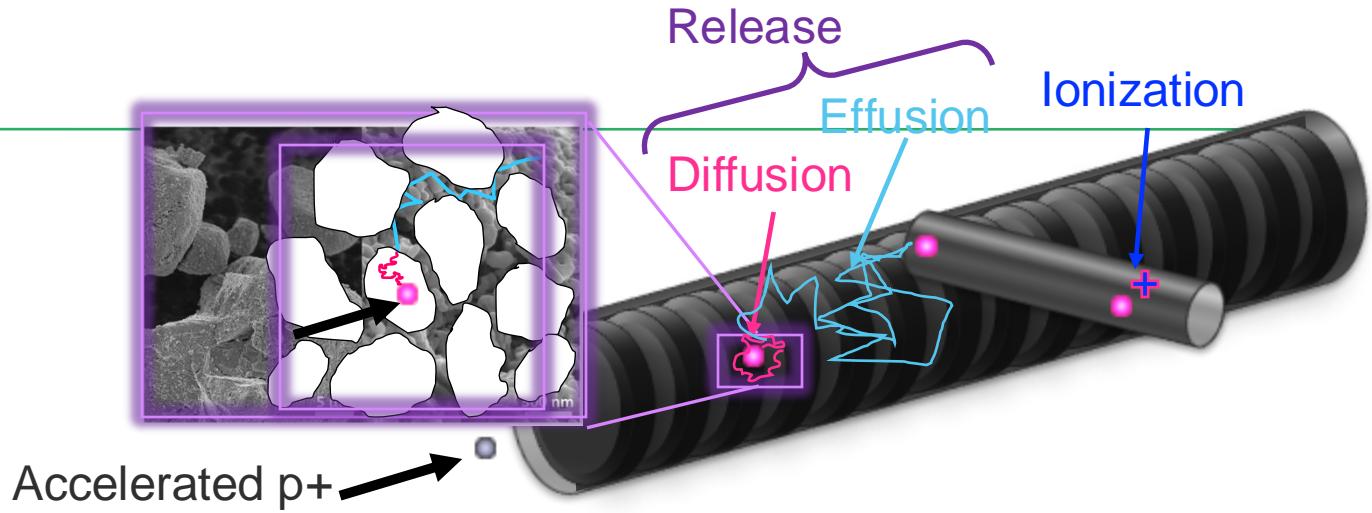
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$$\text{Beam Intensity} = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms

j – Proton flux [cm^{-2}]

σ – Cross section [mb]

ε – Efficiency [%]

μ – diffusion delay parameter

G – grain size

$$\varepsilon = \varepsilon_{\text{diff}} \varepsilon_{\text{eff}} \varepsilon_{\text{is}} \varepsilon_{\text{ext}} \varepsilon_{\text{sep}} \varepsilon_{\text{trans}}$$

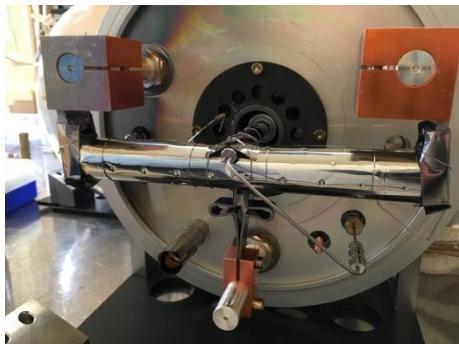
$$\varepsilon_{\text{diff}} \propto \sqrt{\mu \cdot T_{1/2}} \propto \frac{1}{G}$$

$$\mu = \frac{\pi^2 D}{G^2}$$

Material developments

Gas injection

- Reactive/corrosive gases



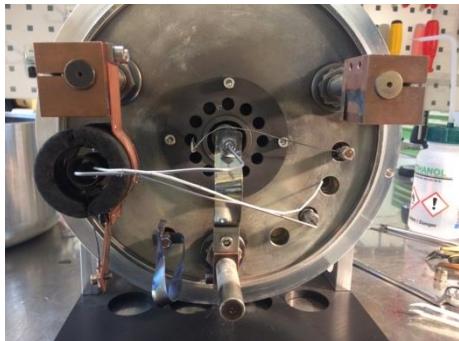
Reactants

Mass markers

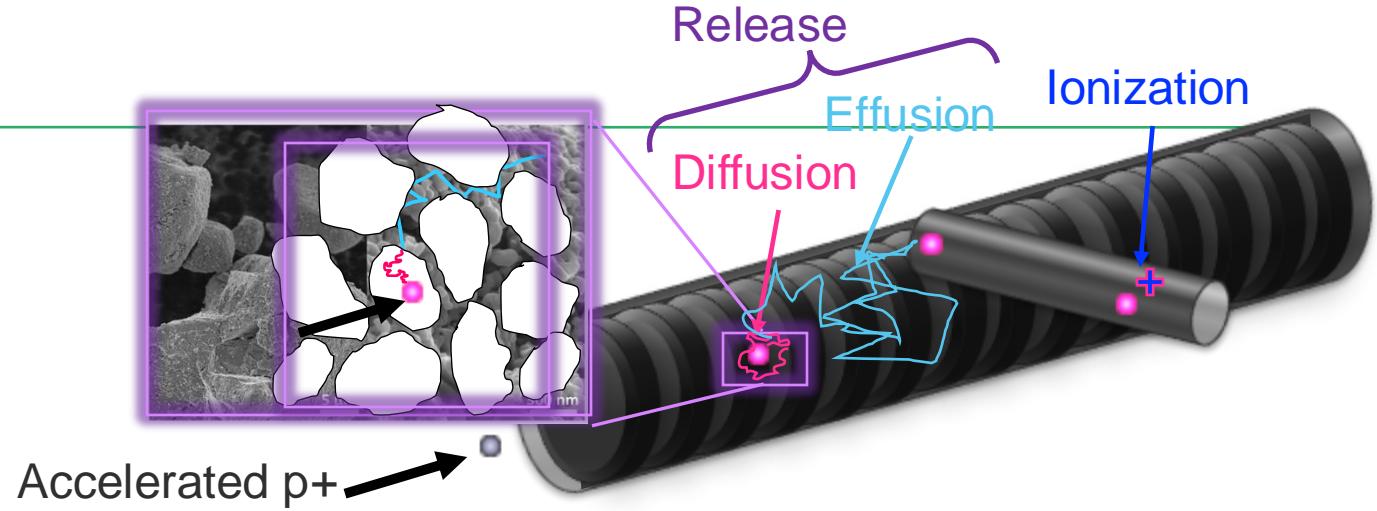
Target materials

Particle size

Open porosity



Adapted from:
J.P. Ramos. EMIS XIII, CERN, Geneva, 2018.



$$\text{Beam Intensity} = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms

j – Proton flux [cm^{-2}]

σ – Cross section [mb]

ε – Efficiency [%]

μ – diffusion delay parameter

G – grain size

$$\varepsilon = \varepsilon_{\text{diff}} \varepsilon_{\text{eff}} \varepsilon_{\text{is}} \varepsilon_{\text{ext}} \varepsilon_{\text{sep}} \varepsilon_{\text{trans}}$$

$$\varepsilon_{\text{diff}} \propto \sqrt{\mu \cdot T_{1/2}} \propto \frac{1}{G}$$

$$\mu = \frac{\pi^2 D}{G^2}$$

Small G , high T \rightarrow Increased $\varepsilon_{\text{diff}}$

Increased $\varepsilon_{\text{diff}}$ \leftrightarrow Increased sintering and grain growth

Ion source developments

Molecular breakup and characterization studies

- FEBIAD-type ion sources [1,2]
- Electron energy and source optimization
- Ion source systematics

Photocathode ion sources [3]

- Cold (room-temperature) environments

In-source spectroscopy [4]

- PI-LIST: sub-Doppler hot-cavity in-source spectroscopy
- CERN-ISOLDE implementation

[1] Maldonado (2023) PhD thesis

[2] Martinez Palenzuela (2020) PhD thesis

[3] Ballof . et al., 2022) *J. Phys.: Conf. Ser.* **2244** 012072

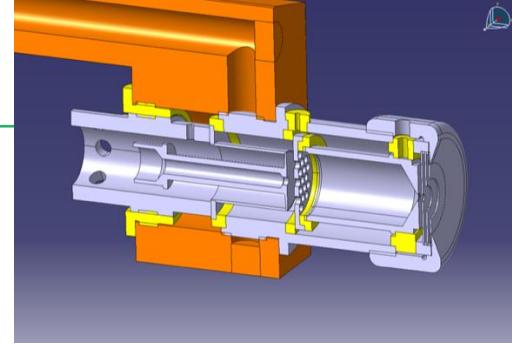
[4] Heinke et al. (2023) *NIM B.* **541** (8-12)



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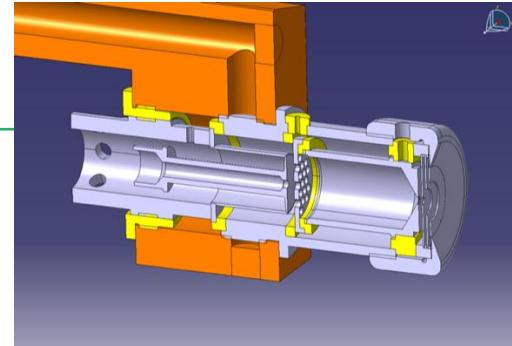
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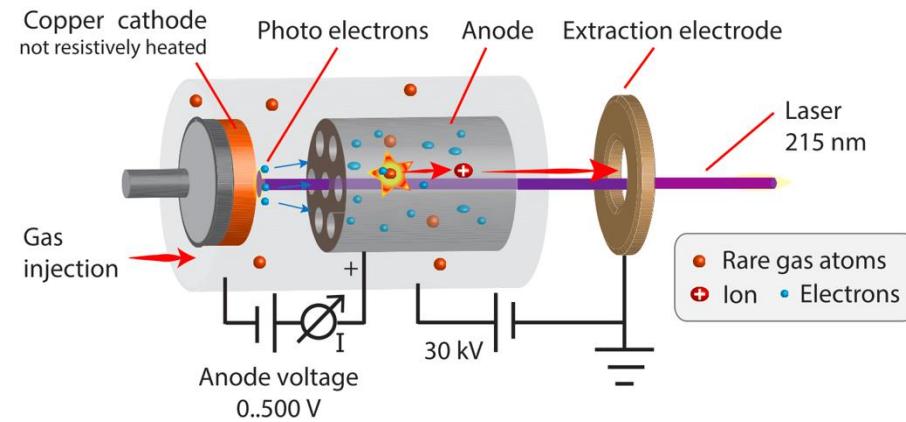
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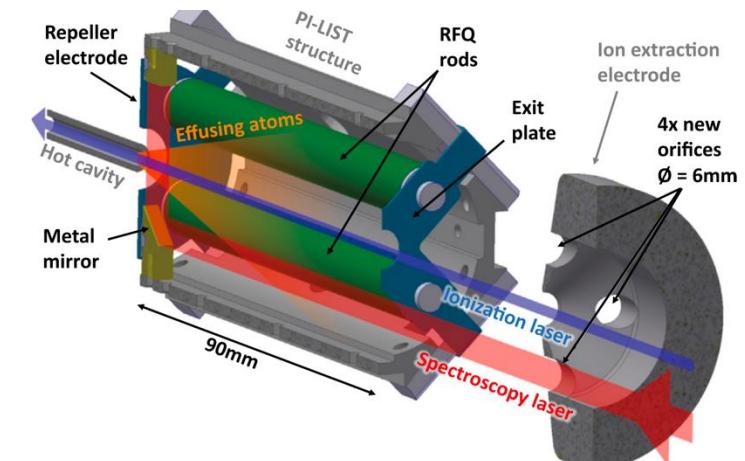
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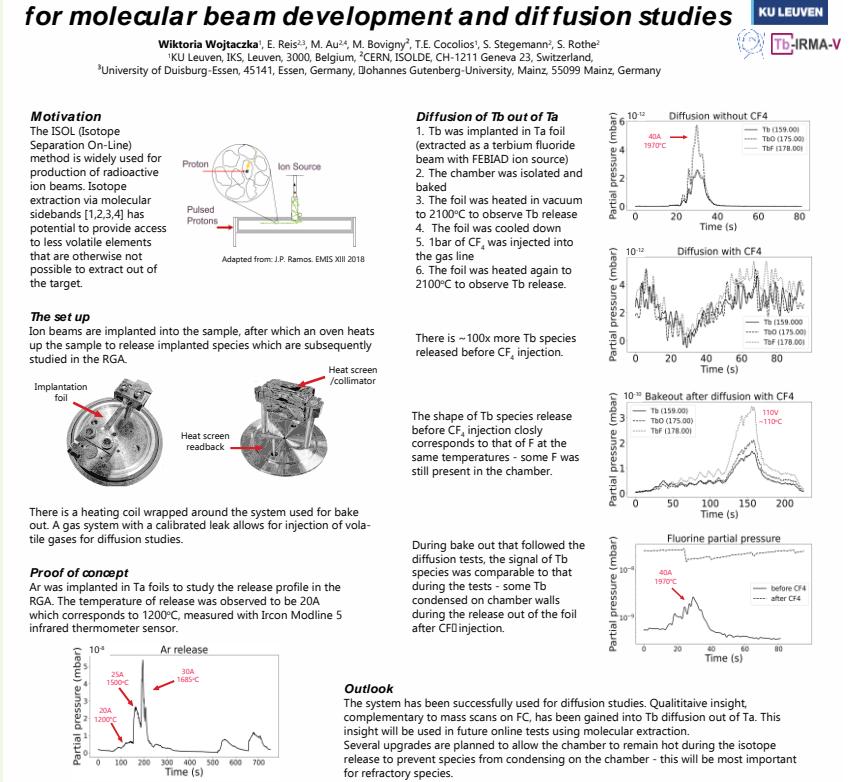
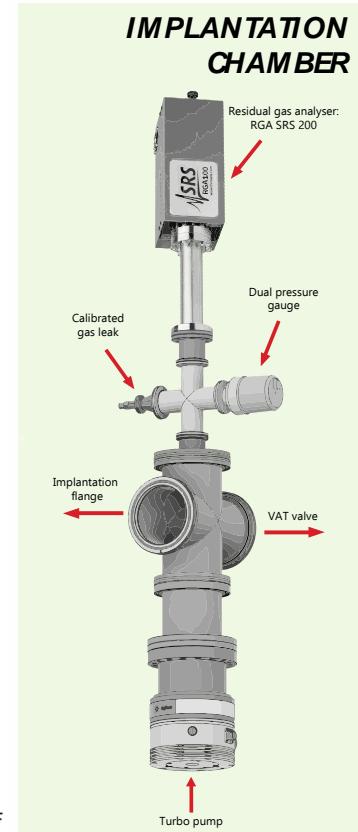
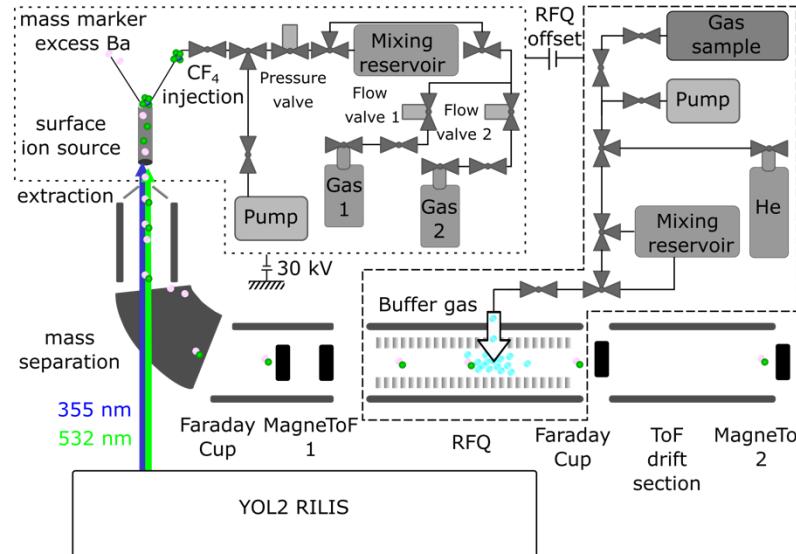
Offline upgrades for molecular beam development

Detection, implantation, ion counting

RILIS for molecules

Gas injection and mixing

- Remote control of HV gas systems and partial pressures



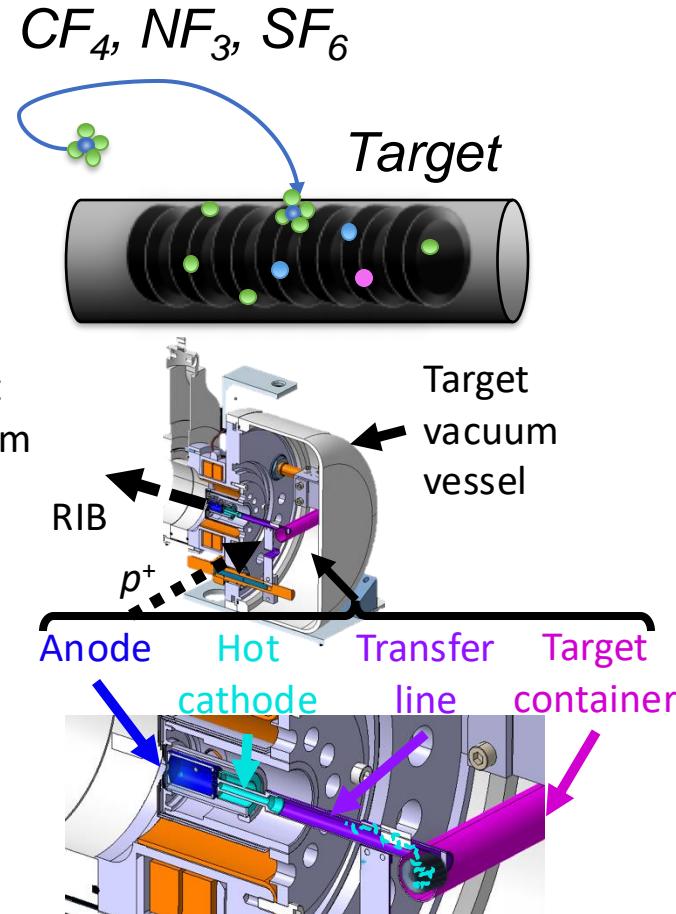
[1] Au et al. (2023) *NIM B*. **541** (144-147)

[2] Wojtaczka et al. (2023) *ICIS'23*, Victoria, Canada

Formation: how do we make molecules?

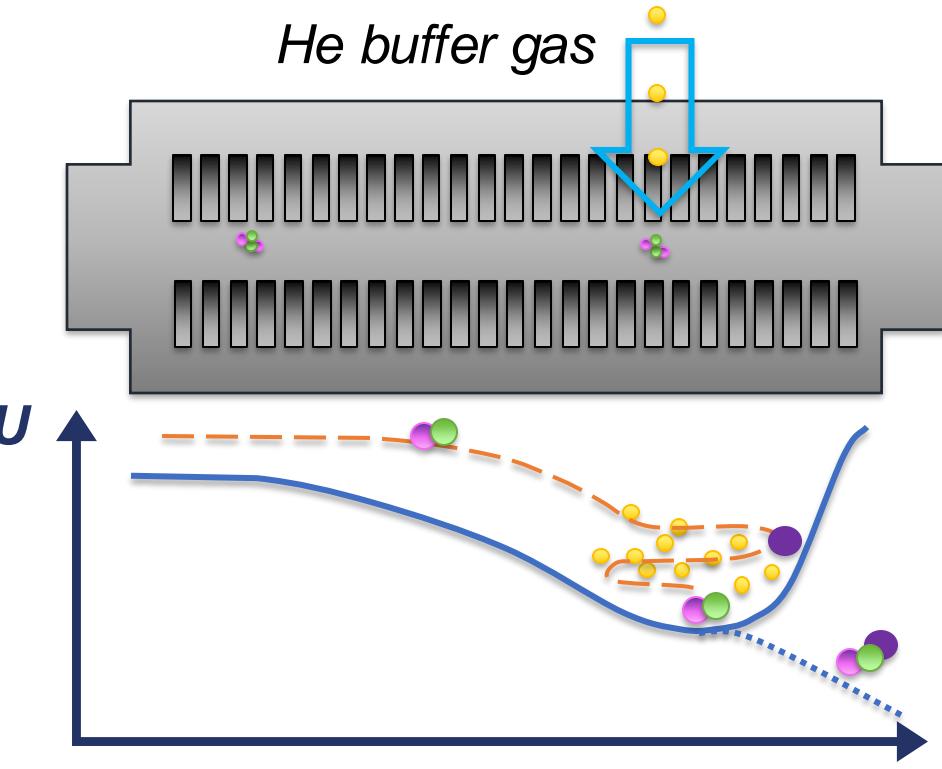
In-source

- Reactive gas



In-trap

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



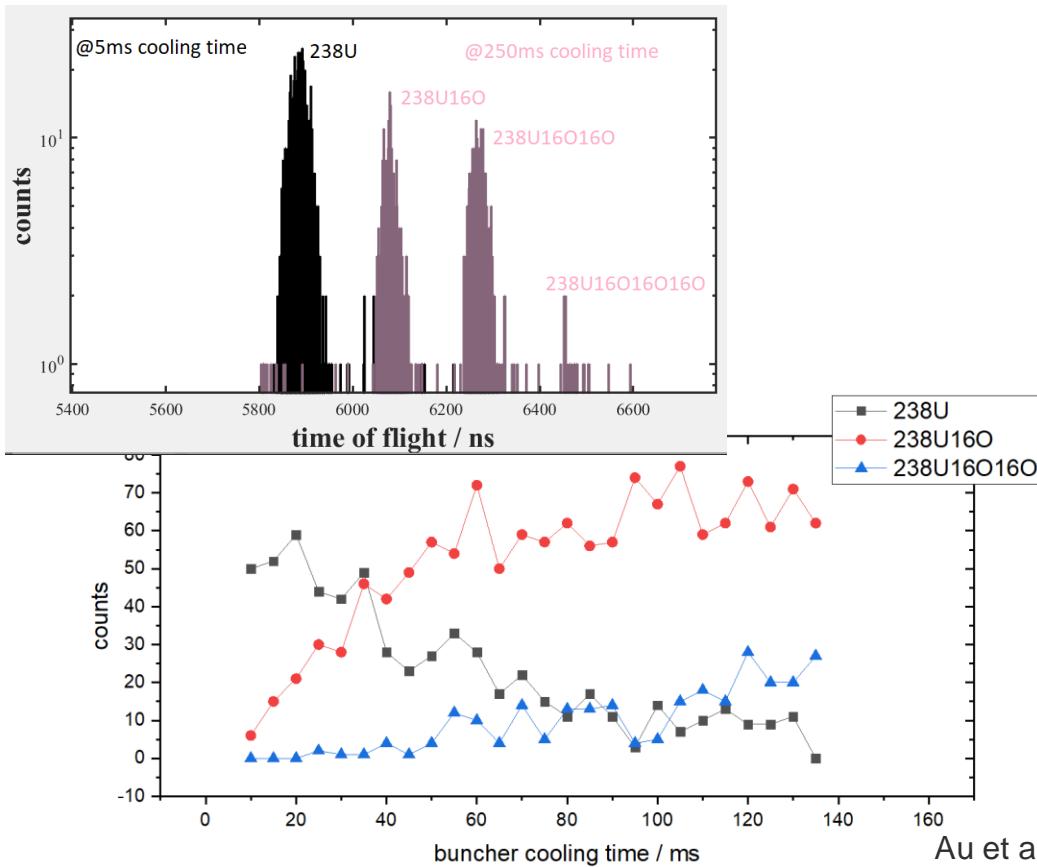
Au et al. (2023) NIM B. 541 (375-379)



In-trap: identification

UO_x , TaO_x

- Residual gas and primary mass-separated beam
- ID by ToF and revs vs ToF



Au et al. (2023) NIM B. 541 (375-379)

