Caltech

Precision Measurements with Laser Cooled Polyatomic Molecules

Nick Hutzler

Caltech

NIST

HEISING-SIMONS FOUNDATION

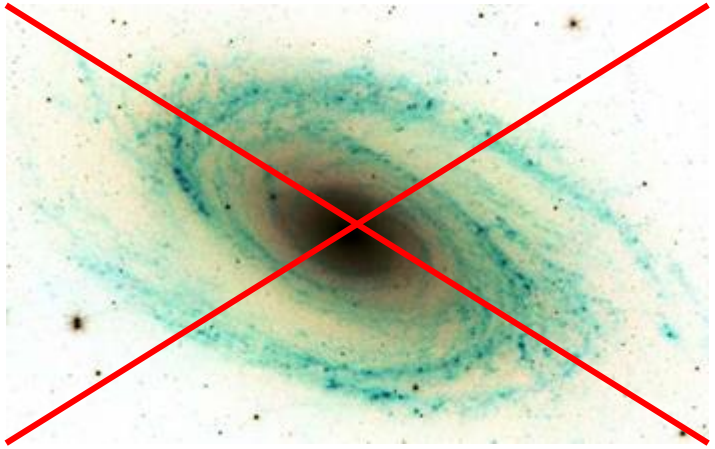
U.S. DEPARTMENT OF ENERGY

Office of Science

NSF

An Asymmetric Universe

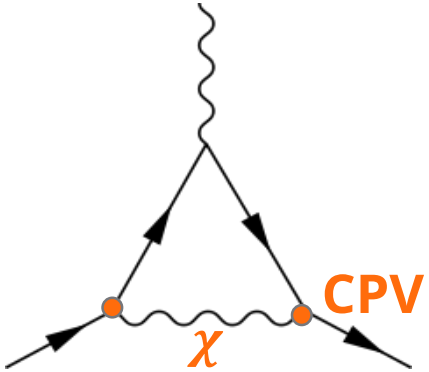
- The universe is made out of matter
- There is no free anti-matter in the universe
- One possible solution: Sakharov conditions, requires new sources of CP violation
- One of the motivations to search for CP violation beyond the Standard Model

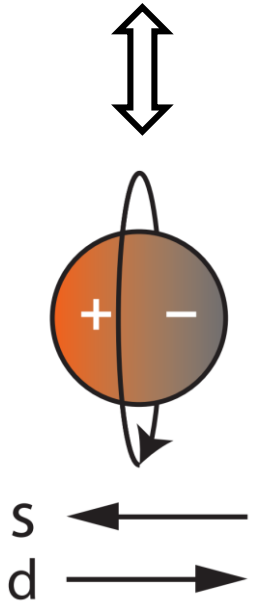


?

CP Violating Observables

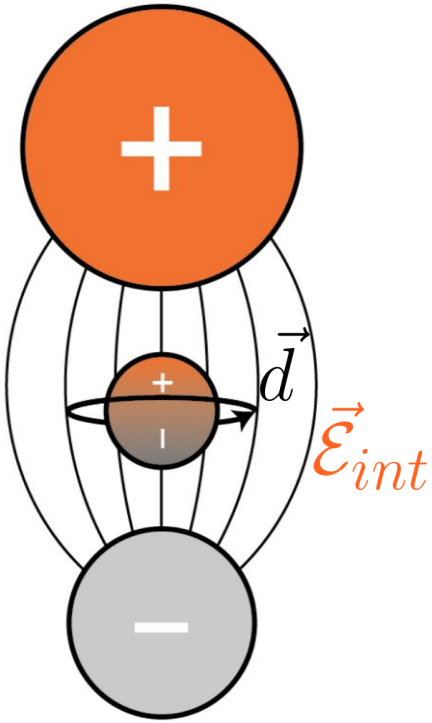
- New CPV physics creates CPV observables at **low energies**
  - (Also high energies)
- Classic example: permanent electric dipole moments (EDMs)
  - **The existence of EDMs requires CPV**
  - Other electromagnetic moments as well

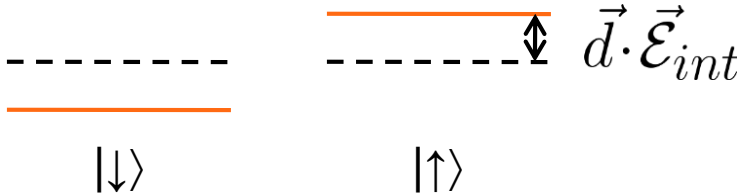




Molecular Internal Fields

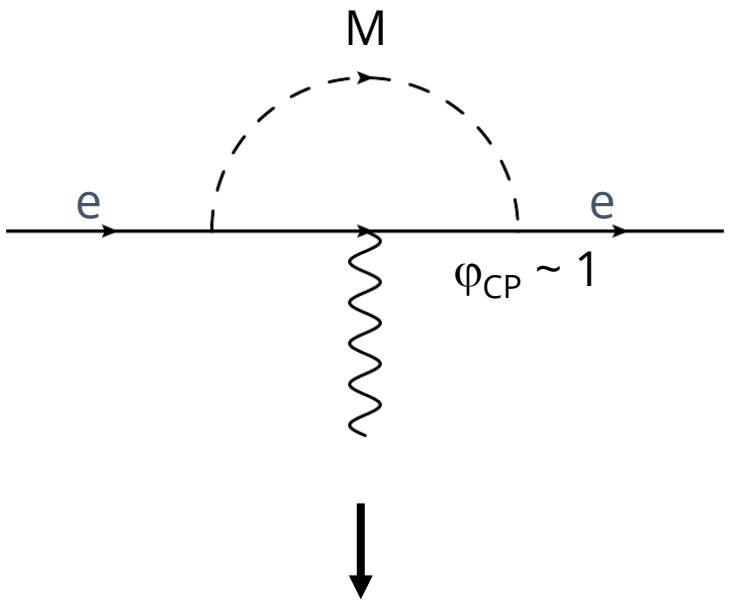
- Molecules have **extremely large** fields
  - Amplifies signatures of CPV electromagnetic moments
  - 10-100 GV/cm for heavy (~$Z^{2\text{-}3}$)
  - >1,000x compared to atoms
  - Maximum lab field ~100 kV/cm
- CPV moments cause CPV energy shifts
  - Example: eEDM
  - $H = -\vec{d} \cdot \vec{\mathcal{E}}_{int}$
- Study effect of internal fields on molecular constituents





Sensitivity

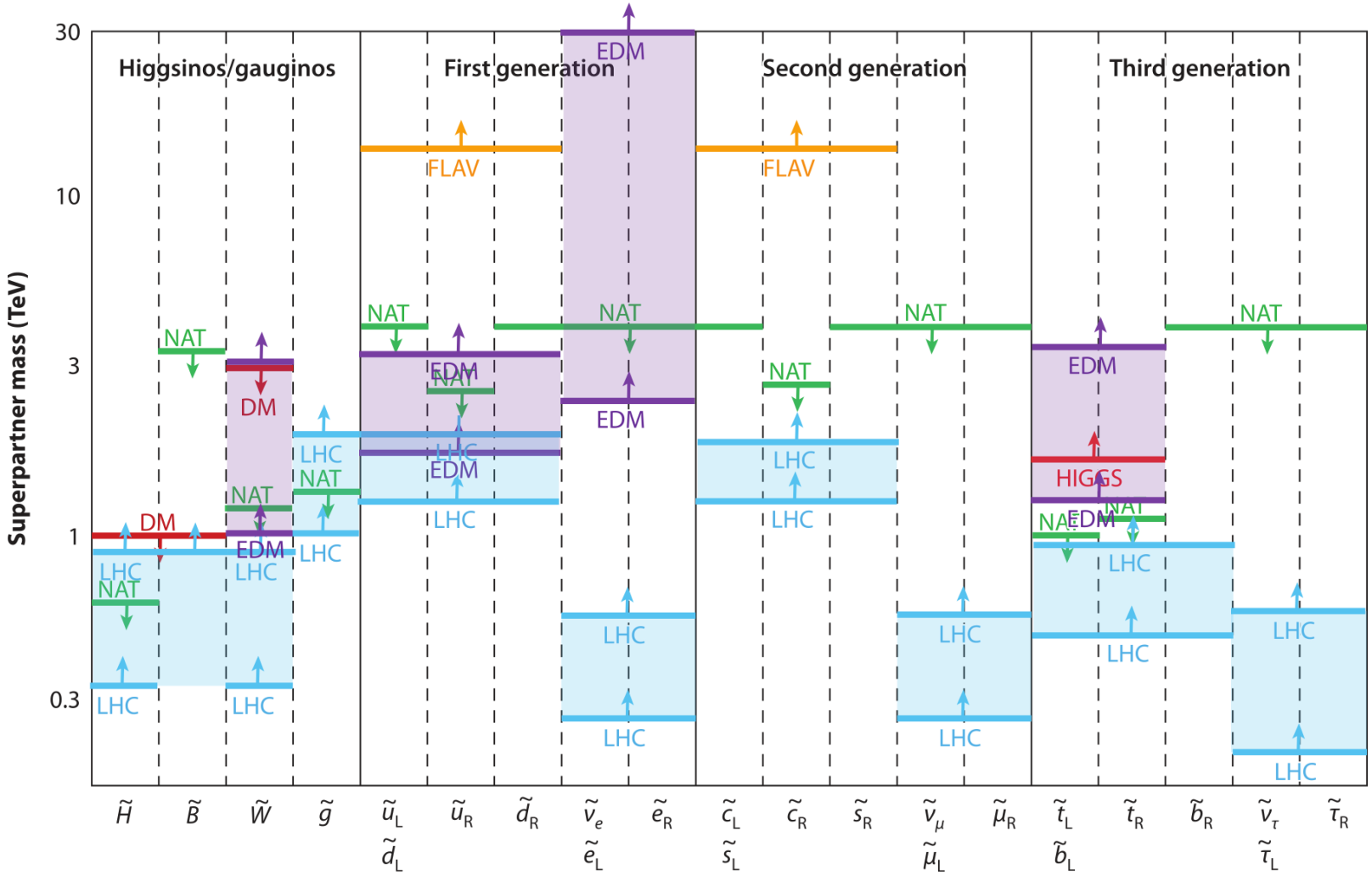
- eEDM sensitivity has improved by 100x in 10 years due to advances with molecules
  - ACME II: $|d_e| < 1.1 \times 10^{-29}$ e cm
- Broad sensitivity to new physics at high energies
  - Background free for many orders of magnitude
- *Complements the many exciting other approaches at this workshop!*
- **Significant further advances in near future from improved quantum control**



M < ~30 TeV

ACME Collaboration. Nature 562, 355 (2019)

Many approaches



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

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

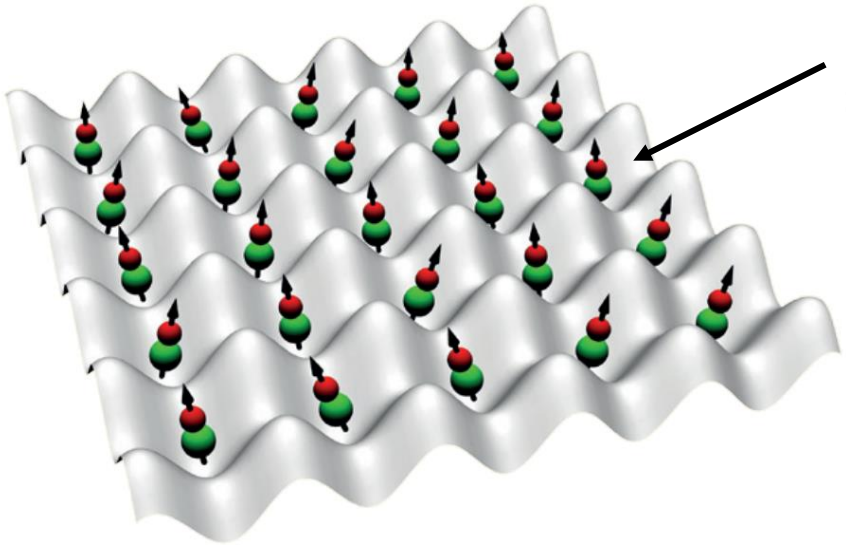
Adapted and updated from J. Feng, Ann. Rev. Nuc. Part. Sci. 63, 351 (2019) with help from D. DeMille

Where we are going…

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

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

Even before implementing advanced quantum control, such as entanglement-based squeezing



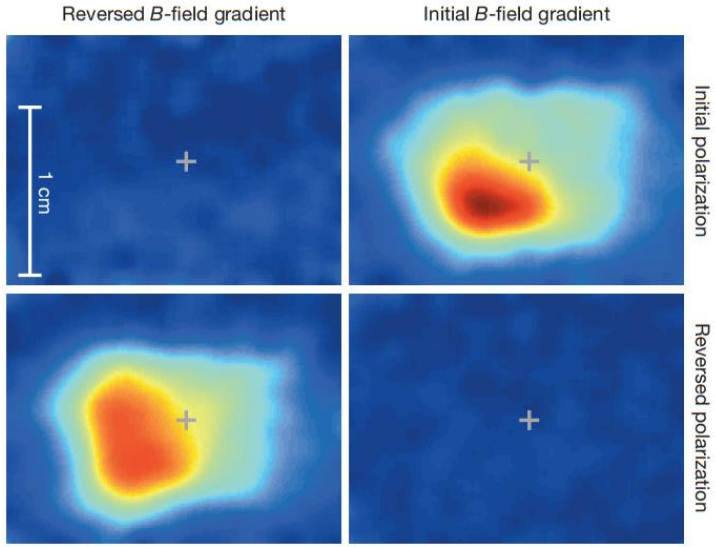
Heavy, polar molecule sensitive to new physics

Need to trap at ultracold temperatures

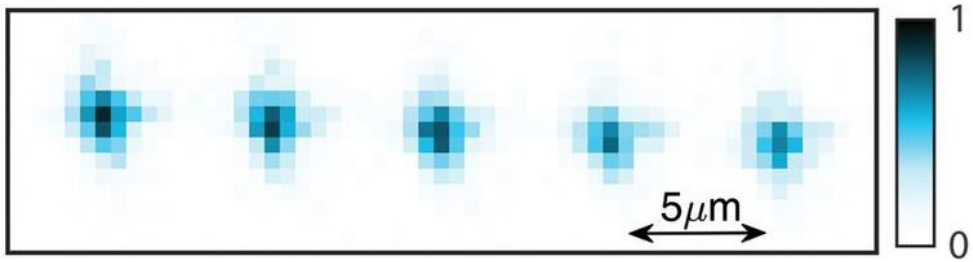
Figure adapted from A. J. Daley, Nature **501**, 497 (2013)

Laser cooling molecules

- Laser cooling is critical to quantum science with atoms and molecules
- Challenging in molecules due to their complex structure
  - Photons can “accidentally” excite internal modes, leading to loss
- Recent, rapid advances!
  - Since 2009: Optical forces, laser cooling, magneto-optical trapping, sub-Doppler cooling (multiple), magnetic trapping, optical trapping, single molecule control, …
- Requires particular electronic structure
  - SrF (DeMille @ Yale), CaF (Tarbutt @ Imperial, Doyle @ Harvard), YO (Ye @ JILA) cooled and trapped… so far



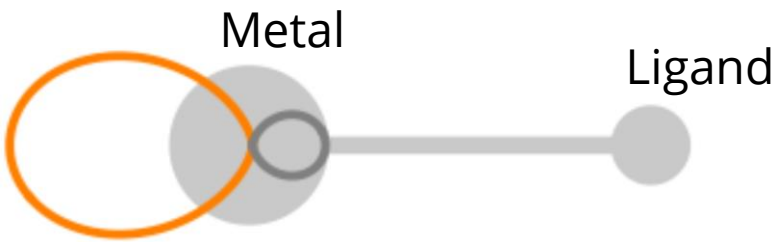
J. F. Barry *et al.*, Nature 512, 286 (2014)



L. Anderegg *et al.*, Science 365, 1156 (2019)

Laser Coolable Molecules

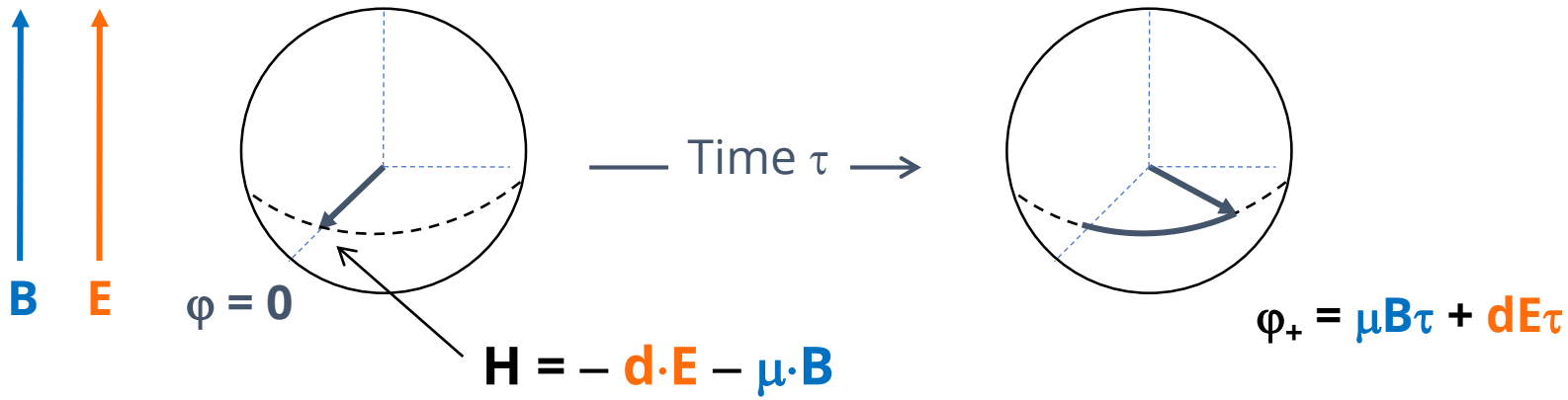
- Recipe: single, metal-centered s electron*
  - $\bullet Sr \bullet \ \circ F \longrightarrow \bullet Sr - F$
  - Decouples electronic from vibrational excitation
- This electronic structure gives good CPV sensitivity
  - Need heavy species (~$Z^{2\text{-}3}$)
  - Several laser-coolable options
- Polyatomic analogues should also work!
  - $\bullet Sr \bullet \ \circ OH \longrightarrow \bullet Sr - OH$
- **Why polyatomics?**



Laser coolable molecules with sensitivity to fundamental symmetry violations	
BaF	EJPD **72**, 197 (2018)
RaF	PRA **82**, 52521 (2010)
TlF	PRA **95**, 62506 (2017)
YbF	PRL **120**, 123201 (2018)
	…

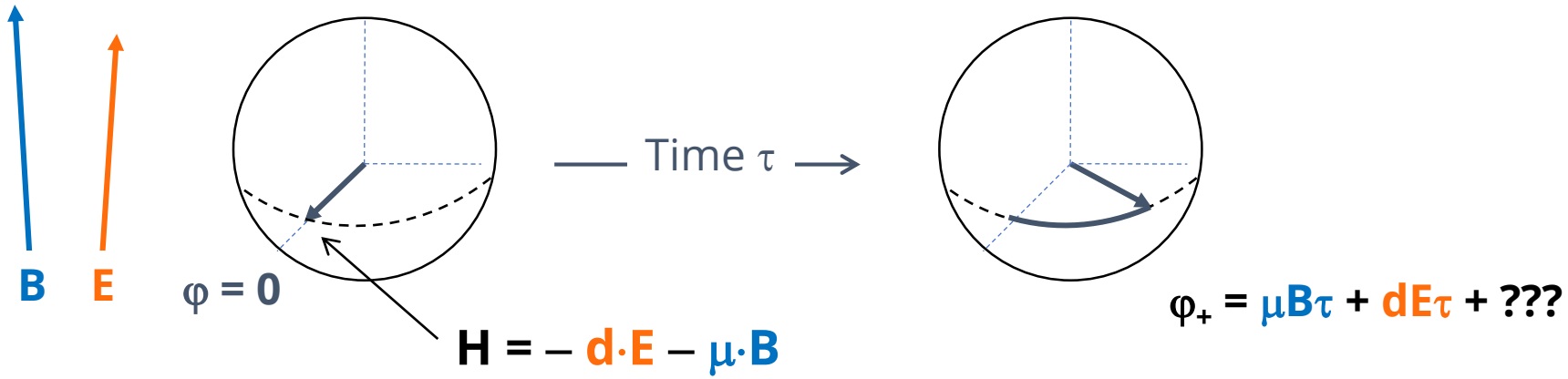
**Actually, neither necessary nor sufficient… results from complex bond hybridization*

Spin precession



$$\Delta\varphi \propto dE\tau$$

Nothing is perfect...

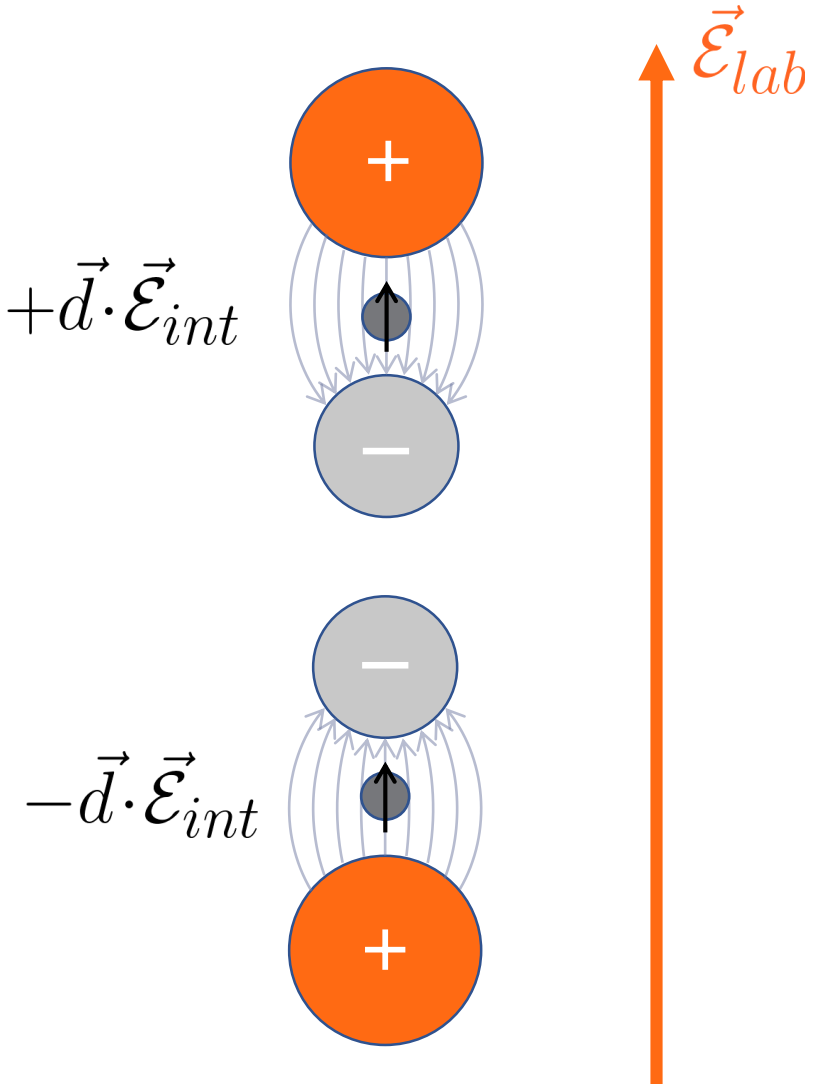


$\mathbf{dE / \mu B < 10^{-6}}$

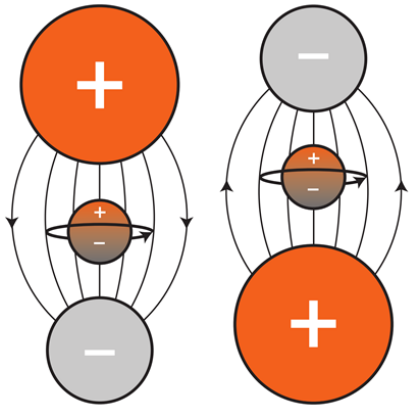
$\Delta\varphi \propto \mathbf{dE\tau + ???}$

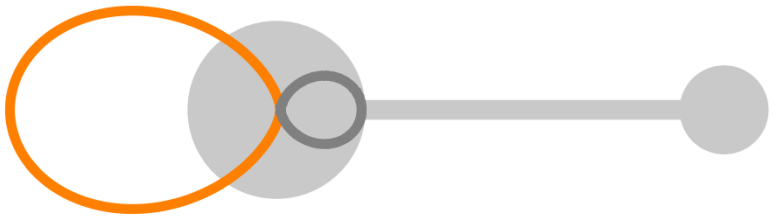
Internal Comagnetometers

- Some molecules can be *fully* polarized in the lab
  - "Internal co-magnetometer"
- Non-CPV effects cancel
  - No external field change + small fields → **Extreme suppression of motional fields, geometric phases, leakage currents, …**
- Best eEDM limits (ACME, JILA) already depend on this
- Requires "exotic" electronic structure in diatomic molecules
  - Parity doubling – internal states split into opposite parity doublets



Incompatible Features





Internal Comagnetometers

- ThO, WC, TaN, HfF^+, …
- Complex electronic structure
- **Interferes with laser cooling**

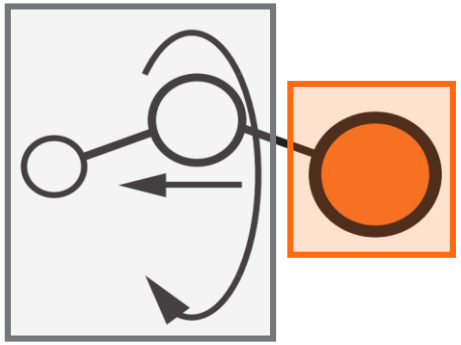
Only in diatomics!

Laser Cooling

- YbF, BaF, RaF, TlF, …
- Simple electronic structure
- **No internal comagnetometers**

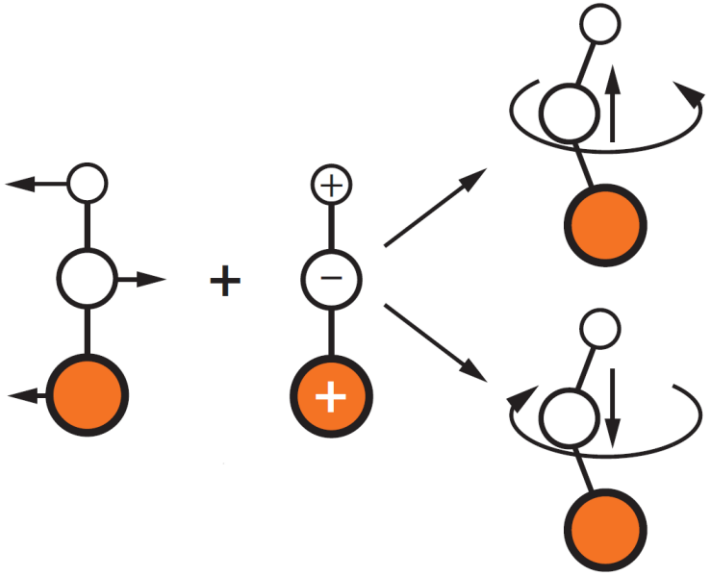
Polyatomic molecules

- Polyatomics generically have parity doublets
  - From mechanical modes
  - Polarize in <1 kV/cm
  - Internal co-magnetometers
  - *Does not destroy laser cooling (unlike diatomics)*
- Example: bending mode in linear triatomic is (nearly) doubly degenerate
- **Polyatomic molecules can be laser cooled, fully polarized, and have internal co-magnetometers**



New physics
Laser cooling

Polarization, Co-magnetometers

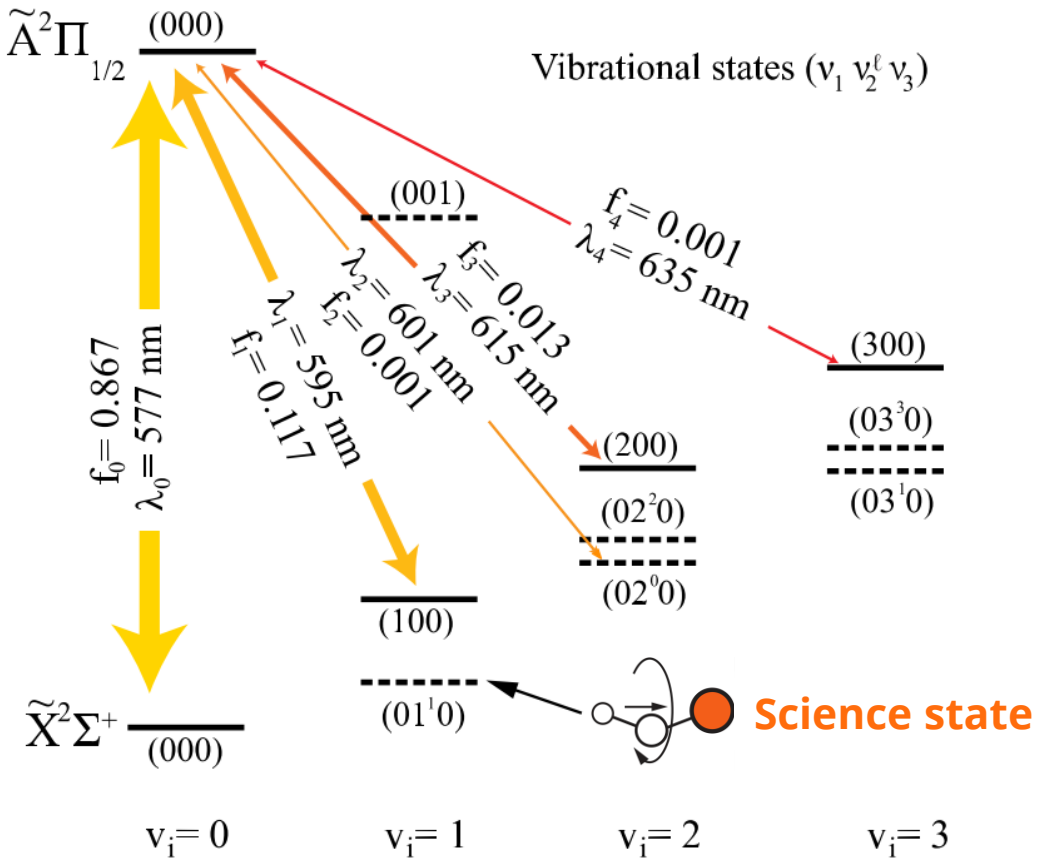


I. Kozyryev and NRH, PRL 119, 133002 (2017)

YbOH

- YbOH is an ideal candidate
  - Imperial group has proven new physics sensitivity, laser cooling in YbF
- Recent calculations of YbOH E_{eff} by several groups
  - E_{eff} = 23.4 GV/cm

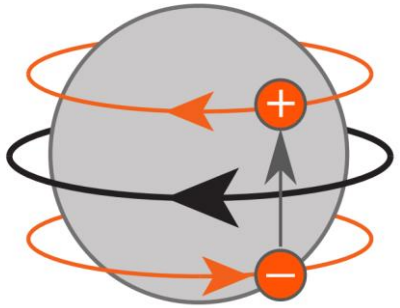
    M. Denis, P. A. B. Haase, R. G. E. Timmermans, E. Eliav, NRH, and A. Borschevsky, Phys. Rev. A **99**, 042512 (2019)
- Broad sensitivity to via multiple stable isotopes
  - **eEDM, NMQM**, NSM, AM, …
- Lots of exciting future directions with other species
  - Ra, Ba, Tl, Ac, Pa, Lu, … ?



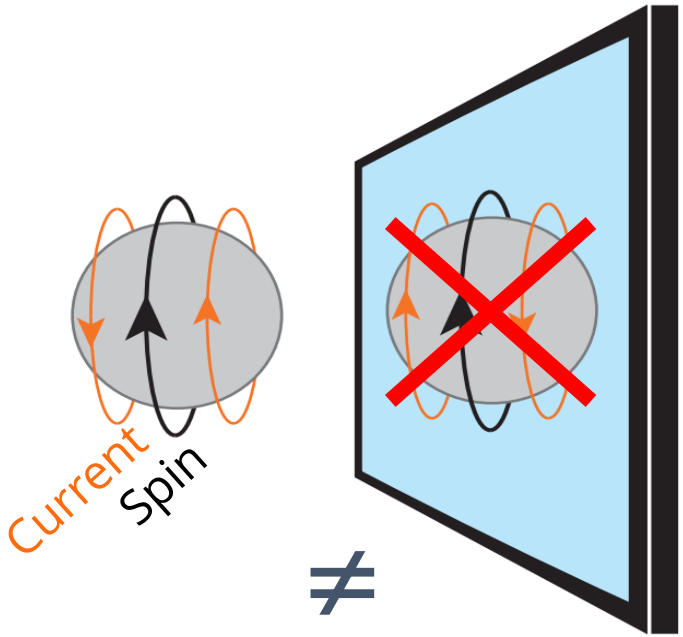
I. Kozyryev and NRH, Phys. Rev. Lett. 119, 133002 (2017)

Nuclear Magnetic Quadrupole Moments

- NMQM Arises from hadronic sources
  - Nucleon EDM
  - quark EDM/chromo-EDM
  - CPV nuclear forces
  - Strong CPV (θ_{QCD})
  - …
- Interacts with internal molecular field gradients
- Quadrupole deformation (β_2) enhances MQM
  - Collective enhancement
  - Typically $\beta_2 Z \sim 10$
  - **Yb**, Ta, Hf, Lu, Ra, Th, …

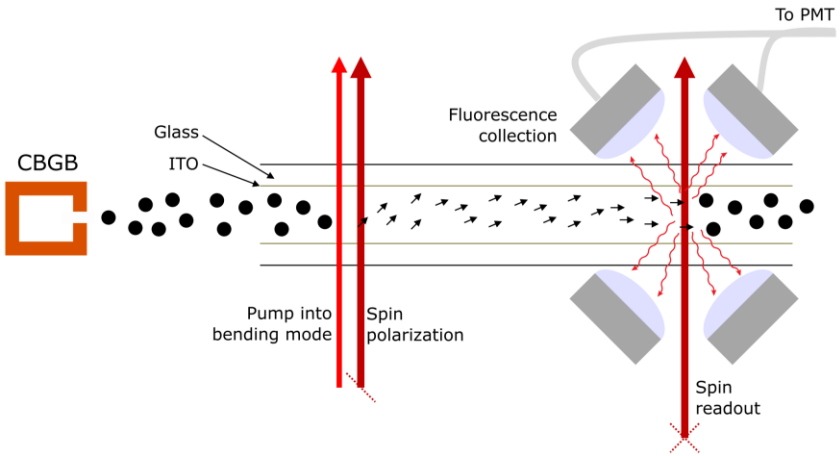


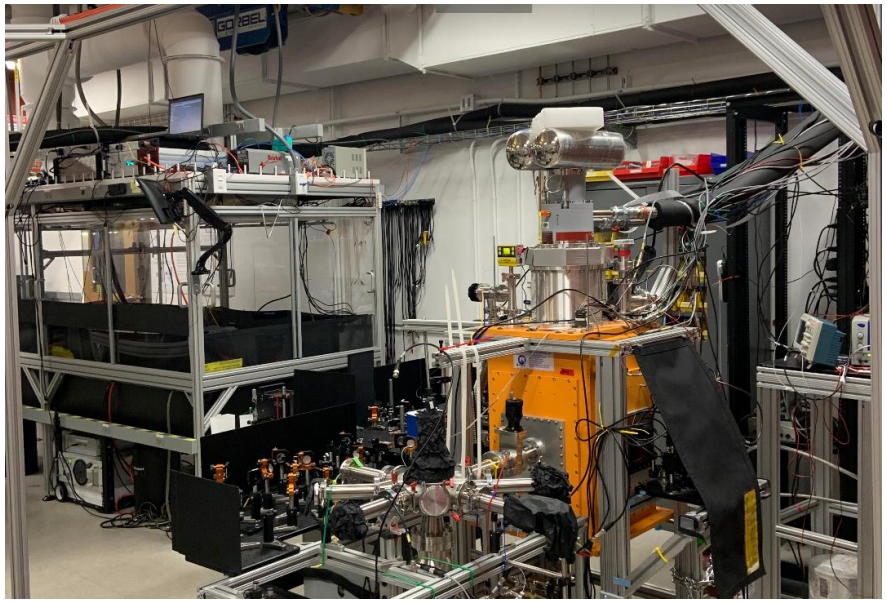
Rotating EDM produces MQM



^{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?
- Under way!



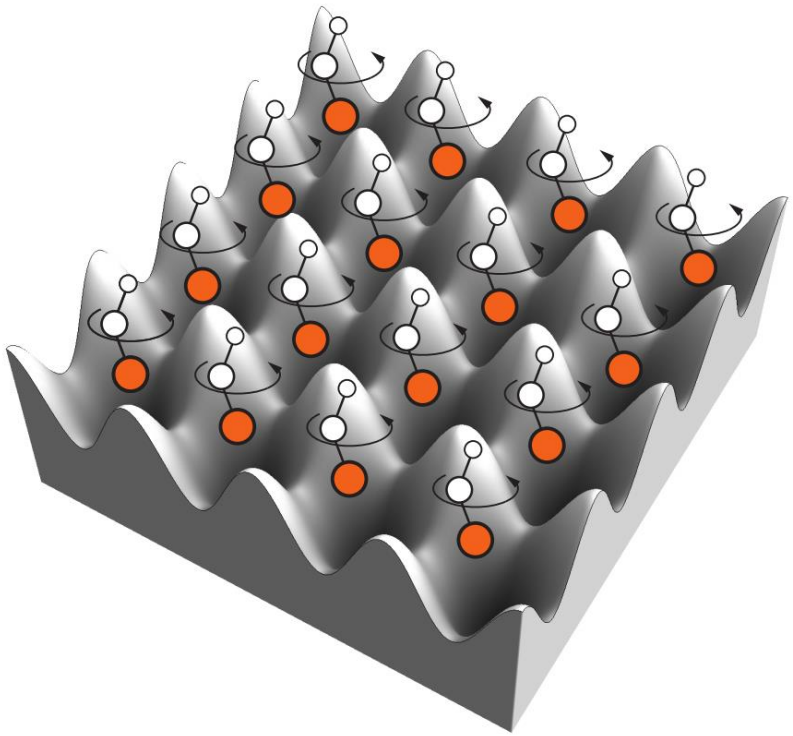


I. Kozyryev and NRH, PRL 119, 133002 (2017)
D. E. Maison, L. V. Skripnikov, and V. V. Flambaum, PRA 100, 032514 (2019)

Polyatomic eEDM Experiment

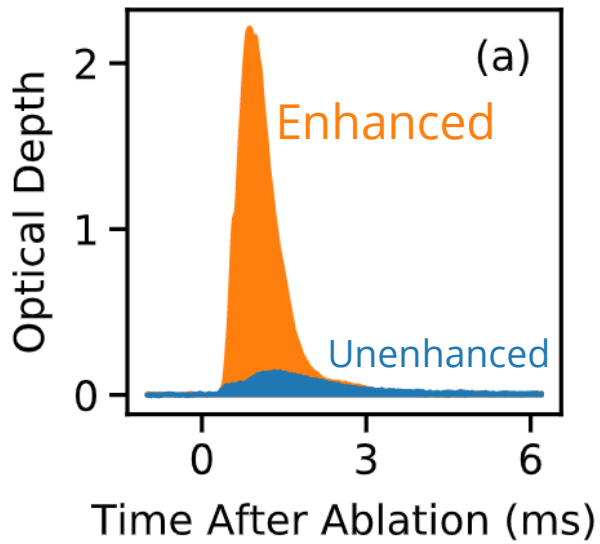
- Electron EDM search in laser cooled and trapped polyatomic molecules
  - Me @ Caltech
  - John Doyle @ Harvard
  - Tim Steimle @ ASU
  - Amar Vutha @ Toronto
- Goal – explore PeV-scale physics in a system with quantum control
- *www.polyedm.com*

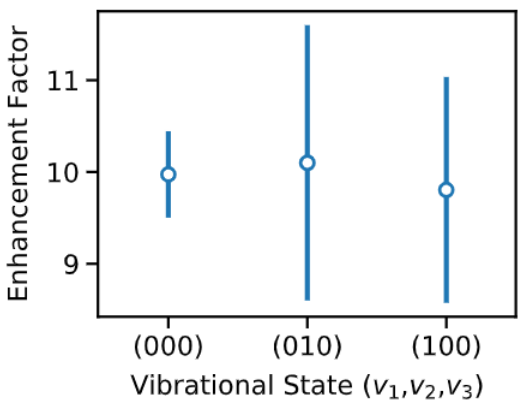
PEDM

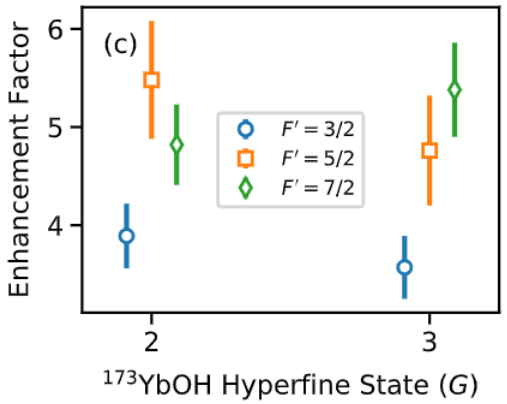


Creating (More) Molecules

- Demonstrated enhanced chemical production of YbOH in buffer gas beam
  - Ablate Yb+Yb$(OH)_3$
  - Excite Yb $^1S_0 \rightarrow {}^3P_1$
  - Yb($^3P_1$) reacts with ablation products to create YbOH
- ~10x enhancement in beam signal
  - Typically ~$10^{10\text{-}11}$ out of cell
  - More in future? Plenty of unreacted Yb…
- Populates science state!



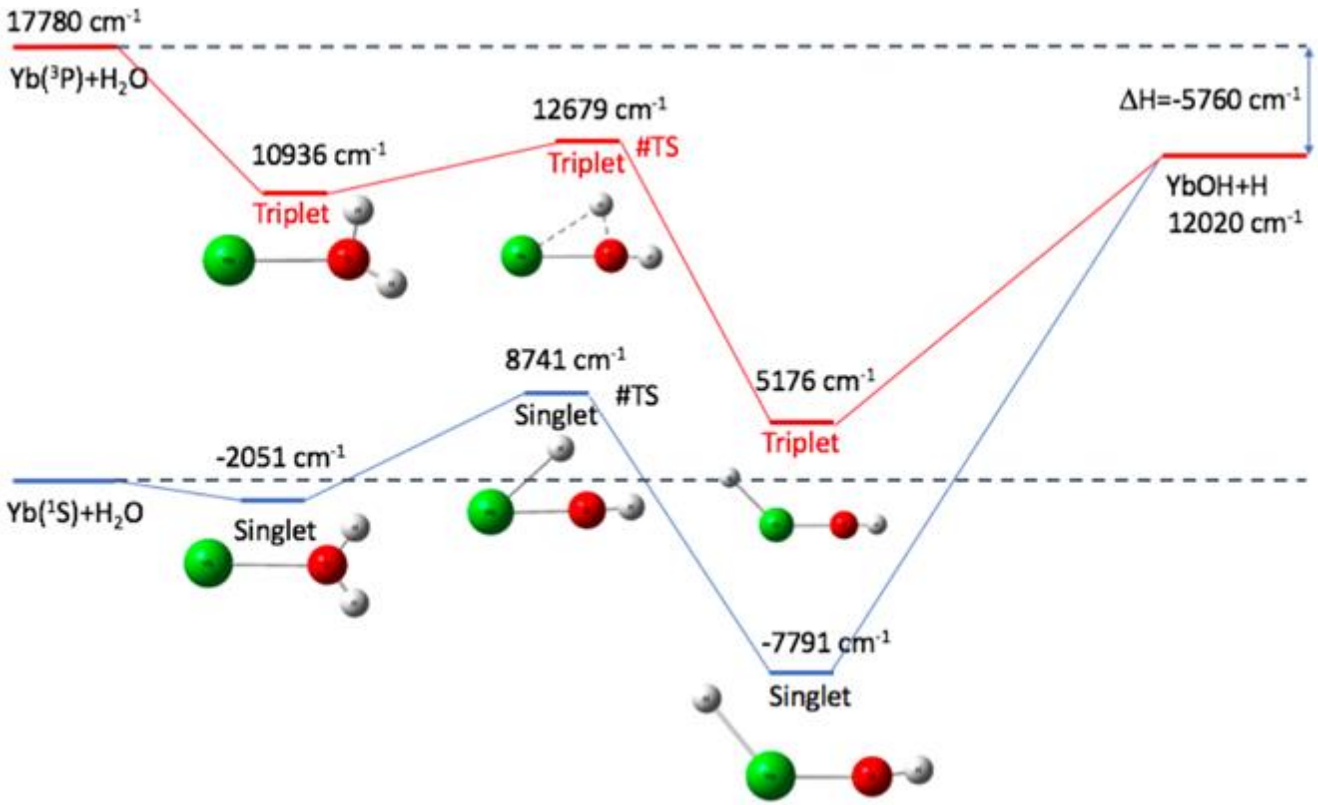




A. Jadbabaie, N. Pilgram, J. Kłos, S. Kotochigova, NRH, in preparation

YbOH Reaction Pathways

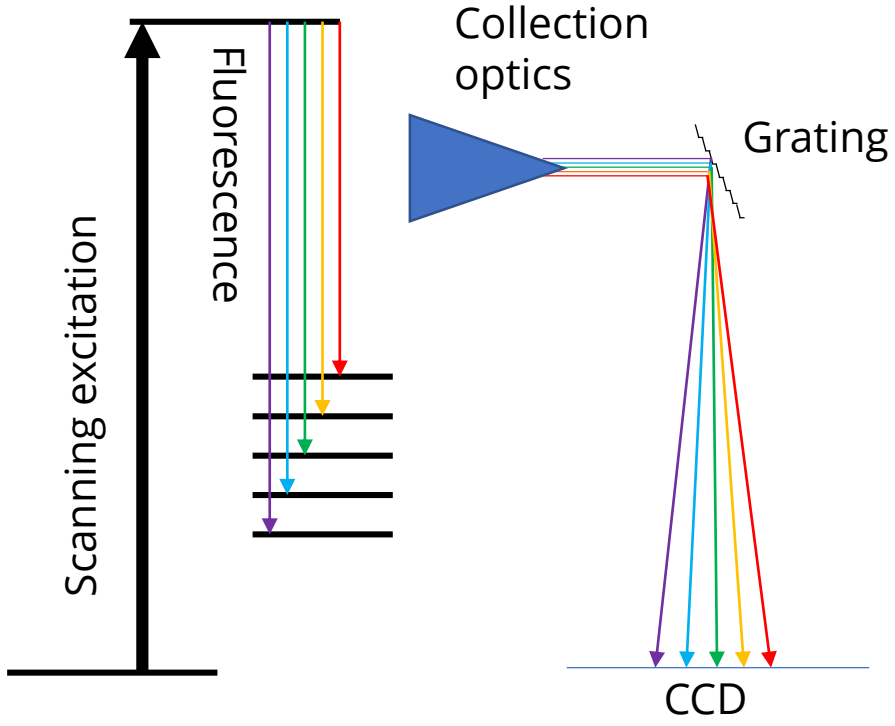
- $^3P_1$ state provides energy to overcome reaction barrier
  - Long enough lifetime to react (~850 ns)
- Populates many vibrational levels
  - Excitated vibrational states generally "empty" in a cold beam…
  - Need population to measure precisely
- Produces ~10^{10} molecules in bending mode X(010)
  - Science state
  - Observed in cryogenic molecular beam
  - Starting detailed study



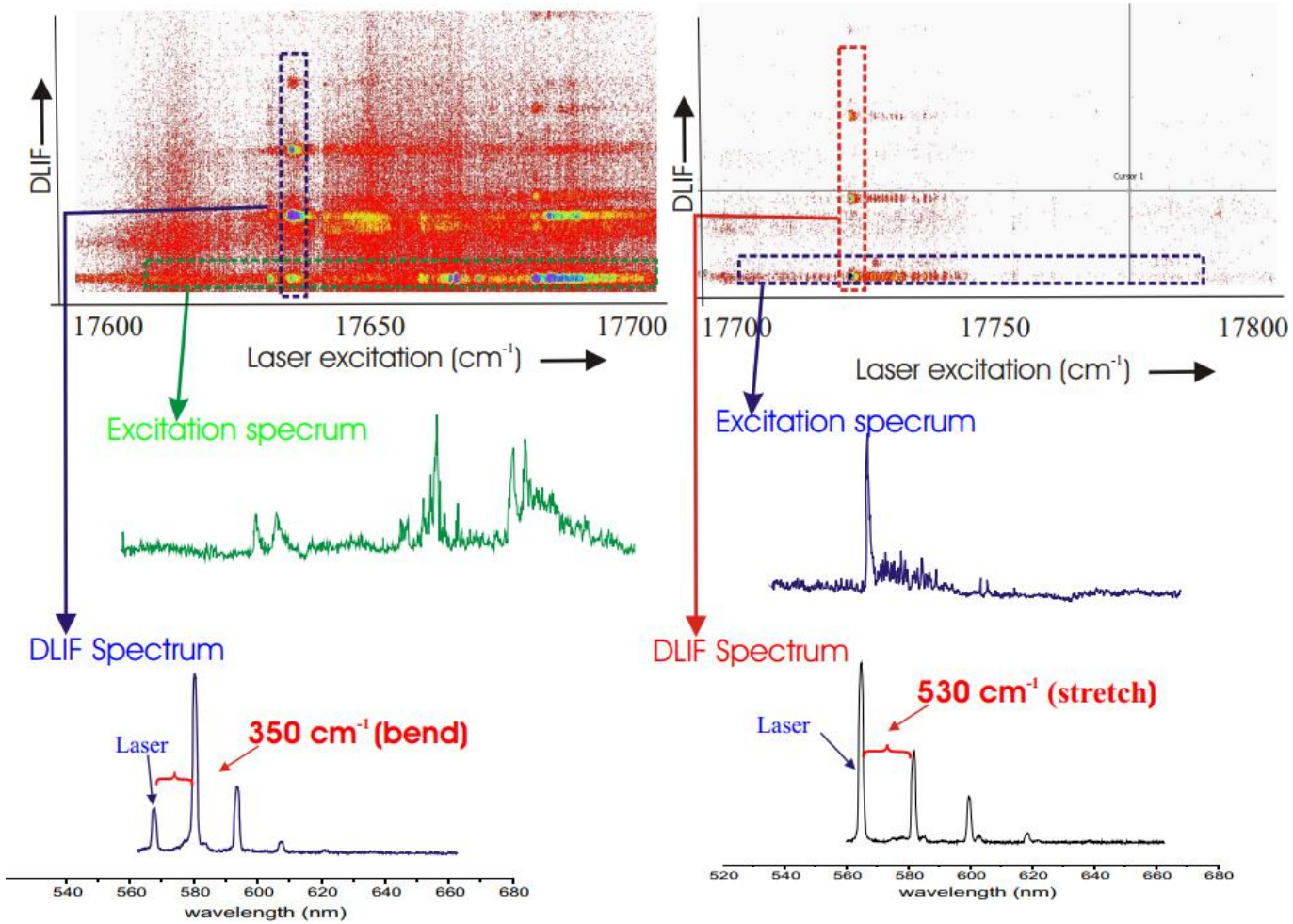
Calculations by Jacek Kłos and Svetlana Kotochigova

Dispersed Fluorescence

- Need to study many levels in the molecule
- Dispersed fluorescence setup @ ASU
  - Gives frequencies and strengths over broad regions
- Over 30 branching ratios measured
- Team effort!
  - Steimle apparatus and people, with regular visits from Caltech and Harvard for running and analysis

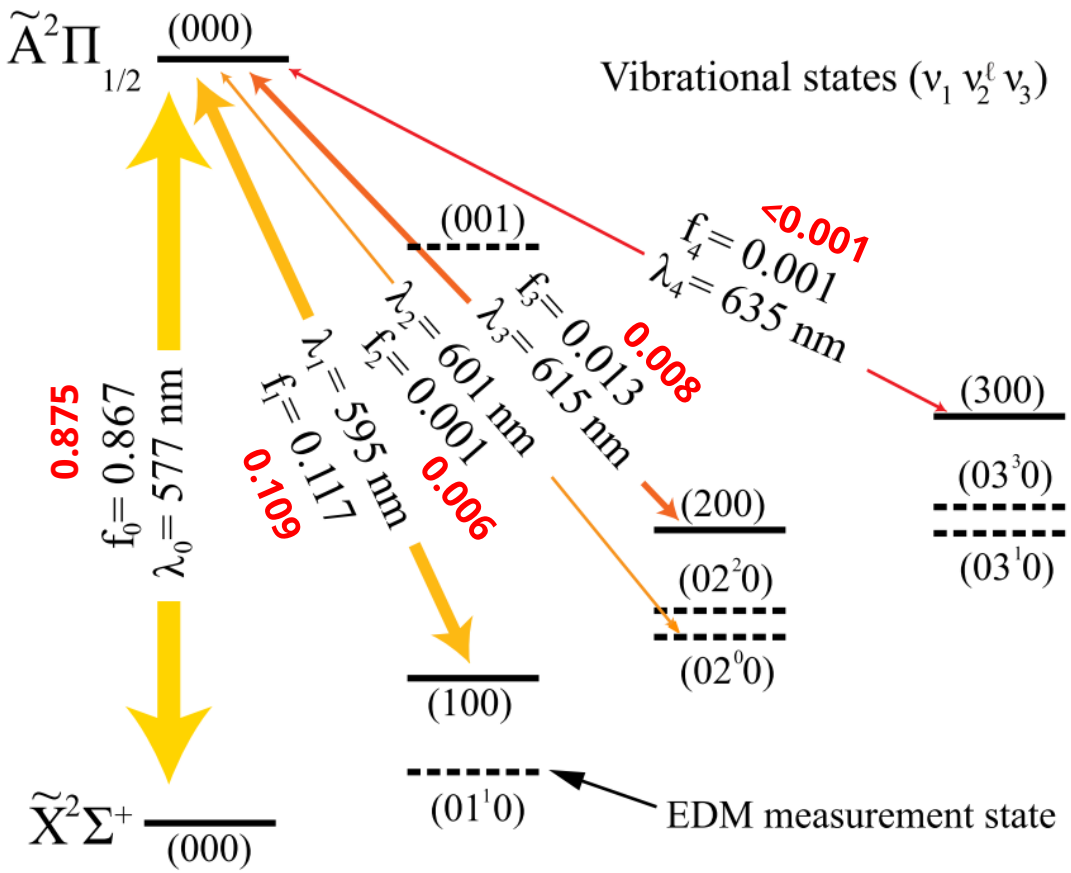


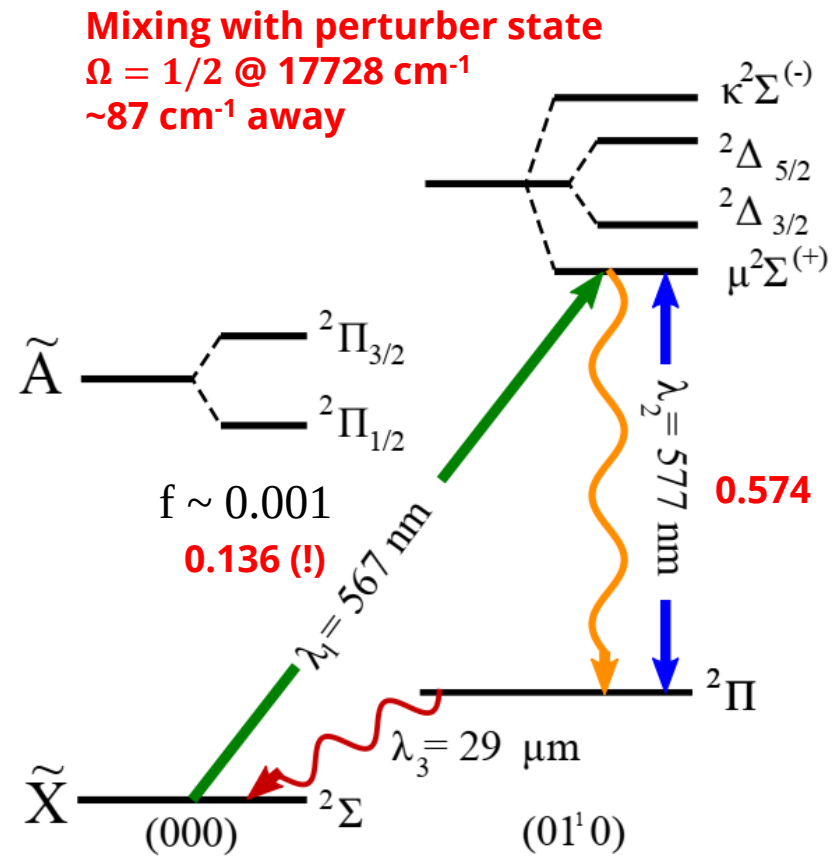
Dispersed Fluorescence in YbOH



Steimle Group @ ASU

Branching Ratios (Preliminary)



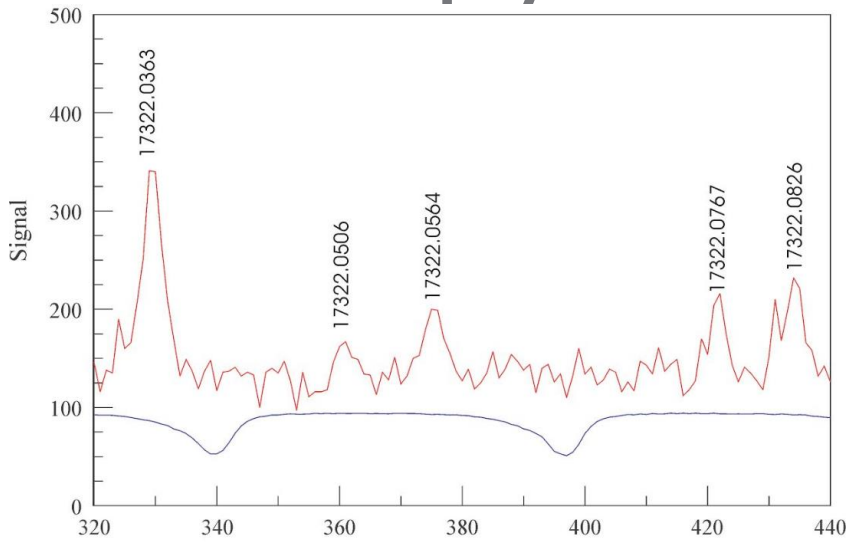


Estimates from I. Kozyryev and NRH, PRL **119**, 133002 (2017). **Measurements** from Steimle Group @ ASU.

E. T. Mengesha, A. T. Le, B. Augenbraun, Z. Lasner, J. M. Doyle, T. C. Steimle, in preparation

High Resolution Spectroscopy

- Dispersed fluorescence gives “low resolution” spectral information
- Follow up with LIF, double resonance, etc.
- Completed/ongoing high resolution spectroscopy of ground state in ^{173,174}YbOH
- Ongoing high resolution spectroscopy of pumping /repumping transitions, Stark, Zeeman, …



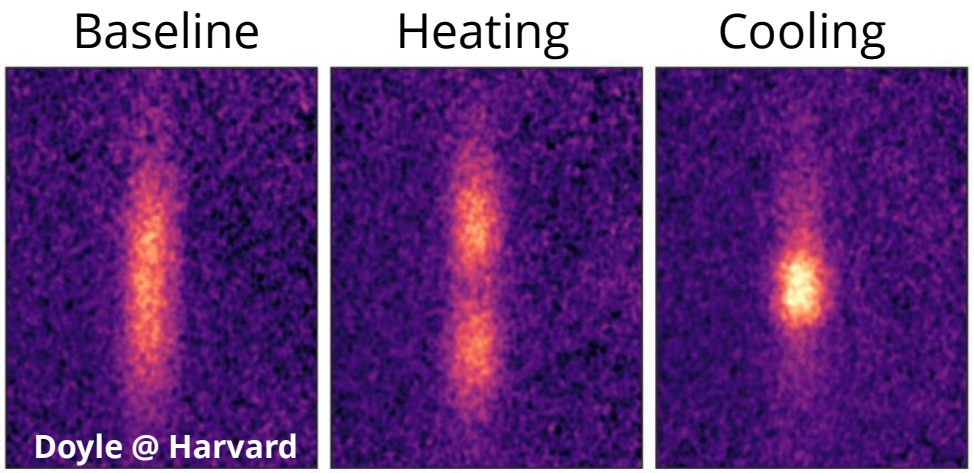
Spectroscopic constants for the $\tilde{X}^2\Sigma^+(0, 0, 0)$ state of ^{174}YbOH (MHz)

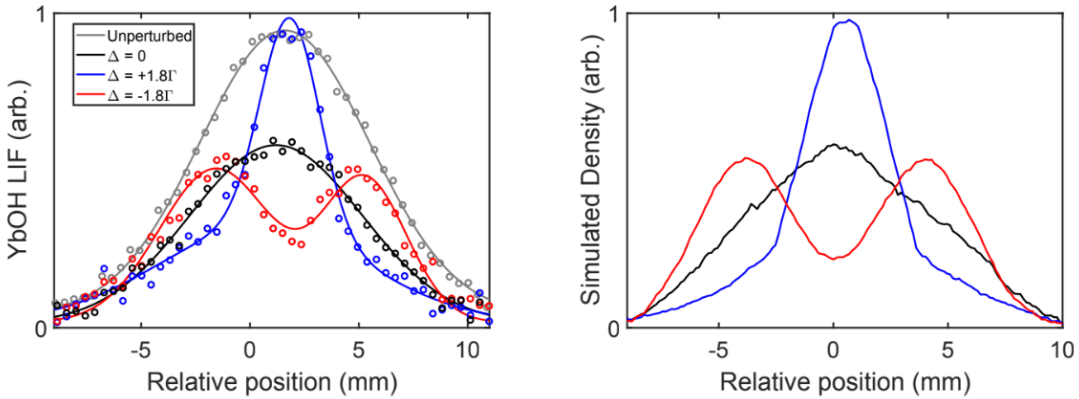
Parameter	Values[a]	
	Present	Previous[b]
B	7348.40053(29)	7357.92(39)
D	0.006084(39)	0.006535(84)
γ	−81.150(57)	28.90(42)
γ_D	0.00476(56)	
b_F	4.80(18)	
c	2.46(48)	

S. Nakhate, T. C. Steimle, N. H. Pilgram, NRH
Chem. Phys. Lett. 715, 105 (2019)

Laser Cooling

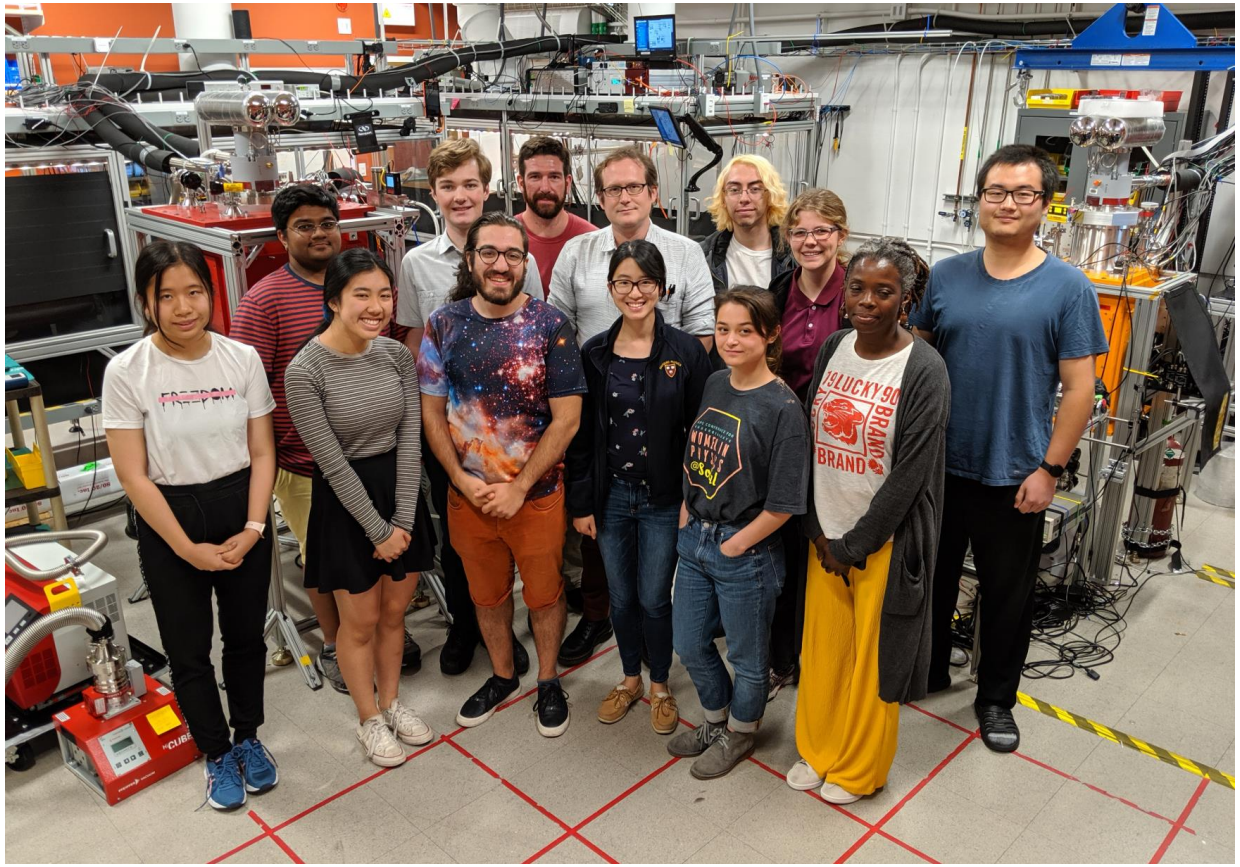
- Doyle Group @ Harvard
- Demonstrated Doppler and Sisyphus cooling of YbOH in 1D
  - Scattering ~500 photons with 3 repumps
  - Consistent with estimates
- Measured T < 600 μK
  - Probably much colder, limited by time of flight
  - More photons on the way…





B. L. Augenbraun, Z. D. Lasner, A. Frenett, H. Sawaoka, C. Miller, T. C. Steimle, J. M. Doyle. In preparation.

Caltech



Hutzler Lab, Summer 2019

Thanks for your attention!

Special thanks for the invitation

Come visit!

www.hutzlerlab.com

www.polyedm.com

HEISING-SIMONS FOUNDATION

NIST

U.S. DEPARTMENT OF ENERGY

Office of Science

NSF