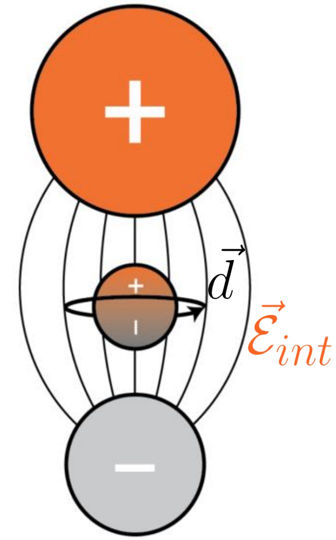
Caltech

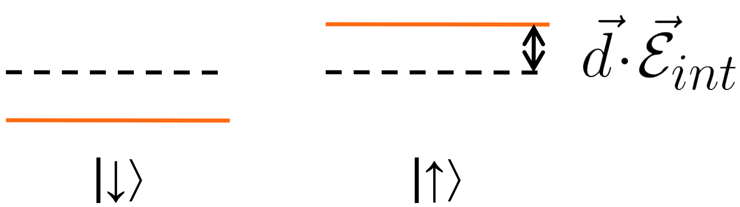
Searching for New Fundamental Physics with Polyatomic Molecules

Part I: Nick Hutzler, Caltech
Part II: Ben Augenbraun, Harvard

Why molecules for CPV?

- Molecules have **extremely large** fields
  - 10-100 GV/cm
  - Relativistic ~Z^3 enhancement
  - ~10^3 larger than fields in atoms
- CPV moments cause CPV energy shifts
- Molecule experiments probing ~10 TeV scales for new physics
  - ACME, Nature **562**, 355 (2018)

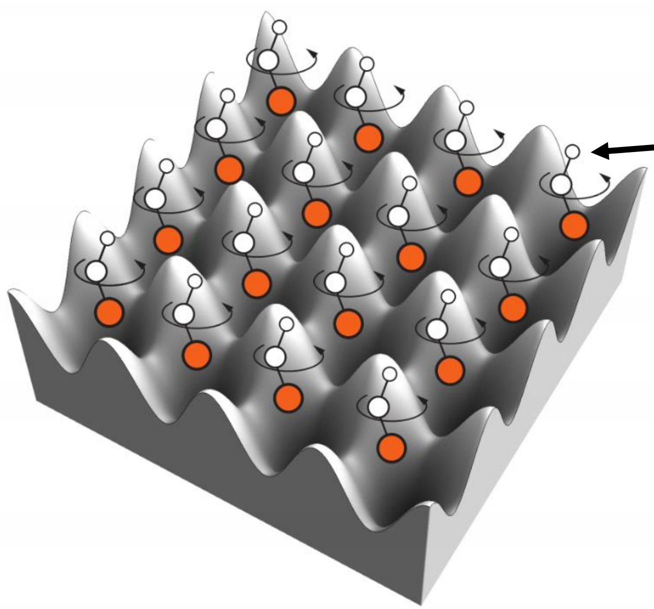




How can we keep improving?

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

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

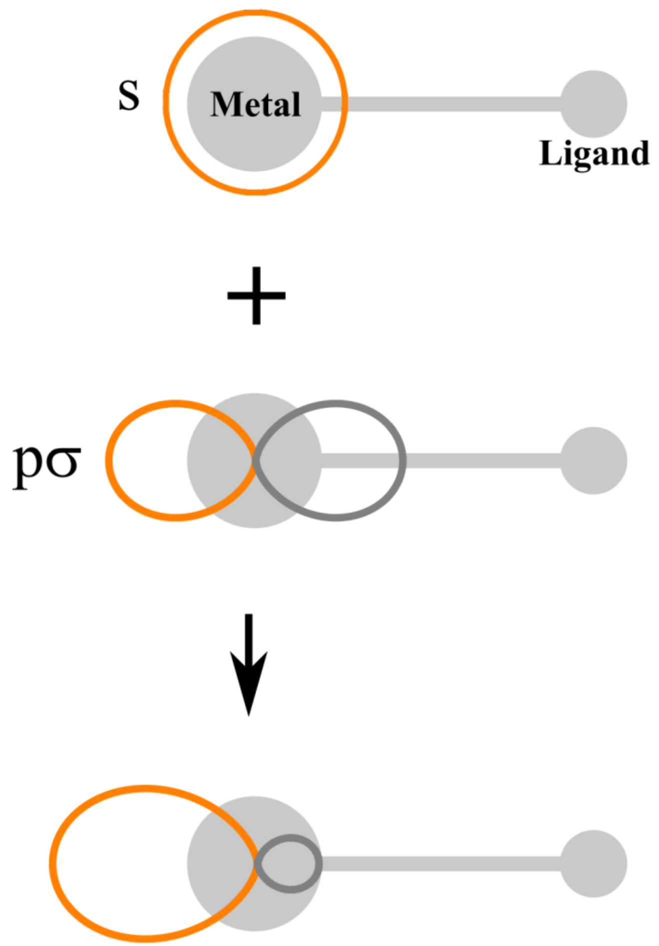


Heavy, polar molecule with advanced quantum control

Need techniques for ultracold quantum control
→ Laser Cooling

Electronic Structure for Laser Cooling

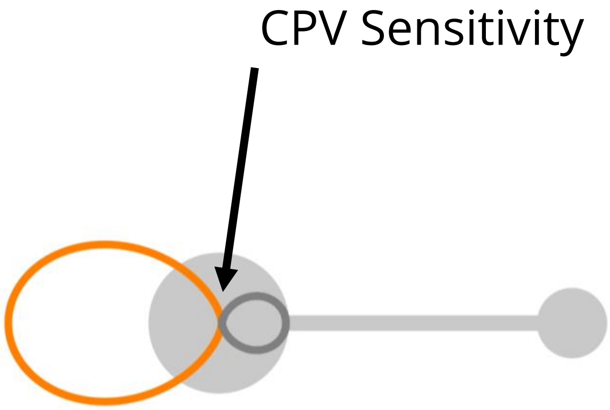
- See Tarbutt, Doyle talks!
- Generally works for molecules with single, metal-centered *s* electron
  - Alkaline-earth (s^2)
  - Single bond to halogen (F)
- Orbital hybridization pushes electron away from bond
- Works for "any" bonding partner – polyatomics
  - Metal-centered electronic structure is robust
  - F → -OH, -CCH, $-OCH_3$, ...



A. M. Ellis, Int. Rev. Phys. Chem. 20, 551 (2001)
M. D. Di Rosa, Eur. Phys. J. D 31, 395 (2004)
M. V. Ivanov, F. H. Bangerter, and A. I. Krylov, PCCP 21, 19447 (2019)
T. A. Isaev and R. Berger, PRL 116, 63006 (2016)
I. Kozyryev, L. Baum, K. Matsuda, and J. M. Doyle, ChemPhysChem 17, 3641 (2016)
I. Kozyryev et al., PRL 118, 173201 (2017)

Compatibility with CPV Searches

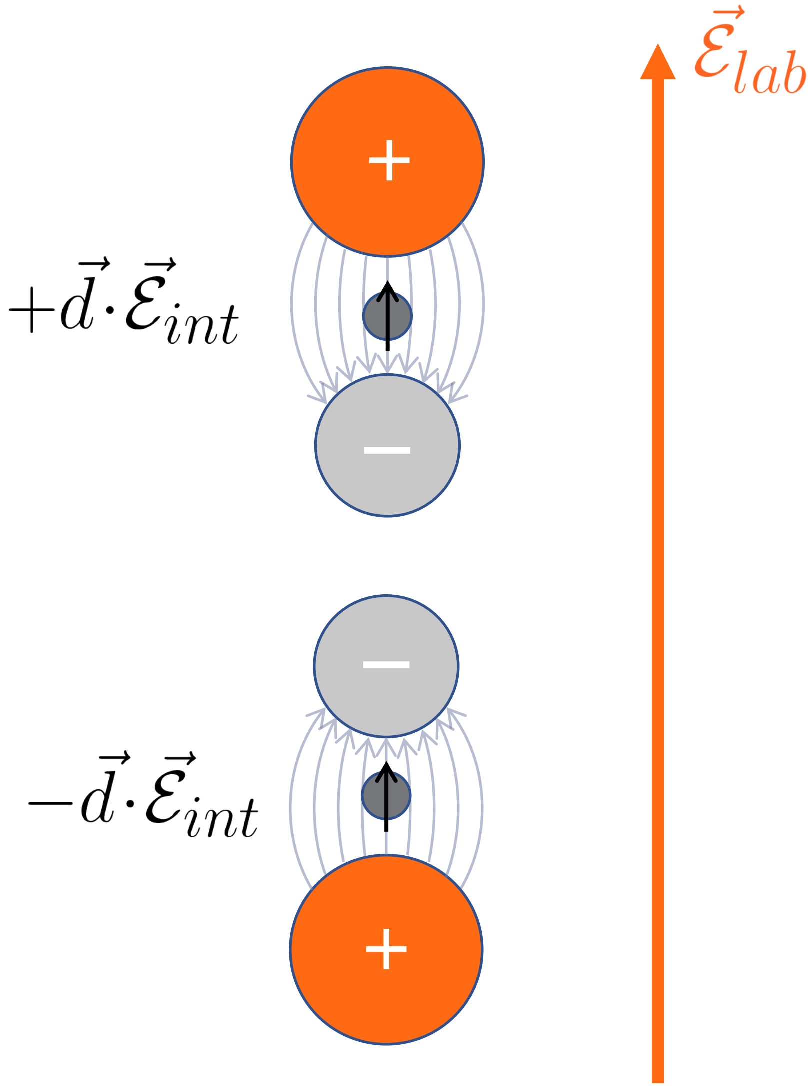
- s electrons are good for CP violation searches
  - Relativistic ~Z^3 enhancement from nuclear core penetration
- BaF, HgF, RaF, TlF, YbF, ...
  - Sensitive to CPV, laser-coolable
  - Several being actively pursued (and discussed at this workshop)
- Generally seems to hold for polyatomic analogues
  - F → -OH, -CCH, -OCH_3, ...
- **So why *polyatomic*?**



T. A. Isaev, A. V. Zaitsevskii, & E. Eliav, J. Phys B. 50, 225101 (2017)

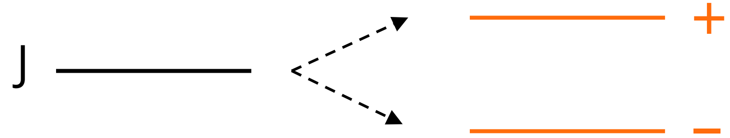
Internal Comagnetometers

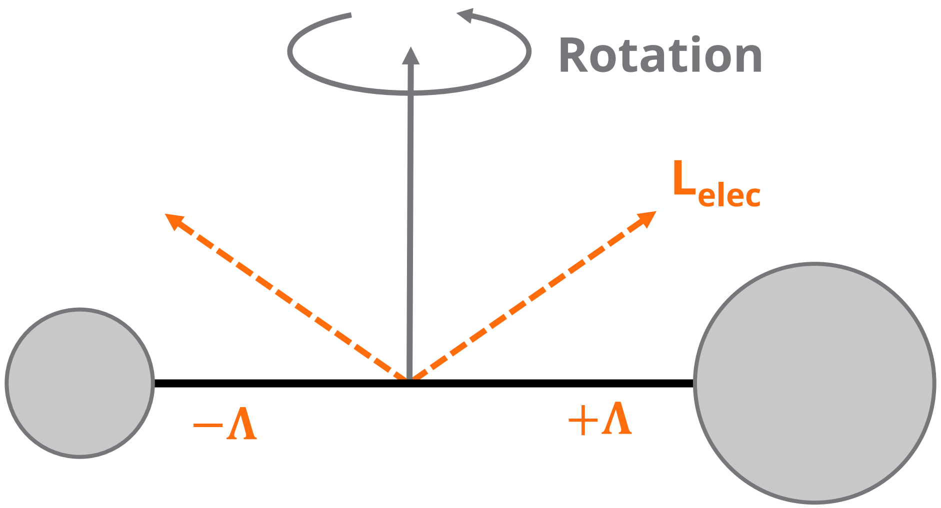
- ACME, JILA experiments rely on full polarization
  - Measure CPV in each state
  - “Internal comagnetometer”
  - No external field change required
- Requires particular structure – parity doublets
  - Closely-spaced states of opposite parity, fully mixed in small fields



Parity Doublets

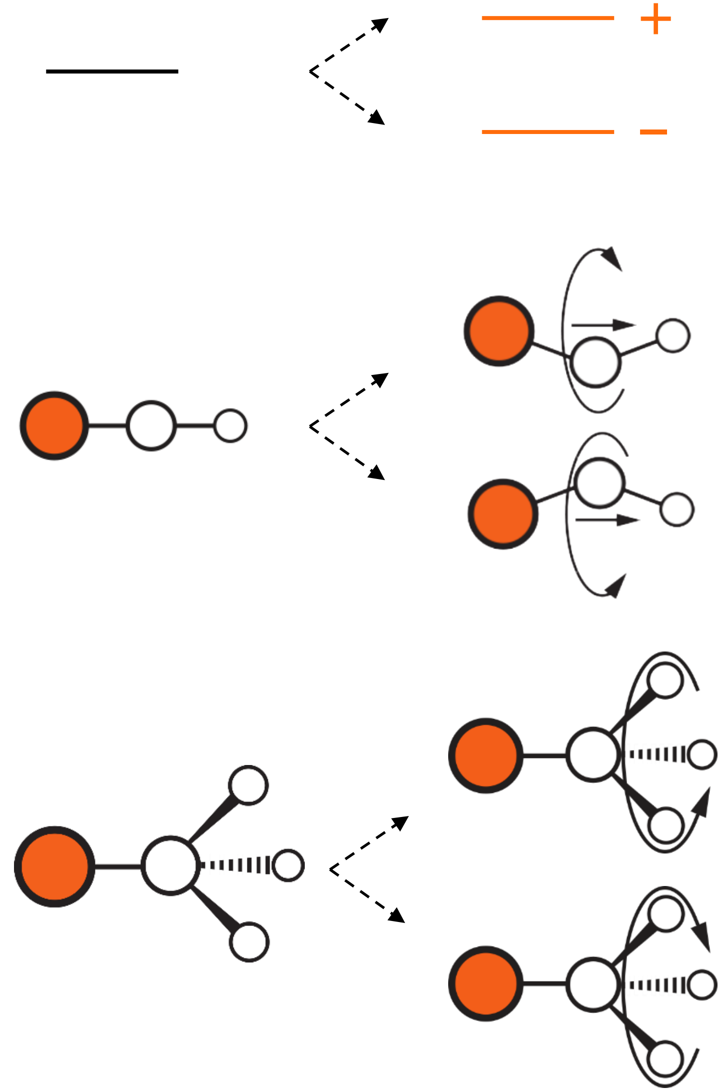
- Hard to find in diatomics
  - Requires $L_{elec}>0$ (Π, Δ, …)
- Limits options
  - At odds with simple structure needed for laser cooling, ultracold assembly
  - Many won't have doublets
    - BaF, HgF, RaF, TlF, YbF, …





Polyatomic Molecules

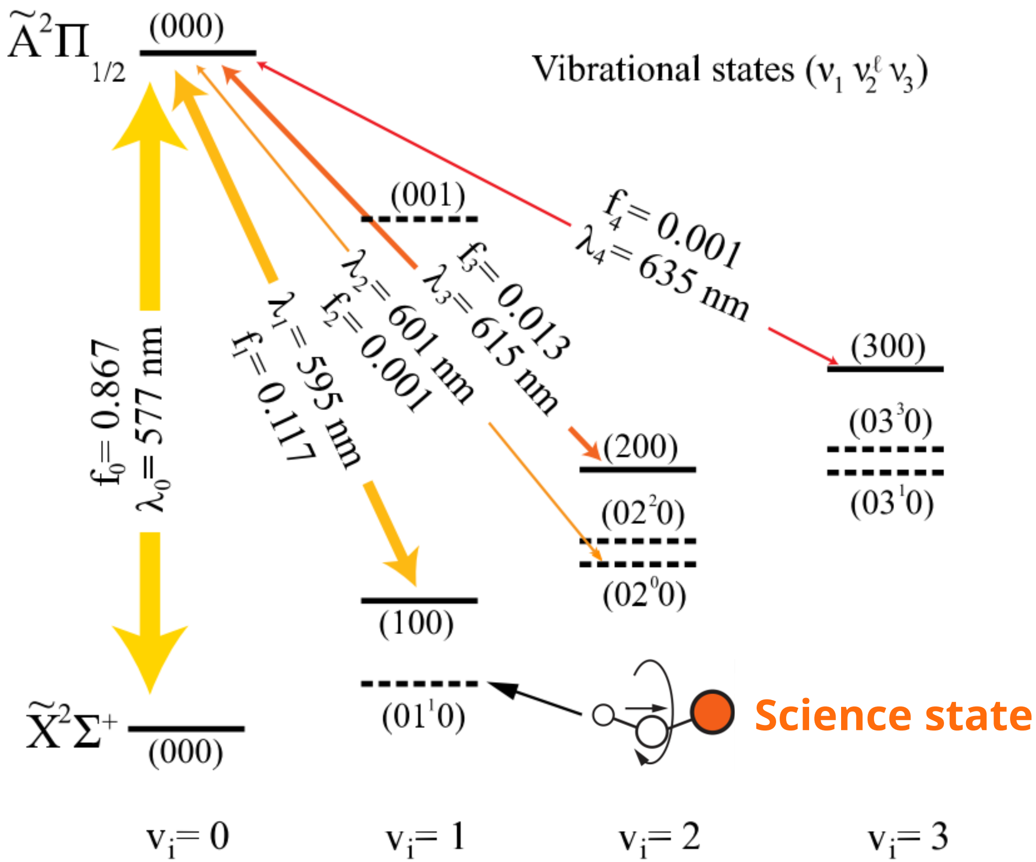
- Polyatomics *generically* have parity doublets
  - (>3 atoms)
  - Arise from symmetry-lowering mechanical modes
- Available for any atomic species
  - Neutrals and ions
  - Only way for some, like Ra
- Many future directions with other species, ligands
- Many applications outside what is discussed here
  - See Doyle talk
  - Review: NRH, Quantum Science and Technology 5, 044011 (2020)



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

YbOH

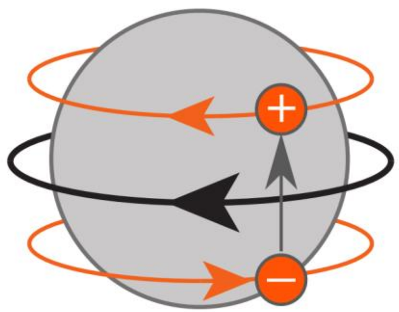
- YbOH combines laser cooling, sensitivity to symmetry violation, and high polarizability
  - Yb is similar to an alkaline earth (s^2 valence)
  - OH is similar to F
  - Highly polarizable, metastable (~1 s), dynamically bent state
- Broad physics reach via multiple stable isotopes

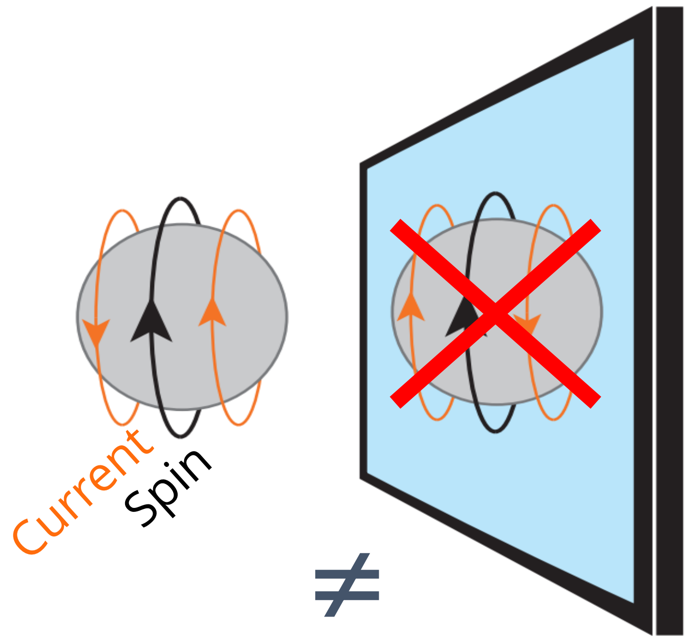


I. Kozyryev and NRH, Phys. Rev. Lett. 119, 133002 (2017)

Nuclear Magnetic Quadrupole Moments

- Violates CP, enhanced in molecules
- NMQM arises from hadronic sources
  - Nucleon EDM
  - quark EDM/chromo-EDM
  - CPV nuclear forces
  - Strong CPV (θ_{QCD})
  - …
- Quadrupole deformation (β_2) enhances MQM
  - Collective enhancement
  - Typically $\beta_2 Z \sim 10$

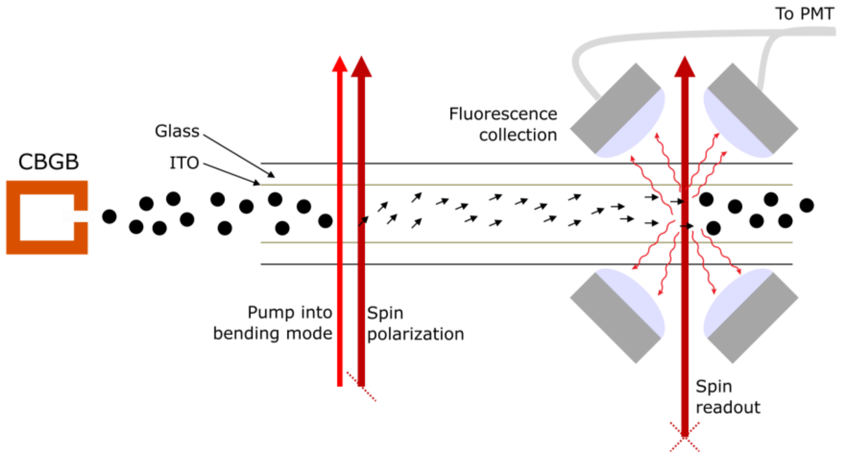


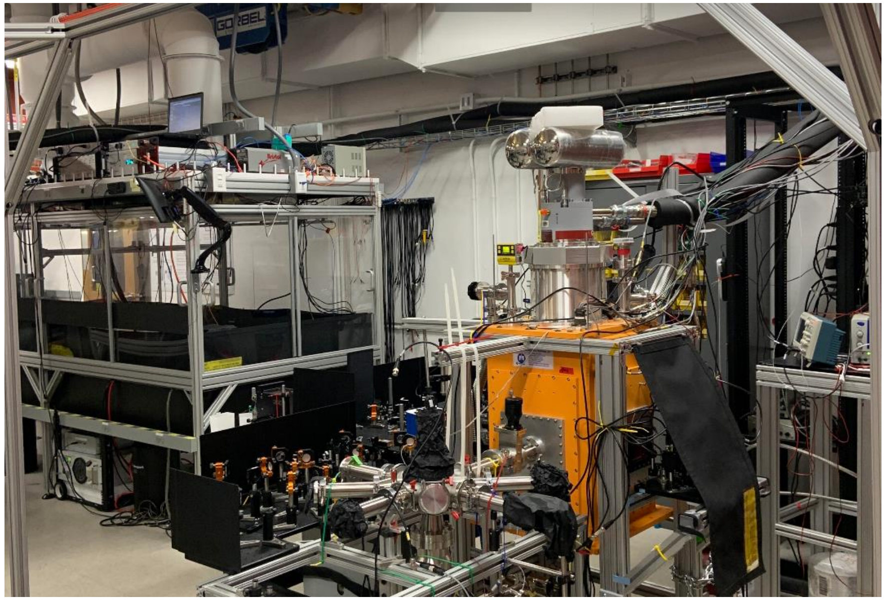


V. V. Flambaum, D. DeMille, and M. G. Kozlov, PRL 113, 103003 (2014)

^{173}YbOH NMQM Experiment @ Caltech

- Building a NMQM search in ^{173}YbOH at Caltech
  - ^{173}Yb (I=5/2), large quadrupole deformation ($\beta_2 \approx 0.3$)
  - Cryogenic buffer gas beam experiment
  - Laser cooling, trapping in future generations

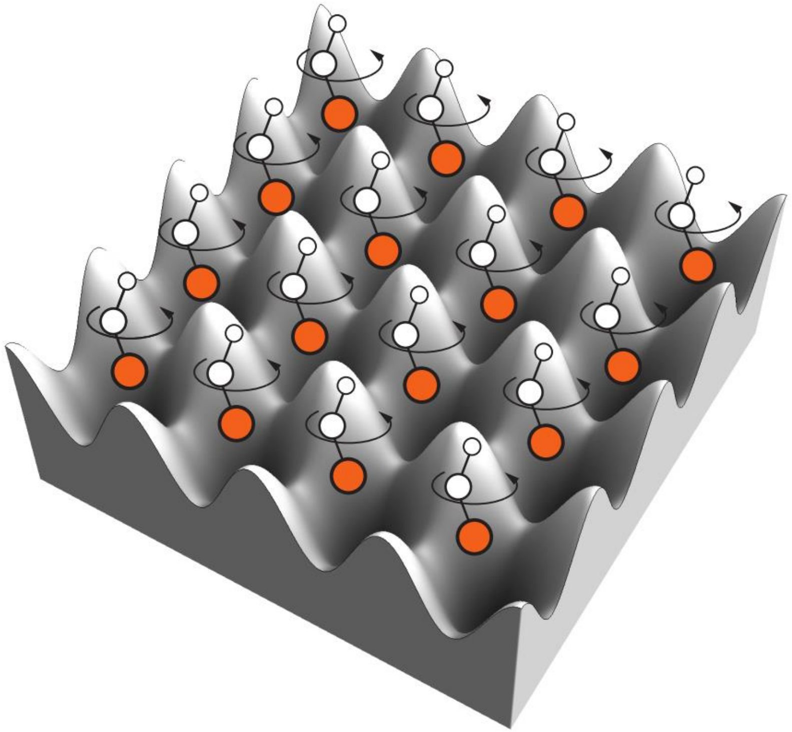




I. Kozyryev and NRH, PRL 119, 133002 (2017)
D. E. Maison, L. V. Skripnikov, and V. V. Flambaum, PRA 100, 032514 (2019)
M. Denis, Y. Hao, E. Eliav, NRH, M. K. Nayak, R. G. E. Timmermans, A. Borschesvky, J. Chem. Phys. 152, 084303 (2020)

Polyatomic eEDM Experiment

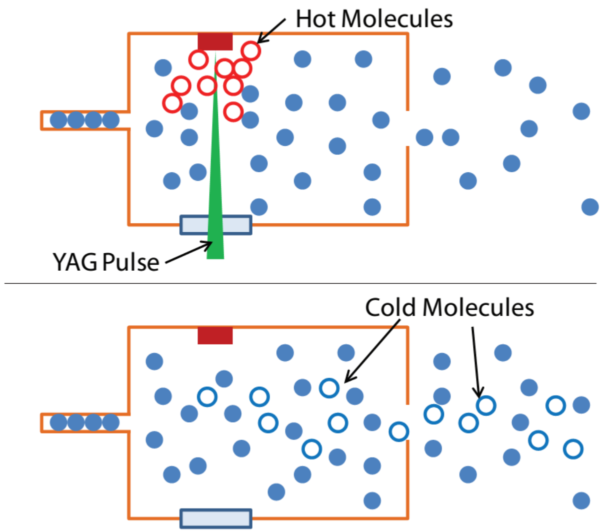
- Electron EDM search in laser cooled and trapped polyatomic molecules
  - NRH @ Caltech
  - John Doyle @ Harvard
  - Tim Steimle @ ASU
  - Amar Vutha @ Toronto
- ^{174}YbOH (I_{Yb}=0)
- *www.polyedm.com*



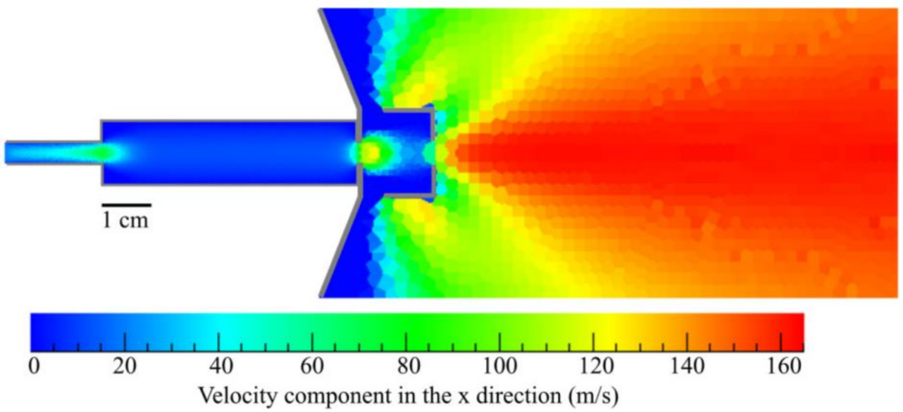
Recent/Current Work

Buffer gas cooling

- Use cryogenic inert gas to cool via collisions
- Surprisingly hard to simulate
  - Length scales vary by ~10^5
  - Non-equilibrium, mixed mass, …
- Developed and implemented a mean field approach
- Accurately reproduces non-trivial behavior
  - Next up – use to improve!
  - See work at Harvard
- Generally useful for modeling gas thermalization



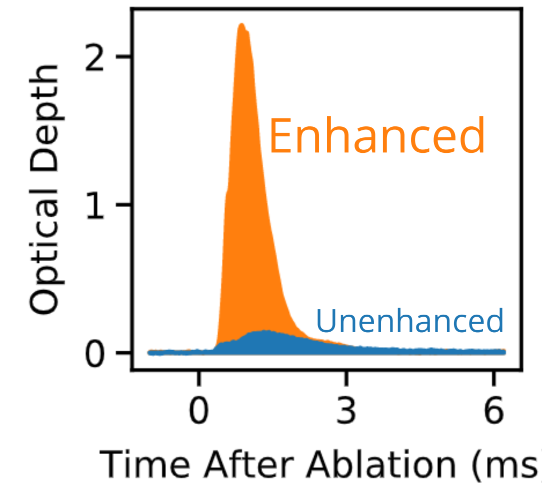
NRH, H. Lu, and J. M. Doyle, Chem Rev 112, 4803 (2012)

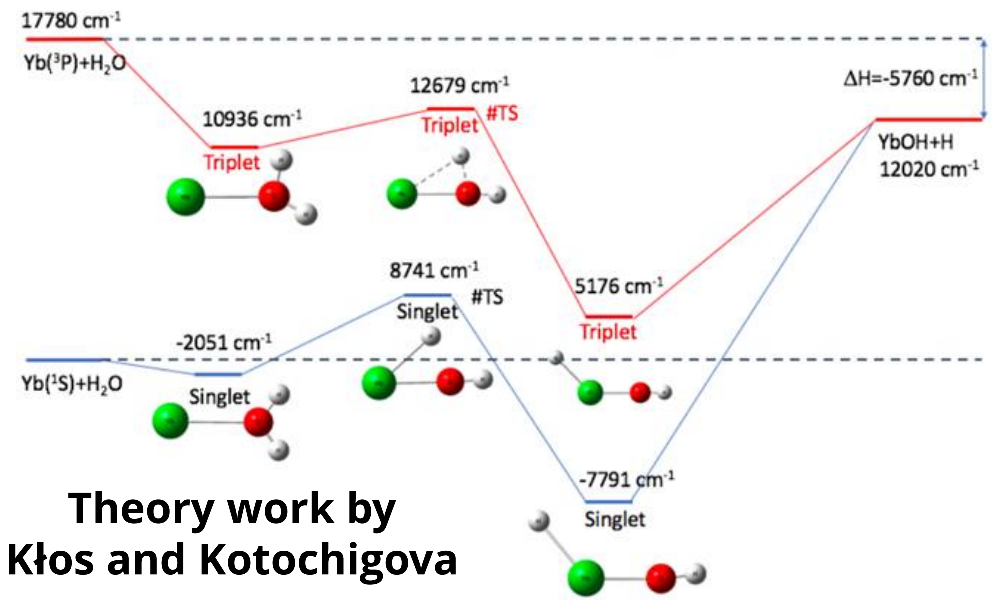


Y. Takahashi, D. Shlivko, G. Woolls, NRH
Phys. Rev. Research 3, 023018 (2021)

Molecular Production via Optically-Driven Cold Chemistry

- Enhanced chemical production in cryogenic molecular source
  - Vaporize Yb+$Yb(OH)_3$
  - Excite Yb $^1S_0 \rightarrow {}^3P_1$
  - $Yb(^3P_1)$ reacts with ablation products to create YbOH
- ~10x enhancement
- Fairly general method
  - Immediately applicable to alkaline earth molecules (Ca, Sr, Ba, Ra, …)





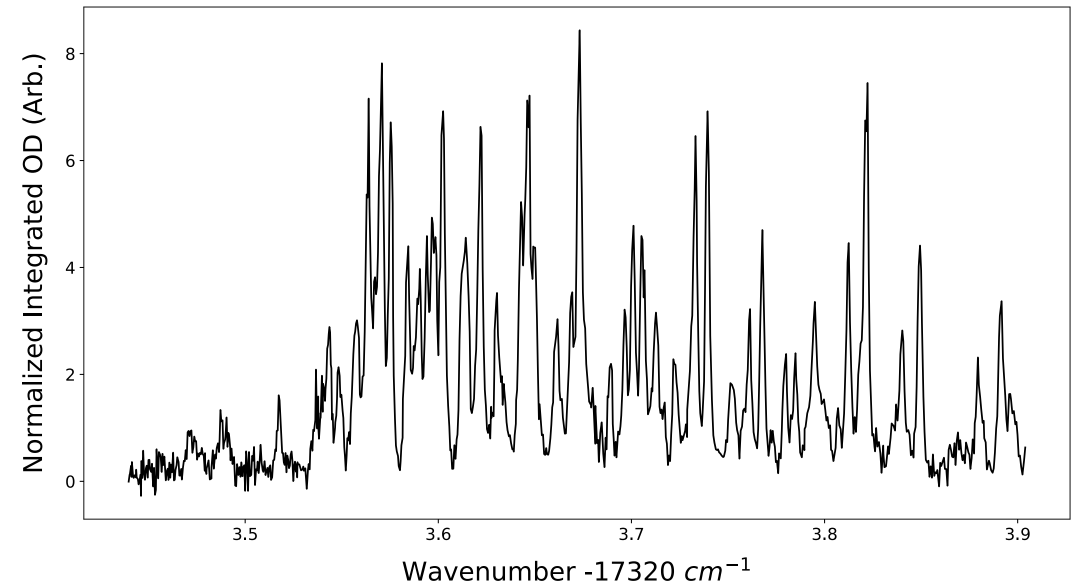
Theory work by Kłos and Kotochigova

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

^{173}YbOH Spectroscopy

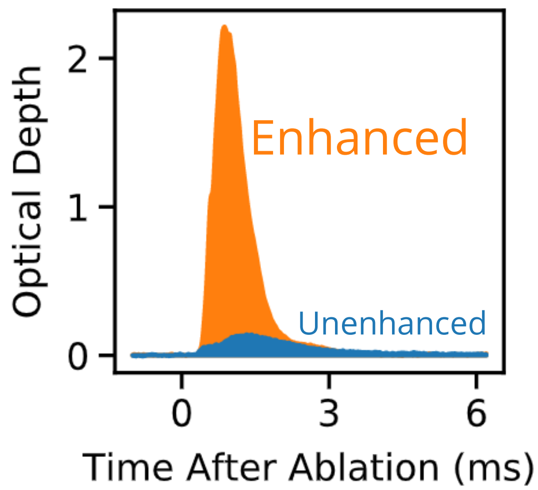
- YbOH is complicated
  - Rotational, hyperfine, and isotope shifts (6 abundant) are ~same
- Recently characterized main electronic transition in ^{173}YbOH
  - Need for MQM
  - Collaboration with Tim Steimle @ ASU
  - Had to a develop a new method

Mix of several isotopes; rotational and hyperfine transitions

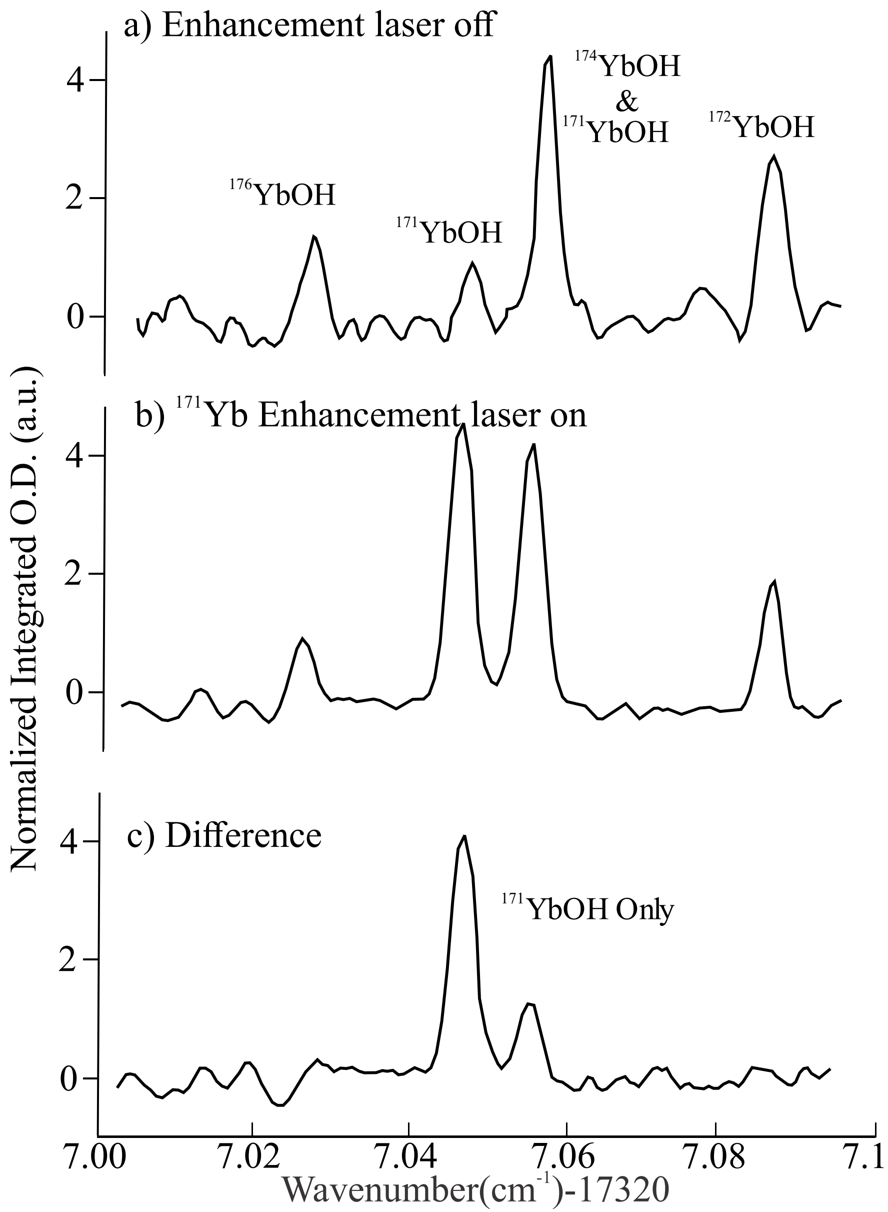


S. Nakhate, T. C. Steimle, N. H. Pilgram, NRH. Chem. Phys. Lett. 715, 105 (2019)
N. H. Pilgram, A. Jadbabaie, Y. Zeng, NRH, T. C. Steimle arXiv:2104.11769 (2021) Accepted to J. Chem. Phys.

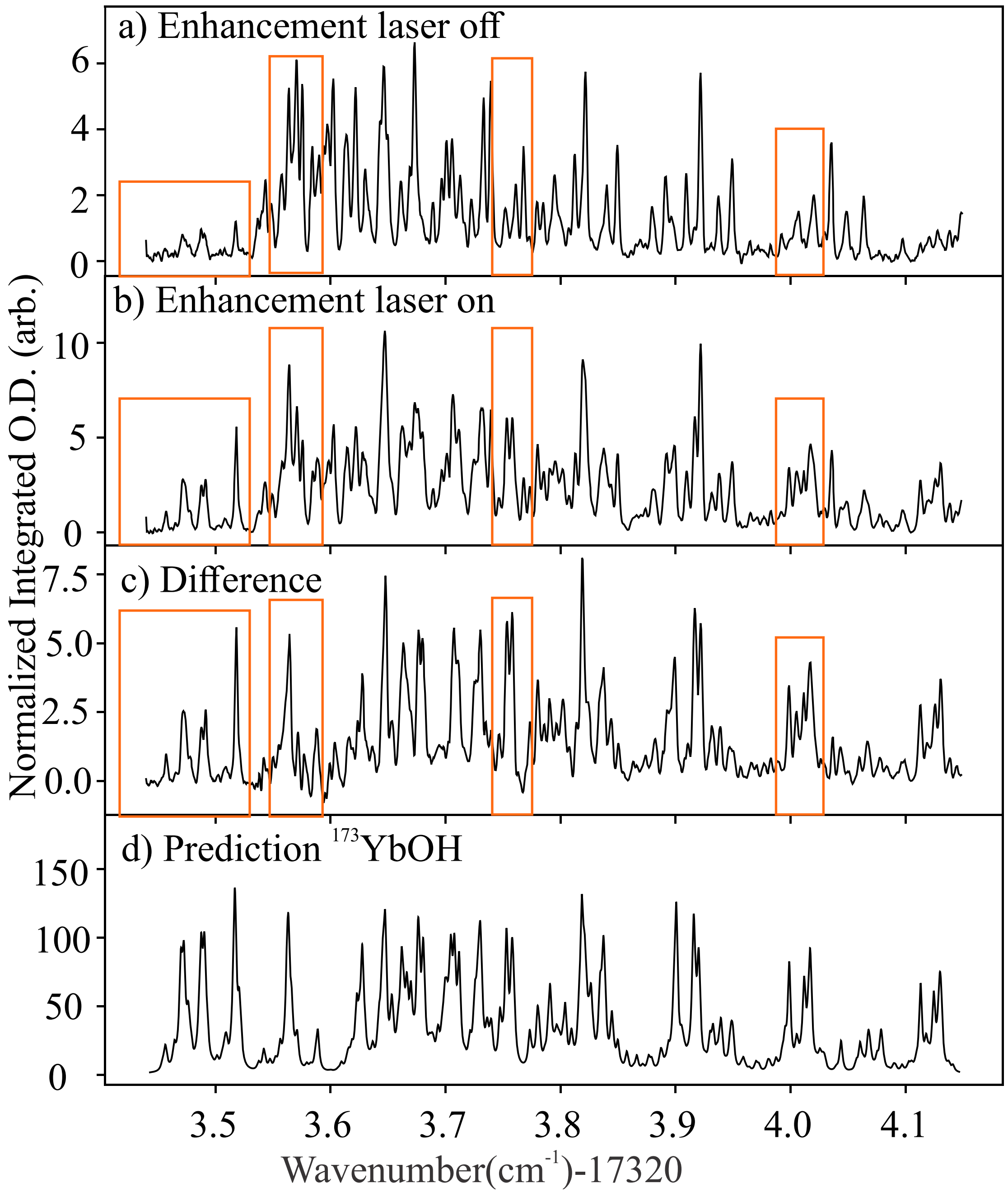
Chemical Enhancement for Spectroscopy

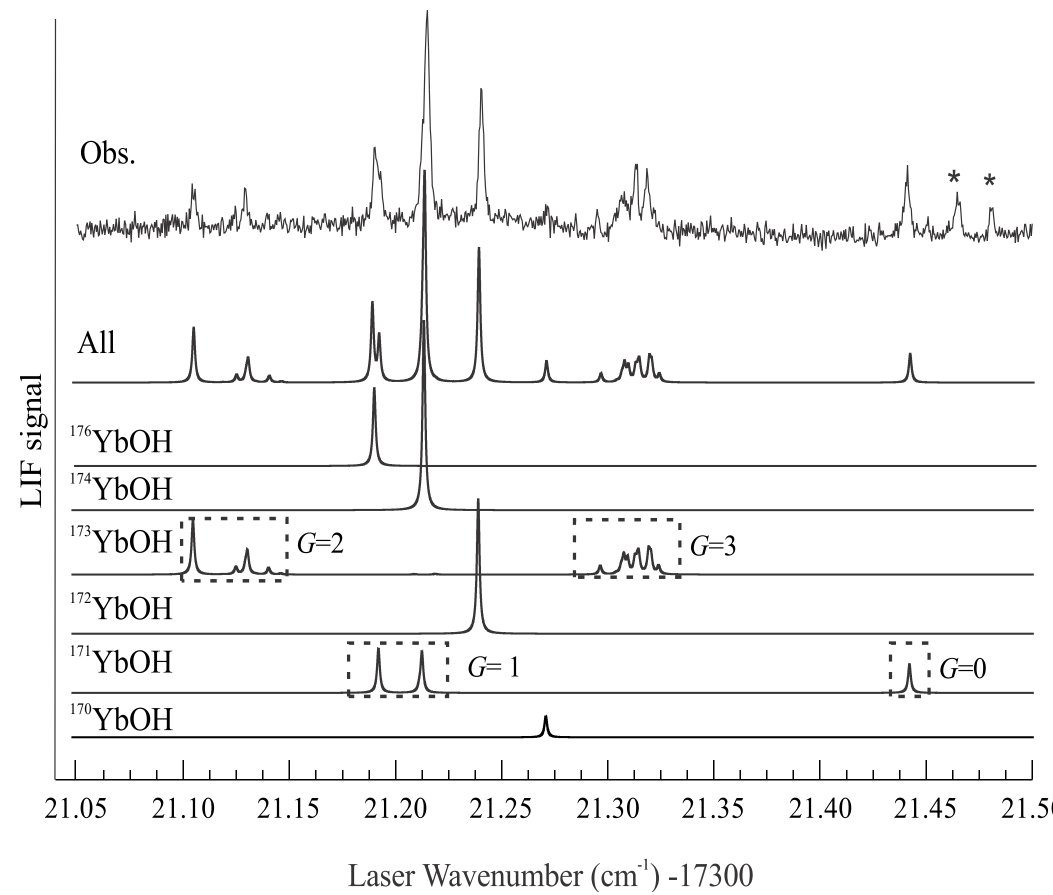


Able to directly isolate signals of different isotopologues using chemical enhancement



N. H. Pilgram, A. Jadbabaie, Y. Zeng, NRH, T. C. Steimle arXiv:2104.11769 (2021) Accepted to J. Chem. Phys.



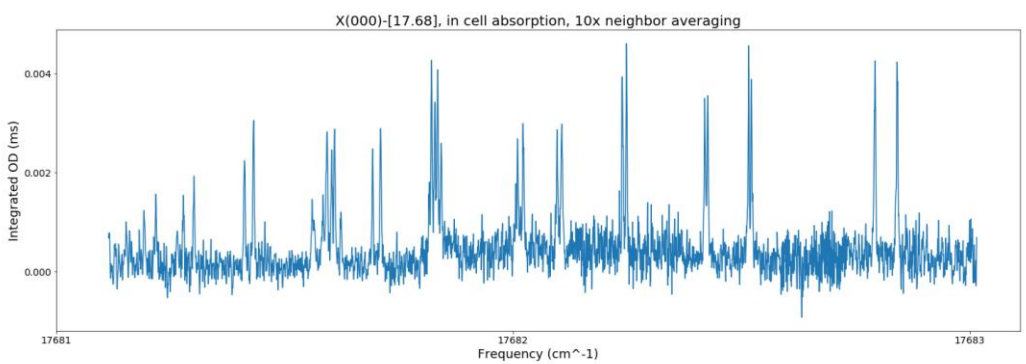


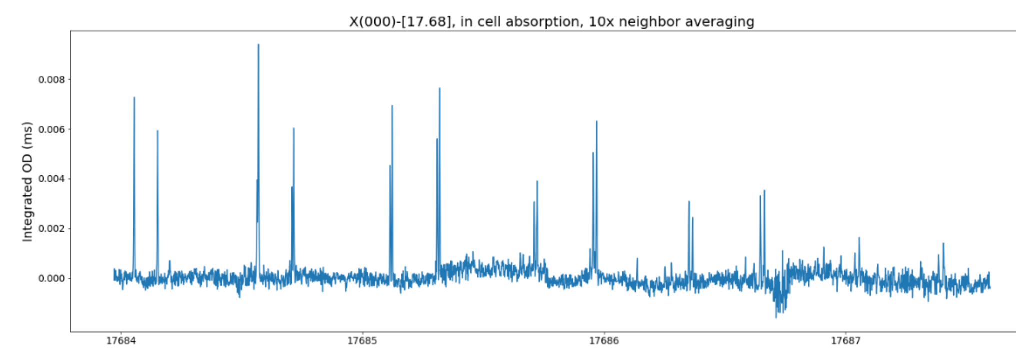
N. H. Pilgram, A. Jadbabaie, Y. Zeng, NRH, T. C. Steimle arXiv:2104.11769 (2021) Accepted to J. Chem. Phys.

Current work: Science State

Collaboration with
Tim Steimle

- Chemical production populates science state
  - ~10^{10} molecules
- High resolution spectra on three transitions involving bending states in $X^2\Sigma$, $A^2\Pi_{1/2}$
- Very complicated due to f-shell states, vibronic mixing
- Currently analyzing, taking more data

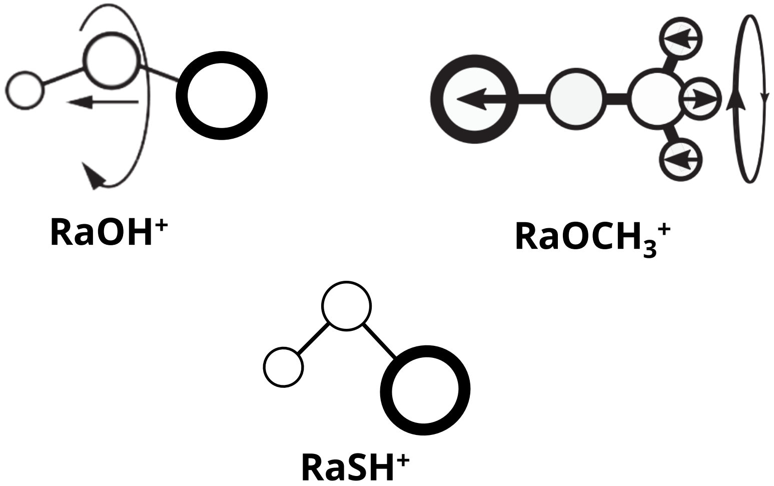




Portions of X(0,0,0) → [17.68] [nominal $A^2\Pi_{1/2}(0,1,0)$]

Theoretical Work

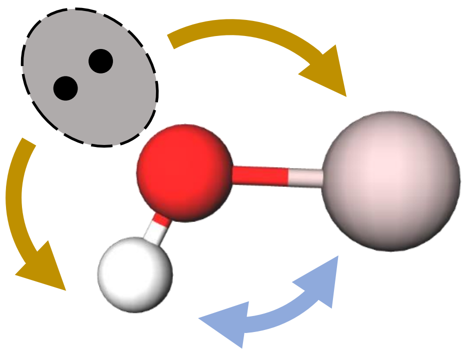
Radium polyatomic molecular ions



Combining deformed nuclei with ion trapping and polyatomic advantages

P. Yu and NRH, PRL 126, 023003 (2021)
I. Kozyryev and NRH, PRL 119, 133002 (2017)

New recipes for laser-coolable molecules



Other approaches for when F → pseudohalogen fails is doesn't apply

P. Yu, A. Lopez, W. A. Goddard, NRH, *in preparation*

Caltech

Hutzler Lab
Summer 2020

Collaborators

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

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

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

Come visit… some time!

www.hutzlerlab.com
www.polyedm.com

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NIST

U.S. DEPARTMENT OF ENERGY
Office of Science

NSF

Part II

Ben Augenbraun
Harvard University

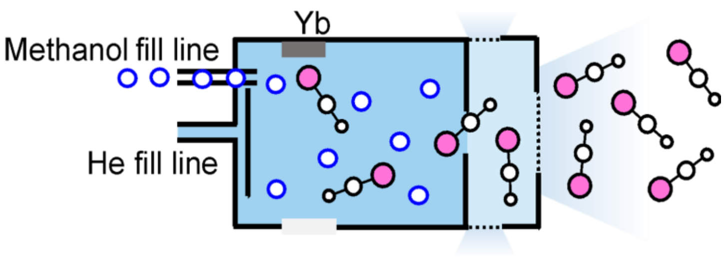
The Path to Ultracold Molecules

Our goal: 10^5 trapped YbOH molecules,
>1 s coherence time,
$<10^{-31}$ e.cm sensitivity…

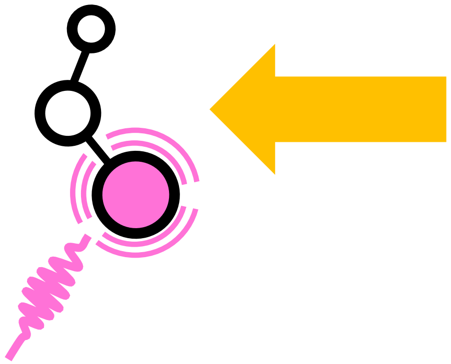
… which requires ultracold temperatures, typically in the μK regime…

… which requires molecules produced with low forward velocities, and further slowed to trap's capture velocity.

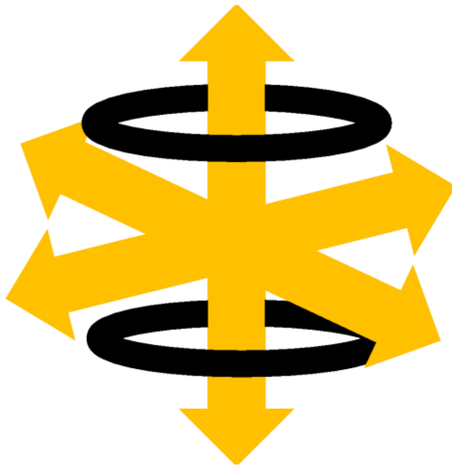
Buffer-gas beam source



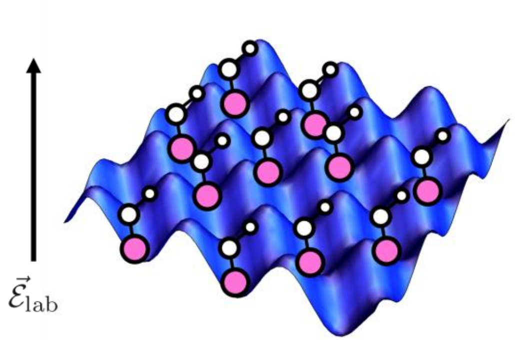
Slowing



Laser Cooling

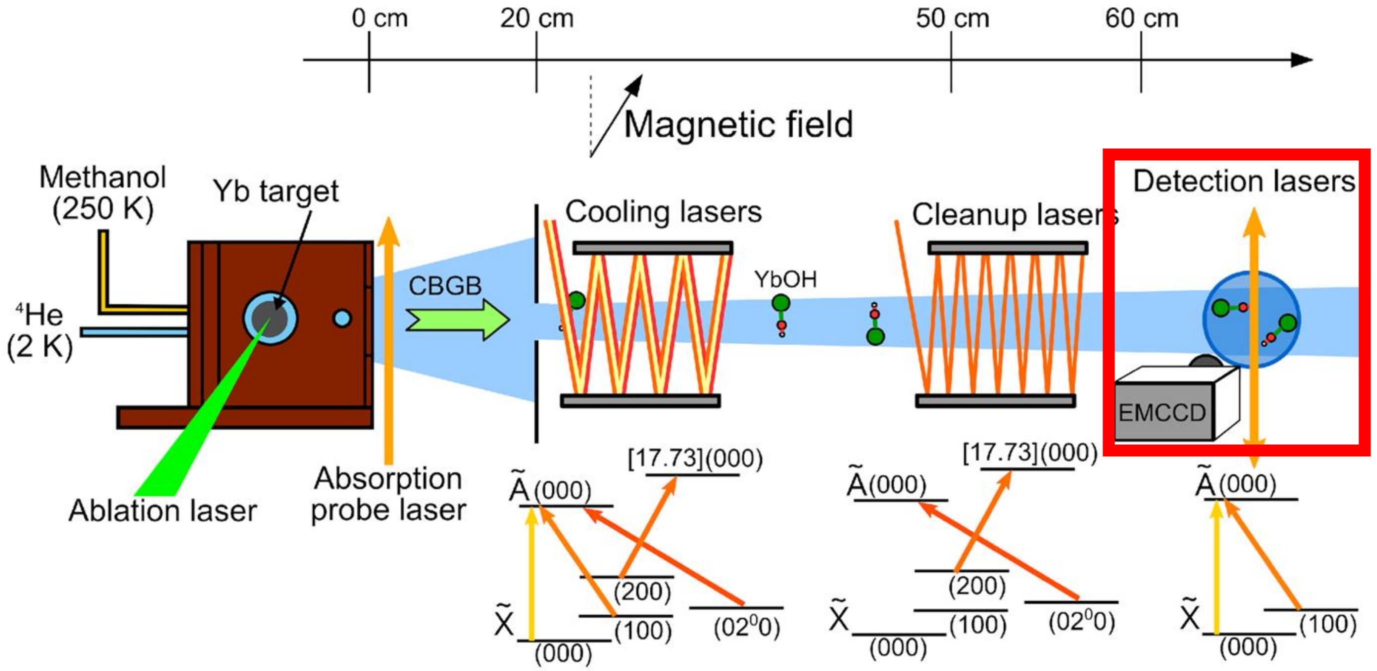


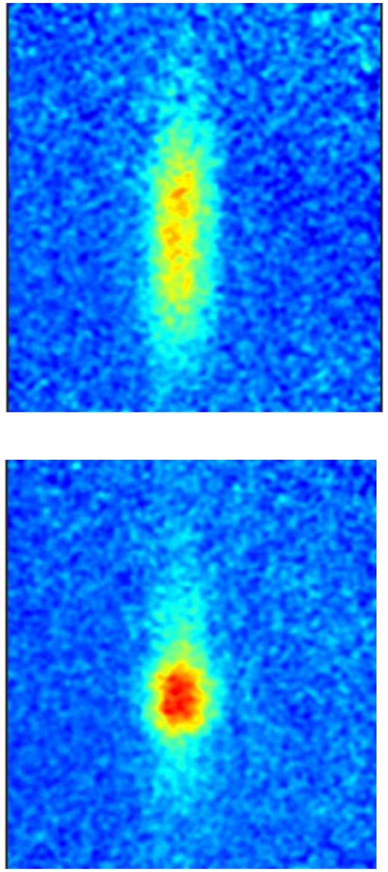
Optical Trapping



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

Laser Cooling of YbOH [from Doyle talk]



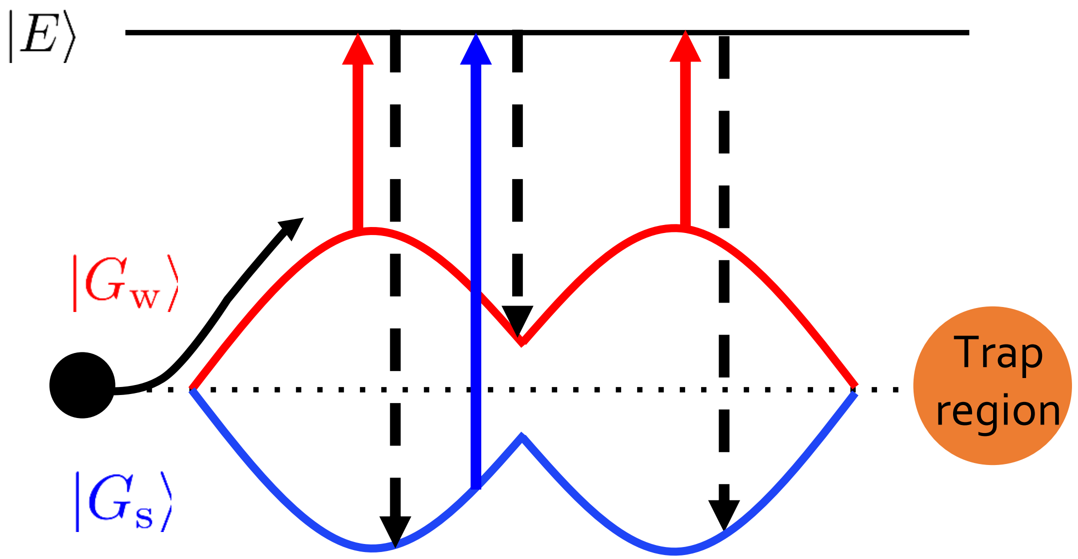


- Efficient 1D cooling of YbOH observed
- Extending to 3D requires ~10-100x more photon scatters
  - That's **possible** but ***hard*!** Don't want to waste the photons all on slowing to capture velocity…

BLA, …, Doyle, New J. Phys. 22 022003 (2020)

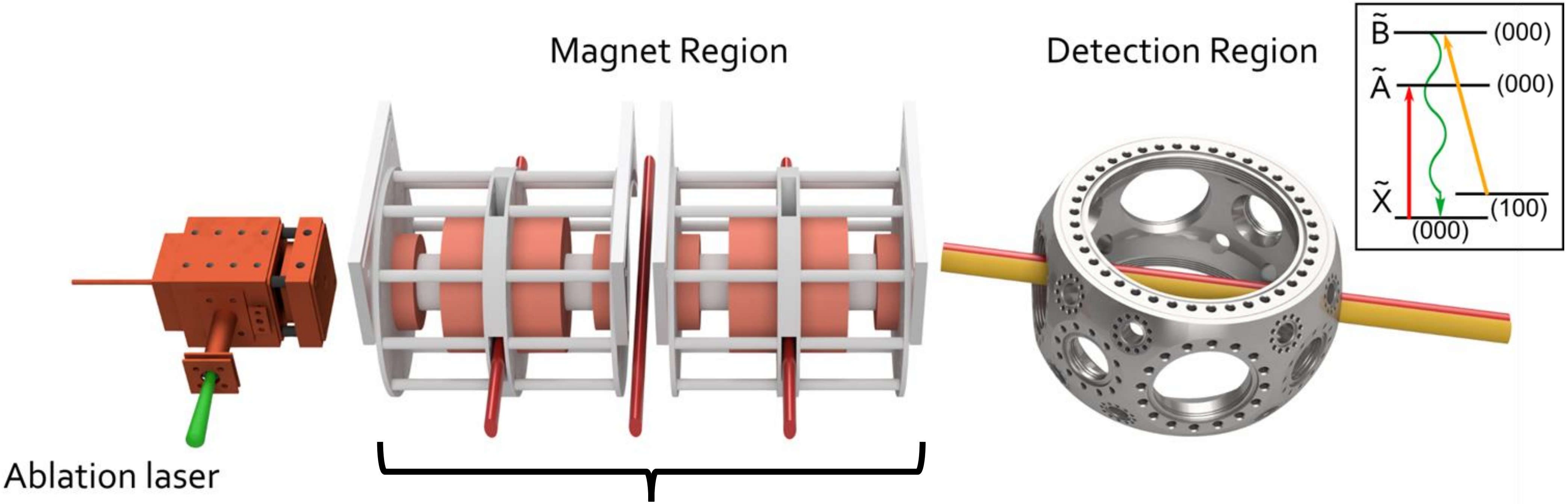
Deceleration using Zeeman effect

- Photons don't carry a lot of momentum, but magnetic fields can cause huge energy shifts
- Let's leverage a Sisyphus-type effect for *deceleration*
- Previously used for directly loading a 4 T deep trap, but that's not a good environment for an EDM experiment



Similar to trapping technique: Lu, …, Doyle, Phys. Rev. Lett. 112, 113006 (2014)
See also: Fitch and Tarbutt, ChemPhysChem 17, 3609-3623 (2016)

Experimental Apparatus



Why superconducting?

More energy removal per stage (bigger B)

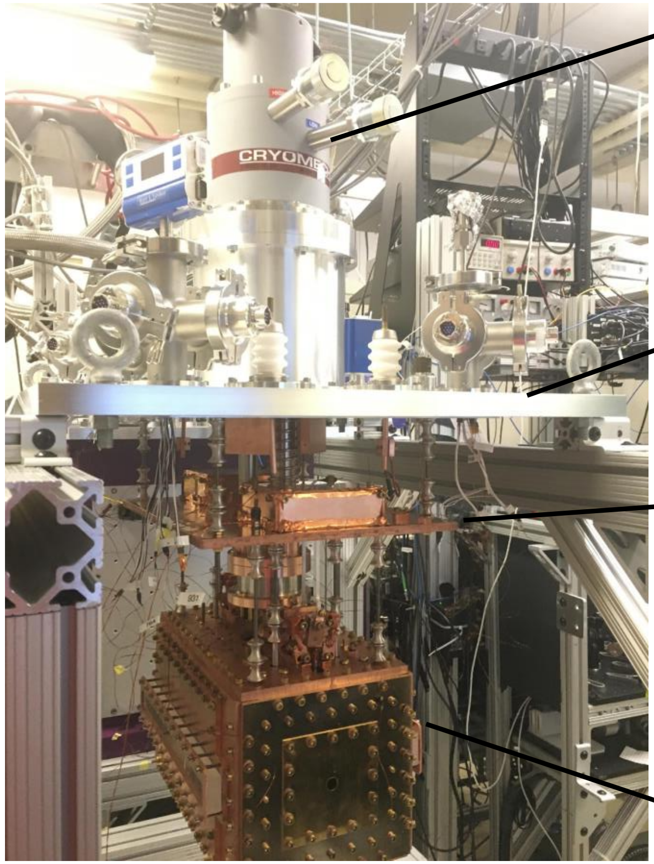
Transverse optical access (“clean” transitions)

Larger bore, excellent vacuum (high acceptance, low loss)

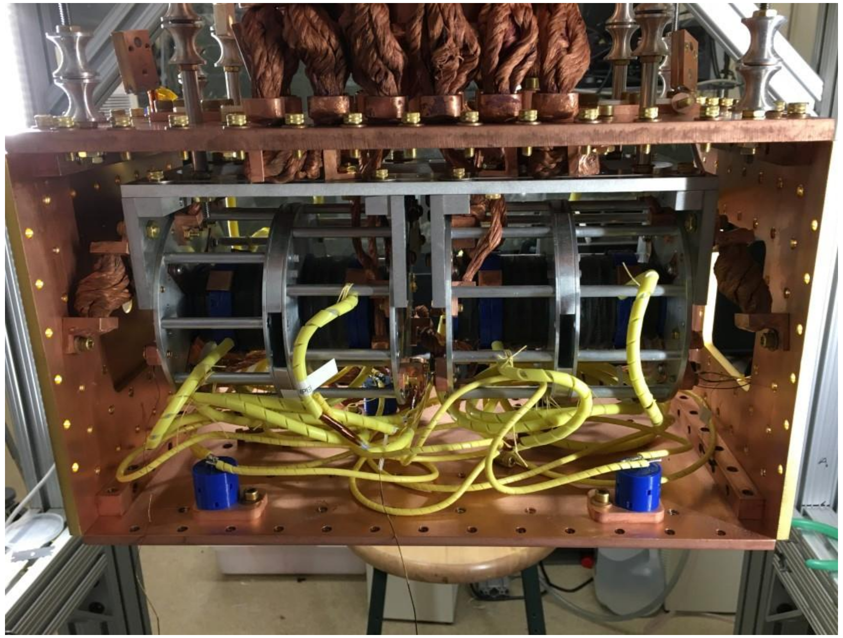
Similar to trapping techinque: Lu, …, Doyle, Phys. Rev. Lett. 112, 113006 (2014)

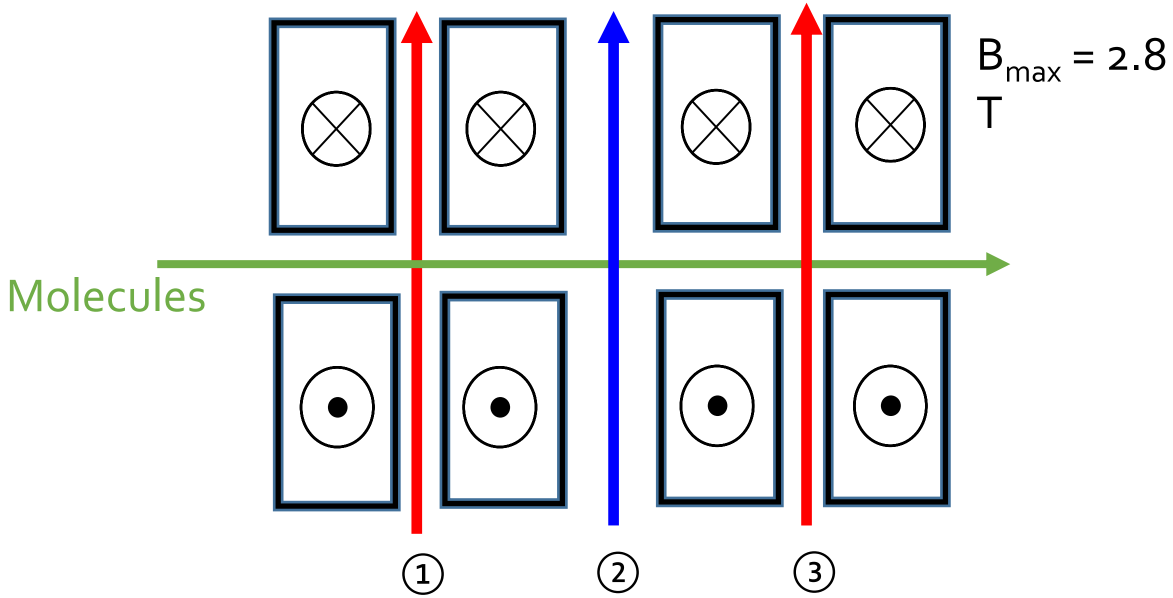
See also: Fitch and Tarbutt, ChemPhysChem 17, 3609-3623 (2016)

Cryogenic System



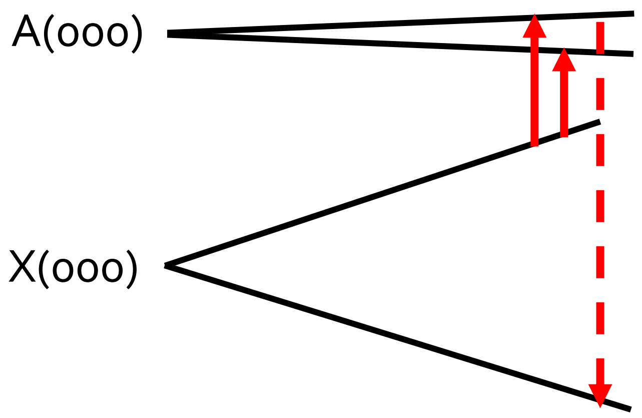
4 K shield interior



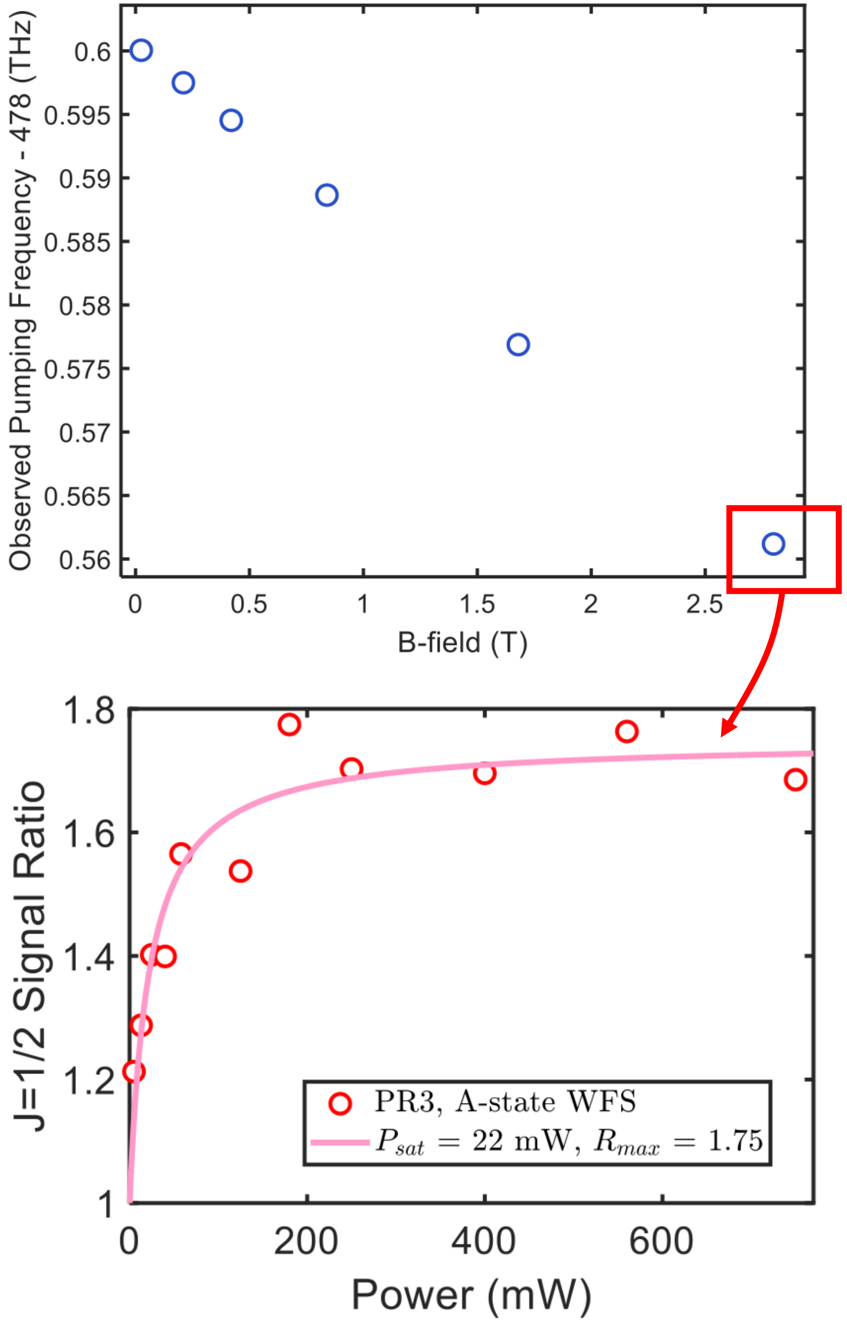


Initial Tests with CaOH

- Start with CaOH instead of YbOH
  - Simpler excited-state Zeeman structure, more mature spectroscopy
- First, verify all optical pumping transitions
  - Example: verify accumulation in SFS

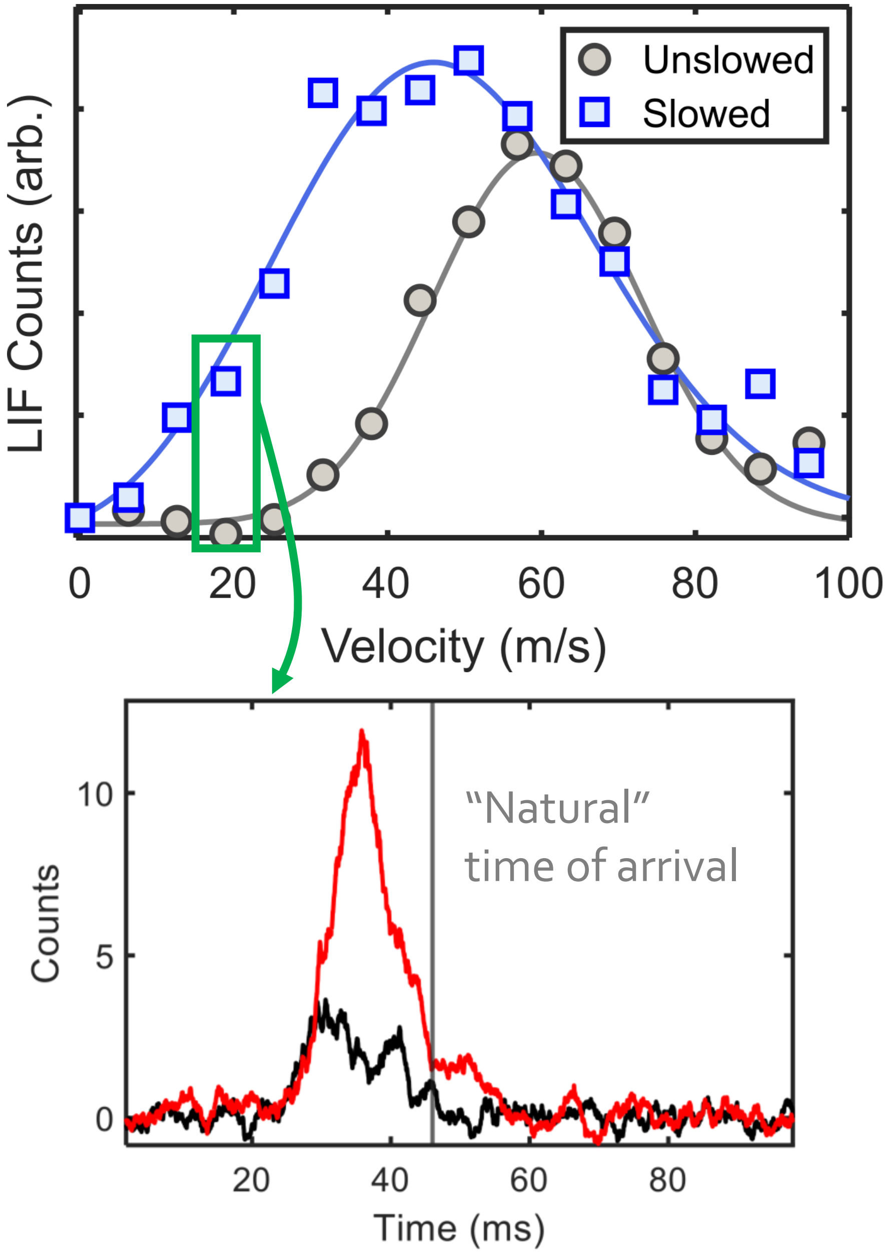


- **Key points: 1) not particularly power hungry, 2) each pumping step takes ~1.5 photons on average**



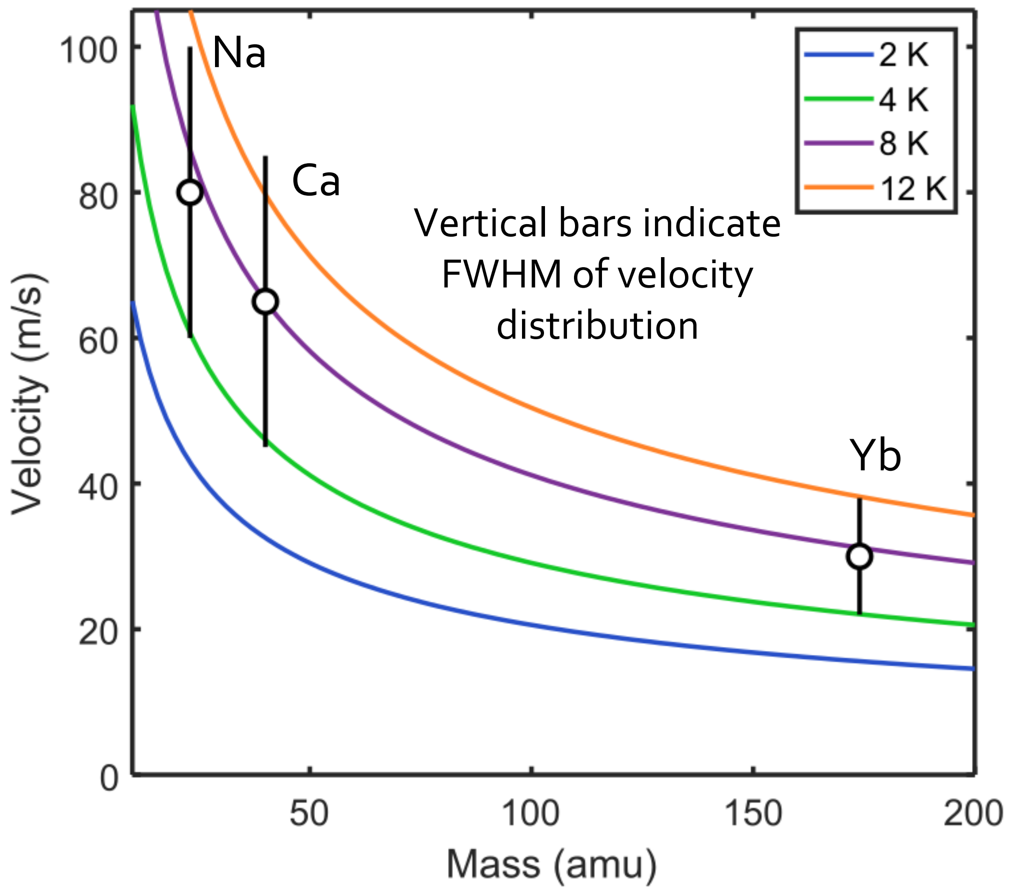
Zeeman-Sisyphus Deceleration of CaOH

- Clear enhancement of low-velocity molecules observed!
  - Overall number enhancement because under certain conditions magnets have net lensing effect
  - **Population < 20 m/s increased from 0.1% to 11% of unslowed beam!**
- Many areas to do even better:
  - Shorter beamline
  - Magnetic guiding after decelerator
  - Vacuum can be improved



Zeeman-Sisyphus for YbOH

- YbOH has complex excited-state Zeeman structure
  - Will spin flip transitions remain efficient?
  - Will B-field-induced mixing among J states lead to loss?
- Outlook appears good:
  - Required energy removal demonstrated
  - Transverse optical access likely critical
  - Lots of tricks enabled in a two-stage decelerator!

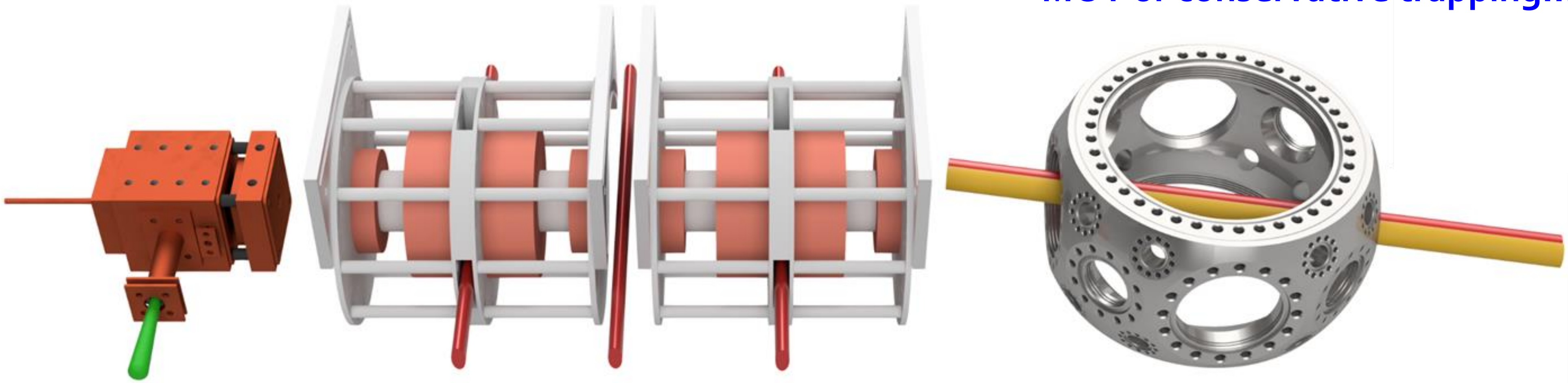


Summary and Outlook

YbOH beams < 25 m/s

Deceleration of CaOH

Slow molecules in ideal environment for MOT or conservative trapping…



- Also exploring other candidates for P,T-violation measurements, e.g. $YbOCH_3$
  - See: BLA, …, Doyle, Steimle, Phys. Rev. A 103, 022814 (2021)

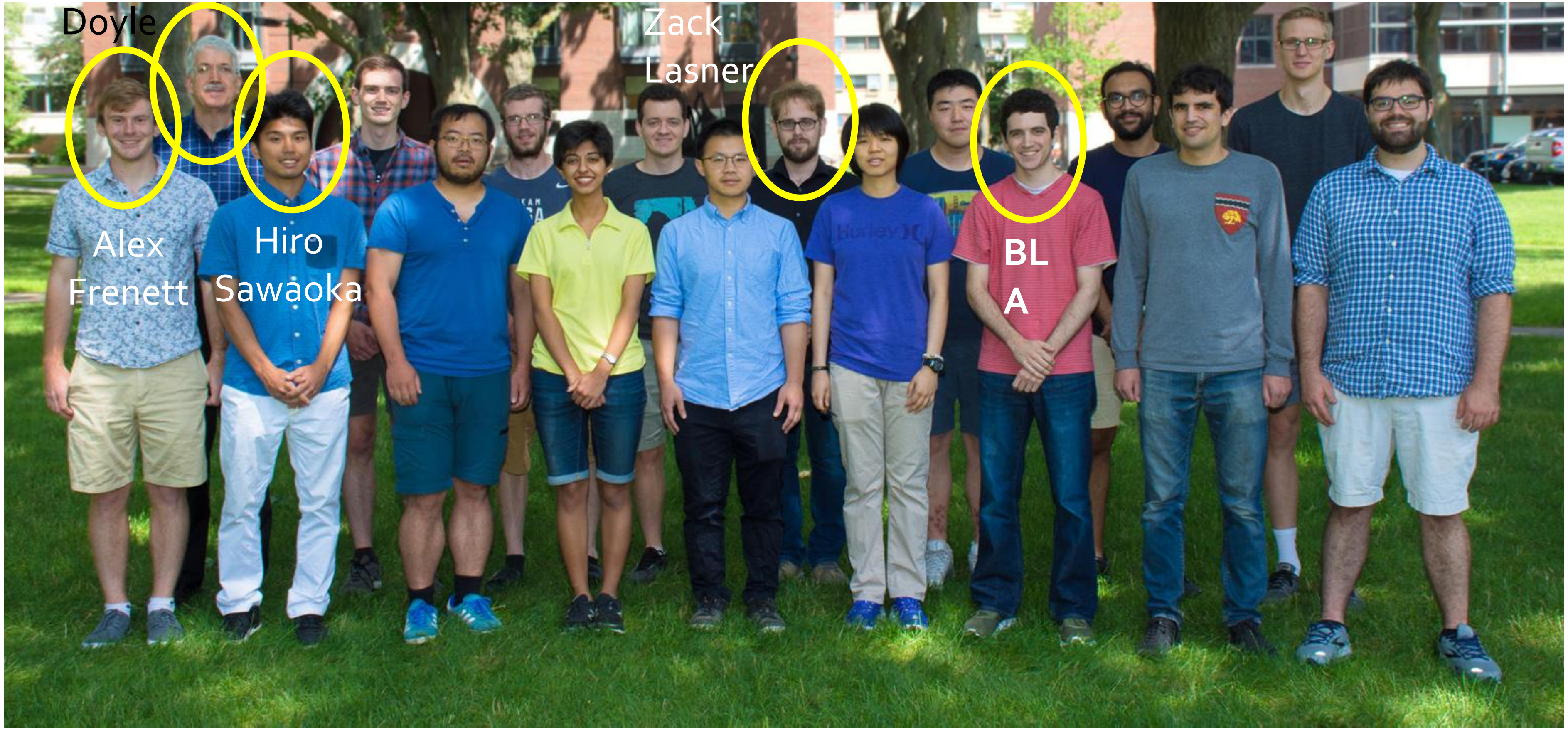
- **Key conclusion: rapid progress toward trapped molecules of interest to probing fundamental physics**

Thank you!

CUA

VE RI TAS

John M. Doyle



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