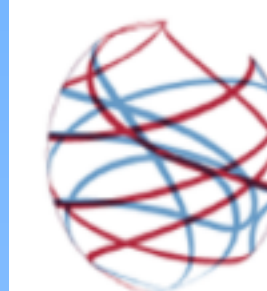




*Center for Ultracold Atoms
At Harvard and MIT*



*Harvard
University*

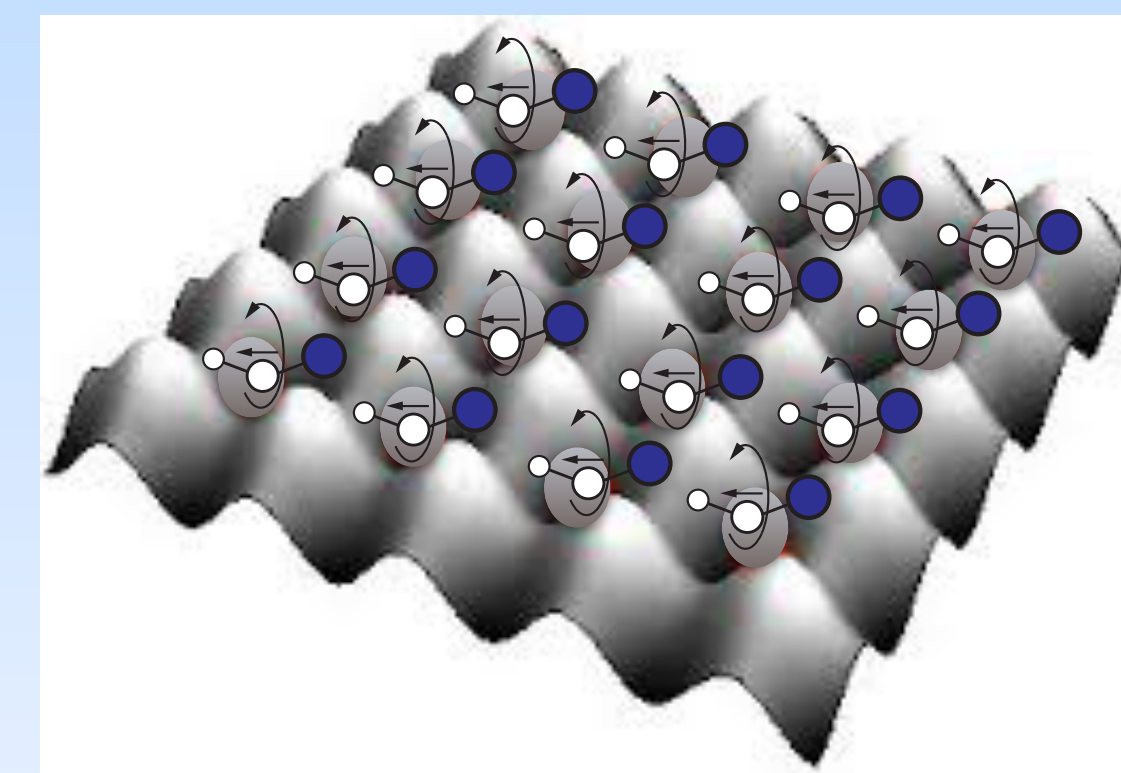
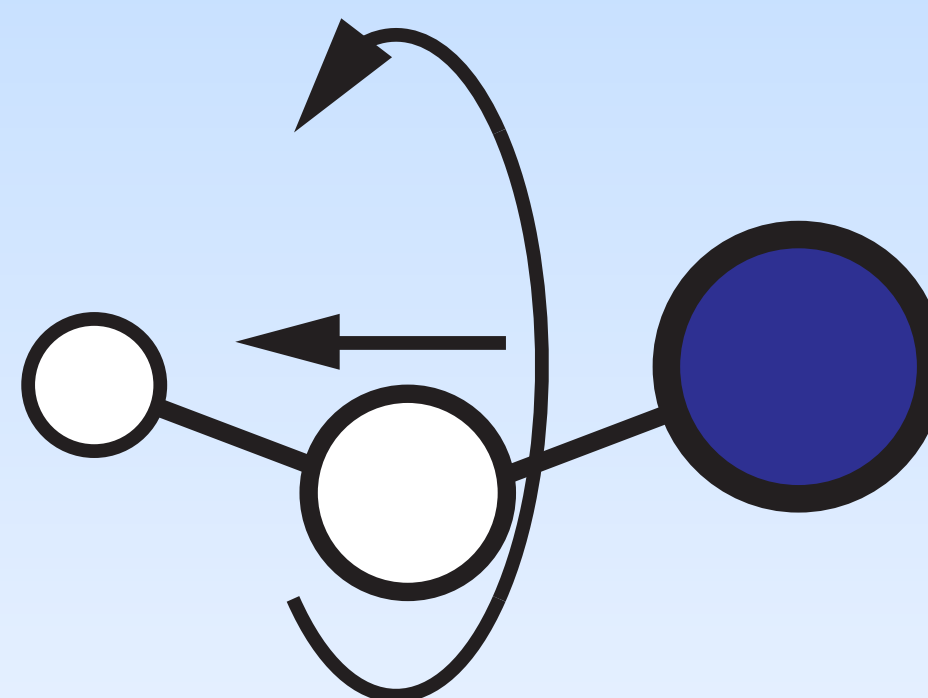
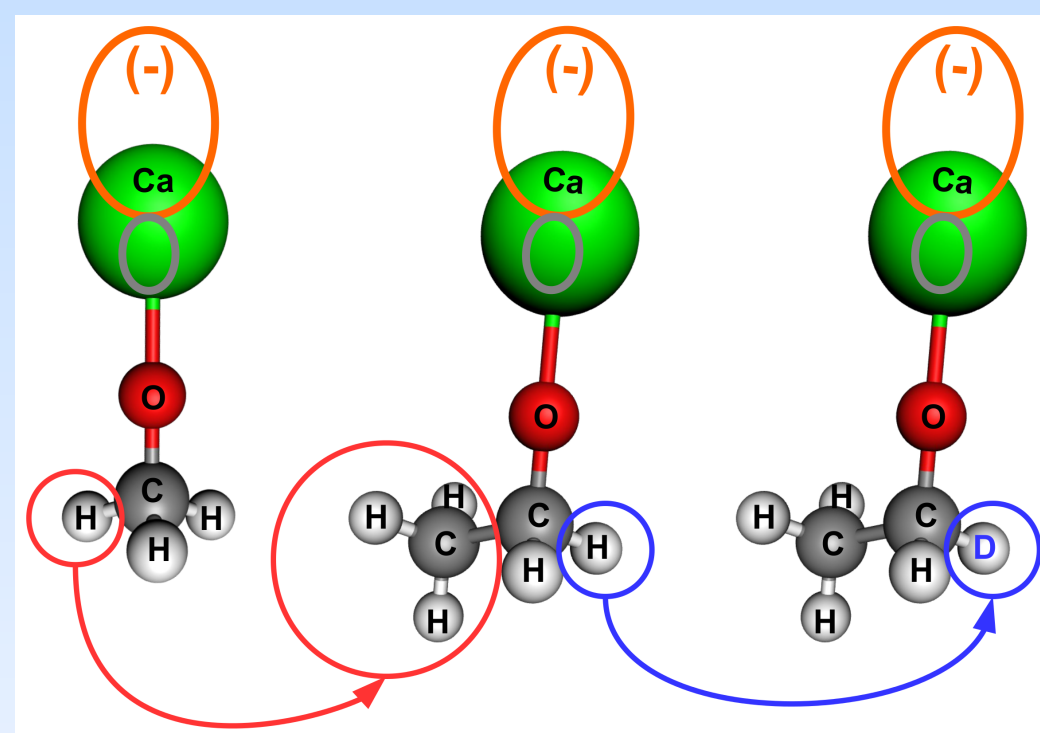


Harvard **Quantum** Initiative
IN SCIENCE AND ENGINEERING

*Harvard
Quantum Initiative*

Ultracold Molecules (and Spectroscopy) for Quantum Science

100 Years of Quantum Physics
ICISE, Quy Nhon, 8 October 2025



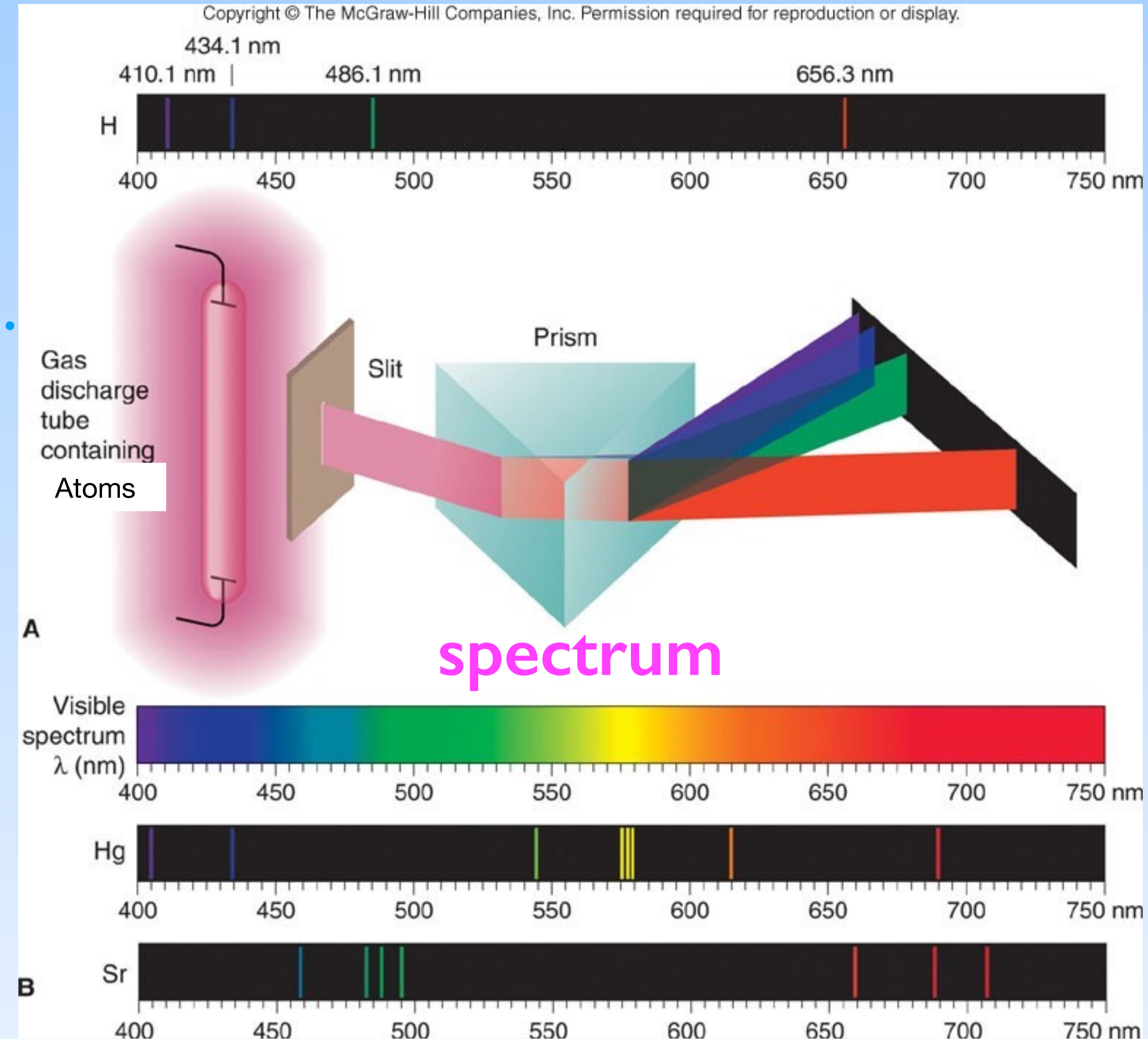
The Beginning of AMO* was Spectroscopy

AMO started before
the advent of quantum theory...

Spectroscopy

*AMO
is

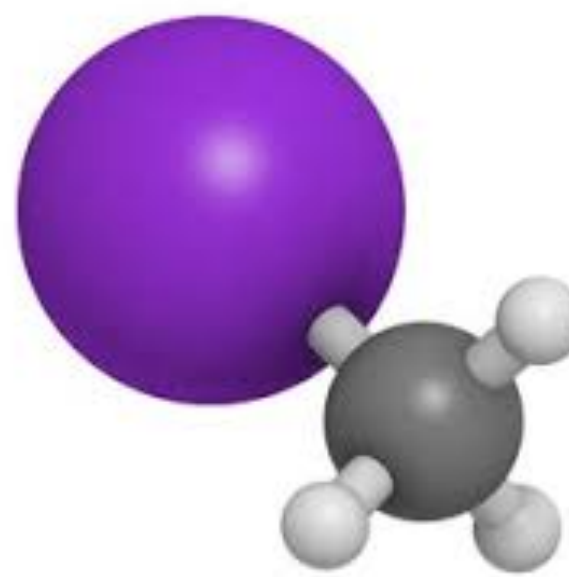
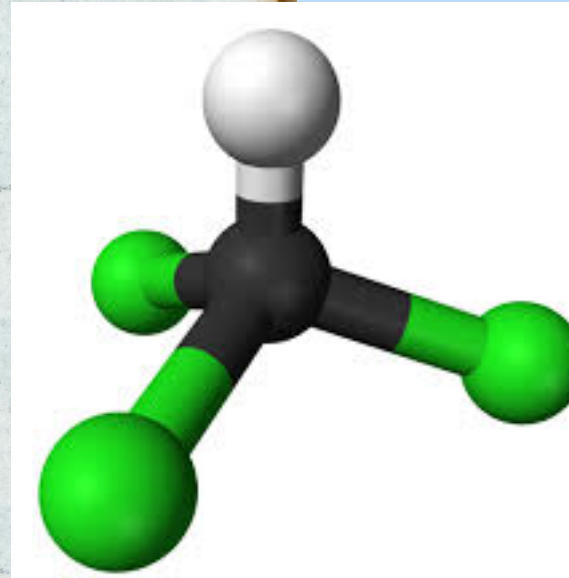
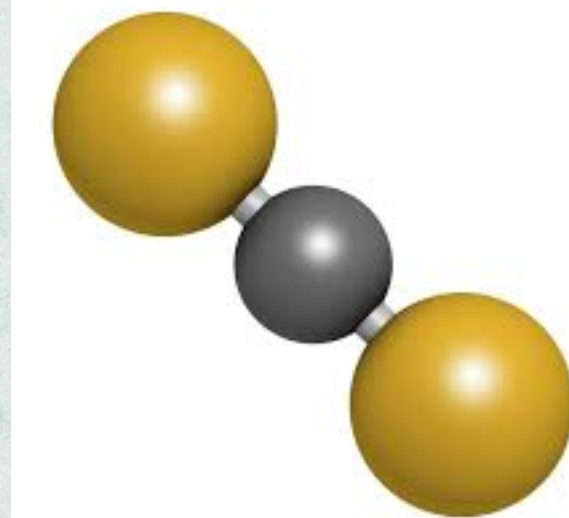
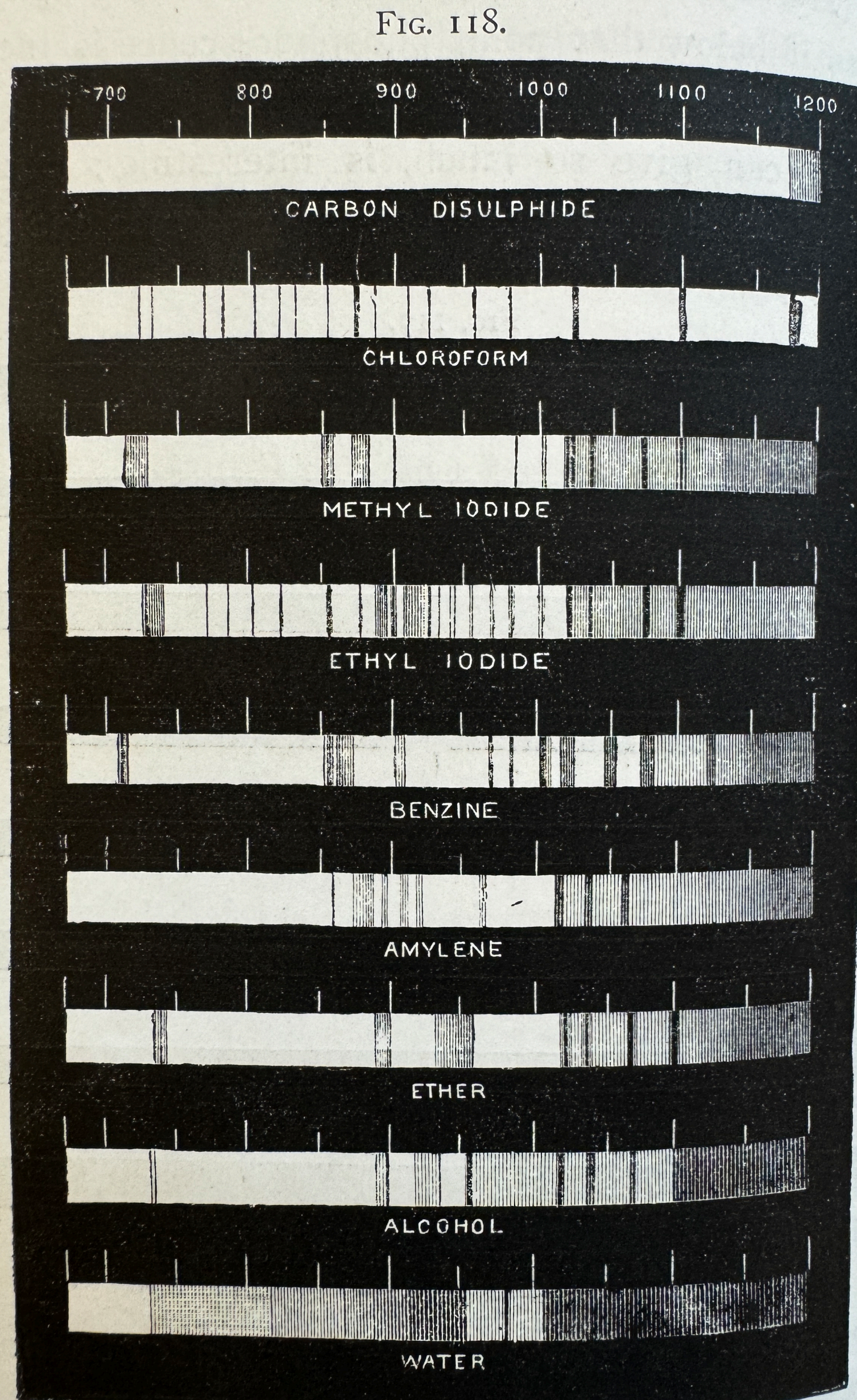
Atomic, Molecular and Optical Physics



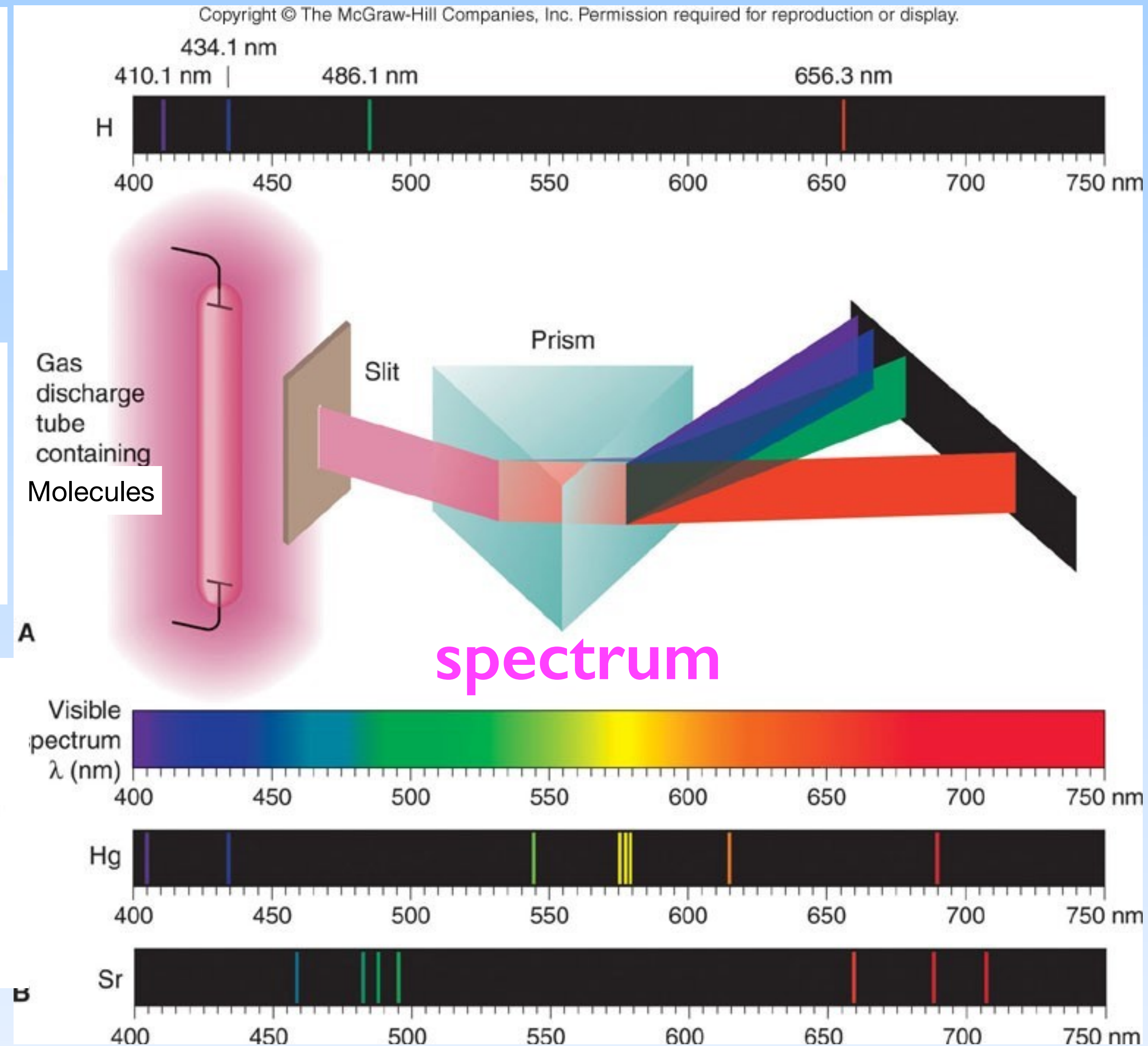
The Beginning of AMO* was Spectroscopy



The Beginning of AMO* was Spectroscopy



shutterstock.com · 131939129



SPECTRUM ANALYSIS

IN ITS APPLICATION TO
TERRESTRIAL SUBSTANCES,

AND
THE PHYSICAL CONSTITUTION OF THE
HEAVENLY BODIES.

FAMILIARLY EXPLAINED BY THE LATE
DR. H. SCHELLEN,

DIRECTOR DER REALSCHULE I. O. COLOGNE, RITTER DES ROTHEN ADLERORDENS IV. KL.,
ASSOCIATE OF SEVERAL LEARNED SOCIETIES.

TRANSLATED FROM THE THIRD ENLARGED AND REVISED GERMAN EDITION BY
JANE AND CAROLINE LASSELL.

EDITED, WITH NOTES, BY
CAPTAIN W. de W. ABNEY, R.E., F.R.S.

With Numerous Woodcuts, Coloured Plates,

AND
ÅNGSTRÖM'S AND CORNU'S MAPS.

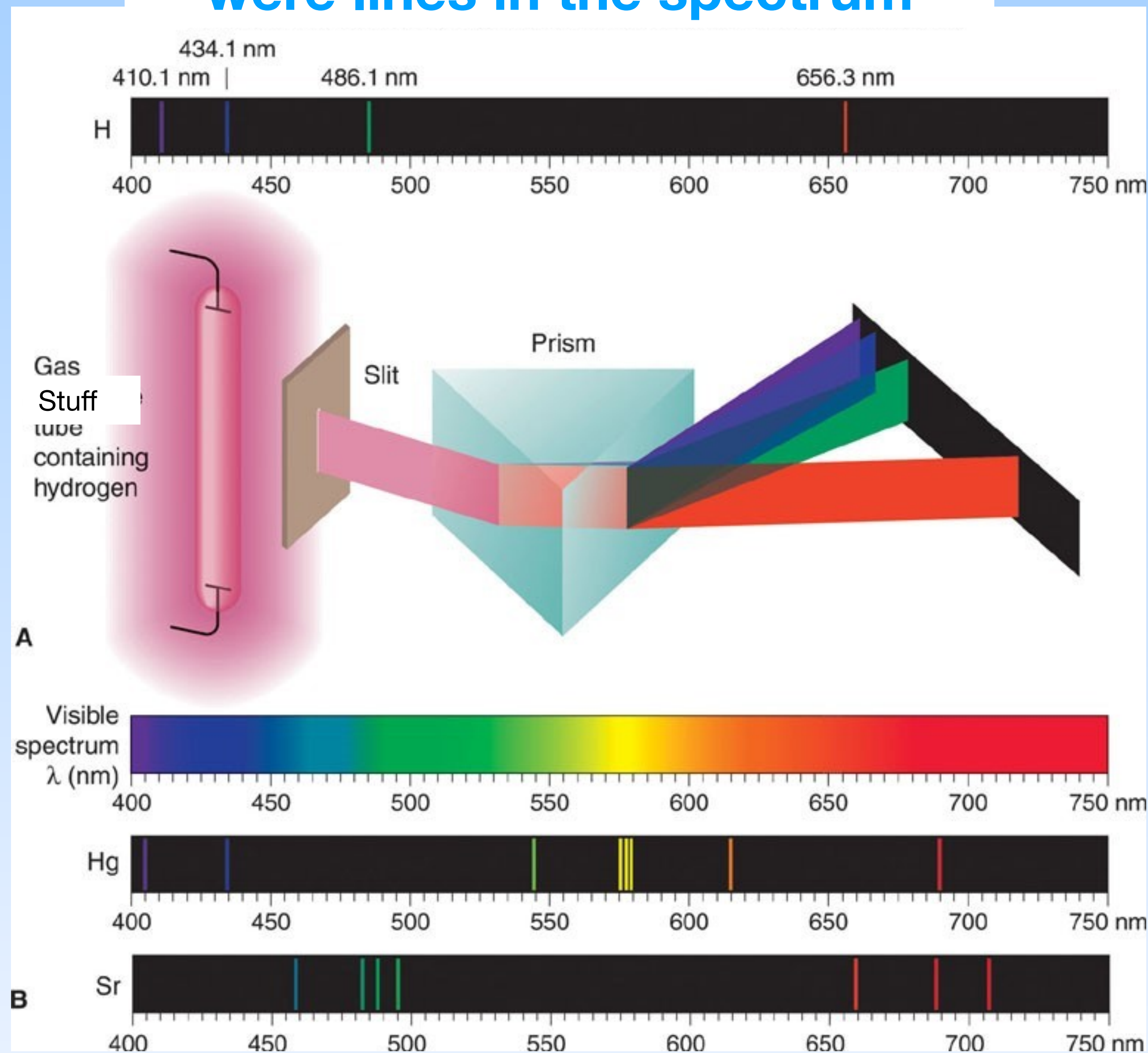
SECOND EDITION.

LONDON:
LONGMANS, GREEN, & CO.

1885.

1885

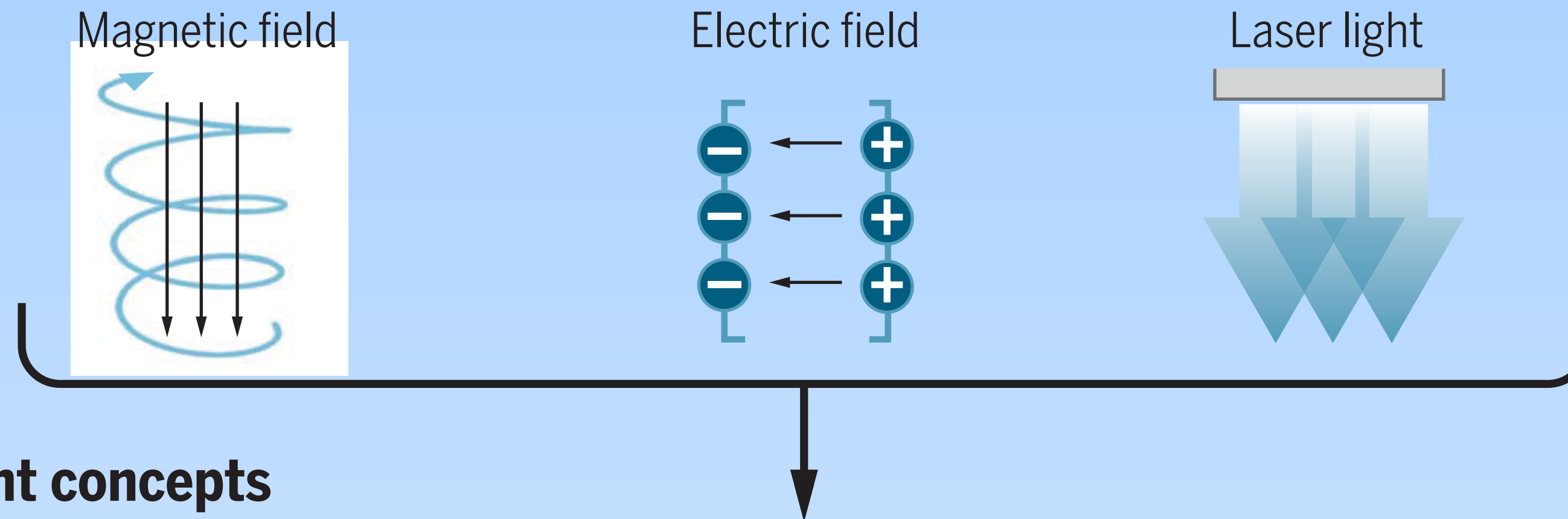
Scientists developed key ideas
about why there
were lines in the spectrum



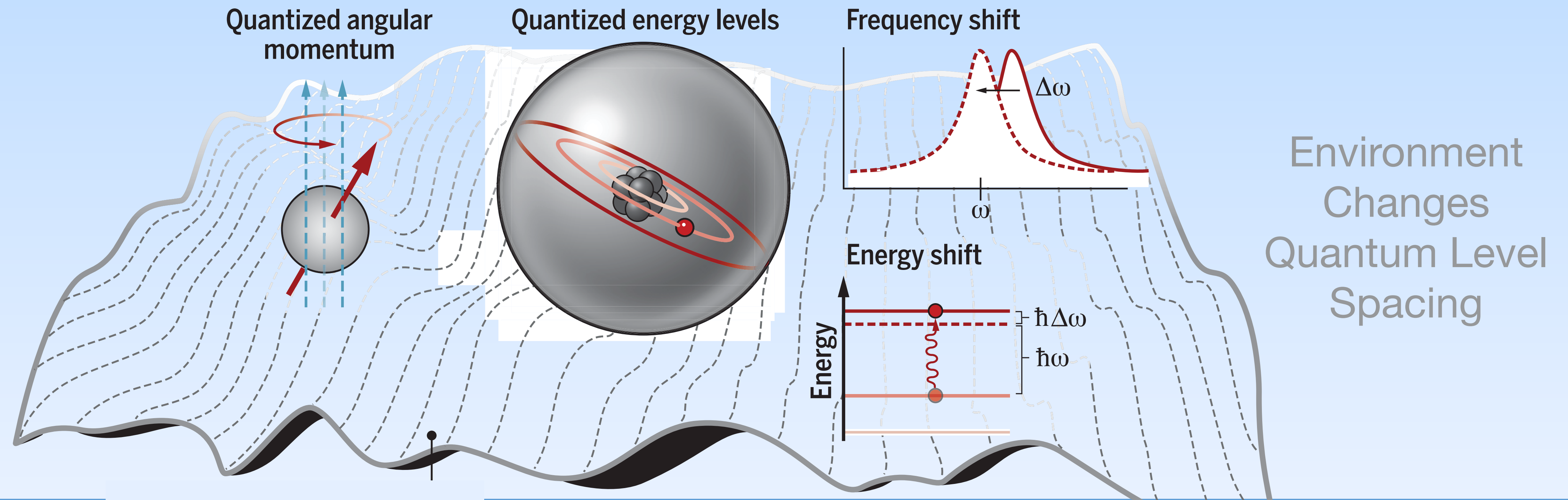
Spectroscopy

Quantum Mechanics

Methods of probing



Measurement concepts

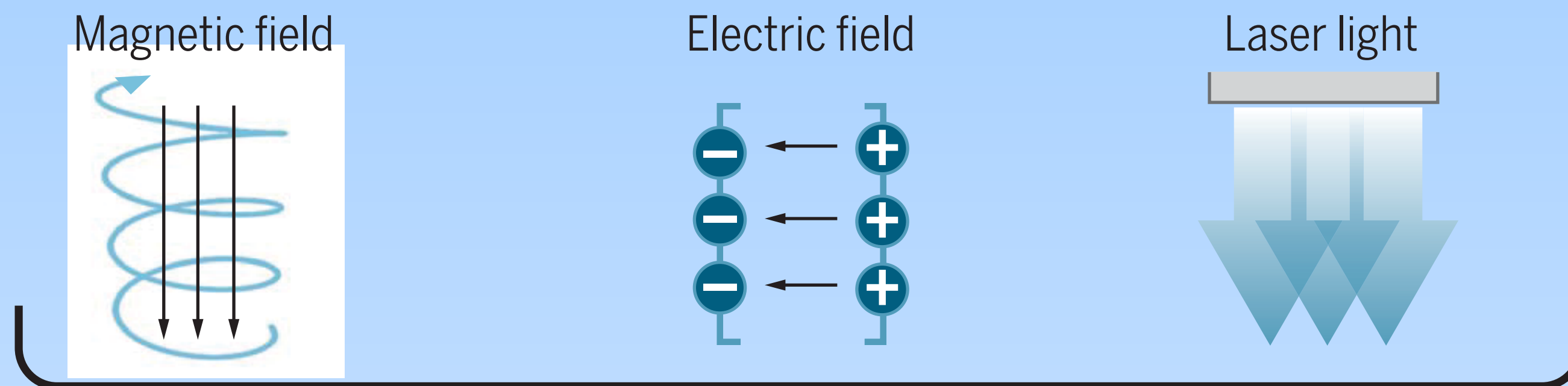


Spectroscopy

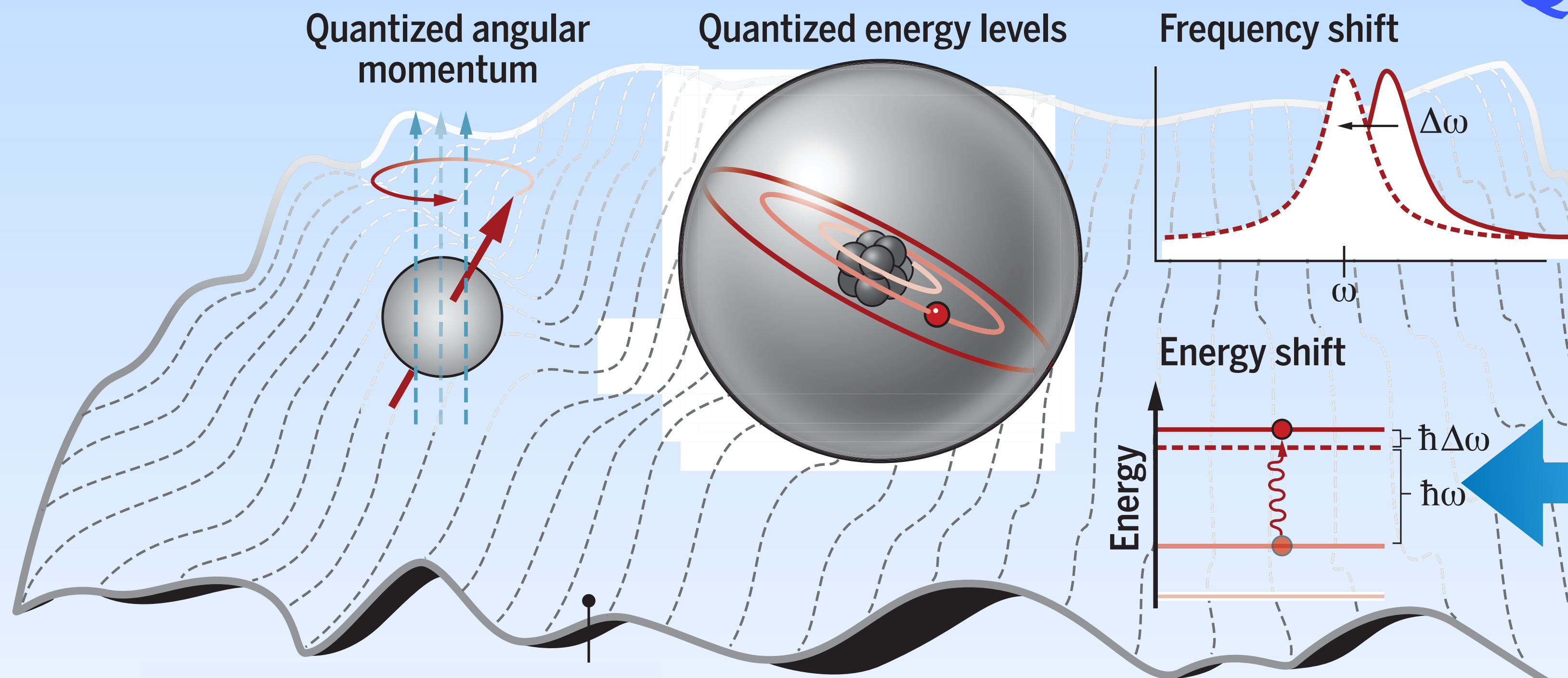
Coherent Radiation Sources

Control

Methods of probing



Measurement concepts



Quantum Control

1940s-1970s
Laser,
Microwaves, etc.

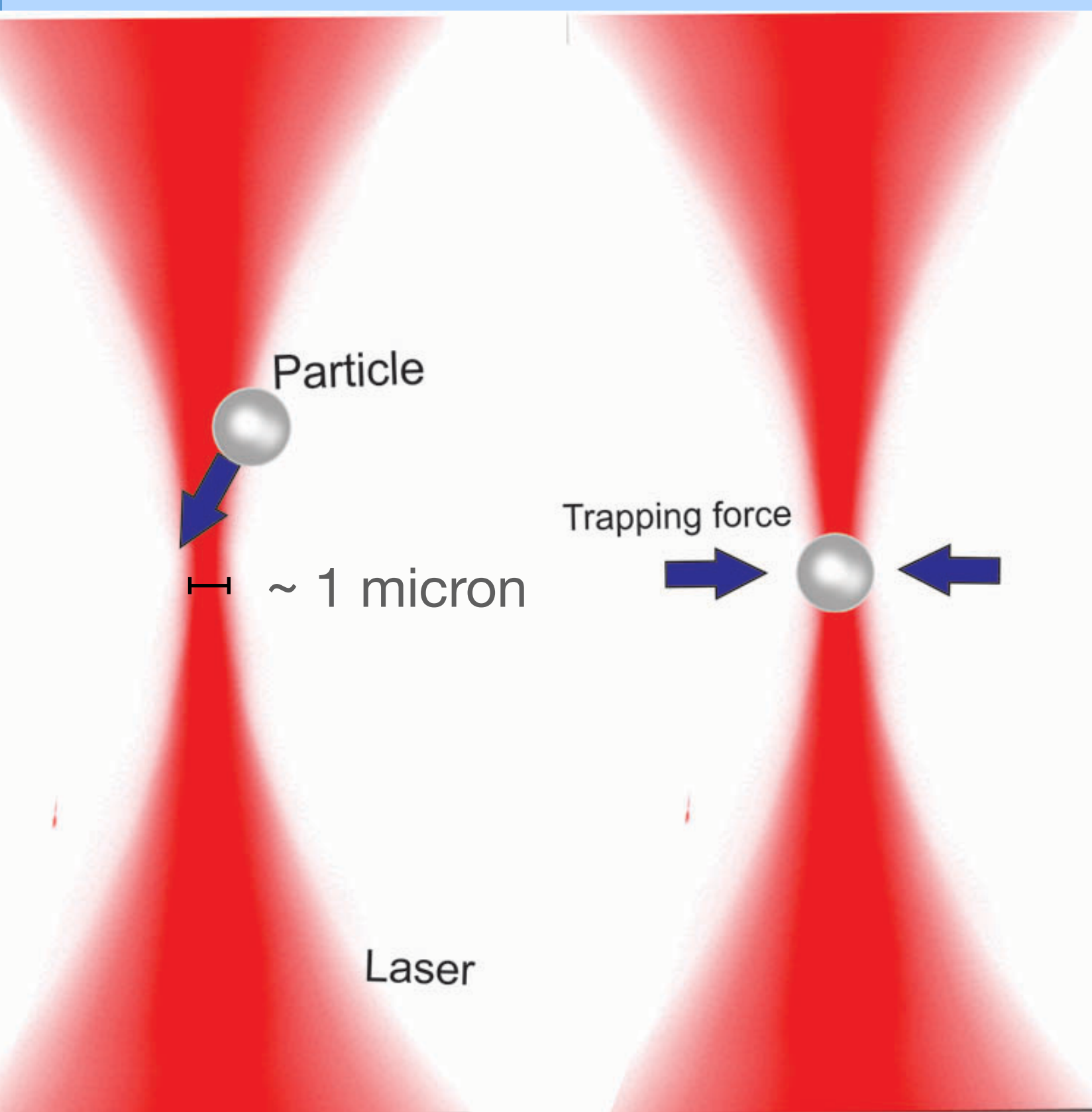
2014

Spectroscopy

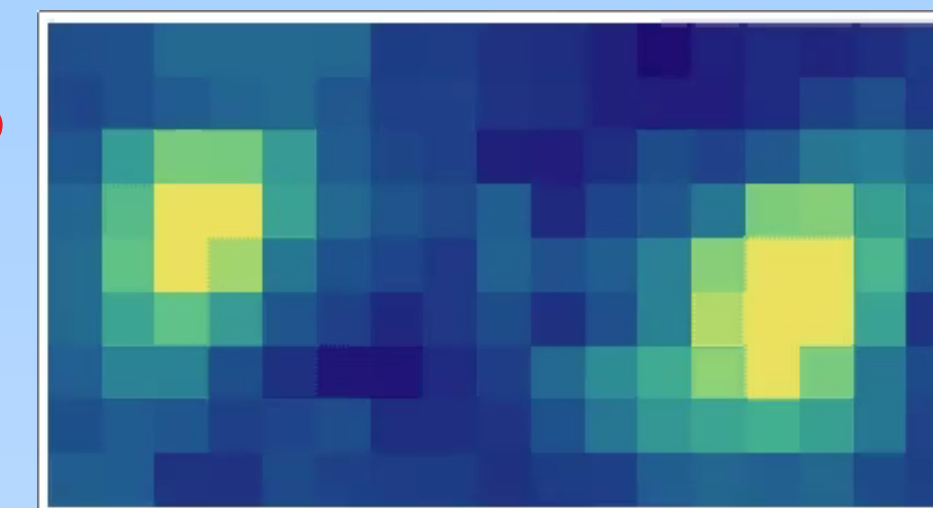
Coherent Radiation Sources

Control

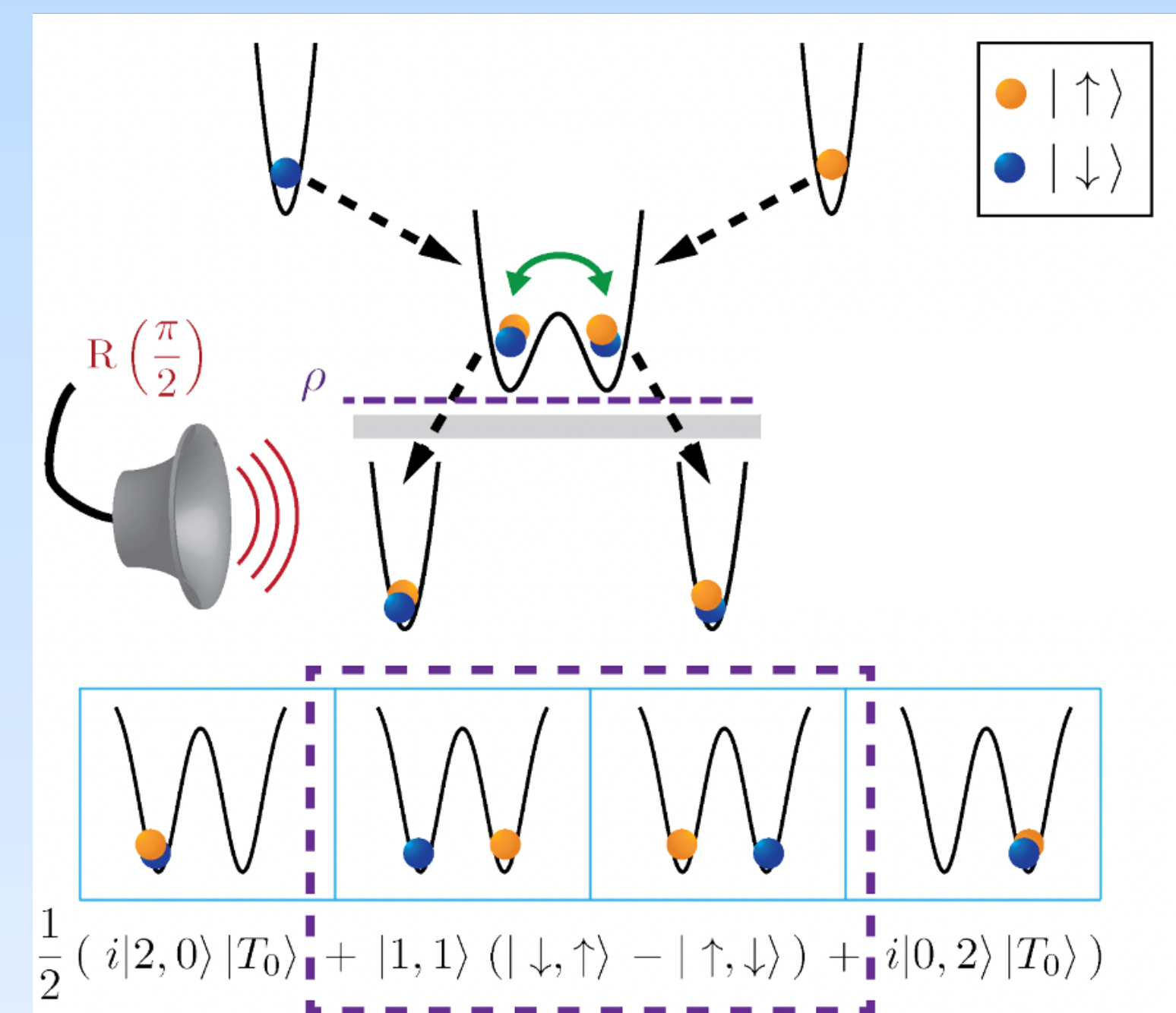
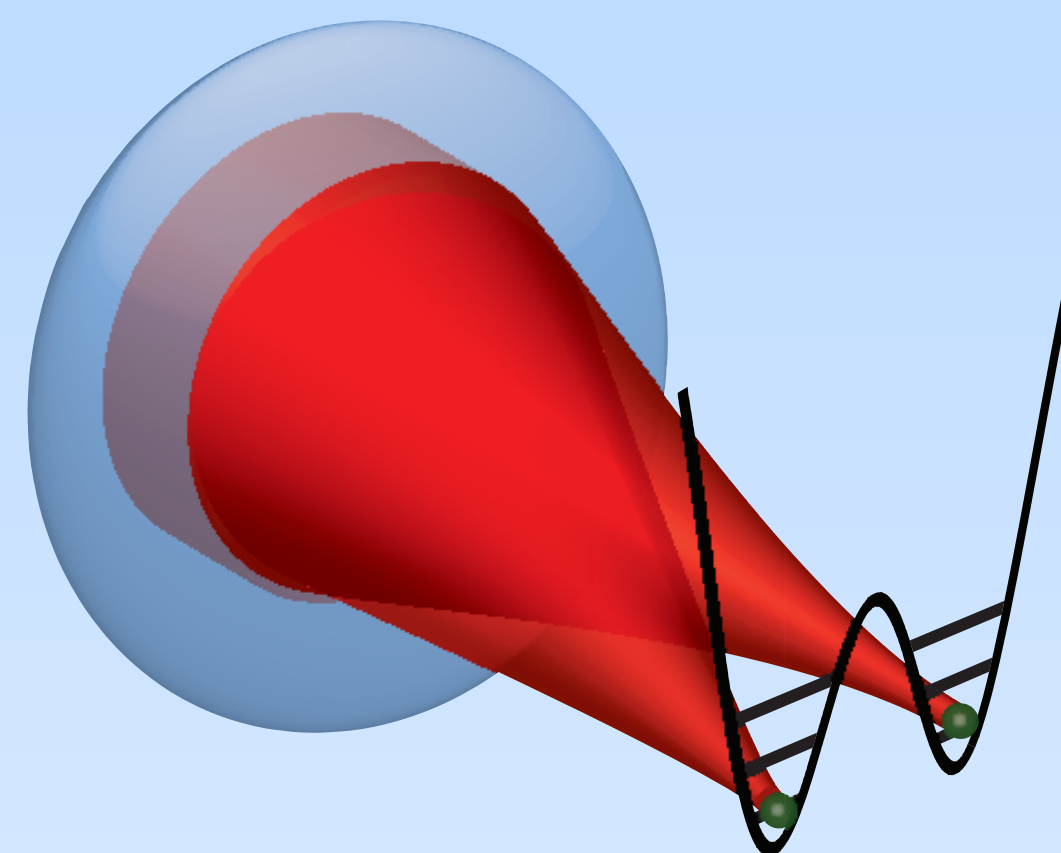
Neutral Atoms
in
“Optical Tweezers”



Control of Single Neutral Atoms
milliKelvin temperatures
Gotten from laser cooling



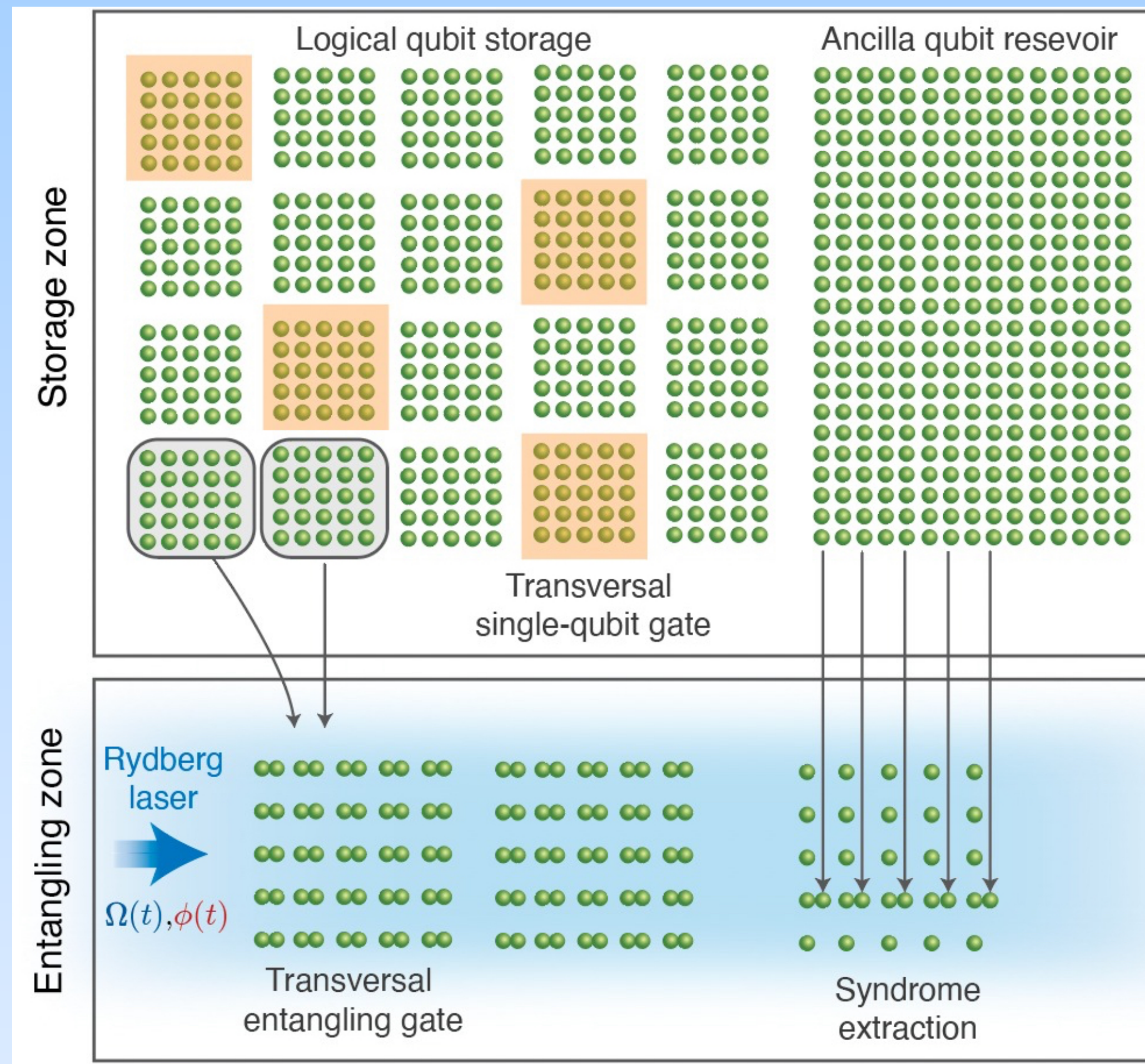
A. M. Kaufman et al, Two-particle quantum interference in tunnel-coupled optical tweezers, Science (2014)



B. J. Lester et al, Measurement based entanglement of non-interacting bosonic atoms, PRL (2018)

Fast Forward
10 Years.... To 2024

Logical Quantum Processor



**Zoned architecture with
280 physical qubit, control over 48
logical qubits**

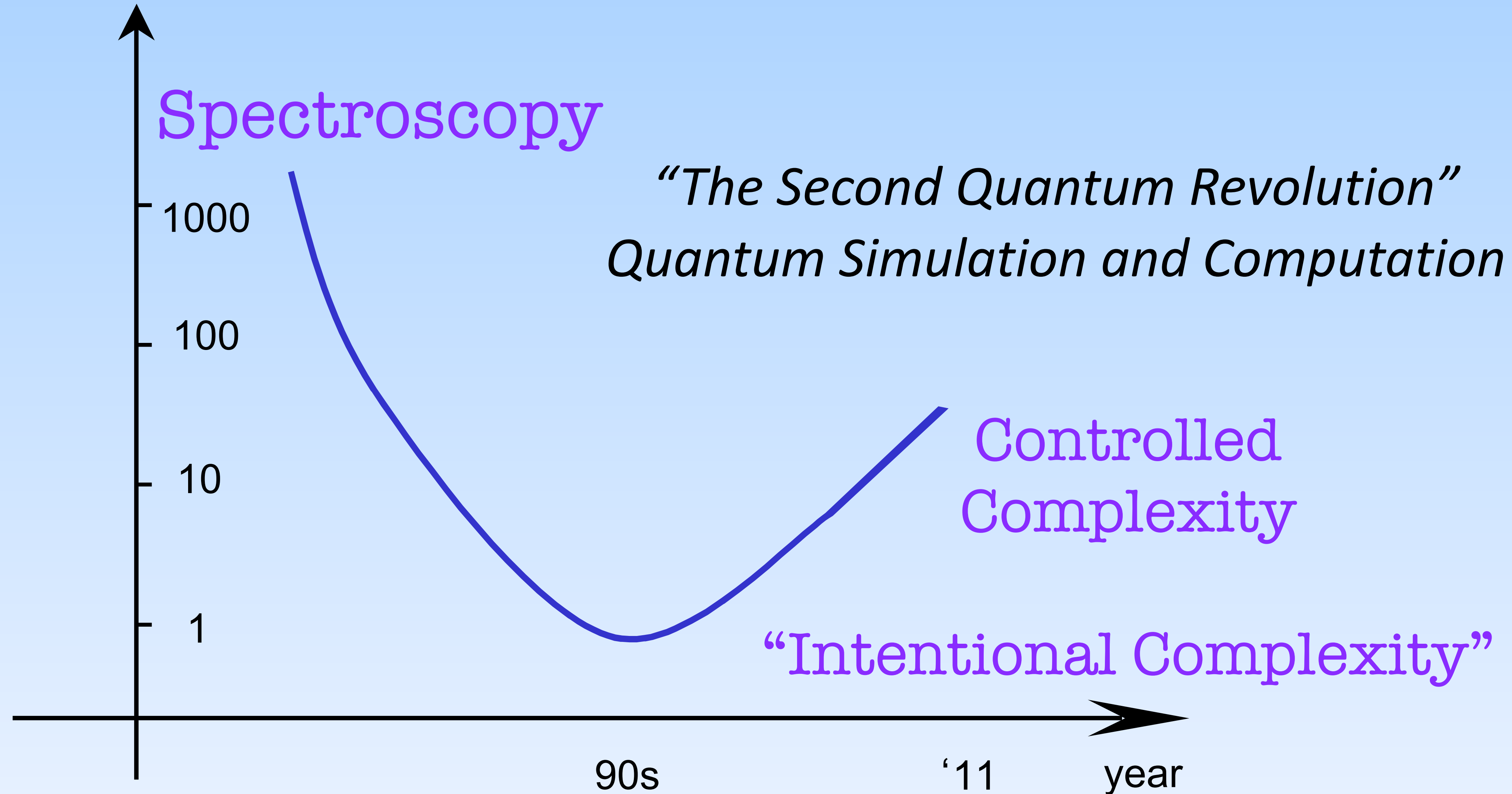
Dolev Bluvstein et al, Nature 2024, Harvard, MIT (Vuletic, Lukin, Greiner)



Spectroscopy ↔ **Control** ↔ **Complexity**

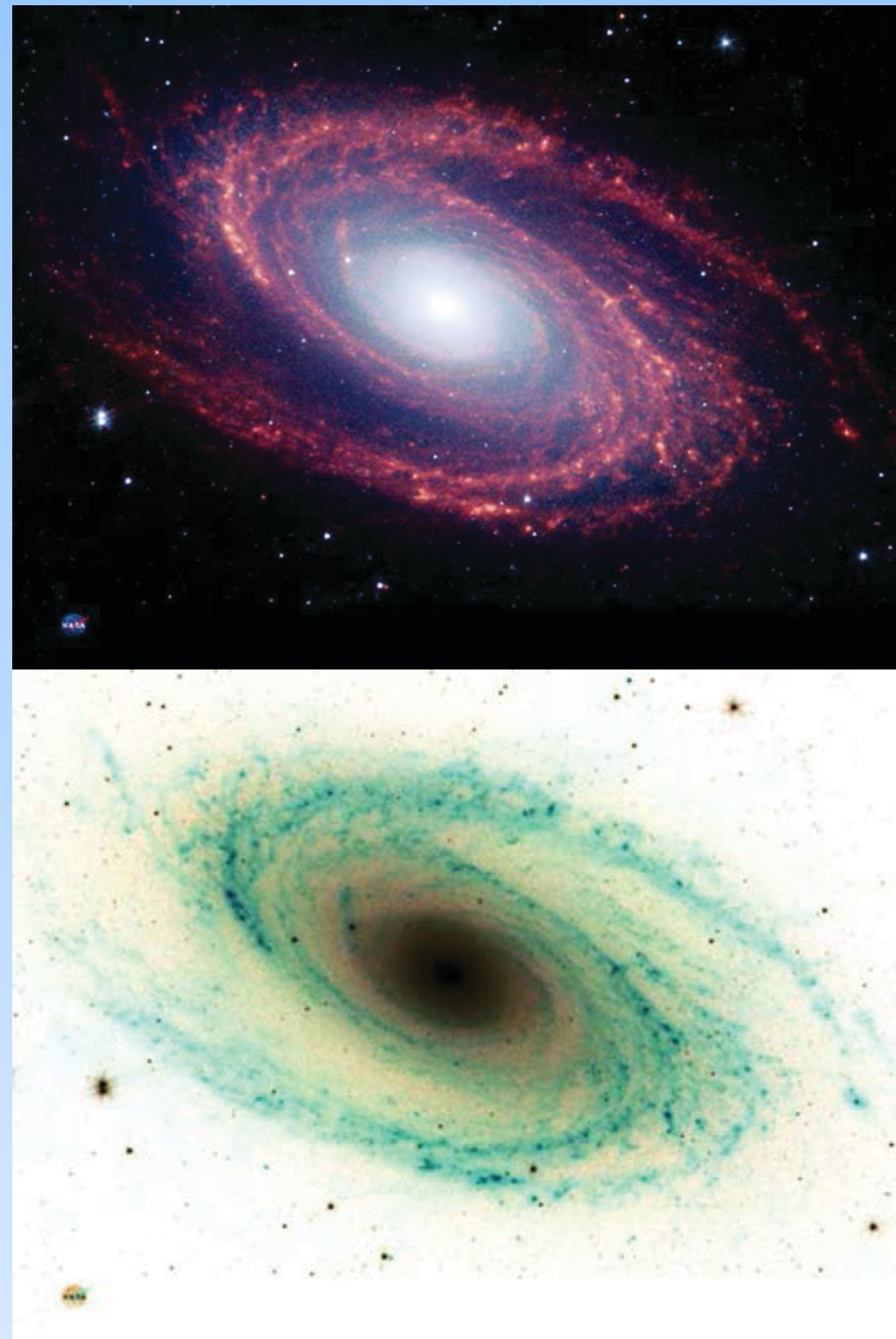
AMO History

Number of quanta in experiments
(atoms, molecules, ions, photons, electrons...)

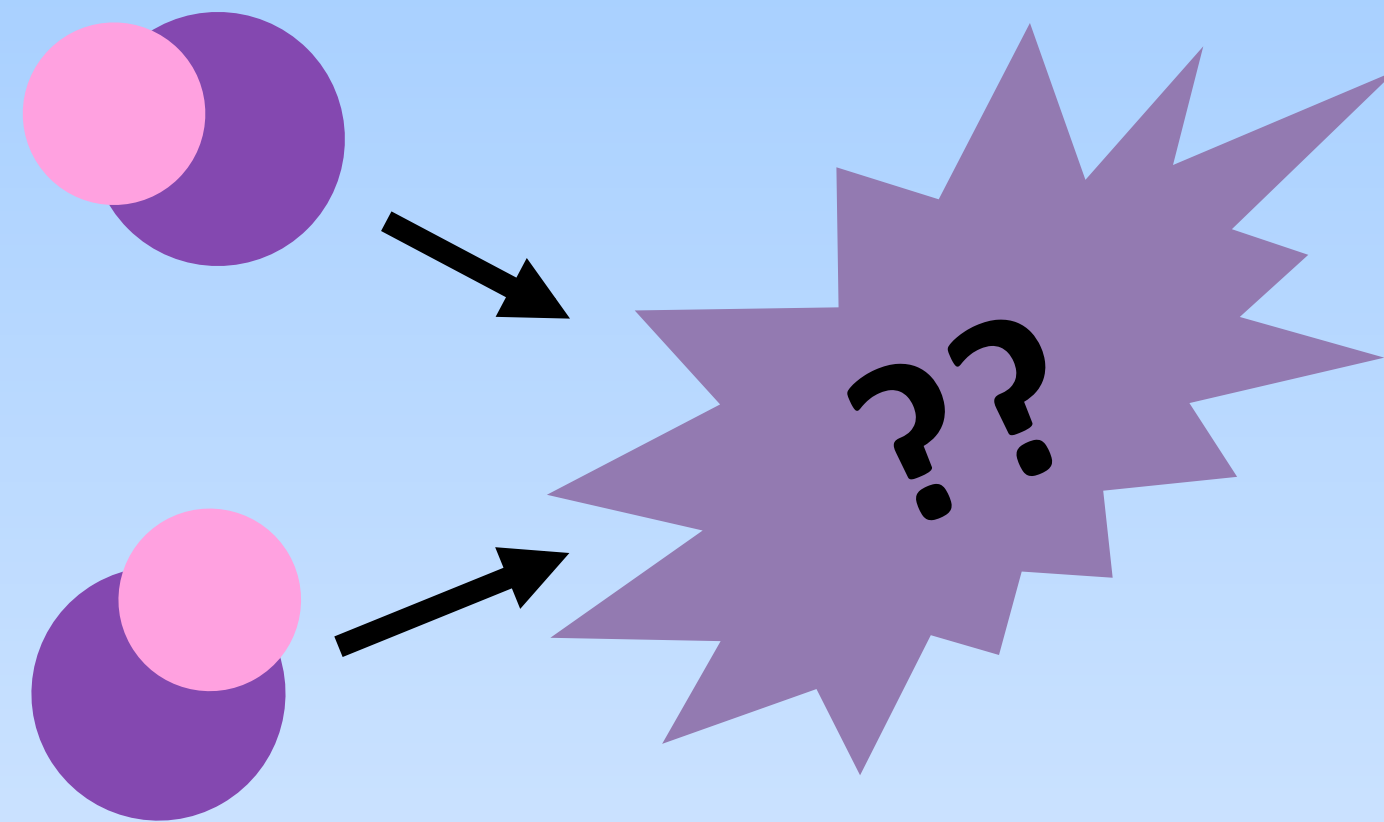


Doyle Group Research “Strategy”

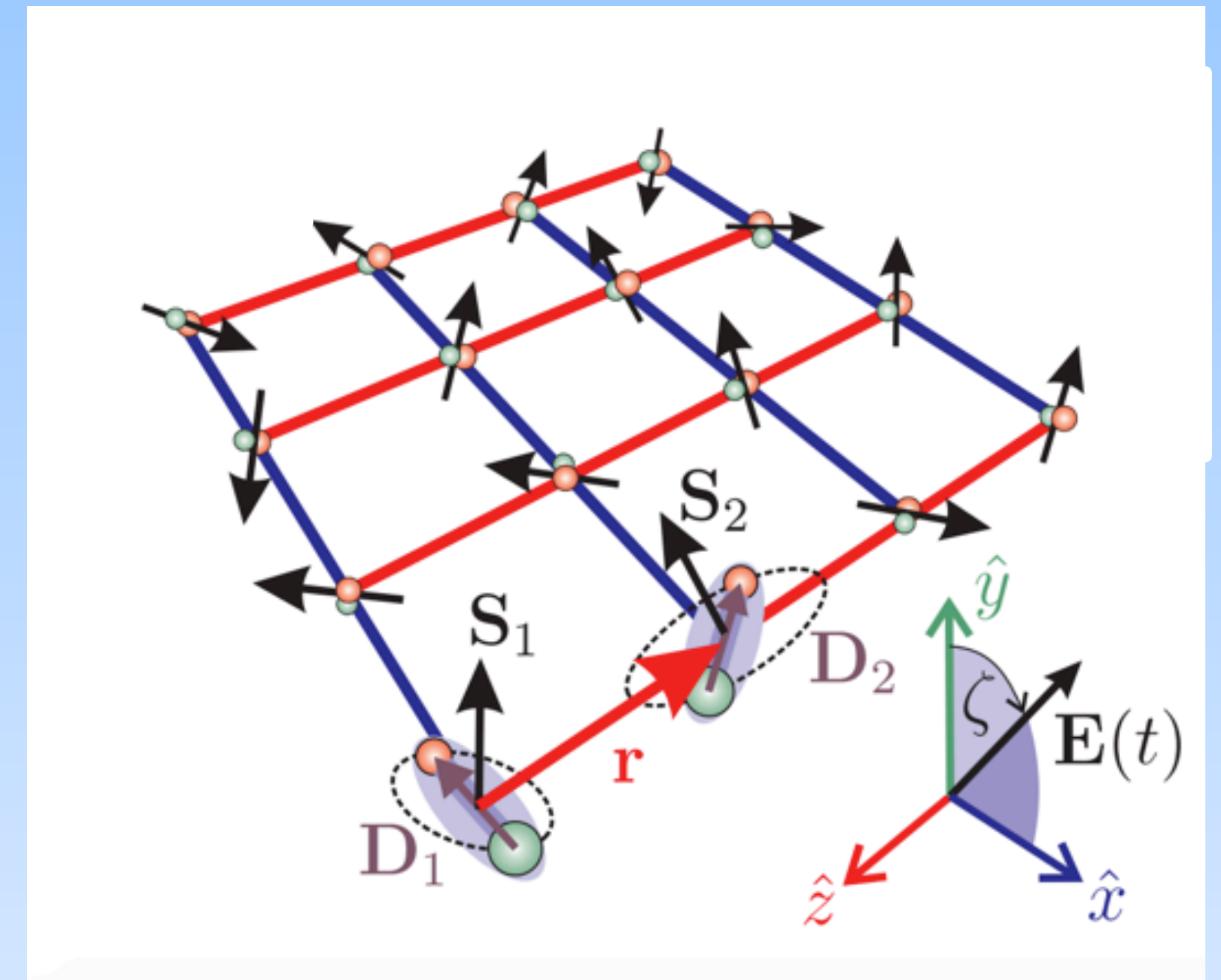
Molecular Complexity might be Harnessed to Advance Areas of AMO



**Metrology-based
Particle Physics**



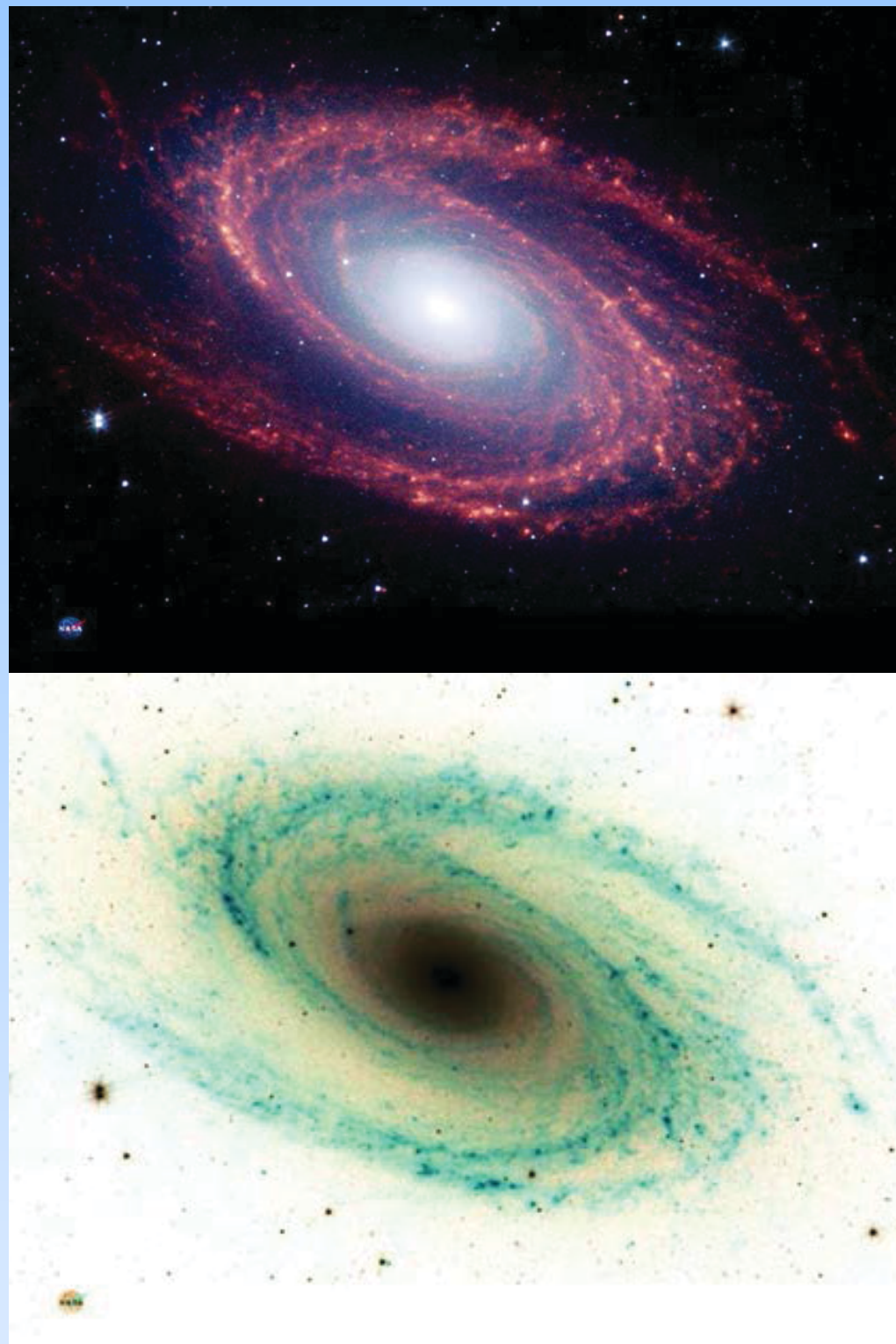
Collisions and Chemistry



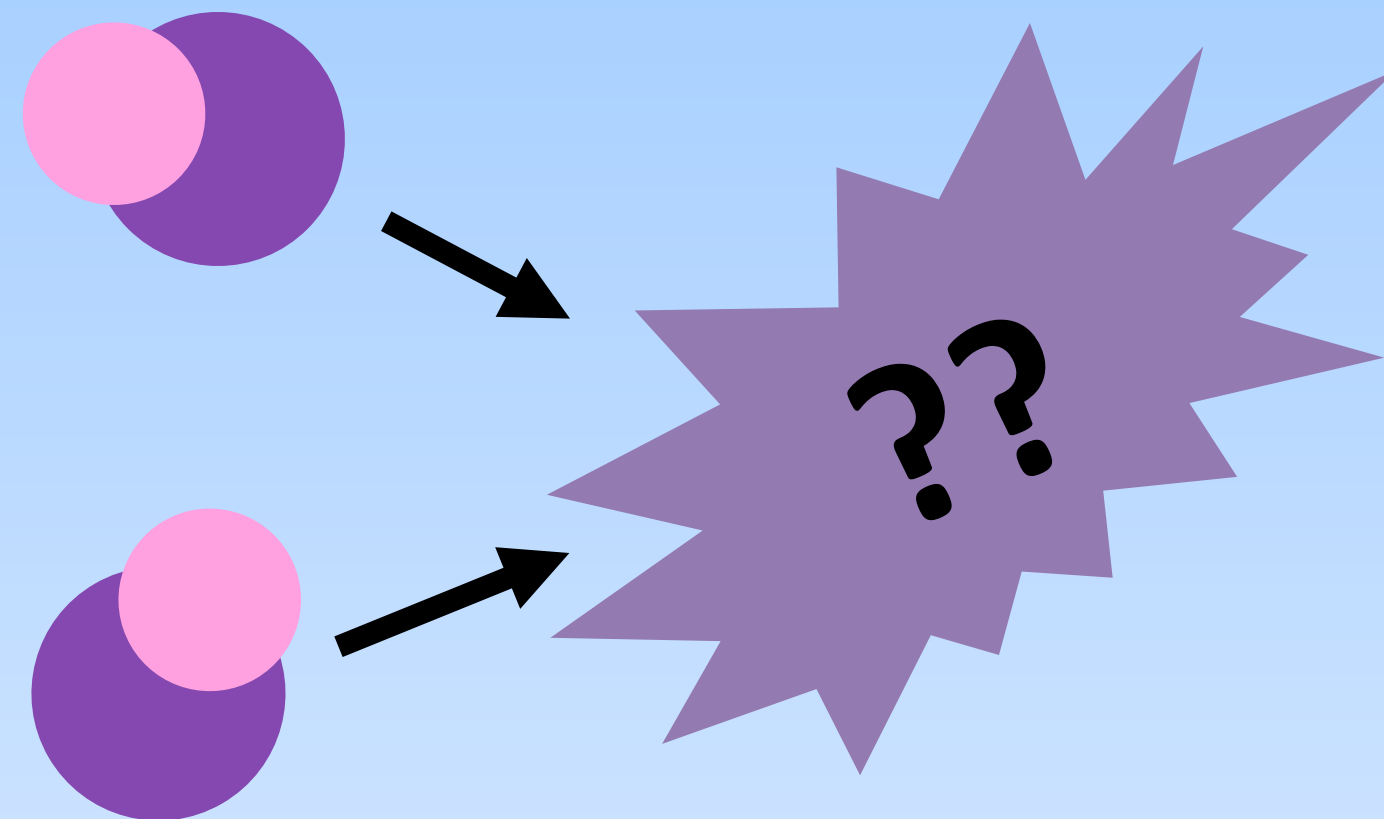
**Quantum Simulation/
Information**

Doyle Group Research “Strategy”

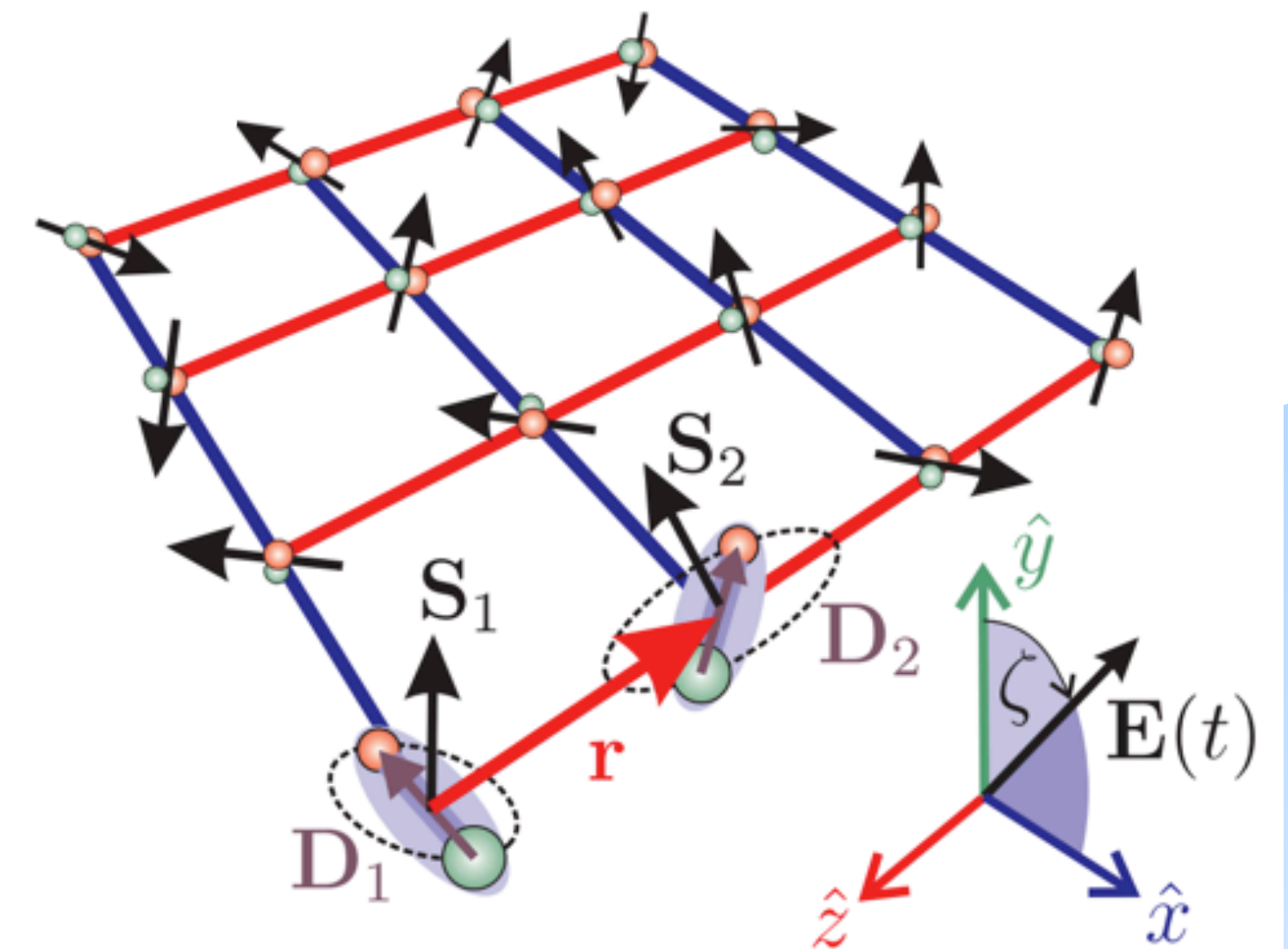
Molecular Complexity might be Harnessed to Advance Areas of AMO



Precision Measurements

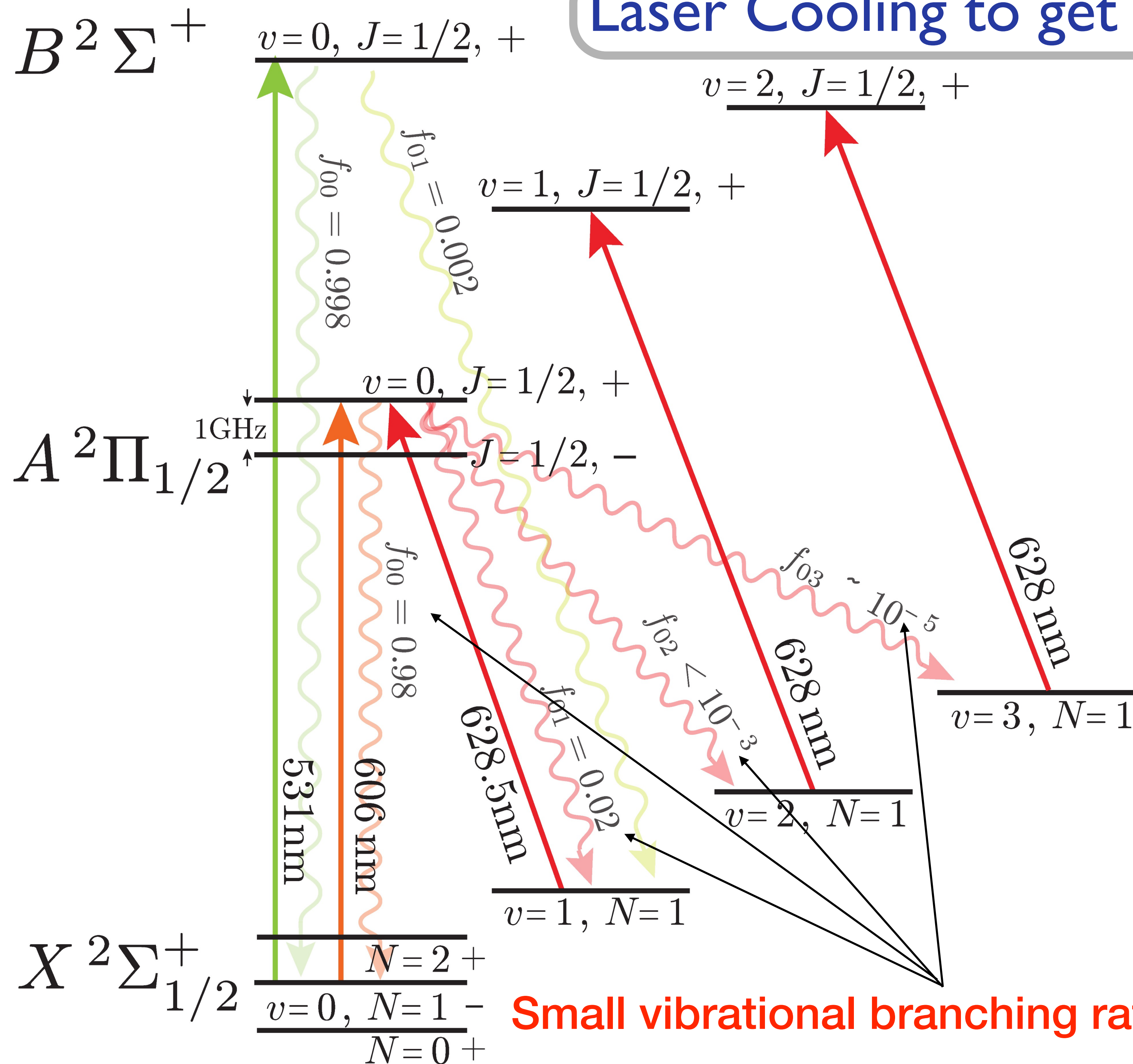


Collisions and Chemistry



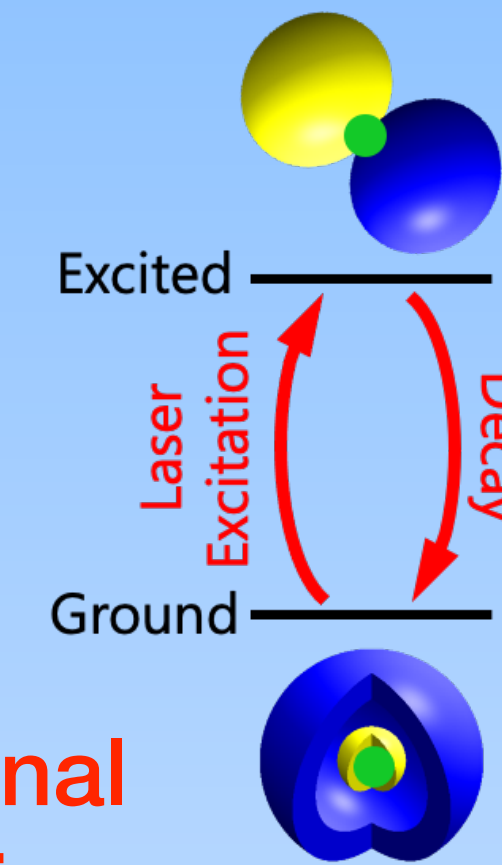
Quantum Simulation/
Information

Laser Cooling to get milliKelvin CaF Molecules



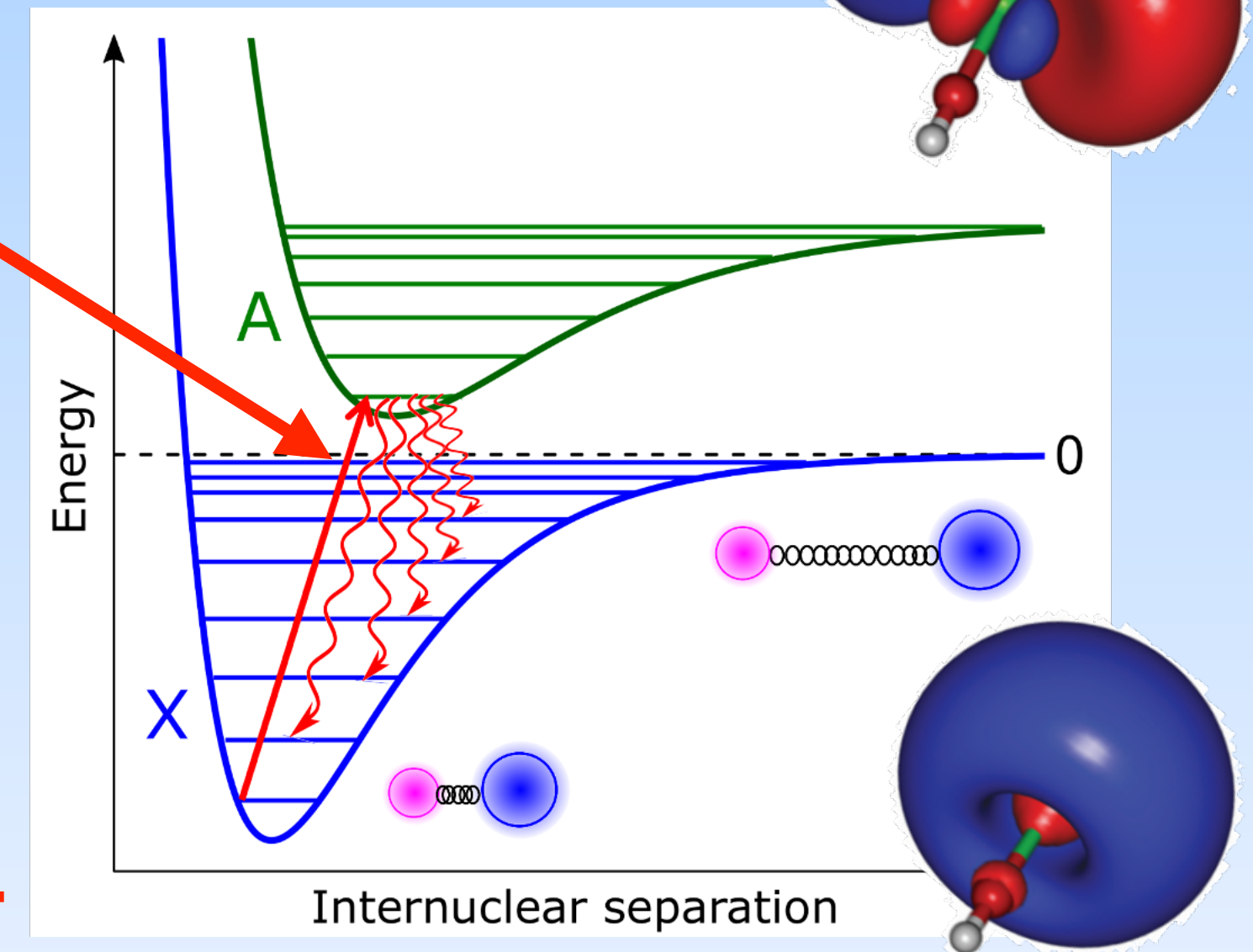
Small vibrational branching ratios for CaF

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



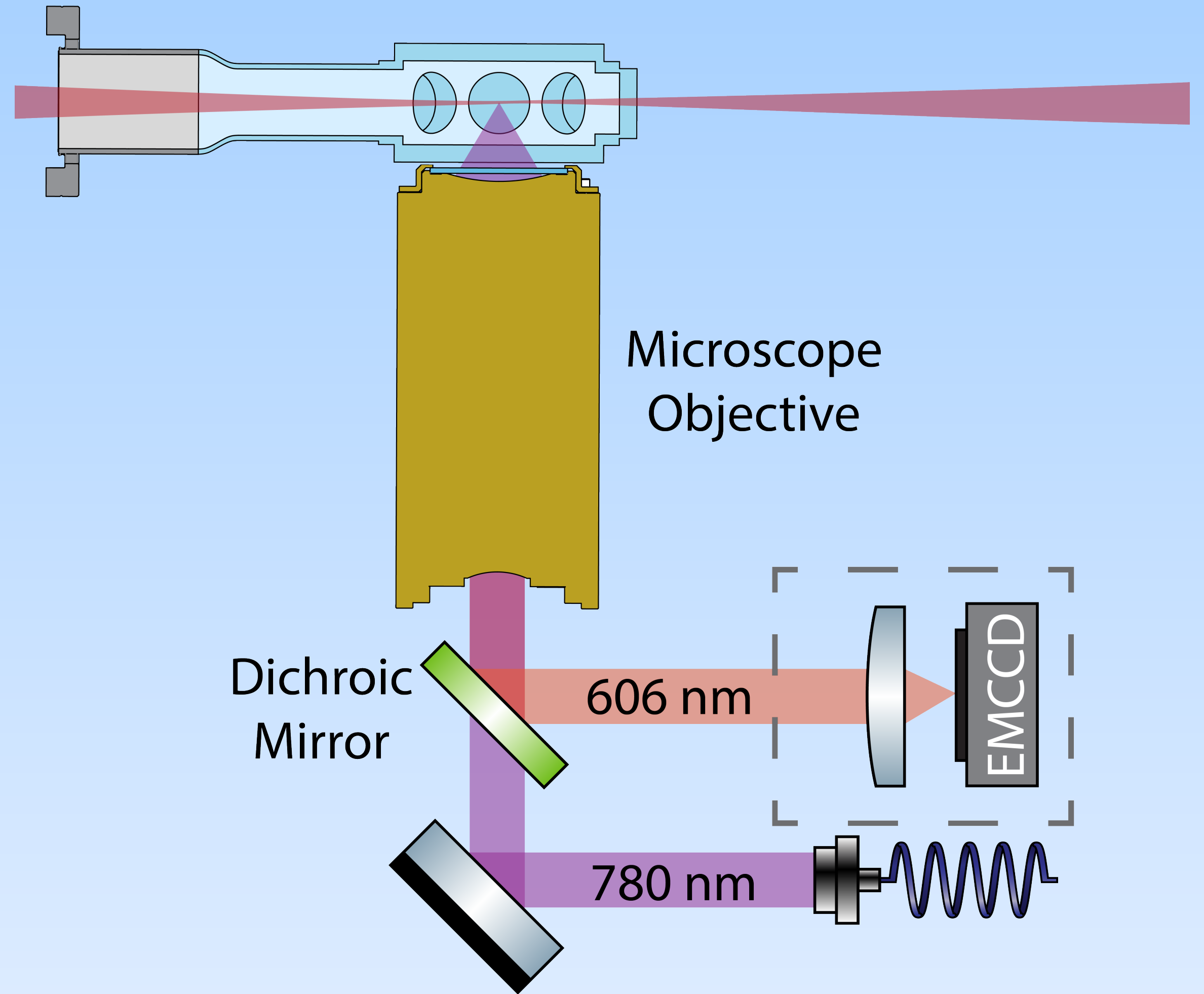
~3,000-7,000 Photon Cycles Needed for Full Laser Cooling

Vibrational Branching Ratios

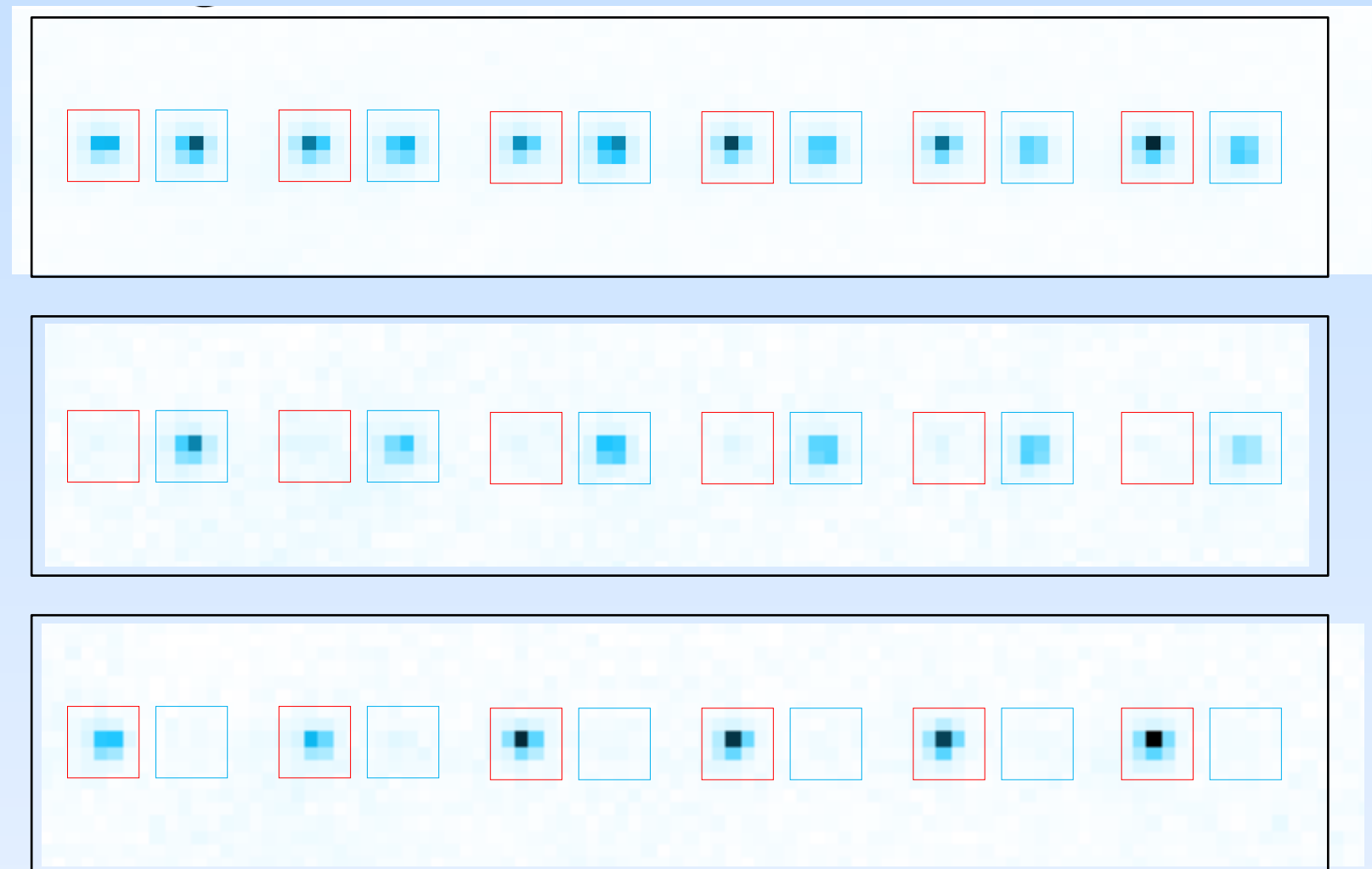


CaF in Tweezer Arrays

Rearrange



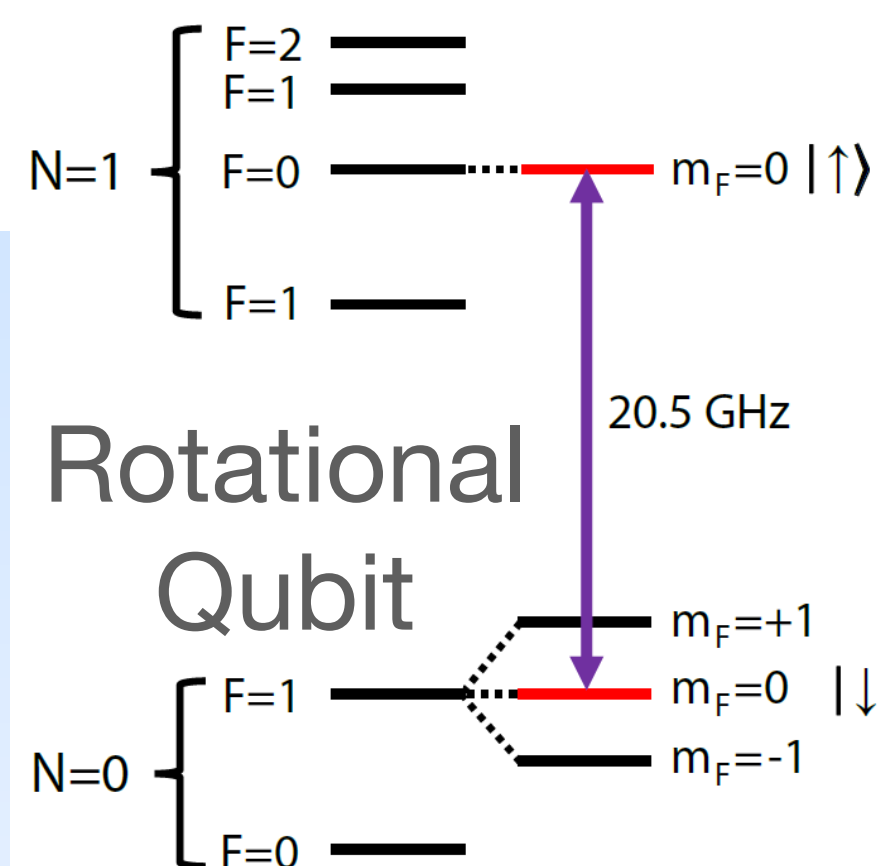
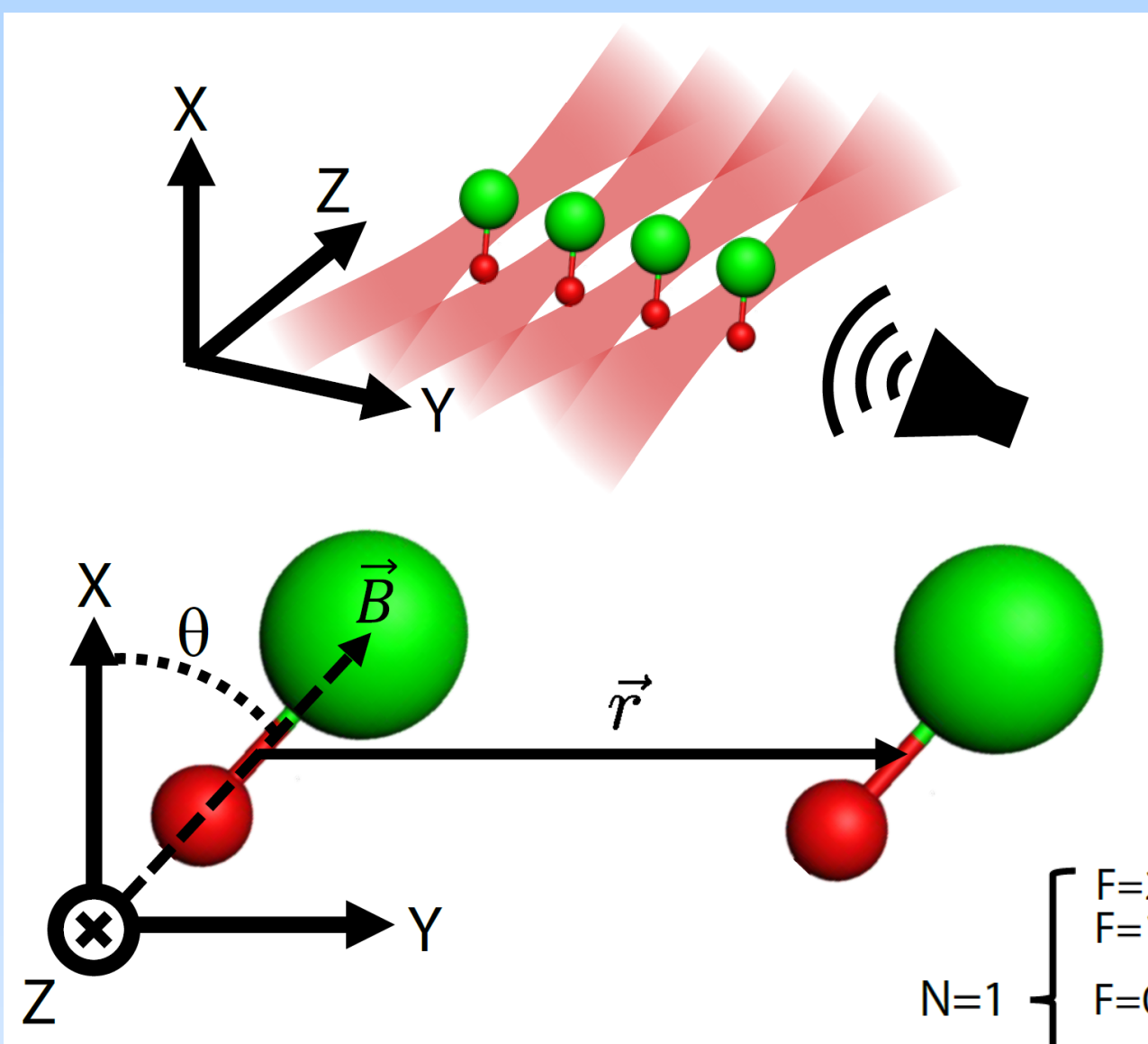
Site Resolved Addressing



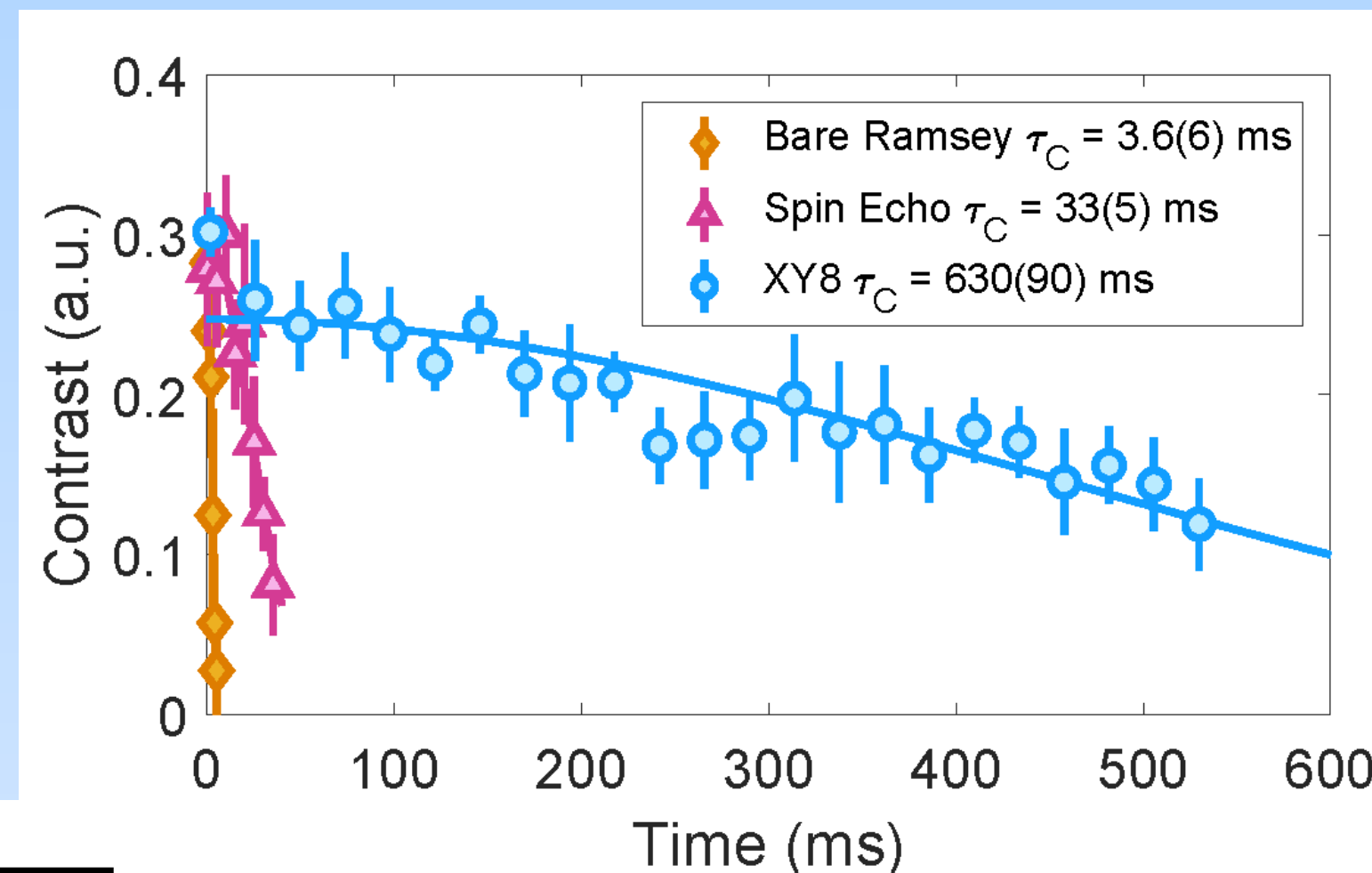
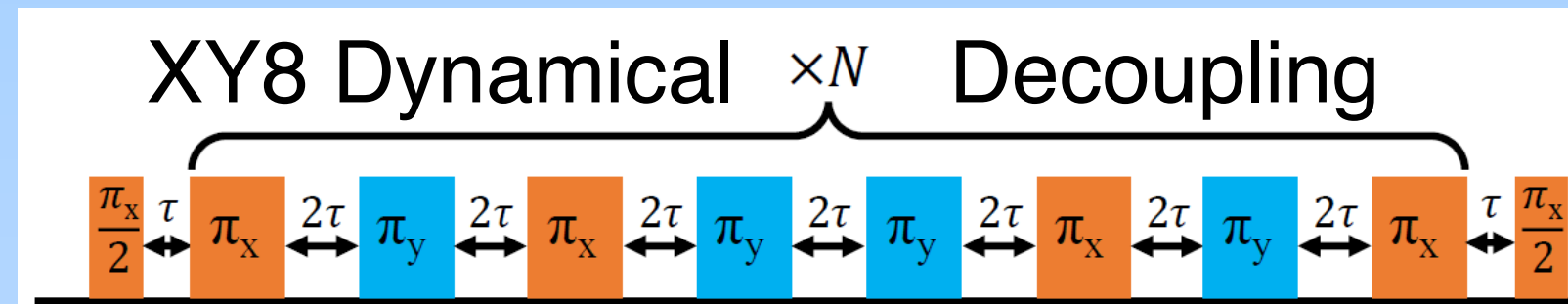
CaF - Dipolar Interactions in Tweezer Arrays

Dipolar Interaction Two Molecules in Nearby Tweezers

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

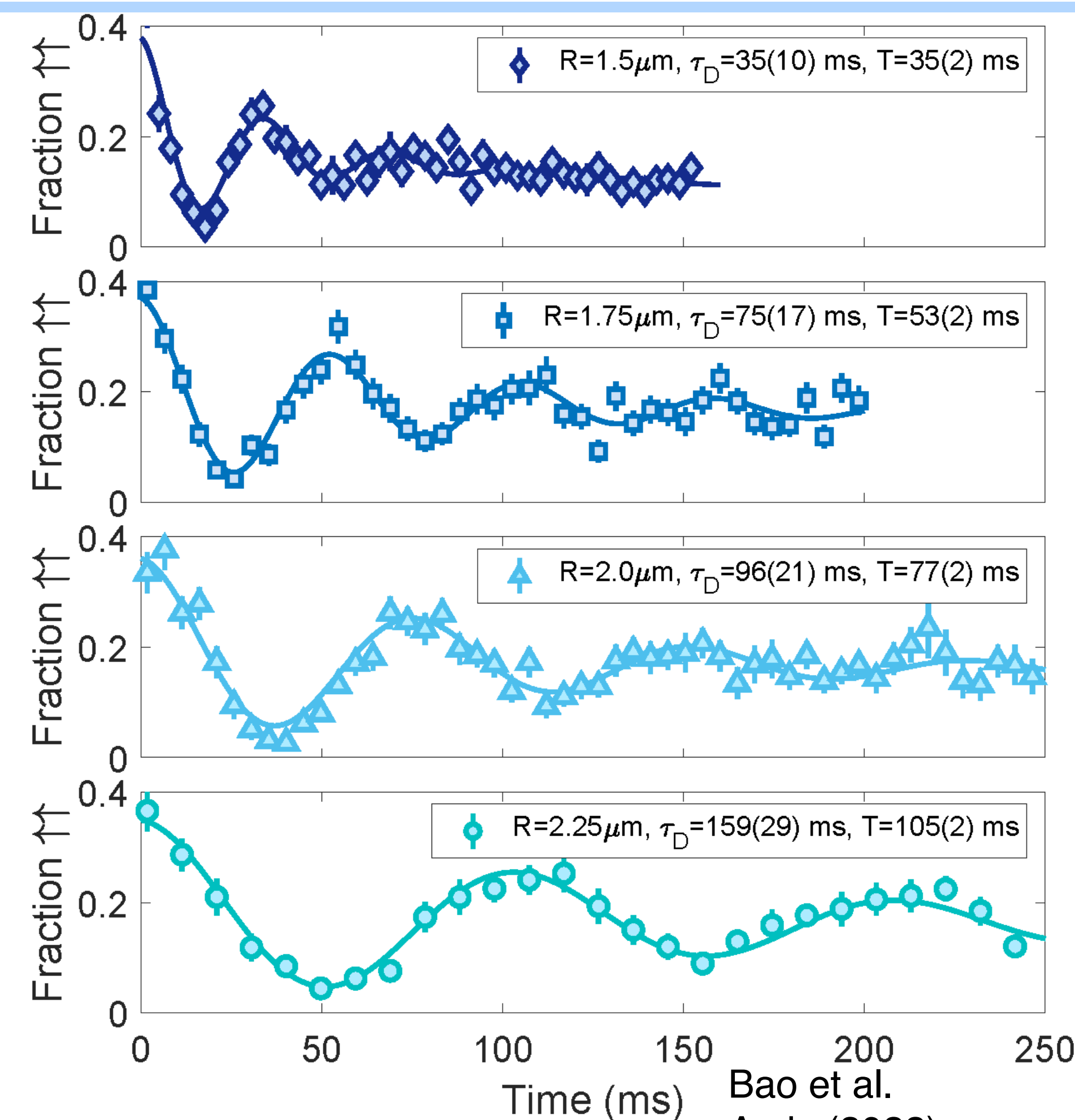


Single Molecule Rotational Qbit Coherence Time



Spin Exchange Between Two Molecules

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

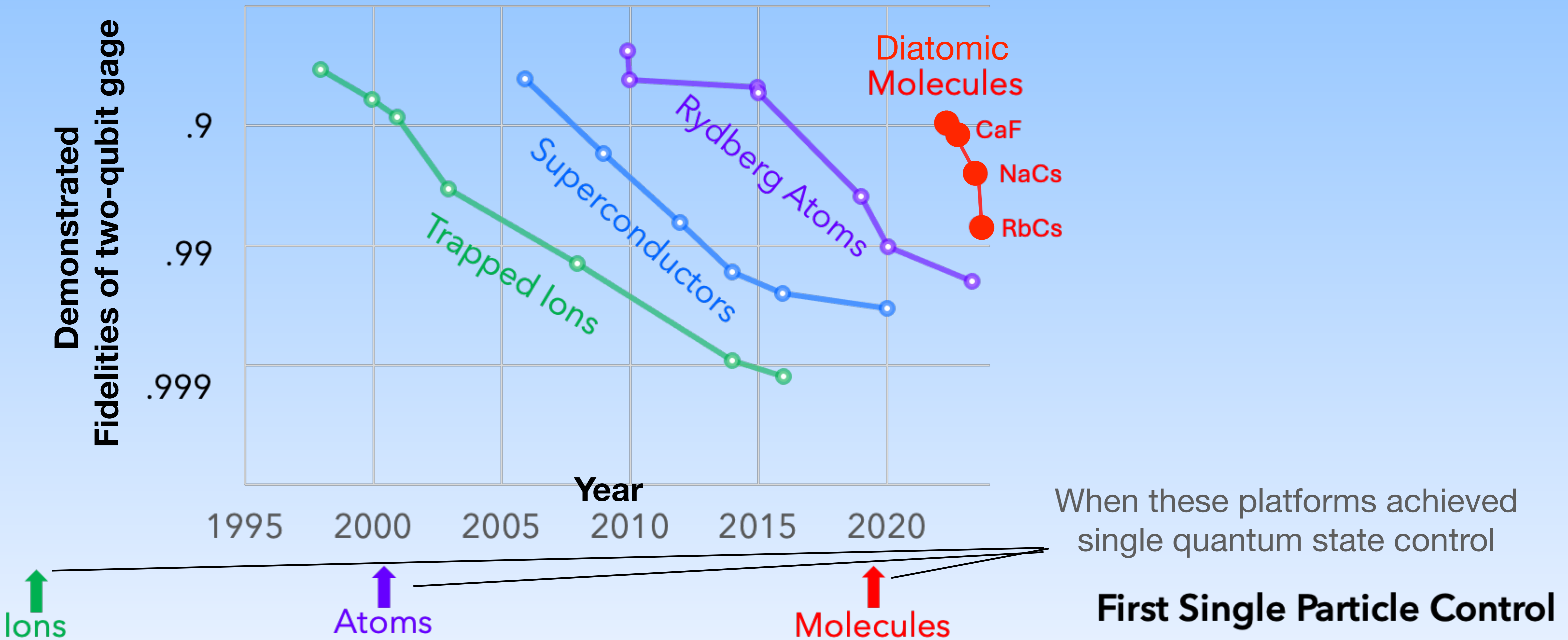


Bell State Fidelity 0.87

A New Qubit Platform

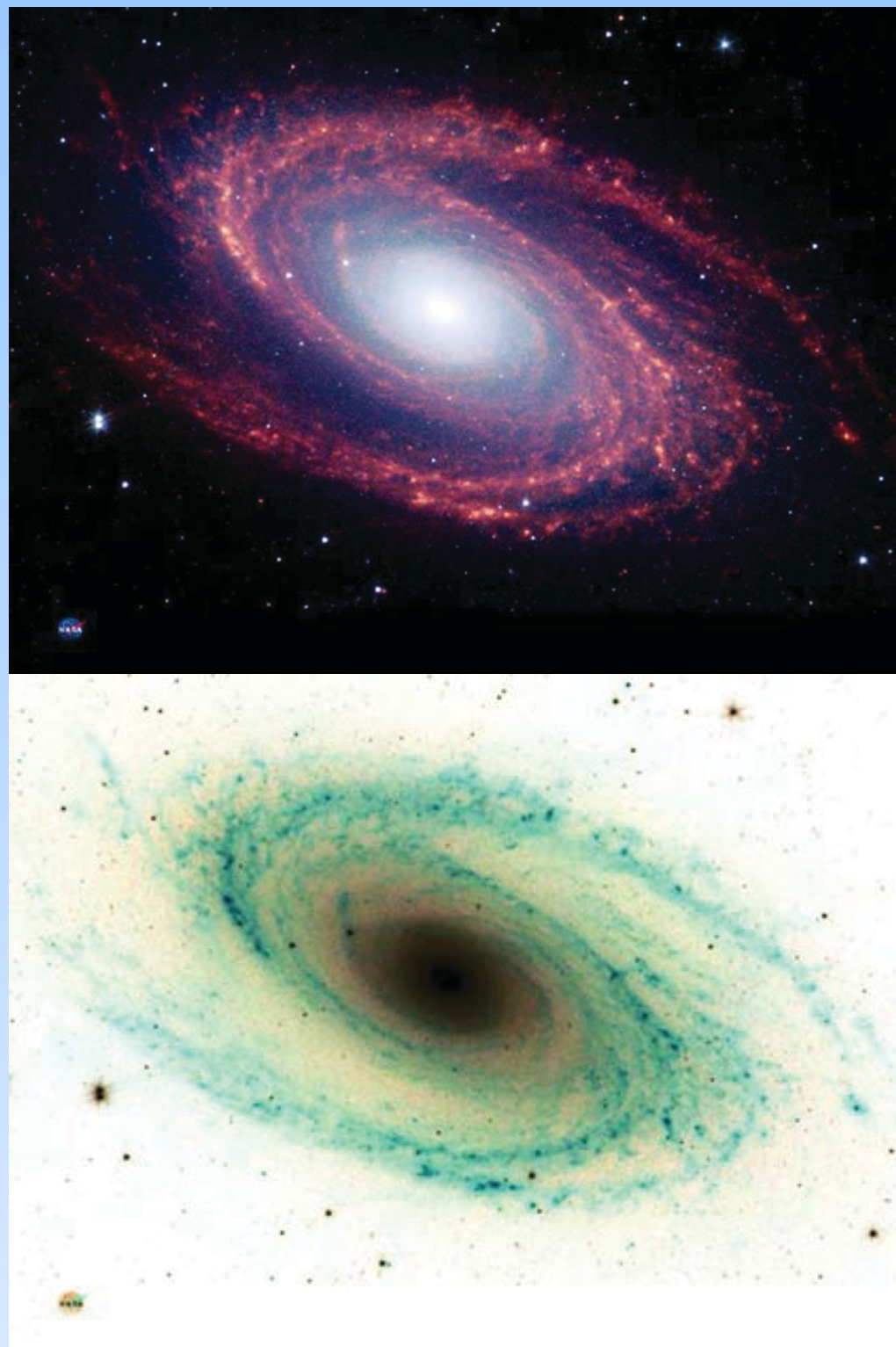
An Extraordinary
Development

Two Qubit Gate Fidelities

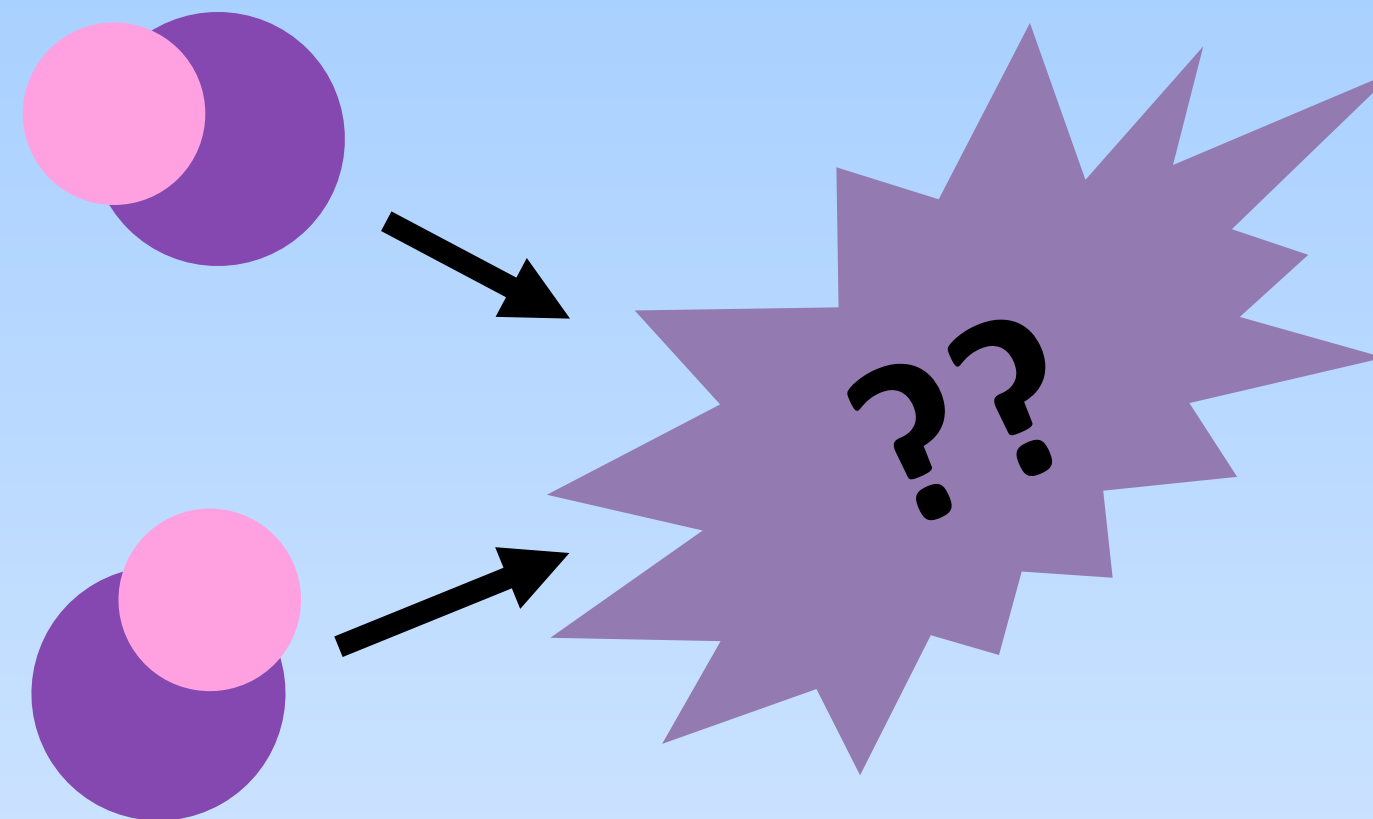


Doyle Group Research “Strategy”

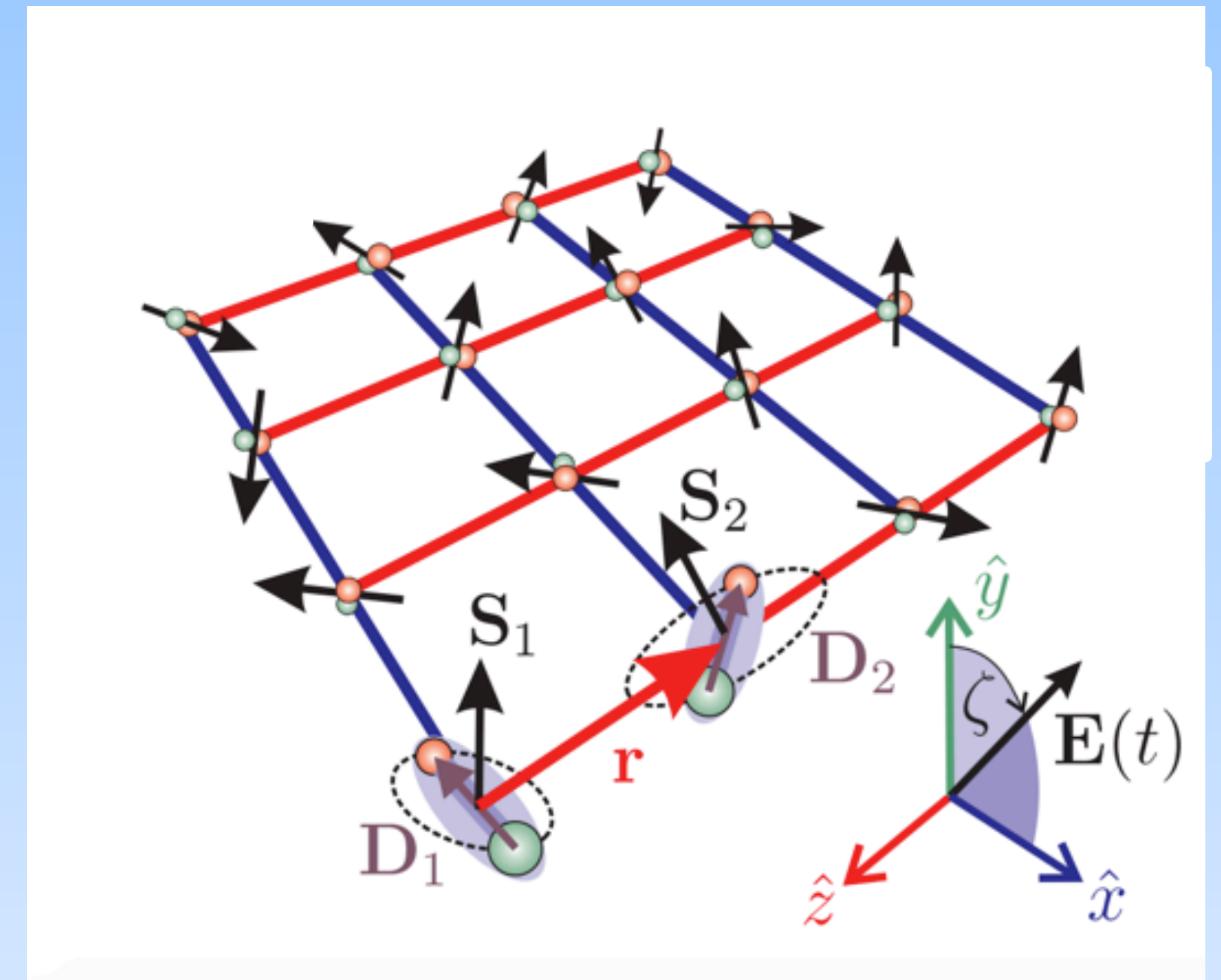
Molecular Complexity might be Harnessed to Advance Areas of AMO



**Metrology-based
Particle Physics**



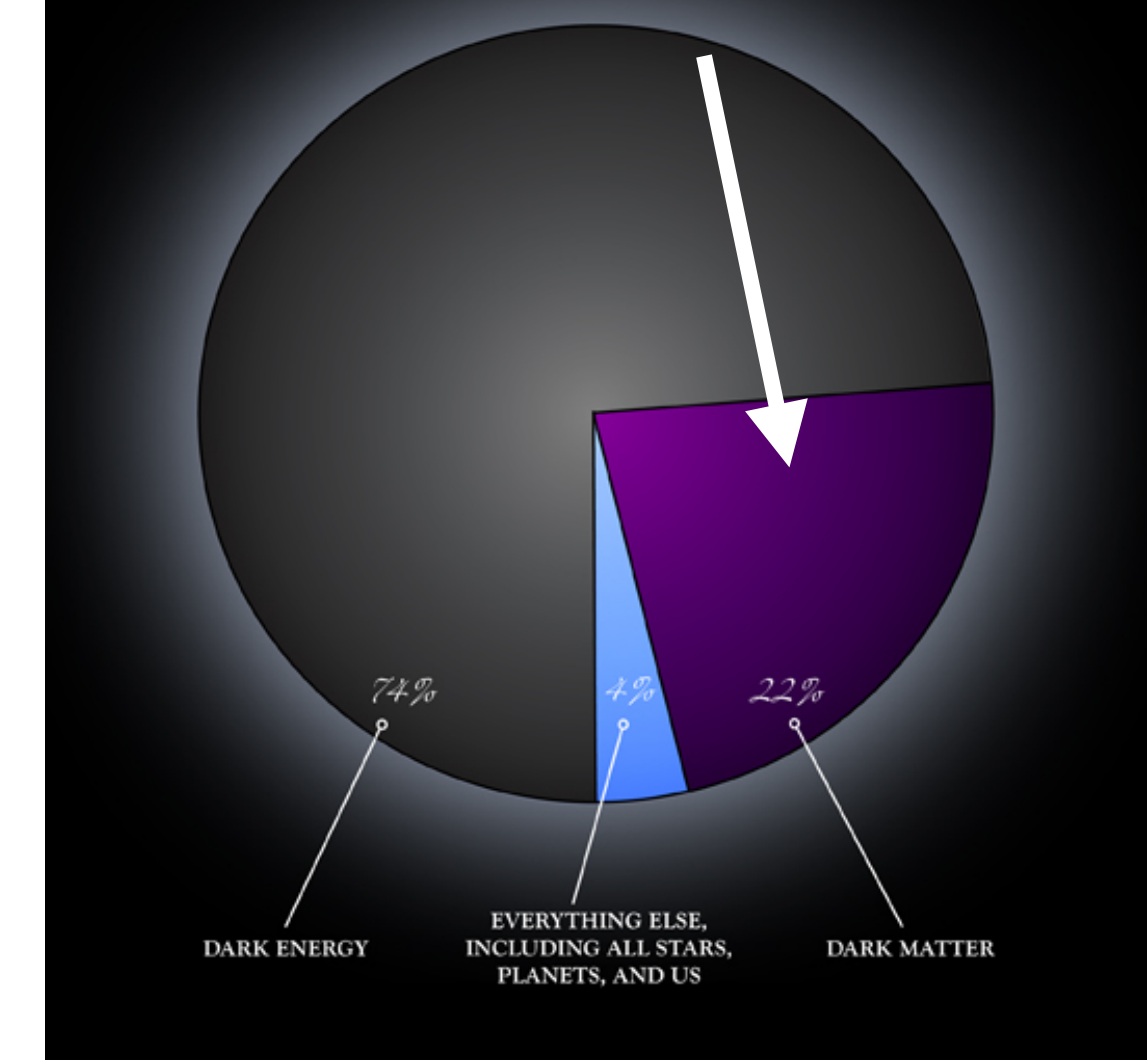
Collisions and Chemistry



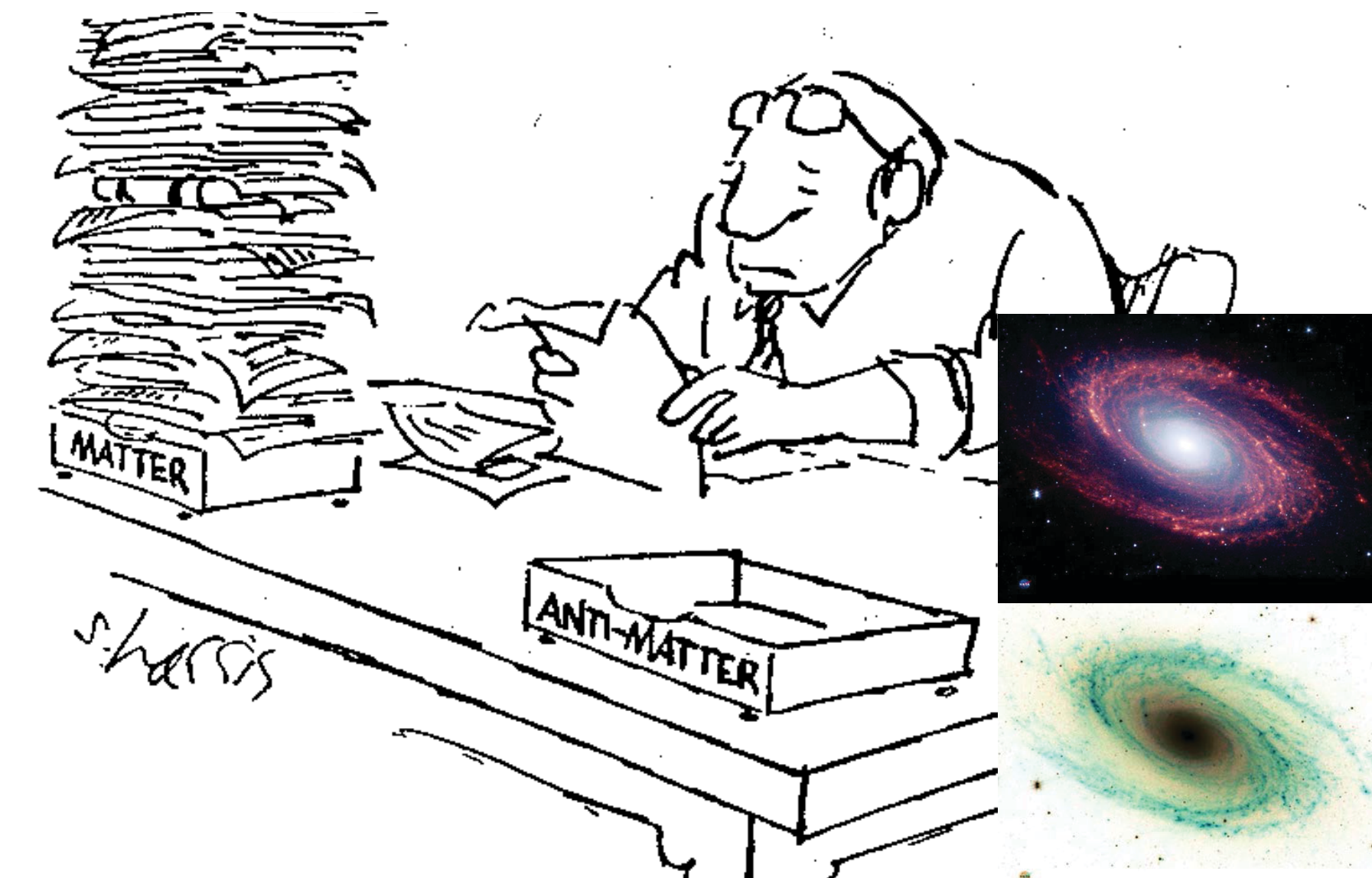
**Quantum Simulation/
Information**

Two Mysteries

1. Dark Matter



2. Matter-Antimatter Asymmetry



YOU ARE HERE

ACCELERATING EXPANSION

A little more than 5 billion years ago, dark energy caused the universe to expand increasingly fast.

INFLATION

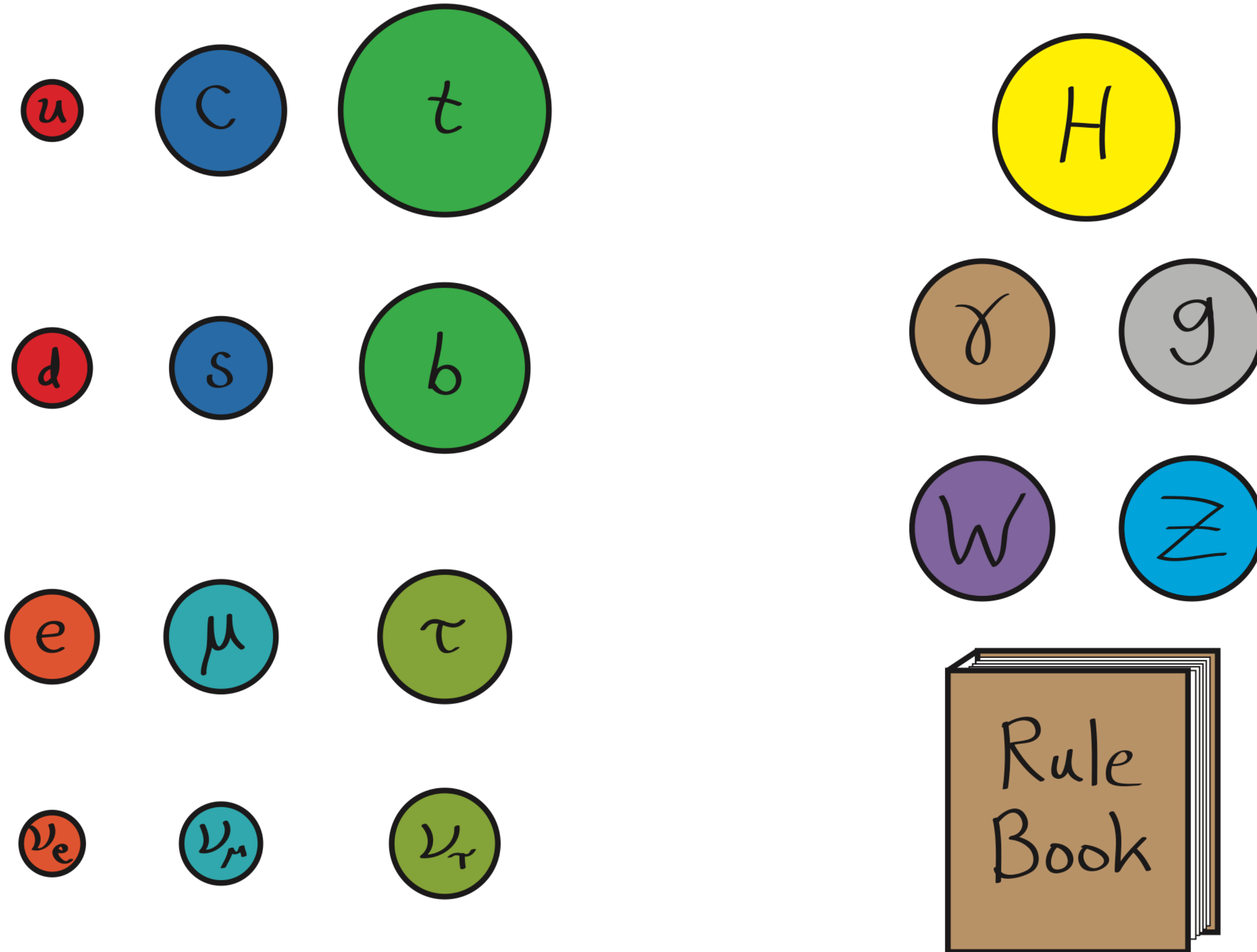
In less than 10^{-30} of a second after the Big Bang, the universe burst open, expanding faster than the speed of light and flinging all the matter and energy in the universe apart in all directions.

BIG BANG

The universe expanded violently from an extremely hot and dense initial state some 13.7 billion years ago.

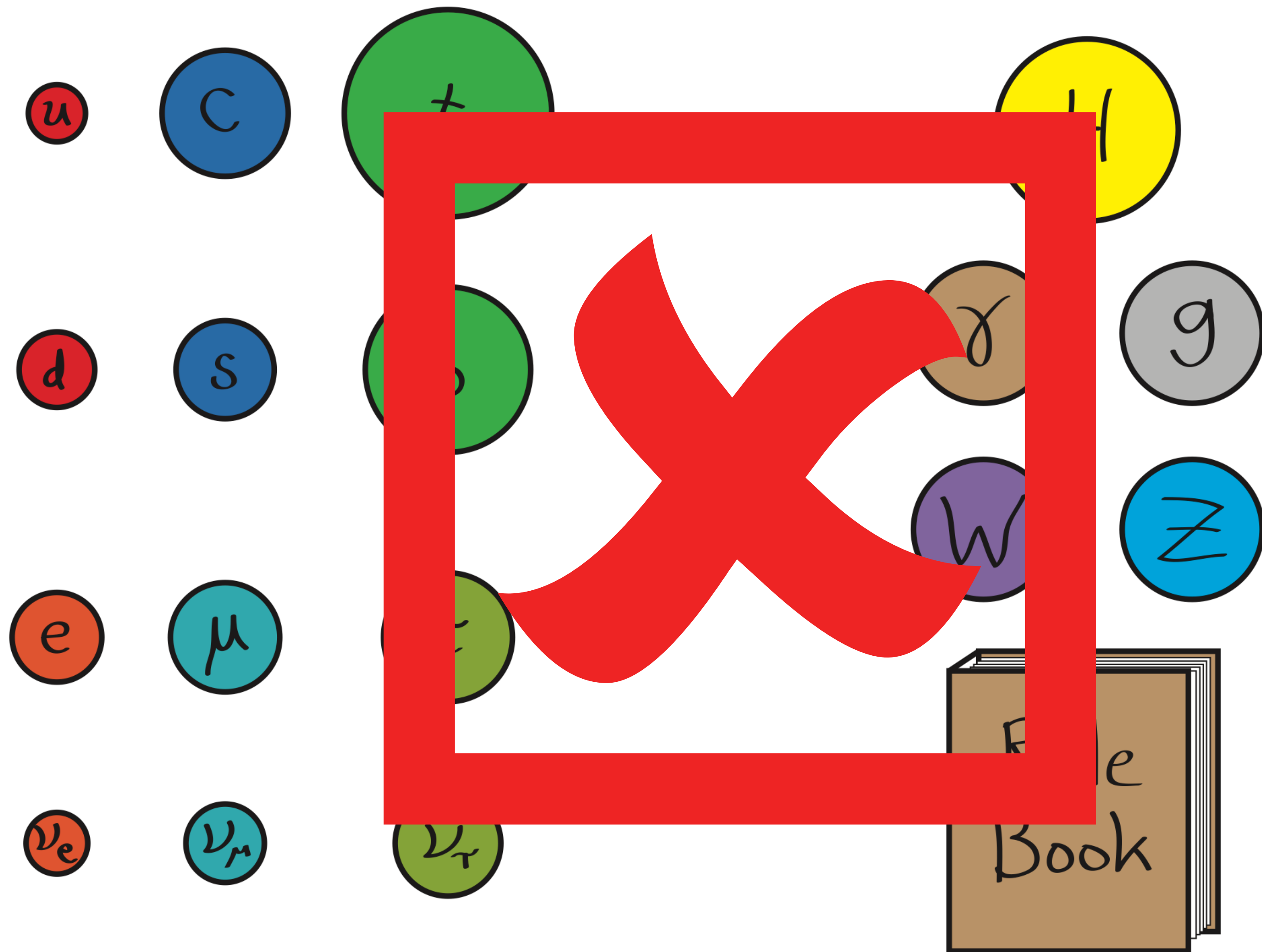


Standard Model



Standard Model
Describes
All Lab-based
Particle Physics
Experiments

Standard Model



Key Unresolved Questions



Dark Matter



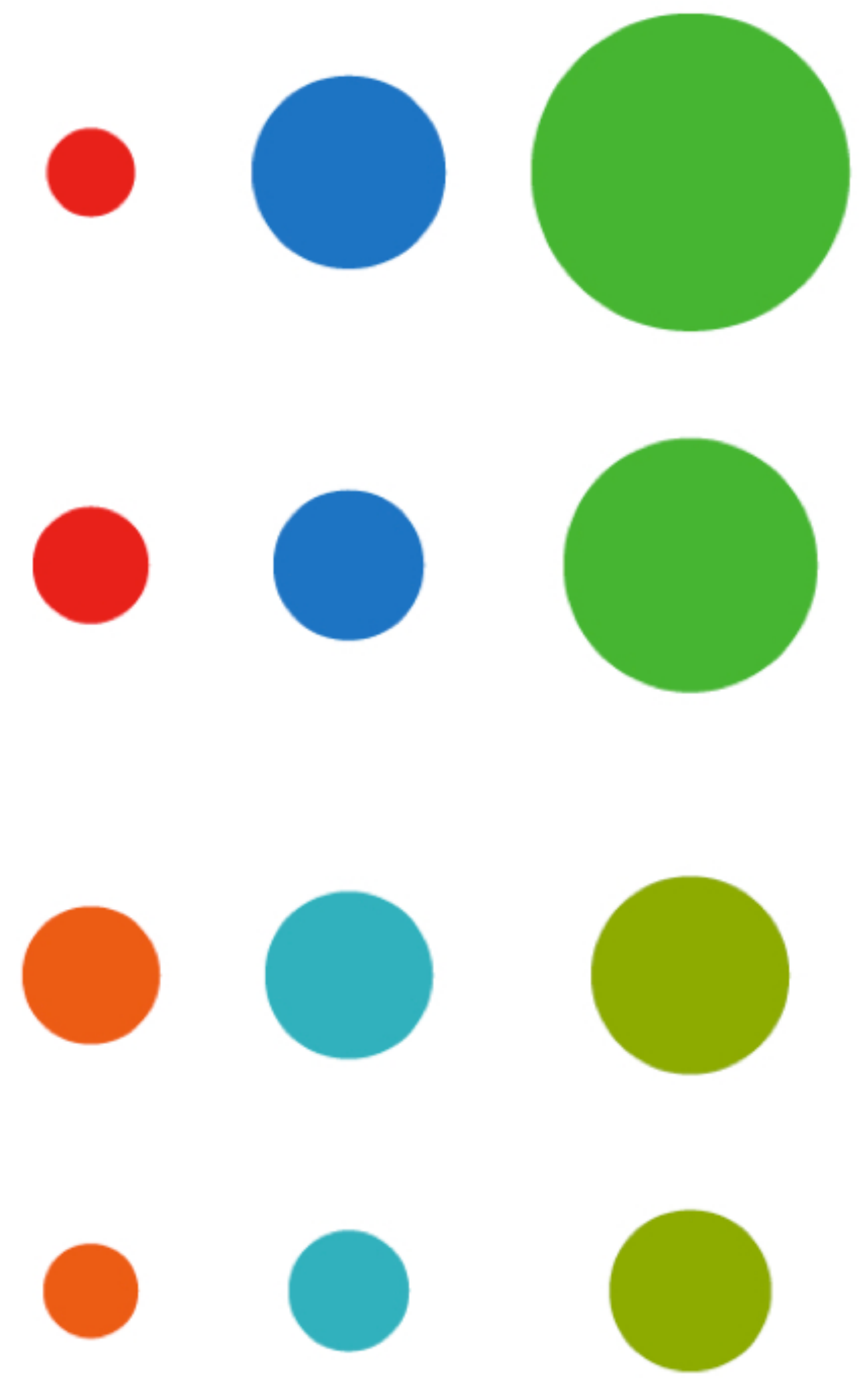
Matter-Anti Matter



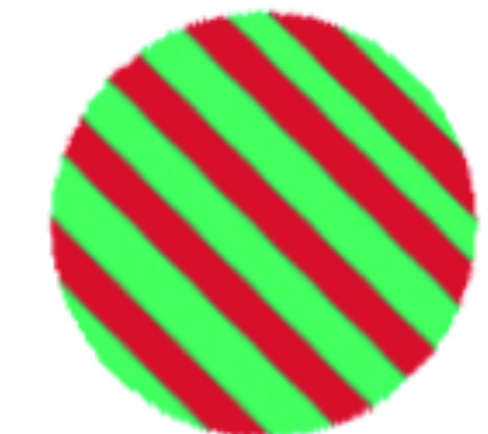
Hierarchy Problem

Possible Solution

Beyond
The
Standard
Model
(BSM)

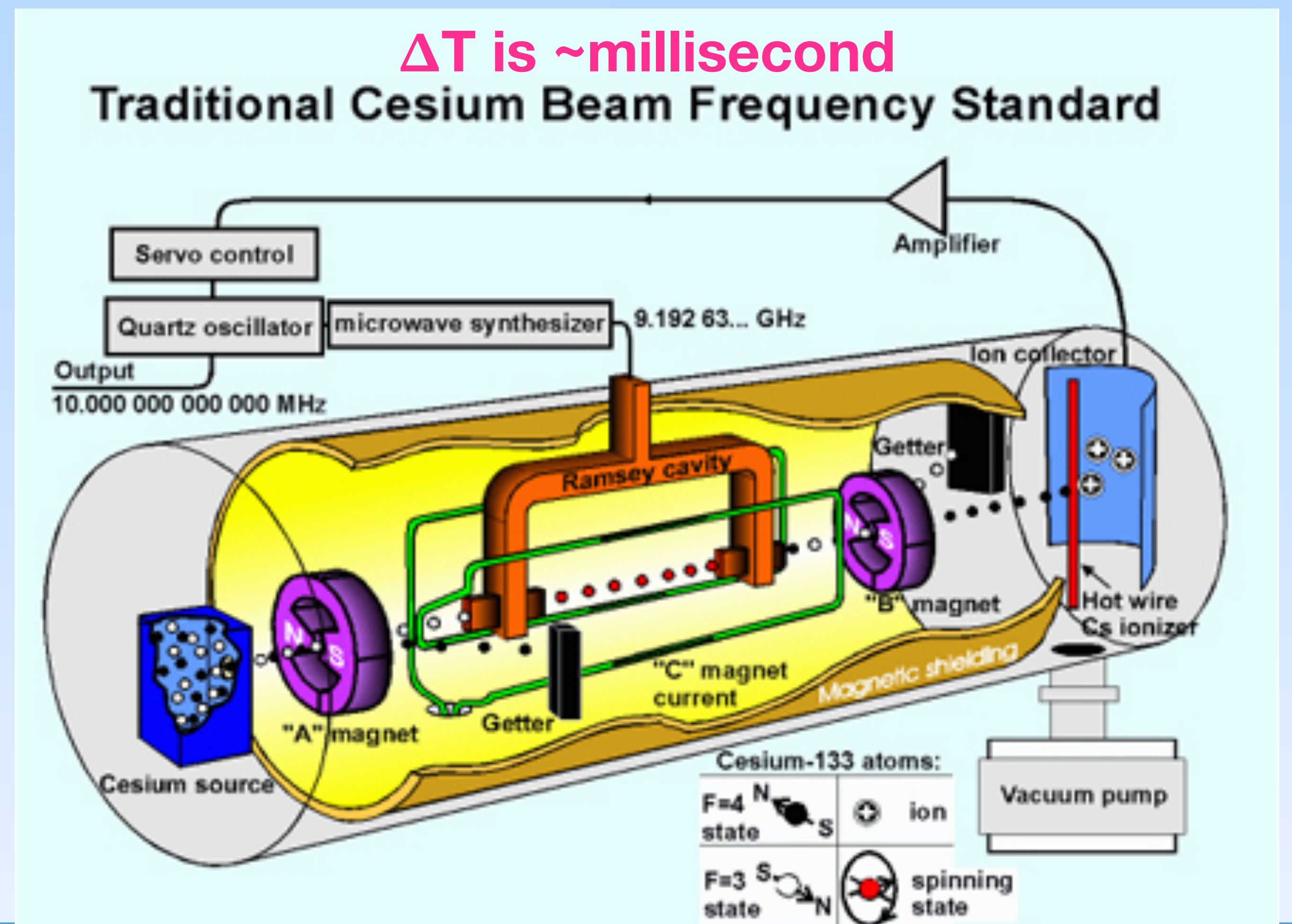
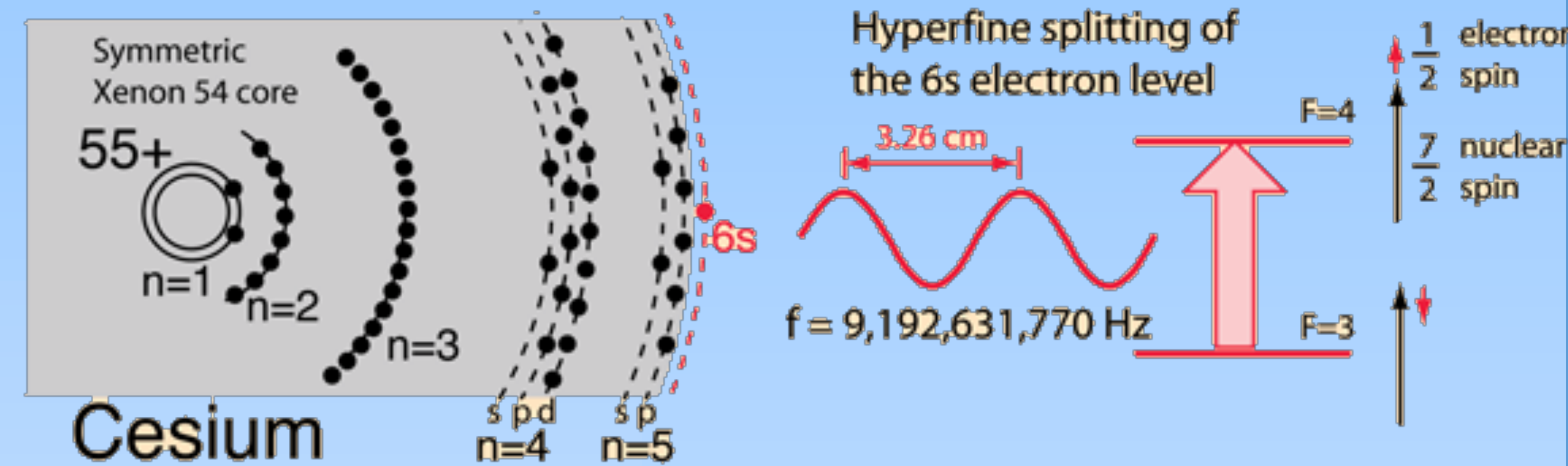


exotic
new
particles

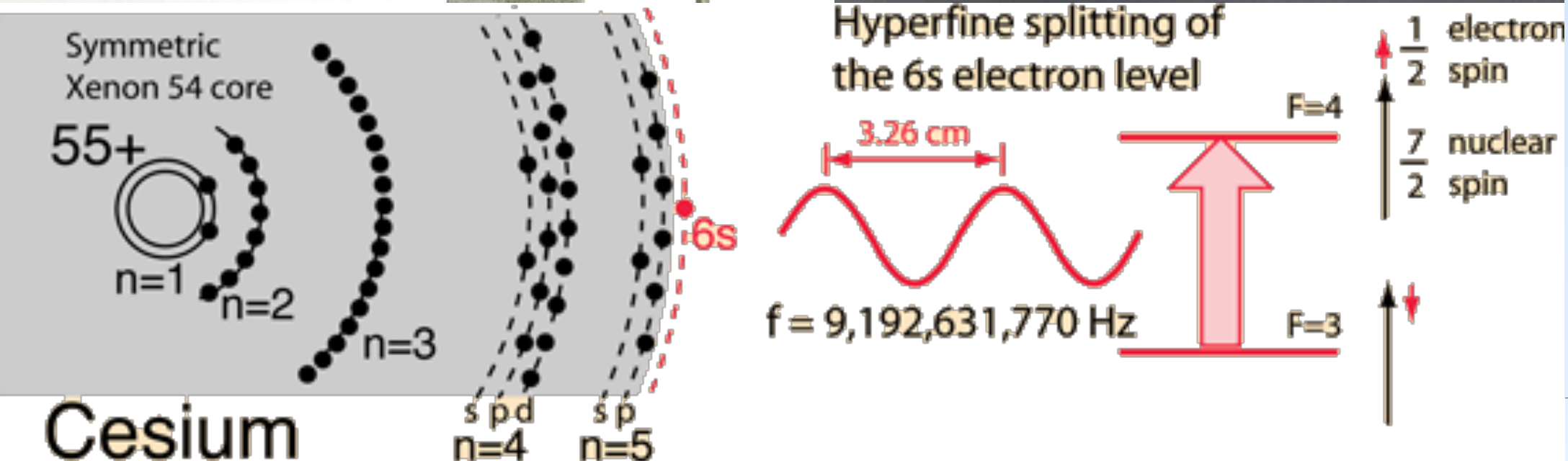
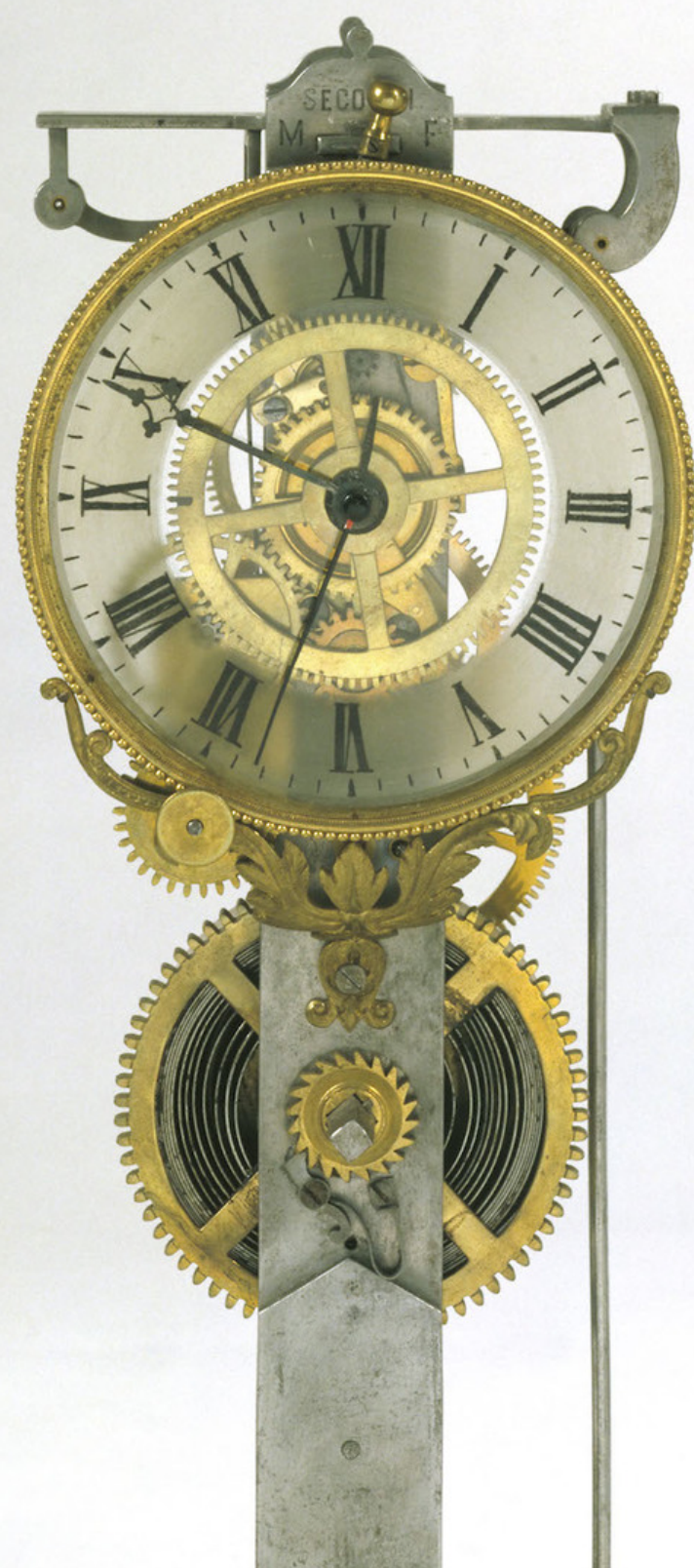


= X

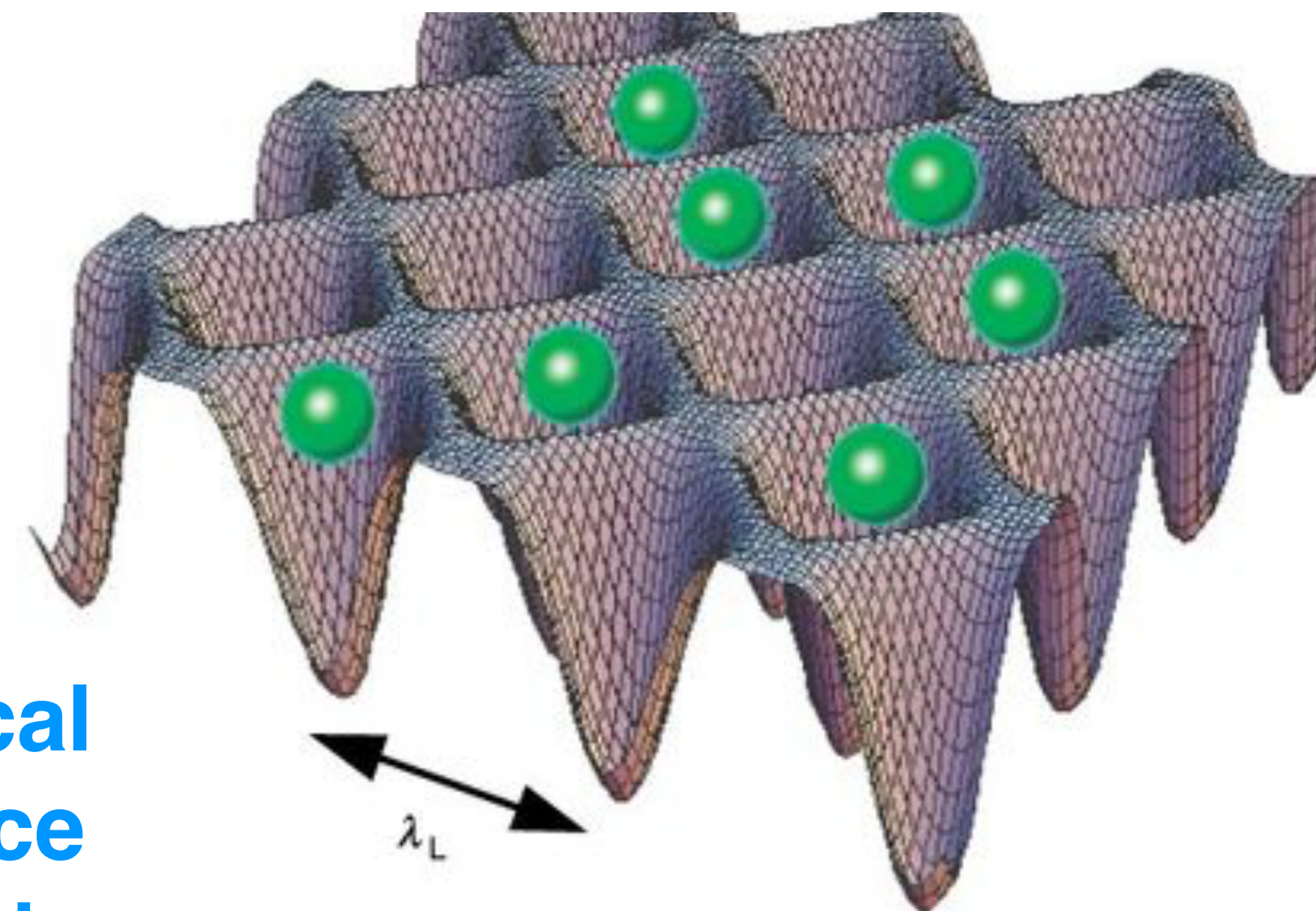
Detect Particle X with a Clock



Detect Particle X with a Clock



OPTICAL TWEEZERS (LATTICE)

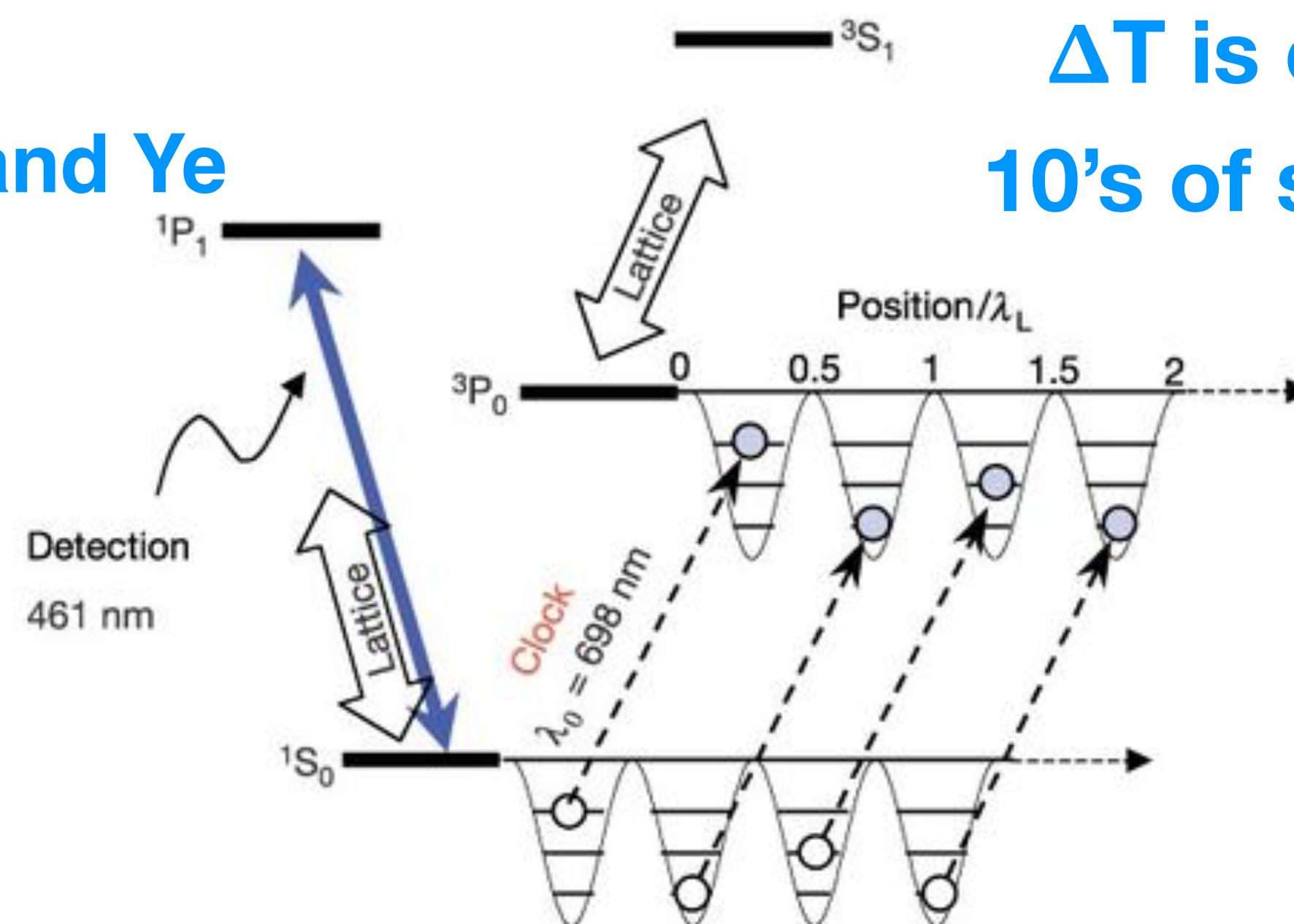


Ultracold Atoms
($\ll 1 \text{ mK}$)

Optical
Lattice
Clock

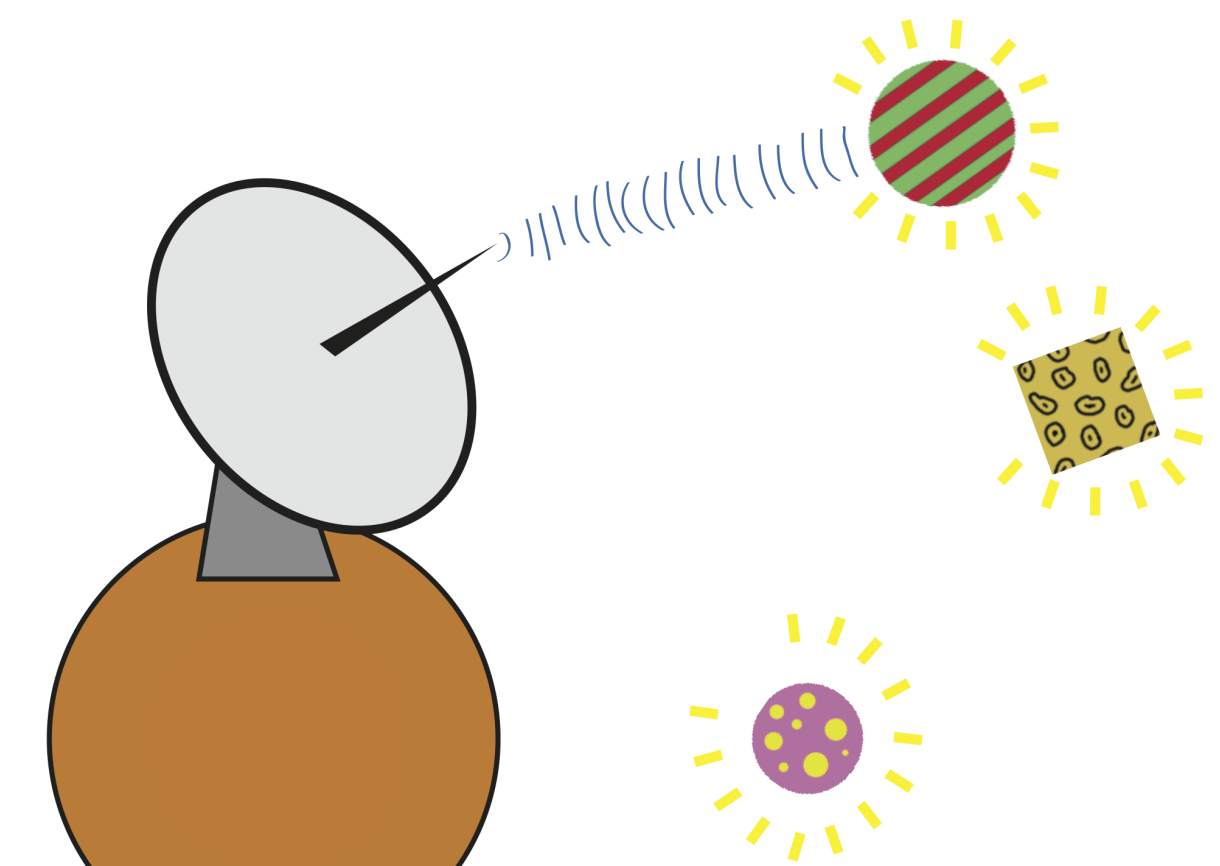
Katori and Ye

ΔT is can be
10's of seconds

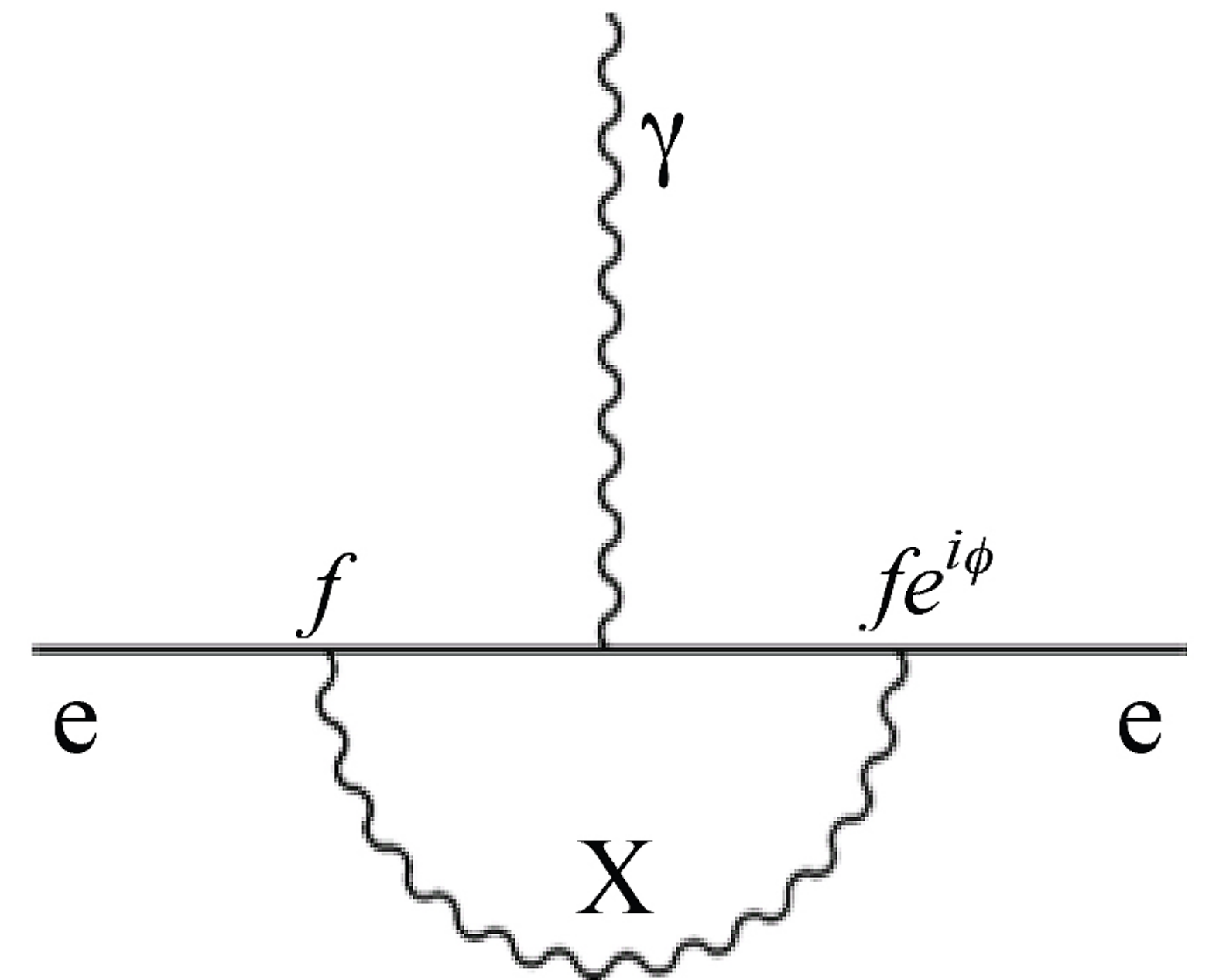


Amazing Fact:
any X particles
are right here, right now.

Already well known...g-2

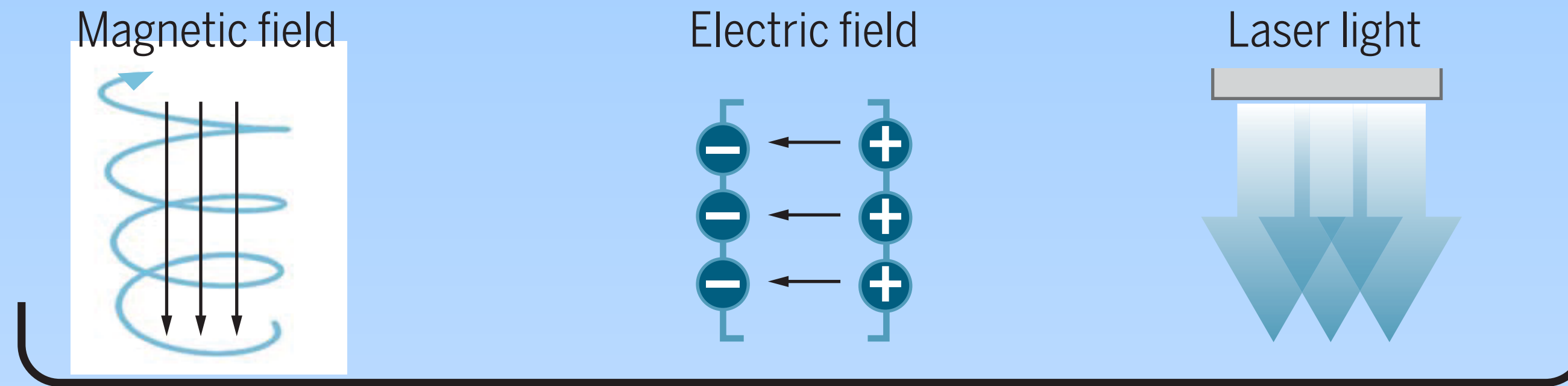


X particles

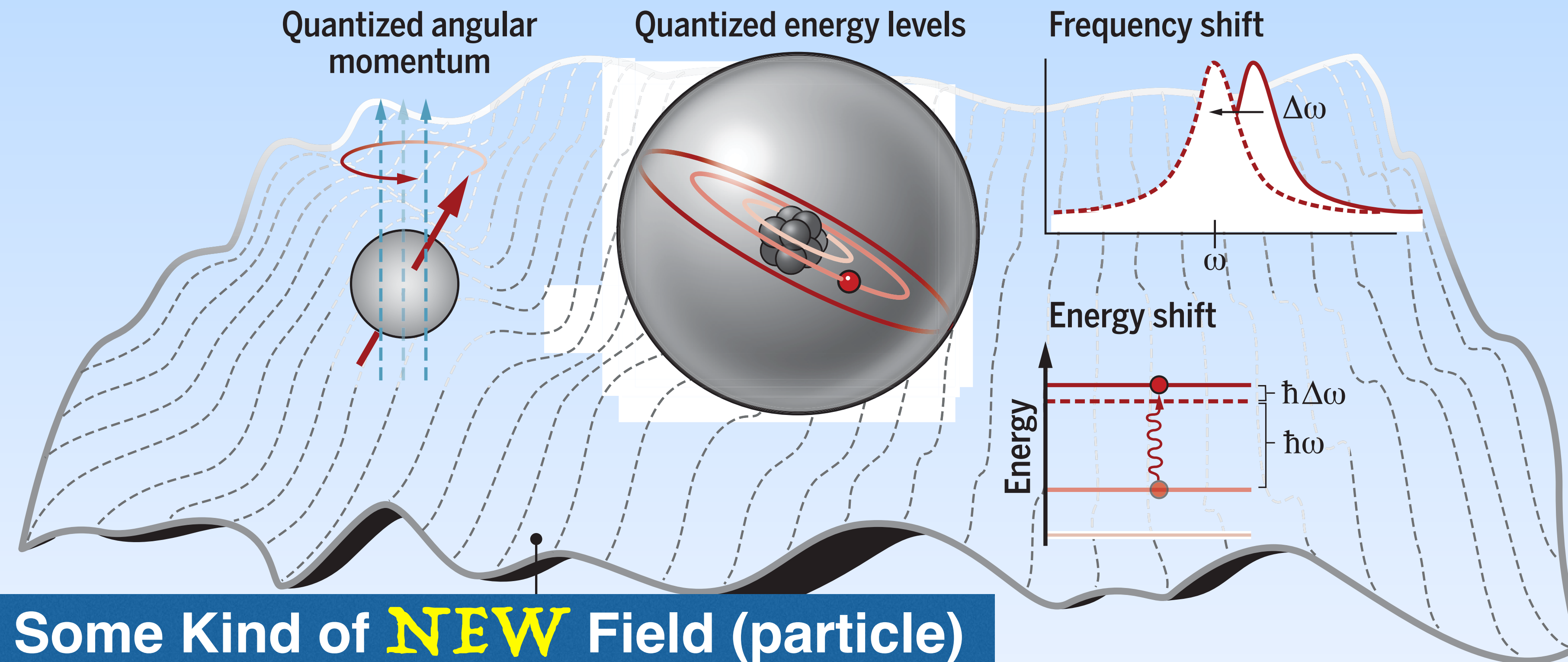


Clocks are Sensors of the NEW PARTICLES

Methods of probing



Measurement concepts



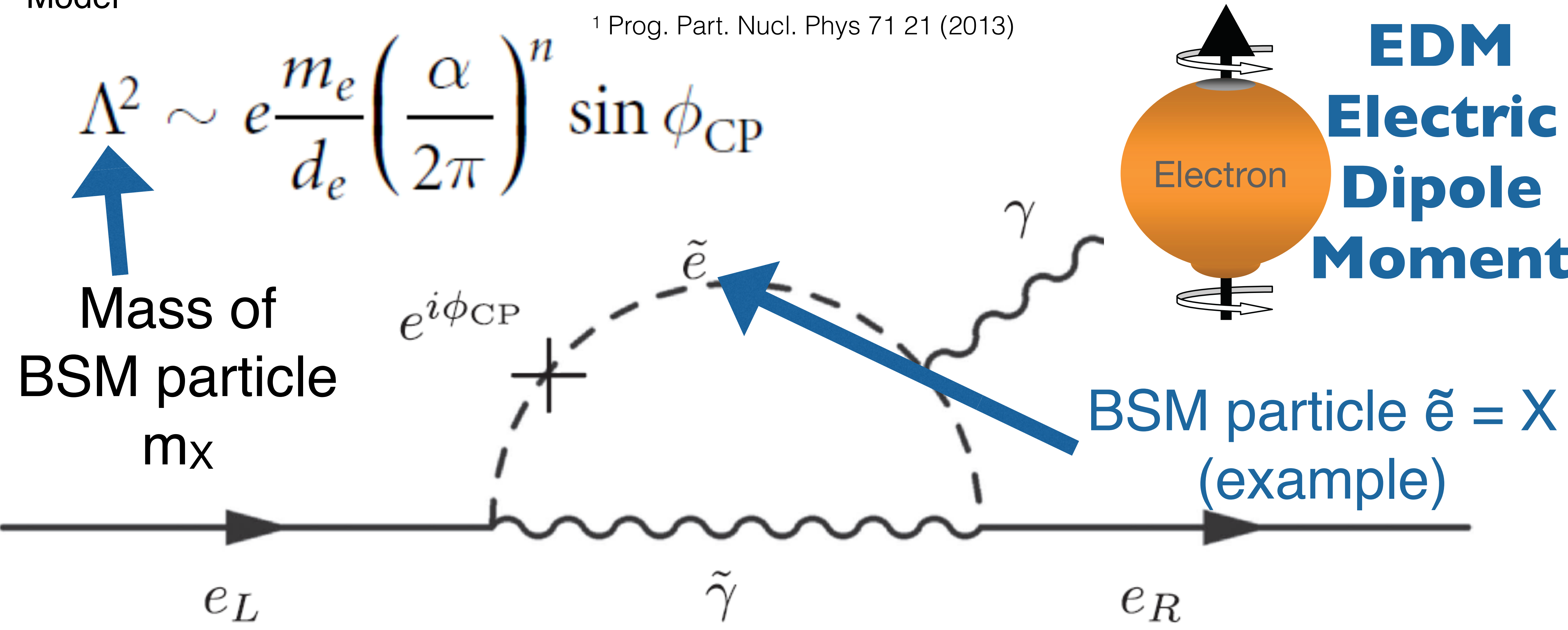
BSM =
Beyond
Standard
Model

In BSM theories, EDM (d_e) is created generically
The mass scale of BSM physics and d_e can be estimated via¹...

¹ Prog. Part. Nucl. Phys 71 21 (2013)

$$\Lambda^2 \sim e \frac{m_e}{d_e} \left(\frac{\alpha}{2\pi} \right)^n \sin \phi_{CP}$$

Mass of
BSM particle
 m_X



Look for the ELECTRIC dipole of the electron

Standard Model Predicts

EDMs $\times 10^6$

lower than current experiment

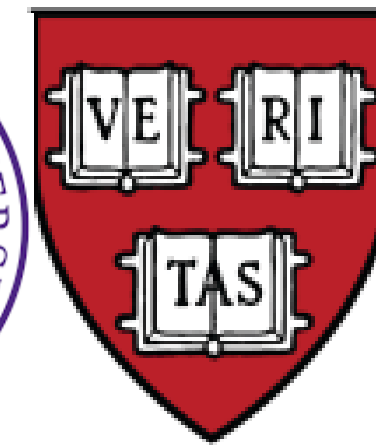
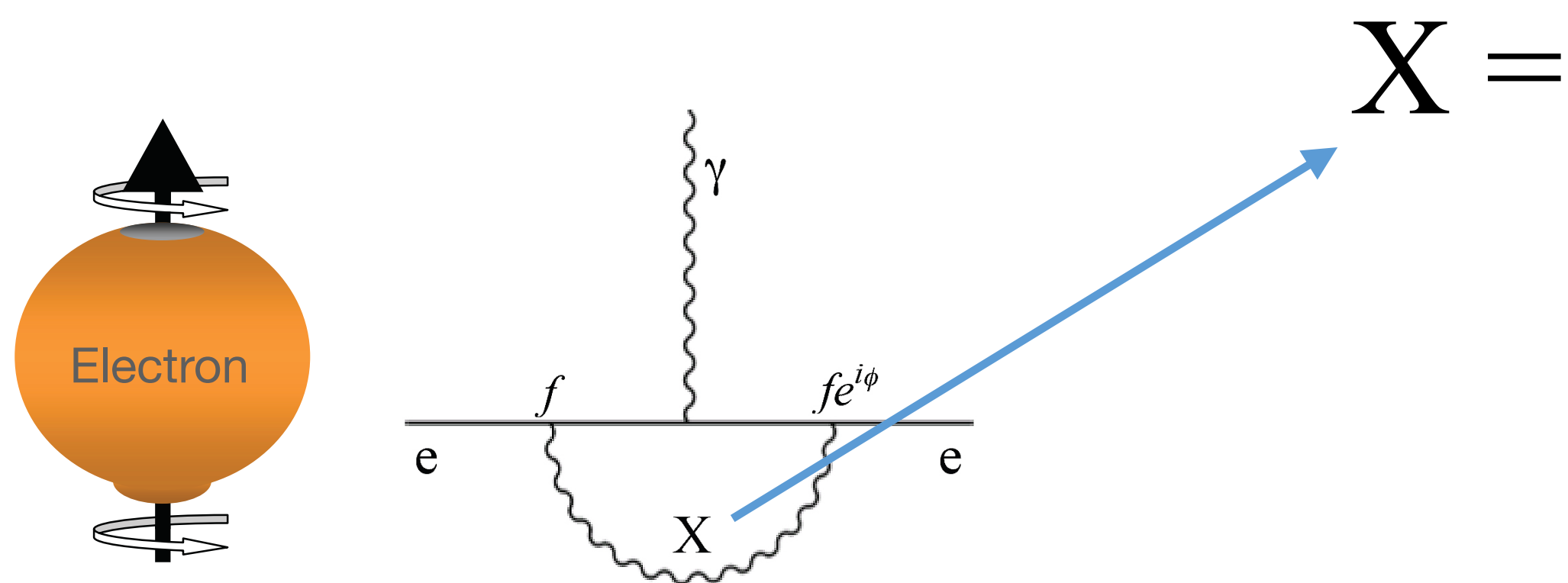
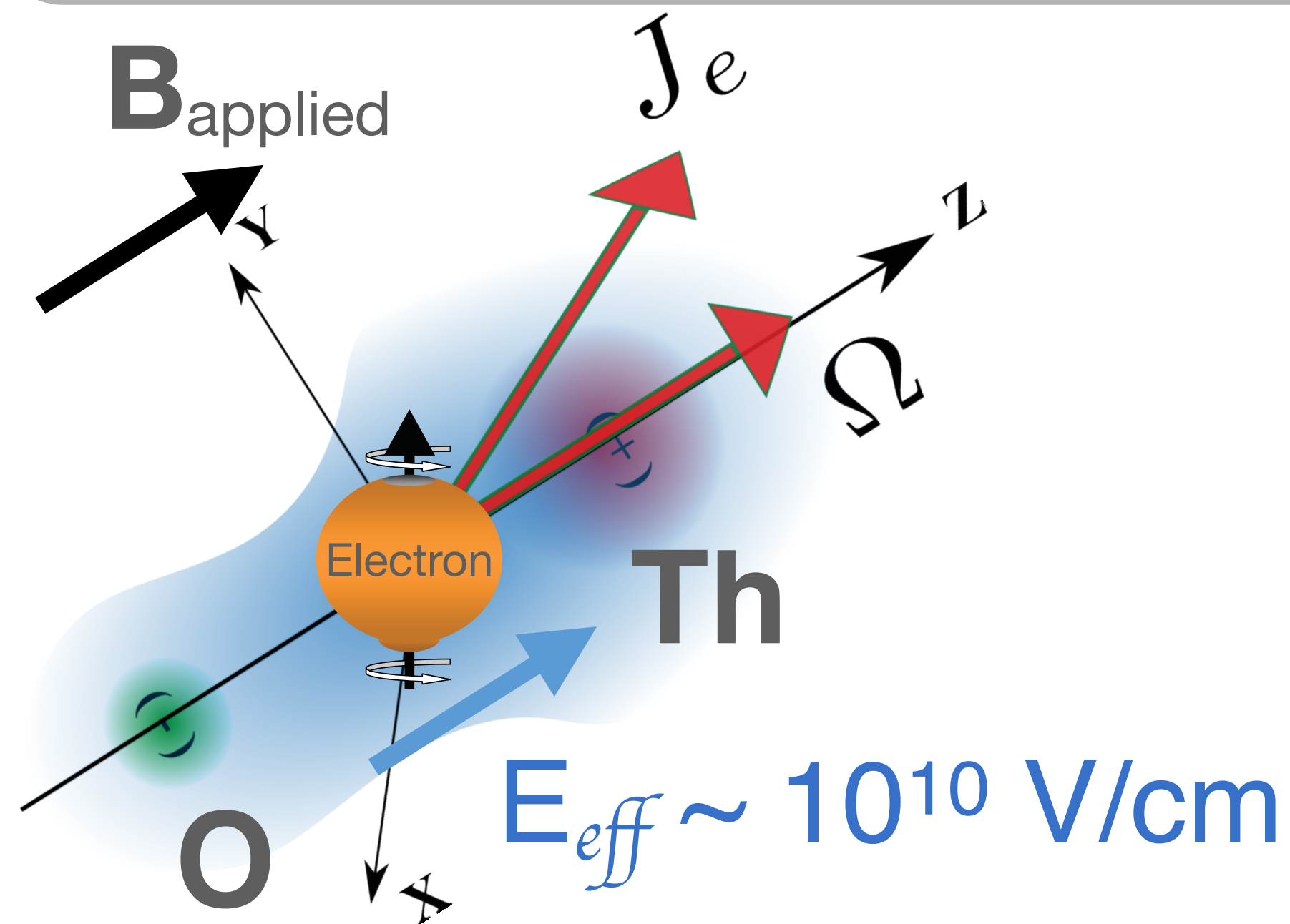
*not enough CP violation

EDM searches are

ZERO BACKGROUND

Searches for New Physics

EDM experiment using cold **ThO** molecules



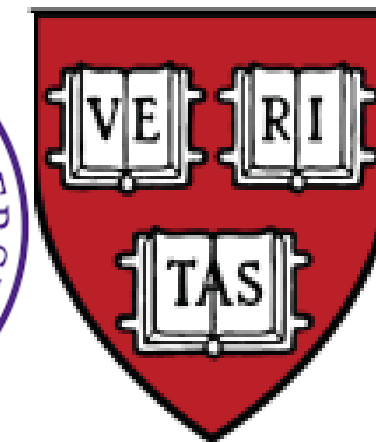
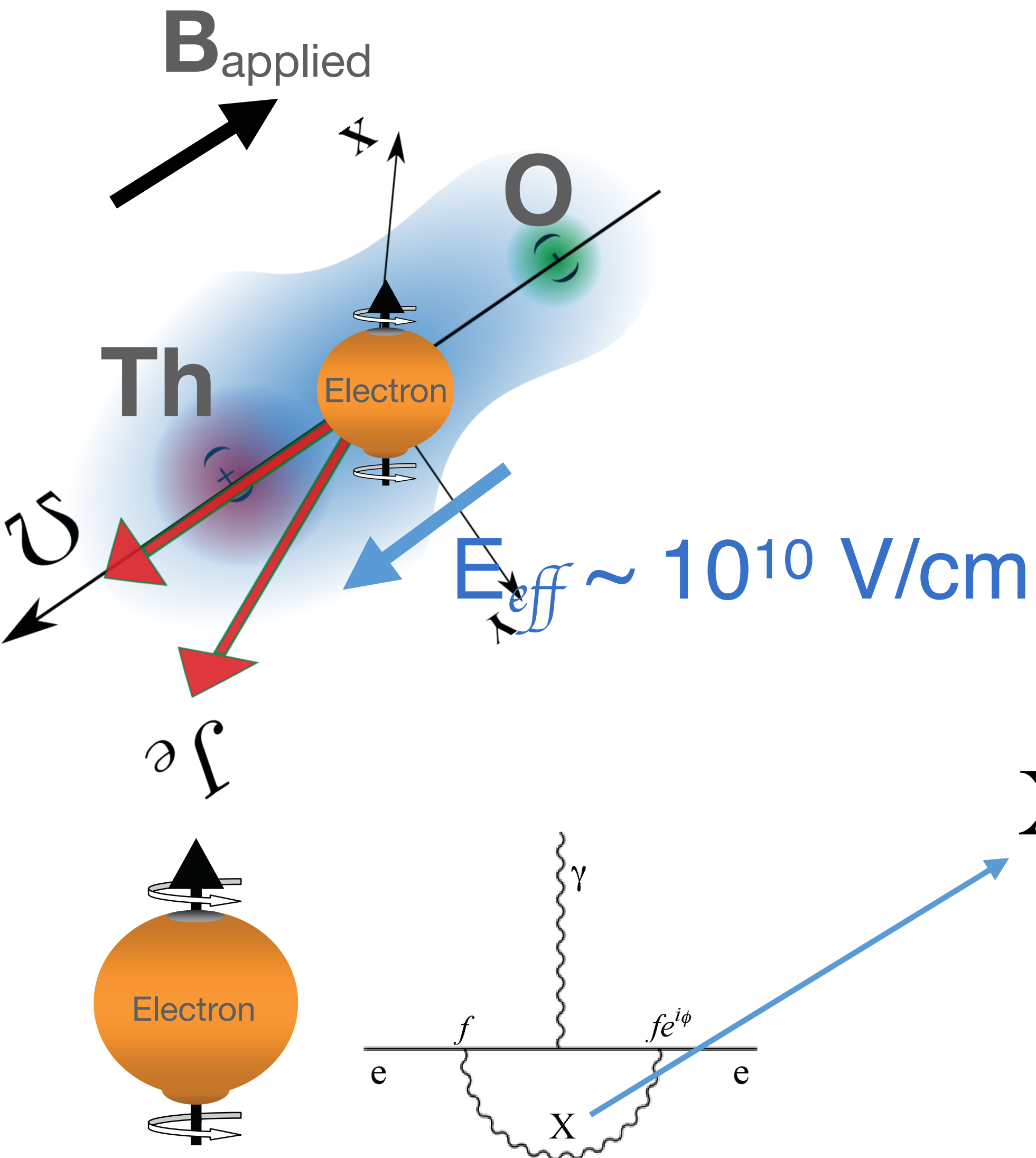
ACME

PIs: Doyle, DeMille and Gabrielse

NEED: Large Mass Nucleus
(For $\times 10^6$ enhancement
internal effective
electric field)
We use Thorium

WANT: Parity Doublet,
e.g. Omega doublet
We use Thorium Oxide

EDM experiment using cold **ThO** molecules



ACME

PIs: Doyle, DeMille and Gabrielse

NEED: Large Mass Nucleus
(For $\times 10^6$ enhancement
internal effective
electric field)
We use Thorium

$$X = \left(\begin{array}{c} \text{purple circle with dots} \\ \text{yellow square with dots} \\ \text{red and green striped circle} \\ \text{brown triangle with antlers} \end{array} \right)$$

WANT: Parity Doublet,
e.g. Omega doublet
We use Thorium Oxide

ACME II 2018

Beam Experiment 1 ms Coherence Time

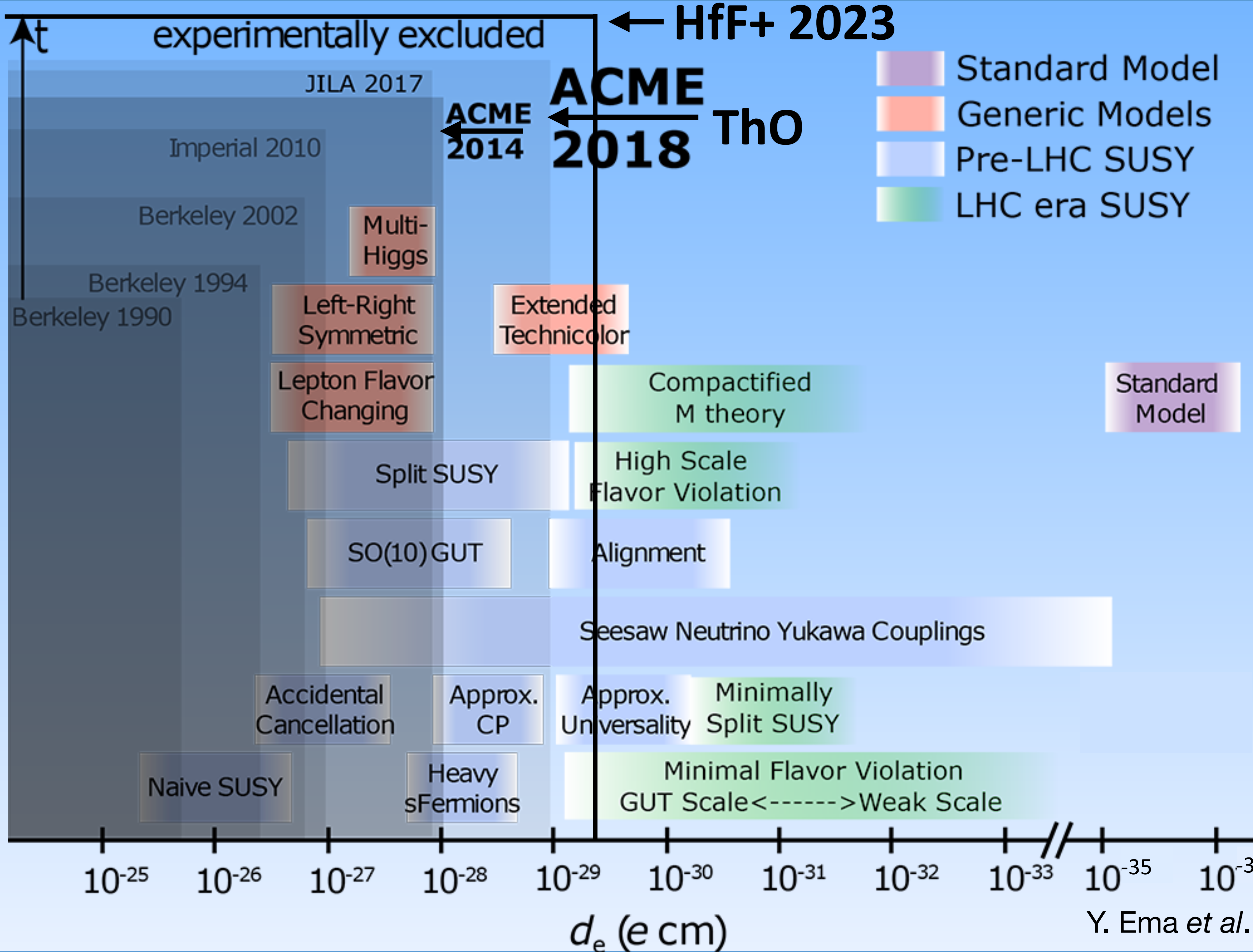
And a
radioactive
“tent” in other
building

CBGB

**Interaction
Region**

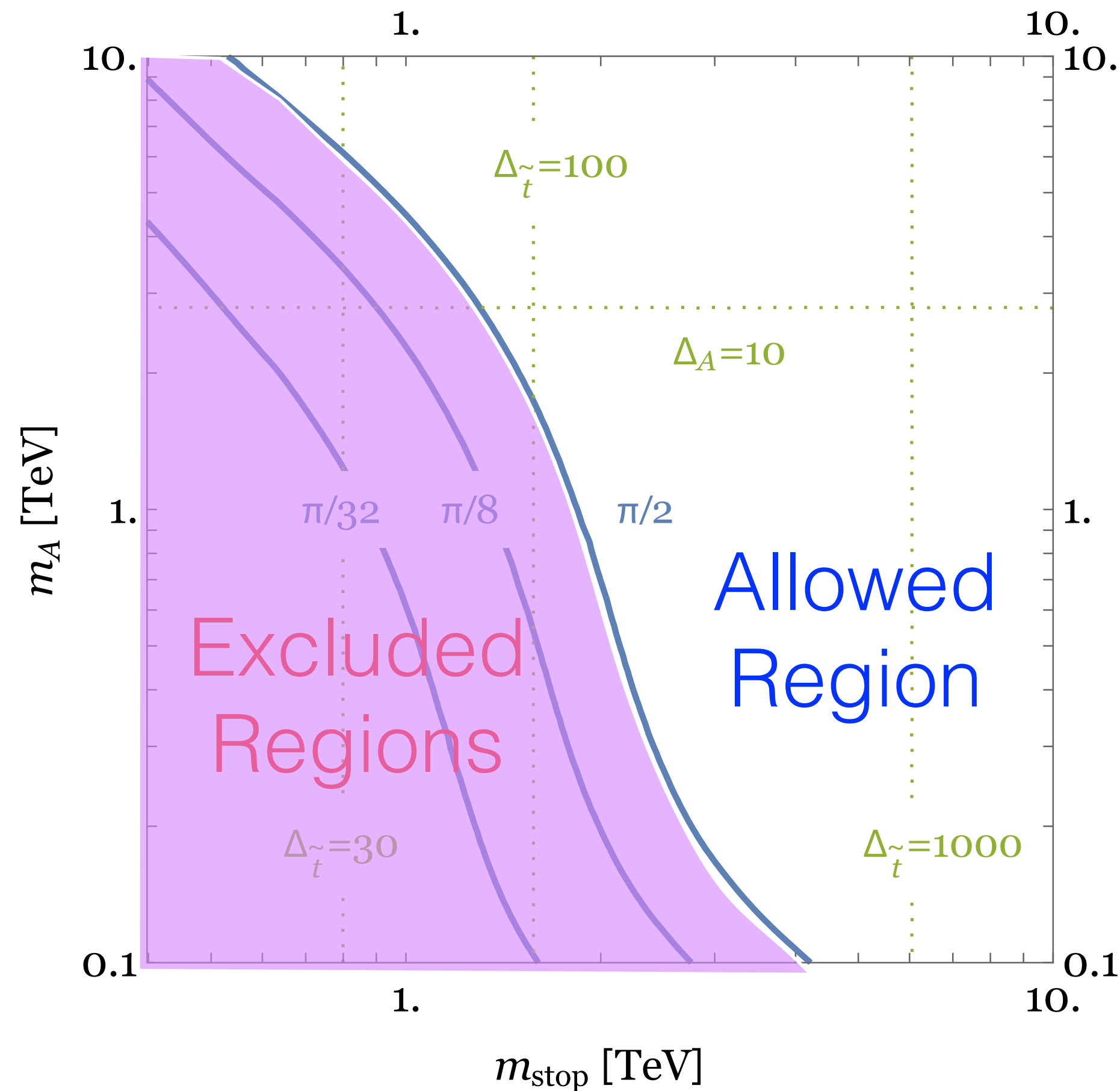
And a 5'x10'
table of optics in
another room

And compressors
pulsed laser
power supplies

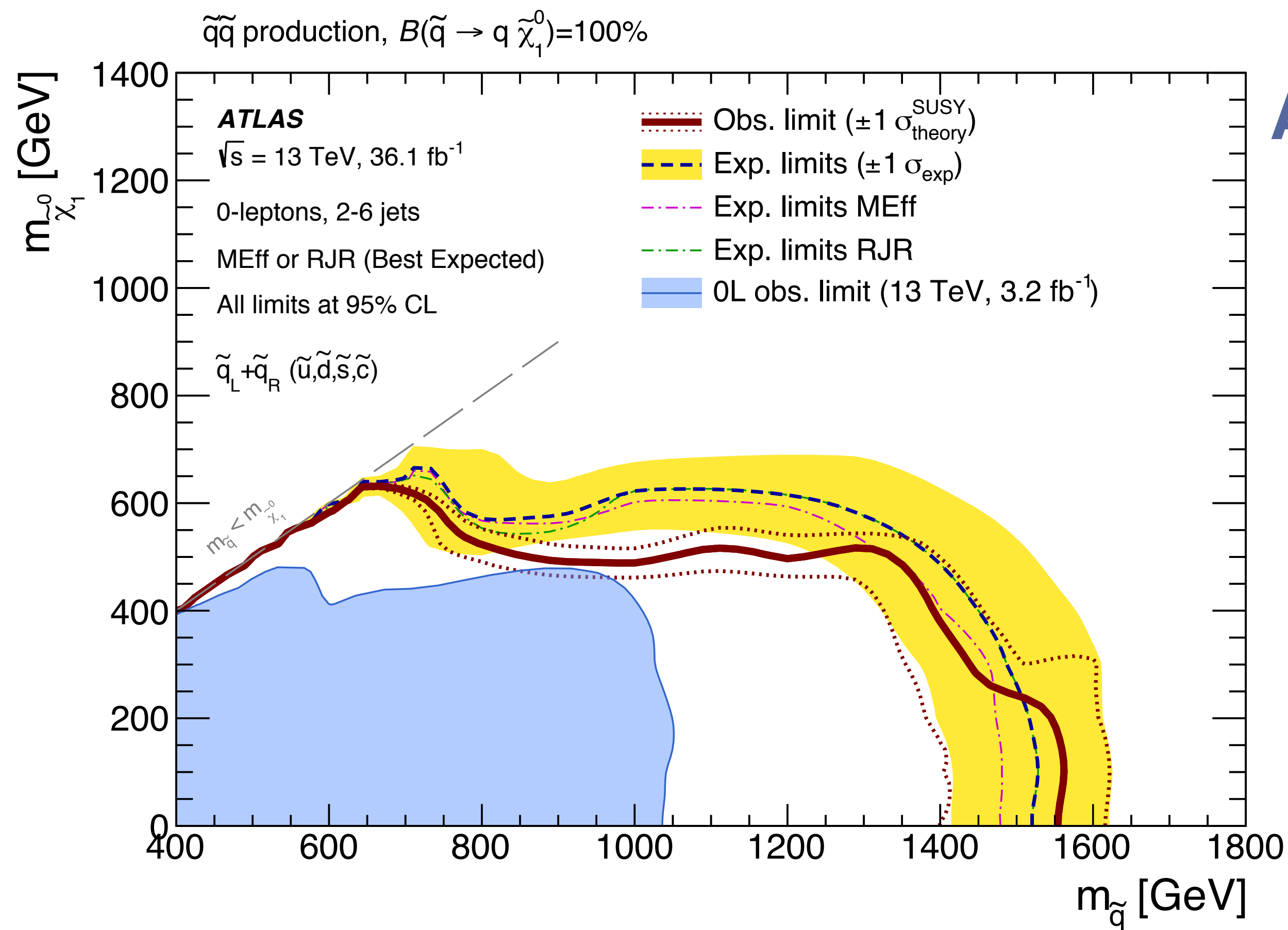


ACME II EDM Limit

$$\arg(A_t\mu)=\pi/2, \pi/8, \pi/32, \tan\beta = 10$$



LHC Limit



Two Loop
Electron EDM
Result
ACME 2

Interpreting
the
Electron
EDM
Constraint

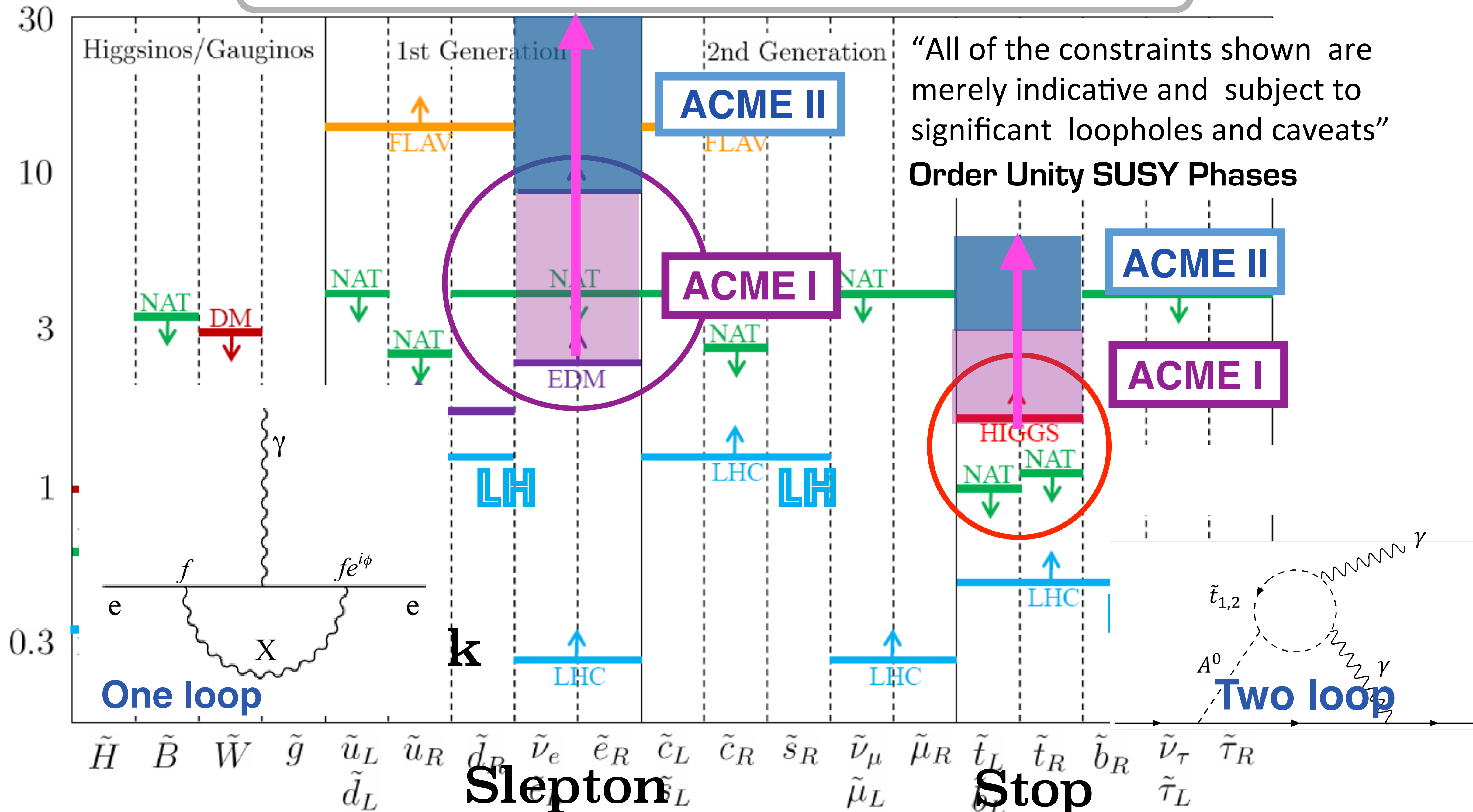
2018

Qianshu
Lu

LHC search in general only depends on particle mass, while eEDM depends on CPV phase.

ACME II result SUSY LIMITS

Superpartner Mass (TeV)



↑
t

experimentally excluded

2014-2017

ACME

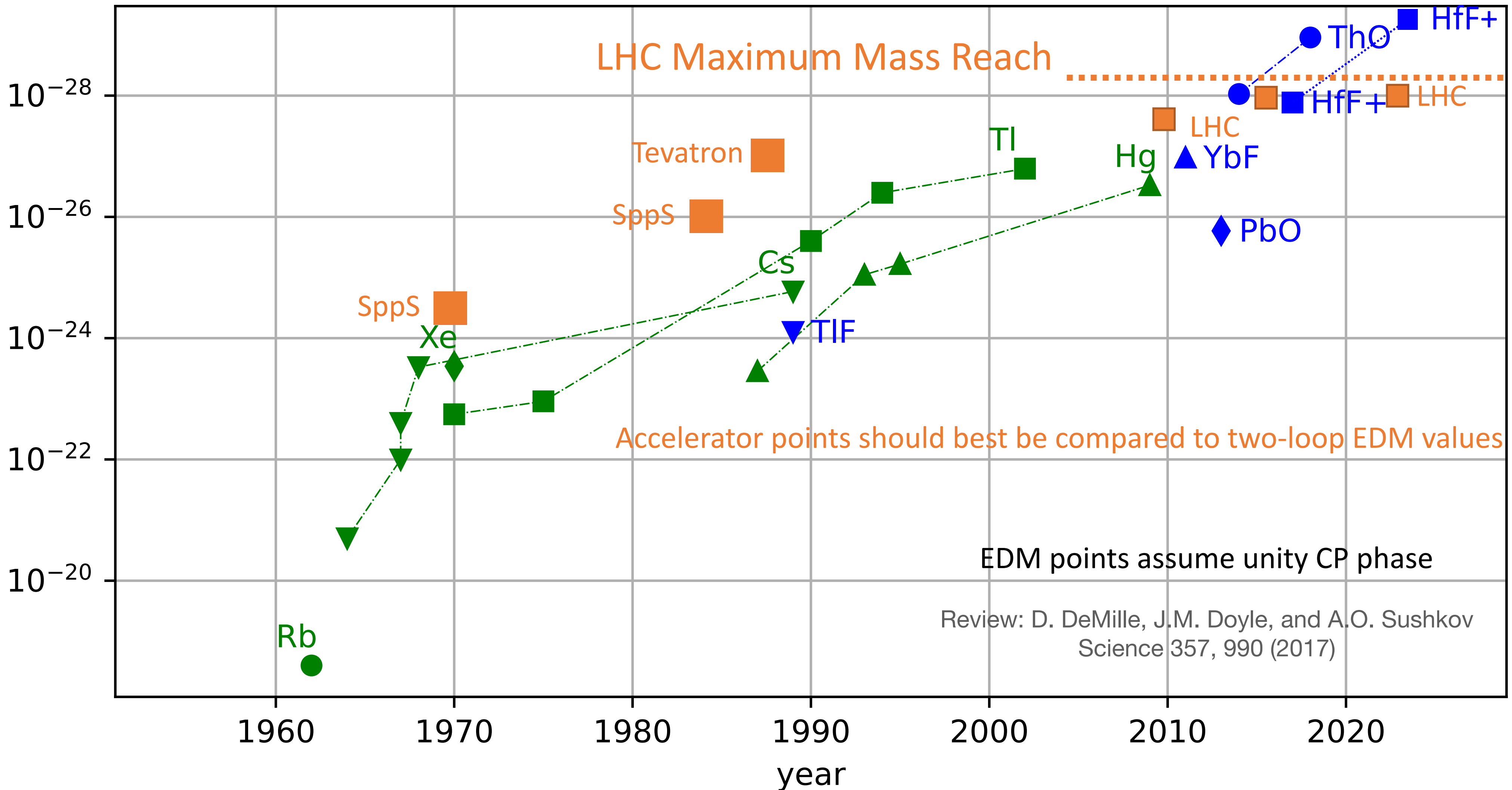
Standard Model

● ThO ACME 2018

■ HfF+ Jila 2023

TeV

Limit
 $d_e(\text{ecm})$



30 3

3 0.3

0.3 0.03

GeV

30 3

3 0.3

0.3 0.03

One Loop Two Loop



ACME III 2023

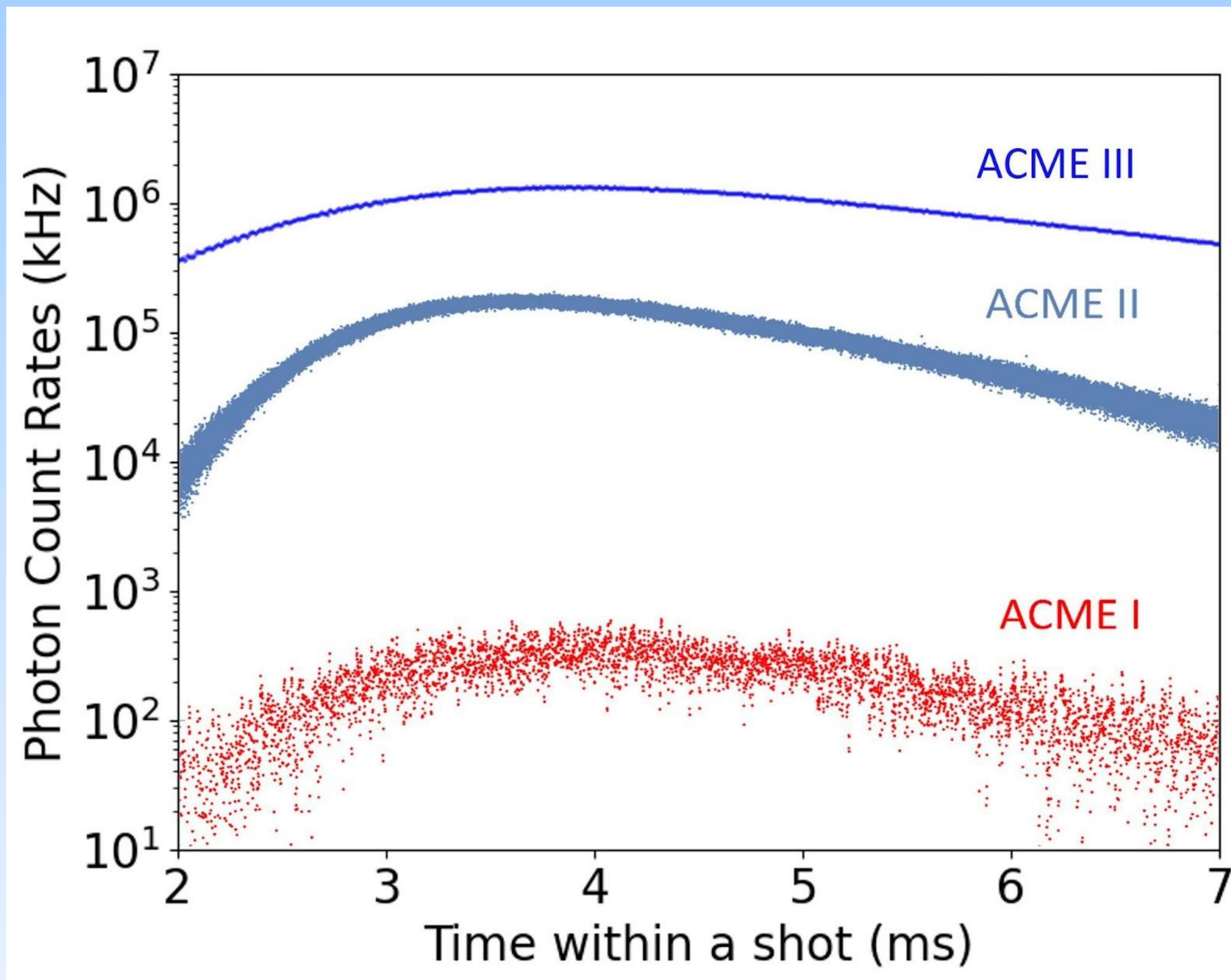
**Interaction
Region**

CBGB

**Molecular
Lens**

5 ms Coherence Time

ACME III signal compared with ACME I and II



Previously Planned Signal Gain is Achieved

Demonstrated Improvement	Signal Gain	T Gain	EDM sensitivity Gain
Longer Precession Time ¹	0.3	5	2.7
Electrostatic Lens ²	12	1	3.5
SiPM Detector ^{3,4}	2.7	1	1.6
Improved collection optics	1.7	1	1.3
Eliminated Timing Jitter Noise ⁵	1	1	1.7
Load-lock target change	1.4	1	1.2
Total	23	5	39
ACME II Daily 1 σ Statistical Sensitivity (e cm)			$\sim 1 \times 10^{-29}$
ACME III Daily 1σ Sensitivity (e cm)			$\sim 3 \times 10^{-31}$

x30 EDM Sensitivity
Based on
Current Running
Conditions in ACME III

Photoelectron counts per ablation shot (50Hz)	Photoelectron counts averaged over 1 second
ACME II $\sim 2.4 \times 10^5$	$\sim 1.2 \times 10^7$
ACME III $\sim 6 \times 10^6$	$\sim 3 \times 10^8$

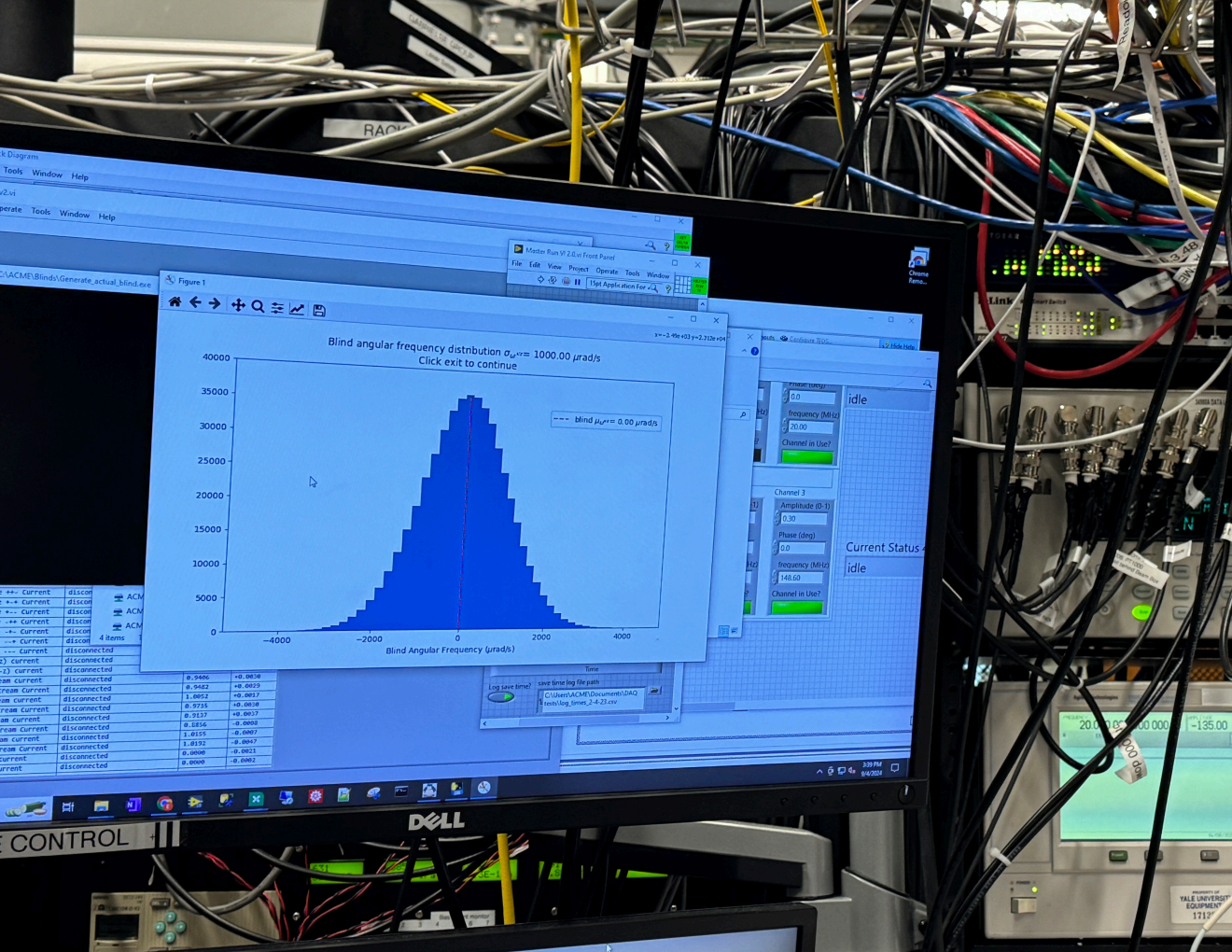
1. D. G. Ang *et al* 2022 *Phys. Rev. A* **106**, 022808

2. X Wu *et al* 2020 *New J. Phys.* **22** 023013

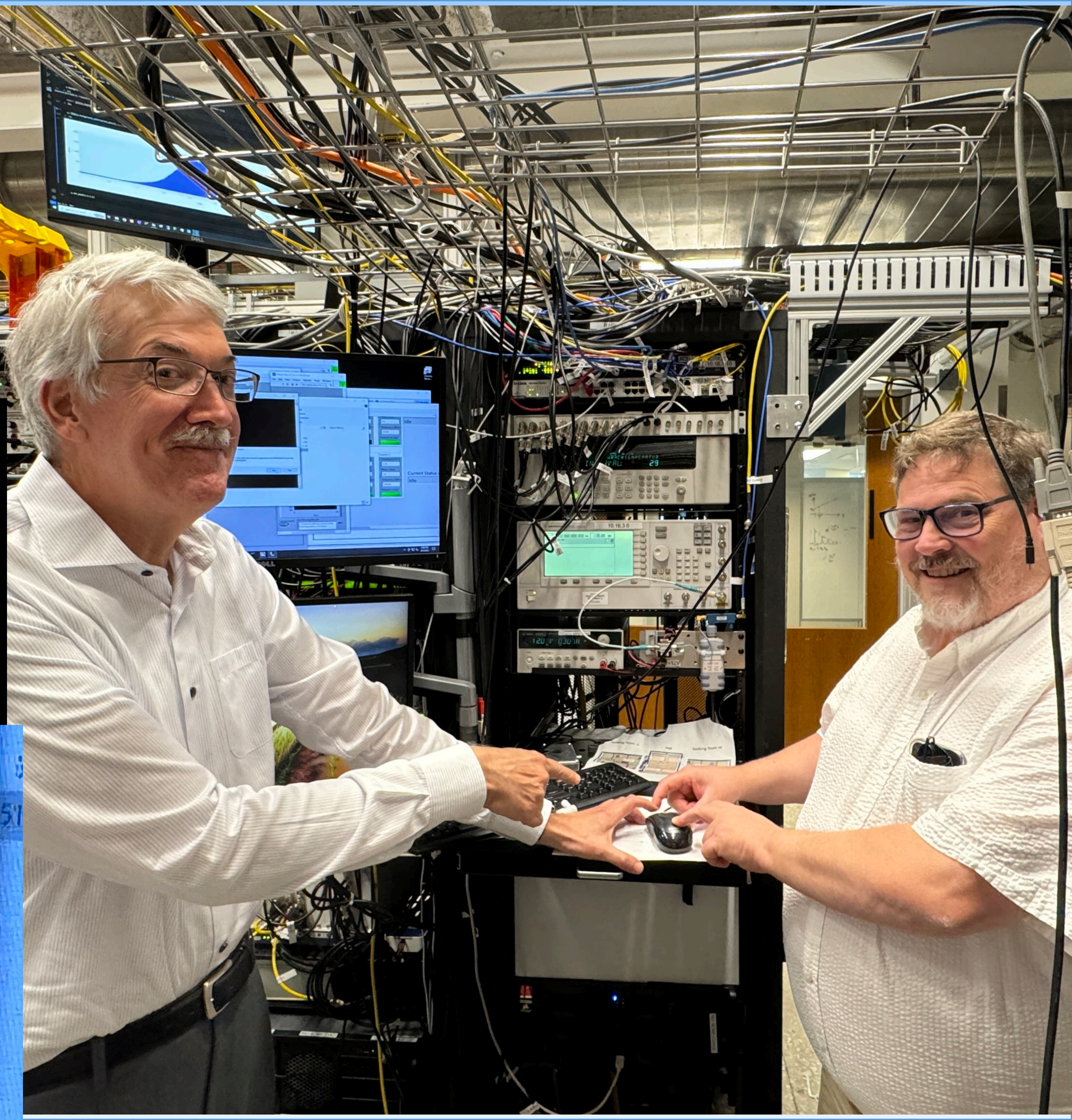
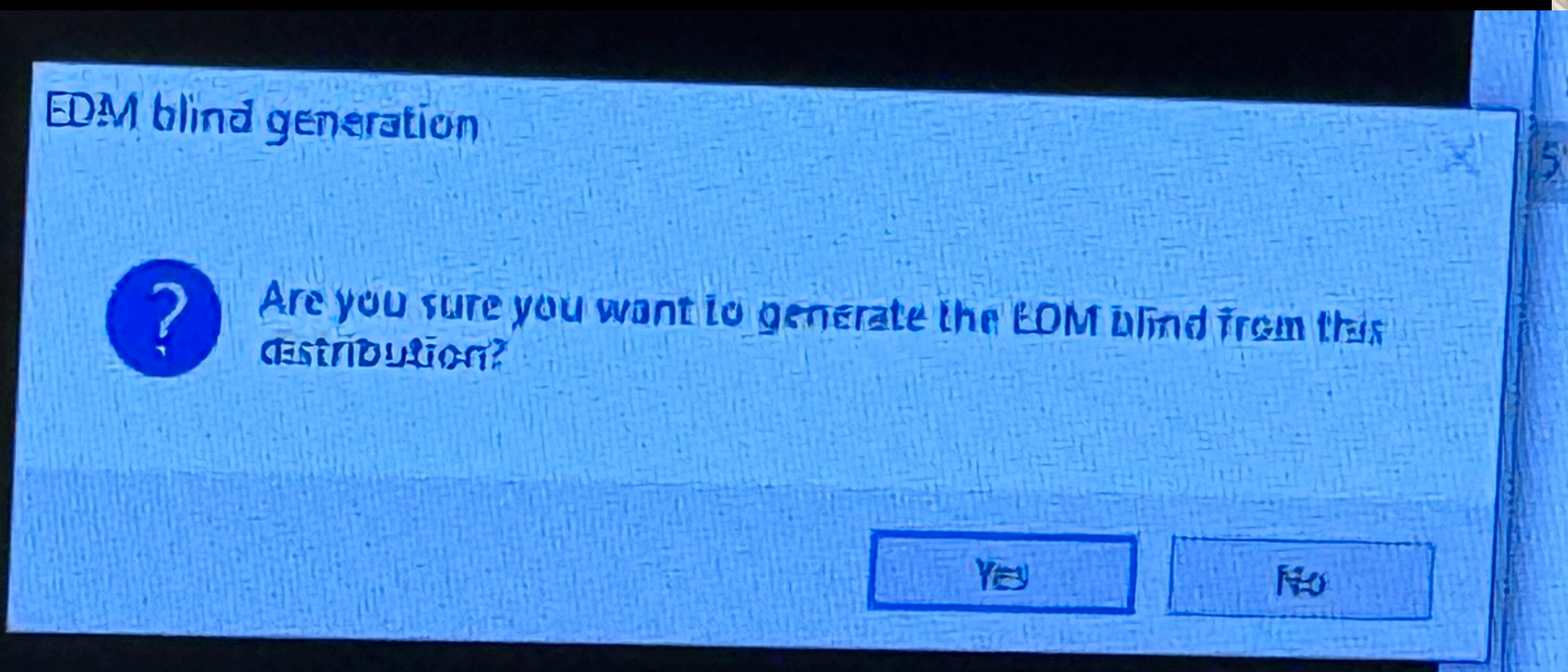
3. T Masuda *et al* 2021 *Optics Express*, **29**(13), 16914-16926

4. A. Hiramoto *et al* 2023 *Nucl. Instrum. Methods Phys. Res., Sect. A*, **1045**, 167513

5. C D Panda *et al* 2019 *J. Phys. B: At. Mol. Opt. Phys.* **52** 235003



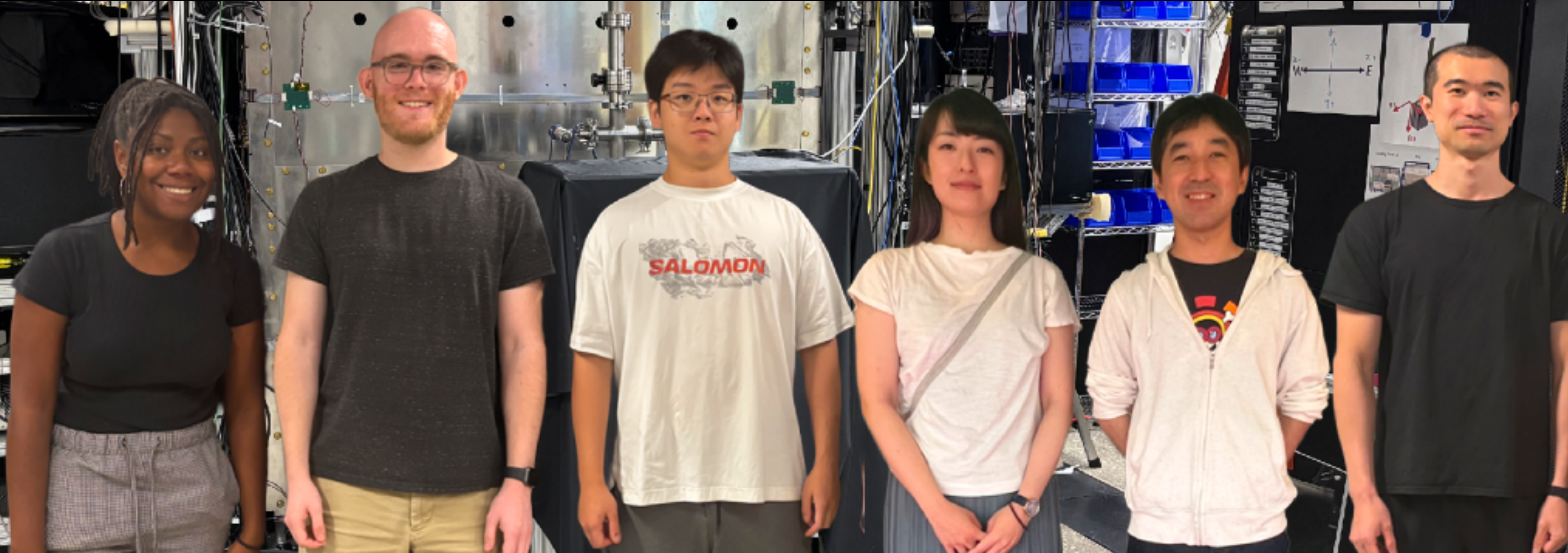
Blind was set one year ago
We are taking EDM data
First systematic checks underway



ACME III
“On The Ground” Team
Now

Ayami Hiramoto
Maya Watts
Collin Driver

Peiran Hu
Zhen Han
Takahiko Masuda



PIs
DeMille, Doyle, Fan, Gabrielse

Collaborators
Hutzler, Panda, Uetake, Wu, Yoshimura

What is the future for EDM Searches?

Why do an EDM experiment now?

CP- ν = CP violation

No sign of BSM physics anywhere but...

- Matter-antimatter asymmetry still unexplained
- Microscopic nature of Dark Matter still unknown
- Hierarch Problem did not go away

How to improve?

- Increase NUMBER OF MOLECULES
- Increase COHERENCE TIME
- Increase inherent SENSITIVITY to CP- ν

$$\delta d_e = \frac{1}{2} \frac{\hbar}{\beta \tau \sqrt{\dot{N} T}}$$

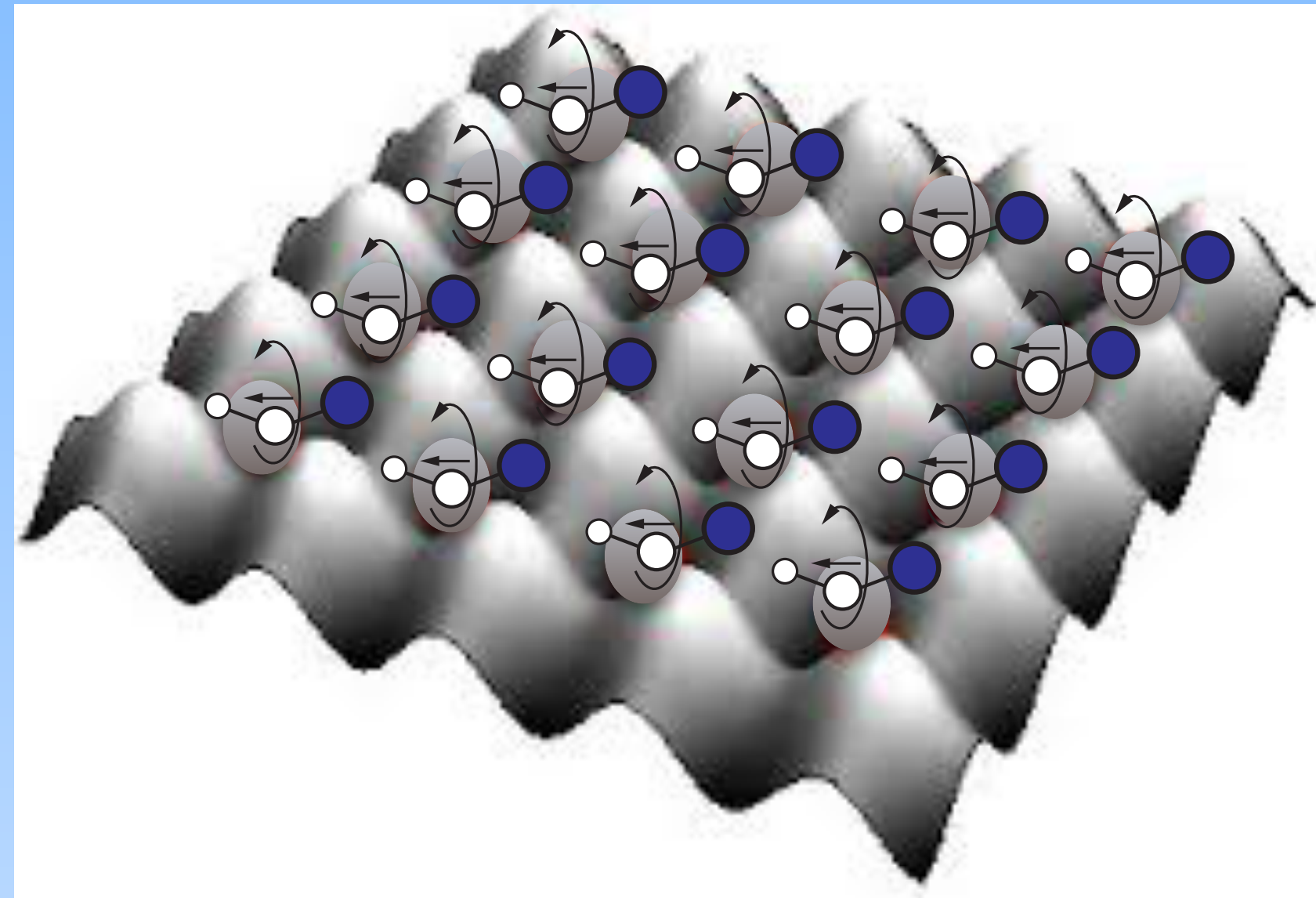
Sensitivity To CP- ν Physics

Coherence Time

(Photon) Counting Rate

Integration time

Increased complexity and increased COHERENCE TIME



Optical Trapping - Tweezers or Ion Trap

ACME III

5 ms



10 s

New Experiments

CP-v Pathfinder - CaOH

(EDM and Schiff Moment Pathfinder experiment)

Doyle/Hutzler
Harvard/Caltech

Poly EDM -SrOH

(Electron EDM)

Doyle/Hutzler/Steimle/Vutha
Harvard/Caltech/Toronto

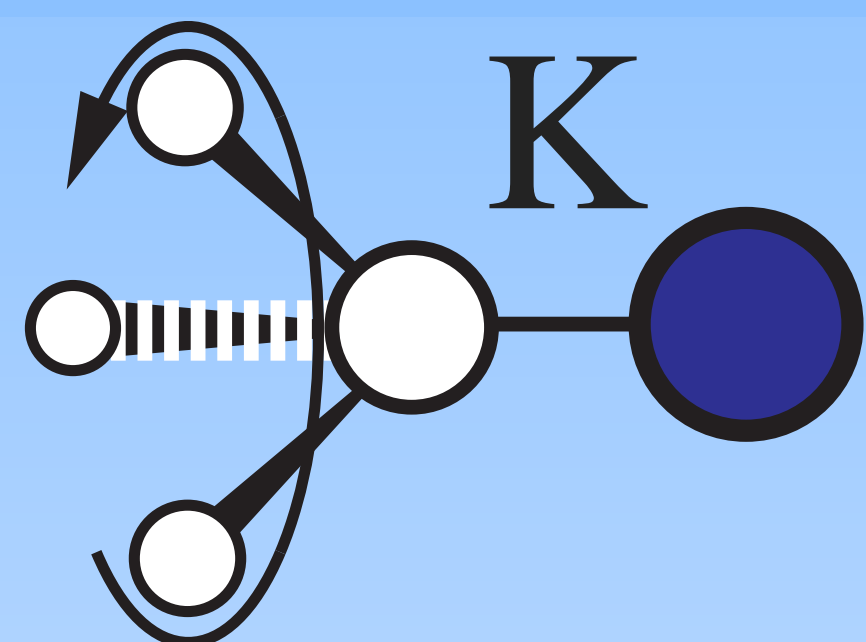
RaX - RaOH, RaF, RaOCH₃

(Nuclear Schiff Moment)

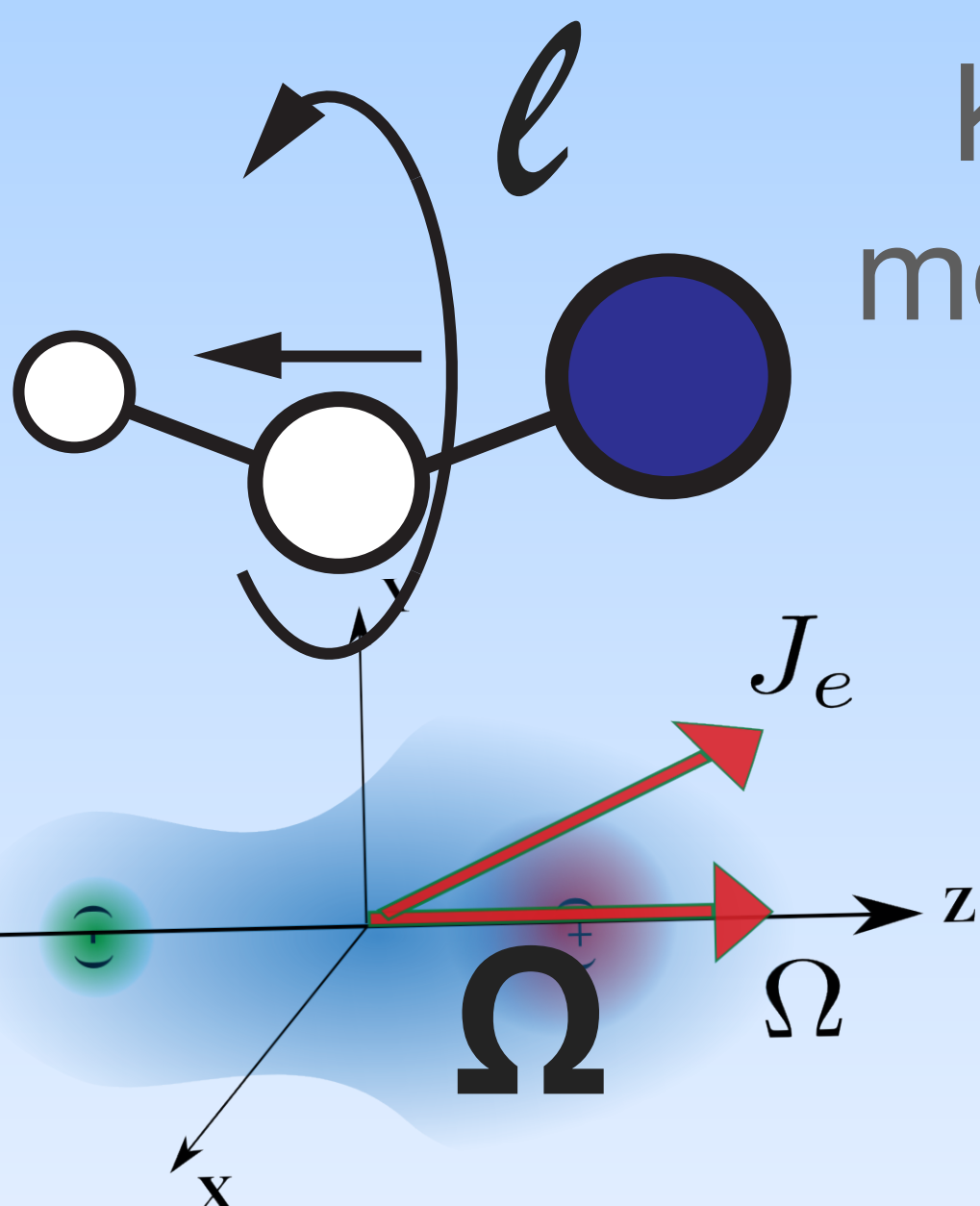
Doyle/Hutzler/Garcia-Ruiz
Harvard/Caltech/MIT

Orbital Angular Momentum Along the Internuclear Axis

Molecular Frame
Quantum Numbers

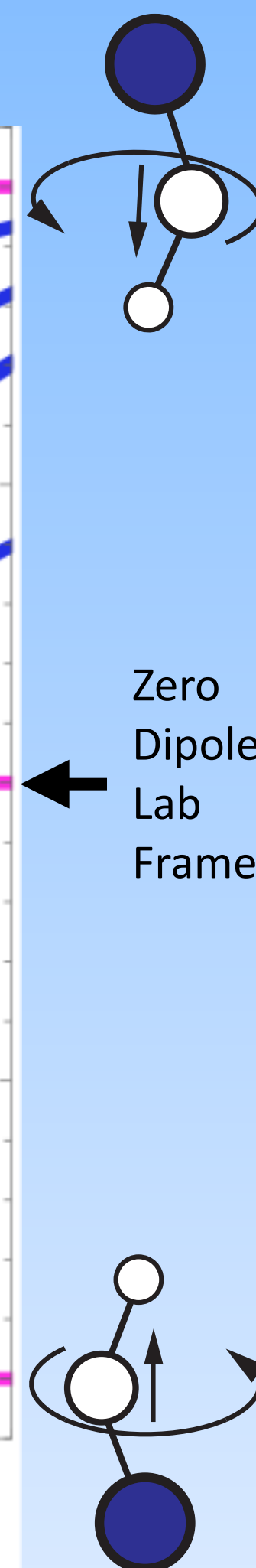
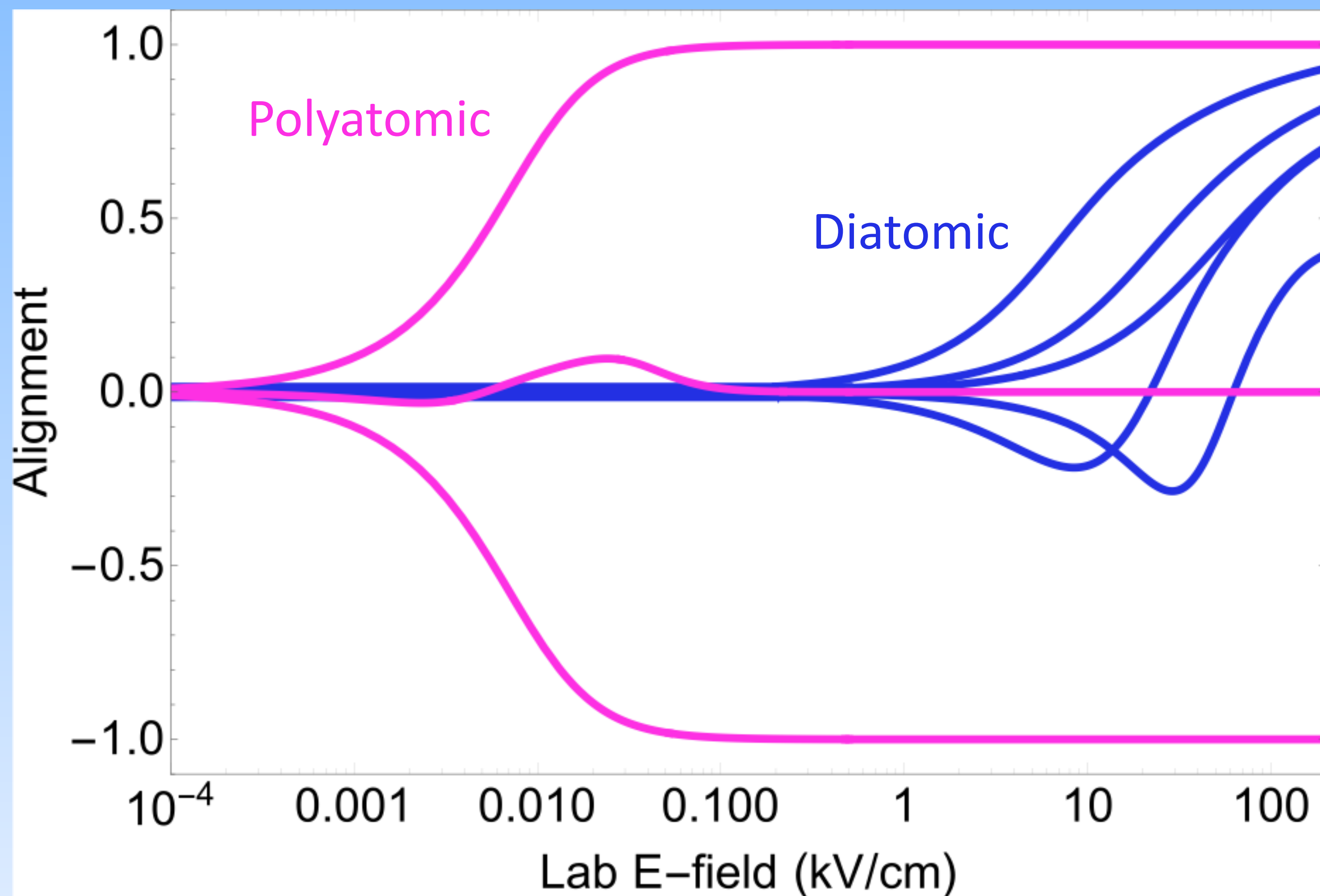


Can
laser
cool
 K and l
molecules



Can't laser cool Ω molecules

“The power of the Parity Doublet”



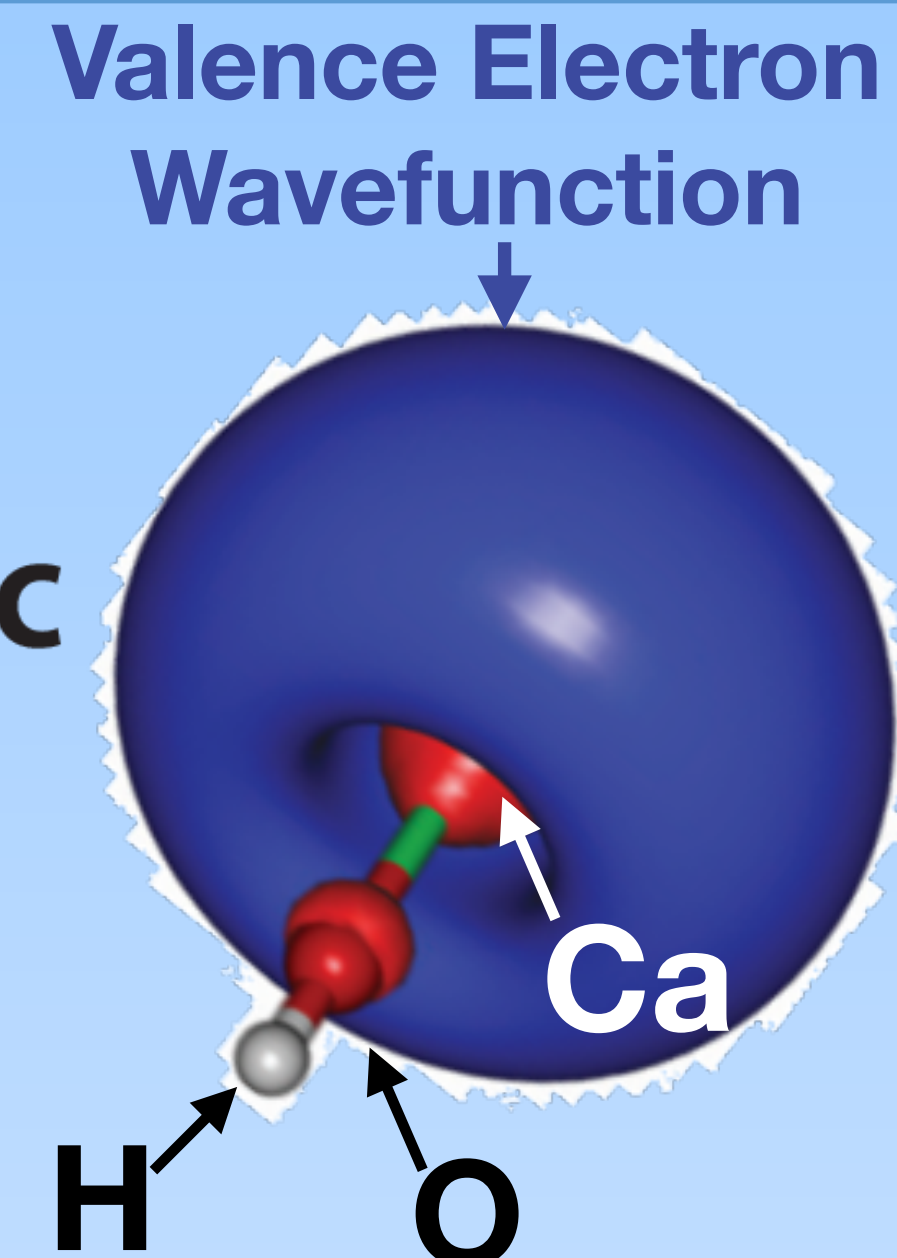
Laser Cooling Polyatomic Molecules

October 2016 ChemPhysChem Paper. See also Isaev and Berger 2016



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]



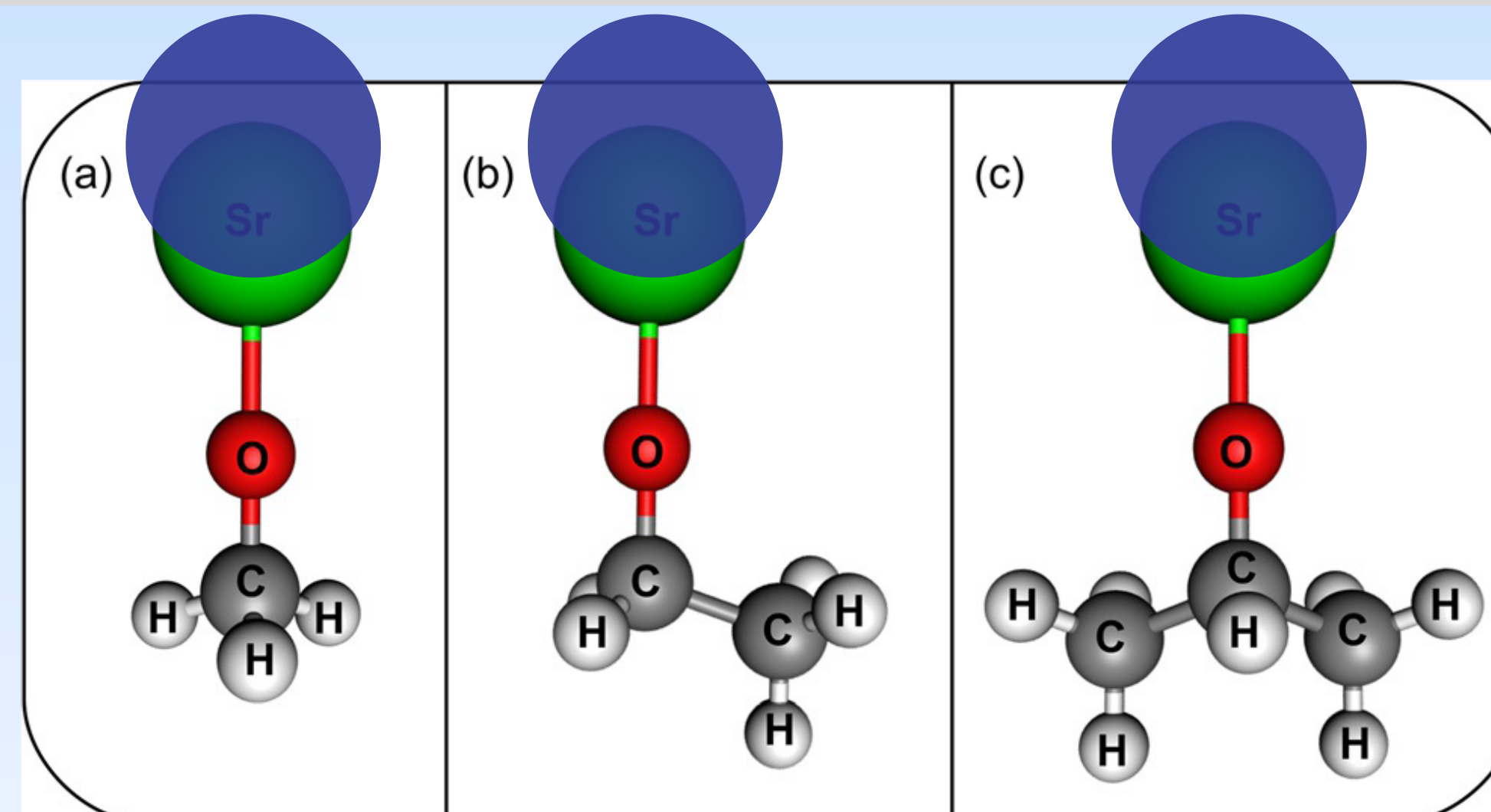
An experimentally feasible strategy for direct laser cooling of polyatomic molecules with six or more atoms is presented. Our approach relies on the attachment of a metal atom to a complex molecule, where it acts as an active photon cycling site. We describe a laser cooling scheme for alkaline earth

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.

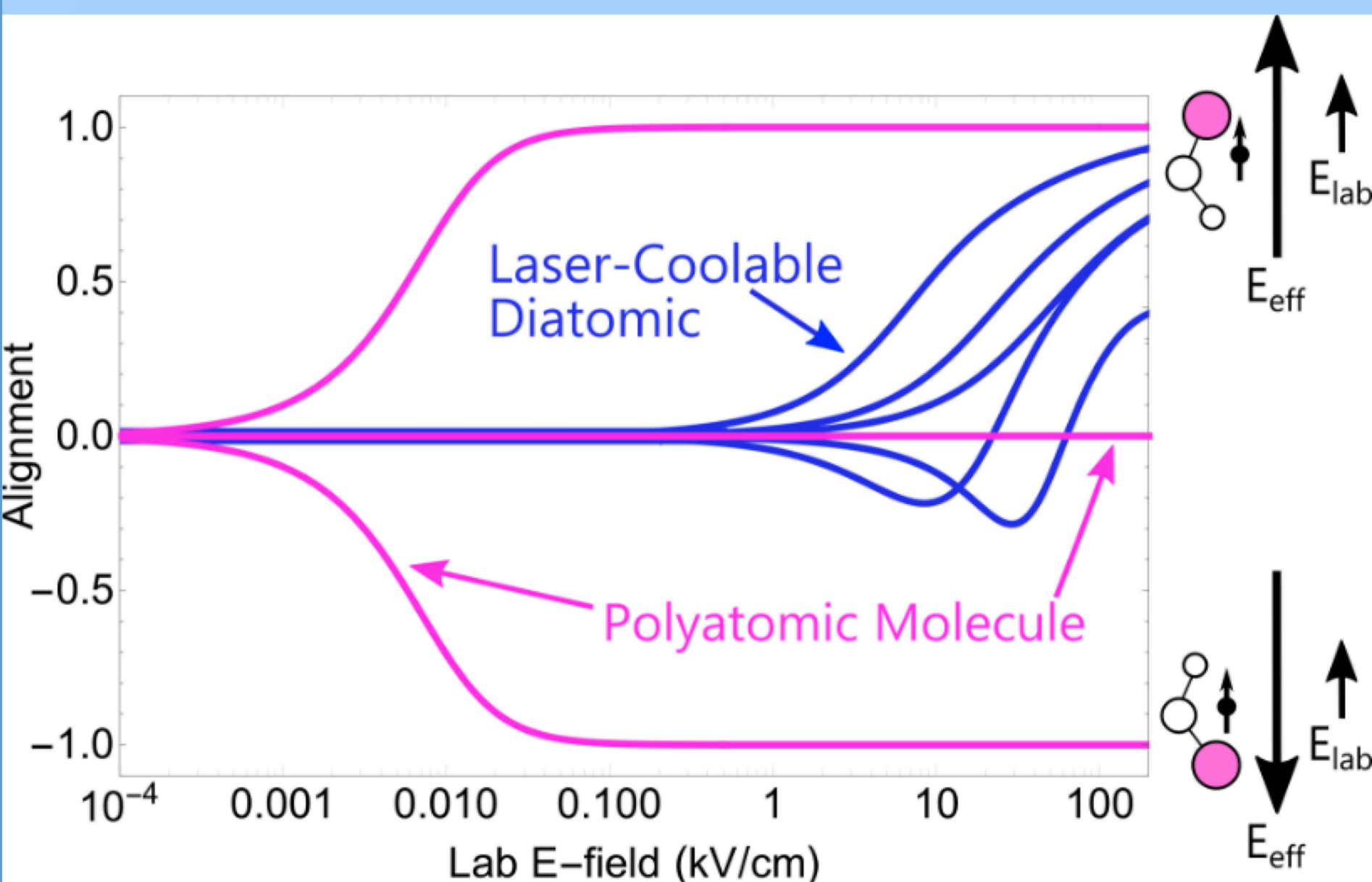
Metal-Oxide-Radical
“MOR”

Molecule Motif

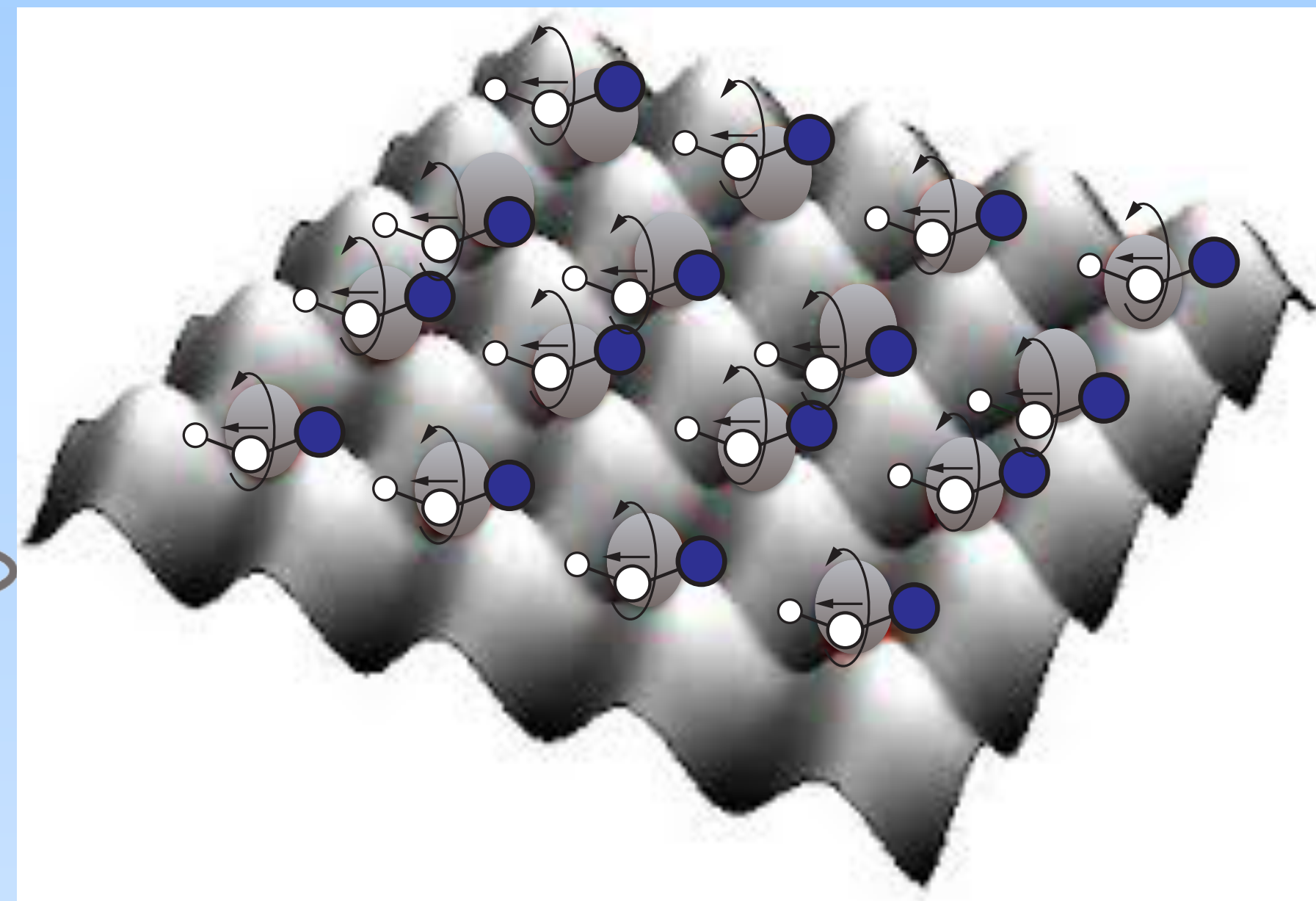
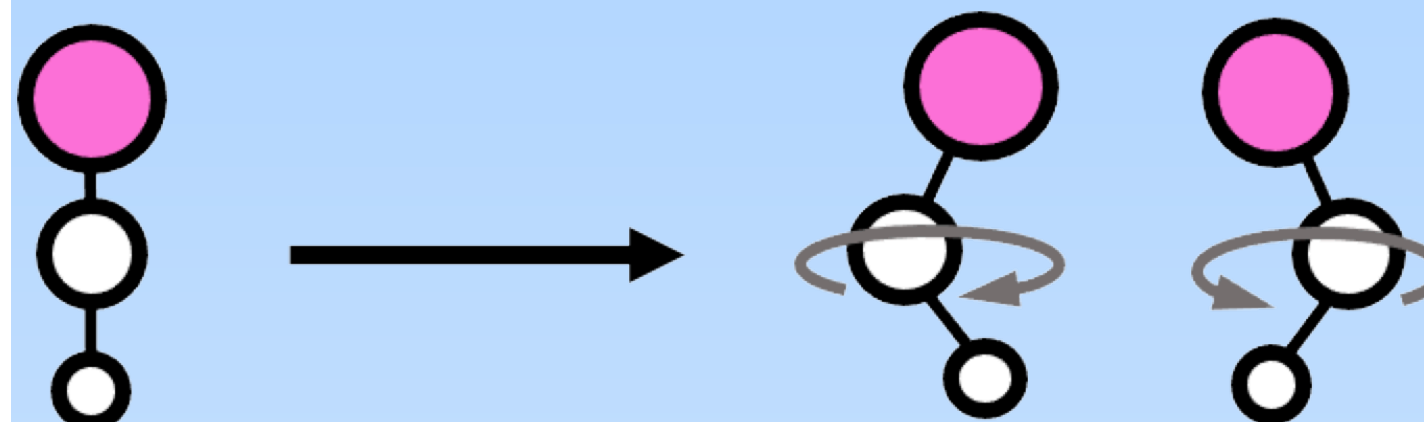
a.k.a Alkaline Earth Atom
Pseudofluoride Molecules



EDM Search Strategy - Laser Cooled Polyatomic Molecules

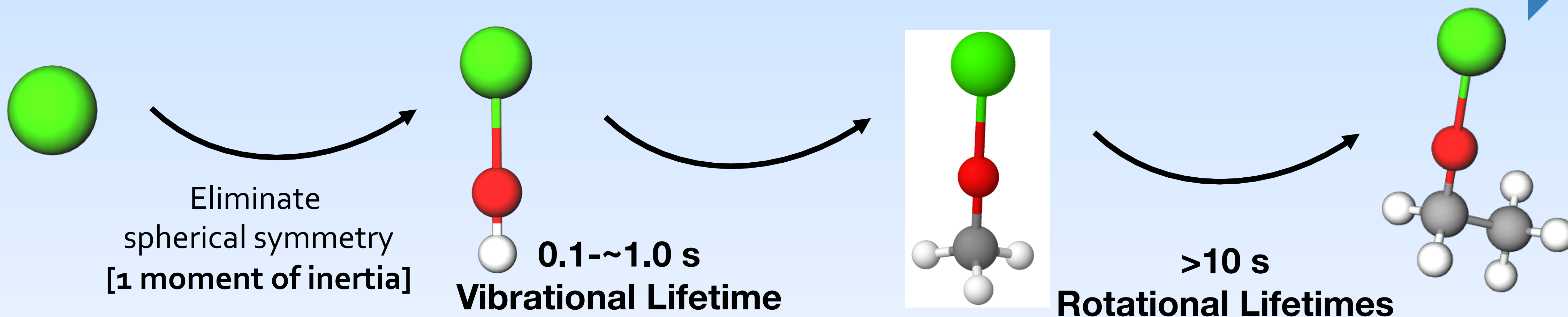


● Sr, Yb, Ba, Ra...



[Precision Measurement of Time-Reversal Symmetry Violation with Laser-Cooled Polyatomic Molecules](#) Kozyryev and Hutzler, PRL, 119 (2017)

Complexity [reduced molecular symmetry]



Our Laser Cooling Path to Ultracold Molecules

Typical values for
atoms

10^{12}

10^8

10^6

Molecule Number

10^{10}

10^6

1.3×10^5

2×10^4

Cryogenic Buffer
gas cooling

Laser
Slowing

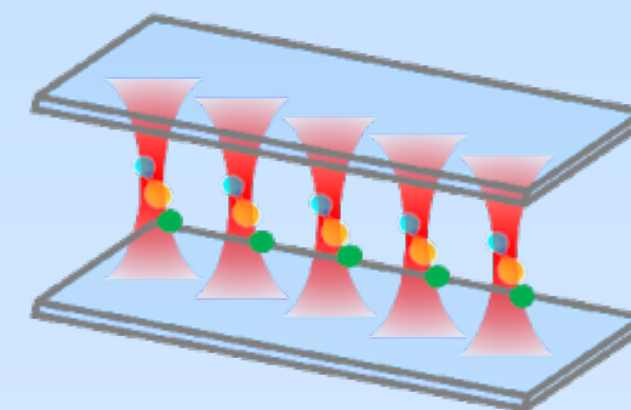
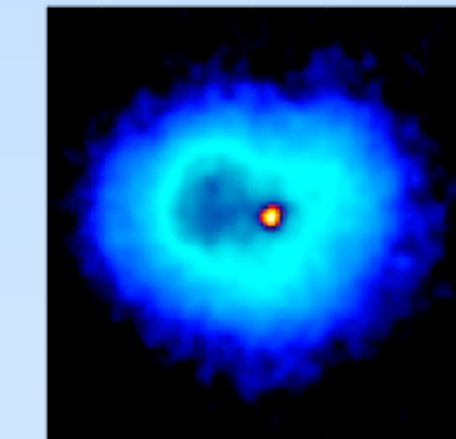
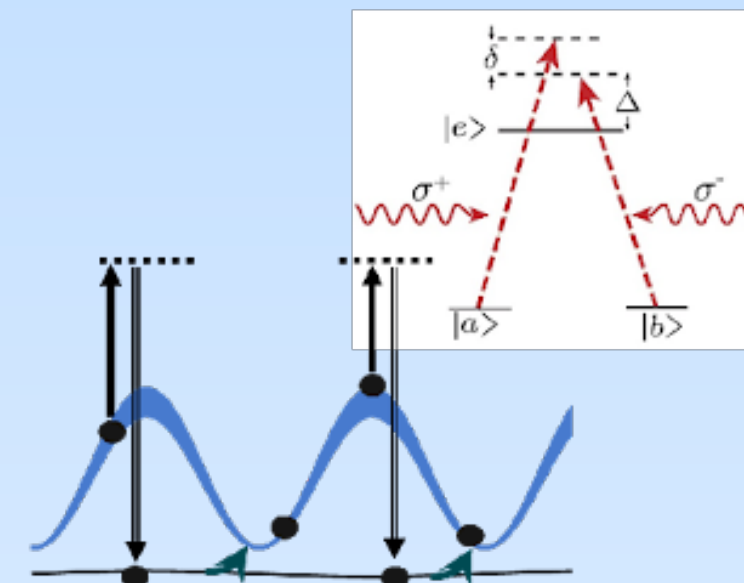
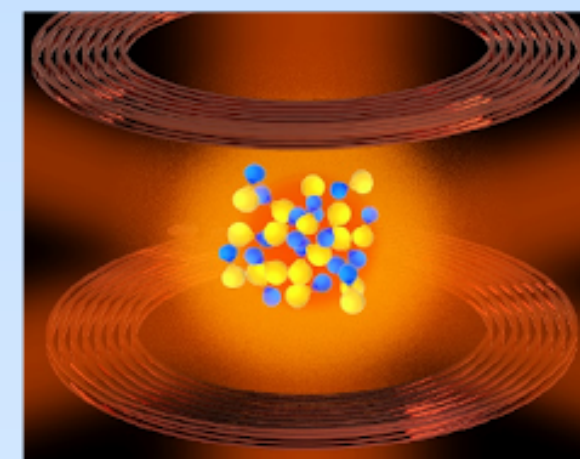
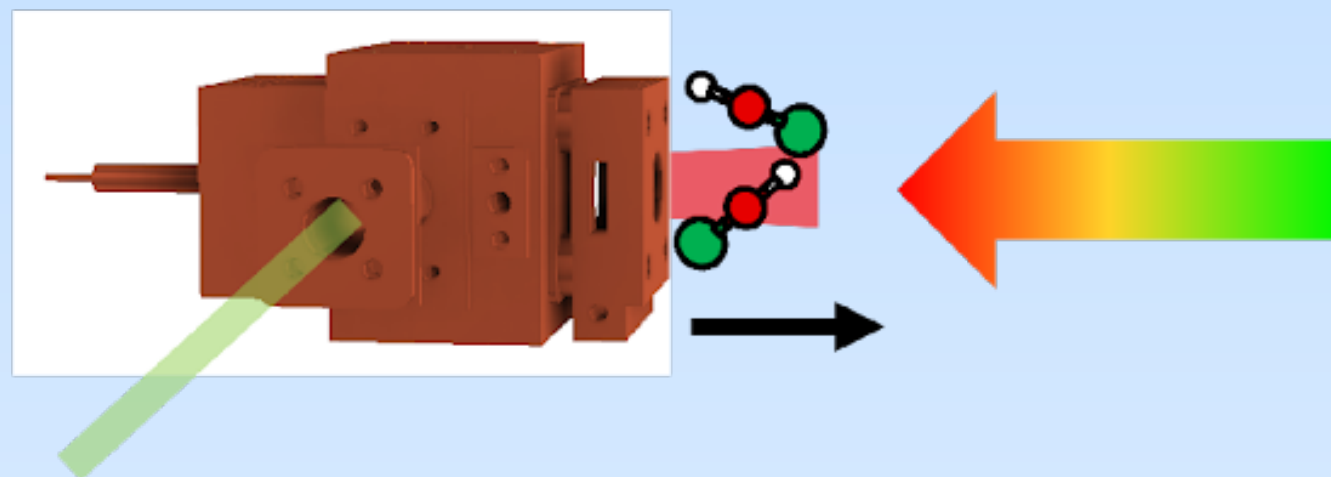
Red-detuned
RF MOT

Sub-Doppler
Cooling

Conveyor
MOT

ODT

Optical
Tweezers



>1000 K

2 K

2 mK

$5 \mu\text{K}$

$100 \mu\text{K}$

Temperature

Phase Space Density

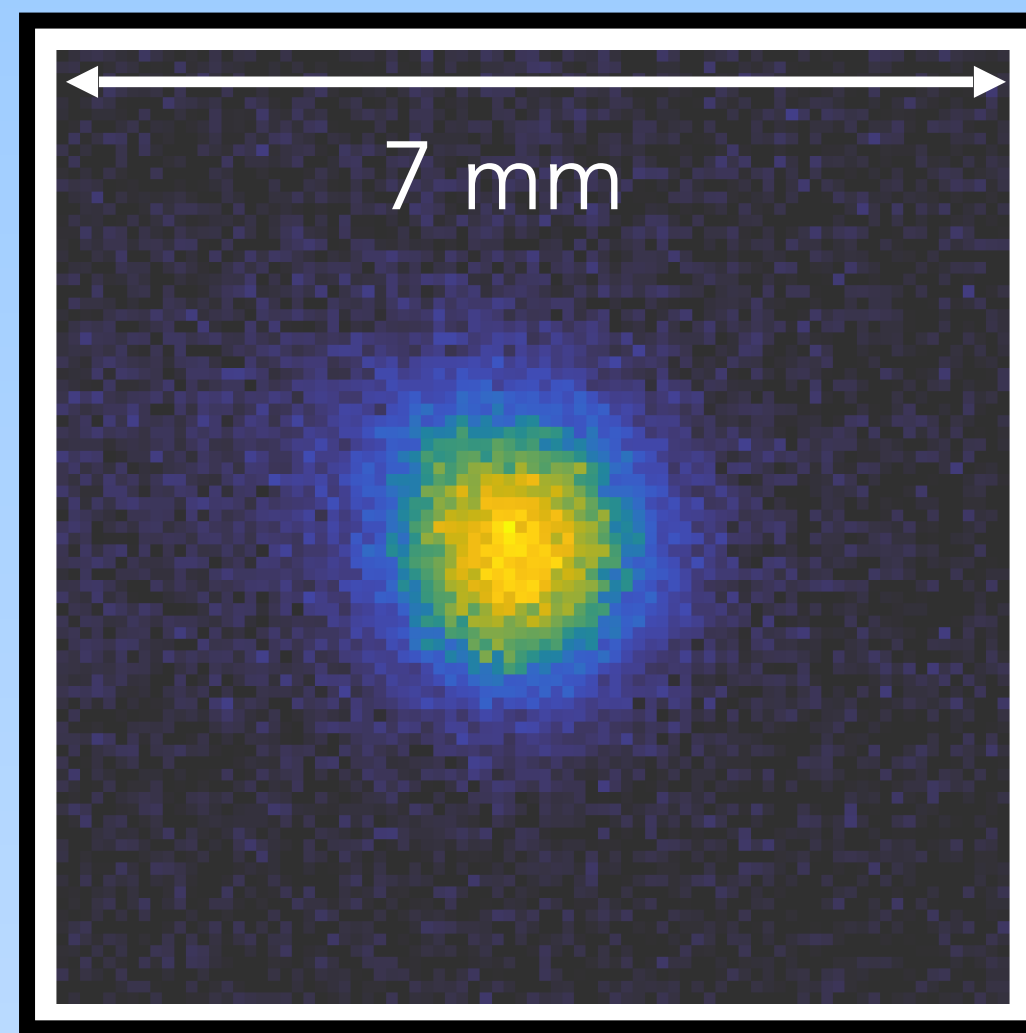
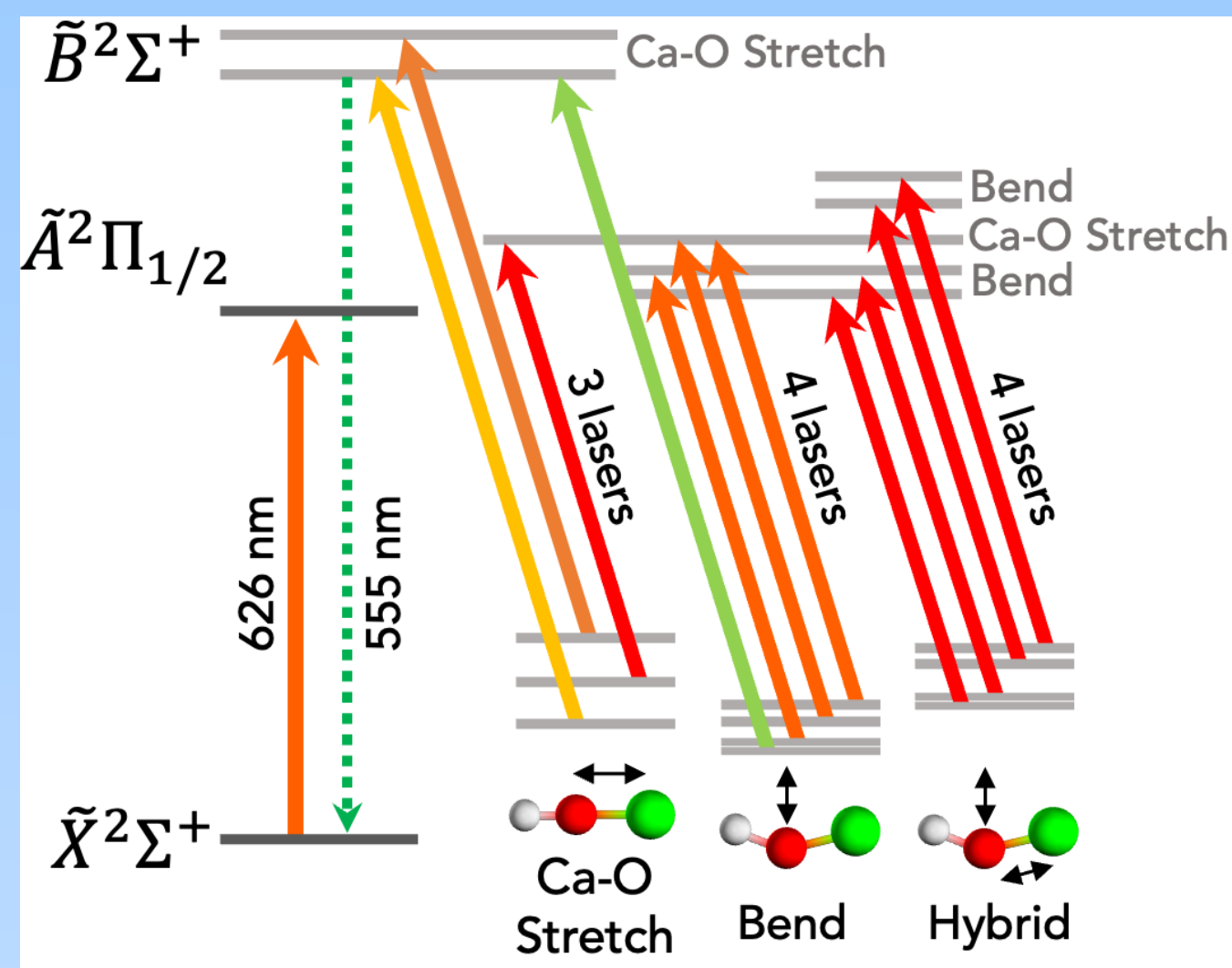
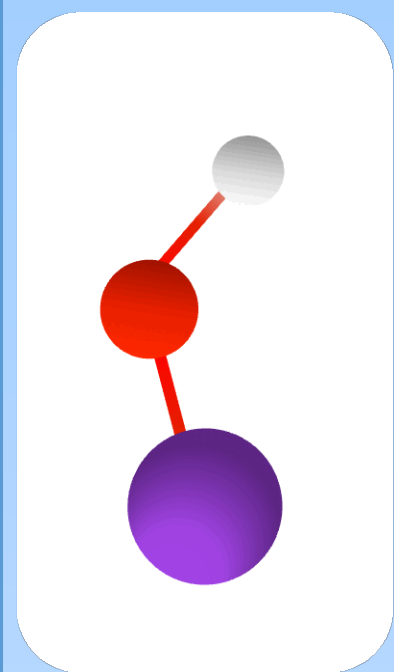
10^{-14}

10^{-11}

10^{-8}

10^{-6}

10^{-4}

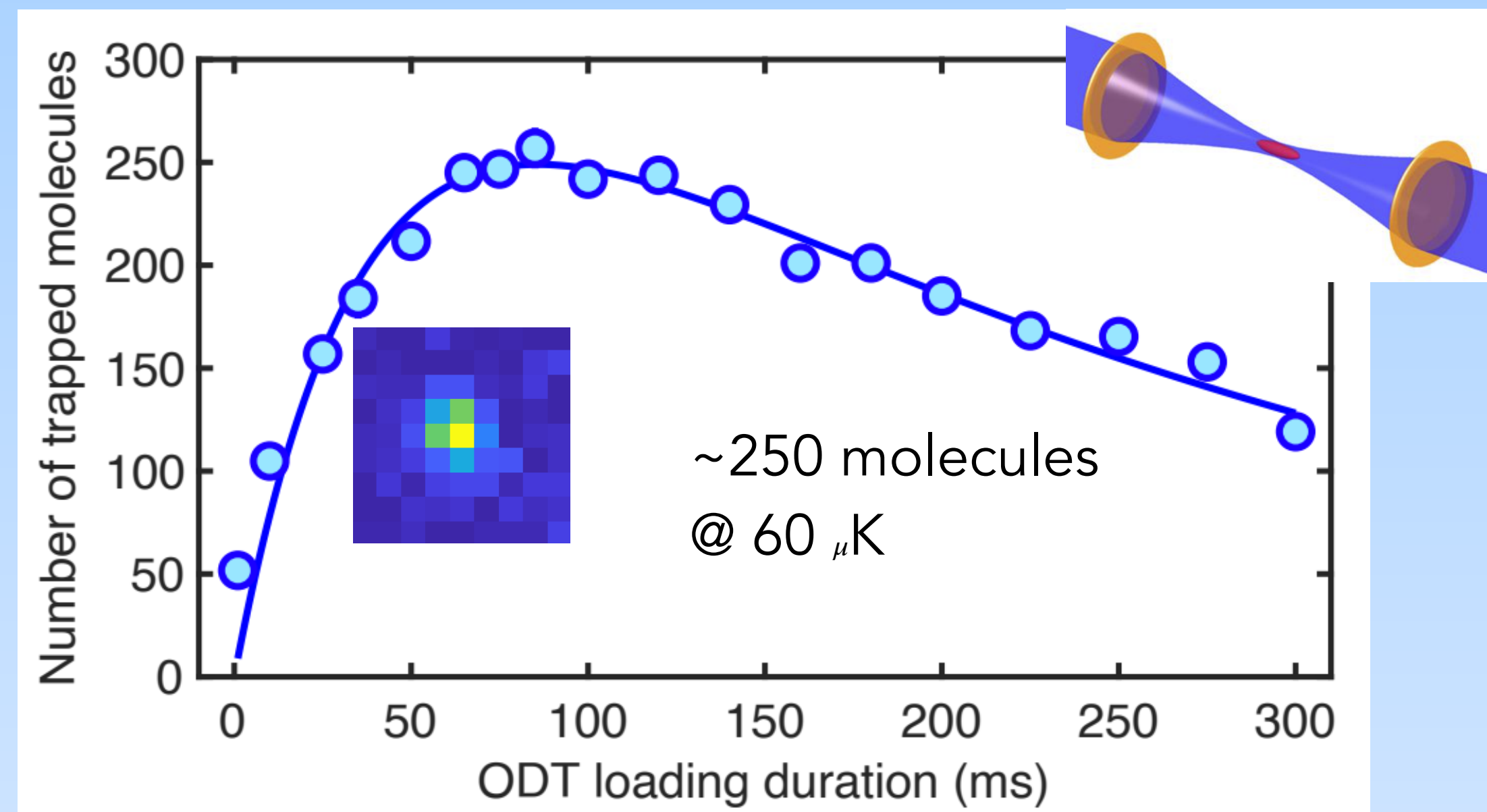


Scatter ~15000 photons by repumping vibrational decay pathways

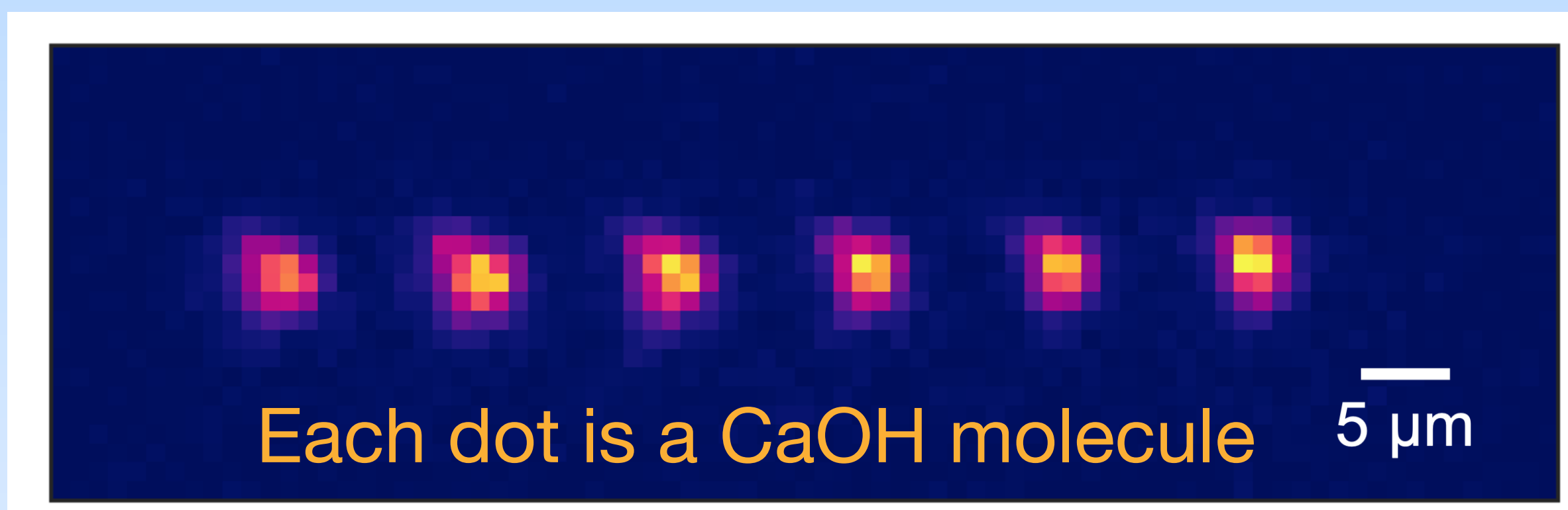
Magneto-Optical Trap of 20,000 CaOH Molecules

CaOH

CaOH in Optical Dipole Trap



Sub-Doppler Cooling and Optical Dipole Trap (ODT) Loading



Tweezer Array of Single CaOH Molecules

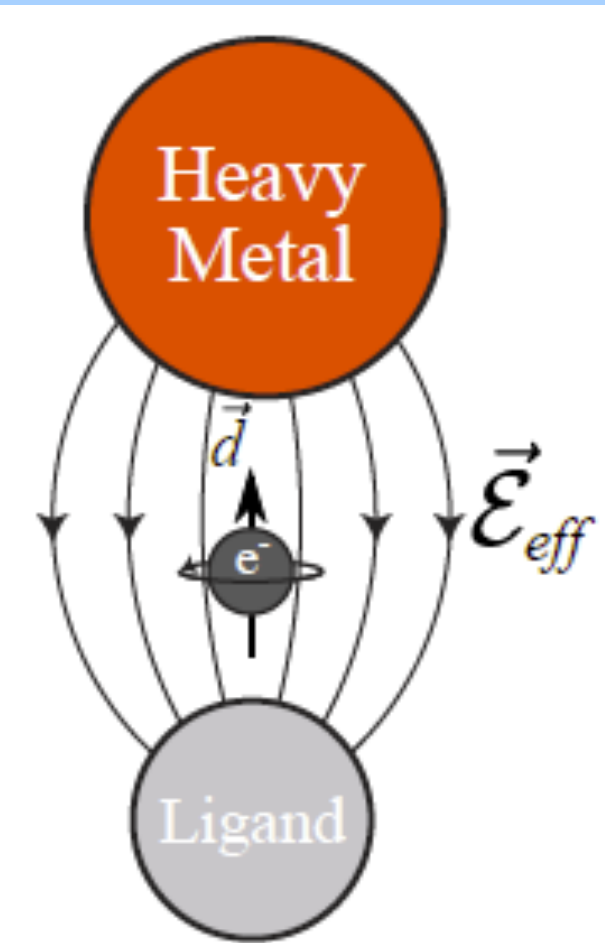
[Magneto-Optical Trapping and Sub-Doppler Cooling of a Polyatomic Molecule](#) N. B. Vilas, C. Hallas, L. Anderegg, P. Robichaud, A. Winnicki, D. Mitra, J.M.D. *Nature* 606, 70-74 (2022).

[Optical Trapping of a Polyatomic Molecule in an I-Type Parity Doublet State](#), C. Hallas, N. B. Vilas, L. Anderegg, P. Robichaud, A. Winnicki, C. Zhang, L. Cheng, J.M.D, *Phys. Rev. Lett.* 130, 153202 (2023).

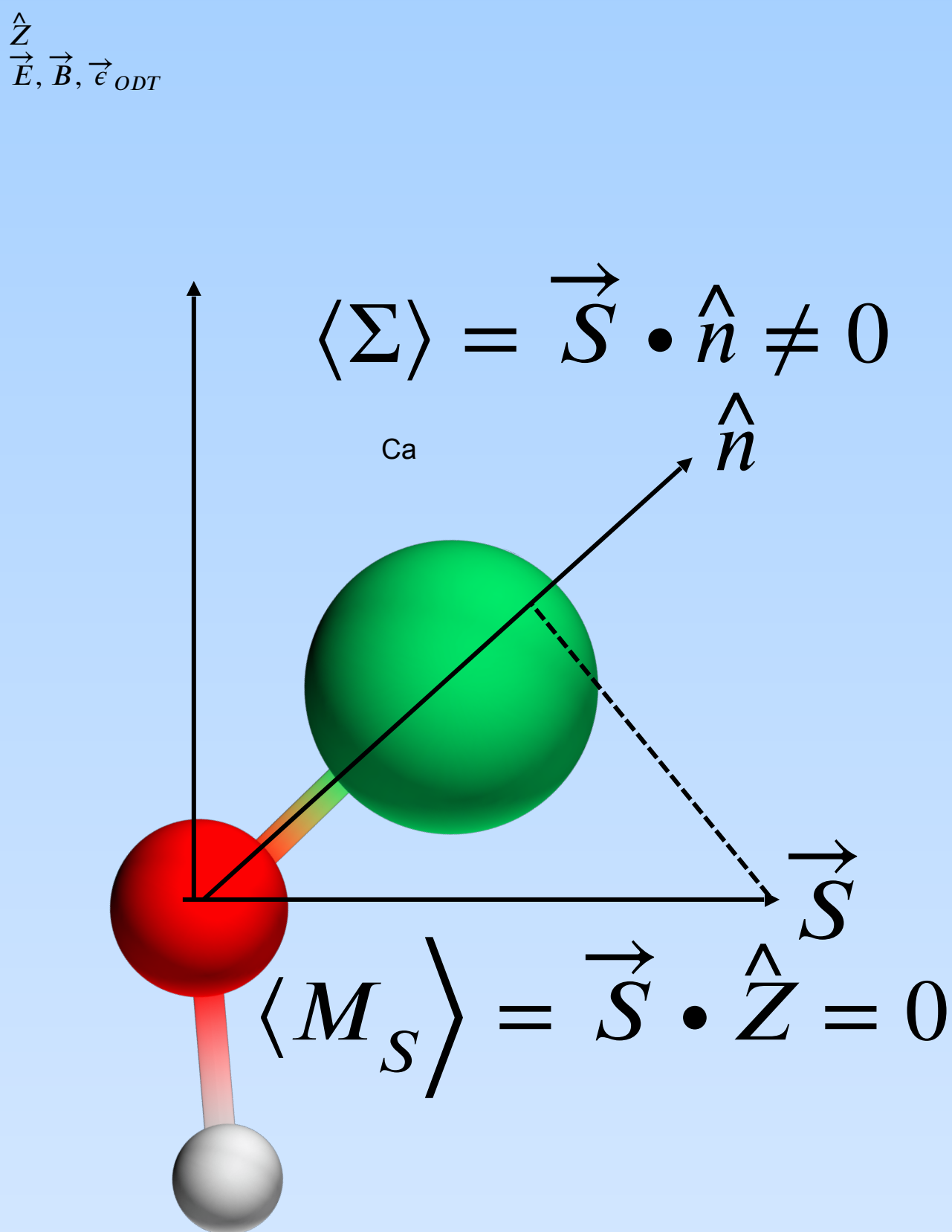
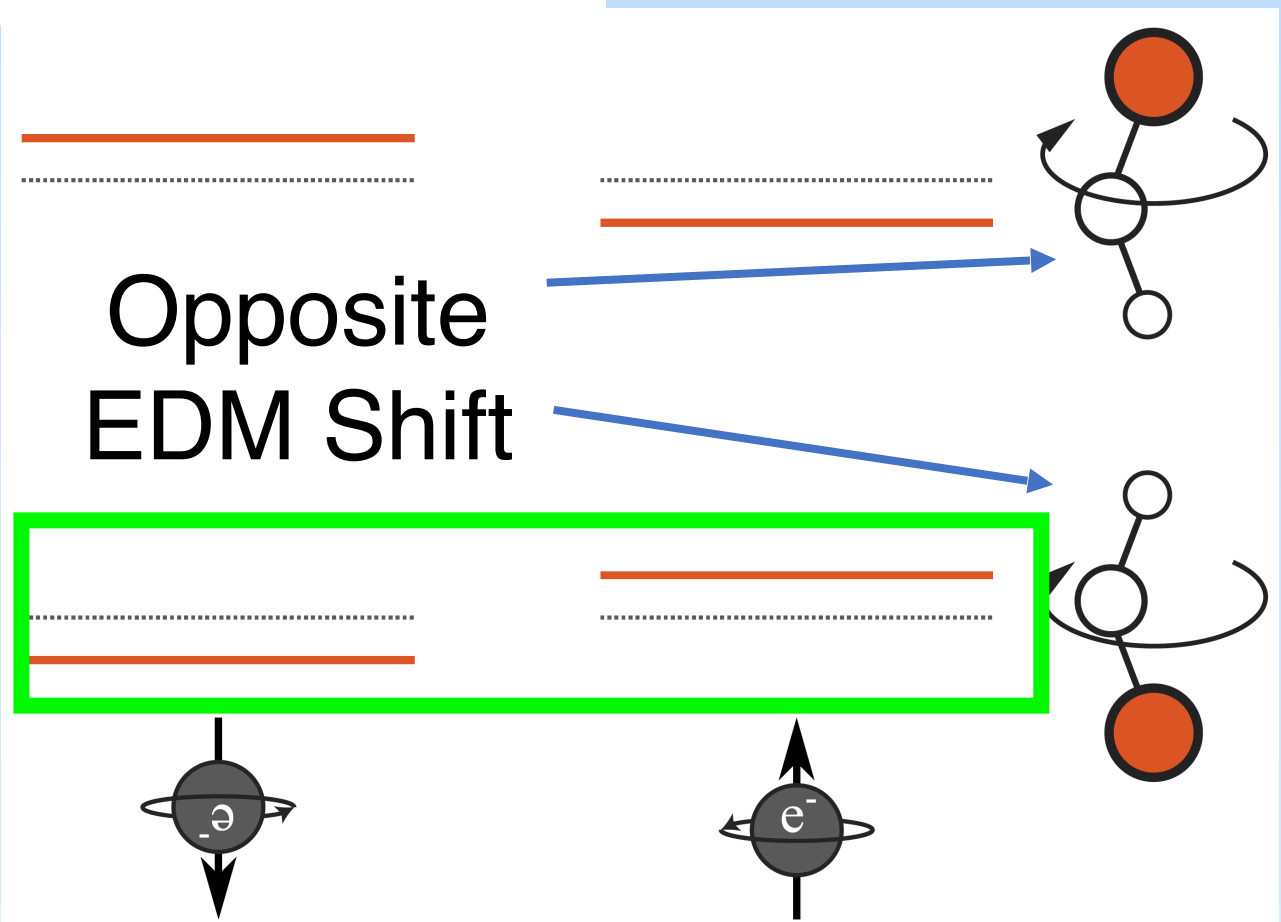
[An optical tweezer array of ultracold polyatomic molecules](#), N. B. Vilas, P. Robichaud, C. Hallas, G. K. Li, L. Anderegg, J.M.D. *Nature* 628, 282-286 (2024).

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

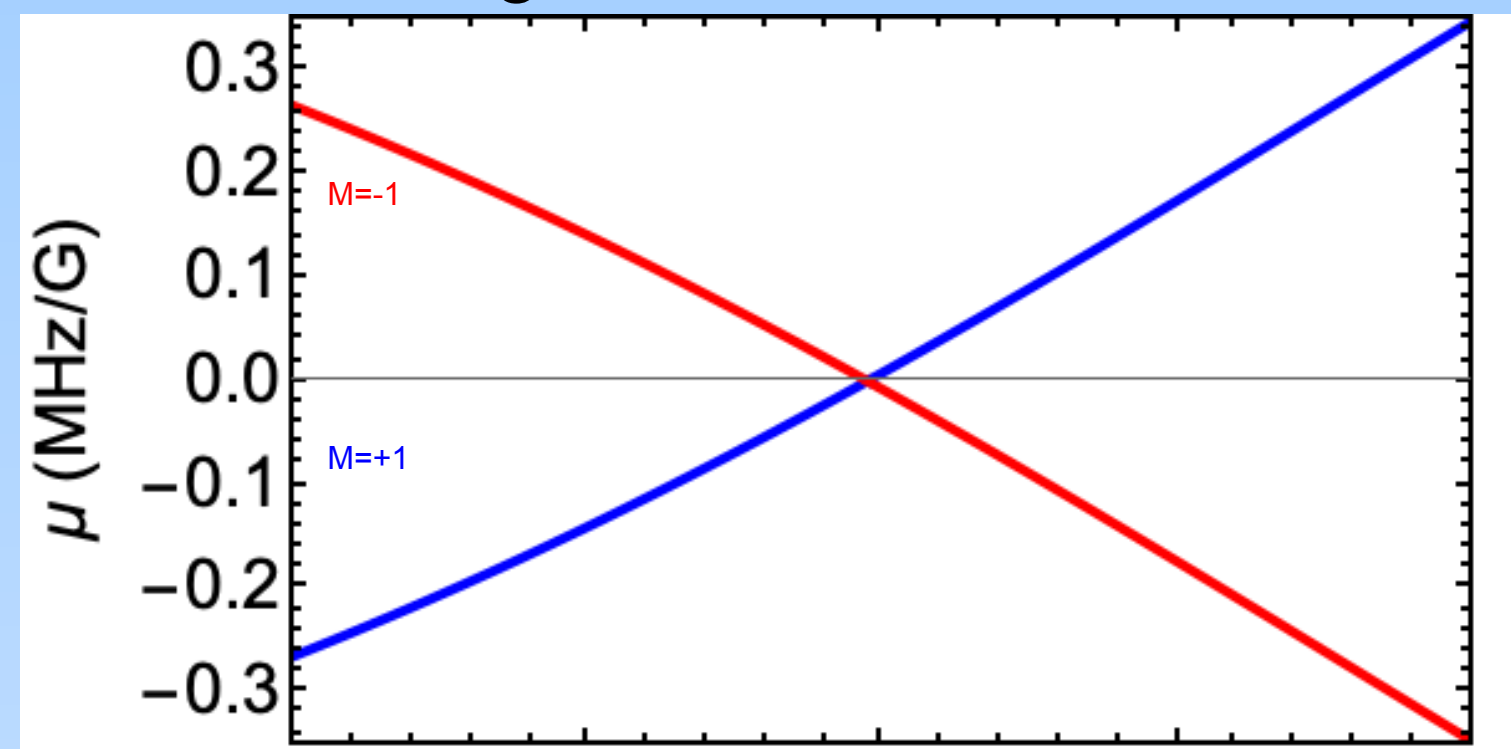
Pathfinder Experiment to EDM CaOH



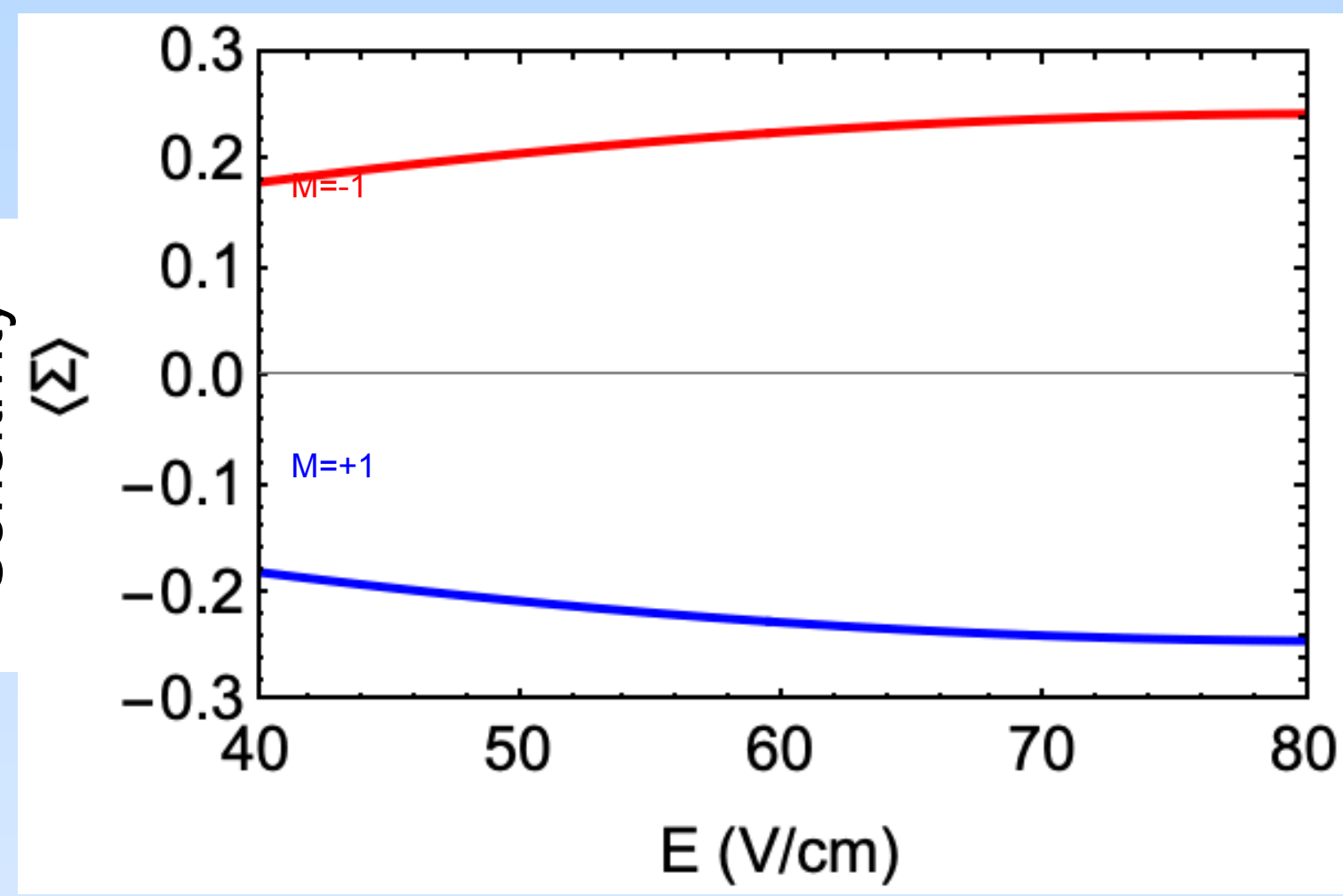
- Application of a lab E_Z results in states with “zero” electron g-factor
 - Still sensitive to EDM interaction!
- Useful for rejecting systematic errors from stray B_Z fields



Zero g-factor EDM states

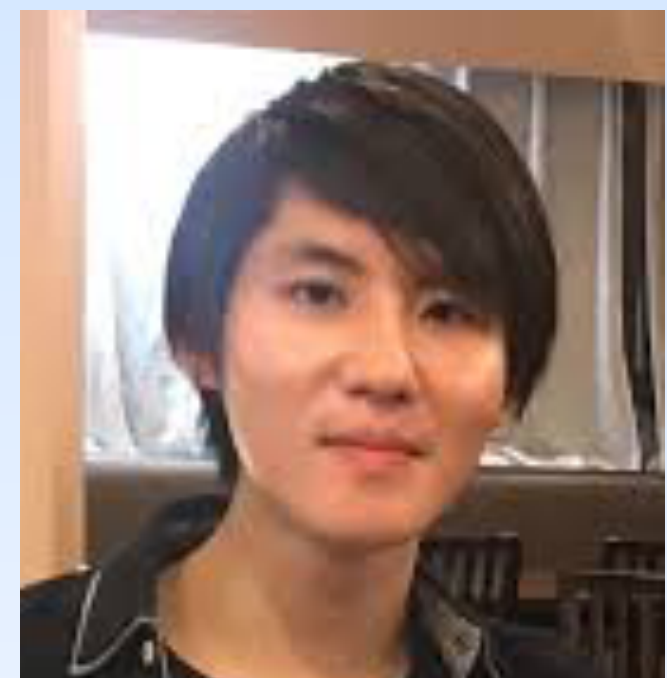


EDM Sensitivity

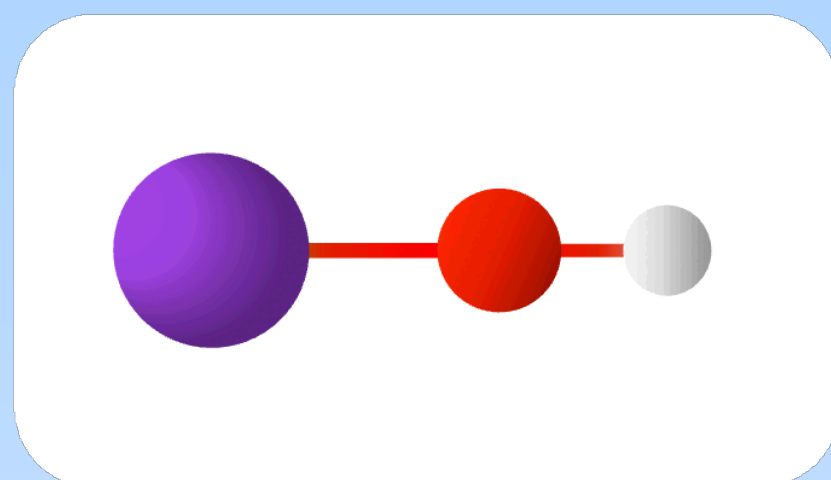


Engineering Field-Insensitive Molecular Clock Transitions for Symmetry Violation Searches

Yuiki Takahashi, Chi Zhang, Arian Jadbabaie, and Nicholas R. Hutzler
 Phys. Rev. Lett. **131**, 183003 – Published 31 October 2023



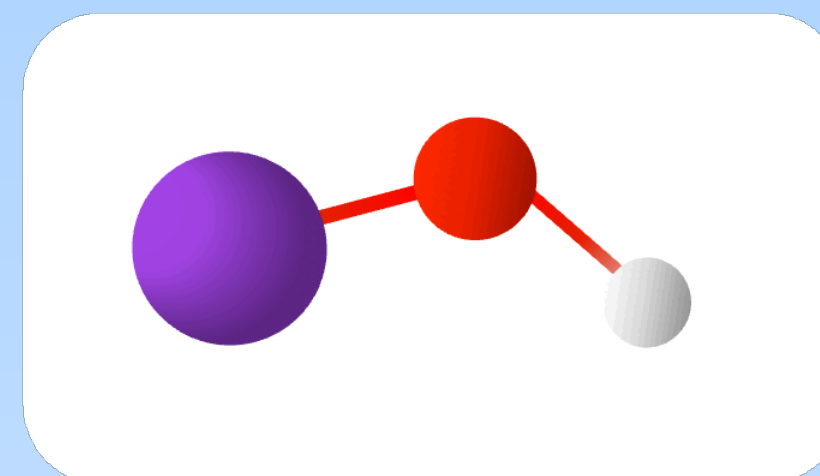
Triatomic Vibrational Quantum Numbers



$X^2\Sigma^{+}(200)$

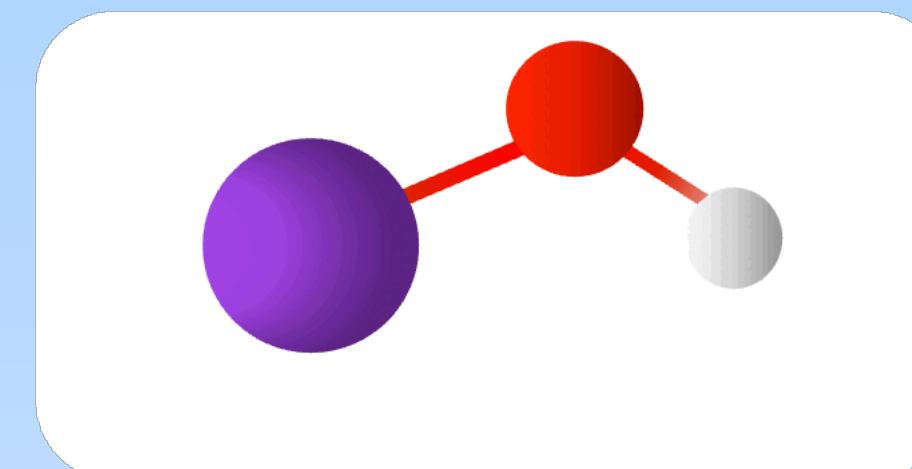
Sr-O stretch mode
Quantum number

O- H stretch mode
Quantum number



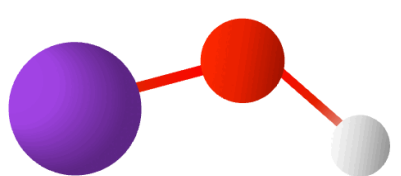
$X^2\Sigma^{+}(010)$

Sr-O-H bending mode
Quantum number
($v=1$ on left, $v=3$ on right)



$X^2\Sigma^{+}(03^1 0)$

Angular Momentum
of Vibration
Quantum Number
($\mathcal{L} = 1$ in here)

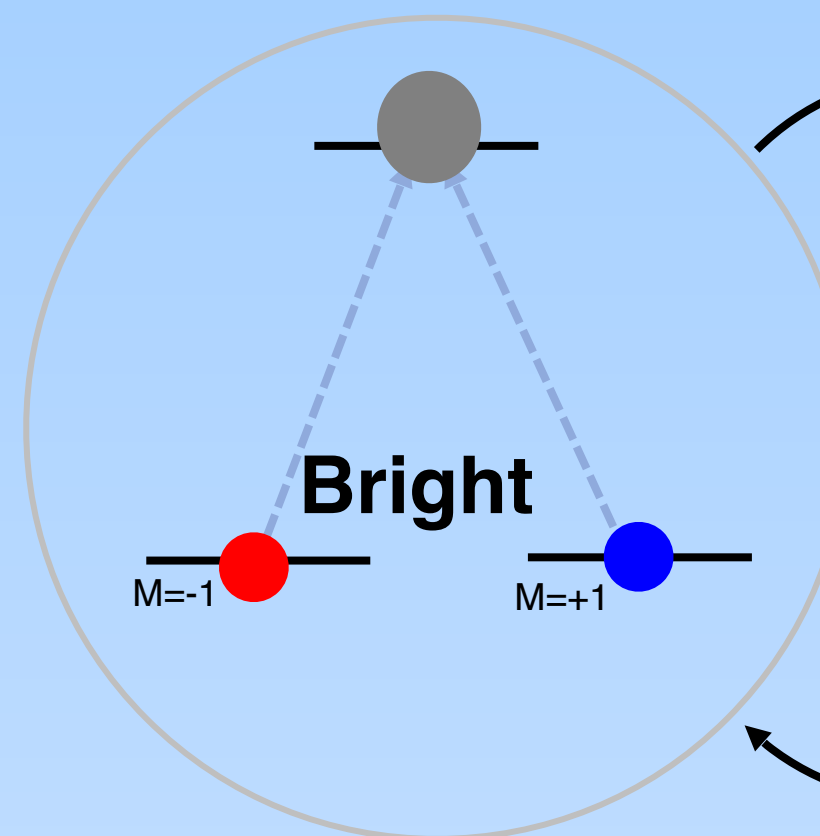
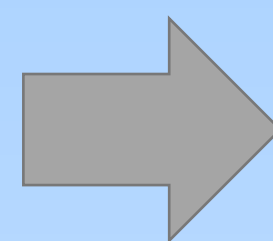
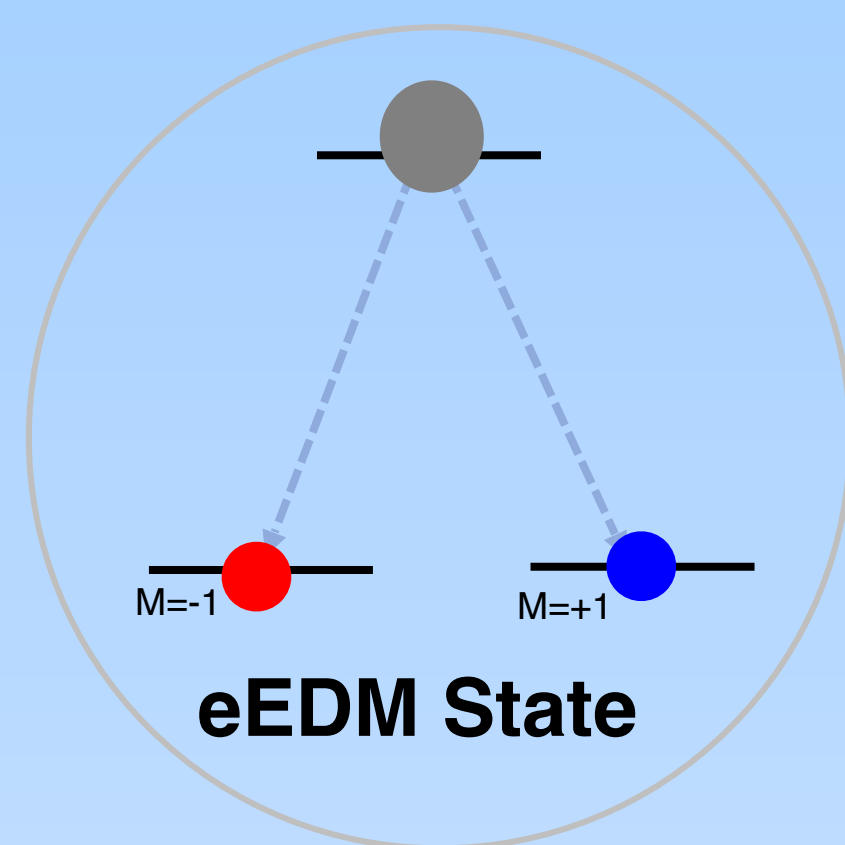
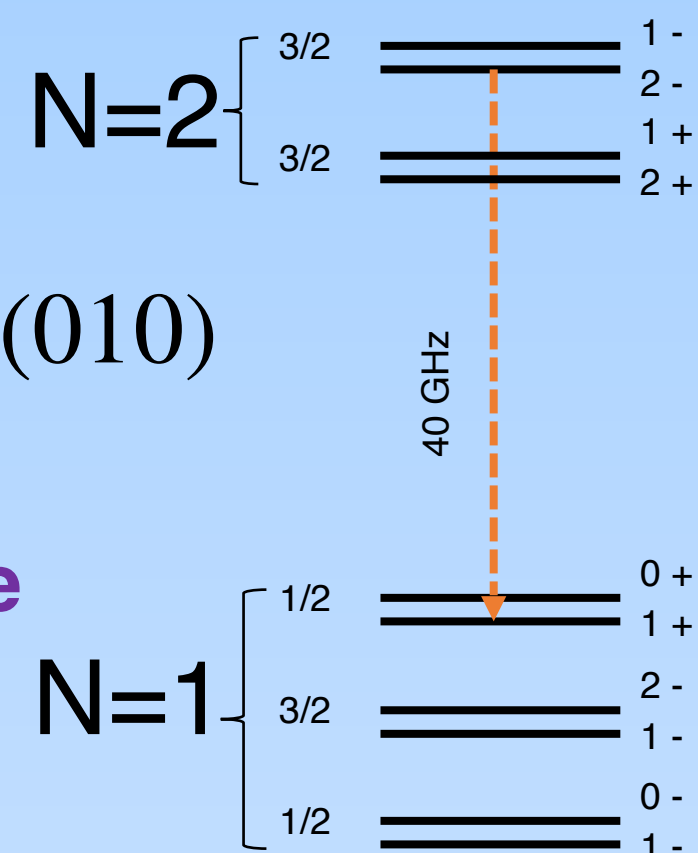


Electron Spin Precession in the CaOH EDM State

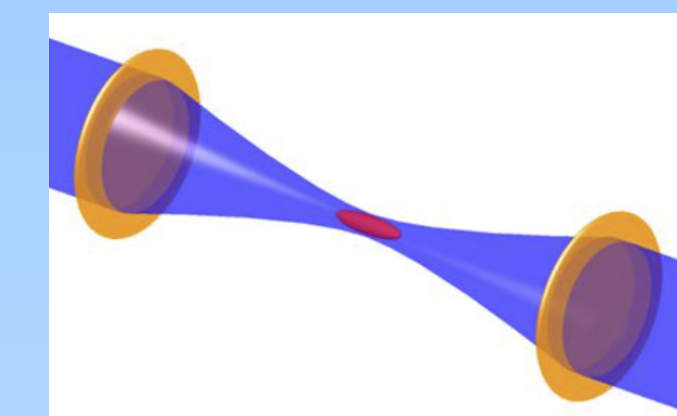
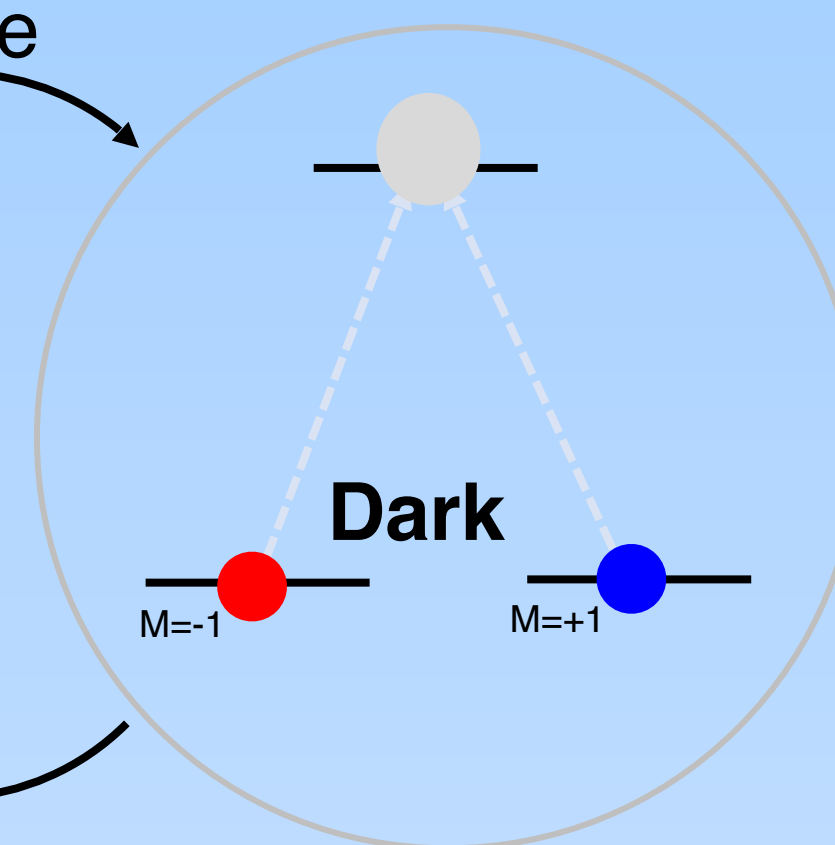
Bending Mode: EDM States

$X^2\Sigma^{+}(010)$

EDM Sensitive State

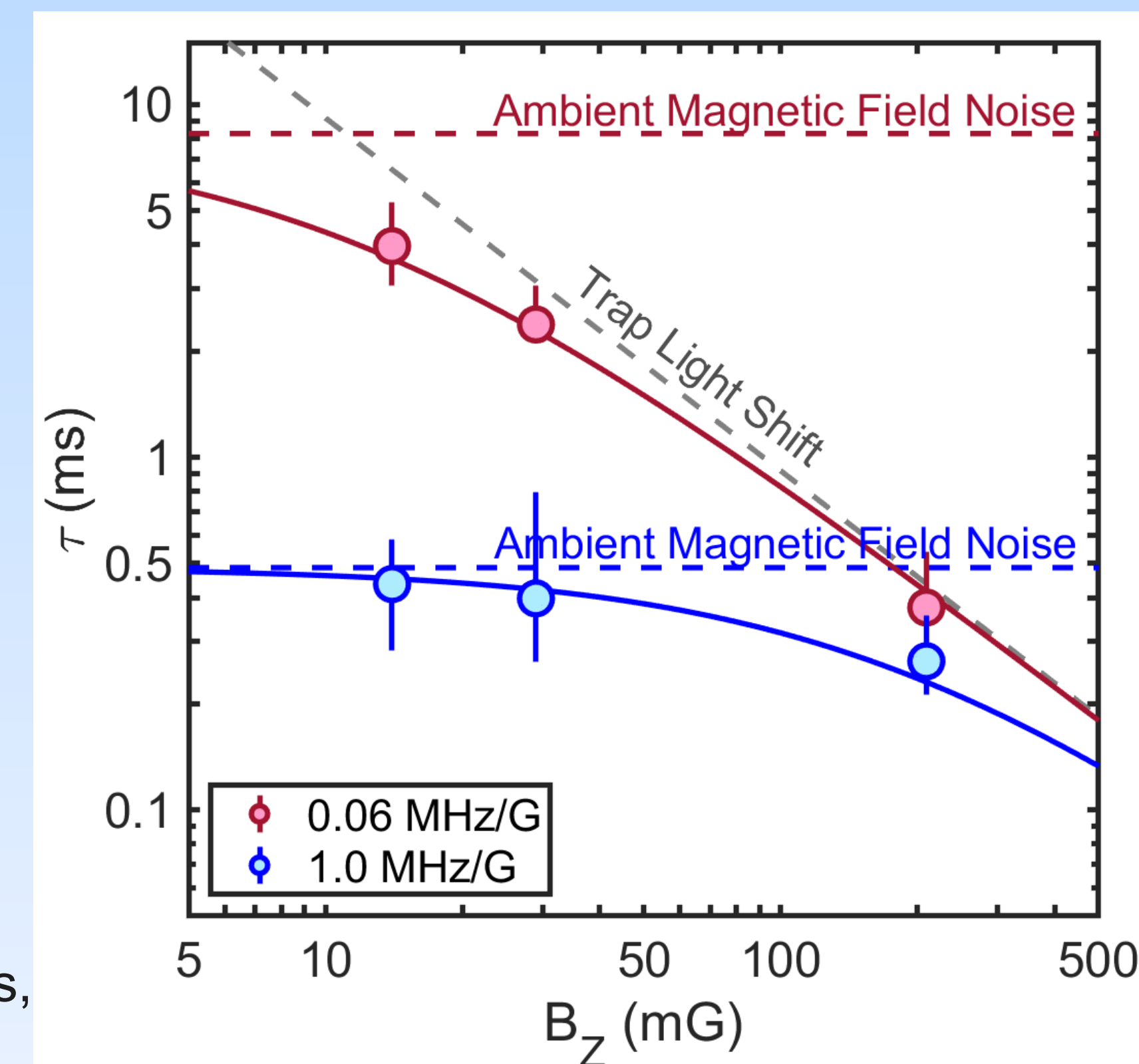
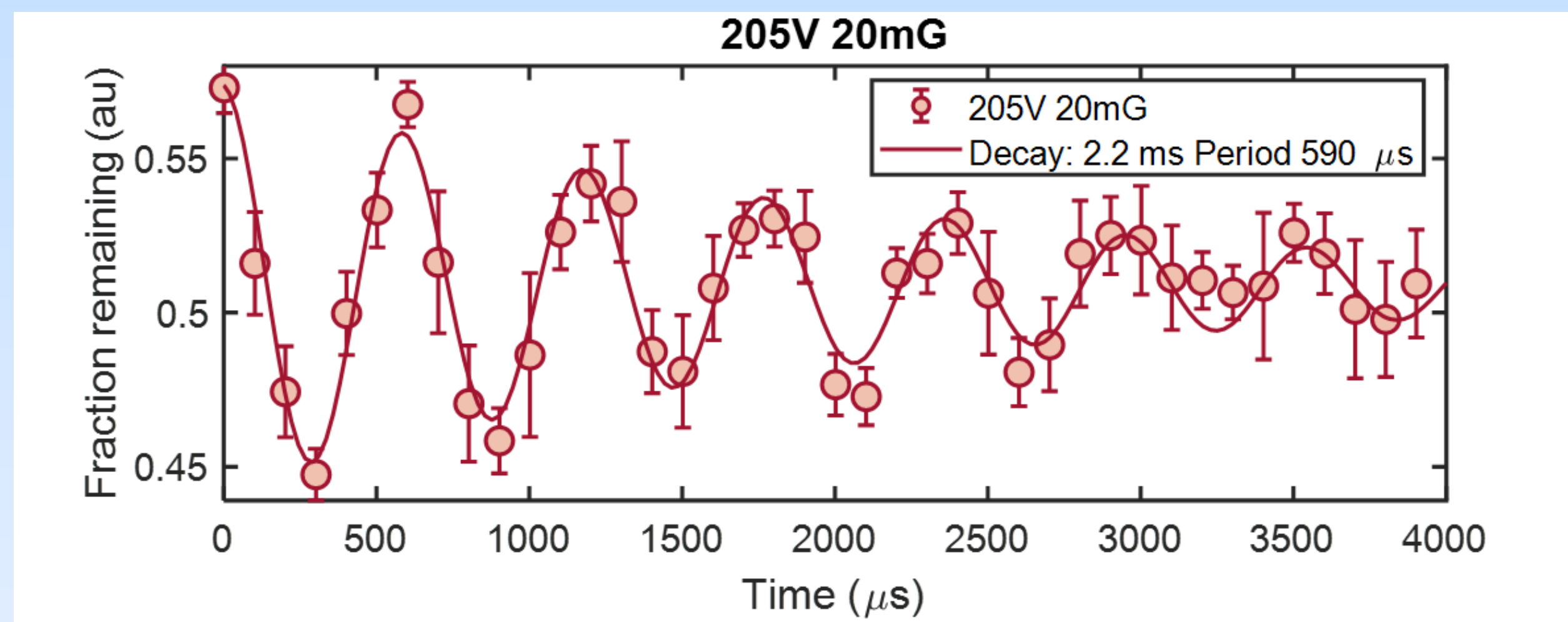


Precession Time



Increased Coherence Near Zero g-factor

eEDM Sensitive Spin Precession

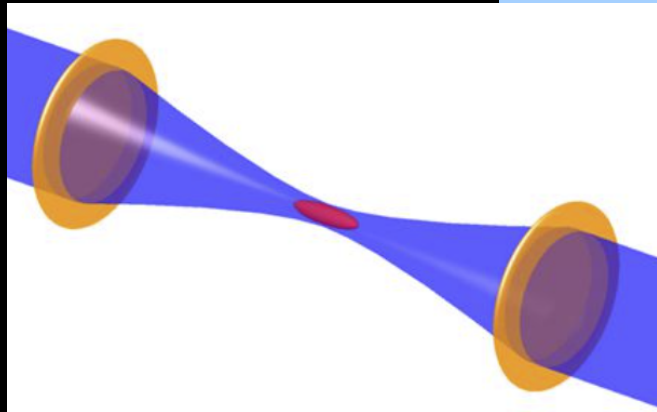


eEDMs... CaOH is too light for eEDM sensitivity...
But SrOH pretty good....

Molecule	ϵ_{eff} (GV/cm)
CaOH	0.30
SrOH	2.2
BaOH	6.8
YbOH	23.6
RaOH	56.9

10.1103/PhysRevA.99.042512
10.1103/PhysRevA.101.012508

Molecule	E_{eff} [GV/cm]	# in trap for $\sim 10^{-29}$ e cm
CaOH	0.30	100,000
SrOH	2.2	2,000
BaOH	6.8	200
YbOH	23.6	20
RaOH	56.9	3



Nick Hutzler - Caltech



Amar Vutha - Toronto



Tim Steimle ASU — Caltech

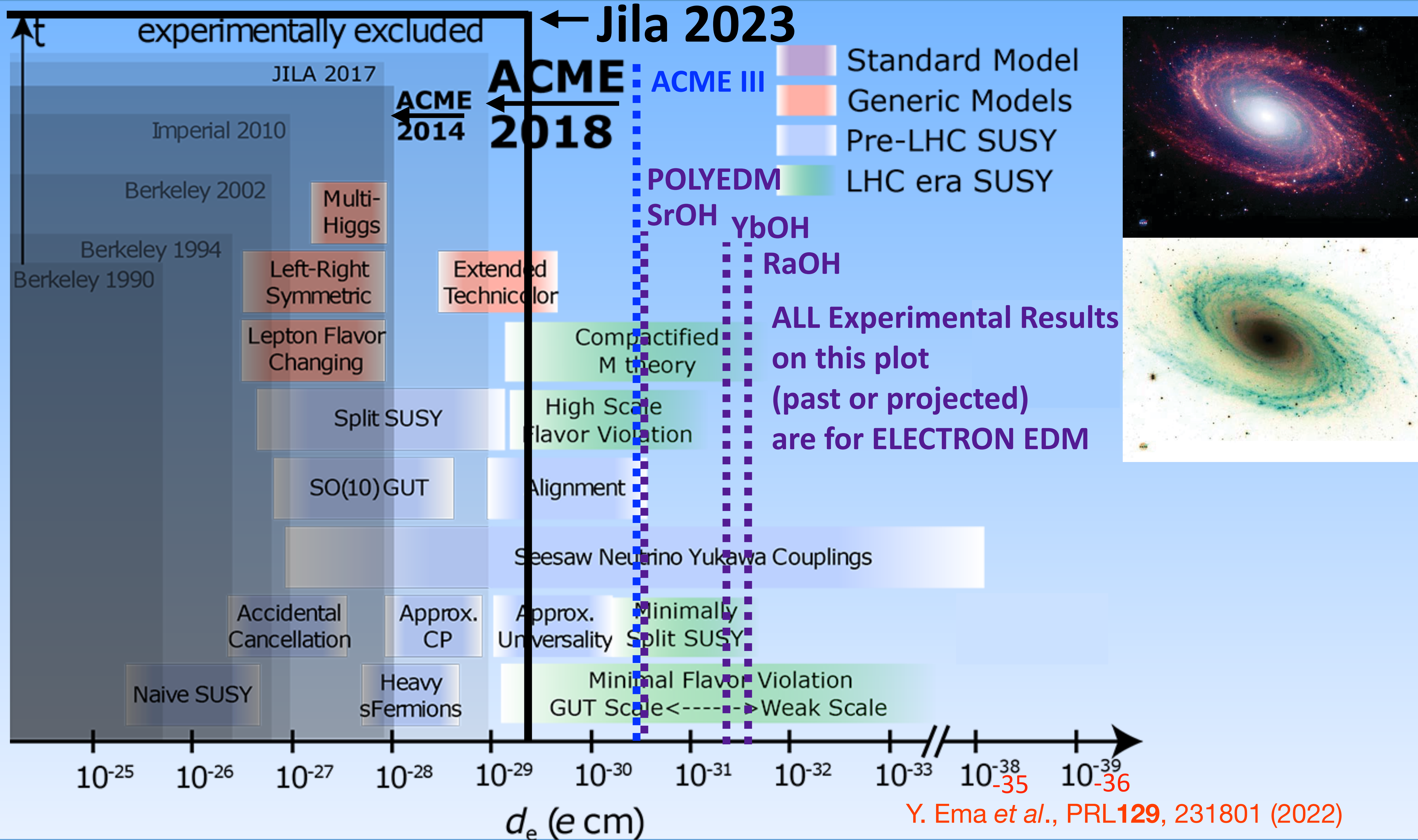
POLYEDM
Collaboration

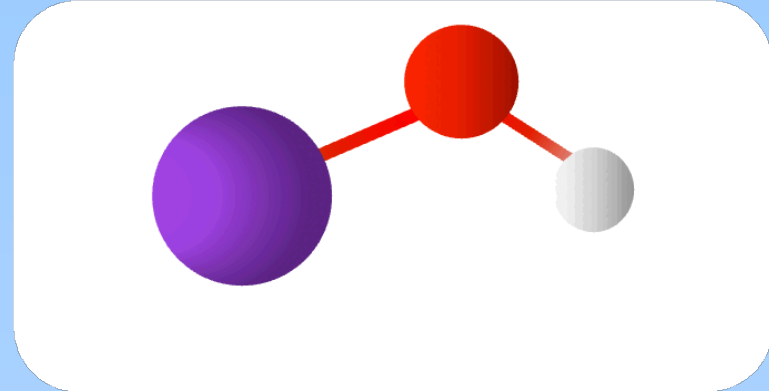
Fraction remaining (au)

$X^2\Sigma^+$

EDM Sen
State

ear
or

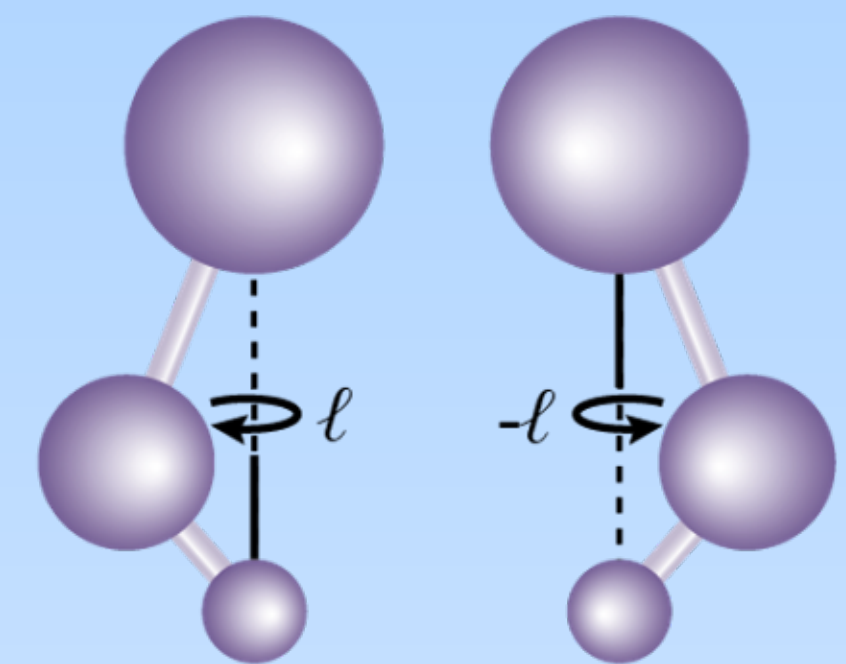




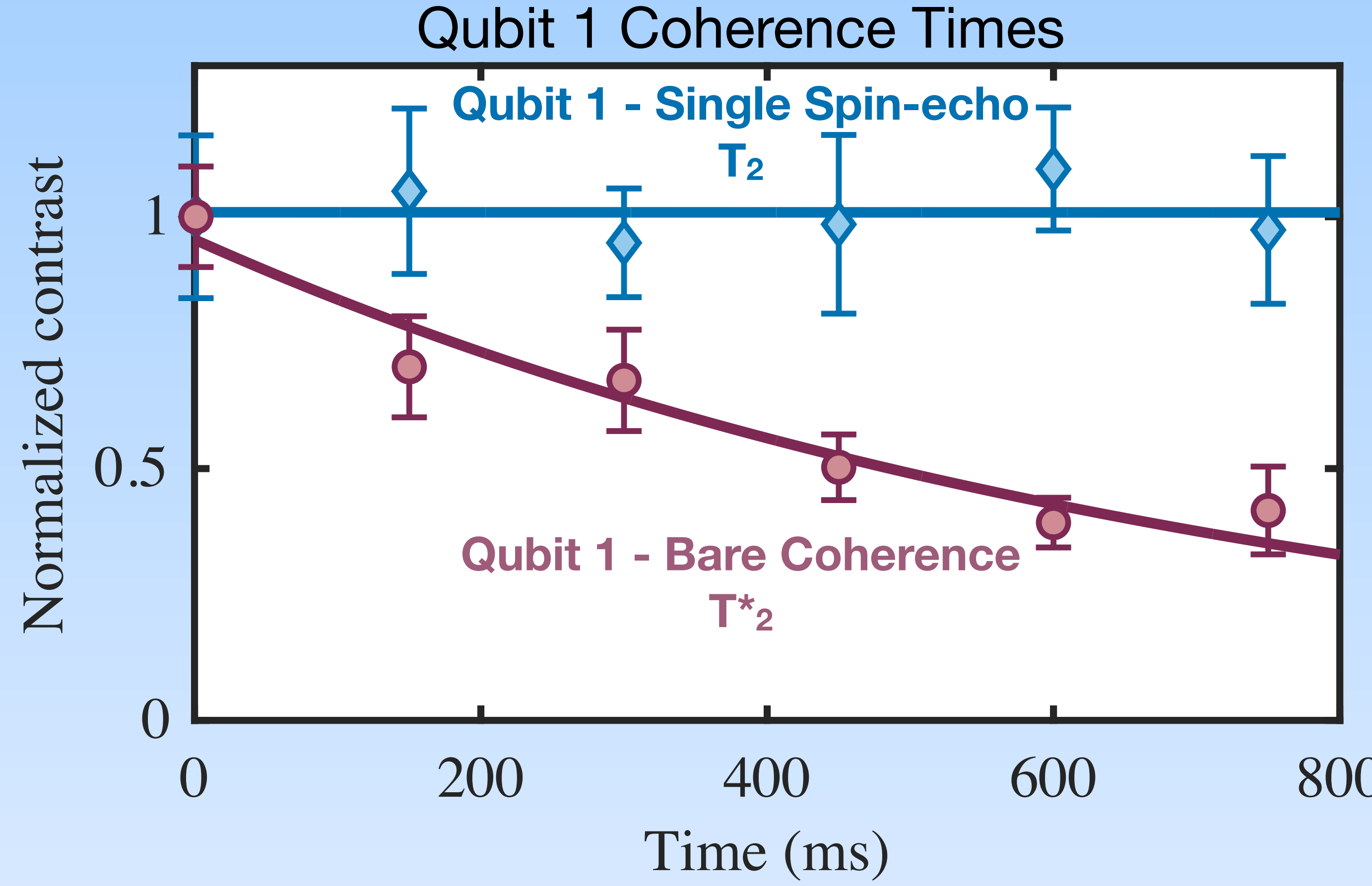
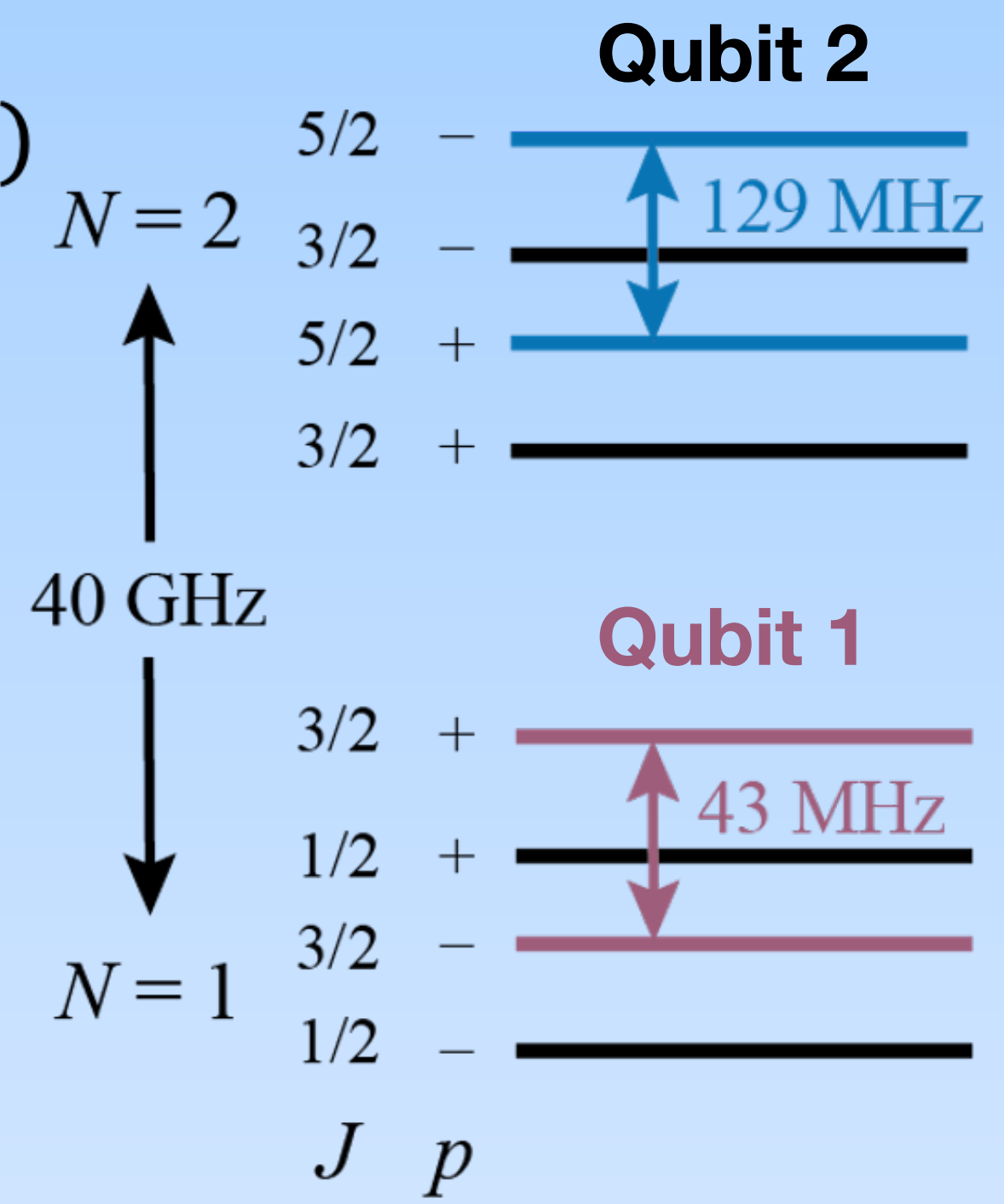
Parity Precession in the CaOH Qubit State

Bending Mode: Qubit States

$$|\pm\rangle = \frac{1}{\sqrt{2}} (|\ell\rangle \pm |-\ell\rangle)$$



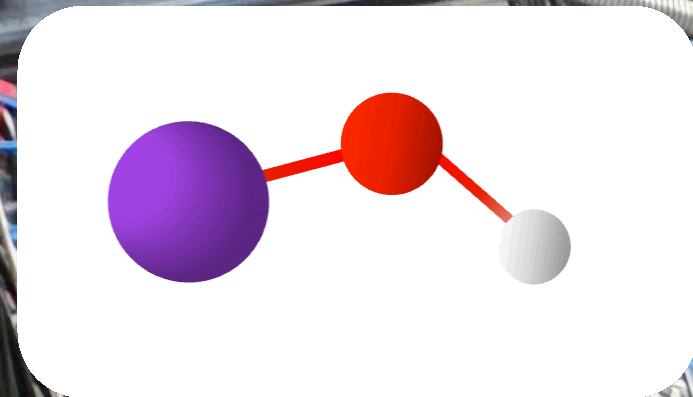
*** bending mode lifetime in our experiment is ~350 ms ***



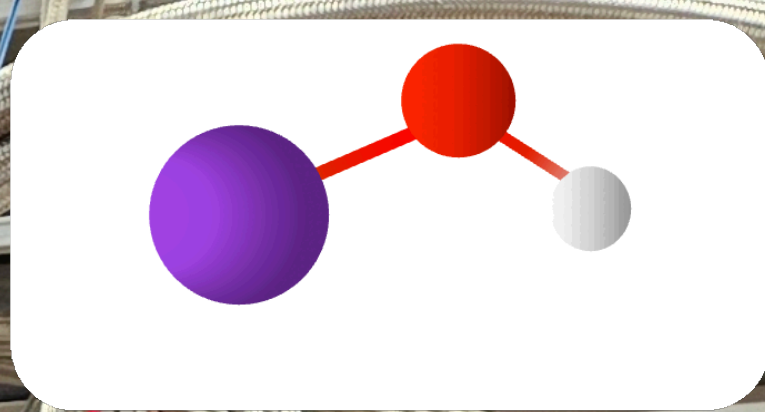
Bare coherence $T^*_2 = 0.8(2)$ s

Spin-echo coherence $T_2 > 2.9$ s

1064 nm optical dipole trap (ODT)



CaOH Current Team



Christian



Giseok



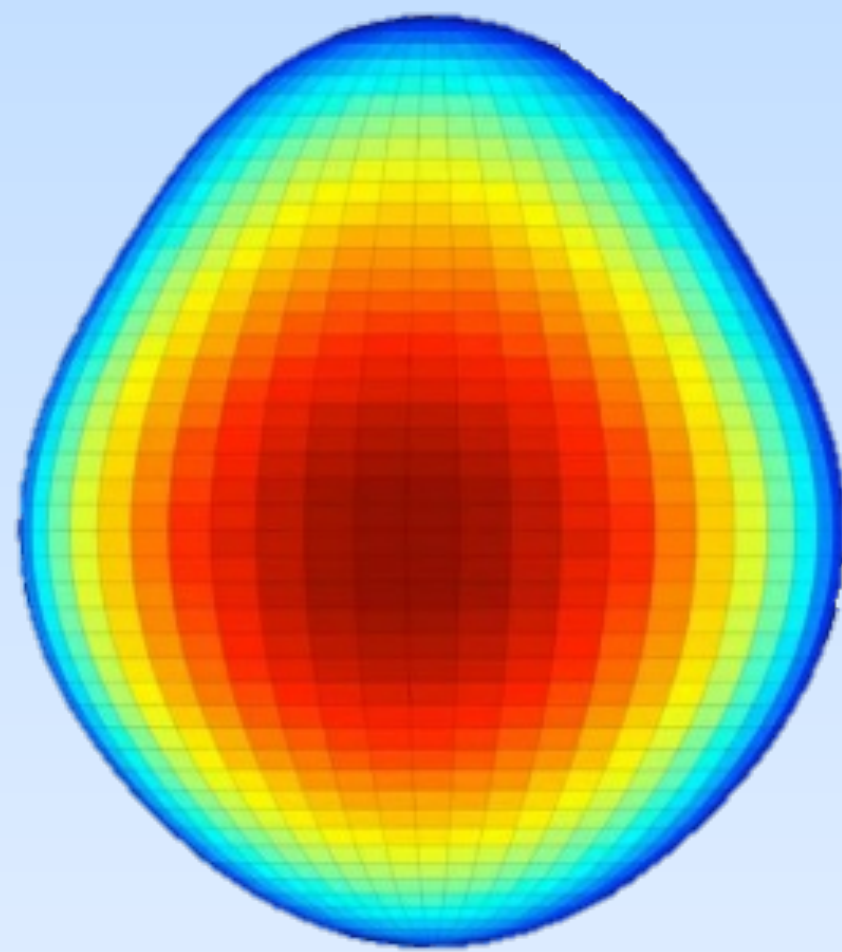
Junheng



Paige

Nuclear CP-violation Enhancement

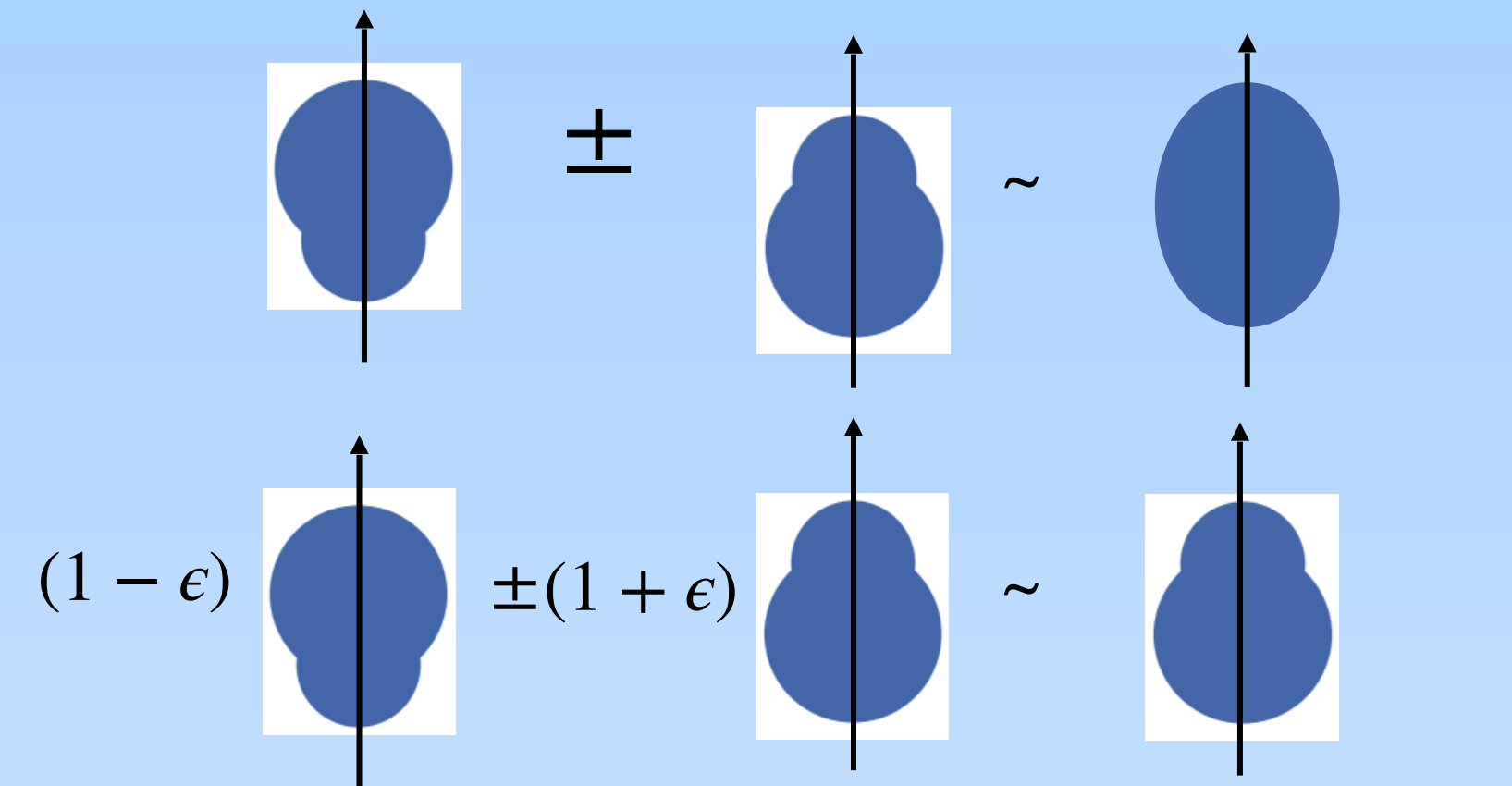
Molecules with Octupole Deformed Nuclei



**Octupole
Deformation “CP-violation Amplifier”**

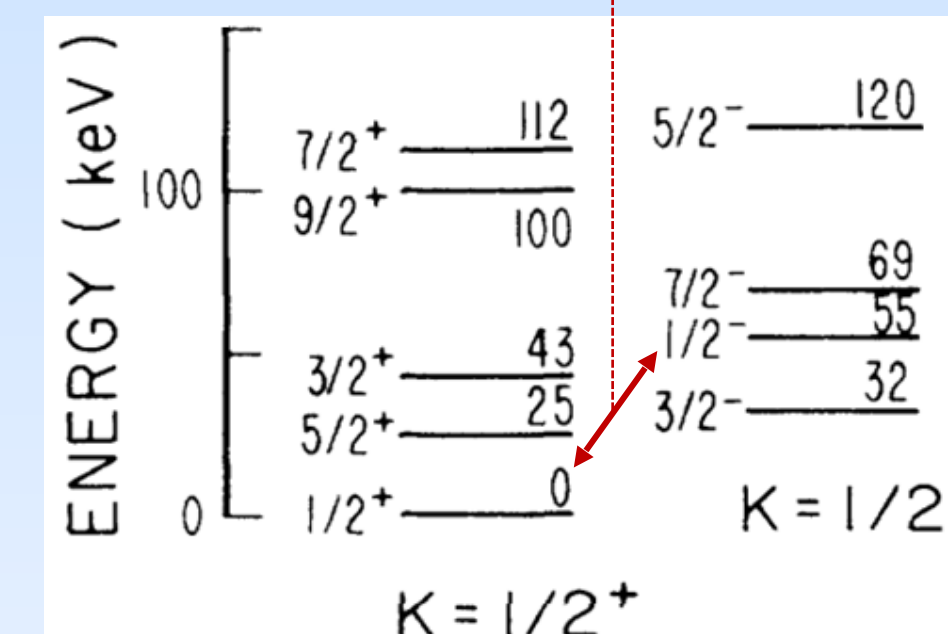
Increased SENSITIVITY to new CP-ν Physics

- In absence of symmetry violation, nucleus should be in superposition
→ octupole shape averages away
- PV, TV nuclear forces can mix these states
- Large charge, mass mismatch from asymmetric shape
- Deformed nuclei often have low-lying opposite parity states, ≈ 100 keV



$$\propto \sum_i \left(r_i^3 - \frac{5}{3} r_i \langle r_{ch}^2 \rangle \right) z_i$$

$$S \approx -2 \frac{\langle \Psi_0 | \hat{S}_0 | \bar{\Psi}_0 \rangle \langle \bar{\Psi}_0 | \hat{V}_{PT} | \Psi_0 \rangle}{\Delta E}$$



N. Auerbach, V. V. Flambaum, & V. Spevak, PRL 76, 4316 (1996)

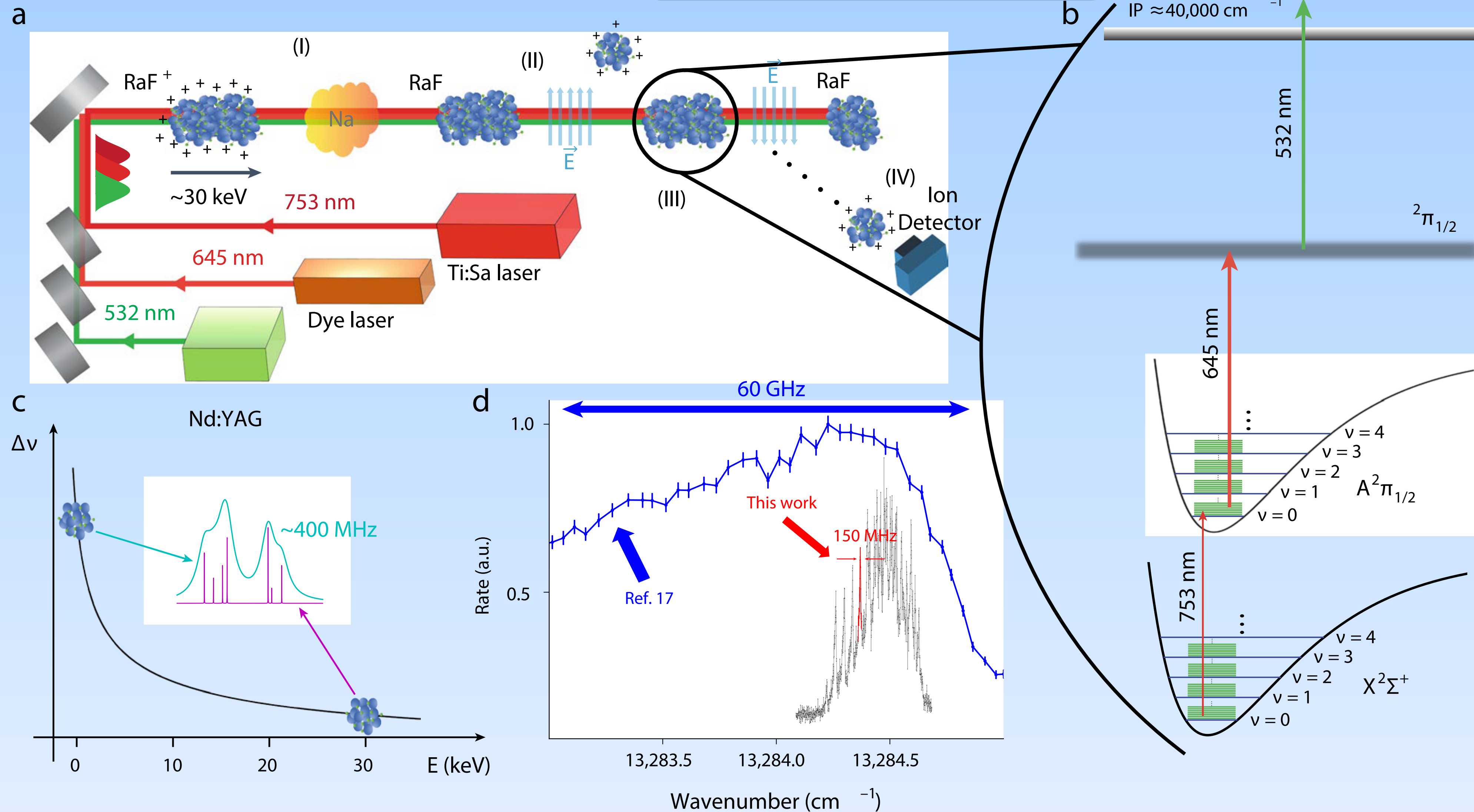
J. Dobaczewski, J. Engel, M. Kortelainen & P. Becker, PRL 121, 232501 (2018), J. Engel et al., PRC 35, 1973R (1987)



RaF at MIT



Ronald Garcia Ruiz
MIT

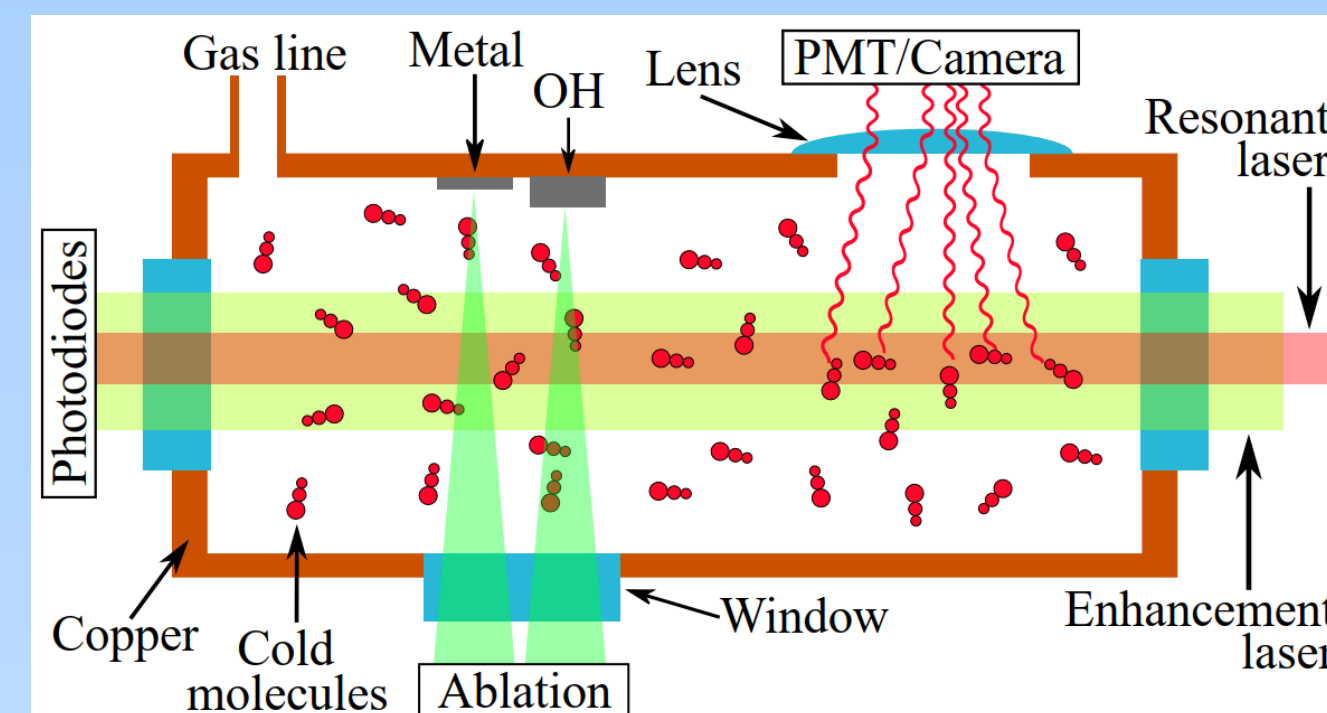
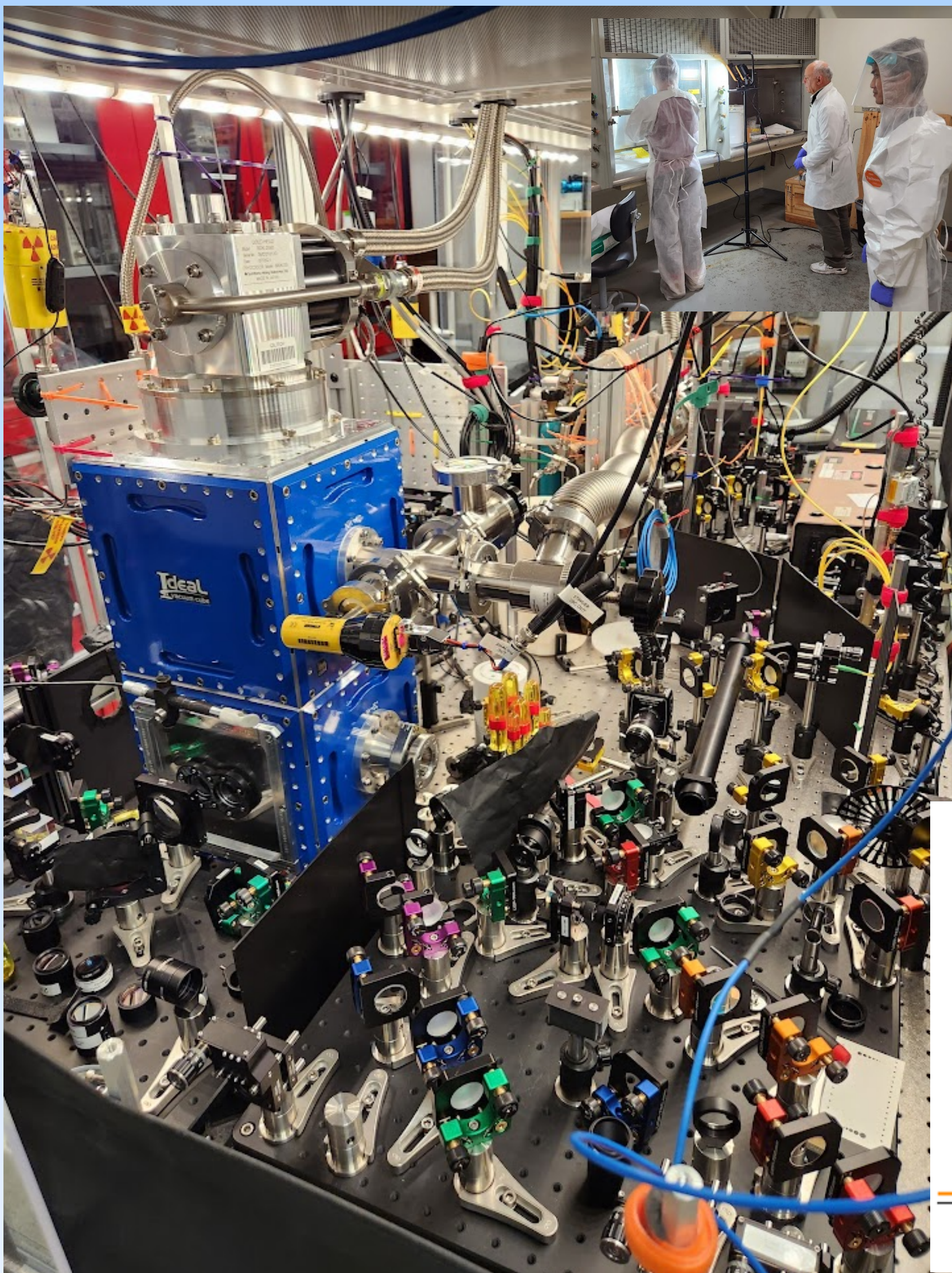


Udrescu, S.M., Wilkins, S.G., Breier, A.A. *et al.* Precision spectroscopy and laser-cooling scheme of a radium-containing molecule. *Nat. Phys.* **20**, 202–207 (2024).

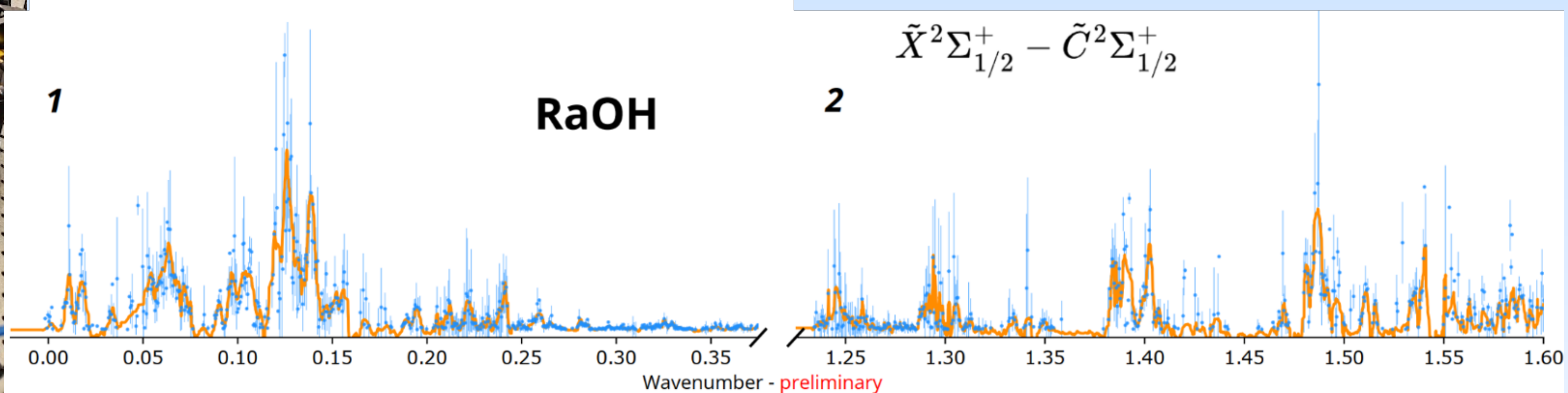
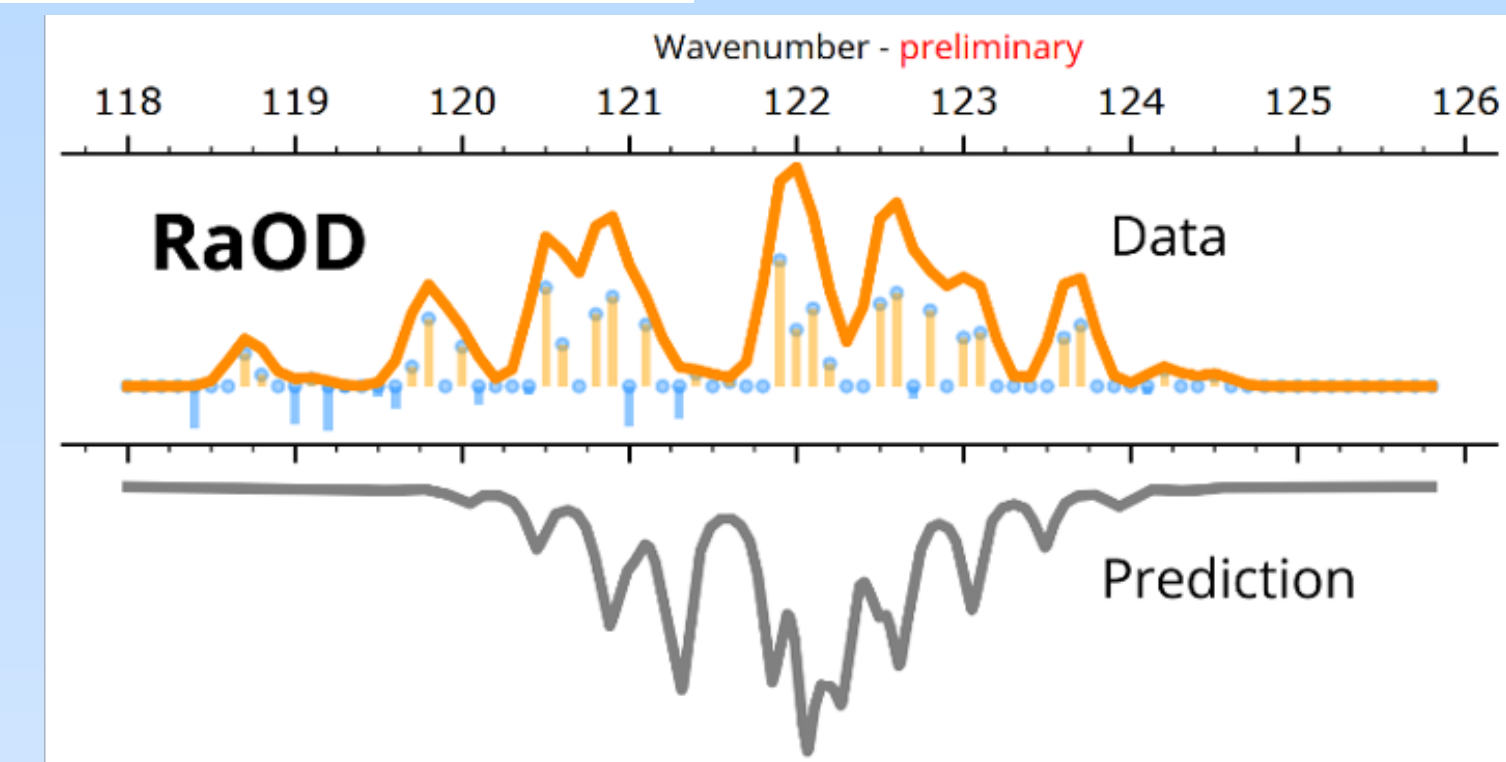
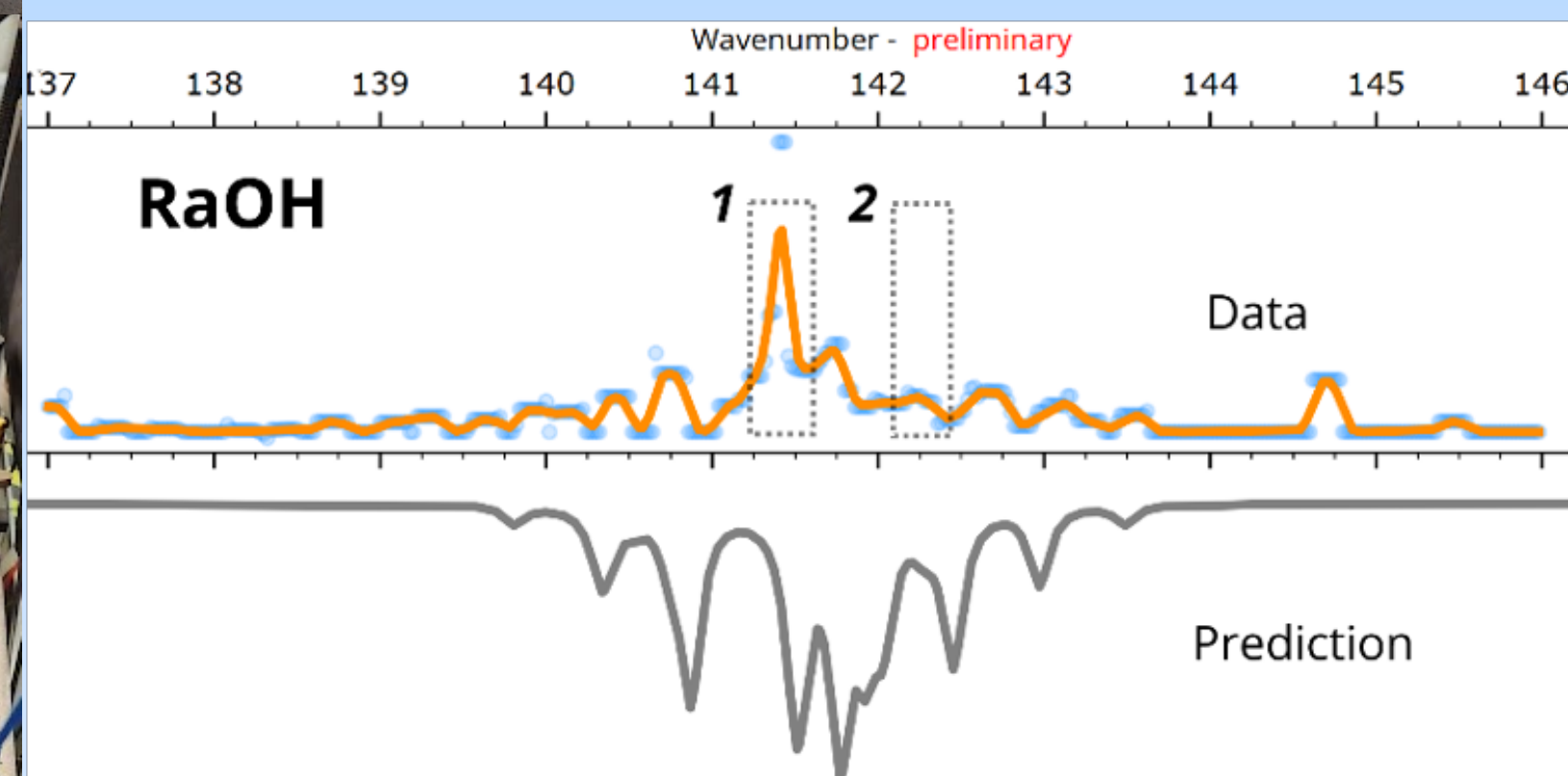


RaOH at Caltech

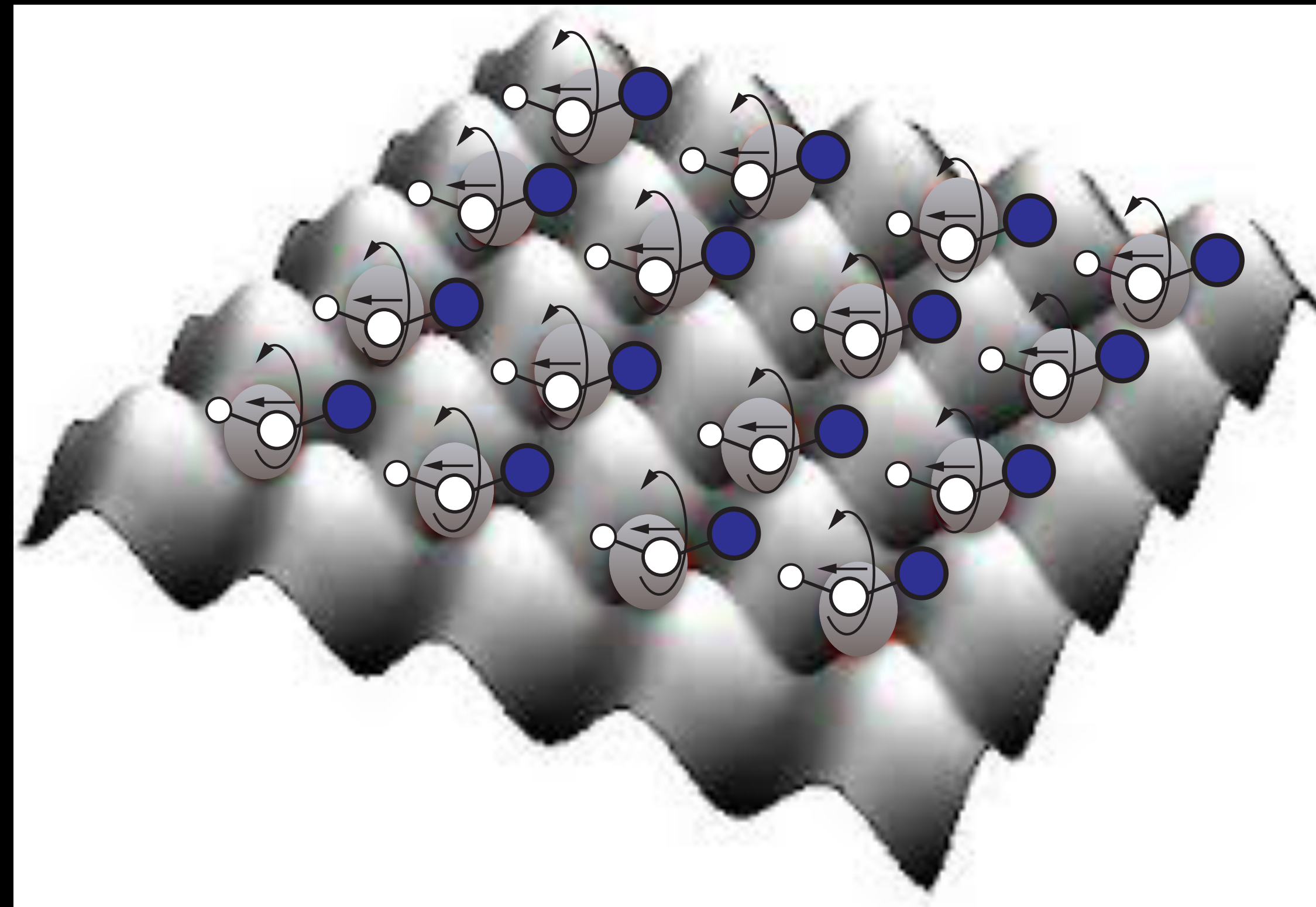
Buffer-gas cooled $^{226}\text{RaOH}$ Spectroscopy



Nick Hutzler
Caltech



But we need them ultra cold
To load into an optical lattice

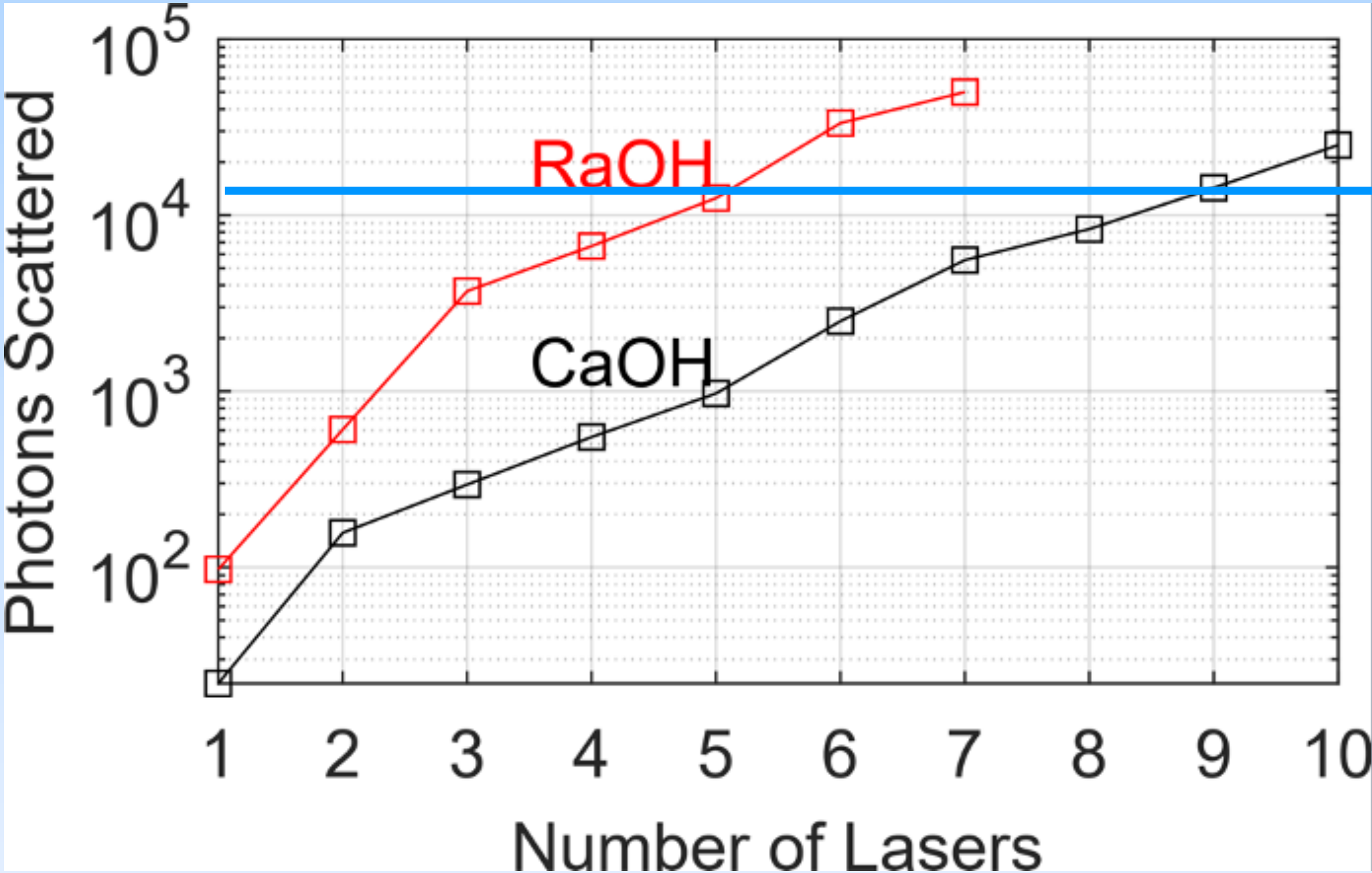
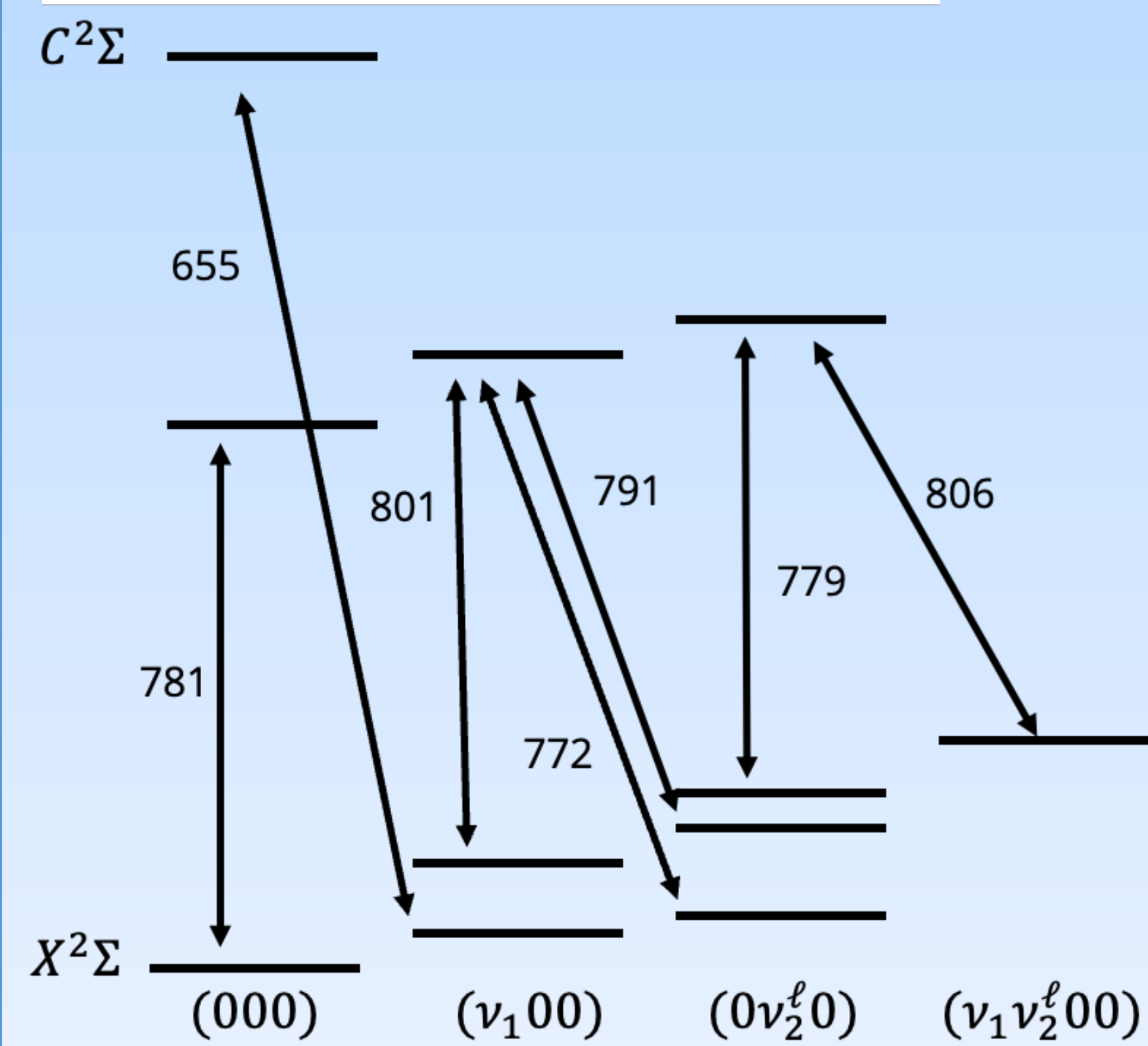


MOT
etc.

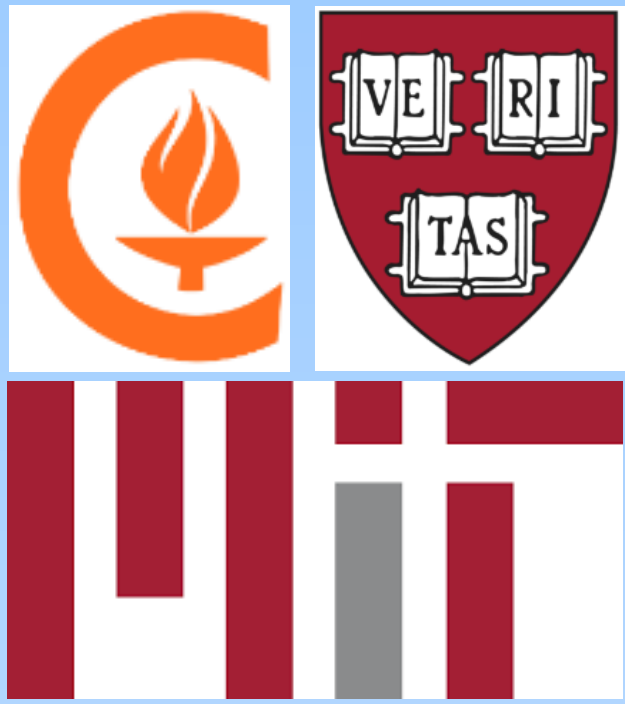
RaOH		
State	VBR (%)	Photons
(000)	98.972	97
(100)	0.863	606
(02 ⁰ 0)	0.138	3704
(200)	0.012	6667
(02 ² 0)	0.007	12500
(12 ⁰ 0)	0.005	33333
(010)	0.001	50000*

Laser Cooling of RaOH, theory

Easier than SrOH and CaOH it appears



MOT



RaX Collaboration



Plan

(RaOH and/or RaF, RaOCH₃)

Laser slow $\rightarrow$ MOT $\rightarrow$ ODT

Nuclear Schiff Moment measurement
~1 s coherence time, ~1000 molecules

“RaX Collaboration”



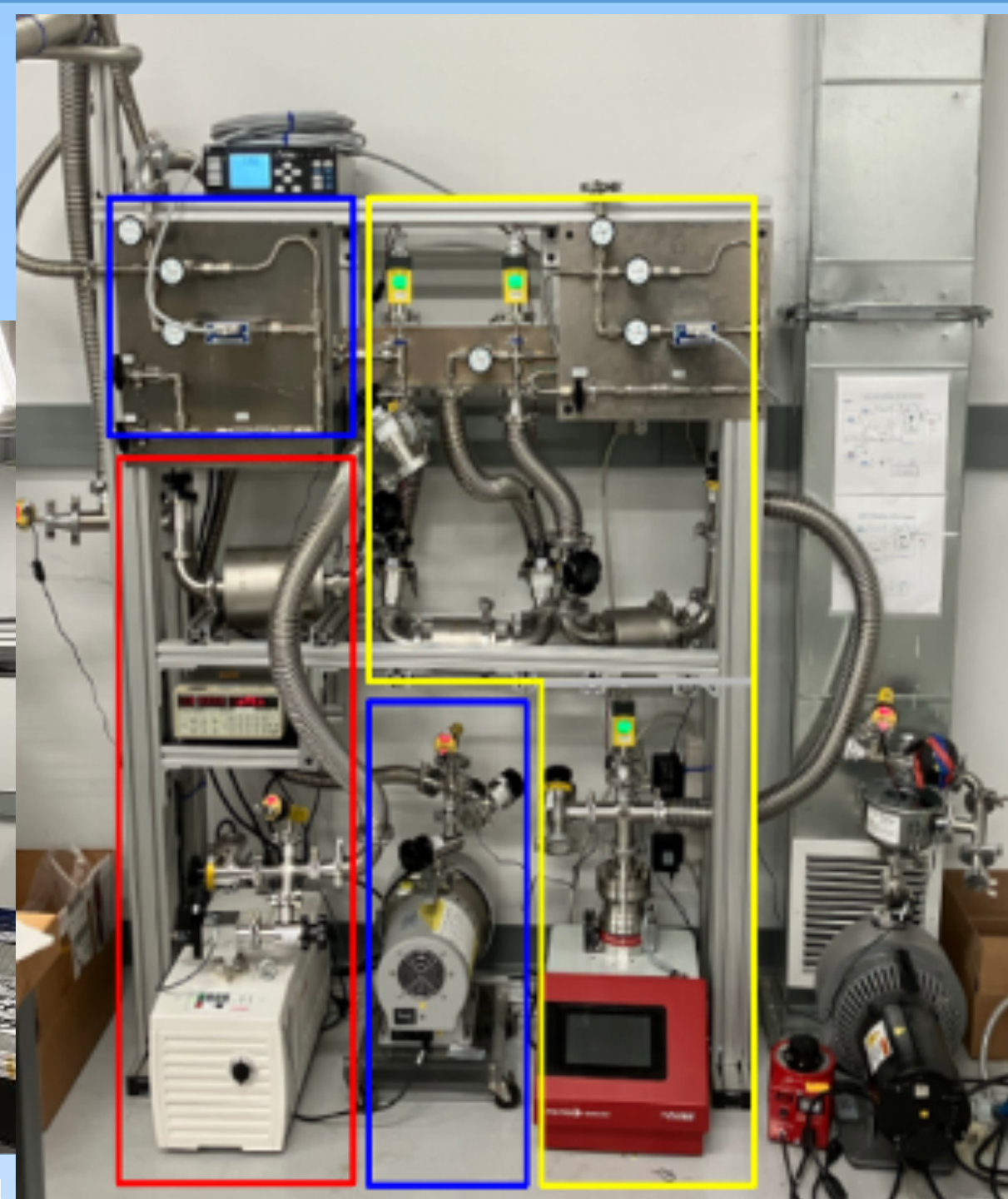
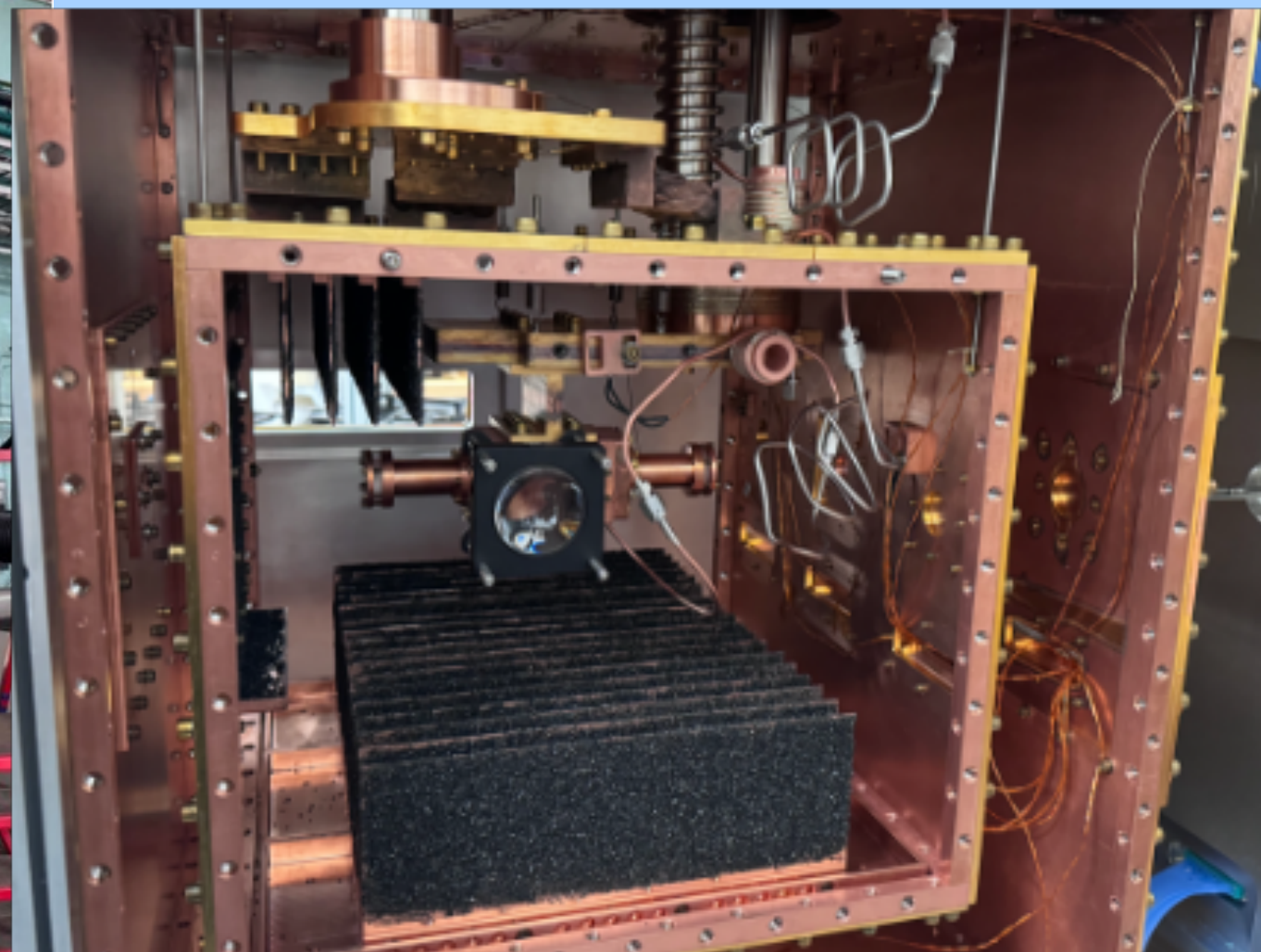
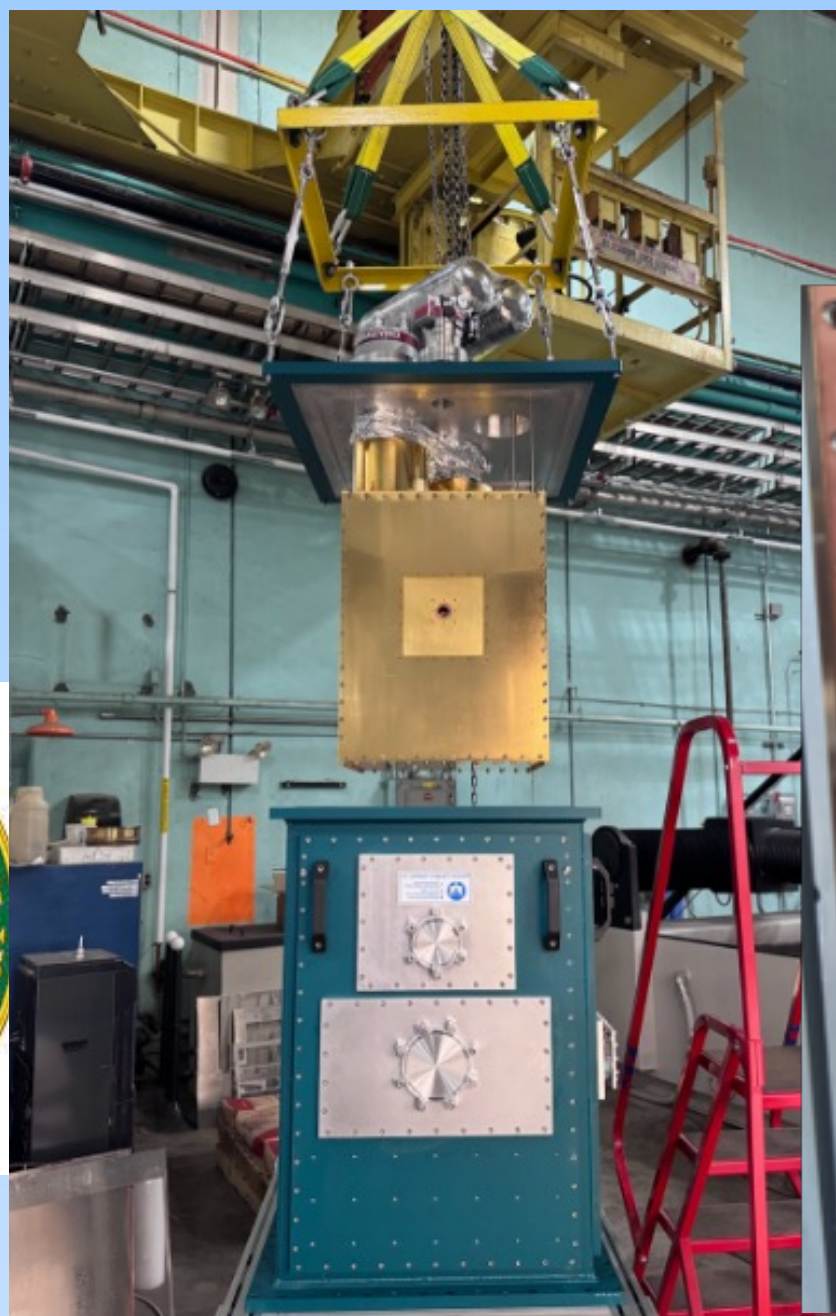
Nick Hutzler
Caltech



Ronald Garcia Ruiz
MIT



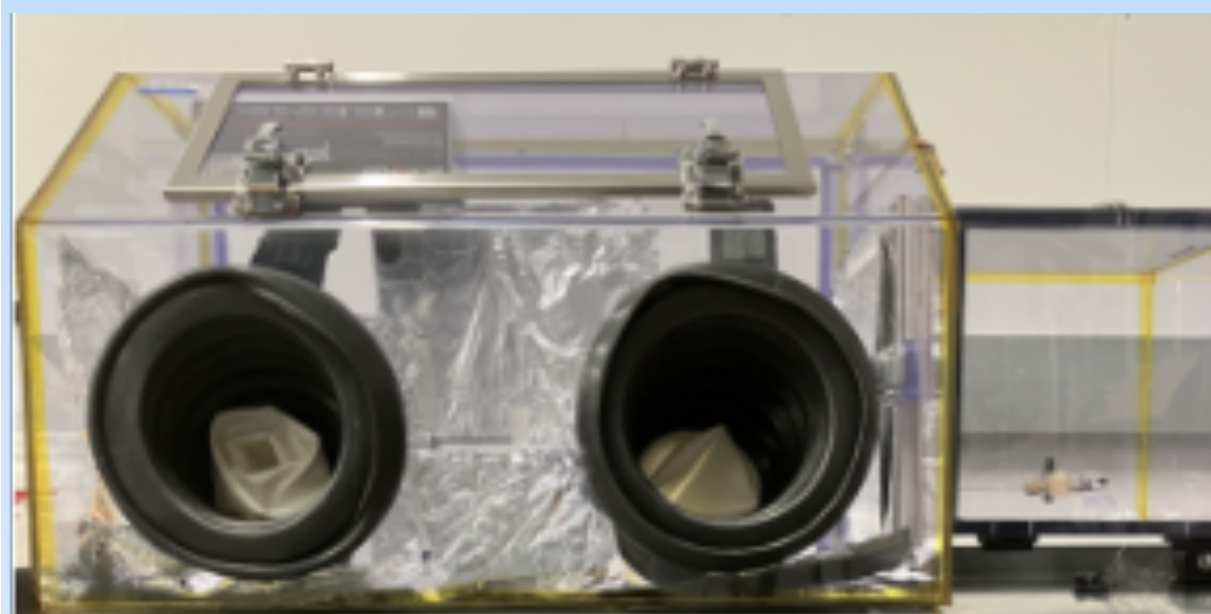
RaX - Experimental Setup



April 2025

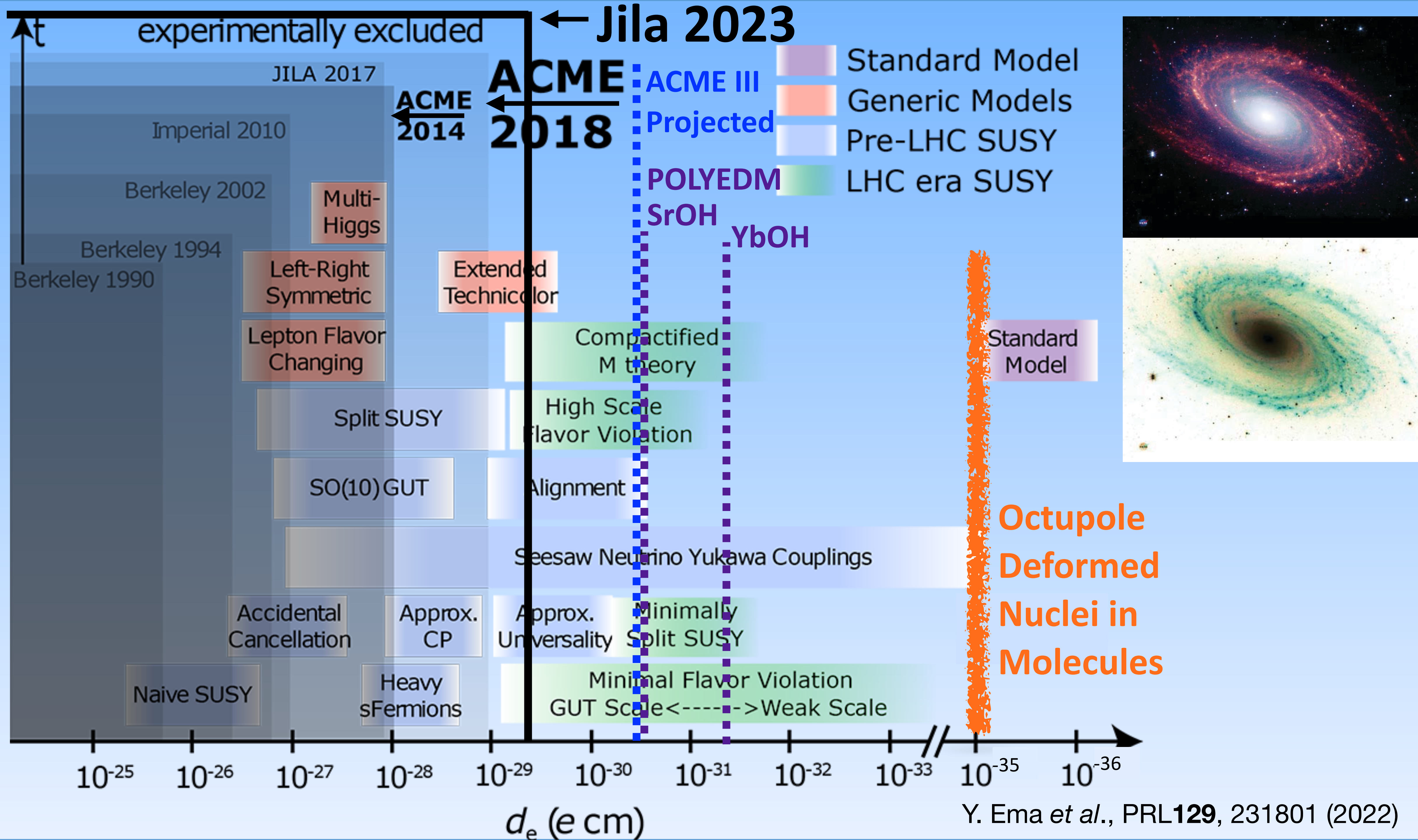


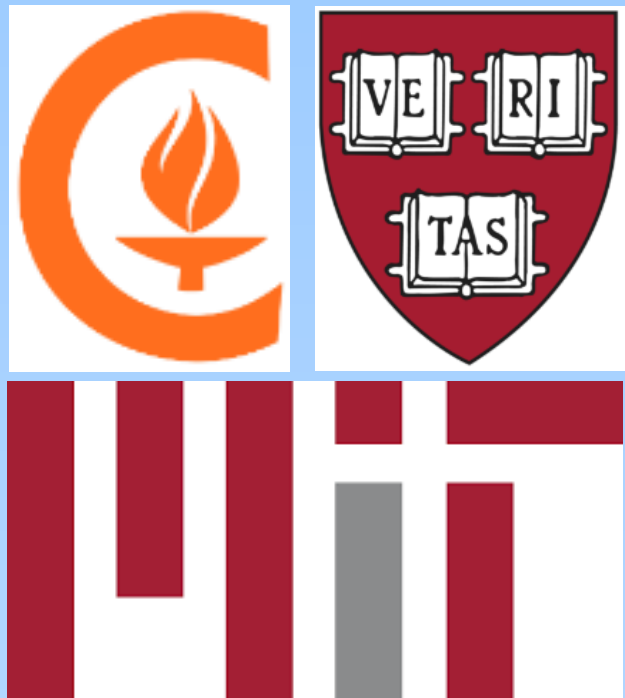
October 2025



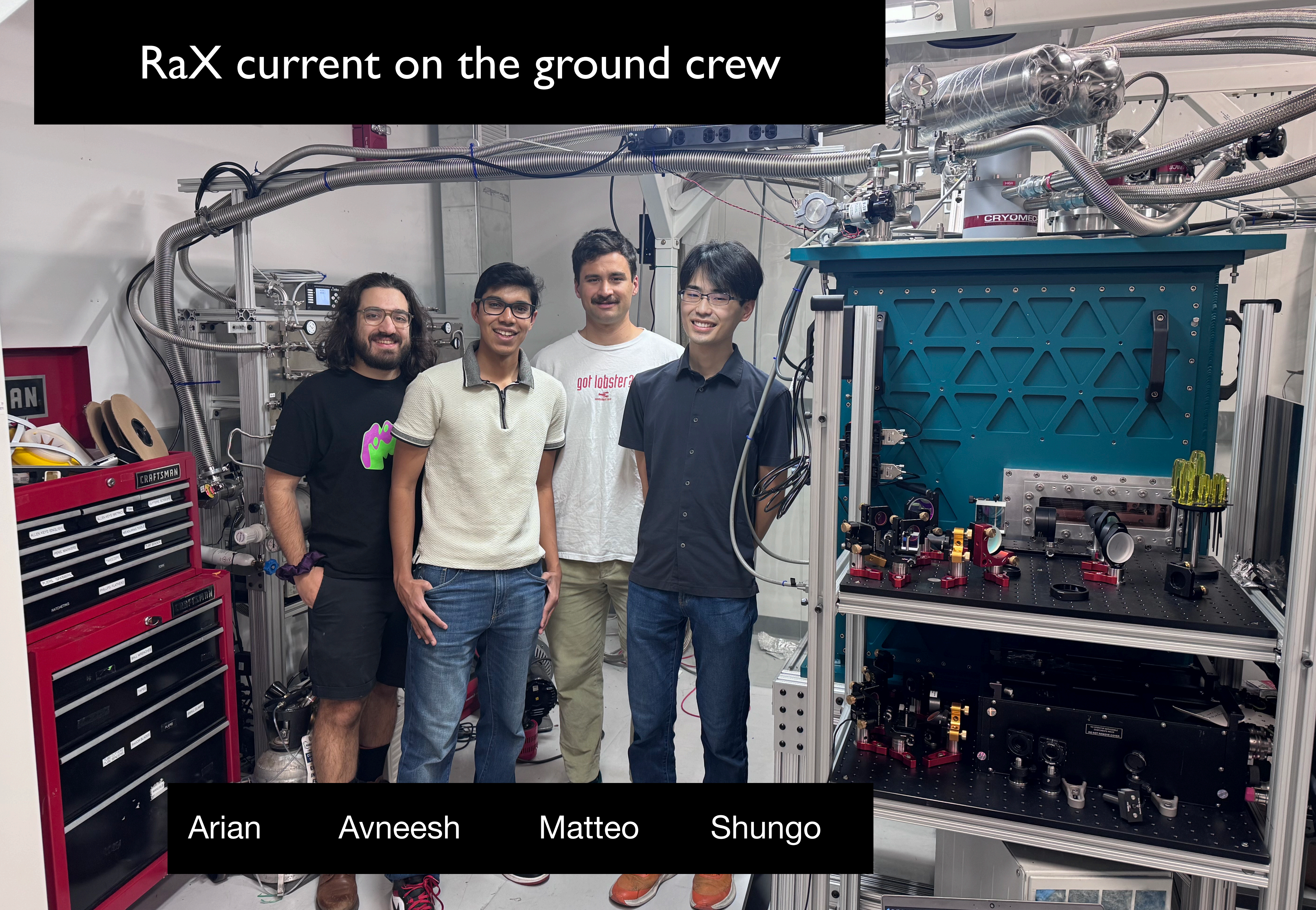
Radon Monitor
AlphaGuard







RaX current on the ground crew



Arian

Avneesh

Matteo

Shungo

arXiv > hep-ph > arXiv:2509.23531

High Energy Physics – Phenomenology

[Submitted on 27 Sep 2025]

Electric Dipole Moments and New Physics

On ArXiv
Last week

Electric Dipole Moments and New Physics

Maxim Pospelov,^a and Adam Ritz^b

^aUniversity of Minnesota, William I. Fine Theoretical Physics Institute, School of Physics and Astronomy, Minneapolis, MN 55455, USA

^bUniversity of Victoria, Department of Physics and Astronomy, Victoria, BC V8P 5C2, Canada

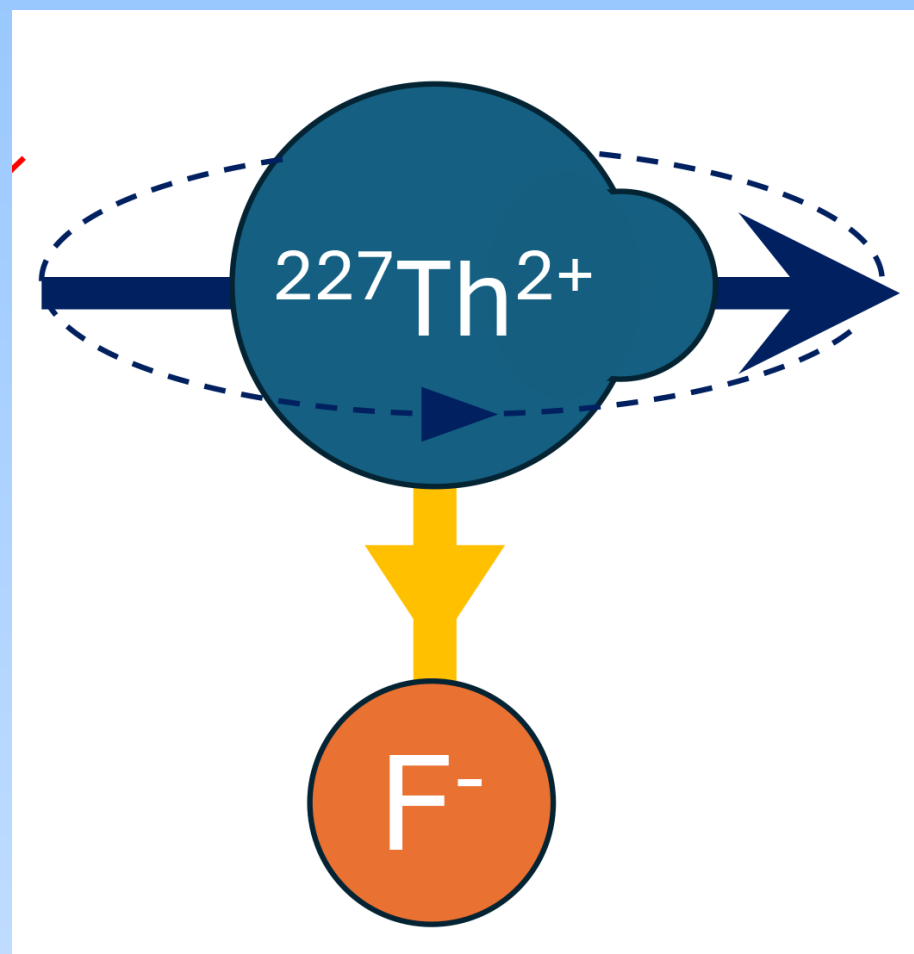
t. Models that induce such interactions typically contain additional BSM particles charged under the SM groups which generically allow for CP -violating interactions. As one prominent application, the sensitivity of current EDM observables to TeV scale new physics, even when EDMs are generated only at the two-loop level, exerts pressure on viable models of electroweak baryogenesis, excluding a large fraction of model-space. The predominant pre-LHC paradigm for new high-scale physics, weak-scale supersymmetry, is now generically pushed to scales above 10 TeV. Current EDM sensitivity reaches these scales, and may extend as high as 100 TeV and above in cases of generic flavor non-universality. At this moment in time, EDMs are one of the few tools available to probe such high-scale models of physics beyond the EW scale. We also reviewed the implications of current EDM sensitivity for new physics in light dark sectors (*e.g.* dark photons, sterile

Lots of new collaborations, new experiments!

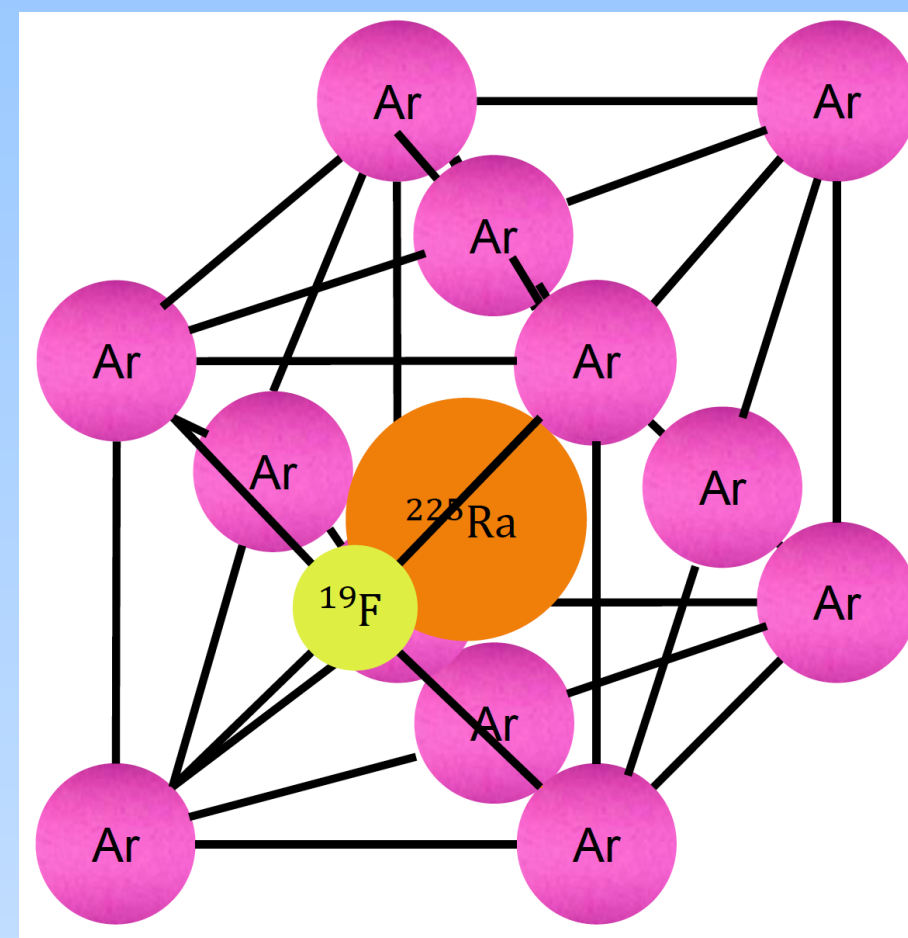
Use molecules that have nuclei x100-1000 more sensitive to CP- ν BSM physics

Octupole Deformed Nuclei/Nuclear Schiff Moment

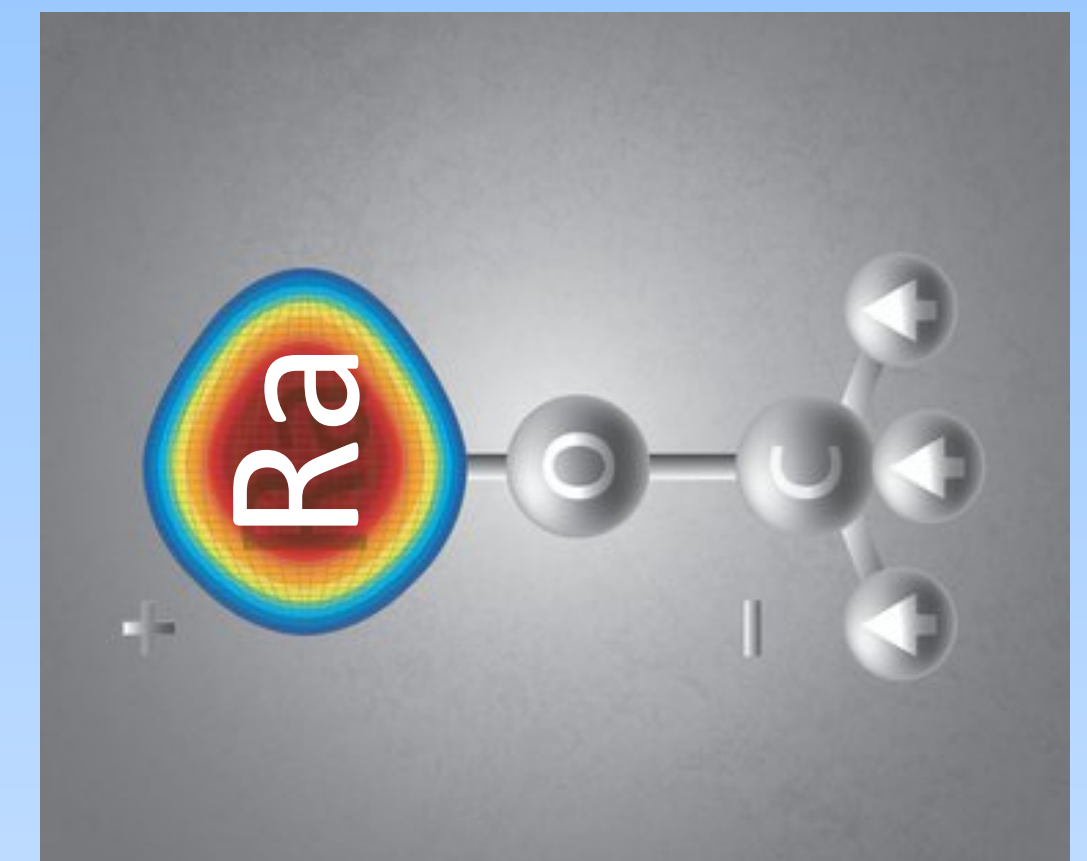
BUT want to keep long coherence times (~ 10 s)!



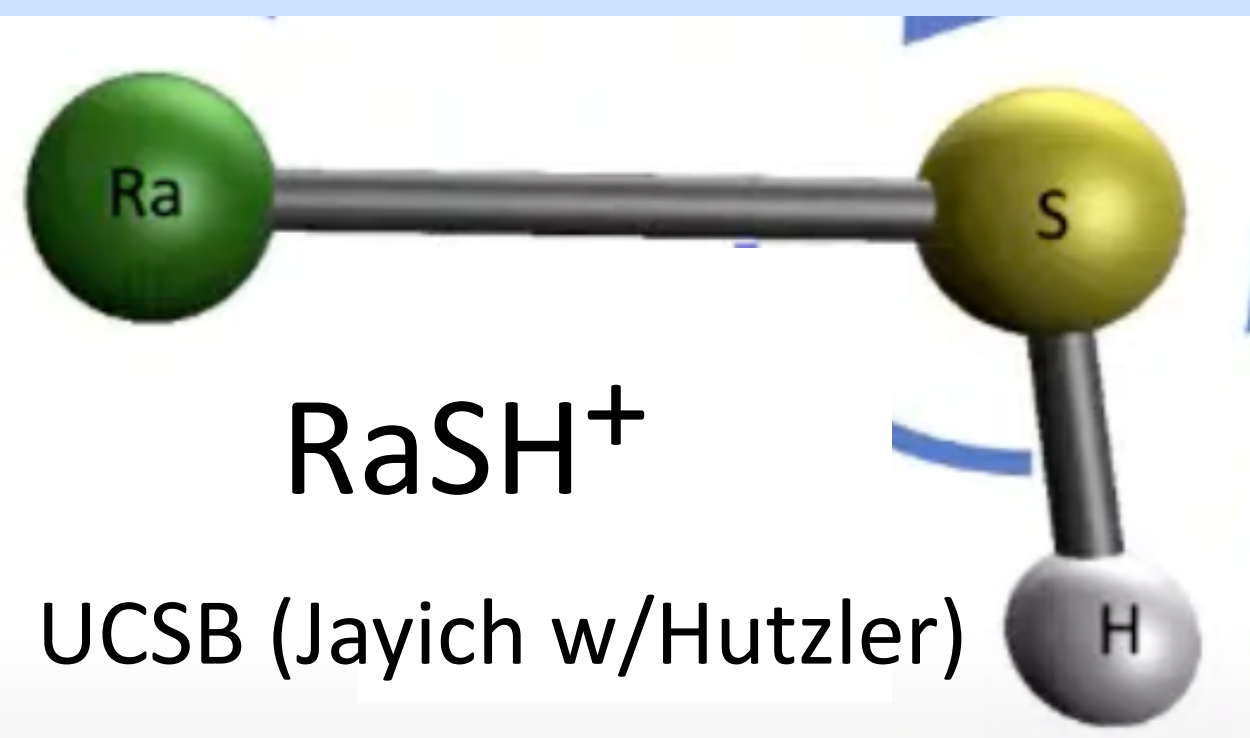
$^{227}\text{ThF}^+$ (Fan/Cornell/Ng/Malbrunot)



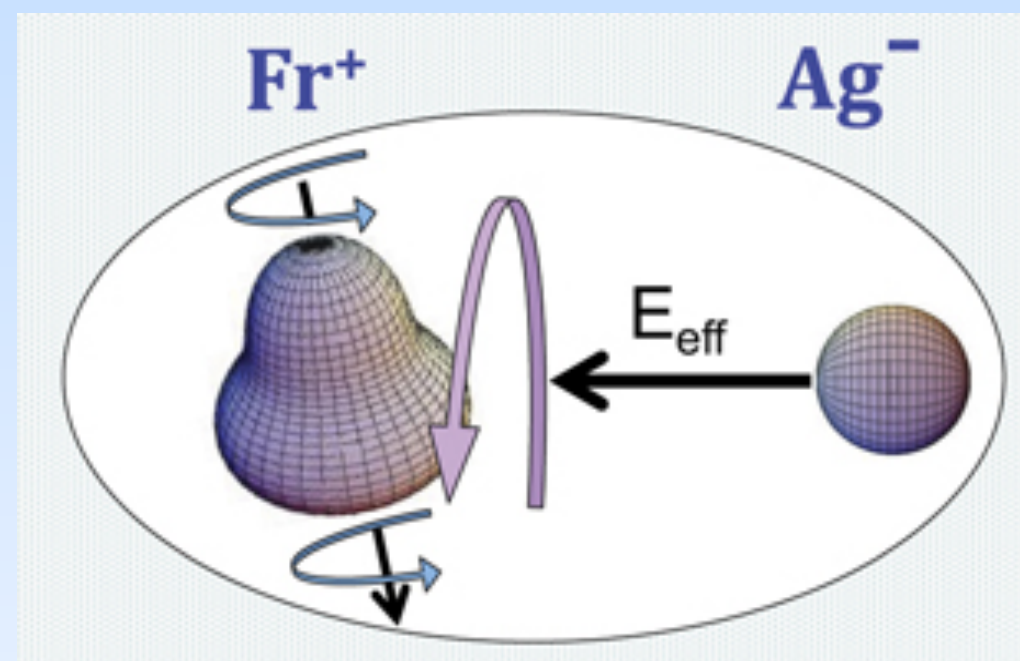
EDM_{cubed} (Vutha and collaborators)



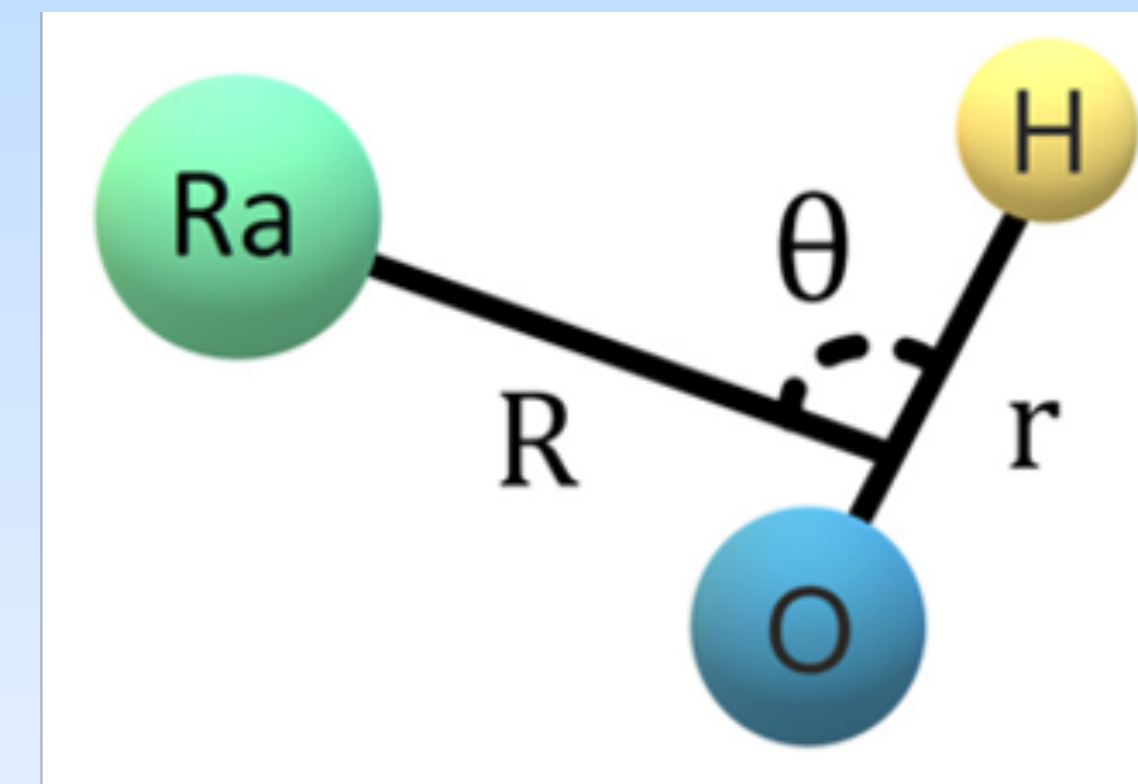
QuEST (Wu)



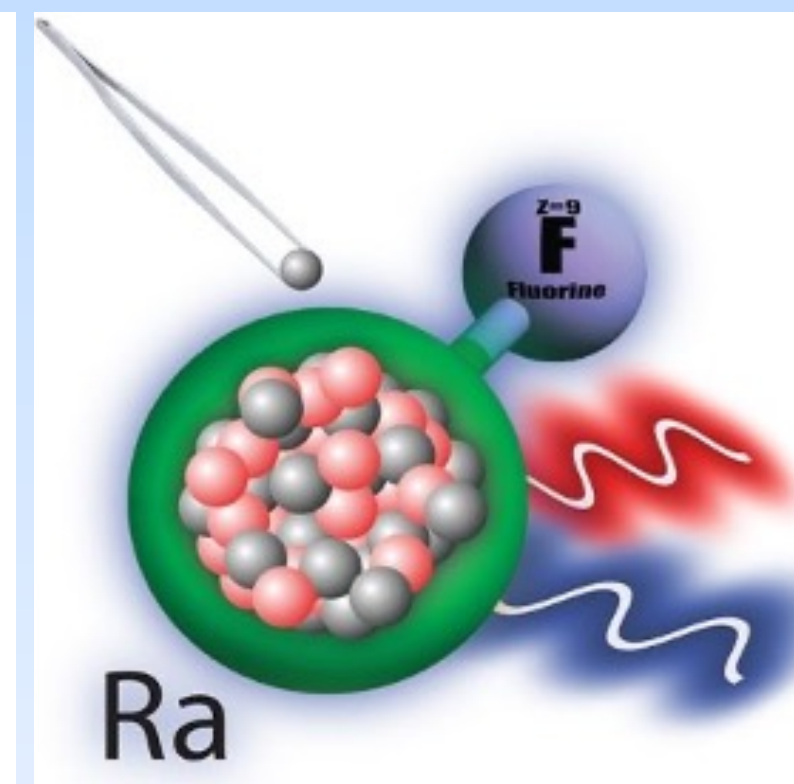
UCSB (Jayich w/Hutzler)

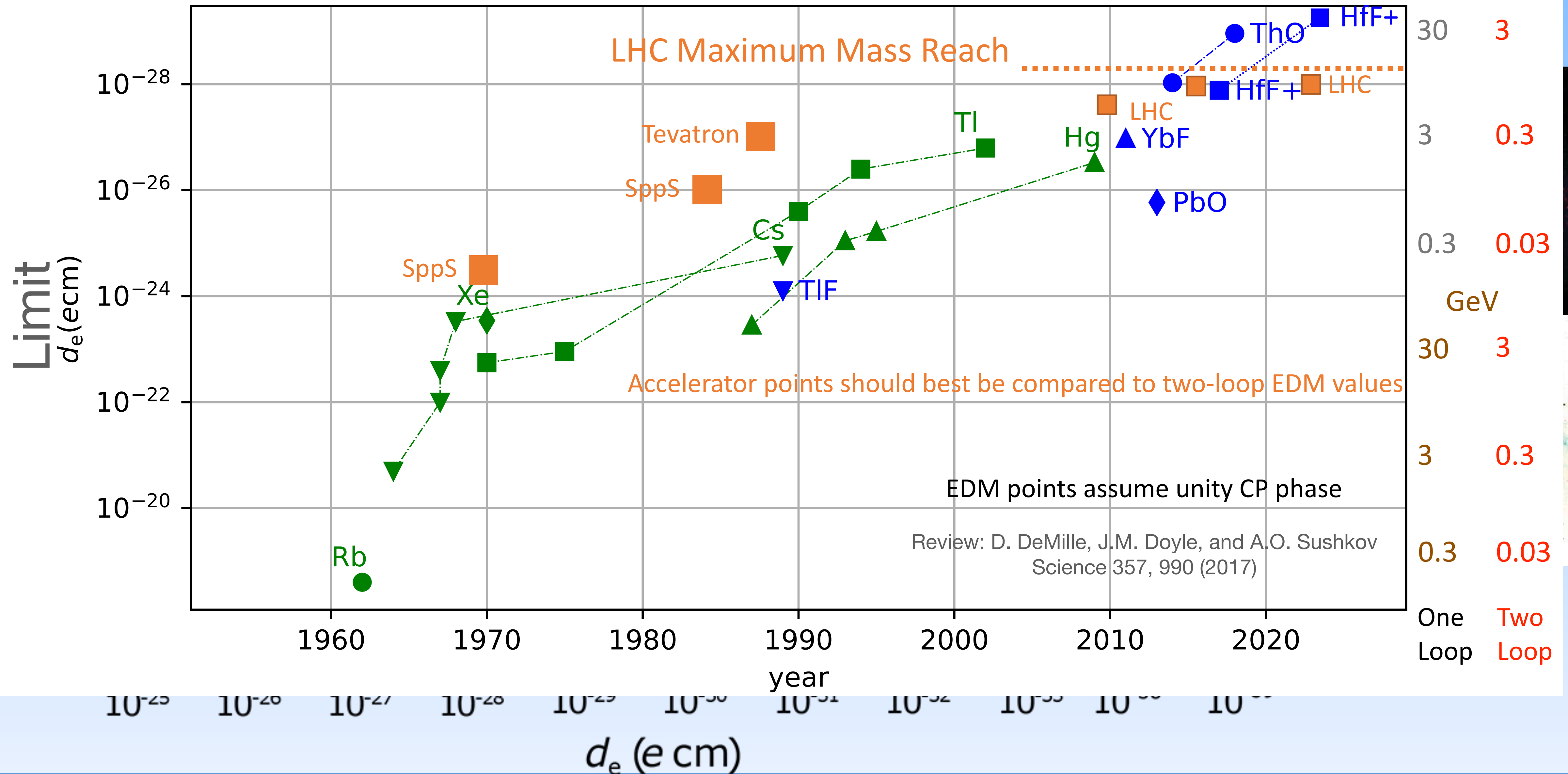


REDRUM (DeMille and collaborators)



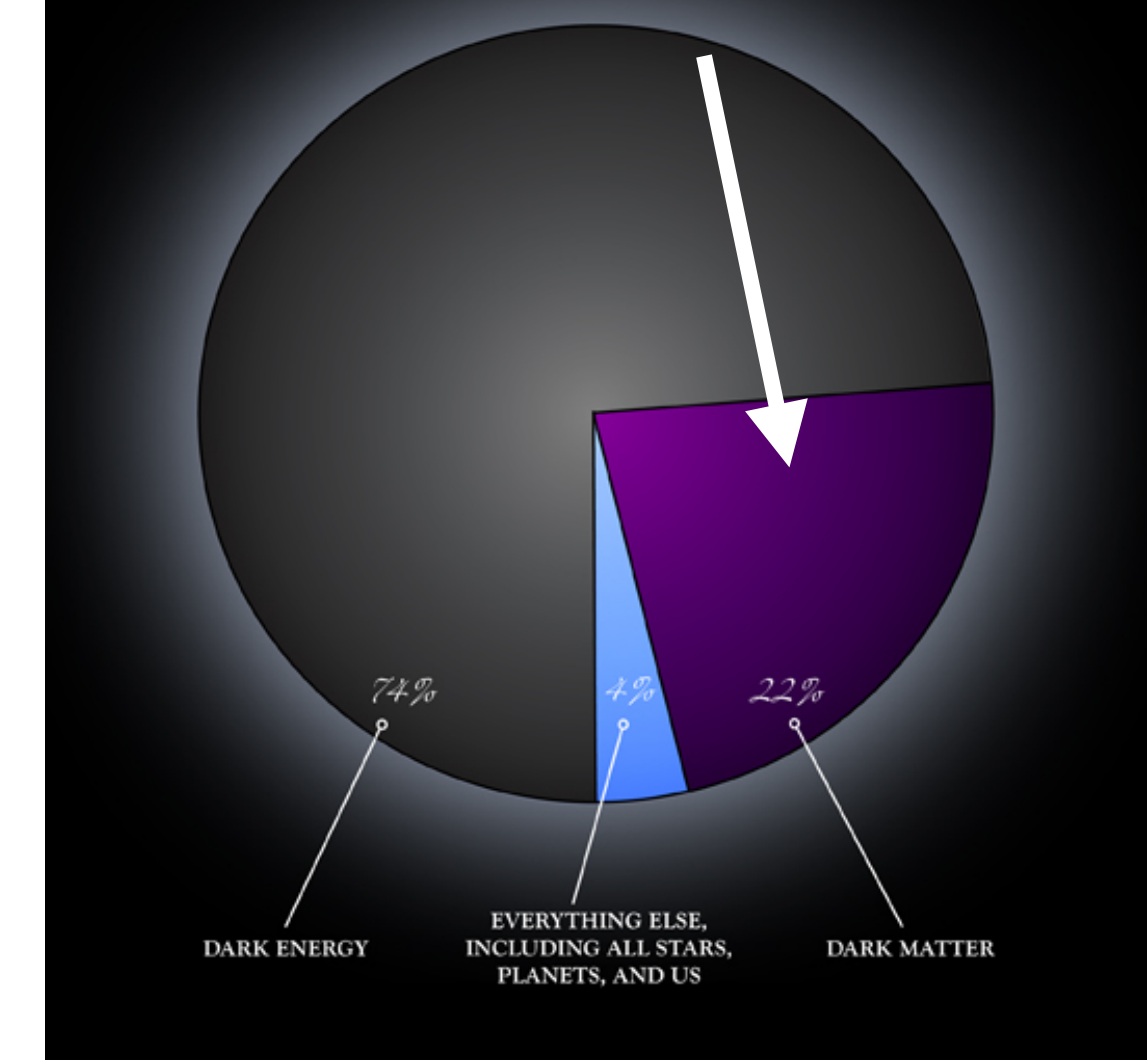
RaX (Hutzler/Doyle/Garcia Ruiz)



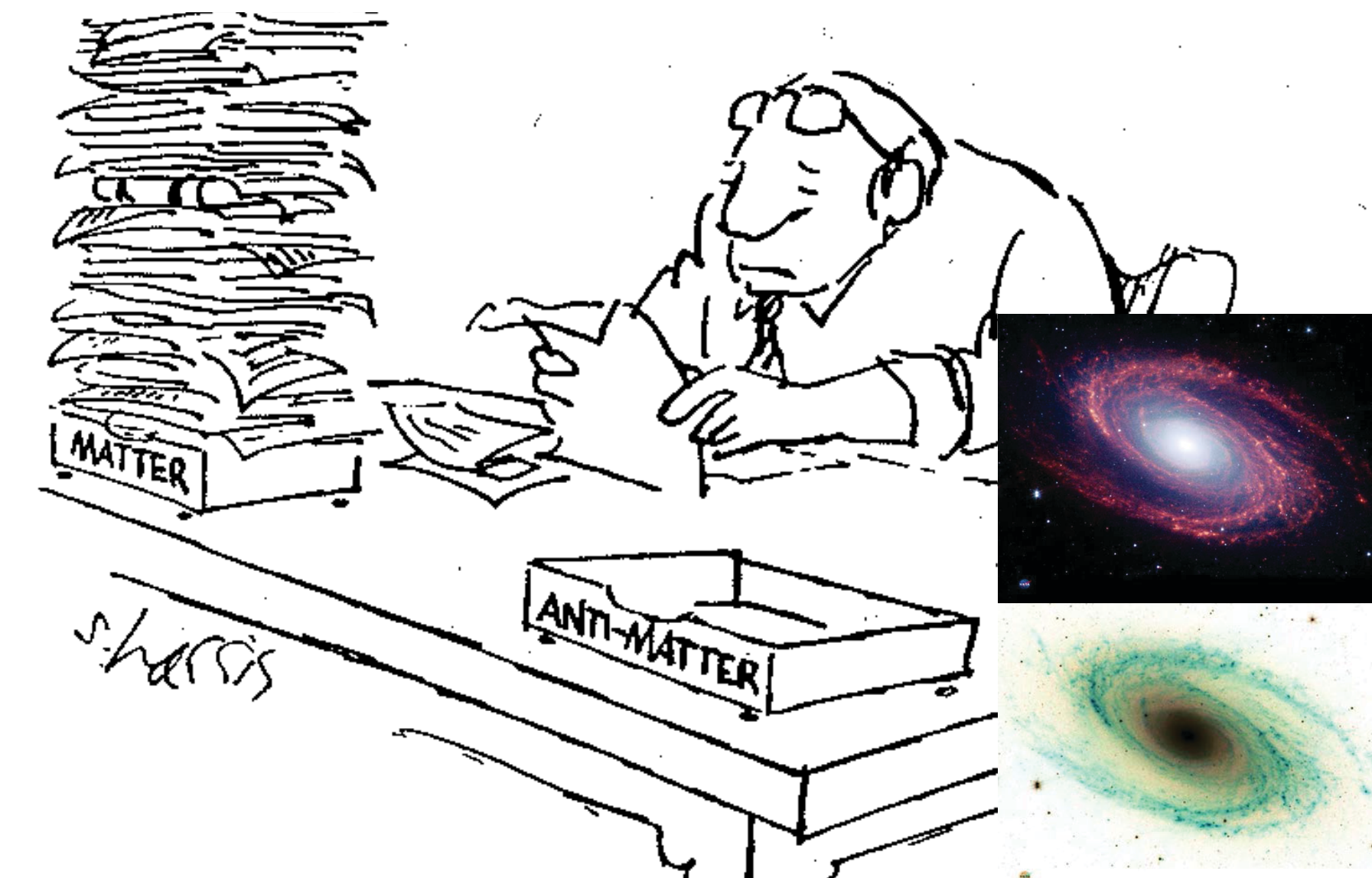


Two Mysteries

1. Dark Matter



2. Matter-Antimatter Asymmetry



YOU ARE HERE

ACCELERATING EXPANSION

A little more than 5 billion years ago, dark energy caused the universe to expand increasingly fast.

INFLATION

In less than 10^{-30} of a second after the Big Bang, the universe burst open, expanding faster than the speed of light and flinging all the matter and energy in the universe apart in all directions.

BIG BANG

The universe expanded violently from an extremely hot and dense initial state some 13.7 billion years ago.



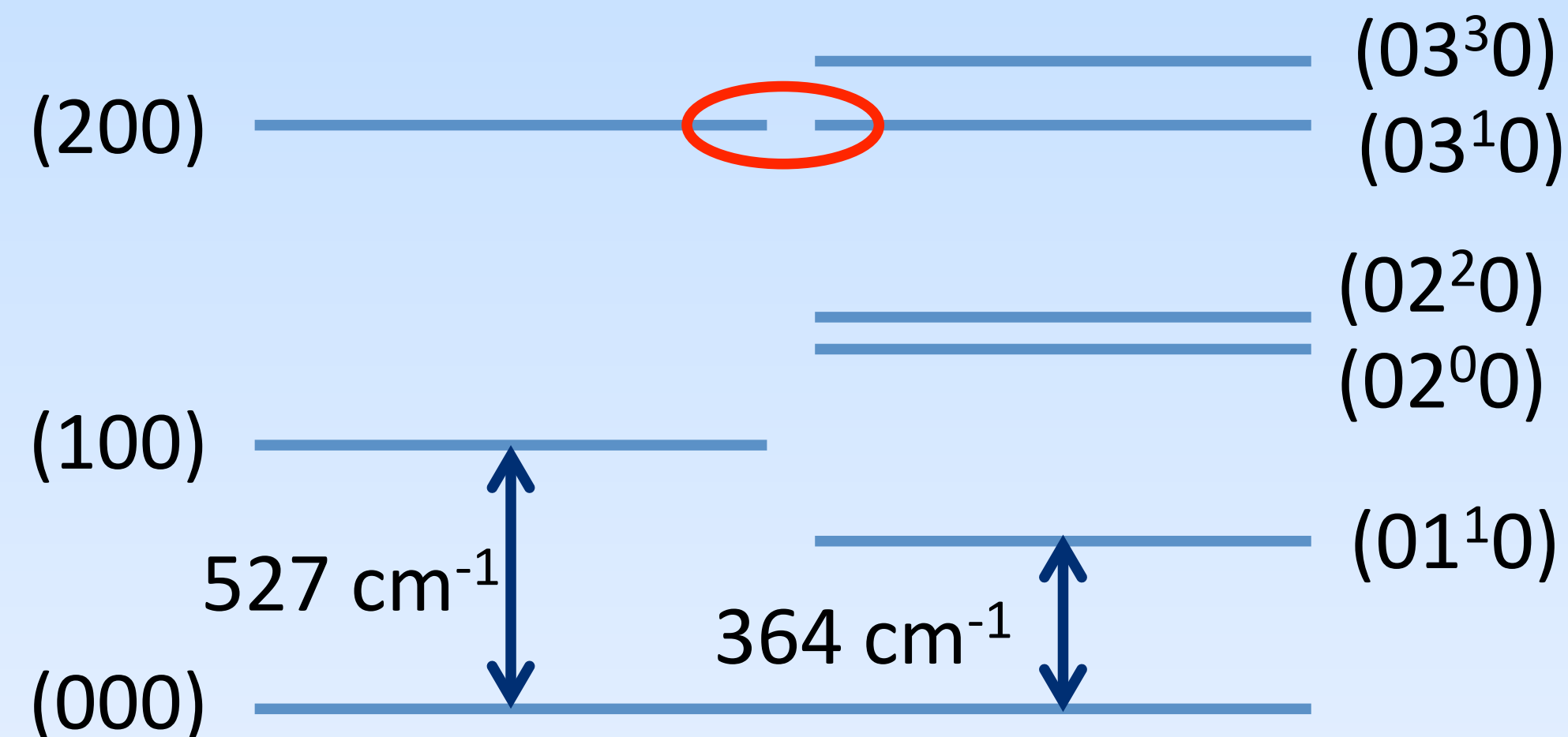
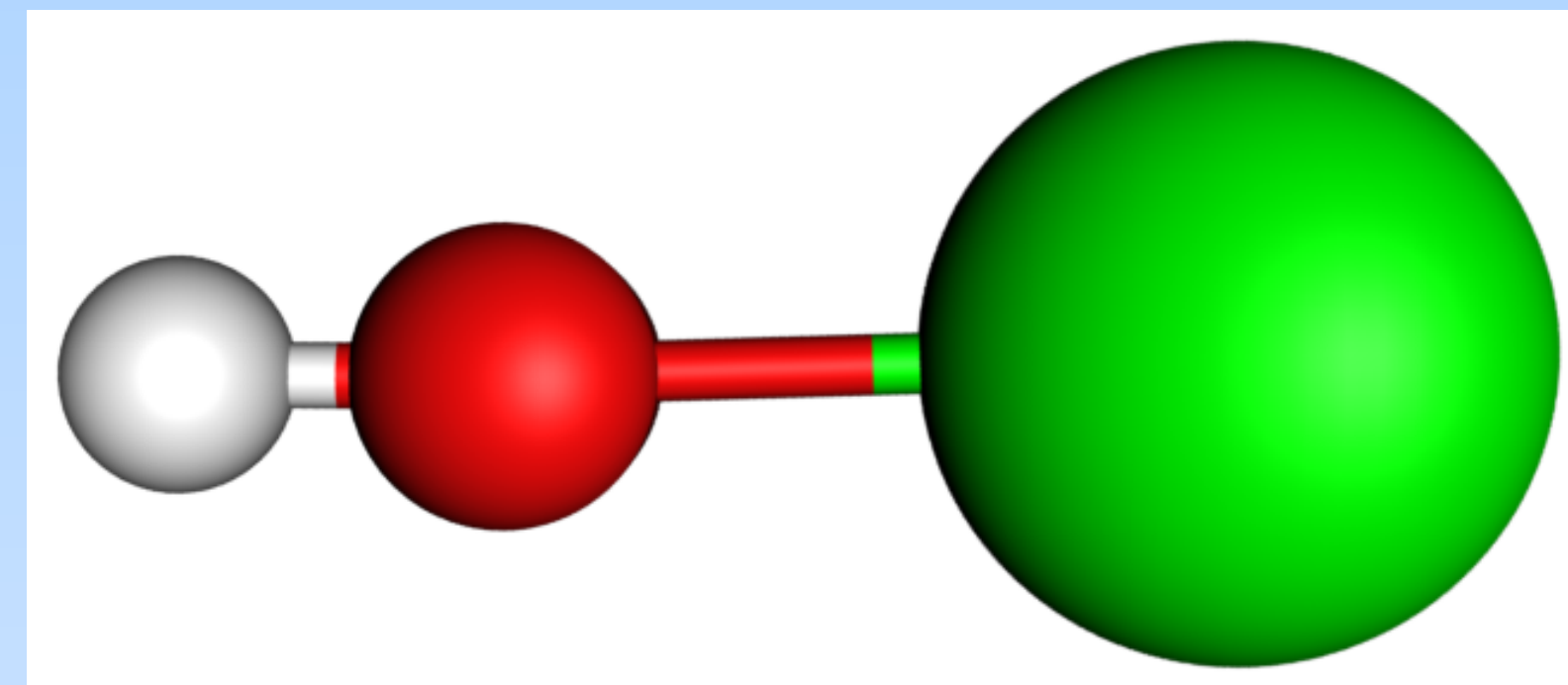
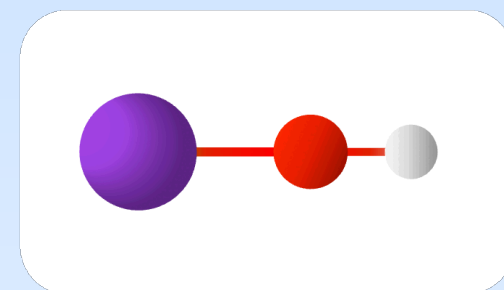
Ultracold polyatomic molecules - Complexity to search for Dark Matter

Dark Matter
Experiment

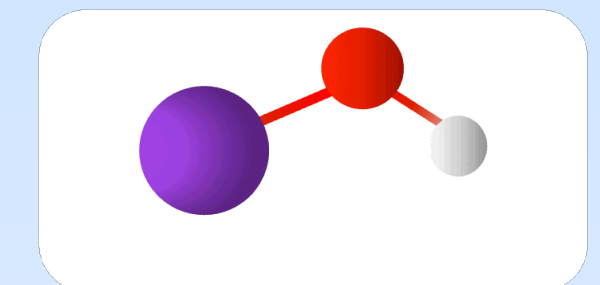
Oscillating Physical Constants due to Dark Matter



Level spacing will change if fundamental constants change



“accidental”
degeneracy



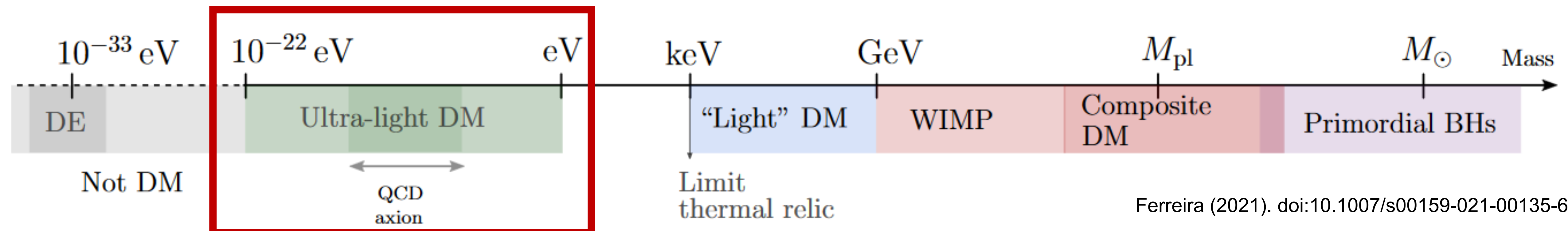
From Dark Matter to Oscillating Physical Constants

*Unknown
Ultralight Dark
Matter Field*

$$\phi_{DM}$$



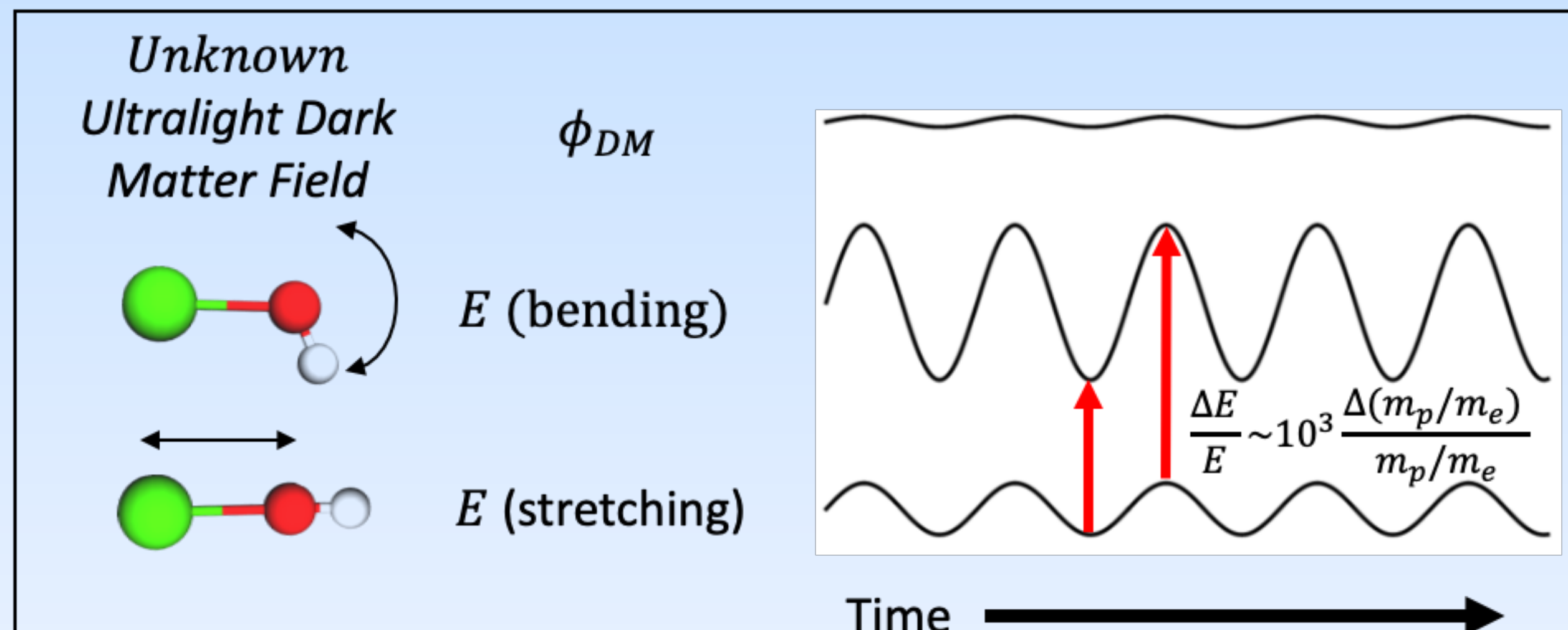
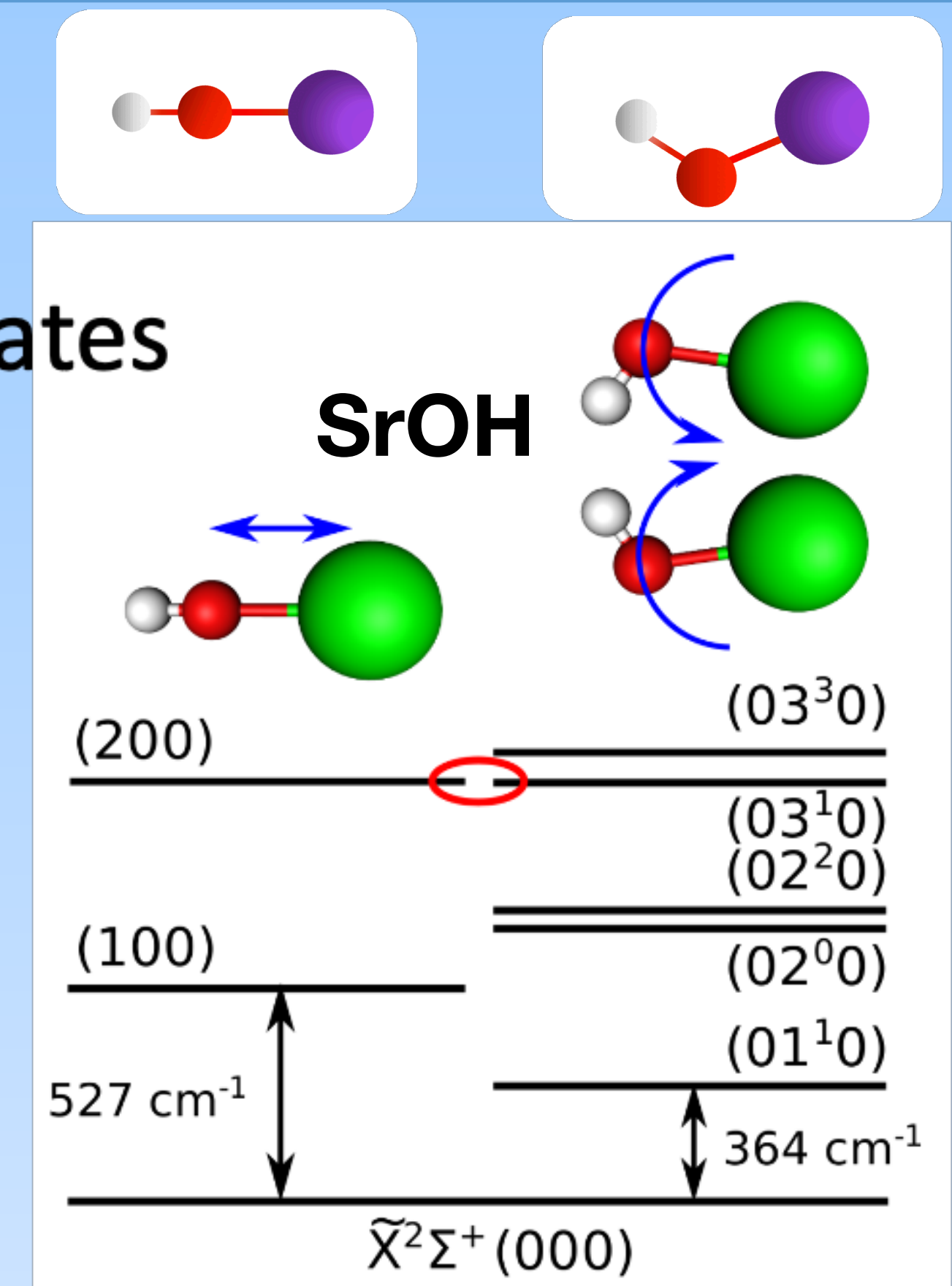
- Extremely diverse range of models could account for dark matter
- Scalar DM particles < 1 eV would be cold, have large Compton wavelengths, form BEC or superfluid
- Results in an oscillating background field
- Coupling of this field to “ordinary” matter results in oscillating physical properties, “constants”



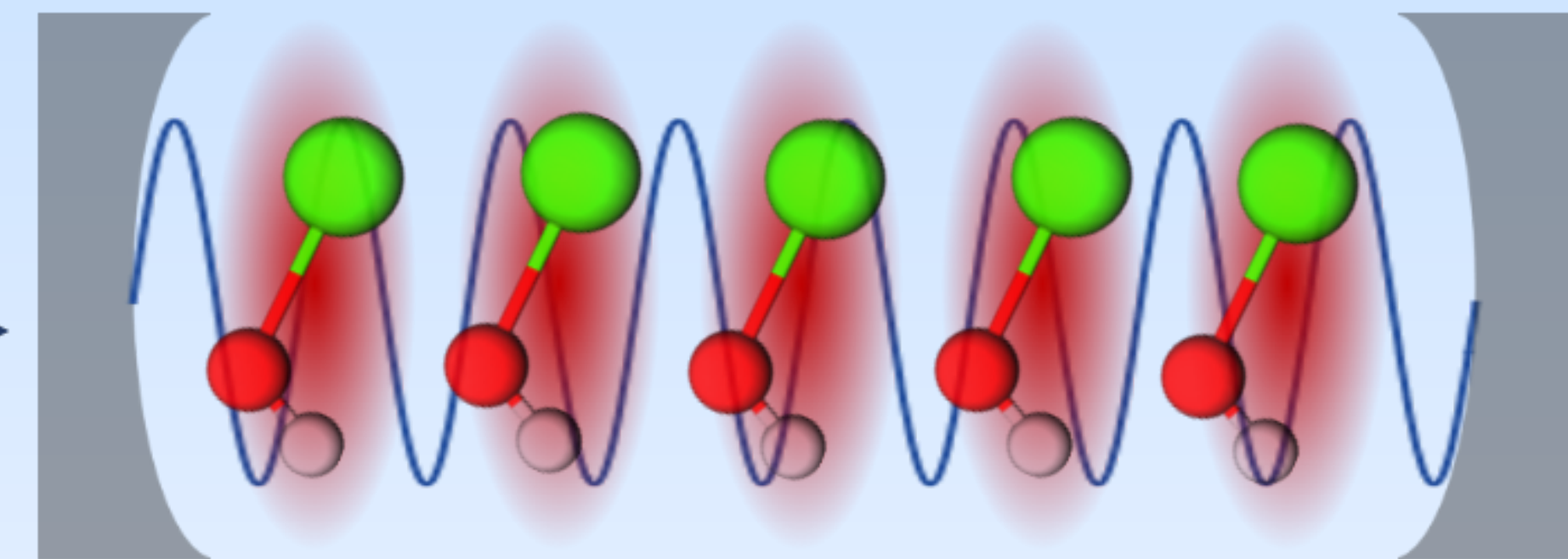
Ultralight Dark Matter

Ultralight Dark Matter has different effects on excited states
Can take advantage of fortuitous near-degeneracies

- For $\tilde{X}(200) \leftrightarrow \tilde{X}(03^10)$ transition in SrOH has $Q_\mu \approx 10 - 10^3$ with $\omega \approx 1 - 50$ GHz
- Requires stable microwaves only
 - no need for optical frequency comb
- Suppressed systematic errors
- Previously laser cooled in Doyle lab
- $\sim 10^{-17} / \sqrt{\text{day}}$ fractional uncertainty in $\delta\mu/\mu$



Trap
Probe



Sensitivity can be enhanced with spin squeezing in a cavity

SrOH Dark Matter Discovery Space

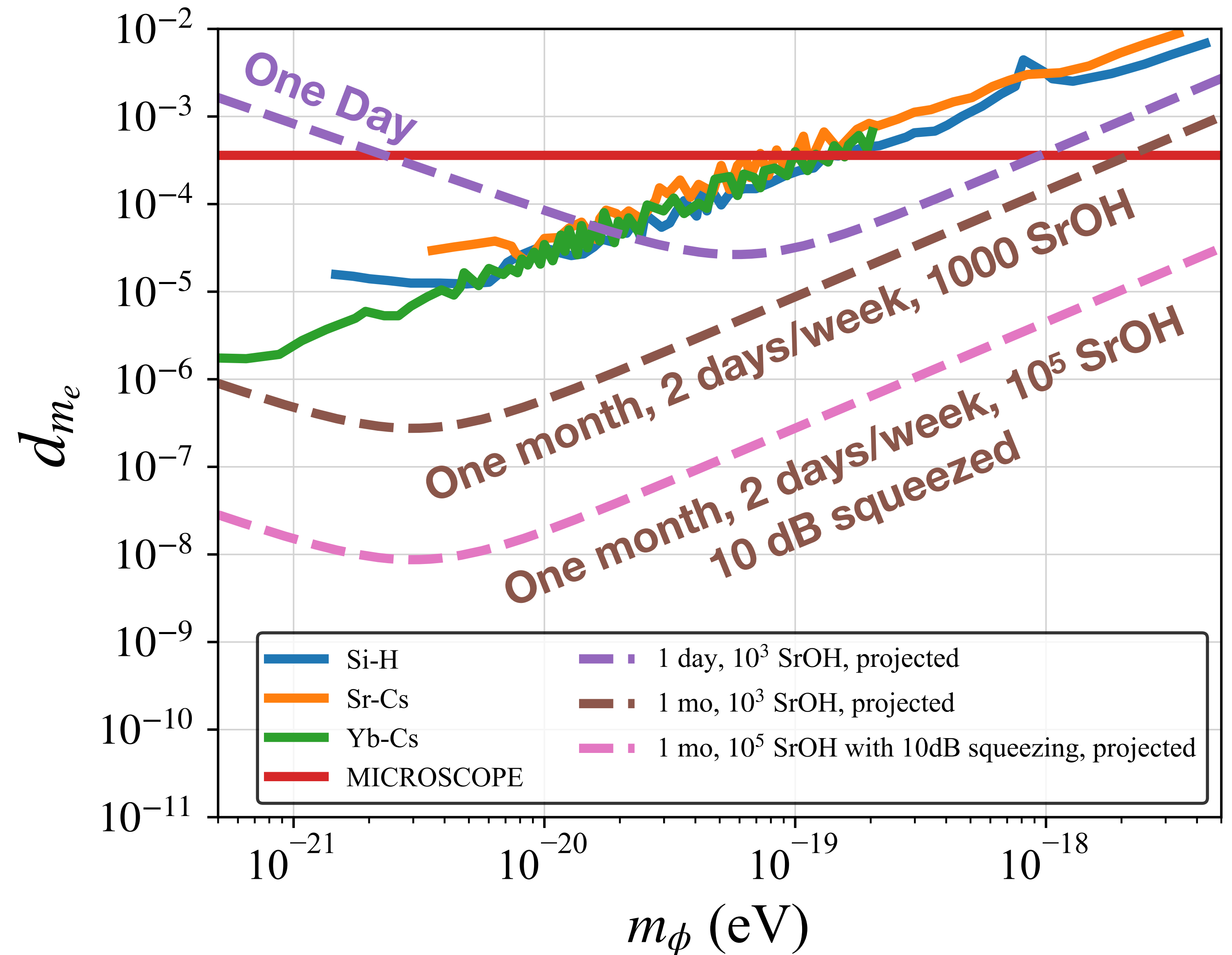
SrOH in ODT

1 day curve is continuous 24 hours of data

1 month curves are for
48 hours of data per week,
4 weeks of data taking.

Optimal sensitivity versus mass varies
because
the longest period measured changes
(here, 30 days vs. 1 day).

We currently have 1000 SrOH in ODT

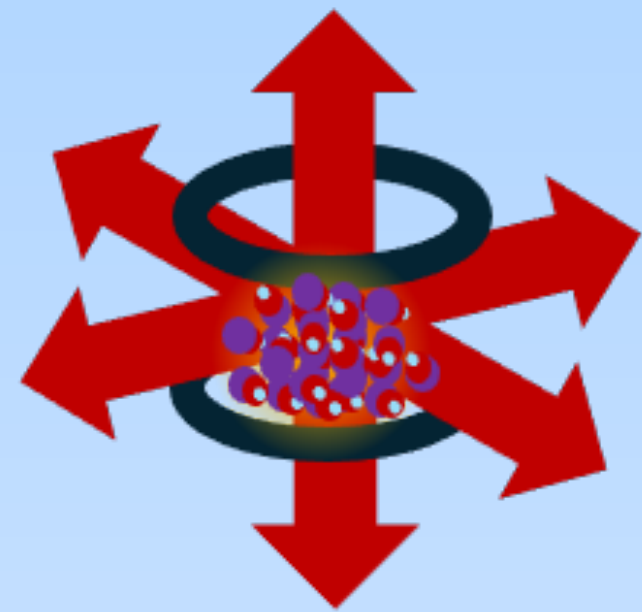


SrOH Optical Trapping

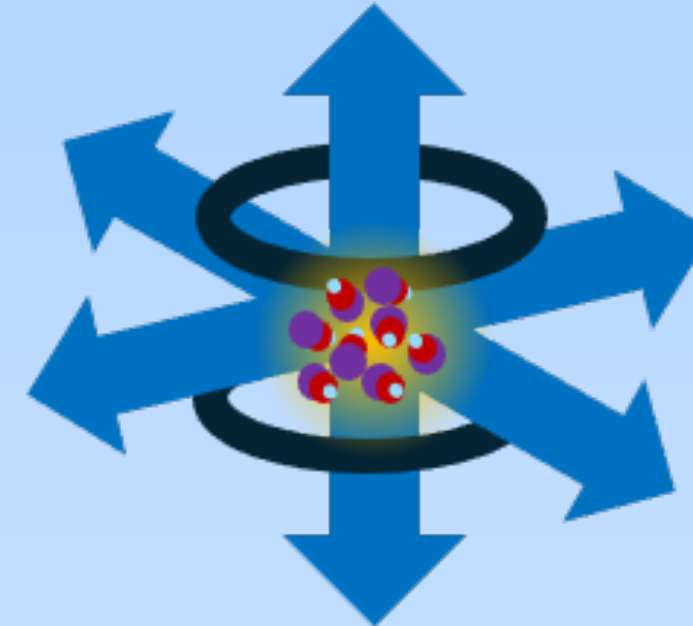
SrOH in ODT

**ODT lifetime (1.2 s) for
1300 Ground State Molecules**

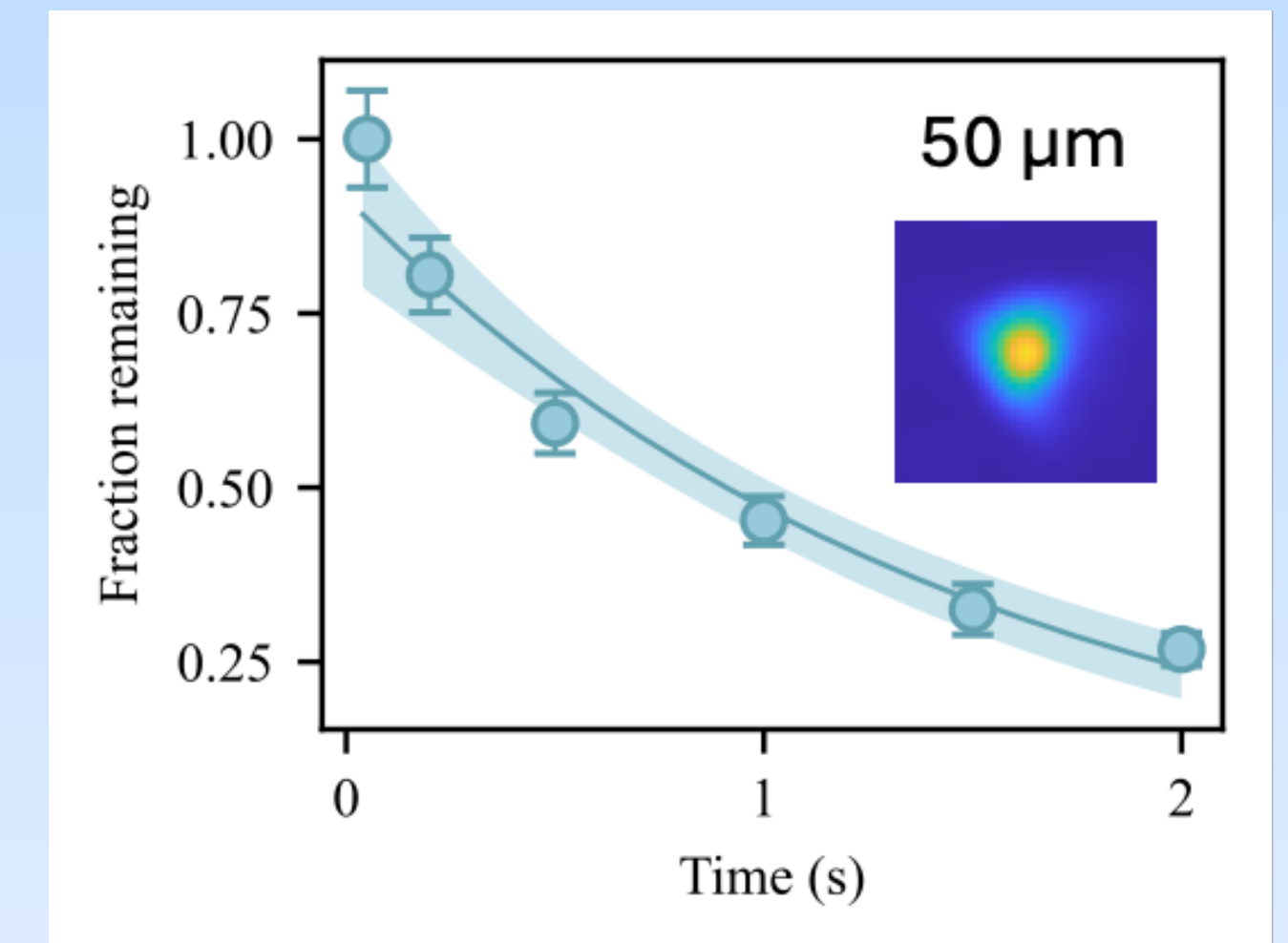
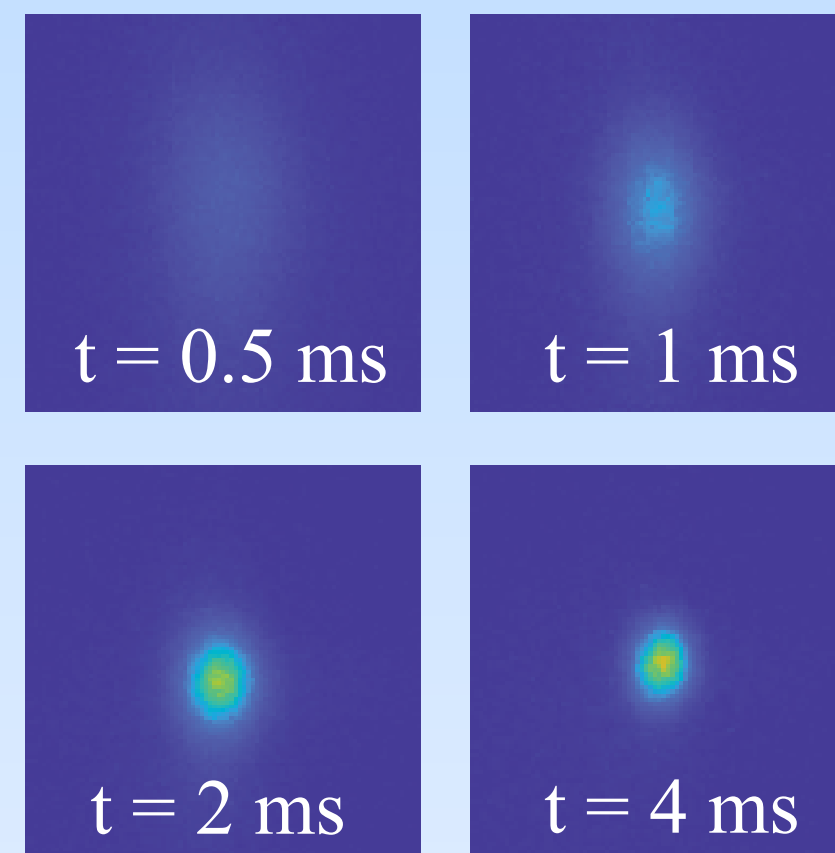
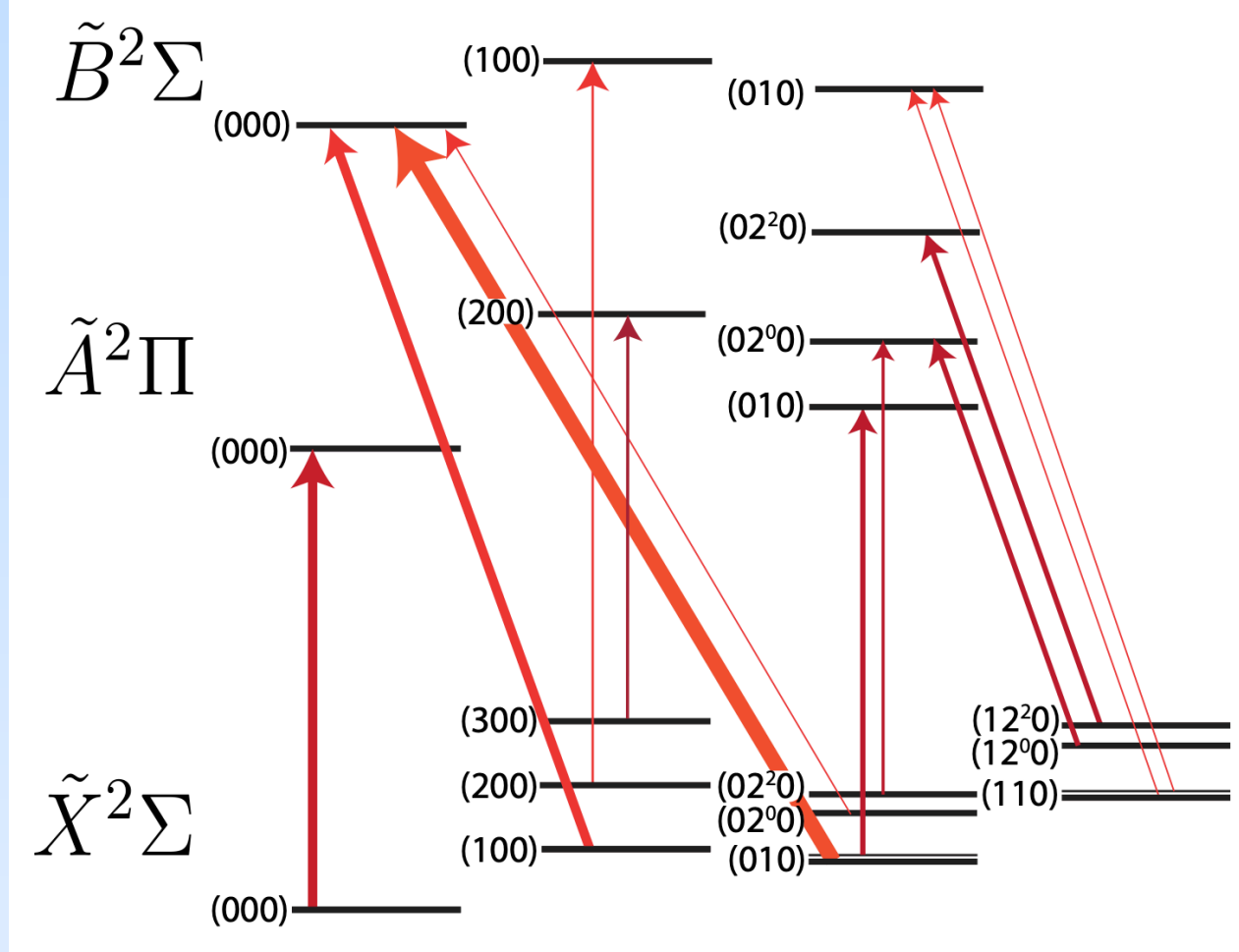
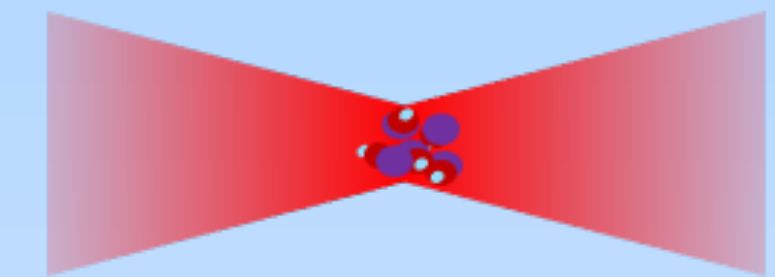
Red MOT,
 10^4 molecules, ~ 1 mm



Conveyor Belt MOT,
 $\sim 5,000$ molecules, ~ 80 μm



Optical dipole trap,
 10^3 molecules, ~ 50 μm



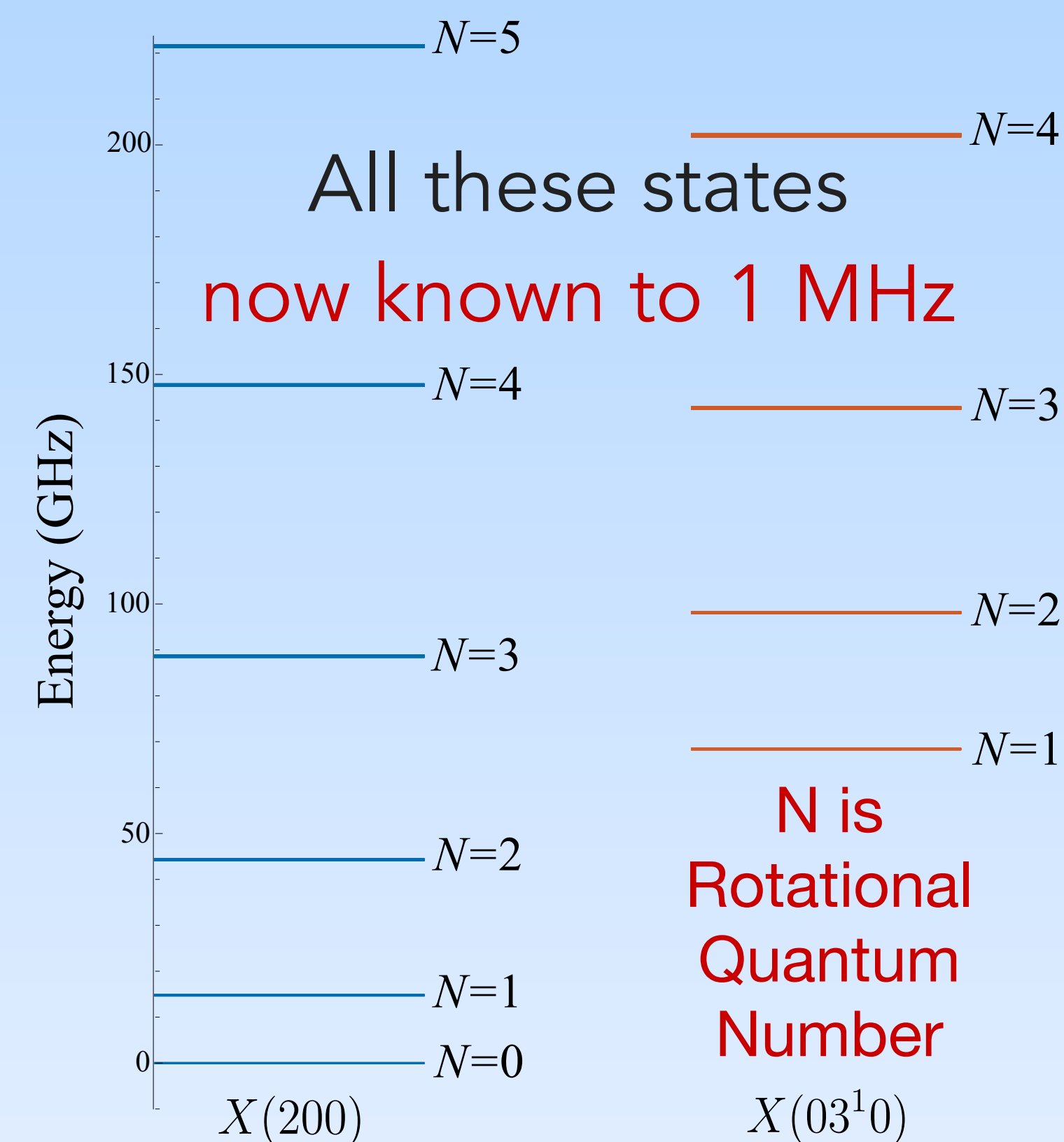
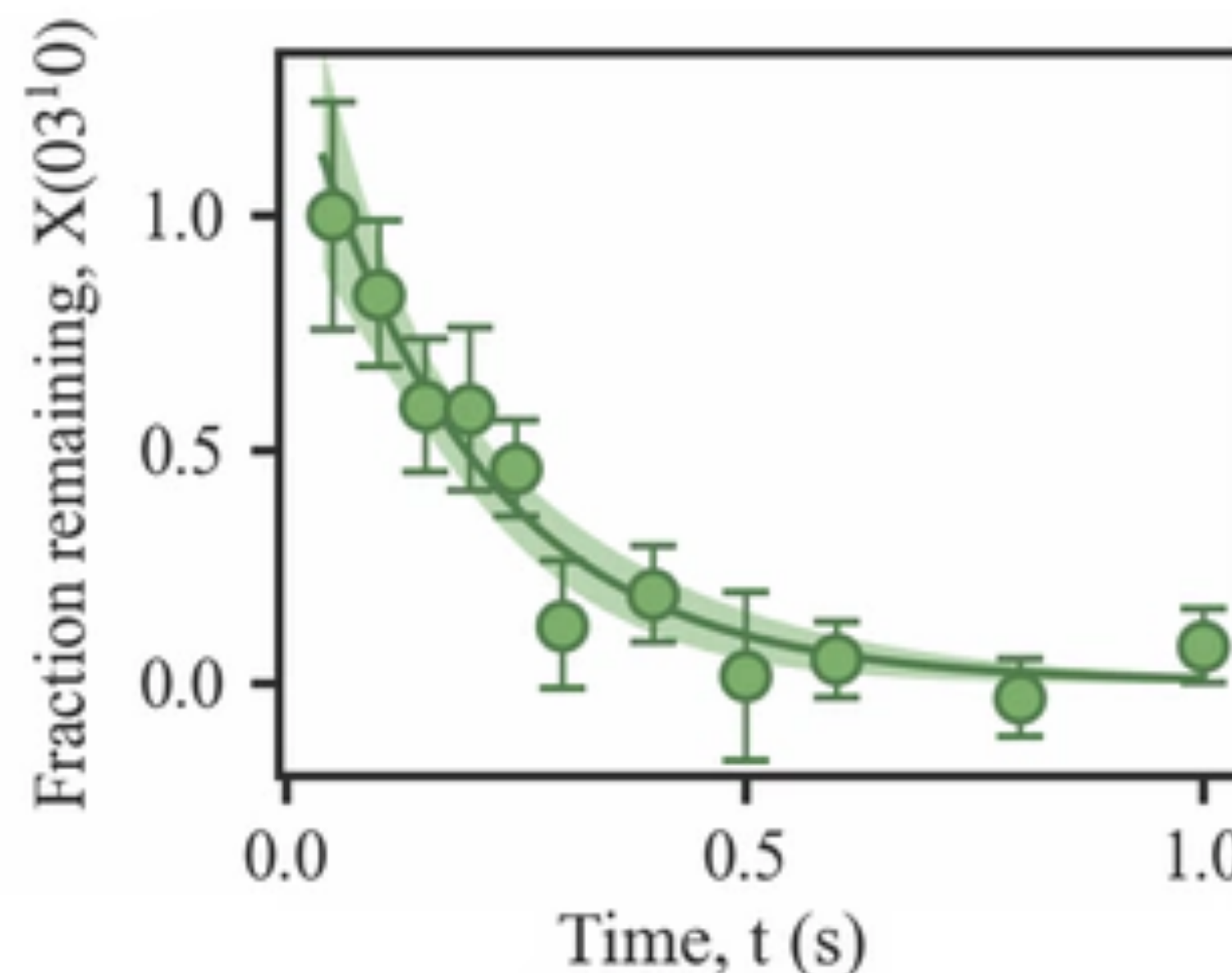
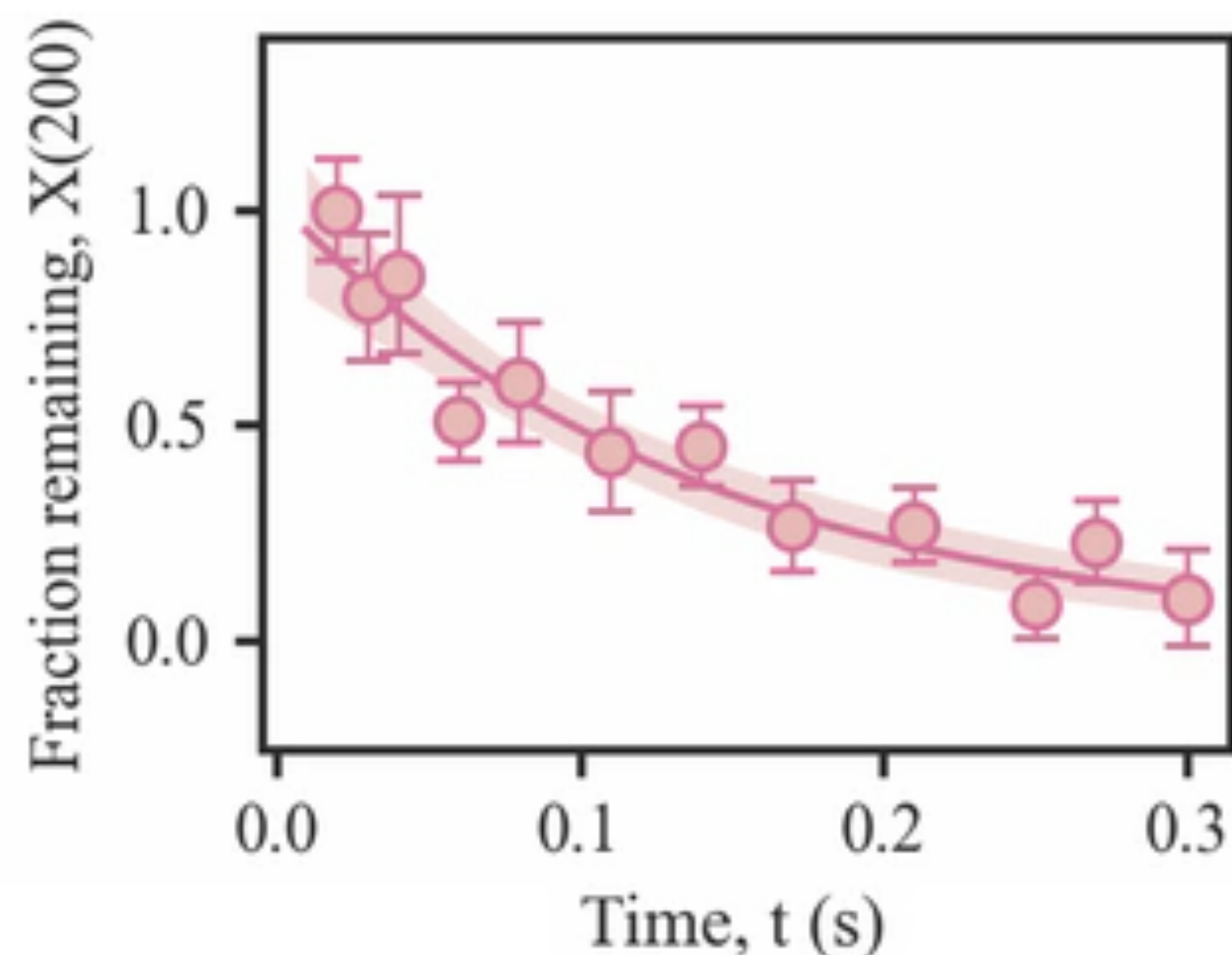
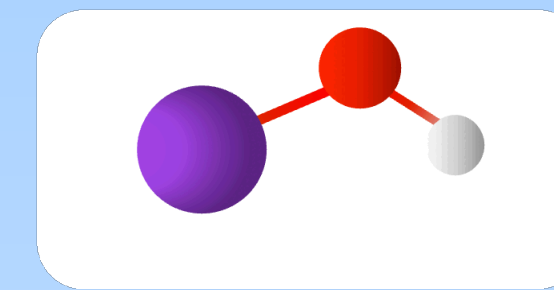
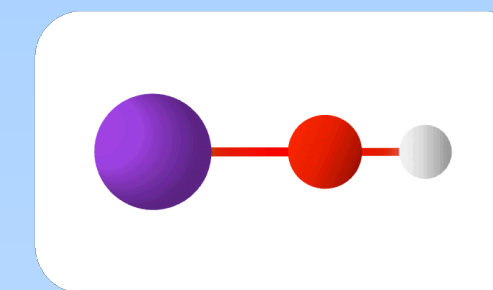
SrOH in ODT

SrOH Dark Matter Science States Found

Lifetimes of SrOH Dark Matter

Science States in ODT (1000 Molecules)

Precise Energies of
SrOH Dark Matter
Science States Determined



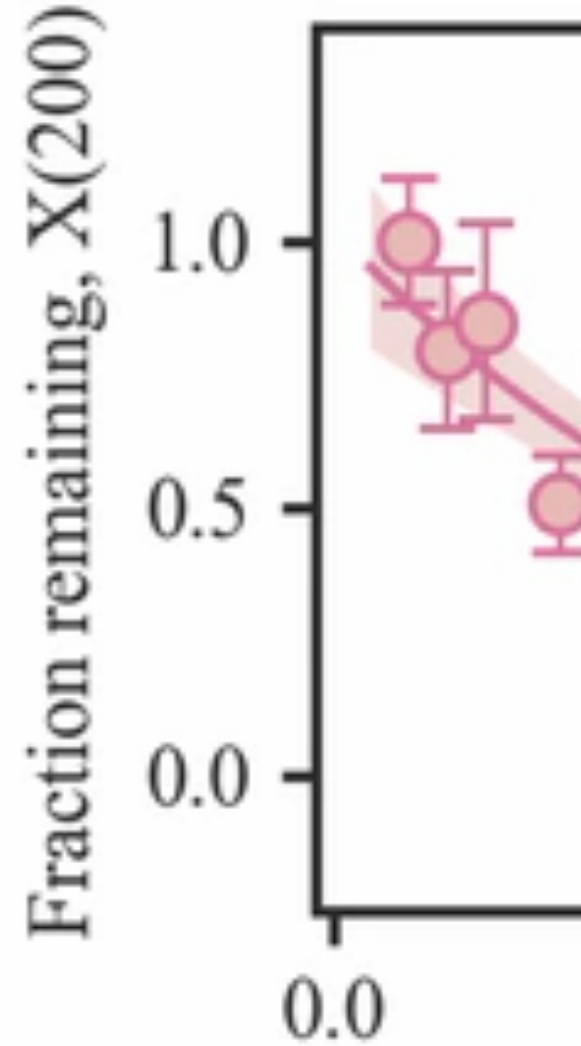
[Optical Trapping of SrOH Molecules for Dark Matter and T-violation Searches](#), H. Sawaoka, A. Nasir, A. Lunstad, M. Li, J. Mango, Z.D. Lasner, and J.M.D, submitted, arXiv:2509.01618 (2025).

[High-sensitivity molecular spectroscopy of SrOH using magneto-optical trapping](#), A. Lunstad, H. Sawaoka, Z.D. Lasner, A. Nasir, M. Li, J. Mango, R. Fields, and J.M.D., submitted, arXiv:2509.09786 (2025).

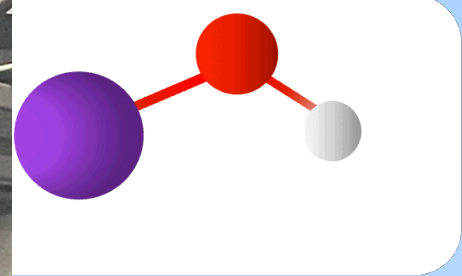
Dark Matter and EDM with SrOH current team

SrOH in O

Lifetimes



gies of
Matter
etermined



ates

1 MHz

N=4

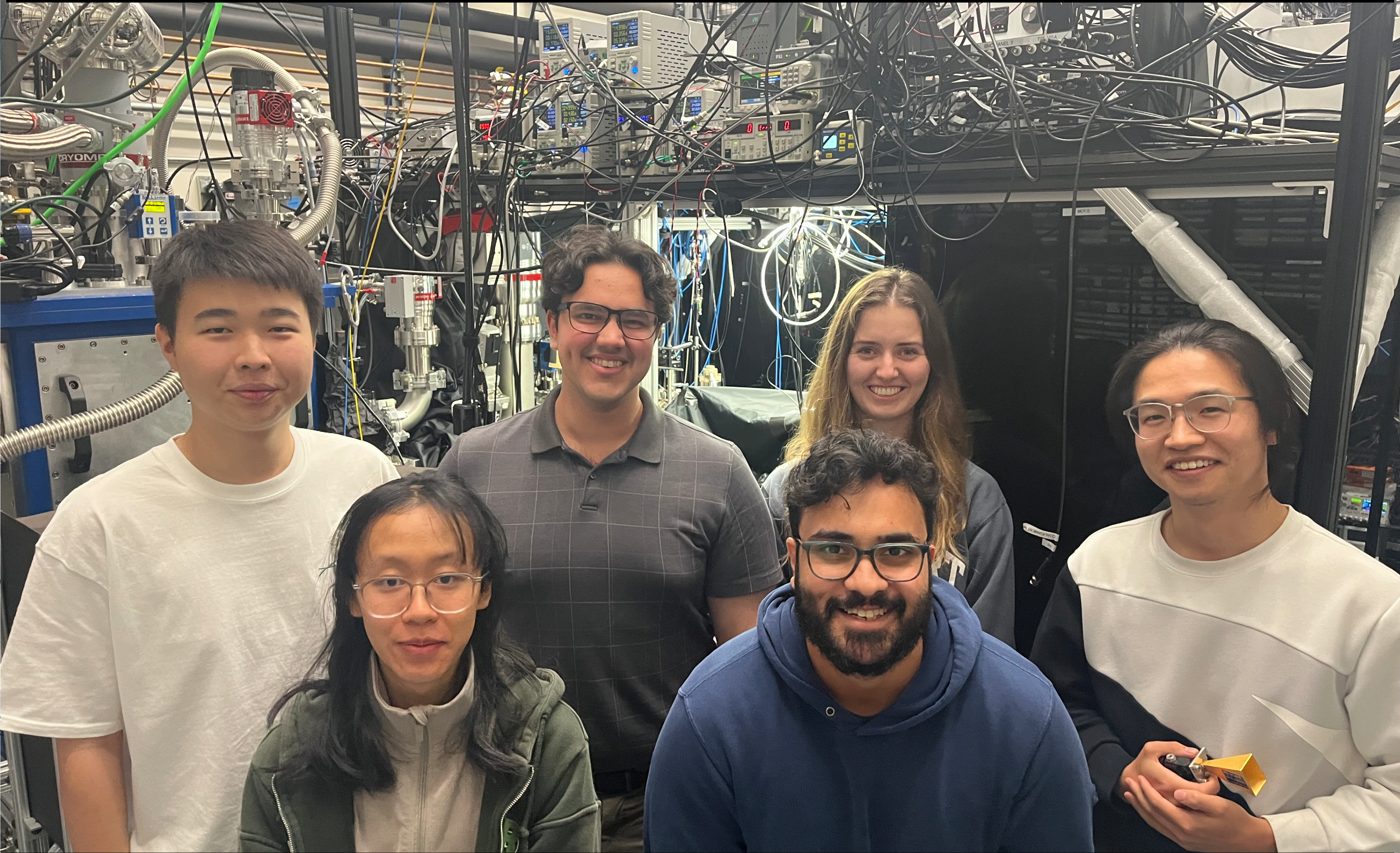
N=3

N=2

N=1

X(03¹0)

ad, H. Sawaoka,
9786 (2025).



Ray

Shuqi

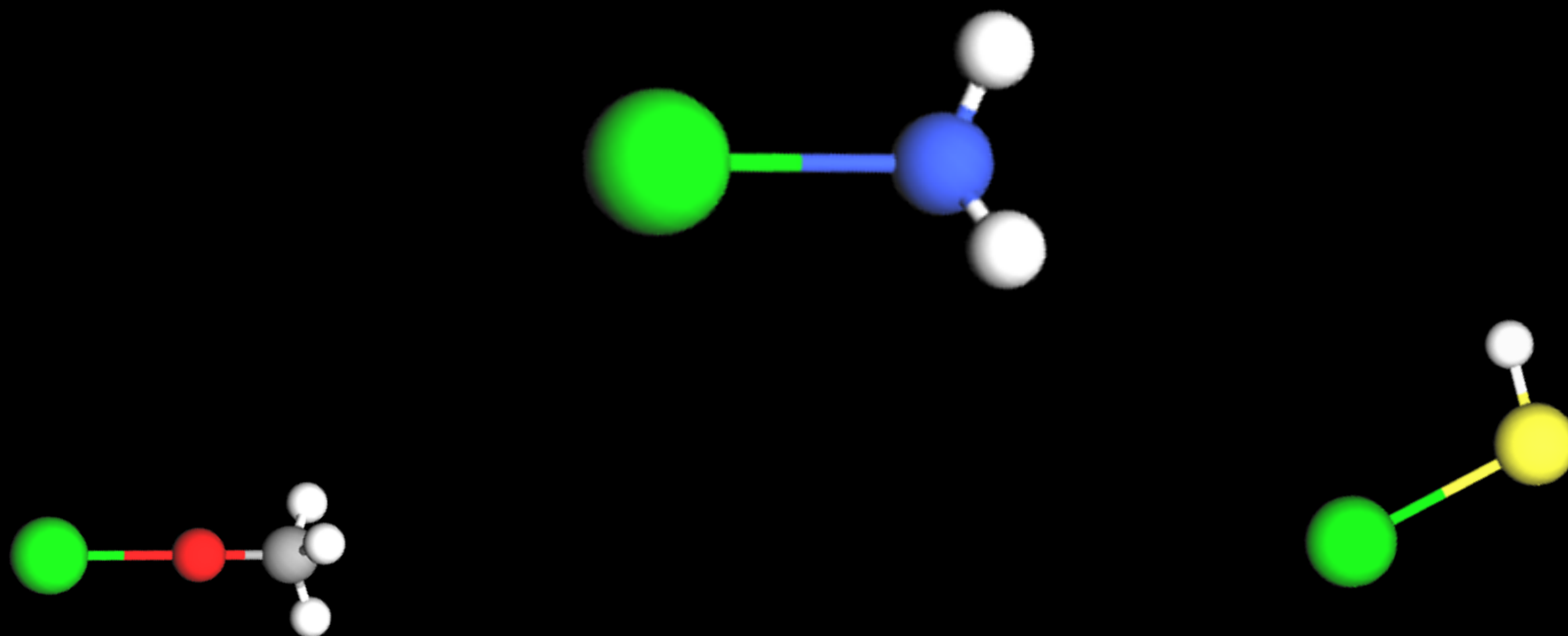
Saif

Abdullah

Annika

Mingda
holding
Mr. Microwave Horn

We need to explore the molecular complexity frontier... etc.
What happens when you excite a molecule?

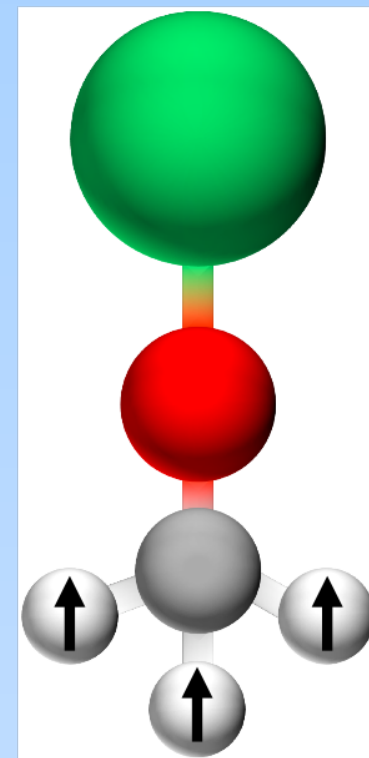


Laser Cooling of a Symmetric Top - CaOCH_3

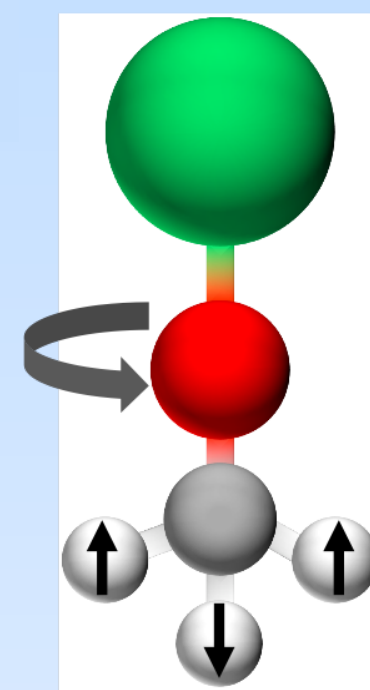
A teeny bit more complicated....

Two new challenges:

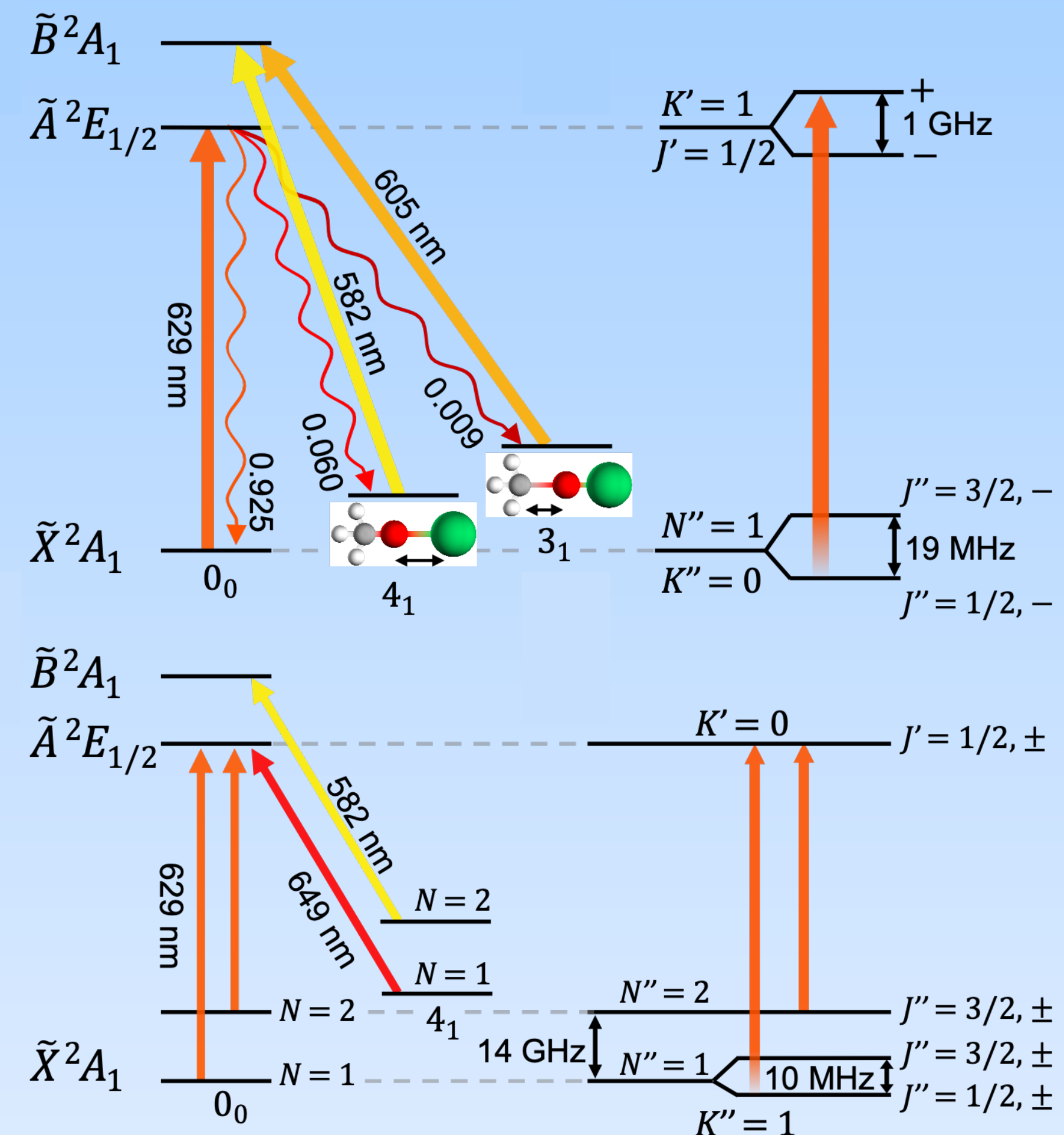
- 12 vibrational modes in play
 - Favorable **FCFs** measured¹
 - "Old hat" by now... just add vibrational repumps
- Novel rotational structure due to rigid body rotation K
 - Rotations about "a" principal axis allowed
 - No help from parity!**



$K = 0$



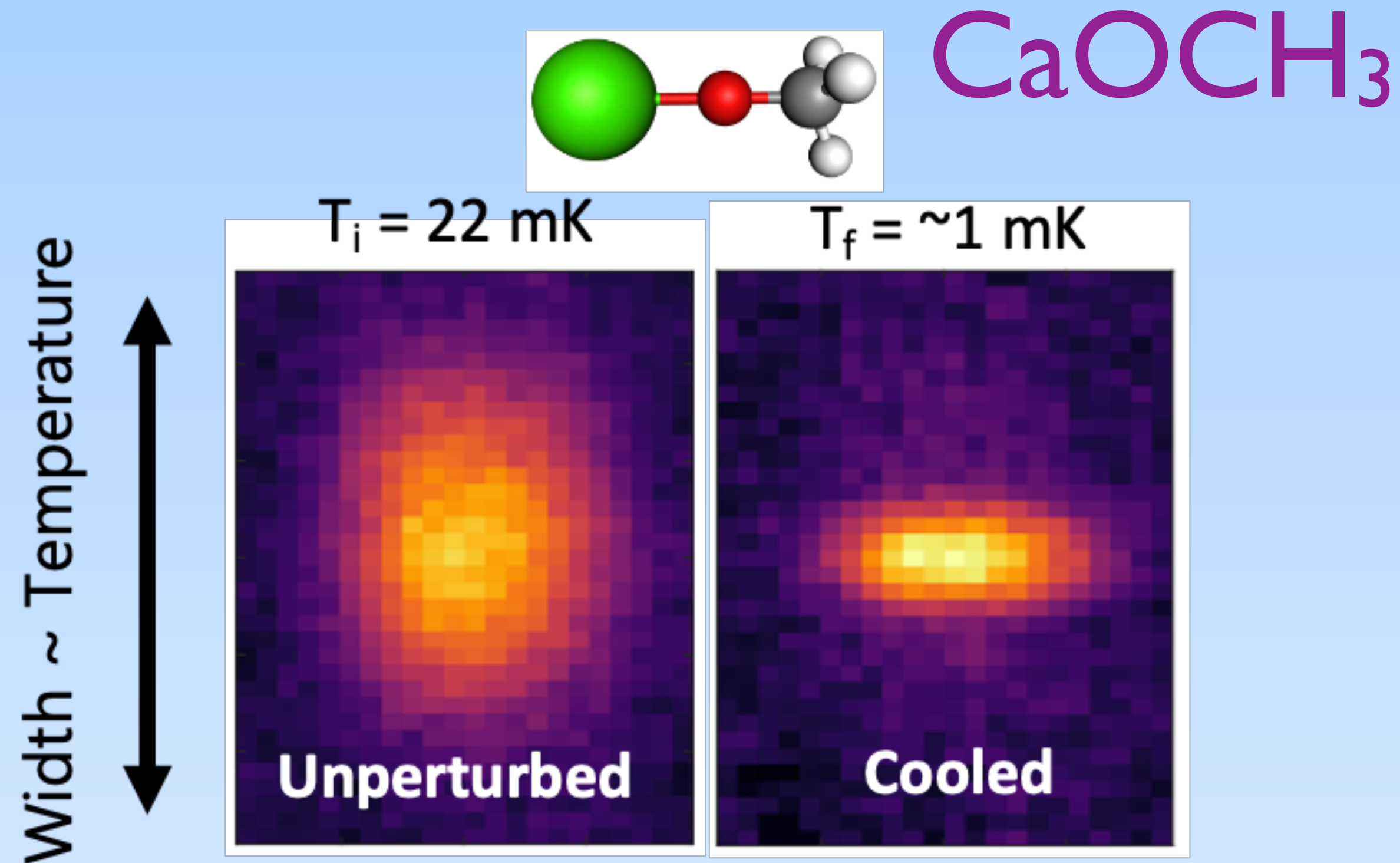
$K = 1$



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.D. 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.D. Science 369, 1366 (2020). [arXiv:2004.02848](#)

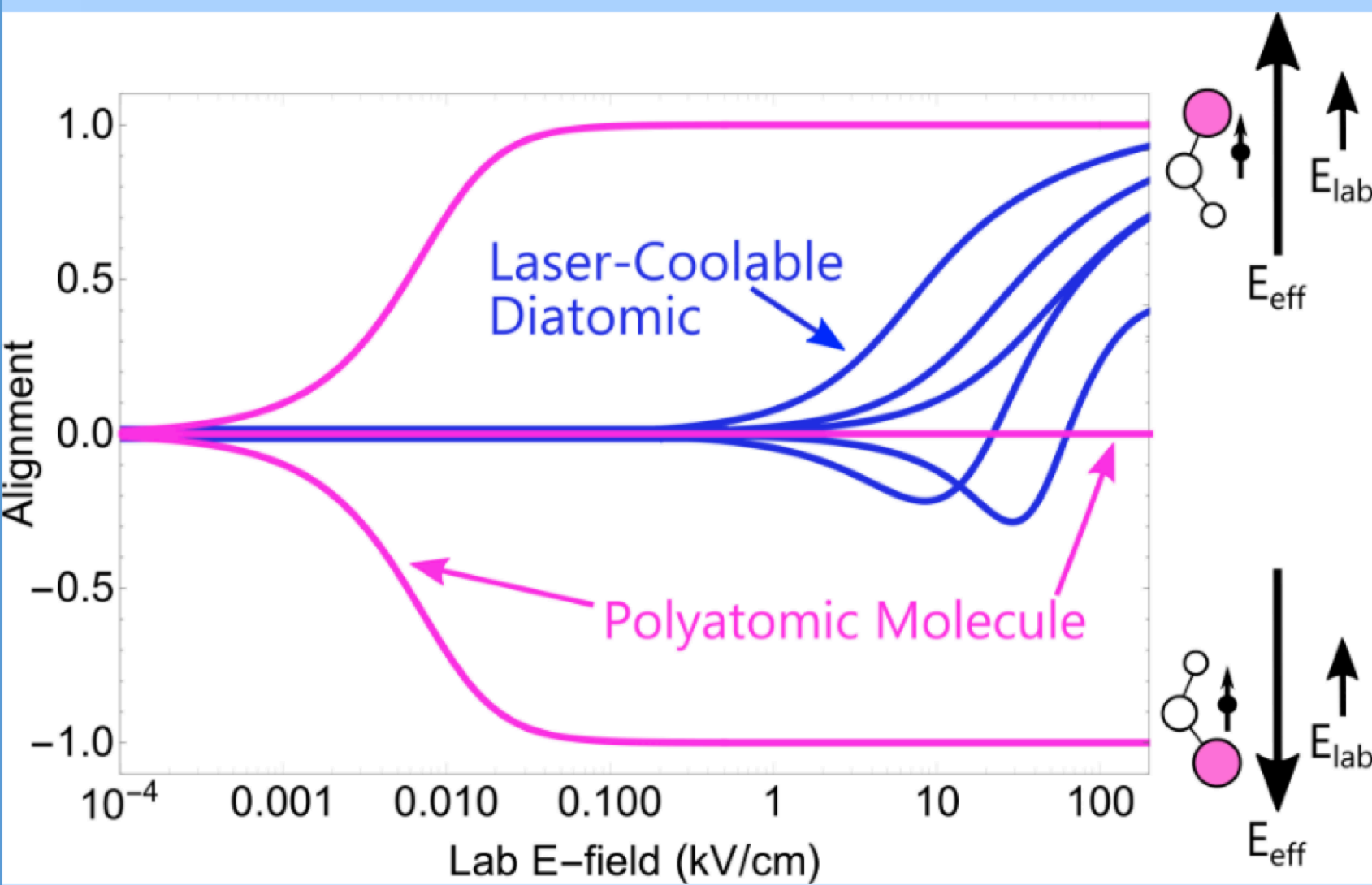
1-D Laser Cooling of a Symmetric Top Molecule



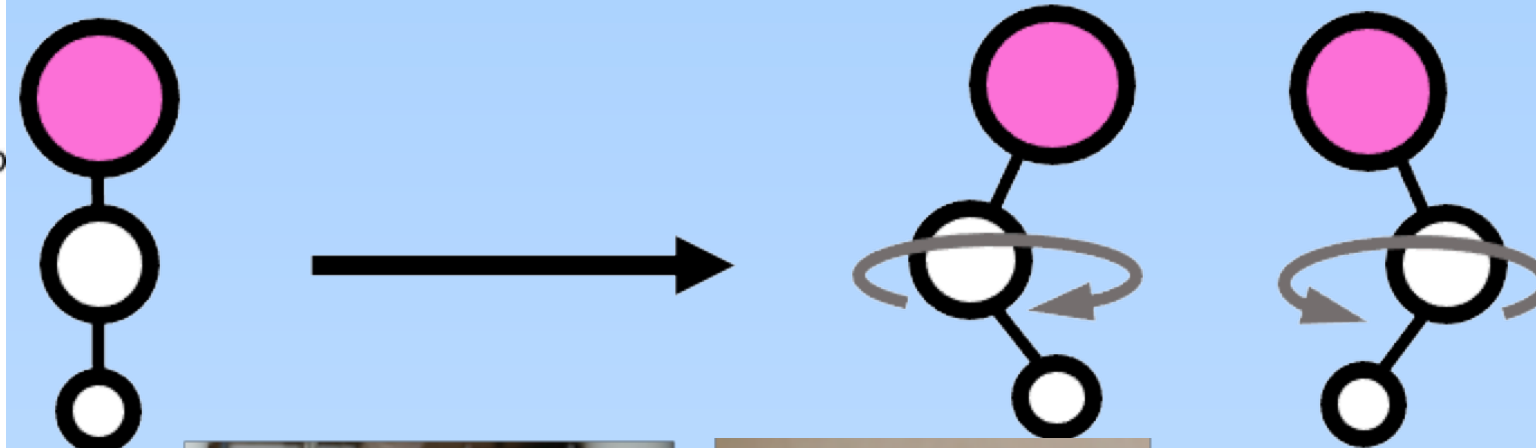
Cooling performance essentially identical to that of simpler linear molecules, despite greatly increased molecular complexity.

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

A CP-v Search Strategy



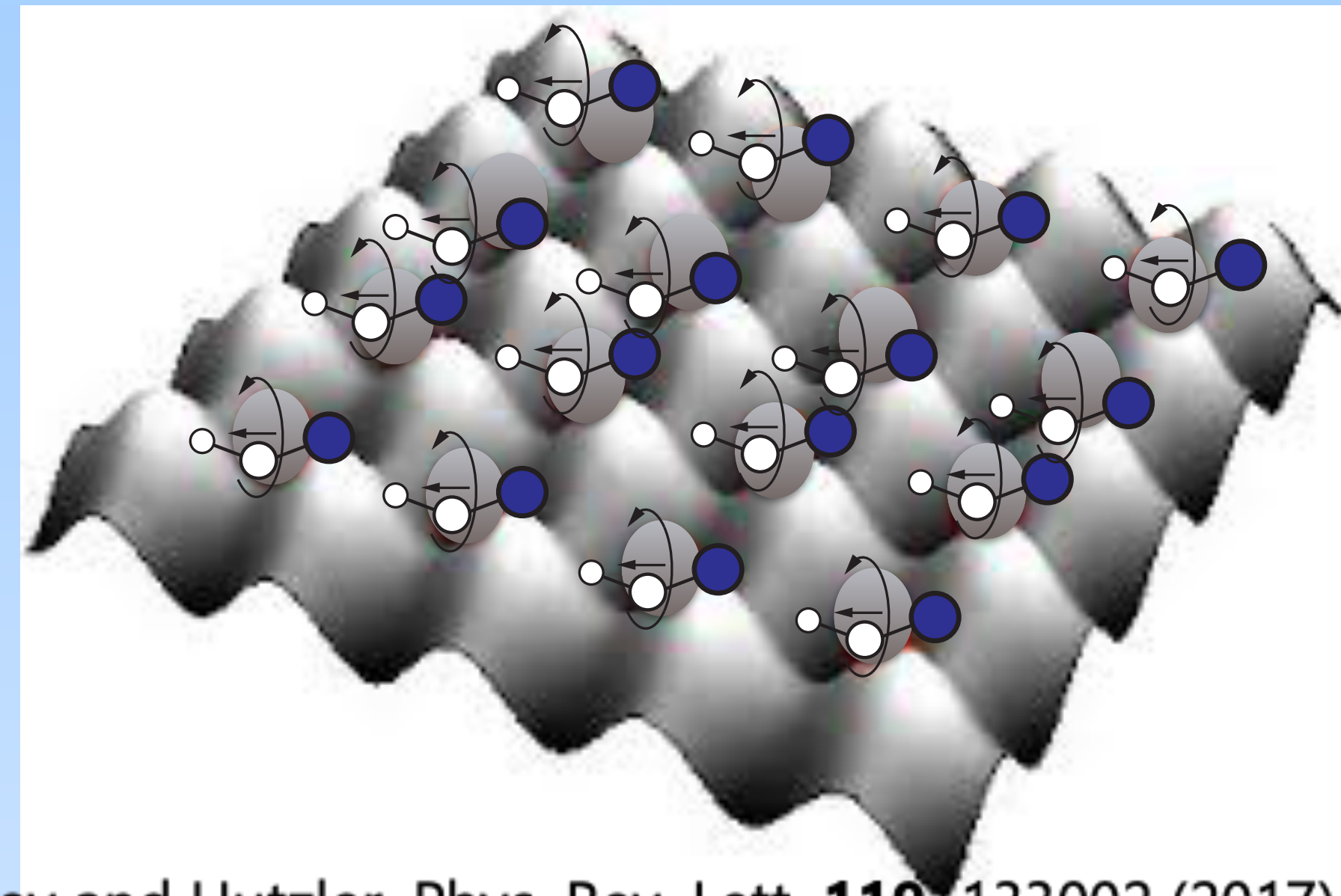
● Sr, Yb, Ba, Ra...



Nick Hutzler

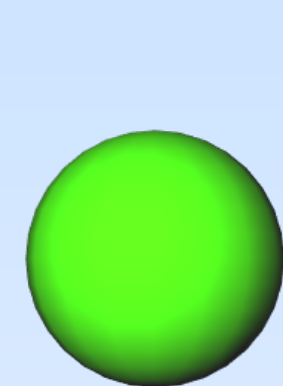


Ivan Kozyryev

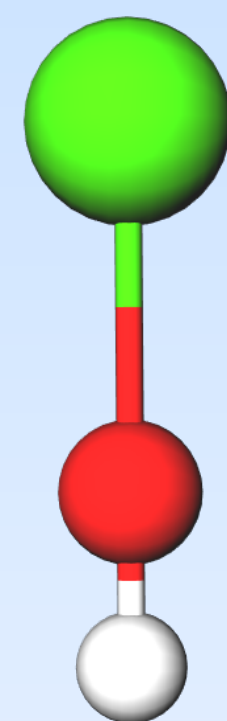


Kozyryev and Hutzler, Phys. Rev. Lett. **119**, 133002 (2017)

Complexity [reduced molecular symmetry]

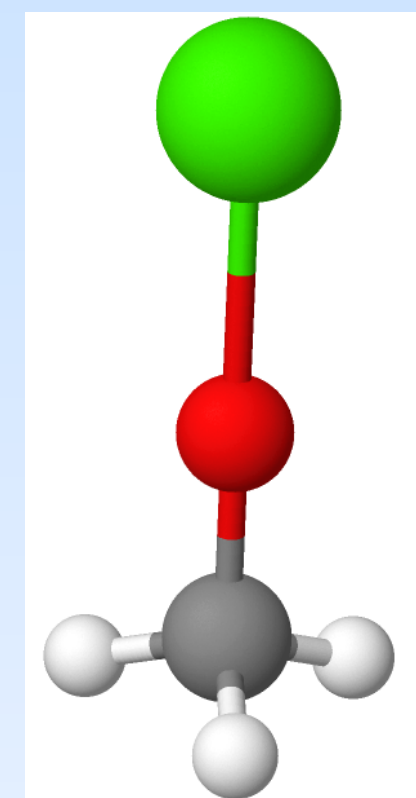


Eliminate
spherical symmetry
[1 moment of inertia]



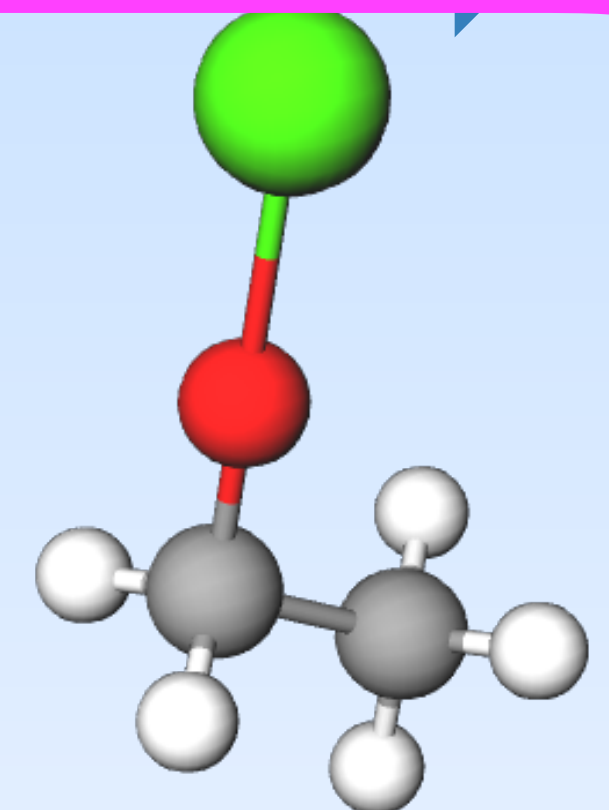
0.1-~1.0 s

Vibrational Lifetime

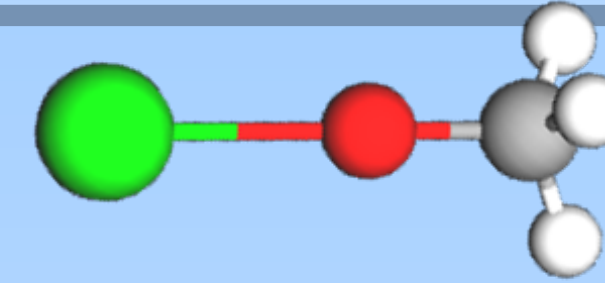
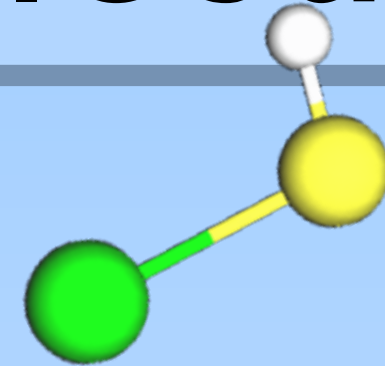
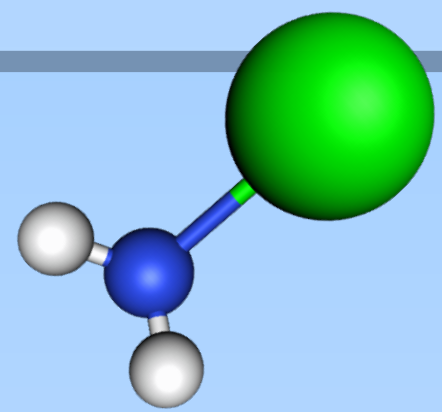


>10 s

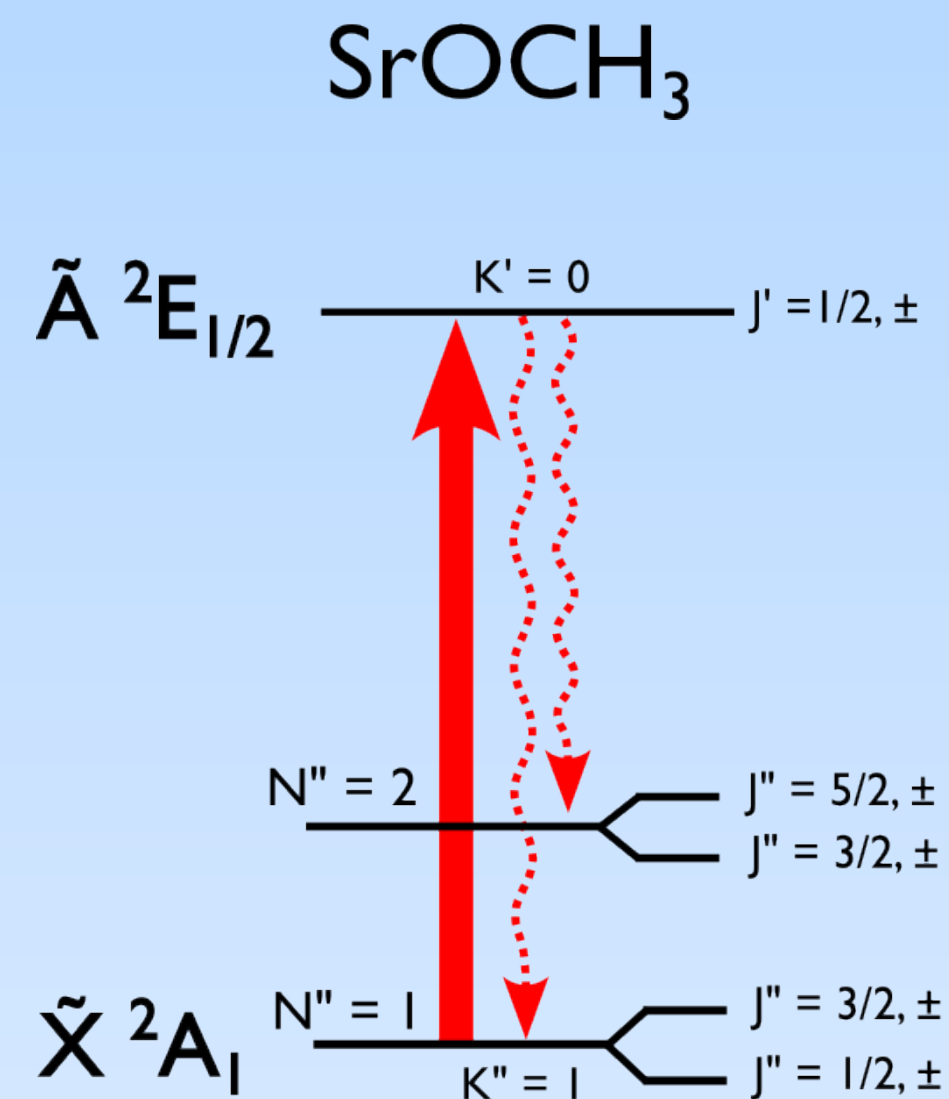
Rotational Lifetimes



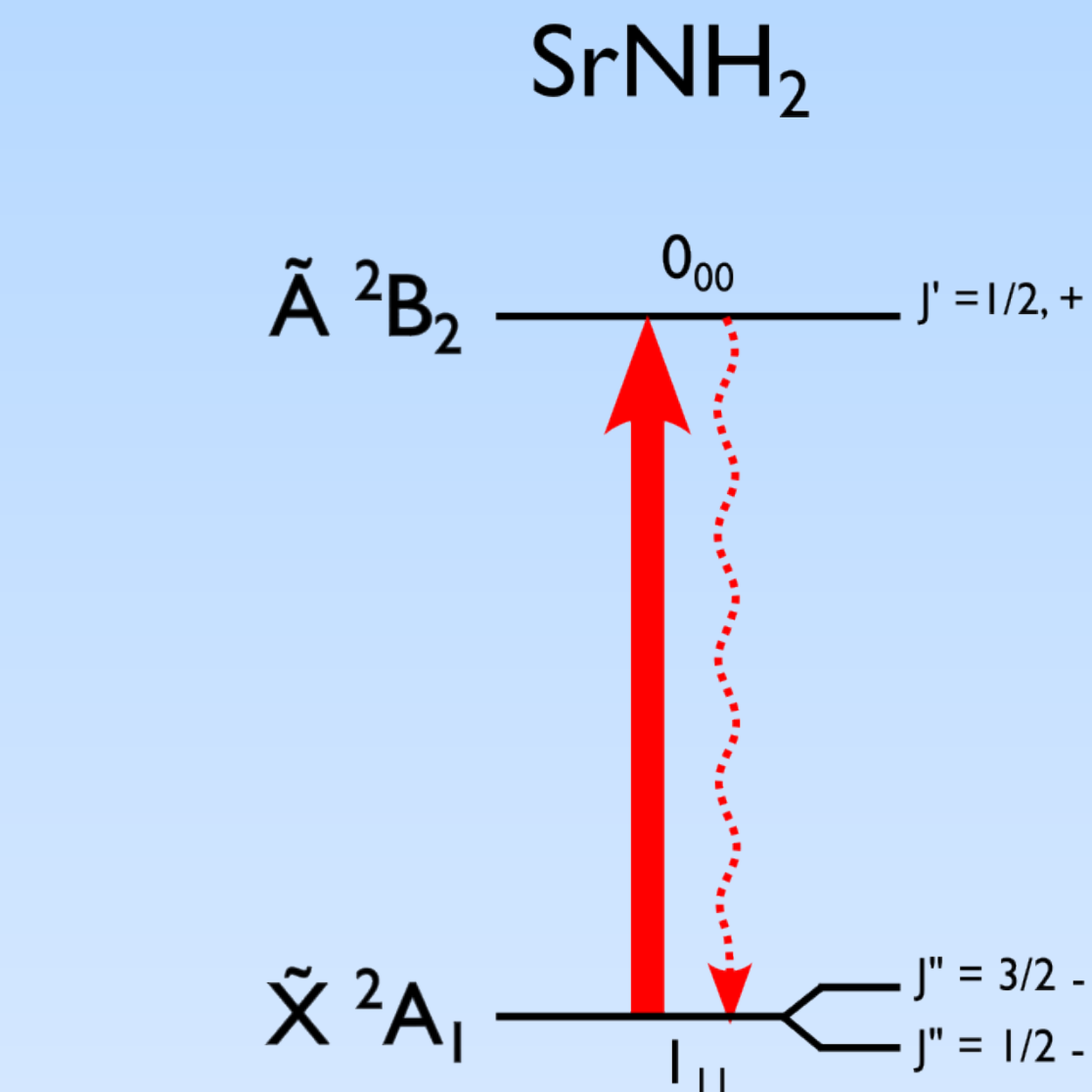
Rotational Closure in Nonlinear Molecules



❖ Symmetry-group specific: not only nominal scheme but also allowed perturbations

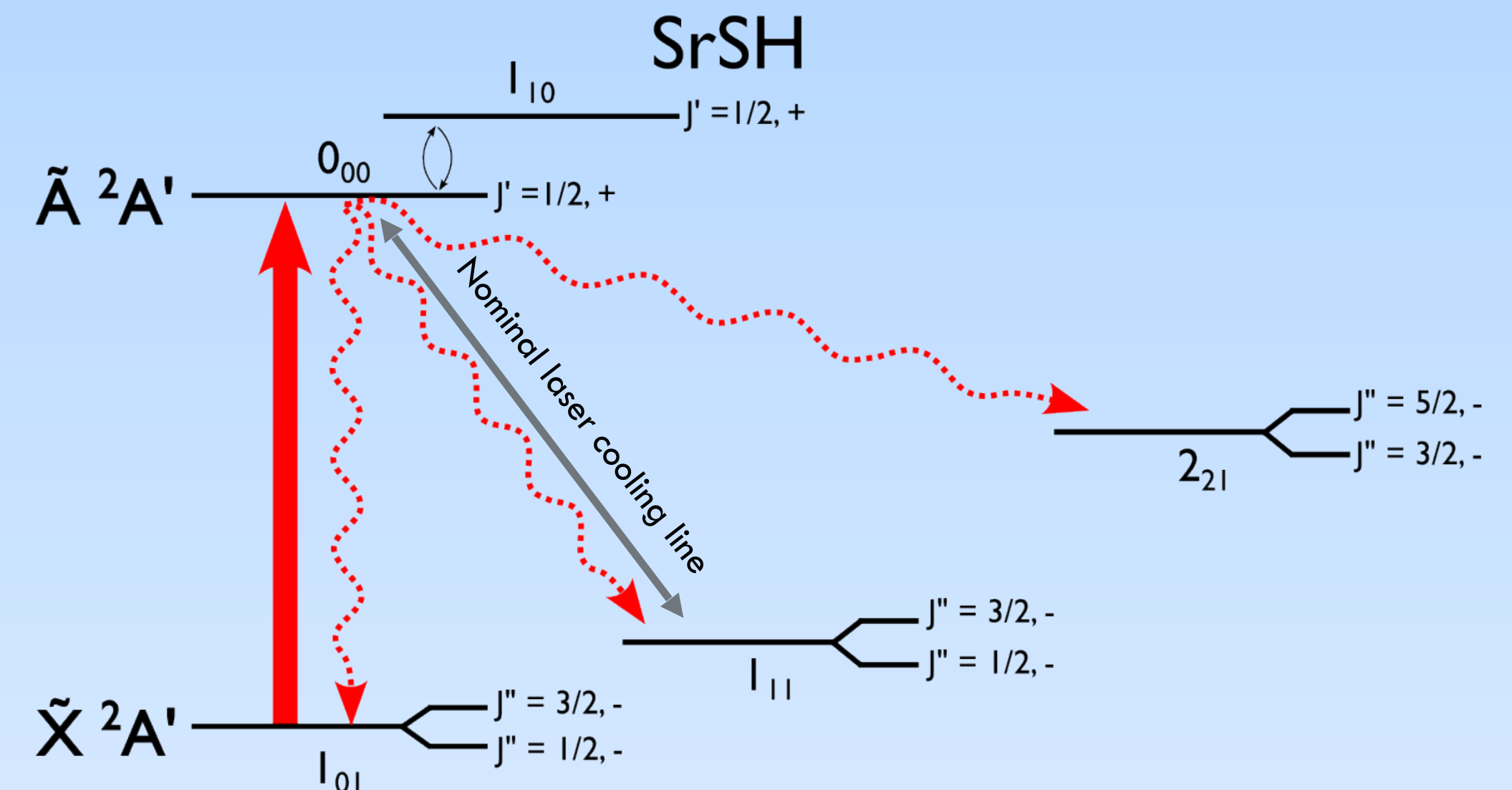


❖ $K''=1$ state requires a rotational repump



❖ $K_a''=1$ state *does not* require a repump

❖ Nearby states of the same J' are different symmetry; mixing forbidden



❖ Nominal scheme is not useful

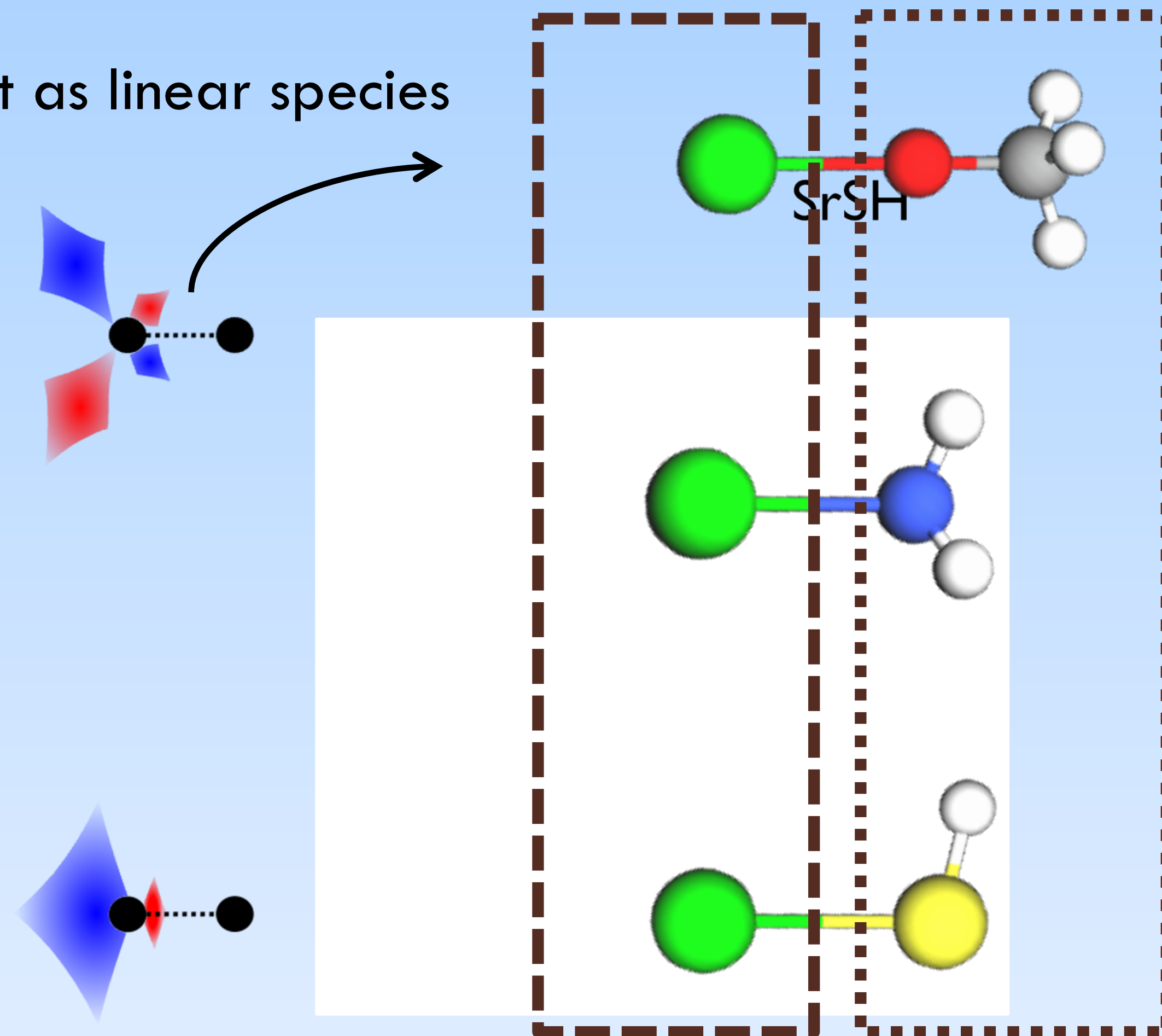
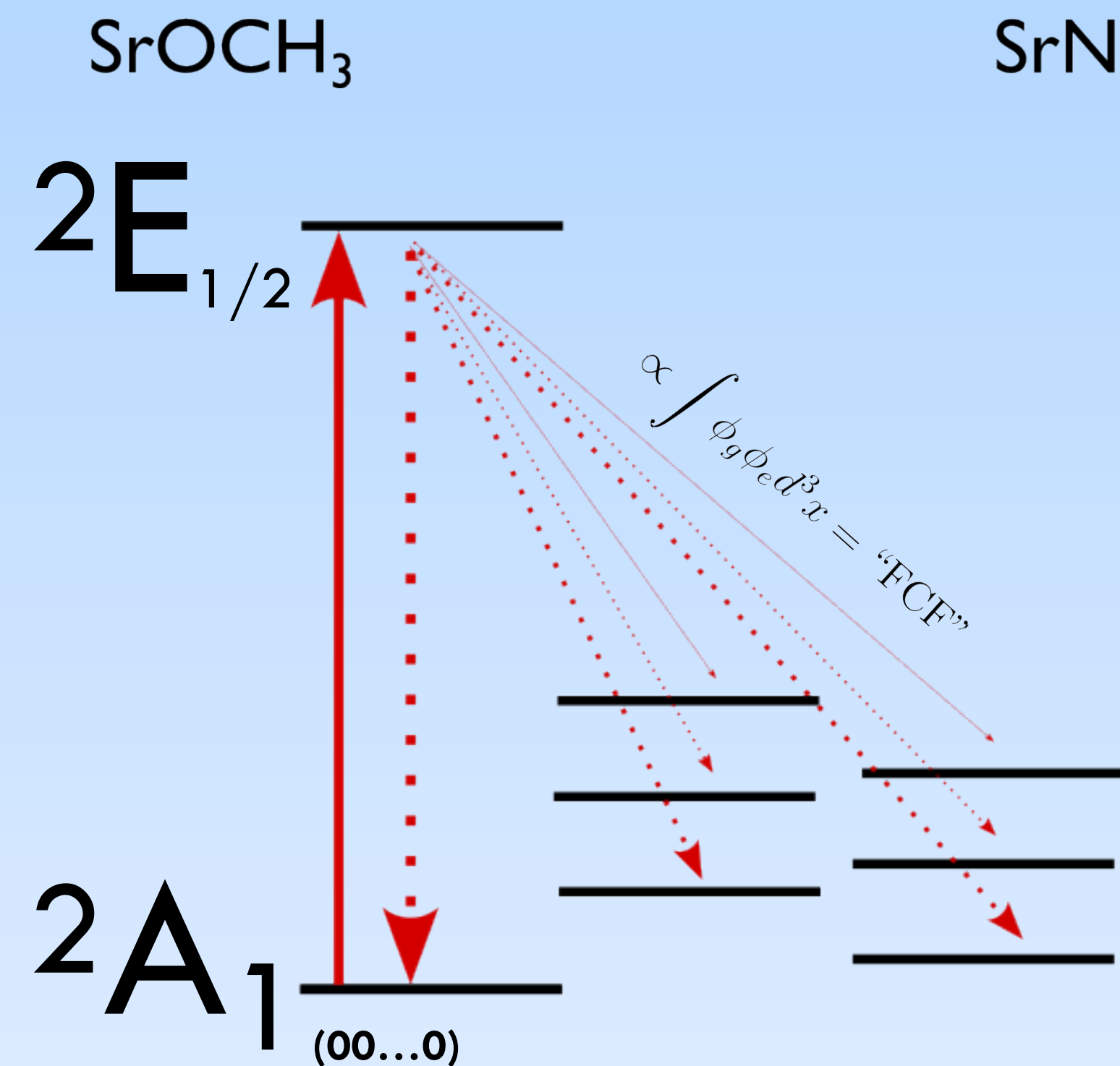
❖ Mixing with nearby J' states is symmetry allowed (and strongly occurs)

[Proposal for laser cooling of complex polyatomic molecules](#) I. Kozyryev, L. Baum, K. Matsuda, and J.M.D, ChemPhysChem 17, 3641 (2016).

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

Vibrational Closure in Nonlinear Molecules

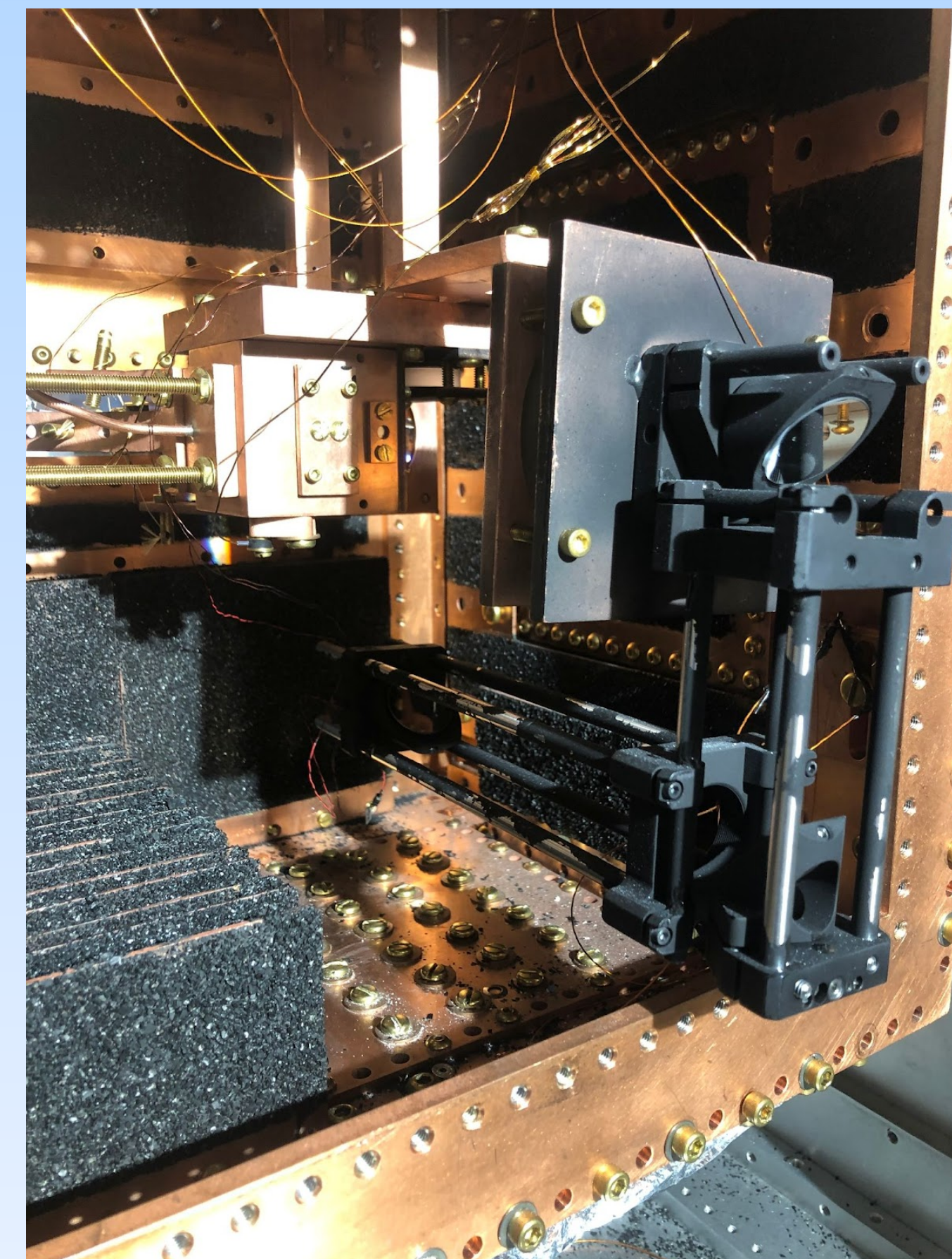
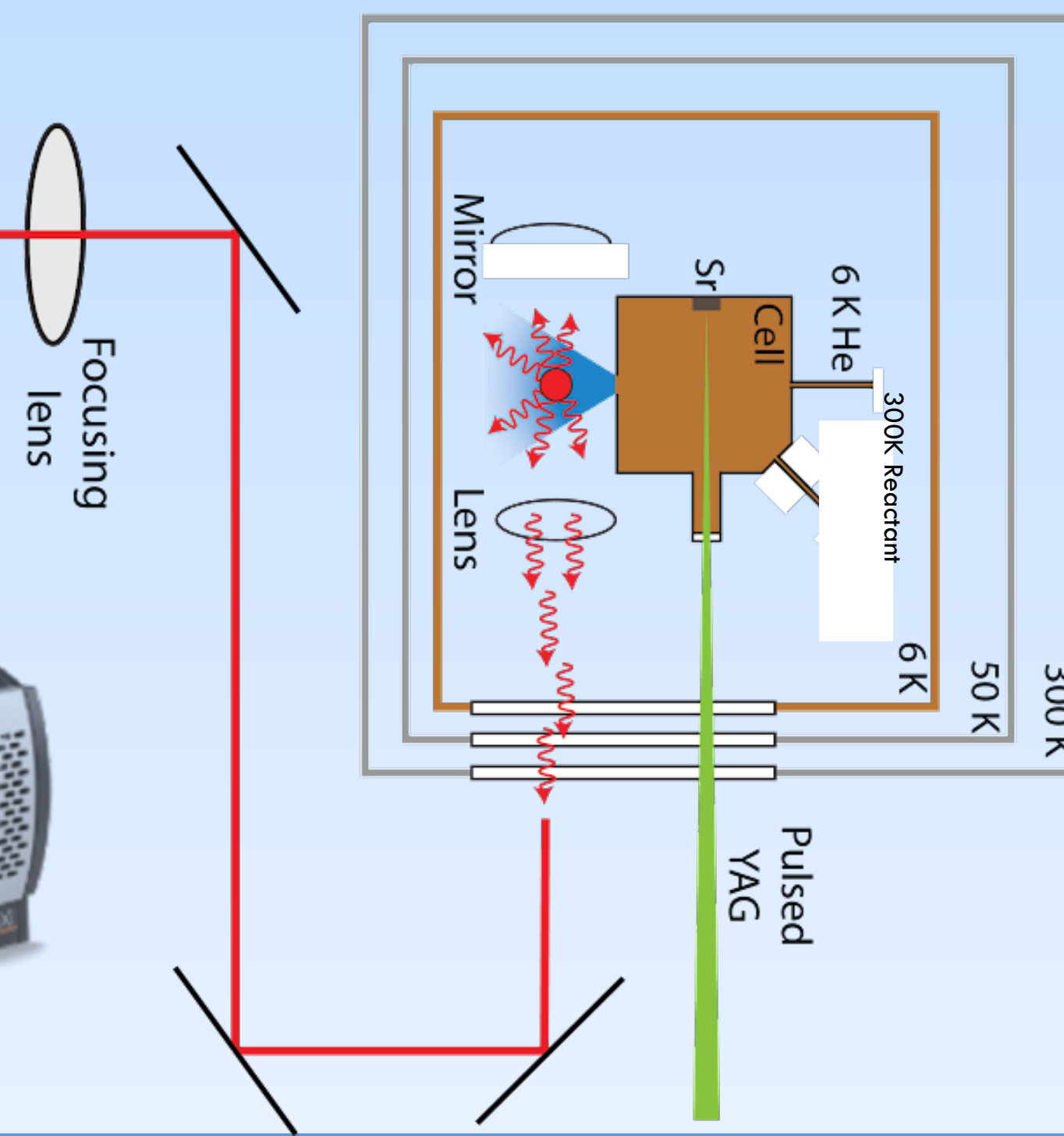
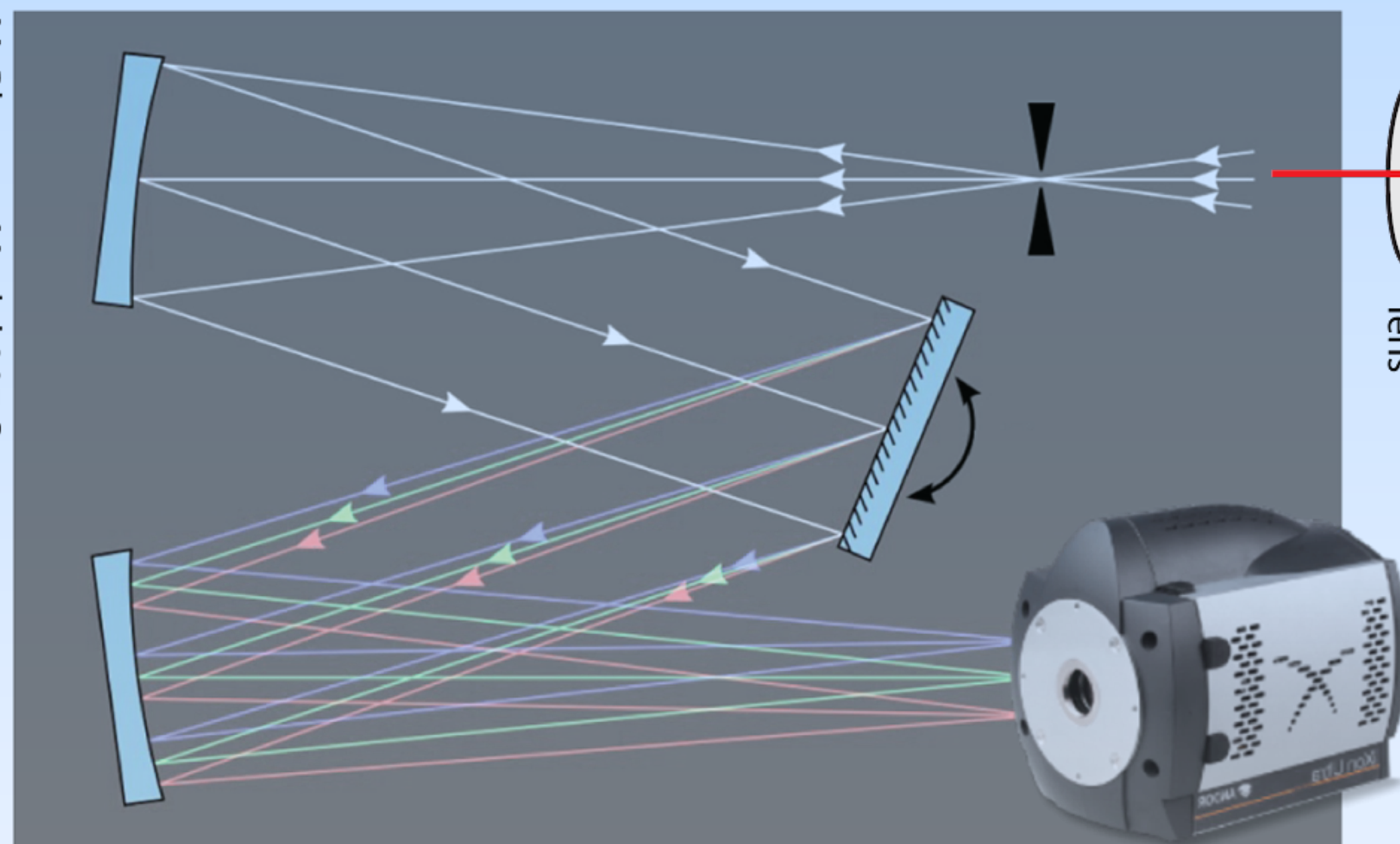
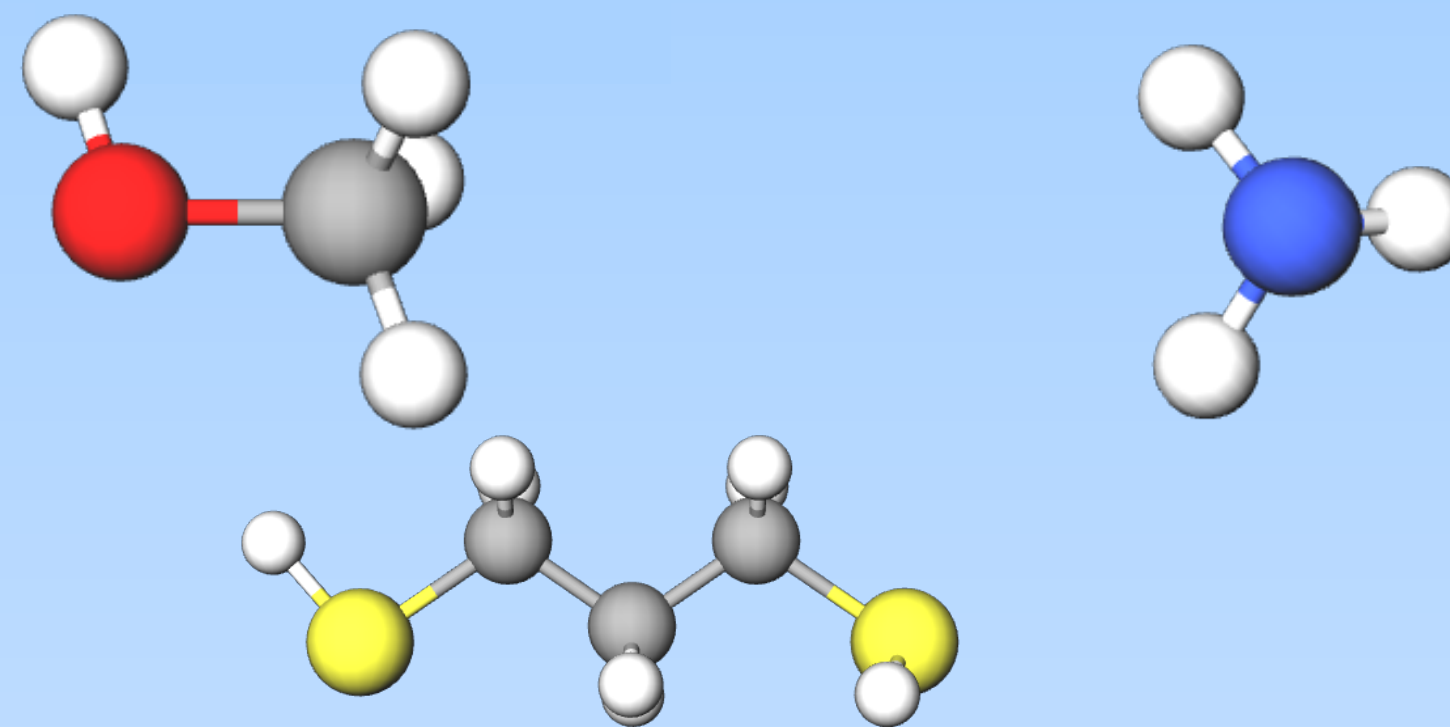
❖ Vibrational Closure: same concept as linear species



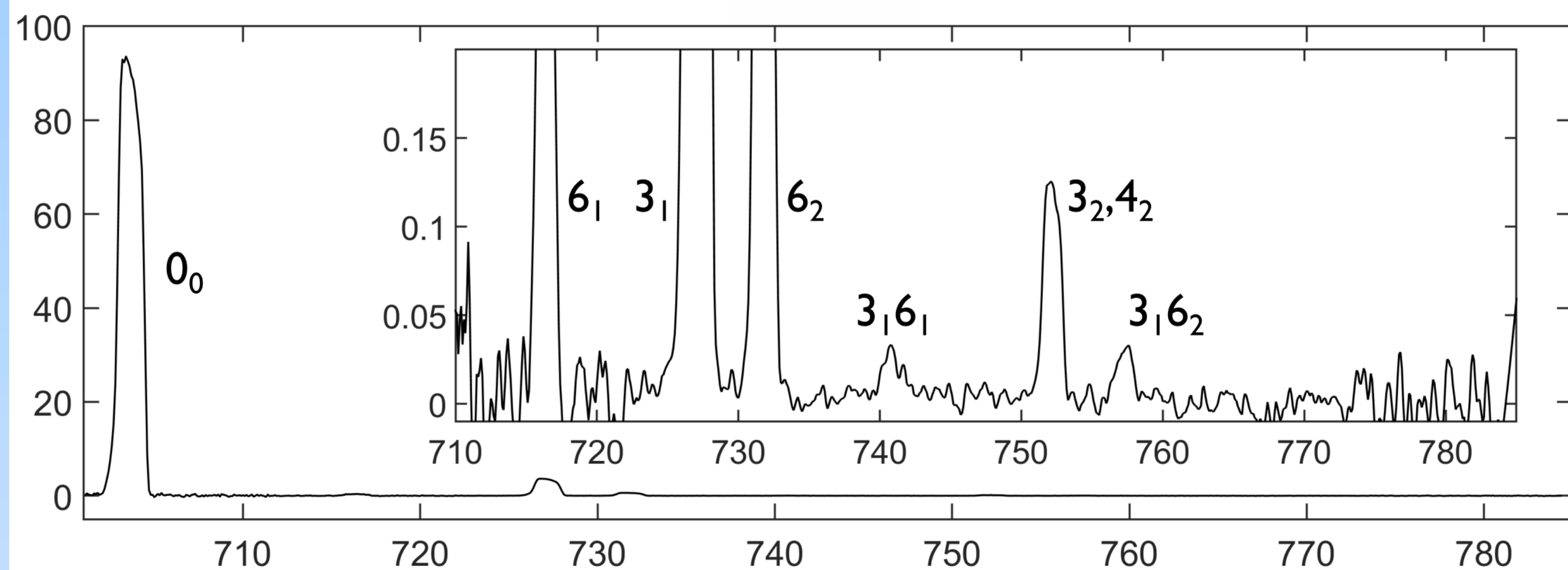
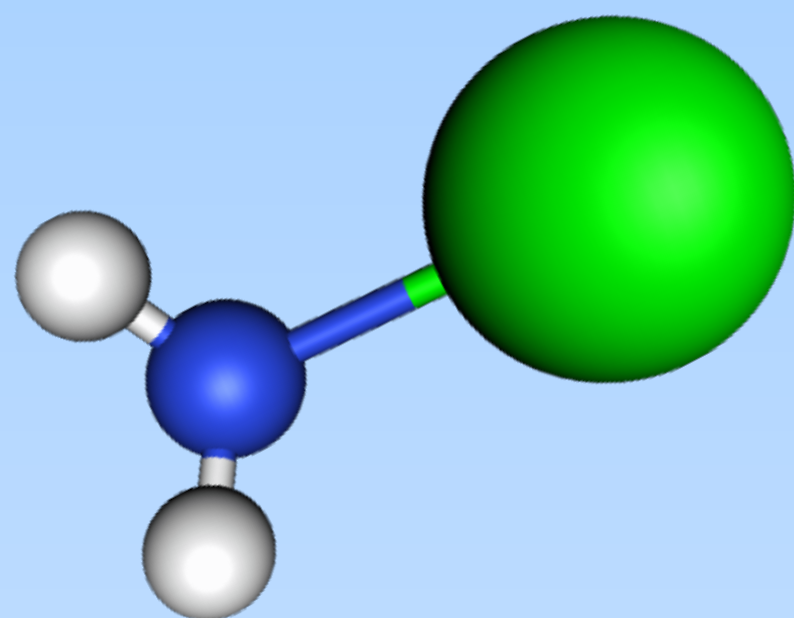
[Proposal for laser cooling of complex polyatomic molecules](#) I. Kozyryev, L. Baum, K. Matsuda, and J.M.D, ChemPhysChem 17, 3641 (2016).
[Polyatomic Candidates for Cooling of Molecules with Lasers from Simple Theoretical Concepts](#), Isaev and Berger, Phys. Rev. Lett. **116** 2016
[Molecular Asymmetry and Optical Cycling: Laser Cooling Asymmetric Top Molecules](#) B. L. Augenbraun, J.M.D., T. Zelevinsky, I. Kozyryev, Phys. Rev. X 10, 031022 (2020).

Apparatus for Measuring Branching Ratios

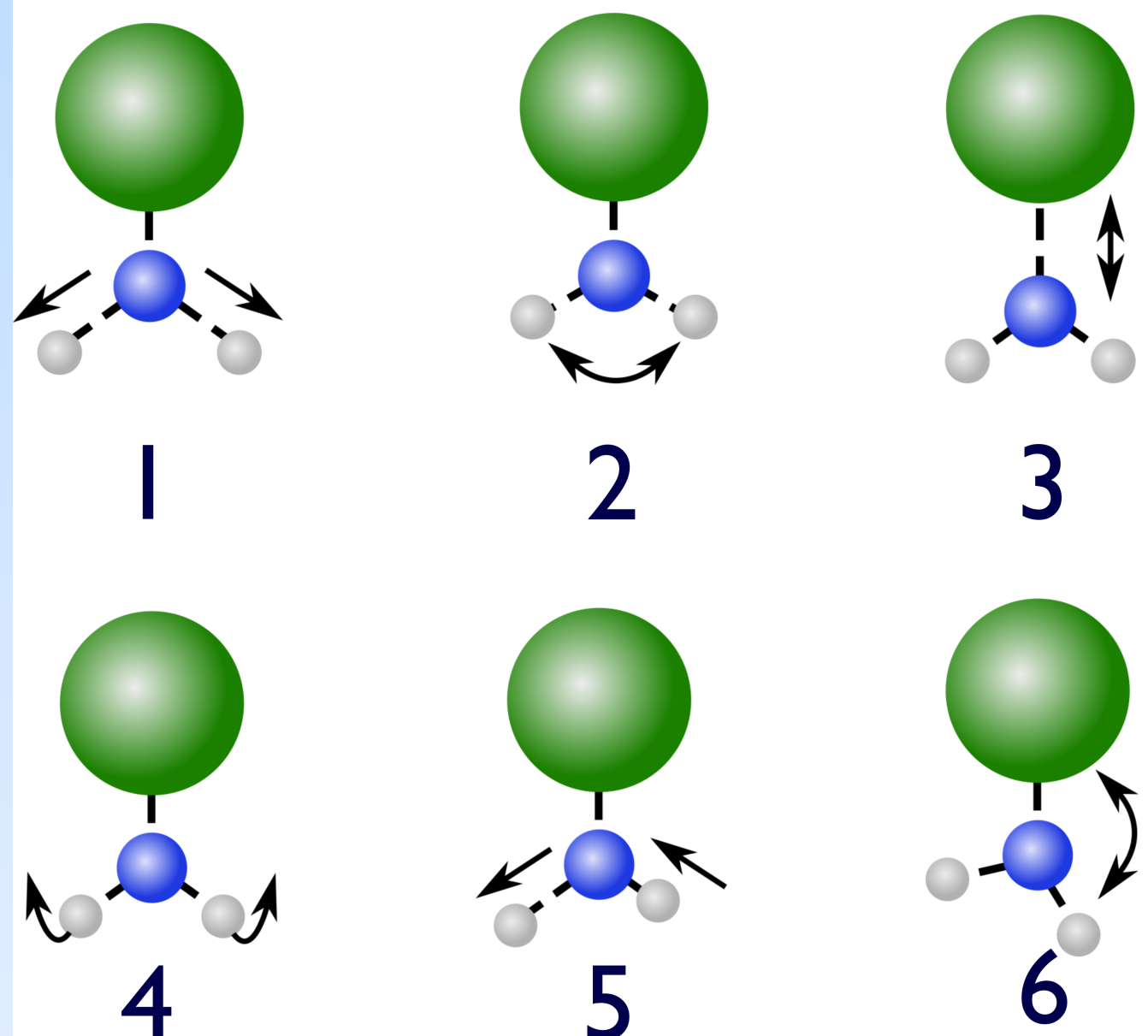
- ❖ Modified cryogenic buffer gas beam source with in-vacuum optics and collection into a Czerny-Turner style interferometer
- ❖ Different reactants for different molecules: methanol, ammonia, and 1,3-propanedithiol



Vibrational Branching Results SrNH_2



Modes



State	Energy (cm^{-1})		VBR (%)	
	Calc.	Exp.	Calc.	Exp.
0_0	0	0	94.024	95.1(2)
6_1	260	252(4)	0.351	0.35(3)
3_1	454	459(1)	4.627	3.7(2)
6_2	531	547(2)	0.869	0.63(3)
3_16_1	699	717(9)	0.019	0.04(1)
3_2	904	918(4)	0.048	0.11(1)
4_2	888	~ 918	0.012	0.02(1)
3_16_2	965	1007(2)	0.030	0.03(1)
2_1	1540	—	0.014	—
1_1	3331	—	0.003	—

Notes:

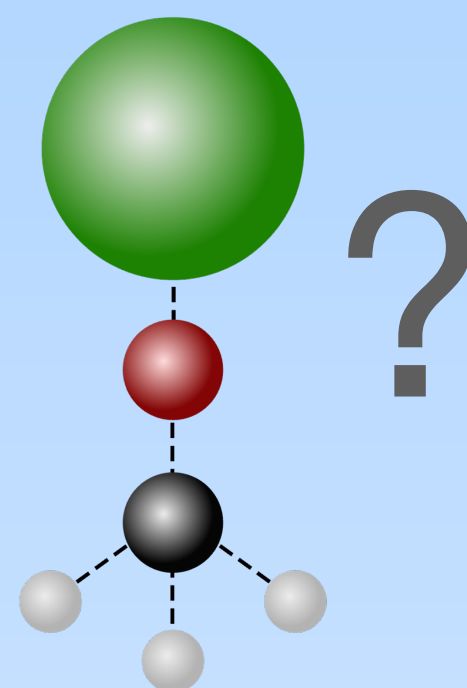
- ❖ 8 states populated at the 0.01% level (5 in SrOH)
- ❖ More atoms $\Rightarrow$ more modes
- ❖ Bending mode is split – but only one of the two is active
- ❖ Resonances in ground state affect strengths, but not # of channels
- ❖ Similar complexity to SrOH : only 1 more repump!

Conclusions and Outlook Nonlinear Sr Molecules For Laser Cooling

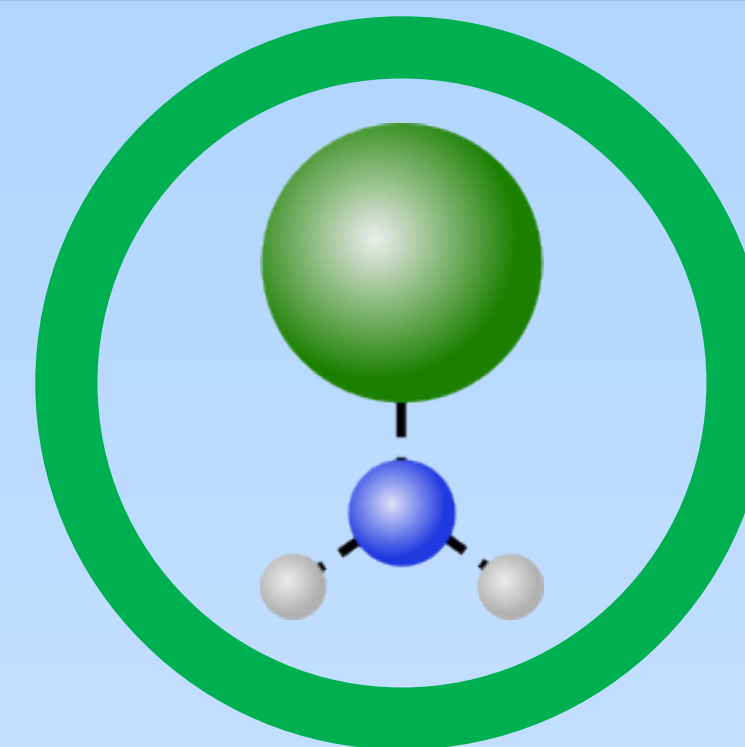
❖ Analyzed all three molecules' vibrational branching to $<0.1\%$
-Varies from 3-15+ frequencies to 1D Sisyphus cool

❖ Combination of VBRs & rotational closure point to SrNH_2 ❖ as the simplest of the three

❖ Future measurements of parts in 10^5 VBRs are well motivated to identify any serious perturbations below current sensitivity early

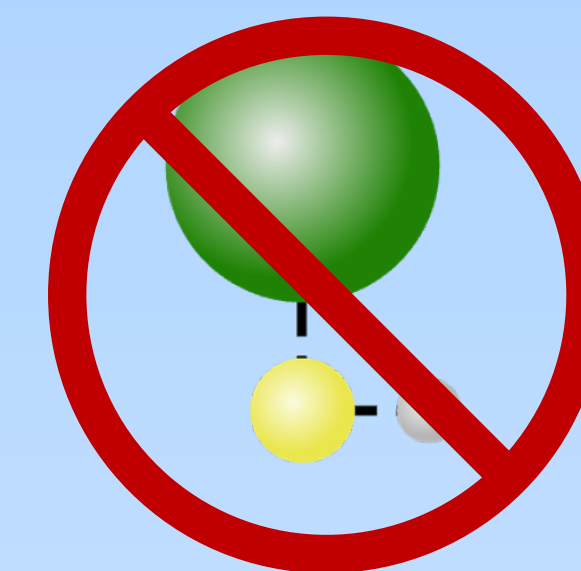


“Nonideal” rotational closure



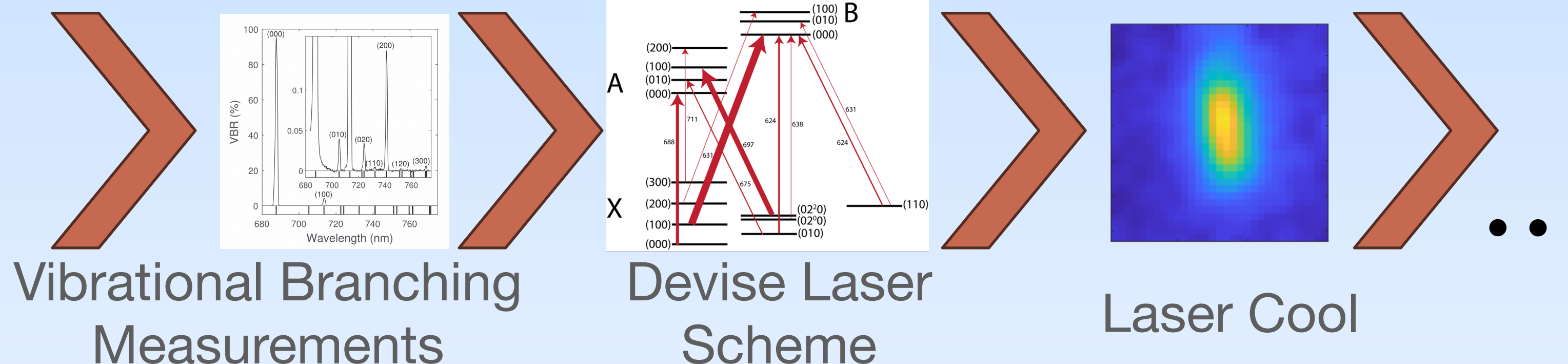
❖ Manageable number of modes

❖ Rotational closure(!)



❖ Far too many modes

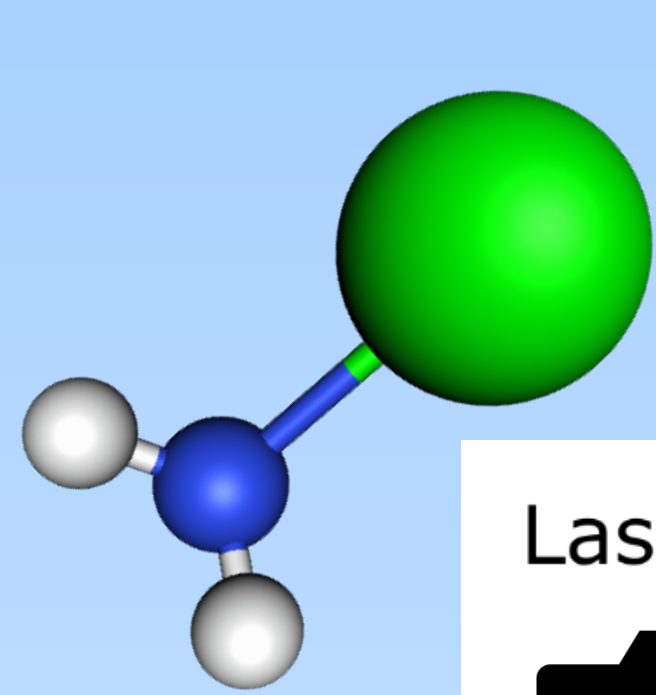
❖ Very poor rotational closure



Vibrational Branching Ratios for Laser-Cooling of Nonlinear Strontium-Containing Molecules

[Alexander Frenett](#), [Zack Lasner](#), [Lan Cheng](#), [J.M.D.](#) Phys. Rev. A 110, 022811 (2024)

I-D Laser Cooling of an **Asymmetric Top** Molecule



CaNH₂ Following the pioneering lead of SrF, CaF, CaOH, CaOCH₃ etc. we have demonstrated...

... radiative force deflection and...

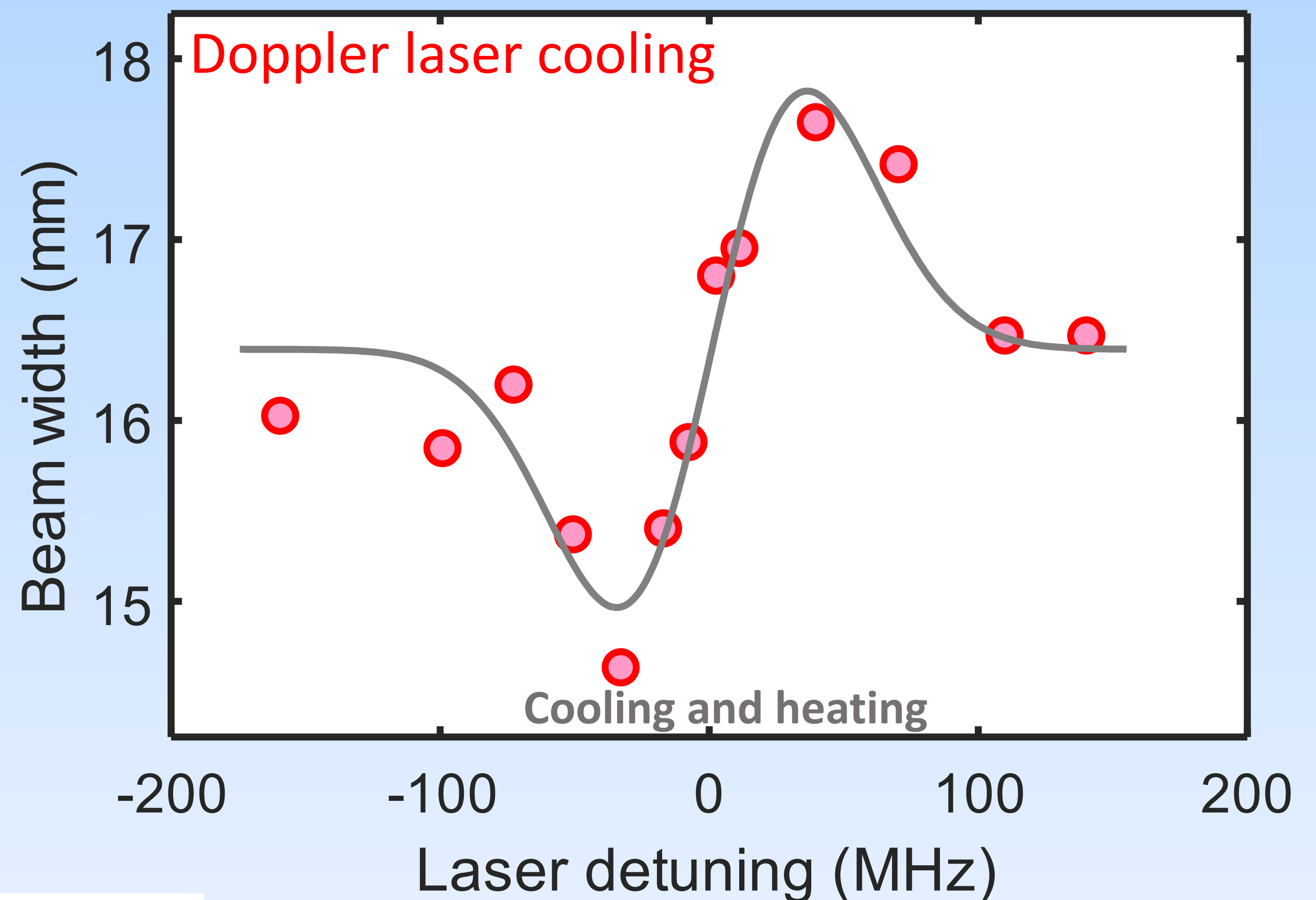
Laser-induced fluorescence (LIF)



Laser

Direction of
Molecule
Flow

Lasers



DAMOP

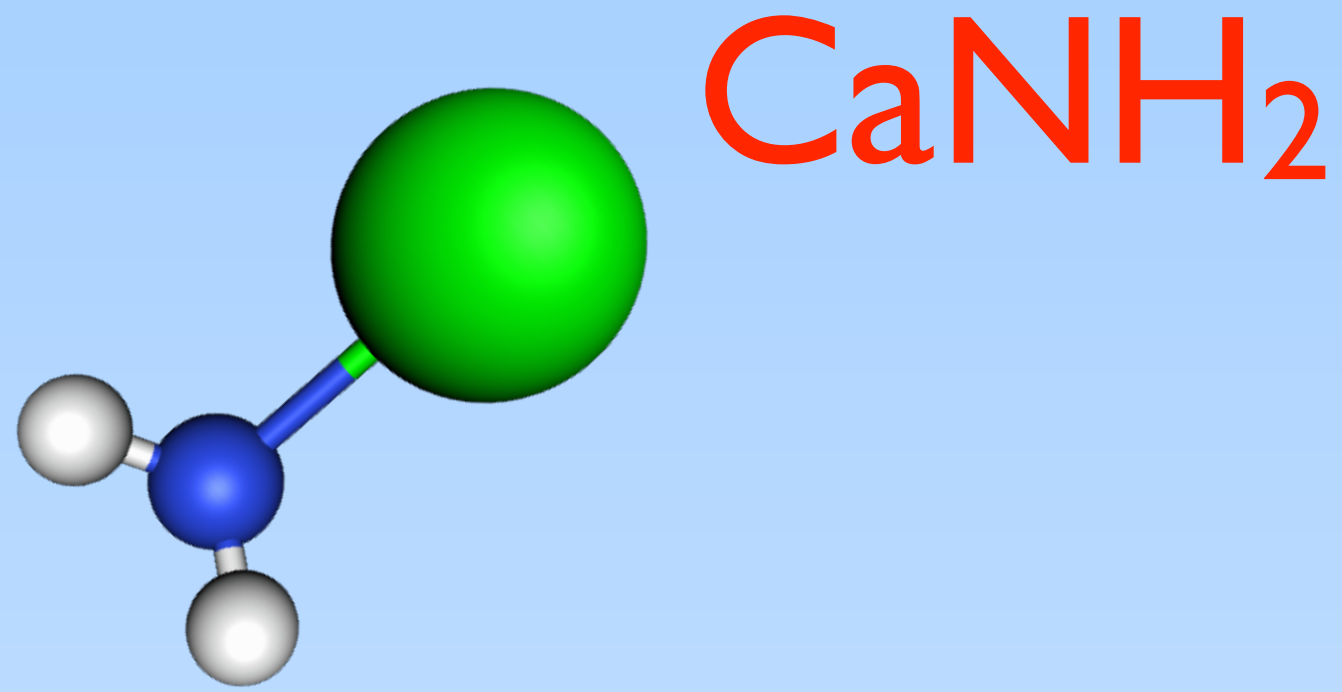
Laser Cooling of Molecules

9:00 AM–9:12 AM, Wednesday, June 5, 2024

Andrew Winnicki

1D Laser Cooling of the Asymmetric Top Molecule CaNH₂

Towards a MOT of an **AS**ymmetric Top Molecules



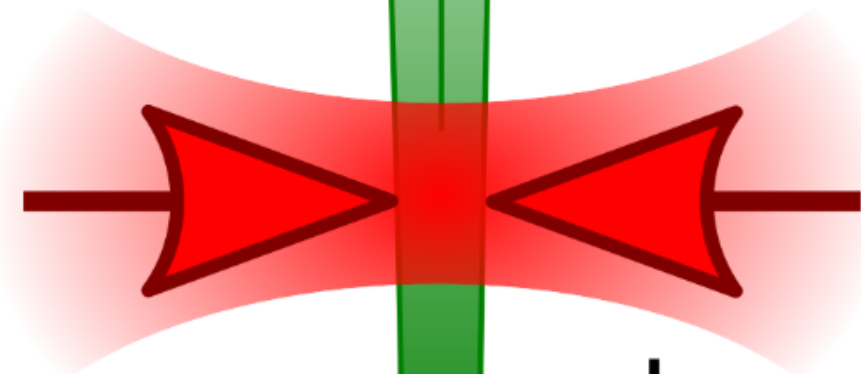
We are “going for it”!

Laser-induced fluorescence (LIF)

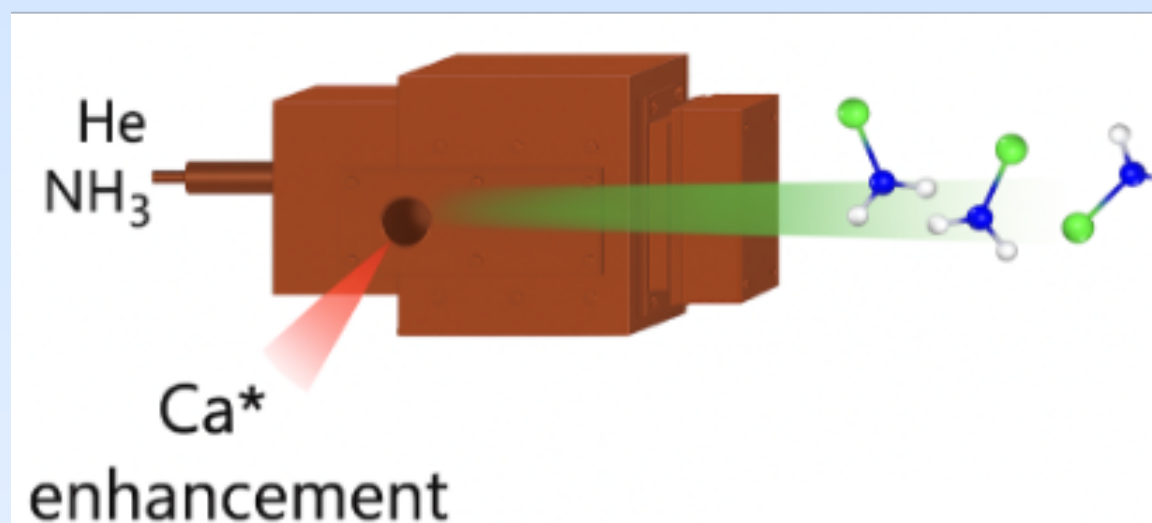
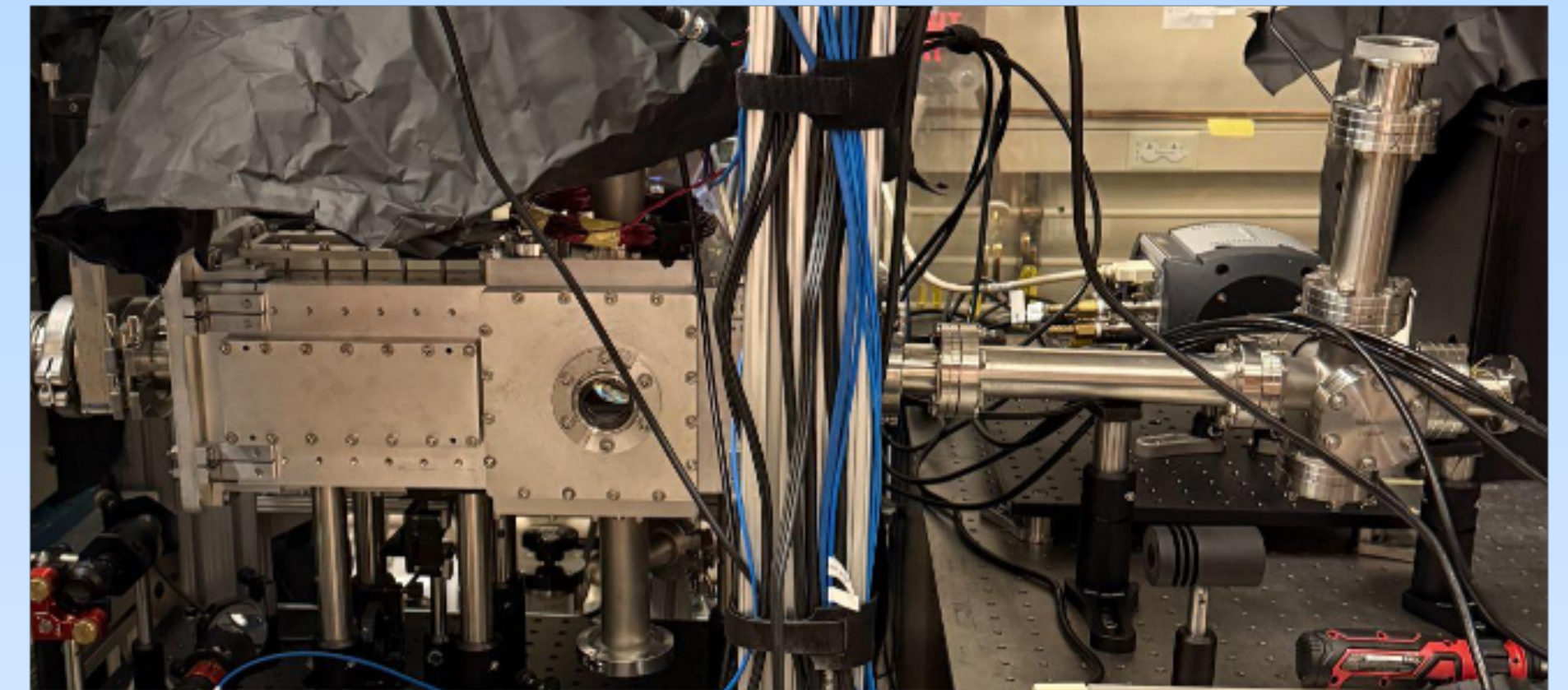
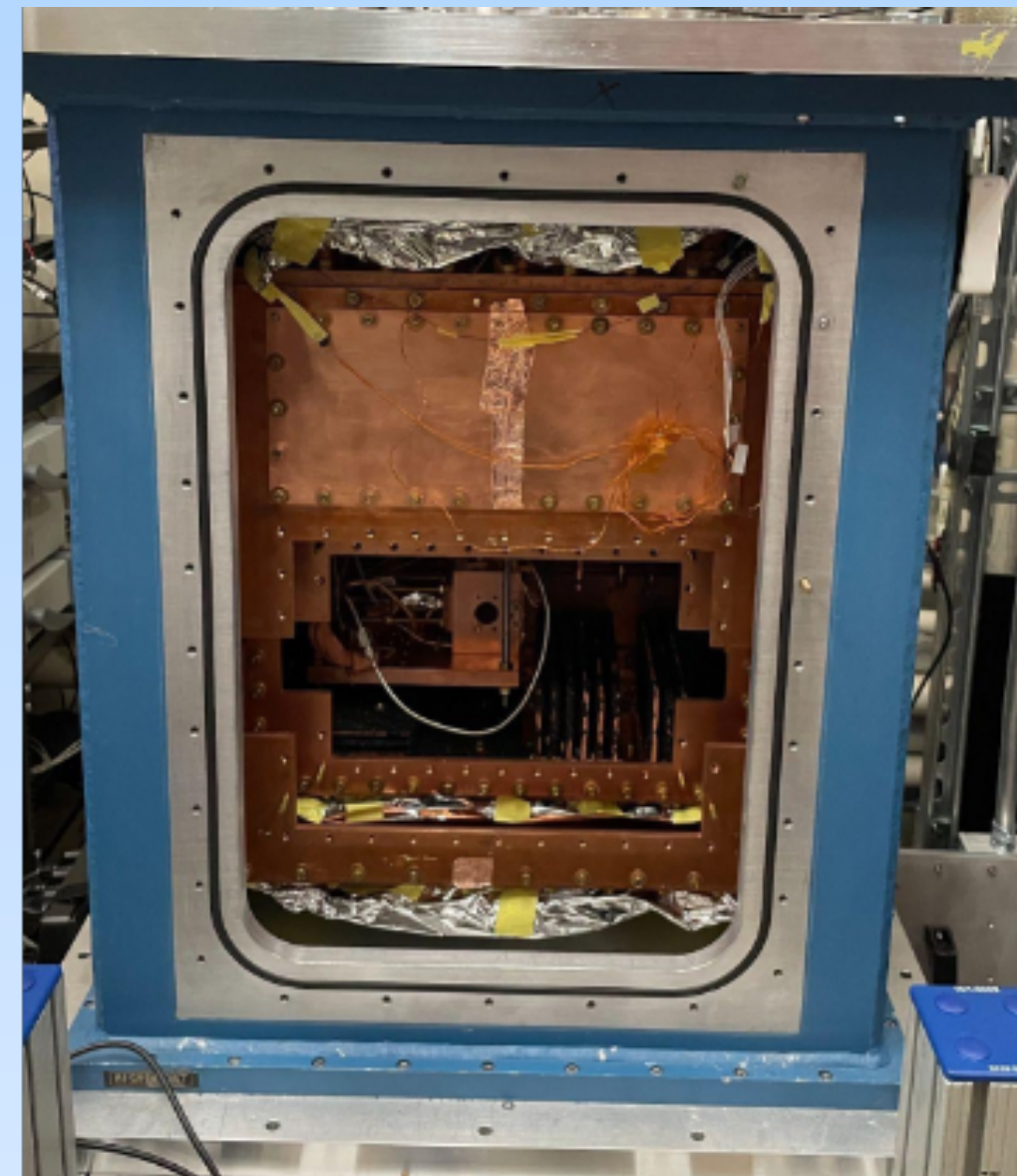


Laser

Direction of
Molecule
Flow



Lasers

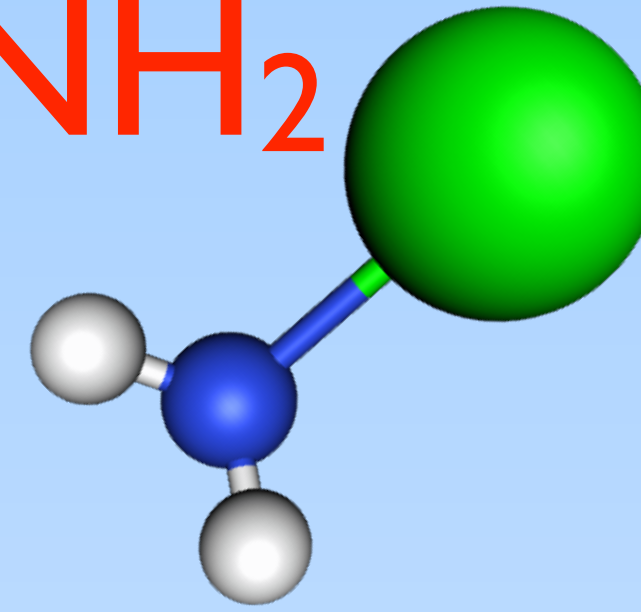


Molecule beam

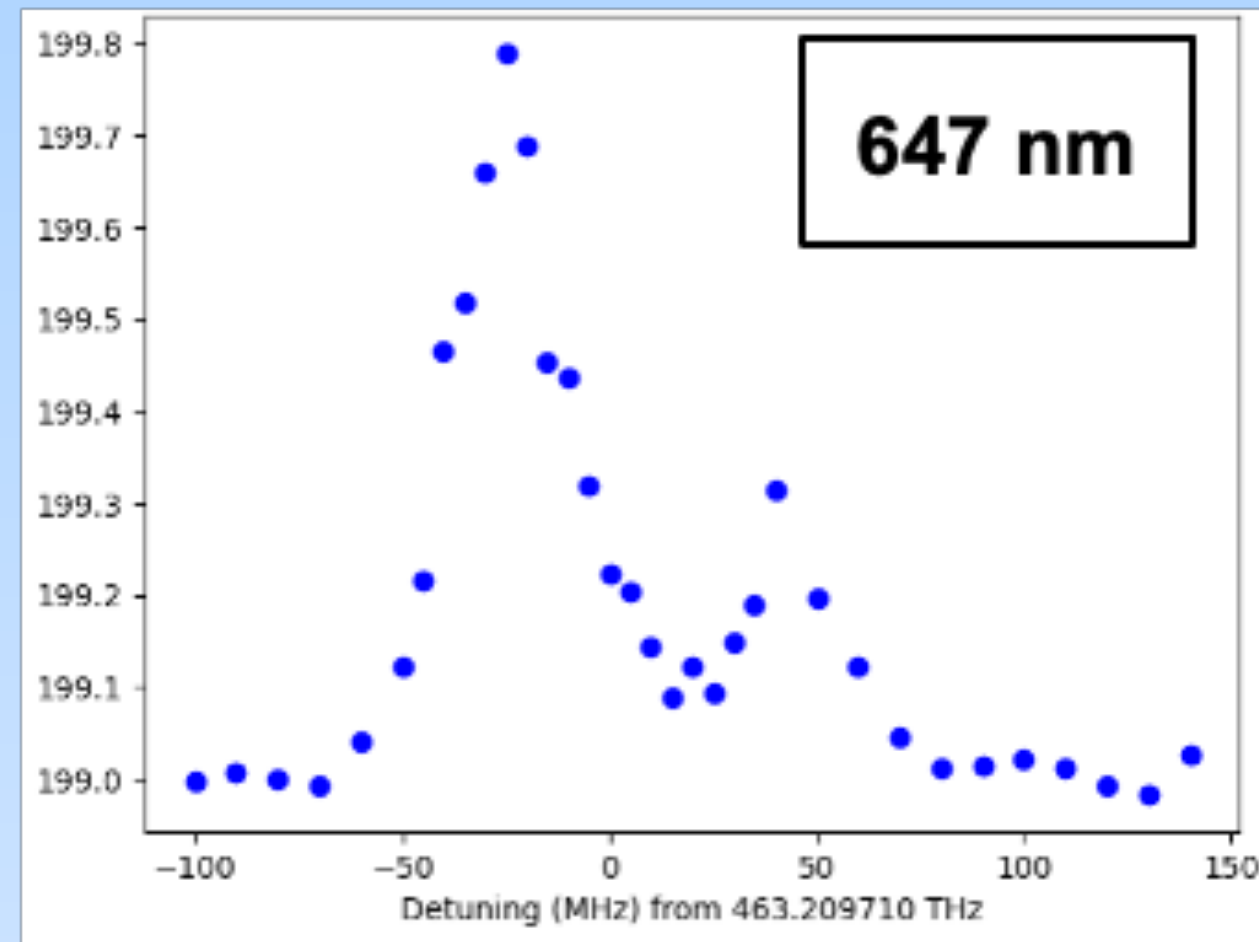


Towards a MOT of an **A**Symmetric Top Molecules

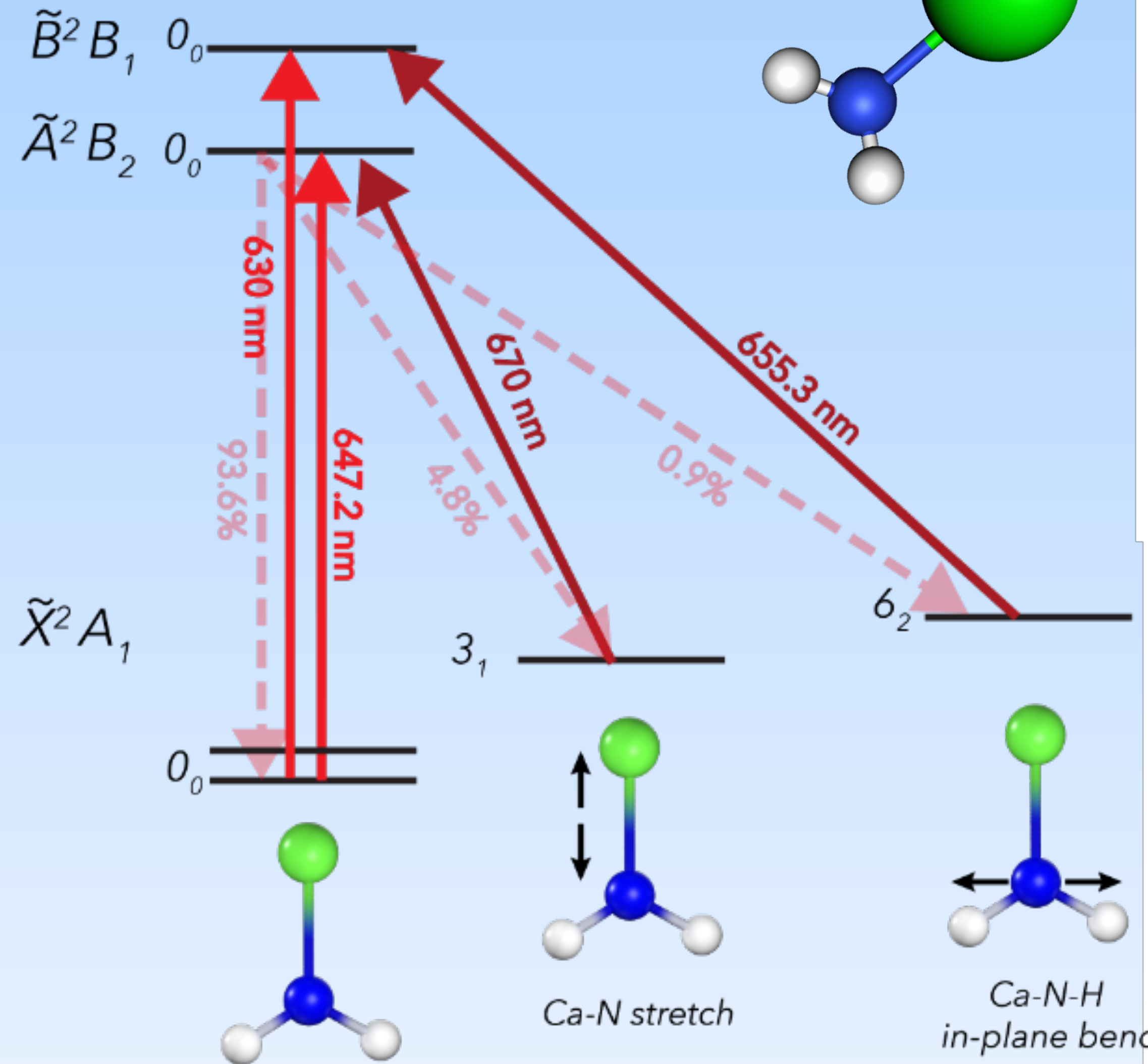
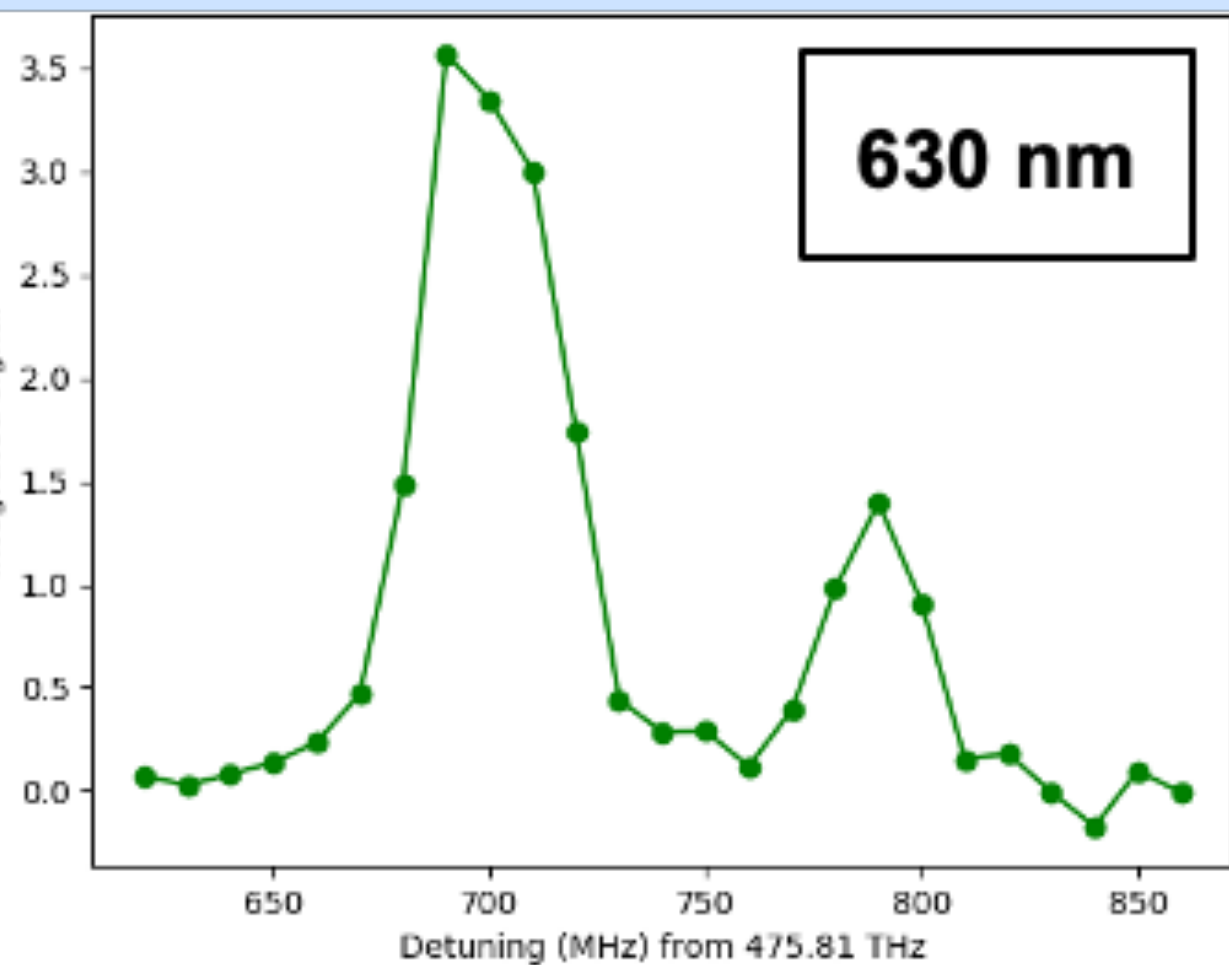
CaNH₂



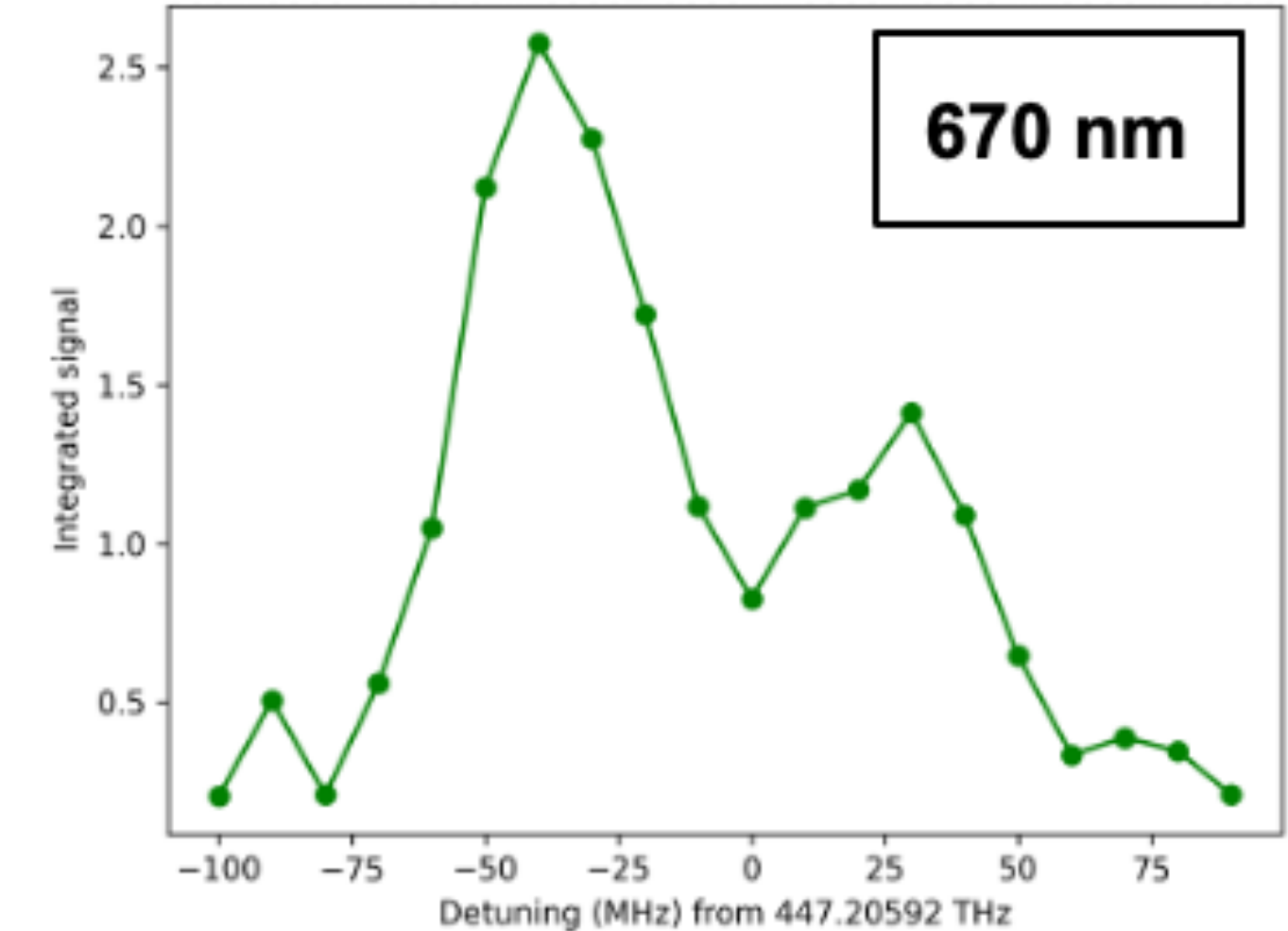
Mainline transition



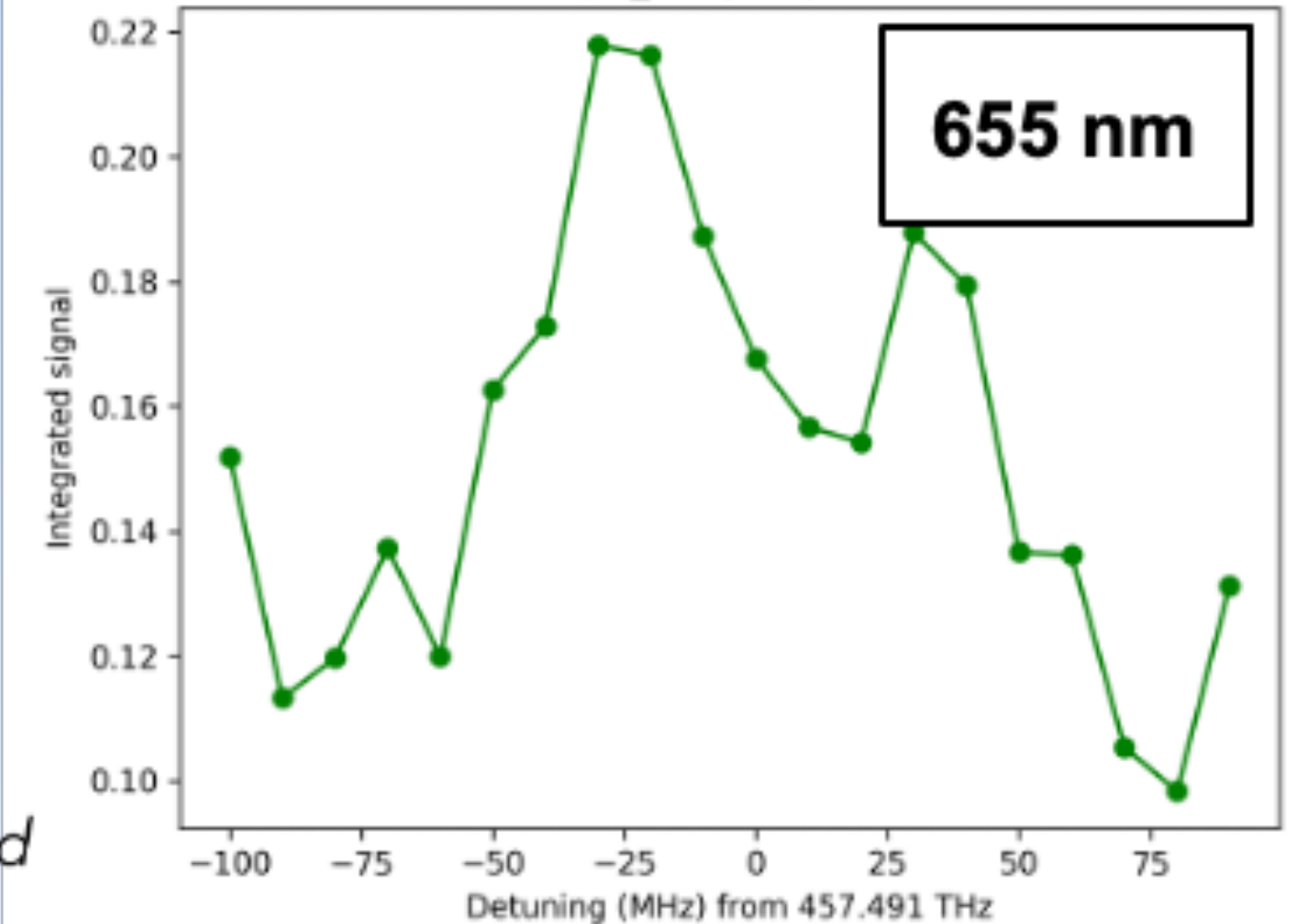
$0_0, 2_{11}$ repump



CaNH₂ 3_1 repump SR scan

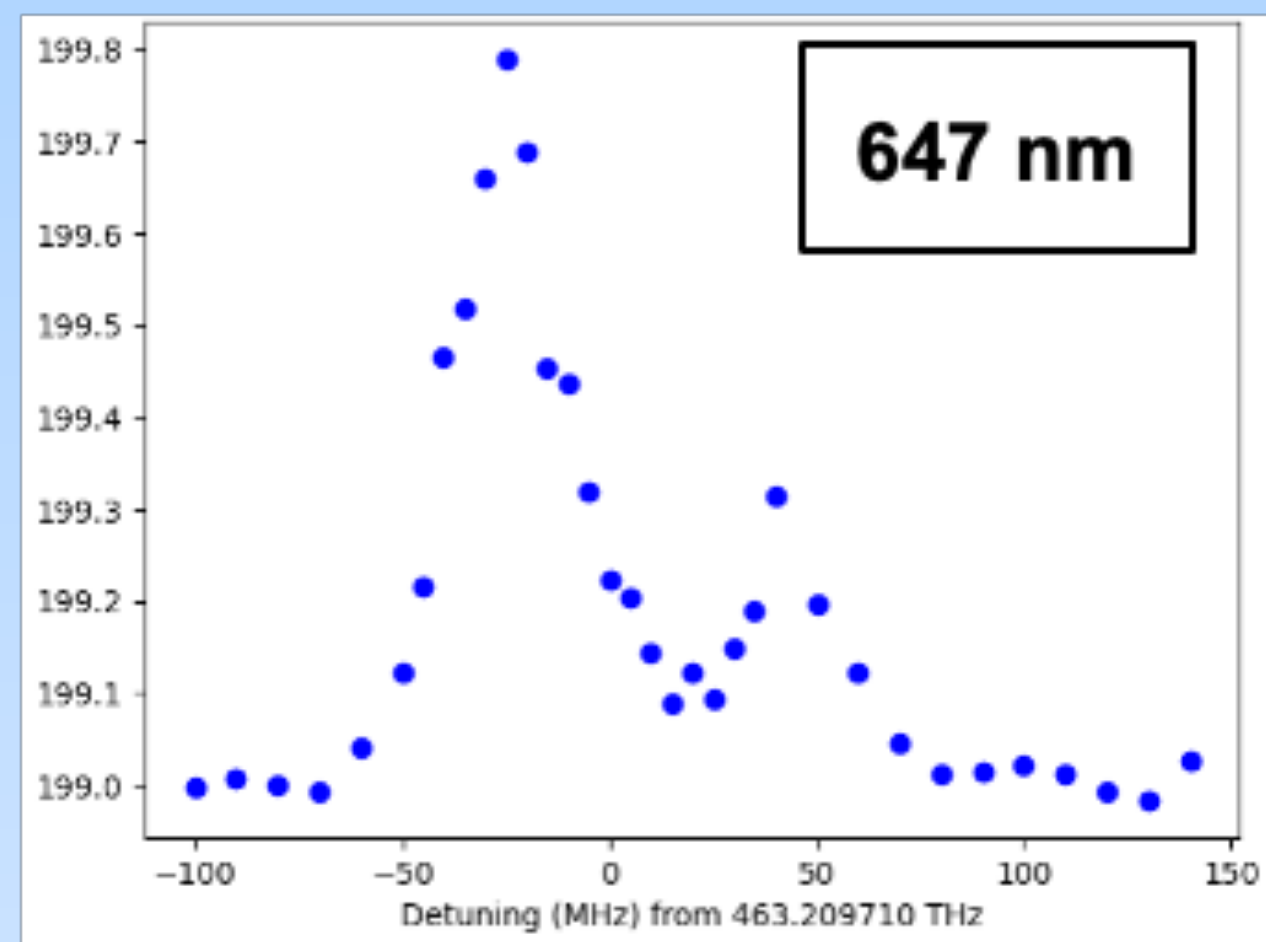


CaNH₂ 6_2 repump SR scan

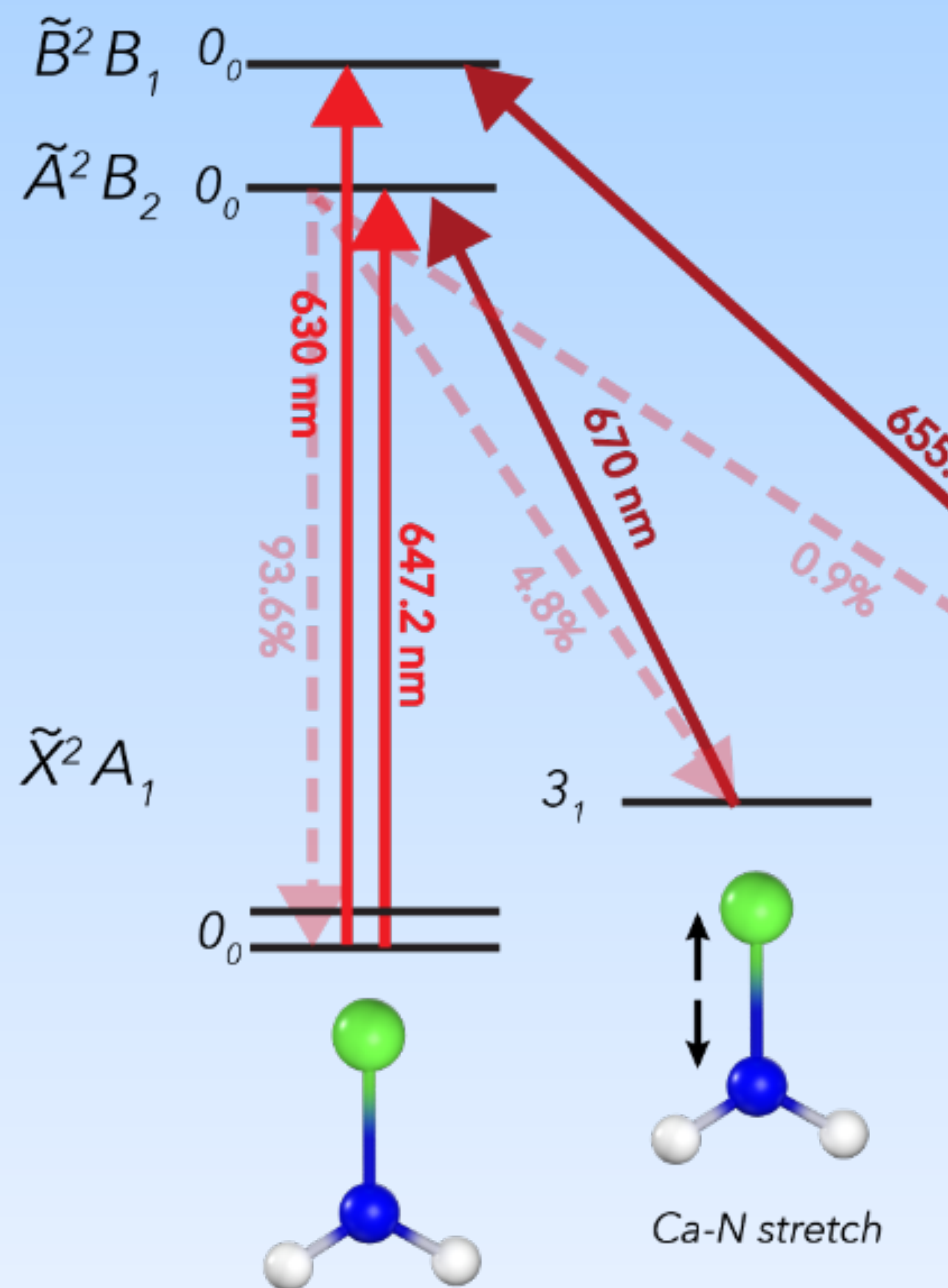
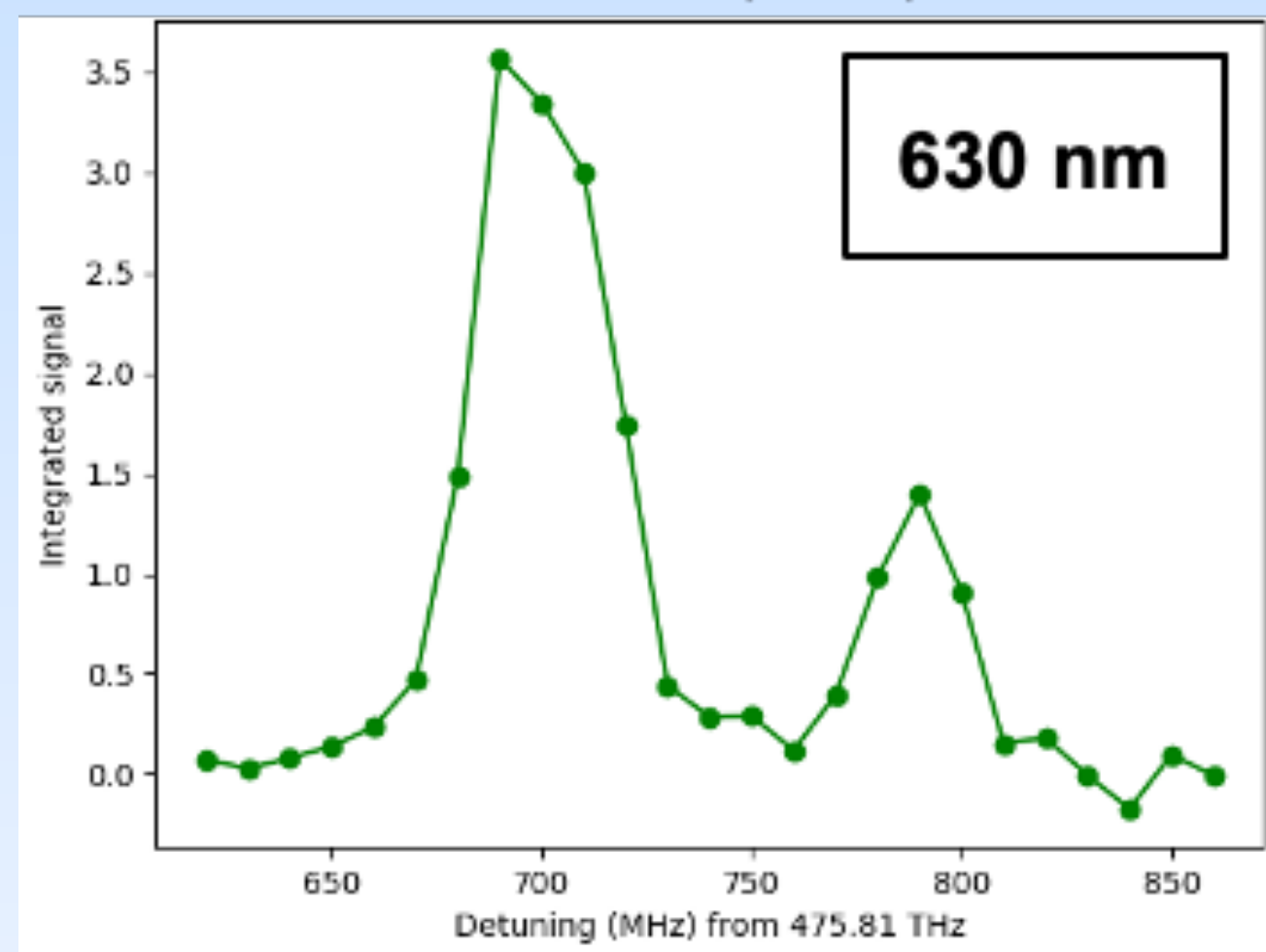


Towards a MOT of an Asymmetric Top Molecules

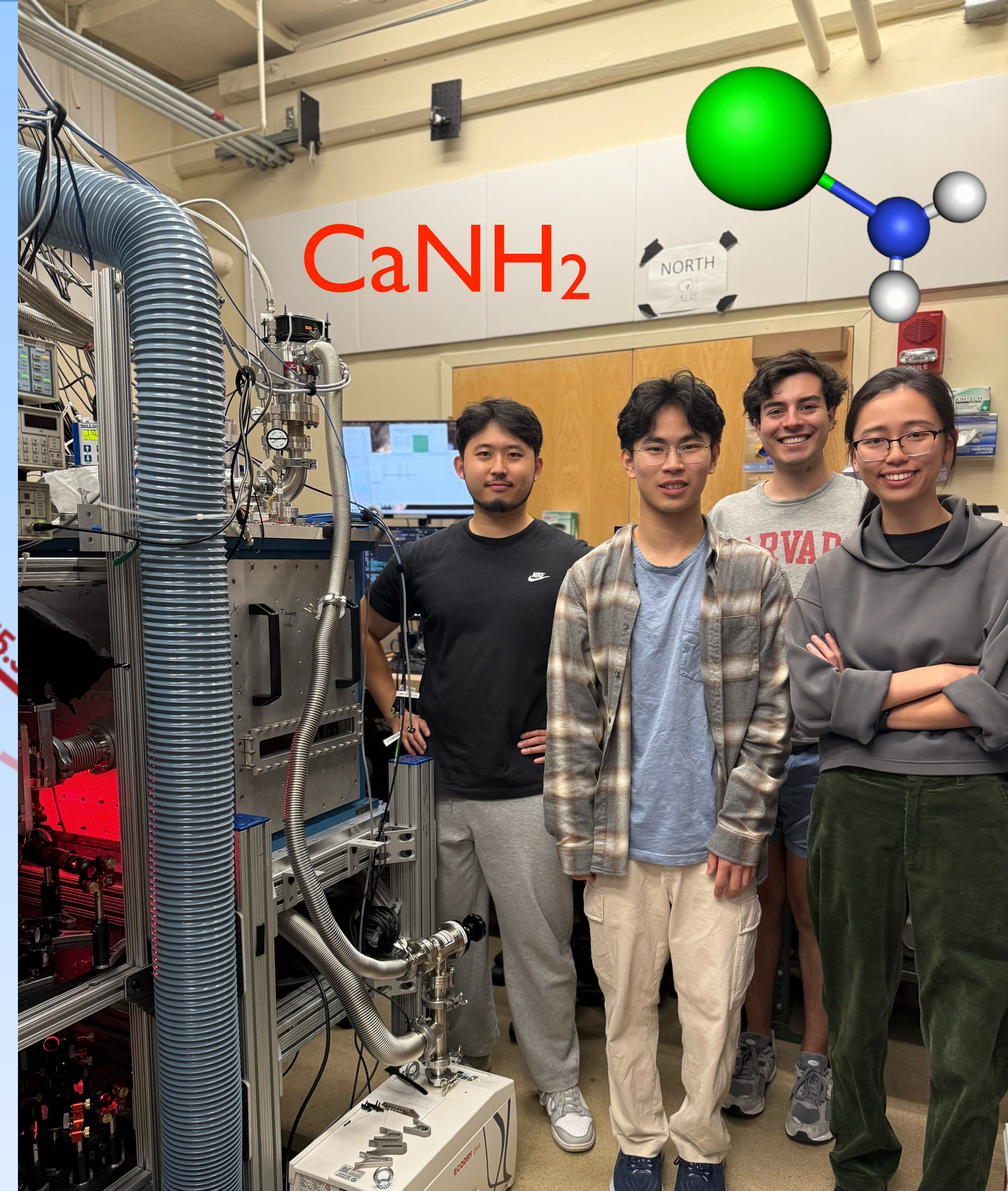
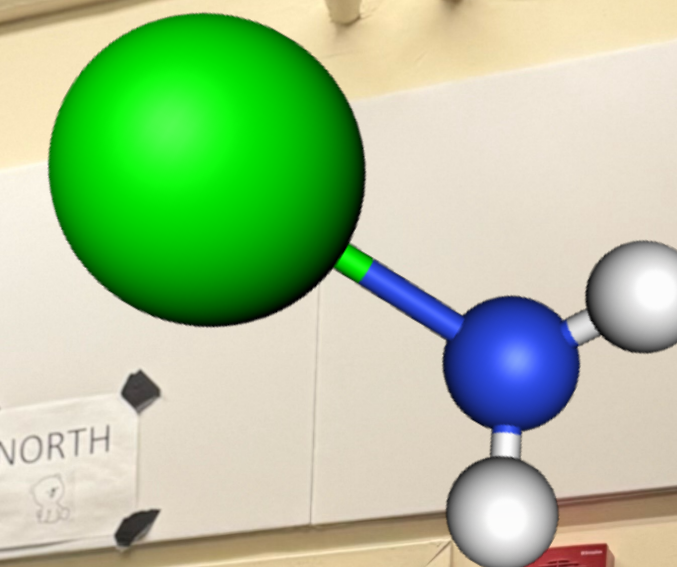
Mainline transition



$0_0, 2_{11}$ repump



CaNH_2



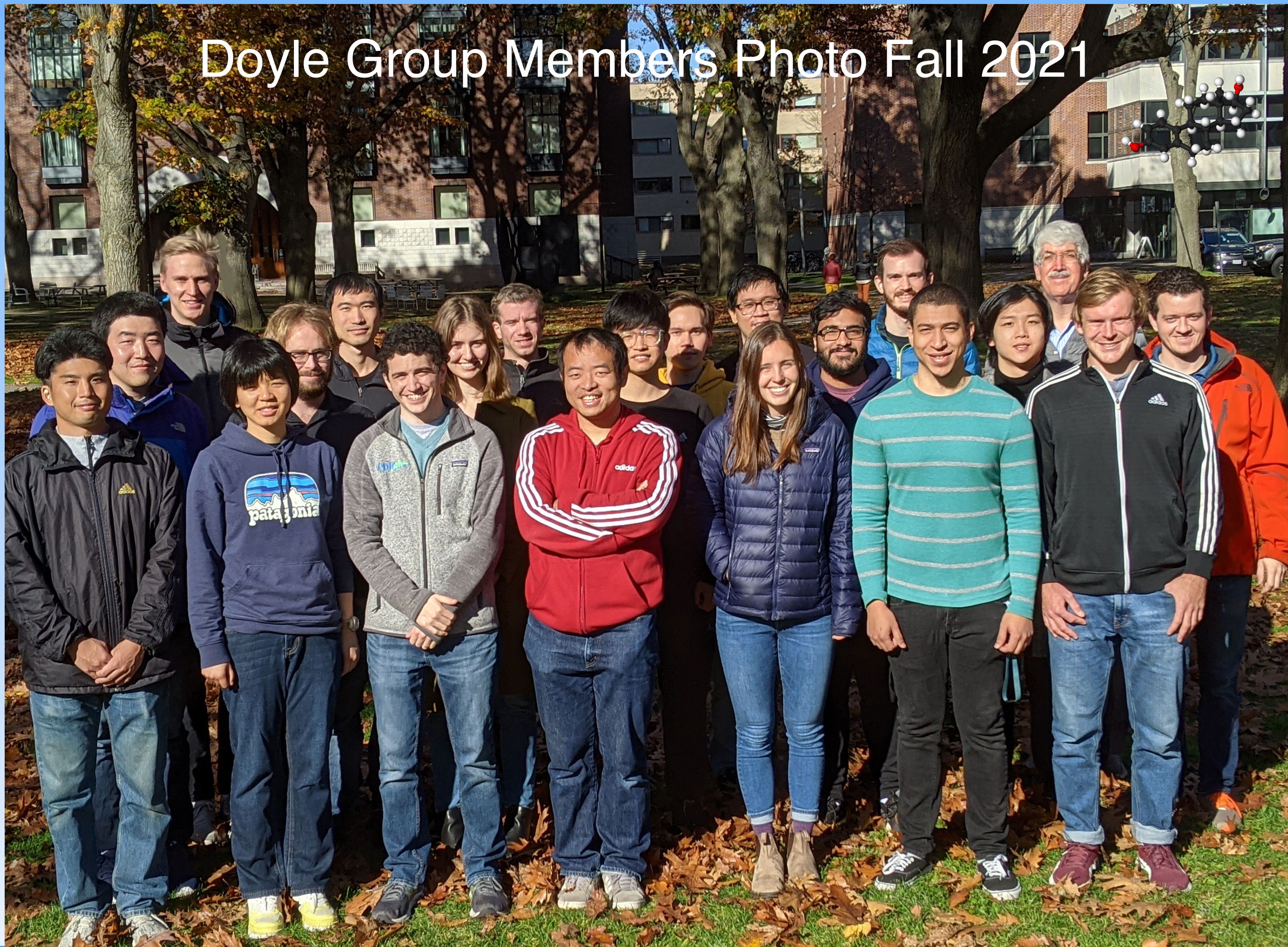
Giseok

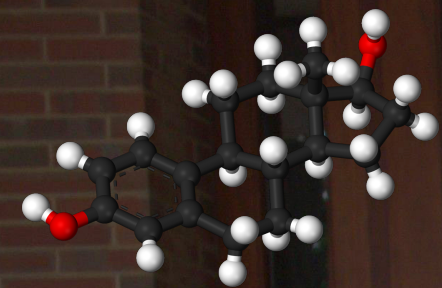
Winston

Jack

Grace

Doyle Group Members Photo Fall 2021





Doyle Group Members Photo Fall 2025

