Quantum science with molecules

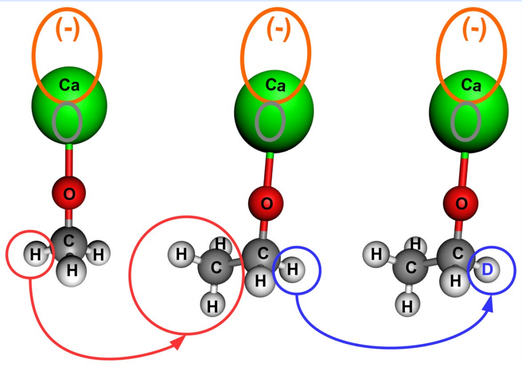
Doyle Group latest results

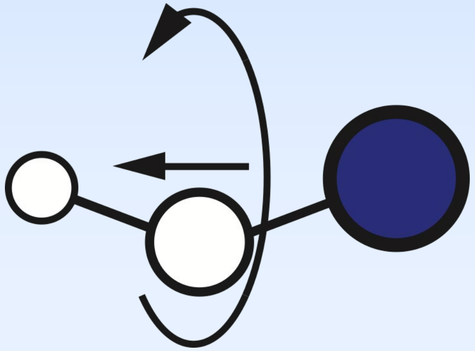
2023

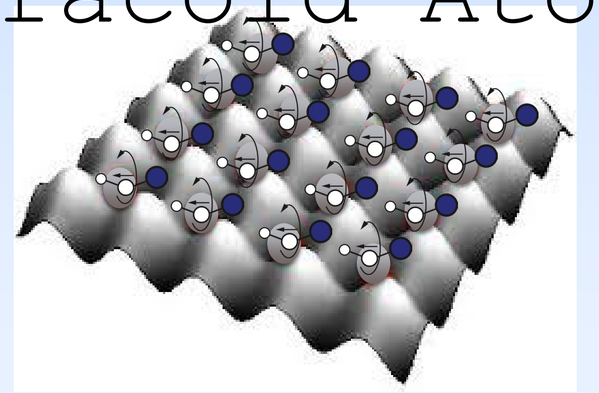
John Doyle

Harvard University Department of Physics

Harvard/MIT Center for Ultracold Atoms



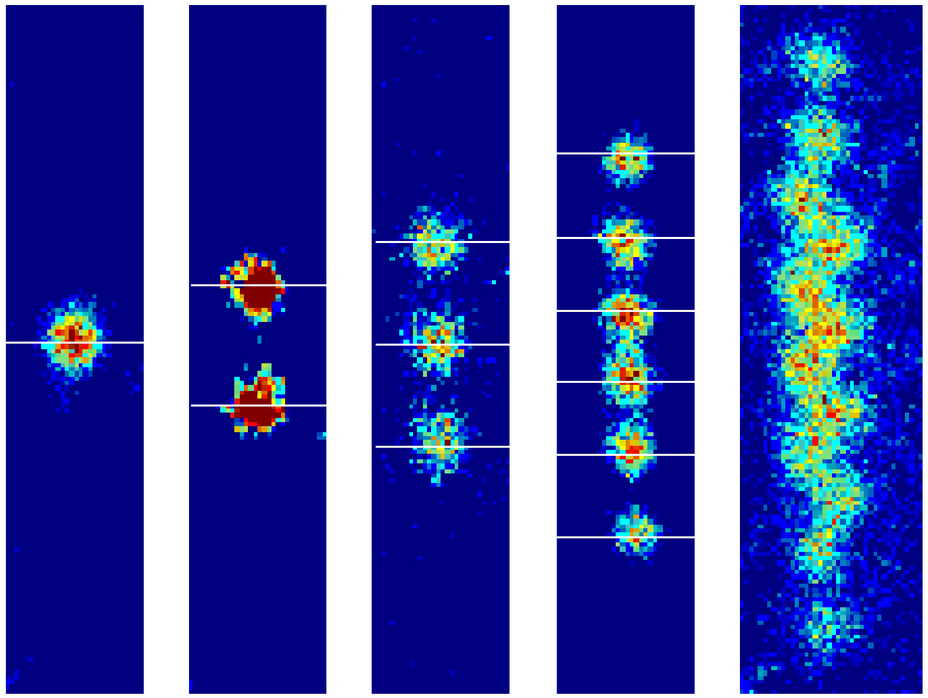




Why do Quantum Science with Molecules ??

Spectroscopy ⟷ Control ⟷ Complexity

Magnesium Ions in Ion Traps



Atoms make better quantum computers.

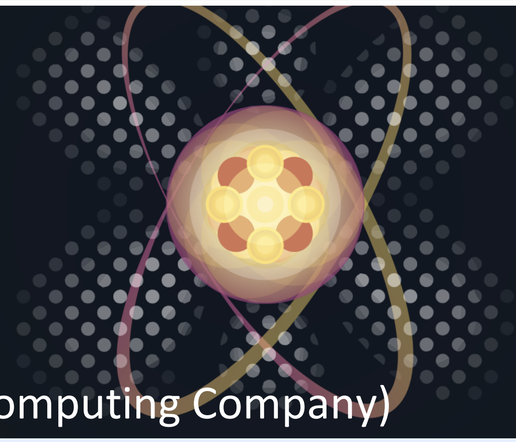
At its core, a quantum computer is a machine that uses a quantum system, like the spin of an electron, to do a very specific type of math.

This math takes advantage of the uniquely complex behavior of quantum systems, namely *entanglement* and *superposition*, to perform calculations that are fundamentally unlike the calculations ordinary computers based on classical physics can perform. Once quantum computers are powerful and stable enough, their unique computational power will solve world-changing problems that are beyond the capabilities of even the largest supercomputers.

Many quantum hardware developers use "synthetic" quantum systems for their quantum bits (*qubits* for short), like loops of supercooled superconducting wire, intentional imperfections in crystalline silicon, or other designs carefully coaxed to behave as quantum systems. At IonQ, we take a different approach. We use a naturally occurring quantum system: individual atoms.

These atoms are the heart of our quantum processing units. We trap them in 3D space, and then use lasers to do everything from initial preparation to final readout. It requires counterintuitive physics, precision optical and mechanical engineering, and fine-grained firmware control over a variety of components, but the superior results speak for themselves.

Read on to understand exactly how our trapped ion quantum cores work, and why they're the most promising platform for quantum computing in development today.



IonQ
Webpage
(Ion Quantum Computing Company)

Doyle Group Research “Strategy”

Molecular Complexity might be Harnessed to Advance Areas of AMO

1) Beyond the Standard Model physics

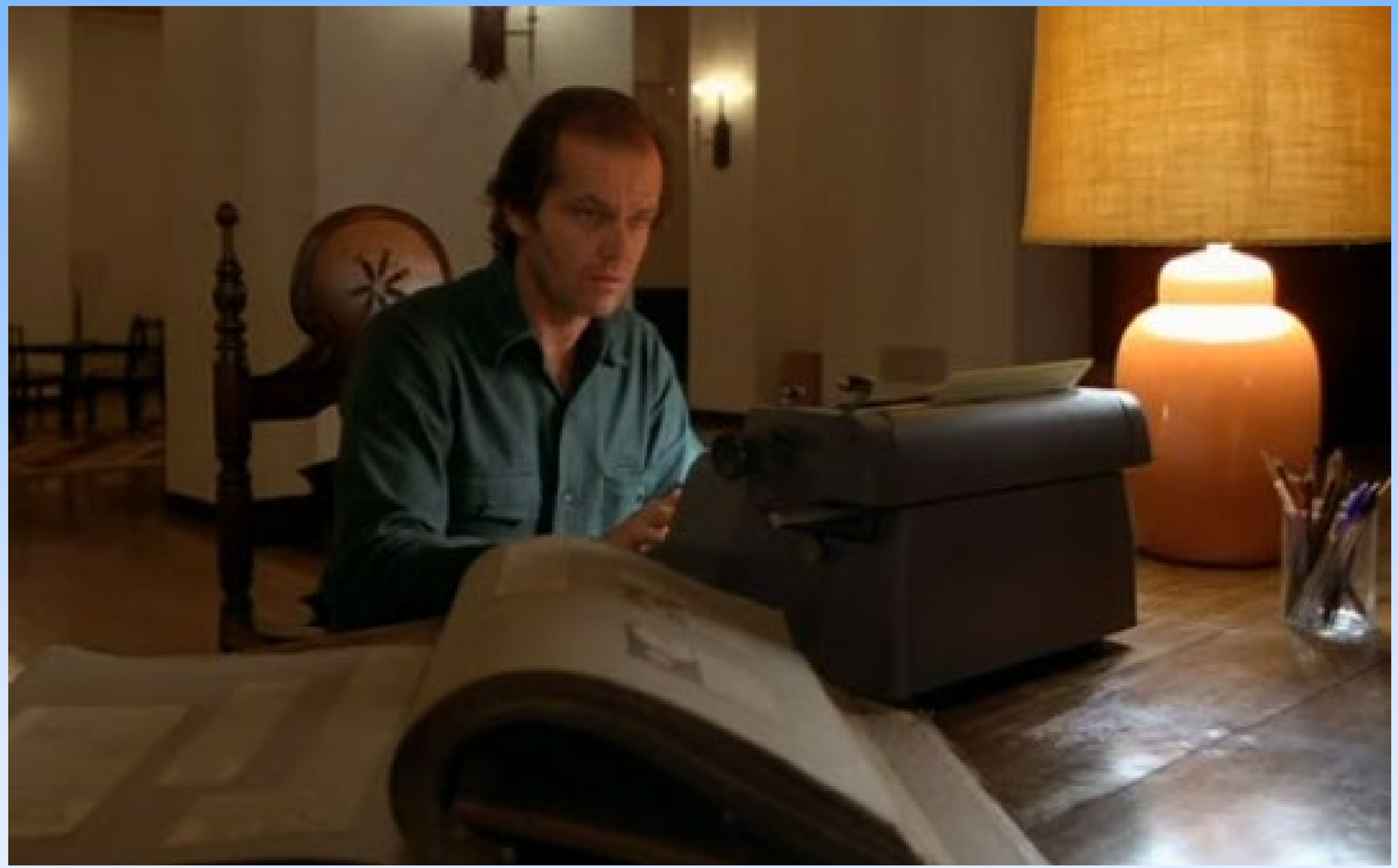
2) Collisions

3) Quantum Information

Are Molecules Scary John ??

“Well, I am glad you asked that!”

Are Molecules Scary John ??



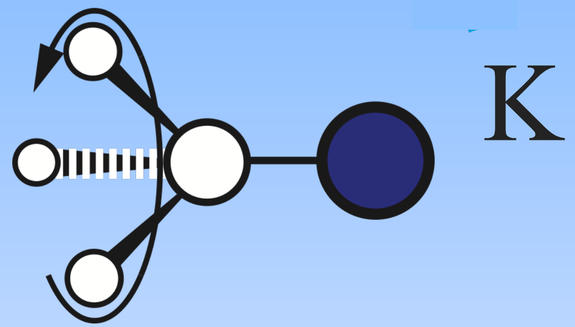
“Maybe ask me at the end of my talk. I’m very busy now. Giving a talk.
To the fine guests of this hotel.”

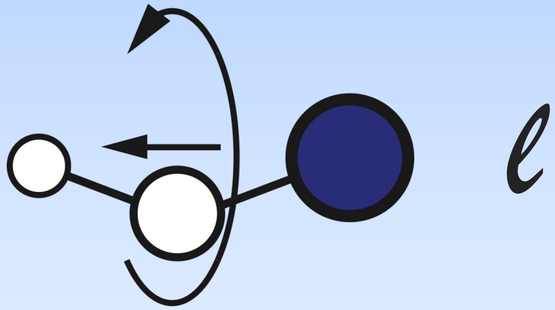
What's So Great About Molecules?

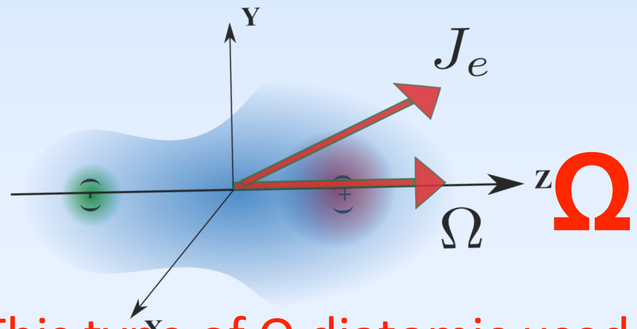
1) Complex Internal Structure - useful?

Orbital Angular Momentum Along the Internuclear Axis

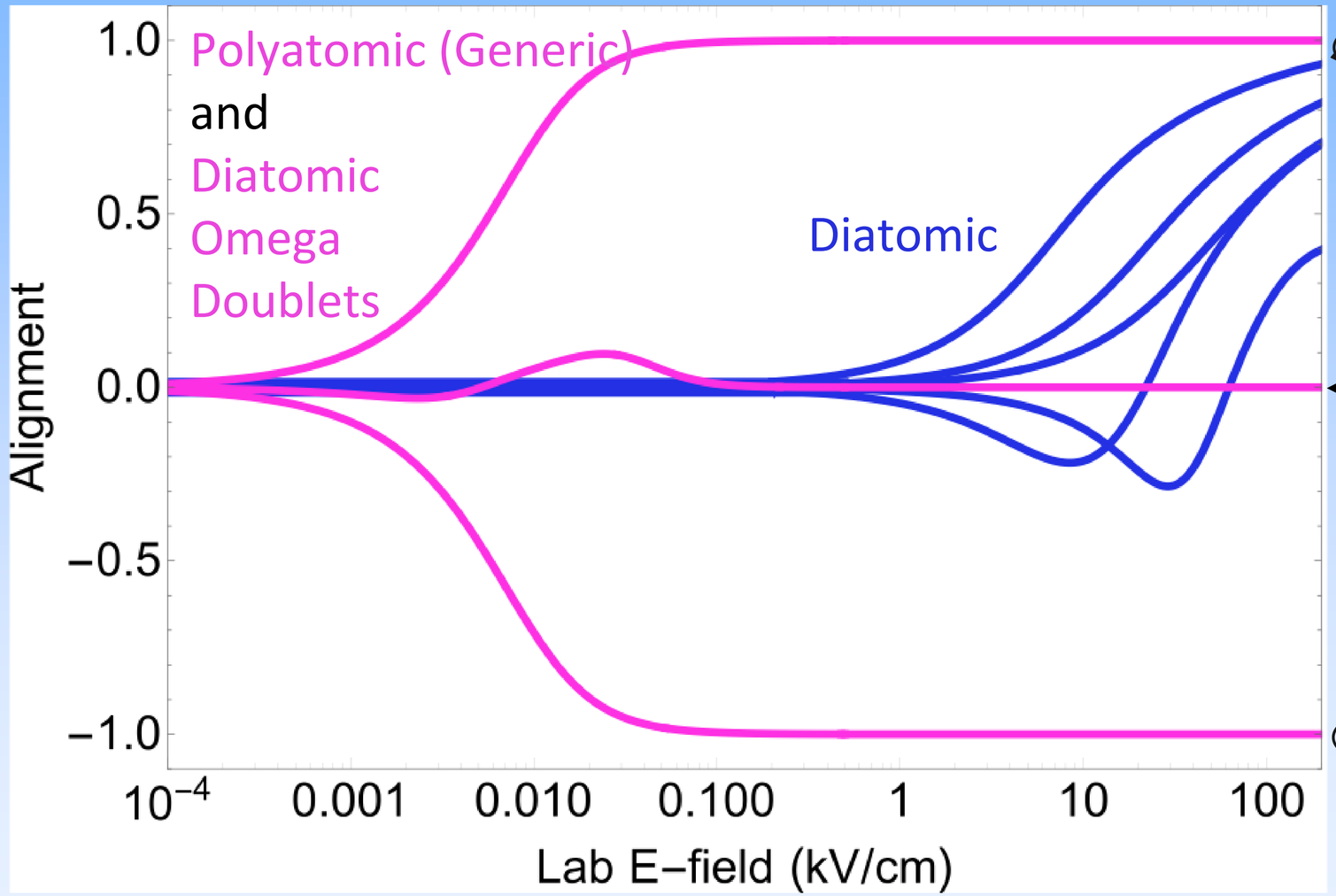
Molecular Frame Quantum Numbers

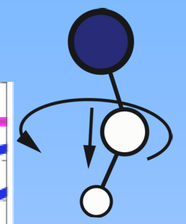




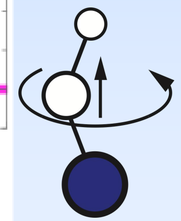


"Our Sponsor, the Parity Doublet"





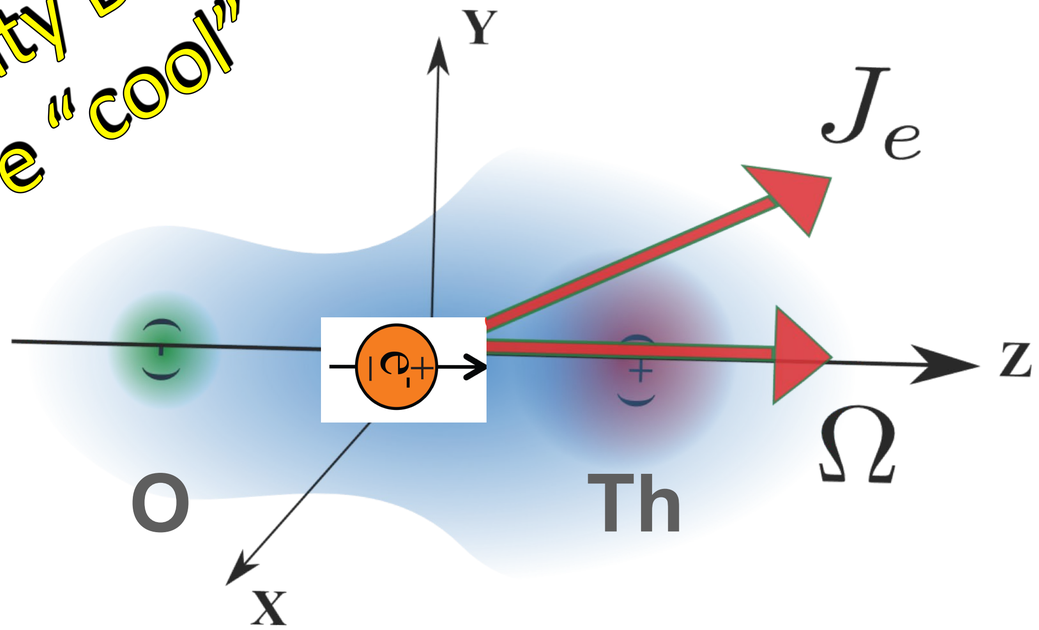
Zero Dipole Lab Frame



This type of Ω diatomic used in current best electron EDM searches

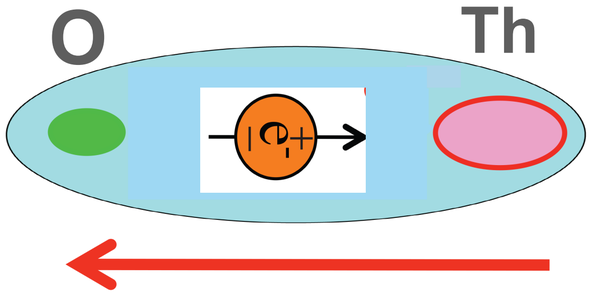
How to do an EDM experiment - example

Parity Doublets are "cool"



Large electric field
Interacts with
Purported electron EDM

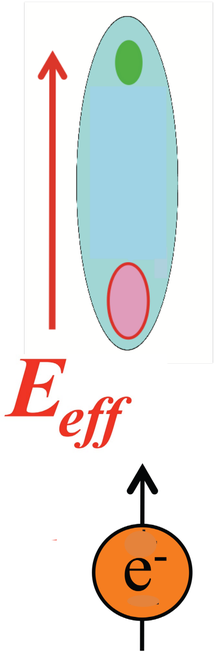
Energy Shift



E_{eff} Molecular "Effective" Electric Field

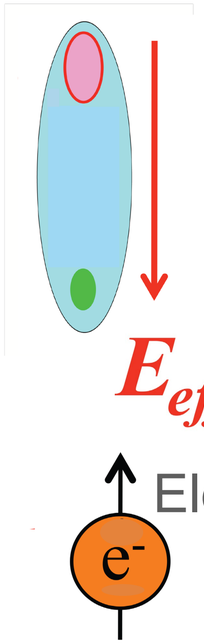
Electron (with its spin) Inside an Omega Doublet Molecule

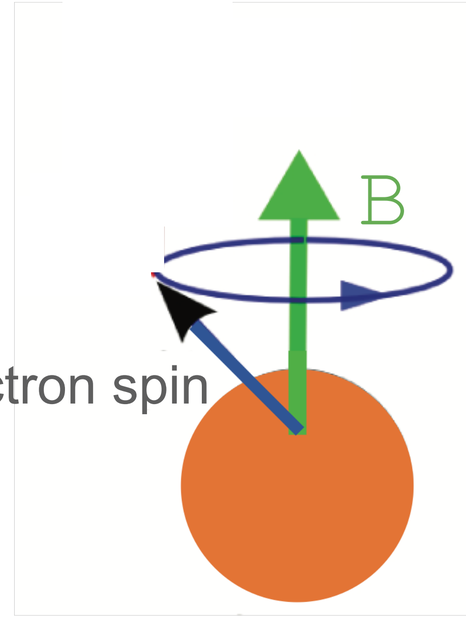
Energy



$\Omega = +1$

$\Omega = -1$





What if Electron has EDM de ?

Energy

E_{eff}

Electric Dipole of Electron d_e

$\Omega = +1$

$\Omega = -1$

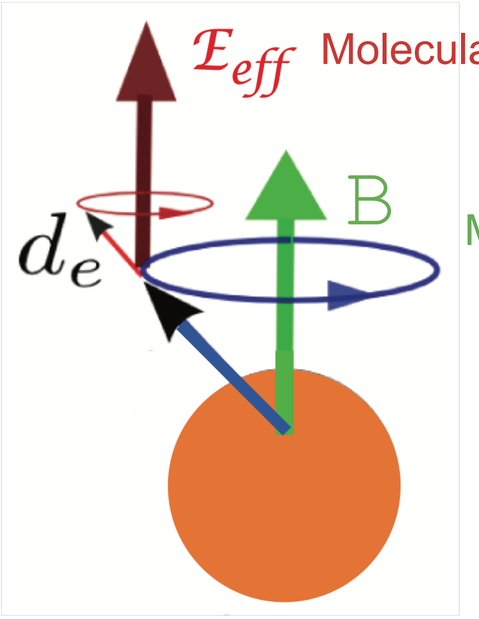
Level shift Due to H'

$H'_{de} = - d_e E_{eff}$

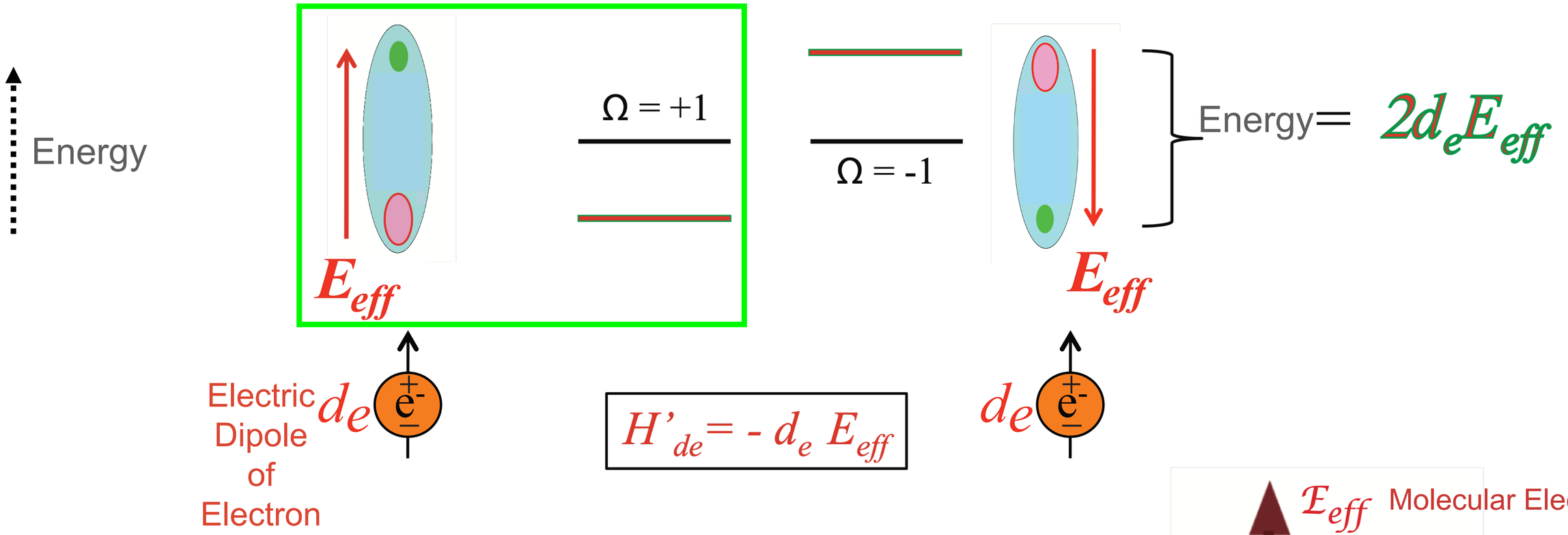
E_{eff}

d_e

Energy = $2 d_e E_{eff}$



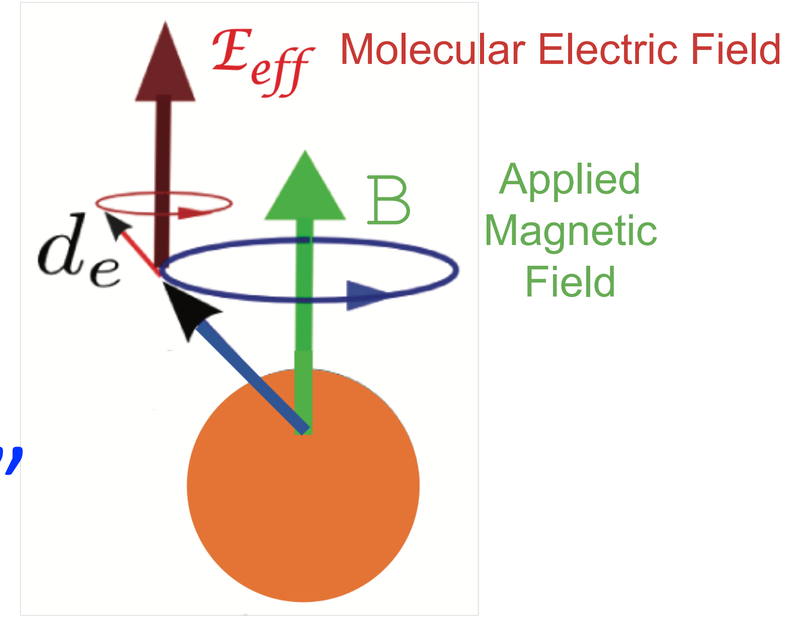
Simplest Picture of How to Measure an EDM of the Electron



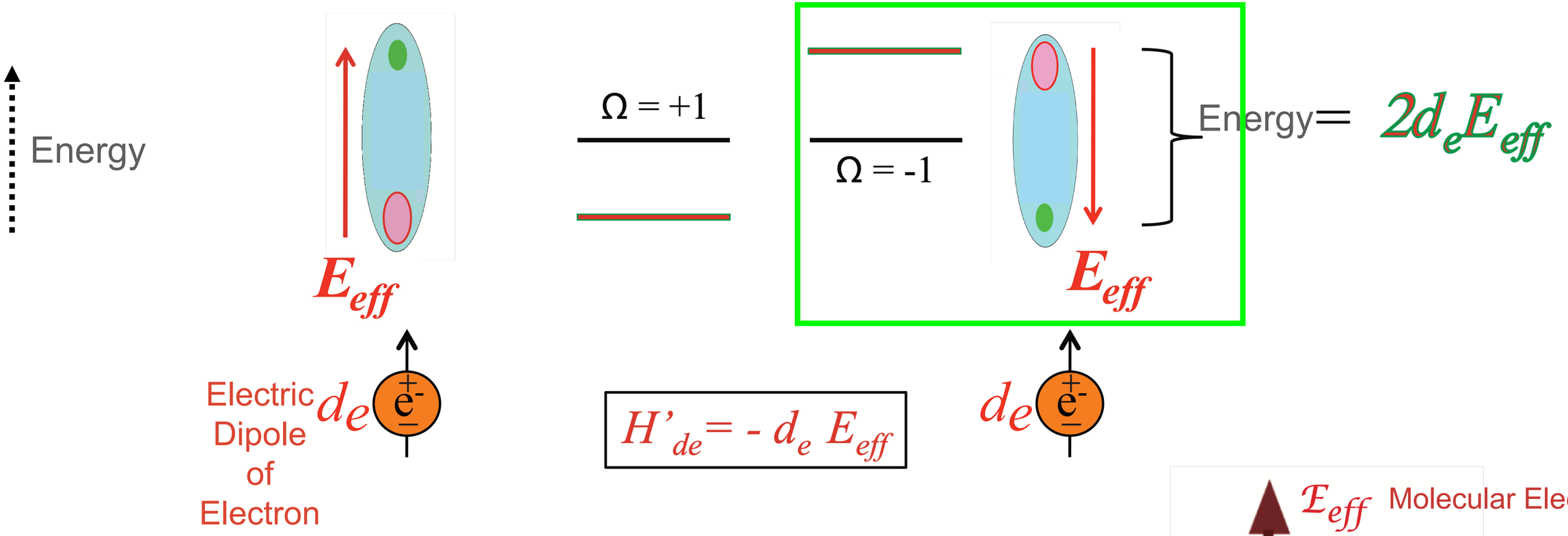
How to measure the eEDM:

Ramsey Spin Precession

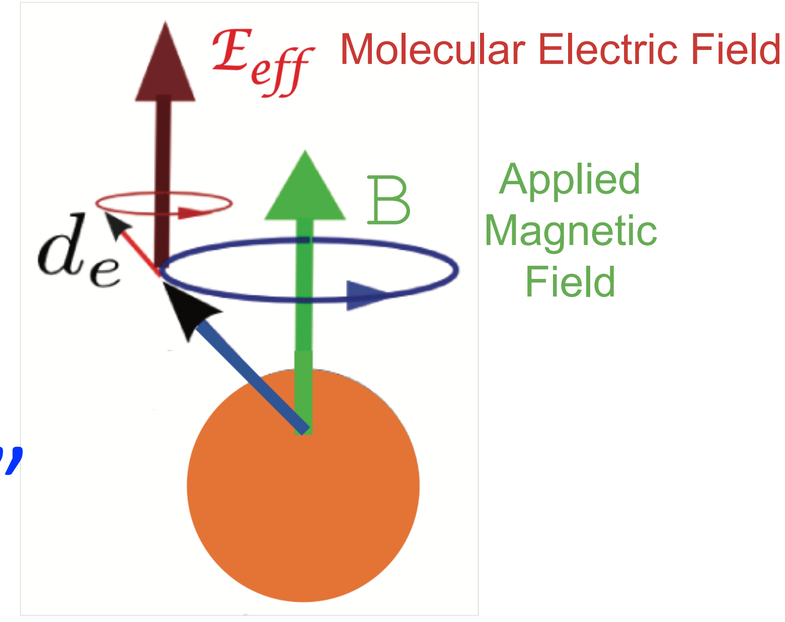
It's a "clock with a *switch ("chop")*"



Simplest Picture of How to Measure an EDM of the Electron



$$H'_{de} = -d_e E_{eff}$$

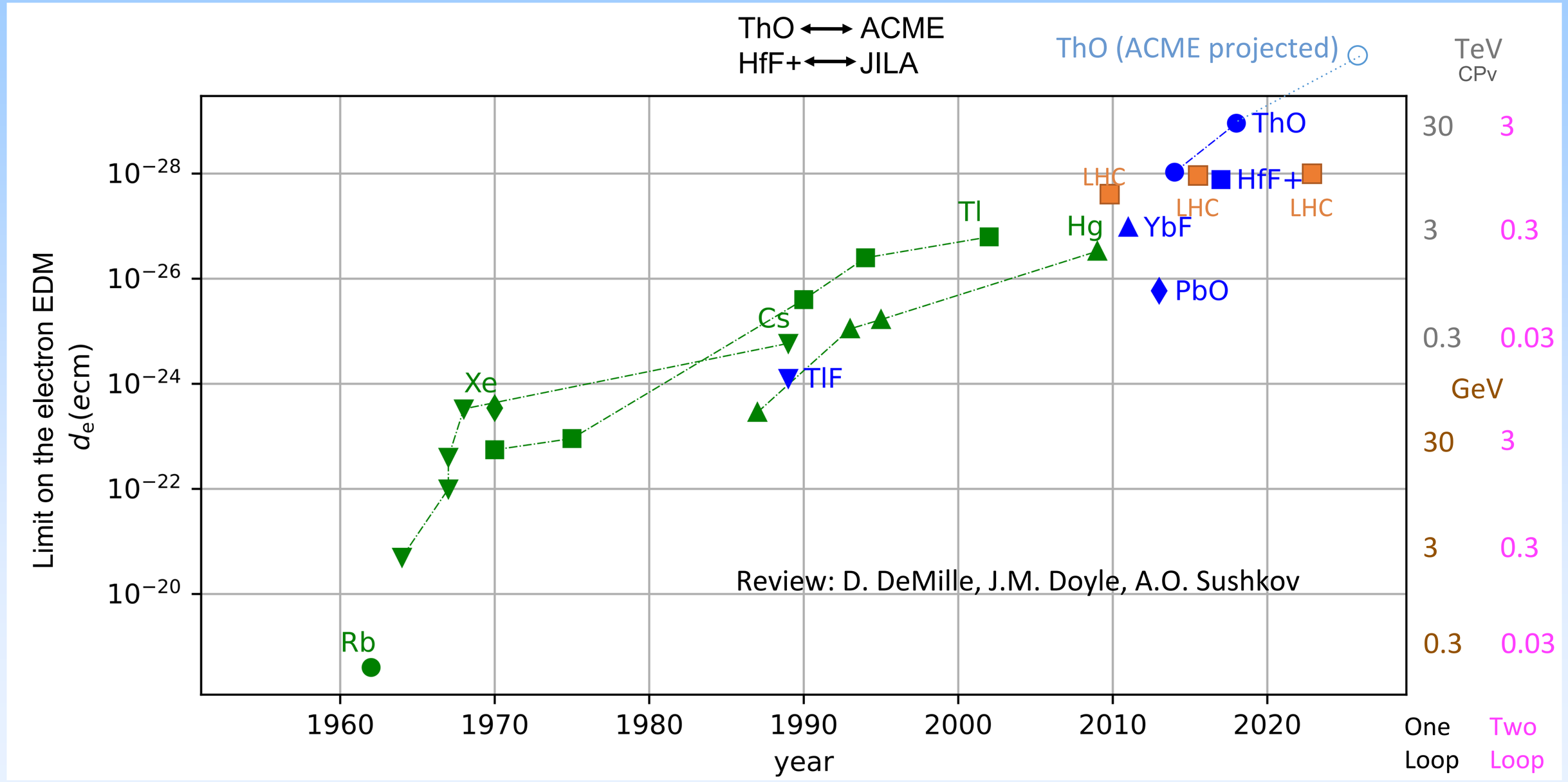


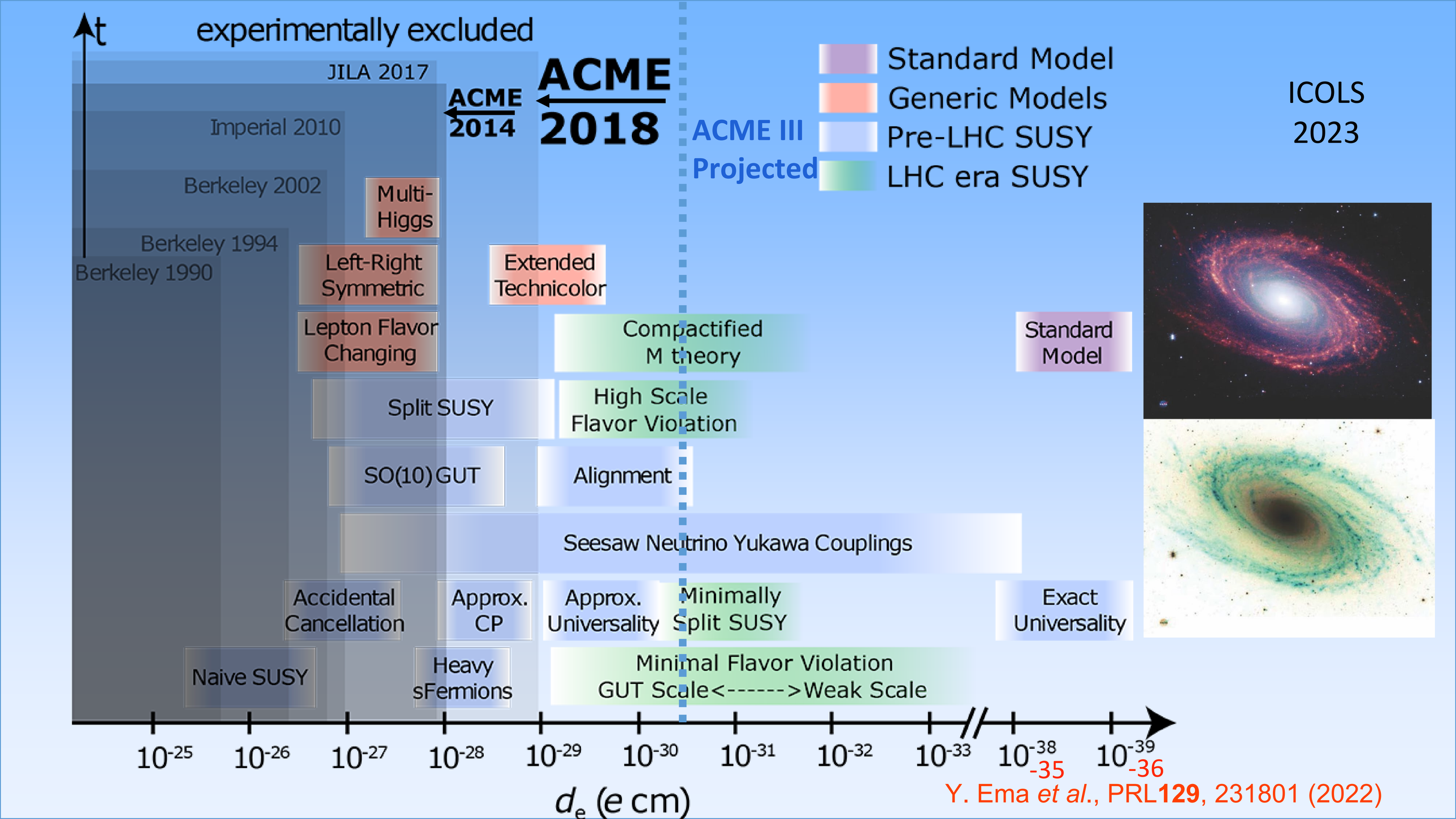
How to measure the eEDM:

Ramsey Spin Precession

It's a "clock with a *switch ("chop")*"

Electron EDM Searches as of 2018





Y. Ema *et al.*, PRL**129**, 231801 (2022)

ThO - ACME


David DeMille


Gerald Gabrielse


John Doyle


Koji Yoshimura

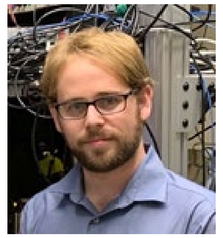

Peiran Hu

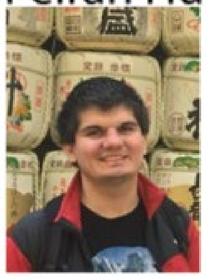

Xing Wu

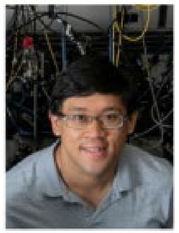

Zhen Han


Siyuan Liu


Collin Diver


Zack Lasner


Cris Panda


Daniel Ang


Cole Meisenhelder


John Mitchell


Maya Watts


Xing Fan

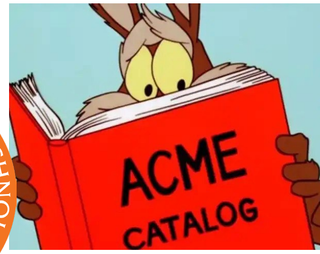

Takahiko Masuda


Satoshi Uetake


Naboru Sasao


Ayami Hiramoto


Nick Hutzler



D. Ang
D. DeMille*
J.M. Doyle*
Z. Han
A. Hiramoto
P. Hu
G. Gabrielse*
N. Hutzler
Z. Lasner
S. Liu
T. Masuda*
C. Meisenhelder
C. Panda
N. Sasao
S. Uetake
X. Wu
K. Yoshimura*

*Senior Leaders/ PIs

See Talk
Friday for
Latest eEDM News!

Jila 2023

ICOLS
2023

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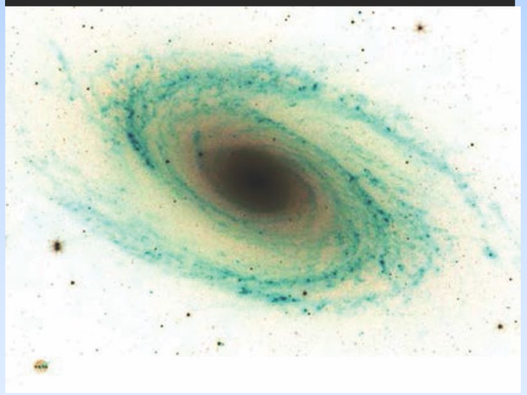


10^{-25} 10^{-26} 10^{-27} 10^{-28} 10^{-29} 10^{-30} 10^{-31} 10^{-32} 10^{-33} 10^{-38} 10^{-39}

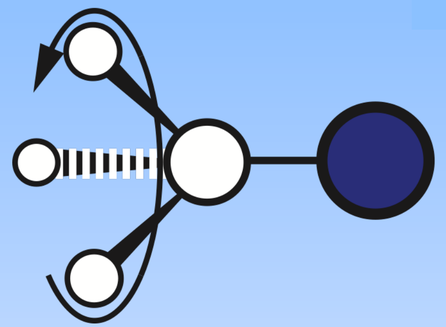
-35 -36

d_e (e cm)

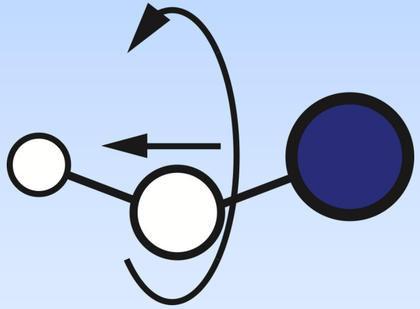
Y. Ema *et al.*, PRL**129**, 231801 (2022)

Orbital Angular Momentum Along the Internuclear Axis

Molecular Frame Quantum Numbers



K



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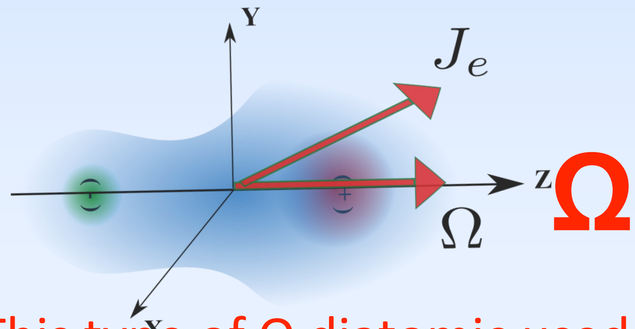
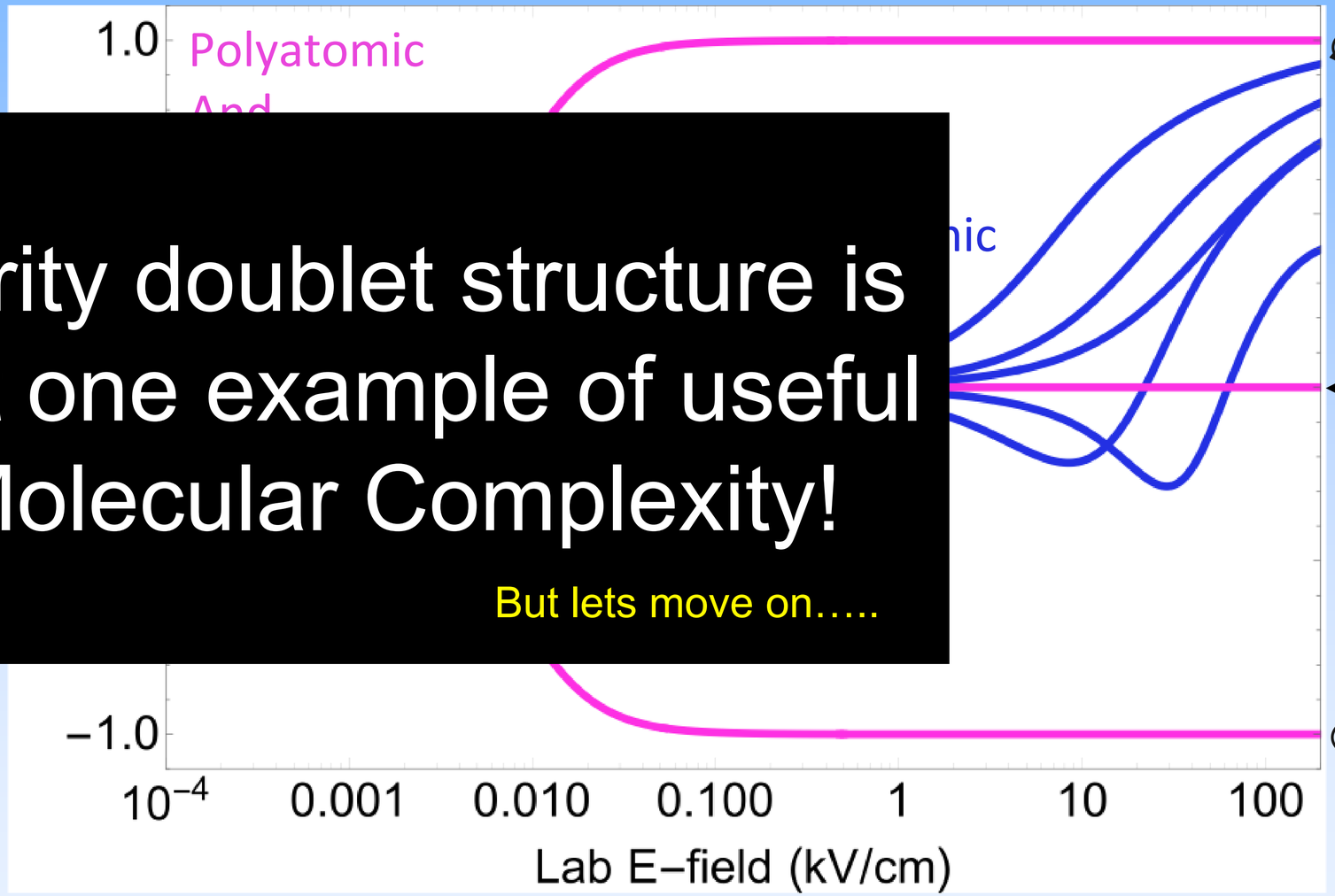


"Our Sponsor, the Parity Doublet"



Parity doublet structure is just one example of useful Molecular Complexity!

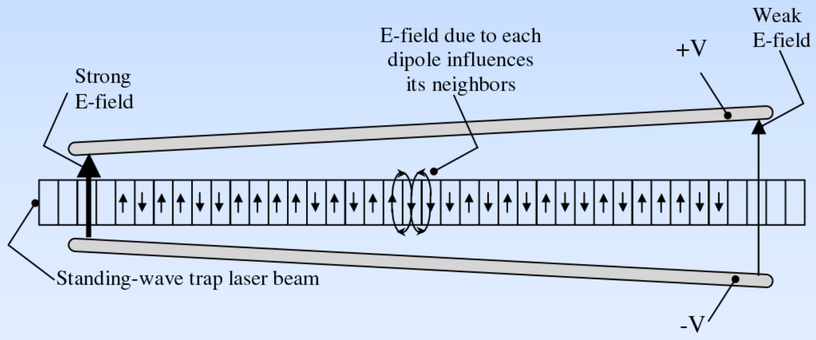
But lets move on…..

This type of Ω diatomic used in current best electron EDM searches

What's So Great About Molecules?

1) Complex Internal Structure - useful?
2) External Electric Dipole Moment

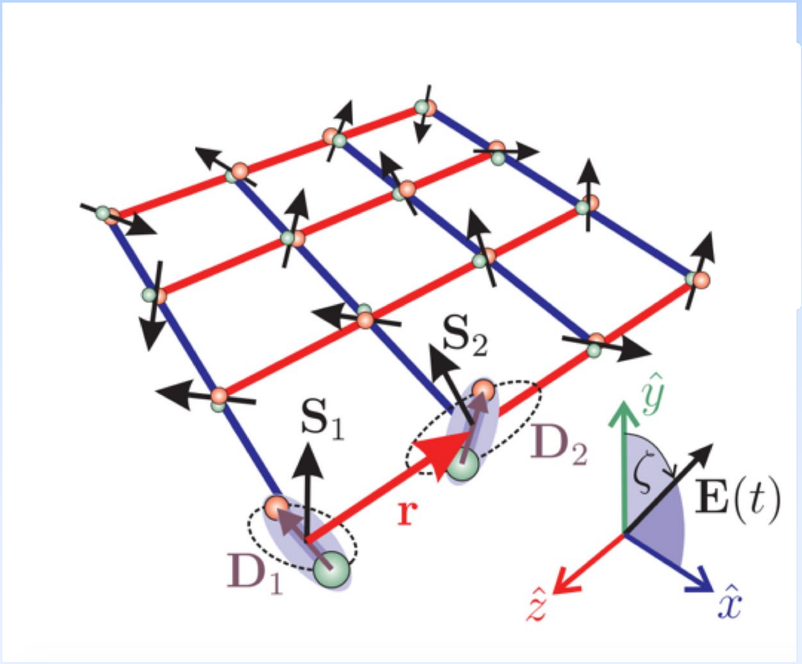
brought to you by the magic of chemical bonding (i.e. quantum mechanics)



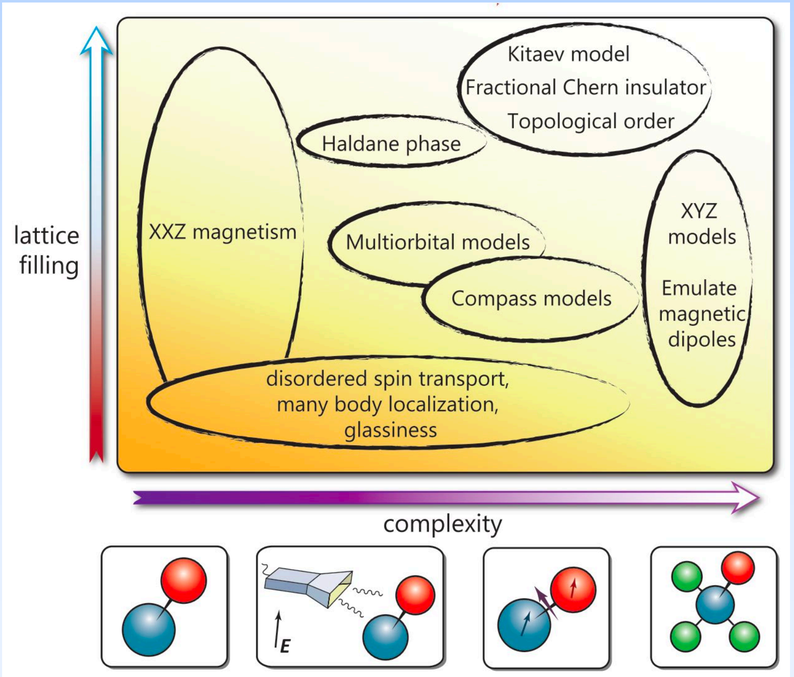
BER 6 PHYSICAL REVIEW LETTERS 11 FEBRUARY 2002

Quantum Computation with Trapped Polar Molecules

D. DeMille



Micheli, Brennan, Zoller, Nature Physics 2006



Wall, Hazzard, Rey, Atomic to Mesoscale book (2015)

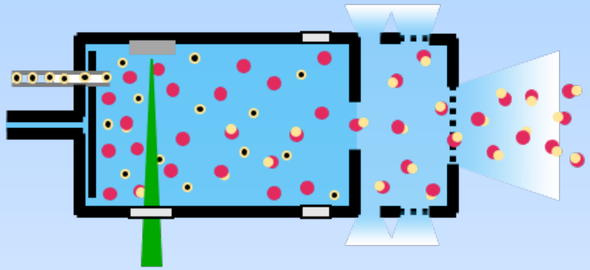
Doyle Group Research “Strategy”

Molecular Complexity might be Harnessed but….

Bringing molecules into the Cold or Ultracold Regime
is
Necessary
or
Advantageous

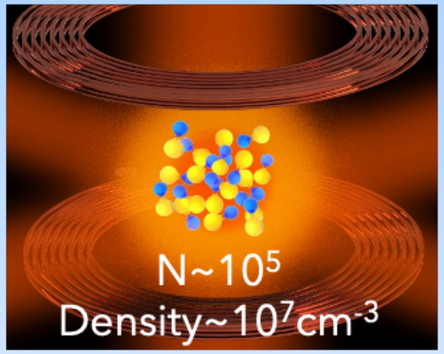
Our Laser Cooling Path to Ultracold Molecules

Molecular Source
(Buffer gas cooling)

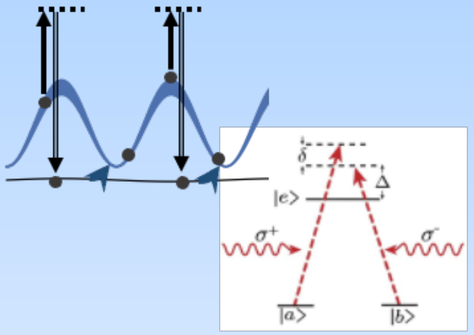


Laser Slowing

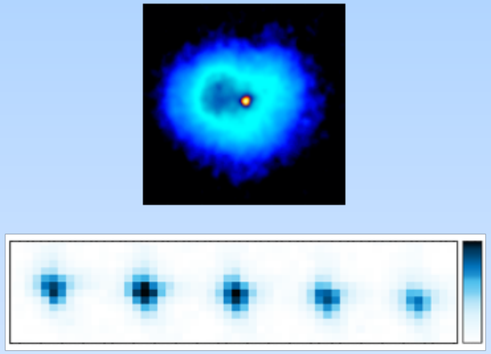
DC/RF MOT



Sub - Doppler Cooling



Optical Trapping



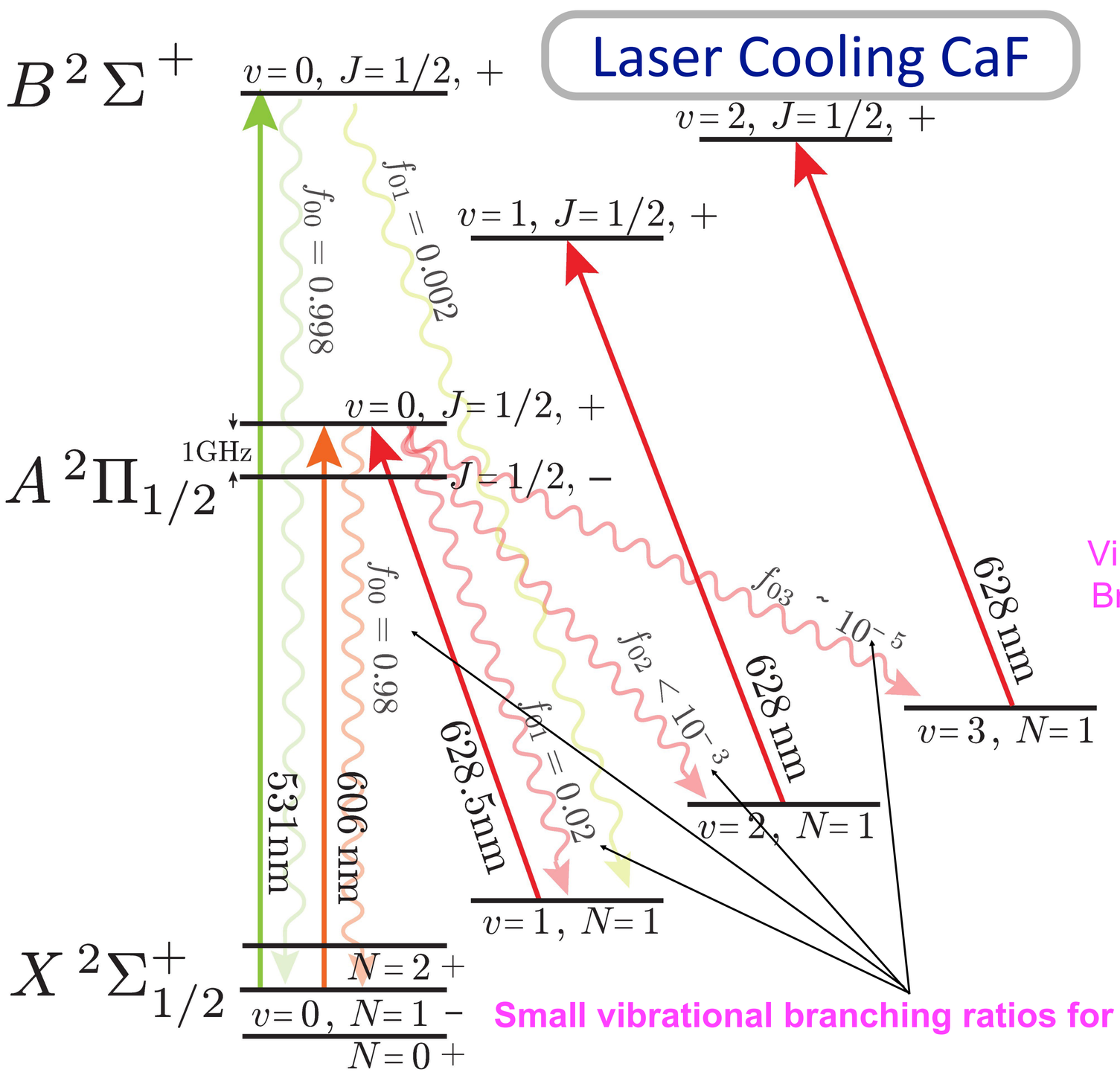
Temperature

>1000 K | 2 K | 300 μK | 5 μK

Phase Space Density

10^{-14} | 10^{-11} | 10^{-8} | 10^{-6} | 10^{-4}

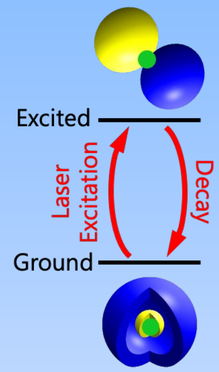
Laser Cooling CaF



Small vibrational branching ratios for CaF

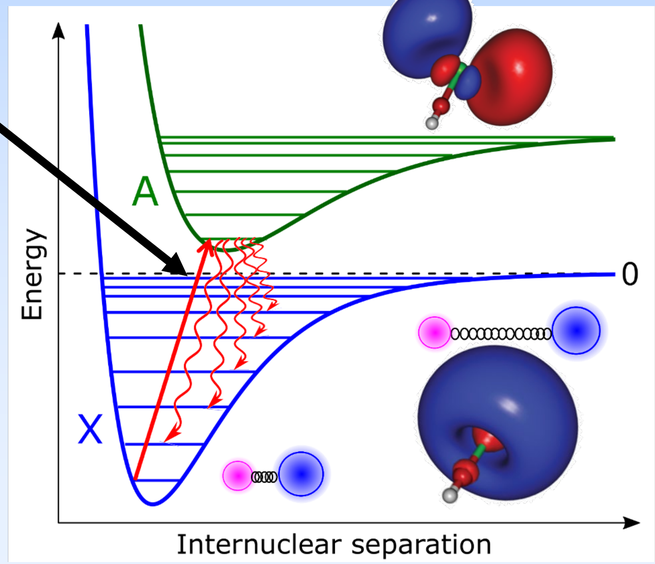
Photon Cycling - Laser Cooling

- Photons carry both linear and angular momentum
  - Cooling
  - Quantum state prep/readout

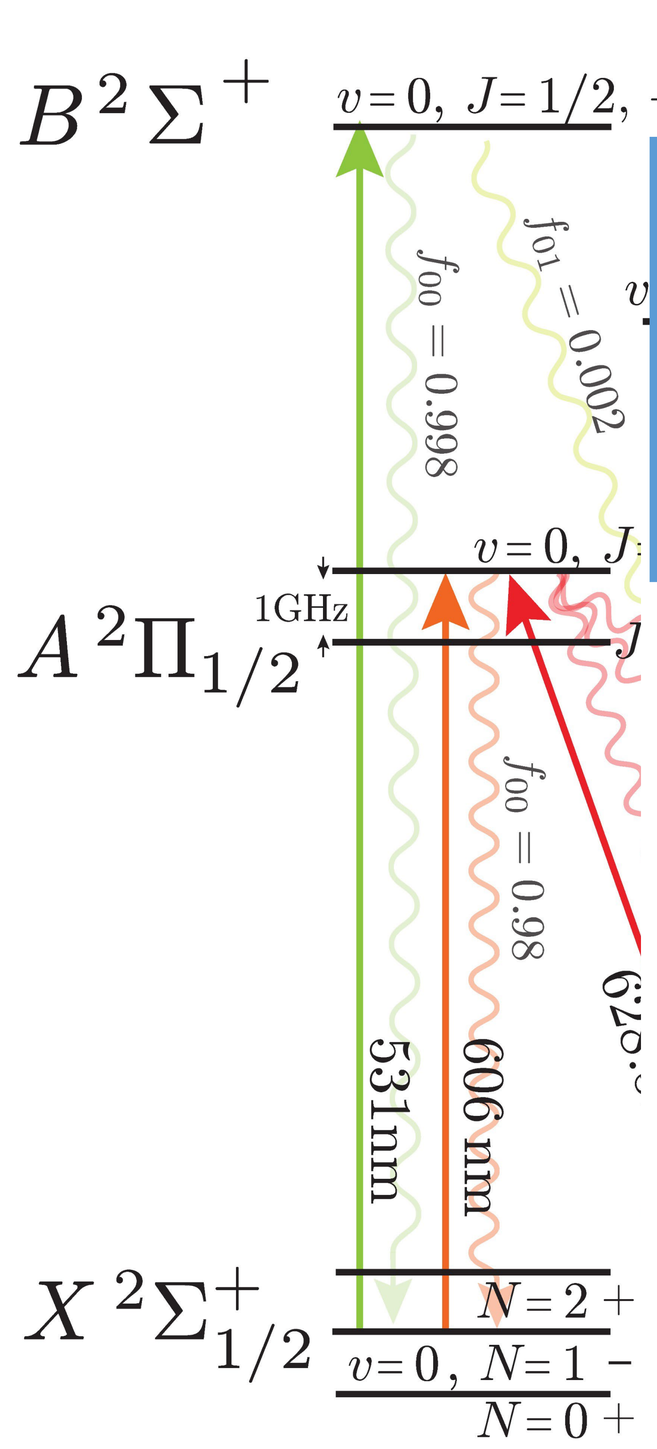


~3,000-7,000 Photon Cycles Needed for Full Laser Cooling

Vibrational Branching Ratios



Prettiest AMO Experiment Ever?



50 cm

We developed methods of laser cooling of molecules and with CaF applied to collisions and qubit behavior in optical tweezers

Dipolar spin-exchange and entanglement between molecules in an optical tweezer array, Y. Bao, S. S. Yu, L. Anderegg, E. Chae, W. Ketterle, K. Ni, JMD (2022). arXiv:2211.09780

Fast optical transport of ultracold molecules over long distances, Y. Bao, S. S. Yu, L. Anderegg, S. Burchesky, D. Gonzalez-Acevedo, E. Chae, W. Ketterle, K.-K. Ni, JMD. New J. Phys. 24 093028 (2022).

Observation of Microwave Shielding of Ultracold Molecules L. Anderegg, S. Burchesky, Y. Bao, S. Yu, T. Karman, E. Chae, K.-K. Ni, W. Ketterle, JMD. Science 373, 6556 (2021).

Rotational Coherence Times of Polar Molecules in Optical Tweezers S.Burchesky, L. Anderegg, Y. Bao, S. S. Yu, E. Chae, W. Ketterle, K.-K. Ni, JMD. Phys. Rev. Lett. 127, 123202 (2021).

Observation of Collisions between Two Ultracold Ground-State CaF Molecules L. **Cheuk**, L. Anderegg, Y. Bao, S. Burchesky, Scarlett Yu, W. Ketterle, K.-K. Ni, JMD. Phys Rev. Lett. 125, 043401 (2020).

An Optical Tweezer Array of Ultracold Molecules L. Anderegg, L. **Cheuk**, Y. Bao, S. Burchesky, W. Ketterle, K.-K. Ni, JMD. Science, 365 (2019)

Λ-Enhanced Imaging of Molecules in an Optical Trap L.**Cheuk**, L.Anderegg, B.Augenbraun, Y. Bao, S. Burchesky, W. Ketterle, JMD. Phys. Rev. Lett. 121, 083201 (2018).

Laser Cooling of Optically Trapped Molecules L. Anderegg, B. Augenbraun, Y. Bao, S. Burchesky, L. **Cheuk**, W. Ketterle, JMD. Nature Physics 14, 890-893 (2018).

Radio Frequency Magneto-Optical Trapping of CaF with High Density L. Anderegg, B. Augenbraun, E. Chae, B. Hemmerling, N. Hutzler, A. Ravi, A. Collopy, J. Ye, W. Ketterle, JMD. Phys. Rev. Lett. 119, 103201 (2017).

One-dimensional magneto-optical compression of a cold CaF molecular beam E. Chae, L. Anderegg, B. L. Augenbraun, A. Ravi, B. Hemmerling, N. R. Hutzler, A. L. Collopy, J. Ye, W. Ketterle, and JMD. New Journal of Physics 19, 033035 (2017)

Laser slowing of CaF molecules to near the capture velocity of a molecular MOT. B. Hemmerling, E. Chae, A. Ravi, L. Anderegg, G. K. Drayna, N. R. Hutzler, A. L. Collopy, J. Ye, W. Ketterle and JMD, J. Phys. B: At. Mol. Opt. Phys. 49, 174001 (2016)

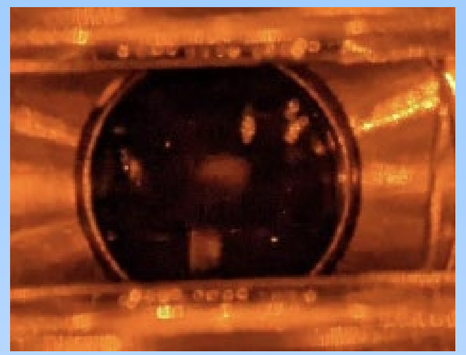
Rotational state microwave mixing for laser cooling of complex diatomic molecules. Mark Yeo, Matthew T. Hummon, Alejandra L. Collopy, Bo Yan, Boerge Hemmerling, Eunmi Chae, JMD, Jun Ye. Phys. Rev. Lett. 114, 223003 (2015).

Buffer-gas loaded magneto-optical traps for Yb, Tm, Er and Ho. B. Hemmerling, G. K. Drayna, E. Chae, A. Ravi, and JMD. New Journal of Physics 16 063070 (2014).

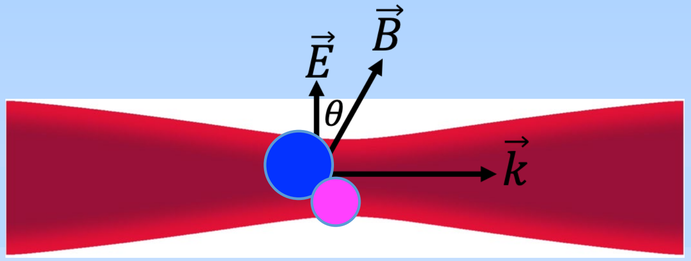
CaF - Doyle Group Results Summary through 2021

Generation 1 Apparatus….

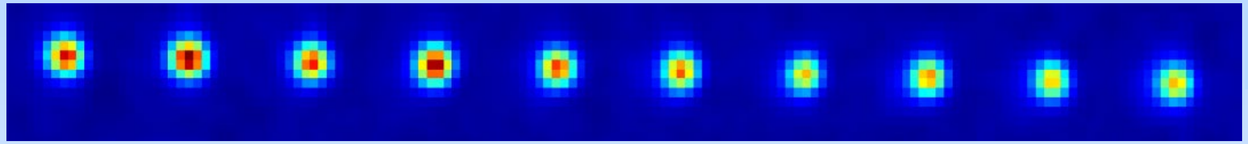
Robust MOT (mK)
and ODT (uK)



100 ms Ramsey
Coherence times
Of Rotational Qubits

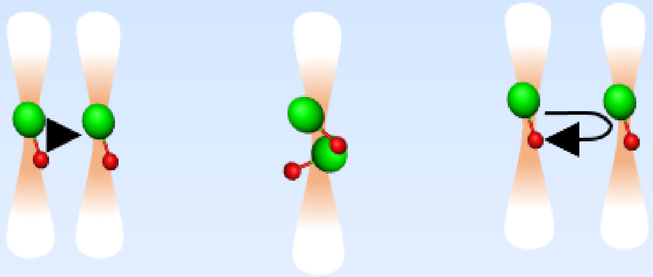


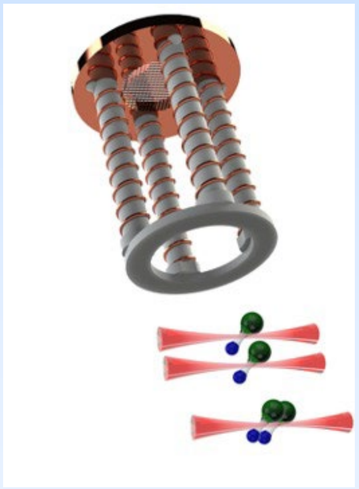
Single
Molecules in Optical
Tweezer Array



Optical Tweezer
with a  inside

Two Molecule
Collisions,
Merged Tweezers





Suppression
of inelastic collisions using Microwaves

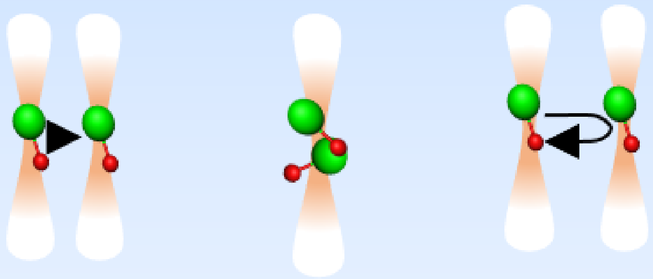
Microwave Shielding is Useful…

Next Talk!

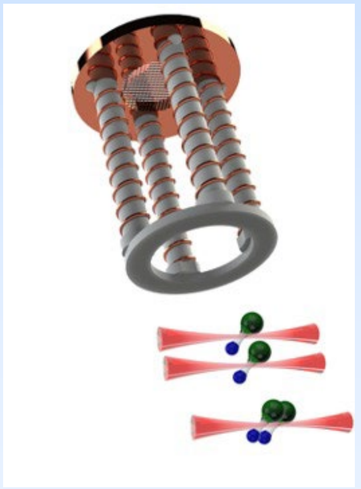
Schindewolf, A.; **Bause, R.**; **Chen, X.**; **Duda, M.**; **Karman, T.**; **Bloch, I.**; **Luo, Xinyu.**: Evaporation of microwave-shiel

with a inside

Two Molecule Collisions, Merged Tweezers

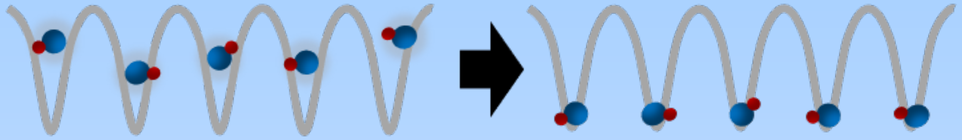


Suppression of inelastic collisions using Microwaves

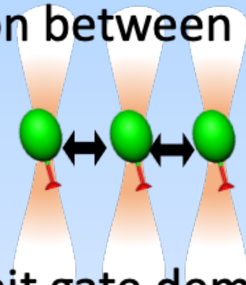


CaF - Near Term Goals from ~1 year ago

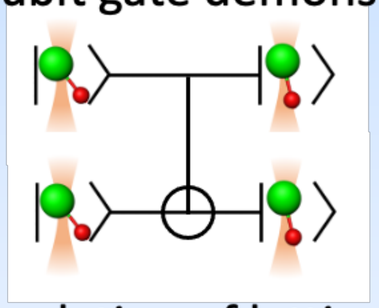
Raman sideband cooling of CaF molecules in optical tweezers



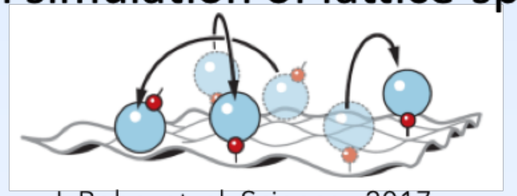
Dipolar interaction between CaF in tweezer sites



Two qubit gate demonstration



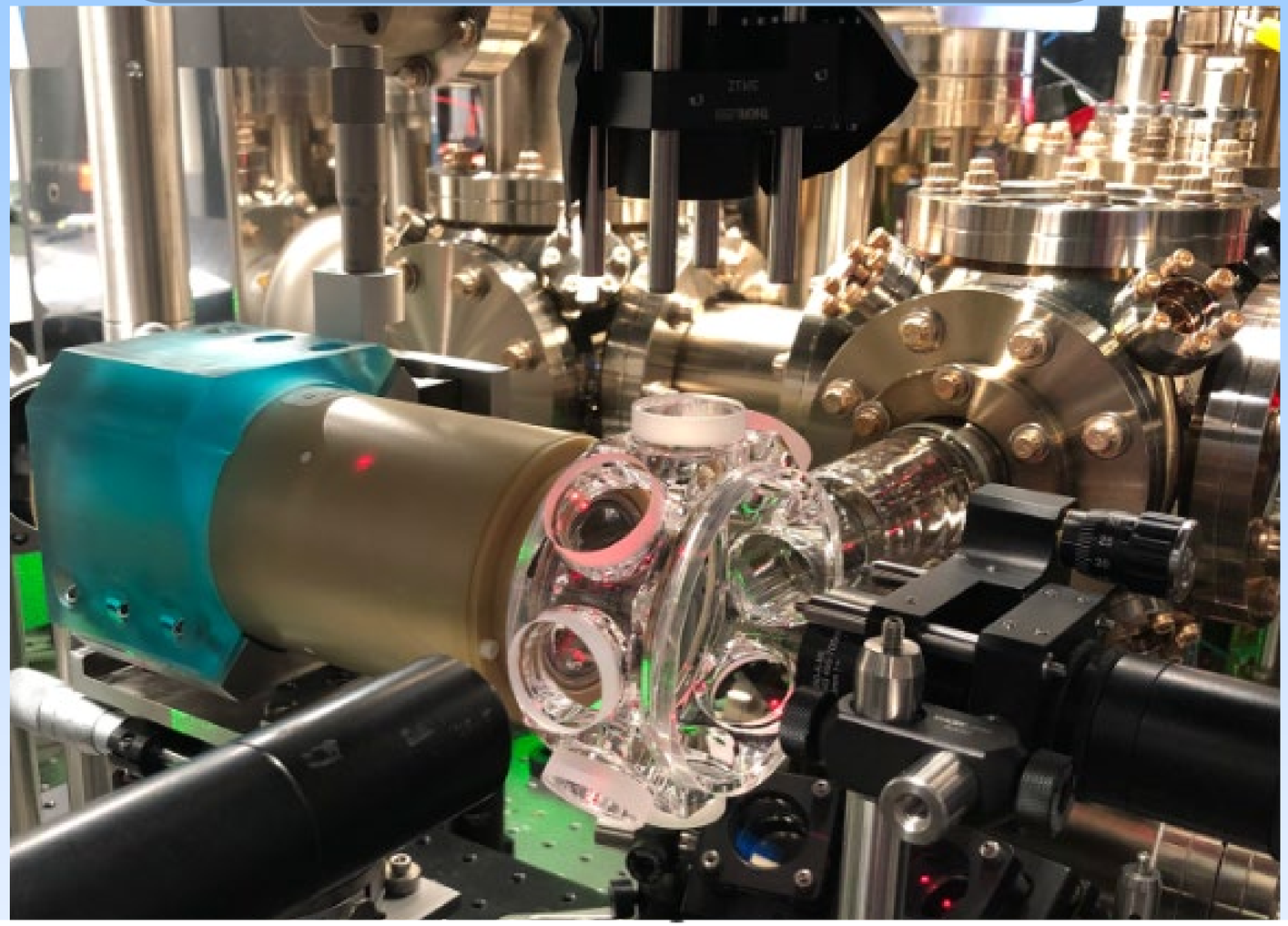
Quantum simulation of lattice-spin models



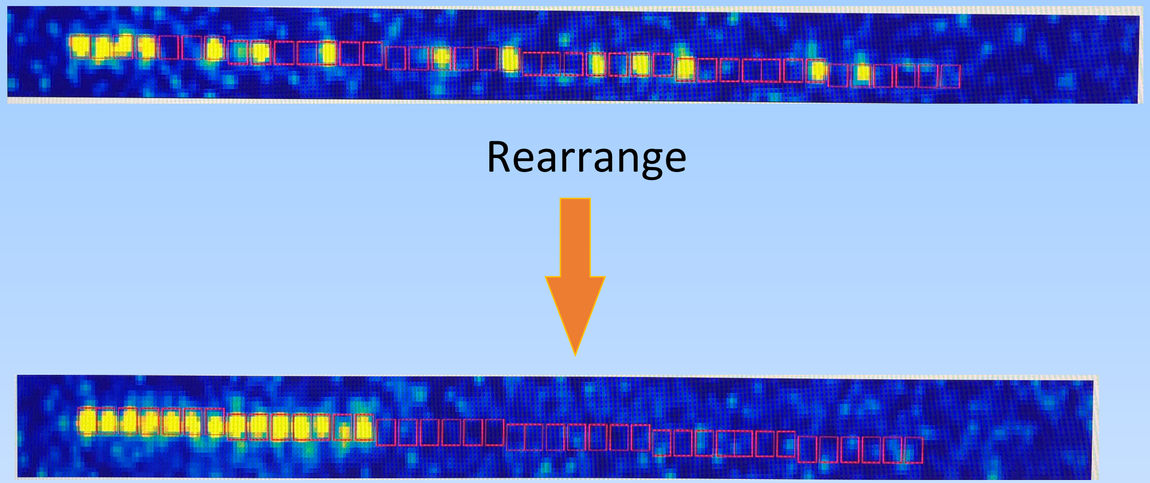
J. Bohn, et. al. Science, 2017

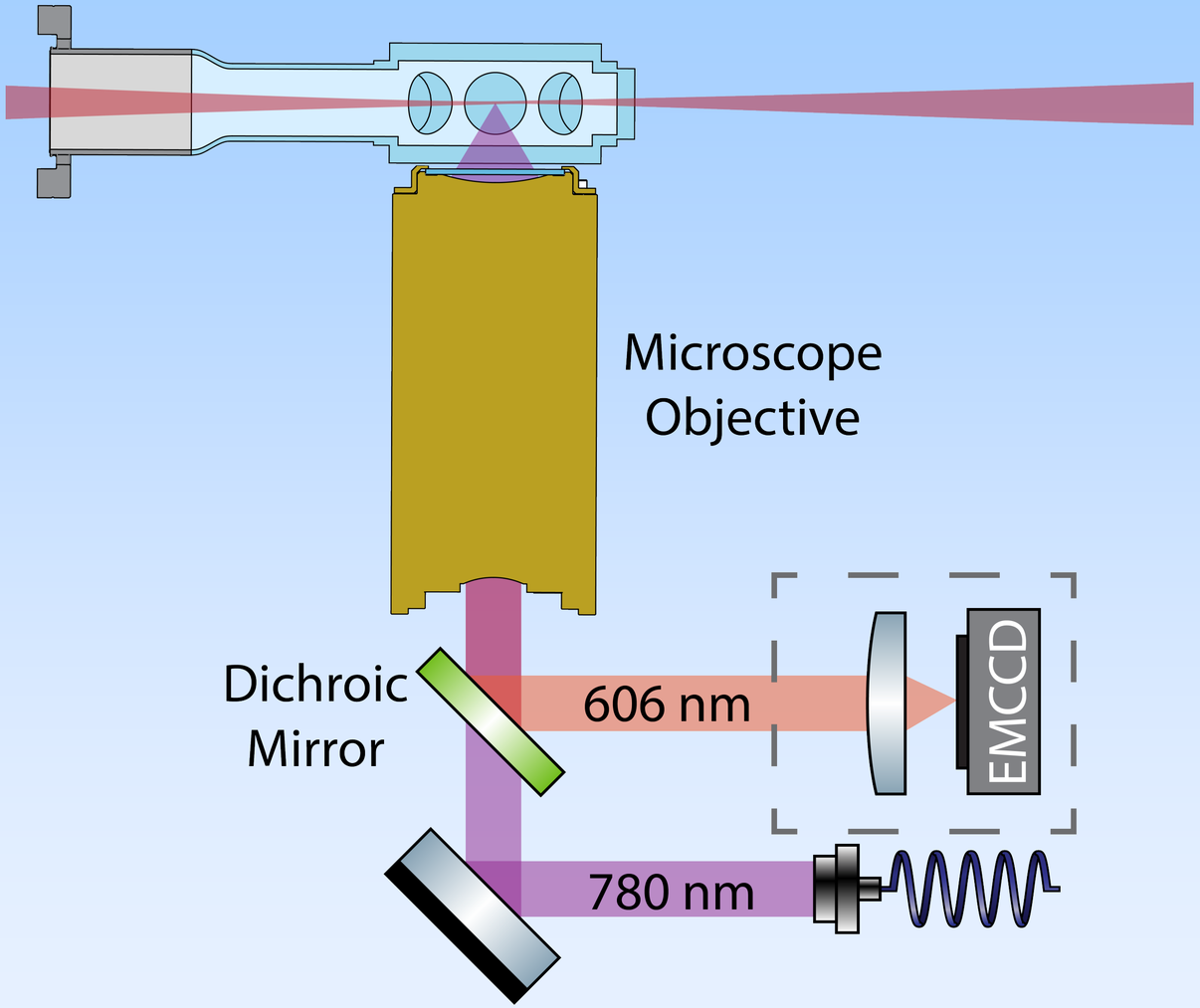
Generation 2 Apparatus….

CaF - Generation II Apparatus



CaF - Rearrangement in Science Cell



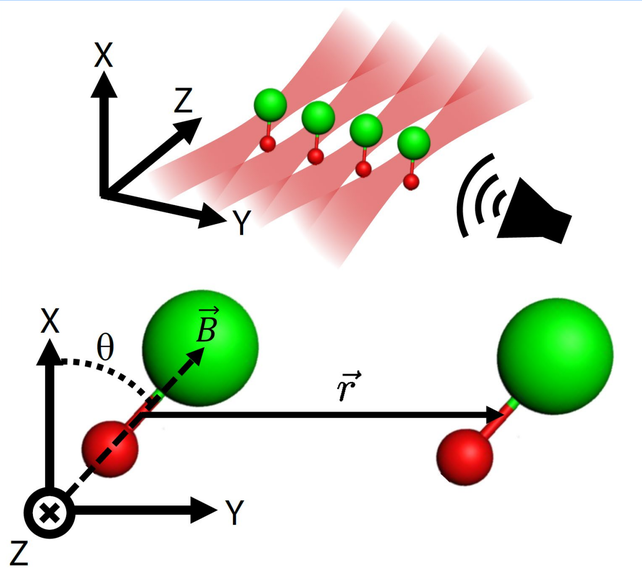


Site Resolved Addressing

CaF - Dipolar Interactions in Tweezer Arrays

Dipolar InteractionTwo Molecules in Nearby Tweezers

$$J = \frac{d^2}{4\pi\epsilon_0 r^3}(1 - 3\cos^2\theta)$$



Y. Bao, et al Arxiv (November 2022)

CaF - Dipolar Interactions in Tweezer Arrays

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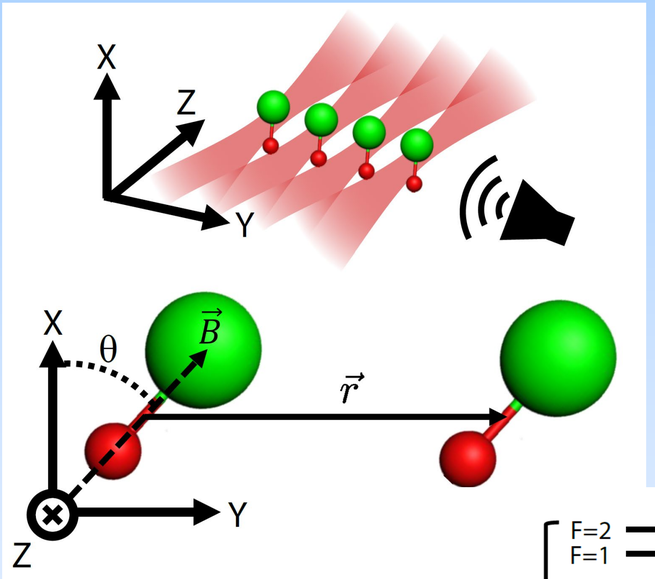

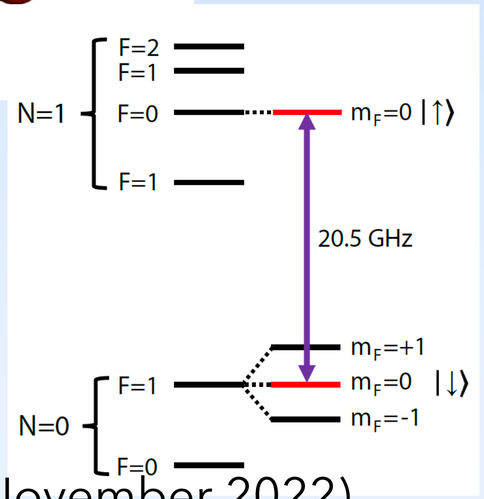

Rotational Qubit

Y. Bao, et al Arxiv (November 2022)

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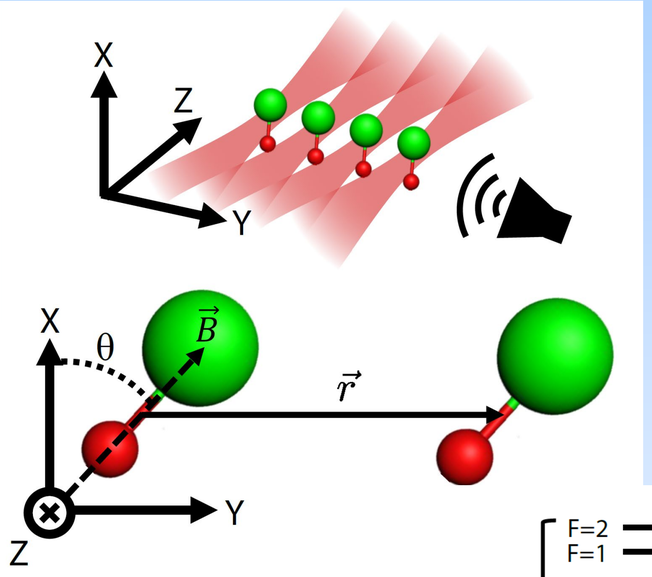
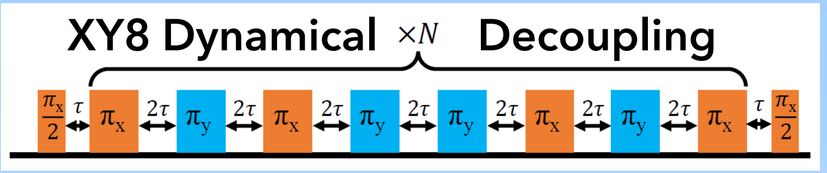
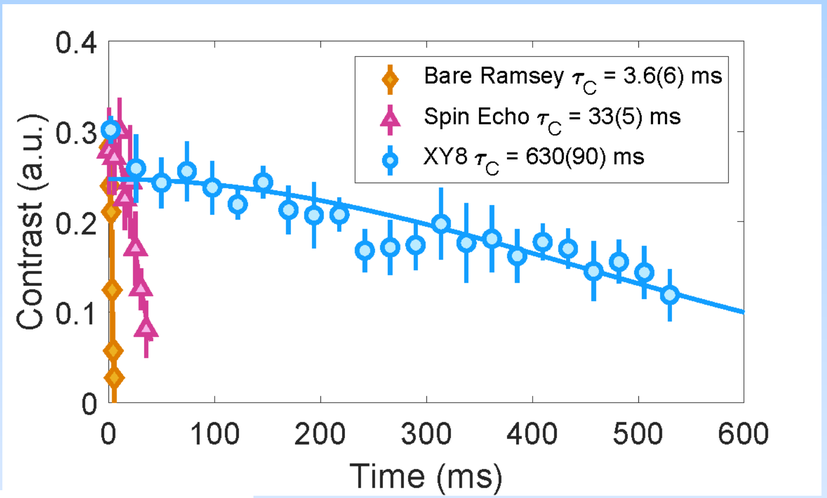
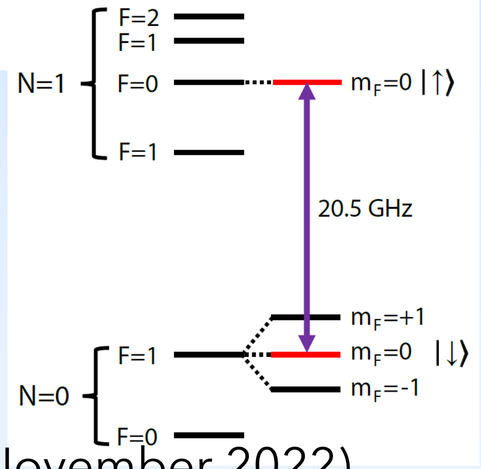


Single Molecule Rotational Qbit Coherence Time







Rotational Qubit

Y. Bao, et al Arxiv (November 2022)

CaF - Dipolar Interactions in Tweezer Arrays

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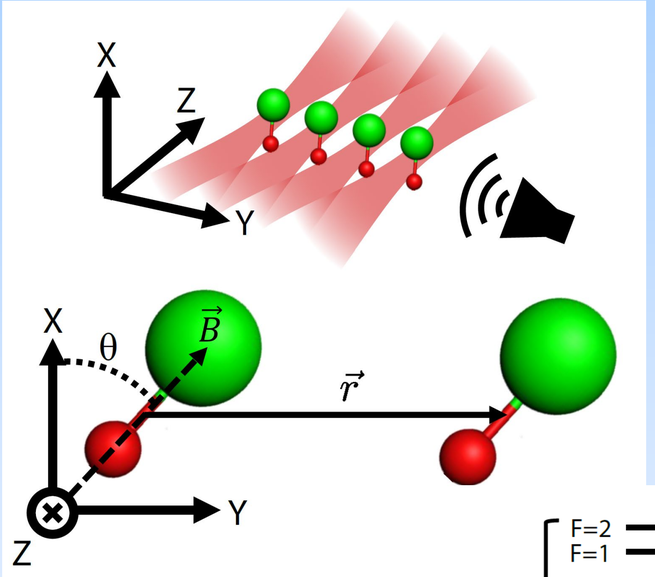
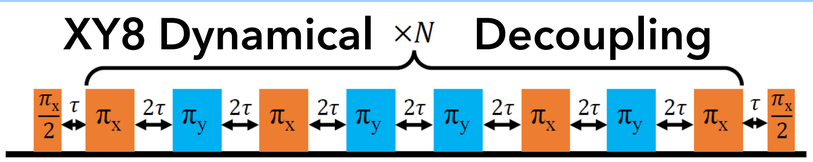
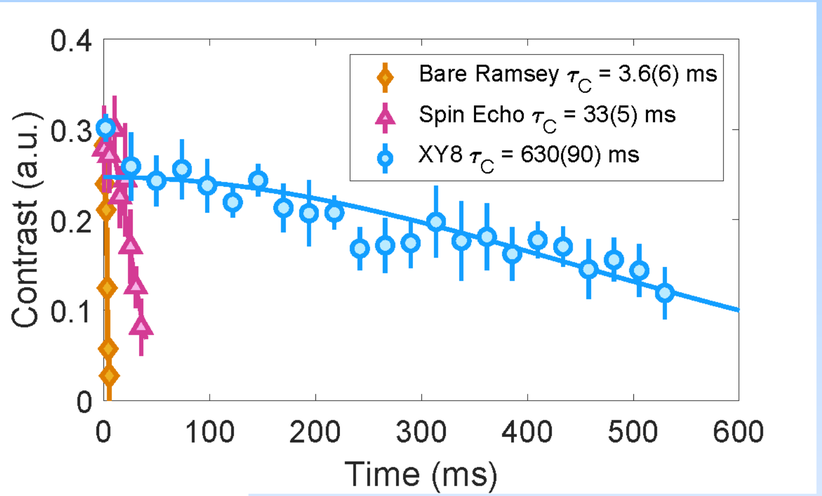
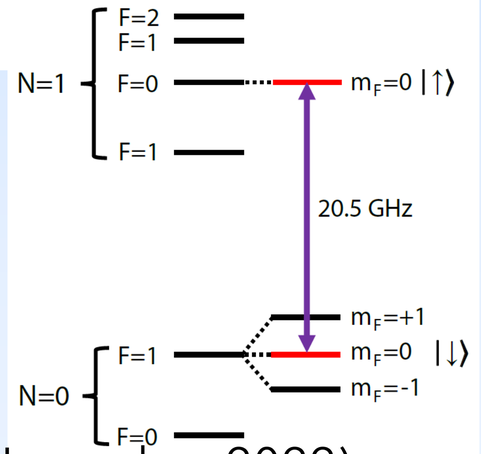


Single Molecule Rotational Qbit Coherence Time



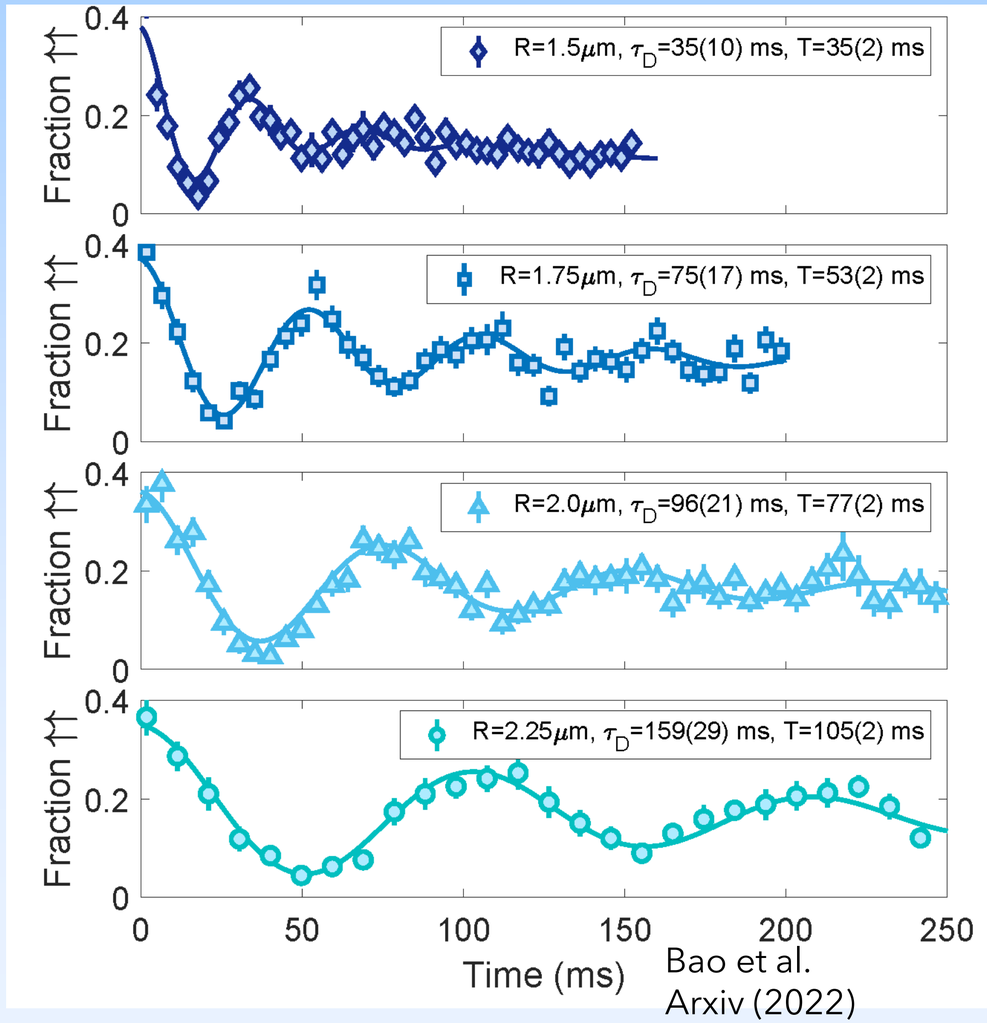




Rotational Qubit

Spin Exchange Between Two Molecules

$$H_{dip} = \frac{J}{2}(\hat{S}_1^+\hat{S}_2^- + \hat{S}_1^-\hat{S}_2^+) = J(\hat{S}_1^x\hat{S}_2^x + \hat{S}_1^y\hat{S}_2^y)$$

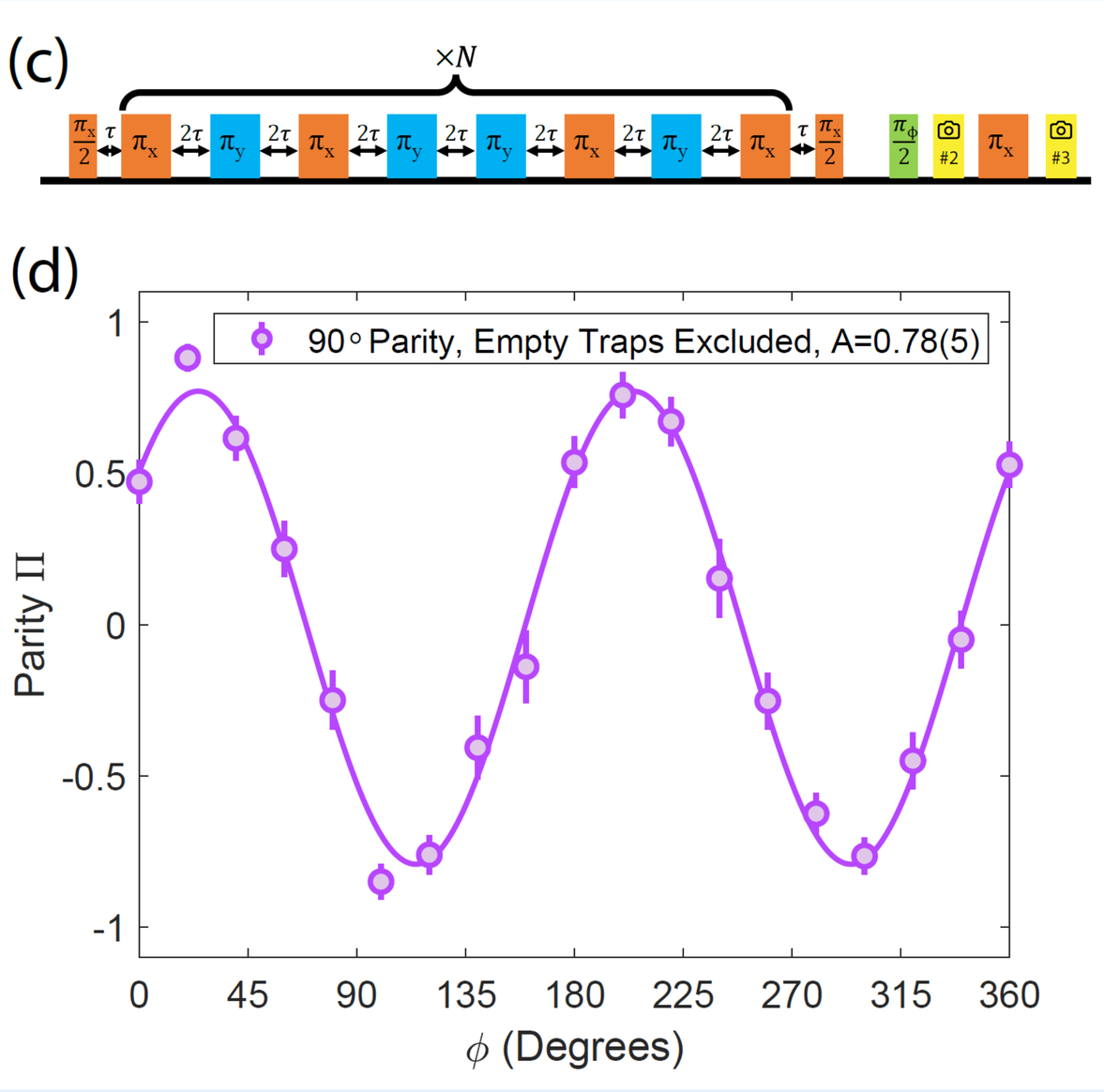


Bao et al. Arxiv (2022)

Y. Bao, et al Arxiv (November 2022)

Bell State Fidelity

- Low state preparation (optical pumping) efficiency results in empty tweezer traps
- Cannot distinguish between empty trap or zero qubit state
- Excluding empty traps using an additional π-pulse and imaging pulse
- Bell state fidelity after exclusion of empty traps reaches 0.87(6)
- Agrees well with SPAM corrected fidelity

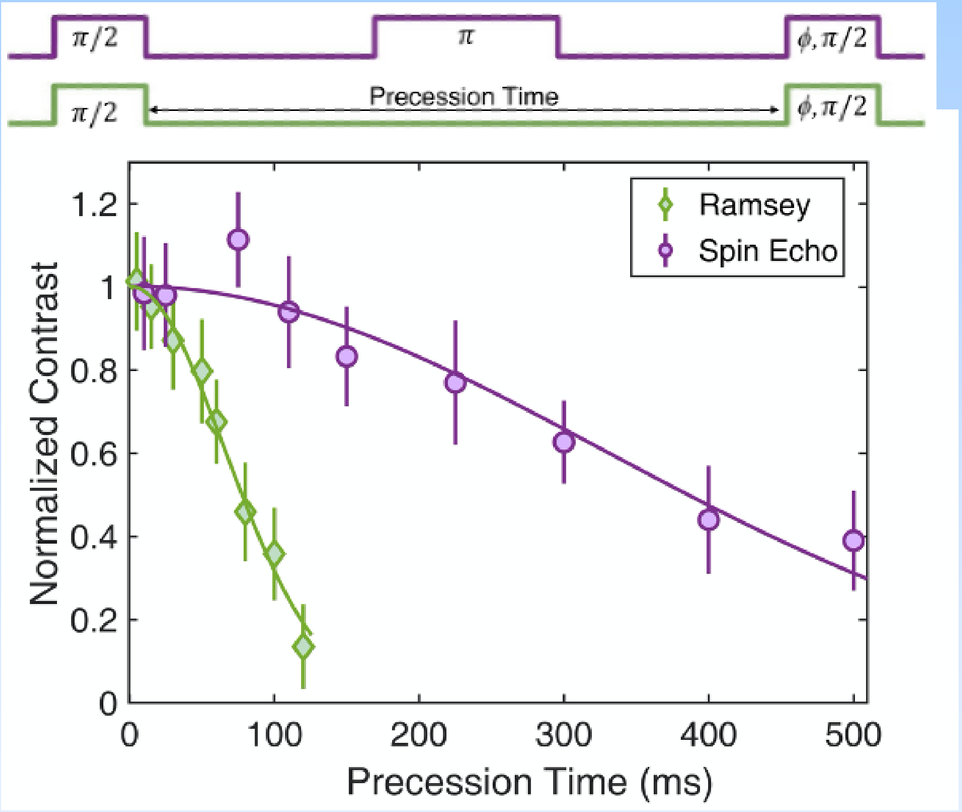


$$\Pi = \langle \hat{\Pi} \rangle = \langle \hat{S}_1^z \hat{S}_2^z \rangle = P_{\uparrow\uparrow} + P_{\downarrow\downarrow} - P_{\uparrow\downarrow} - P_{\downarrow\uparrow}$$

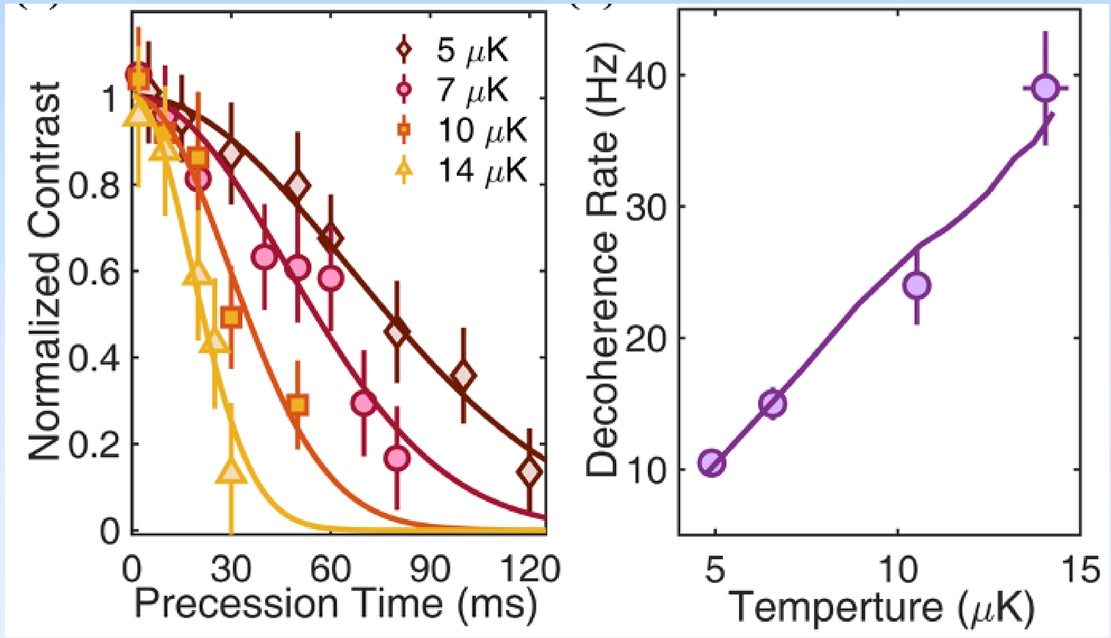
Y. Bao, et al Arxiv (November 2022)

CaF - Qbit Coherence Times from Generation 1 Apparatus

S.Burchesky, et al. PRL (2021)



Further improvements in cooling (ex. Raman sideband) expected to increase coherence times



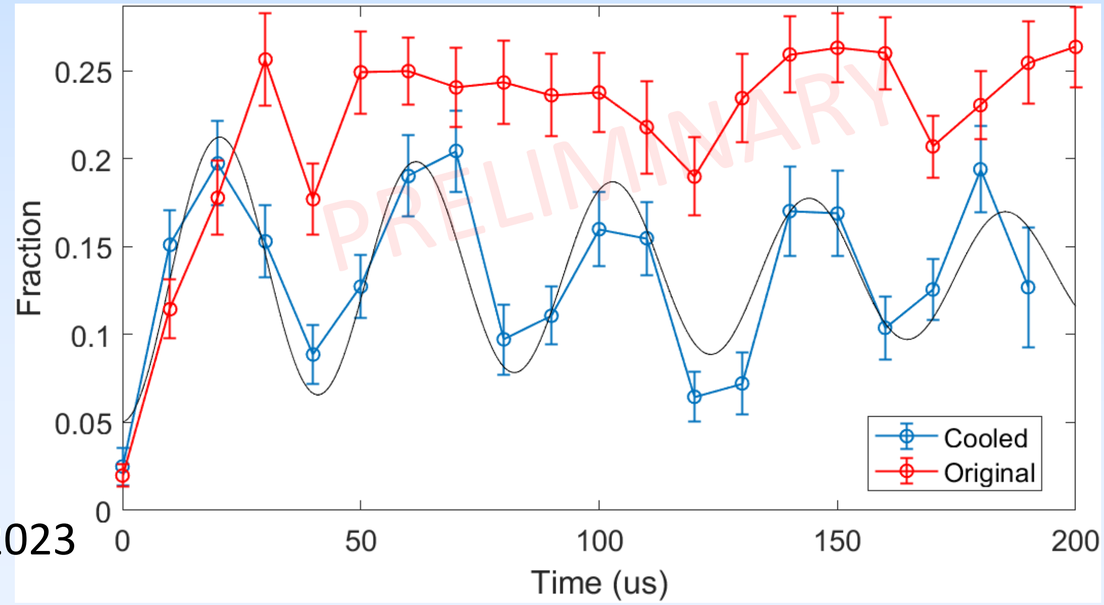
- **100 ms Ramsey coherence time**
- **500 ms Spin Echo coherence time**

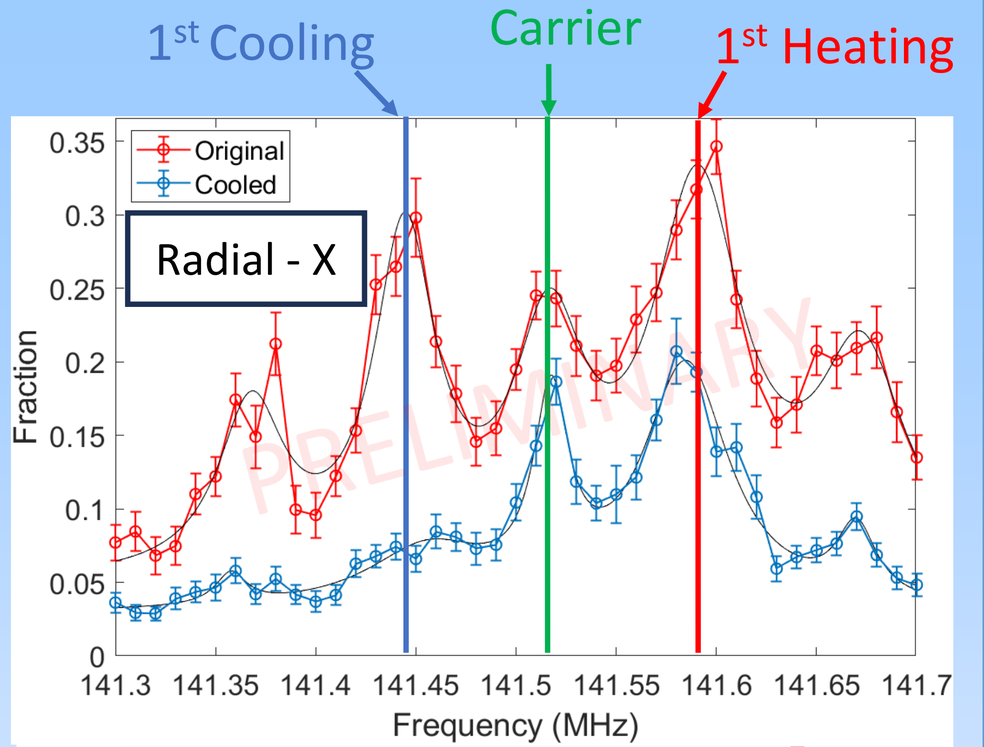
Back to Generation 2 Apparatus….

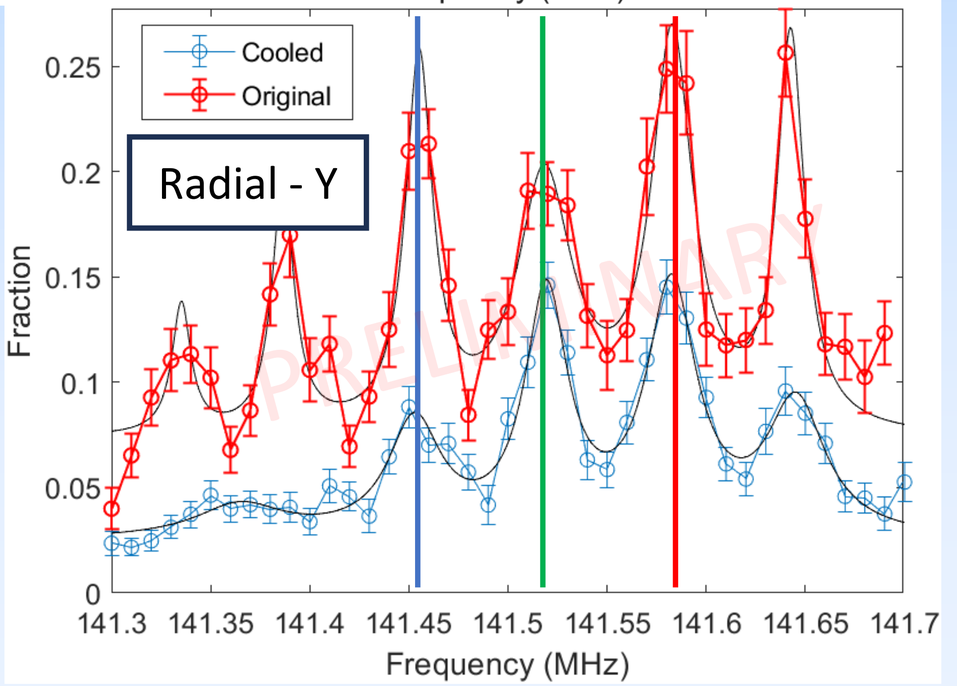
Raman Sideband Cooling for Higher Fidelity

Proposal: Caldwell and Tarbutt, Phy. Rev. Res. 2, 013251 (2020)

- Drive high-order sidebands for initial cooling
- Use shaped Raman pulse (Blackman)
- Cooling to $n_{rad} \approx 0.3$ on both radial axes
- Coherence of the Rabi oscillation on carrier is greatly improved







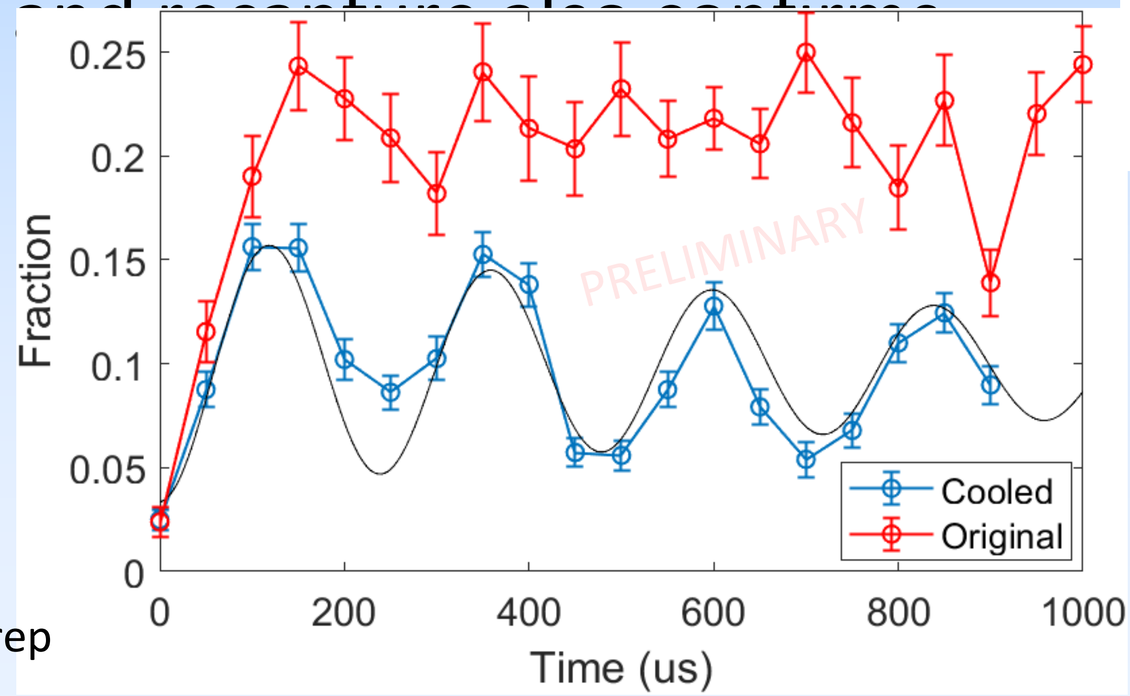
Y. Bao et al. DAMOP 2023 (June 2023)

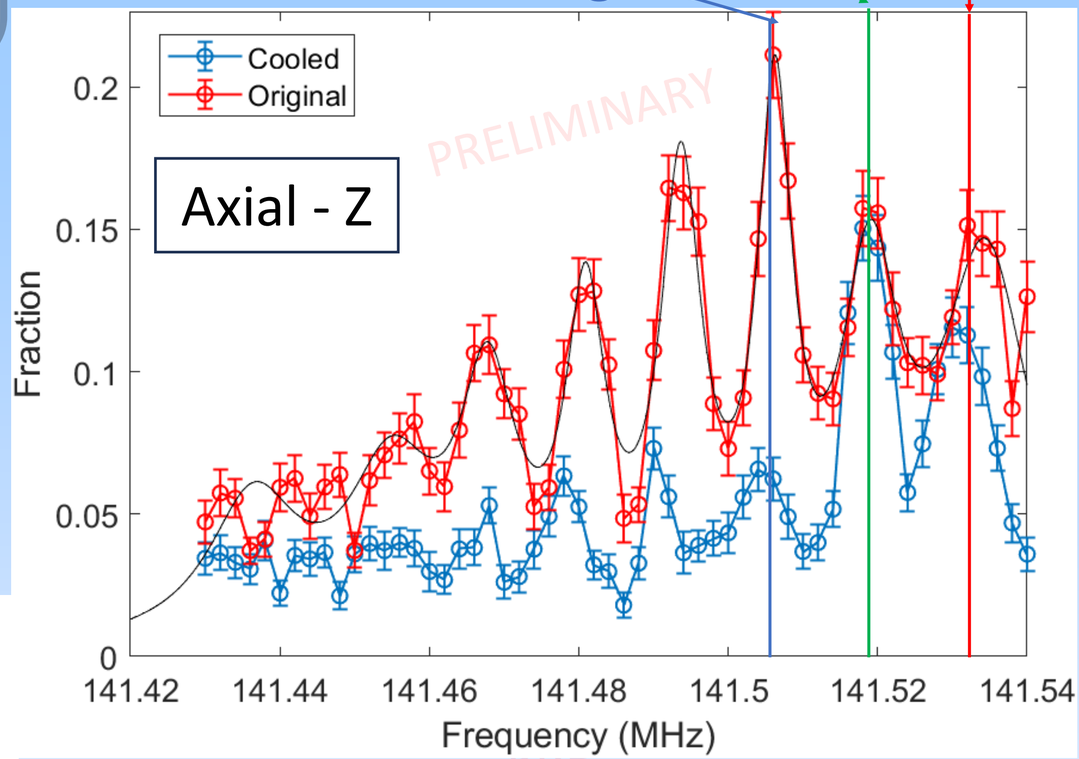
Back to Generation 2 Apparatus….

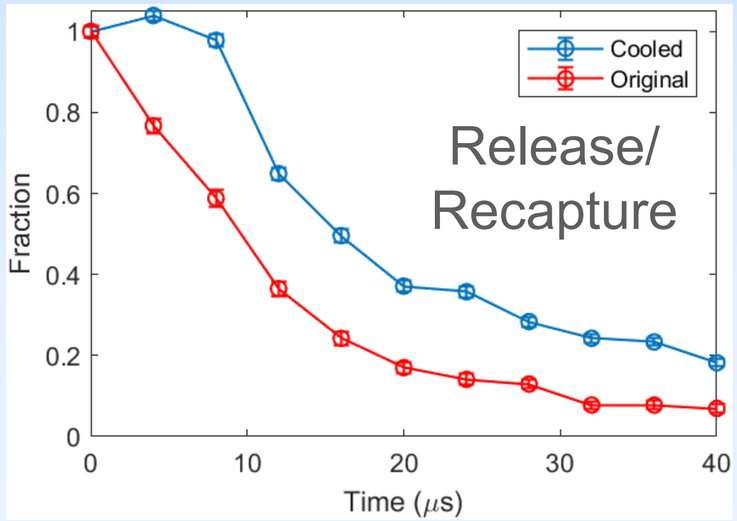
Raman Sideband Cooling for Higher Fidelity

Proposal: Caldwell and Tarbutt, Phy. Rev. Res. 2, 013251 (2020)

- Cooling to $n_{ax} < 1$ on axial direction
- Coherence of the Rabi oscillation on carrier is greatly improved
- Release and recapture also confirms cooling



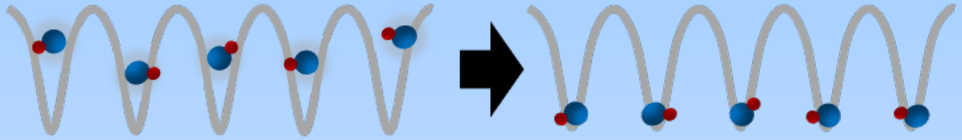




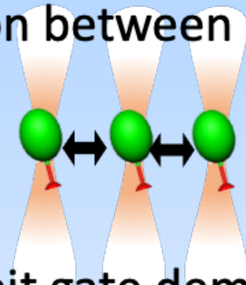
Y. Bao et al. in prep (June 2023)

CaF - Near Term Goals

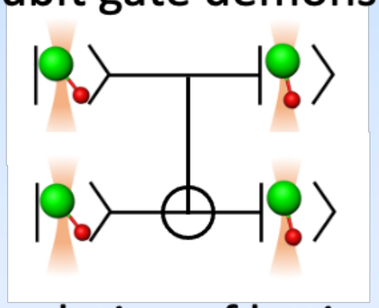
Raman sideband cooling of CaF molecules in optical tweezers



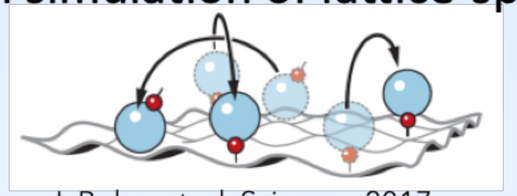
Dipolar interaction between CaF in tweezer sites



Two qubit gate demonstration

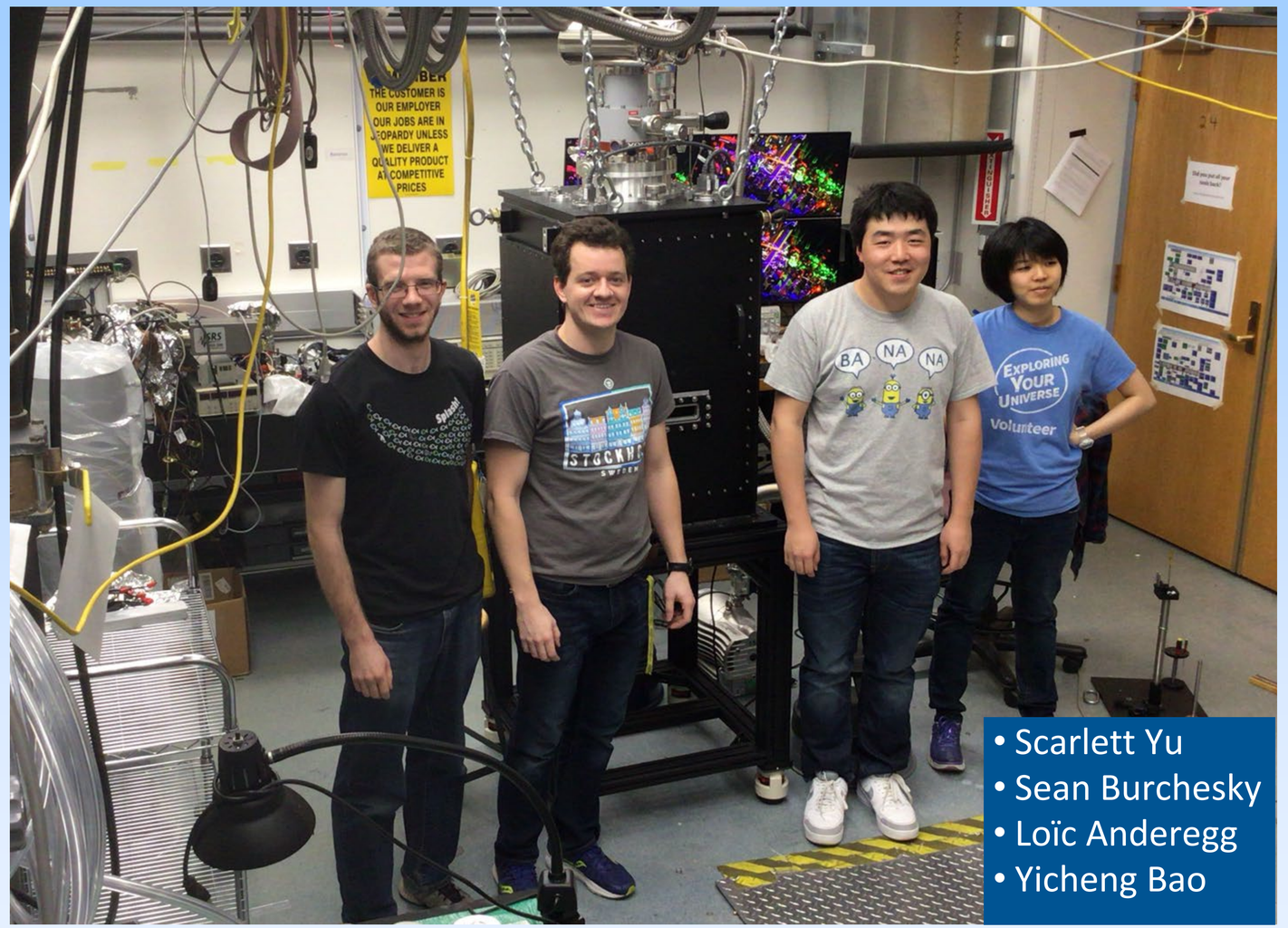


Quantum simulation of lattice-spin models



J. Bohn, et. al. Science, 2017

CaF Team and Funding



- Scarlett Yu
- Sean Burchesky
- Loïc Anderegg
- Yicheng Bao

Eunmi Chae

Korea University

Wolfgang Ketterle

MIT

Kang-Kuen Ni

Harvard Chemistry

Harvard Quantum Initiative
IN SCIENCE AND ENGINEERING

Cheuk Group, Similar Results

Dipolar Entanglemen
and RSB of CaF

- Similar work is
  lab @ Princeto versity
- Rearrange imp nted
- Better state pr tion
- Studies on the stness of
  the Bell state

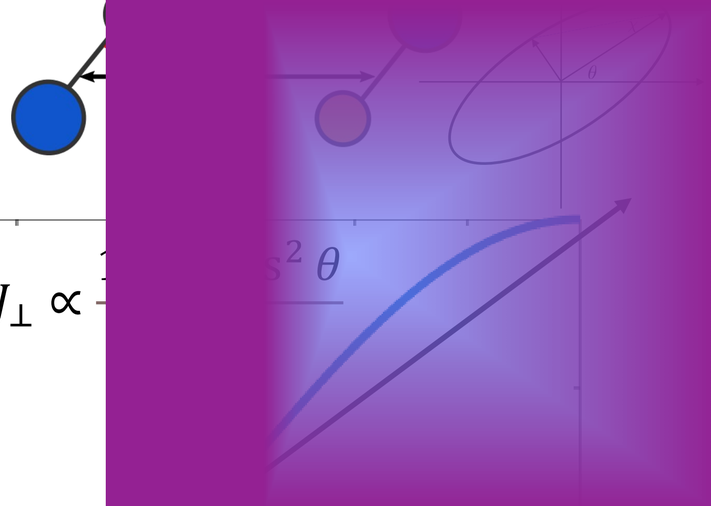
See Yesterday's Talk
for
Latest Princeton CaF News!

Holland, et al. *arXiv:2210.06309* (October 2022)

Lu, Li, Holland, Cheuk,
arXiv:2306.02455 (June 2023)
and DAMOP 2023 (June 2023)

Ni Group,
Similar Results

T



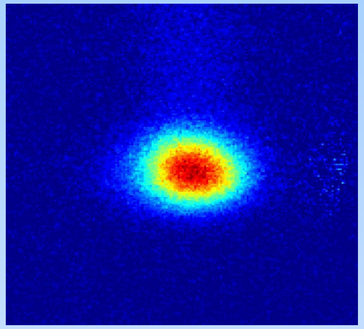
With indivi
er

See Talk this afternoon
for
Latest NaCs News!

Diatomics: Expanding Field, Driven By Quantum Science

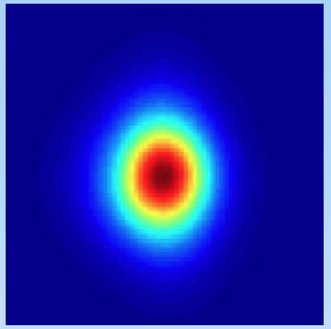
And even more species are being considered!

CaF



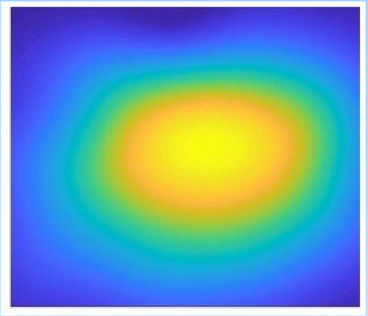
Tarbutt, Doyle
Cheuk, Langen, Ospelkaus, …

SrF



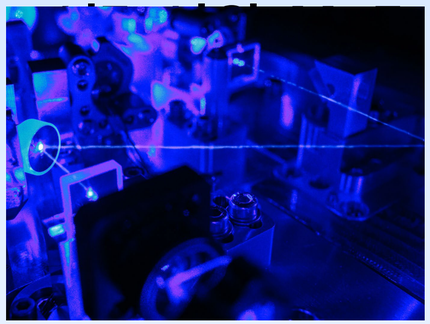
DeMille

Narrow Line Transitions YO



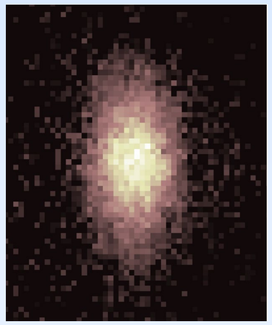
Ye

Low Mass with UV Transitions



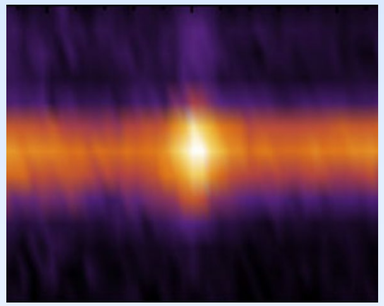
Truppe, Hemmerling, McCarron, Chae, Yin, Norrgard

Chemistry and Precision Measurement Relevant CH, CaH, BaH



Zelevinsky, McCarron

EDM Sensitive Molecules YbF, BaF



Tarbutt, Langen

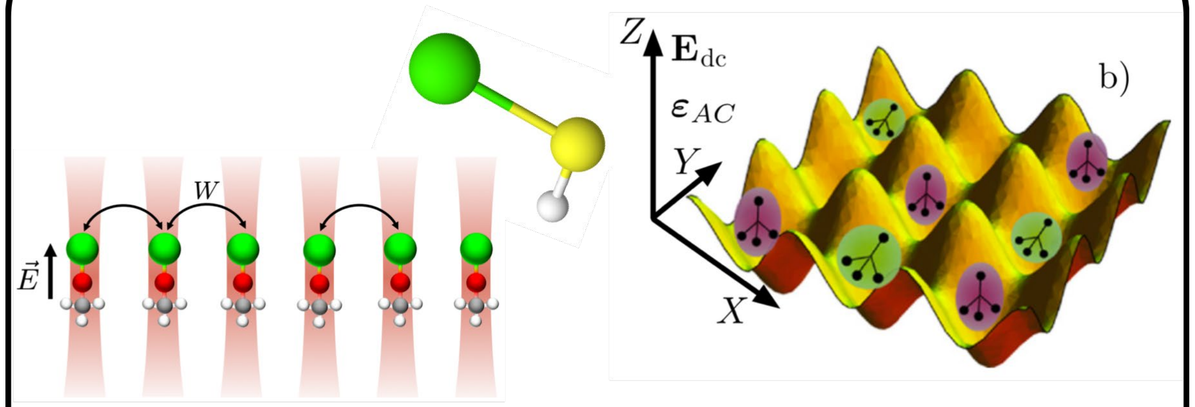
Italics Name means MOT achieved

Now to Polyatomic Molecules

2018 Porsche 718 Cayman GTS

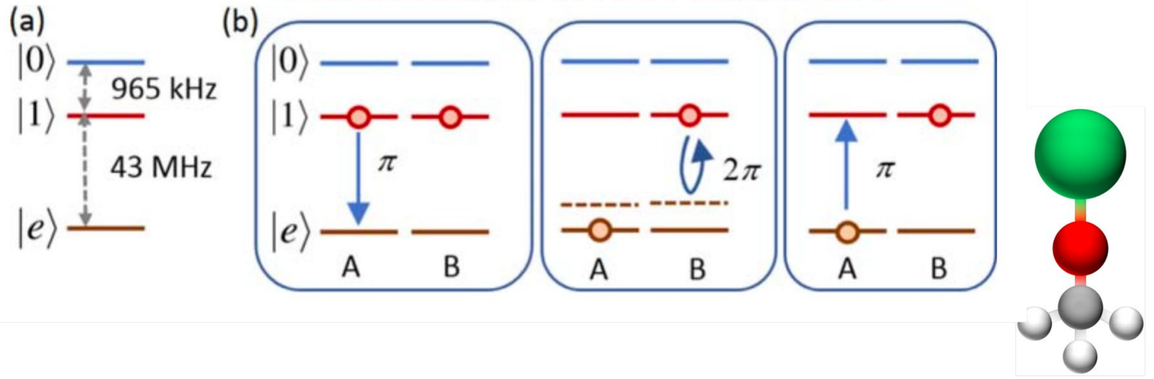
Motivations for Polyatomic Molecules

Quantum Simulation



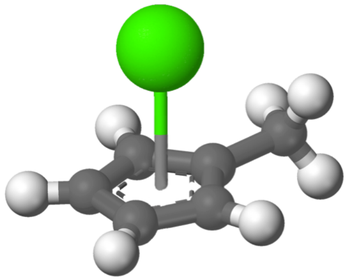
Wall, Maeda, and Carr, Ann. Phys. **525**, 845 (2013)
Wall, Maeda, and Carr, NJP **17**, 021001 (2015)

Quantum Information

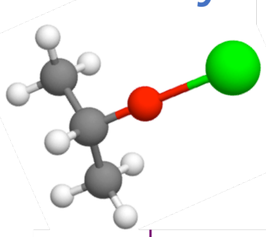


Yu,….JMD, NJP 21, 093049 (2019)
Albert et al., arXiv:1911.00099

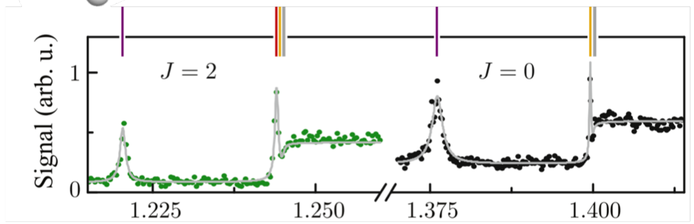
Ultracold Chemistry and Collisions



Augenbraun, Zelevinsky, Kozyryev, JMD, PRX 10, 031022 (2020)
Bohn, Rey, Ye, Science **357** (2017)

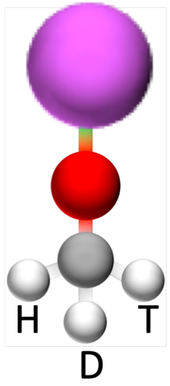


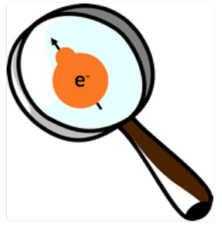
Augustovičová and Bohn, NJP **21** (2019)

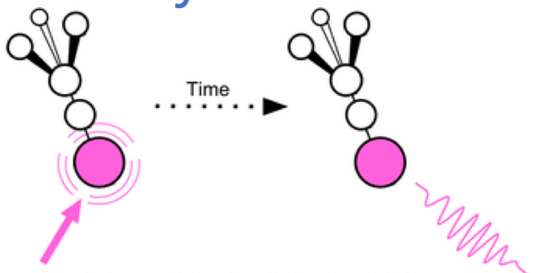


McGuyer et al., NJP **357** (2015)

Precision Measurement/ Fundamental Physics

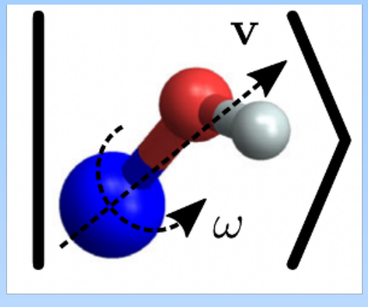




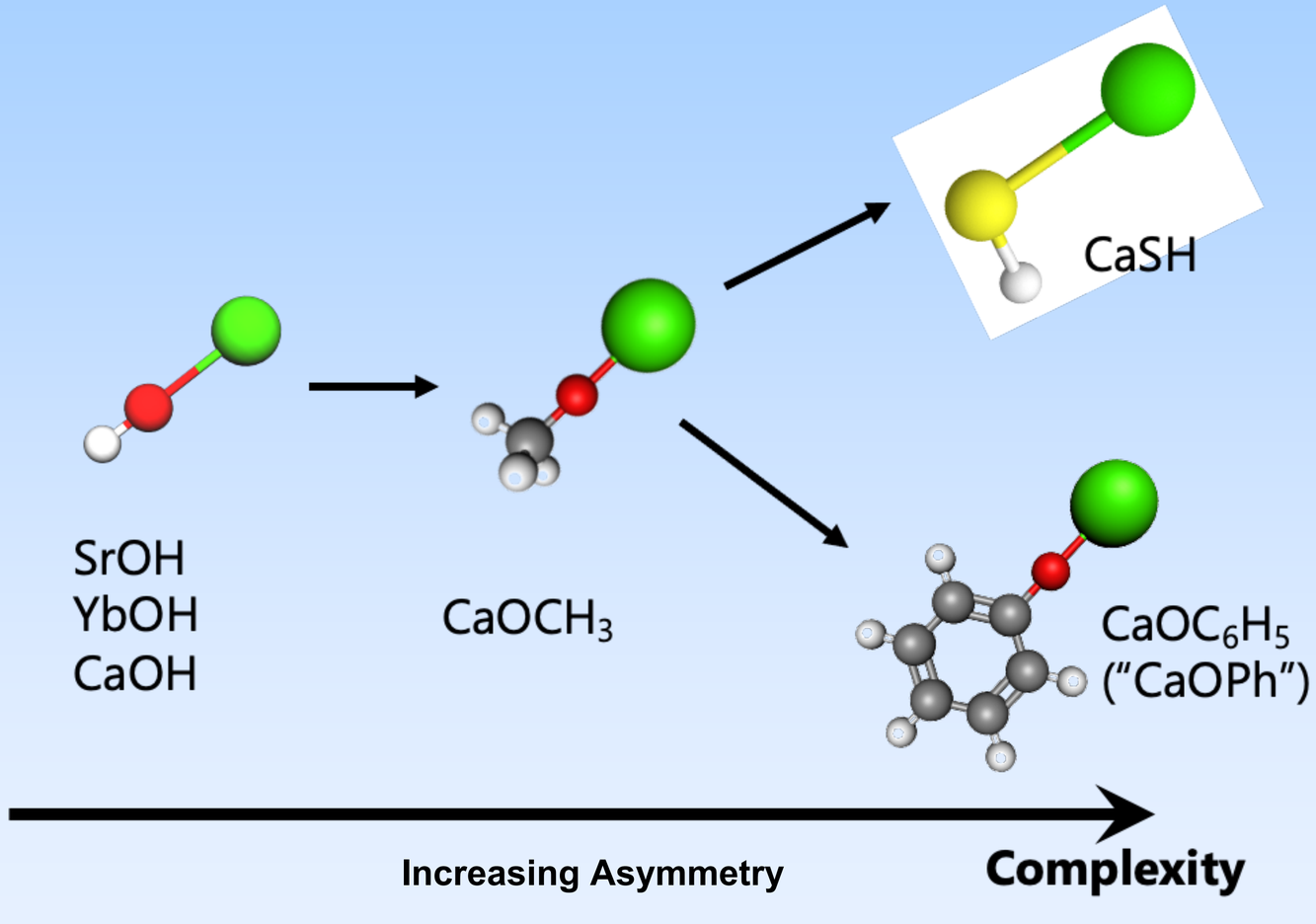


Kozyryev, Lasner, JMD, PRA 103, 043313 (2021)
Kozyryev and Hutzler PRL 119, 133002 (2017)
Augenbraun, Zelevinsky, Kozyryev, JMD, PRX 10, 031022 (2020)

Large and Asymmetric Top Molecules? (And Organic Molecules)

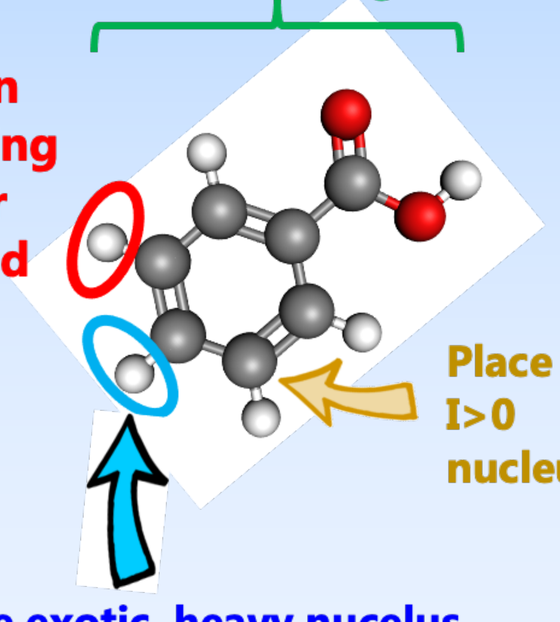


See Preskill group paper:
PRX **10**, 31050 (2020)
Quantum Error Correction



"Ultracold orgo"

Append an optical cycling center for cooling and control



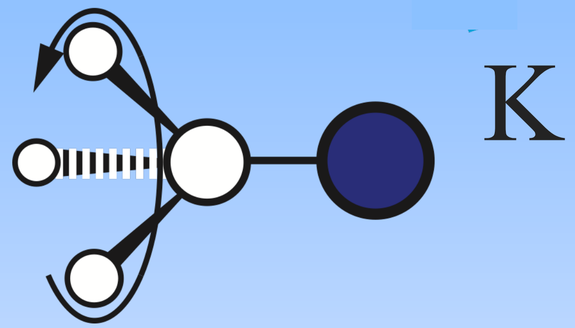
Place an I>0 nucleus

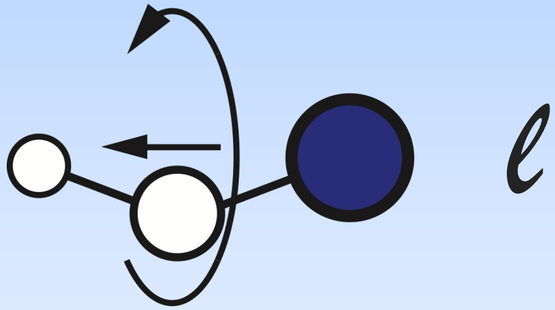
Substitute exotic, heavy nucelus

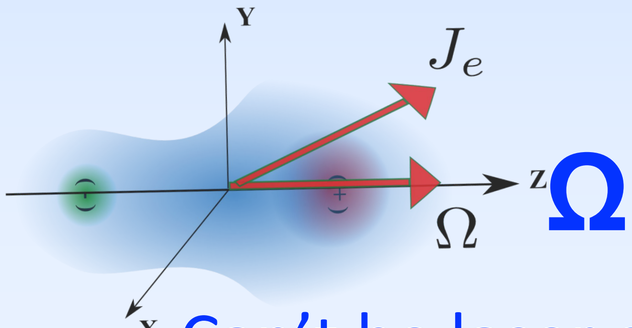
Molecular Asymmetry and Optical Cycling: Laser Cooling Asymmetric Top Molecules B. L. Augenbraun, JMD, T. Zelevinsky, I. Kozyryev, Phys. Rev. X 10,

Orbital Angular Momentum Along the Internuclear Axis

Molecular Frame Quantum Numbers

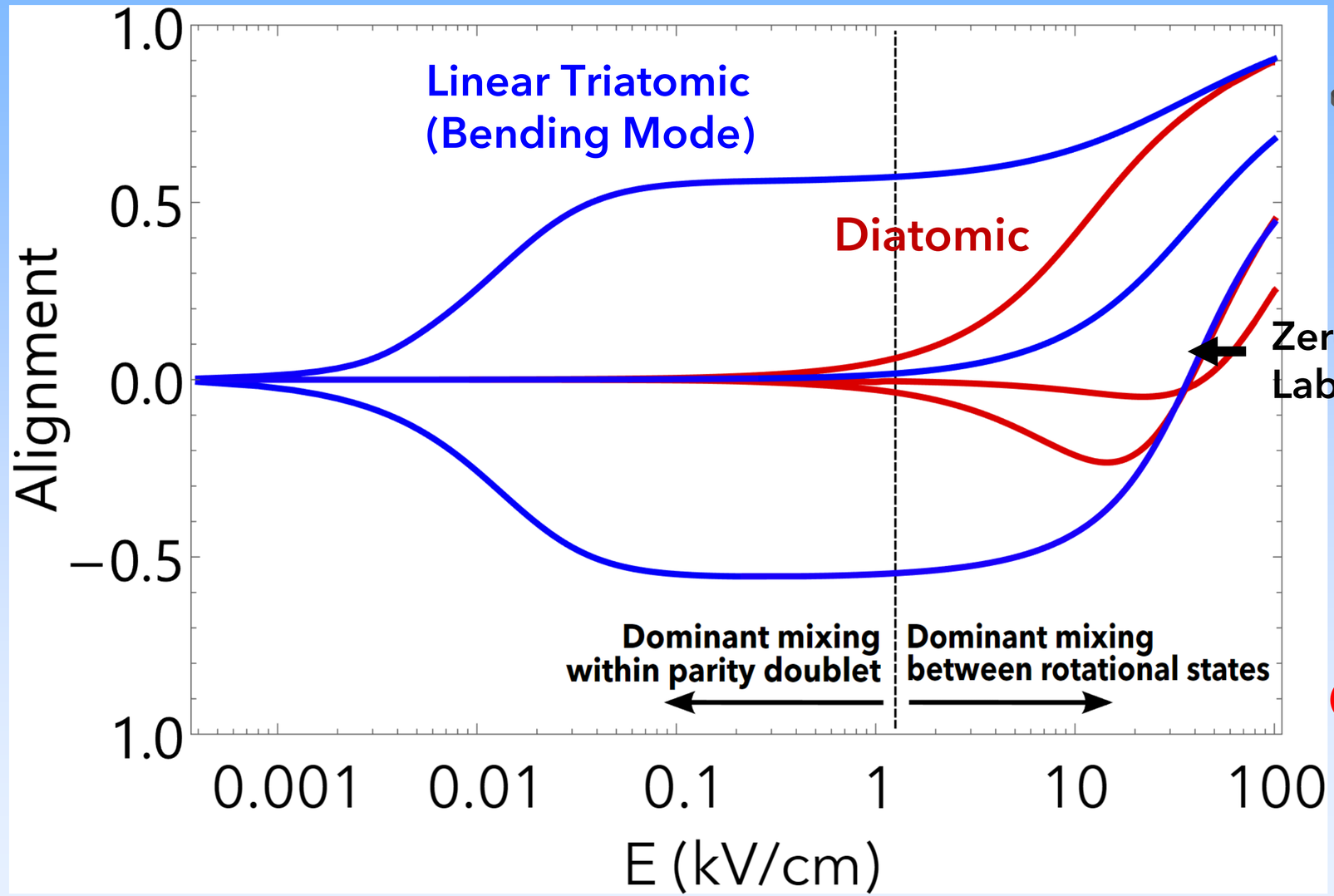


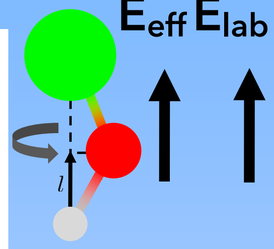


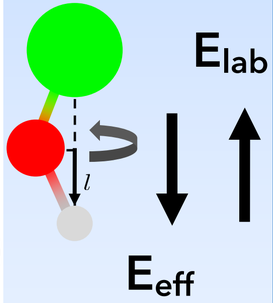


Can't be laser cooled !

Why polyatomic molecules?







Laser Cooling Polyatomic *Molecules*

October 2016 ChemPhysChem Paper. See also Isaev and Berger 2016

Special Issue

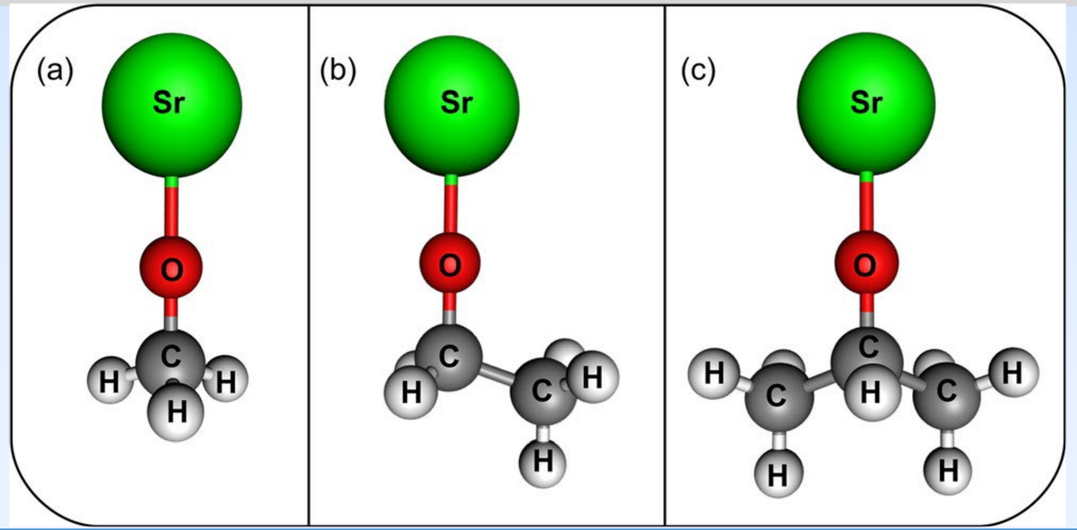
Proposal for Laser Cooling of Complex Polyatomic Molecules

Ivan Kozyryev,*[a, b] Louis Baum,[a, b] Kyle Matsuda,[a, b] and John M. Doyle[a, b]

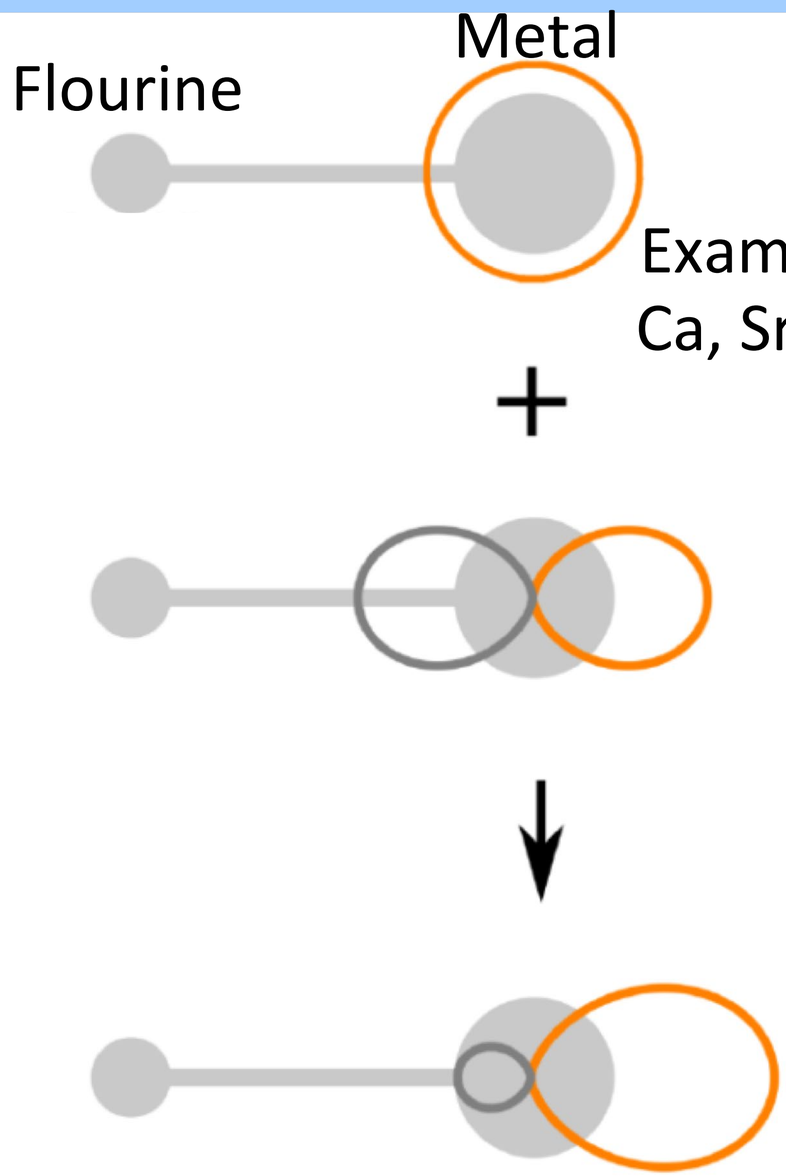
An experimentally feasible strategy for direct laser cooling of polyatomic molecules with six or more atoms is presented. Our approach relies on the attachment of a metal atom to a complex molecule, where it acts as an active photon cycling site. We describe a laser cooling scheme for alkaline earth monoalkoxide free radicals taking advantage of the phase space compression of a cryogenic buffer-gas beam. Possible applications are presented including laser cooling of chiral molecules and slowing of molecular beams using coherent photon processes.

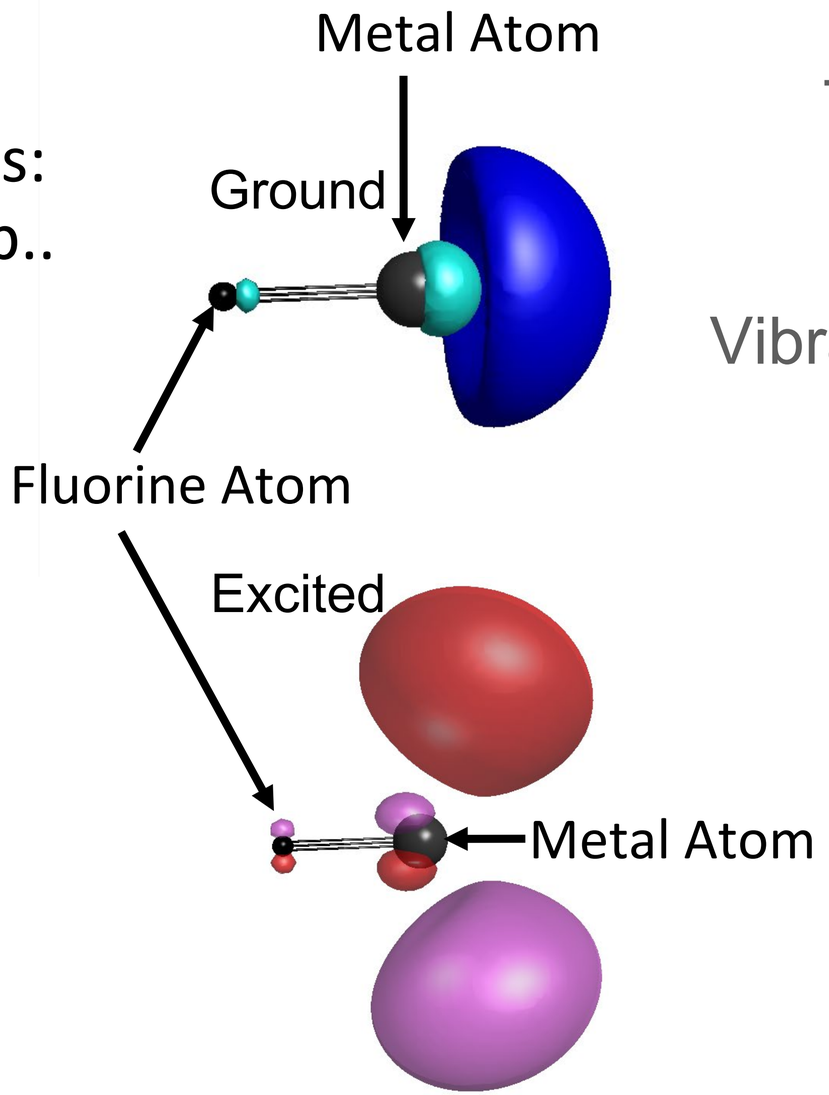
Metal-Oxide-Radical
"MOR"
Molecules

a.k.a Alkaline Earth Atom Pseudofluoride Molecules



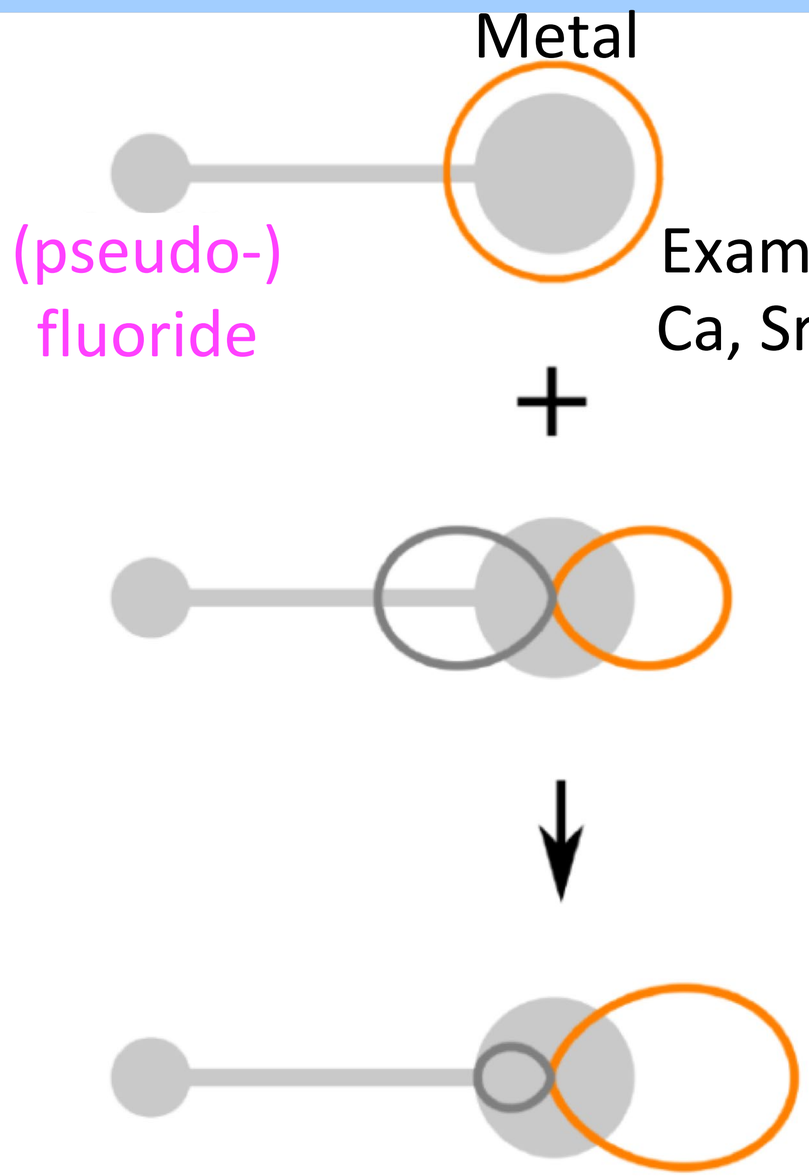
Diatomic Molecules that can be Laser Cooled

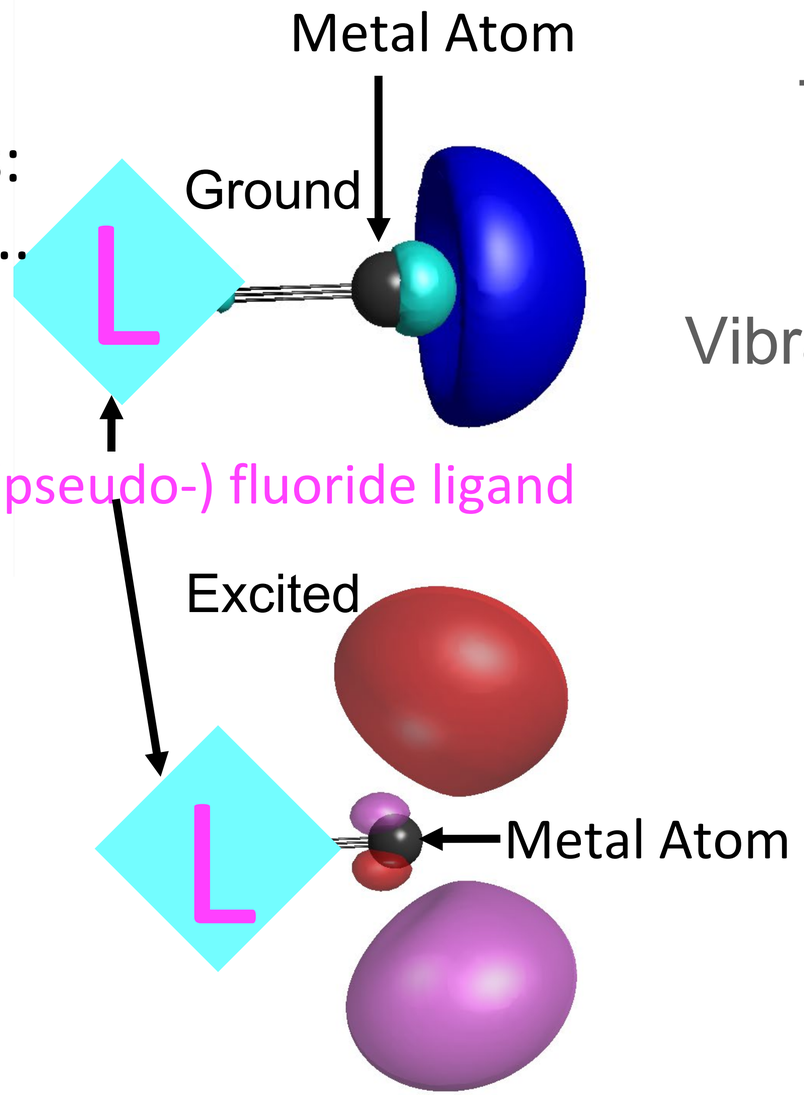




This bonding motif is
The reason that
CaF has small
Vibrational Branching Ratios

Class of Molecules that can be Laser Cooled





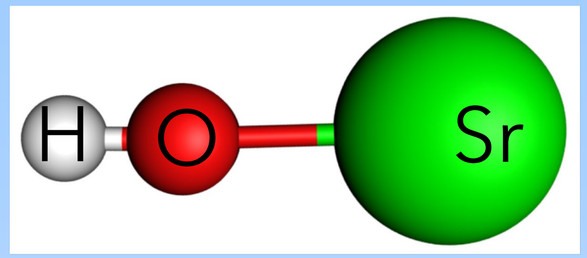
This bonding motif is
The reason that
CaF has small
Vibrational Branching Ratios

If this motif
is kept,
then…
small VBRs
should remain

What to do with them? We have a plan!… ultracold polyatomic EDM search

EDM Experiment

PolyEDM



SrOH in an ODT

Then

YbOH, RaOH, etc..

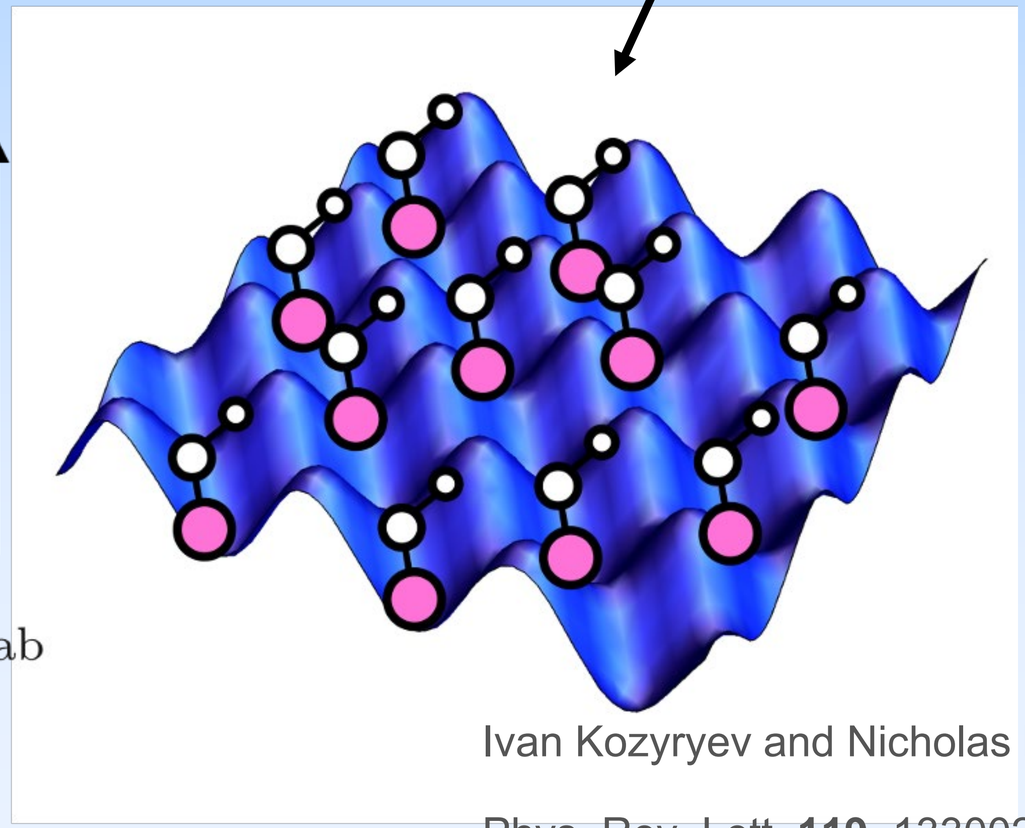
Heavy, polar molecule sensitive to new physics

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

Mass reach for New Physics ~ 1,000 TeV

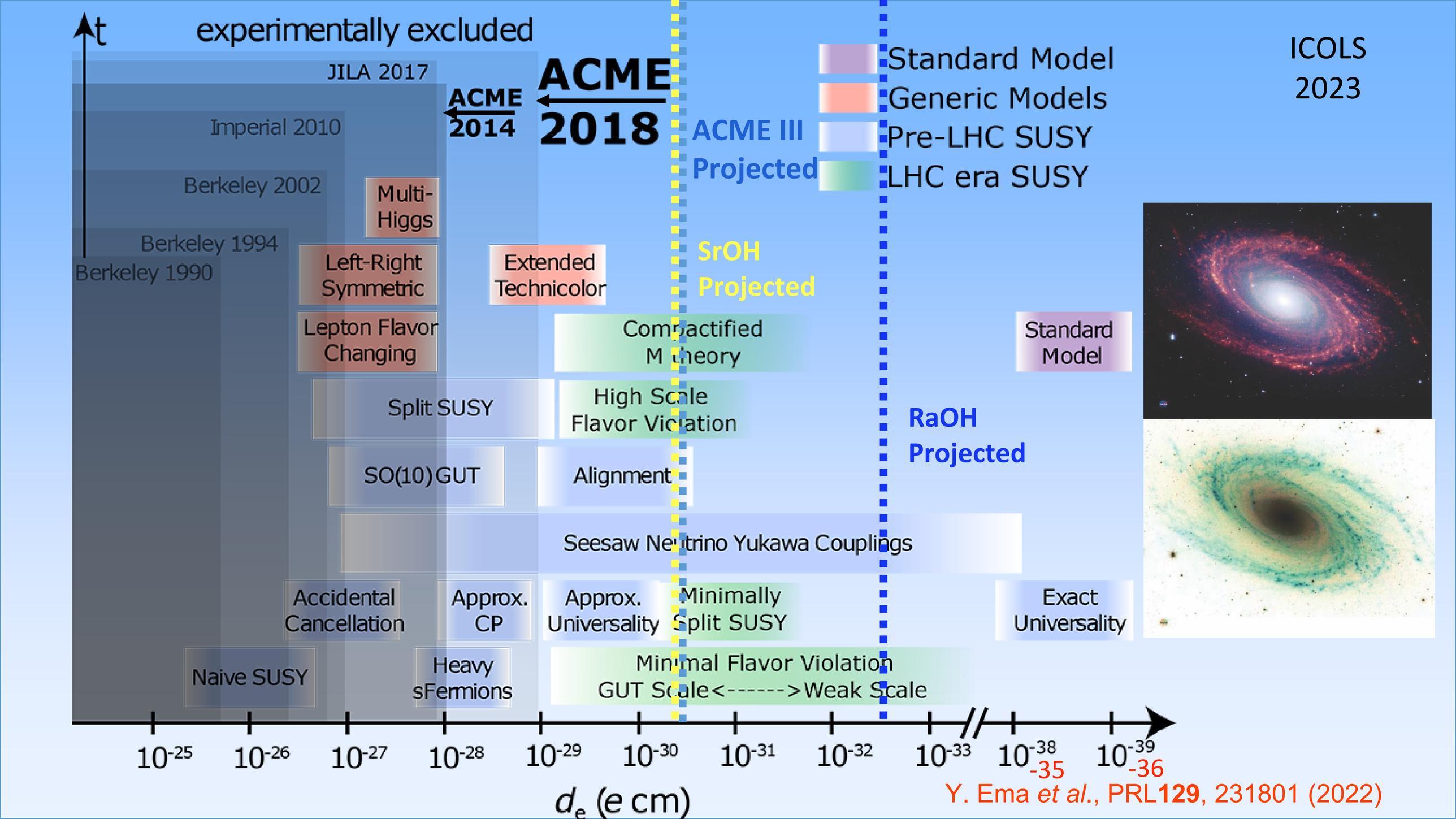
Optical Lattice "Clock"

Ultracold Heavy Polyatomic Molecules



Ivan Kozyryev and Nicholas R. Hutzler

Phys. Rev. Lett. **119**, 133002 (2017)

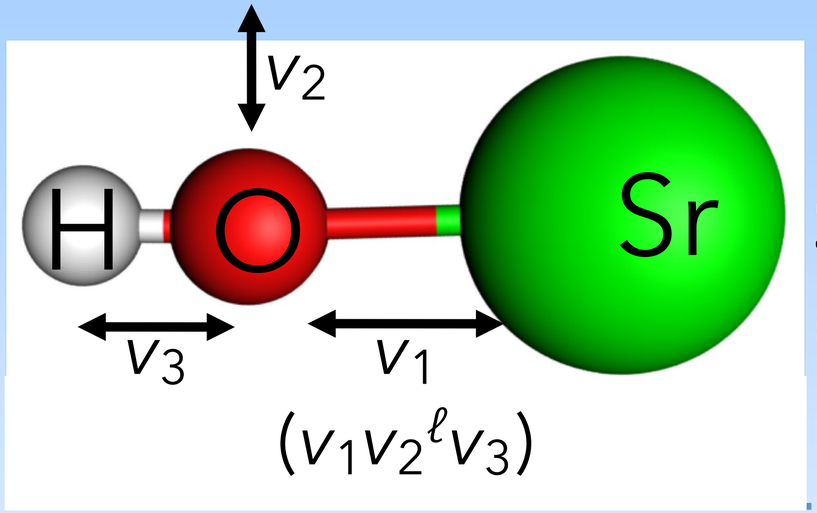

t
experimentally excluded
JILA 2017
ACME 2014
ACME 2018
Imperial 2010
Berkeley 2002
Berkeley 1994
Berkeley 1990
Multi-Higgs
Left-Right Symmetric
Extended Technicolor
Lepton Flavor Changing
Compactified M theory
Split SUSY
High Scale Flavor Violation
SO(10) GUT
Alignment
Seesaw Neutrino Yukawa Couplings
Accidental Cancellation
Approx. CP
Approx. Universality
Minimally Split SUSY
Exact Universality
Naive SUSY
Heavy sFermions
Minimal Flavor Violation
GUT Scale<------>Weak Scale
ACME III Projected
SrOH Projected
RaOH Projected
Standard Model
Generic Models
Pre-LHC SUSY
LHC era SUSY
Standard Model
ICOLS 2023
10-25
10-26
10-27
10-28
10-29
10-30
10-31
10-32
10-33
10-38
10-39
-35
-36
de (e cm)
Y. Ema et al., PRL129, 231801 (2022)

What to do with them? We have a plan!… ultracold polyatomic Dark Matter search

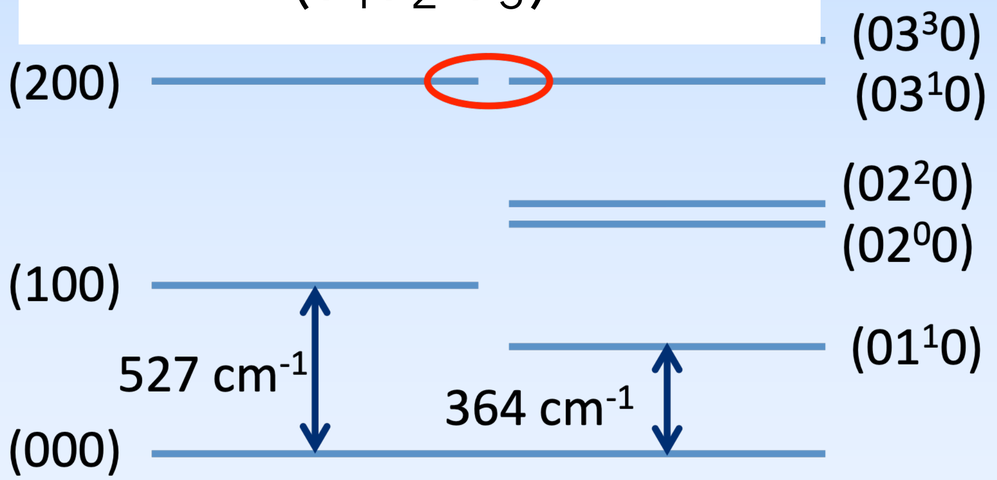
Dark Matter Experiment

New Dark Matter experiment, Oscillating Physical Constants

Level spacing will change if fundamental constants change



SrOH in an Optical Dipole Trap

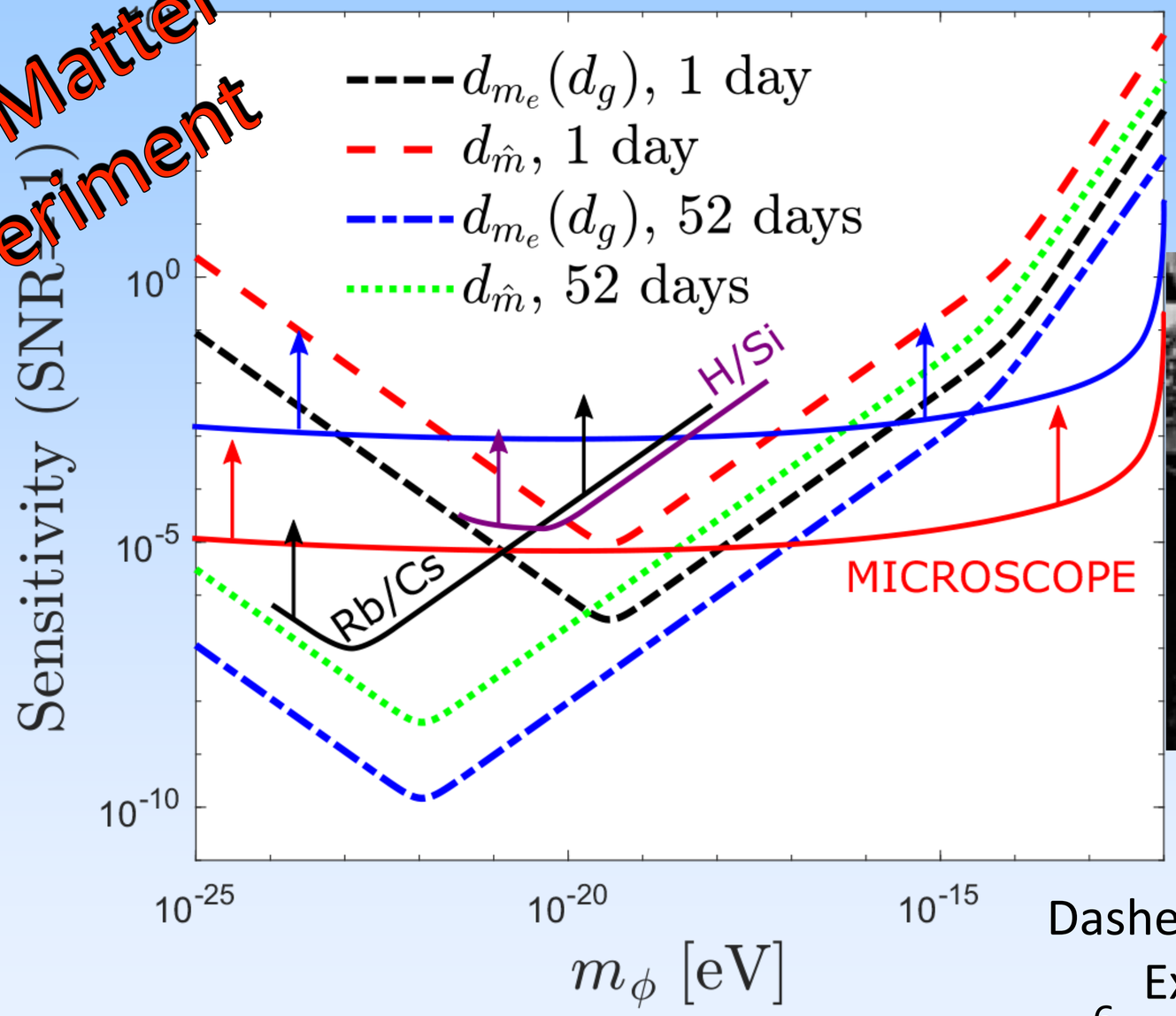


"accidental" degeneracy

Kozyryev, Lasner, JMD PRA 103, 043313 (2021)

Example 2: SrOH Dark Matter Experiment Sensitivity

Dark Matter Experiment



New experiment,
Ultracold SrOH in Optical Lattice

Kozyryev, Lasner, JMD PRA 103, 043313 (2021)

Dashed Lines are For **SrOH** Dark Matter Experiment

10^6 Molecules, ~100 ms coherence time.

Funding from Q-sense (QLCI from NSF) Project

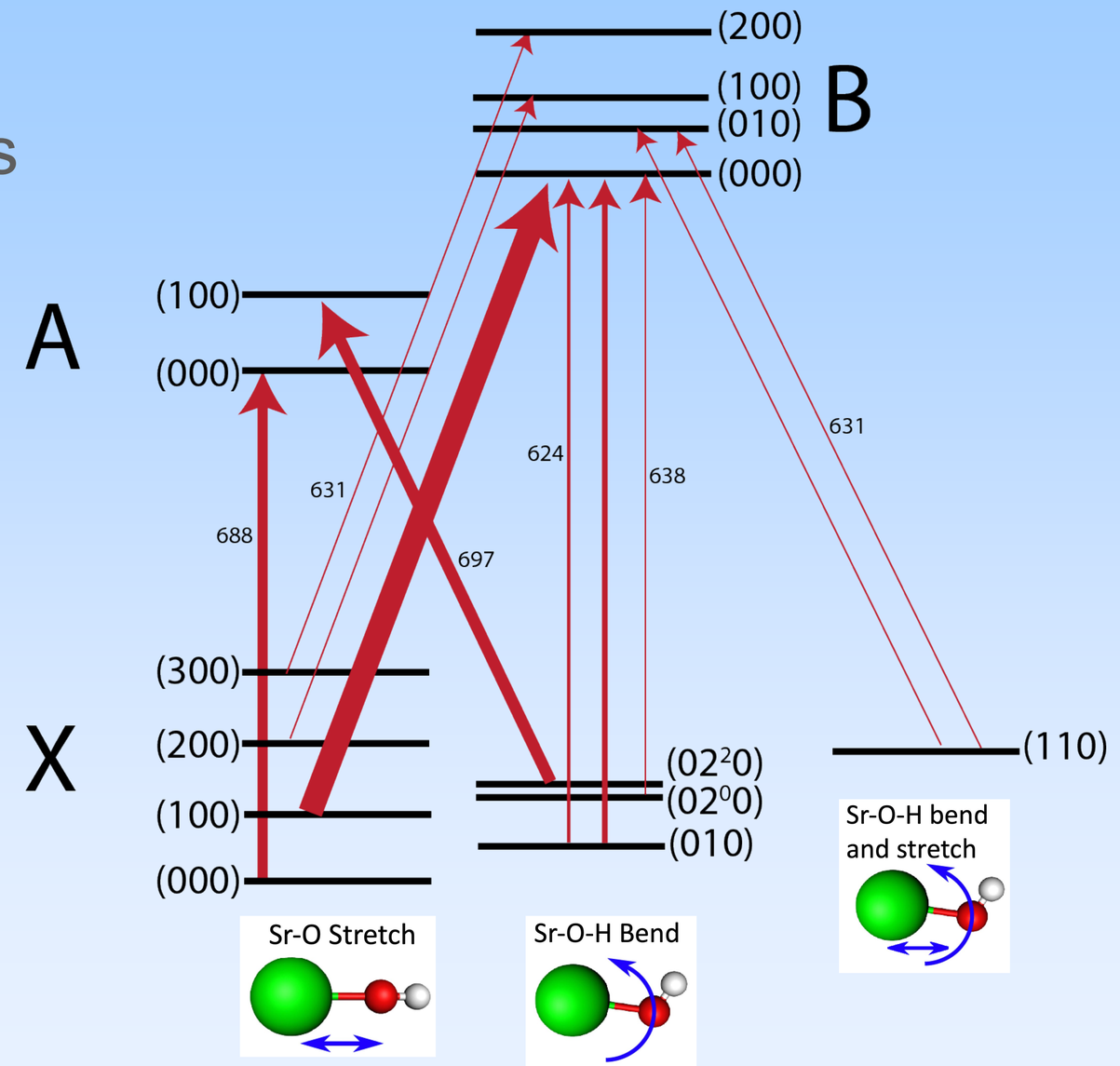
Scheme for Full 3-D Laser Cooling of SrOH

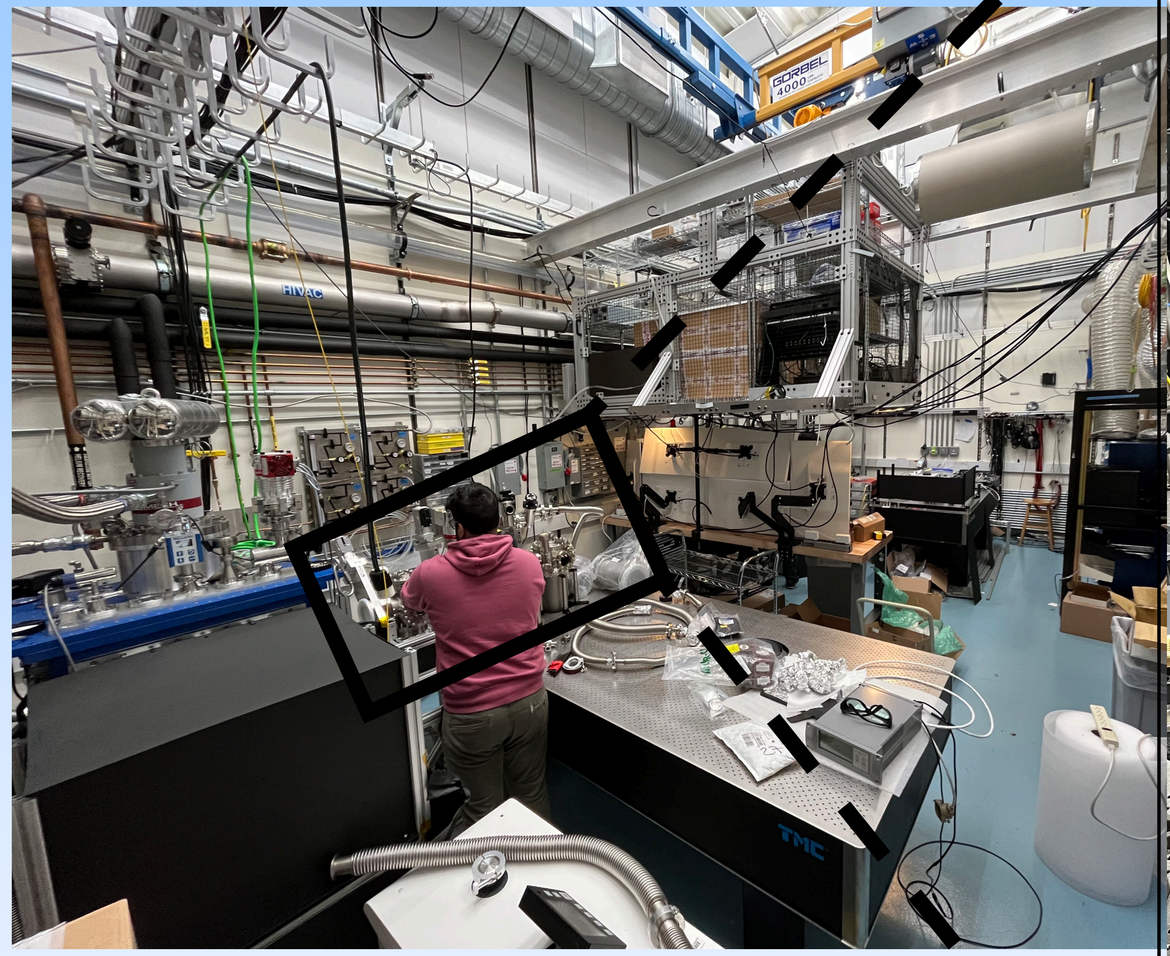
1-D laser cooling. ~200 photons

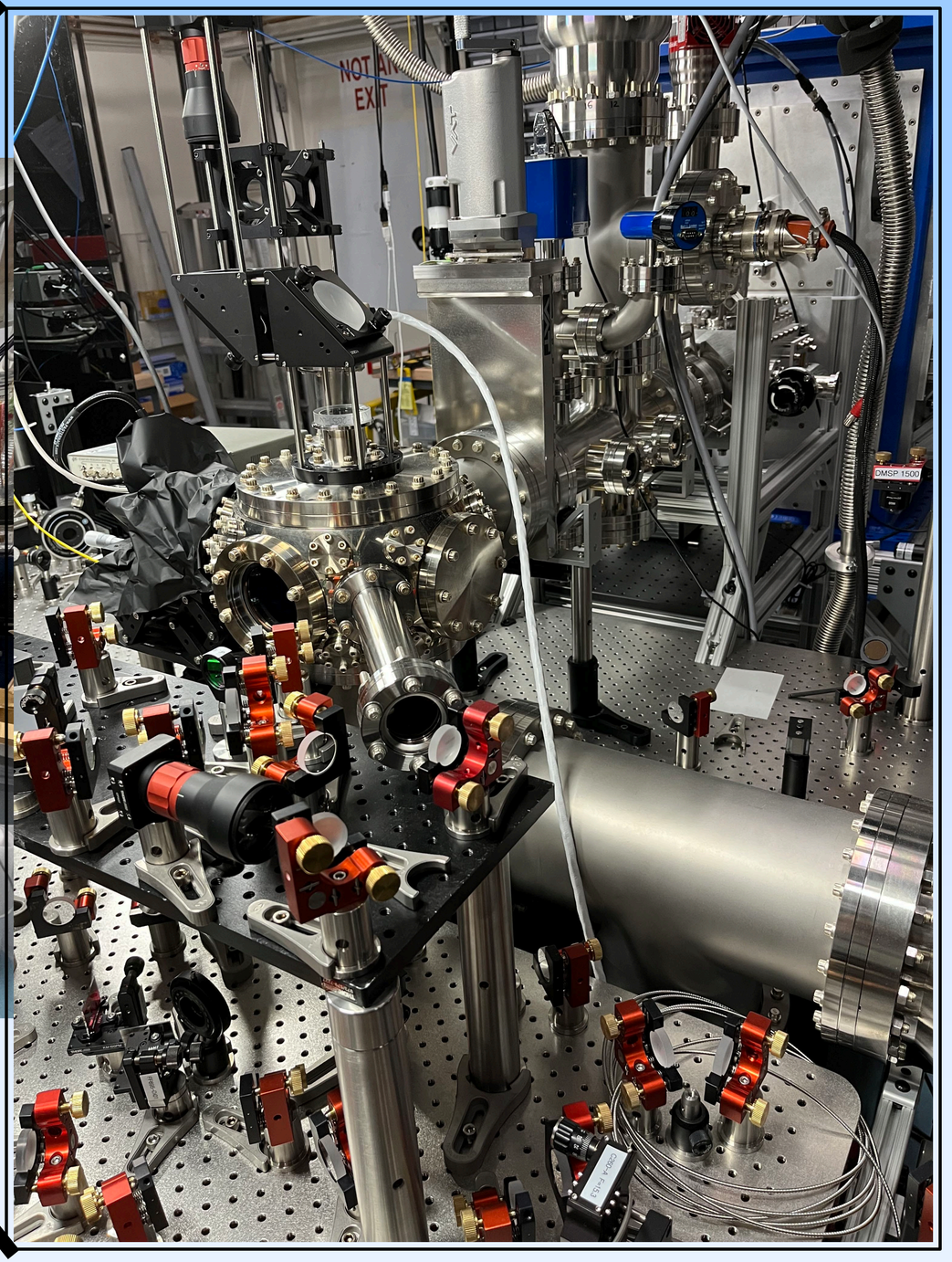
3-D laser cooling. ~20000 photons

Number Of Photons γ

Gnd	Exc	λ	
$X(000)$	$A(000)$	688 nm	20
$X(100)$	$B(000)$	631 nm	
$X(02^20)$	$A(100)$	697 nm	200
$X(010), N=2$	$B(100)$	625 nm	<10 W
$X(010), N=1$	$B(000)$	624 nm	
$X(02^00)$	$B(000)$	638 nm	<1 W
$X(200)$	$B(100)$	630 nm	2000
$X(300)$	$B(200)$	630 nm	
$X(110), N=1$	$B(010)$	630 nm	
$X(110), N=2$	$B(010)$	630 nm	<100 mW
			20000

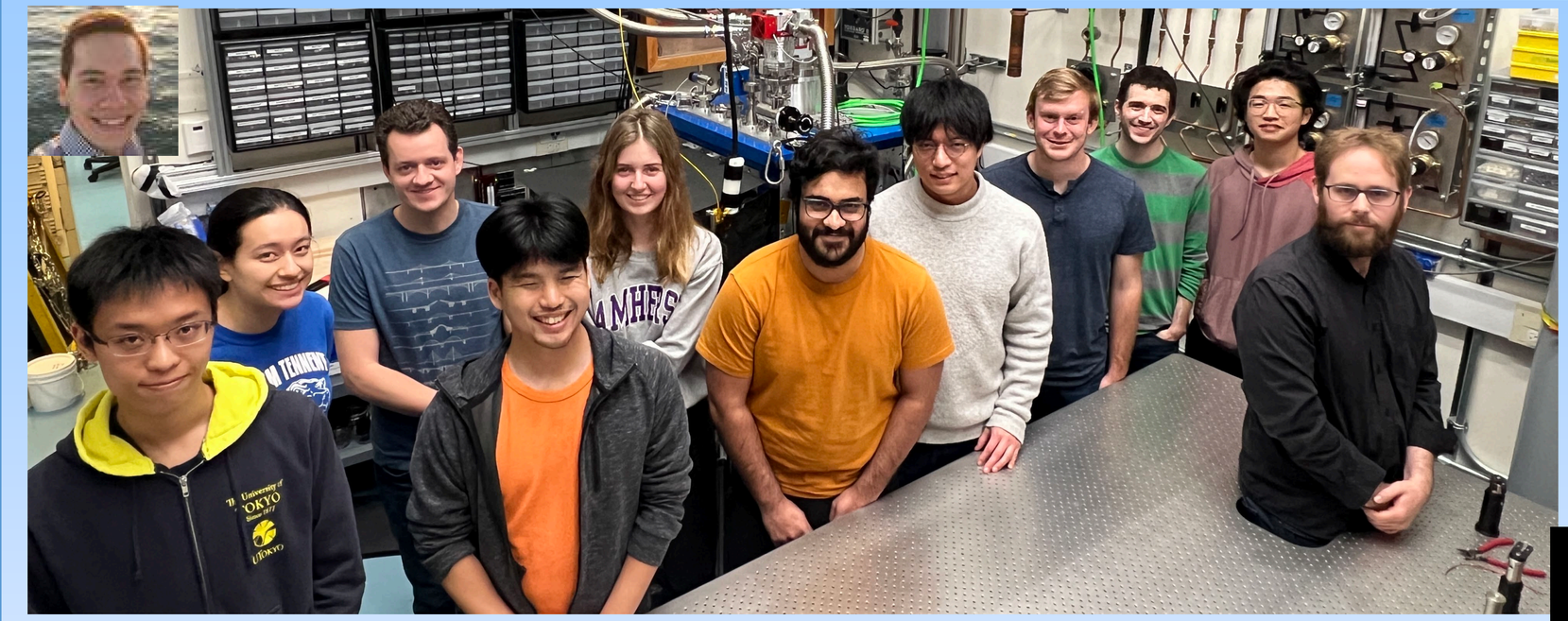






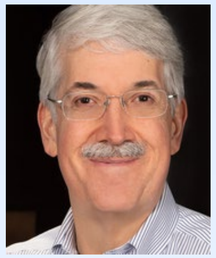
Our SrOH lab
~now

SrOH EDM and Dark Matter Team

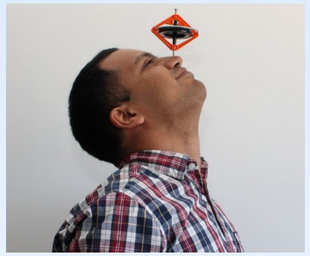


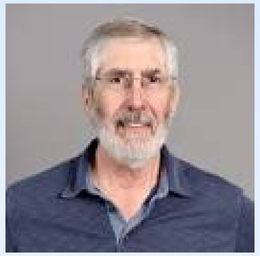
Andrew Winnicki, Tasuku Ono, Hana Lampson, Loic Anderegg, Hiromitsu Sawaoka, Annika Lunstad, Abdullah Nasir, Takashi Sakamoto, Alexander Frenett, Benjamin Augenbraun, Mingda Li, ZL

See Poster Today!

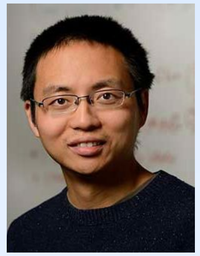


PI
John Doyle





PolyEDM collaborators
Nick Hutzler, Amar Vutha, Tim Steimle



VBR theory
Lan Cheng

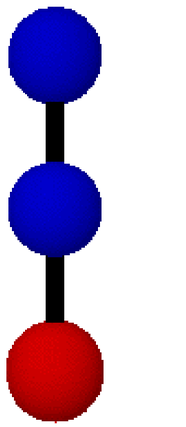
Q-SEnSE
An NSF Quantum Leap Challenge Institute

GORDON AND BETTY MOORE FOUNDATION

ALFRED P. SLOAN FOUNDATION 1934

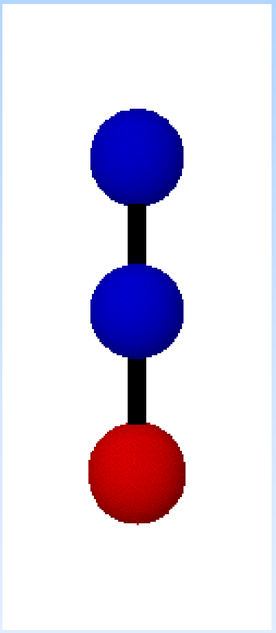
HEISING-SIMONS FOUNDATION

3-D *Laser Cooling of Triatomic Molecules*



Why are we so confident that it will all work for SrOH?

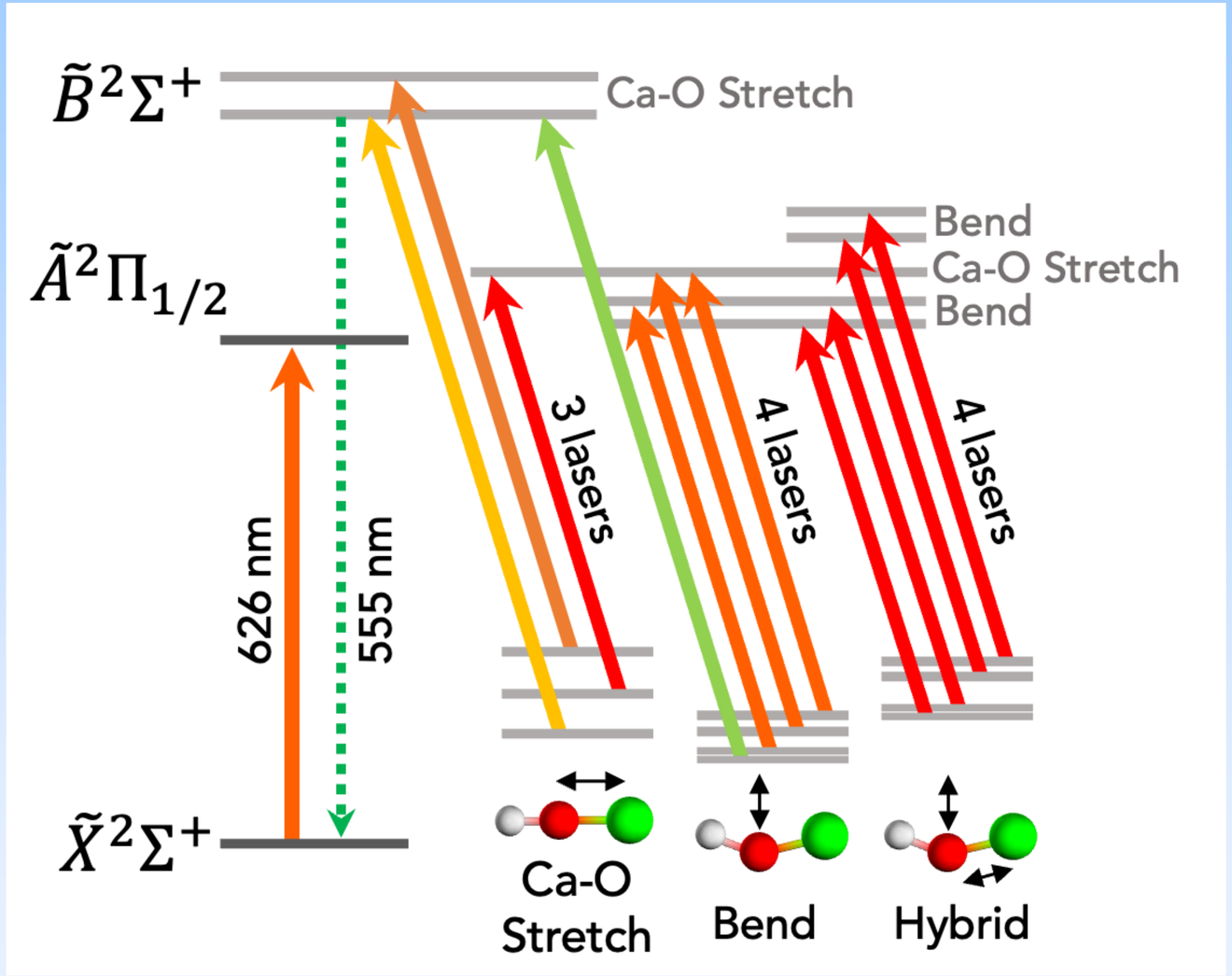
3-D *Laser Cooling of Triatomic Molecules*



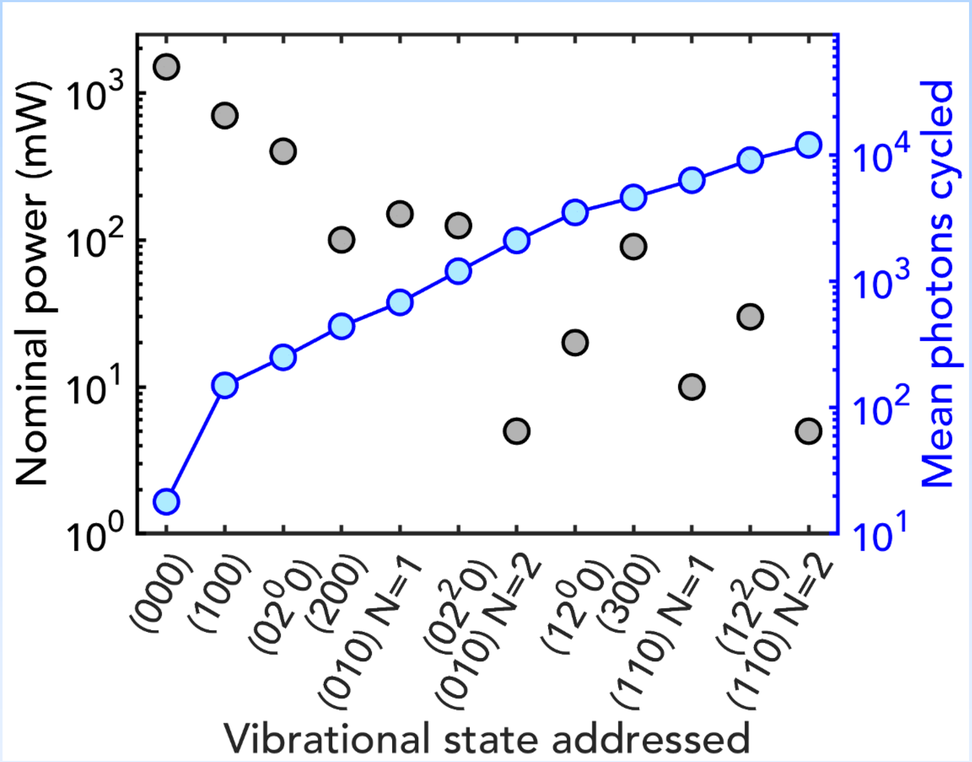
Why are we so confident that it will all work for SrOH?

We have done it for CaOH.

3-D Laser Cooling of Triatomic Molecules CaOH



We Cycle >10,000 photons per molecule!

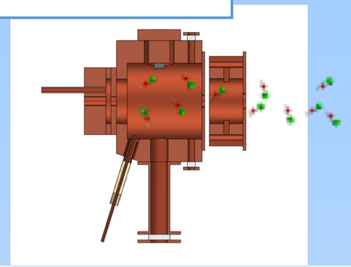


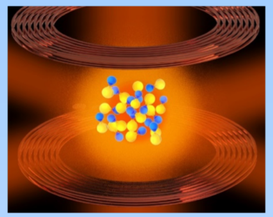
Magneto-Optical Trapping and Sub-Doppler Cooling of a Polyatomic Molecule N. B. Vilas, C. Hallas, L. Anderegg, P. Robichaud, A. Winnic

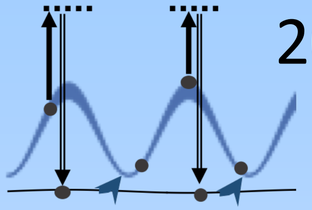
3-D Laser Cooling and Optical Trapping CaOH

2021 -2022

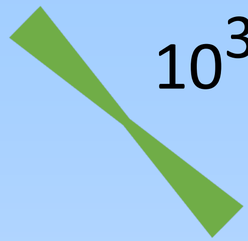
Experimental sequence for optical trapping of CaOH molecules…





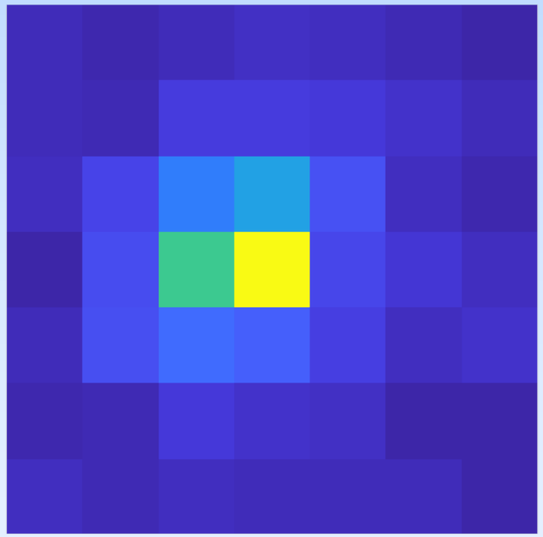


20 uK



10^3 molecules

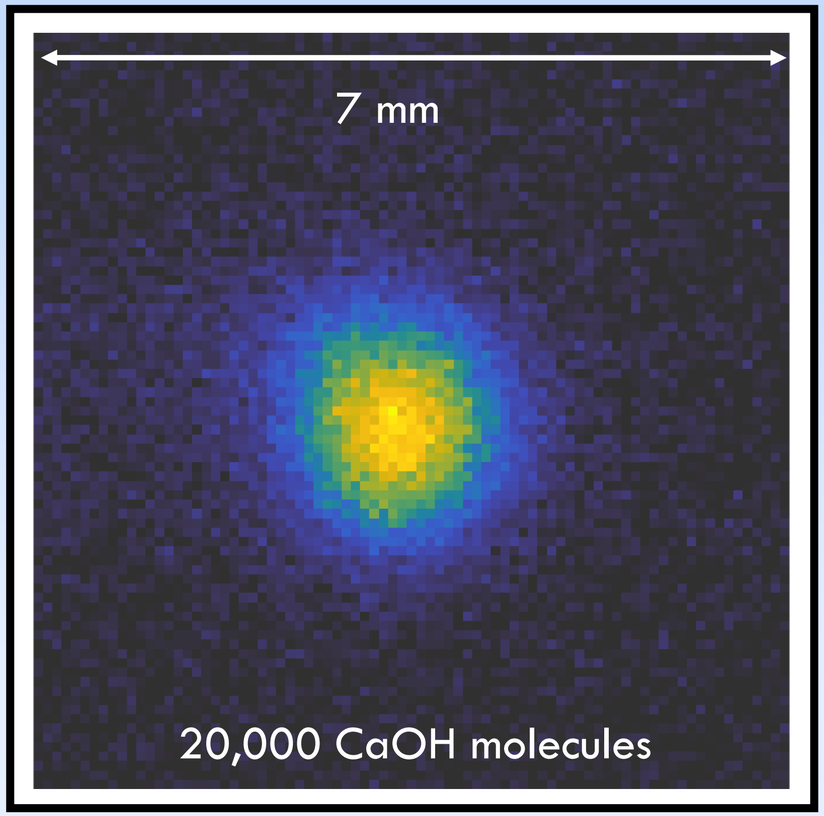
Ablation Produce CaOH	**Slowing and MOT**	**Sub-Doppler cooling**	**Optical trapping**



T ~ 40 uK
In an ODT (Optical Dipole Trap)

EMCCD image of optically trapped CaOH molecules

"Single Shot" image

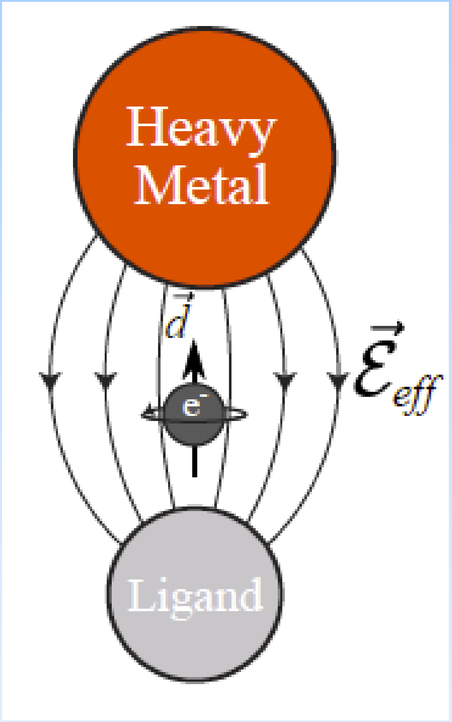


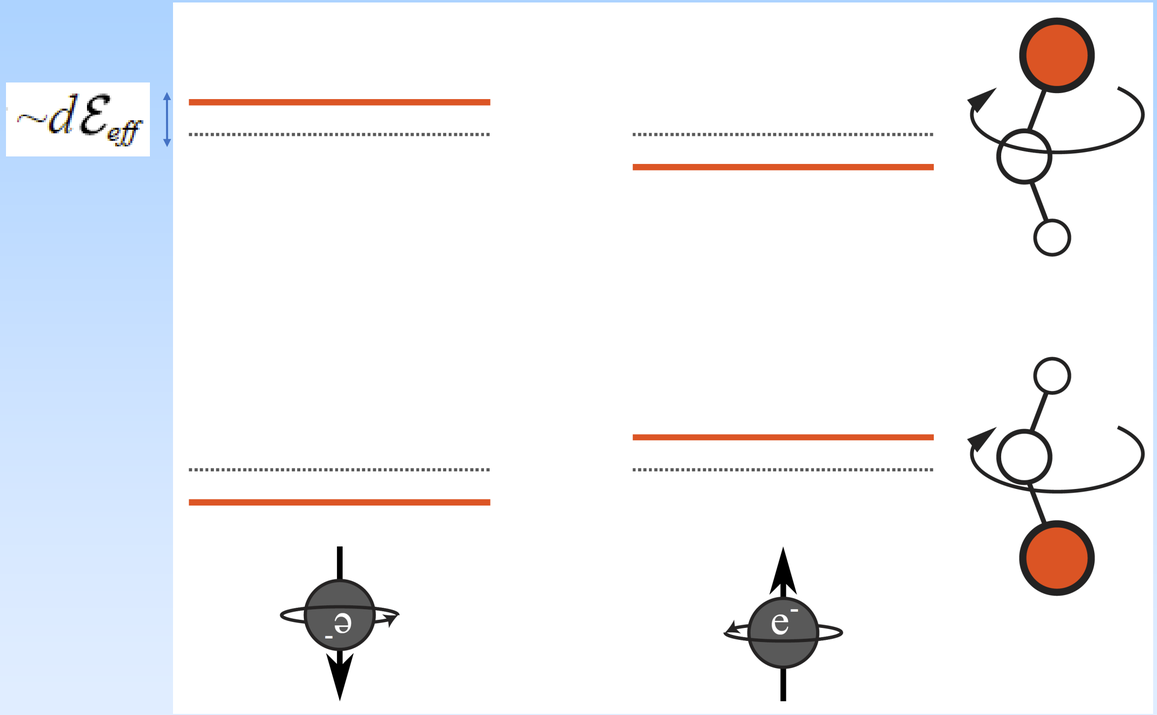
T ~ 1 mK
In a MOT (Magneto-Optical Trap)

NSF and AFOSR

Pathfinder Experiment to EDM CaOH

- Demonstrate the principles of a polyatomic eEDM search using trapped CaOH





Caltech Collaborators

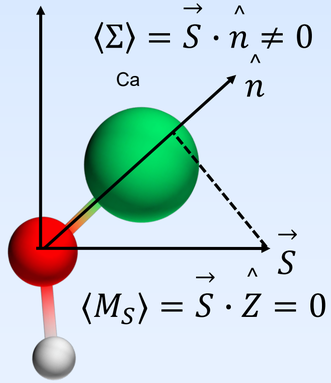
Arian Jadbabaie

Nick Hutzler

PolyEDM

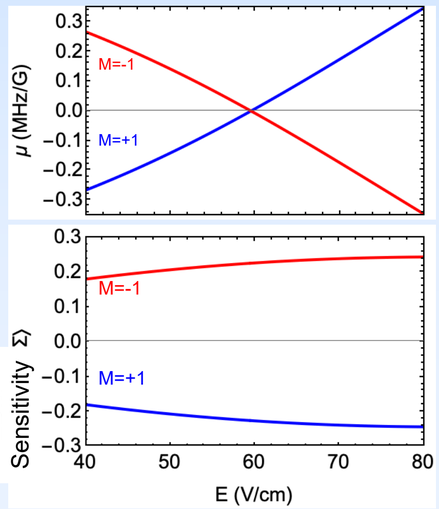
Opposite EDM Shift

$\hat{Z}$
$\vec{E}, \vec{B}, \vec{\epsilon}_{ODT}$



- Application of a lab E_Z results in states with "zero" electron g-factor
  - Still sensitive to EDM interaction!
- Useful for rejecting systematic errors from stray B_Z fields

Zero g-factor EDM states

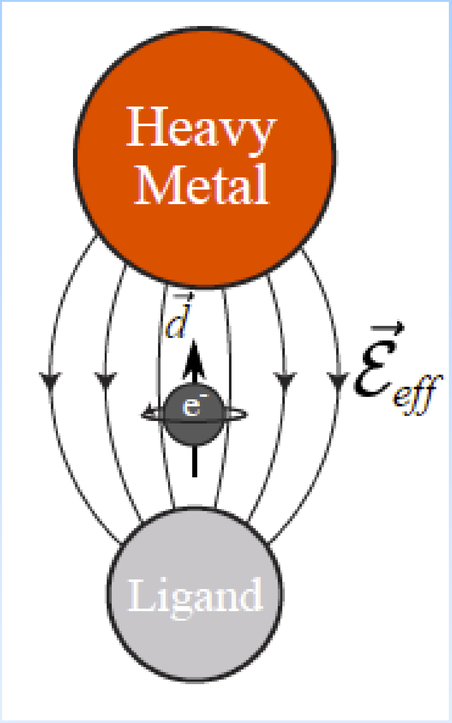


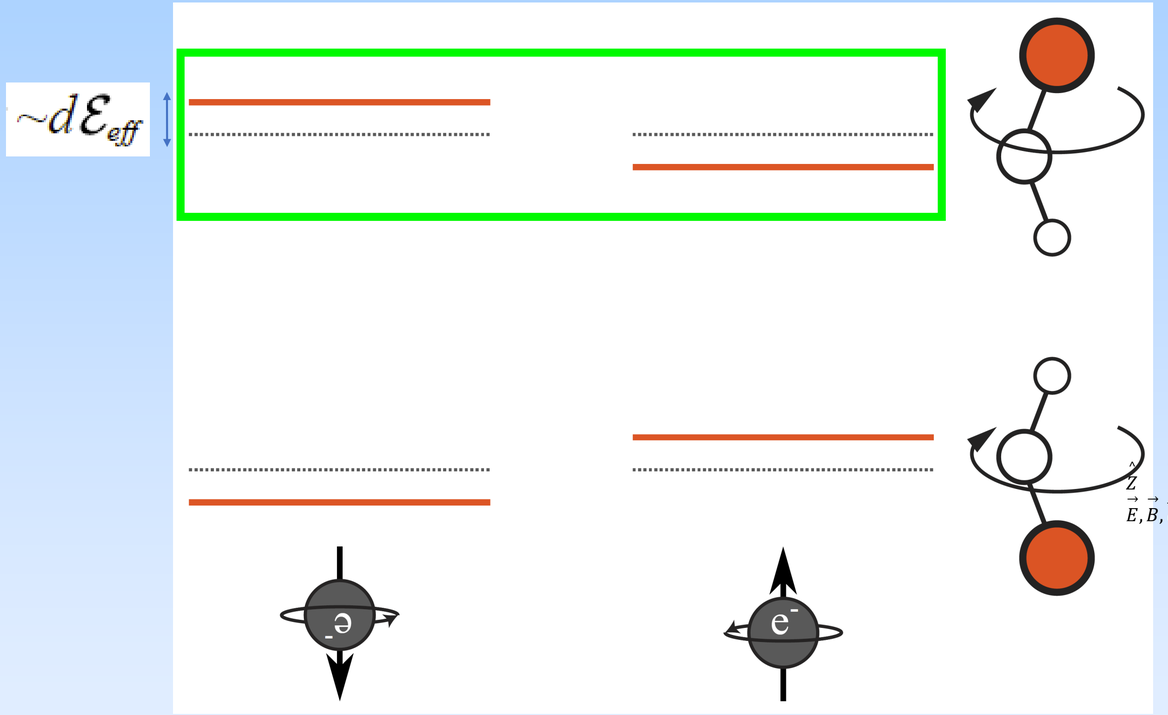
Aligned "comagnetometer" states have opposite EDM shifts, aiding systematic error rejection

Pathfinder Experiment to EDM CaOH

- Demonstrate the principles of a polyatomic eEDM search using trapped CaOH

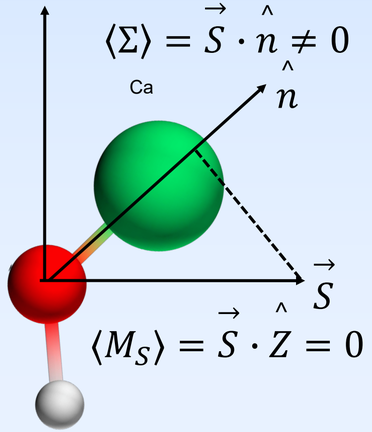
PolyEDM



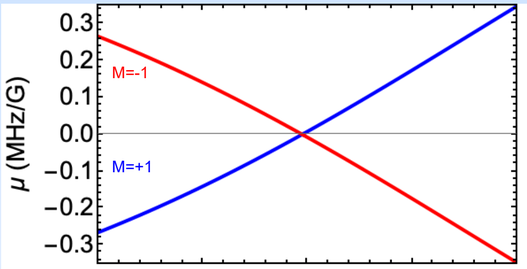


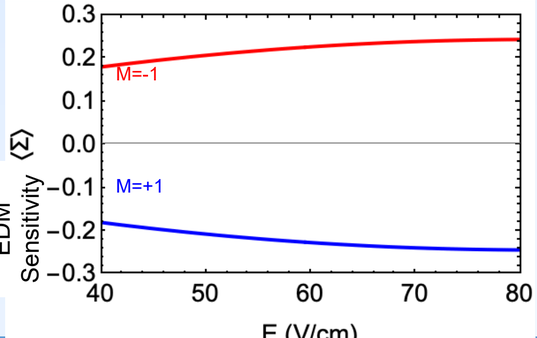
Opposite EDM Shift

$\vec{E}, \vec{B}, \vec{\epsilon}_{ODT}$



Zero g-factor EDM states



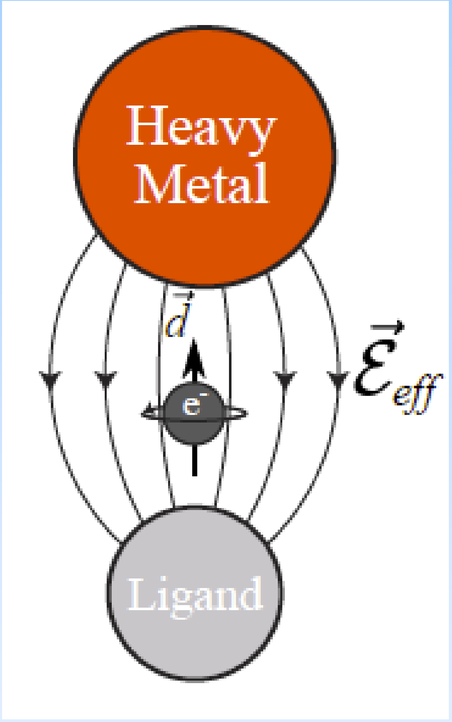


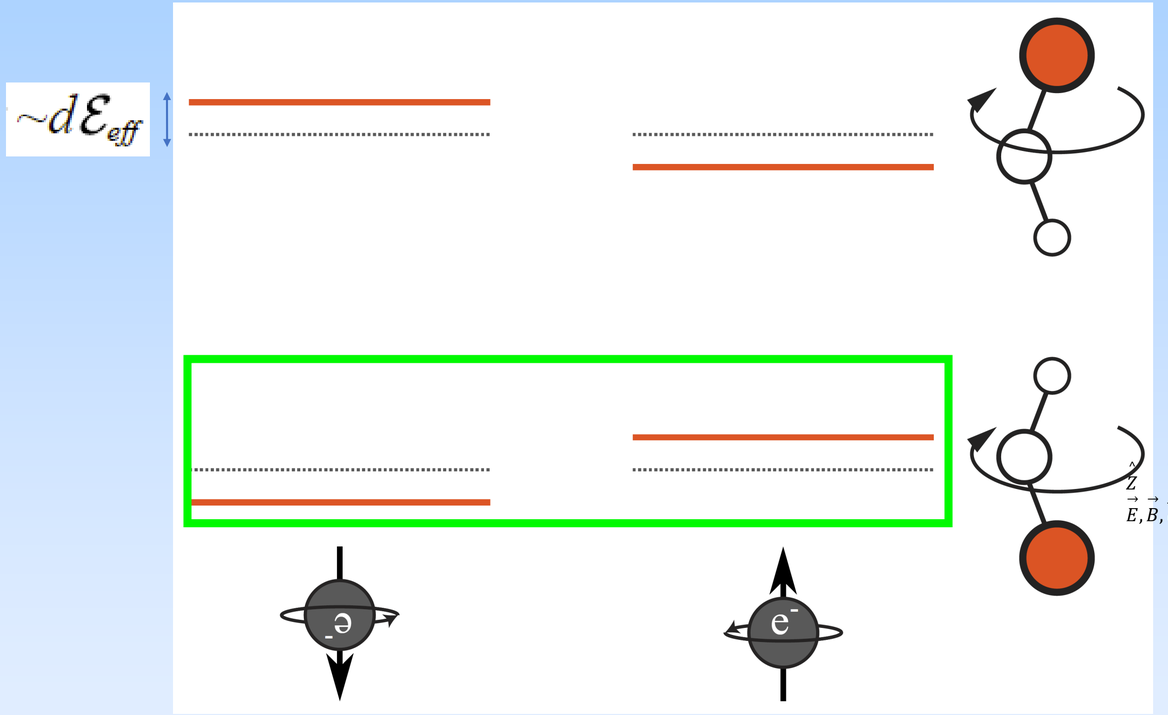
Aligned "comagnetometer" states have opposite EDM shifts, aiding systematic error rejection

Pathfinder Experiment to EDM CaOH

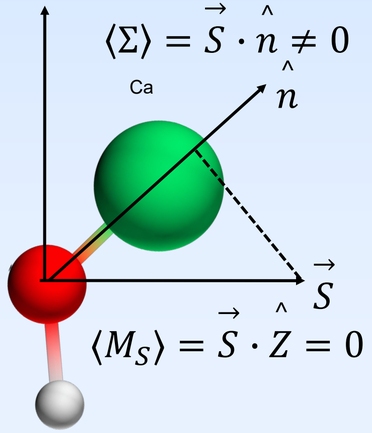
- Demonstrate the principles of a polyatomic eEDM search using trapped CaOH

PolyEDM

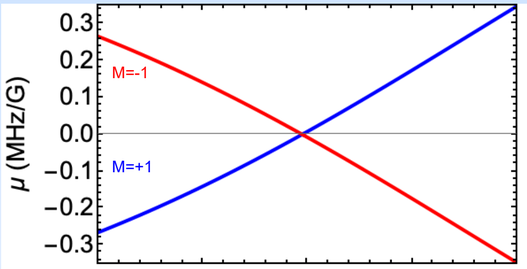


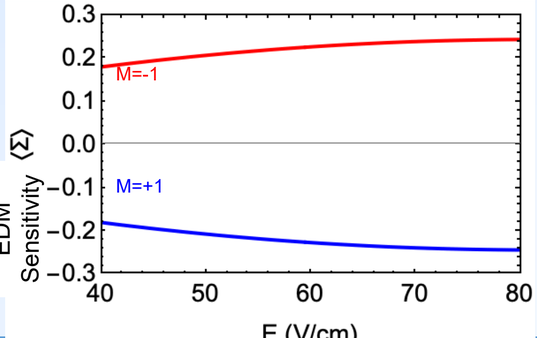


Opposite EDM Shift



Zero g-factor EDM states



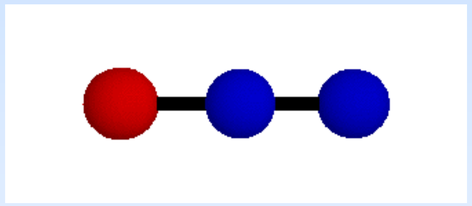


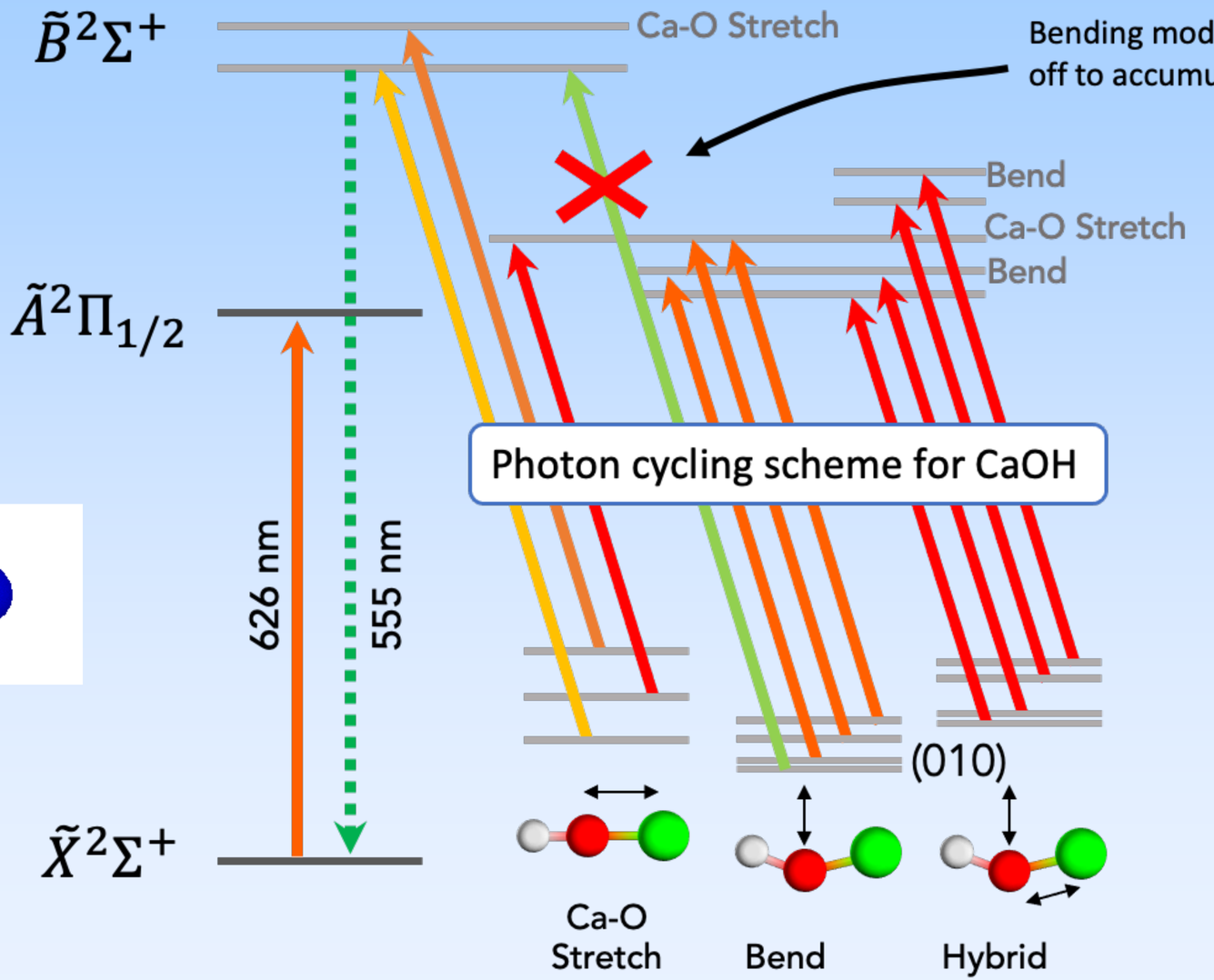
Aligned "comagnetometer" states have opposite EDM shifts, aiding systematic error rejection

Optically Pump into the Bending Mode (010) CaOH

April 2022

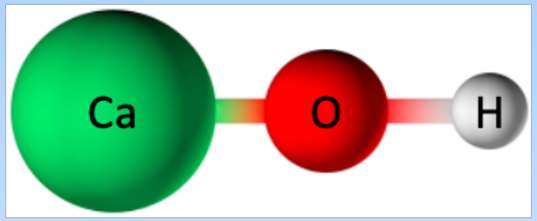
CaOH molecules trapped in ODT, then transferred to bending mode via optical pumping



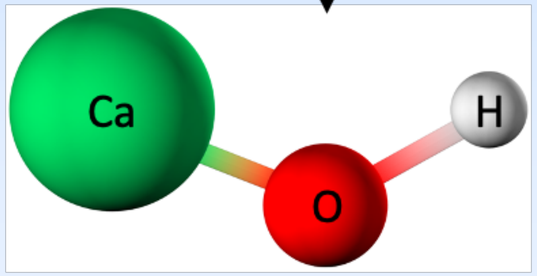


Bending mode repumper turned off to accumulate population

Photon cycling scheme for CaOH

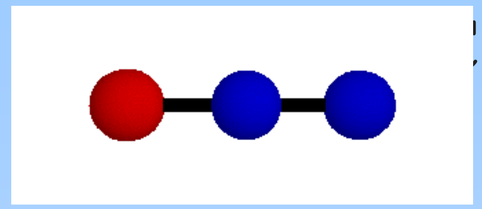


Optical pumping transfer with >90% efficiency

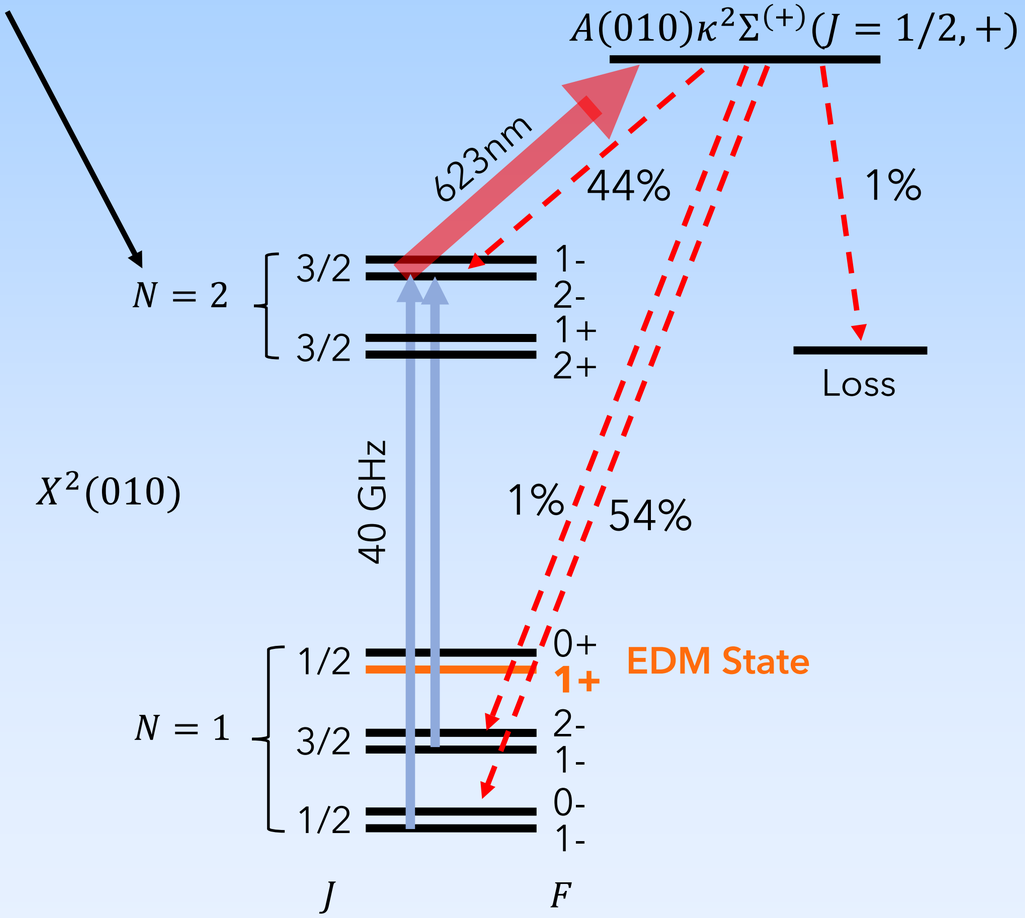


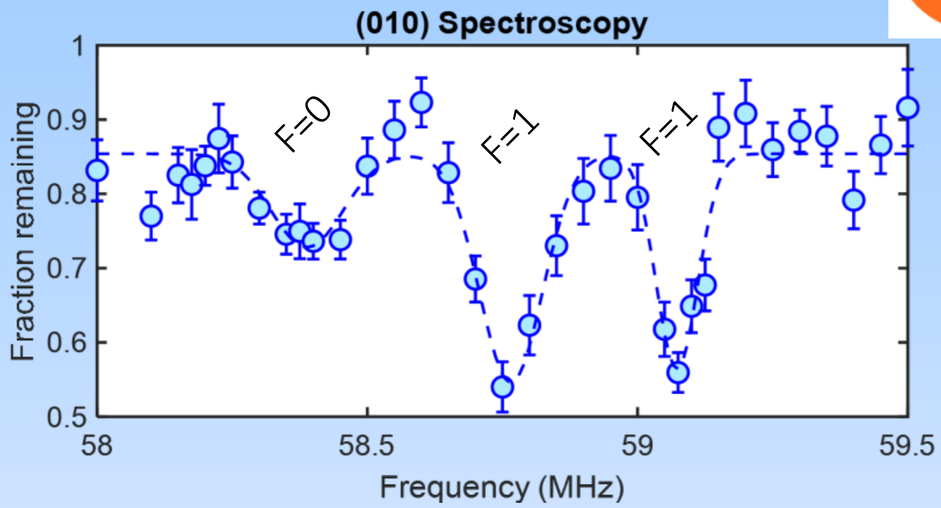
NSF and AFOSR

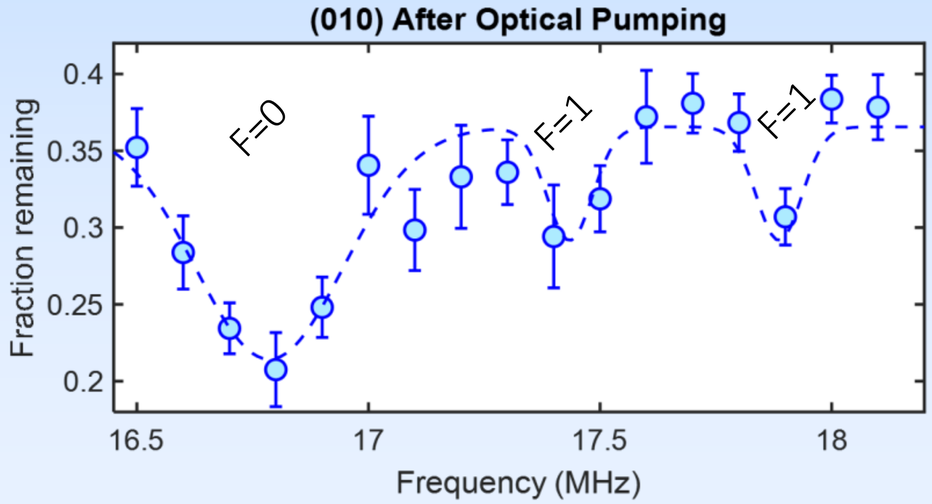
Single Quantum State Control Bending Mode (010) CaOH



N is Rotational Quantum Number





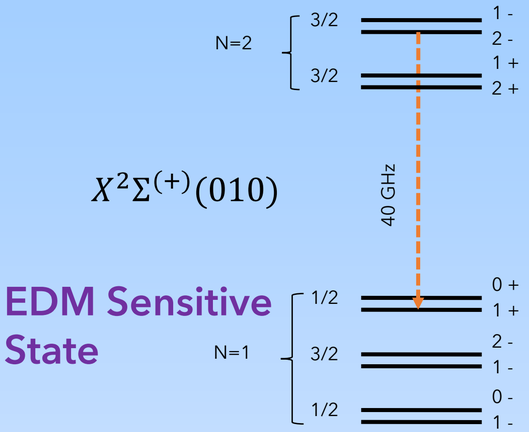


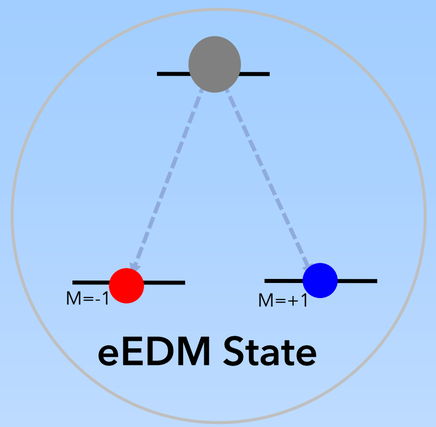
PolyEDM

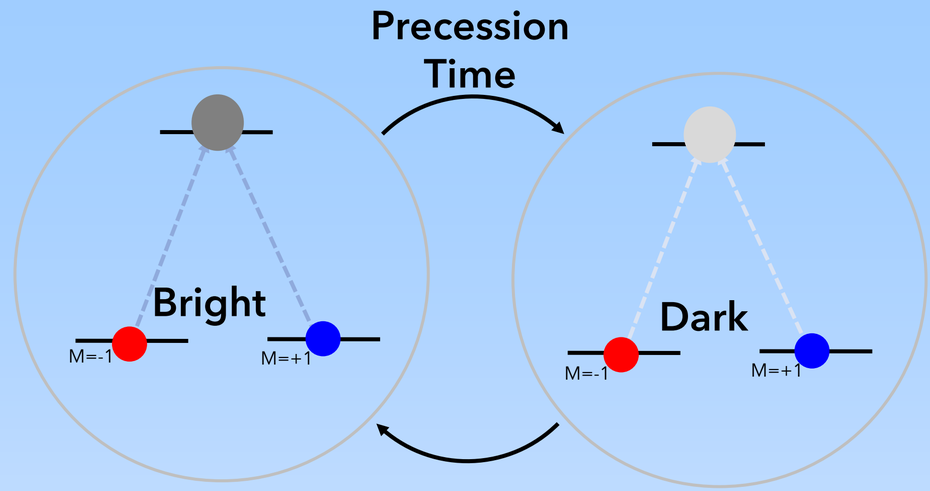
CaOH @ Harvard

Bending Mode EDM State

Electron Spin Precession in the EDM Sta



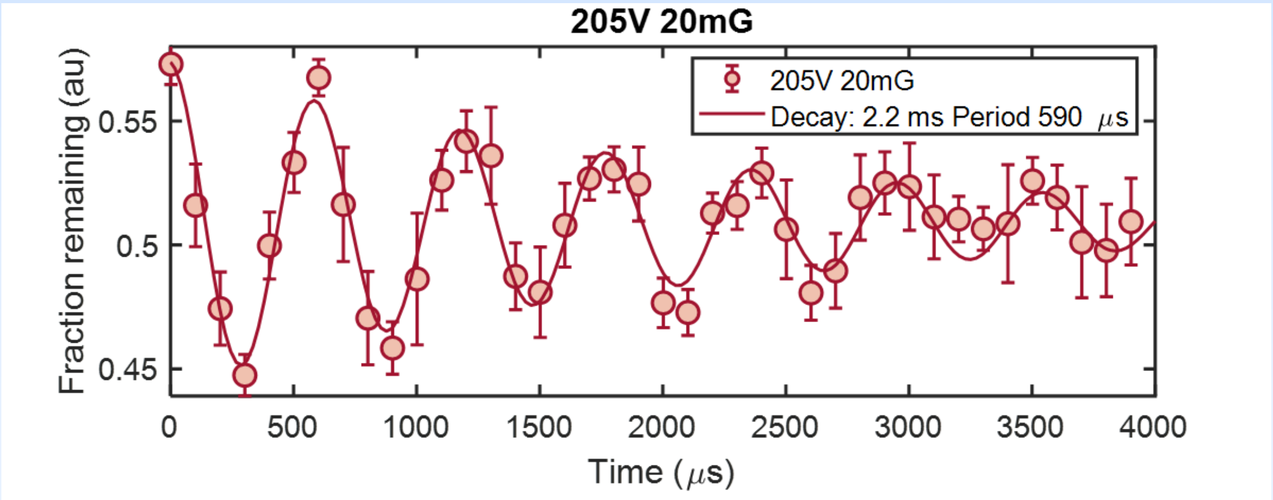


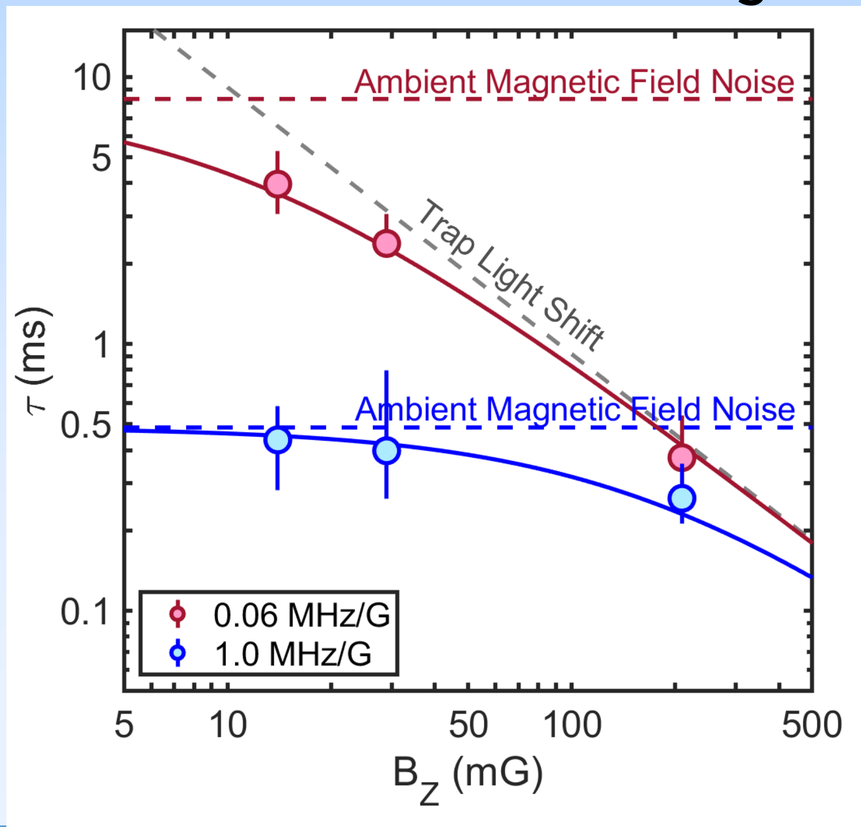


PolyEDM

Increased Coherence Near Zero g-factor

eEDM Sensitive Spin Precession



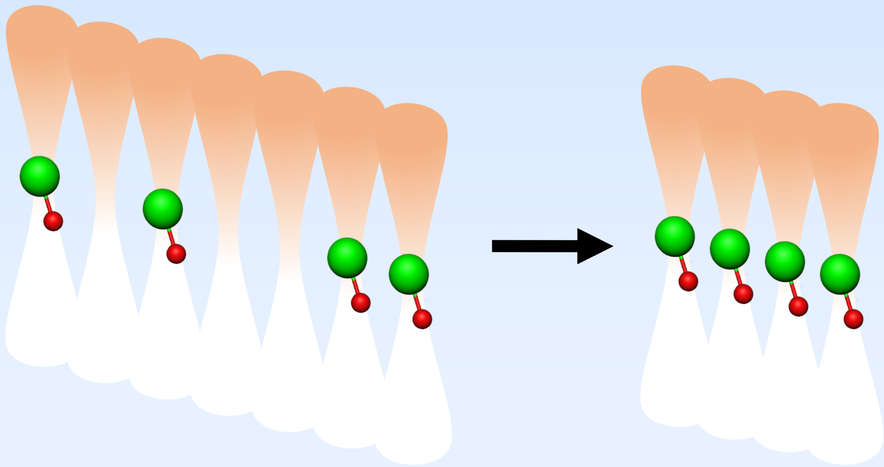


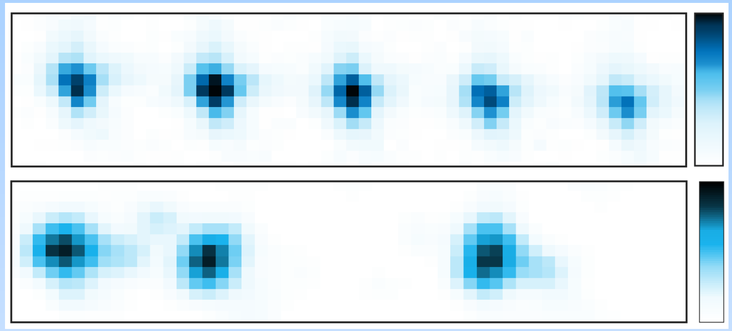
Optical Tweezer Arrays

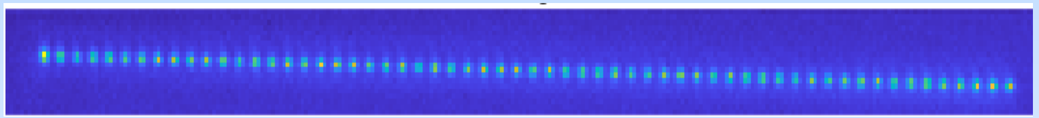
Molecules in Tweezers

But wait, there's more with CaOH!

Rearrangeable Optical Tweezer Arrays

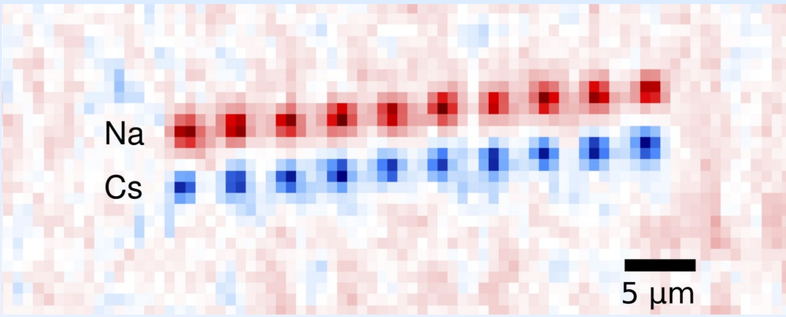






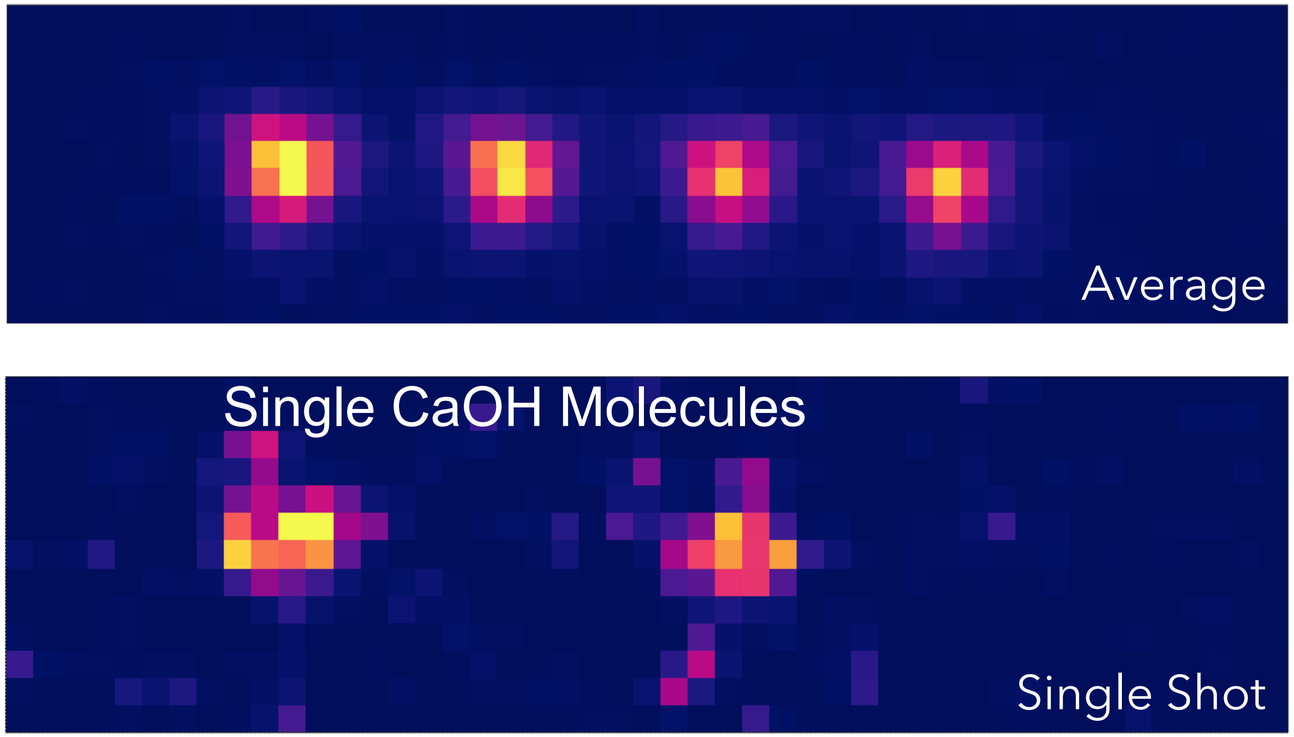
CaF - Harvard - Doyle Group

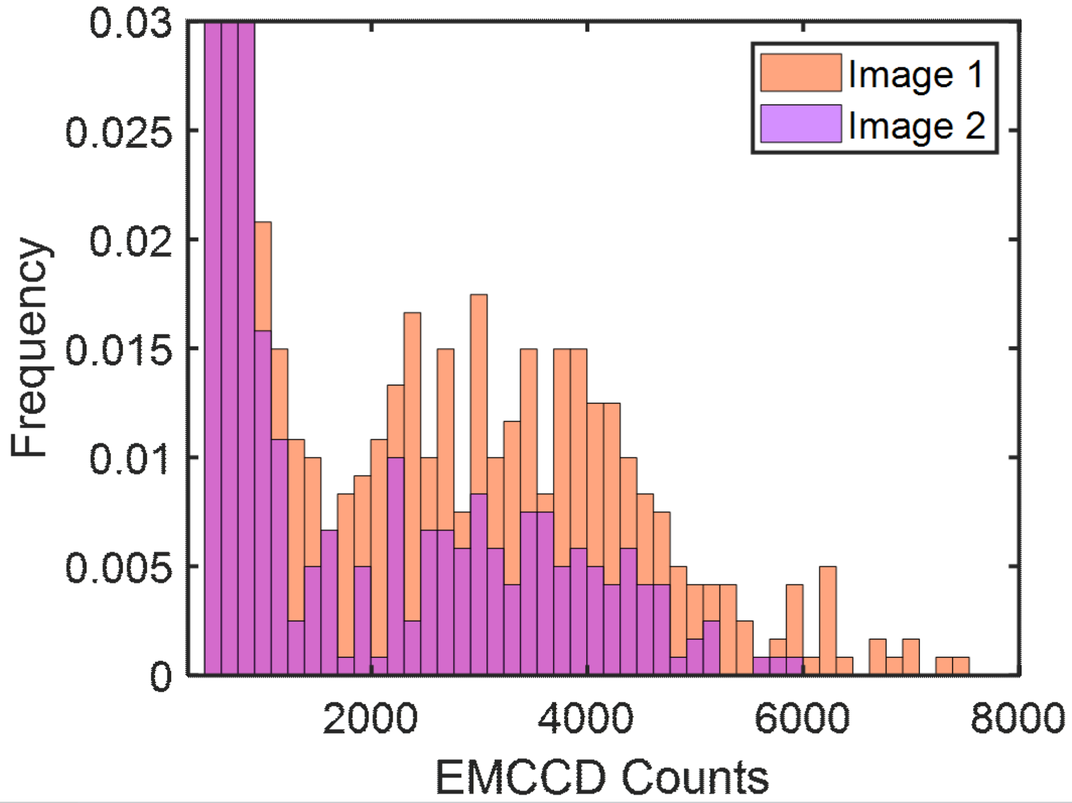
CaF - Princeton - Cheuk Group



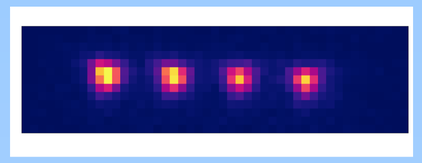
NaCs - Harvard - Ni Group

Optical Tweezer Array of CaOH Molecules





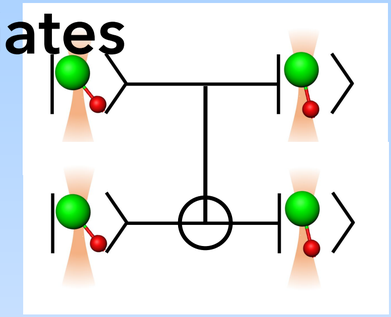
May 2023 unpublished
DAMOP (June 2023)



Single CaOH molecules
in an Optical Tweezer Array

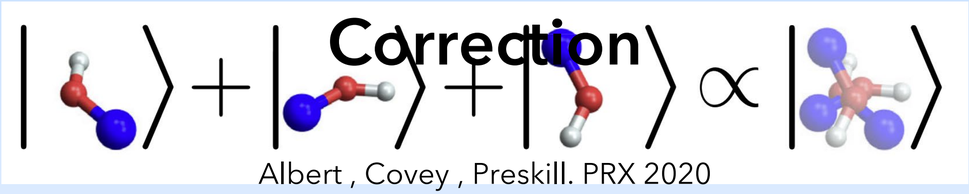
CaOH (and other Triatomic Molecules) Outlook

Two Qubit Gates



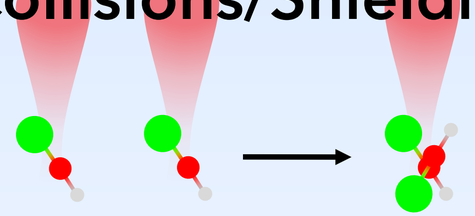
Quantum Simulation

Error Correction

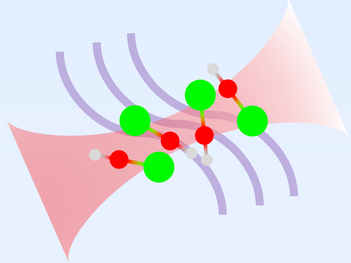


Albert , Covey , Preskill. PRX 2020

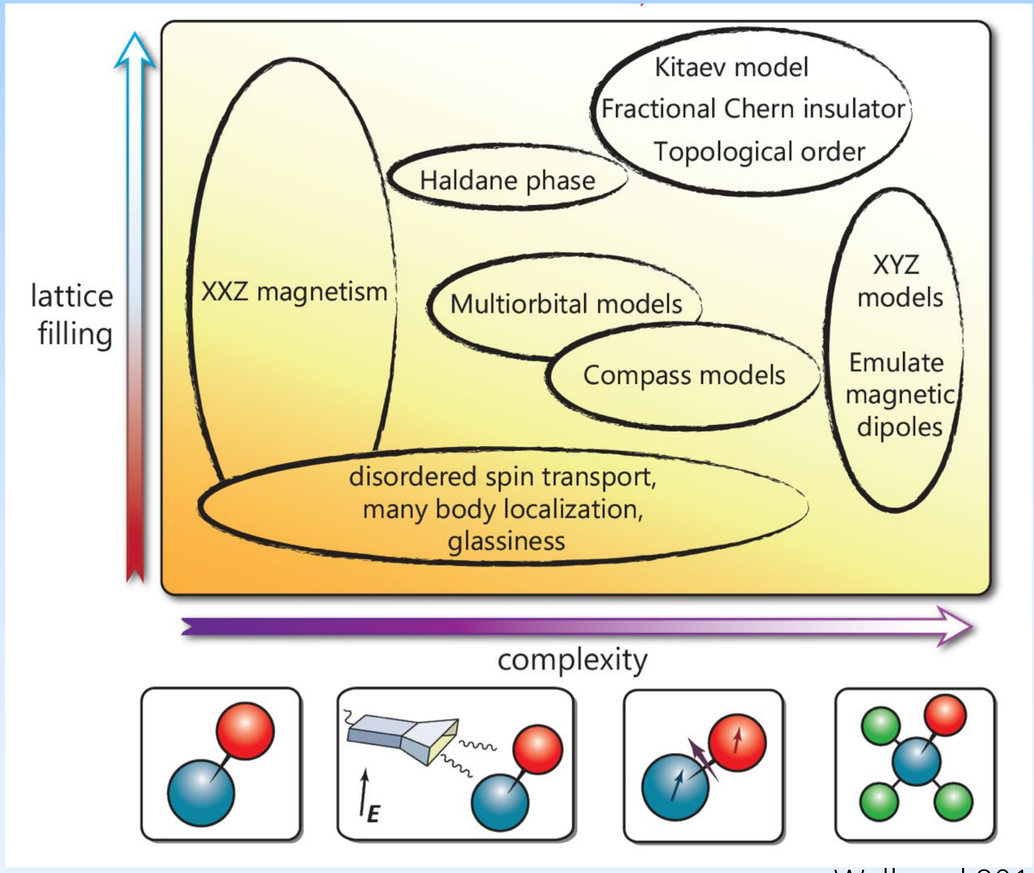
Collisions/Shielding



BSM Physics Searches



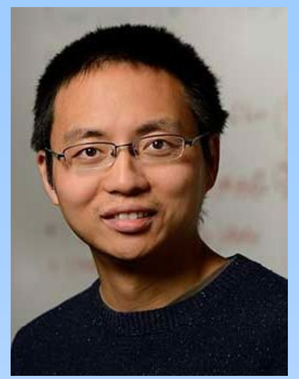
Complexity Adds Possibilities



Wall et al 2014

CaOH Acknowledgments

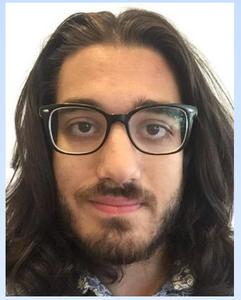
Theory Collaborators



Lan Cheng Chaoqun Zhang

Johns Hopkins University

EDM Pathfinder Collaborators



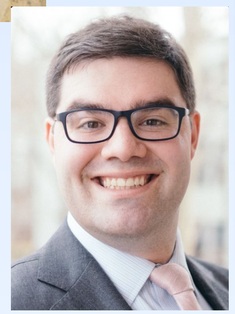
Arian Jadbabaie

Nick Hutzler

Nathaniel Vilas
Christian Hallas
Paige Robichaud

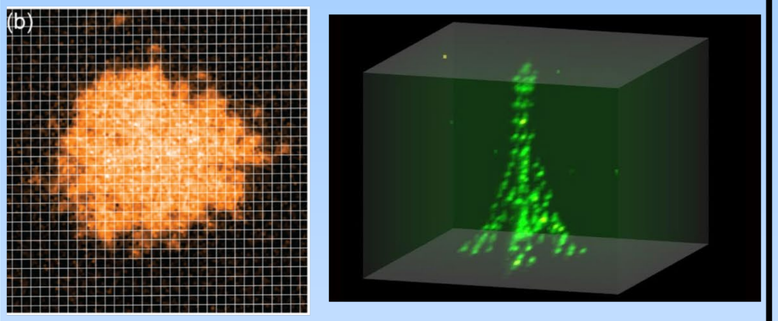
Andrew Winnicki
Loic Anderegg
JMD

Former:
Louis Baum
Debayan Mitra



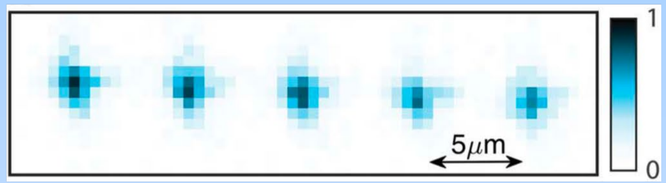
The Complexity Theme

Atoms (N=1)



Cheuk, …, Zwierlein PRL **116** (2016)
Barredo, …, Browaeys Nature **561** (2018)

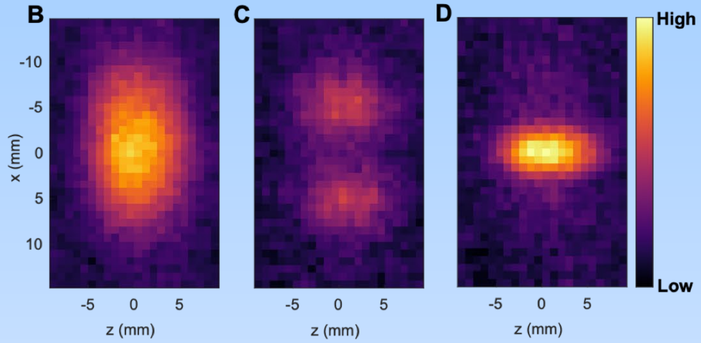
Diatomic Molecules (N=2)



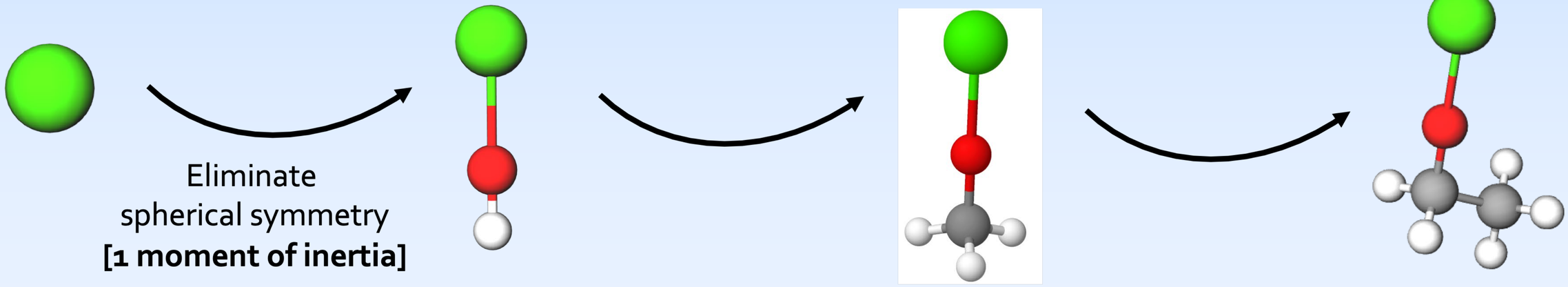
Anderegg, Cheuk, …, Doyle Science **365** (2019)

Harvard, Yale, Imperial, Boulder,
Columbia, UConn, UC Riverside, …

Polyatomic Molecules (N=3,4,5…)



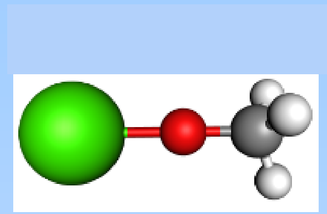
Complexity [reduced molecular symmetry]



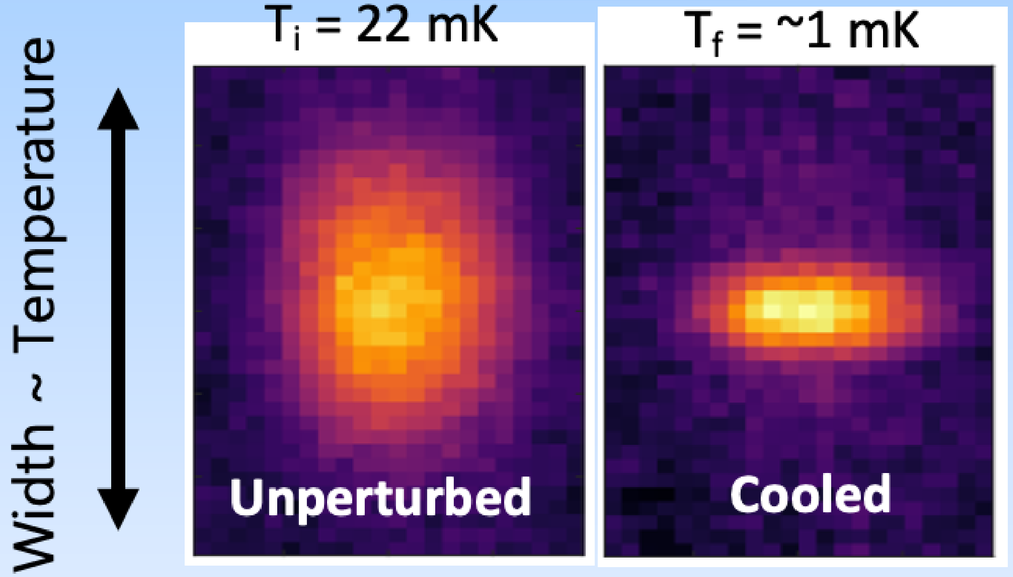
Eliminate
spherical symmetry
[1 moment of inertia]

[Molecular Asymmetry and Optical Cycling: Laser Cooling Asymmetric Top Molecules] B. L. Augenbraun, JMD, T. Zelevinsky, I. Kozyryev, Phys. Rev. X

1-D Laser Cooling of a Symmetric Top Molecule



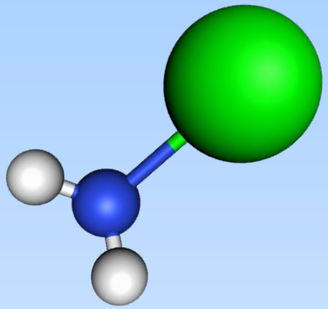
$CaOCH_3$



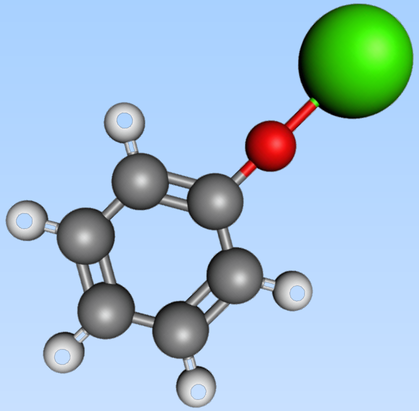
Cooling performance essentially identical to that of simpler linear molecules, despite greatly increased molecular complexity.

Direct Laser Cooling of a Symmetric Top Molecule D. Mitra, N. B. Vilas, C. Hallas, L. Anderegg, B. L. Augenbraun, L. Baum, C. Miller, S. Raval, and JMD. Science 369, 1366 (2020).

Can we cool an ASymmetric Top Molecule?



$CaNH_2$



CaOPh

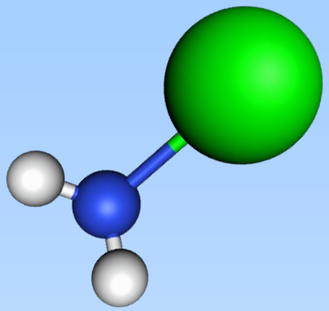
Calcium Phenoxide

Our Theory says Yes!

Molecular Asymmetry and Optical Cycling: Laser Cooling Asymmetric Top Molecules

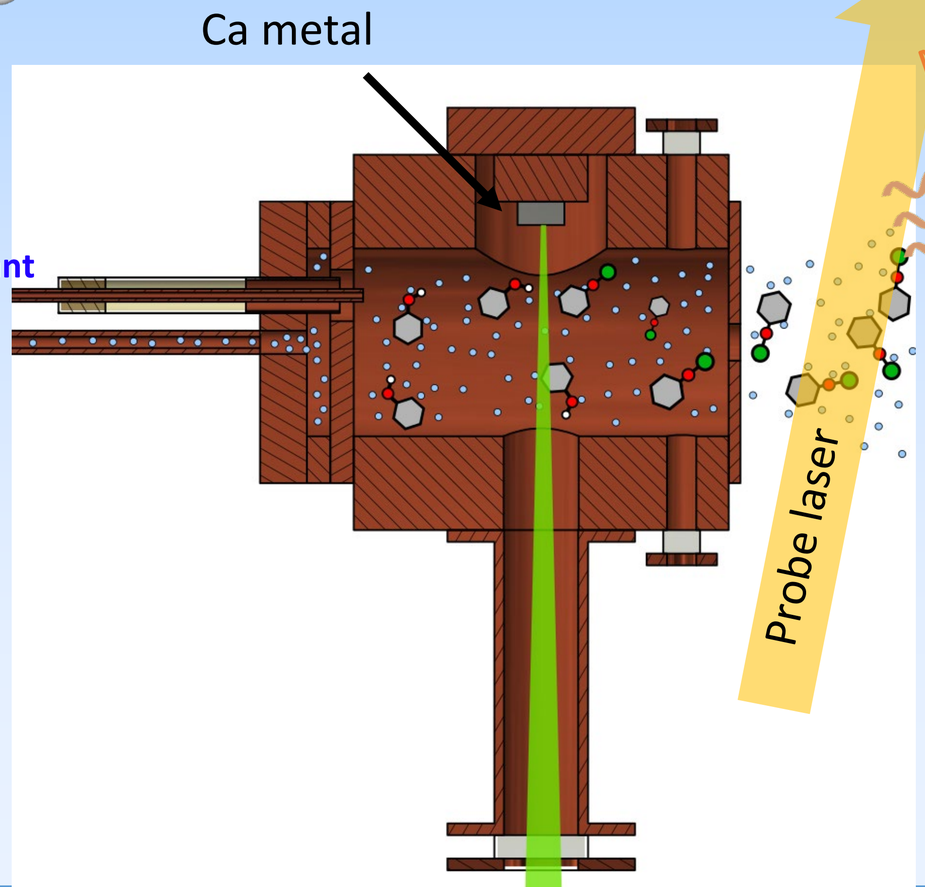
B. L. Augenbraun, J. M. Doyle, T. Zelevinsky, I. Kozyryev, Phys. Rev. X 10, 031022 (2020).

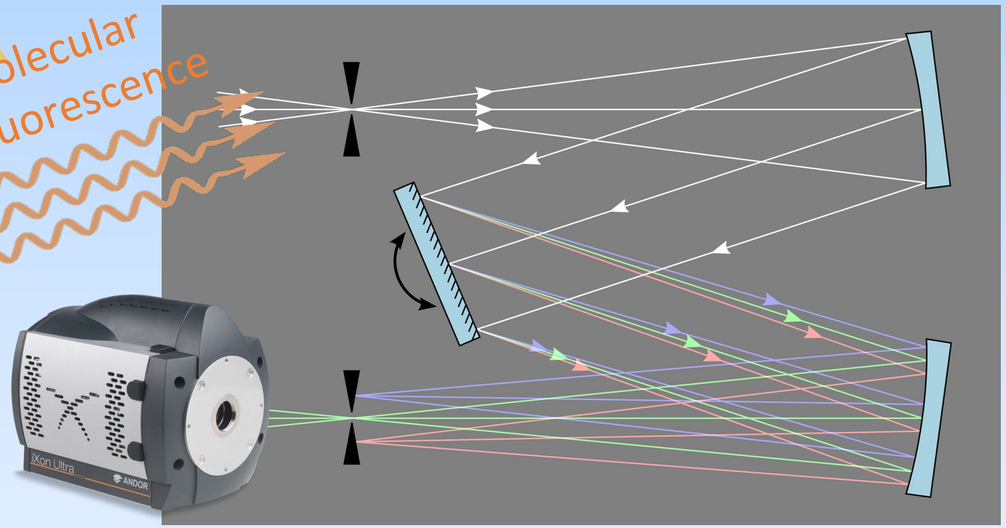
1-D Laser Cooling of an ASymmetric Top Molecule



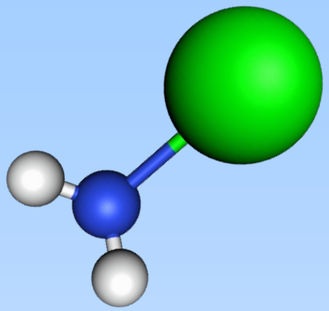
$CaNH_2$

Vibrational Branching Ratio Measurements



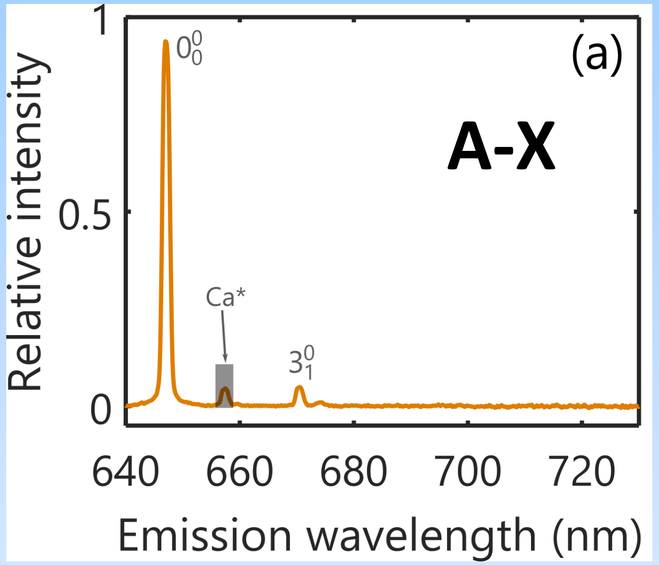


1-D Laser Cooling of an ASymmetric Top Molecule

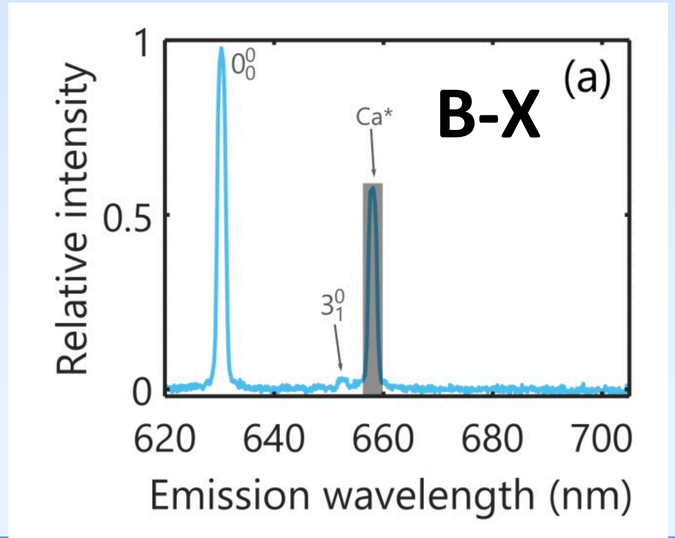


$CaNH_2$

Vibrational Branching Ratio Measurements

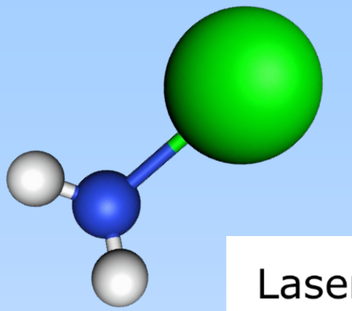


Branch	Measured	Calculated
	95.8%	96.3%
(Ca-N stretch)	4.1%	3.5%
(2x Ca-N stretch)	< 0.5%	0.1%



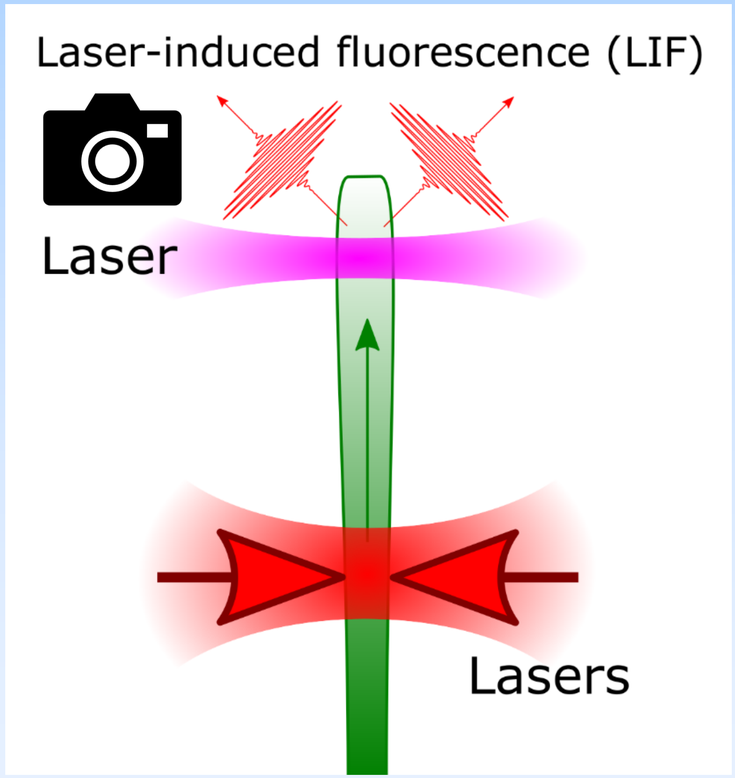
Branch	Measured	Calculated
	97.5%	98.1%
(Ca-N stretch)	2.48%	1.9%
(2x Ca-N stretch)	< 0.1%	0.01%

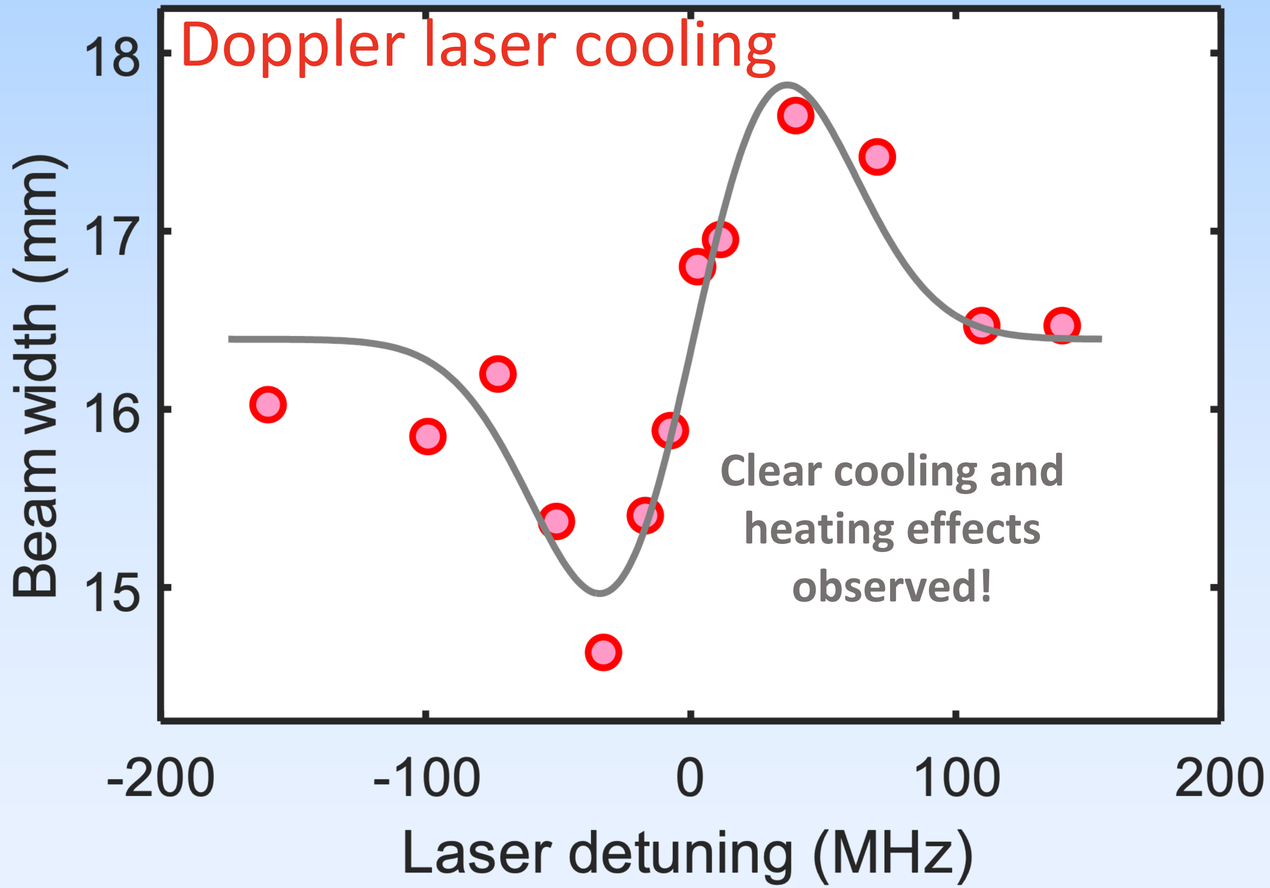
1-D Laser Cooling of an ASymmetric Top Molecule



$CaNH_2$ Following the pioneering lead of SrF, CaF, CaOH, CaOCH3 etc. we have demonstrated…

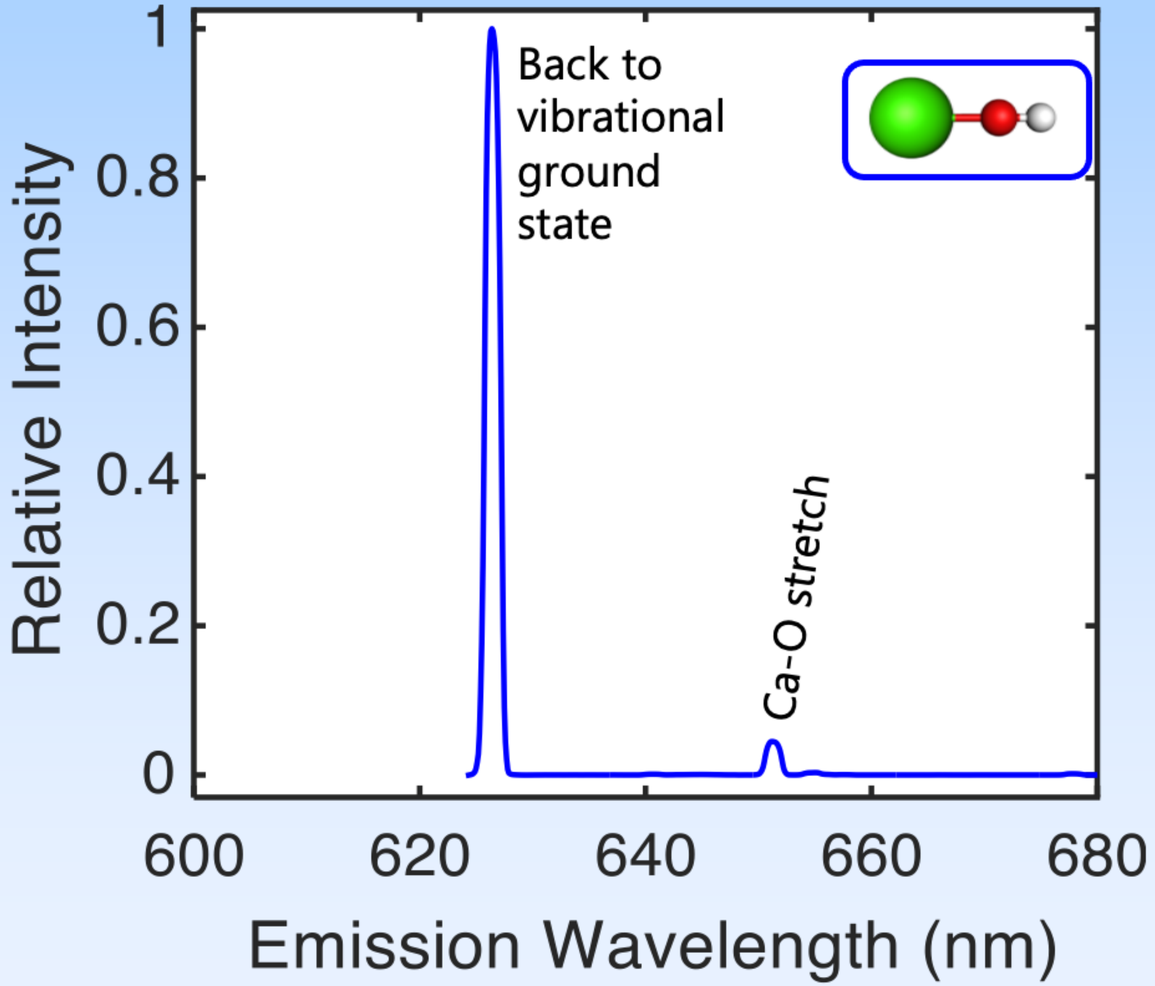
… radiative force deflection and…





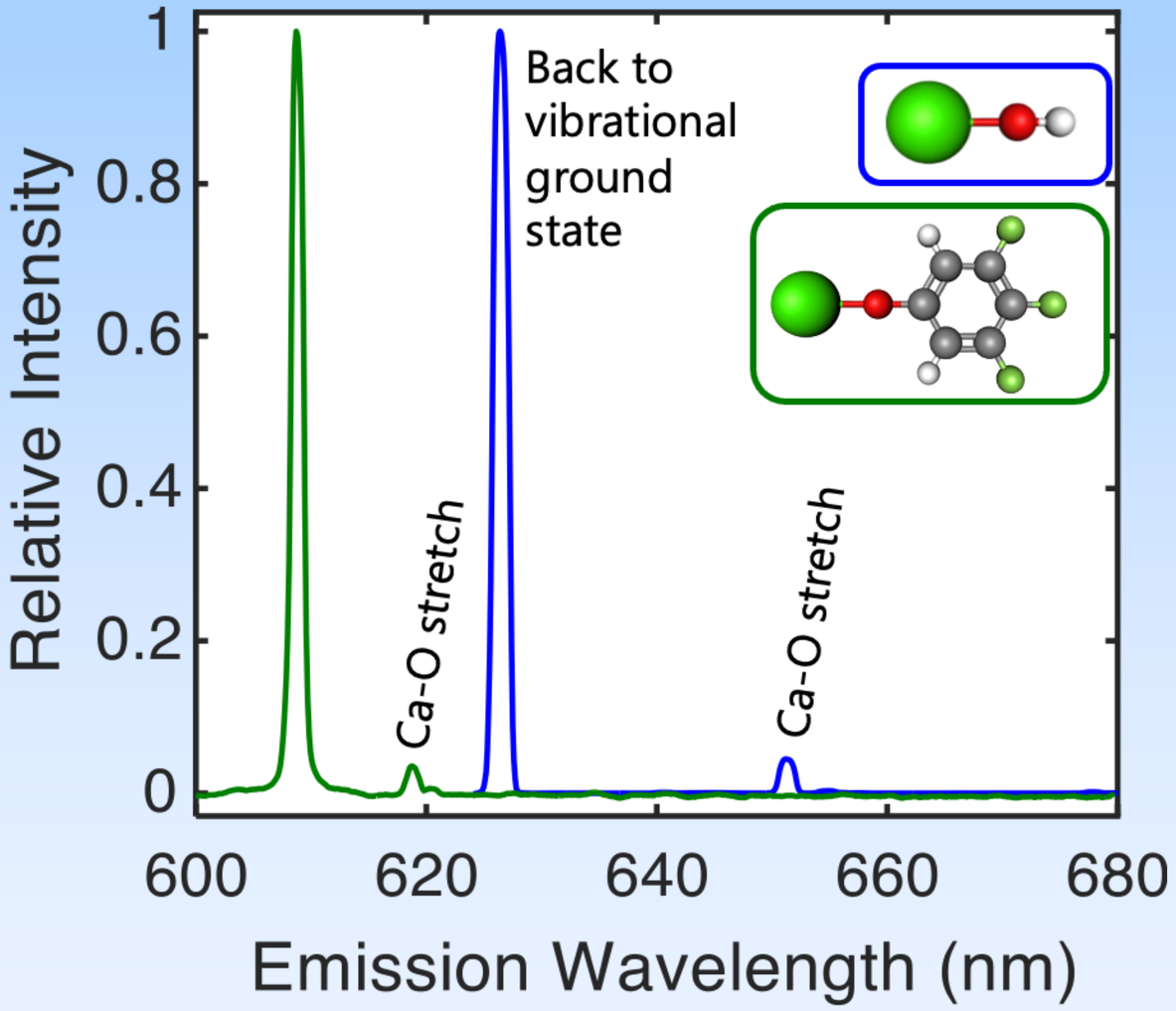
Can Big Molecules Be Laser Cooled?

Vibrational Branching Ratio Measurements



Can Big Molecules Be Laser Cooled?

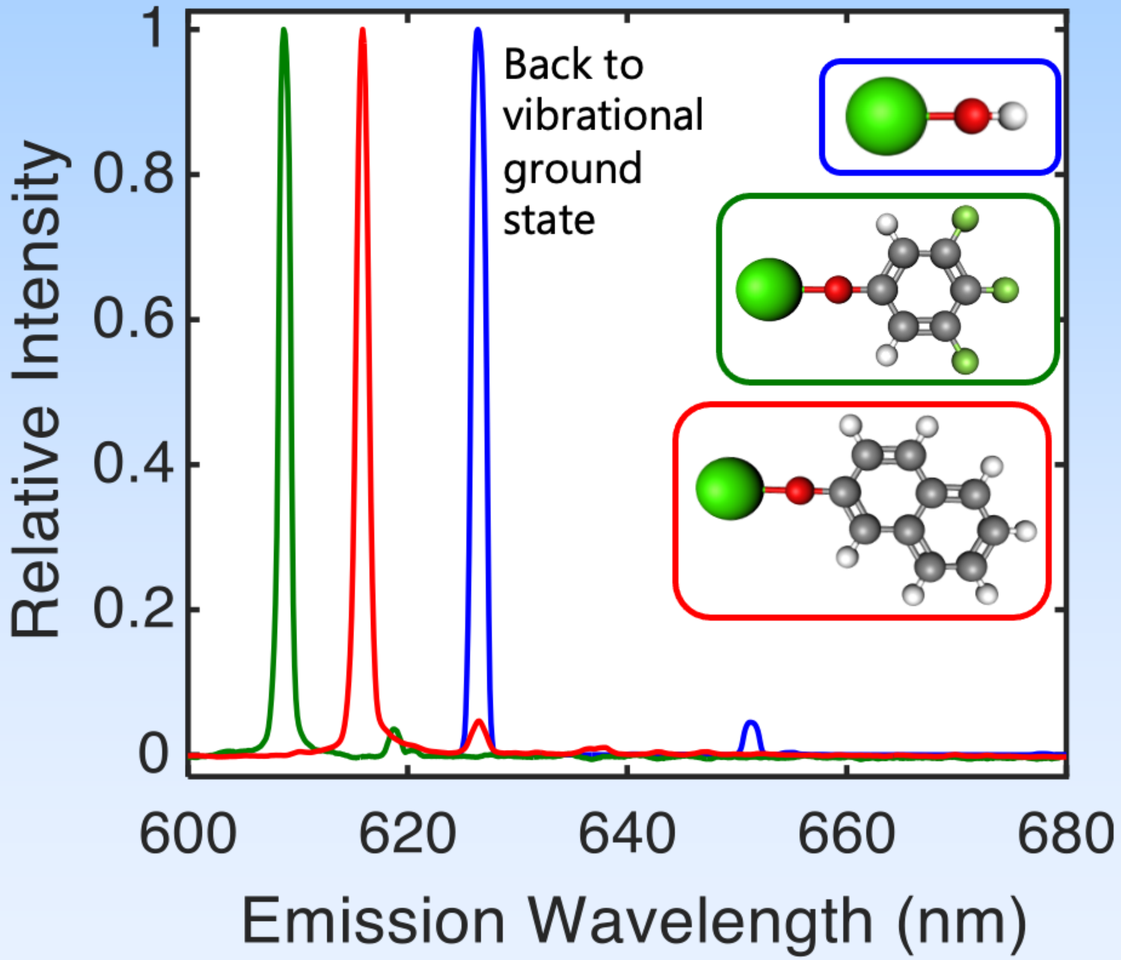
Vibrational Branching Ratio Measurements



Functionalizing Aromatic Compounds with Optical Cycling Centers, G.-Z. Zhu, D. Mitra, B. L. Augenbraun, C. E. Dickerson, M. J. Frim, G. Lao, Z. D. Lasner, A. N. Alexan

Can Big Molecules Be Laser Cooled?

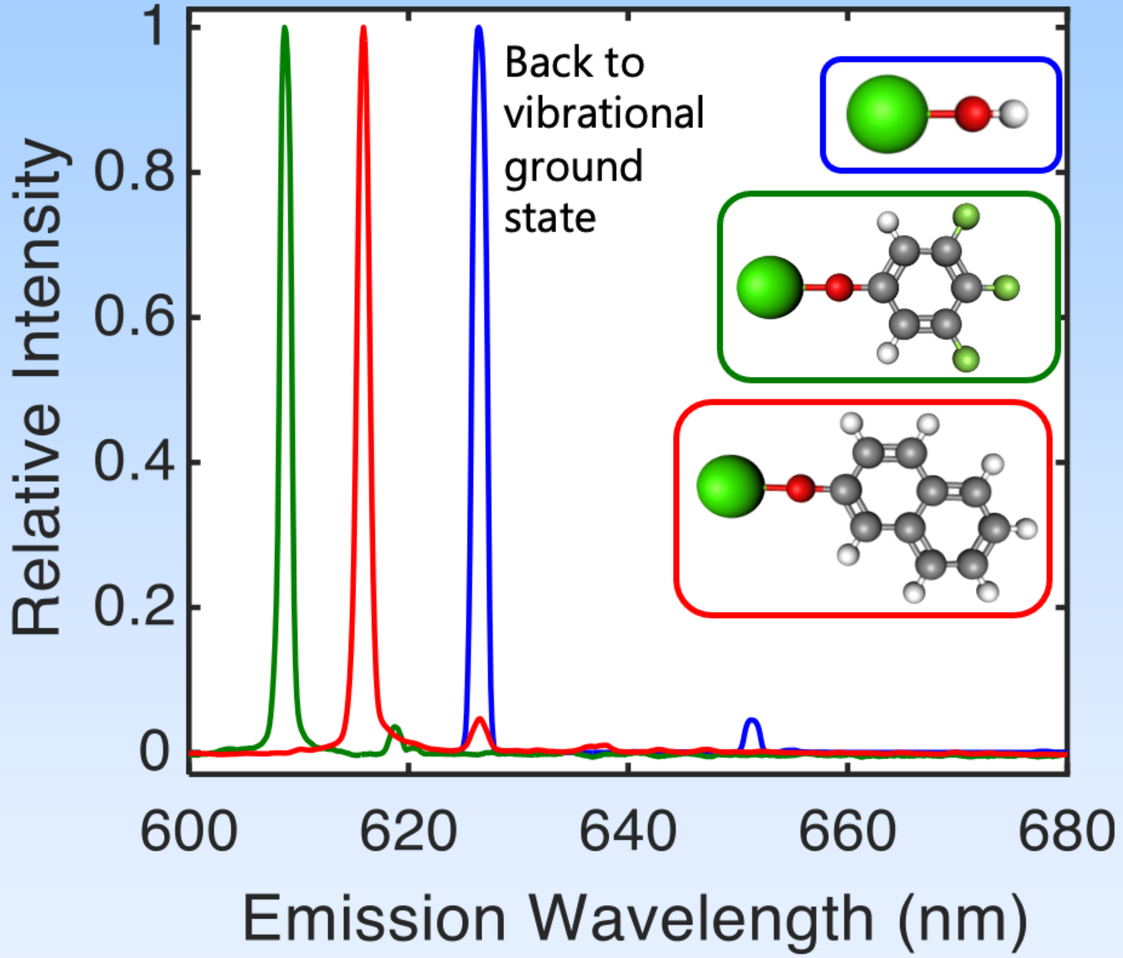
Vibrational Branching Ratio Measurements

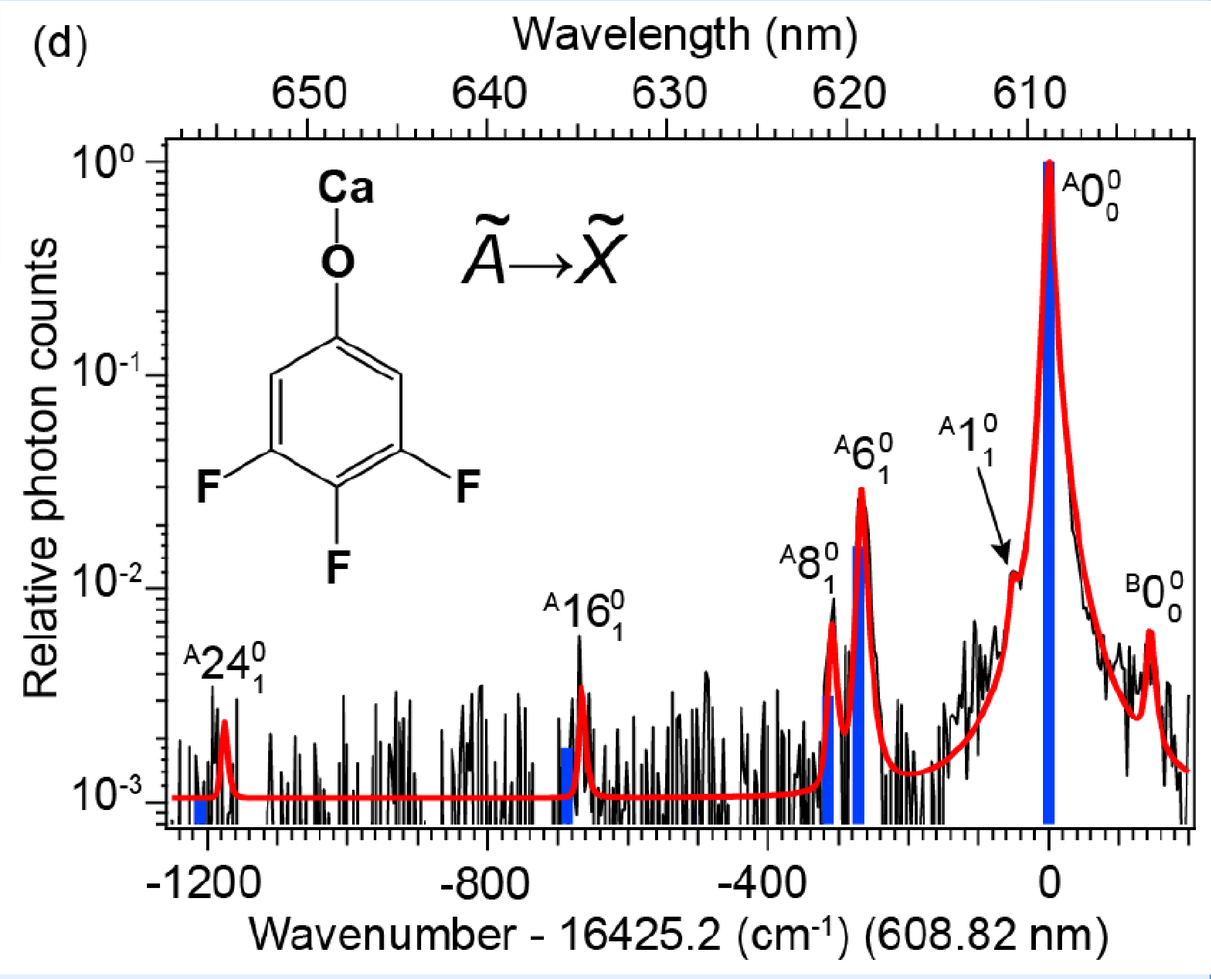


Pathway Towards Optical Cycling and Laser Cooling of Functionalized Arenes, D. Mitra, Z. D. Lasner, G.-Z. Zhu, C. E. Dickerson, B. L. Augenbraun, A. D. Bailey, A. N. Al

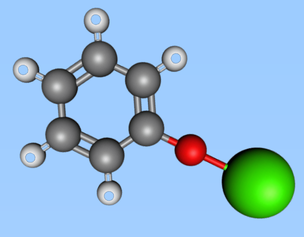
Can Big Molecules Be Laser Cooled?

Vibrational Branching Ratio Measurements



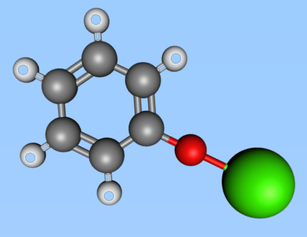


Pathway Towards Optical Cycling and Laser Cooling of Functionalized Arenes, D. Mitra, Z. D. Lasner, G.-Z. Zhu, C. E. Dickerson, B. L. Augenbraun, A. D.



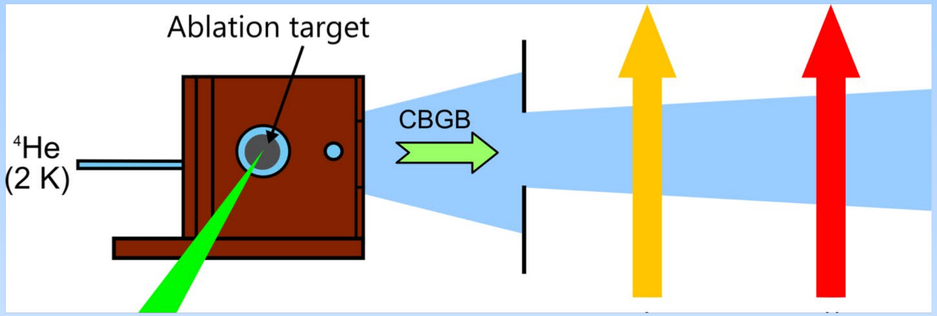
What to do next if we want to laser cool CaOPh?

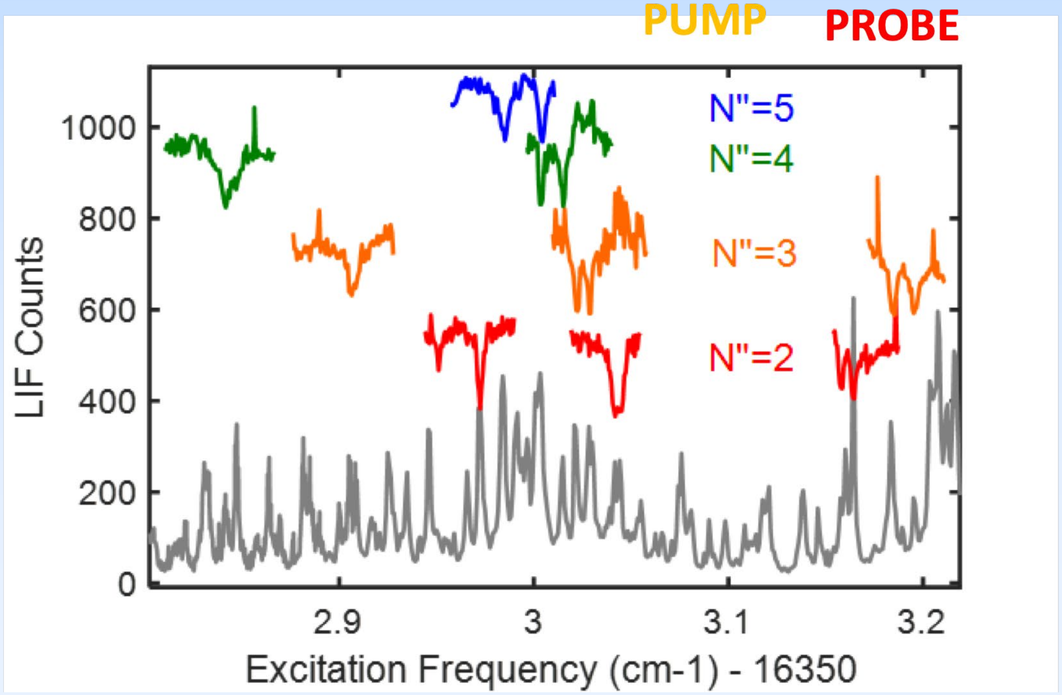
Need rotationally resolved spectroscopy to assign individual lines (like the laser cooling transition(s)!)



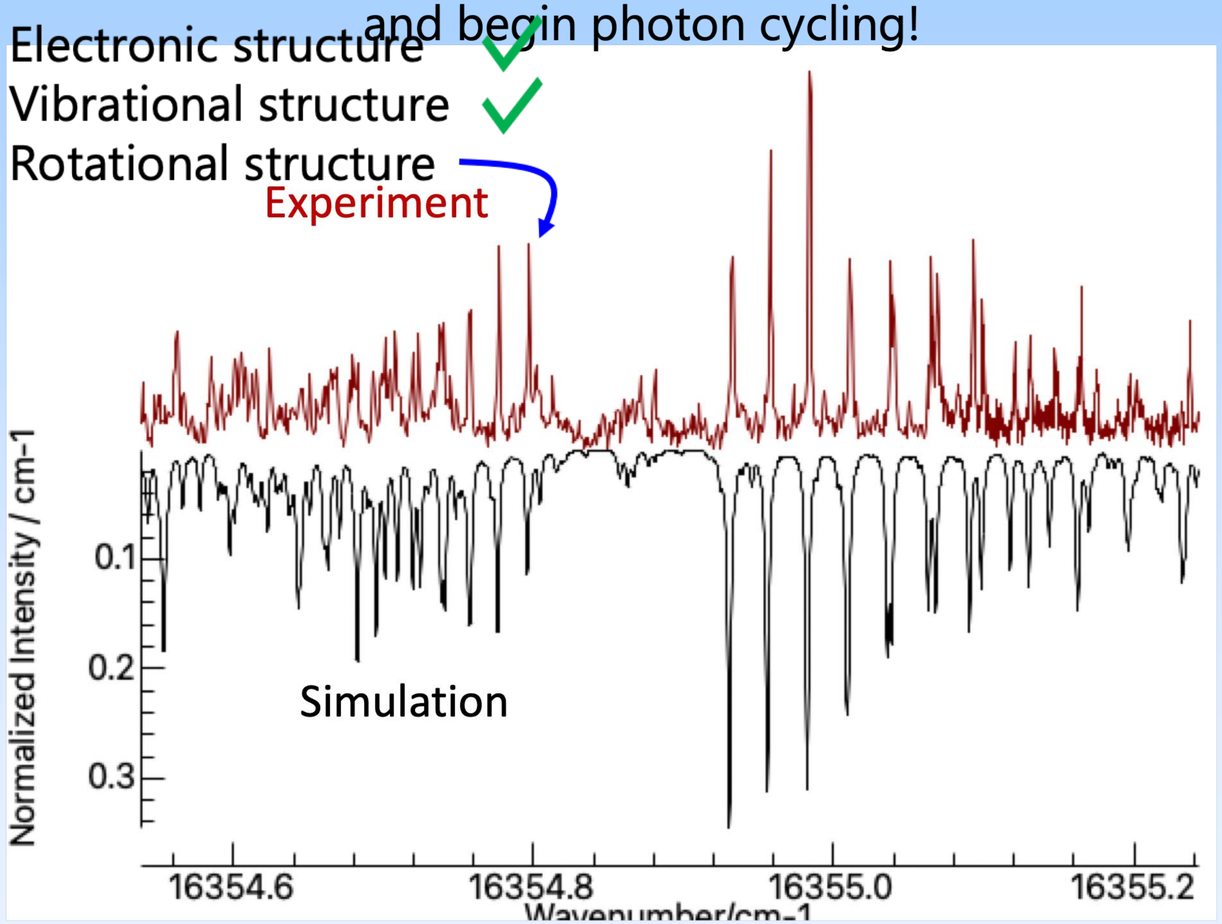
High Resolution (Rotational) Spectroscopy of CaOPh

Using double resonance depletion spectroscopy (pump-probe) to assign rotational quantum numbers





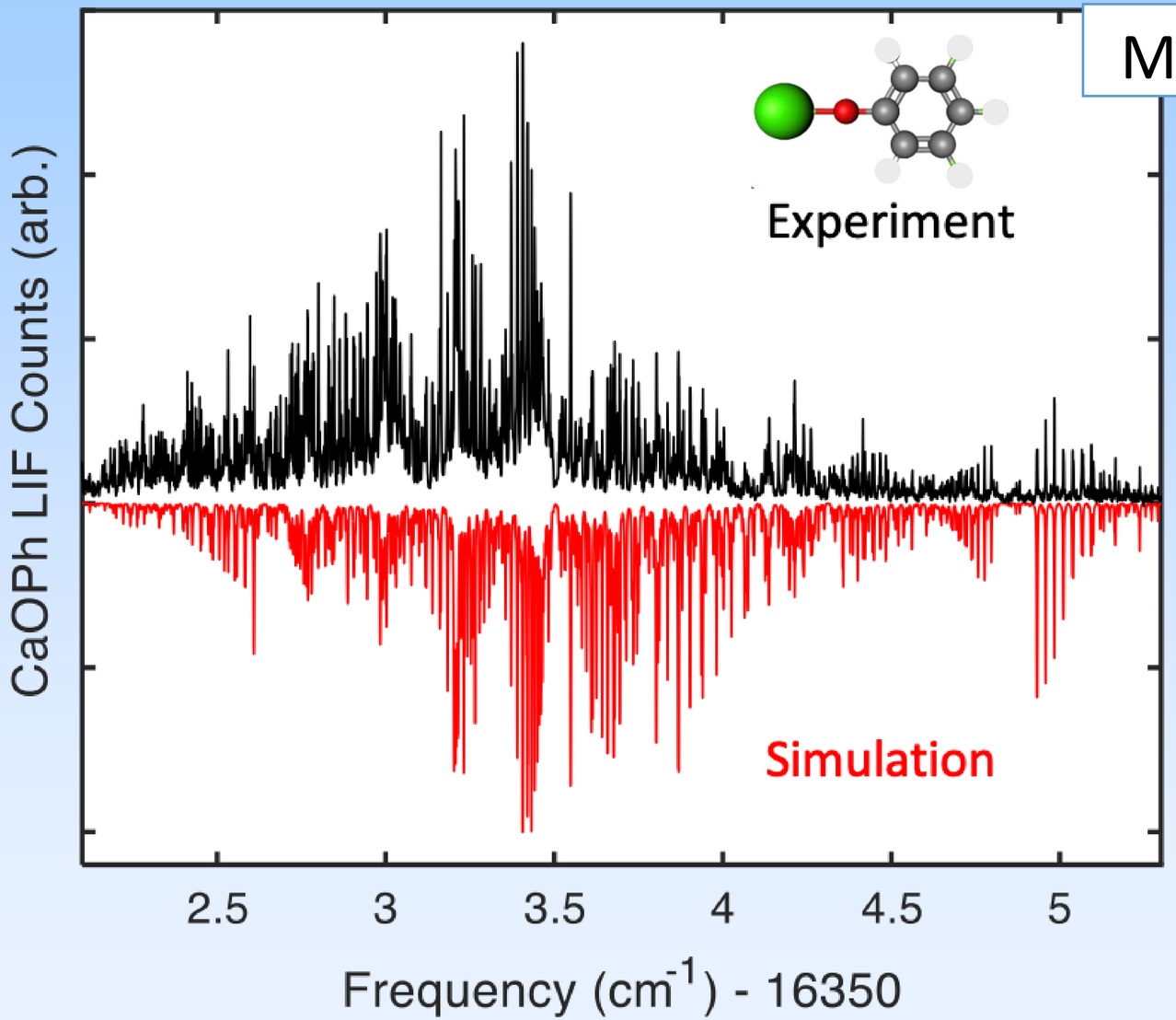
Simulation beginning to converge with experiment: soon we can pick out the “laser cooling” transitions and begin photon cycling!



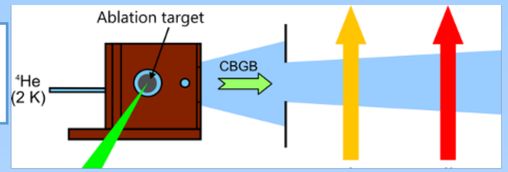
High-Resolution Laser Spectroscopy of a Functionalized Aromatic Molecule, B. L. Augenbraun, S. Burchesky, A. Winnicki, and J. M. Doyle, J. Chem. Phys. Lett. 13, 46, 10771-10777 (2022)

Can Big Molecules Be Laser Cooled?

Electronic structure ✓
Vibrational structure
Rotational structure

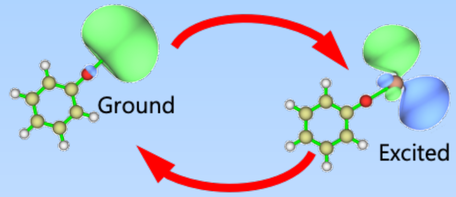


March 2022



Optimize source,
Do spectroscopy

Months-year

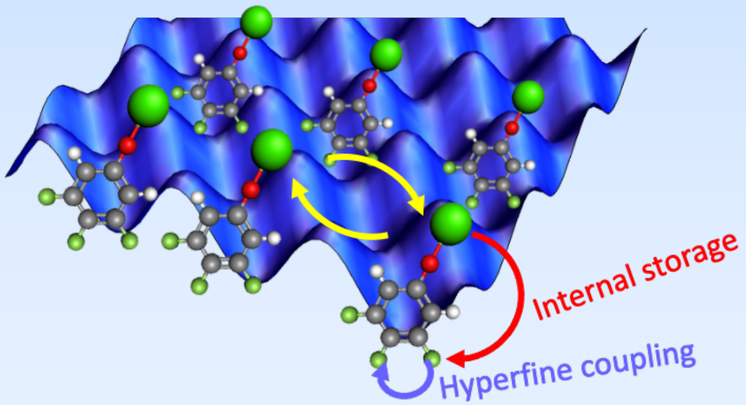


Photon cycling,
1D laser cooling

~Few years

"High order" spectroscopy, slowing, trap loading

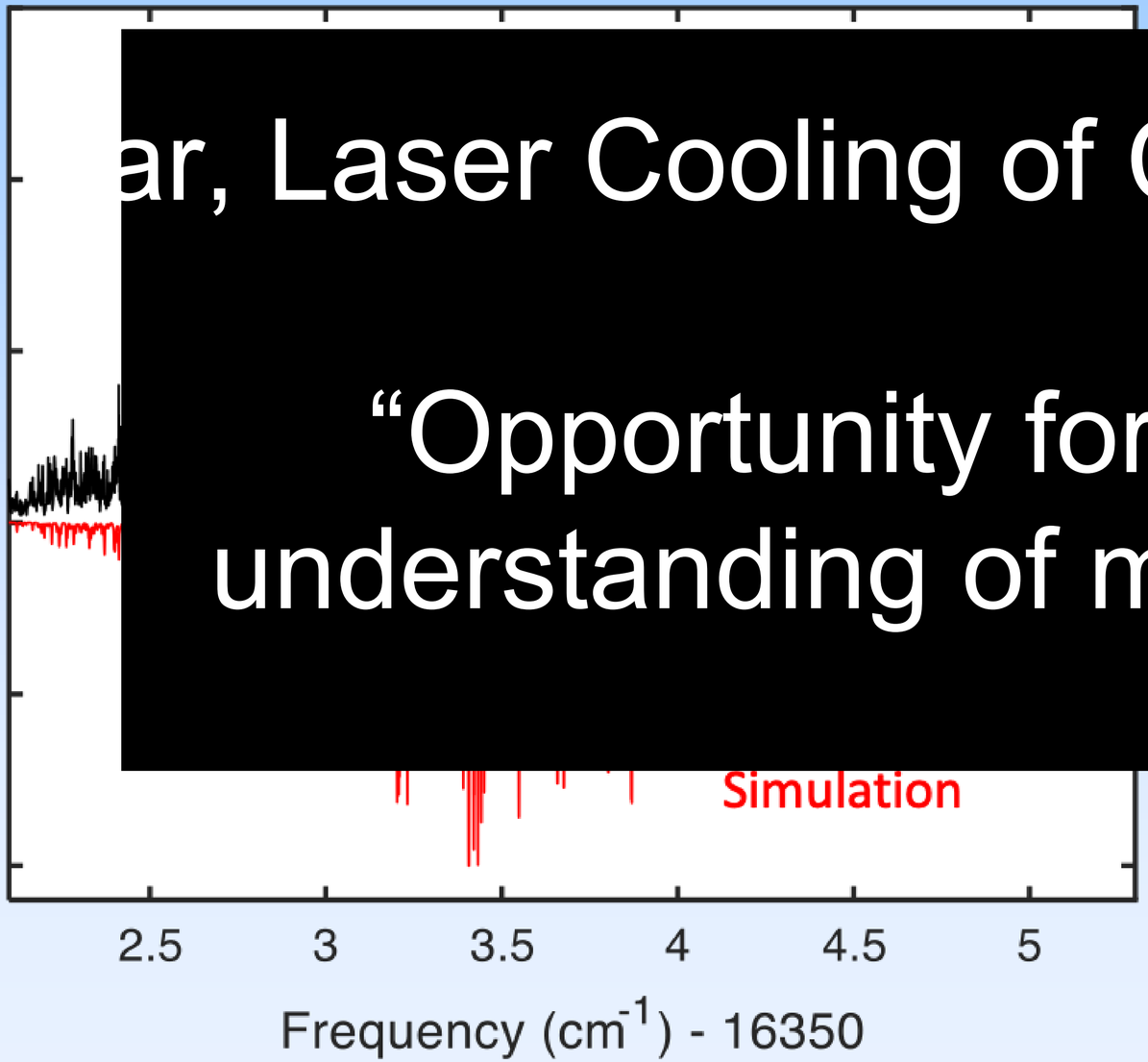
Future?

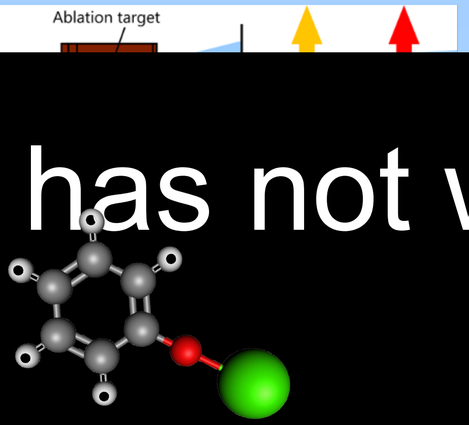


Electronic structure
Vibrational structure
Rotational structure

Can Big Molecules Be Laser Cooled?

So far, Laser Cooling of CaOPh has not worked.





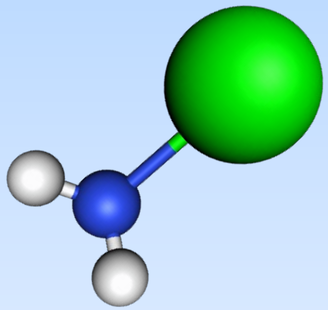
"Opportunity for improving our understanding of molecular systems"

Progress Cooling of Asymmetric Top Molecules

We have accomplished 1-D Doppler cooling of an Asymmetric Top Molecule

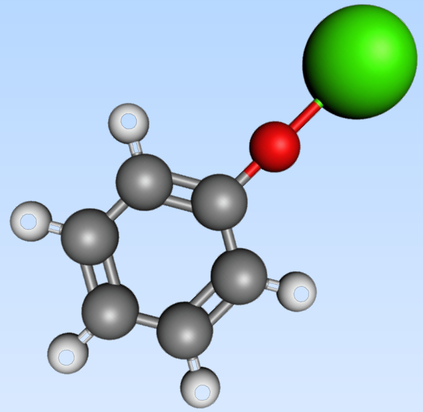
We tried to laser cool this.
No success so far….

$CaNH_2$



1

CaOPh



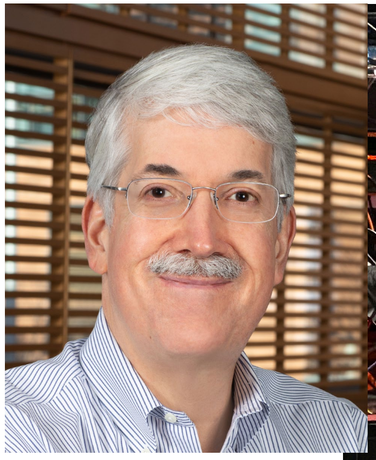
2

Augenbraun,….et al. unpublished 2023

Burchesky, Ph.D. thesis 2023

Direct Laser Cooling of Polyatomic Molecules, B. L. Augenbraun, L. Anderegg, C. Hallas, Z. D. Lasner, N. B. Vilas, and J. M. Doyle (2023).

CaOPh High Resolution and $CaNH_2$ Team

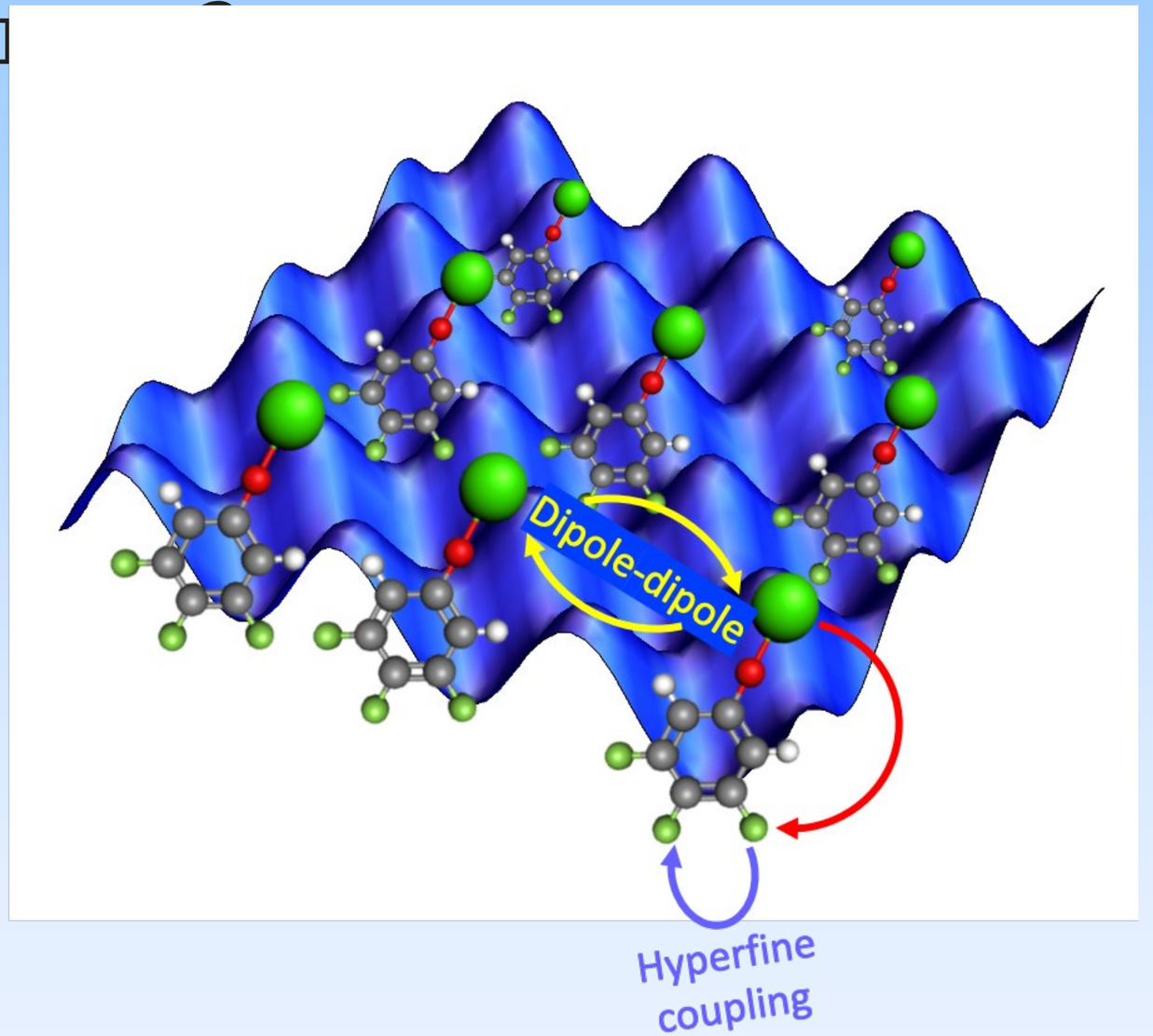


Sean Burchesky
Derick Gonzalez
Andrew Winnicki

Ben Augenbraun
JMD

Dear Kind Speaker, Please Skip to Slide 101

A Future of Ultracold Quantum Scien



If you will it dude….it is no dream…

Current Collaborators, Group Members, Funding

Quantum Simulation (diatomic, CaF)

ARO and Quantum Systems Accelerator

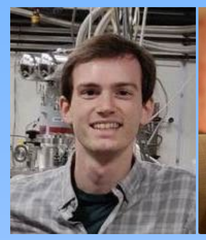

Yicheng Bao


Scarlett Yu


Loic Anderegg

Quantum Simulation (triatomic, CaOH, and bending mode)

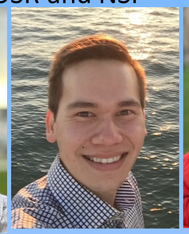
AFOSR and NSF


Nathaniel Vilas


Paige Robichaud


Christian Hallas


Loic Anderegg

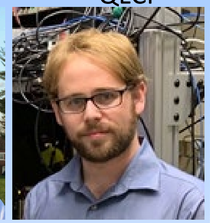

Andrew Winnicki (U)


Arian (Caltech) Jadbabaie

PolyDarkMatter (SrOH)

QLCI


Annika Lundstad


Zack Lasner


Hana Lampson (U)


Derick Gonzalez-Acevedo

PolyEDM (SrOH, YbOH. Harvard-based Researchers)

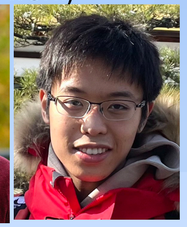
Moore and Sloan (Heising-Simons early work)


Alex Frenett


Hiromitsu Sawaoka


Abdullah Nasir

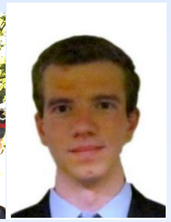

Ben Augenbraun

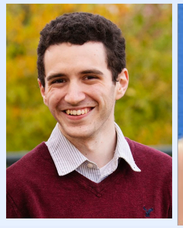

Tasuku Ono (U)

High Resolution Spectroscopy and Laser Cooling of Phenols

Heising-Simons


Grace Li

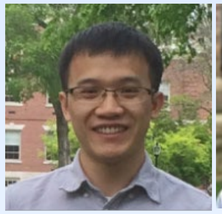

Sean Burchesky


Ben Augenbraun


Claire(UCLA) Dickerson

Low Resolution Spectroscopy and VBR Theory of Aromatics

DOE


Guozhu (UCLA) Zhu


Debayan Mitra


Zack Lasner

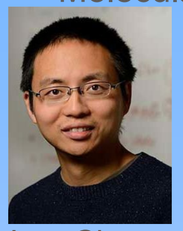
ACME, see other slide

Doyle Group Research also funded by
- Keck
- Templeton
- HQI

U = Undergraduate

Professor Corner

Molecular Theory


Lan Cheng Johns Hopkins


Tanya Zelevinsky

PolyEDM co-PIs


Nick Hutzler (Caltech)

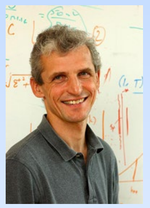

Tim Steimle (ASU)


Amar Vutha (Toronto)

CaF Collaborators


Kang-Kuen Ni (Harvard/CUA)

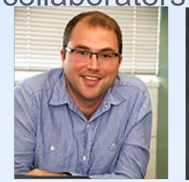

Eunmi Chae (Korea Univ.)

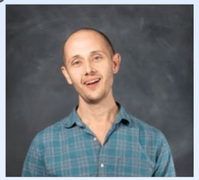

Wolfgang Ketterle (MIT/CUA)

Aromatic collaborators

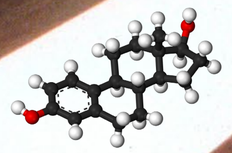

Anastassia Alexandrova (UCLA)


Wes Campbell (UCLA)

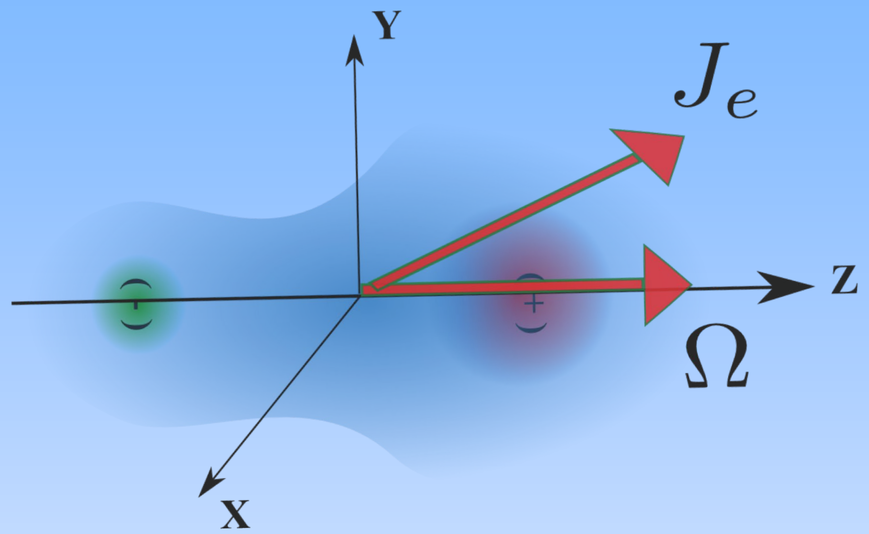

Justin Caram (UCLA)


Eric Hudson (UCLA)

Questions?…..Join me at the bar.



Molecules - Scary??



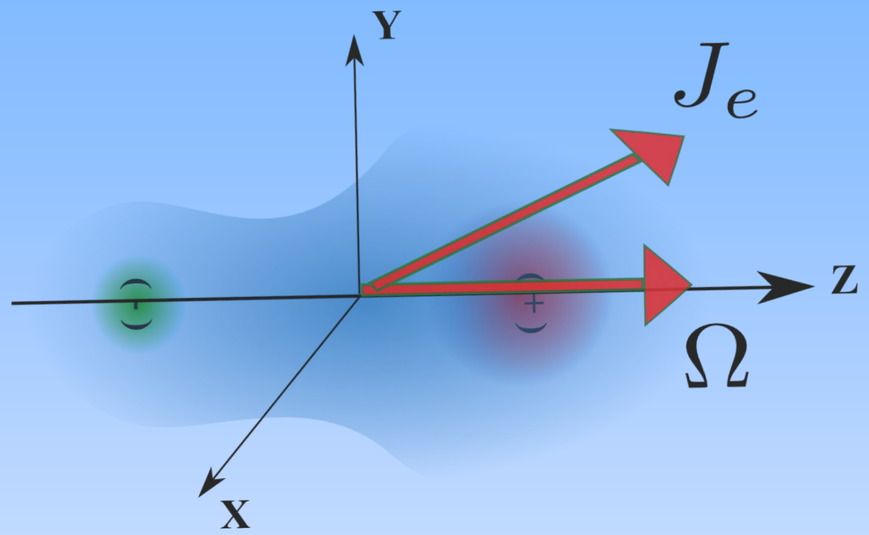
Diatomics

Vibrations

Rotations

A few new Quantum Numbers…

Molecules - Scary??



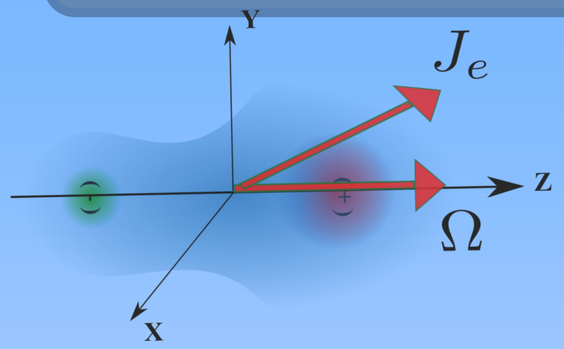
Diatomics

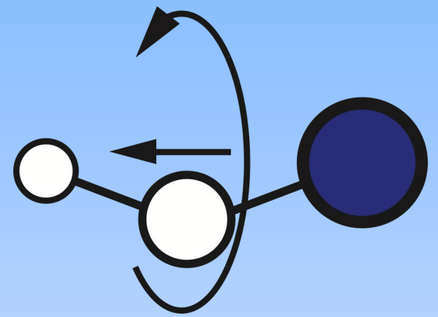
Vibrations
Rotations
A few new Quantum Numbers…

What's the big deal?

Molecules - Scary??

Molecules - Scary??



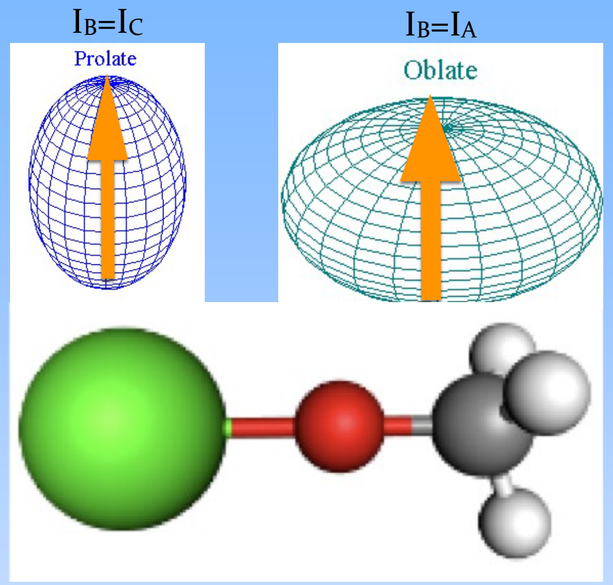


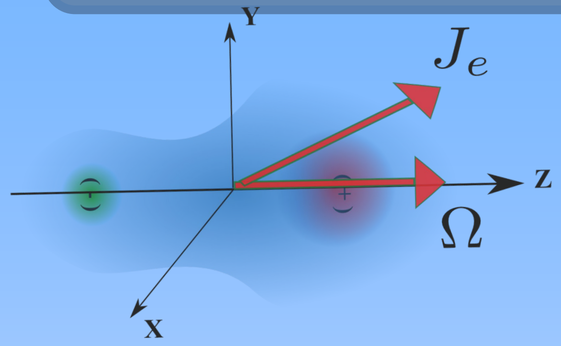
Bending Modes!

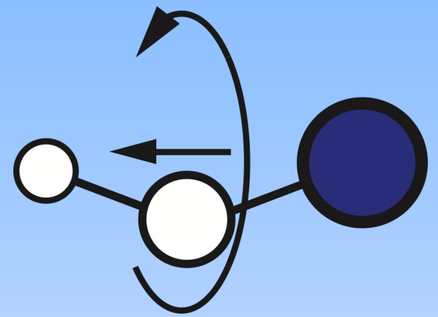
Molecules - Scary??

Symmetric Tops!

$$\text{Symmetric Top Ket} = |J, K, M\rangle$$







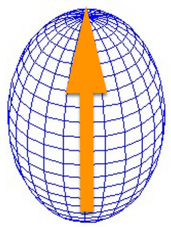
Bending Modes!

Molecules - Scary??

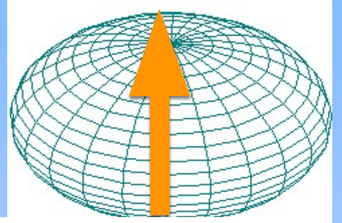
Symmetric Tops!

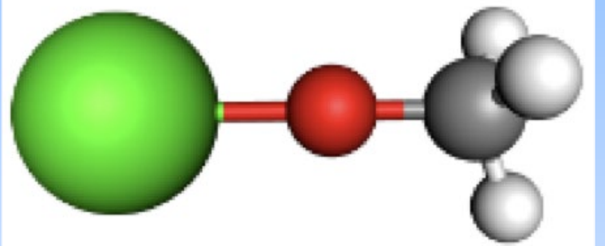
$$\text{Symmetric Top Ket} = |J, K, M\rangle$$

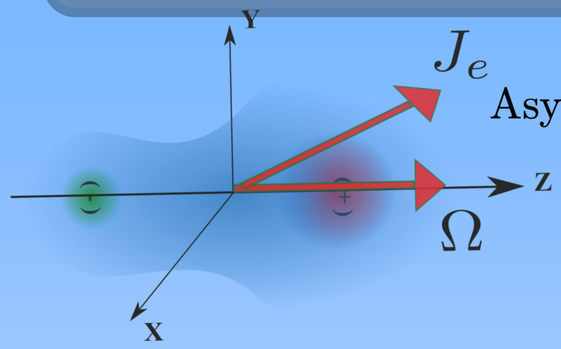
$I_B = I_C$ Prolate



$I_B = I_A$ Oblate







Asymmetric Top Wavefunctions are Simple Sums of Symmetric Top Wavefunctions

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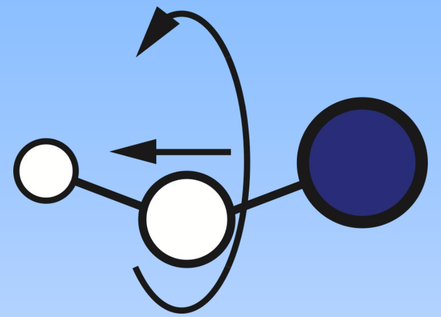
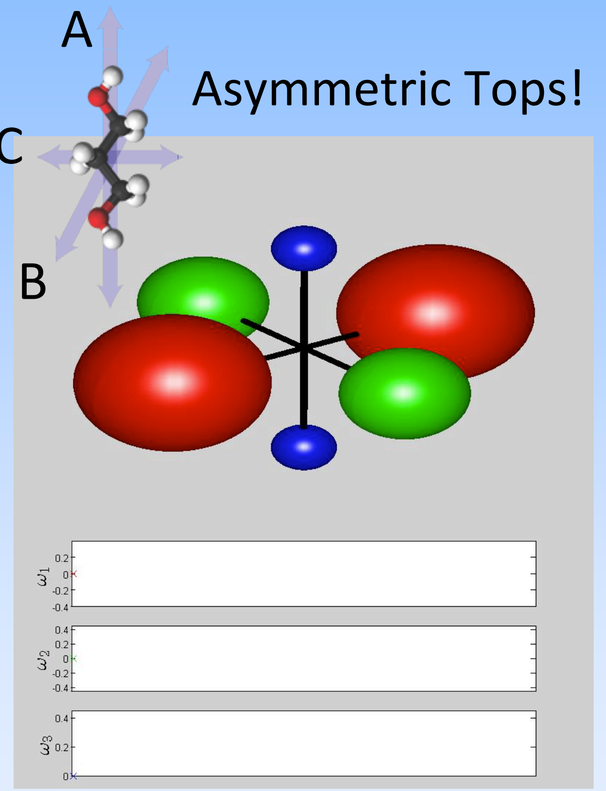
Labels, not quantum numbers



Bending Modes!

Asymmetric Tops!



Molecules - Scary??

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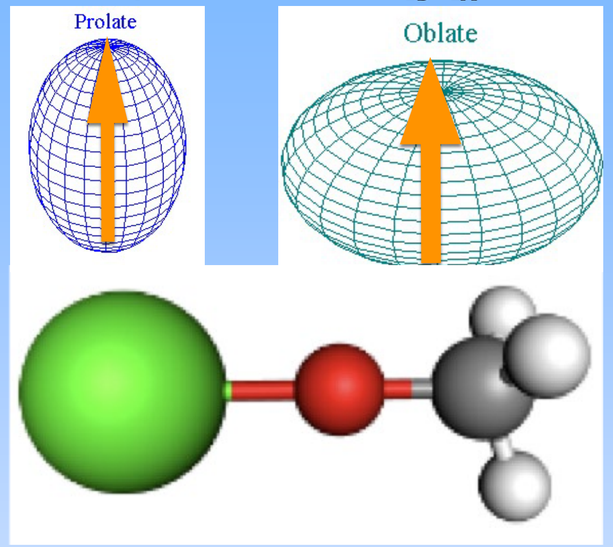

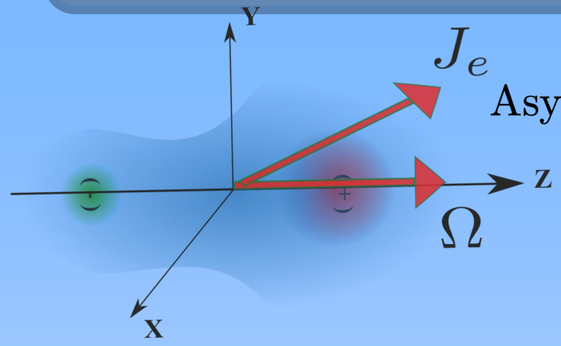

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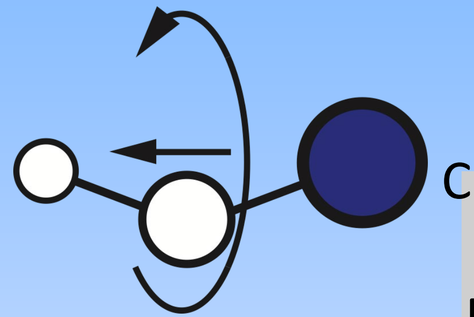

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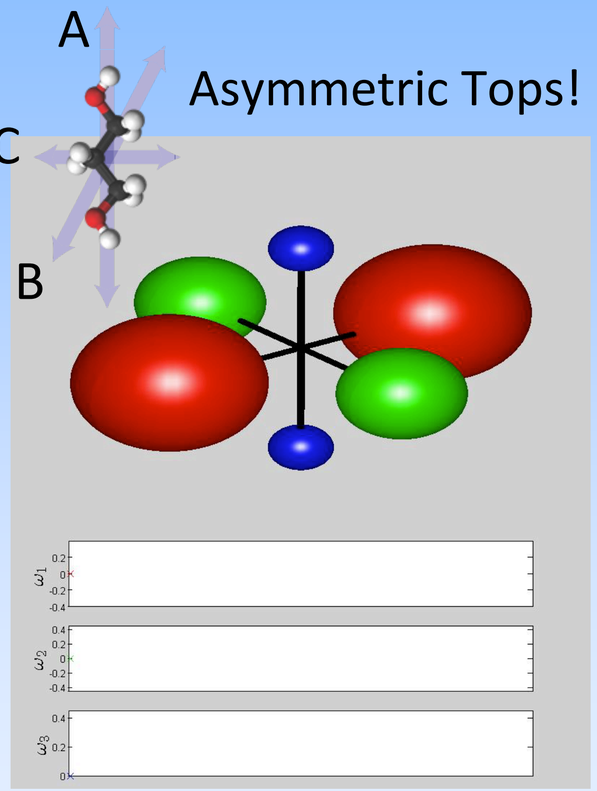

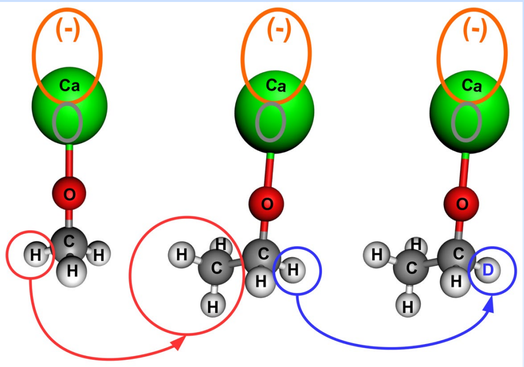

Substitutions, conformers…

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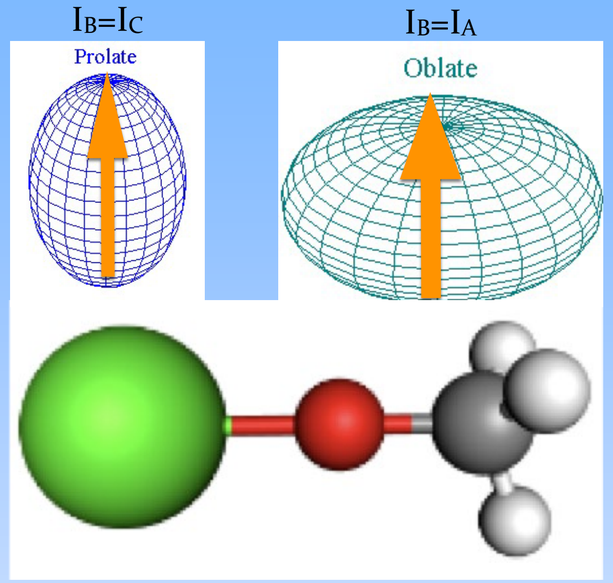

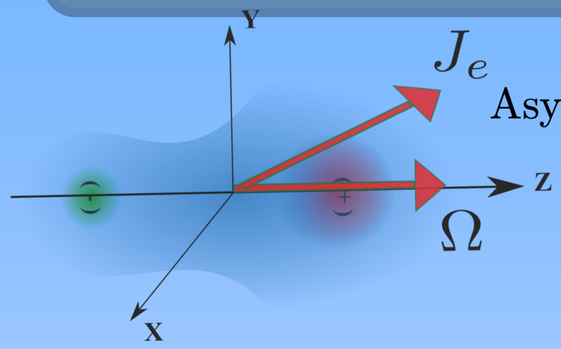

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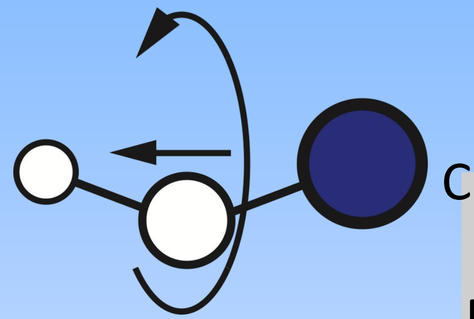

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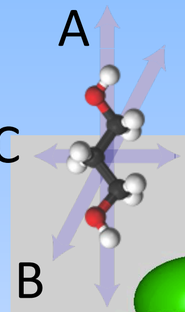

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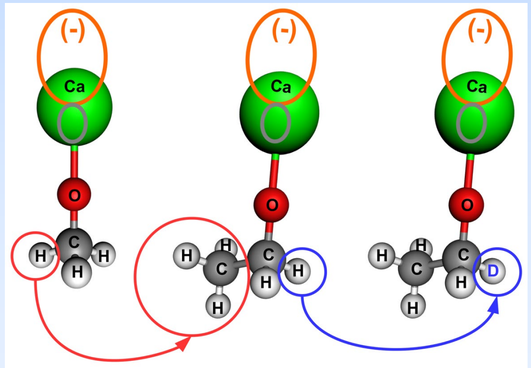
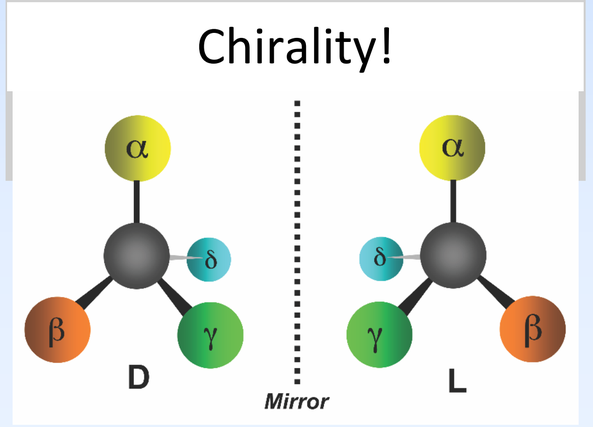


Substitutions, conformers…

Chirality!



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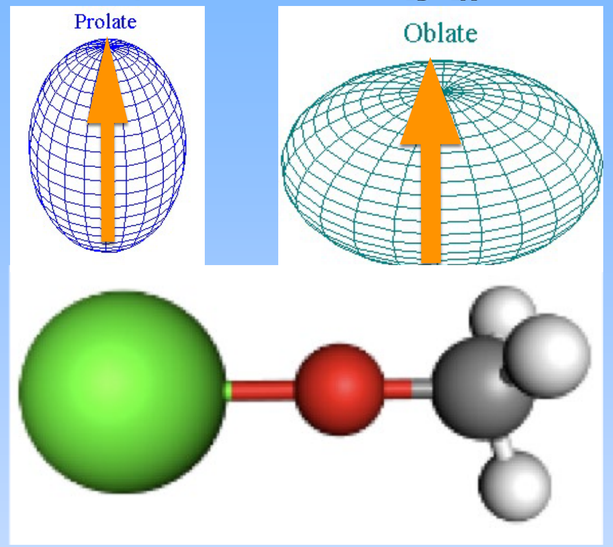

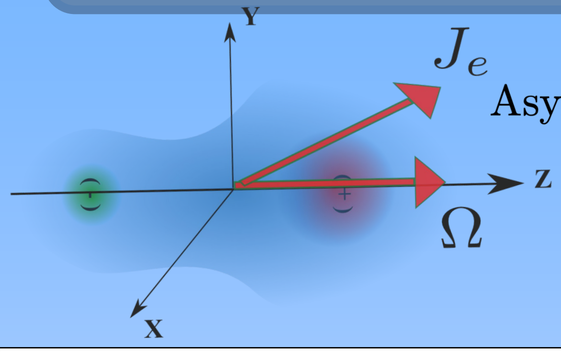

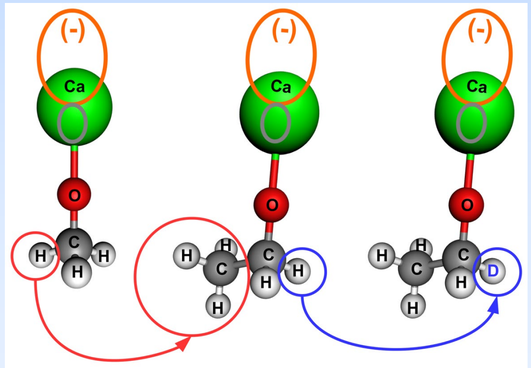

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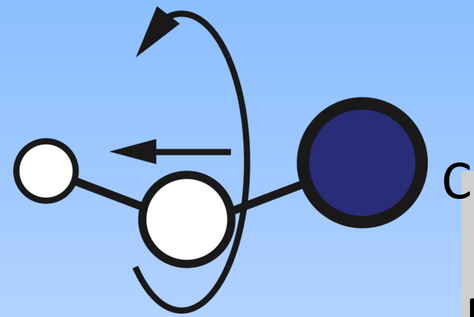
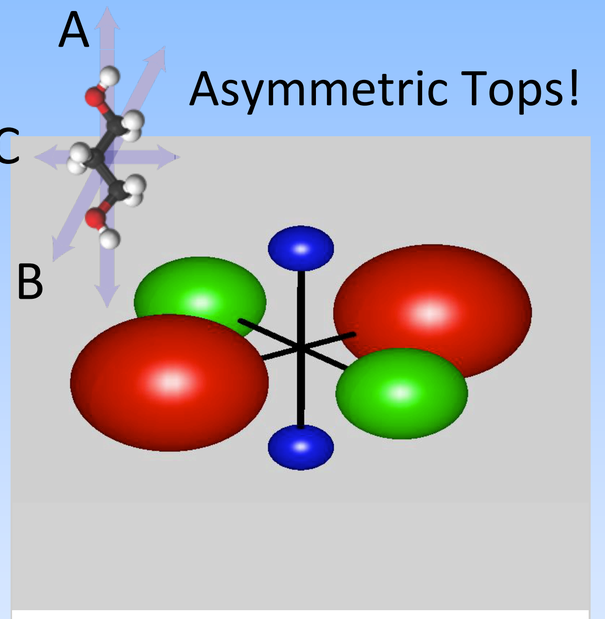


Bending Modes!

Asymmetric Tops!



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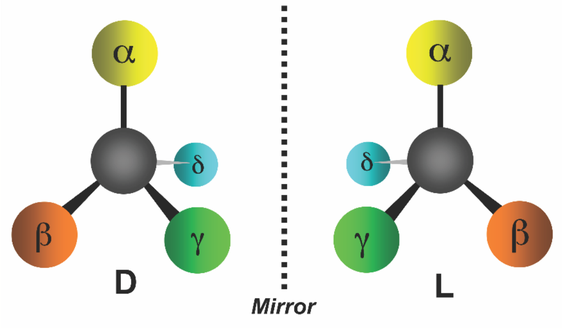

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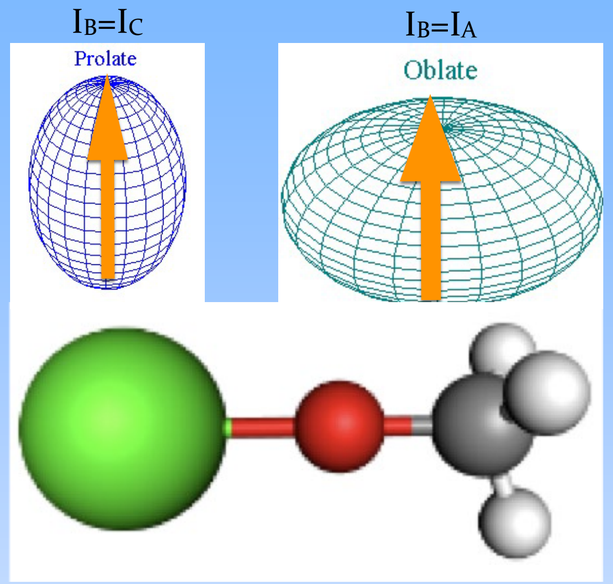

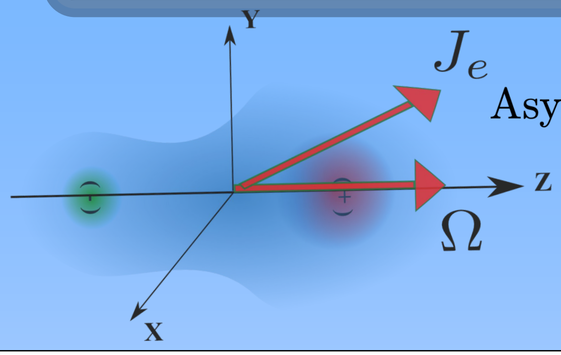

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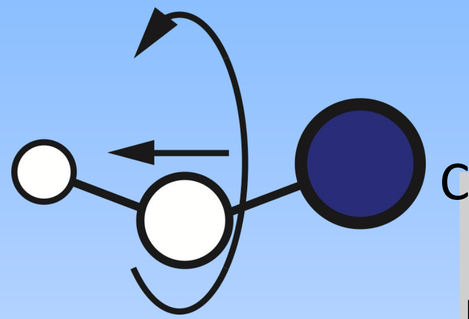
Optical Tensor Stark Shifts!



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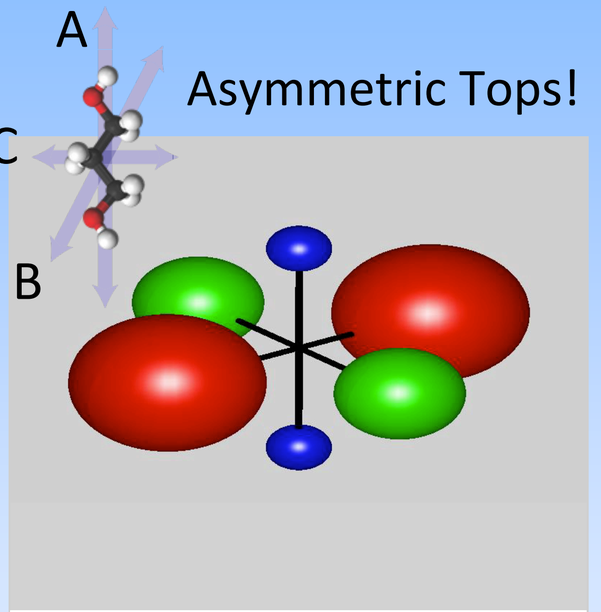

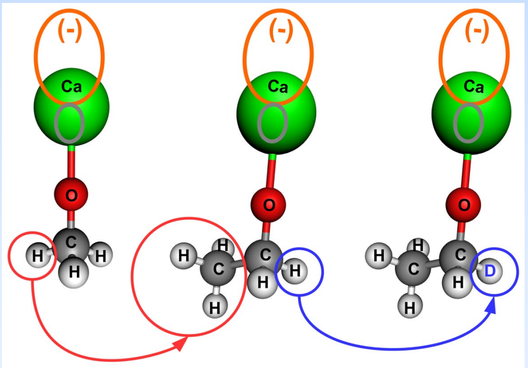

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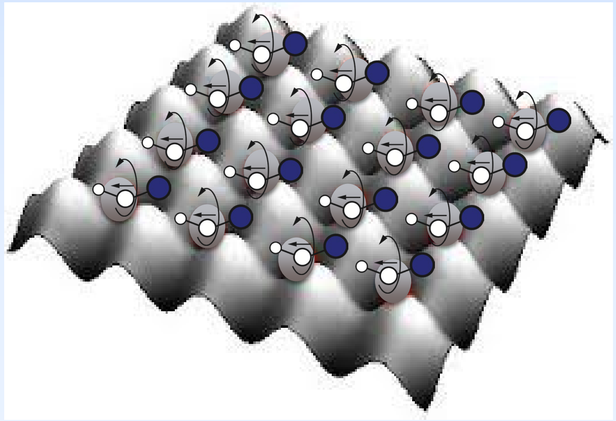

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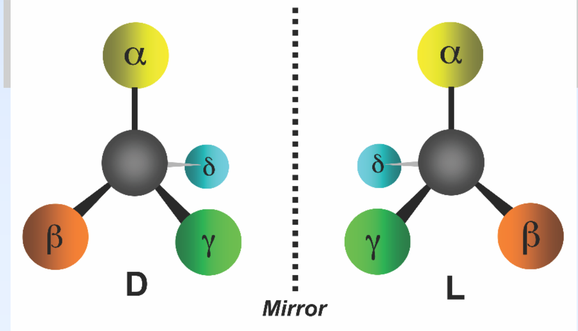

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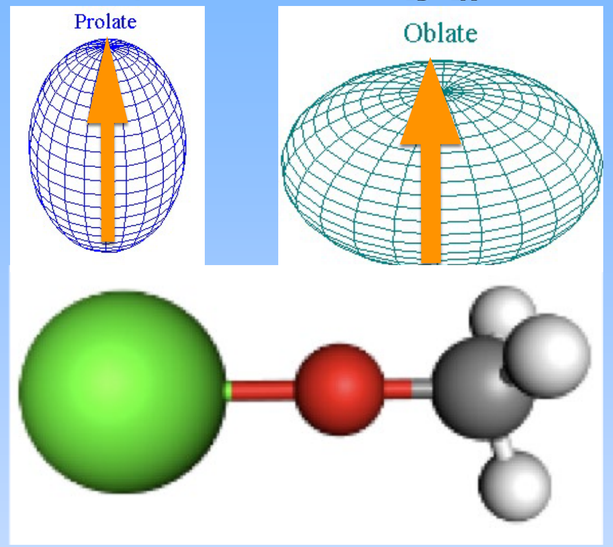

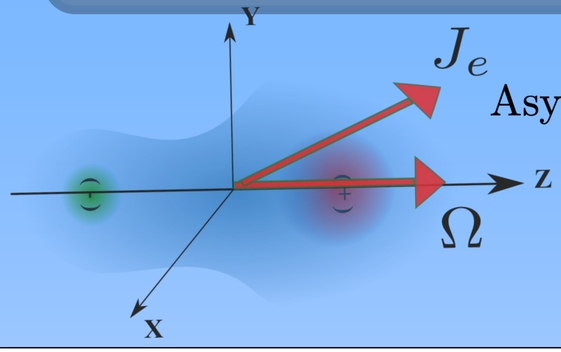

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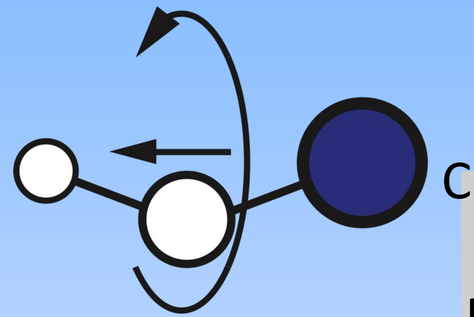

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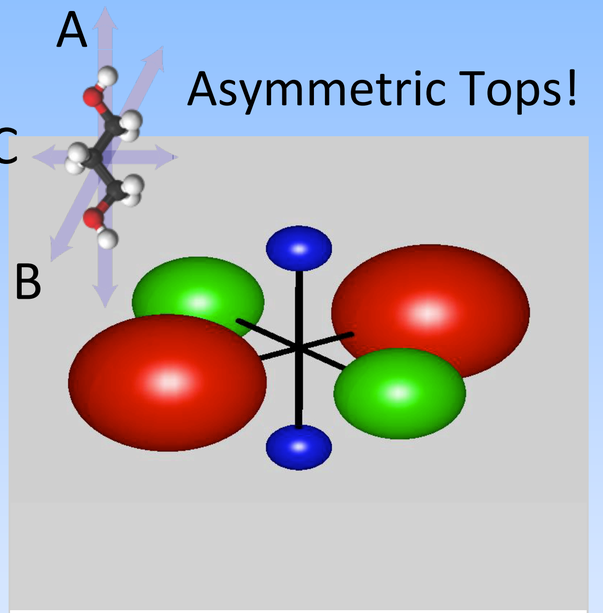

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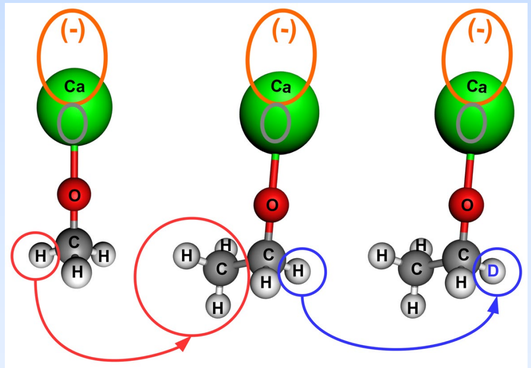

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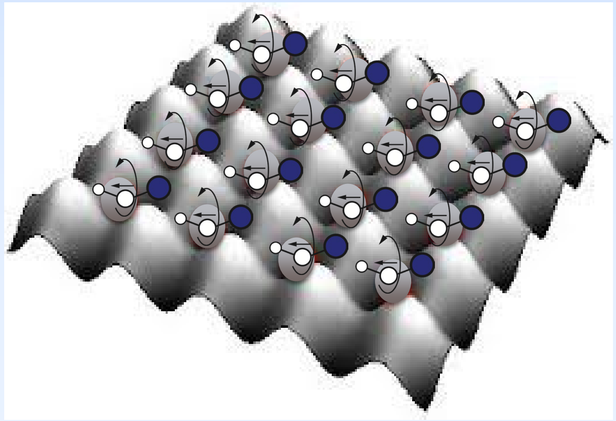
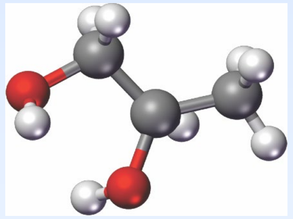
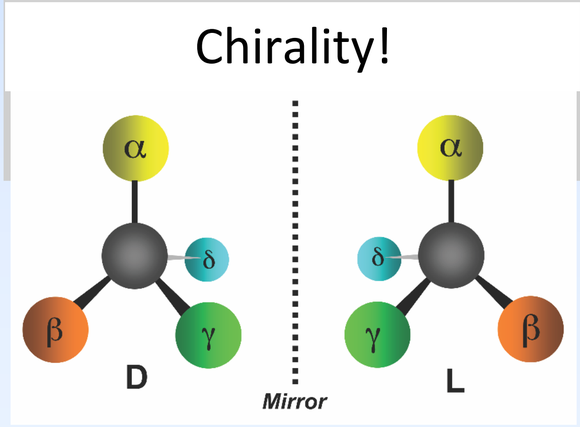




Weird names!

R 1-2 propanediol



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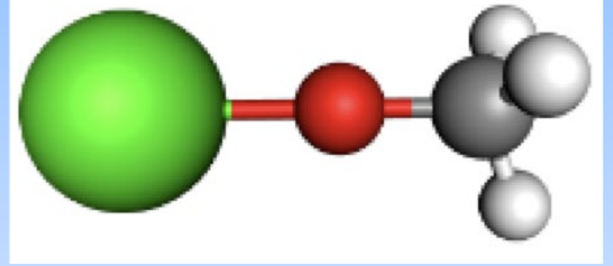

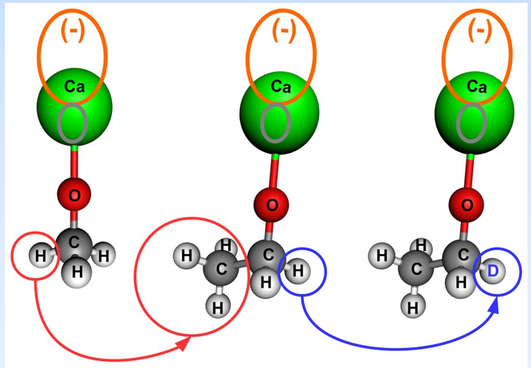

Substitutions, conformers…

YbOH [$\tilde{A}$]

(010) – Spin-orbit/Renner-Teller
(120) – Fermi resonance

Typical scale ~ 2x10^{-4}

CaOH [$\tilde{A}$]

o) – Fermi resonance
er Renner-Teller
/Renner-Teller
~ 2X10^{-4}

$CaOCH_3$ [$\tilde{B}$]

Pseudo-Jahn-Teller
cale ~ 1X10^{-2}

CaSH [$\tilde{C}$]

(020) – Pseudo-Jahn-Teller
Typical scale ~ 10^{-1}

$YbOCH_3$ [$\tilde{A}$]

(010) – Pseudo-Jahn-Teller
Typical scale ~ 10^{-1}

CaOH: Baum, …, Doyle arXiv:2006.01769 (2020)
$CaOCH_3$: Kozyryev, …, Steimle NJP **21** (2019)
$CaSH/CaNH_2$: Augenbraun, …, Doyle, In prep. (2020)
$YbOCH_3$: Augenbraun, …, Steimle, In prep. (2020)

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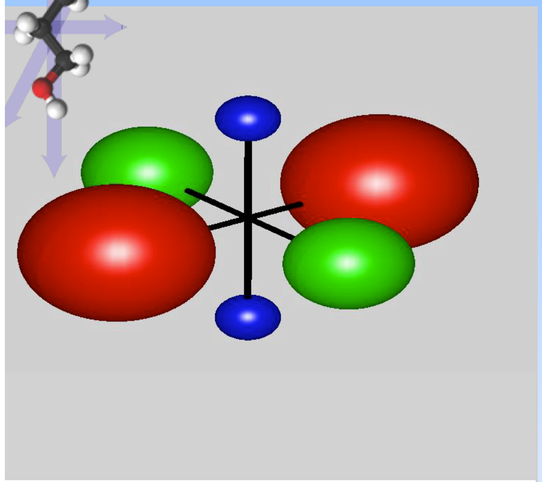

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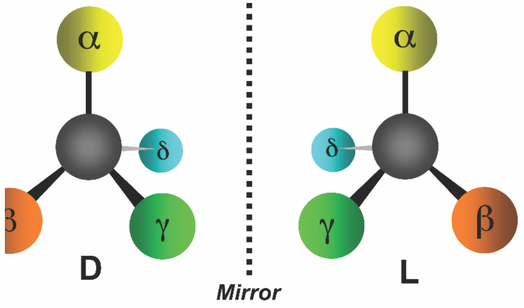

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