Experimental Perspectives on Fundamental Physics with Molecules

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Outline

- Electron EDM experiments with molecules
  - Prototypical example of modern, rapidly-evolving experiments with molecules
  - ~100x improvement on limit in past ~10 years
- Next-generation tools
  - Molecules offer orders-of-magnitude improvements in multiple sectors through multiple avenues

EDM Experiments with Molecules

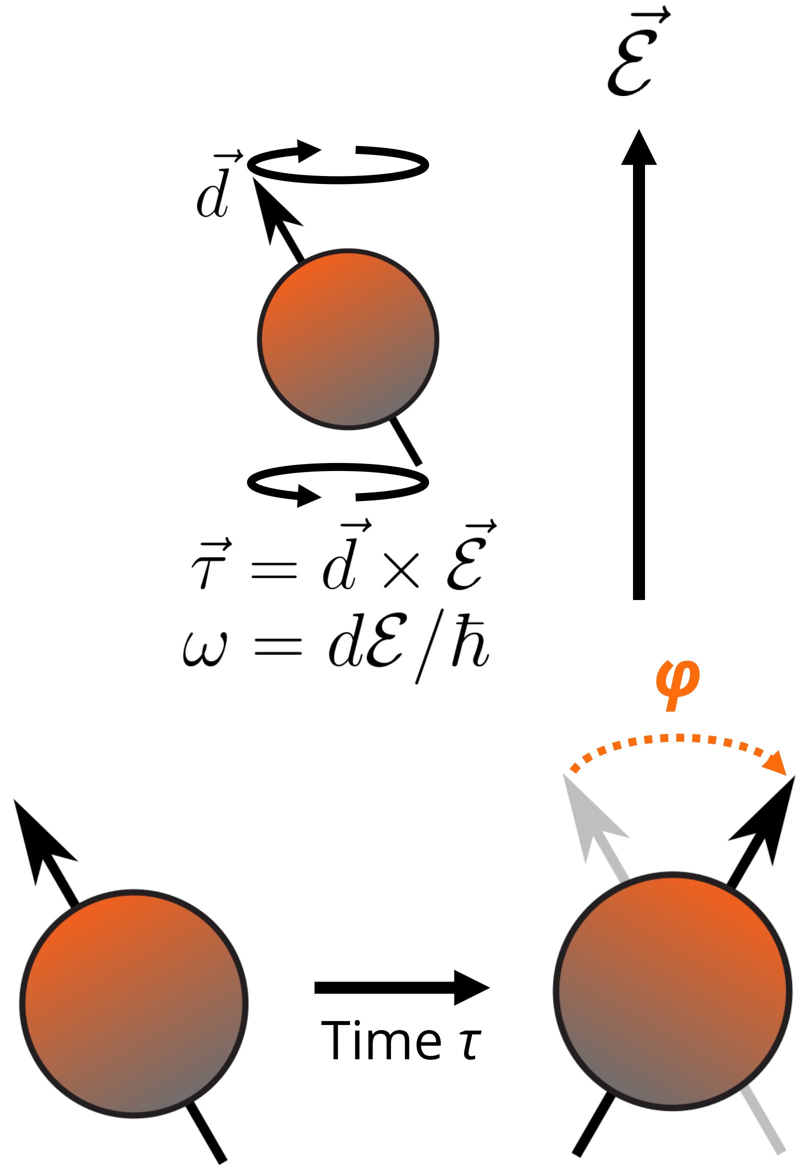
Measuring EDMs

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

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

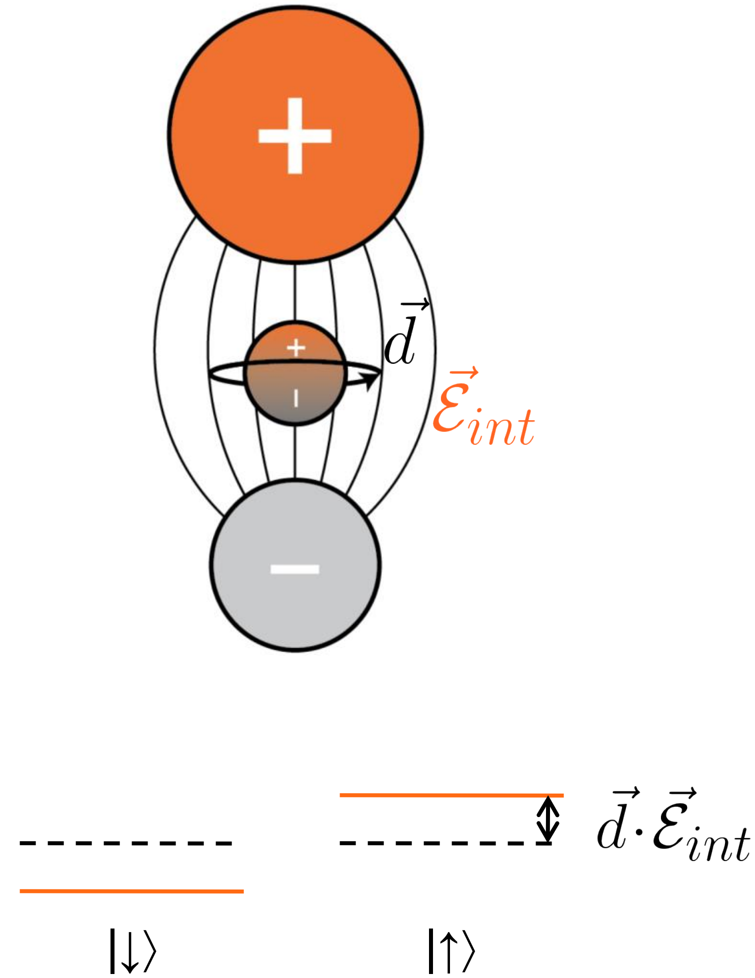
Want large electric field (↑ $\mathcal{E}$)

Want long interaction time (↑ τ)

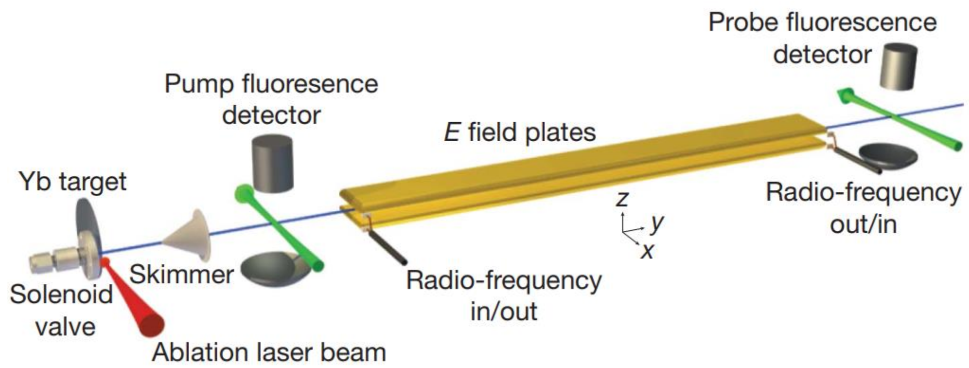


Electric field?

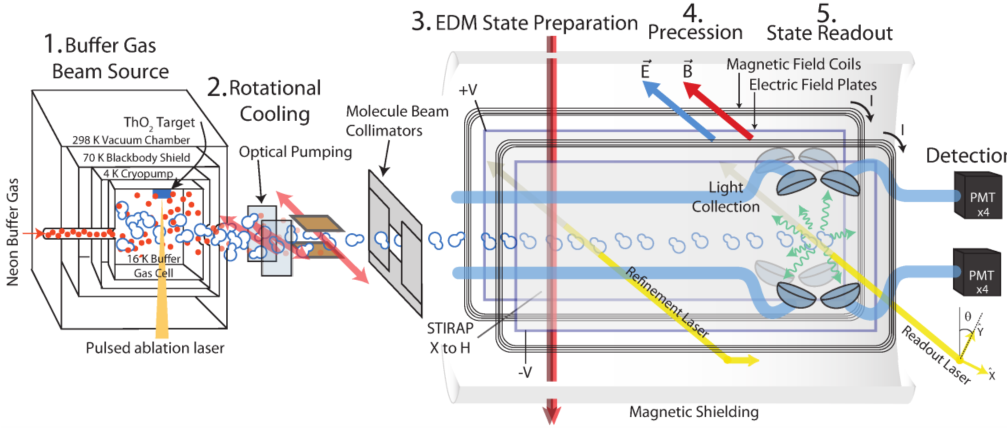
- Atoms/molecules have **extremely large** fields
  - 10-100 GV/cm for heavy species
  - Maximum lab field ~100 kV/cm
- Permanent EDM causes symmetry-violating energy shifts
- Molecular polarizability enhances sensitivity by ~1,000 vs. atoms
  - Atoms set best limits until 2011 – molecules are complicated!
  - Atoms still best in many areas… *but watch your back!*



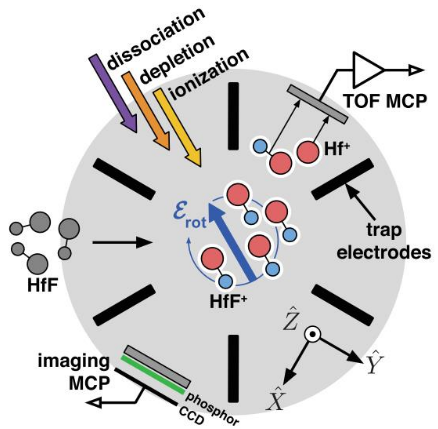
Atom smashers



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/Yale/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)

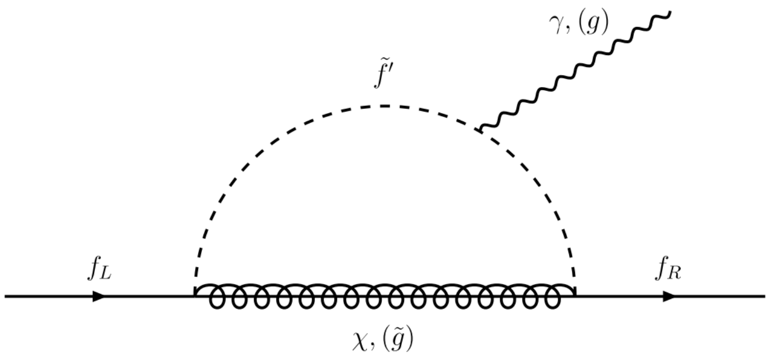


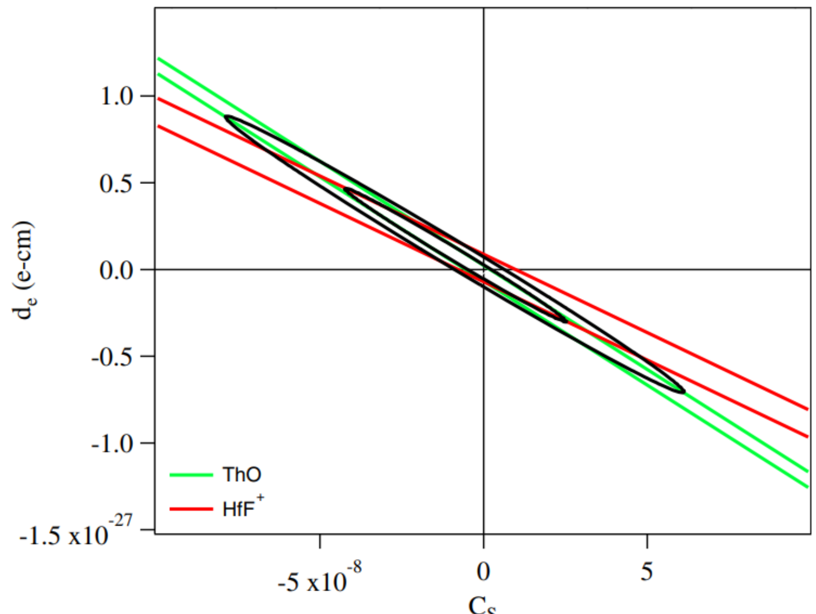
HfF^+, JILA/Boulder
- Spin precession in ion trap
- Long coherence time from trapping
- $|d_e| < 1.3 \times 10^{-28}$ e cm (2017)

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

Interpreting EDM Constraints

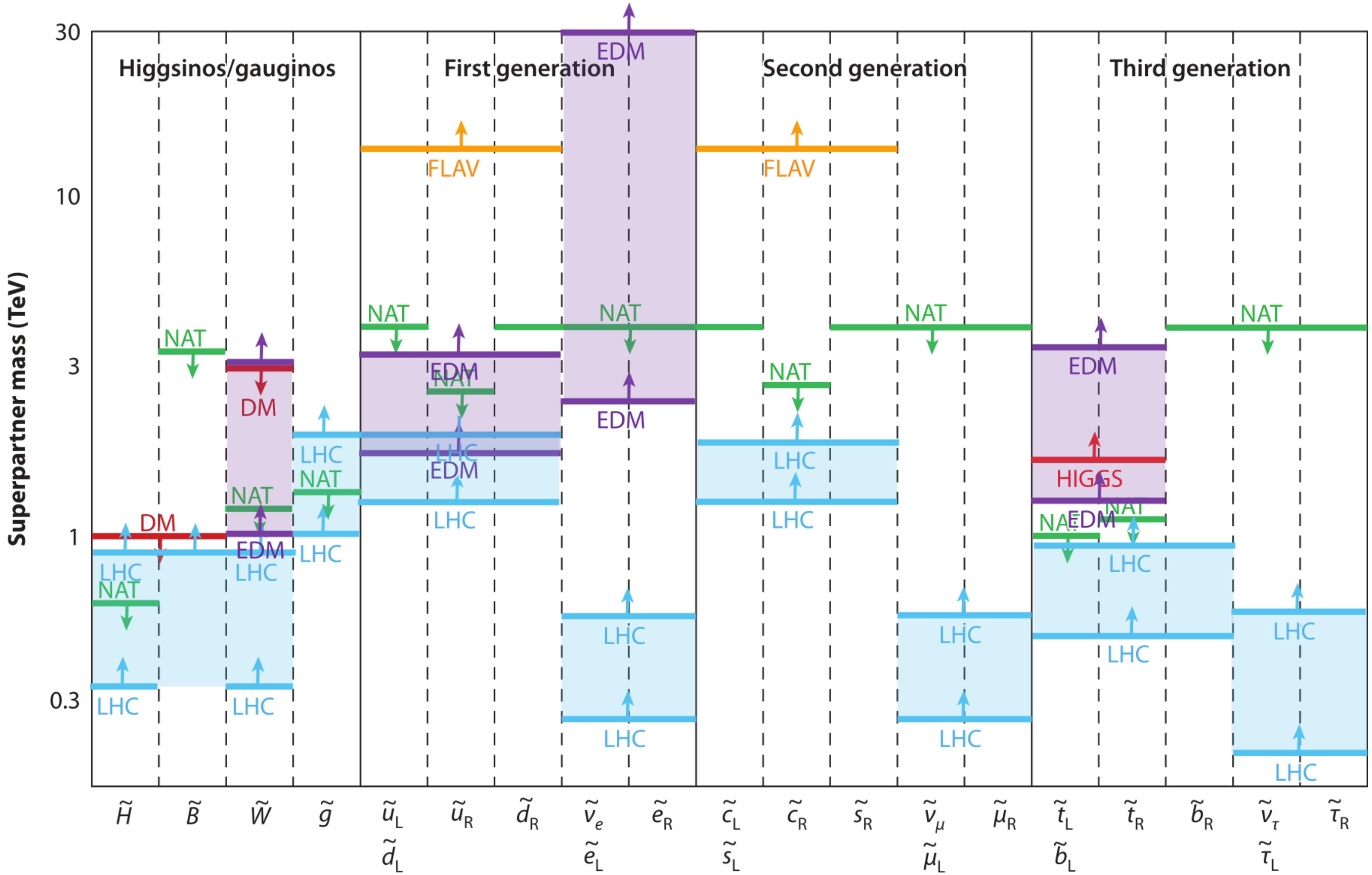
- SM background free
- Generic constraint
  - New particle mass M
  - CPV coupling φ~1
  - M $\gtrsim$ 30 TeV
    - ~3 TeV for 2 loops
- Much higher (>PeV) for specific models
- Multiple sources of CPV
  - **Multiple experiments are *needed* to disentangle**
  - Especially true for hadronic CPV searches





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)

Many complementary approaches



Shading shows progress since 2013 (LHC, ACME, 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

Next-generation tools

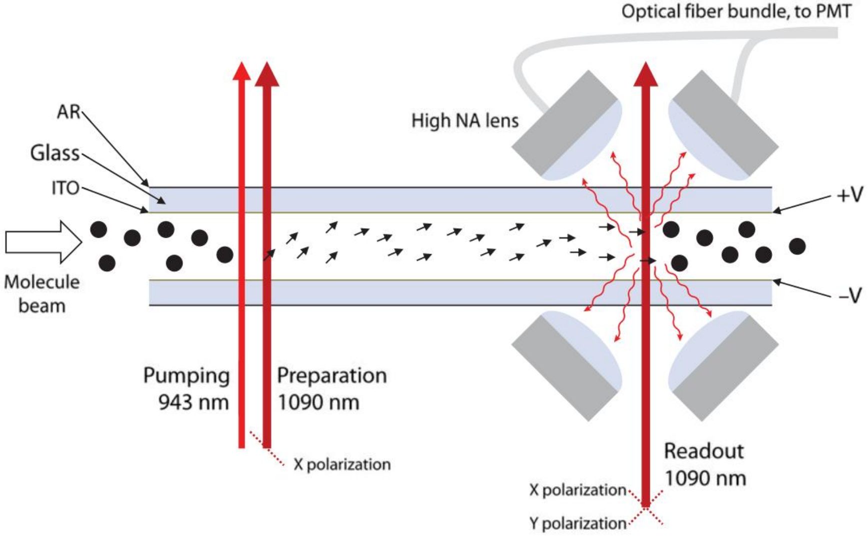
Sensitivity

- 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, …
- Highly symbiotic with quantum information science (QIS)
  - Same requirement: Coherent quantum control
  - Huge and active field (that I won't talk about)
- We will frame our discussion largely around EDMs, but the experimental advances will have broad applicability
- Our focus: new approaches and new systems

Laser cooling

Motivation for laser cooling

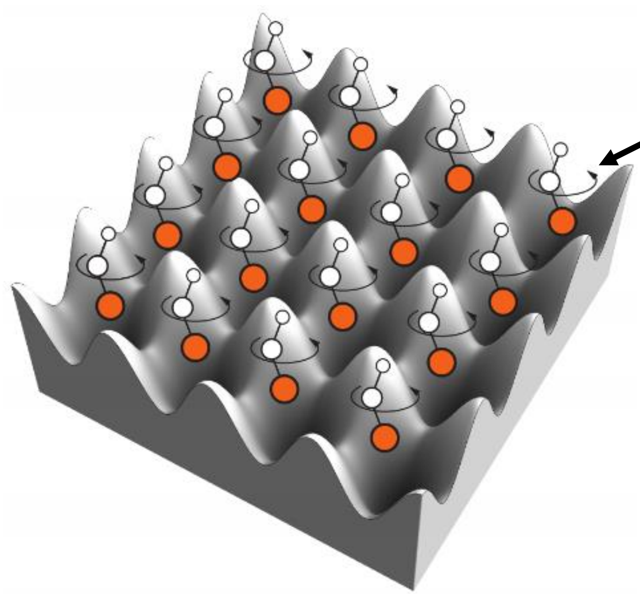
- Beam experiments (ThO, YbF) limited by time of flight, τ ~ few ms
- Can extend by slowing and compressing beam
- Trapping can yield orders of magnitude improvement
  - Critical for long coherence time of HfF^{+}, Ra experiments
- For neutral species, requires ultra-cold temperatures <1 mK
  - Suitable conservative traps are shallow
  - Free molecules (fountains) must be very slow
- → Laser cooling



Ultracold CPV Searches

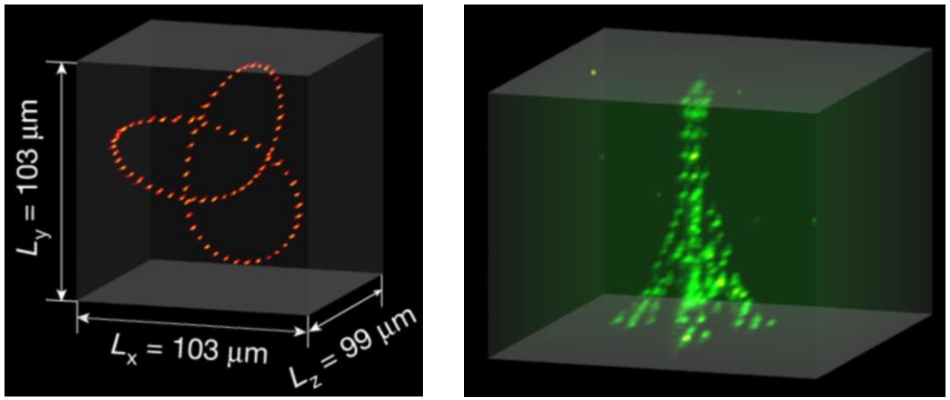
- 10^6 molecules
- 10 s coherence
- Large enhancement(s)
- Robust error rejection
- 1 week averaging

$M_{new\ phys}$ ~ 1,000 TeV

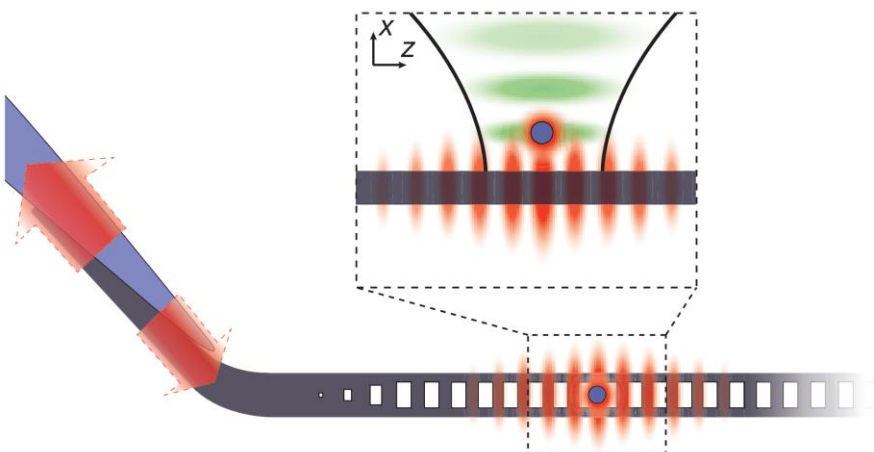


Heavy, polar molecule sensitive to new physics

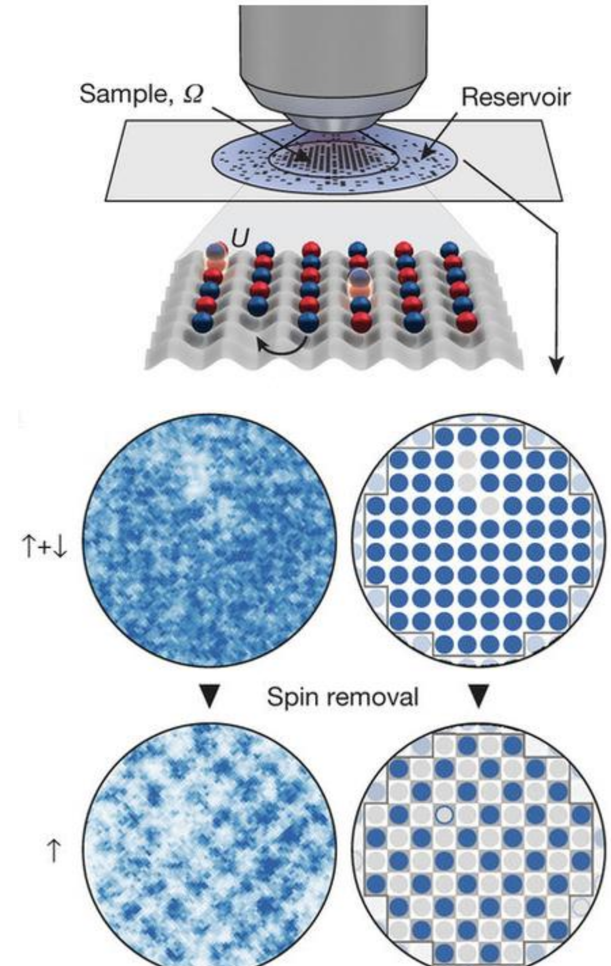
Quantum Control with Atoms



D. Barredo *et al*., Nature **561**, 79–82 (2018)



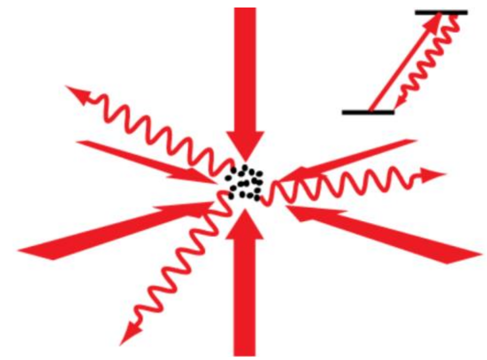
T. G. Tiecke, *et al.*, Nature **508**, 241 (2014).



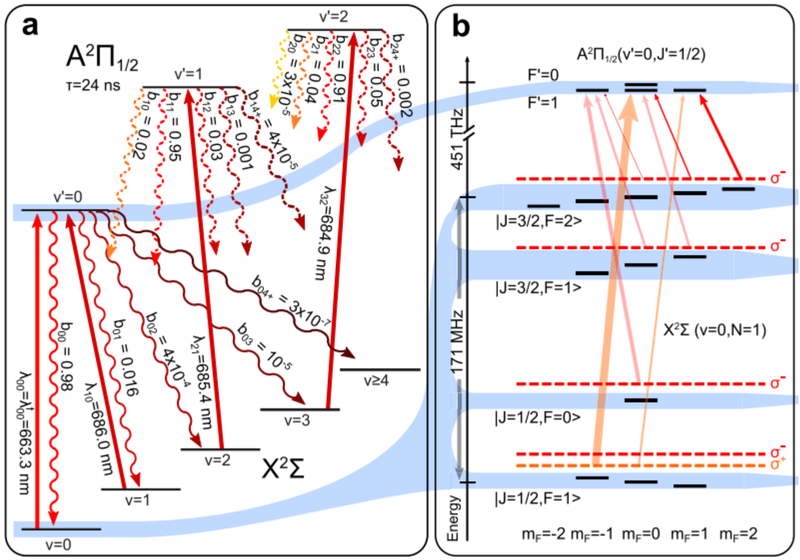
A. Mazurenko *et al.*, Nature 545, 462-466 (2017)

Laser cooling/trapping

- Lasers can be used to cool atomic gases to < μK
  - Major driver of AMO, QIS
  - ~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!*



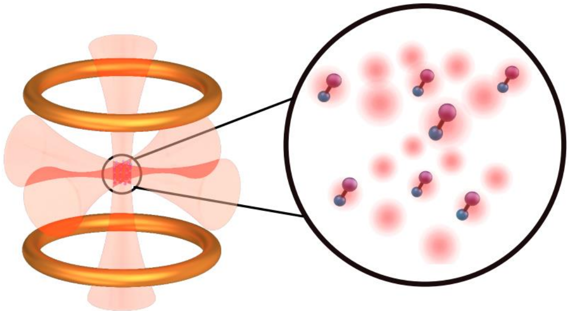
“Atoms”

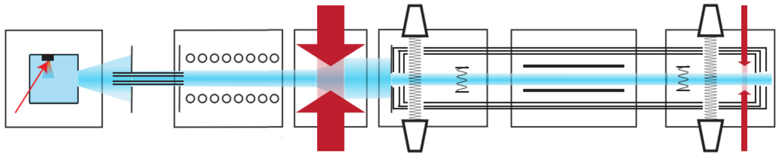


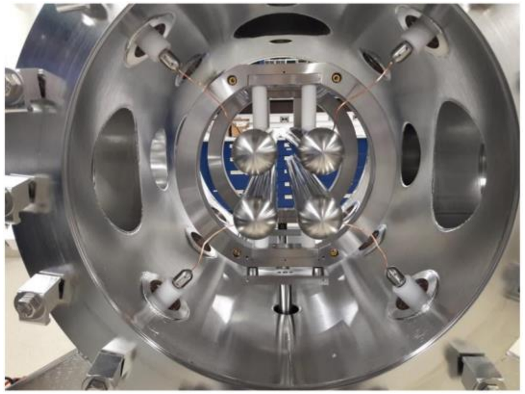
“Molecules”

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

Three Examples







YbF

- eEDM @ Imperial College London
- Laser cooling demonstrated
- N. J. Fitch et al., 2009.00346 (2020)

BaF

- NL-eEDM Collaboration
- Advanced deceleration techniques
- P. Aggarwal et al., Eur. Phys. J. D 72, 197 (2018).

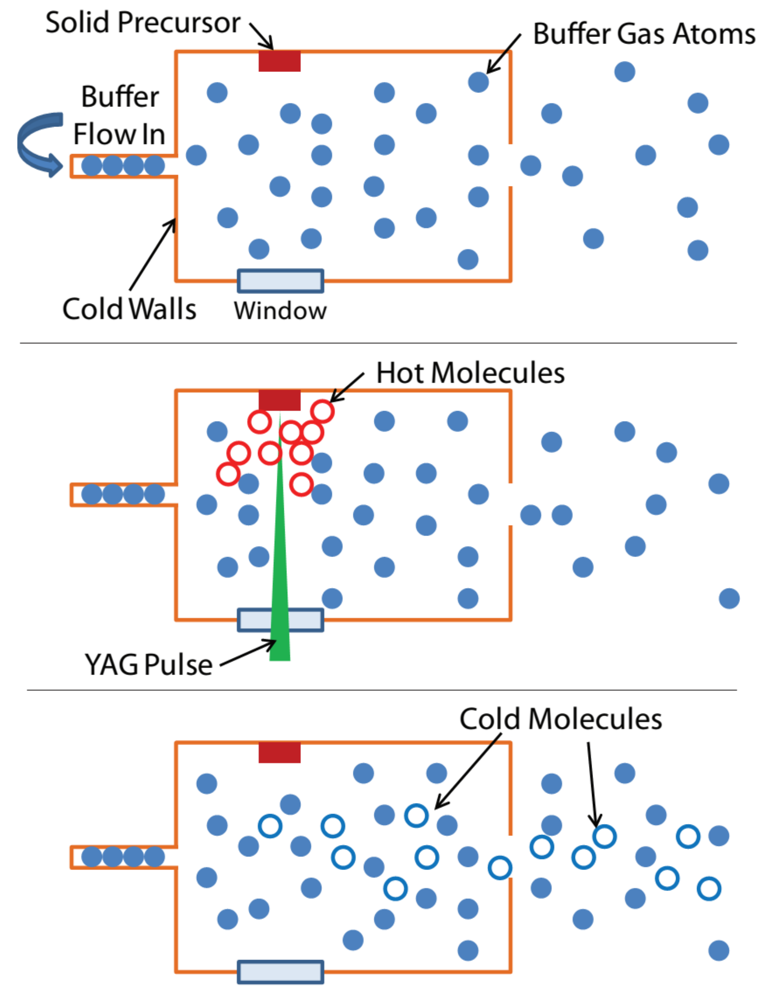
TlF

- CeNTREX Collaboration
- Tl Schiff moment (~proton edm)
- O. Grasdijk et al., 2010.01451 (2020)

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)

Laser-coolable species

- Either directly, or in a molecule
- Incomplete, and will continue to grow!

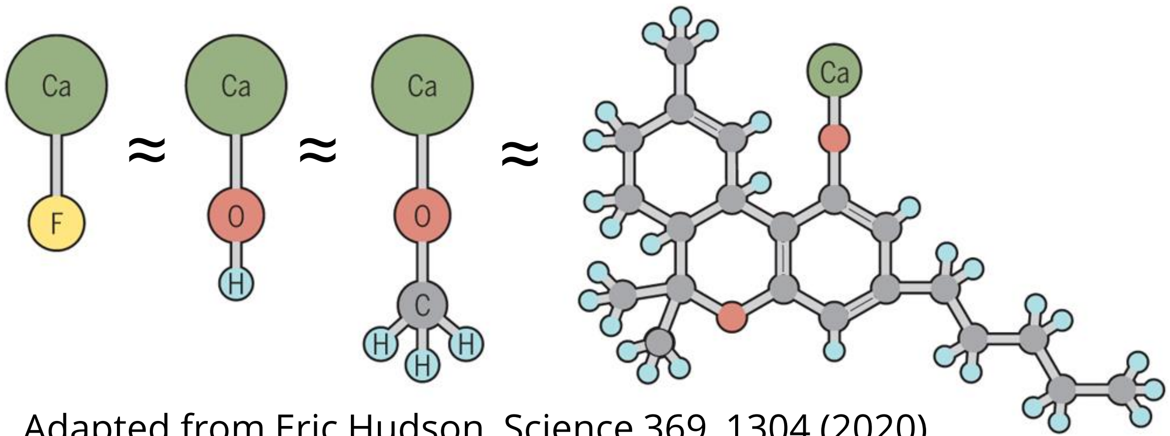
1 H Hydrogen 1																	**4 He Helium 2**
7 Li Lithium 3	**9 Be Beryllium 4**											**11 B Boron 5**	**12 C Carbon 6**	**14 N Nitrogen 7**	**16 O Oxygen 8**	**19 F Fluorine 9**	**20 Ne Neon 10**
23 Na Sodium 11	**24 Mg Magnesium 12**											**27 Al Aluminium 13**	**28 Si Silicon 14**	31 P Phosphorus 15	**32 S Sulphur 16**	**35.5 Cl Chlorine 17**	**40 Ar Argon 18**
39 K Potassium 19	**40 Ca Calcium 20**	45 Sc Scandium 21	**48 Ti Titanium 22**	51 V Vanadium 23	**52 Cr Chromium 24**	55 Mn Manganese 25	56 Fe Iron 26	59 Co Cobalt 27	59 Ni Nickel 28	63.5 Cu Copper 29	65.4 Zn Zinc 30	**70 Ga Gallium 31**	73 Ge Germanium 32	75 As Arsenic 33	79 Se Selenium 34	**80 Br Bromine 35**	**84 Kr Krypton 36**
85 Rb Rubidium 37	**88 Sr Strontium 38**	**89 Y Yttrium 39**	91 Zr Zirconium 40	93 Nb Niobium 41	96 Mo Molybdenum 42	99 Tc Technetium 43	101 Ru Ruthenium 44	103 Rh Rhodium 45	106 Pd Palladium 46	**108 Ag Silver 47**	**112 Cd Cadmium 48**	**115 In Indium 49**	119 Sn Tin 50	122 Sb Antimony 51	128 Te Tellurium 52	**127 I Iodine 53**	**131 Xe Xenon 54**
133 Cs Caesium 55	**137 Ba Barium 56**	57-71	178 Hf Hafnium 72	181 Ta Tantalum 73	184 W Tungsten 74	186 Re Rhenium 75	190 Os Osmium 76	192 Ir Iridium 77	195 Pt Platinum 78	197 Au Gold 79	**201 Hg Mercury 80**	**204 Tl Thallium 81**	207 Pb Lead 82	209 Bi Bismuth 83	210 Po Polonium 84	210 At Astatine 85	222 Rn Radon 86
223 Fr Francium 87	**226 Ra Radium 88**	89-103	267 Rf Rutherfordium 104	268 Db Dubnium 105	269 Sg Seaborgium 106	270 Bh Bohrium 107	277 Hs Hassium 108	278 Mt Meitnerium 109	281 Ds Darmstadtium 110	282 Rg Roentgenium 111	285 Cn Copernicium 112	286 Nh Nihonium 113	289 Fl Flerovium 114	290 Mc Moscovium 115	293 Lv Livermorium 116	294 Ts Tennessine 117	294 Og Oganesson 118

139 La Lanthanum 57	140 Ce Cereum 58	141 Pr Praseodymium 59	144 Nd Neodymium 60	147 Pm Promethium 61	150 Sm Samarium 62	152 Eu Europium 63	157 Gd Gadolinium 64	159 Tb Terbium 65	**163 Dy Dysprosium 66**	**165 Ho Holmium 67**	**167 Er Erbium 68**	**169 Tm Thulium 69**	**173 Yb Ytterbium 70**	175 Lu Lutetium 71
227 Ac Actinium 89	232 Th Thorium 90	231 Pa Protactinium 91	238 U Uranium 92	237 Np Neptunium 93	247 Pu Plutonium 94	243 Am Americium 95	247 Cm Curium 96	247 Bk Berkelium 97	251 Cf Californium 98	254 Es Einsteinium 99	253 Fm Fermium 100	256 Md Mendelevium 101	254 No Nobelium 102	257 Lr Lawrencium 103

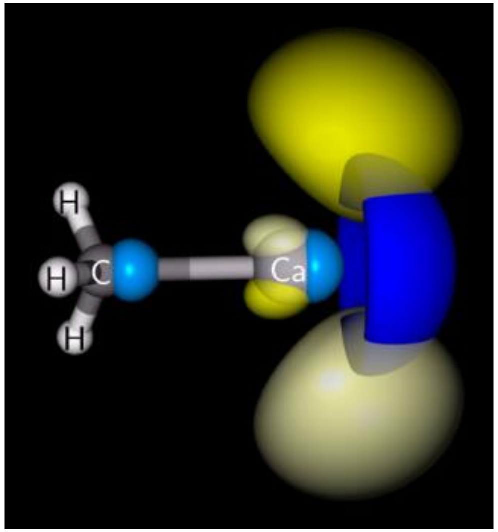
Polyatomic Molecules

Polyatomic Molecules

- Additional degrees of freedom to engineer desirable properties
  - Electric and magnetic field interactions
  - High polarizability
  - 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

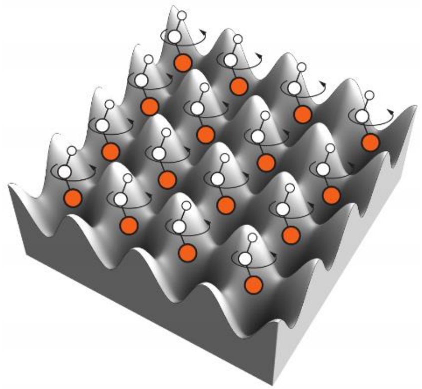


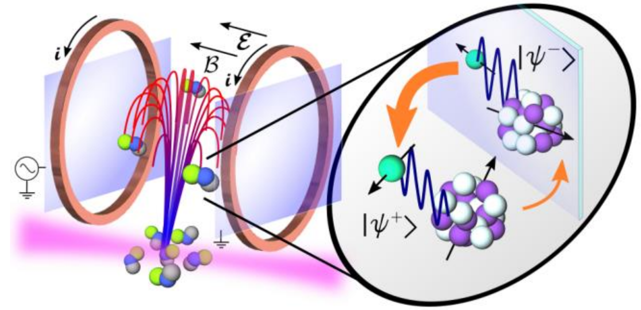
Adapted from Eric Hudson, Science 369, 1304 (2020)

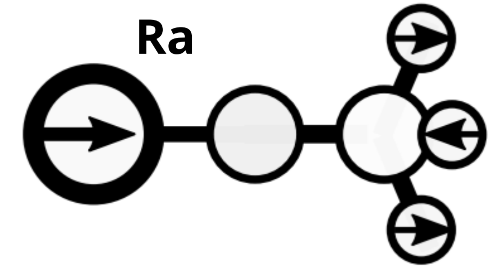


T. A. Isaev and R. Berger
PRL **116**, 063006 (2016)

Three Examples







YbOH

- Combine laser cooling, high polarizability
- PolyEDM: NRH, Doyle, Steimle, Vutha,
- Visit *polyedm.com* to see what we are up to!
- I. Kozyryev and NRH, PRL 119, 133002 (2017)

MgNC

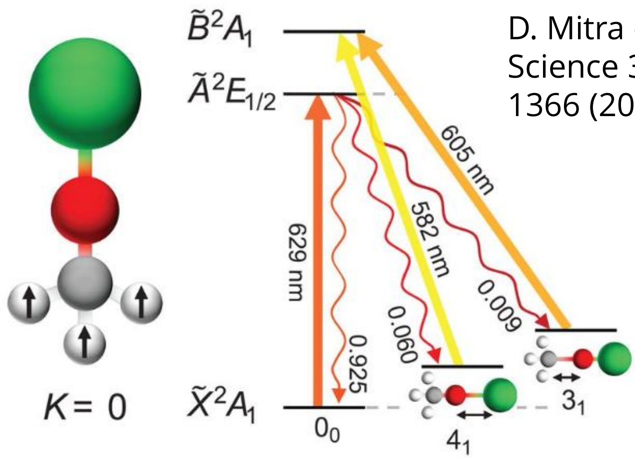
- Engineer magnetic field interactions for PV
- Reduces B field, adds many systematic checks
- E. B. Norrgard, et al, Nat. Comm. Phys. 2, 77 (2019)

$RaOCH_3^+$

- Combines deformed nucleus with ion trap EDM approach (more later)
- Recently created in Jayich Lab @ UCSB
- M. Fan et al., 2007.11614 (2020)
  P. Yu and NRH, 2008.08803 (2020)

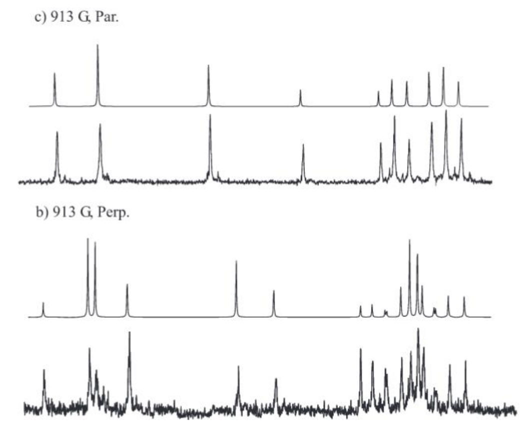
… Many, many more!

Selected Experimental Advances

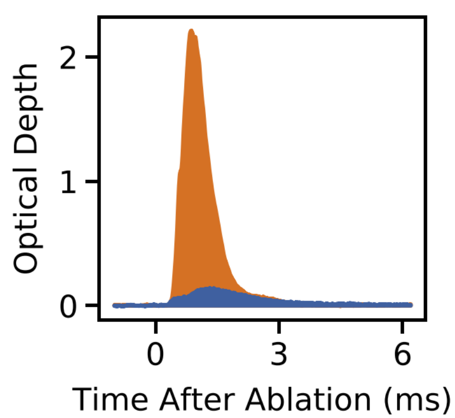


D. Mitra et al., Science 369, 1366 (2020)

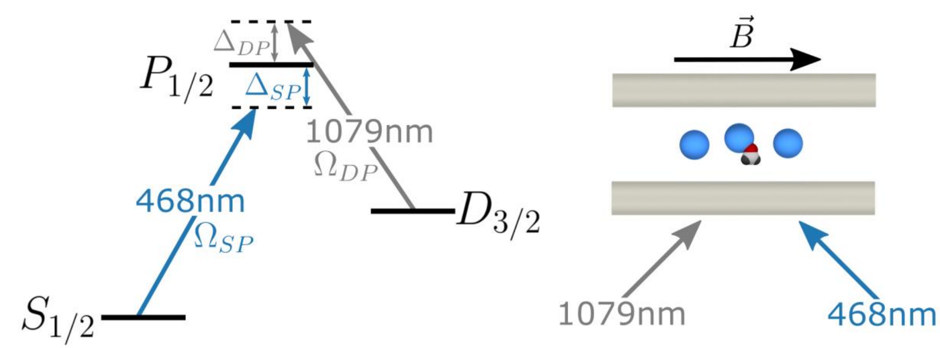
Laser cooling (Doyle @ Harvard)
SrOH, CaOH, YbOH, $CaOCH_3$



High resolution, broadband spectroscopy
D. Nguyen T. C. Steimle et al., J. Mol. Spec. 347, 7 (2018)



Production methods
A. Jadbabaie et al., New J. Phys. 22, 022002 (2020)

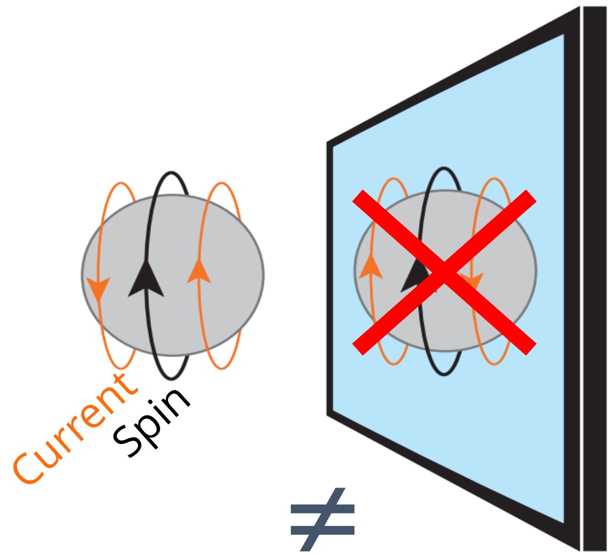


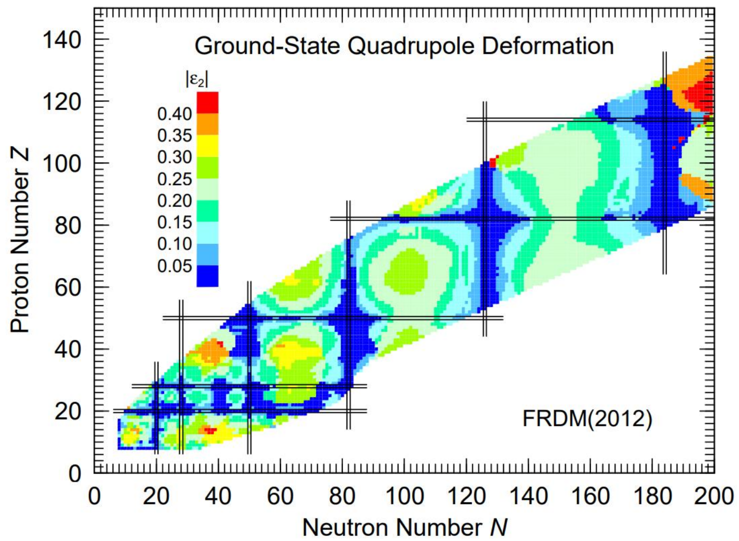
Ion trapping, cooling, control
Fan et al., 2007.11614 (2020)

Deformed Nuclei

Hadronic CPV Enhancement

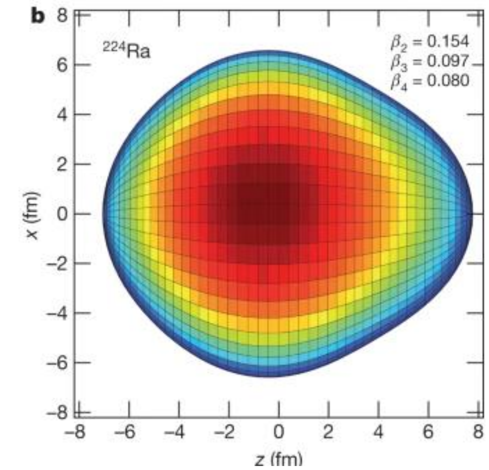
- Quadrupole (β_2) and octupole (β_3) deformations enhance hadronic CPV
  - θ_{QCD}, chromo-EDMs, nucleon EDMs, CPV forces, …
  - Combines with molecular enhancements
- β_2: Magnetic quadrupole moments (MQMs)
  - Collective enhancement, typically ~10
  - Yb, Ta, Hf, Th, Ra, …
    - V. V. Flambaum, et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



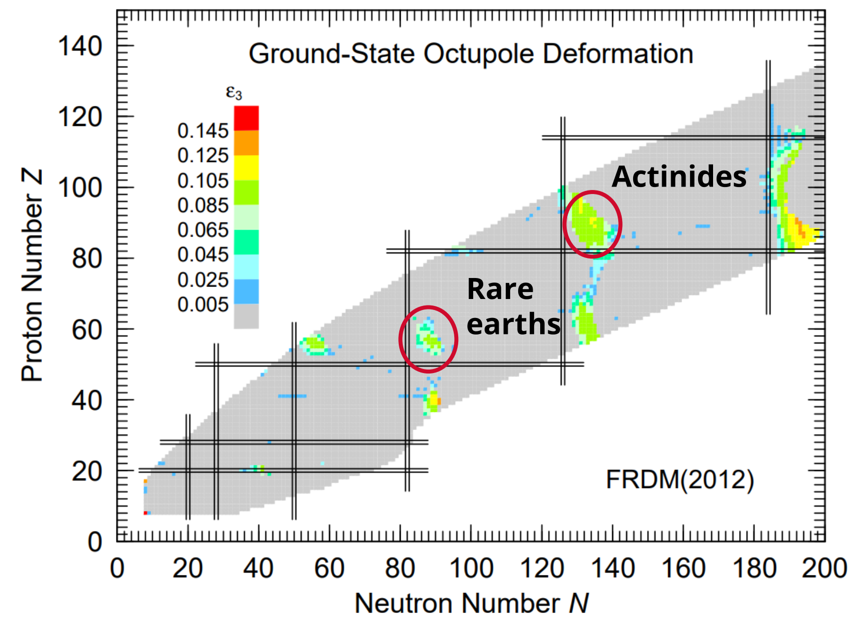


Octupole Deformations

- β_3: Schiff Moments (NSMs) enhanced by ~100-1,000
  - "Hard to come by"
  - Ra, Ac, Th, …
  - Heavy, spinful, deformed species are short-lived
- Combines with molecular enhancements → $10^{5\text{-}6}$ 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)



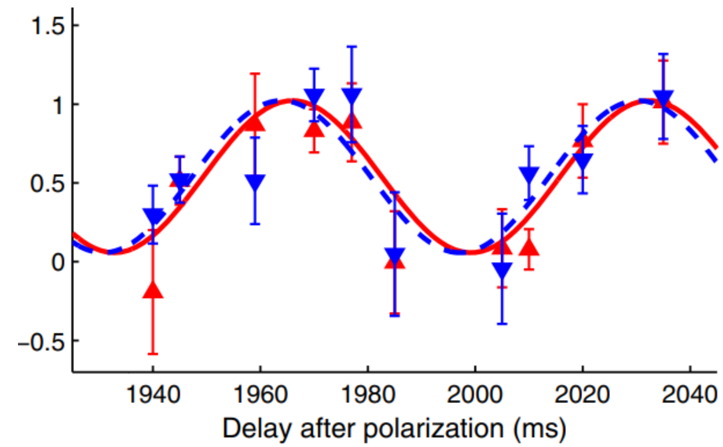
L. P. Gaffney *et al.*, Nature 497, 199 (2013)



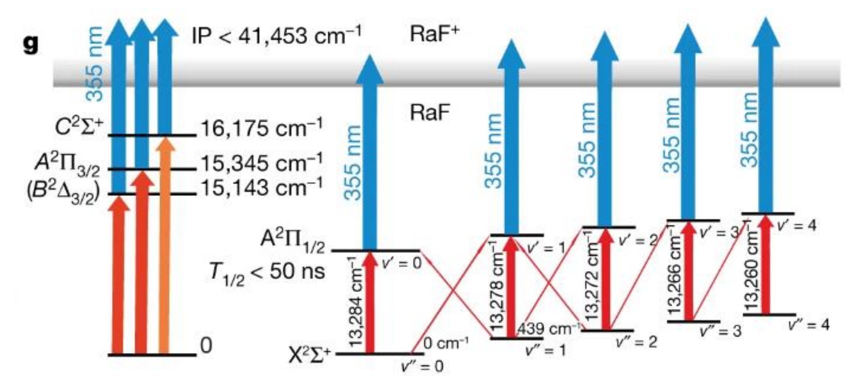
Radium

- Ra is especially interesting!
  - Ra, Ra^+, Ra molecules can be laser cooled
  - 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
RaAg	Assemble from laser-coolable atoms See work by Fleig and DeMille
$RaOCH_3^+$	Trapped, cooled/controlled with co-trapped Ra+ [Fan et al., 2007.11614 (2020)] Single ion could reach frontiers of hadronic CPV [Yu and NRH, 2008.08803 (2020)]
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)



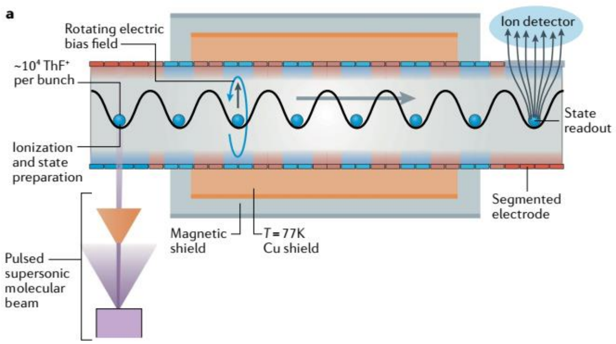
Ra EDM @ ANL
R. H. Parker, et al., PRL 114, 233002 (2015)



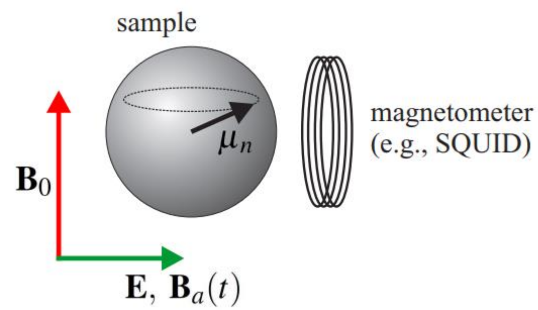
High-resolution RaF spectroscopy
R. F. Garcia Ruiz *et al.*, Nature 581, 396 (2020)

Other Directions

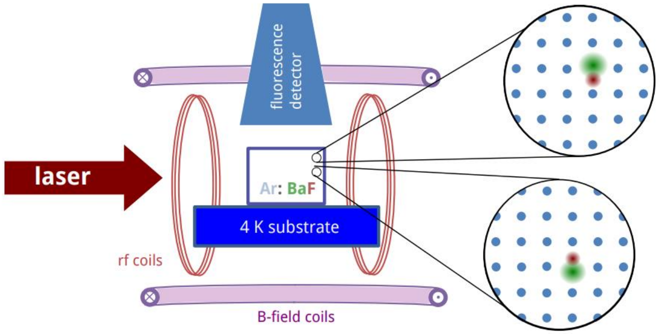
Other CPV Approaches



W. B. Cairncross and J. Ye, Nat. Rev. Phys. **1**, 510 (2019).



CASPEr: Budker et al., Phys. Rev. X 4 021030 (2014)



EDM^3 :A. C. Vutha et al., Atoms 6, 3 (2018)

Next-gen ion trapping

- Combine long coherence time with large count rates
- Suitable for eEDM, NSM, MQM

Oscillating EDMs

- Probe new axion (and axion-like) parameter space
- Use sensitive NMR techniques to search for oscillating CPV
- “Static” EDM experiments also provide sensitivity

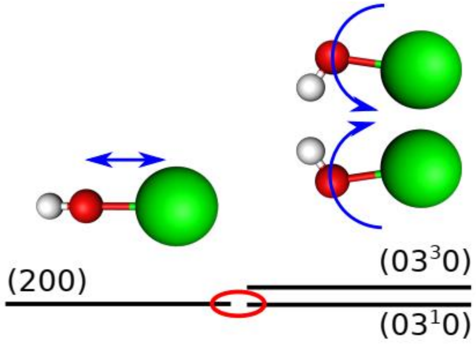
Graham and Rajendran, PRD 84, 055013 (2011), Stadnik and Flambaum PRD 89, 043522 (2014), Stadnik et al., PRL 120 013202 (2018)

Noble gas matrices

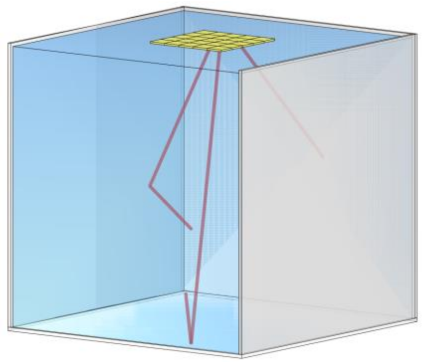
- Extreme count rates
- Suitable for molecules and atoms, including rare isotopes

J. T. Singh, Hyperfine Interact. 240, 29 (2019).

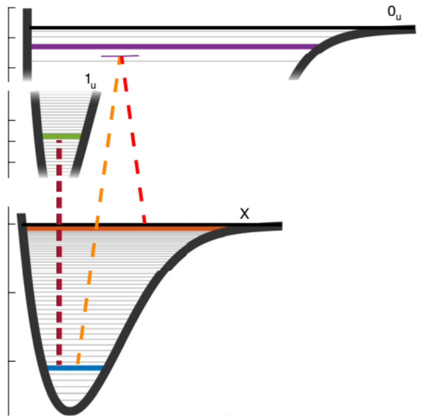
New Fields and Forces



I Kozyryev, et al.,1805.08185 (2020)



Arvanitaki et al., PRX 8, 041001 (2018)



S. S. Kondov et al., Nat. Phys. 15, 1118 (2019)

Drifting constants

- Can arise due to new light fields
- Molecules especially sensitive to m_p/m_e
  - Rotation, vibration
- Enhanced at nearly-degenerate levels
  - Complexity is advantageous!

Flambaum and Kozlov, PRL 99 (2007), DeMille et al., PRL 100 043202 (2008150801), Kobayashi et al., Nat. Commun. 10, 3771 (2019).

New fields

- New heavy fields can oscillate on laboratory timescales
- Resonant absorption, scattering, precession, mixing, …
- Molecular levels highly tunable with fields

Stadnik and Flambaum, PRD 89, 043522 (2014), R. Essig et al., PRR 1, 033105 (2019), Flambaum et al. PRD, 101 073004 (2020)

New forces

- New short-range forces will modify molecular binding
- Use precision molecular clocks to look for deviations from theory

Summary

Precision measurements with molecules have made tremendous advances in the last decade, and will lead to orders-of-magnitude improvements in many BSM searches in the not-too-distant future

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)
- **Interpretation of EDM limits**
  - See list of references in Safronova, Chupp reviews
  - J. Engel et al., Prog. Part. Nucl. Phys. 71, 21 (2013)
- **Laser cooling molecules**
  - M. R. Tarbutt, Contemp. Phys. 59, 356 (2018)
  - D. McCarron, J. Phys. B At. Mol. Opt. Phys. 51, 212001 (2018)
- **Email me!**

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Summer 2020

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