Title: EDM measurements

Speakers: Nick Hutzler

Collection: School on Table-Top Experiments for Fundamental Physics

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Searches for Fundamental Symmetry Violations with Atoms and Molecules

Nick Hutzler
Caltech

Notes About the Content

- We will focus mostly on CP-violation searches (EDMs) in atoms and molecules
  - Mainly because this is what I do and what I know!
  - Many of the ideas and techniques are directly applicable to a wide range of atomic/molecular science, and relevant for a very wide range of research, from HEP to QIS to Chemistry

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- The slides are on the Indico website

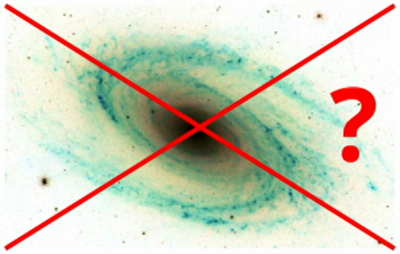
Part I: Motivation and Background

Why are we interested in CP-violation at low energy?

How are atoms and molecules used for these searches?

An Asymmetric Universe

- There is a large imbalance between matter and anti-matter in the universe
  - Baryon Asymmetry of the Universe (BAU)
- How can we explain this?



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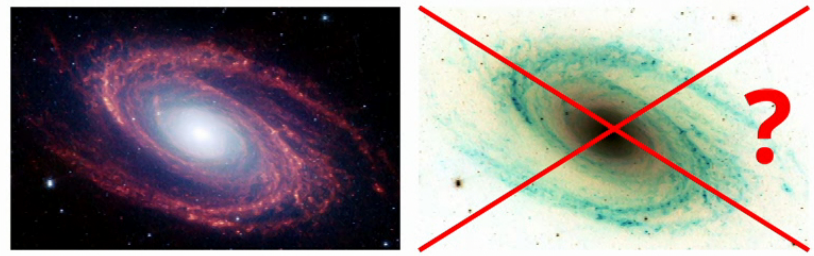

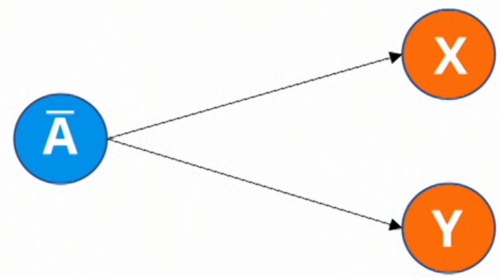

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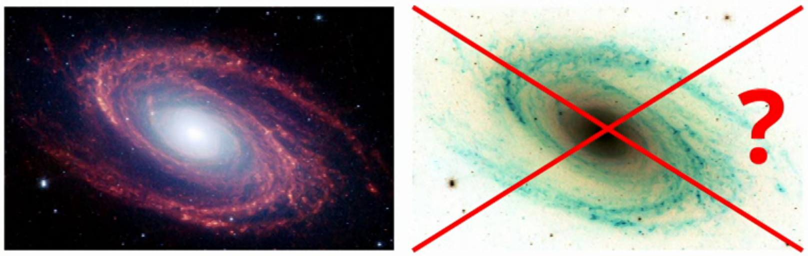
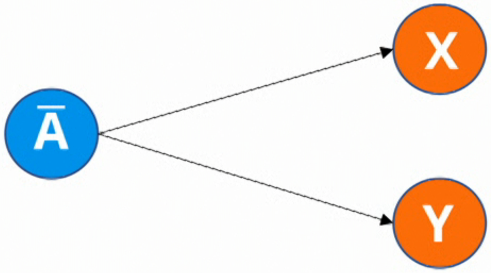
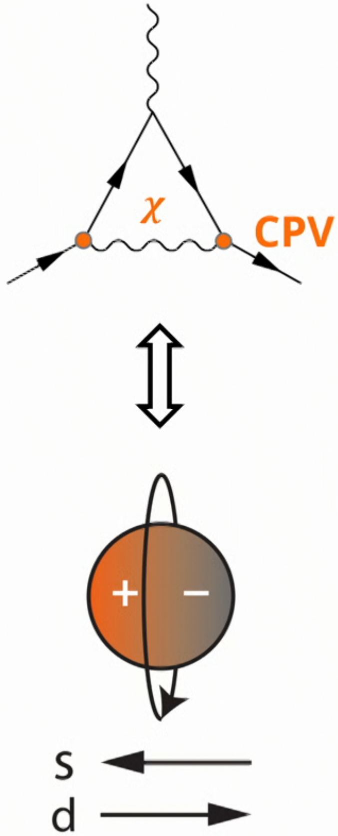
- Sakharov conditions:
  - B-violating processes
  - Departure from thermal equilibrium
  - C- and CP-violation ← our focus!
  - Must be outside Standard Model





Low Energy Observables

- New CPV physics with SM couplings generically gives rise to CPV electromagnetic moments in SM objects
  - Fundamental particles:
    - Electric dipole moment (EDM)
  - Nuclei:
    - Nuclear Schiff moment (NSM)
    - Magnetic quadrupole moment (MQM)



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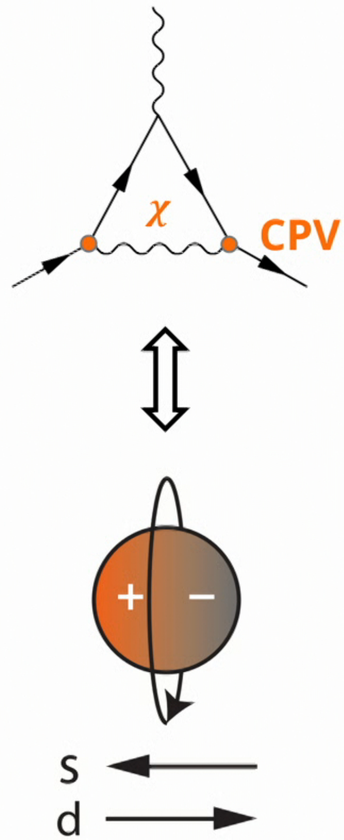
- Moments must lie along intrinsic spin (no internal other vectors!)
- $\vec{d} \propto \vec{s}$ → problem
  - $\vec{d}$ is P-odd, T-even
  - $\vec{s}$ is P-even, T-odd



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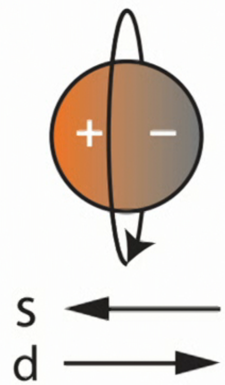

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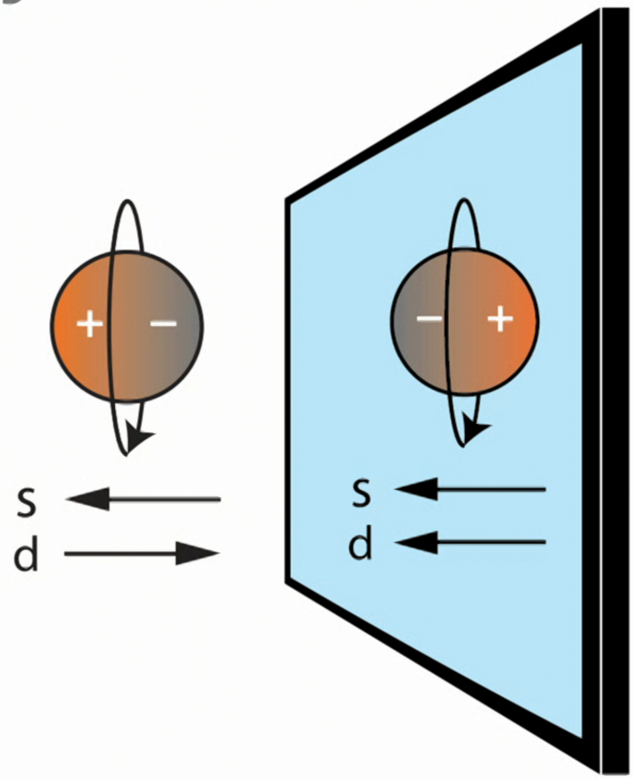

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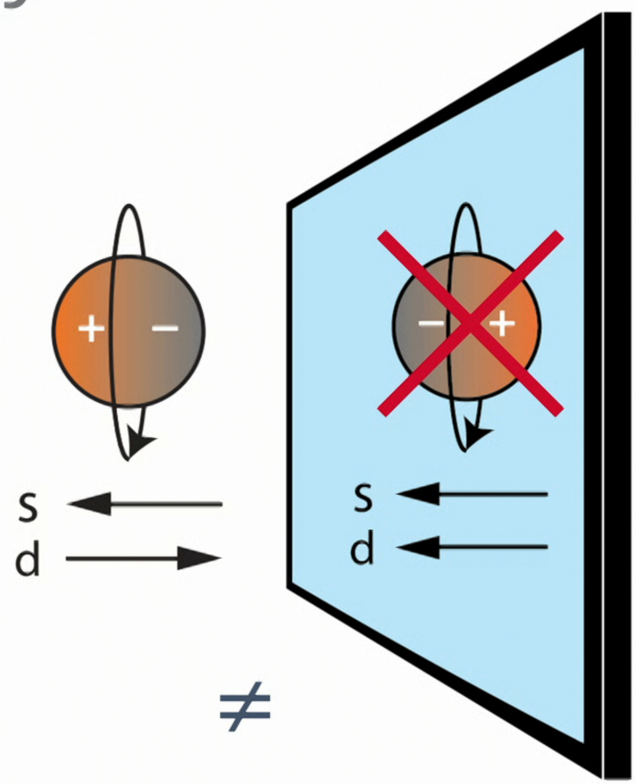

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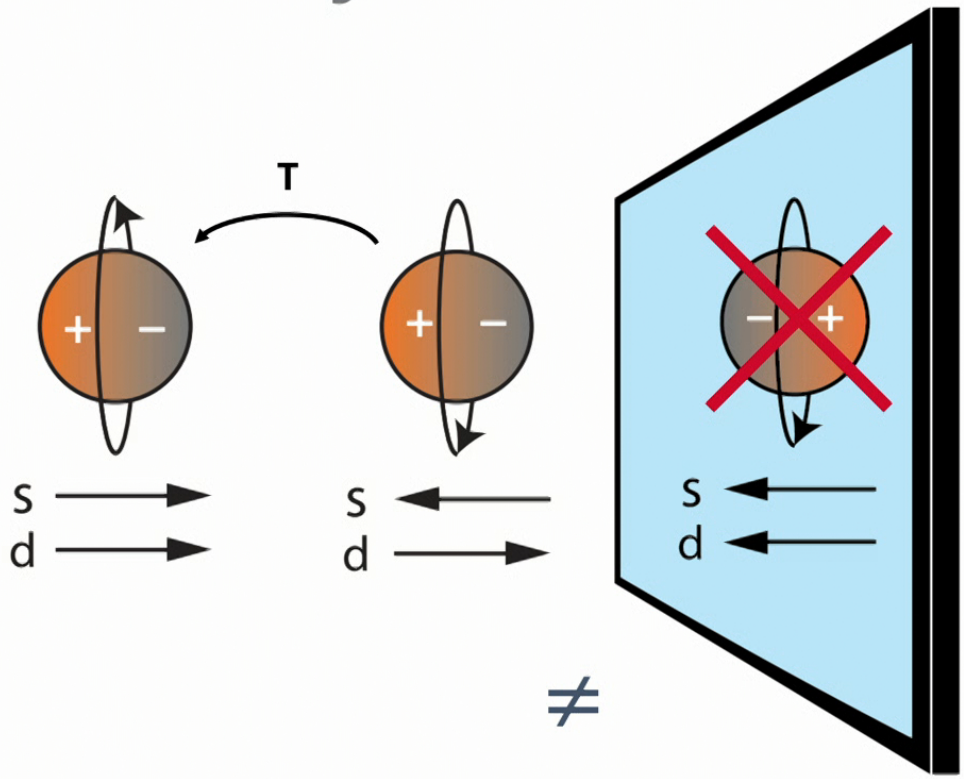

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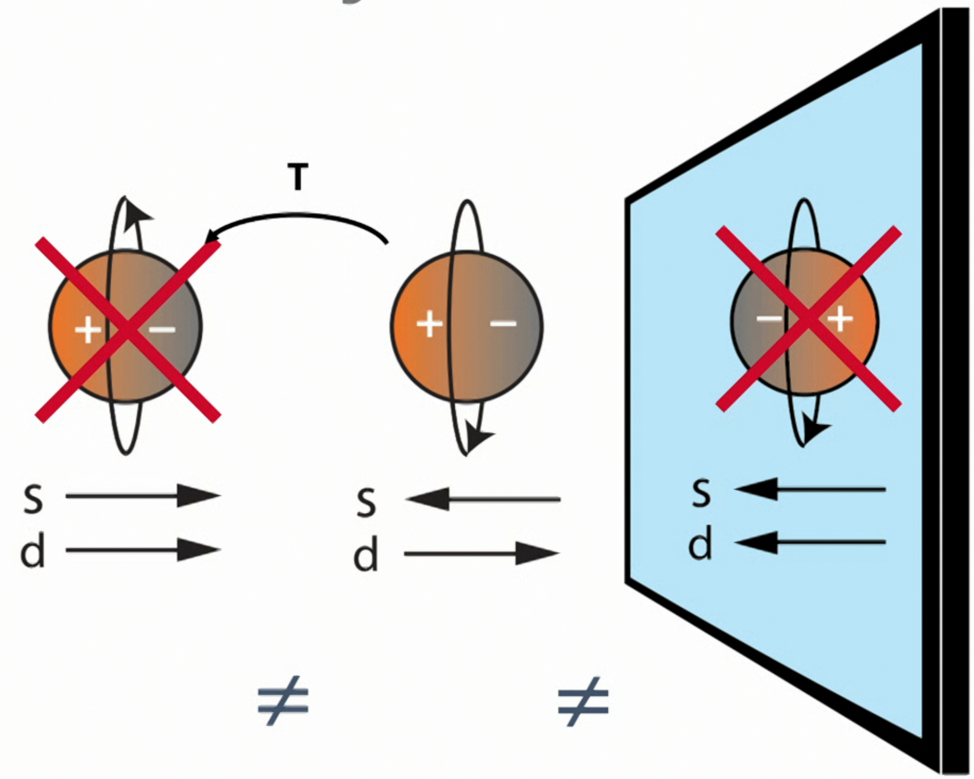

EDMs violate P, T, CP*

(*Assuming conservation of CPT…)

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Electron EDM

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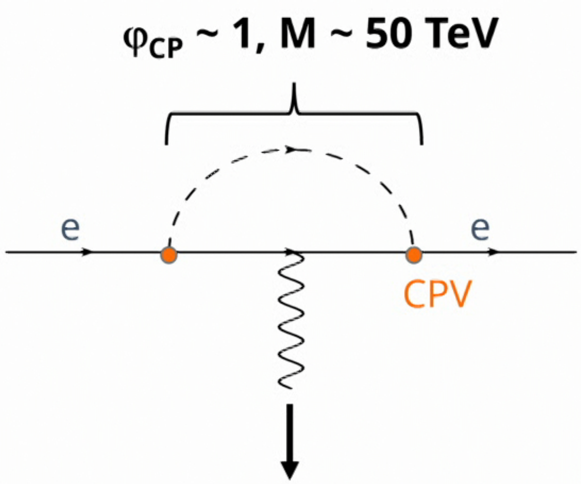
  - One loop ~ 10-50 TeV
  - Two loop ~ 0.5-2 TeV



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(current experimental limit)

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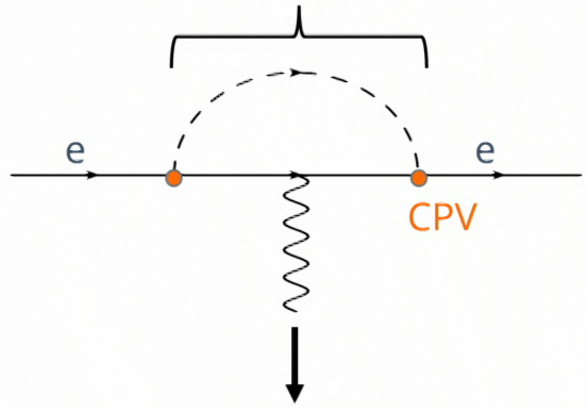
    - (Complicated)
  - Arises from CKM @ 4 loops

φ_{CP} ~ 1, M ~ 50 TeV



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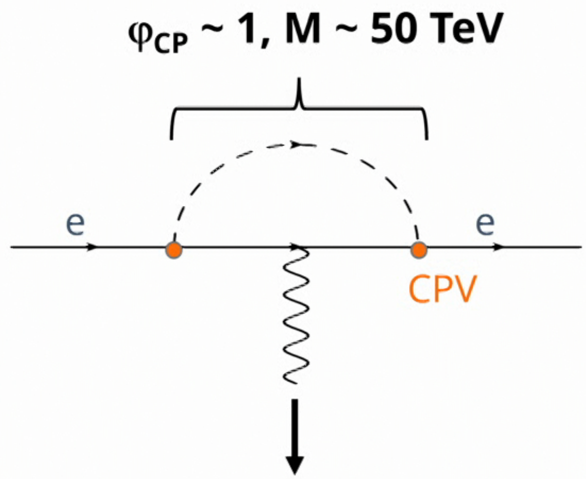
- For specific models, energy reach can be even higher (or lower!)



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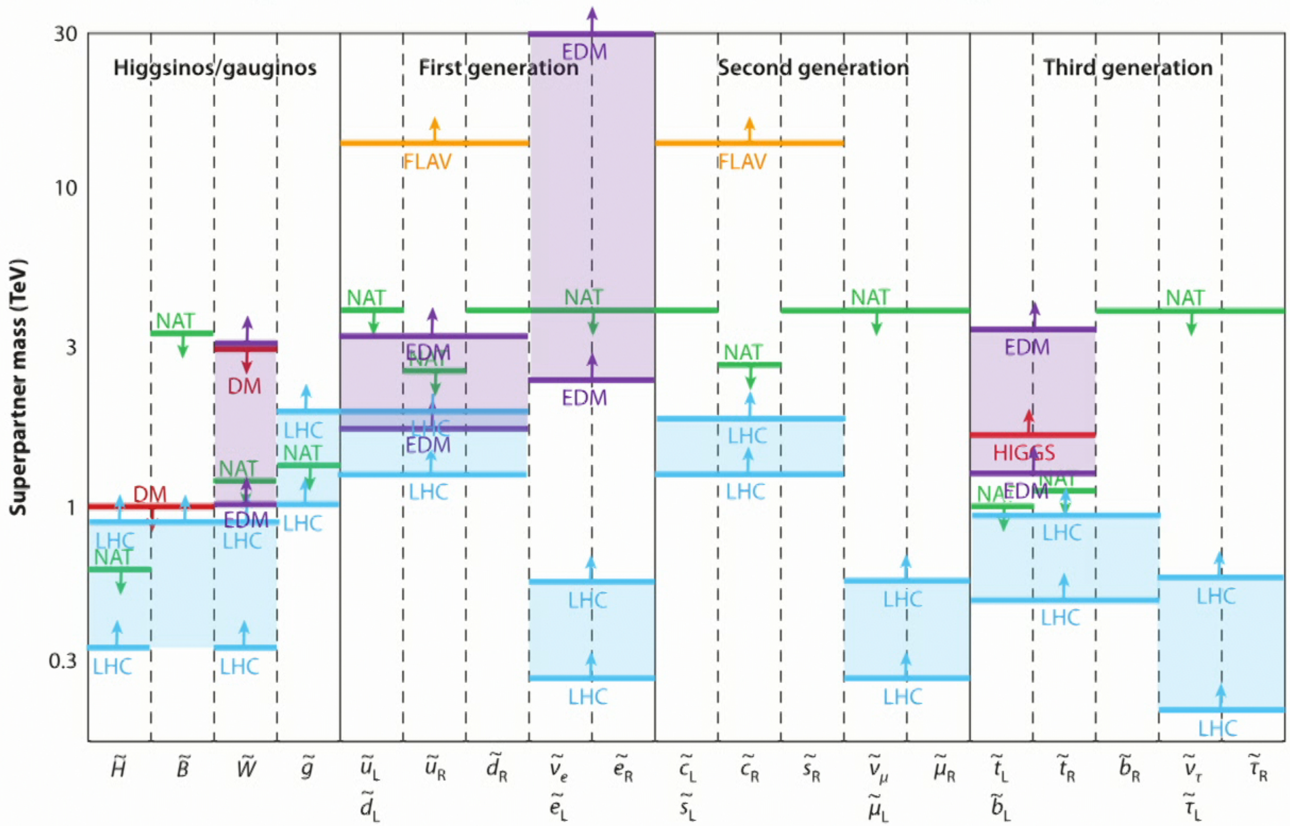
Many complementary approaches

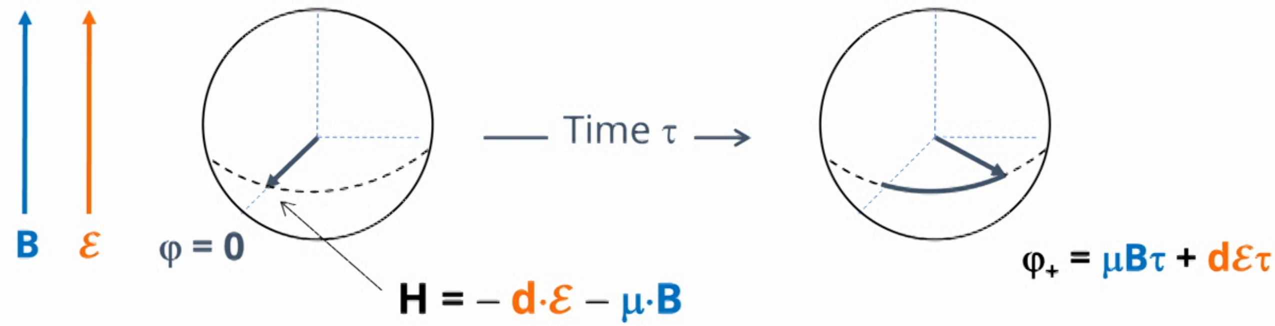


Shading shows progress since 2013 (LHC, ACME, nEDM, ^{199}Hg)

"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

An Idealized EDM Experiment



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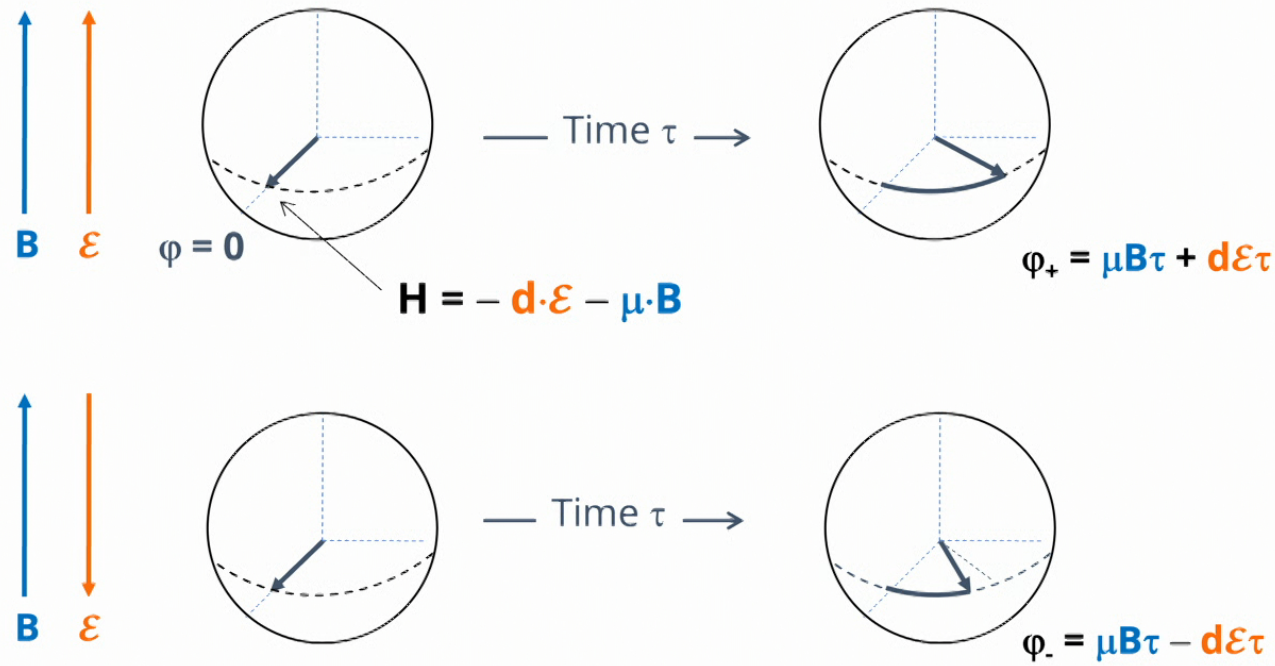

$$\Delta\varphi \propto d\mathcal{E}\tau$$

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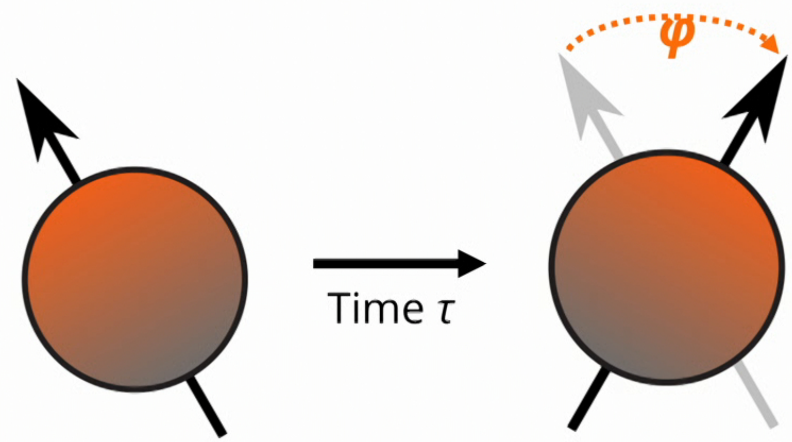
Sensitivity

- Experimental observable is angle φ (phase),

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

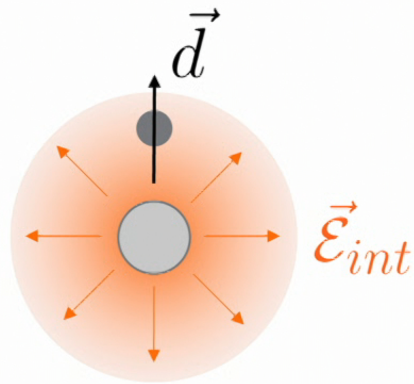
- Repeated measurements:

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



Internal Fields

- Basic idea: Atoms/molecules have **extremely large** fields
  - $e/4\pi\epsilon_0 a_0^2 \sim$ GV/cm
  - Up to ~100 GV/cm for heavy species
  - Much larger than "maximum" lab field of ~100 kV/cm



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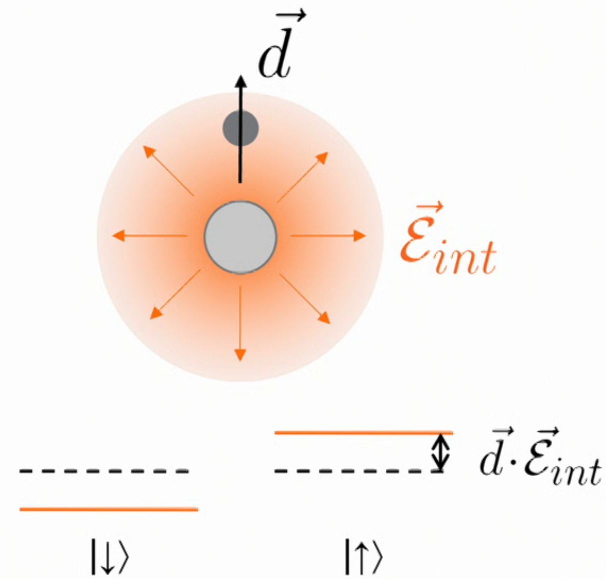
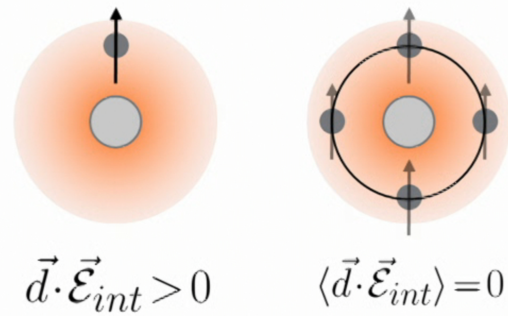
- Permanent EDM causes splitting of energy levels
  - Amplified by internal fields



Polarization

- First "internal field" picture caveat – electrons and nucleus experience zero average field!
  - Always the case: eigenstates have well-defined parity, so $\langle\psi|\vec{\mathcal{E}}|\psi\rangle \propto \langle\psi|\vec{r}|\psi\rangle = 0$



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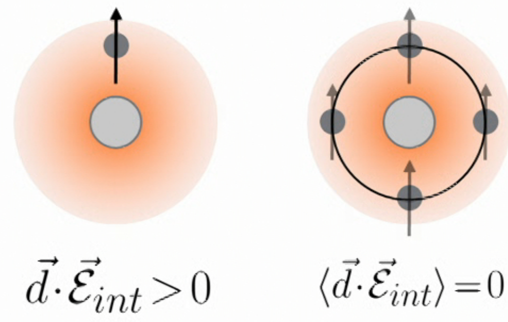
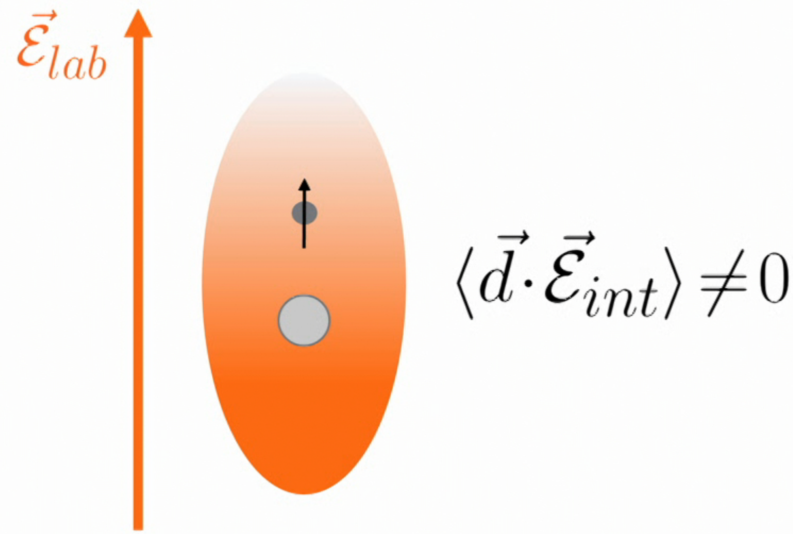
- **Solution: polarize**
  - Apply lab field to polarize atom/molecule
  - Interaction no longer averages to zero





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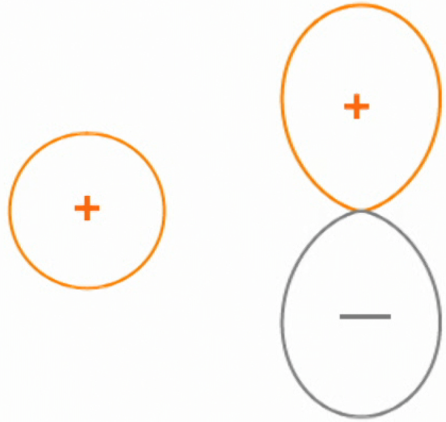

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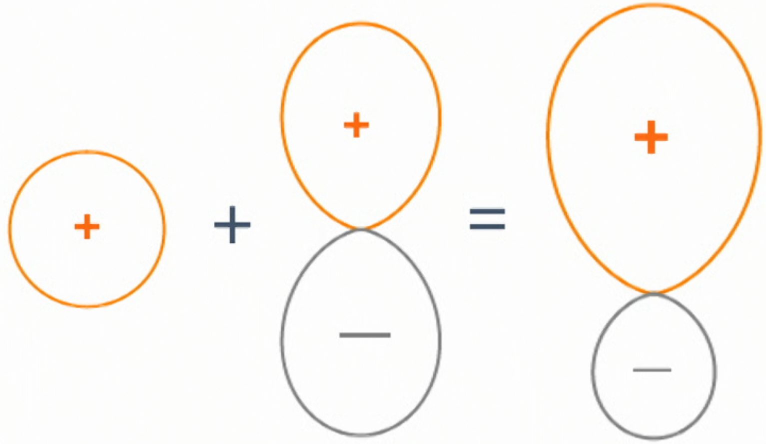
- Must overcome energy splitting Δ between the states to polarize
  - Therefore an induced dipole moment
- EDM sensitivity is ∝ P, fractional polarization



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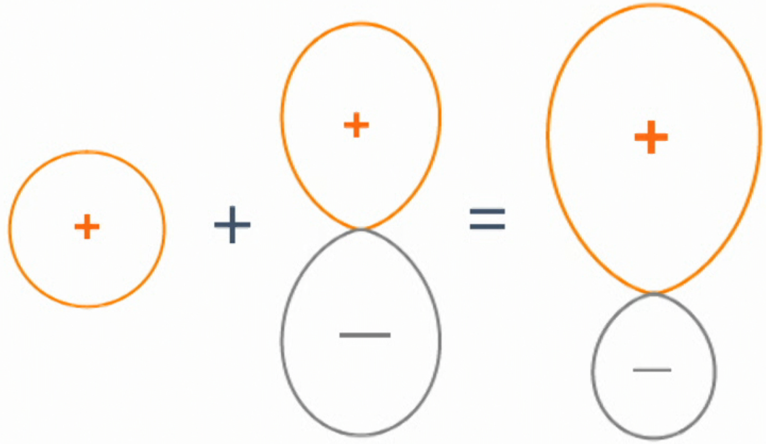

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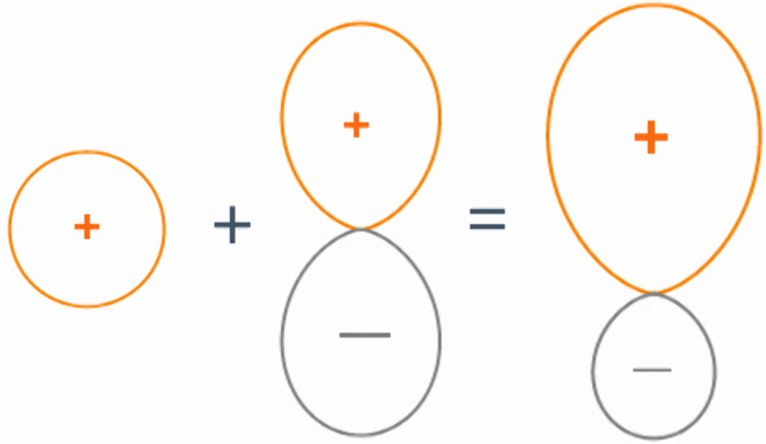

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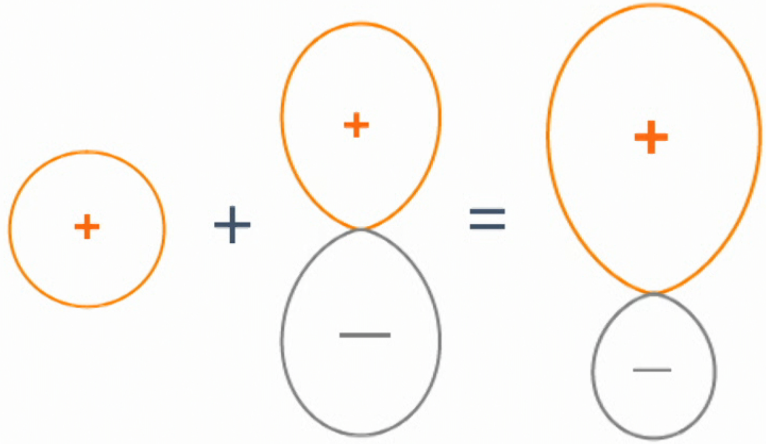
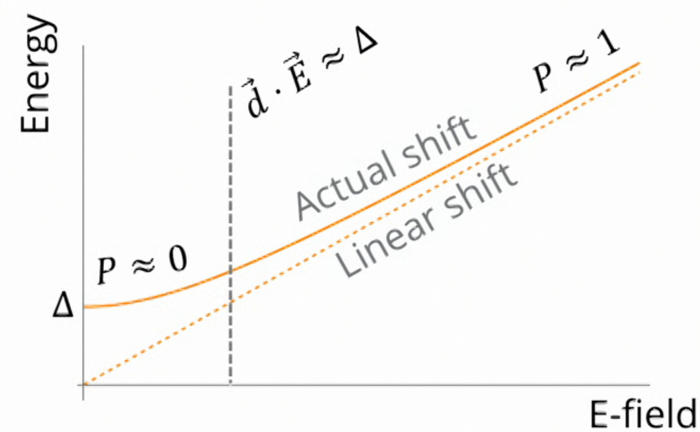
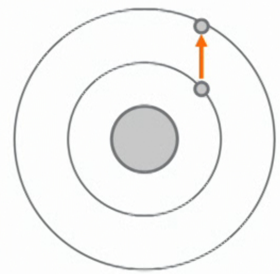


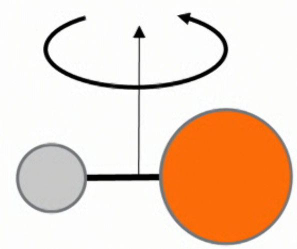


Atoms vs. molecules

- Atoms
  - Δ ~ 100 THz (electronic)
  - P ~ 10^{-3} @ 100 kV/cm
- Molecules
  - Δ ~ 10 GHz (rotational)
    - Sometimes even smaller, more on that later
  - P ~ $\mathcal{O}(1)$ @ 10 kV/cm
- **"Molecules are 1000x more sensitive"**



Atoms
Δ ~ 100 THz



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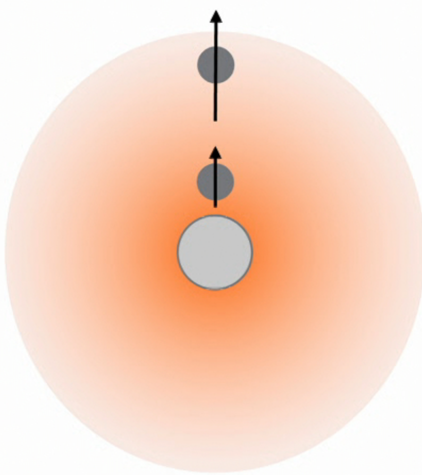
- This is true! But there are evasions:
  - Electrons move relativistically
  - Nuclei have complex shape

Relativistic Effects

- Dipole moment $\vec{d}$ experiences Lorentz contraction
- Correlation of velocity, position relative to nucleus
- $\vec{d}$ is not a constant in the atom



Commins *et al.*, Am. J. Phys. 75, 532 (2007)

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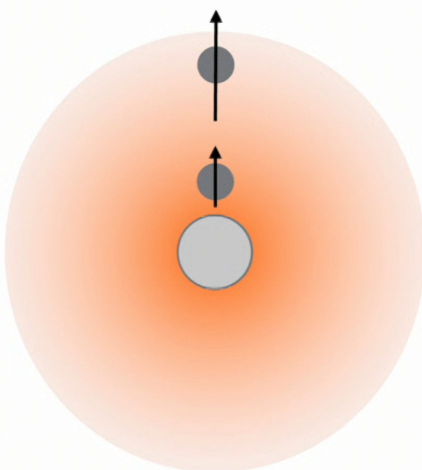

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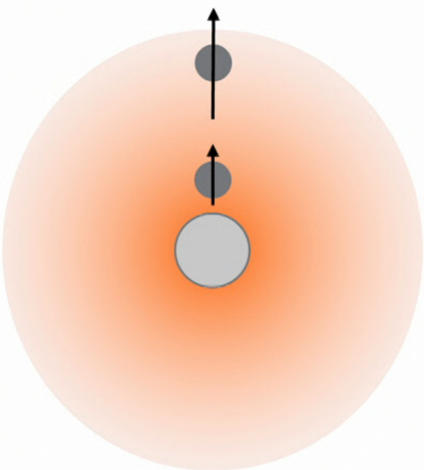
- Depends on very short-range electronic wavefunction near nucleus – very relativistic quantum mechanics
  - Requires core-penetrating electron orbitals



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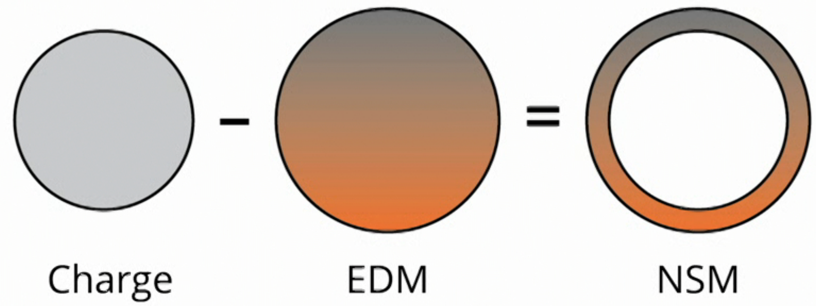

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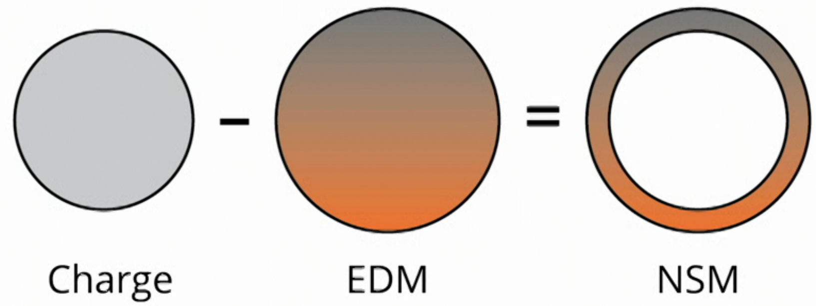
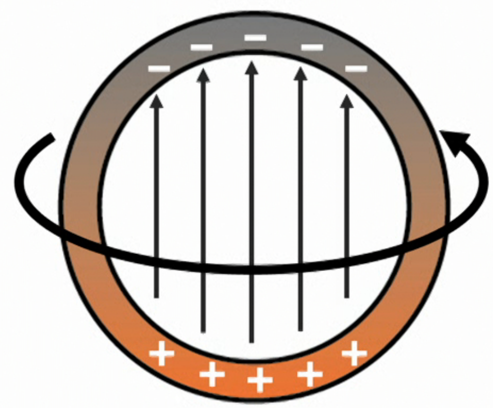
- Gives rise to a Nuclear Schiff Moment (NSM)
  - Depends on mismatch between charge and mass distributions in nucleus
  - Looks like an E field correlated with nuclear spin (CPV)
  - Mixes opposite parity, core-penetrating electron orbitals
  - Enhanced in high Z nuclei
  - Requires nuclear spin >0





Octupole Deformations

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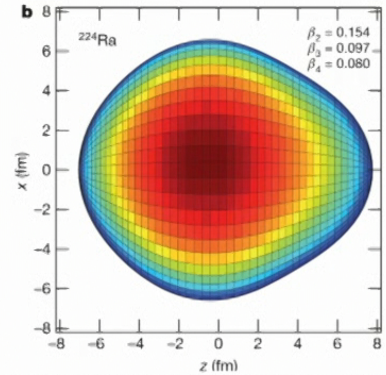
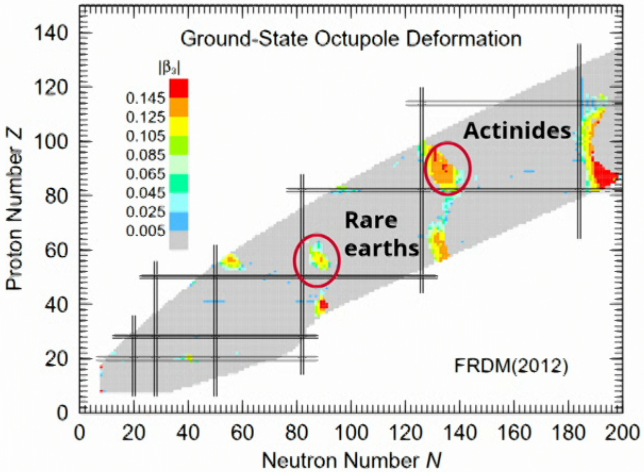


L. P. Gaffney *et al.*, Nature 497, 199 (2013)



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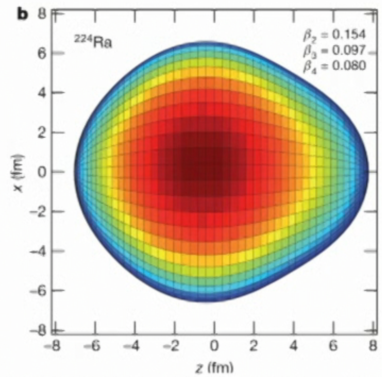

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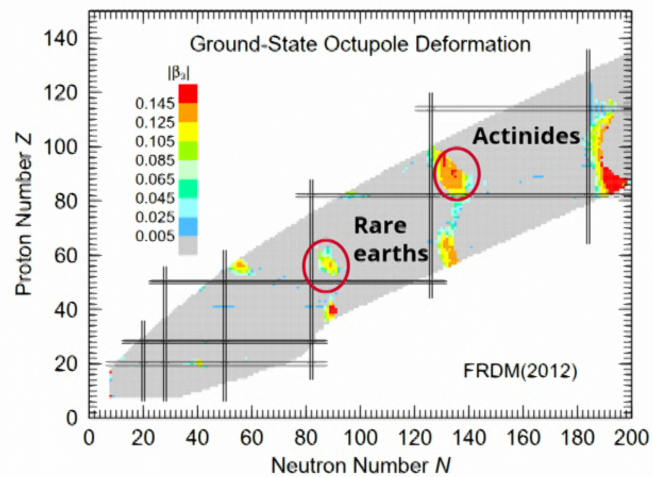

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  - Heavy, spinful, deformed species are short-lived
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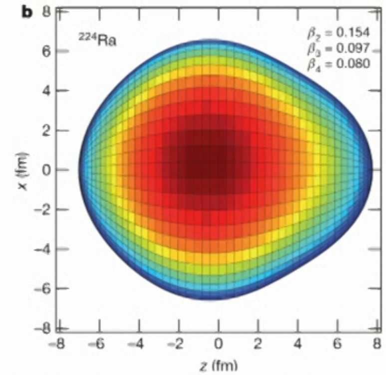
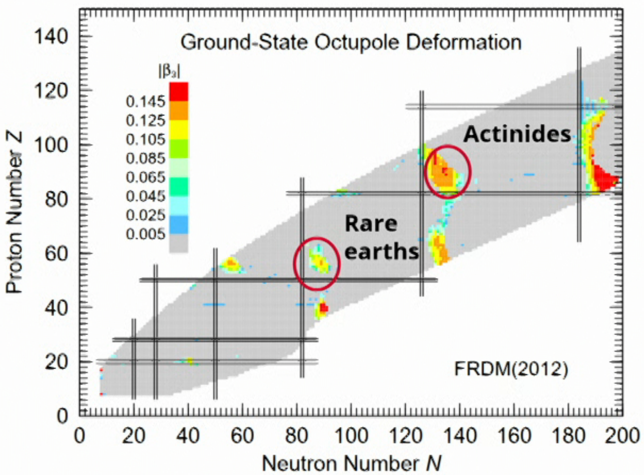
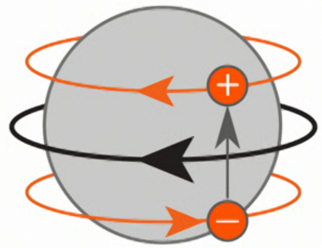


L. P. Gaffney *et al.*, Nature 497, 199 (2013)



Nuclear Magnetic Quadrupole Moments

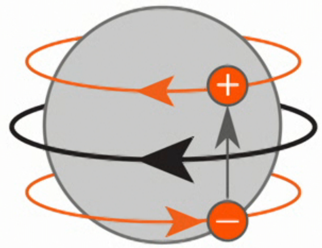
- Another evasion of Schiff shielding: magnetic effects
- Nuclear magnetic quadrupole moments (MQM) are not screened
  - Intuitive origin: orbiting nucleon with EDM creates an MQM ~ $\vec{I}$



Rotating EDM produces MQM

Nuclear Magnetic Quadrupole Moments

- Another evasion of Schiff shielding: magnetic effects
- Nuclear magnetic quadrupole moments (MQM) are not screened
  - Intuitive origin: orbiting nucleon with EDM creates an MQM $\sim \vec{I}$
  - Violates T, P, CP
- Need $I \geq 1$
- Enhanced in high Z nuclei
- Quadrupole deformation (β_2) enhances MQM



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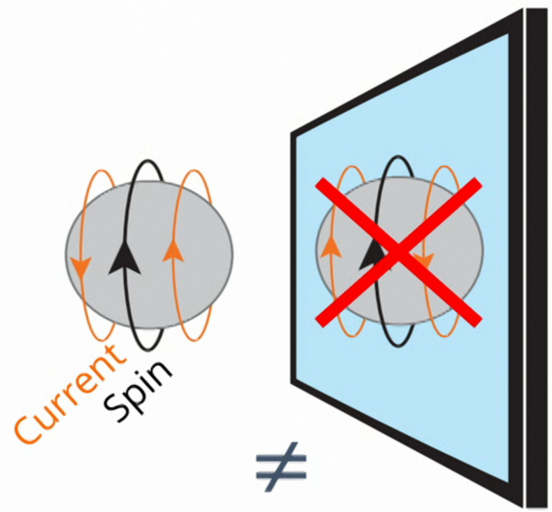
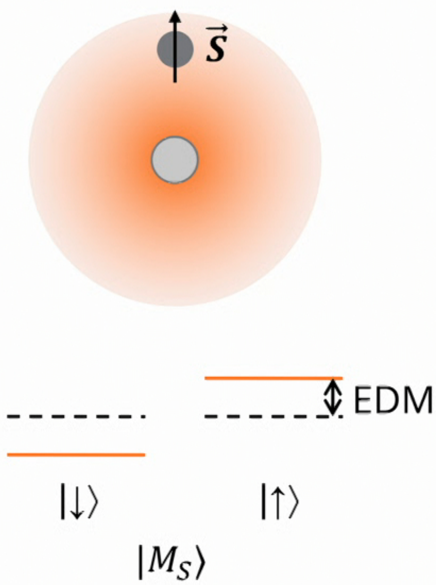
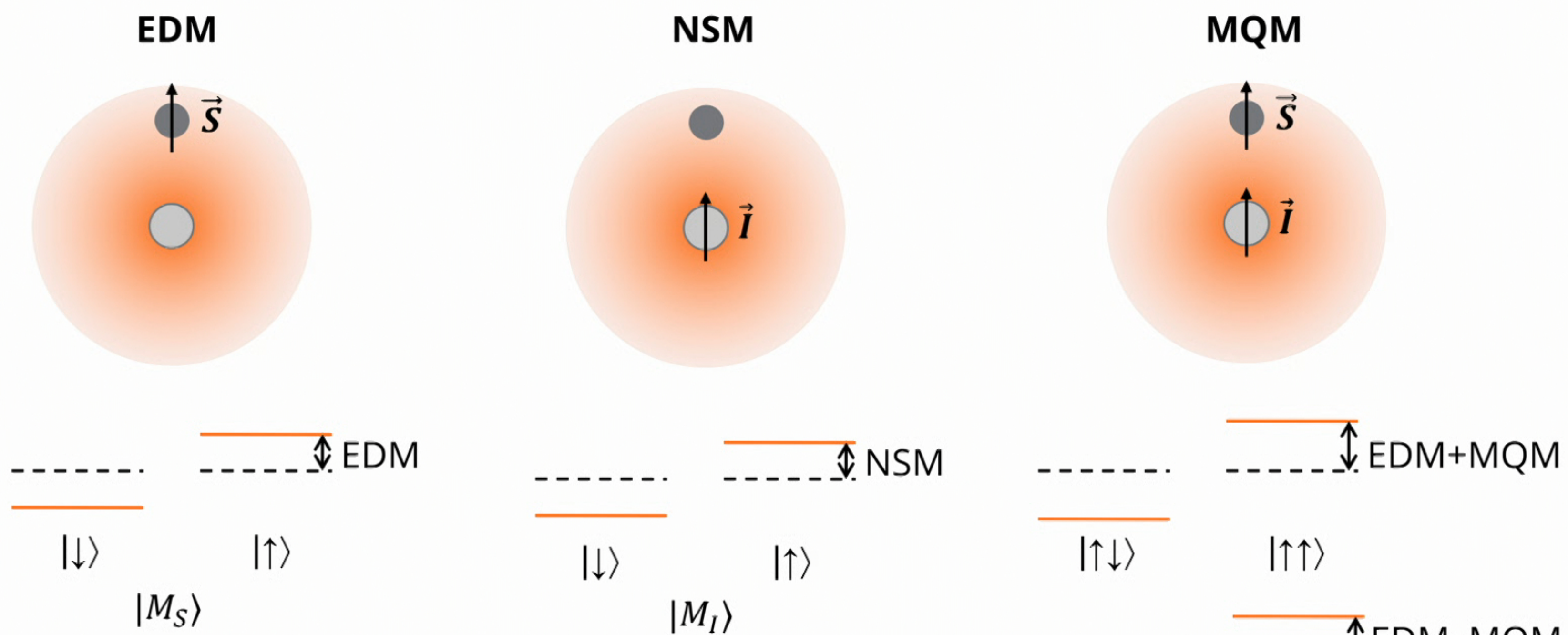


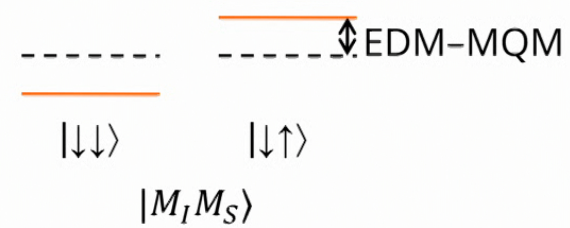
EDM vs. NSM vs. MQM shifts

EDM



EDM vs. NSM vs. MQM shifts



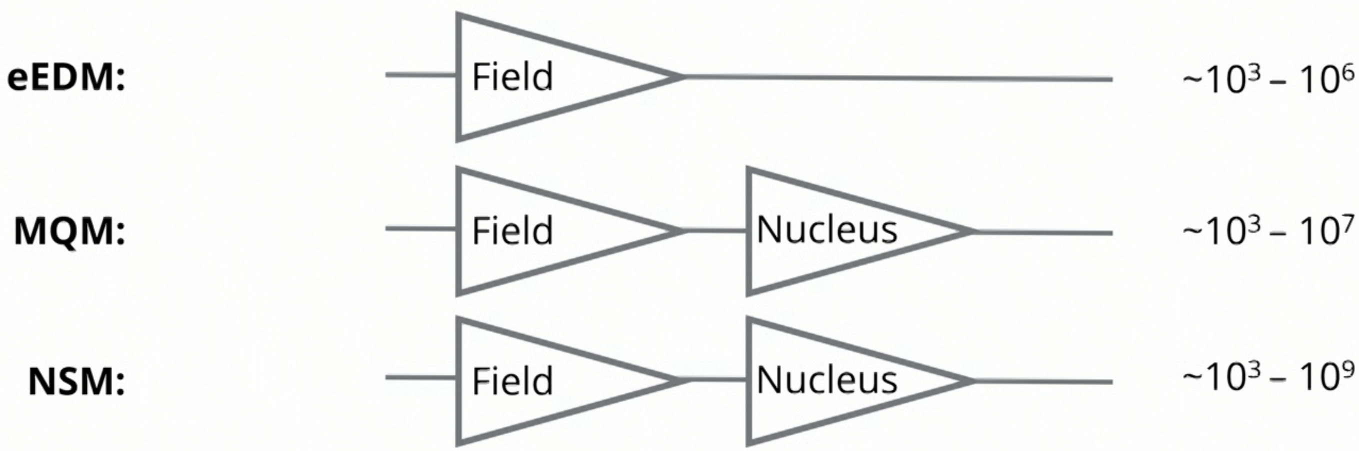


- **Very similar methods, requirements, approaches**
- **These are often all just called "EDM Experiments"**
- **All shifts are proportional to polarization**

Field + Nuclear Enhancements

	Internal Field Enhancement	Nuclear Enhancement
Atom	~1,000	
Molecule	~1,000,000	
Quadrupole nucleus (MQM)		~10
Octupole nucleus (NSM)		~1,000

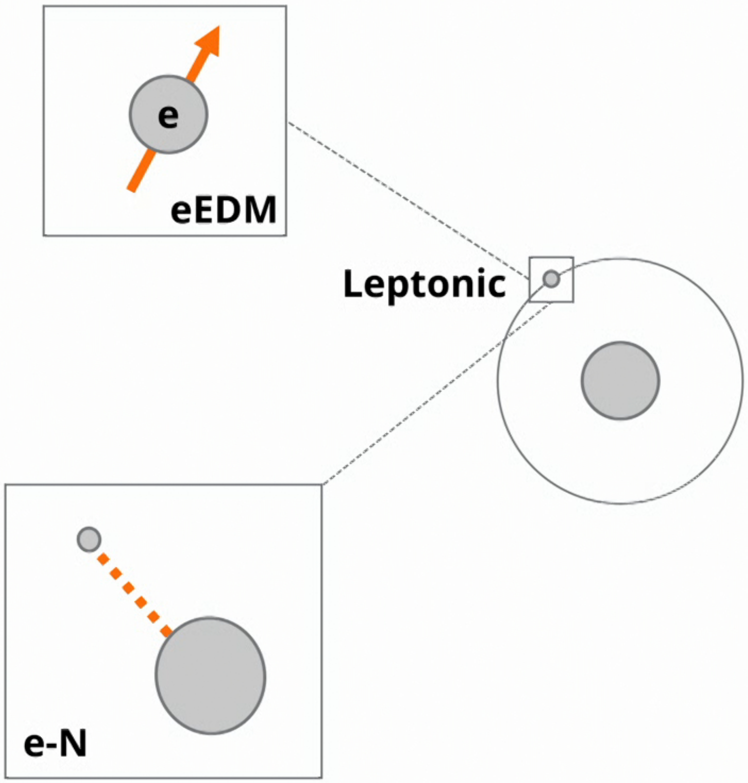
Both NSM, MQM *potentially* have additional ~1,000x enhancement in ^{229}Pa



Summary: Structure Requirements

Feature	eEDM	NSM	MQM	Reason
Large Z?	Yes!	Yes!	Yes!	Gives large atomic/molecular enhancement
Core-penetrating s/p-like electrons?	Yes!	Yes!	Yes!	Depends on short-distance electronic wave function (for different reasons)
Quadrupole nucleus?	Not necessary	Yes!	Yes!	Collective nuclear enhancement
Octupole nucleus?	Not necessary	Yes!	Not necessary	Collective nuclear enhancement
Open shell?	Necessary	Not necessary	Necessary	Electron spin density near nucleus
Nuclear spin?	Not necessary	$I \geq 1/2$	$I \geq 1$	Need nuclear spin to define moments

Many Sources



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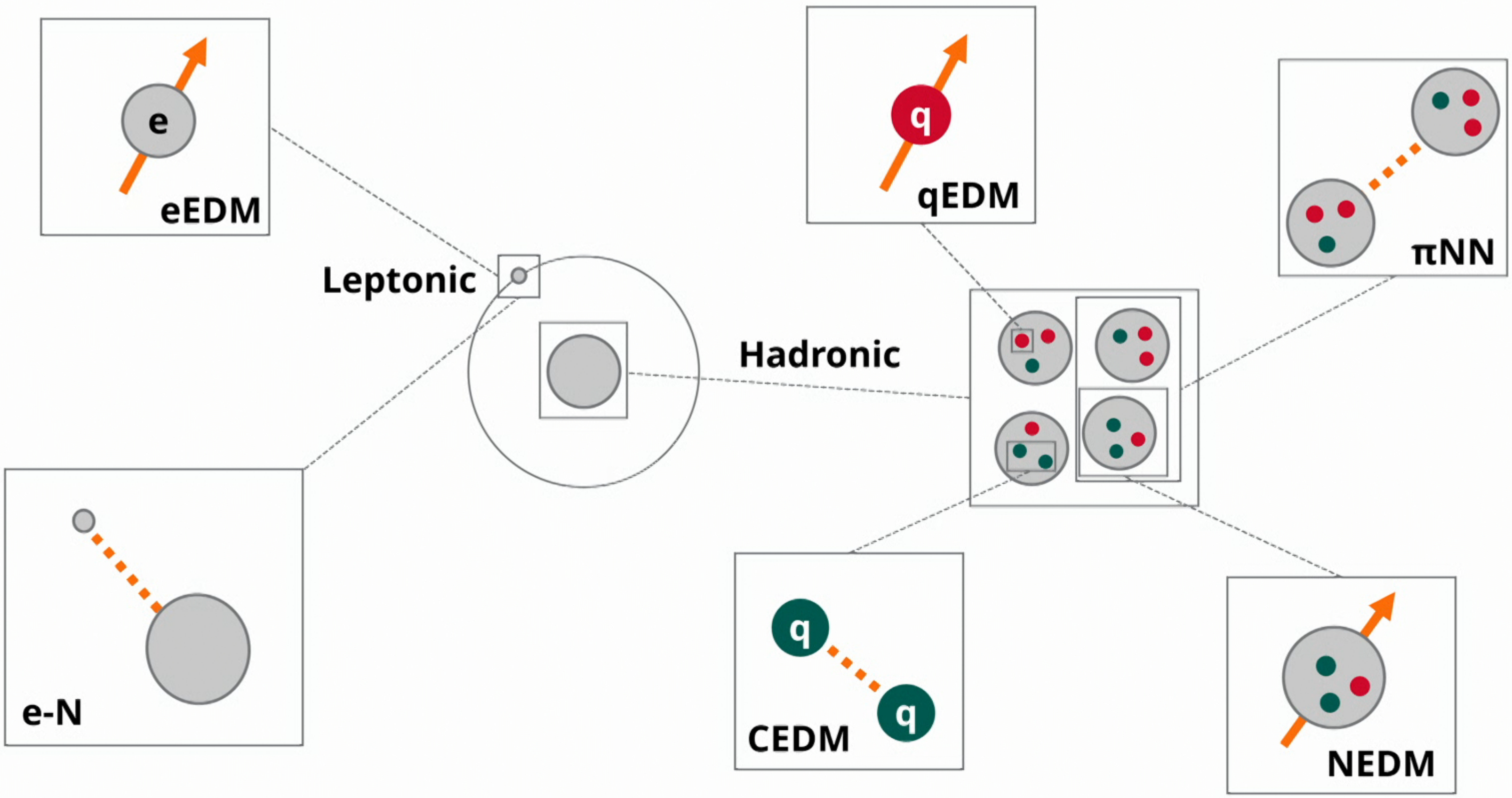
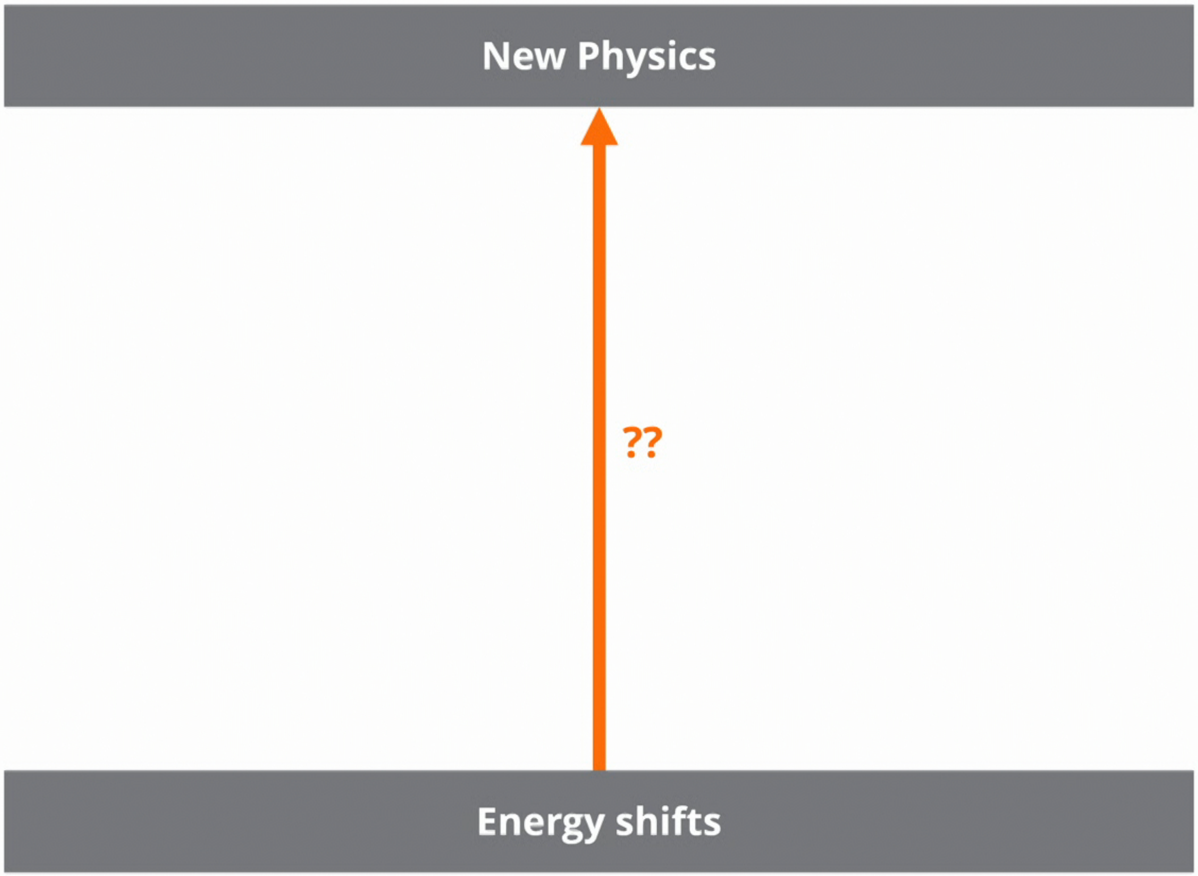
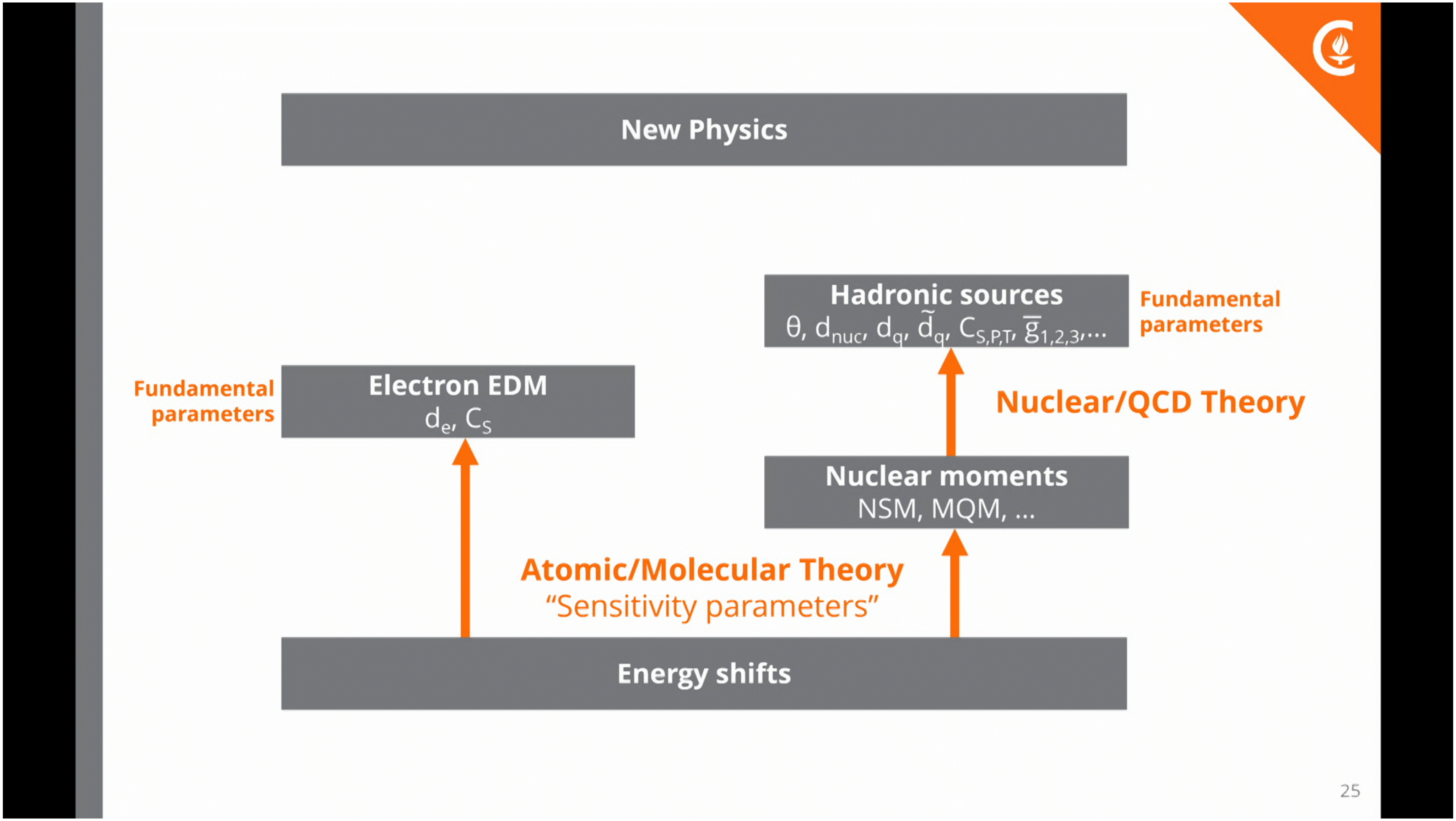



New Physics
??
Energy shifts


New Physics
Hadronic sources
θ, d_{nuc}, d_q, $\tilde{d}_q$, $C_{S,P,T}$, $\bar{g}_{1,2,3}$,...
Fundamental parameters
Fundamental parameters
Electron EDM
d_e, C_S
Nuclear/QCD Theory
Nuclear moments
NSM, MQM, ...
Atomic/Molecular Theory
"Sensitivity parameters"
Energy shifts
25

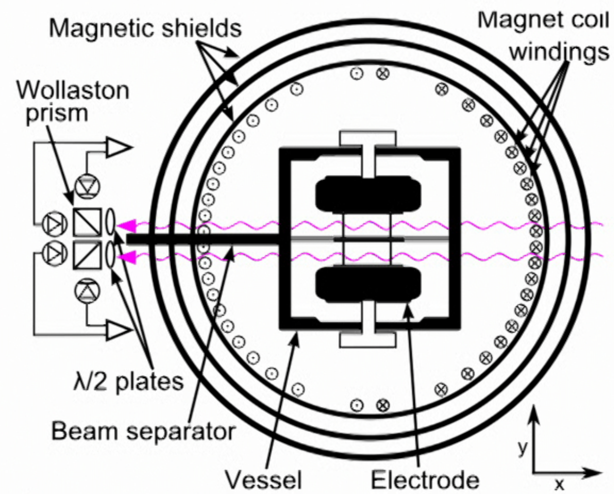
Part II: Selected Contemporary Experiments

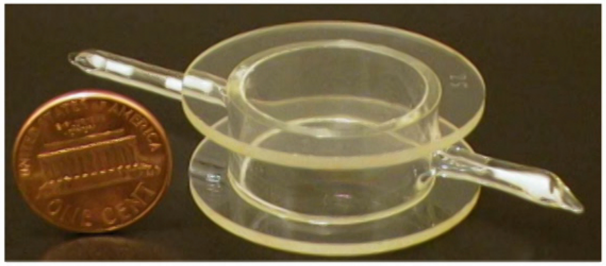
Not a review, but instead using them as a vehicle to explain modern experimental methods, and motivate ongoing and future improvements.

We will not discuss all ongoing EDM experiments!

^{199}Hg EDM

- University of Washington
- ^{199}Hg in a vapor cell
- $I = 1/2$ due to valence *n*
  - Spherical nucleus → no "nuclear enhancement"
- Heavy nucleus, highly relativistic electrons
  - Large "atomic enhancement"
  - Sensitive to NSM





B. Graner, Y. Chen, E. G. Lindahl, and B. R. Heckel, PRL 116, 161601 (2016)
Photo from Y. Chen FRIB Presentation, 2019
https://indico.frib.msu.edu/event/13/

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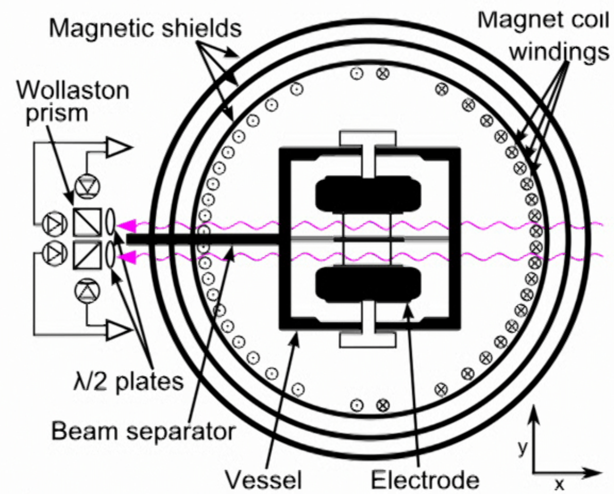
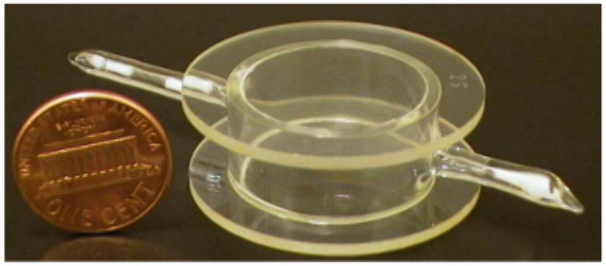
  - ~0.1 nHz (!!)





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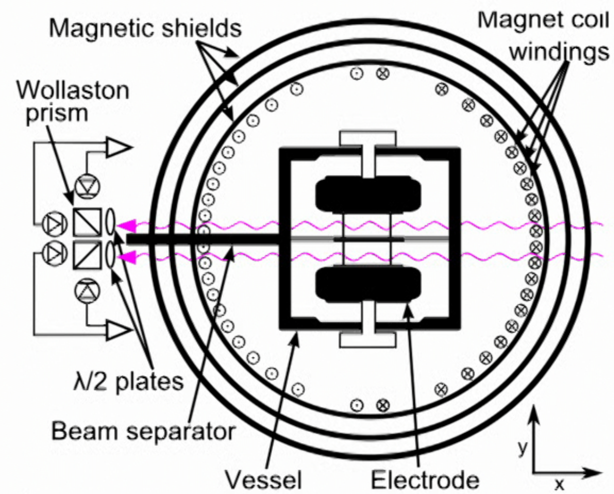
- Very high count rates (vapor cell) + very long coherence times (minutes) → extremely good frequency resolution
  - ~0.1 nHz (!!)
- Sensitive primarily to hadronic CPV
  - Example: $\theta_{QCD} < 1.5 \times 10^{-10}$ (single source assumption)
- ^{129}Xe EDM experiments are similar, but use different techniques



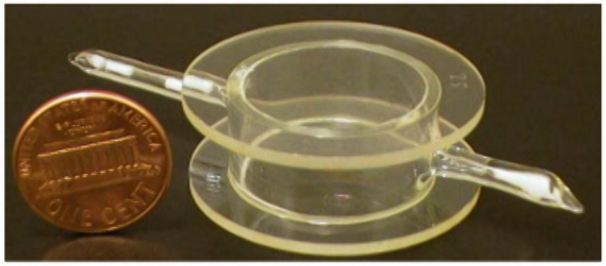


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Photo from Y. Chen FRIB Presentation, 2019
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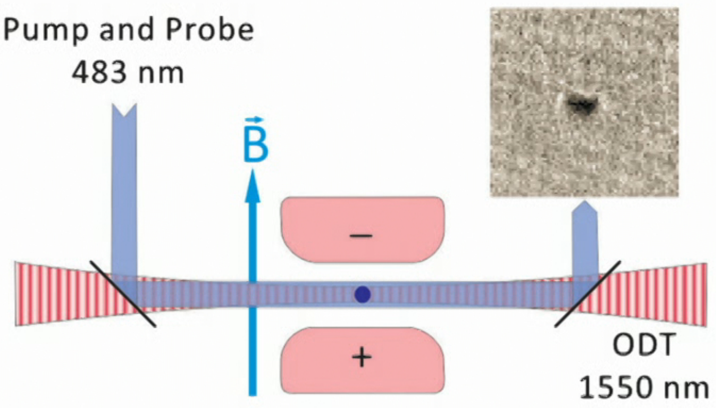
^{225}Ra EDM

- Argonne National Lab
- ^{225}Ra, $I = 1/2$
  - Large atomic enhancement
  - Large static octupole deformation, ~1,000x more intrinsic (nuclear) NSM sensitivity vs. Hg
  - Challenges: $t_{1/2}$ ~ 2 weeks, no vapor cells



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

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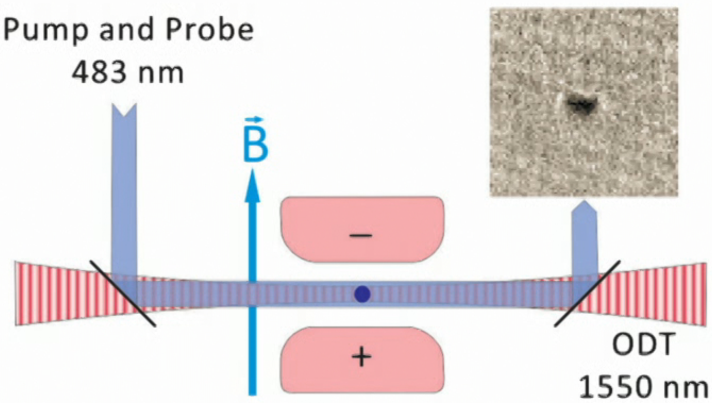
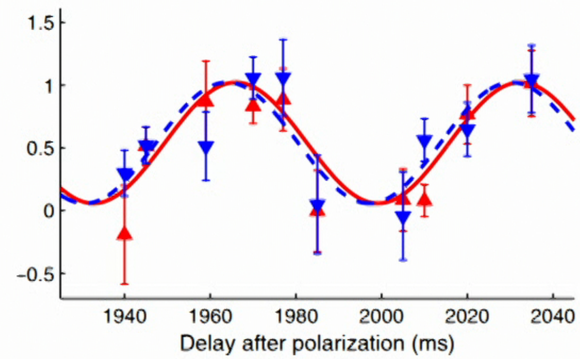
  - Trap in gas phase at ultracold temperatures
  - Low temperature → highly coherent





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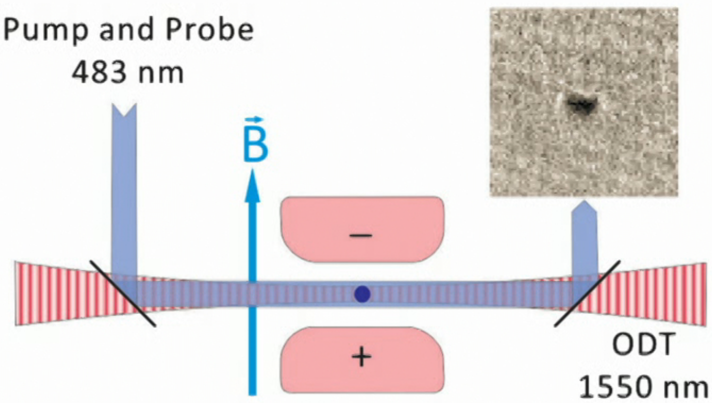
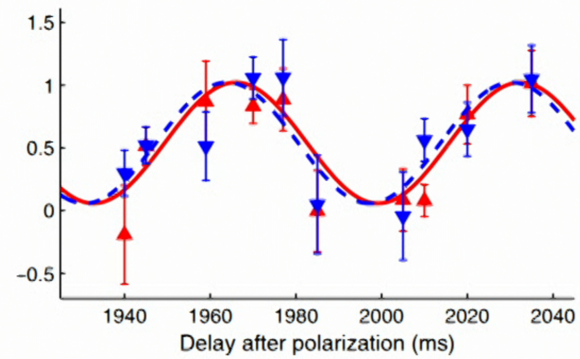
  - Challenges: $t_{1/2}$ ~ 2 weeks, no vapor cells
- Laser-coolable
  - Trap in gas phase at ultracold temperatures
  - Low temperature → highly coherent
- ^{171}Yb @ USTC
  - "Test bed" for ^{225}Ra
  - Similar structure, not radioactive
  - Recently demonstrated some advanced quantum methods for EDM measurement
  - arXiv: 2207.08140





Ra EDM @ ANL
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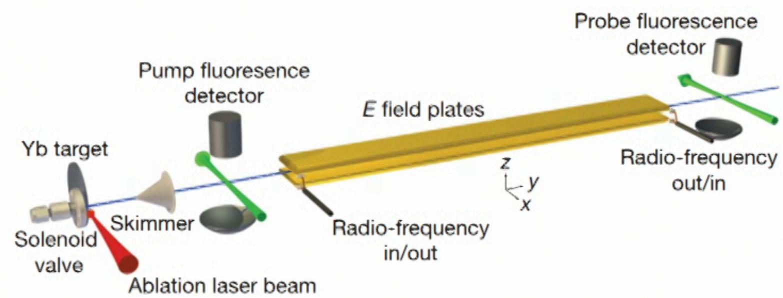
YbF

- Imperial College London
- ^{174}YbF has single unpaired valence electron, no nuclear spin
  - Sensitive to eEDM
- Spin precession in pulsed supersonic beam



J. J. Hudson, D. M. Kara, I. J. Smallman, B. E. Sauer, M. R. Tarbutt, and E. A. Hinds, Nature 473, 493 (2011)

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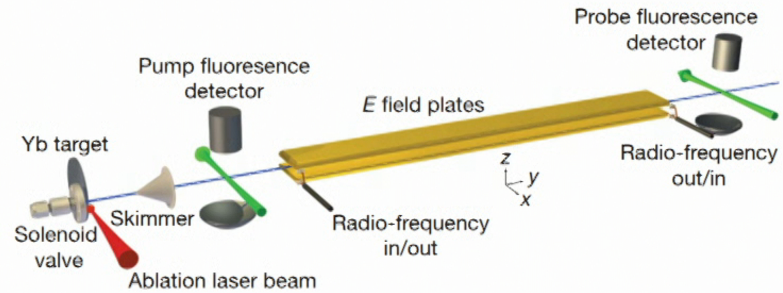
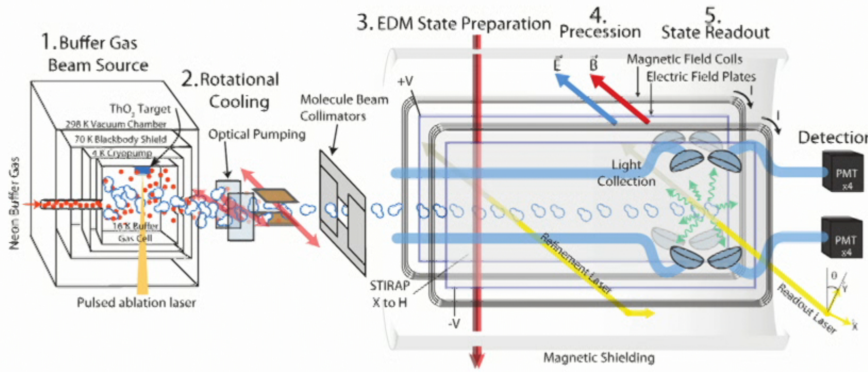
- ^{174}YbF has single unpaired valence electron, no nuclear spin
  - Sensitive to eEDM
- Spin precession in pulsed supersonic beam
- First to beat atomic experiments
  - Berkeley, Tl, 2002
- $|d_e| < 1.1 \times 10^{-27}$ e cm (2011)
  - Statistics limited
- Being upgraded (more later)

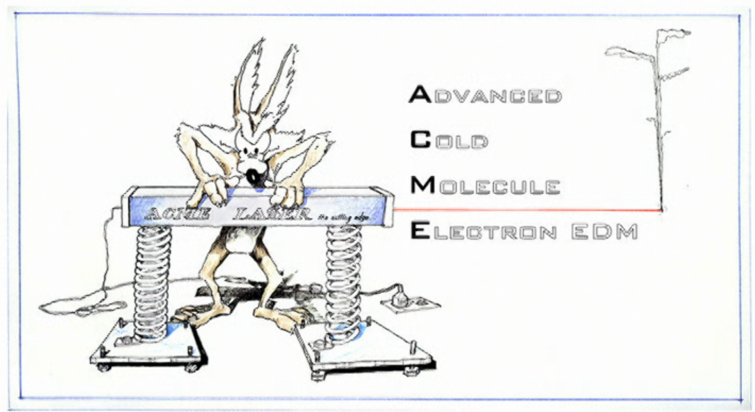


J. J. Hudson, D. M. Kara, I. J. Smallman, B. E. Sauer, M. R. Tarbutt, and E. A. Hinds, Nature 473, 493 (2011)

ThO

- ACME Collaboration (Harvard, Chicago, Northwestern)
- Spin precession in cryogenic beam





Beam Experiments

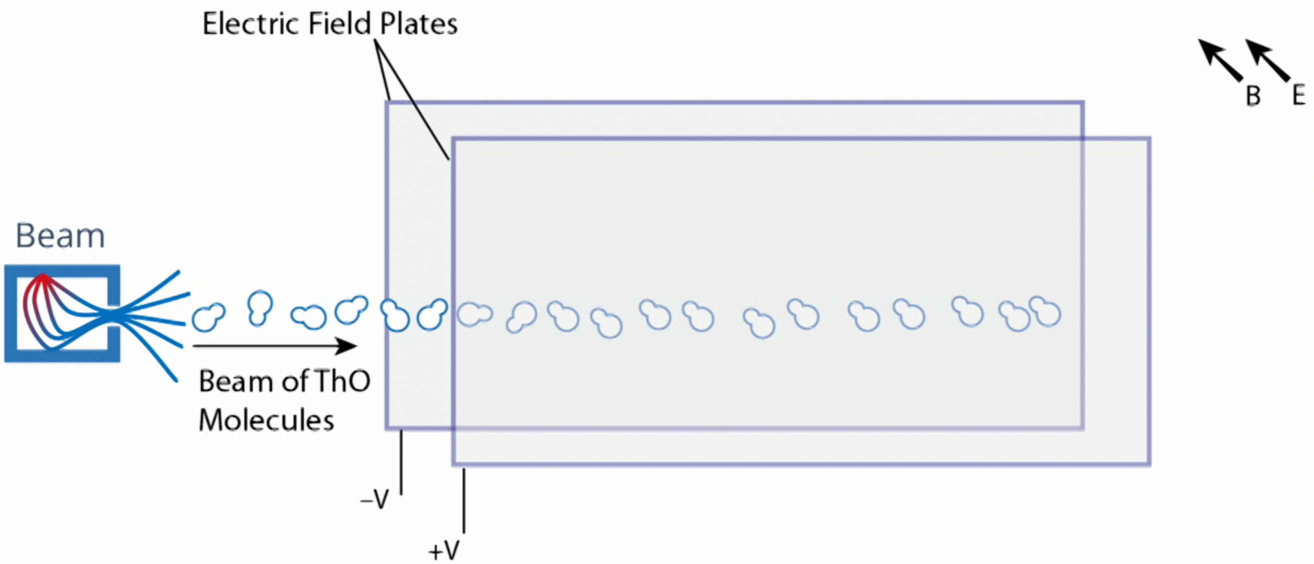
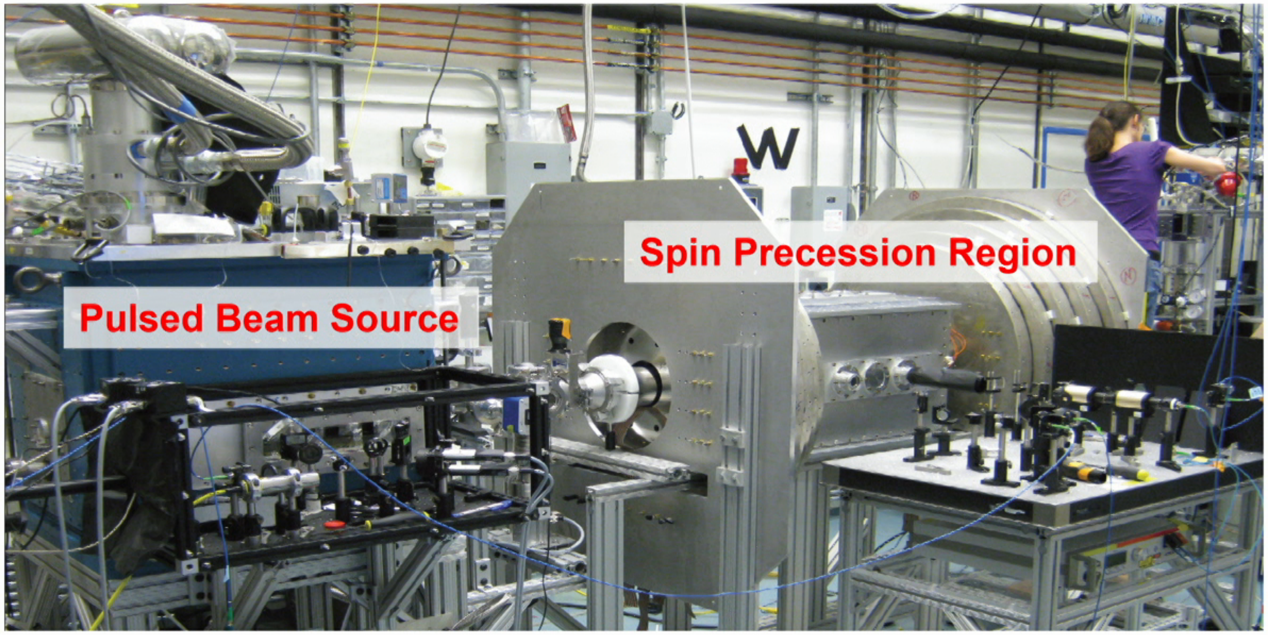


Photo of ACME Gen I



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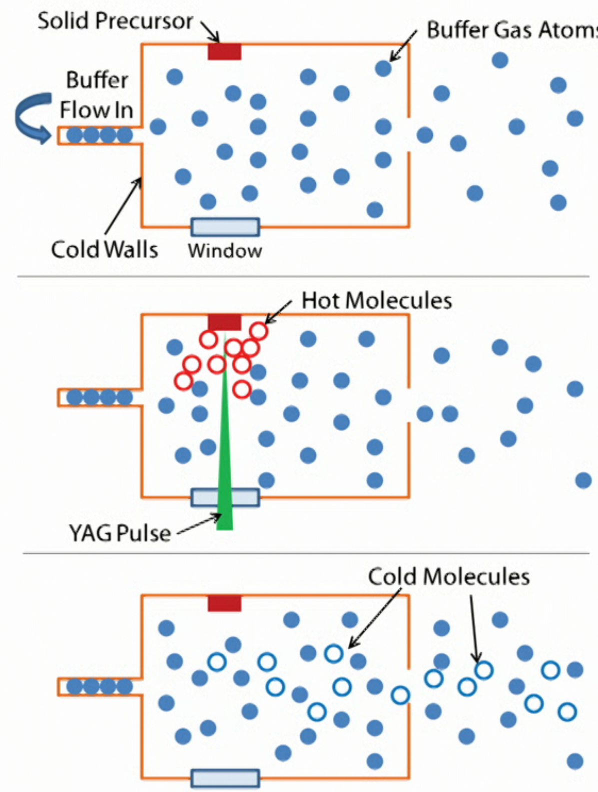
Cryogenic Buffer Gas Beams

- 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
- First step for molecular laser cooling as well (more on that later)



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

Parity Doublets

- Certain molecules have *parity doublets*
  - Each rotational state is split into a doublet of opposite parity states, split by <100 MHz
  - Small splittings mean high polarization in ~100 V/cm

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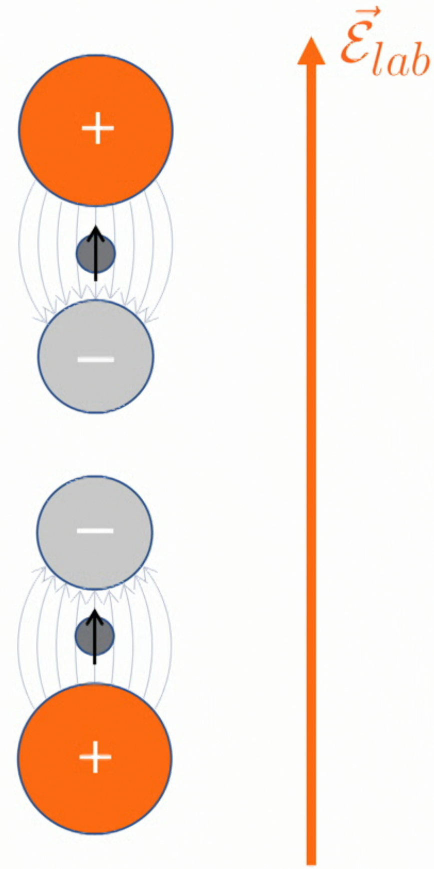
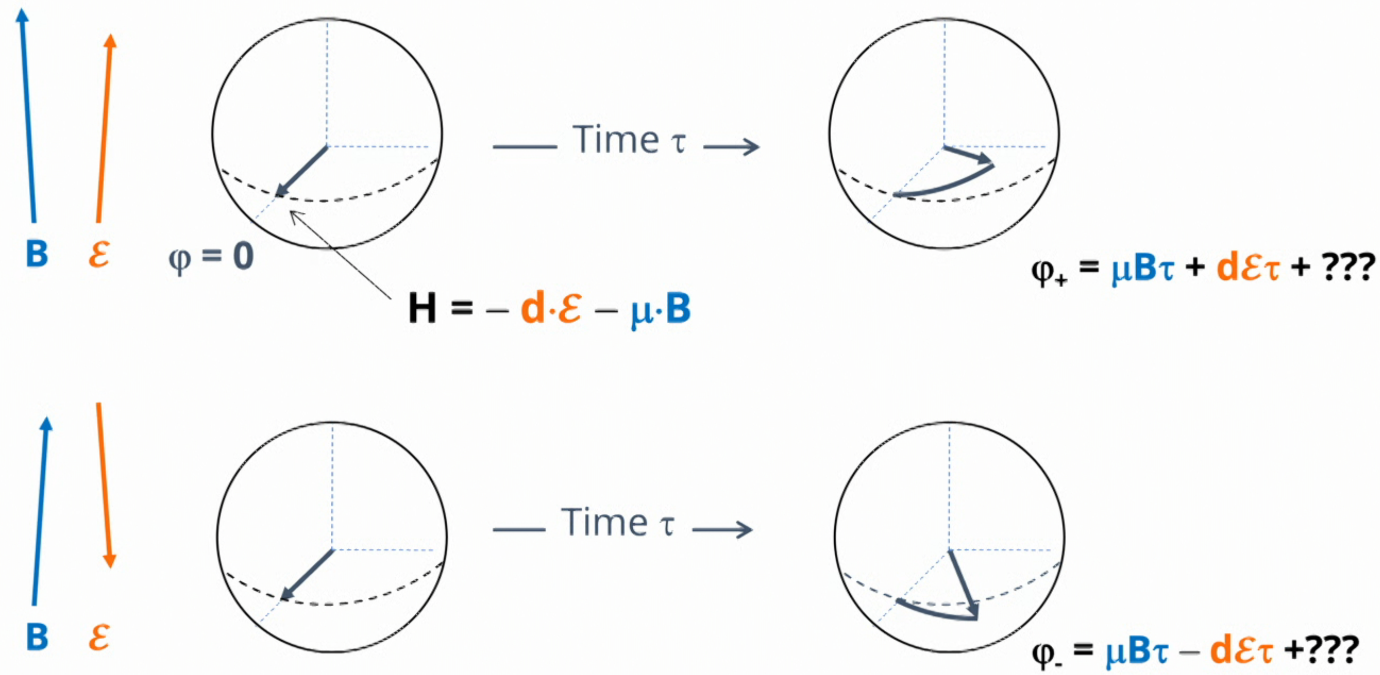
- Gives rise to two, fully-polarized states with opposite alignment of internal fields



Nothing is perfect...



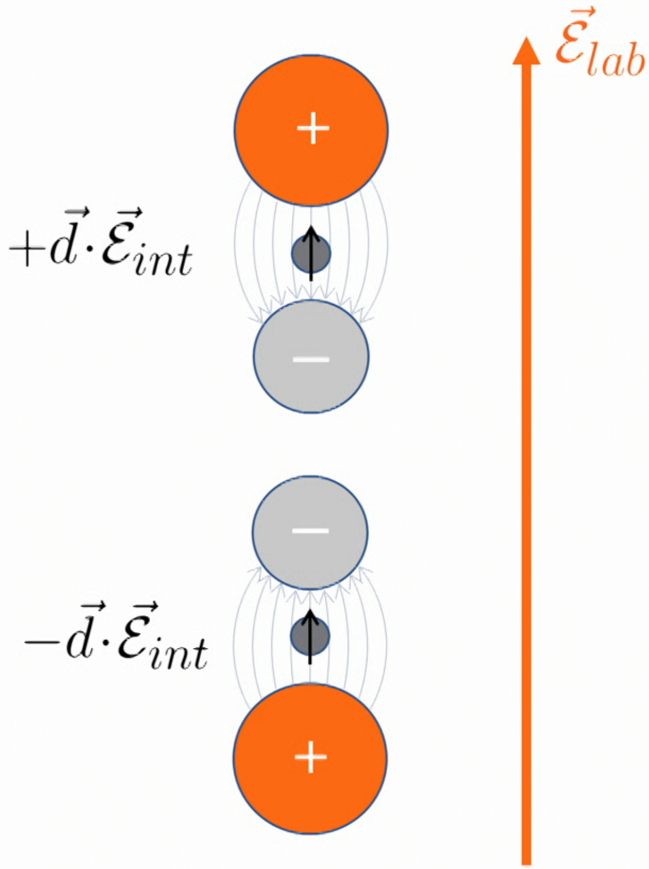
$d\mathcal{E} / \mu B < 10^{-6}$

$\Delta\varphi \propto d\mathcal{E}\tau + ???$

38

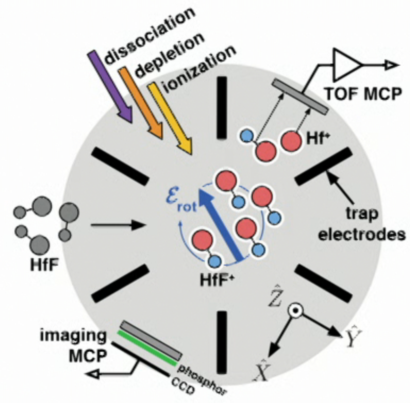
Parity Doublets

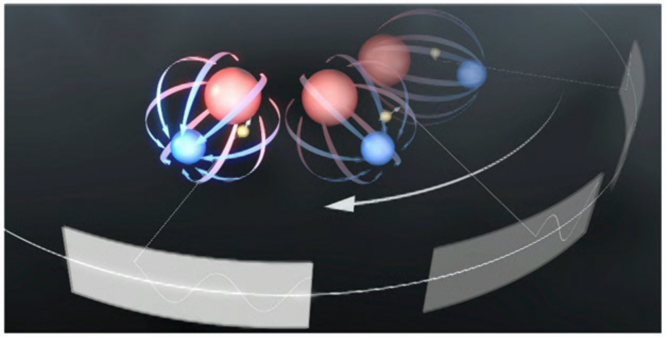
- Certain molecules have *parity doublets*
  - Each rotational state is split into a doublet of opposite parity states, split by <100 MHz
  - Small splittings mean high polarization in ~100 V/cm
- Gives rise to two, fully-polarized states with opposite alignment of internal fields
- Change EDM shift without changing external fields – just tune lasers to address different states
- Very powerful for rejection of systematic errors



HfF+

- JILA and CU Boulder
- Spin precession in molecular ion trap
  - Rotating electric field prevents charged ions from escaping
- Molecular structure with parity doublets
  - Critical – can't reverse electric field! Trap → Anti-trap

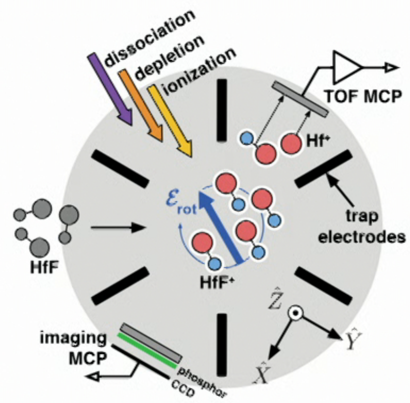


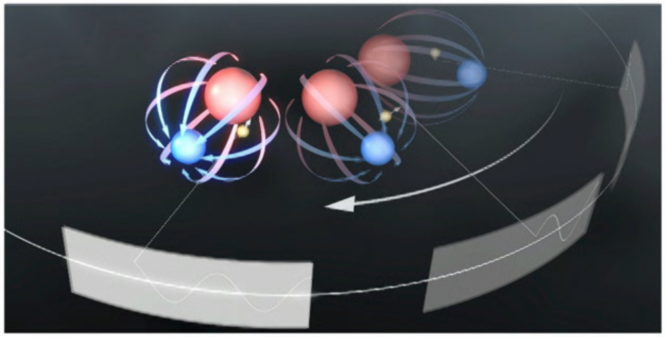


W. B. Cairncross, D. N. Gresh, M. Grau, K. C. Cossel, T. S. Roussy, Y. Ni, Y. Zhou, J. Ye, and E. A. Cornell, Phys. Rev. Lett. **119**, 153001 (2017)

HfF^+

- JILA and CU Boulder
- Spin precession in molecular ion trap
  - Rotating electric field prevents charged ions from escaping
- Molecular structure with parity doublets
  - Critical – can't reverse electric field! Trap → Anti-trap
- Long coherence time
- $|d_e| < 1.3 \times 10^{-28}$ e cm (2017)
- Being upgraded!
  - Demonstrated order of magnitude improvement, result in next ~year





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Part III: Selected Ongoing and Future Developments

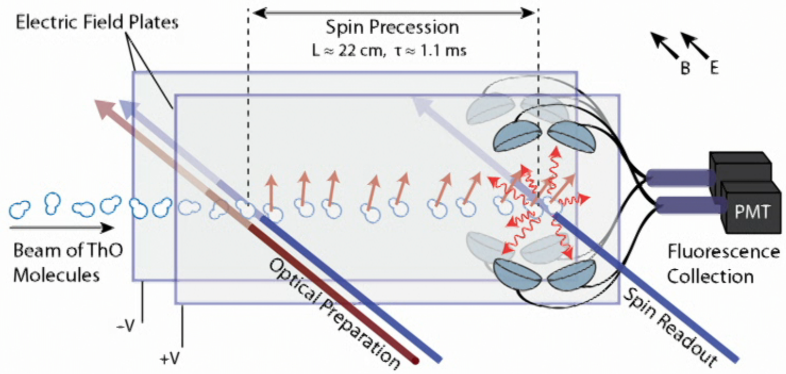
Not a review, but instead using them as a vehicle to explain experimental methods under development

We will not discuss all ongoing and future developments!

Laser cooling

Motivation for molecular 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



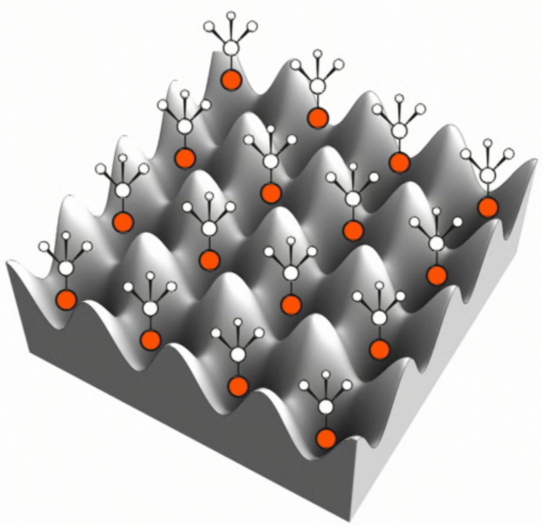
10^6 molecules
100 s coherence time
Heavy, deformed nucleus
Quantum control
Robust error rejection
Two weeks integration

→

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

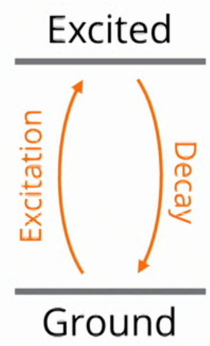
↓

Future orders-of-magnitude improvements from quantum-enhanced metrology, highly exotic nuclei, ...
+ ~5-10 year time scale?

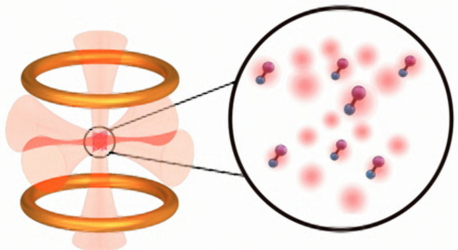


Laser cooling molecules

- Apply forces by photon scattering
- Requires many (~10^5) cycles of absorption, spontaneous decay



Three Examples

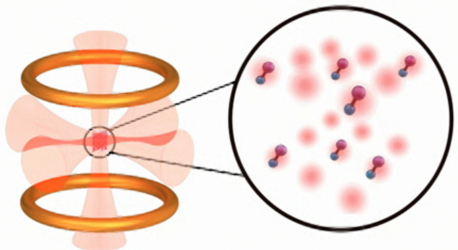


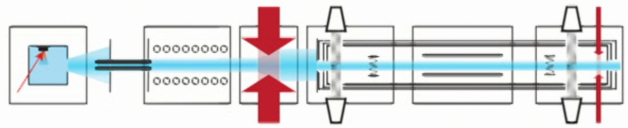
YbF

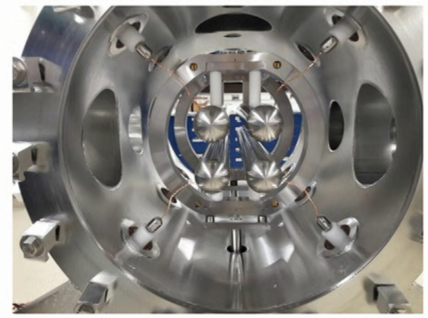
- eEDM @ Imperial College London
- Laser cooling, other upgrades demonstrated
- X. Alauze *et al.*, Q. Sci. & Tech. 6, 044005 (2021)
- N. J. Fitch *et al.*, Q. Sci. & Tech. 6, 014006 (2021)

Several more laser cooling examples later

Three Examples







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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)

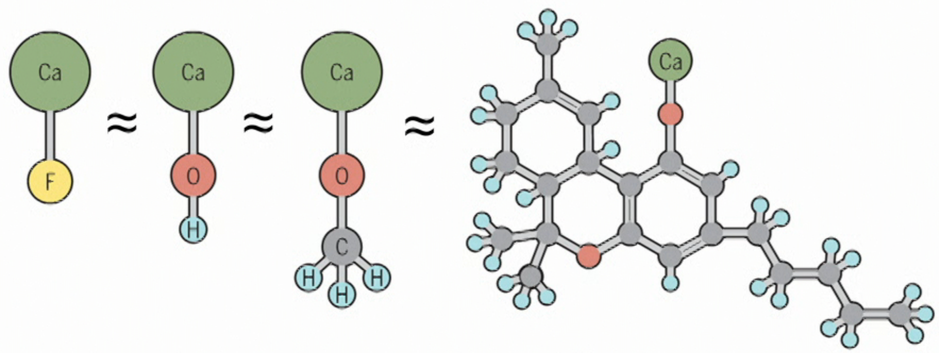
TlF

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

Several more laser cooling examples later

Polyatomic Molecules

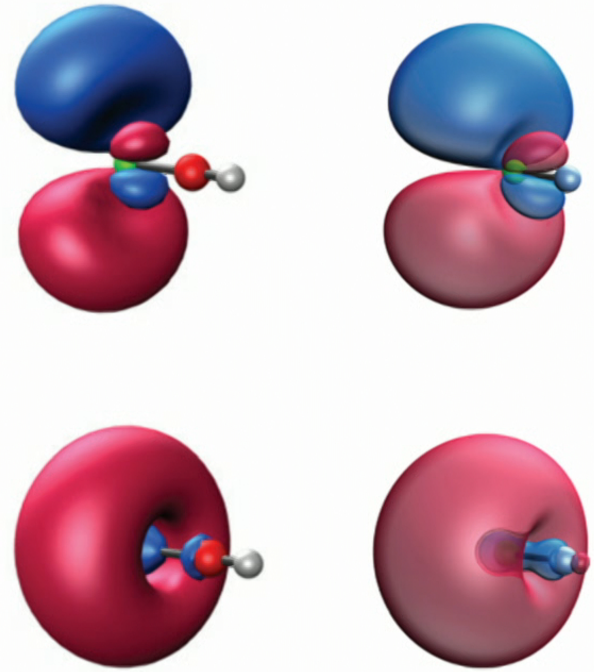
- Additional degrees of freedom to engineer desirable properties
  - Electric and magnetic field interactions
  - High polarizability
  - Species in ligand
  - Frequencies of rotation and vibration
  - ...
- Review: 2008.03398



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

Polyatomic Molecules

- Many bonding partners "bond similarly"
  - – F ≈ –OH, – –CCH, – OCH_3, …
  - Not in general, but for s^2 atoms it typically holds
    - Ca, Sr, Ba, Yb, Ra
  - Since electron wavefunction is metal-centered, ligand matters much less
- Similar electronic structure implies similar:
  - Laser cooling/photon cycling
  - CPV sensitivity
  - Measurement methods

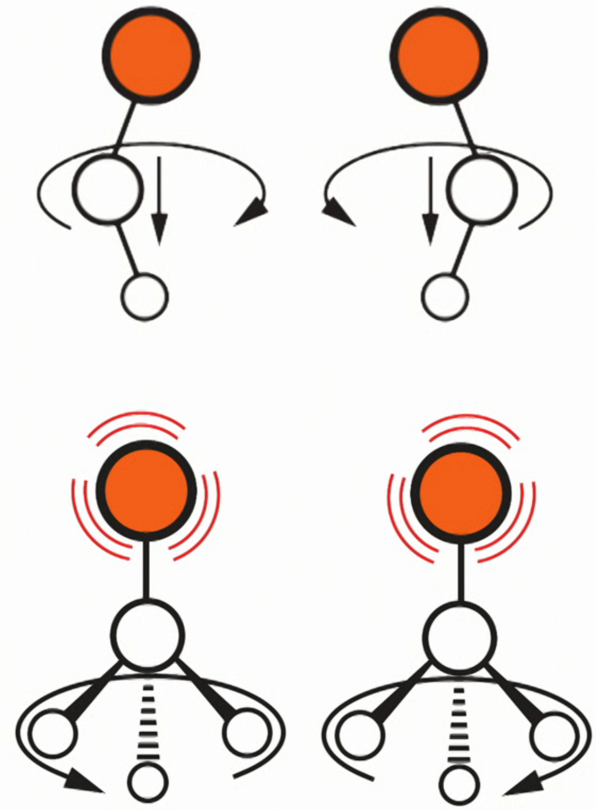


CaOH CaF

T. A. Isaev and R. Berger, PRL 116, 063006 (2016) T. A. Isaev, A. V. Zaitsevskii, and E. Eliav, J. Phys. B 50, 225101 (2017)
M. V. Ivanov, F. H. Bangerter, and A. I. Krylov, Phys. Chem. Chem. Phys. 21, 19447 (2019)

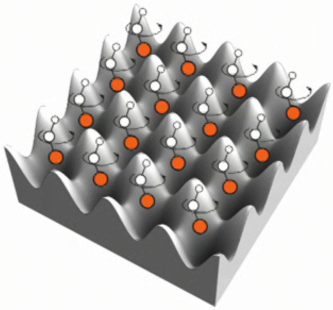
Polyatomic Molecules

- Polyatomics *generically* have parity doublets
  - (>3 atoms)
  - Arise from symmetry-lowering mechanical modes
  - As opposed to diatomics, which rely on "exotic" electronic structure
  - Generically realize high polarizability and systematic robustness advantages
- Useful – Laser-coolable diatomic molecules don't have parity doublets!
- Available for any atomic species



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

Three Examples

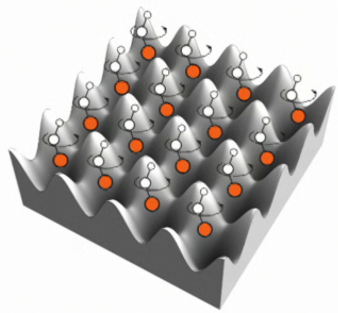


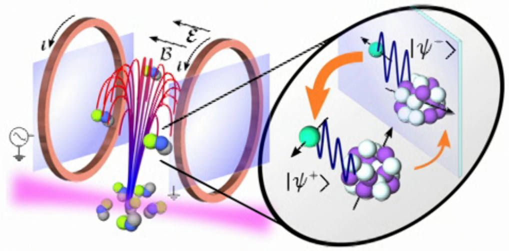
CaOH/SrOH/YbOH

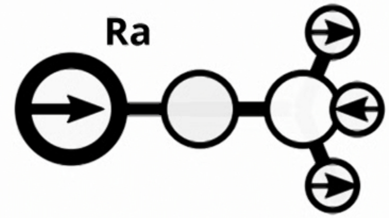
- Combine laser cooling, high polarizability
- PolyEDM: eEDM search, NRH, Doyle, Steimle, Vutha
- ^{173}YbOH MQM @ Caltech
- I. Kozyryev and NRH, PRL 119, 133002 (2017)

… Many, many more!

Three Examples







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- I. Kozyryev and NRH, PRL 119, 133002 (2017)

MgNC

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

$RaOCH_3^+$

- Combines deformed nucleus with ion trapping
- 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!

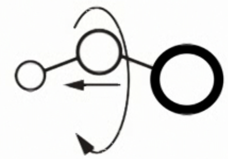
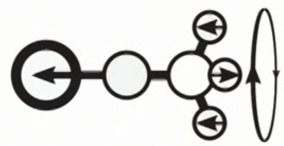
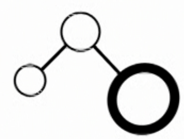
Radium

- Ra is highly versatile
  - Ra has well-understood octupole deformation
  - Ra, Ra^{+}, Ra molecules can be laser cooled
  - Venue to combine laser cooling, polyatomics, ion trapping, deformed nuclei

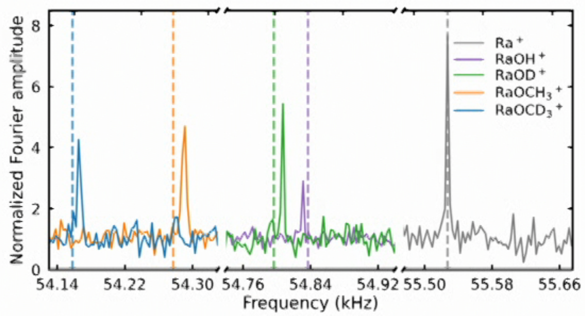
Radium Molecular Ions

- RaF^+ → Not highly polarizable
  - No parity doubling
  - True for any RaX^+ diatomic
- Radium polyatomics
  - High polarizability due to symmetry-lowering motions
  - Ability to tune properties to match experimental needs
  - Quantum logic, clock measurement, spin precession
- Already created and trapped in Jayich Lab @ UCSB
- A *single* trapped molecular ion could probe the frontiers of symmetry violation

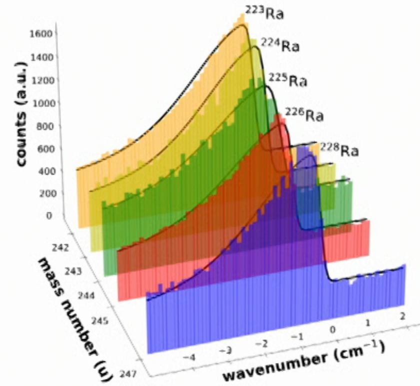
P. Yu and NRH, PRL 126, 023003 (2021)
M. Fan *et al.*, PRL126, 023002 (2021)

	Doubling	*Energy*	*Lifetime*
 **$RaOH^+$**	~10 MHz	~ 10 THz	~1 s
 **$RaOCH_3^+$**	~1 kHz	~160 GHz	>> 1 h
 **$RaSH^+$**	~5 MHz	~300 GHz	>1 m

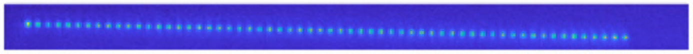
Progress in the last ~year



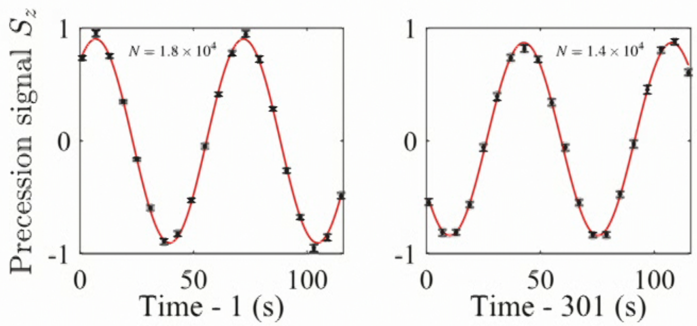
Creation, trapping, cooling, control of radioactive molecular ions



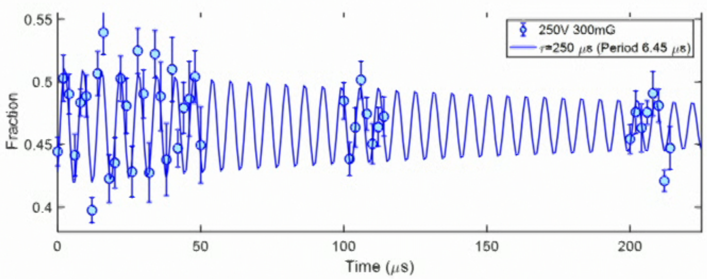
Precision spectroscopy of radioactive molecules



Quantum-controlled ultracold molecules



Atomic EDM searches with long coherence times in optical traps



Electron spin precession in optically-trapped polyatomic molecules

Fan *et al.*, PRL 126, 023002 (2021)
Doyle Group @ Harvard
PolyEDM Collaboration

Udrescu *et al.*, PRL 127, 033001 (2021)
Zheng *et al.*, arXiv:2207.08140 (2022)

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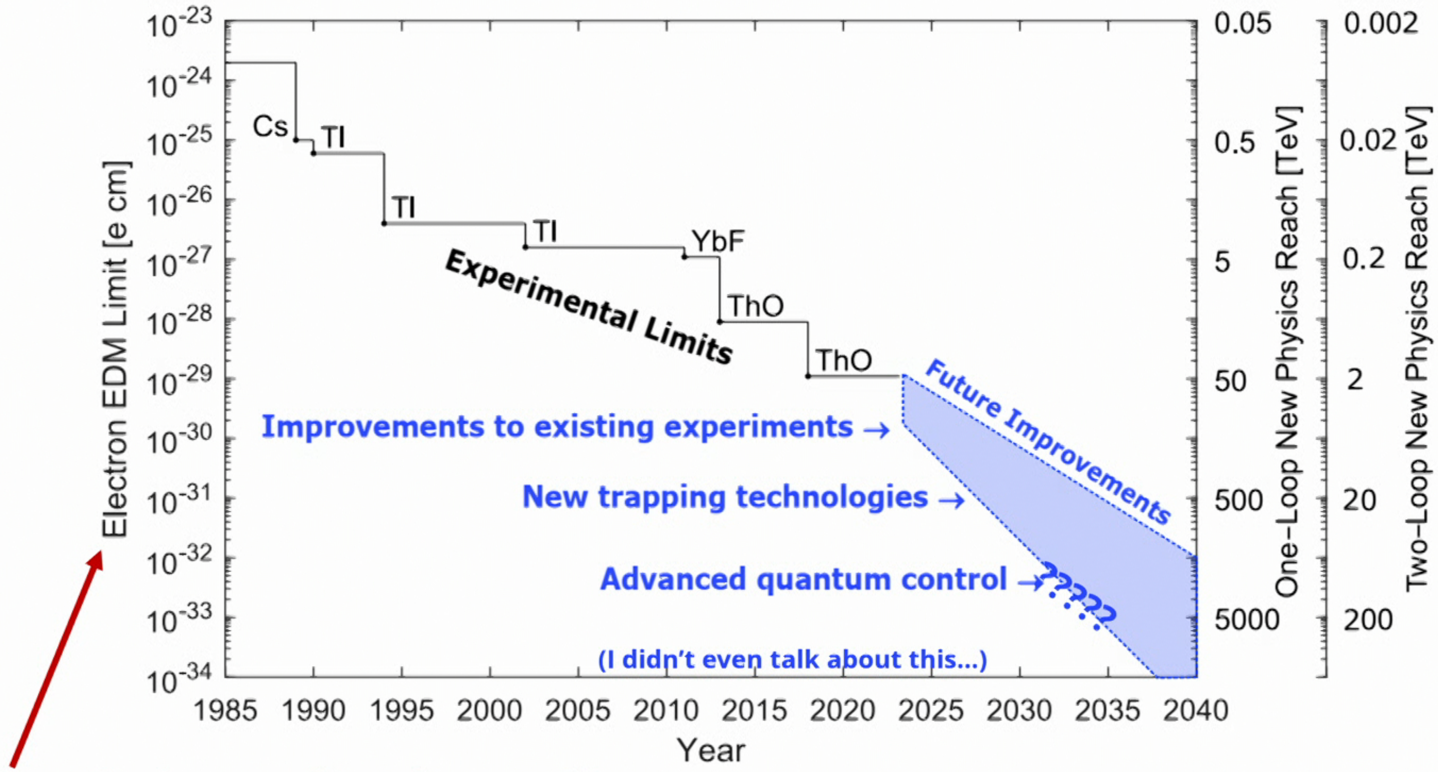
A Positive Outlook



Similar gains in a very broad range of areas –
hadronic CPV, ultralight DM, precision electroweak, ...

From Snowmass EDM whitepaper, arXiv:2203:08103

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
  - *hutzlerlab.com* → Teaching → TRISEP 2022
- **Interpretation of EDM limits**
  - See Safronova, Chupp, Snowmass reviews
  - J. Engel et al., Prog. Part. Nucl. Phys. 71, 21 (2013)
- **Email me!**