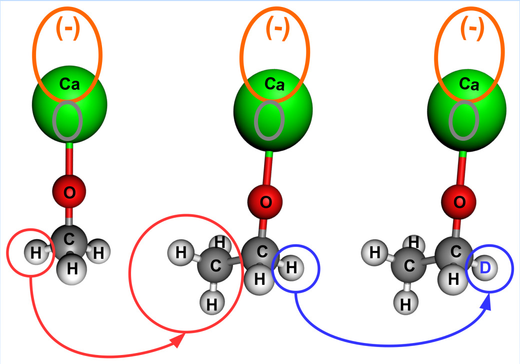
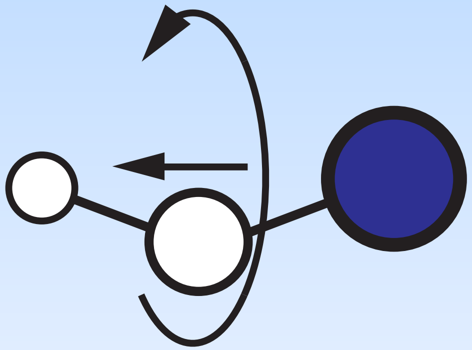
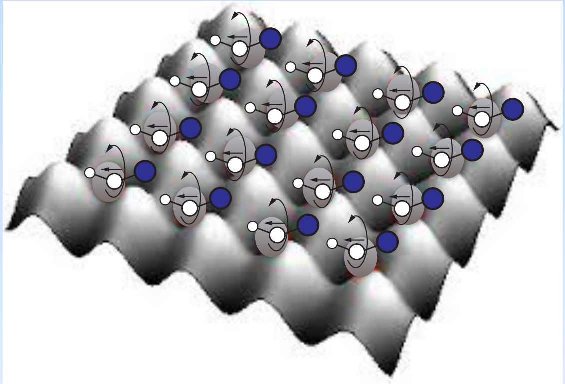
# SPIN
# 2021
# John Doyle

Harvard/MIT Center for Ultracold Atoms



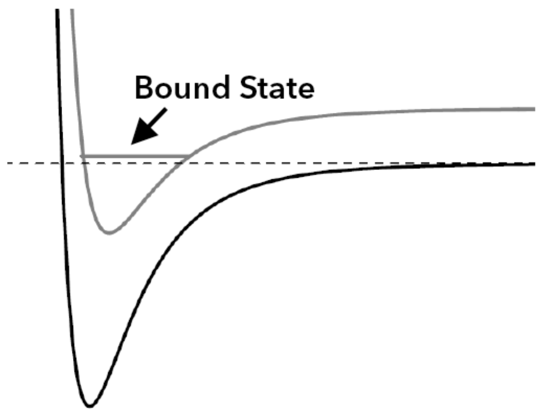


**I have been working with cold and ultracold molecules since 1993.**

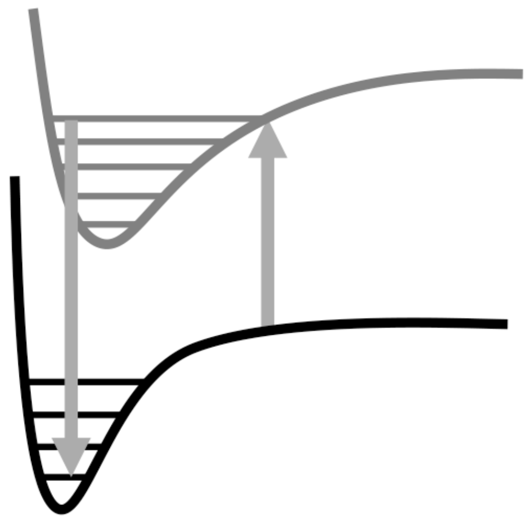
Research with Cold Molecules
has grown

| Molecule Association Groups | | |
|---|---|---|
| **Molecule** | **PI** | **Location** |
| **KRb** | Jin/Ye | JILA |
| **NaK** | Zwierlein | MIT |
| **NaLi** | Ketterle | MIT |
| **RbCs** | Demille | Yale/Chicago |
| **NaK** | Bloch | MPQ |
| **NaCs** | Will | Columbia |
| **NaK** | Pan/Zhao | USTC |
| **YbLi** | Gupta | Washington |
| **RbCs/KCs** | Naegerl | Innsbruck |
| **RbCs/CsYb** | Cornish | Durham |
| **KRb/NaCs** | Ni | Harvard |
| **LiCs** | Weidemüller | Heidelberg |
| **NaK** | Ospelkaus | Hannover |
| **NaRb** | Dajun Wang | Hong Kong |
| **NaCs** | Jia | Shanxi |
| **CsYb** | Cong | Dalian |
| **NaCs** | Bigelow | Rochester |
| **LiK** | Dieckmann | Singapore |
| **NaRb** | Bakr | Princeton |
| **NaK** | Loh | Singapore |
| **LiCs** | Hood | Perdue |
| **RbLi** | Stamper-Kurn | Berkeley |
| **LiCs** | Chin | UChicago |
| **NaRb** | Gadway | Illinois |
| **NaK** | Park | Postech |
| **RbSr** | Deulieu | CNRS Orsay |

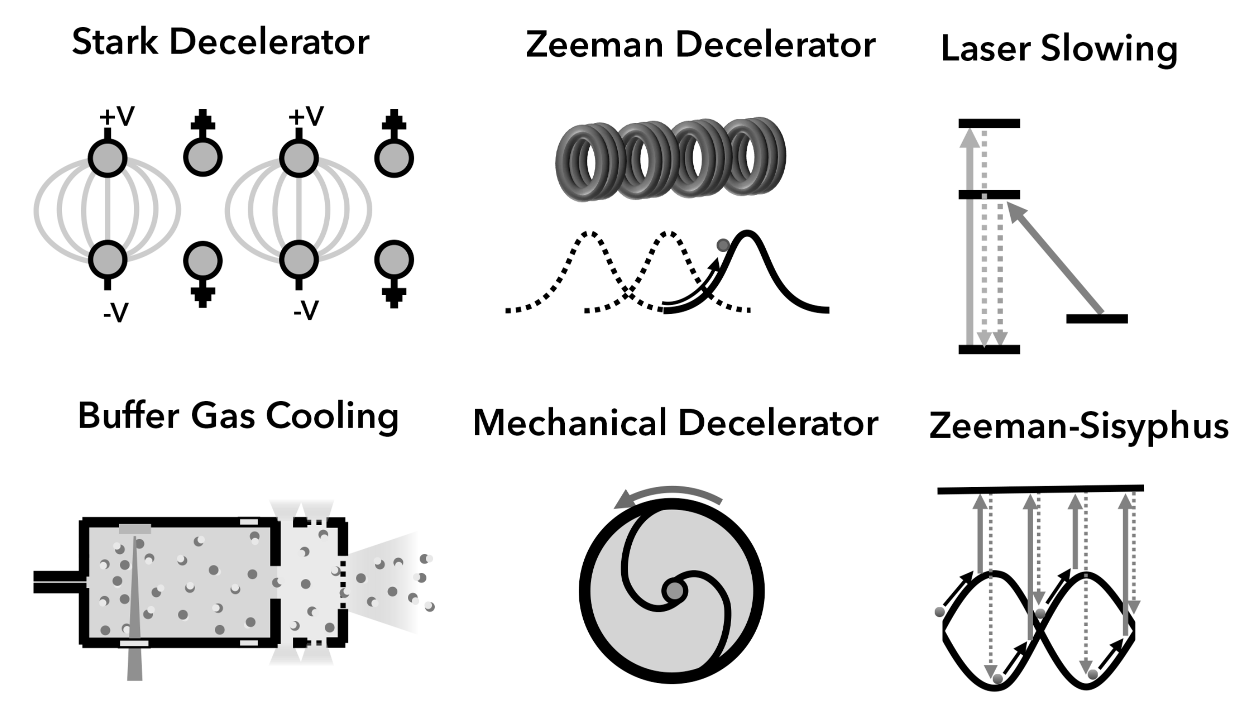
# Feshbach Resonance




# Photo-association



## Beam Slowing Methods




| Direct Laser Cooling of Molecules | | |
|---|---|---|
| **Molecule** | **PI** | **Location** |
| **CaF/SrOH/CaOH/YbOH/$CaOCH_3$** | Doyle | Harvard |
| **CaF/YbF** | Tarbutt | Imperial |
| **CaF** | Osplekaus | Hannover |
| **CaF** | Cheuk | Princeton |
| **SrF/TlF** | Demille | Yale |
| **YO** | Ye | JILA |
| **BaF** | Yan | Zhejiang |
| **TlF** | Hunter | Amherst |
| **AlCl/SrF/CH** | McCarron | UConn |
| **MgF** | Chae | Korea |
| **MgF** | Yin | ECN- Shanghai |
| **AlF** | Truppe | FHI |
| **AlCl** | Hemmerling | UCR |
| **BaH/CaH** | Zelivinski | Columbia |
| **BaF** | Langen | Stuttgart |
| **YbOH** | Hutzler | Caltech |

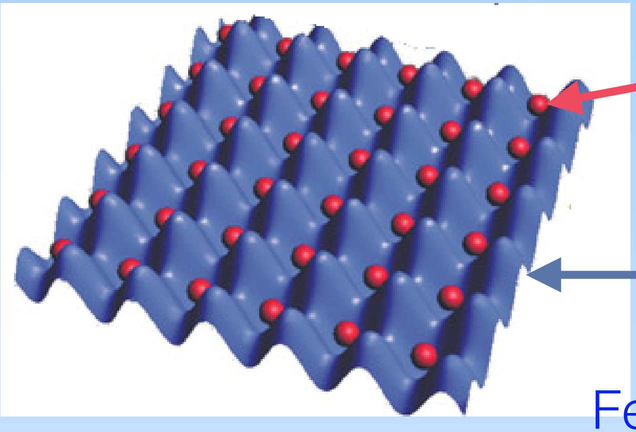
| Molecules - Trapped - Other Methods | | |
|---|---|---|
| **Molecule** | **PI** | **Location** |
| **$ND_3$** | Meijer | FHI |
| **NO** | Barker | ICL |
| **$CH_3$** | Momose | UBC |
| **$CH_3F$, $H_2CO$** | Rempe | MPQ |
| **$ND_3$** | Lewandowski | JILA |
| **OH** | Ye | JILA |
| **$O_2$** | Narevicius | Weizmann |
| | | |

Why do research on/with Cold and Ultracold molecules?

Is there something *wrong* with atoms?

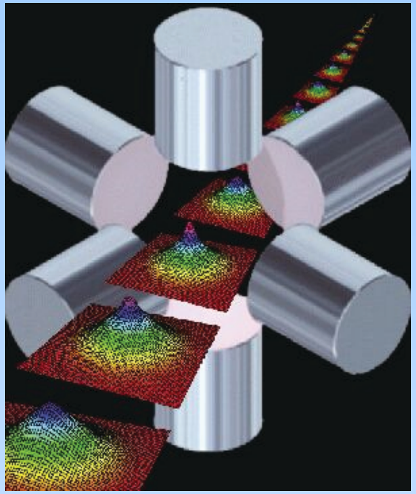
# What has been done with cold atoms?

Best Clocks in the World
Optical Lattice Clock (OLC)

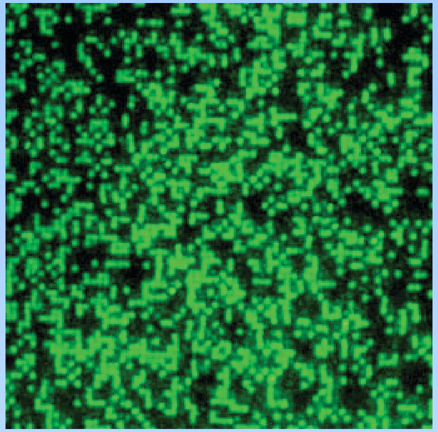


Ultracold atoms
T~microKelvin

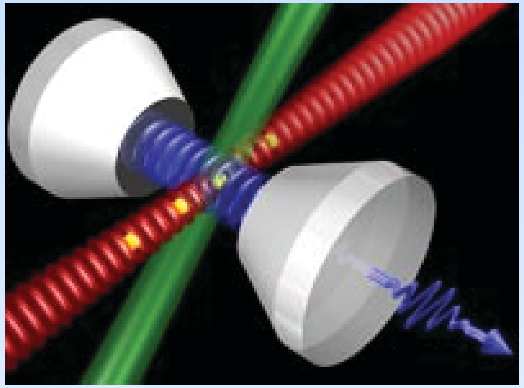
Optical Lattice



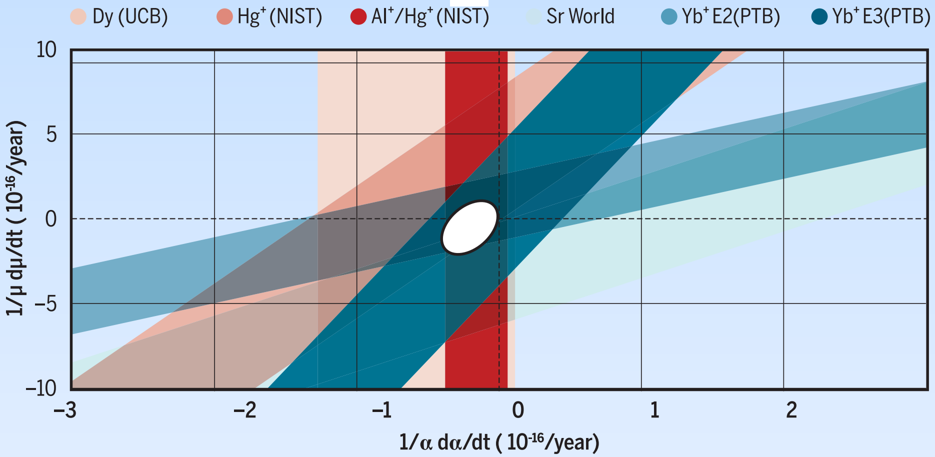
New Quantum Fluids
Fermionic, Bosonic, vortices,
fluid circuits, ….



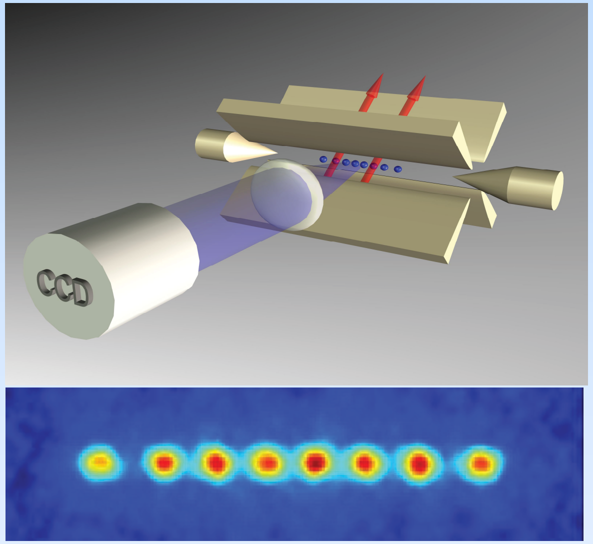
Quantum simulation
Fermi-Hubbard Model…



Quantum Cavity Dynamics
squeezing for measurement, etc

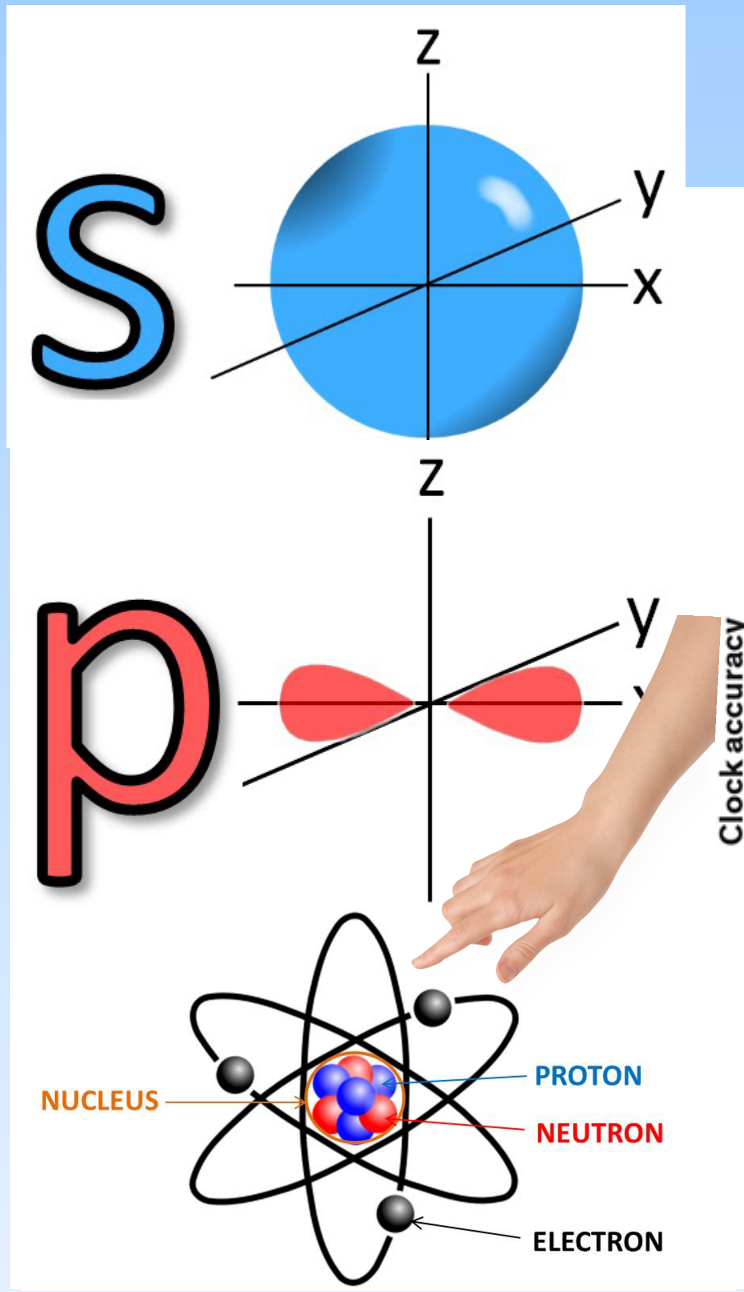


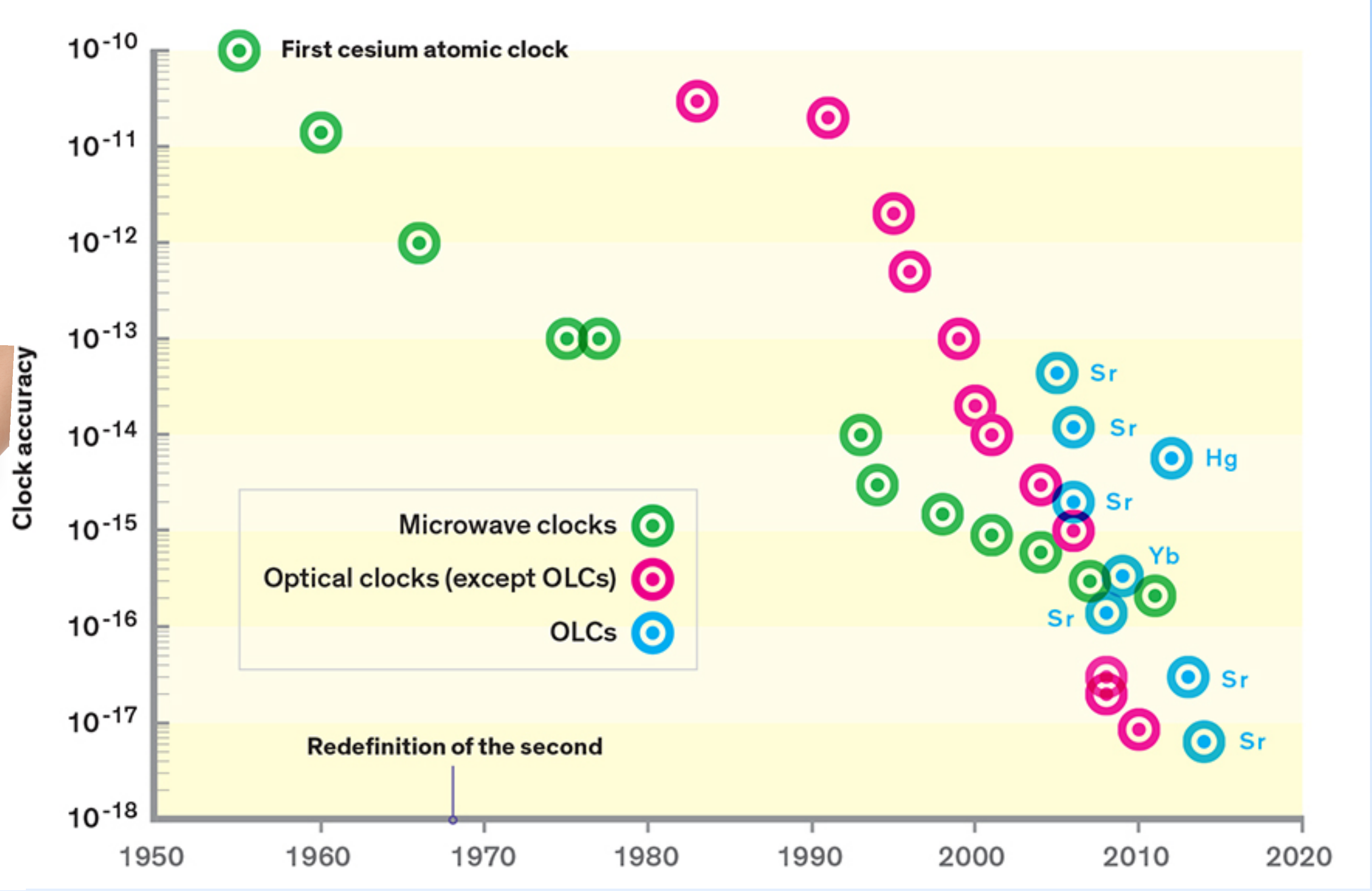

Dark Energy, Dark Matter,
BSM, CP violation




Quantum computers

# Atomic Clock History



# Why *Cold* Molecules??

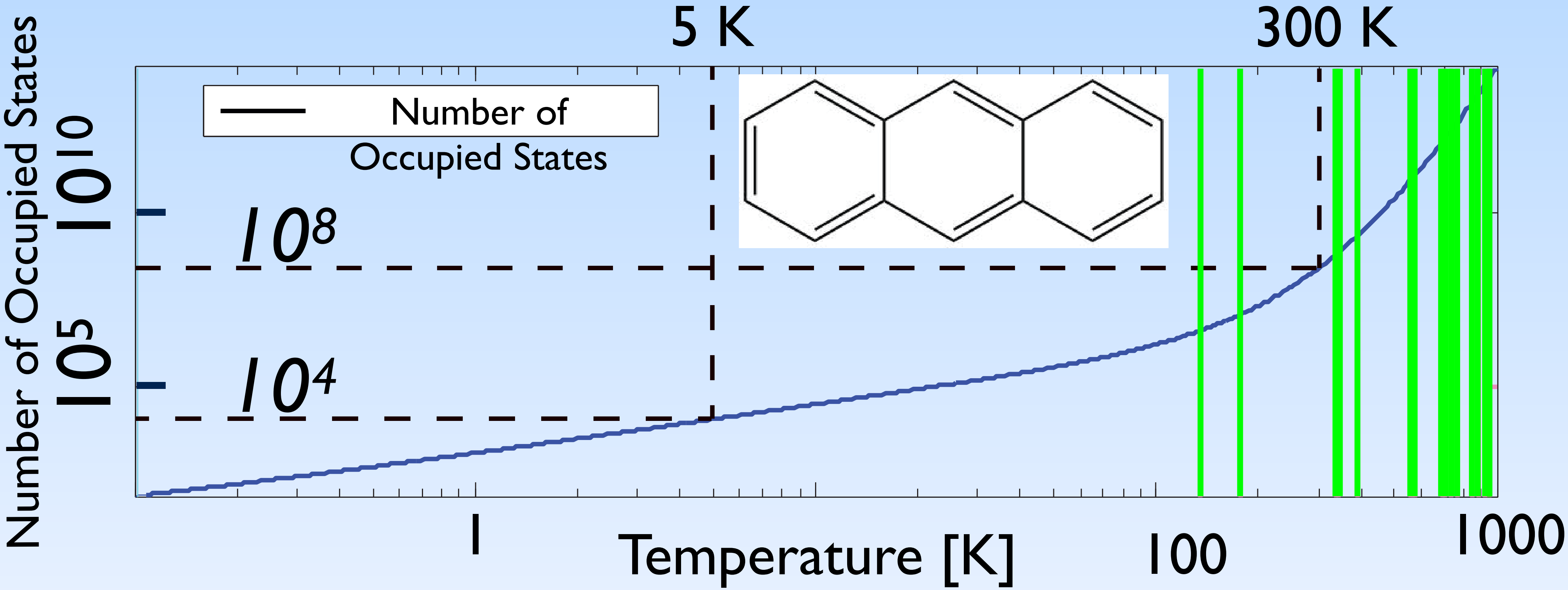
## The Universe* is Made of Molecules

SPECTROSCOPY, INTERACTIONS,….

Why cold?

# Cooling: Phase Space Compression


Important for Spectroscopy!



# More Recently, Molecular Spectroscopy with Modern Twist

**Accurate prediction and measurement of vibronic branching ratios for laser-coolable linear polyatomic molecules**

Chaoqun Zhang,[1] Benjamin L. Augenbraun,[2,3,*] Zack D. Lasner,[2,3]
Nathaniel B. Vilas,[2,3] John M. Doyle,[2,3] and Lan Cheng[1,†]

[1] *Department of Chemistry, The Johns Hopkins University, Baltimore, MD 21218, USA*
[2] *Department of Physics, Harvard University, Cambridge, MA 02138, USA*
[3] *Harvard-MIT Center for Ultracold Atoms, Cambridge, MA 02138, USA*

We report a generally applicable computational and experimental approach to determine vibronic branching ratios in linear polyatomic molecules to the $10^{-5}$ level, including for nominally symmetry-forbidden transitions. These methods are demonstrated in CaOH and YbOH, showing approximately two orders of magnitude improved sensitivity compared with the previous state of the art. Knowledge of branching ratios at this level is needed for the successful deep laser cooling of a broad range of molecular species.

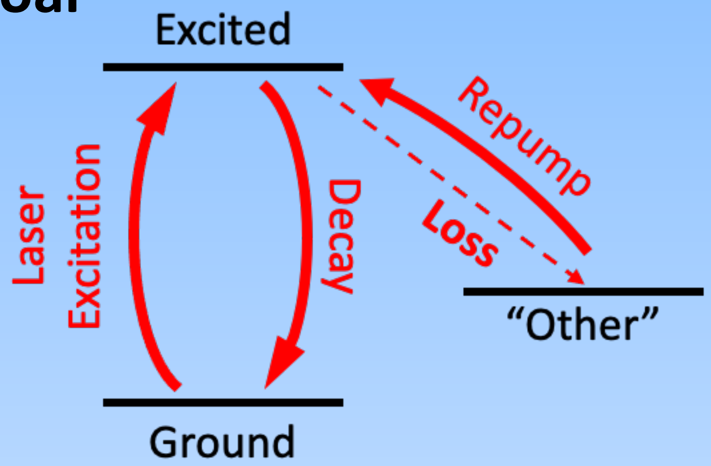
Submitted three weeks ago

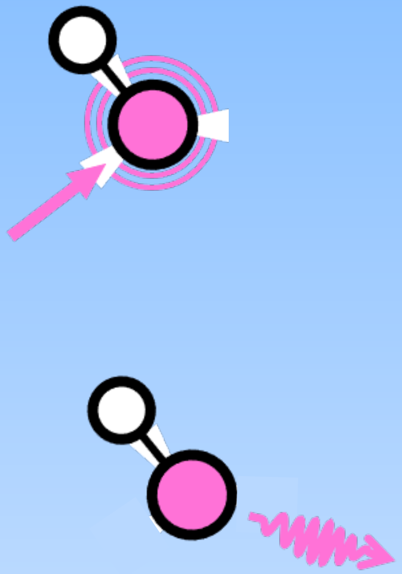
Two orders of magnitude better than previous state of the art.

# Molecular Spectroscopy with Optical Cycling

## Photon Cycling Molecules

**Goal**



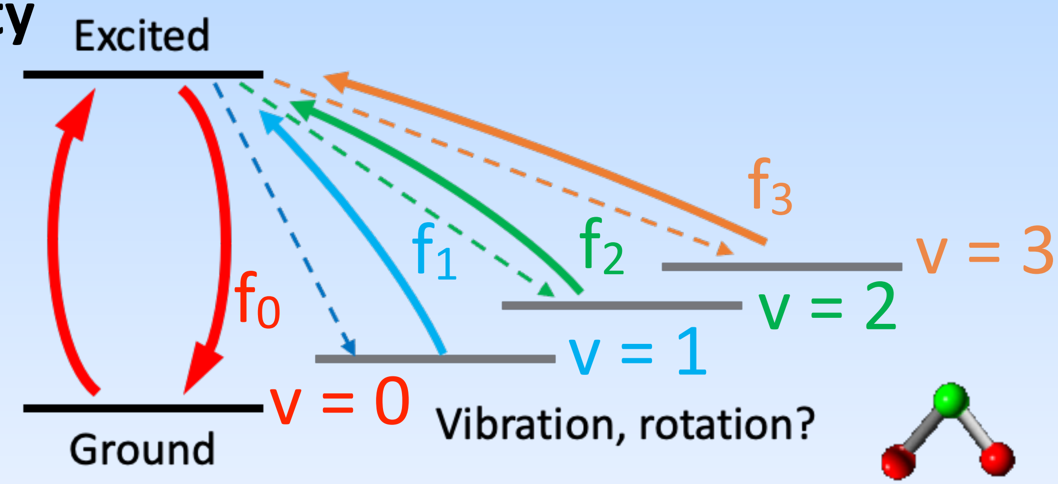
Fraction of molecules decaying
These other ro-vibrational states
Is quantified by "Frank Condon Factors" (FCF)

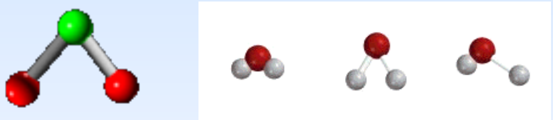
**Example of a "Highly Diagonal" Molecule**

$f_1$ = 0.02, $f_2$=0.01, $f_3$=0.003 means

means about 4% of the time the molecule decays from the excited state to vibrationally excited "off diagonal" states

**Reality**

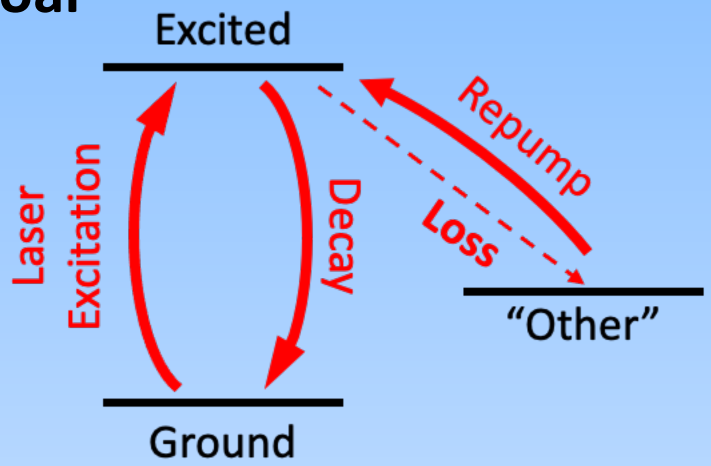


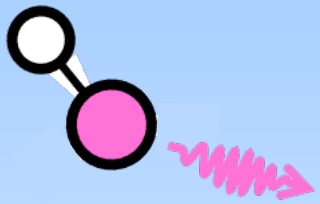



# Molecular Spectroscopy with Optical Cycling

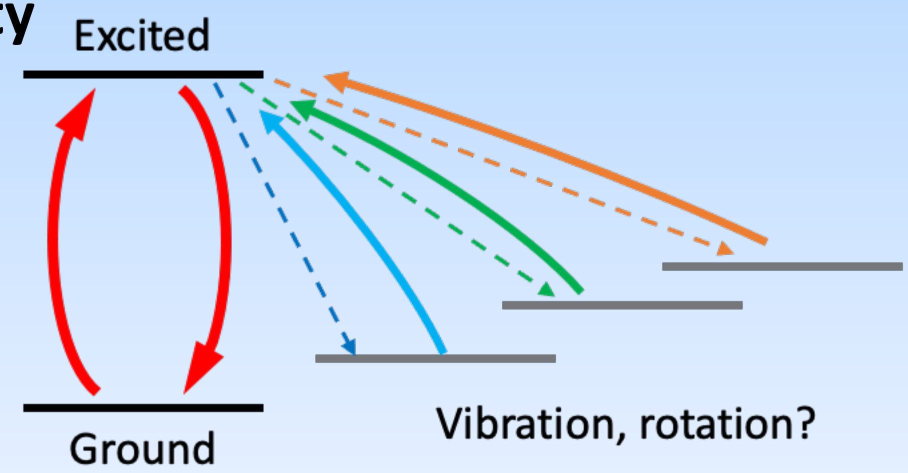
## Photon Cycling Molecules

**Goal**



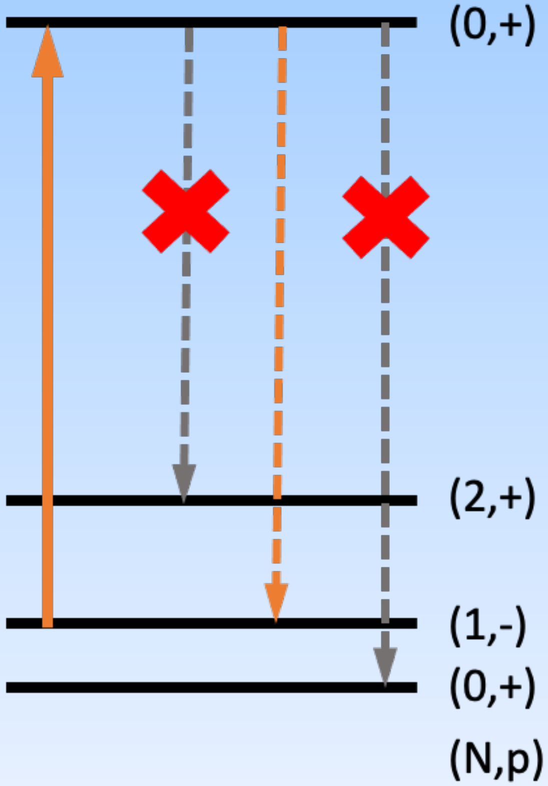



**Diatomic Reality**



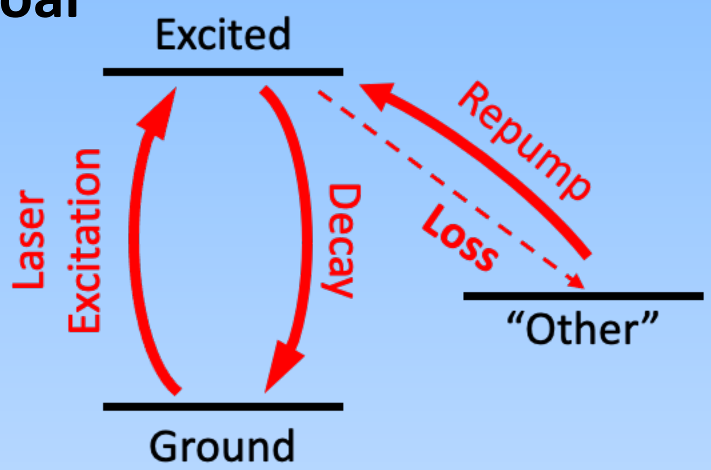

### "Diatomic Reality"

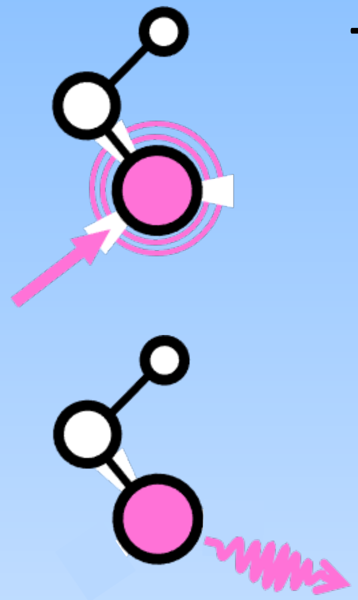
- Key factors for diatomic molecules:
  - Diagonal FCFs (of course…)
  - General **lack of excited-state perturbations**
  - **Rotationally closed transitions**



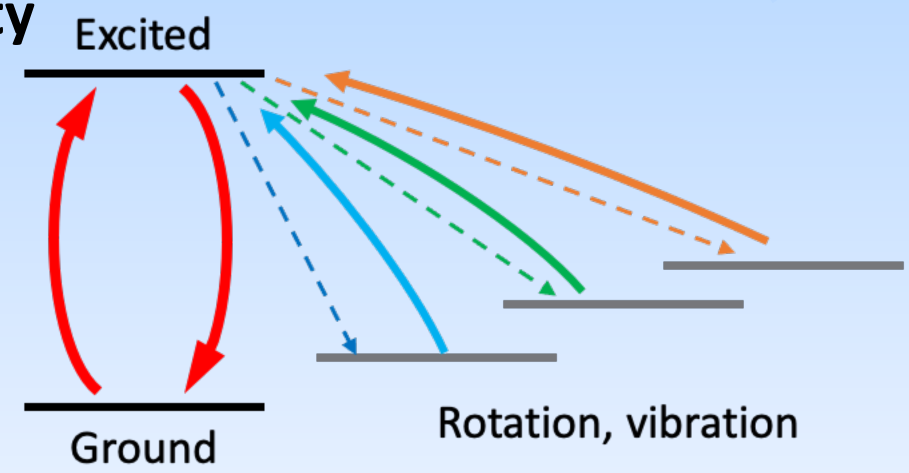
# Photon Cycling Molecules

**Goal**



**Polyatomic Reality**

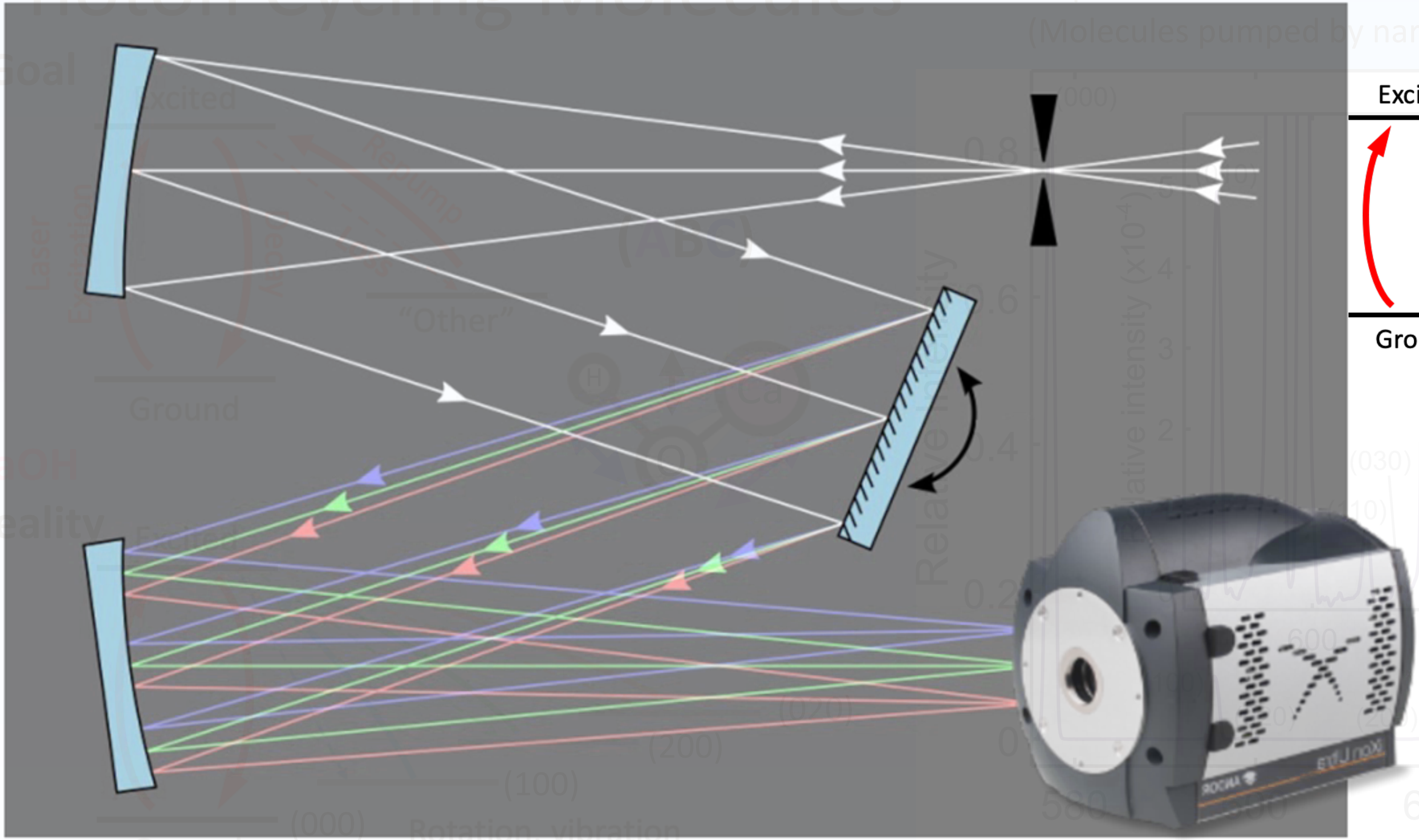


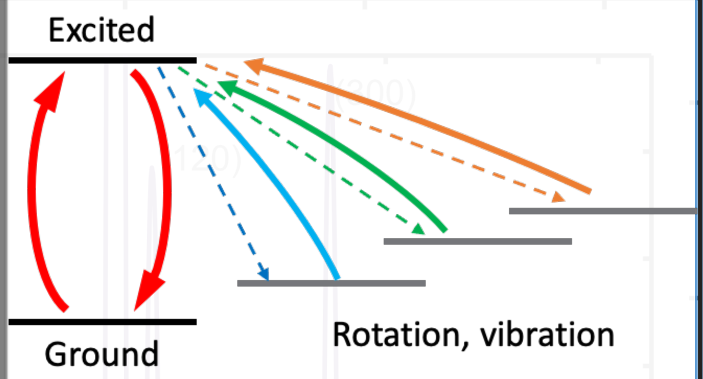

## "Polyatomic Reality?"

To photon cycle POLYATOMIC molecules, need to ask:

- Diagonal FCFs?
- **Electronic structure? Vibronic coupling?**
- **Can rotational branching be controlled?**

# Molecular Spectroscopy with Optical Cycling and a Grating Spectrometer!



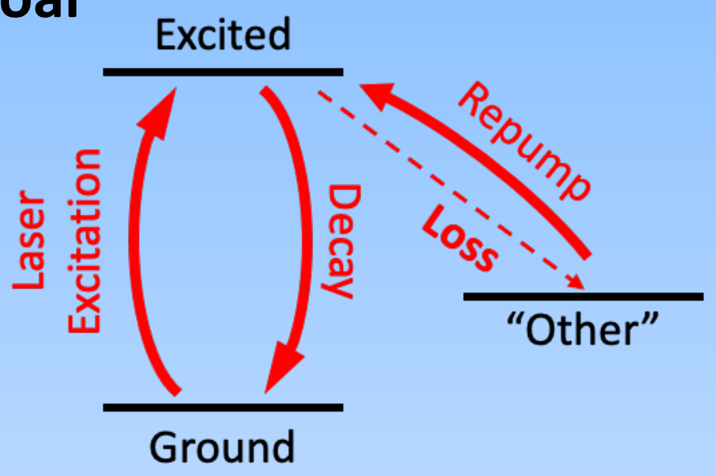



We collect the light shown
By dashed lines above

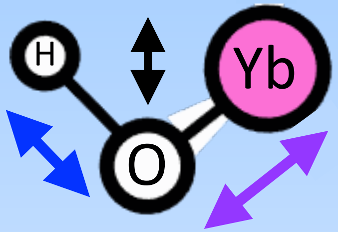
# Molecular Spectroscopy with Optical Cycling

## Photon Cycling Molecules

**Goal**

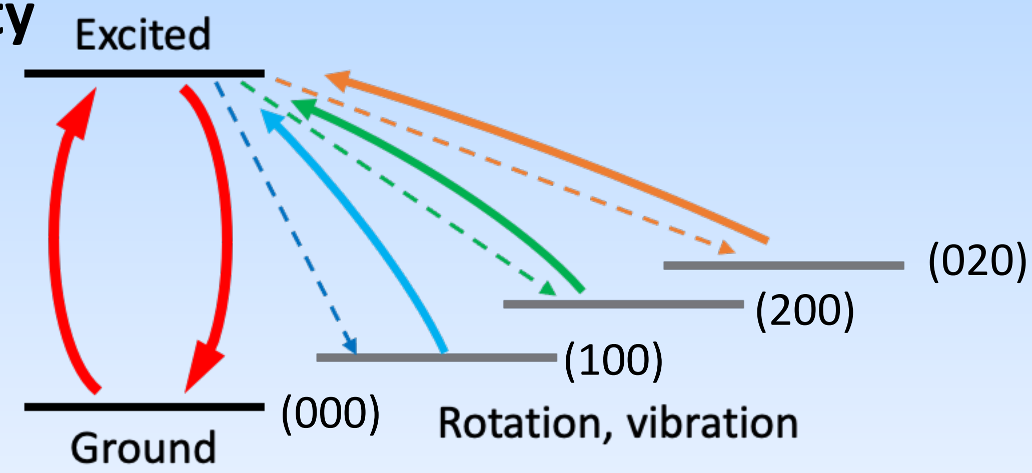



(ABC)




**CaOH**

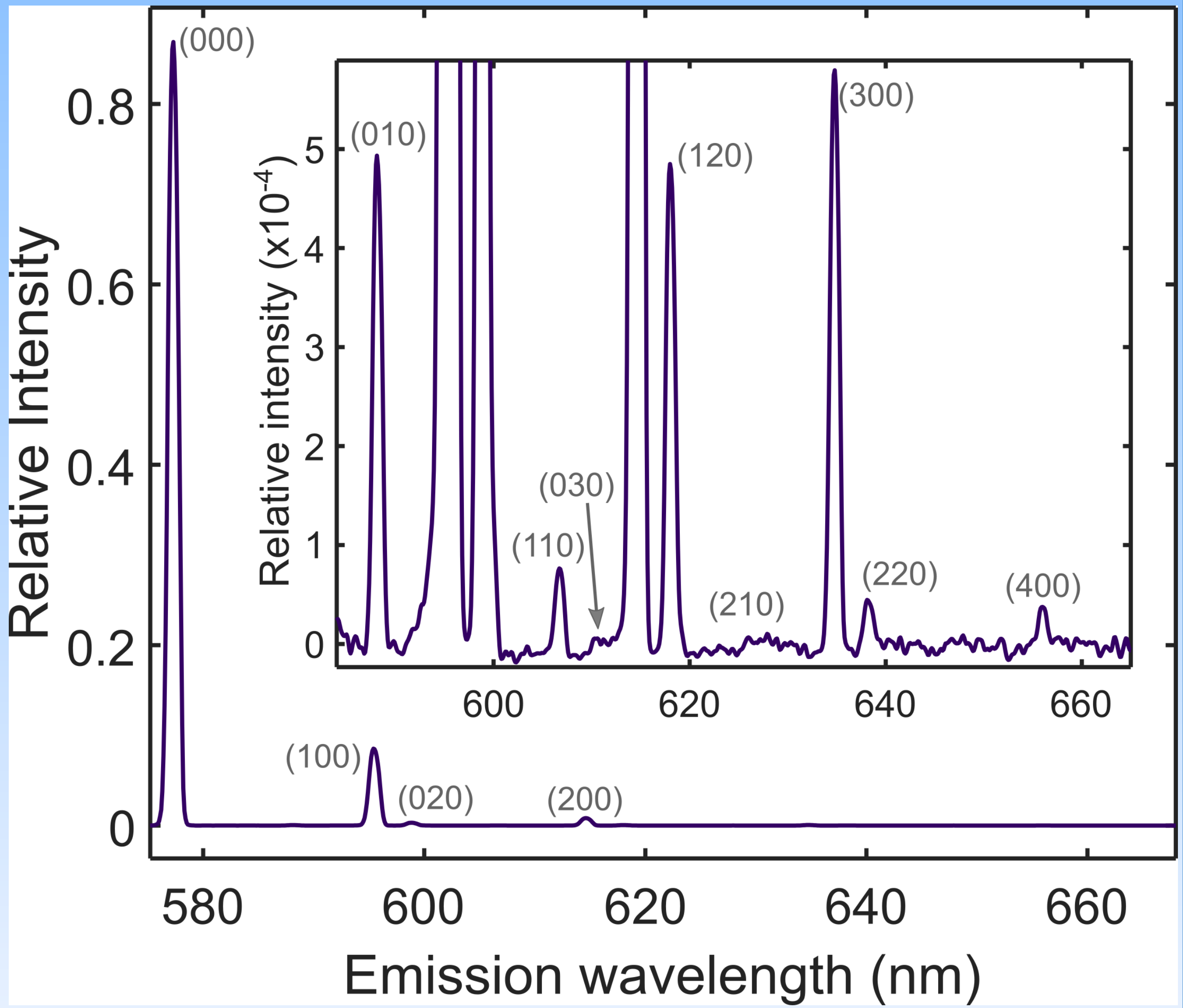
**Reality**




**YbOH Reality**

Dispersed Fluorescence - GRATING SPECTROMETER!

(Molecules pumped by narrow band CW laser :-)



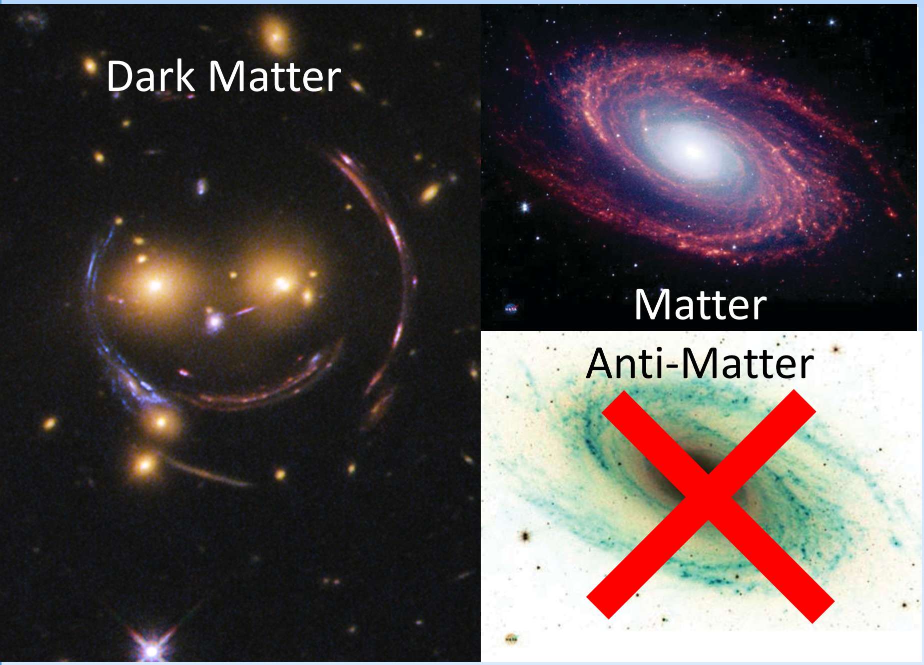
# Molecular Spectroscopy with Optical Cycling

What have we learned?

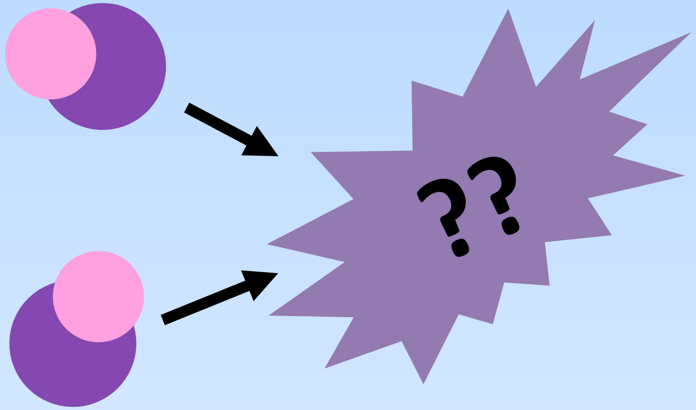
Stay tuned to the end of this talk….

# Why Cold Molecules ?? - Physics Motivation

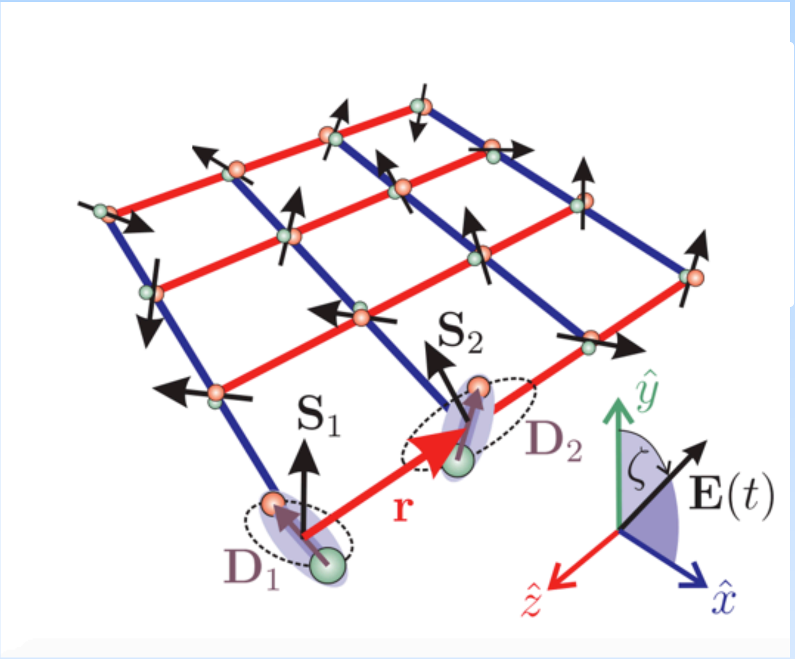
**Precision Measurements**




**Collisions and Chemistry**




**Quantum Simulation/ Information**

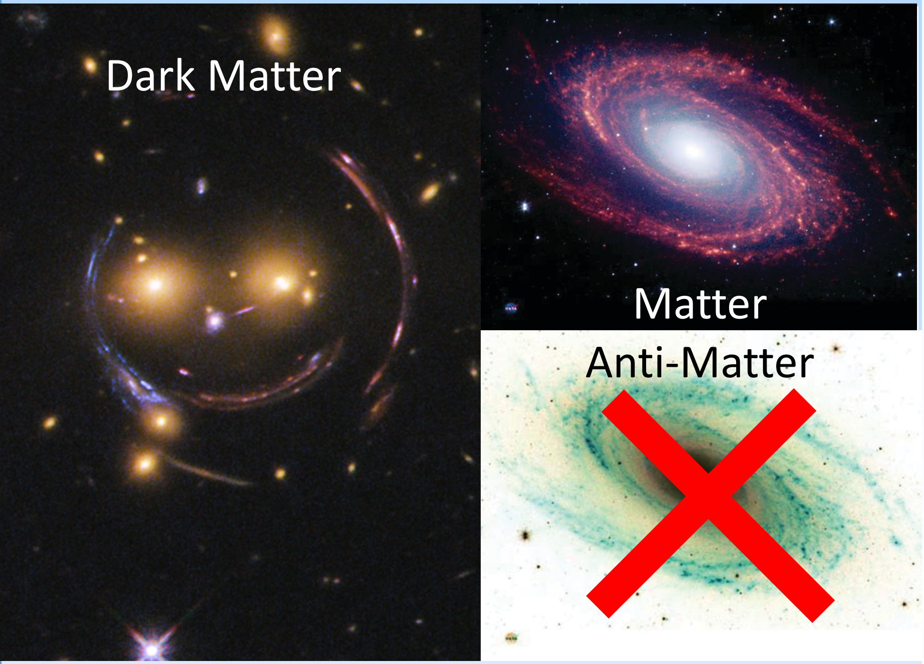



Cold → Colder → Coldest

# Answer 2

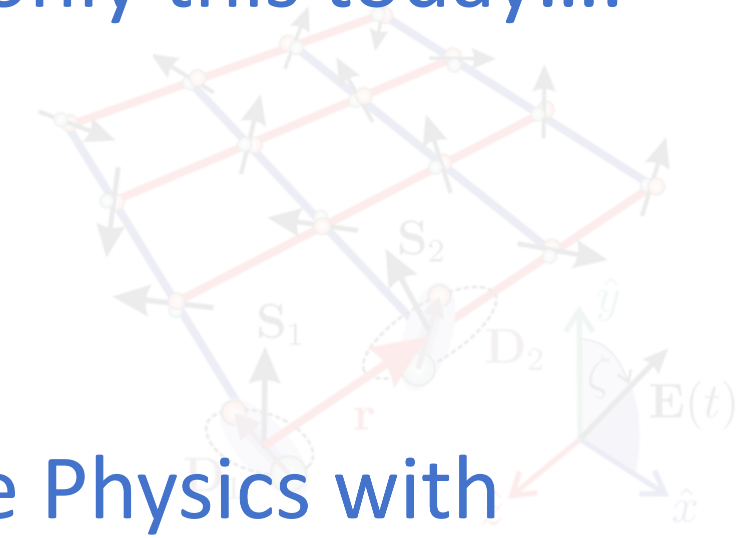
# Why Cold Molecules ??

## Precision Measurements




Collisions and Chemistry

Quantum Simulation/ Information



Let's talk about only this today….

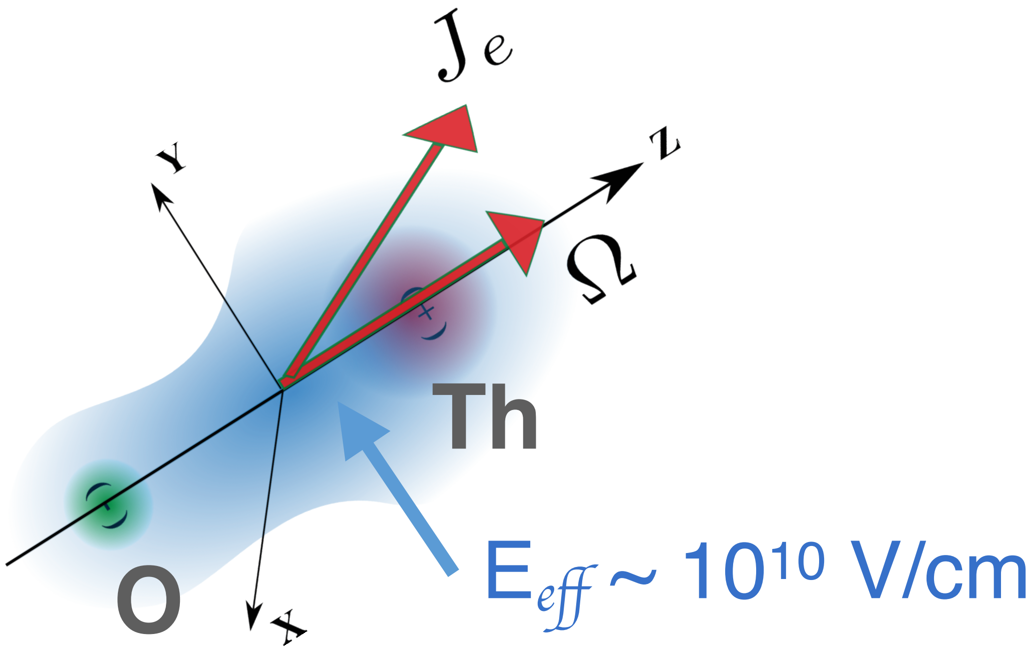
Precision Particle Physics with Cold Molecules

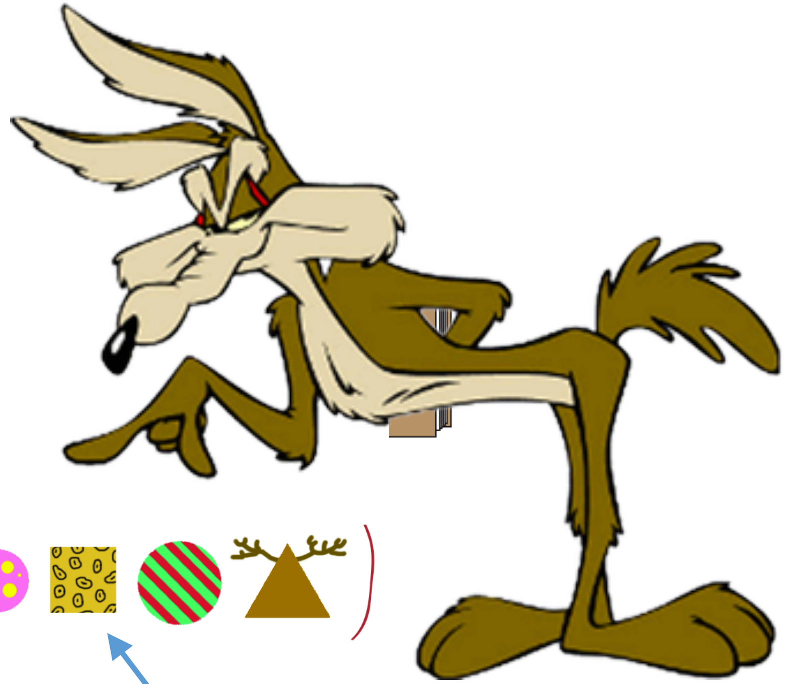
Cold

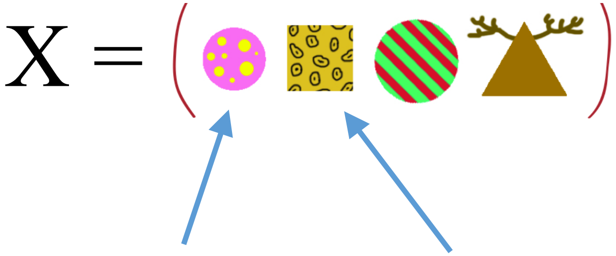
Colder

Coldest

# The ACME Experiment Using Cold ThO Molecules

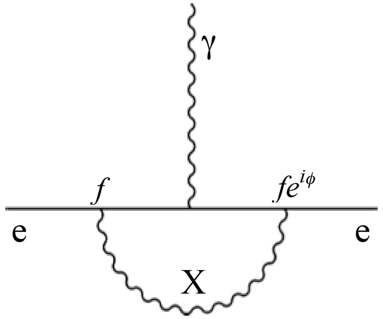


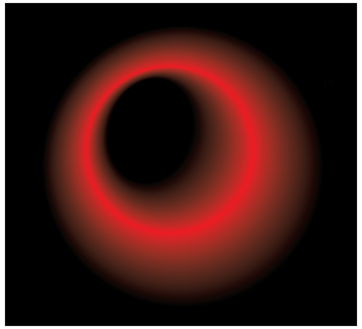






sElectron sTop

(proposed new beyond the Standard Model particles)

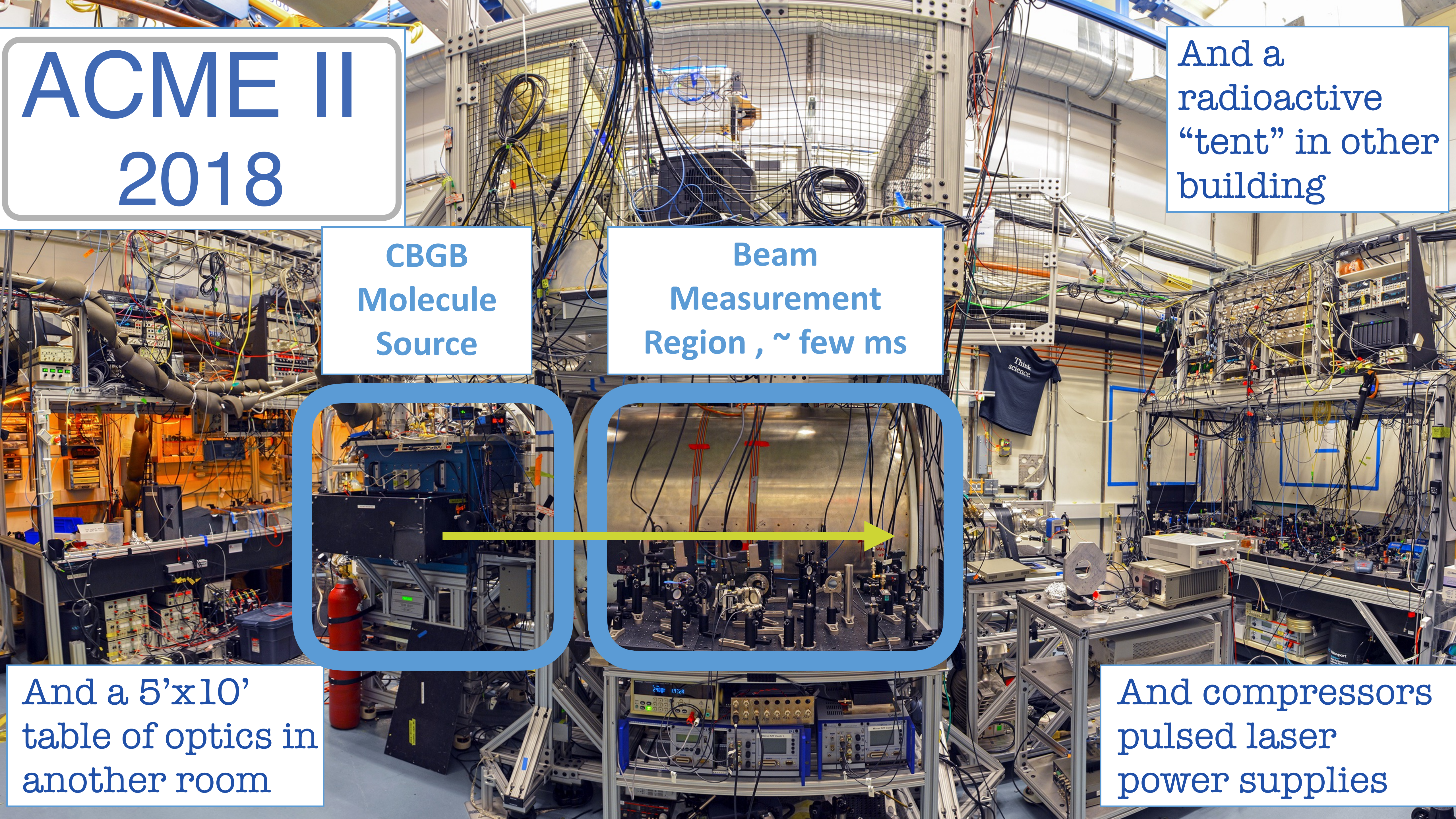



D. Ang
D. DeMille*
J.M. Doyle*
Z. Han
B. Hao
A. Hiramoto
P. Hu
G. Gabrielse*
N. Hutzler
D. Lascar
Z. Lasner
S. Liu
T. Masuda*
C. Meisenhelder
C. Panda
N. Sasao
S. Uetake
X. Wu
K. Yoshimura*

*Senior Leaders/ PIs

# ACME II 2018



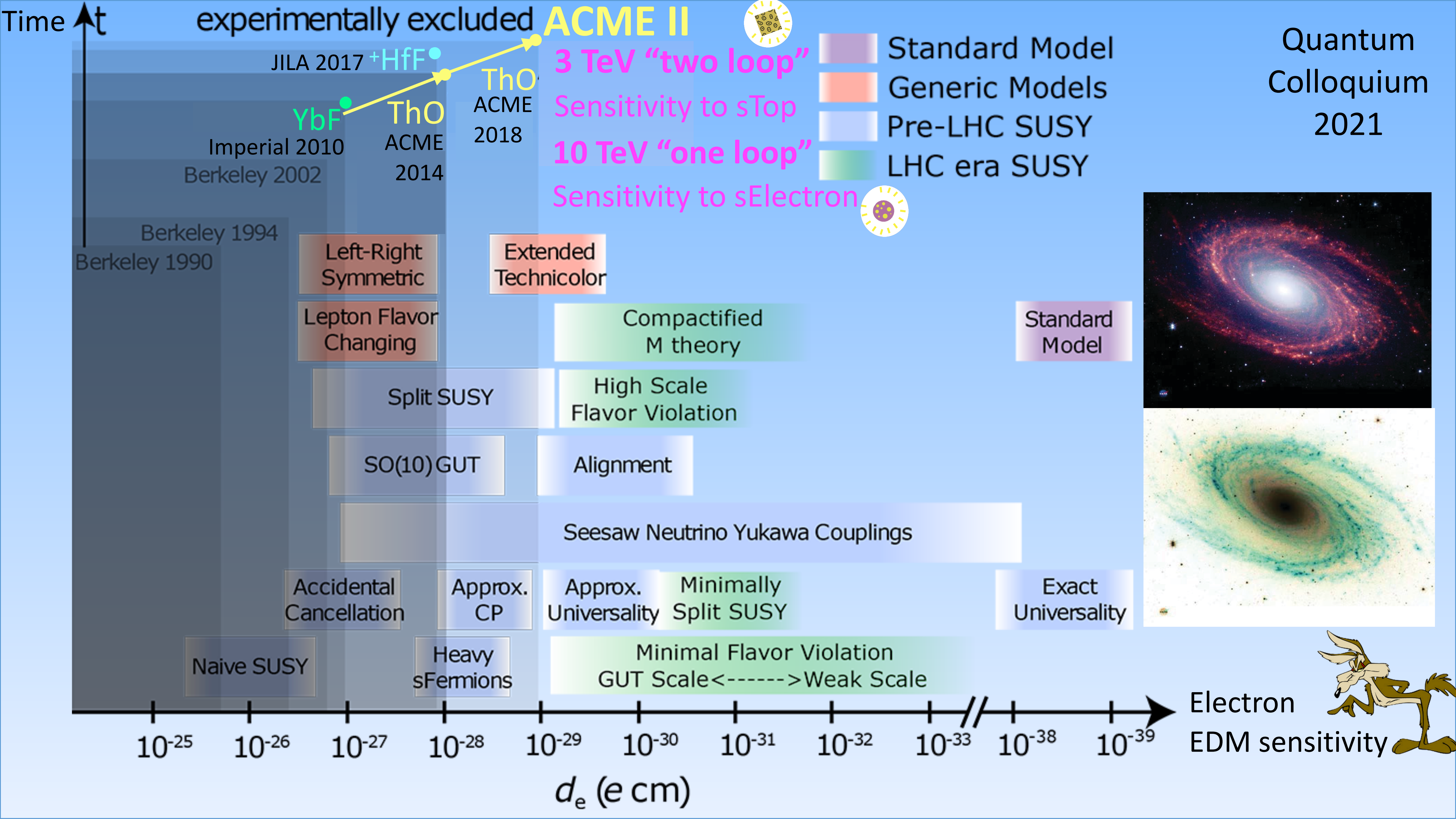
And a radioactive "tent" in other building

CBGB Molecule Source

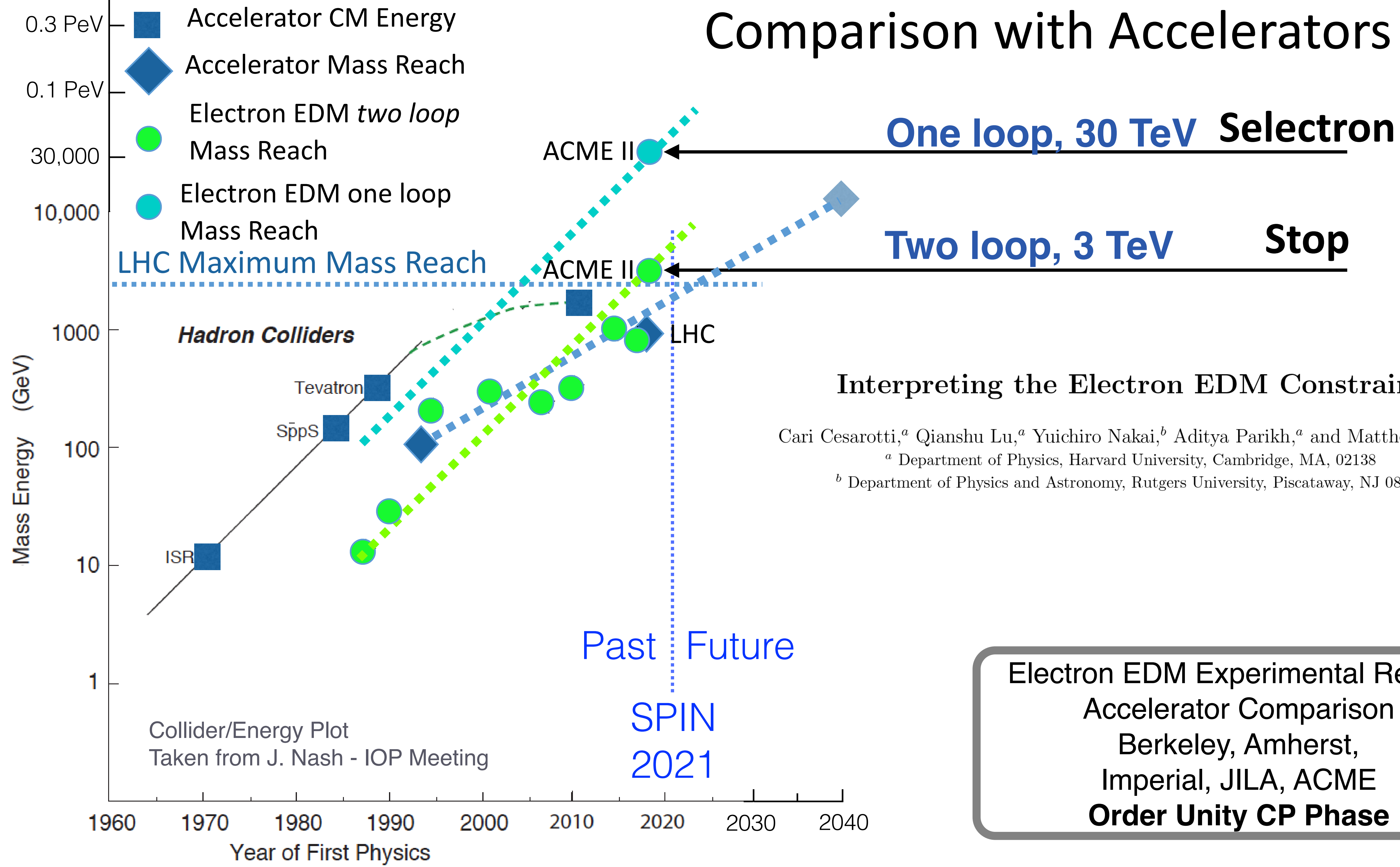
Beam Measurement Region , ~ few ms

And a 5'x10' table of optics in another room

And compressors pulsed laser power supplies


Time t
experimentally excluded
ACME II
JILA 2017 +HfF
ThO
ACME
2018
YbF
ThO
ACME
2014
Imperial 2010
Berkeley 2002
Berkeley 1994
Berkeley 1990
3 TeV "two loop"
Sensitivity to sTop
10 TeV "one loop"
Sensitivity to sElectron
Standard Model
Generic Models
Pre-LHC SUSY
LHC era SUSY
Quantum
Colloquium
2021
Left-Right
Symmetric
Extended
Technicolor
Lepton Flavor
Changing
Compactified
M theory
Standard
Model
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
$10^{-25}$ $10^{-26}$ $10^{-27}$ $10^{-28}$ $10^{-29}$ $10^{-30}$ $10^{-31}$ $10^{-32}$ $10^{-33}$ $10^{-38}$ $10^{-39}$
Electron
EDM sensitivity
$d_e$ (e cm)

# Comparison with Accelerators




Interpreting the Electron EDM Constraint

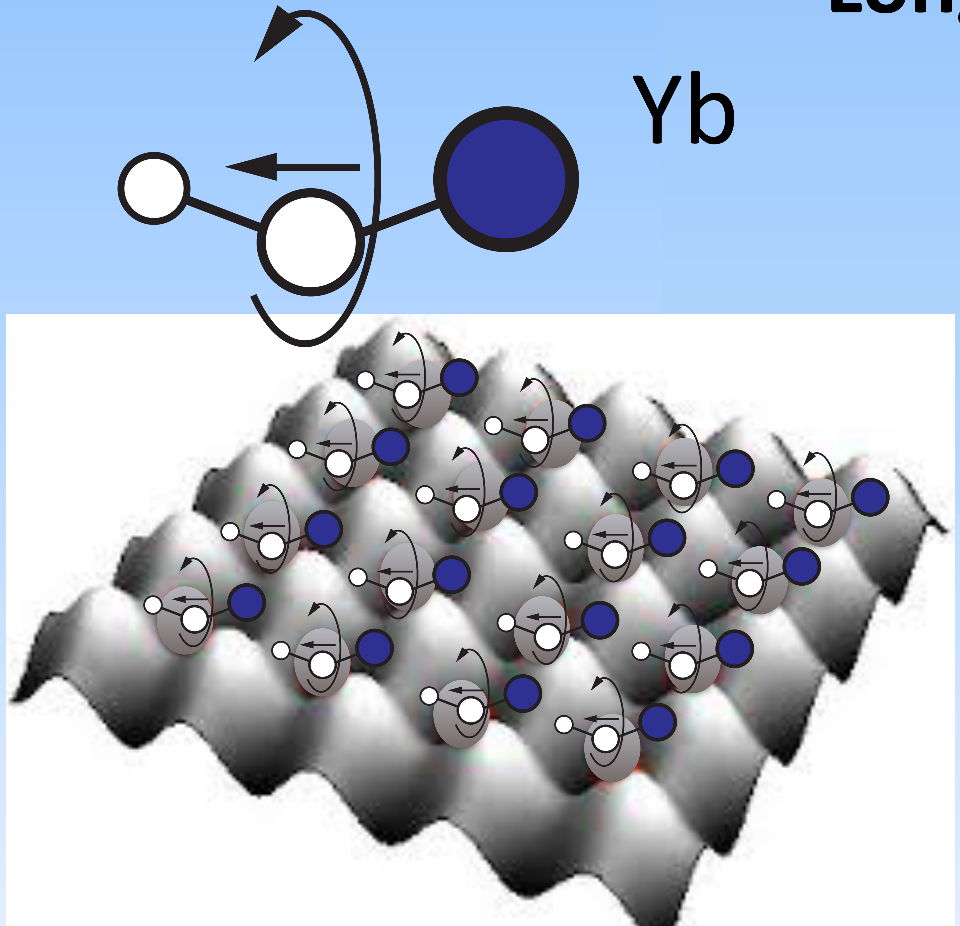
Cari Cesarotti,[a] Qianshu Lu,[a] Yuichiro Nakai,[b] Aditya Parikh,[a] and Matthew Reece[a]
[a] Department of Physics, Harvard University, Cambridge, MA, 02138
[b] Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08854

Electron EDM Experimental Results
Accelerator Comparison
Berkeley, Amherst,
Imperial, JILA, ACME
**Order Unity CP Phase**

# YbOH - A Future EDM Experiment - Polyatomics in Optical Trap

An approach based on ultracold molecules

**Long Coherence Time (~1 s), Large Numbers**
**Small Volume (< 1 mm^3)**



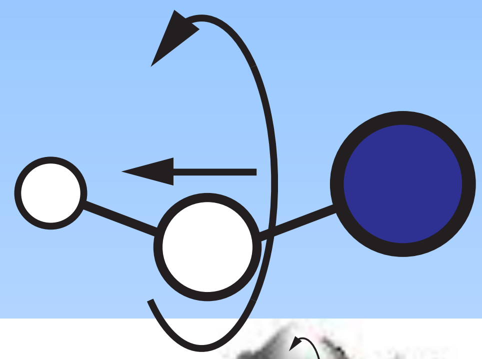

*Kozyryev and Hutzler, PRL (2017)*

1) Heavy Molecule
2) **$\ell$ doubling**
3) Large Numbers
4) Ultracold - ODT - Long $\tau$

PolyEDM Collaboration
Caltech (Hutzler and Steimle)
Harvard (Doyle)
Toronto (Vutha)

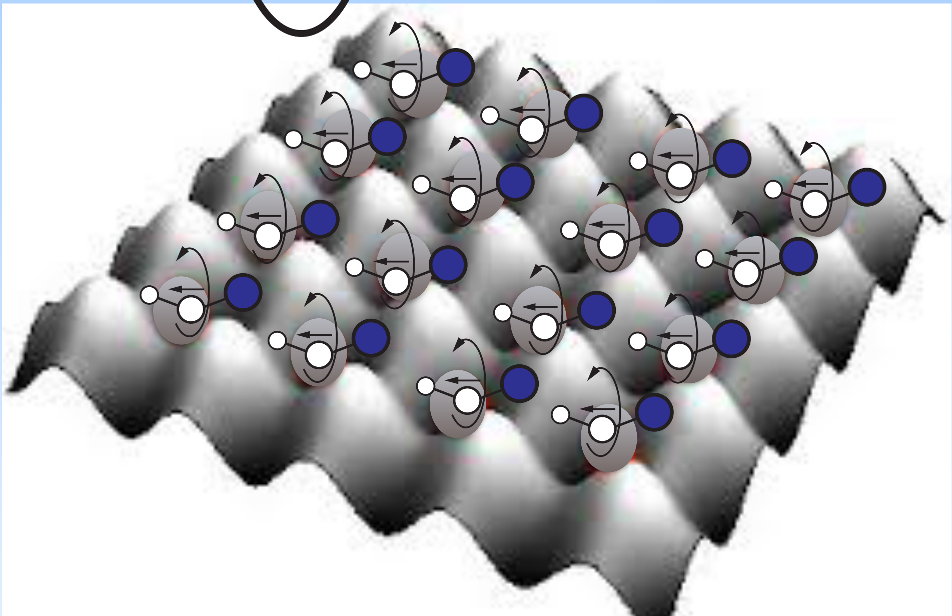
# YbOH - A Future EDM Experiment - Polyatomics in Optical Trap

An approach based on ultracold molecules




**Long Coherence Time (~1 s), Large Numbers**
**Small Volume (< 1 mm^3)**

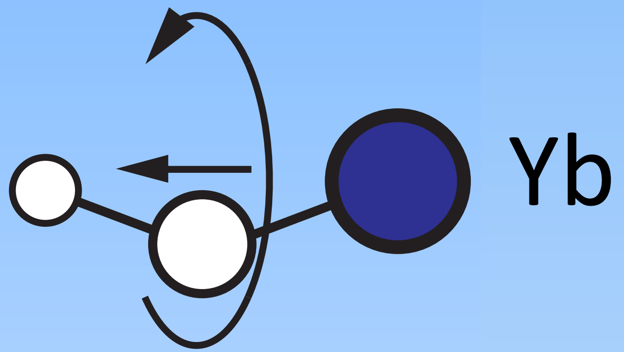
1) Heavy Molecule
2) **$\ell$ doubling**
3) Large Numbers
4) Ultracold - Optical Lattice - Long $\tau$



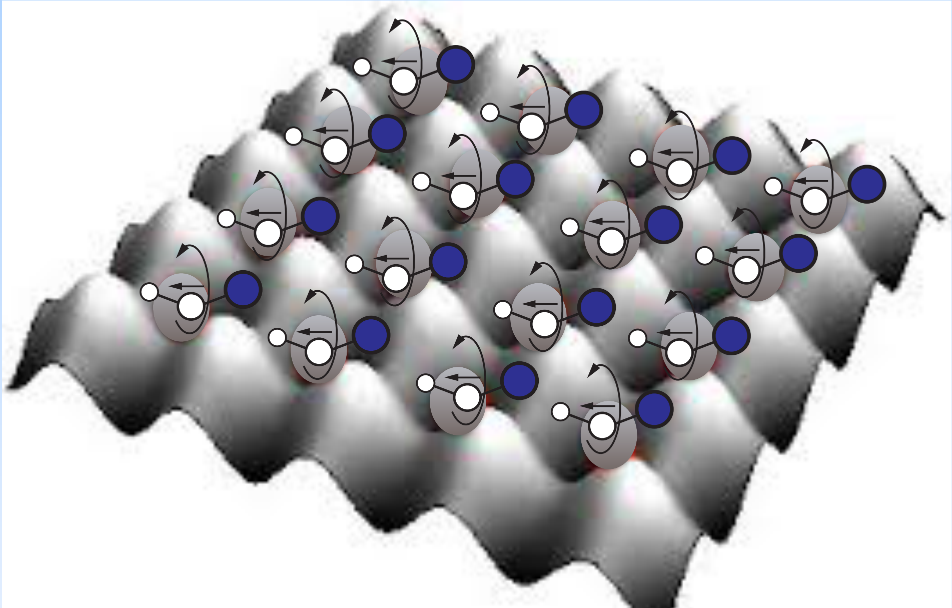
*Kozyryev and Hutzler, PRL (2017)*

**PolyEDM Collaboration**
**Caltech (Hutzler and Steimle)**
**Harvard (Doyle)**
**Toronto (Vutha)**

# YbOH - Future EDM - Polyatomics in Optical Trap



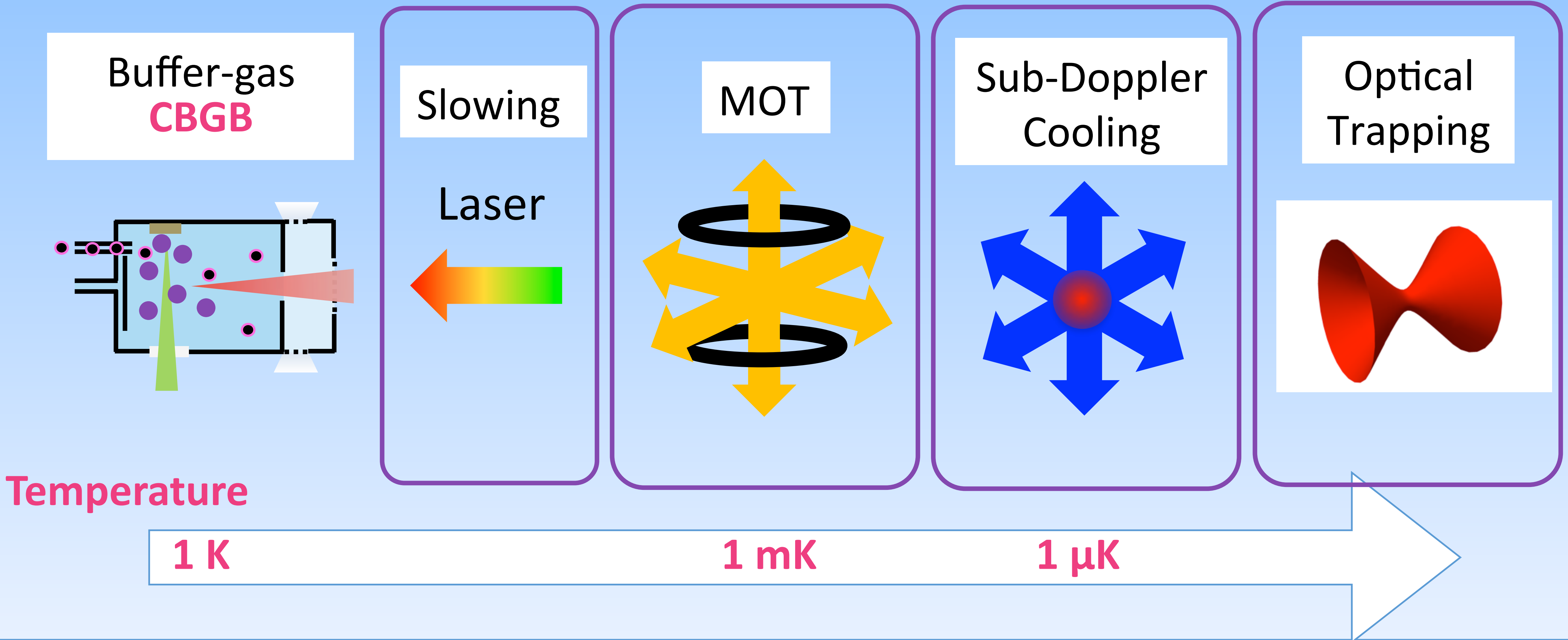

“PolyEDM” Experiment



*Kozyryev and Hutzler, PRL (2017)*

? How to get Ultracold ?

# Molecule Laser Cooling and Trapping
# Molecule Roadmap



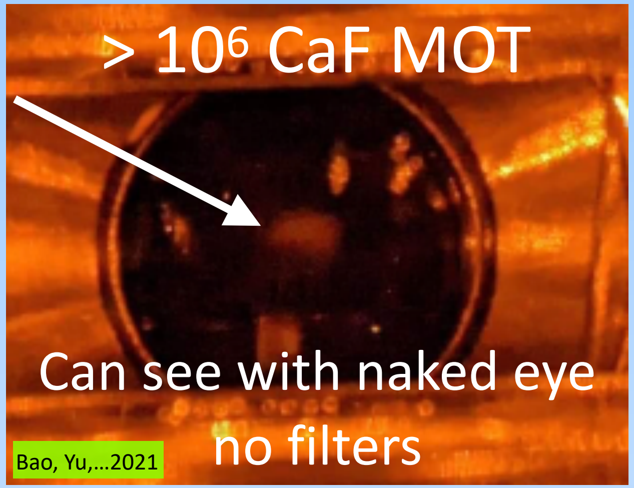
# 2021: MOT, Sub-Doppler, Mag Trap, ODT, *MORE*

| Yale<br>(SrF) | Imperial<br>(CaF) | Harvard<br>(CaF) | JILA<br>(YO) |
|---|---|---|---|
| **First diatomic MOT<br>Sub-Doppler cooled<br>Magnetically trapped** | **First sub-Doppler cooling<br>Magnetically trapped** | **First optically trapped<br>First Λ-Enhanced<br>Largest MOT number<br>Highest density** | **First oxide molecule<br>Potential for narrow line cooling** |
| **MOT: 1 x $10^4$ molecules<br>Sub-Doppler: 50 μK<br>Mag trap: $10^3$ molecules<br>@ 3 x $10^4$ $cm^{-3}$ (in single state)<br>Polarization enhanced cooling in optical trap 2021** | **MOT: 2 x $10^4$ molecules<br>Sub-Doppler: 50 μK<br>Mag trap: 5 x $10^3$ molecules<br>@ 1.2 x $10^5$ $cm^{-3}$<br>Rotational Coherence,<br>Magnetic Trap 2020<br>Atom-molecule Collisions 2021** | **MOT: $10^5$ molecules<br>Λ-Enhanced SD: 5 μK<br>ODTrap: $10^3$ molecules<br>@ 6 x $10^8$ $cm^{-3}$<br>Best MOT is $10^6$ Molecules<br>And…see upcoming slides, 2021** | **MOT: $10^3$ molecules,<br>20 ms lifetime<br>Best MOT is $10^5$ Molecules<br>Sub-Doppler cooling and compression, 4 μK 2020<br>Optical lattice 2021** |
| Barry…Nature (2014)<br>Steinecker… ChemPhysChem (2016)<br>McCarron… PRL (2018) | Truppe…Nat. Phys. (2017)<br>Williams…PRL (2018) | Anderegg…PRL (2017)<br>Anderegg…Nat. Phys. (2018)<br>Cheuk…PRL (2018) | Collopy…PRL (2018) |

# CaF - Doyle Group Super Quick Summary

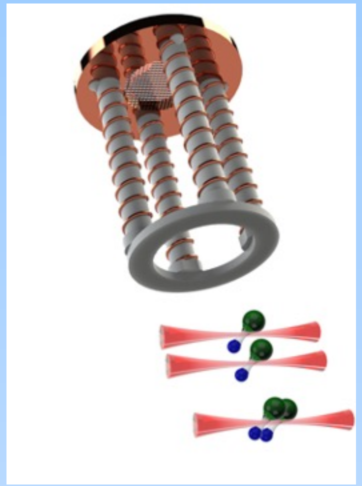
Look!

Robust MOT
and ODT
of CaF



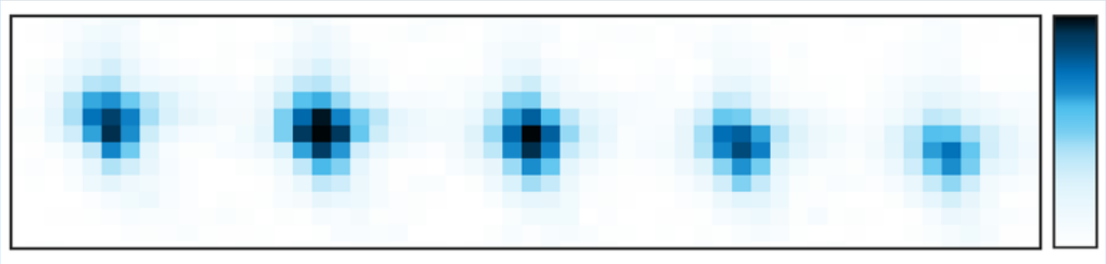

L.Anderegg, B. L Augenbraun, E. Chae, B. Hemmerling, N. R Hutzler, A. Ravi, A. Collopy, J. Ye, W. Ketterle, J. M Doyle. PRL (2017)

Suppression
Of collisions using
Microwaves



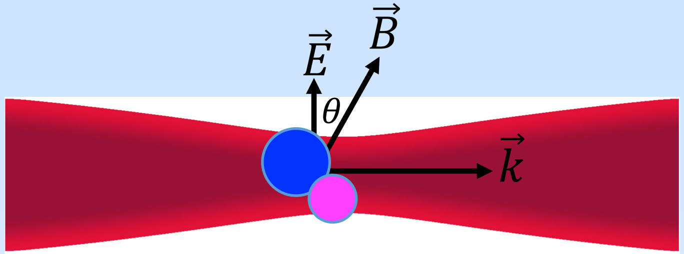
L. Anderegg, S. Burchesky, Y. Bao, S. Yu, T. Karman, E. Chae, K.-K. Ni, W. Ketterle, J.M. Doyle, Science 2021

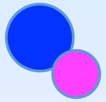
Single
Molecules
In Optical
Tweezer Array



L. Anderegg, L. W. Cheuk, Y. Bao, S. Burchesky, W. Ketterle, K-K. Ni, J. M. Doyle.. Science 2019

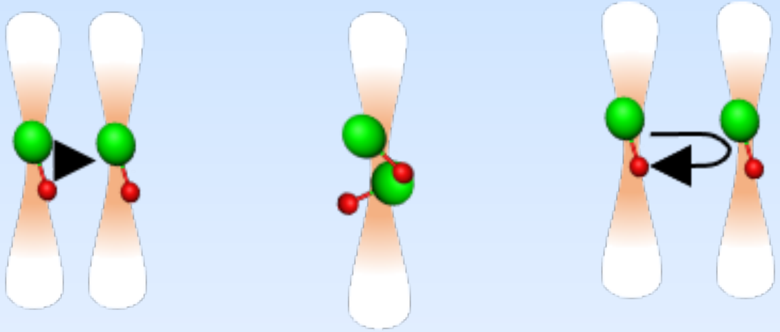
100 ms Ramsey
Coherence times
Of Rotational Qubits




Optical Tweezer
with a  inside

S. Burchesky, L. Anderegg, Y. Bao, S. Yu, E. Chae, W. Ketterle, K.-K. Ni, J.M. Doyle, PRL 2021

Two Molecule
Collisions,
Merged Tweezers

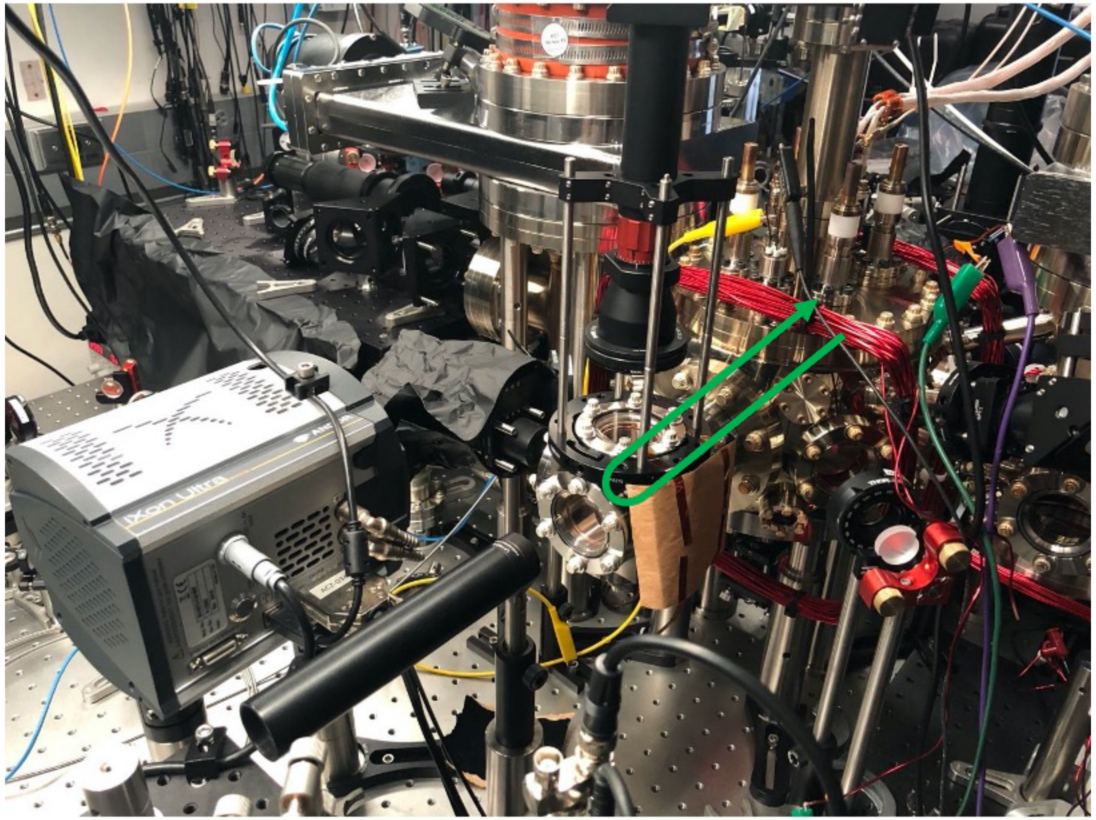


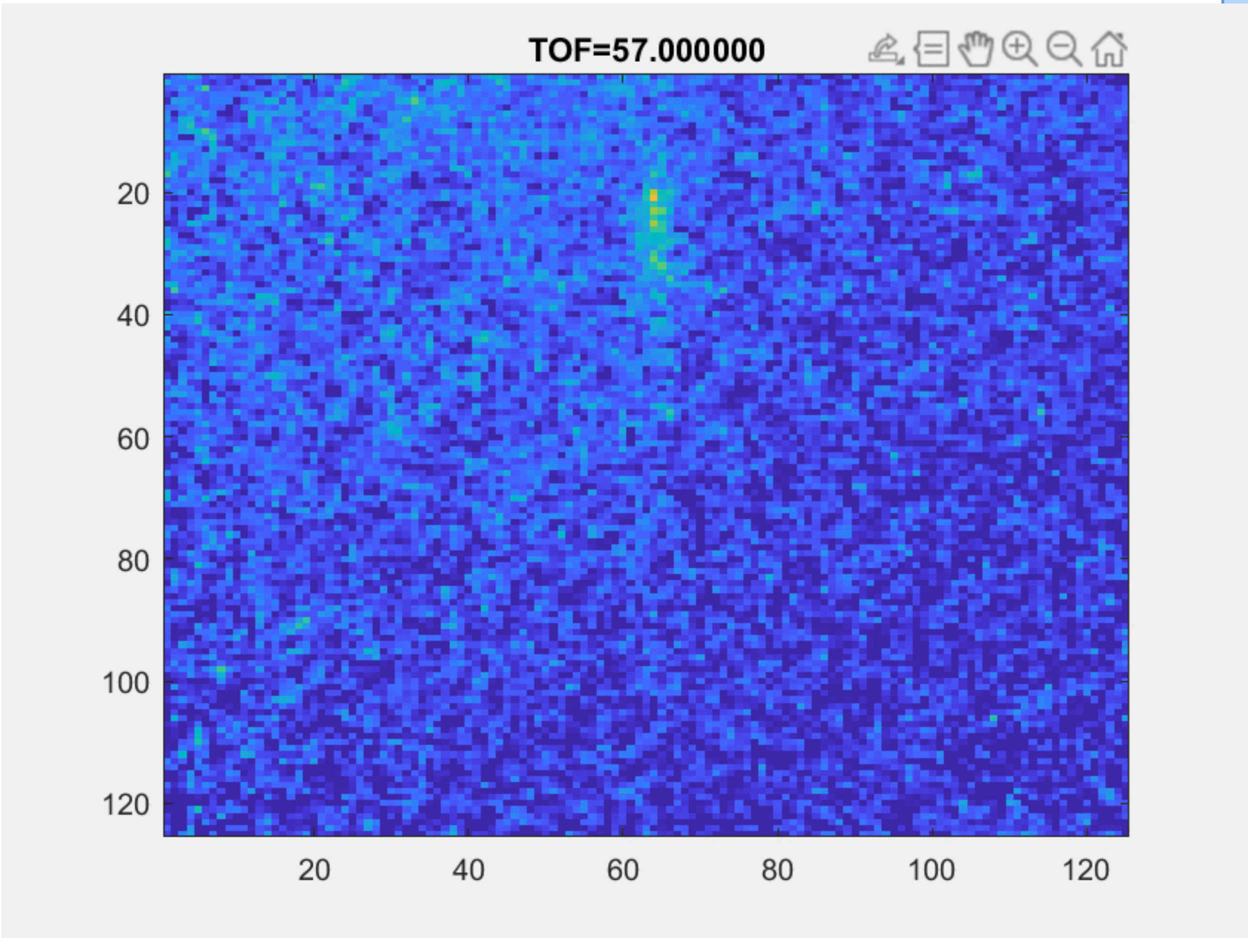
L. W. Cheuk, L. Anderegg, Y. Bao, S. Burchesky, S. Yu, W. Ketterle, K-K. Ni, J. M. Doyle. PRL 2020

# CaF - Doyle Group Super Quick Summary

Two Weeks Ago

## Transport in the Optical Dipole Trap, ~40 cm





# CaF - Doyle Group Summary


Look!

Robust MC
and ODT
of CaF

Single
Molecules
In Optical
Tweezer Ar

Two Mole
Collision
Merged Twe

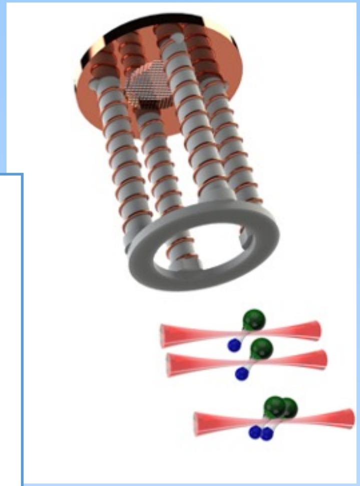
Optical Cycling is Very Powerful

Quantum State Readout

Opens up the toolbox of the ultracold,
e.g. VSCPT, Grey Molasses, etc.,
Just like atoms

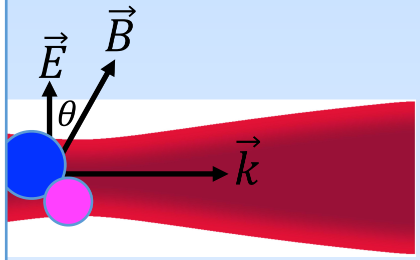
L. W. Cheuk, L. Anderegg, Y. Bao, S. Burchesky, S. Yu,
W. Ketterle, K-K. Ni, J. M. Doyle. PRL 2020

Suppression

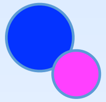


S. Burchesky, Y. Bao, S. Yu, T. Karman,
. Ni, W. Ketterle, J.M. Doyle, Science 2021

s Ramsey
nce times
onal Qubits




cal Tweezer

a  inside

S. Burchesky, L. Anderegg, Y. Bao, S. Yu, E. Chae, W. Ketterle, K.-K. Ni, J.M. Doyle, PRL 2021

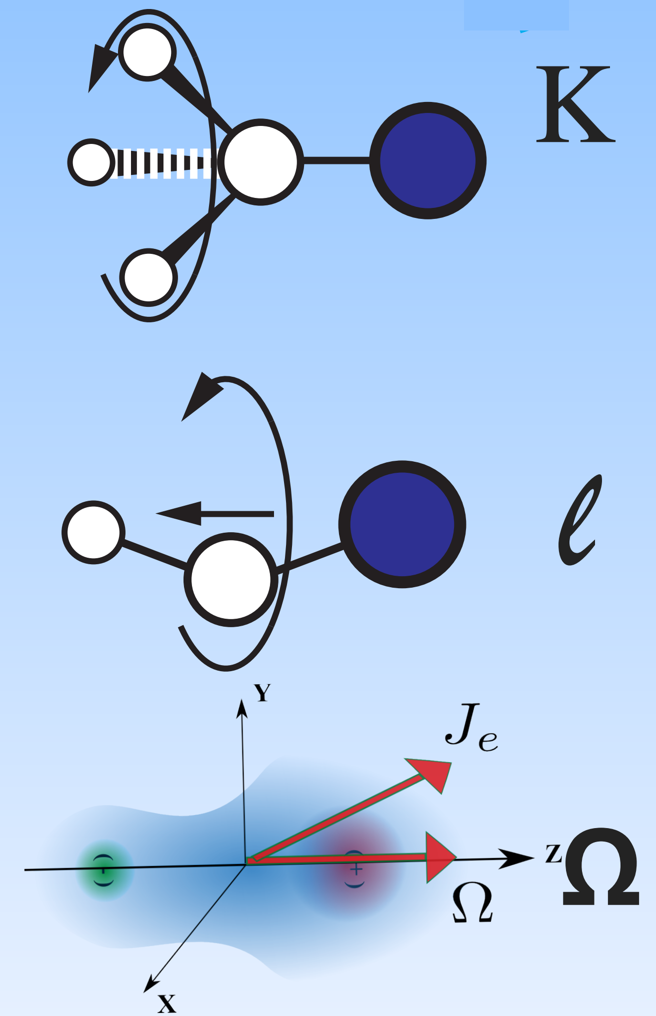
# Now to Polyatomic Molecules (Hold on to the steering wheel...)

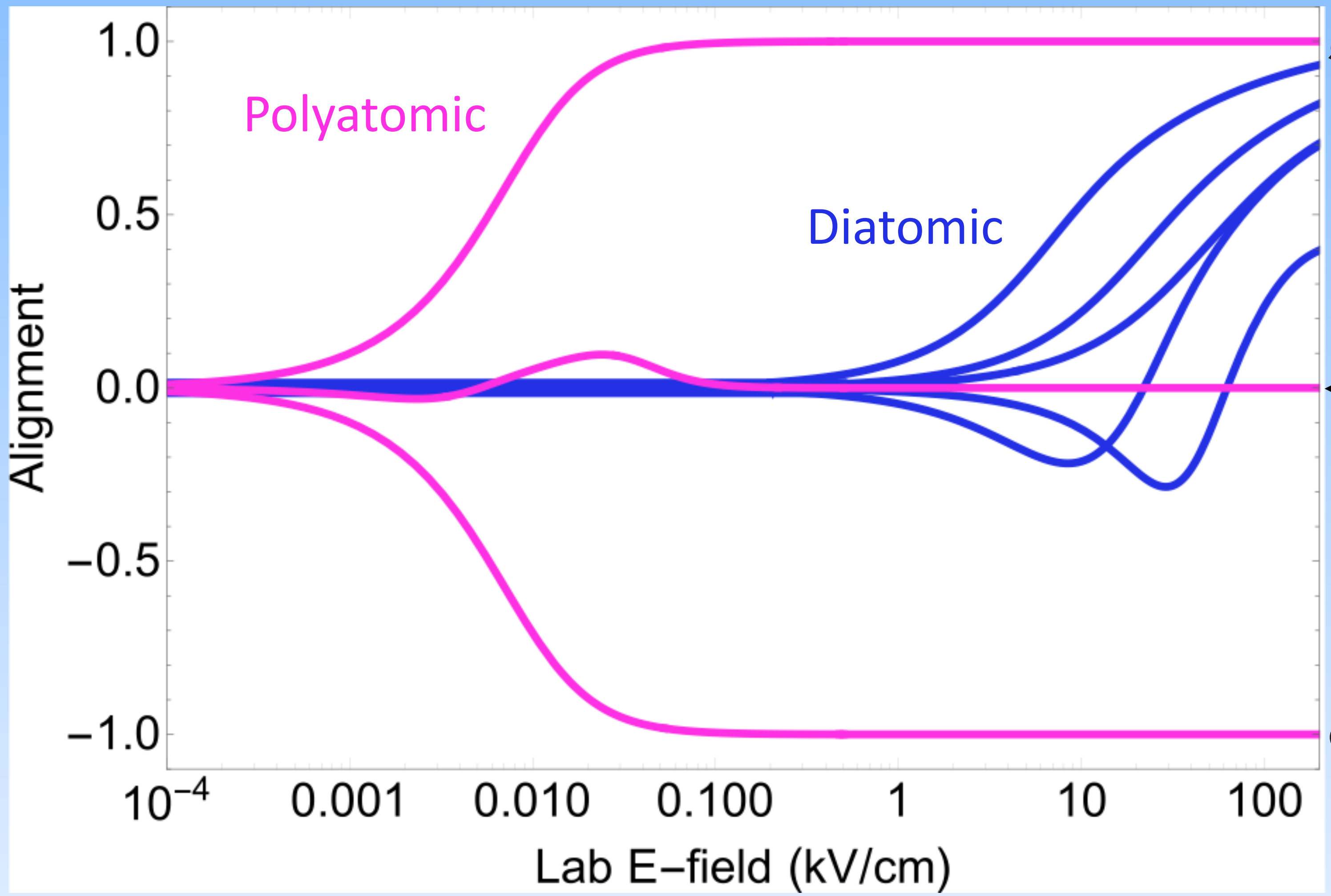

2018 Porsche 718 Cayman GTS

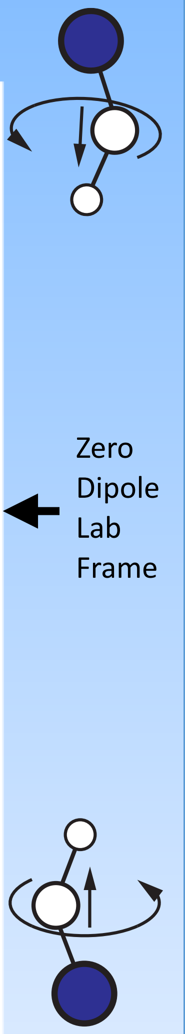
# Very Important Generic Feature of Polyatomic Molecules

# Finite Orbital Angular Momentum Along the Internuclear Axis

## Molecular Frame Quantum Numbers

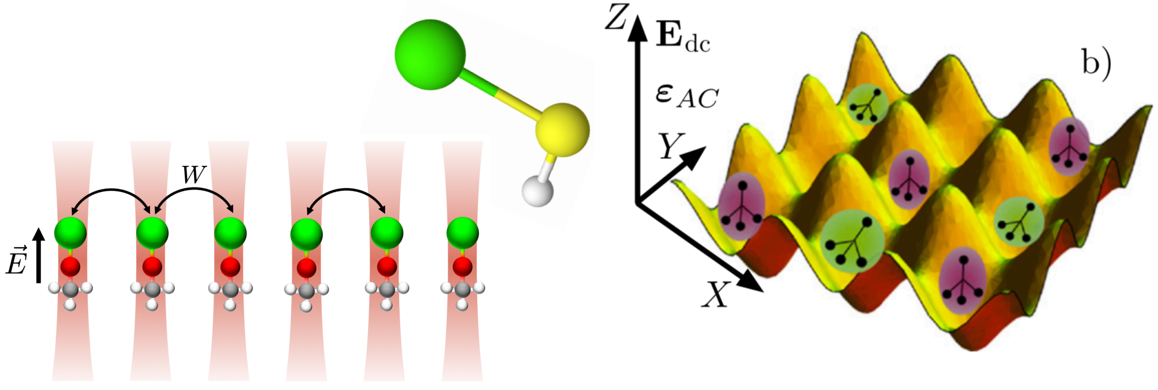


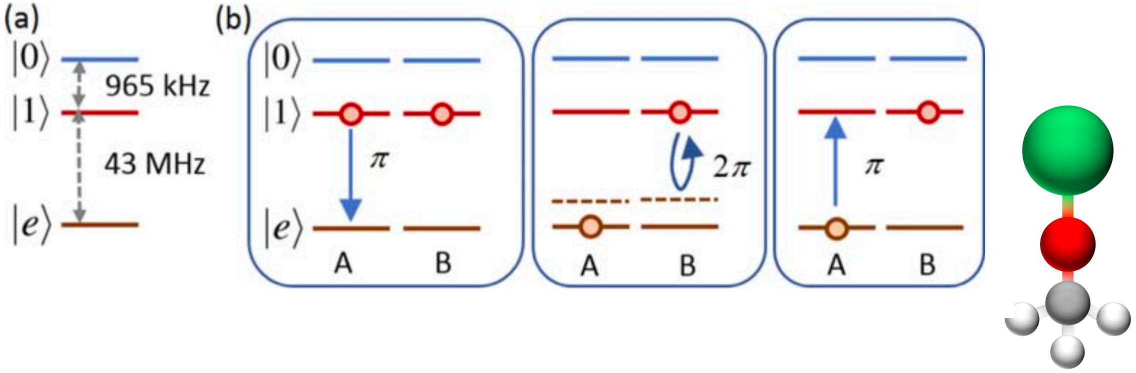
# Motivations for Polyatomic Molecules

## Quantum Simulation



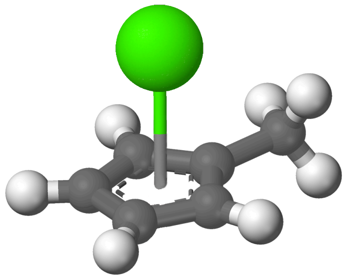

Wall, Maeda, and Carr, Ann. Phys. **525**, 845 (2013)
Wall, Maeda, and Carr, NJP **17**, 021001 (2015)

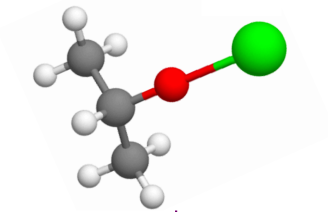
## Quantum Information

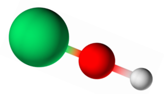



Yu,….JMD, NJP 21, 093049 (2019)
Albert et al., arXiv:1911.00099

## Ultracold Chemistry and Collisions

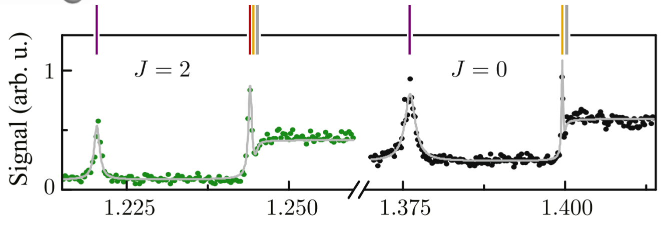






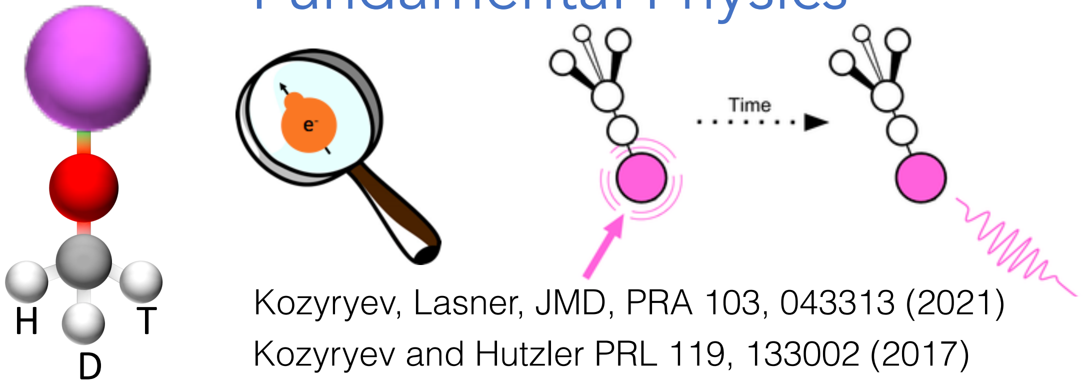
Augustovičová and Bohn, NJP **21** (2019)

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




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)

# *Laser Cooling* Polyatomic *Molecules*

October 2016 ChemPhysChem Paper

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]

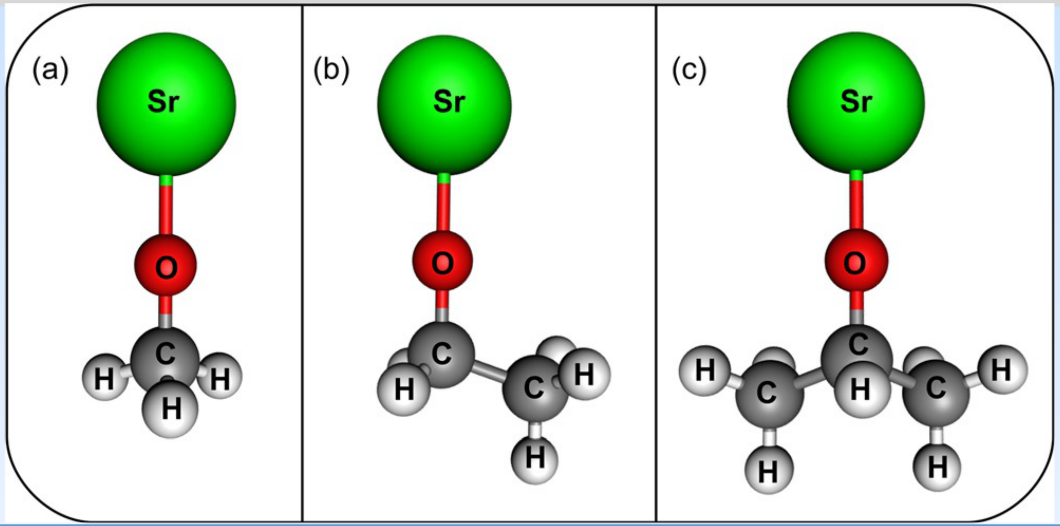
An experimentally feasible strategy for direct laser cooling of polyatomic molecules with six or more atoms is presented. Our approach relies on the attachment of a metal atom to a complex molecule, where it acts as an active photon cycling site. We describe a laser cooling scheme for alkaline earth 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

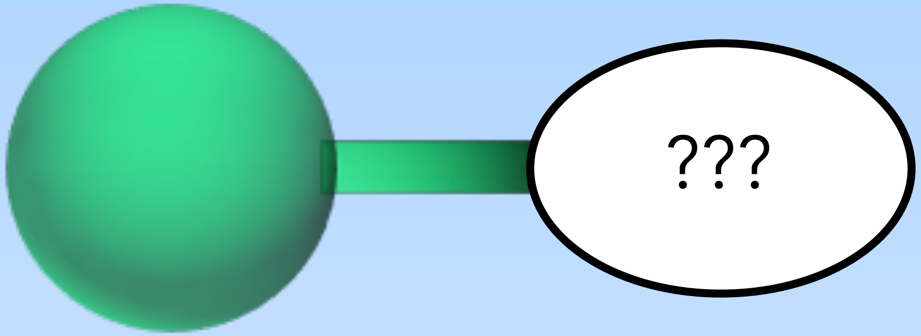
"MOR"

Molecules

a.k.a Alkaline Earth Atom Pseudofluoride Molecules



# *Molecule Laser Cooling is Generalizable*

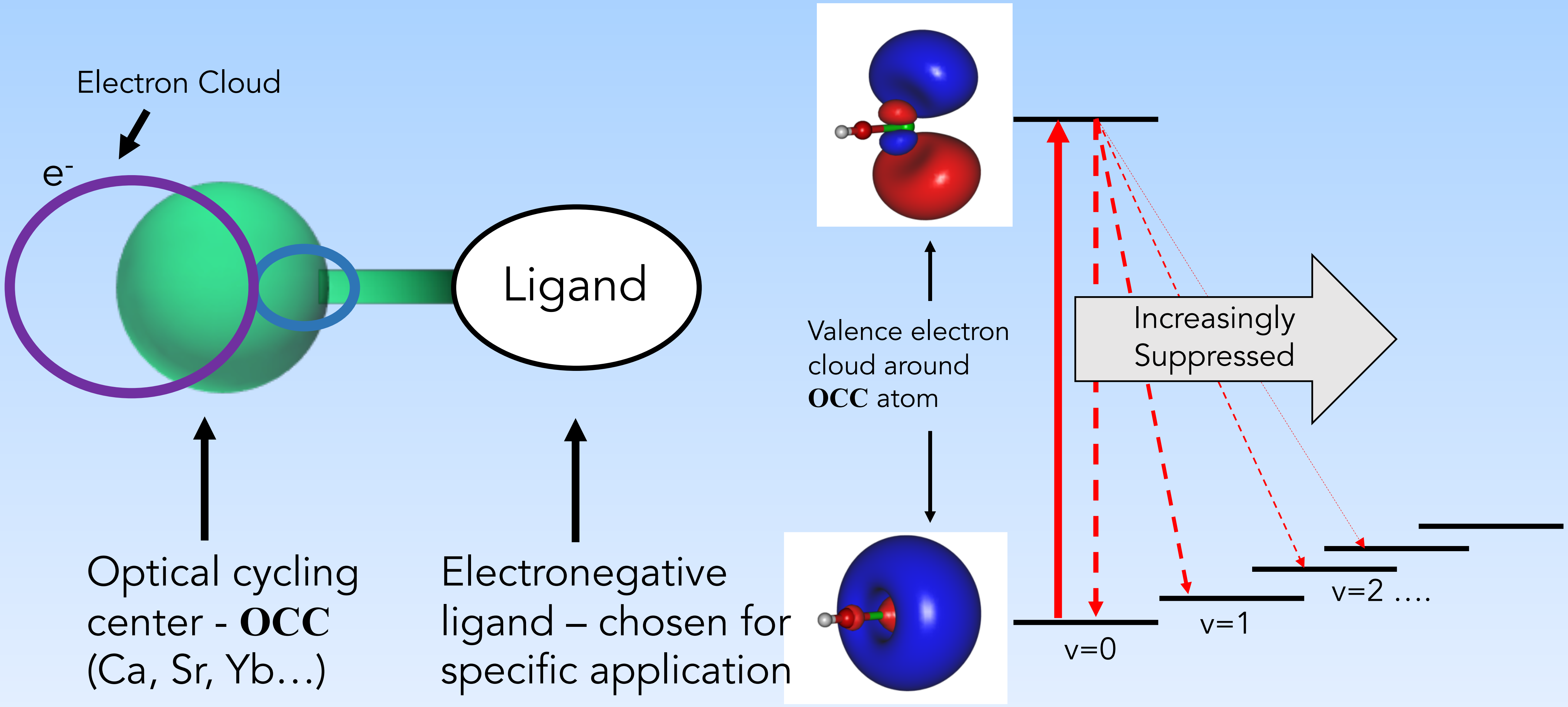



Optical cycling center - **OCC** (Ca, Sr, Yb…)

Electronegative ligand – chosen for specific application

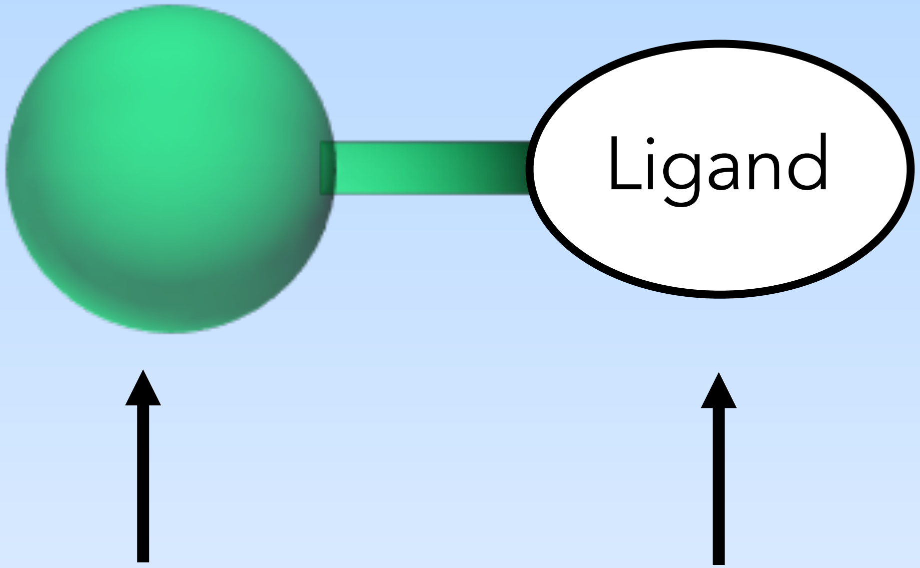
Isaev and Berger, PRL **116** (2016); Kozyryev et al., ChemPhysChem **17** (2016); Augenbraun et al., PRX (in press), arXiv:2001.11020

# *Molecule Laser Cooling is Generalizable*



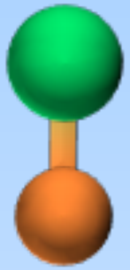

Isaev and Berger, PRL **116** (2016); Kozyryev et al., ChemPhysChem **17** (2016); Augenbraun et al., PRX (in press), arXiv:2001.11020

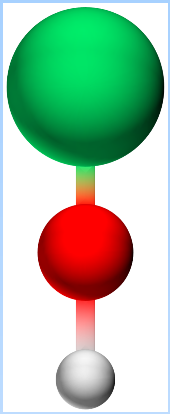
# *Molecule Laser Cooling is Generalizable*

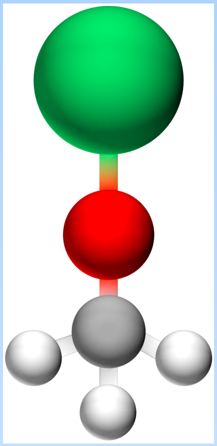



Optical cycling center - **OCC** (Ca, Sr, Yb…)

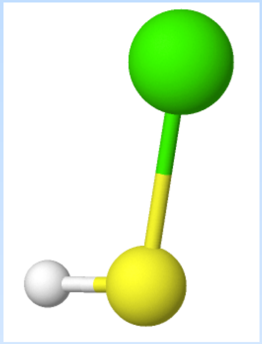
Electronegative ligand – chosen for specific application

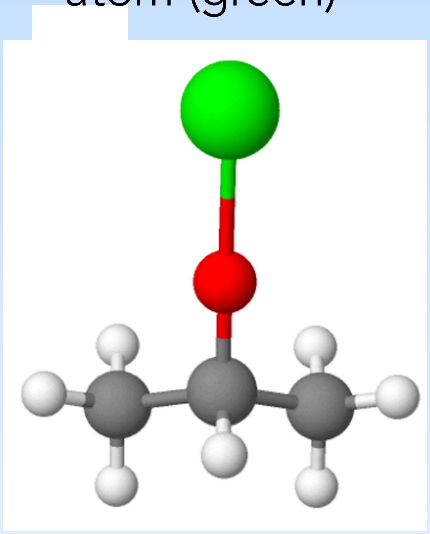


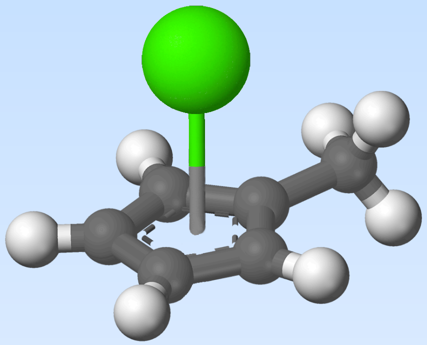




Different **ligands** attached to an **OCC** atom (green)

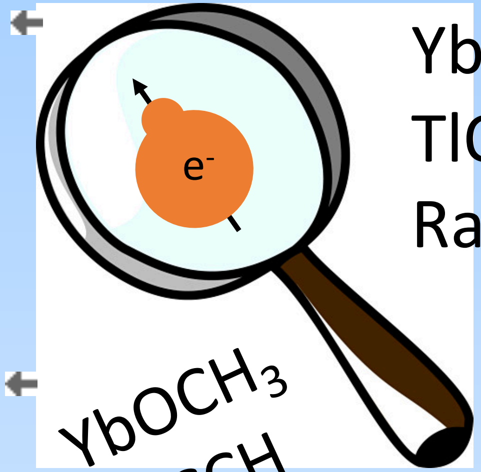






Isaev and Berger, PRL **116** (2016); Kozyryev et al., ChemPhysChem **17** (2016); Augenbraun et al., PRX (in press), arXiv:2001.11020

# Laser Cooling Generalizable to Lots of Polyatomics



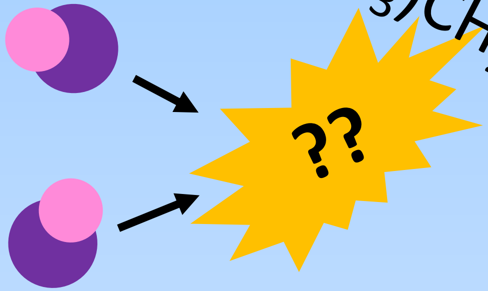

YbOH
TlOH
RaOH

$^{9}$BeNC
$^{25}$MgNC
$^{43}$CaOH

CaNCO
SrNCO
$SrNH_2$
$CaNH_2$

BaCCH
$MgOCH_3$
CaOH
SrOH
BaNCO

$CaOCH(CH_3)CH_2CH_3$




$YbOCH_3$
YbCCH

$CaOC_2H_5$
SrOCHDT
BaNC
BaOBO

PbOH
HfCH
LuCO
$BeOCH_3$

$CaCCCH_3$
SrSH
CaSH
CaNC

SrOBO
CaOBO

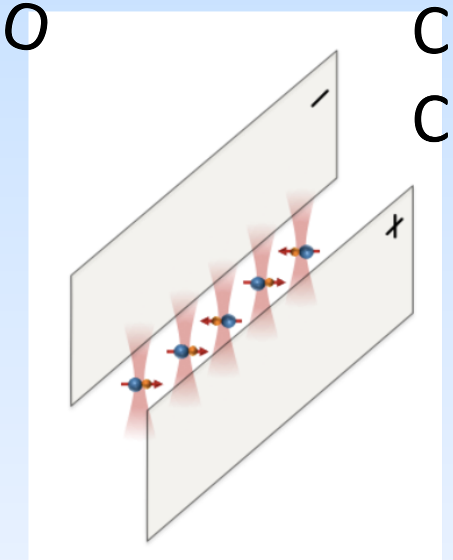
$SrO(CH_2)_3CH_3$
$SrOC(CH_3)_3$
$SrOC(CH_3)=CH_2$

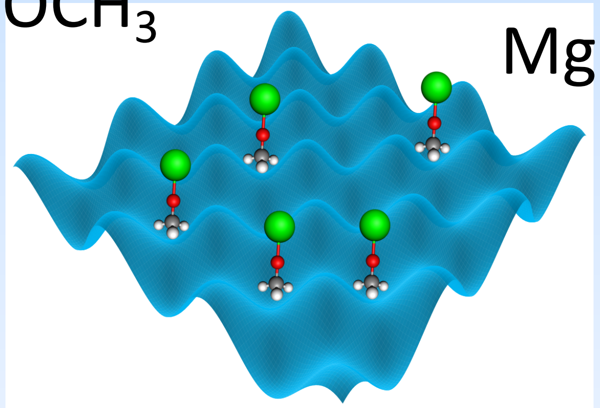
$^{87}$SrOH
$^{137}$BaOH
$^{171}$YbOH

$LuOH^+$
TaCOCa
ThCH

CaOH
$CaOCH_3$

MgCCH
CaNC
MgNC

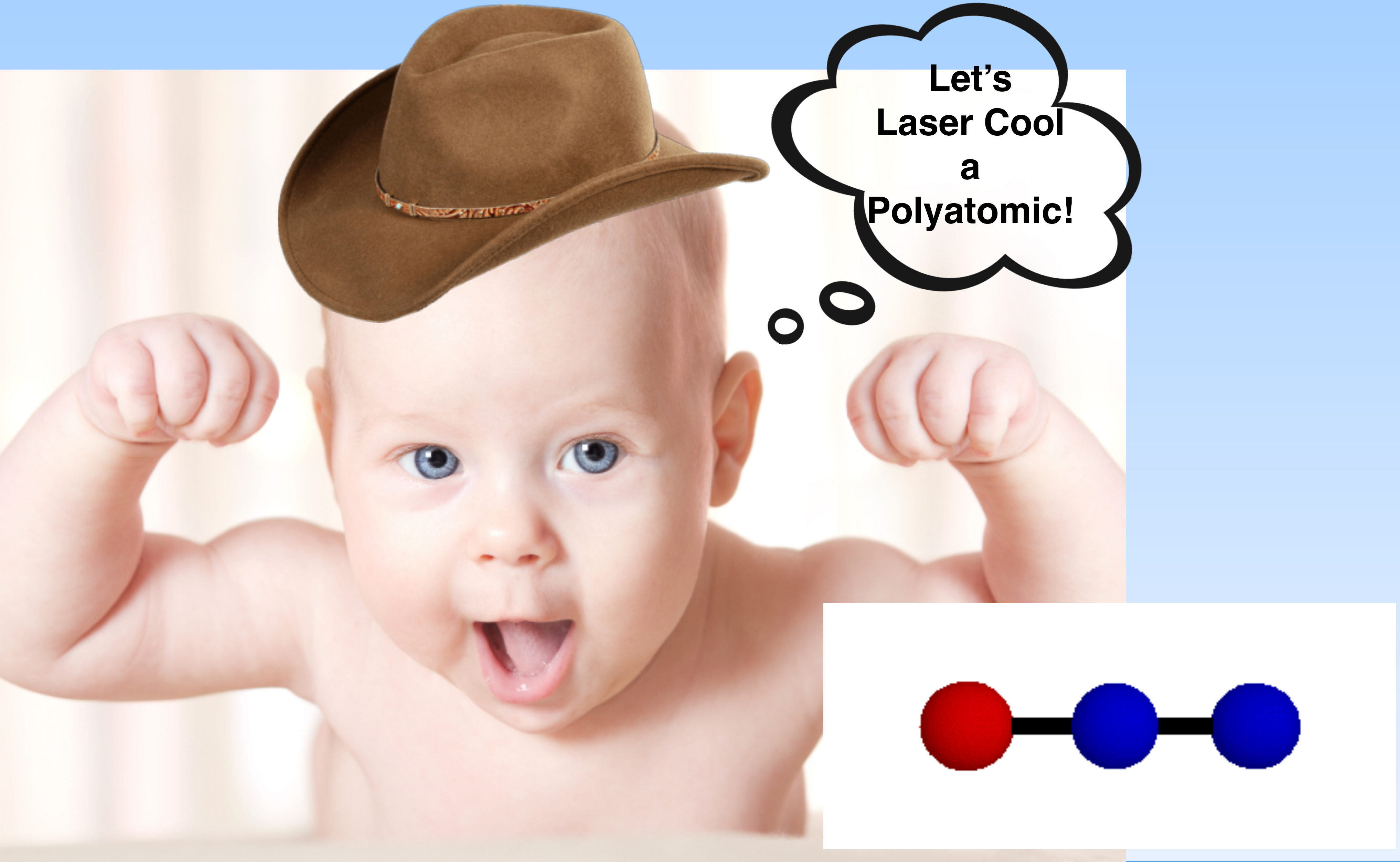




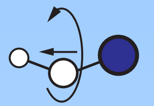
Norrgard, ..., Scherschlight, Comm. Phys. 2 (2019)
Kozyryev and Hutzler, PRL 119 (2017)
Kozyryev, ..., Doyle, ChemPhysChem 17 (2016)
Ivanov, ..., Krylov, PhysChemChemPhys (2019)

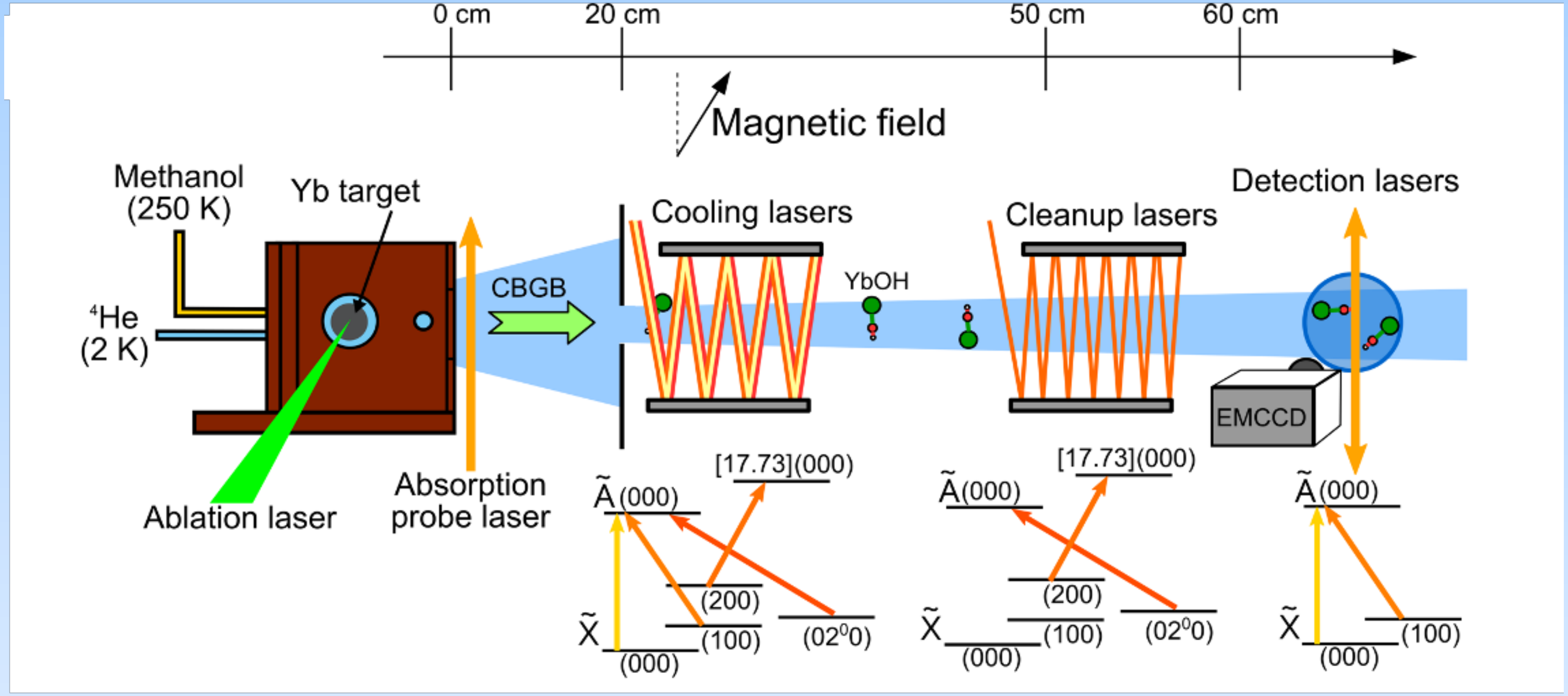
# Sounds Hard to the Seasoned AMO Experimentalist….

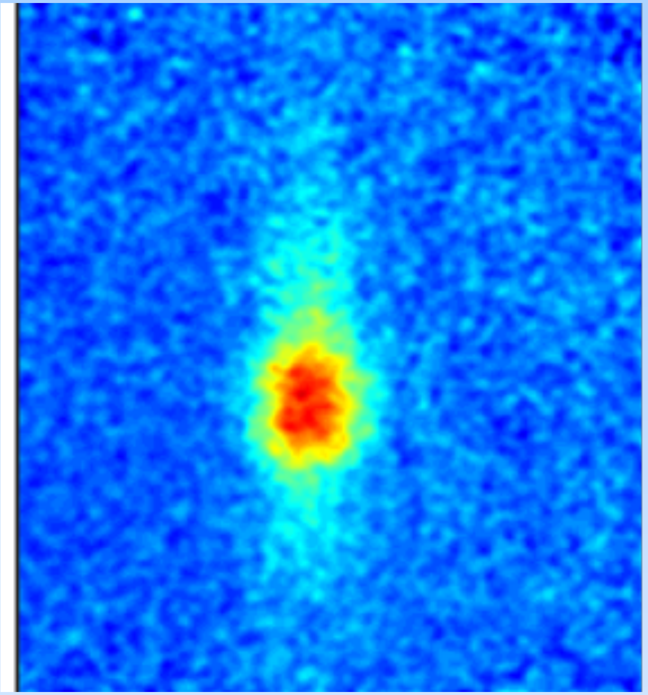
# So what!



# Sisyphus Laser Cooling of YbOH for *PolyEDM* Search

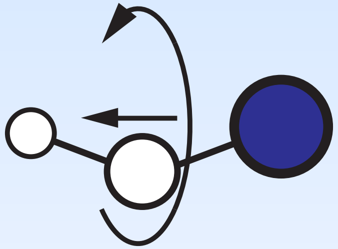


**Blue-Detuned**
**Cooing 20 mK to <600 μK**
~500 photons scattered

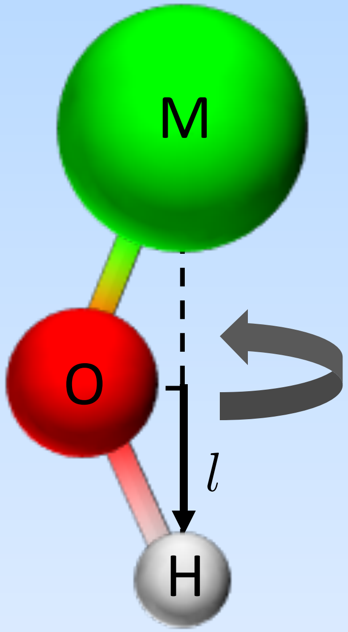
**Laser cooling of YbOH works efficiently despite vibrational and electronic complex structure**



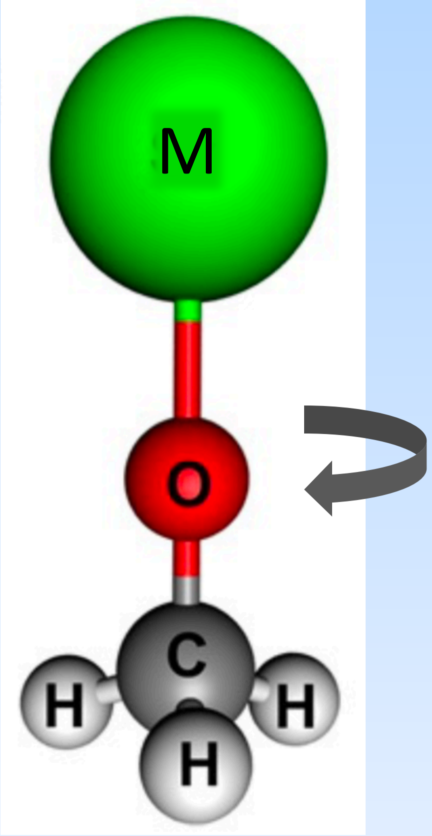
# Why Stop There?

Bending Mode Angular Momentum and Symmetric Top Angular Momentum

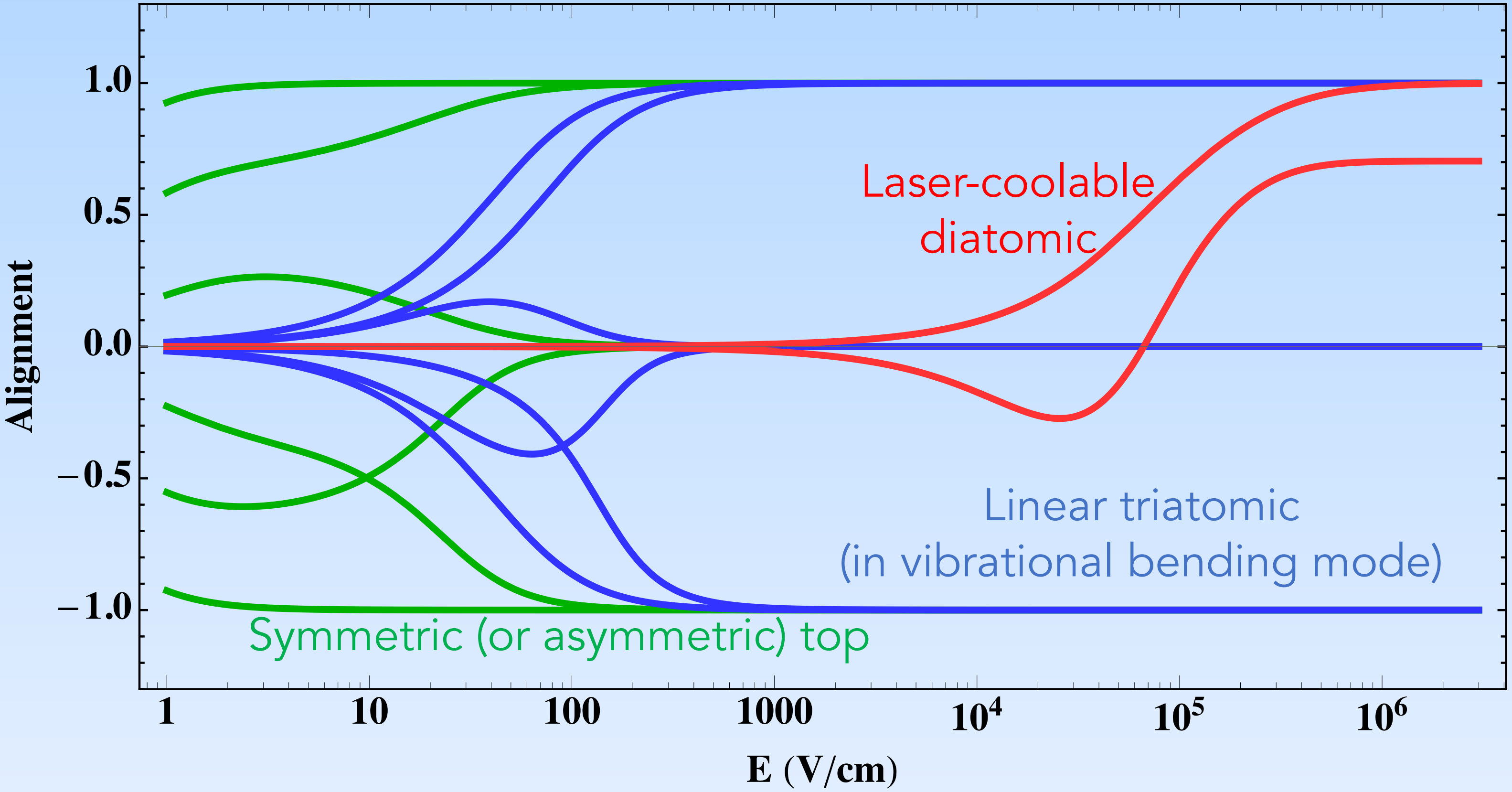
Parity doublets




$\Delta E \sim 10$ MHz




$\Delta E \sim 100$ kHz

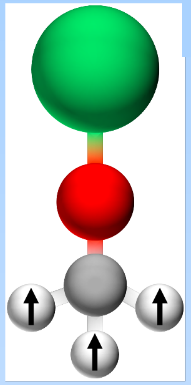


# Laser Cooling of a Symmetric Top - $CaOCH_3$

## A *teeny bit* more complicated….

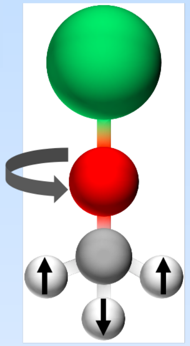
Two new challenges:

1. 12 vibrational modes in play
   - Favorable **FCFs** measured[1]
   - "Old hat" by now… just add vibrational repumps

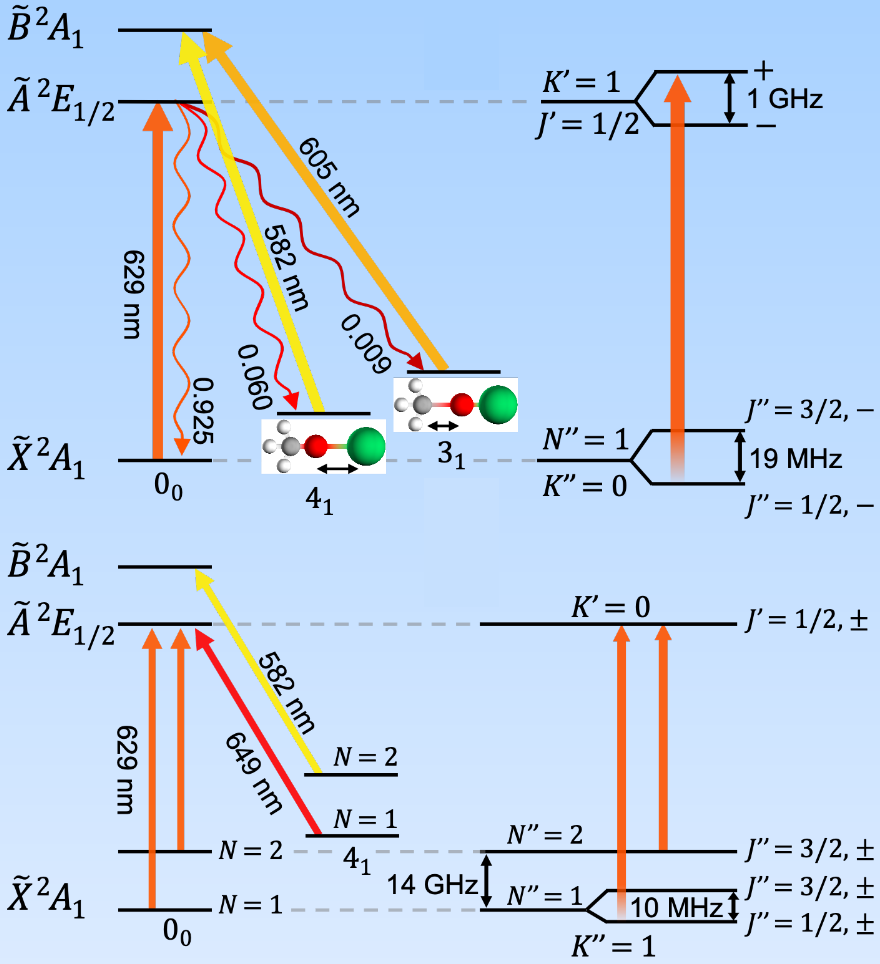


$K = 0$

2. Novel rotational structure due to rigid body rotation $K$
   - Rotations about "a" principal axis allowed
   - **No help from parity!**



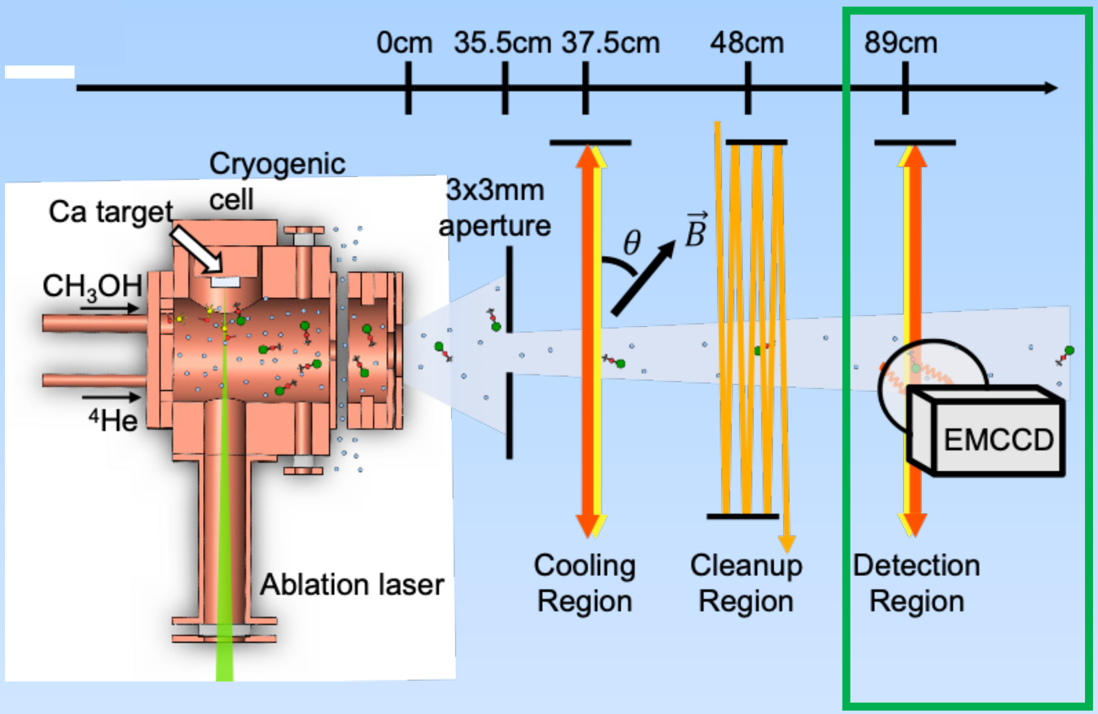
$K = 1$

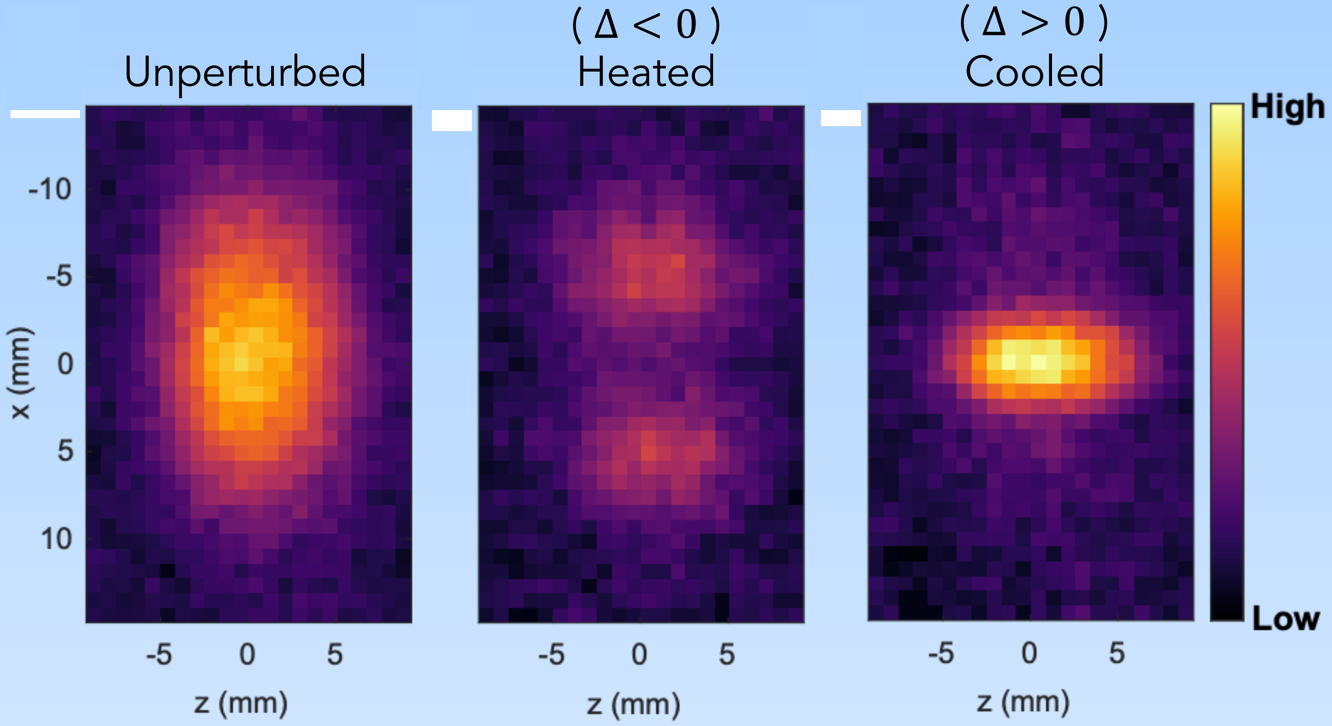



1 [Determination of CaOH and CaOCH3 vibrational branching ratios for direct laser cooling and trapping] I. Kozyryev, T. C. Steimle, P. Yu, D.-T. Nguyen, J.M. Doyle. New J. Phys. 21, 052002 (2019).

[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 J.M. Doyle. Science 369, 1366 (2020).

# Laser Cooling of a Symmetric Top - $CaOCH_3$



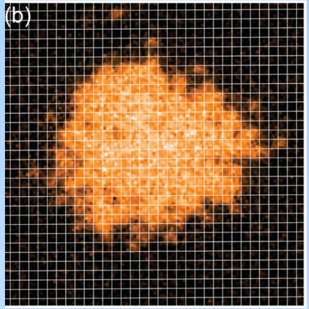

Ca + $CH_3OH$ → CaOH + …
Ca + $CH_3OH$ → $CaOCH_3$ + …

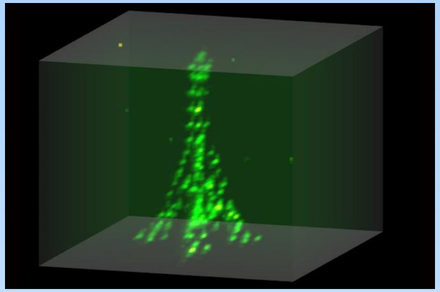
- Cooled ~$10^4$ molecules from 22(1) mK to 1.8(7) mK
- ~100 photons scattered
- Scattering rates ~$2\times10^6$ $s^{-1}$
- Works equally well for both nuclear spin isomers
- **All comparable to early demonstrations with diatomics!**

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 J. M. Doyle. Science 369, 1366 (2020).

# Complexity

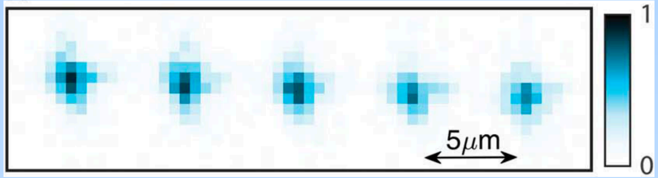
## Atoms
## (N=1)



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

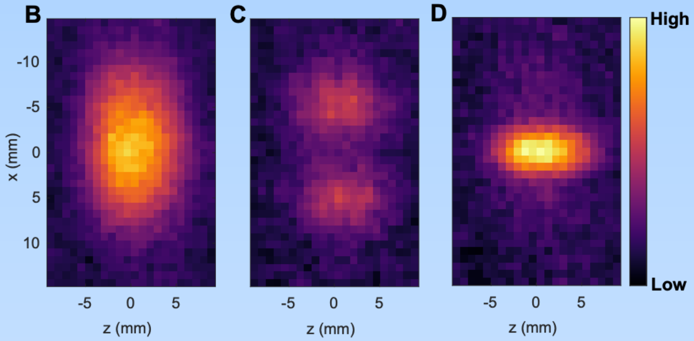
## Diatomic Molecules
## (N=2)




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

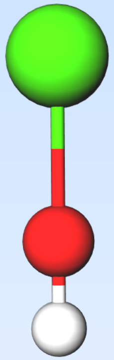
Harvard, Yale, Imperial, Boulder, Columbia, UConn, UC Riverside, ...

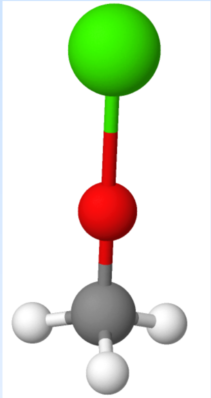
## Polyatomic Molecules
## (N=3,4,5...)

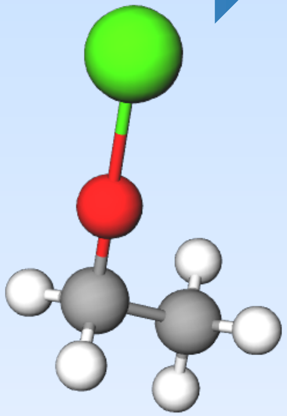



Complexity [reduced molecular symmetry]

Eliminate
spherical symmetry
**[1 moment of inertia]**





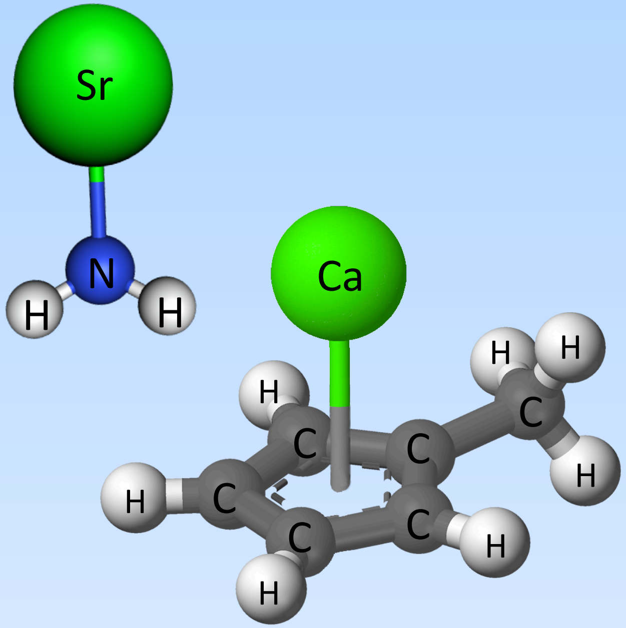


Molecular Asymmetry and Optical Cycling: Laser Cooling Asymmetric Top Molecules B. L. Augenbraun, J. M. Doyle, T. Zelevinsky, I. Kozyryev, Phys. Rev. X 10, 031022 (2020).

# Asymmetric Top Molecules - Possible Applications

## Cold chemistry

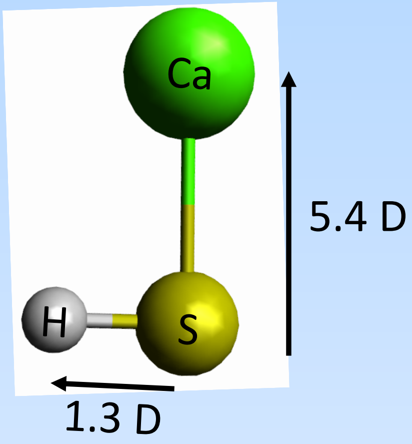
Astrophysical relevance
Chemically interesting ligands




Bond-selective dissociation,
Can tailor ligand to science goal

## Quantum simulation

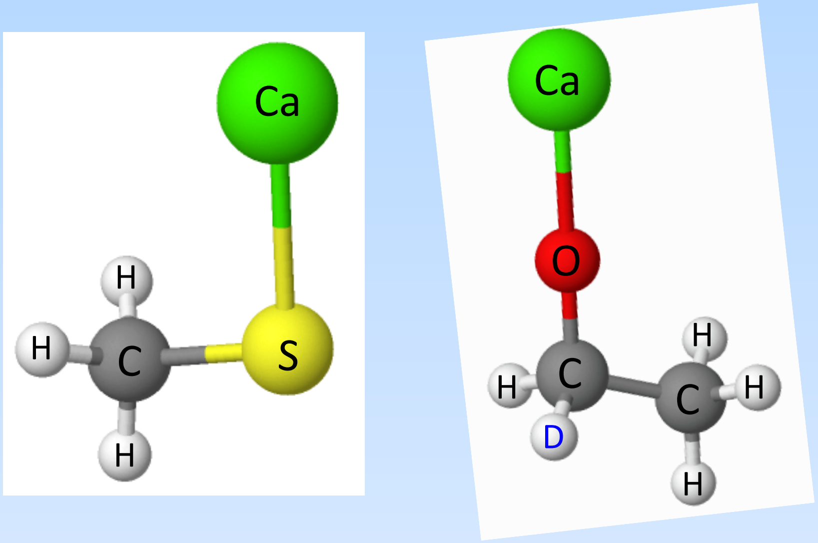
Large dipole moments
Multiple (permanent) projections




Molecule-molecule couplings
10-100x stronger than
comparable diatomics

## Fundamental Symmetries

$m_p/m_e$ variation, Parity violation,
CP-violating electromagnetic moments, ...




Internal rotors and chiral structures ***only***
exist in asymmetric top molecules


van Dishoeck, et al. *Astro. J. Lett.*, 416 (1993)
Robles, Ellis, and Miller, J. Am. Chem. Soc. 114, 7171 (1992)

Scurlock, ..., Steimle. *J. Chem. Phys.*, 100, 5481 (1994)

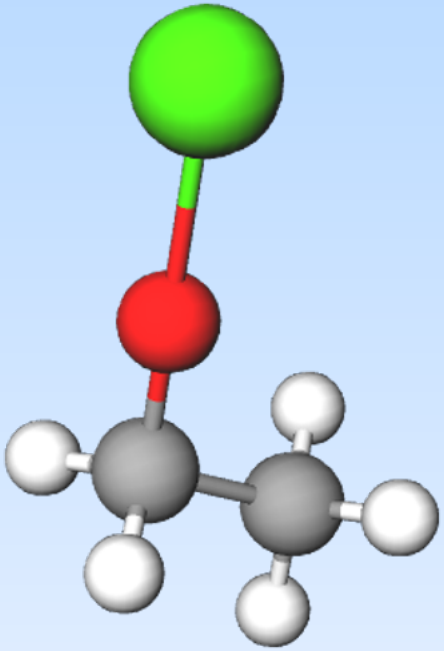
Jansen, Bethlem, Ubachs, J. Chem. Phys. 140, 010901 (2014)
Fernando, ..., Bernath, *J. Phys. Chem.* 95, 7 (1991)
Paul, Reza, Liu. *J. Mol. Spec.*, 330 (2016)

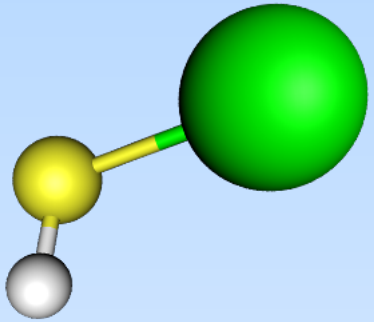
## *Questions of the moment*

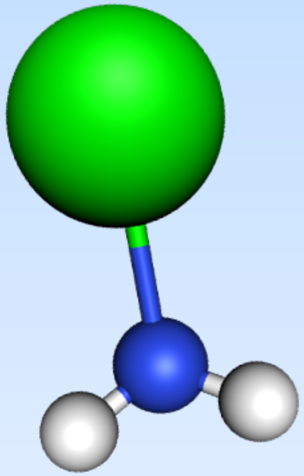
*Can one laser cool asymmetric top molecules?*

*Can one make them microKelvin temperatures?*

*Can we achieve 10 s coherence times?*

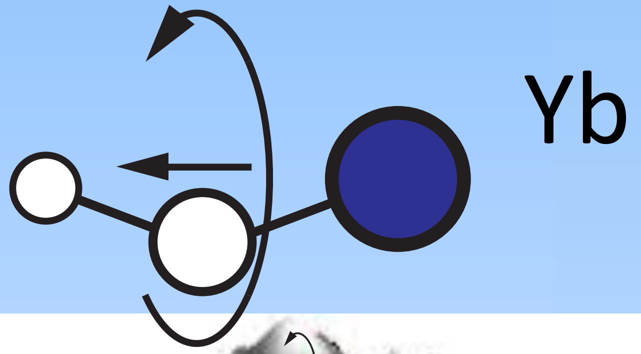


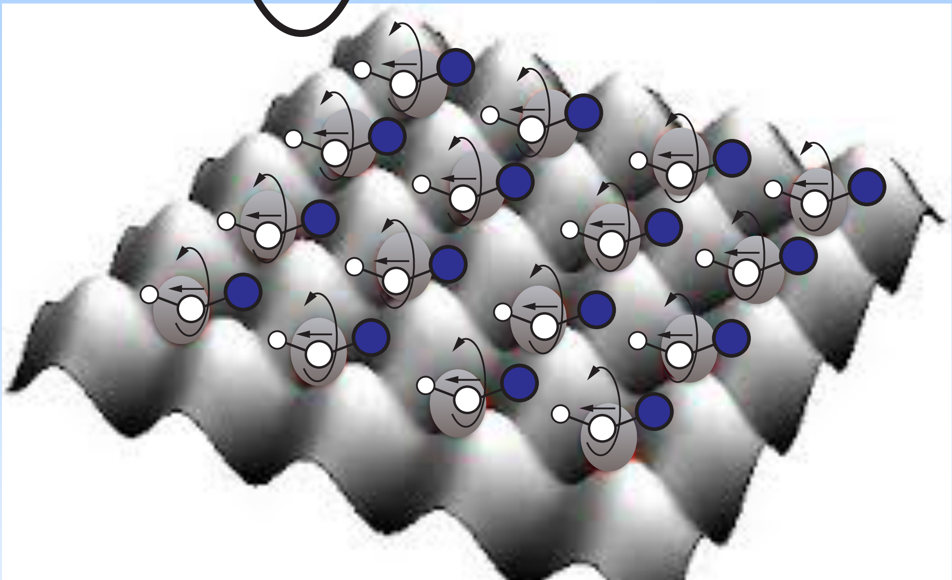




# YbOH - Future EDM - Polyatomics in Optical Trap

Now We Have a Path to Realize a new EDM experiment…



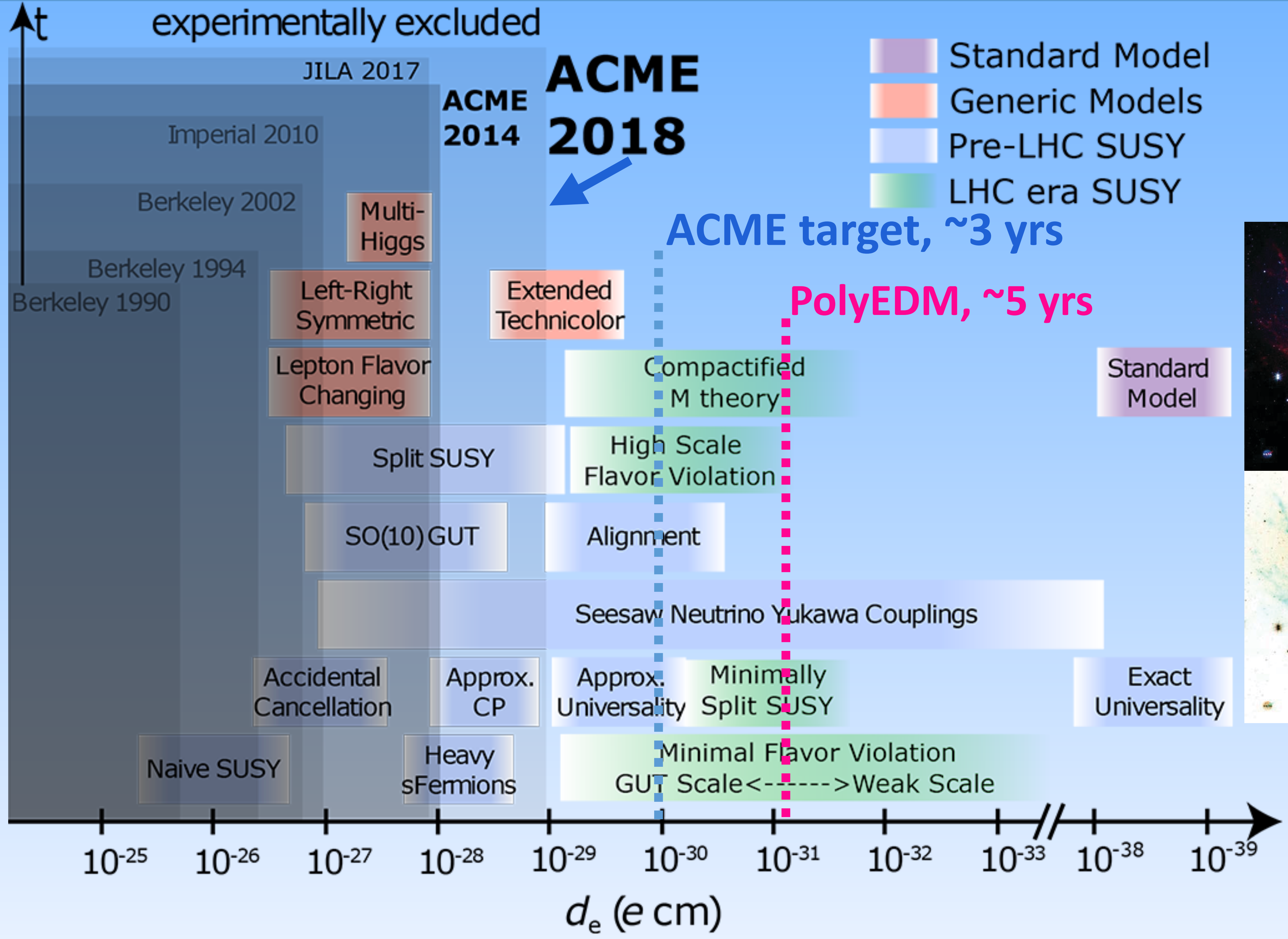
*Kozyryev and Hutzler, PRL (2017)*

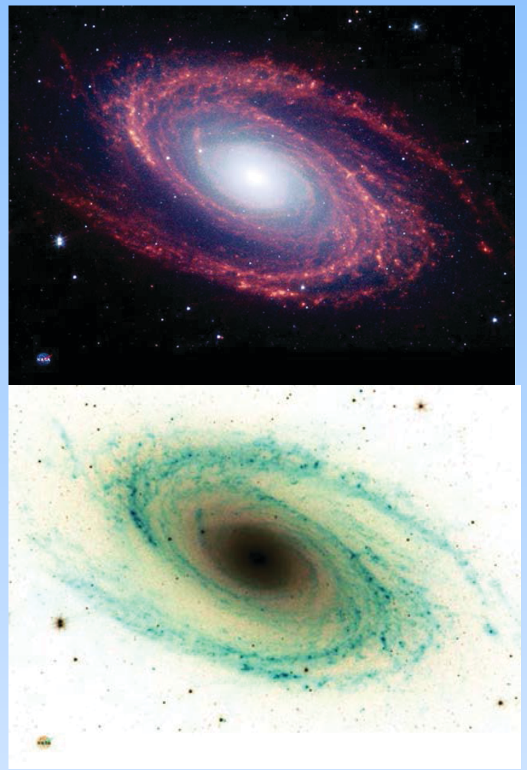
**Long Coherence Time, Large Numbers**
**Small Volume**

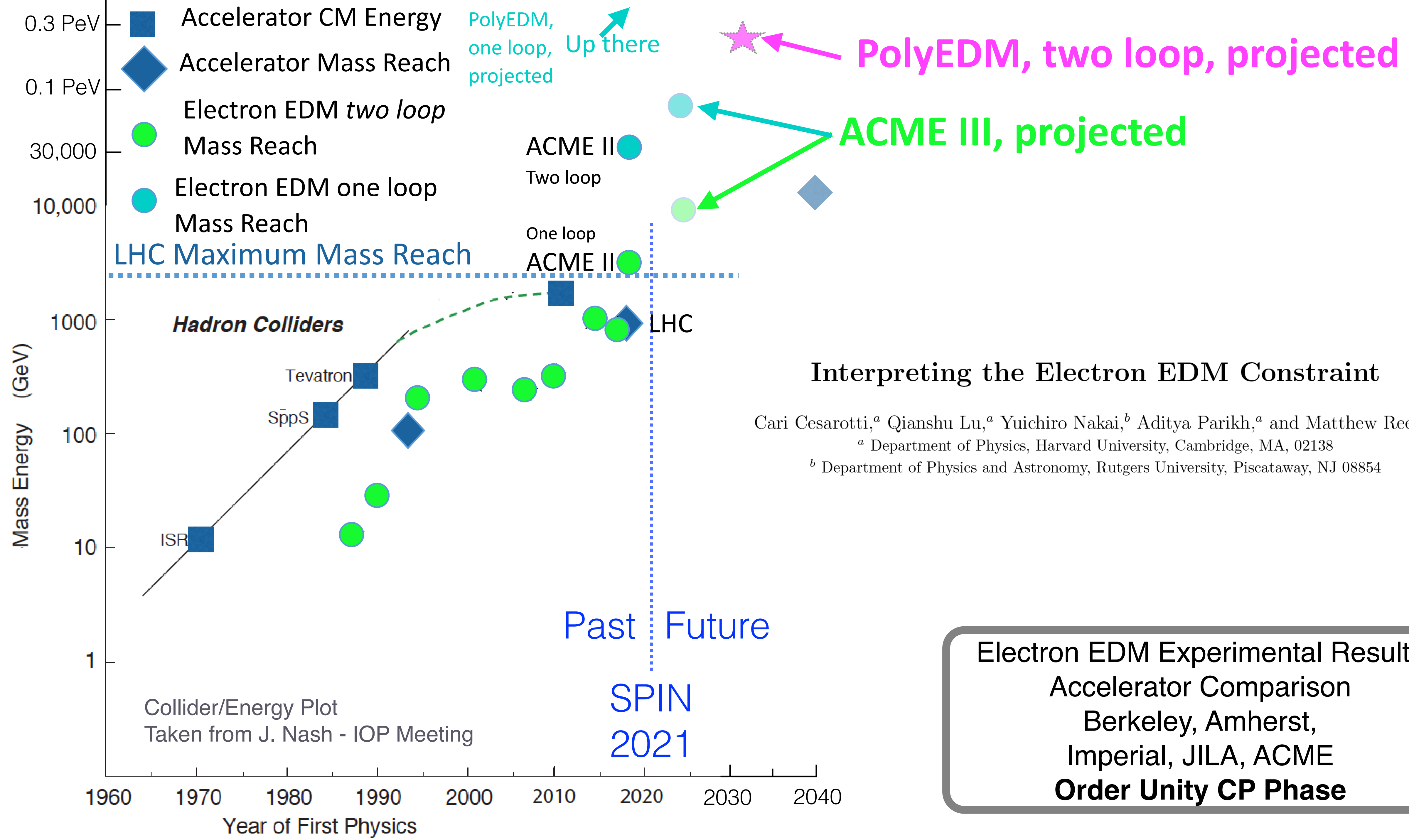
YbOH can be laser cooled

1) Heavy Molecule
2) **$\ell$ doubling**
3) Large Numbers
4) Ultracold - ODT - Long $\tau$

Hopkins
2021






## Interpreting the Electron EDM Constraint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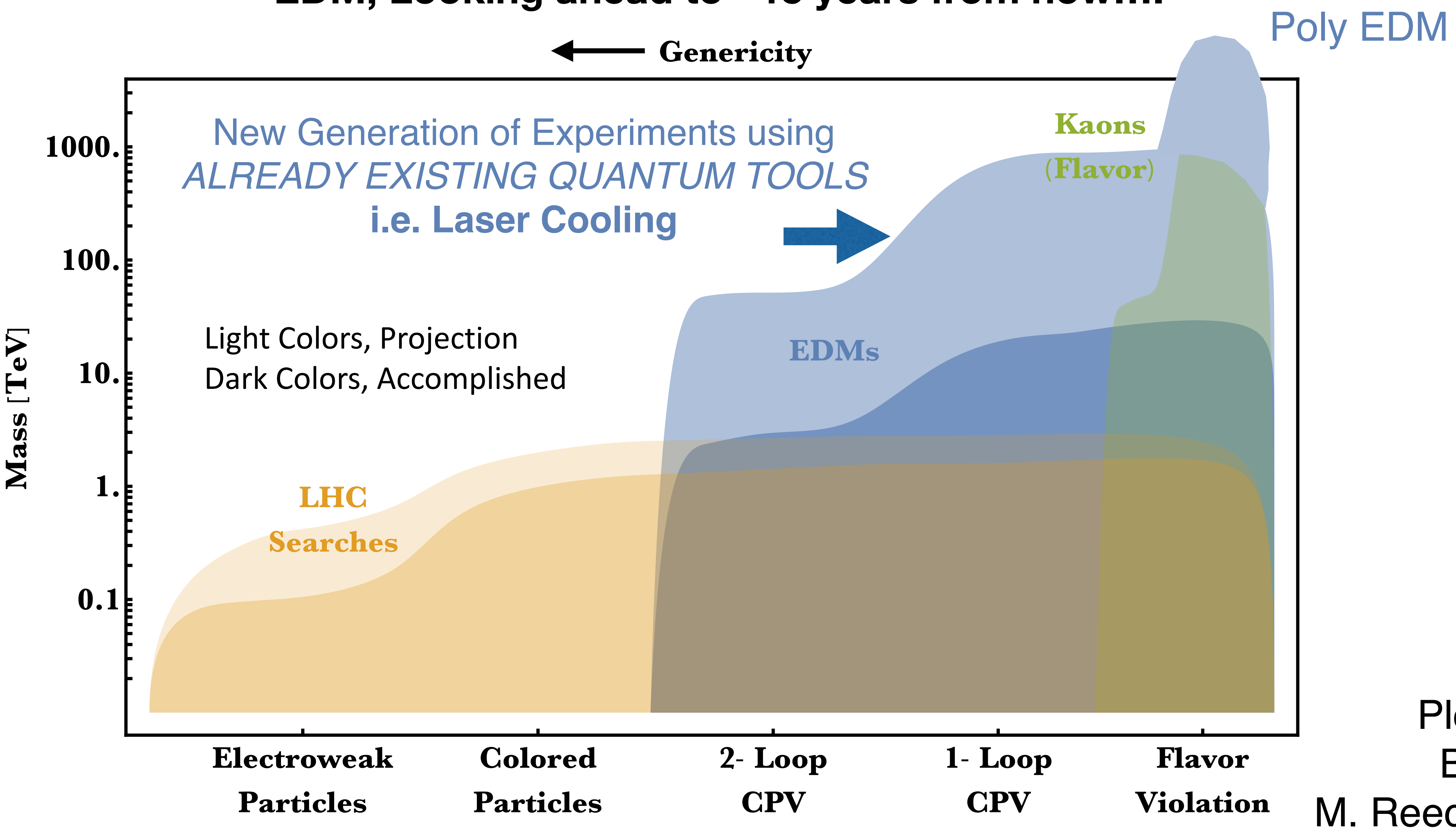
Electron EDM Experimental Results
Accelerator Comparison
Berkeley, Amherst,
Imperial, JILA, ACME
**Order Unity CP Phase**

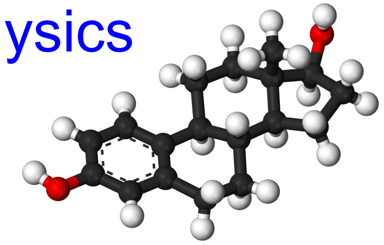
# EDM, Looking ahead to ~15 years from now….



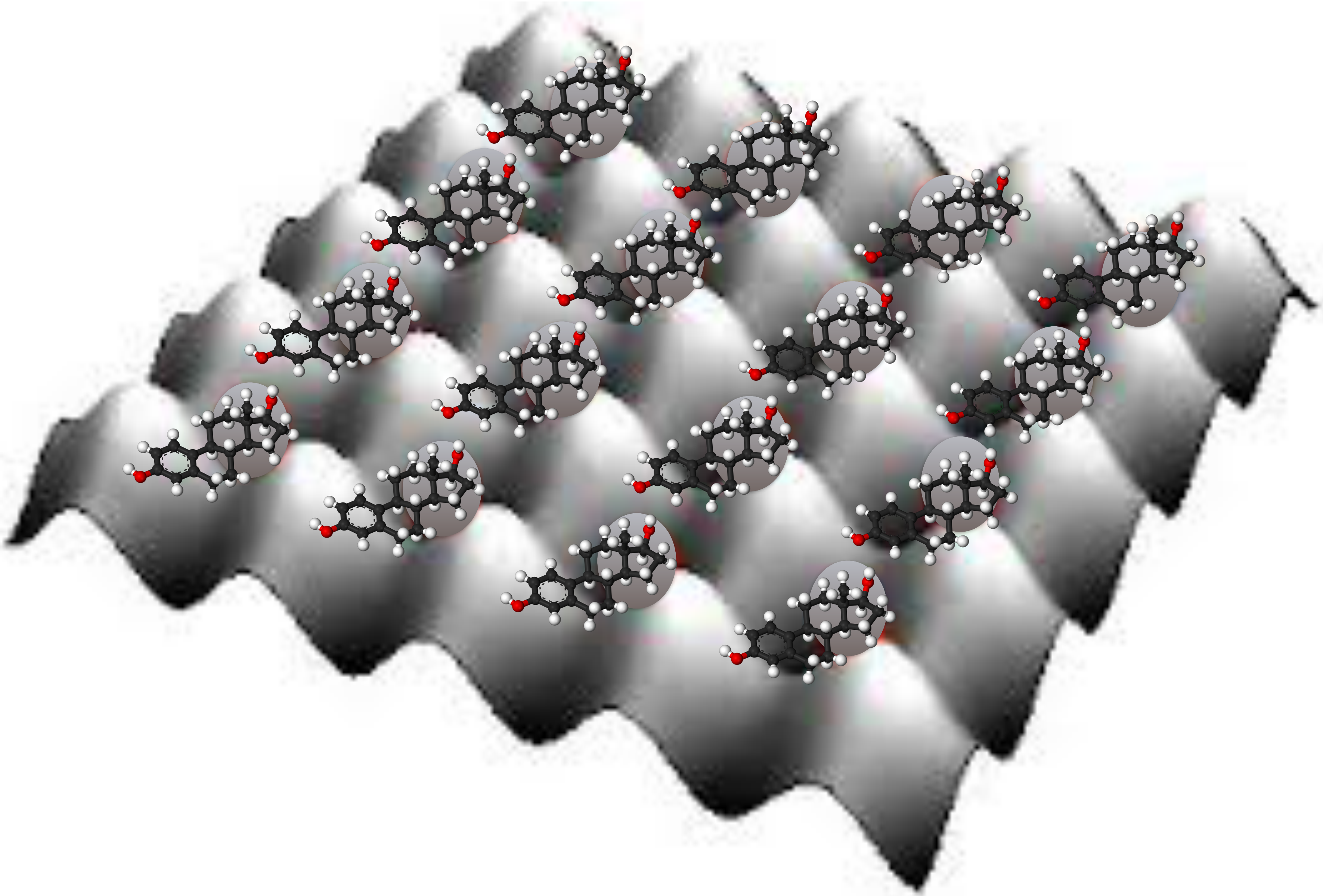

Plot By M. Reece

# The Future is Calling

Designer
Molecule
For
Particle
Physics



No longer “if”.



# Doyle Group Members Photo Summer 2019