# Towards CP-Violation Measurements with RaX Molecules

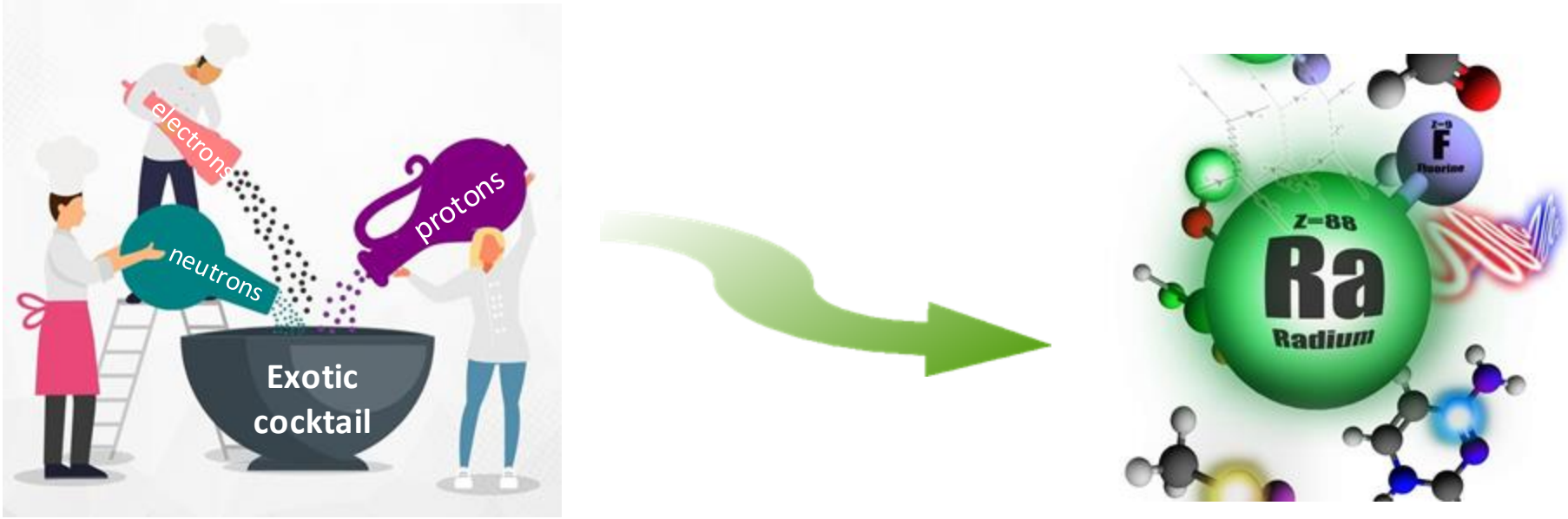


Figure modified from https://sphereofinfluence360.com/

**Ronald Fernando Garcia Ruiz**, ***MIT***

**Electric Dipole Moments: Experimental and Theoretical Horizons**
**Caltech-TDLI Workshop, Los Angeles, May 2025**

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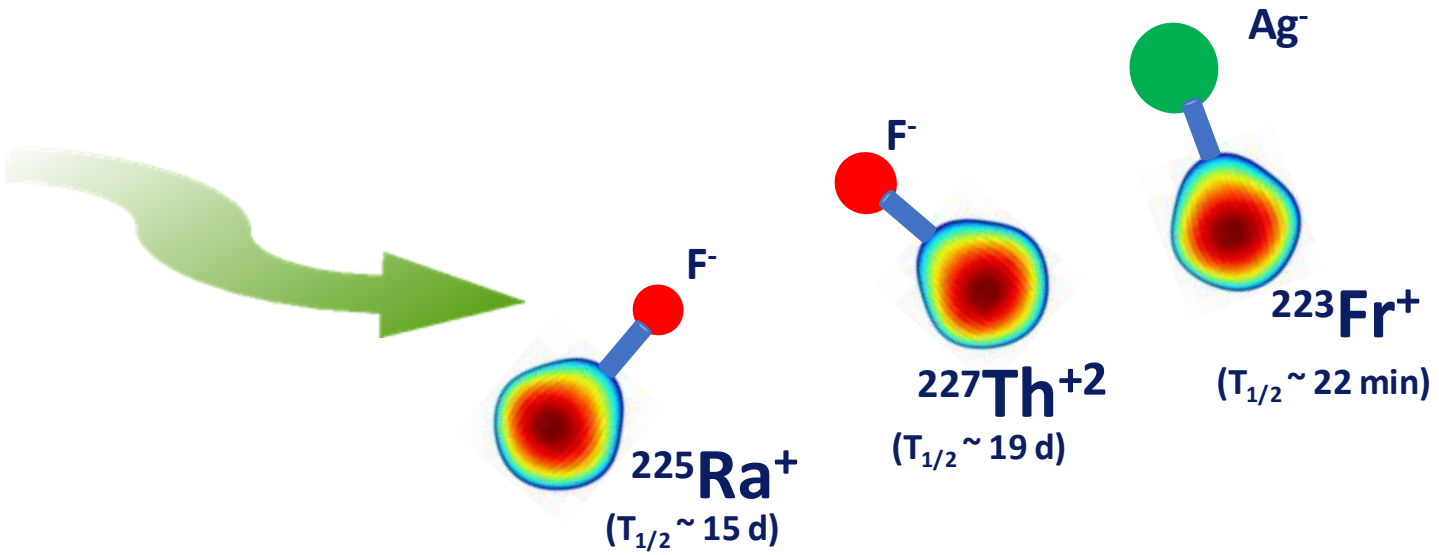


**Ronald Fernando Garcia Ruiz**, ***MIT***

**Electric Dipole Moments: Experimental and Theoretical Horizons**
**Caltech-TDLI Workshop, Los Angeles, May 2025**

## Graduate Students

**S. Udrescu**
(-> U. Chicago)

**A. Brinson**
NSF Fellowship

**H. Kakiota**
Ezoe Memorial Fellowship

**S. Moroch**
Hertz Foundation Fellowship

**F. Pastrana**
Santo Domingo Fellowship

**D. Gonzales**
(Harvard)

**J. Munoz**
MIT Fellowship

**M. Fulghieri**

**F. Shungo**

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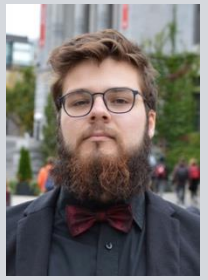
**S. Wilkins**

**J. Karthein**
Texas A&M

**S. Ebadi**
Pappalardo Fellowship

**A. Jadbabaie**
NSF MPS Ascend Fellow



**A. Belley**
NSERC Fellow

## PI

**R.F. Garcia Ruiz**

## Undergraduate Students

**N. Ching-Hau**

**S. Munoz**

**Y. Lewis**

**R. Hernandez**

**S. Becerra**

FRIB

Bates Lab

# Thanks to…

CRIS

MANCHESTER 1824

KU LEUVEN

CERN

Massachusetts Institute of Technology

北京大学 PEKING UNIVERSITY

UNIVERSITY OF GOTHENBURG

DEPARTMENT OF ENERGY · UNITED STATES OF AMERICA

FRIB

## RaX Collaboration

Caltech

Nuclear & Atomic & Molecular

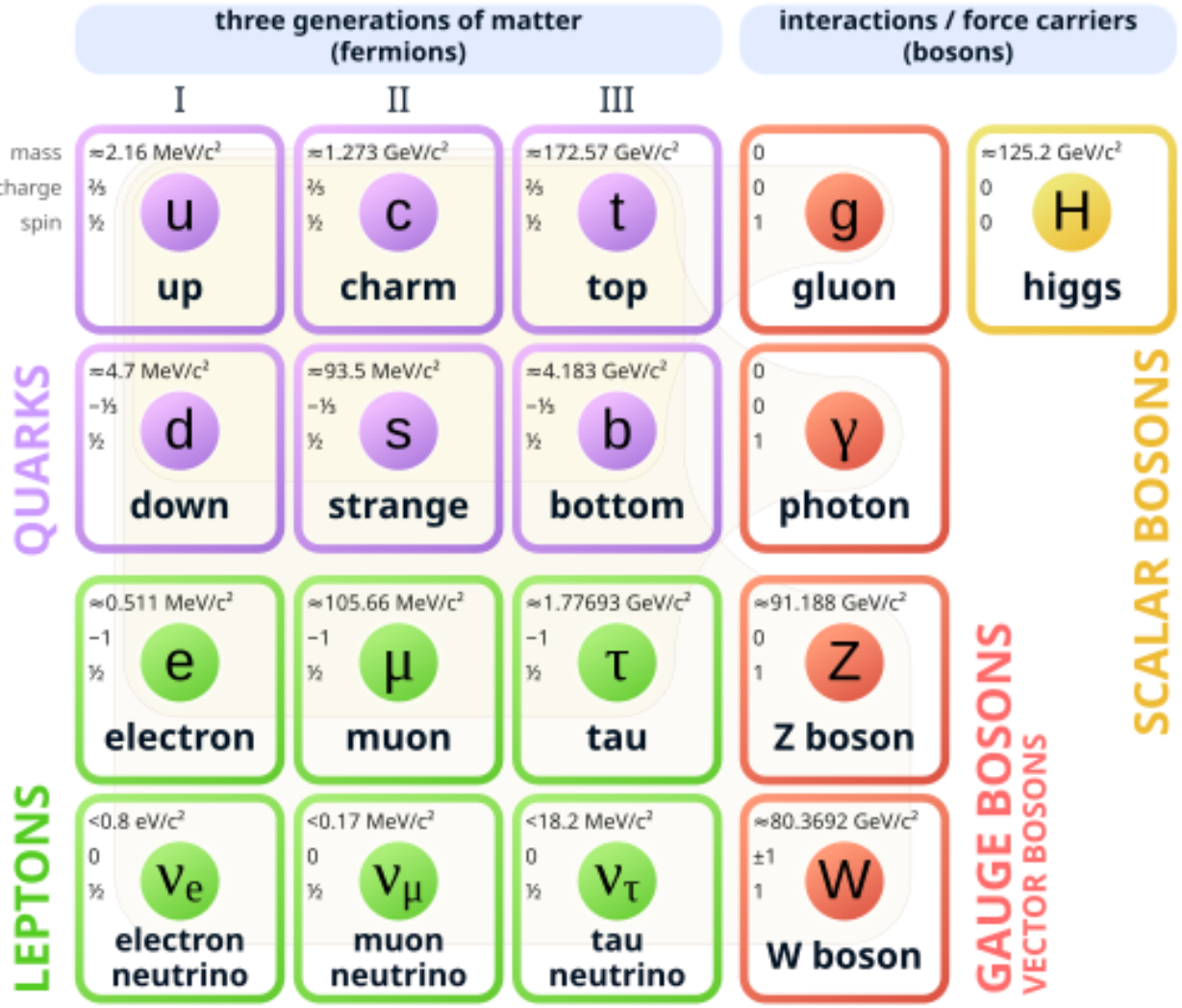
Theory

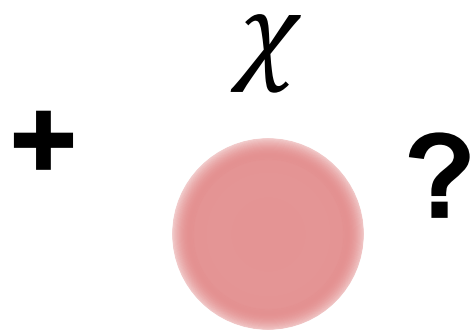
Experiment

J. Dobaczewski (York)
J. Holt (TRIUMF)
R. Stroberg (U ND)
W. Nazarewicz (FRIB/MSU)…

R. Berger (Malbroune)
A. Borchevsky (Groningen)
L. Skripnikov (Petersburg)

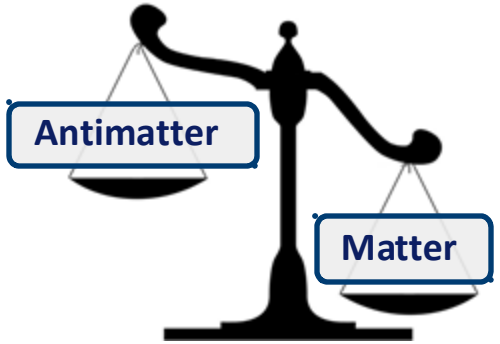
# What are the fundamental particles and forces of nature?



# Major Open Questions in Physics and Cosmology

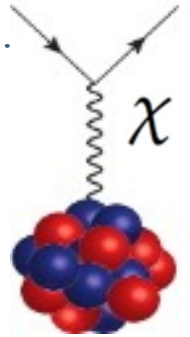
**Why is there more matter?**




**Strong CP problem**

$$\mathcal{L} = \theta \frac{1}{16\pi^2} F^a_{\mu\nu} \tilde{F}^{\mu\nu a}$$

**Are there new particles?**




**New sources of Time-reversal (CP) violation in nuclei can provide answers to these problems**

Fundamental Symmetries

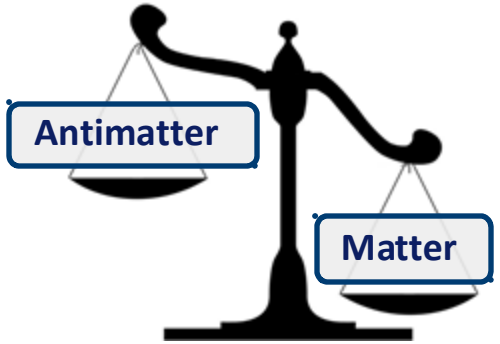
Parity (P): $r \rightarrow -r$

Time (T): $t \rightarrow -t$

Charge (C): $c \rightarrow \bar{c}$

# Major Open Questions in Physics and Cosmology

**Why is there more matter?**

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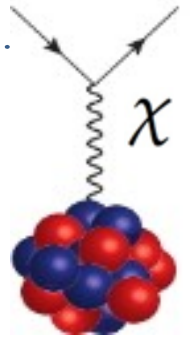
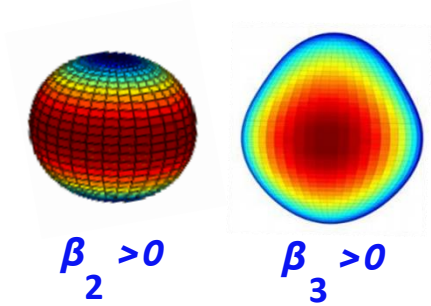
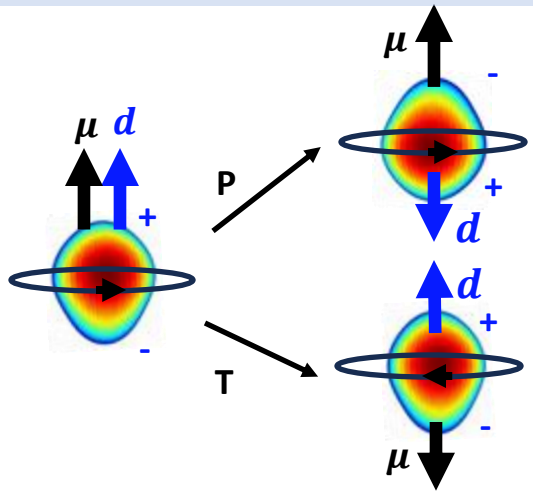



**New sources of Time-reversal (CP) violation in nuclei can provide answers to these problems**

**Electric Dipole Moment (d)** ⇄ **Time-reversal violation**




Fundamental Symmetries

Parity (P): $r \rightarrow -r$

Time (T): $t \rightarrow -t$

Charge (C): $c \rightarrow \bar{c}$

# Why Radioactive molecules?

[Sandars Phys. Rev. Lett. 18, 1396 (1967)]
[Hudson et al. Nature 473 493 (2011)]

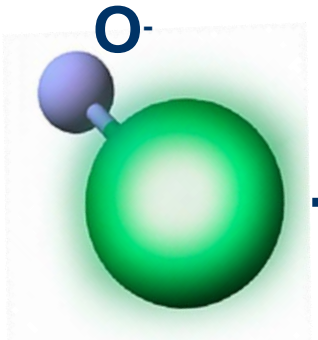
**Nuclear spin = 0**

$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + d_e E_{eff}$$

[1 eV=241.8 THz]

eV ~2 $10^{-2}$ $10^{-5}$ $<10^{-20}$

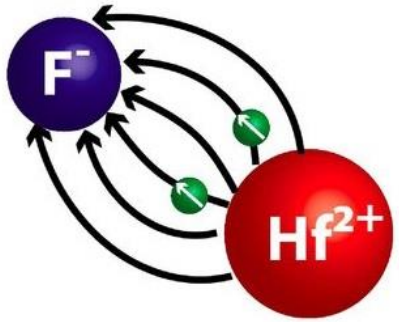
Talks: Kia Boon Ng
Nick Hutzler




$Th^+$

$E_{eff}$~80 GV/cm

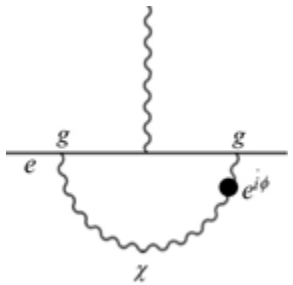
[ACME, Nature 562, 355 (2018)]




$E_{eff}$~20 GV/cm

[Roussy et al. Science 381, 46 (2023)]

$|d_e| \leq 4.1 \times 10^{-30}\ e \cdot cm$

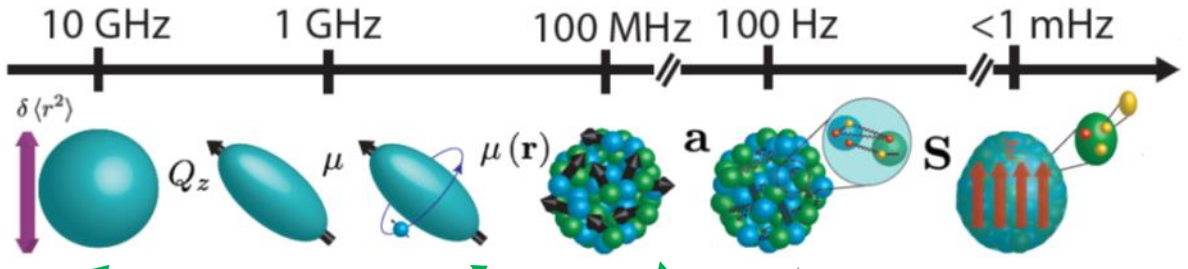



$$d_e \sim \mu_B \left(\frac{g^2}{2\pi}\right)^N \left(\frac{m_e}{m_\chi}\right)^2 \sin\phi$$

Probing physics @ TeV scale!

# Why Radioactive molecules?

Molecules with heavy, exotic nuclei: Ra(Z=88)




## Nuclear spin > 0

$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTVe} + d\,E_{eff}$$

[1 eV=241.8 THz]

| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-18}$ | $<10^{-20}$ |
|---|---|---|---|---|---|---|---|

$$\sim O_{Nucl}\, E_{mol}$$

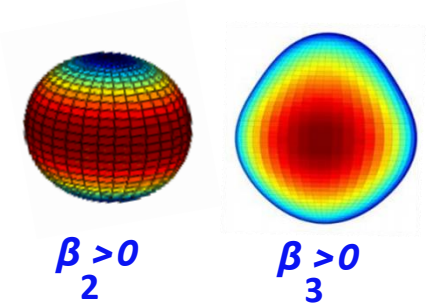
- ✓ Large Z, A
- ✓ Nuclear spin I >0
- ✓ $\beta_2$ , $\beta_3 > 0$

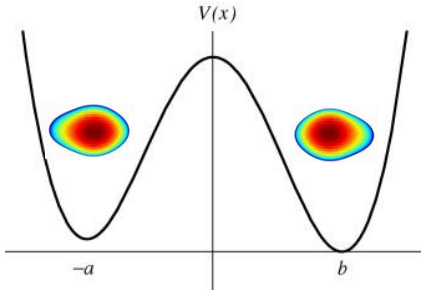
**Nuclear**

$$\sim Z A^{2/3} \beta_2 \beta_3^2 / (E^N_+ - E^N_-)$$

**Molecule**

$$\sim Z^3 / (E^e_+ - E^e_-)$$



# Radioactive molecules => Best of all worlds!

- ✓ Large Z, A
- ✓ Nuclear spin I >0
- ✓ $\beta_2$ , $\beta_3$ > 0

**Enhanced in actinide, short-lived nuclei:**
**Fr (Z=87), Ra (Z=88), Th (Z=90), …**

($T_{1/2}$ ~ 22 min) ($T_{1/2}$ ~ 15 d) ($T_{1/2}$ ~ 19 d)

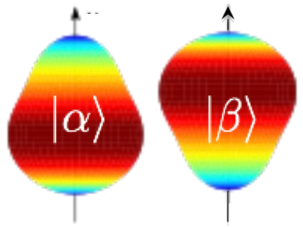
**Nuclear X Molecule**

$>10^3$ $>10^3$ => $>10^6$

[Gaffney et al. Nature 497, 199 (2013)]
Ra @ ANL [Parker et al. PRL 114, 233002 (2015)]
[Auerbach et al. PRL 76, 4316 (1996)]

$^{225}$Ra (Z=88)

$T_{1/2}$= 15 days




$^{225}$Ra

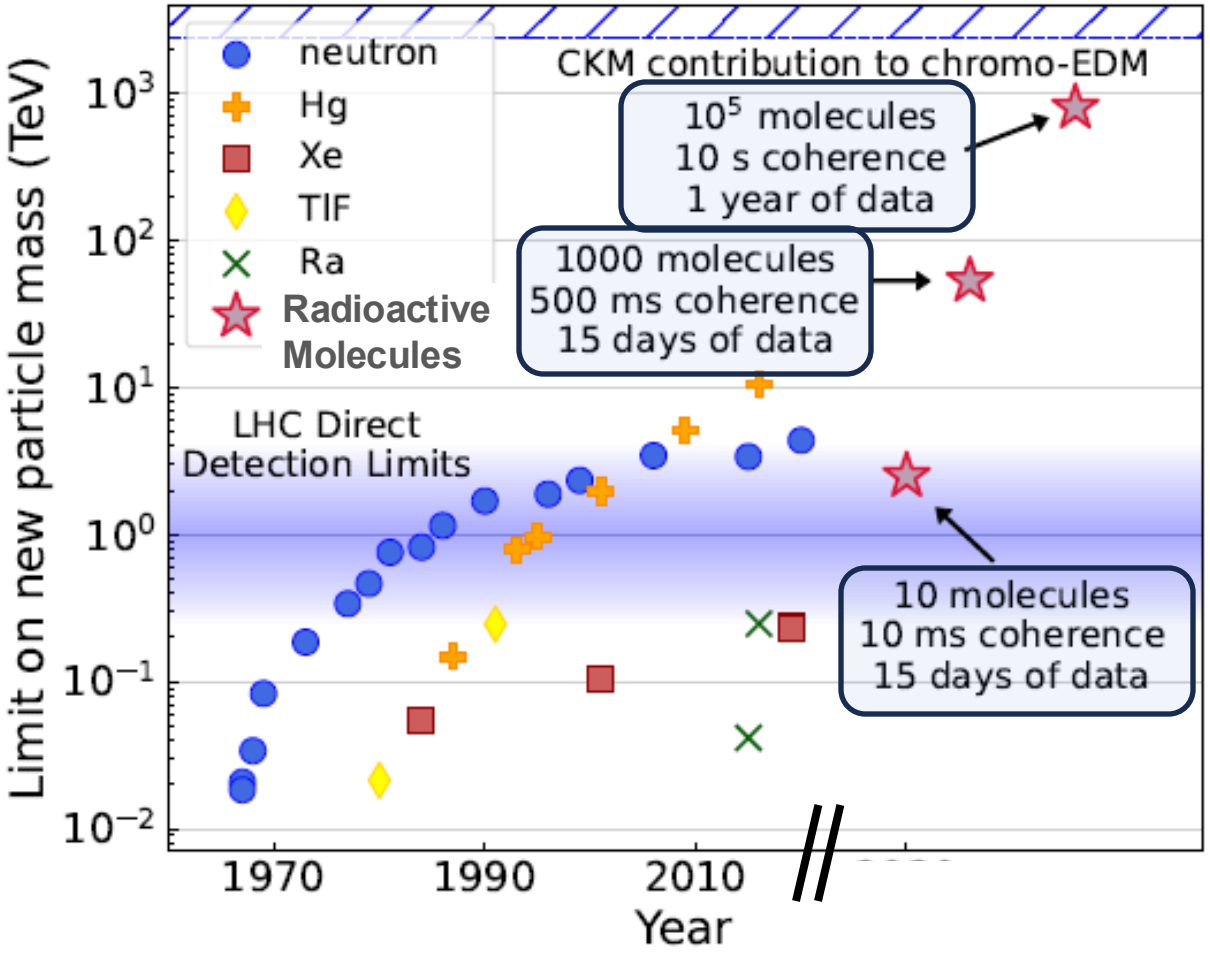

$^{225}Ra^+$ $F^-$

RaF: [Isaev et al. Phys. Rev. A 82, 052521 (2010)]

# Radioactive molecules => Best of all worlds!


Weak Force

Hadronic EDM Limits

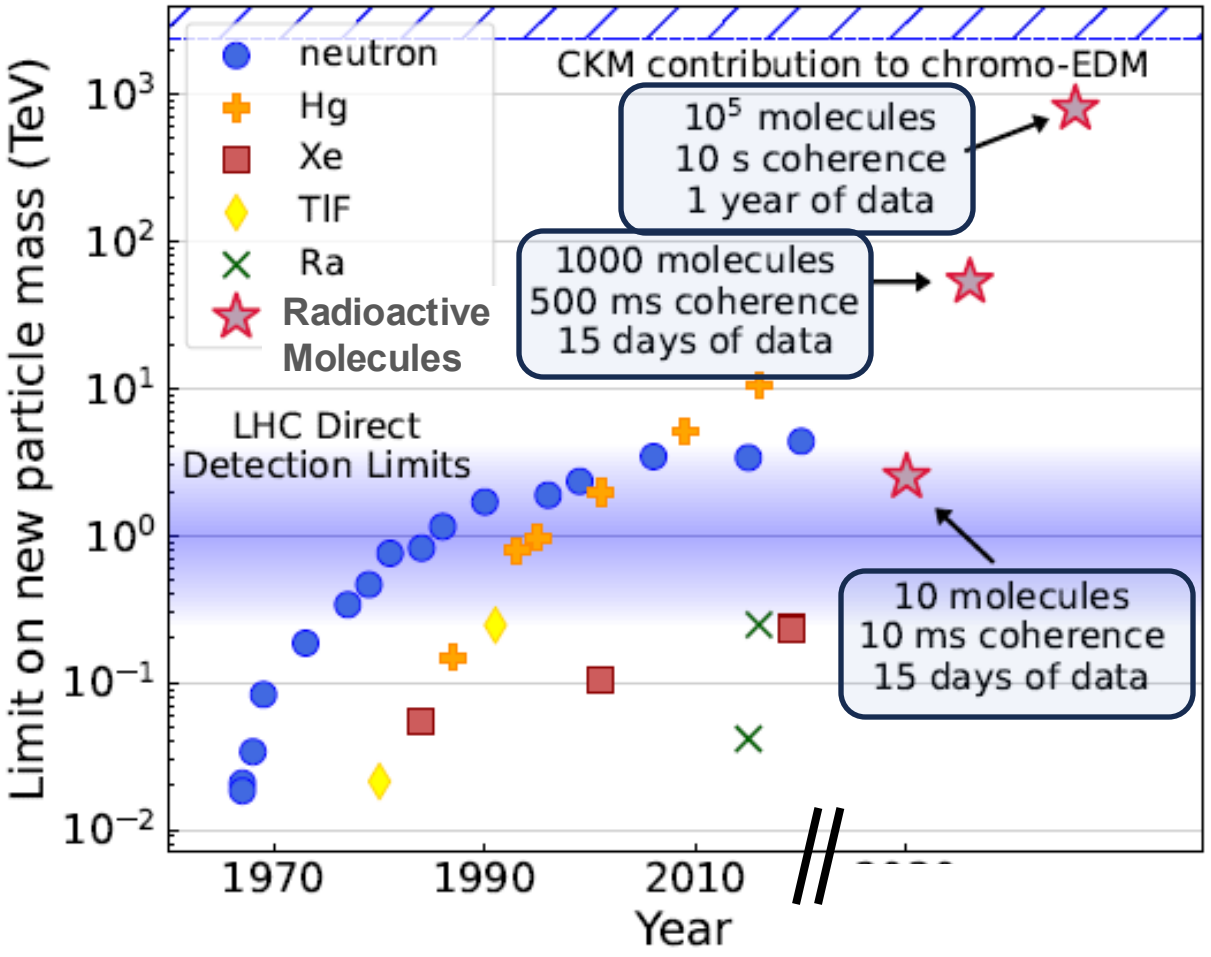



Window of Discovery

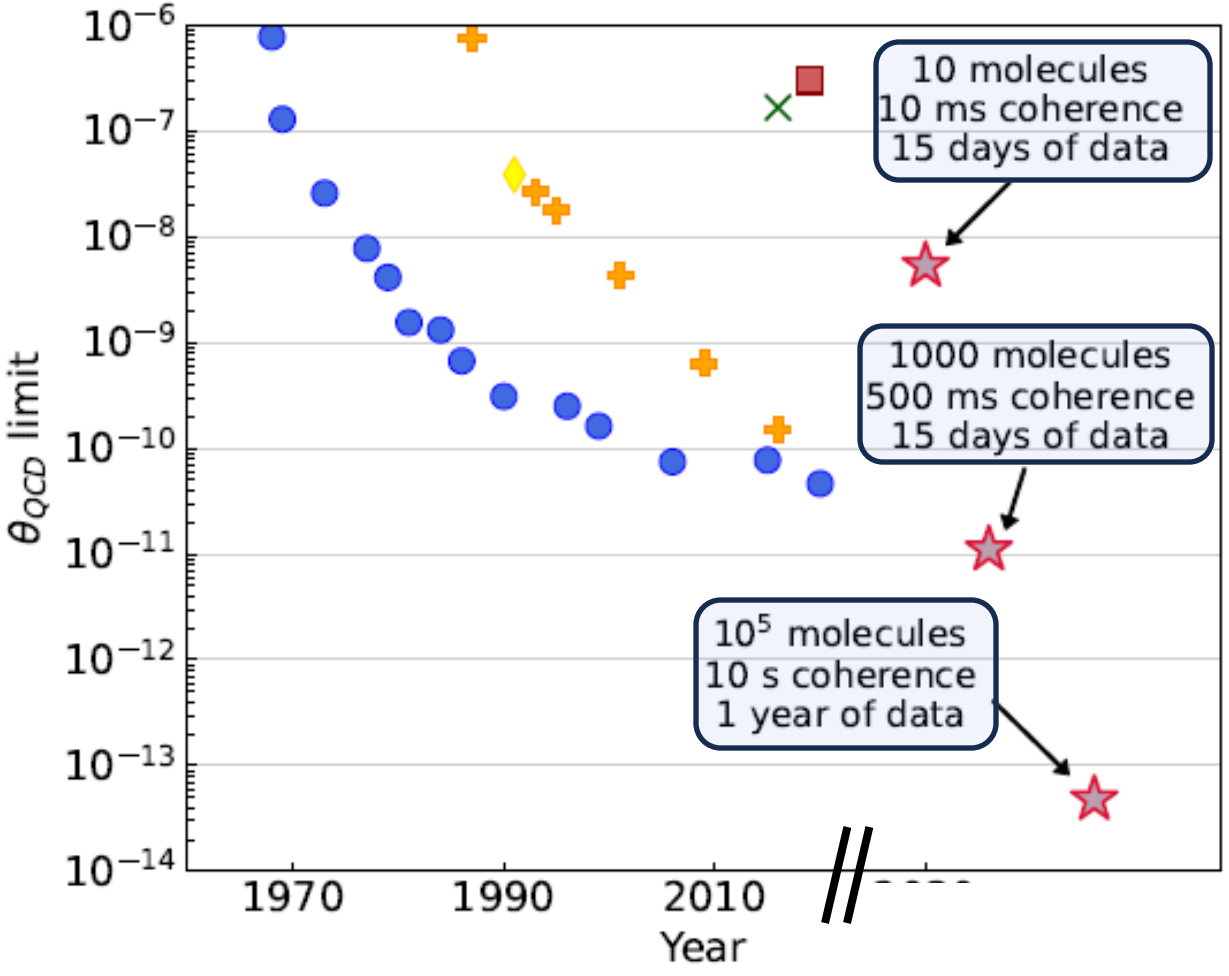
# Radioactive molecules => Best of all worlds!


## Hadronic EDM Limits

**Weak Force**

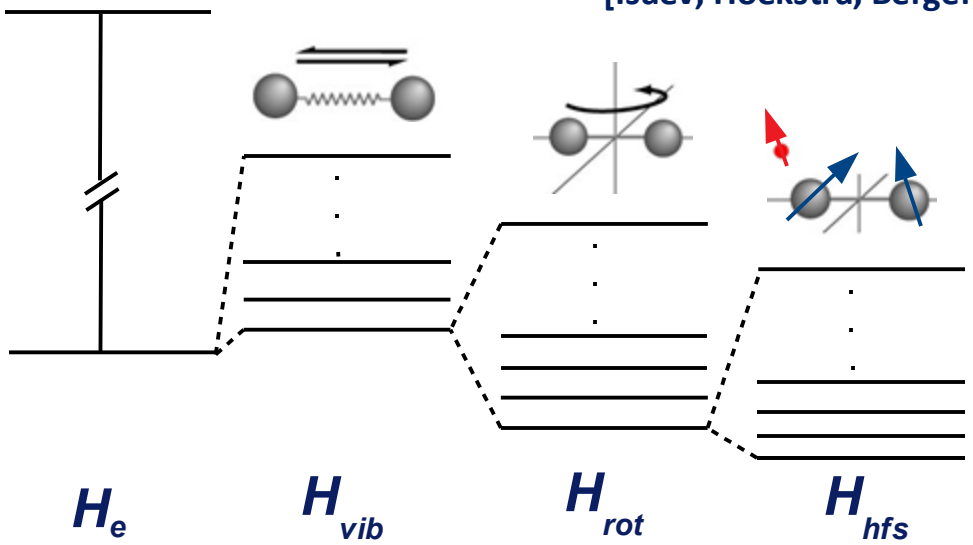



**Strong Force**



# Recent Results (RaF)


[Garcia Ruiz, Berger et al. CERN-INTC-2018-017 / INTC-P-546 (2018)]
[Isaev, Hoekstra, Berger, Phys. Rev. A 82, 052521 (2010)]



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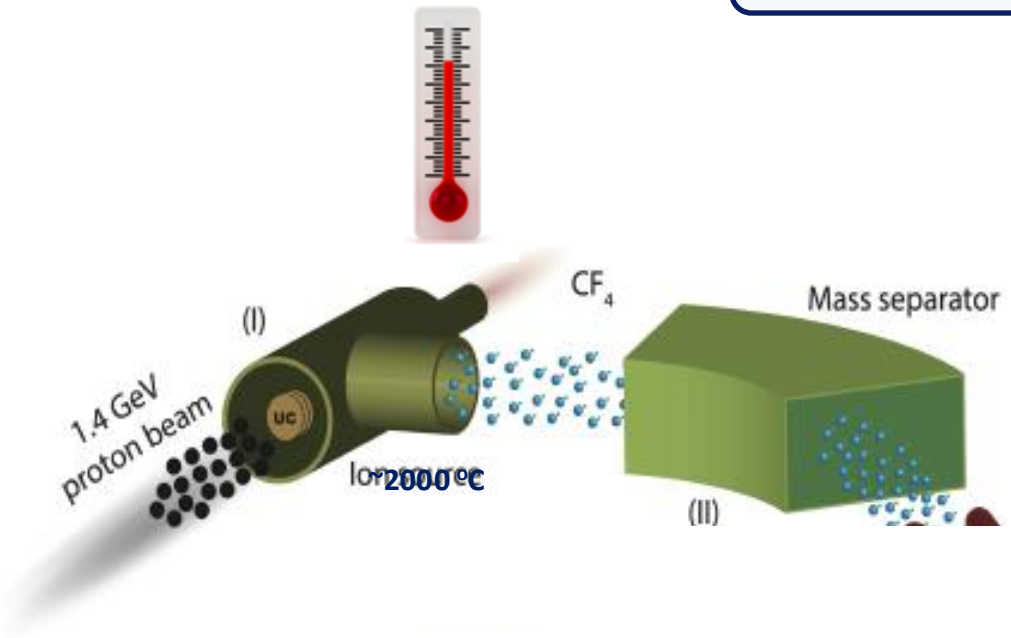


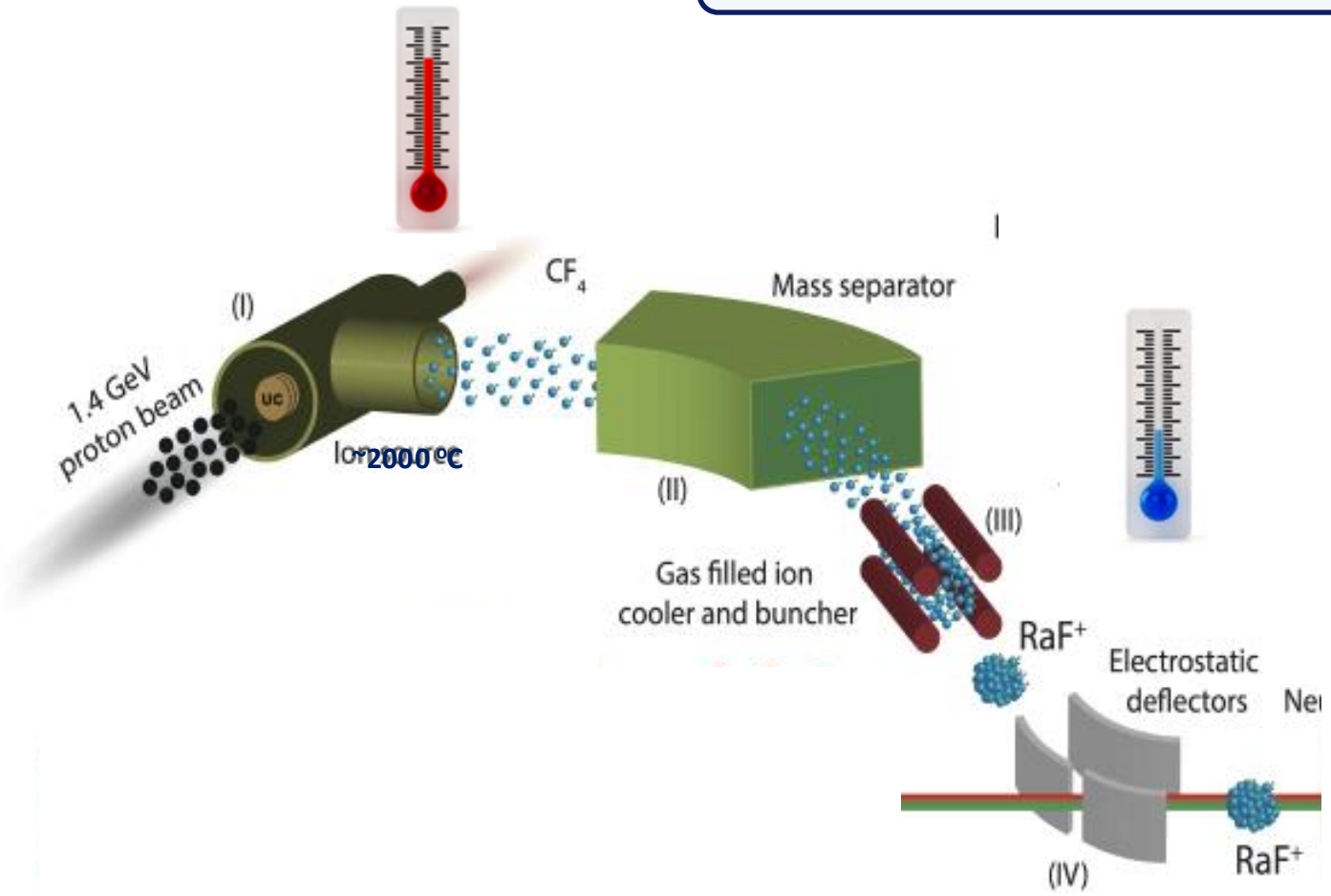

**Experimental Challenges:**

1. Extreme environments (>2000 °C)
2. Highly contaminated (1:$10^8$)
3. Have short lifetimes ( ~ days)
4. Small quantities (< 1 ng)

$$\mathrm{Ra}^{+} \xrightarrow{\mathrm{CF_4}} \mathrm{RaF}^{+}$$

[Garcia Ruiz et al. Nature 581, 396 (2020)]

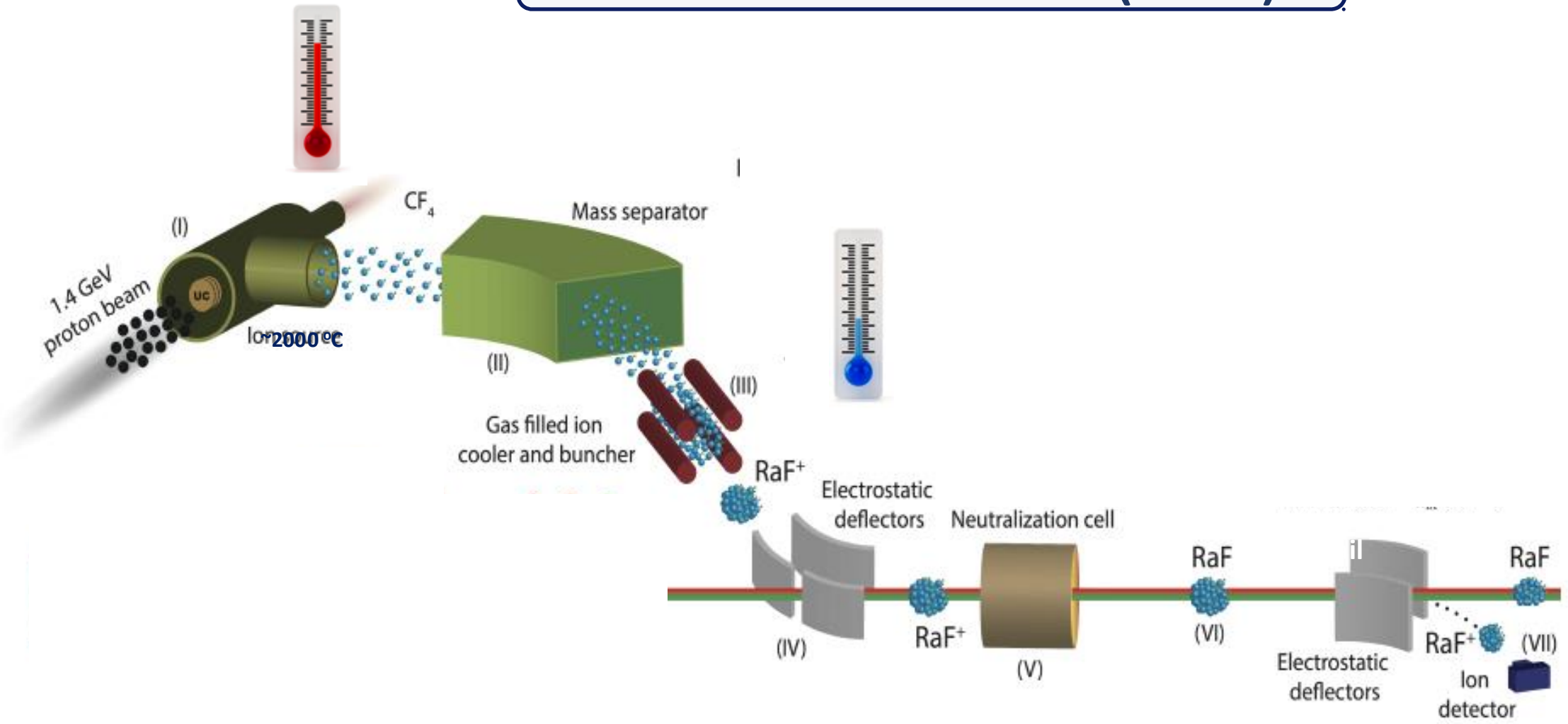
# Recent Results (RaF)




$$Ra^+ \xrightarrow{CF_4} RaF^+$$

[Garcia Ruiz et al. Nature 581, 396 (2020)]

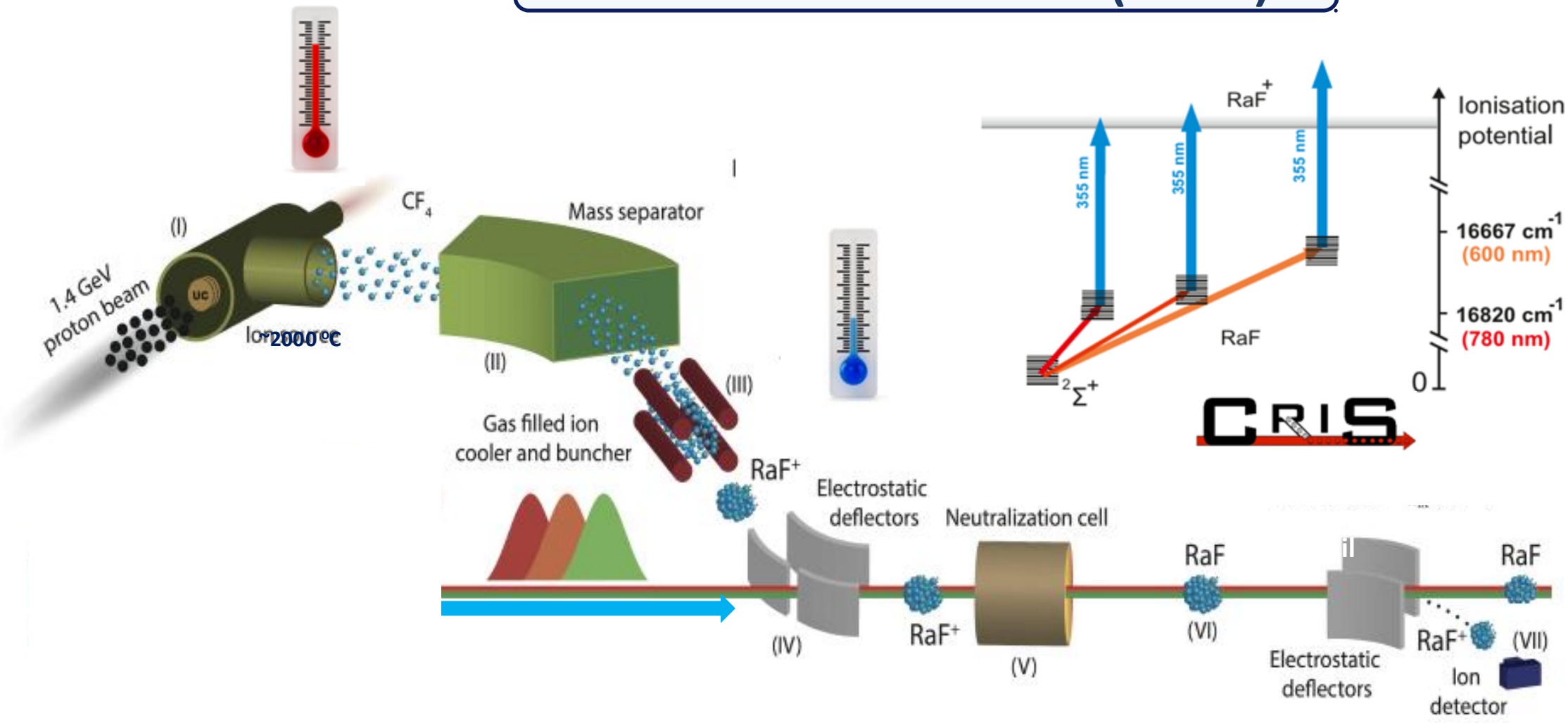
# Recent Results (RaF)




$$\mathrm{Ra}^+ \xrightarrow{\mathrm{CF_4}} \mathrm{RaF}^+ \xrightarrow{\mathrm{Na}} \mathrm{RaF}(^2\Sigma)$$

[Garcia Ruiz et al. Nature 581, 396 (2020)]

# Recent Results (RaF)

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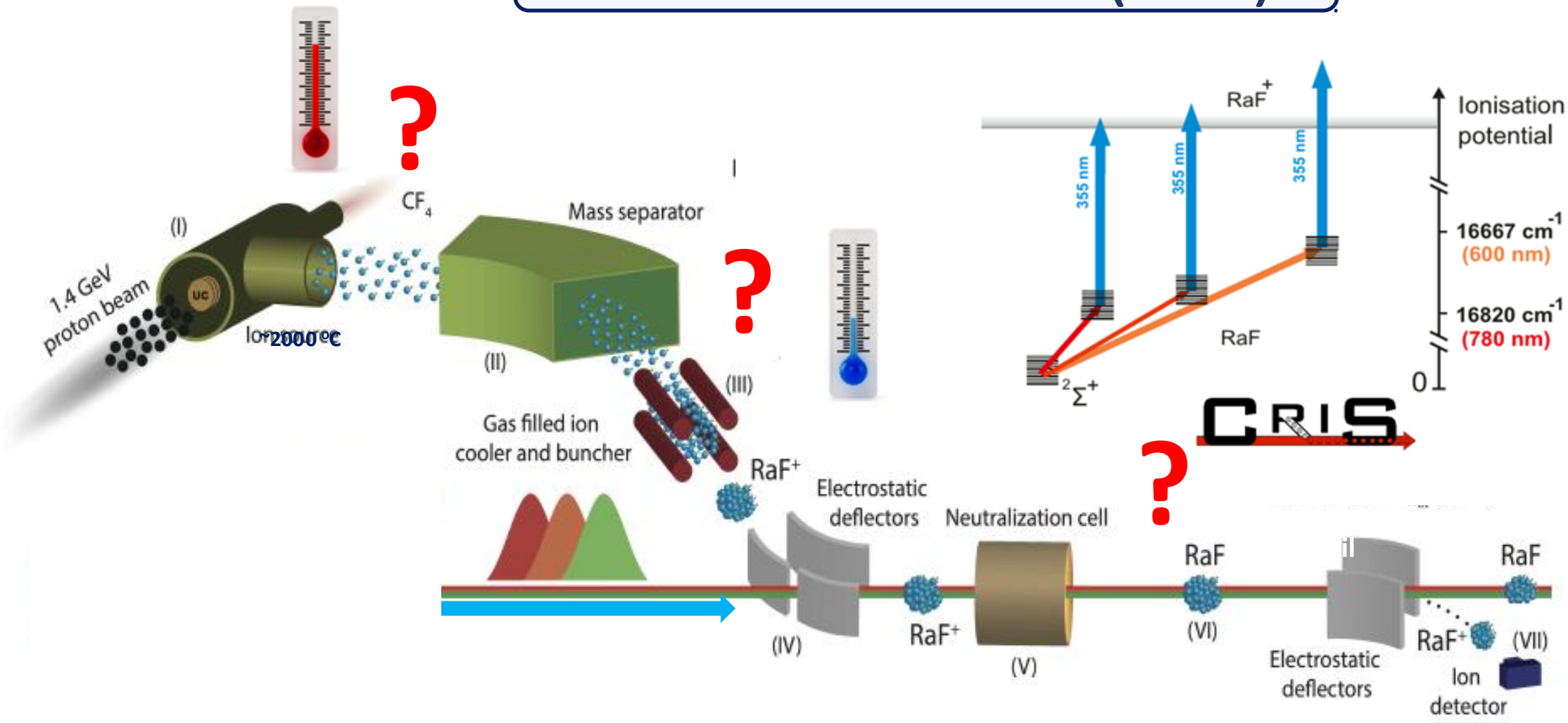
[Garcia Ruiz et al. Nature 581, 396 (2020)]

# Recent Results (RaF)




## A lot of unknowns!

**Total efficiency:** $\varepsilon = \varepsilon_{Ra}\,\varepsilon_{RaF}\,\varepsilon_{RFQ}\,\varepsilon_{neut}\,\varepsilon_{vib}\,\varepsilon_{rot}\,\varepsilon_{exc}\,\varepsilon_{ion}$

$(10^{-7} < \varepsilon < 1)$

Talk: Silviu Udrescu
Thursday 3-6 pm

# Recent Results (RaF)


nature

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Article | Open Access | Published: 27 May 2020

## Spectroscopy of short-lived radioactive molecules

R. F. Garcia Ruiz, R. Berger, [...]

*Nature* **581**, 396–400 (2020) | Cite this article

*Nature 581, 396 (2020)*

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### Spectroscopy of molecules with unstable nuclei

Pinning down the energy transitions of radium monofluoride, and eventually other short-lived molecules, could reveal the ways they are influenced by the properties of heavy radioactive nuclei.

Andrew Grant

CHEMISTRY WORLD

### Molecular experiments hope to reveal new physics

BY ANDY EXTANCE | 5 JUNE 2020

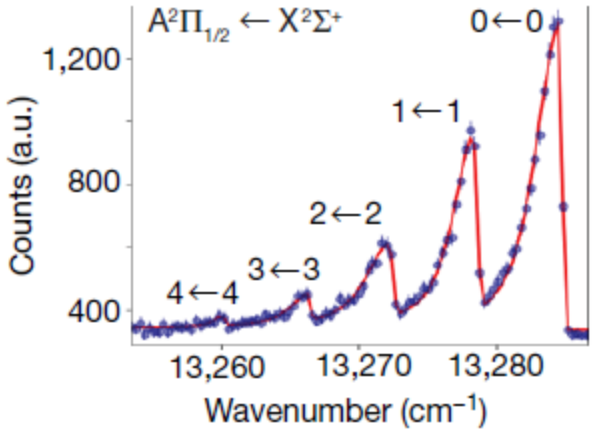
Detecting extremely short-lived radium fluoride can explore standard model's limits

physicsworld

ATOMIC AND MOLECULAR | RESEARCH UPDATE

Exotic radioactive molecules could reveal physics beyond the Standard Model

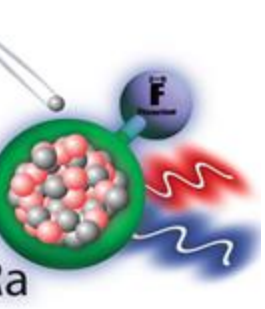
05 Jun 2020




$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-18}$ |
|---|---|---|---|---|---|---|

# Recent Results (RaF)

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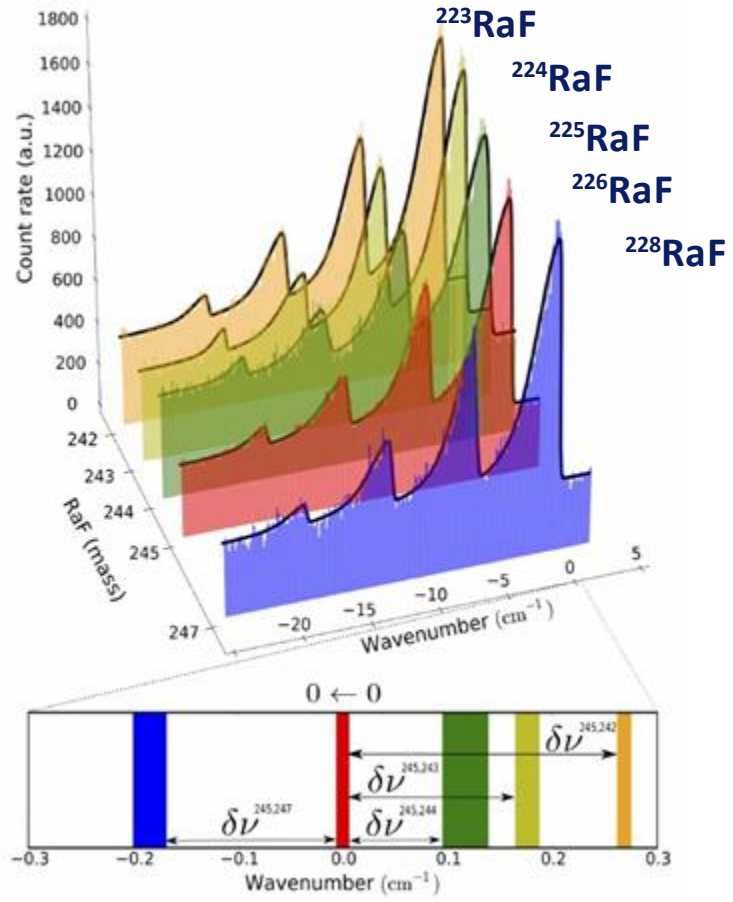
Isotope Shifts of Radium Monofluoride Molecules

S. M. Udrescu *et al.*

Phys. Rev. Lett. **127**, 033001 – Published 14 July 2021

Physics See Viewpoint: Sizing up Exotic Nuclei with Radioactive Molecules





*Nature 581, 396 (2020)*

*PRL 127, 033001 (2021)*

$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-18}$ |
|---|---|---|---|---|---|---|

# Recent Results (RaF)


nature physics

nature > nature physics > articles > article

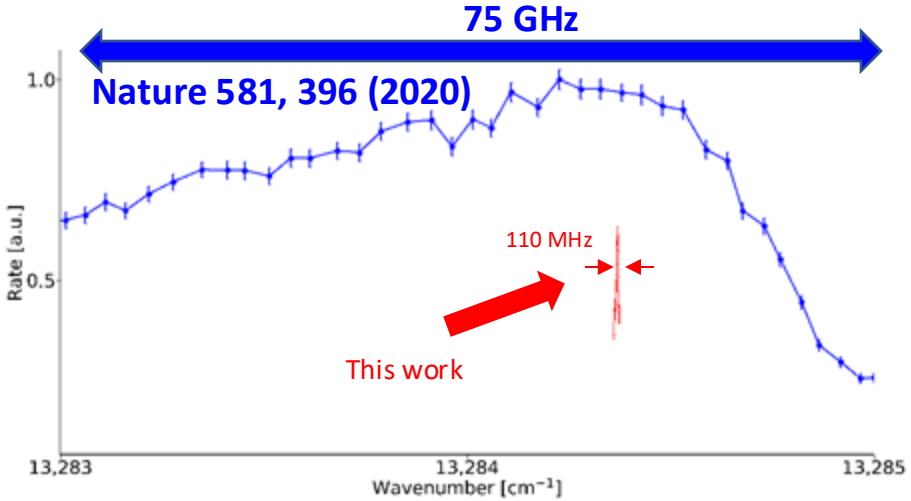
Article | Published: 09 January 2024

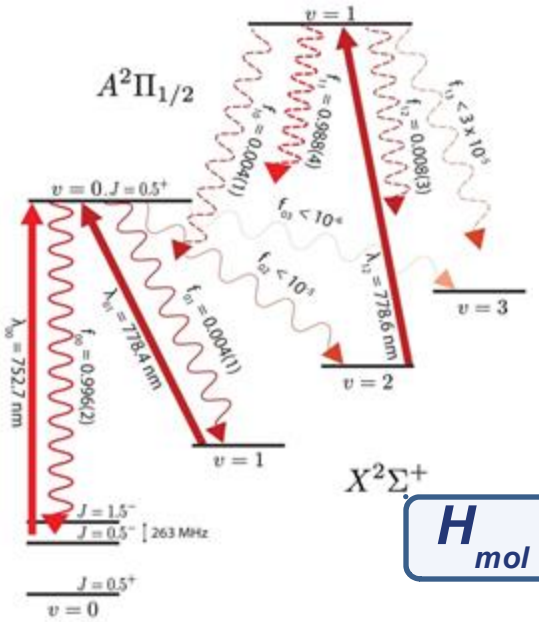
## Precision spectroscopy and laser-cooling scheme of a radium-containing molecule

S. M. Udrescu, S. G. Wilkins, A. A. Breier, M. Athanasakis-Kaklamanakis, R. F. Garcia Ruiz, M. Au, I.

+ Show authors

Nature Physics 20, 202–207 (2024) | Cite this article




Nature 581, 396 (2020)

PRL 127, 033001 (2021)

Nature Phys 20, 202 (2024)

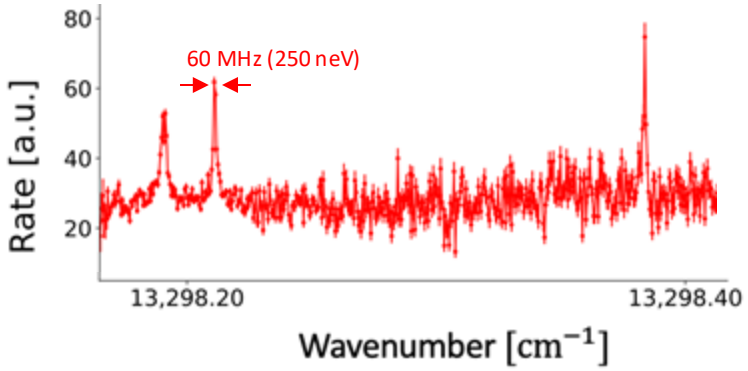
$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

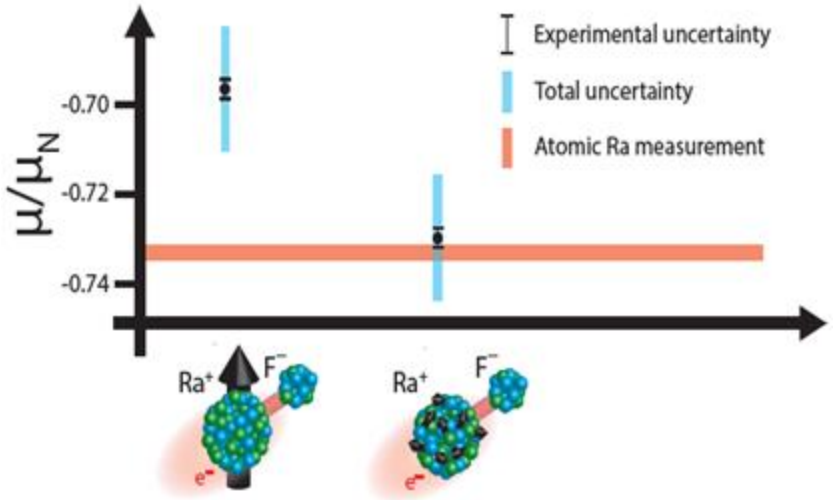
| | | | | | | |
|---|---|---|---|---|---|---|
| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-18}$ |

Talk: Silviu Udrescu
Thursday 3-6 pm

# Recent Results (RaF)

$^{225}$RaF hyperfine spectrum




$$\int B_e \, d\mu \neq \mu B_e$$


Nature 581, 396 (2020)
PRL 127, 033001 (2021)
Nature Phys 20, 202 (2024)
(2025) Science

$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 $10^{-2}$ $10^{-5}$ $10^{-8}$ $<10^{-12}$ $<10^{-18}$

Talk: Silviu Udrescu
Thursday 3-6 pm



[Wilkins, Udrescu et al. arxiv:2311.04121 (2024)]

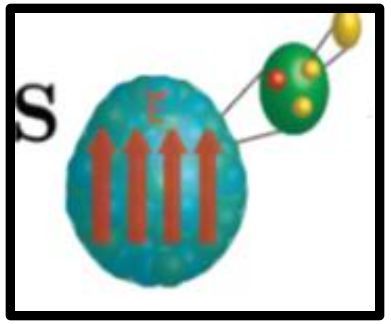
# Recent Results (RaF)

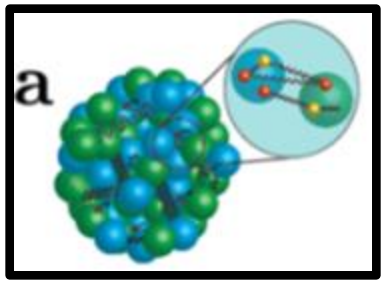

Nature 581, 396 (2020)
PRL 127, 033001 (2021)
Nature Phys 20, 202 (2024)
(2025) Science

$$H_{mol} = H_e + H_{vib} + H_{rot} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-18}$ |
|---|---|---|---|---|---|---|

$$\Delta O \sim \frac{1}{F_{mol}\tau\sqrt{N}}$$

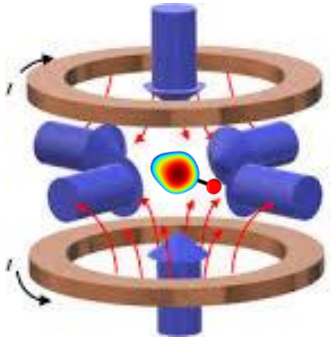


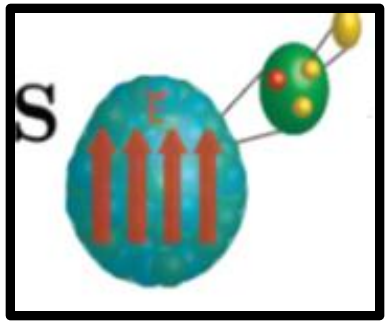




NEPTUNE Project

RaX Collaboration

# P,T-violation measurements with RaX Molecules






## RaX Collaboration

*J. Doyle (Harvard), N. Hutzler (Caltech), R.F. Garcia Ruiz (MIT) & FRIB*

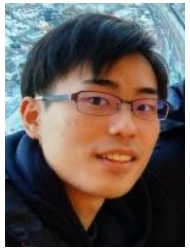
FRIB

Caltech


S. Ebadi


A. Jadbabaie


M. Fulghieri

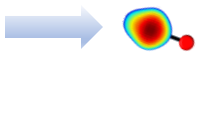

F. Shungo

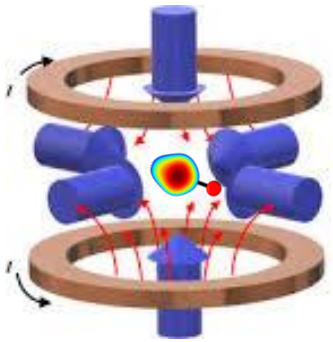

K. Khusainova

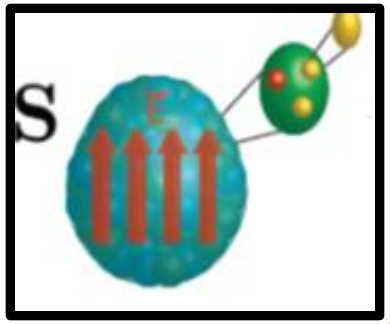

S. Munoz

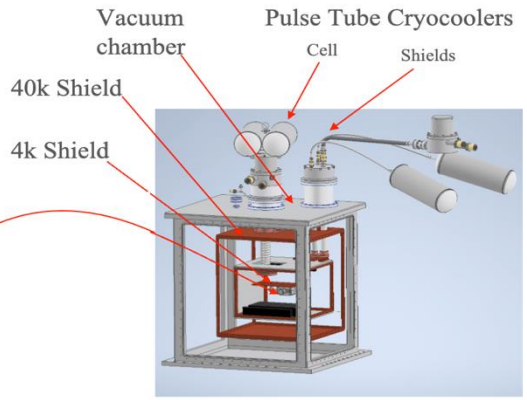
Talk: Nick Hutzler

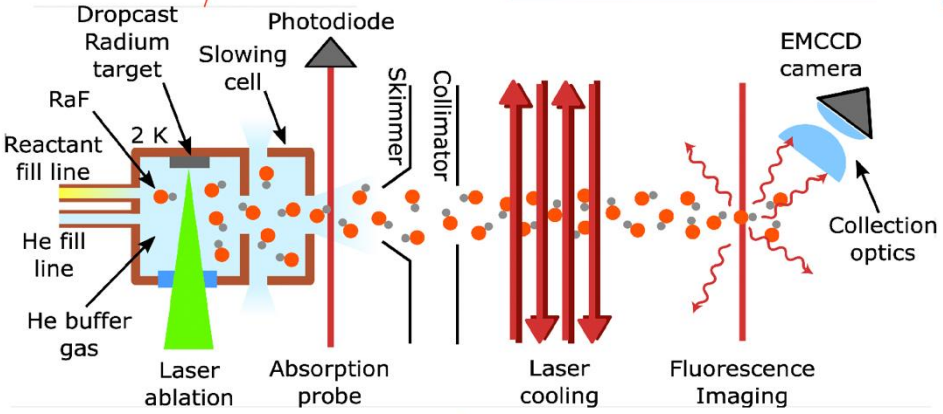
# P,T-violation measurements with RaX Molecules





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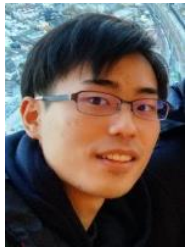
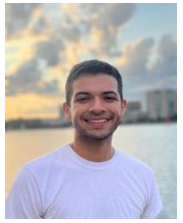

F. Shungo

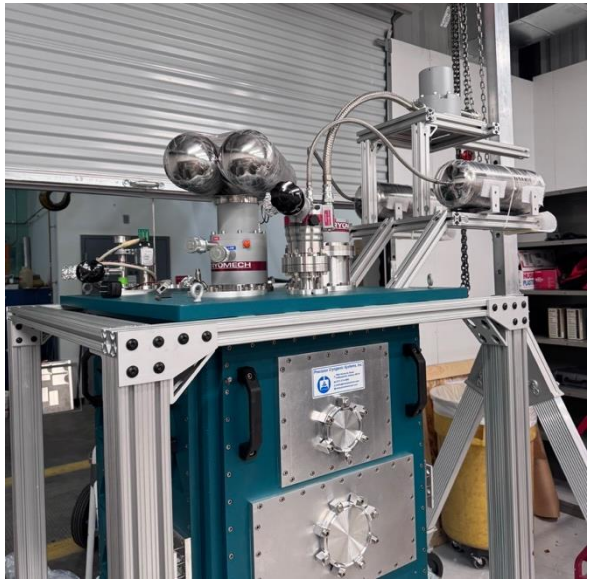

S. Ganguly


K. Khusainova

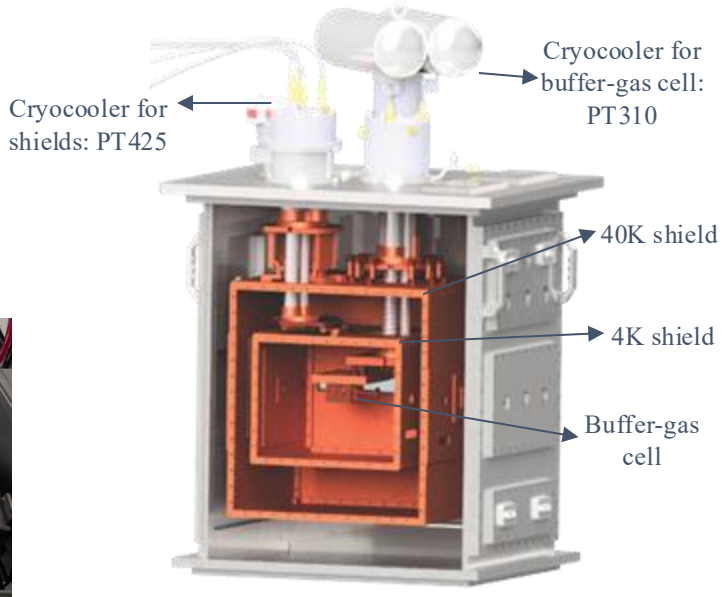

S. Munoz

# RaX Collaboration

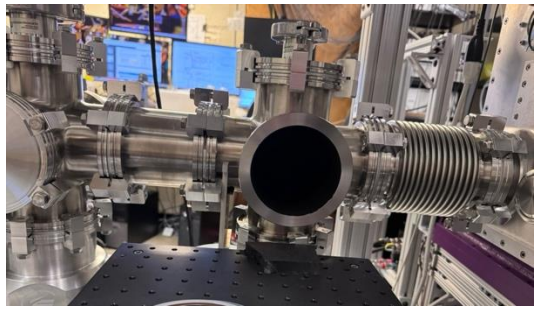
- ✓ Beam box fabrication
- ✓ Cryocooler testing
- ✓ Laser Installation
- ✓ Beamline construction



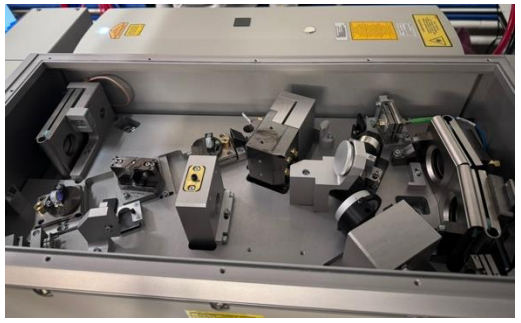
RaX @ Harvard




PT310 cryocooler:
2.2K base temp,
1W@3K, 51W@34K



Blackened beamline for fluorescence collection

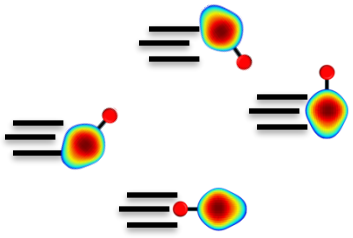


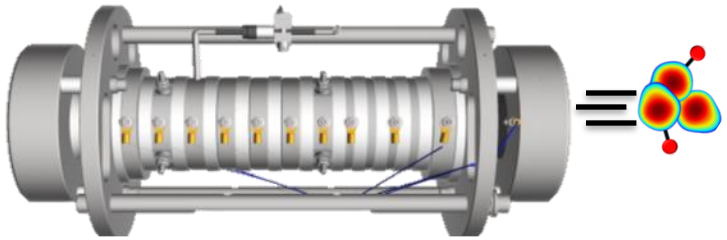
Ti:Sa laser installed

# Towards Efficient RaF Trapping Via $RaF^-$

T > 1000 K

$RaF^+$





**Gas filled ion Cooler and buncher**

T~300 K

$RaF^+$

(Charge exchange)

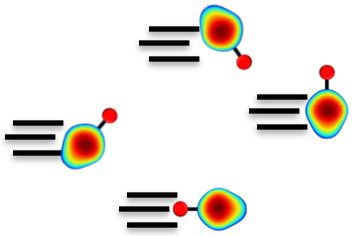
$RaF^+ + Na \Rightarrow Na^+ + RaF^+$

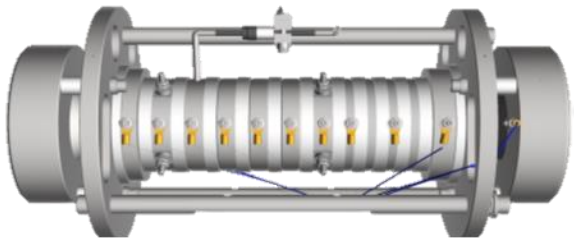
$RaF^+$

# Towards Efficient RaF Trapping Via $RaF^-$

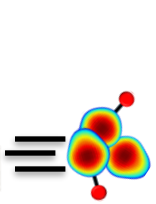
T > 1000 K

$RaF^+$





**Gas filled ion Cooler and buncher**



T~300 K

$RaF^+$

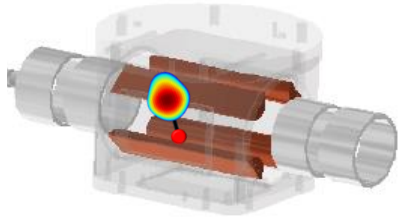
(Charge exchange)

$RaF^+ + 2Na \Rightarrow 2Na^+ + RaF^-$

$RaF^-$

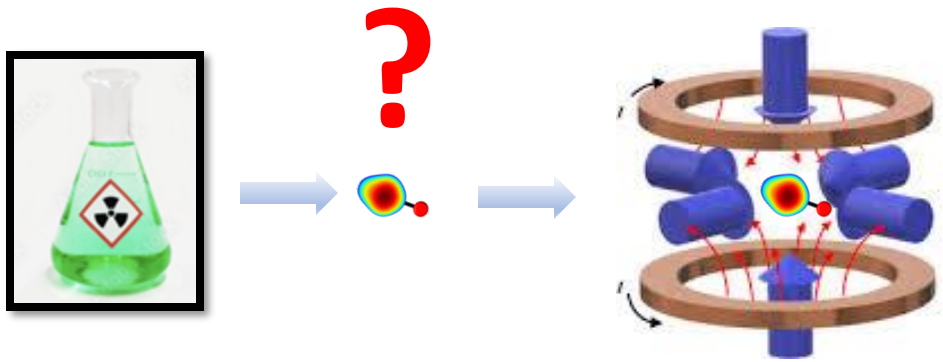
T < 4 K

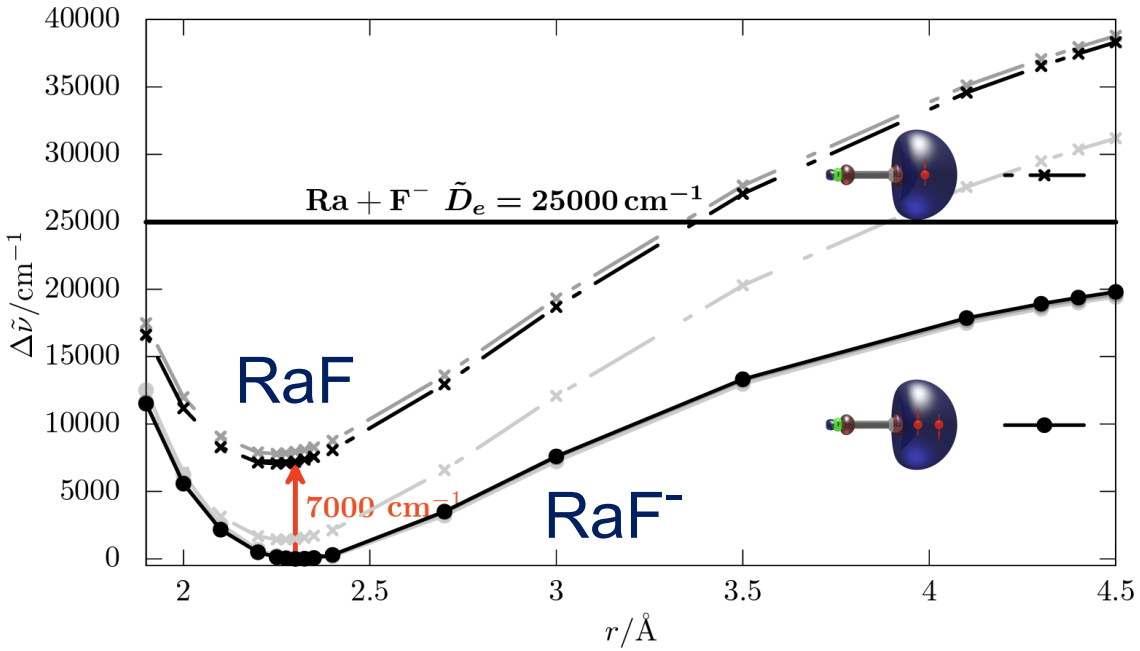
RaF



$RaF^- + \gamma \Rightarrow RaF + e$

# Towards Efficient RaF Trapping Via $RaF^-$

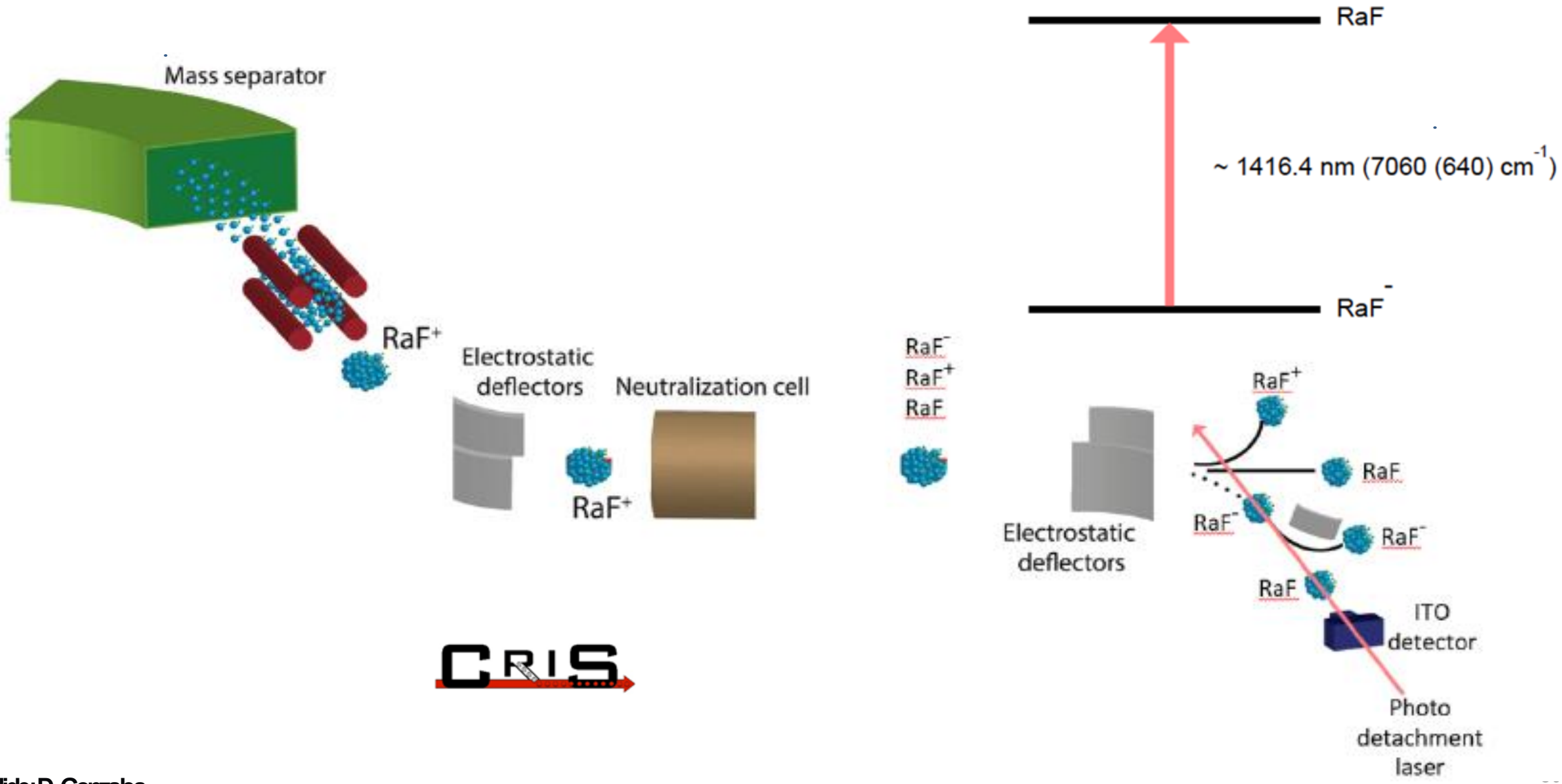



Gaul, Garcia Ruiz, Berger. arXiv:2403.09320 (2024)

Slide: D. Gonzales

# Towards Efficient RaF Trapping Via $RaF^-$




Slide: D. Gonzales

# Towards Efficient RaF Trapping Via $RaF^-$

PRELIMINARY

RaF

$RaF^-$

Table I. Dissociation wavenumbers $\tilde{D}_e$, adiabatic photoelectron detachment wavenumbers (ADW), equilibrium bond length $r_e$ and harmonic vibrational wavenumber $\tilde{\omega}_e$ of $MF^-$ computed at the level of RECP-CCSD(T)

| Molecule | $\tilde{D}_e/cm^{-1}$ | ADW/$cm^{-1}$ | $r_e$/Å | $\tilde{\omega}_e/cm^{-1}$ |
|---|---|---|---|---|
| $BeF^-$ | 28 700 | 8590 | 1.42 | 1060 |
| $MgF^-$ | 21 600 | 10 800 | 1.82 | 577 |
| $CaF^-$ | 26 200 | 8300 | 2.01 | 516 |
| $SrF^-$ | 25 000 | 7950 | 2.15 | 434 |
| $BaF^-$ | 26 800 | 6770 | 2.27 | 439 |
| $RaF^-$ | 23 600 | 7210 | 2.36 | 401 |

Gaul, Garcia Ruiz, Berger. arXiv:2403.09320 (2024)

Slide: D. Gonzales

# Radioactive Molecules for Fundamental-Symmetry Tests

→ **RaOH, $RaOH^+$ and $RaOCH_3^+$**

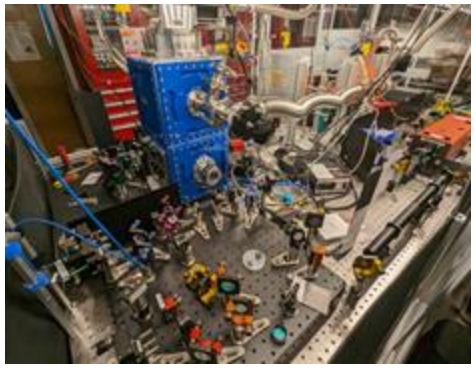
**[Hutzler, Jayich, …]**

Talk: Andrew Jayich

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

**[Yu & Hutzler Phys. Rev. Lett. 126, 023003 (2021)]**

**Hutzler's group: RaOH @ Caltech**

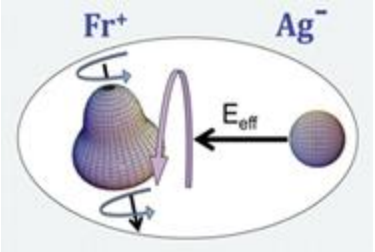


Talk: Nick Hutzler

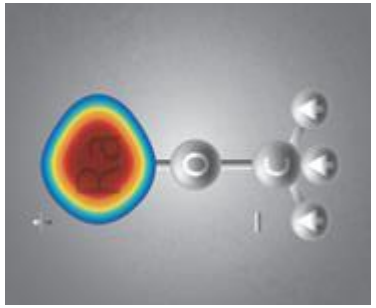
→ **Assembling molecules from cold atoms [Demille]**

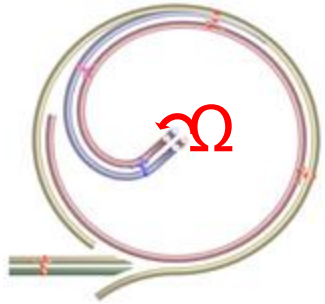
- **RaAg**
- **FrAg**

**[New J. Phys. 23, 113039 (2021)]**




→ **Centrifuge Deceleration**

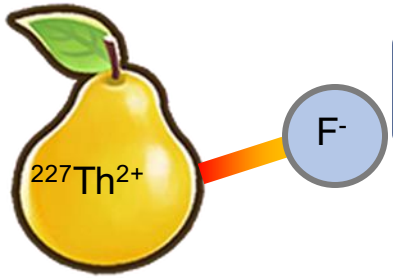





Xing Wu
$RaOCH_3$ @ FRIB/MSU

→ **$^{227}ThF^+$**

**[Boon Ng, Cornell, Fang, Malbrunot …]**

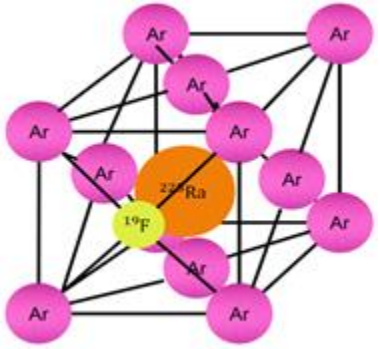



Talk: Kia Boon Ng

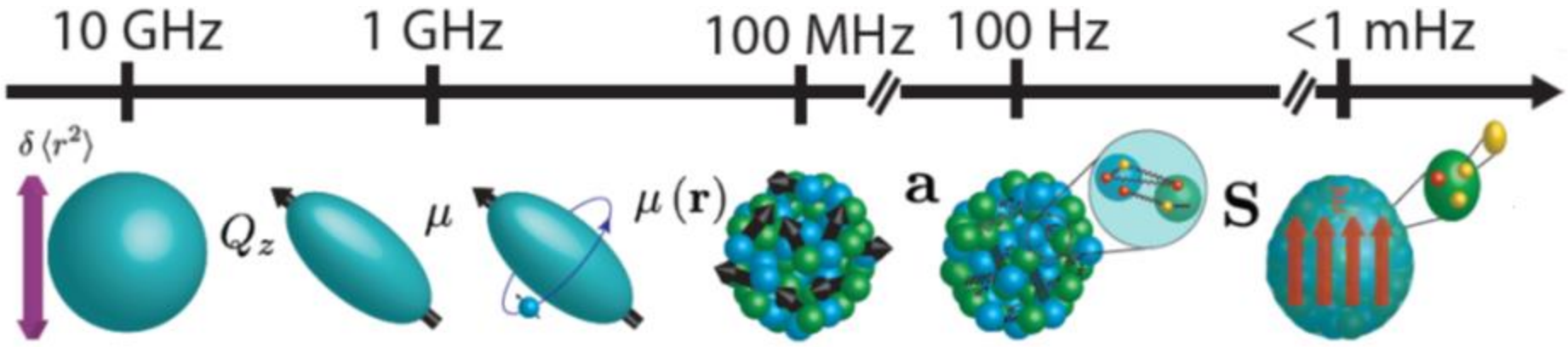
**[Singh, Vutha,…]**

**[Vutha et al. PRA 98:032513 (2018)]**

Talk: Jaideep Singh

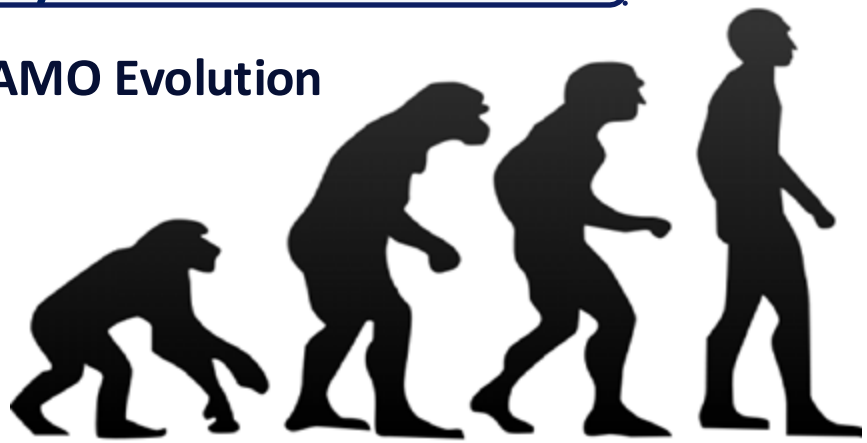


# Summary and Outlook

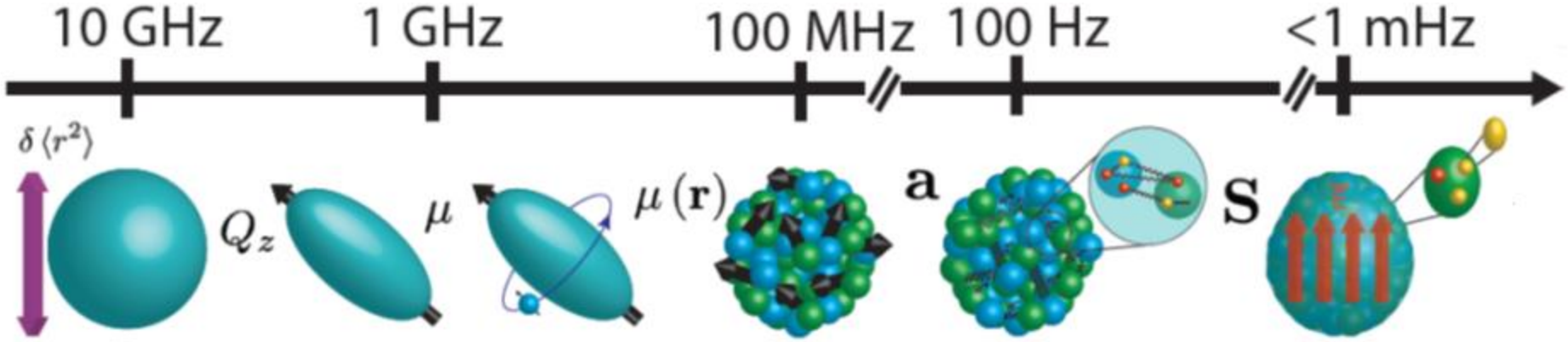



**Radioactive molecules offer a new window into our exploration of electroweak nuclear properties**

# Summary and Outlook




We are here!





$$H_{mol} = H_e + H_{vib} + H_{rot} + H_{sr} + \ldots + H_{hfs} + H_{PV} + H_{PTV}$$

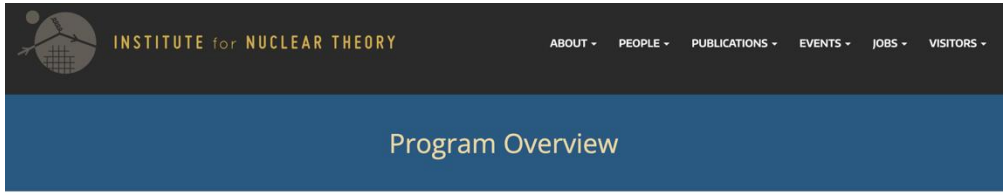
| eV | ~ 2 | $10^{-2}$ | $10^{-5}$ | $10^{-6}$ | $10^{-8}$ | $<10^{-12}$ | $<10^{-15}$ |
|---|---|---|---|---|---|---|---|

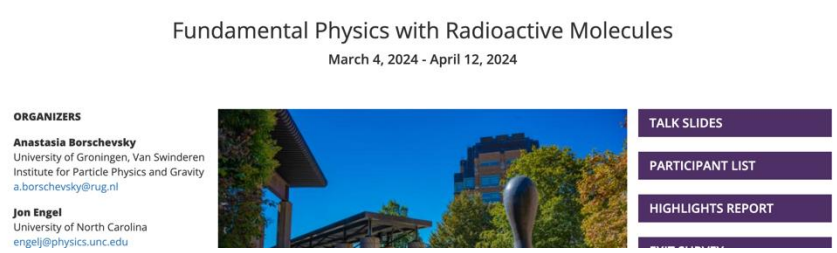
# Summary and Outlook

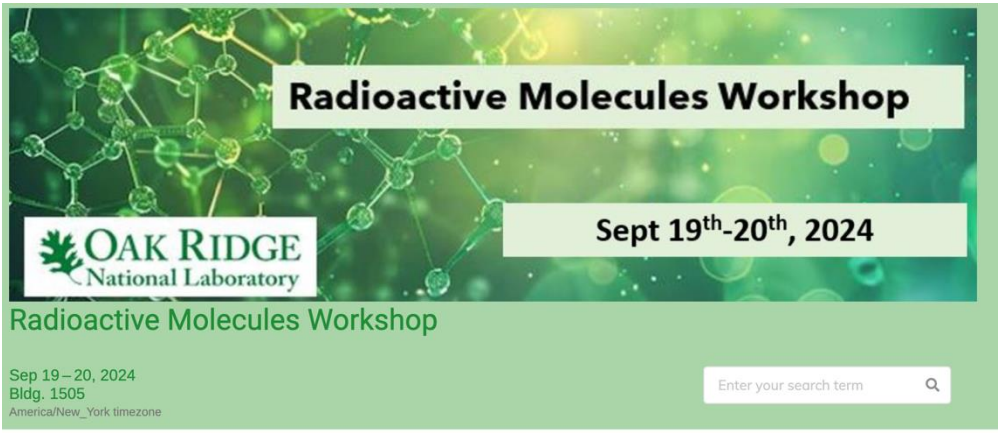
**New opportunities!**

- **New Facilities (FRIB, TRIUMF, ISOLDE, FAIR,…. )**
- **New Era of Precision (Atomic, Molecular, Nuclear) Physics**

# Radioactive Molecules: A growing community

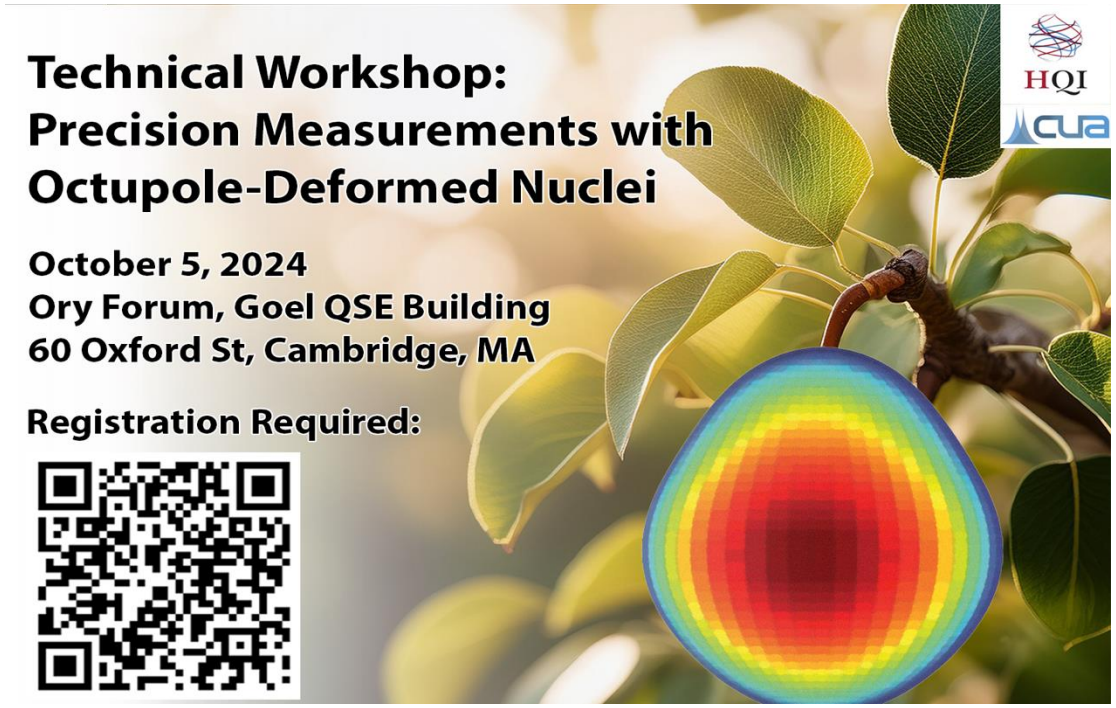




- "Opportunities for Fundamental Physics Research with Radioactive Molecules"
  Review Article: **Prog. Phys. 87 084301 (2024).**

- Invited reviews in preparation for
  **- Rev. Mod. Phys.,**
  **- Nature Physics Reviews**

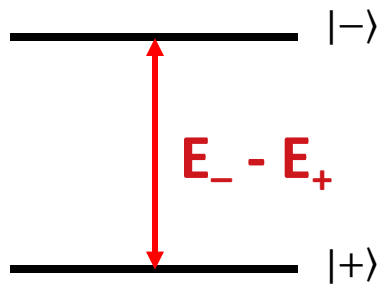


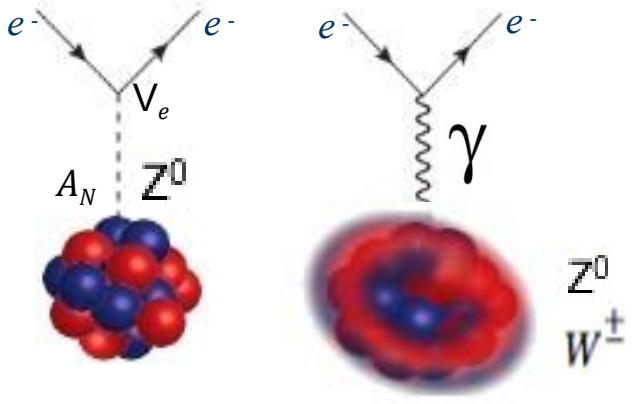
# NEPTUNE – Project

## Nuclear Electroweak Measurements in a Penning Trap Using Near-degenerate Energy States of Molecules

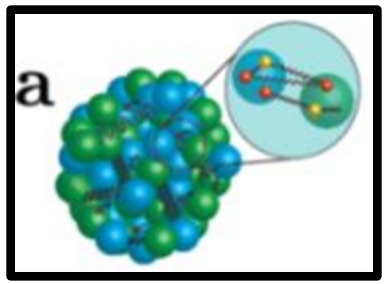
$H_{PV} \sim F(Z^c) / (E_+^{\,e} - E_-^{\,e})$

$$|s'\rangle = |s\rangle + \sum_{\gamma} \frac{\langle s| V_{PV} |p\rangle}{E_- - E_+} |p\rangle$$




$C_{2u}$, $C_{2d}$ Anapole



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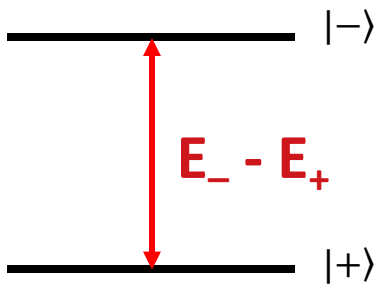


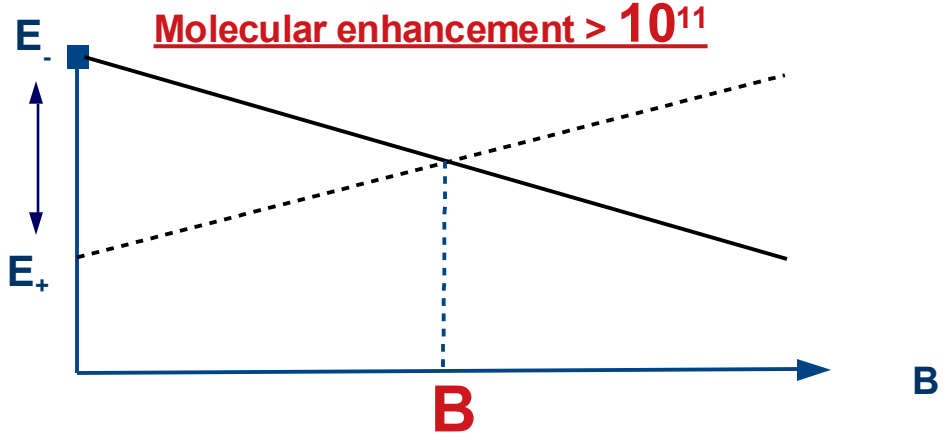


Molecular enhancement > $10^{11}$

**> $10^{11}$ Enhancement sensitivity to electroweak nuclear properties!**

Demille's Group [Altunas et al. Phys. Rev. Lett. 120, 142501 (2018)]

# NEPTUNE – Project

## Nuclear Electroweak Measurements in a Penning Trap Using Near-degenerate Energy States of Molecules

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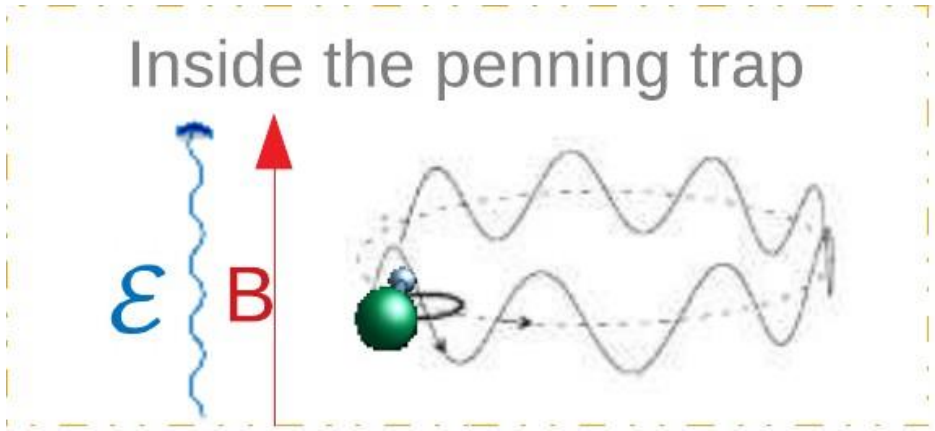



J. Karthein


S. Udrescu


S. Moroch


H. Kakiota

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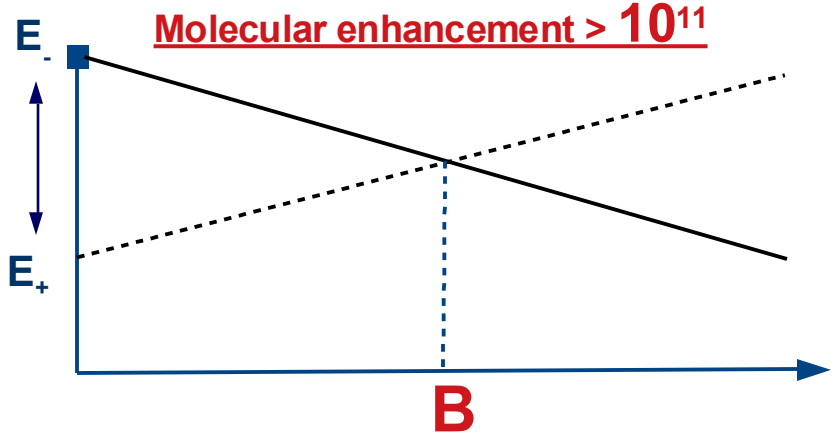
Electroweak Nuclear Properties from Single Molecular Ions in a Penning Trap

J. Karthein, S. M. Udrescu, S. B. Moroch, I. Belosevic, K. Blaum, A. Borschevsky, Y. Chamorro, D. DeMille, J. Dilling, R. F. Garcia Ruiz, N. R. Hutzler, L. F. Pašteka, and R. Ringle
Phys. Rev. Lett. **133**, 033003 – Published 19 July 2024



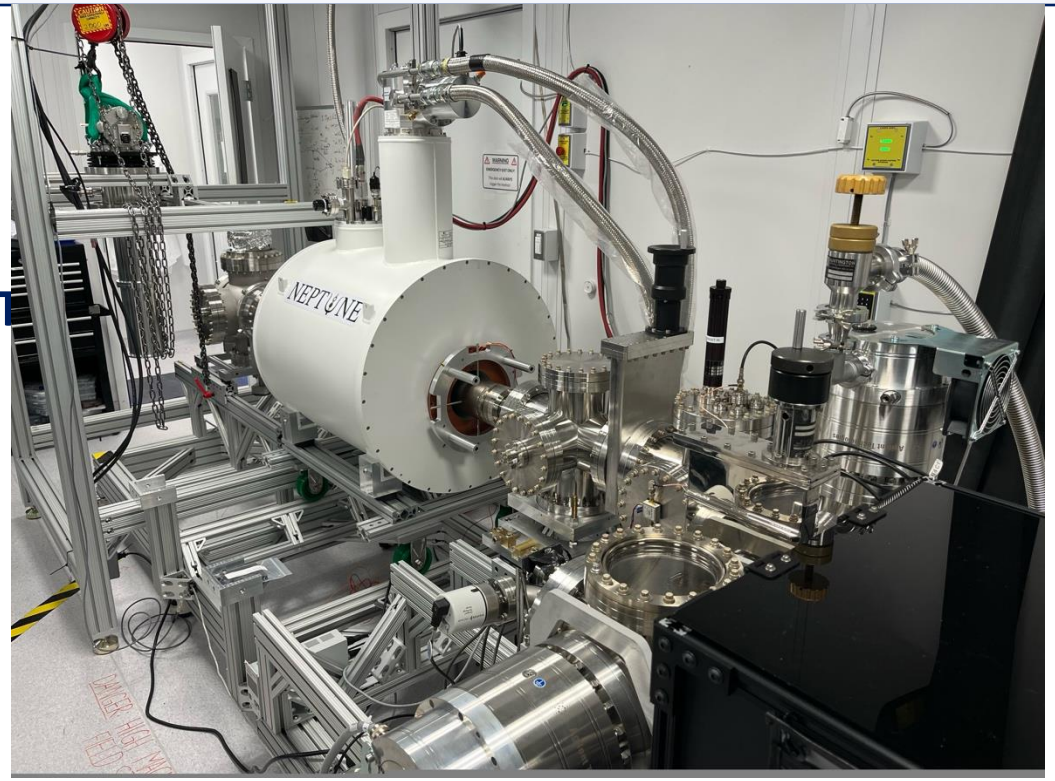

**Molecular enhancement > $10^{11}$**

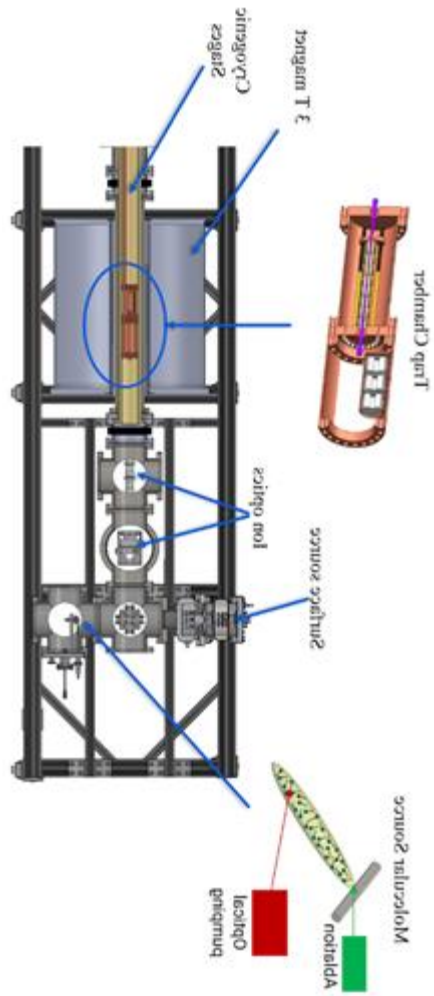
- Parity and Time reversal violation $> 10^3$
- **Nuclear amplification** **$> 10^3$**
- **Parity violation** **$> 10^{11}$**

# NEPTUNE – Project

## Nuclear Electroweak Measurements in a Penning Trap Using Near-degenerate Energy States of Molecules

**Lab @MIT**

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