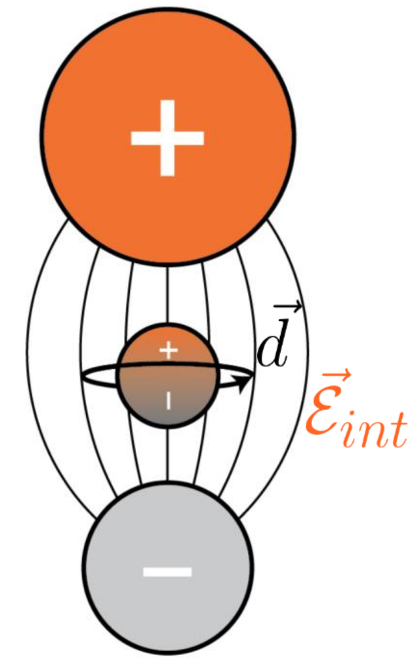
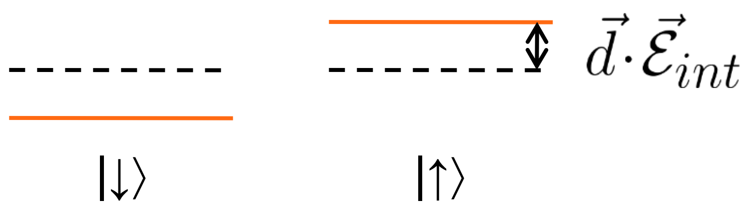
CPV Searches with Polyatomic Molecules: *Quantum Control and Exotic Nuclei*

Nick Hutzler
Caltech

Molecular Sensitivity

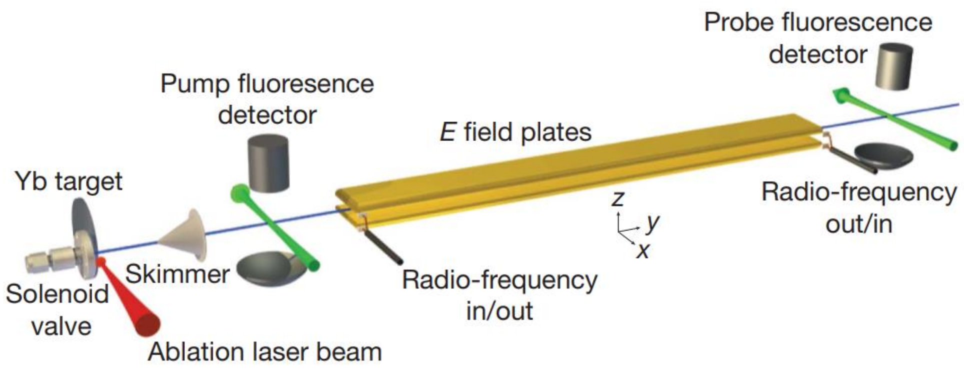
- Molecules contain large fields
  - Amplifies CPV observables
  - ~1-100 GV/cm effective fields
  - Vary roughly as $\sim Z^{2-3}$
  - Measure with coherent methods
- Molecule experiments are already probing beyond the reach of colliders
- Major advances coming!
  - Higher sensitivity
  - Access to new effects
  - **Many exciting approaches**



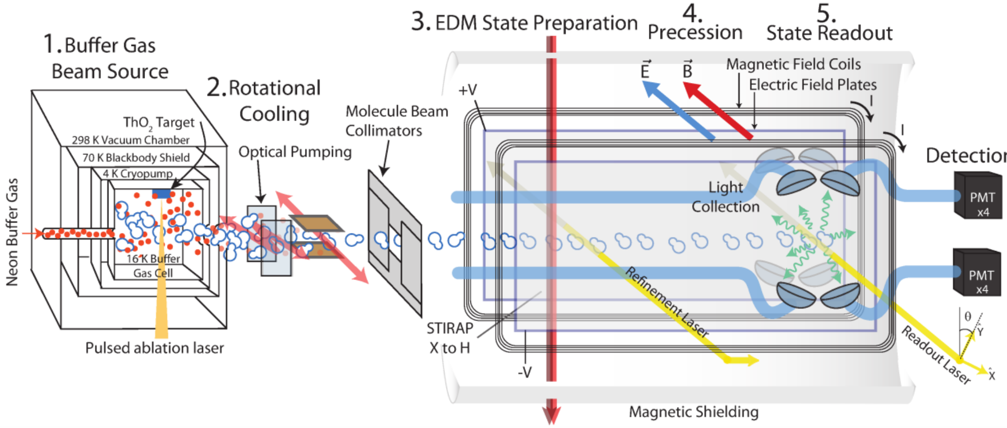


$$|\uparrow\rangle + |\downarrow\rangle \rightarrow |\uparrow\rangle + \boldsymbol{e^{i\phi}}|\downarrow\rangle$$

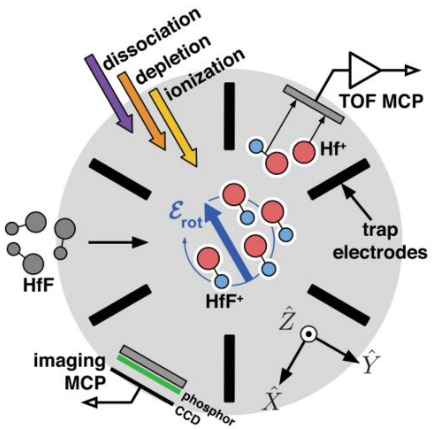
Molecular Experiments



YbF, Imperial
- Spin precession in pulsed supersonic beam
- First to beat atomic Tl limits
- $|d_e| < 1.1 \times 10^{-27}$ e cm (2011)



ACME, ThO, Harvard/Chicago/Northwestern
- Spin precession in cryogenic beam
- $|d_e| < 8.7 \times 10^{-29}$ e cm (2014)
- $|d_e| < 1.1 \times 10^{-29}$ e cm (2018)



HfF^+, JILA/Boulder
- Spin precession in ion trap
- $|d_e| < 1.3 \times 10^{-28}$ e cm (2017)
- **$|d_e| < 4.1 \times 10^{-30}$ e cm (2023)**

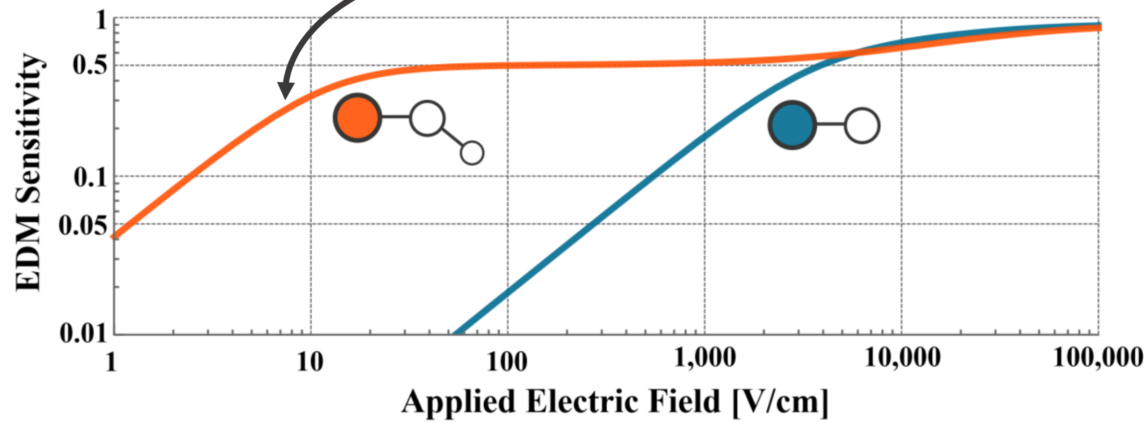
- **~300x in ~12 years**
- **Each is being upgraded**
- **More are under way**
- **Extending to nuclear CPV**

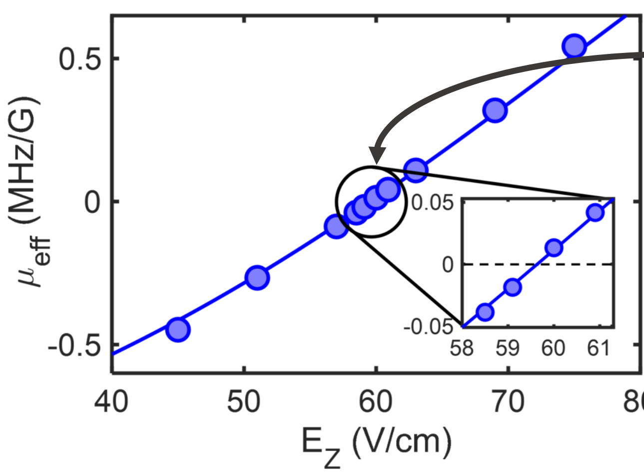
Quantum Control

Advantages of Polyatomics

- Opportunities from additional degrees of freedom
  - High polarizability
  - Tunable electro-magnetic sensitivity
  - Robustness against systematic errors
- Can engineer for the science, protocol, and platform

High polarizability in small fields



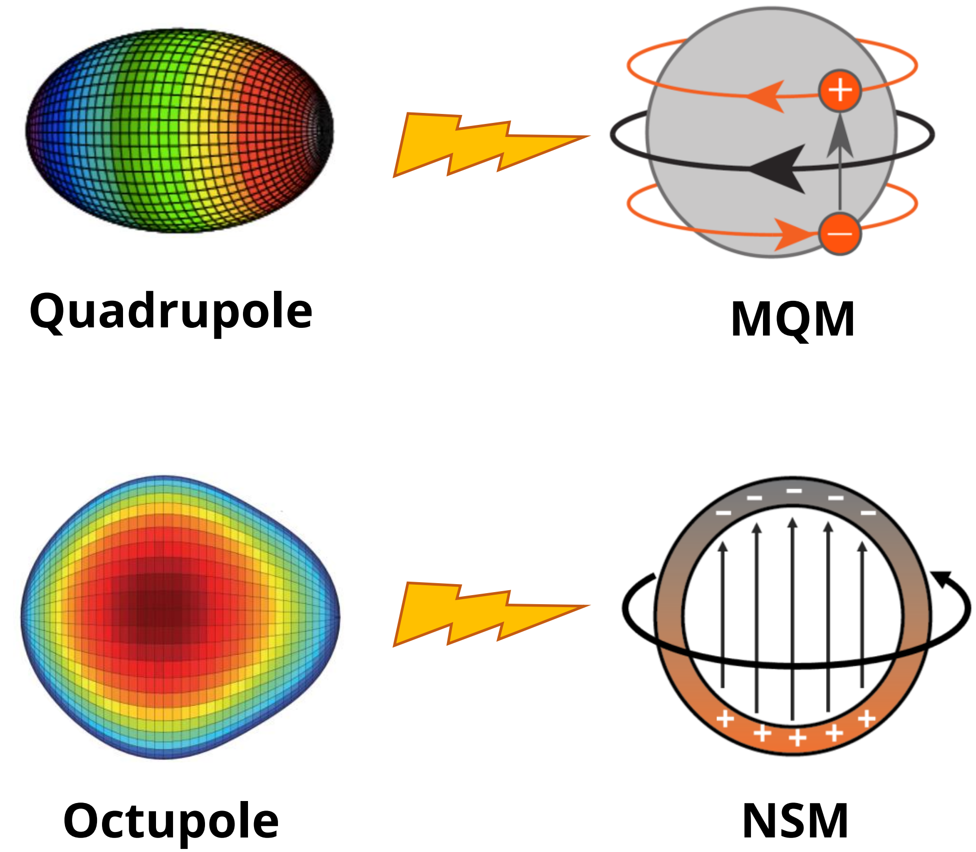


Magnetically insensitive states

L. Anderegg, N. B. Vilas, C. Hallas, P. Robichaud, A. Jadbabaie, J. M. Doyle, NRH, Science 382, 665 (2023)
Y. Takahashi, C. Zhang, A. Jadbabaie, NRH, PRL 131, 183003 (2023)

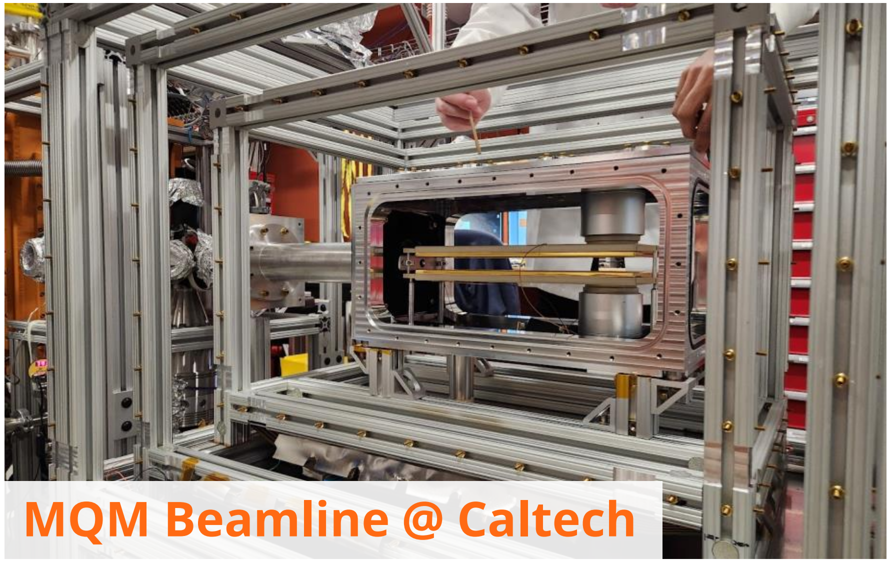
Nuclear Sensitivity

- Access different physics vs. eEDM
  - Nucleon EDMs, quark EDMs, strong force, nuclear forces, …
- Nuclear symmetry violations can be enhanced by nuclear properties
  - In addition to molecular enhancement
- Magnetic quadrupole moment (MQM)
  - Enhanced by quadrupole shape
  - Typically ~10x
- Nuclear Schiff moment (NSM)
  - Enhanced by octupole shape
  - Typically 100-1,000x
- Manifest similarly to EDMs
  - “EDM experiments”



^{173}YbOH NMQM @ Caltech

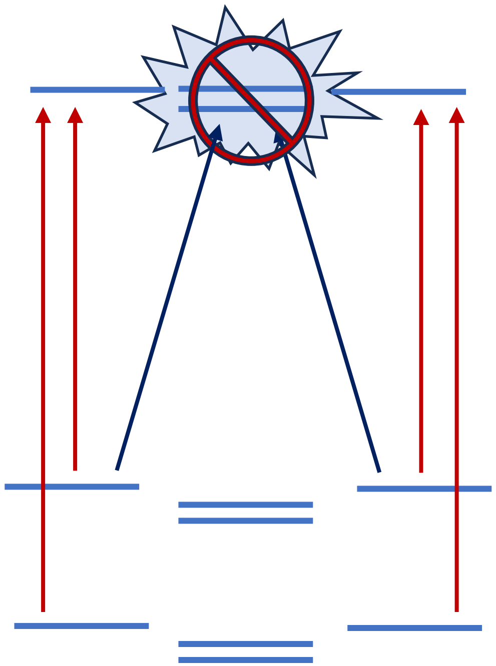
- MQM search in ^{173}YbOH
  - Large quadrupole deformation in Yb
  - Large molecular enhancement
  - Optical control/readout by photon cycling
  - Disentangle EDM, NSM, MQM via hyperfine dependence
- Cryogenic molecular beam experiment
- Laser cooling in future generations
- Currently implementing measurement protocol

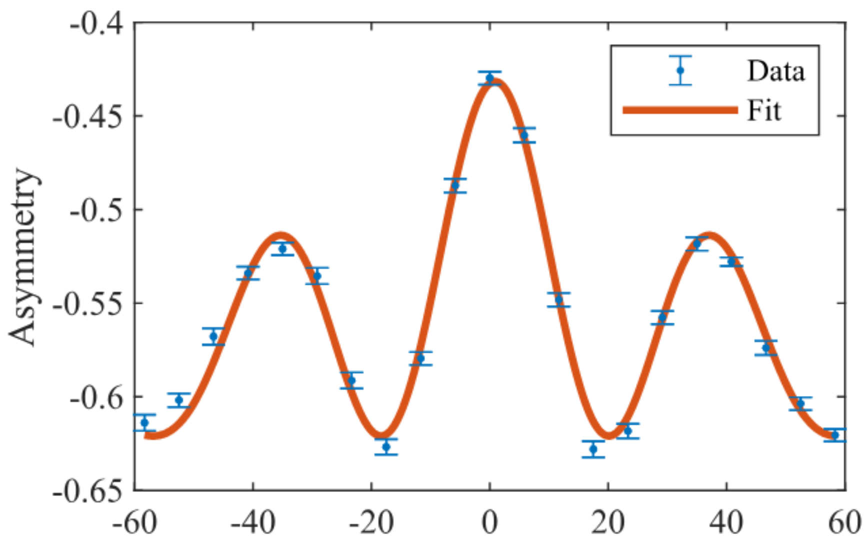


MQM Beamline @ Caltech

Spin Precession Protocol

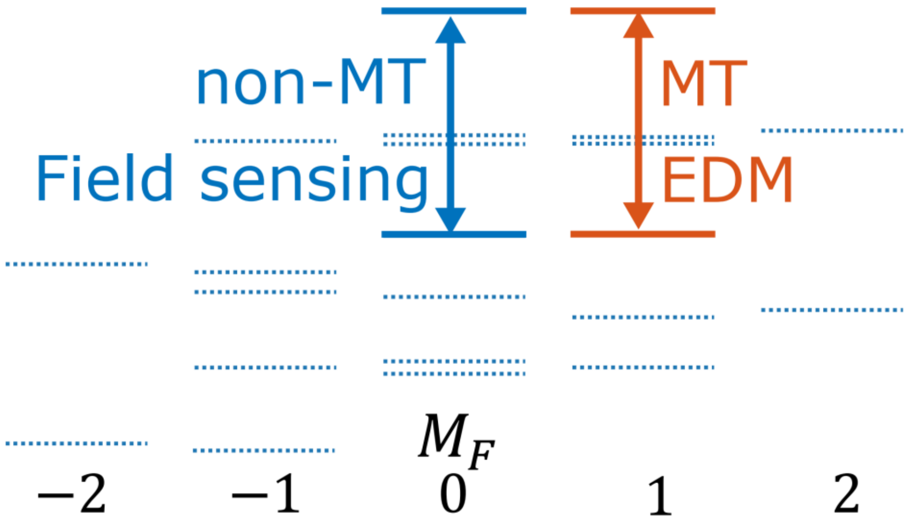
- Problem: $|+M\rangle \pm |-M\rangle$ is hard
  - M is large
    - $I_{Yb} = 5/2, S = 1/2, \; N = 1, I_H \; = \; 1/2$
  - Beam has velocity dispersion and large spatial extent – hard to use microwaves
  - Unresolved excited state hyperfine structure – hard to use lasers
- Solution: Give up
  - Can instead prepare $|M\rangle \pm |M'\rangle$
  - Choose $|M - M'| \leq 2$ to connect with two-photon transition
- This works fine, but has first order Stark and Zeeman shift
  - Large molecular dipole
  - Unpaired electron spin





Polyatomic-Enabled Protocol

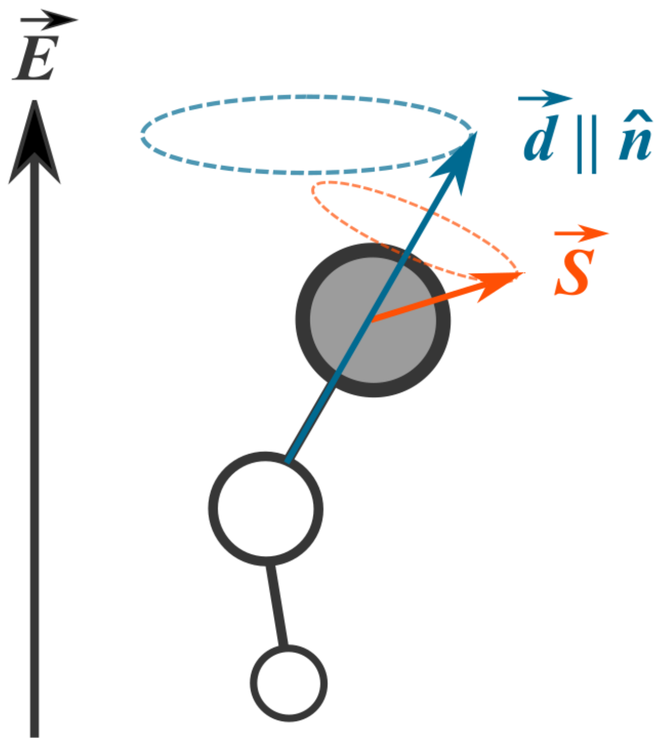
- Solution: Field-insensitive "magic" transitions
  - Can find $\mathcal{E}$ where Stark, Zeeman <0.1%, EDM >30%
  - Static CPV shifts → can use traditional spin precession
  - Can change sign of Stark, Zeeman, EDM, shifts
  - Simultaneous sensing of fields with "non-magic" transitions
- Exist generically in polyatomics – agnostic to details of molecular structure
- Applicable to electron and nuclear symmetry violation
  - EDM, NSM, and MQM



^{174}YbOH science state
E ~ 40 V/cm, B ~ 12 mG

Proposal: Y. Takahashi, C. Zhang, A. Jadbabaie, NRH, PRL 131, 183003 (2023)

Intuitive Picture

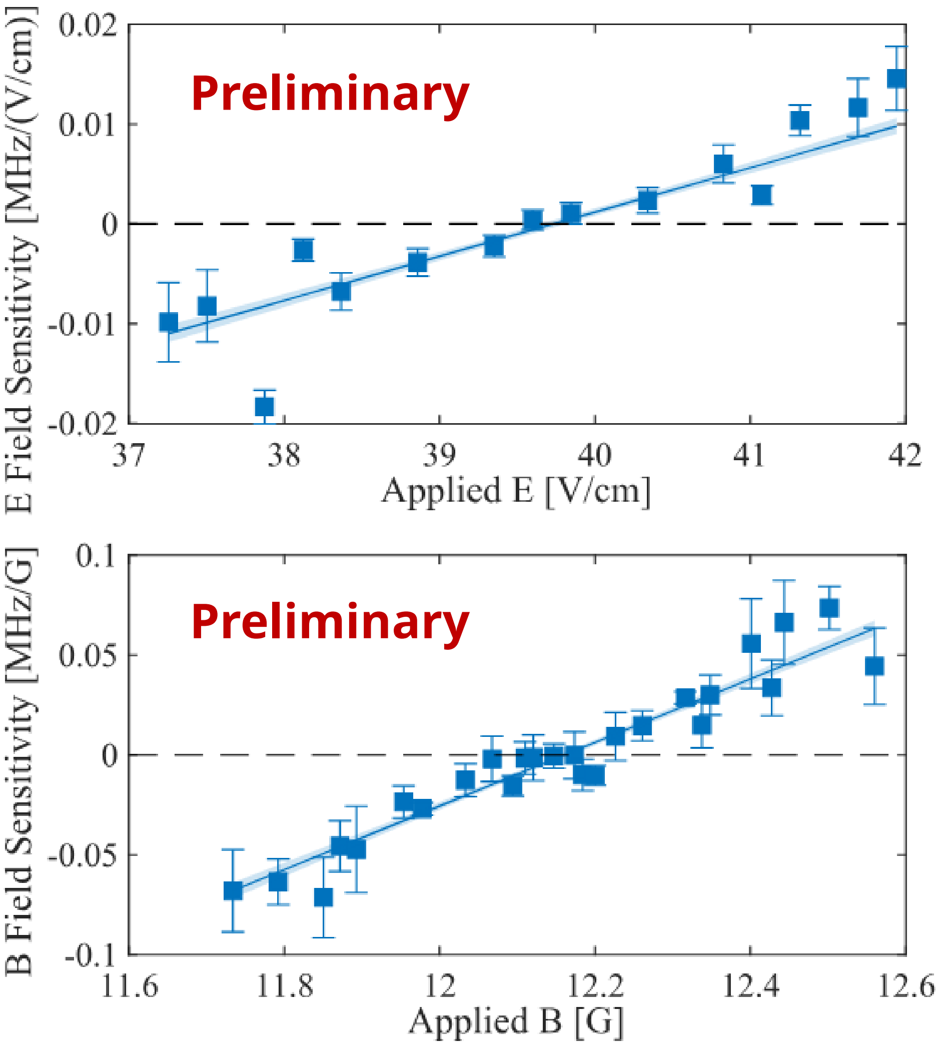


$$|\omega| \sim |\vec{d} \cdot \vec{E}|$$

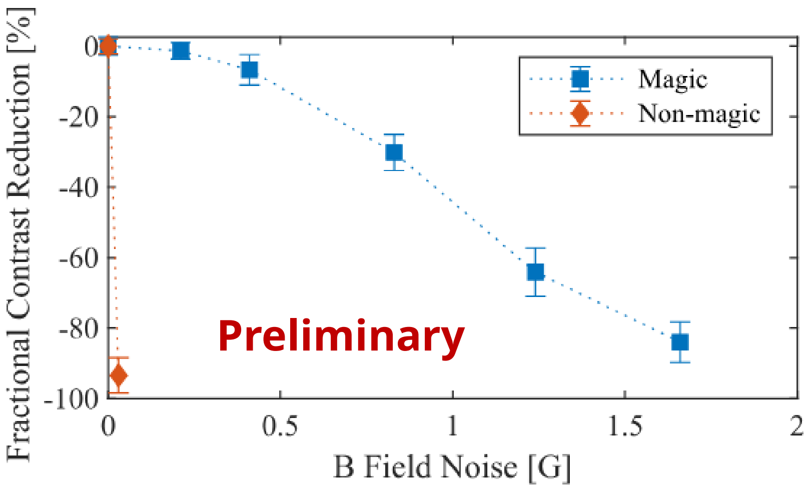
$$|\omega| \sim |\gamma \vec{S} \cdot \hat{n}|$$

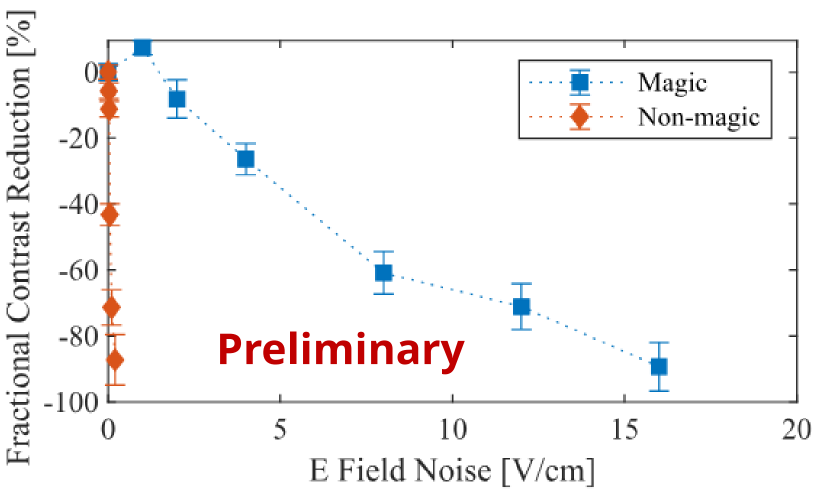
Implementation in ^{174}YbOH

- Recently implemented in ^{174}YbOH science state
  - $\mathcal{E}_{eff} \approx 22$ GV/cm @ 40 V/cm
  - Preliminary – data from the last few months
- Can see EDM-sensitive Ramsey fringes
- Able to simultaneously suppress electric and magnetic sensitivity to <1%
  - < Few μ_N magnetic sensitivity in a molecule with an unpaired electron

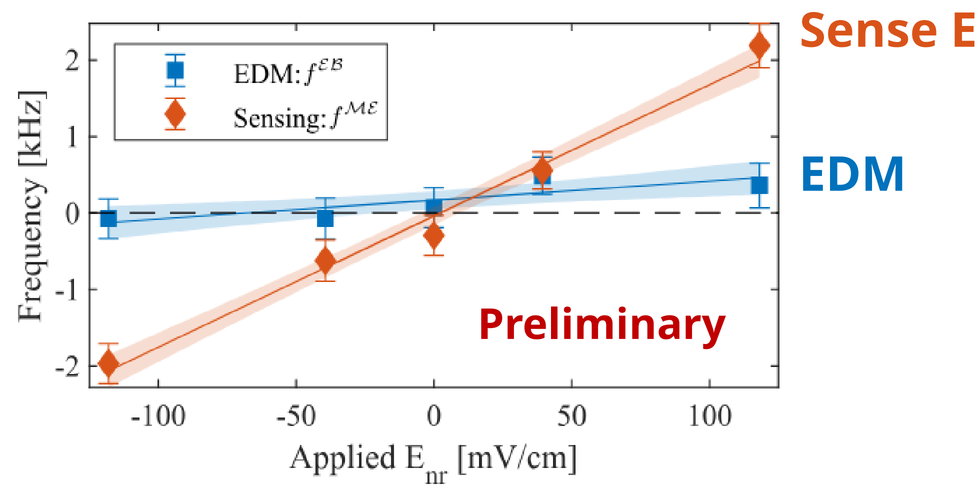


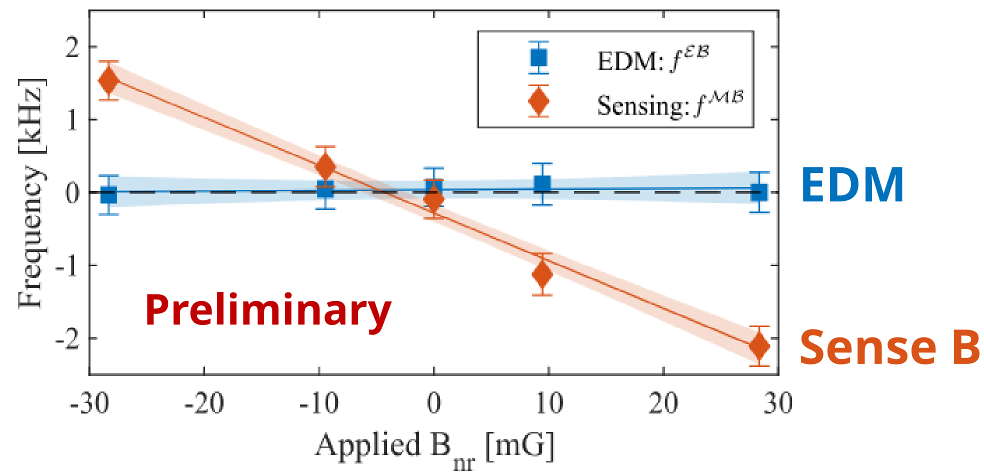
Implementation in ^{174}YbOH





Highly robust against electromagnetic noise

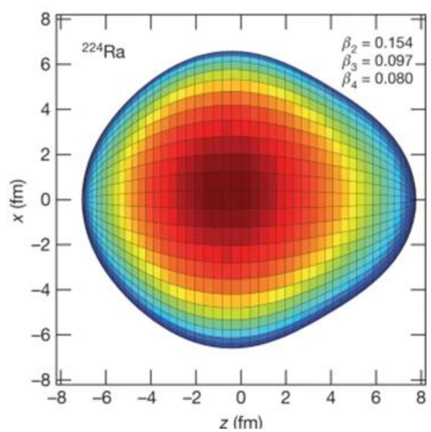




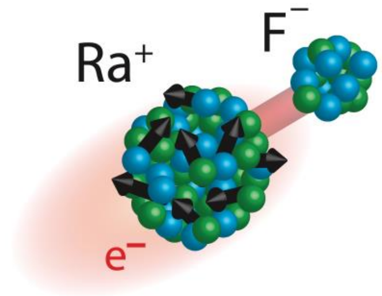
Ability to use field-sensitive transitions to sense fields

Radioactive Molecules

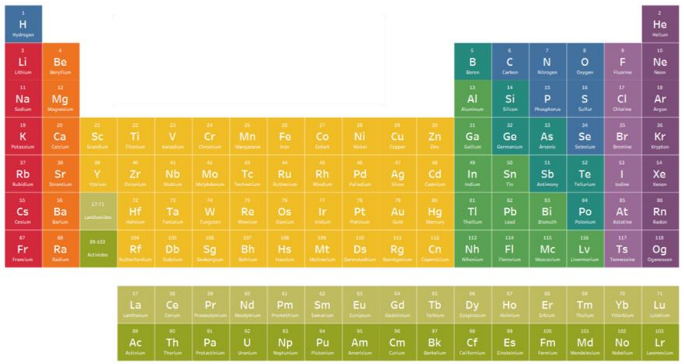
Why Radioactive Molecules?



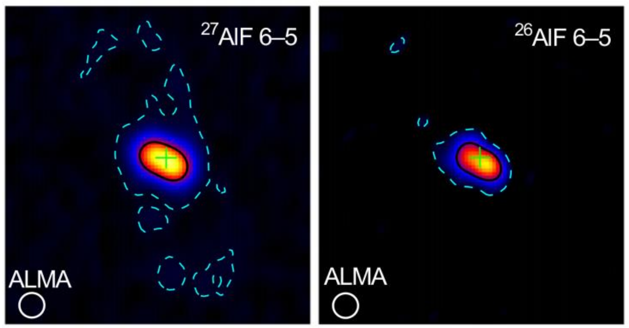
Fundamental symmetries



Nuclear structure



Radiochemistry

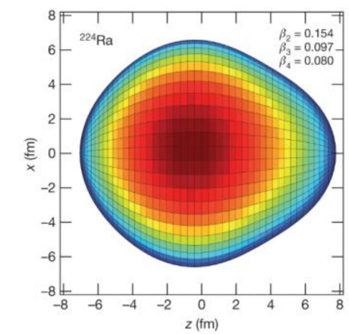


Nuclear astrophysics

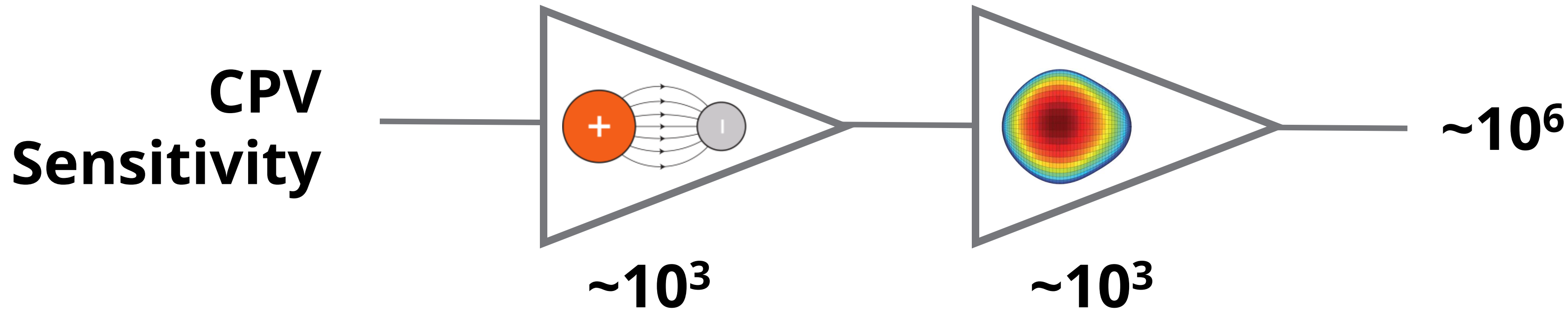
- Molecules containing exotic nuclei have interesting properties [see 2302.02165]
- Fundamental challenge: adapting many laboratory techniques to trace, short-lived species

Enhanced CPV Sensitivity with Exotic Nuclei

- Nuclear symmetry violations enhanced in heavy, octupole-deformed (pear-shaped) nuclei
  - Combines with molecular sensitivity enhancement



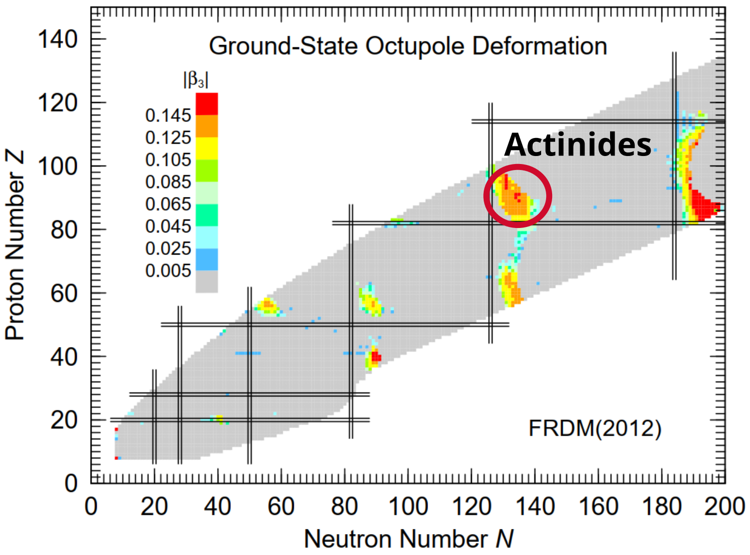
L. P. Gaffney et al., Nature 497, 199 (2013)



- Control *one* molecule at a time → hadronic frontier
- Challenge: even producing radioactive molecules is a research frontier

Radioactive Nuclei

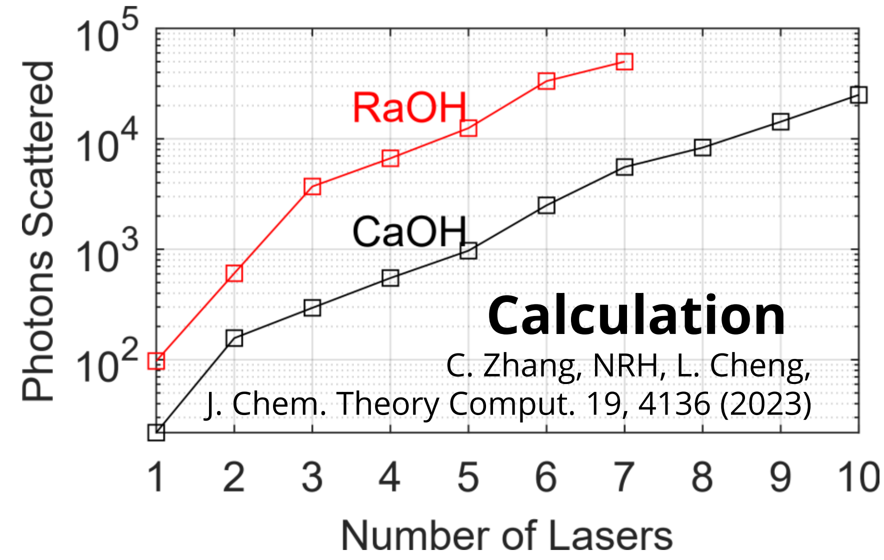
- Main challenge: heavy, spinful, octupole-deformed nuclei are radioactive
- Nuclei must be in a molecule amenable to "precision measurement" methods
  - Synthesis, cooling, spectroscopy, measurement protocol, coherent quantum state control, …

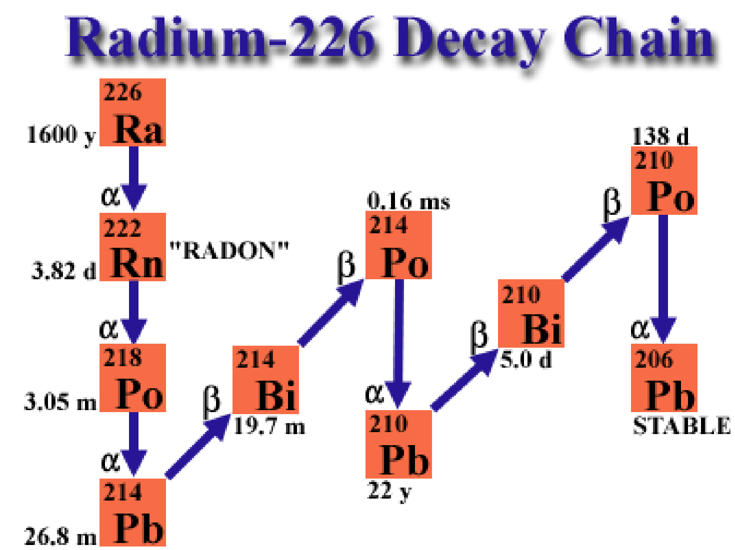


Isotope	Half-life
^{223}Fr	22 min
^{225}Ra	15 d
^{223}Ra	11 d
^{227}Ac	22 yr
^{229}Th	7,900 yr
^{229}Pa	2 d

Radium Molecules

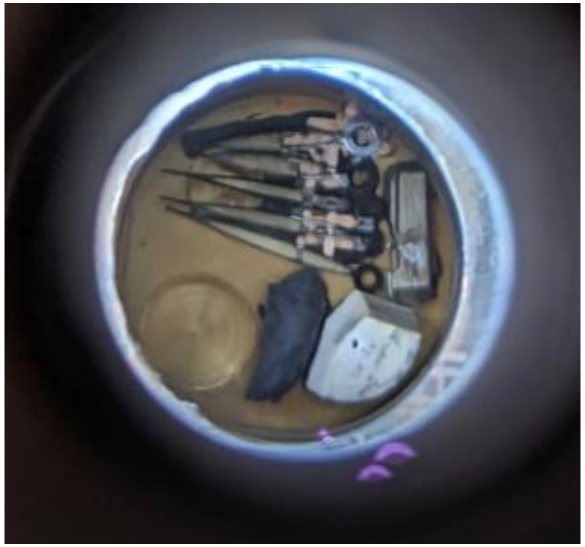
- Radium-containing molecules are interesting
- Advantages:
  - Octupole deformed
  - Very good theory support
  - High CPV sensitivity
  - Makes optically-controllable molecules for advanced atomic/molecular physics methods
  - **Demonstrated measurement protocols**
- Motived groundbreaking RaF spectroscopy work led by R. F. Garcia Ruiz
- Challenges
  - **Hard to get**
  - Small quantities
  - Biologically hazardous
  - Long-lived radon daughters
  - Large theoretical uncertainties
  - Even “normal” molecules take years to study





Radium Supply

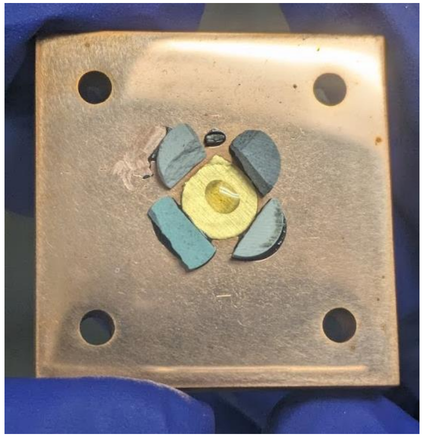
- We have started making cryogenic, radium-containing molecules @ Caltech
- So far Ra-226
  - Will move toward Ra-225, Ra-223
  - Also interested in things like Th, Pa, … ?
- First supply: eBay watch repair store
  - Worked, but not a long-term solution…
- Second: Eckert & Ziegler
  - 10 μCi (370 kBq) $RaCl_2$ solution
  - Worked great, immediately compatible with target production methods
  - Too expensive for long-term use
- Current: NIDC
  - 1 mCi (37 MBq) $Ra(NO_3)_2$ dried salt
  - Needed to dissolve, aliquot to ~50 μCi experimental quantities
  - Thanks Alyssa Gaiser!
- **We are starting to have trouble sourcing material**



Alyssa Gaiser, MSU/FRIB

Ablation Target Production

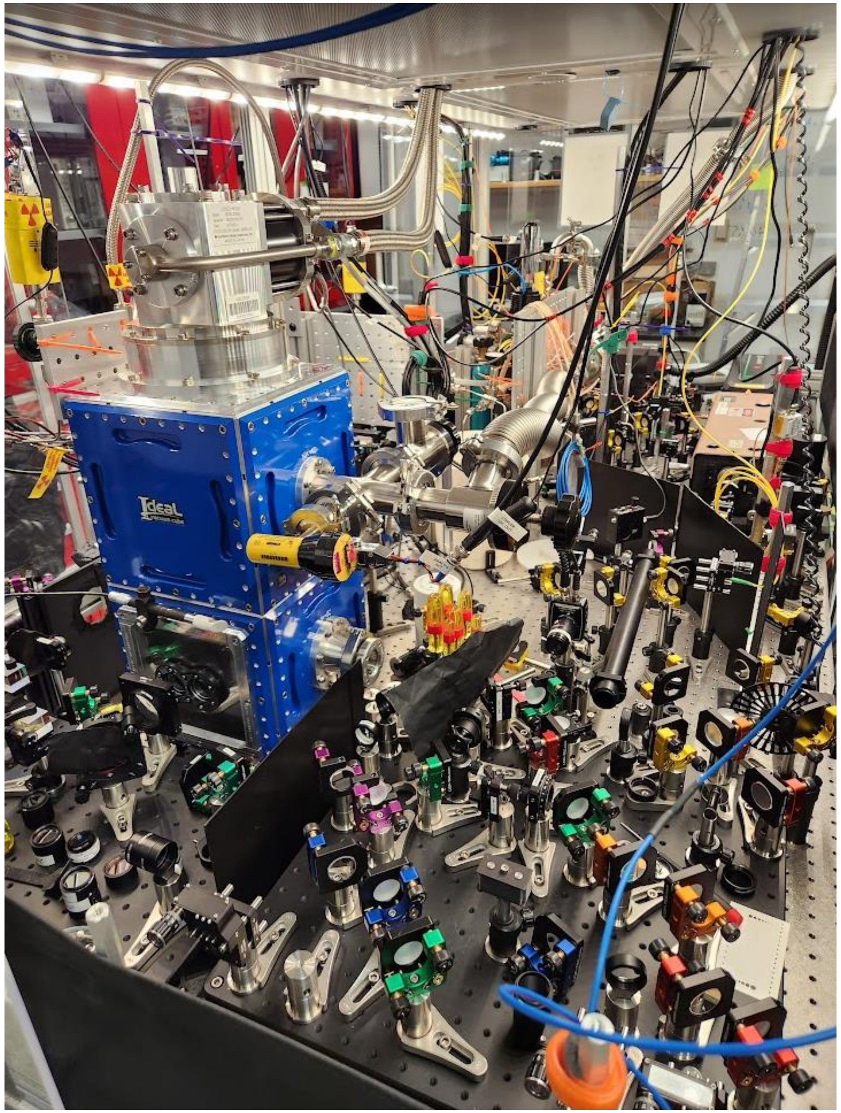
- We want a uniform, repeatable, stable ablation target which survives at 4 K
- Drop cast $RaCl_2$ or $Ra(NO_3)_2$ solution onto hot surface
  - Use isotope lab @ Caltech
- Test with barium, but complicated by radiolysis
  - H_2O + rad → e^-, H^+, OH, …
  - Hot radioactive acid is *nasty*
- Tested many approaches, now very uniform and repeatable



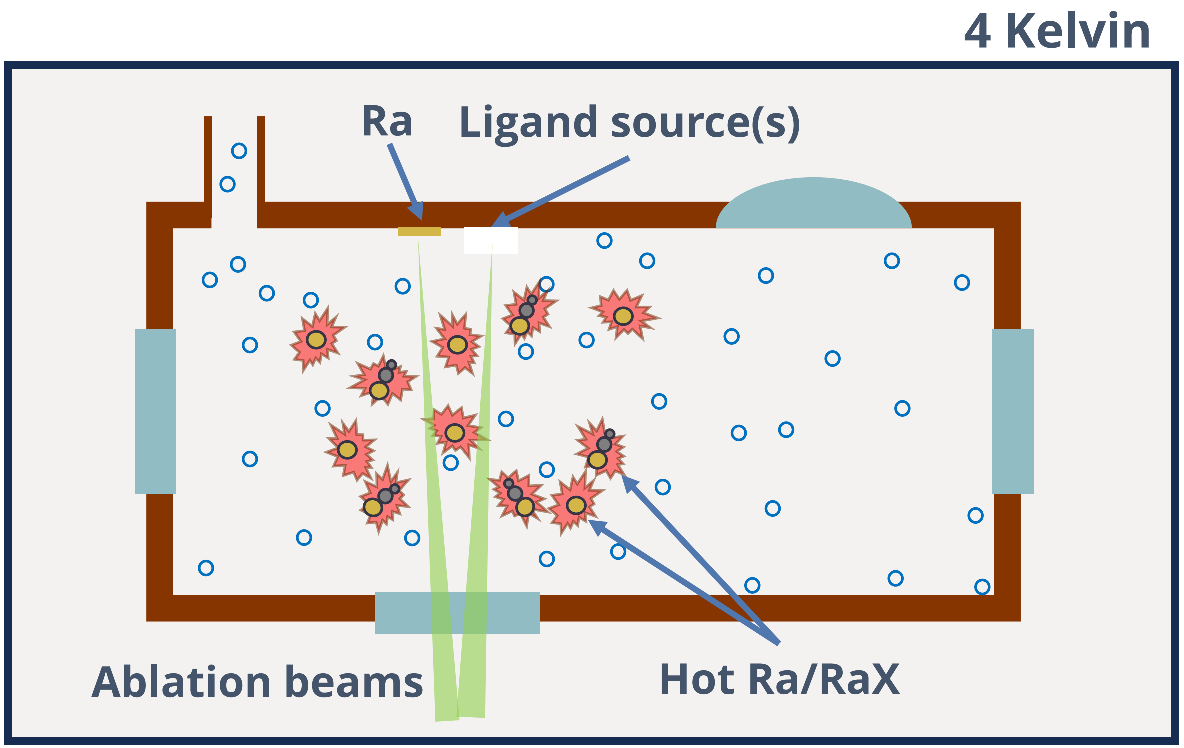
Most recent

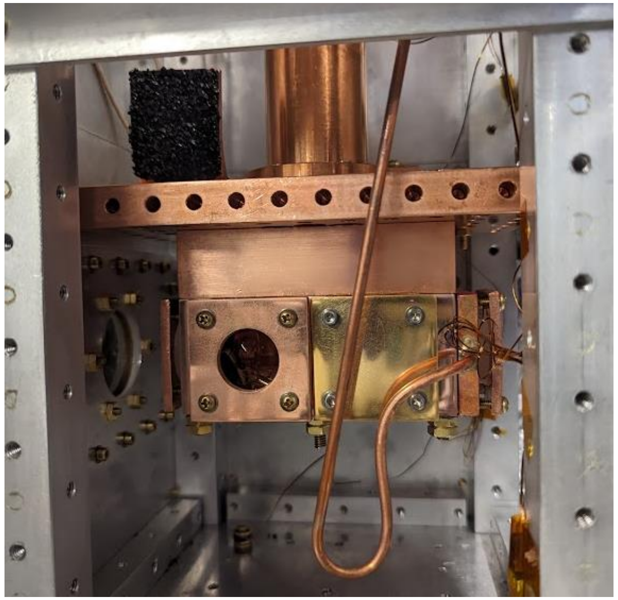
Radium Molecule Apparatus

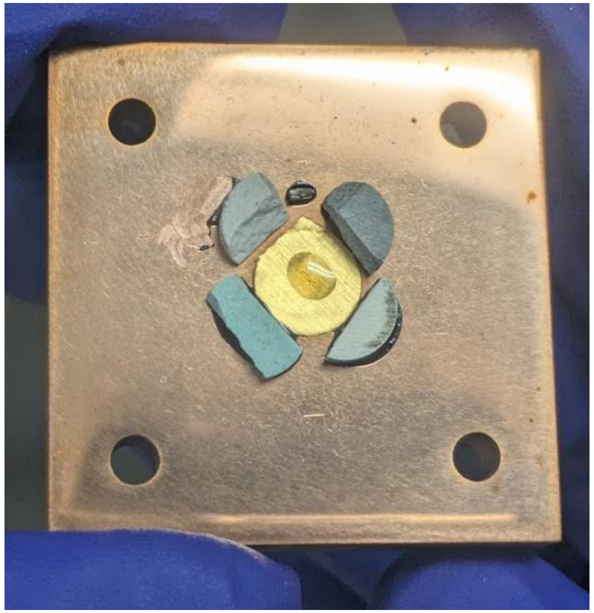
- Tabletop apparatus at Caltech
- 4 K helium cryogenic buffer gas cooling
  - Molecules are cold and stopped
  - Same starting point as many molecular precision measurement and quantum science experiments
- **Approach should be applicable to many other species**



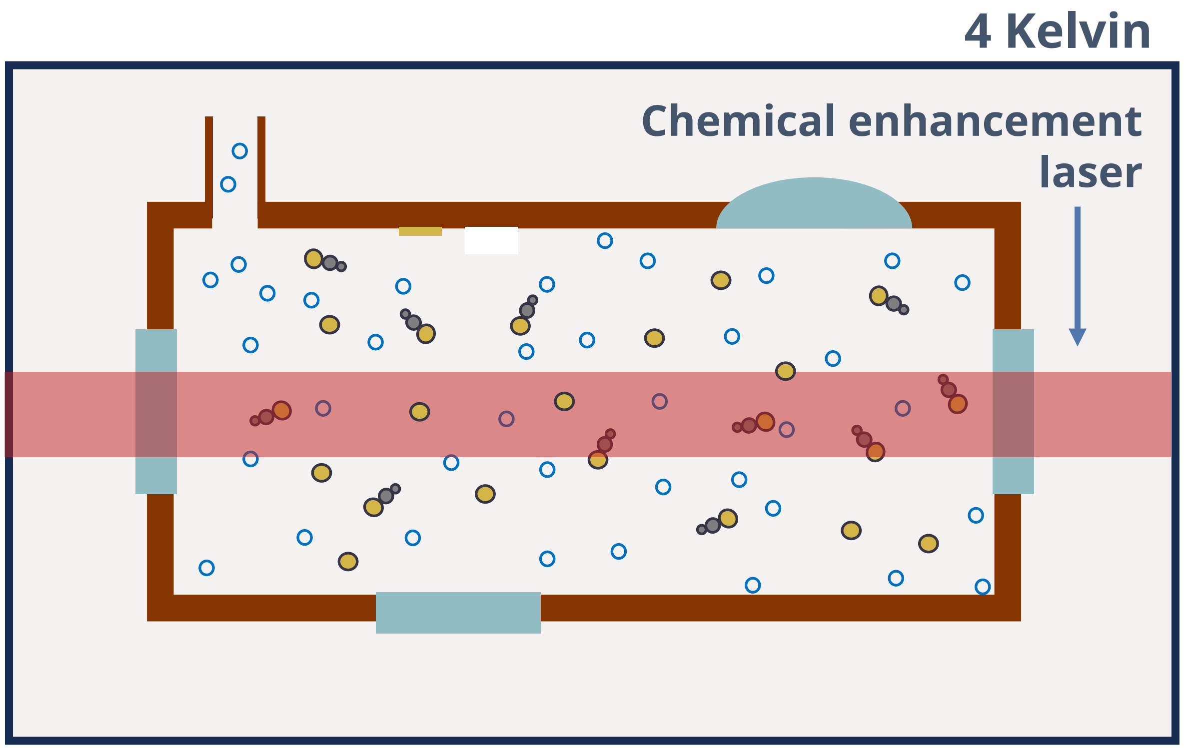
Molecule Production

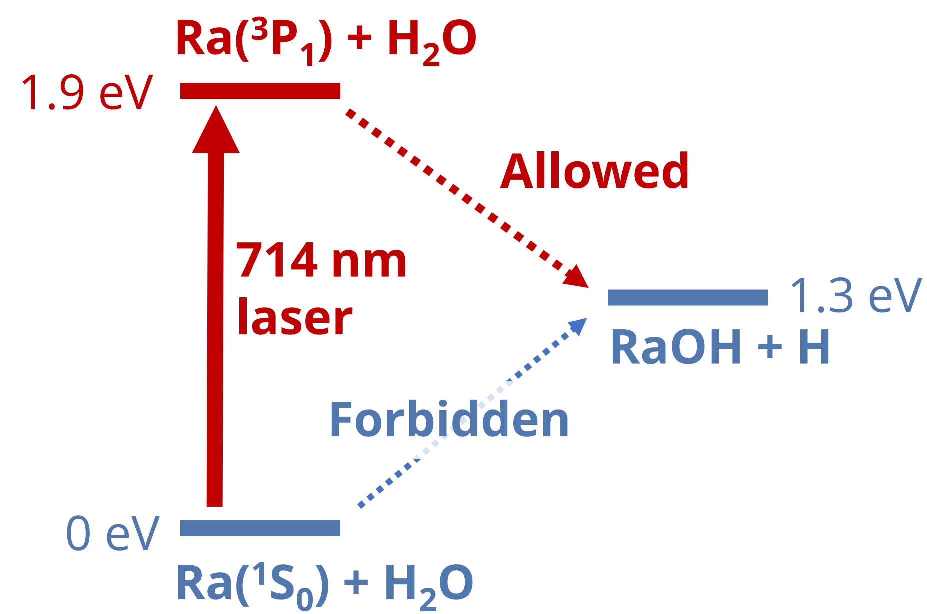




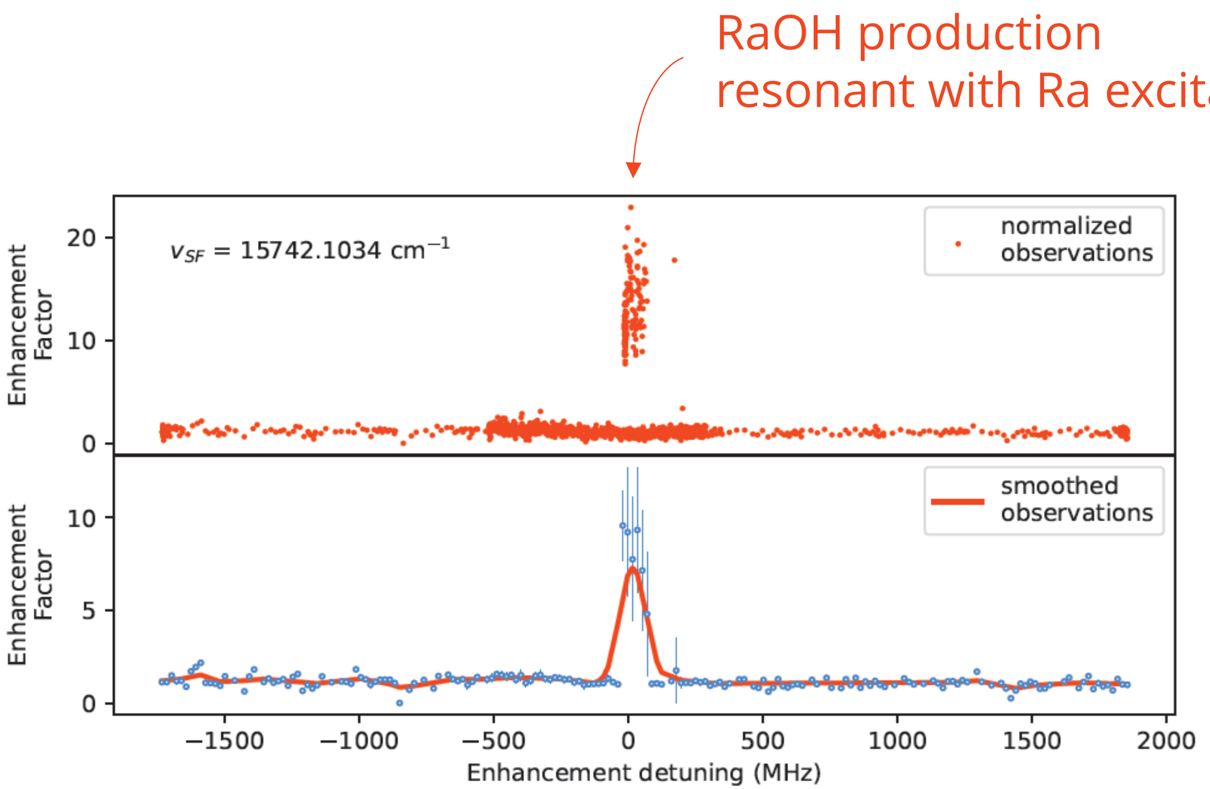


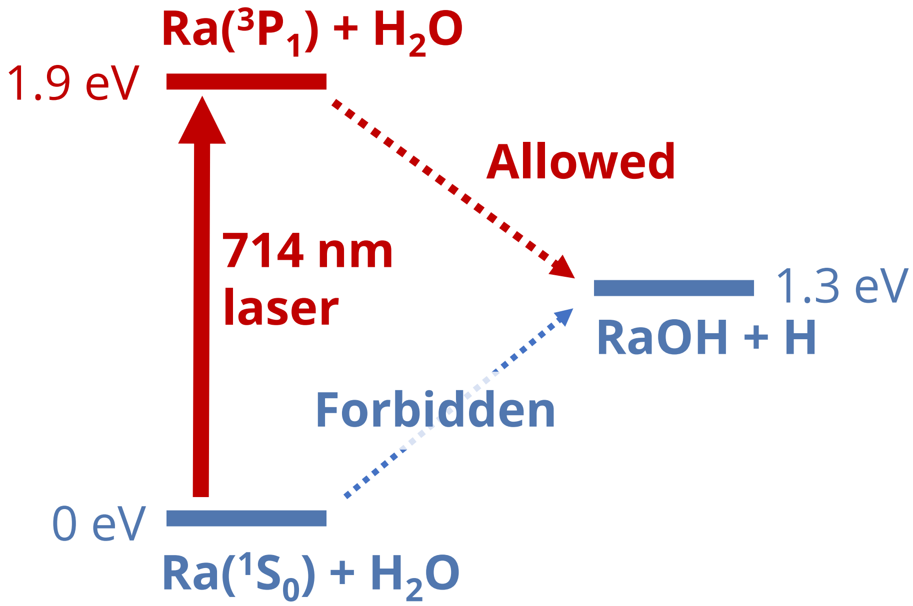
Molecule Production



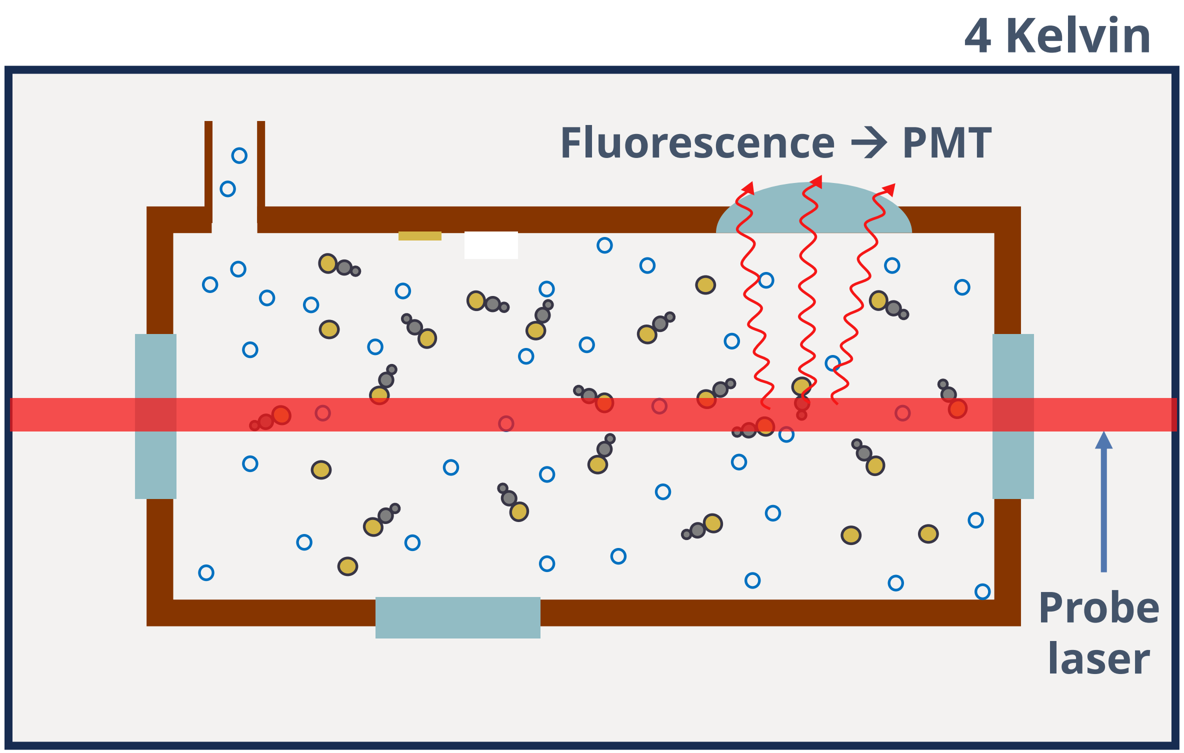


Molecule Production

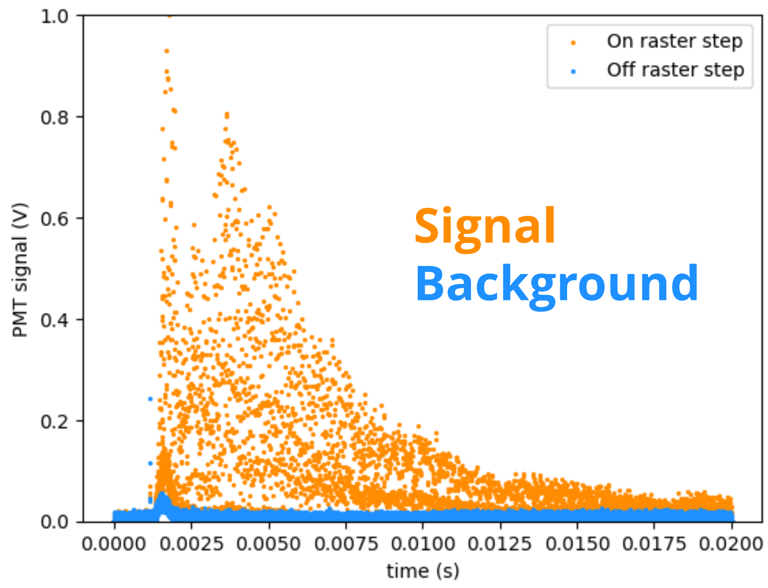




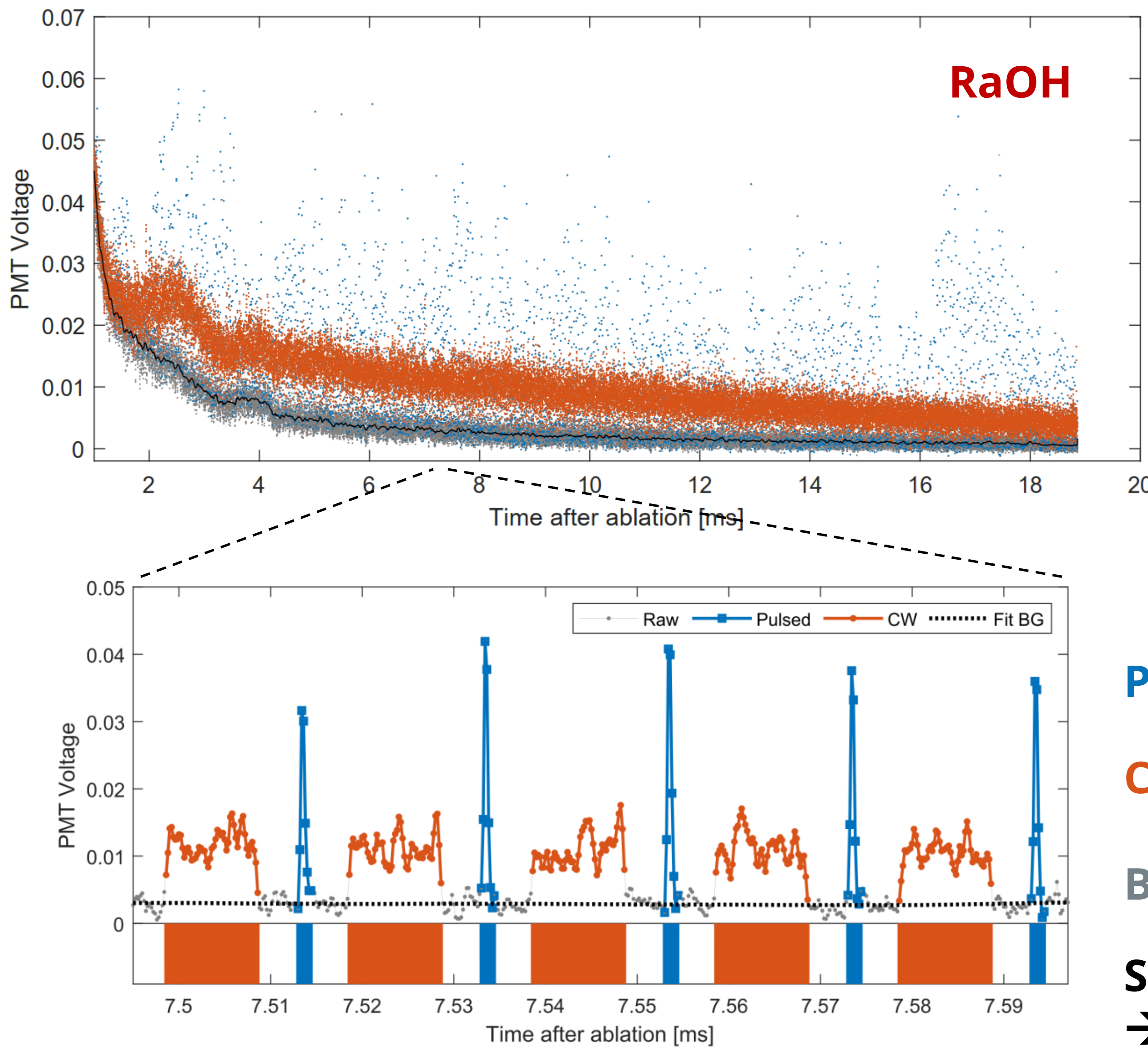
Molecule Detection



Single-shot fluorescence
From 4 K RaF molecules



Single-shot Molecule Detection



Pulsed – broad

Continuous – narrow

Background

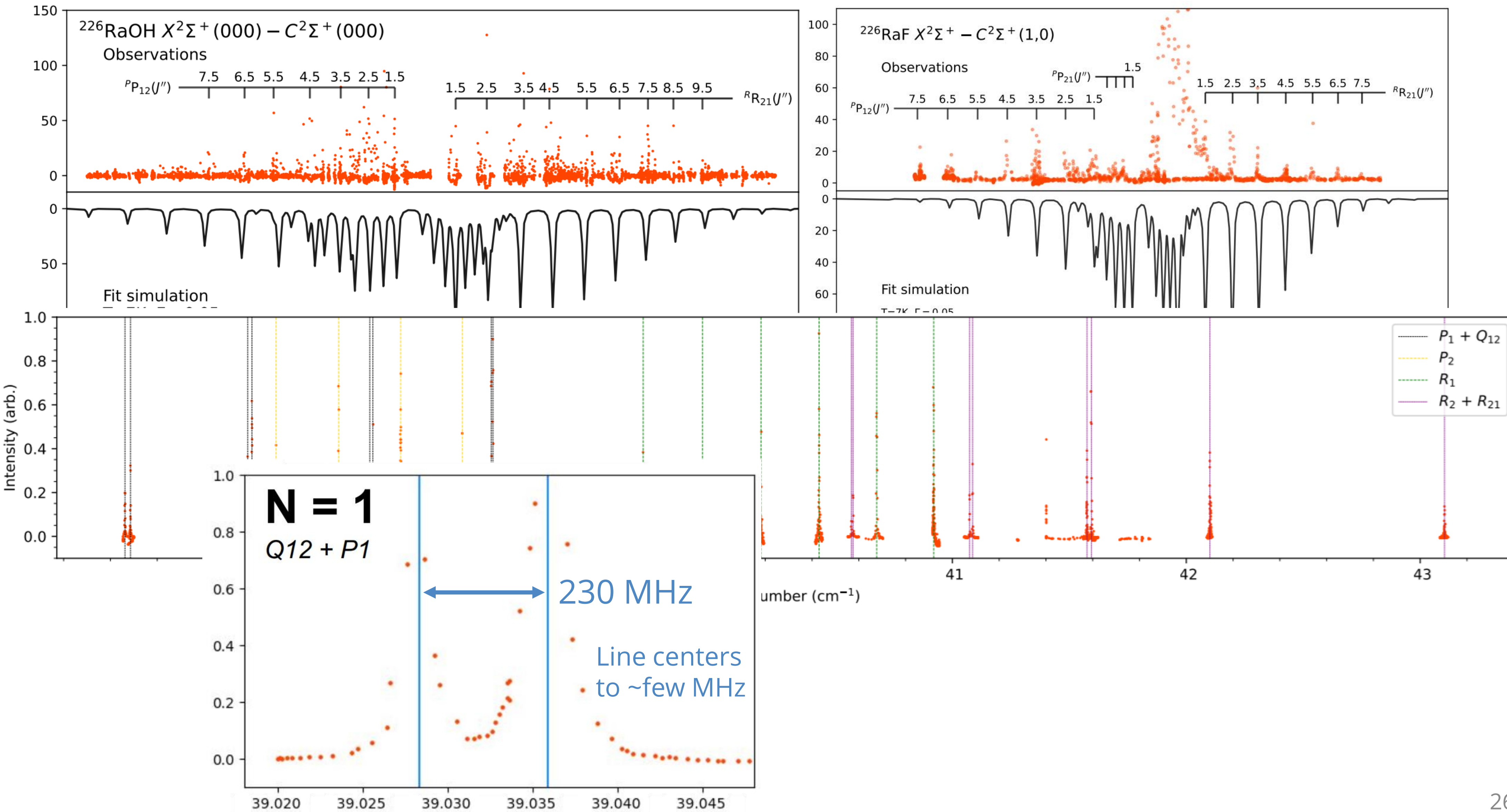
Pulsed – broad

Continuous – narrow

Background

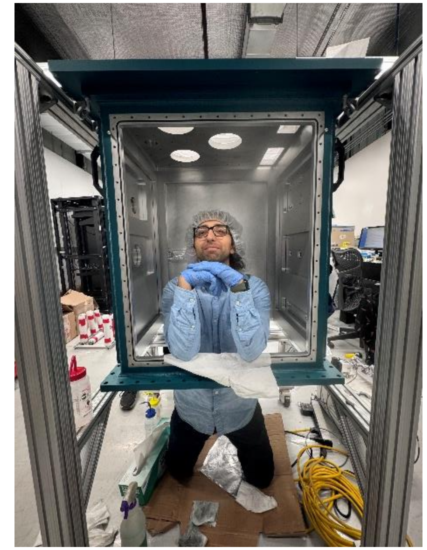
Simultaneous measurement
→ single shot normalization

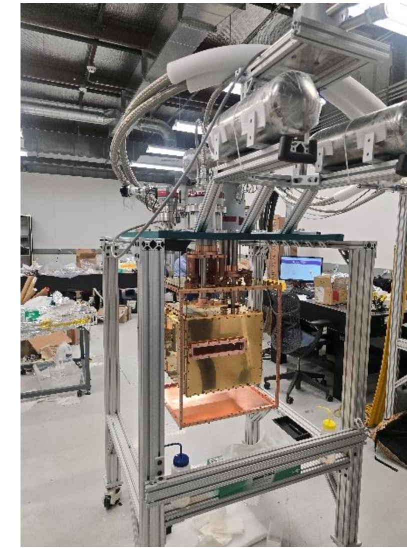
Preliminary Spectra of 4 K RaOH, RaOD, RaF $X^2\Sigma \rightarrow C^2\Sigma$



RaX Collaboration

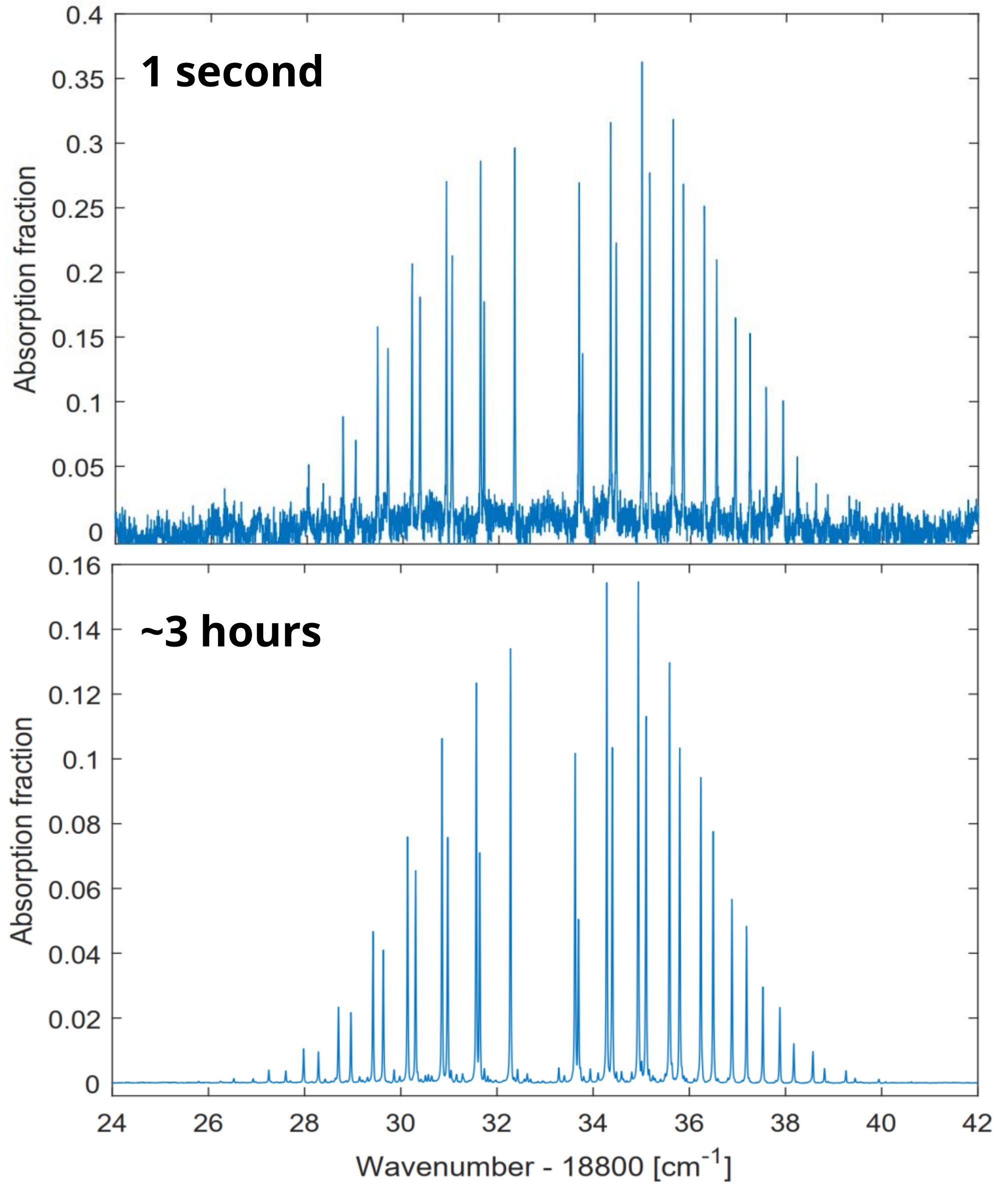
- Cryogenic beam formation for precision measurement is an engineering challenge
- RaX collaboration is on it!
  - John M. Doyle @ Harvard
  - Ronald Garcia Ruiz @ MIT
  - NRH @ Caltech
  - Support from FRIB



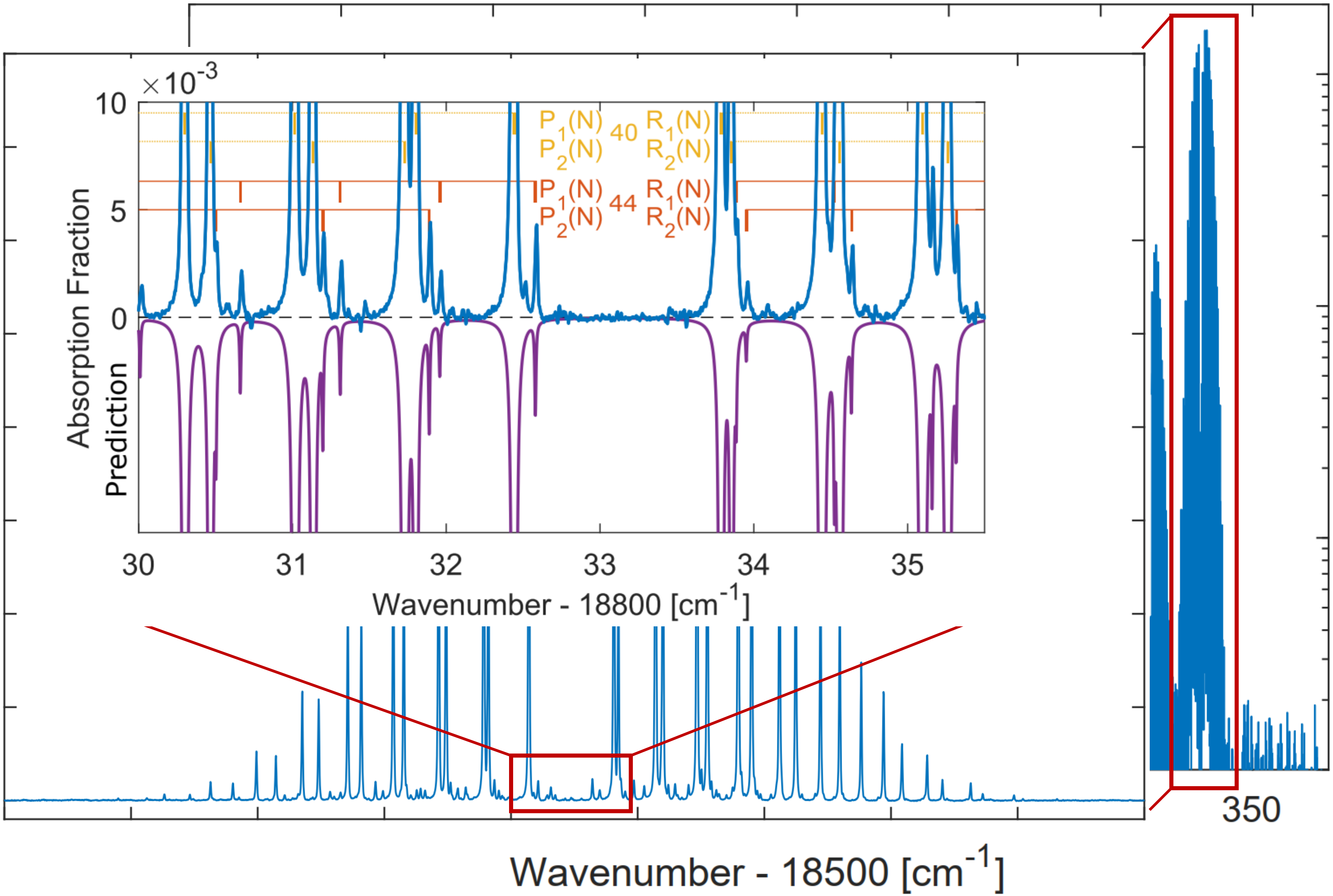


New Spectroscopy Tools

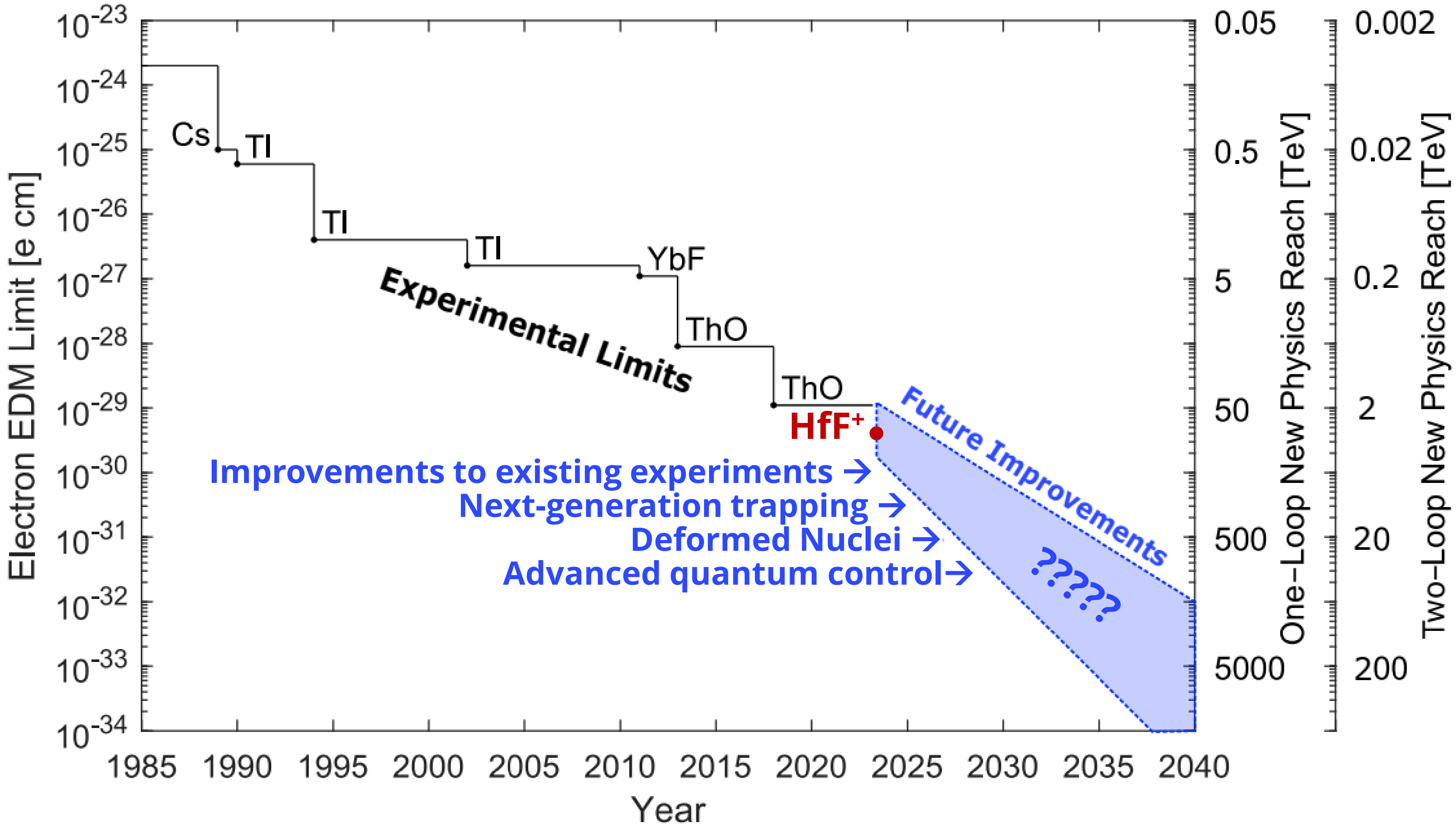
- Spectroscopy is a big barrier for molecule work
  - Typically months → years
- Goal: make it take hours
- Combine buffer gas cooling, absorption spectroscopy, and state of the art spectrometers
- 500 MHz resolution over 15 THz in a single shot
- Prototyped with CaF



A. N. Patel, M. I. Howard, T. C. Steimle, NRH
arXiv:2505.03650 (2025) Submitted


×10^-3
10
5
0
Absorption Fraction
Prediction
P1(N) 40 R1(N)
P2(N) R2(N)
P1(N) 44 R1(N)
P2(N) R2(N)
30
31
32
33
34
35
Wavenumber - 18800 [cm^-1]
350
Wavenumber - 18500 [cm^-1]

Future Improvements

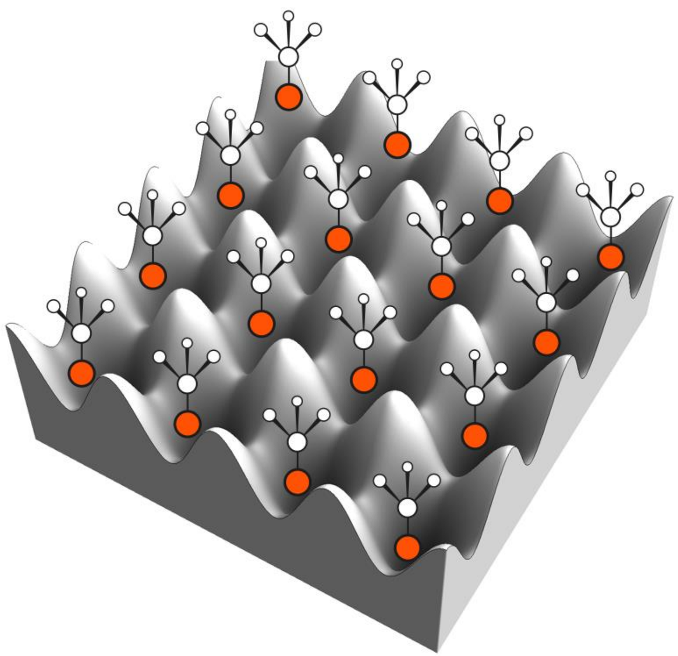


Similar improvements in hadronic CPV are also anticipated

From 2022 Snowmass EDM whitepaper, arXiv:2203:08103 **– Updated**

10^6 molecules
100 s coherence time
Heavy, deformed nucleus
Quantum control
Robust error rejection

~PeV-scale CP-violating physics @ 1 loop
~100 TeV-scale CP-violating physics @ 2 loops
Both leptonic and hadronic sectors
Extreme precision, $\theta_{QCD} \lesssim 10^{-14}$
Near Standard Model CKM value
~10 – 20 year time scales



Future orders-of-magnitude improvements from quantum-enhanced metrology, highly exotic nuclei, …

The pieces are coming together!
With multiple approaches!

Hutzler Lab, Spring 2024
Standing: Adele, Ashay, Chi, Tim, Phelan, Yi, Nick, Harish, Yuiki, Zitian
Sitting: Elizabeth, Madison, Chandler, Nachiket
Hovering: Anya, Yuxi

Collaborators

PolyEDM: **John M. Doyle (Harvard), Tim Steimle (ASU), Amar Vutha (Toronto)**

Molecular Theory: **Anastasia Borschevsky (Groningen), Bill Goddard (Caltech), Jacek Kłos (UMD), Svetlana Kotochigova (Temple)**

SLAM!: **Lan Cheng (JHU), Alyssa Gaiser (MSU), Ronald Garcia Ruiz (MIT), Andrew Jayich (UCSB), Jaideep Singh (MSU/FRIB)**

PolyEDM

polyedm.com

SLAM!

slamcommunity.com

NSF

ALFRED P. SLOAN FOUNDATION

GORDON AND BETTY MOORE FOUNDATION

HEISING-SIMONS FOUNDATION

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The Rose Hills FOUNDATION

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