Fundamental Symmetry Violations with Molecules

An overview of the field with an emphasis on selected ongoing developments

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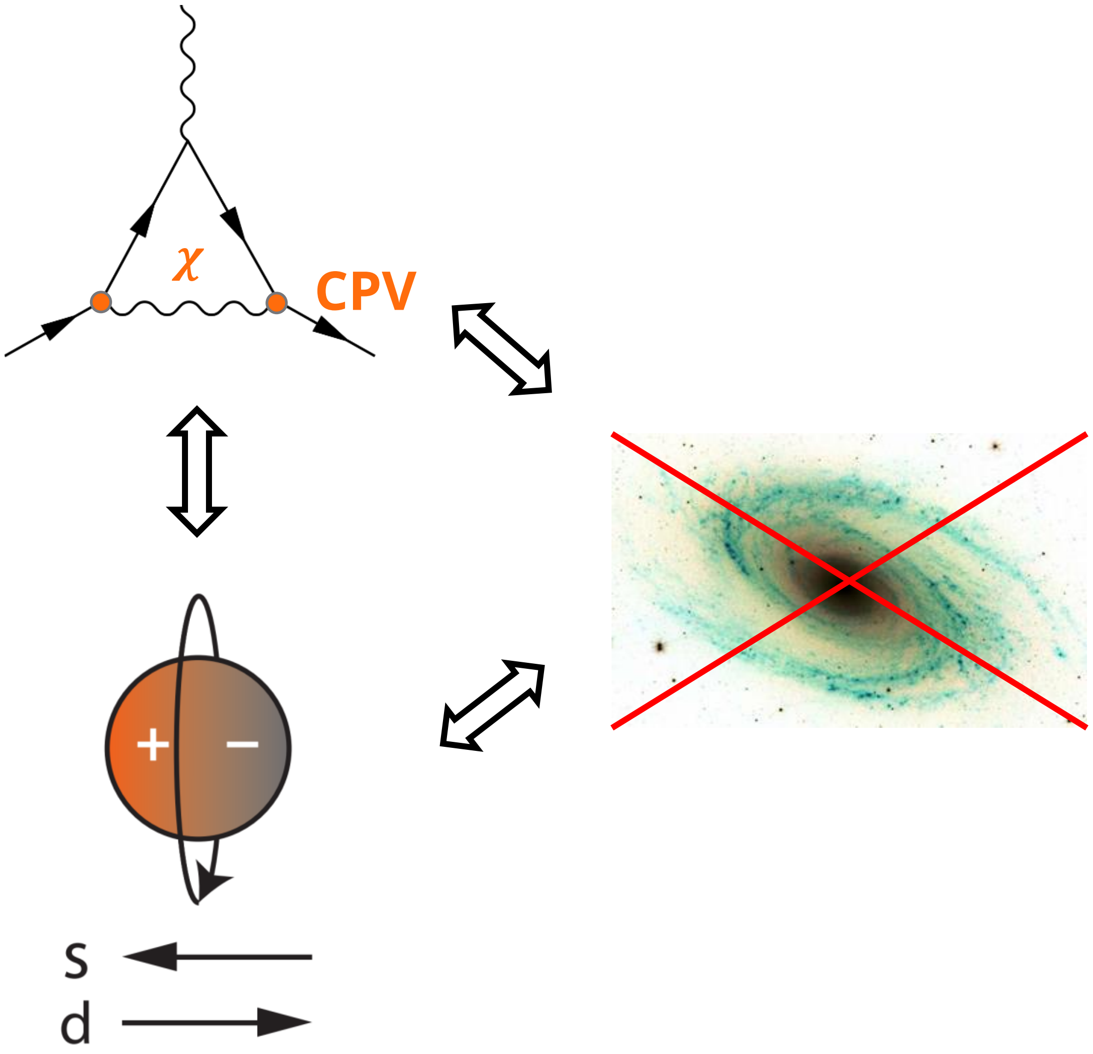
Caltech

Low Energy Observables

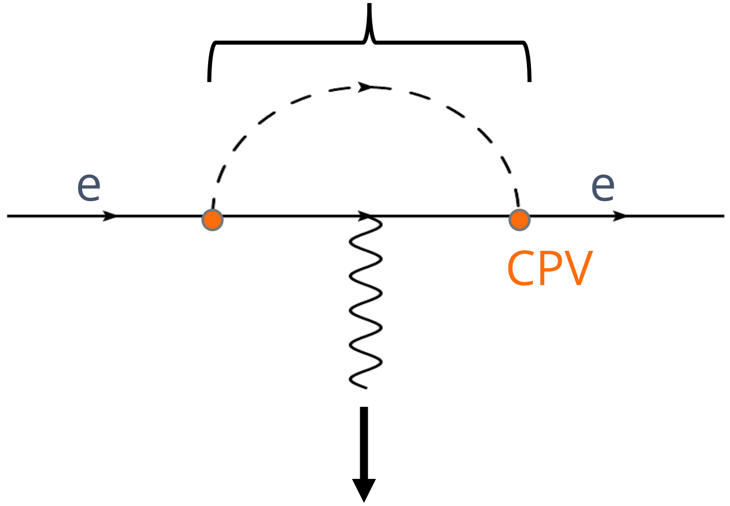
- The baryon asymmetry suggests new CP-violating physics
- Leads to CPV electromagnetic moments of regular matter
  - Electric dipole moment (EDM)
  - Nuclear Schiff moment (NSM)
  - Magnetic quadrupole moment (MQM)
- Enables sensitive probes of new physics



Electron EDM

- Generically sensitive to CPV particles and forces coupling to the electron
  - One loop ~ 10-50 TeV
  - Two loop ~ 0.5-2 TeV
- "Background free"
  - SM value is small
  - $|d_e| \sim 10^{-35}$ e cm
    - (Complicated)
  - Arises from CKM @ 4 loops
- For specific models, energy reach can be even higher (or lower!)

φ_{CP} ~ 1, M ~ 50 TeV



$|d_e| \sim 1{\times}10^{-29}$ e cm
(current experimental limit)

Sensitivity

- An EDM experiences a torque in an electric field
- Experimental observable is angle φ (phase),

$$\varphi = d\mathcal{E}\tau/\hbar$$

- Repeated measurements:

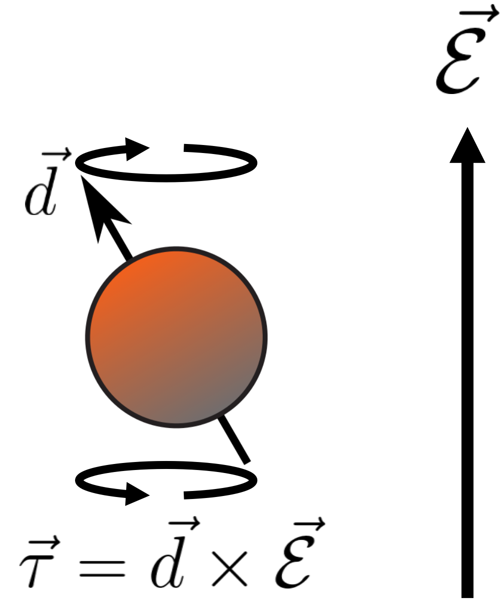
$$\delta d = \hbar/\mathcal{E}\tau\sqrt{N}$$

Effective electric field

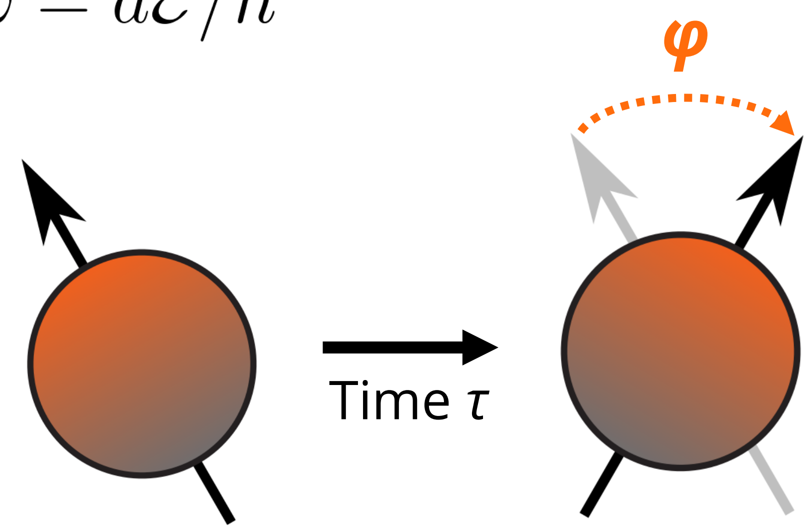
Coherence time

Total measurements

Make these large!

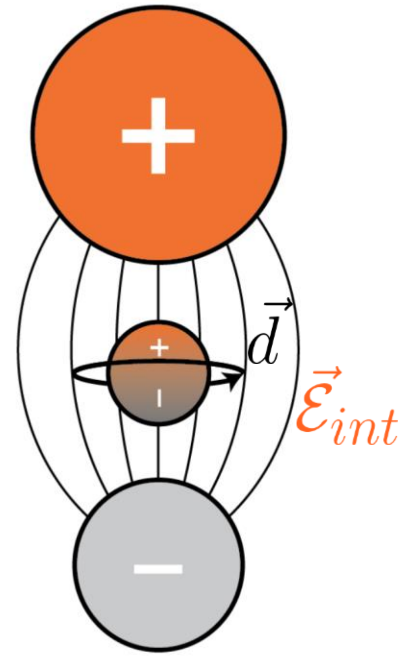


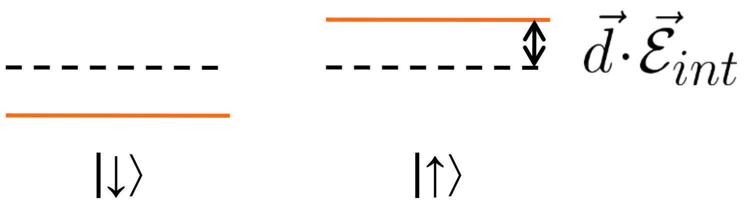
$$\vec{\tau} = \vec{d} \times \vec{\mathcal{E}}$$
$$\omega = d\mathcal{E}/\hbar$$



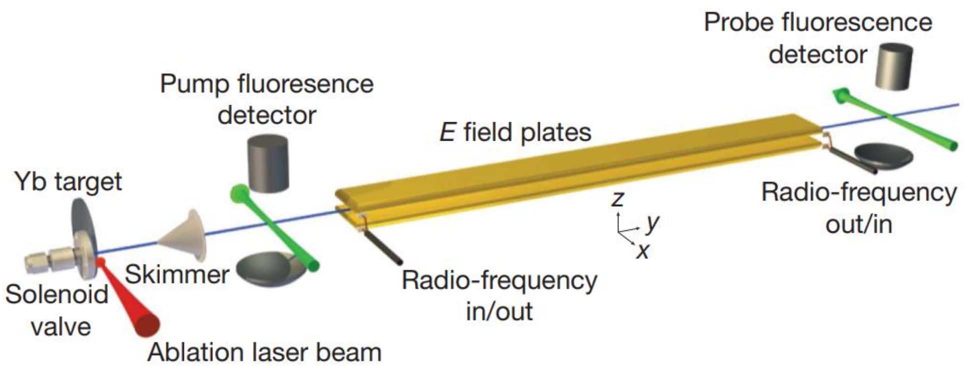
Electric field?

- Atoms/molecules have **extremely large** fields
  - 1-100 GV/cm for heavy species
  - Maximum lab field ~100 kV/cm
- CPV moments cause CPV molecular energy shifts
  - Enhancements for EDM, NSM, and MQM
- Measure using coherent methods (Ramsey)
- Molecular polarizability enhances sensitivity by ~1,000 vs. atoms
  - Atoms set best electron EDM limits until 2011 – molecules are complicated!
  - Atoms still best in other areas… *for now!*

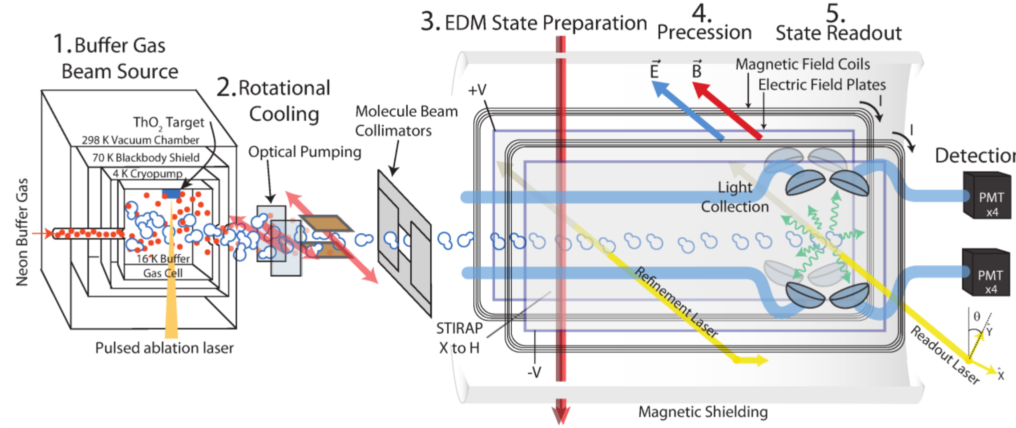




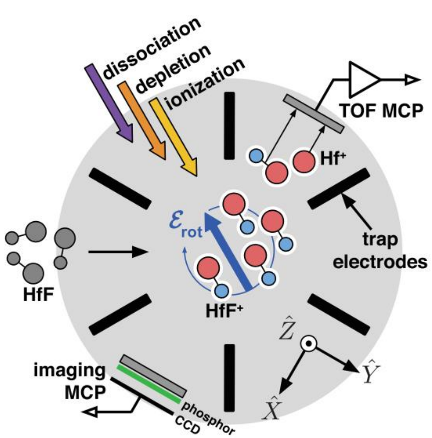
Molecular EDM Searches



YbF, Imperial
- Spin precession in pulsed supersonic beam
- First to beat atomic experiments
- $|d_e| < 1.1 \times 10^{-27}$ 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
- Long coherence time from trapping
- $|d_e| < 1.3 \times 10^{-28}$ e cm (2017)
- **$|d_e| < 4.1 \times 10^{-30}$ e cm (2022)**

- **100x in 10 years**
- **Each experiment is being upgraded**
- **More are under way**
- ***Atom technology is also advancing!***

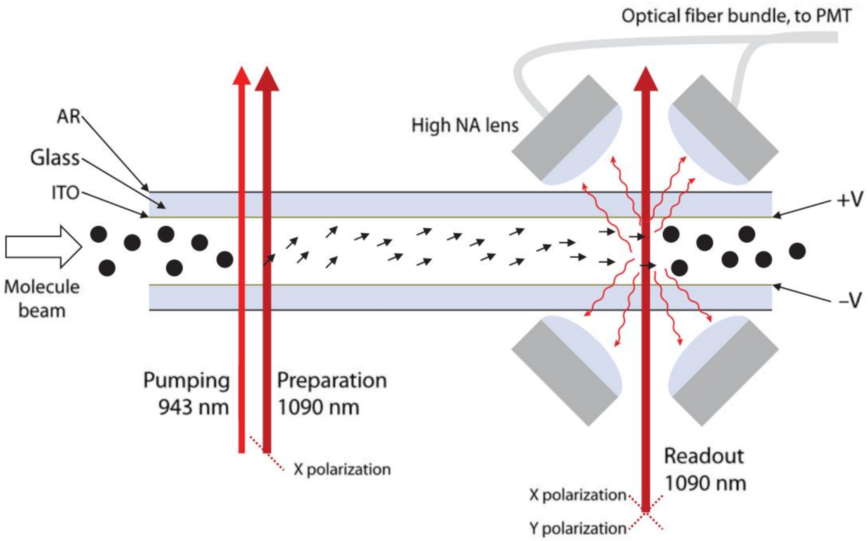
Next-generation tools

Laser cooling

Motivation for laser cooling

- Beam experiments (ThO, YbF) limited by time of flight, few ms
- Trapping can yield orders of magnitude improvement
  - Critical for long coherence time of HfF^+, Ra experiments
  - Ideal platform for quantum-enhanced metrology
- Neutral species must be "ultracold," <1 mK

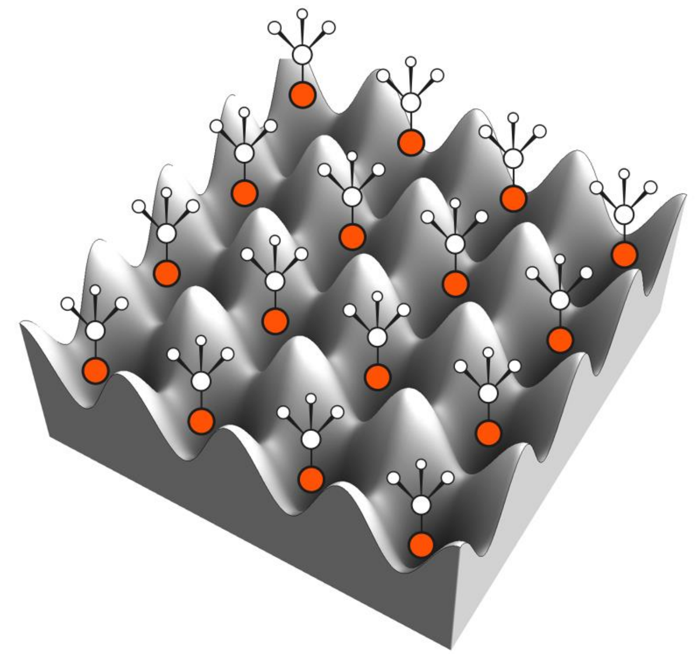
→ **Laser cooling**



ACME Generation I

Let's dream

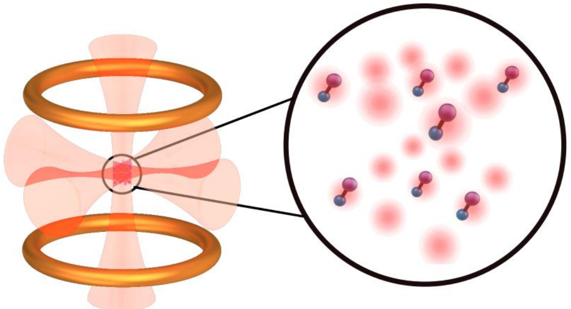
10^6 molecules
100 s coherence time
Deformed nucleus
Quantum control
Robust error rejection
Two weeks integration

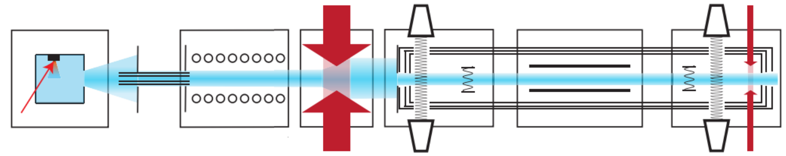


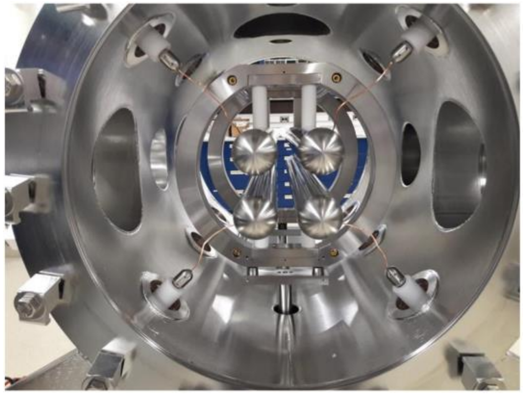
~PeV-scale CP-violating physics @ 1 loop
~100 TeV-scale CP-violating physics @ 2 loops
Both leptonic and hadronic sectors
Extreme precision, $\theta_{QCD} \lesssim 10^{-14}$

Even more room to improve!
Quantum-enhanced metrology
Truly exotic nuclei like ^{229}Pa?

Three Examples







YbF

- eEDM @ Imperial College London
- X. Alauze *et al*., Q. Sci. & Tech. 6, 044005 (2021)
- N. J. Fitch *et al.*, Q. Sci. & Tech. 6, 014006 (2021)

BaF

- NL-eEDM Collaboration
- P. Aggarwal et al., Eur. Phys. J. D 72, 197 (2018).
- P. Aggarwal *et al.*, PRL 127, 173201 (2021)

TlF

- CeNTREX Collaboration
- Tl Schiff moment (~proton edm)
- O. Grasdijk *et al*., Q. Sci. & Tech. 6, 014006 (2021)

Several more laser cooling examples later

Assembled Molecules

- Alternative route: laser cool atoms, coherently associate into molecules
- Directly get ultracold, high density samples
- FrAg, RaAg are excellent candidates
  - Large CPV sensitivity, highly polar, laser-coolable atoms
  - Ag “looks like alkali”

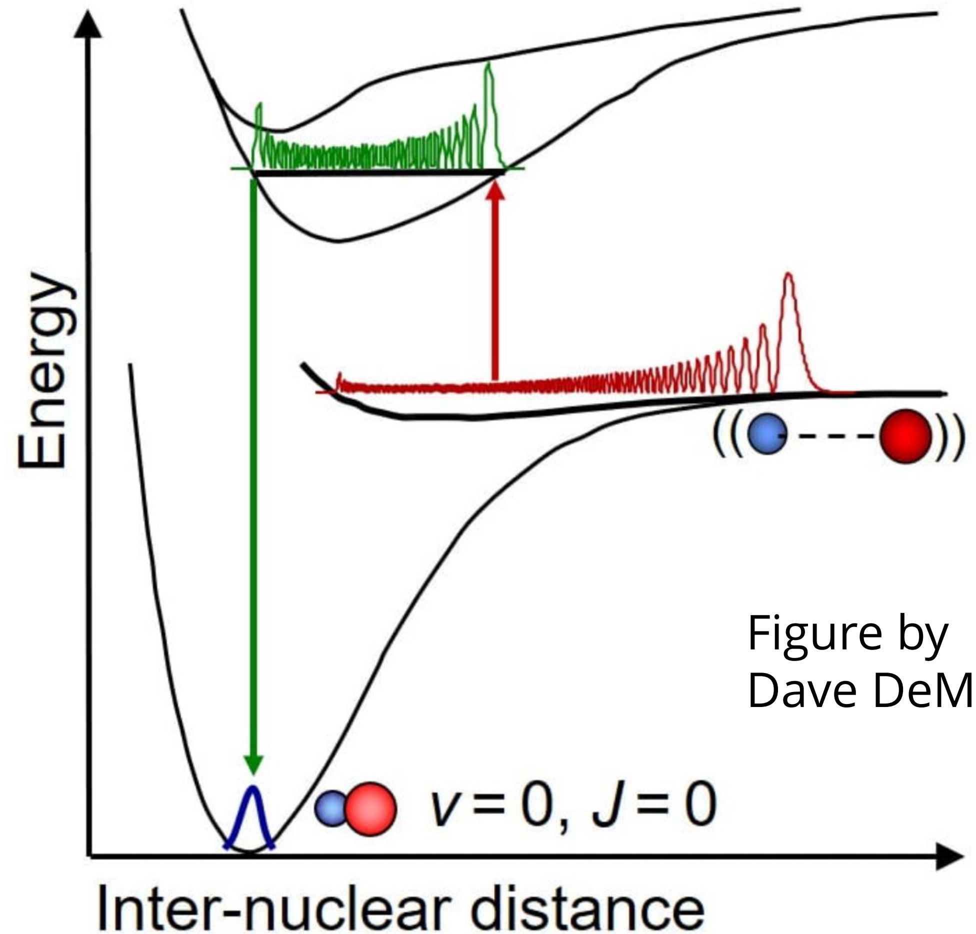


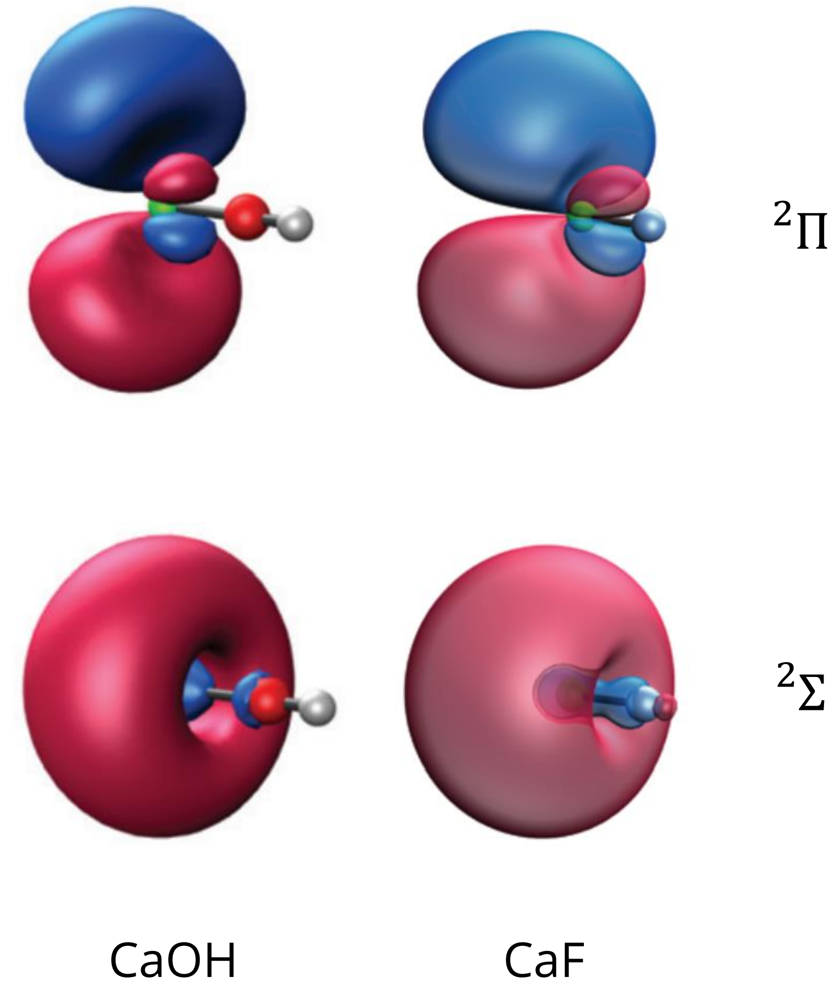
Figure by
Dave DeMille

T. Fleig and D. DeMille, NJP 23, 113039 (2021), J. Kłos, H. Li, E. Tiesinga, S. Kotochigova New J. Phys. 24, 025005 (2022)

Polyatomic Molecules

Polyatomic Molecules

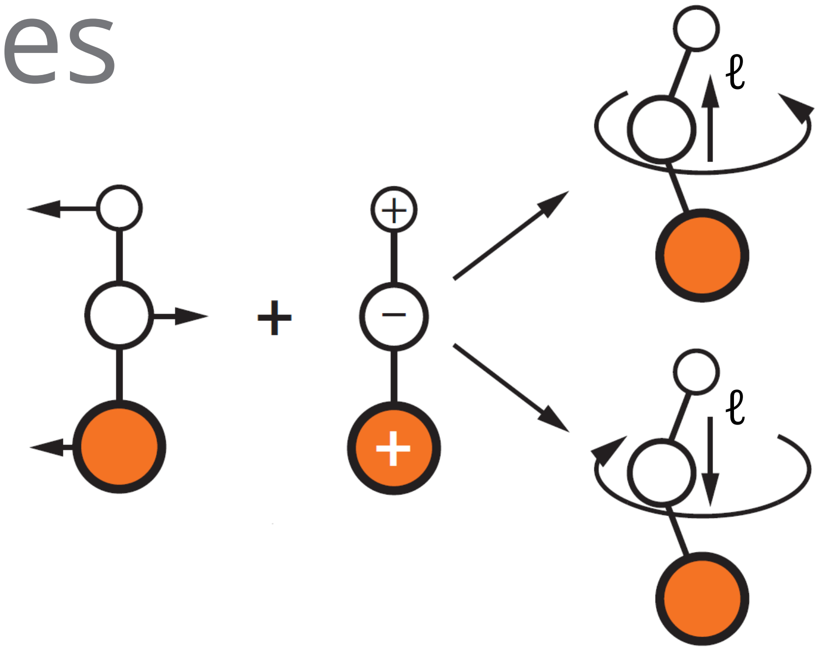
- Many bonding partners “bond similarly”
  - – F ≈ –OH, – –CCH, – OCH_3, …
  - Not in general, but for laser-coolable species it often holds
  - Since electron wavefunction is metal-centered, ligand matters much less
- Similar electronic structure implies similar:
  - Laser cooling/photon cycling
  - CPV sensitivity
  - Measurement methods
- … but with the possibility of engineering other features
  - Electric and magnetic field interactions
  - High polarizability (generic parity doublets)
  - Species in ligand
  - Frequencies of rotation and vibration
  - …

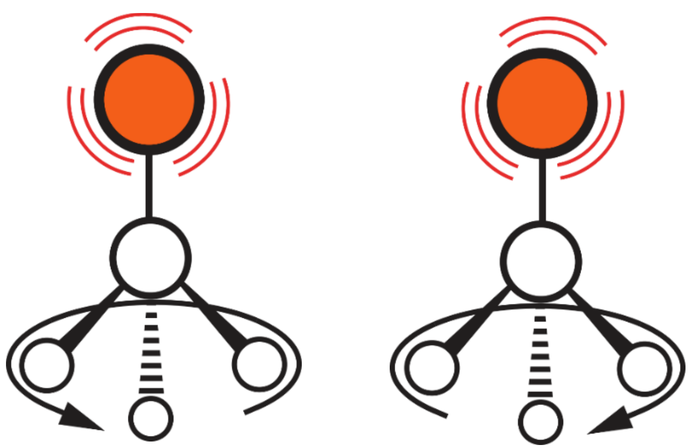


PRL 116, 063006 (2016), J. Phys. B 50, 225101 (2017), Phys. Chem. Chem. Phys. 21, 19447 (2019), Q. Sci. & Tech. 5, 044011 (2020)

Polyatomic Molecules

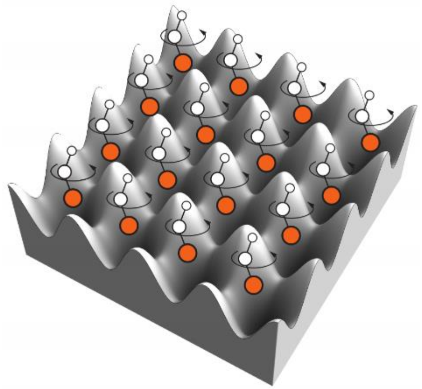
- Laser-coolable molecules have no parity doublets
  - Enable large polarization in small fields, robust systematic rejection
- Polyatomics *generically* have parity doublets
  - Arise from symmetry-lowering mechanical modes
- Parity doubling with incompatible electronic structure
  - For example, laser-coolable molecules
- Available for any atomic species

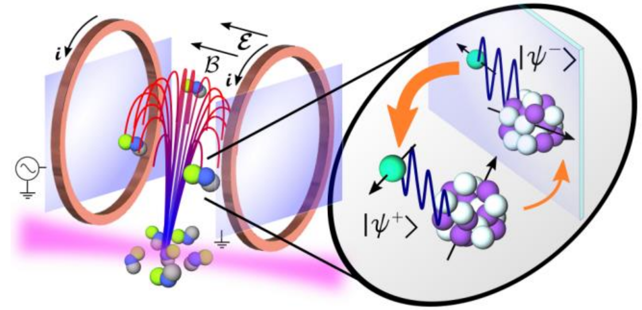


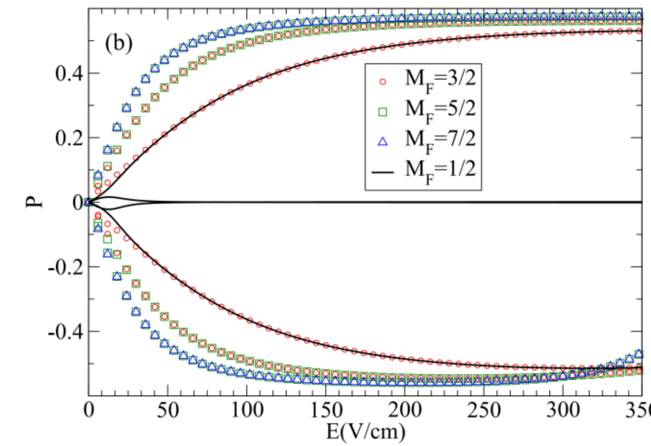


I. Kozyryev and NRH, Phys. Rev. Lett. **119**, 133002 (2017)

Three Examples







PolyEDM

- Combine laser cooling, high polarizability
- (Ca, Sr, Yb, Ra)OH
- SrOH electron EDM
  - PolyEDM: NRH, Doyle, Steimle, Vutha
  - See *polyedm.com*
- ^{173}YbOH nuclear MQM
- I. Kozyryev and NRH, PRL 119, 133002 (2017)

MgNC

- Engineer magnetic field interactions for PV
  - (More later)
- E. B. Norrgard, et al, Nat. Comm. Phys. 2, 77 (2019)

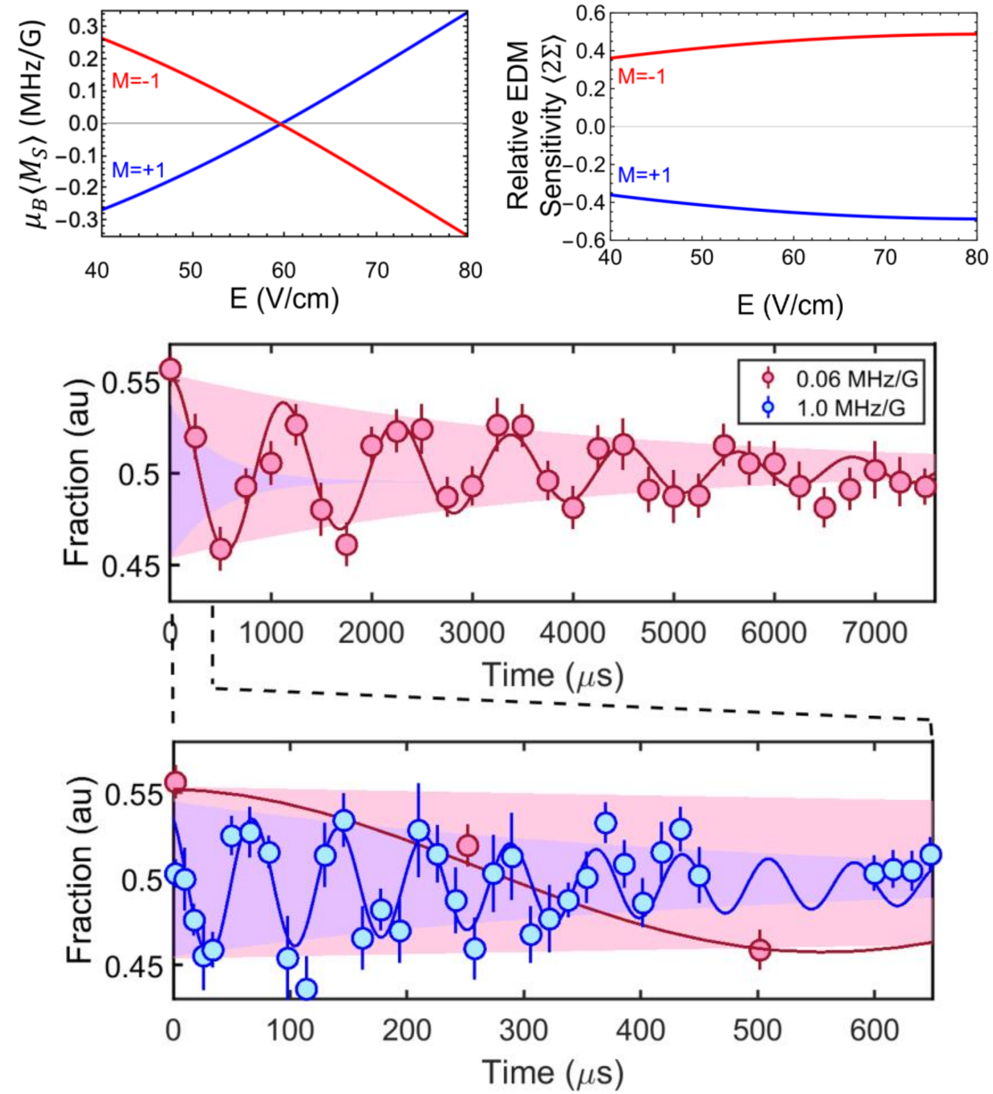
LuOH$^+$

- Combines quadrupole deformed, MQM-sensitive nucleus with ion trapping
- D. E. Maison, L. V. Skripnikov, G. Penyazkov, M. Grau, A. N. Petrov, PRA 106, 062827 (2022)

… and many more! See Q. Sci. Tech. 5, 044011

Tunable Moments

- Polyatomic structure gives tunability of electromagnetic moments
- Zero-crossings of magnetic field sensitivity which maintain EDM sensitivity
  - Can tune-out magnetic field noise, decoherence
  - Can also tune out EDM sensitivity as a check
- Demonstrated in ultracold, trapped CaOH @ Harvard



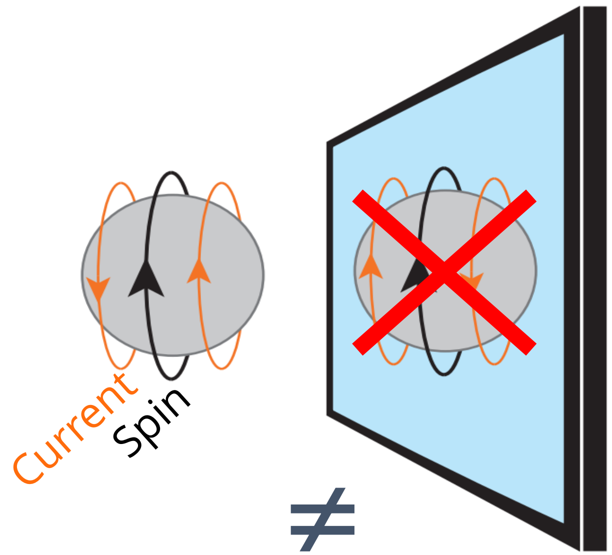
Loïc Anderegg
Harvard University

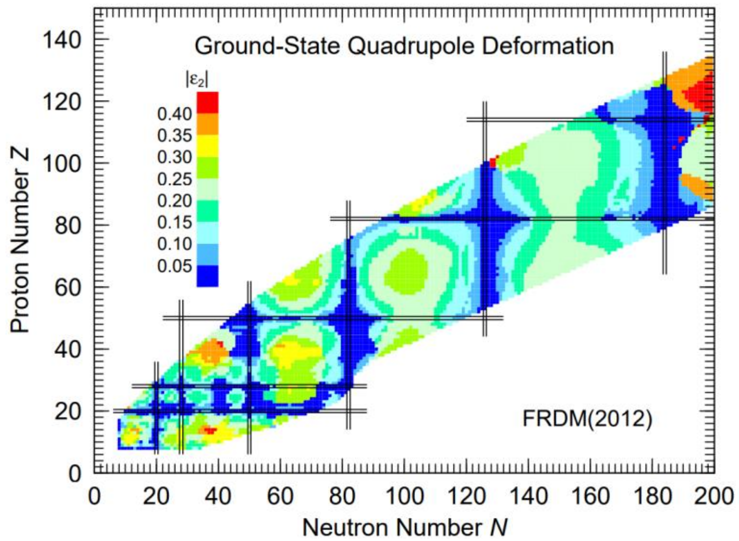
L. Anderegg, N. B. Vilas, C. Hallas, P. Robichaud, A. Jadbabaie, J. M. Doyle, NRH, arXiv:2301.08656 (2023)

Deformed Nuclei

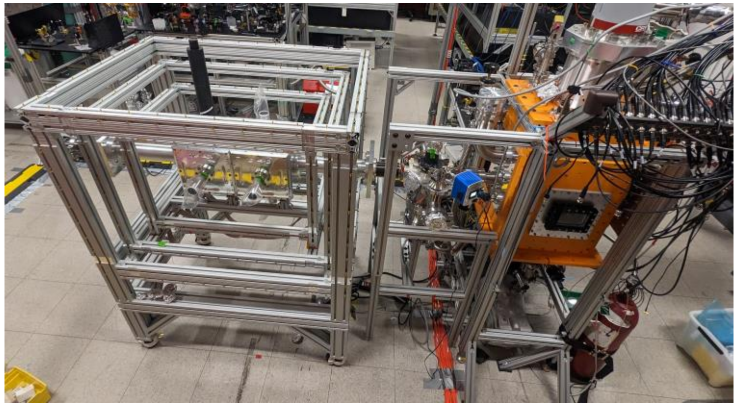
Hadronic CPV Enhancement

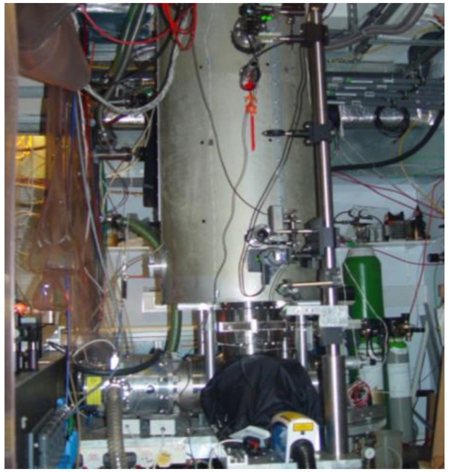
- Quadrupole (β_2) and octupole (β_3) deformations can enhance hadronic CPV
  - θ_{QCD}, chromo-EDMs, nucleon EDMs, CPV forces, …
  - Combines with molecular enhancements
- β_2: Magnetic quadrupole moments (MQMs)
  - Collective enhancement, typically ~10
  - "Many options"
  - Yb, Lu, Ta, Hf, Th, Ra, …
    - V. V. Flambaum, et al., PRL 113, 103003 (2014)

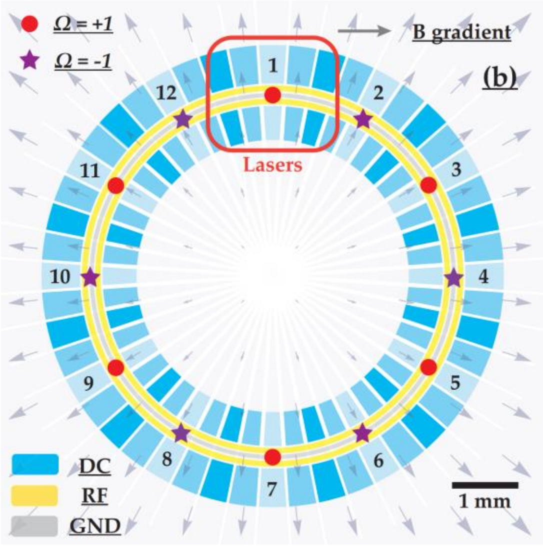




Three Examples







^{173}YbOH

- Combine photon cycling and polarizable polyatomic structure
- Laser cooling in future generations
- I. Kozyryev and NRH, PRL 119, 133002 (2017)

^{173}YbF

- Leveraging advanced experimental methods with YbF (from eEDM)
- C. J. Ho, J. Lim, B. E. Sauer, M. R. Tarbutt, Front. Phys. 2, 11 (2023)

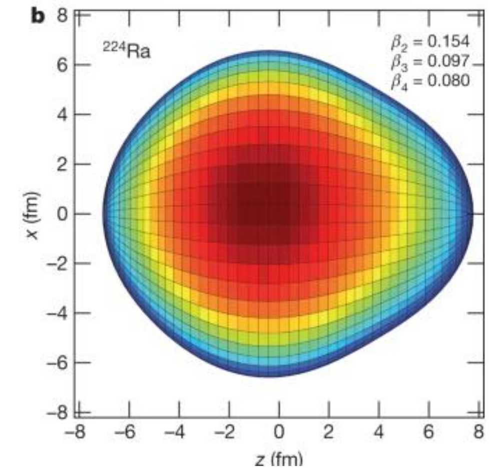
^{181}TaO$^+$

- Ion trapping with advanced quantum logic control
- T. N. Taylor, J. O. Island, Y. Zhou, 2210.11613 (2022)

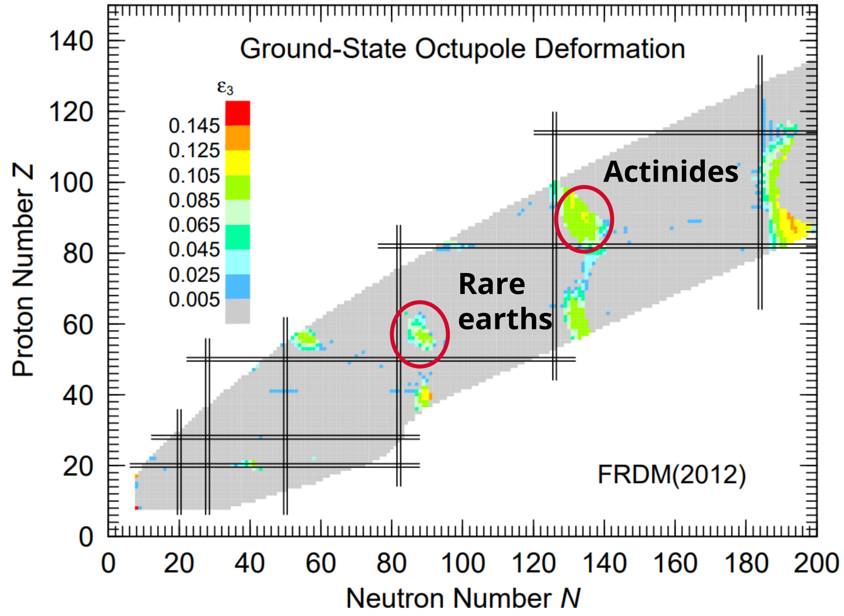
Also LuOH$^+$ mentioned earlier

Octupole Deformations

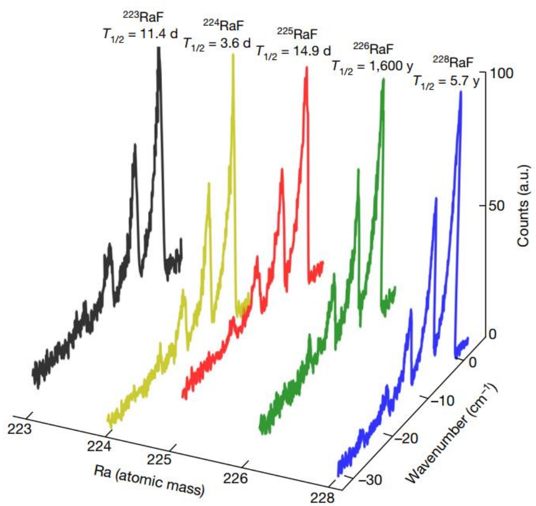
- β_3: Schiff Moments (NSMs) enhanced by ~100-1,000
  - Fr, Ra, Ac, Th, Pa, …
  - Heavy, spinful, deformed species are short-lived
- Combines with molecular enhancements → $10^{5\text{-}6}$ sensitivity gain vs. atoms with spherical nuclei
  - Trapping *one* molecule could probe frontiers of hadronic CPV
- Truly exotic nuclei like ^{229}Pa offer *another* factor of 100-1000 (maybe)
- You will hear more about these deformed nuclei next



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

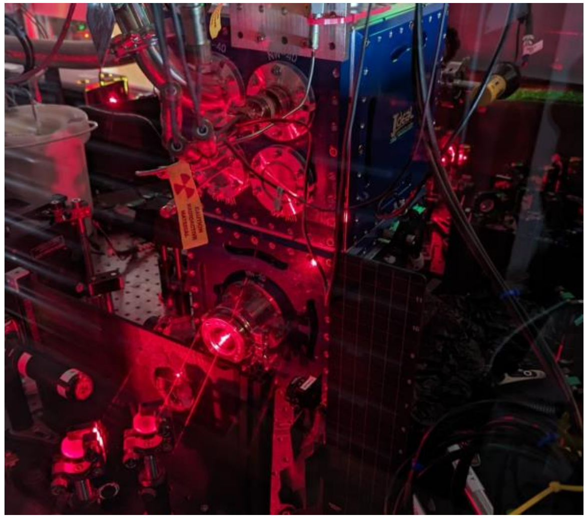


Three Radium Examples



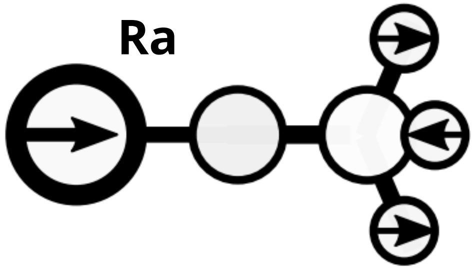
RaF

- First spectroscopically studied radioactive molecule
- Laser-coolable, good for eEDM, NSM, MQM, PV searches
- R. F. Garcia Ruiz *et al.*, Nature 581, 396 (2020)



RaOH

- Laser-coolable, good for eEDM, NSM, MQM, PV searches
  - ...with parity doubling
- Recent studies at Caltech
- OH → OCH_3, SH, ... later?



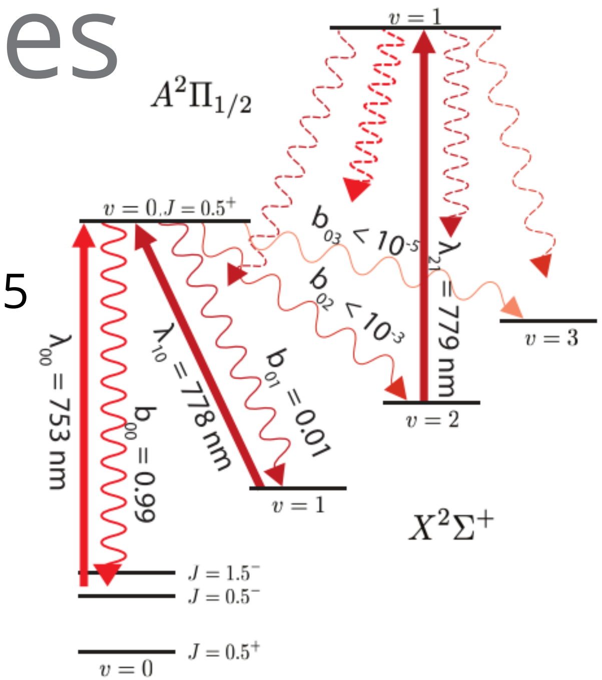
RaX^+ Polyatomics

- X = OH, SH, OCH_3, ...
- Ion trapping with parity doubling
- Created at UCSB (Jayich)
- M. Fan et al., PRL 126, 023002 (2021)
  P. Yu and NRH, PRL 126, 023003 (2021)

Neutral Radium Molecules

- RaF, RaOH, others are expected to be *very* laser-coolable
  - ~98-99% diagonal
  - RaF spectroscopy supports this expectation

RaF Braching (Calculated)
arXiv: 2302.02165



Calculated RaOH A-X Branching Ratios

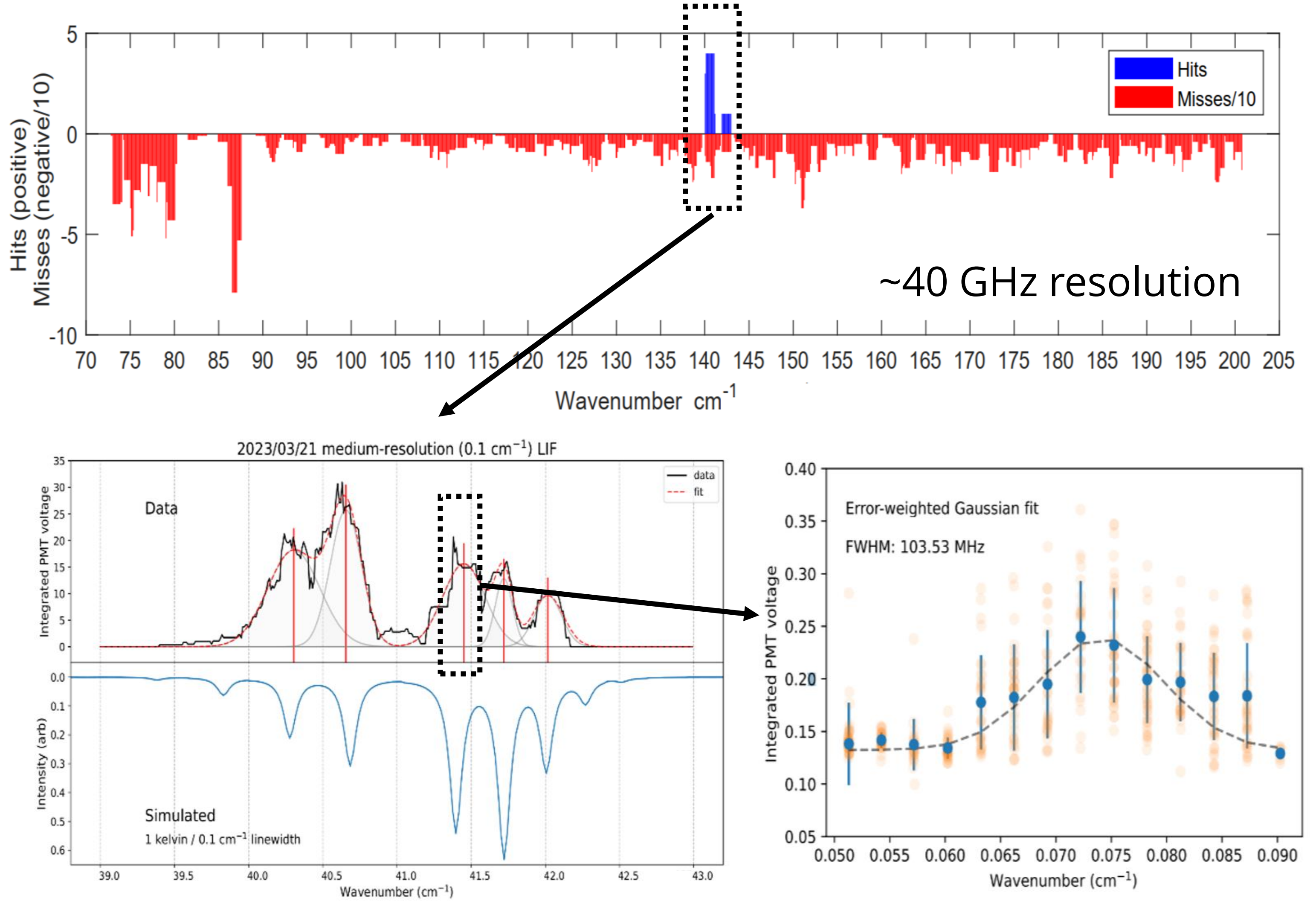
Vibrational states	energy level	branching ratio
(000)	0	98.972
(010)	337	0.001
(100)	475	0.863
(02^00)	646	0.138
(02^20)	678	0.007
(200)	947	0.012
(12^00)	1111	0.005

C. Zhang, NRH, L. Cheng, Submitted (2023)

T. A. Isaev, A. V. Zaitsevskii, E. Eliav, J. Phys. B. 50, 225101 (2017), I. Kozyryev and NRH, Phys. Rev. Lett. 119, 133002 (2017)

RaOH Spectroscopy

- ***Very preliminary***: just started ^{226}RaOH spectroscopy at Caltech
- Cooled in 4 K He
- ~100 MHz linewidth, ~10 ms interrogation
- Tabletop apparatus
- Temperature, density at starting point for precision measurement, laser cooling
- Efficient search strategy (limited quantities!)
- Method should be generalizable to many other radioactive species
- Predictions by Chaoqun Zhang and Lan Cheng (JHU) were *very* accurate



~3 GHz resolution

~100 MHz resolution

Outlook

nEDM, NSM, MQM – Complementarity

- CPV experiments in atoms and molecules are sensitive to multiple underlying sources
- Need to develop broad range of experiments looking for different moments using different species
  - **Required** to disentangle multiple sources for robust limits, and eventual identification of CPV sources
  - Beyond "double-checking" – unavoidable science need

Wilson coefficient	Operator (dimension)	Number	Systems
$\bar{\theta}$	Theta term (4)	1	Hadronic & diamagnetic atoms
δ_e	Electron EDM (6)	1	Paramagnetic atoms & molecules
$\mathrm{Im}\, C_{\ell equ}^{(1,3)}$, $\mathrm{Im}\, C_{\ell eqd}$	Semi-leptonic (6)	3	
δ_q	Quark EDM (6)	2	Hadronic & diamagnetic atoms
$\tilde{\delta}_q$	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	

J. Engel, M. J. Ramsey-Musolf, and U. van Kolck, Prog. Part. Nucl. Phys. **71**, 21 (2013)

SLAM! Community

SLAM!

Physics with Short-Lived Atoms and Molecules

www.slamcommunity.com

What will it take to access CPV?

BSM Phenomenology

Molecular spectroscopy

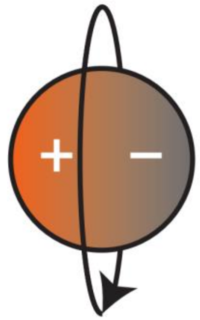
Sensitive Detection

Radiochemistry

Beam facilities

Molecular production

Laser cooling/trapping



Particle Theory

Quantum control

Nuclear structure

QIS

QCD

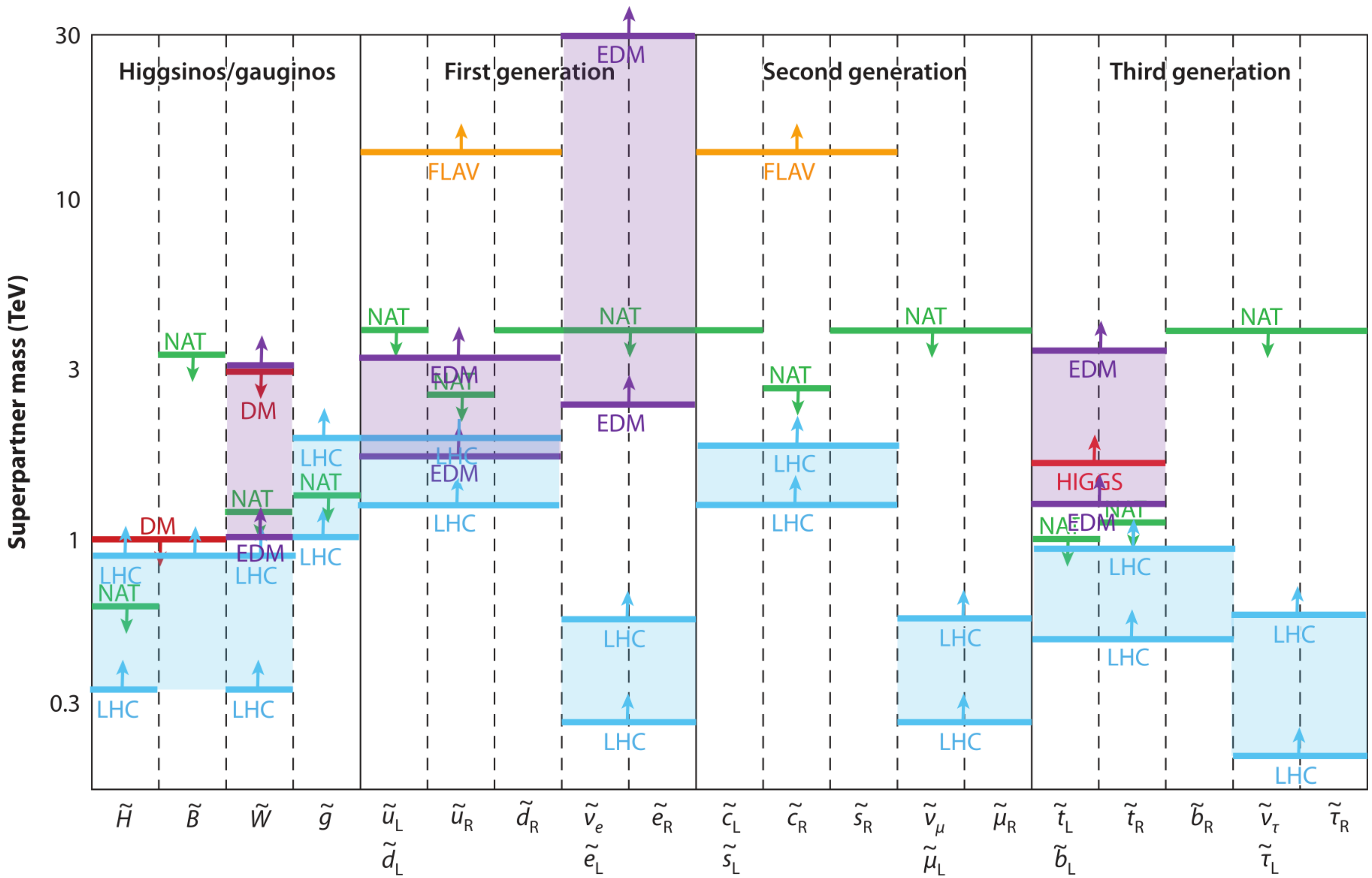
Physical chemistry

Atomic/molecular structure

Relativistic quantum chemistry

And more…

Many complementary approaches

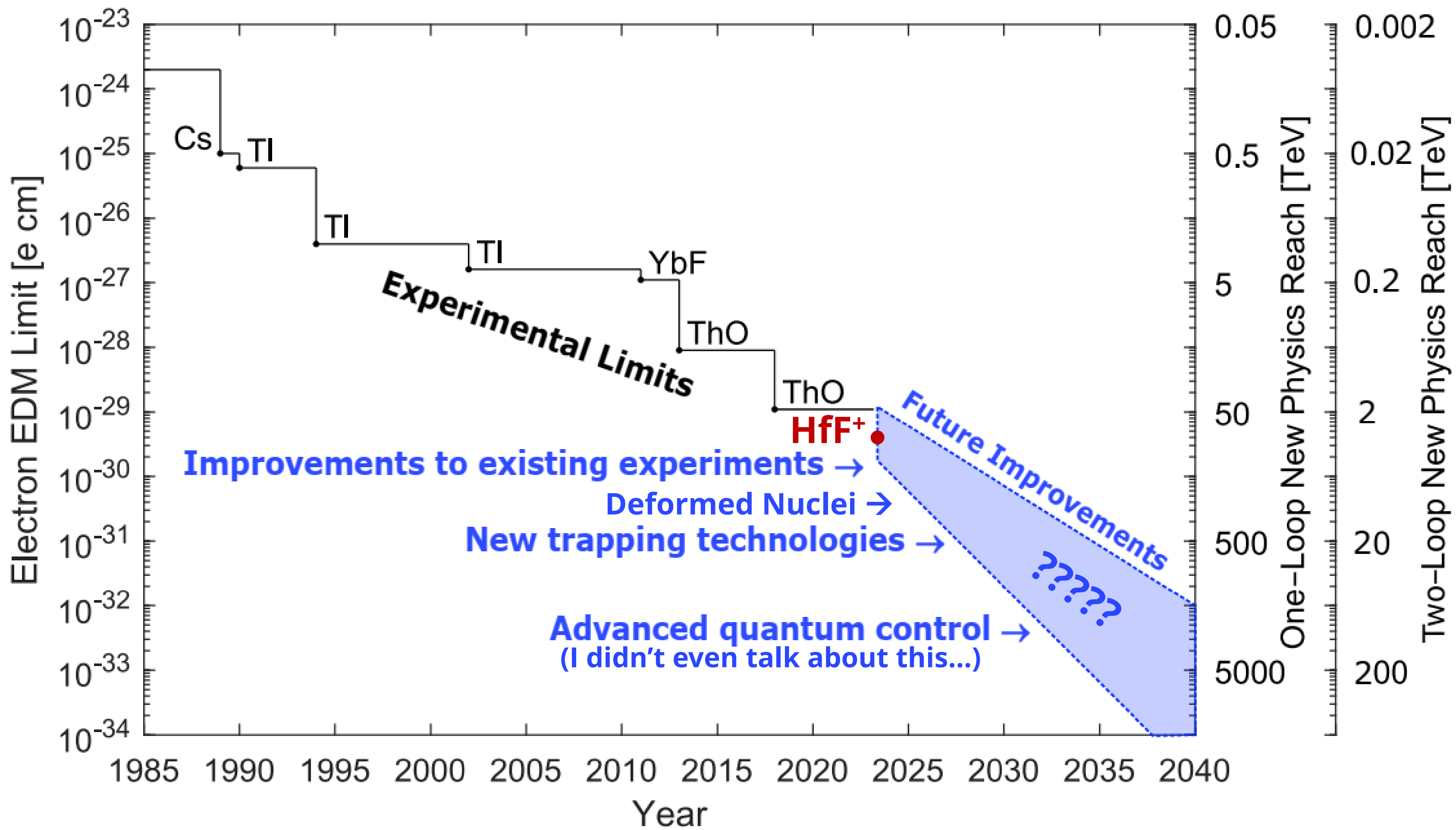


Shading shows progress since 2013 (LHC, ACME, JILA, nEDM, ^{199}Hg)

"All of the constraints shown are merely indicative and are subject to significant loopholes and caveats." –J. Feng

Adapted and updated from J. Feng, Ann. Rev. Nuc. Part. Sci. 63, 351 (2013) with help from D. DeMille

A Positive Outlook



From 2022 Snowmass EDM whitepaper, arXiv:2203:08103 **– Already out of date!**