Production of radioactive ion beams at CERN-ISOLDE

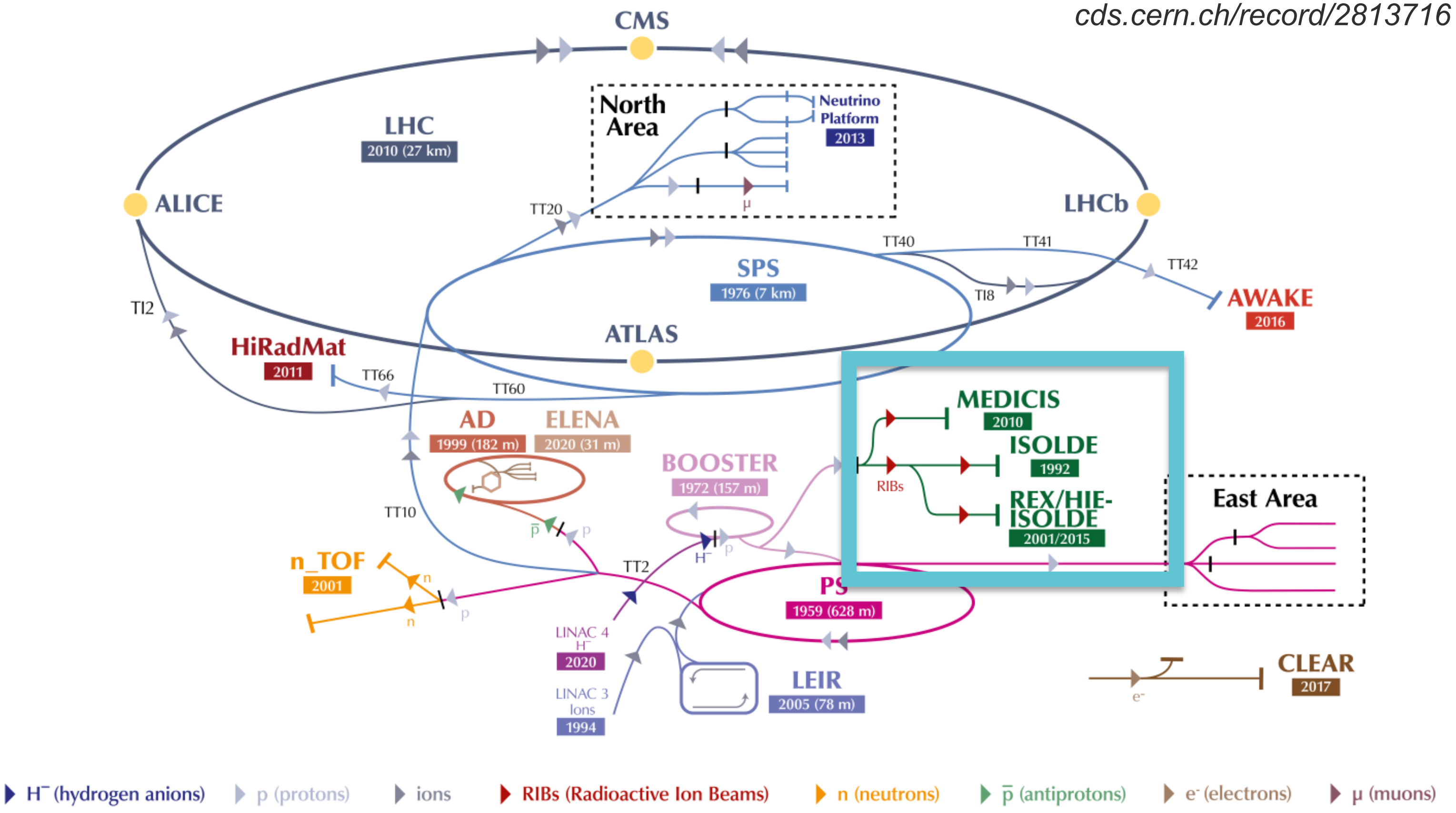
Mia Au

CERN SY-STI

Outline: Radioactive beams at CERN-ISOLDE

1	CERN-ISOLDE and ISOL
2	Radioactive beams
3	Radioactive molecules
4	Offline developments

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



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


Cu
#625
W

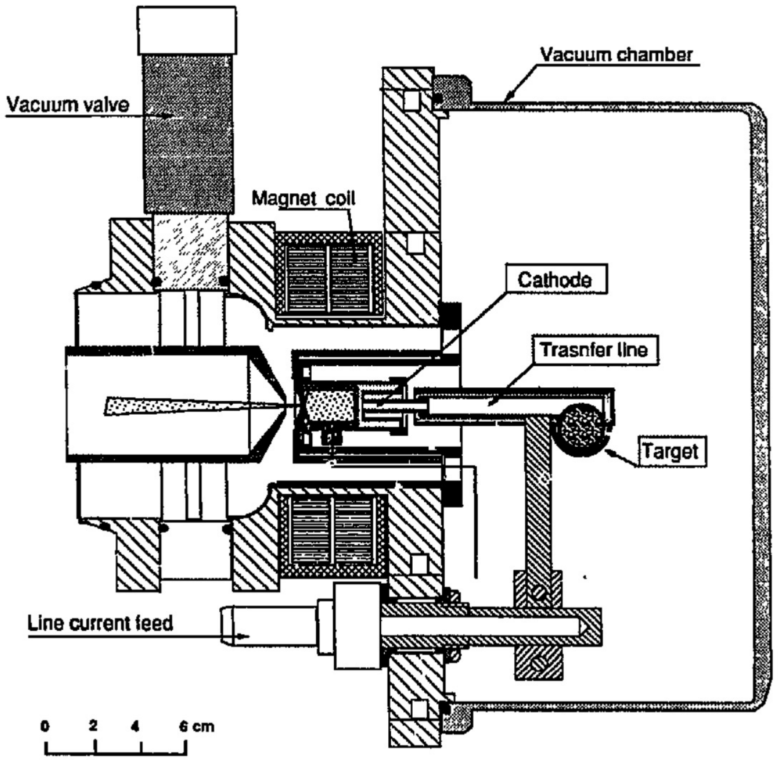


Fig. 1. Target and ion source assembly with plasma ion source MK5. The vacuum valve is part of the assembly.

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The Isotope Separation On-Line (ISOL) method

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

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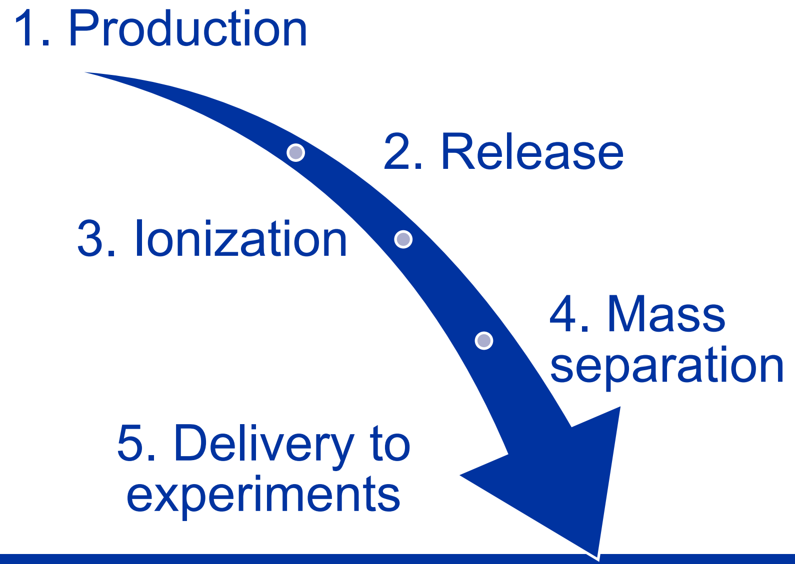

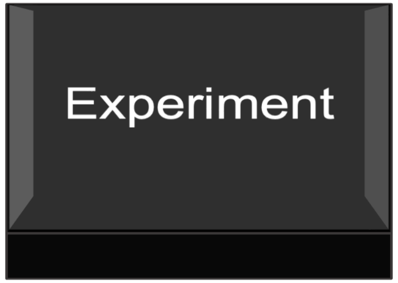

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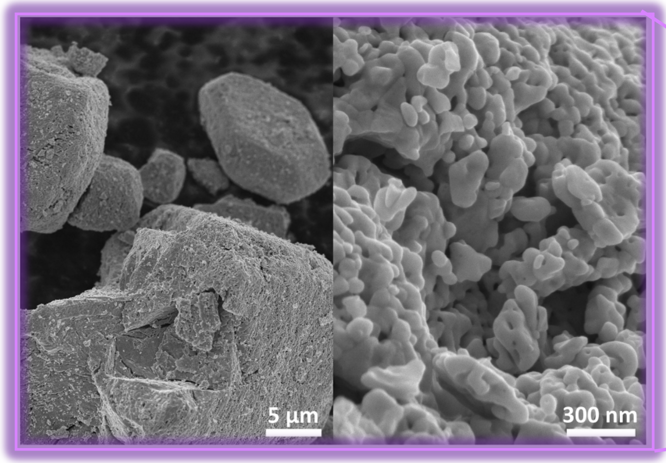

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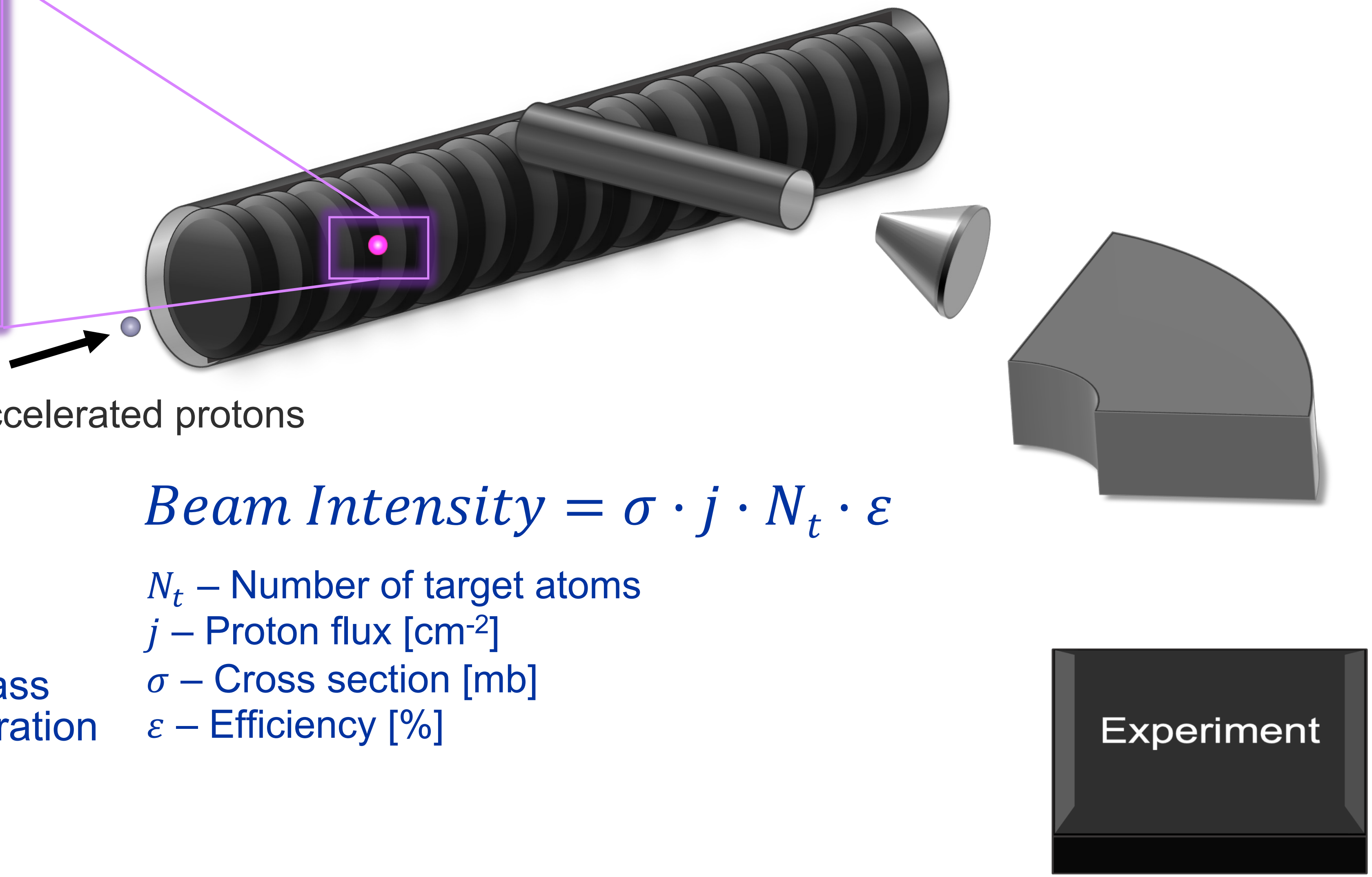
NIM B **463**, 201



1. Production
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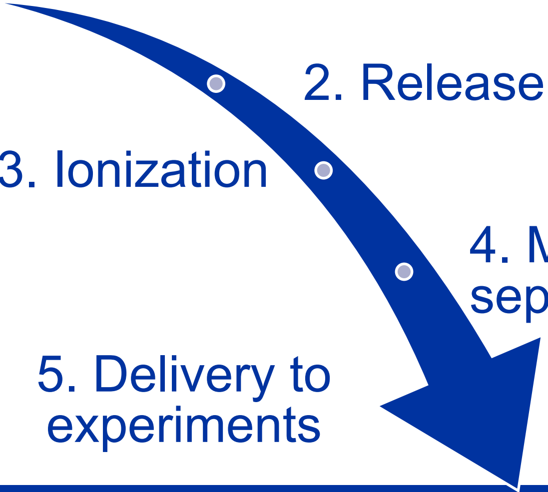

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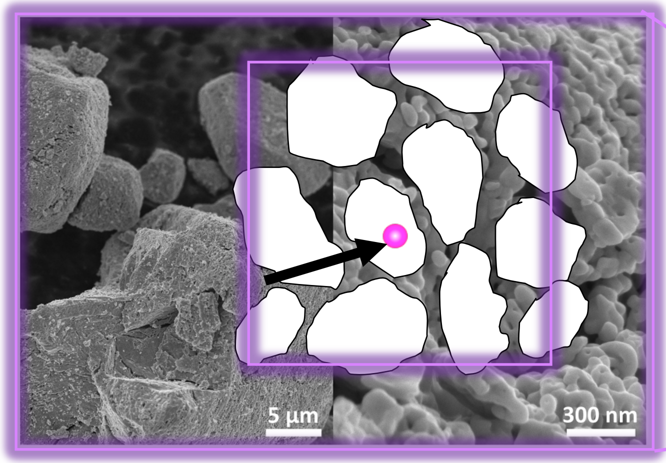

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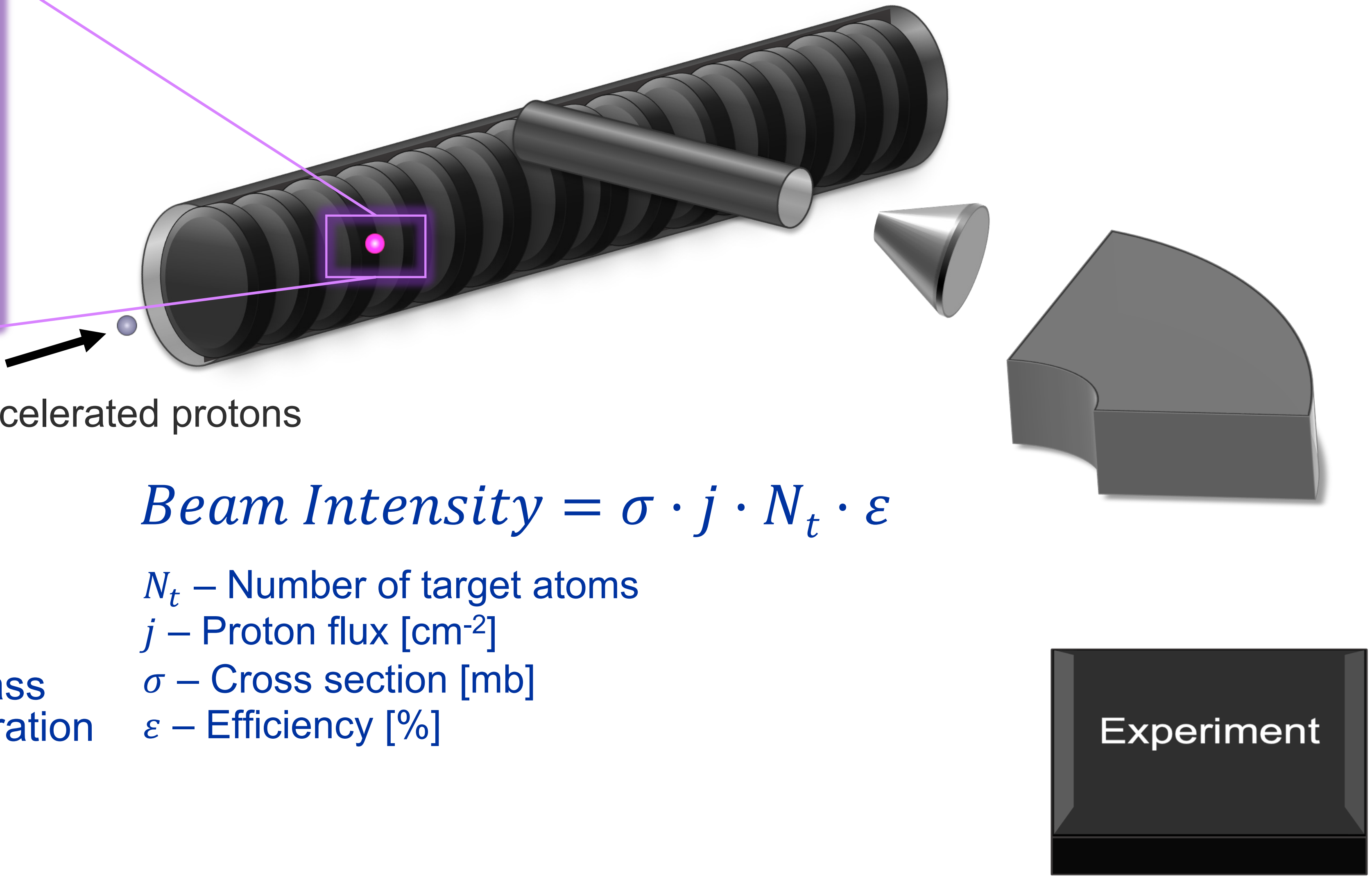

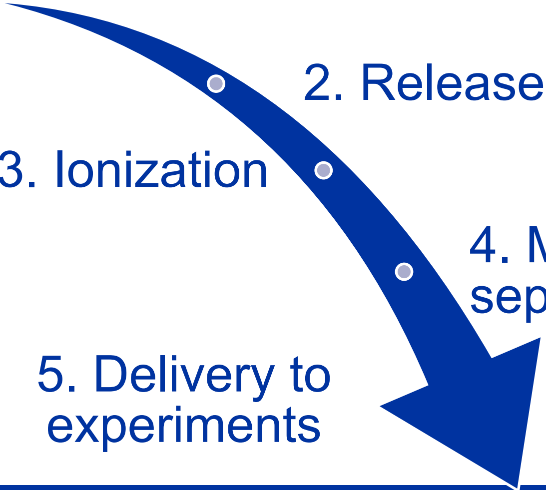

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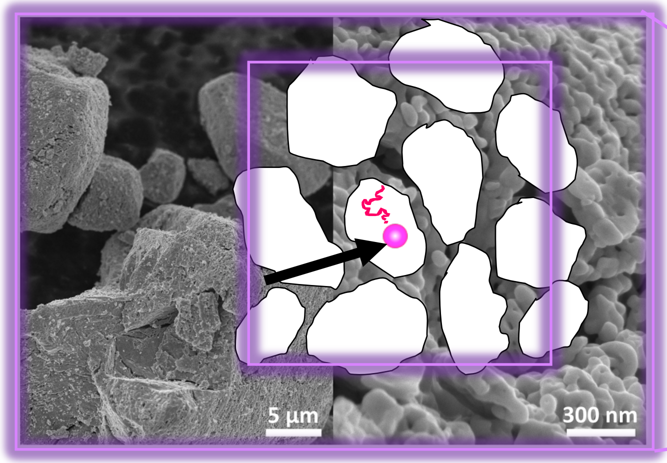

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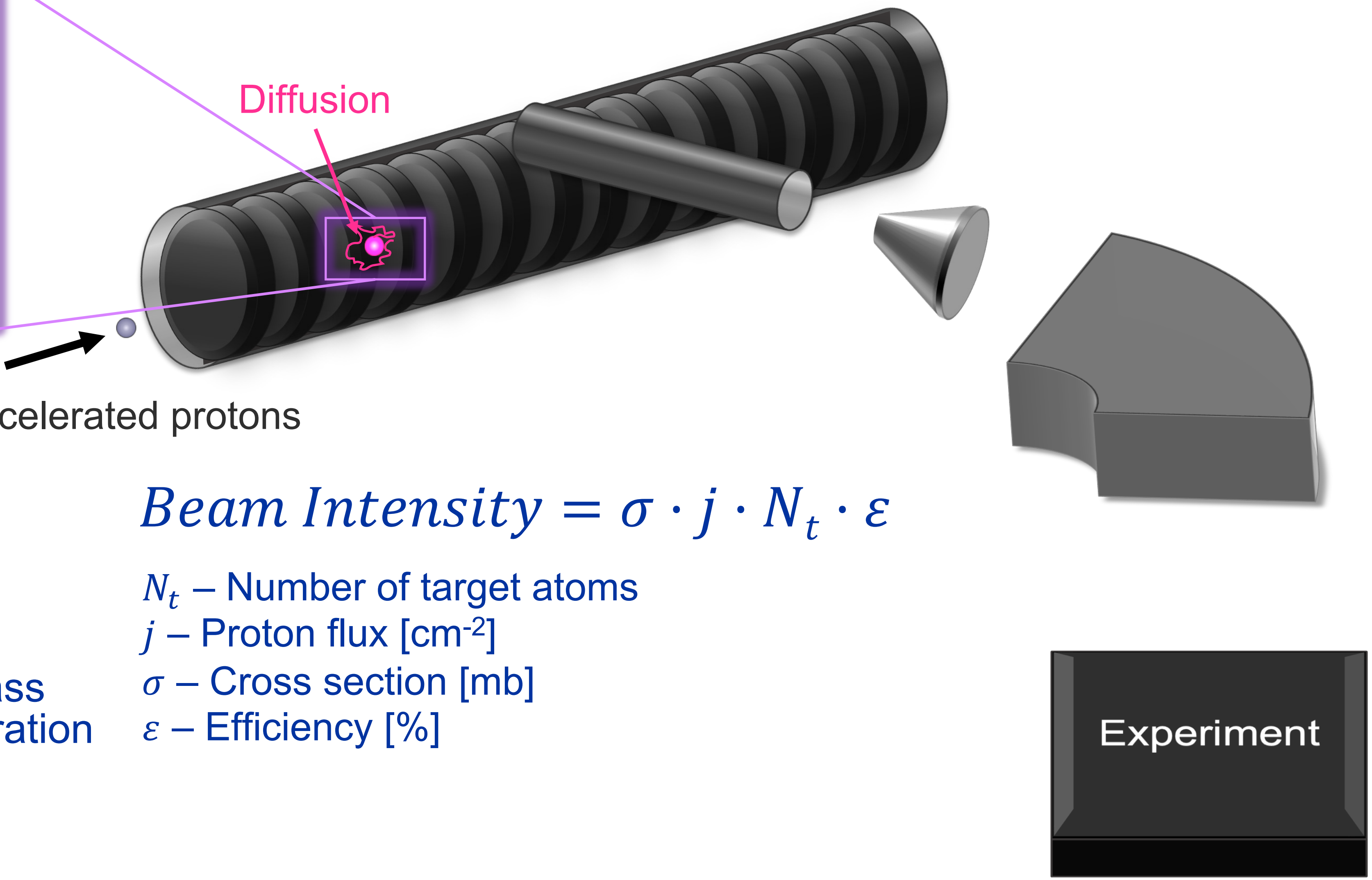

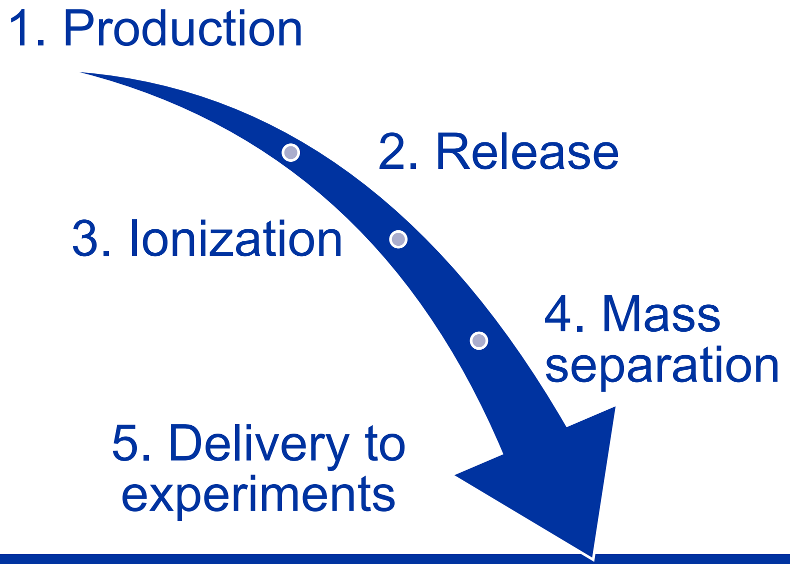

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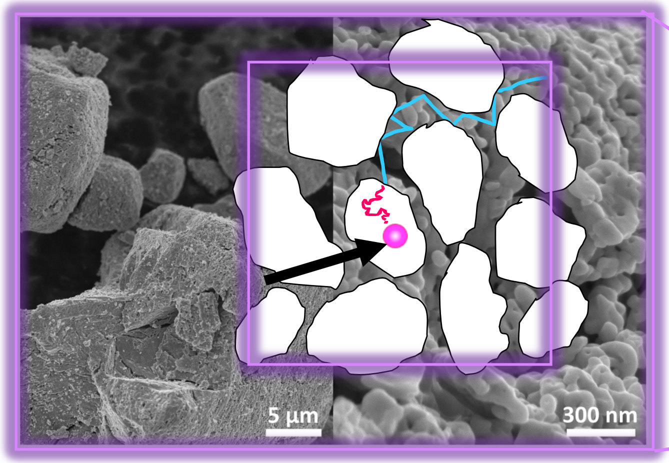

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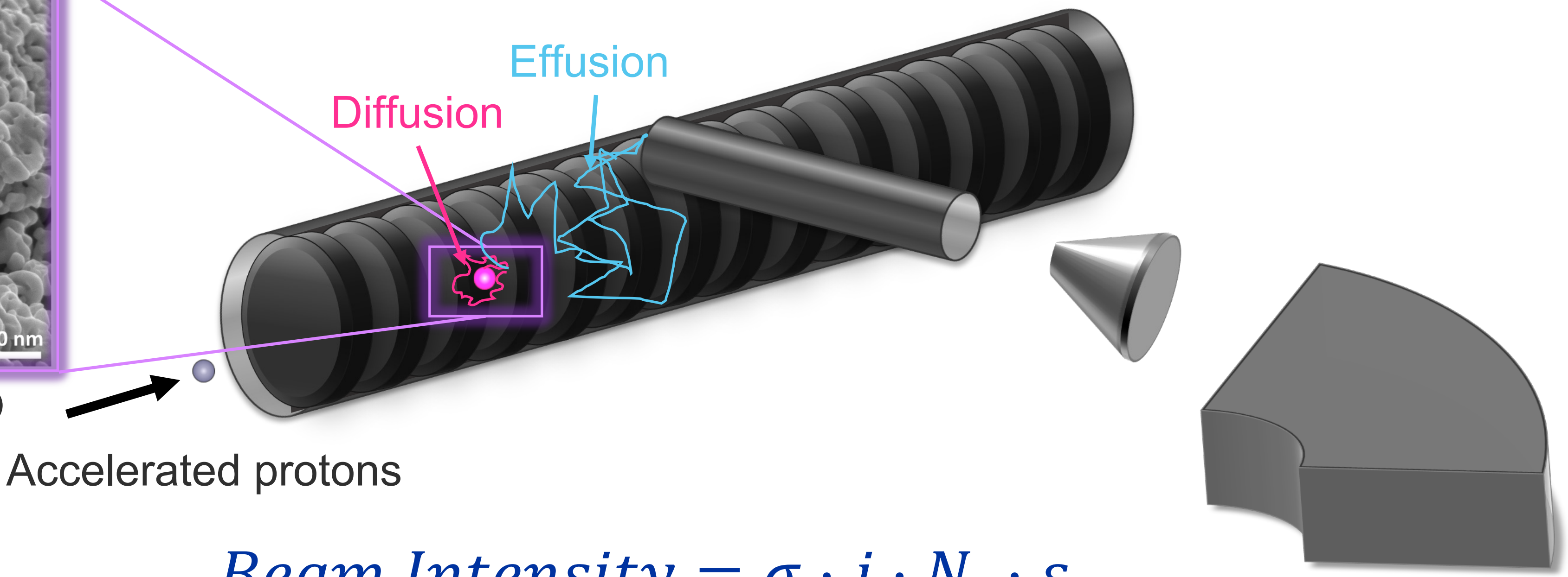

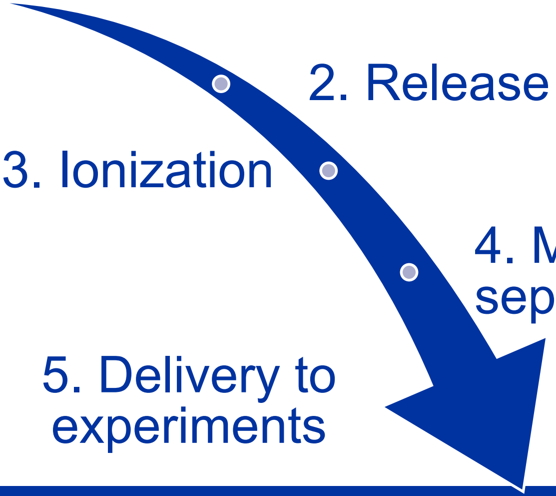

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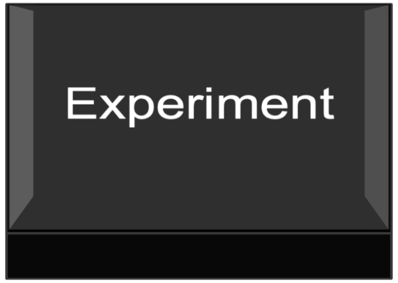

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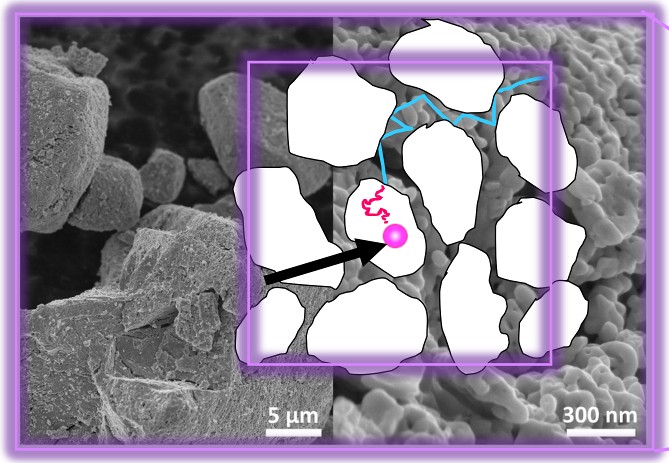

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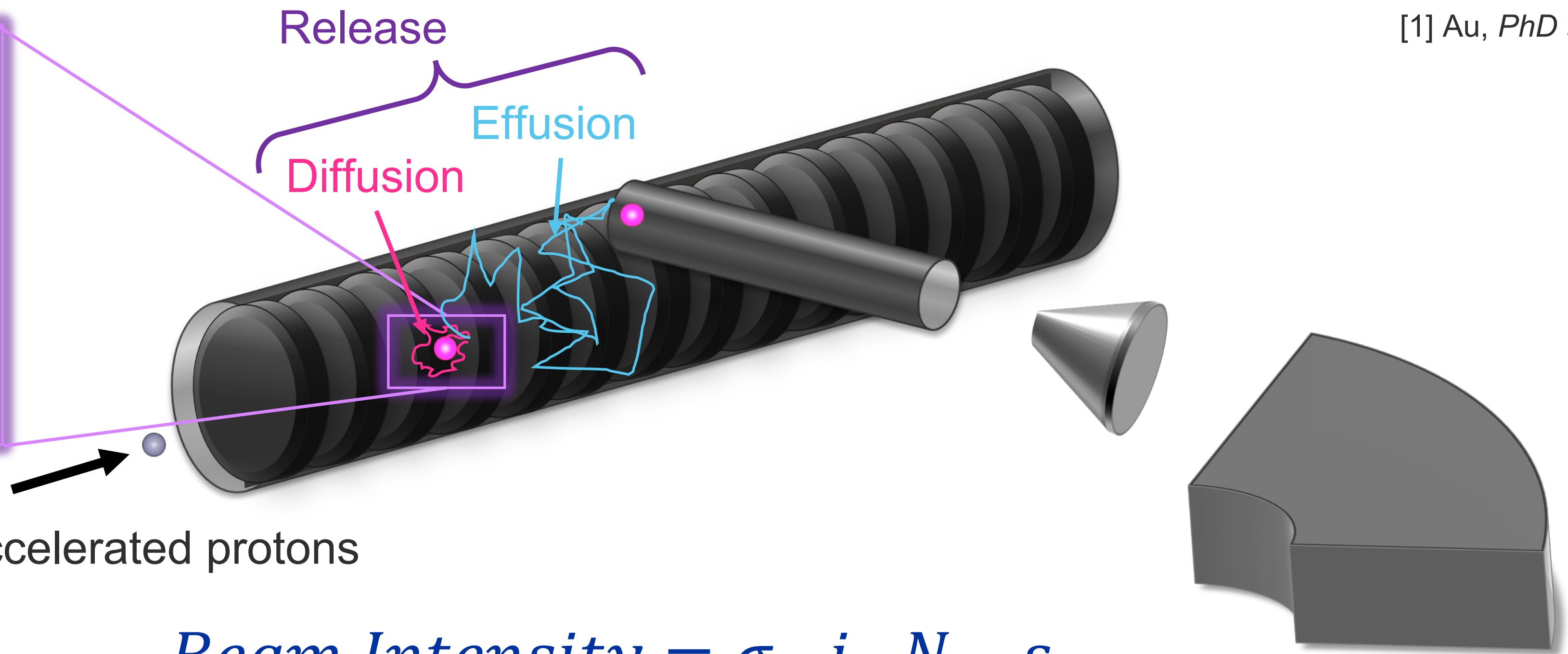

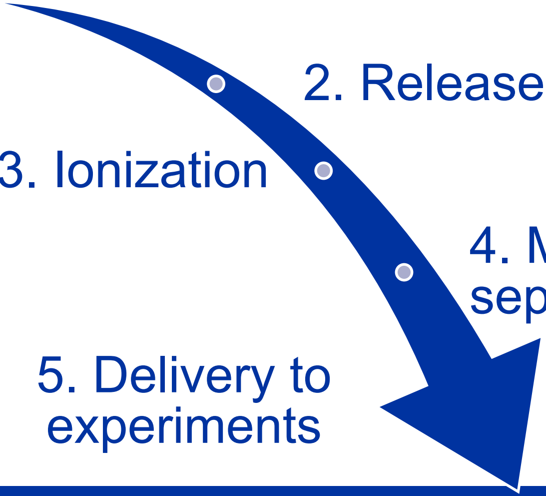

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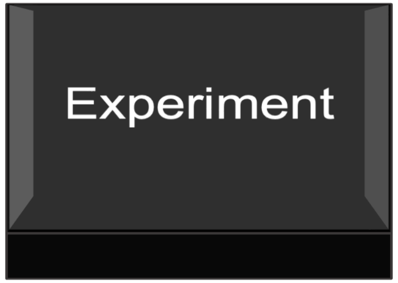

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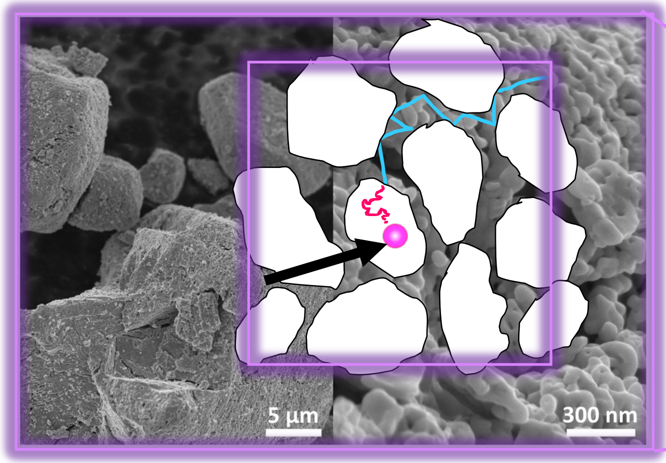

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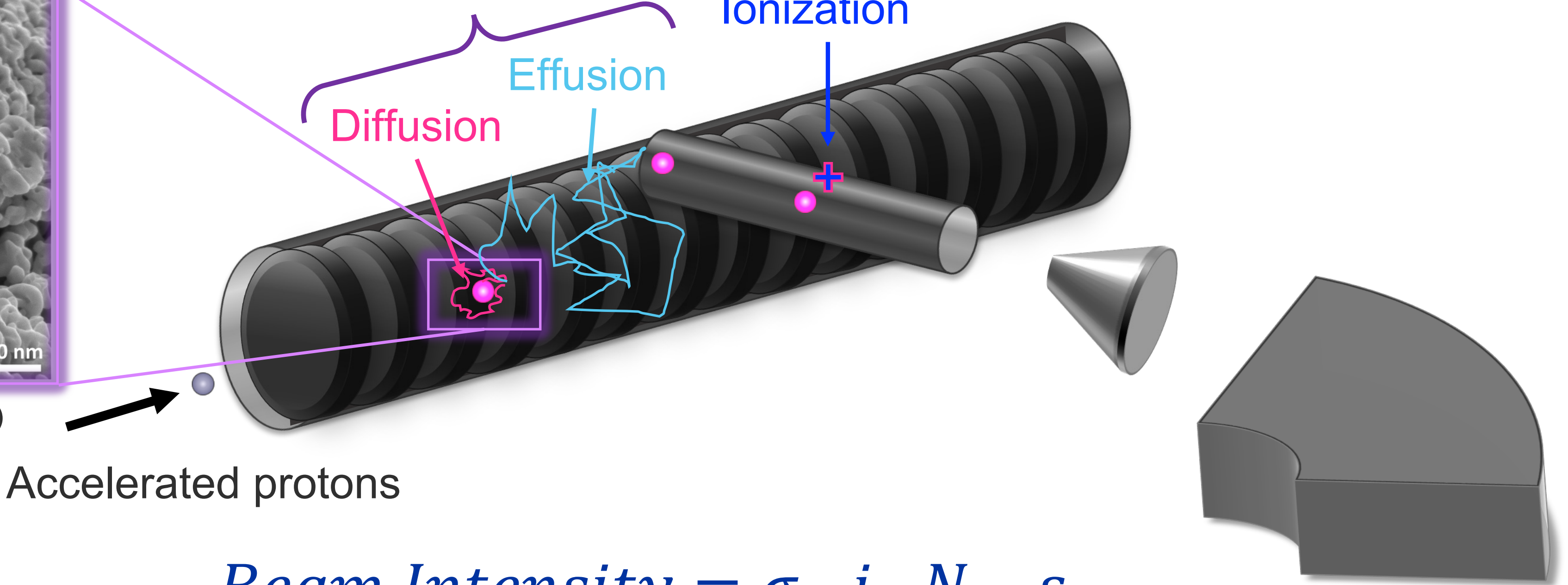

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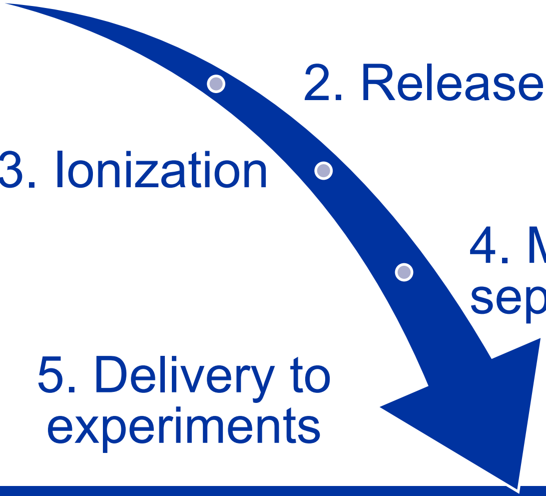

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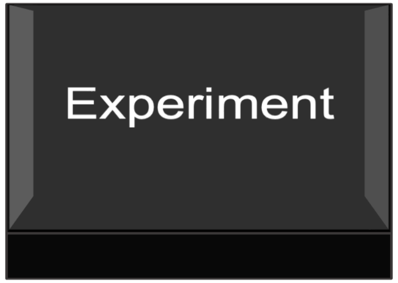

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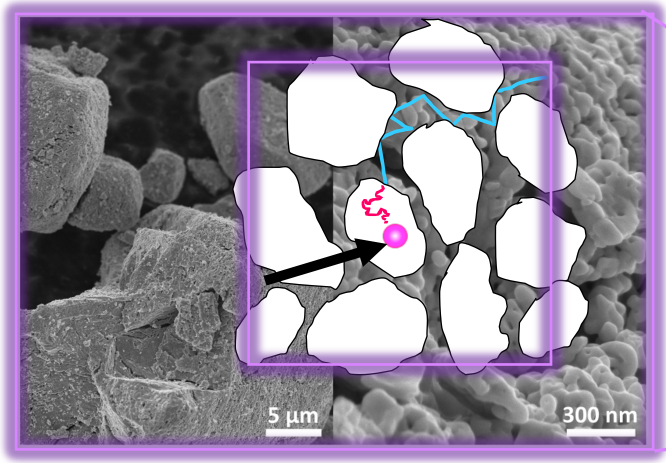

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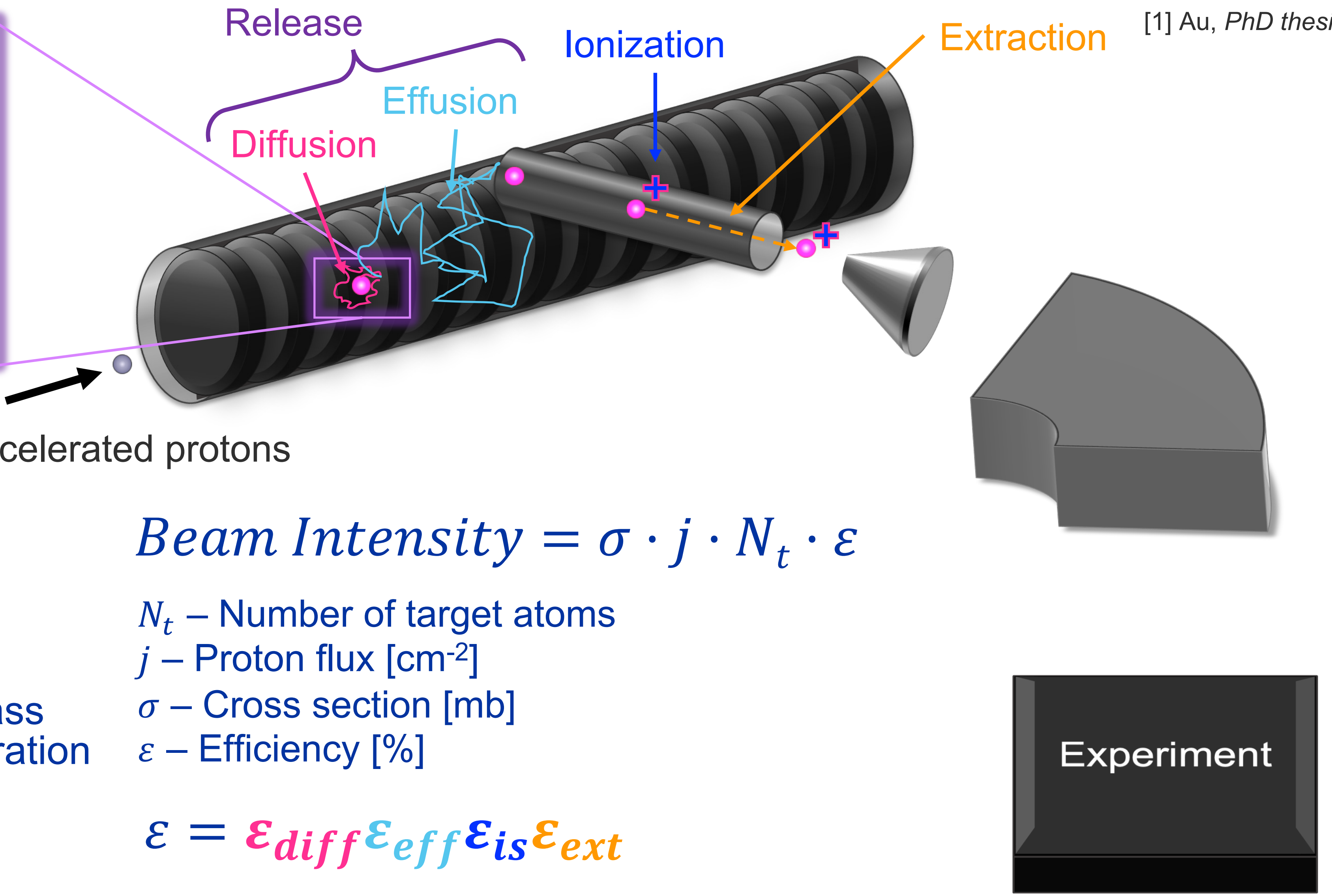

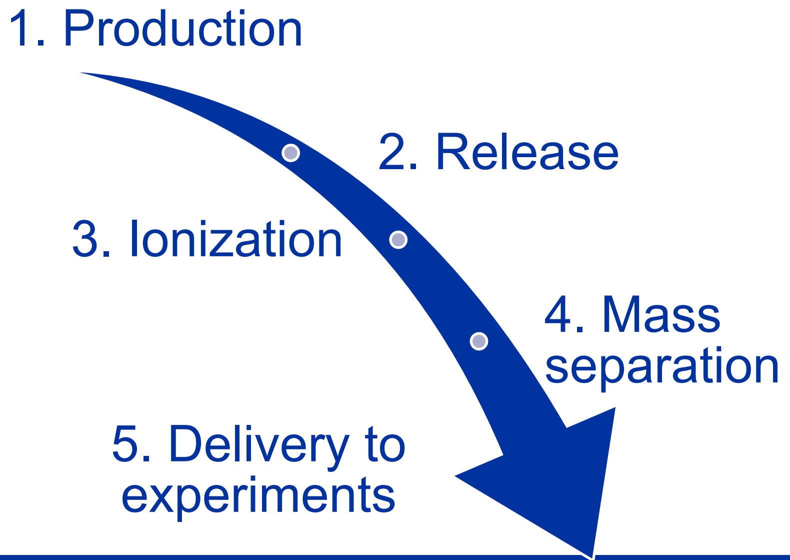

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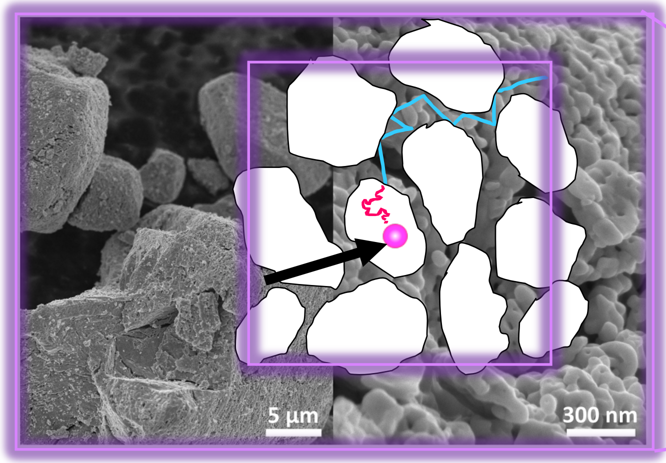

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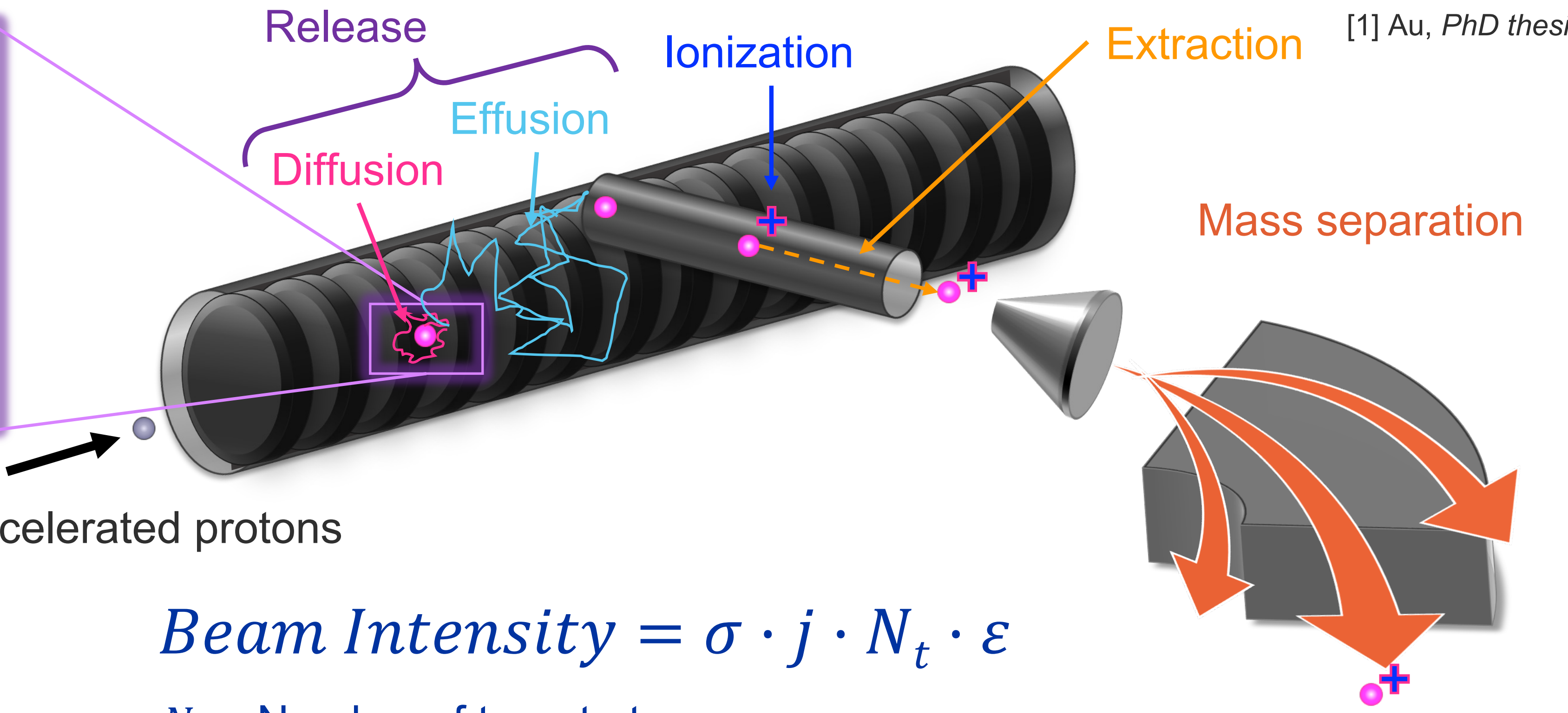

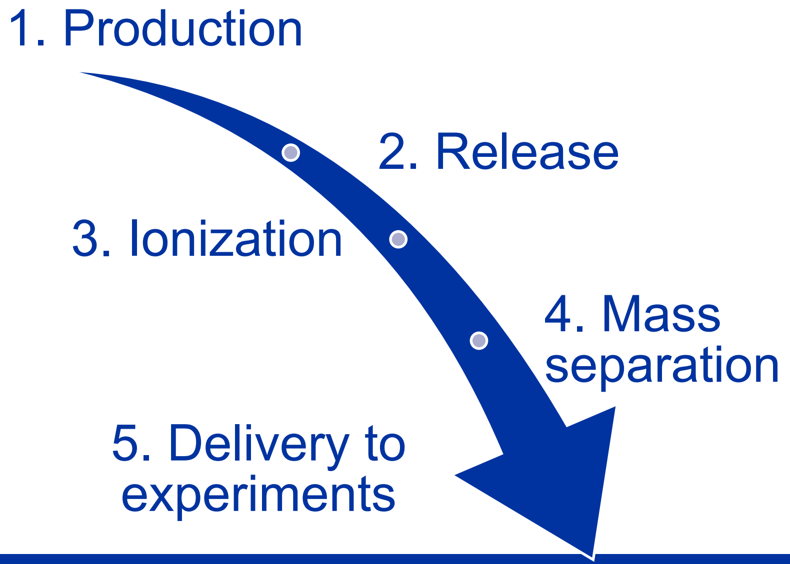

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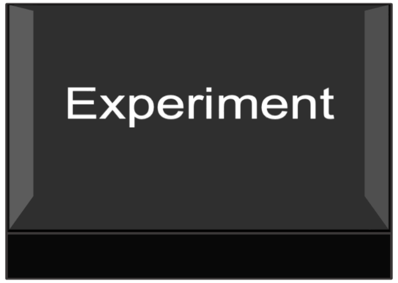

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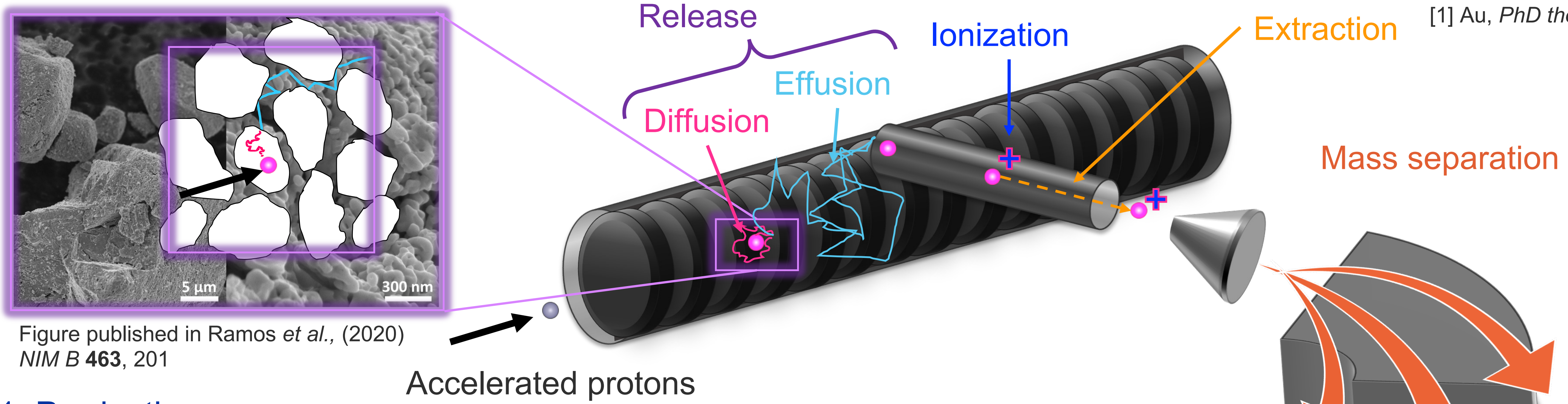

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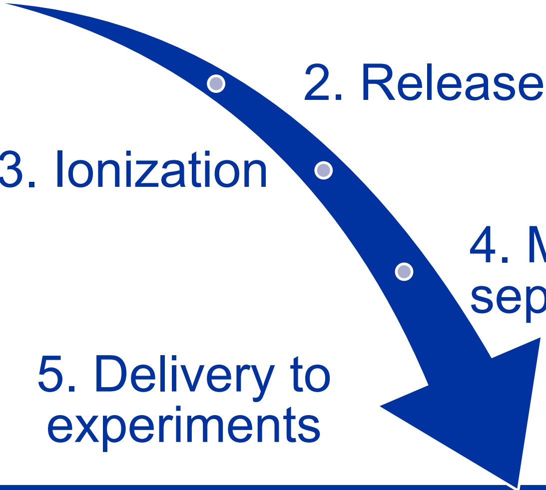

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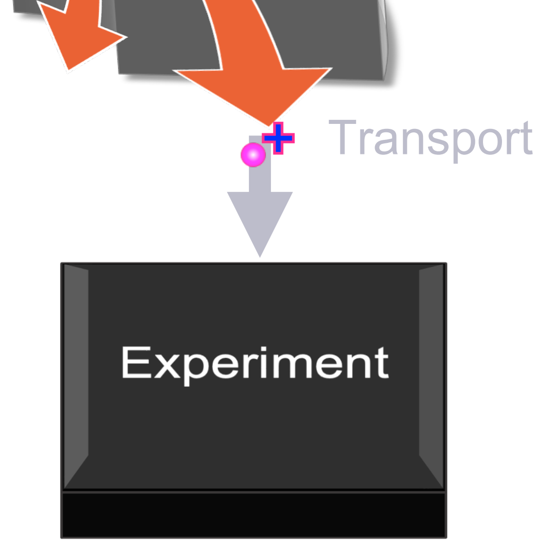


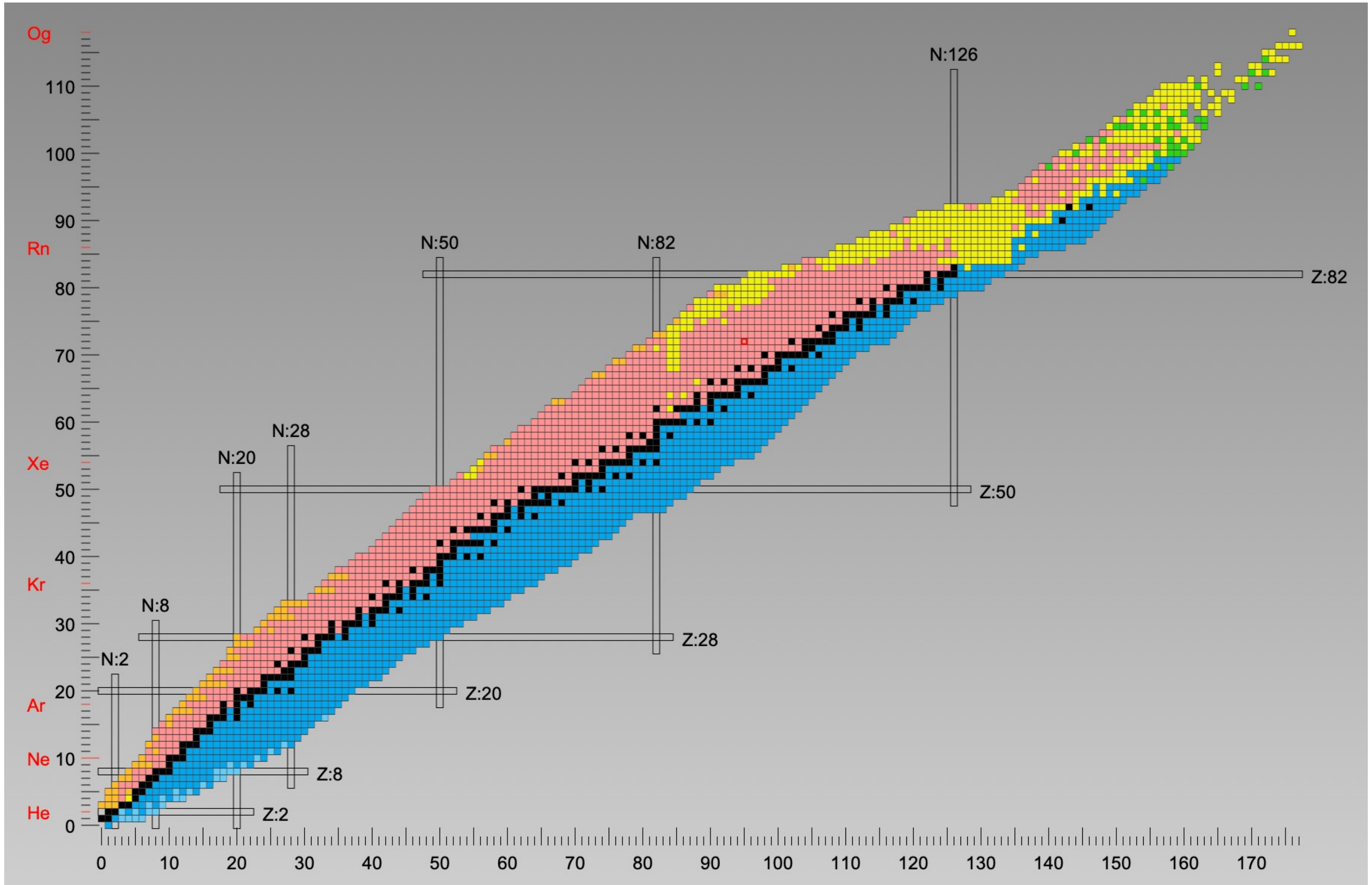
ISOL step 1: Production

Target selection

- Cross sections
- Bulk
- Half-lives

At ISOLDE

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



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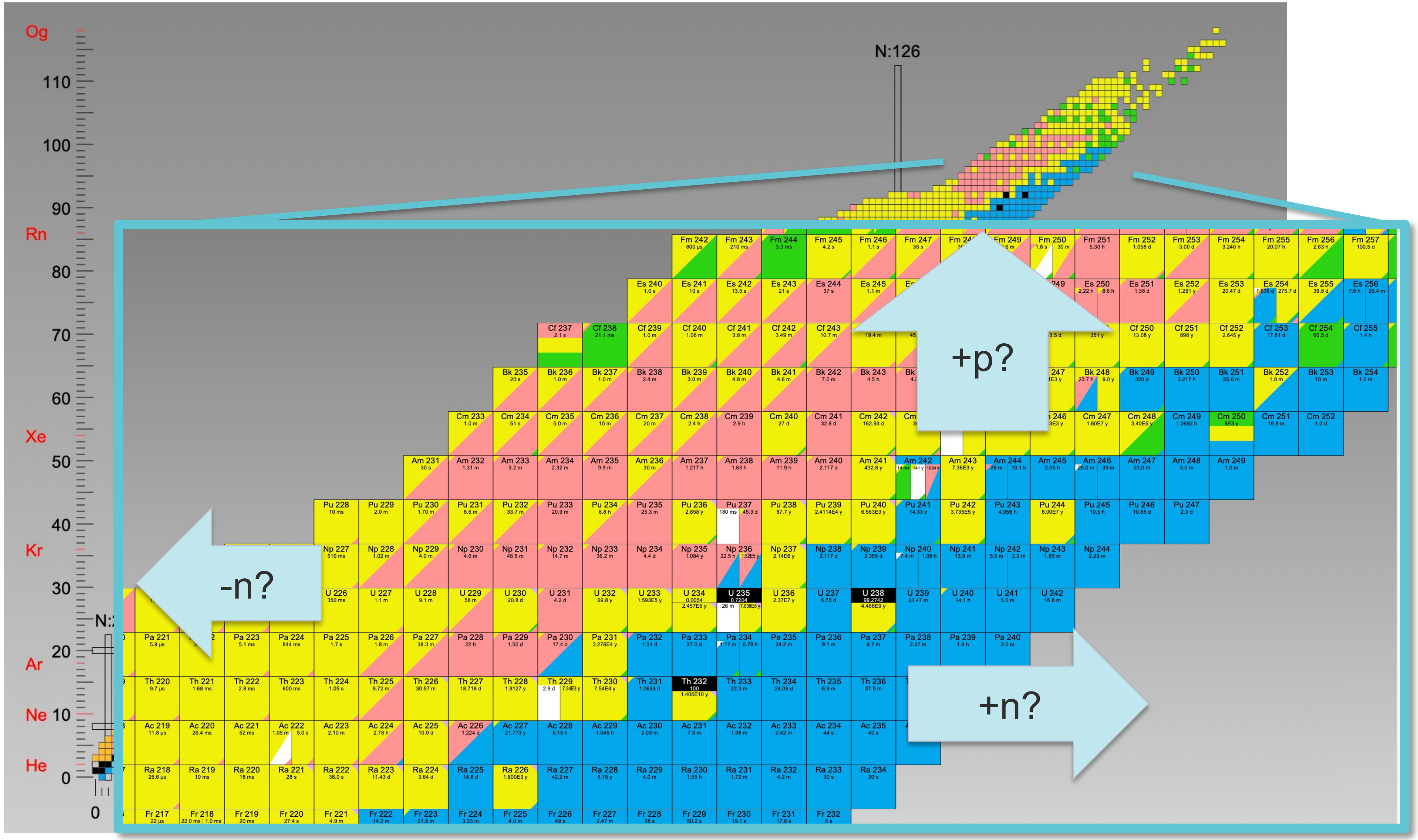

ISOL step 1: Production

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

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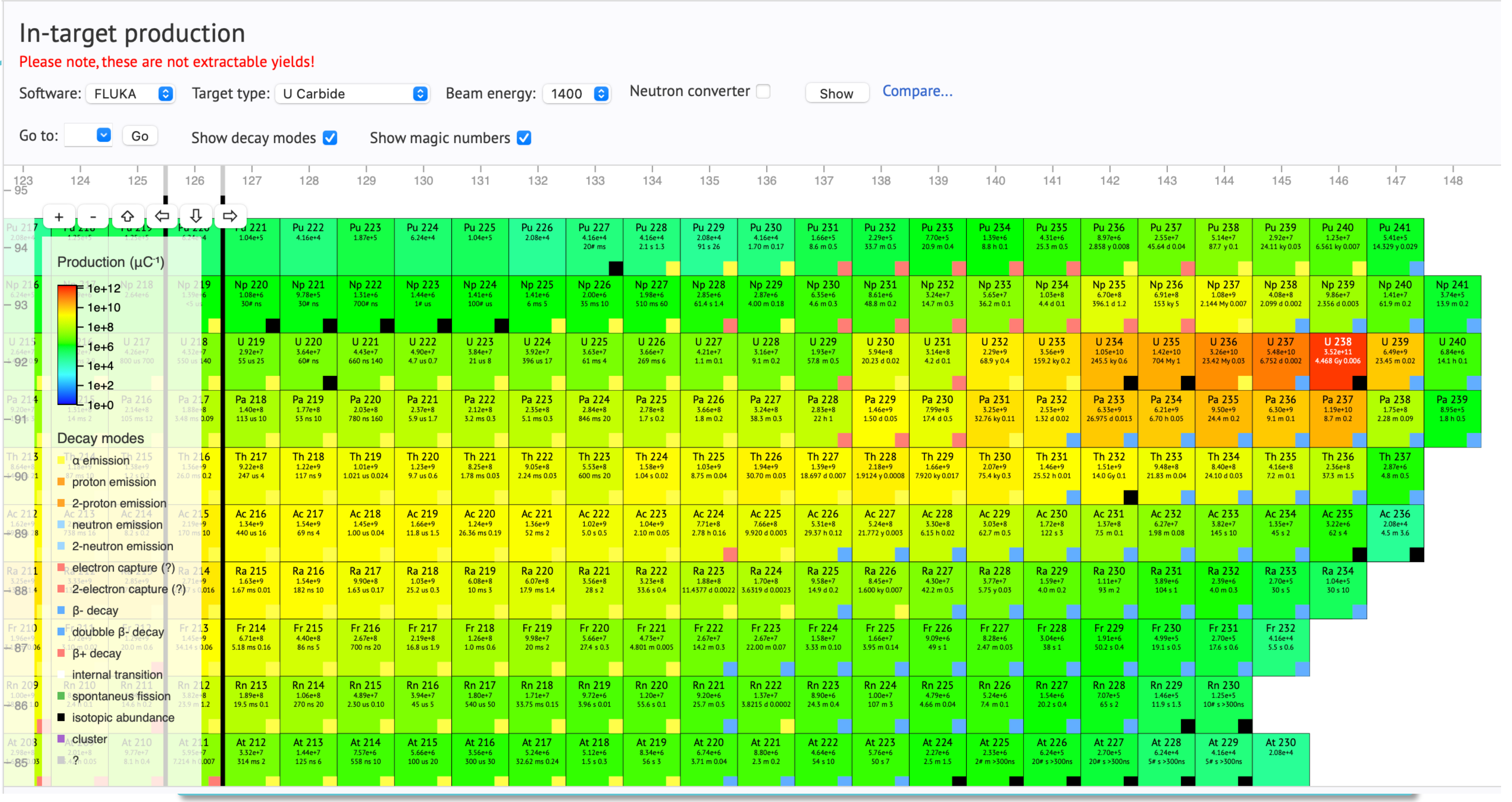

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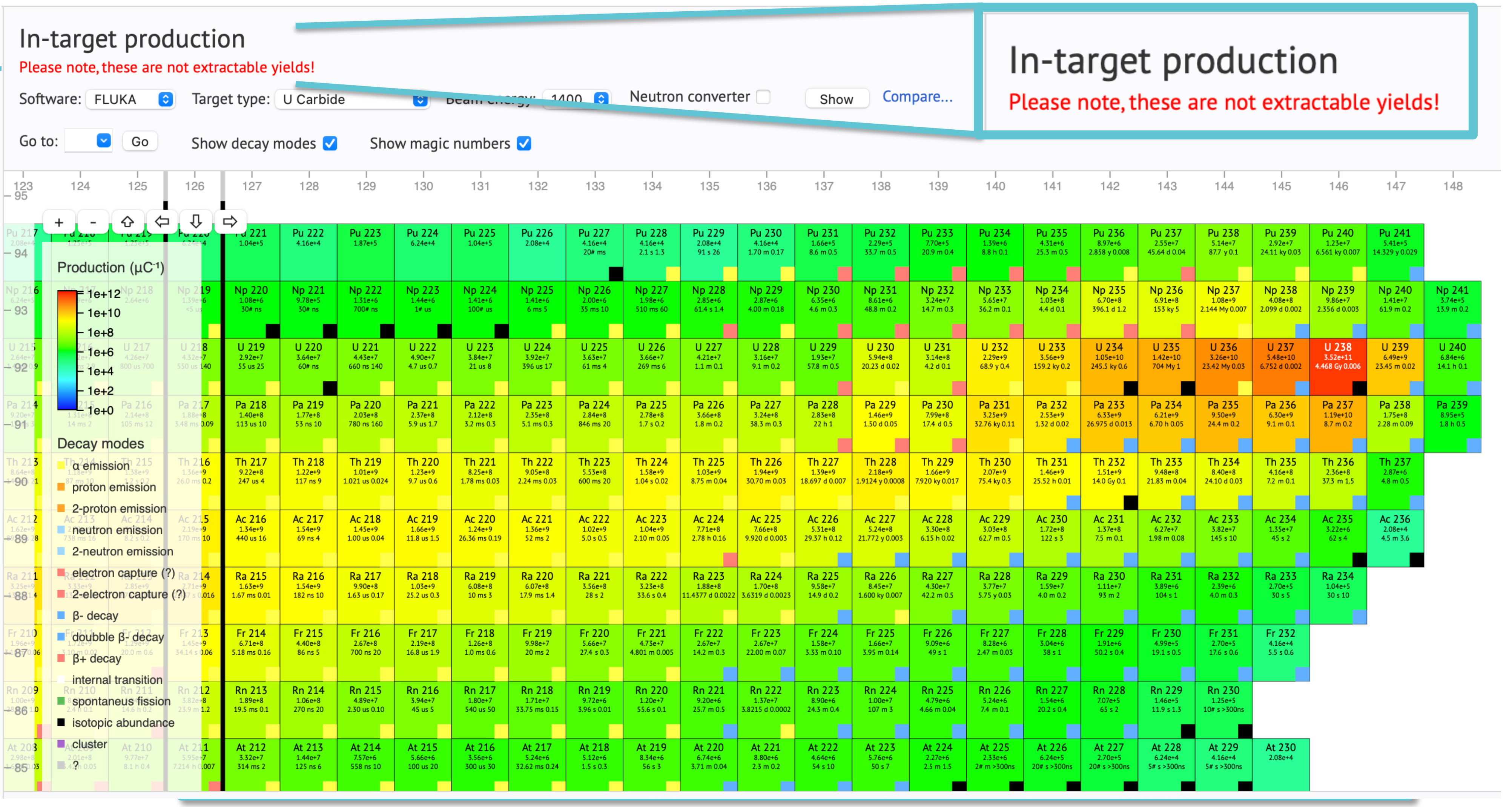

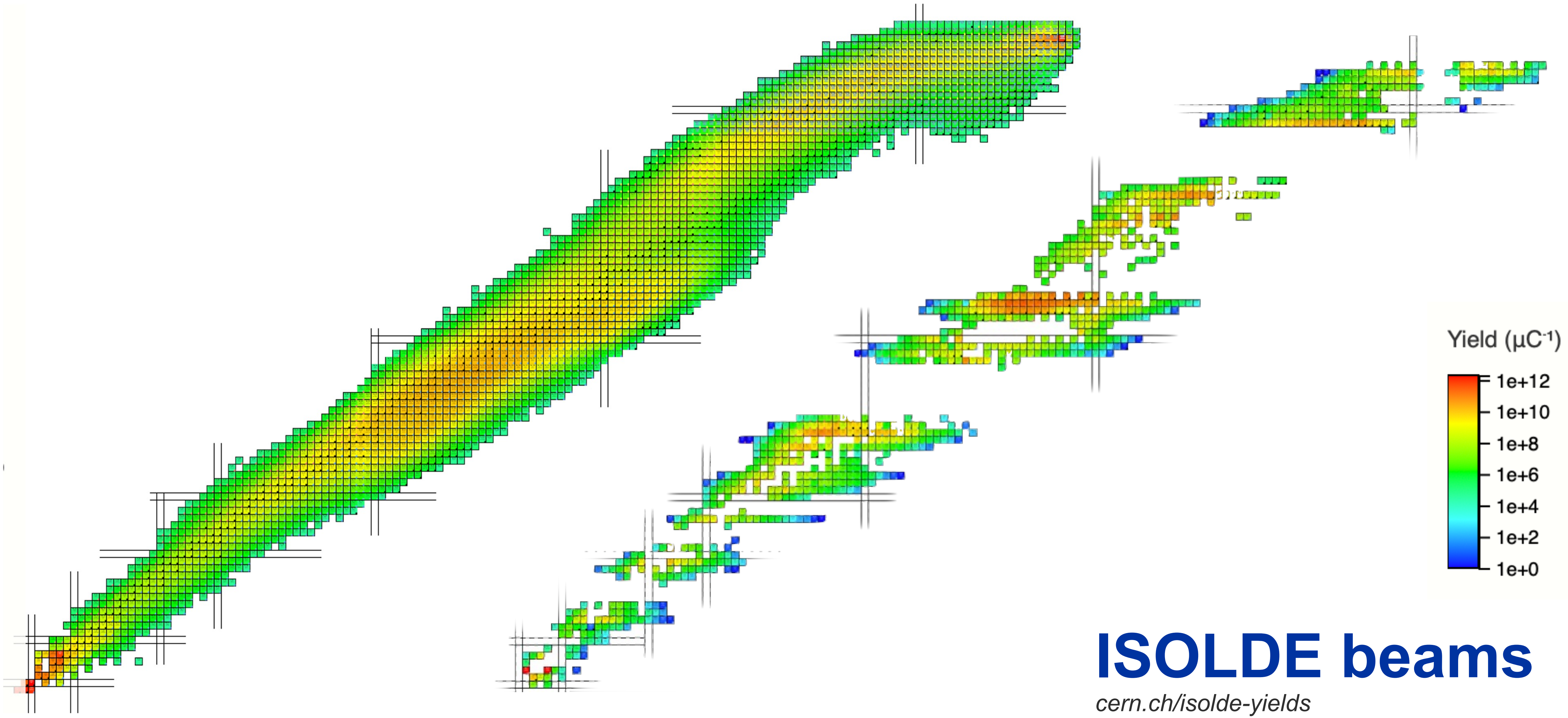

ISOLDE beams

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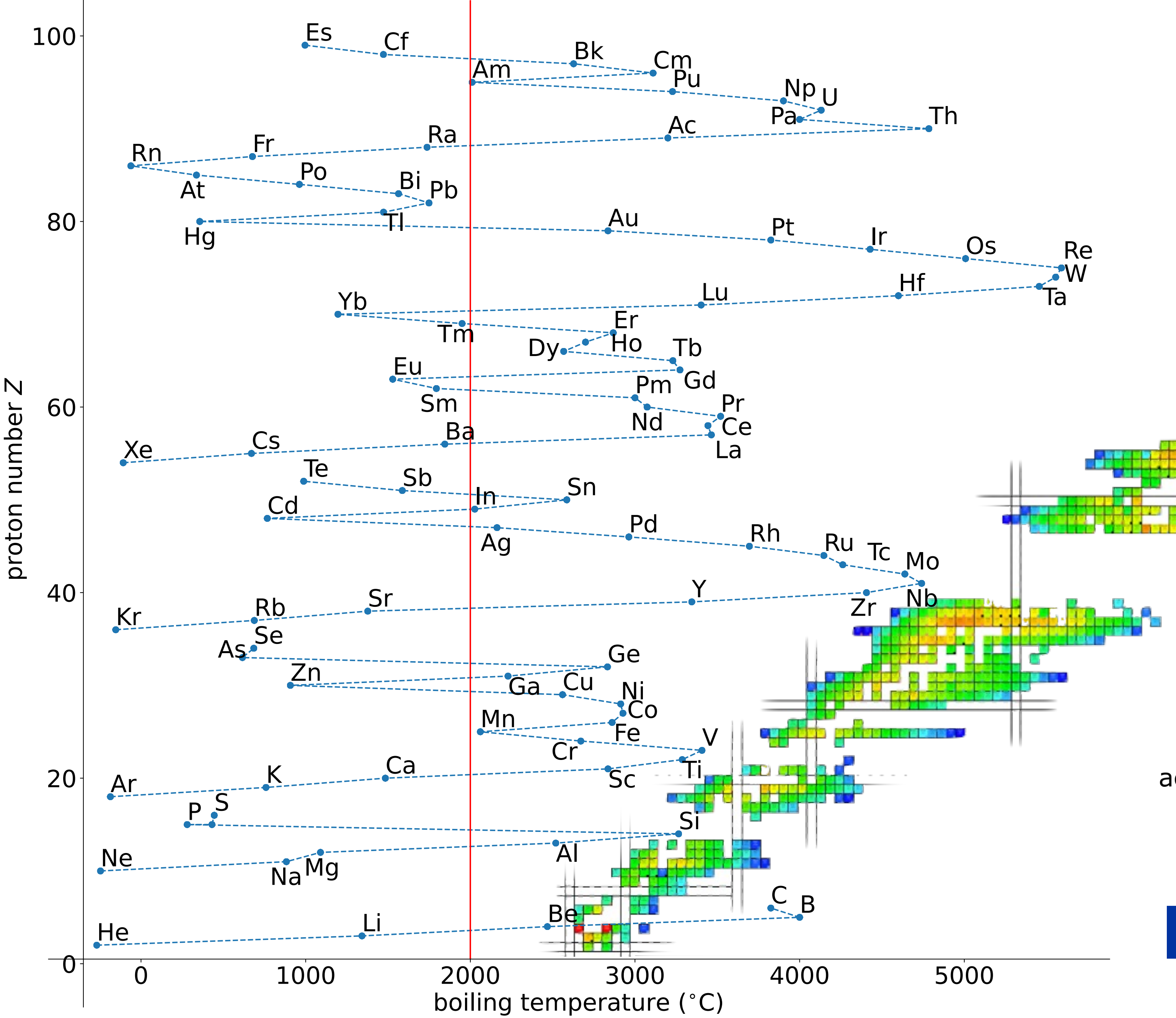

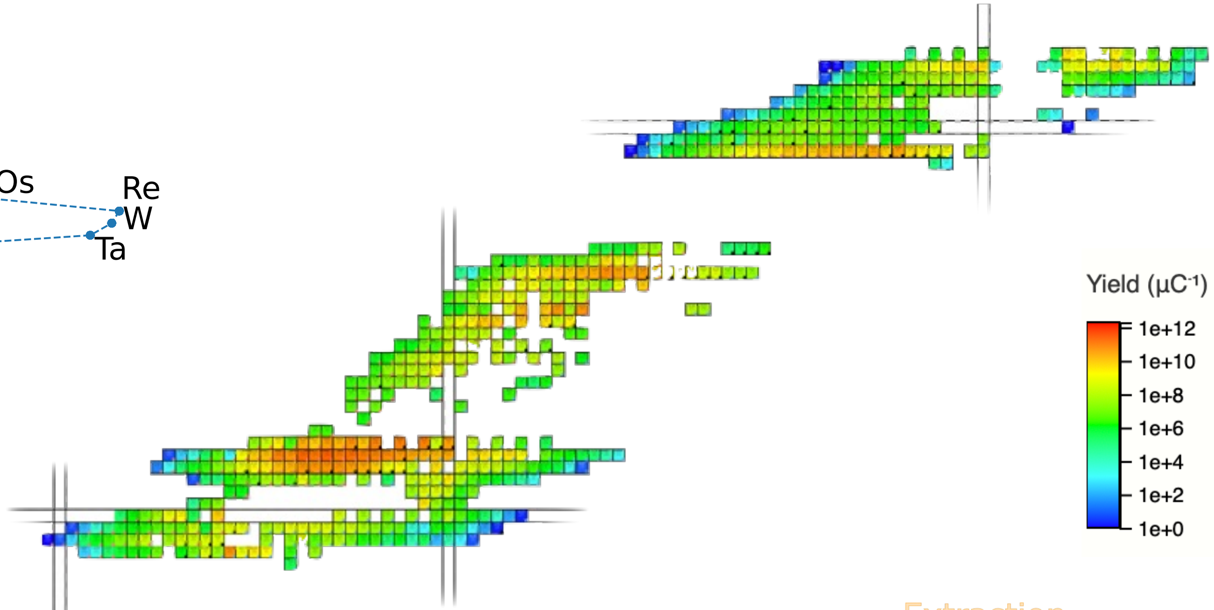

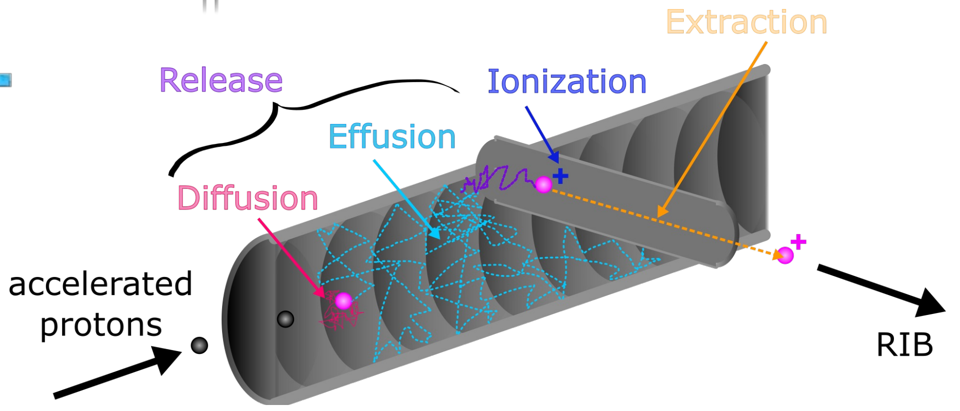

ISOL Step 2: Release

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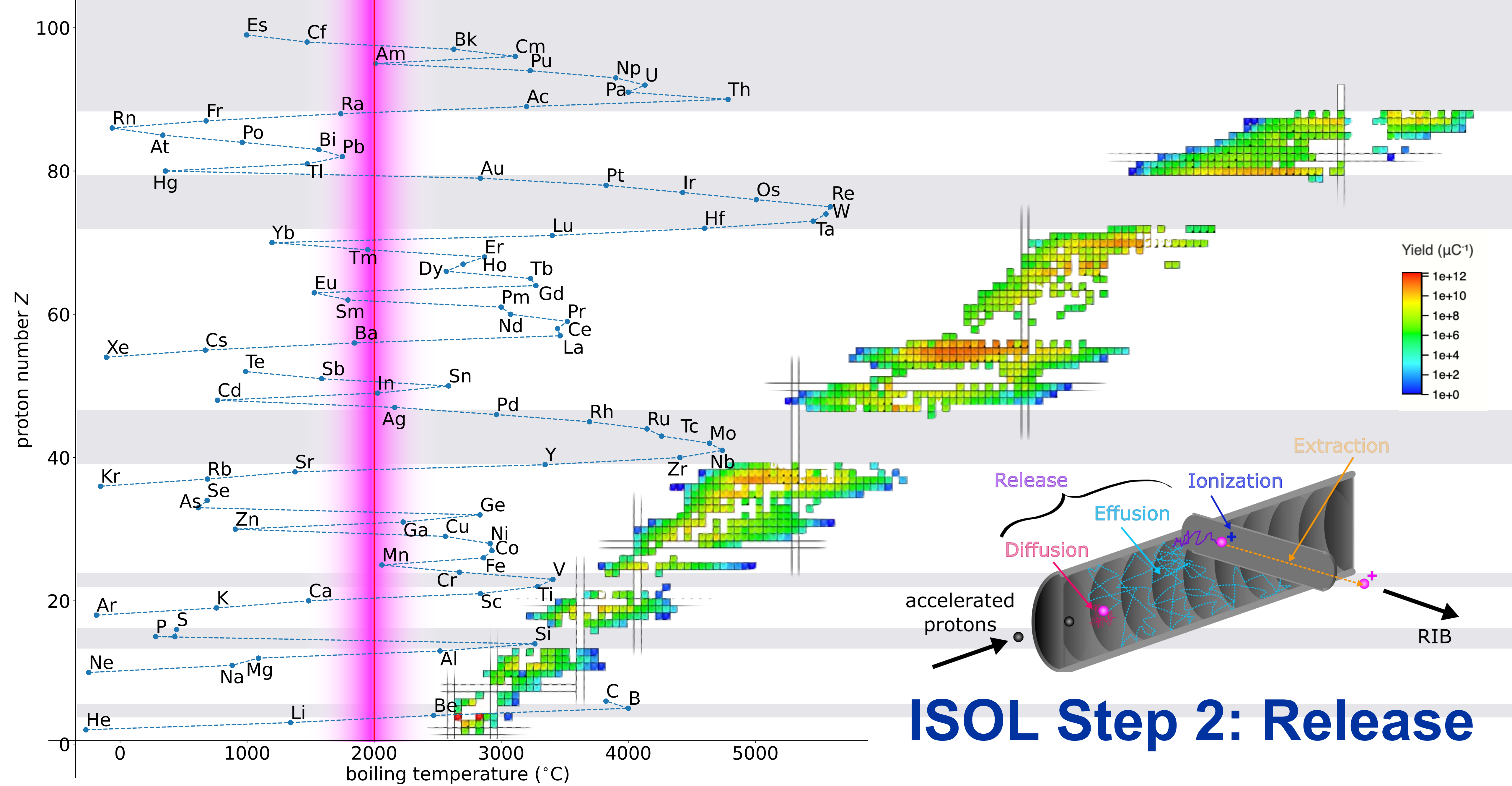
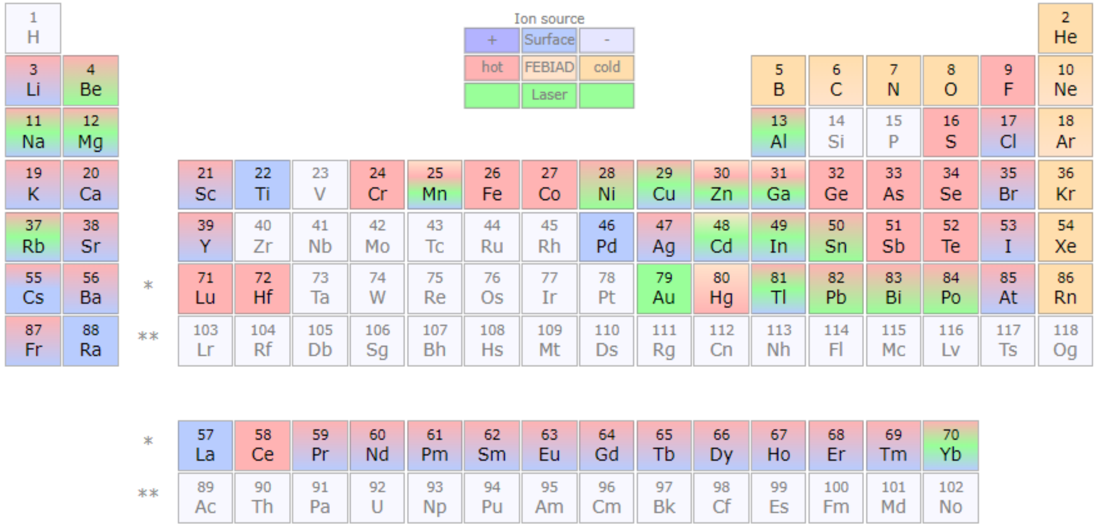


ISOL Step 3: Ionization



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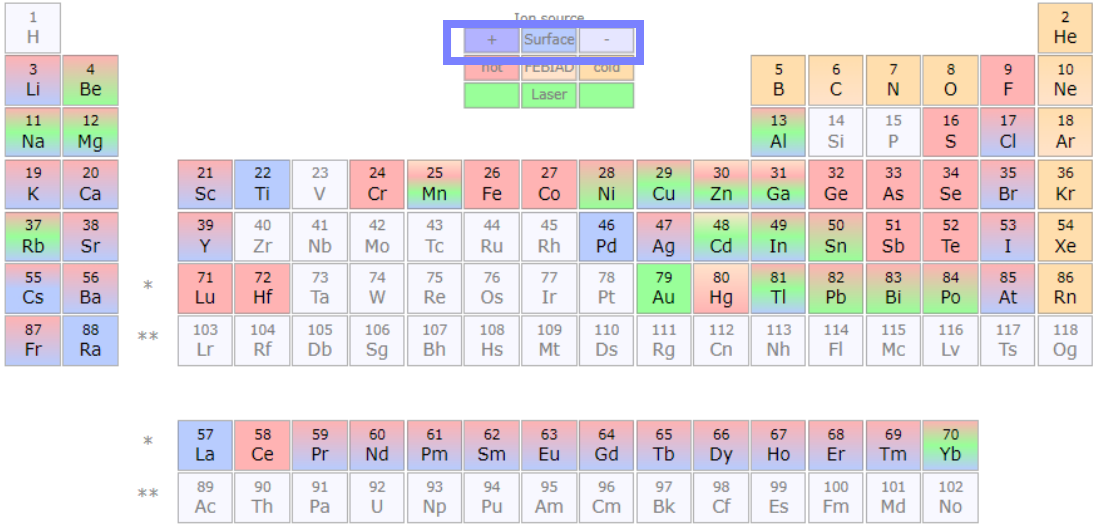
Ion sources

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

ISOL Step 3: Ionization



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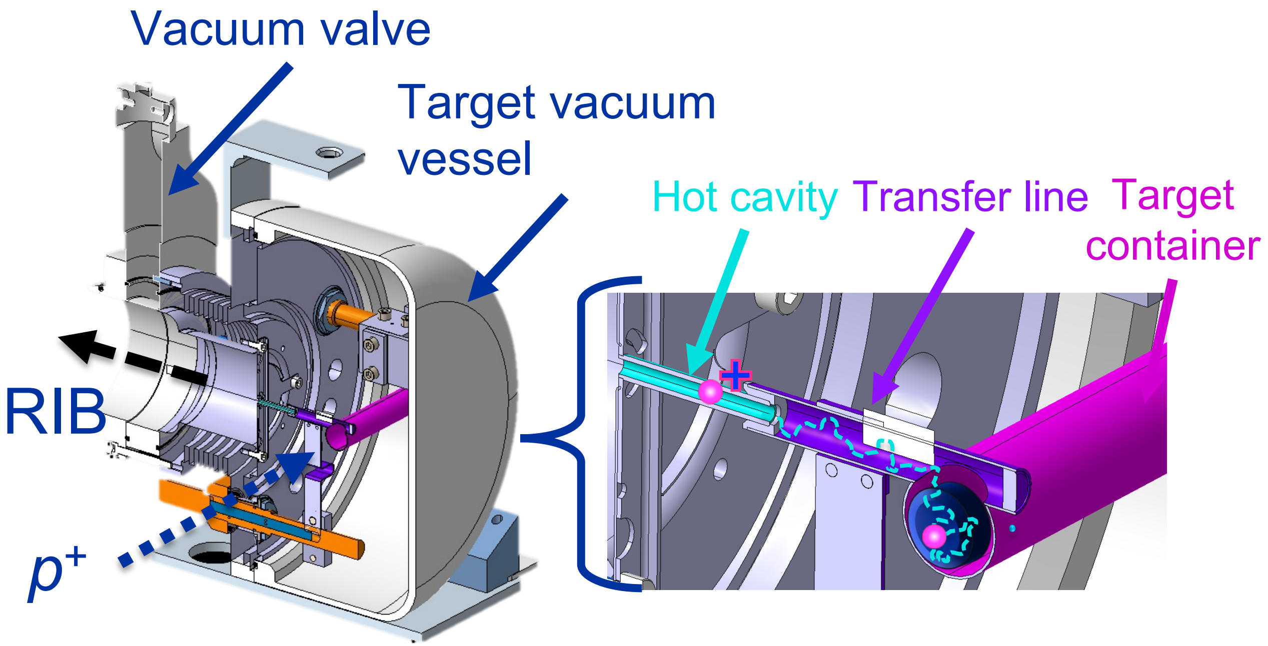

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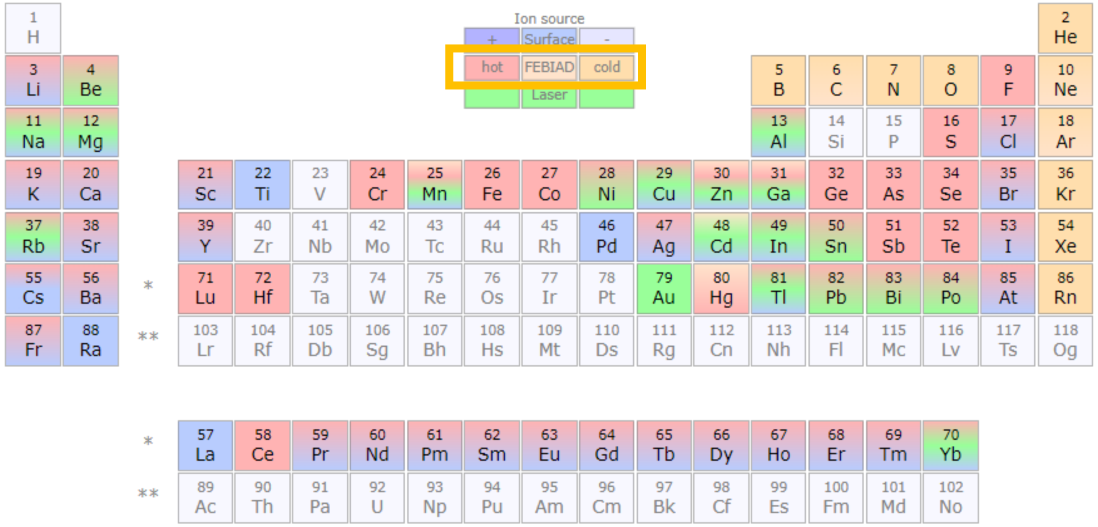

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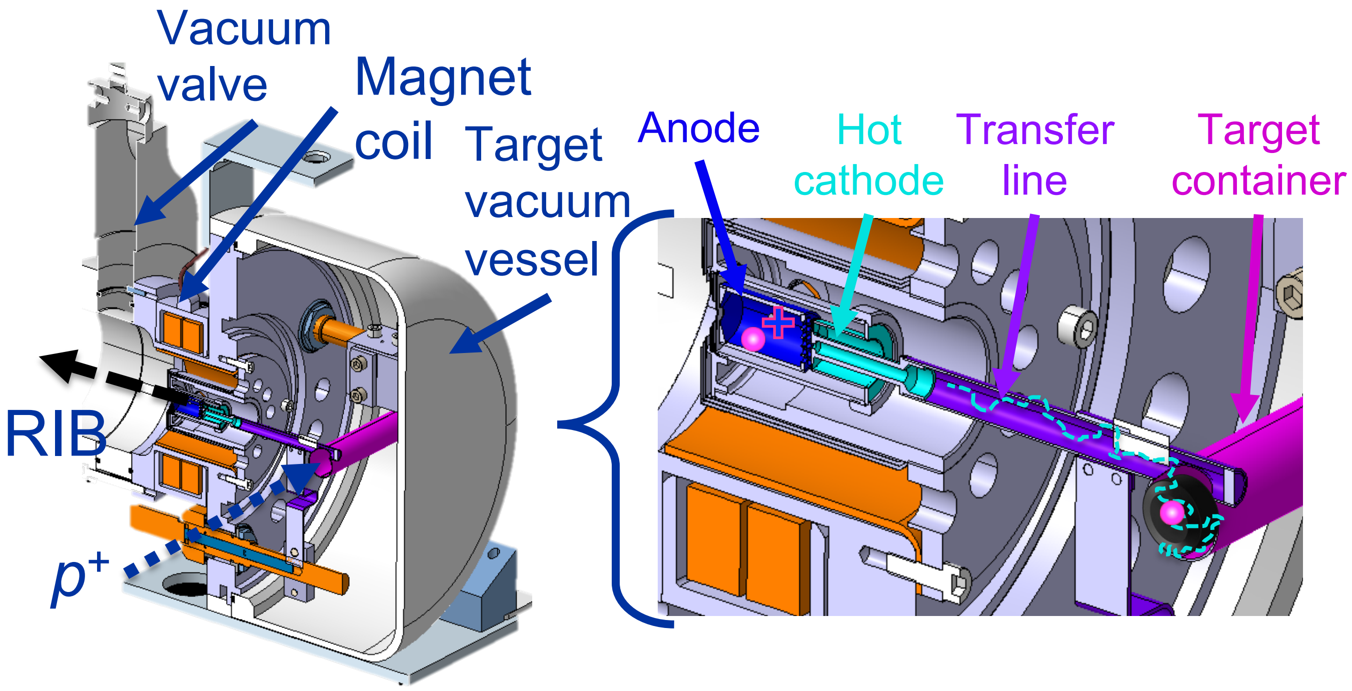

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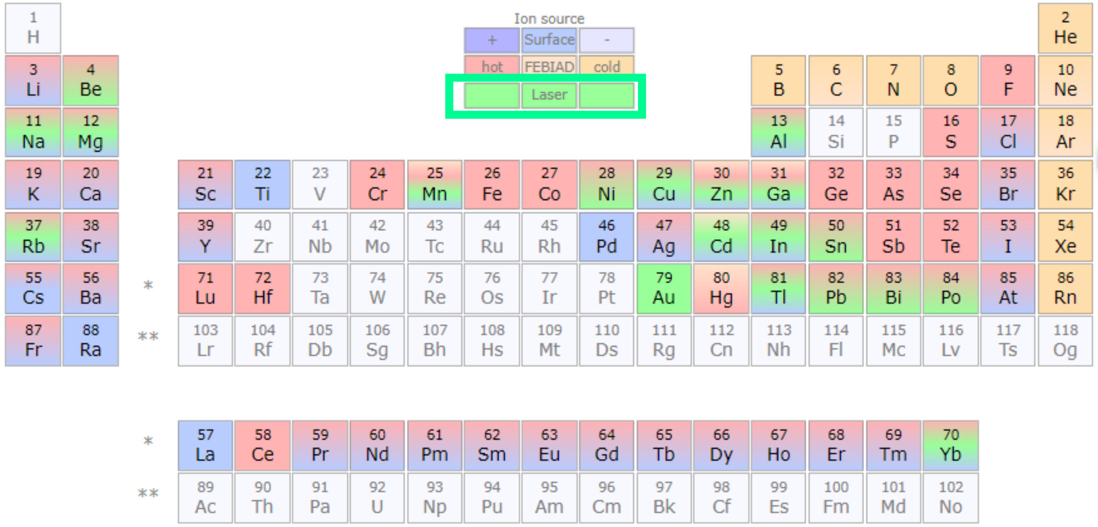

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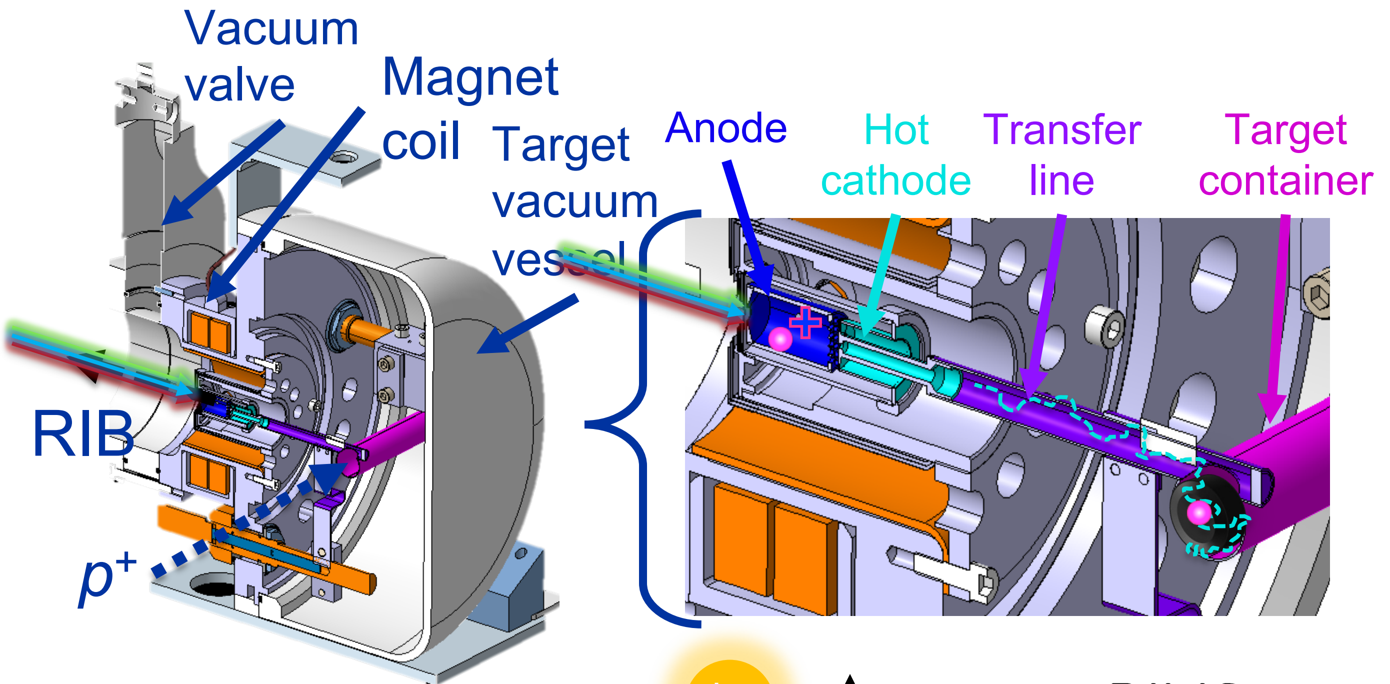

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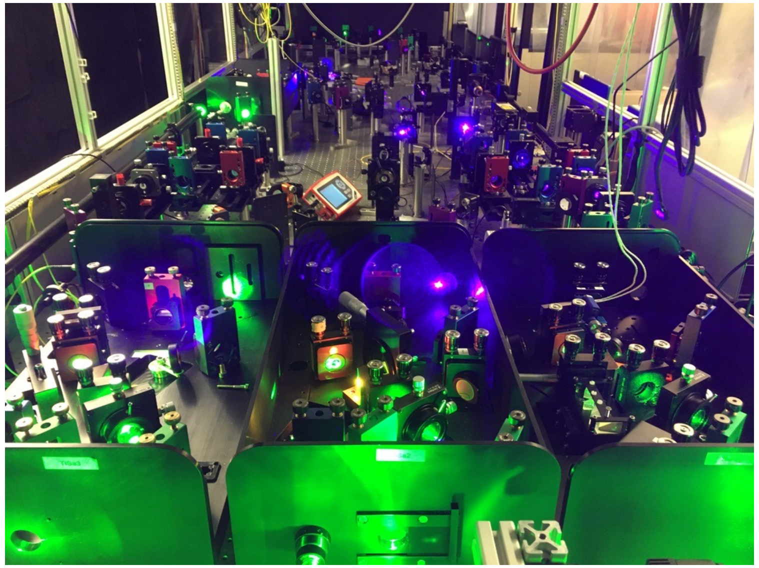

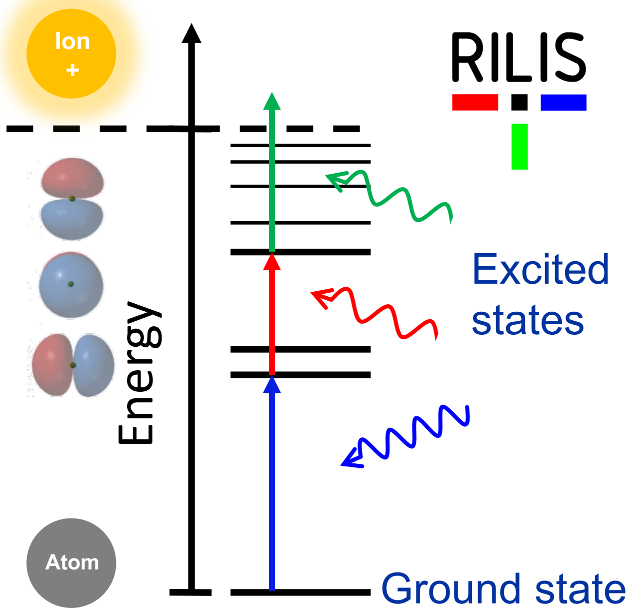

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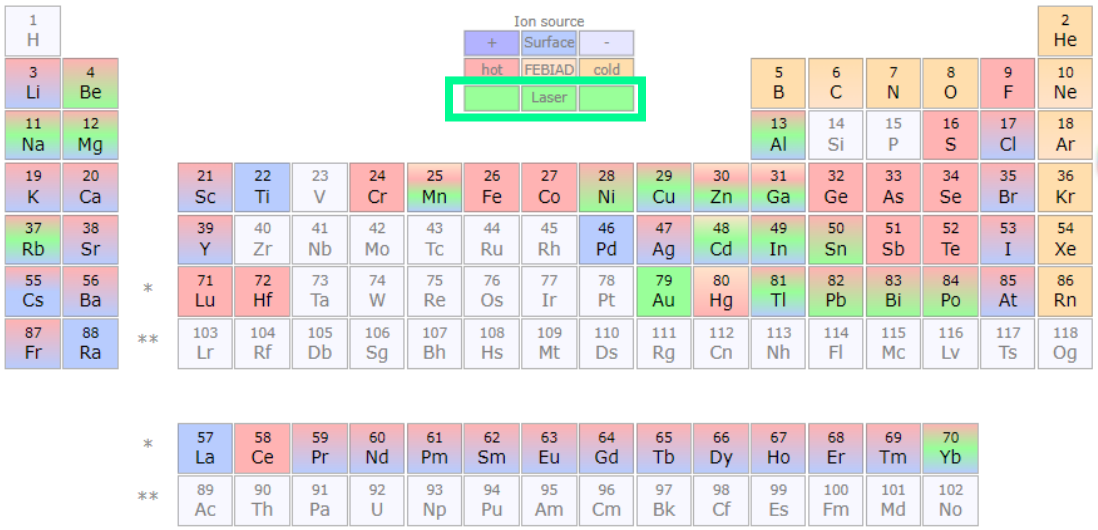

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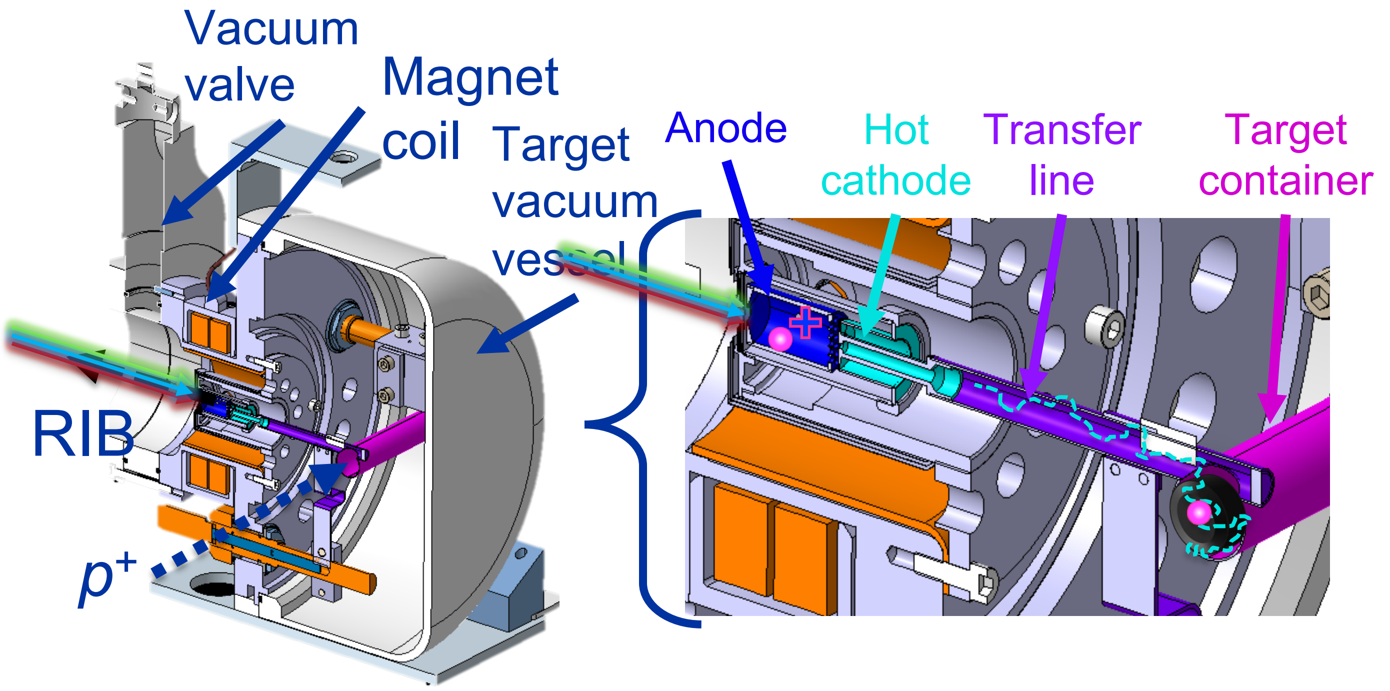

Ion sources

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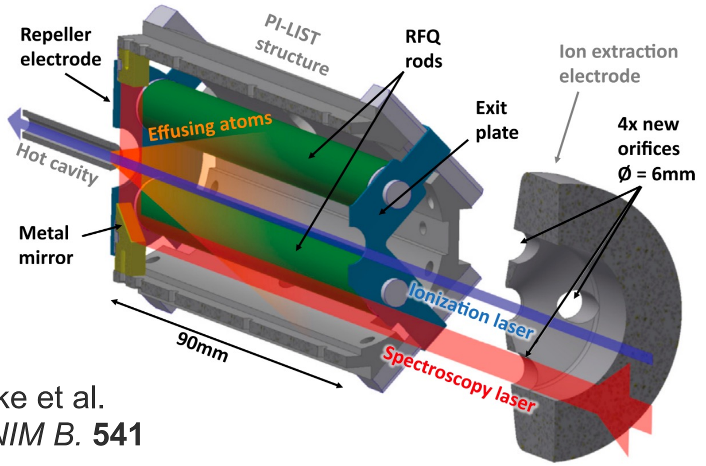
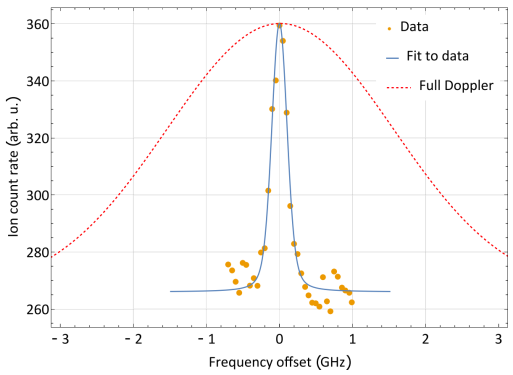
- Plasma / electron impact ionization
- Resonance laser ionization
    - Resonance ionization spectroscopy (RIS)



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

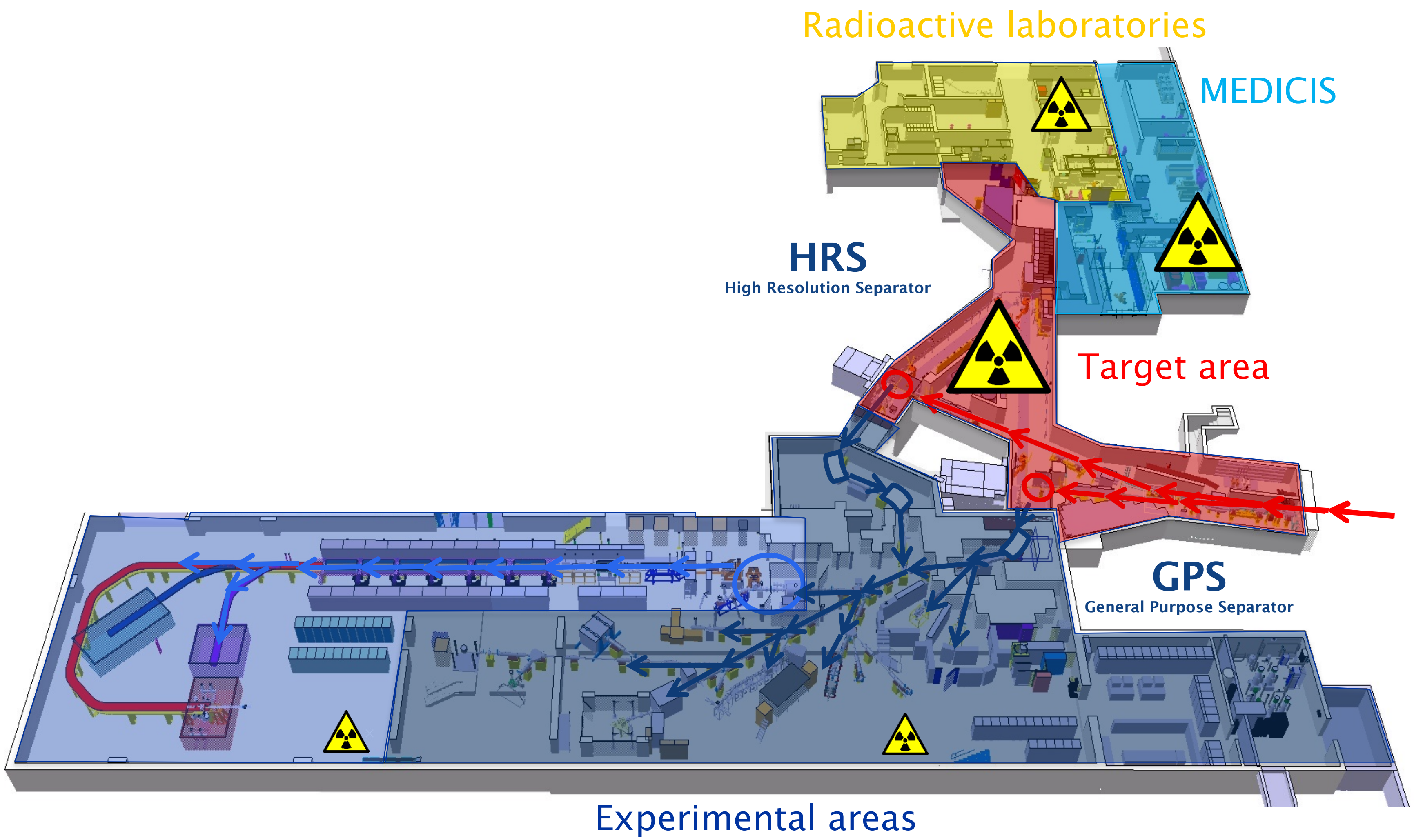


ISOL

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

"On-Line":

- Production
- Release
- Ionization
- Extraction



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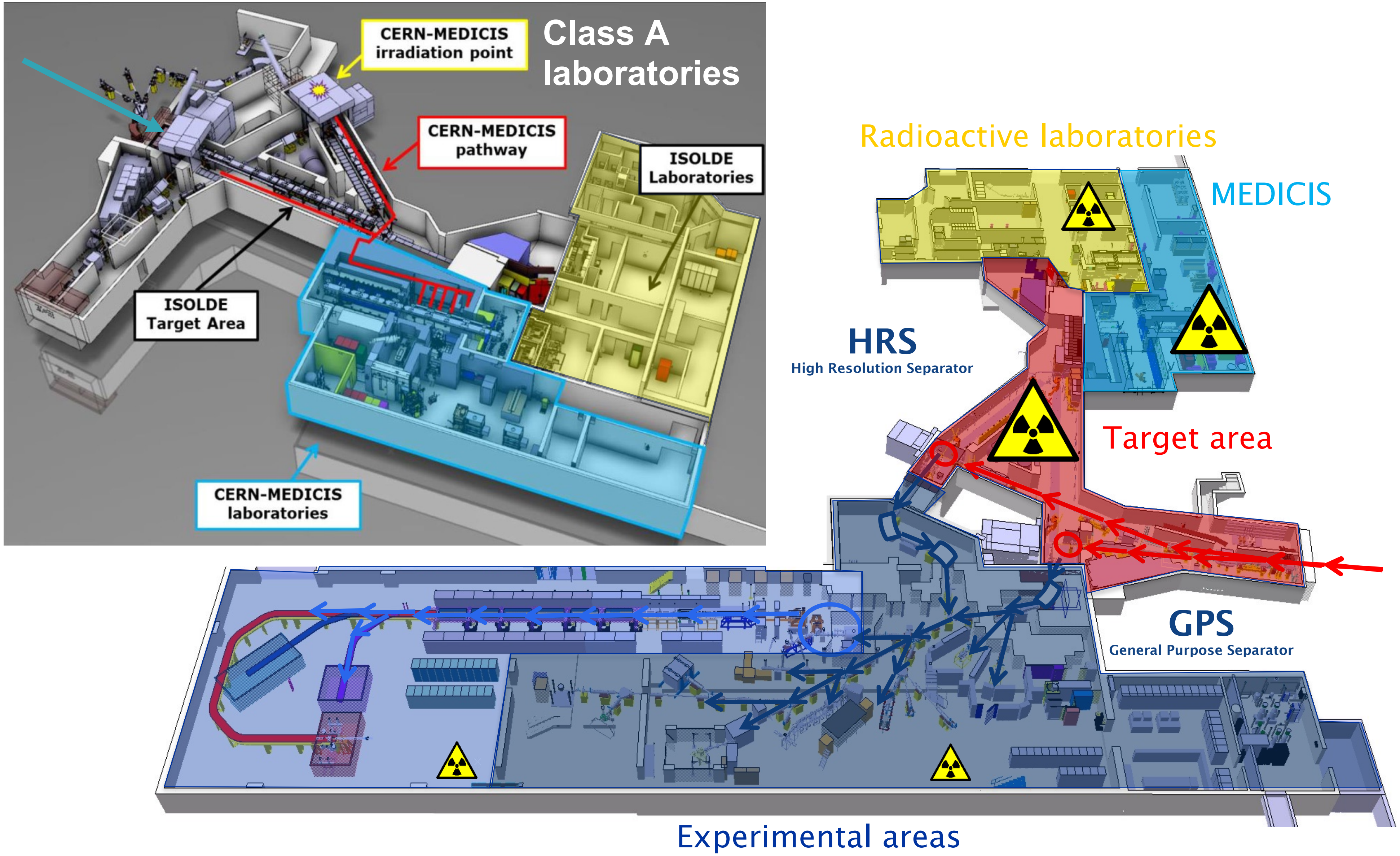

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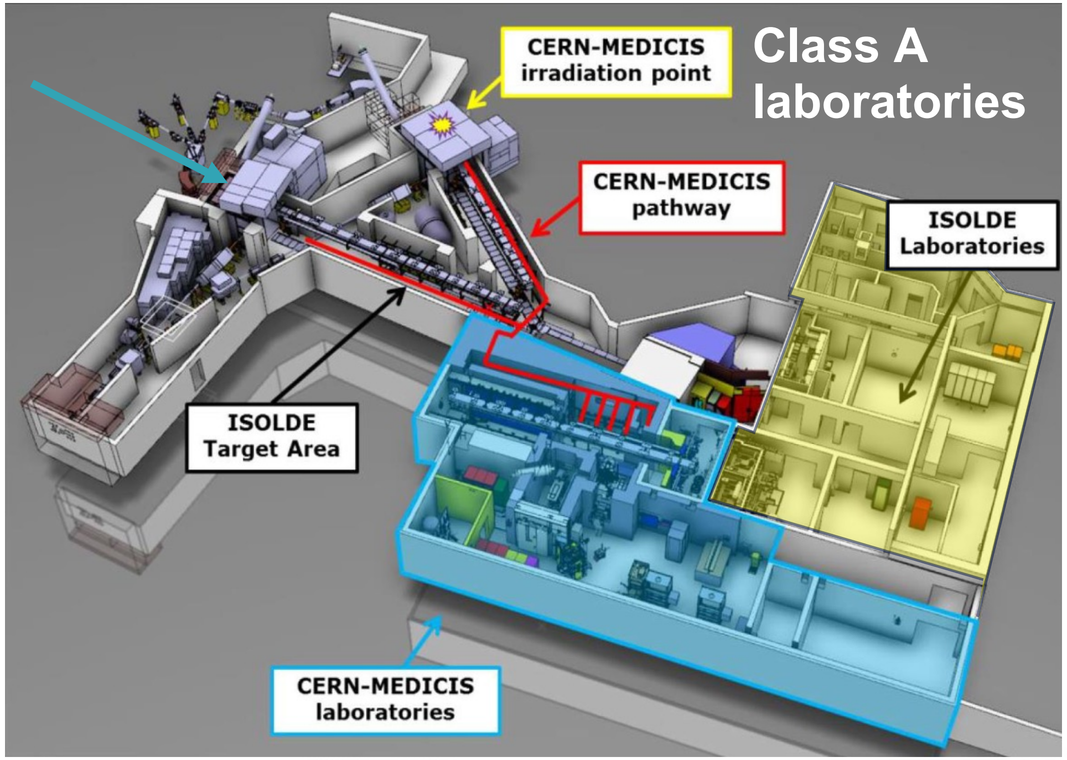

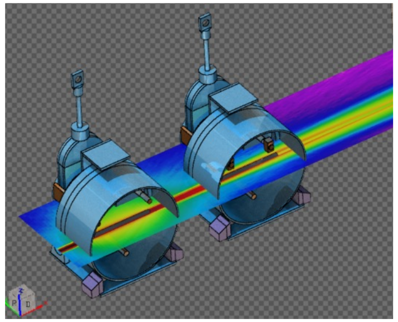

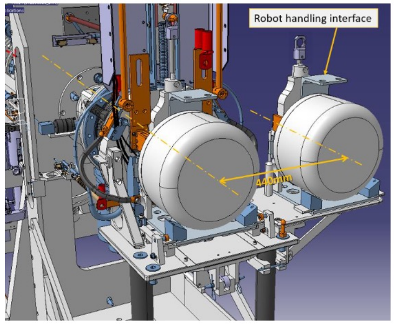

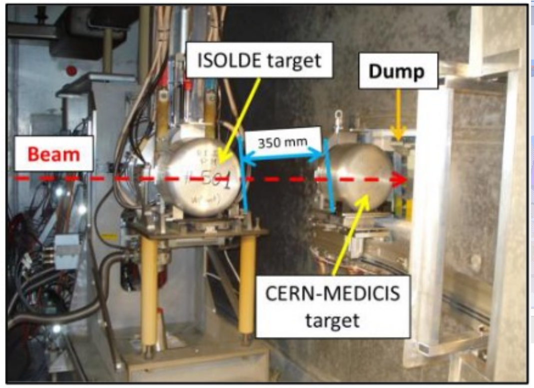

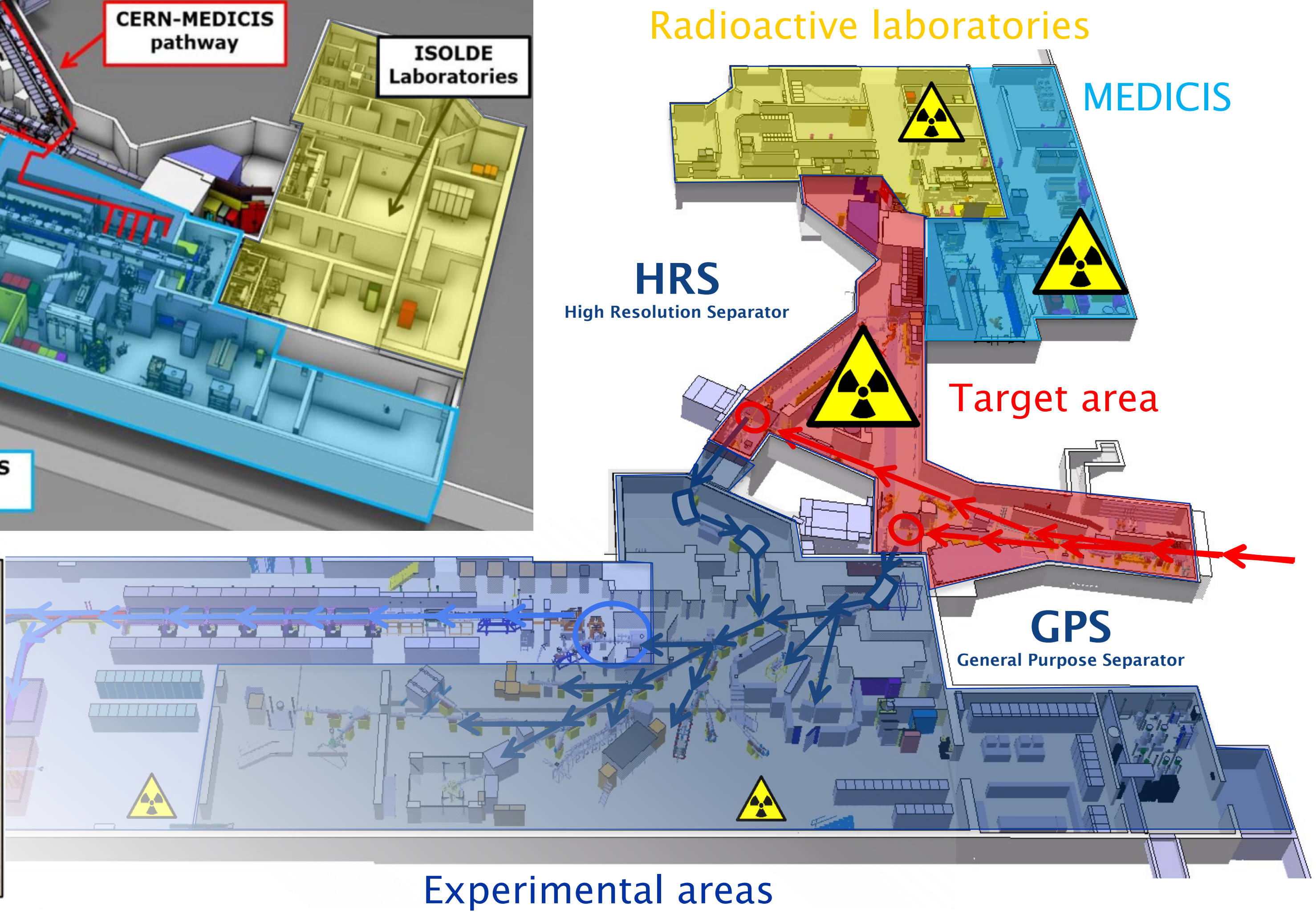

Catherall et al. (2017) *J. Phys G* **44**, 094002
isolde.web.cern.ch

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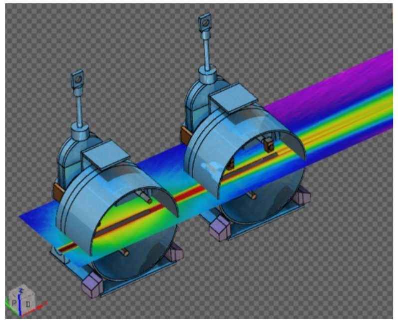
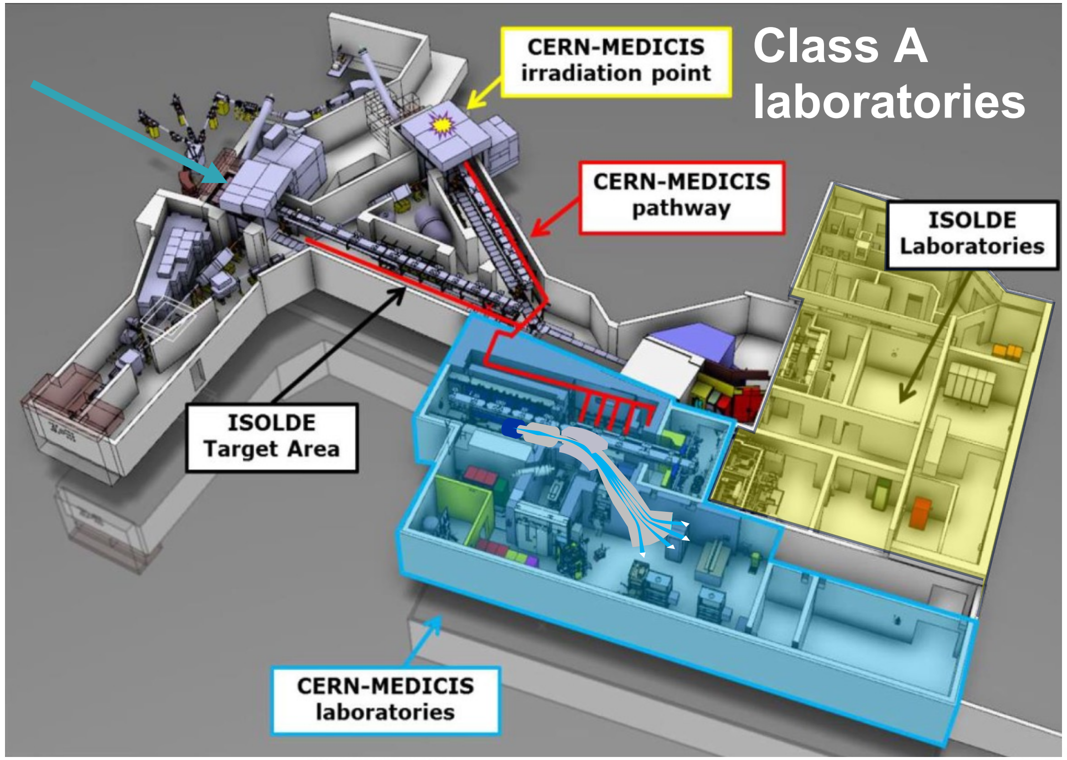
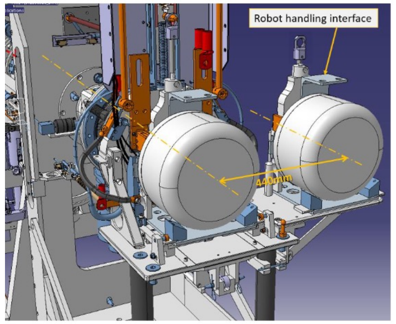
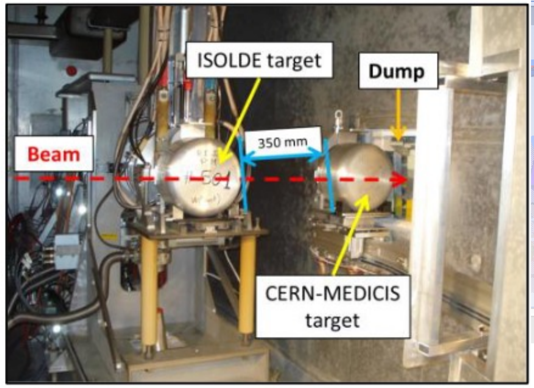
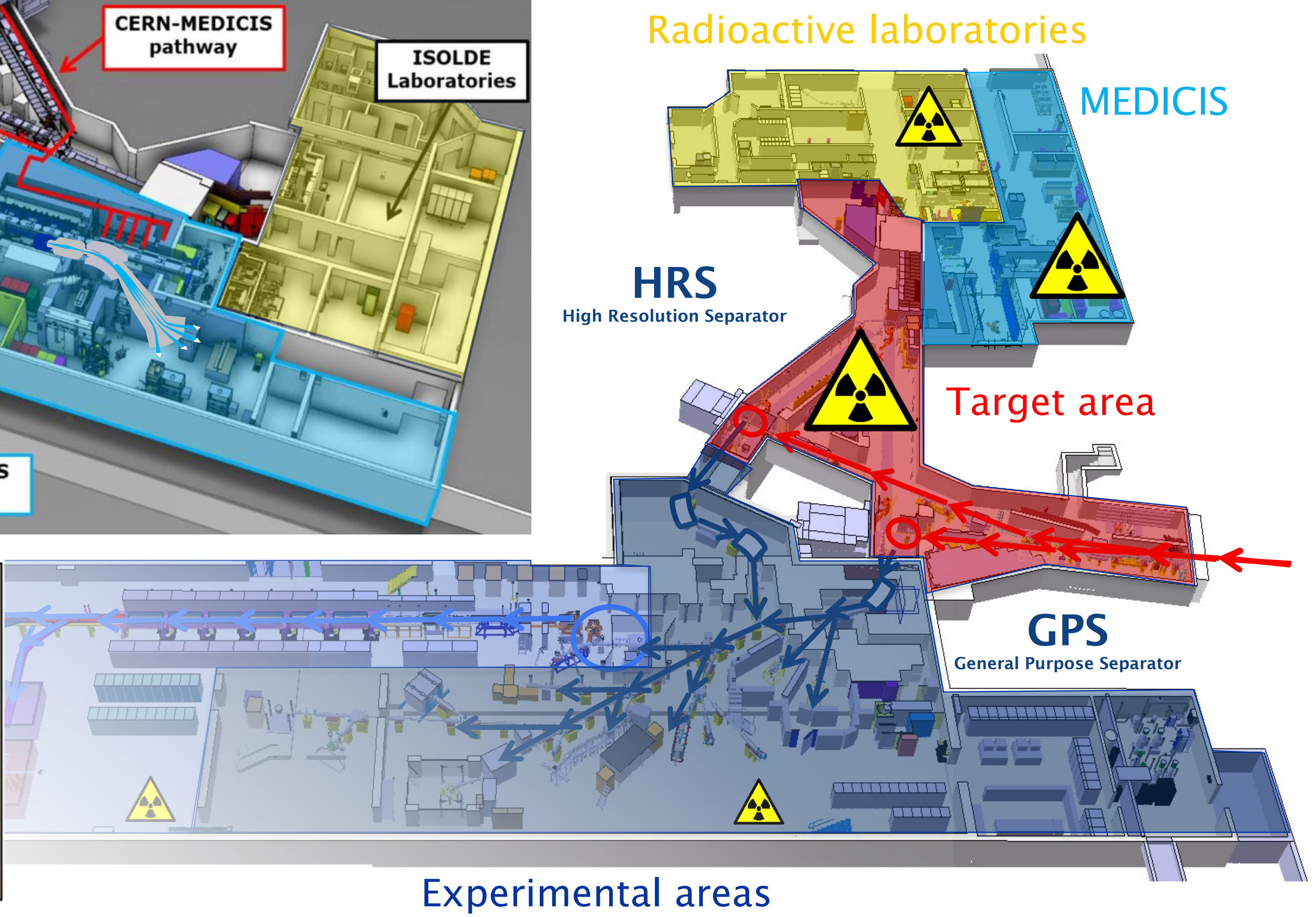




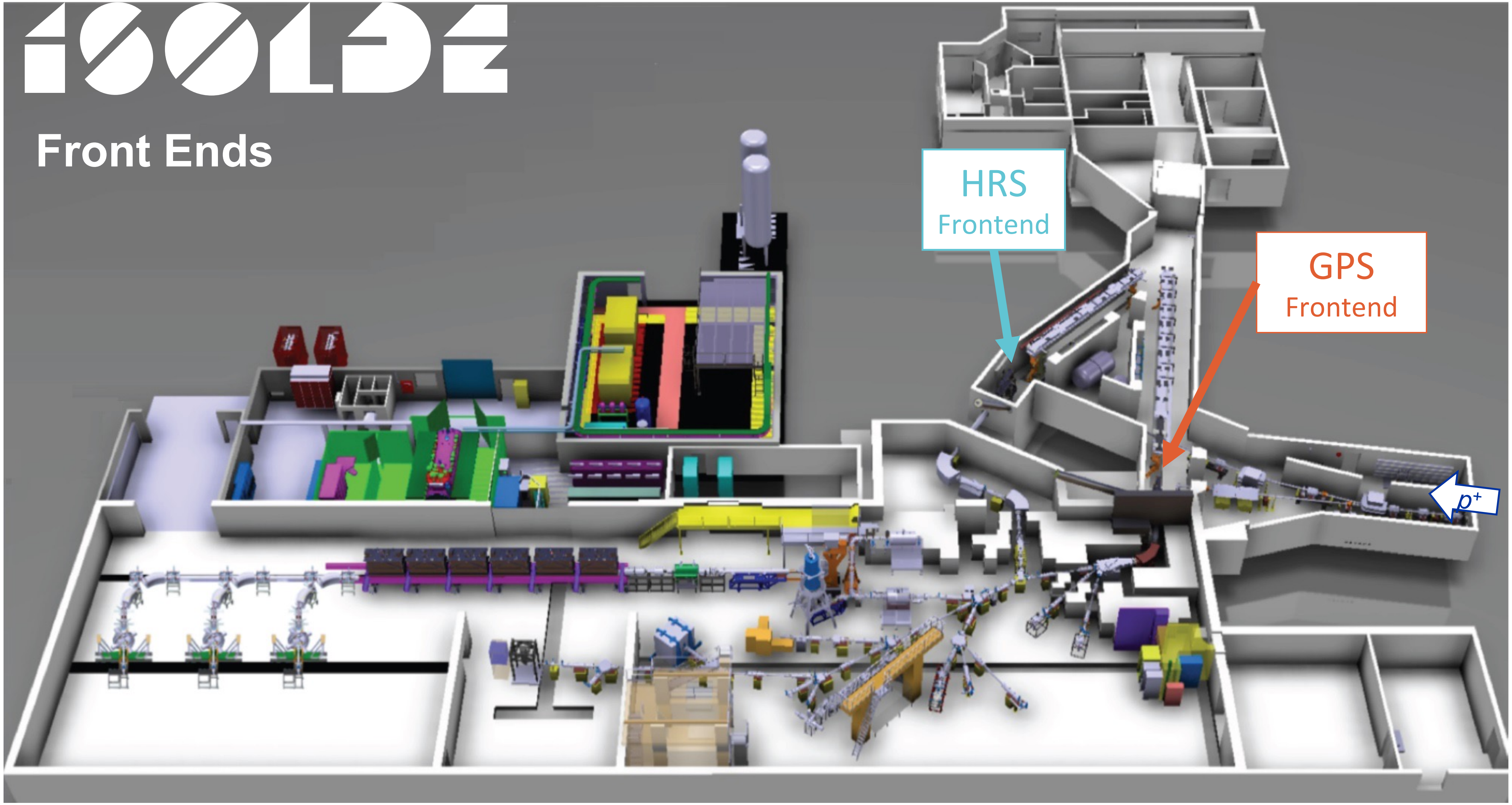






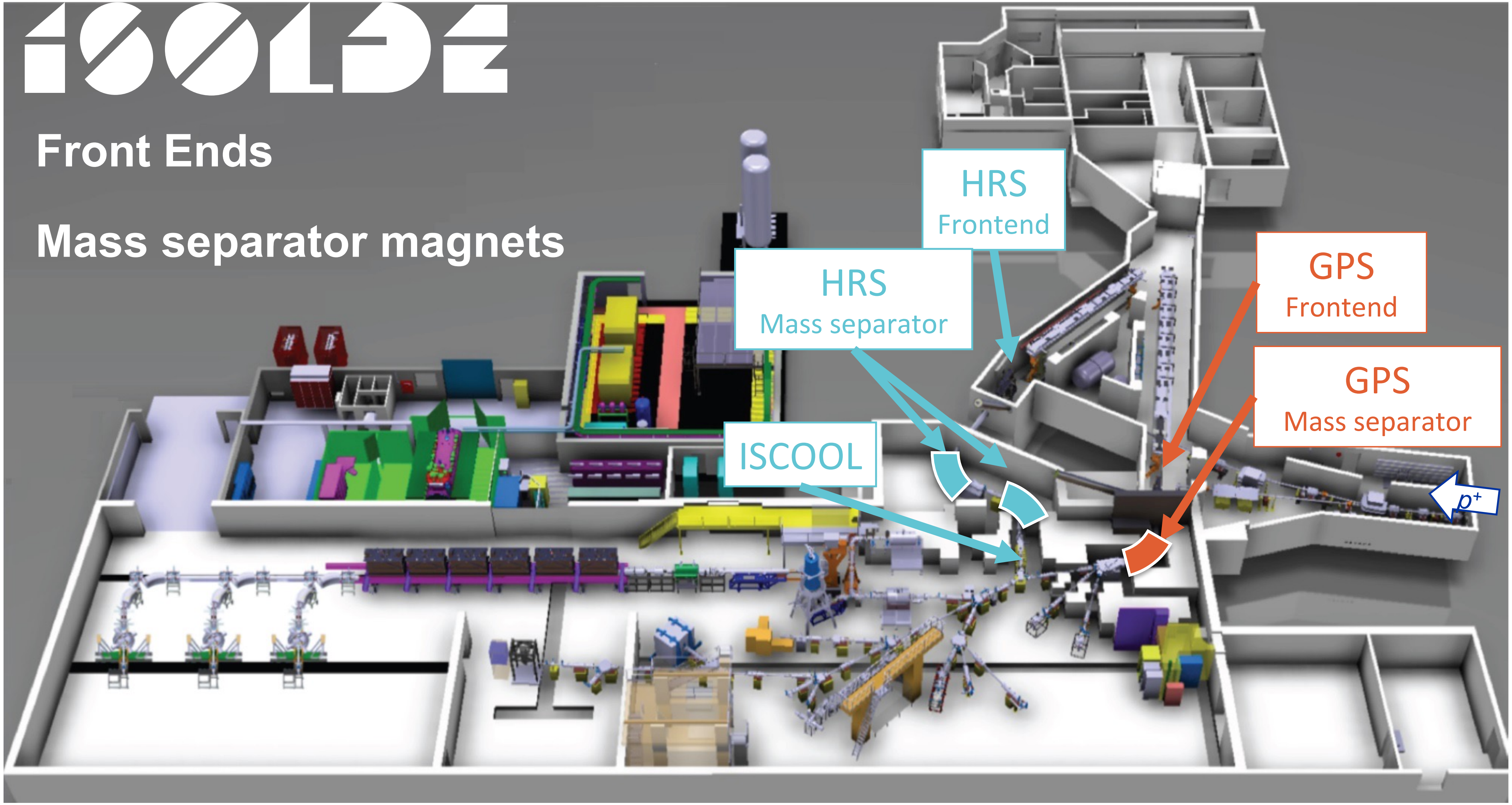
ISOL Step 4: Mass separation

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



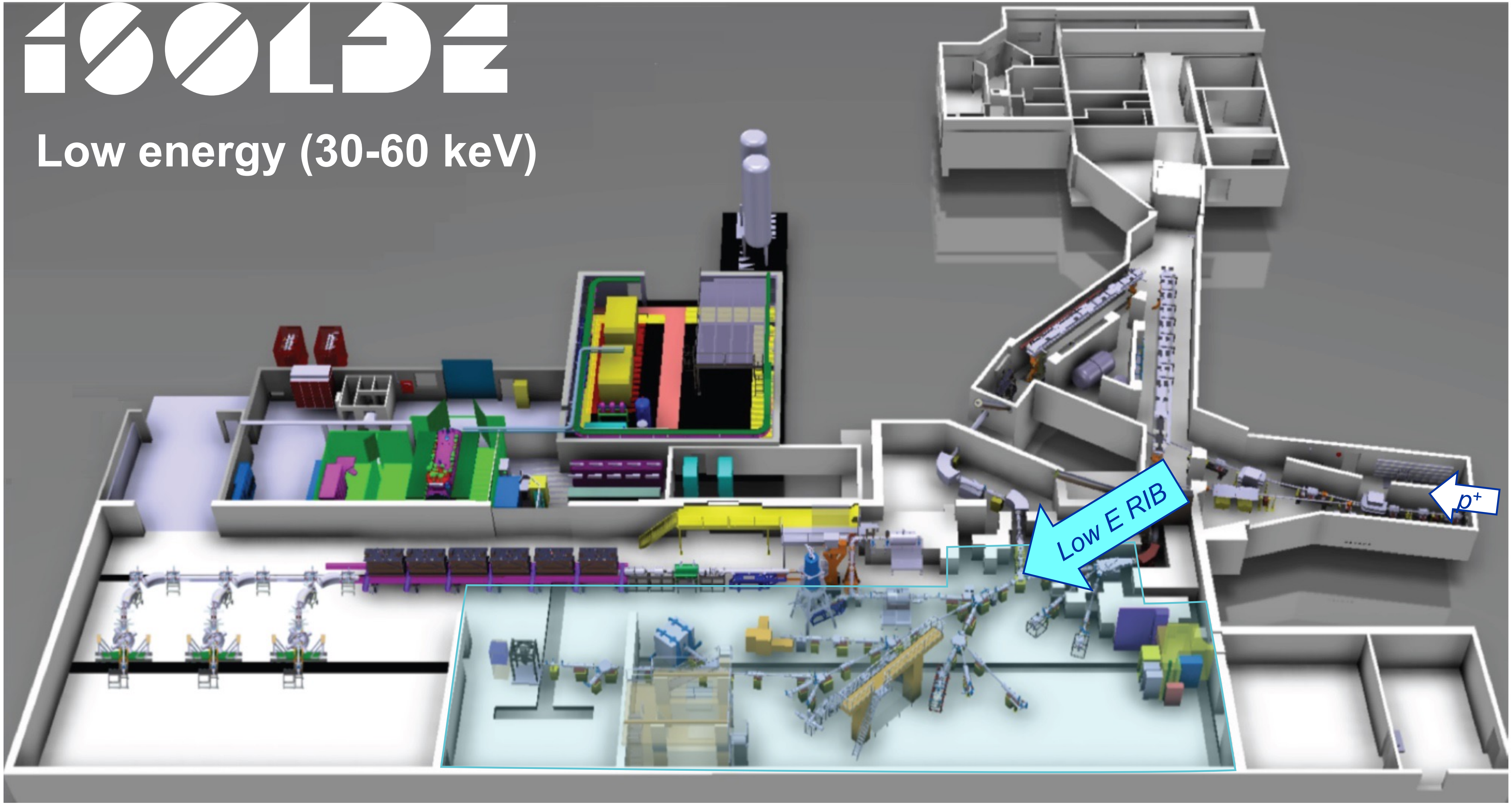
ISOL Step 4: Mass separation

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



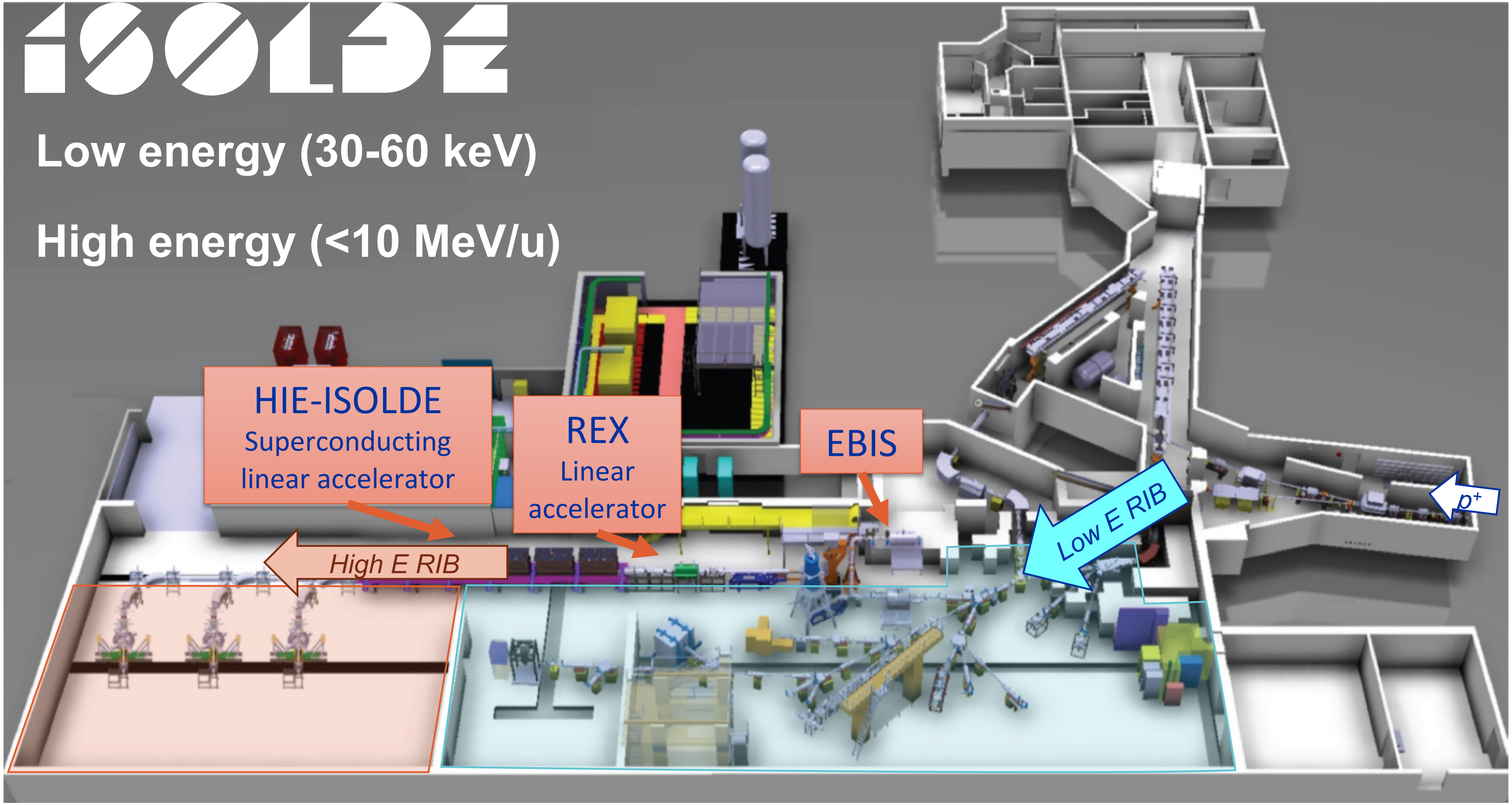
ISOL Step 5: Delivery to Experiments

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



ISOL Step 5: Delivery to Experiments

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



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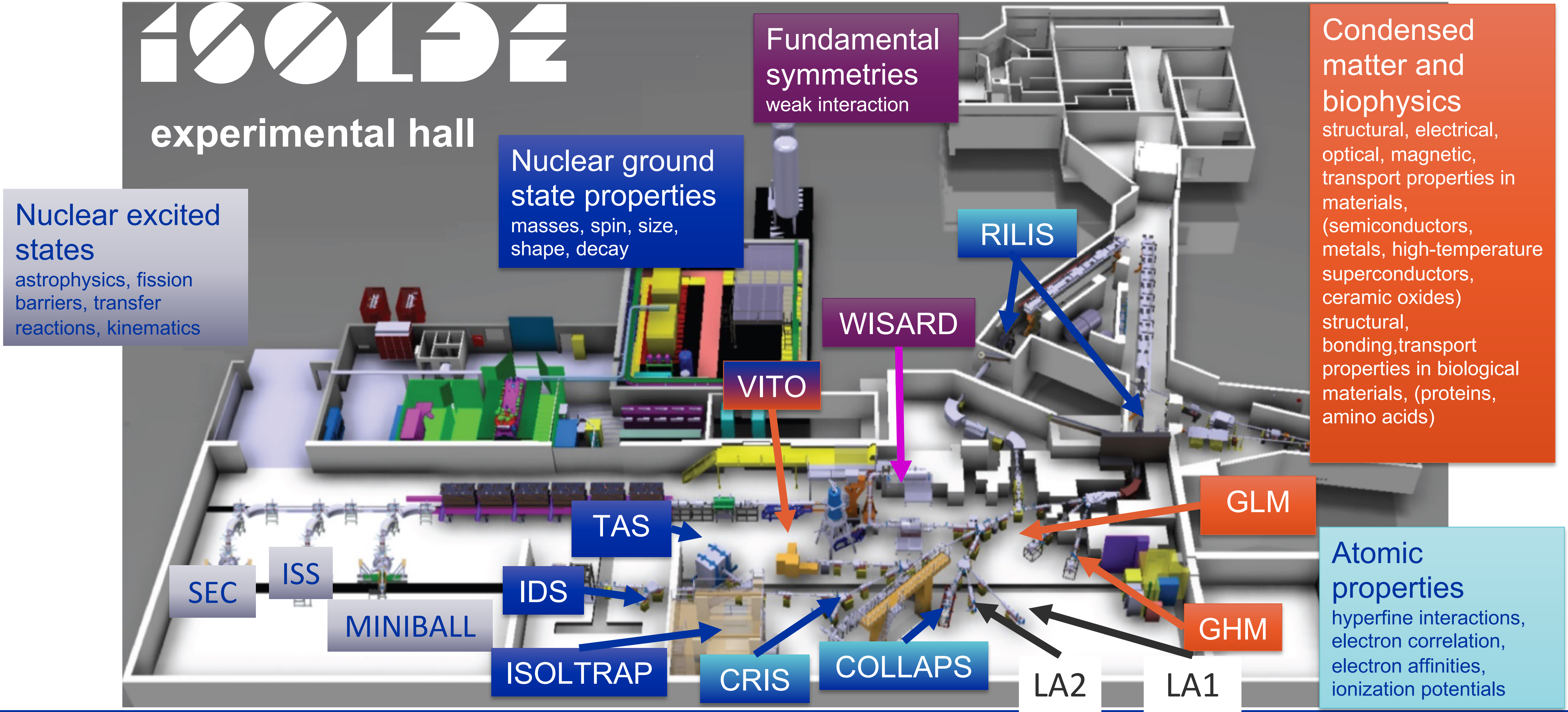
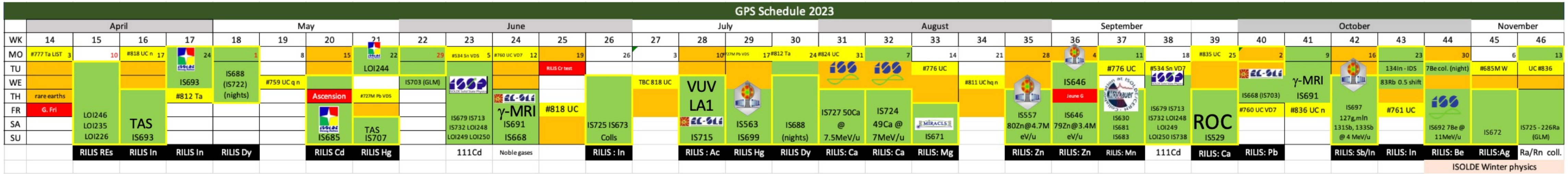
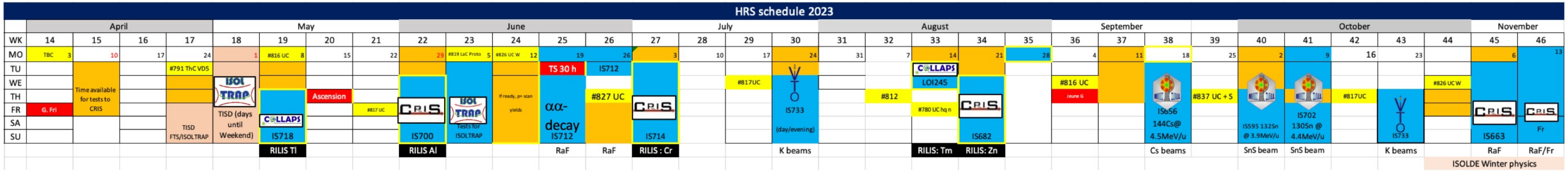
isolde.web.cern.ch



TISD 2023



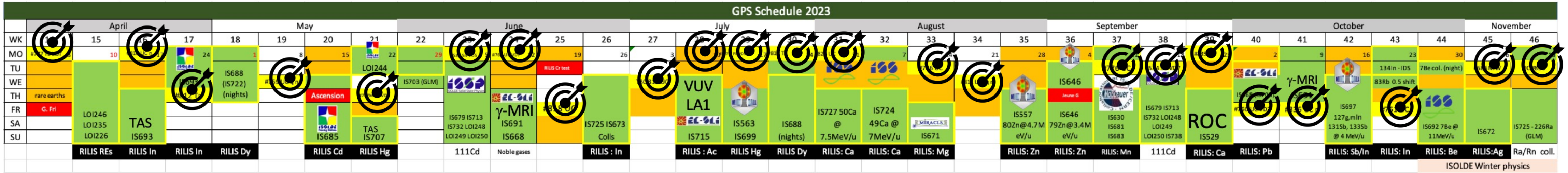


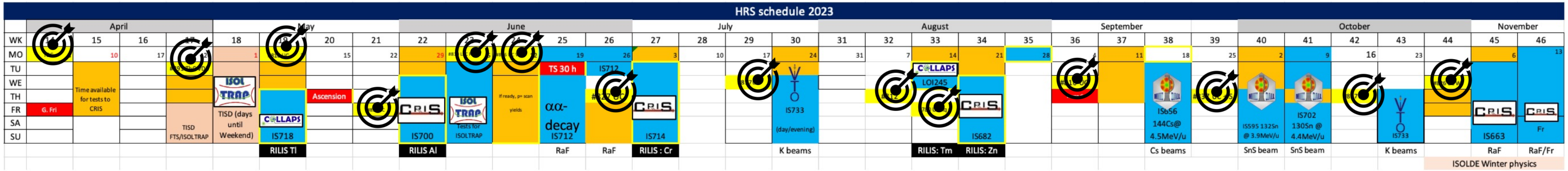
= Yield measurements, proton scans, setup

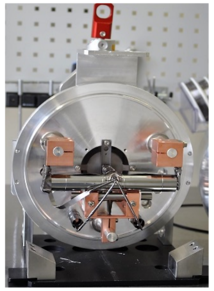
RILIS

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





=  (~ 50 kCHF)

= Yield measurements, proton scans, setup

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

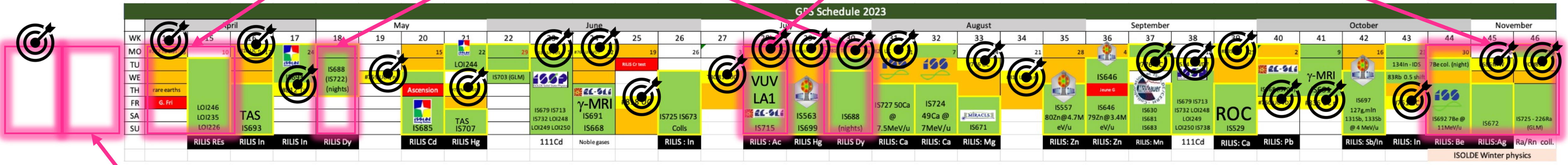
TISD 2023

Back-of-line heating: Dy collections

Batch mode: Ra → Ac

Winter physics: long-lived isotopes + external samples

LIST: lanthanides

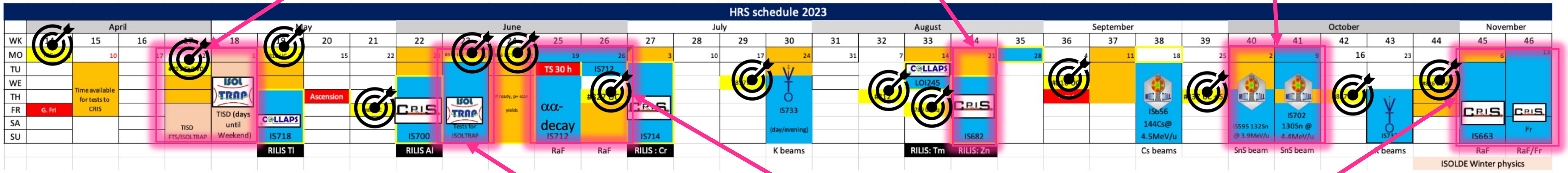


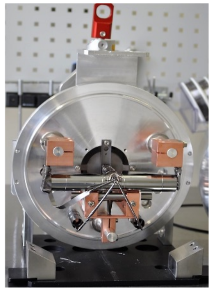
LIST: actinides

ThC$_x$ VD5+CF$_4$

Prototype hot quartz: CRIS Zn

SnS





Prototype target + prototype ion source: ISOLTRAP Cd

RaF online + winter physics now standard beam

= (~ 50 kCHF)

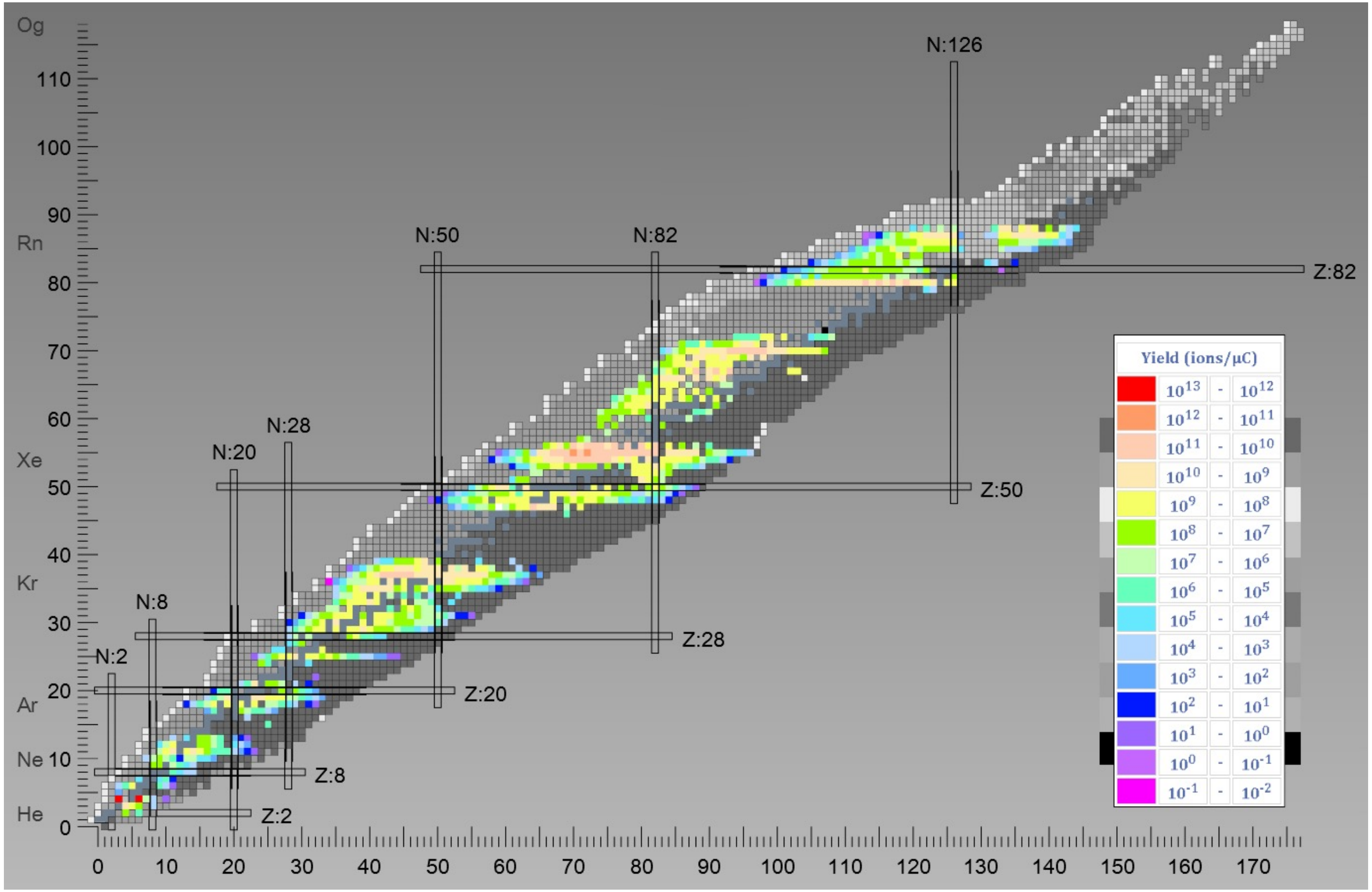
= Yield measurements, proton scans, setup

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

CERN-ISOLDE

>1000 isotopes and isomers

74 elements



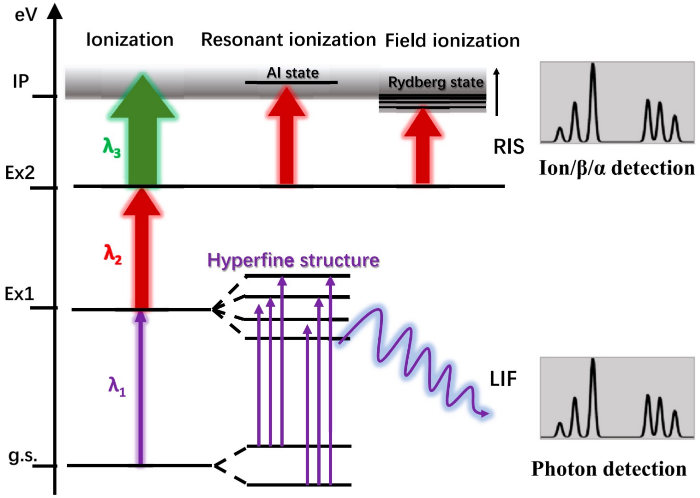
Ballof *et.al*, (2020) *NIM B* **463**, 211-215
cern.ch/isolde-yields

www.nucleonica.com
Dataset: JEFF-3.1 Nuclear Data Library, NEA (2023)

Laser spectroscopy

Collinear

- RIS, LIF



Progress in Particle and Nuclear Physics 129 (2023) 104005

Review

Laser spectroscopy for the study of exotic nuclei

X.F. Yang [a,*], S.J. Wang [a], S.G. Wilkins [b,*], R.F. Garcia Ruiz [b,*]

[a] School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100871, China
[b] Massachusetts Institute of Technology, Cambridge, MA 02139, USA

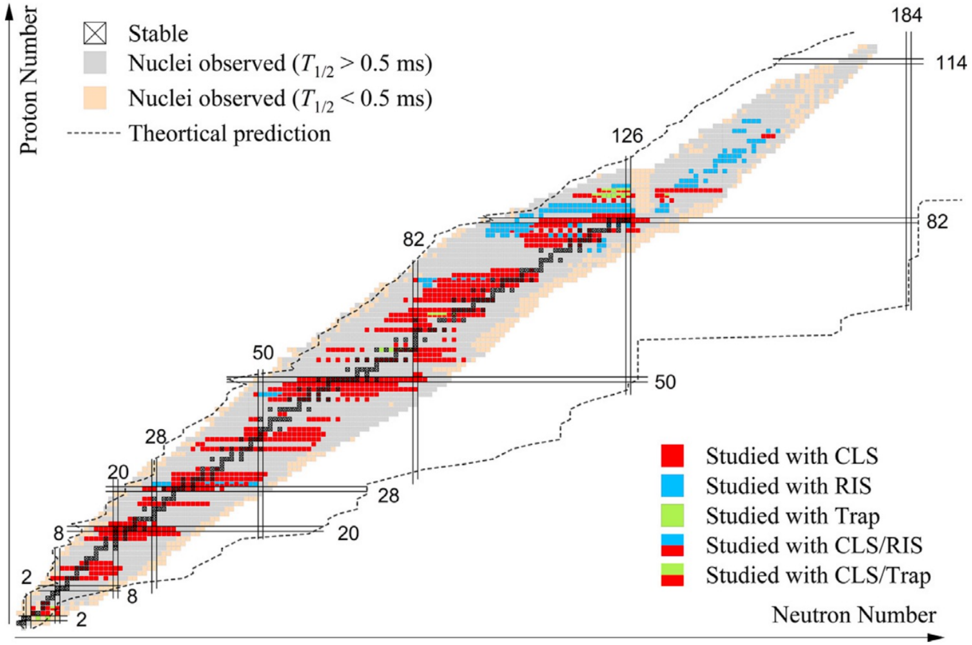
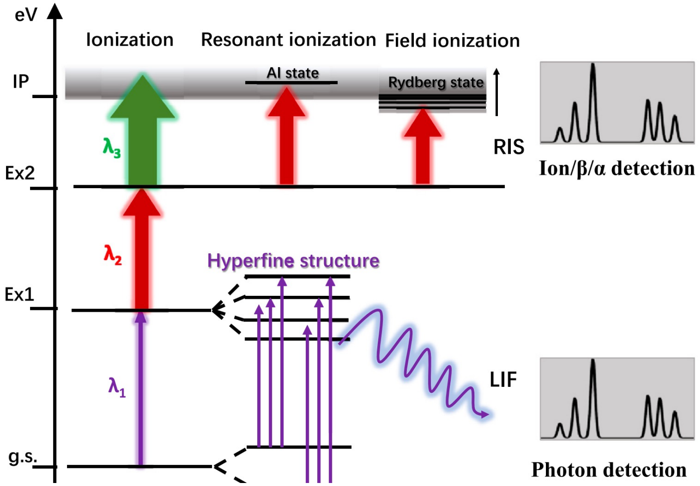


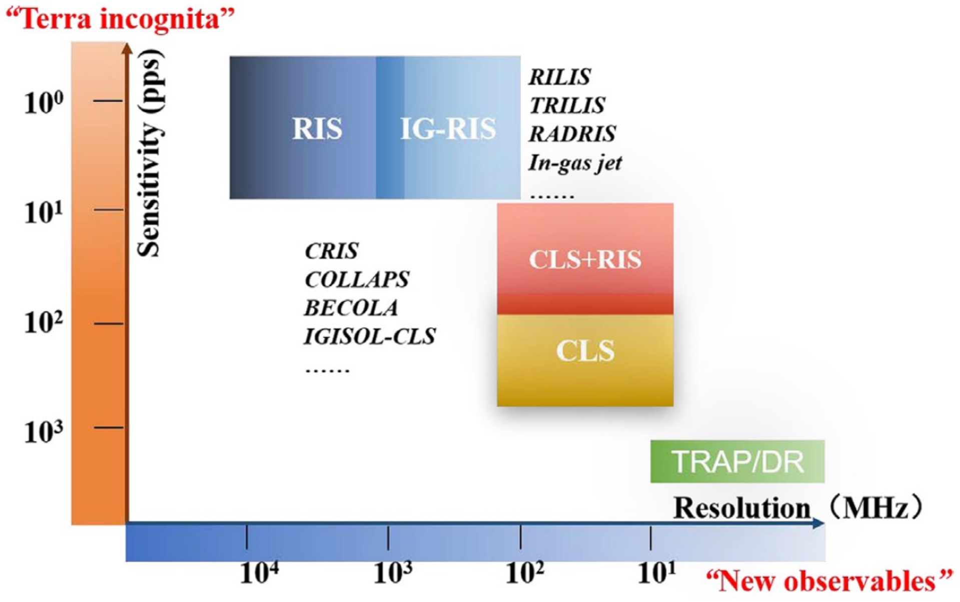
Fig. 1.2. The chart of the nuclides. Stable and long-lived isotopes that exist in large quantities on Earth are indicated in black. The dashed line indicates the region within which bound nuclei are predicted to exist by nuclear theory [65]. Unstable isotopes produced at RIB facilities are shown in gray (for nuclei with $T_{1/2} > 0.5$ ms) and light yellow (for nuclei with $T_{1/2} < 0.5$ ms). Around a thousand ground- and long-lived isomeric states of unstable nuclei have been studied by laser spectroscopy experiments so far, which are indicated with red, blue and green squares, depending on the technique employed.

Laser spectroscopy

Collinear

- RIS, LIF





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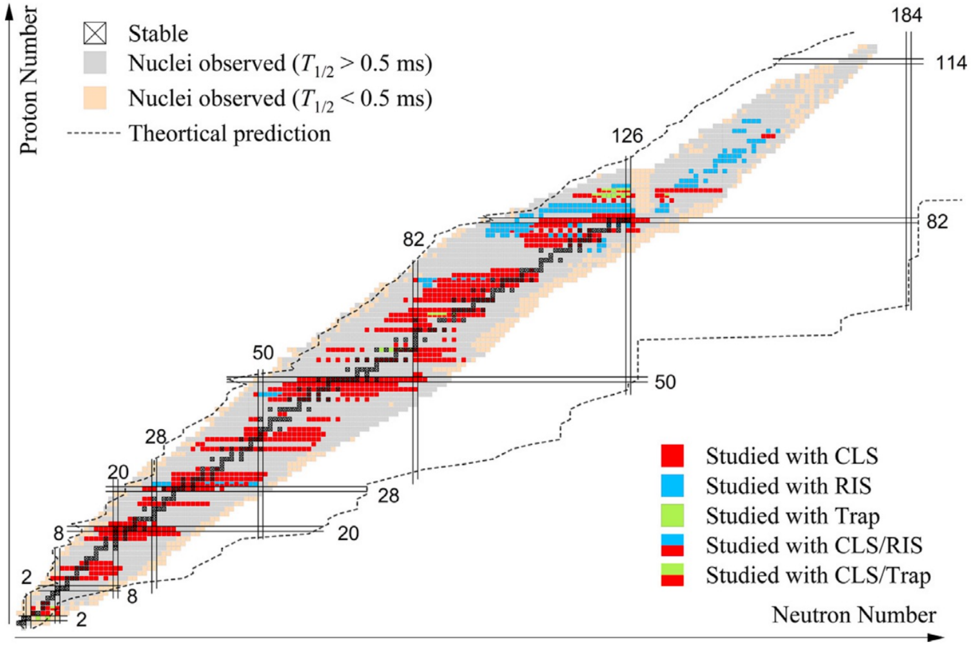

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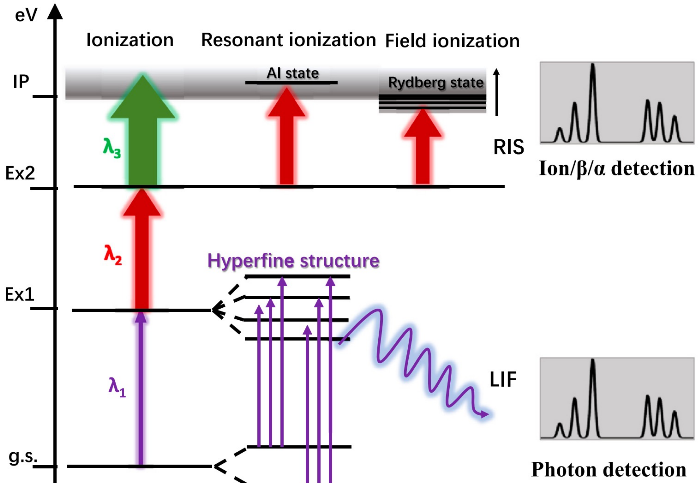
Laser spectroscopy

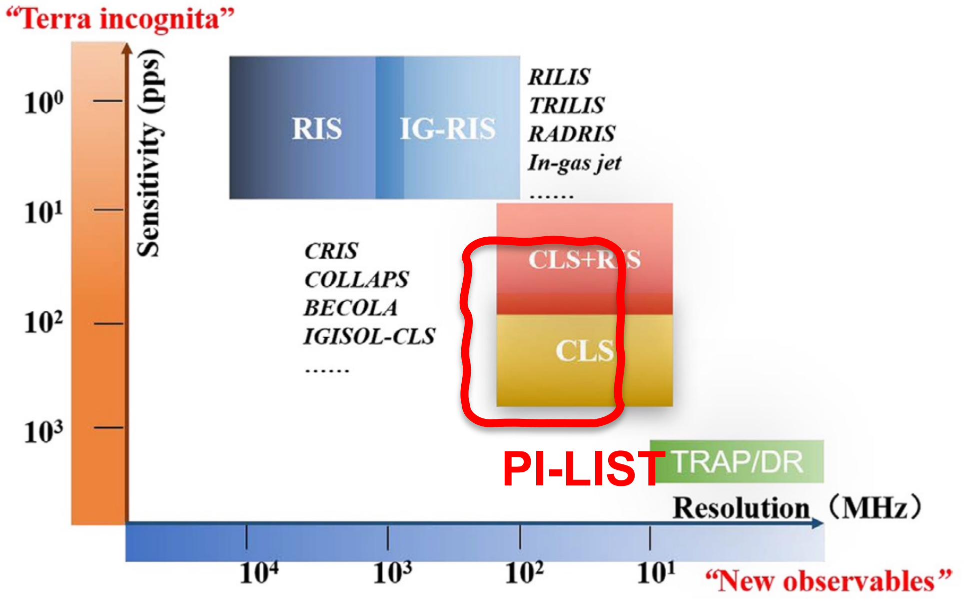
Collinear

- RIS, LIF

In-source

- PI-LIST





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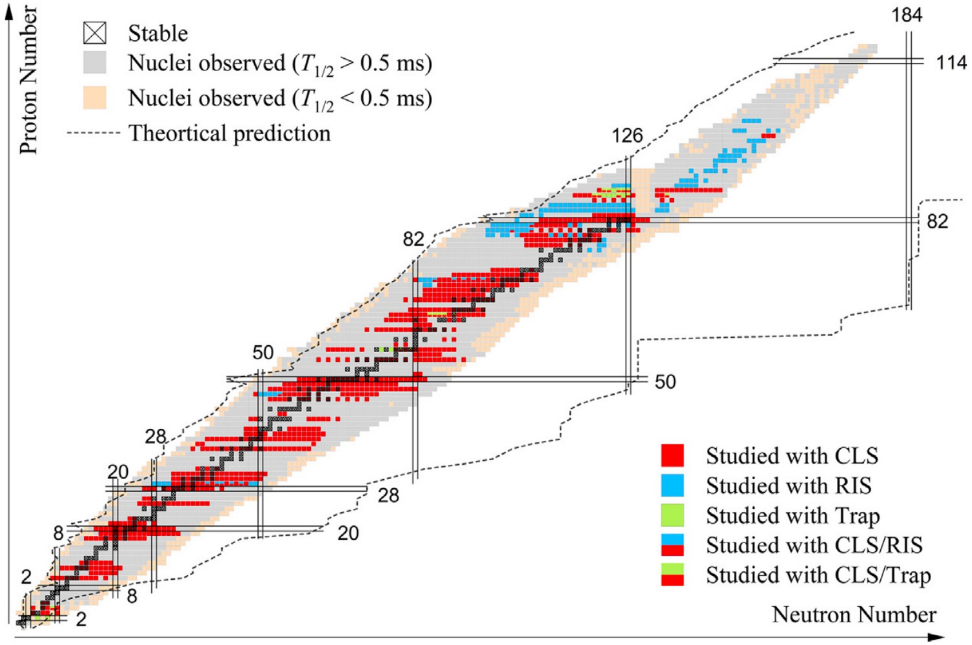

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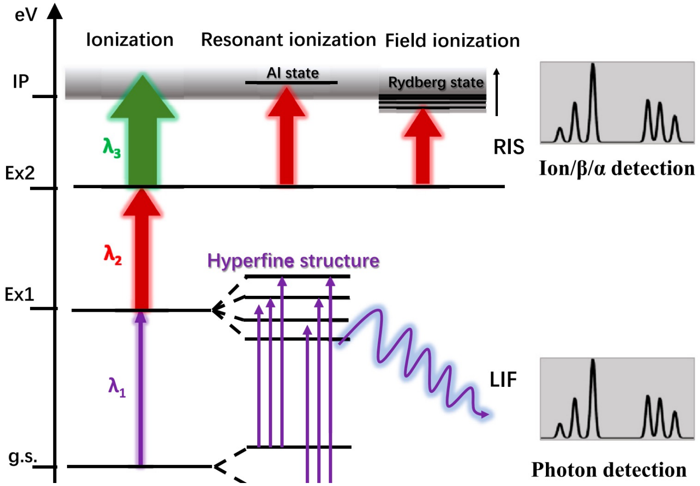

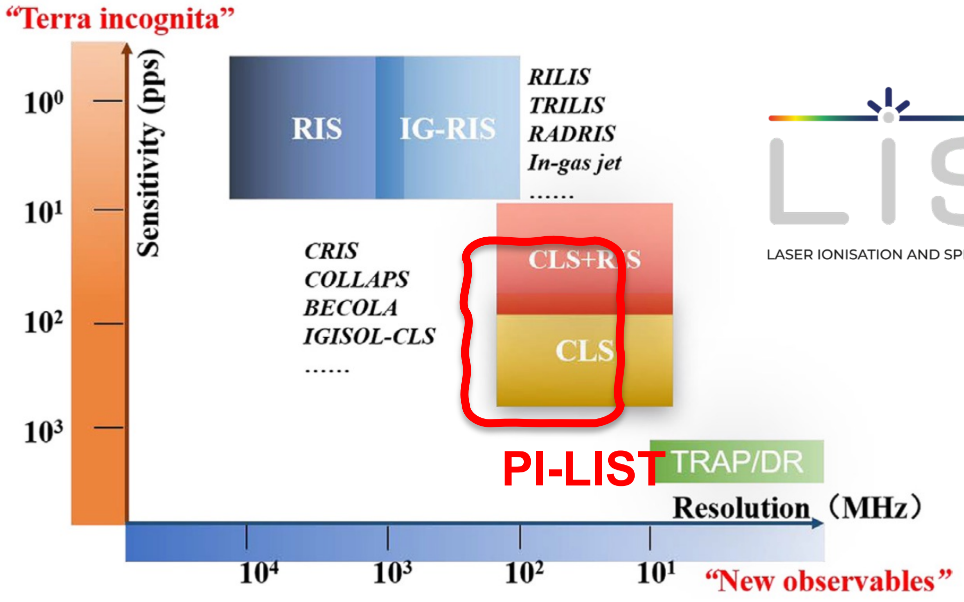

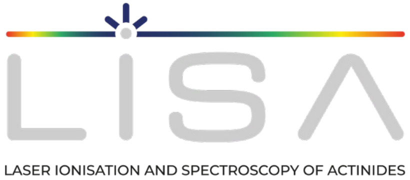

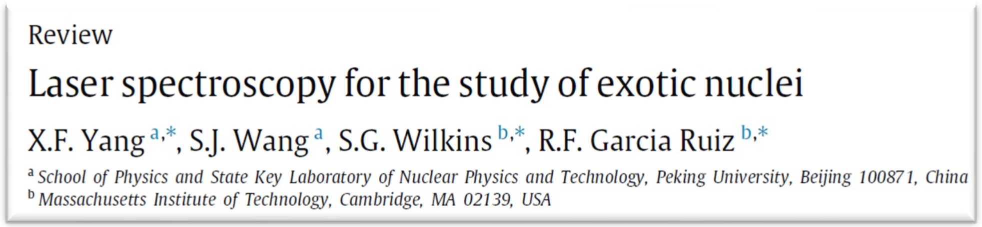
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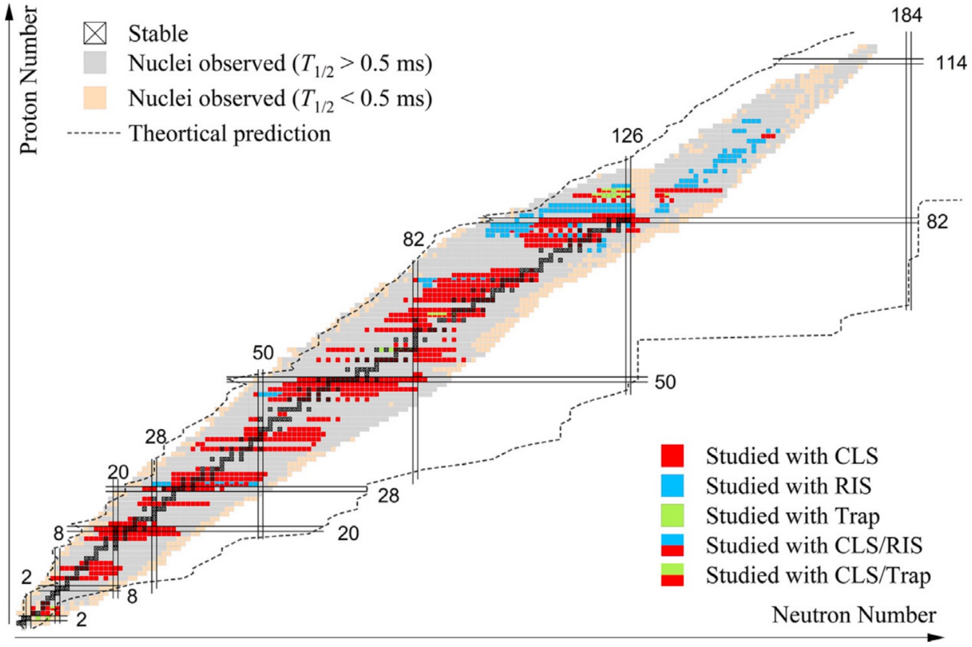

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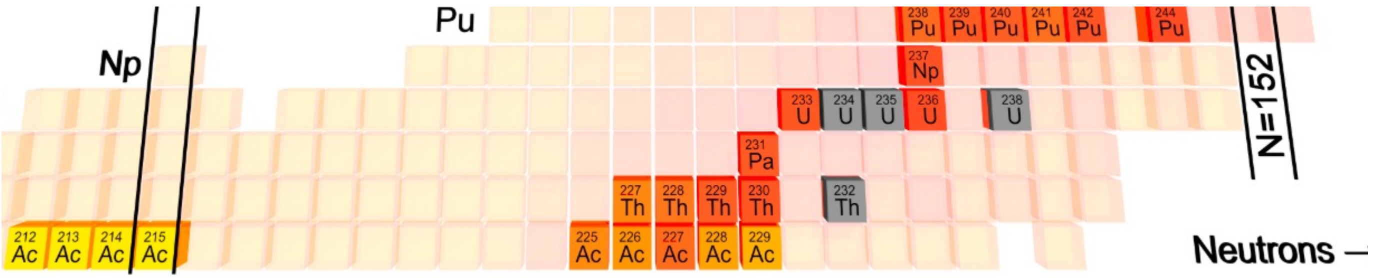
Atomic actinides



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

Up to 2020

Actinium

- Delivered as Ra/Fr: ^{225}Ac [1]
- Laser-ionized: ^{227}Ac [2] IS637, Data under analysis

Now

Actinium

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

Thorium

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

Protactinium

- Laser-ionized: tried, failed

Uranium

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

Neptunium

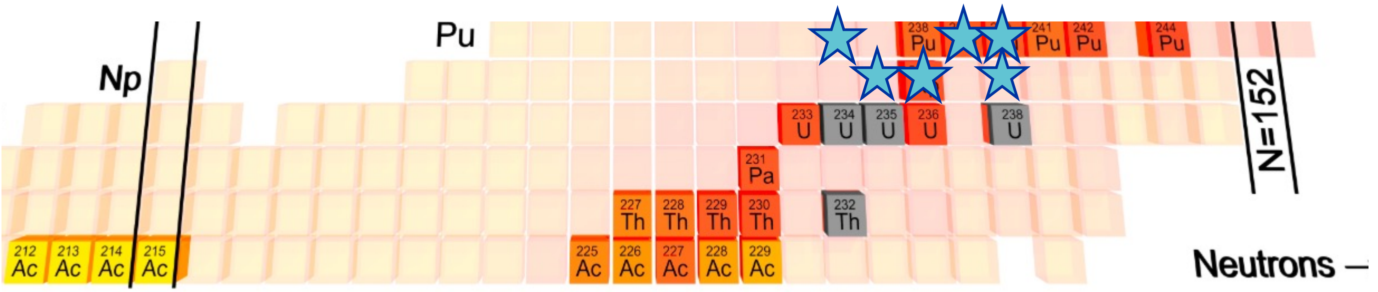
- Laser-ionized: $^{235\text{-}241}$Np

Plutonium

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

[1] Guglielmetti et al, EPJA 12, 383-386 (2002)
[2] Jajčišinova et al, D8.1, Zenodo (2023)
[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 et al, PhD thesis, in preparation (2023)
[9] Au et al, INTC-LOI-243 (2022)

Atomic actinides



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[8] Kaja et al, PhD thesis, in preparation (2023)
[9] Au et al, INTC-LOI-243 (2022)

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

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

M. Au,[1,2,*] M. Athanasakis-Kaklamanakis,[1,3] L. Nies,[1,4] R. Heinke,[1] K. Chrysalidis,[1] U. Köster,[1,5] P. Kunz,[6] B. Marsh,[1] M. Mougeot,[1,7,†] L. Schweikhard,[4] S. Stegemann,[1] Y. Vila Gracia,[1] Ch. E. Düllmann,[2,8,9] and S. Rothe[1]

[1] *European Organization for Nuclear Research (CERN), Meyrin, 1211 Geneva, Switzerland*
[2] *Department of Chemistry, Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany*
[3] *Katholieke Universiteit Leuven, Instituut voor Kern-en Stralingsfysica, B-3001 Leuven, Belgium*
[4] *Institut für Physik, University of Greifswald, 17489 Greifswald, Germany*
[5] *Institut Laue-Langevin, 38000 Grenoble, France*
[6] *TRIUMF, Vancouver, Canada V6T 2A3*
[7] *Max Planck Institut für Kernphysik, 69117 Heidelberg, Germany*
[8] *GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany*
[9] *Helmholtz Institute Mainz, 55099 Mainz, Germany*

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Atomic actinides

Contents lists available at ScienceDirect

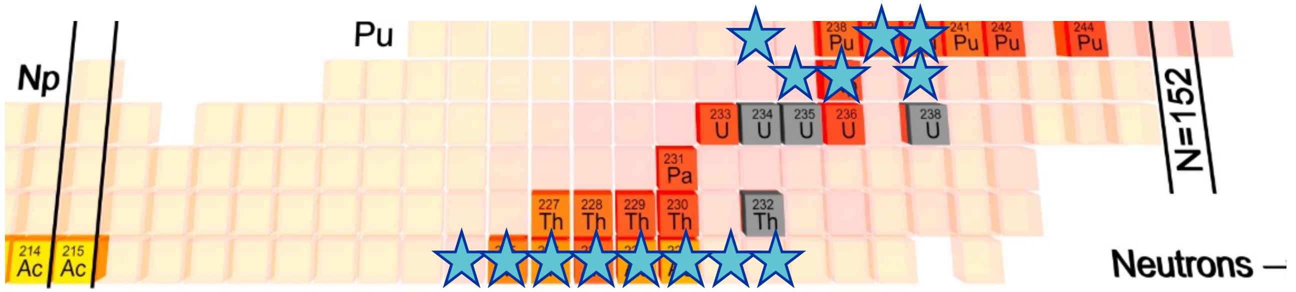
Nuclear Inst. and Methods in Physics Research, B

journal homepage: www.elsevier.com/locate/nimb

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 [a], Thomas E. Cocolios [c], Valentin N. Fedosseev [a], Isabel Hendriks [a,d], Asar A.H. Jaradat [a], Magdalena Kaja [e], Tom Kieck [f,g], Tobias Kron [e], Ralitsa Mancheva [a,c], Bruce A. Marsh [a], Stefano Marzari [a], Sebastian Raeder [f,g], Sebastian Rothe [a], Dominik Studer [f,g], Felix Weber [e], Klaus Wendt [e]

[a] STI group, SY department, CERN, Switzerland
[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
[f] GSI Helmholtzzentrum für Schwerionenforschung, Germany
[g] Helmholtz Institute Mainz, Germany



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

Ac [3]

:

Neptunium

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

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

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

M. Au [1,2,*] M. Athanasakis-Kaklamanakis [1,3] L. Nies [1,4] R. Heinke [1] K. Chrysalidis [1] U. Köster [1,5] P. Kunz [6] B. Marsh [1] M. Mougeot [1,7,†] L. Schweikhard [4] S. Stegemann [1] Y. Vila Gracia [1] Ch. E. Düllmann [2,8,9] and S. Rothe [1]

[1] *European Organization for Nuclear Research (CERN), Meyrin, 1211 Geneva, Switzerland*
[2] *Department of Chemistry, Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany*
[3] *Katholieke Universiteit Leuven, Instituut voor Kern-en Stralingsfysica, B-3001 Leuven, Belgium*
[4] *Institut für Physik, University of Greifswald, 17489 Greifswald, Germany*
[5] *Institut Laue-Langevin, 38000 Grenoble, France*
[6] *TRIUMF, Vancouver, Canada V6T 2A3*
[7] *Max Planck Institut für Kernphysik, 69117 Heidelberg, Germany*
[8] *GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany*
[9] *Helmholtz Institute Mainz, 55099 Mainz, Germany*

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

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

Protactinium

- Laser-ionized: tried, failed

Uranium

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

[1] Guglielmetti et al, EPJA 12, 383-386 (2002)
[2] Jajčišinova et al, D8.1, Zenodo (2023)
[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 et al, PhD thesis, in preparation (2023)
[9] Au et al, INTC-LOI-243 (2022)

Atomic actinides

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, B

journal homepage: www.elsevier.com/locate/nimb

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

Reinhard Heinke [a,*], Mia Au [a,b], Cyril Ber
Valentin N. Fedosseev [a], Isabel Hendriks
Tobias Kron [e], Ralitsa Mancheva [a,c], Bruc
Sebastian Rothe [a], Dominik Studer [f,g], Fe

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[c] Institute for Nuclear and Radiation Physics, KU Leuven, Belgium
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[f] GSI Helmholtzzentrum für Schwerionenforschung, Germany
[g] Helmholtz Institute Mainz, Germany

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

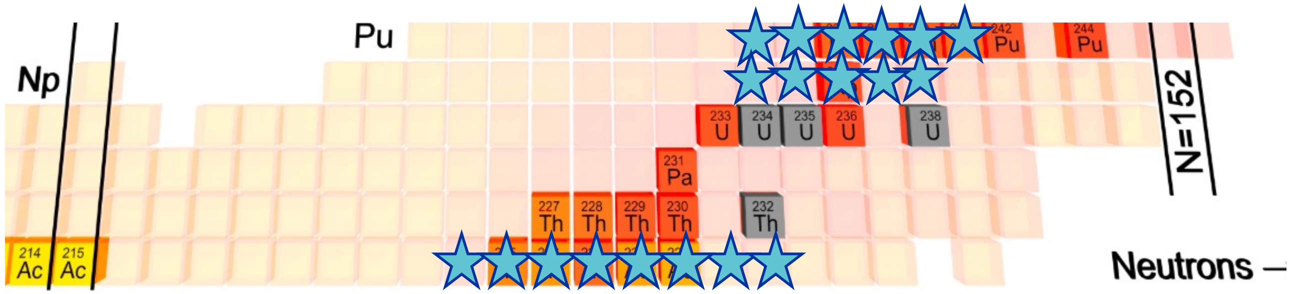
May 13, 2022

Mia Au[1,2], Anastasia Borschevsky[3], Katerina Chrysalidis[1], Raphaël Crosa-Rossa[3], Christoph Düllmann[2,4,5], Reinhard Heinke[1], Asar Jaradat[1], Magdalena Kaja[2], Bruce Marsh[1], Iain Moore[6], Andrea Raggio[6], Sebastian Rothe[1], Simon Stegemann[1], Darcy van Eerten[7], Clemens Walther[7]

[1] SY-STI, CERN, Switzerland
[2] Johannes Gutenberg-Universität Mainz, Germany
[3] Rijksuniversiteit Groningen, Groningen, Netherlands
[4] GSI Helmholtzzentrum für Schwerionenforschung, Germany
[5] Helmholtz Institute Mainz, Germany
[6] University of Jyväskylä, Finland
[7] IRS, 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

March 2023



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

Neptunium

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

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

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

M. Au,[1,2,*] M. Athanasakis-Kaklamanakis,[1,3] L. Nies,[1,4] R. Heinke,[1] K. Chrysalidis,[1] U. Köster,[1,5] P. Kunz,[6] B. Marsh,[1] M. Mougeot,[1,7,†] L. Schweikhard,[4] S. Stegemann,[1] Y. Vila Gracia,[1] Ch. E. Düllmann,[2,8,9] and S. Rothe[1]

[1] European Organization for Nuclear Research (CERN), Meyrin, 1211 Geneva, Switzerland
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[3] Katholieke Universiteit Leuven, Instituut voor Kern-en Stralingsfysica, B-3001 Leuven, Belgium
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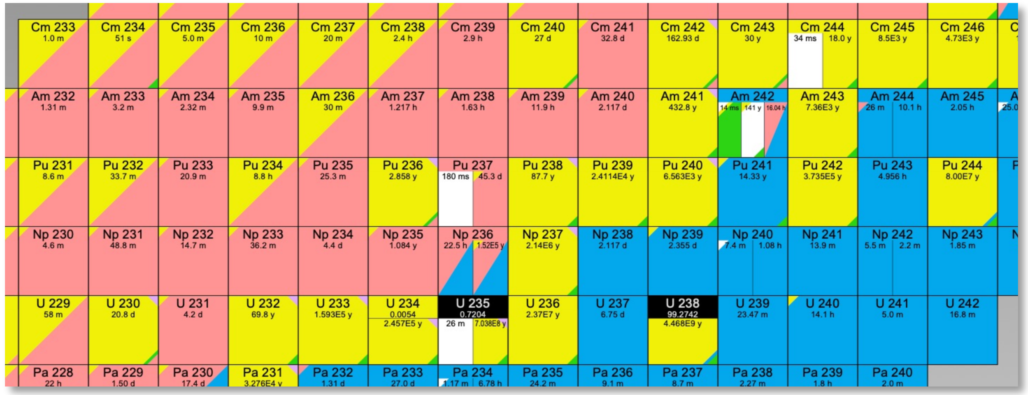
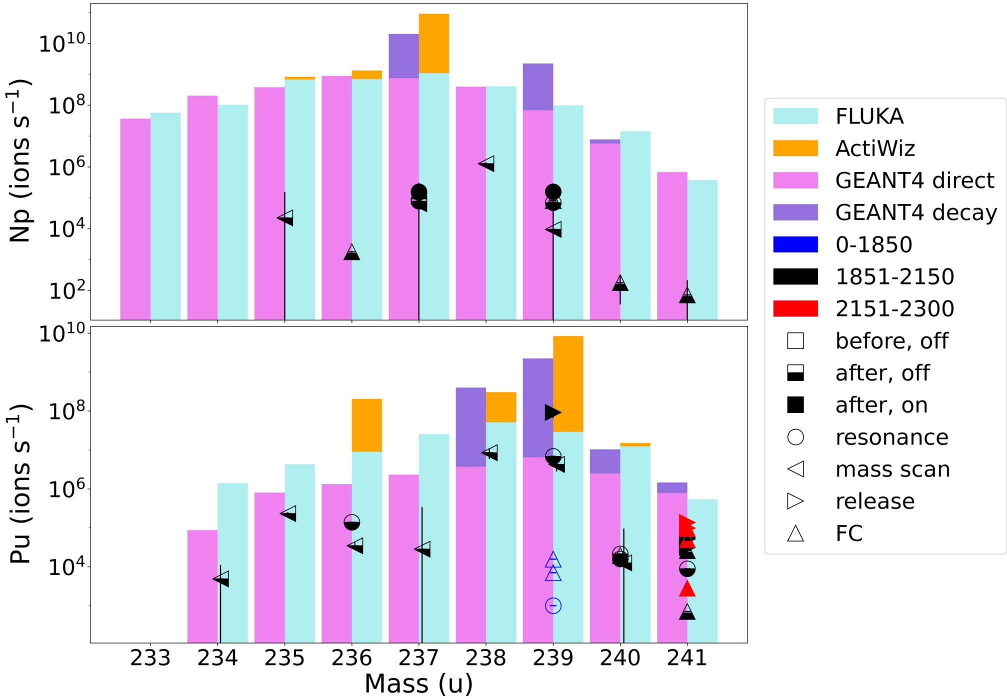
[1] Guglielmetti et al, EPJA 12, 38
[2] Jajčišinova et al, D8.1, Zenod
[3] Andreyev et al, INTC-LOI-216
[4] Heinke et al, NIM B 541, 8-12
[5] Heinke, Jaradat, Zenodo, 782
[6] Kraemer et al, Nature 617, 70
[7] Au et al, NIM B 541, 375-379
[8] Kaja et al, PhD thesis, in prep
[9] Au et al, INTC-LOI-243 (2022)

Transuranium atomic ion beams





Open Access

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

M. Au, M. Athanasakis-Kaklamanakis, L. Nies, R. Heinke, K. Chrysalidis, U. Köster, P. Kunz, B. Marsh, M. Mougeot, L. Schweikhard, S. Stegemann, Y. Vila Gracia, Ch. E. Düllmann, and S. Rothe

Phys. Rev. C **107**, 064604 – Published 8 June 2023

TABLE I. Production mechanisms in % of total events for selected nuclides of interest calculated using GEANT4 QGSP_INCLXX+ABLA with 10^9 1.4-GeV proton primaries. "Inelastic" and "Decay" columns give the sums over inelastic and radioactive decay processes, respectively. The larger contribution is indicated in bold. Various capture reactions are included in the model but not shown in the table. Some processes with event fractions below 1 % (e.g., photonuclear reactions) are not shown and only the dominant parent nucleus and its corresponding percentage fraction of total events are given. The number of total events is scaled from 10^9 protons to obtain the nuclides per μC equivalent.

isotope	Inelastic:	p	n	d	t	^{3}He	α	ions	Decay:	parent		nuclides/μC
^{234}U	36.4	28.4	7.2	0.4	-	-	-	-	**63.6**	^{234}Pa	59.4	6.7×10^9
^{236}U[a]	**65.5**	52.2	11.8	0.8	-	-	-	-	34.3	^{236}Pa	34.3	1.3×10^{10}
^{237}U	**70.5**	61.7	7.4	0.9	0.1	-	-	-	29.0	^{237}Pa	29.0	1.9×10^{10}
^{239}U[a]	1.9	-	-	1.3	0.5	-	0.1	-	-	-	-	2.2×10^9
^{240}U	**100.0**	-	-	-	85.7	-	11.9	2.4	-	-	-	2.1×10^6
^{231}Np	**99.0**	93.1	-	3.5	0.5	-	-	-	1.0	^{231}Pu	1.0	1.3×10^6
^{232}Np	**100.0**	92.3	0.3	5.4	0.3	0.1	-	-	-	-	-	1.3×10^7
^{233}Np	**99.8**	91.1	0.2	6.1	0.5	0.1	-	-	0.2	^{233}Pu	0.2	3.6×10^7
^{234}Np	**100.0**	89.9	0.2	7.7	0.7	0.1	-	-	-	-	-	2.0×10^8
^{235}Np	**99.8**	88.4	0.2	8.9	1.0	0.2	-	-	0.2	^{235}Pu	0.2	3.8×10^8
^{236}Np	**100.0**	86.2	0.1	11.1	1.4	0.2	-	-	-	-	-	8.7×10^8
^{237}Np	3.7	3.0	-	0.6	0.1	-	-	-	**96.3**	^{237}U	96.3	2.0×10^{10}
^{238}Np	**100.0**	61.0	-	27.4	8.8	1.1	0.4	-	-	-	-	3.9×10^8
^{239}Np	3.0	-	-	2.2	0.6	0.2	0.1	-	**97.0**	^{239}U	97.0	2.2×10^9
^{240}Np	**73.0**	-	-	-	47.8	5.2	18.8	1.3	27.0	^{240}U	27.0	7.8×10^6
^{241}Np	**100.0**	-	-	-	-	-	96.3	3.7	-	-	-	6.8×10^5
^{235}Pu	**99.2**	62.5	-	-	-	18.8	10.9	-	0.8	^{235}Am	0.8	8.0×10^5
^{236}Pu	**97.1**	67.8	-	-	-	13.5	12.5	-	2.9	^{236}Np	1.9	1.3×10^6
^{237}Pu	**99.5**	39.7	-	-	-	15.5	39.7	0.3	0.5	-	-	2.3×10^6
^{238}Pu	0.9	0.1	-	-	-	0.1	0.7	-	**99.1**	^{238}Np	99.1	4.0×10^8
^{239}Pu	0.3	-	-	-	-	-	0.3	-	**99.7**	^{239}Np	99.7	2.2×10^9
^{240}Pu	24.0	-	-	-	-	2.5	20.9	0.6	**76.0**	^{240}Np	75.5	1.0×10^7
^{241}Pu	**53.9**	-	-	-	-	-	51.7	2.2	46.1	^{241}Np	46.1	1.4×10^6

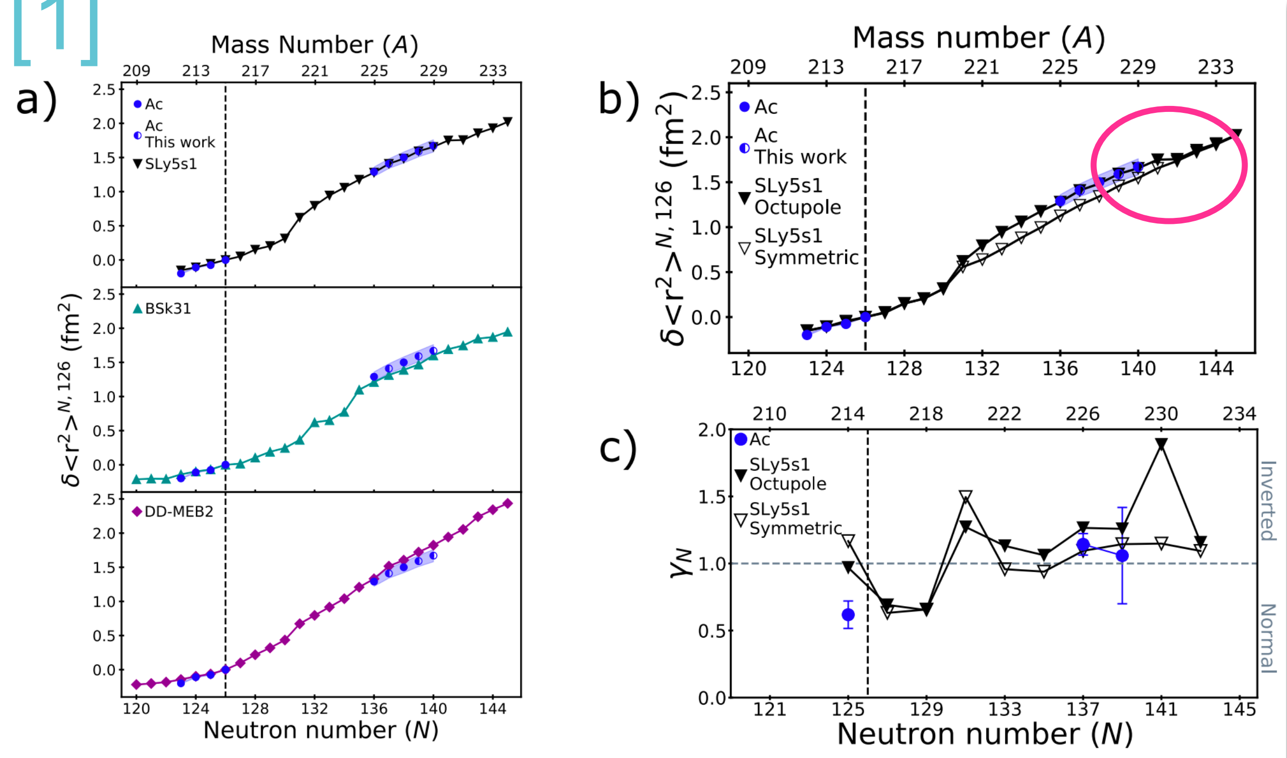
[a] ^{236,239}U are 0.2 % and 98.1 % produced by neutron capture reactions, respectively.

Actinium

PI-LIST [2]

- Octupole deformation

[1]

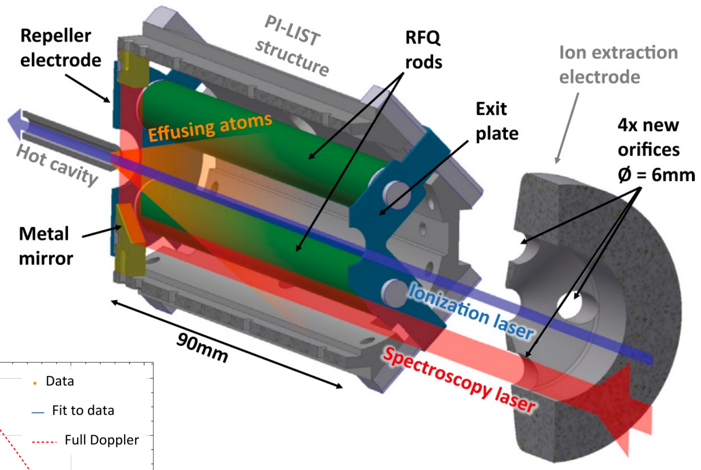


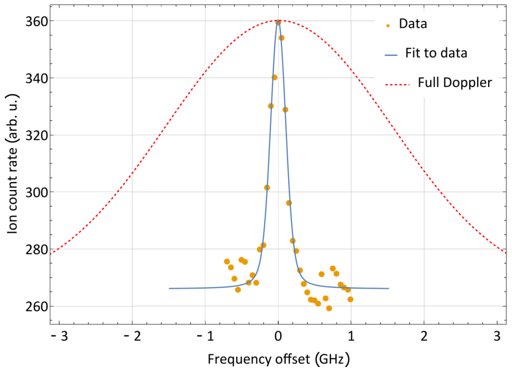
[1] Verstraelen et al., Phys. Rev. C. 100, 044321 (2019)
[2] Heinke et al. (2023) *NIM B.* **541** (8-12)
[3] Heinke et al., CERN-INTC-2020-029, INTC-P-556, https://cds.cern.ch/record/2717945 (2020)
[4] Heinke *et al.,* in preparation (2024)

Actinium

PI-LIST [2]

- Octupole deformation





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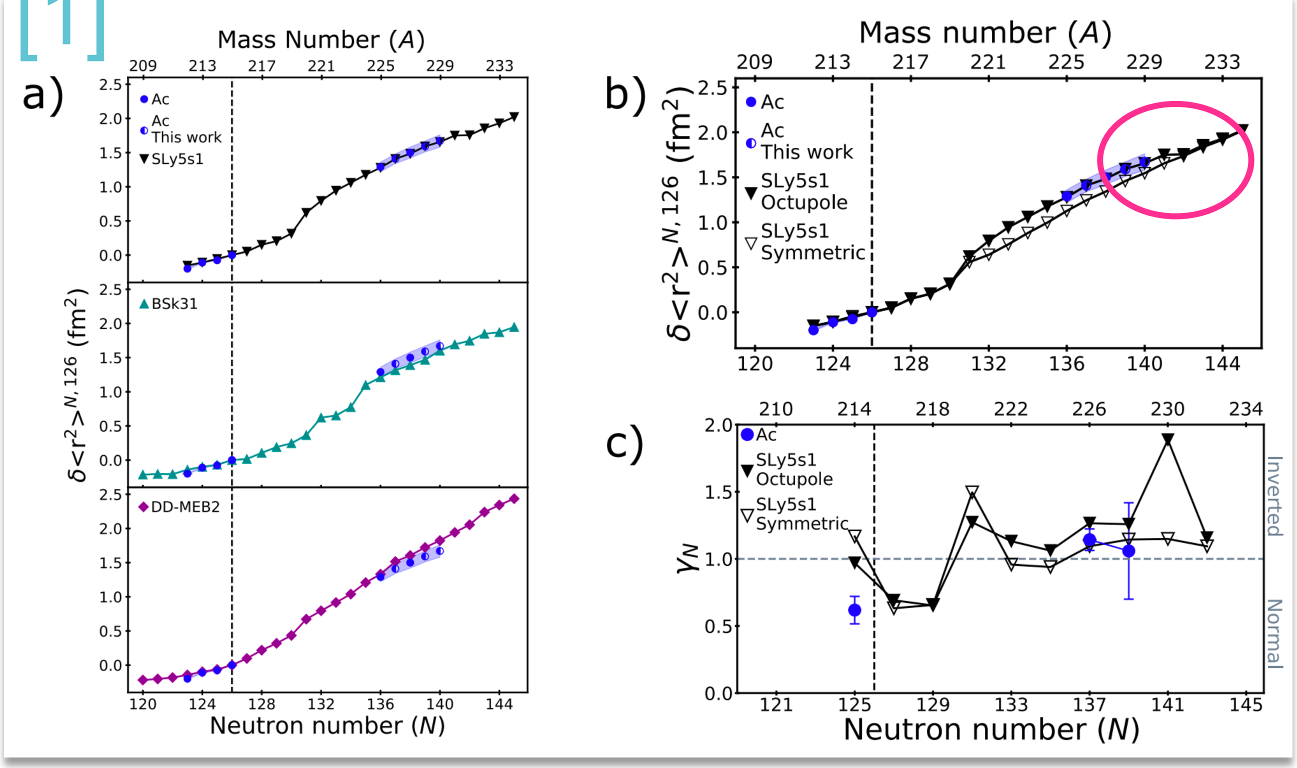

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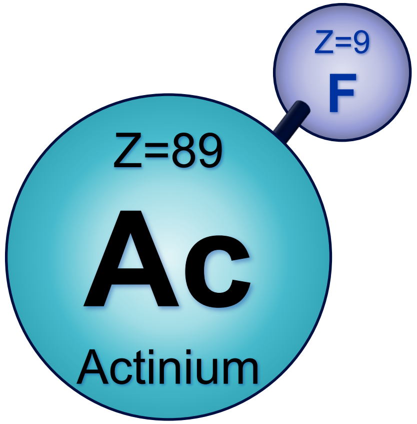
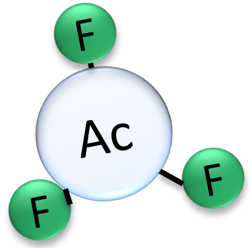
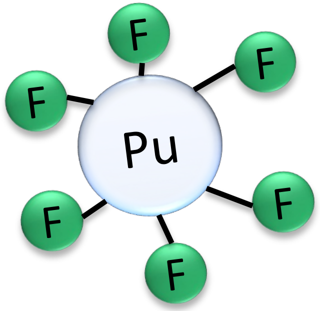
PRELIMINARY [3,4]

Image published in EP Newsletter, CERN (2020)



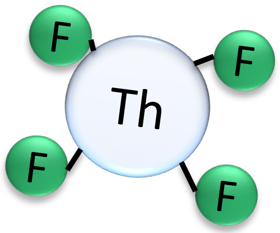


bp 3250˚C

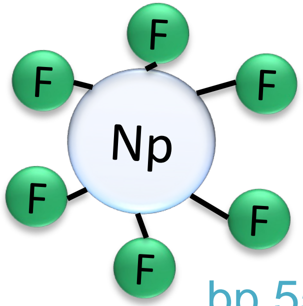


bp 62˚C

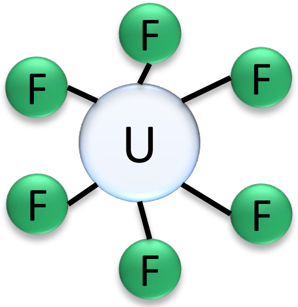
Radioactive molecular beams



bp 1680˚C

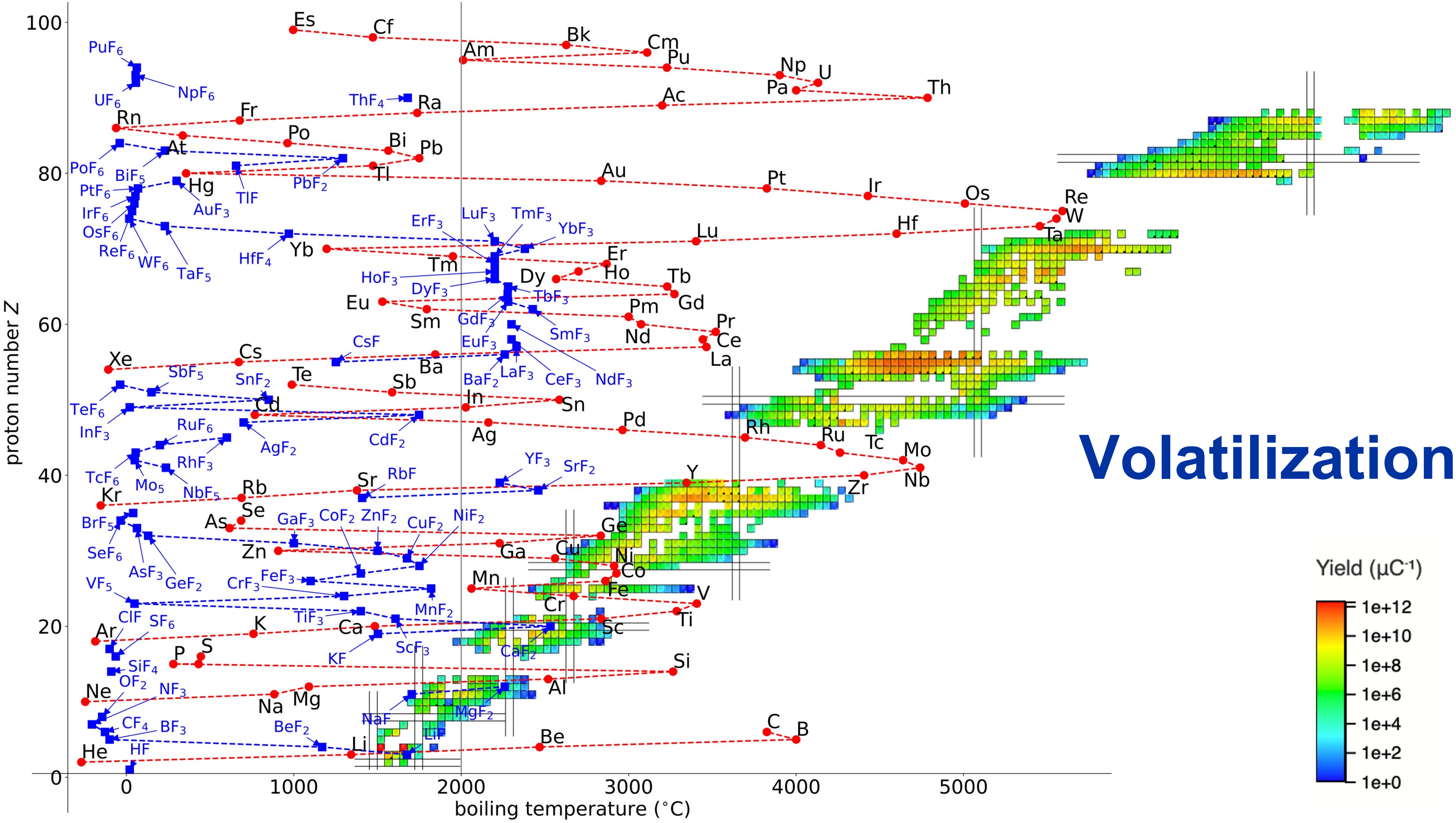


bp 55˚C

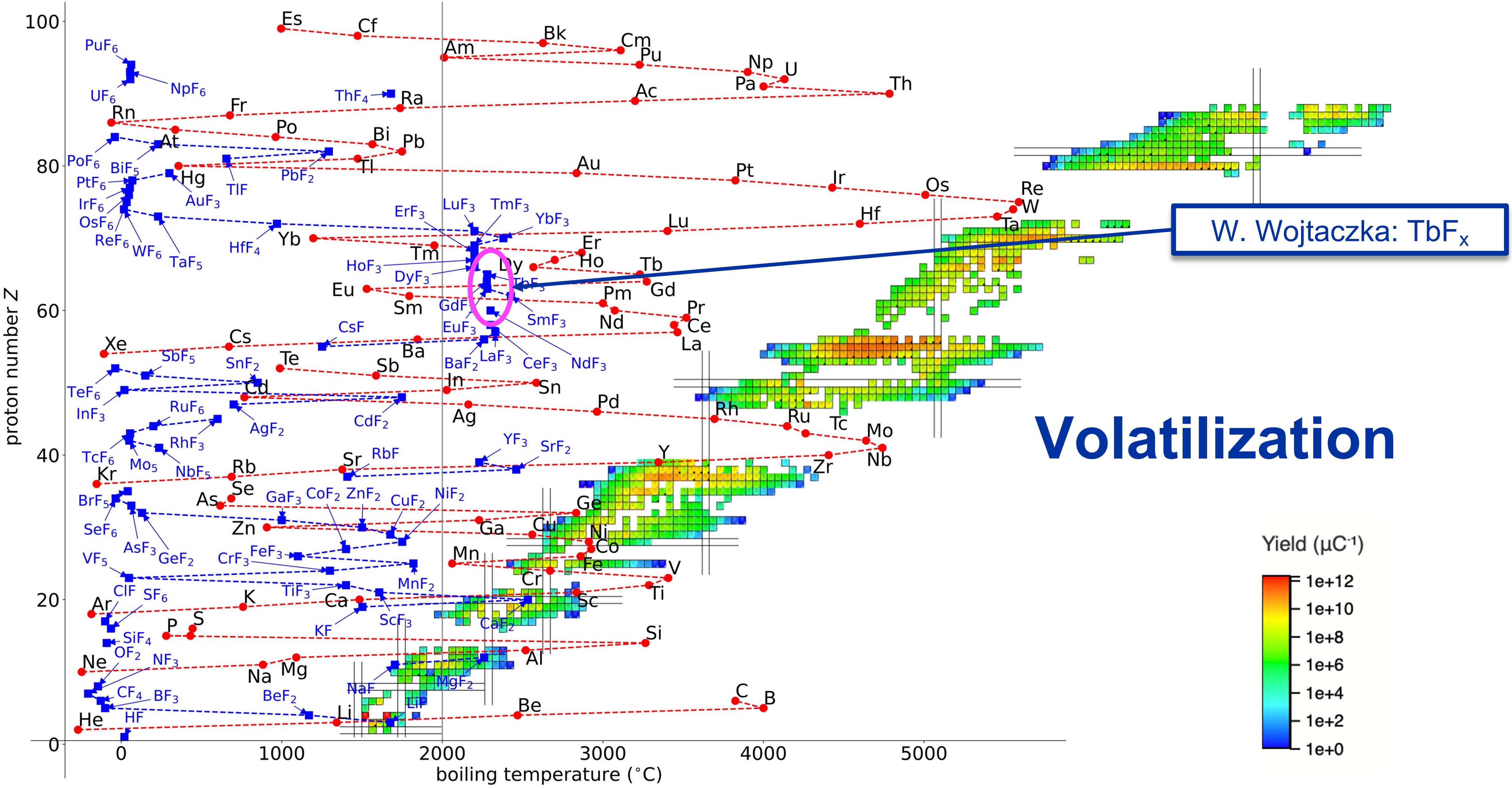


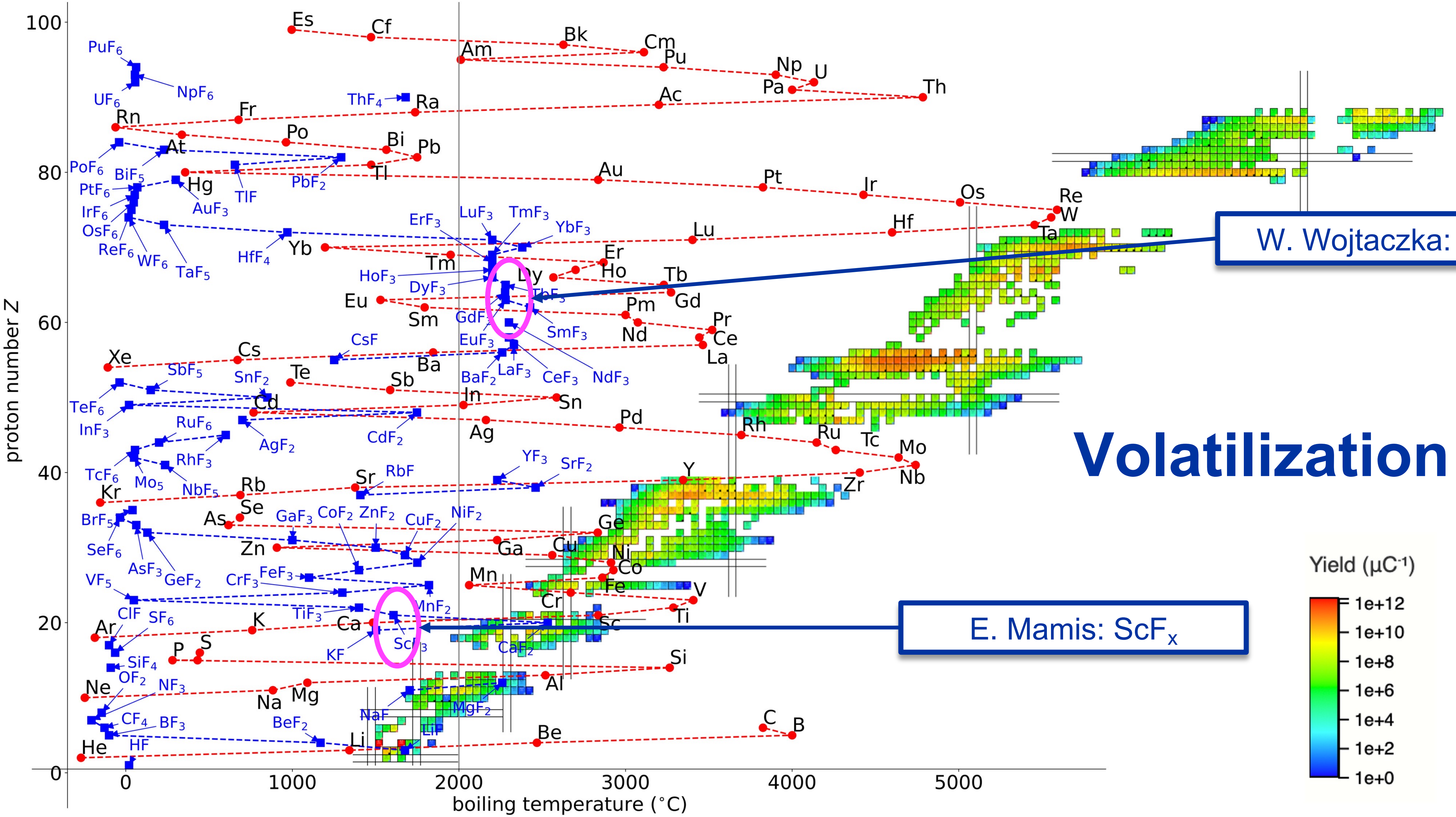
bp 56.5˚C

Volatilization

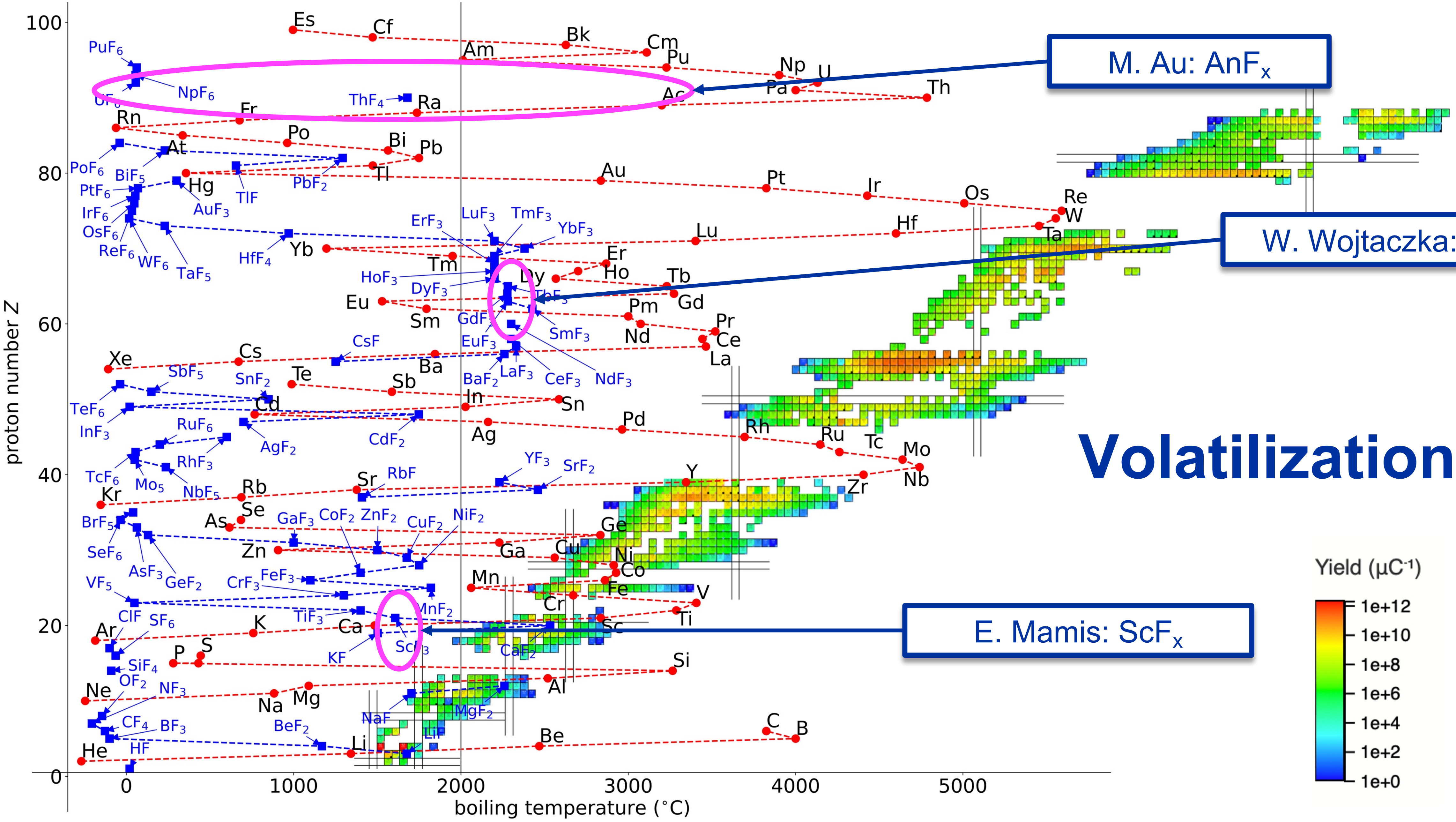


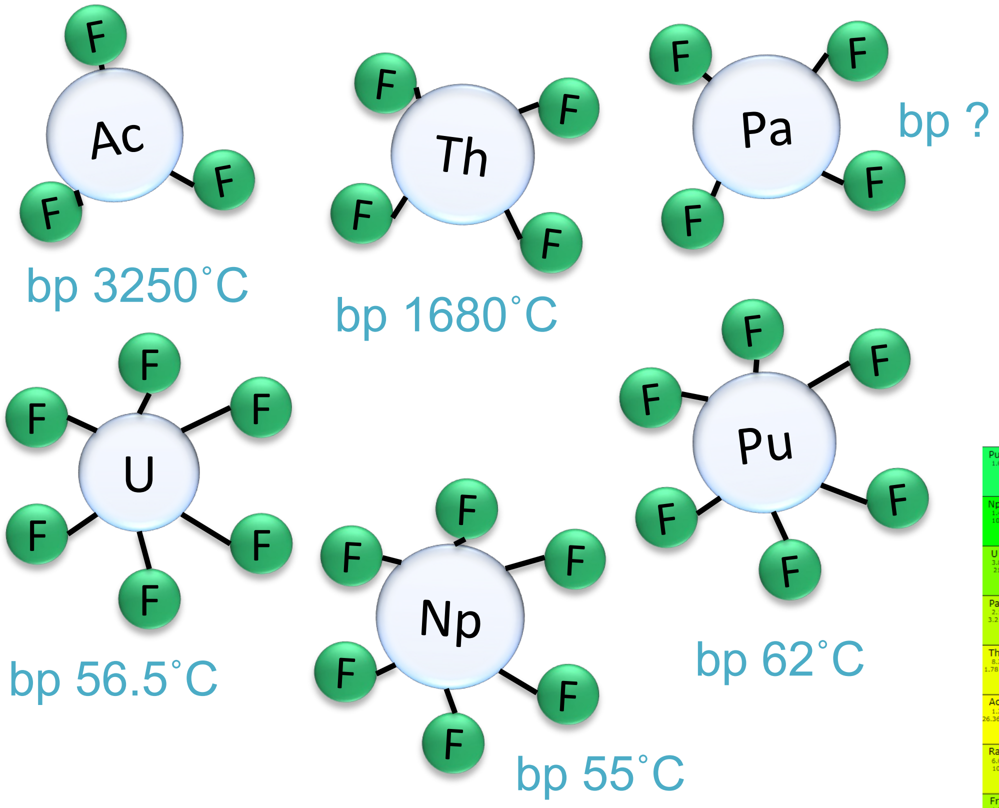
Volatilization

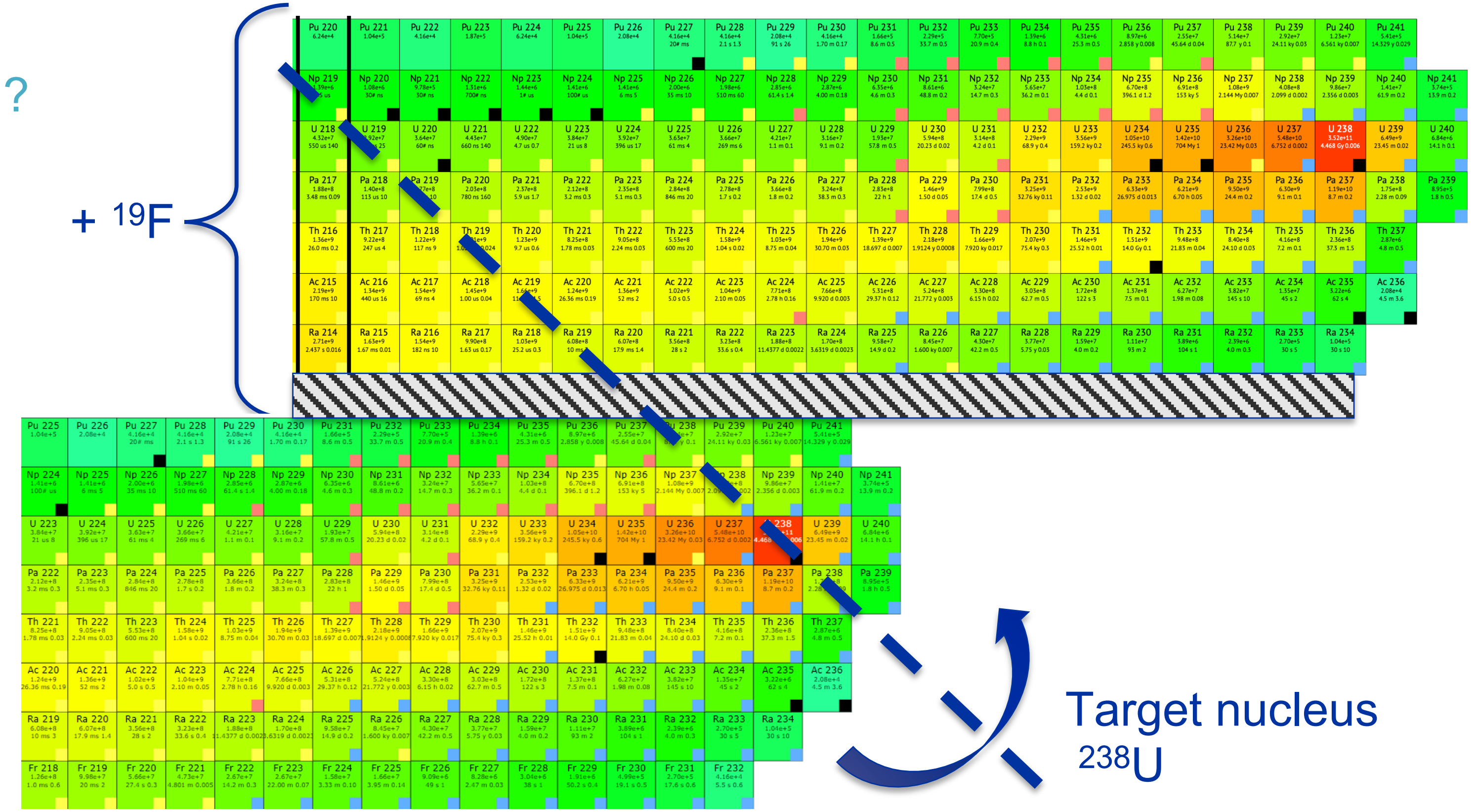



Volatilization
W. Wojtaczka: TbF_x
E. Mamis: ScF_x
proton number Z
boiling temperature (°C)
Yield (μC^{-1})
1e+12
1e+10
1e+8
1e+6
1e+4
1e+2
1e+0
0
20
40
60
80
100
1000
2000
3000
4000
5000

Volatilization





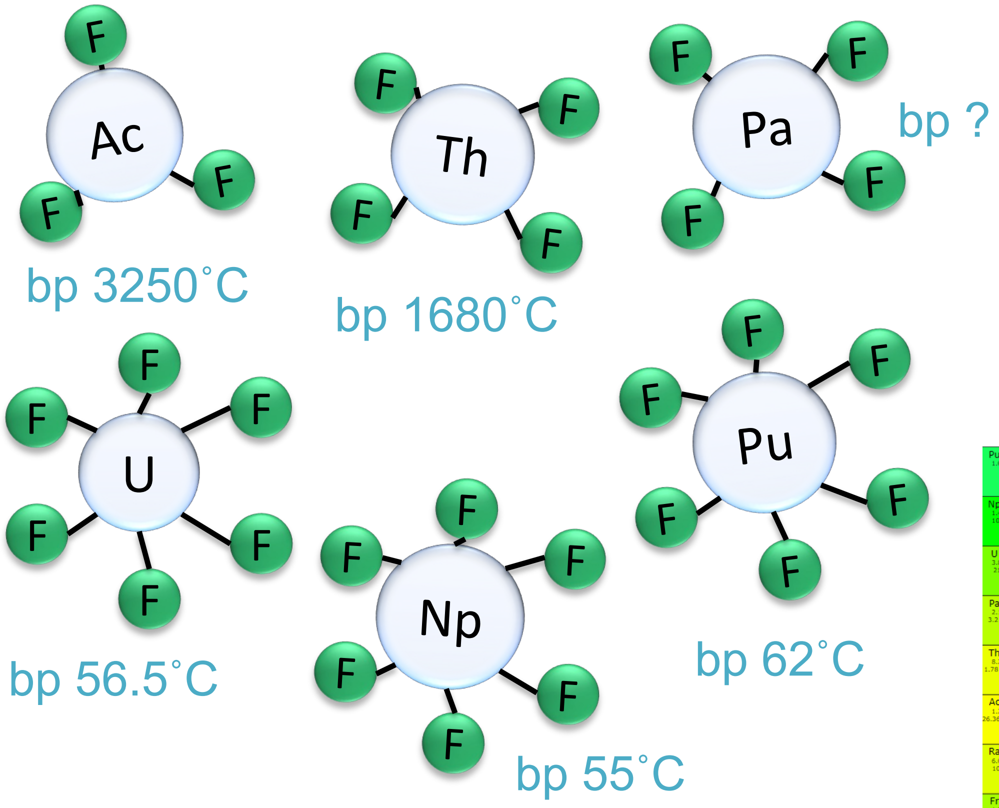


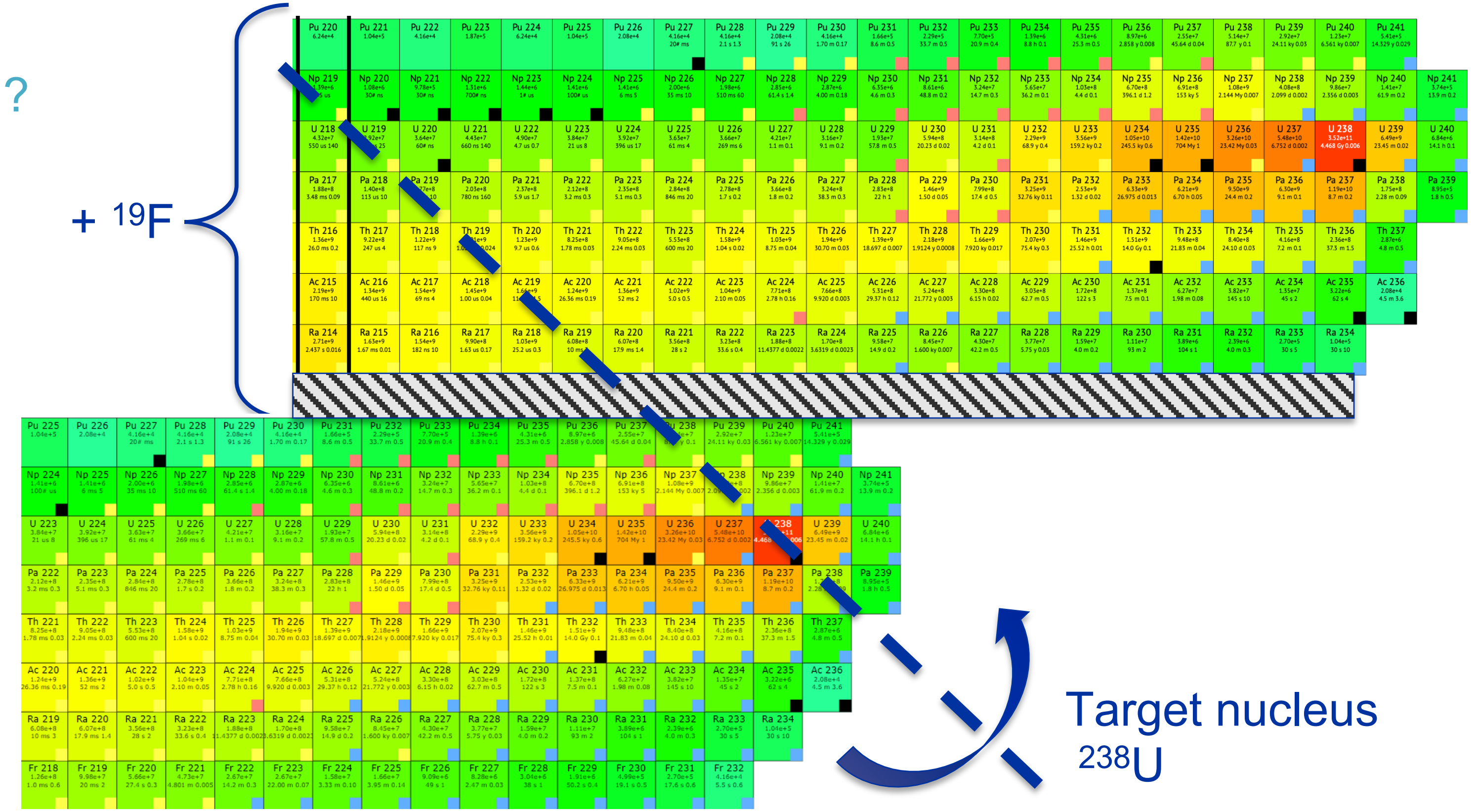
Molecular beams

1. **Volatilization**
2. **Sideband extraction**
3. **Research opportunities**

INSTITUTE for NUCLEAR THEORY

INT Workshop 24-87W Schedule
Fundamental Physics with Radioactive Molecules





Molecular beams

1. Volatilization
2. Sideband extraction
3. Research opportunities

INSTITUTE for NUCLEAR THEORY

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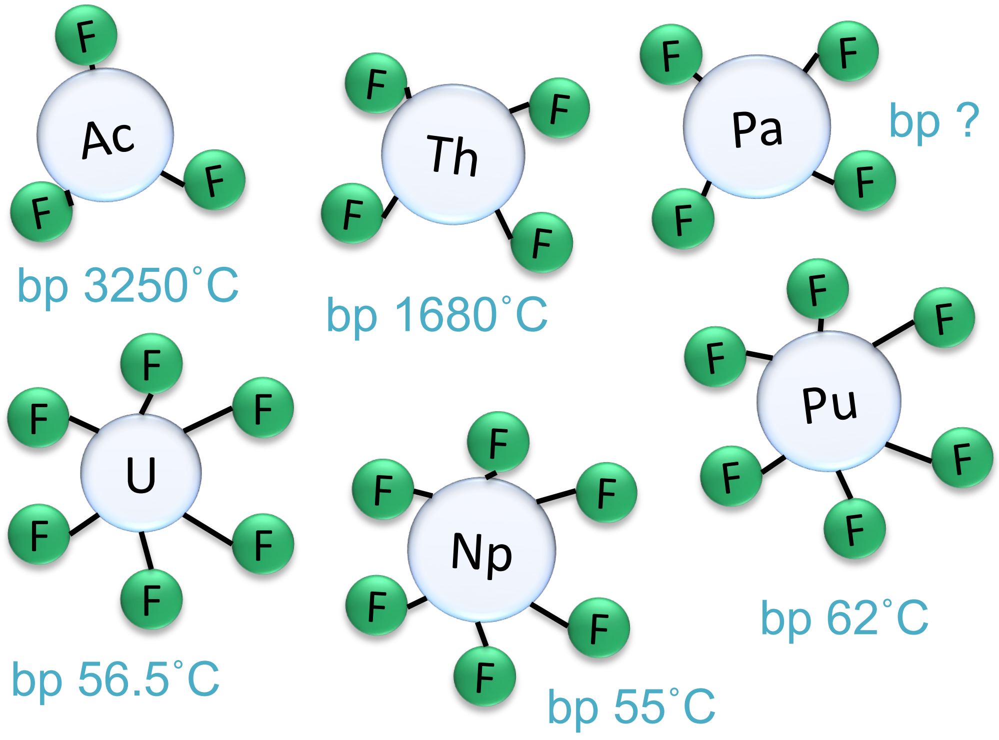
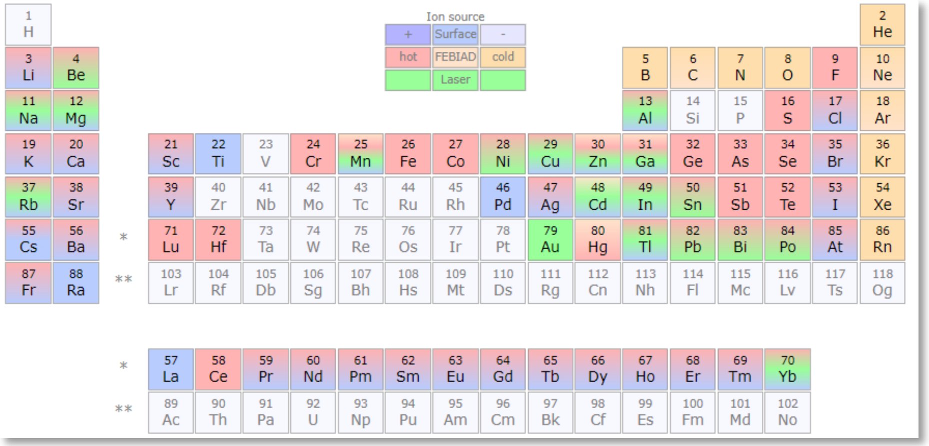
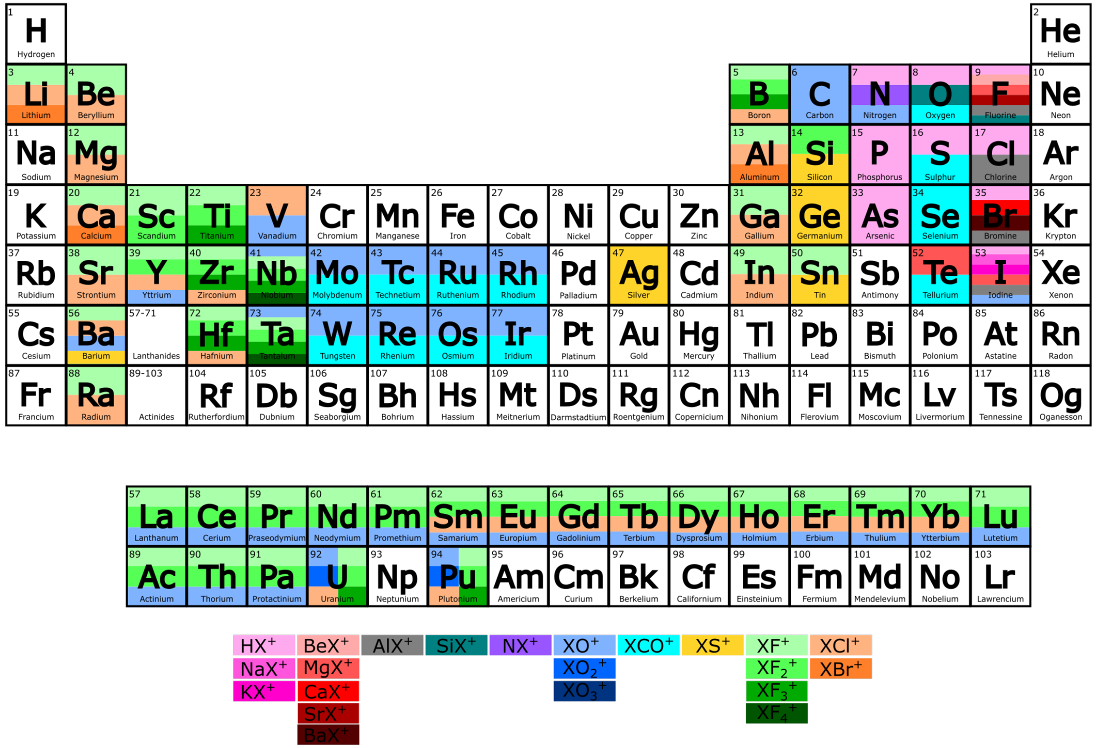
Fundamental Physics with Radioactive Molecules

Opportunities for Fundamental Physics Research with Radioactive Molecules, arXiV 2302.02165 (2023)







Molecular beams

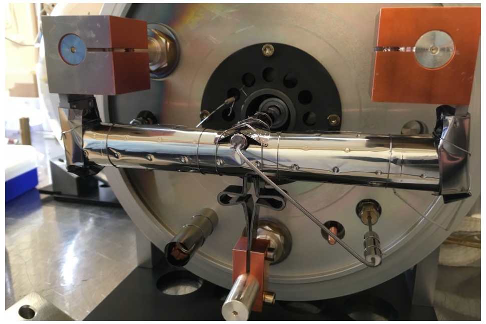
1. Formation
2. Detection and identification
3. Characterization

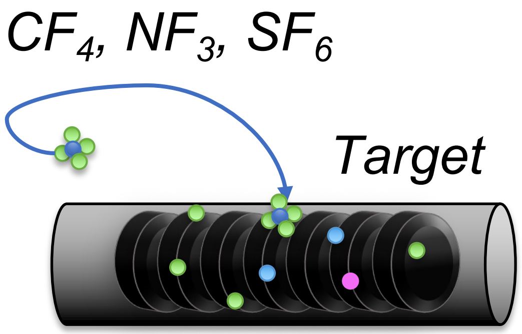
Au and Ballof, (2022) *Zenodo* 10.5281/zenodo.6884293 DOI 10.5281/zenodo.6884293

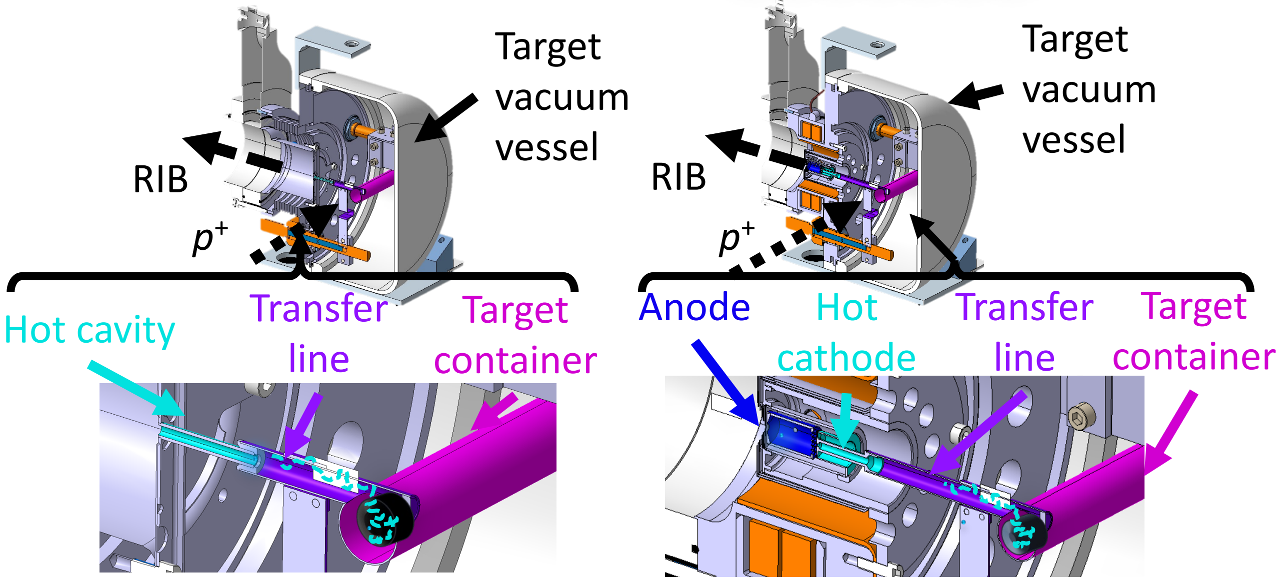
Formation: how do we make the molecules?

In-source

- Reactive gas

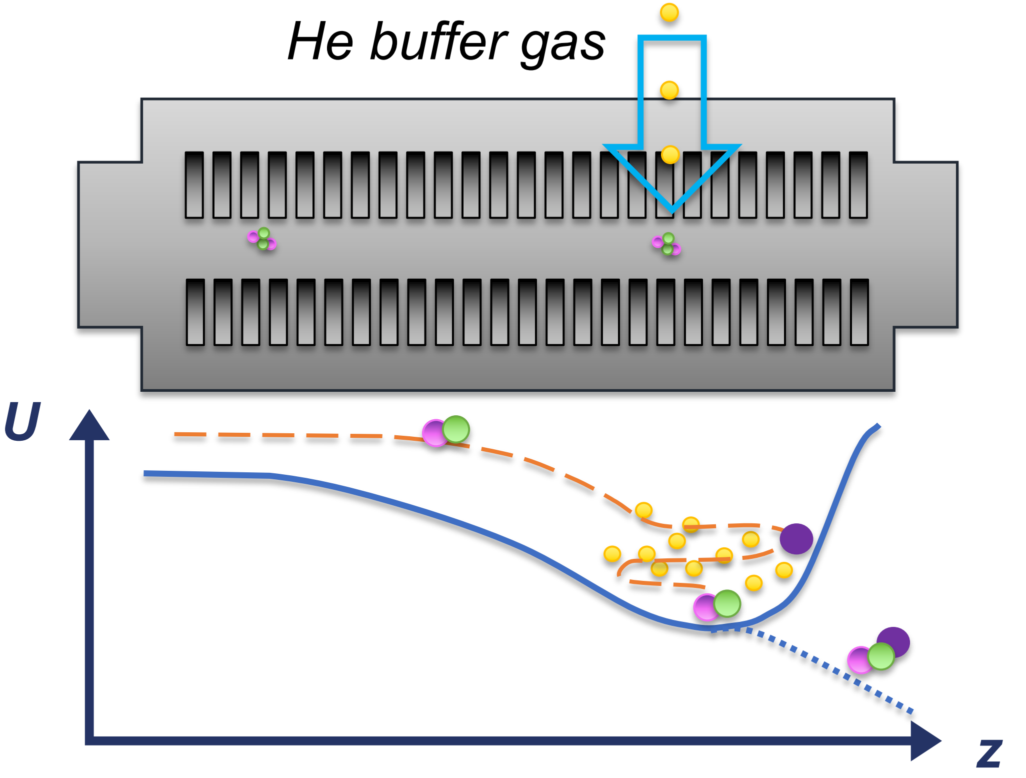






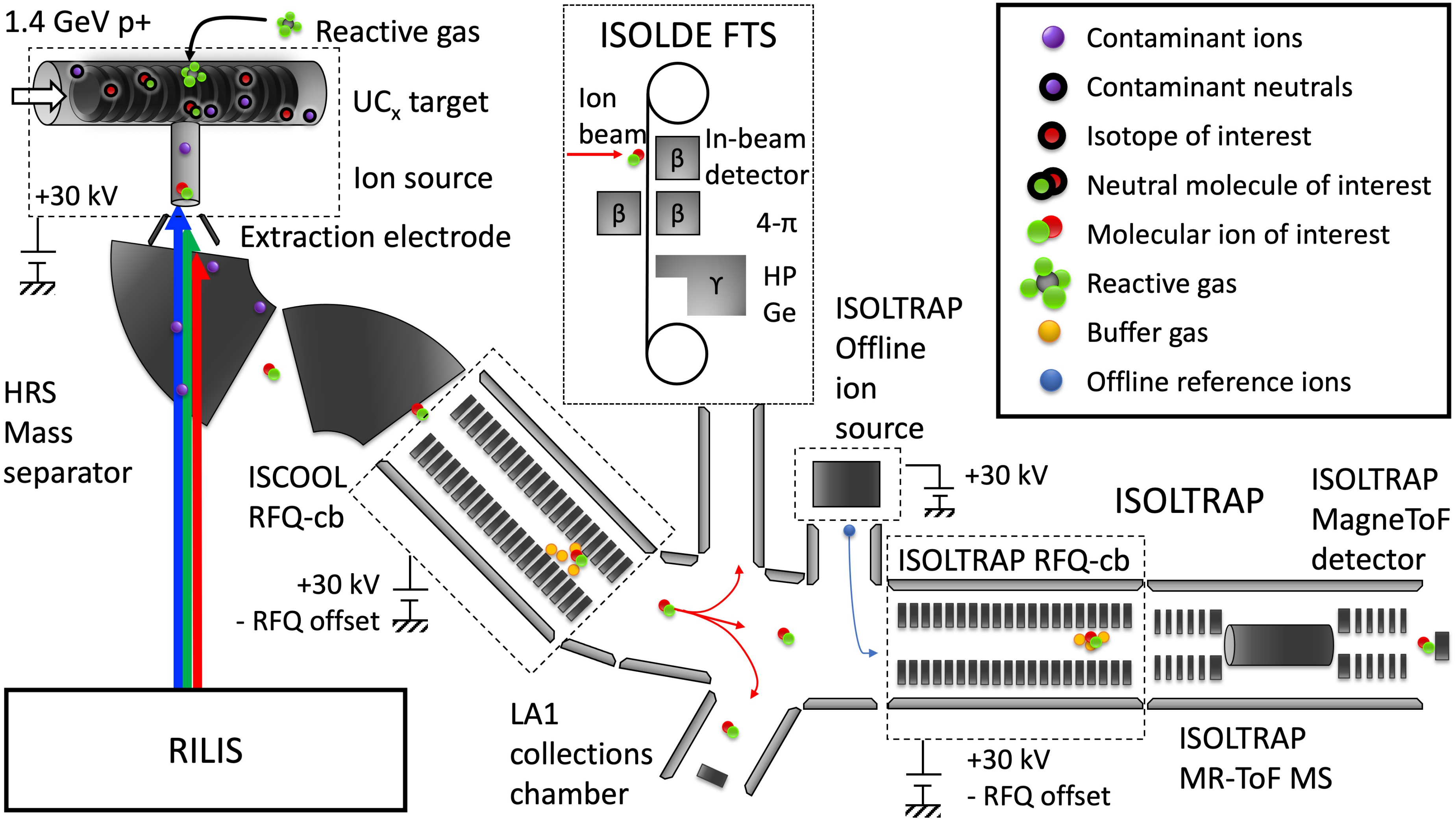
In-trap

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



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

Detection and identification



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

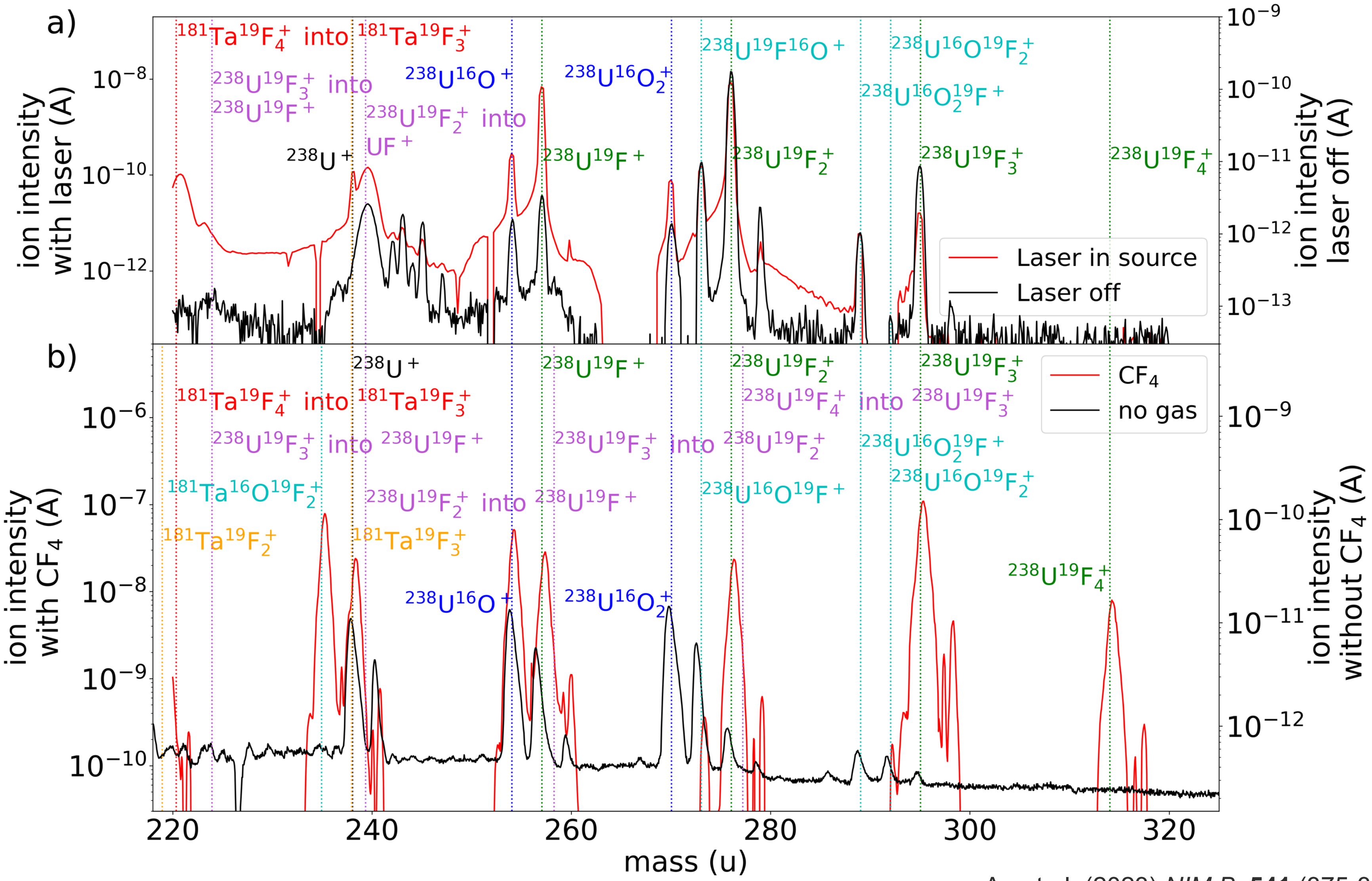
Ion sources and effects

Surface:

- Low IPs
- Surface ionization efficiency
- Production of RaF^+

FEBIAD:

- High/unknown IPs
- High efficiency
- Dissociation
- Production of AcF^+



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

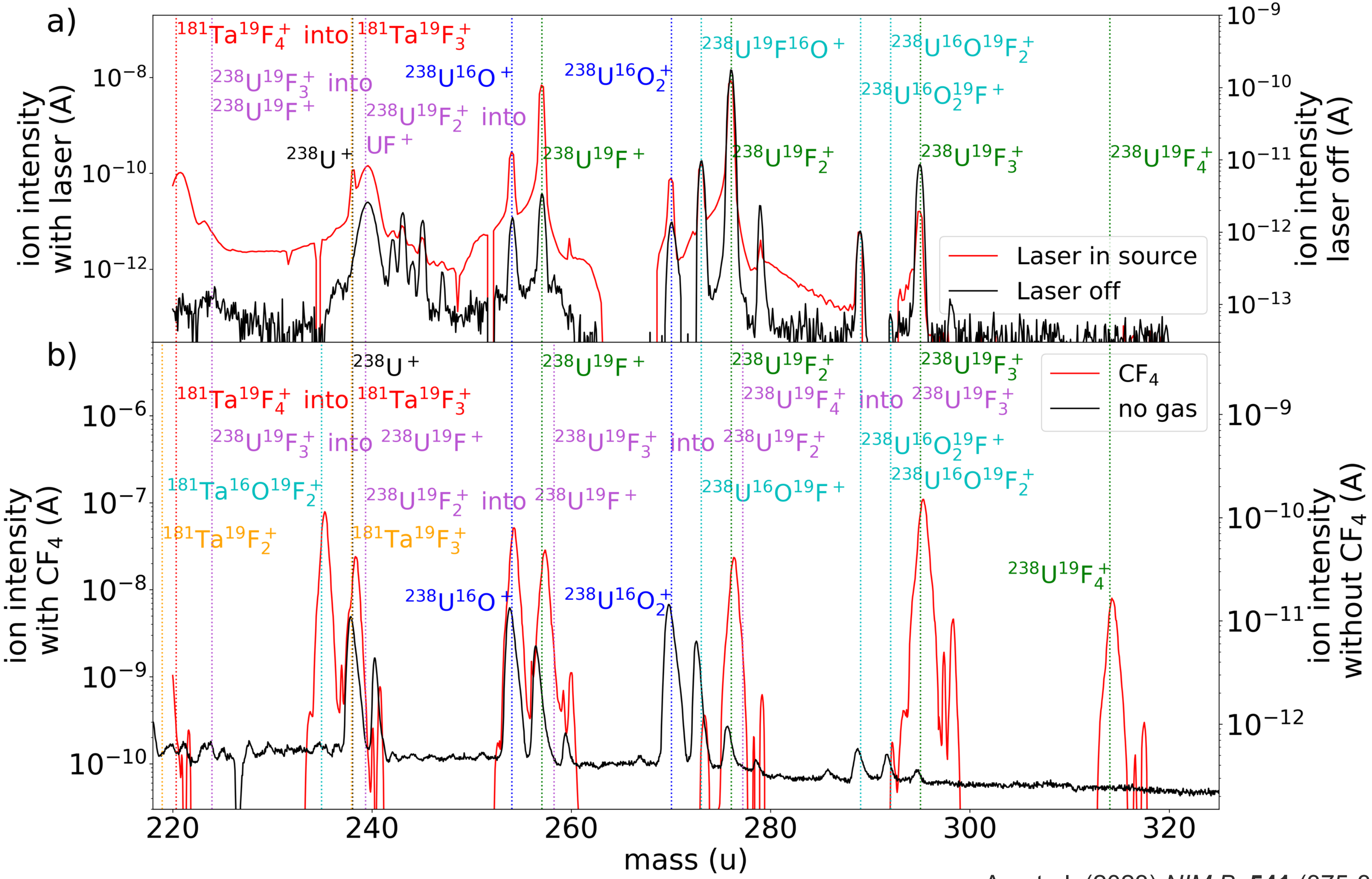
Ion sources and effects

Surface:

- Low IPs
- Surface ionization efficiency
- Production of RaF⁺
  - IP: ~4.9 eV

FEBIAD:

- High/unknown IPs
- High efficiency
- Dissociation
- Production of AcF⁺



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

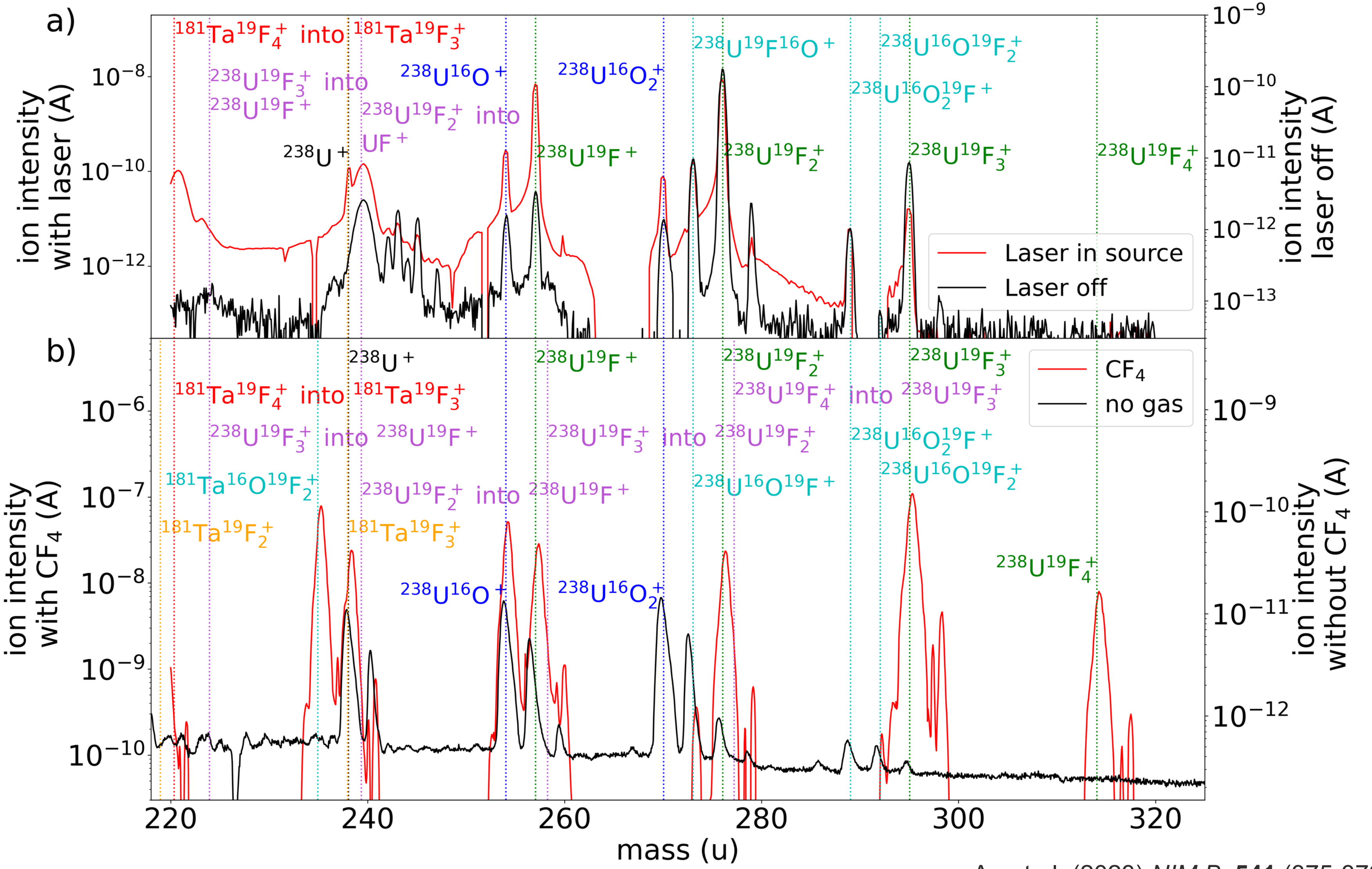
Ion sources and effects

Surface:

- Low IPs
- Surface ionization efficiency
- Production of RaF^+
  - IP: ~4.9 eV

FEBIAD:

- High/unknown IPs
- High efficiency
- Dissociation
- Production of AcF^+
  - IP: ? D_e: ?



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

RaF production and CRIS

Collinear Resonance Ionization Spectroscopy (CRIS) technique

- Fast (10s keV) beams reduce velocity spread
- Collinear geometry: linewidth dominated by laser linewidth

RaF production

- Surface ion source
- CF_4 injection

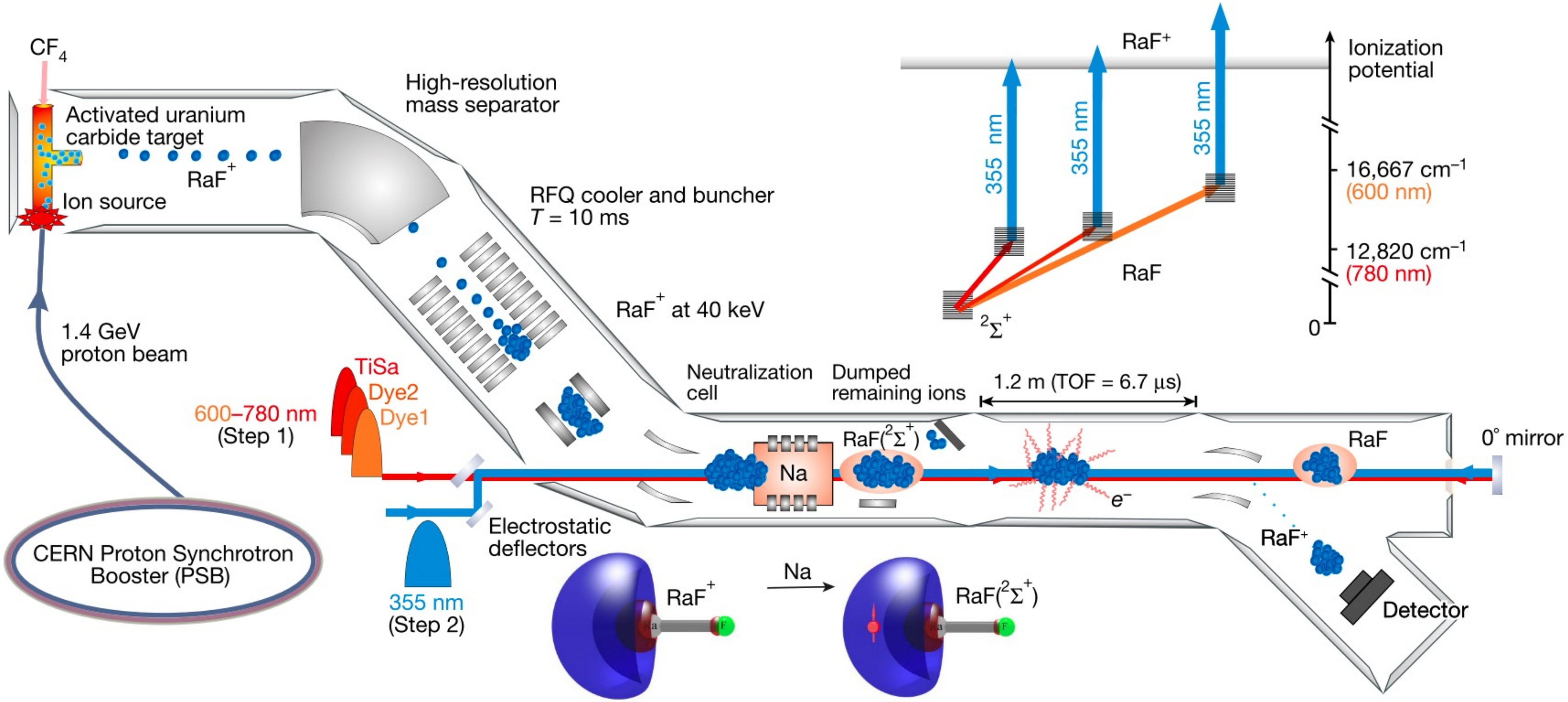
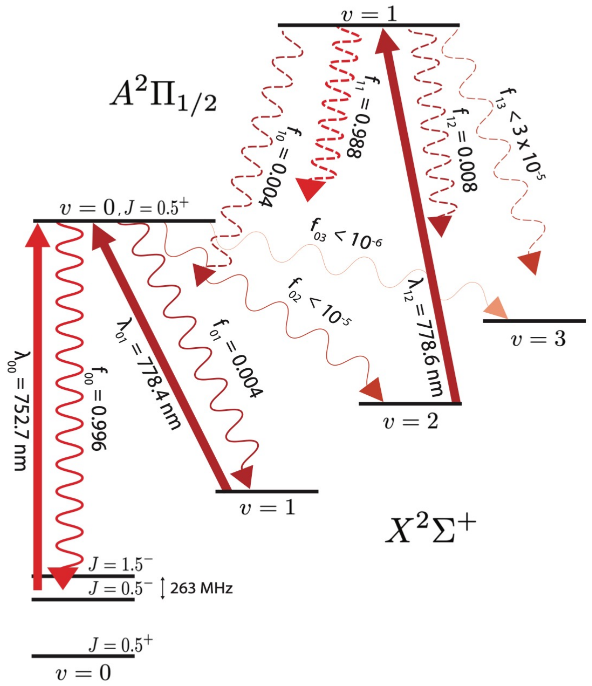


Image published in Garcia Ruiz et al, (2020) *Nature* **581**

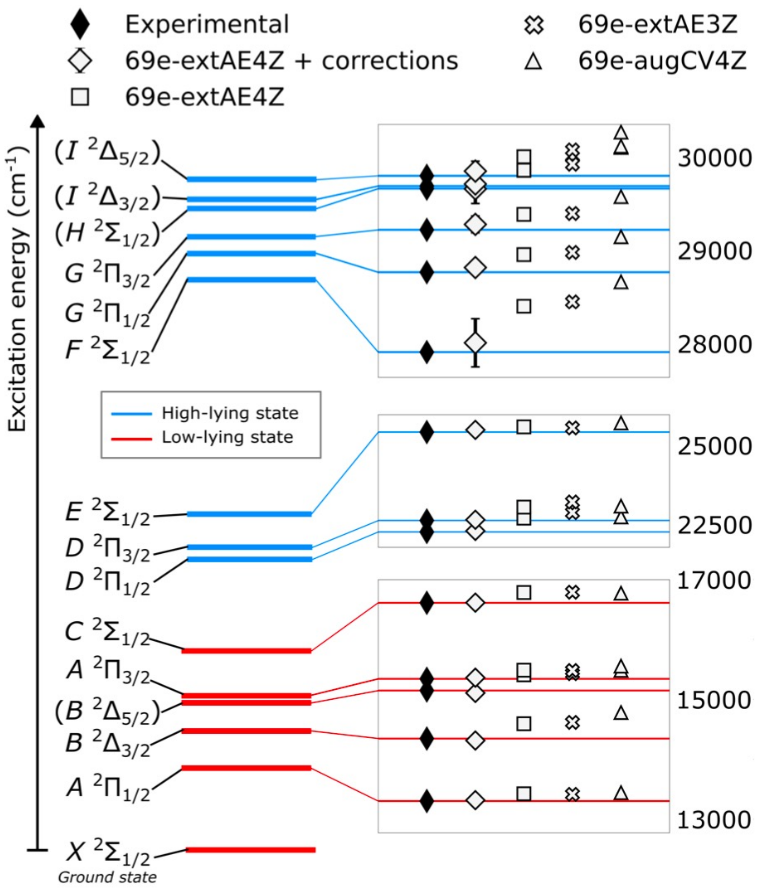
RaF characterization

Laser cooling [1]



Excited states [2]

- agreement ≥ 99.64% (~12 meV)

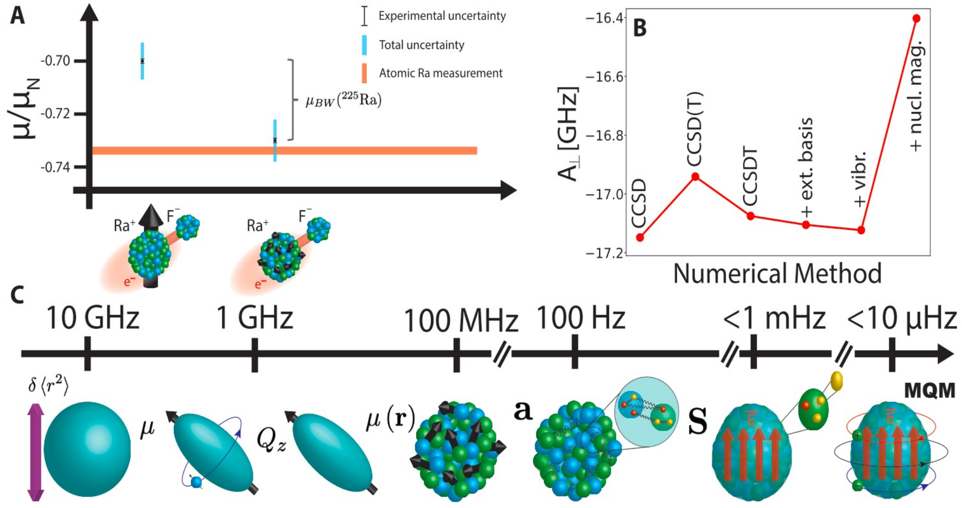


State lifetimes [3]

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

Nuclear magnetization effect [4]

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



[1] Udrescu et al., Research Square 10.21203/rs.3.rs-2648482/v1 accepted in Nat. Phys. (2023)
[2] Athanasakis-Kaklamanakis *et al.*, arXiv 2308.14862 submitted to PRL (2023)
[3] Athanasakis-Kaklamanakis *et al.*, in preparation (2024)
[4] Wilkins *et al.*, arXiV 2311.04121 submitted to Science (2024)

Actinium

T-αT (targeted alpha therapy)

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

9-10 MBq

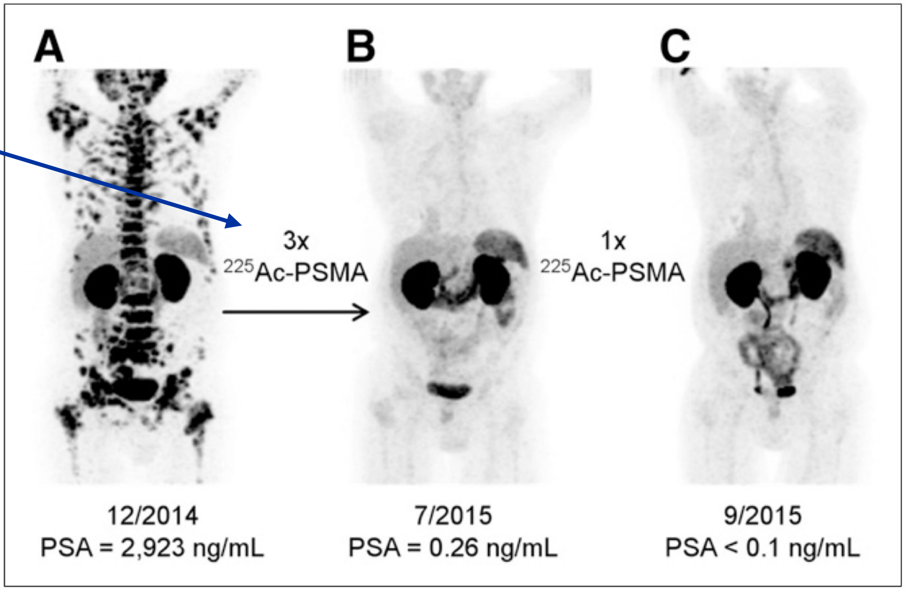


Figure published in: Kratchowil *et al.* (2016) J. Nucl. Med. **57** 1941-1944

Production routes

- **^{226}Ra**
  - ^{226}Ra(p,2n)^{225}Ac

^{225}Ra (generator)

- ^{226}Ra(γ,n)^{225}Ra → ^{225}Ac
- ^{226}Ra(n,2n)^{225}Ra → ^{225}Ac

Actinide

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

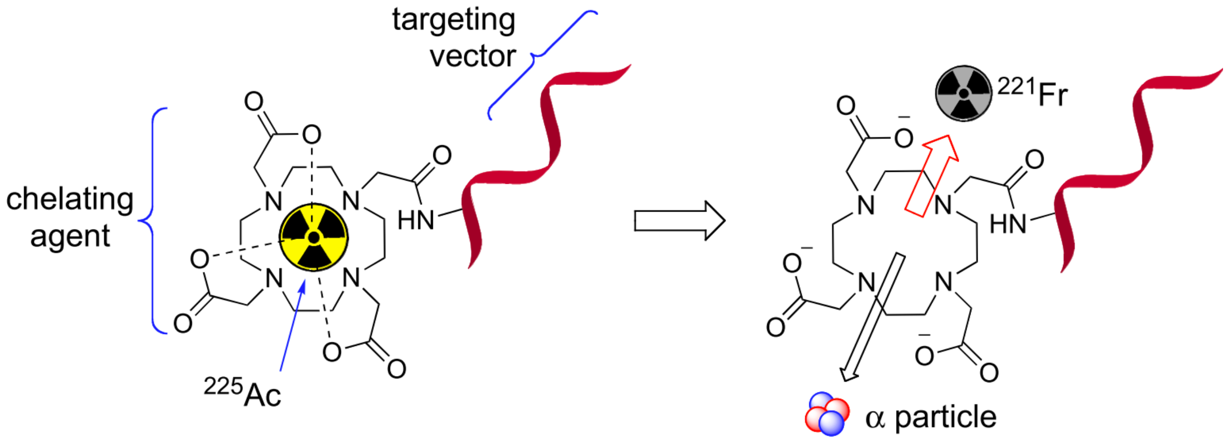


Figure published in: Robertson *et al.* (2018) Current Radiopharmaceuticals. **11** 156-172

Actinium (Fluoride)

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]

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

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

The energy shift is

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

[1] Verstraelen et al., Phys. Rev. C. 100, 044321 (2019)
[2] Heinke et al., CERN-INTC-2020-029, INTC-P-556, https://cds.cern.ch/record/2717945 (2020)
[3] Flambaum, Feldmeier, Phys. Rev. C. 101, 015502 (2020)
[4] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

Actinium (Fluoride)

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: ?

[4]

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 $[e\,\text{fm}^3\bar{\theta}]$	$d_A[e\,\text{cm}]$ $10^{-17}S[e\,\text{fm}^3]$	$10^{-17}\bar{\theta}$
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}Fr^{+}	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	$\lesssim 1$

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

[3]

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

$$W_S \approx 46000 \text{ a.u.} \tag{55}$$

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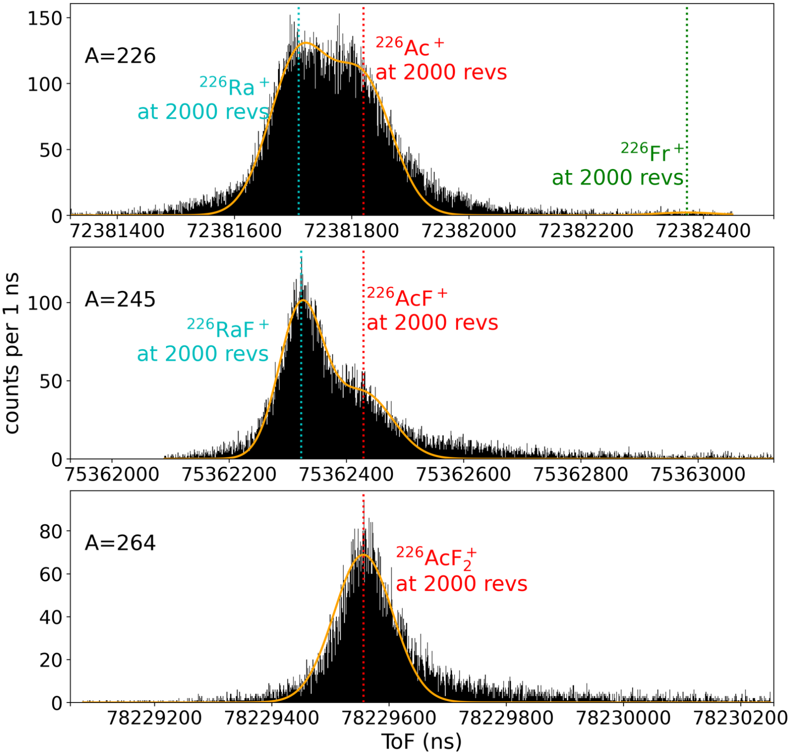
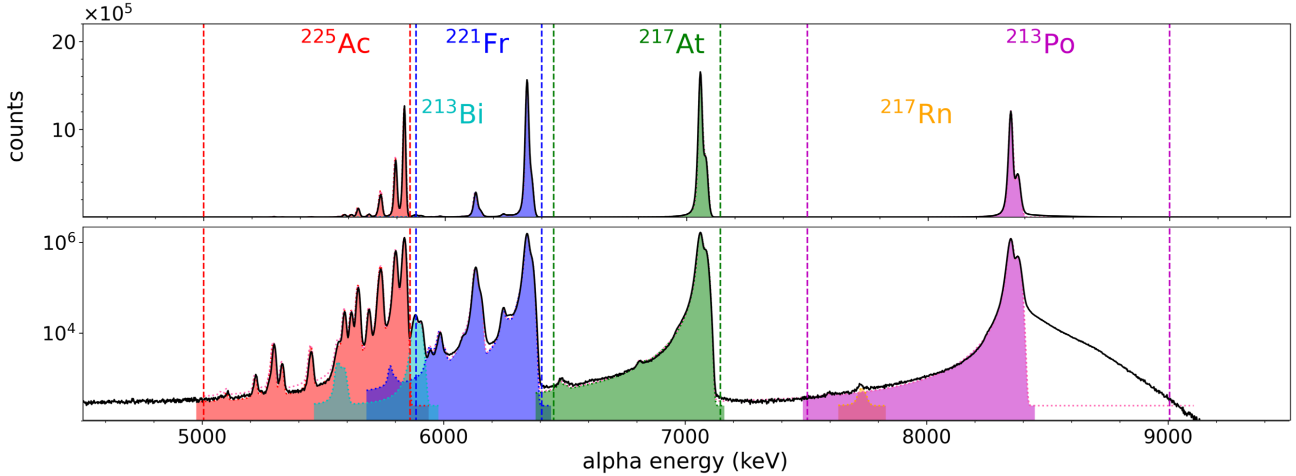
[4] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

Production of AcF_x^+

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





[1] Au, *PhD thesis* (2023)
[2] Au, *submitted to Nat. Comms* (2024)

AcF spectroscopy

Experimental [1,3]

- $(8)\Pi_1 \leftarrow X\ ^1\Sigma_0$

Nuclear theory

- *previous values from scaling factors*
- $S_{int} \leftrightarrow Q_0{}^3$ [2]
- DFT: $S_{int}(^{227}Ac)$ vs. $S_{int}(^{225}Ra)$ = 26.6(19) *e* fm^3 [3]

Molecular theory

- IH-FS-RCCSD
- IP = 48,866 cm^{-1}
- D_e = 57,214 cm^{-1}

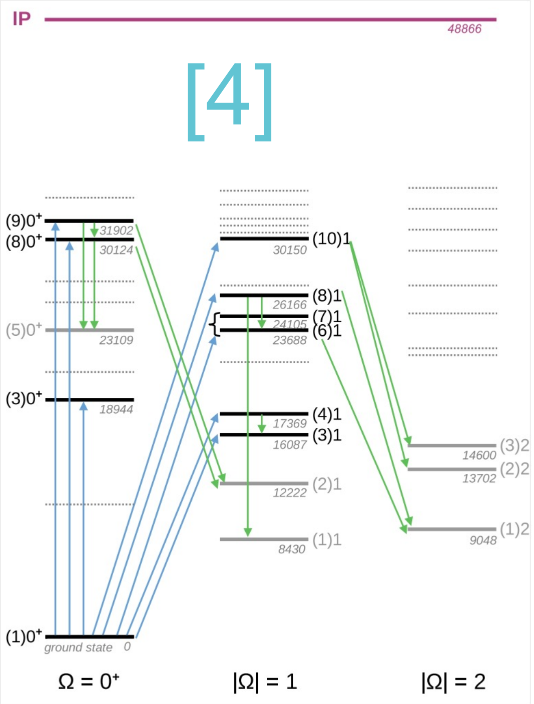


FIG. 3. The strongest transitions (blue arrows) from the $X(1)0^+$ ground state of AcF and the strongest transitions for stimulated emission (green arrows). Levels accessible with two-step excitations are shown with solid gray lines. Dotted lines depict electronic states that are hardly accessible from the ground state with either direct or two-step excitations. It is noted that all transitions to the $\Omega = 0^-$ states have low probabilities and are not shown here. T_e values (cm^{-1}) are shown.

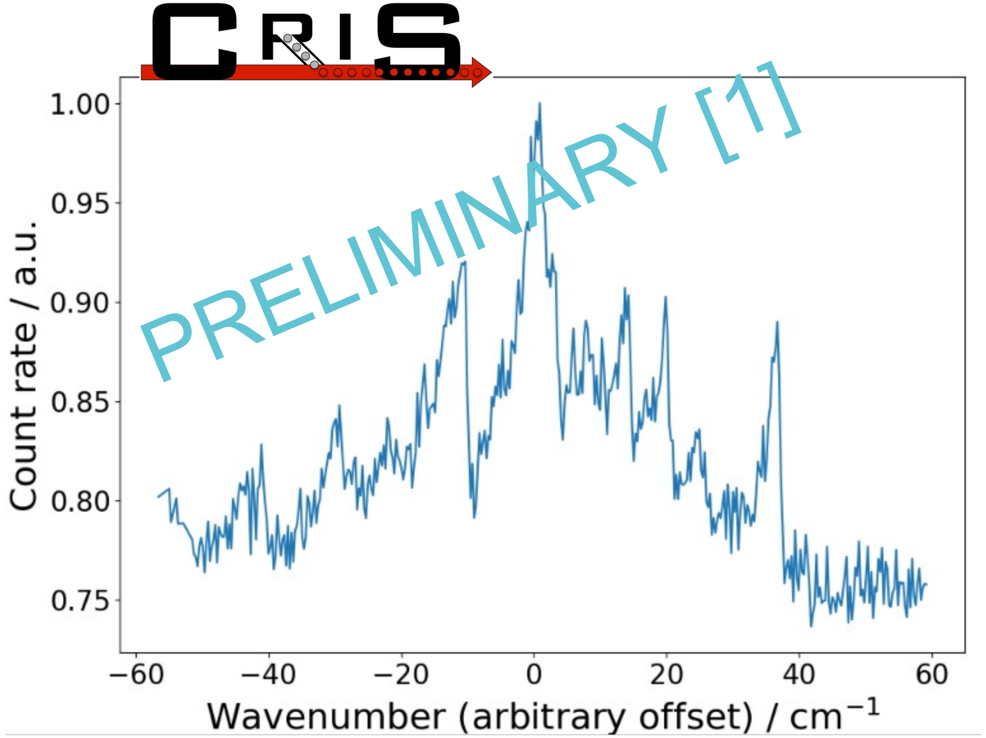


Table 2 Molecular constants X and $W_S^{(2)} = 6X/r^{sp}$ $(e/a_B{}^4$, $a_B = 1$ Bohr) calculated at different levels of theory, given in square brackets

Mol.	State	X [HF]	X [CCSD]	X [CCSD(T)]	r^{sp}	$W_S^{(2)}$ [CCSD(T)]
AcF	$^1\Sigma^+$	−2022	−1569	−1593	1.16	−8240
AcN	$^1\Sigma^+$	−10 580	−9415	−8950	1.16	−46 295
AcO^+	$^1\Sigma^+$	−13 362	−11 600	−11 302	1.16	−58 461
ThO	$^1\Sigma^+$	−3965	−3187	−3332	1.17	−17 085
EuO^+	$(f^6)^a$	-2475^a	-2140^a	-2114^a	1.09	$-11\,677^a$
EuN	$(f^6)^a$	-1975^a	-1847^a	-1890^a	1.09	$-10\,419^a$
TlF	$^1\Sigma^+$	9111	7262	7004	1.13	37 192

[a] The spin–orbit part of the GRECP operator has been omitted in the calculation. Therefore, we give only the configuration of the molecular state.

[5]

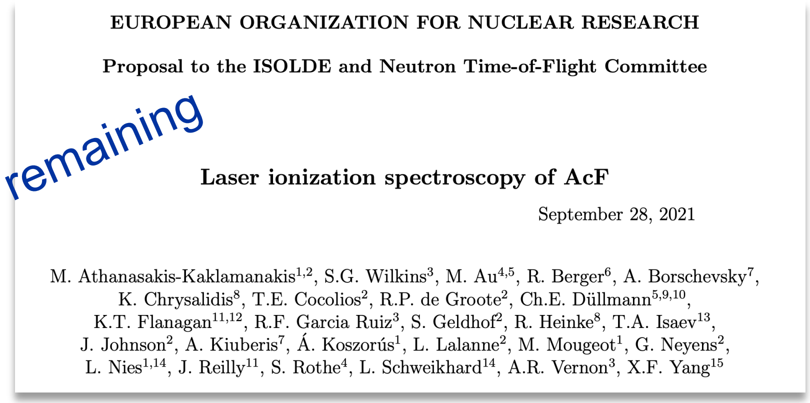
[1] Athanasakis-Kaklamanakis and Au, (2023) *CERN EP newsletter*
[2] Dobaczewski, Engel, Kortelainen, Becker, *Phys. Rev. Lett.* **121,** 232501 (2018)
[3] Athanasakis-Kaklamanakis, Au, Kyuberis, Zülch, Wibowo, Skripnikov, Reilly, Lalanne *et al., in prep.* (2024)
[4] Skripnikov *et al.,* J. Chem. Phys **159** 124301 (2023)
[5] Skripnikov *et al.,* Phys. Chem. Chem. Phys **22** 18374-18380 (2020)

Fluoride beams current status

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

Image published in EP Newsletter, CERN (2020)

15 shifts remaining



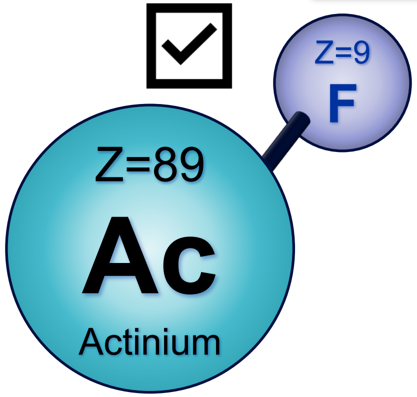
EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

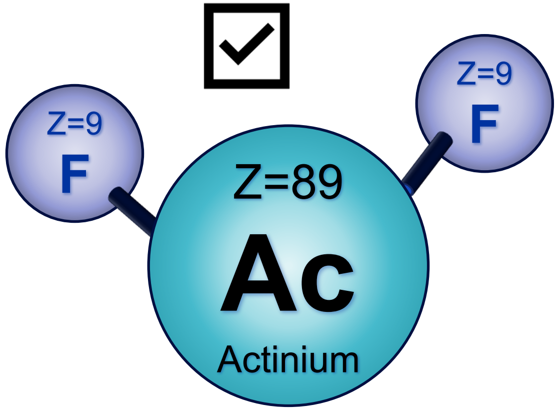
Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Laser ionization spectroscopy of AcF

September 28, 2021

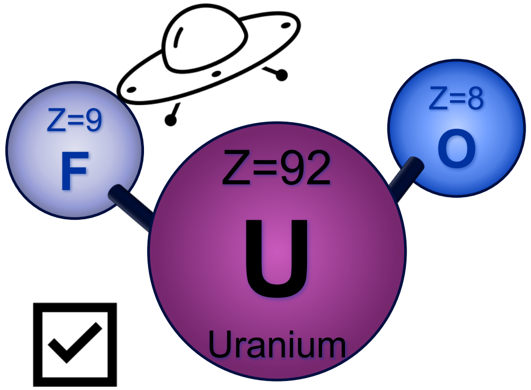
M. Athanasakis-Kaklamanakis[1,2], S.G. Wilkins[3], M. Au[4,5], R. Berger[6], A. Borschevsky[7], K. Chrysalidis[8], T.E. Cocolios[2], R.P. de Groote[2], Ch.E. Düllmann[5,9,10], K.T. Flanagan[11,12], R.F. Garcia Ruiz[3], S. Geldhof[2], R. Heinke[8], T.A. Isaev[13], J. Johnson[2], A. Kiuberis[7], Á. Koszorús[1], L. Lalanne[2], M. Mougeot[1], G. Neyens[2], L. Nies[1,14], J. Reilly[11], S. Rothe[4], L. Schweikhard[14], A.R. Vernon[3], X.F. Yang[15]



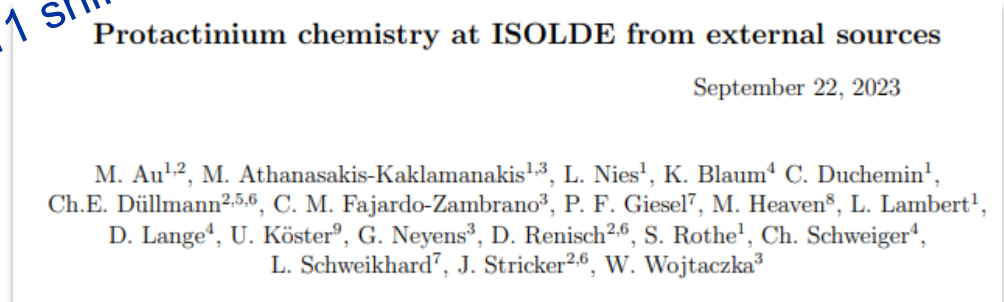


Next steps

- TbF_x TISD beamtime (2024)
- PaF_x from external samples (2024)



11 shifts approved

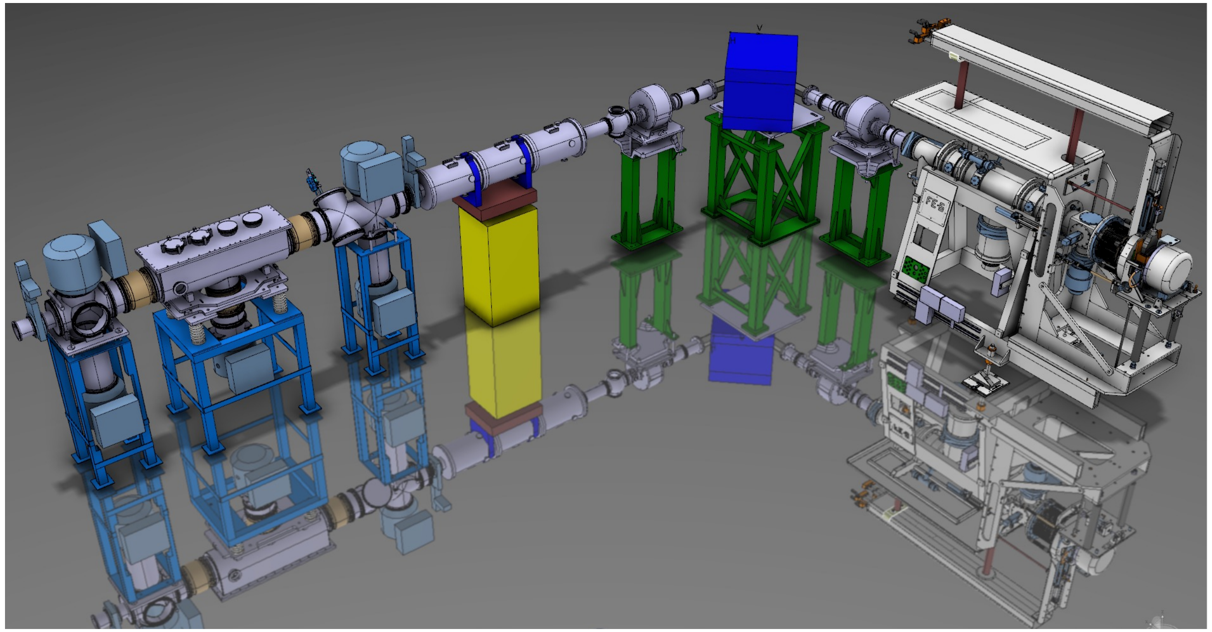


Protactinium chemistry at ISOLDE from external sources

September 22, 2023

M. Au[1,2], M. Athanasakis-Kaklamanakis[1,3], L. Nies[1], K. Blaum[4] C. Duchemin[1], Ch.E. Düllmann[2,5,6], C. M. Fajardo-Zambrano[3], P. F. Giesel[7], M. Heaven[8], L. Lambert[1], D. Lange[4], U. Köster[9], G. Neyens[3], D. Renisch[2,6], S. Rothe[1], Ch. Schweiger[4], L. Schweikhard[7], J. Stricker[2,6], W. Wojtaczka[3]

*Yield publication in progress



ISOLDE OFFLINE 1
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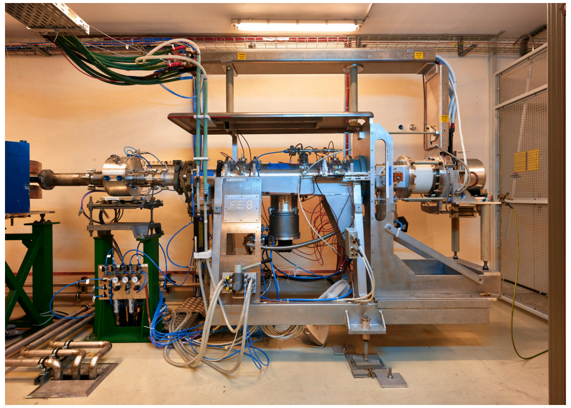

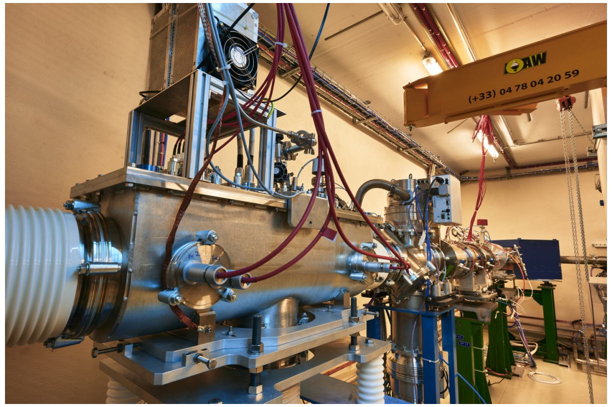

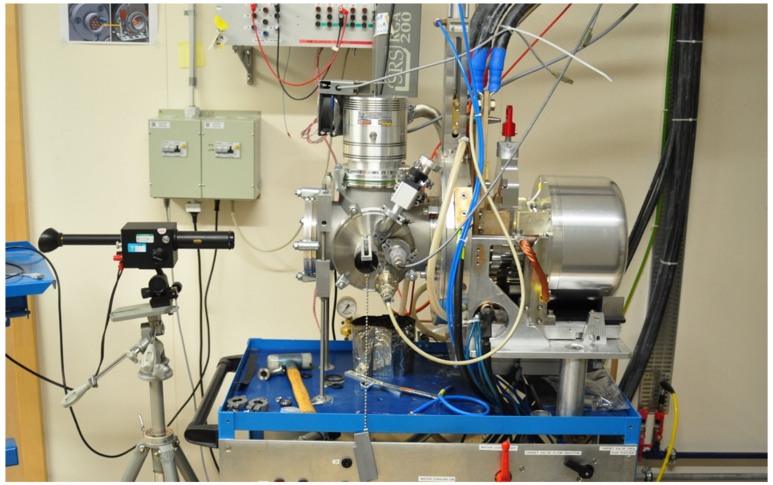

ISOLDE PUMP STAND
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ISOLDE OFFLINE 2
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Behind the scenes: Offline developments

Material developments

Gas injection

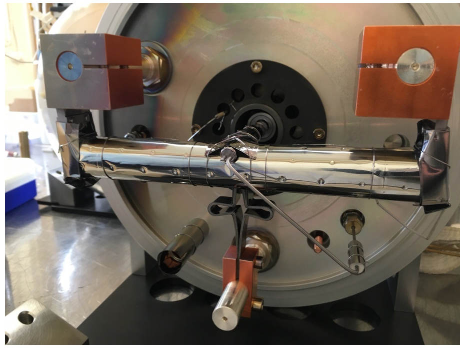
- Reactive/corrosive gases

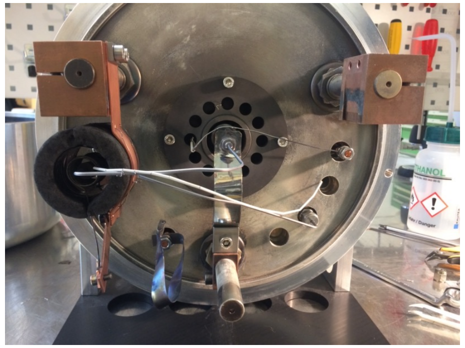
Reactants

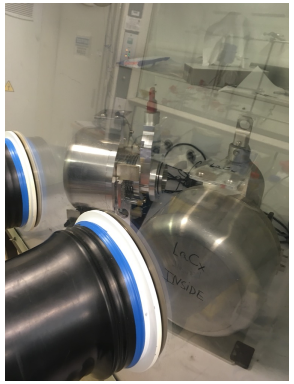
- Mass markers

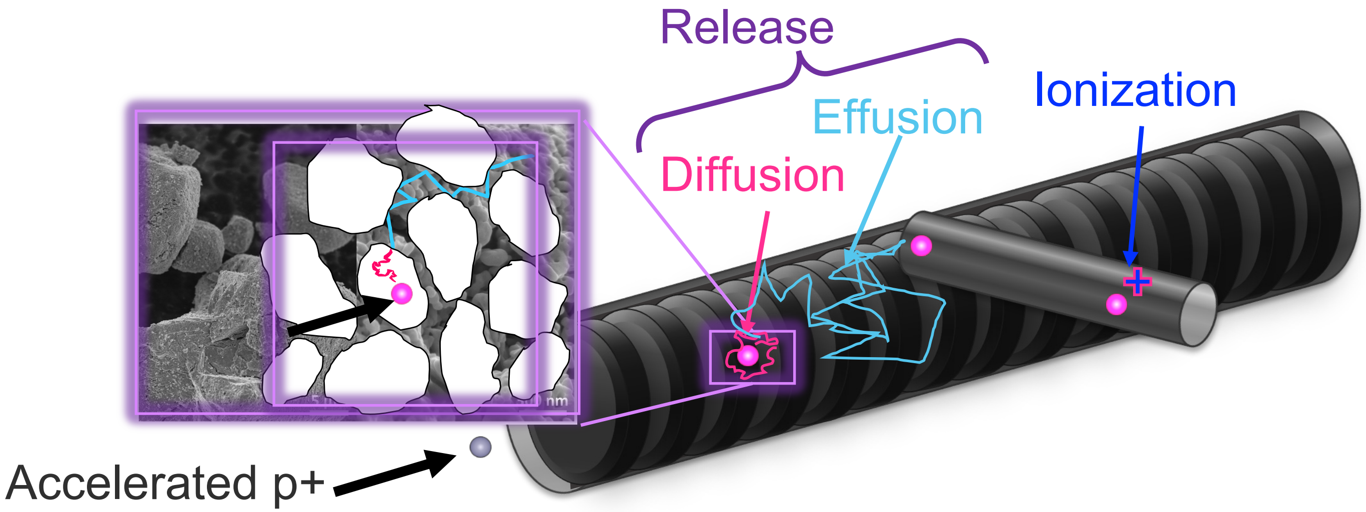
Target materials

- Particle size
- Open porosity









$$Beam\ Intensity = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms
j – Proton flux [cm^{-2}]
σ – Cross section [mb]
ε – Efficiency [%]

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

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

Material developments

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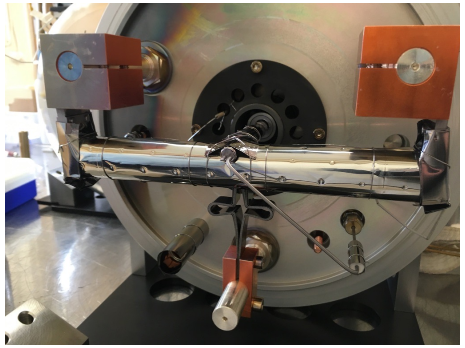

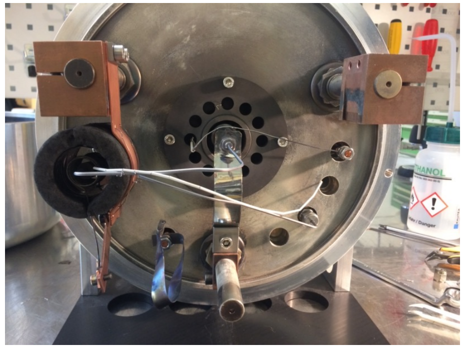

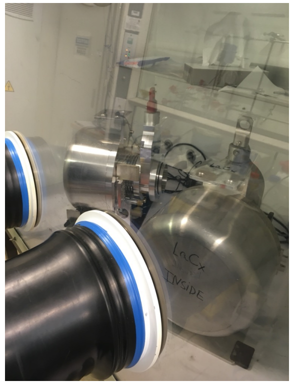

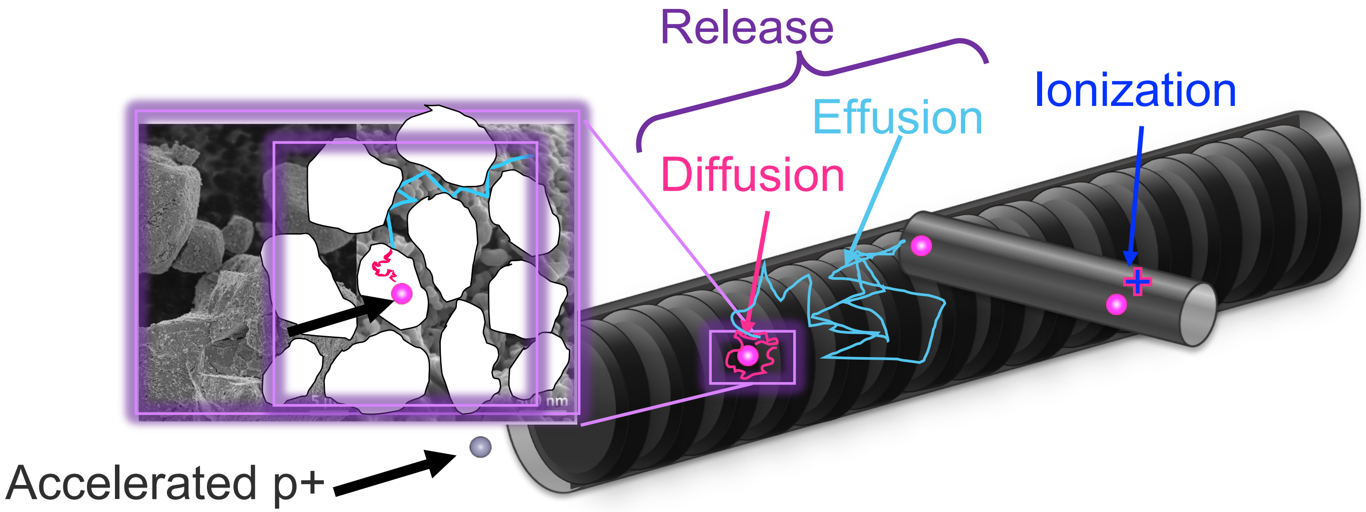

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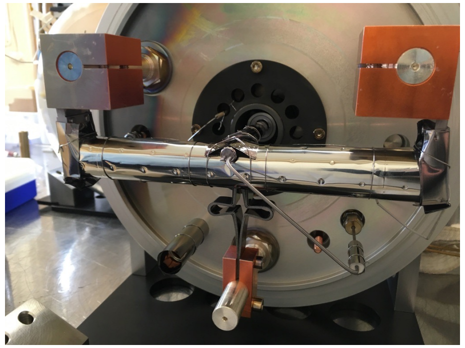

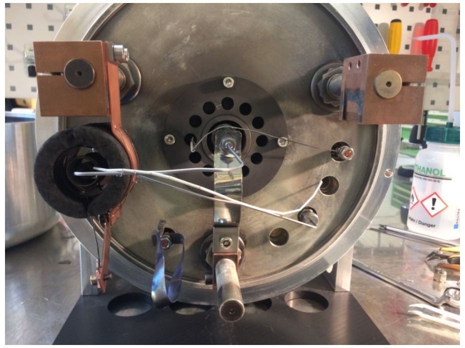

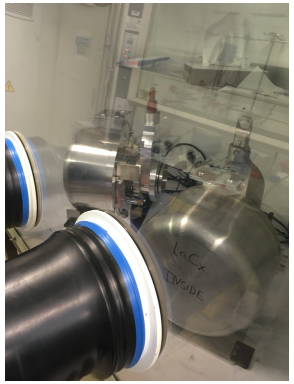

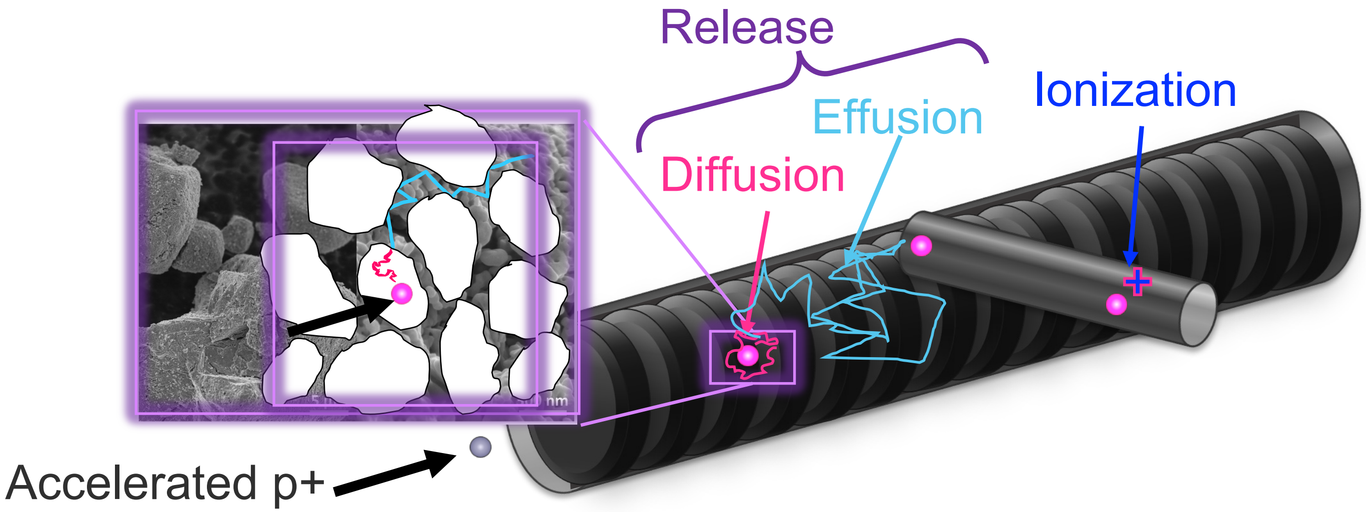

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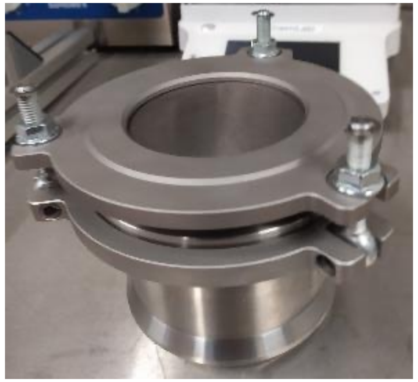
Small G, high T ⇨ Increased $\varepsilon_{\text{diff}}$

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

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

Non-actinide development and characterization lab

Planetary ball mill – Powder particle size reduction



Laser diffraction **particle size analyzer**

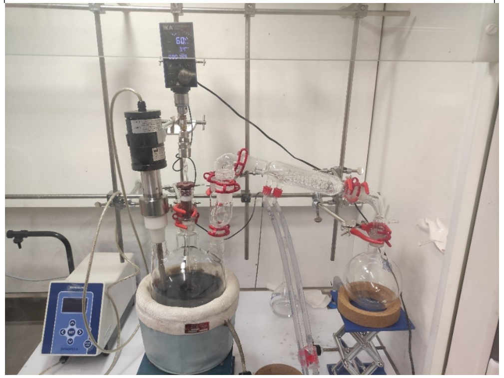
Gas sorption – Pore size distribution (BET)

Date: 2021-07-09

EDMS : 2596262

Operational Procedure

Production of nano-LaOH + multiwalled carbon nanotube powder pellets for ISOLDE target production



Reactor setup

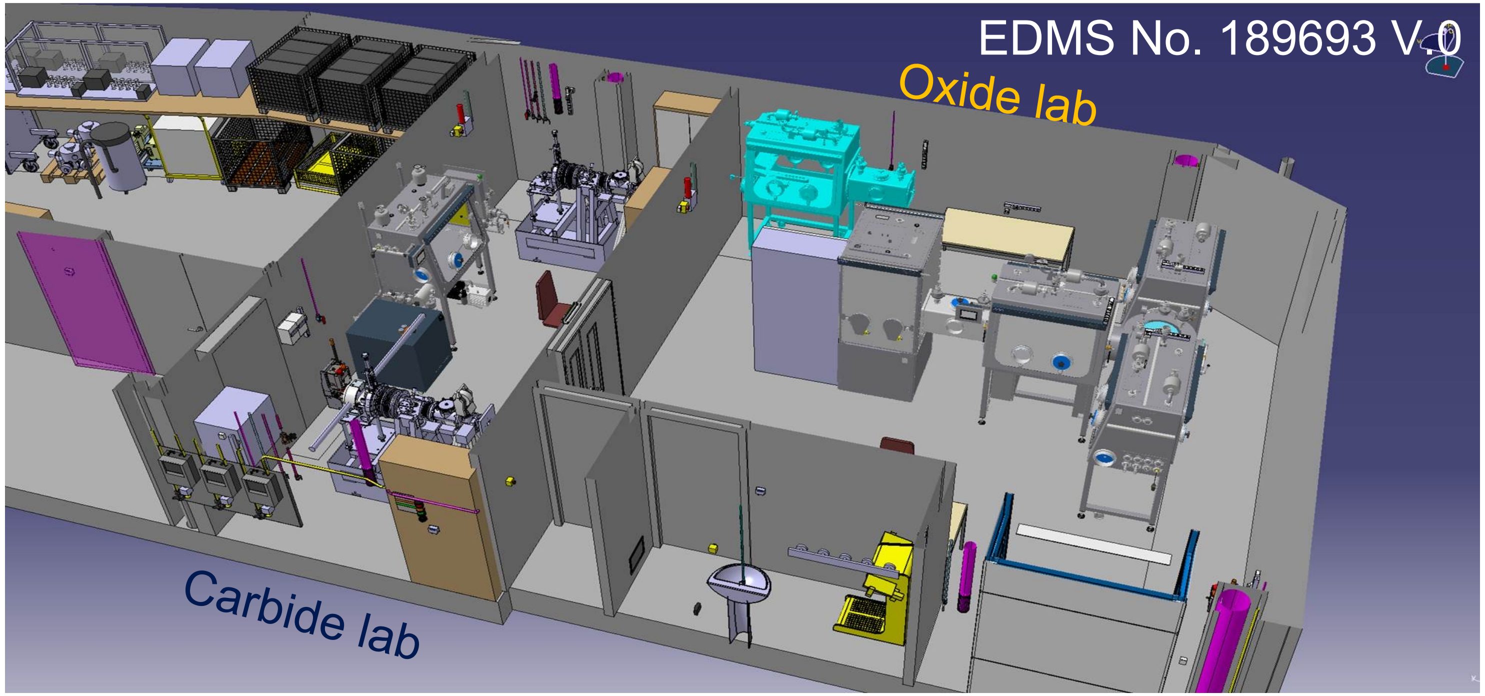
Carburization pumpstand
Target development, sintering studies

Gas pycnometry
Apparent density determination

TGA-MS – Reaction kinetics

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

The Nanolab: Production and Research



Carbide lab

Oxide lab

5 Glove boxes

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

Production alternating with development

Storage capsules

Photos courtesy of L. Lambert

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]

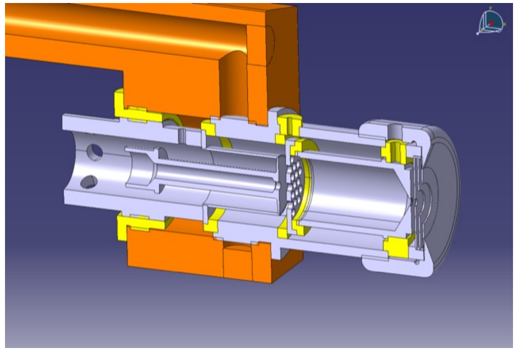
- Cold (room-temperature) environments

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)

Ion source developments



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Ion source developments

Molecular breakup and characterization studies

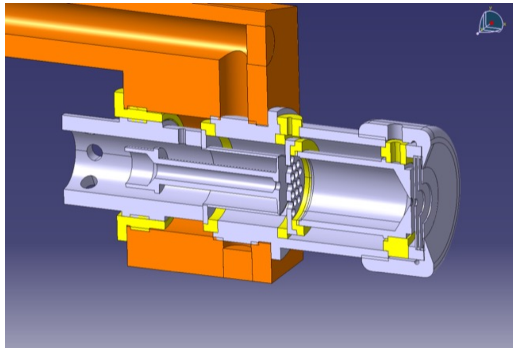
- FEBIAD-type ion sources [1,2]
- Electron energy and source optimization
- Ion source systematics

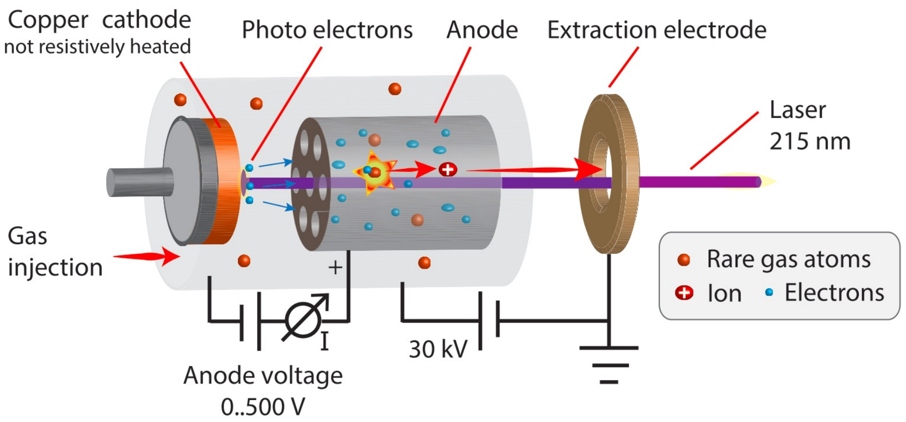
Photocathode ion sources [3]

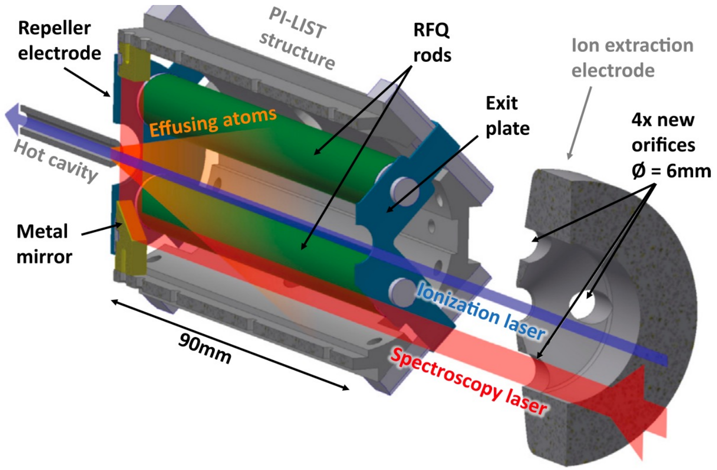
- Cold (room-temperature) environments

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)

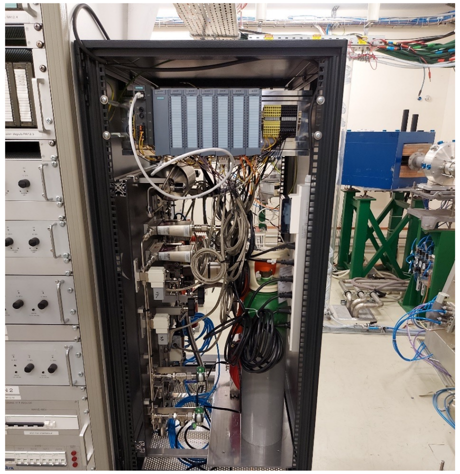
Offline upgrades for molecular beam development

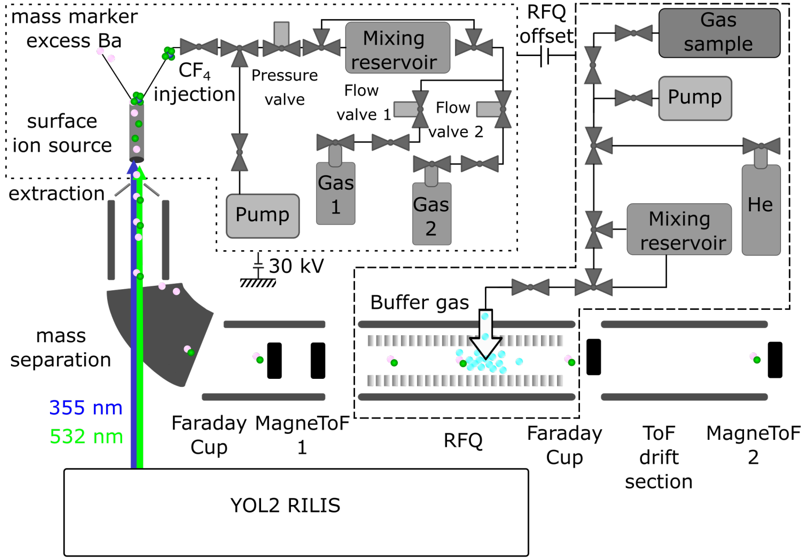
Detection, implantation, ion counting

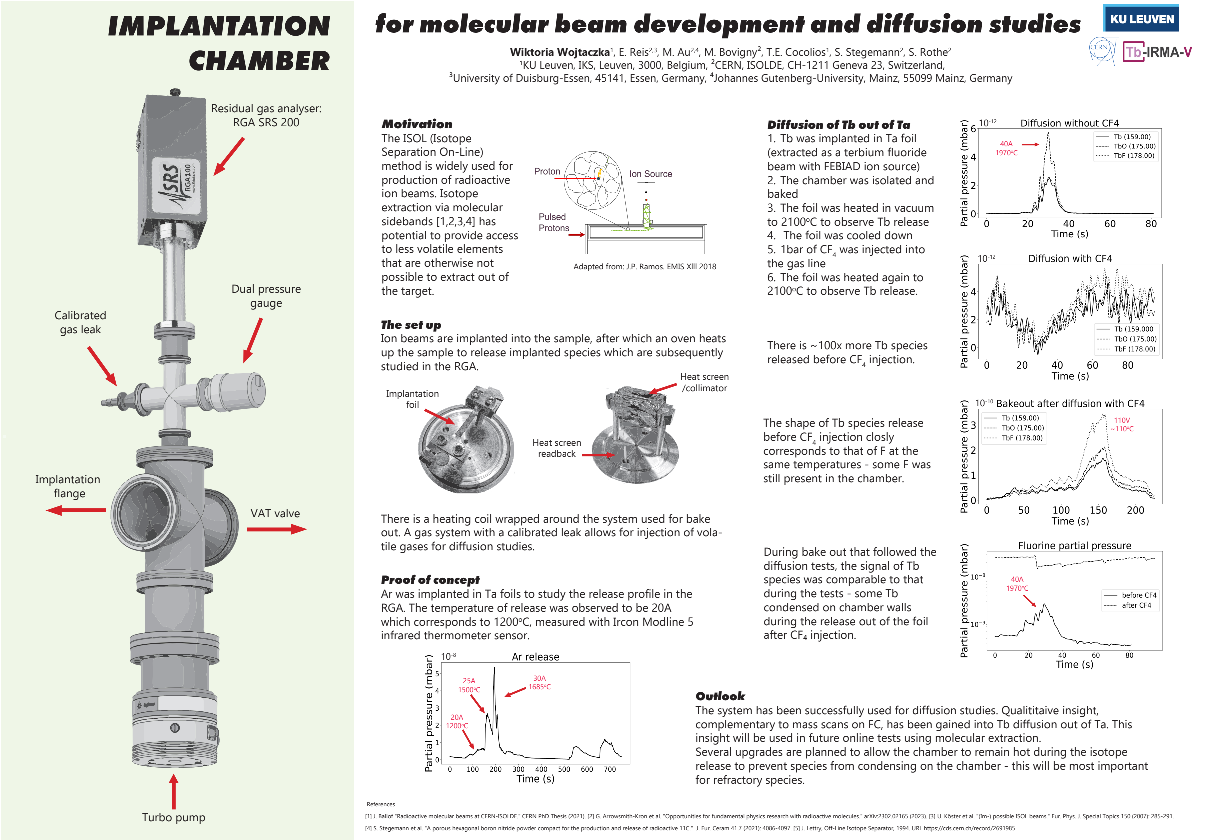
RILIS for molecules

Gas injection and mixing

- Remote control of HV gas systems and partial pressures






IMPLANTATION CHAMBER

for molecular beam development and diffusion studies

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Motivation
The ISOL (Isotope Separation On-Line) method is widely used for production of radioactive ion beams. Isotope extraction via molecular sidebands [1,2,3,4] has potential to provide access to less volatile elements that are otherwise not possible to extract out of the target.

Adapted from: J.P. Ramos. EMIS XIII 2018

The set up
Ion beams are implanted into the sample, after which an oven heats up the sample to release implanted species which are subsequently studied in the RGA.

There is a heating coil wrapped around the system used for bake out. A gas system with a calibrated leak allows for injection of volatile gases for diffusion studies.

Proof of concept
Ar was implanted in Ta foils to study the release profile in the RGA. The temperature of release was observed to be 20A which corresponds to 1200°C, measured with Ircon Modline 5 infrared thermometer sensor.

Diffusion of Tb out of Ta
1. Tb was implanted in Ta foil (extracted as a terbium fluoride beam with FEBIAD ion source)
2. The chamber was isolated and baked
3. The foil was heated in vacuum to 2100°C to observe Tb release
4. The foil was cooled down
5. 1bar of CF_4 was injected into the gas line
6. The foil was heated again to 2100°C to observe Tb release.

There is ~100x more Tb species released before CF_4 injection.

The shape of Tb species release before CF_4 injection closly corresponds to that of F at the same temperatures - some F was still present in the chamber.

During bake out that followed the diffusion tests, the signal of Tb species was comparable to that during the tests - some Tb condensed on chamber walls during the release out of the foil after CF_4 injection.

Outlook
The system has been successfully used for diffusion studies. Qualititaive insight, complementary to mass scans on FC, has been gained into Tb diffusion out of Ta. This insight will be used in future online tests using molecular extraction.
Several upgrades are planned to allow the chamber to remain hot during the isotope release to prevent species from condensing on the chamber - this will be most important for refractory species.

References
[1] J. Ballof "Radioactive molecular beams at CERN-ISOLDE." CERN PhD Thesis (2021). [2] G. Arrowsmith-Kron et al. "Opportunities for fundamental physics research with radioactive molecules." arXiv:2302.02165 (2023). [3] U. Köster et al. "(Im-) possible ISOL beams." Eur. Phys. J. Special Topics 150 (2007): 285-291.
[4] S. Stegemann et al. "A porous hexagonal boron nitride powder compact for the production and release of radioactive 11C." J. Eur. Ceram 41.7 (2021): 4086-4097. [5] J. Lettry, Off-Line Isotope Separator, 1994. URL https://cds.cern.ch/record/2691985

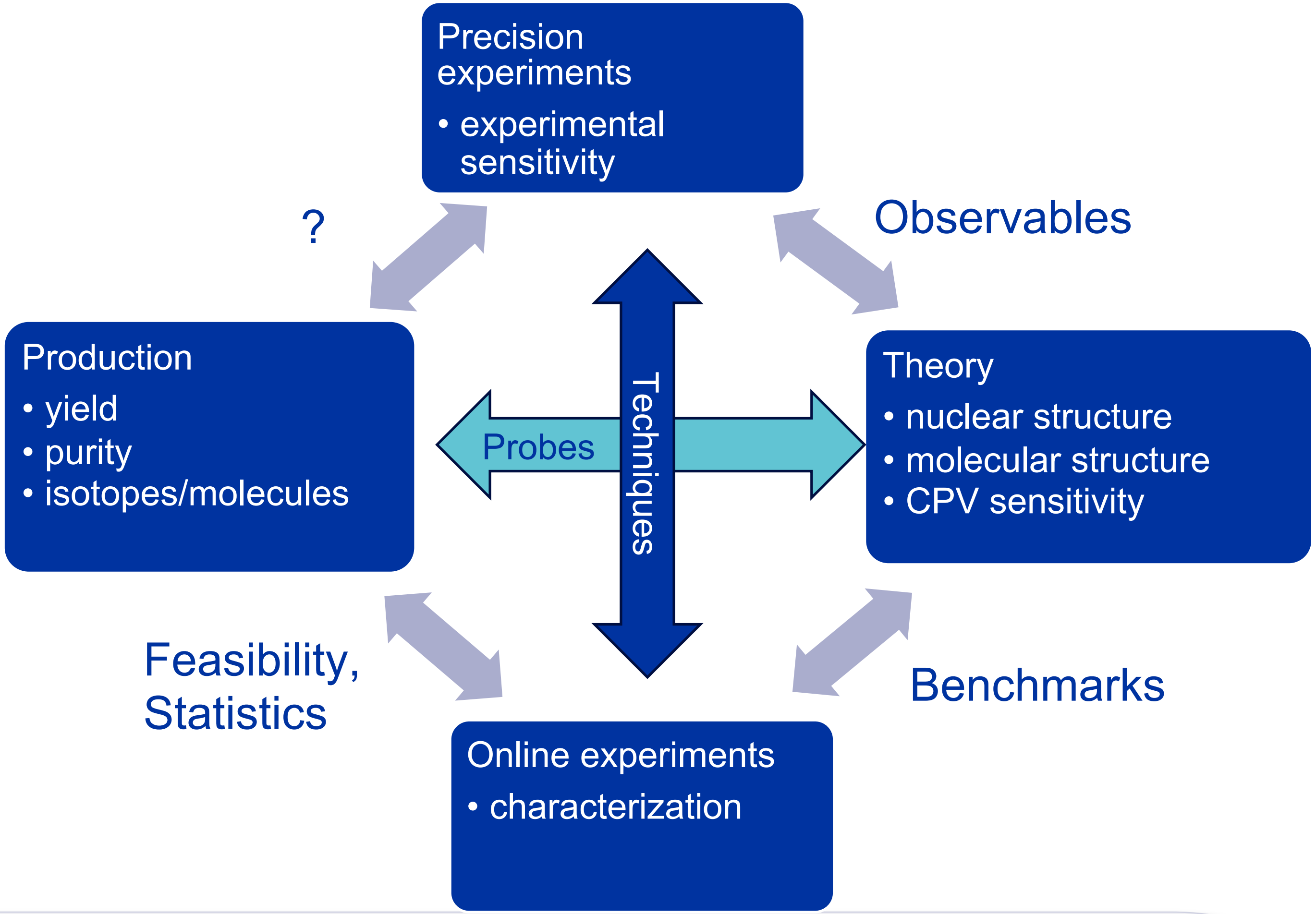
[1] Au et al. (2023) *NIM B.* **541** (144-147)
[2] Wojtaczka et al. (2023) *ICIS'23,* Victoria, Canada

Discussion

1 RIB facilities, ISOL method:
Radioactive species are available

2 RaF, AcFx:
Initial characterization of new probes

3 Offline developments: formation, characterization



4 Towards precision measurements: a question to the workshop?

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LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

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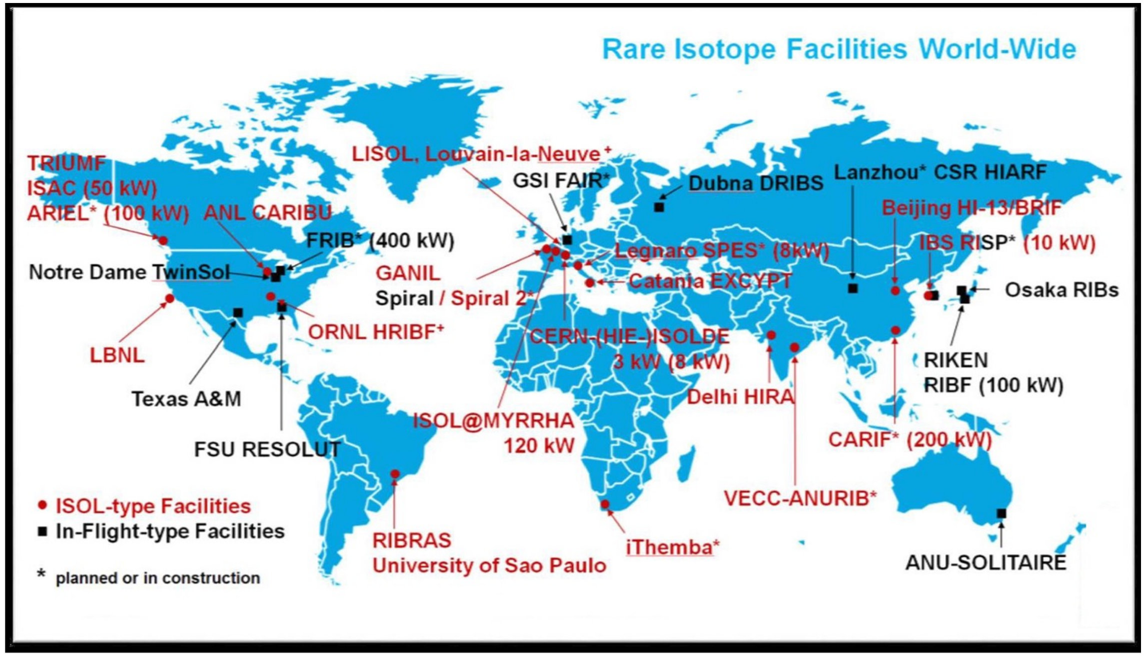

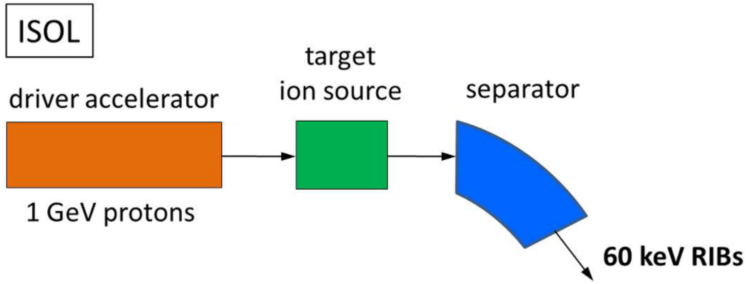

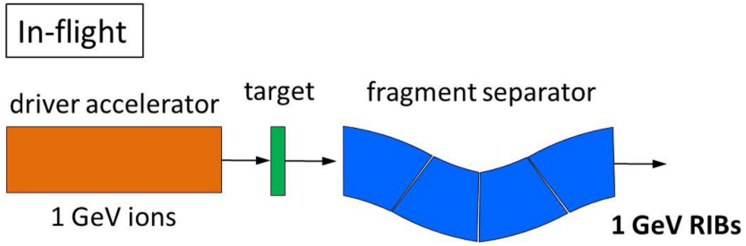

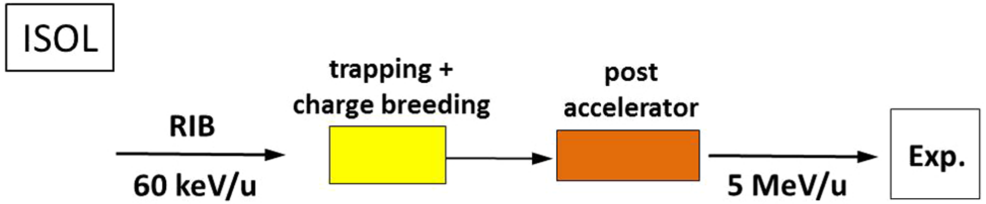

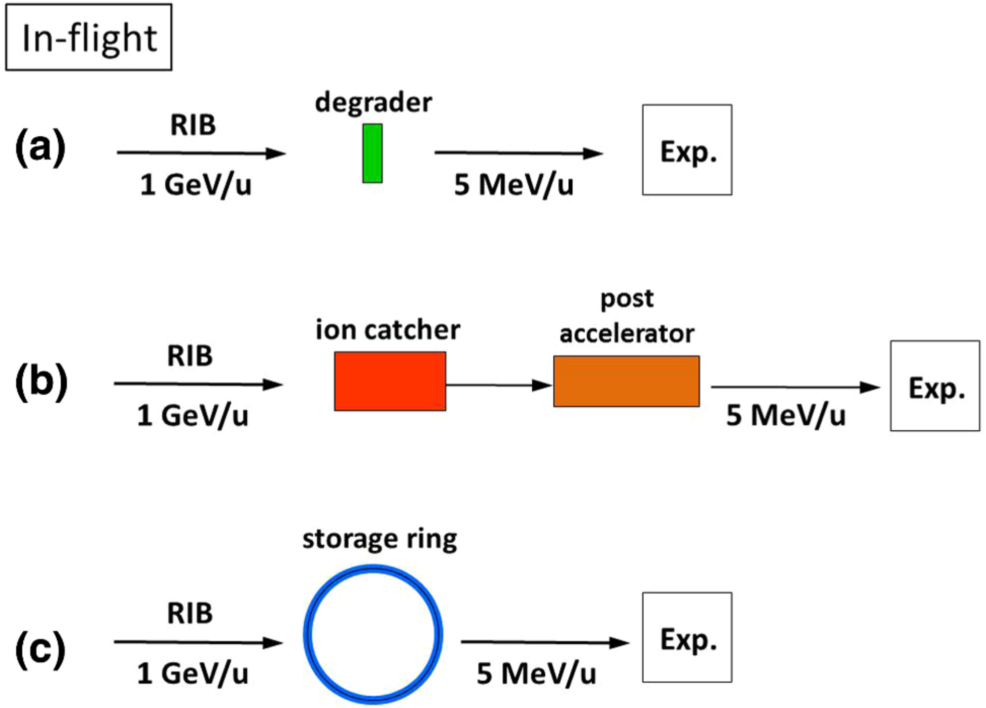

[1] Adamian, Antonenko, Diaz-Torres *et al.*, (2020) *EPJA*. **56**