Radioactive molecular ion beams at CERN-ISOLDE

Mia Au

CERN SY-STI | JGU Mainz FB09

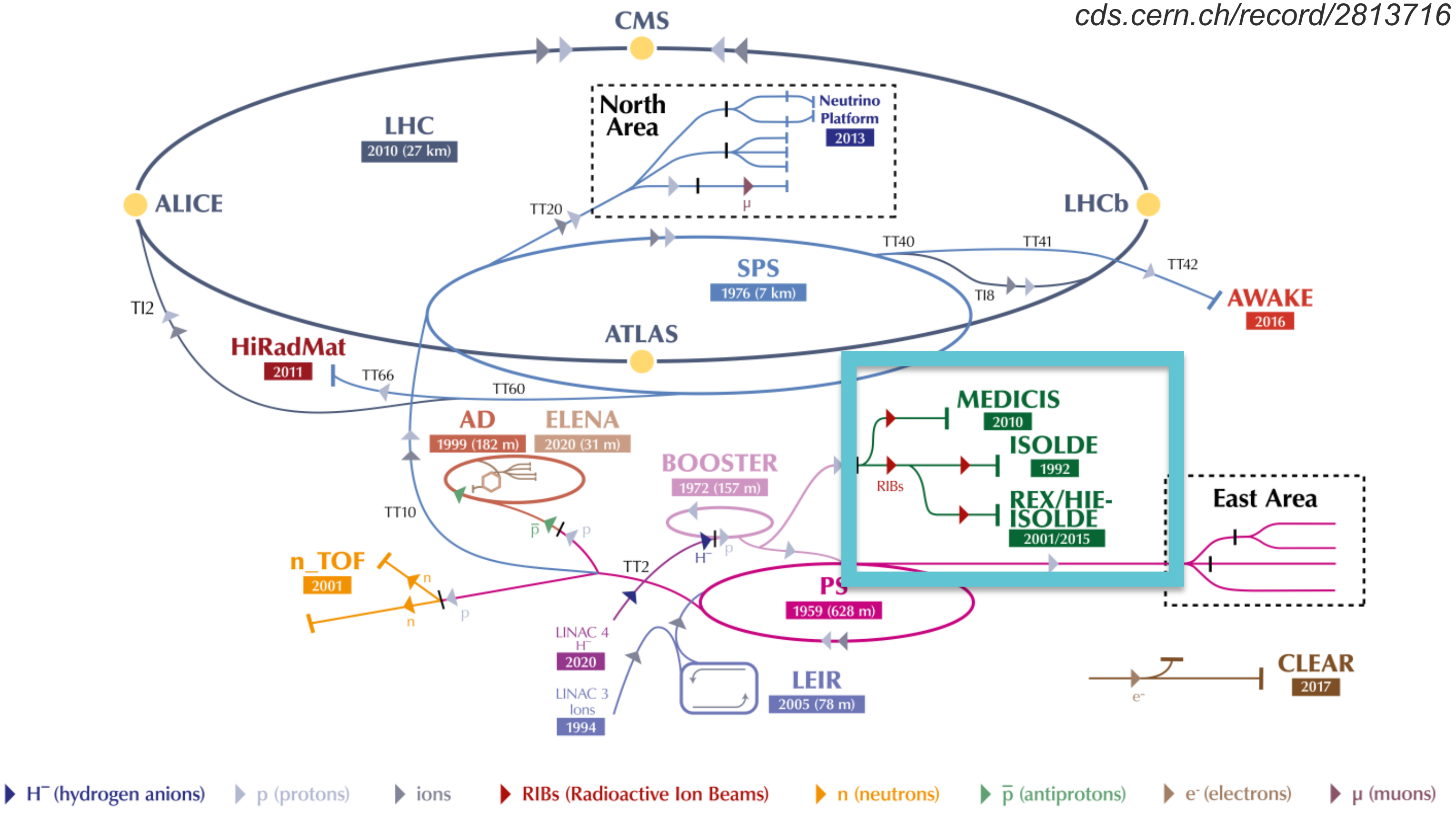
ICIS'23
International Conference on Ion Sources

Victoria, 21 September 2023

Outline: Radioactive molecular ion beams at CERN-ISOLDE

1. Introduction: CERN-ISOLDE
2. Why molecular beams?
3. Production and ionization
4. Results and conclusion

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

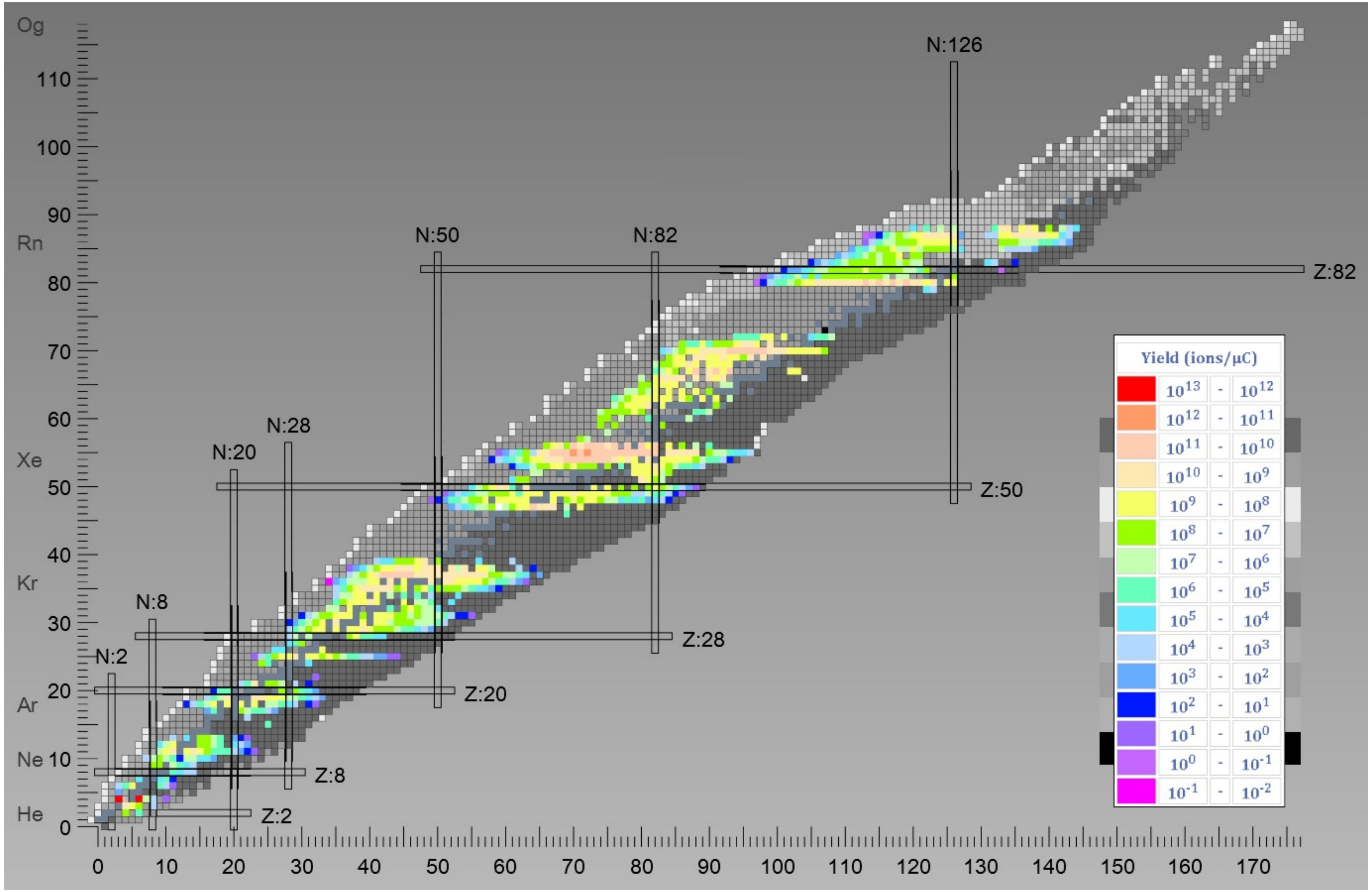


LHC - Large Hadron Collider // SPS - Super Proton Synchrotron // PS - Proton Synchrotron // AD - Antiproton Decelerator // CLEAR - CERN Linear Electron Accelerator for Research // AWAKE - Advanced WAKefield Experiment // ISOLDE - Isotope Separator OnLine // REX/HIE-ISOLDE - Radioactive EXperiment/High 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

CERN-ISOLDE

>1000 isotopes and isomers

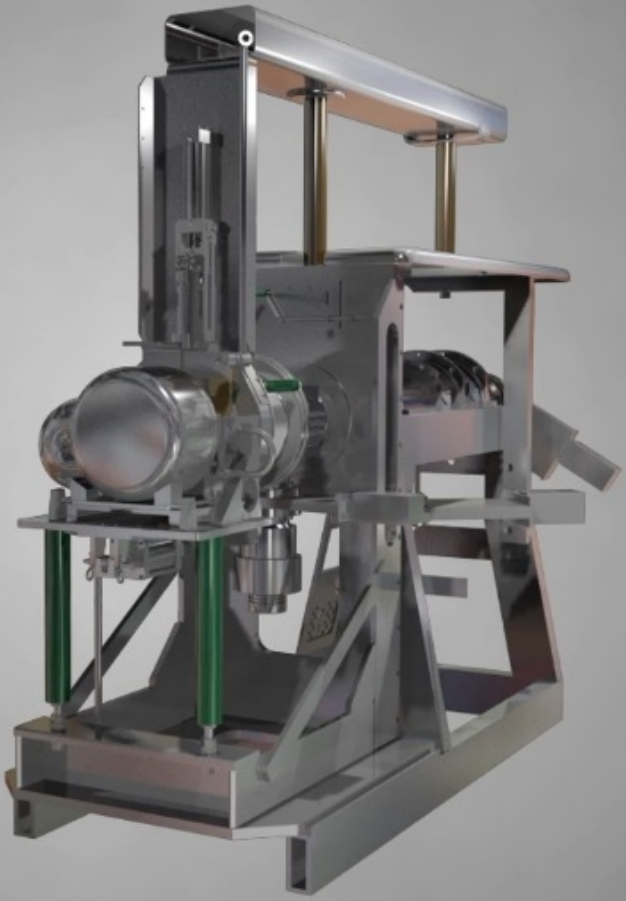
76 elements



Ballof *et.al*, (2020) *NIM B* **463**, 211-215
Stegemann *et al. (2023) NIM B* **541**, 169-172
cern.ch/isolde-yields

3852 nuclides, 226 stable
www.nucleonica.com
Dataset: JEFF-3.1 Nuclear Data Library, NEA (2023)

DOI 10.17181/cds.2289929



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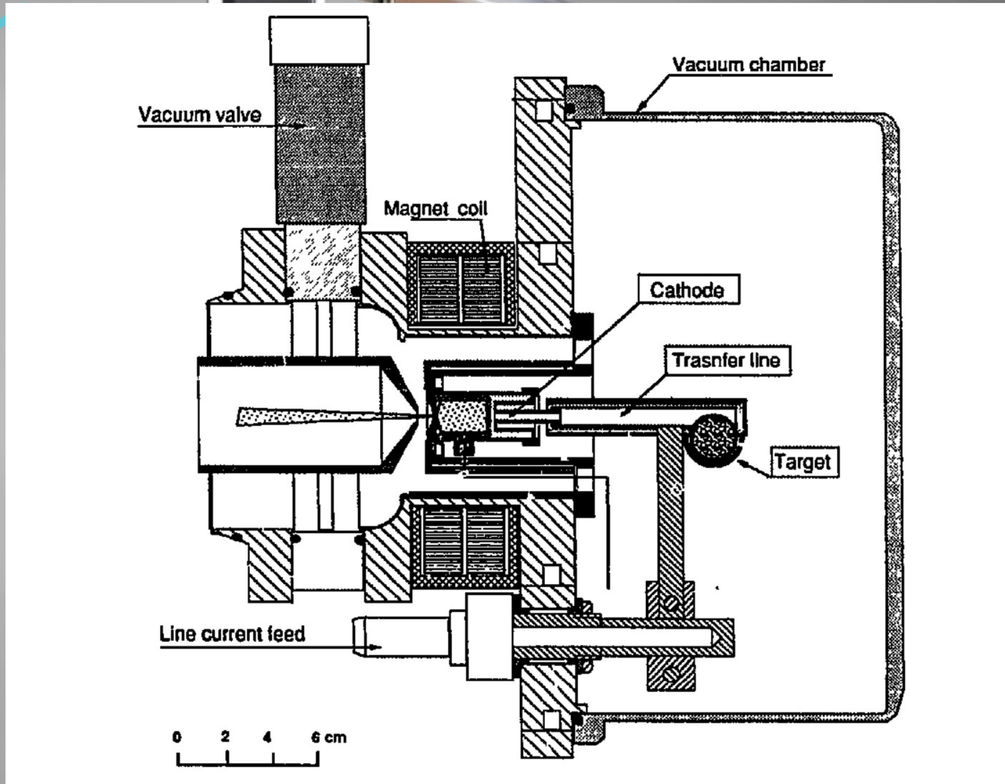

Fig. 1. Target and ion source assembly with plasma ion source MK5. The vacuum valve is part of the assembly.

Sundell, Ravn (1992) *NIM B.* **70** (160-164)

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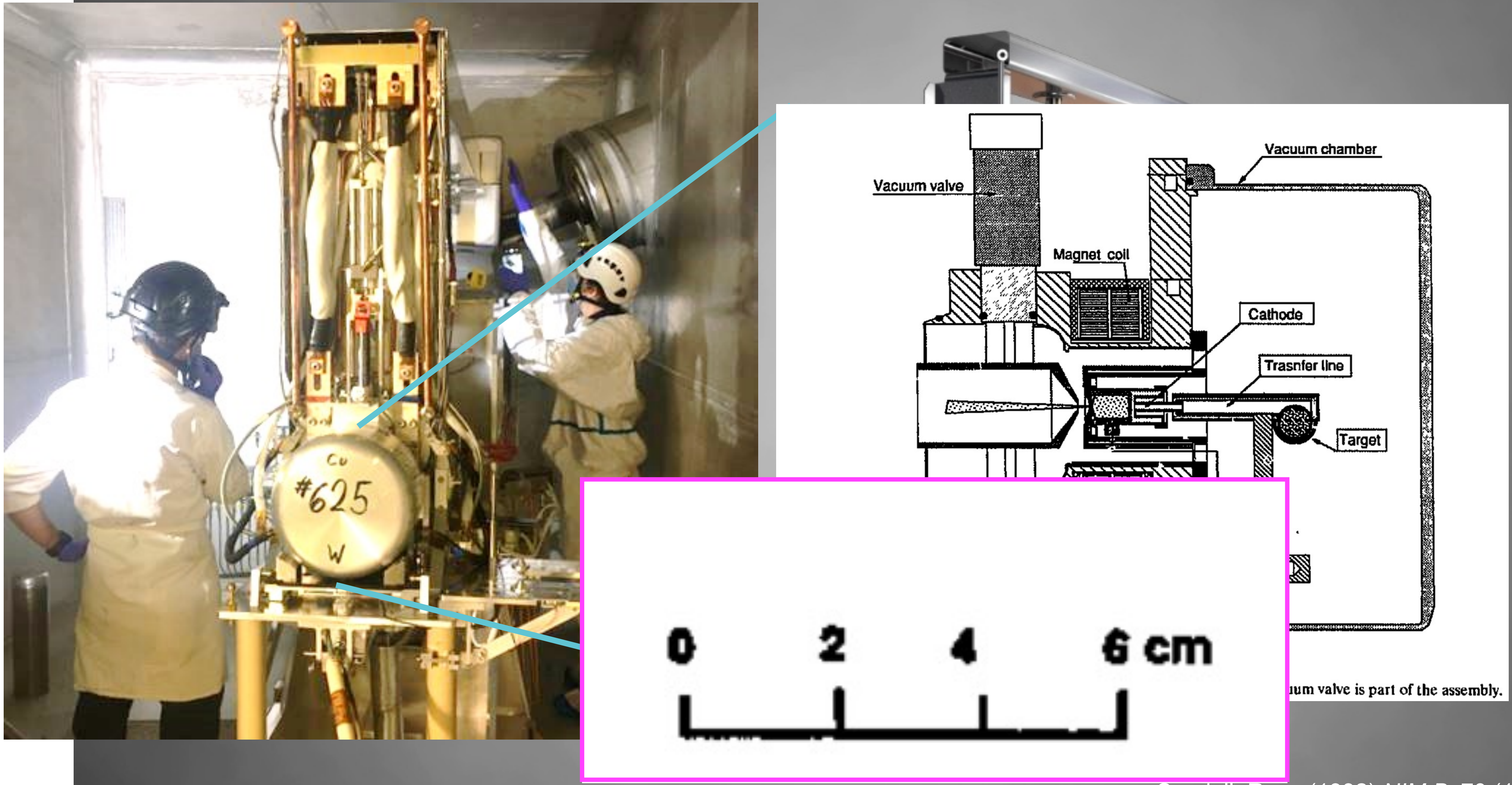

Sundell, Ravn (1992) *NIM B.* **70** (160-164)

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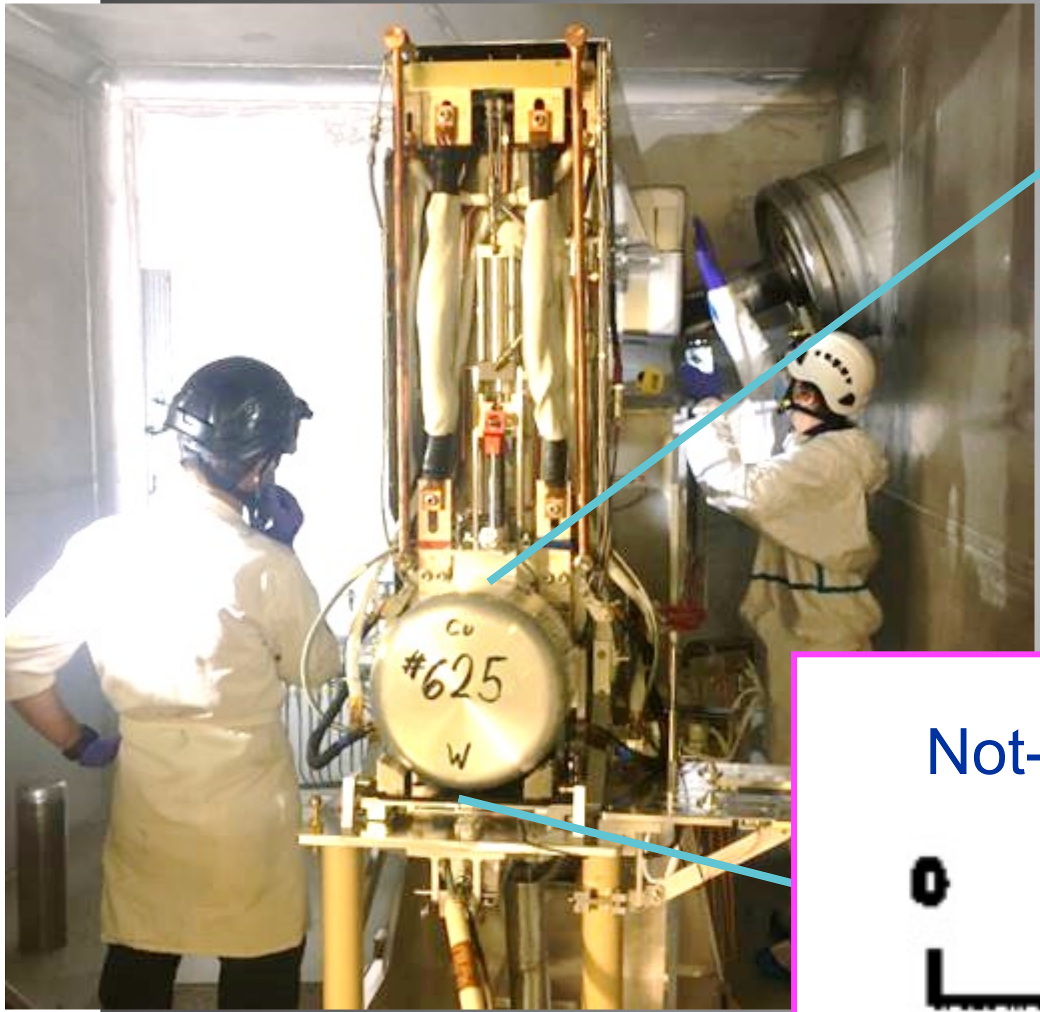

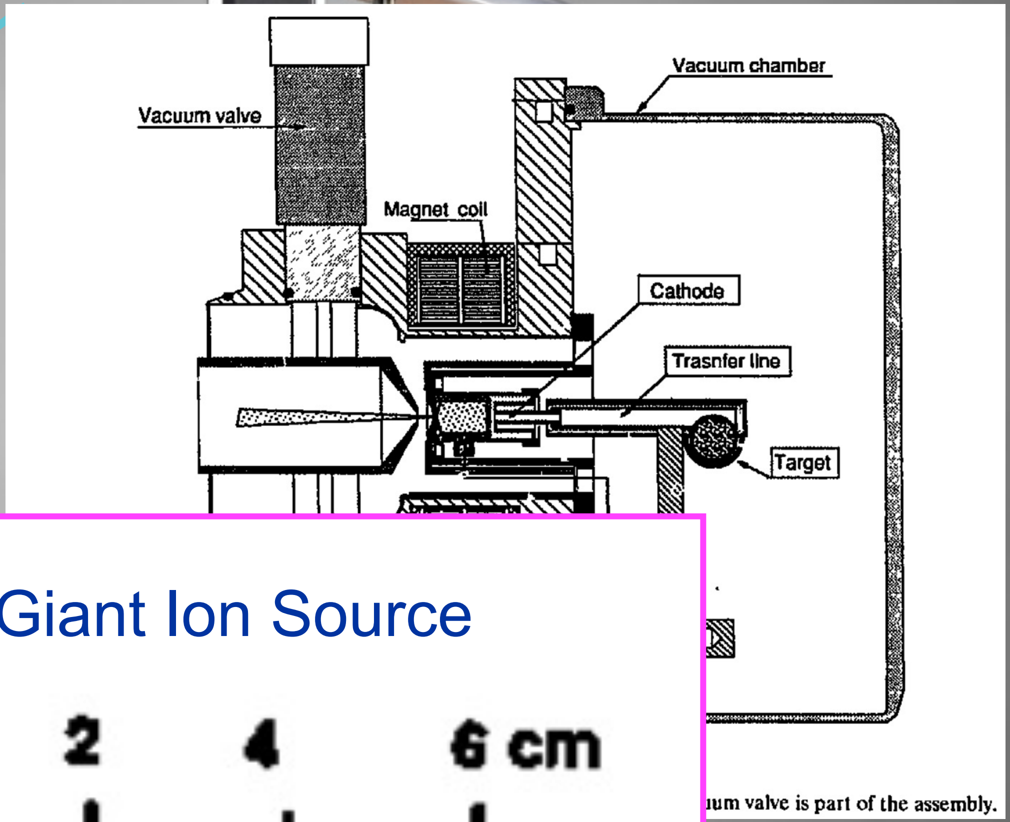

Not-Giant Ion Source

0 2 4 6 cm

Sundell, Ravn (1992) *NIM B.* **70** (160-164)

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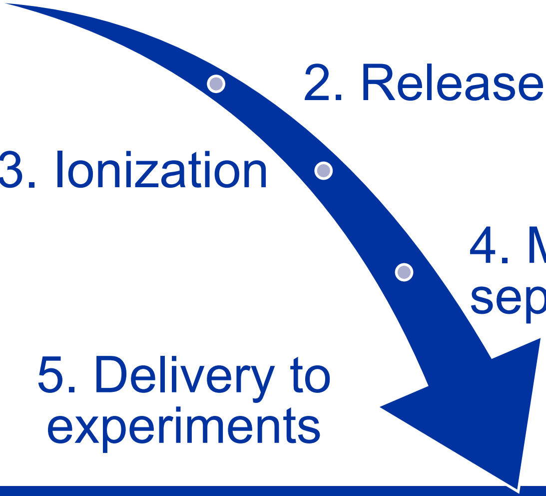
The Isotope Separation On-Line (ISOL) method



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

N_t – Number of target atoms
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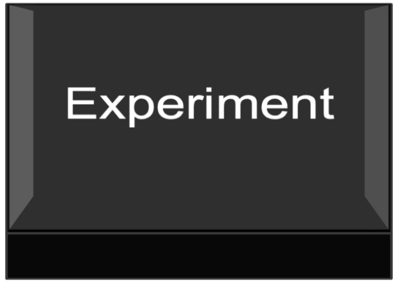

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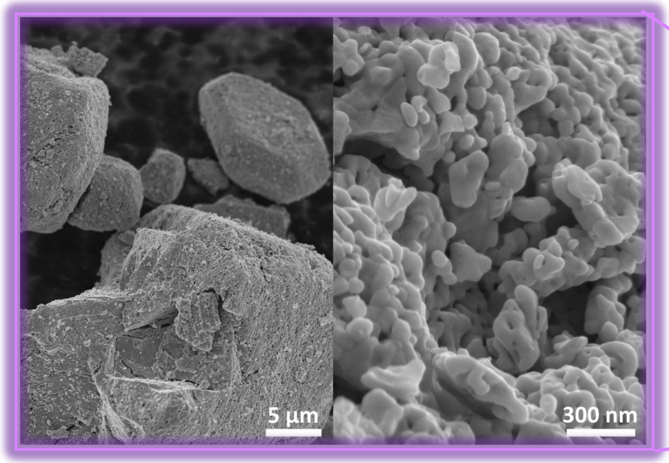
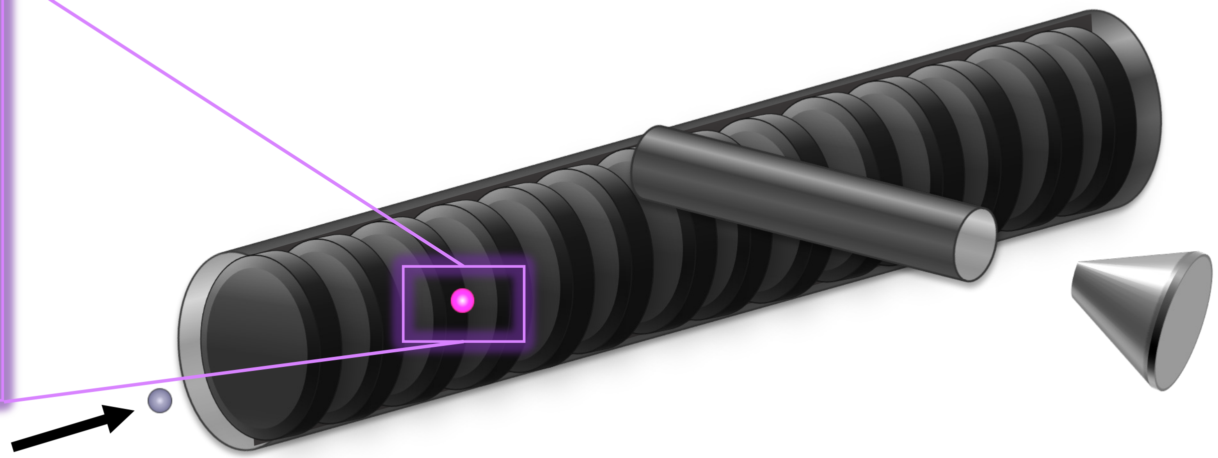
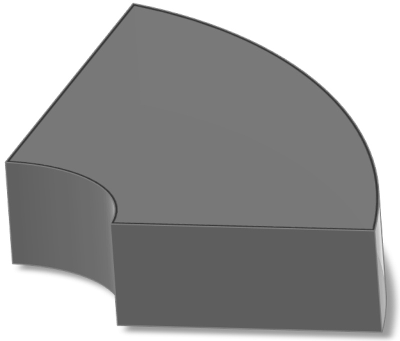
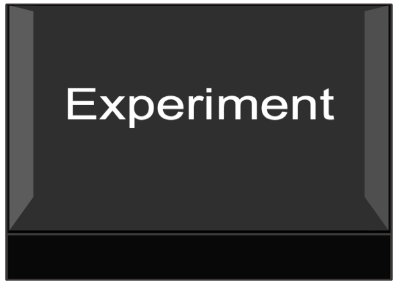


Figure published in Ramos *et al.*, (2020)
NIM B **463**, 201



Accelerated protons





1. Production
2. Release
3. Ionization
4. Mass separation
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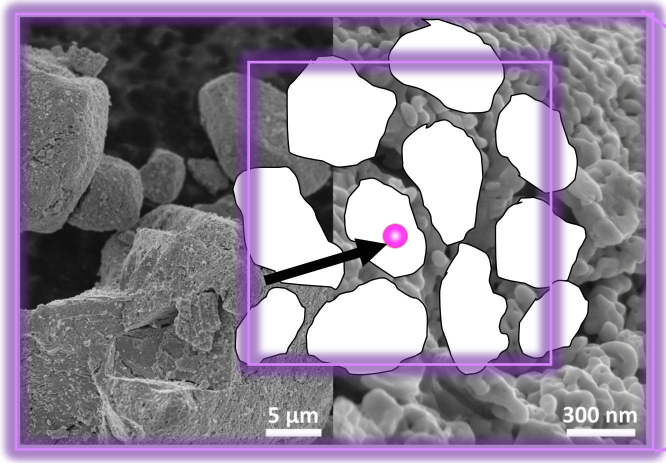

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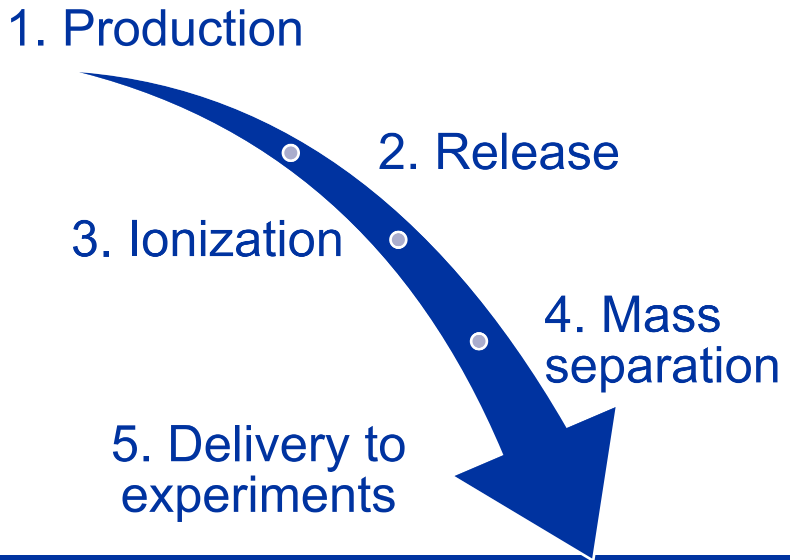

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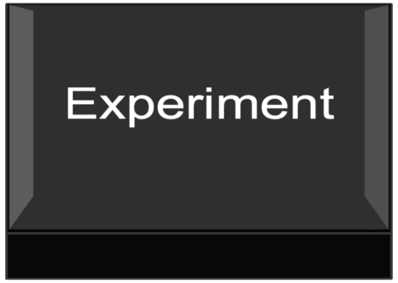

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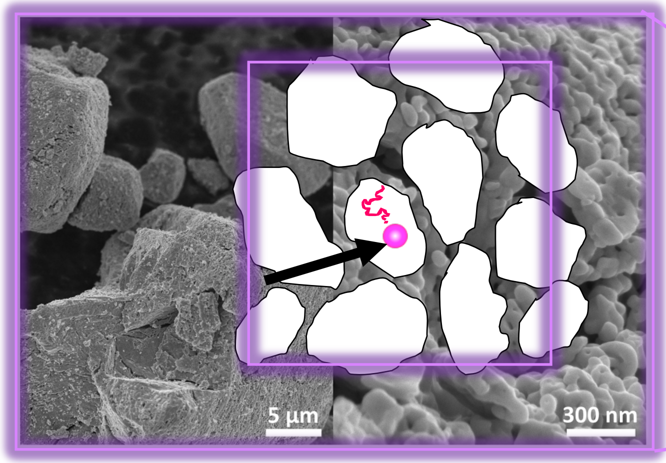

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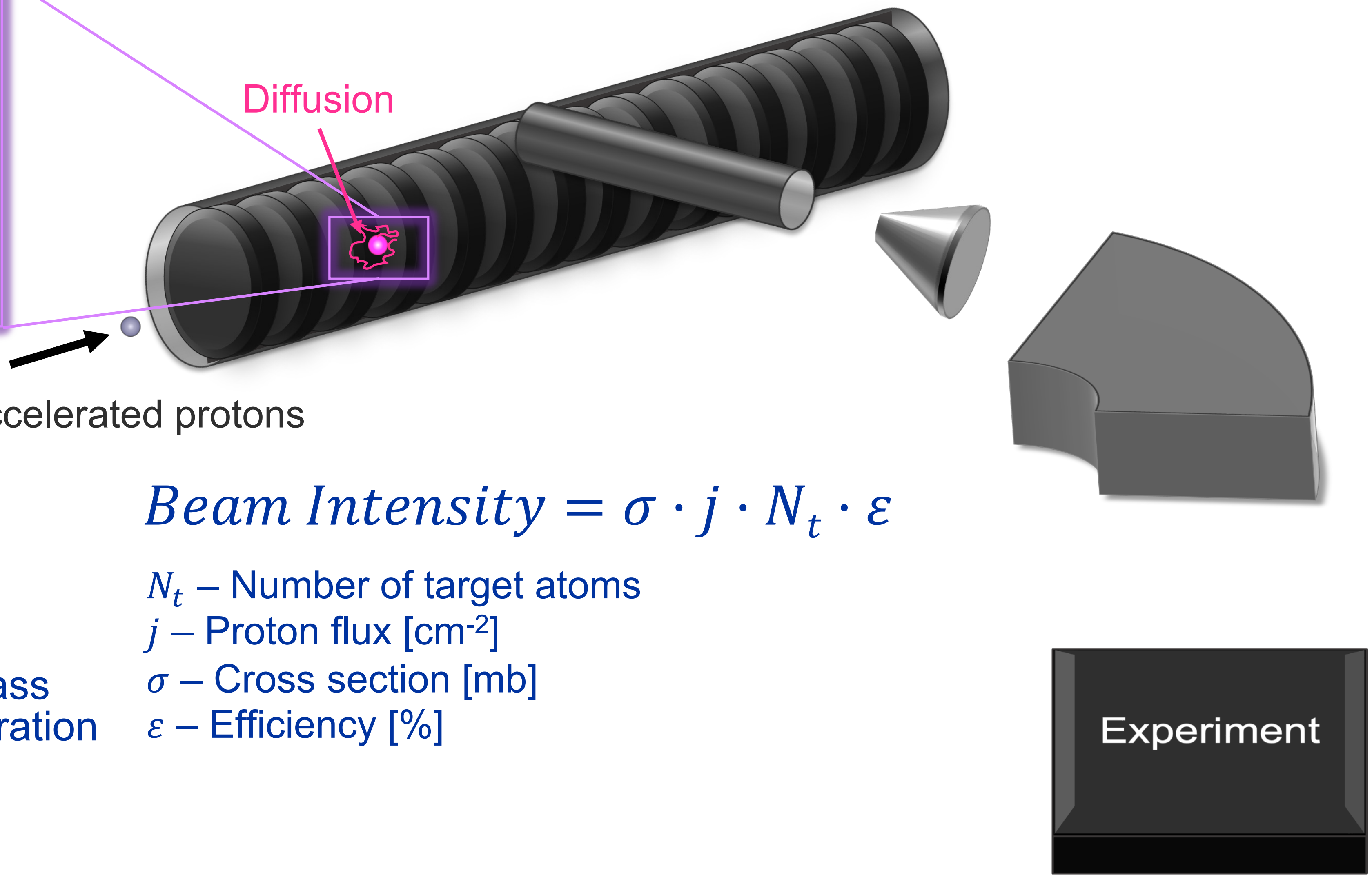

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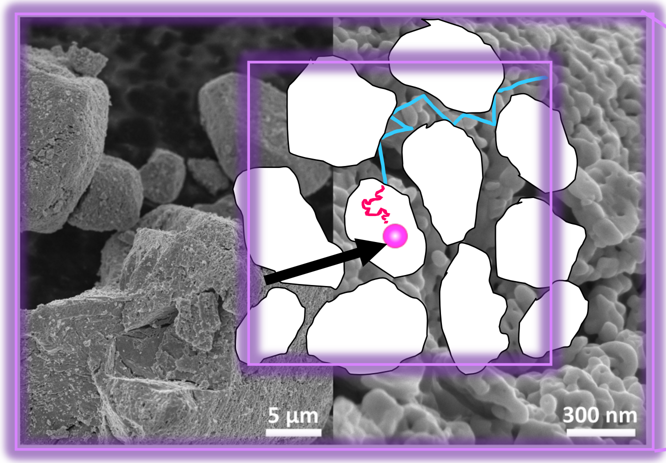

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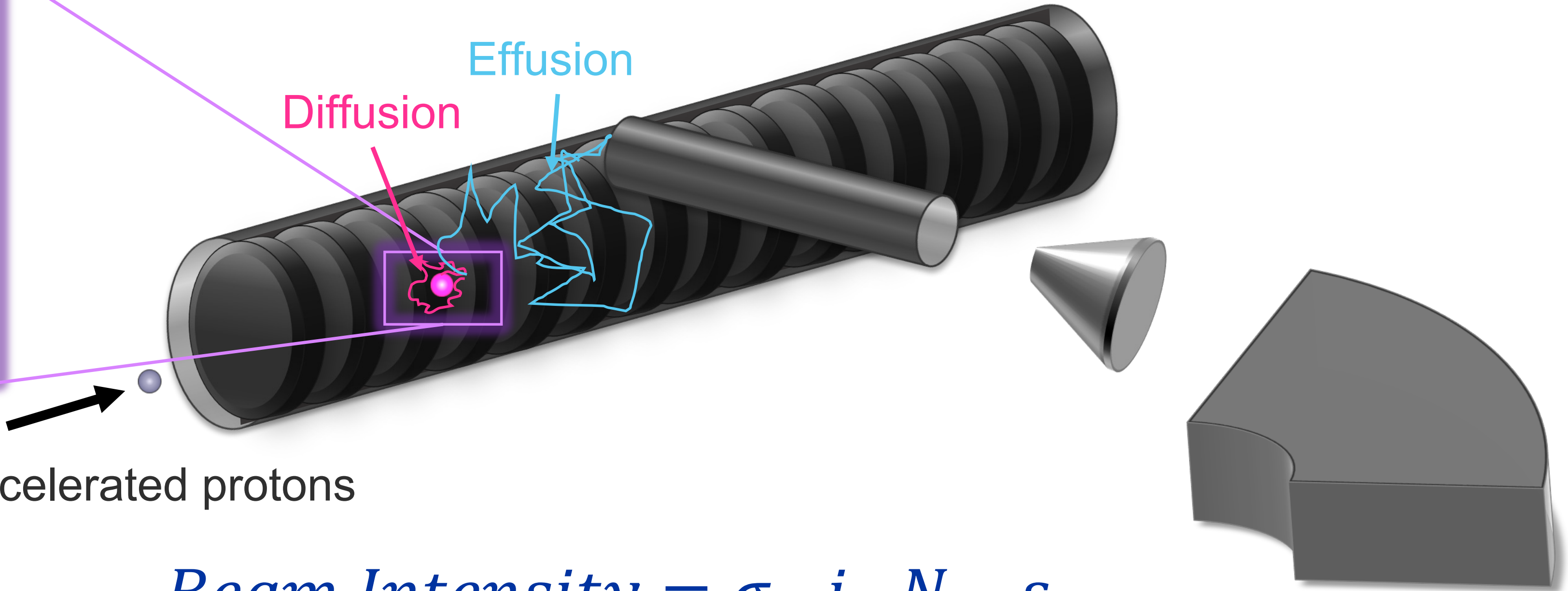

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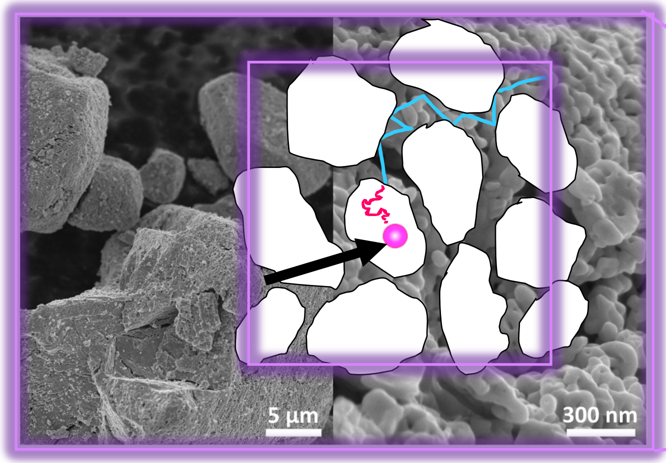
Experiment

The Isotope Separation On-Line (ISOL) method



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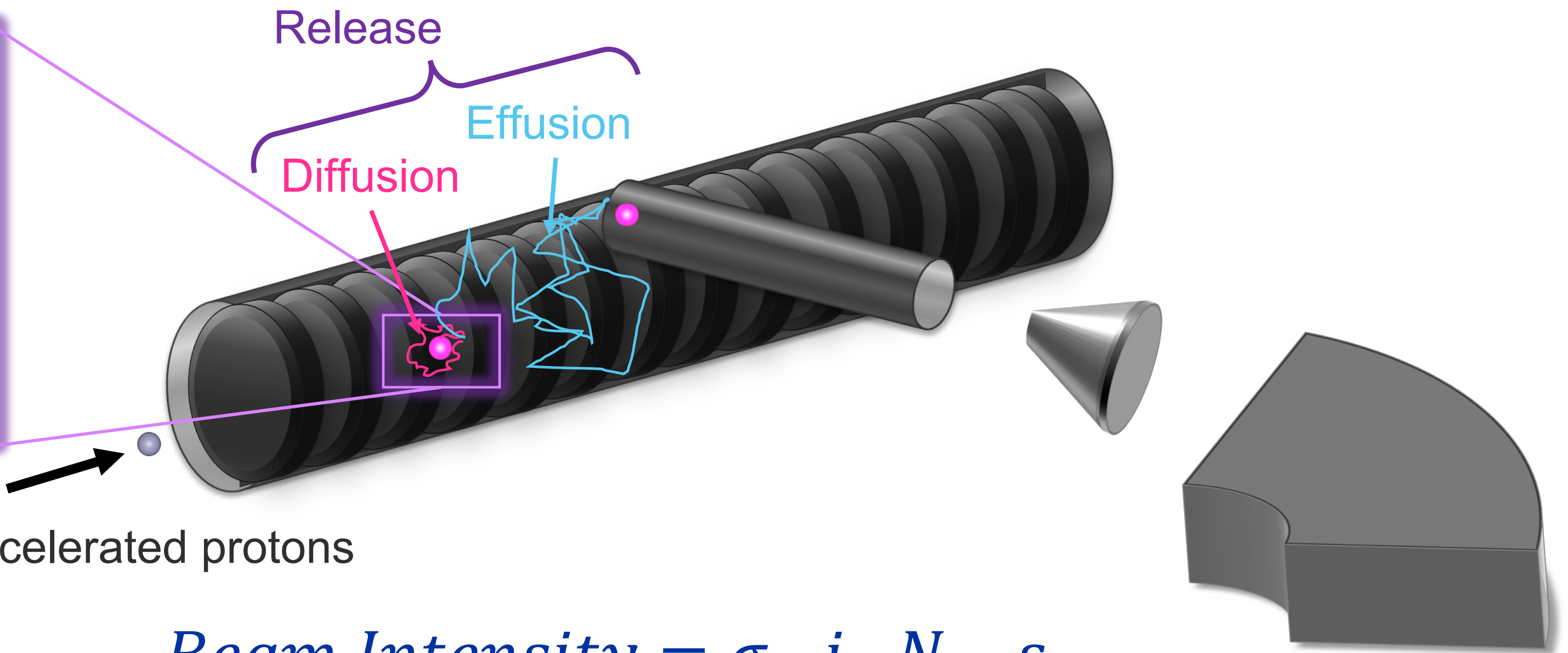

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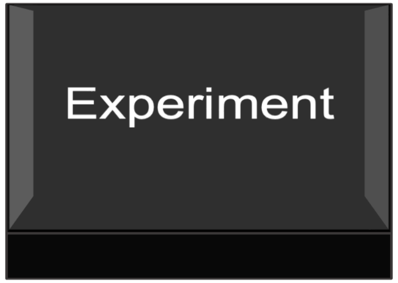
$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}$$



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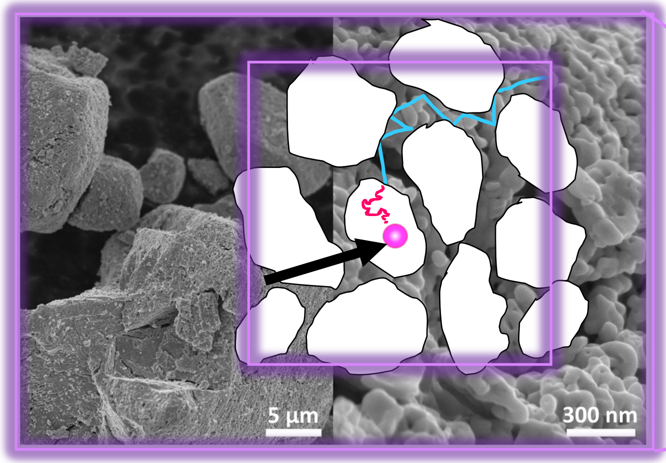

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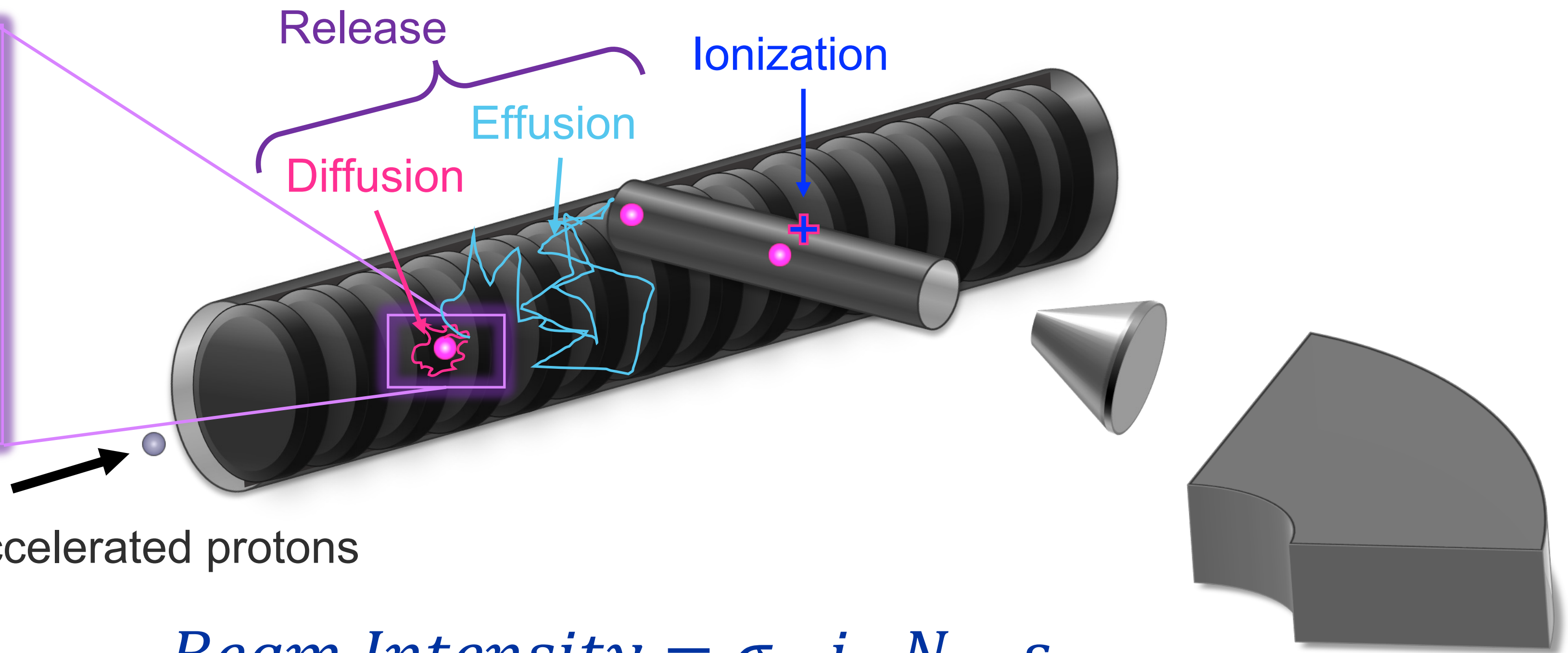

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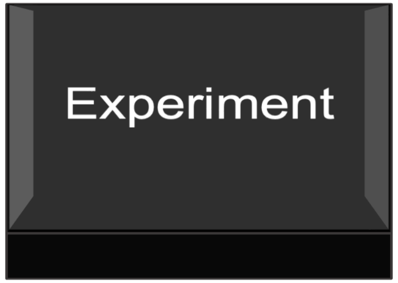

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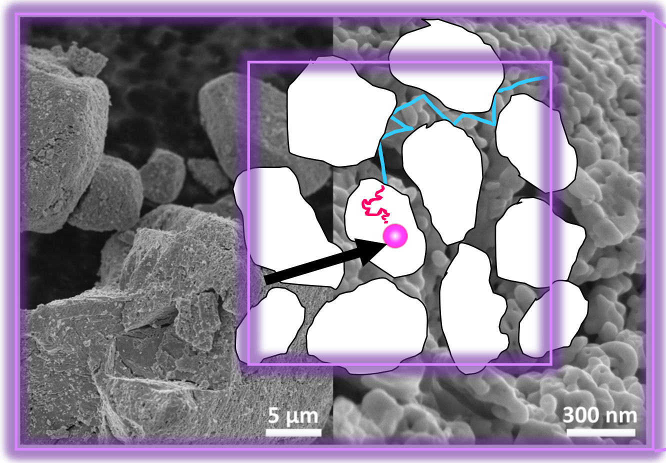

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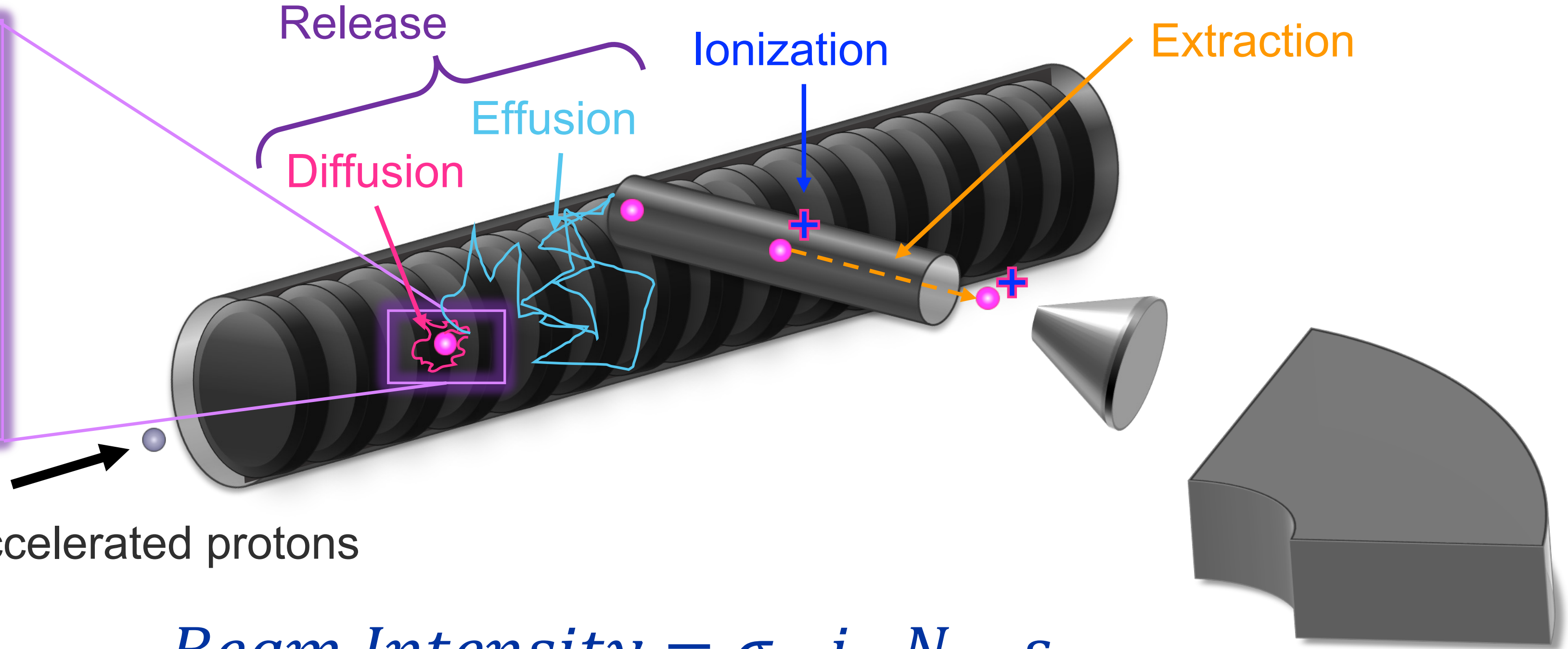

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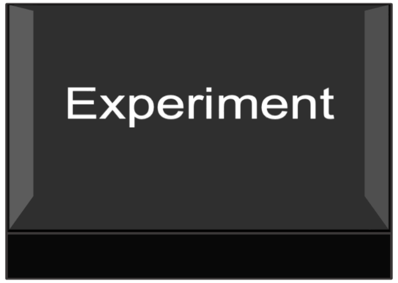

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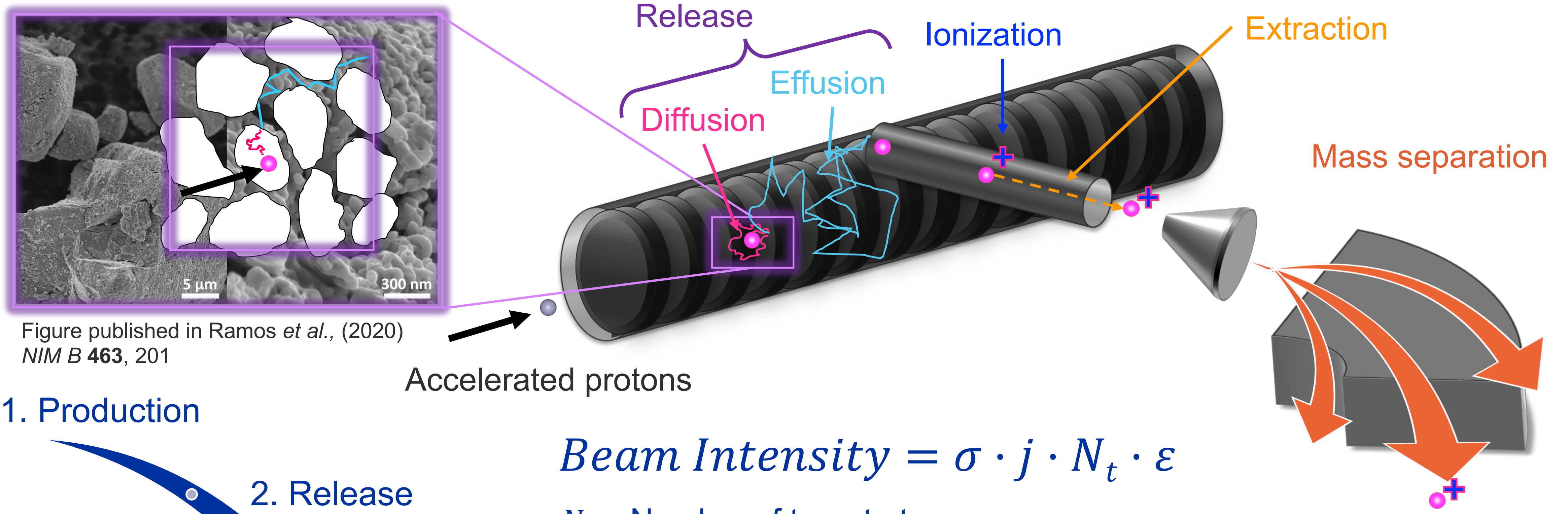

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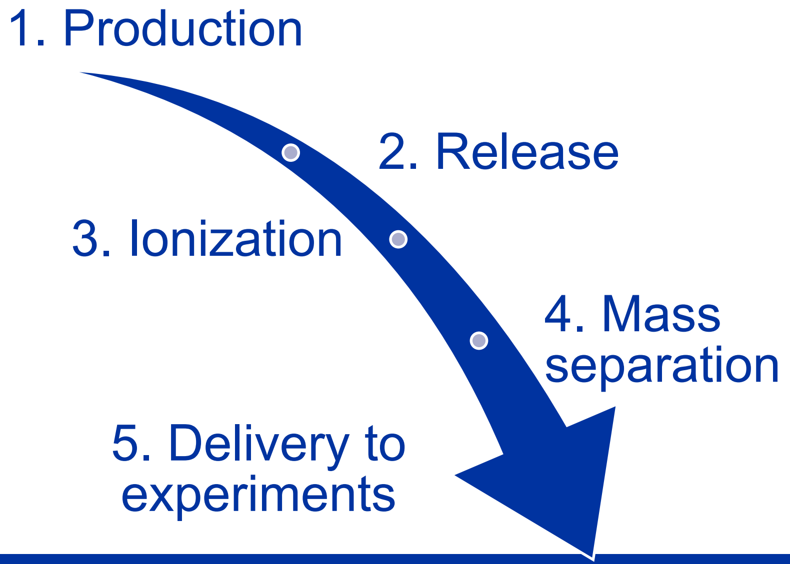

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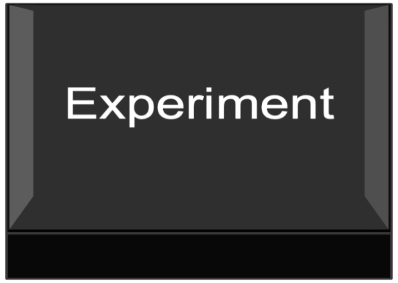

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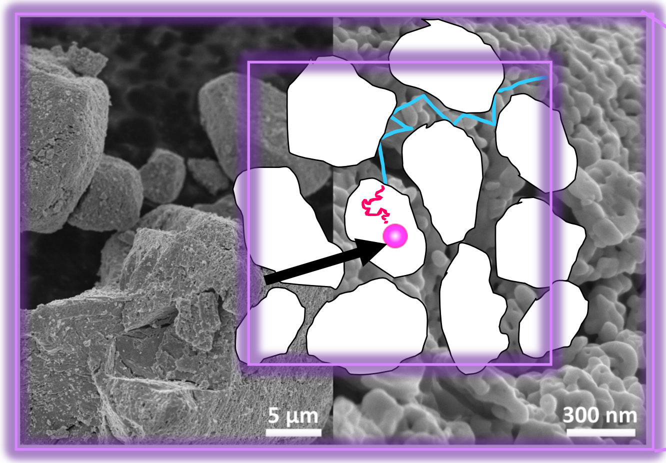

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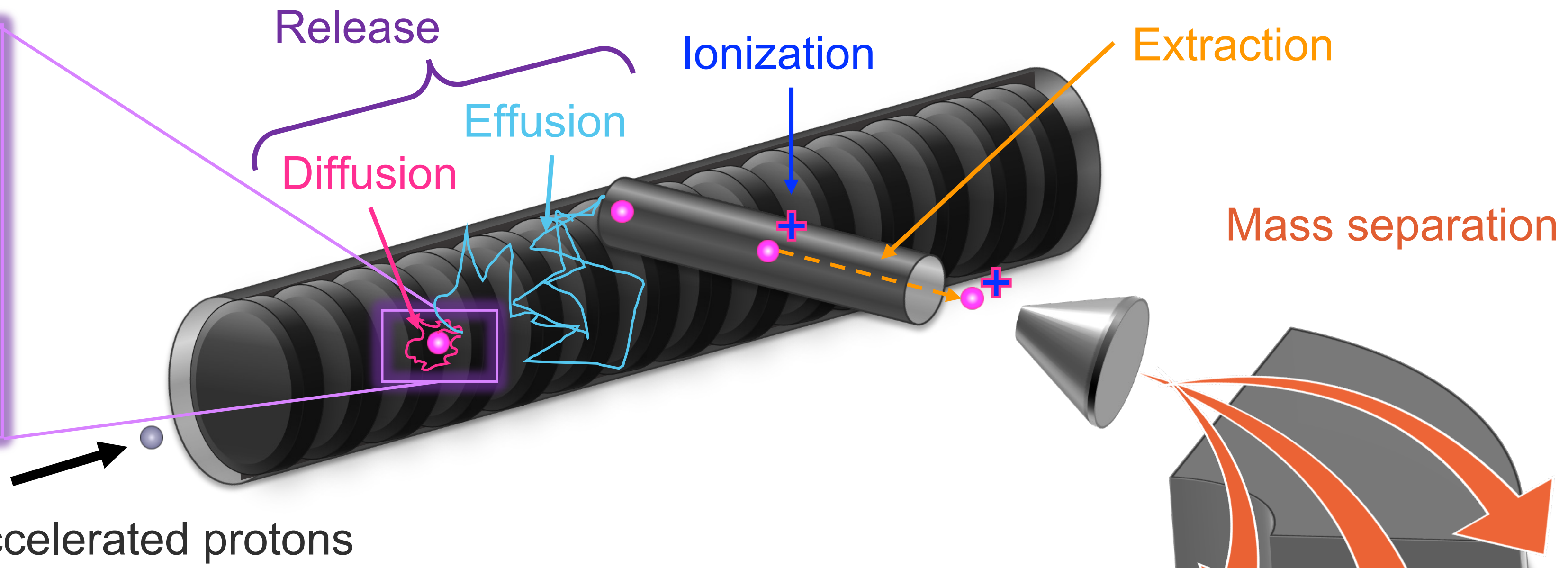

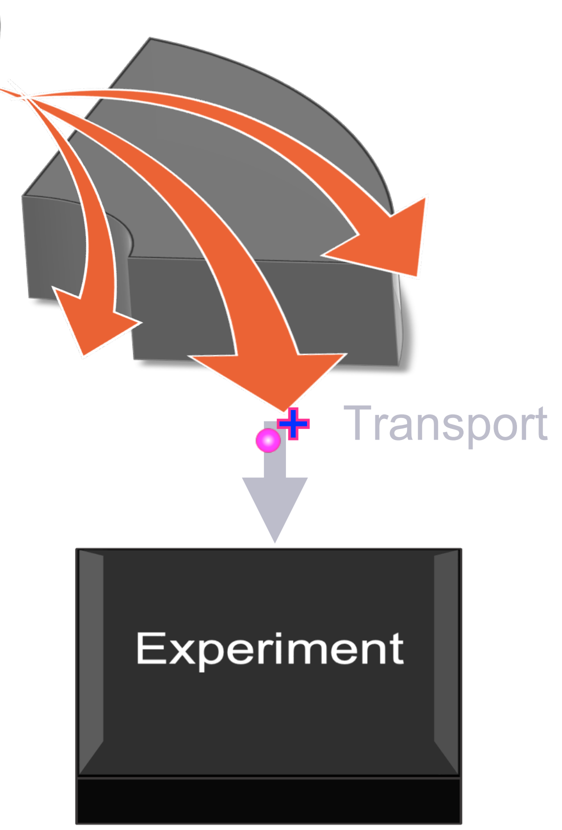

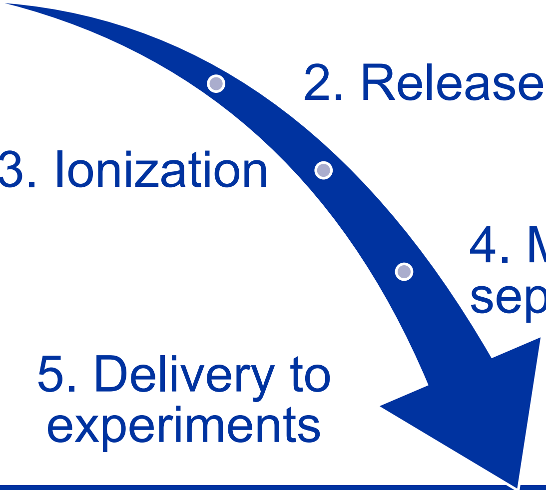

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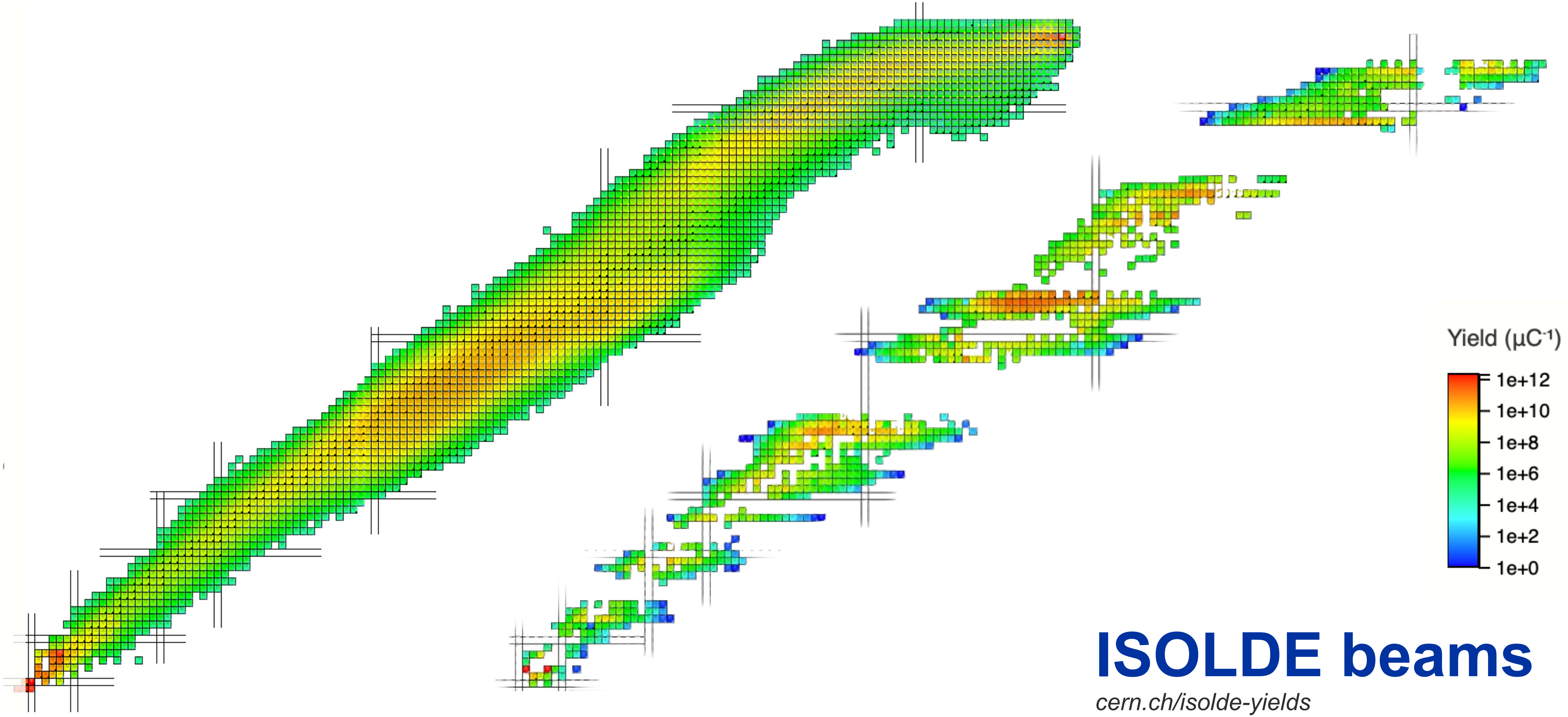
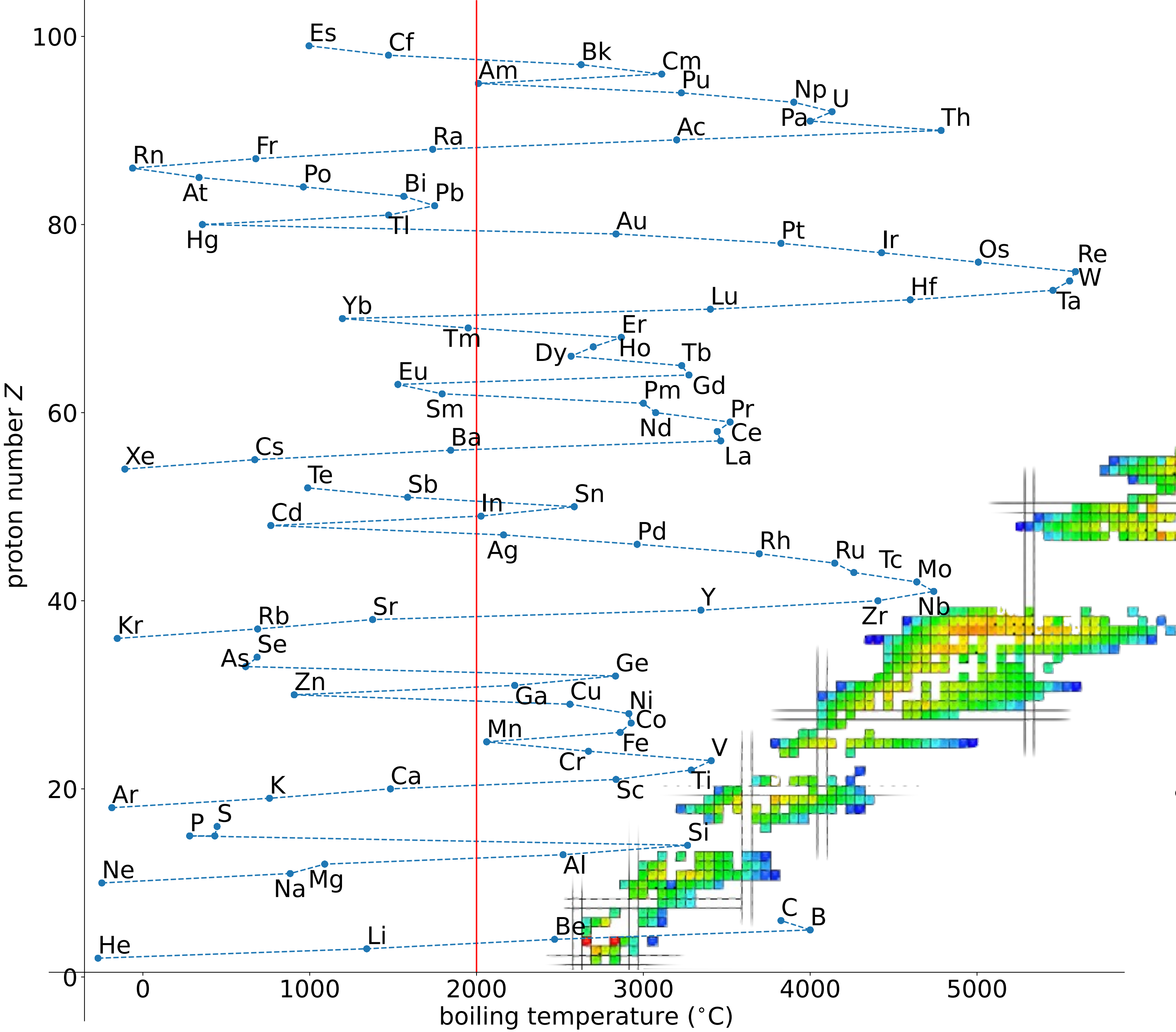
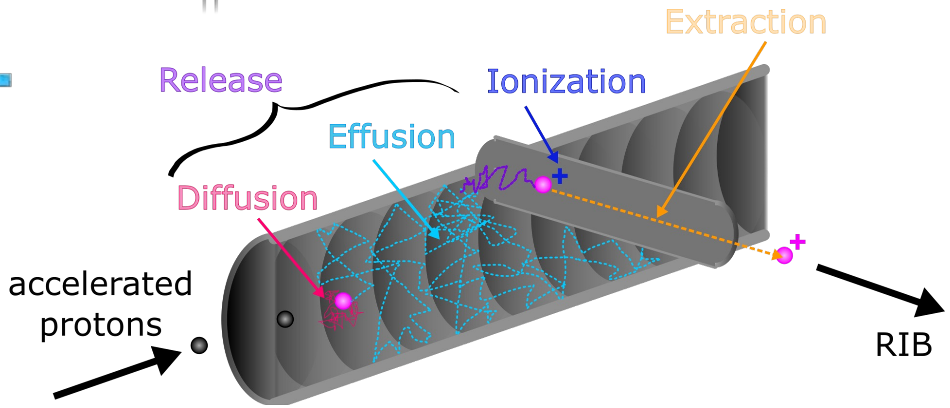


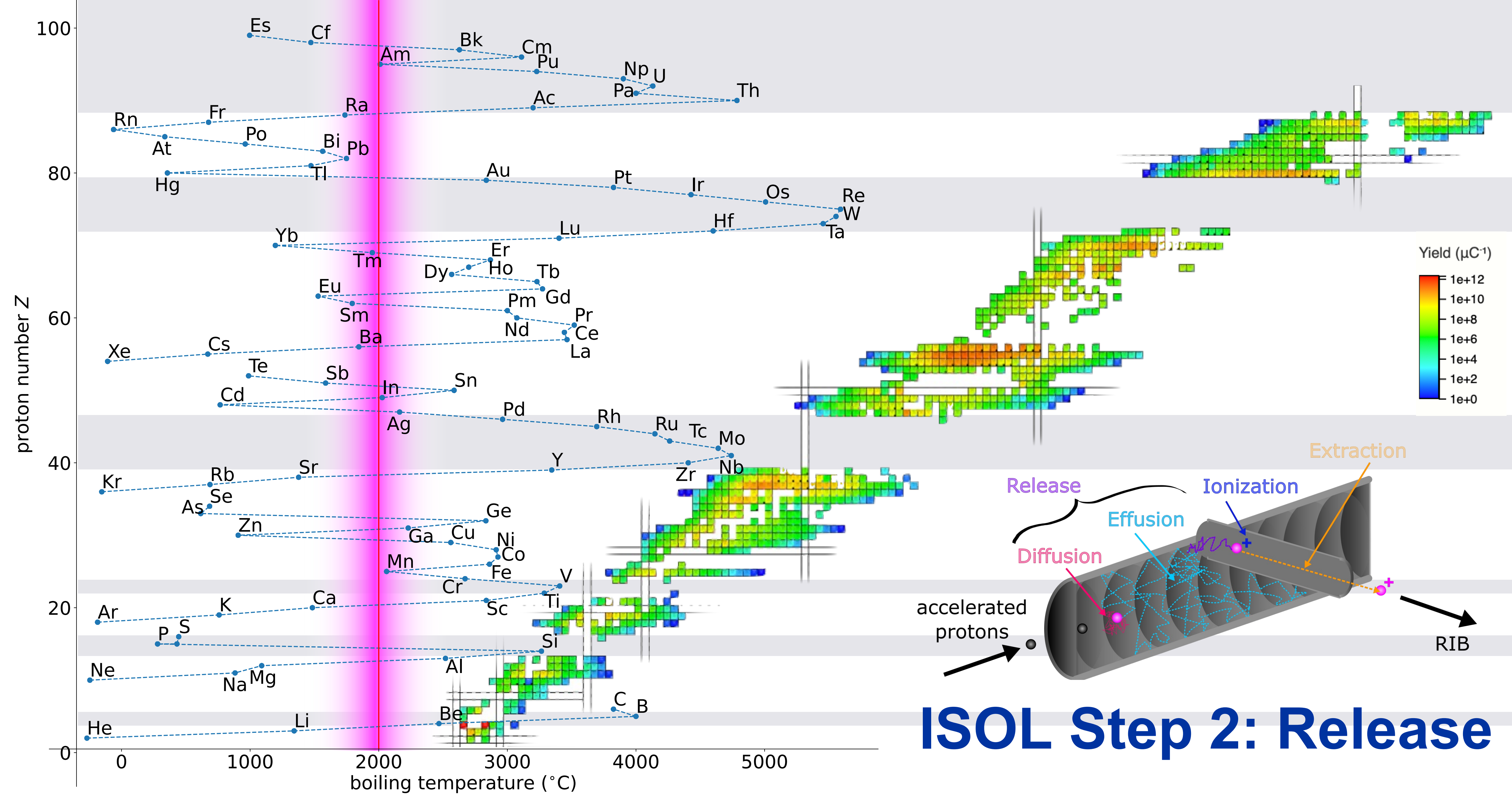
ISOLDE beams

cern.ch/isolde-yields



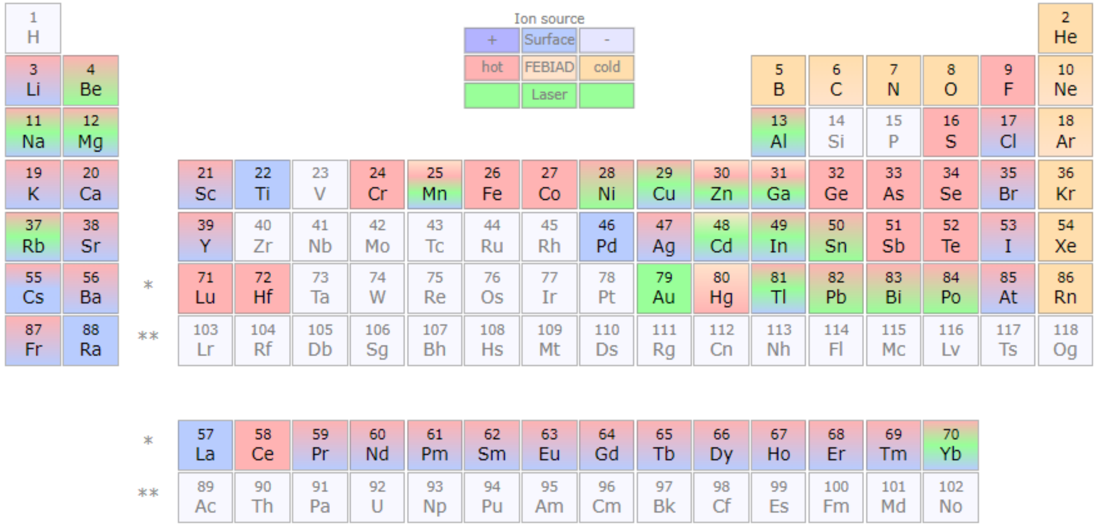


ISOL Step 2: Release



ISOL Step 2: Release

ISOL Step 3: Ionization

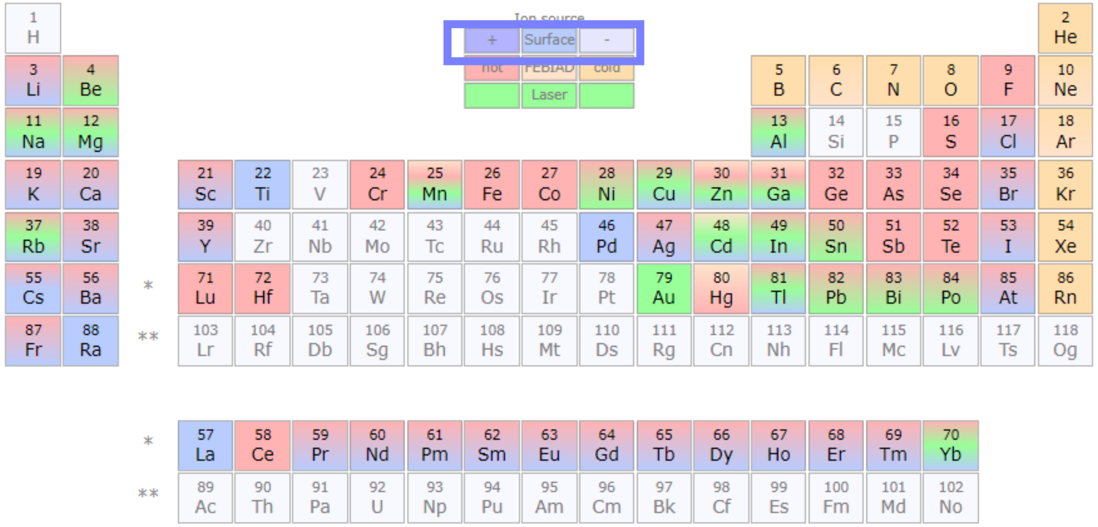


cern.ch/isolde-yields

Ion sources

- Surface ionization
- Electron impact ionization
- Resonance laser ionization

ISOL Step 3: Ionization



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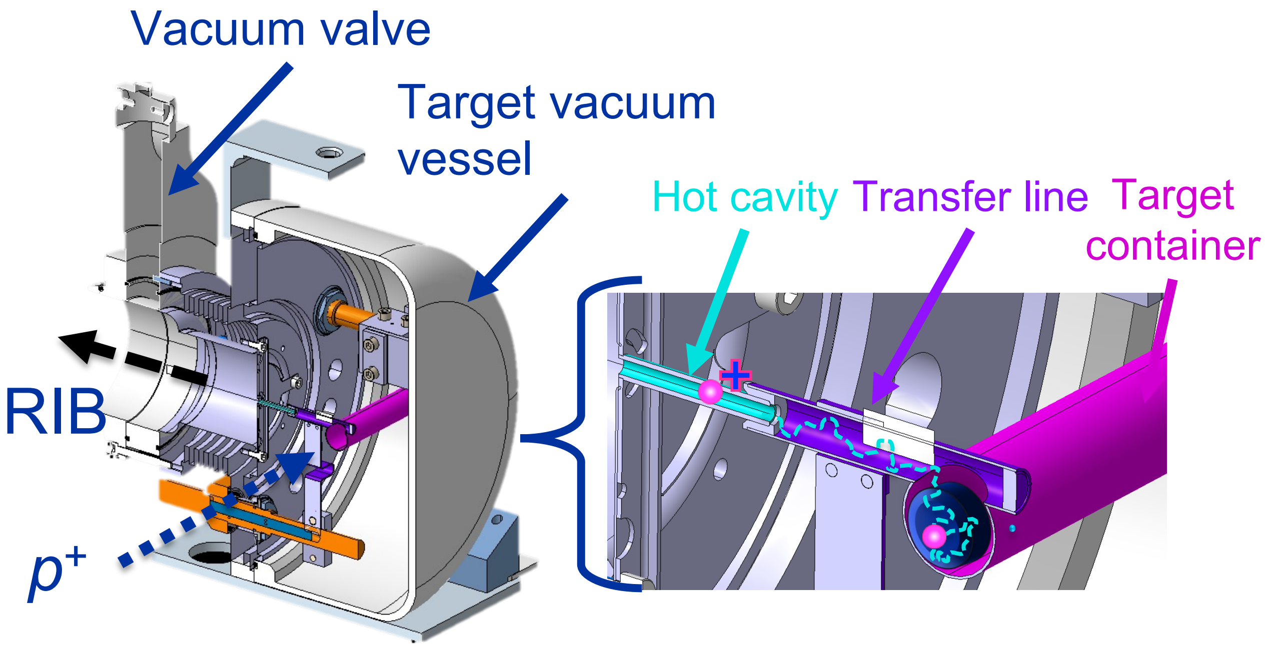
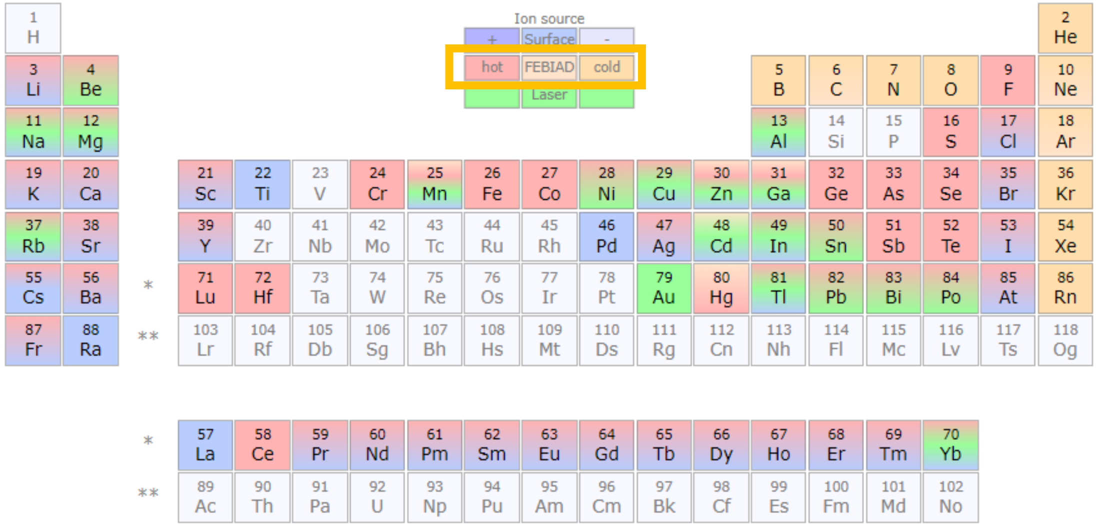


Ion sources

- Surface ionization
- Electron impact ionization
- Reson

First Ion Source at ISOL@MYRRHA with an Improved Thermal Profile - From Prototype to the First Experimental Validati...
Sophie Hurier

ISOL Step 3: Ionization



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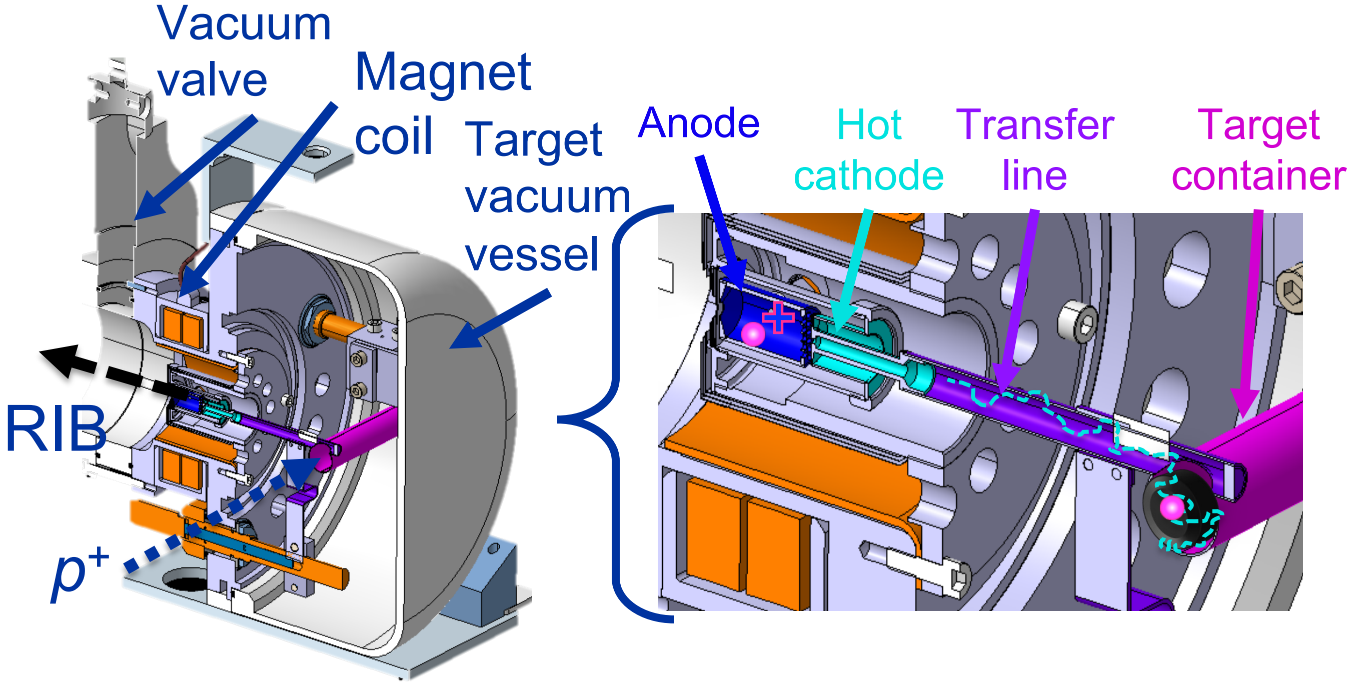

Ion sources

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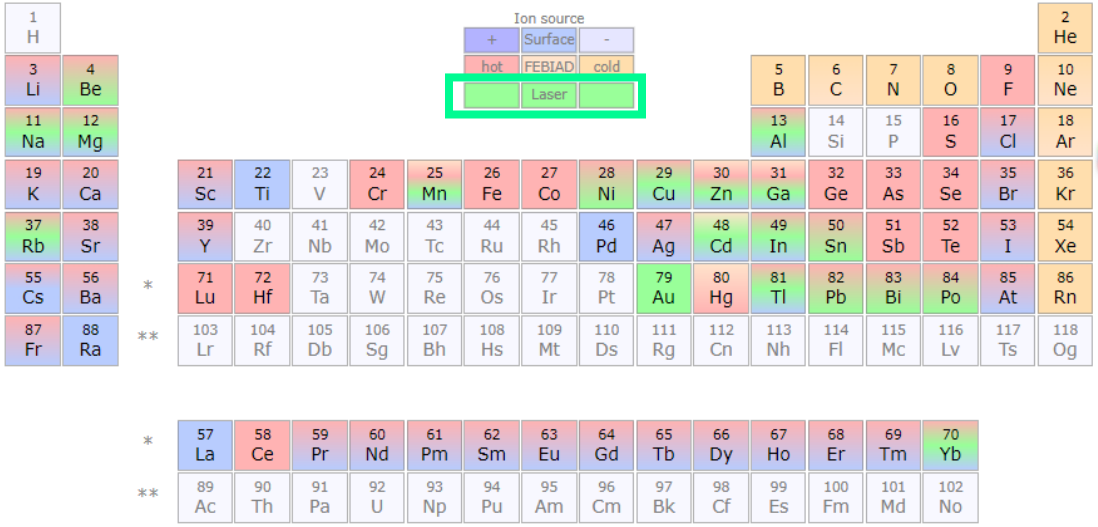
Poster 64,66. F. Maldonado: The FEBIAD at TRIUMF

ISOL Step 3: Ionization



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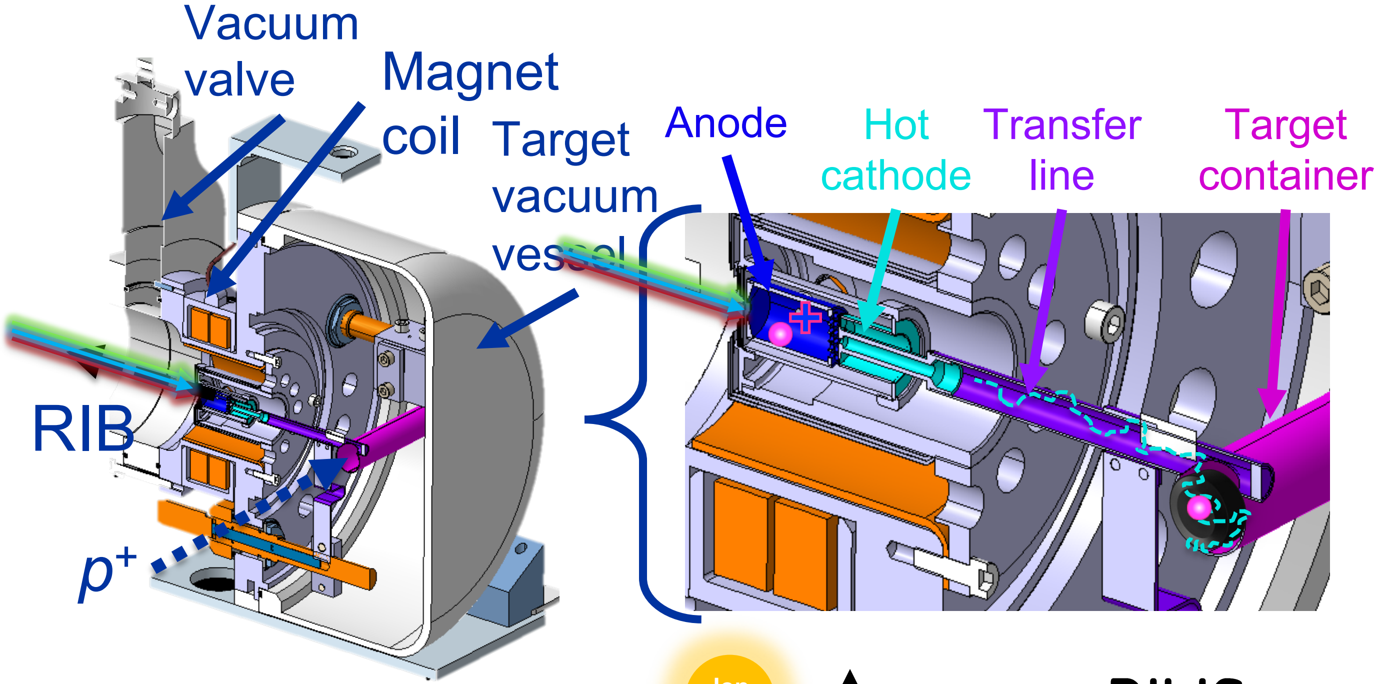

Ion sources

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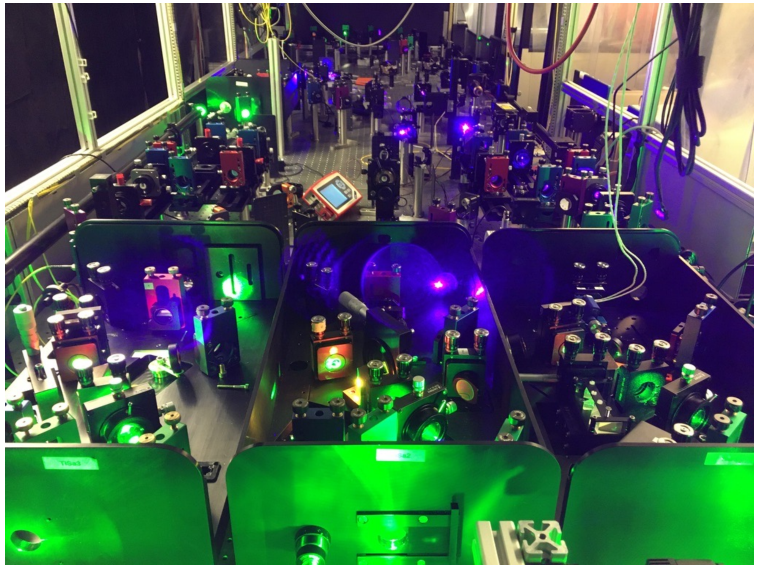
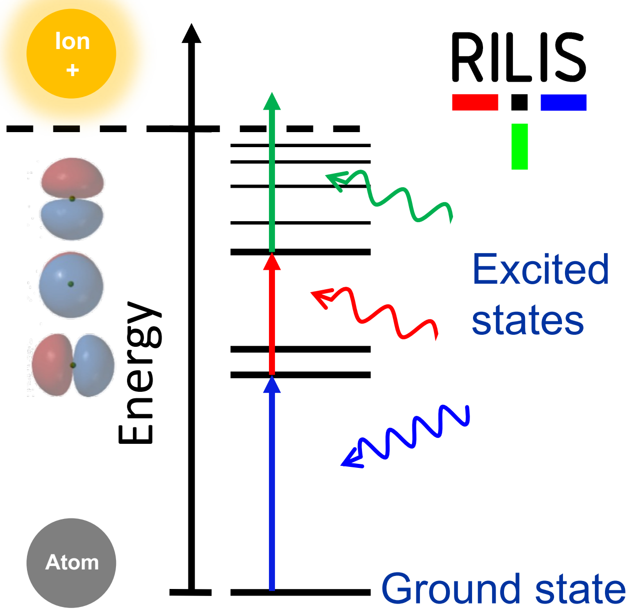
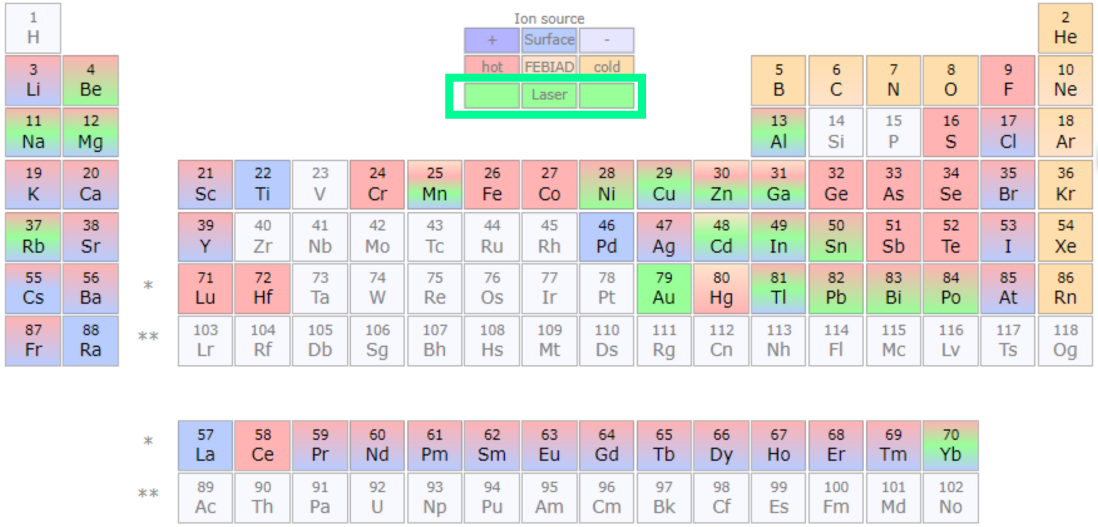




ISOL Step 3: Ionization



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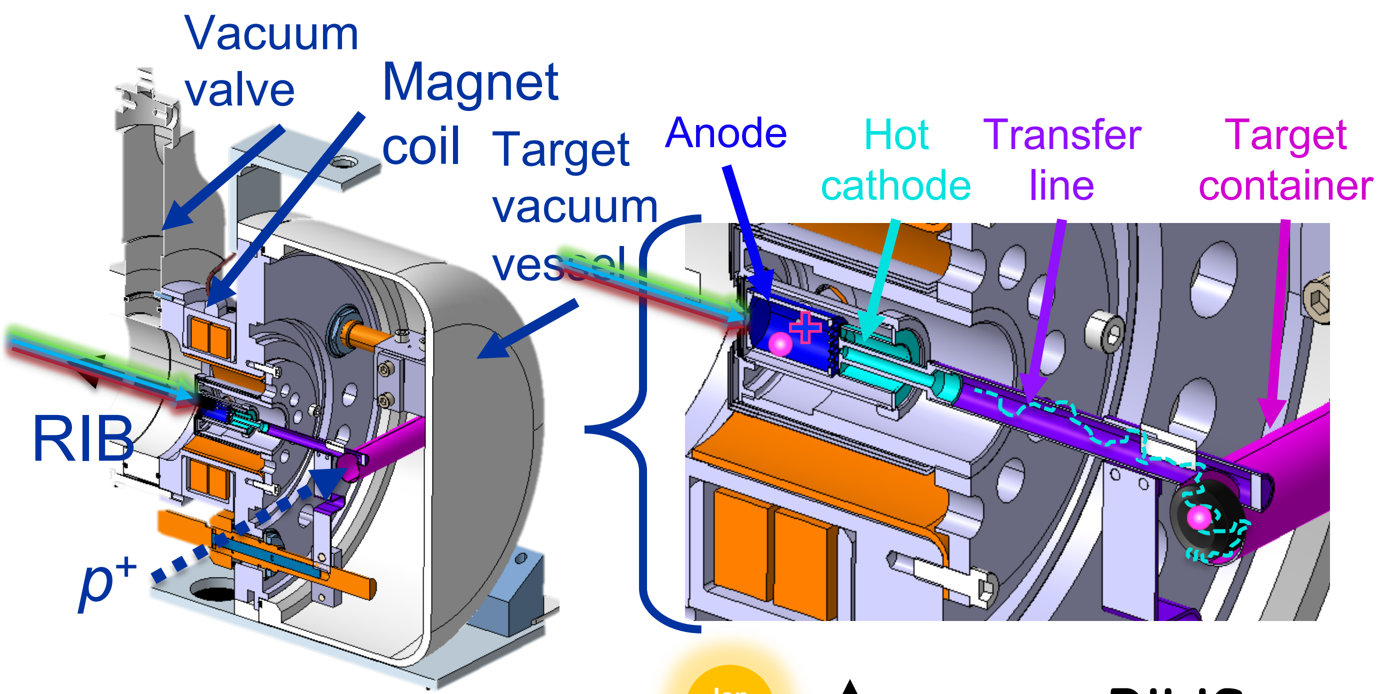
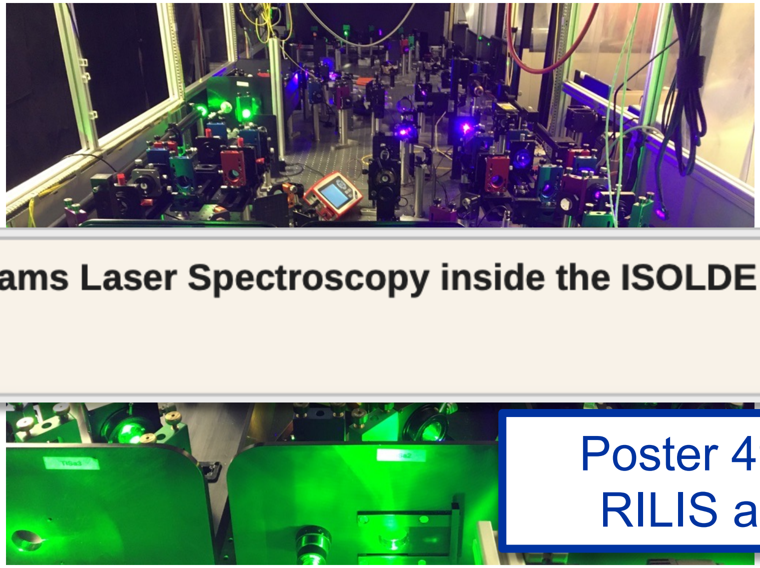
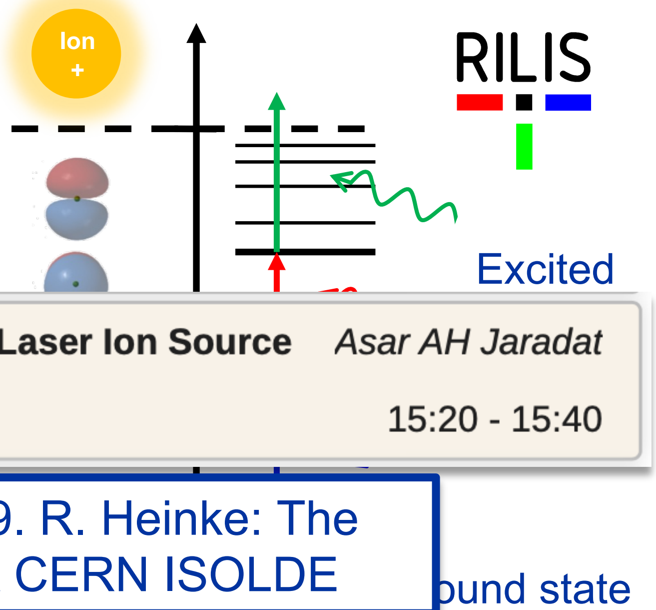


Ion sources

- Surface ionization
- Electro
- Resonance laser ionization



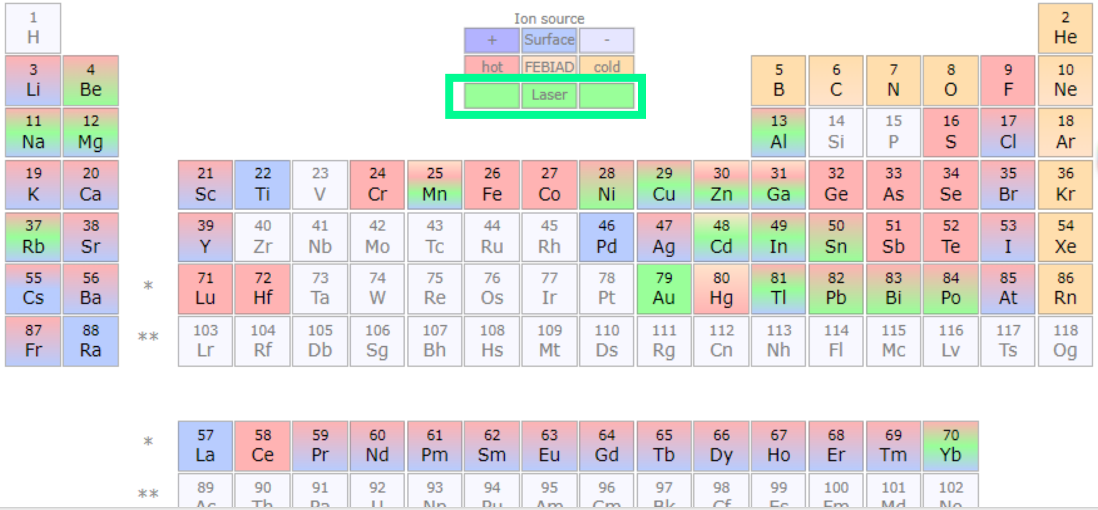


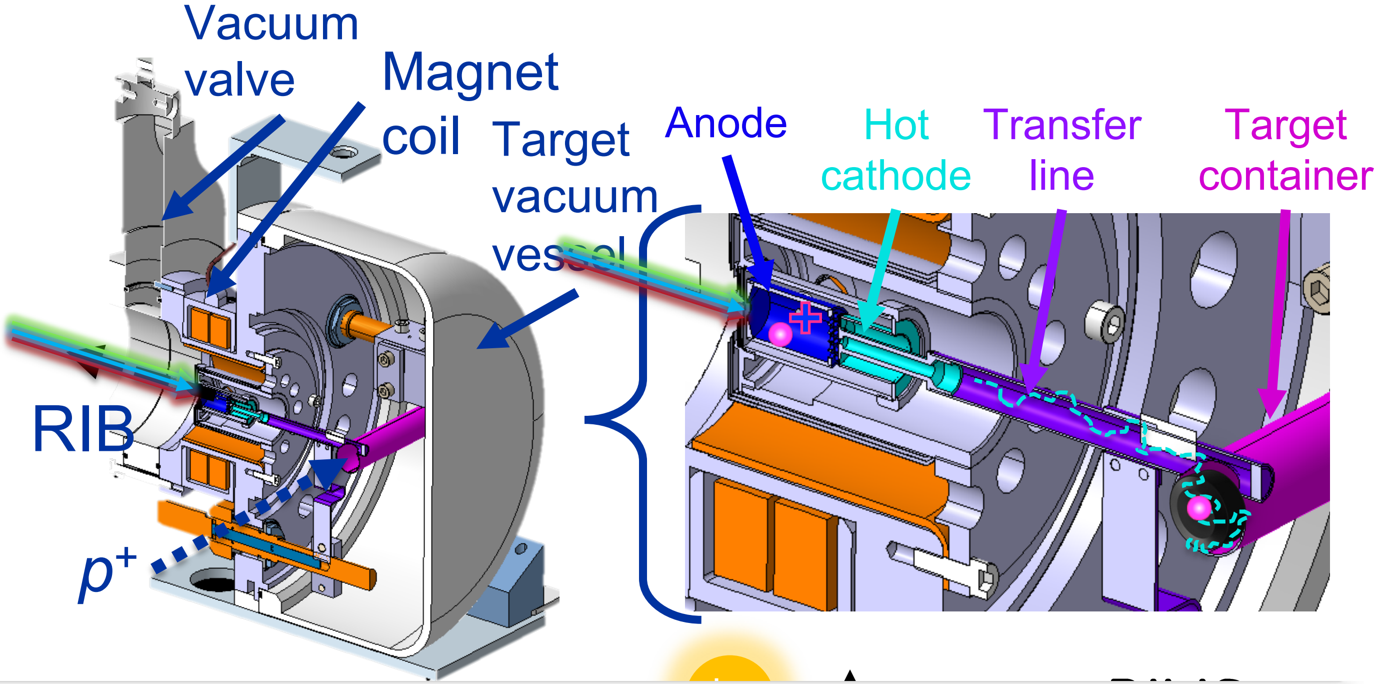
The PI-LIST: High-Resolution Crossed-Beams Laser Spectroscopy inside the ISOLDE Laser Ion Source *Asar AH Jaradat*

Victoria Conference Centre 15:20 - 15:40

Poster 49. R. Heinke: The RILIS at CERN ISOLDE

ISOL Step 3: Ionization

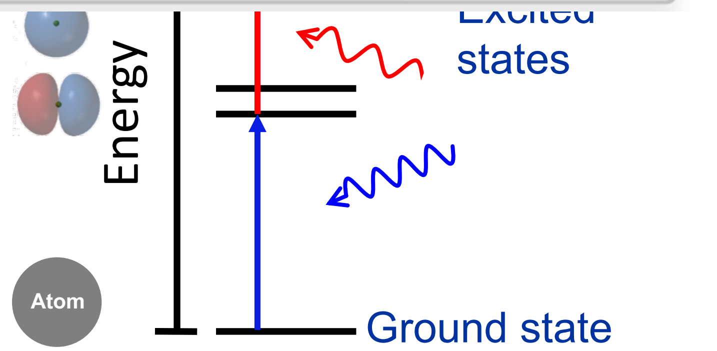




Ion Sources for Radioactive Ion Beam Delivery at CERN-ISOLDE *Sebastian Rothe*

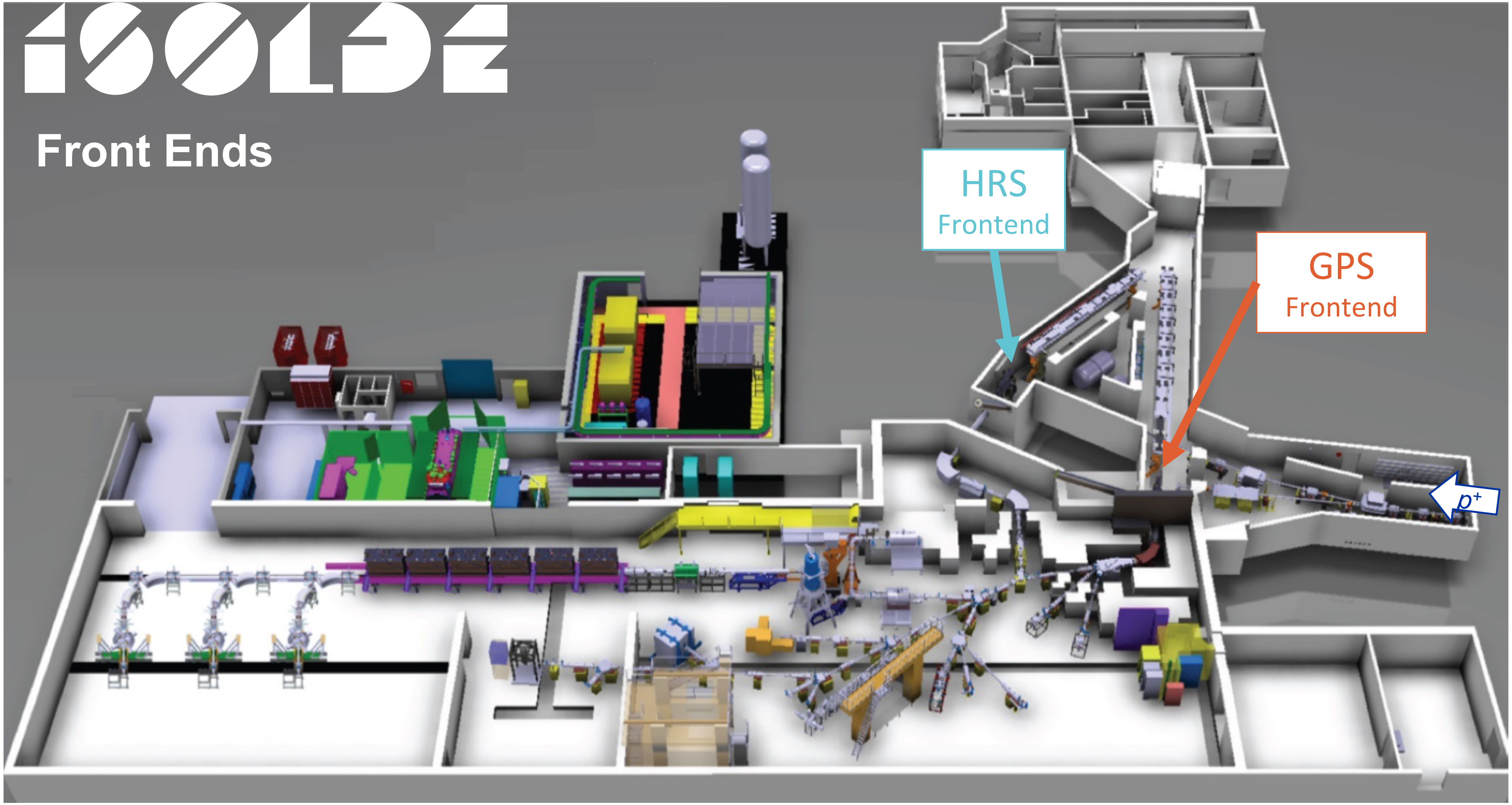
Victoria Conference Centre 11:30 - 12:00

- Surface ionization
- Electron impact ionization
- Resonance laser ionization



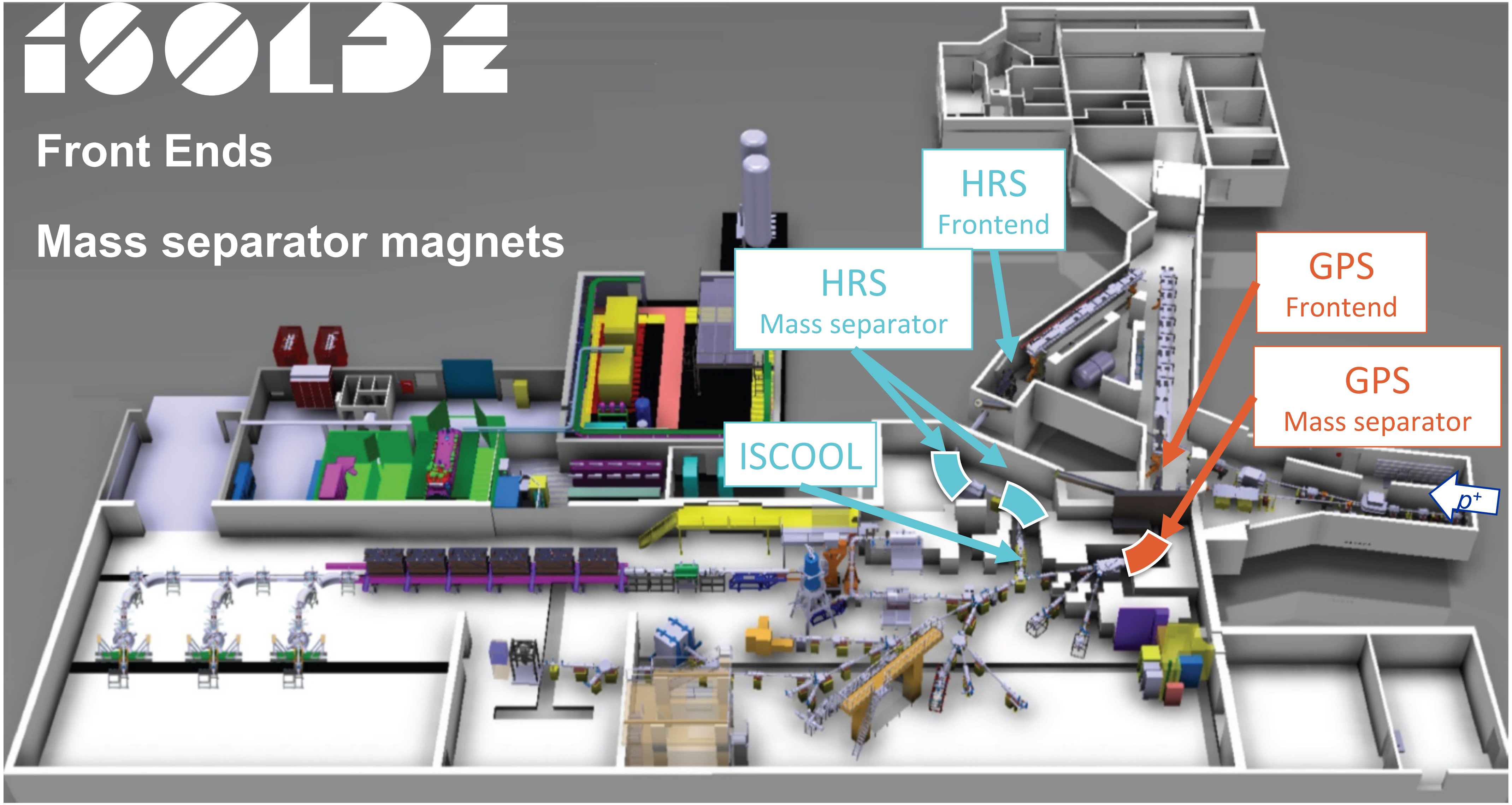
ISOL Step 4: Mass separation

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



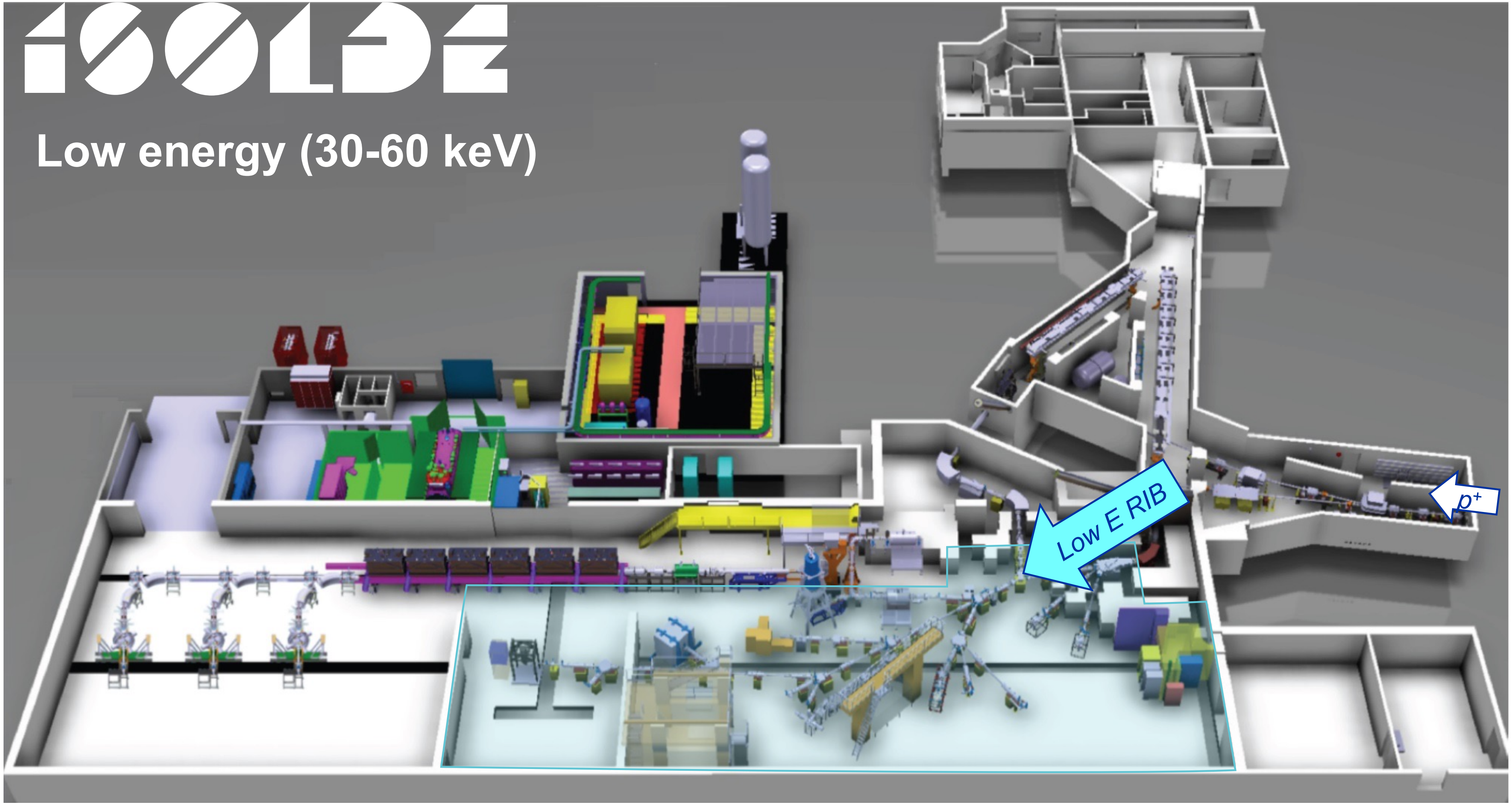
ISOL Step 4: Mass separation

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



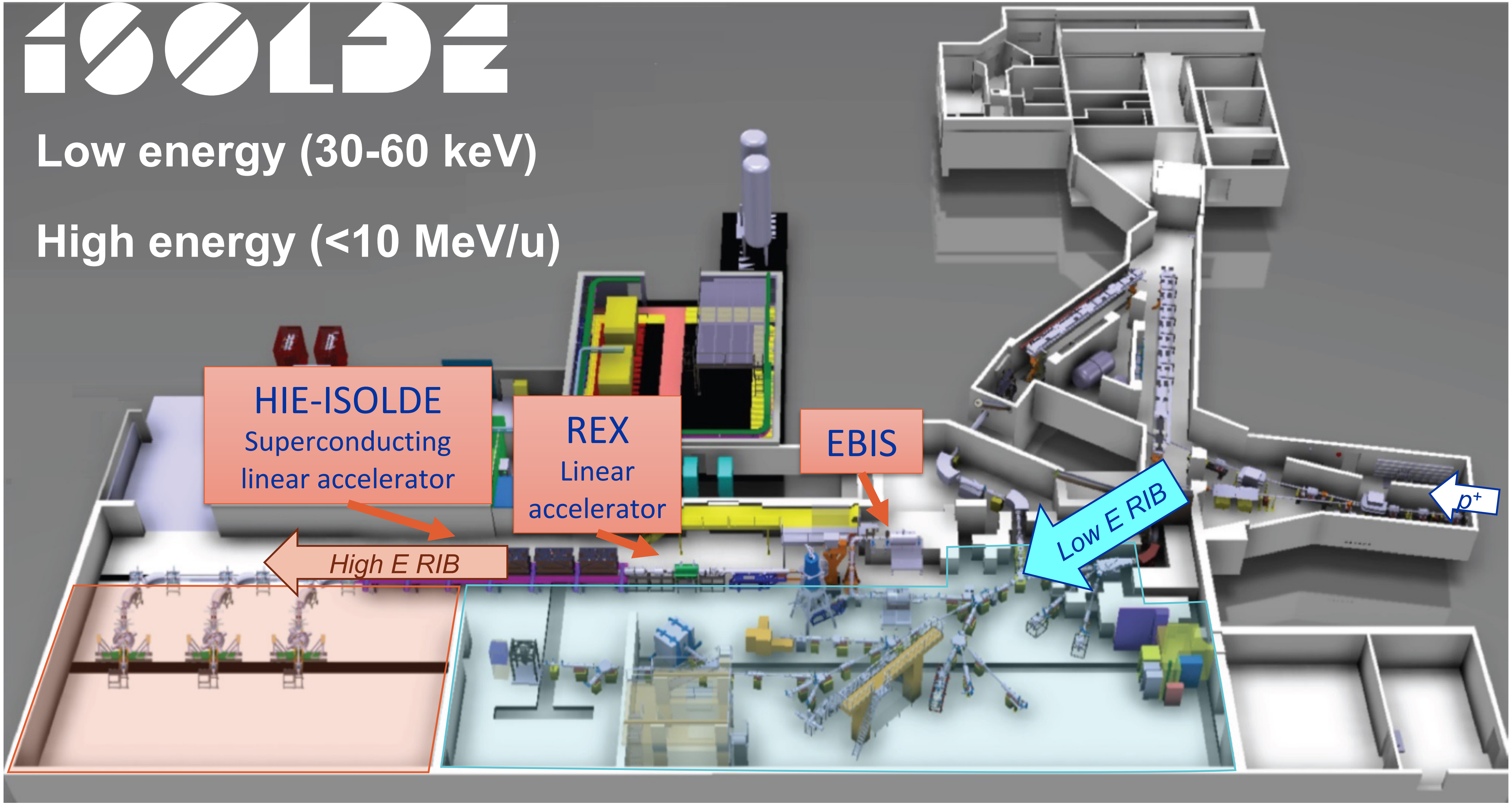
ISOL Step 5: Delivery to Experiments

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



ISOL Step 5: Delivery to Experiments

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

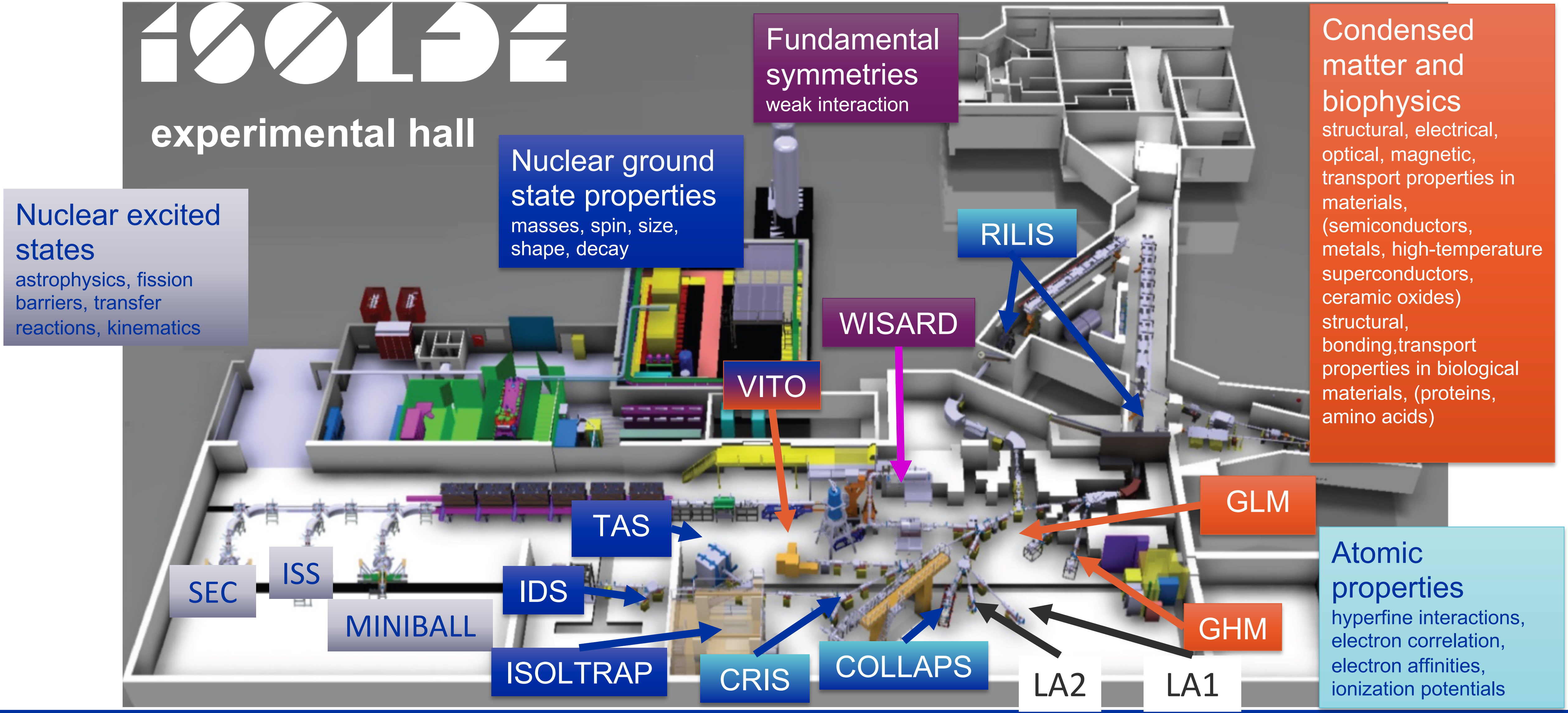
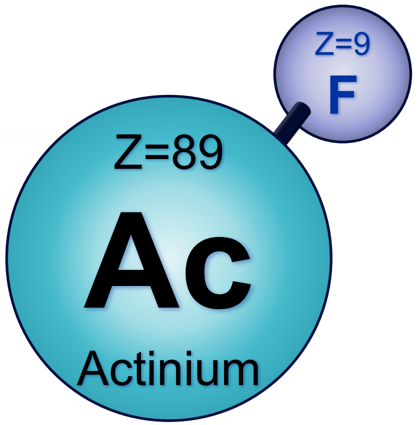


Image published in EP Newsletter, CERN (2020)



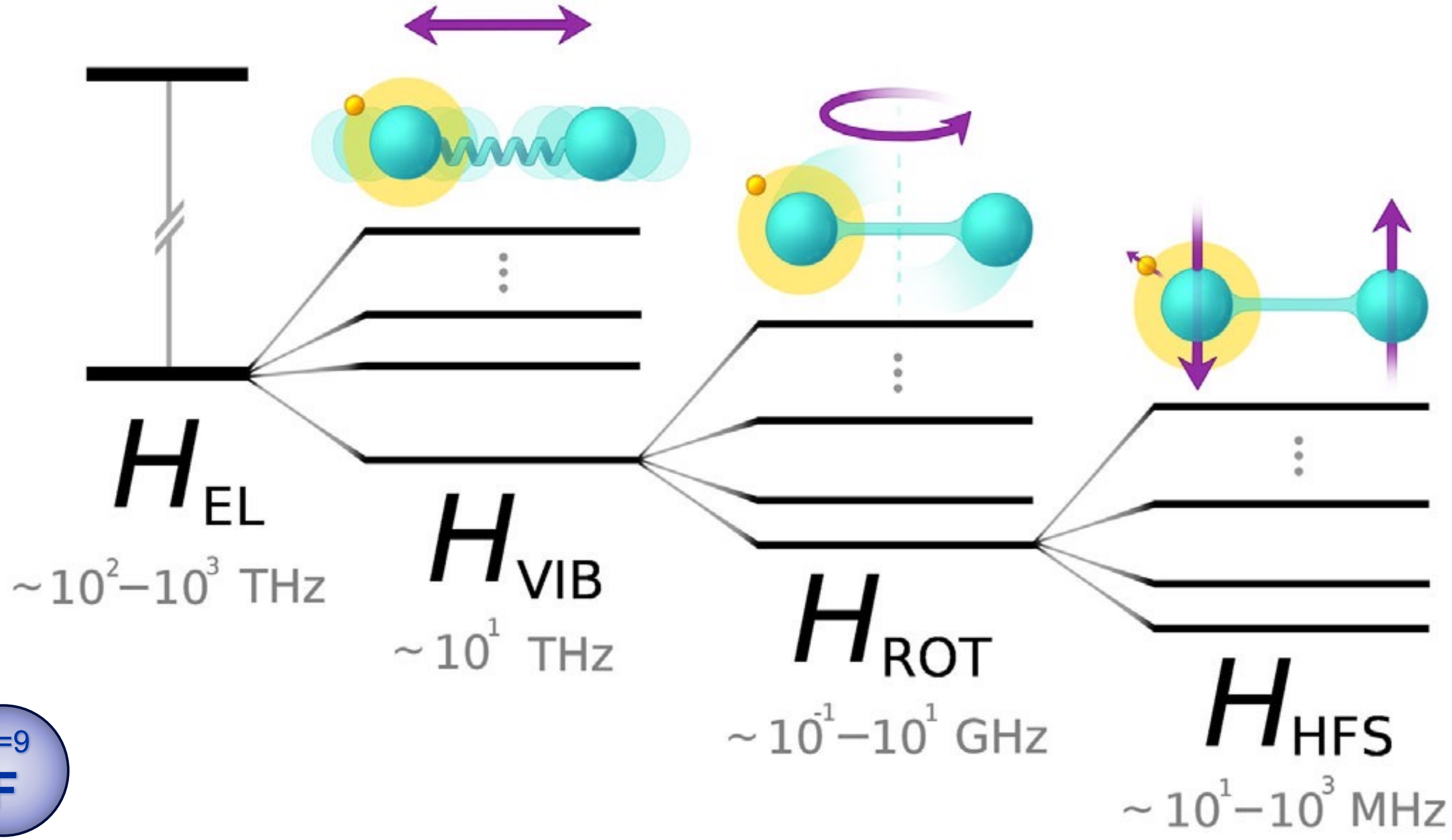
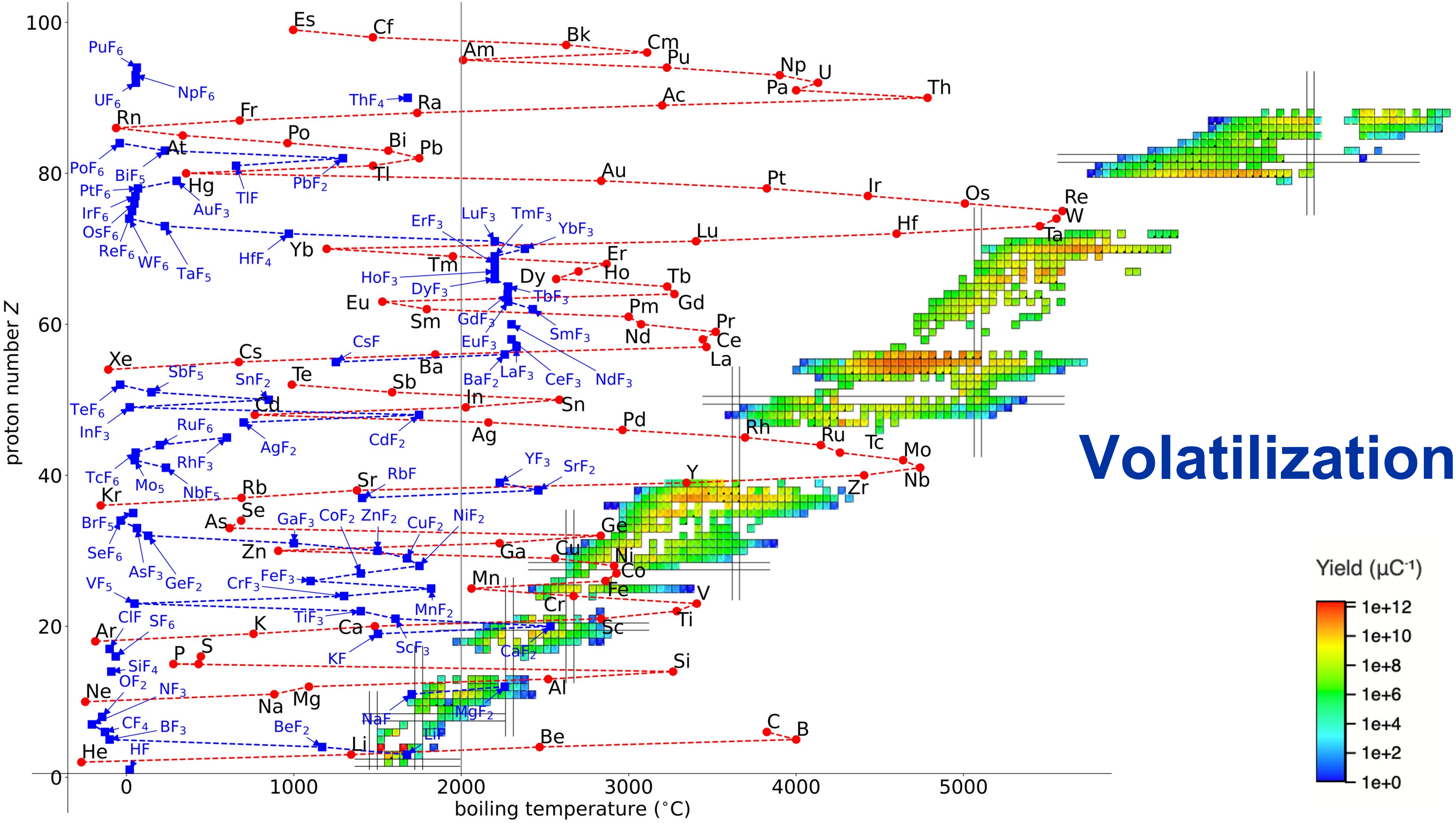


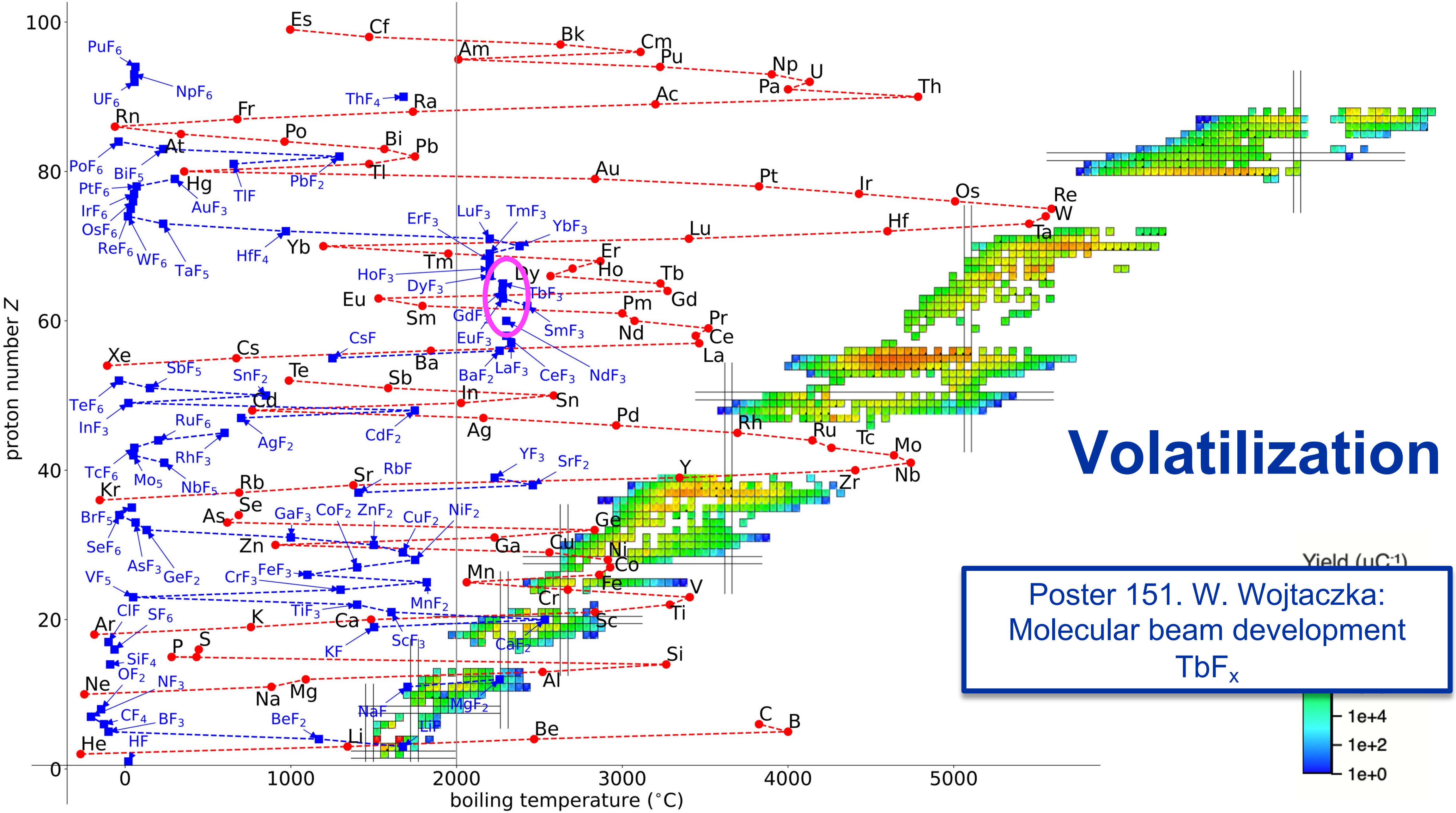
Image published in Athanasakis-Kaklamanakis *et al.*, PRX **13** 011015 (2023)

Radioactive molecular beams

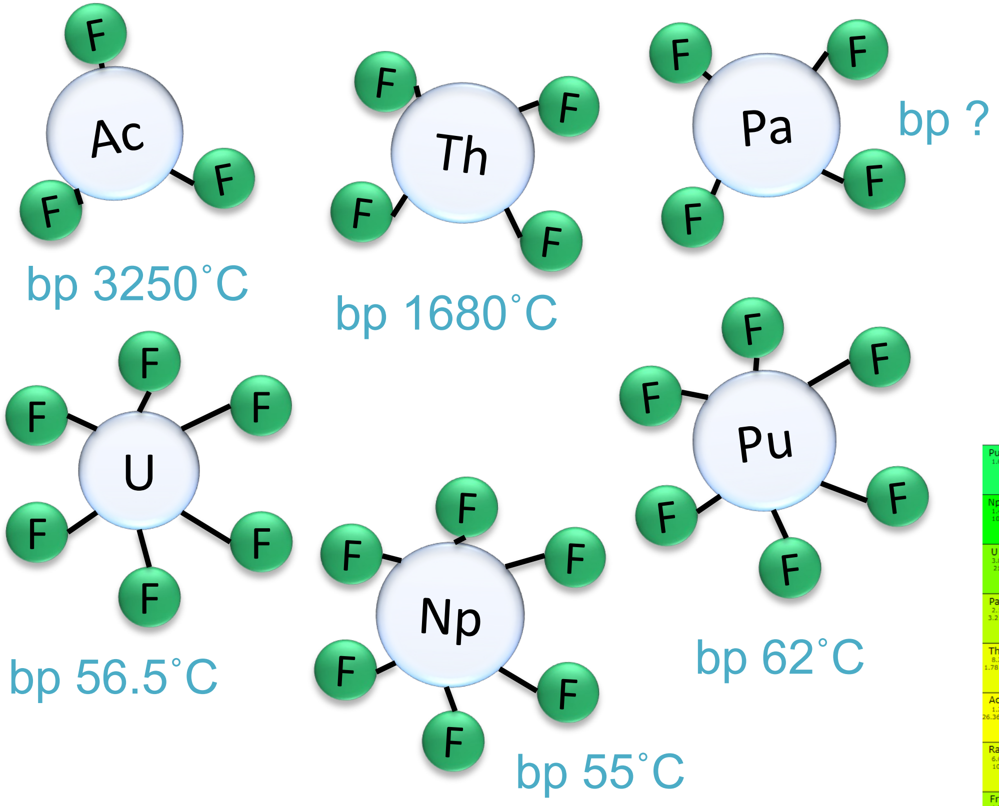
Volatilization

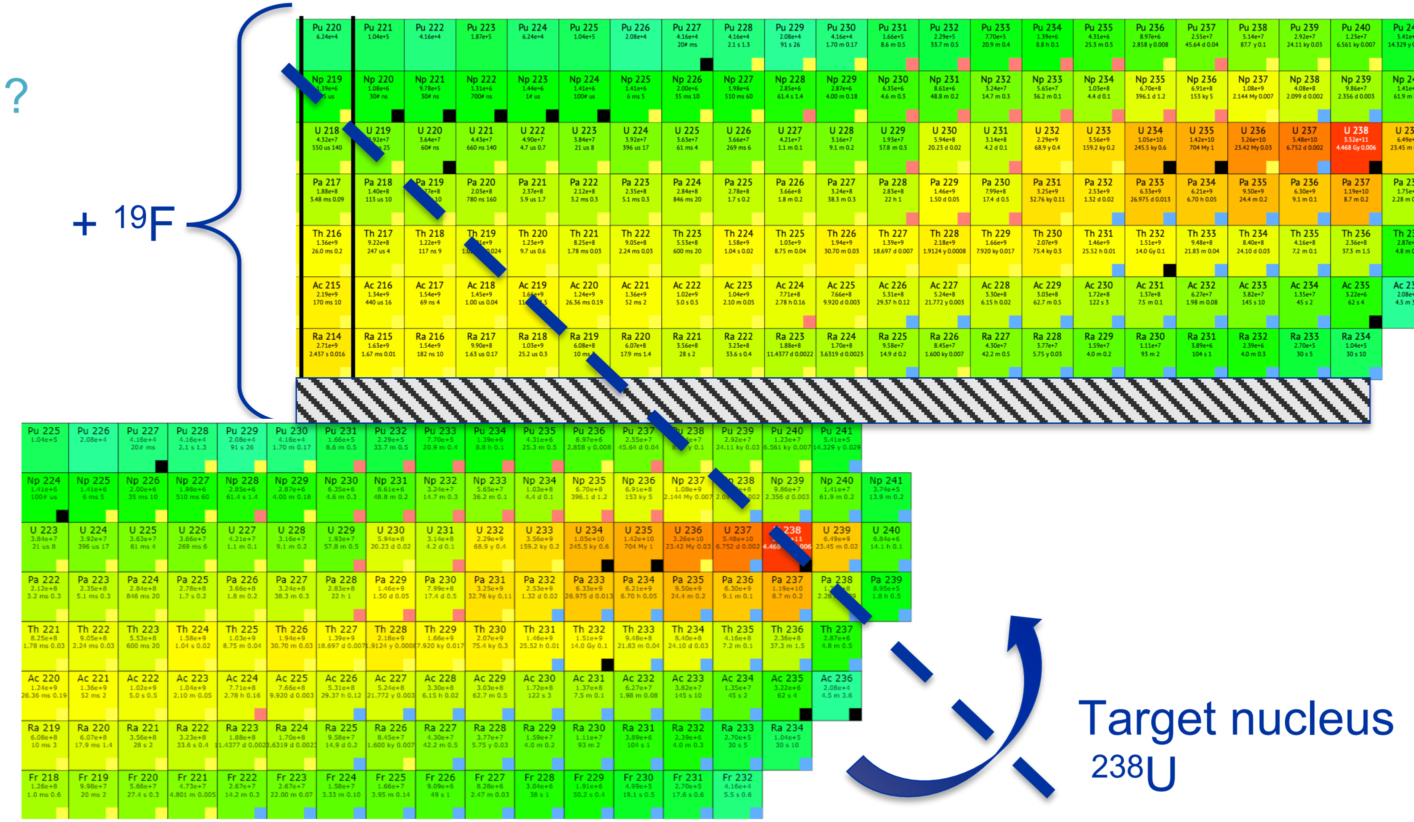


Volatilization



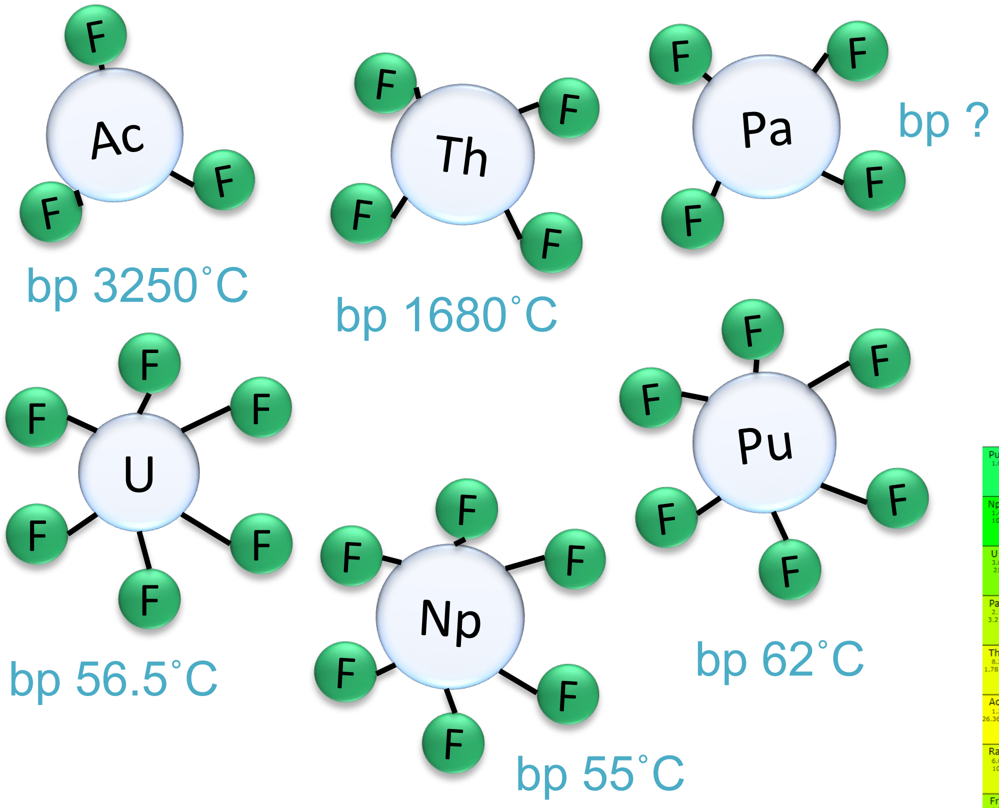
Poster 151. W. Wojtaczka: Molecular beam development TbF_x

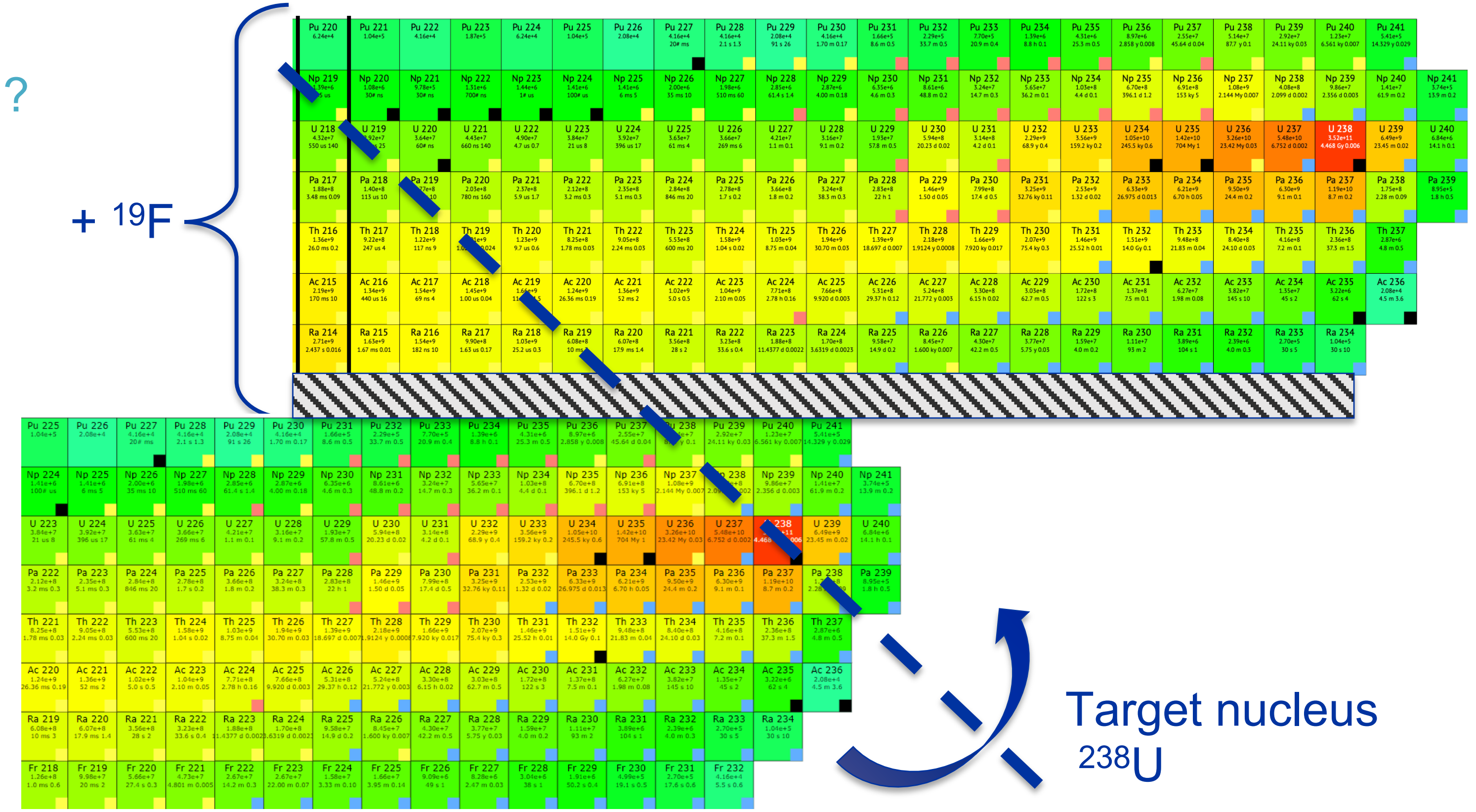




Molecular beams

1. Volatilization
2. Sideband extraction
3. Research opportunities





Molecular beams

1. Volatilization

2. Sideband extraction

3. Research opportunities

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

A small detour: Why is there more matter than antimatter?

Matter-antimatter asymmetry

- Initial conditions of expanding universe

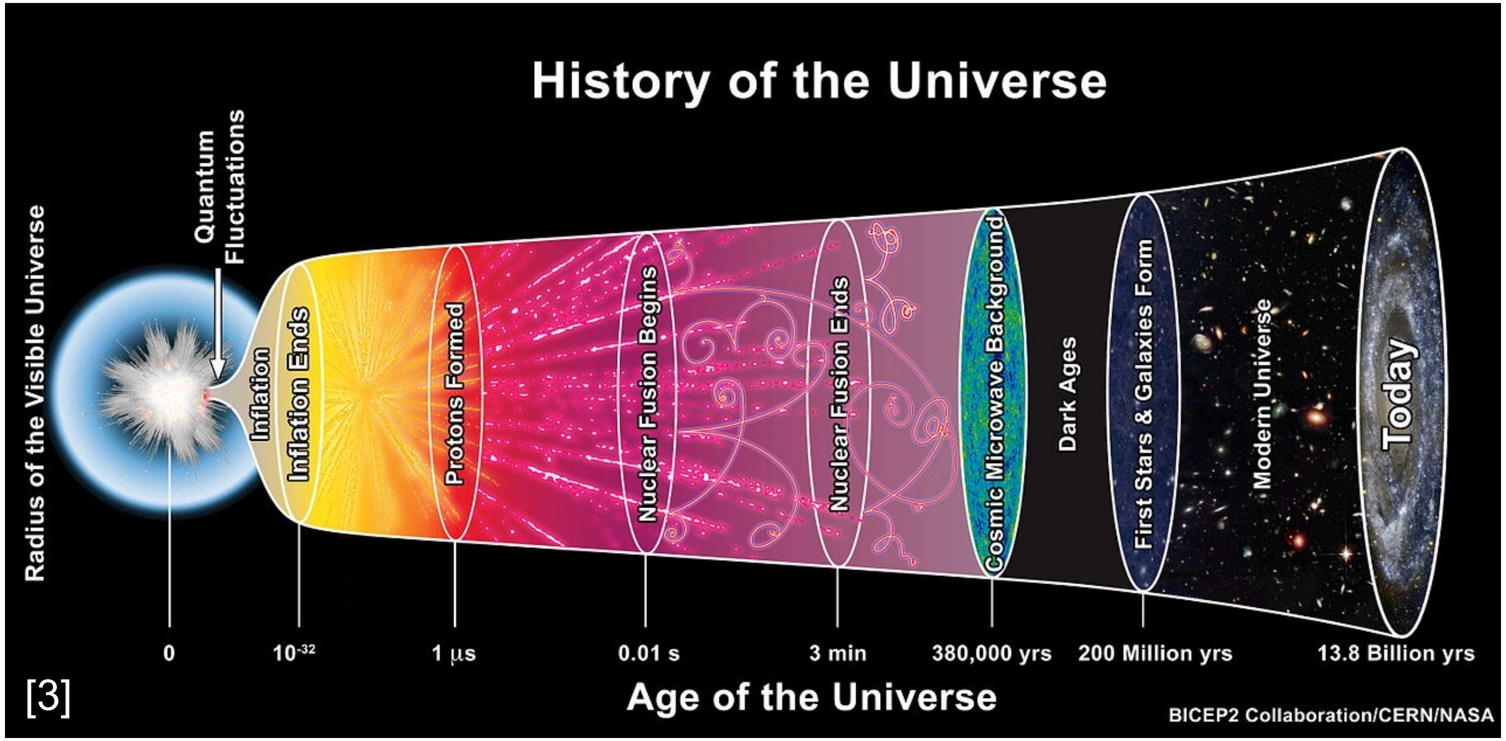
Sakharov's conditions

- *CP* violation for baryogenesis [1]

CPV in the Standard Model

- CKM phase δ_{CKM} (order 1)
- Strong *CP*-phase $\bar{\theta}$ (tiny)
- BSM?

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

EDM searches

The existence of a finite permanent EDM of a particle or atom would violate time reversal (T) and parity (P) symmetry, or equivalently charge conjugation and parity symmetry (CP), needed to solve baryon asymmetry

July 2023
Roussy et al., Science 381, 6653, pp. 46-59 (2023)
Scale: ~10^{13} eV

nEDM $|d_n|$

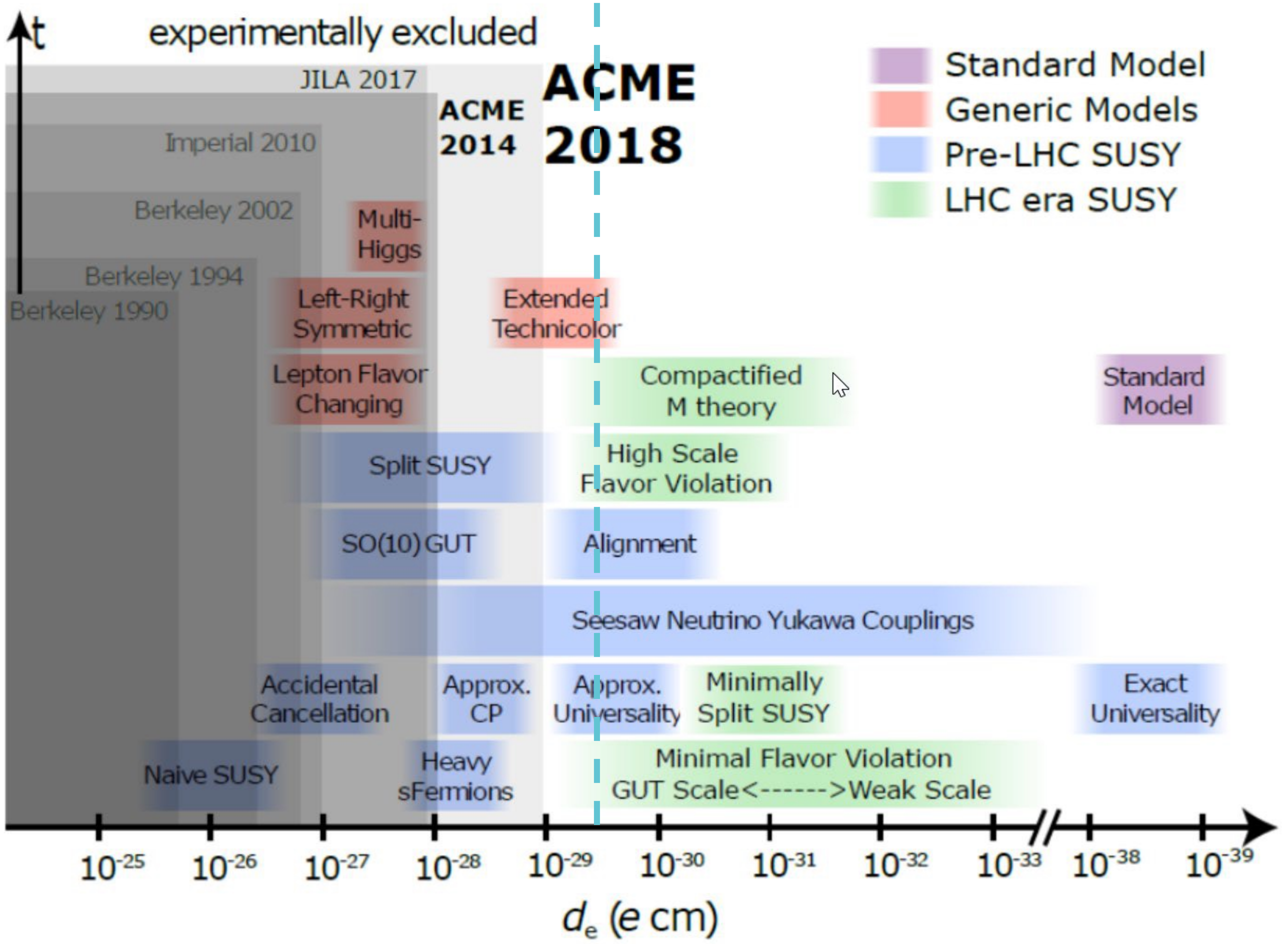
- 2006 ILL UCNs: $|d_n| < 2.9\times10^{-26}$ *e* cm

eEDM $|d_e|$

- 2011 Imperial $^{174}Yb^{19}F$: $|d_e| < 2\times10^{-28}$ *e* cm
- 2018, 2013 ACME $^{232}Th^{16}O$: $|d_e| < 1\times10^{-29}$ *e* cm
- 2023, 2017 JILA $^{180}Hf^{19}F^{+}$: $|d_e| < 4.1\times10^{-30}$ *e* cm

Atomic EDM

- 2009, ^{199}Hg: $|d| < 3.1\times10^{-29}$ *e* cm
- 2015, ^{225}Ra: $|d| < 5\times10^{-22}$ *e* cm

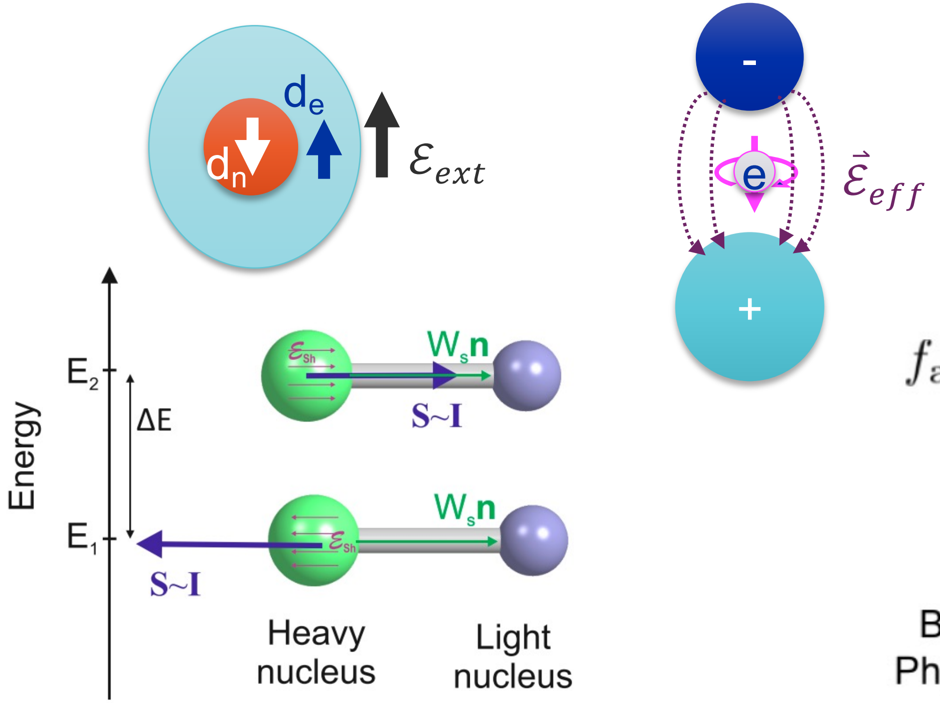


D. DeMille, The ACME Experiment, (2023)
https://cfp.physics.northwestern.edu/gabrielse-group/acme-electron-edm.html

Sources of BSM physics in radioactive molecules

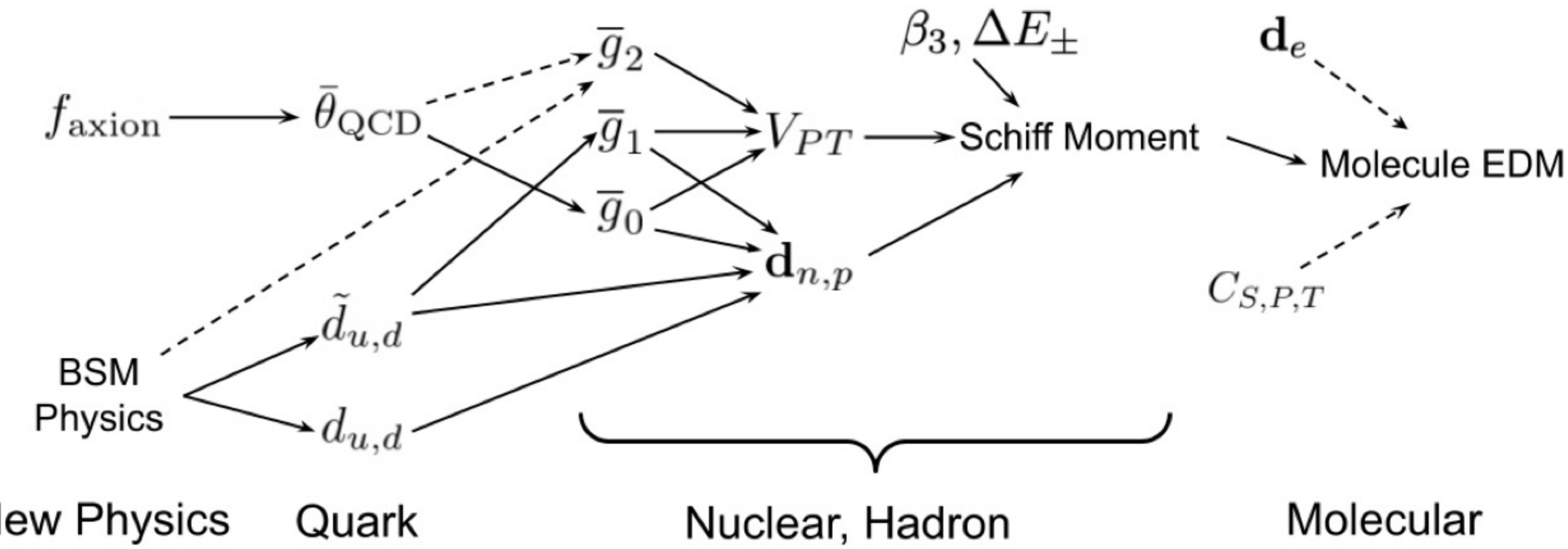
Schiff screening

- non-relativistic, point-like constituents in a bound, neutral system interacting only electrostatically



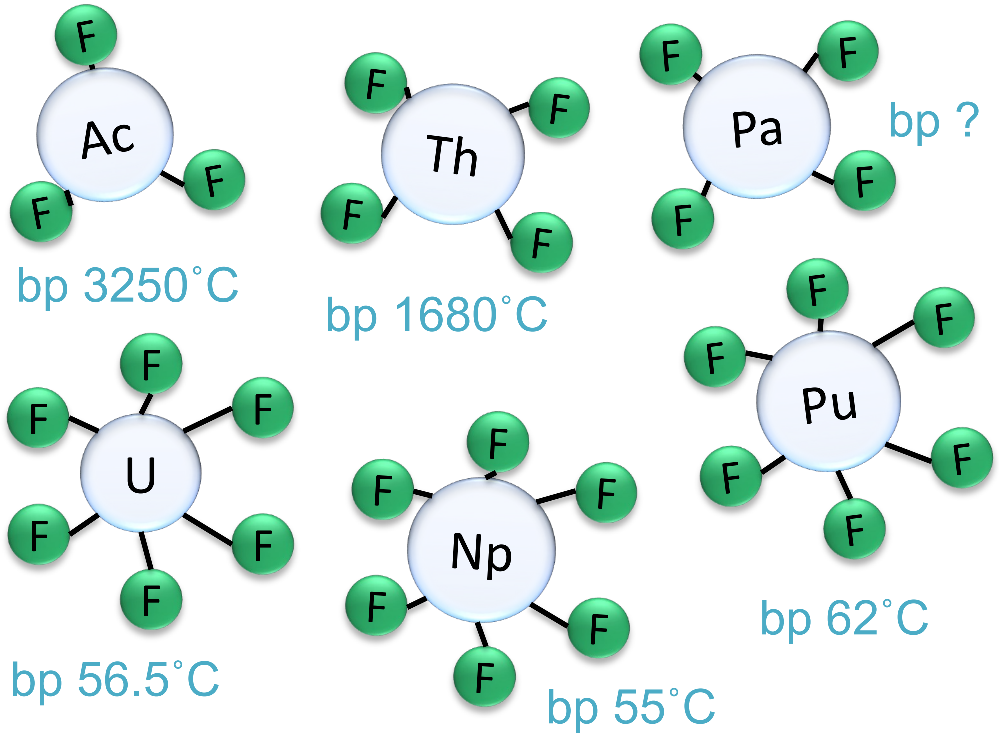
CPV

- Imperfect screening
- Relativistic effects, $\vec{\mathcal{E}}_{eff}$
- Finite-size nucleus: $S \propto \frac{\beta_2 \beta_3^2 Z A^{2/3}}{\Delta E_\pm}$, MQM $(I \geq 1)$



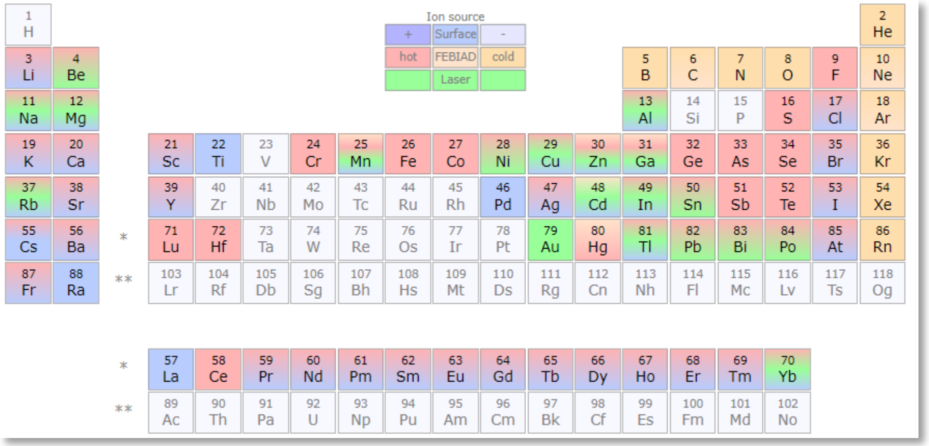
Opportunities for Fundamental Physics Research with Radioactive Molecules, arXiV 2302.02165 (2023)

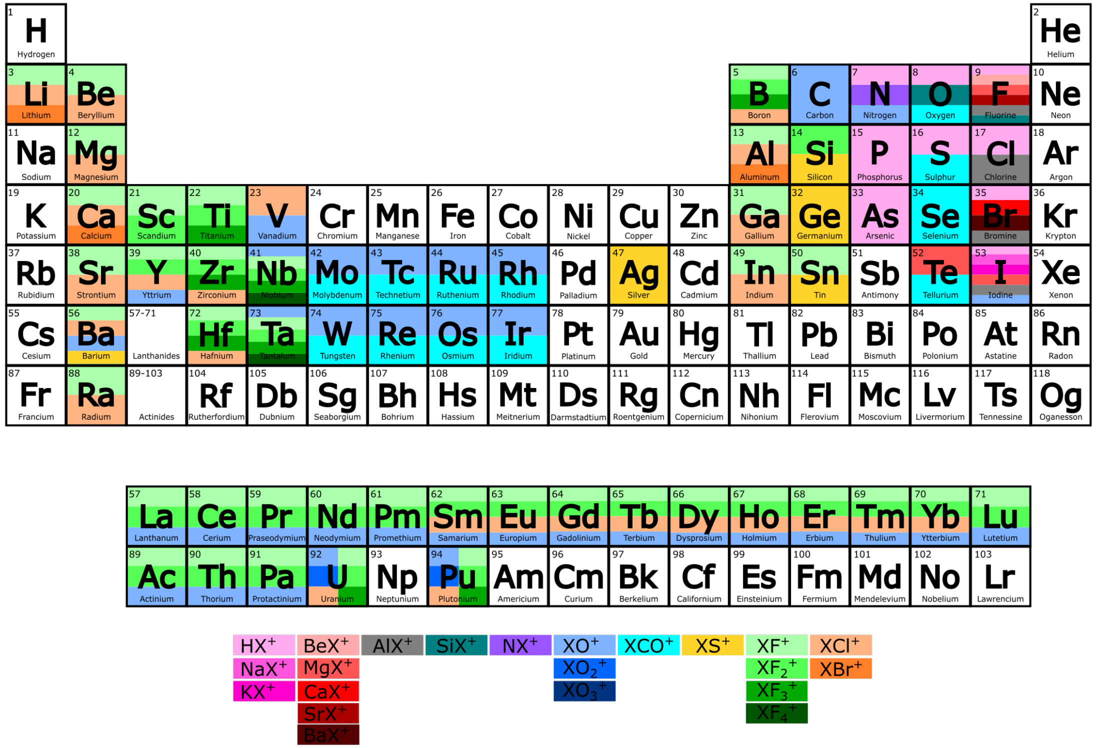
[1] Alarcon *et al*., (2022) *arXiV* 2203.08103
[2] Safronova et al. (2018) Rev. Mod. Phys. 90. 2,
[3] Skripnikov et al. (2020) Phys. Chem. Chem. Phys. 22. 33, 18374



Molecular beams

1. Formation and ionization
2. Detection and identification
3. Characterization



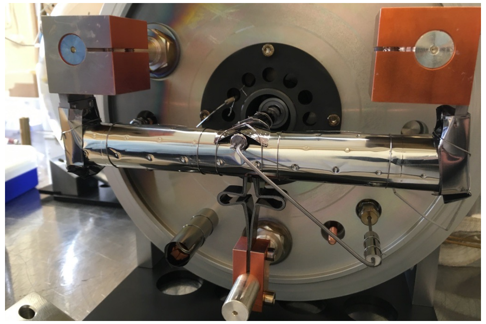


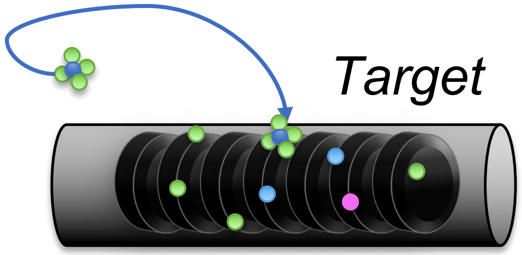
Au and Ballof, (2022) *Zenodo* 10.5281/zenodo.6884293 DOI 10.5281/zenodo.6884293

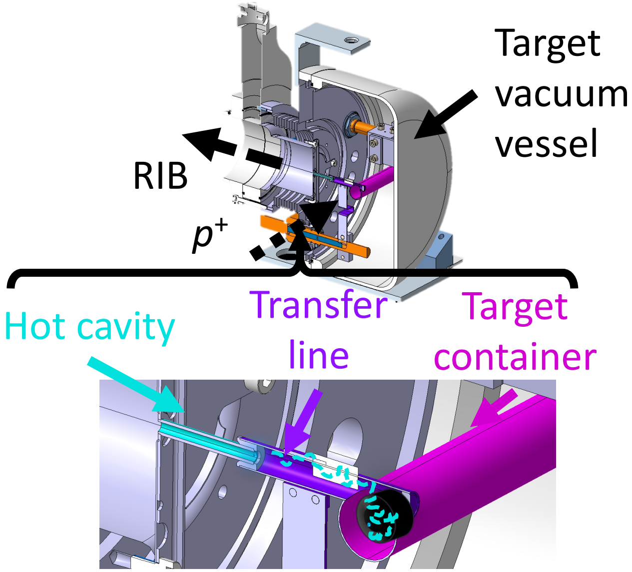
Formation: how do we make the molecules?

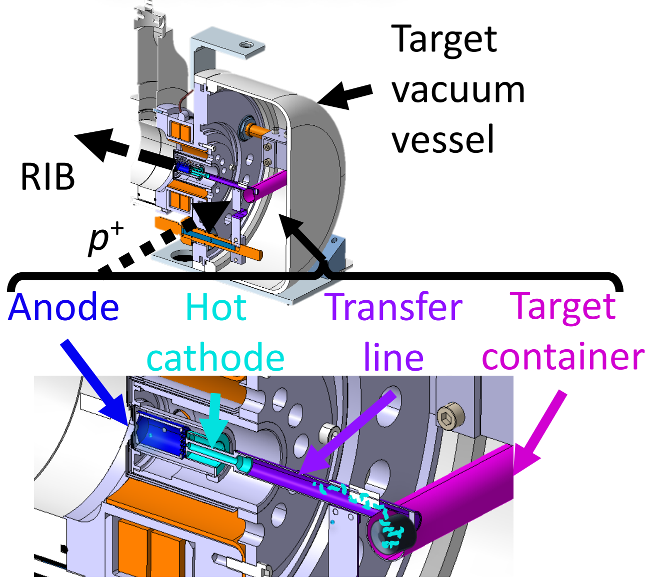
In-source

- Reactive gas



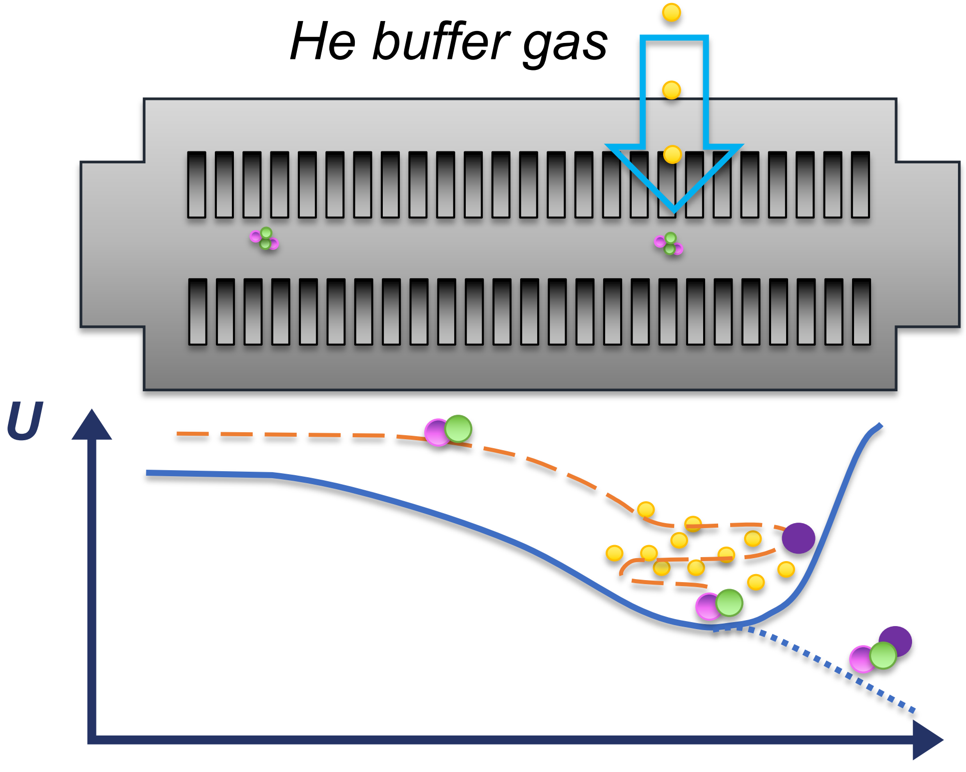






In-trap

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

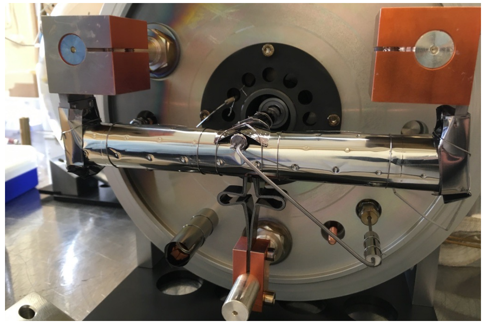


Au et al. (2023) *NIM B.* **541** (375-379)

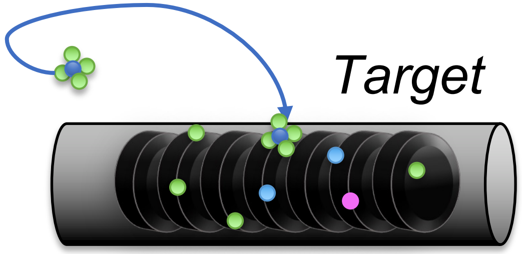
Formation: how do we make the molecules?

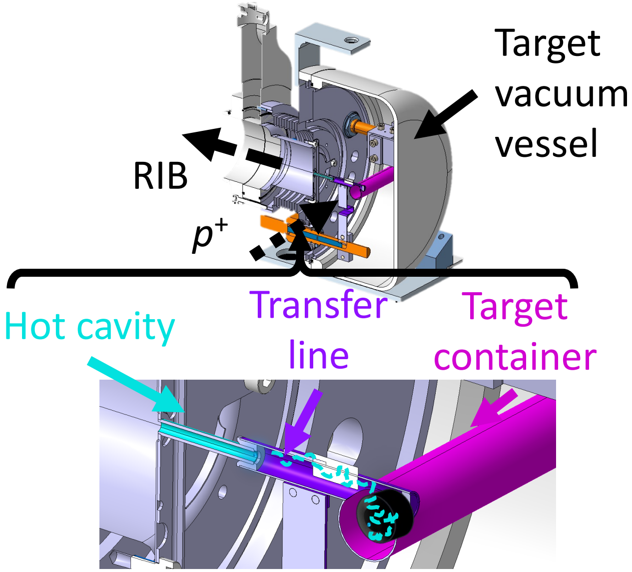
In-source

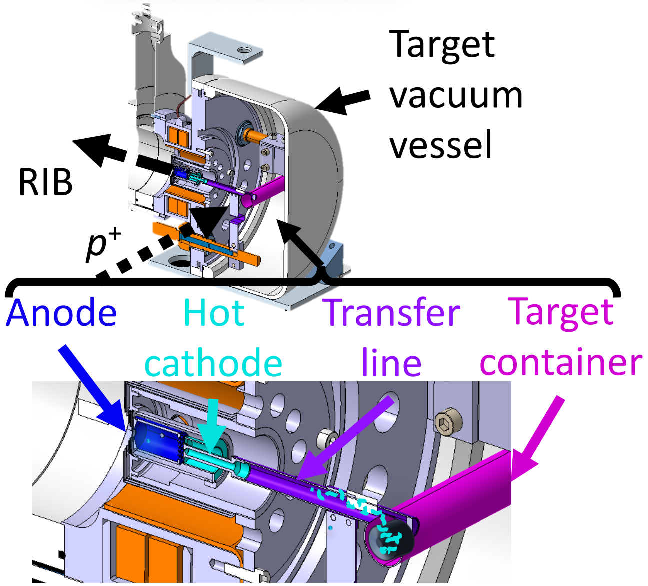
- Reactive gas



CF_4, NF_3, SF_6

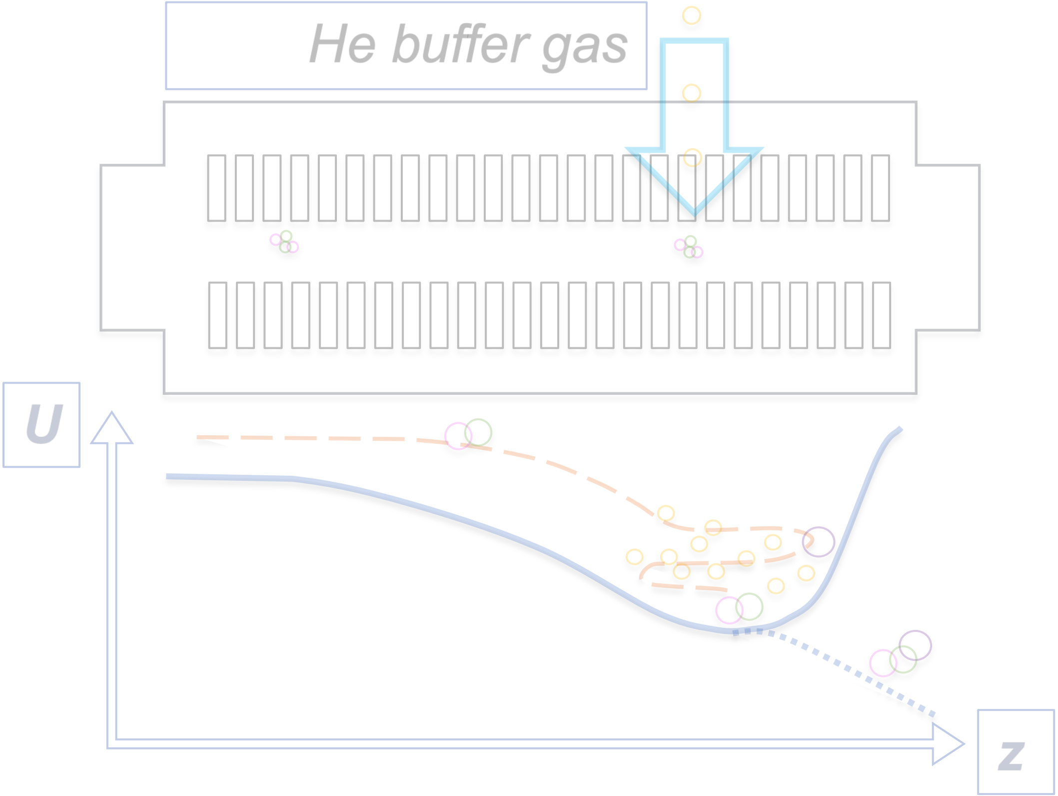






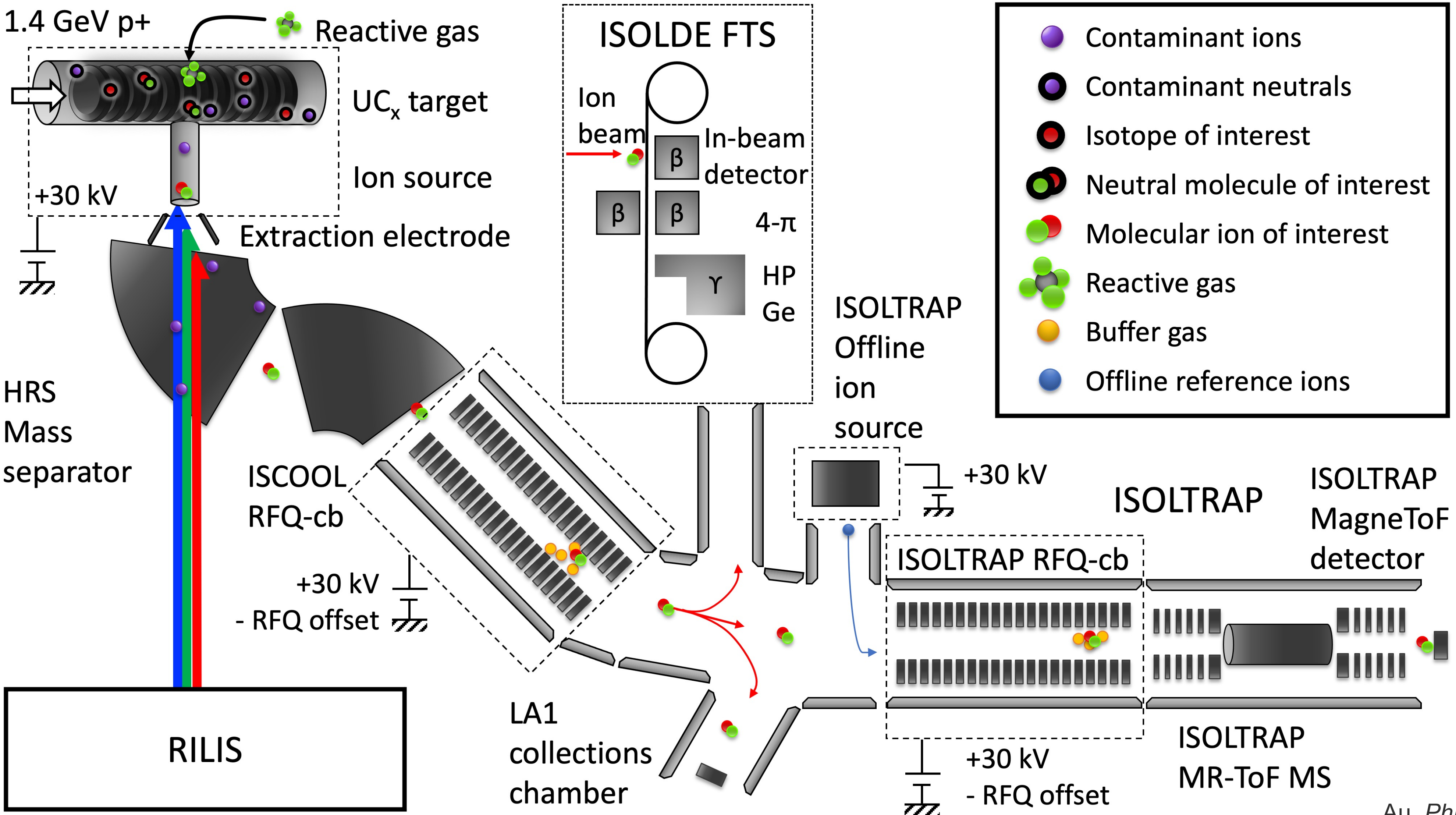
In-trap

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



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

Detection and identification



Au, *PhD thesis* (2023)

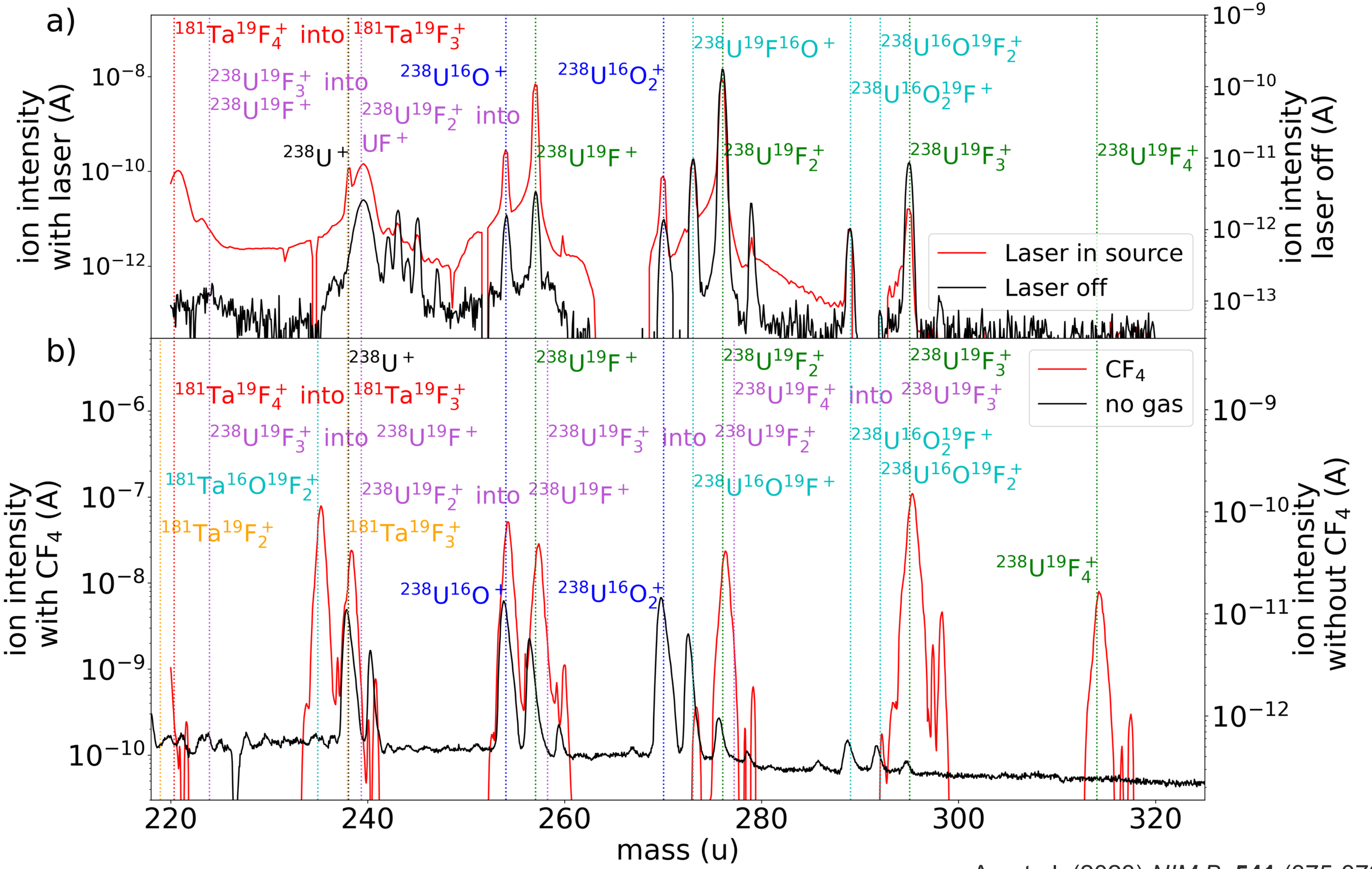
Ion sources and effects

Surface:

- Low IPs
- Surface ionization efficiency

FEBIAD:

- High/unknown IPs
- High efficiency
- Dissociation



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

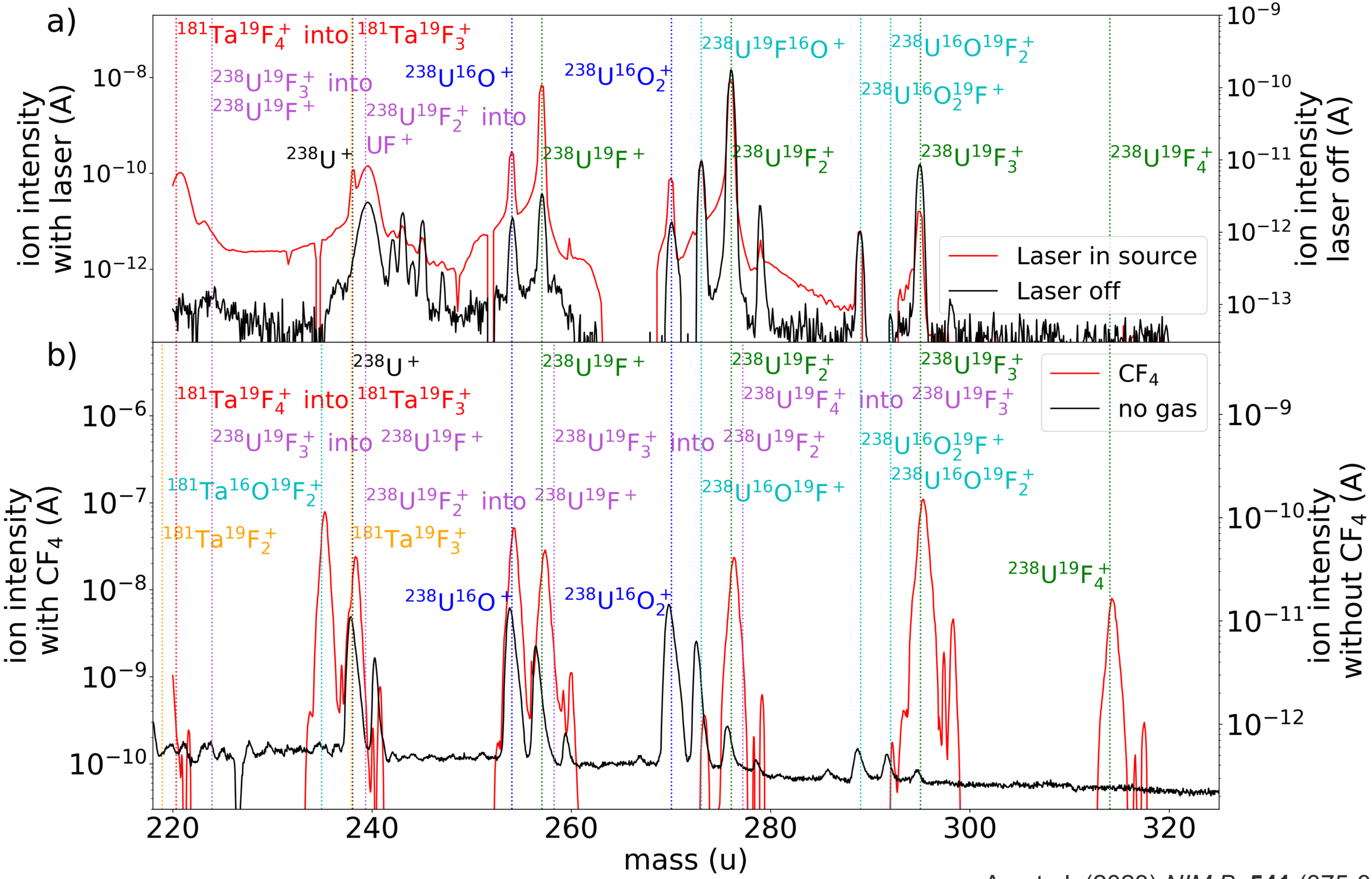
Ion sources and effects

Surface:

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

FEBIAD:

- High/unknown IPs
- High efficiency
- Dissociation



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

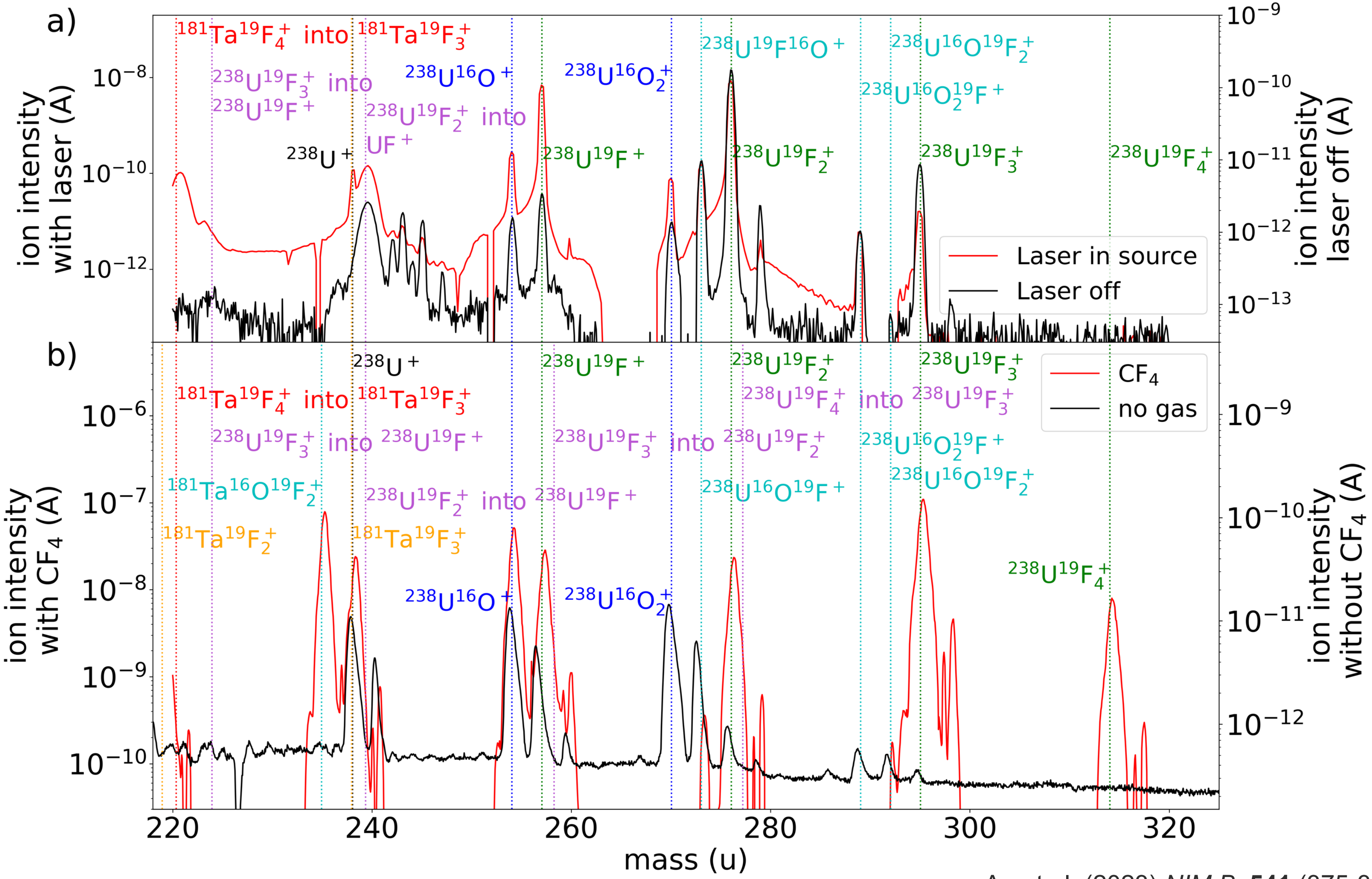
Ion sources and effects

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

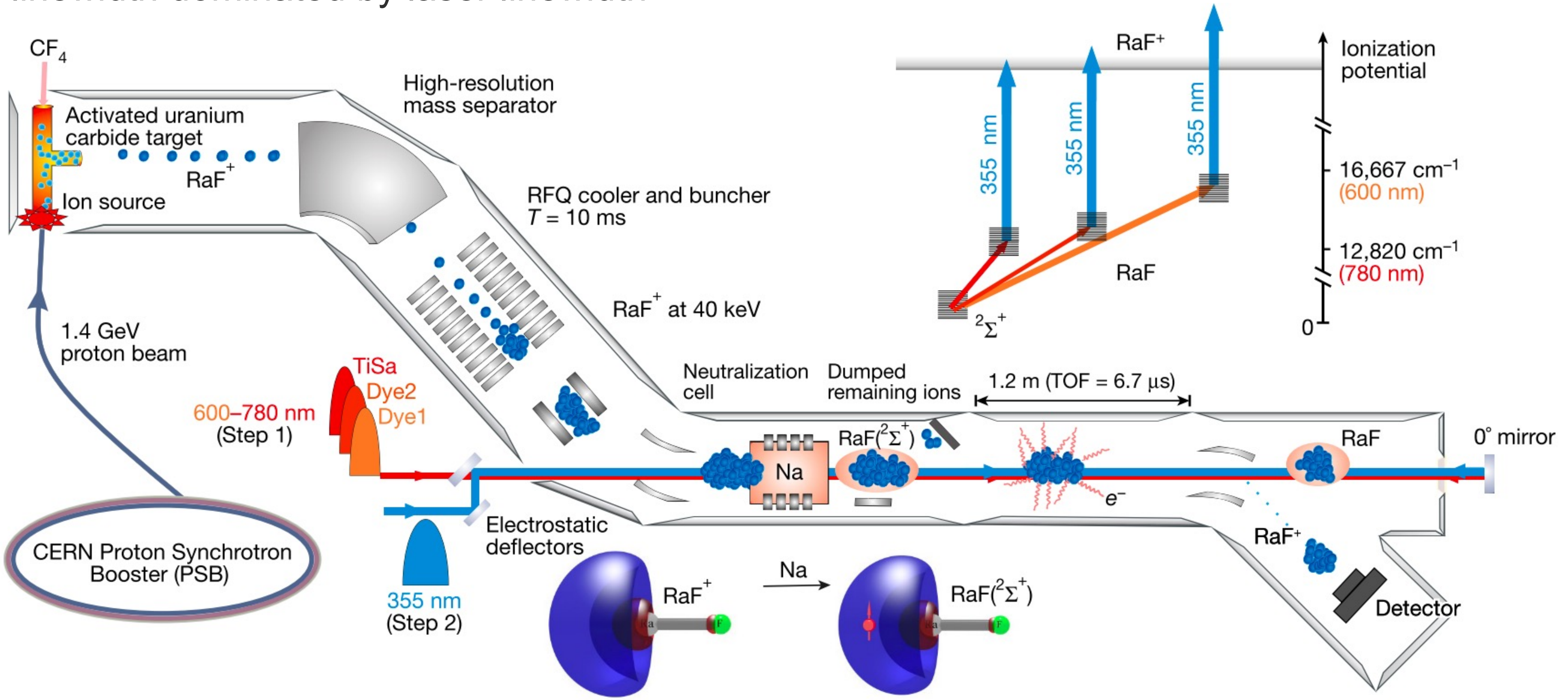
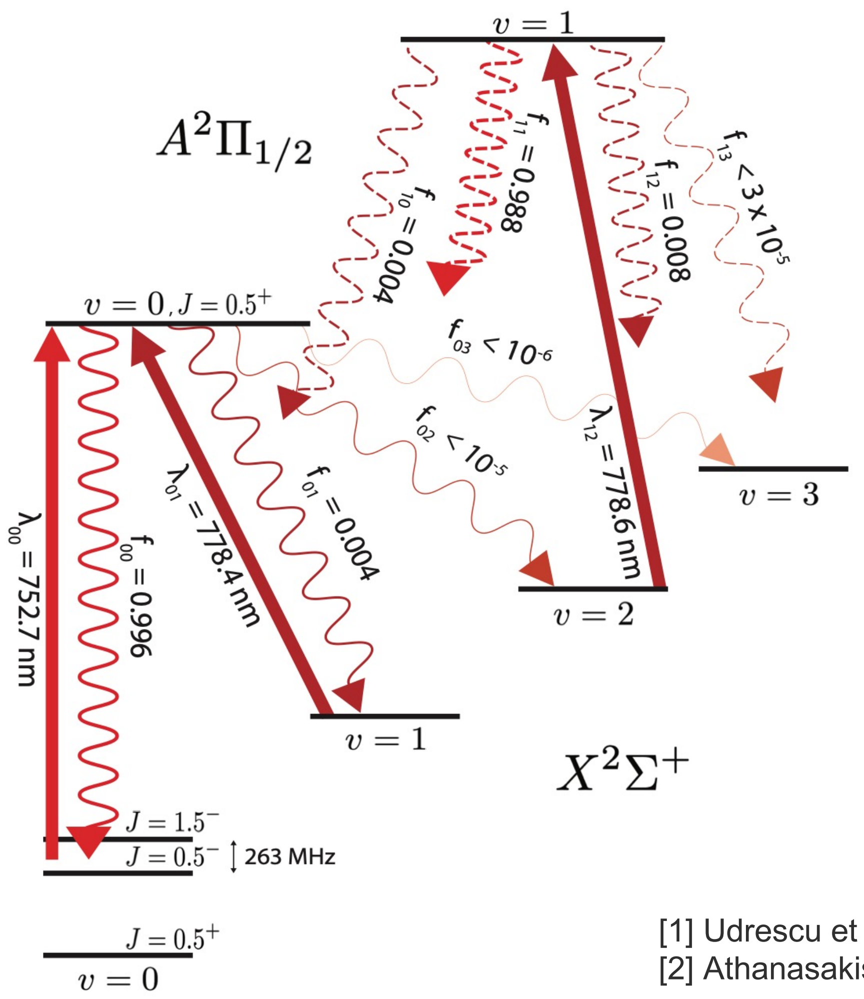


Image published in Garcia Ruiz et al, (2020) *Nature* **581**

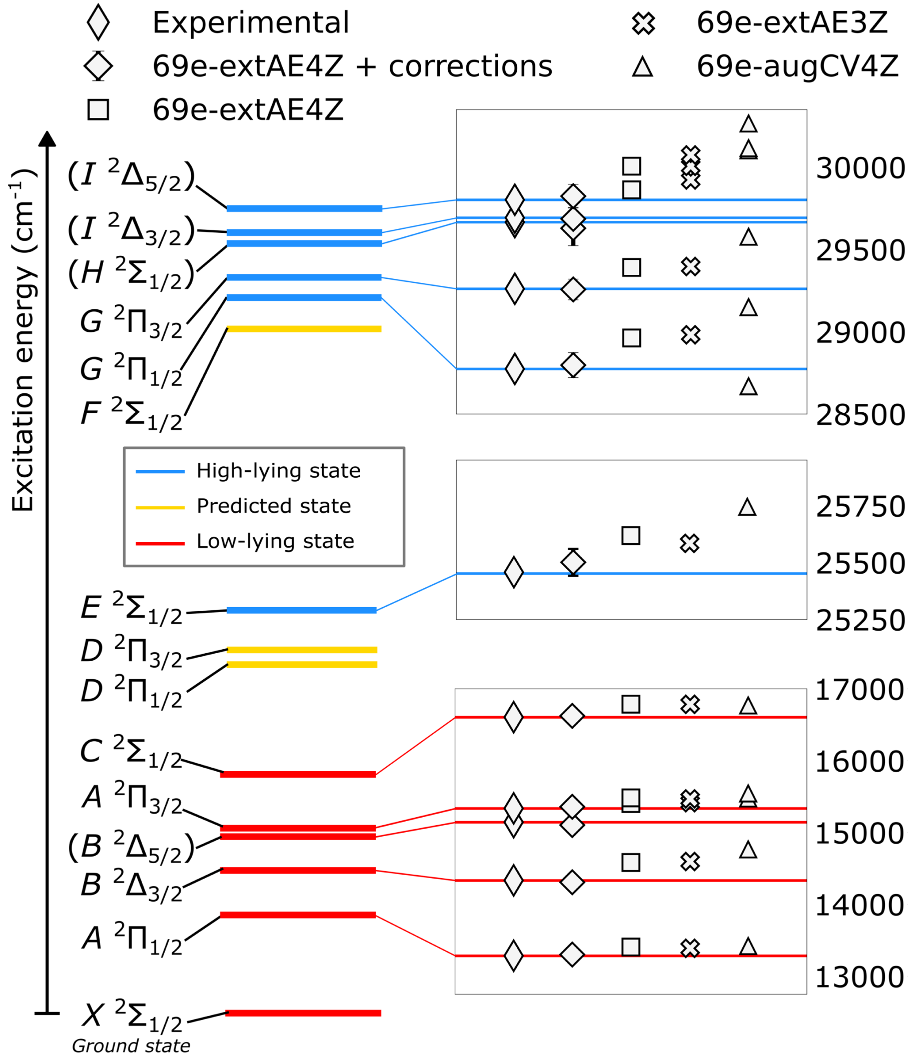
RaF characterization

Laser cooling [1]



Excited states [2]

- High-order electron correlation and QED corrections
- FS-RCC
- calculations: agreement ≥ 99.71% (~8 meV)
- Ground state sensitive to eEDM and nuclear parity violations [4]



[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)

Actinium Fluoride

Ac: Nuclear and atomic properties

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

AcF: molecular enhancement

- Enhanced sensitivity to effective T,P-violating interaction [1]
  - Electronic structure: ?

Production

- IP: ? D_e: ?

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 $|\bar{\theta}| < 10^{-10}$.

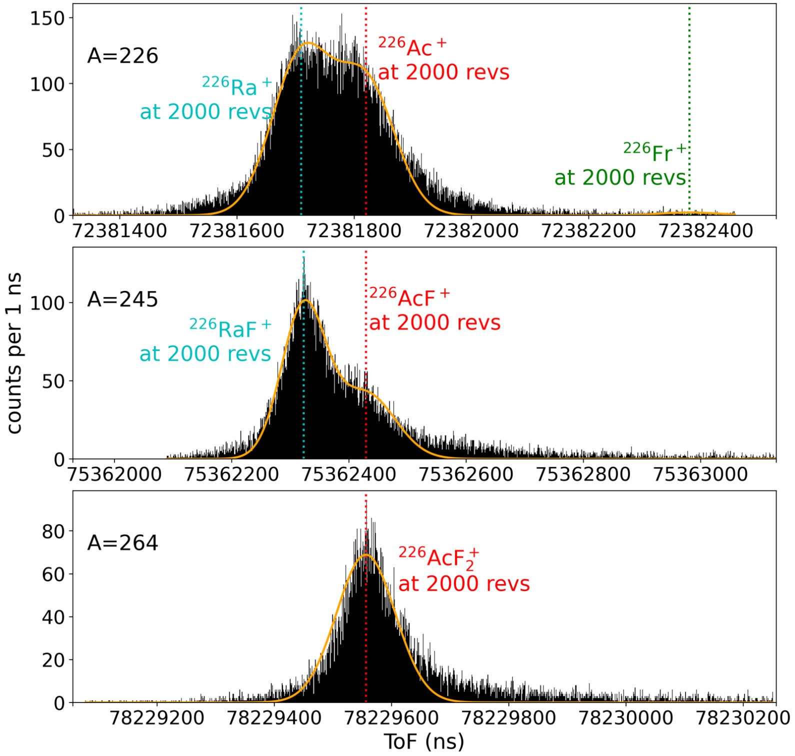
Z	Atom	S $[e\,\mathrm{fm}^3\bar{\theta}]$	$d_A[e\,\mathrm{cm}]$ $10^{-17}S[e\,\mathrm{fm}^3]$	$10^{-17}\bar{\theta}$
63	^{153}Eu	−3.7	−1.63	6
63	^{153}Eu^{3+}	−3.7	0.33	−1.2
66	^{161}Dy	$\lesssim 4$	−2.23	$\lesssim 9$
80	^{199}Hg	0.005	−2.50	−0.013
81	^{205,203}Tl^{+}	0.02	−2.79	−0.06
82	^{207}Pb^{2+}	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	$\lesssim 1$

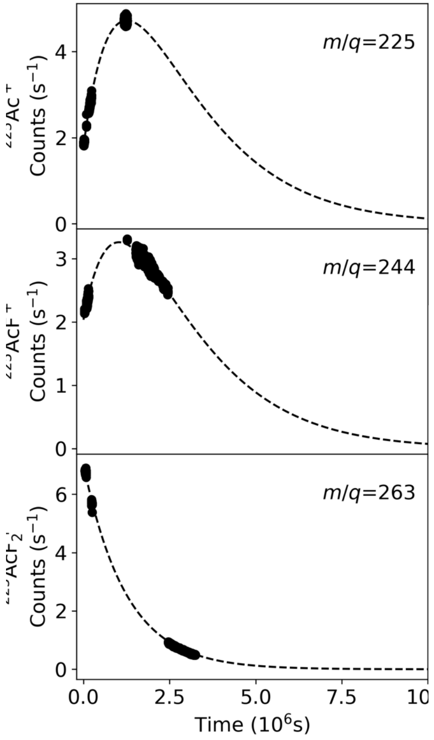
[a]Estimates for ^{229}Pa are presented assuming that the existence of a very close nuclear doublet level will be confirmed.

[1] Flambaum, Feldmeier, Phys. Rev. C. 101, 015502 (2020)
[2] Flambaum, Dzuba, Phys. Rev. A. 101, 042504 (2020)

Production of AcF_x^+

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

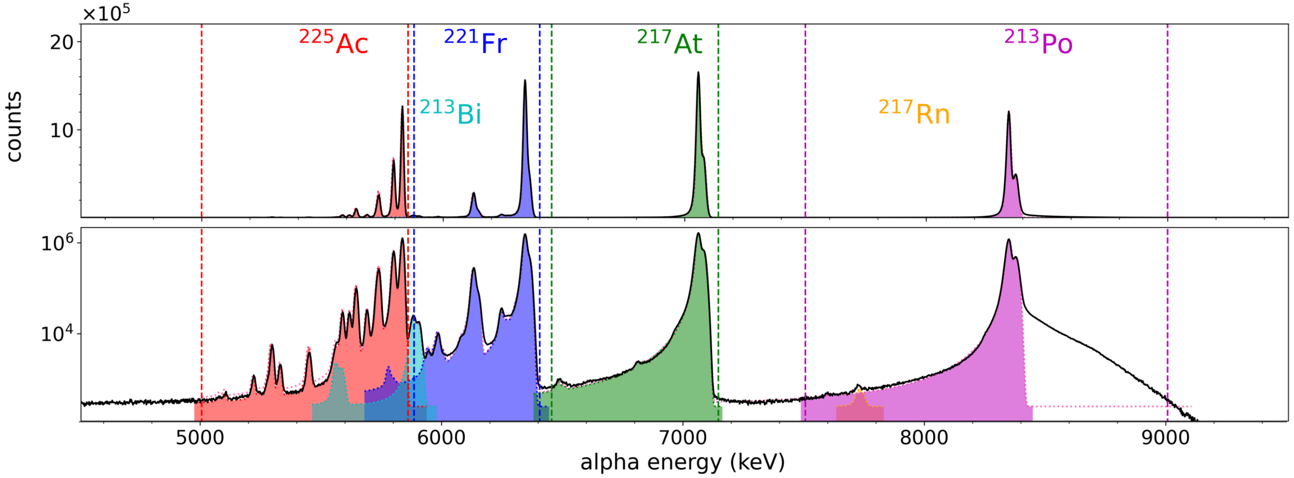




Extraction and separation of actinium molecular ion beams for production of medical radionuclide ^{225}Ac

M. Au,[1,2,*] L. Nies,[1] S. Stegemann,[1,3] M. Athanasakis-Kaklamanakis,[1,4] T.E. Cocolios,[4] P. Fischer,[3] P.F. Giesel,[3] J. Johnson,[4] U. Köster,[5] D. Lange,[6] M. Mougeot,[1,6,†] J. Reilly,[7] M. Schlaich,[8] Ch. Schweiger,[1,6] F. Wienholtz,[8] W. Wojtaczka,[4] Ch. E. Düllmann,[2,9,10] and S. Rothe[1]

In preparation (2023)



Au, *PhD thesis* (2023)

AcF spectroscopy

Experimental

- CRIS

Molecular theory

- IH-FS-RCCSD
- IP = 48,866 cm^{-1}
  - (6.06 eV)
- D_e = 57,214 cm^{-1}
  - (7.09 eV)

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

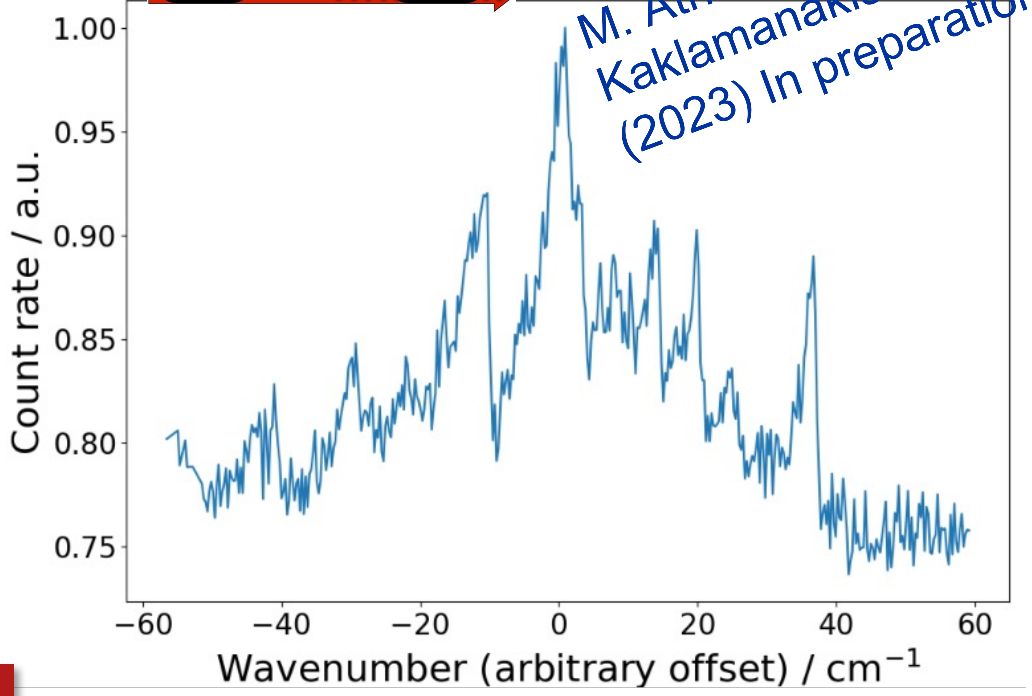
Laser ionization spectroscopy of AcF

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]

Athanasakis-Kaklamanakis et al, INTC-LOI-615 (2021)

CRIS



M. Athanasakis-Kaklamanakis et al., (2023) In preparation

arXiv > physics > arXiv:2305.06932

Physics > Atomic Physics

[Submitted on 11 May 2023]

Ab initio study of electronic states and radiative properties of the AcF molecule

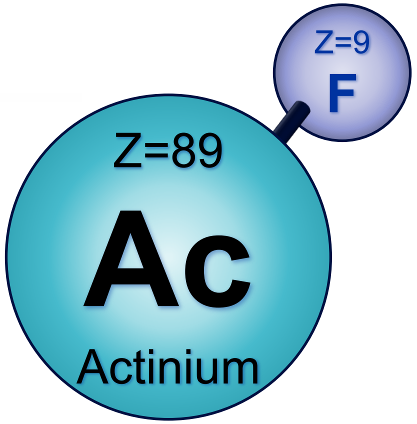
Leonid V. Skripnikov, Alexander V. Oleynichenko, Andréi Zaitsevskii, Nikolai S. Mosyagin, Michail Athanasakis-Kaklamanakis, Mia Au, Gerda Neyens

State	T_e, cm^{-1}	r_e, Å	ω_e, cm^{-1}	$\lvert d\rvert^2$, a.u.	$\langle L_z\rangle$, a.u.	Composition	Leading configurations
$(1)0^+$	0	2.110	541	–	0.0	100% $X(1)^1\Sigma^+$	92% $7s^{\sigma}_{1/2}7s^{\sigma}_{1/2}$
(8)1	26166	2.127	549	3.751	1.1	42% $(2)^1\Pi$ + 30% $(3)^3\Pi$	21% $7s^{\sigma}_{1/2}7p^{\pi}_{3/2}$ 19% $7s^{\sigma}_{1/2}7p^{\pi}_{1/2}$

First spectrum of an electronic transition in AcF obtained with broadband laser spectroscopy at the CRIS experiment,
Figure published in Athanasakis-Kaklamanakis and Au, (2023) *CERN EP newsletter*
https://ep-news.web.cern.ch/content/isolde-lays-ground-cp-violation-tests-radioactive-molecules

Conclusions

1. The ISOL method for molecular beams: In-source and in-trap formation
2. Radioactive molecules for BSM physics
3. RaF production, spectroscopy, characterization
4. AcF_x production, ID, AcF spectroscopy



Acknowledgements

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LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

Metastable molecular ions

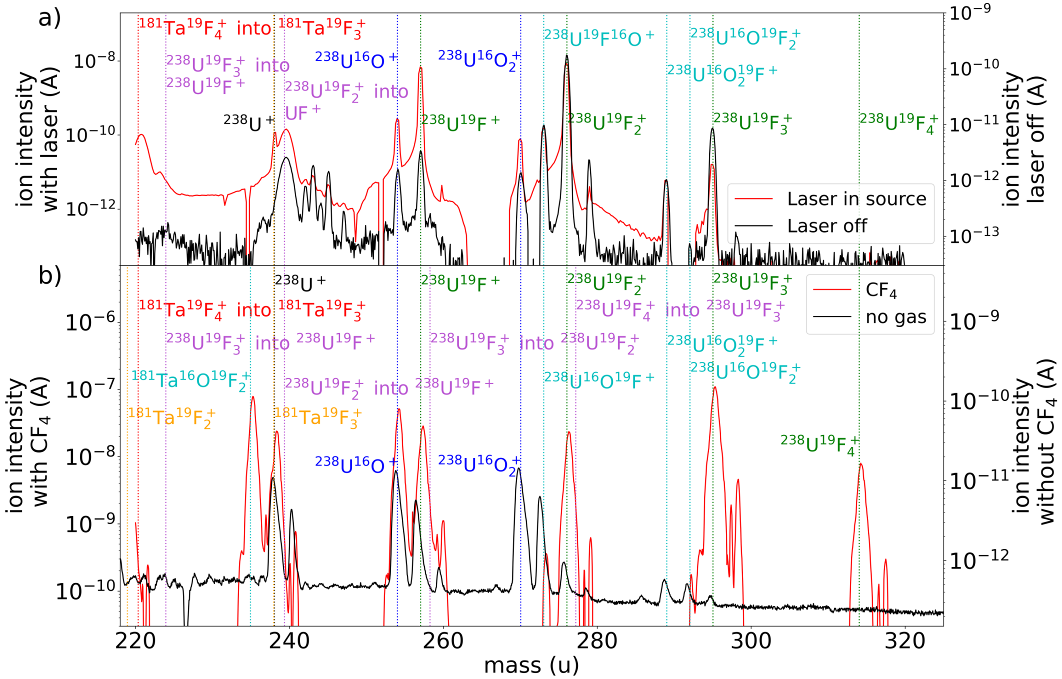
Dissociation **after** acceleration,
before mass separation.

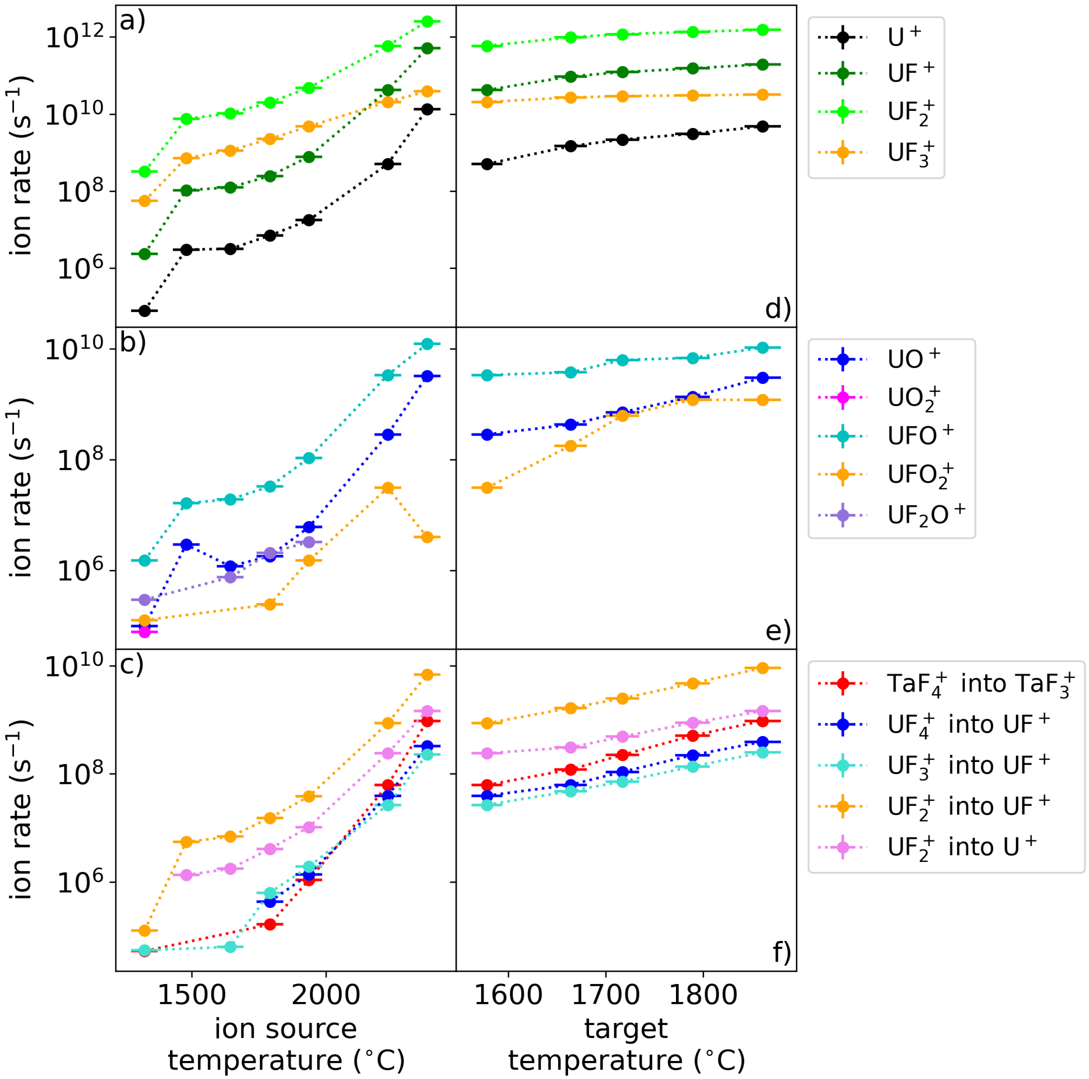
apparent mass: $m^* = \frac{m_f^2}{m_p}$

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

Ex: UF_2^+ into UF^+ + F

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





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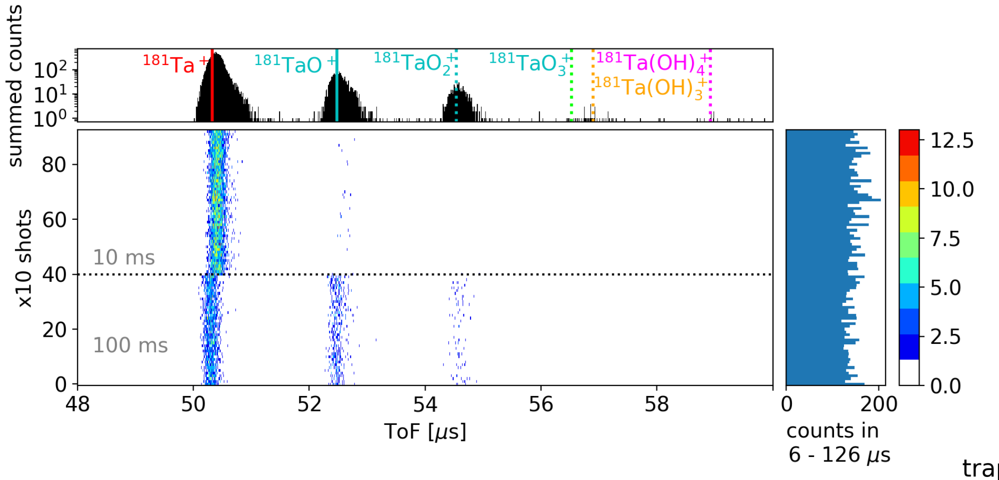
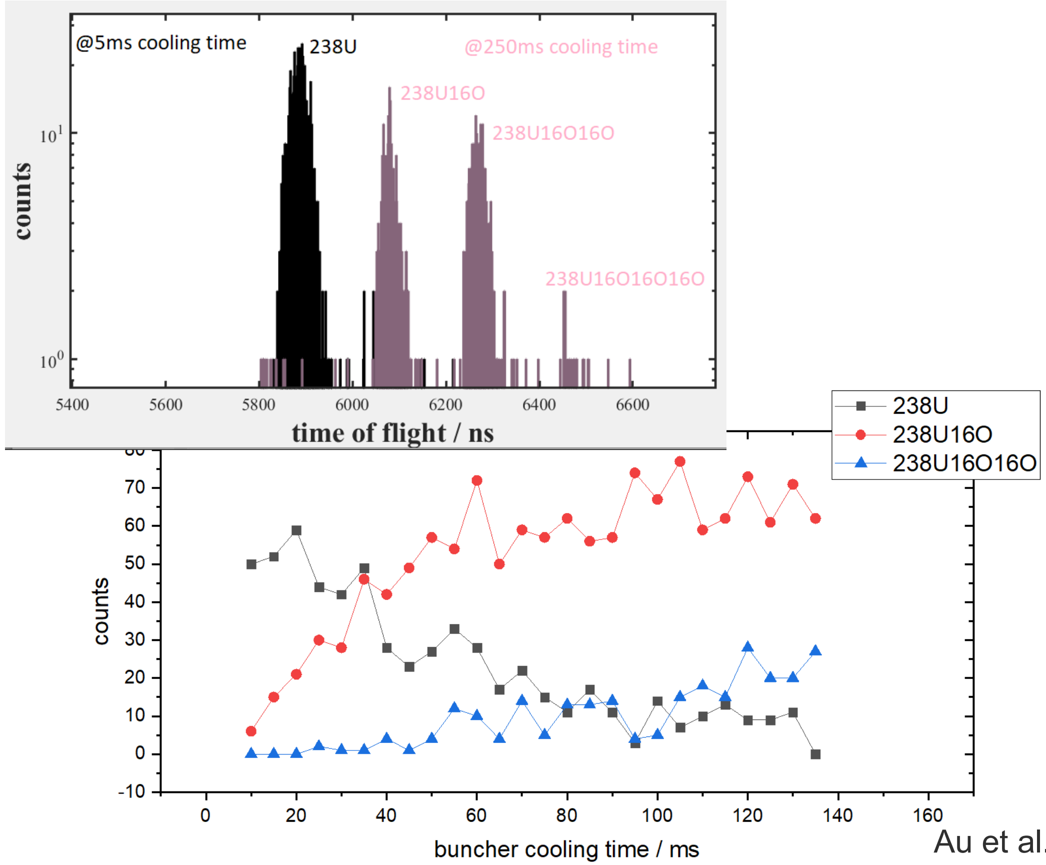
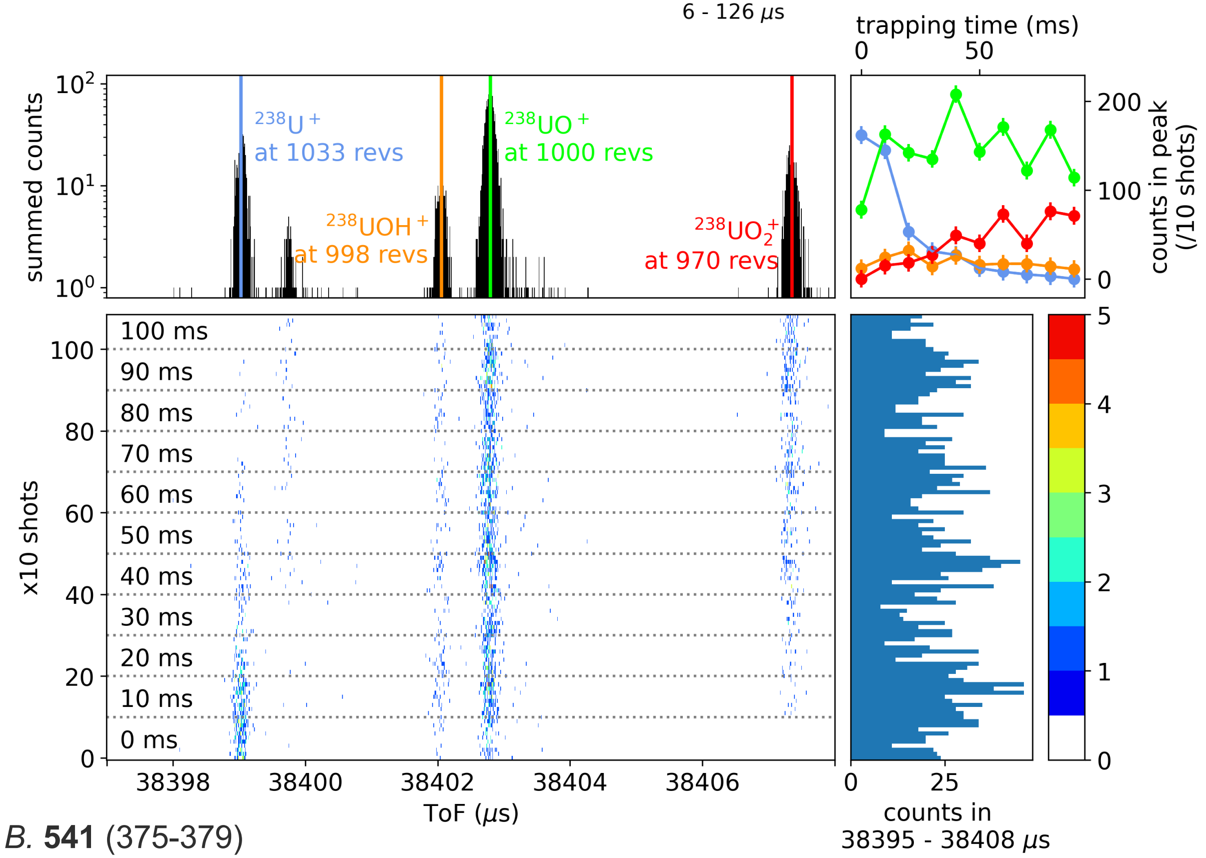
In-trap: identification

UO_x, TaO_x

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







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