

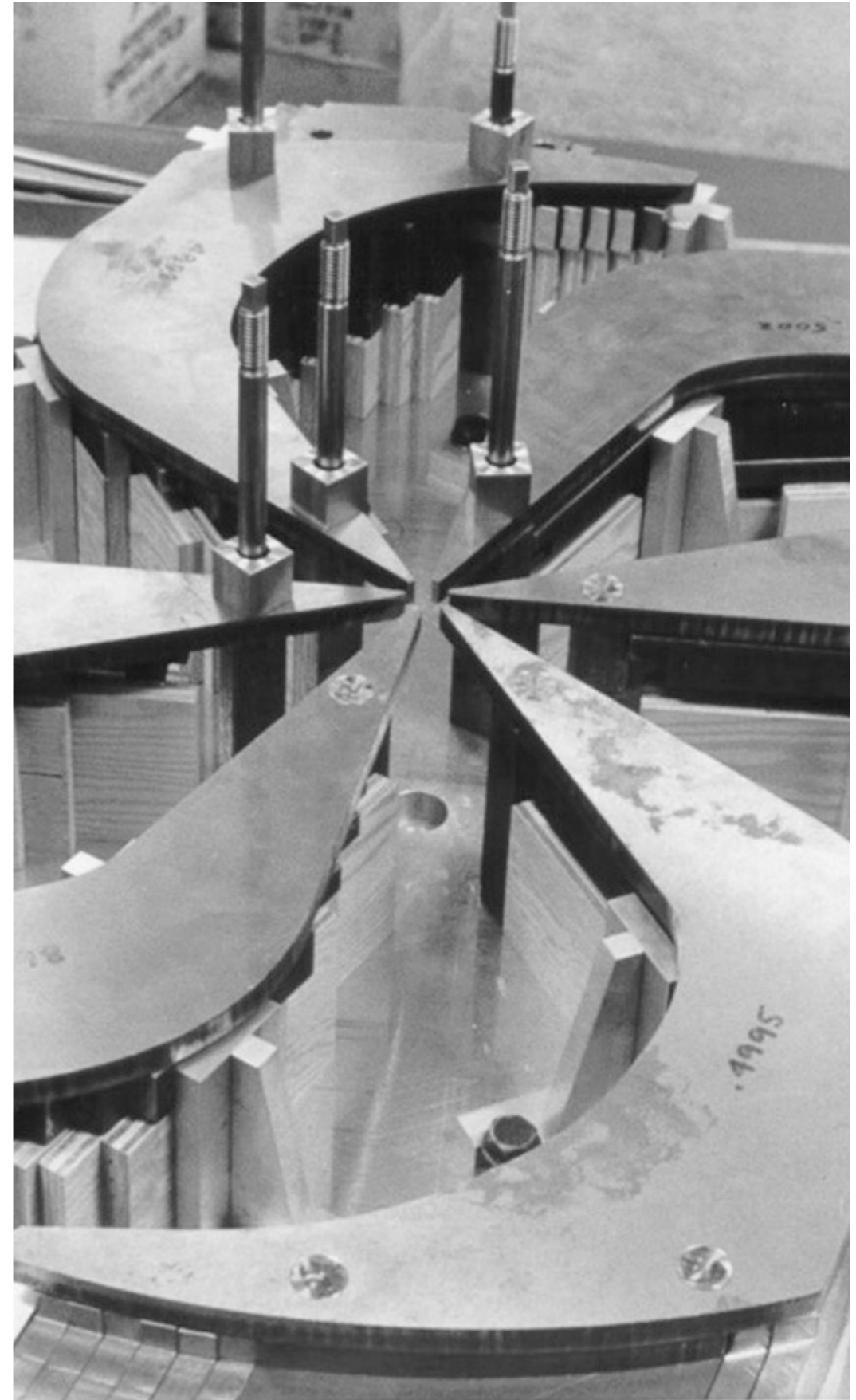


UNIVERSITY OF  
TORONTO

# Radioactive Molecules at TRIUMF

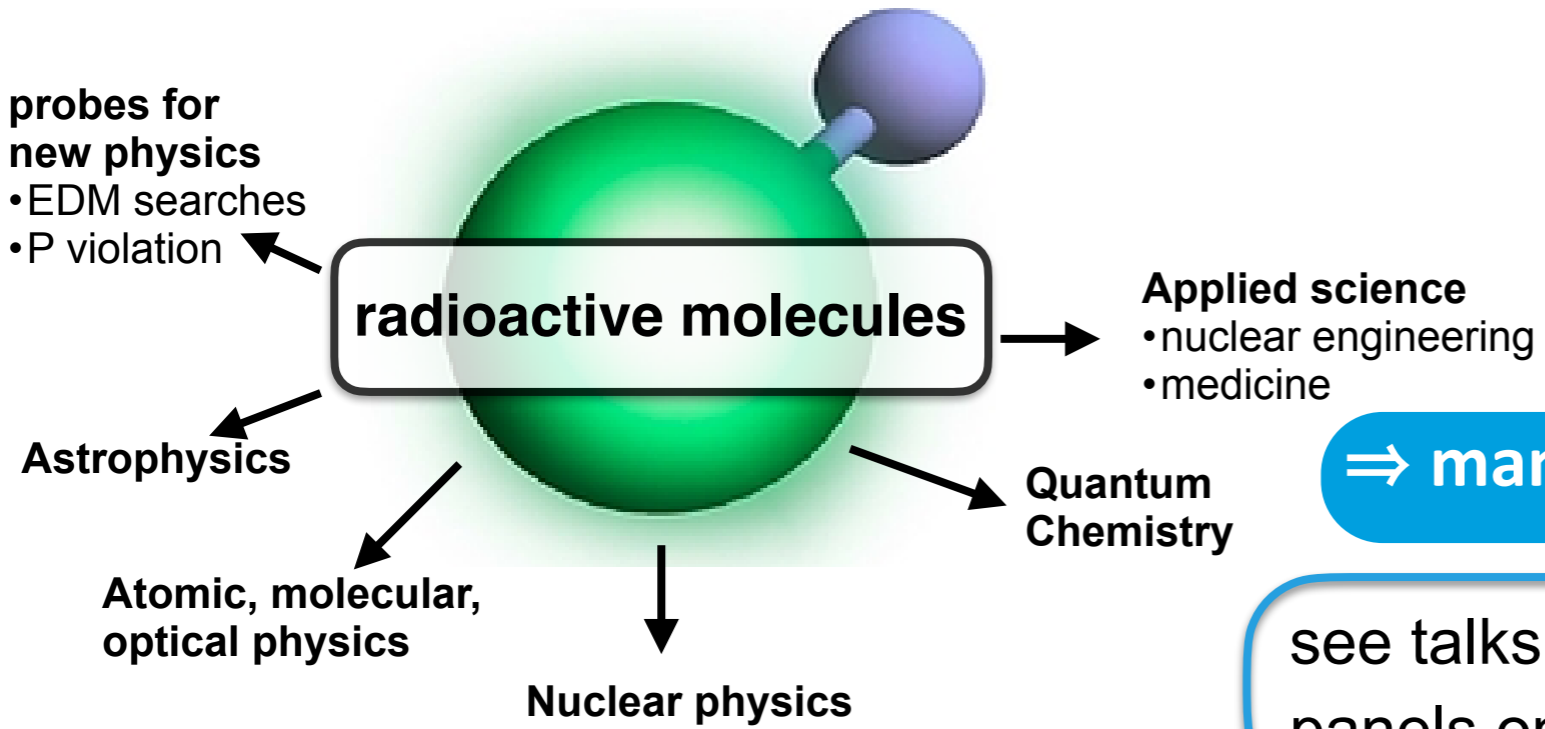
Stephan Malbrunot-Ettenauer  
TRIUMF, University of Toronto

ARIEL Workshop  
CNRS-TRIUMF IRL  
April 2026



Discovery,  
accelerated

# 'Designer Molecules'

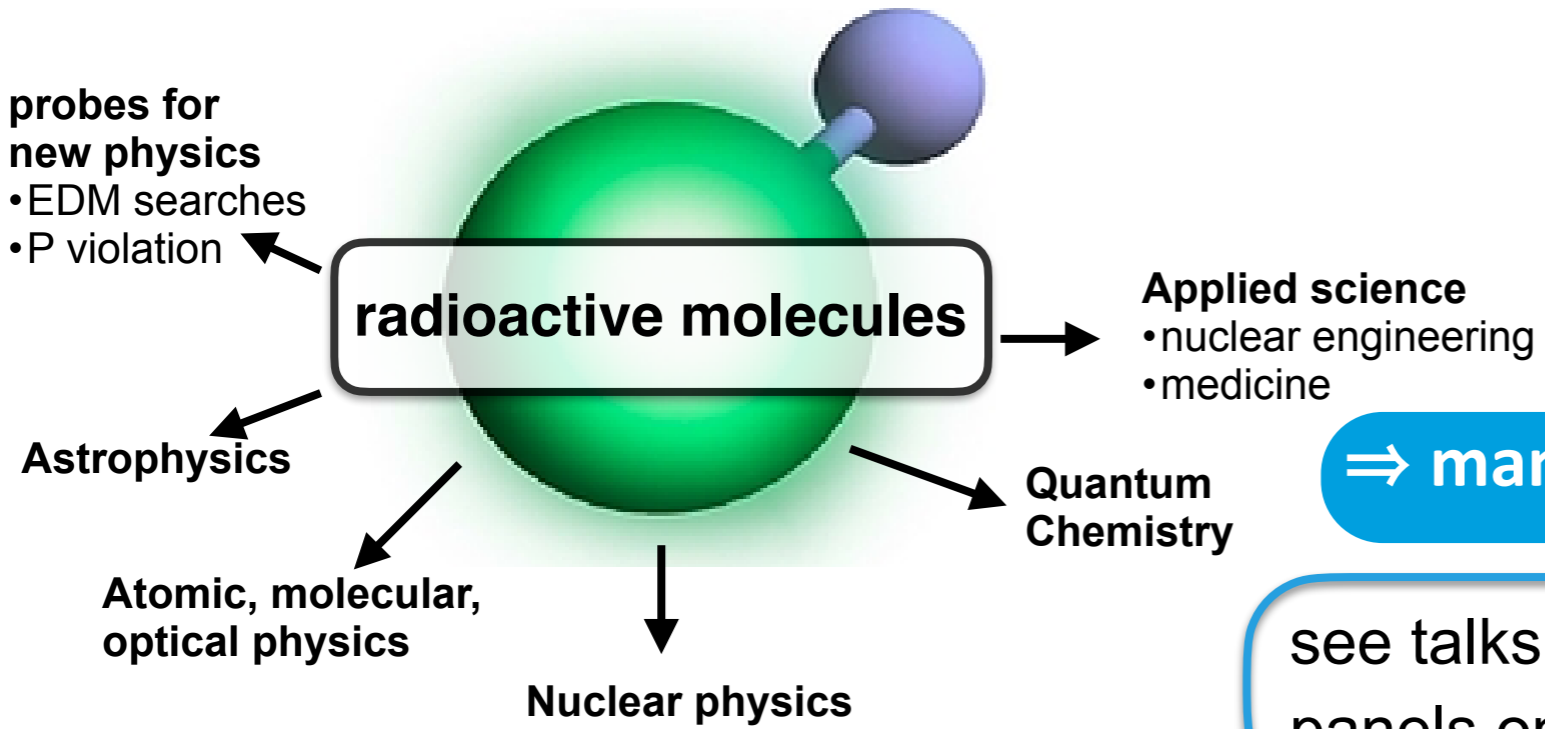


⇒ many, exciting science opportunities

see talks by A. Jamison, M. Au, S. Franchoo  
panels on Tue and Wed

*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: Rep. Prog. Phys. 87 084301 (2024)*

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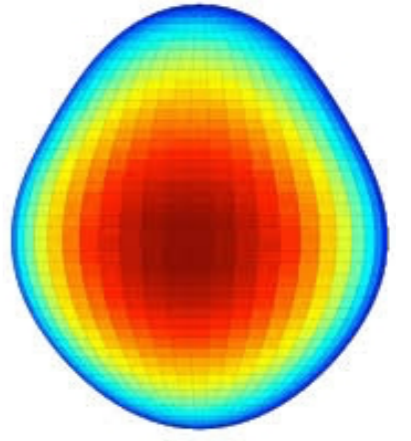
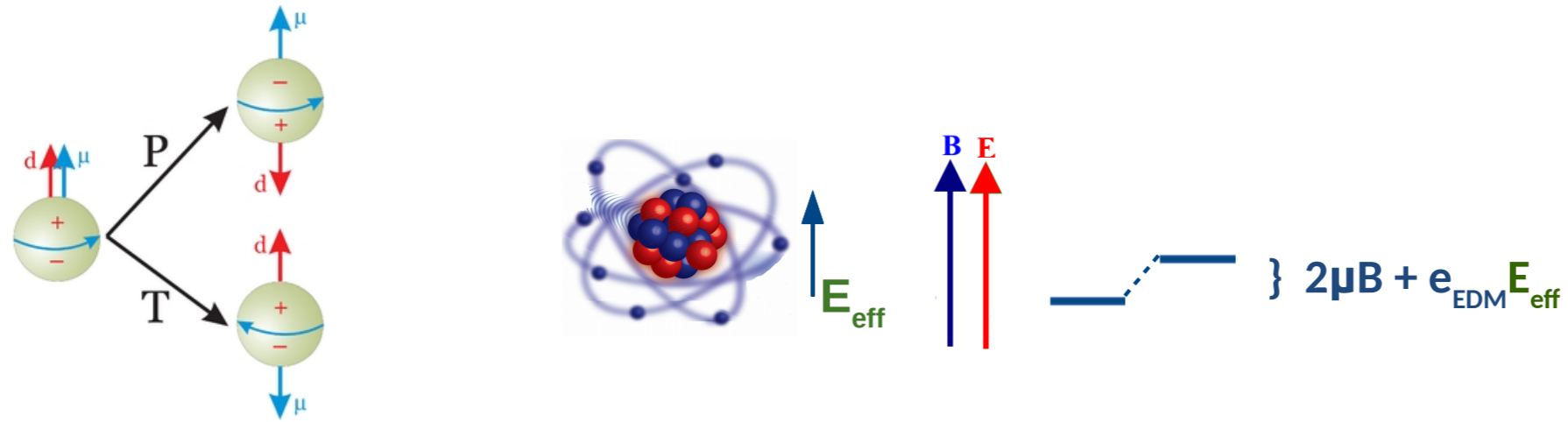


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## Electric Dipole Moments (EDM)



Octupole Deformation

# 'Designer Molecules'

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

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

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

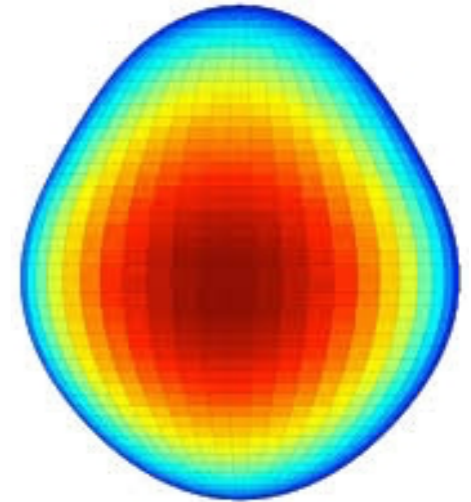
$$|S_{\text{Hg}}| < 3.1 \cdot 10^{-13} \text{ e fm}^3$$

*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
- } compared to  $^{199}\text{Hg}$

all known cases in radionuclides



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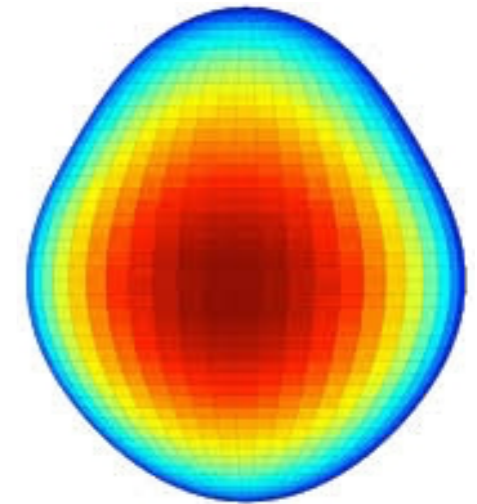
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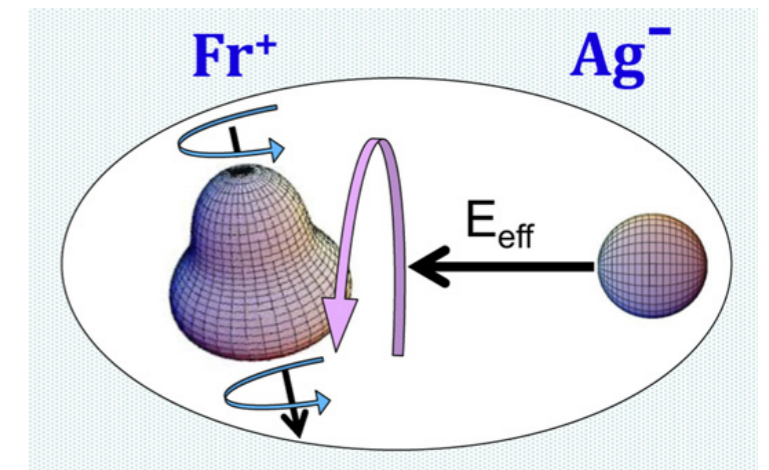
all known cases in radionuclides

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

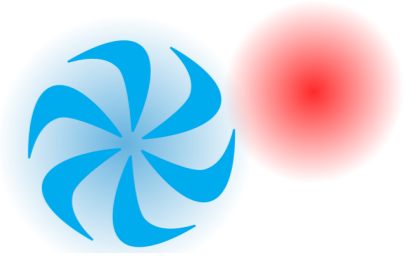
- **intrinsic enhancement of  $10^7$  compared to  $^{199}\text{Hg}$**

*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 radioactive ion beam (RIB) facilities  
    ➔ challenge: reduced availability
- **anticipated gain: x 1,000 for certain CPV-parameters (comp to  $^{199}\text{Hg}$ )**

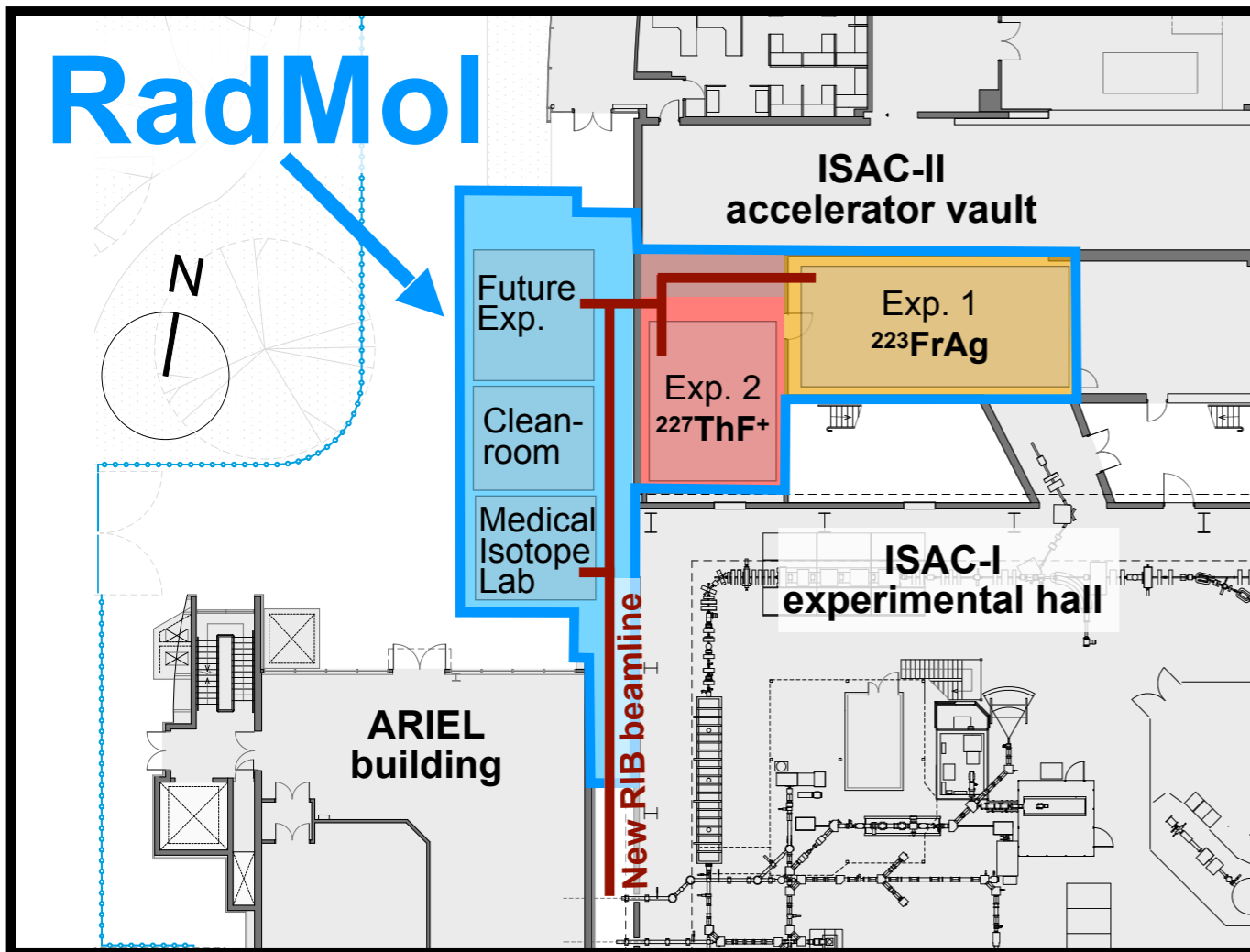


# RadMol



*a radioactive molecule lab for fundamental physics*

## RadMol



### Goal:

- dedicated laboratory to study radioactive molecules<sub>4</sub>
- to host multiple experimental stations
- precision studies for searches for new physics
- Molecular EDM with unprecedented sensitivity to nuclear T-breaking Schiff moments using **octupole deformed nuclei**
- provision for expansions into other fields

### TRIUMF strengths:

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

### NEWS:

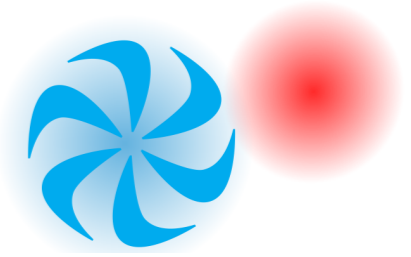
- Funding application approved via



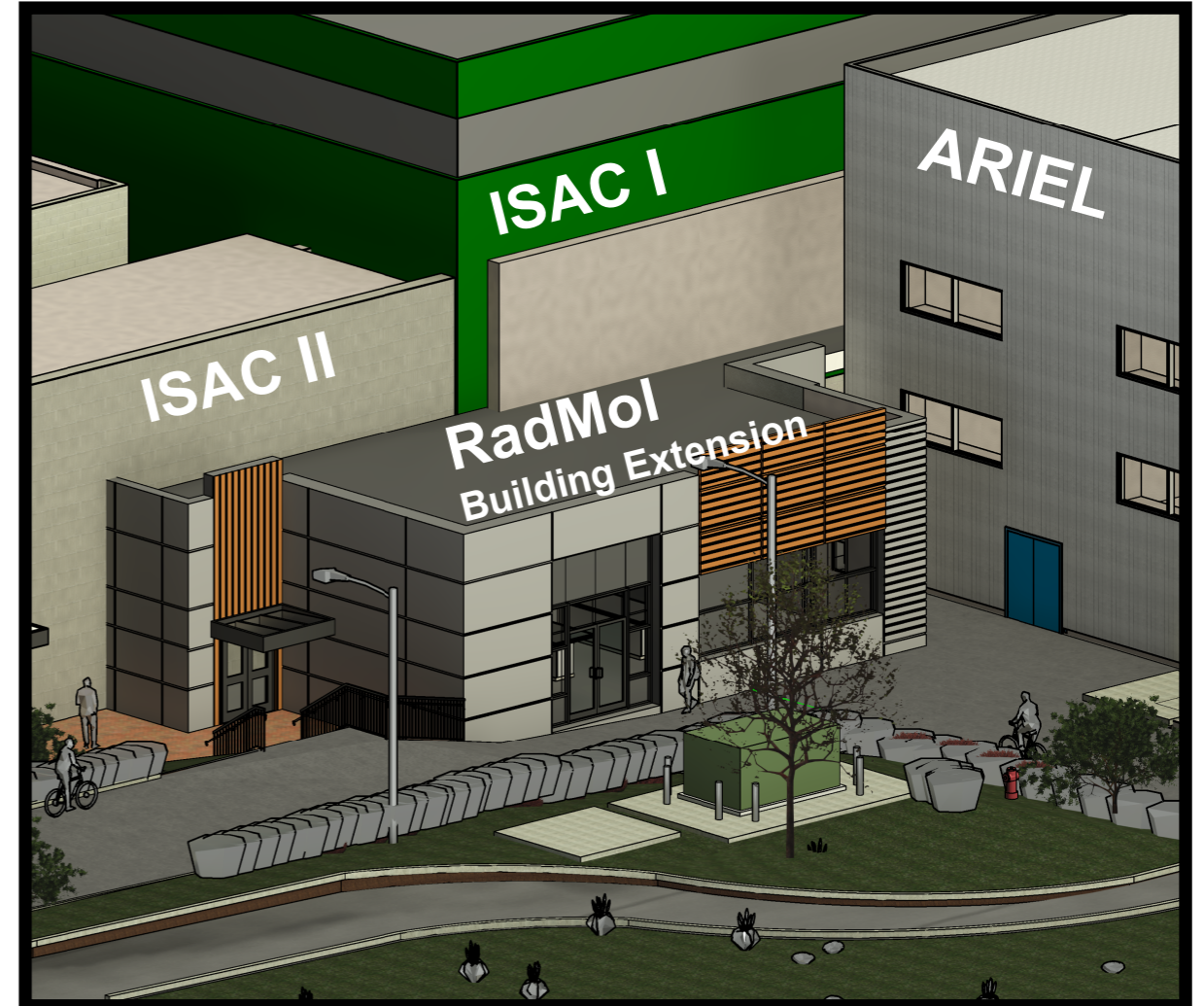
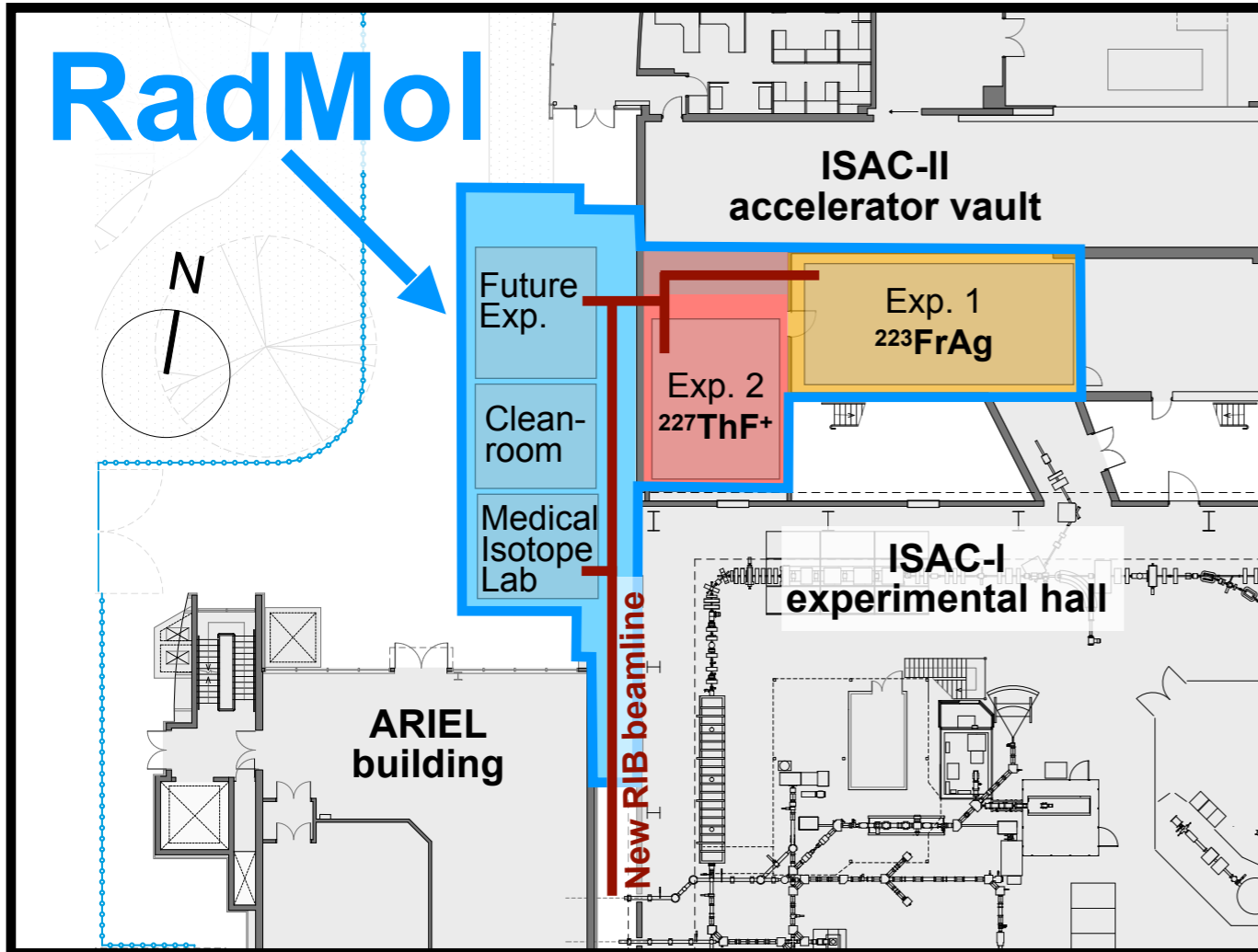
### RadMol Collaboration:



# Scope of present RadMol project

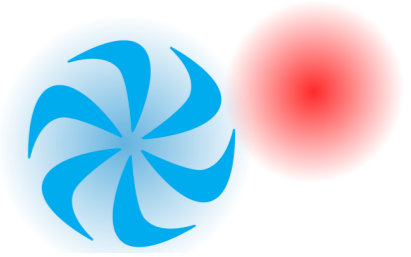


5



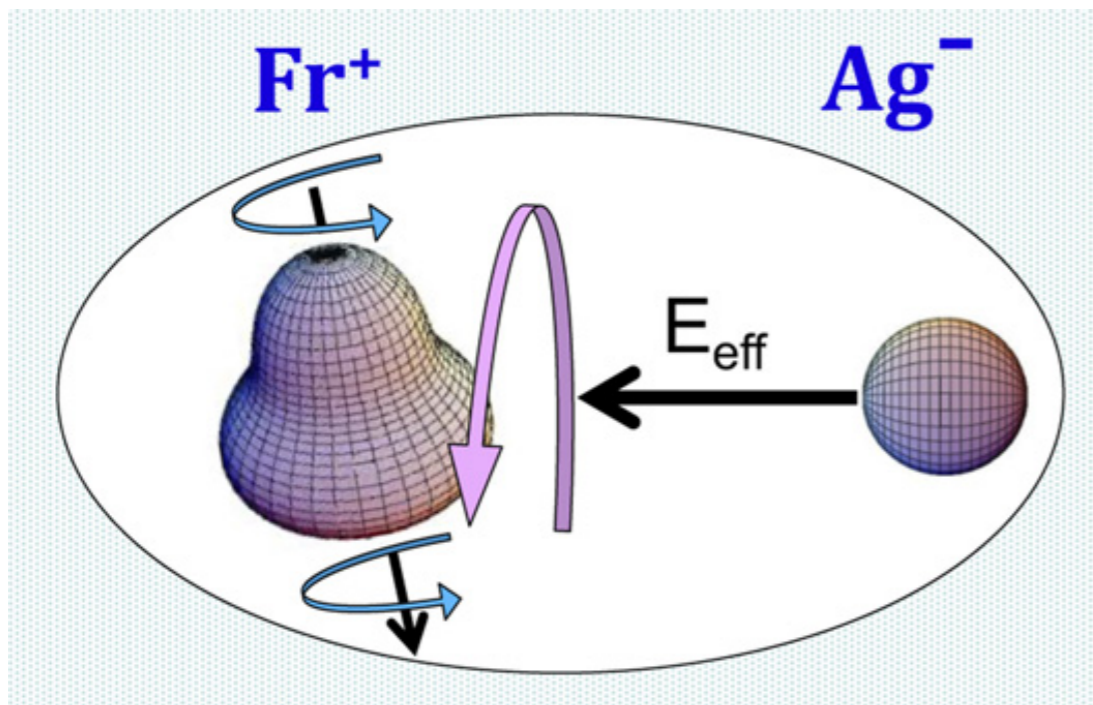
- renovation of existing laboratory space
- 2 experimental stations for Schiff-moment searches
- RIB beam lines to RadMol labs
- RadMol building extension

Initial focus of RadMol@TRIUMF

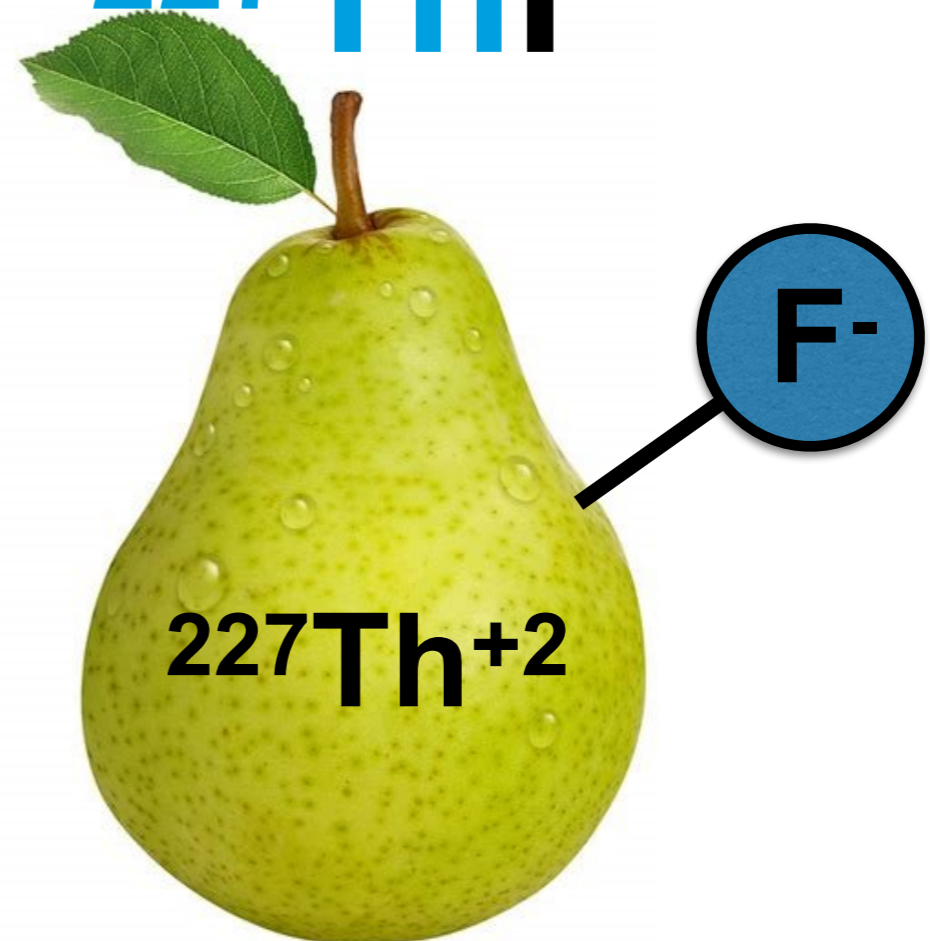


# Precision Experiments on

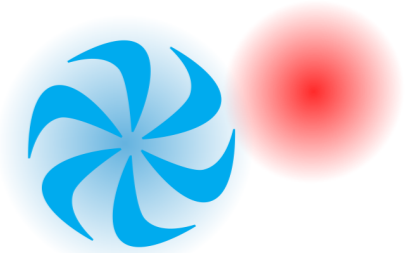
$^{223}\text{FrX}$



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

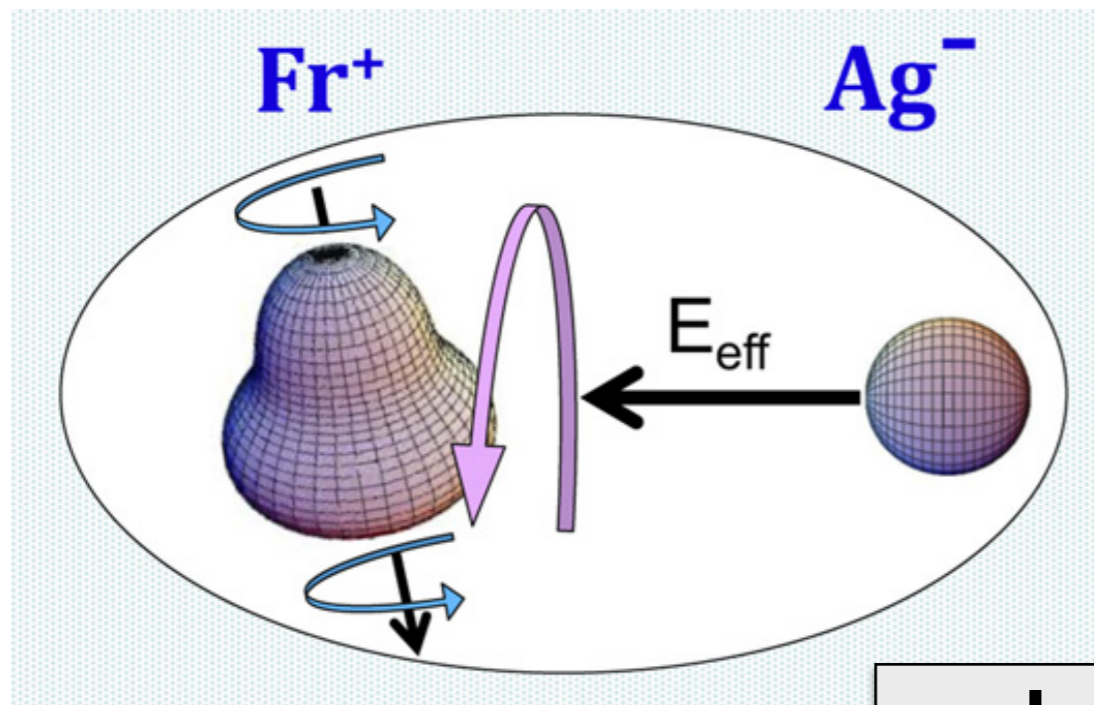


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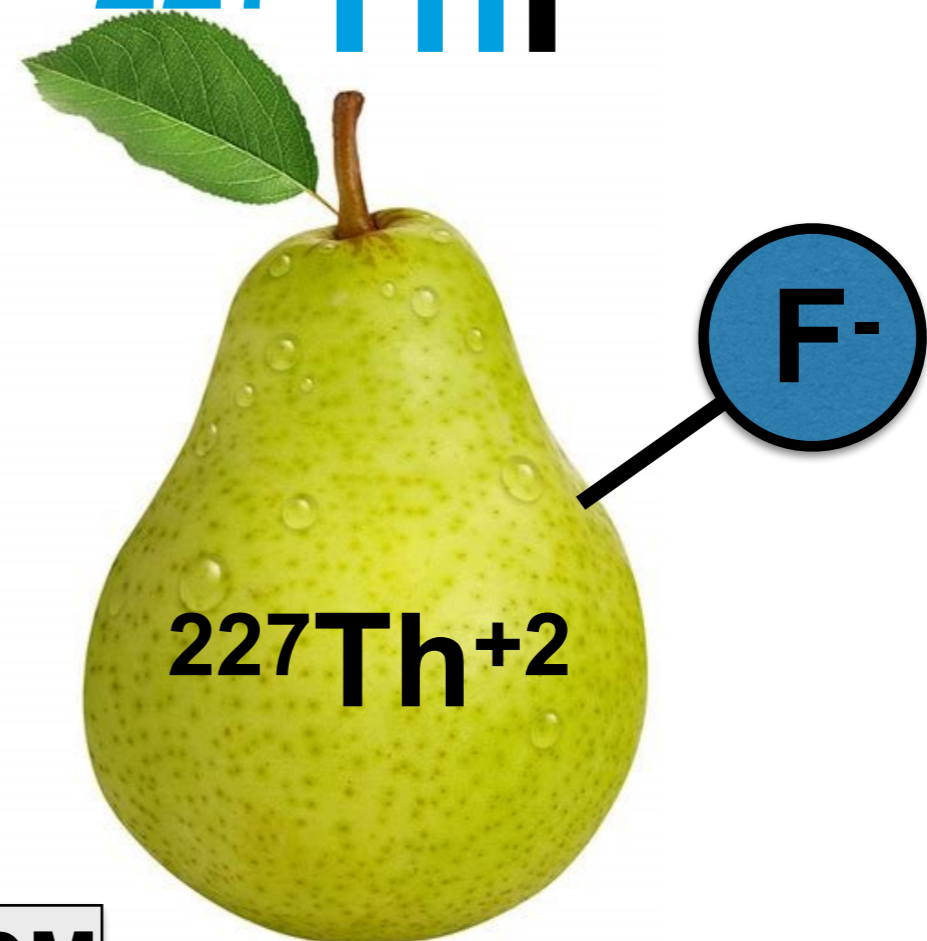


# Precision Experiments on

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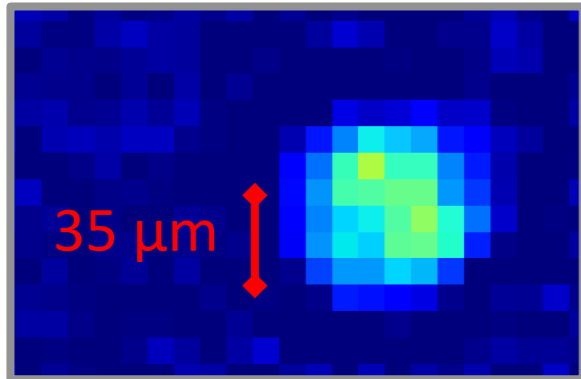
molecular EDM

nuclear Schiff moment

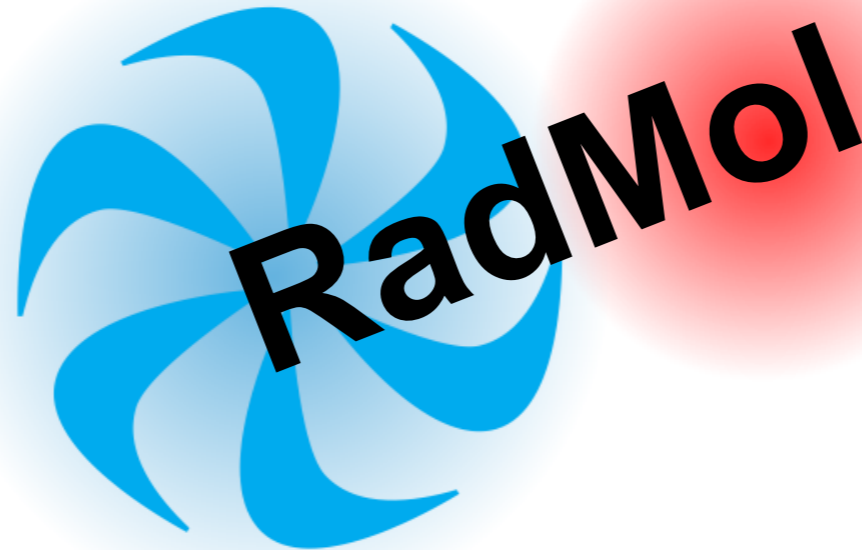
new physics

# Foundational RadMol projects

**Fr: low-energy scattering behaviour**  
@ TRIUMF's Fr trapping facility

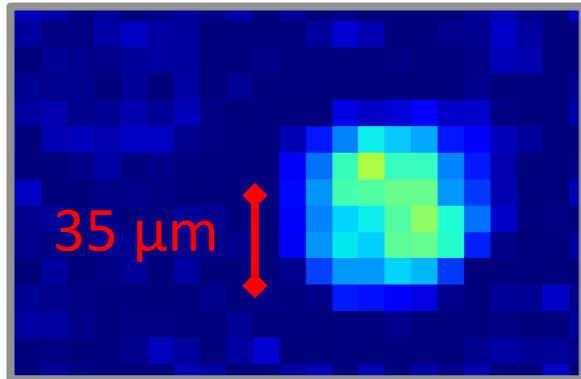


see talk by A. Jamison



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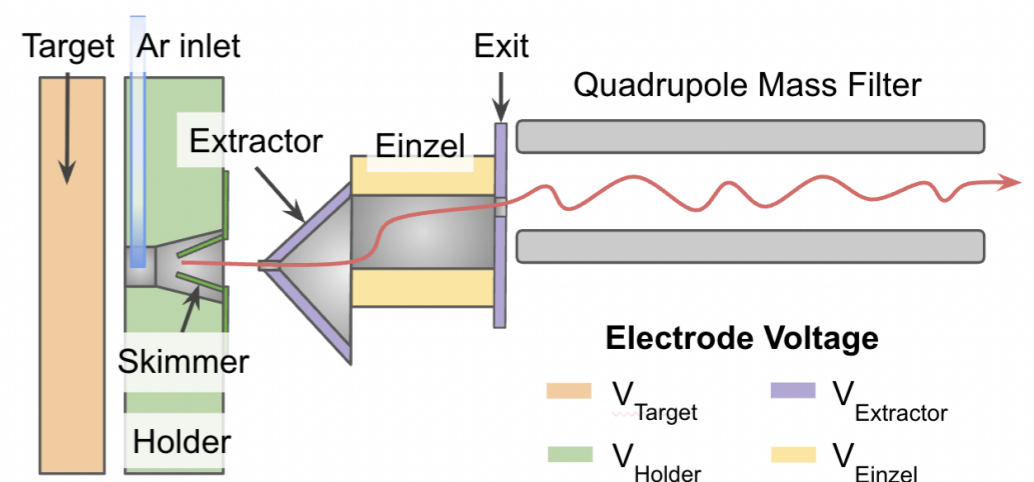


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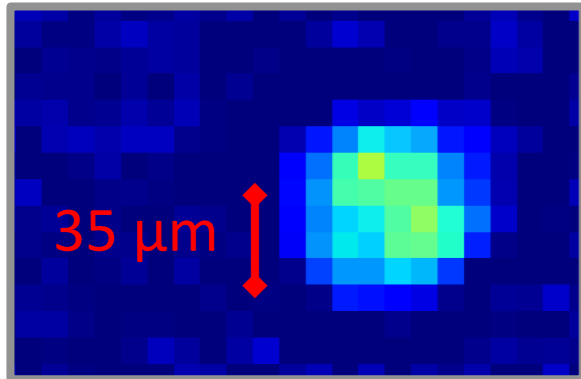
RadMol

development of  $\text{Th}^+$  /  $\text{ThF}^+$  beams  
@ existing RadMol lab



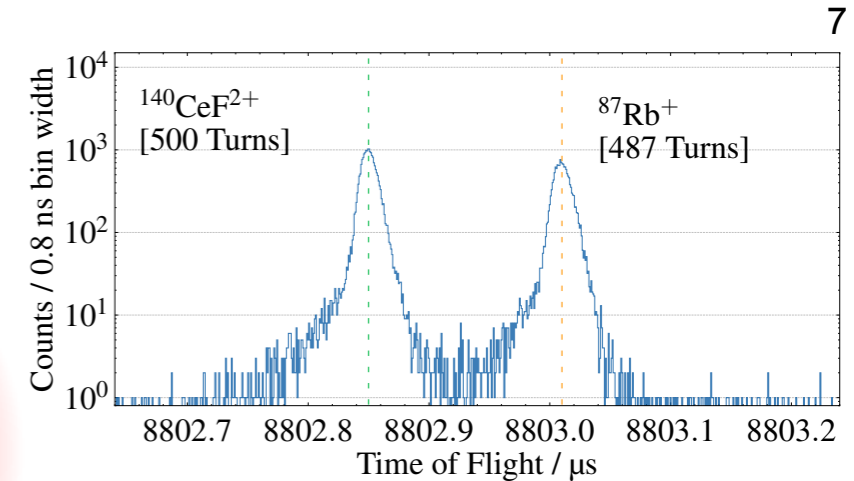
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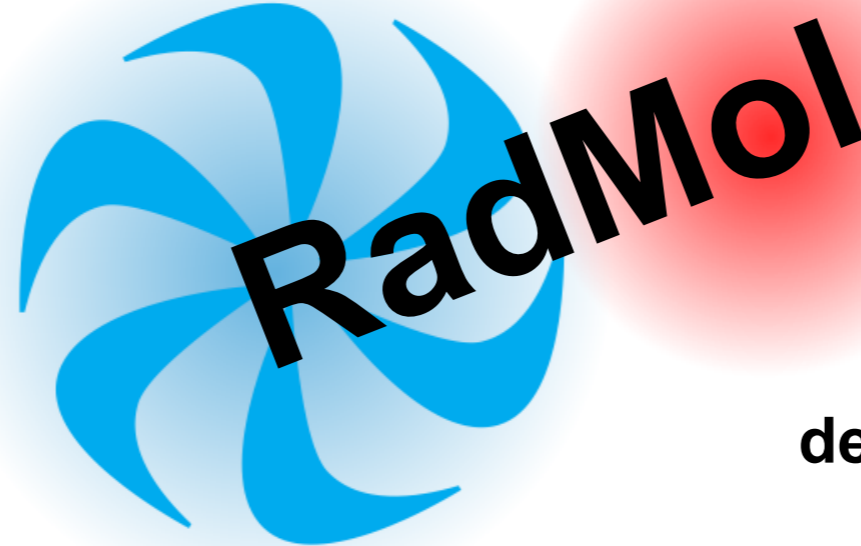


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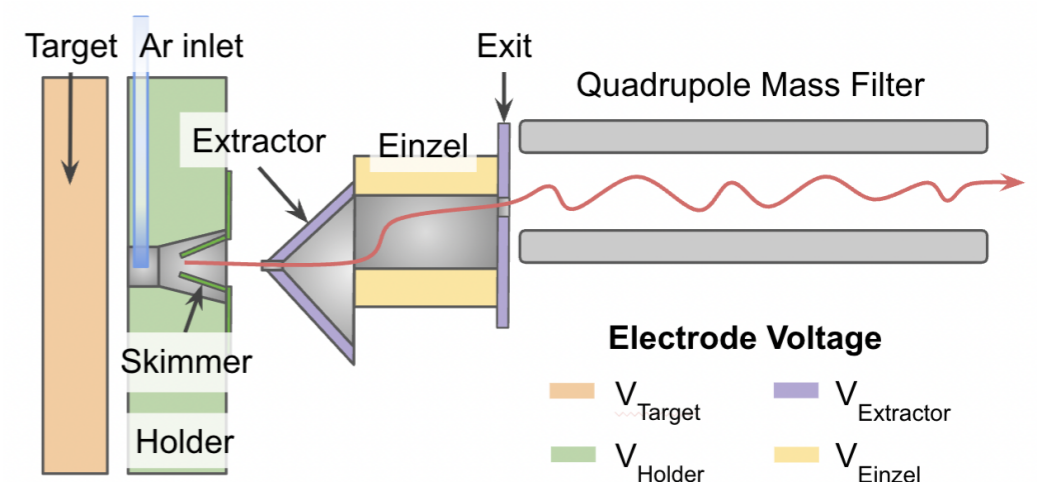
formation of ('highly' charged) molecules  
@ TITAN + OLIS or TITAN EBIT



*R. Simpson et al., submitted*

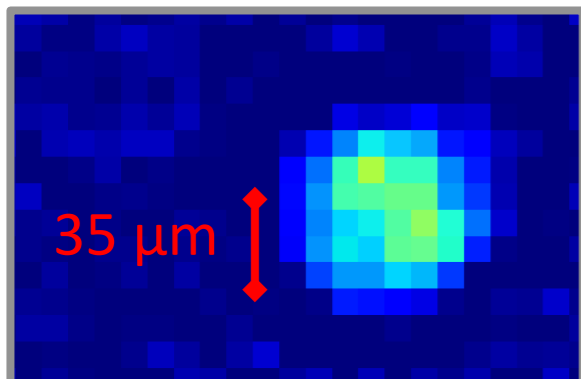


development of Th<sup>+</sup> / ThF<sup>+</sup> beams  
@ existing RadMol lab



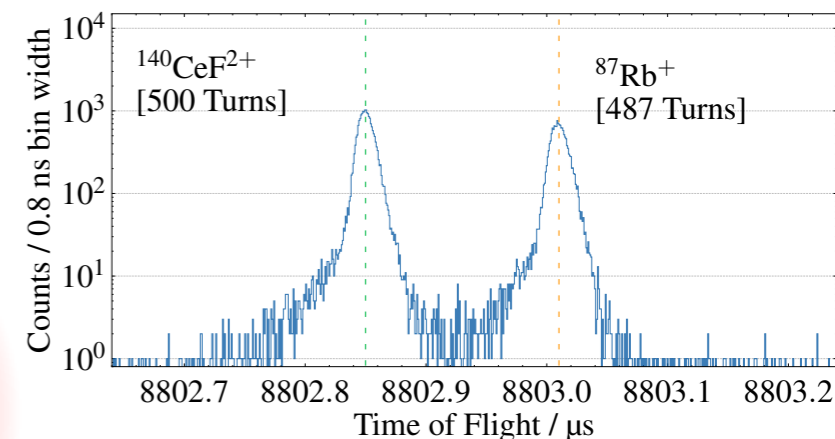
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R. Simpson et al., submitted

physics interpretation &  
collaboration:

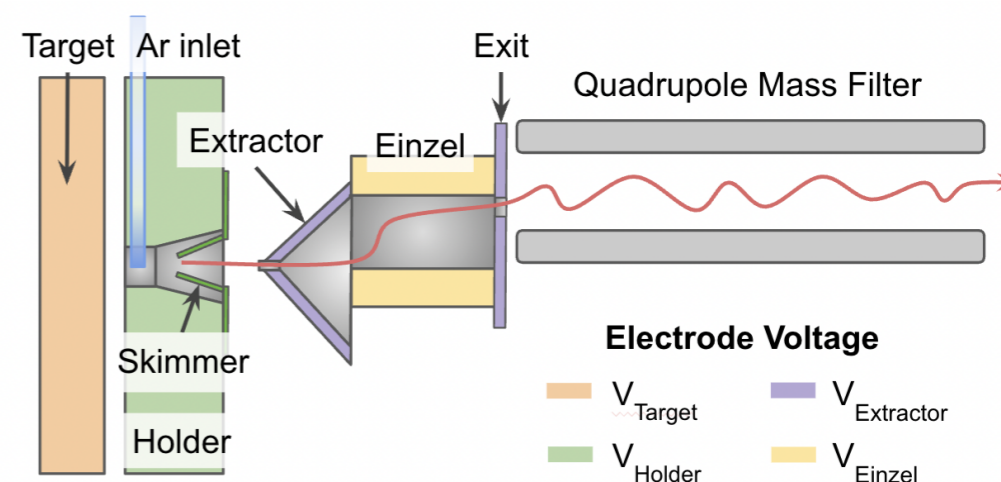
exp - nuclear theory - quantum chemistry

## Nuclear Schiff Moment of $^{19}\text{F}$

- (re-)interpret  $\text{HfF}^+$  as NSM
- 1<sup>st</sup> nuclear *ab initio* calculation of a NSM

K.B. Ng et al., *Phys. Rev. Lett.* 136, 112501 (2026)

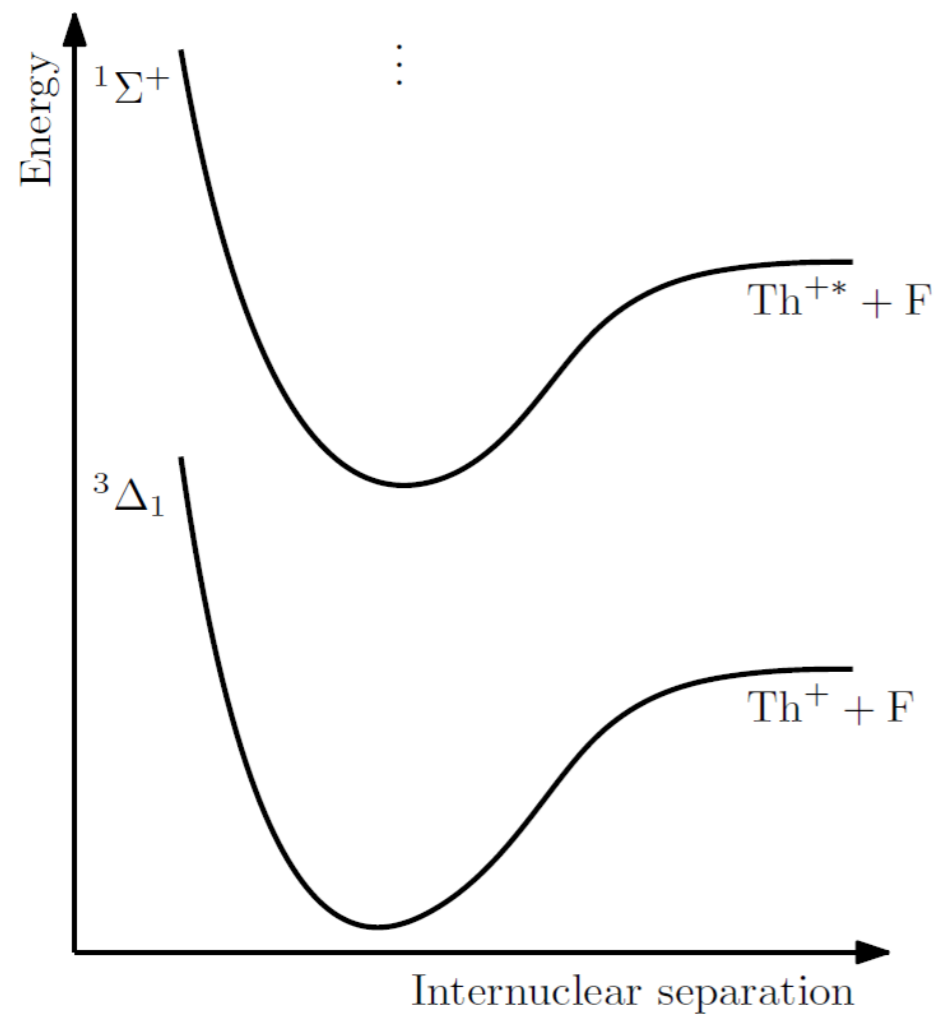
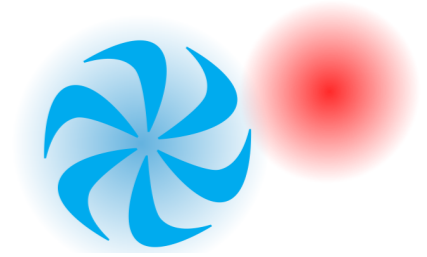
development of  $\text{Th}^+$  /  $\text{ThF}^+$  beams  
@ existing RadMol lab



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

half-life:  $\approx 19$  days

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

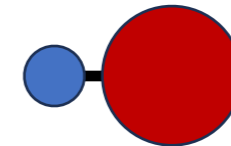
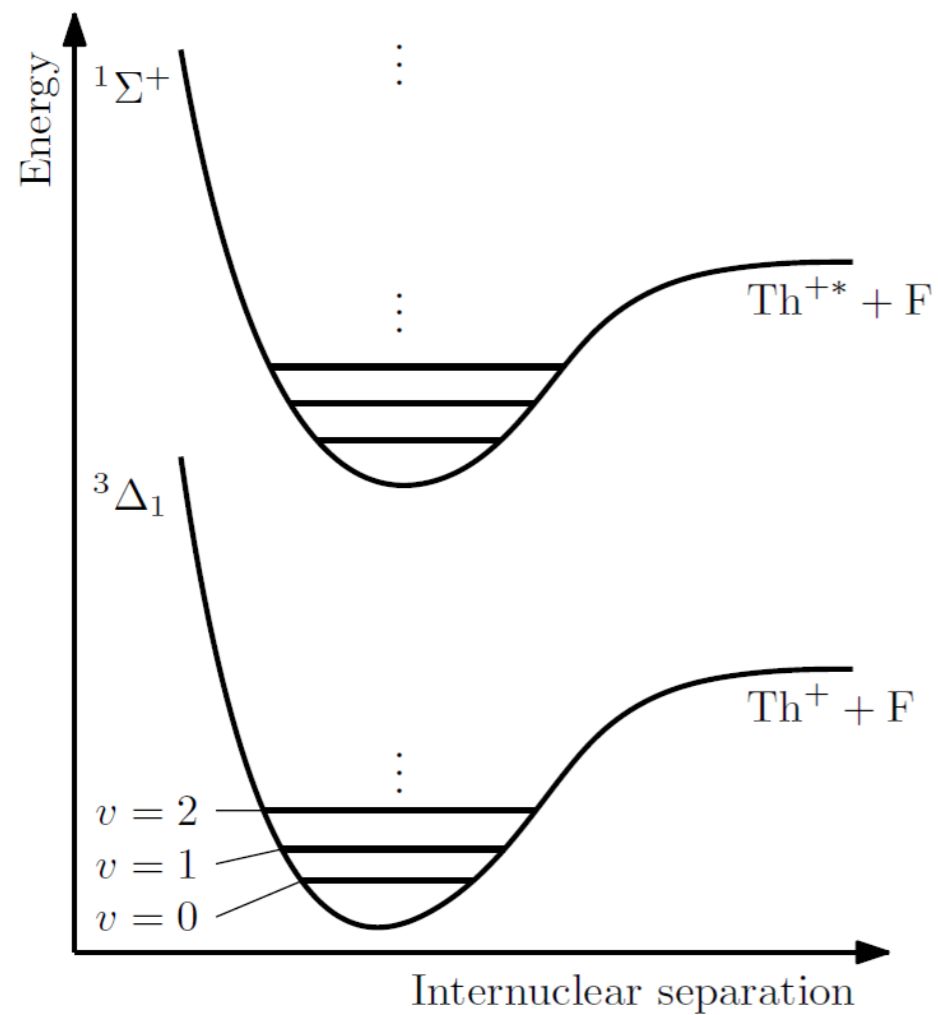
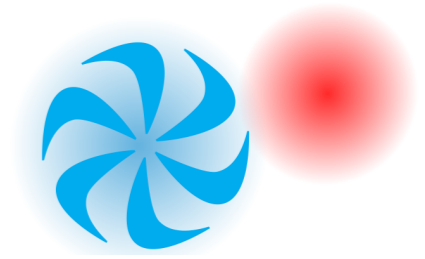


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→ Vibrational manifold ( $600\text{ cm}^{-1} \sim 800\text{ K}$ )



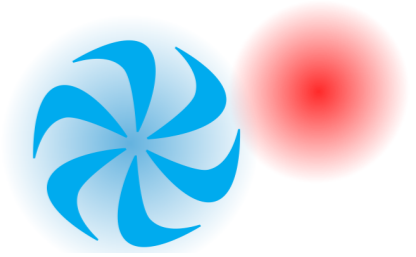
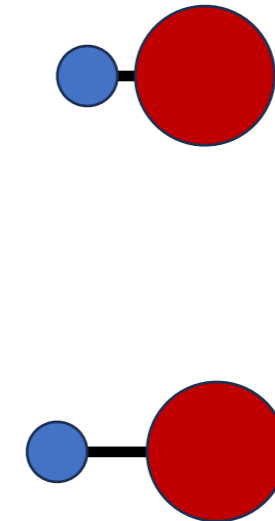
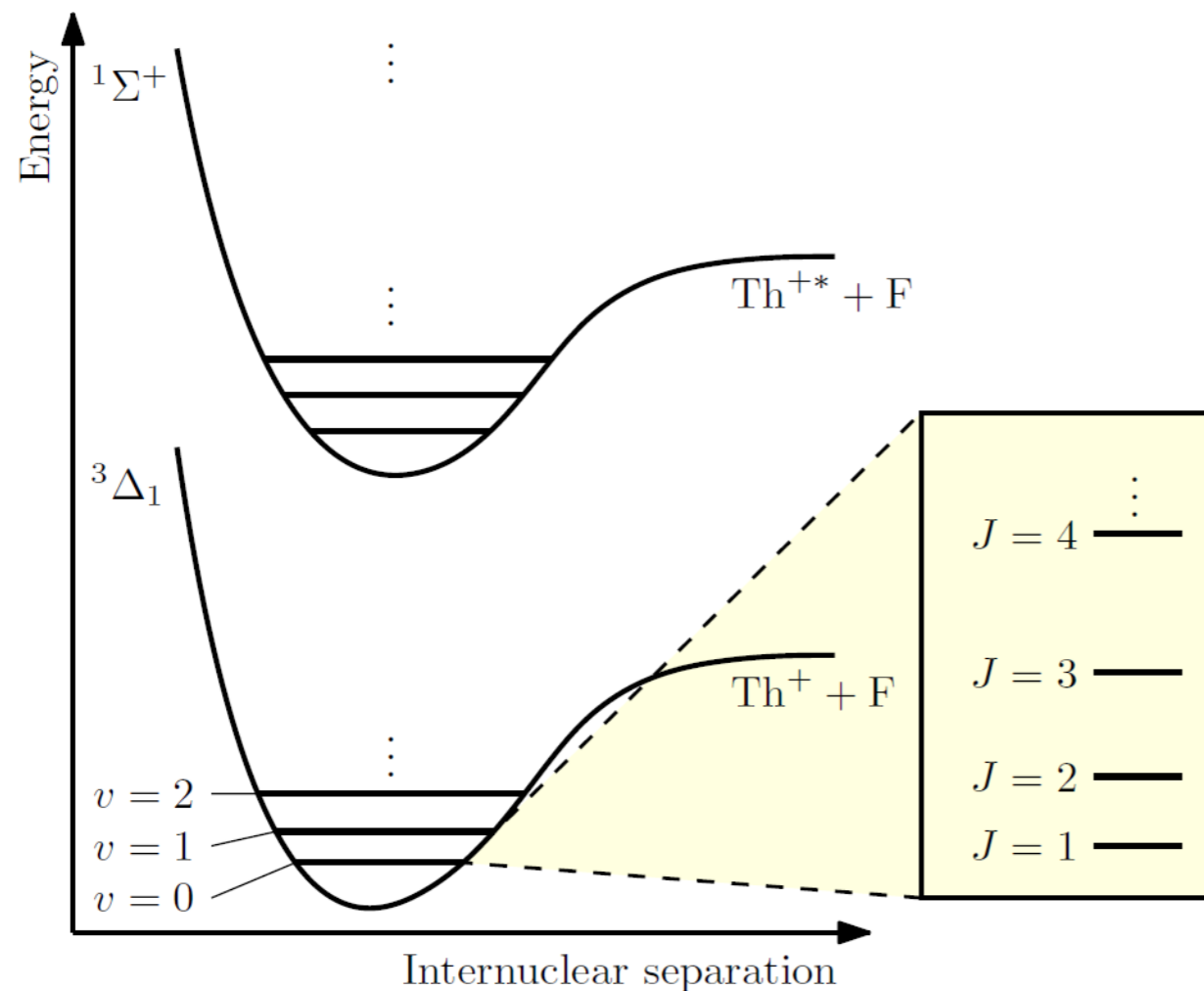
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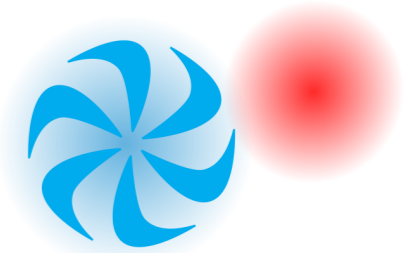
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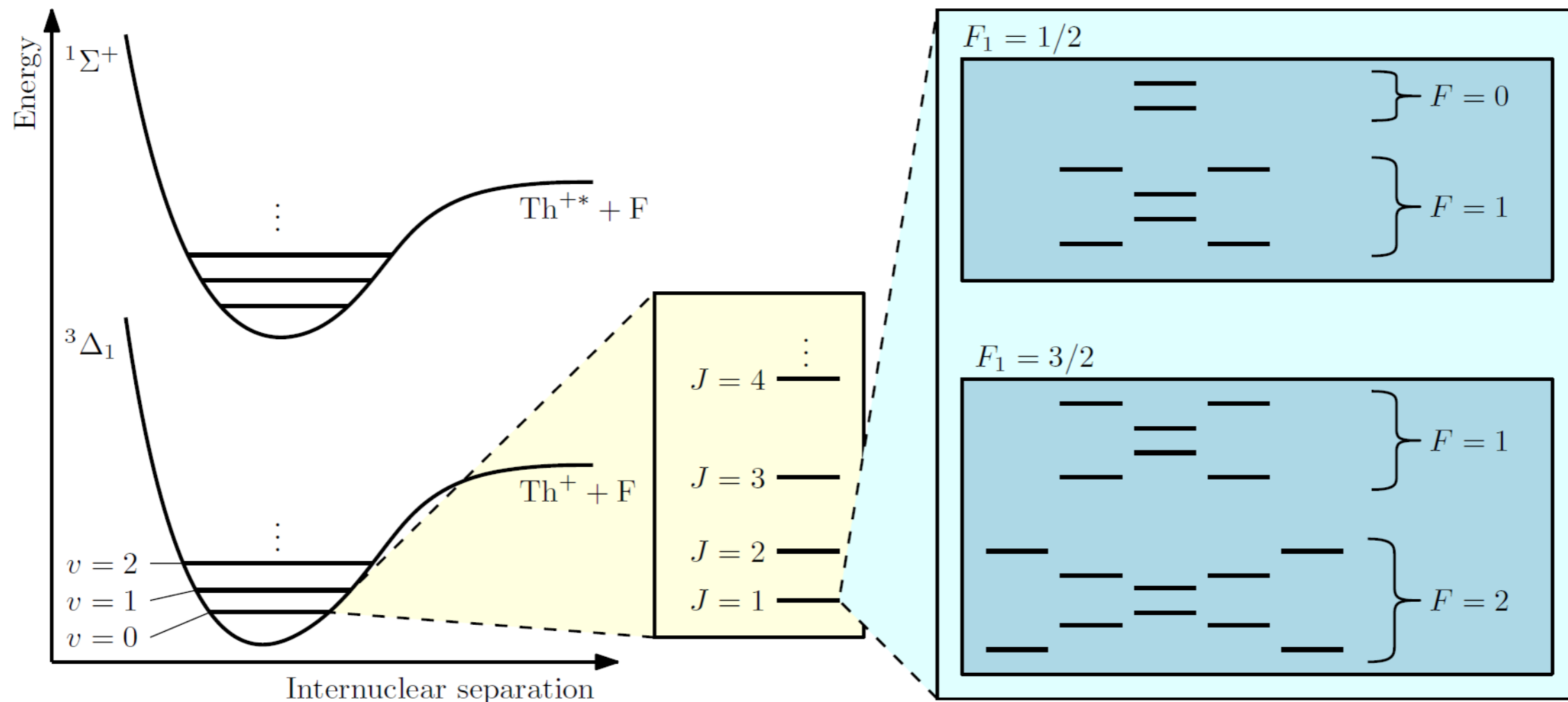
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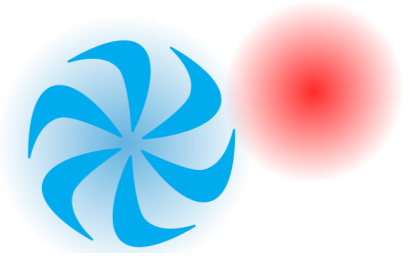
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- Parity doublets, Stark, Zeeman ( $10 - 100 \text{ MHz}$ )



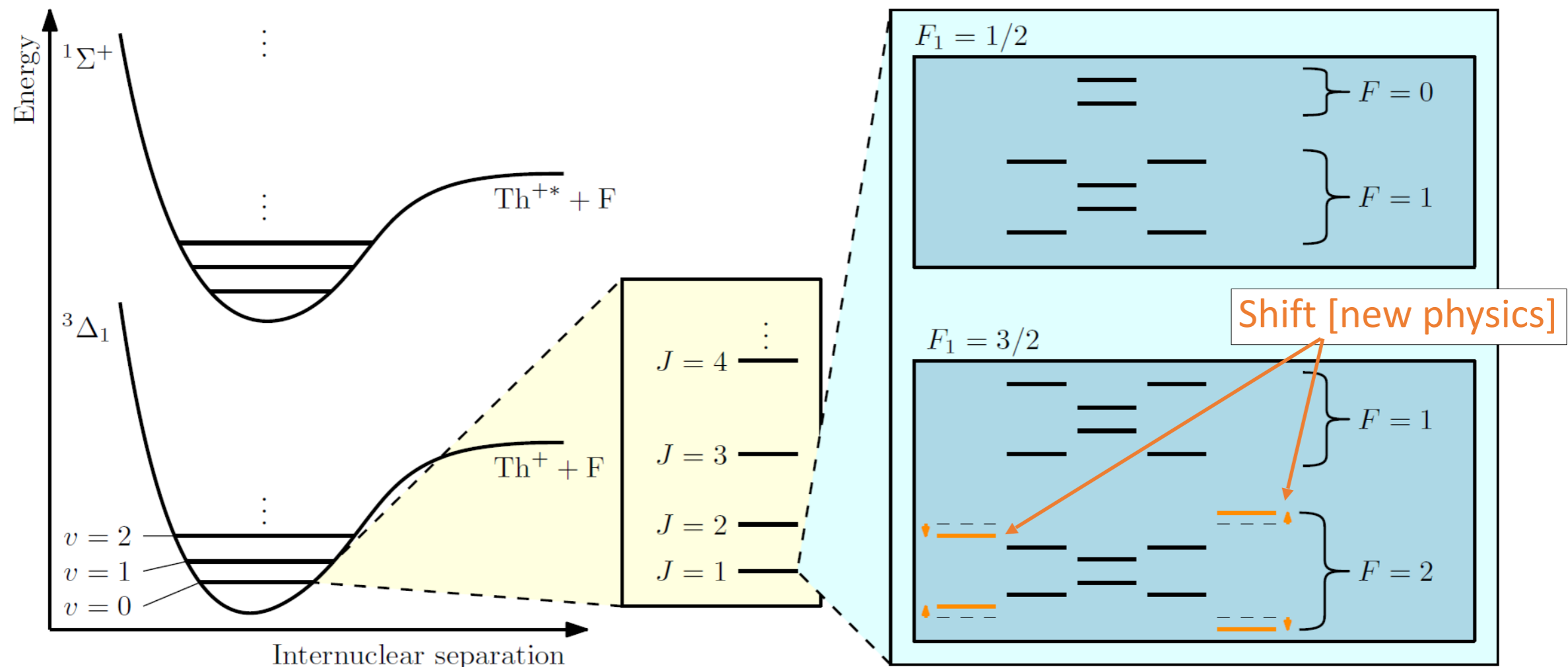
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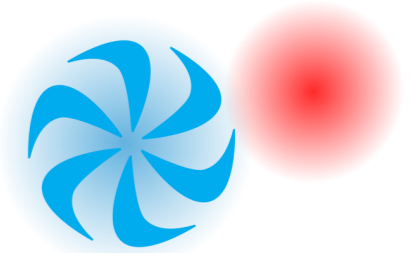
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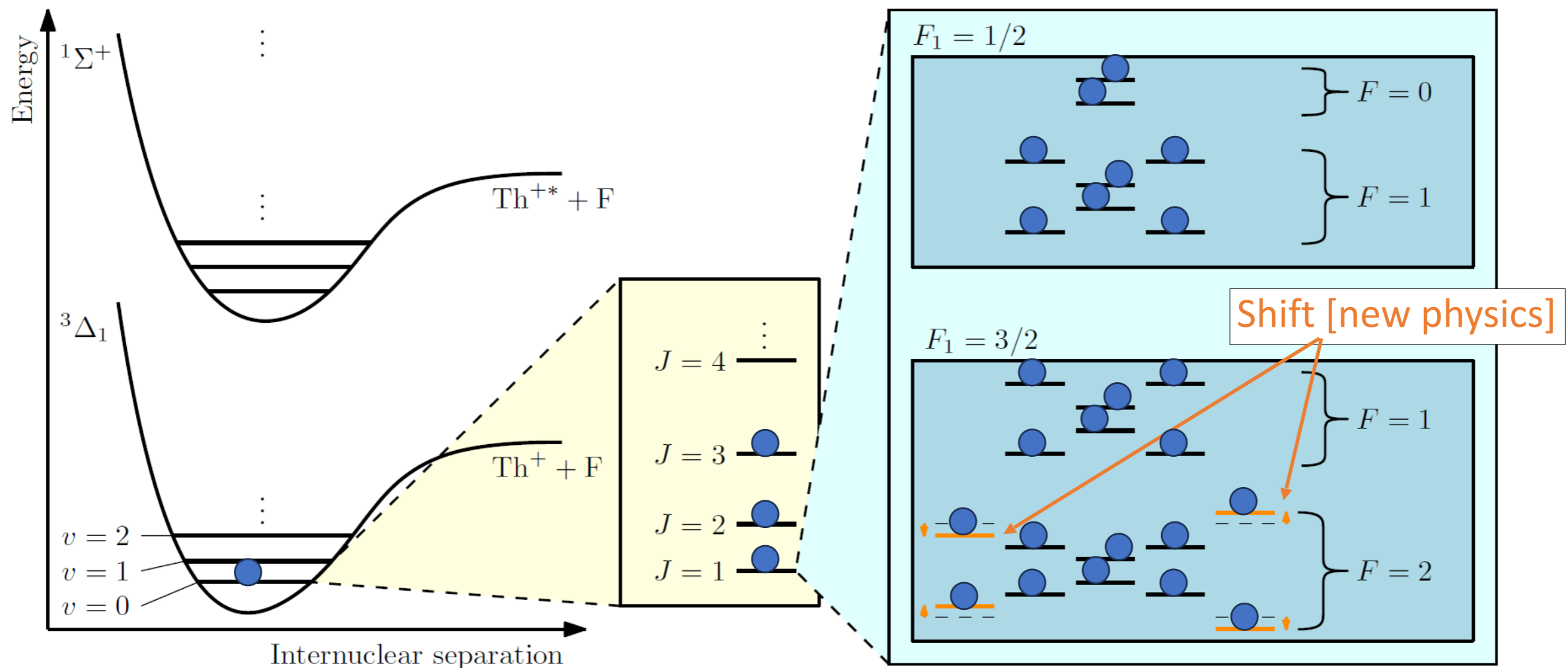


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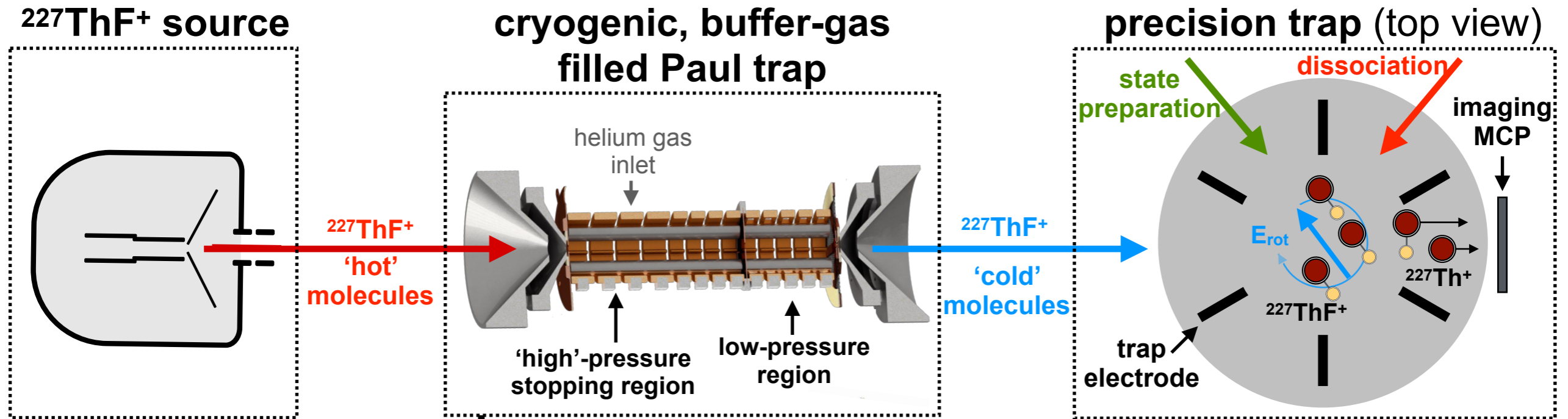
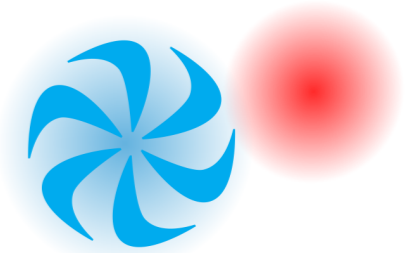
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Ions typically produced/cooled to  $\sim 10\text{ K}$   
 $\Rightarrow$  distributed across multiple  $J$  (and finer structures).



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



Cooling of

- centre of mass motion
- inner degrees of freedom

- experimental EDM technique analogous to  $\text{HfF}^+$  electron EDM experiment (worldwide best)

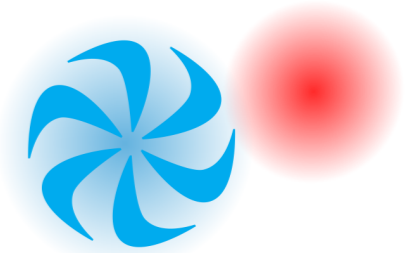
*T. S. Roussy et al. Science 381.6653 (2023), pp. 46–50.*

- molecular structure of  $^{232}\text{ThF}^+$  known from spectroscopy at JILA

*K.B. Ng et al. Phys. Rev. A 105, 022823 (2022)*

- access to  $^{227}\text{Th}$  via  $^{227}\text{Ac}$  sample ( $T_{1/2} = 22$  years)

# Th ion source

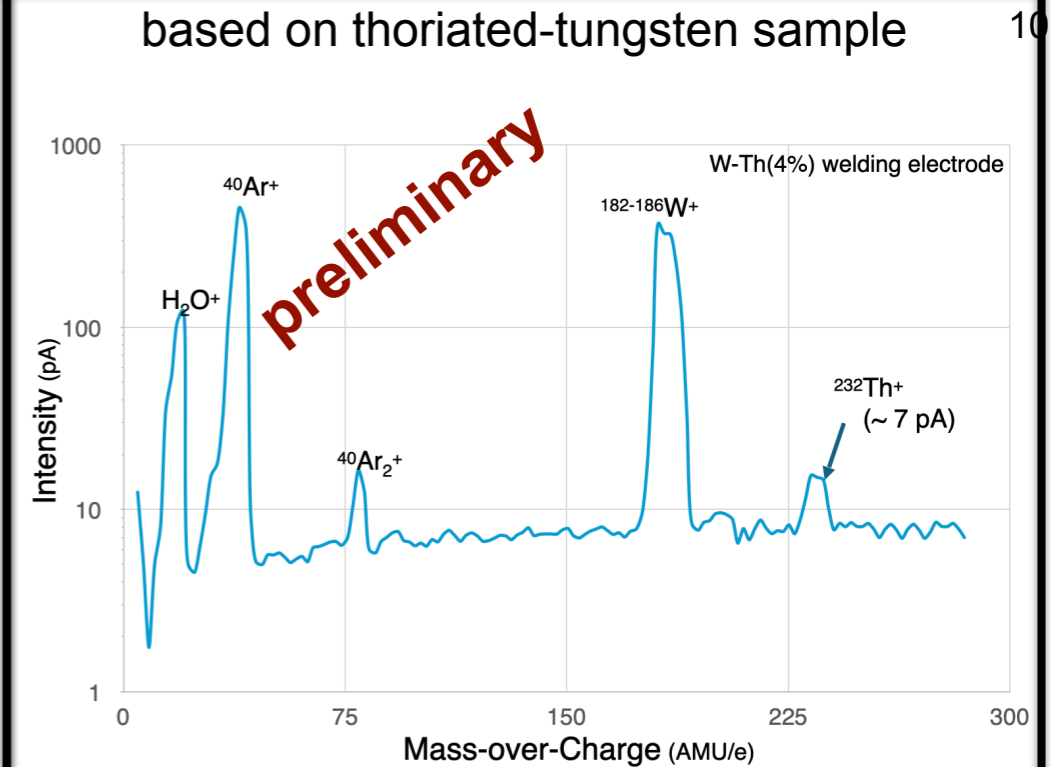
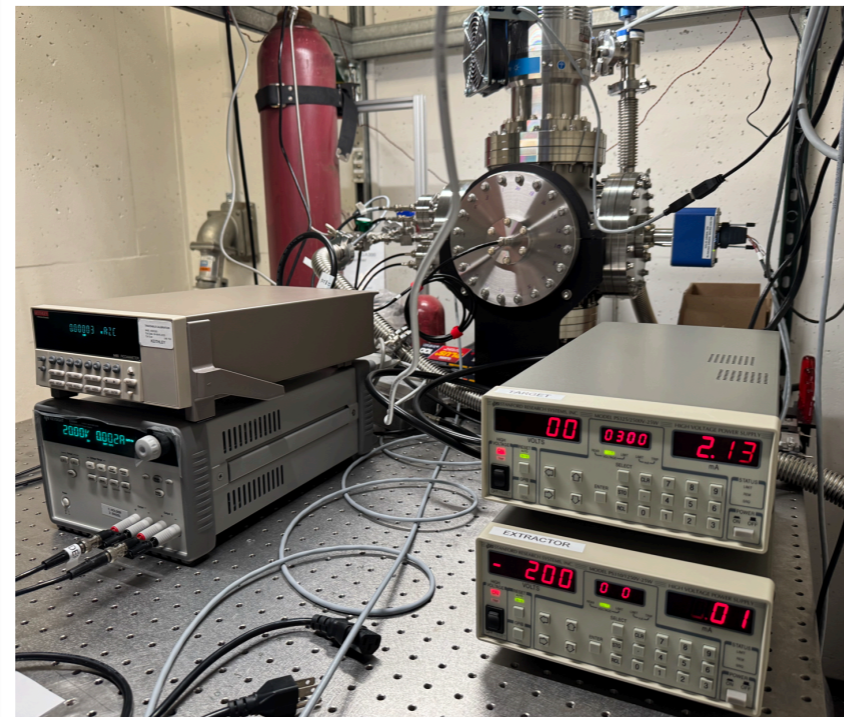
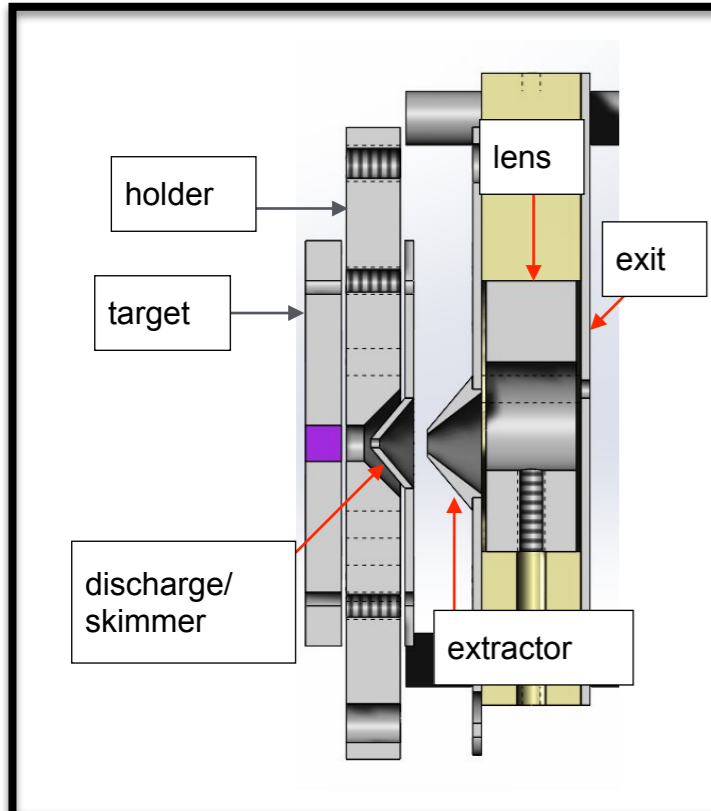


- glow-discharge ion source (design by M. P. Reiter/ Edinburgh)

ion-source schematics

setup @ TRIUMF

first  $^{232}\text{Th}^+$  ion beam @ Edinburgh

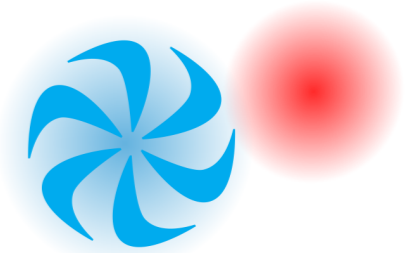


## Objective:

- efficient, compact & low-maintenance source for short-lived  $^{227}\text{Th}^+$
- test in-source formation of  $\text{ThF}^+$
- expand to other refractory elements (including Pa)

S. Cui et al., to be submitted

# Th ion source

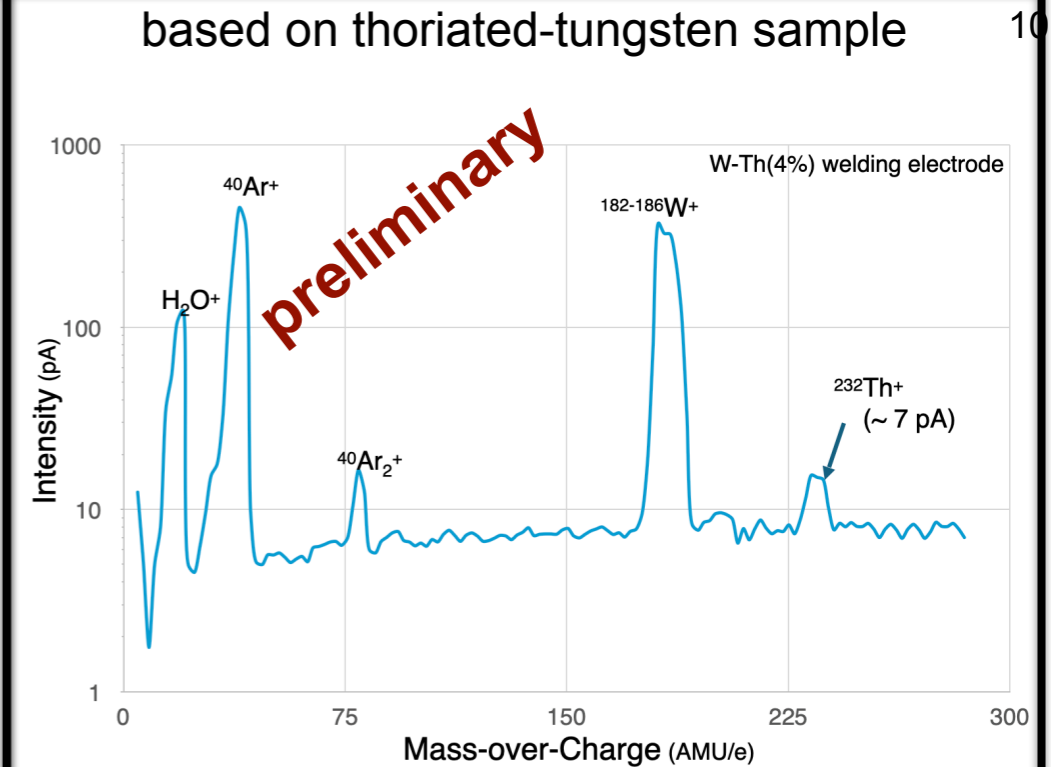
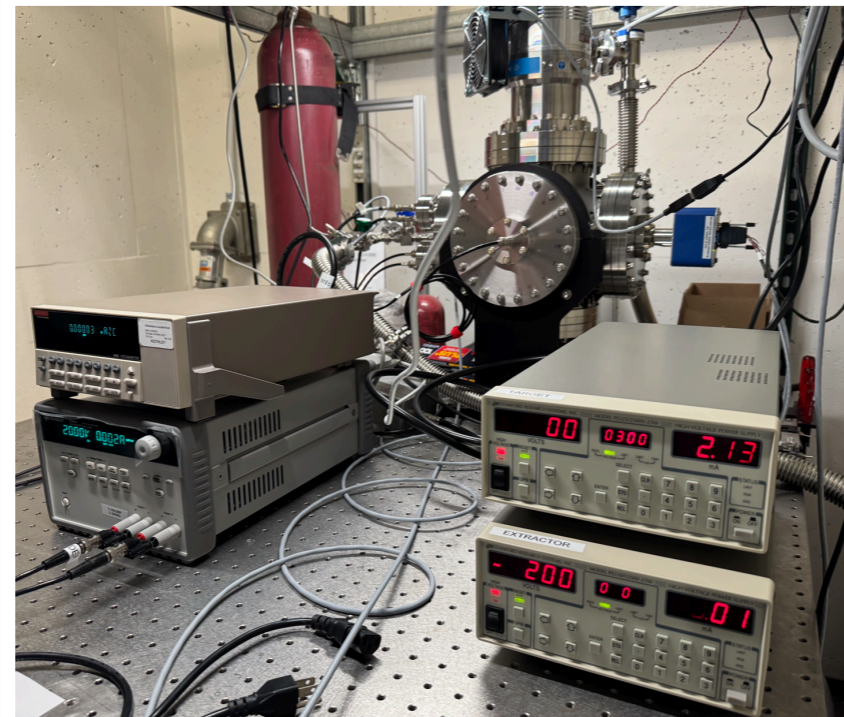
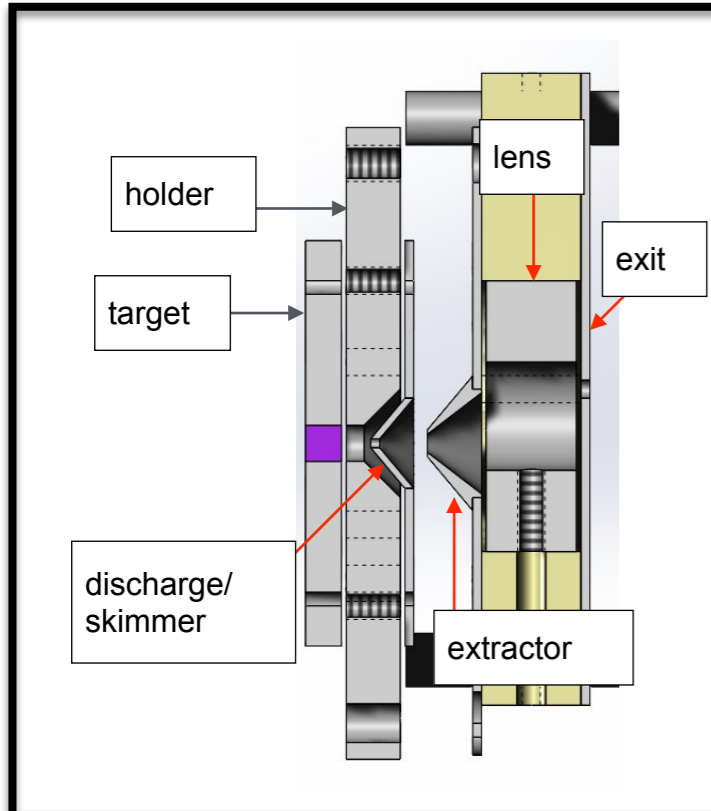


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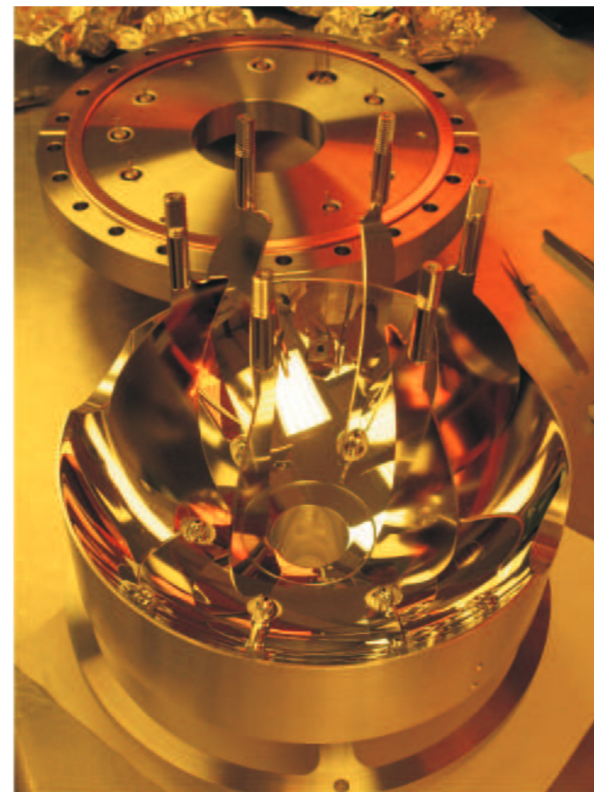
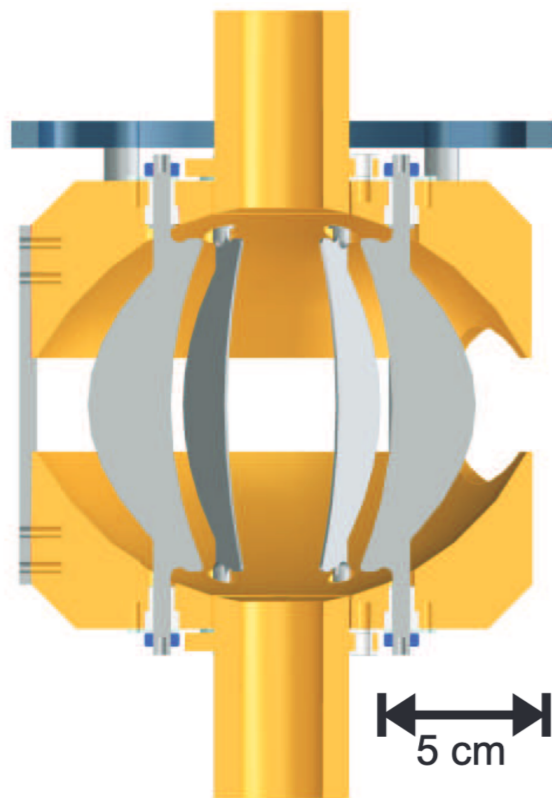
use of  $^{225,227}\text{Ac}$  samples for offline Fr source ?  
(to complement high-yield ISAC/ARIEL beam)

GANIL

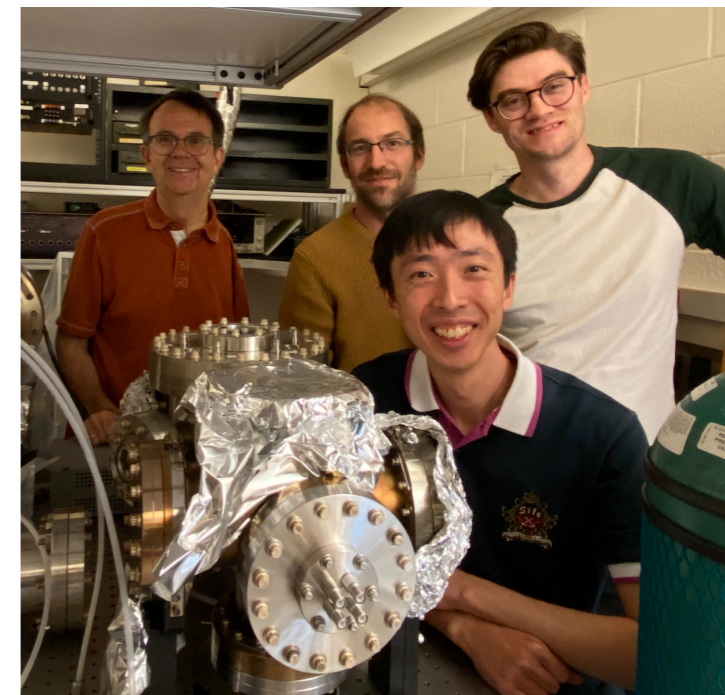
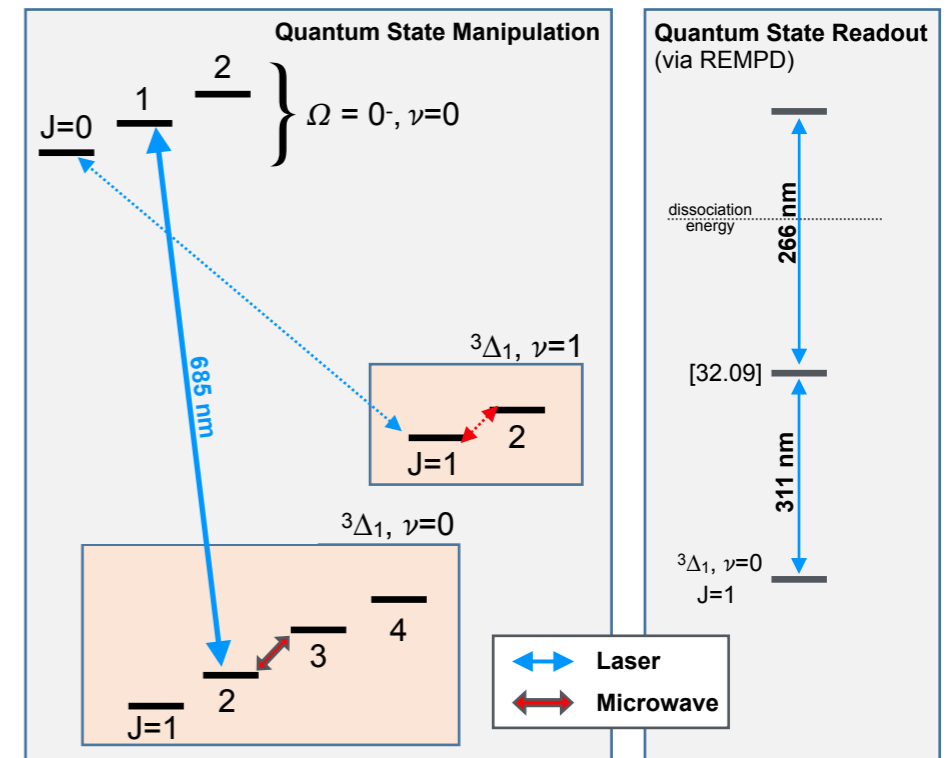
ijc Lab  
Irène Joliot-Curie  
Laboratoire de Physique  
des 2 Infinis

# Toward 1<sup>st</sup> quantum state manipulation and readout of $^{227}\text{ThF}^+$

- Ion trap for first  $\text{ThF}^+$  experiments at TRIUMF  
initially: naturally-occurring  $^{232}\text{Th}$ ,  
later:  $^{227}\text{Th}$  ( $T_{1/2}=19$  d)
- In existing, operational laboratory

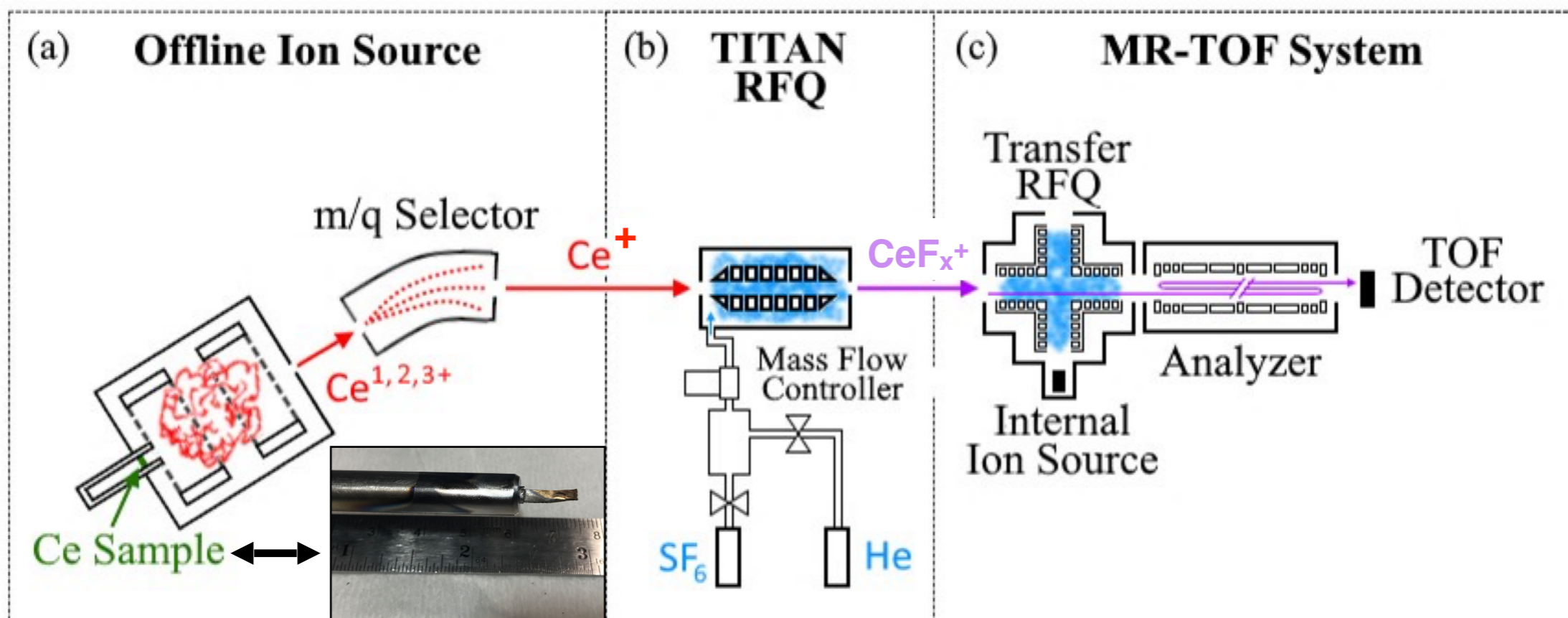


**JILA Gen 1 EDM ion trap**



recent progress of JILA-TRIUMF collaboration: improved efficiency in quantum state readout

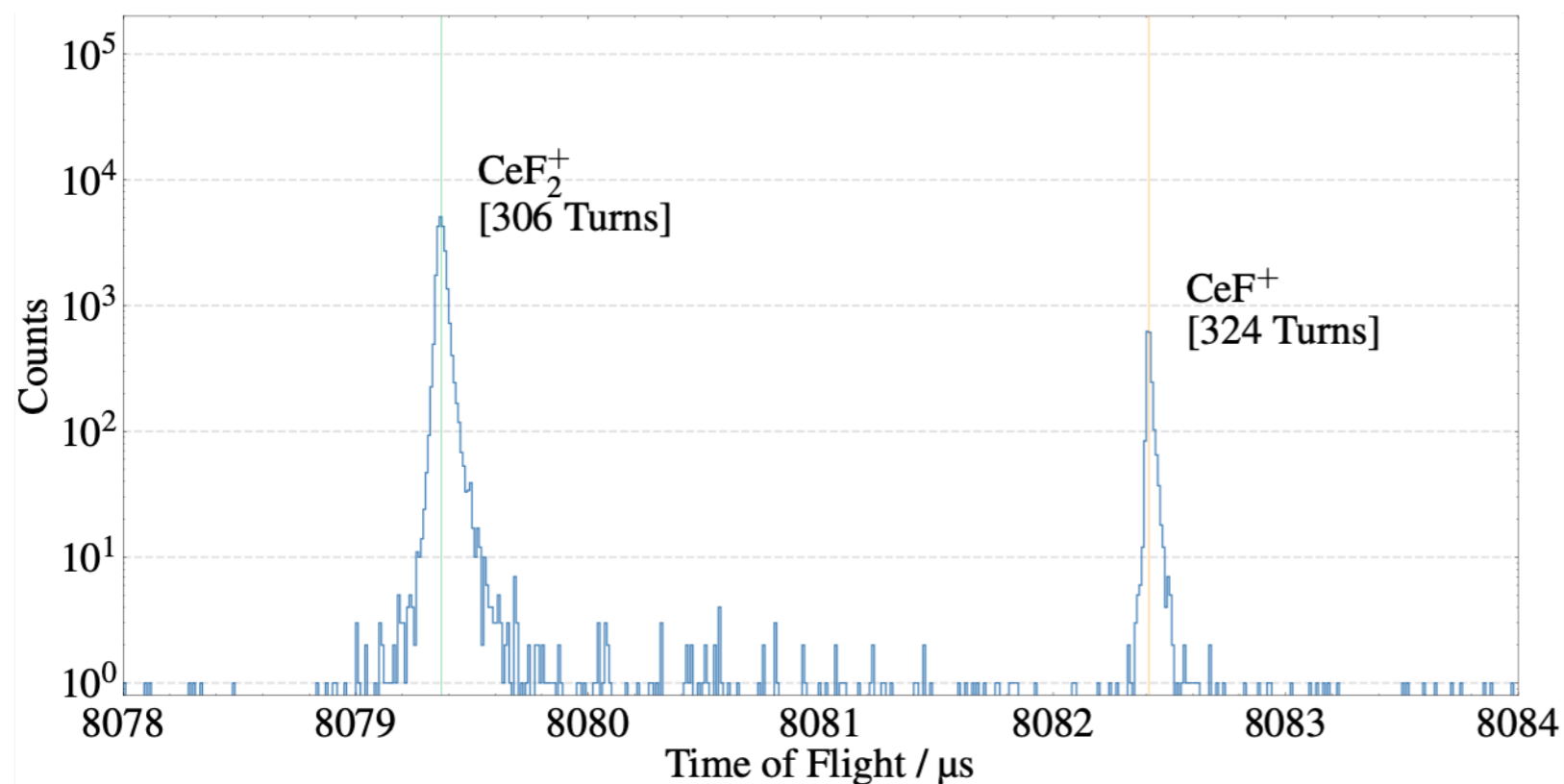
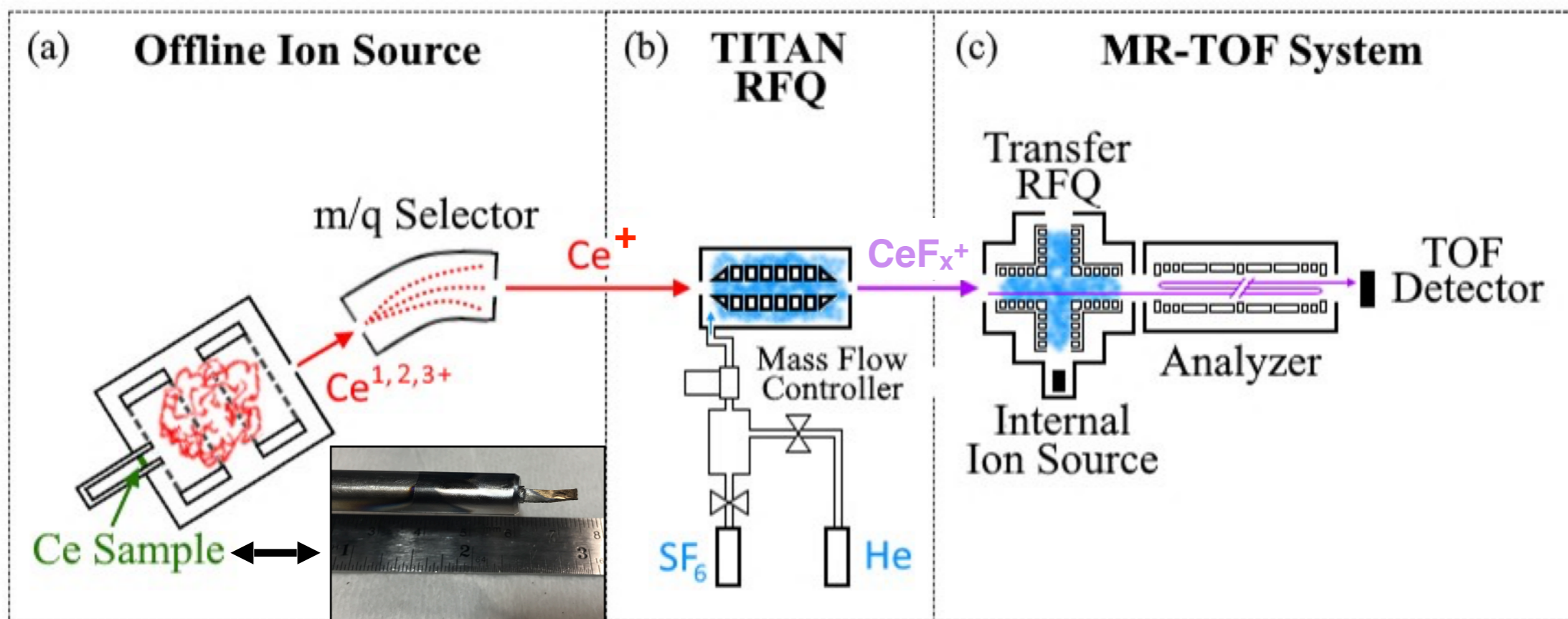
# Molecular formation@TITAN



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

actinides

# Molecular formation@TITAN

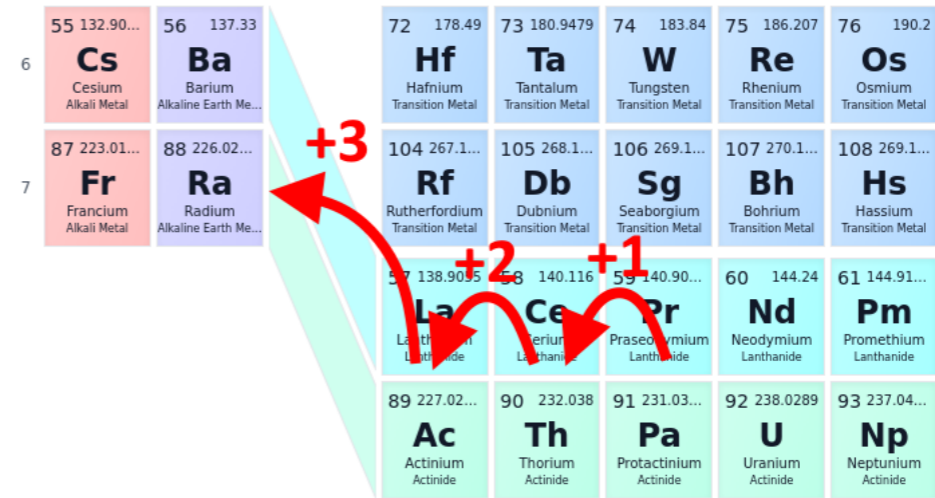
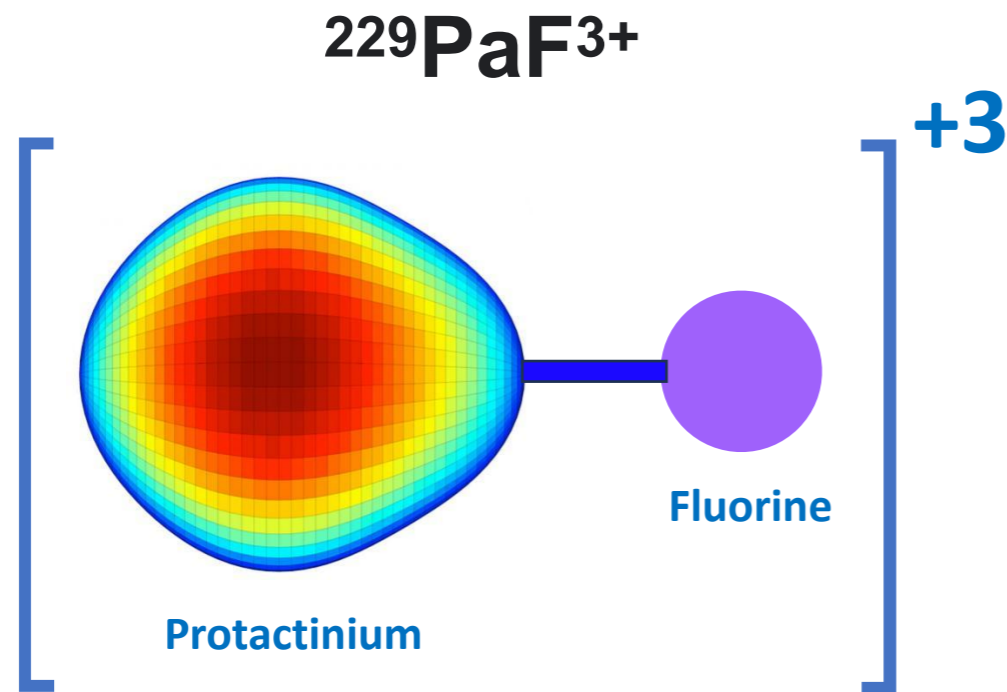


57 La	58 Ce	59 Pr	60 Nd	<b>lanthanides</b>
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## Conclusions:

- $\text{CeF}^+$  successfully formed
- excellent prospect for  $\text{ThF}^+$

# 'Highly' charged radioactive molecules



13

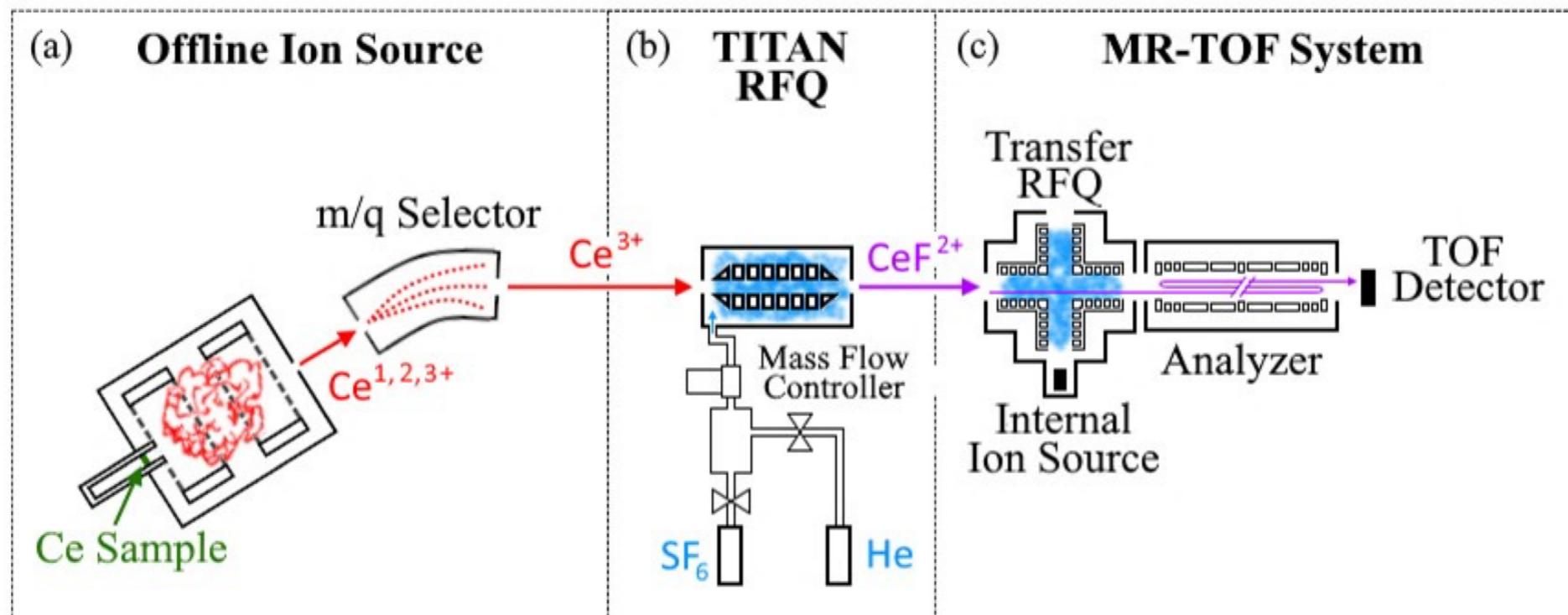
C. Zülch et al., arXiv 2203.10333 (2022)

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

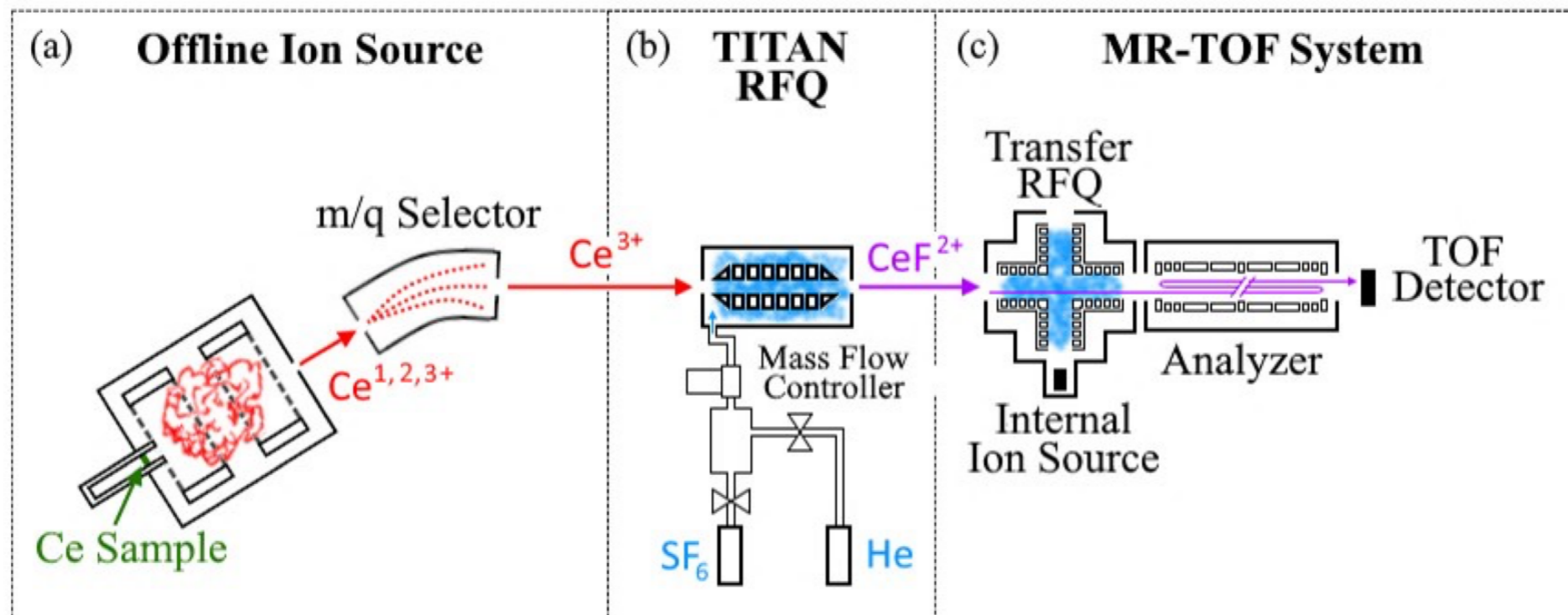
Species	Schiff Scaling Factor (relative to $^{225}\text{Ra}$ )
$^{225}\text{Ra}$	:= 1 (~200x larger than 199Hg)
$^{229}\text{Th}$	2
$^{227}\text{Ac}$	6
$^{229}\text{Pa}$	40

other iso-electronic molecules:  $\text{AcF}^+$ ,  $\text{ThF}^{+2}$

# Formation of $\text{CeF}^{2+}$ at TITAN



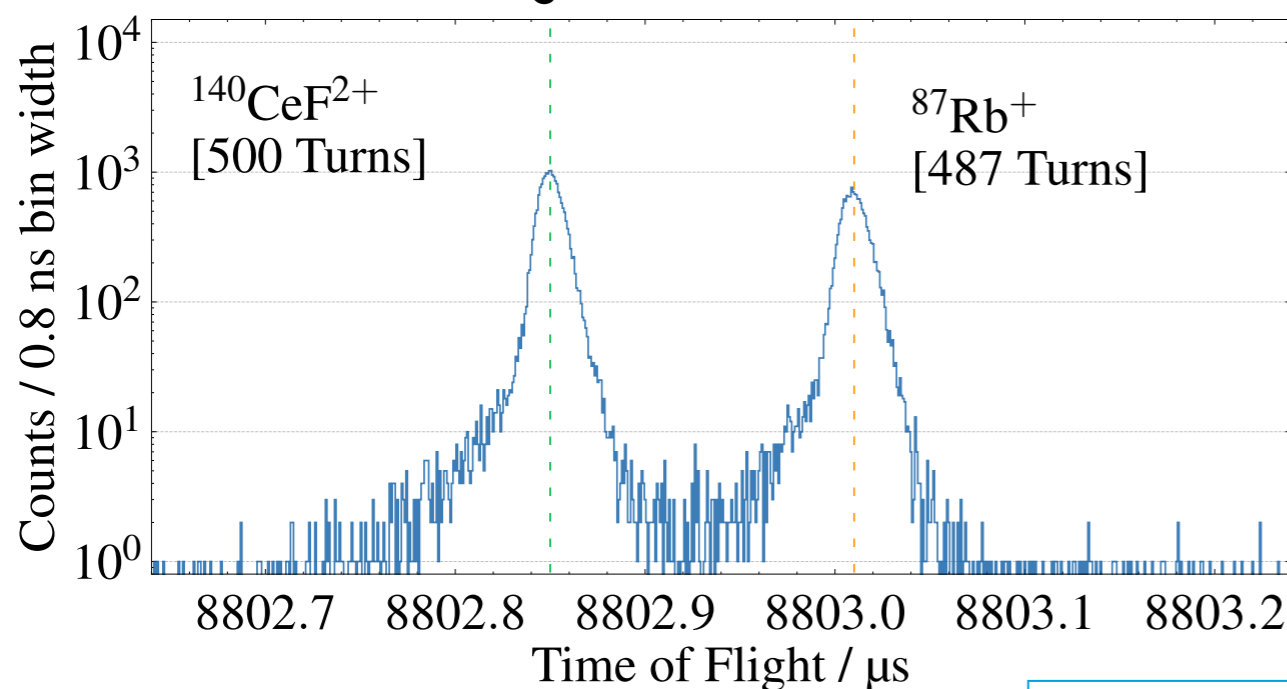
# Formation of $\text{CeF}^{2+}$ at TITAN



14

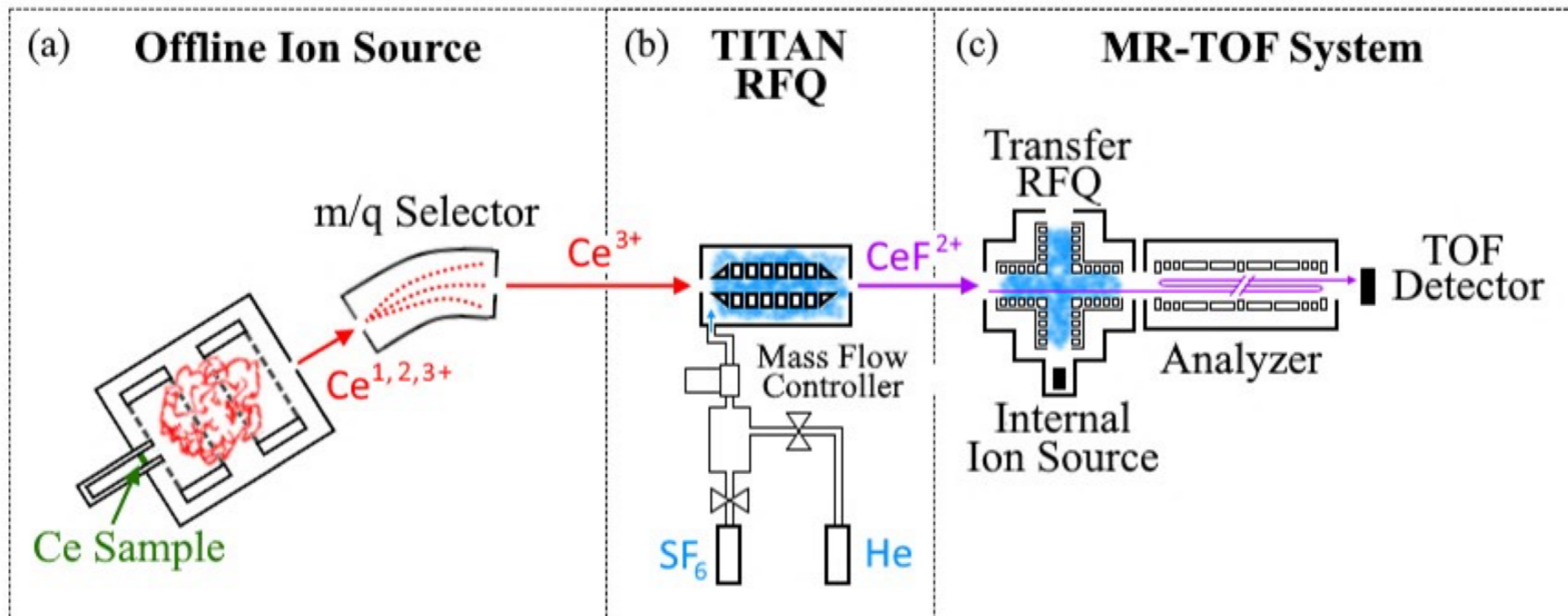
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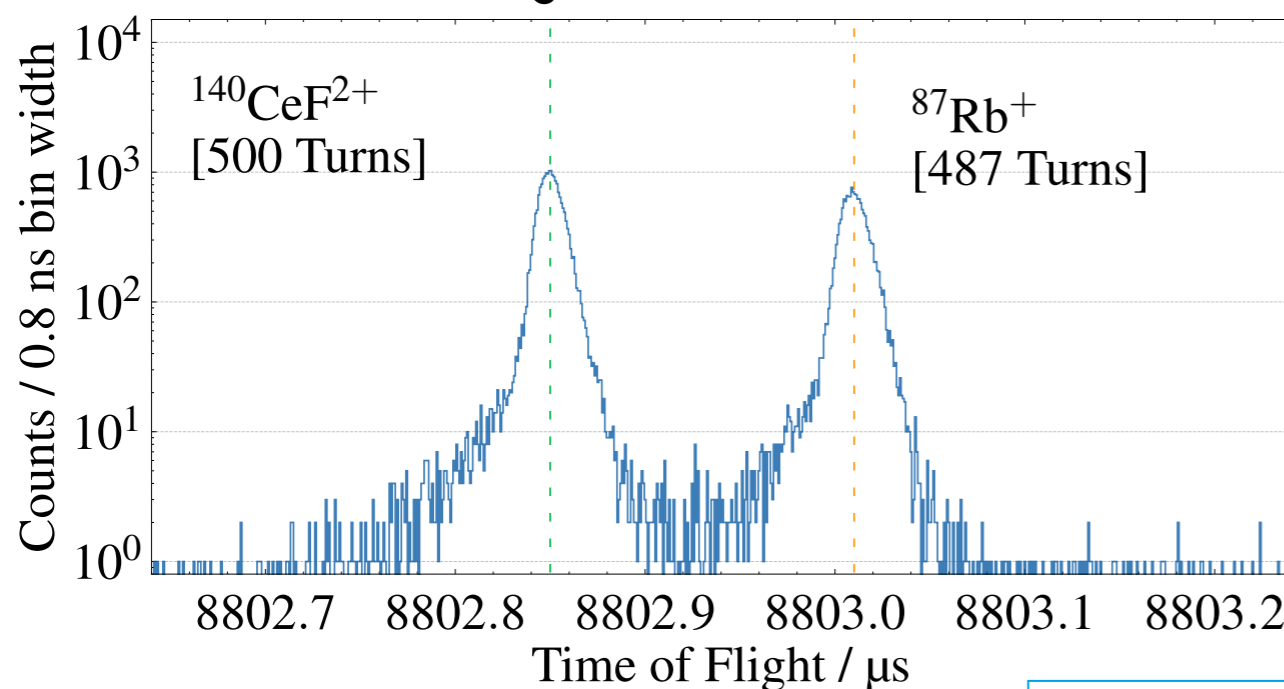


*R. Simpson, et al., submitted*

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14



*R. Simpson, et al., submitted*

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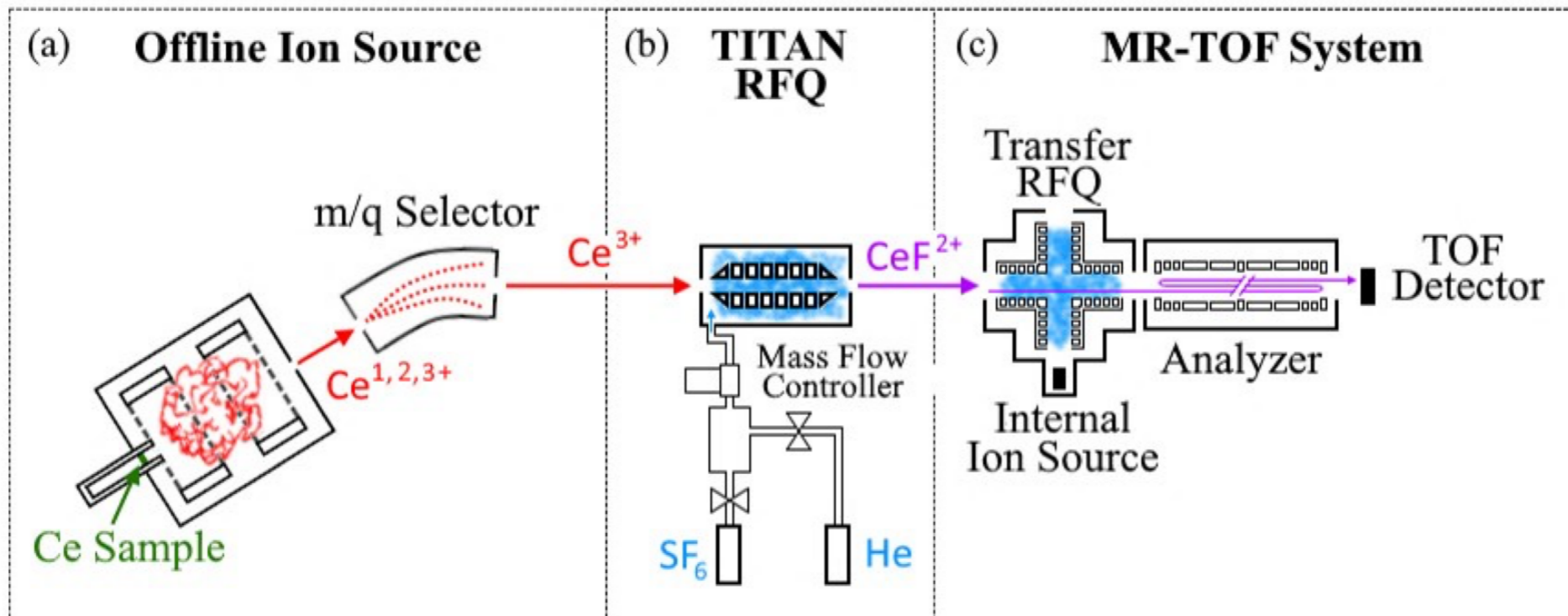
## Additional work:

- theoretical sensitivity factors
  - ➔  $\text{CeF}^{2+}$  interesting by itself
  - ➔ surrogate for  $\text{PaF}^{3+}$

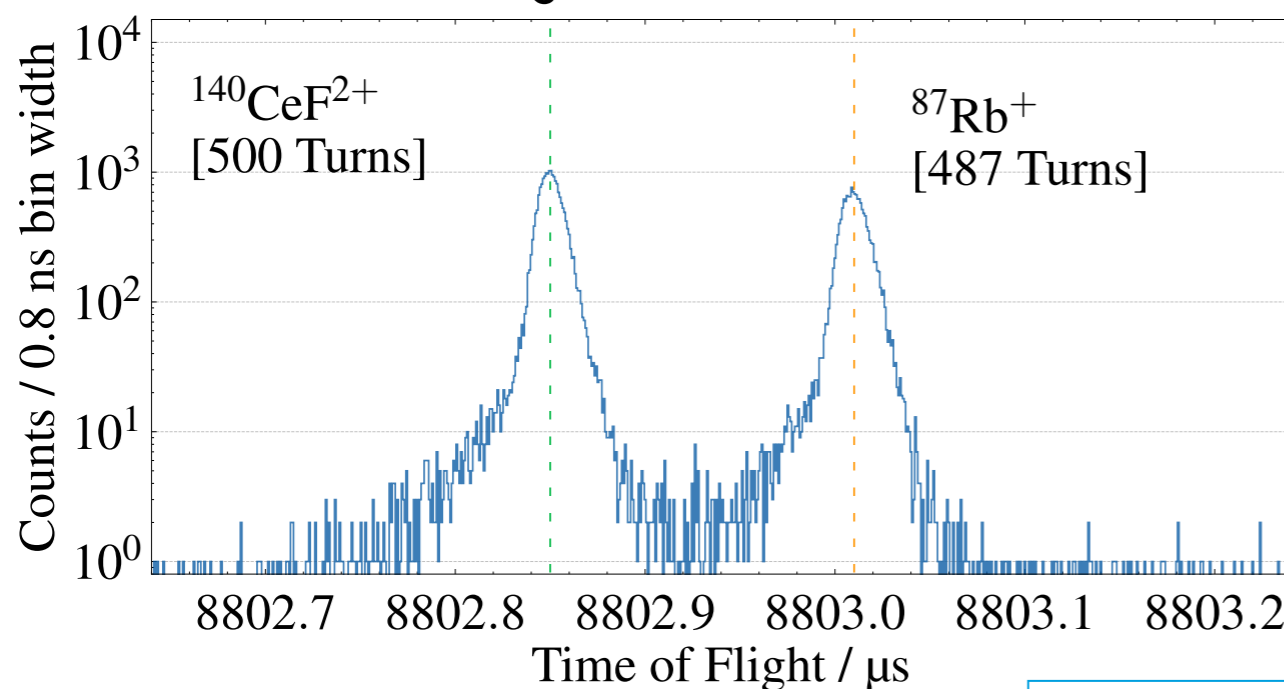
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*C. Zülch et al., arXiv 2203.10333*

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## complementary approach:

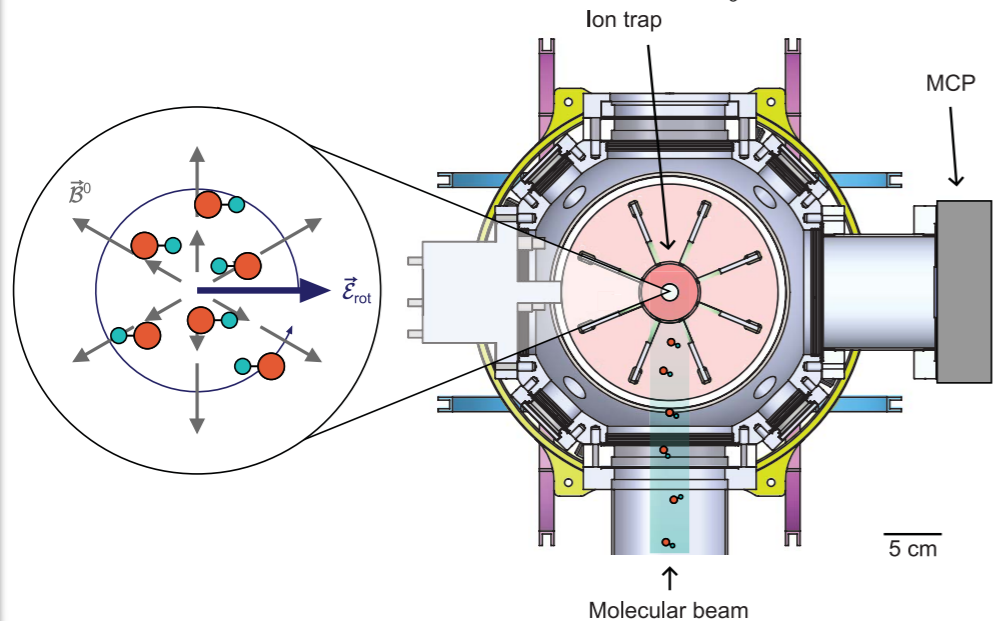
- charge breeding of molecule
- successfully employed at TITAN EBIT

# Nuclear Schiff Moment (NSM) of $^{19}\text{F}$

## $^{180}\text{Hf}^{19}\text{F}^+$ @ JILA

energy splitting between EDM-sensitive molecular states

$$hf = (-14.6 \pm 22.8_{\text{stat}} \pm 6.9_{\text{syst}}) h \mu\text{Hz},$$



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

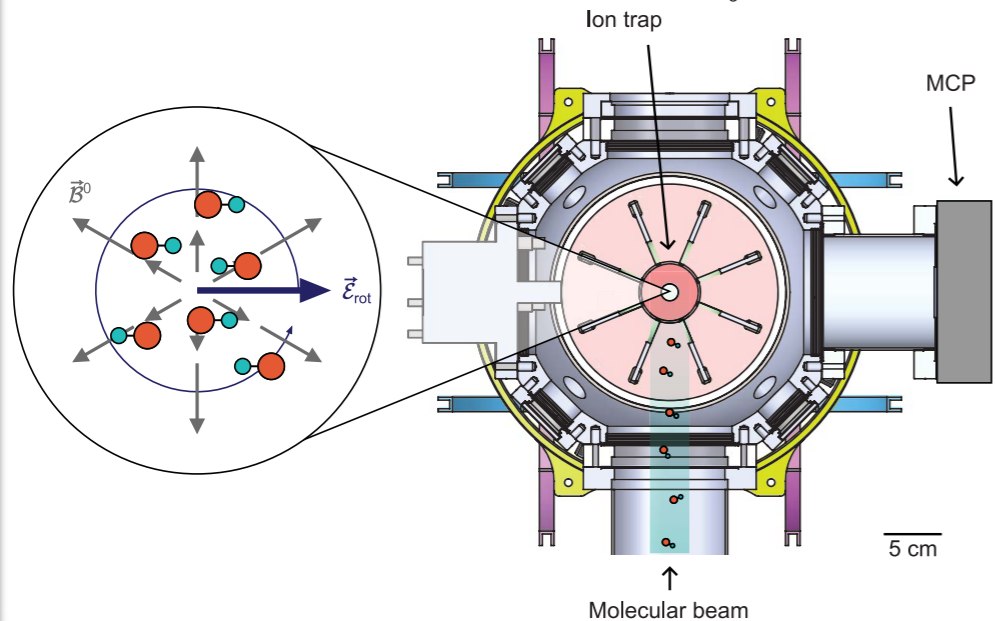
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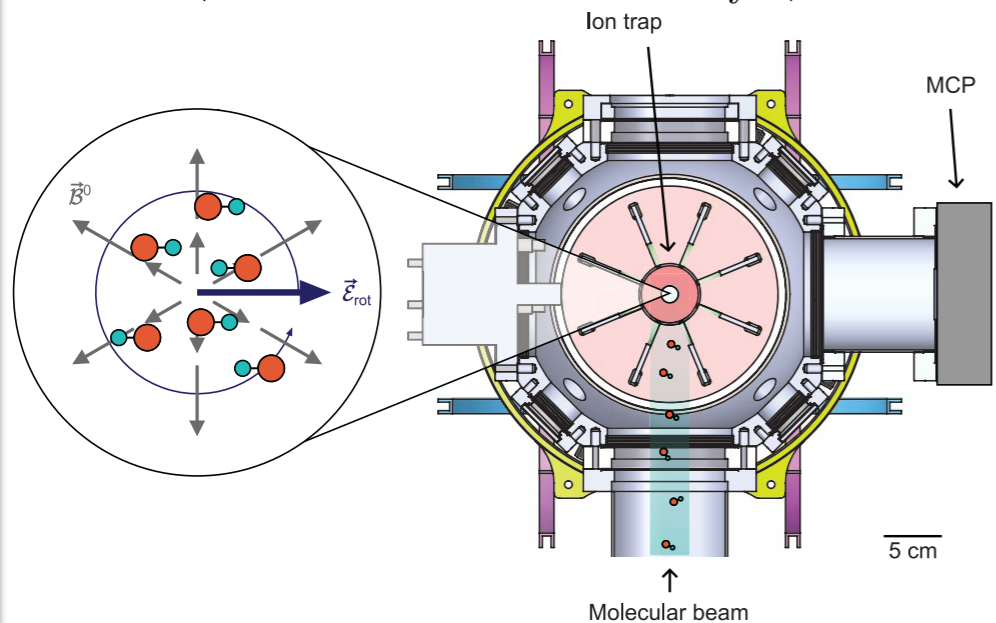
$$\Delta E_{\text{NSM}} = W_S S \left\langle \frac{\mathbf{I} \cdot \hat{n}}{I} \right\rangle$$

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molecular sensitivity  $W_S$ : **(quantum chemistry)**

Molecule	Molecular state	$W_S \left( \frac{e}{4\pi\epsilon_0 a_0^4} \right)$	
$\text{HfF}^+$	$a^3\Delta_1$	115	by L. Cheng

$$|S(^{19}\text{F})| < 9.0 \times 10^{-9} e \text{ fm}^3 \quad (90\% \text{ confidence level})$$

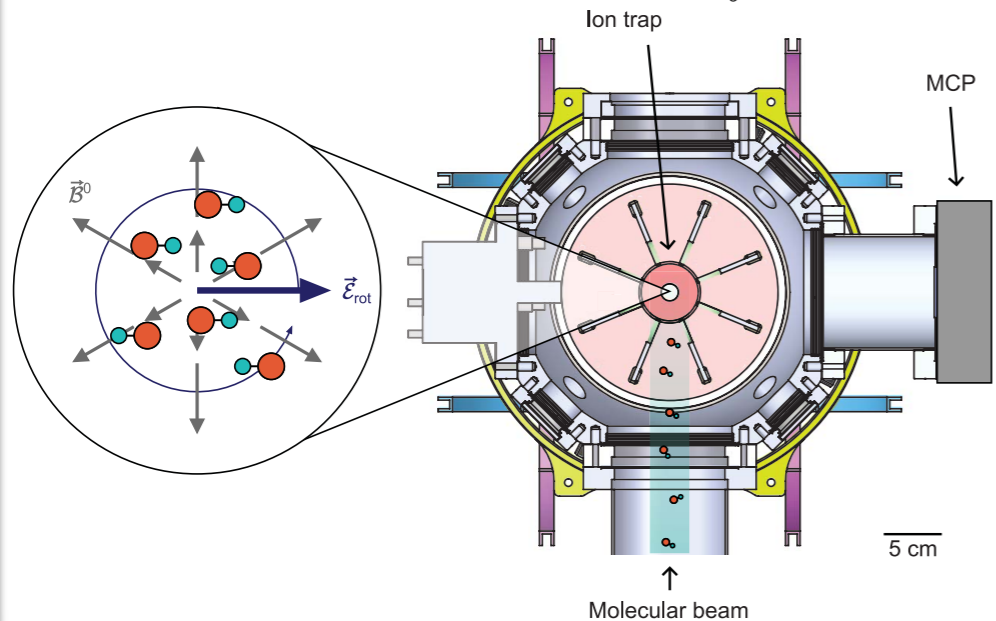
*K.B. Ng et al., Phys. Rev. Lett. 136, 112501 (2026)*

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### 1<sup>st</sup> nuclear *ab initio* calculation of a NSM

no-core shell model by P. Navratil & S. Foster

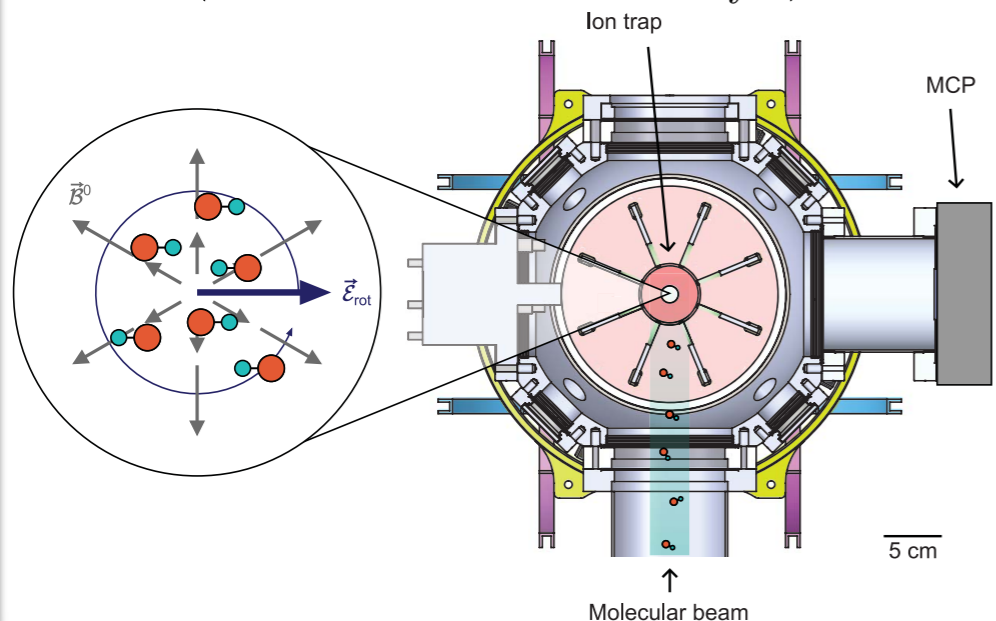
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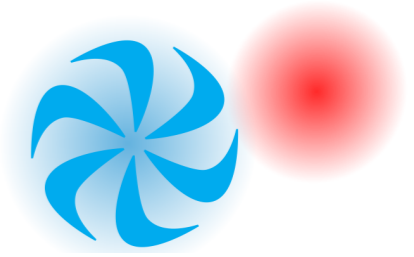
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Quantity	Limit
$ \bar{g}_0 $	$2.3 \times 10^{-8}$
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$ \bar{g}_2 $	$3.3 \times 10^{-8}$

# Summary



- **Radioactive Molecules**

- new science path
- intriguing & unexplored **probes for New Physics**
- **time-reversal-violating nuclear Schiff moments**
  - ◆ **octupole deformed nuclide**
  - ◆ **in polar molecule**

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- dedicated laboratory for radioactive molecules & precision studies
- FrX, ThF<sup>+</sup>, and towards PaF<sup>+3</sup>

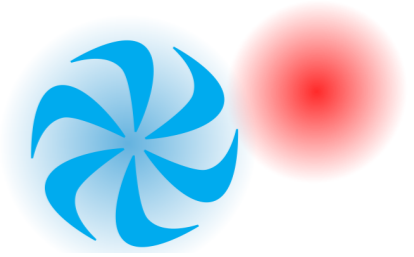
- synergies & collaboration to CNRS

**GANIL**



↕  
Fr offline ion-source

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→ synergies & collaboration to CNRS

**GANIL**



Fr offline ion-source

**support to strengthen & expand?**

# Present RadMol@TRIUMF collaboration

## TRIUMF

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C. Hoehr  
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 **JOHNS HOPKINS**  
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 **MIT**  
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 university of  
 groningen S. Hoekstra

 UNIVERSITY OF  
MARYLAND L. Orozco


## Theory Support

Philipps  Universität  
Marburg R. Berger

 **TEMPLE**  
UNIVERSITY® S. Kotochigova

 UNIVERSITY OF  
DELAWARE M. Safronova

 **JOHNS HOPKINS**  
UNIVERSITY L. Cheng

 university of  
 groningen A. Borschevsky

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 **LAURIER** S. Ghose  
WILFRID LAURIER UNIVERSITY

## New UBC-CNRS collaboration

 **iJC Lab**  
Irène Joliot-Curie  
Laboratoire de Physique  
des 2 Infinis S. Franchoo  
V. Manea  
A. Vicente

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TRIUMF-local RadMol team

A. Carpentier, C. Charles, L. Croquette, S. Cui, R. Simpson,  
L. Croquette, K.B. Ng, S. Malbrunot-Ettenauer  
Former team members: I. Belosevic, E. Frieling, P. Justus, E. Riley,  
A. Teigelhöfer



## TITAN



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## FrPNC

