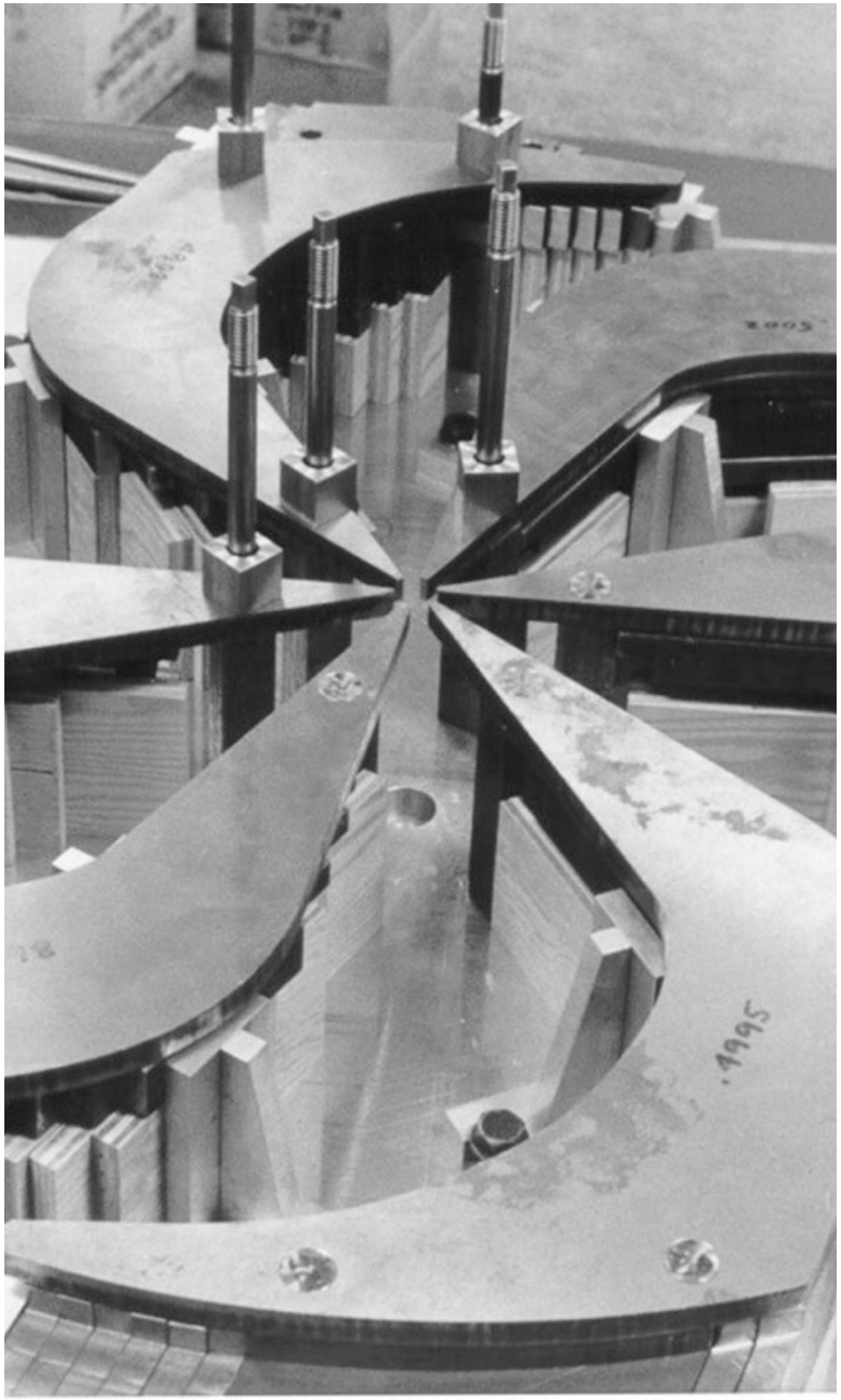
Radioactive Molecules
Novel Probes for New Physics



Stephan Malbrunot-Ettenauer
TRIUMF, University of Toronto

CAP conference 2024

Discovery, accelerated

‘Designer Molecules’

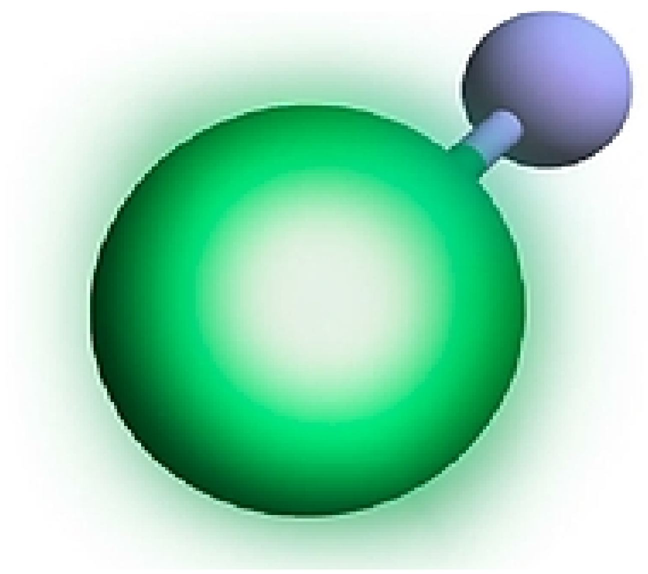
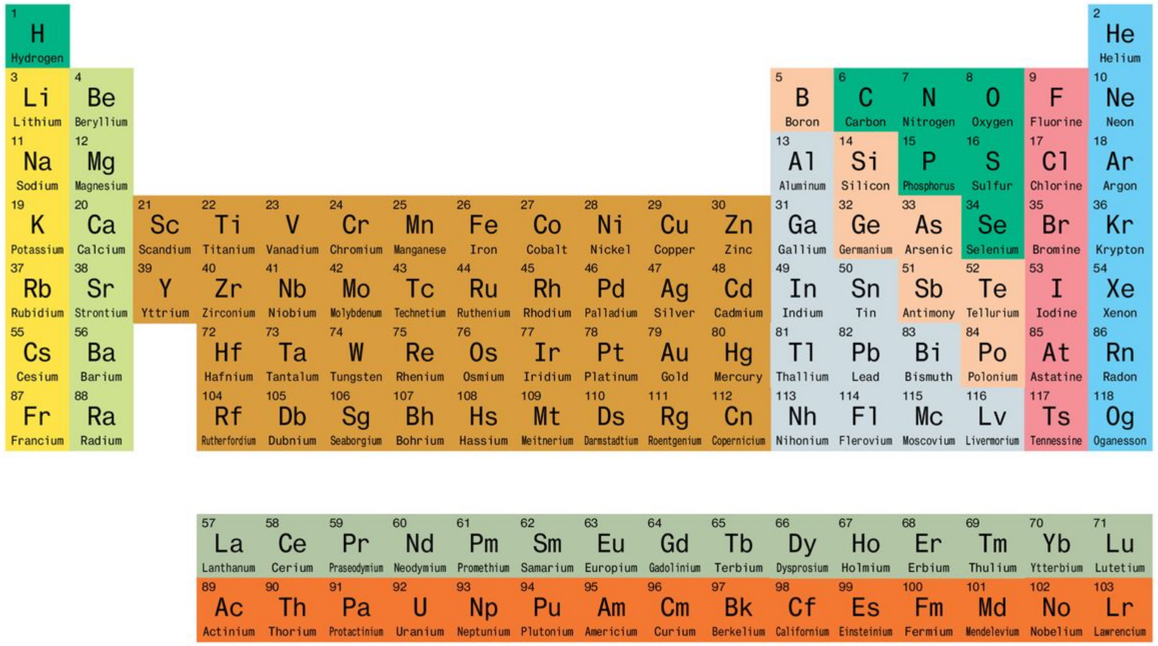


Table of Elements

80 chemical elements
(with stable nuclides)



‘Designer Molecules’

Table of Isotopes

252 stable

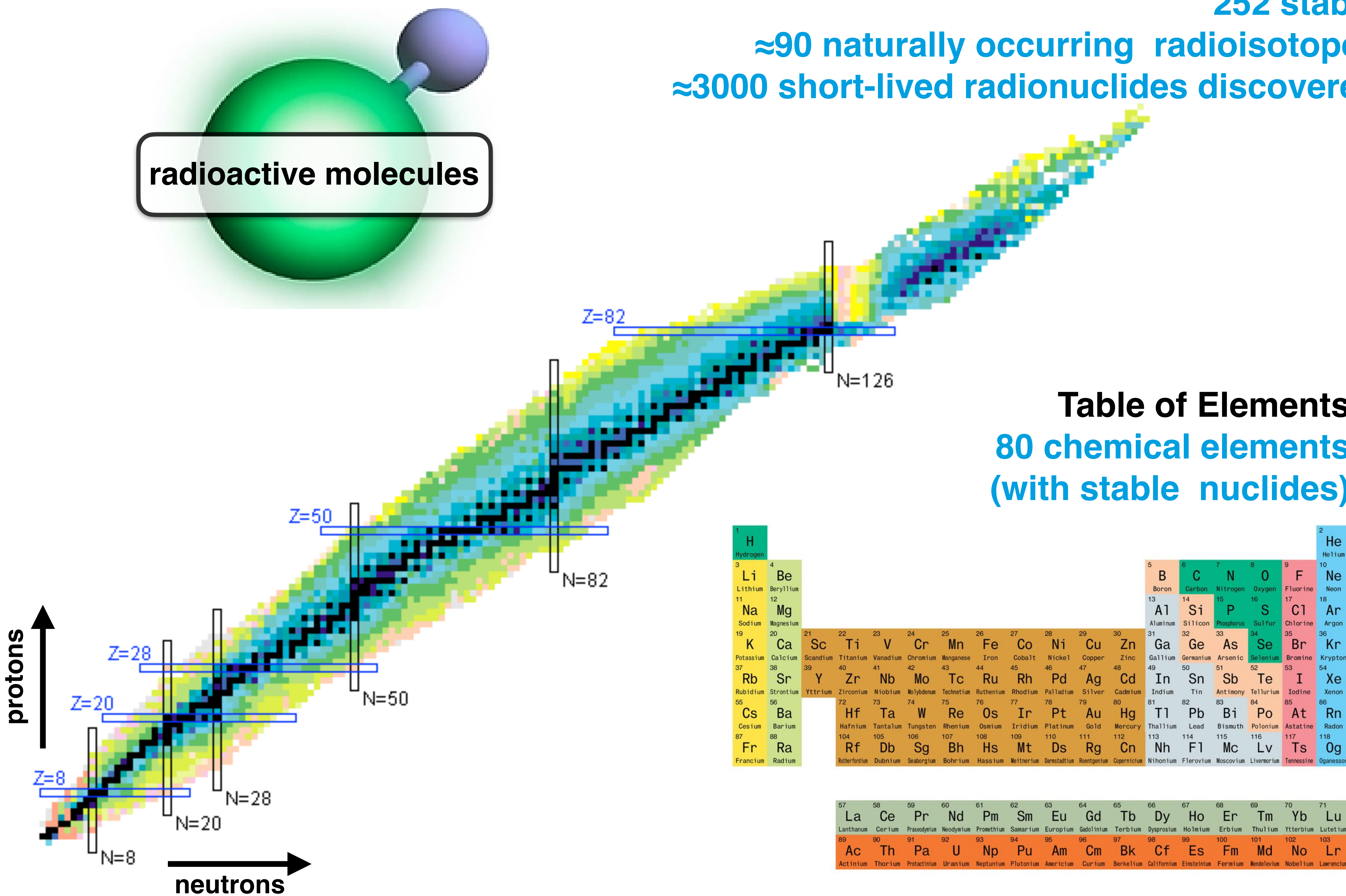
≈90 naturally occurring radioisotopes

≈3000 short-lived radionuclides discovered

2

Table of Elements

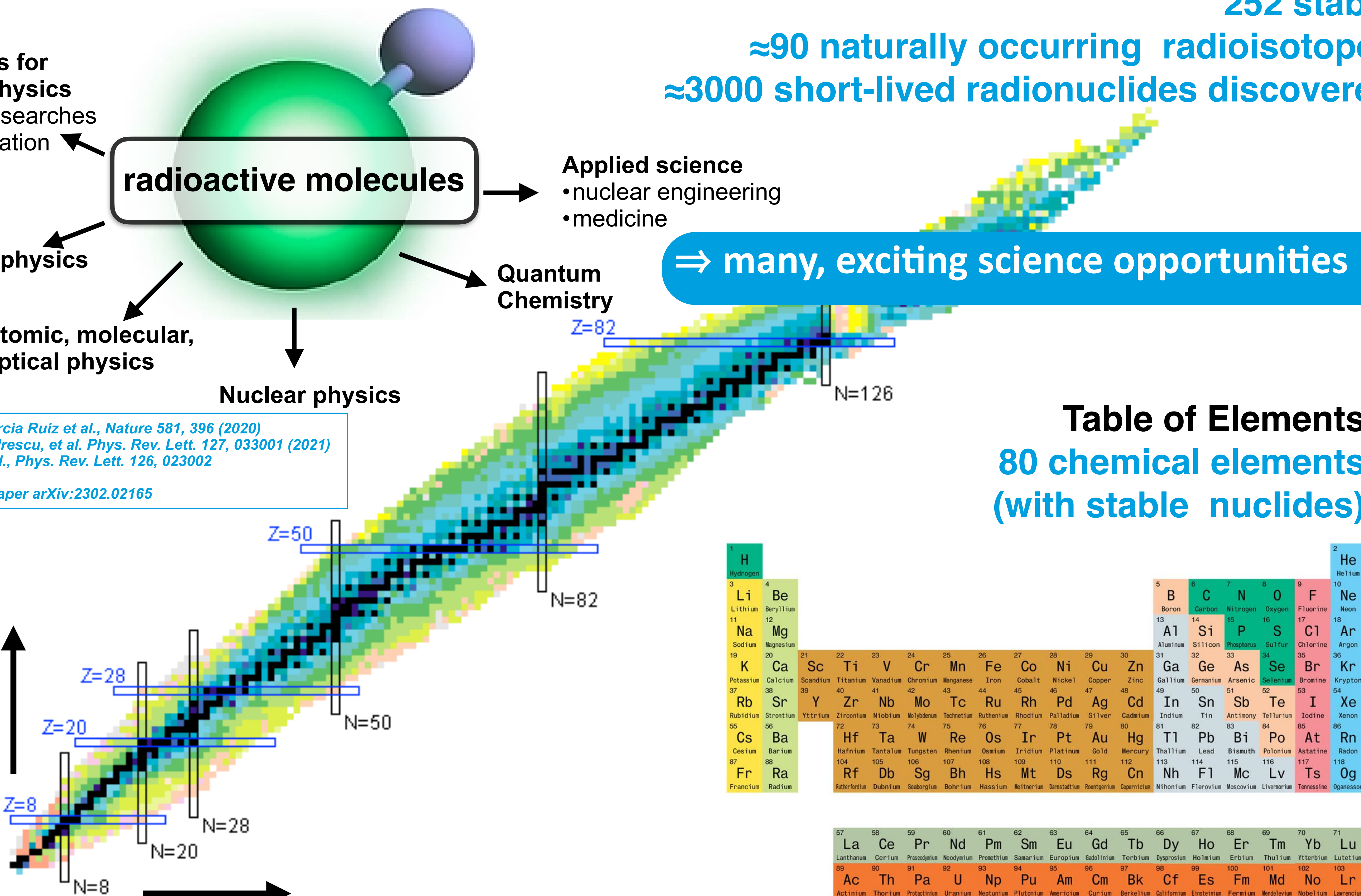
80 chemical elements (with stable nuclides)



‘Designer Molecules’

Table of Isotopes
252 stable
≈90 naturally occurring radioisotopes
≈3000 short-lived radionuclides discovered

2



probes for new physics
- EDM searches
- P violation

Applied science
- nuclear engineering
- medicine

Astrophysics

Quantum Chemistry

Atomic, molecular, optical physics

Nuclear physics

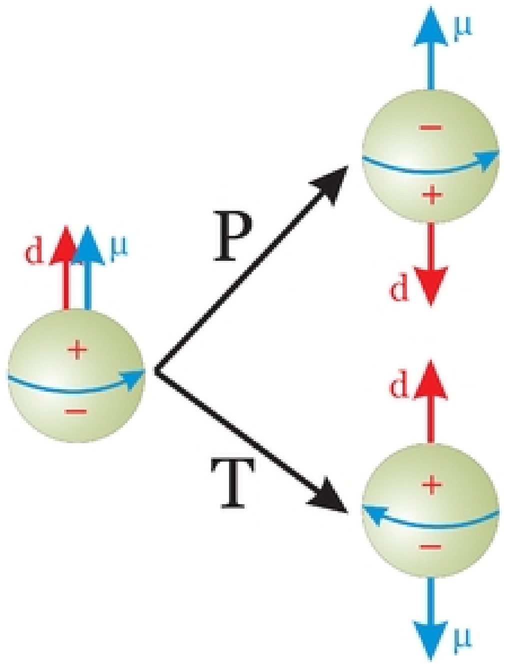
⇒ many, exciting science opportunities

R. F. Garcia Ruiz et al., Nature 581, 396 (2020)
S. M. Udrescu, et al. Phys. Rev. Lett. 127, 033001 (2021)
Fan et al., Phys. Rev. Lett. 126, 023002

White Paper arXiv:2302.02165

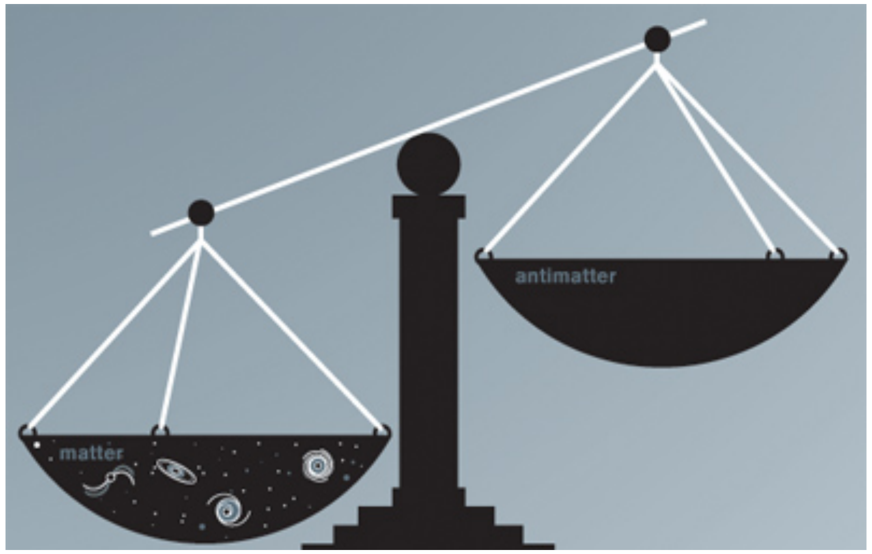
Table of Elements
80 chemical elements
(with stable nuclides)

Permanent Electric Dipole Moments (EDM)

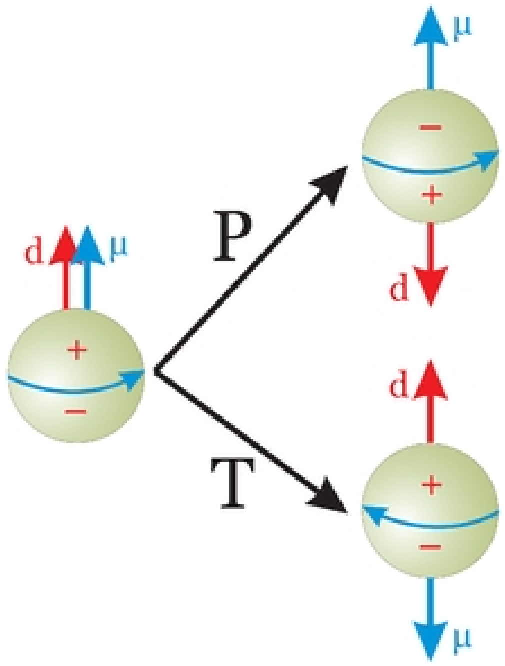


- local separation of the electric charge along a particle’s spin axis
- implies time-reversal (T) violation ⇒ violation of CP symmetry

matter-antimatter asymmetry in the universe



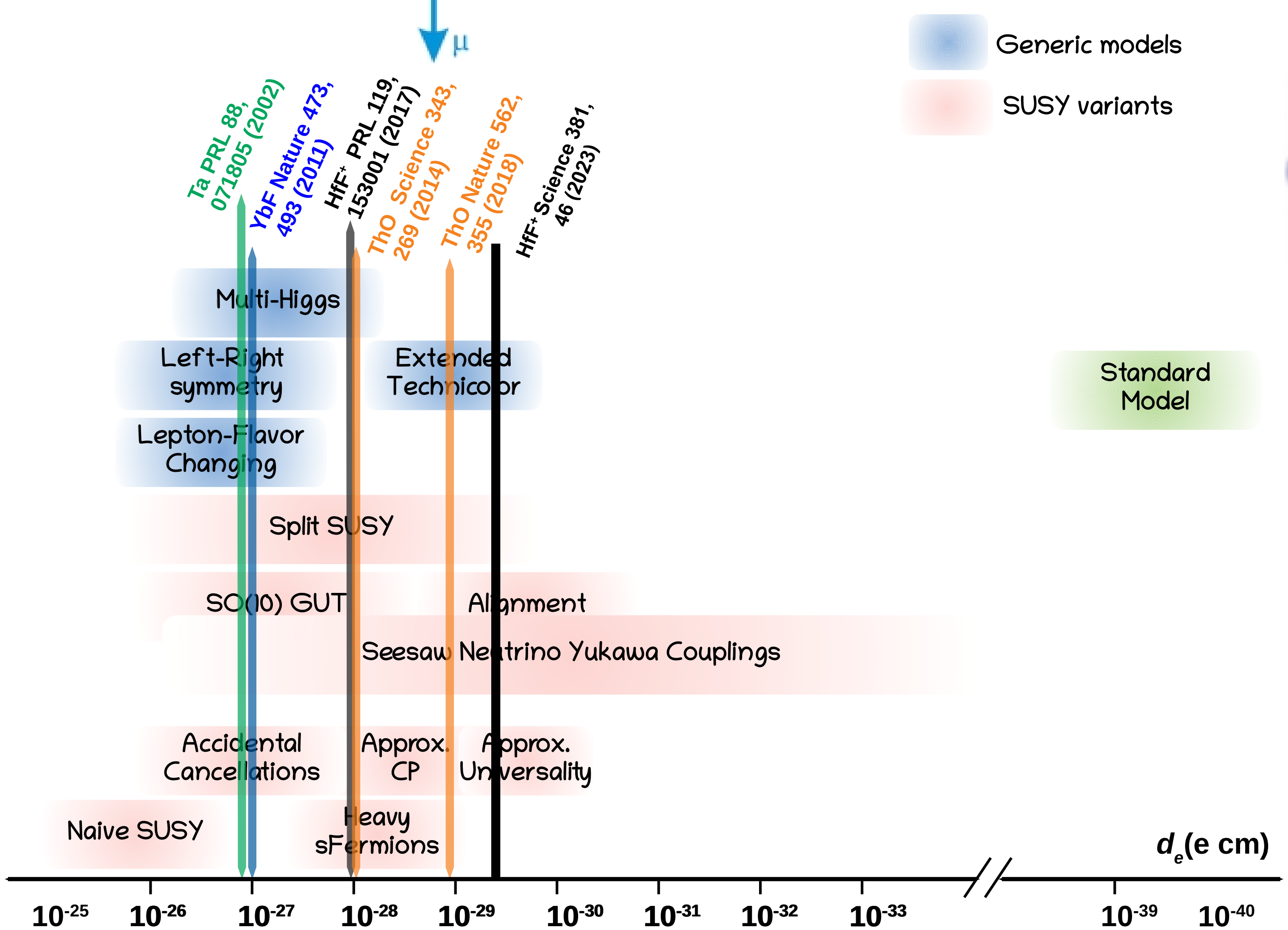
Permanent Electric Dipole Moments (EDM)



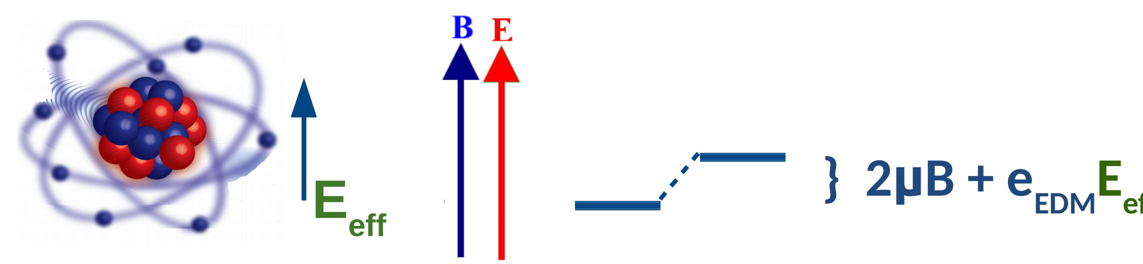
- local separation of the electric charge along a particle's spin axis
- implies time-reversal (T) violation ⇒ violation of CP symmetry

↕

matter-antimatter asymmetry in the universe



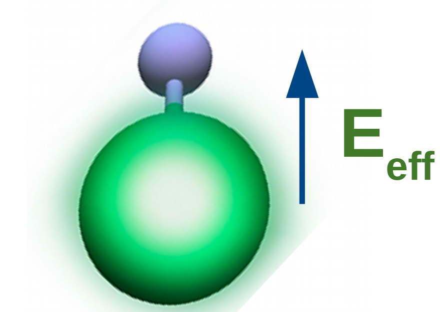
electron EDM



EDM sensitivity:

$$\sigma_d \sim \frac{1}{E_{\text{eff}}\,\tau\sqrt{\dot{N}T}}$$

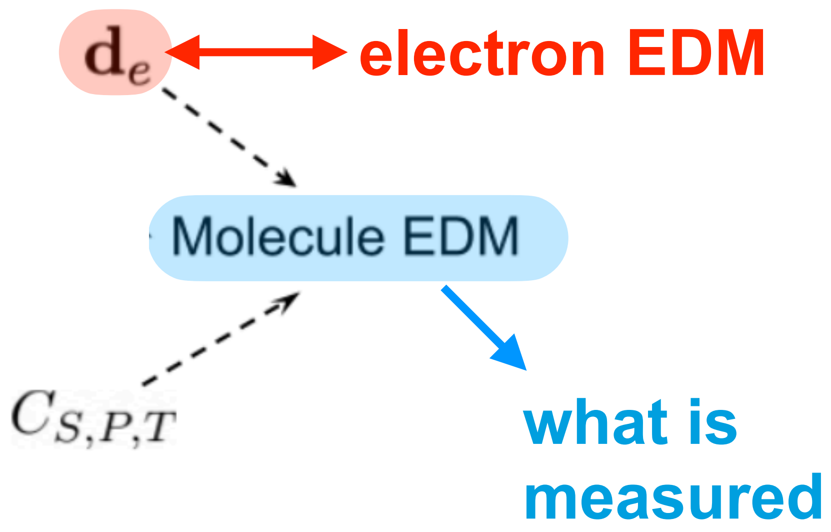
Advantage of molecules:



E_{ext} ~1 V/cm

E_{eff} ~80 GV/cm

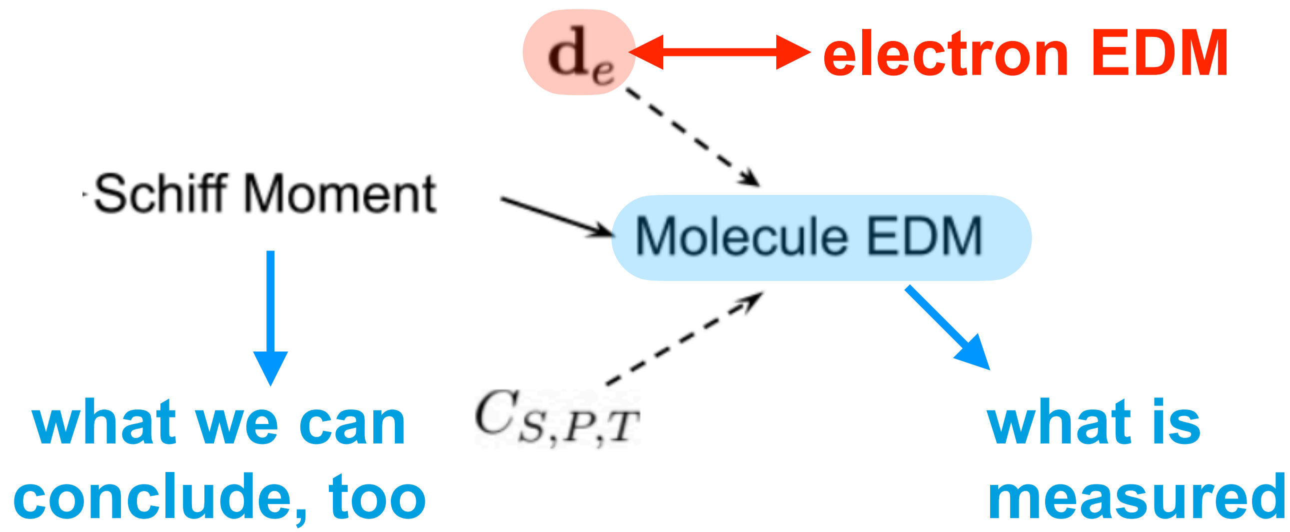
Complementarity of different probes



4

Molecular

Complementarity of different probes



4

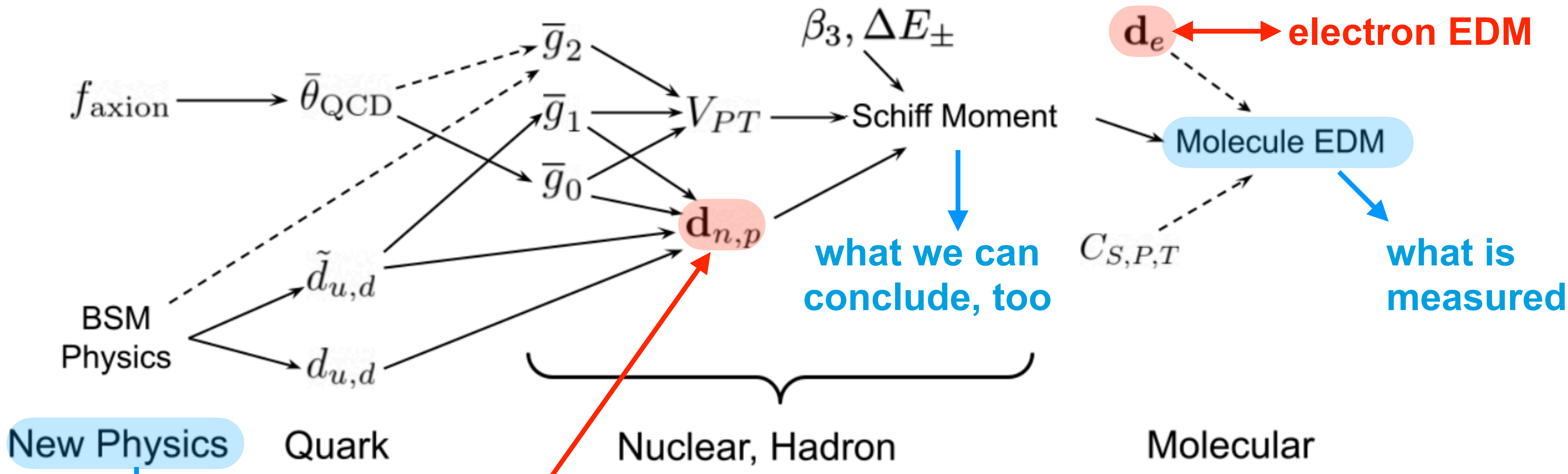
Molecular

enhancement factors for different mol. EDM contributions

State	Term Symbol	$W_S/\frac{e}{4\pi\epsilon_0 a_0^4}$	$W_\mathrm{d}/\frac{10^{24}\mathrm{Hz}}{e\mathrm{cm}}$	$W_\mathrm{s}/h\mathrm{kHz}$	$W_\mathrm{T}/h\mathrm{Hz}$	$W_\mathrm{M}/\frac{10^{33}h\mathrm{Hz}}{c\ e\mathrm{cm}^2}$
HfF^+	$^3\Delta_1$	$-15\,000$	6.4	23	-1200	0.57
ThO	$^3\Delta_1$	$-35\,000$	24	140	-3500	1.4
PaF^{3+}	$^2\Phi_{5/2}$	$-72\,000$	0.66	4.2	-6700	0.038

R. Berger et al.

Complementarity of different probes



what we are ultimately interested in

what neutron EDM experiments measure e.g.

enhancement factors for different mol. EDM contributions

State	Term Symbol	$W_S/\frac{e}{4\pi\epsilon_0 a_0^4}$	$W_\mathrm{d}/\frac{10^{24}\mathrm{Hz}}{e\mathrm{cm}}$	$W_\mathrm{s}/h\mathrm{kHz}$	$W_\mathrm{T}/h\mathrm{Hz}$	$W_\mathrm{M}/\frac{10^{33}h\mathrm{Hz}}{c\ e\mathrm{cm}^2}$
HfF^+	$^3\Delta_1$	−15 000	6.4	23	−1200	0.57
ThO	$^3\Delta_1$	−35 000	24	140	−3500	1.4
PaF^{3+}	$^2\Phi_{5/2}$	−72 000	0.66	4.2	−6700	0.038

R. Berger et al.

Schiff Theorem

nuclear dipole moment
shielded by electrons' dipole moment } nothing to detect

Assumptions:

- non-relativistic electrons
- a point nucleus the electrons

Schiff Theorem

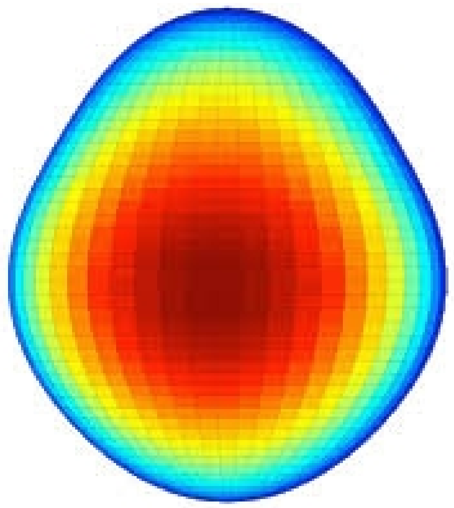
nuclear dipole moment
shielded by electrons' dipole moment } nothing to detect

Assumptions:

- non-relativistic electrons
- a point nucleus the electrons

↕

!! octupole deformation !!



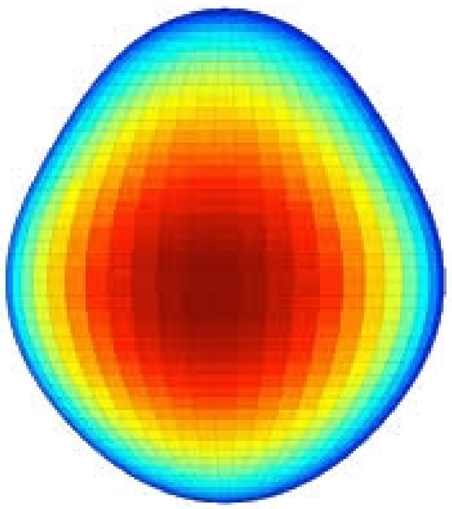
‘Designer Molecules’

… for searches for CP violation in atomic nuclei

^{199}Hg present ‘gold standard’ for limit on nuclear Schiff moment

$|d_{Hg}| < 7.4 \cdot 10^{-30}$ e cm (95% confidence limit)

$|S_{Hg}| < 3.1 \cdot 10^{-13}$ e fm^3 *B. Graner et al., Phys. Rev. Lett. 116, 161601 (2016)*



Enhancement factors in our approach:

- **octupole** deformed nuclide x 100-1,000
- in polar molecule x 1,000-10,000

} compared to ^{199}Hg

- in atom or ion trap x 1,000 compared to beam experiments

all known cases in radionuclides

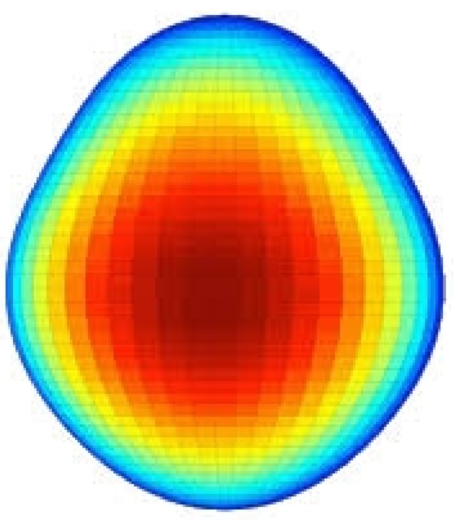
‘Designer Molecules’

… for searches for CP violation in atomic nuclei

^{199}Hg present ‘gold standard’ for limit on nuclear Schiff moment

$|d_{Hg}| < 7.4 \cdot 10^{-30}$ e cm (95% confidence limit)

$|S_{Hg}| < 3.1 \cdot 10^{-13}$ e fm^3 *B. Graner et al., Phys. Rev. Lett. 116, 161601 (2016)*



Enhancement factors in our approach:

- octupole deformed nuclide x 100-1,000 } compared to ^{199}Hg
- in polar molecule x 1,000-10,000 } compared to ^{199}Hg
- in atom or ion trap x 1,000 compared to beam experiments

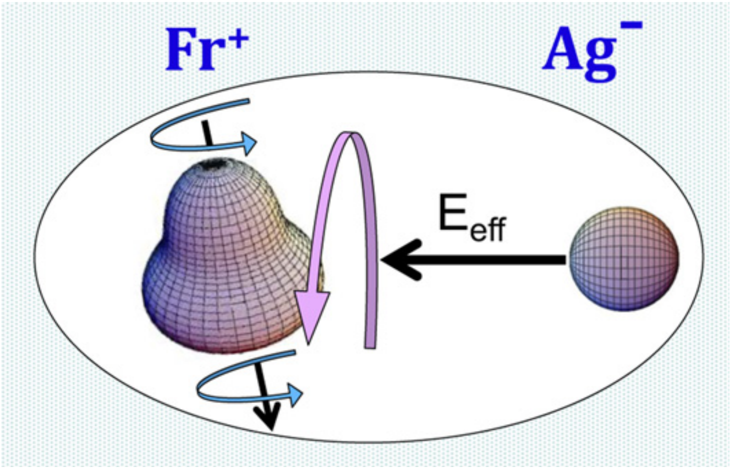
all known cases in radionuclides

Example: ^{223}FrAg

- **intrinsic enhancement of 10^7 compared to ^{199}Hg**

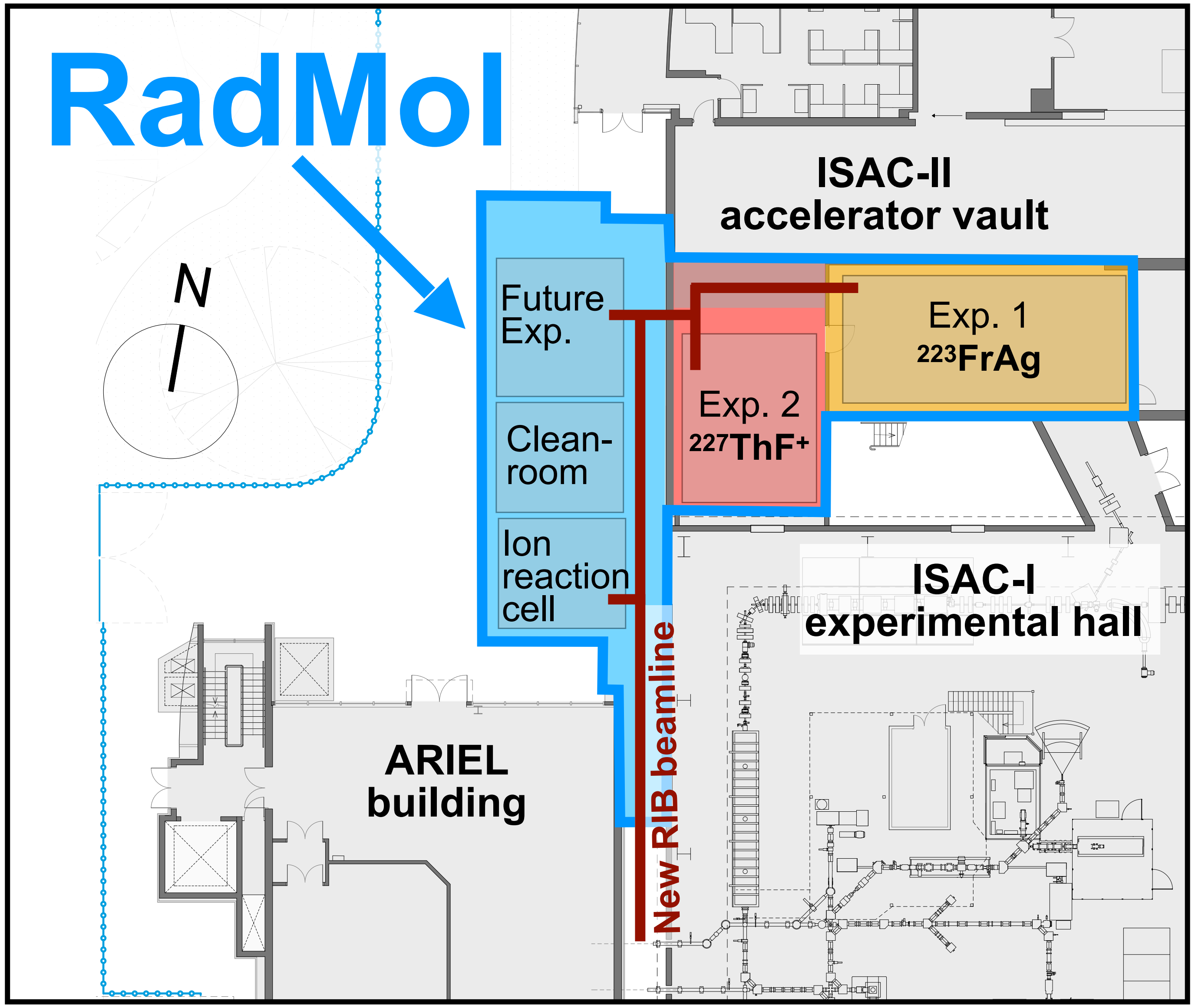
V. V. Flambaum and V. A. Dzuba. Phys. Rev. A 101, 042504 (2020)
T. Fleig. private communications with D. DeMille (2022)

- need to be produced at TRIUMF



RadMol

a radioactive molecule lab for fundamental physics



Goal:

- dedicated laboratory for radioactive molecules
- to host 3 experimental stations
- precision studies for searches for new physics
- Molecular EDM with unprecedented sensitivity to nuclear Schiff moments
- provision for expansions into other fields

TRIUMF advantages:

- large variety in radioactive ion beams (RIB)
- high beamtime availability (3 RIBs)
- existing laboratory space am

Current Canadian Team:

- 12 faculty and staff physicists

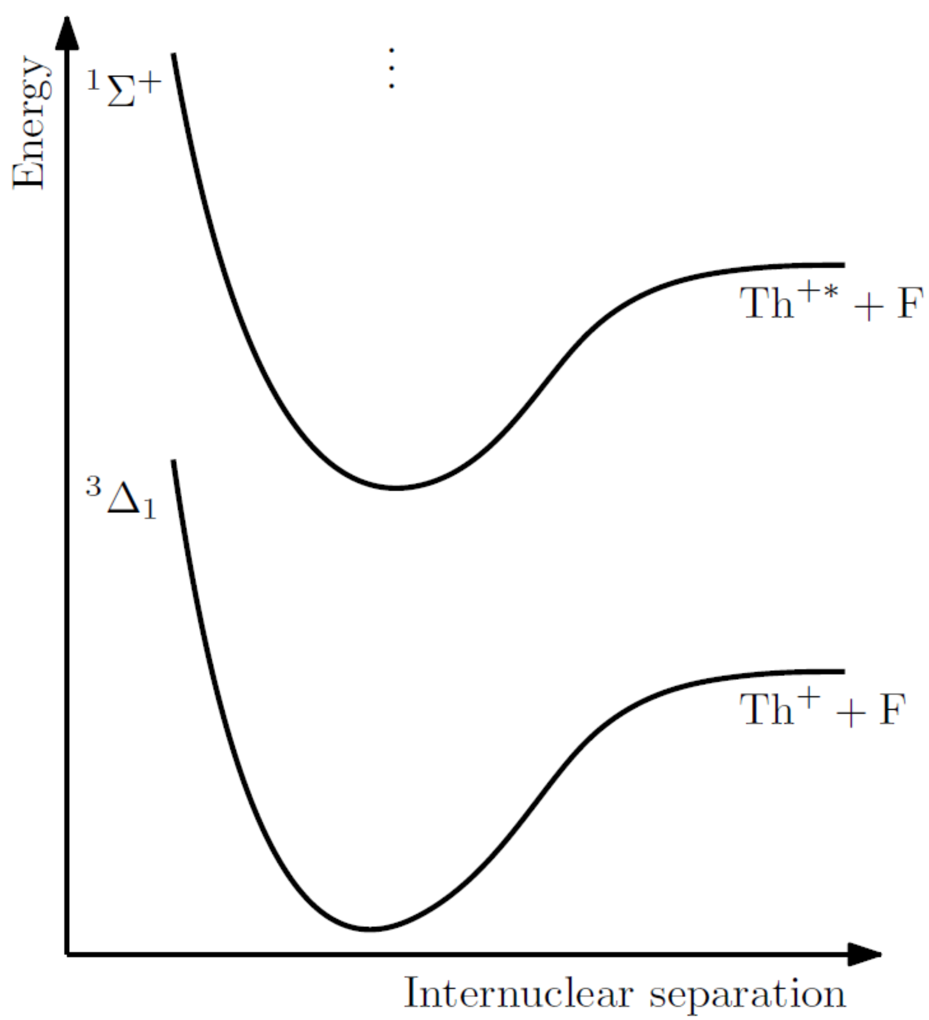
RadMol Collaboration:

UBC, University of Manitoba, TRIUMF, University of Waterloo, McGill, The University of Chicago, University of Colorado Boulder, Massachusetts Institute of Technology, Harvard, University of Toronto, uOttawa, Johns Hopkins University, University of Maryland, Philipps Universität Marburg, Temple University, university of groningen, The University of Edinburgh

$^{227}ThF^{+}$ molecule

half-life: ≈19 days

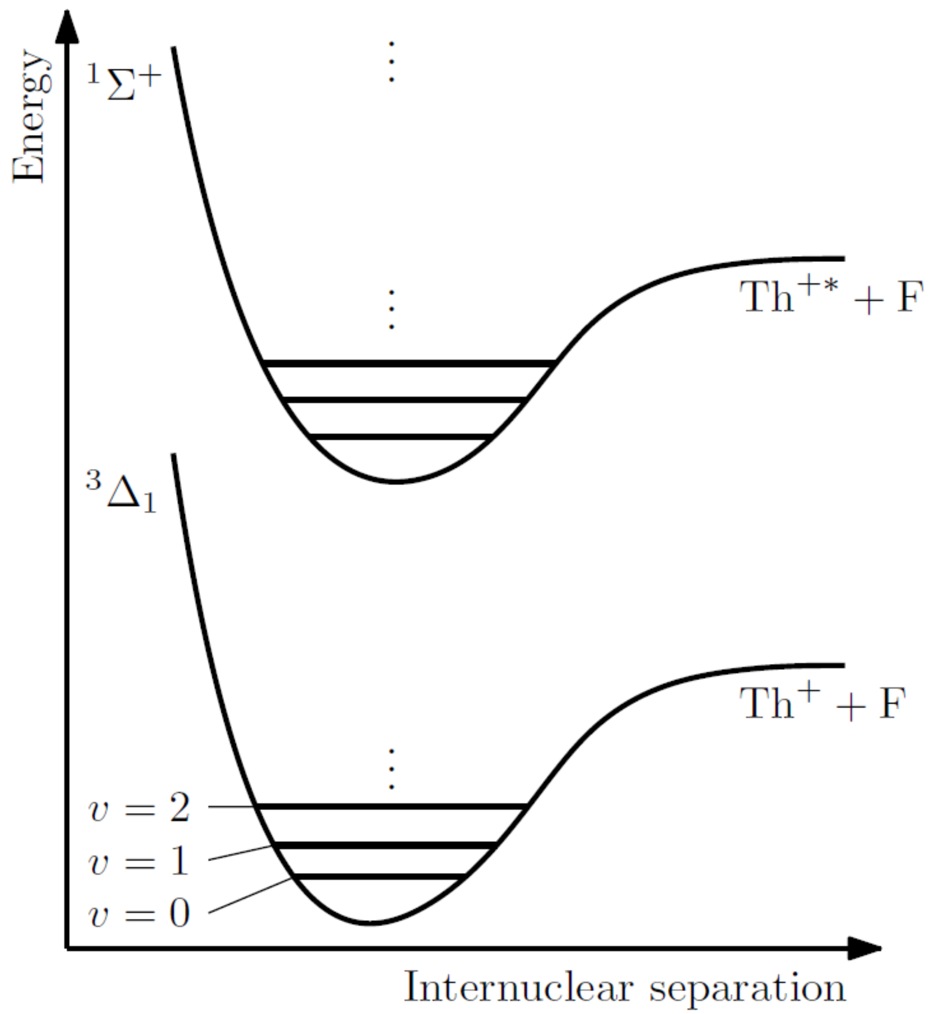
Electronic level ($1000\ cm^{-1}$ ~ 1400 K ~ 0.1 eV)

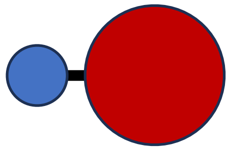


$^{227}ThF^{+}$ molecule

half-life: ≈19 days

Electronic level ($1000\ cm^{-1}$ ~ 1400 K ~ 0.1 eV)
→ Vibrational manifold ($600\ cm^{-1}$ ~ 800 K)





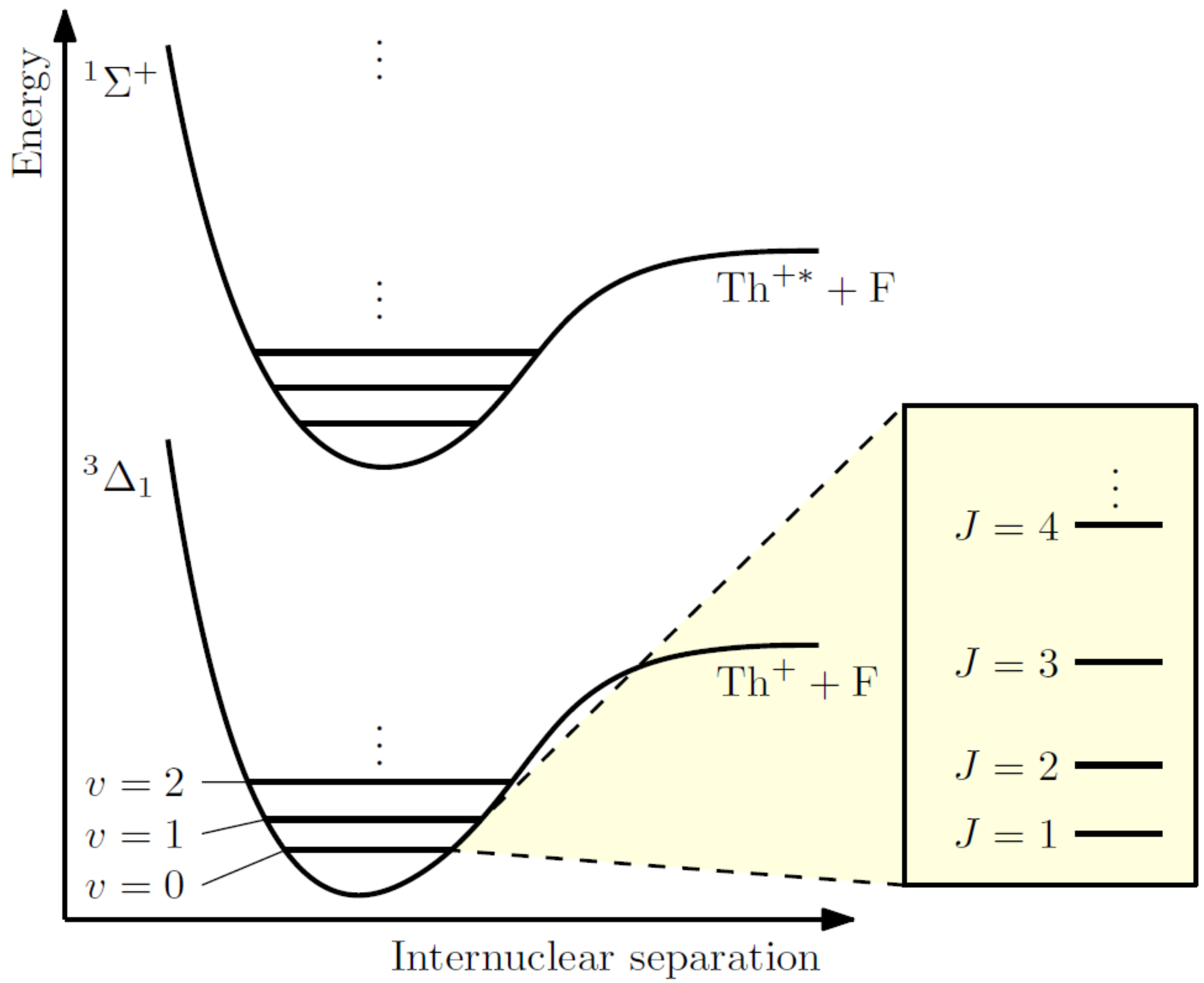
$^{227}ThF^+$ molecule

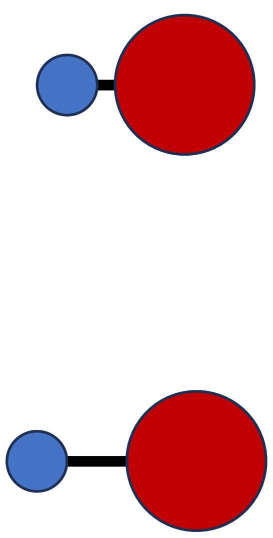
half-life: ≈19 days

Electronic level (1000 cm^{-1} ~ 1400 K ~ 0.1 eV)

→ Vibrational manifold (600 cm^{-1} ~ 800 K)

→ Rotational manifold (1 cm^{-1} ~ 30 GHz ~ 1 K)





$^{227}ThF^{+}$ molecule

half-life: ≈19 days

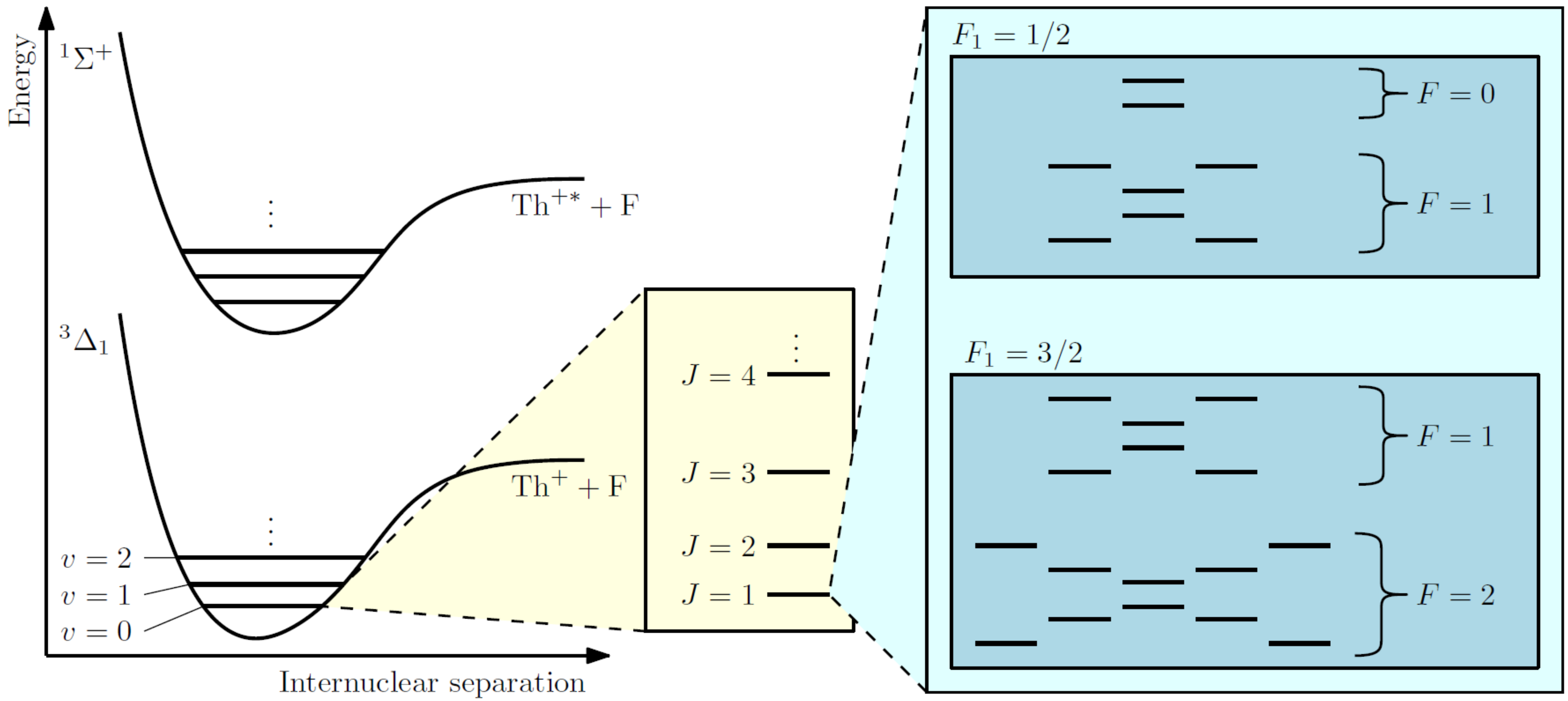
Electronic level (1000 cm^{-1} ~ 1400 K ~ 0.1 eV)

→ Vibrational manifold (600 cm^{-1} ~ 800 K)

→ Rotational manifold (1 cm^{-1} ~ 30 GHz ~ 1 K)

→ Nested hyperfine (100 MHz – 1 GHz ~ 3 – 30 mK)

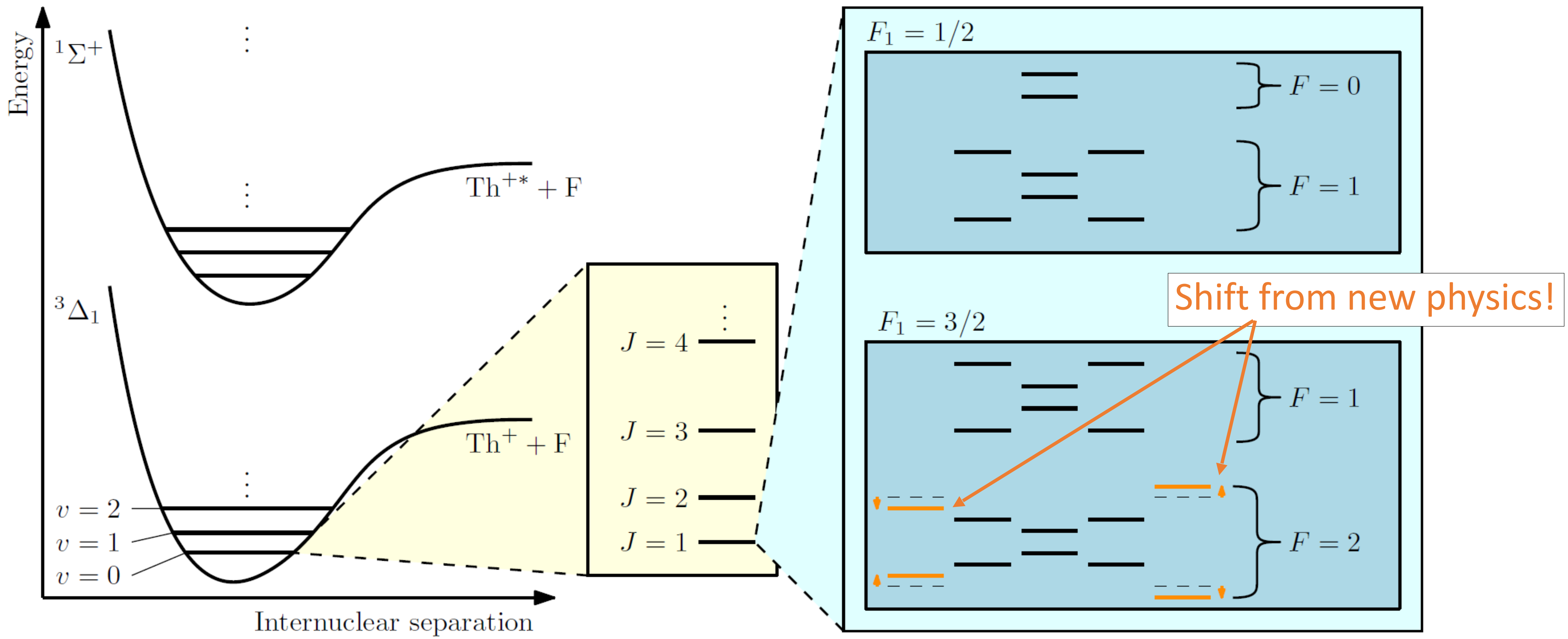
→ Parity doublets, Stark, Zeeman (10 – 100 MHz)



$^{227}ThF^{+}$ molecule

half-life: ≈19 days

Electronic level (1000 cm^{-1} ~ 1400 K ~ 0.1 eV)
→ Vibrational manifold (600 cm^{-1} ~ 800 K)
→ Rotational manifold (1 cm^{-1} ~ 30 GHz ~ 1 K)
→ Nested hyperfine (100 MHz – 1 GHz ~ 3 – 30 mK)
→ Parity doublets, Stark, Zeeman (10 – 100 MHz)

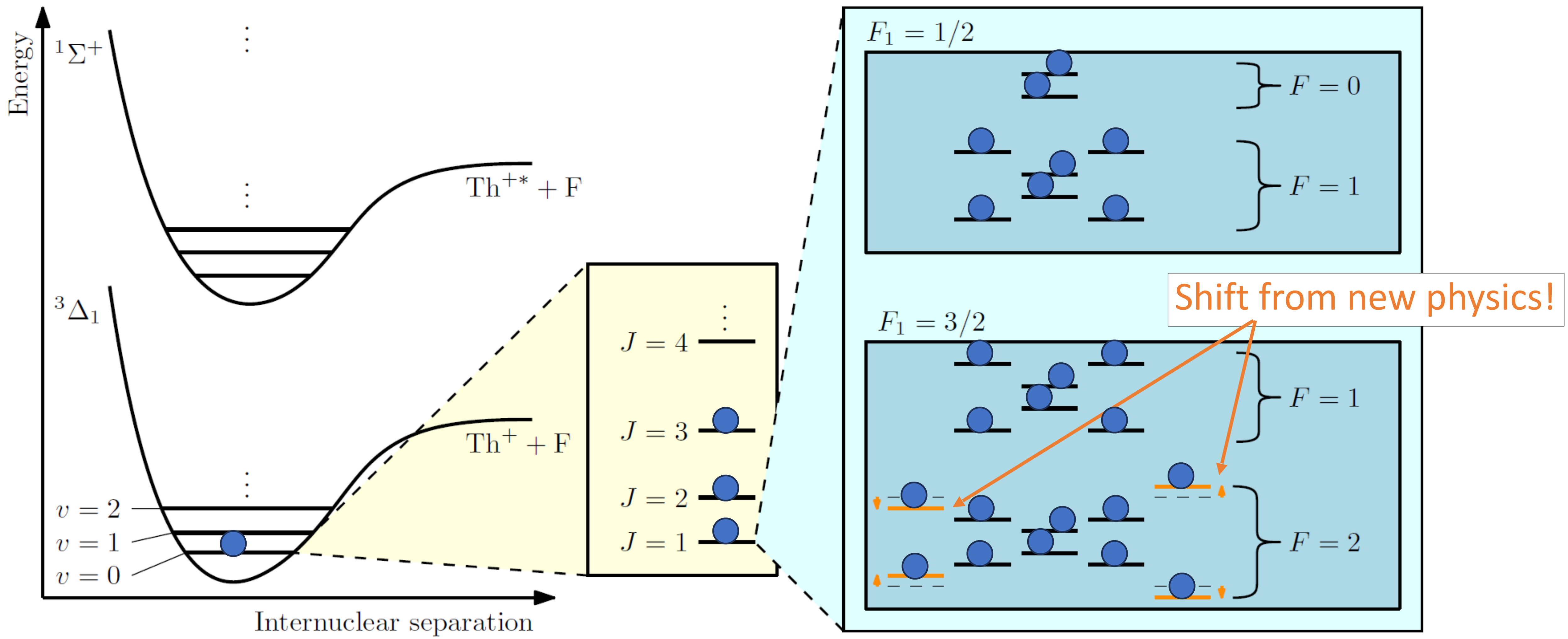


$^{227}ThF^+$ molecule

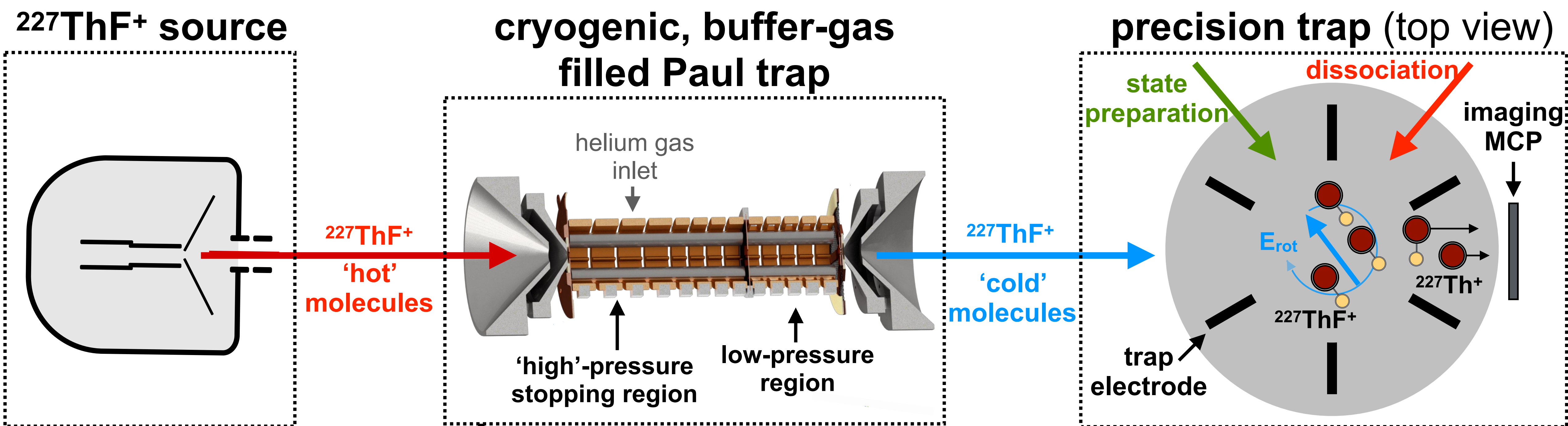
half-life: ≈19 days

Electronic level (1000 cm^{-1} ~ 1400 K ~ 0.1 eV)
→ Vibrational manifold (600 cm^{-1} ~ 800 K)
→ Rotational manifold (1 cm^{-1} ~ 30 GHz ~ 1 K)
→ Nested hyperfine (100 MHz – 1 GHz ~ 3 – 30 mK)
→ Parity doublets, Stark, Zeeman (10 – 100 MHz)

Ions typically produced/cooled to ~10 K
⇒ distributed across multiple *J* (and finer structures).



$^{227}ThF^+$ experiment



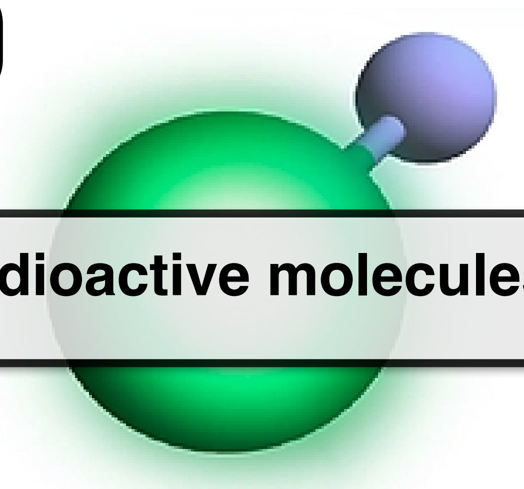
Cooling of
- centre of mass motion
- inner degrees of freedom

- experimental EDM technique analogous to HfF^+ electron EDM experiment

  T. S. Roussy et al. Science 381.6653 (2023), pp. 46–50.
- molecular structure of $^{232}ThF^+$ known from spectroscopy at JILA

  K.B. Ng et al. Phys. Rev. A 105, 022823 (2022)
- access to ^{227}Th via ^{227}Ac sample from TRIUMF life sciences

Multidisciplinary



nuclear theory

BSM interpretation

EDM expertise

particle physics

molecular physics

atomic physics techniques

quantum chemistry

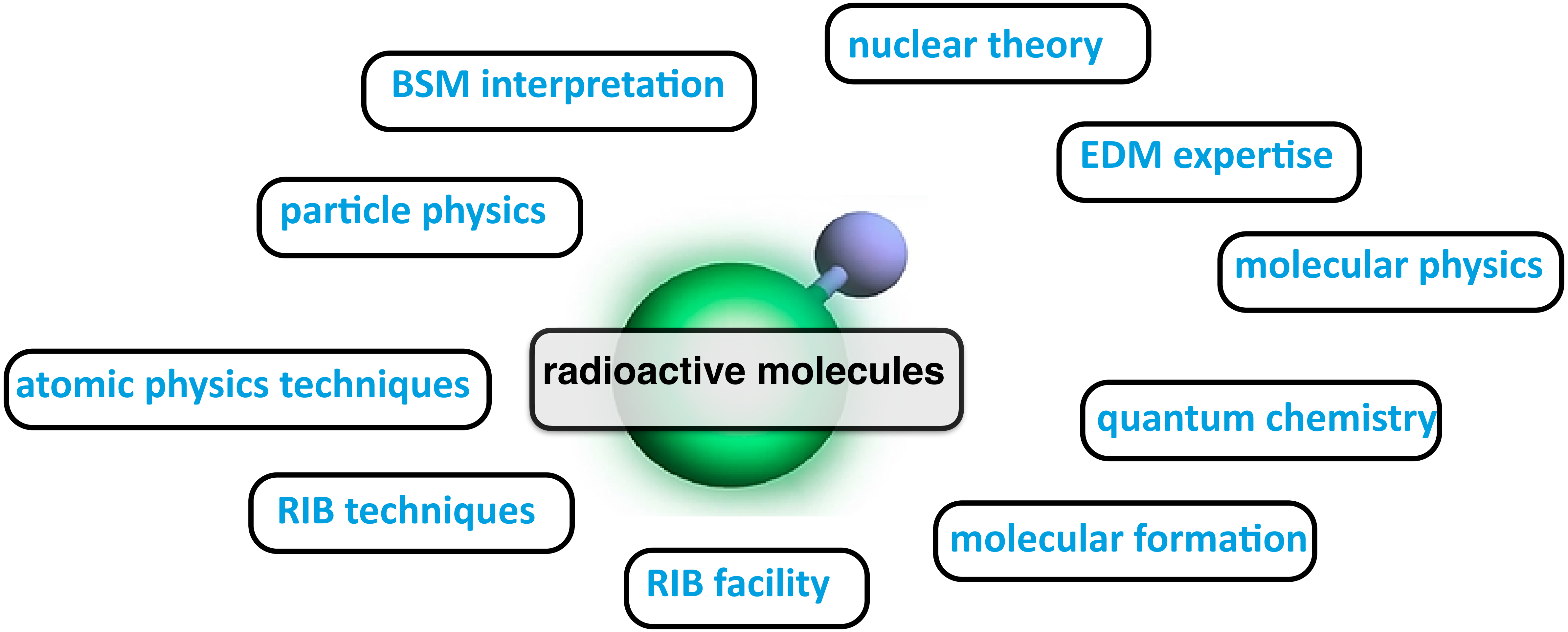
RIB techniques

molecular formation

RIB facility

10

Multidisciplinary



10

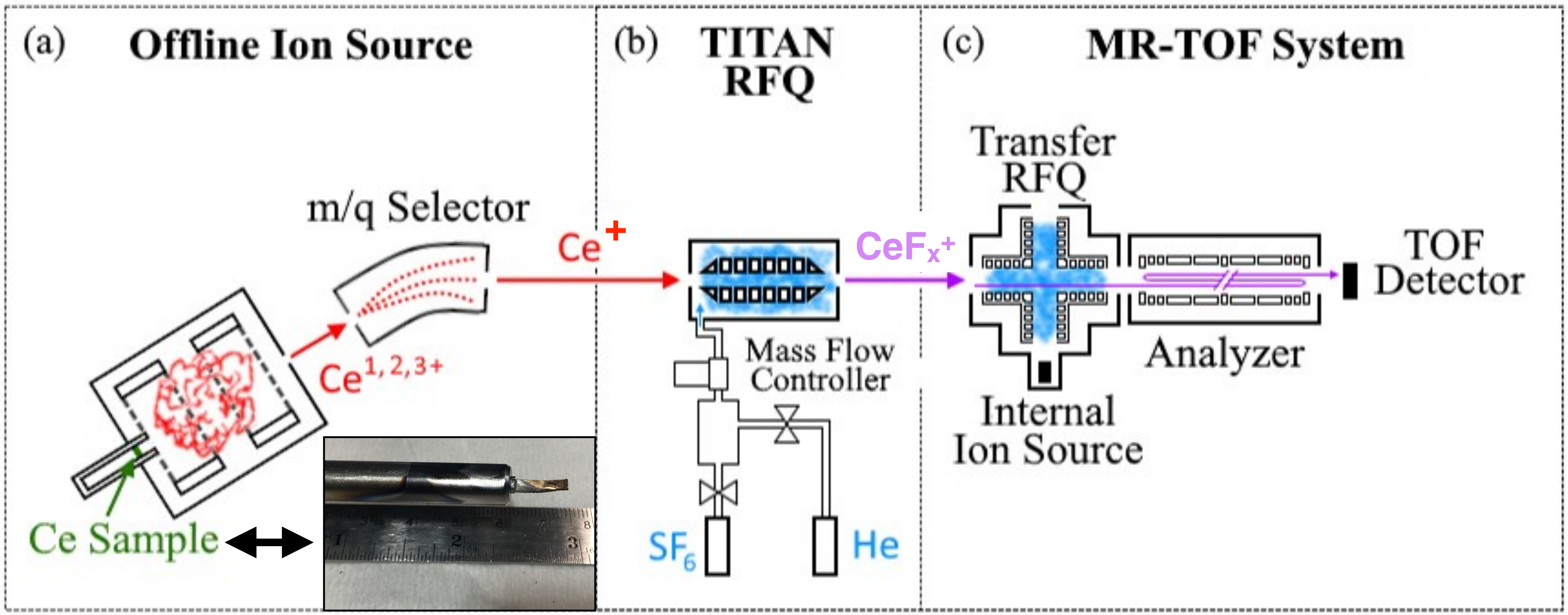
General theme for experiment:

translate high-precision AMO techniques into accelerator lab

Exemplary topics for today:

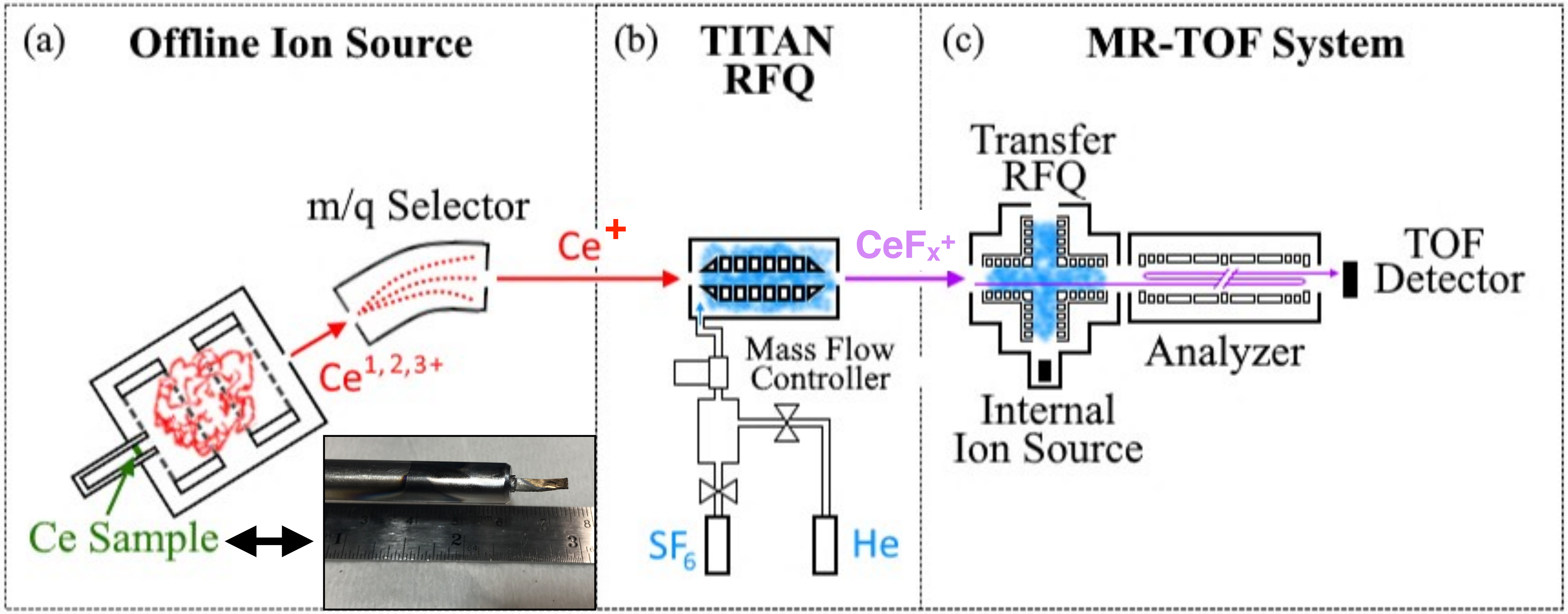
- Formation of ionic molecules
- Cooling of ionic molecules

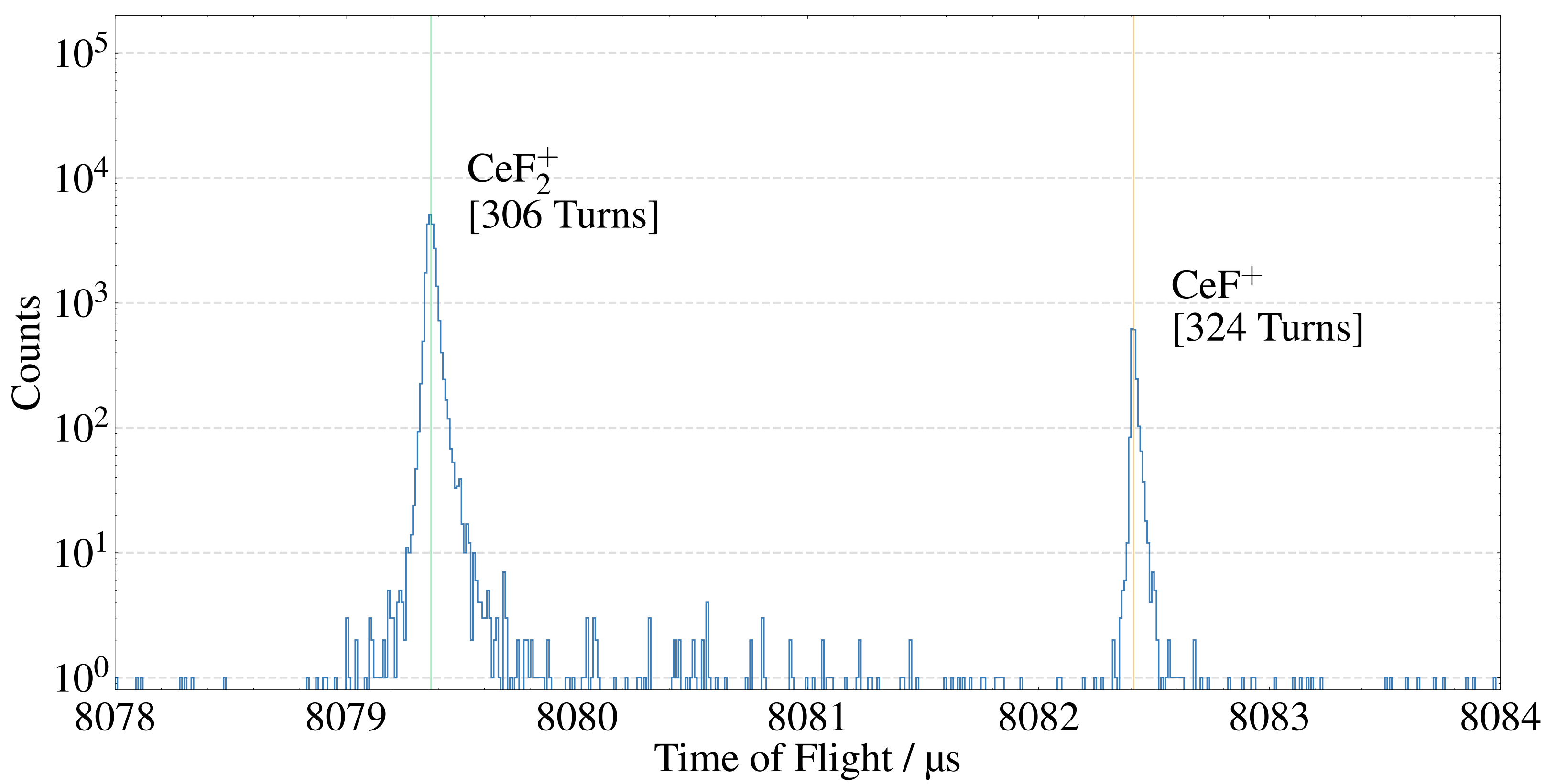
Molecular formation@TITAN

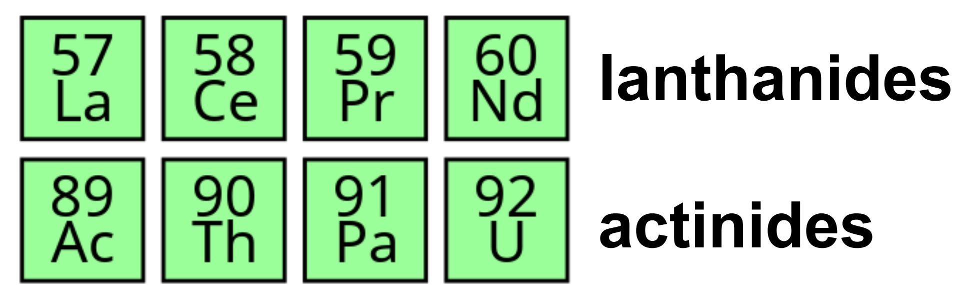


57 La	58 Ce	59 Pr	60 Nd	**lanthanides**
89 Ac	90 Th	91 Pa	92 U	**actinides**

Molecular formation@TITAN



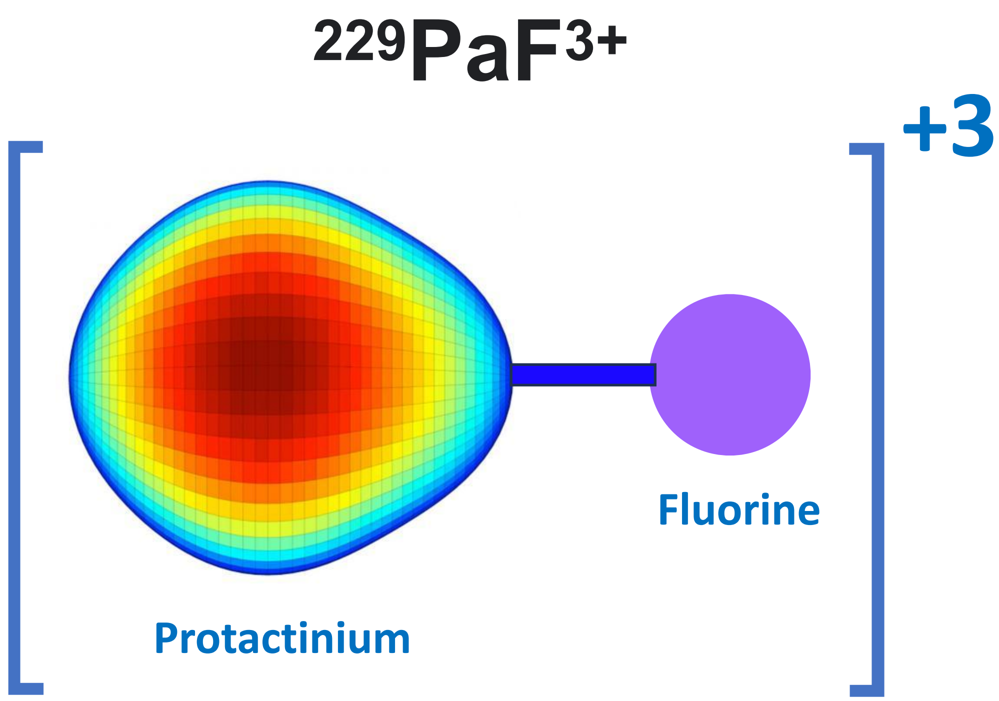


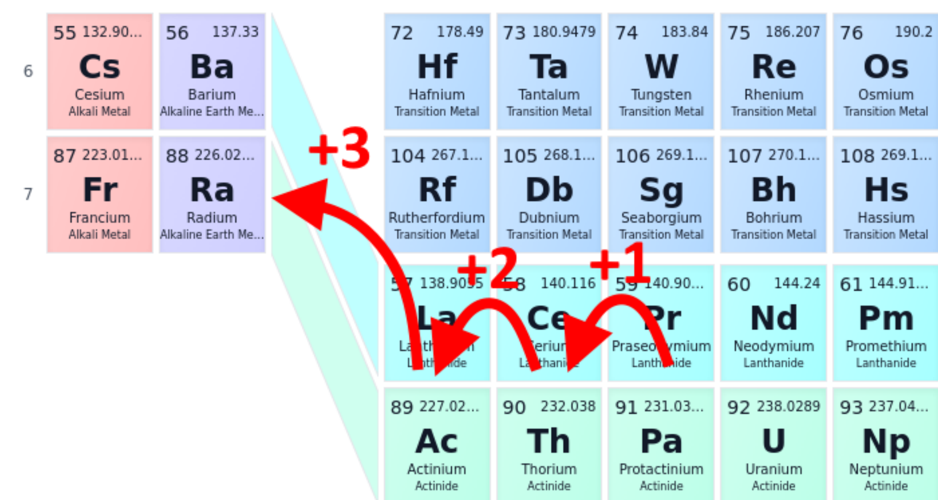


Conclusions:

- CeF^+ successfully formed
- excellent prospect for ThF^+

'Highly' charged radioactive molecules





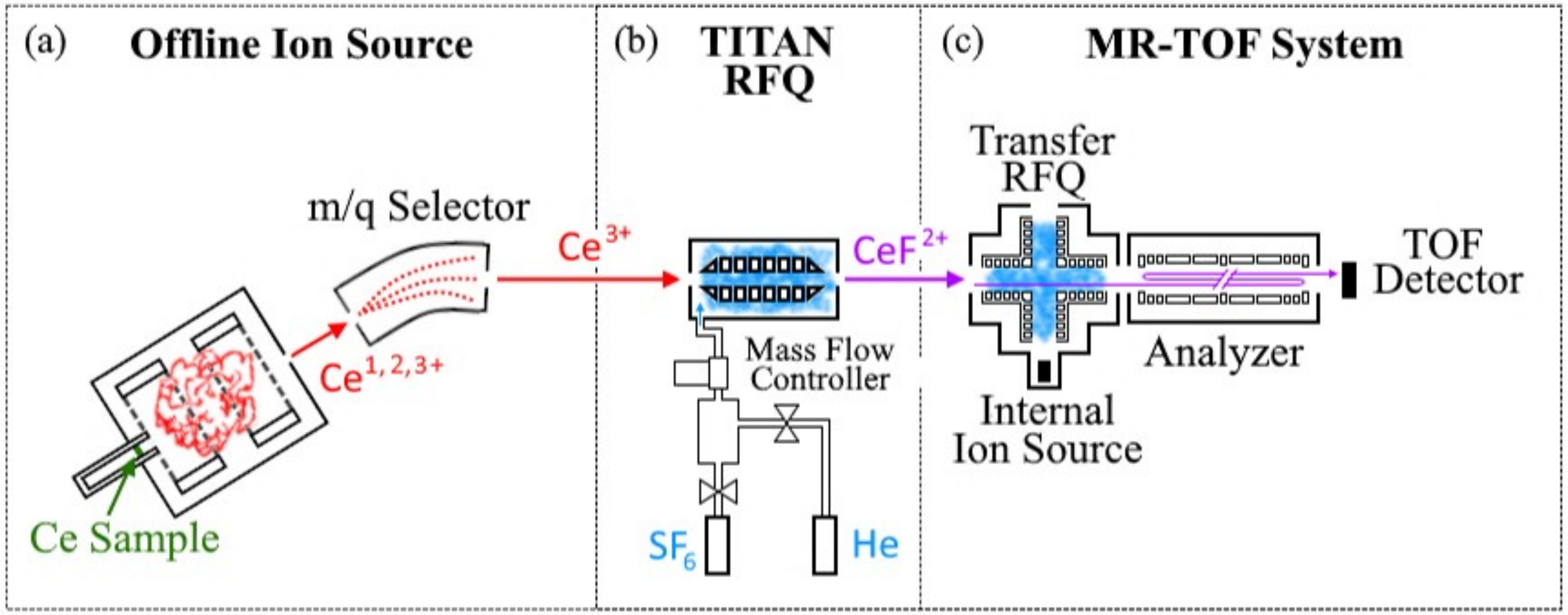
C. Zülch et al., arXiv 2203.10333 (2022)

- notable sensitivity increase for new physics
- iso-electronic to (neutral) RaF
- easily trap-able
- potential for direct laser cooling?

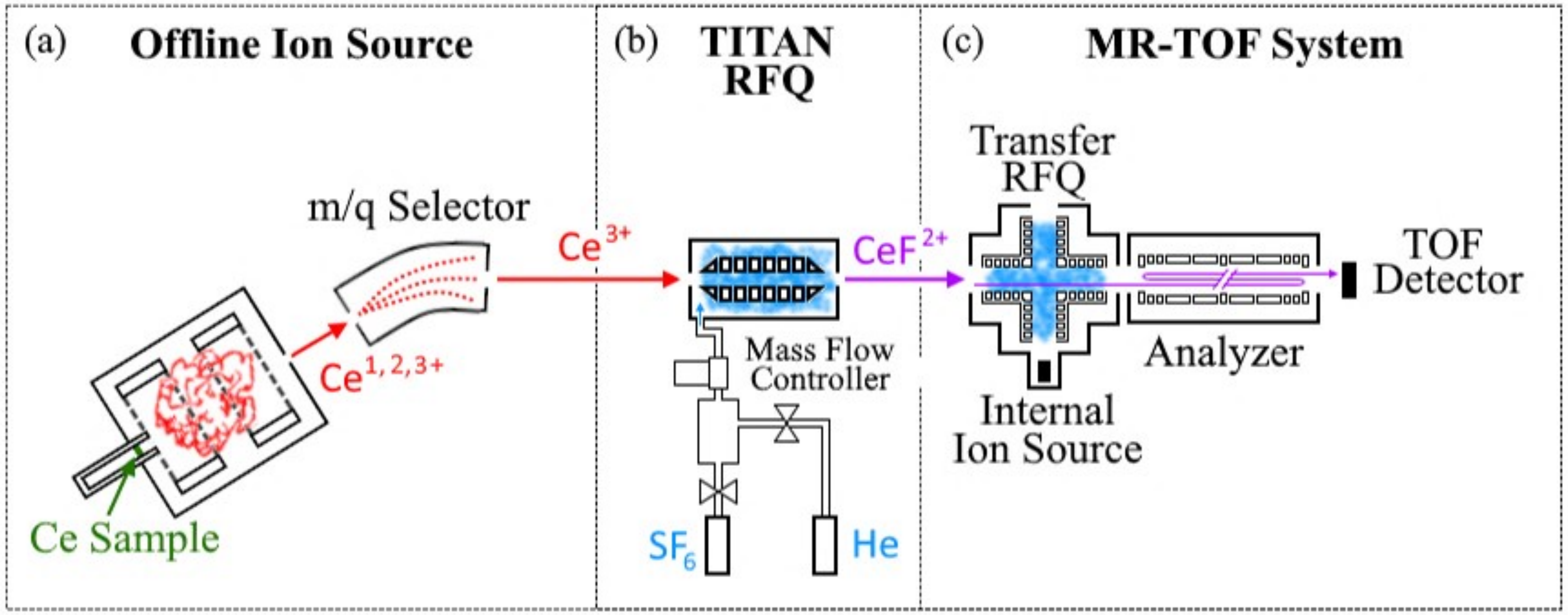
other iso-electronic molecules: ThF^{+2}

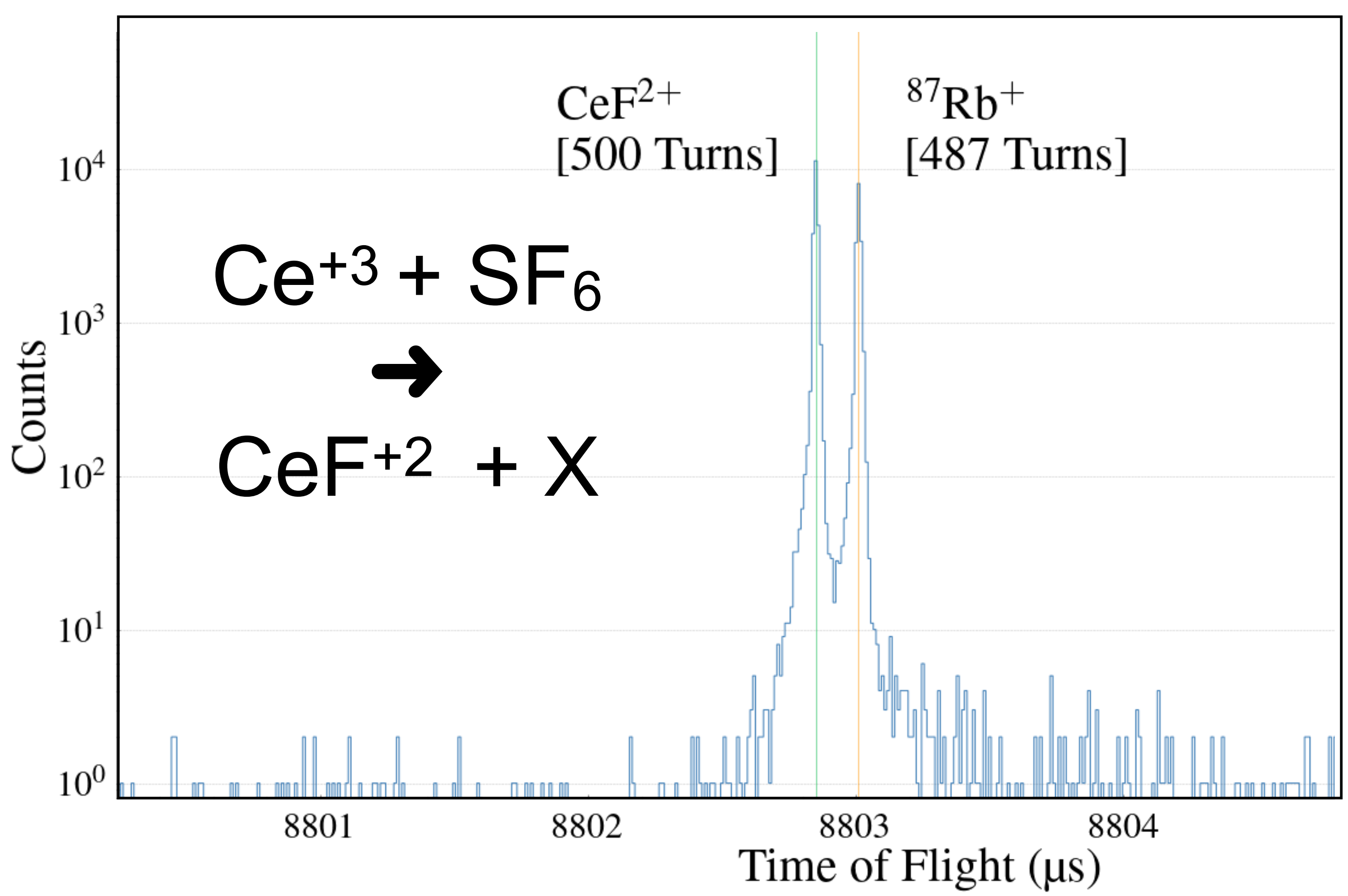
Species	Schiff Scaling Factor (relative to ^{225}Ra)
^{225}Ra	:= 1 (~200x larger than 199Hg)
^{229}Th	2
^{227}Ac	6
^{229}Pa	40

CeF^{2+} in TITAN cooler-buncher



CeF^{2+} in TITAN cooler-buncher





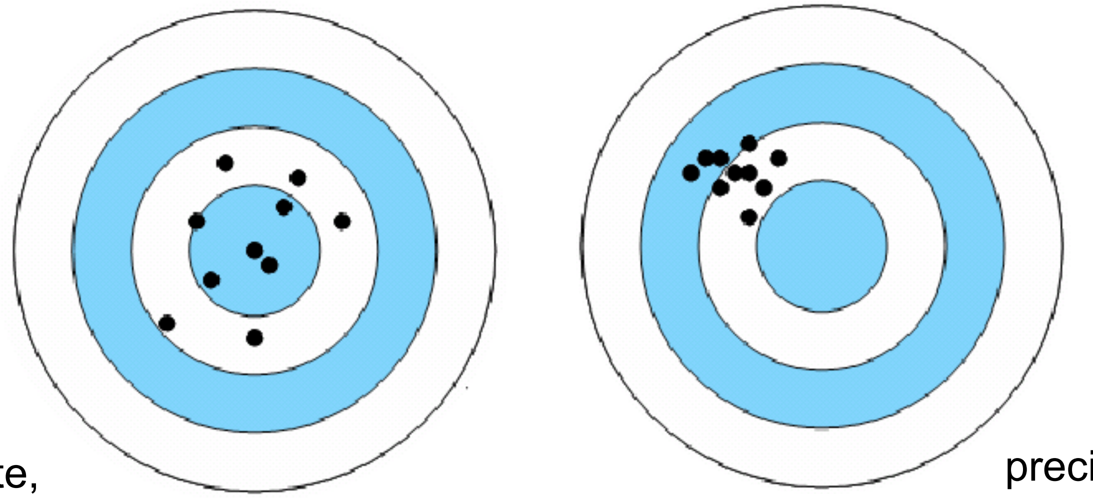
Conclusions:

- CeF^{+2} successfully formed
- Excellent prospect for ThF^{+2}

Atomic physics techniques at RIB facilities

high **precision and accuracy**

K. Blaum, et al., Phys. Scr. T152, 014017 (2013)
P. Campbell et al., Prog. Part. and Nucl. Phys. 86, 127-180 (2016)
J. Dilling et al., Annu. Rev. Nucl. Part. Sci. 68, 45 (2018)



accurate,
but not precise

precise,
but not accurate

ion traps
- masses
- RIB preparations
- mass separation
- in-trap decay

laser spectroscopy
- hyperfine structure
- isotope shifts
- optical pumping

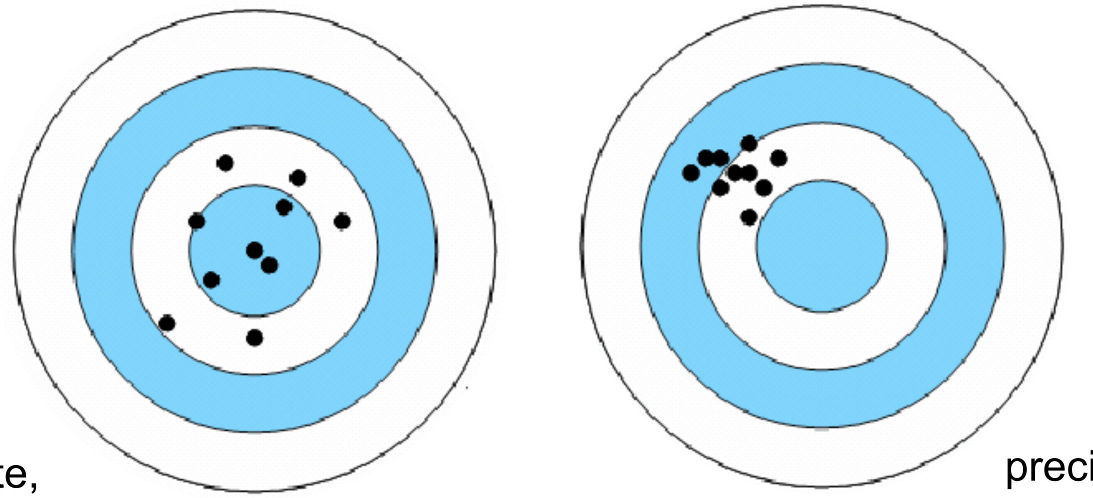
atom traps
- in-trap decay
- laser spectroscopy
- APV

strong programs at TRIUMF:
FrNPC, TITAN, TRINAT, TRILIS, collinear LS

Atomic physics techniques at RIB facilities

high **precision and accuracy**

K. Blaum, et al., Phys. Scr. T152, 014017 (2013)
P. Campbell et al., Prog. Part. and Nucl. Phys. 86, 127-180 (2016)
J. Dilling et al., Annu. Rev. Nucl. Part. Sci. 68, 45 (2018)



accurate,
but not precise

precise,
but not accurate

ion traps
- masses
- RIB preparations
- mass separation
- in-trap decay

laser spectroscopy
- hyperfine structure
- isotope shifts
- optical pumping

atom traps
- in-trap decay
- laser spectroscopy
- APV

Challenges

short half-lives

$T_{1/2} < 10$ ms

($\Delta m/m = 6 \cdot 10^{-8}$)

M. Smith et al., PRL 101, 202501 (2008)

low intensity

masses: 0.5 ions / h

M. Block et al., Nature 463, 785 (2010)
E. Minaya Ramirez et al., Science 337, 1207(2012)

temperature

buffer gas cooling

(selected cases of laser cooling)

300 K

↕

μK - mK - K

purity

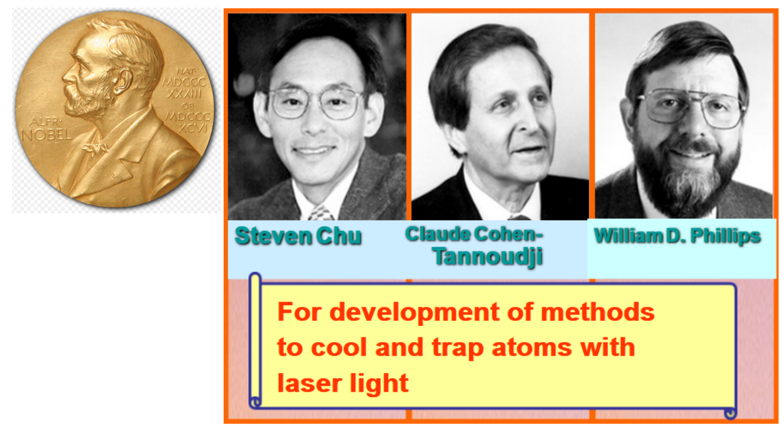
$R = m/\Delta m > 5 \cdot 10^6$

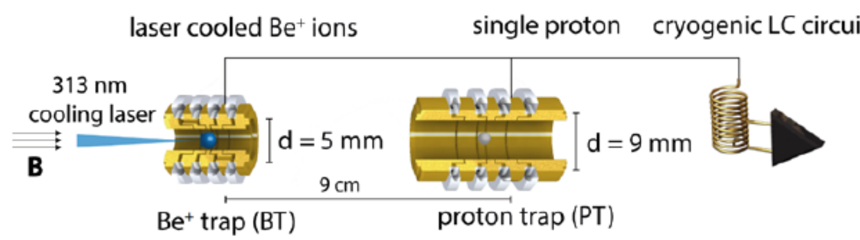
limited ion capacity

S. Eliseev et al., PRL 110, 082501 (2013)

Doppler Cooling

- Powerful technique to reach sub-K atom and ion temperatures [1]
- Standard tool for high-precision measurements: atomic clocks [2], quantum information science [3], physics beyond the standard model [4]



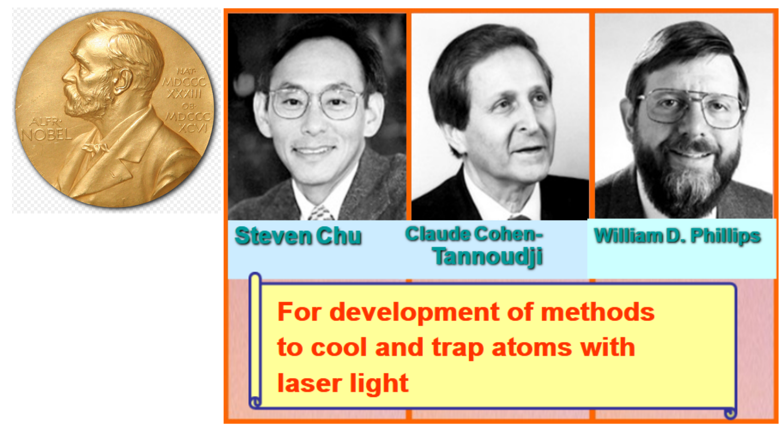


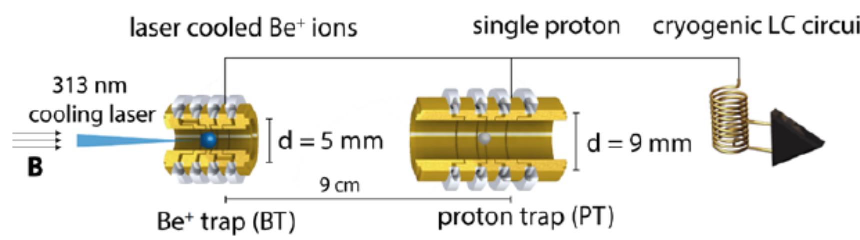
[1] T. Haensch and A. Schawlow, Optics Communications 13, 68 (1975).
D. J. Wineland and W. M. Itano, Phys. Rev. A 20, 1521 (1979).
J. Eschner et al, J. Opt. Soc. Am. B20, 1003 (2003).

[2] D. Ludlow et al, Rev. Mod. Phys. 87, 637 (2015).
[3] C. D. Bruzewicz et al, Applied Physics Reviews 6, 021314 (2019).
[4] M. S. Safronova et al, Rev. Mod. Phys. 90, 025008 (2018).

Doppler Cooling

- Powerful technique to reach sub-K atom and ion temperatures [1]
- Standard tool for high-precision measurements: atomic clocks [2], quantum information science [3], physics beyond the standard model [4]





[1] T. Haensch and A. Schawlow, Optics Communications 13, 68 (1975).
D. J. Wineland and W. M. Itano, Phys. Rev. A 20, 1521 (1979).
J. Eschner et al, J. Opt. Soc. Am. B20, 1003 (2003).

[2] D. Ludlow et al, Rev. Mod. Phys. 87, 637 (2015).
[3] C. D. Bruzewicz et al, Applied Physics Reviews 6, 021314 (2019).
[4] M. S. Safronova et al, Rev. Mod. Phys. 90, 025008 (2018).

- Specific applications with RIBs

G. D. Sprouse and L. A. Orozco, Annu. Rev. Nucl. Part. Sci.. 47, 429 (1997)
J. A. Behr et al., Phys. Rev. Lett. 79, 375 (1997).
M. Trinczek et al., Phys. Rev. Lett. 90, 012501 (2003).
L. B. Wang et al., Phys. Rev. Lett. 93, 142501 (2004).
P. A. Vetter et al., Phys. Rev. C 77, 035502 (2008).
J. R. A. Pitcairn et al., RRC 79, 015501 (2009)
A. Takamine et al., Phys. Rev. Lett. 112, 162502 (2014)
B. Fenker et al., Phys. Rev. Lett. 120, 062502 (2018)

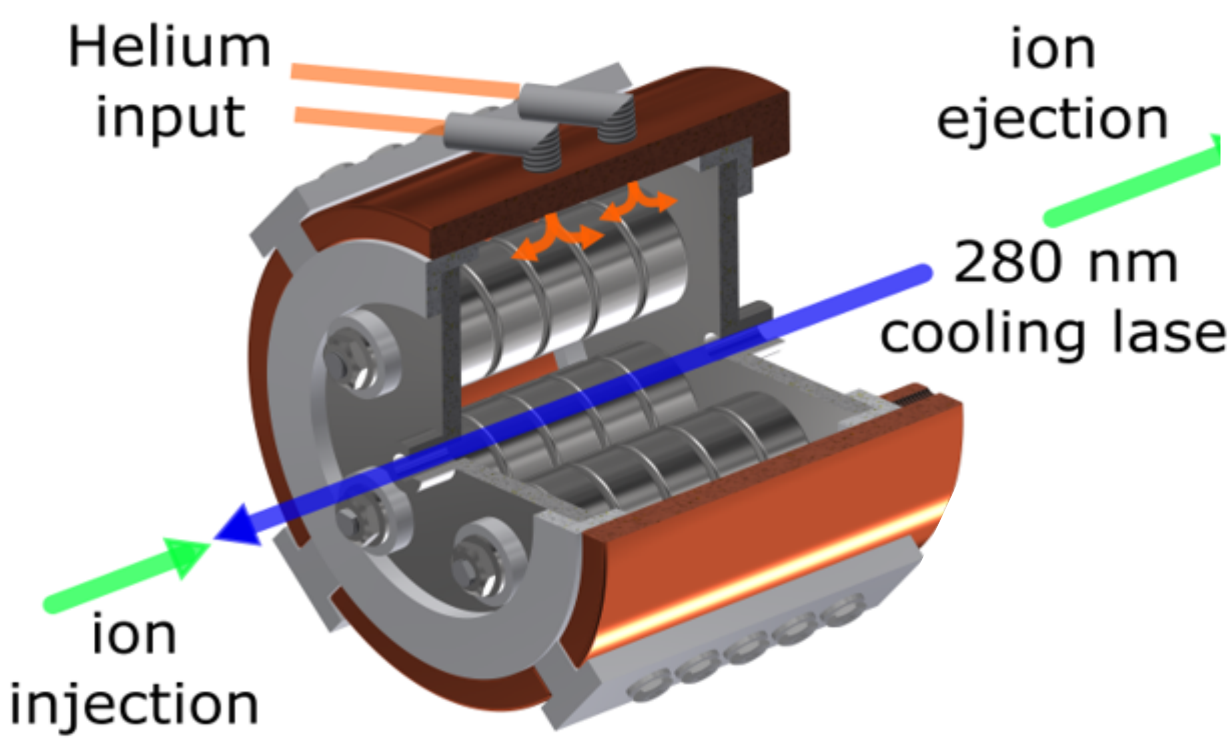
- unexplored as cooling technique to deliver high quality (molecular) RIBs

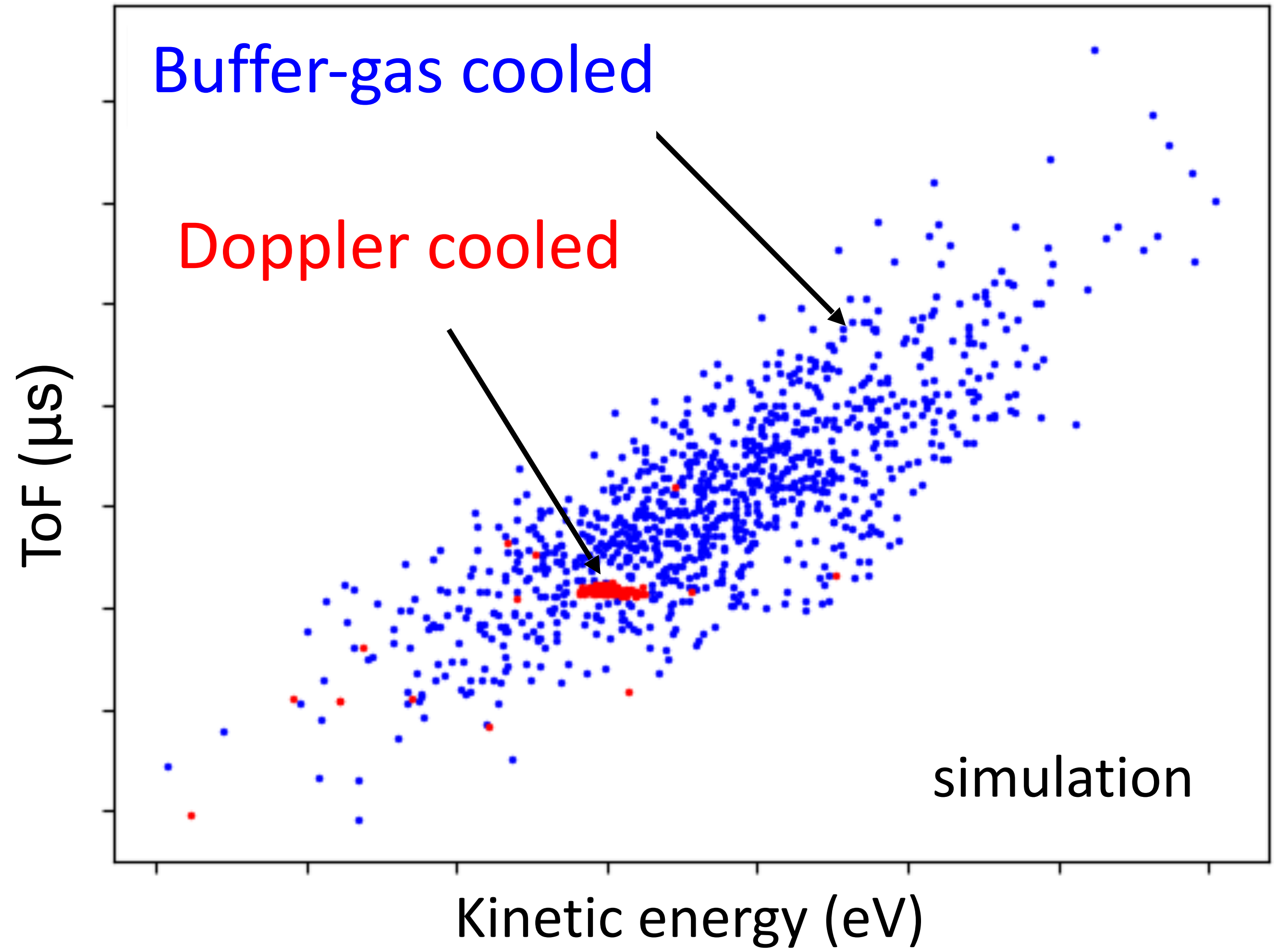
Goal: provide ultra-cold (molecular) RIBs

- ... compatible with short half-lives
- ... universally applicable (via sympathetic cooling)

Experimental Demonstration at MIRACLS
(with TITAN contributions)

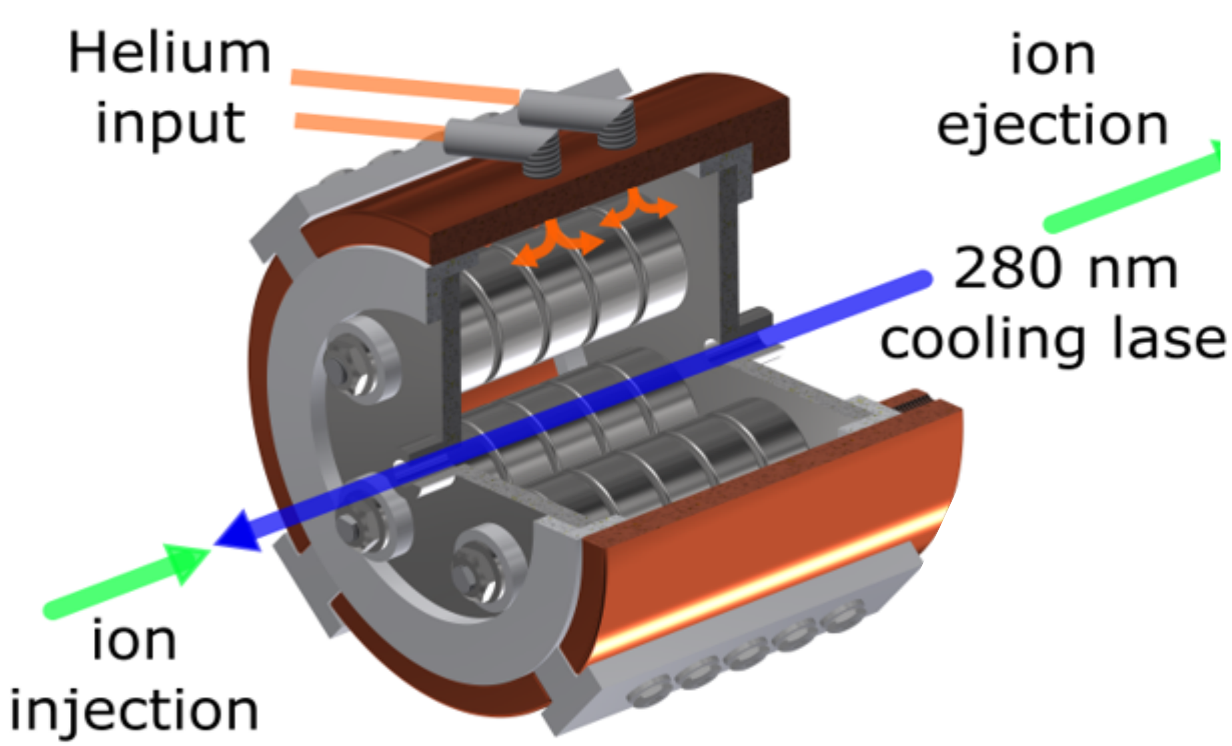
Paul trap:



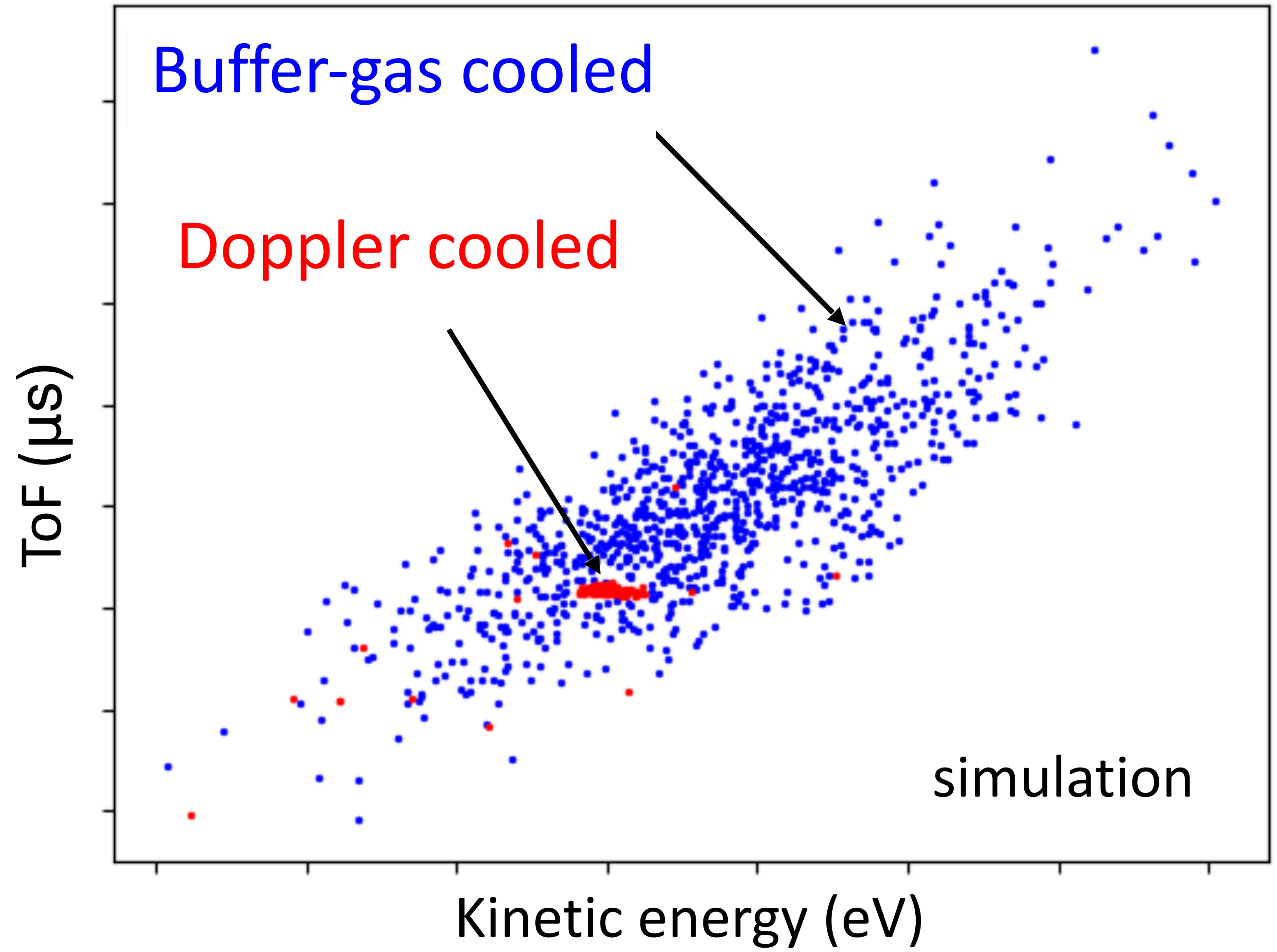


Experimental Demonstration at MIRACLS
(with TITAN contributions)

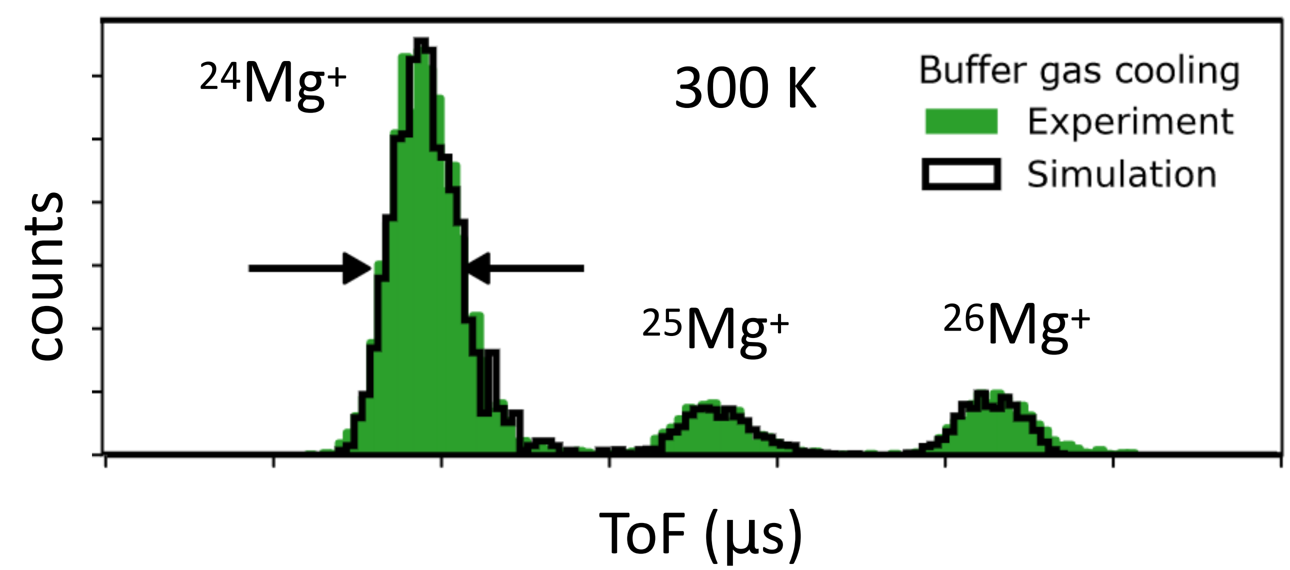
Paul trap:



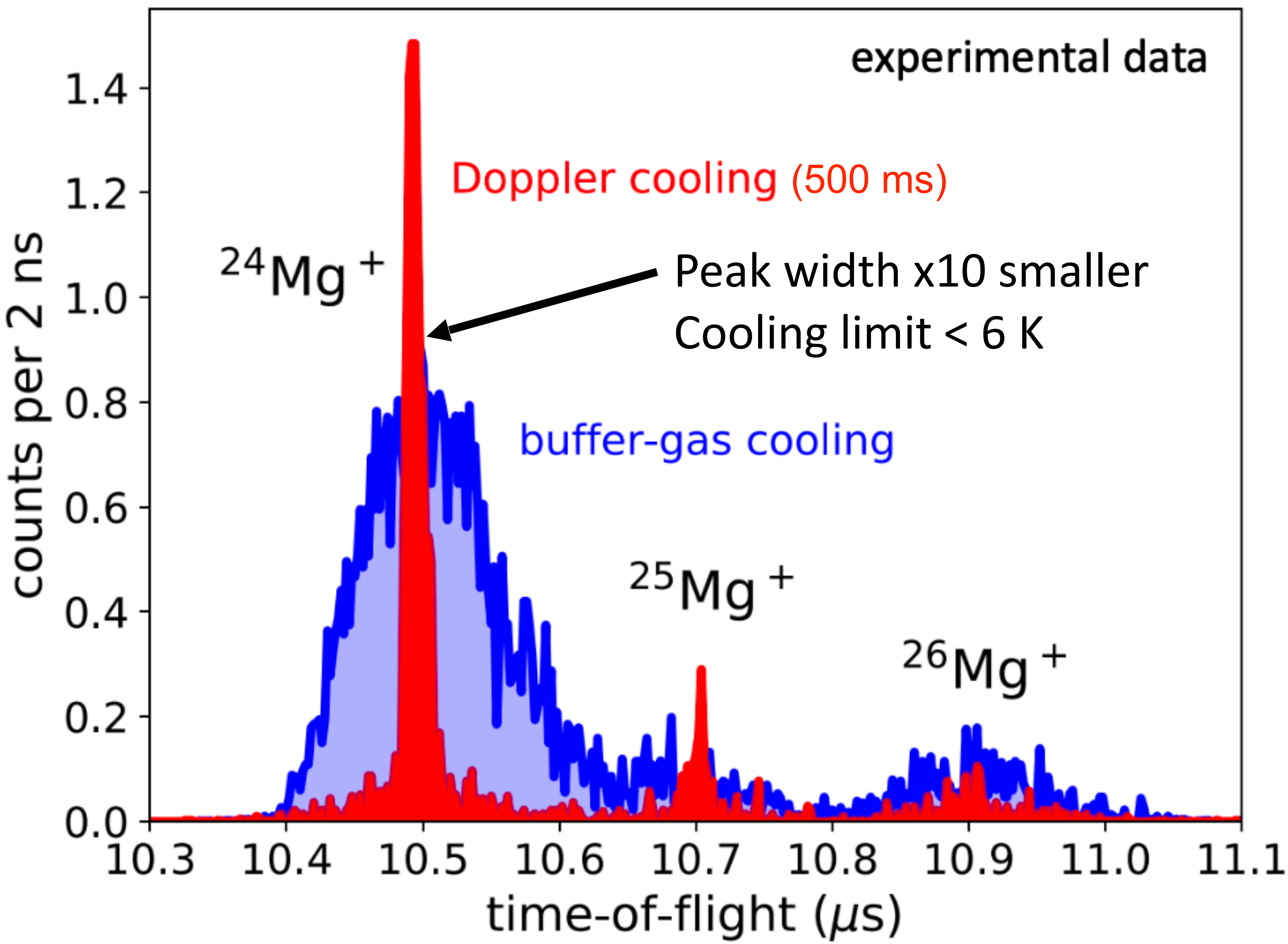
externally produced, fast and 'hot' Mg^+ ion beam



17



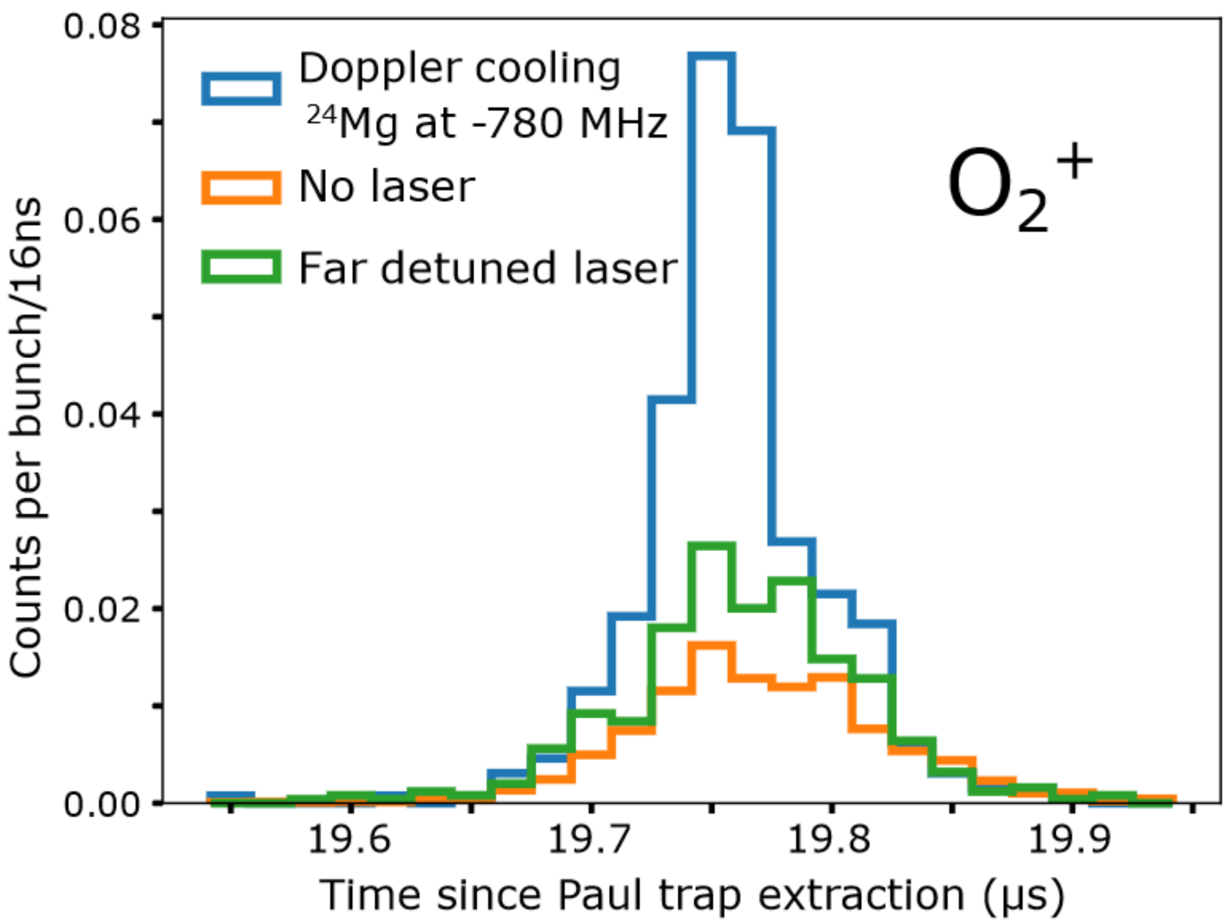
Experimental results



S. Sels, F. Maier et al., Phys. Rev. Research 4, 033229 (2022)

Sympathetic cooling

- ‘universal’ availability of cold ion ensembles
- including ionic systems which cannot be directly laser-cooled



opportunity for cold molecular RIBs

	O_2^+
Peak width residual-gas or buffer-gas cooling	113(5) ns
Sympathetic cooling	58(4) ns
Improvement in countrate	Factor 2.6

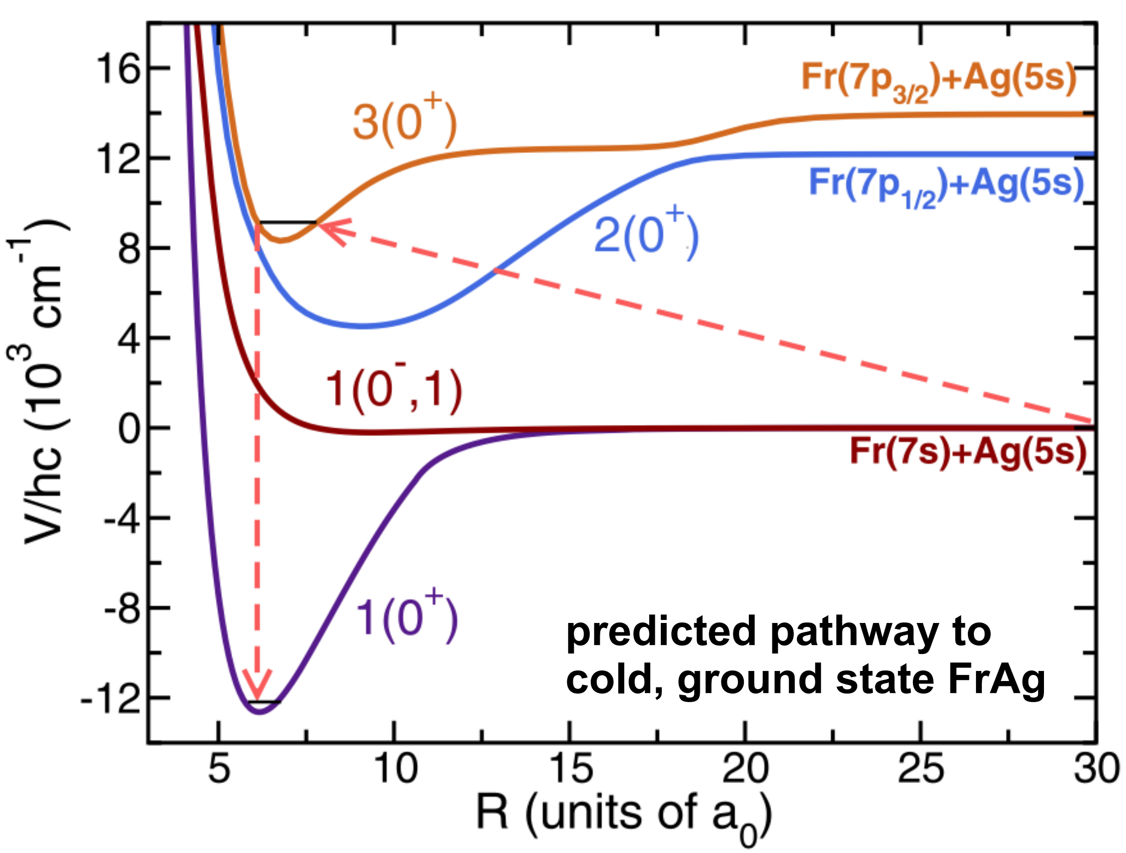
S. Sels, F. Maier et al., Phys. Rev. Research 4, 033229 (2022)

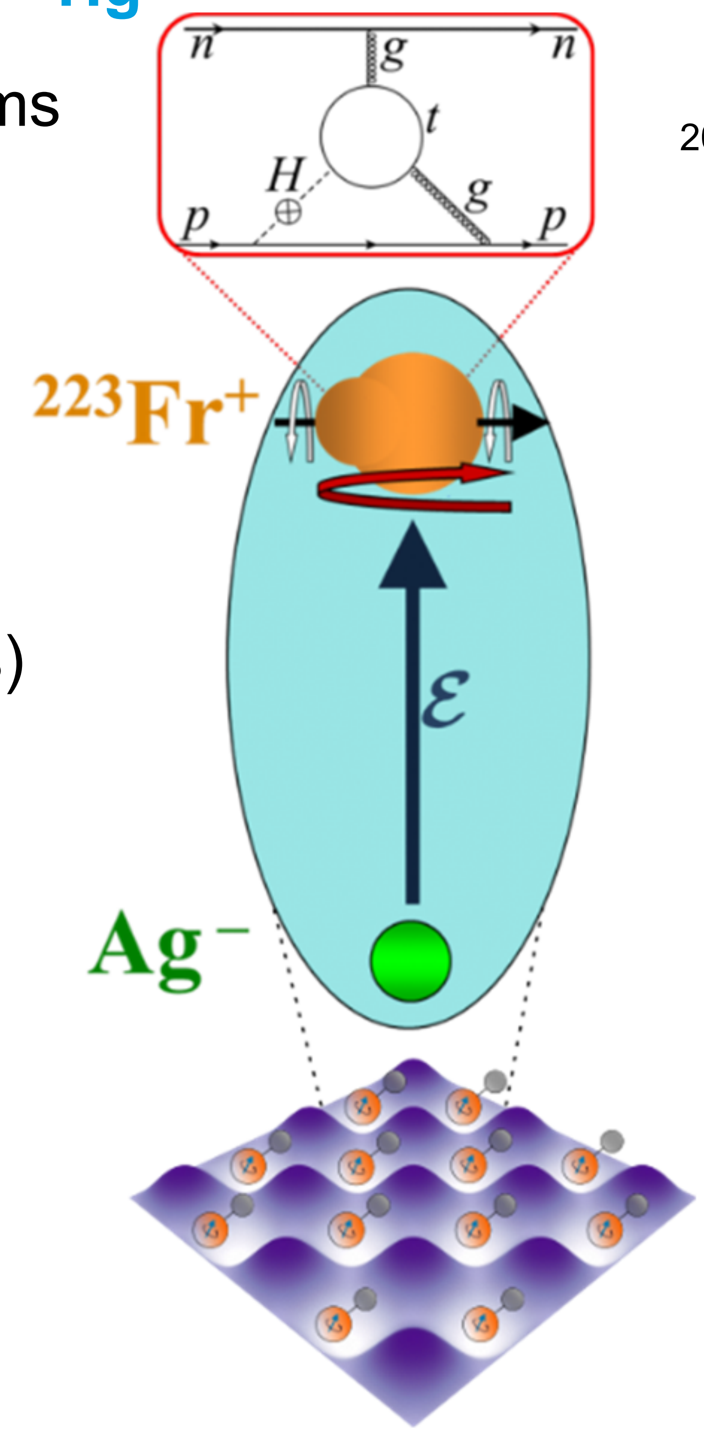
Can be done better analogous to existing work, e.g. [1],[2]

J. Wuebbena et al, Phys. Rev. A 85, 043412,2012.
[2] M. Guggemos, New Journal of Physics 17, 103001, 2015.

Neutral molecules: FrAg

- **Schiff moment: intrinsic enhancement of 10^7 compared to ^{199}Hg**
- ultracold molecule assembled from laser-cooled Fr and Ag atoms
- confined in optical lattice
- ^{223}Fr ($T_{1/2}$=22 min) at ISAC: $1.3 \cdot 10^7$ ions/sec
- infrastructure and expertise at TRIUMF's Fr trapping facility
- first exp. goal: measurement of Fr s-wave scattering length
  (input for formation of Bose Einstein Condensate with Fr atoms)





J Kłos et al., New J. Phys. 24, 025005 (2022)

Summary

- **Radioactive Molecules**
  - ➡ entirely new science path
  - ➡ intriguing & unexplored **probes for New Physics**

- **RadMol**
  - ➡ dedicated laboratory for radioactive molecules & precision studies at TRIUMF
  - ➡ initial focus: **CP-violating nuclear Schiff moments**
    - ✦ **octupole deformed** nuclide
    - ✦ in **polar molecule**
    - ✦ In atom or ion **trap**
  - ➡ requires multidisciplinary approach & technical developments (today: formation, cooling)

RadMol Collaboration

Institution	Department	Principal Investigators
TRIUMF	Physical Sciences	Behr, Holt, Malbrunot-Ettenauer, Kwiatkowski, Teigelhöfer
	Accelerator Division	Babcock, Charles
	Life Sciences Division	Radchenko
University of British Columbia	Physics&Astronomy	Madison
	Chemistry	Momose, Krems
University of Toronto	Physics	Vutha
University of Waterloo	Physics&Astronomy	Jamison
University of Manitoba	Physics&Astronomy	Gwinner
McGill University	Physics	Buchinger
University of Ottawa	Physics	Stolow
University of Chicago / USA	Physics	DeMille
University of Colorado, Boulder / USA	Physics	Cornell
University of Edinburgh	Physics&Astronomy	Reiter
University of Groningen / NL	Physics	Borschevsky, Hoekstra
Harvard University	Physics	Fan
Johns Hopkins University/USA	Chemistry	Cheng
Massachusetts Institute of Technology / USA	Physics	Garcia Ruiz
University of Maryland / USA	Physics	Orozco
University of Marburg / GER	Chemistry	Berger
Temple University / USA	Physics	Kotochigova

Acknowledgement

R. Simpson, I. Belosevic, A. Mollaebrahimi, C. Walls, C. Chambers, M. Au, P. Justus, C.Charles, L. Croquette, K.B. Ng, K. Madison, SME, A. Kwiatkowski
and all members of the **RadMol** and **TITAN** and collaborations

TITAN

University of Manitoba, University of Calgary, SFU, McGill, Max-Planck-Institut für Kernphysik Heidelberg, University of Victoria, Justus-Liebig-Universität Giessen, Colorado School of Mines, University of Notre Dame, UBC, Westfälische Wilhelms-Universität Münster, university of groningen (kvi - center for advanced radiation technology), CNRS, GANIL Spiral2, Technische Universität Dresden

NSERC CRSNG

European Research Council (erc)

ATTRACT

MIRACLS

I. Belosevic, L. Croquette, P. Fischer, C. Kanitz, F. Hummer, E. Leistenschneider, S. Lechner, F. Maier, P. Plattner, A. Roitman, M. Rosenbusch, S. Sels, F. Wienholtz, M. Vilen, R. Wolf, F. Buchinger, W. Nörtershäuser, L. Schweikhard, SME

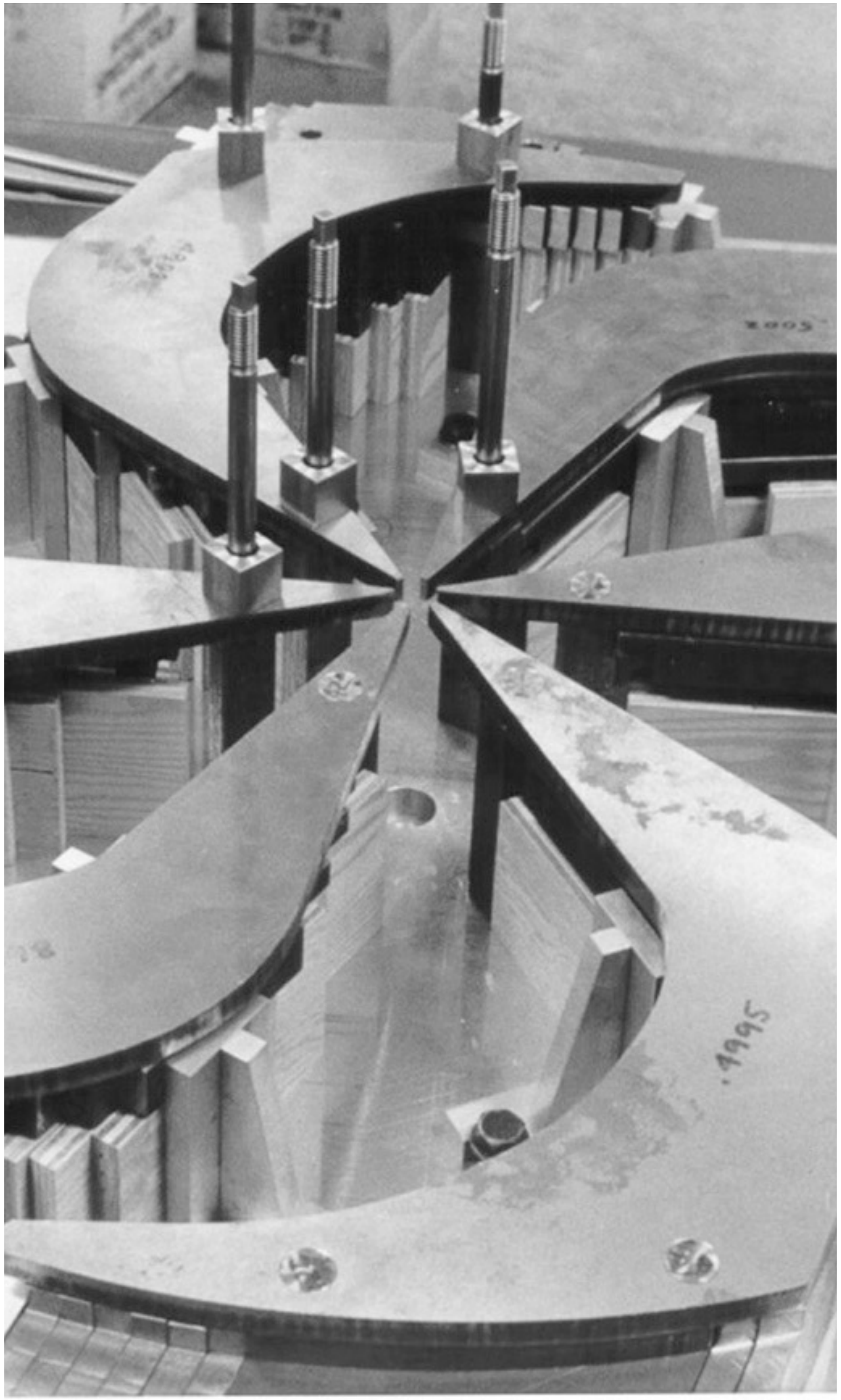
CERN, Universität Greifswald – Wissen lockt. Seit 1456, Technische Universität Darmstadt, McGill, TRIUMF

TRIUMF

Thank you
Merci

www.triumf.ca

Follow us **@TRIUMFLab**



Discovery, accelerated