

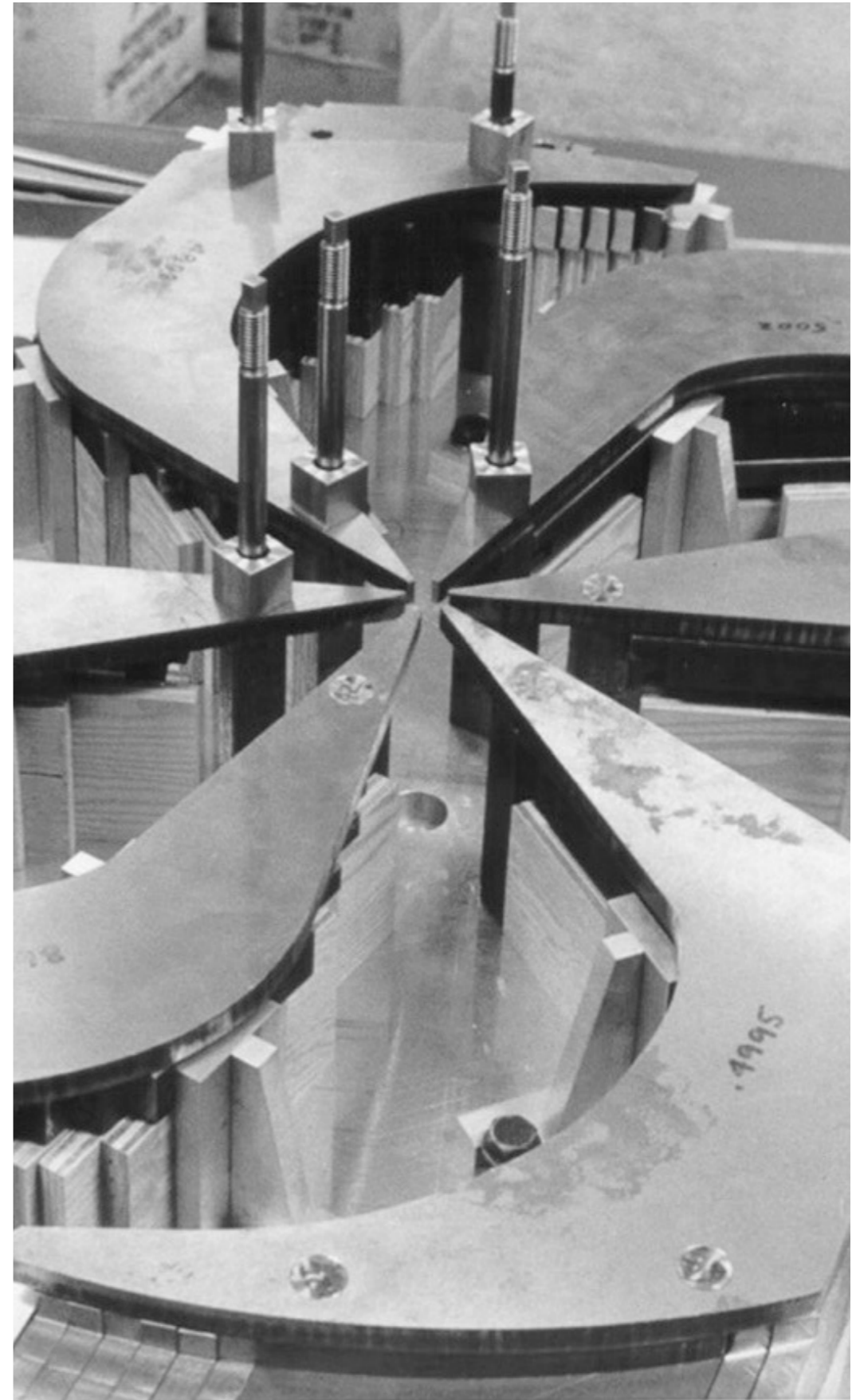


UNIVERSITY OF  
TORONTO

# Radioactive Molecules

Stephan Malbrunot-Ettenauer  
TRIUMF, University of Toronto

PSD BAE Retreat 2024



Discovery,  
accelerated

# '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

**radioactive molecules**

**Applied science**  
•nuclear engineering  
•medicine

**Astrophysics**

**Quantum Chemistry**

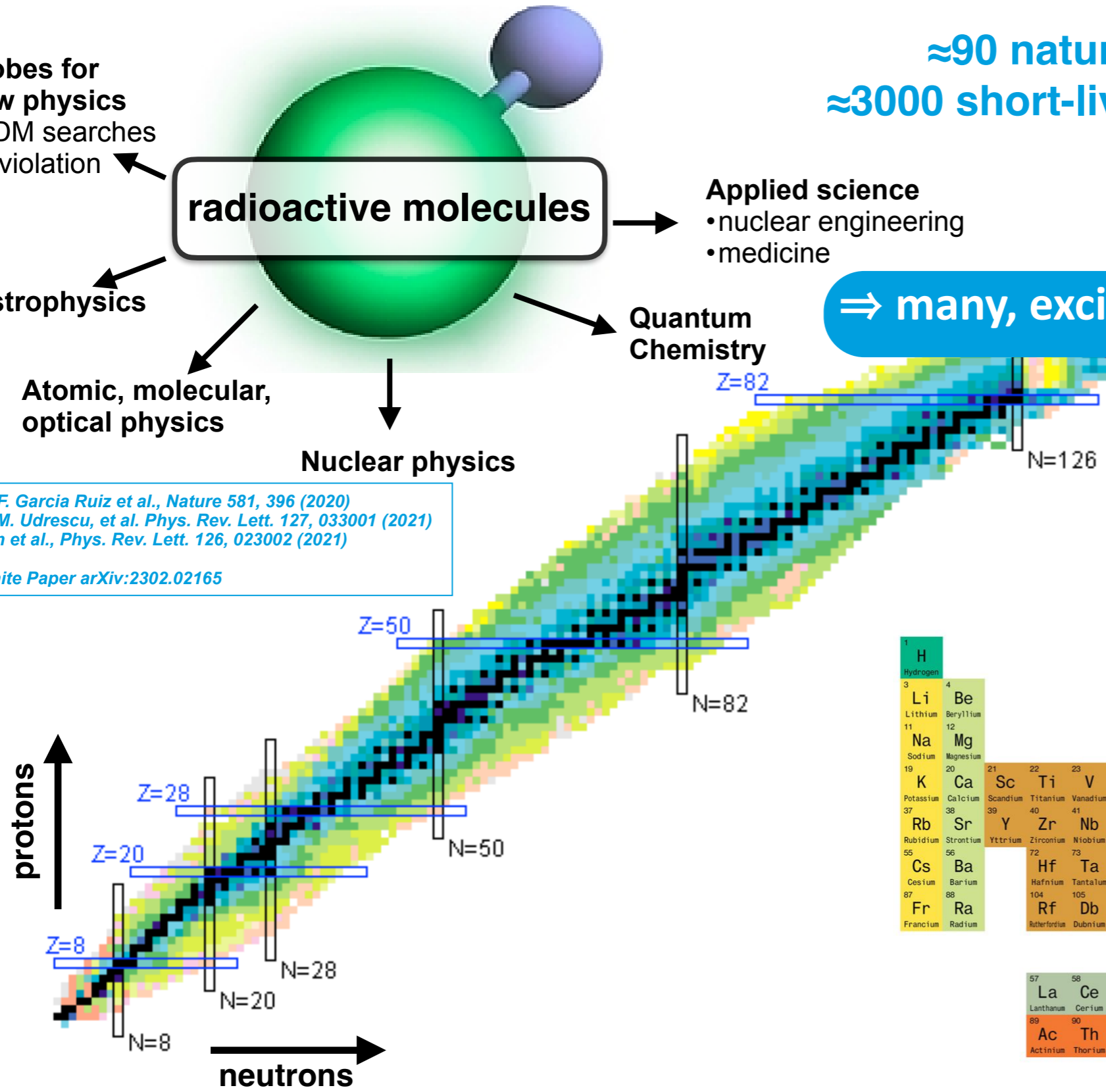
⇒ many, exciting science opportunities

**Atomic, molecular, optical physics**

**Nuclear physics**

*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 (2021)*

White Paper arXiv:2302.02165



**Table of Elements**  
80 chemical elements  
(with stable nuclides)

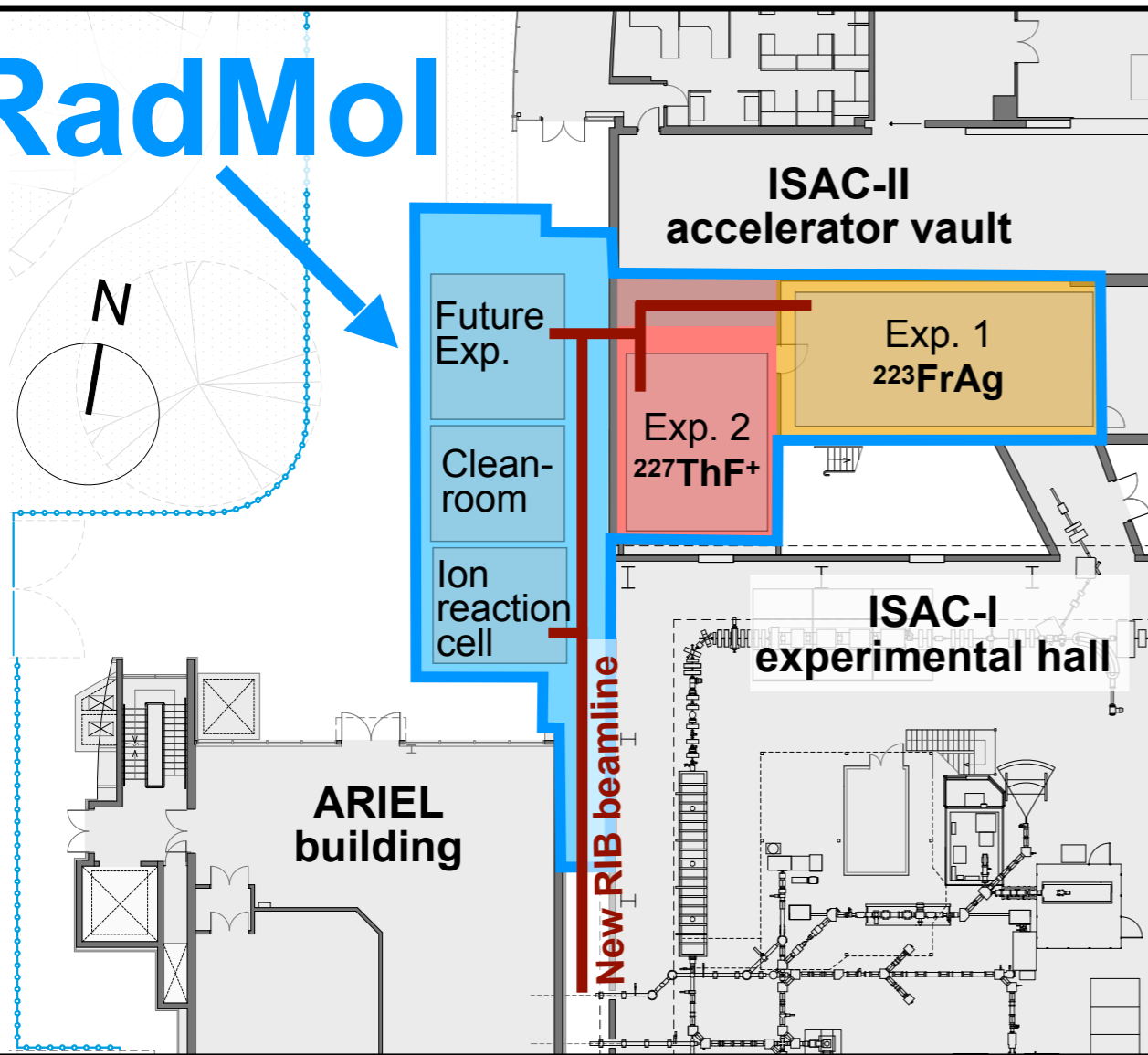
1 H Hydrogen																	2 He Helium
3 Li Lithium	4 Be Beryllium											5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
11 Na Sodium	12 Mg Magnesium											13 Al Aluminum	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
19 K Potassium	20 Ca Calcium	21 Sc Scandium	22 Ti Titanium	23 V Vanadium	24 Cr Chromium	25 Mn Manganese	26 Fe Iron	27 Co Cobalt	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
37 Rb Rubidium	38 Sr Strontium	39 Y Yttrium	40 Zr Zirconium	41 Nb Niobium	42 Mo Molybdenum	43 Tc Technetium	44 Ru Ruthenium	45 Rh Rhodium	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
55 Cs Cesium	56 Ba Barium	57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium	86 Rn Radon
87 Fr Francium	88 Ra Radium	89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium	118 Og Oganesson

57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium
89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium

# RadMol

*a radioactive molecule lab for fundamental physics*

## RadMol



### Goal:

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

### TRIUMF advantages:

- large variety in radioactive ion beams (RIB)
- high beamtime availability (3 independent RIBs)
- existing laboratory space for large, multi-station program

### Current Canadian Team:

- 12 faculty and staff physicists

### RadMol Collaboration:





# ‘Designer Molecules’

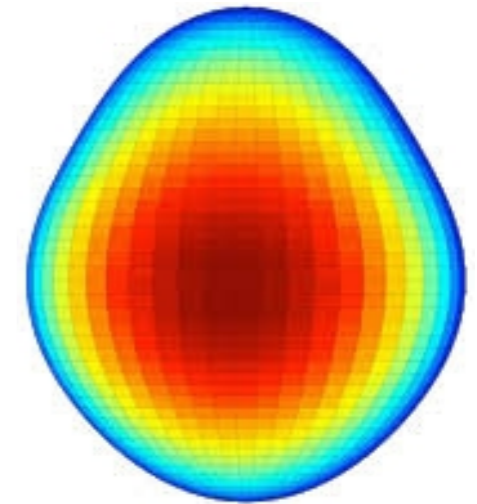
... for searches for time-reversal violation in atomic nuclei

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

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

$$|S_{\text{Hg}}| < 3.1 \cdot 10^{-13} \text{ e fm}^3$$

*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
  - in atom or ion trap x 1,000 compared to beam experiments
- } compared to  $^{199}\text{Hg}$

all known cases in radionuclides



# 'Designer Molecules'

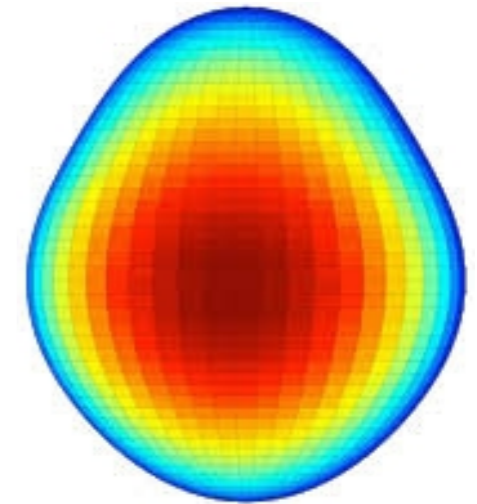
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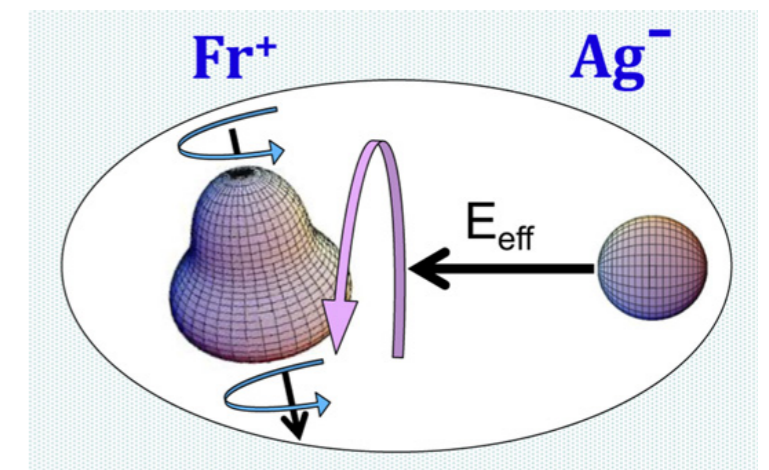
Example:  $^{223}\text{FrAg}$

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

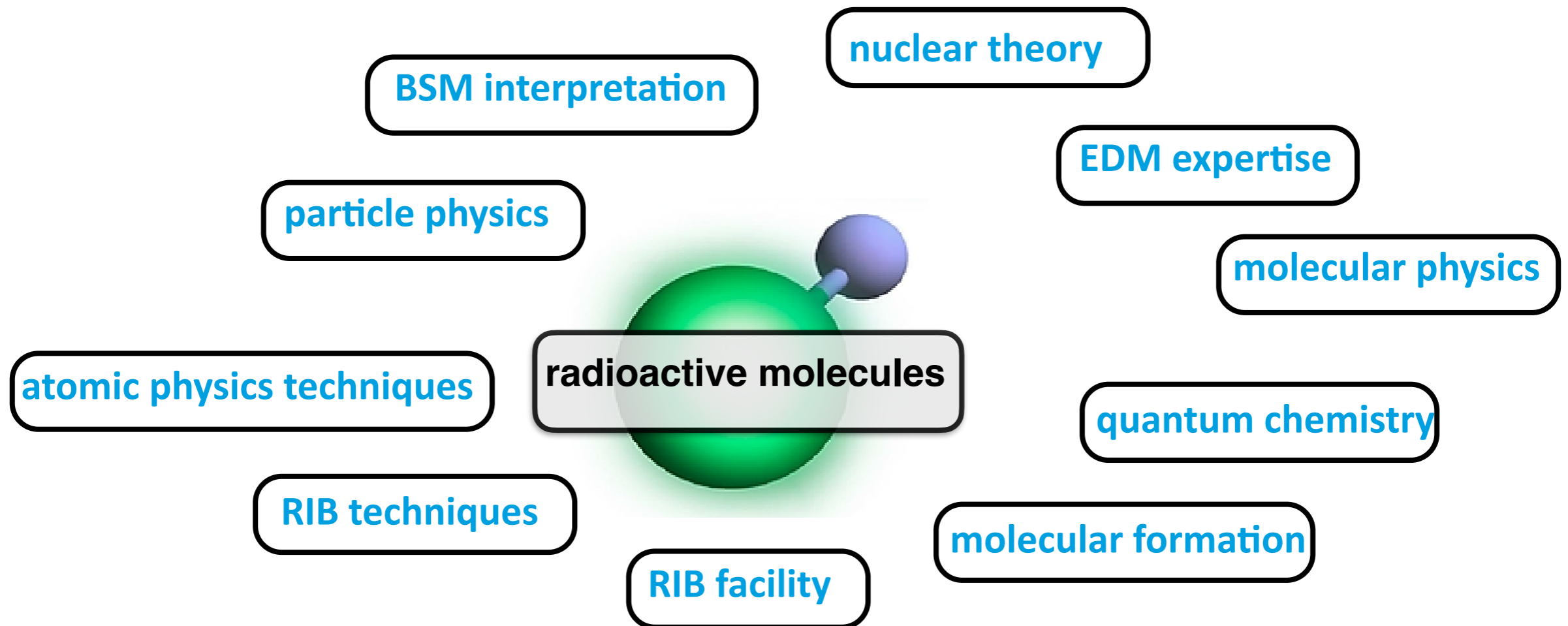
*V. V. Flambaum and V. A. Dzuba. Phys. Rev. A 101, 042504 (2020)*  
*T. Fleig. private communications with D. DeMille (2022)*

- need to be produced at TRIUMF  
    ➔ challenge: reduced availability

- **anticipated gain: x 1,000 for certain CPV-parameters (comp to  $^{199}\text{Hg}$ )**



# Multidisciplinary



## General theme for experiments:

translate high-precision AMO techniques into accelerator lab

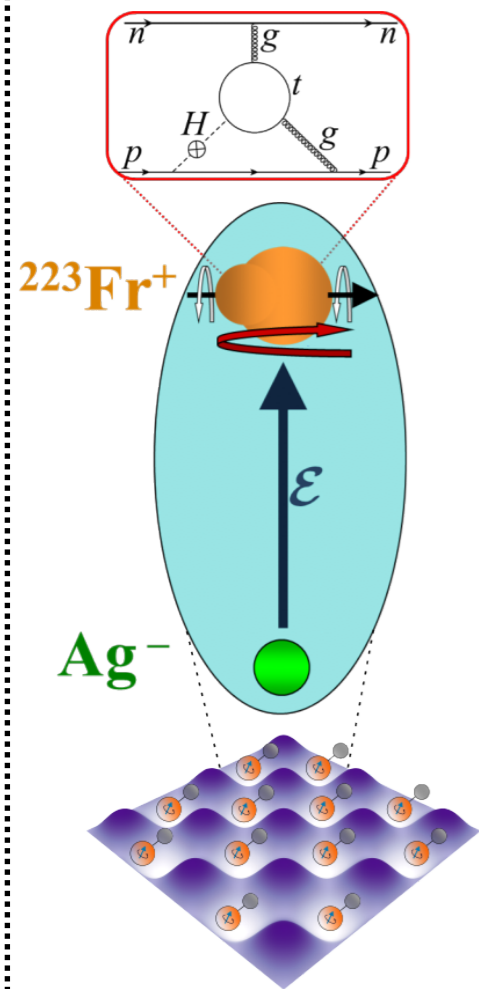
# RadMol collaboration

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

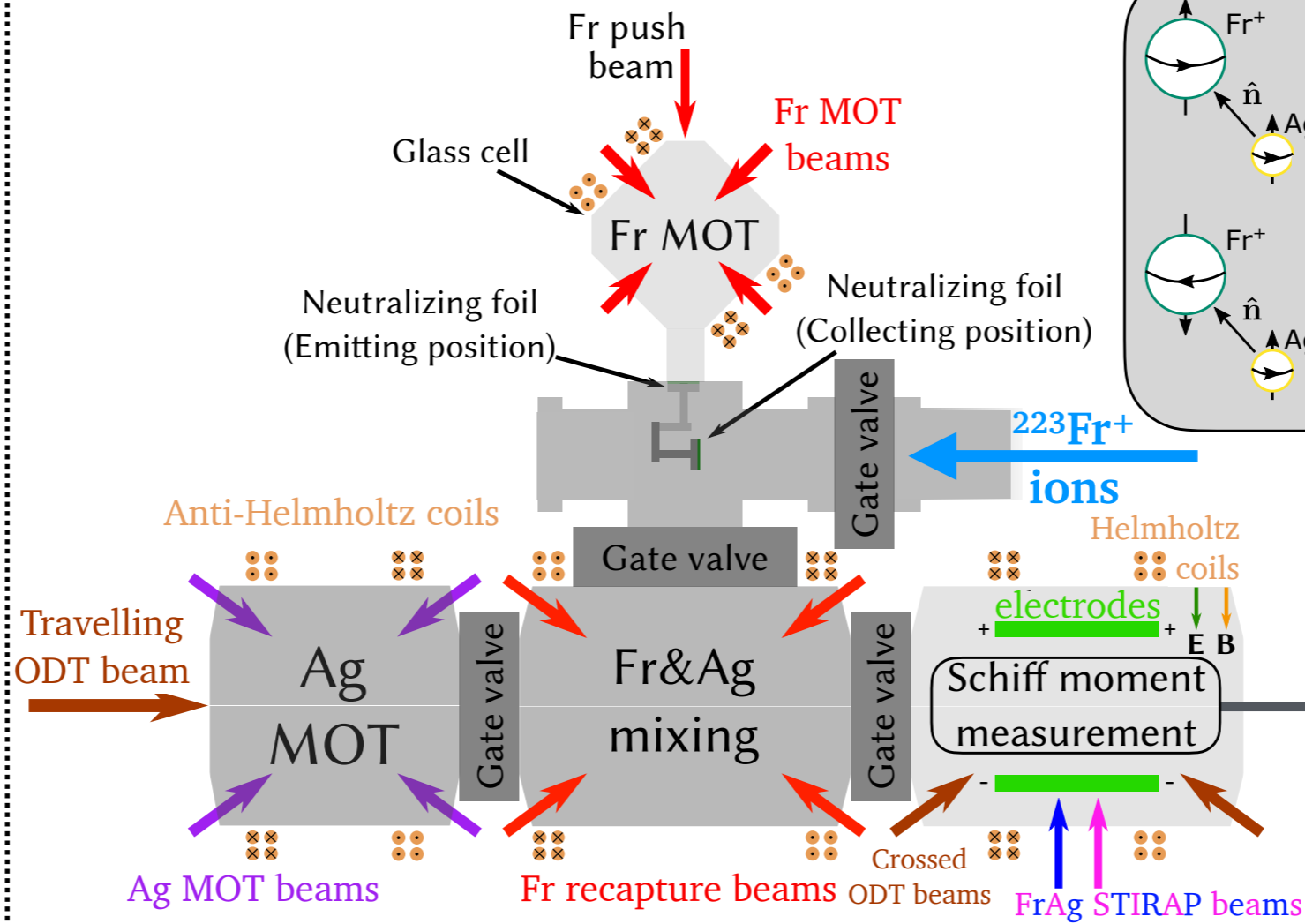


# $^{223}\text{FrAg}$ experiment

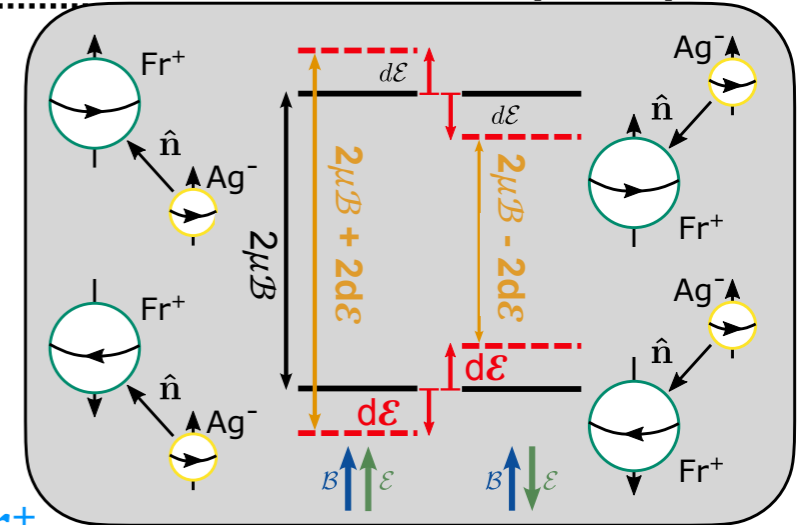
## concept



## experimental apparatus

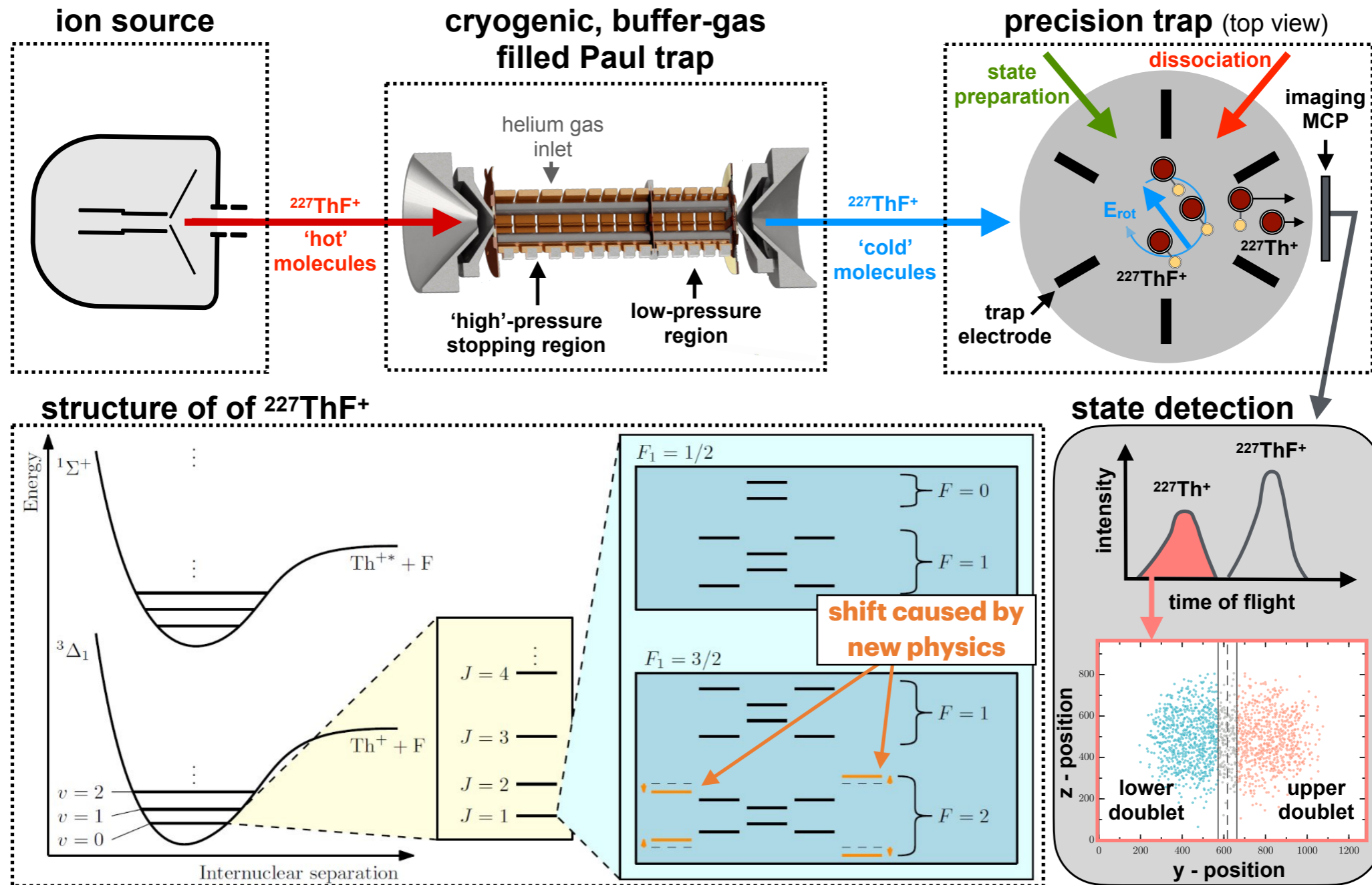


## EDM measurement principle



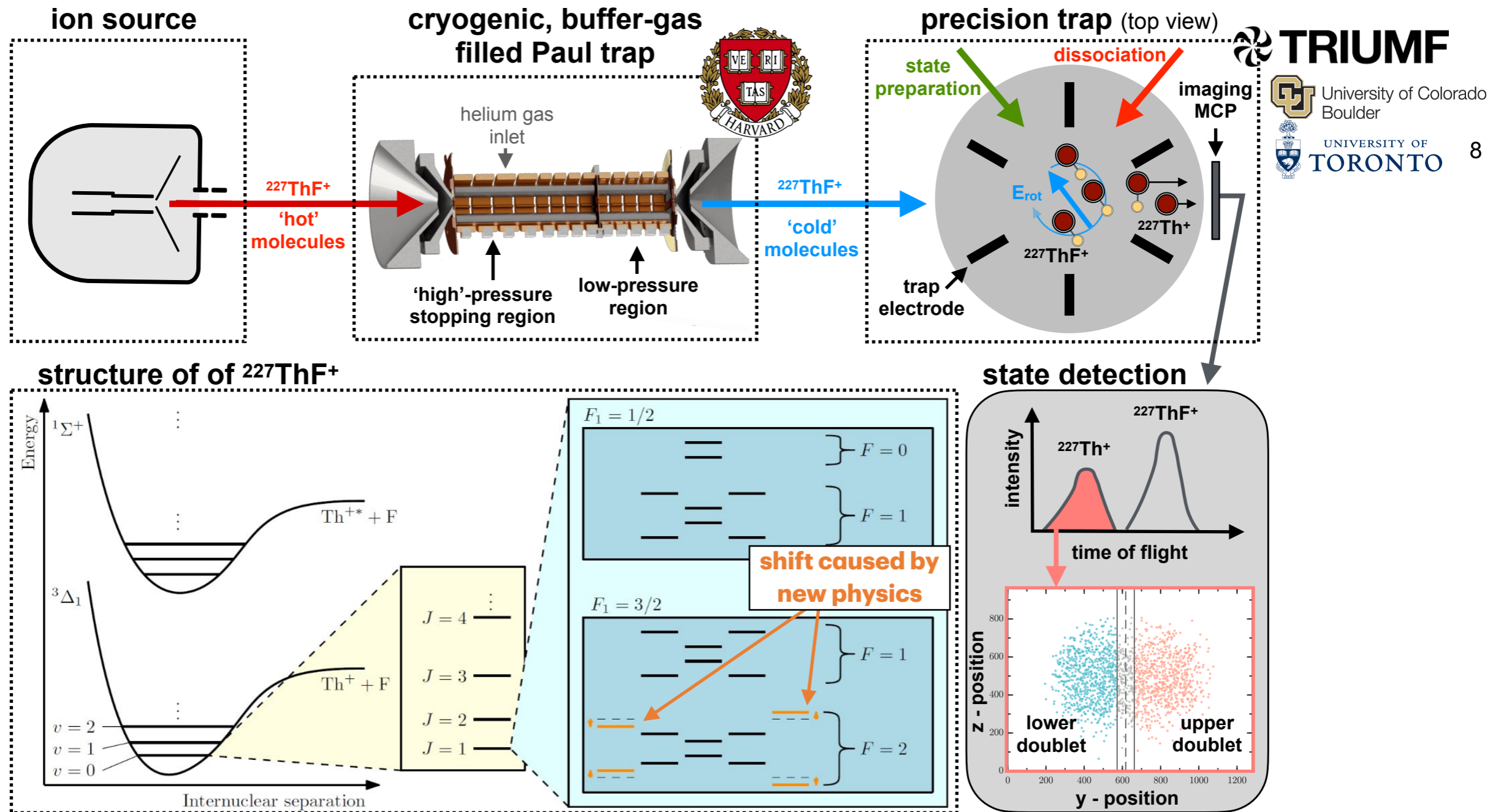
- led by Chicago (DeMille)
- benefits from Fr trapping knowhow at TRIUMF/Manitoba
- obtained 2.8 MUSD grant by Moore foundation (ca. 400 kCAD directly to TRIUMF)
- $^{223}\text{Fr}$ 's half-life 23 min: high-intensity online access + low-intensity offline source ( $^{227}\text{Ac}$ )

# $^{227}\text{ThF}^+$ experiment



- experimental EDM techniques analogous to JILA electron EDM experiment
- molecular structure of  $^{232}\text{ThF}^+$  known from spectroscopy at JILA
- access to  $^{227}\text{Th}$  via  $^{227}\text{Ac}$  sample from life sciences  $\Rightarrow$  strong inter-divisional effort  
(Accelerator, Life Sciences, Physical Sciences)

# $^{227}\text{ThF}^+$ experiment

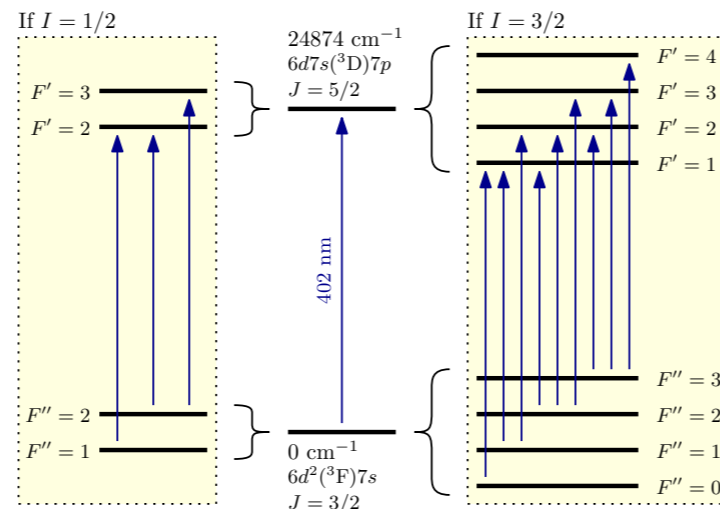


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# 1st step: laser spectroscopy of $^{227}\text{Th}^+$

**Goal:** measure nuclear spin & magnetic moment of  $^{227}\text{Th}^+$



9

**Challenge:** Th beams at ISAC (and globally)

**Solution:**

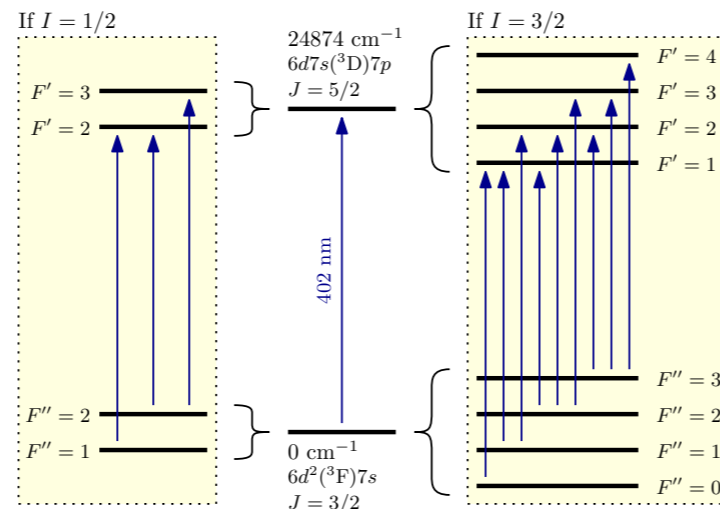
- $^{227}\text{Ac}$  sample in Life Sciences: generator of  $^{227}\text{Th}$  via chemical separation
- form 'offline' beam in ISAC target
- mass separation in ISAC magnetic mass separator
- laser spectroscopy in polarizer line (using TITAN buncher)

**Competition on  $^{227}\text{Th}^+$ :**

- TRIUMF has leading edge
- others will follow, e.g. Pa LOI at ISOLDE on similar concept

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- mass separation in ISAC magnetic mass separator
- laser spectroscopy in polarizer line (using TITAN buncher)

Vision for future RadMol laboratory:

Infrastructure for chemical and mass separation for fundamental physics and medical isotope research

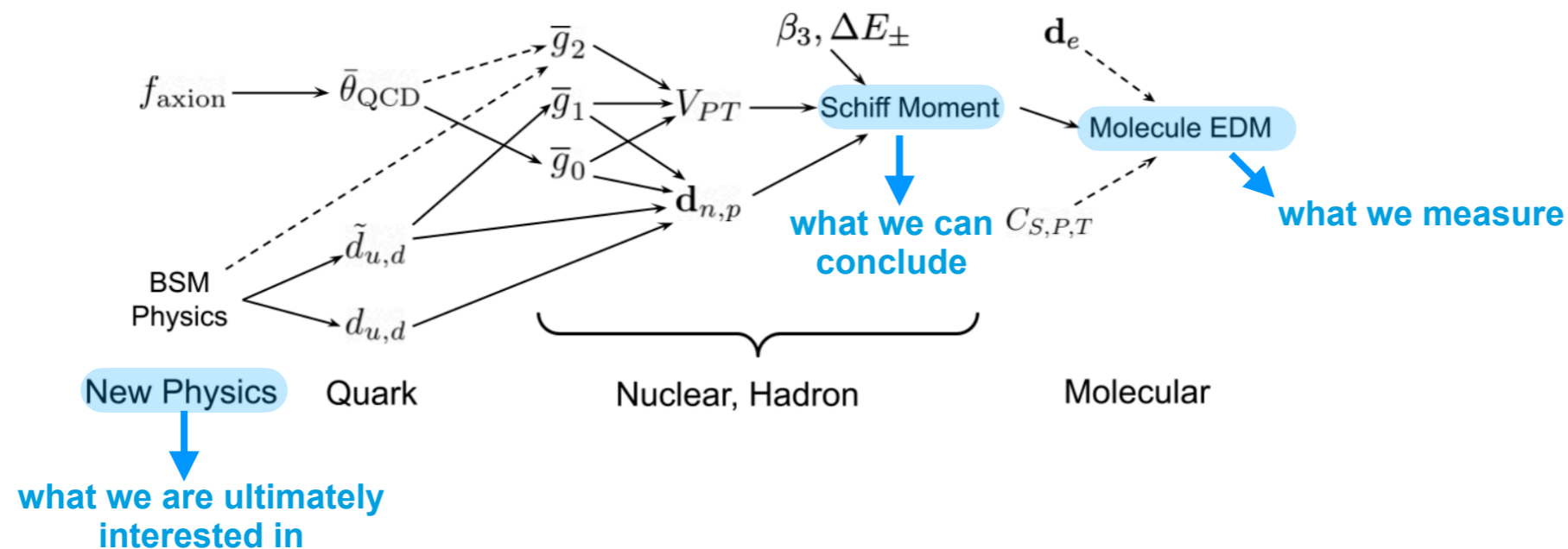
**Competition on  $^{227}\text{Th}^+$ :**

- TRIUMF has leading edge
- others will follow, e.g. Pa LOI at ISOLDE on similar concept

# why 2 experiments?

- 2 complementary techniques: neutral atoms  $\Leftrightarrow$  singly charged ions
- different systematics
- different sensitivity to underlying physics

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- TRIUMF's attractiveness to this new, emerging field
  - ➔ beam availability & strong beams (for Fr) or unique sample (for Th)
  - ➔ in-house expertise in sample and beam preparations
  - ➔ existing expertise (FrPNC, TRINAT, TITAN, laser, target and ion sources)
  - ➔ existing laboratory space  $\Rightarrow$  experimental work has already started

**S2068LOI - S2171LOI S2279LOI - S2309 -**

- FrAg apparatus: unique approach  $\Leftrightarrow$  ThF<sup>+</sup> apparatus: 'universal' spectroscopy setup



# spectroscopy of other ionic molecules

## Examples for other radioactive molecules:

RaOCH<sub>3</sub><sup>+</sup>

Fan et al., Phys. Rev. Lett. 126, 023002 (2021)  
Yu and Hutzler, Phys. Rev. Lett. 126, 023003 (2021)

PaF<sup>3+</sup>

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

AcO<sup>+</sup>

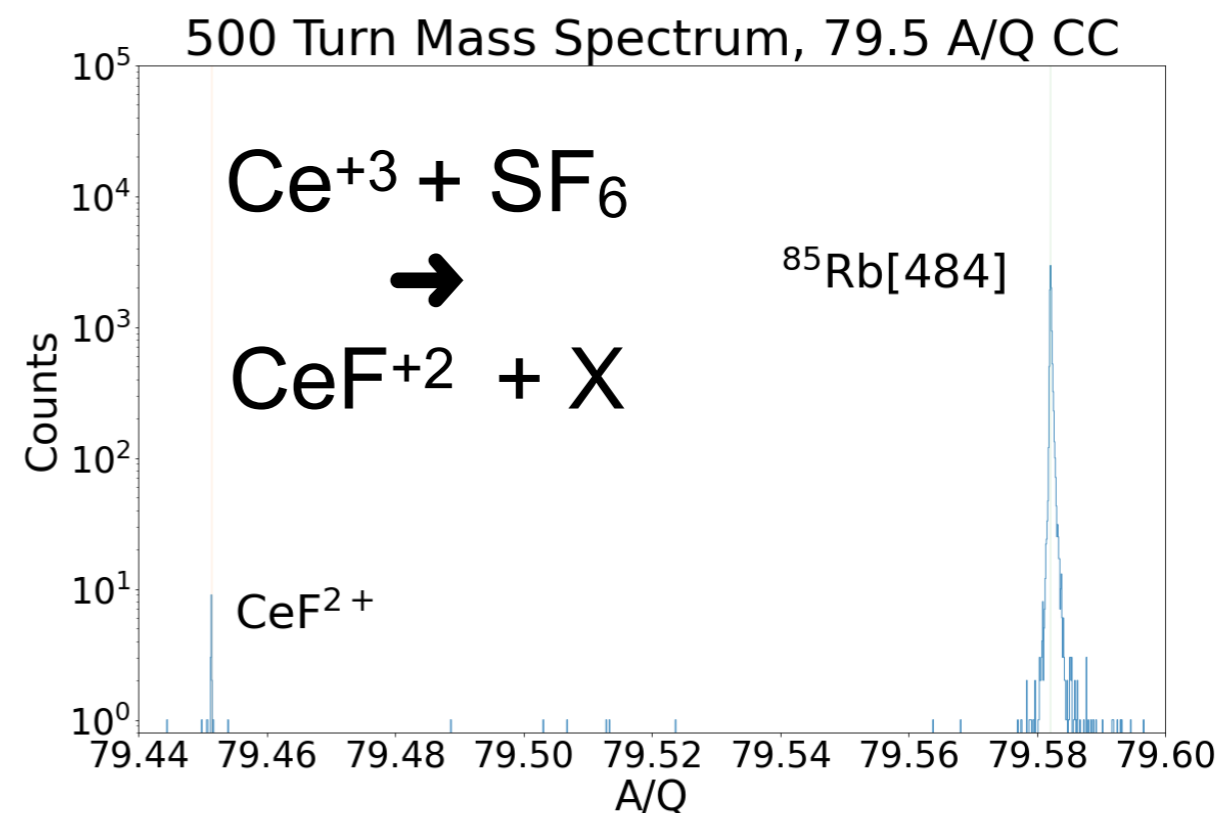
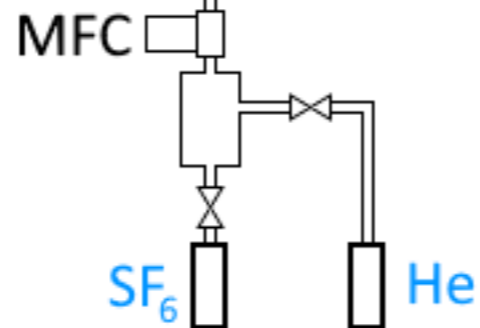
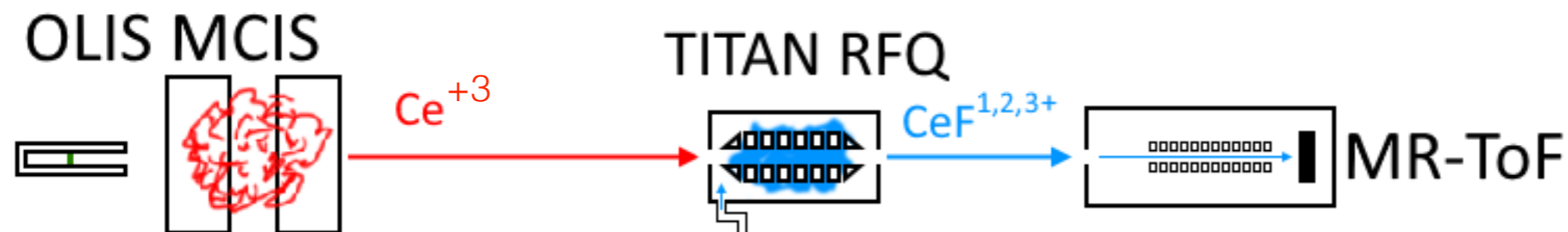
Flambaum and V. A. Dzuba, Phys. Rev. A 101, 042504 (2020).

CeF<sup>+2</sup>, AcF<sup>+</sup>

R. Berger, private communications

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## First step to spectroscopy: Molecular Formation / Present



Manuscript (almost) completed

Successful collaboration between  
Accelerator, Life Sciences, Physical  
Sciences Divisions

# spectroscopy of other ionic molecules

## Examples for other radioactive molecules:



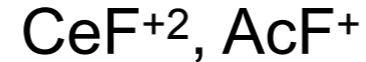
Fan et al., *Phys. Rev. Lett.* 126, 023002 (2021)  
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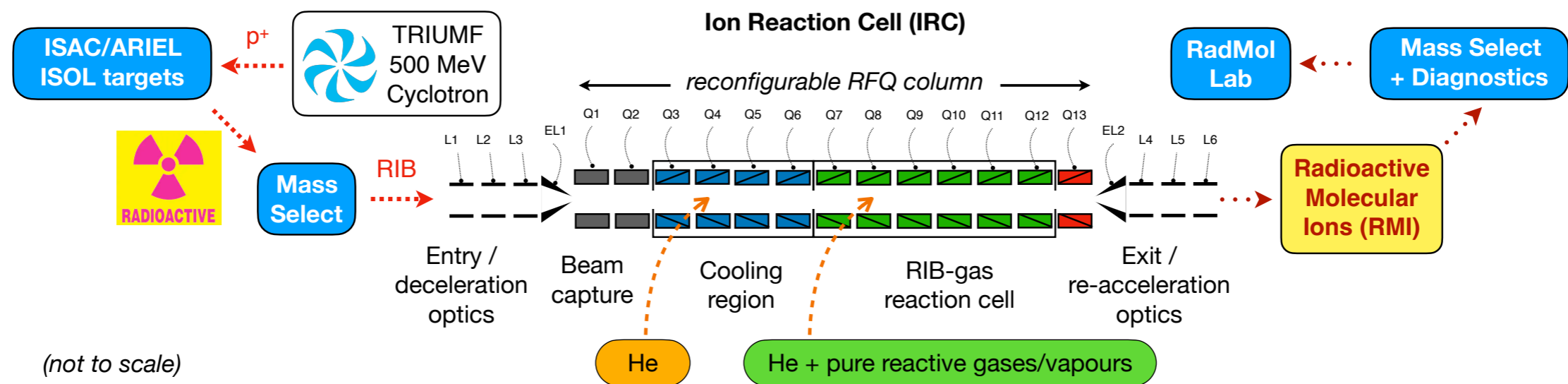


R. Berger, private communications

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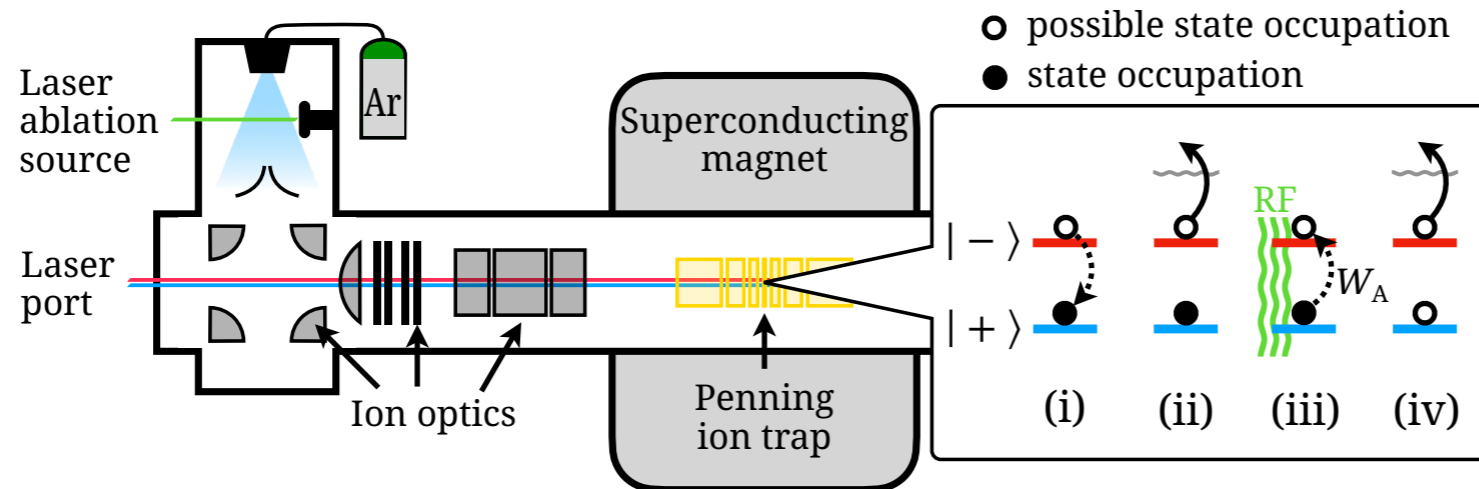
## First step to spectroscopy: Molecular Formation / Future

Dedicated Ion Reaction Cell (led by C. Charles)



# Provisions for future experiments

## Nuclear Anapole Moments

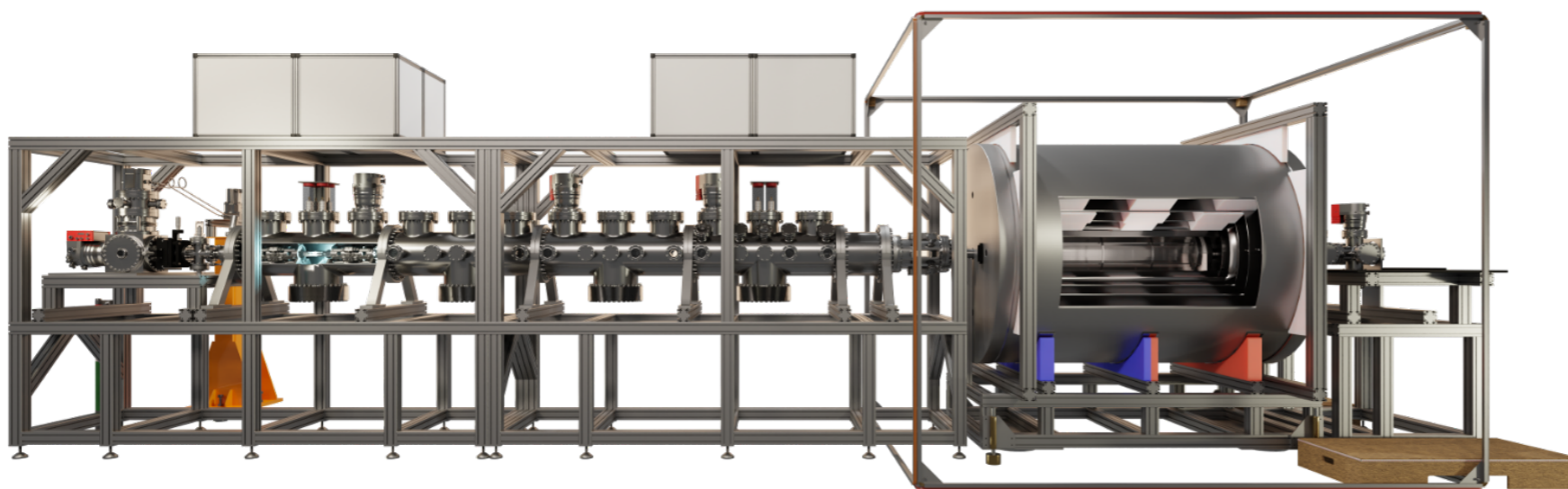


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*Karthein et al., accepted in Phys. Rev. Lett.*

## Spectroscopy of ultra-cold radioactive molecules



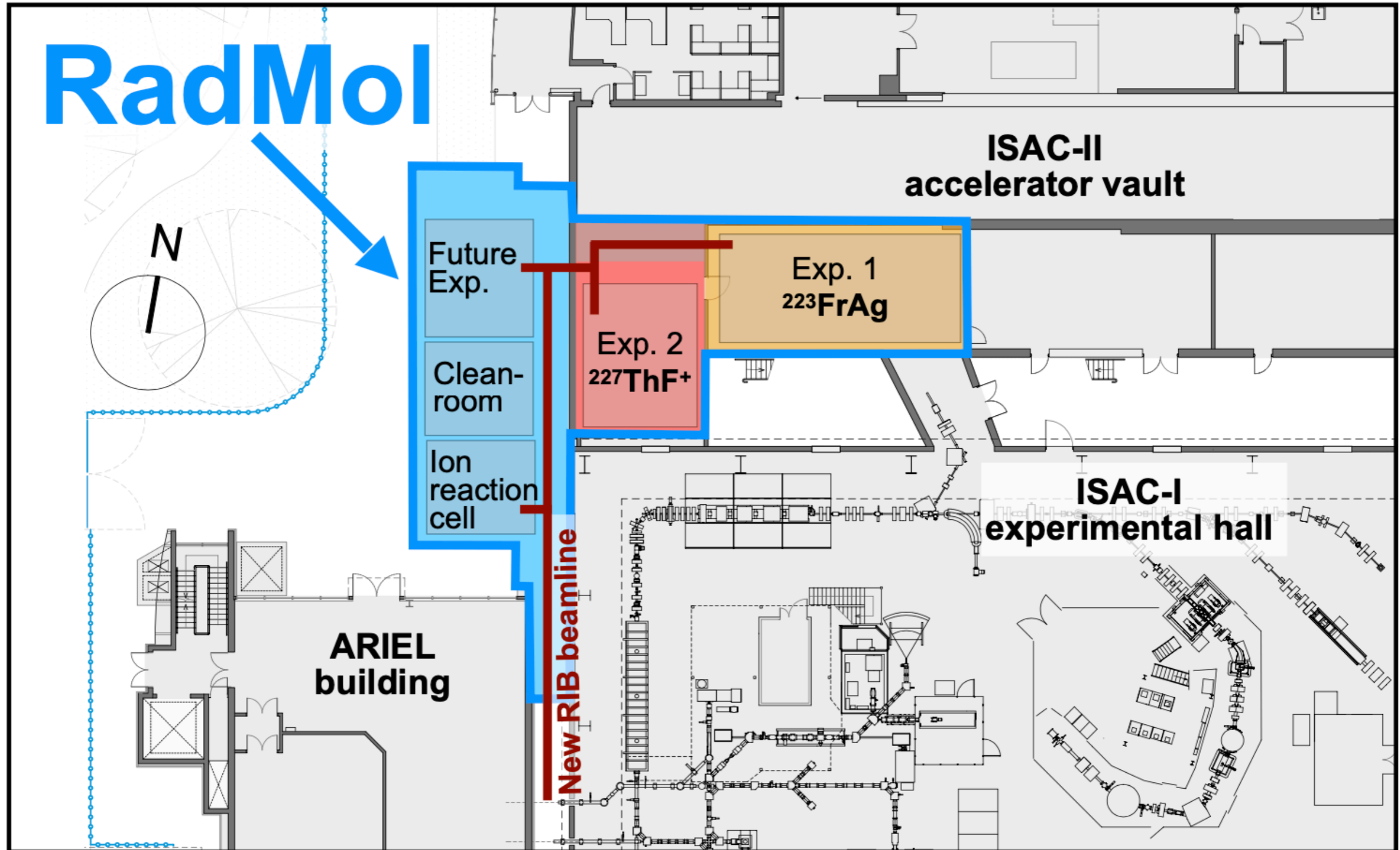
- Quantum chemistry
- Astrophysics
- BSM physics



university of  
 groningen

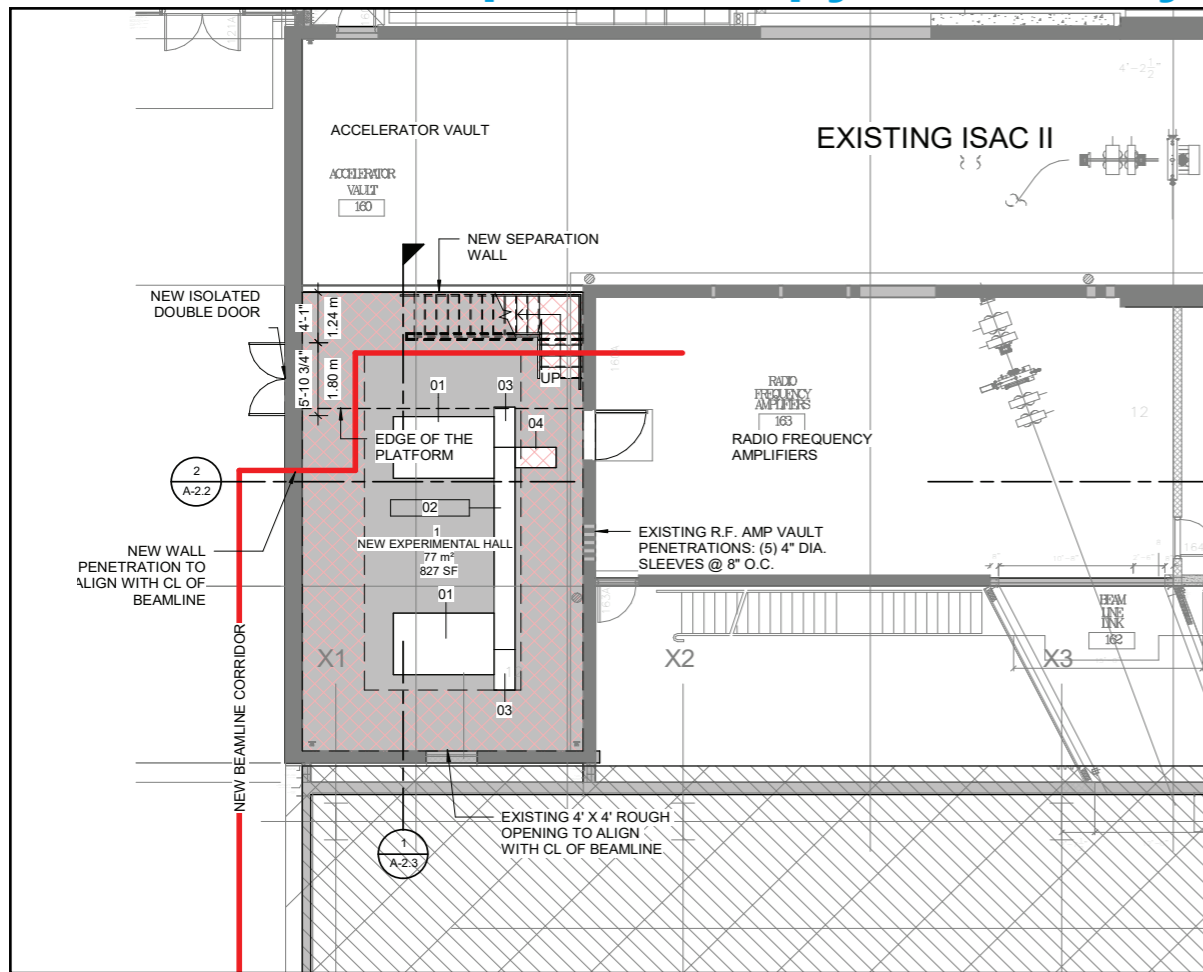
Steven Hoekstra for BaF

# Laboratory Layout



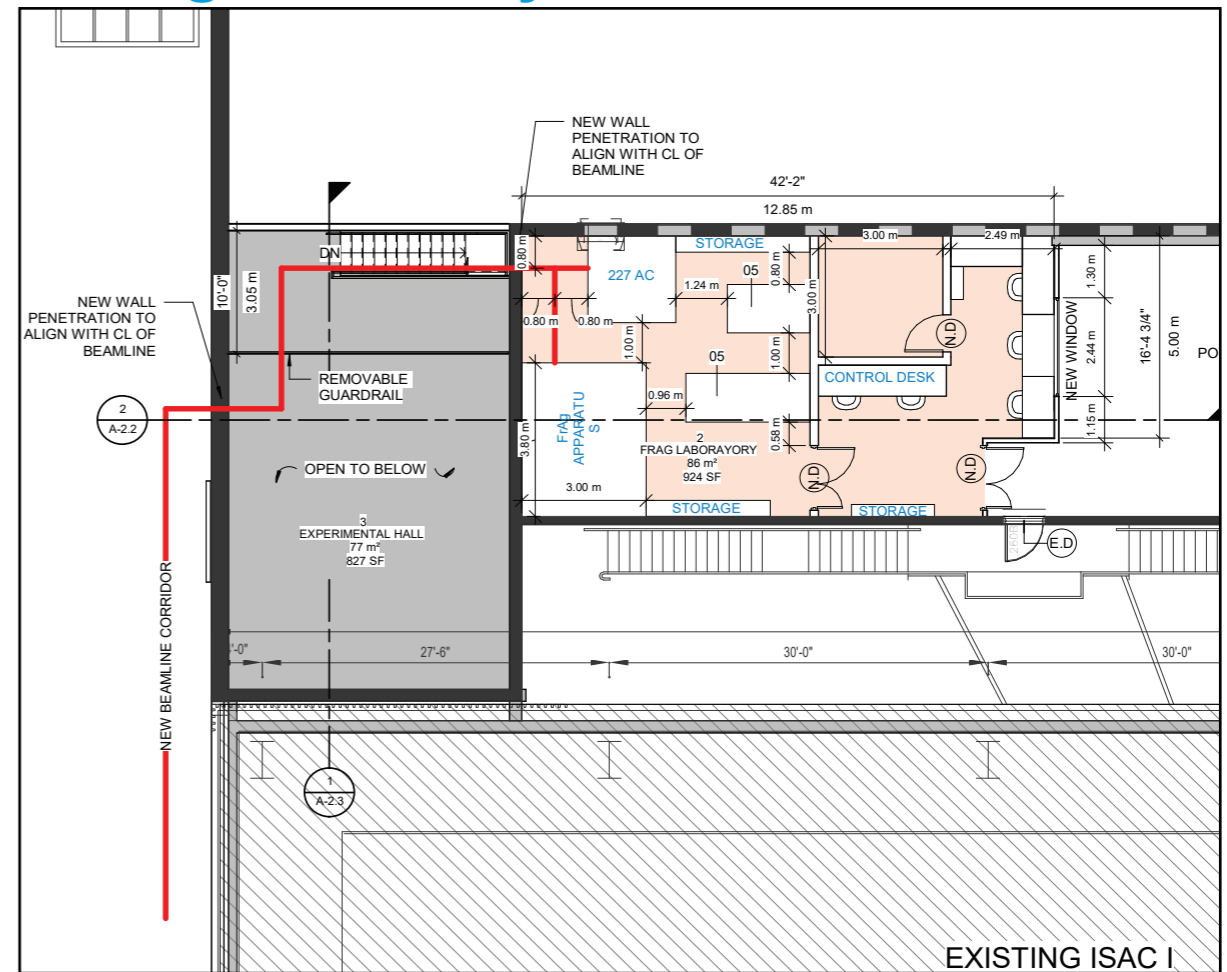
# Renovation of existing laboratory space

## ThF<sup>+</sup> and ion spectroscopy laboratory



Ground floor

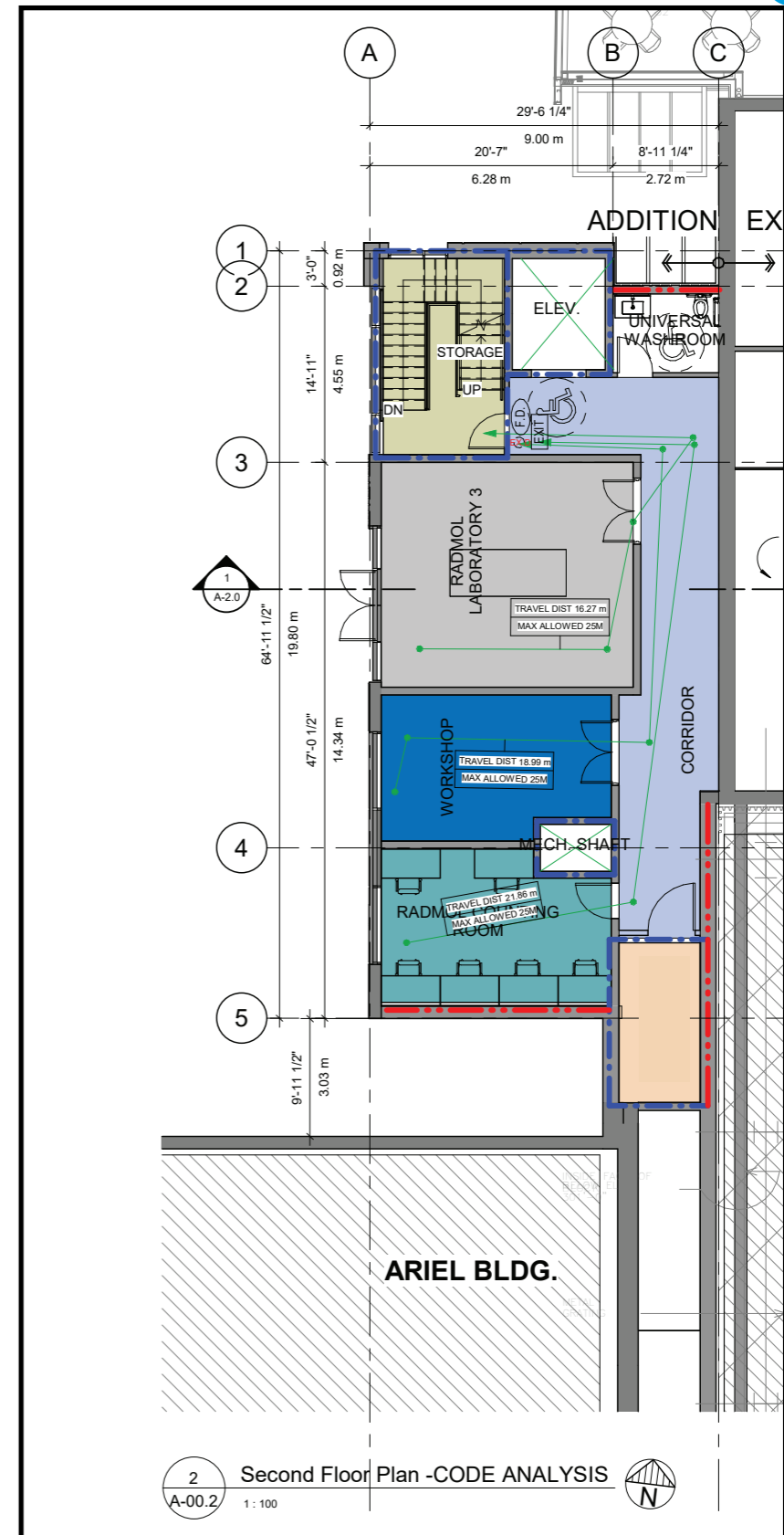
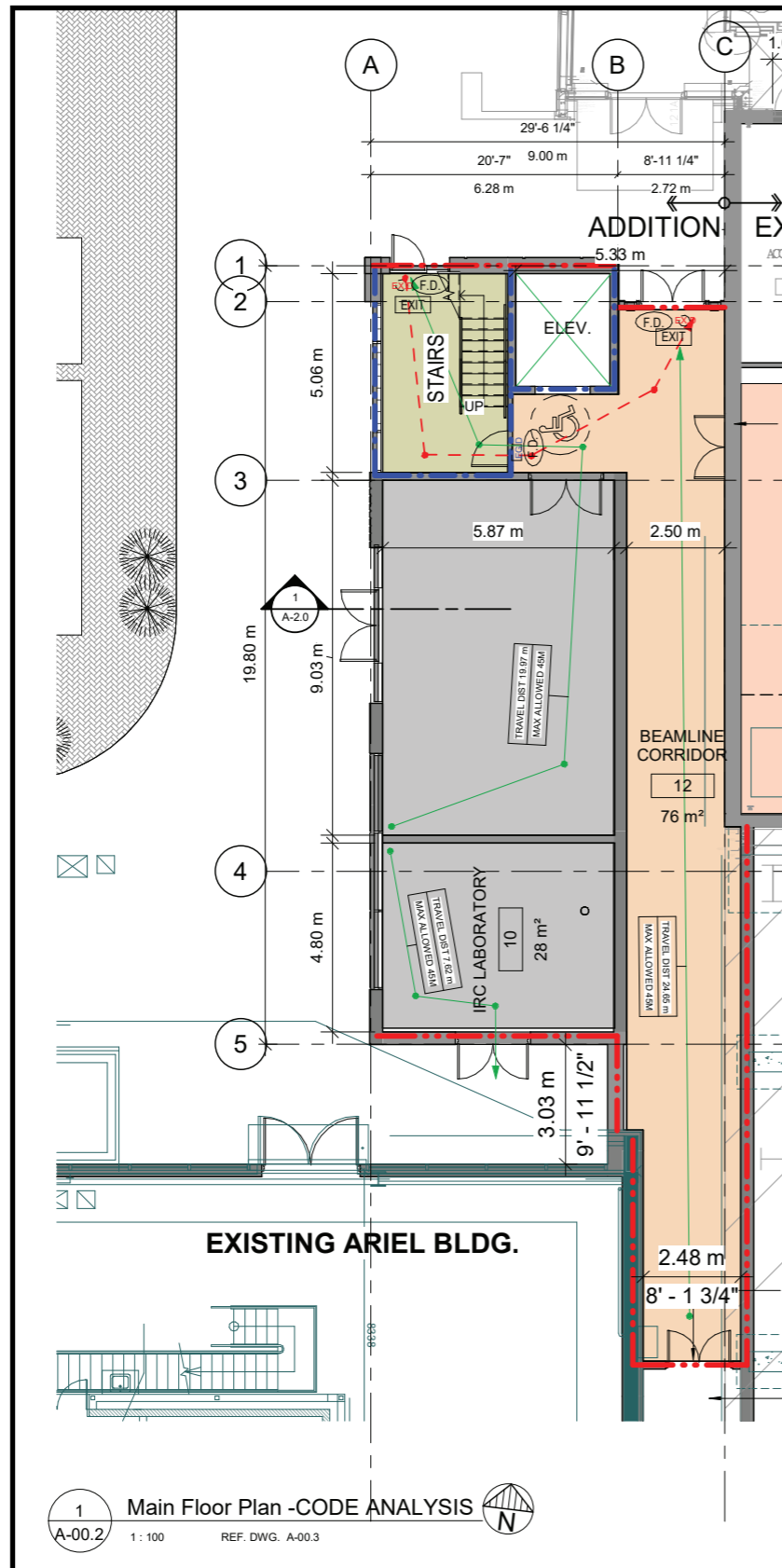
## FrAg laboratory



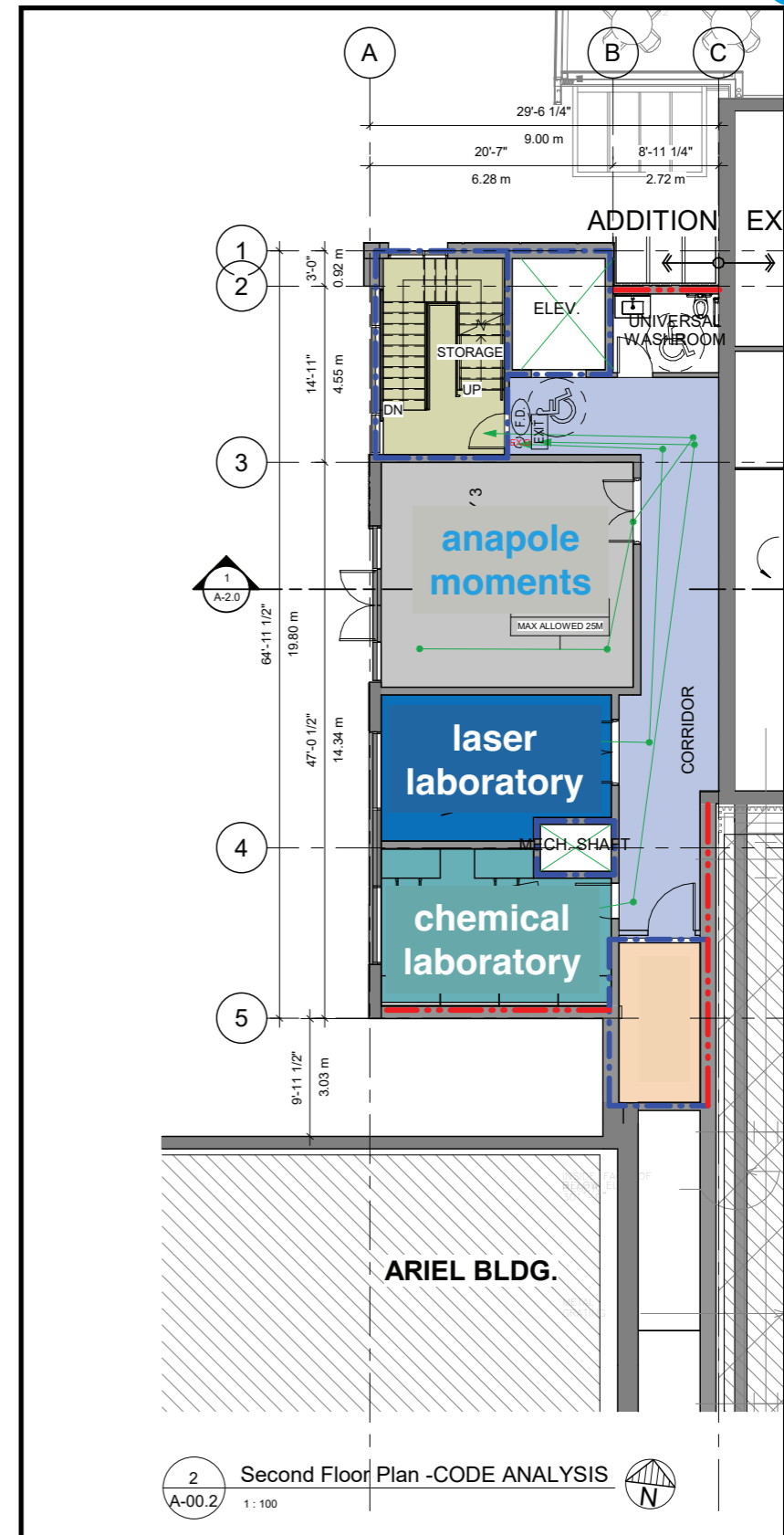
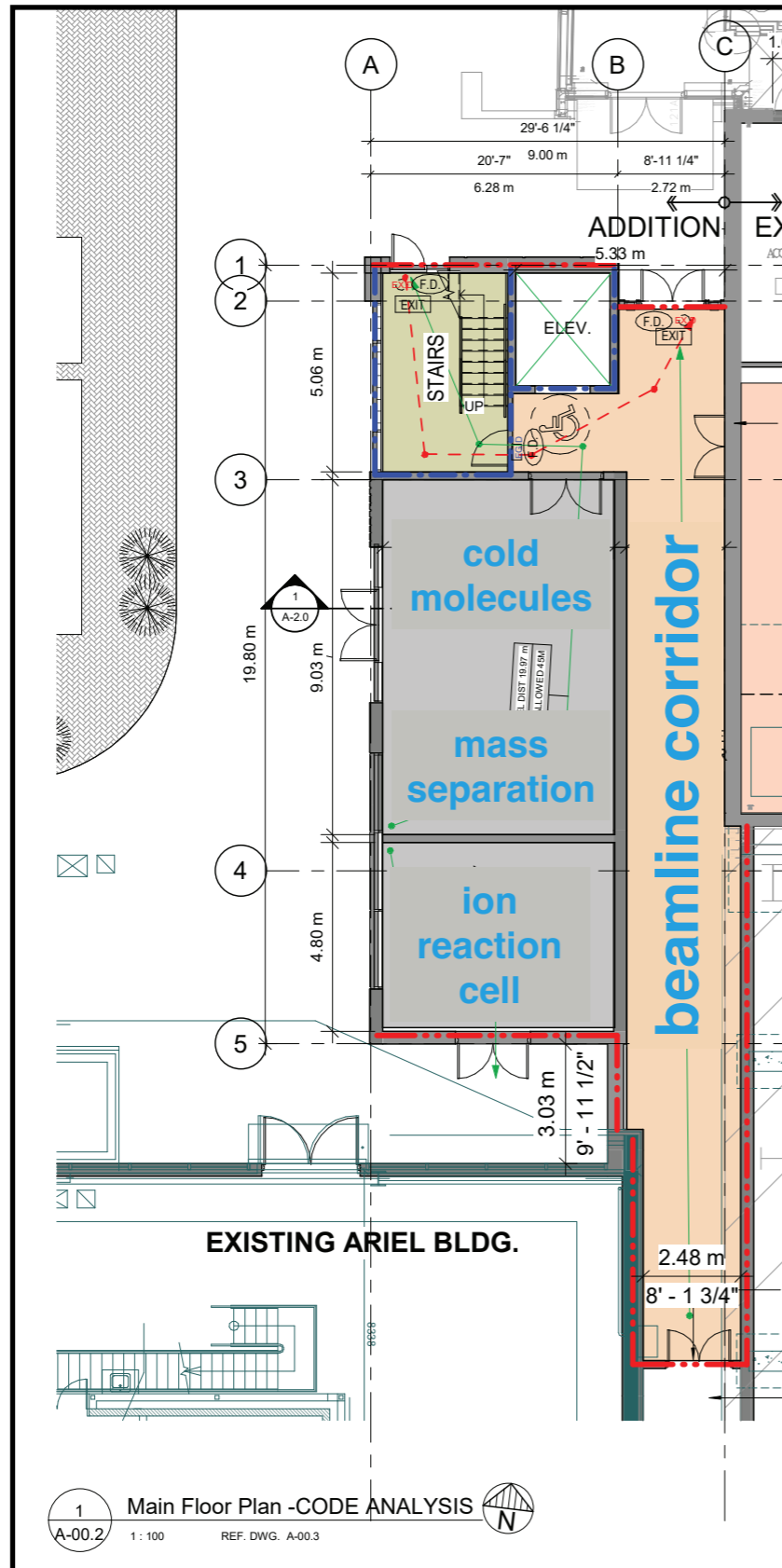
Second Floor



# New ISAC II extension building



# New ISAC II extension building





# New ISAC II extension building



# budget

Infrastructure or equipment requested	Costs [\$MCAD]
renovation of laboratory space	1.2
building extension	10.8
RIB beam-lines	2.5
Ion Reaction Cell & integration	0.5
Experimental Apparatus - $^{223}\text{FrAg}$	3.2
Experimental Apparatus - $^{227}\text{ThF}^+$	2.8
<b>TOTAL</b>	<b>21.0</b>

**12 MCAD**

have to come from BC institutions

Funding	Amount [\$MCAD]
CFI envelope	
UBC	4.0
University of Toronto	3.5
University of Waterloo	0.4
University of Manitoba	0.5
Provincial contributions	
BC	4.0
Ontario	3.9
Manitoba	0.5
Other	
TRIUMF	4.0
Vendor Discounts	0.2
<b>TOTAL</b>	<b>21.0</b>

$\Sigma = 12 \text{ MCAD}$



# Consequences of staging CFI proposal

## Scenario A: full proposal

- leading Schiff moment experiments at TRIUMF
  - attract leading AMO scientists in EDM searches
  - allows exploitation across TRIUMF divisions
  - clear vision for expansion into wider program
  - cement TRIUMF as the place for radioactive molecules
  - required TRIUMF investment: 4 MCAD
- ion reaction cell  
Mass separation capabilities for medical isotopes  
beamline for new RFQ accelerator in ISAC-II

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## Scenario B: renovation + ThF<sup>+</sup> + FrAg

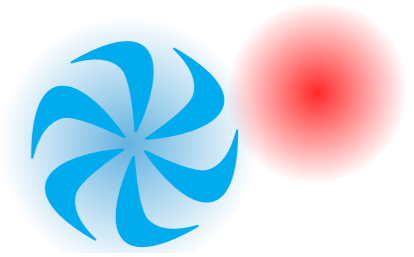
- required TRIUMF investment: 1.2 MCAD
- Synergy in source development between ThF<sup>+</sup> and FrAg (both <sup>227</sup>Ac based)
- ThF<sup>+</sup> in full science program, but no general spectroscopy program
- FrAg only with offline source: why at TRIUMF?
- prospect for later online beam: earliest in 2032 ⇒ not compatible with AMO schedules
- Limited mutual benefits across divisions (while collaboration remains essential)

## Scenario C: 1/2 renovation + ThF<sup>+</sup>

- required TRIUMF investment: 0.6 MCAD (?)
- only ThF<sup>+</sup> in full science program, but no general spectroscopy program
- loss of major collaborators - including UBC
- difficult to argue why UBC is CFI lead



# 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**
  - ➔ requires multidisciplinary approach & technical developments
  - ➔ strong benefits for collaboration across divisions
  - ➔ attract leading AMO scientists to TRIUMF
  - ➔ TRIUMF is place of choice (if we proceed full scale now)

# RadMol Collaboration:



Thank you  
Merci

[www.triumf.ca](http://www.triumf.ca)

Follow us **@TRIUMFLab**

