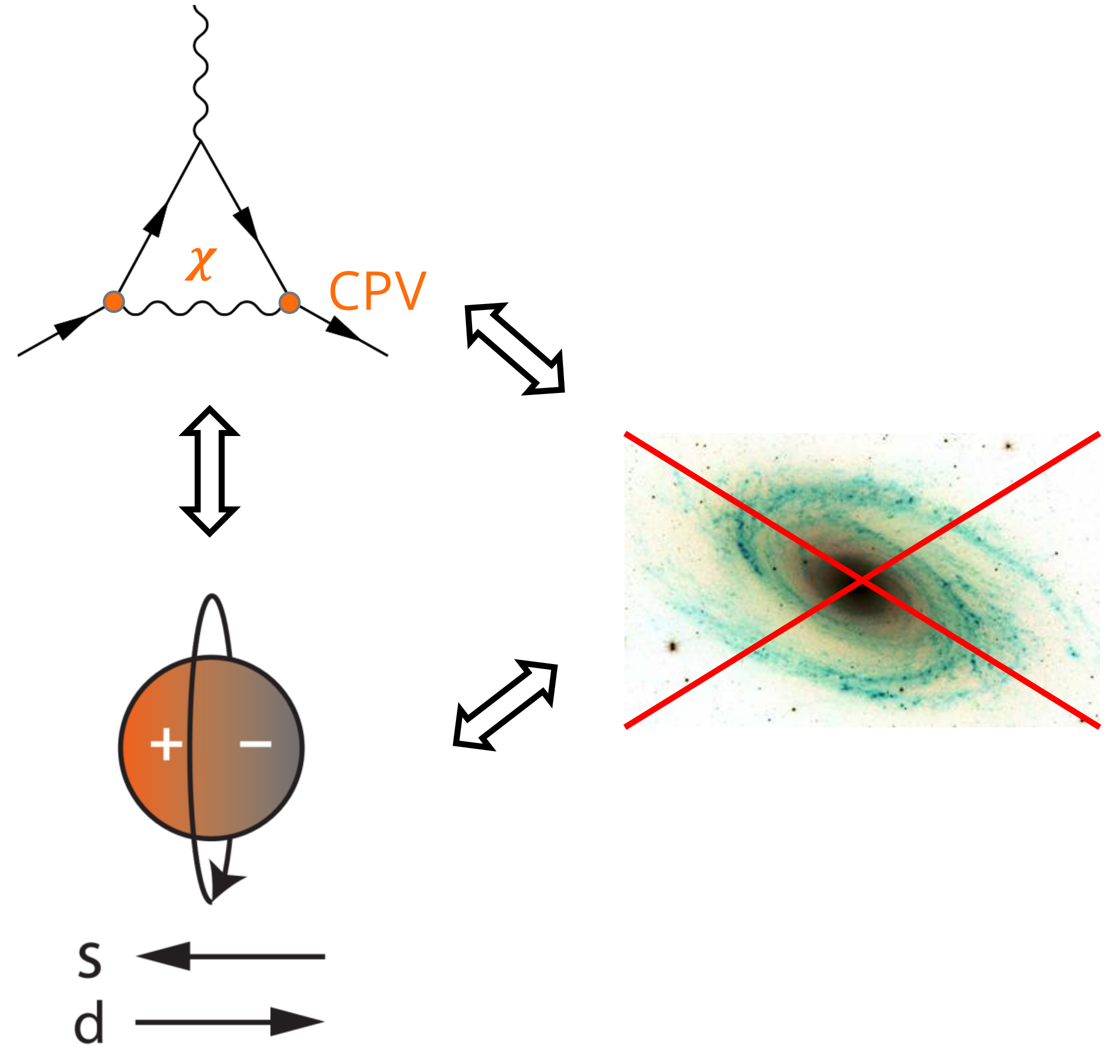
Caltech

Cold Radioactive Molecules for Fundamental Physics

Nick Hutzler
Caltech

Low Energy CPV Observables

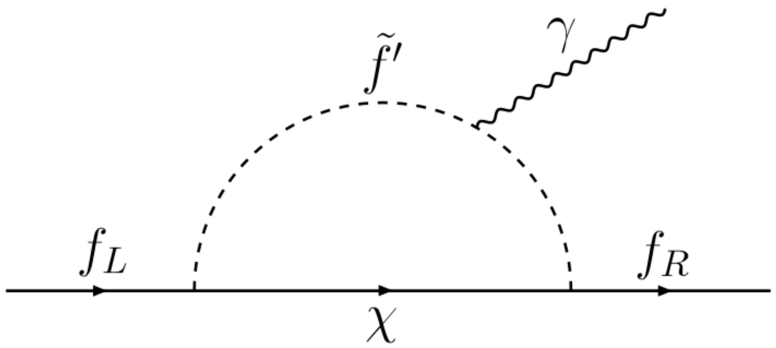
- The baryon asymmetry suggests new CP-violating physics
- Leads to CP-violating electromagnetic moments of regular matter
  - EDMs, CP-violating nuclear moments (“EDMs”)
- Enables sensitive probes of new physics



Energy Reach

- EDMs are generically sensitive to CPV sources
- Electron EDM:
  - HfF⁺, $d_e < 4.1 \times 10^{-30}\ e\ cm$
  - One loop ~ 70 TeV
  - Two loop ~ 3 TeV
  - "Background free"
- Nuclear symmetry violation probes similar energy scales in hadronic sector
  - Quark EDMs, strong force, nuclear forces, …
- Complex parameter space *requires* multiple experiments in multiple systems!

$$d_f \sim e q_f \sin(\delta_{CPV}) \left(\frac{g^2}{16\,\pi^2}\right)^{\ell} \frac{m_f}{M_{NP}^2}$$

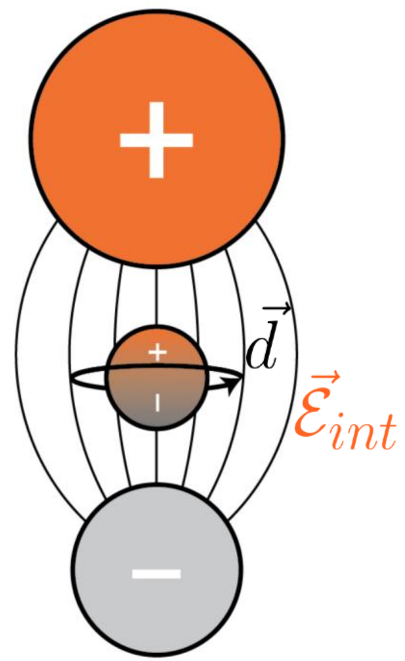


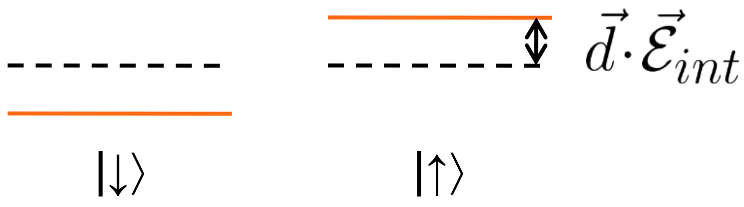
Wilson coefficient	Operator (dimension)	Number
$\bar{\theta}$	Theta term (4)	1
δ_e	Electron EDM (6)	1
$\mathrm{Im}\, C_{\ell equ}^{(1,3)}$, $\mathrm{Im}\, C_{\ell eqd}$	Semi-leptonic (6)	3
δ_q	Quark EDM (6)	2
$\tilde{\delta}_q$	Quark chromo EDM (6)	2
$C_{\tilde{G}}$	Three-gluon (6)	1
$\mathrm{Im}\, C_{quqd}^{(1,8)}$	Four-quark (6)	2
$\mathrm{Im}\, C_{\varphi ud}$	Induced four-quark (6)	1
Total		13

Snowmass EDM white paper, arXiv:2203.08103
J. Engel, M. J. Ramsey-Musolf, and U. van Kolck, Prog. Part. Nucl. Phys. 71, 21 (2013)

Molecular CPV Sensitivity

- Molecules contain large fields
  - Amplify electron, nuclear CPV
  - ~10^3 as large as atoms
  - ~10^6 as large as lab fields
  - “Molecular Enhancement”
- Measure with spin precession
  - Conceptually similar to nEDM
- So far only eEDM
  - Nuclear CPV requires nuclear spin → complicates structure
- Many developments underway
  - Improvements to existing experiments
  - New eEDM and nuclear CPV experiments
  - Major sensitivity increases coming

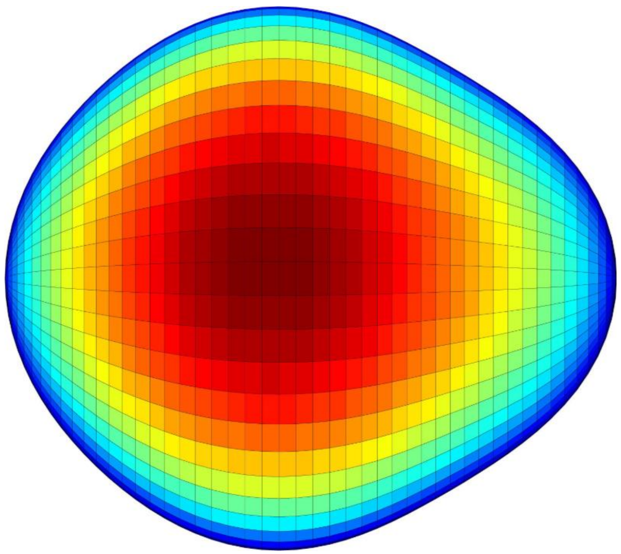




$$|\uparrow\rangle + |\downarrow\rangle \rightarrow |\uparrow\rangle + e^{i\phi}|\downarrow\rangle$$

Paths to Further Improvement

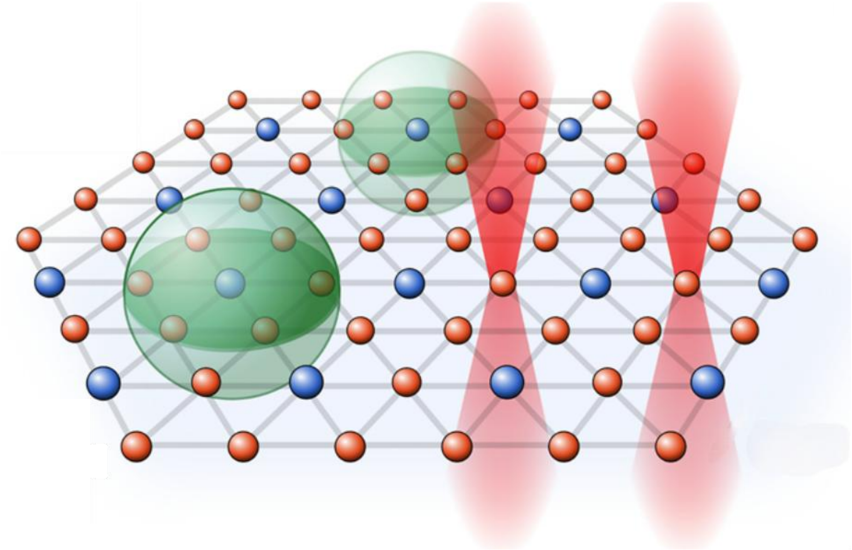
Exotic Nuclei



Use the heaviest, most sensitive nuclei to amplify CPV effects

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

Modern Quantum Tools



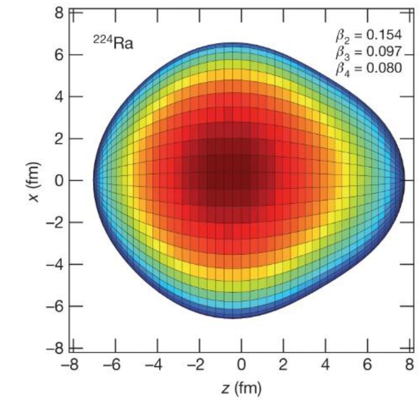
Apply methods from quantum science to increase resolution

AVS Quantum Sci. 3, 023501 (2021)

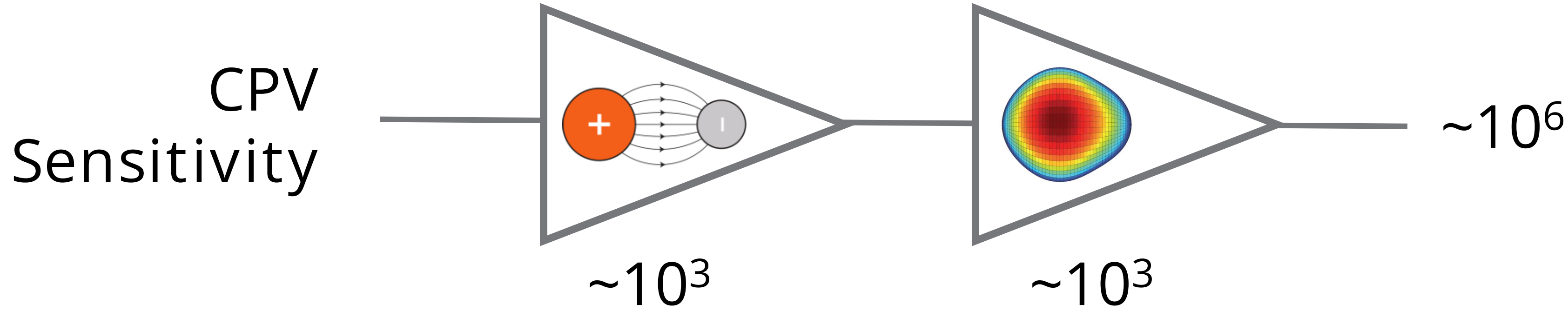
Exotic Nuclei

Exotic Nuclei

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

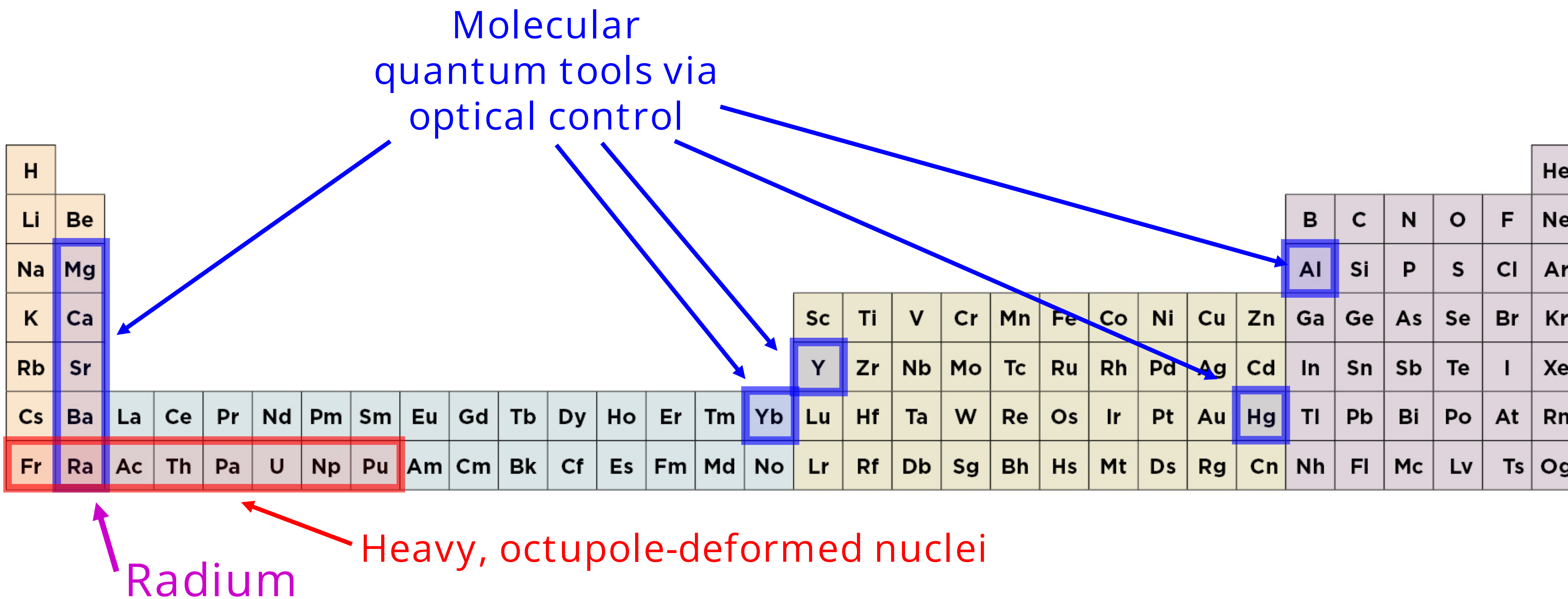


L. P. Gaffney et al., Nature 497, 199 (2013)



- Control *one* molecule at a time → hadronic frontier
- Challenge: radioactive, rare, hard to work with

Which Species?

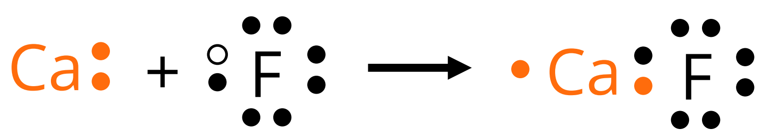


Motivated RaF spectroscopy at CERN ISOLDE, e.g. R. F. Garcia Ruiz *et al.*, Nature (2020)

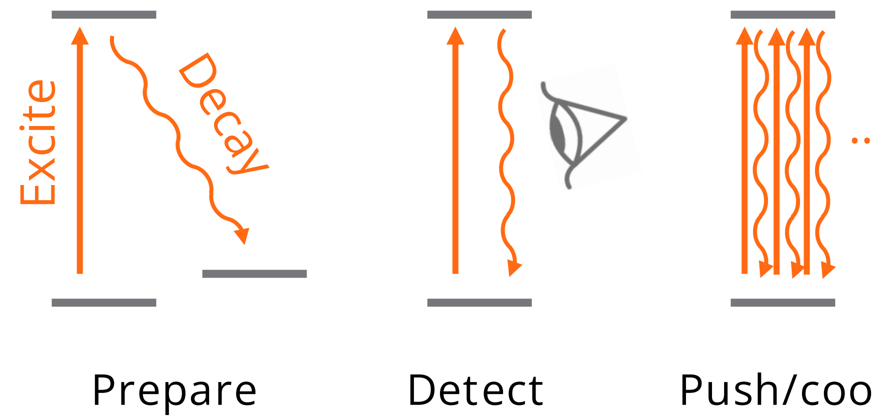
Very exciting work going on with many other species as well!

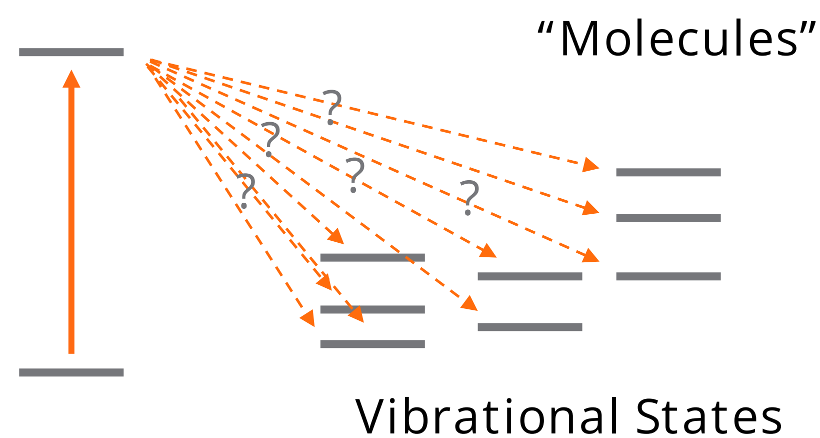
Molecular Quantum Tools

- Atomic quantum science relies heavily on optical control
  - Scatter ("cycle") photons for state preparation/readout, cooling, trapping
- Internal complexity of molecules makes this challenging
  - Unwanted vibrational excitation
- Some molecules offer optical control
  - Single unpaired *s* electron (Ca, Sr, Ba, Yb, Ra) + (F, OH, NH_2, …)



- Radium molecules have this!
- Major advances in last decade: laser cooling, trapping, entanglement, quantum engineering, …

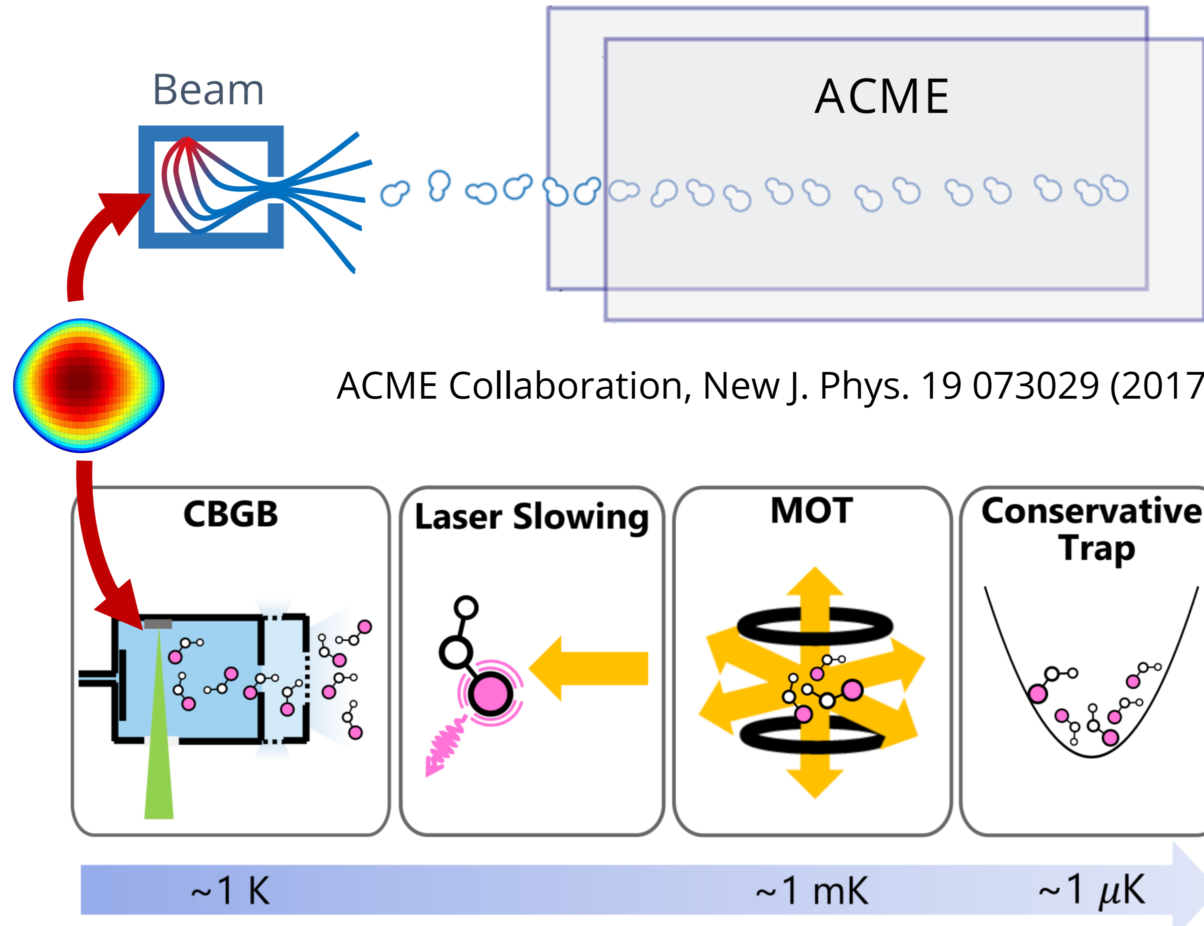




Review: B. L. Augenbraun *et al.*, Adv. At. Mol. Opt. Phys. 72, 89 (2023)

Precision Molecular Tools with Exotic Nuclei

- Molecular precision measurements start at ≲100 K, low velocities
- Introduce exotic nuclei directly into molecular precision measurement
  - Cryogenic Buffer Gas Cooling
  - Can use for beams, laser cooling, ion trapping, …
  - Applicable to essentially any species

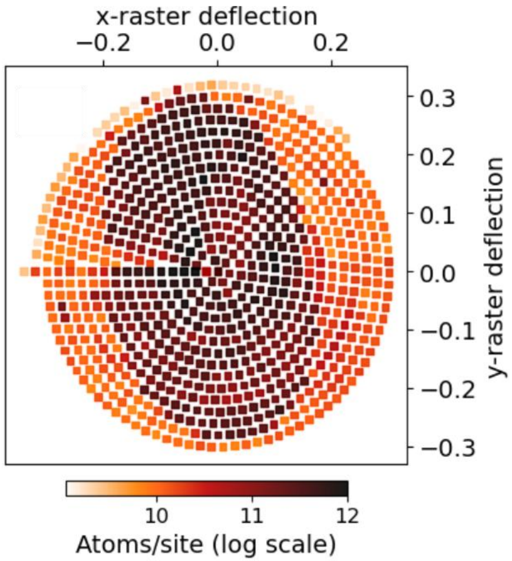


ACME Collaboration, New J. Phys. 19 073029 (2017)

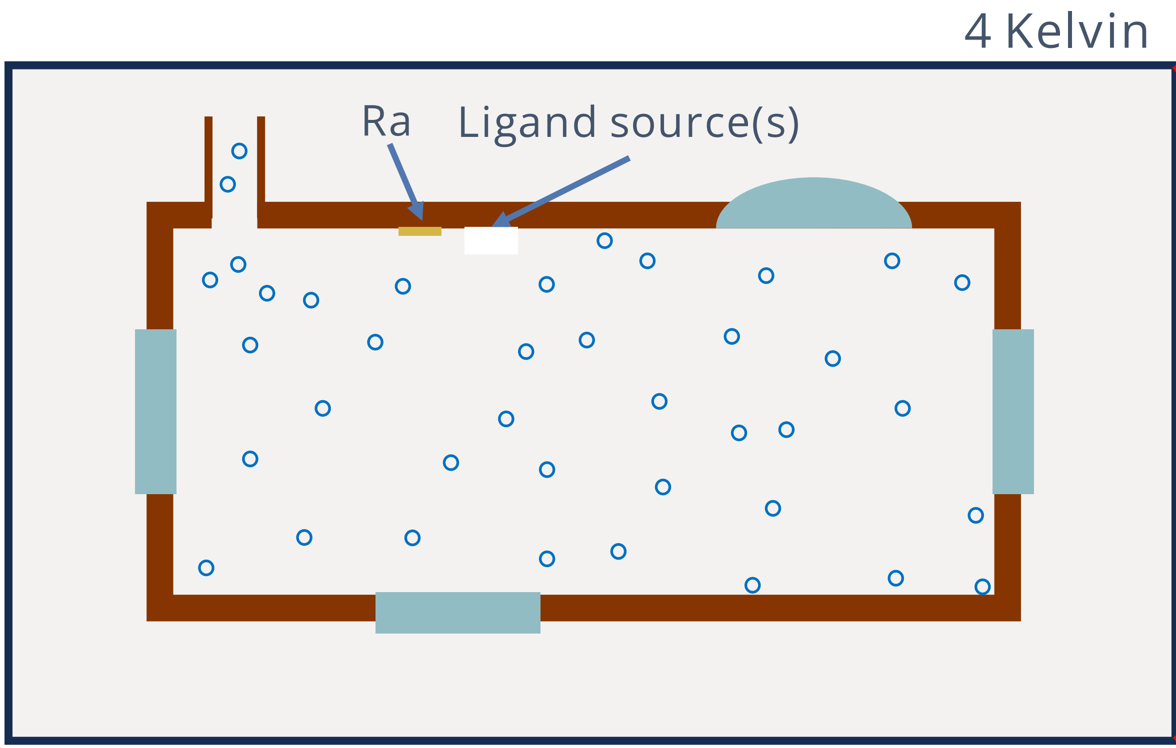
B. L. Augenbraun *et al.*, Adv. At. Mol. Opt. Phys. 72, 89 (2023)

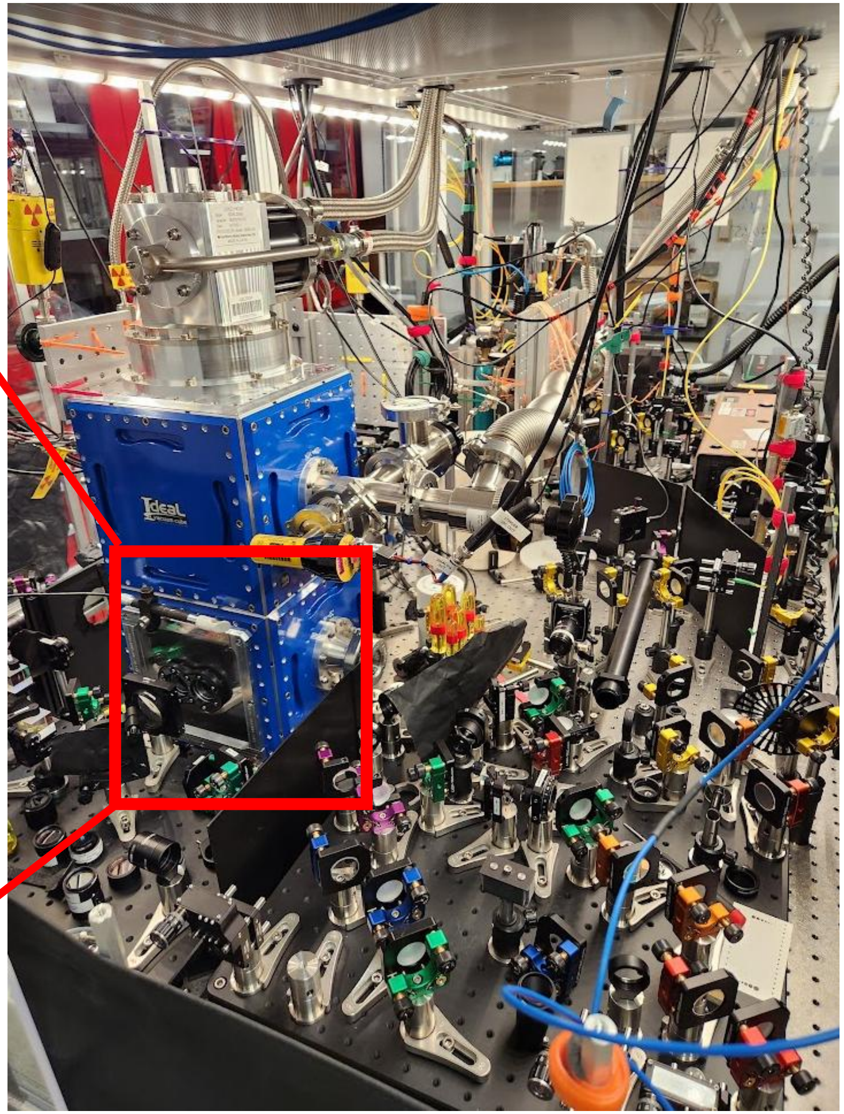
Radium Ablation Targets

- Start: 1 mg Ra-226 nitrate salt
  - 1 mg = 1mCi = 37 MBq
  - US Department of Energy (DOE) National Isotope Development Center (NIDC) and Oak Ridge National Laboratory
- Divided into ~50 μg quantities
  - Thanks Alyssa Gaiser, MSU/FRIB
- Deposit radium solution
  - Contains xylitol to form uniform, repeatable, stable matrix at 4 K
  - Ablate to make cold Ra atoms
  - $\mathcal{O}(1)$ material conversion

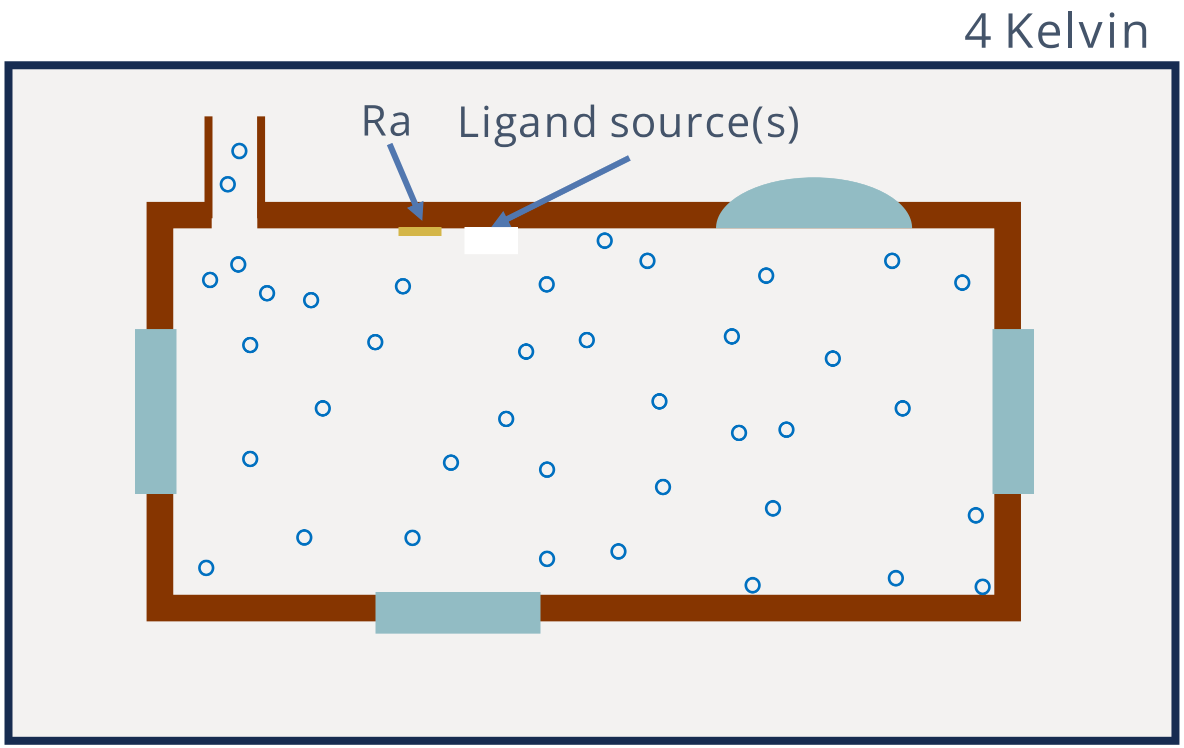


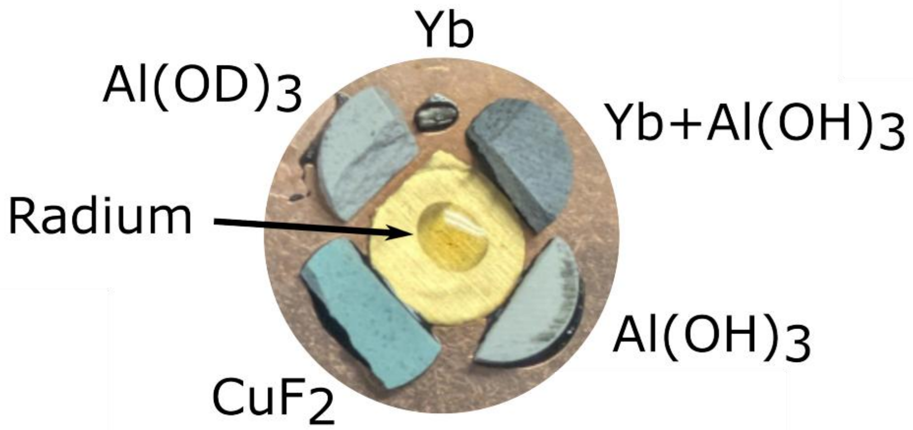
Molecule Production



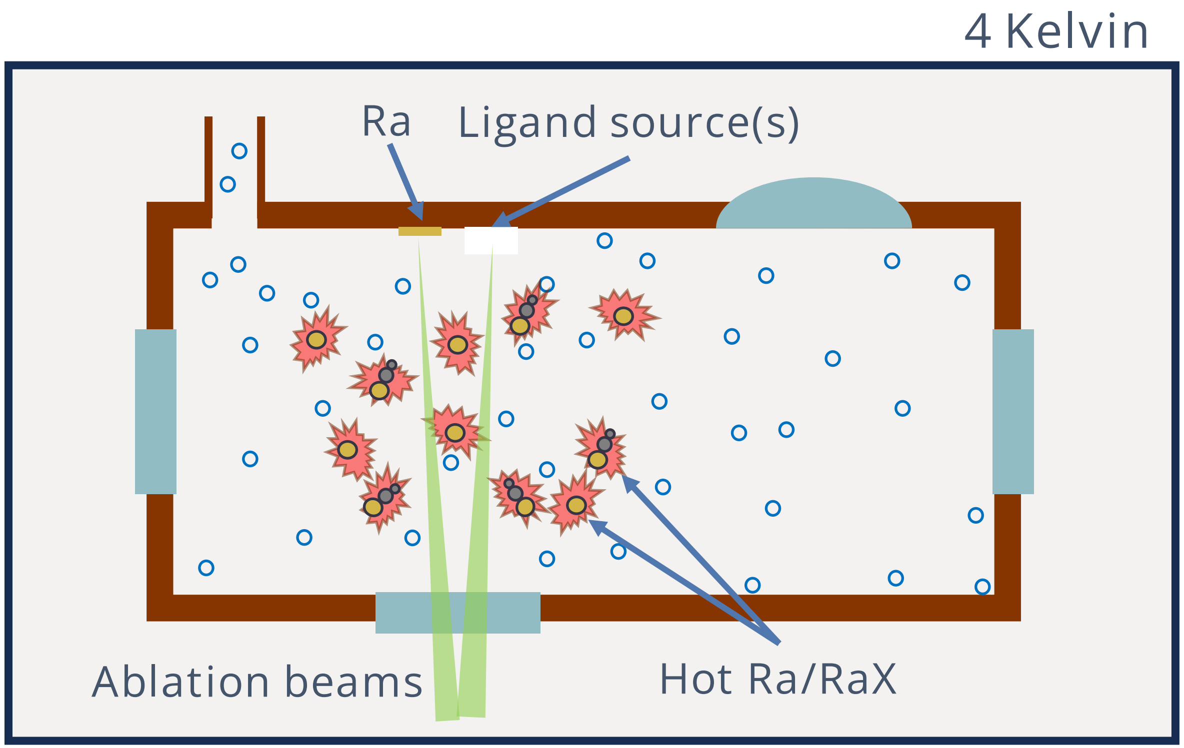


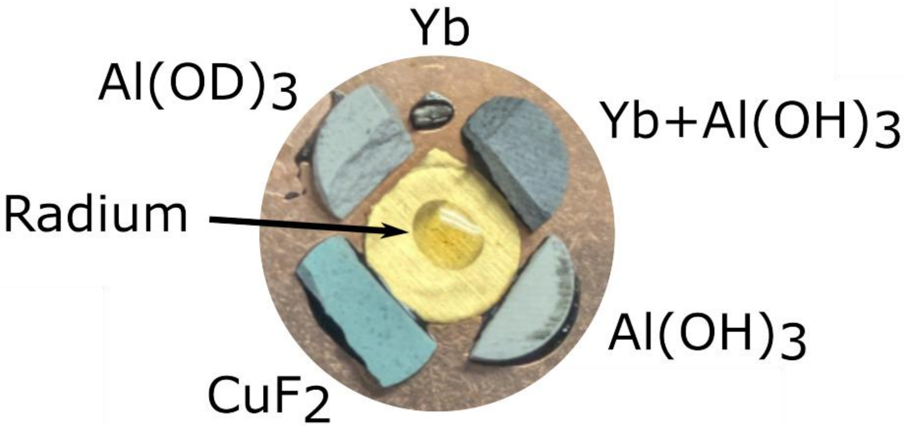
Molecule Production



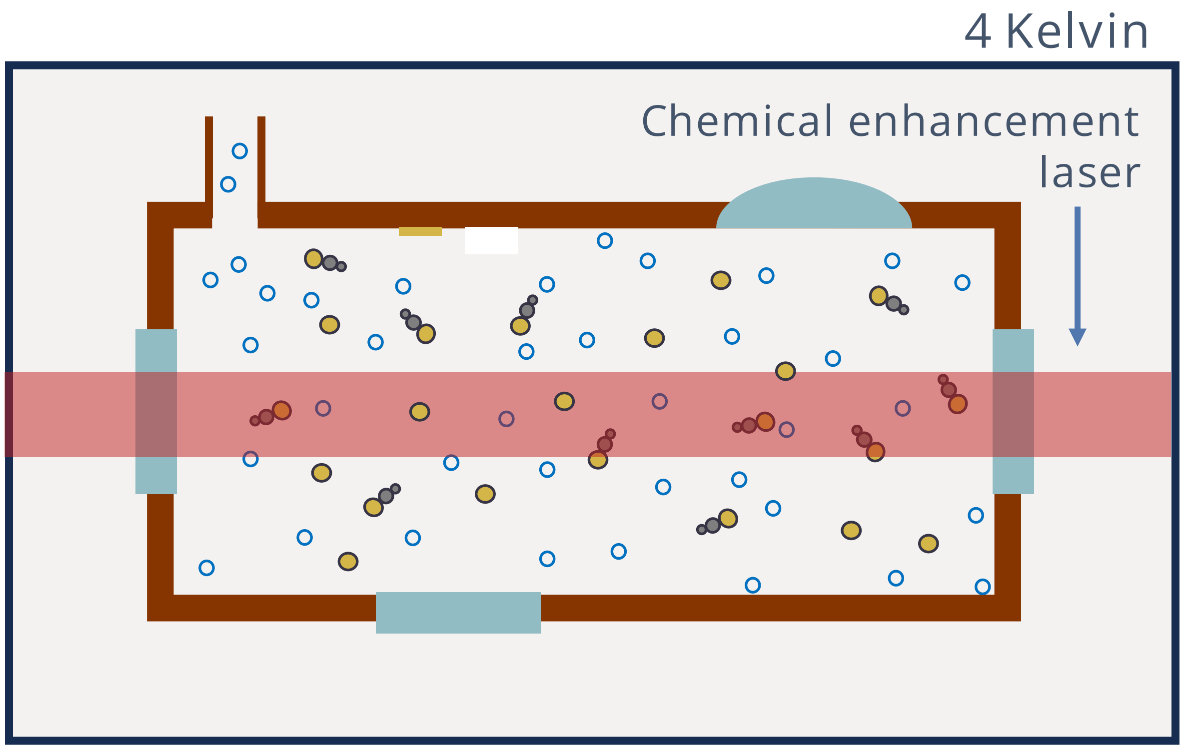


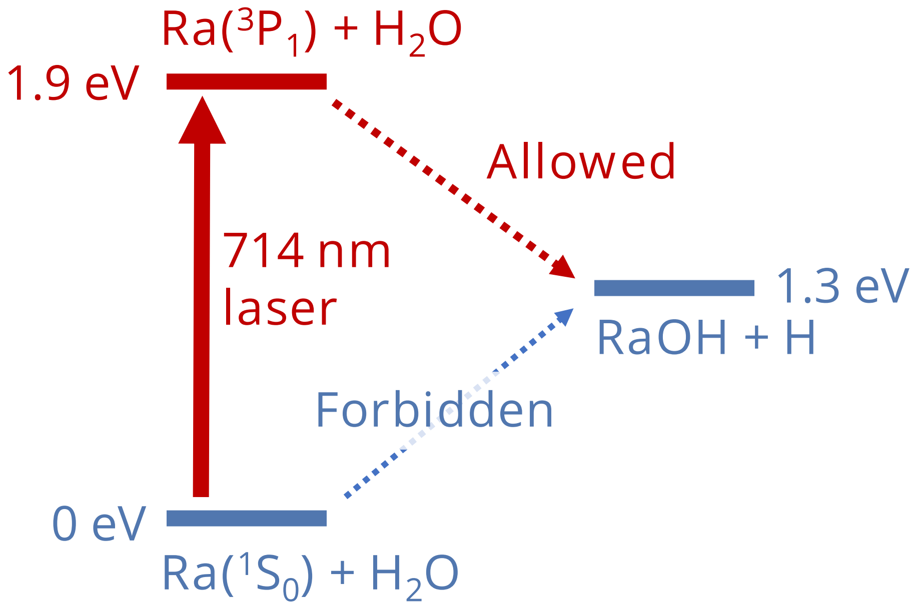
Molecule Production



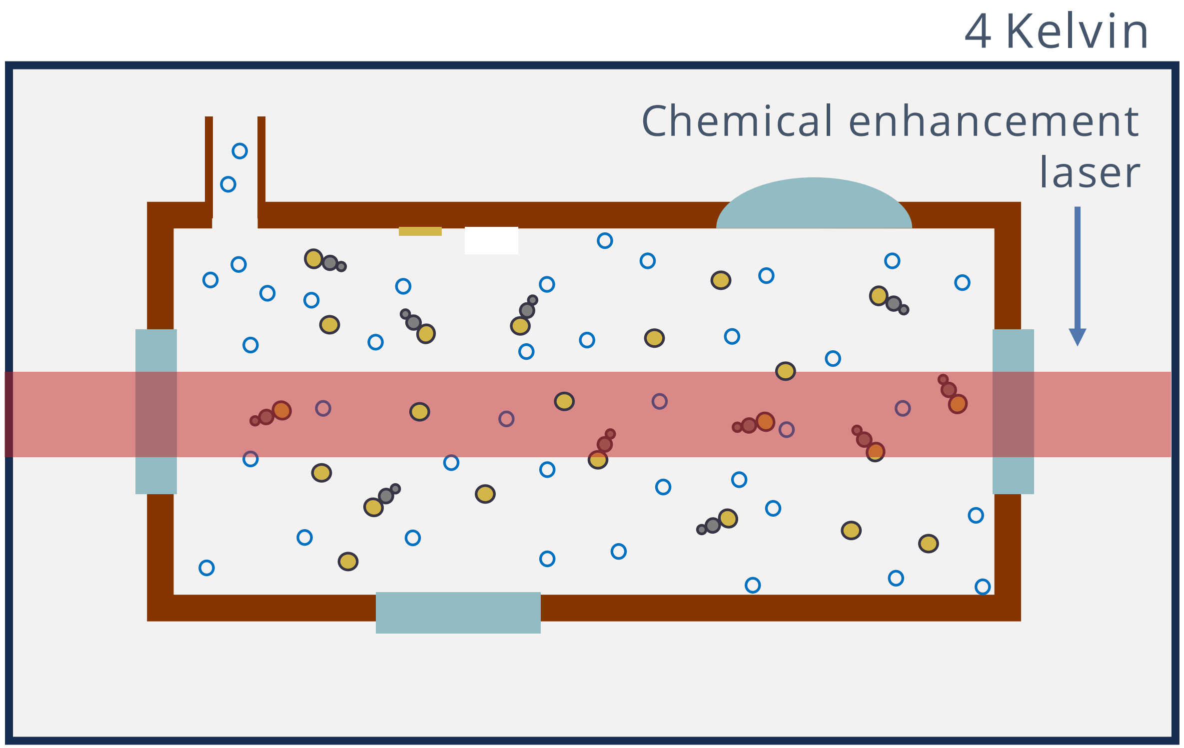


Molecule Production

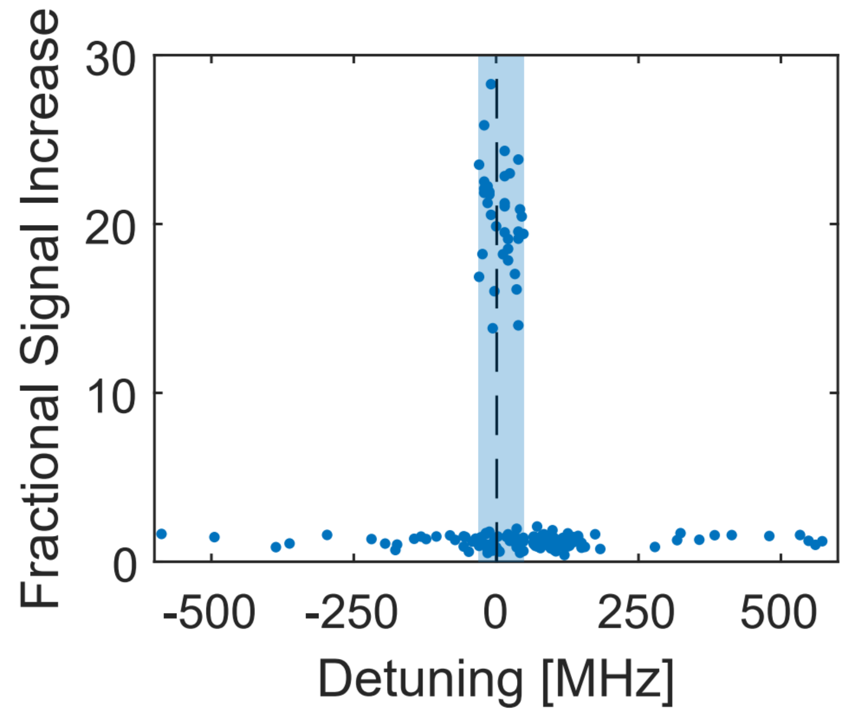




Molecule Production

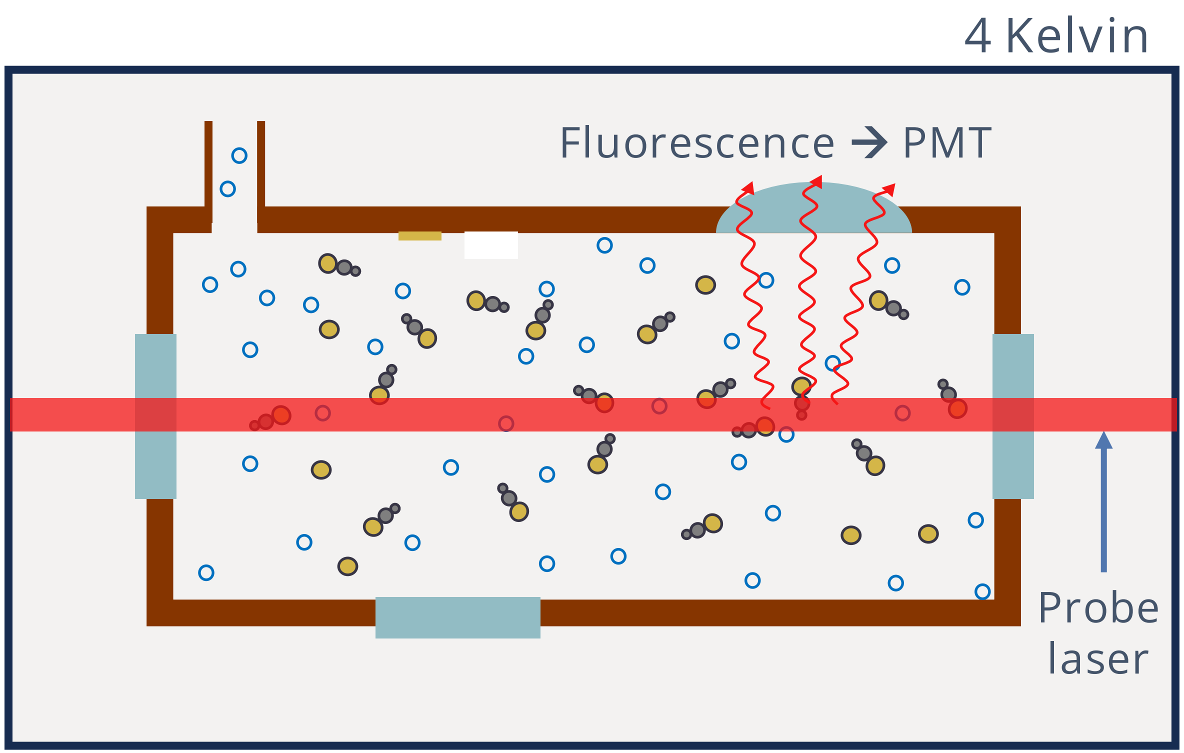


Resonant laser-driven production
Insensitive to contamination
No need to isolate, purify, etc.

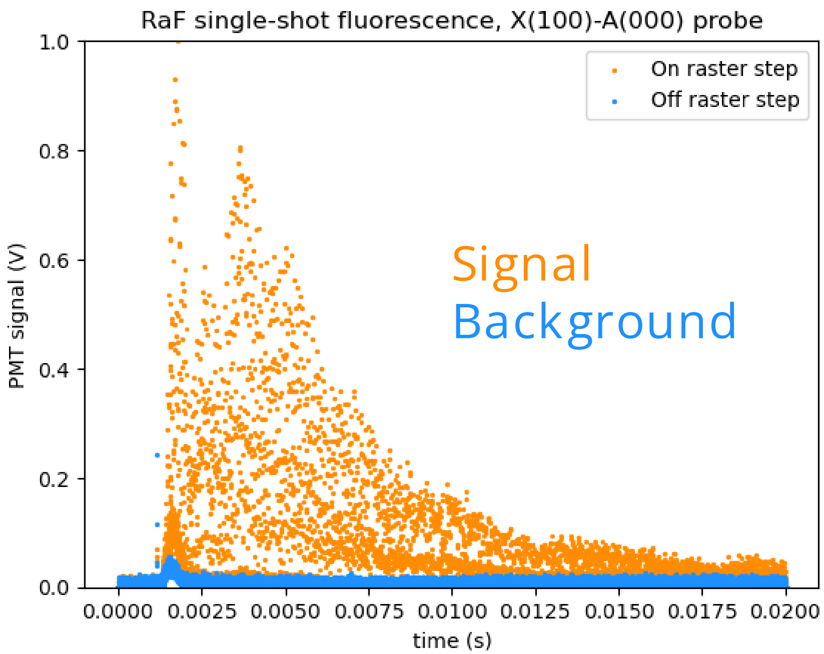


From Ra $^1S_0 \rightarrow {}^3P_1$

Molecule Detection

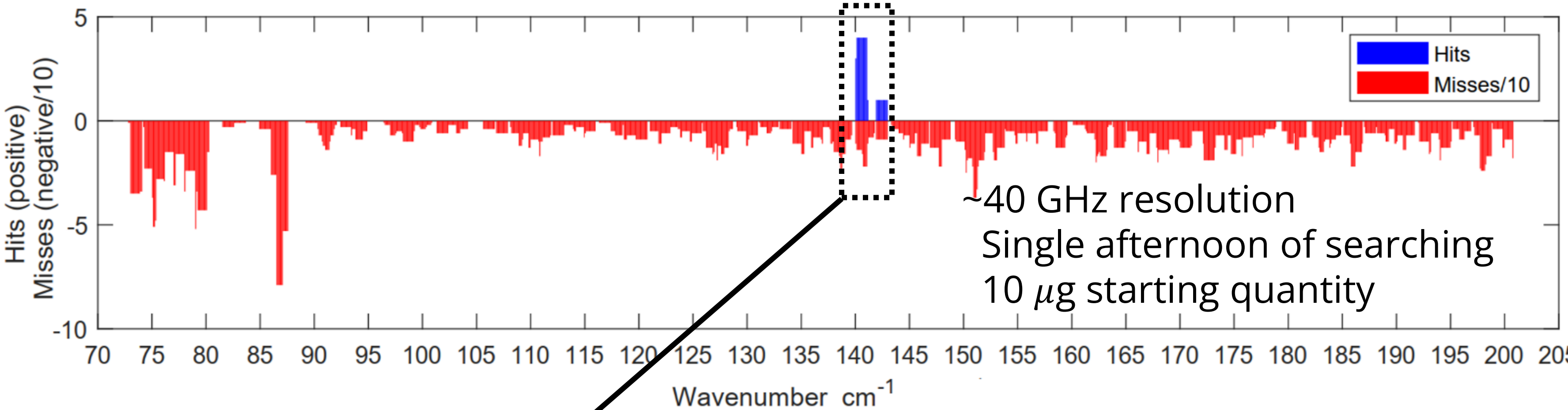


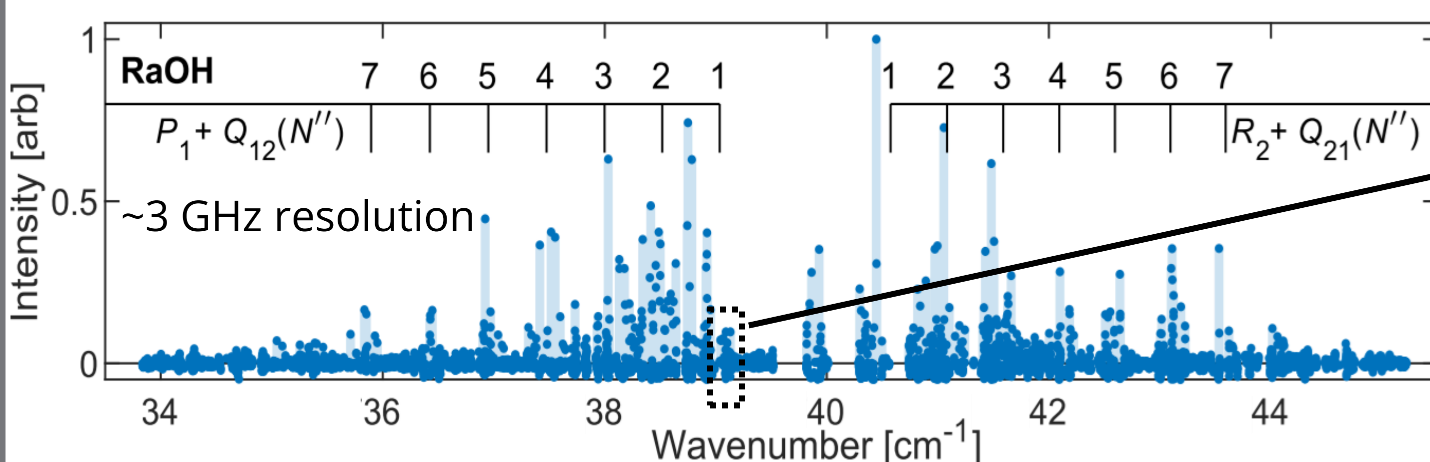
Single-shot fluorescence
From 4 K RaF molecules

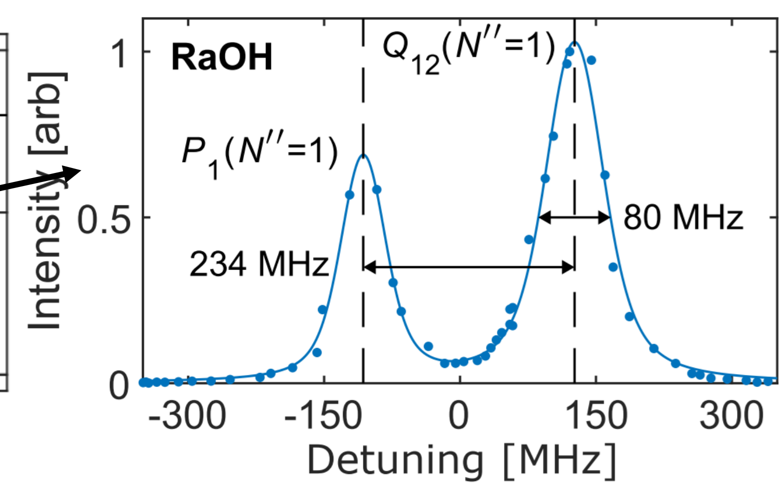


Cold RaOH Spectroscopic Search

Initial theoretical uncertainty ~10 THz, Lan Cheng @ Johns Hopkins



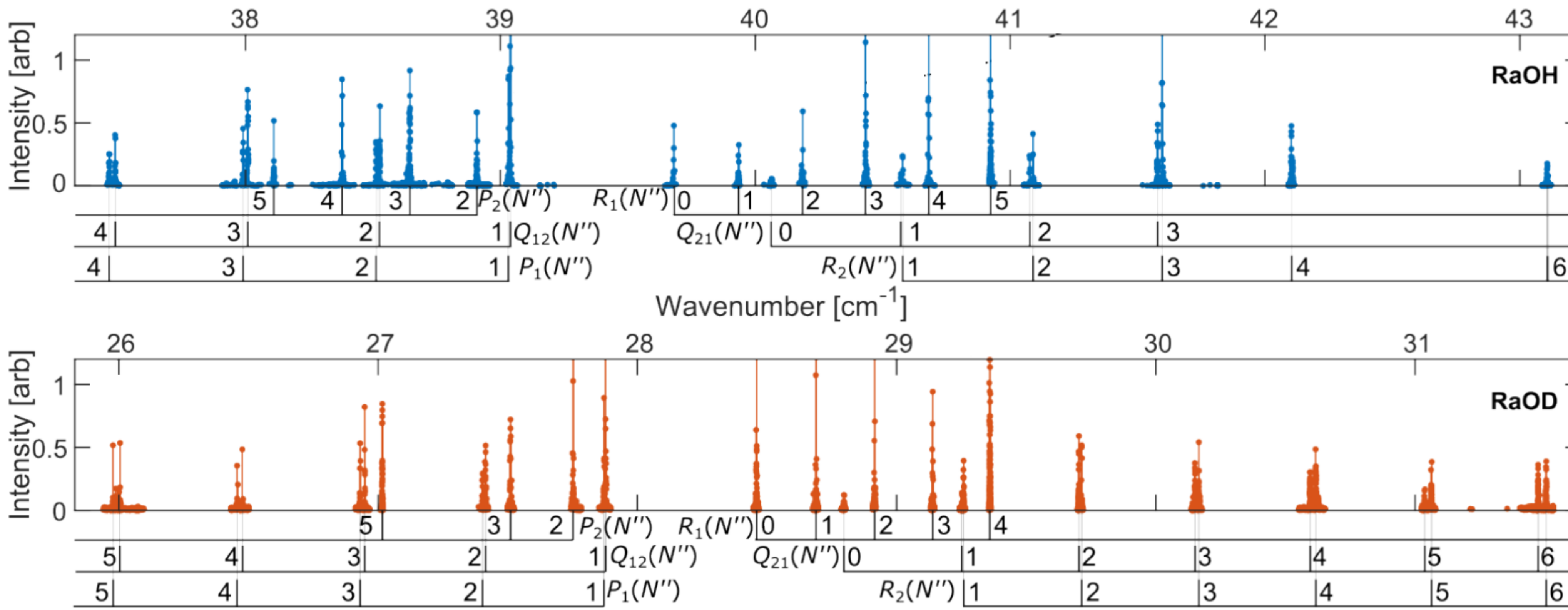




Single-frequency, Doppler limited

C. J. Conn,* P. Yu,* M. I. Howard, Y. Yang, C. Zhang, A. Jadbabaie, A. Gorou, A. N. Gaiser, T. C. Steimle, L. Cheng, NRH, arXiv:2508.08368 (2025)

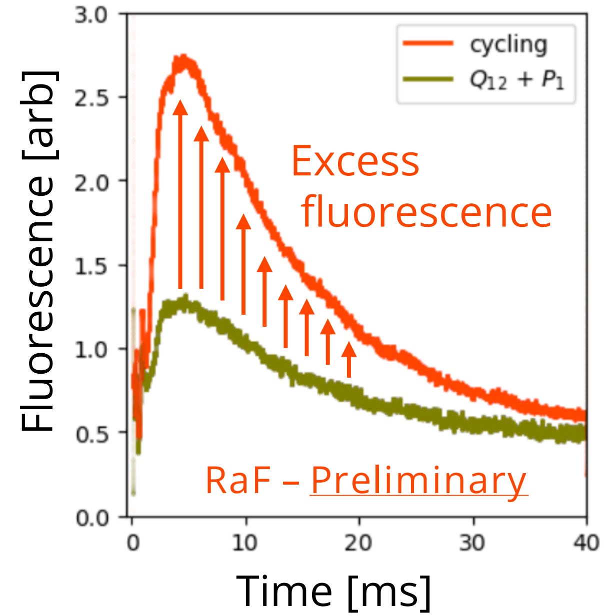
Spectra of 4 K RaOH, RaOD, RaF

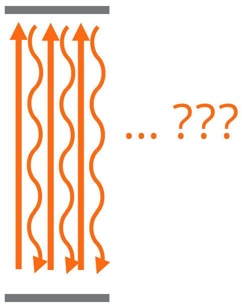


C. J. Conn,* P. Yu,* M. I. Howard, Y. Yang, C. Zhang, A. Jadbabaie, A. Gorou, A. N. Gaiser, T. C. Steimle, L. Cheng, NRH, arXiv:2508.08368 (2025)

Next Steps

- Main laser cooling transition, photon cycling
  - Preliminary observations
- Ra-225 and Ra-223
  - Can just “drop in”
  - Buy directly from NIDC or make locally with “cows”
- RaX Collaboration will laser cool radium molecules for precision measurements
  - NRH @ Caltech
  - John M. Doyle @ Harvard
  - Ronald Garcia Ruiz @ MIT
  - Support from FRIB
  - Commissioning beam





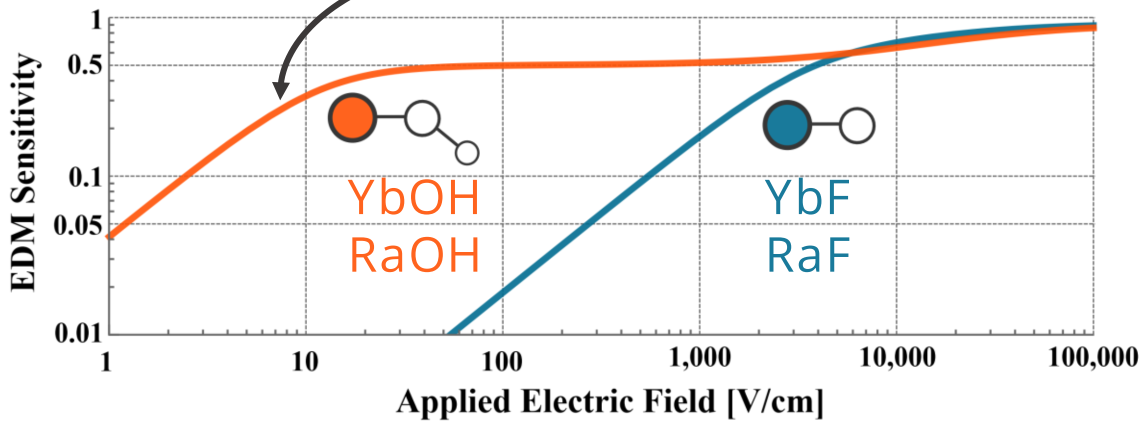
John M Doyle

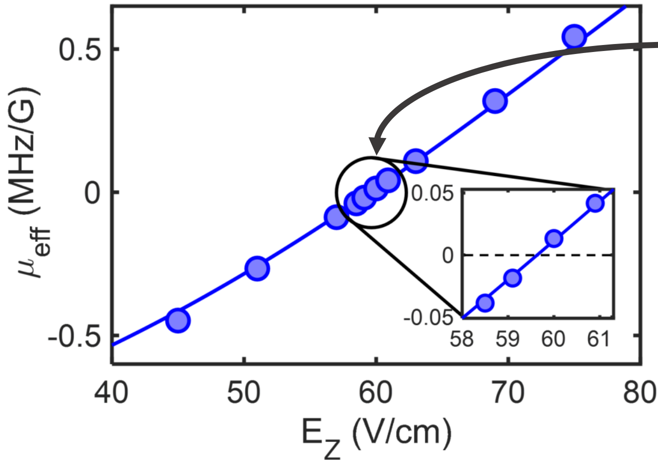
Ronald F Garcia Ruiz

Quantum Tools

Advantages of Polyatomics

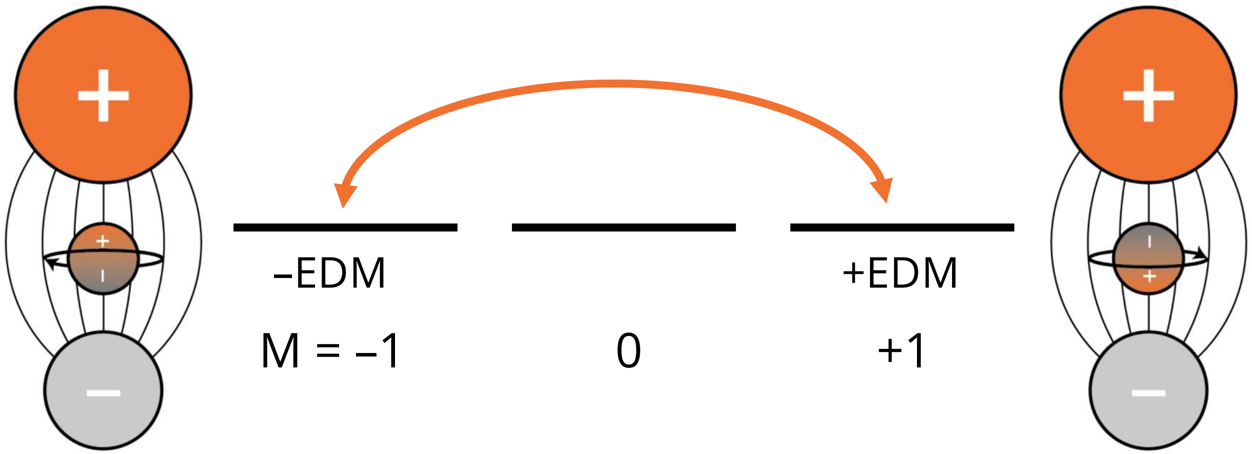
- Why polyatomic over diatomic?
- Polyatomics provide opportunities from additional degrees of freedom
  - High polarizability
  - Tunable electro-magnetic sensitivity
  - Many tools to reject systematic errors





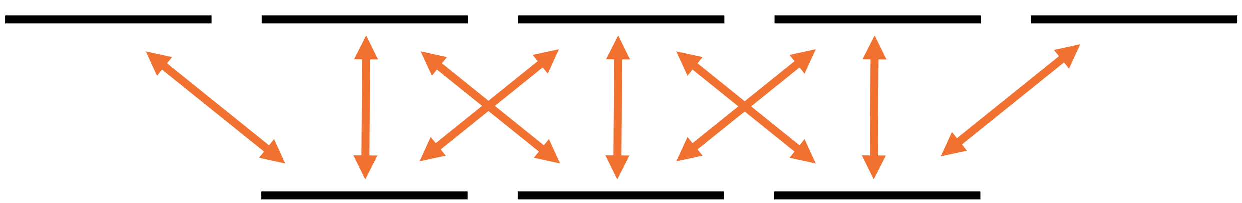
L. Anderegg, N. B. Vilas, C. Hallas, P. Robichaud, A. Jadbabaie, J. M. Doyle, NRH, Science 382, 665 (2023)
Y. Takahashi, C. Zhang, A. Jadbabaie, NRH, PRL 131, 183003 (2023)

New Measurement Approach



Spin precession

Analogous to Ramsey method



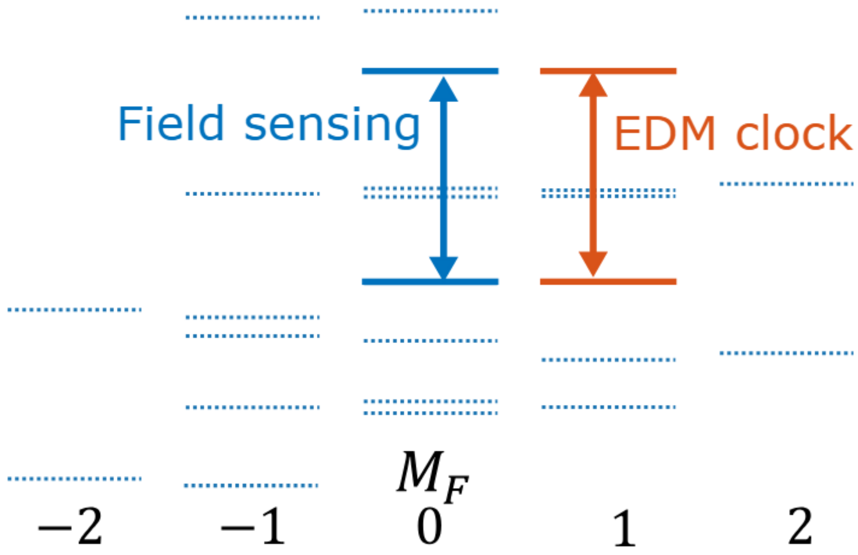
Clock measurement

Analogous to atomic clocks

Multiple measurement channels with tunable properties

Engineered Clock Transitions

- Independently, in-situ tunable E, B, CPV sensitivities
  - Tune with applied fields
  - Including making field sensitivities zero, flipped sign, …
  - Also many others: AC/DC polarizability, field polarization, transverse field sensitivity, …
- Measure using clock methods
- Generic in polyatomics
  - Including complex hyperfine structure needed for nuclear CPV (original motivation)

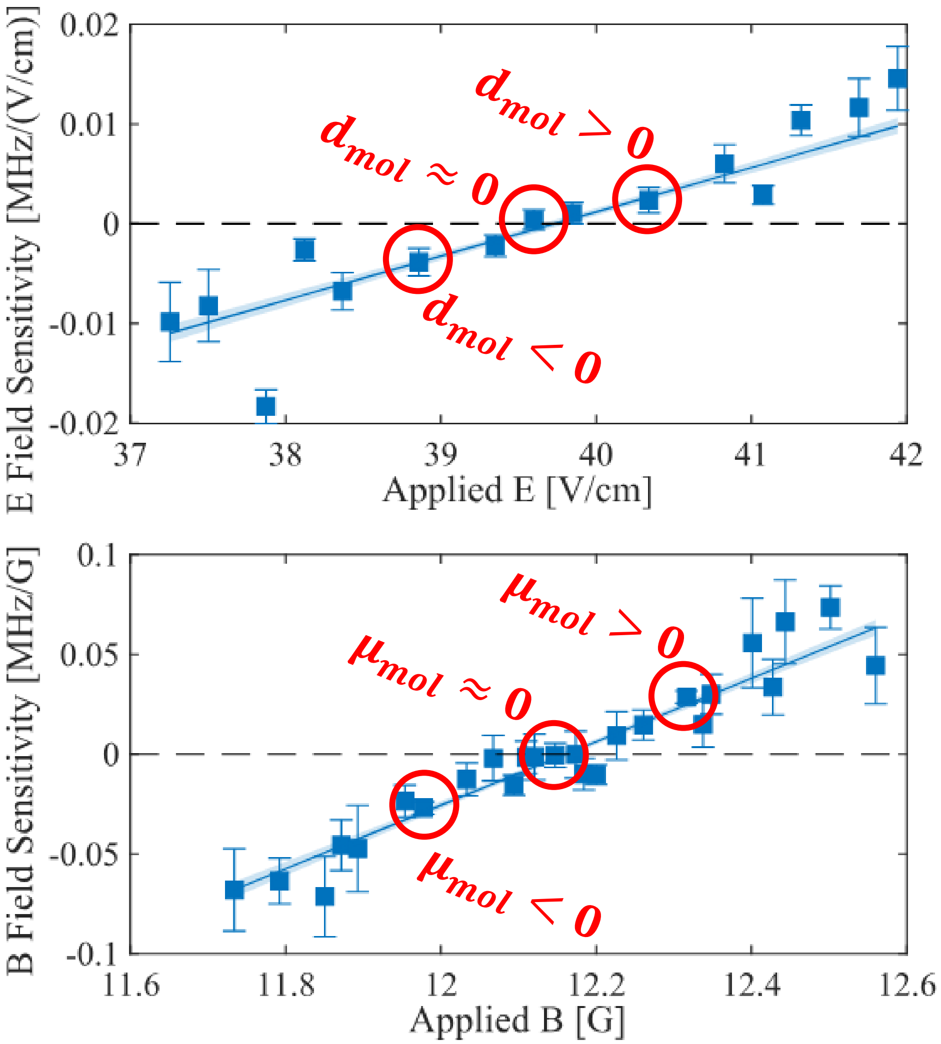


^{174}YbOH science state
E ~ 40 V/cm, B ~ 12 mG
~22 GV/cm internal field

Proposal: Y. Takahashi, C. Zhang, A. Jadbabaie, NRH, PRL 131, 183003 (2023)
Experiment: Y. Takahashi, H. D. Ramachandran, A. Jadbabaie, Y. Zeng, C. Zhang, NRH, arXiv:2508.06787 (2025)

Implementation in ^{174}YbOH

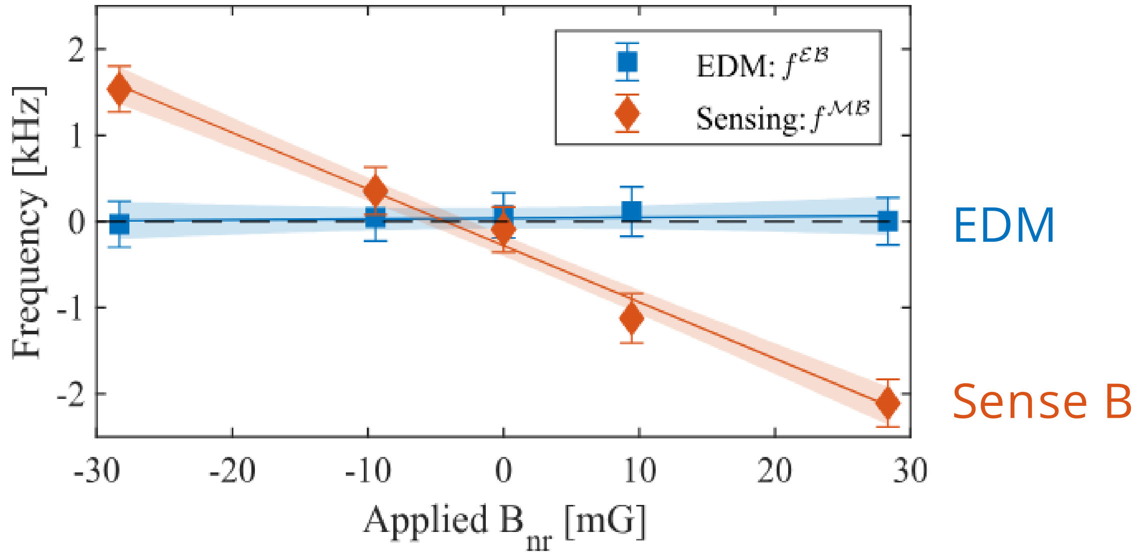
- Able to suppress electric, magnetic sensitivity to <1%
  - Likely lower – statistics limited
- Can independently set E, B CPV shifts to zero, or change sign/magnitude

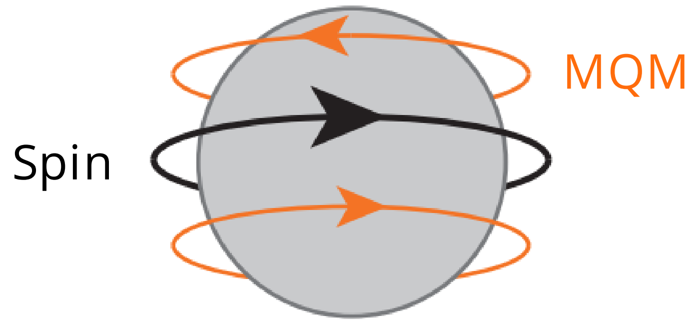


Y. Takahashi, H. D. Ramachandran, A. Jadbabaie, Y. Zeng, C. Zhang, NRH, arXiv:2508.06787 (2025)

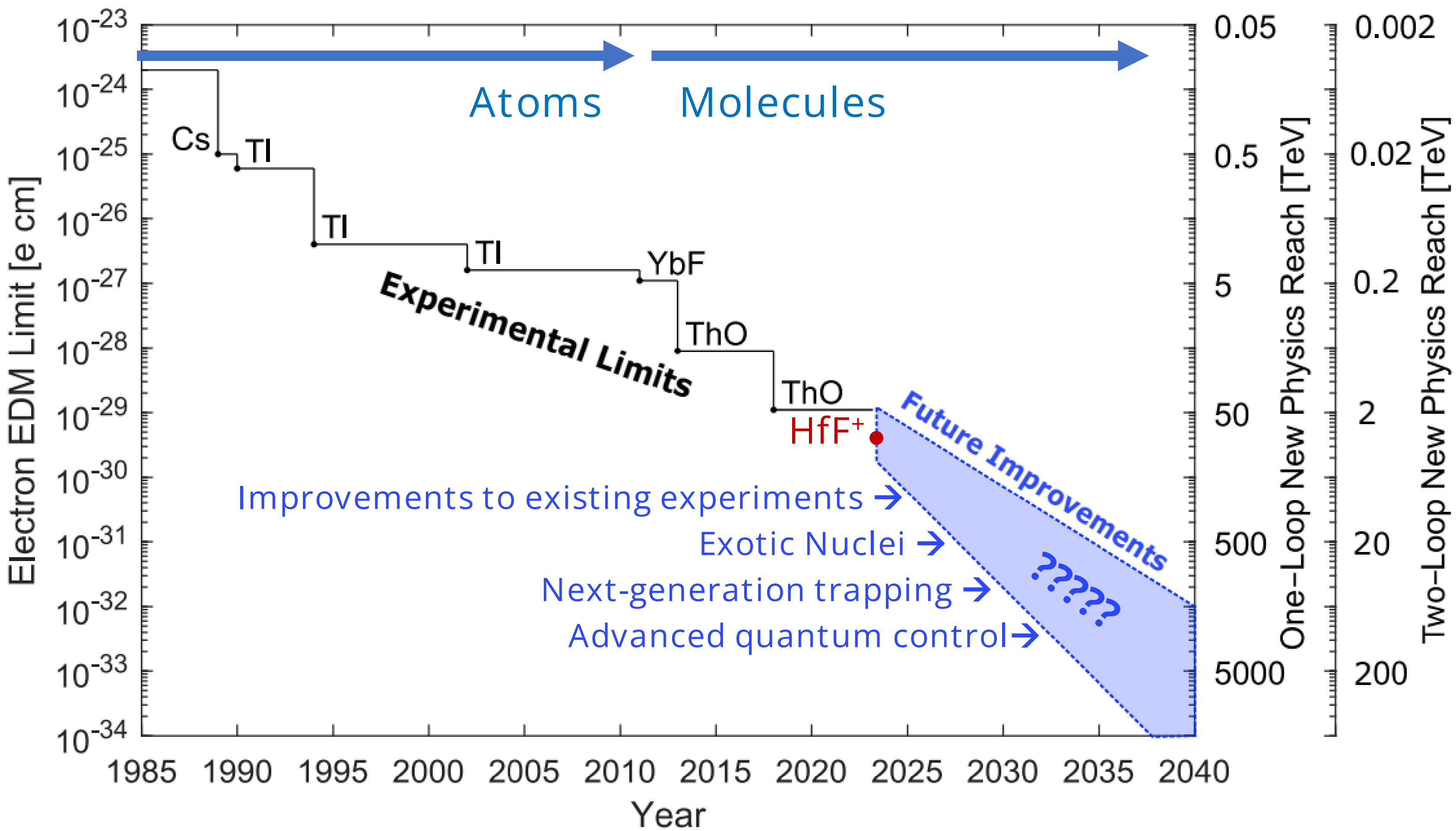
Implementation in ^{174}YbOH

- Can use tunable moments to directly sense fields
  - Like a co-magnetometer, but same molecule, states, lasers, etc.
- Currently using ^{173}YbOH – motivated by nuclear symmetry violation
  - ^{173}Yb(I=5/2) nuclear magnetic quadrupole moment (MQM)
  - Taking data, improving count rate





Future Improvements



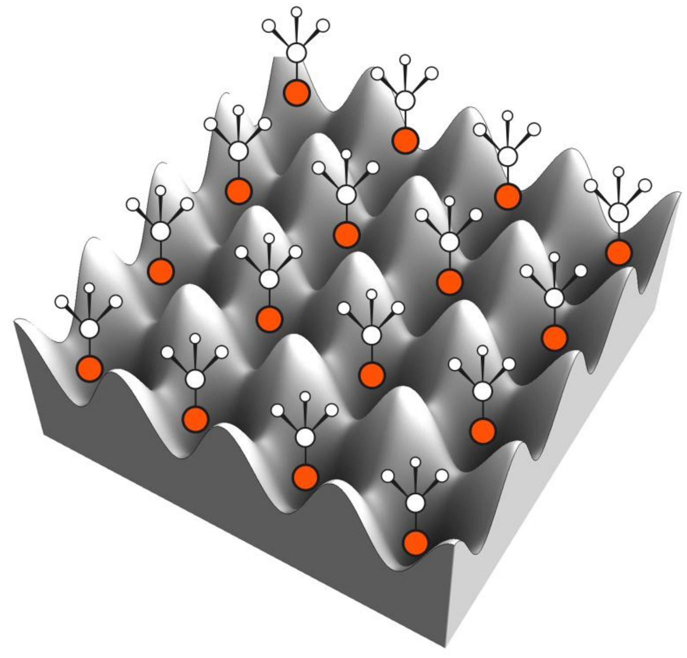
Similar improvements in hadronic CPV are also anticipated

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, $\boldsymbol{\theta_{QCD} \lesssim 10^{-14}}$
Near Standard Model CKM value
~10 – 20 year time scales

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



Hutzler Lab, Summer 2025
www.hutzlerlab.com
Left to Right: Tim, Chi, Nick, Sophie, Chandler, Madison, Yuxi, Kim, Yuiki, Steph, Phelan, Tatyana

Caltech

Collaborators

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

Molecular Theory: Anastasia Borschevsky (Groningen), Bill Goddard (Caltech), Lan Cheng (JHU)

Radium Molecules: Lan Cheng (JHU), John M. Doyle (Harvard), Alyssa Gaiser (MSU), Ronald Garcia Ruiz (MIT)

PolyEDM
polyedm.com

SLAM!
slamcommunity.com

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