Measuring EDMs with trapped molecular ions: HfF^+ and ThF^+

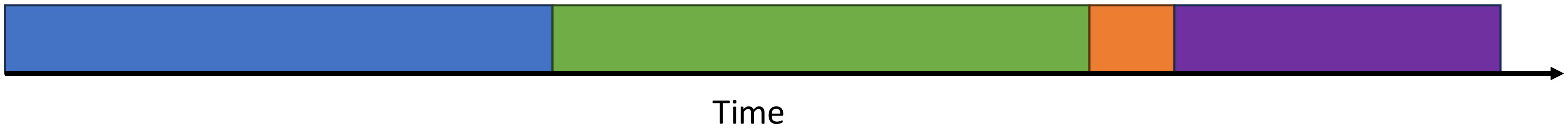
Kia Boon Ng, TRIUMF (present) and JILA (past)

Electric Dipole Moments: Experimental and Theoretical Horizons

12 May 2025

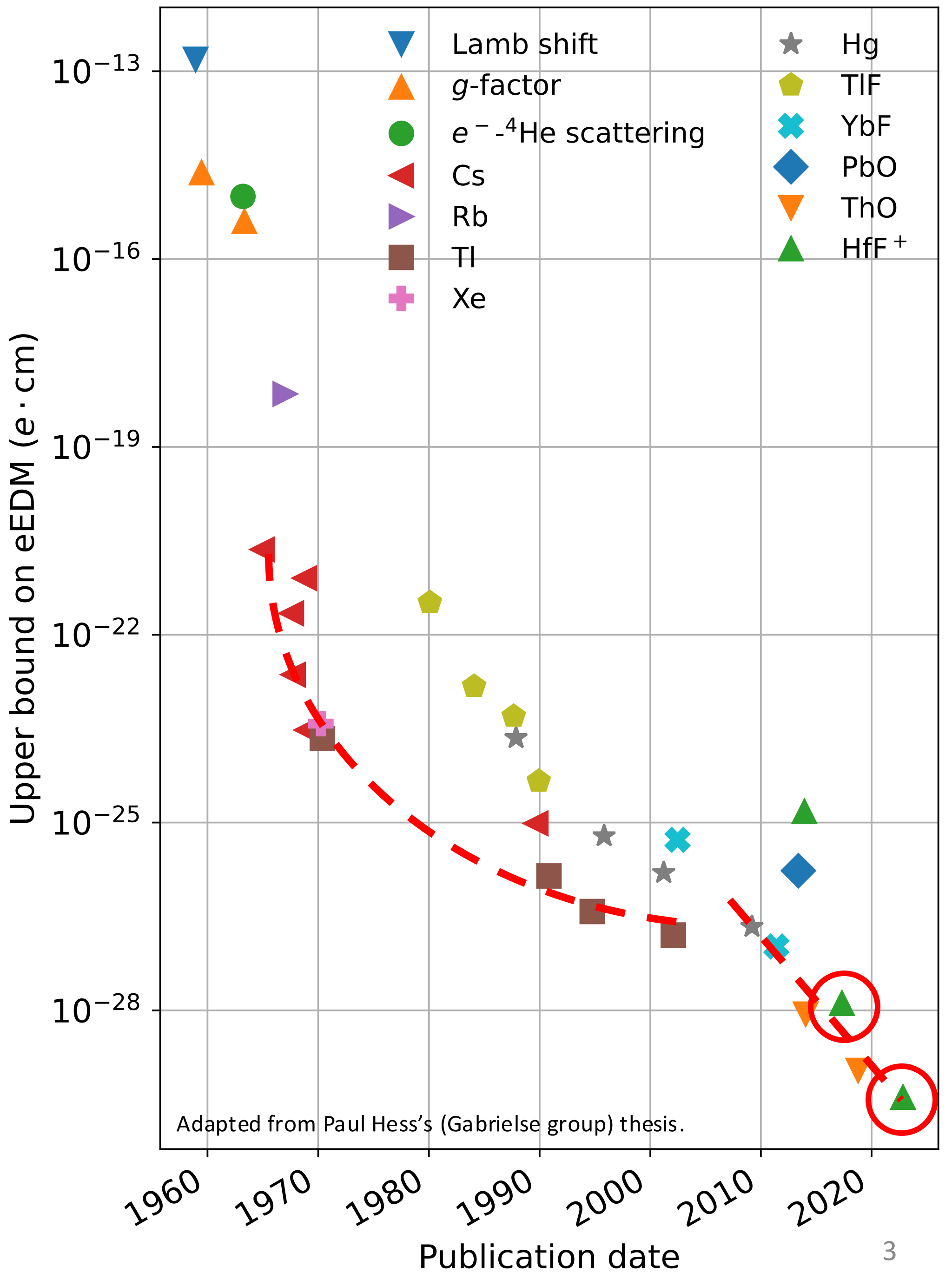
Overview

- HfF^+ (JILA)
  - Current best bound on eEDM.
  - Two-state detection.
  - Comparison with past results.
- ThF^+ (JILA)
  - Vs HfF^+.
  - Multiplexing with Bucket Brigade.
- $^{227}ThF^+$ (TRIUMF)
  - Nuclear Schiff moment.
  - Efficiency is key!
- EDM with molecular ions
  - Rotating electric field.
  - Non-inertial frame.



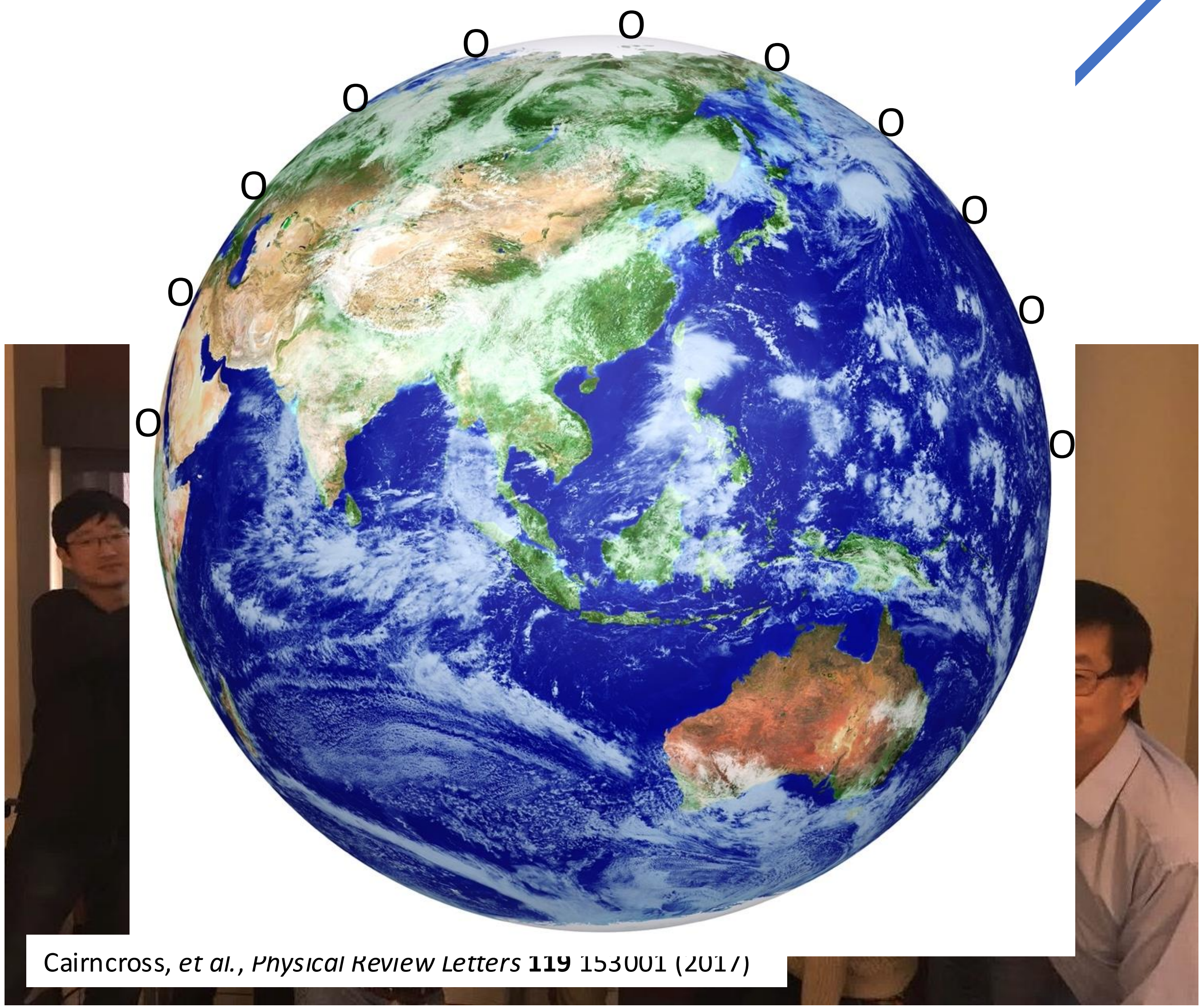
Reminder: eEDM

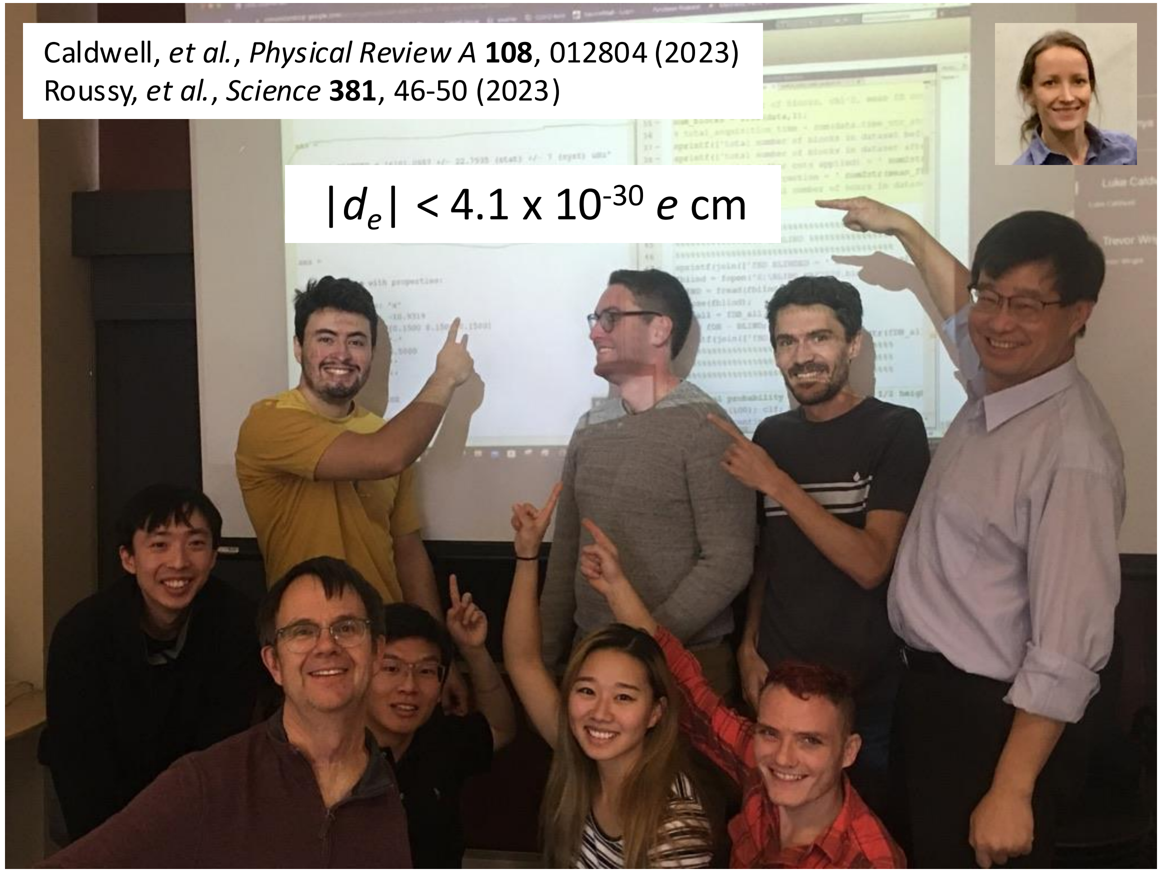
- Polar molecules
  - Strong internal electric field.
  - Easily polarizable.
- Standard Model background
  - ~ 10^{-35} e cm.



Hudson, *et al.*, *Physical Review Letters* **89** 023003 (2002)
Eckel, *et al.*, *Physical Review A* **87** 052130 (2013)
Pospelov and Ritz, *Physical Review D* **89** 056006 (2014)
Ema, *et al.*, *Physical Review Letters* **129** 231801 (2022)

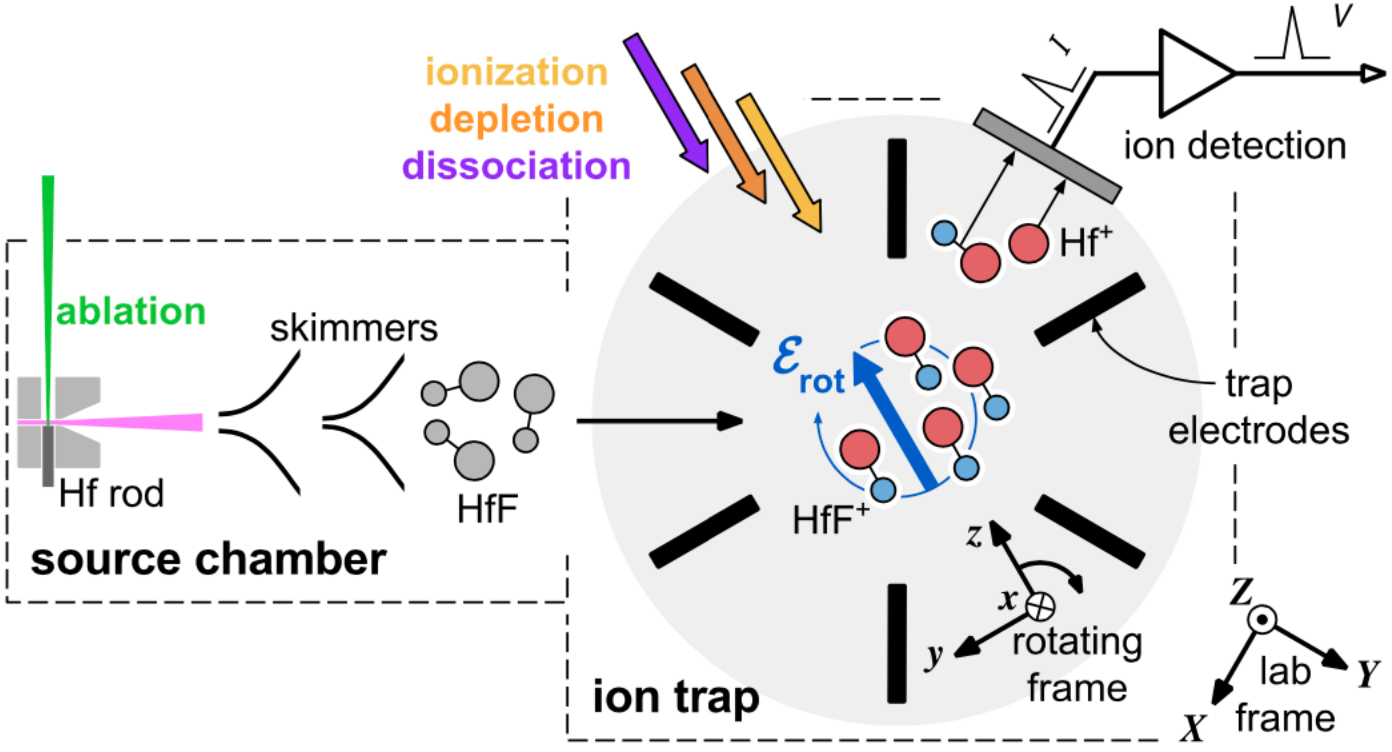
JILA eEDM with HfF^+ (Gen. I 2017, Gen. II 2022)





Equivalent to measuring the Earth's roundness with a precision of a single layer of atoms on one of its hemispheres!

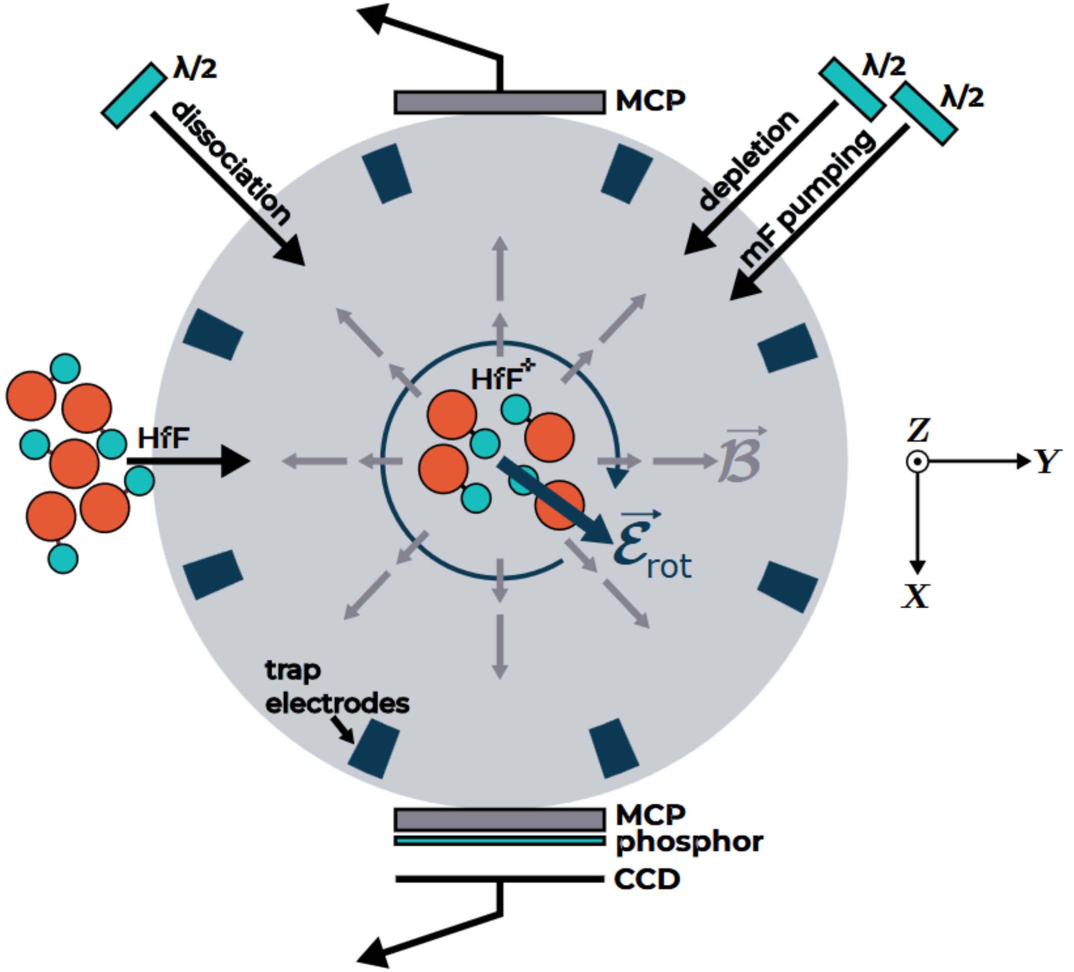
JILA eEDM Gen. I



Cairncross, *et al.*, *Physical Review Letters* **119** 153001 (2017)

Upgrade to Gen. II

- Better trap design
  - Field homogeneity.
  - Larger trapping volume.
- Rotational cooling
  - More useful ions.
- Two-state detection
  - Suppression of systematics.
  - Faster ion number statistics.

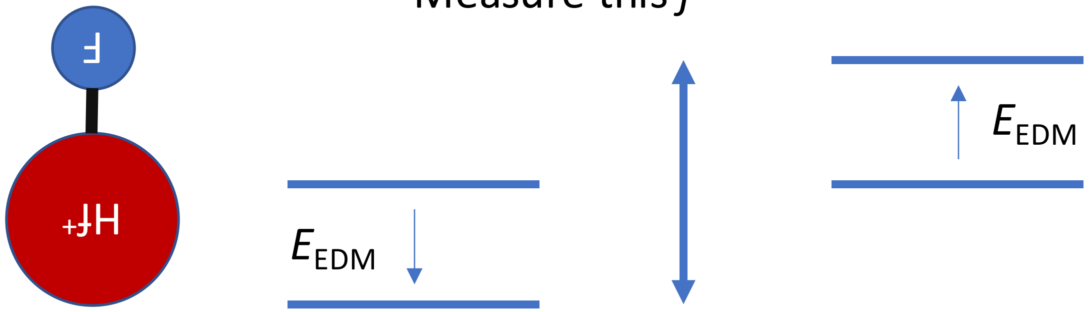


Roussy, PhD thesis (2022)

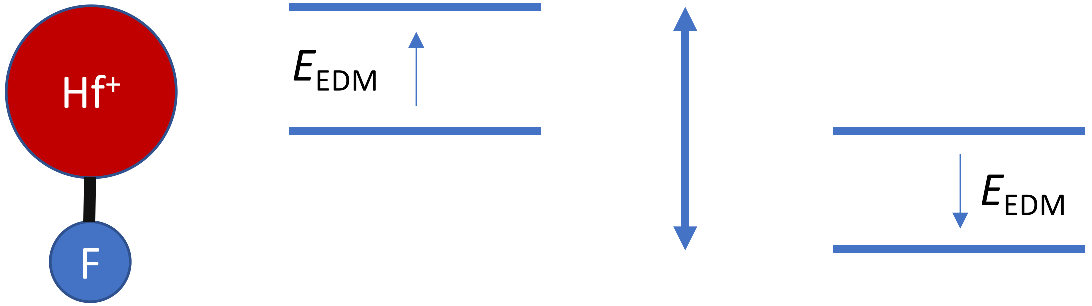
Two-state detection: the idea

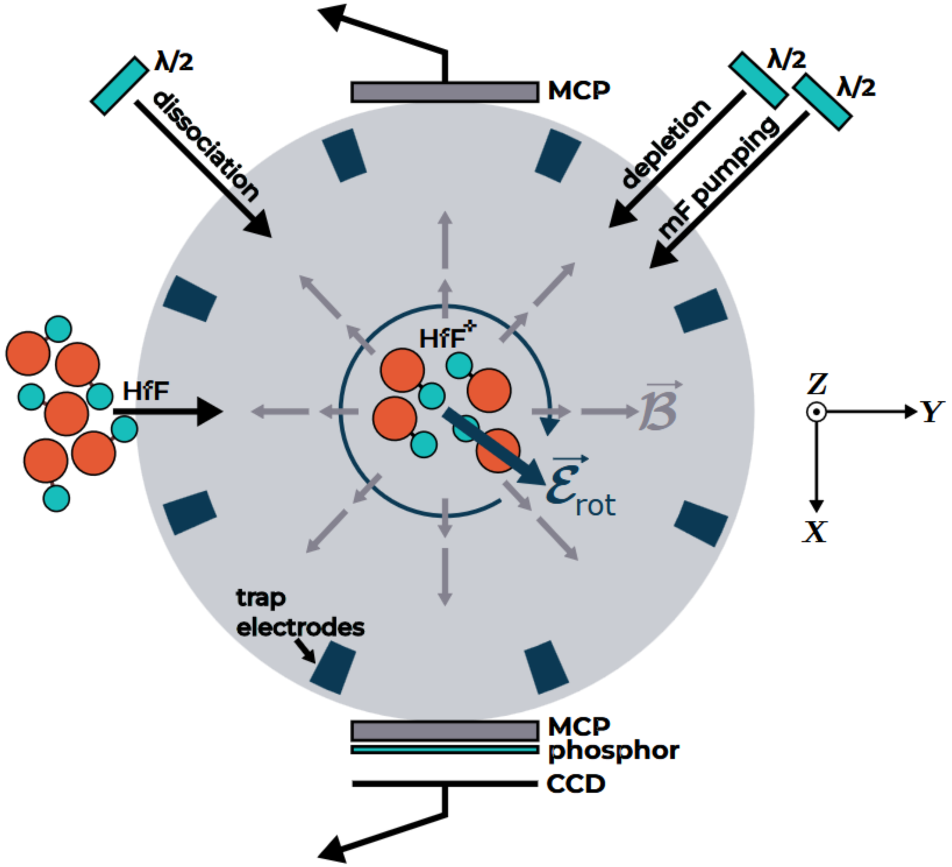
Two-state detection: the reason

Measure this *f*



Measure this *f*, *simultaneously!*

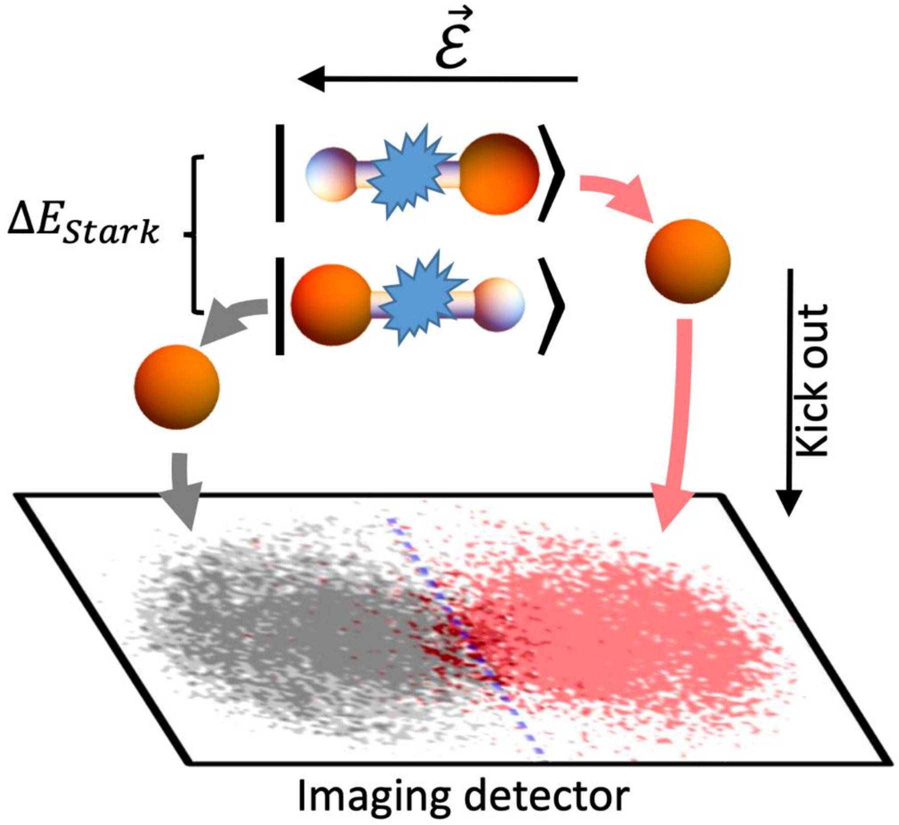




Two-state detection: the paper

- Dissociate molecules.
- Kick ions towards ion detector.
- Draw a calibrated line.
- Associate ions on one side with one state.
- Collaboration with Tanya Zelevinsky (Columbia University).

- Developed for HfF^+ and ThF^+.
- Recent TRIUMF-JILA collaboration experiment increased dissociation efficiency in ThF^+ by x10!



Zhou, *et al.*, *Physical Review Letter* **124** 053201 (2020)

Ng, *et al.*, submitted to PRA (2025)

JILA EDM Gen. II

RESEARCH ARTICLE

PHYSICS Roussy *et al.*, *Science* **381**, 46–50 (2023) 7 July 2023

An improved bound on the electron's electric dipole moment

Tanya S. Roussy[1,2]†, Luke Caldwell[1,2]†, Trevor Wright[1,2], William B. Cairncross[1,2]‡, Yuval Shagam[1,2]§, Kia Boon Ng[1,2], Noah Schlossberger[1,2], Sun Yool Park[1,2], Anzhou Wang[1,2], Jun Ye[1,2], Eric A. Cornell[1,2]*

Systematic and statistical uncertainty evaluation of the HfF^+ electron electric dipole moment experiment

Luke Caldwell,[1,2,*] Tanya S. Roussy,[1,2,*] Trevor Wright,[1,2] William B. Cairncross,[1,2,†] Yuval Shagam,[1,2,‡] Kia Boon Ng,[1,2] Noah Schlossberger,[1,2] Sun Yool Park,[1,2] Anzhou Wang,[1,2] Jun Ye,[1,2] and Eric A. Cornell[1,2]
[1]*JILA, NIST, and University of Colorado, Boulder, Colorado 80309, USA*
[2]*Department of Physics, University of Colorado, Boulder, Colorado 80309, USA*

(Received 2 December 2022; revised 21 February 2023; accepted 10 May 2023; published 10 July 2023)

We have completed an improved precision measurement of the electron's electric dipole moment using trapped HfF^+ in rotating bias fields. We report on the accuracy evaluation of this measurement, describing the mechanisms behind our systematic shifts. Our systematic uncertainty is reduced by a factor of 30 compared to the first generation of this measurement [Cairncross *et al.* Phys. Rev. Lett. **119**, 153001 (2017)]. Our combined statistical and systematic accuracy is improved by a factor of 2 relative to any previous measurement [Nature (London) **562**, 355 (2018)].

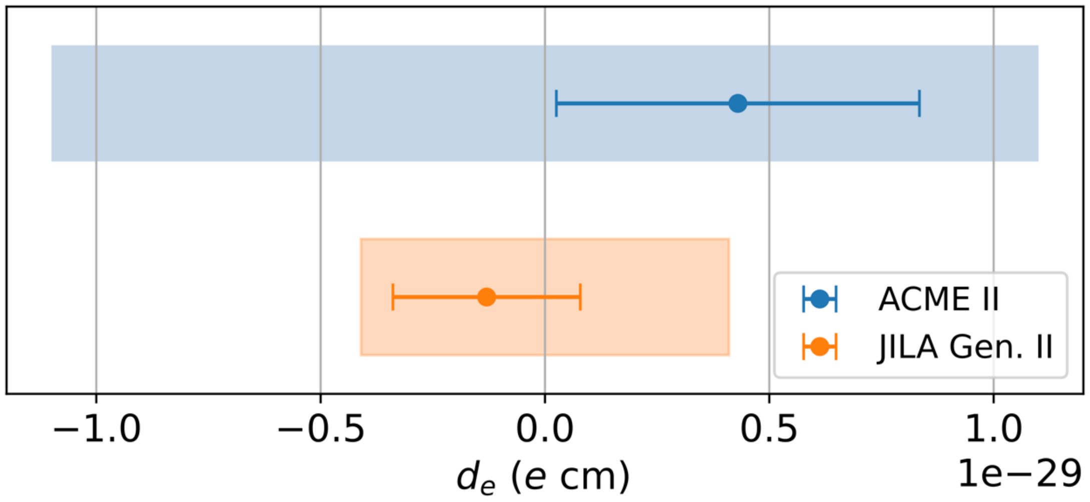
DOI: 10.1103/PhysRevA.108.012804

Experiment	Interrogation time	1σ statistical	1σ systematic	1σ total	90% confidence
JILA Gen. I (2017)	314 hours	77×10^{-30} e cm	1.7×10^{-30} e cm	79×10^{-30} e cm	130×10^{-30} e cm
ACME Gen. II (2018)	350 hours	3.1×10^{-30} e cm	2.6×10^{-30} e cm	4.0×10^{-30} e cm	11×10^{-30} e cm
JILA Gen. II (Nov, 2022)	550 hours	2.0×10^{-30} e cm	0.6×10^{-30} e cm	2.1×10^{-30} e cm	4.1×10^{-30} e cm

JILA EDM Gen. II

Official announcement paper:
Roussy, *et al.*, *Science* **381**, 46-50 (2023)

Systematics analyses paper:
Caldwell, *et al.*, *Physical Review A* **108**, 012804 (2023)



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  - Rotating electric field.
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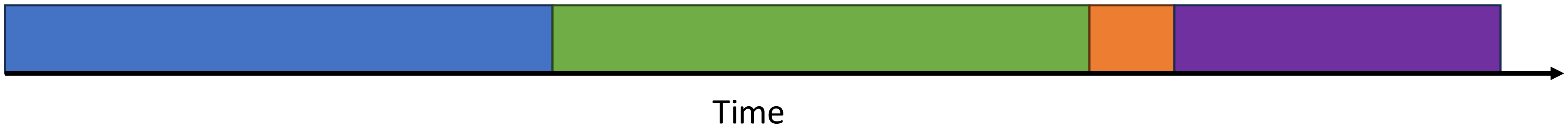
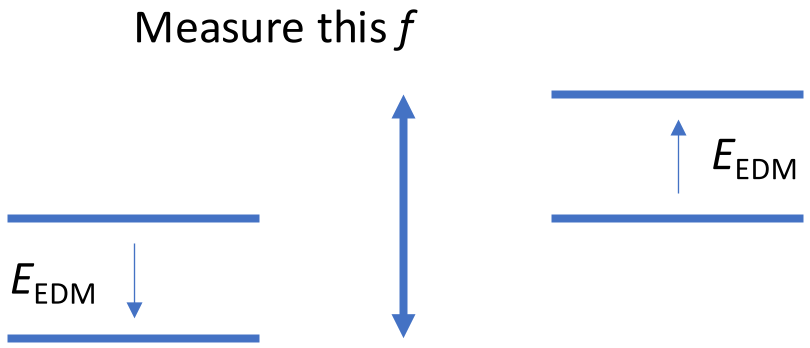


Statistical sensitivity



Ramsey spectroscopy
(a spin precession experiment)

$$\delta f \sim \frac{1}{\tau \sqrt{N}}$$

$$\delta d_e \sim \frac{1}{\underbrace{\mathcal{E}_{\mathrm{eff}}\, \tau}_{\text{Molecule specific}} \sqrt{N}}$$

Why ThF^+?

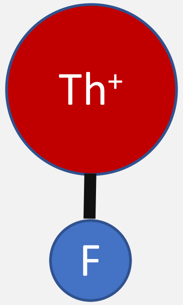
	ThF^+	HfF^+
Effective *E*-field	35.2 GV/cm	23.4 GV/cm
eEDM-sensitive state	(Ground state) 0 cm^{-1}	(Excited state) 977 cm^{-1}
Expected Coherence	~10 s	2 s

This restricts this

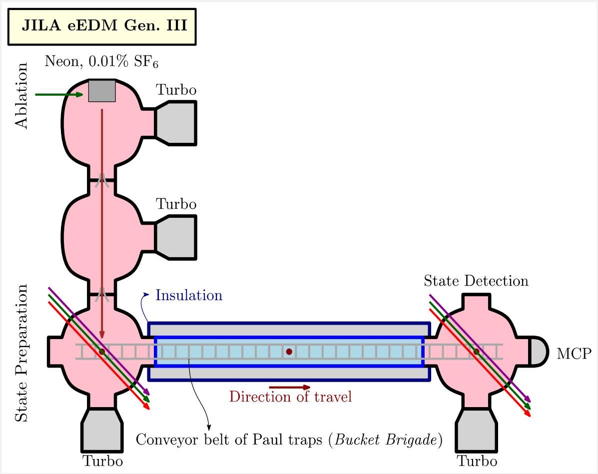
Denis, *et al.*, *New Journal of Physics* **17** 043005 (2015)
Skripnikov, *et al.*, *Physical Review A* **91** 042504 (2015)
Petrov, *et al.*, *Physical Review A* **76** 030501 (2007)
Gresh *et al.*, *Journal of Molecular Spectroscopy* **319**, 1-9 (2016).
Cossel *et al.*, *Chemical Physics Letters* **546**, 1-11 (2012)

JILA Gen. III with ThF^+

- How ThF^+? Two things remain:

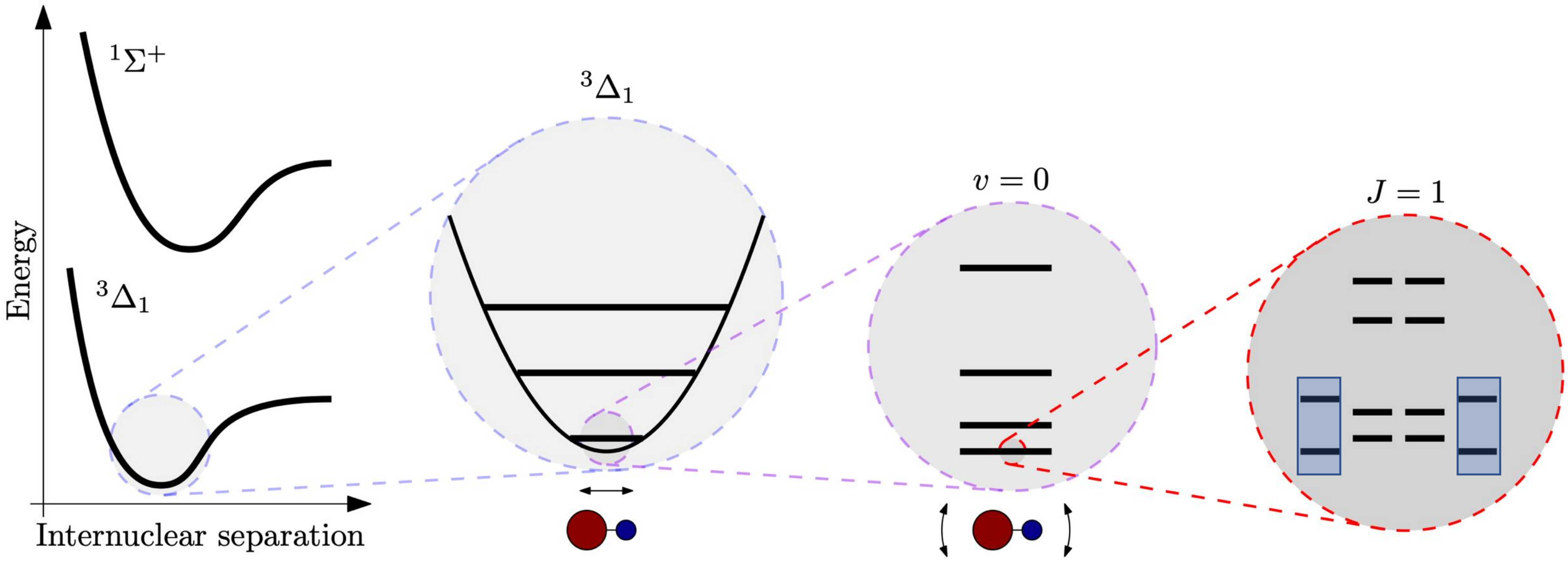


Behavior:
- What are the quantum numbers?
- What are the transition frequencies?



Implementation:
- How do we maximize the potential of the molecule?
- What experimental protocol?

How ThF^+ (behavior)?



Electronic
$\gtrsim 1000\ \mathrm{cm}^{-1}$

Vibrational
$\sim 600\ \mathrm{cm}^{-1}$

Rotational
$\gtrsim 1\ \mathrm{cm}^{-1} \sim 30$ GHz

Hyperfine, parity, and m_F
$\sim$ MHz

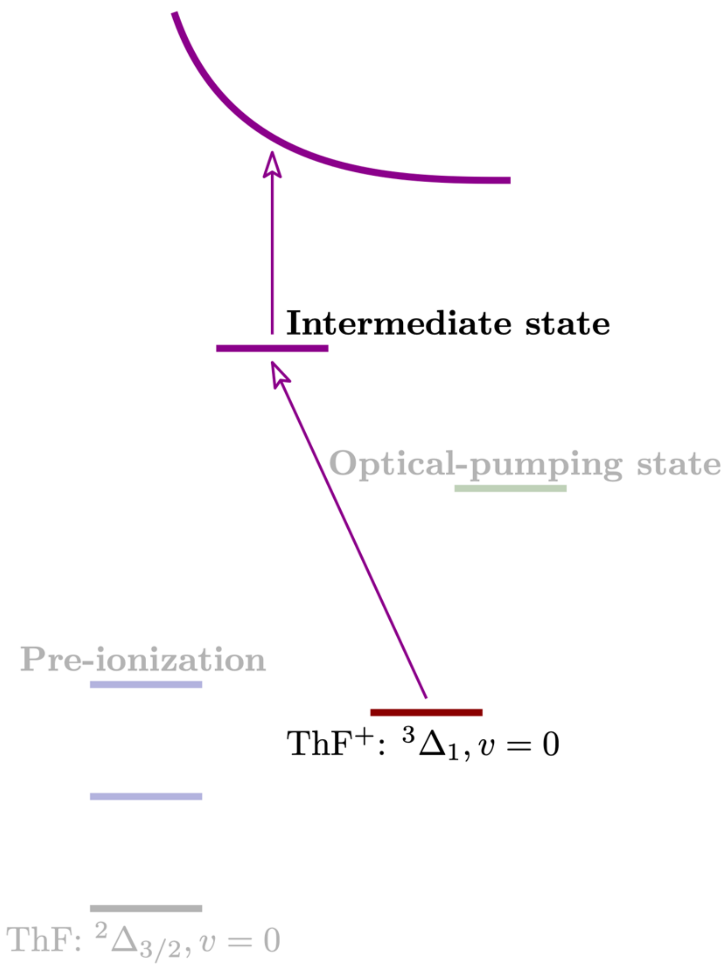
How ThF^+ (behavior)?

1. Create ThF^+ in X $^3\Delta_1(v=0)$
   - State-selective REMPI

2. Optical + μwave pump to the *science state:* X $^3\Delta_1(v=0, J=1, F=3/2, m_F=\pm3/2)$

3. EDM Ramsey spectroscopy

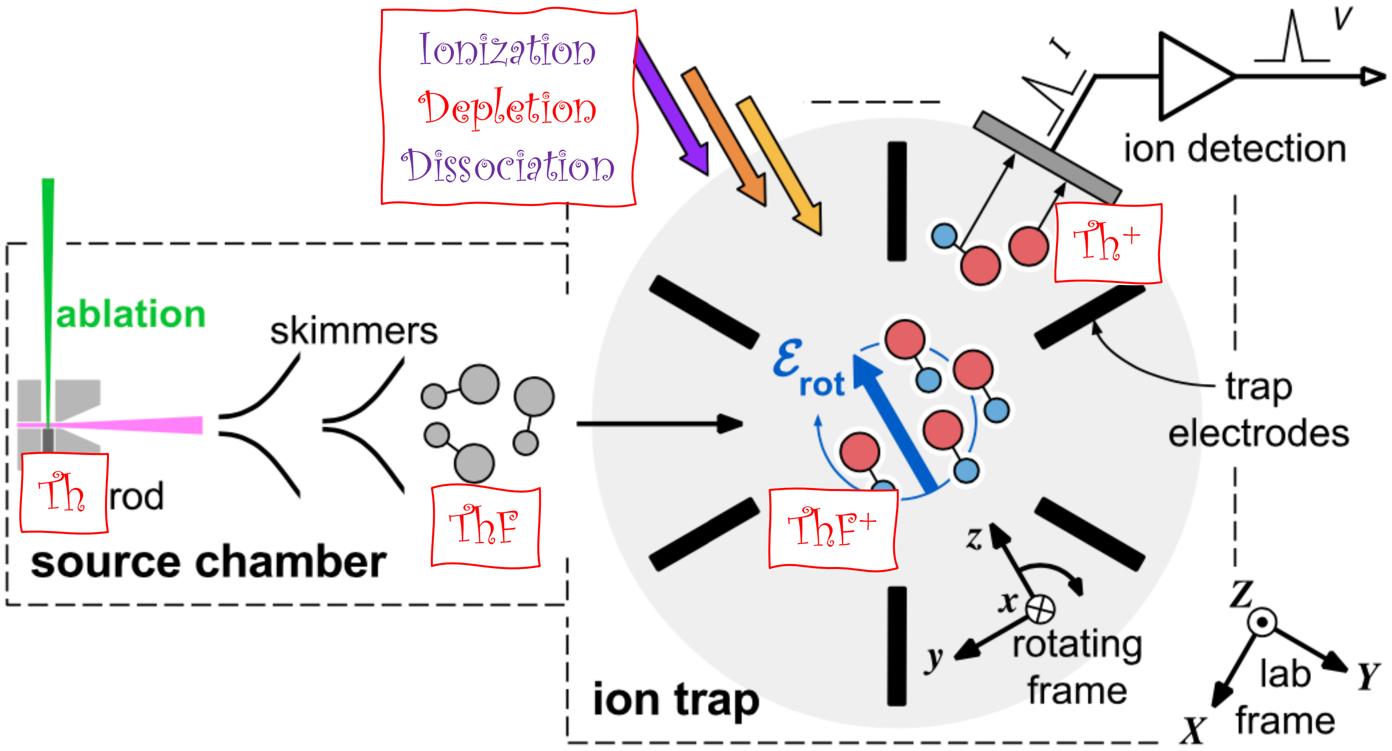
4. State readout
   - State-selective REMPD



Zhou, *et al.*, *Journal of Molecular Spectroscopy*, **358** (2019) 1-16.
Gresh, *et al.*, *Journal of Molecular Spectroscopy*, **319** (2016), 1-9.
Barker, *et al.*, *Journal of Chemical Physics*, **136** (2012) 104305.
Ng, *et al.*, *Physical Review A* **105** (2022) 022823.

← Many papers and PhD theses involved!

How ThF^+ (implementation)?



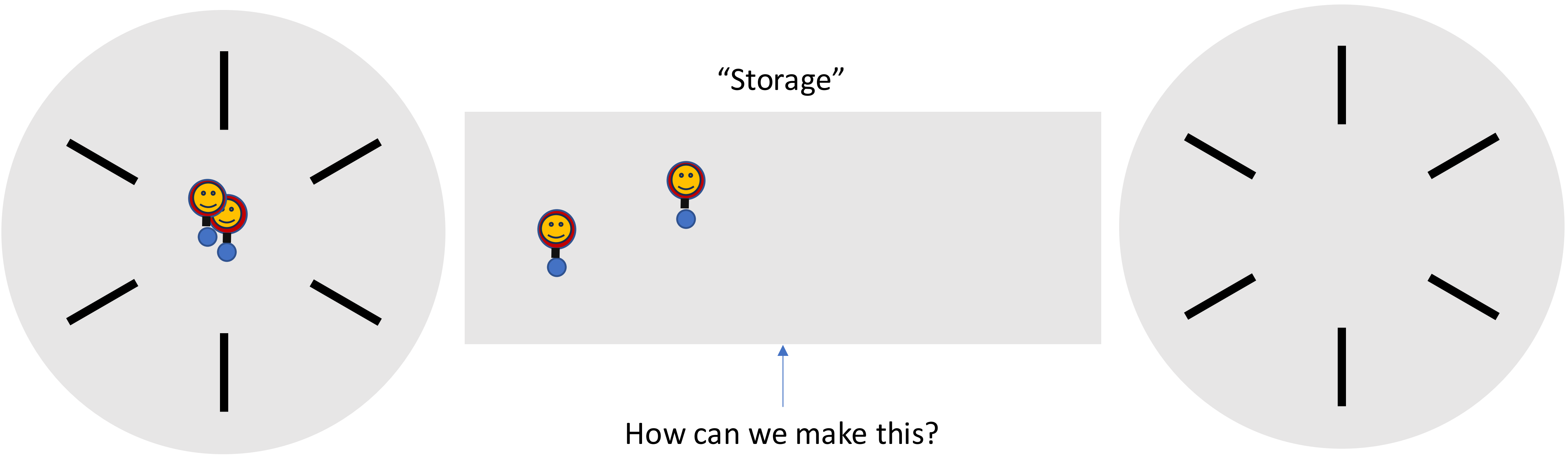
Cairncross, et al. *Physical review letters* 119.15 (2017): 153001.

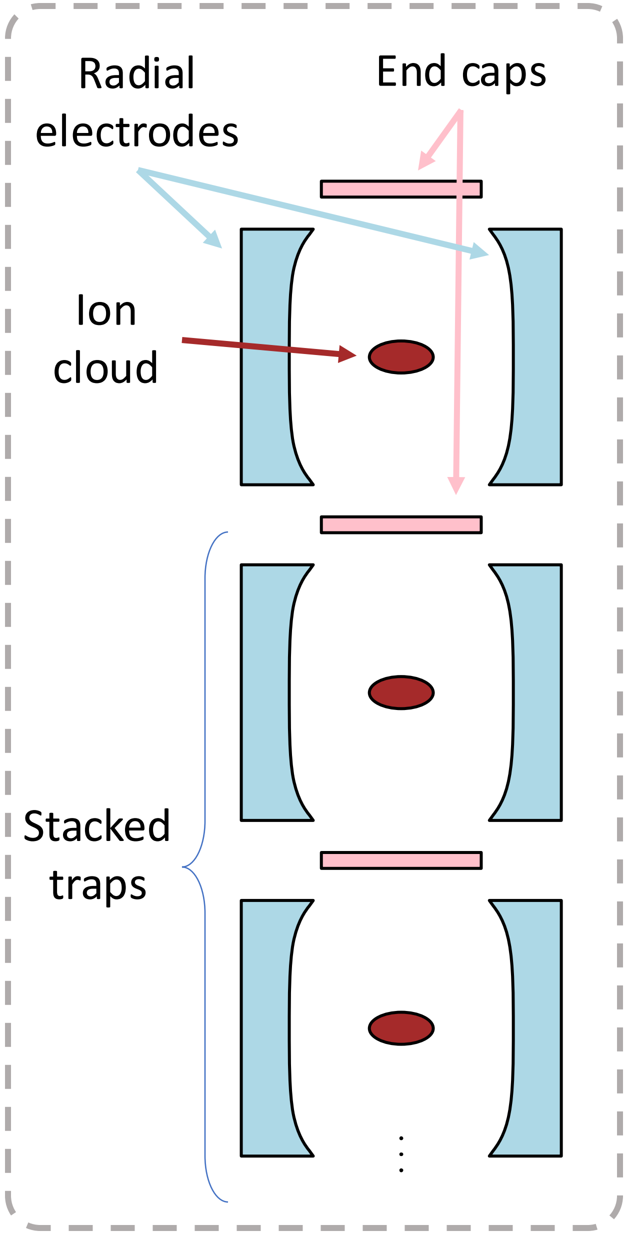
How ThF^+ (implementation): multiplexing

State preparation
- ~ 100 ms

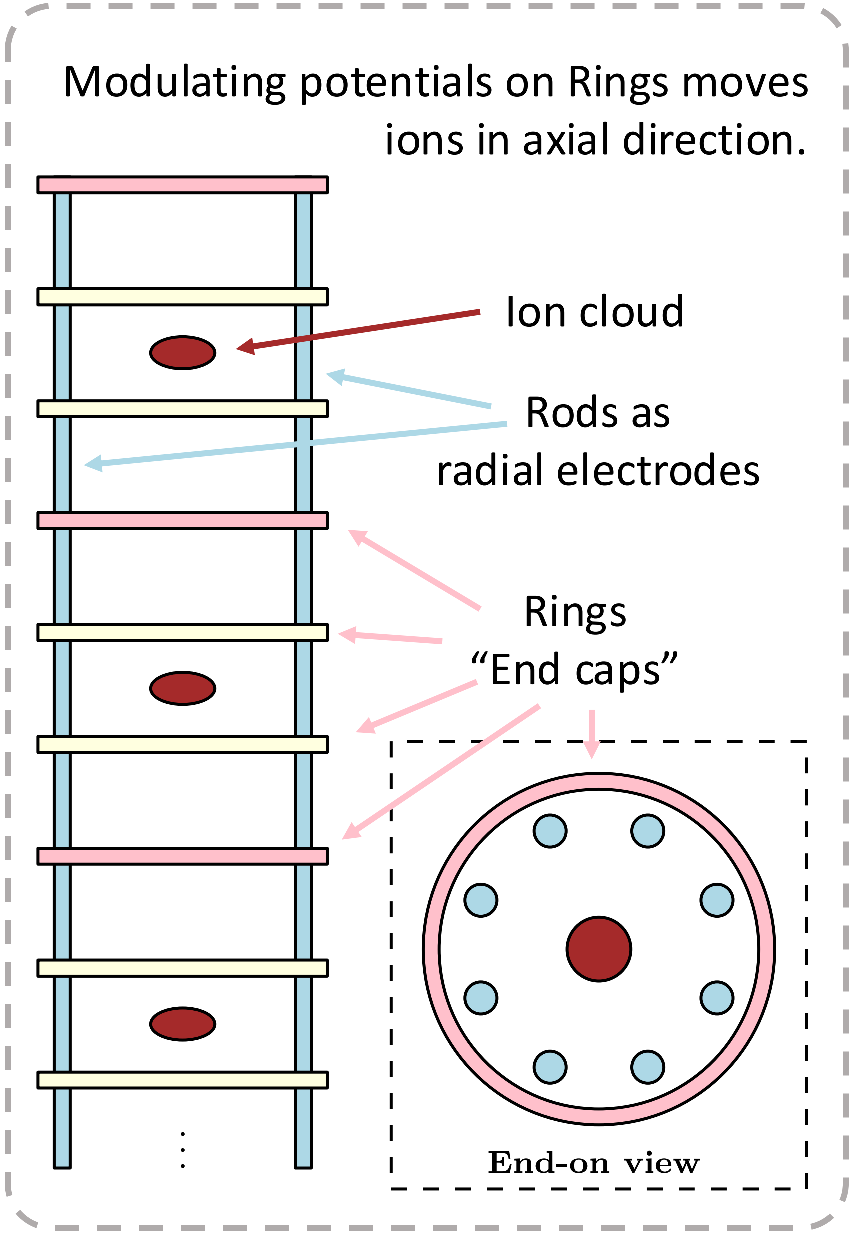
Spin precession
- ~ 10 s

State readout
- ~ 100 ms

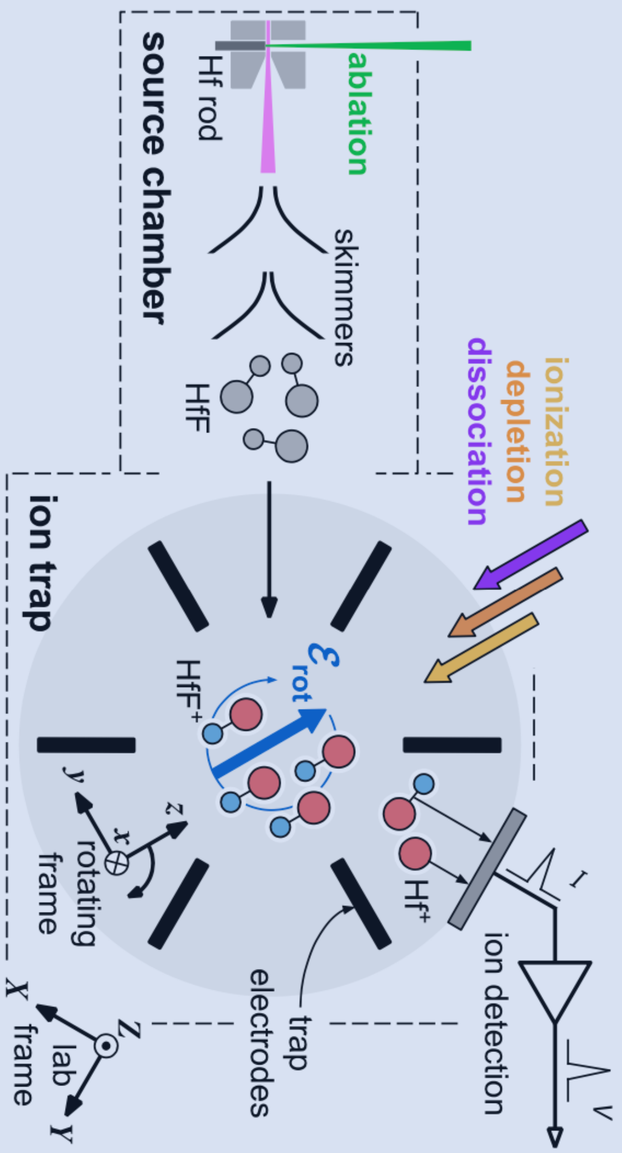



Radial electrodes
End caps
Ion cloud
Stacked traps

≡

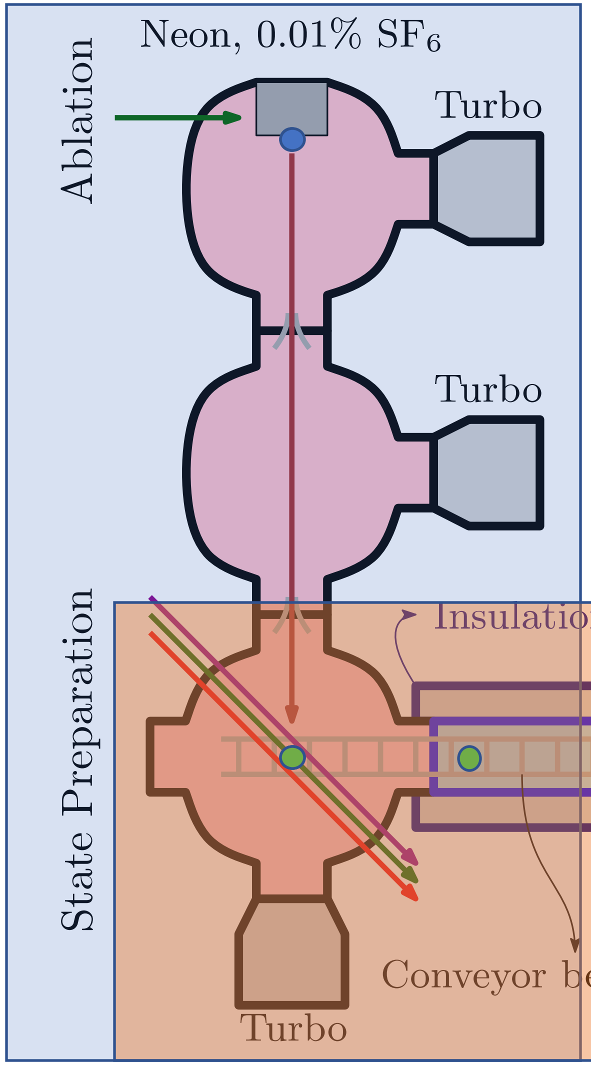

Modulating potentials on Rings moves ions in axial direction.
Ion cloud
Rods as radial electrodes
Rings
"End caps"
End-on view

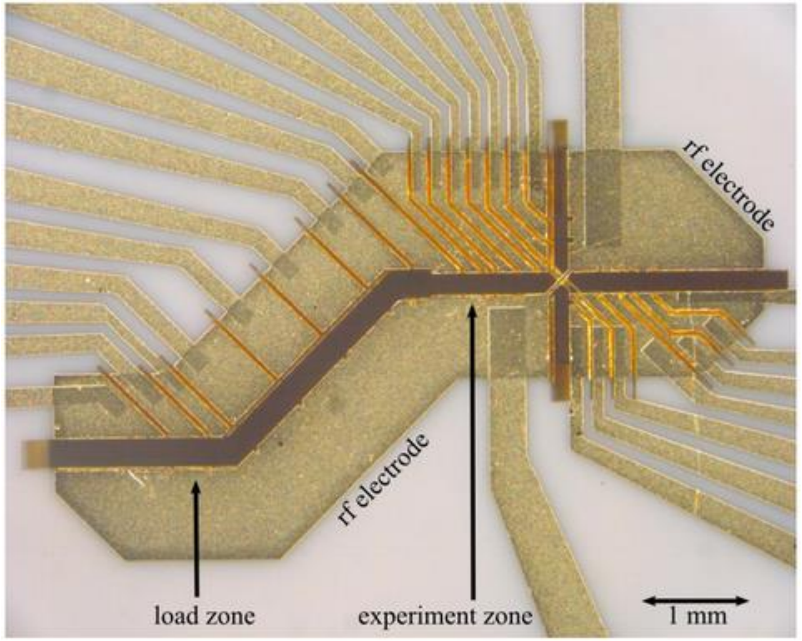
JILA eEDM Gens. I/II



≡

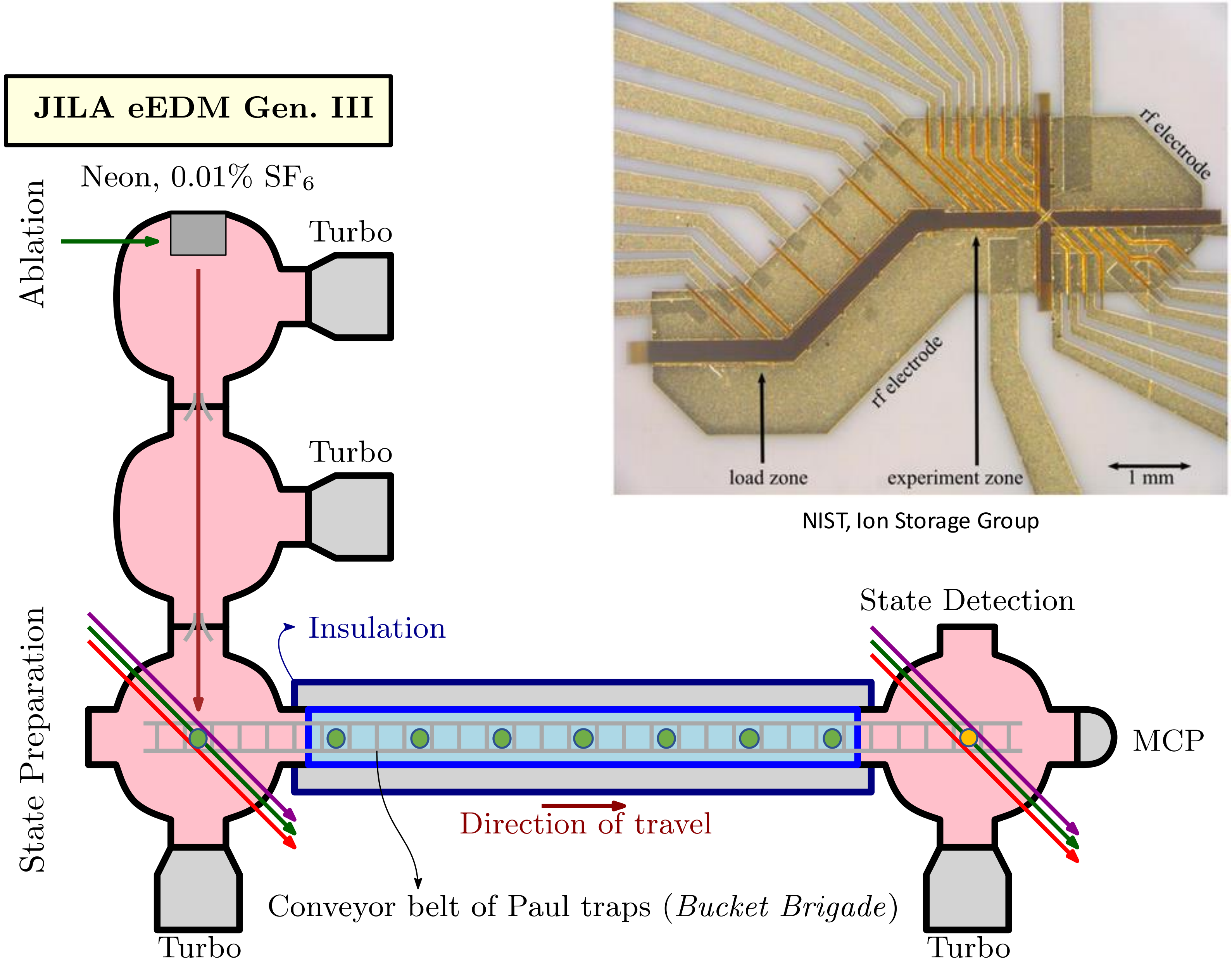
JILA eEDM Gen. III





NIST, Ion Storage Group

New in JILA eEDM Gen. III (*Bucket Brigade*)



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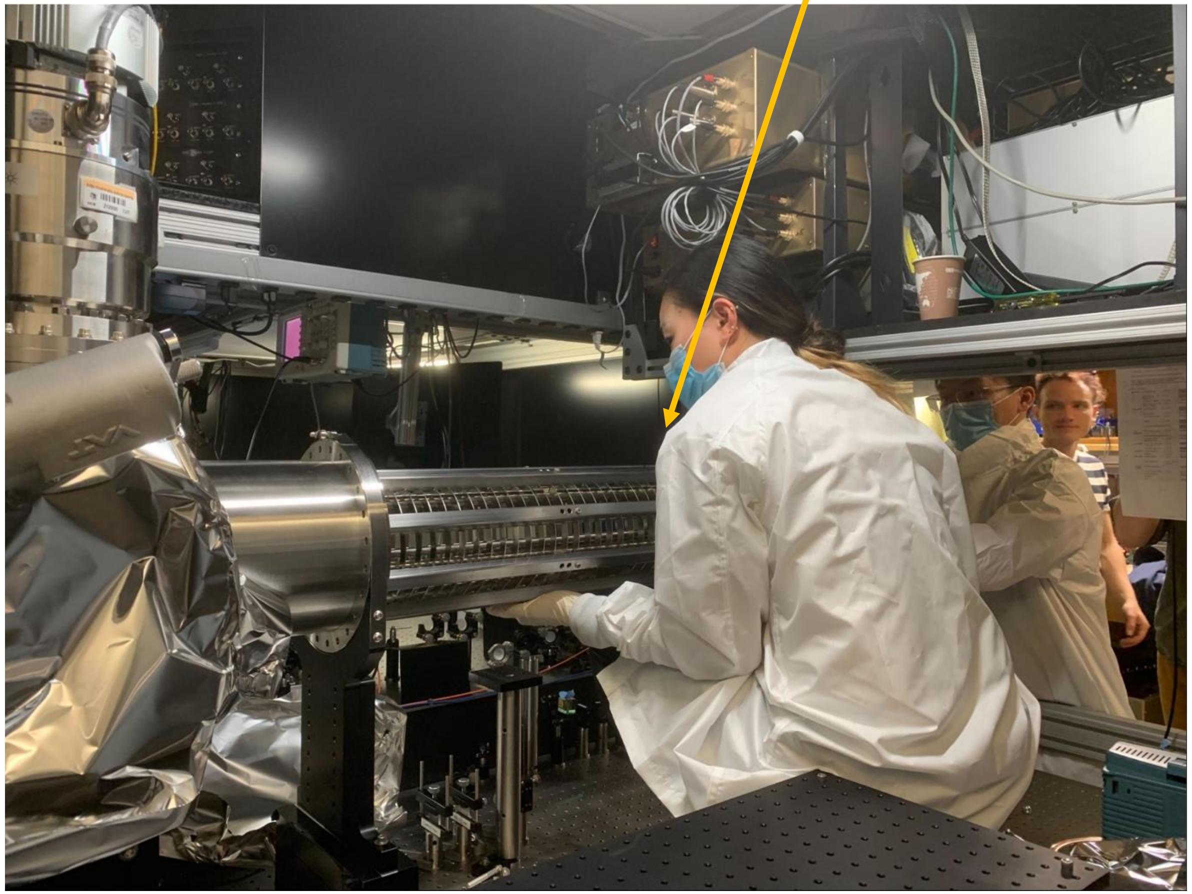
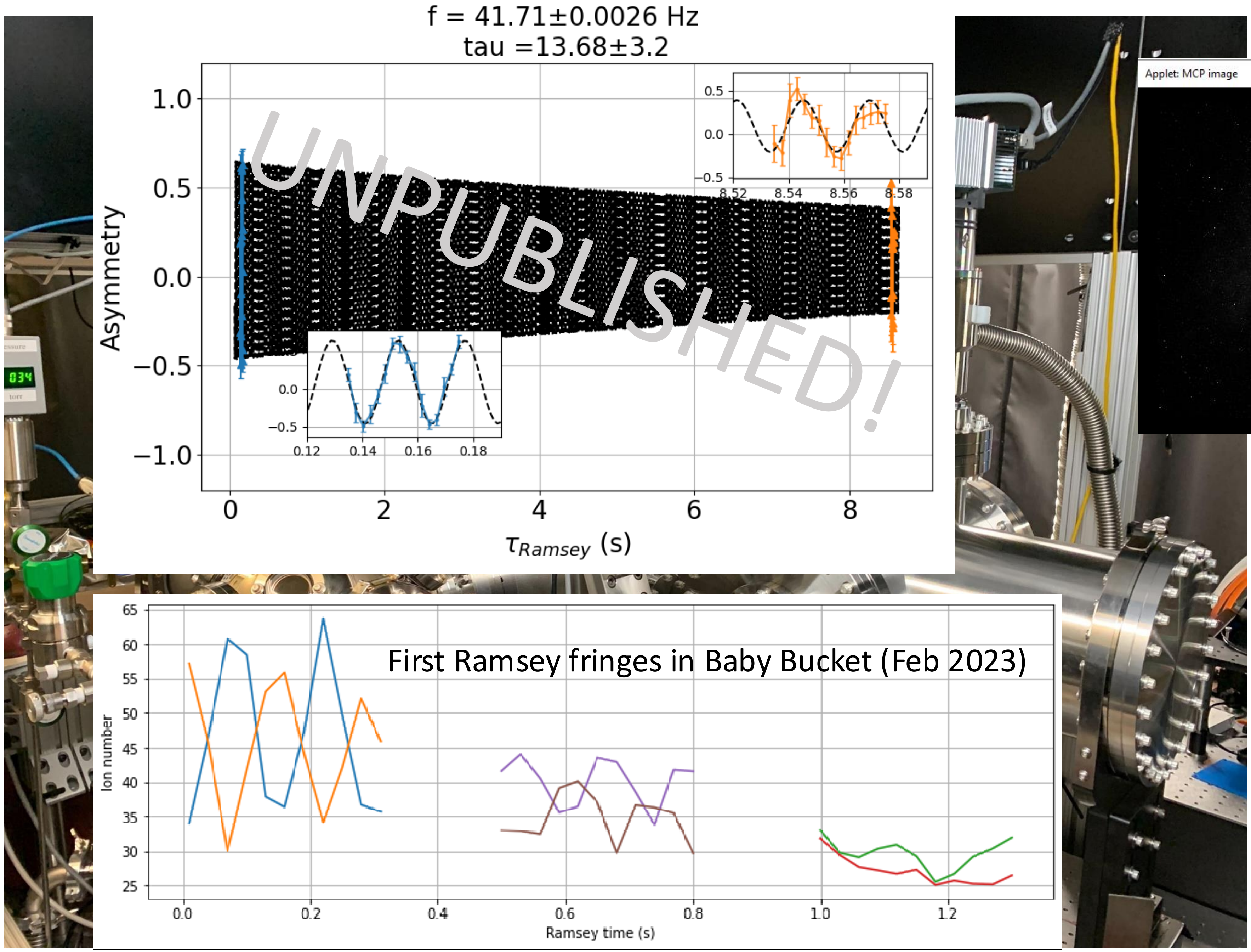
New in JILA eEDM Gen. III (*Bucket Brigade*)

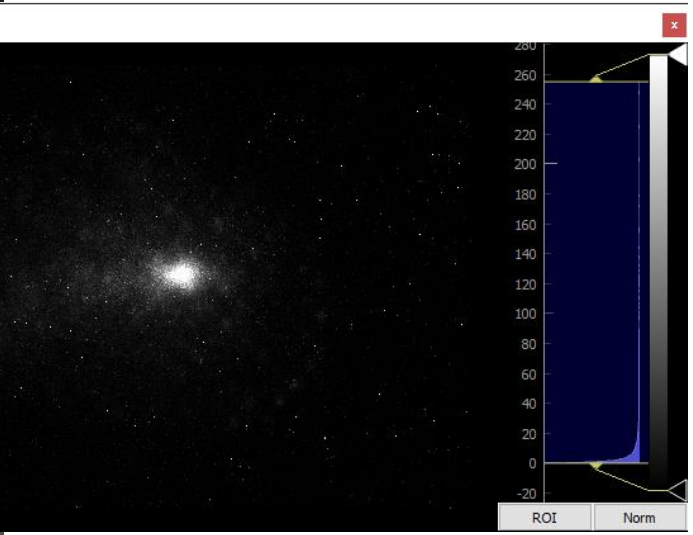
Electronics and structural design of the Baby Bucket



Loading the Baby Bucket into the vacuum chamber

Advisor included for scale





Above: we have ions!
(June 2022)

Left: overview of setup

Not shown: we have ion transport
(July 2022)

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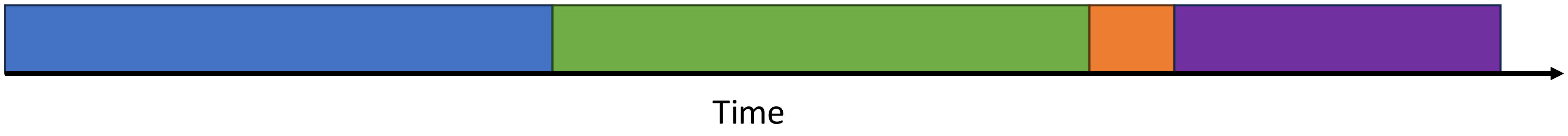

Why $^{227}ThF^+$

Complementary system to ongoing experiments like CeNTREX, FrAg, RaX, etc.

Sensitive to nuclear new physics

- Nuclear Schiff moment of ^{227}Th
  - Odd-*A* nucleus.
  - Octupole enhancement factor similar to ^{225}Ra.

Magnifies effects of new physics

- Molecular enhancement with ThF^+
  - Polarization of molecular orientation.
  - Polarization of electron cloud enhances molecular sensitivity to NSM.

Things are figured out, just do it!

- Quantum control scheme for ThF^+ already developed by JILA

Great minds think alike:
upcoming $^{229}ThF^+$ experiment at Harvard.

Flambaum, *et al.*, *Physical Review A* **101** 042504 (2020)
Liang *et al.*, *Physical Review C* **51** 1199 (1995)
Hammond *et al.*, *Physical Review C* **65** 064315 (2002)
Minkov, *et al.*, *Physical Review C* **110** 034327 (2024)
Eckel, *et al.*, *Physical Review A* **87** 052130 (2013)
Chen *et al.*, *Journal of Physical Chemistry A* **128** 31 (2024)

^{232}Th vs ^{227}Th

Developing efficient ion source with
Moritz Pascal Reiter (University of Edinburgh).

	^{232}Th	^{227}Th
Half life	14 billion years (~age of the universe)	19 days
Quantity	~10^{22} atoms 	~10^{12} No meaningful photo to share ; _ ;

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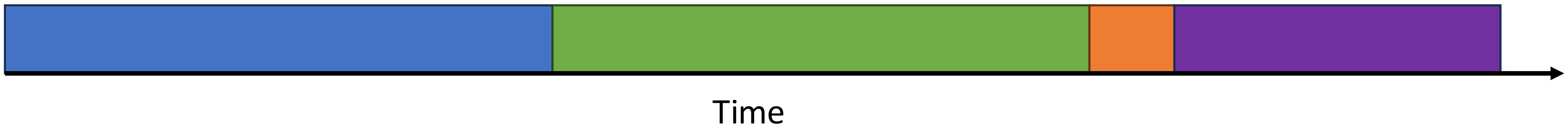


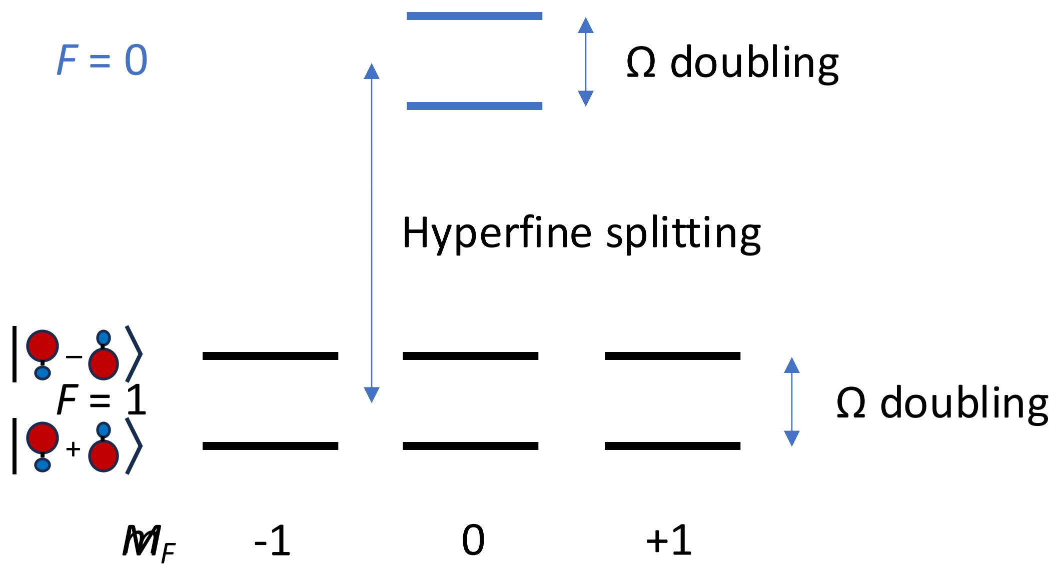
Molecular ions in a rotating electric field

- Polarizing E-field must rotate to keep ions confined
    - Fast compared to typical motion in ion trap.
    - Adiabatic compared to typical energy scales in molecule.

- Spectroscopy performed in rotating frame
    - Non-inertial interactions.

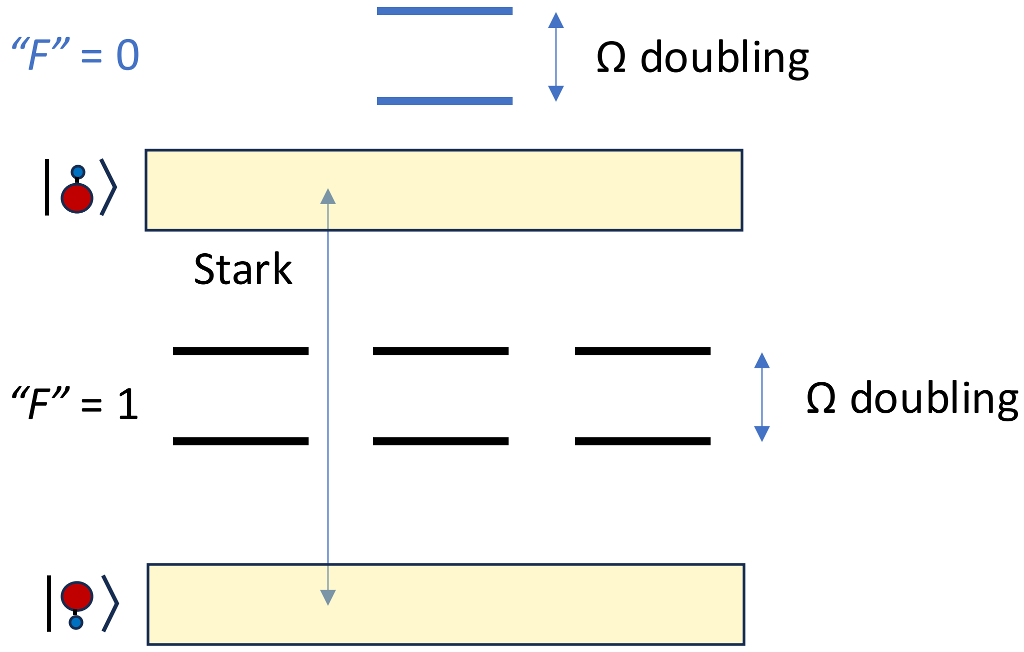
Molecular ions in a rotating electric field

- Molecular level structure (no E-field)
  - Zeeman sublevels.
  - Parity doublets.
  - Hyperfine.



Molecular ions in a rotating electric field

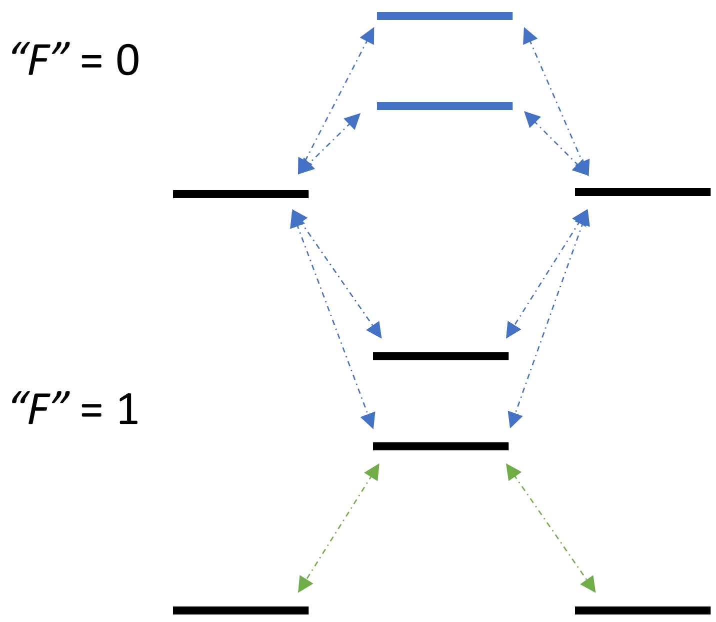
- Molecular level structure (with E-field)
  - *F* is an asymptotic quantum number.
  - Eigenstates have good orientation when $E_{\text{Stark}} >> E_{\Omega}$.



Measure EDM here because maximal spin projection

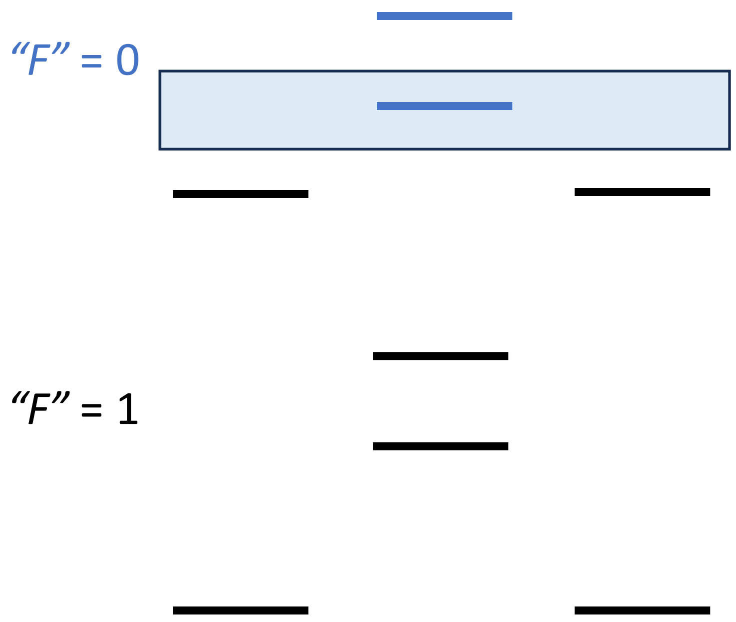
Molecular ions in a rotating electric field

- Non–inertial-frame coupling
  - Couples states of neighboring m_F.
- Degeneracy lifted by coupling.
- Magnitude of coupling effect
  - Sum of all possible $2m_F$-order perturbations.



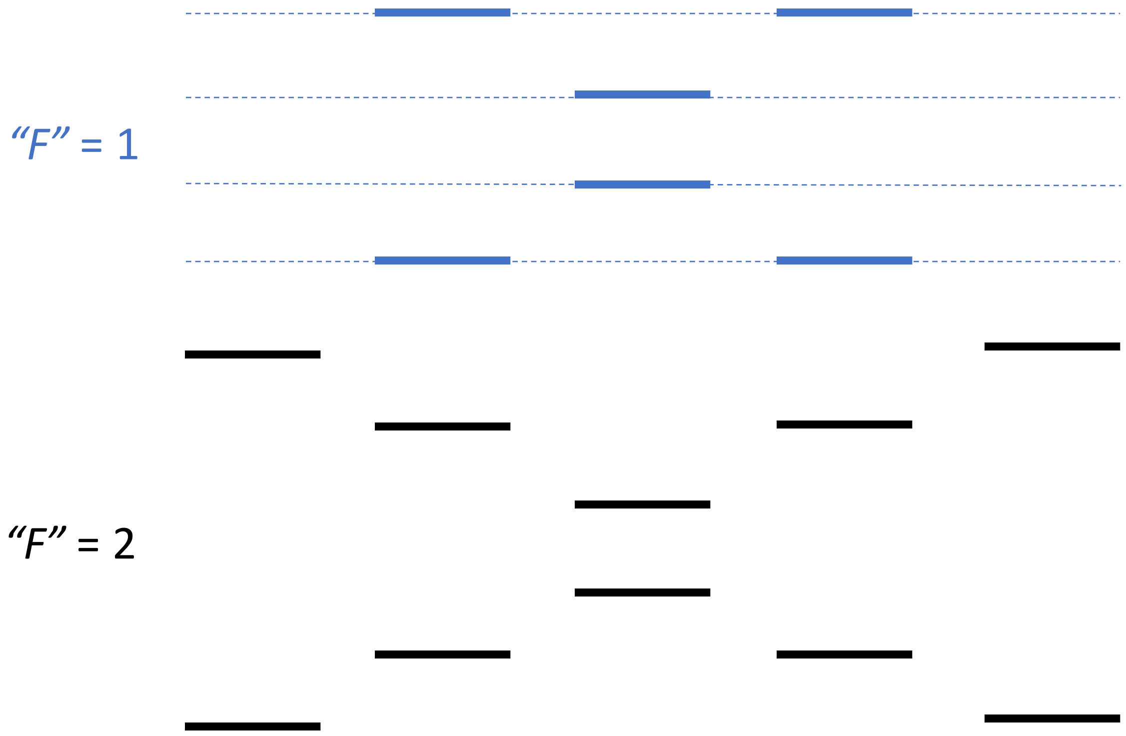
Molecular ions in a rotating electric field

- Non–inertial-frame coupling
  - Couples states of neighboring m_F.
- Hyperfine crossing
  - Stretched states are strongly mixed with each other.



Molecular ions in a rotating electric field

- Non–inertial-frame coupling
  - Couples states of neighboring m_F.
- Hyperfine crossing
  - Stretched states are strongly mixed with each other.
  - More hyperfine crossings in larger Hilbert spaces.



Molecular ions in a rotating electric field

- Non–inertial-frame coupling
  - Couples states of neighboring m_F.

- Bad news
  - Strategic choice of E-field strength.
  - Unstretched states harder to use than stretched ones.
  - Electronics are move involved.

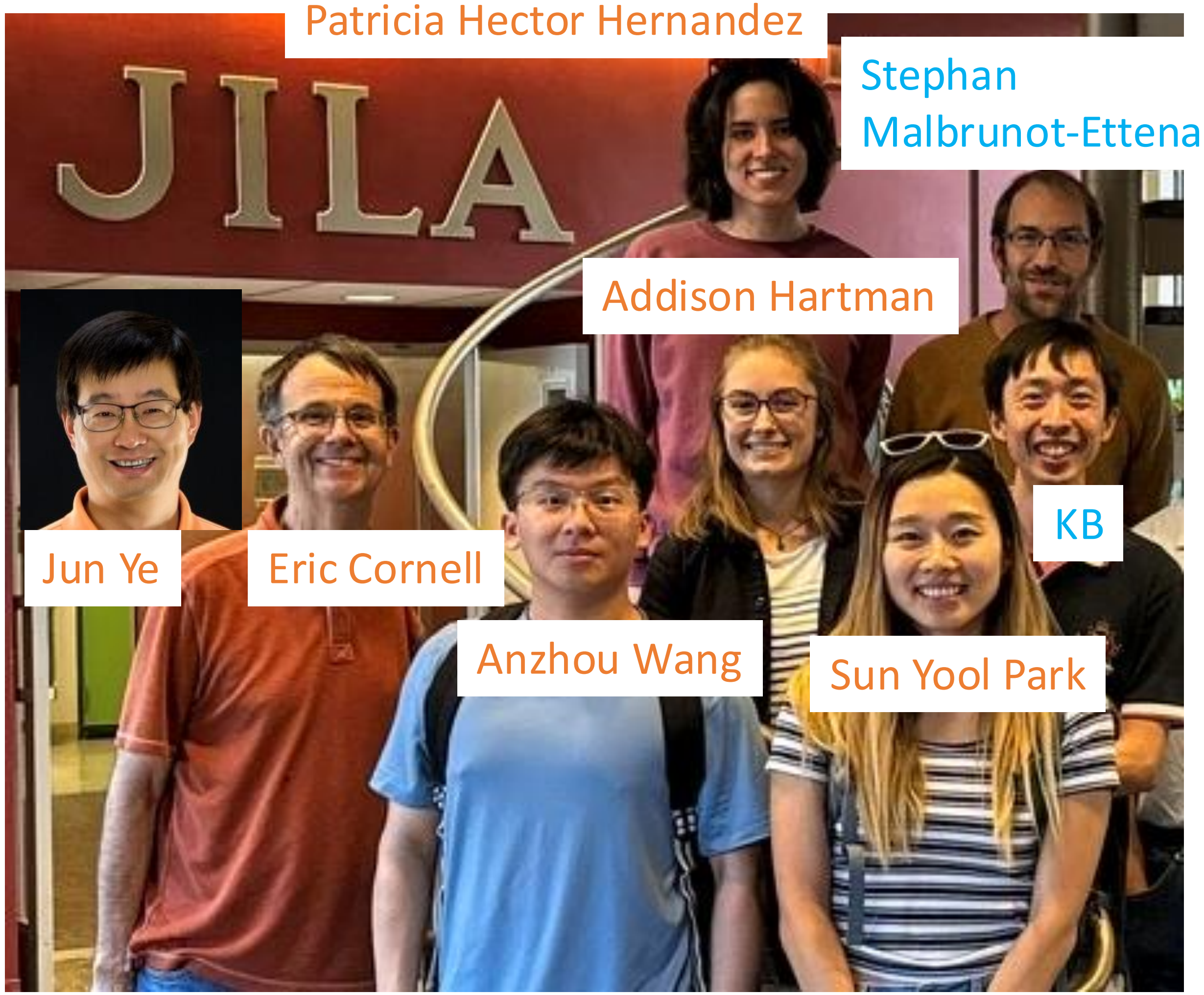
- Good news
  - Resource to introduce coupling between stretched states.
  - Powerful systematics rejection with "lock-in detection".
  - Opportunities for more quantum state manipulation techniques!

Ng, *et al.*, submitted to PRA (2025)

Thank you!

We are hiring students!

TRIUMF
NSERC CRSNG
NSF
NIST



Lan Cheng

Valery Radchenko

Ed Riley

Stephanie Cui

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