Caltech

Searching for New Particles and Forces with Polar Molecules

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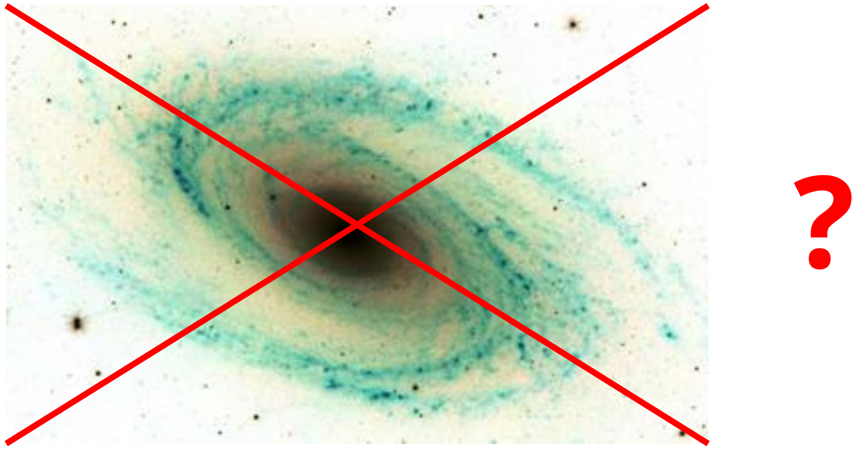
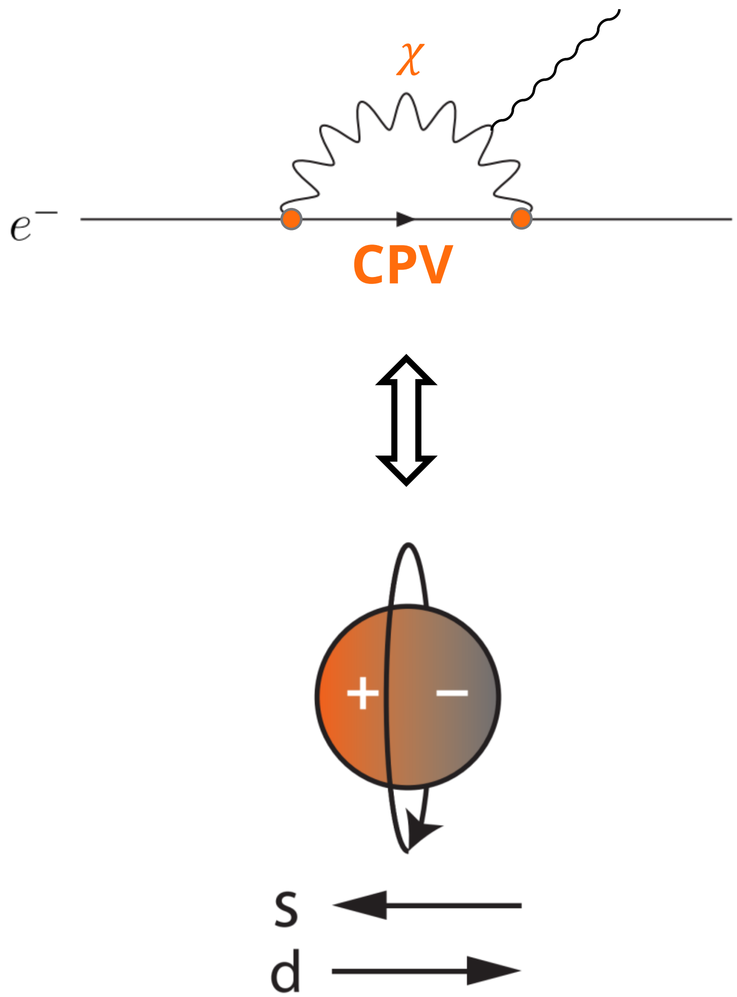
An Asymmetric Universe

- The universe is made out of matter
- There is no free anti-matter in the universe
- Problem: all **known** physical laws treat them equally!
  - Where did matter come from?
  - Where did anti-matter go?
  - **There must be processes that favor matter over anti-matter**
- Baryon Asymmetry of the Universe (BAU)

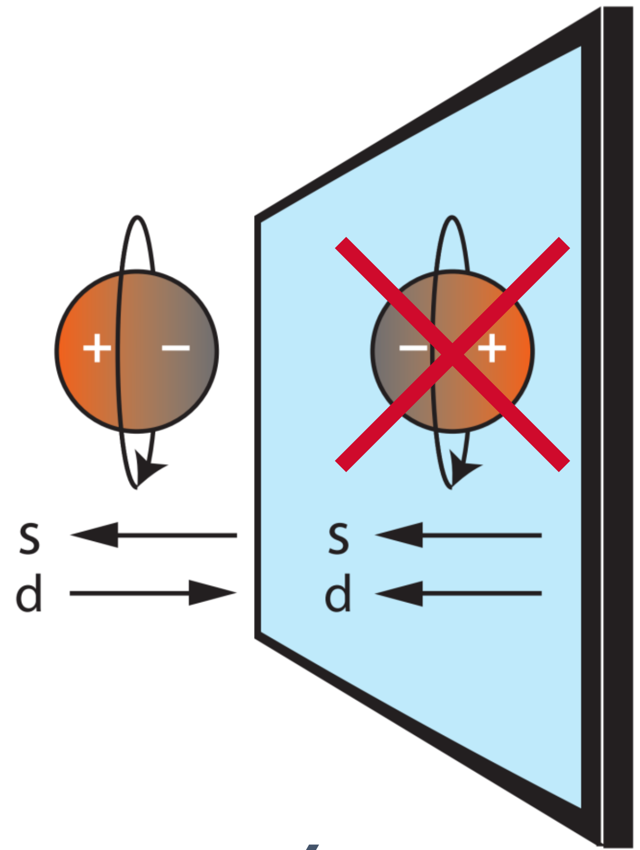


CP Violating Observables

- The BAU requires new CP-violating physics
  - Leads to CPV observables at **low energies**
    - (Also high energies)
- Classic example: permanent electric dipole moments (EDMs)
  - **The existence of EDMs requires CPV**
  - Other moments as well
    - I will use “EDM” to mean generic CPV electro-magnetic moments
  - **Generically sensitive to new CPV physics**



EDMs violate symmetries



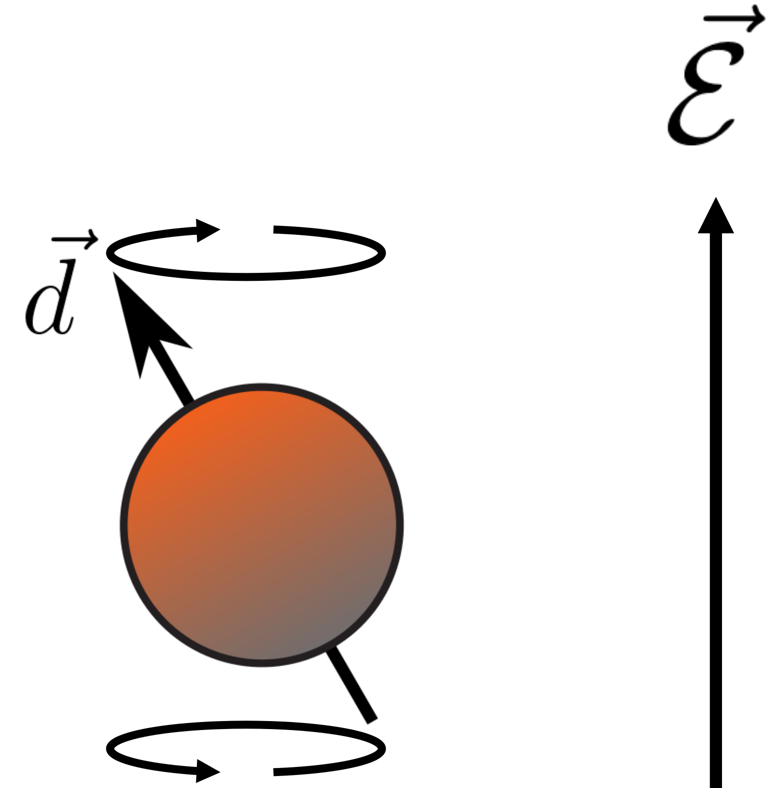
≠

EDMs violate P, T, CP*

(*Assuming conservation of CPT…)

Measuring EDMs

- An EDM experiences a torque in an electric field
  - This torque causes the spin to precess
- Experiment:
  - Initialize spin
  - Precession in electric field
  - Measure spin
  - Repeat…



$$\vec{\tau} = \vec{d} \times \vec{\mathcal{E}}$$

$$\omega = d\mathcal{E}/\hbar$$

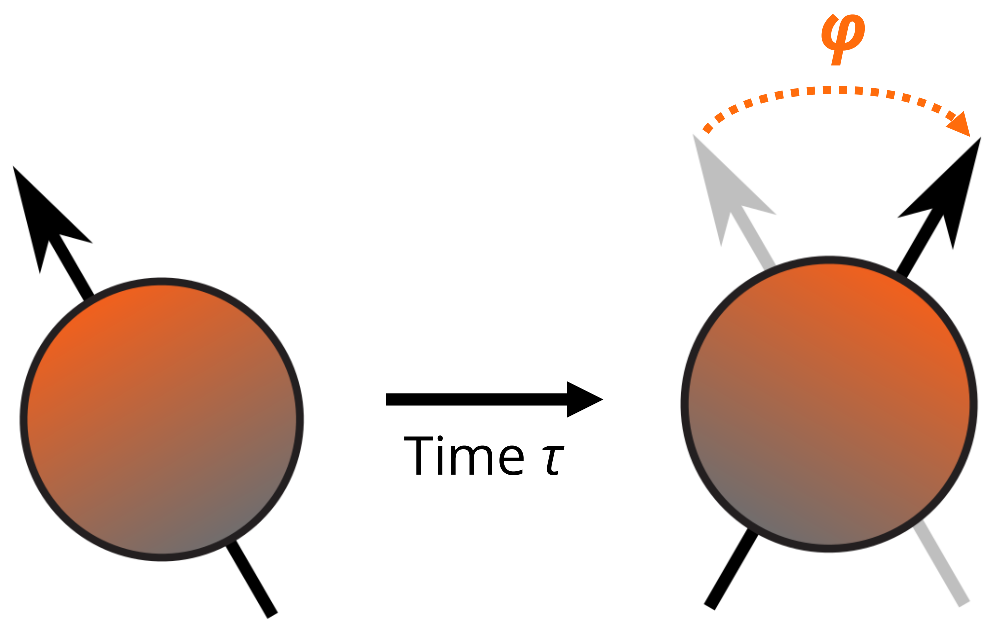
Sensitivity

- Experimental observable is angle φ (phase)

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

Want large electric field

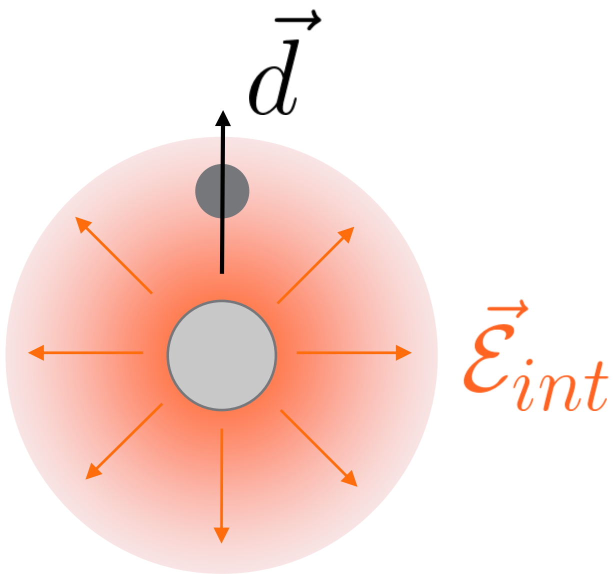
Want long interaction time

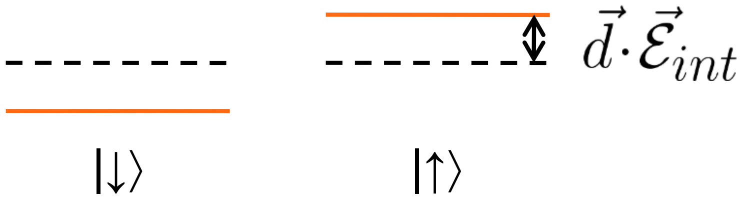


Electric field?

- Atoms/molecules have **extremely large** fields
  - $e/4\pi\epsilon_0 a_0^2 \sim$ GV/cm
  - Relativistic ~Z^3 enhancement
  - 10-100 GV/cm for heavy species
  - Maximum lab field ~100 kV/cm
- Permanent EDM causes splitting of energy levels
  - Induces permanent EDM in atom/molecule

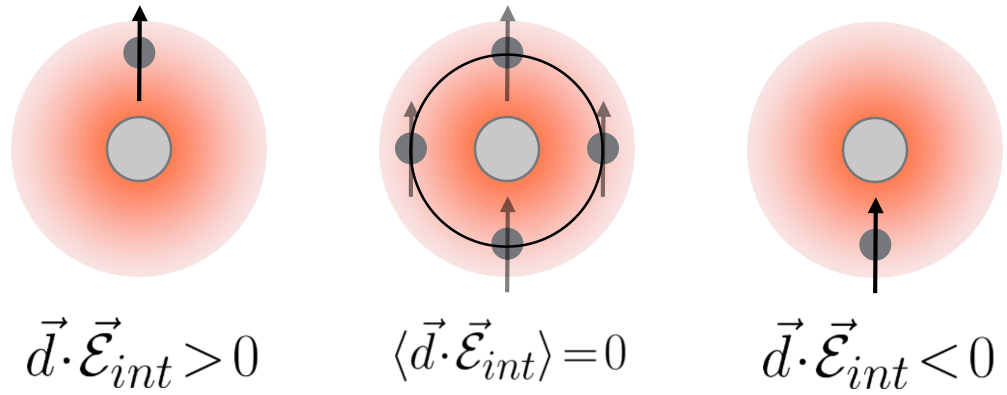
$$H = -\vec{d} \cdot \vec{\mathcal{E}}_{int}$$

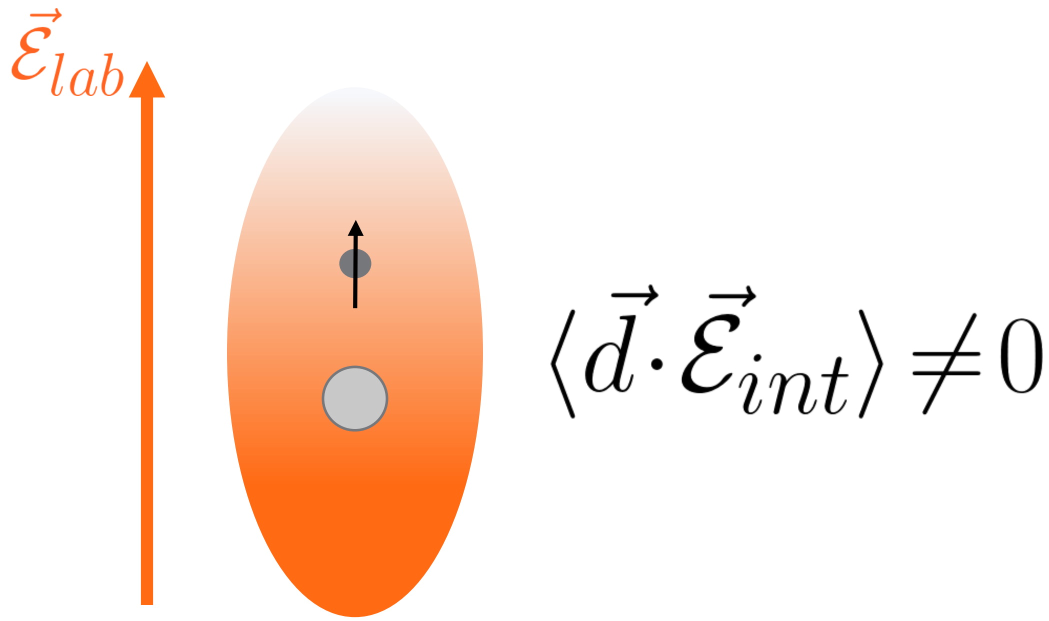




Polarization

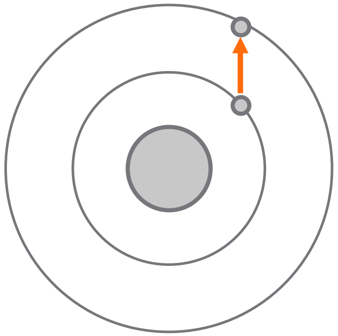
- Problem – constituents experiences zero average field!
  - Atom/molecule states always have this symmetry in free space
- **Solution: polarize**
  - Apply lab field to orient atom/molecule
  - Interaction no longer averages to zero
  - Sensitivity $\propto$ polarization P



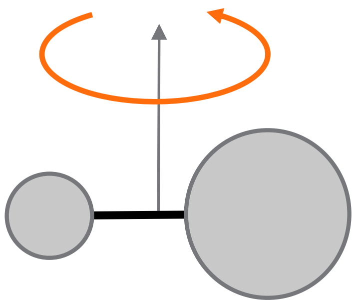


Atoms vs. molecules

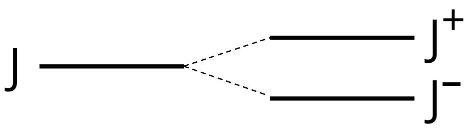
- Atoms
  - Δ ~ 10-100 THz (electronic)
  - P ~ 10^{-3} @ 100 kV/cm
- Molecules
  - Δ ~ 10 GHz (rotational)
  - P ~ $\mathcal{O}(1)$ @ 10 kV/cm
- **"Molecules are 1000x more sensitive"**
- Some molecules have **parity doublets**, Δ<10 MHz
  - Enables full polarization in small fields… and more!



Atoms
Δ ~ 100 THz



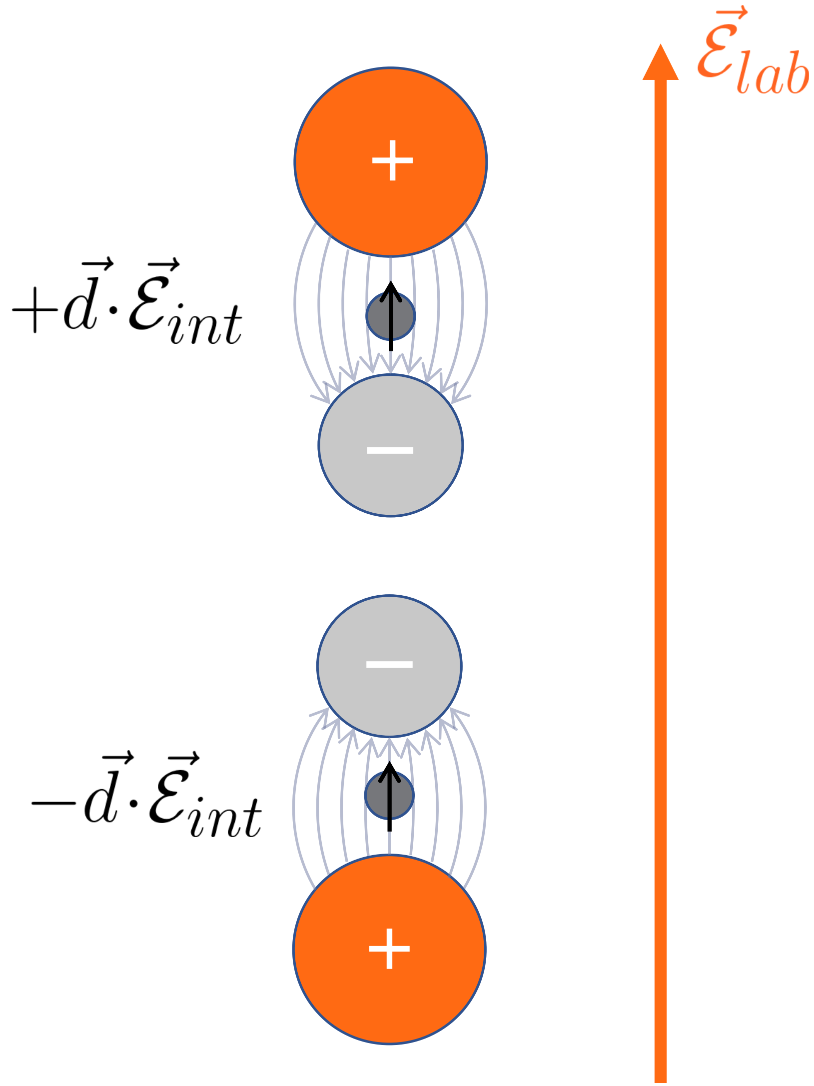
Molecules
Δ ~ 10 GHz



Parity Doubling
Δ ~ 10 MHz

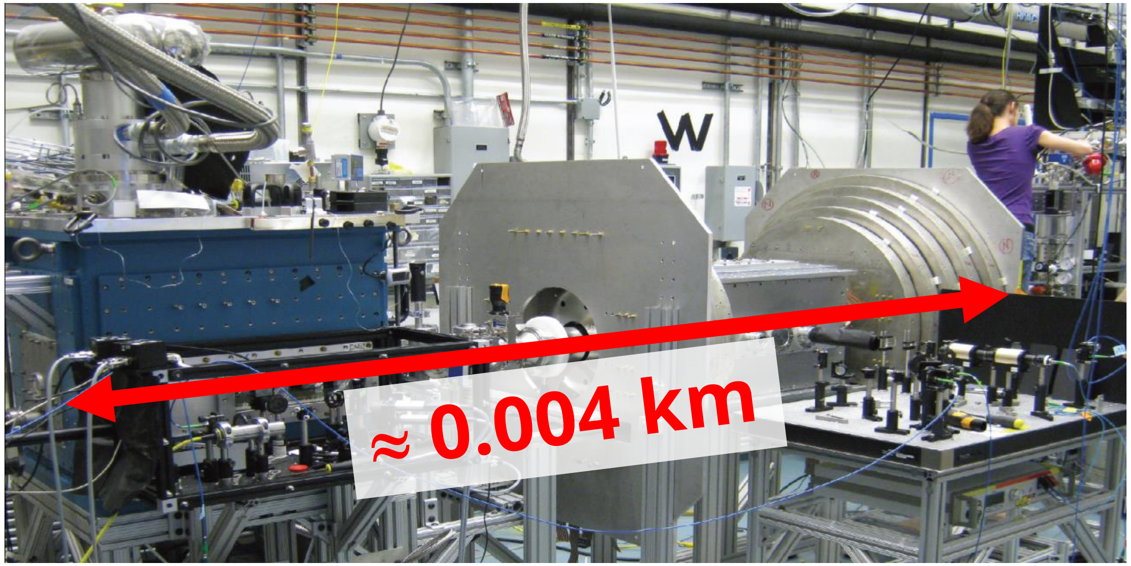
Internal Comagnetometers

- Parity doublets enable full polarization in the lab
  - **"Internal comagnetometer"**
  - Measure CPV in each state
  - Sensitivity equal/opposite!
  - Non-CPV effects cancel
  - **Suppresses important systematic effect**
  - Additional suppression from smaller fields
- Requires particular electronic structure
  - Arises from coupling between electron L and molecular rotation



Molecule State of the Art: ACME

- Harvard/Yale Collaboration
  - DeMille, Doyle, Gabrielse
- Molecular beam spin precession in ThO
- Combination of several new ideas and techniques
  - Cryogenic buffer gas beam
  - Internal comagnetometers
- Current best limit on the electron EDM (2nd gen.)
  - $|d_e| < 1.1 \times 10^{-29}$ e cm
  - Statistics limited
- **Already probing the TeV scale – beyond the LHC**
  - $>$10-100 M_{higgs}



ACME Collaboration, Nature **562**, 355 (2018)

Where can we improve?

- Shot noise limited sensitivity

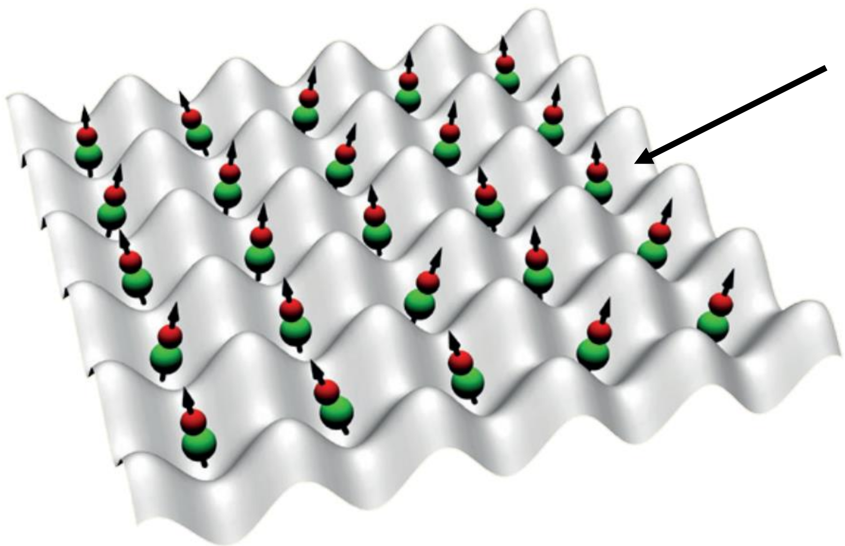
$$\delta d_{\mathrm{e}} = \frac{\hbar}{2\mathcal{E}_{\mathrm{eff}}\tau\sqrt{N}}$$

Beams have τ ~ 1 ms

Traps can have τ > 1 s...

Where we are going…

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



Heavy, polar molecule sensitive to new physics

Figure adapted from A. J. Daley, Nature **501**, 497 (2013)

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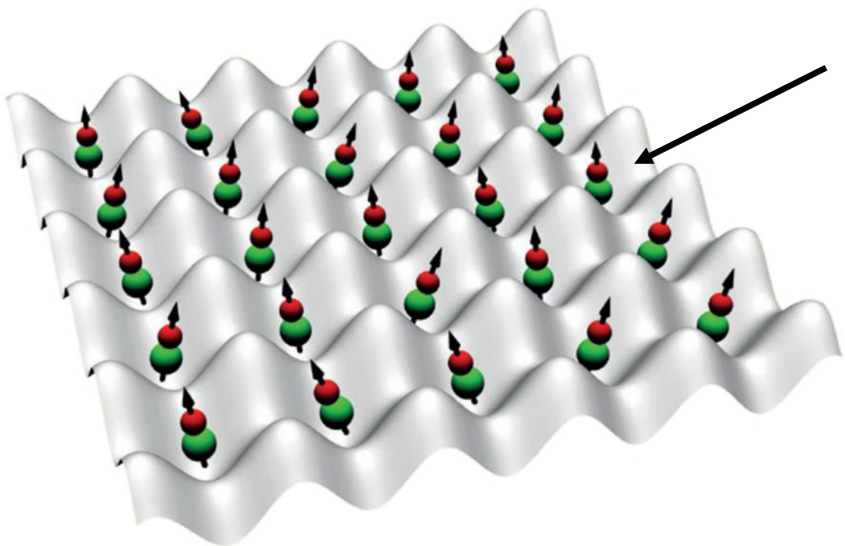
$M_{new\ phys} \sim 1{,}000$ TeV (!)



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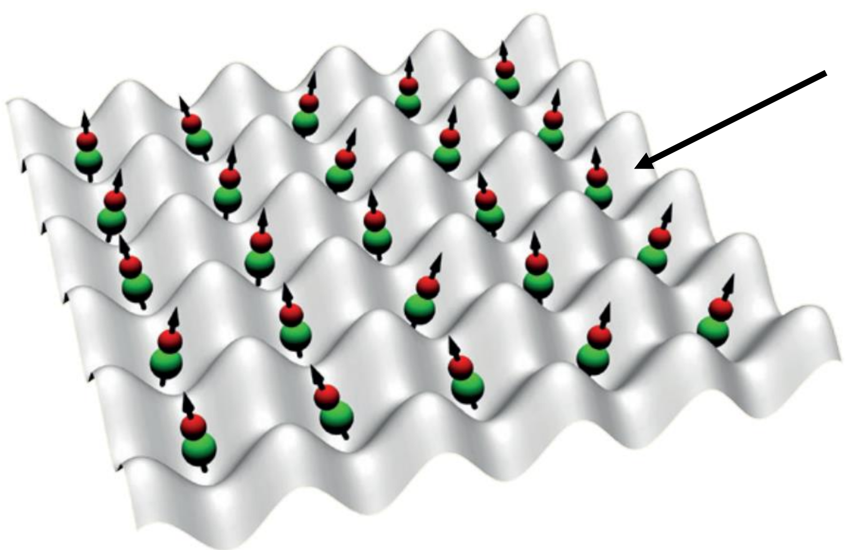
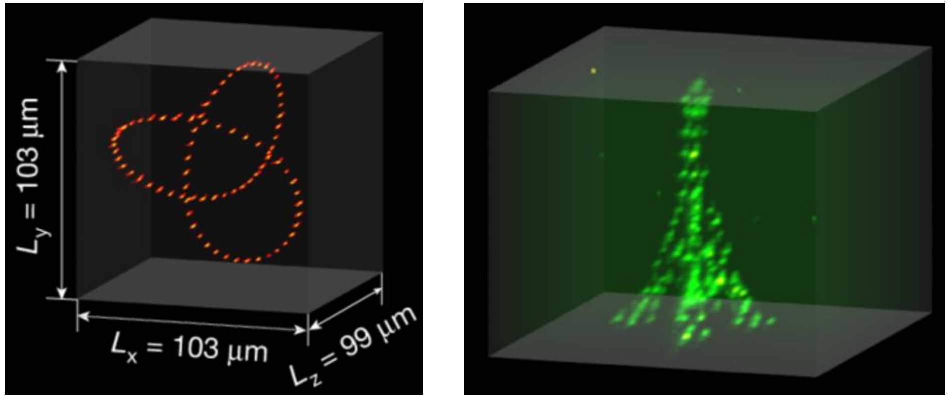


Heavy, polar molecule sensitive to new physics

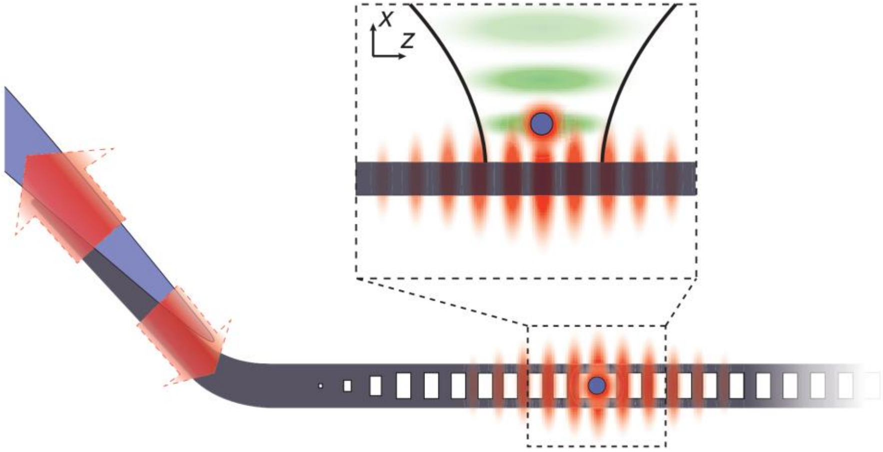
…but how do we get there?

Figure adapted from A. J. Daley, Nature **501**, 497 (2013)

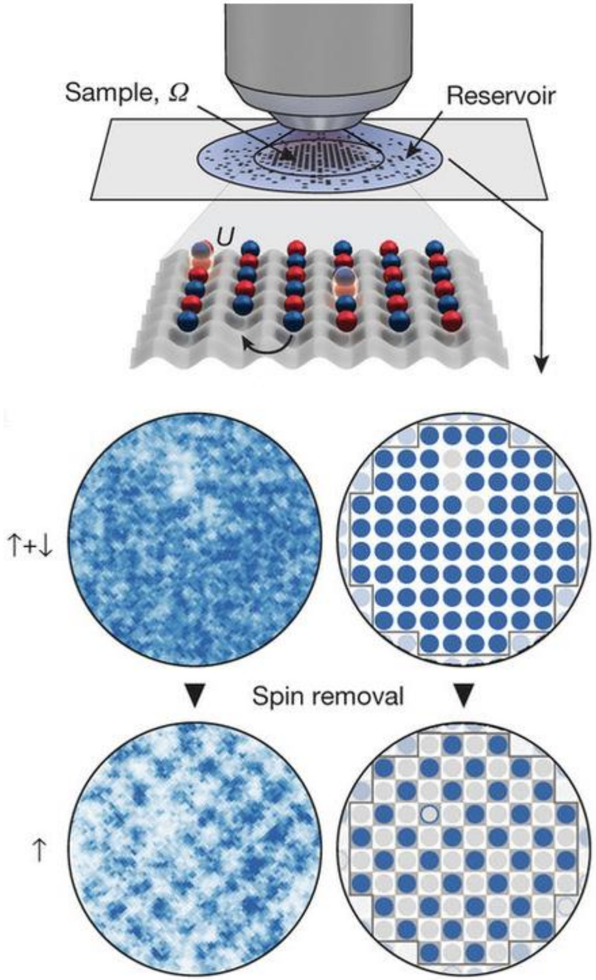
Quantum Control with Atoms



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

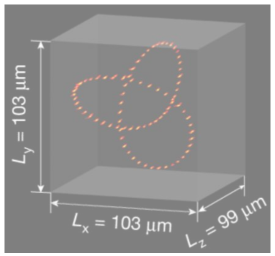


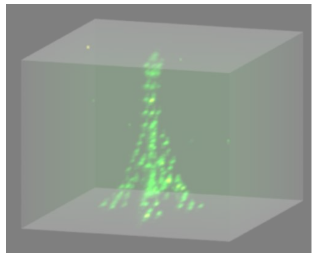
T. G. Tiecke, *et al.*, Nature **508**, 241 (2014).

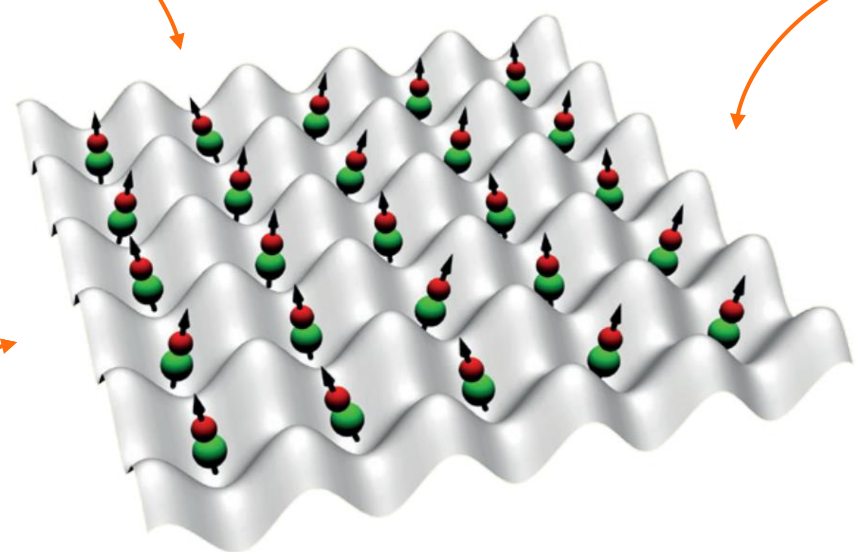


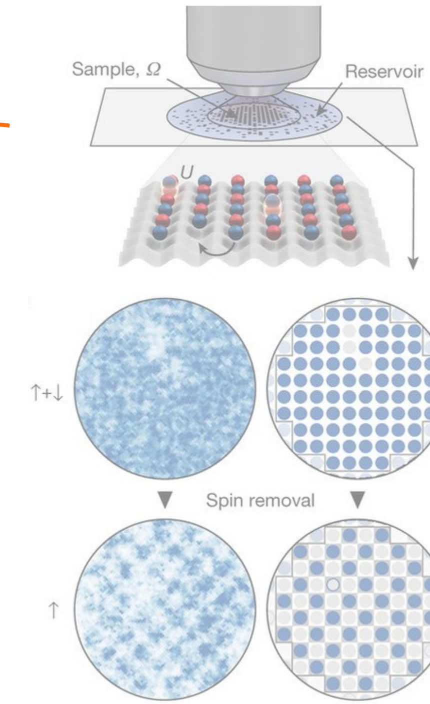
A. Mazurenko *et al*., Nature 545, 462-466 (2017)

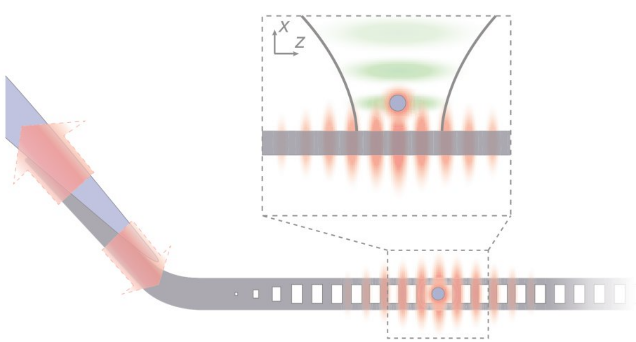
Quantum Control with Molecules?







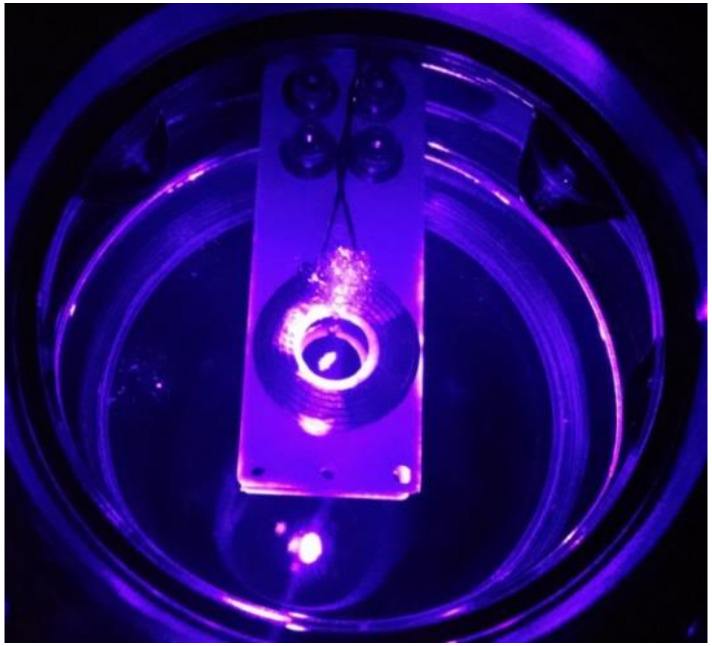




Many people are working on this,
with many recent and exciting results!

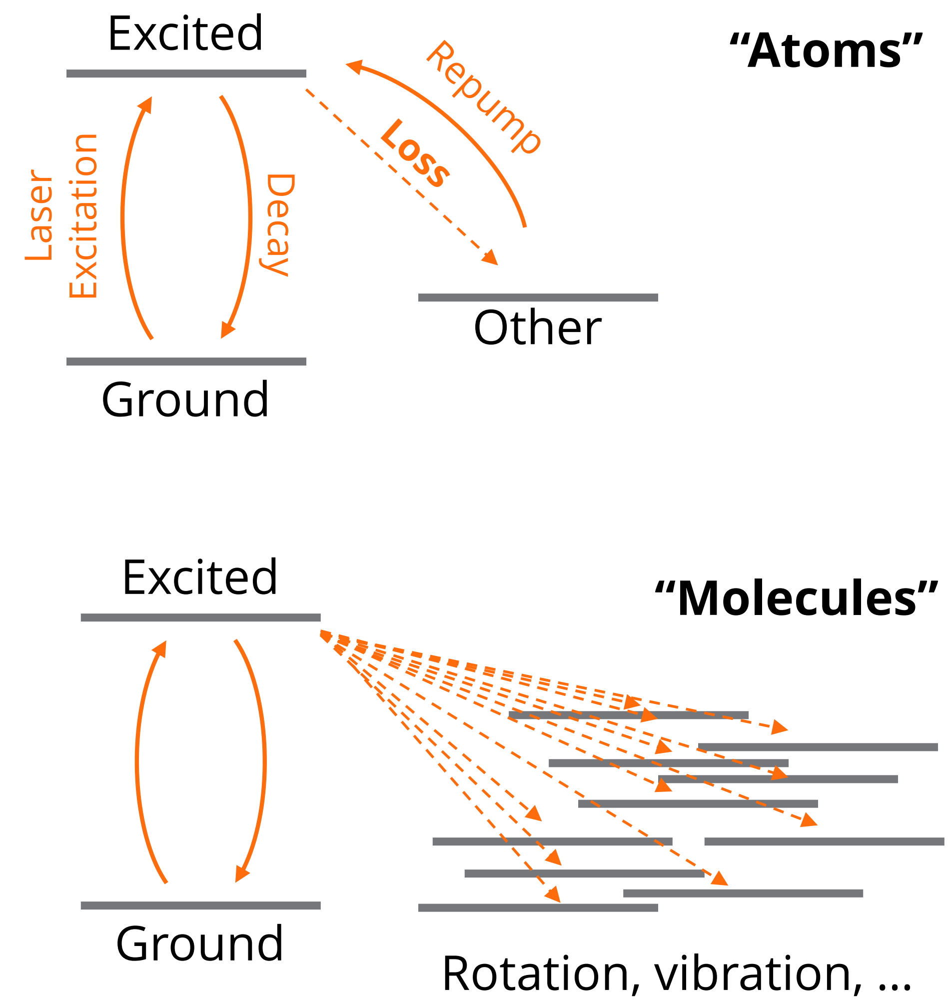
Laser cooling/trapping

- Lasers can be used to cool and trap < mK gases
- Important driver of many quantum techniques
  - Quantum information, quantum devices, synthetic quantum matter, fundamental chemistry, sensing, …
- Claim: only (proven) suitable method for us



Laser cooling molecules

- Requires many (~$10^{4\text{-}5}$) cycles of absorption, spontaneous decay
- Decay to other states stops the cooling process
- Internal vibrational, rotational levels are excited in decay



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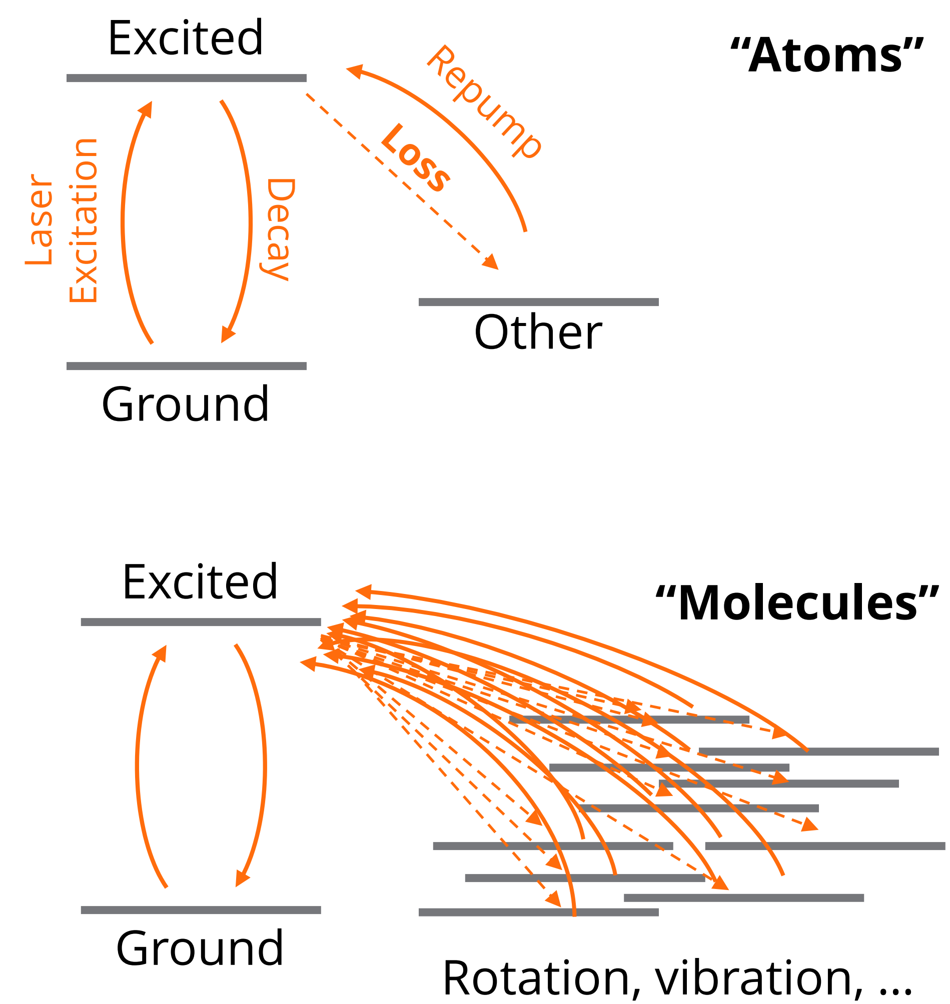

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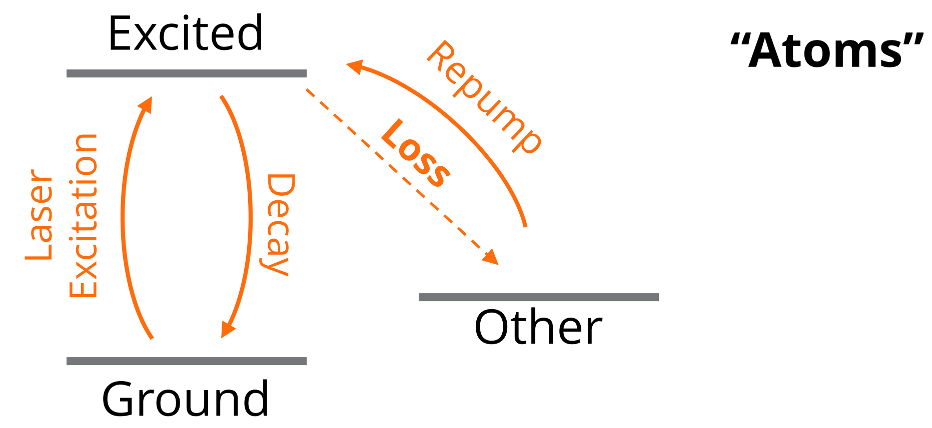
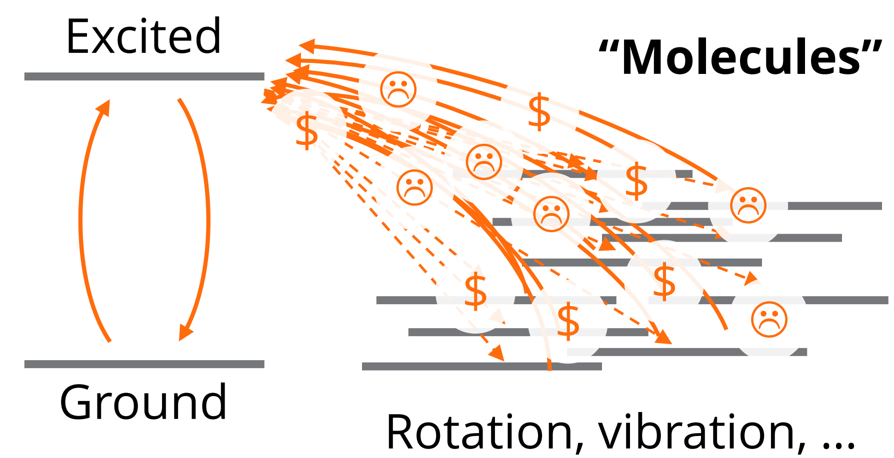
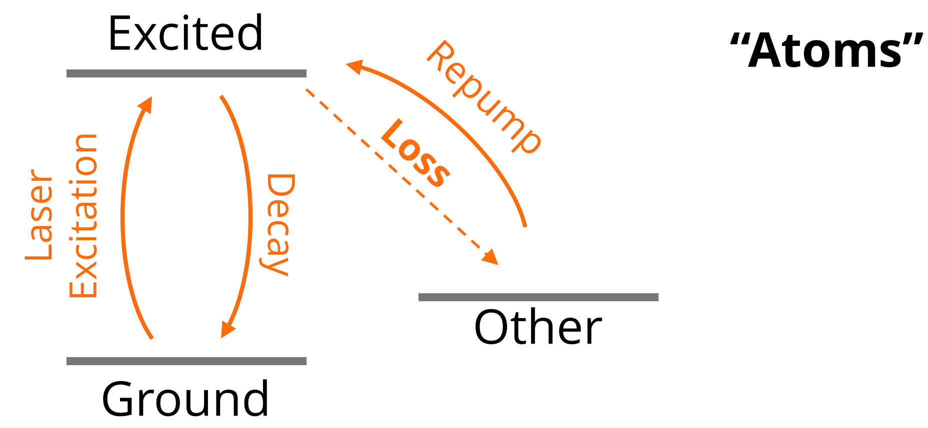
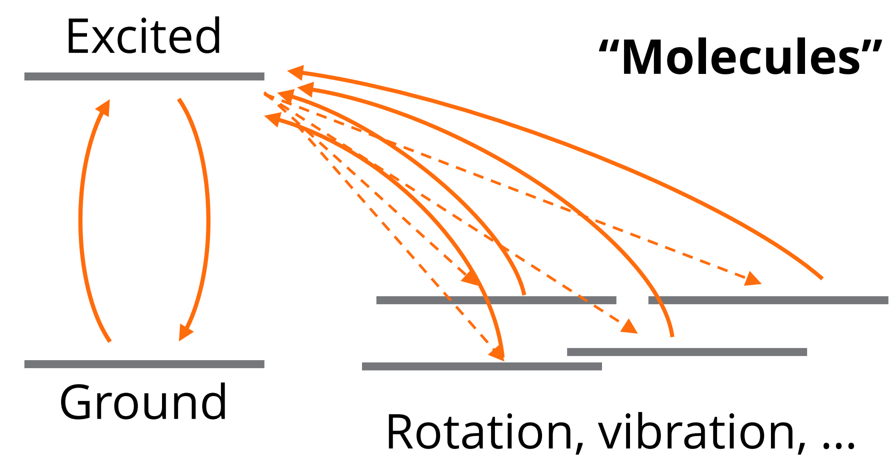




Laser cooling molecules

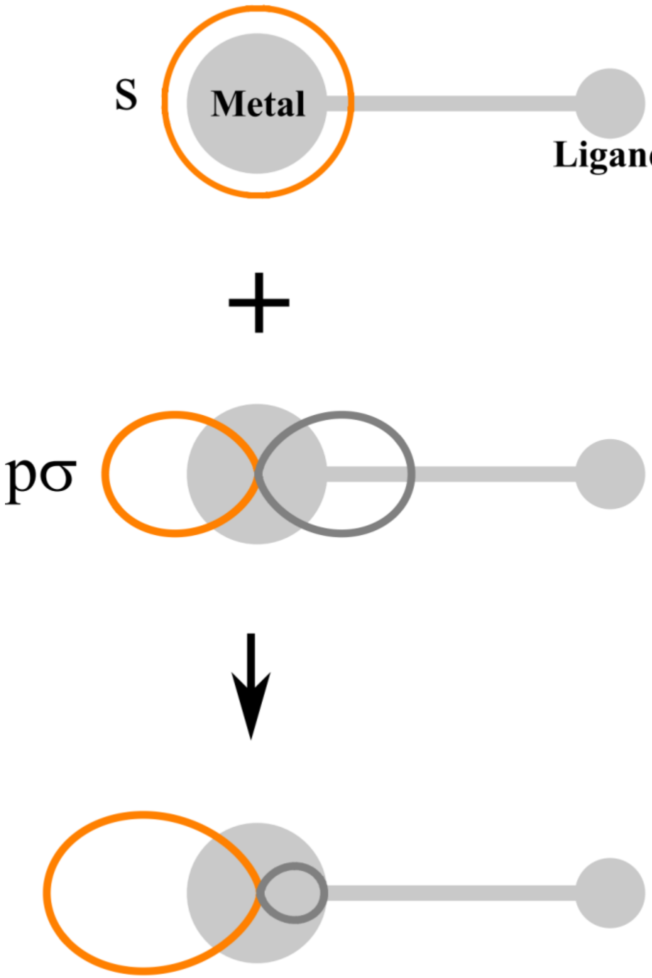
- Requires many ($\sim10^{4\text{-}5}$) cycles of absorption, spontaneous decay
- Decay to other states stops the cooling process
- Internal vibrational, rotational levels are excited in decay
- For carefully chosen molecules, this is manageable!



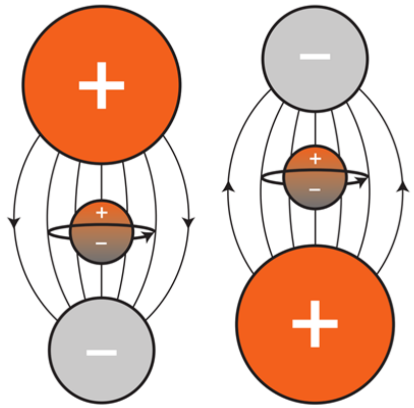


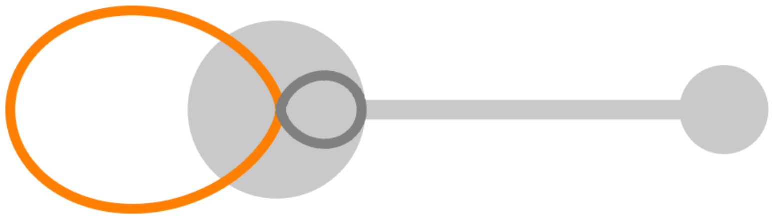
Electronic Structure for Laser Cooling

- Generally works for molecules with single, metal-centered s orbital
  - Alkaline-earth (s^2)
  - Single bond to halogen (F)
- Orbital hybridization pushes electron away from chemical bond
  - Decouples electronic structure from molecular excitations



Incompatible Features





Internal Comagnetometers

- ThO, WC, TaN, HfF^+, ...
- "Requires" $L_{elec} > 0$
- **Interferes with laser cooling**

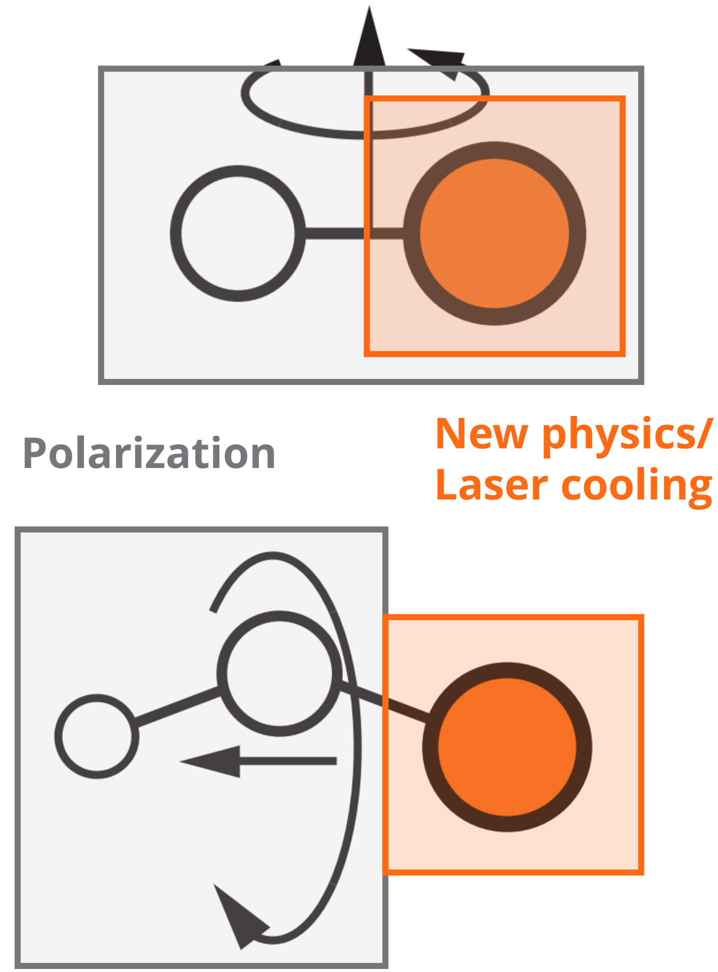
Only in diatomics!

Laser Cooling

- YbF, BaF, RaF, TlF, ...
- "Requires" $L_{elec} = 0$
- **No internal comagnetometers**

Polyatomic molecules

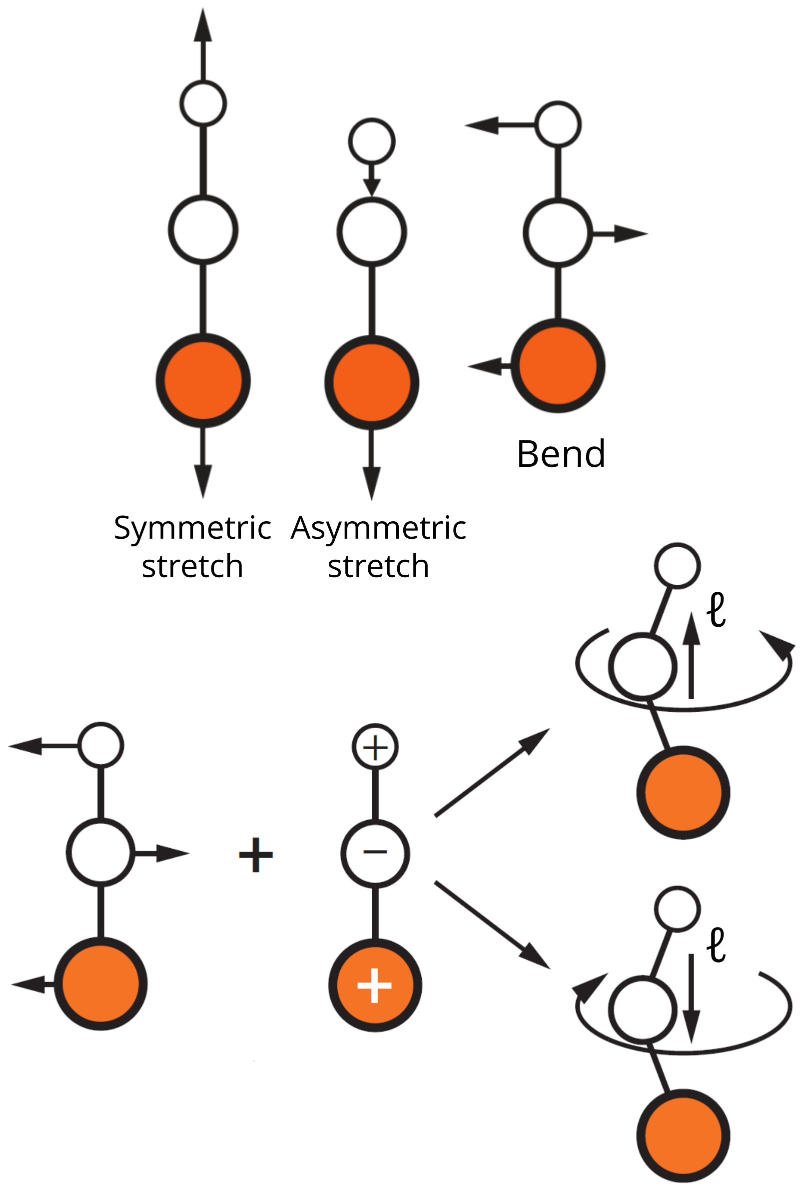
- In *diatomics*, full polarization and laser cooling conflict
- In *polyatomics*, these features can be decoupled
  - Laser cooling, new physics sensitivity via metal
  - Polarization and co-magnetometers from ligand (with >1 atom)



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

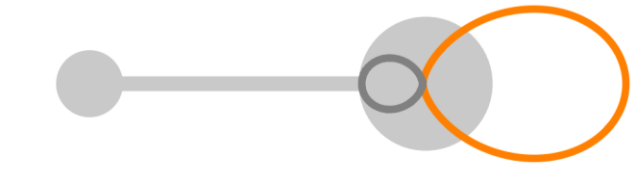
ℓ-doublets

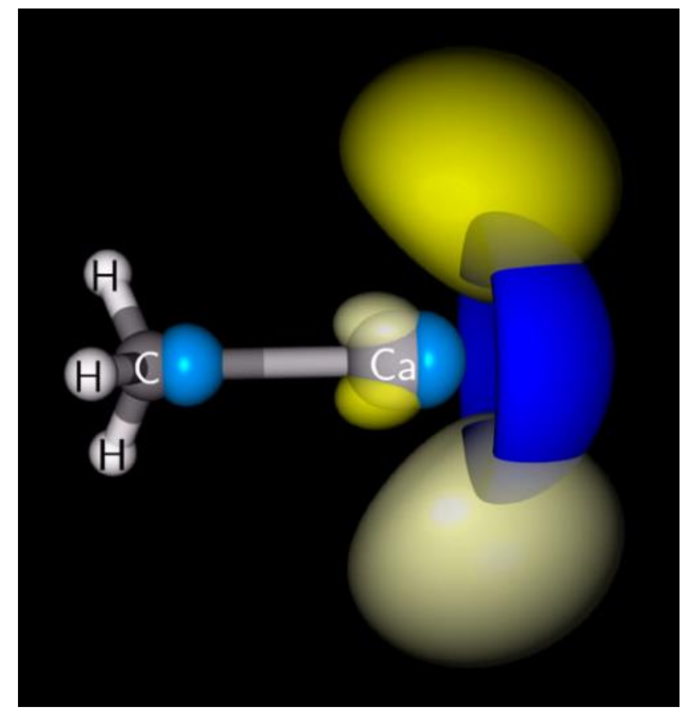
- Example: linear triatomic
- Three mechanical modes
- Bending mode is doubly degenerate
- Eigenstates have orbital angular momentum ℓ
- Coupling of ℓ to rotation gives rise to parity doublet!
  - Typical ~ 10 MHz
  - Independent of electronic structure!
- Symmetric tops even better? (K-doubling)



Laser cooling polyatomics

- Laser cooling still works!
  - Electron wavefunction *really is* decoupled from bond
  - Still looks like single, metal-centered electron
  - Essentially any monovalent, ionic bond
- Recently demonstrated
  - SrOH – I. Kozyryev, *et al.*, PRL **118**, 173201 (2017)
- Readily created
- Lots of spectroscopy





T. A. Isaev and R. Berger, PRL **116**, 63006 (2016)

Pathway to PeV Physics

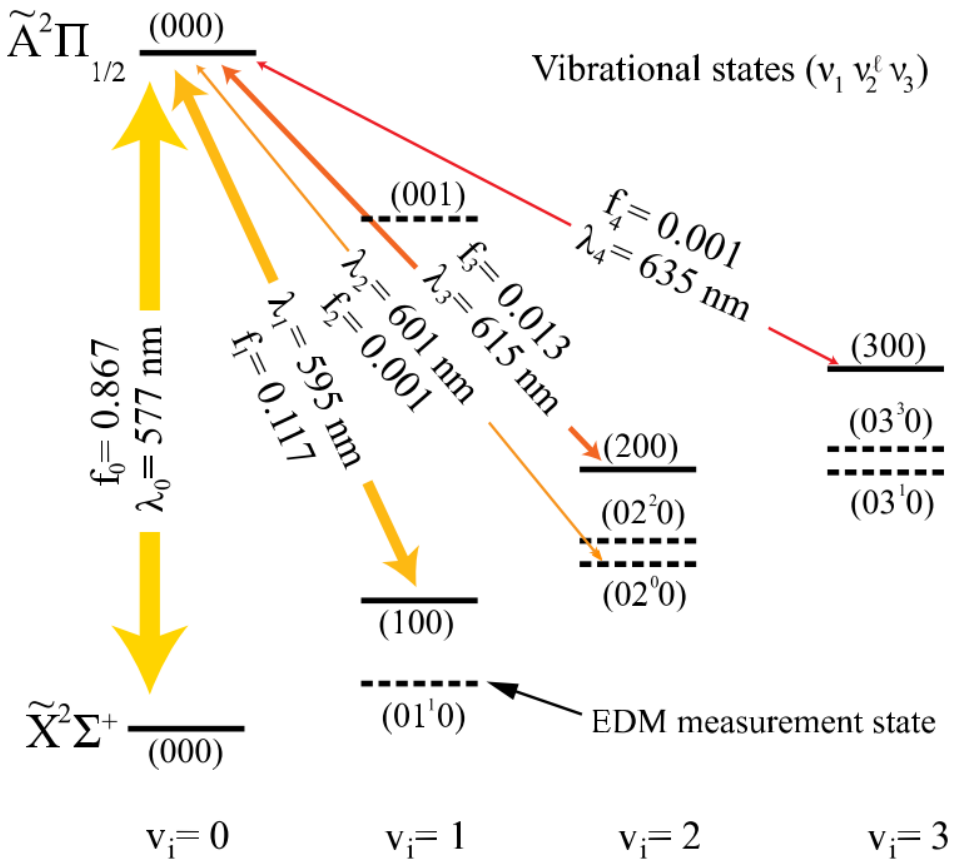
- Realistic pathway to PeV-scale physics
- General approach, applicable for many measurements
  - Electron EDM:
    YbF → YbOH, BaF → BaOH, ...
  - Nuclear Schiff:
    TlF → TlOH, RaF → RaOH, ...
  - Nuclear Anapole:
    BaF → BaOH, ...
  - Ion trap searches:
    $RaOH^+$, $ThOH^+$, ...
- Lots of new directions!

Feature	**Polyatomics**
Laser cooling	✓
Full polarization	✓
Internal co-mag.	✓
>1 s lifetime	✓
Scalable (Large #)	✓

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

YbOH

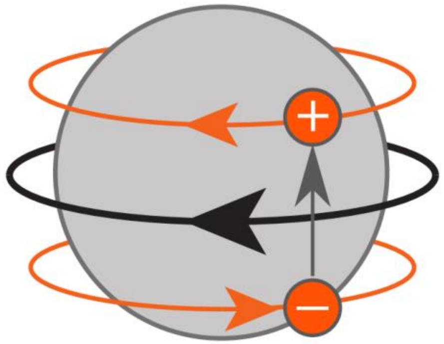
- YbOH is ideal candidate for first experiment
  - Existing preliminary spectroscopy
  - Recent additional spectroscopy
  - Good CPV sensitivity
  - Laser coolable
  - Multiple stable isotopes
- Sensitive to hadronic and leptonic CPV
  - … and more!



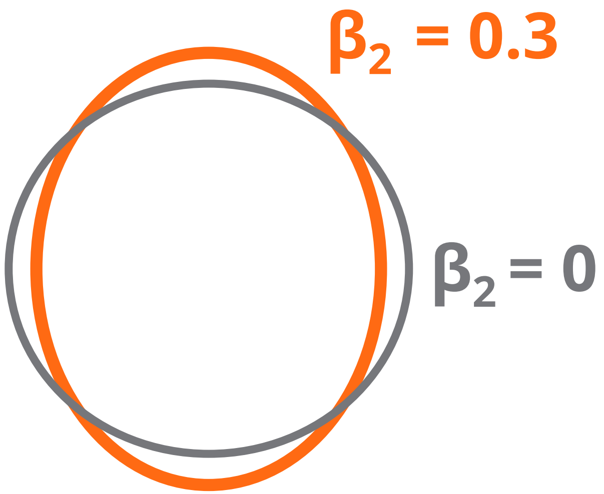
I. Kozyryev and NRH, Phys. Rev. Lett. **119**, 133002 (2017)

Nuclear MQM

- Nuclear magnetic quadrupole moments are sensitive to hadronic CPV
  - Nucleon EDM
  - quark EDM/chromo-EDM
  - CPV nuclear forces
  - Strong CPV (θ_{QCD})
  - …
- Orthogonal to eEDM
  - Current eEDM molecules are not sensitive
- Quadrupole deformation (β_2) enhances MQM
  - Collective enhancement
  - Ta, Yb, Hf, Lu, Th, …

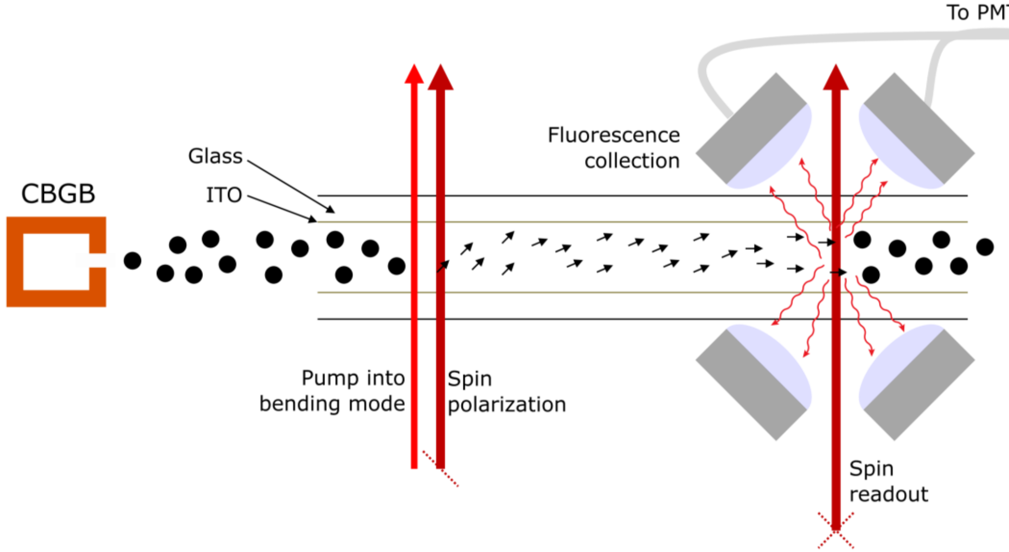


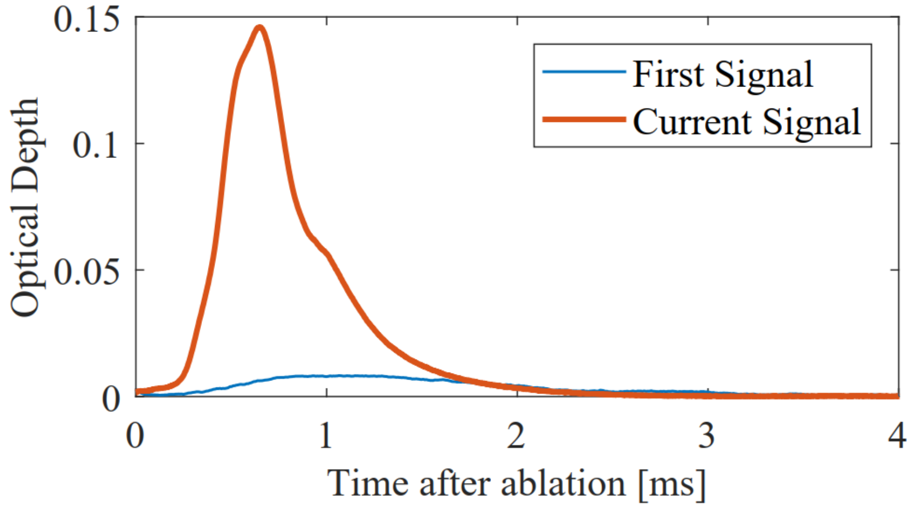
Rotating EDM produces MQM



^{173}YbOH NMQM Experiment @ Caltech

- Building a NMQM search in ^{173}YbOH at Caltech
  - ^{173}Yb (I=5/2), highly deformed
  - Cryogenic buffer gas beam experiment
  - Laser cooling, trapping in future generations?
- Construction underway!
  - We have a YbOH beam
  - Currently optimizing and characterizing



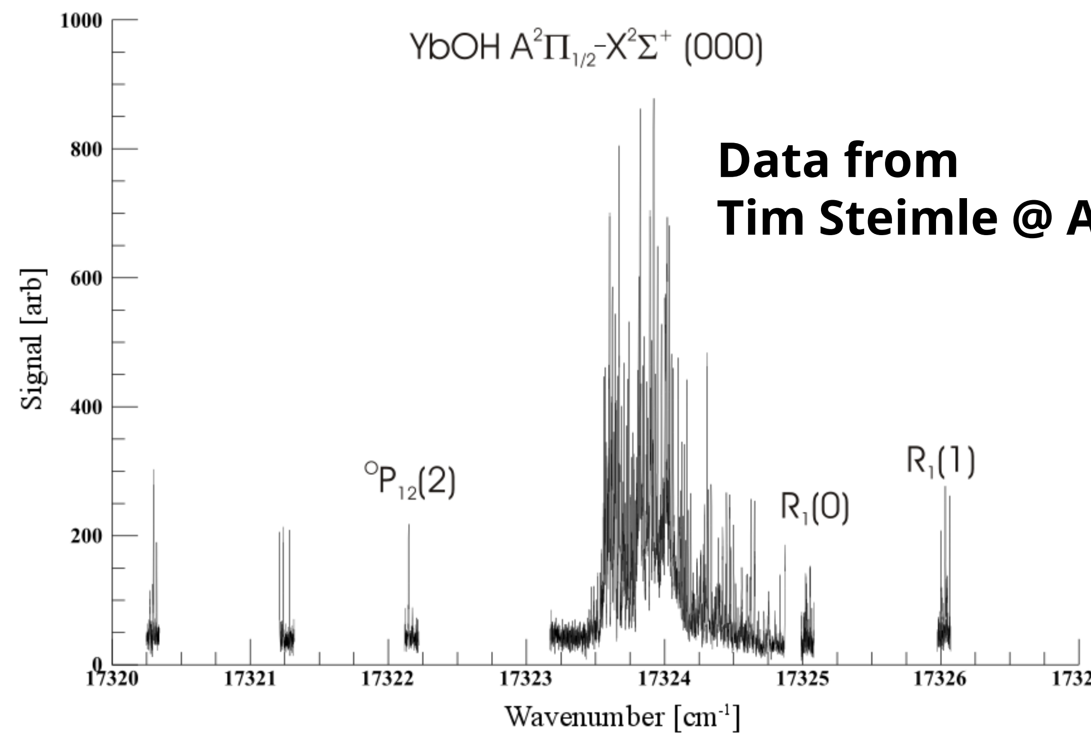


Polyatomic eEDM Experiment

- Electron EDM search in laser cooled and trapped ^{174}YbOH
- ^{174}Yb (I=0), simpler structure
- Just getting started!
  - NRH @ Caltech
  - John Doyle @ Harvard
  - Tim Steimle @ ASU
  - Amar Vutha @ Toronto
- Goal: explore PeV-scale fundamental symmetry violating physics
- *Stay tuned!*

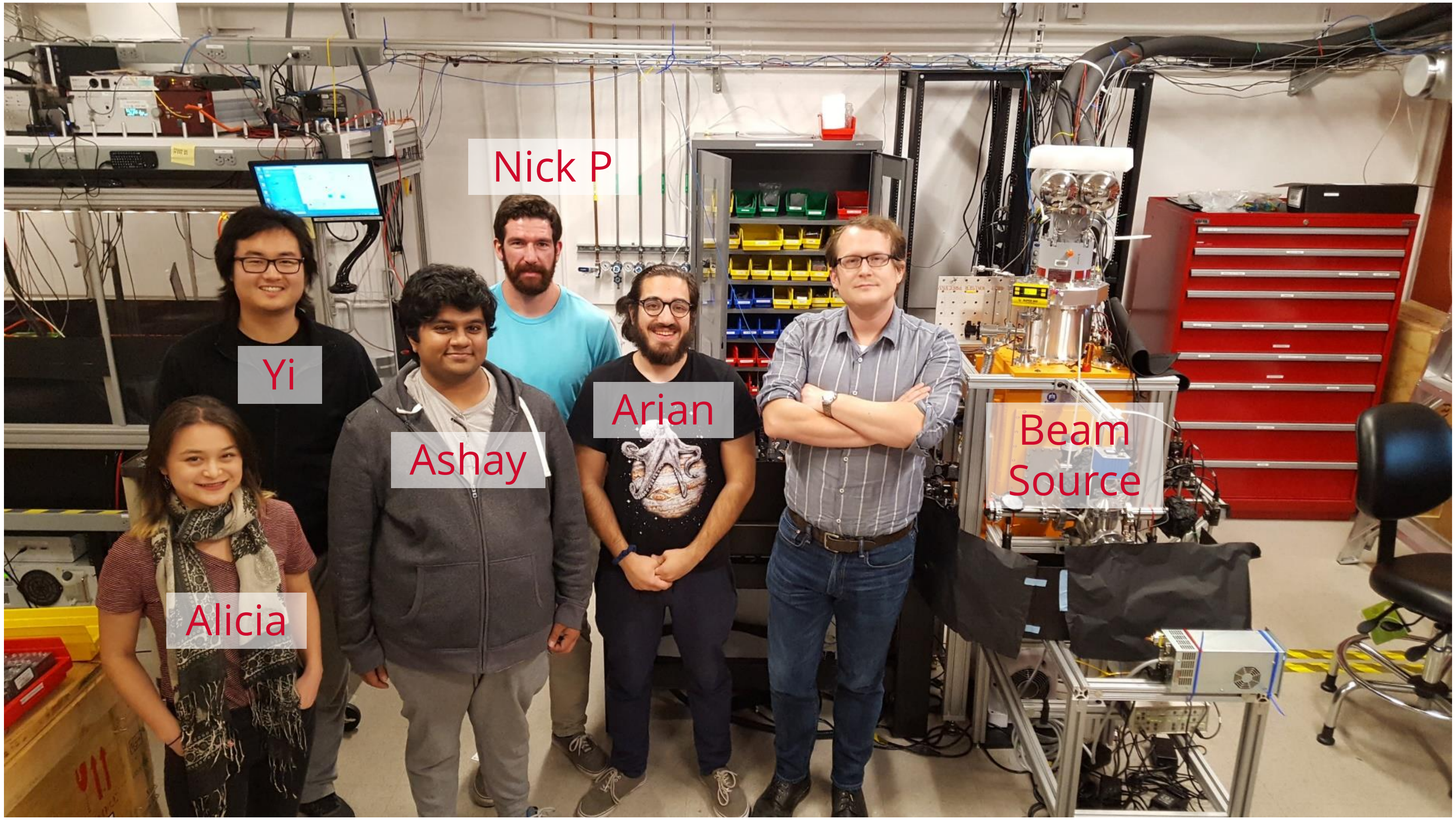
EDM

www.polyedm.com



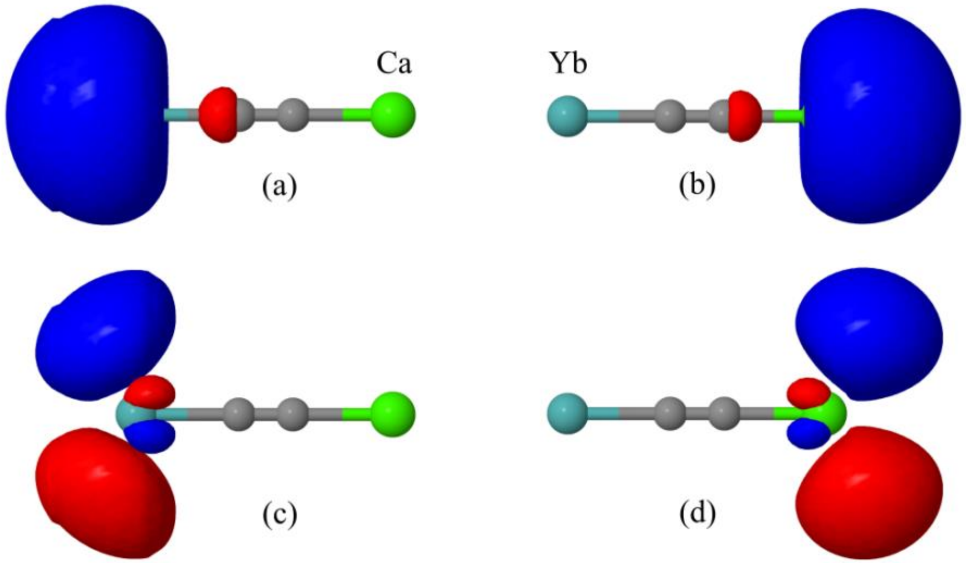
Data from Tim Steimle @ ASU

The Group, Fall 2018



A Future Direction: MFOCCs

- Why not make ligand optically active?
  - Enhanced optical forces
  - New state preparation/readout approaches
  - More co-magnetometry
  - Access to non-cycling species
  - …
  - ***New avenues for quantum control!***
- Molecules Functionalized with Optical Cycling Centers (MFOCCs)
  - New collaboration!
  - Wes Campbell, Anastassia Alexandrova, Justin Caram, John Doyle, Eric Hudson, NRH, Anna Krylov



Yb−C≡C−Ca

Behaves like two cycling centers!

Calculations by Matt O'Rourke

Thanks for your attention!

Come visit!

Questions?