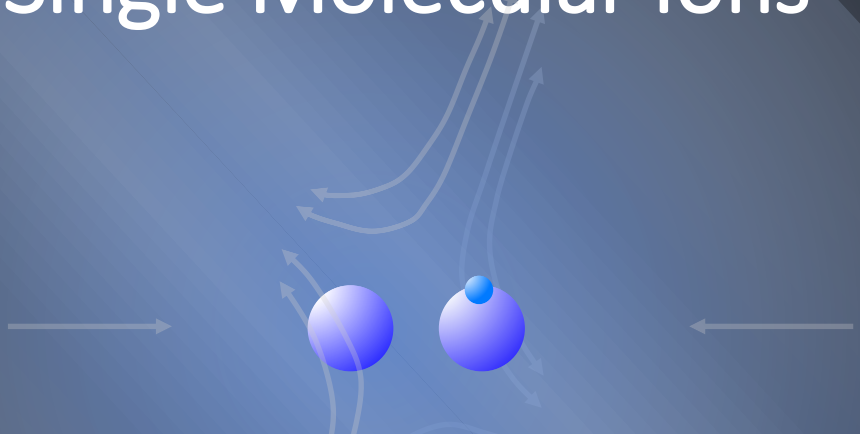
Quantum State Control and Precision Spectroscopy of Single Molecular Ions



Chin-wen (James) Chou[1,2]

Alejandra Collopy[1], Yiheng Lin[3], Christoph Kurz[1], Michael E. Harding[4], Philipp N. Plessow[4], Tara Fortier[1], Scott Diddams[2], Yu Liu[1,2], Zhimin Liu[1,2], Julian Schmidt[1,2,5], Dalton Chaffee[1,2],

Dietrich Leibfried[1,2], David R. Leibrandt[1,2,6]

[1]National Institute of Standards and Technology, [2]University of Colorado, USA

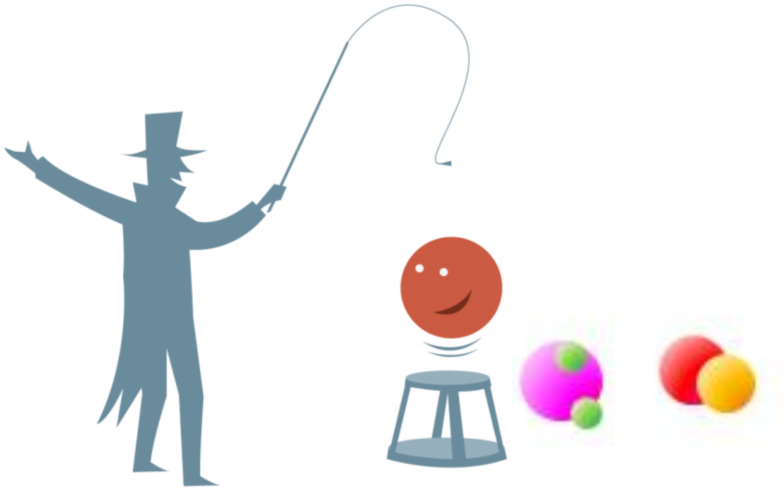
[3]University of Science and Technology of China, China, [4]Karlsruhe Institute of Technology, Germany

[5]Paul Scherrer Institute, Switzerland [6]University of California, Los Angeles, USA

Motivation

A general protocol for coherent quantum control of molecular ions

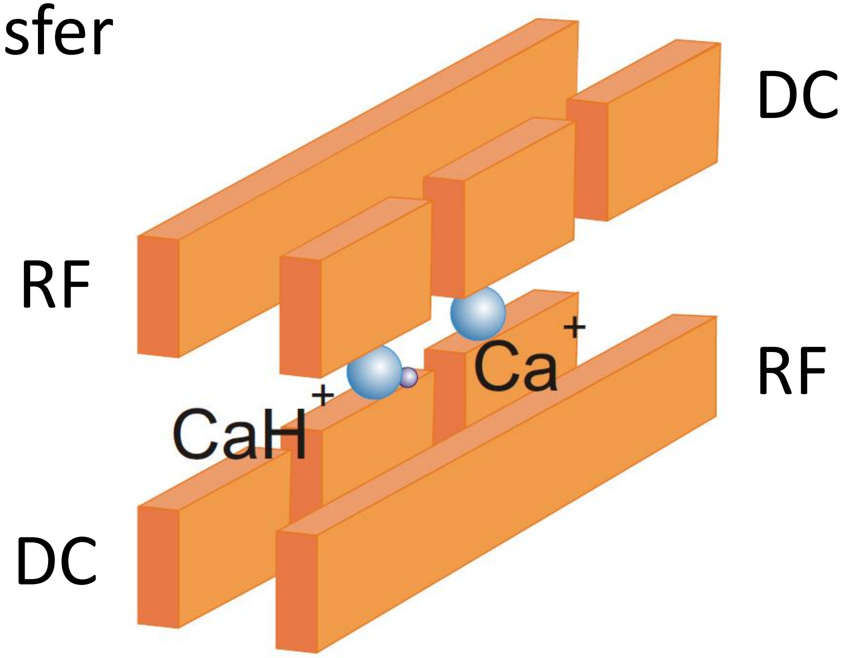
Exploiting the AMO toolbox and reach the same level of **control** and measurement **precision** for **many species** of molecular ions as on atomic ions.



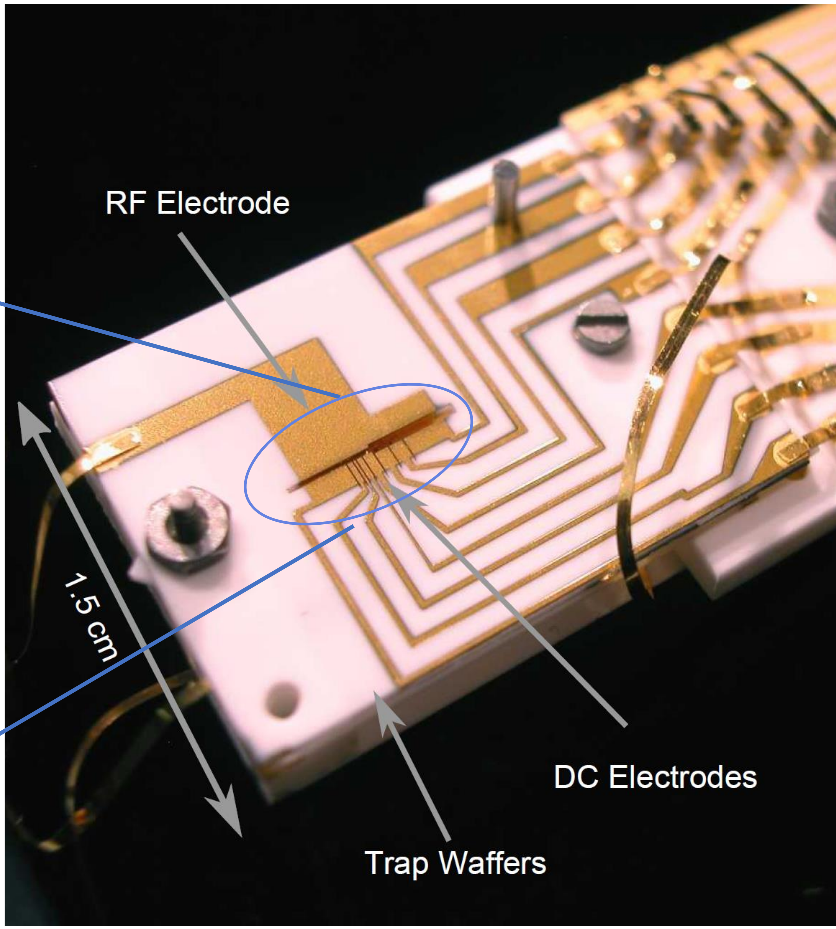
Credit: nobelprize.org

Quantum-Logic Spectroscopy of $^{40}CaH^+$

- Convenient to work with $^{40}Ca^+$
- Form a $^{40}CaH^+$ by trapping two $^{40}Ca^+$ ions and leaking in H_2
- Coupled motion allows sympathetic cooling and information transfer



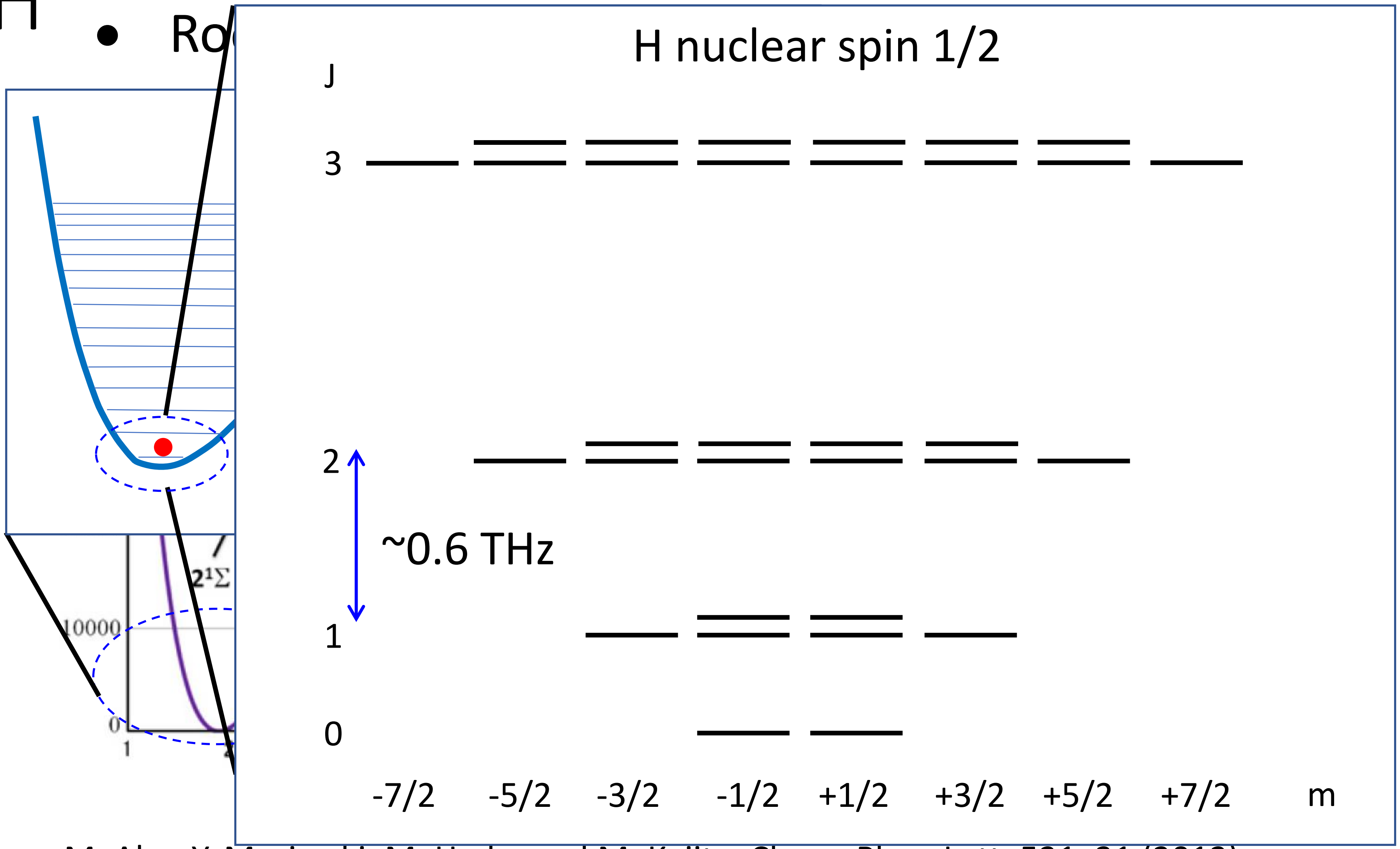
P.O. Schmidt, et al. Science 309, 749 (2005)



Credit: John Jost thesis

Proof-of-principle molecular ion $^{40}CaH^+$

- Ro



M. Abe, Y. Moriwaki, M. Hada, and M. Kajita, Chem. Phys. Lett. 521, 31 (2012)

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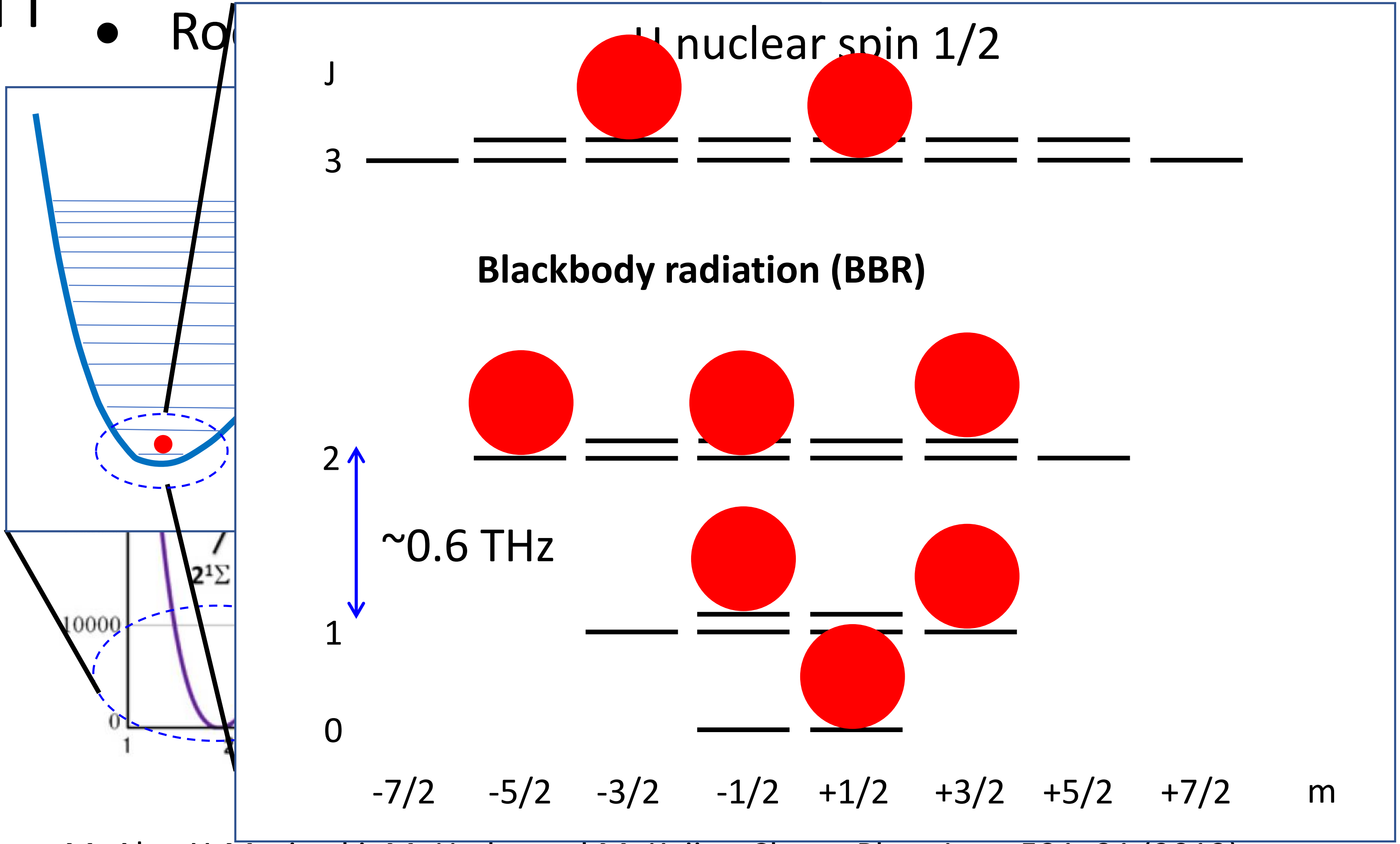

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Proof-of-principle molecular ion $^{40}CaH^+$

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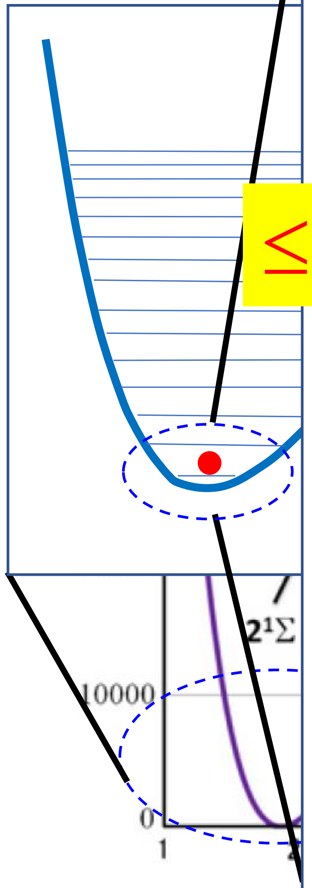
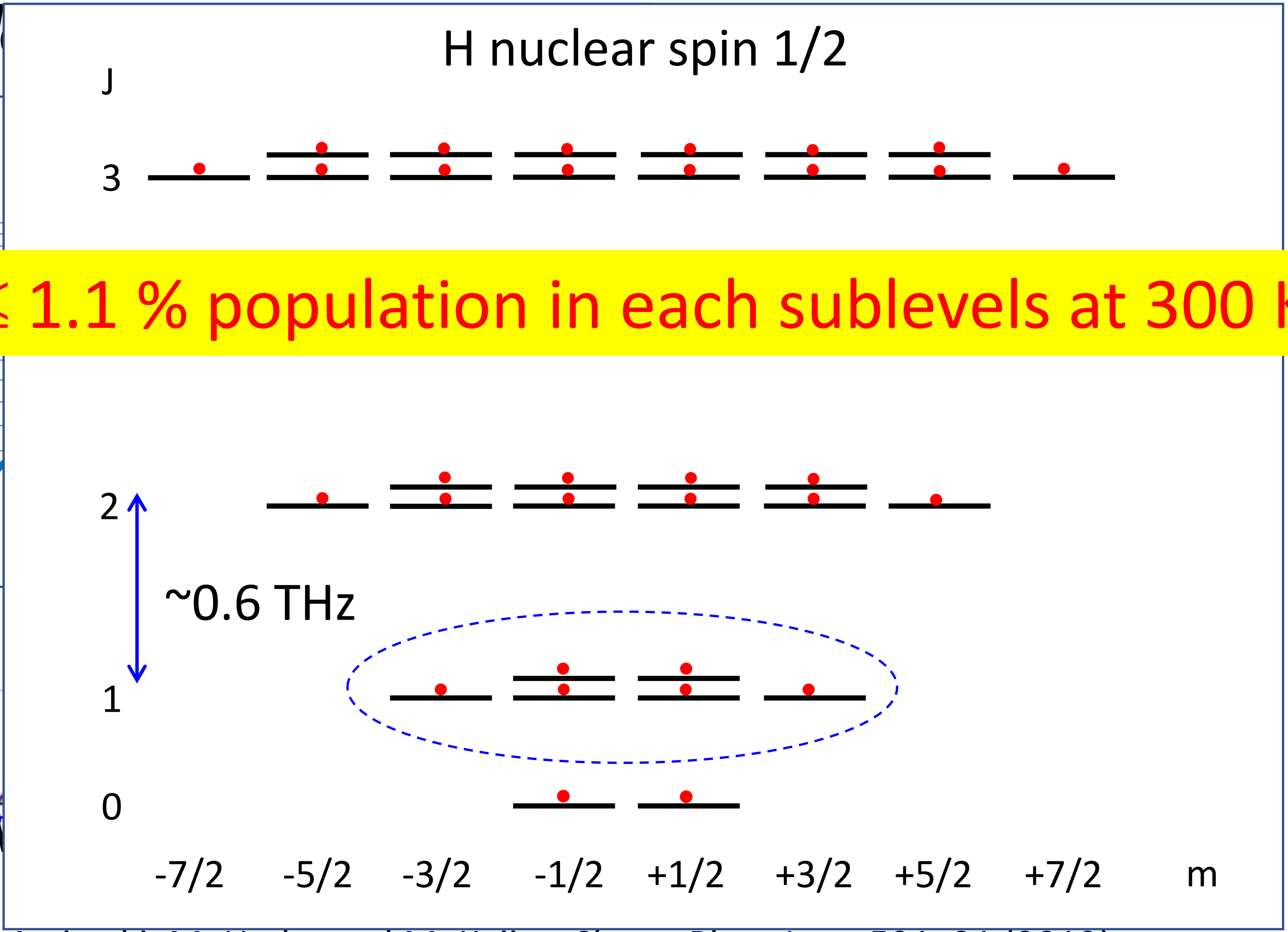
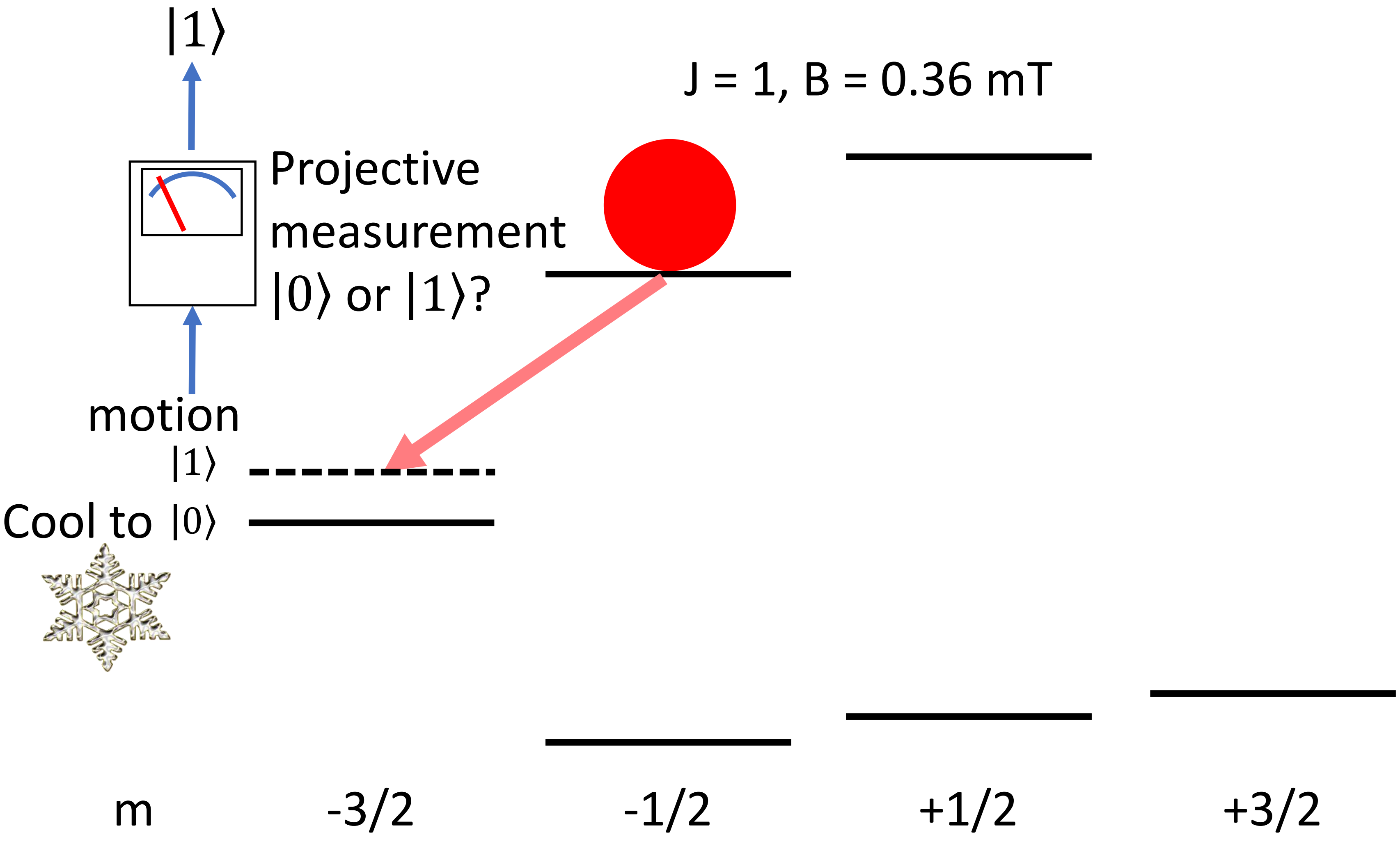




≤ 1.1 % population in each sublevels at 300 K

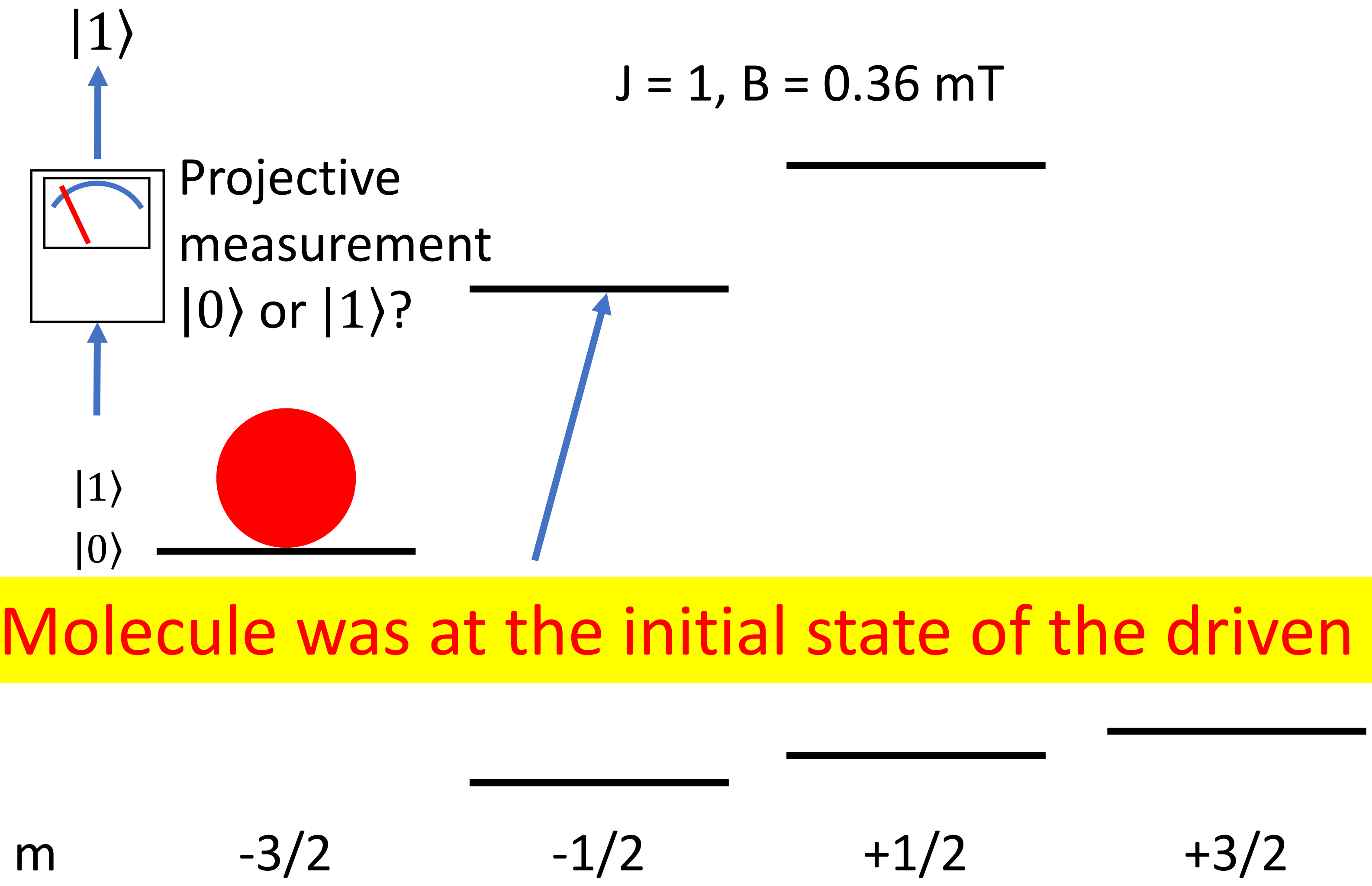
M. Abe, Y. Moriwaki, M. Hada, and M. Kajita, Chem. Phys. Lett. 521, 31 (2012)

Projecting into a Pure State



I. S. Vogelius, L. B. Madsen, and M. Drewsen, J. Phys. B, 39, S1259 (2006). Nature **545**, 203 (2017)

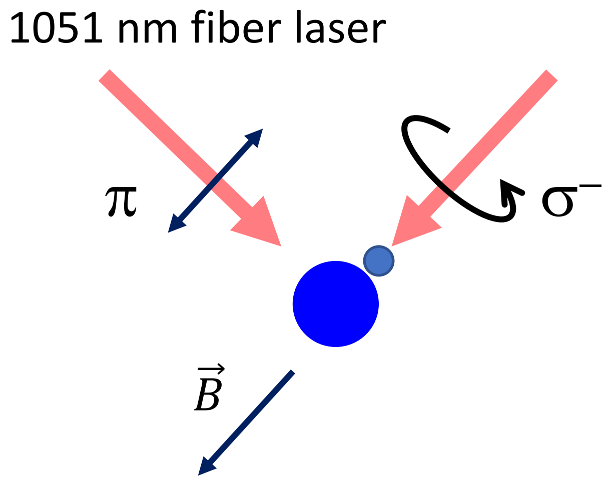
Nondestructive State Detection

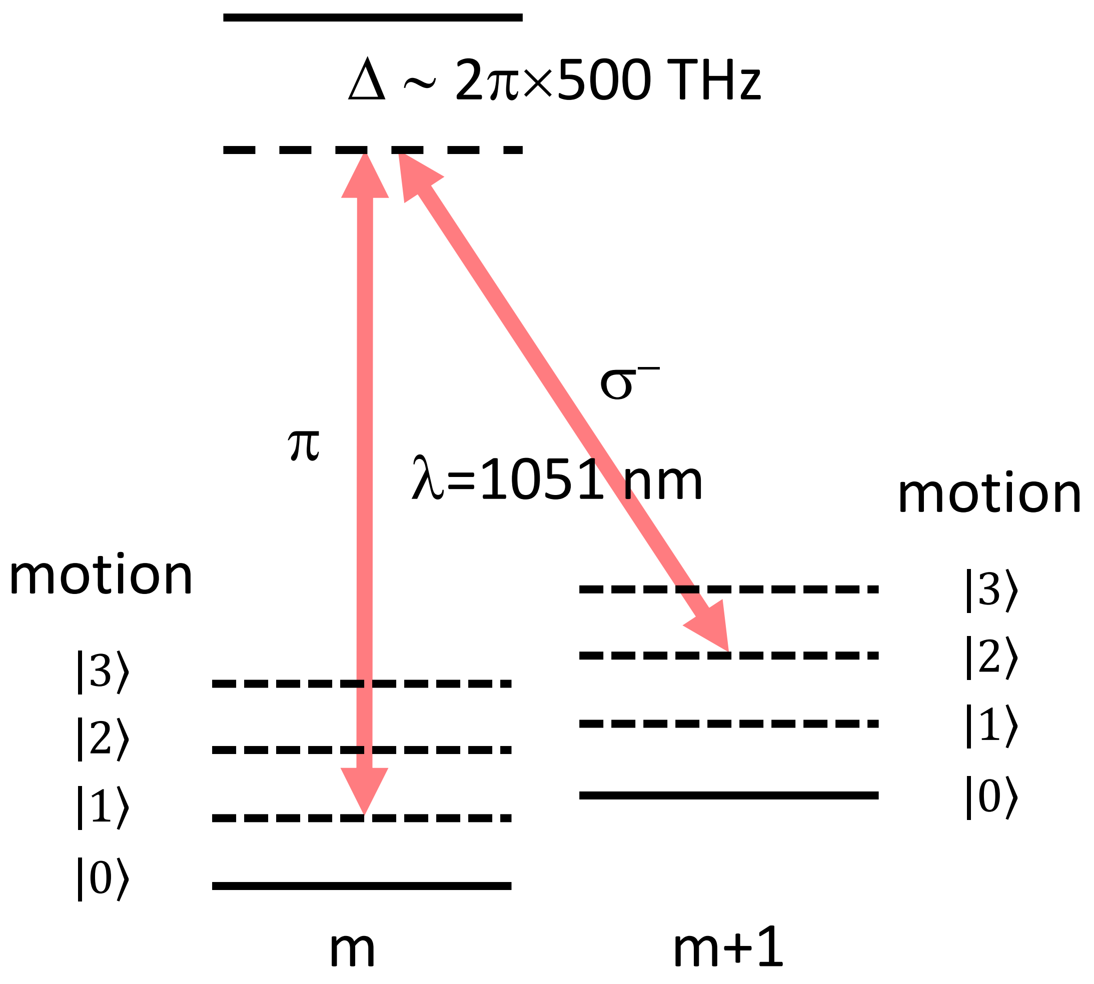


Molecule was at the initial state of the driven transition

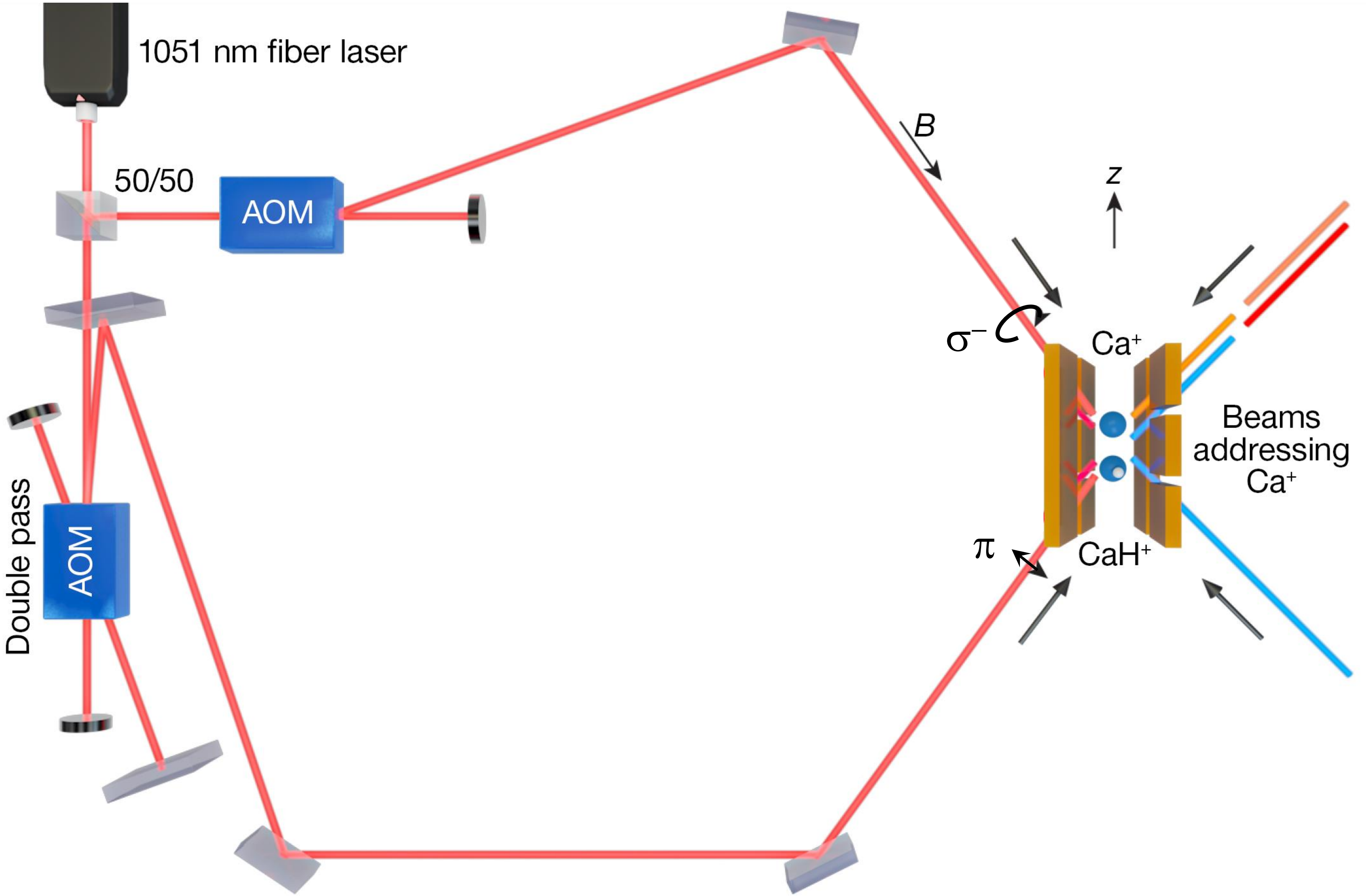
F. Wolf et al., Nature 530, 457 (2016)   M. Sinhal et al., Science 367, 1213 (2020)   Nature **545**, 203 (2017)

Stimulated Raman Transition





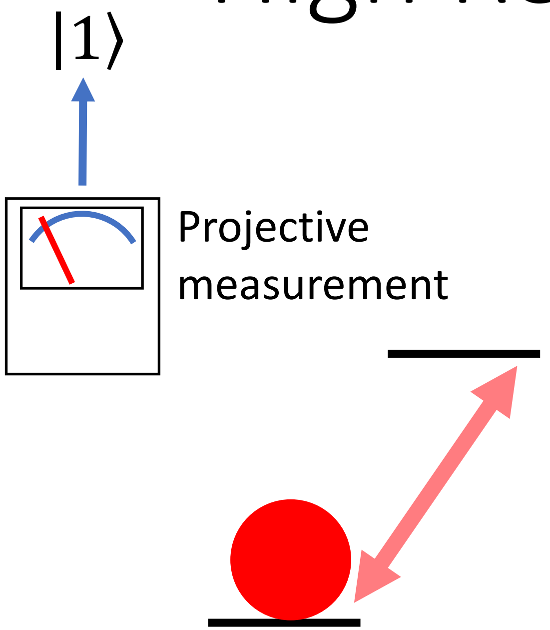
- Coherent process
- Applicable to many molecular ion species

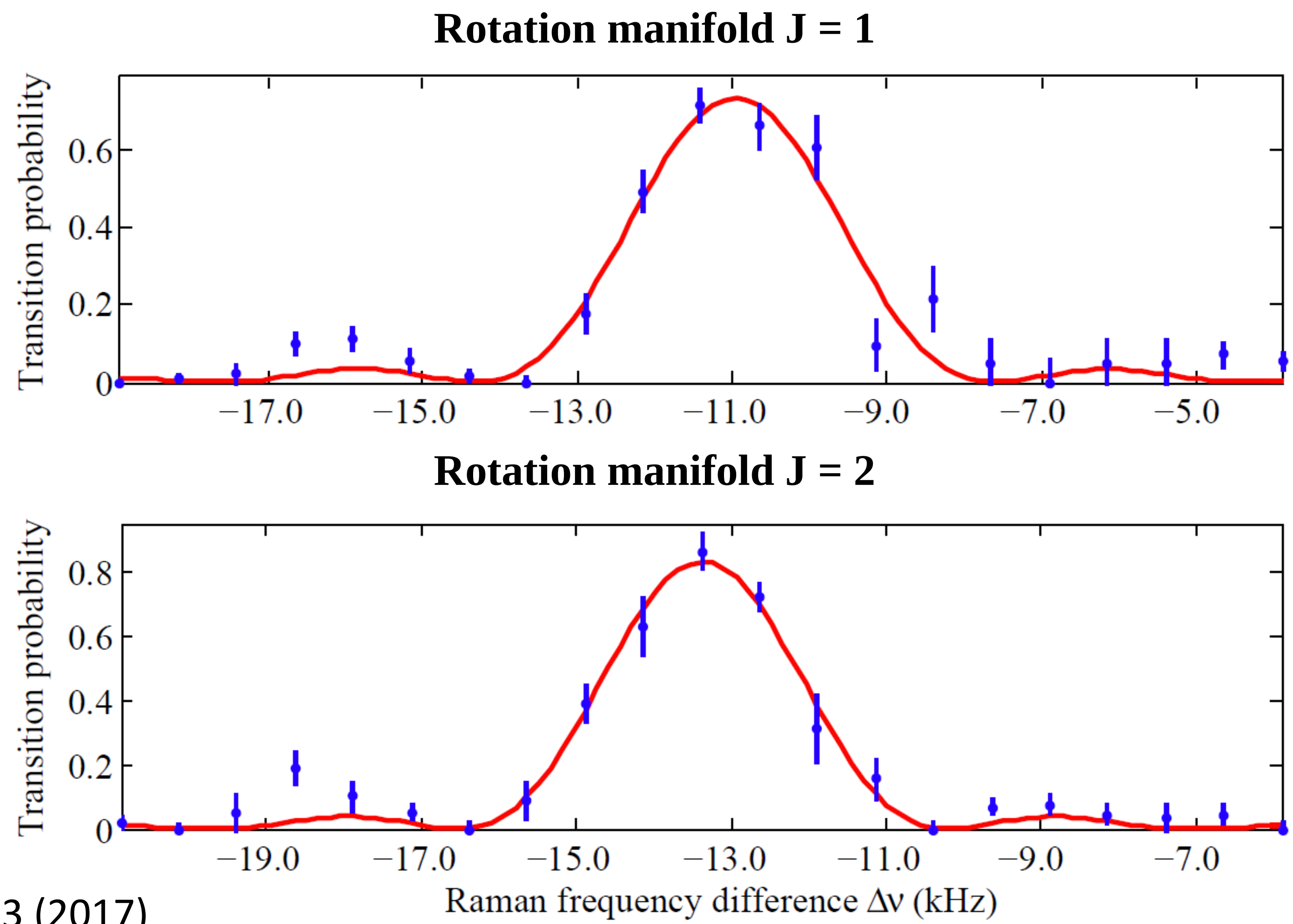


1051 nm fiber laser
50/50
AOM
AOM
Double pass
B
z
σ−
Ca+
CaH+
π
Beams addressing Ca+

Initializing into a Pure State

High Resolution Coherent Spectra

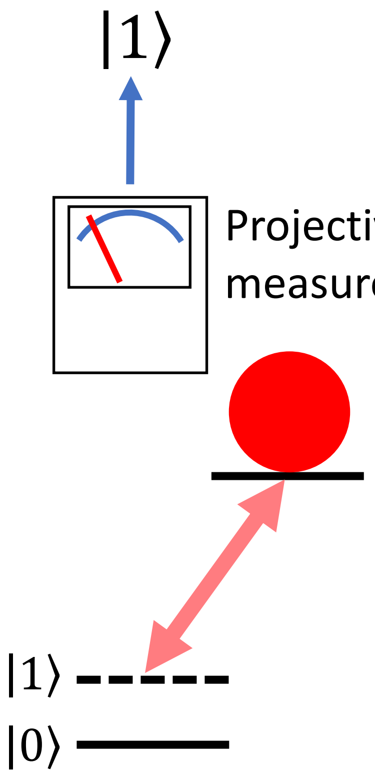


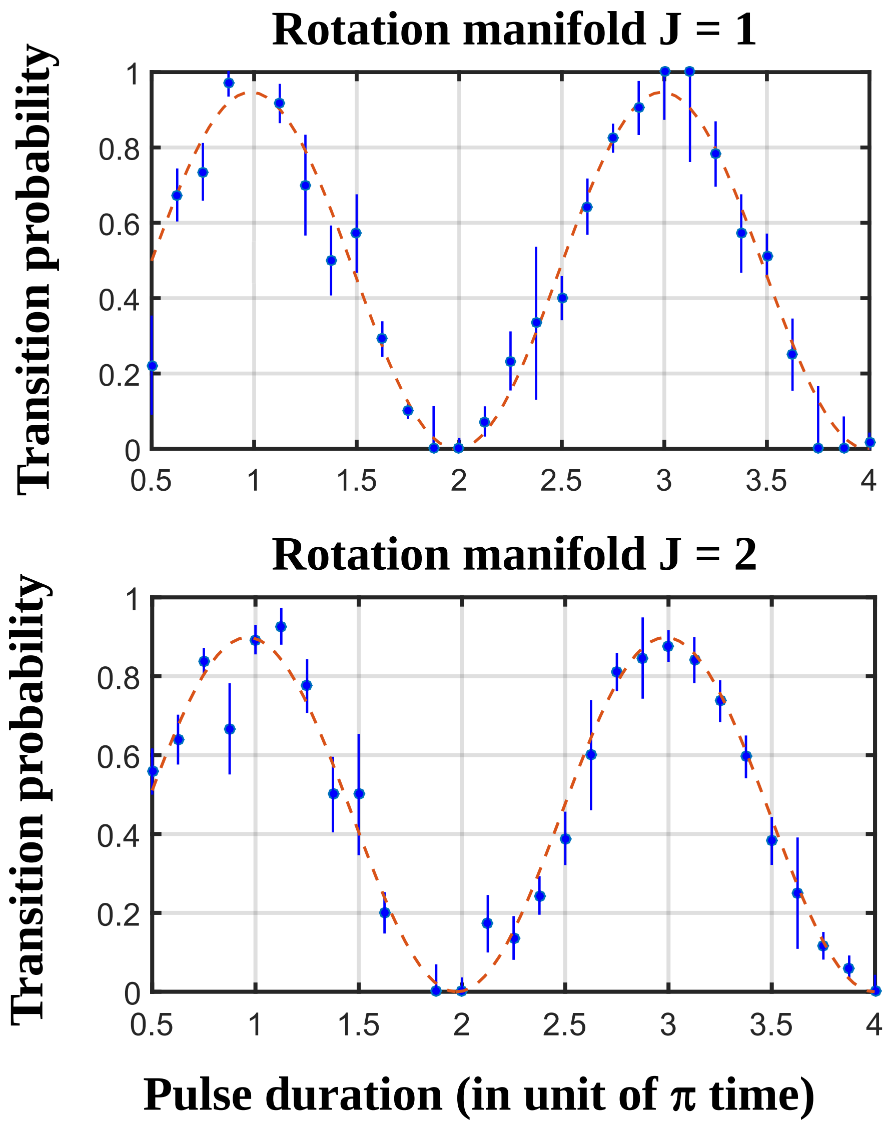


Nature **545**, 203 (2017)

Coherent Manipulation

Rabi Flopping between Molecular Levels

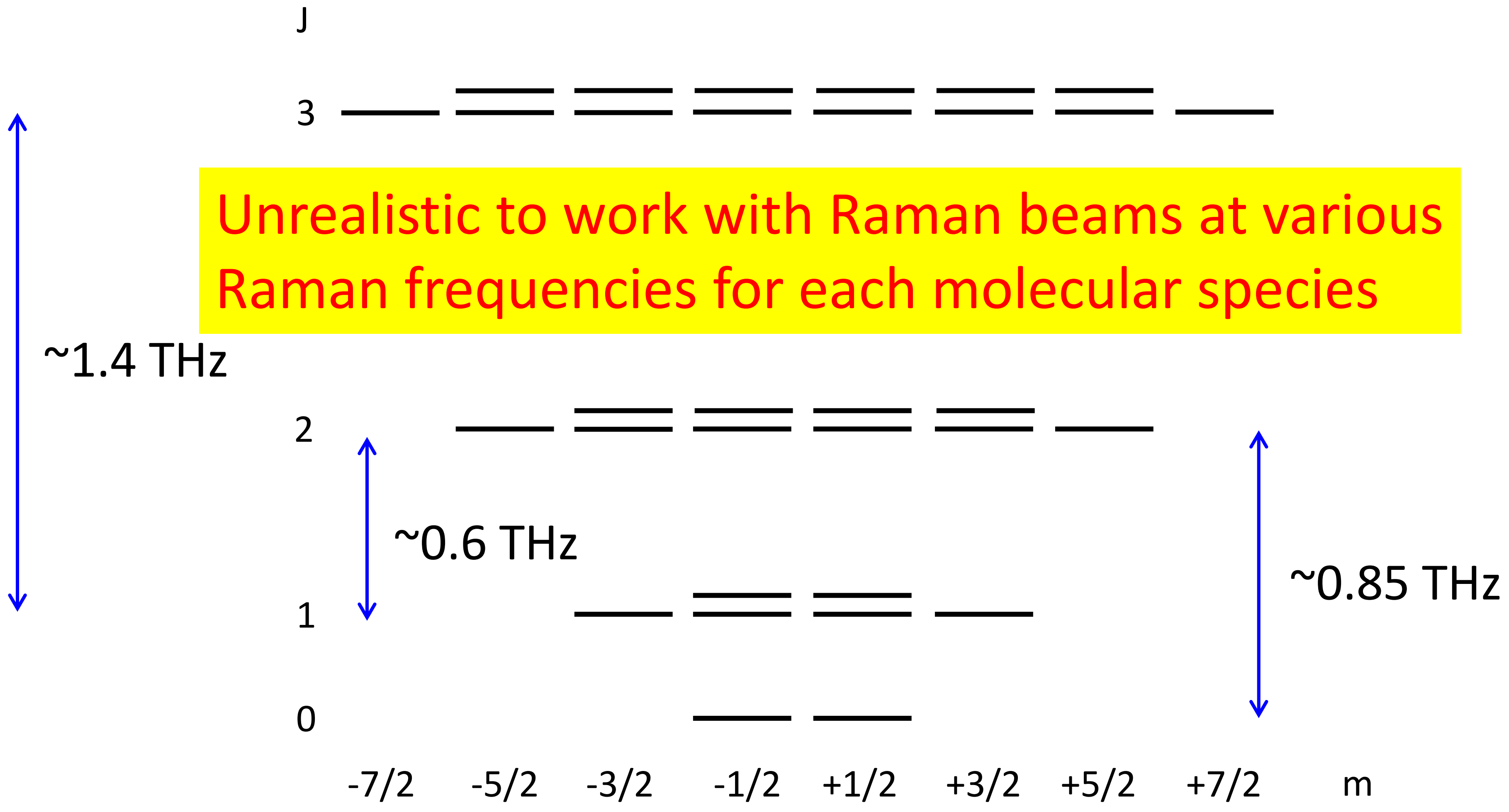




t_π = 700 μs

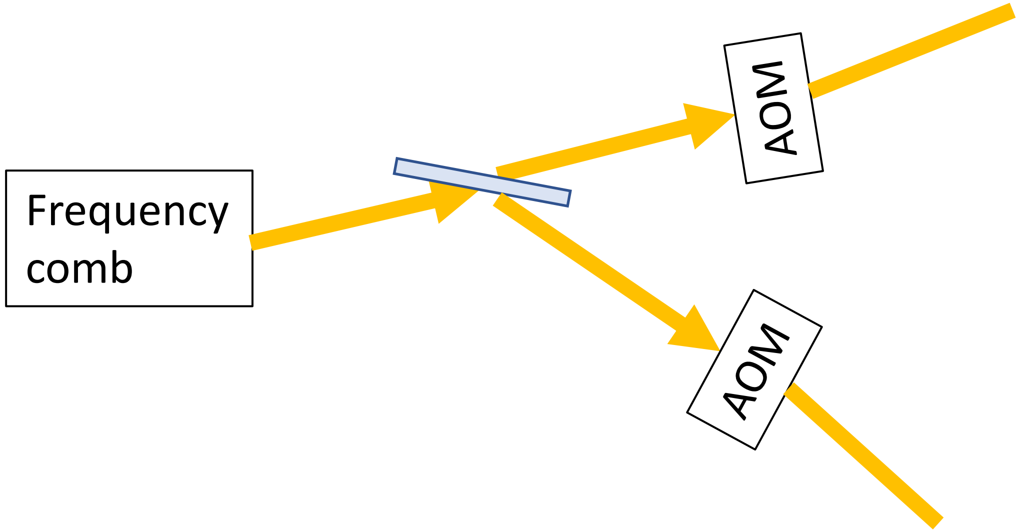
t_π = 788 μs

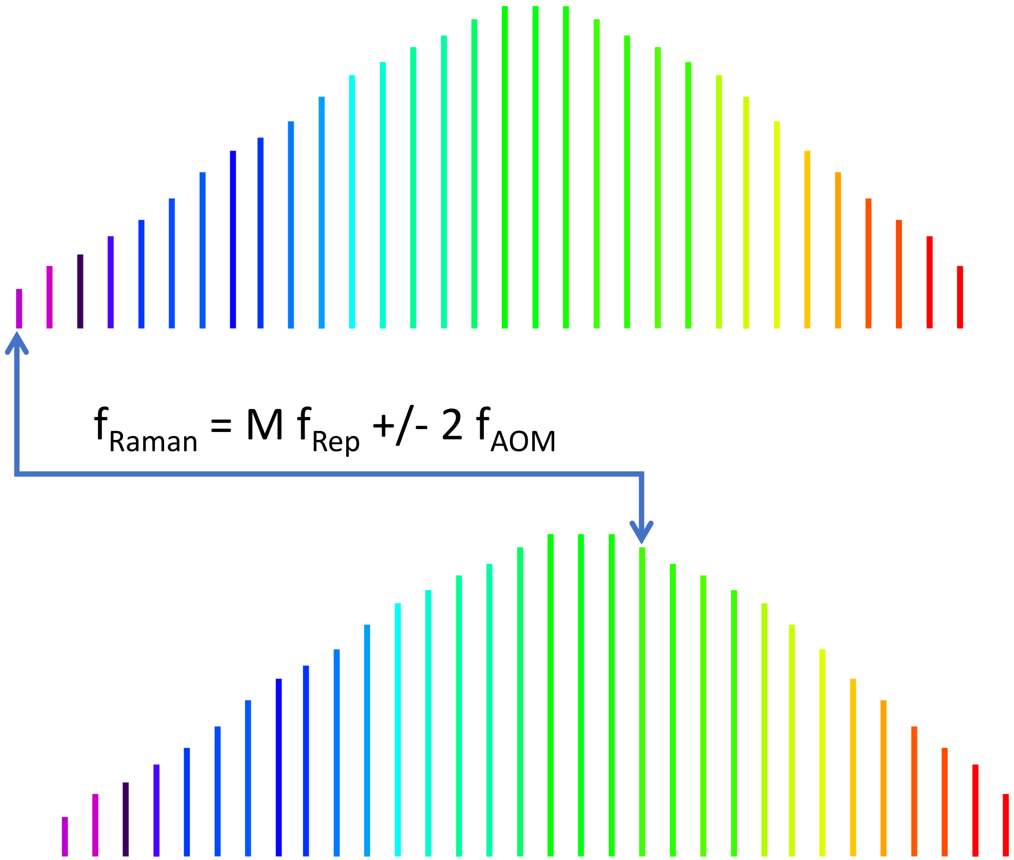
Expanding the Control Toolbox
Controlling Molecular Rotation



Unrealistic to work with Raman beams at various Raman frequencies for each molecular species

Driving THz Rotational Transitions with a Frequency Comb

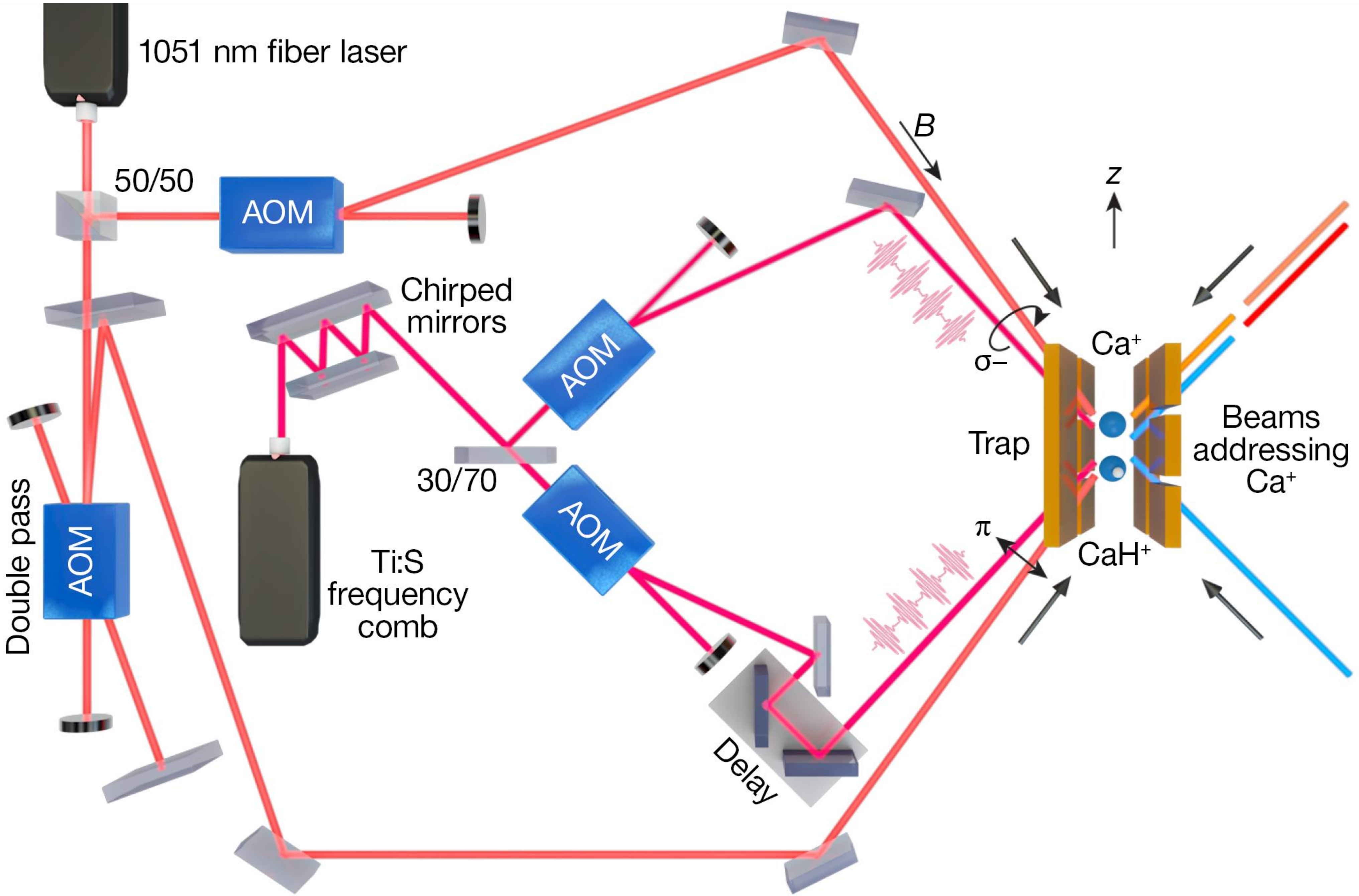




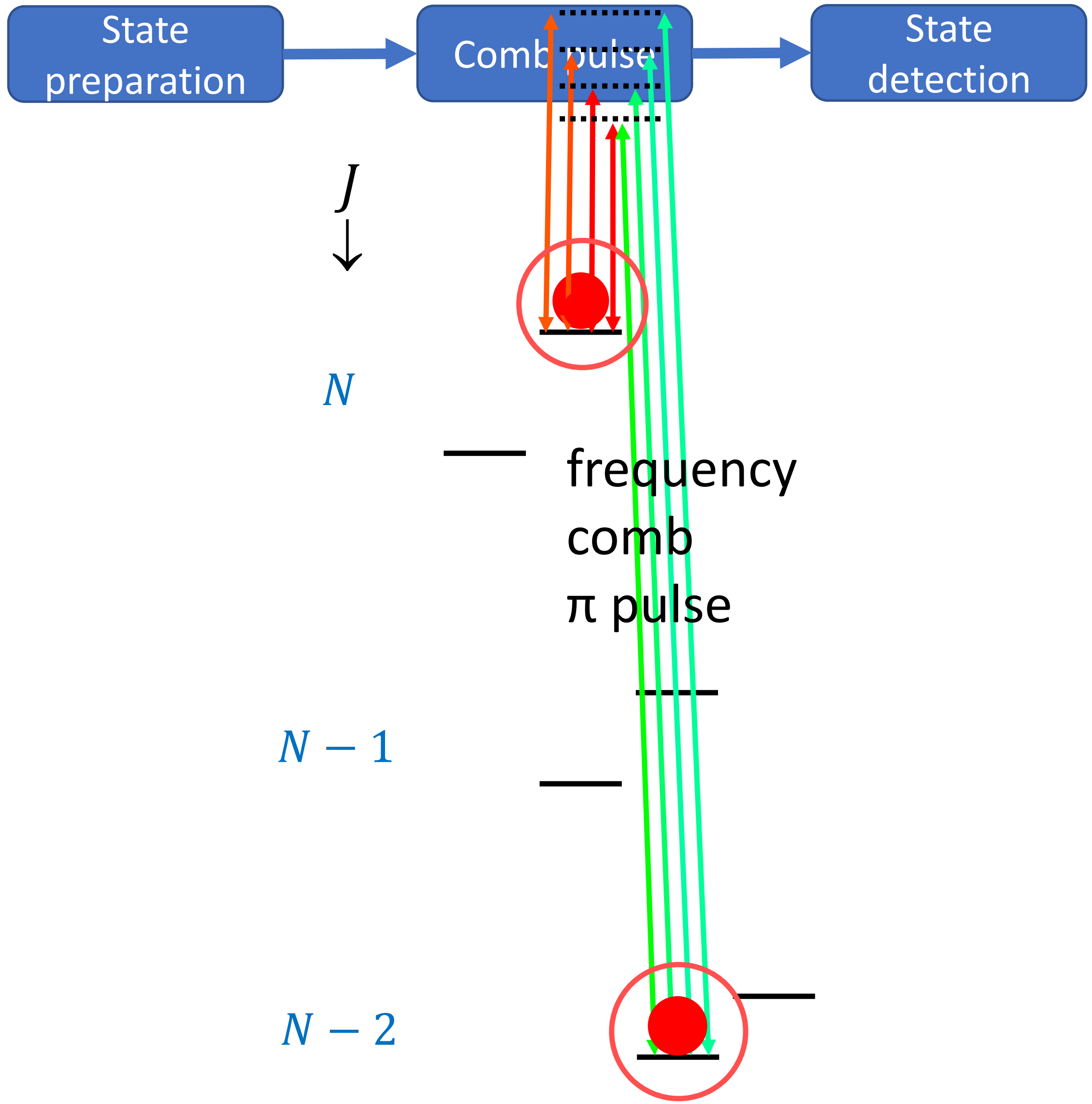
Tara Fortier
Precision Photonic Synthesis, NIST

Scott Diddams
University of Colorado

D. Hayes, et al., Phys. Rev. Lett. 104, 140501 (2010)
S. Ding and D. Matsukevich, New J. Phys. 14 023028 (2012)
D. Leibfried, New J. Phys. 14 023029 (2012)
C. Solaro, et al., Phys. Rev. Lett. 120, 253601 (2018)

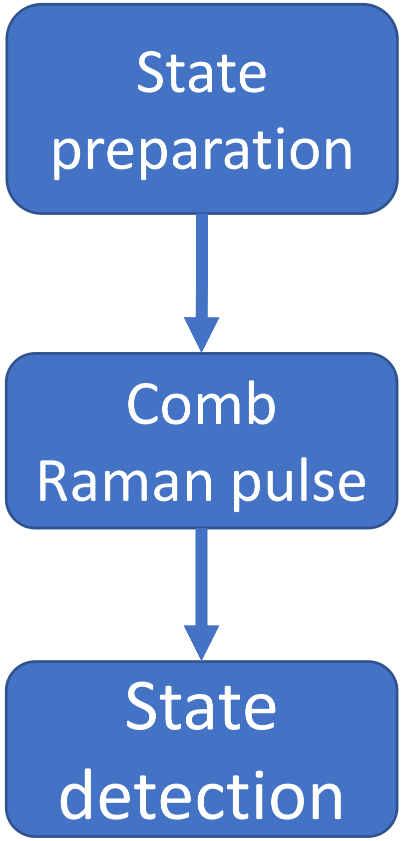

1051 nm fiber laser
50/50
AOM
Chirped mirrors
AOM
B
z
Ca+
σ−
Trap
Beams addressing Ca+
30/70
AOM
Double pass
AOM
Ti:S frequency comb
π
CaH+
Delay

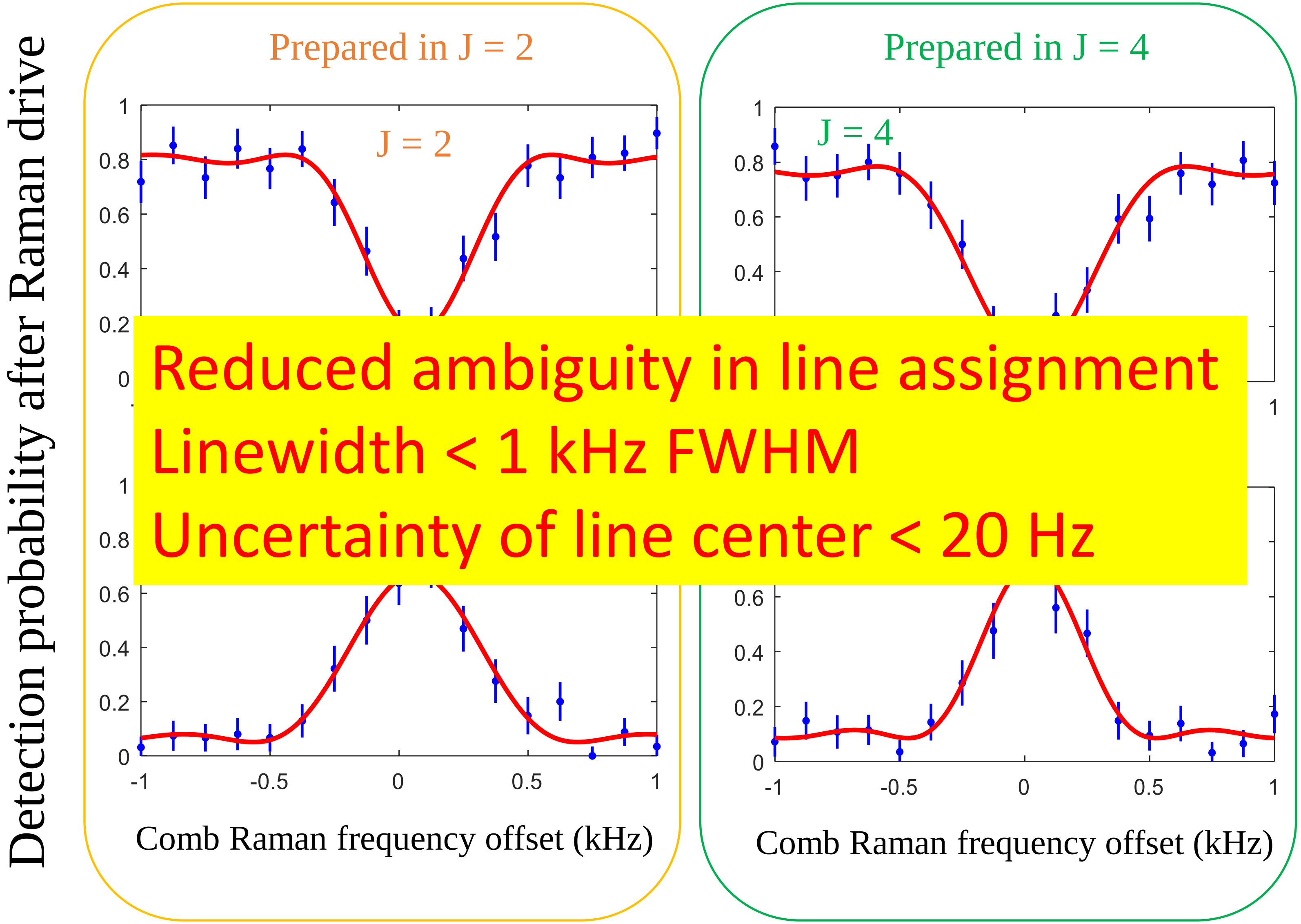
Rotational Spectroscopy



Complementary Rotational Spectra

Science 367, 1458 (2020)





Reduced ambiguity in line assignment

Linewidth < 1 kHz FWHM

Uncertainty of line center < 20 Hz

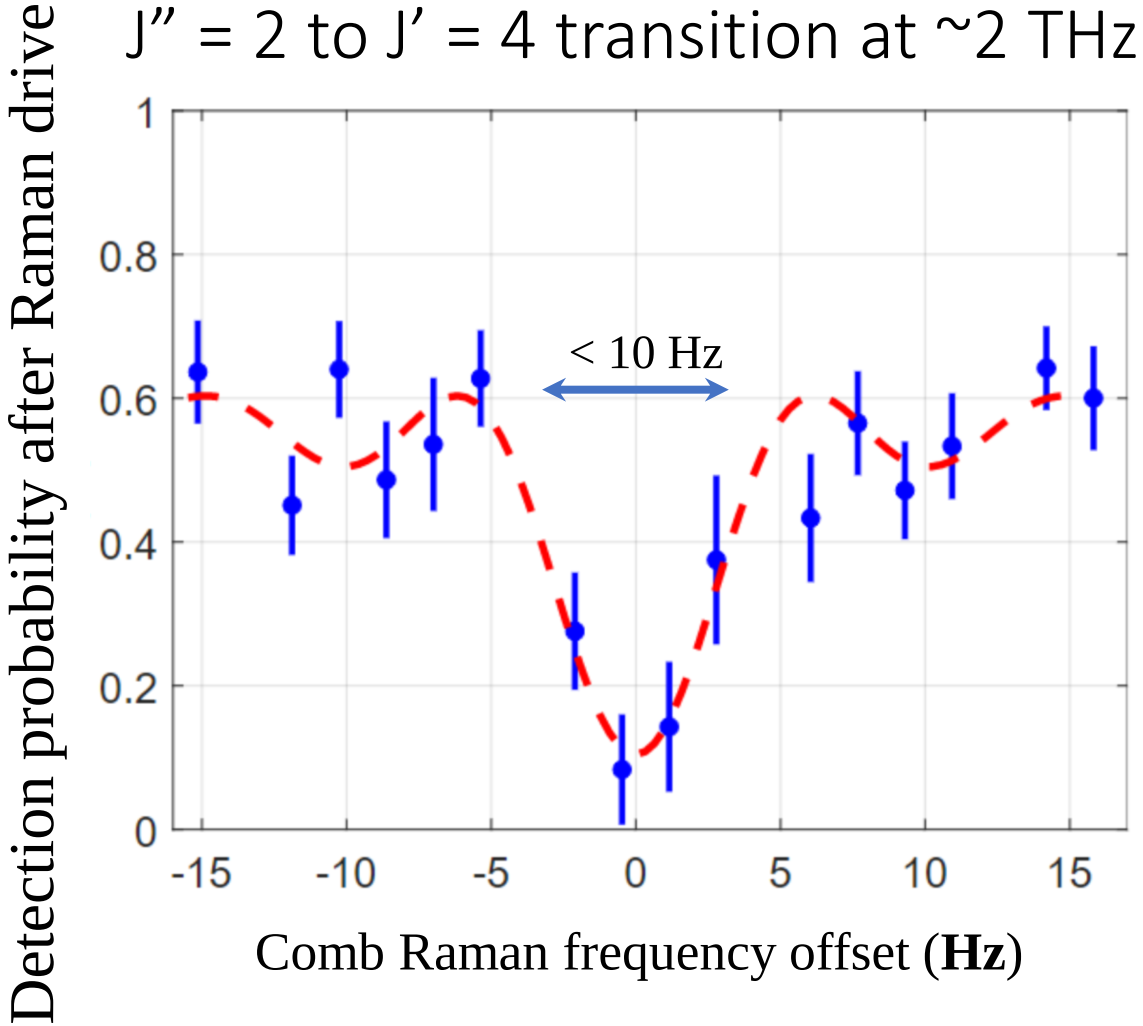
Improved Precision

- Improved comb coherence:

microwave reference

$\Rightarrow$ Al^+ clock laser

- 128 ms probe time shown
- Sub-Hz/sub-ppt statistical line-center uncertainty



A.L. Collopy et al., Phys. Rev. Lett. **130**, 223201 (2023)

Article

Precise test of quantum electrodynamics and determination of fundamental constants with HD+ ions

https://doi.org/10.1038/s41586-020-2261-5

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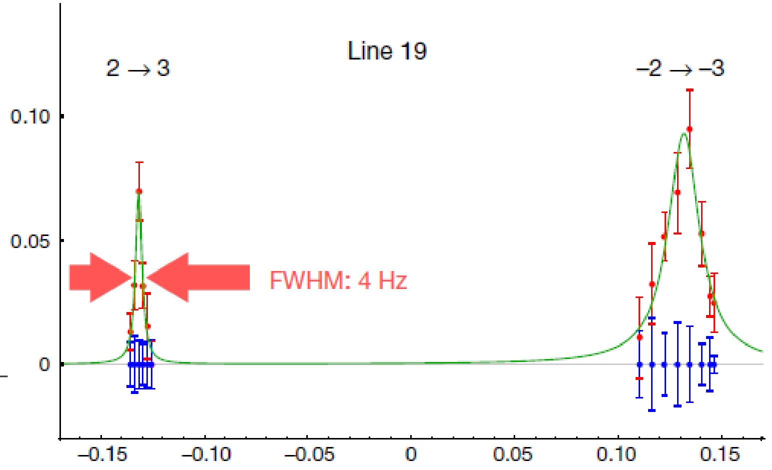
Published online: 06 May 2020

S. Alighanbari[1], G. S. Giri[1], F. L. Constantin[1,2], V. I. Korobov[3] & S. Schiller[1]✉

Bound three-body quantum systems are important for fundamental physics[1,2] because they enable tests of quantum electrodynamics theory and provide access to



Science

REPORTS

Cite as: S. Patra *et al.*, *Science* 10.1126/science.aba0453 (2020).

Proton-electron mass ratio from laser spectroscopy of HD+ at the part-per-trillion level

Sayan Patra[1], M. Germann[1*], J.-Ph. Karr[2,3], M. Haidar[2], L. Hilico[2,3], V. I. Korobov[4], F. M. J. Cozijn[1], K. S. E. Eikema[1,5], W. Ubachs[1,5], J. C. J. Koelemeij[1†]

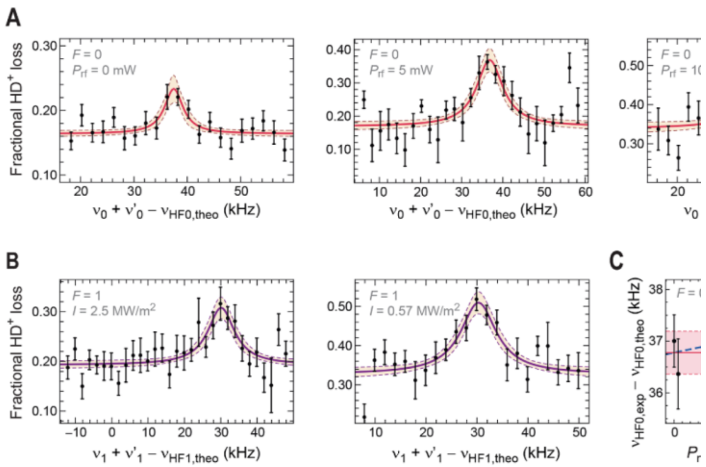


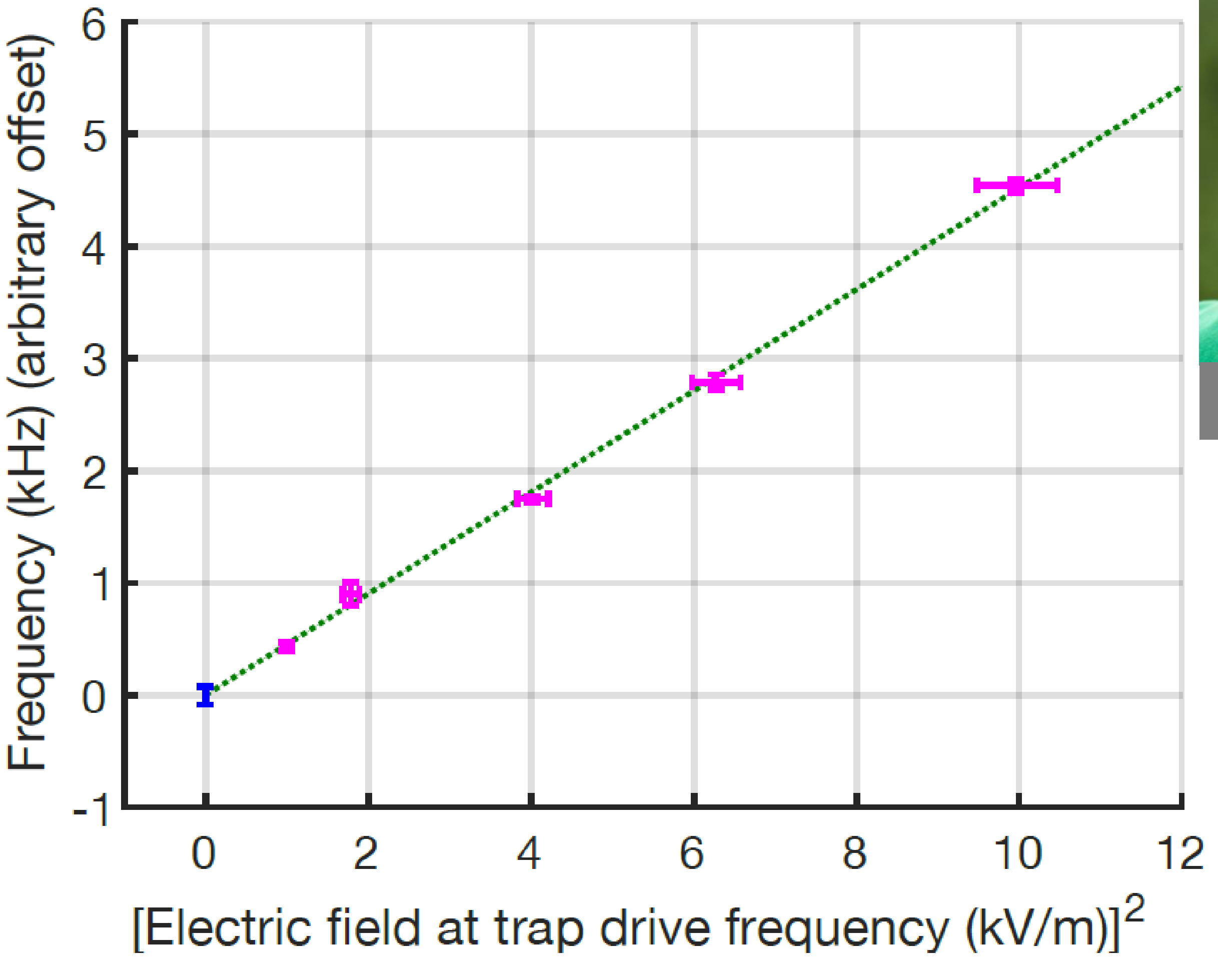
Fig. 2. Spectra of the two-photon transition at 415 THz.

Measuring the dipole moment of CaH^+

- RF-field dependence of J"=0 to J'=2 transition
- J'=2 sublevel nearly unaffected
- J"=0 shift:

$$\Delta f_{J=0} = -\frac{d^2 |E_{rf}|^2}{12\ h^2 B_{\mathrm{rot}}}$$

- CaH^+ electric dipole moment *d*:
  5.5 Debye (5.35-5.75)

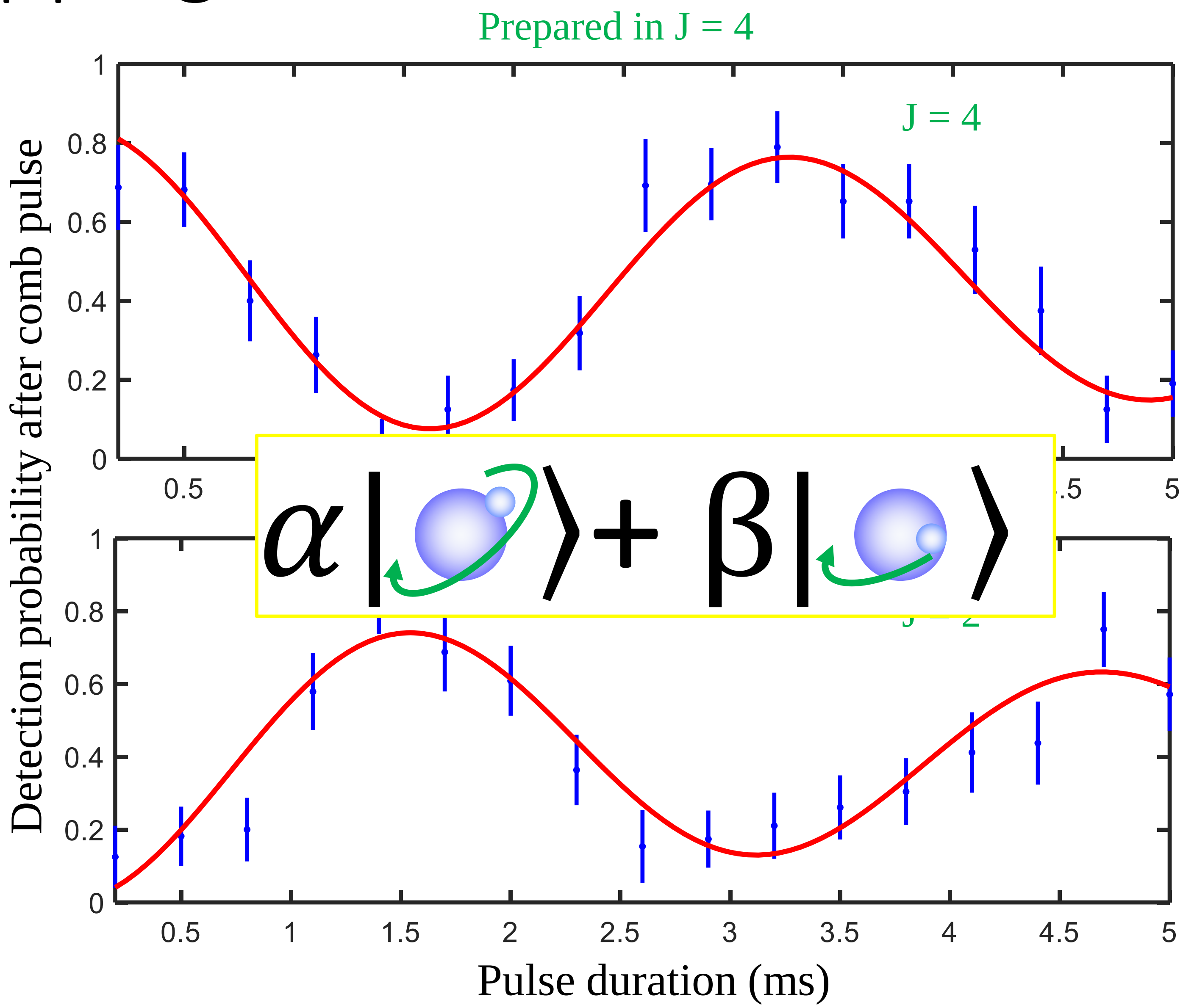



Alejandra Collopy

A.L. Collopy et al., Phys. Rev. Lett. **130**, 223201 (2023)

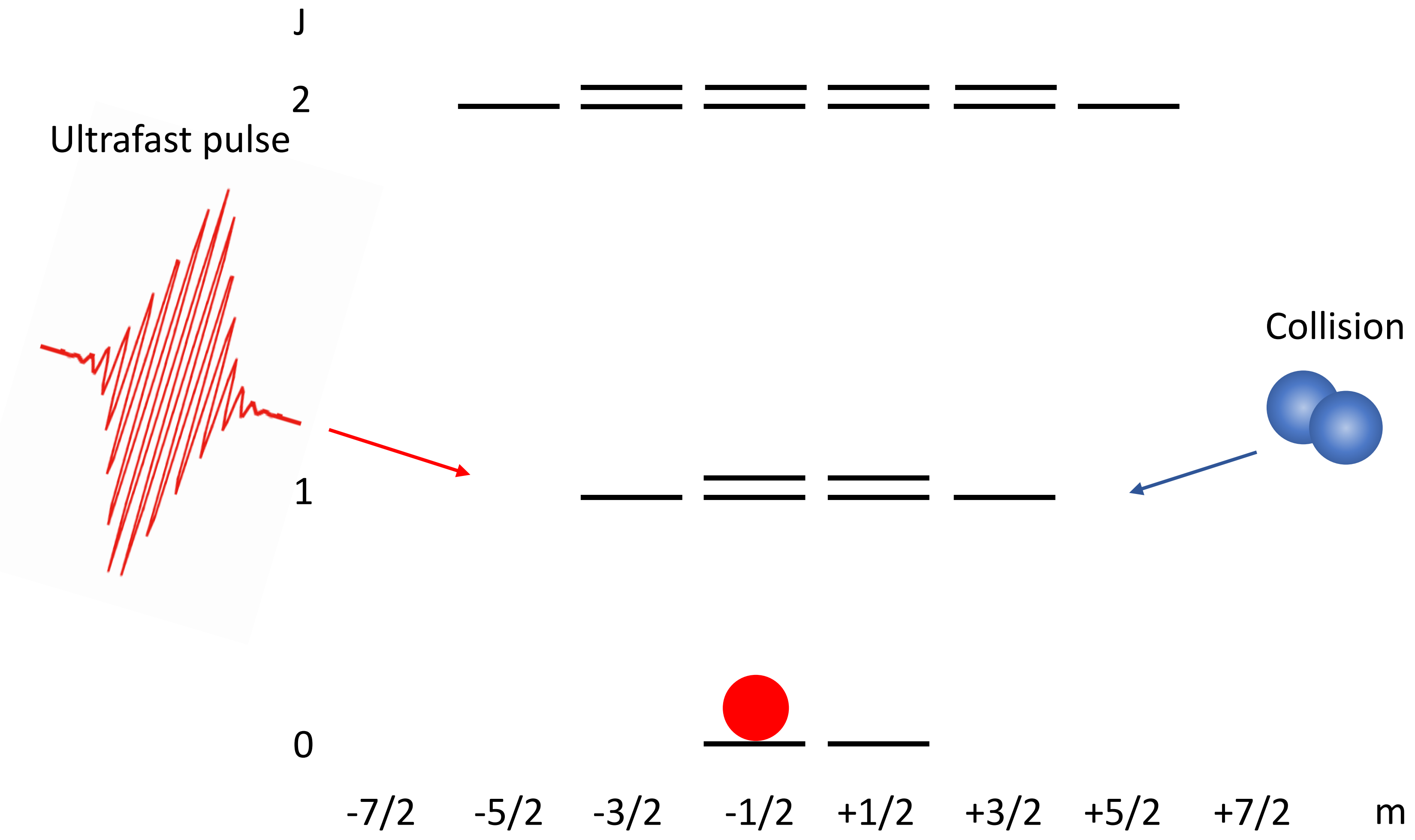
Coherent Manipulation

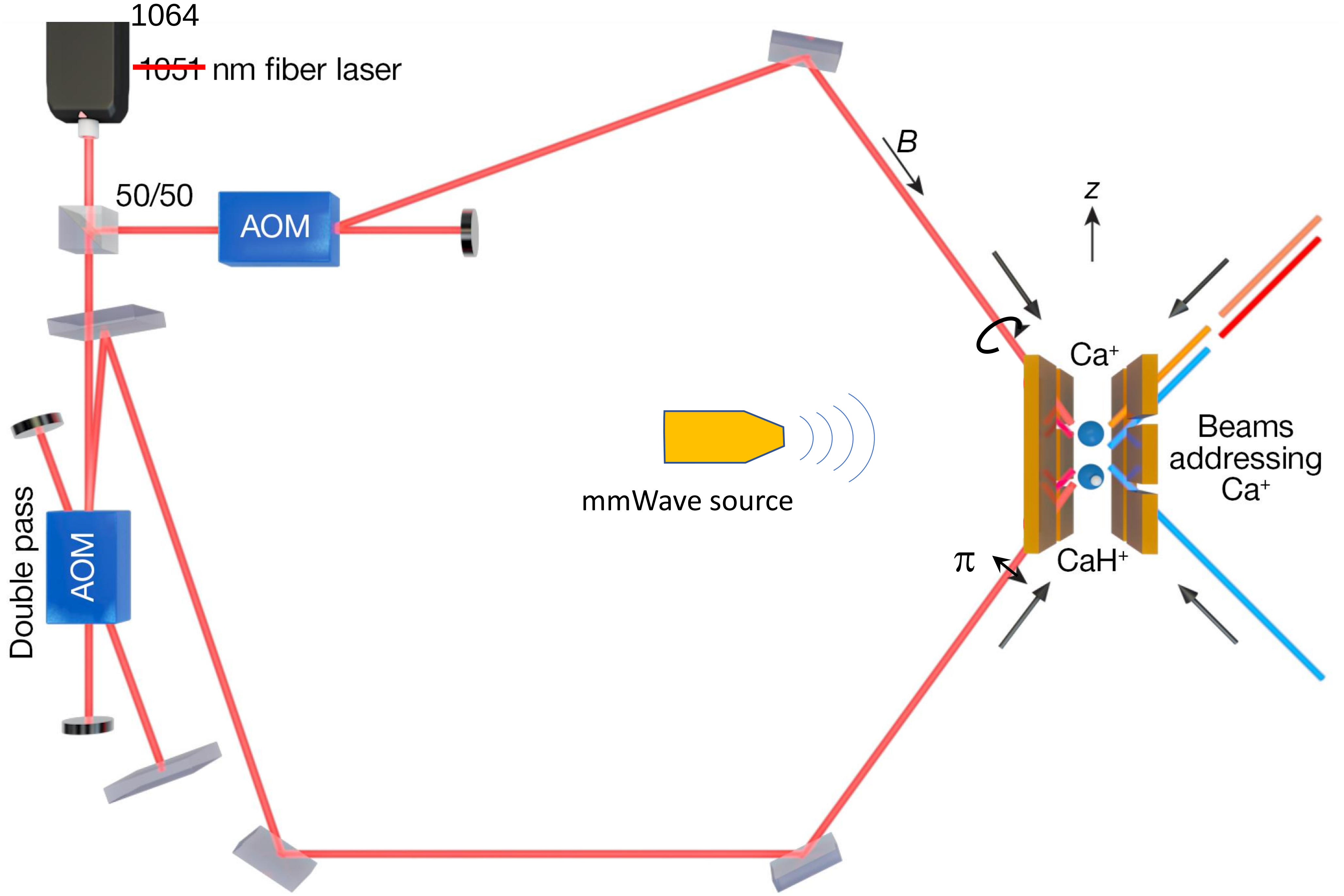
Rabi flopping between J = 2 and J = 4 levels



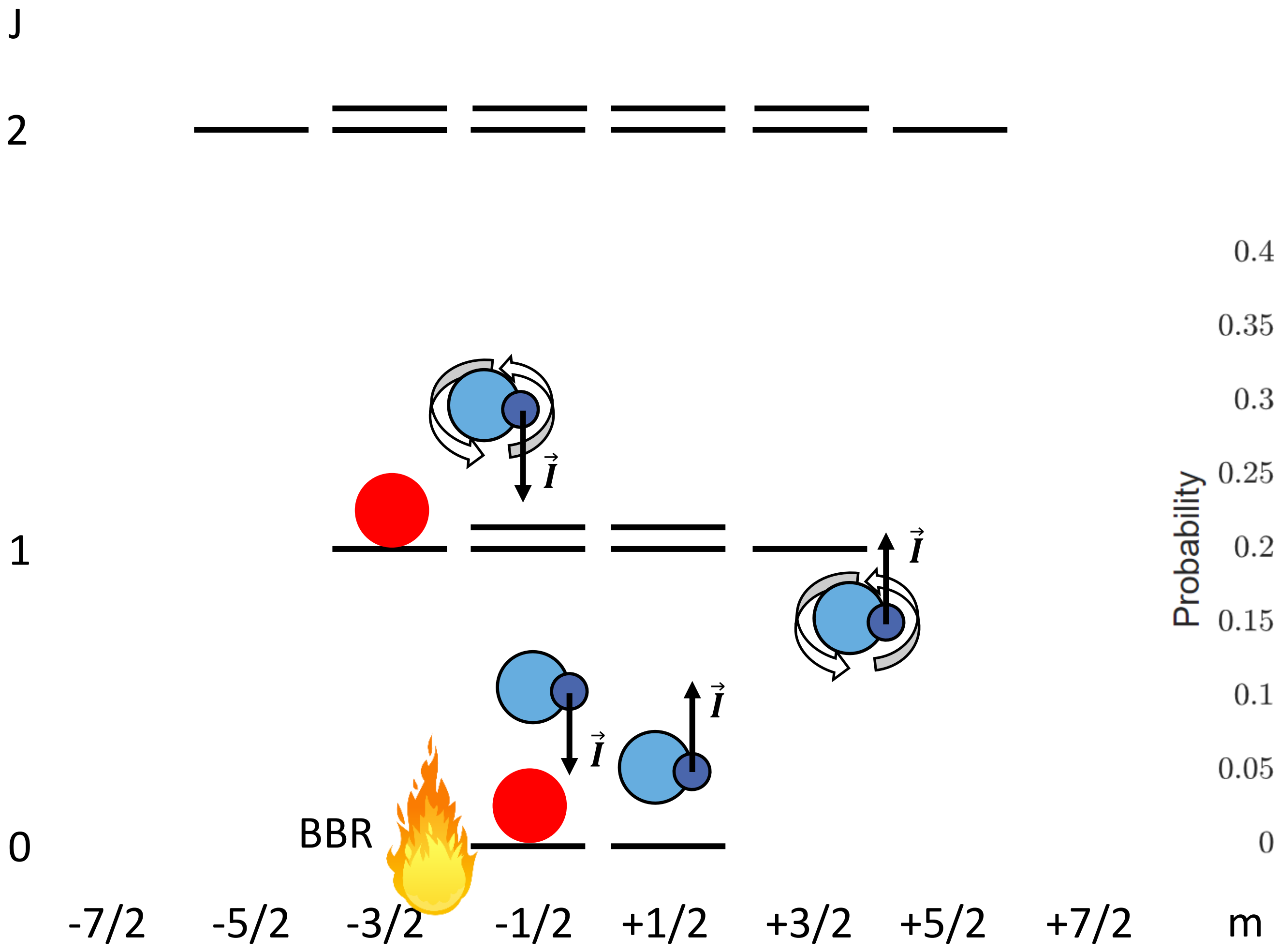
Science 367, 1458 (2020)

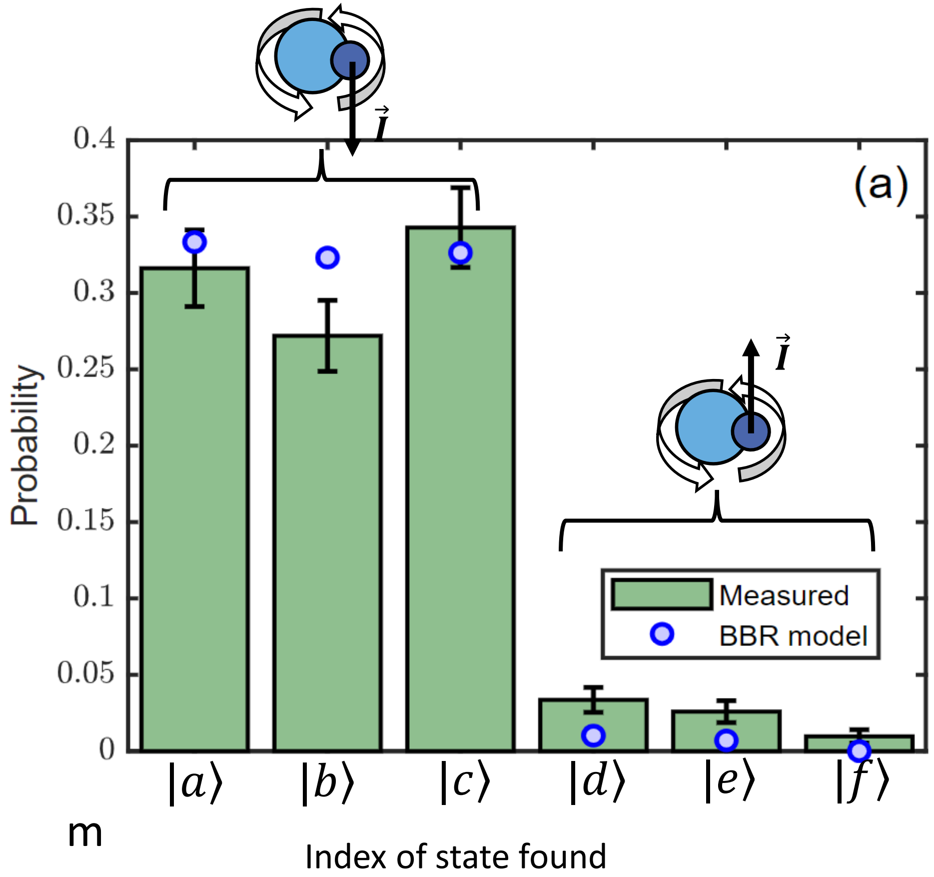
Tracking Molecular State




1064
1051 nm fiber laser
50/50
AOM
Double pass
AOM
B
z
Ca+
CaH+
π
mmWave source
Beams addressing Ca+

Tracking Molecular State

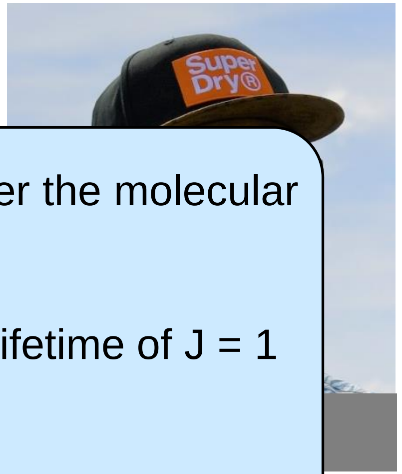


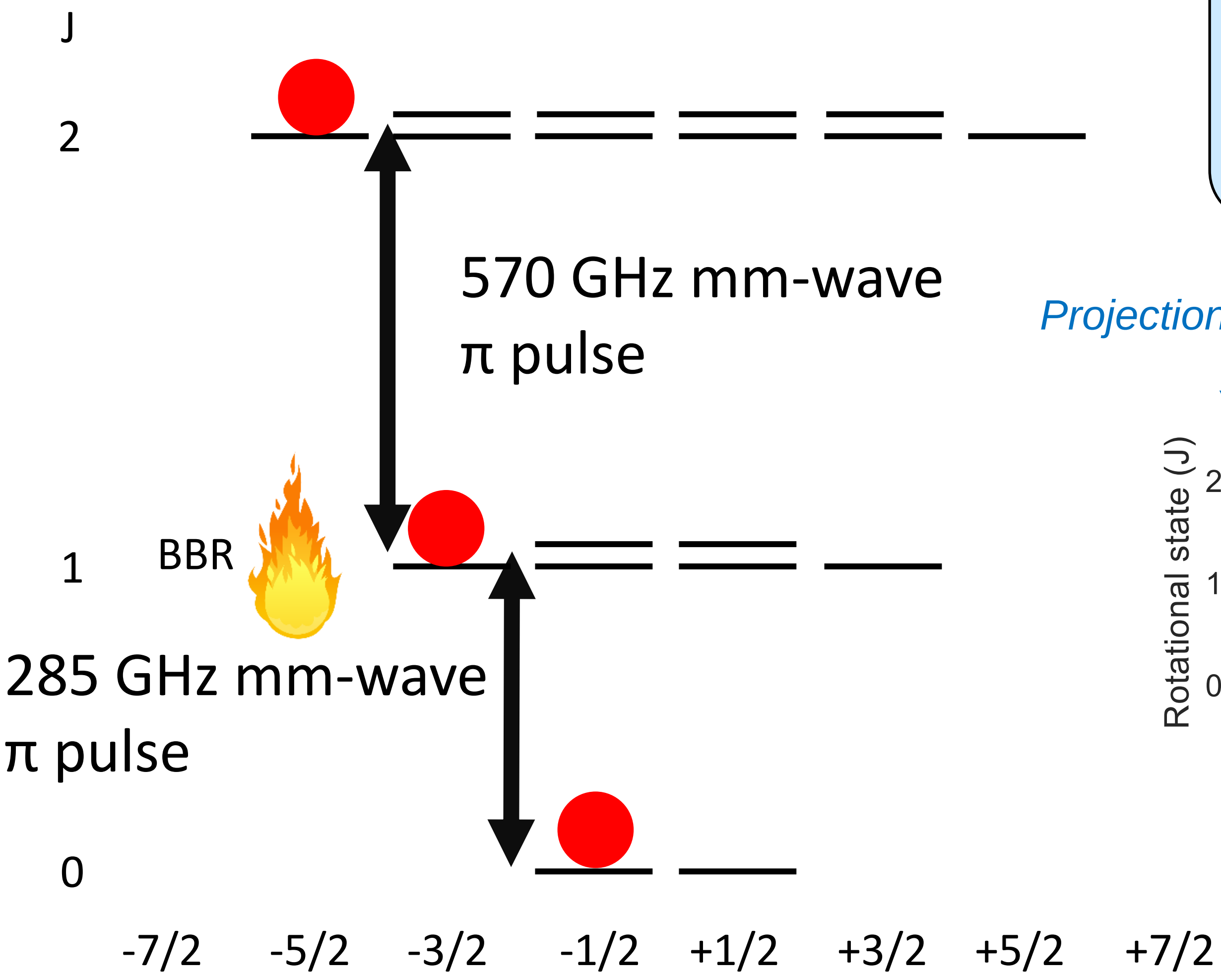


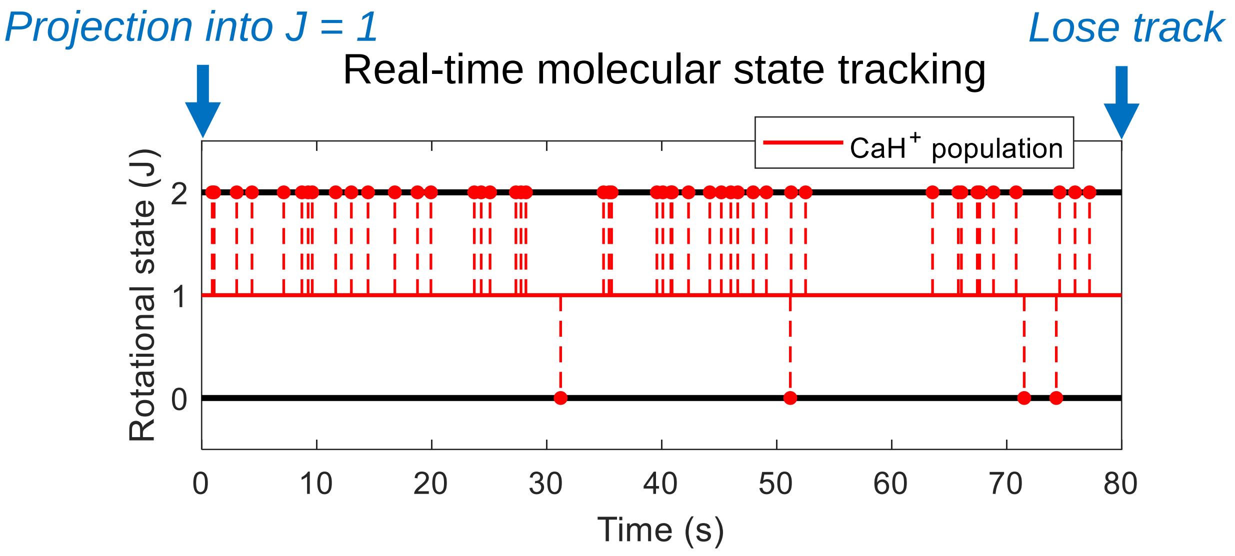
Yu Liu

Trapping Molecular State

- For each projection can recover the molecular state avg. ~ **20 times**
- Increased the effective mean lifetime of J = 1 from 1.7 s to **35 s**
- Increased the duty cycle of J = 1 experiments from 5% to **65%**







Previous work in Penning traps:

D. J. Fink and E. G. Myers, PRL **124**, 013001 (2020)

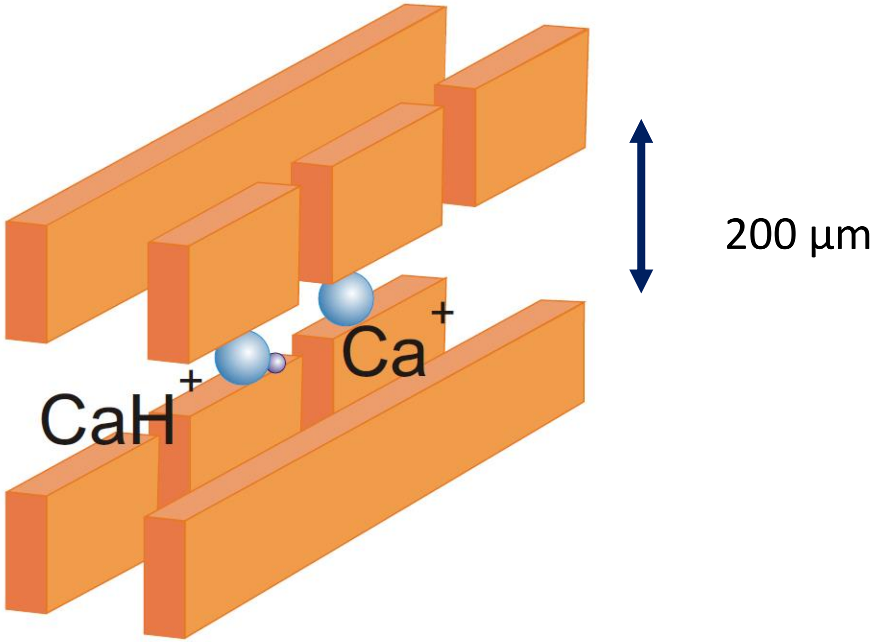
J. K. Thompson, et al., Nature **430**, 58 (2004)

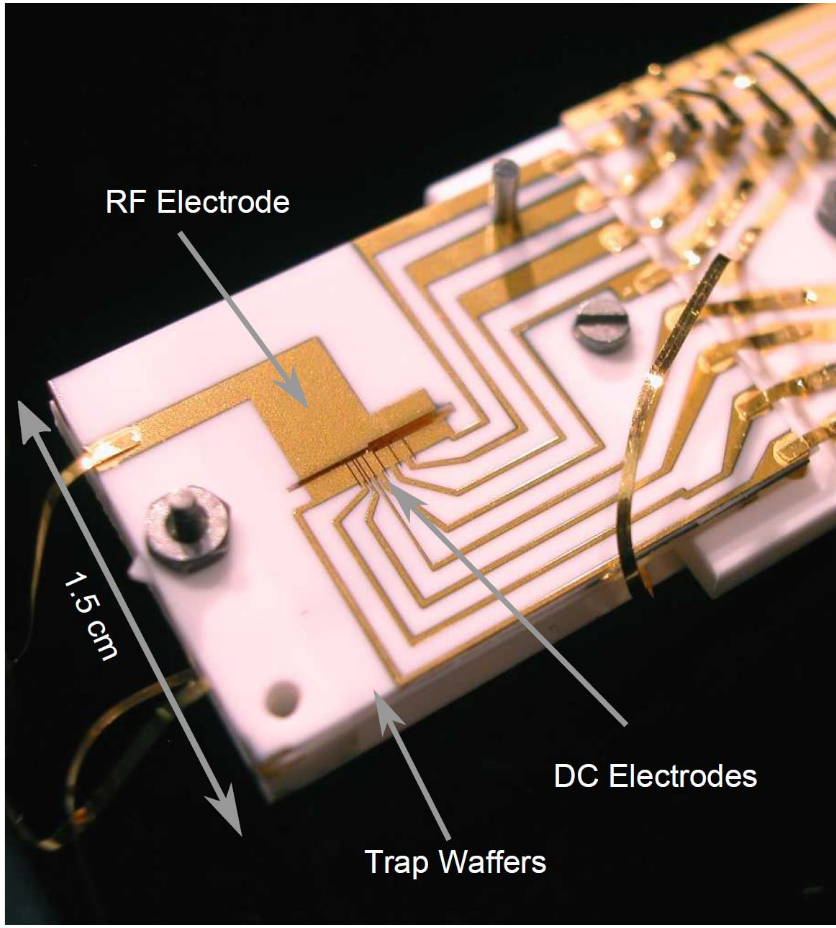
Inferring Temperature from Transition Rates

Transition rates imply different BBR temperatures and are not consistent with room temperature environment.

Measuring lifetimes of specific sublevels can infer strengths of different polarizations in the background thermal radiation.

The significantly different intensities of the polarizations might be the result of sub-mm dimensions of the trap structure.



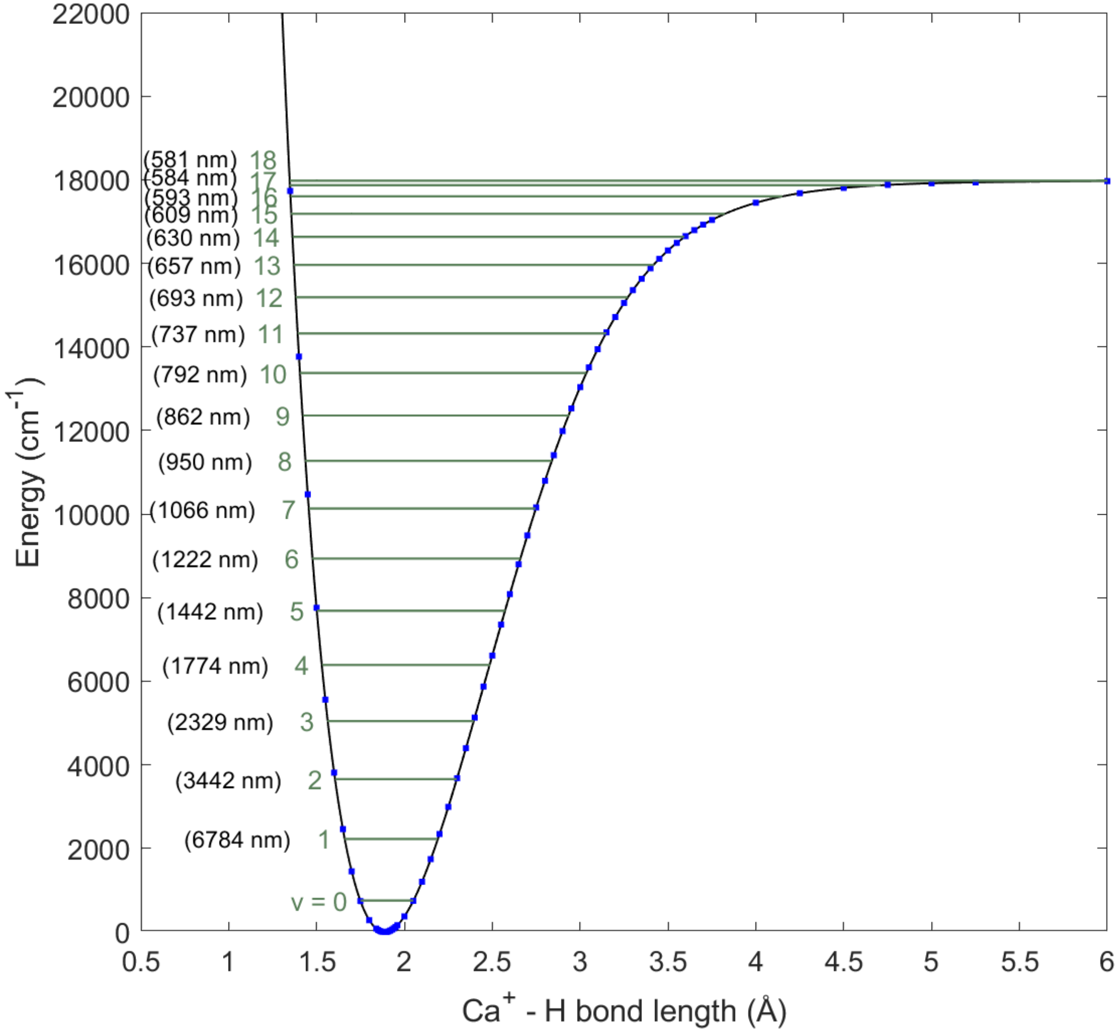


Credit: John Jost thesis

Vibrational Spectroscopy

- Telecom transitions
- Uncertainties on the order of THz from theoretical calculations

Vibrational levels of CaH^+



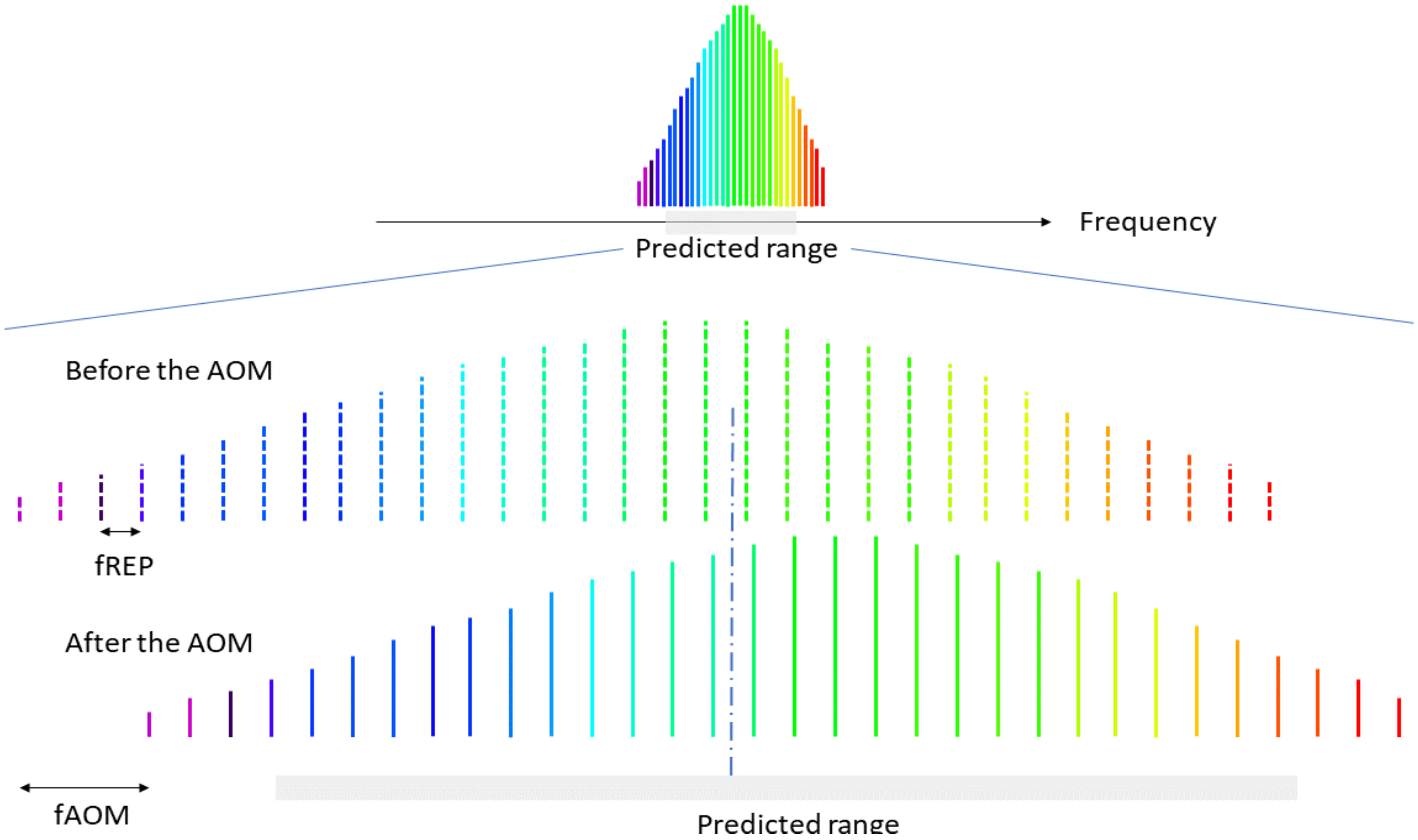
Direct-comb vibrational spectroscopy

Searching for a “needle in the haystack”

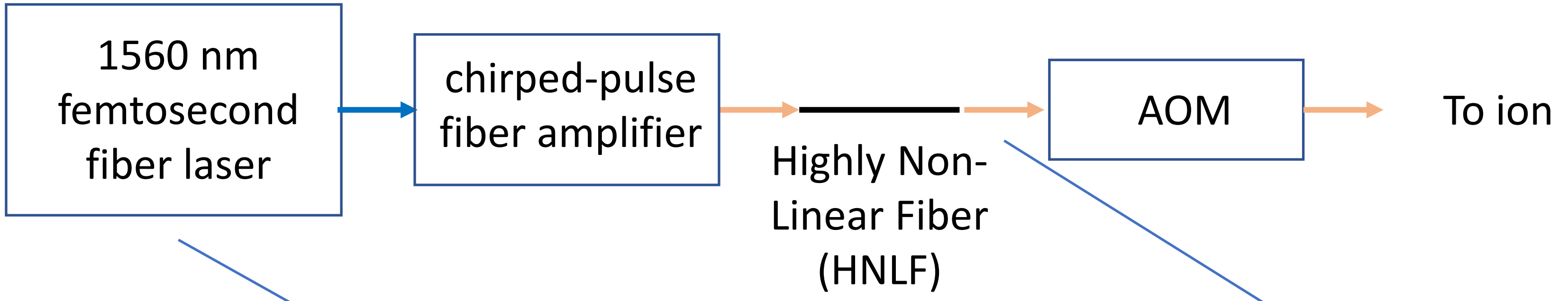
A frequency comb with 200 MHz repetition rate fREP and ~1 THz spectral width can simplify the search by 1 THz/200 MHz = 5×10^3

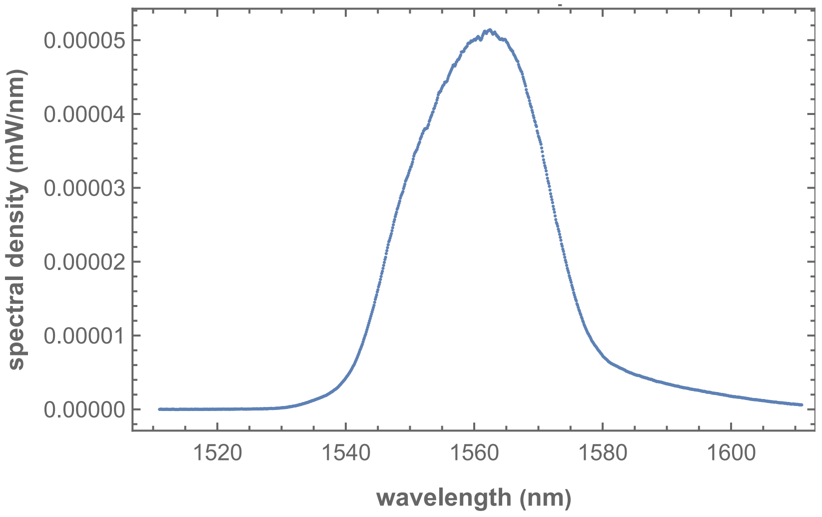
The frequency of each comb tooth is known to high precision.

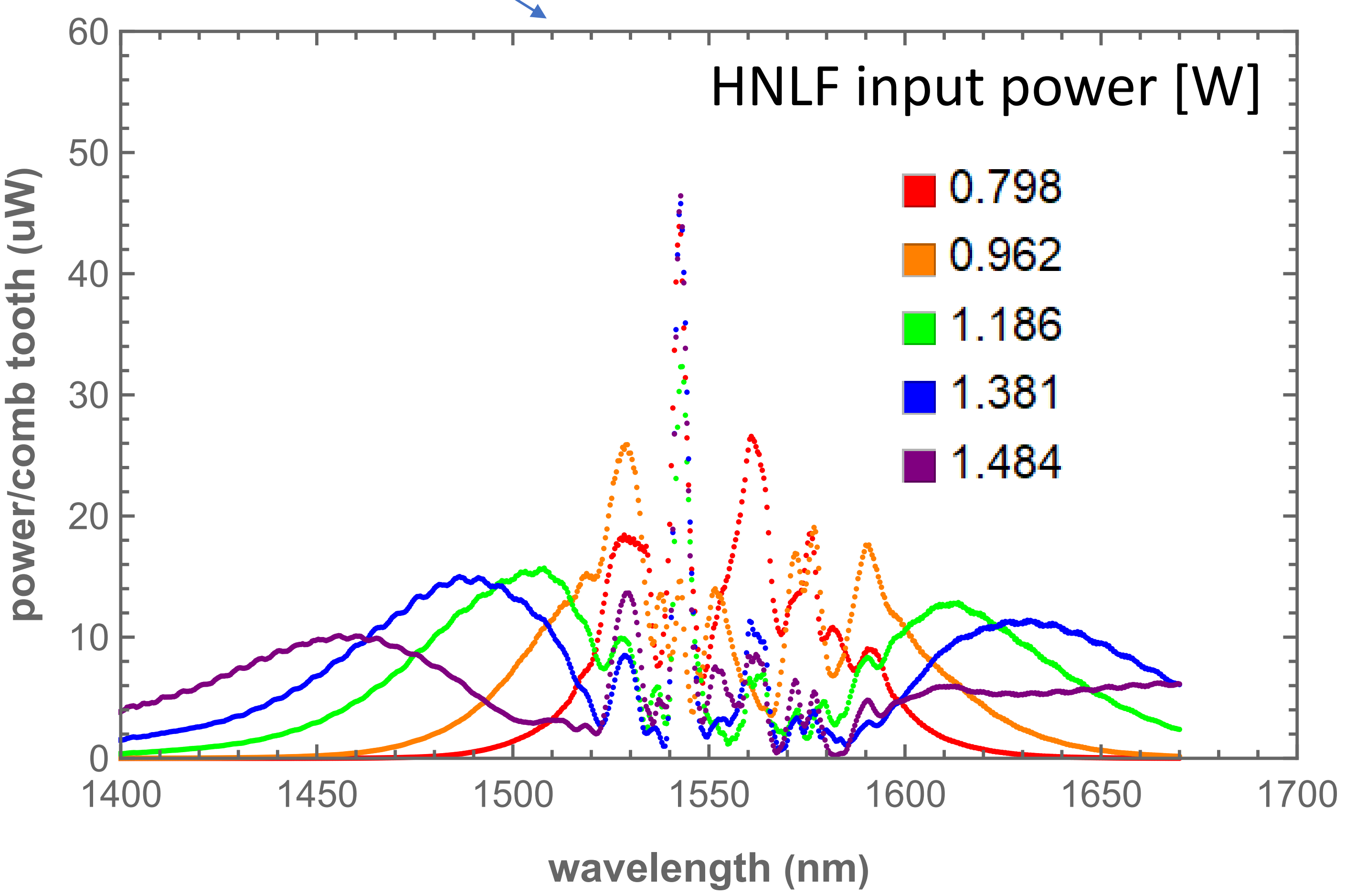
Performing spectroscopy with the molecule in a pure initial state yields few spectroscopic features easier to interpret.

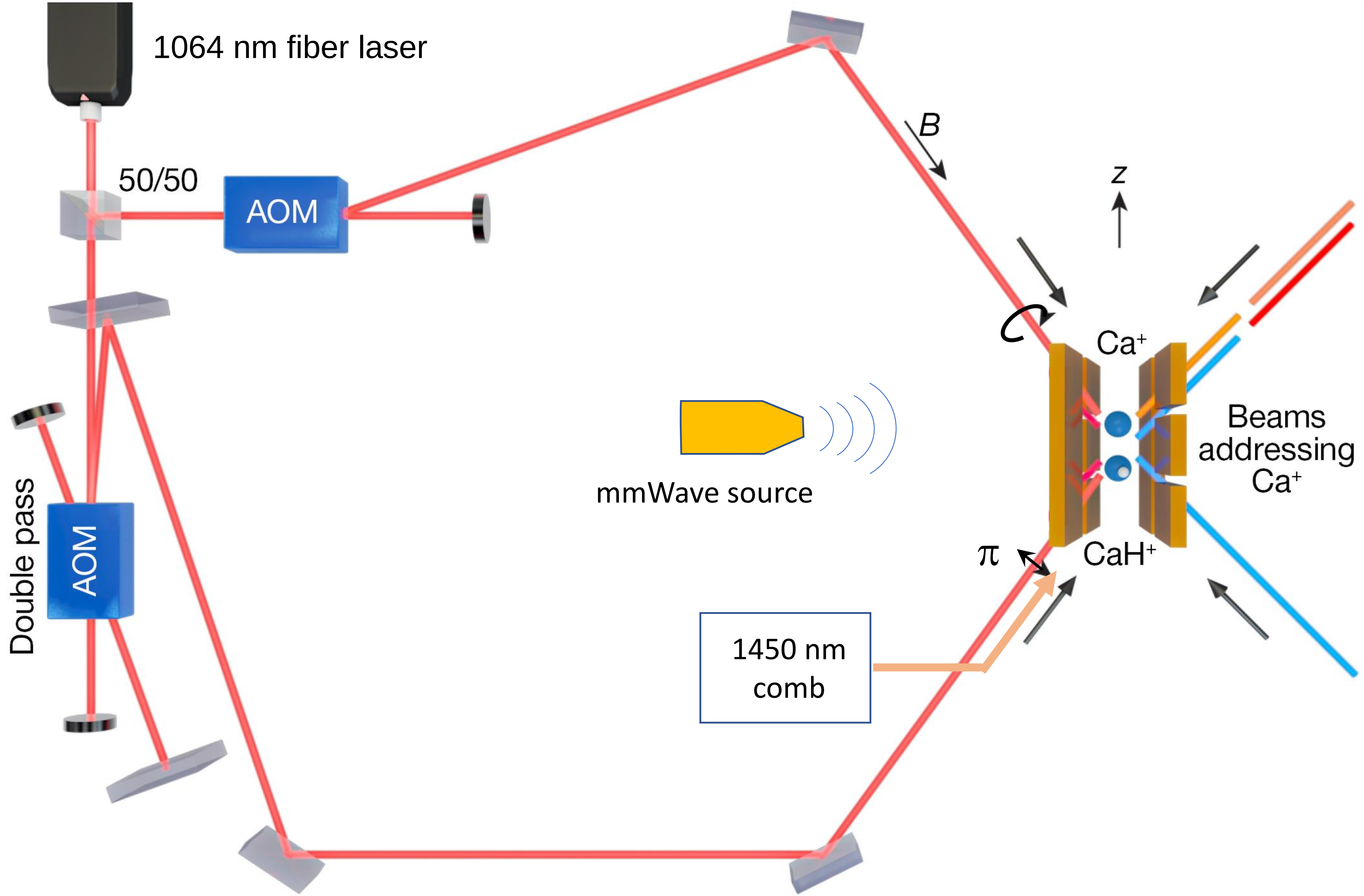


Frequency Comb with a Tunable Spectrum



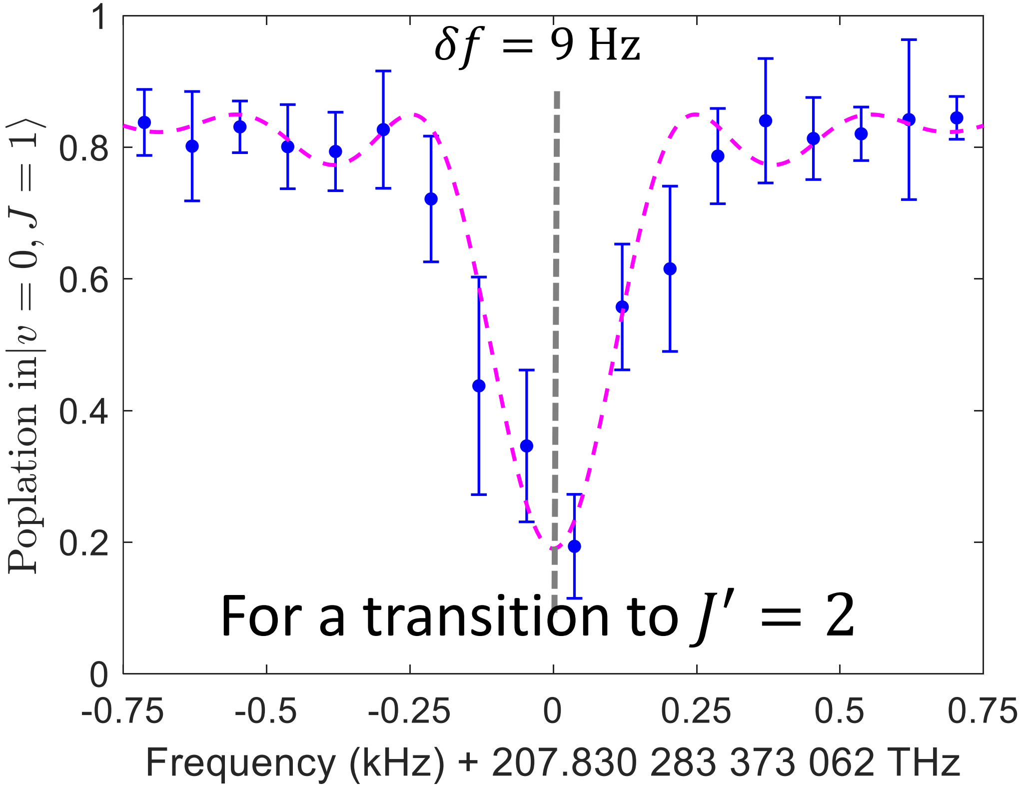


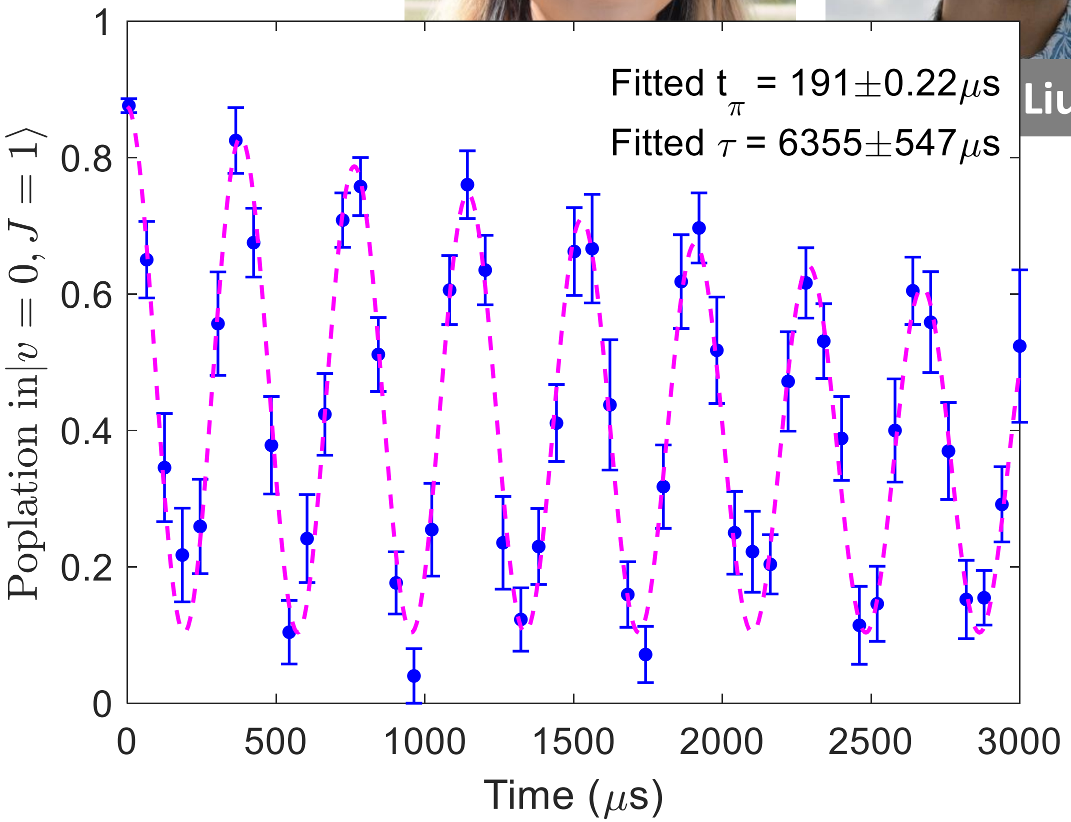



1064 nm fiber laser
50/50
AOM
Double pass
AOM
B
z
Ca+
CaH+
π
Beams addressing Ca+
mmWave source
1450 nm comb

Coherent Vibrational Spectroscopy

With 10^{-14} (statistical) uncertainty





The frequency of the comb tooth driving the transition can be determined precisely by taking the spectra at various comb repetition rates.

K. H. Leung et al., Terahertz Vibrational Molecular Clock with Systematic Uncertainty at the 10^{-14} Level, PRX 13, 011047 (2023)

Summary

Probabilistic projective **preparation** and **nondestructive detection** of molecular state with CW Raman beams

Coherent manipulation of molecular states with CW and comb Raman beams

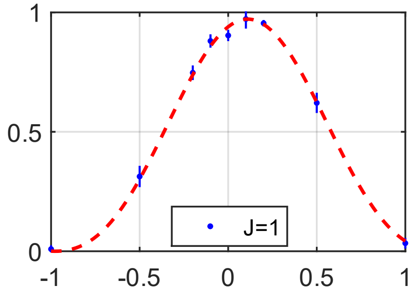
Precision spectroscopy of **THz rotational** transitions, and **telecom vibrational** transitions

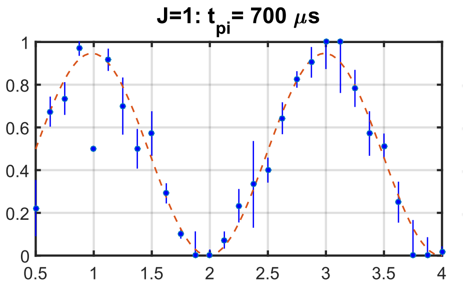
Measure dipole moments of molecular ions

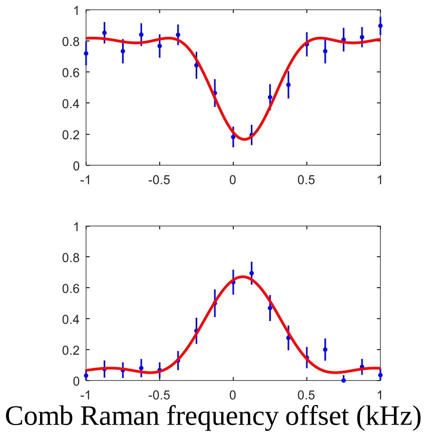
Using far-detuned Raman beams to manipulate molecular states, **applicable to other molecular species**

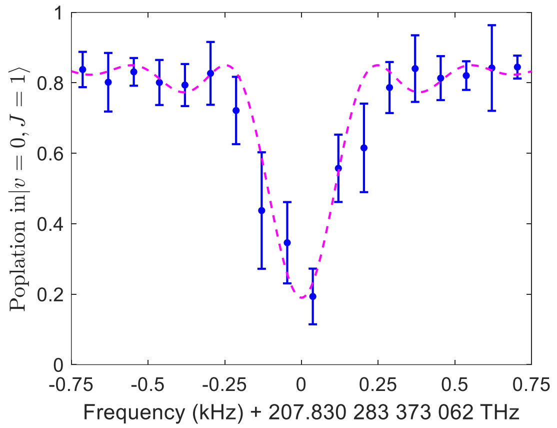
Tracking and trapping molecular population

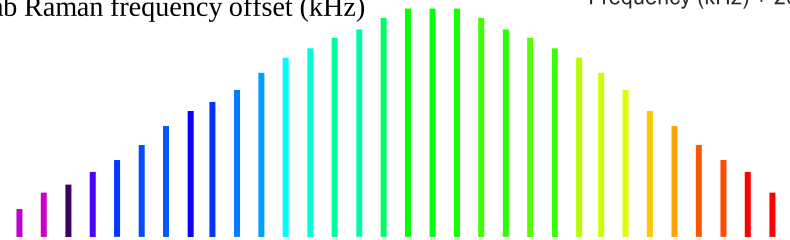
Atom-molecule **entanglement**

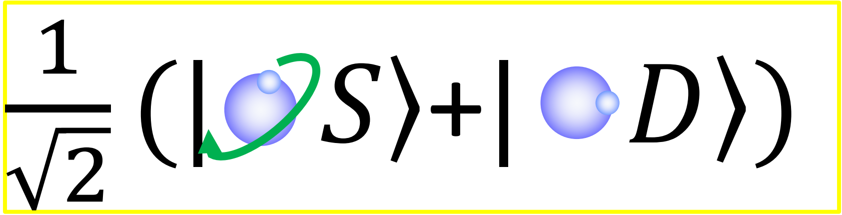


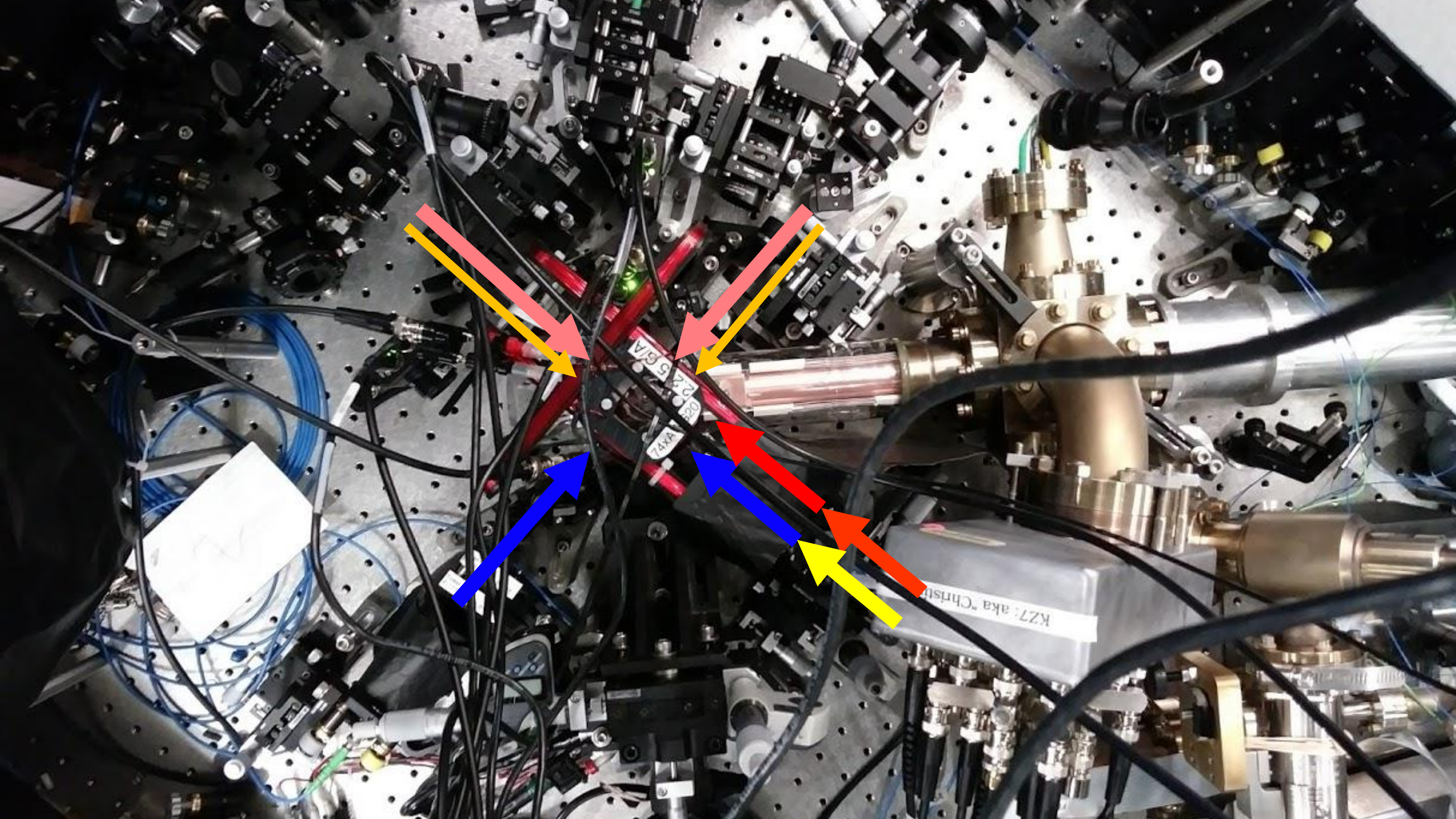




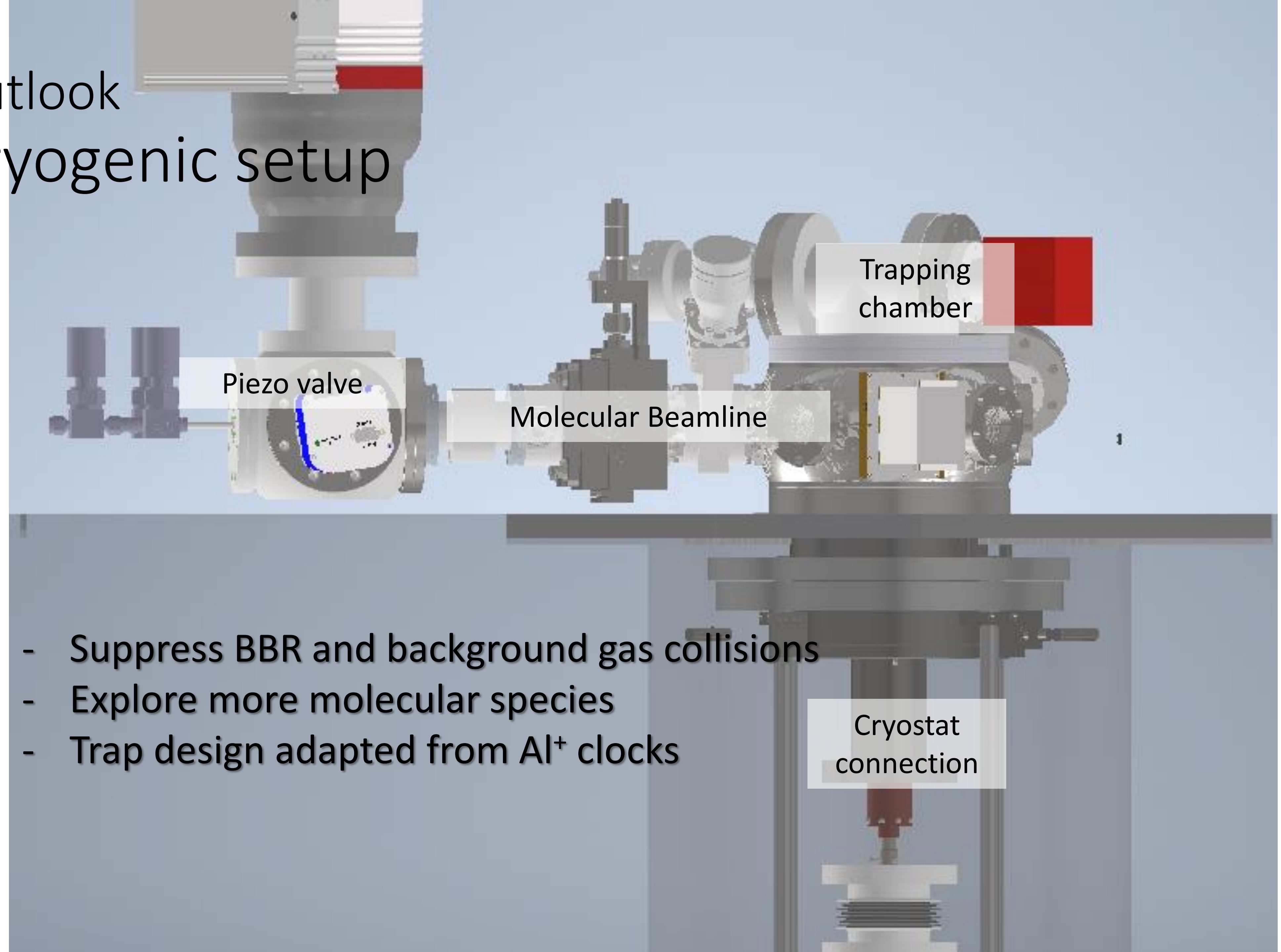




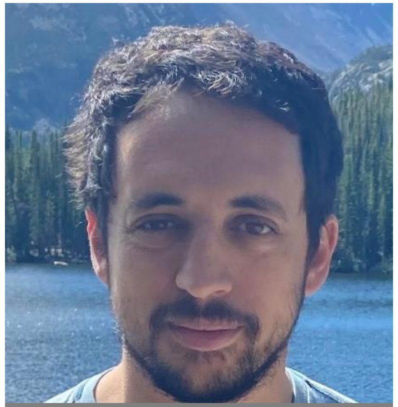




Outlook
Cryogenic setup



- Suppress BBR and background gas collisions
- Explore more molecular species
- Trap design adapted from Al^+ clocks

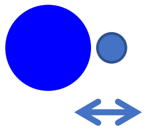

Baruch Margulis

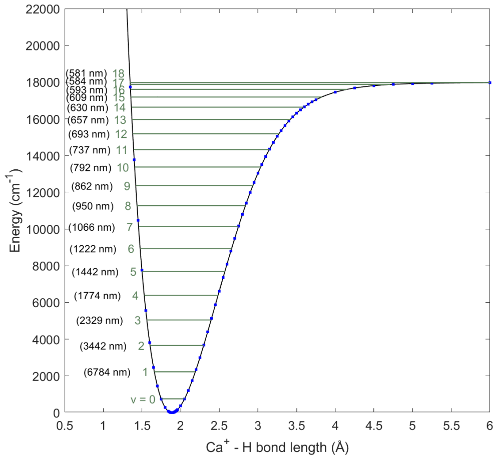

Julian Schmidt


Dalton Chaffee

Outlook

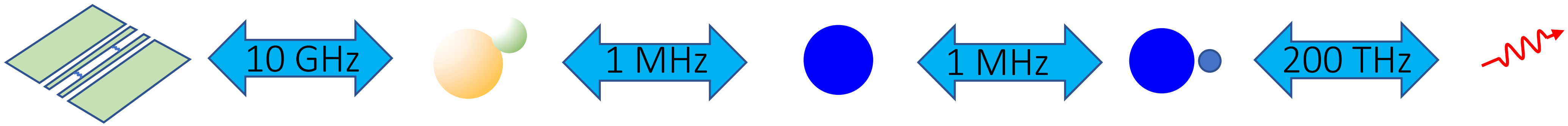
Further improve precision in vibration spectroscopy
- Characterize overtones and the potential energy curve
- Variation of fundamental constants





Polyatomic molecules
- Parity violation in chiral molecules
- Isomer identification

Hybrid systems for quantum information
- Transducing between platforms, e. g. telecom photons



Wide selection of qubit frequencies, efficient transduction between ions and telecom photons

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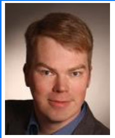
Michael Harding
Karlsruhe Institute of Technology


Scott Diddams
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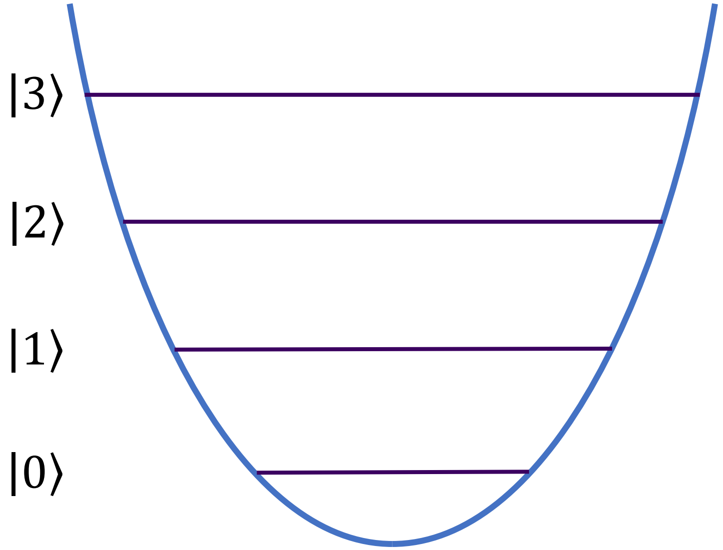
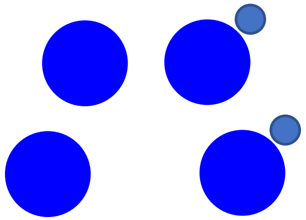
Quantum-Logic Spectroscopy: Information Bus
Quantized Harmonic Motion

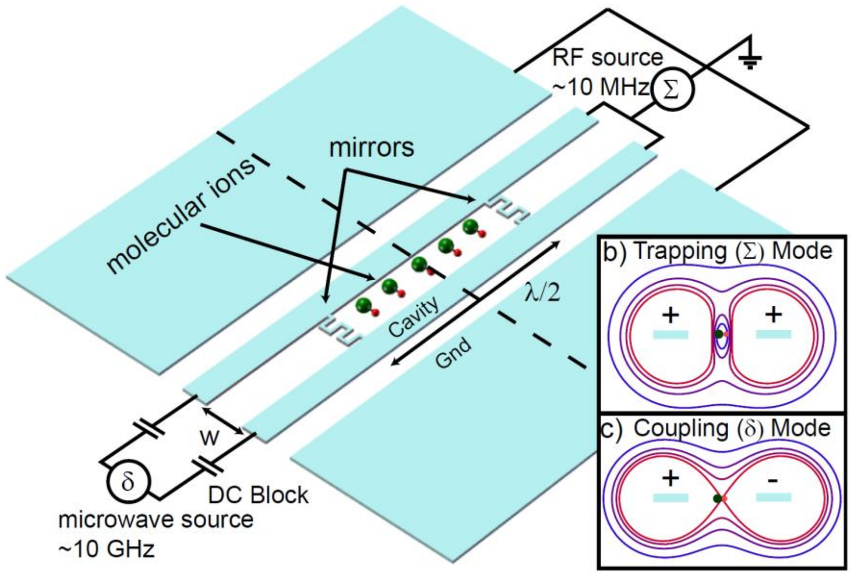


Use the atom for high-fidelity operation and detection on the states of the shared harmonic motion.

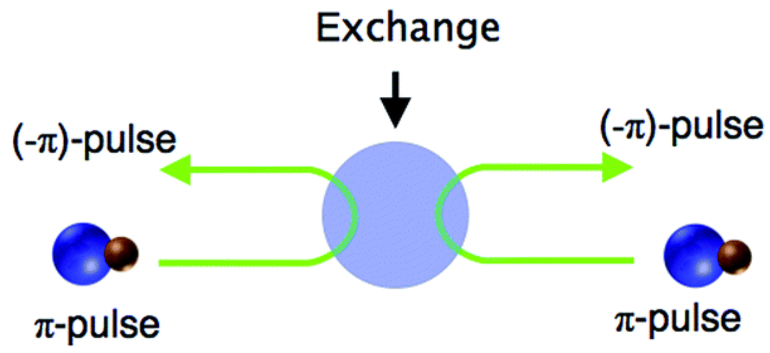


P.O. Schmidt, et al. Science 309, 749 (2005)

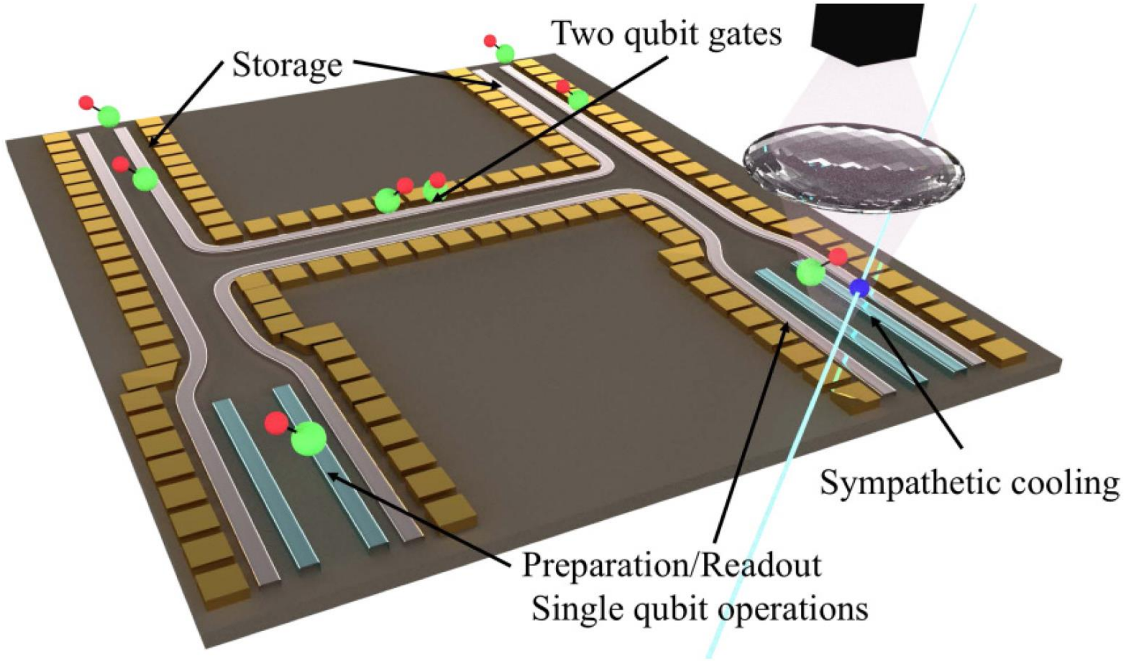
Molecules as Qubits



D. I. Schuster et al., Phys. Rev. A 83, 012311 (2011)



K.-K. Ni, T. Rosenband and D. D. Grimes, Chem. Sci. 9, 6830 (2018)



E. R. Hudson and W. C. Campbell, RA 98, 040302(R) (2018)

Entanglement between CaH^+ and Ca^+

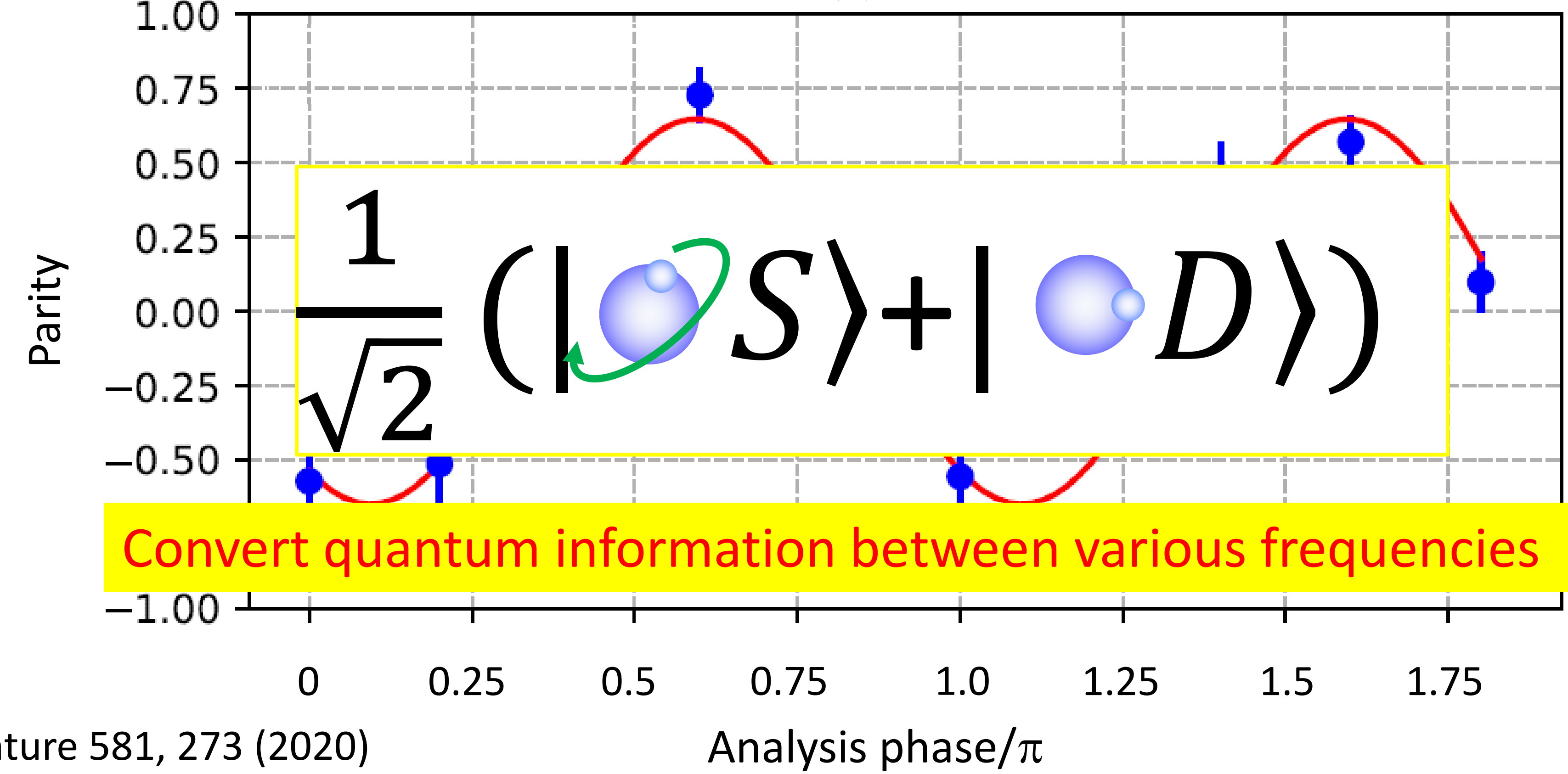
Fidelity

CaH^+ ion, one qubit state in J = 0 and the other in J = 2:

$|\downarrow'\rangle$: $J = 0, |\mathrm{m} = -1/2, -\rangle \leftrightarrow |\uparrow\rangle$: $J = 2, |\mathrm{m} = -3/2, -\rangle$, ~855 GHz

Entangled state $(|\uparrow, S\rangle + e^{i\phi}|\downarrow', D\rangle)/\sqrt{2}$ with ~76 % fidelity

$\mathrm{P}(|\downarrow', S\rangle) + \mathrm{P}(|\uparrow', D\rangle) = 0.87(3)$ Parity fringe contrast $= 0.65(5)$



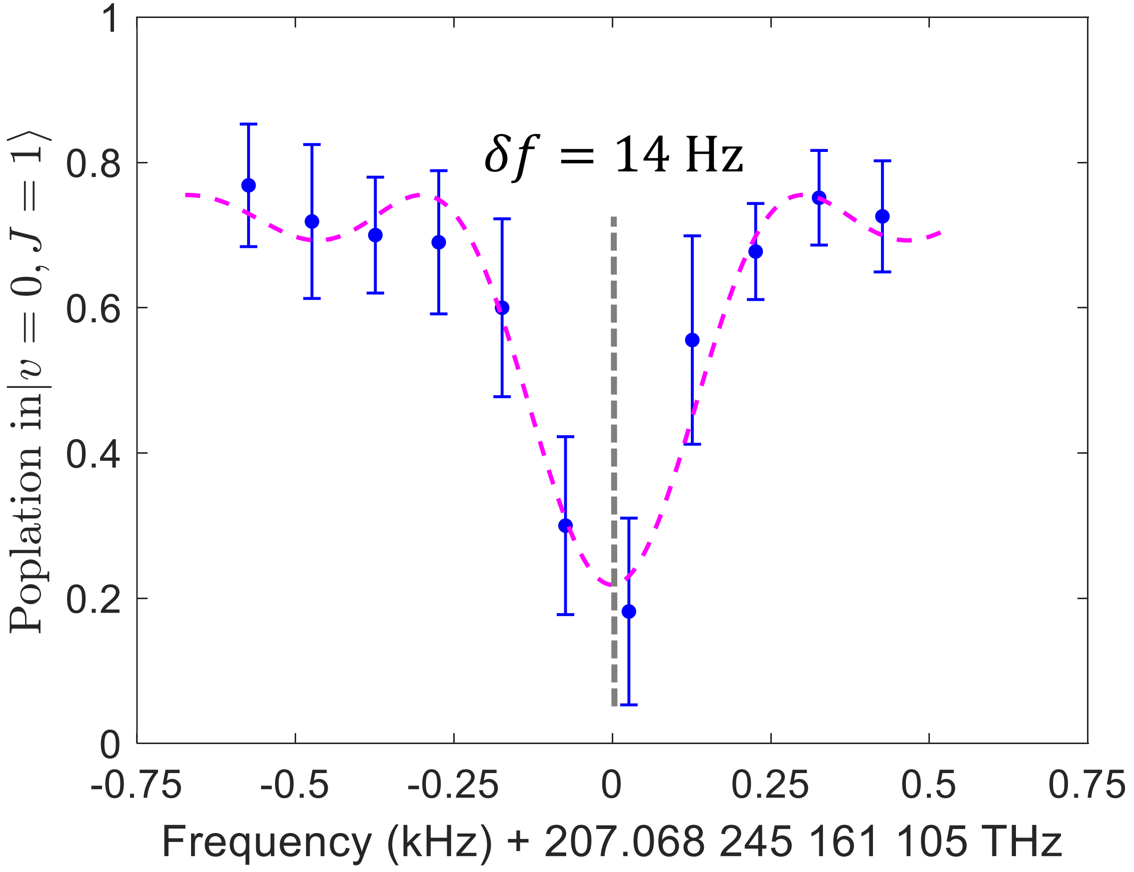
Convert quantum information between various frequencies

Yiheng Lin

Y. Lin et al., Nature 581, 273 (2020)

Coherent vibrational spectroscopy
Molecular properties for excited vibrational levels

For a transition to $J' = 0$



$J' = 0$ and $J' = 2$
frequency difference:
762.038211957(20) GHz
Rotational constant of v = 5
(coarse estimate) 129 GHz