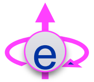
Fundamental physics with radioactive atoms and molecules at CERN-ISOLDE

Mia Au and Michail Athanasakis-Kaklamanakis

Low energy observables for BSM physics



eEDM

- Sensitive to electron-coupled CPV

Nuclear MQM

- Sensitive to nuclear CPV
- Enhanced in heavy nuclei
- Enhanced in quadrupole-deformed nuclei

Nuclear Schiff moment

- Sensitive to nuclear CPV
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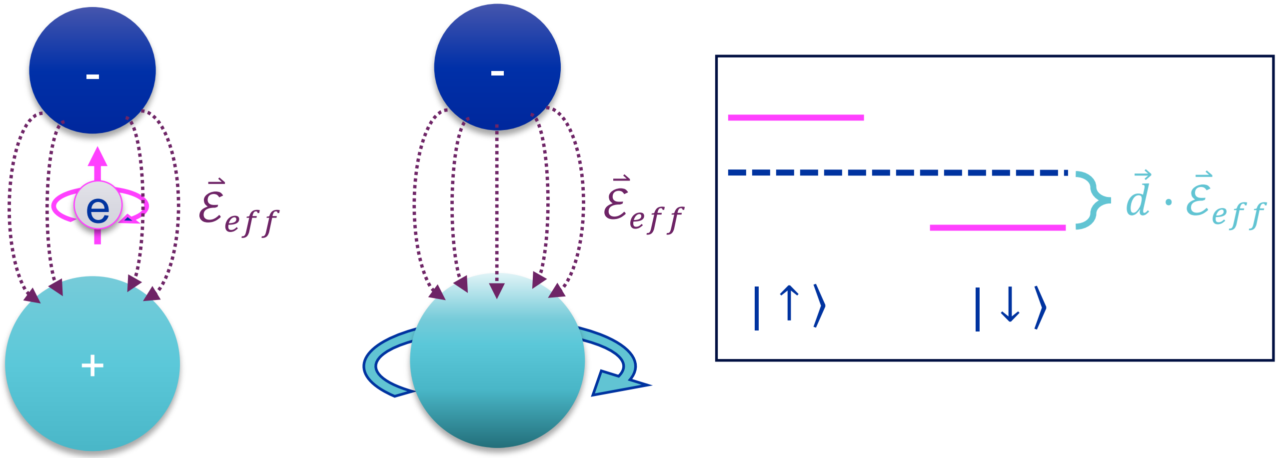
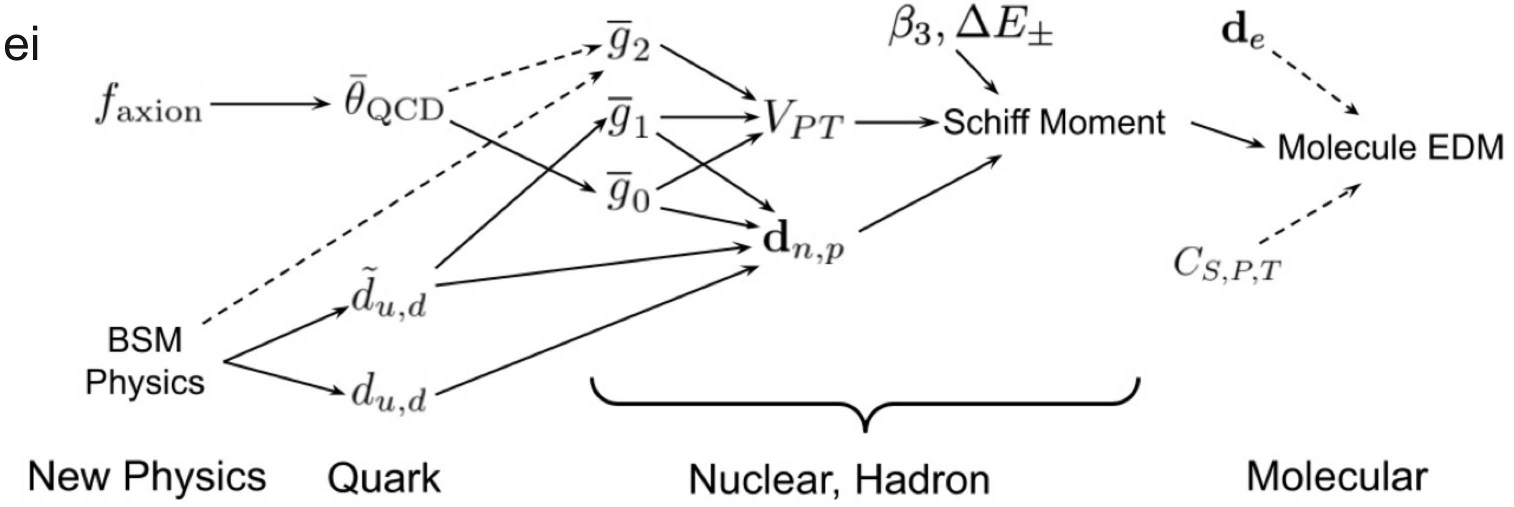
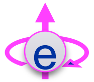




[1] Contribution to Snowmass 2021: *EDMs and the search for new physics*, arXiV 2203.08103 (2021)
[2] Safronova et al. (2018) *Rev. Mod. Phys.* 90. 2
[3] *Opportunities for Fundamental Physics Research with Radioactive Molecules*, arXiV 2302.02165 (2023), *Rep. Prog. Phys.* (2024) doi: *10.1088/1361-6633/ad1e39*

Low energy observables for BSM physics

eEDM



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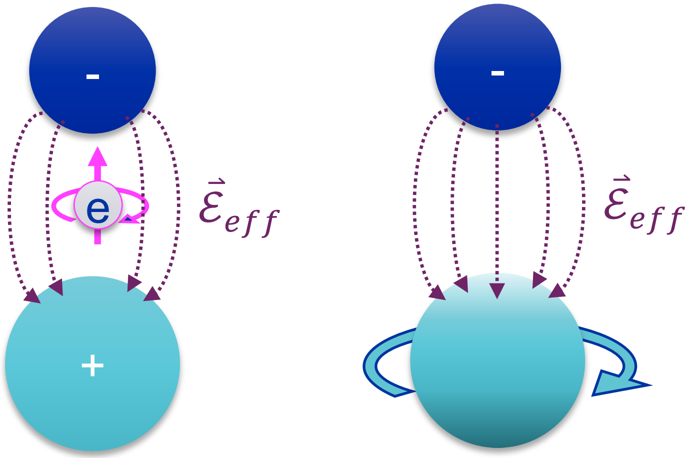

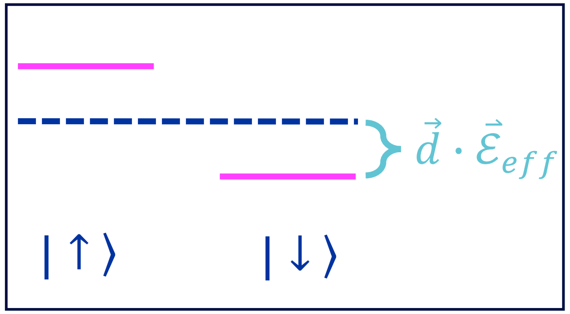

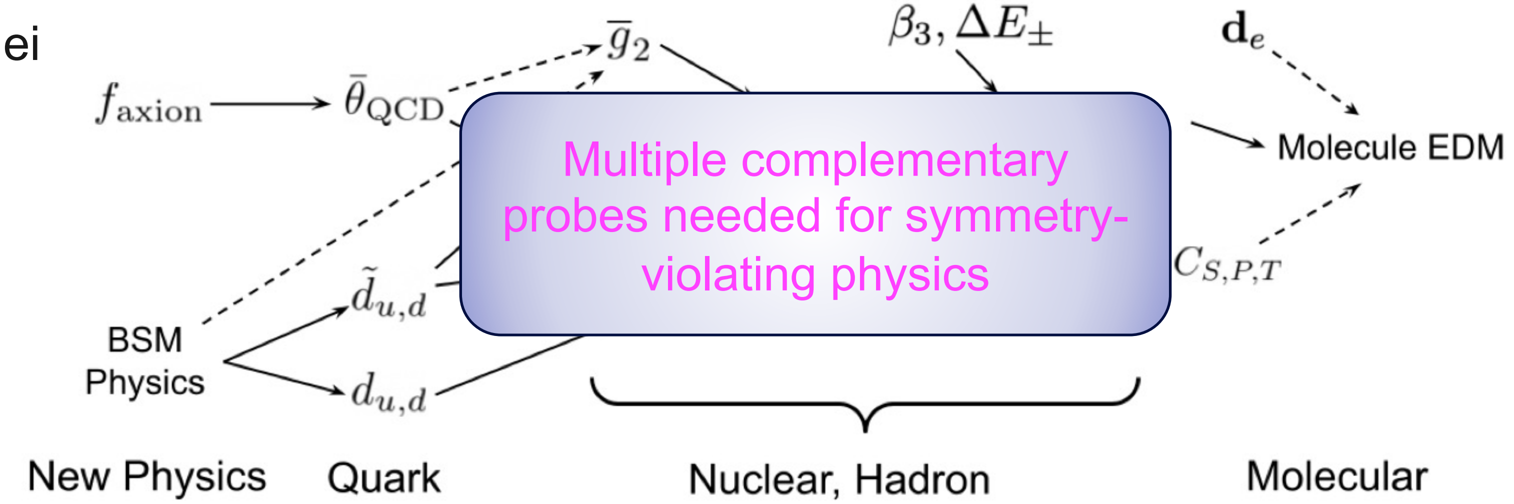

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EDM searches

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

nEDM $|d_n|$

- 2006 ILL UCNs: $|d_n| < 2.9\times10^{-26}$ *e* cm
- 2020 PSI [1]: $|d_n| < 1.8\times10^{-26}$ *e* cm

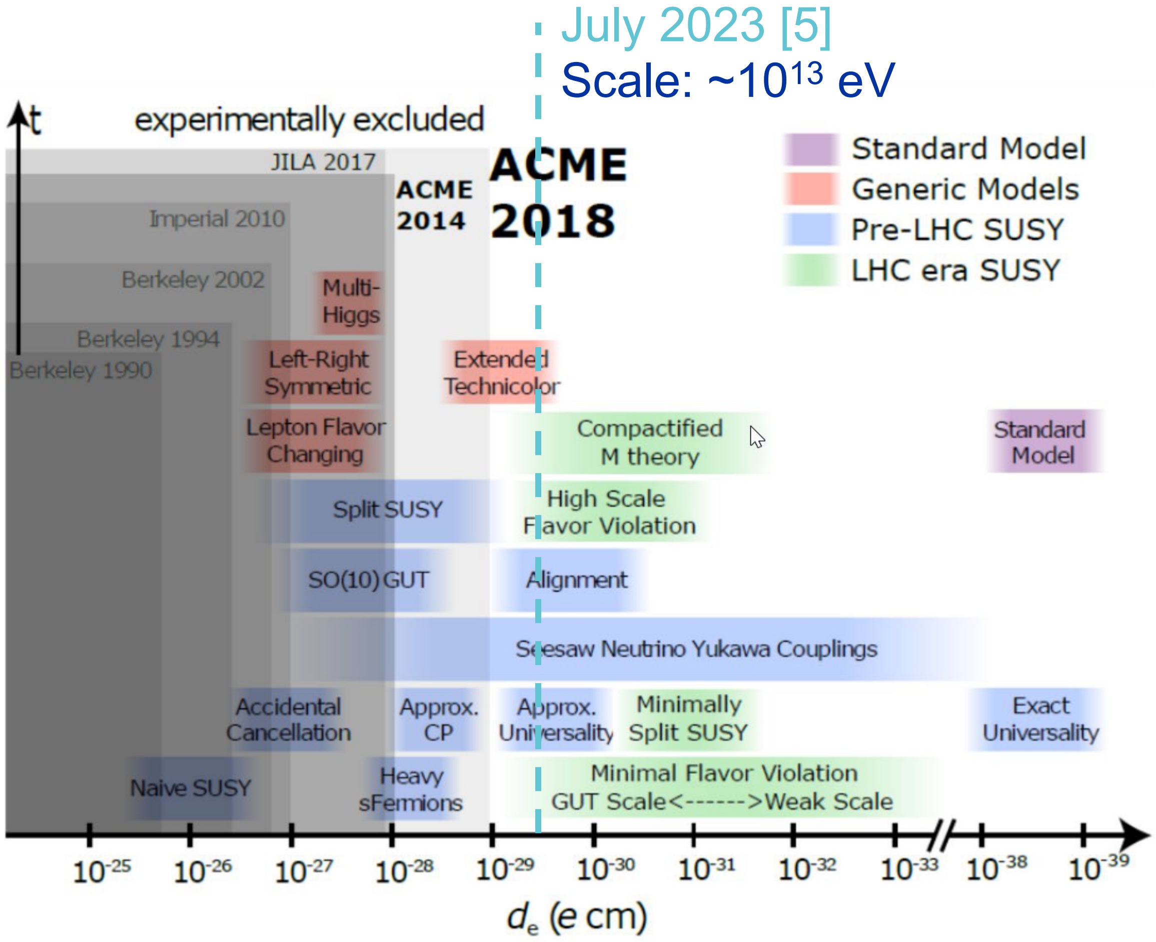
eEDM $|d_e|$

- 2011 Imperial $^{174}Yb^{19}F$ [2]: $|d_e| < 2\times10^{-28}$ *e* cm
- 2018, 2013 ACME $^{232}Th^{16}O$ [3,4]: $|d_e| < 1\times10^{-29}$ *e* cm
- 2023, 2017 JILA $^{180}Hf^{19}F^{+}$ [5]: $|d_e| < 4.1\times10^{-30}$ *e* cm

Atomic EDM

- 2009, ^{199}Hg: $|d| < 3.1\times10^{-29}$ *e* cm
- 2015, ^{225}Ra [6]: $|d| < 5\times10^{-22}$ *e* cm
- 2020, ^{171}Yb [7]: $|d| < 1.5\times10^{-26}$ *e* cm

[1] PRL 124, 081803 (2020)
[2] Nature 473, 493 (2011)
[3] Science 343, 269 (2014)
[4] Nature 562, 355 (2018)
[5] Science 381, 46 (2023)
[6] PRC 94, 025501 (2016)
[7] PRL 129, 083001 (2020)



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

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- 2011 Imperial $^{174}Yb^{19}F$ [2]: $|d_e| < 2{\times}10^{-28}$
- 2018, 2013 ACME $^{232}Th^{16}O$ [3,4]: $|d_e| < 1$
- 2023, 2017 JILA $^{180}Hf^{19}F^{+}$ [5]: $|d_e| < 4.1{\times}1$

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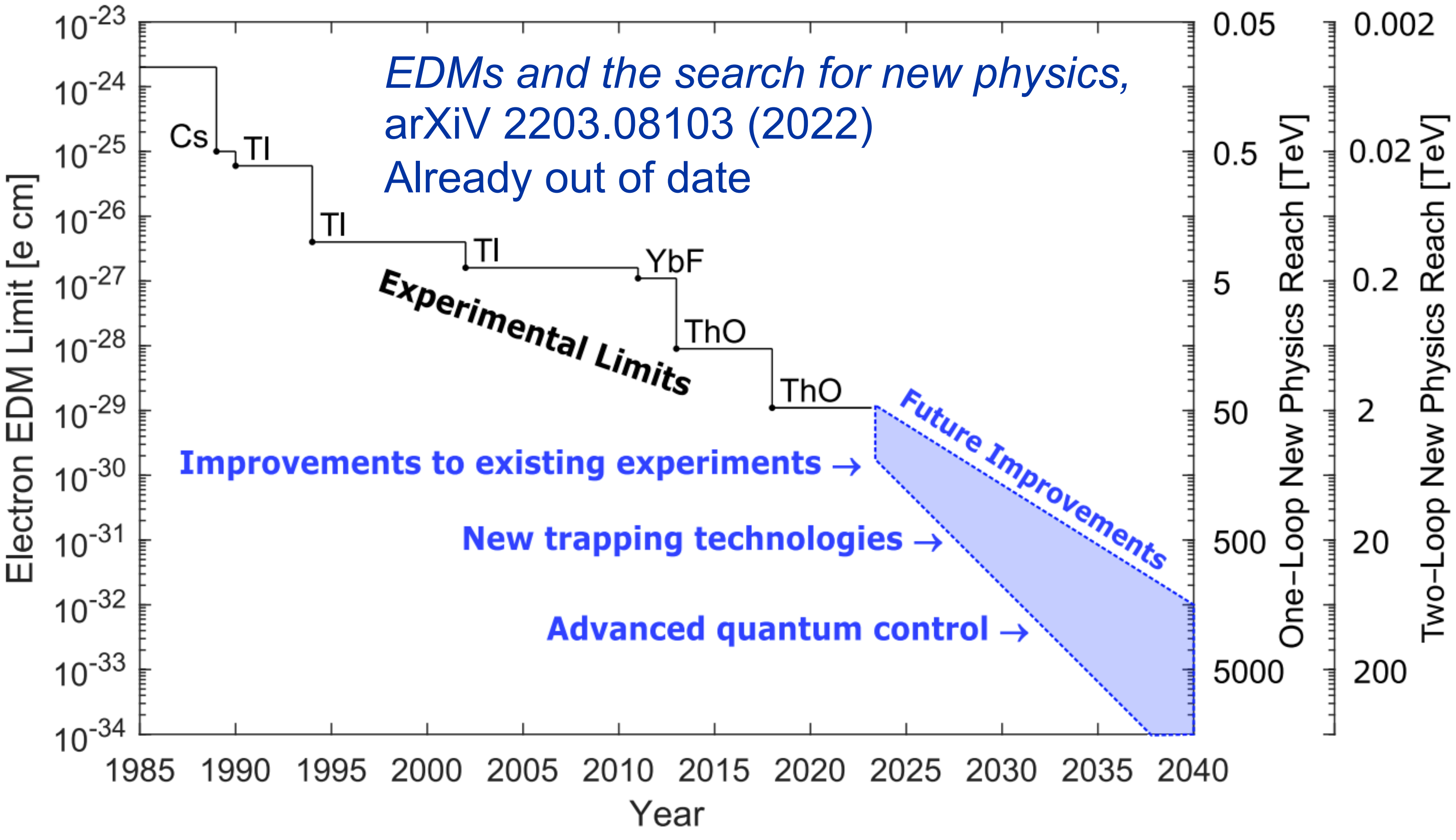


D. DeMille, The ACME Experiment, (2023)
https://cfp.physics.northwestern.edu/gabrielse-group/acme-electron-edm.html

CPV in molecules

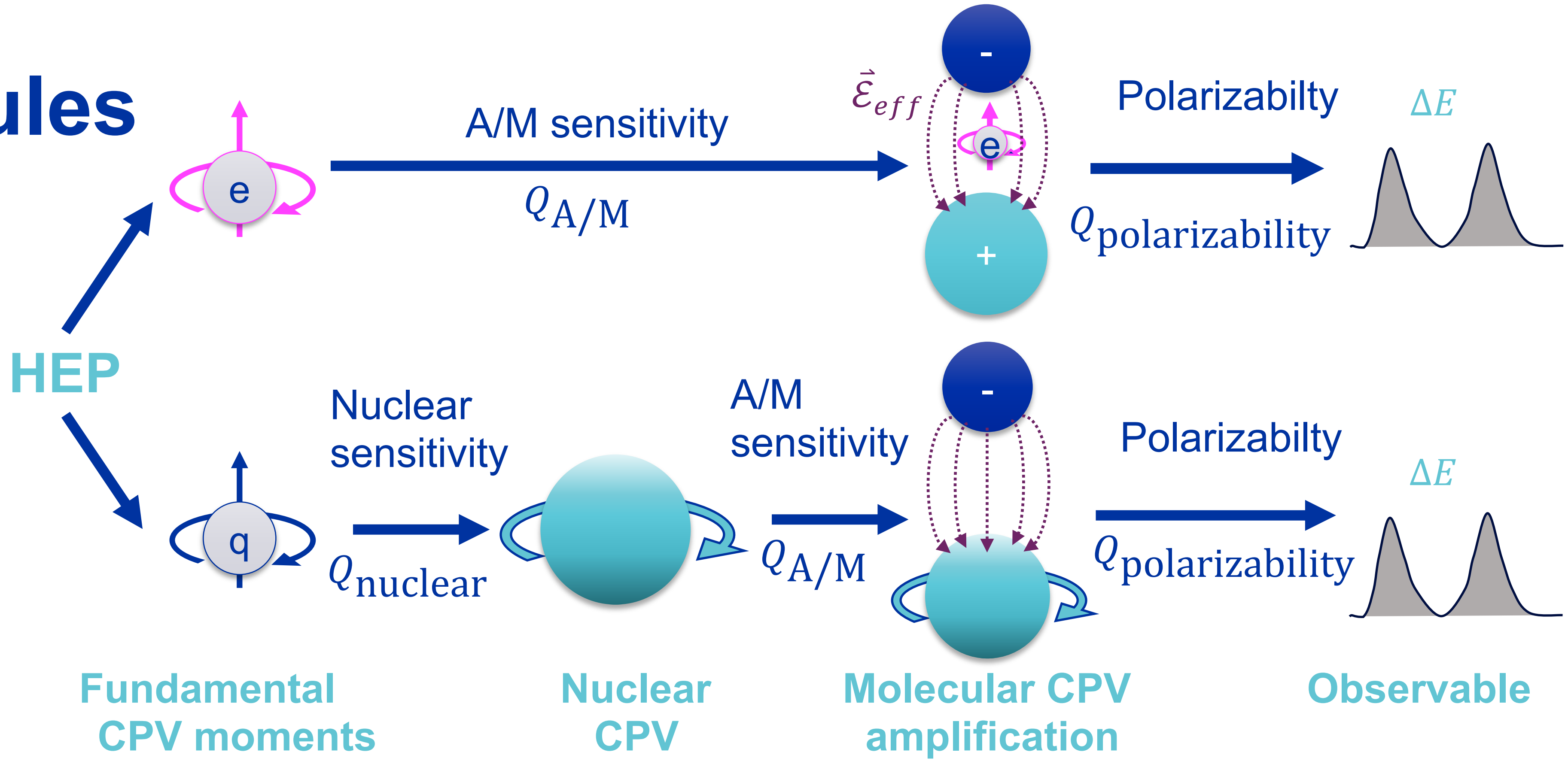
CPV searches (EDM searches)

- using a strong electric field and looking for spin precession

Molecular enhancement

- $\vec{\mathcal{E}}_{eff}$ along molecular axis ~GV / cm
- Polar molecules can be aligned with lab axis with weak fields (~V / cm)

→ Molecular searches are exceptionally sensitive to leptonic, hadronic, and nuclear EDMs



Statistical uncertainty scales as

$$\frac{1}{Q_{\mathrm{eff}}\,\tau\,\sqrt{N}}$$

Sensitivity

Extremely large for polar radioactive molecules

Spin precession time

Number of particles

Pathway to improved limits on EDMs

Statistical uncertainty scales as

$$\frac{1}{Q_{\mathrm{eff}}\,\tau\,\sqrt{N}}$$

Efficiently produced and trapped radioactive molecules

$\sqrt{N}$ → produced

τ → trapped

$Q_{\mathrm{nuclear,AM,polarizability}}$ → radioactive

Pathway to improved limits on EDMs

Statistical uncertainty scales as

$$\frac{1}{Q_{\text{eff}}\,\tau\,\sqrt{N}}$$

Efficiently produced and trapped radioactive molecules

$\sqrt{N}$ → produced; τ → trapped; $Q_{\text{nuclear,AM,polarizability}}$ → radioactive

CPV

Atom (spherical nucleus)	$Q_{\text{A/M}}$ →	**State of the art**
Atom (deformed nucleus)	$Q_{\text{A/M}}$ → Q_{nuclear} →	**~1000x**
Molecule (spherical nucleus)	$Q_{\text{A/M}}$ → $Q_{\text{polarizability}}$	**~1000x**
Molecule (deformed nucleus)	$Q_{\text{A/M}}$ → Q_{nuclear} → $Q_{\text{polarizability}}$ →	**~10^6 x**

Conceptualization courtesy of N. Hutzler (2024)

Pathway to improved limits on EDMs

Statistical uncertainty scales as

$$\frac{1}{Q_{\text{eff}}\, \tau \sqrt{N}}$$

Efficiently produced and trapped radioactive molecules

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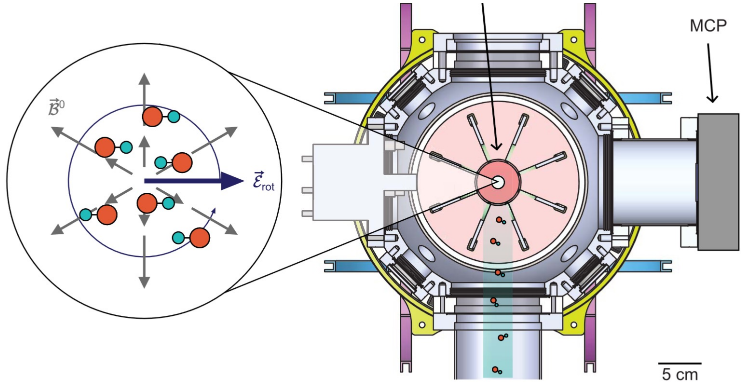
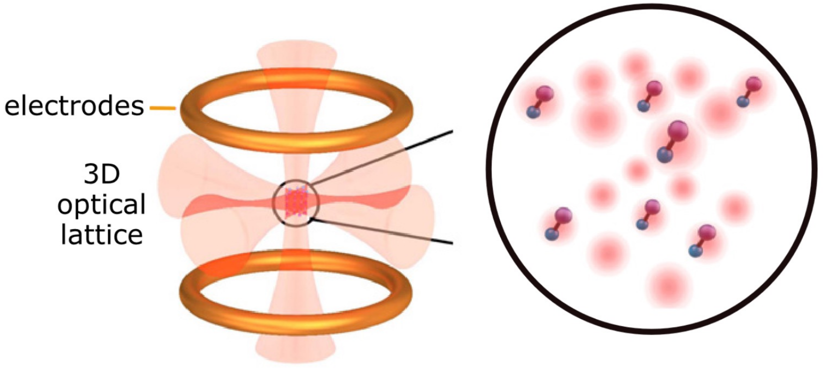
Ion trapping



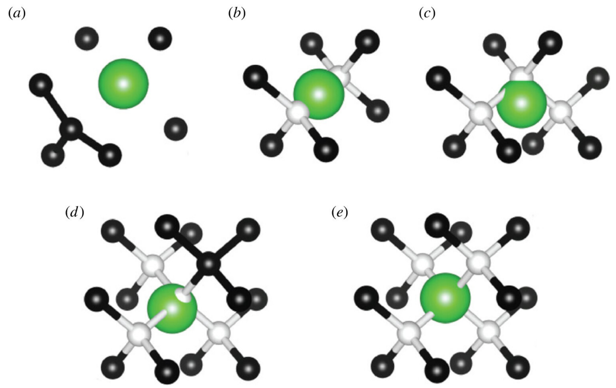
Roussy et al., *Science* 381, 46 (2023)

MOT → Optical lattice



Fitch et al., *Q.Sci.T.* 6, 014006 (2021)

Solid-state ensembles



Morris et al., *Phil. Trans. R. Soc. A.* 3822 (2023)

Pathway to improved limits on EDMs

Statistical uncertainty scales as $\dfrac{1}{Q_{\mathrm{eff}}\,\tau\,\sqrt{N}}$

Efficiently produced and trapped radioactive molecules

$\sqrt{N}$ → produced τ → trapped $Q_{\mathrm{nuclear,AM,polarizability}}$ → radioactive

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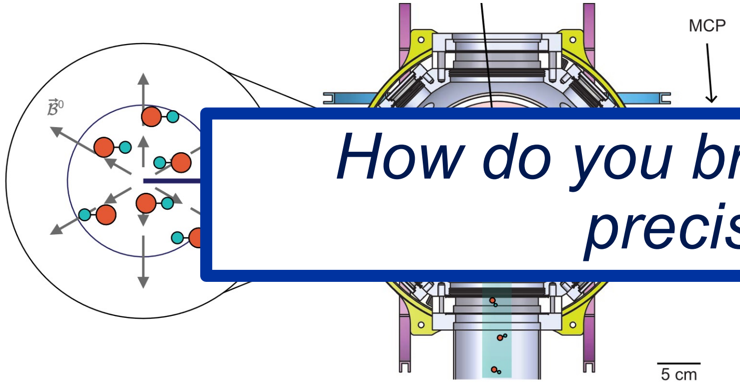

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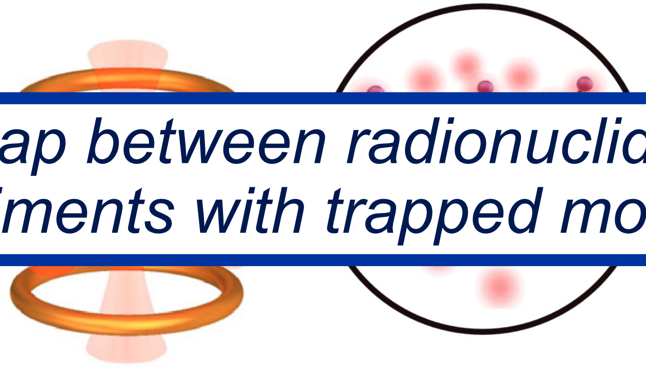

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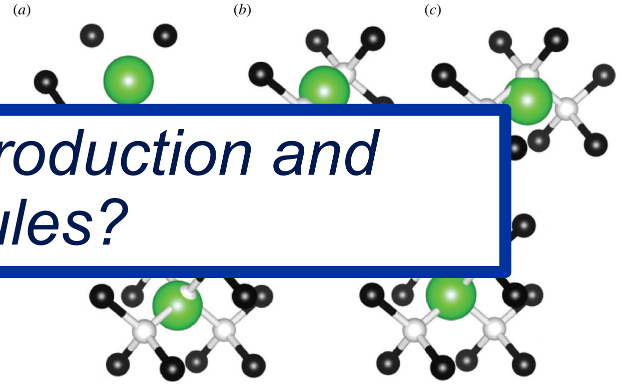


Morris et al., *Phil. Trans. R. Soc. A.* 3822 (2023)

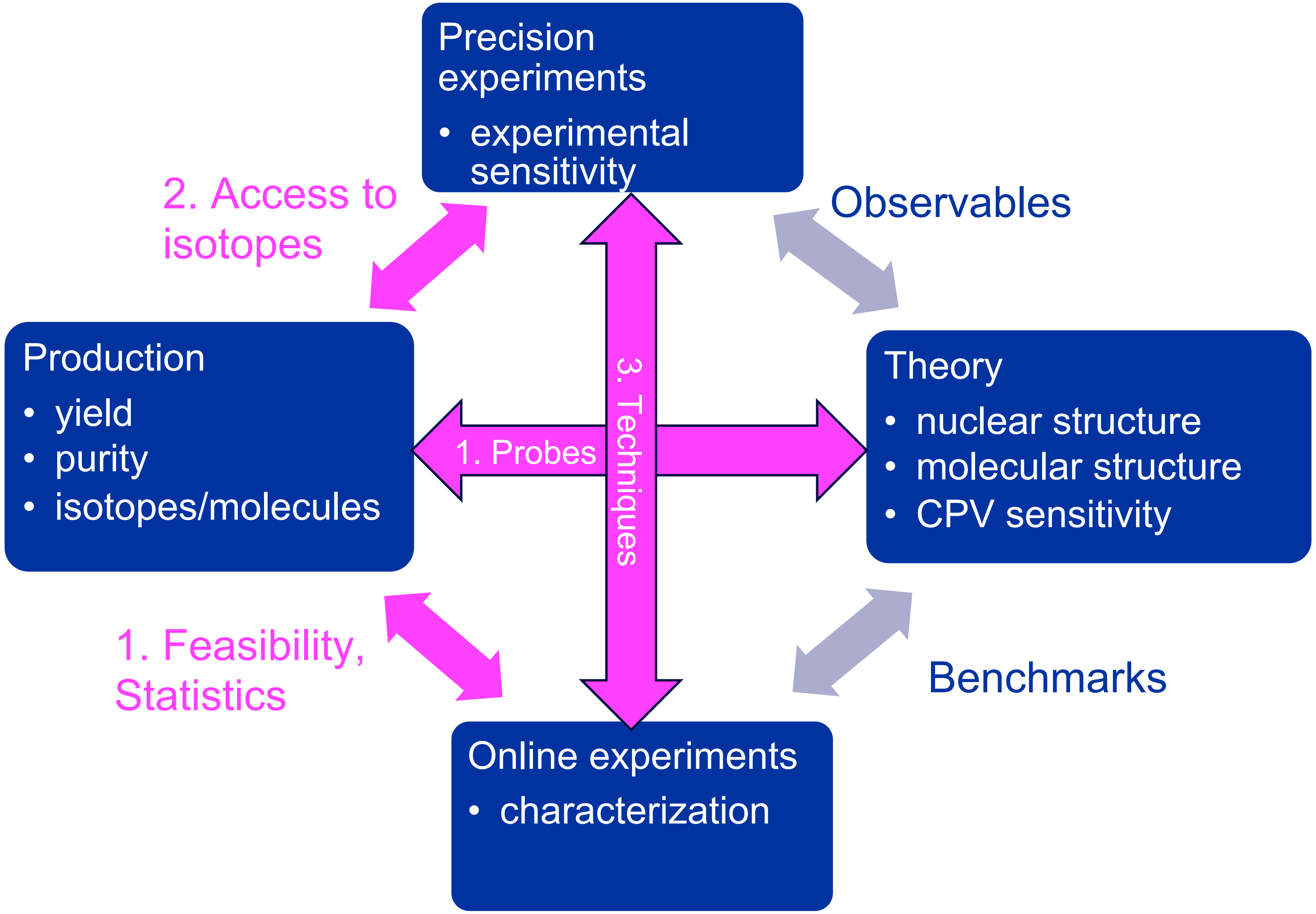
How do you bridge the gap between radionuclide production and precision experiments with trapped molecules?

Proposal

1 Access to new probes for online experiments

2 Production for offline experiments

3 Precision for online experiments

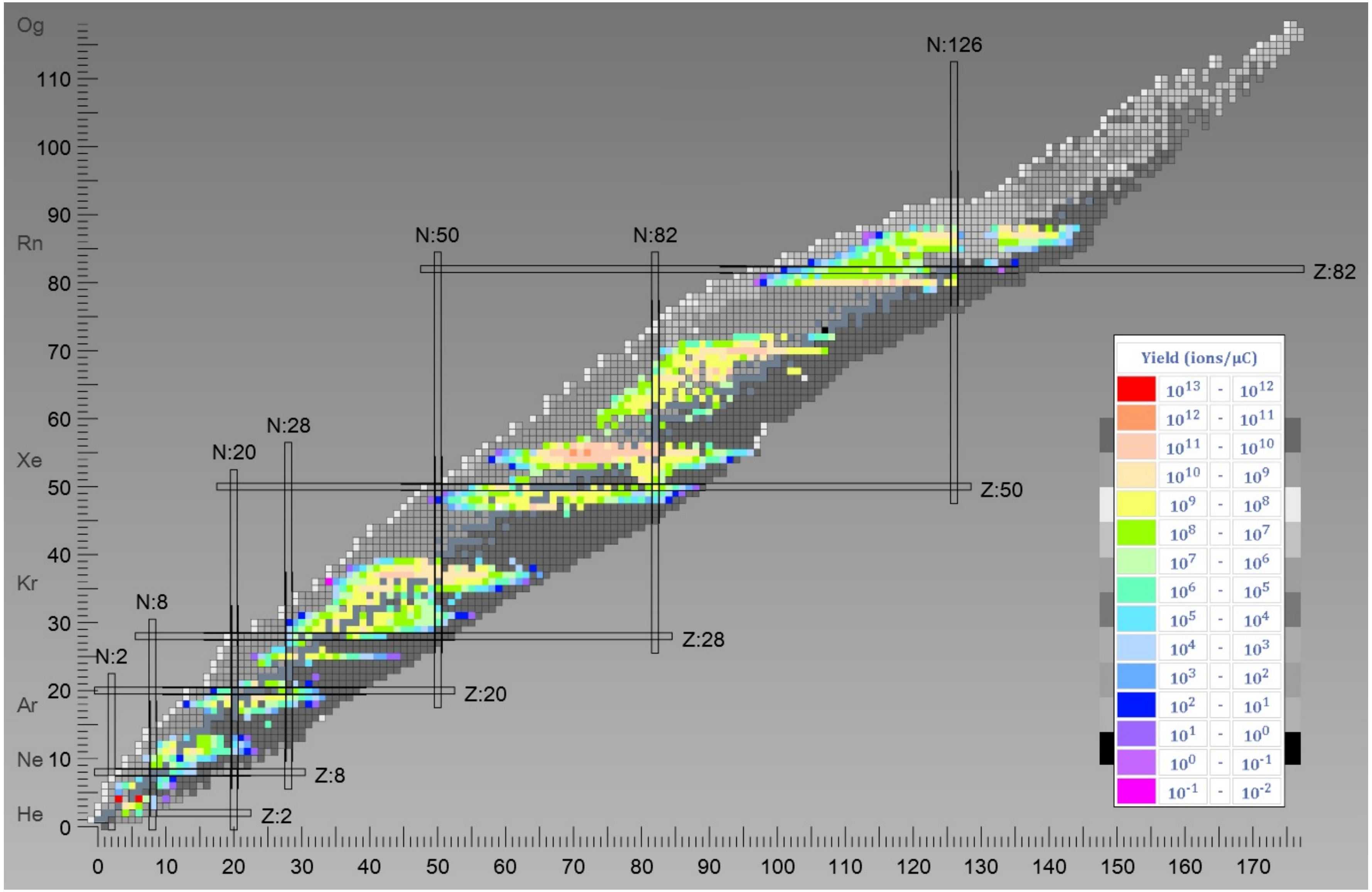


4 Towards precision measurements with radioactive molecules

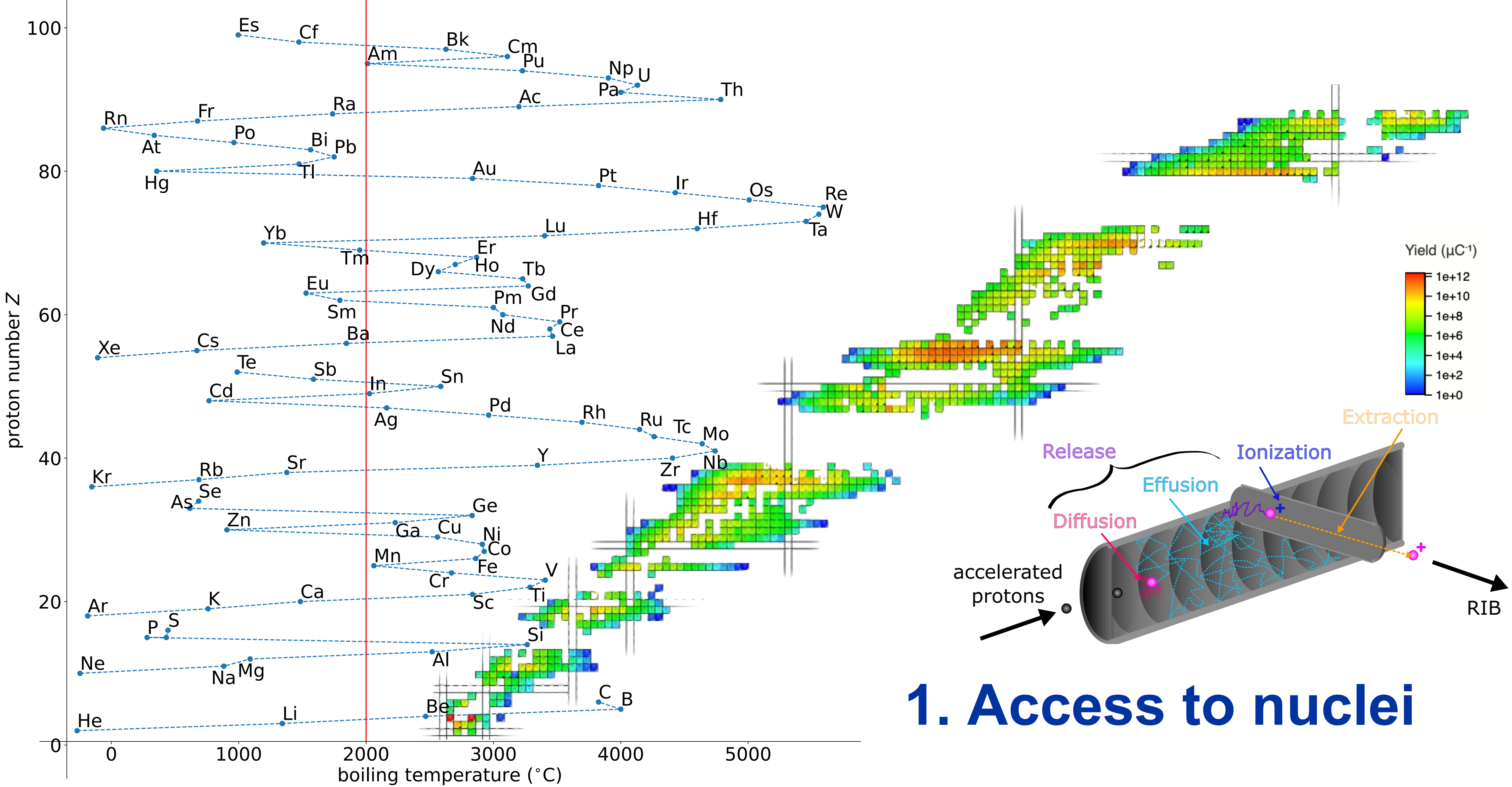
CERN-ISOLDE

>1000 isotopes and isomers

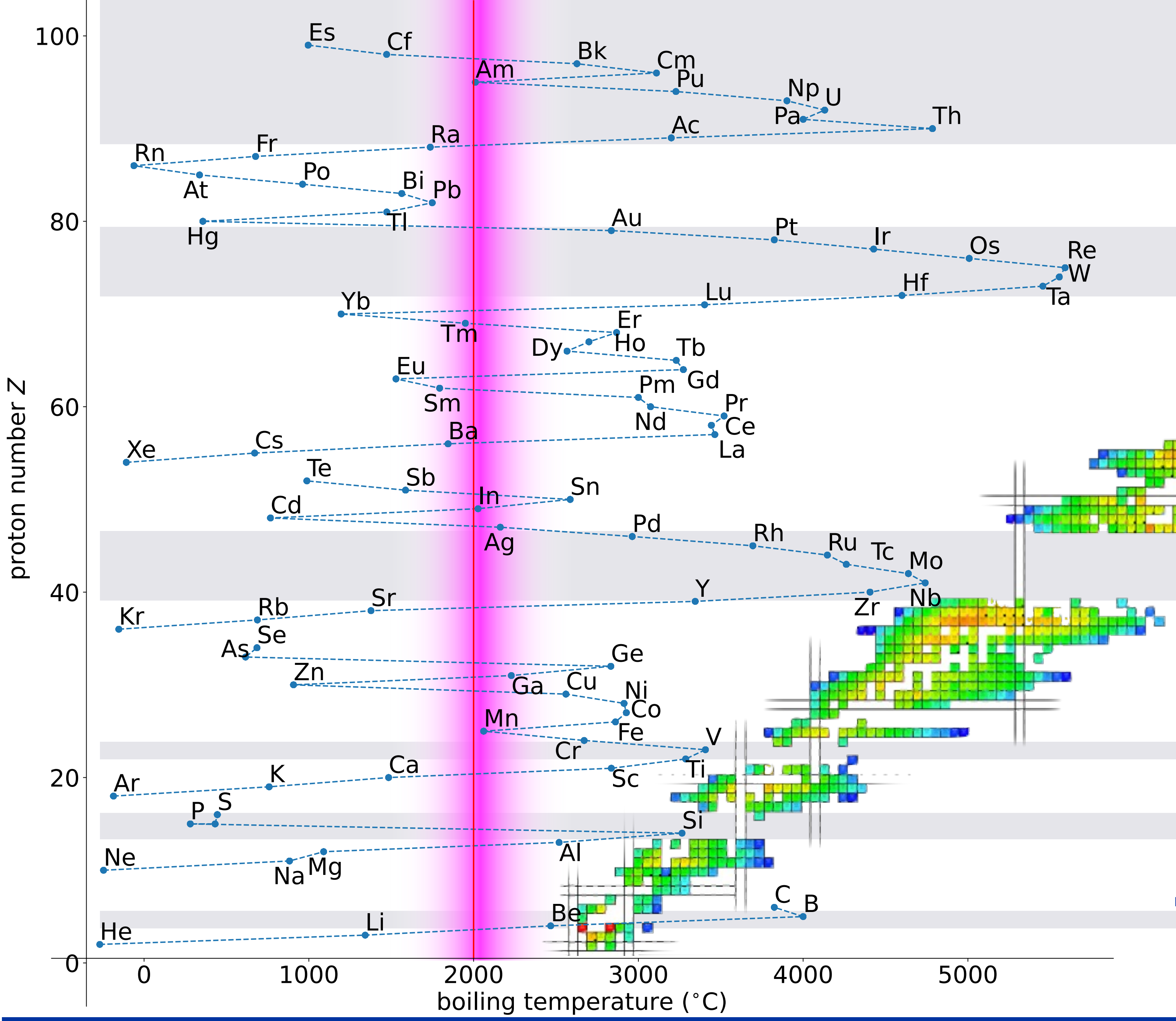
74 elements

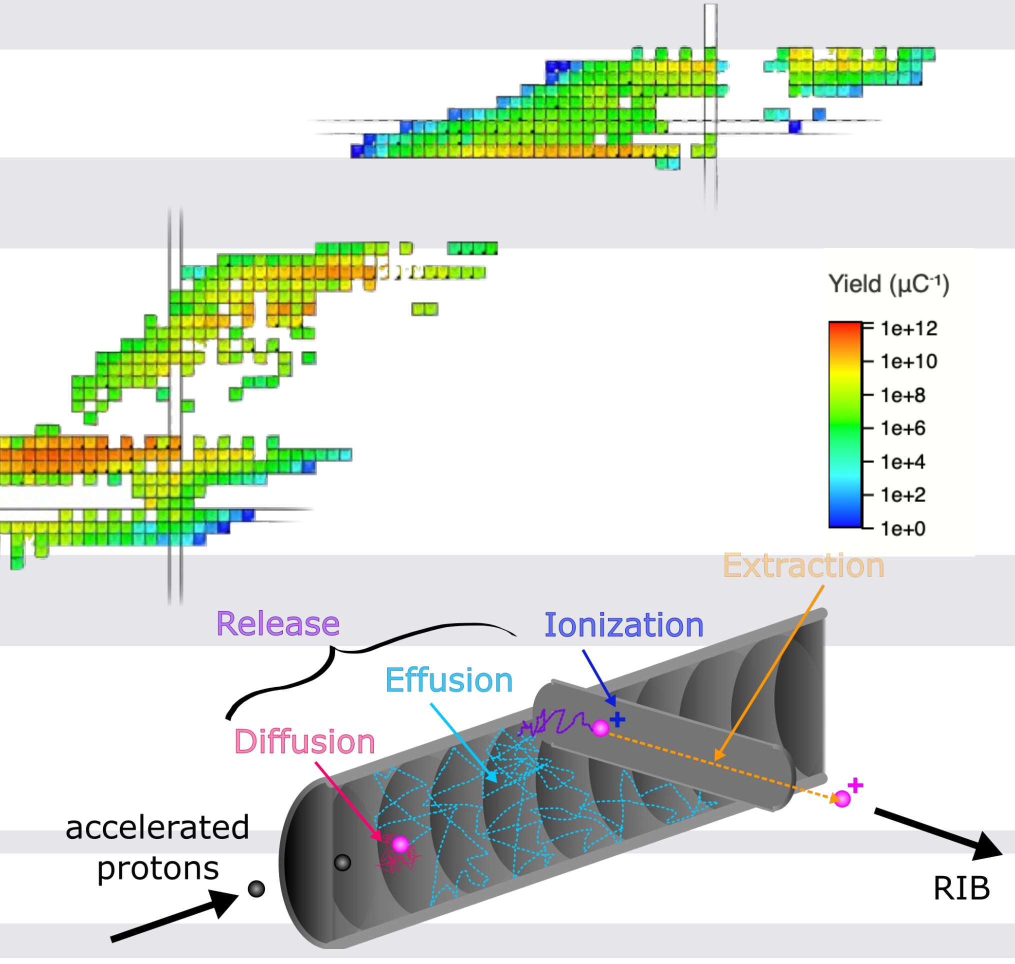


www.nucleonica.com, *Dataset: JEFF-3.1 Nuclear Data Library, NEA (2023)*
Ballof *et.al,* (2020) *NIM B* **463**, 211-215
cern.ch/isolde-yields



1. Access to nuclei





1. Access to nuclei

2. Production of radioactive molecules for offline experiments

Facilities for ISOLDE TISD

- MEDICIS, GLM, LA1, YOL

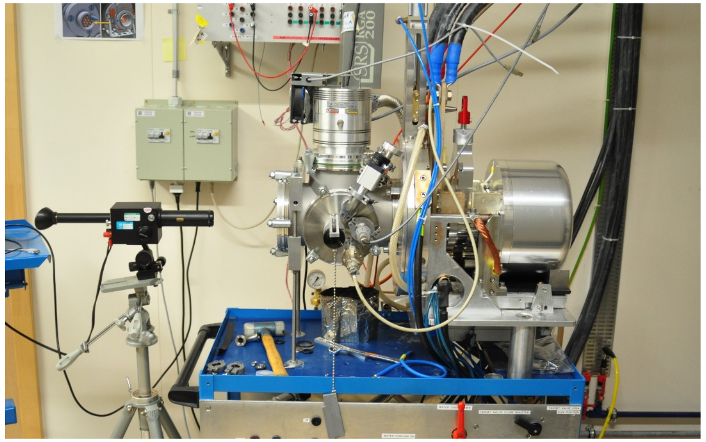
Proposal

- Feasibility studies, efficiency characterization of isotope collection and transport

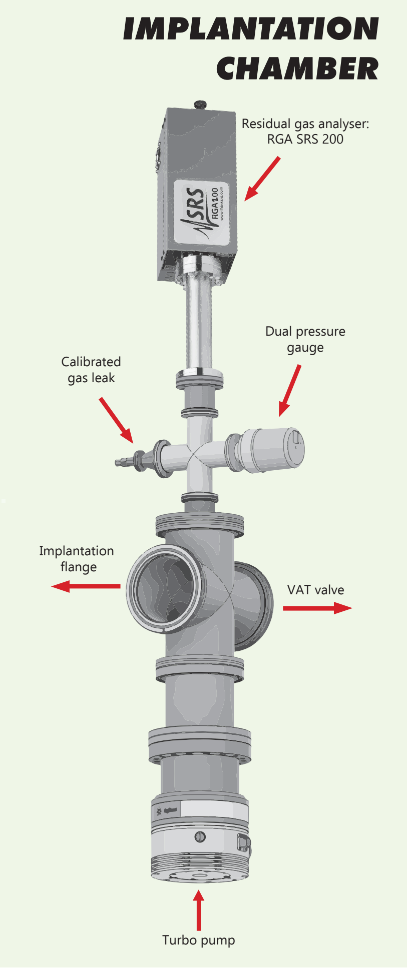
Extension and collaboration

- Ablation and measurement:
- Imperial College London, Hutzler lab (Caltech), EMA (MIT), RAFICI (University of Edinburgh)

ISOLDE OFFLINE 1 © 2019-2022 CERN



ISOLDE PUMP STAND © 2019-2022 CERN

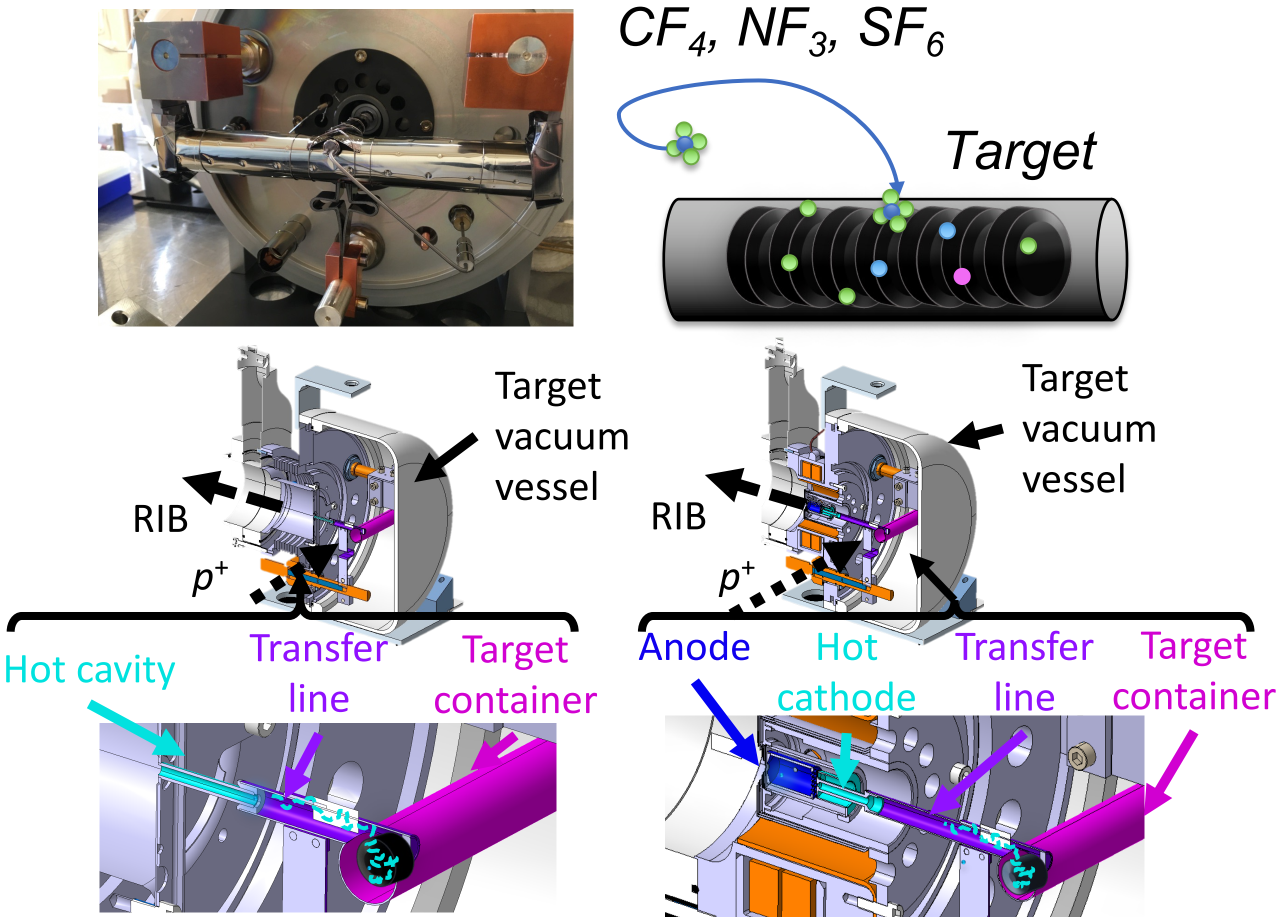


[1] Au et al. (2023) *NIM B.* **541** (144-147)
[2] Wojtaczka et al. (2023) *ICIS'23,* Victoria, Canada

3. Online experiments

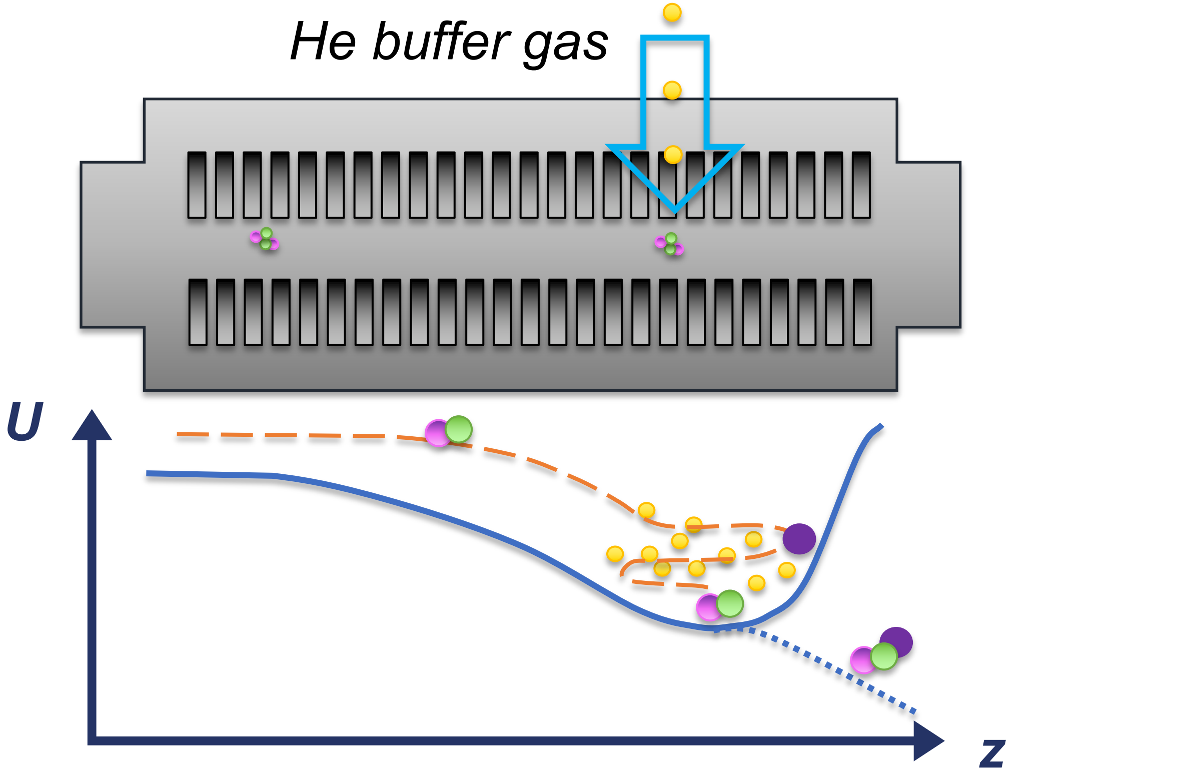
In-source

- Reactive gas



In-trap

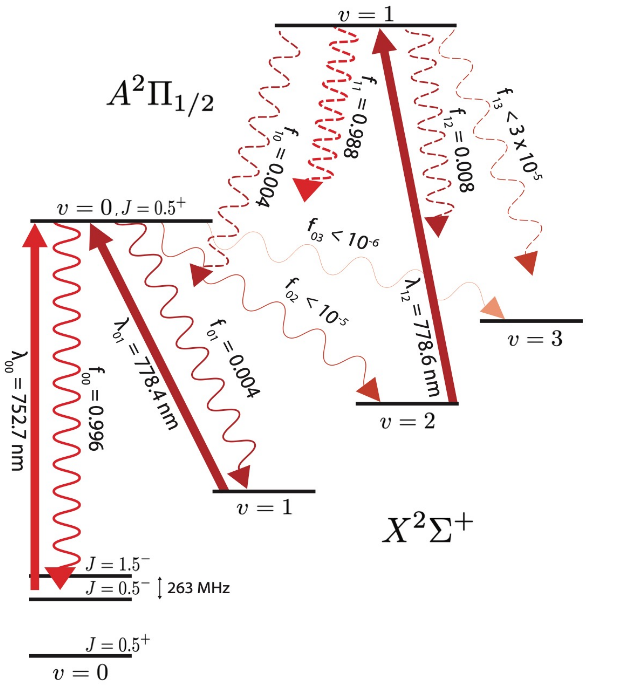
- Radio-frequency quadrupole cooler-buncher (RFQ-cb)



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

RaF characterization at CERN-ISOLDE

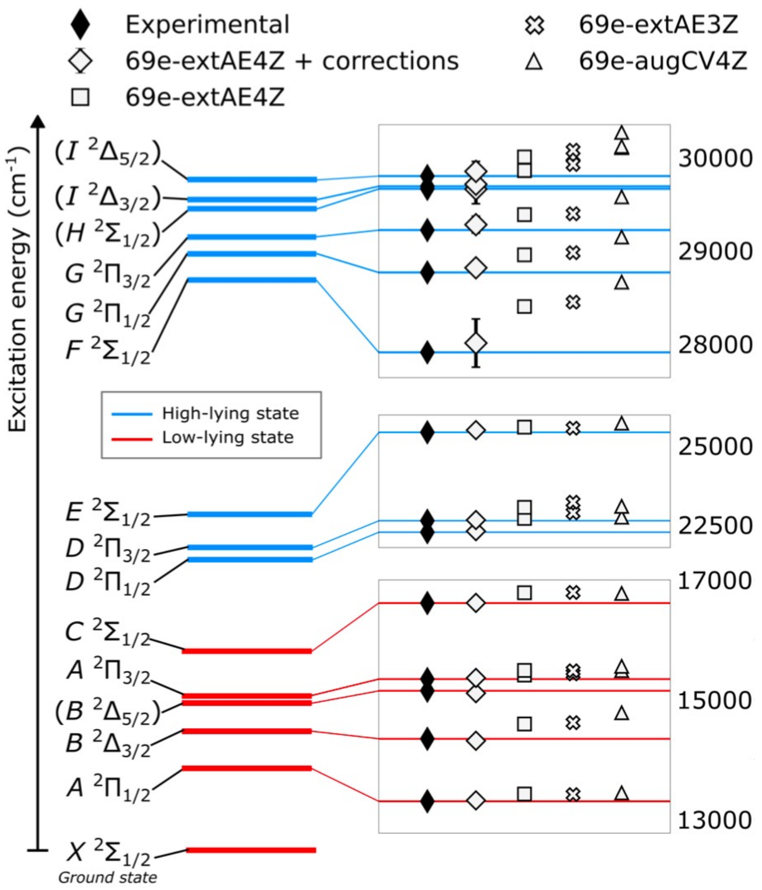
Laser cooling [1]



[1] Udrescu et al., Research Square 10.21203/rs.3.r 2648482/v1 accepted in Nat. Phys. (2023)
[2] Athanasakis-Kaklamanakis *et al.*, arXiv 2308.14862 submitted to PRL (2023)
[3] Athanasakis-Kaklamanakis *et al.*, arXiV 2403.09336 submitted to PRA (2024)
[4] Wilkins *et al.*, arXiV 2311.04121 submitted to Science (2024)

Excited states [2]

- agreement ≥ 99.64% (~12 meV)

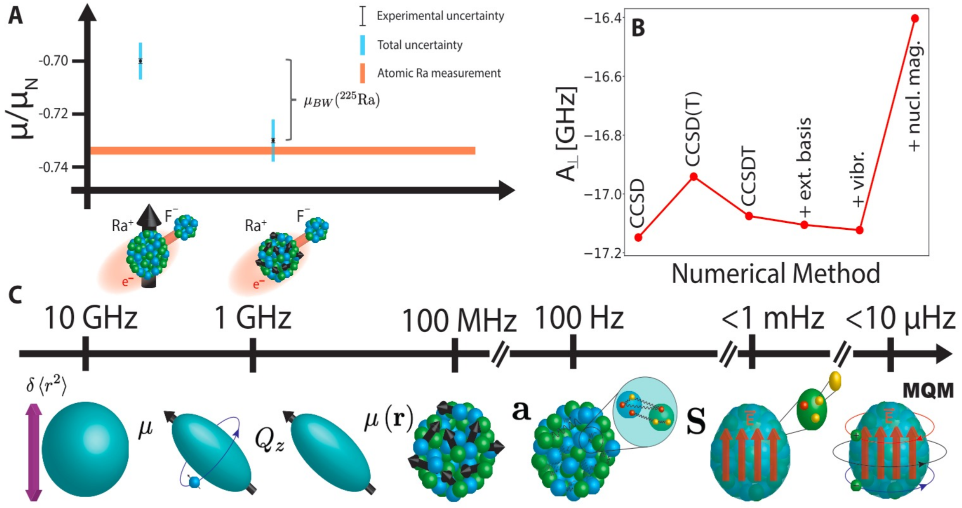


State lifetimes [3]

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

Nuclear magnetization effect [4]

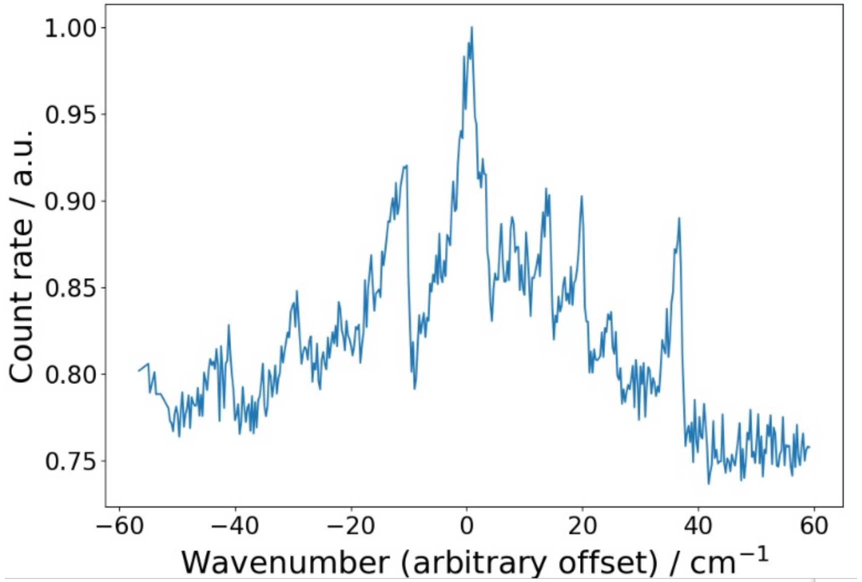
- $\mu(^{225}Ra)$



Multiple probes: AcF

Characterization

- $t_{1/2}$ and radioactivity challenging for offline setups
- 1 y proposal to beamtime



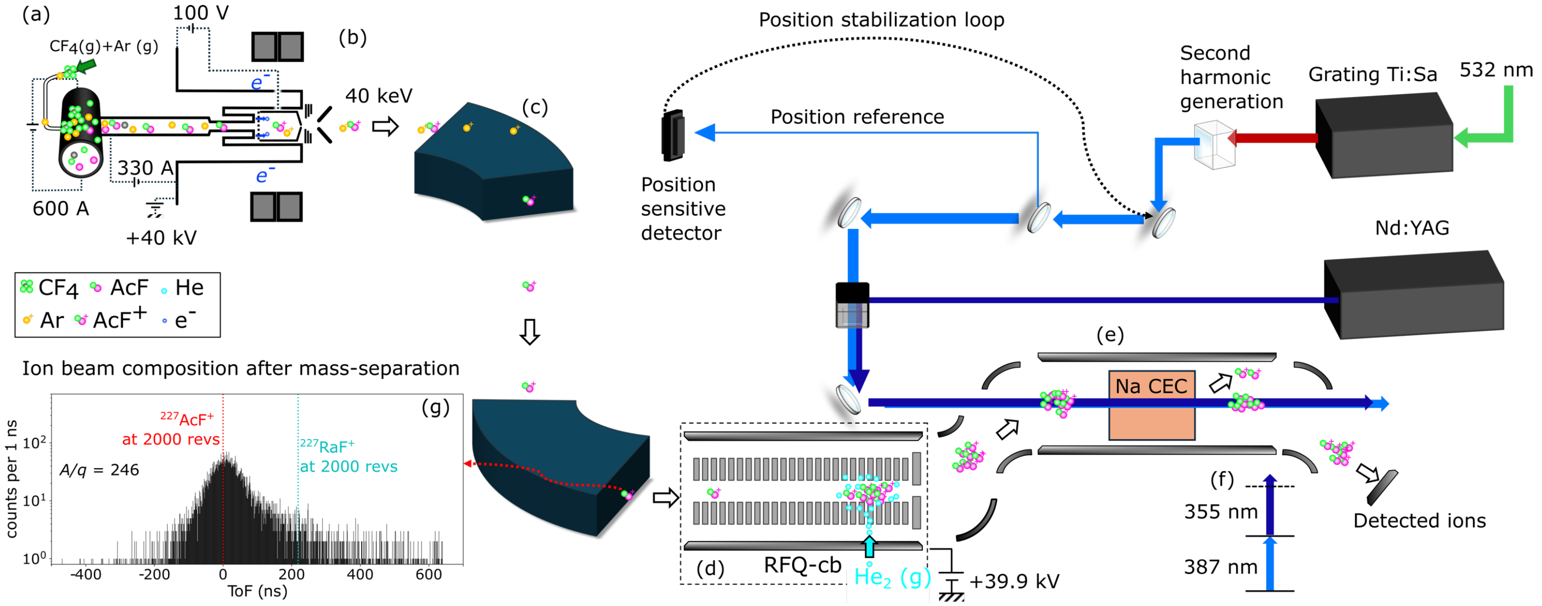
EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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]



[1] Athanasakis-Kaklamanakis, Wilkins, Au *et al.*, (2021) https://cds.cern.ch/record/2782407, INTC-P-615

[2] Athanasakis-Kaklamanakis and Au, (2023) *CERN EP newsletter*

[3] Athanasakis-Kaklamanakis, Au, Kyuberis, Zülch, Wibowo, Skripnikov, Reilly, Lalanne *et al., in preparation* 2024

Towards precision experiments at RIB facilities

Preparation of molecule

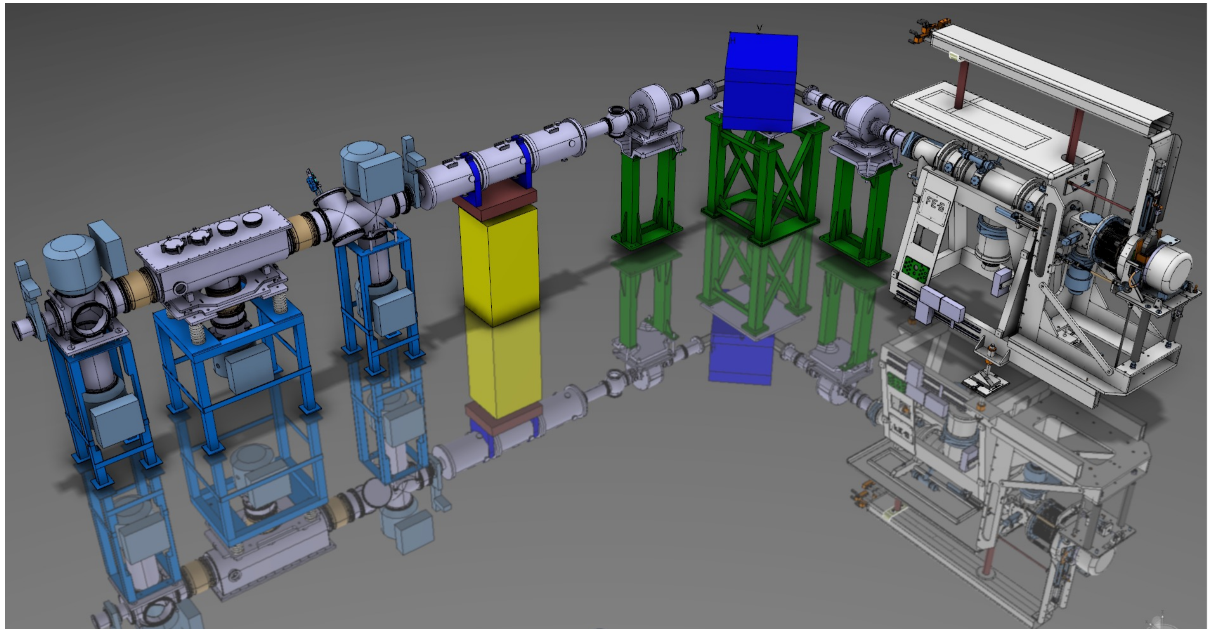
- Maximizing polarizability, inherent sensitivity
- Low-temperature molecular formation
- Control and selectivity of chemical reactions

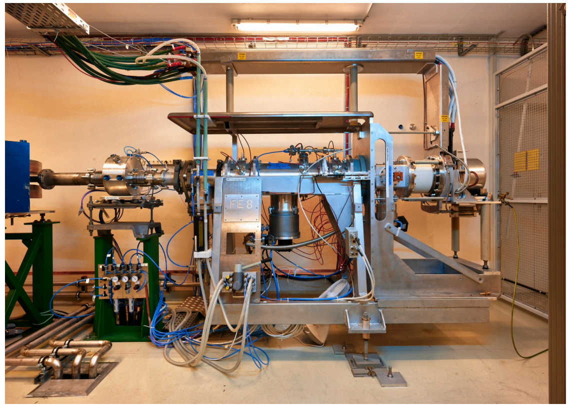
Preparation of "science state"

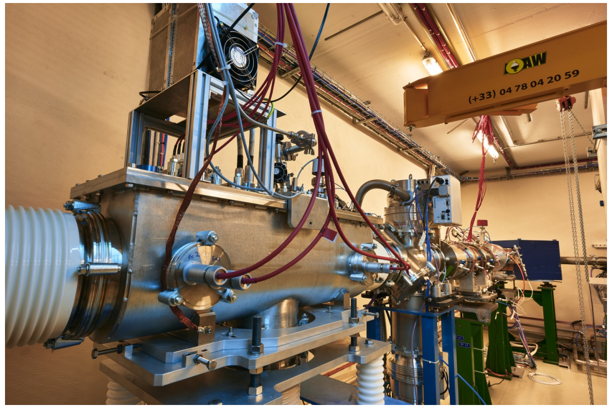
- Deceleration from extraction energy
- Cooling
- Noise – E, B systematics

Proposal

- test feasibility of delivering cold, prepared ensembles for precision measurements
- initial upgrades to existing infrastructure





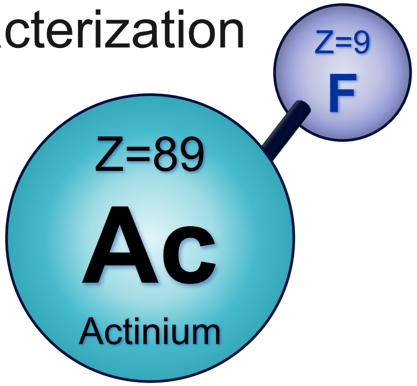


Photos: ISOLDE OFFLINE 2 © 2019-2022 CERN

Summary: radioactive molecules for PBC

1. Beam development at CERN-ISOLDE

- Provision of heavy, octupole deformed nuclei with sensitivity to symmetry violations
- Molecular formation techniques for delivery of sensitive, polarizable complementary probes for characterization



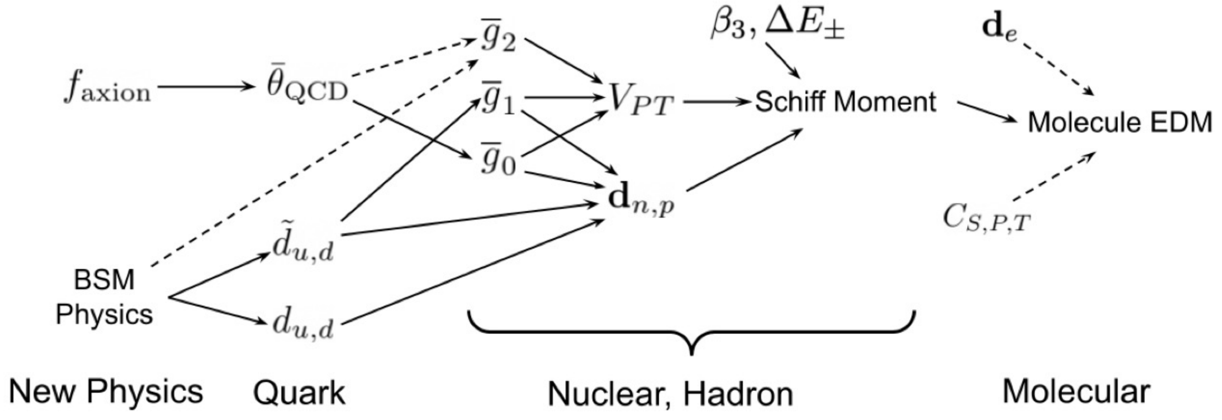
2. Radioactive species for offline precision experiments

- Collection and transportation of radioactive nuclei to precision experiments

Image published in EP Newsletter, CERN (2020)

3. Beam preparation towards precision for online experiments

- Feasibility of delivering cold, prepared ensembles for precision measurements



Opportunities for Fundamental Physics Research with Radioactive Molecules, arXiV 2302.02165 (2023), Rep. Prog. Phys. (2024) doi: 10.1088/1361-6633/ad1e39

Conclusion: a tipping point

Community: rapidly growing, particle, nuclear, AMO physics, experimental and theory

- “In the next 10 years gains in sensitivity by several orders of magnitude over current bounds are possible and even likely for electrons, nucleons, atoms and molecules, with the very real chance of discovery.” – Snowmass contribution, 2021
- INT Program INT-24 (March 2024) https://www.int.washington.edu/programs-and-workshops/24-1
- New Opportunities for Fundamental Physics Research with Radioactive Molecules, Virtual Meeting (2021)

Given the low relative cost of these experiments in terms of both funding ($\lesssim$ \$10 M, and often $\lesssim$1 M) and personnel (typically $\lesssim$ 10 people), pursuing many simultaneously is feasible. However, advancing to the next generation will require increases in scale and complexity. Many of the new approaches discussed here require sustained R&D budgets, theory support, and access to facilities when working with exotic nuclei, continued over several experimental generations to fully realize their projected gains. The field is moving very rapidly and requires risk tolerance, but it has proven that it can deliver results from a variety of novel approaches.

European EDM community

- ECT*, Trento (March 2024)
- Outcome: Formation of Europe-wide matterEDM network where experiments with unstable nuclei will be a major aspect for future work

EDMs: complementary experiments and theory connections

Mar 4 – 8, 2024
ECT*
Europe/Rome timezone

Thank you!

Acknowledgements

J. Ballof, R. Berger, A. Borschevsky, A. Breier, K. Chrysalidis, R.P. de Groote, Ch.E. Düllmann, C. Fajardo-Zambrano, P. Fischer, K. Flanagan, R.F. Garcia Ruiz, K. Gaul, P.F. Giesel, S. Gilardoni, R. Heinke, S. Hoekstra, N. Hutzler, Á. Koszorus, A. Kyuberis, D. Lange, B.A. Marsh, G. Neyens, L. Nies, E. Reis, S. Rothe, A. Oleynichenko, M. P. Reiter, P. Schmidt-Wellenburg, C. Schweiger, L. Skripnikov, M. Tarbutt, S. Wilkins, W. Wojtaczka,
The CRIS collaboration, The ISOLDE collaboration

Radioactive molecules community: growing every day

LISA
LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

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

Funding request

Catalyst to solidify an active and rapidly growing community with expertise in fundamental symmetries, nuclear, atomic, molecular, and optical physics, experimentalists and theorists.

Beam development

- 1 postdoc, 2-3 years

Feasibility and precision techniques

- 1 postdoc, 2-3 years
- Collaboration with leaders in the European precision EDM community
  - S. Hoekstra (Precision Frontier, University of Groningen)
  - M. Tarbutt (Centre for Cold Matter, Imperial College London)
  - M. P. Reiter (The Univerity of Edinburgh)
  - P. Schmidt-Wellenburg (PSI)

Infrastructure and consumables

- ~200k

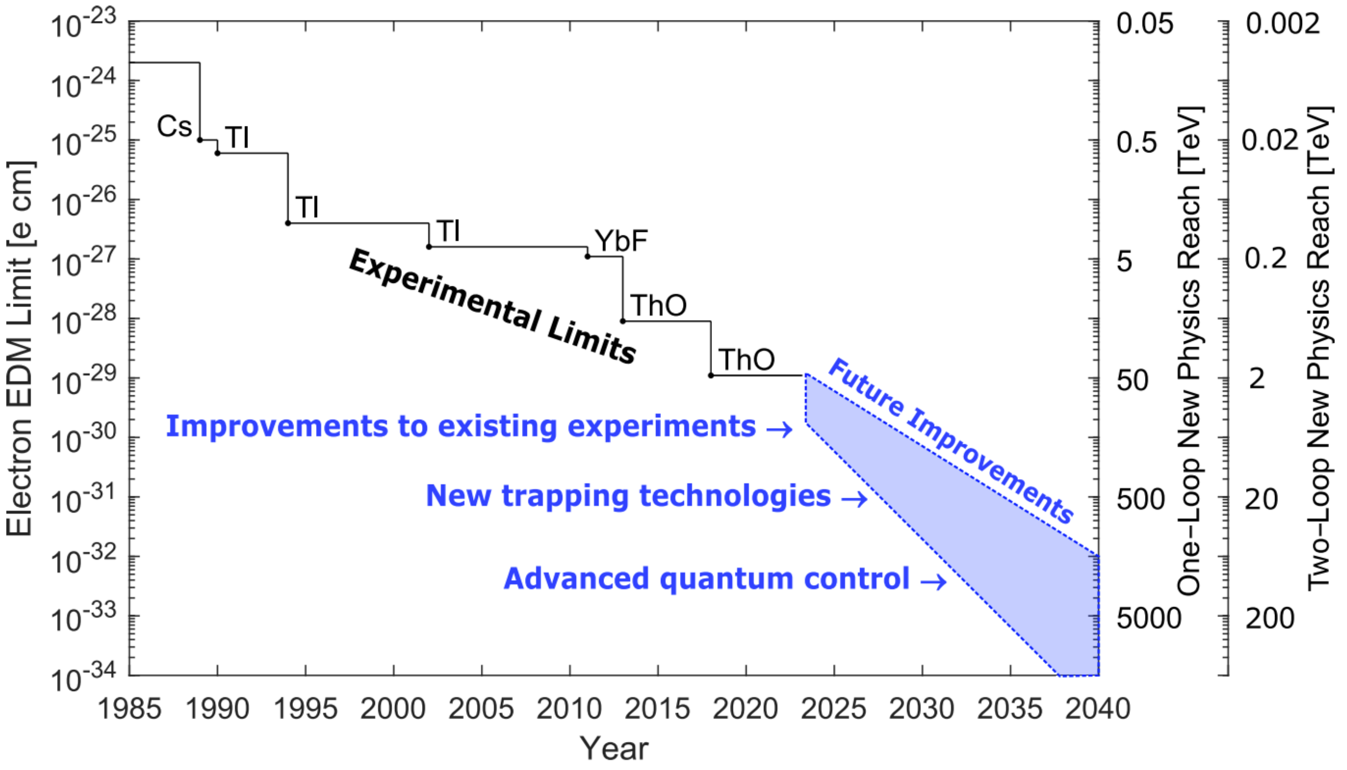
Existing and planned searches

Published:

- eEDM: Cs, Tl, YbF (Imperial) [1], ThO (ACME) [2,3], HfF+ (JILA) [4]
- Schiff, MQM: ^{129}Xe, ^{199}Hg, ^{225}Ra [5], ^{171}Yb [6]

Planned / under development:

- ^{225}Ra, ^{223}Ra molecules, FrAg, $^{221/210}$Fr, RIKEN, ^{229}Pa, PaF^{3+} MSU/FRIB, $^{225/227}$AcF, ^{229}Th, ^{175}Lu, Old Dominion, Grau , ^{181}Ta, UNLV, Zhou



[1] Nature 473, 493 (2011)
[2] Science 343, 269 (2014)
[3] Nature 562, 355 (2018)
[4] Science 381, 46 (2023)
[5] PRC 94, 025501 (2016)
[6] PRL 129, 083001 (2020)

Material developments

Gas injection

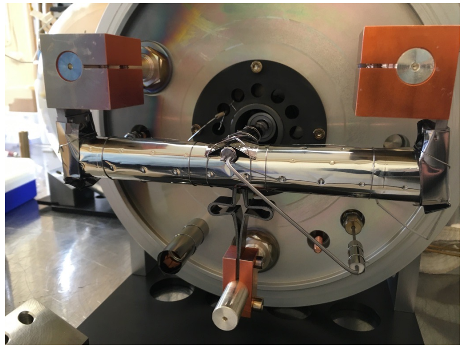
- Reactive/corrosive gases

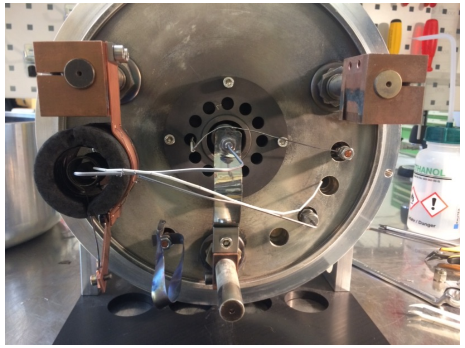
Reactants

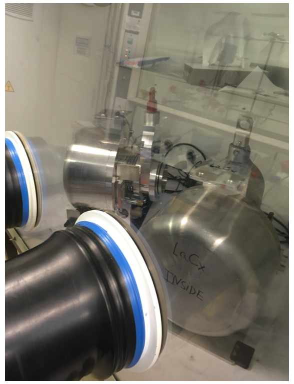
- Mass markers

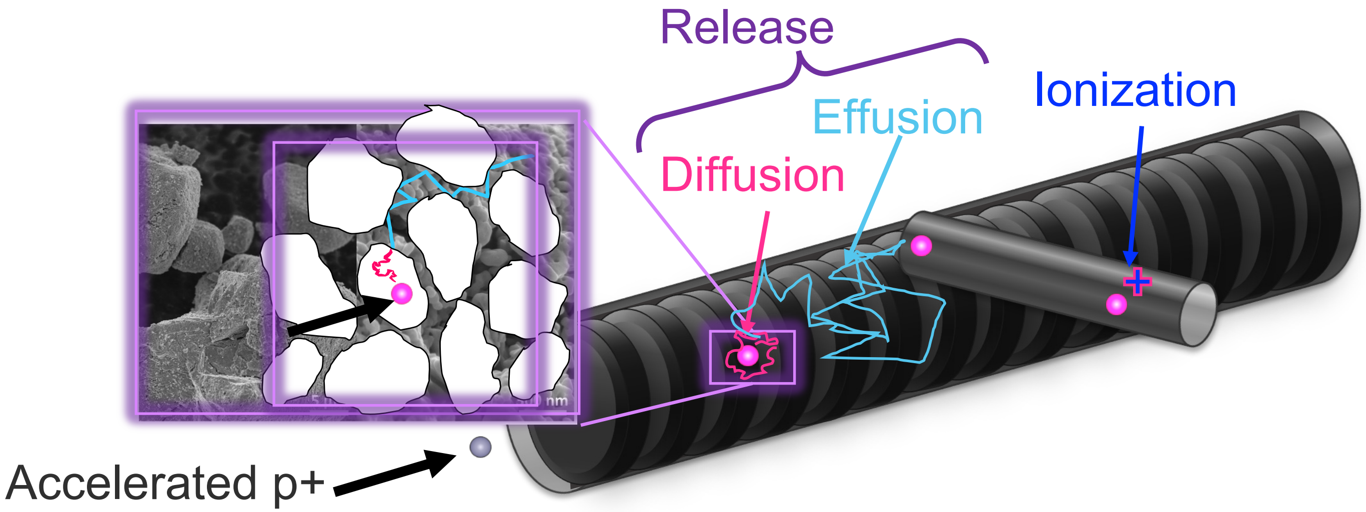
Target materials

- 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 [%]

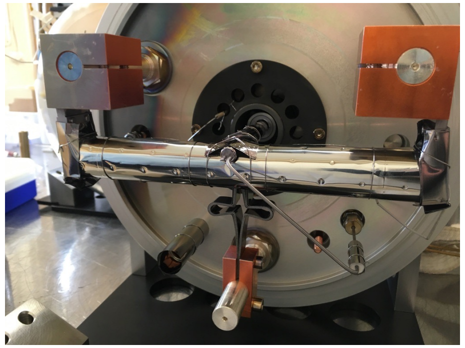
$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}\varepsilon_{is}\varepsilon_{ext}\varepsilon_{sep}\varepsilon_{trans}$$

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

Material developments

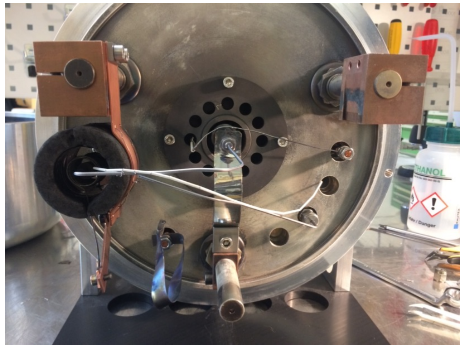
Gas injection

- Reactive/corrosive gases



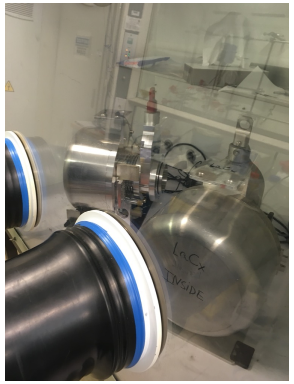
Reactants

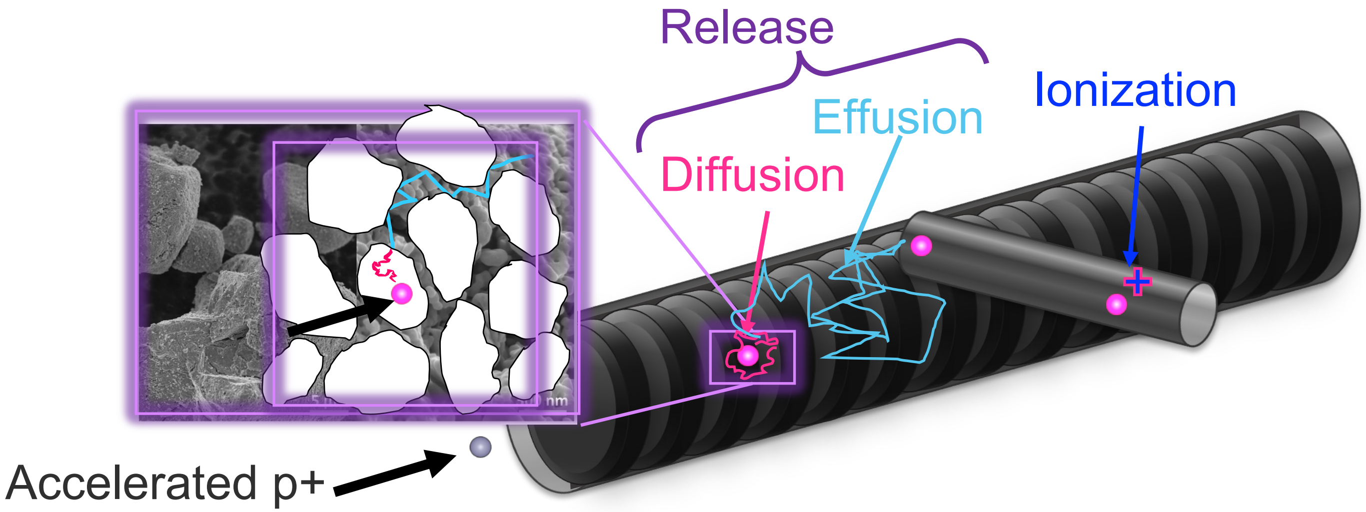
- Mass markers



Target materials

- Particle size
- Open porosity





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

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

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

Material developments

Gas injection

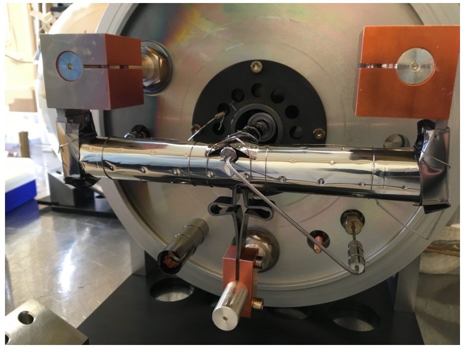
- Reactive/corrosive gases

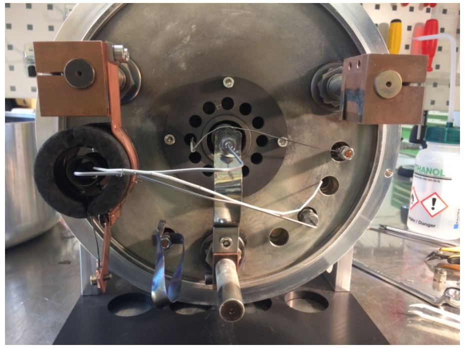
Reactants

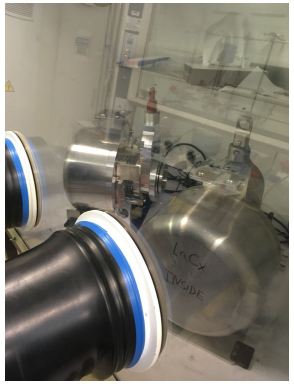
- Mass markers

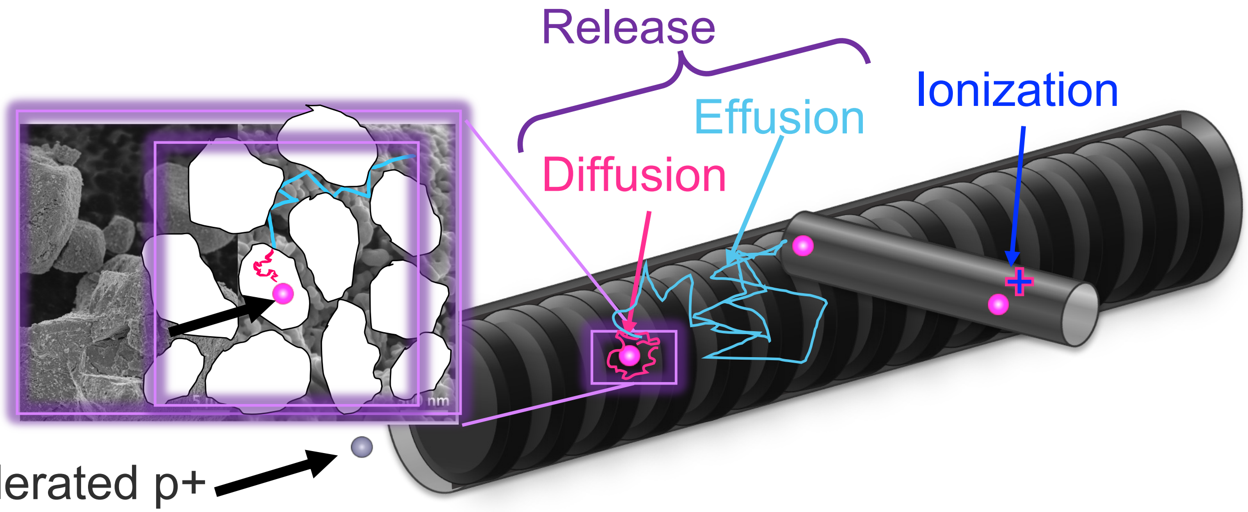
Target materials

- Particle size
- Open porosity









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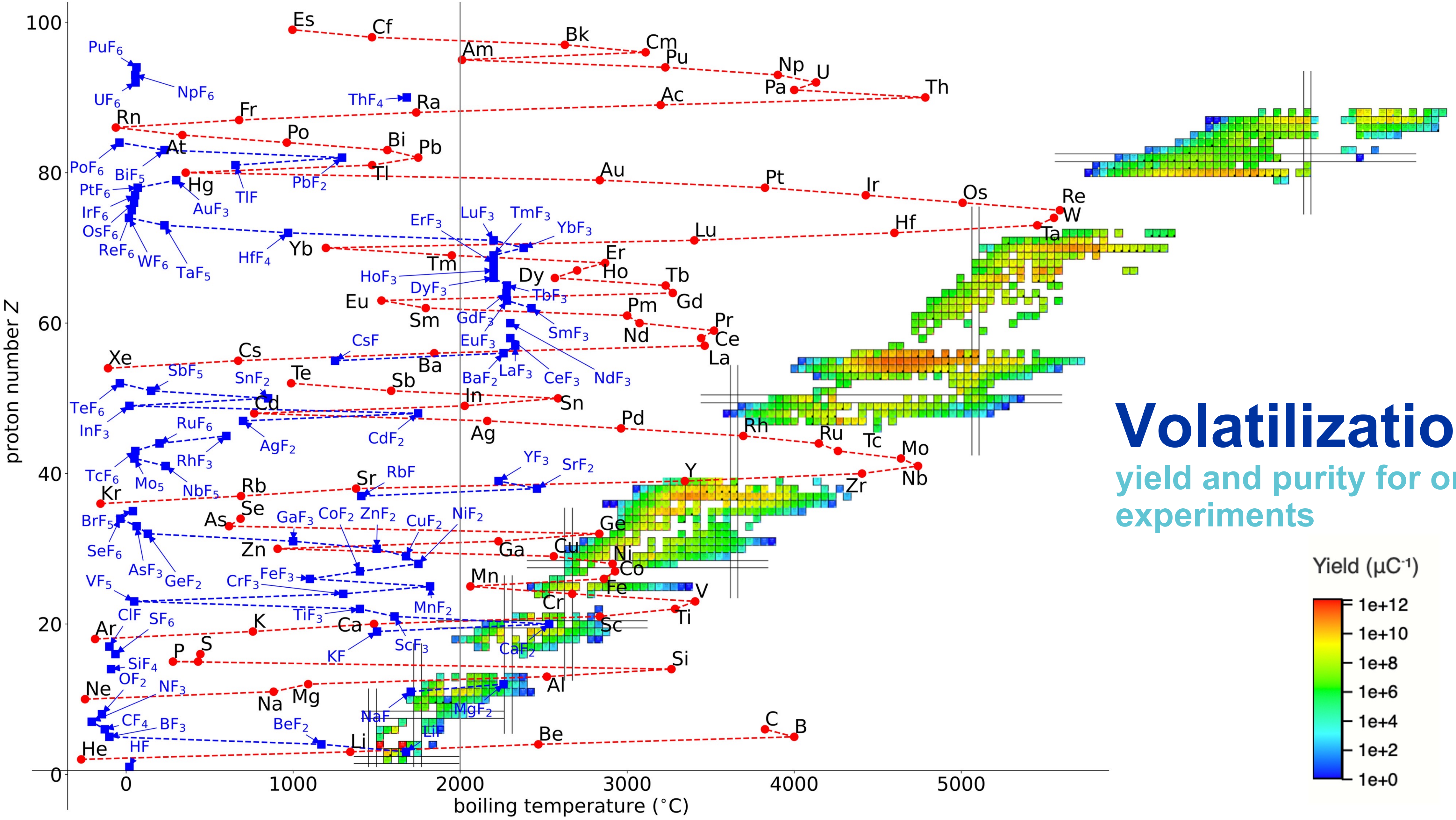
$$\mu = \frac{\pi^2 D}{G^2}$$

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

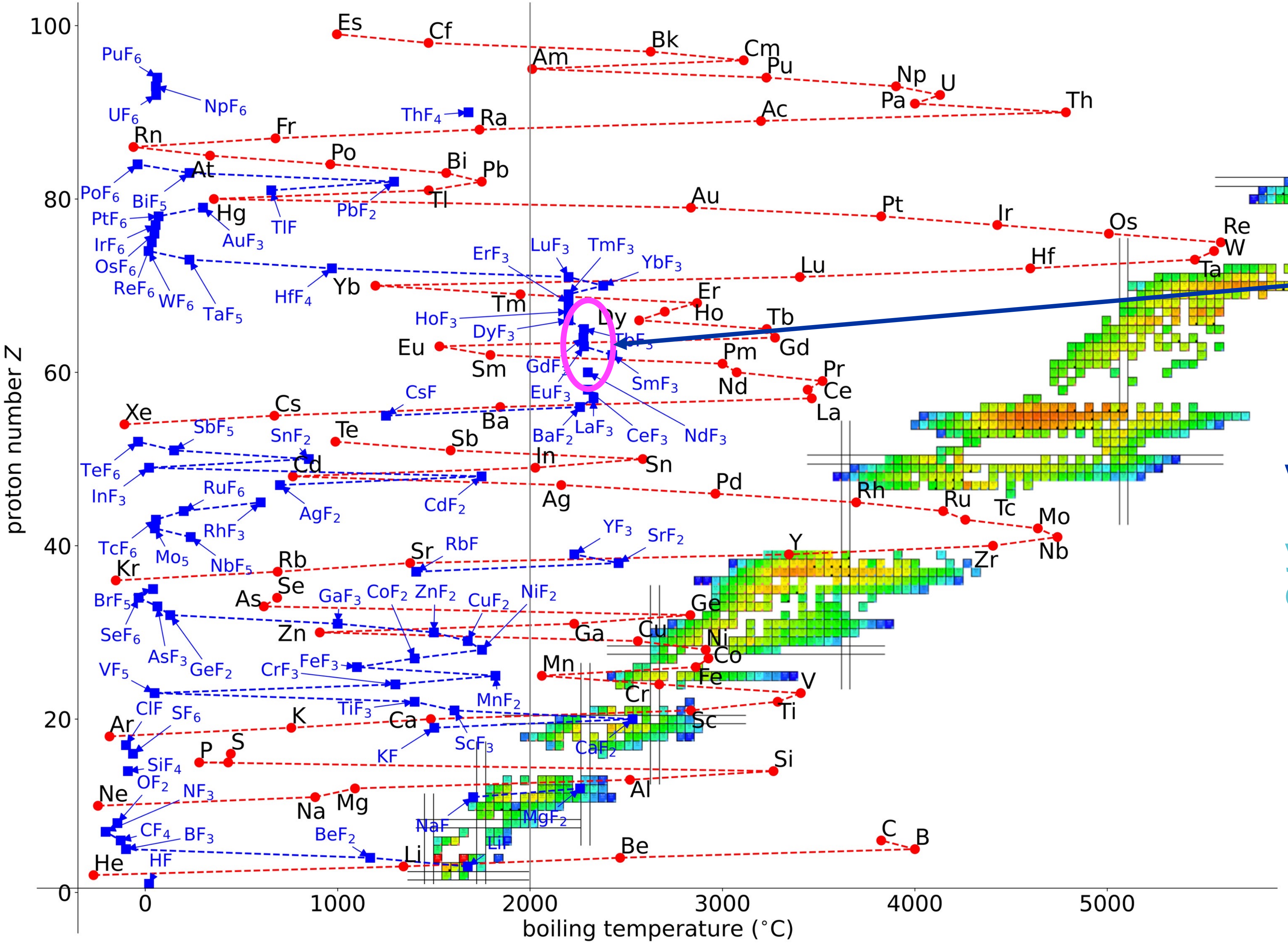
Increased $\varepsilon_{\text{diff}}$ ⟷ Increased sintering and grain growth

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



Volatilization

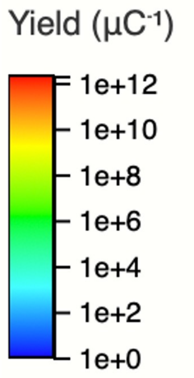
yield and purity for online experiments

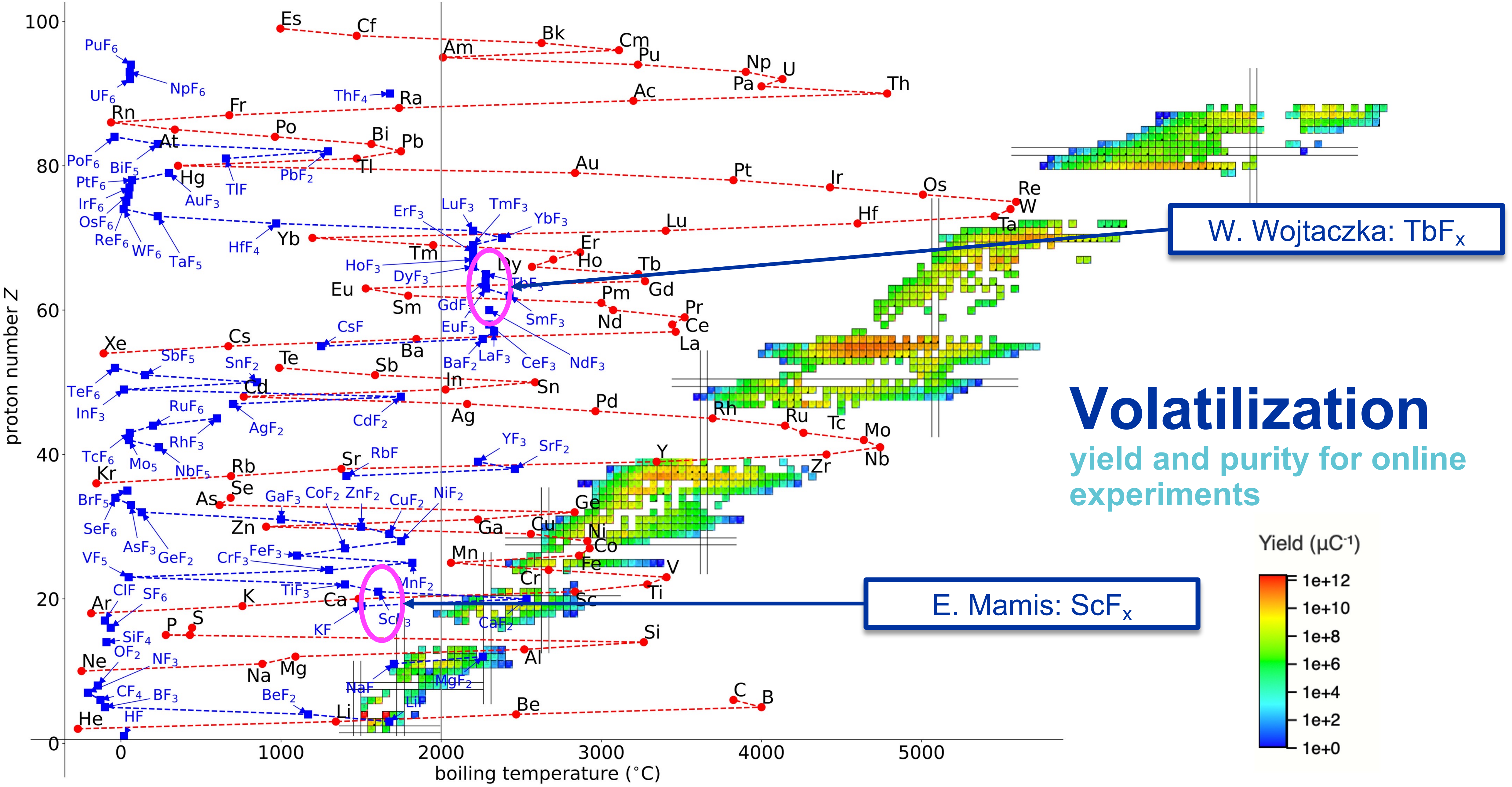


W. Wojtaczka: TbF_x

Volatilization

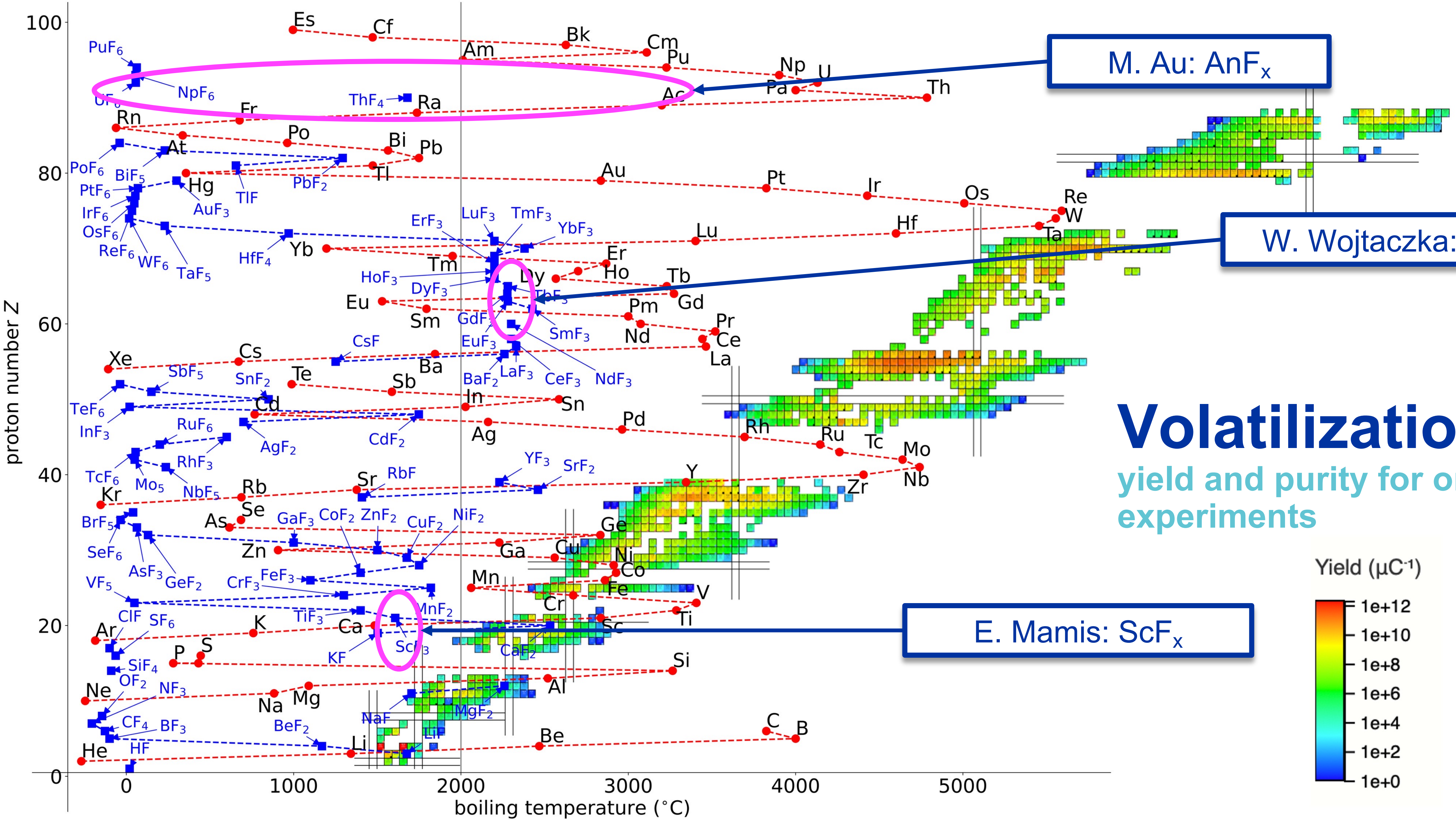
yield and purity for online experiments





Volatilization

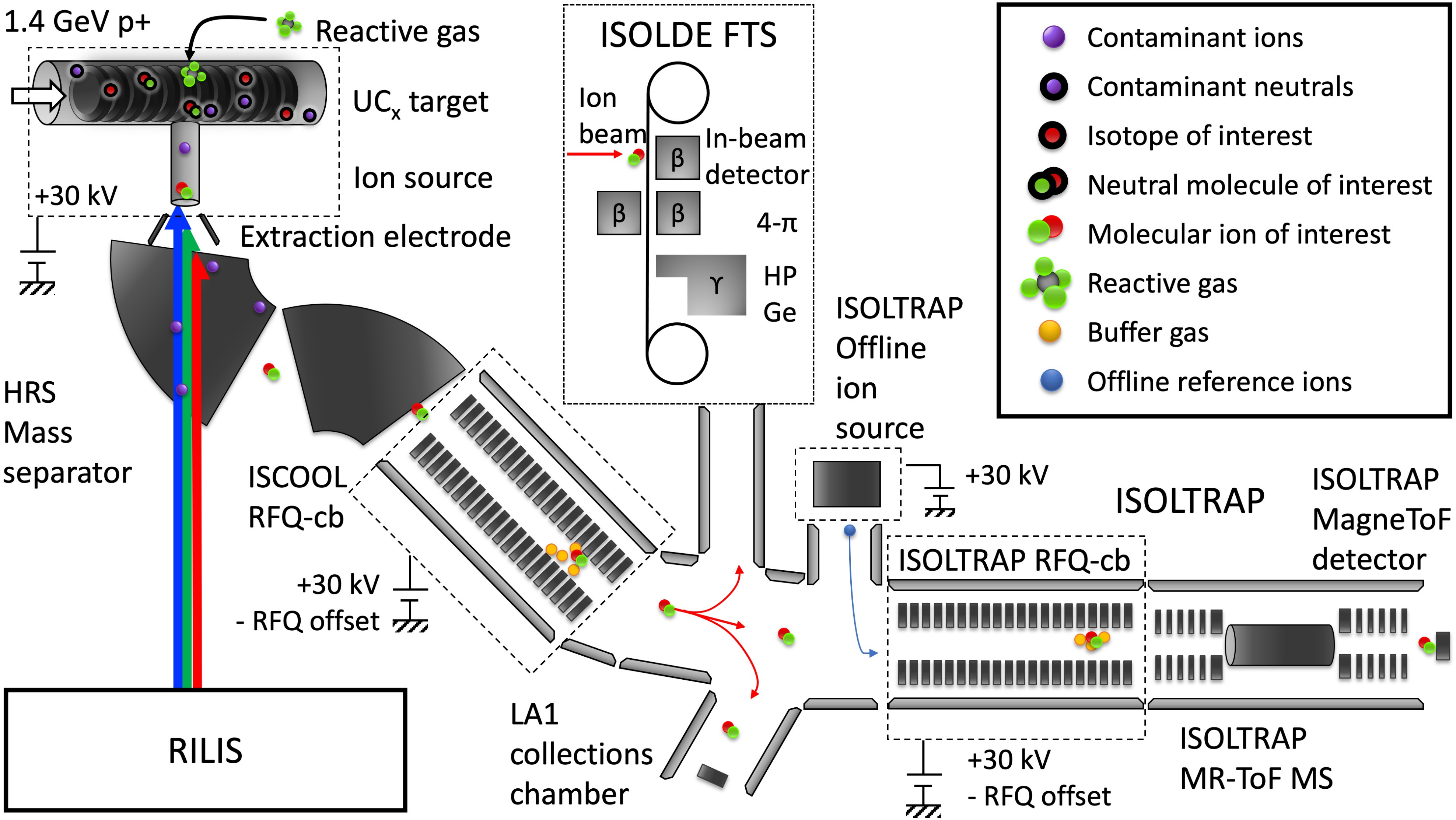
yield and purity for online experiments



Volatilization

yield and purity for online experiments

Detection and identification



Molecular structure

$$E = E_{el} + E_{vib} + E_{rot} + E_{hf}$$

Electronic

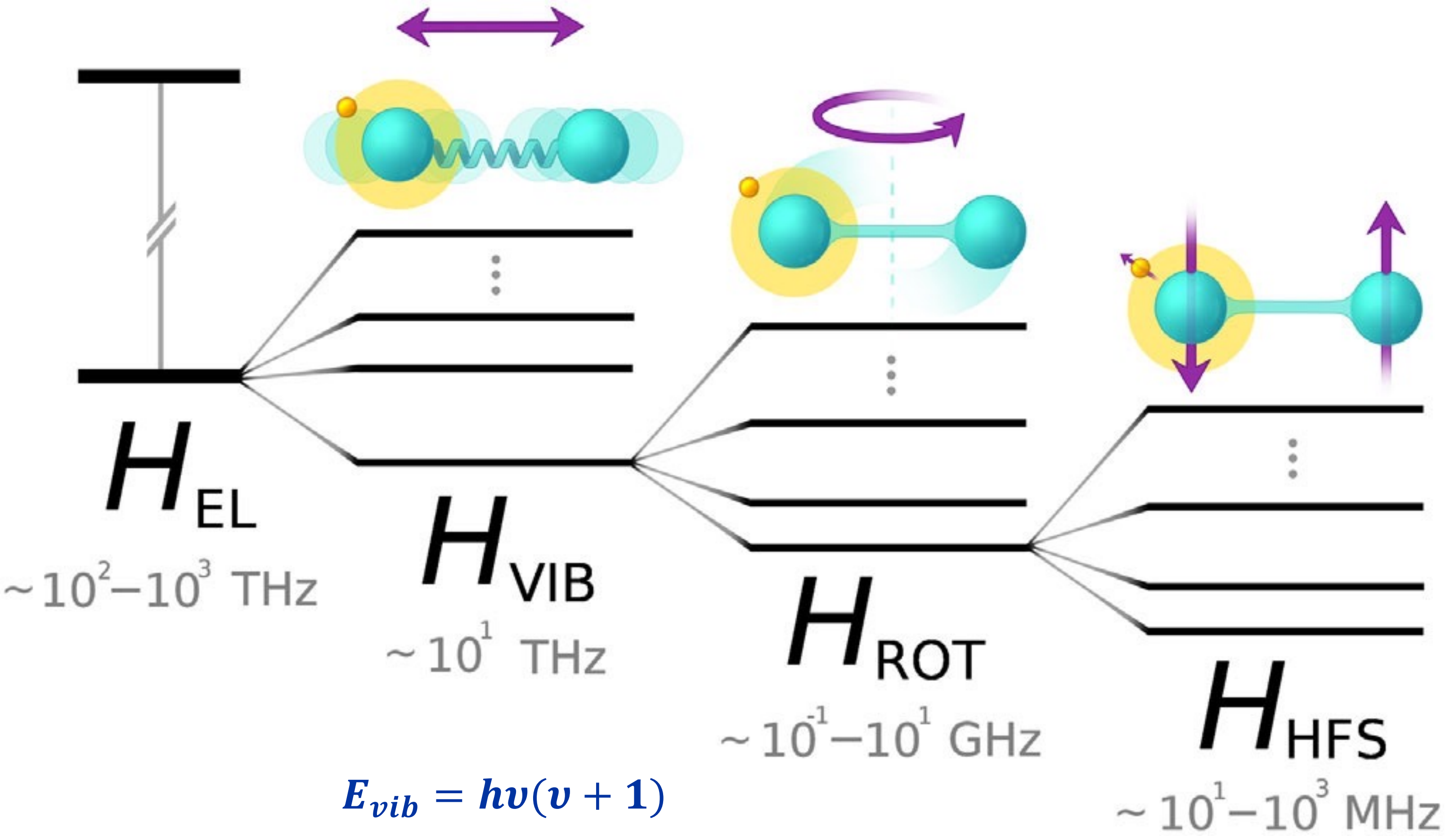
- $> 5000\ cm^{-1}$

Vibrational

- $300 - 3000\ cm^{-1}$

Rotational

- $1 - 100\ cm^{-1}$



$$E_{vib} = h\nu(v + 1)$$

$$E_{rot} = hcBJ(J + 1)$$

Figure published in [1]

[1] Athanasakis-Kaklamanakis *et al.*, PRX **13** 011015 (2023)

AcF spectroscopy

Experimental [1]

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

Nuclear theory

- *previous values from scaling factors*
- $S_{int} \leftrightarrow Q_0^3$ [2]
- DFT: $S_{int}(^{227}Ac) = 37.1(16)\ e\ fm^3$, $=1.4\ S_{int}(^{225}Ra) = 26.6(19)\ e\ fm^3$ [3]

Molecular theory

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

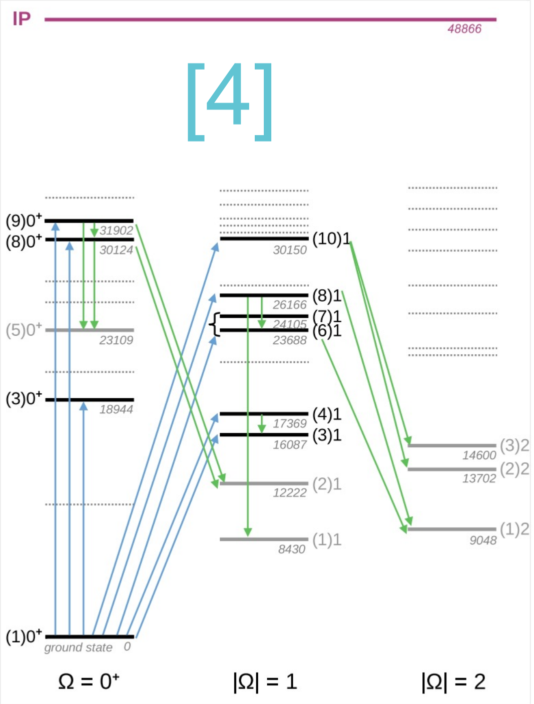


FIG. 3. The strongest transitions (blue arrows) from the $X(1)0^+$ ground state of AcF and the strongest transitions for stimulated emission (green arrows). Levels accessible with two-step excitations are shown with solid gray lines. Dotted lines depict electronic states that are hardly accessible from 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.

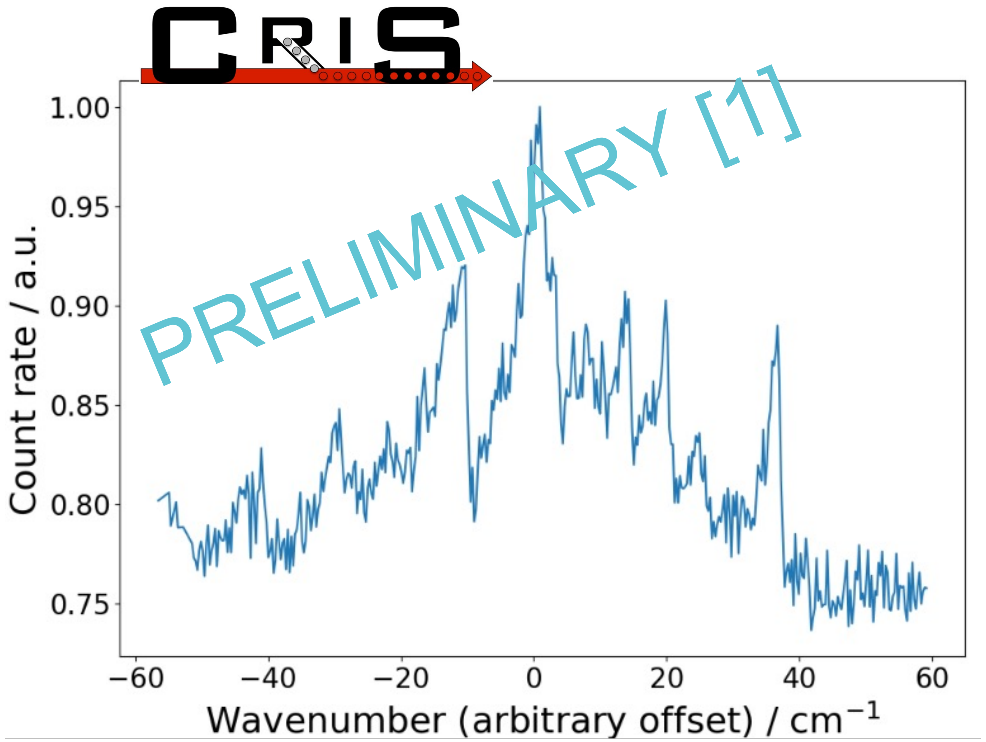


Table 2 Molecular constants X and $W_S^{(2)} = 6X/r^{sp}$ $(e/a_B^4,\ a_B = 1$ Bohr) calculated at different levels of theory, given in square brackets

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)

Beam preparation

Molecular formation

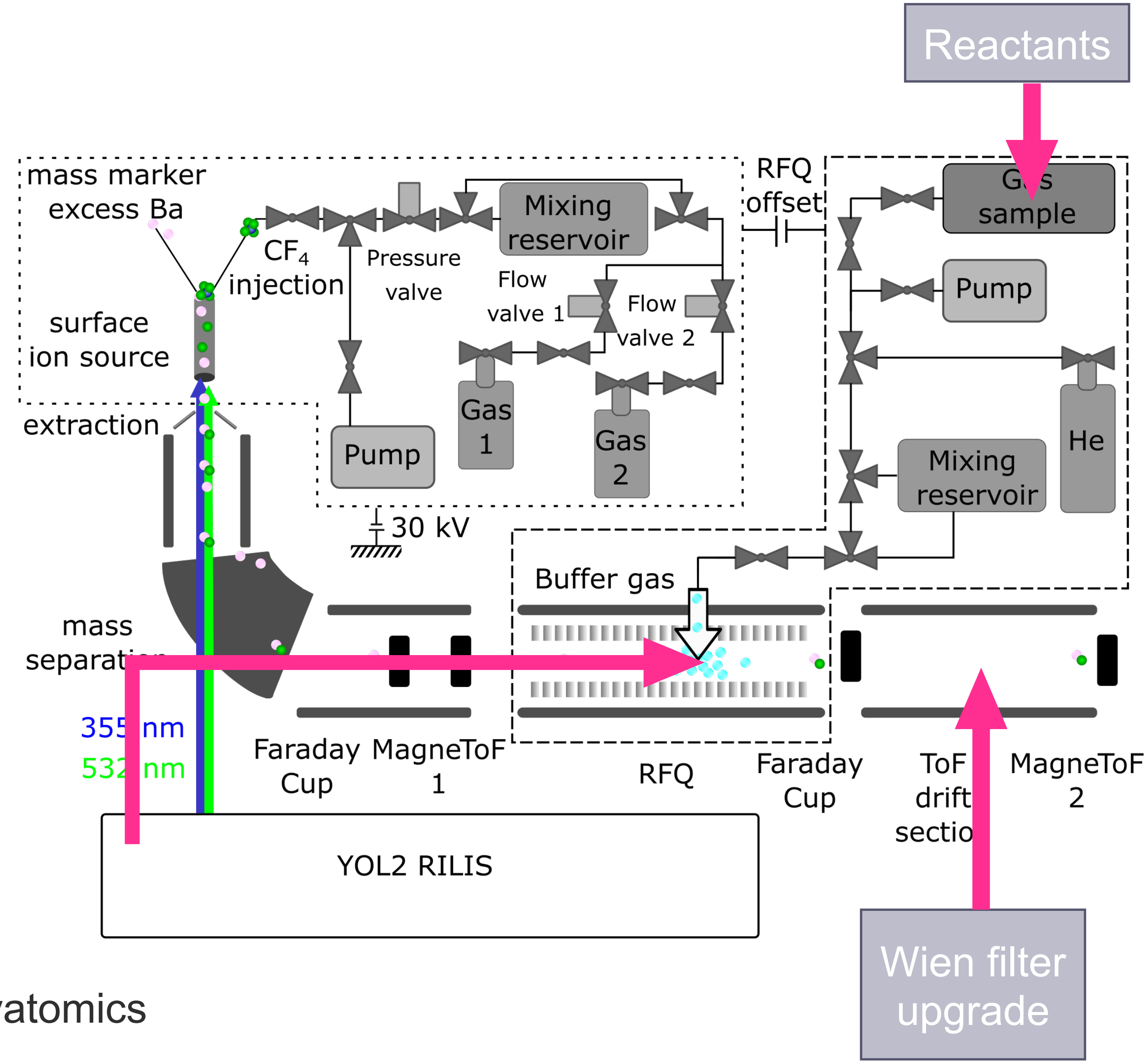
- Ion source developments:
  - FEBIAD, photocathode, LIST
- RFQ gas mixing and injection tests (+TRIUMF)
- Implantation/ablation (+KUL)

Laser setup

- YOL2 laser lab development (+KUL)
- New end of beamline
- Laser path to RFQ

Scheme development / spectroscopy

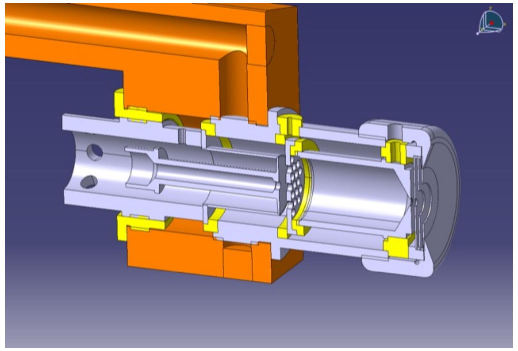
- In-source: Offline LIST – fluorides, oxides
- In-trap: pending molecular formation studies – polyatomics



Ion source developments

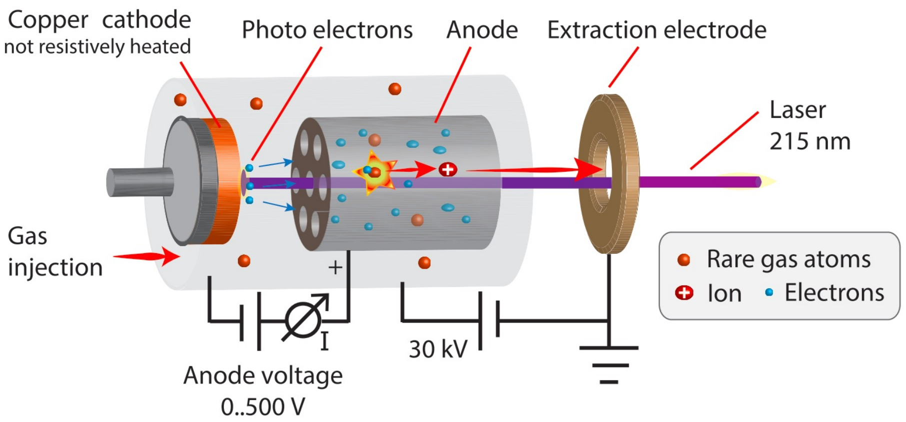
Molecular breakup and characterization studies

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



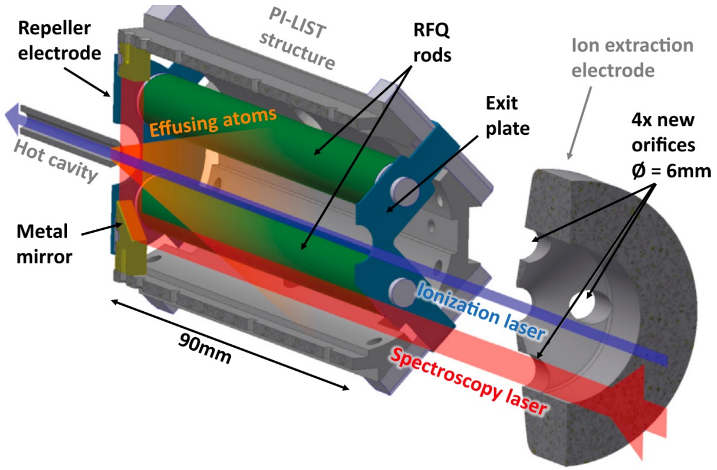
Photocathode ion sources [3]

- 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)