Caltech

Experimental Searches for Fundamental Symmetry Violations with Molecules

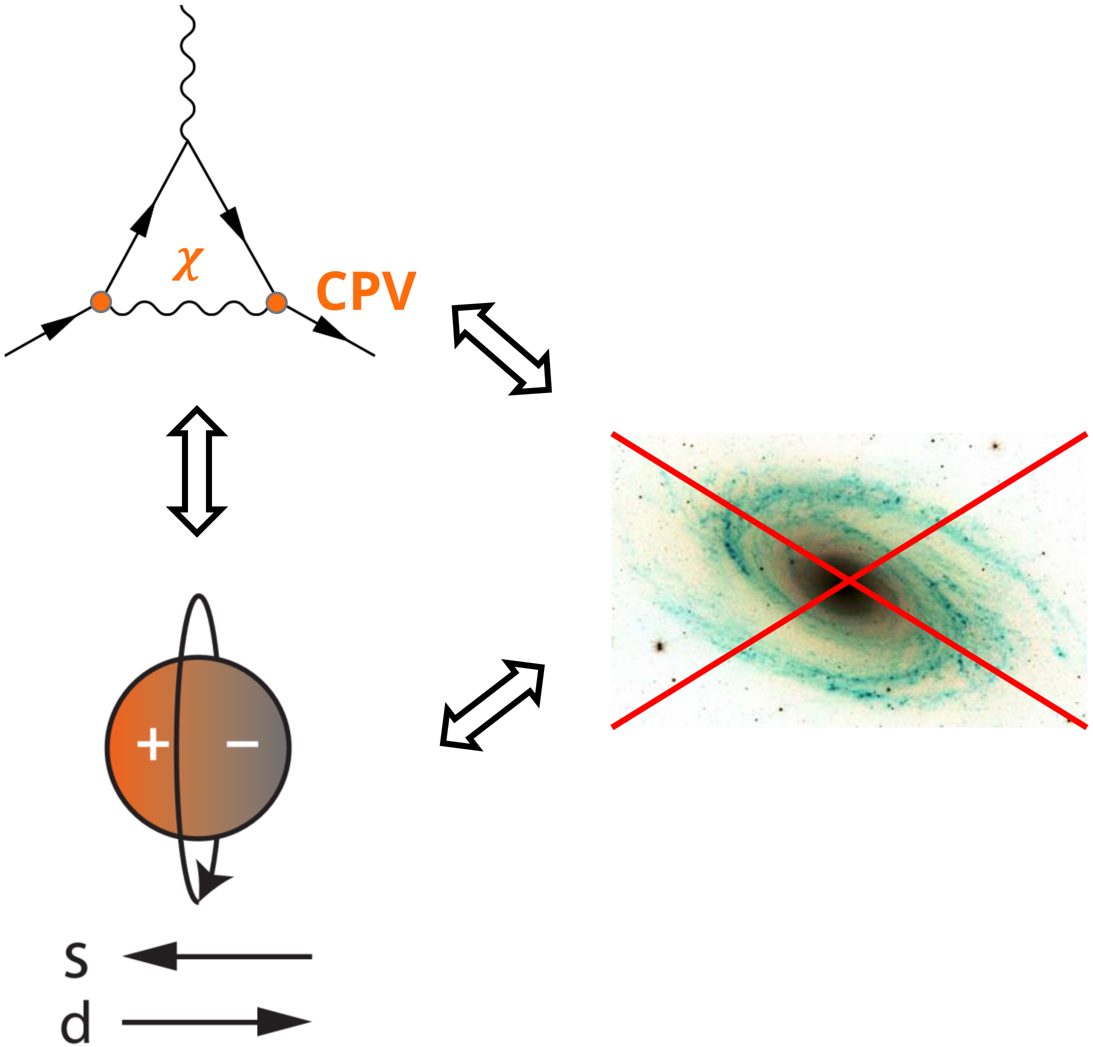
An overview with emphasis on selected current trends

Nick Hutzler

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
- These effects are enhanced in molecules



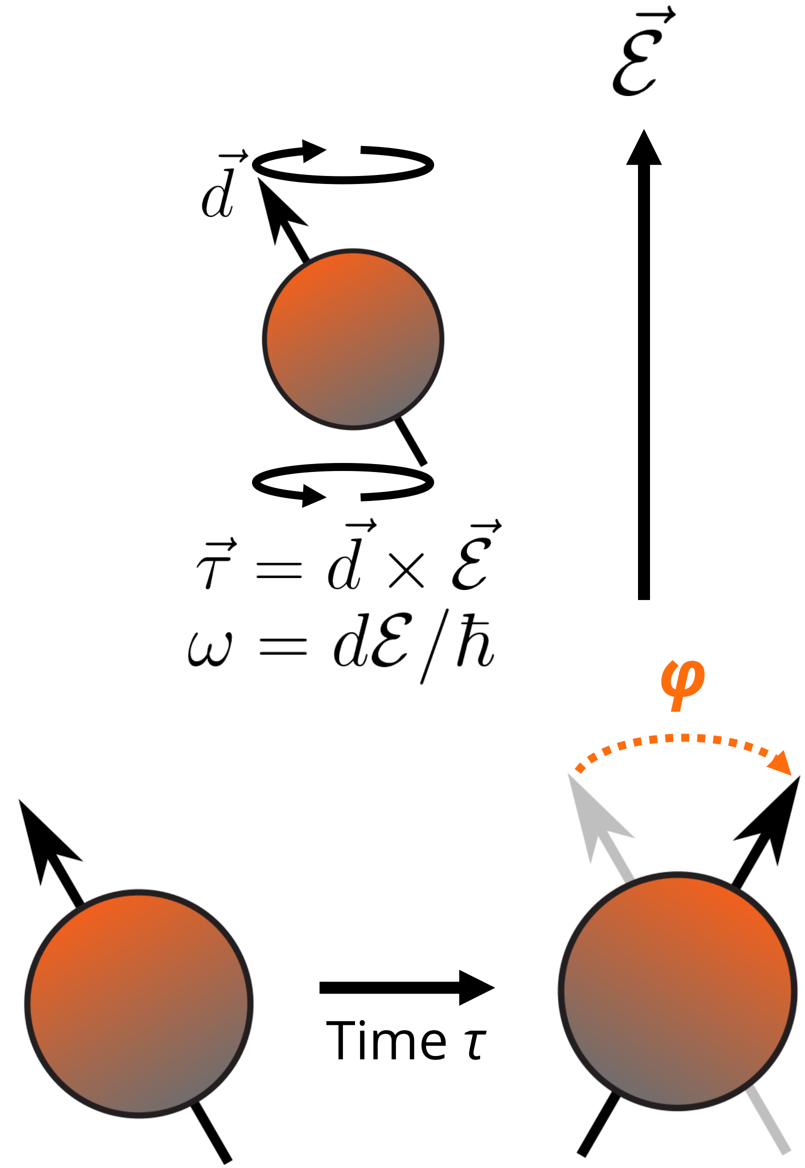
Measuring EDMs

- An EDM experiences a torque in an electric field
- Experiment:
  - Initialize, precess, measure, repeat...

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

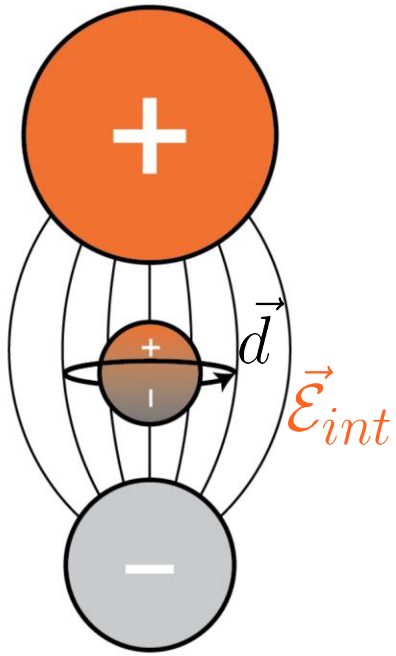
Want large electric field

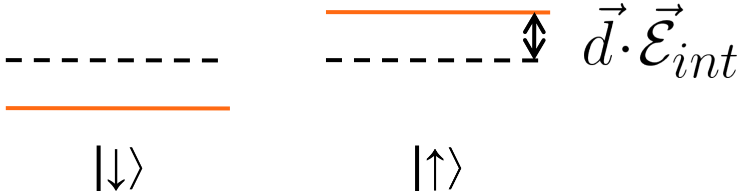
Want long interaction time



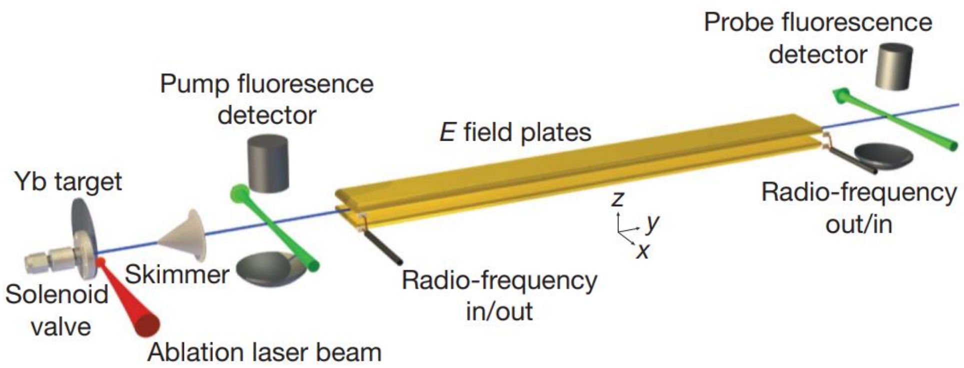
Electric field?

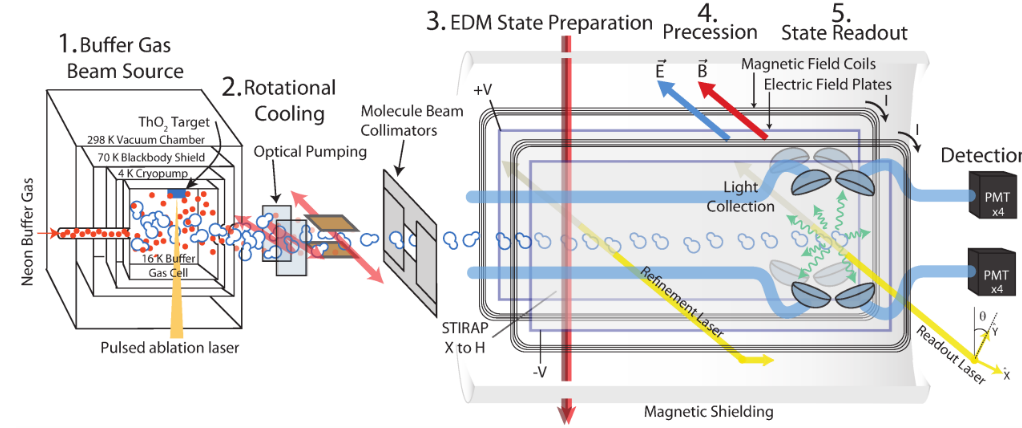
- Atoms/molecules have **extremely large** fields
  - 10-100 GV/cm for heavy species
  - Maximum lab field ~100 kV/cm
- CPV moments cause CPV molecular energy shifts
  - Enhancements for EDM, NSM, and MQM
- Molecular polarizability enhances sensitivity by ~1,000 vs. atoms
  - Atoms set best electron EDM limits until 2011 – molecules are complicated!
  - Atoms still best in other areas… *for now!*





Molecular EDM Searches



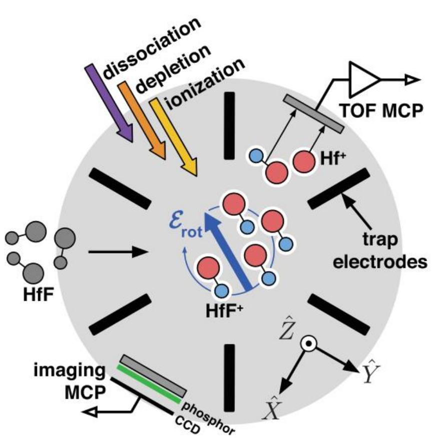


YbF, Imperial

- 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
- Current most sensitive limit
- $|d_e| < 8.7 \times 10^{-29}$ e cm (2014)
- $|d_e| < 1.1 \times 10^{-29}$ e cm (2018)



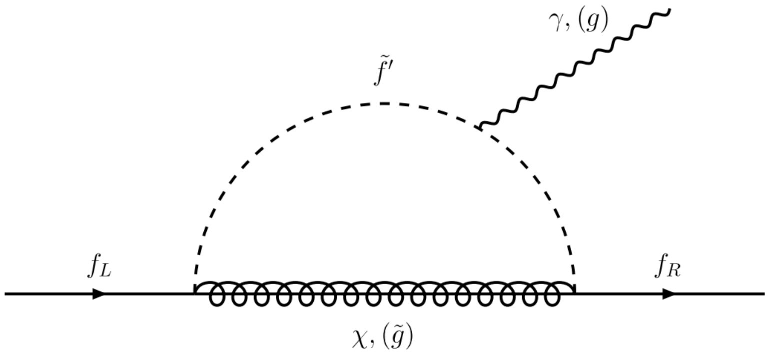
HfF⁺, JILA/Boulder

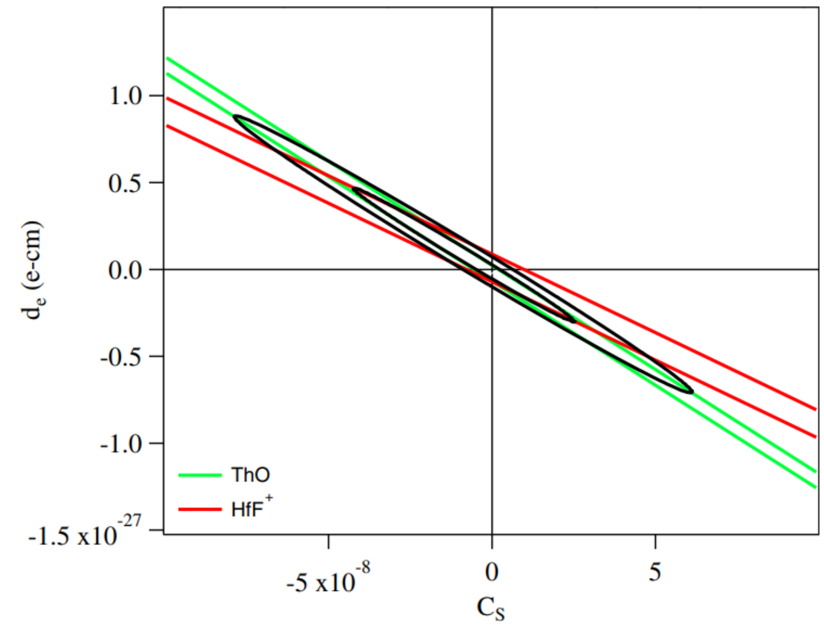
- Spin precession in ion trap
- Long coherence time from trapping
- $|d_e| < 1.3 \times 10^{-28}$ e cm (2017)

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

Interpreting EDM Constraints

- SM "background free"
  - (for several orders of magnitude)
- Generic constraint
  - New particle mass M
  - CPV coupling φ~1
  - M $\gtrsim$ 50 TeV (1 loop), 5 TeV (2 loop)
- Much higher (>PeV) for specific models
- Multiple sources of CPV
  - **Multiple experiments are *needed* to disentangle**
  - Especially true for hadronic CPV searches
  - See 2203.08103



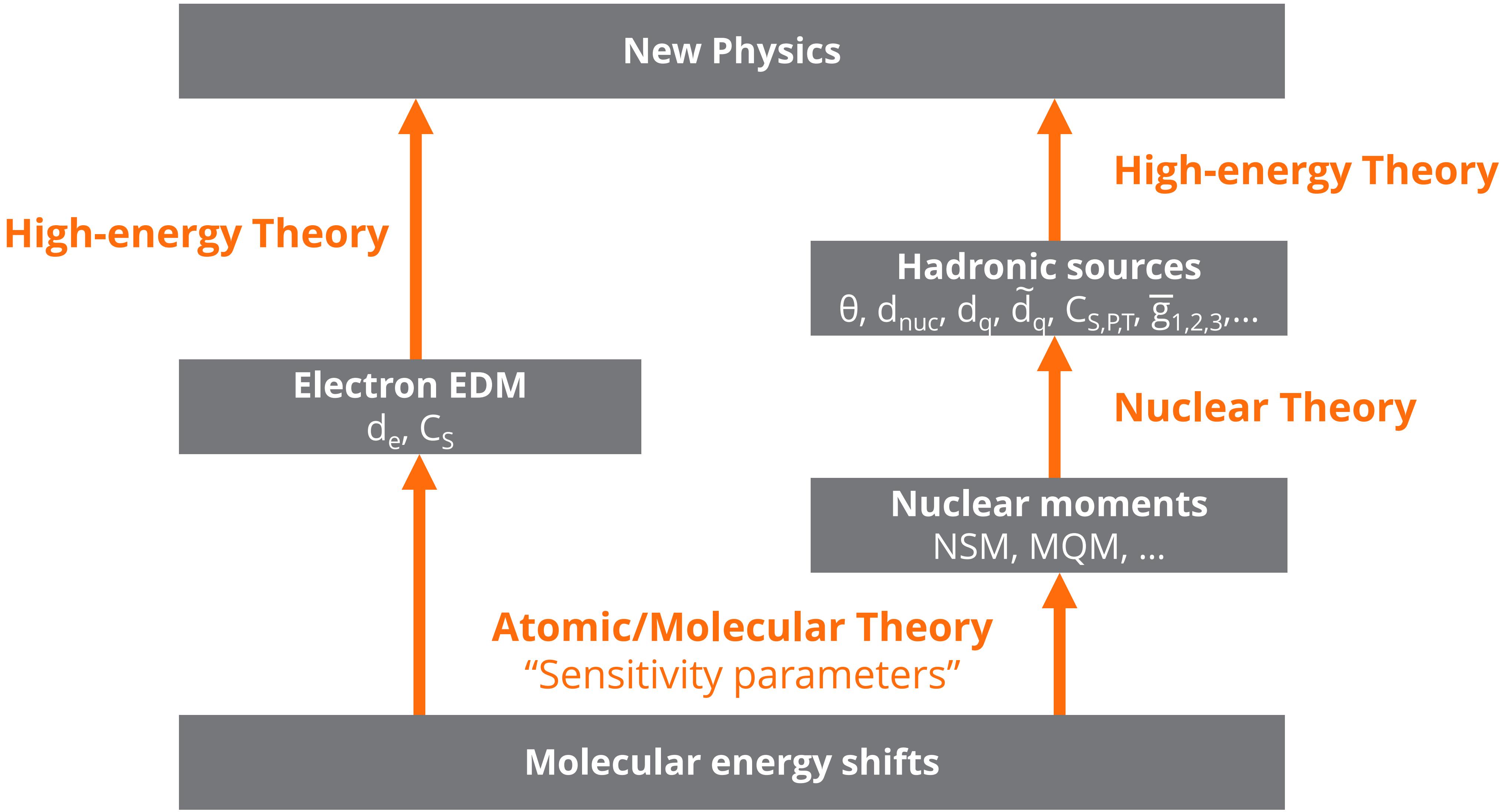


J. Engel, M. J. Ramsey-Musolf, and U. van Kolck, Prog. Part. Nucl. Phys. 71, 21 (2013)
T. E. Chupp, P. Fierlinger, M. J. Ramsey-Musolf, and J. T. Singh, Rev. Mod. Phys. 91, 015001 (2019)
Snowmass 2021 EDM white paper: arXiv:2203.08103 (2022)

New Physics

??

Molecular energy shifts



Many types of theory are critical for these experiments!

This workshop includes leaders in each of these areas

Next-generation tools

Sensitivity

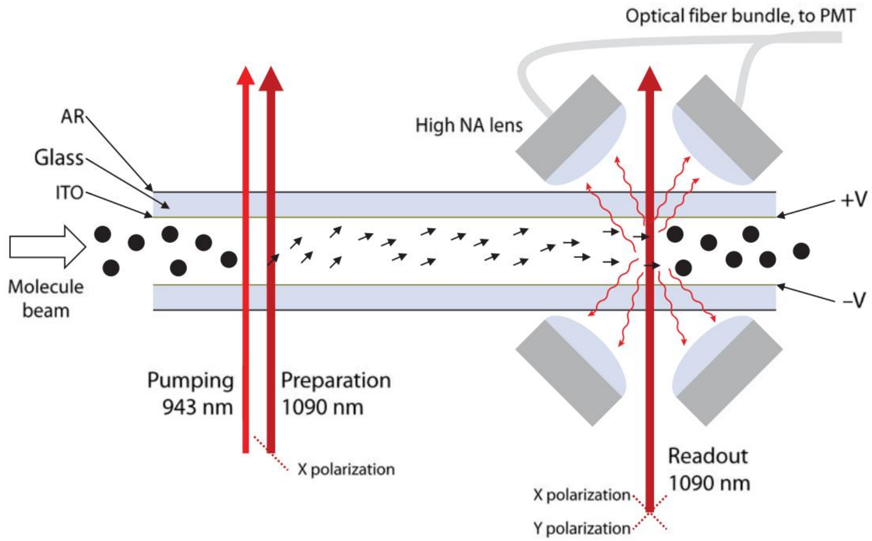
- Sensitivity to new physics scales as
  [Intrinsic sensitivity] × [Coherence time] × [Count rate]$^{1/2}$
- ***Molecular experiments can combine significant enhancements in all of these areas***
  - Orders-of-magnitude improvement for wide range of BSM
  - Leptonic/hadronic CPV, dark matter, parity violation, new forces, weakly-coupled sectors, …
- Will benefit from concurrent advances in other areas
  - Quantum information science (QIS)
    - Same basic requirement: Coherent quantum control
  - Access to short-lived, exotic nuclei
    - Offer further, dramatic increases in sensitivity

Laser cooling

Motivation for laser cooling

- Beam experiments (ThO, YbF) limited by time of flight, few ms
- Can extend by slowing and compressing beam
- Trapping can yield orders of magnitude improvement
  - Critical for long coherence time of HfF^+, Ra experiments
- Neutral species must be "ultracold," <1 mK

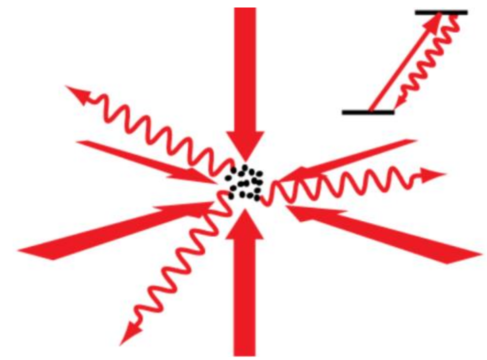
→ **Laser cooling**



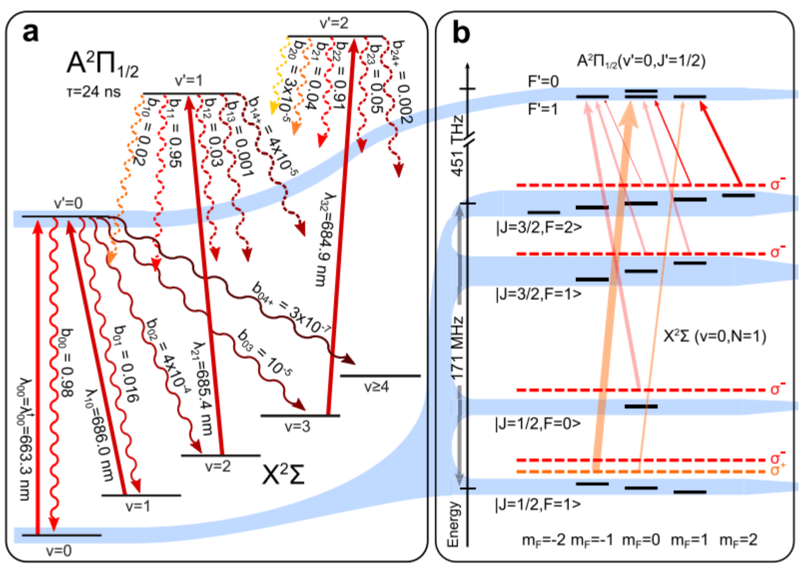
ACME Generation I

Laser cooling/trapping

- Lasers can be used to cool atomic gases to < μK
  - Major driver of AMO, QIS
  - *Extremely advanced*
  - ~10^5 cycles of absorption, spontaneous decay
- Some molecules can be directly laser cooled
  - Complexity → challenging
  - SrF, CaF, YO, YbF, BaF, …
  - Polyatomics (later)
- Can assemble molecules from ultracold atoms
  - Rb, Cs, Ba, Ra, Yb, Hg, …
  - KRb, RbCs, NaCs, NaRb, …
- *Many recent, rapid advances!*



"Atoms"



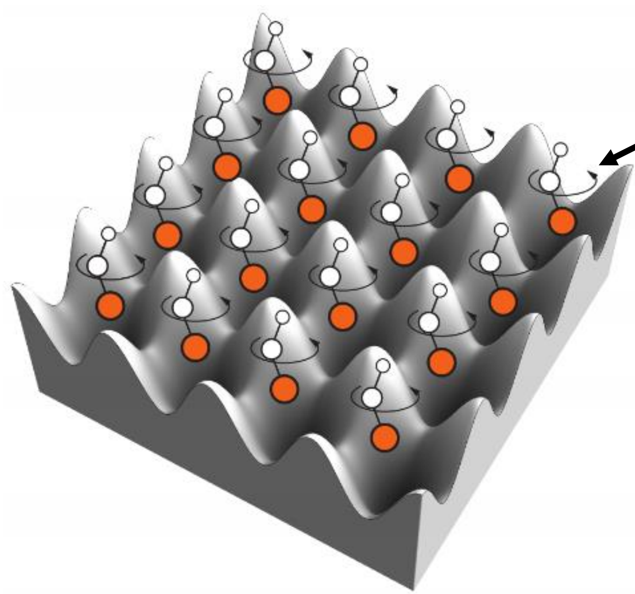
"Molecules"

First molecule MOT: SrF, DeMille Group
J. F. Barry et al, Nature 512, 286 (2014)

Ultracold CPV Searches

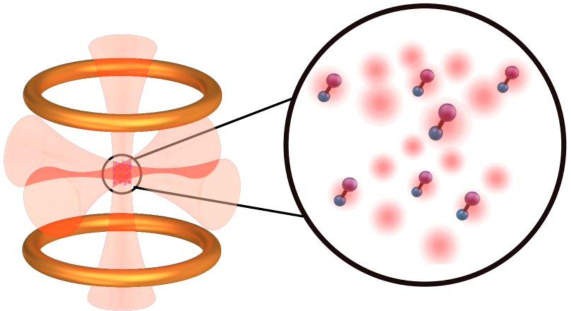
- 10^6 molecules
- 10 s coherence
- Large enhancement(s)
- Robust error rejection
- 1 week averaging

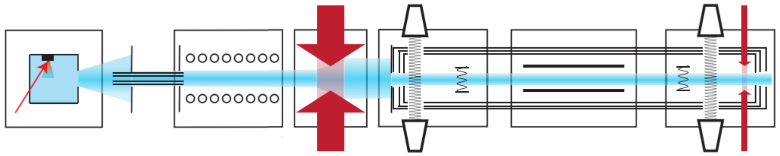
$M_{\text{new phys}} \sim$ **1,000 TeV**

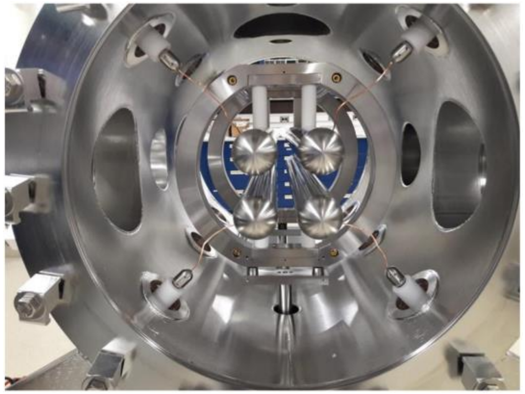


Heavy, polar molecule sensitive to new physics

Three Examples







YbF

- eEDM @ Imperial College London
- Laser cooling, other upgrades demonstrated
- 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
- Advanced deceleration techniques
- P. Aggarwal et al., Eur. Phys. J. D 72, 197 (2018).
- P. Aggarwal *et al.*, PRL 127, 173201 (2021)

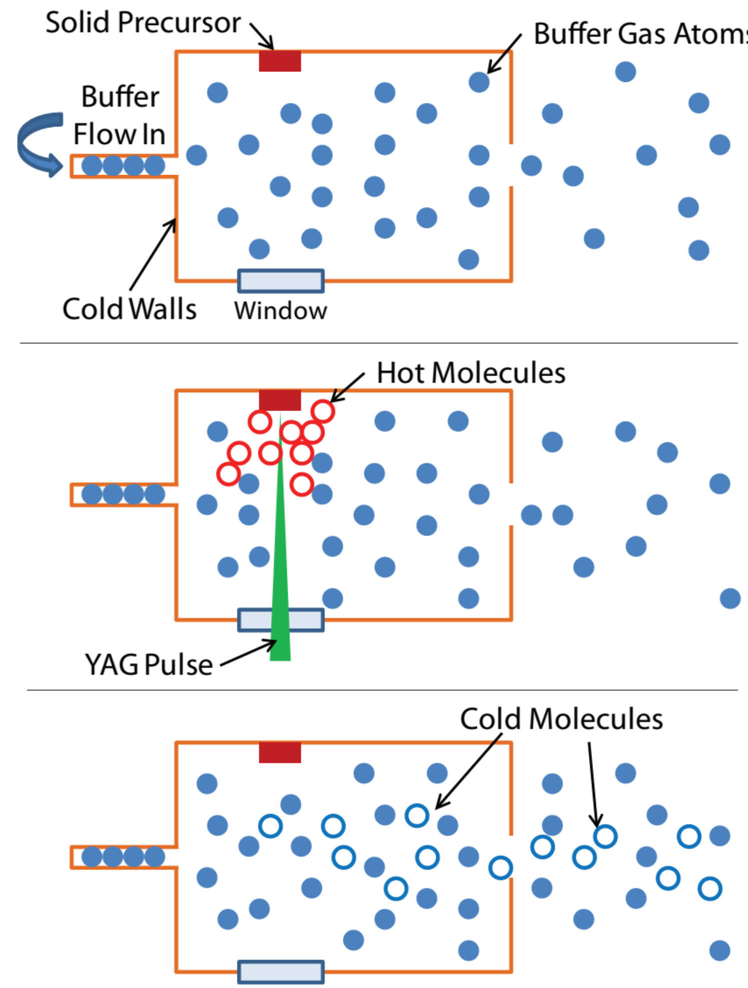
TlF

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

Several more laser cooling examples later

Buffer gas cooling

- These molecules are free radicals with low vapor pressure – challenging
- Use inert gas in cryogenic environment to cool via collisions
  - CBGB – Cryogenic buffer gas beam
- “Works for anything”
- Cold, slow, high flux
- Critical for ACME, all neutral molecule laser cooling/trapping

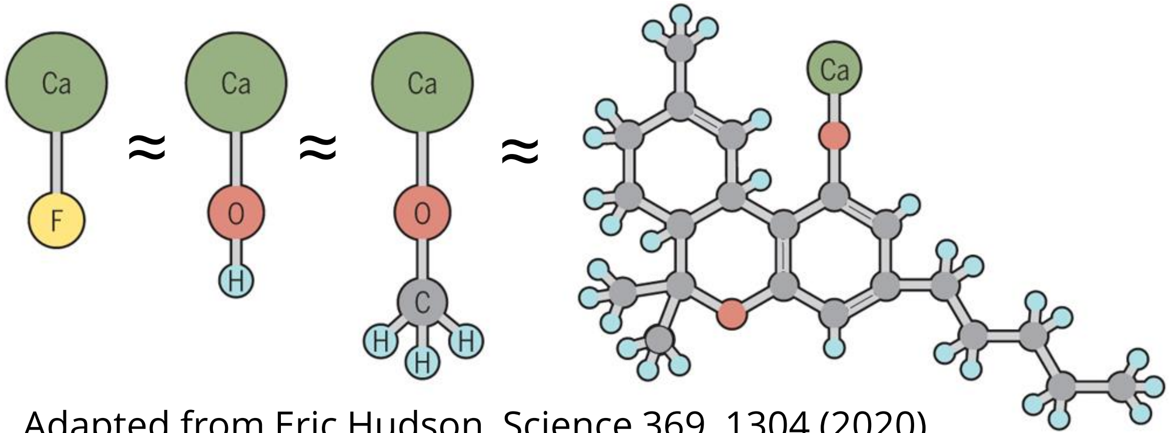


NRH, H. Lu, and J. M. Doyle, Chem. Rev. 112, 4803 (2012)

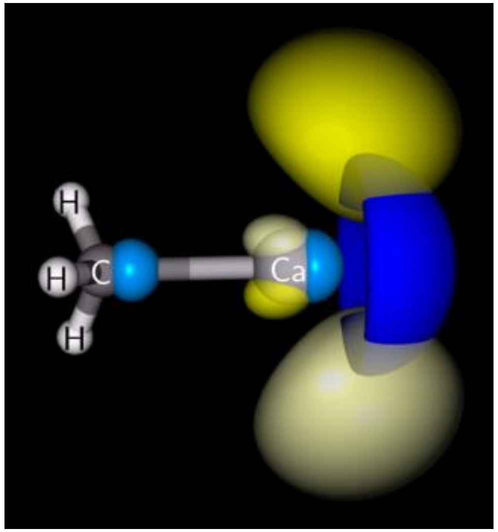
Polyatomic Molecules

Polyatomic Molecules

- Additional degrees of freedom to engineer desirable properties
  - Electric and magnetic field interactions
  - High polarizability (generic parity doublets)
  - Species in ligand
  - Frequencies of rotation and vibration
  - …
- Other desirable properties are often preserved
  - (… with suitable ligand)
  - Laser cooling/photon cycling
  - Intrinsic sensitivity
  - Exotic nuclei
- Review: 2008.03398
  Q. Sci. & Tech. 5, 044011 (2020)

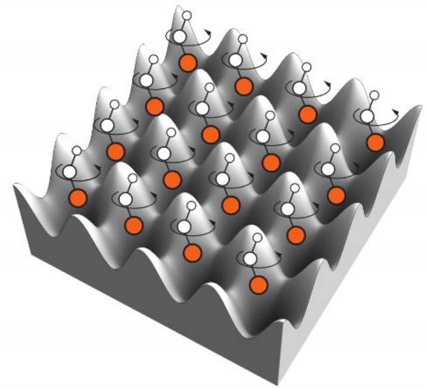


Adapted from Eric Hudson, Science 369, 1304 (2020)



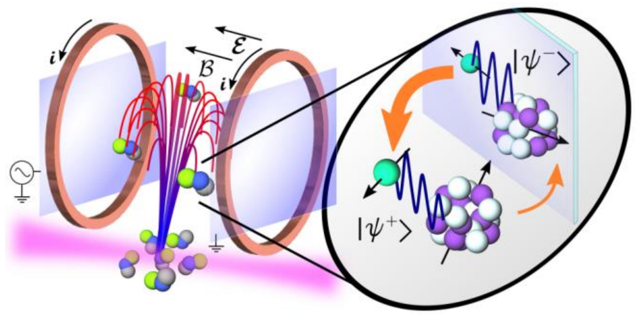
T. A. Isaev and R. Berger
PRL **116**, 063006 (2016)

Three Examples



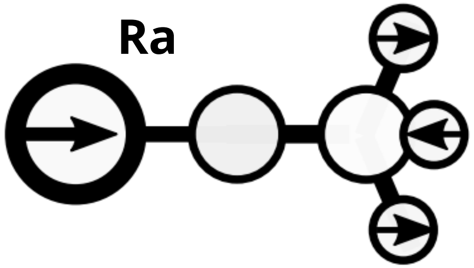
YbOH

- Combine laser cooling, high polarizability
- PolyEDM: NRH, Doyle, Steimle, Vutha
- See *polyedm.com*
- I. Kozyryev and NRH, PRL 119, 133002 (2017)



MgNC

- Engineer magnetic field interactions for PV
- See also Silviu Udrescu talk on Thursday
- E. B. Norrgard, et al, Nat. Comm. Phys. 2, 77 (2019)



$RaOCH_3^+$

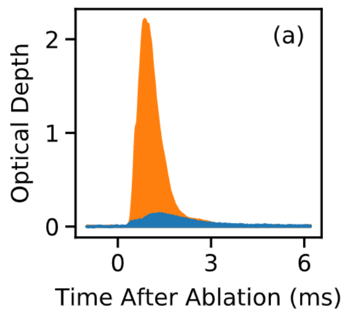
- Combines deformed nucleus with ion trapping
- Created and controlled in Jayich Lab at UCSB
- Other molecules as well, ex. PaF^{3+} [2203.10333]
- M. Fan et al., PRL 126, 023002 (2021)
  P. Yu and NRH, PRL 126, 023003 (2021)

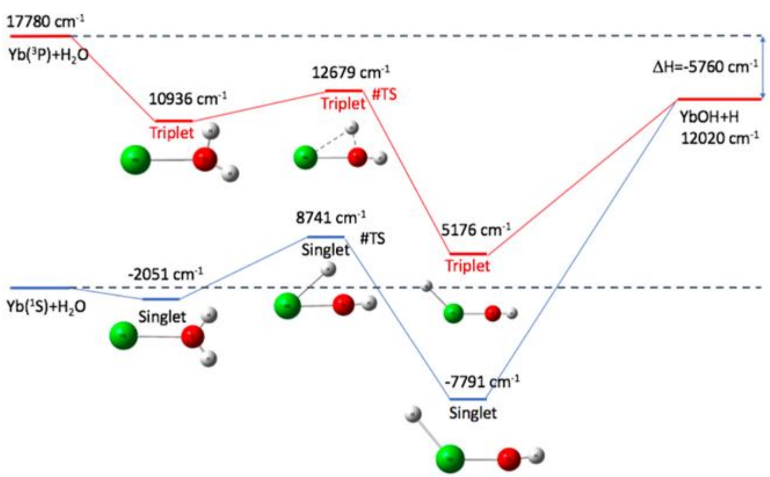
… Many, many more!

Selected (Biased) Advances

Production and spectroscopy

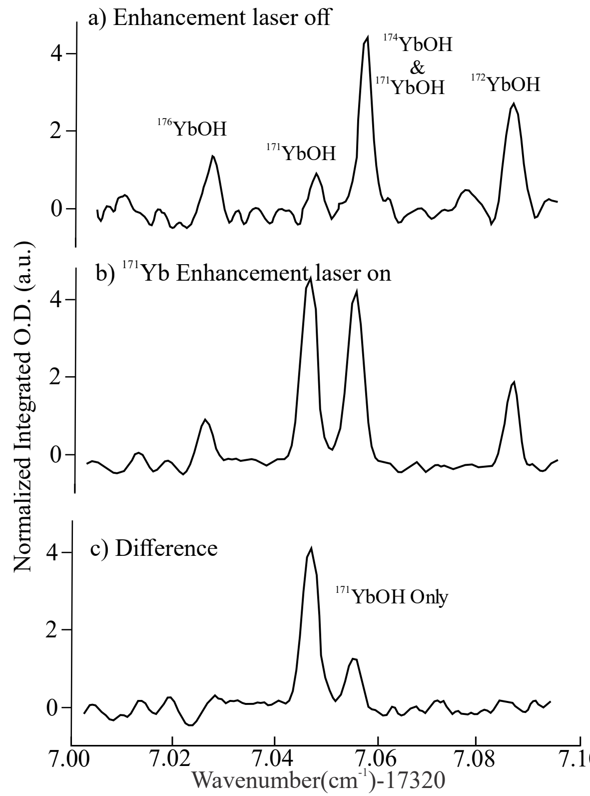
Optically-enhanced chemical production





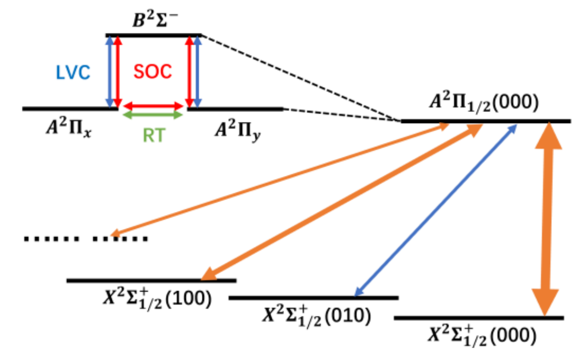
A. Jadbabaie, N. Pilgram, J. Kłos, S. Kotochigova, NRH, New J. Phys. 22, 022002 (2020)

New methods for dense and overlapped spectra



N. H. Pilgram, A. Jadbabaie, Y. Zeng, NRH, T. C. Steimle. J. Chem Phys. 154, 244309 (2021)

High precision/accuracy structure and branching



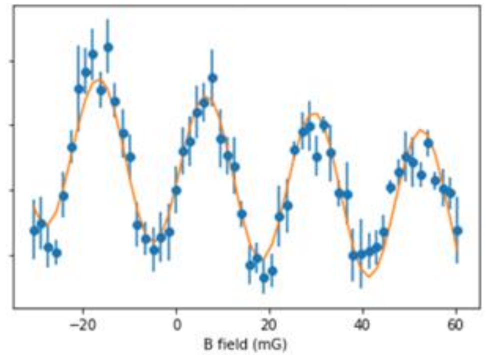
	Level		VBR	
State	Calculated	Experiment	Calculated (%)	Experiment (%)
(000)	0	0	87.691	89.44(61)
(010)	322	319	0.053	0.054(4)
(100)	532	528	10.774	9.11(55)
(02^00)	631	627	0.457	0.335(20)
(02^20)	654	…	0.022	…
(110)	846	840	0.007	0.0100(13)
(03^10)	952	947	<0.001	0.0020(9)
(200)	1062	1052	0.863	0.914(62)
(12^00)	1154	1144	0.063	0.055(4)
(12^20)	1173	…	0.004	…
(210)	1369	1369	<0.001	0.0019(12)
(300)	1600	1572	0.054	0.067(4)
(22^00)	1680	1651	0.007	0.0050(9)
(400)	2160	2079	0.002	0.0045(9)

C. Zhang, B. L. Augenbraun, Z. D. Lasner, N. B. Vilas, J. M. Doyle, and L. Cheng, J. Chem. Phys. 155, 091101 (2021).

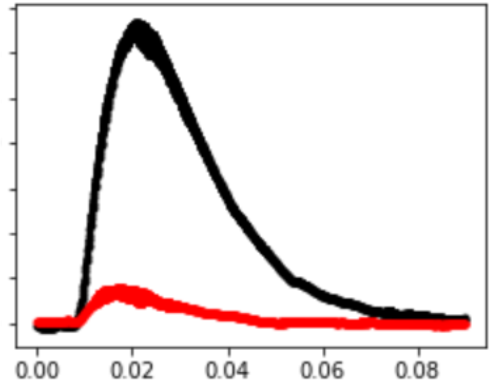
What's new with YbOH?

Recent developments at Caltech

- Coherent spin precession (preliminary)
- Photon cycling in all isotoplogues
  - Odd isotopes complicated by hyperfine structure
- High-resolution spectroscopy
  - Including of science state (below)

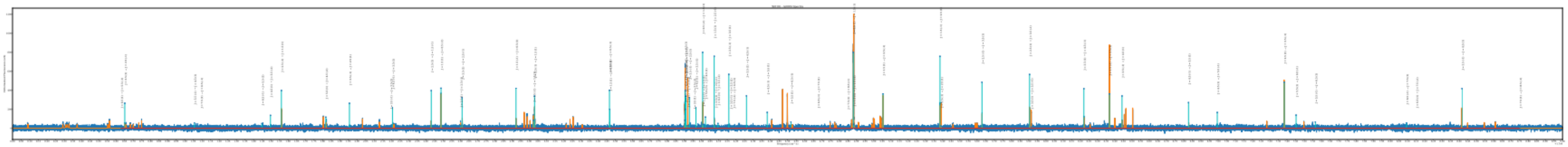


Spin precession



Photon cycling

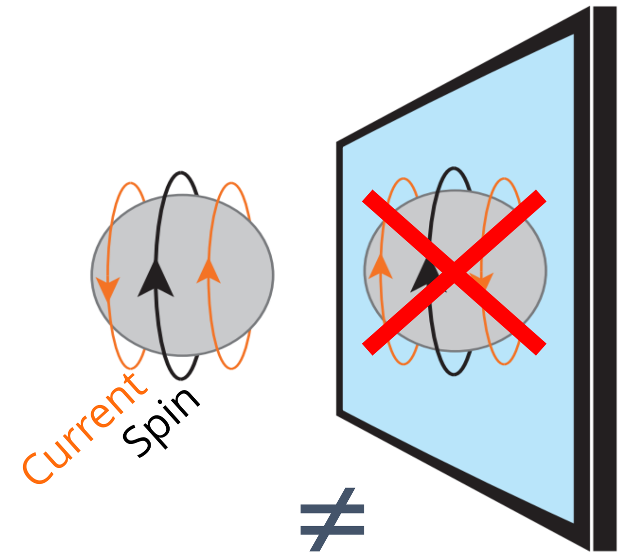
High-resolution spectroscopy

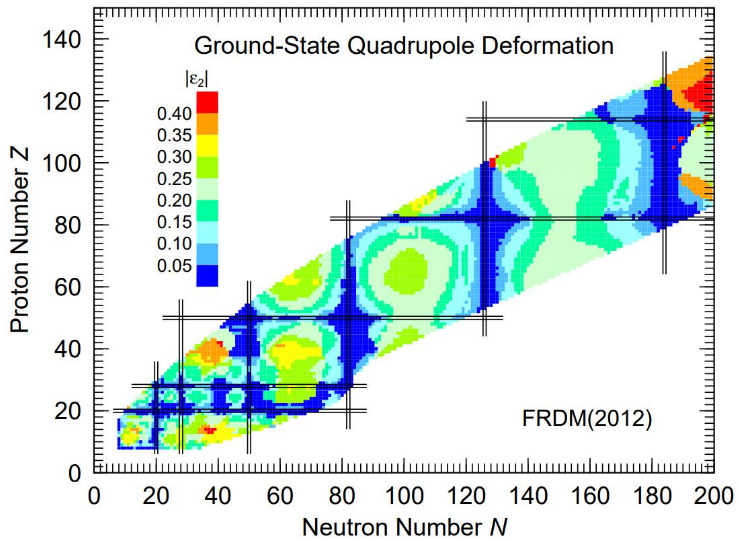


Deformed Nuclei

Hadronic CPV Enhancement

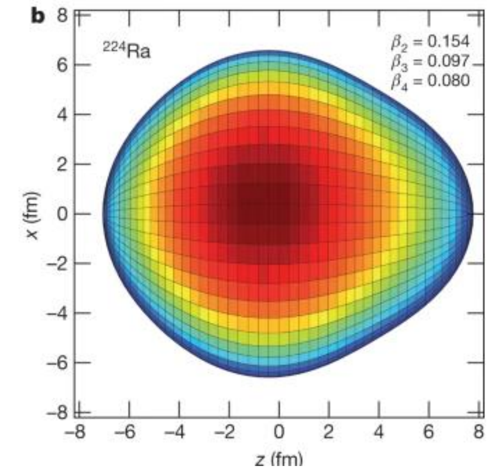
- Quadrupole (β_2) and octupole (β_3) deformations can enhance hadronic CPV
  - θ_{QCD}, chromo-EDMs, nucleon EDMs, CPV forces, …
  - Combines with molecular enhancements
- β_2: Magnetic quadrupole moments (MQMs)
  - Collective enhancement, typically ~10
  - Yb, Ta, Hf, Th, Ra, …
    - V. V. Flambaum, et al., PRL 113, 103003 (2014)
  - Ex: ^{173}YbOH (my lab)



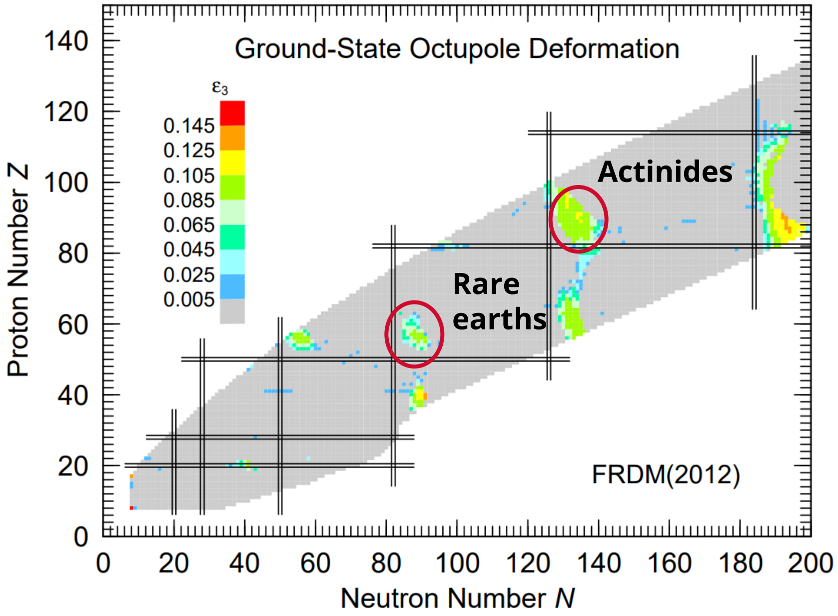


Octupole Deformations

- β_3: Schiff Moments (NSMs) enhanced by ~100-1,000
  - Ra, Ac, Th, …
  - Heavy, spinful, deformed species are short-lived
- Combines with molecular enhancements → 10^{5-6} sensitivity gain vs. atoms with spherical nuclei
  - Hg, Xe (highly advanced experiments, hard to beat)
  - Many CPV sources → need multiple experiments
- Truly exotic nuclei like ^{229}Pa offer *another* factor of 100-1000 (maybe)



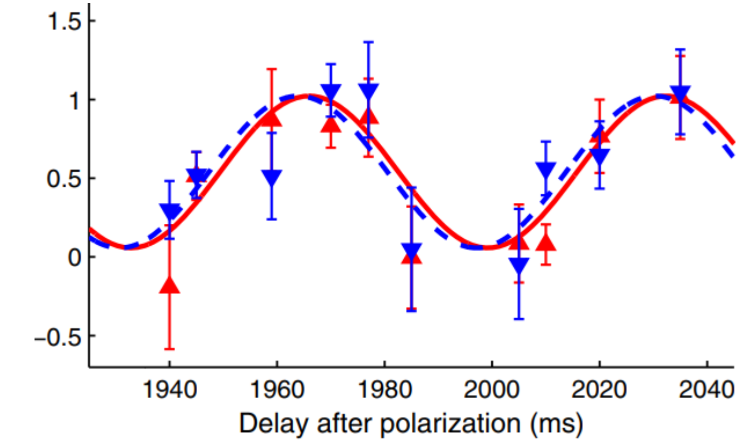
L. P. Gaffney *et al.*, Nature 497, 199 (2013)



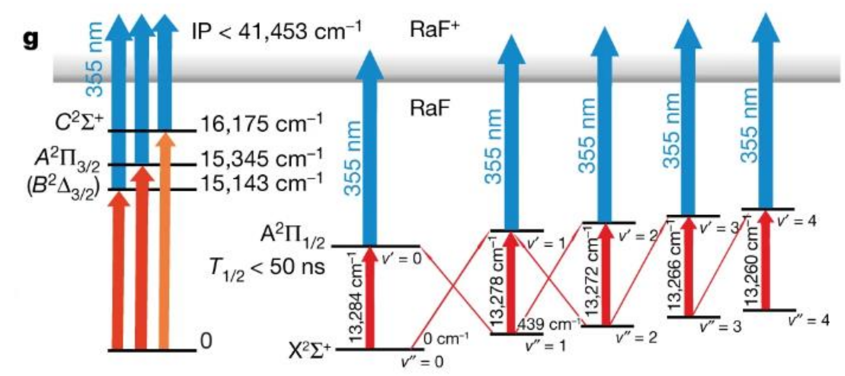
Radium

- Ra is especially interesting!
  - Ra, Ra^+, Ra molecules can be laser cooled
  - Nucleus is well-understood
  - Venue to combine laser cooling, polyatomics, ion trapping, deformed nuclei

Ra	Laser-cooled, trapped EDM experiment @ ANL
RaF	Laser-coolable [Isaev et al., PRA 82, 052521 (2010)] Recent high-resolution spectroscopy [right]
Ra^+	Laser-cooled, trapped, controlled @ UCSB
RaAg	Assemble from laser-coolable atoms See Fleig and DeMille, 2108.02809
$RaOCH_3^+$, $RaOH^+$, $RaSH^+$, …	Trapped, cooled/controlled with co-trapped Ra^+ [M. Fan et al., PRL 126, 023002 (2021)] Single ion could reach frontiers of hadronic CPV [P. Yu and NRH, PRL 126, 023003 (2021)]
RaOH, $RaOCH_3$, …	Laser coolable, high polarizability T. A. Isaev, et al., J. Phys. B 50, 225101 (2017) I. Kozyryev and NRH, PRL 119, 133002 (2017)



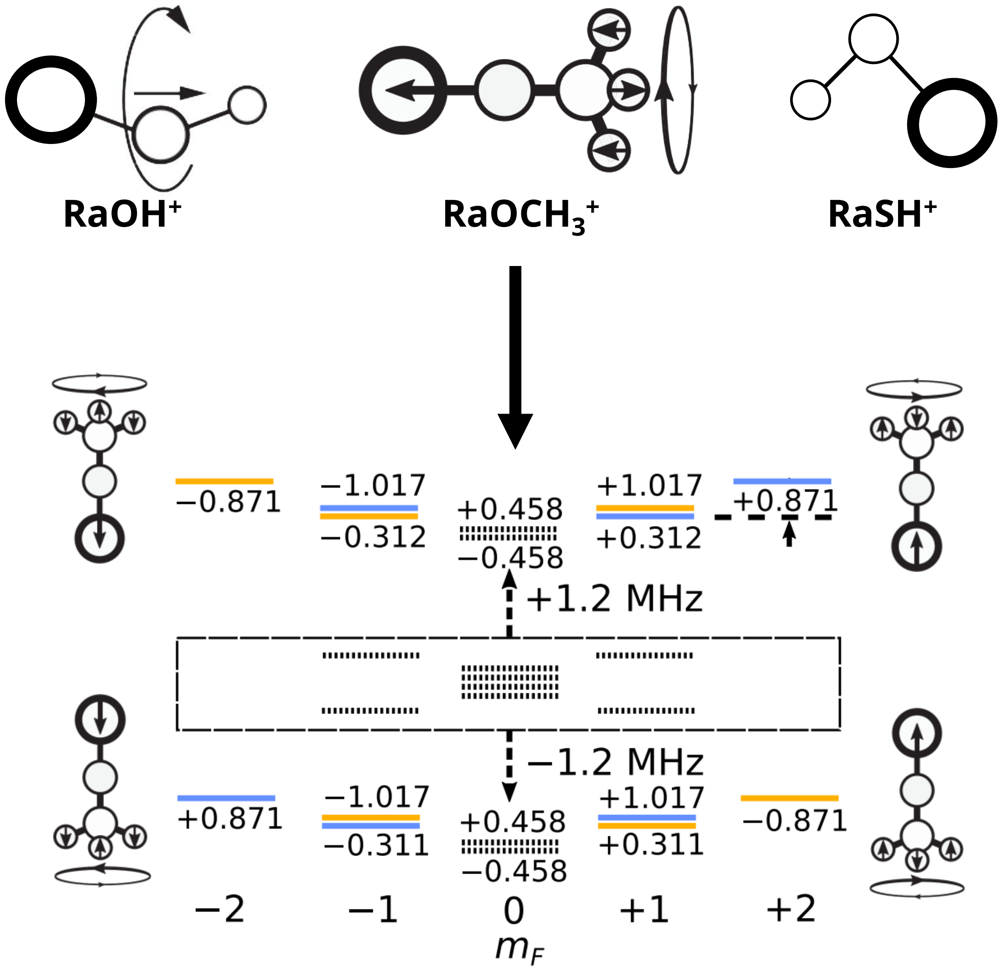
Ra EDM @ ANL
R. H. Parker, et al., PRL 114, 233002 (2015)



High-resolution RaF spectroscopy
R. F. Garcia Ruiz *et al.*, Nature 581, 396 (2020)

Radium Molecular Ions

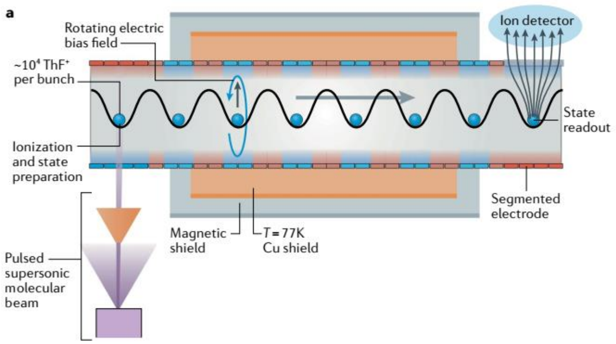
- Radium polyatomics
  - High polarizability due to symmetry-lowering motions
  - Engineer ligand to tune properties, match experimental needs
  - Spin precession, quantum logic, clock transitions, …
- Created, cooled, controlled via co-trapped Ra^+ in Jayich Lab (UCSB)
- A *single* trapped molecular ion could probe the frontiers of symmetry violation

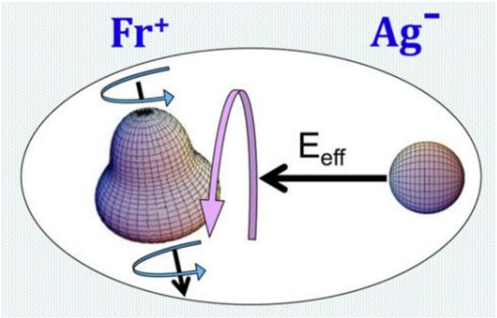


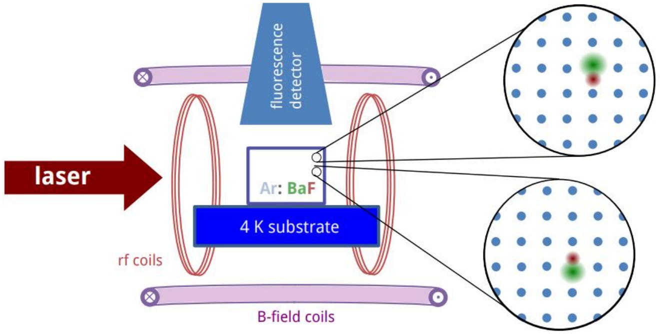
I. Kozyryev and NRH, PRL 119, 133002 (2017)
P. Yu and NRH, PRL 126, 023003 (2021)
M. Fan, C. A. Holliman, X. Shi, H. Zhang, M. W. Straus, X. Li, S. W. Buechele, and A. M. Jayich, PRL 126, 023002 (2021)

Other Directions

Other CPV Approaches







Next-gen ion trapping

- Combine long coherence time with large count rates
- Suitable for eEDM, NSM, MQM
- W. B. Cairncross and J. Ye, Nat. Rev. Phys. **1**, 510 (2019).

Ultracold assembly

- Create ultracold molecules out of ultracold atoms
- Benefits from highly advanced atomic techniques
- RaAg, FrAg promising
- T. Fleig and D. DeMille, arXiv:2108.02809
- J. Kłos *et al.*, New J. Phys. 24 (2022) 025005

Noble gas matrices

- Extreme count rates
- Suitable for molecules and atoms, including rare isotopes
- A. C. Vutha et al., Atoms 6, 3 (2018)
- J. T. Singh, Hyperfine Interact. 240, 29 (2019).

WOULD YOU LIKE TO KNOW MORE?

- **Precision measurements in atoms/molecules**
  - M. S. Safronova et al., Rev. Mod. Phys. 90, 025008 (2018)
  - N. R. Hutzler, Quantum Sci. Technol. 5, 044011 (2020)
- **EDMs**
  - T. E. Chupp, et al., Rev. Mod. Phys. 91, 015001 (2019)
  - W. B. Cairncross and J. Ye, Nat. Rev. Phys. 1, 510 (2019)
  - Snowmass 2021 EDM white paper: arXiv:2203.08103
- **Interpretation of EDM limits**
  - See Safronova, Chupp, Snowmass reviews
  - J. Engel et al., Prog. Part. Nucl. Phys. 71, 21 (2013)
- **Many other talks in this workshop!**

Caltech

Hutzler Lab
Fall 2021

Collaborators

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

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

Photon Cycling: Anastassia Alexandrova (UCLA), Wesley Campbell (UCLA), Justin Caram (UCLA), John M. Doyle (Harvard), Eric Hudson (UCLA), Anna Krylov (USC)

Come visit... some time!

www.hutzlerlab.com
www.polyedm.com

GORDON AND BETTY MOORE FOUNDATION
ALFRED P. SLOAN FOUNDATION
HEISING-SIMONS FOUNDATION
NIST
U.S. DEPARTMENT OF ENERGY Office of Science
NSF