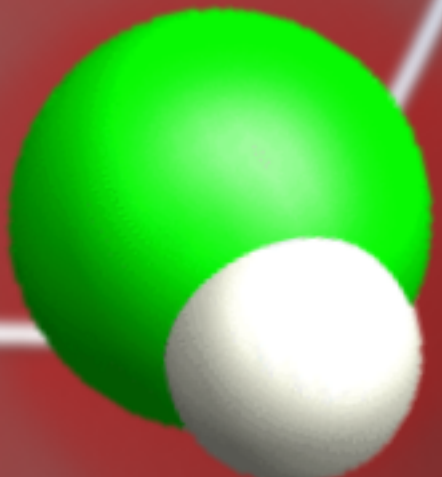
Quantum Logic Control of a Single Molecular Ion

Dietrich Leibfried

Chin Wen (James) Chou, David Leibrandt
Alejandra Collopy, Dalton Chaffee, Christoph Kurz, Yiheng Lin, Yu Liu, Julian Schmidt
Ion Storage Group

"*. . . we never experiment with just one electron or atom or (small) molecule.*"—E. Schrödinger, 1952

D. J. Wineland *et al.*, Proc. 6th Symp. Freq. Stand. Metr., 361 (2002).

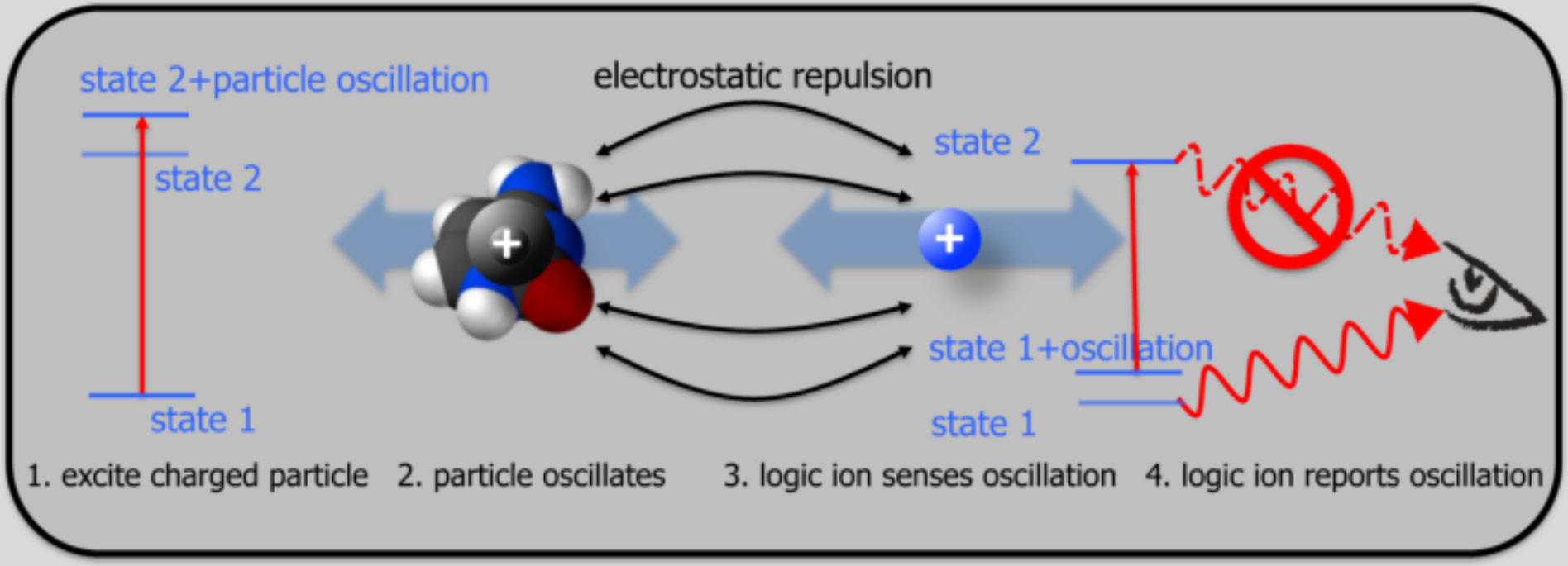


applications (existing and potential):

- optical clocks based on a single Al^{+} ion
- study and control electronic state of highly charged ion Ar^{13+} [PTB, Schmidt, Crespo,…]
- study and control rotation of a diatomic molecular ion
- study and control rotation and vibration of diatomic and polyatomic molecular ions
- couple to electrons, (anti-)protons or other charged systems without optical transitions
- …

motion as the messenger

infer the state of a charged particle by coupling its motion to an atomic "logic" ion with all the features we know and love

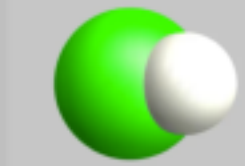


proposal: D. J. Wineland *et al.*, Proc. 6th Symp. Freq. Stand. Metr., 361 (2002)
realization: P. O. Schmidt, T. Rosenband *et al.*, Science 309 749 (2005)

molecules do more

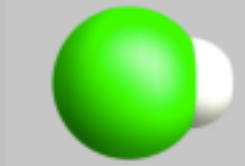
electronic transitions:
typically > 460 THz, eq. to > 22000 K
(electron spin)-orbit, fine structure
(electron spin)-(nuclear spin), hyperfine structure

atoms&molecules

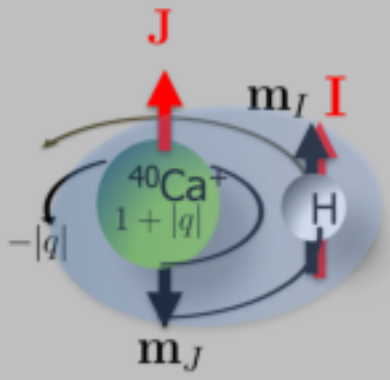


vibrational transitions:
first order: harmonic oscillator with energies $h\nu(n+1/2)$
typically ν > 6 THz, eq. to > 287 K

only molecules



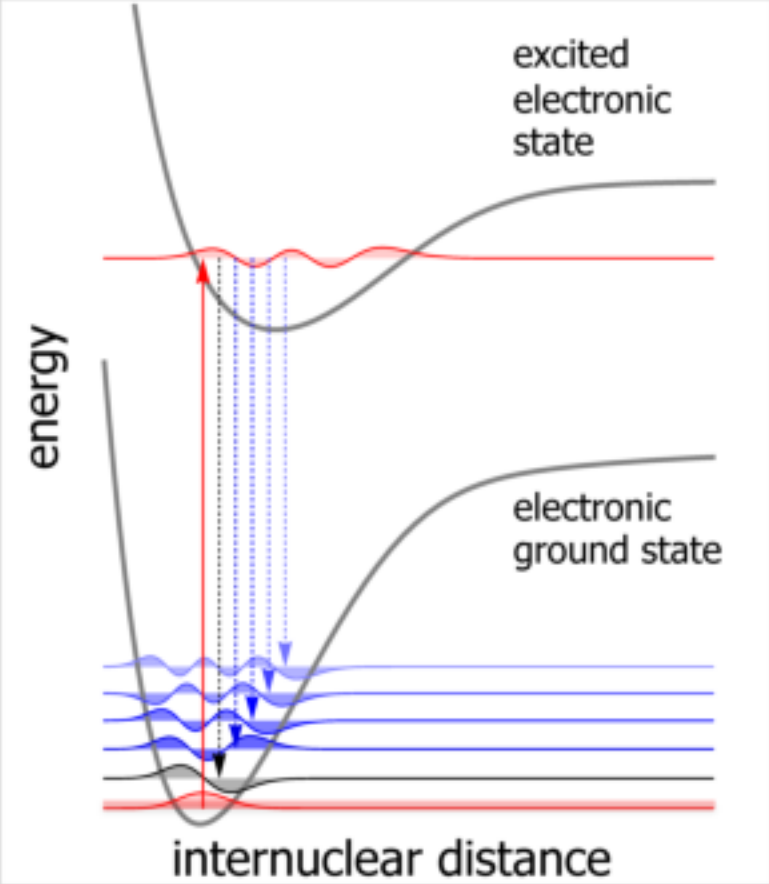
rotational transitions:
first order: rigid rotation with energies $hB\ J(J+1)$
rotational constant typically B < 1 THz, < 48 K



further degrees of freedom:
(electron spin)-rotation coupling (≈ MHz-GHz < 1 K)
(nuclear spin)-(nuclear spin) coupling (≈ kHz-MHz < 1 mK)
(nuclear spin)-rotation coupling (≈ kHz, < 1 μK)
present in nearly all molecules , frequencies easy to synthesize

room temperature: typical molecule in electronic and vibrational ground states

lost in Hilbert space



absorption and emission of photons similarly likely between many different vibrational levels of different electronic states

generally, no conservation laws that allow for cycling transitions to scatter many photons

only **very special** molecules can "mimic" very special atoms

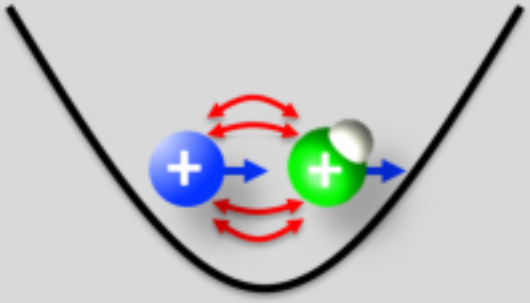
ways out:

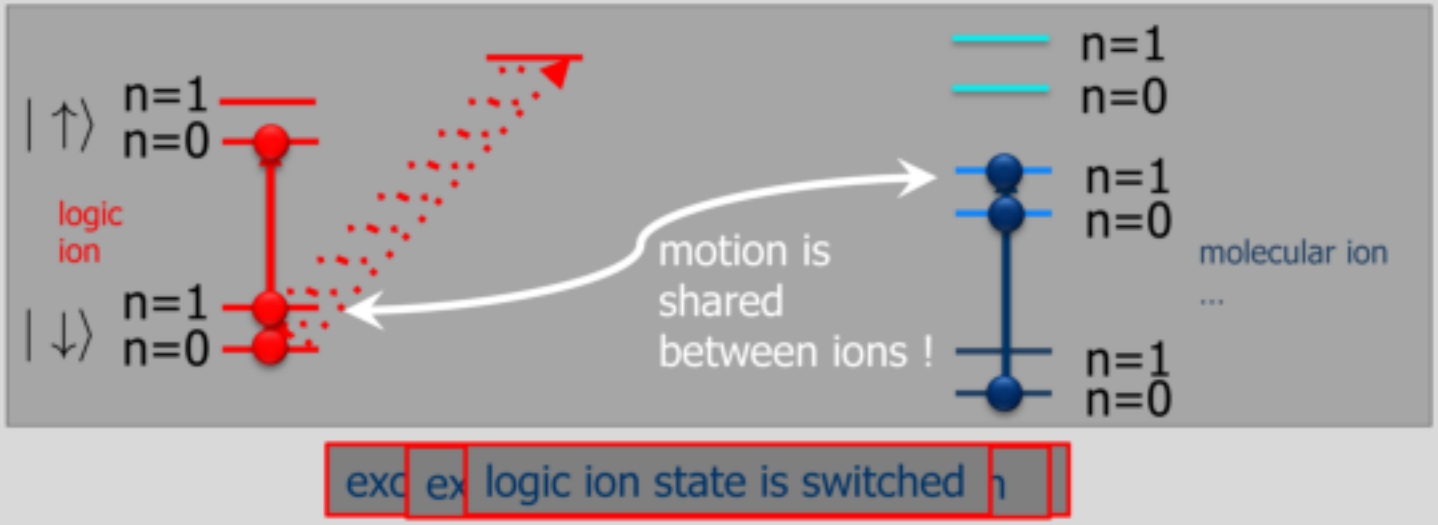
- restrict choices to "special atom-like" molecules
- build molecules from well controlled atoms
- avoid spontaneous emission of molecule

… but that means no light to observe

quantum-logic spectroscopy protocol

- logic and target ion in common potential
- Coulomb repulsion → normal modes
- select one shared mode and cool to ground state (n=0) on logic ion



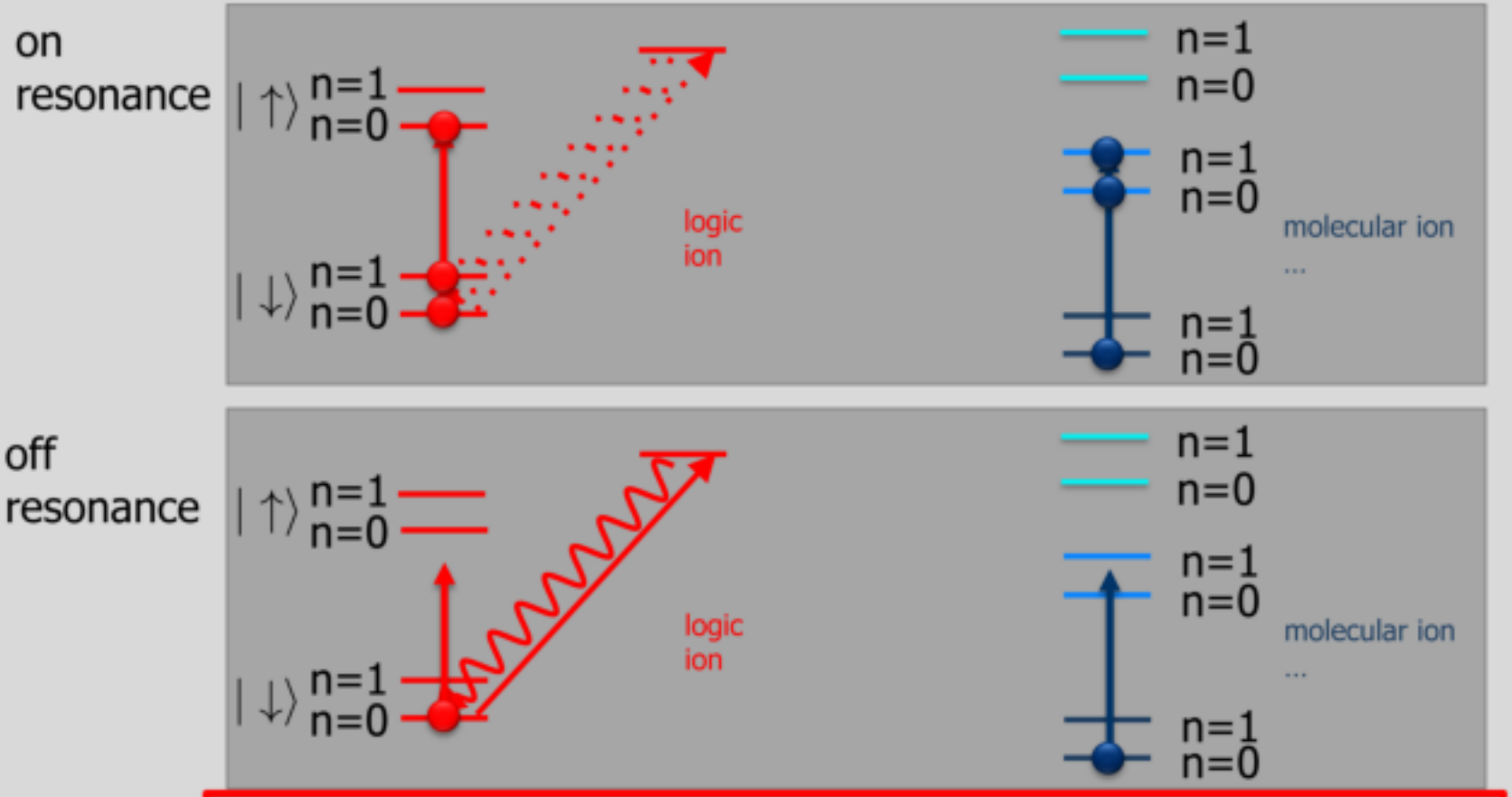


logic ion ("easy" to manipulate)

"invisible" system

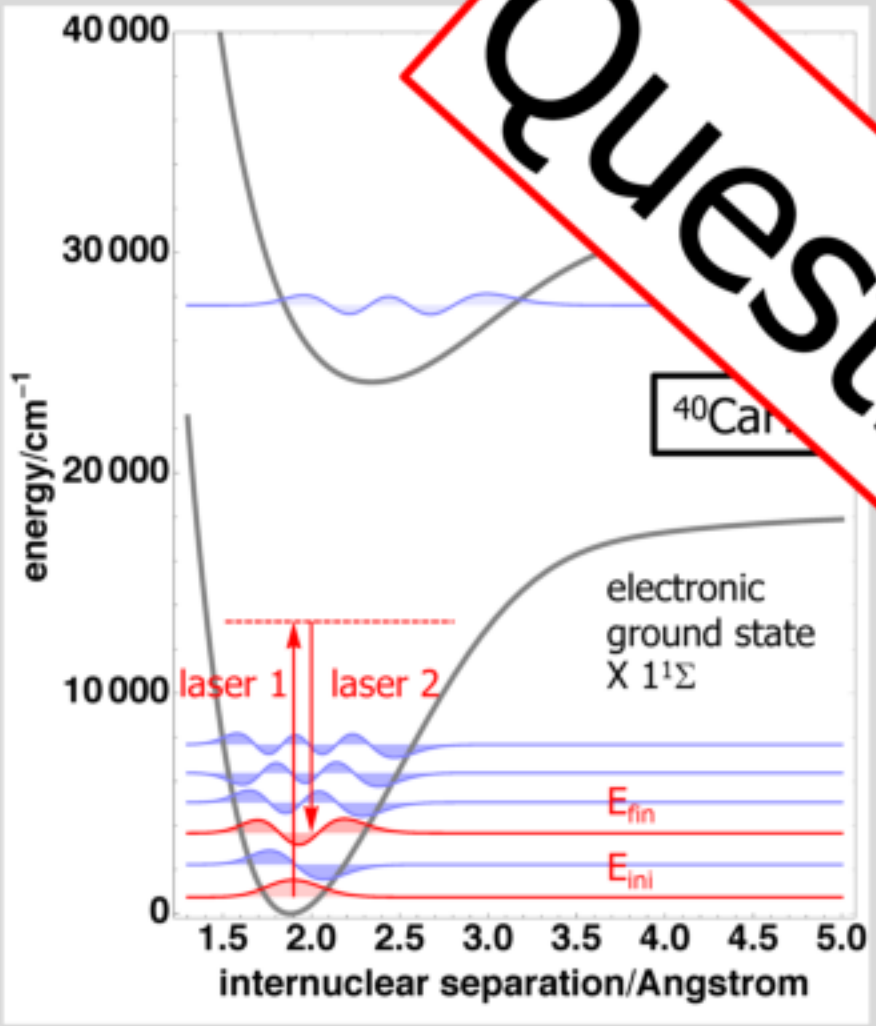
P. O. Schmidt *et al.*, Science 309 749 (2005)

quantum-logic spectroscopy protocol



logic ion state switches IFF molecular ion state switches
synchronous switch can be detected on logic ion
molecular ion is projected into pure final state

no photon left behind
(in the molecule)



Questions ?

stimulated* (laser1 + laser 2) two-photon Raman transitions

oid populating excited electronic state

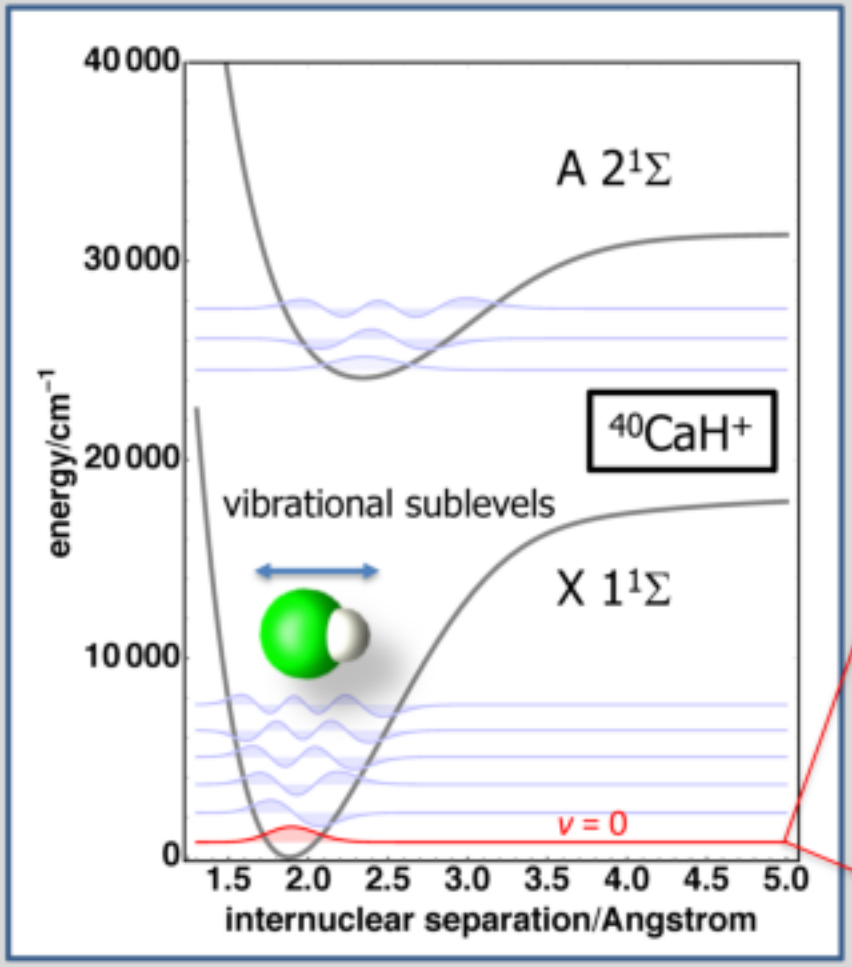
ifference between initial and final state

$E_{ini} = h (\nu_{laser\ 1} - \nu_{laser\ 2})$

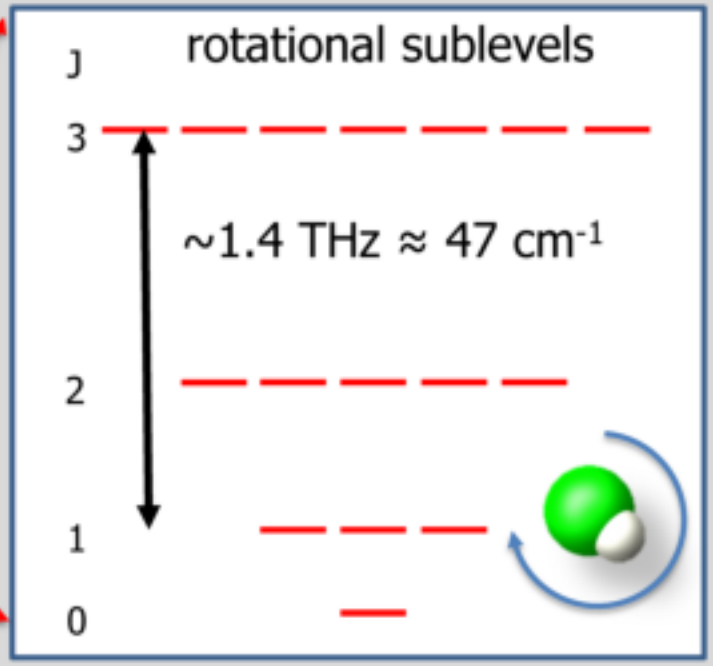
om electronic transition,
ion in higher electronic state

* traditional molecular Raman spectroscopy techniques involve at least one spontaneously emitted photon

proof of principle molecule $^{40}CaH^+$



- Load two Ca^+
- increase H_2 background pressure
- $(Ca^+)^* + H_2 \rightarrow CaH^+ + H$
- molecule decays to X $1^1\Sigma$, $v = 0$



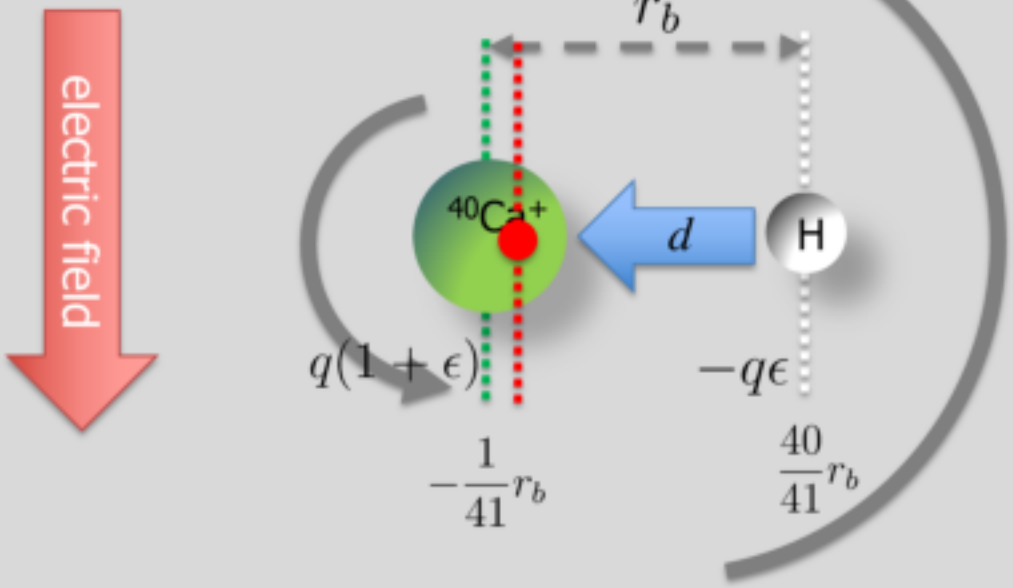
dipole moment

$^{40}Ca^{+}$ closed shells, one valence electron

valence electron from Ca^{+} moves closer to H to form a singlet state with H valence electron

H, one valence electron, needs 2nd electron to close shell

torque in electric field



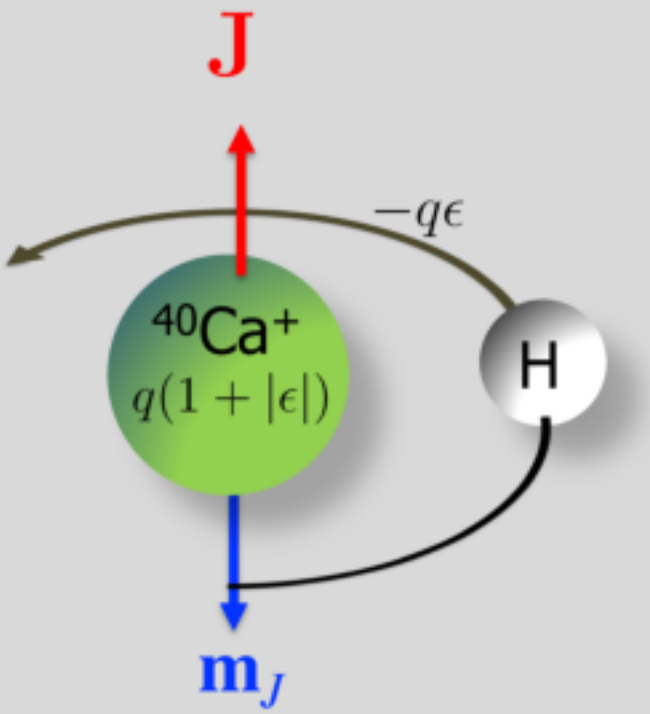
$0 < \epsilon < 1$

system with net charge and no radial symmetry

center of mass ≠ center of charge

rotational magnetic moment

CaH^+ rotates perpendicular to bond axis around COM, rotation vector **J**

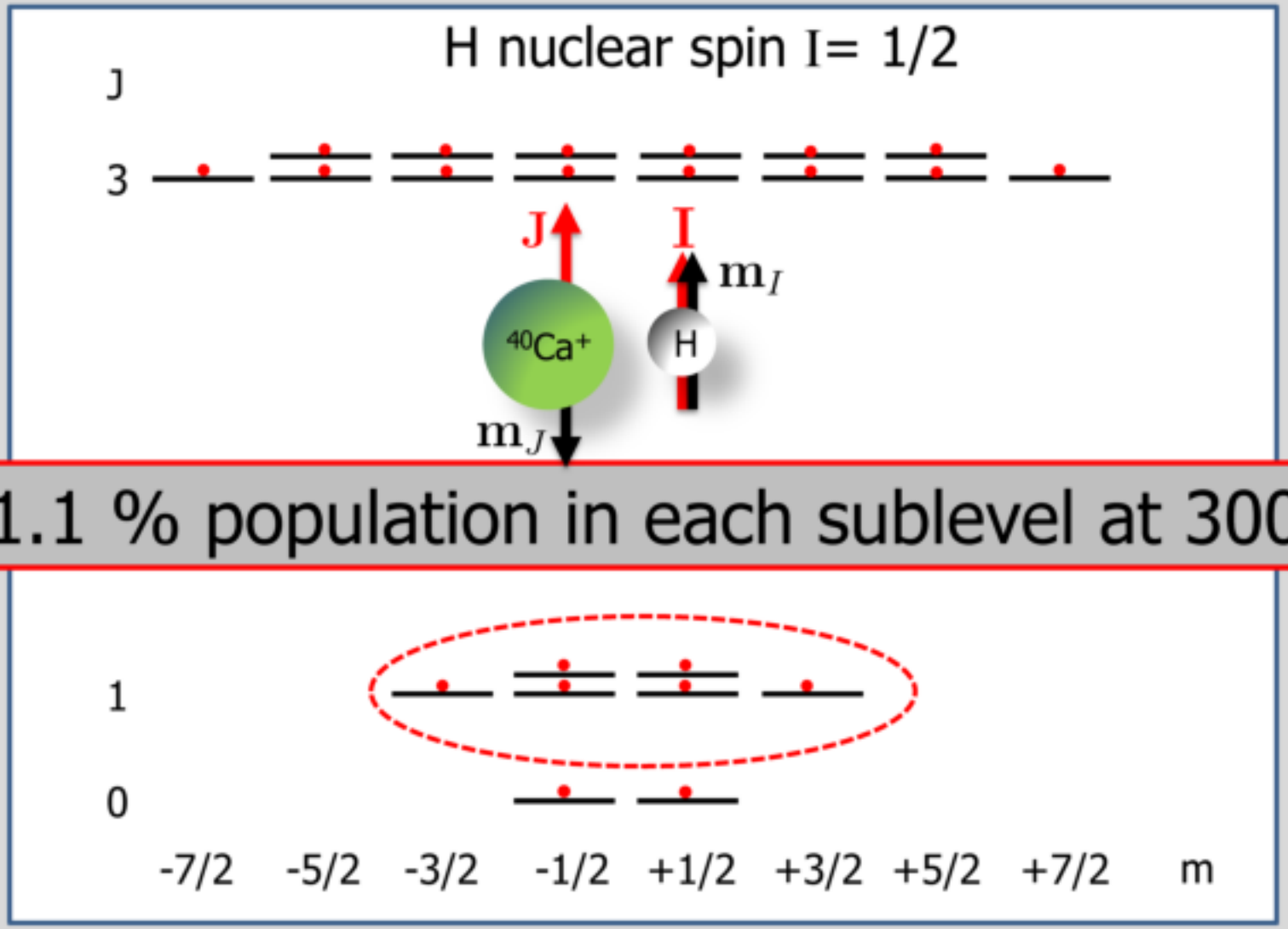


rotating charge distribution creates net negative current

induced rotational magnetic moment $\mathbf{m}_J$ anti-parallel to rotation vector **J**

$^{40}CaH^+$

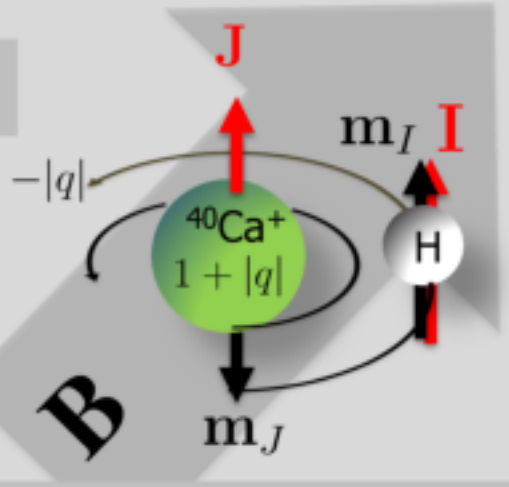
rotational and nuclear structure



≤ 1.1 % population in each sublevel at 300 K

finite magnetic field

J = 1, B = 0.36 mT (intermediate regime)



"signature" transition ~ 10 kHz

$$H = -g_J \frac{\mu_n}{\hbar}\mathbf{J}\cdot\mathbf{B} - g_p \frac{\mu_n}{\hbar}\mathbf{I}\cdot\mathbf{B} - \frac{2\pi c_{IJ}}{\hbar}\mathbf{I}\cdot\mathbf{J}$$

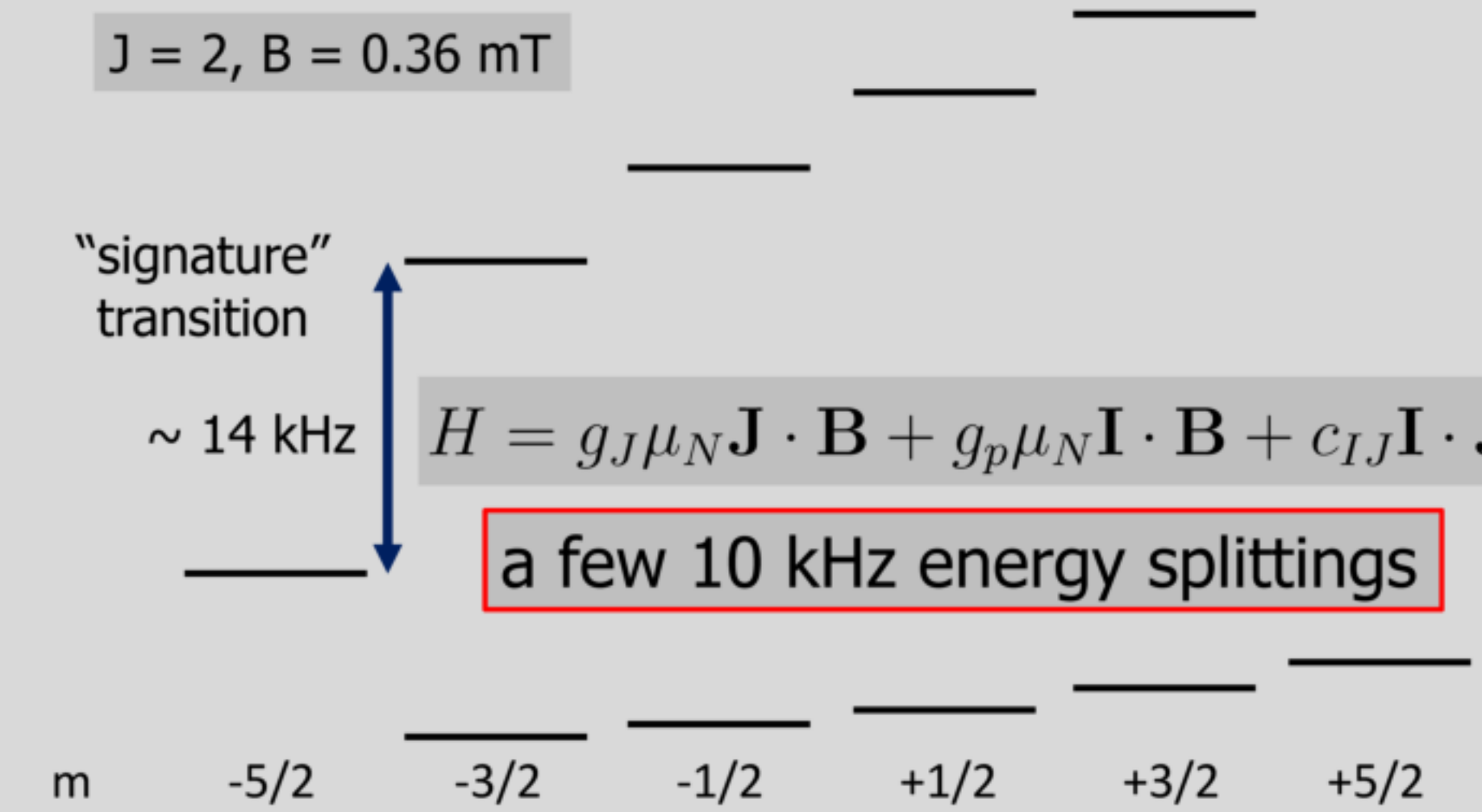
$$g_J\mu_N/h \simeq -10.2\ \mathrm{kHz/mT} \quad c_{IJ} \approx 8.5\ \mathrm{kHz}$$

$$g_p\mu_N/h \simeq 42.6\ \mathrm{kHz/mT}$$

m -3/2 -1/2 +1/2 +3/2

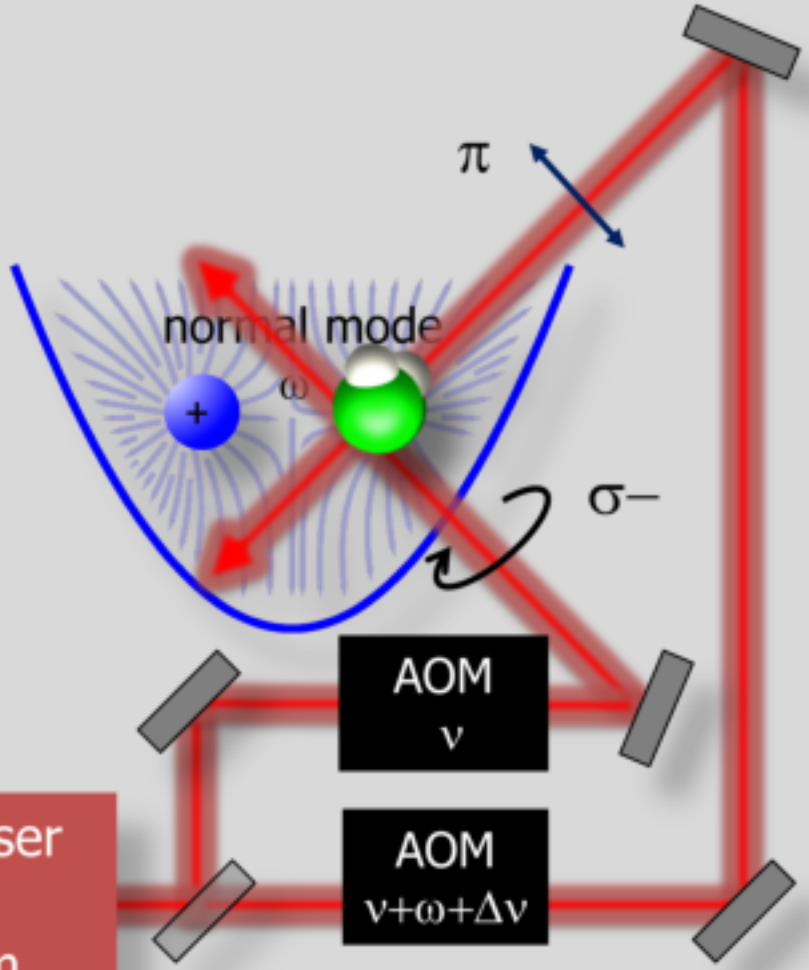
C.W. Chou et al., Nature 545, 203 (2017)

J=2 signature transition



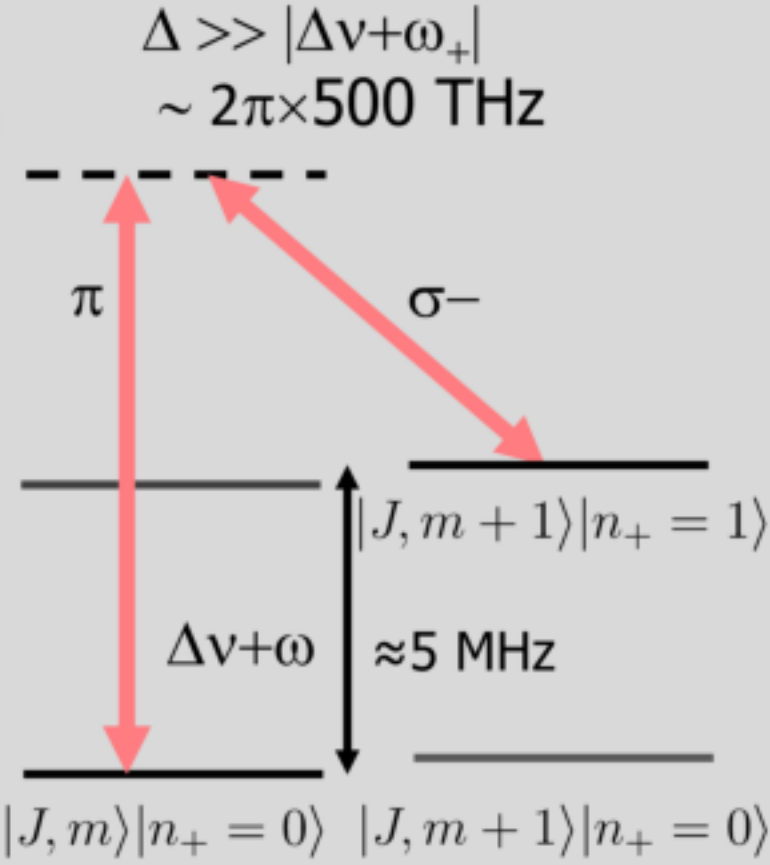
C.W. Chou et al., Nature 545, 203 (2017)

blue sideband transition



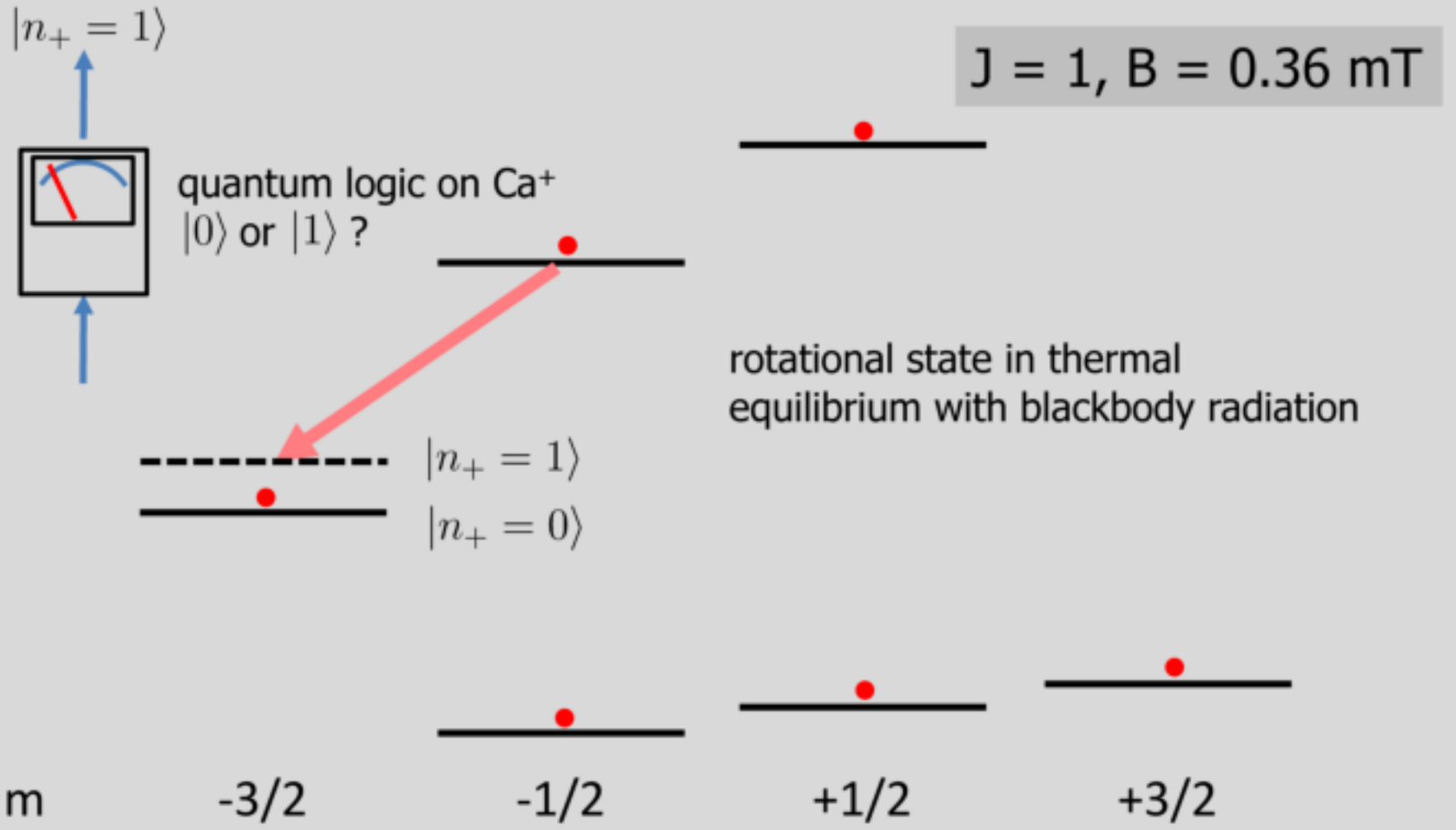
Raman laser

1051 nm



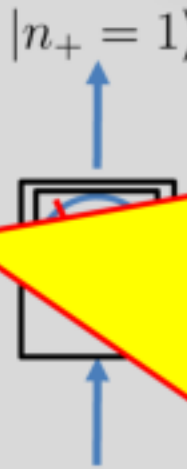
angular momentum and motion change

projecting into a pure state



D. Leibfried, New J. Phys. 14, 023029 (2012)

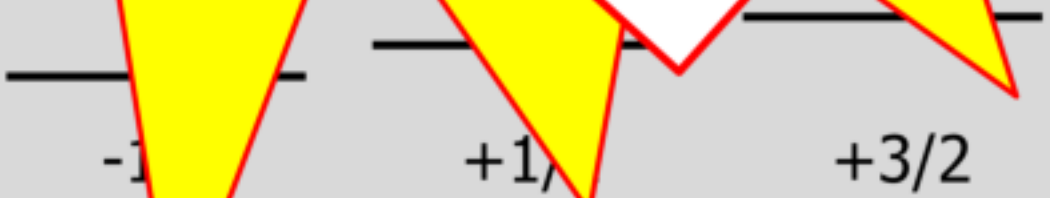
projecting into a ure state



- 0.36 mT

Questions ?

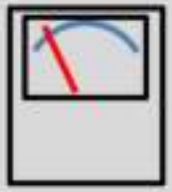
proje ible dynamics
leaves m re quantum state
cool motion - also irreversible
→ rotational pu



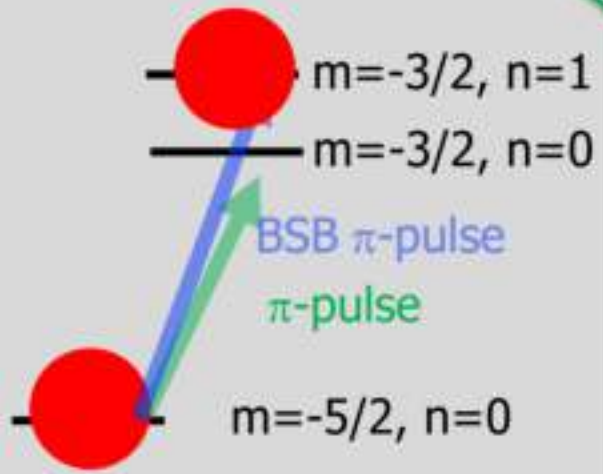
D. Leibfried, ew J. Phys. 14, 023029 (2012)

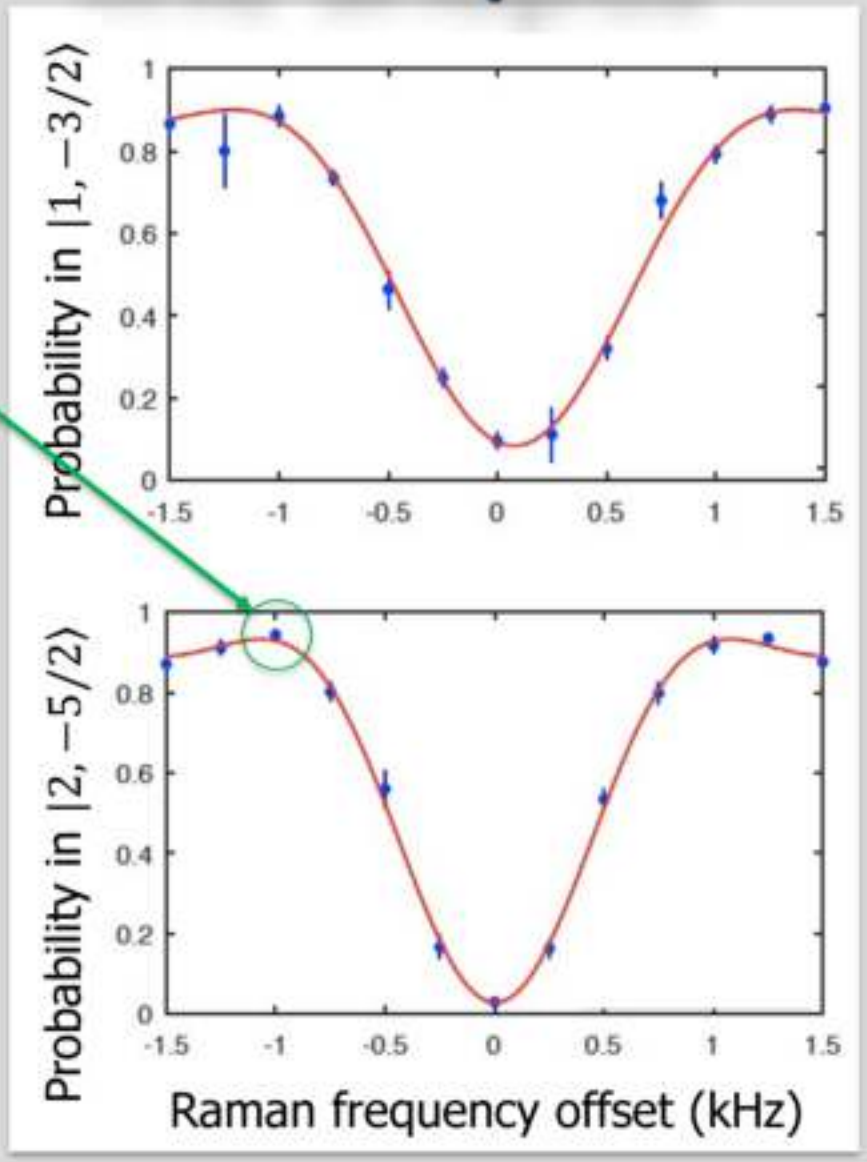
starting from a pure state
line shapes

quantum logic
on Ca^+, $|0\rangle$ or $|1\rangle$?



$|1\rangle$





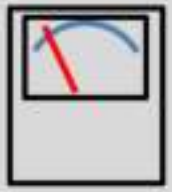
J=1

J=2

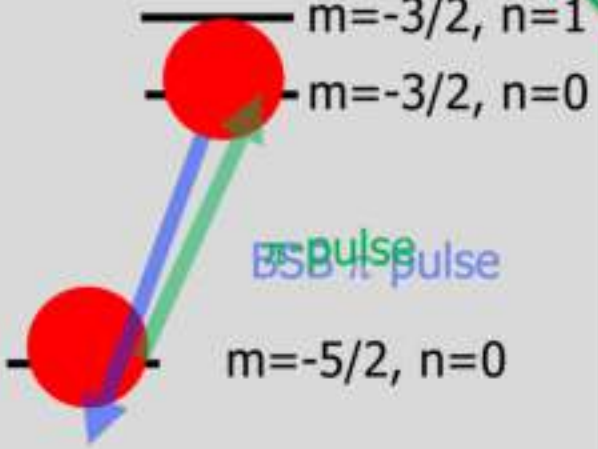
C.W. Chou et al.,
Nature 545, 203
(2017)

starting from a pure state
line shapes

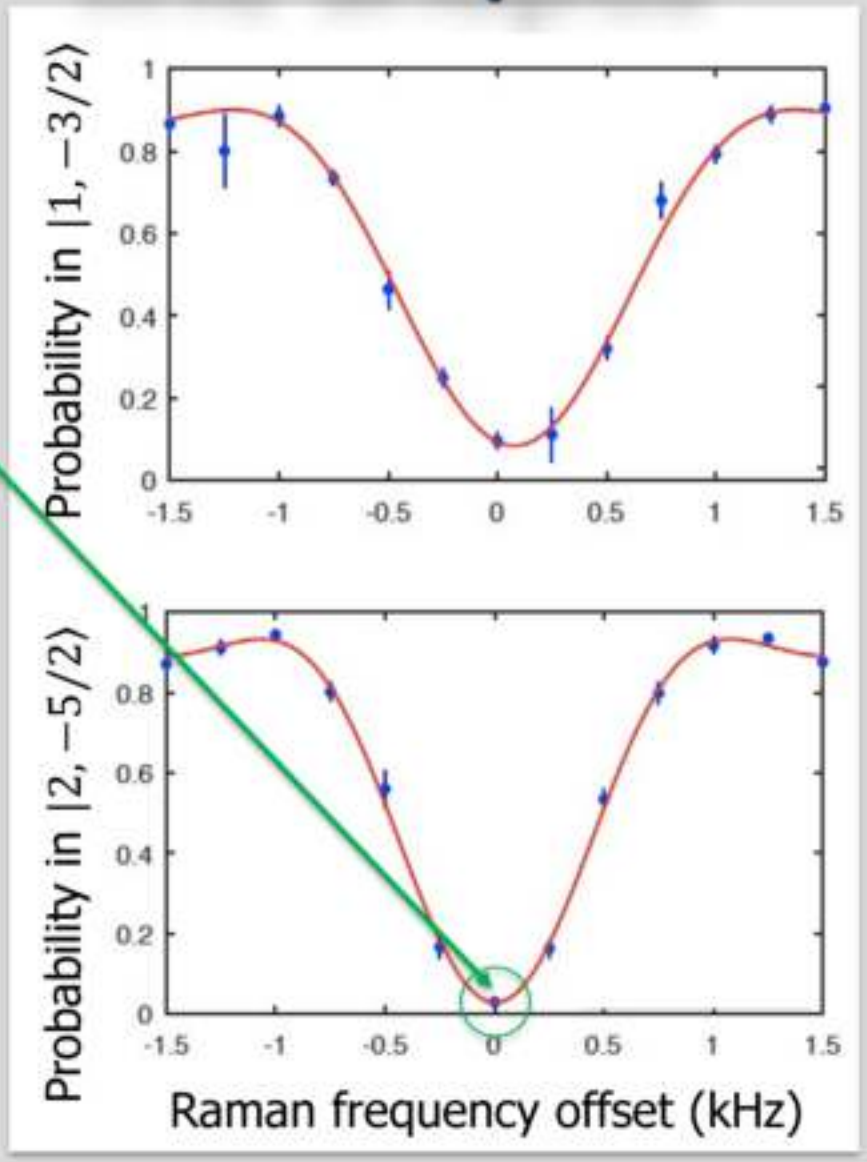
quantum logic
on Ca^+, $|0\rangle$ or $|1\rangle$?



$|0\rangle$



high contrast
transform limited
carrier transitions



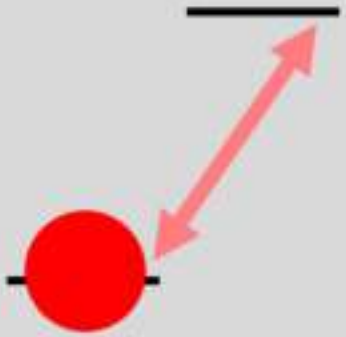
J=1

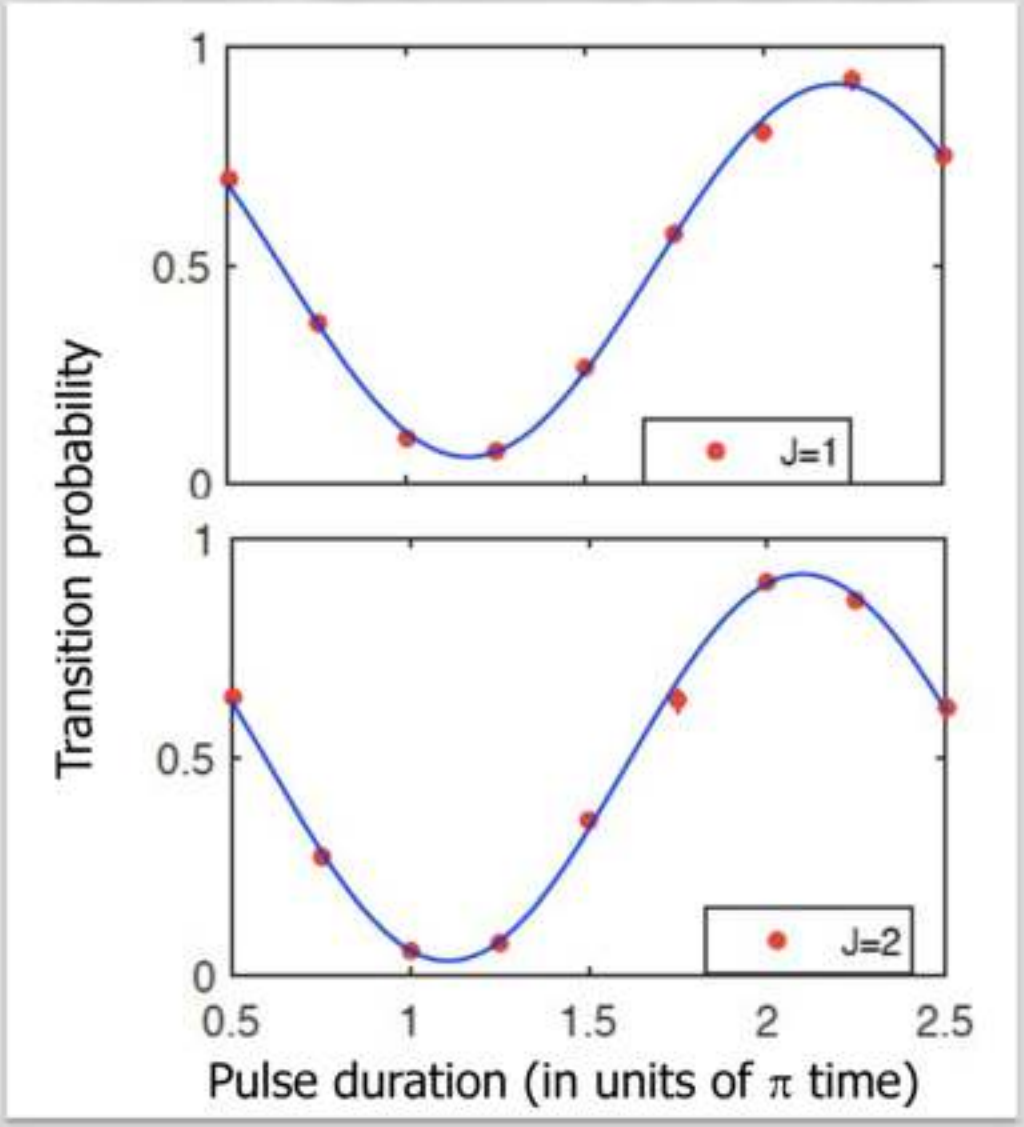
J=2

C.W. Chou et al.,
Nature 545, 203
(2017)

coherent manipulation
Rabi oscillations

quantum logic
on Ca^+, $|0\rangle$ or $|1\rangle$?





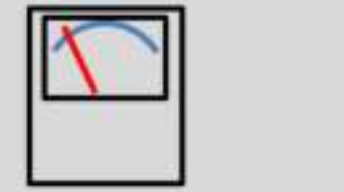
J=1
t_π=806 μs

J=2
t_π=955 μs

C.W. Chou et al., Nature 545, 203 (2017)

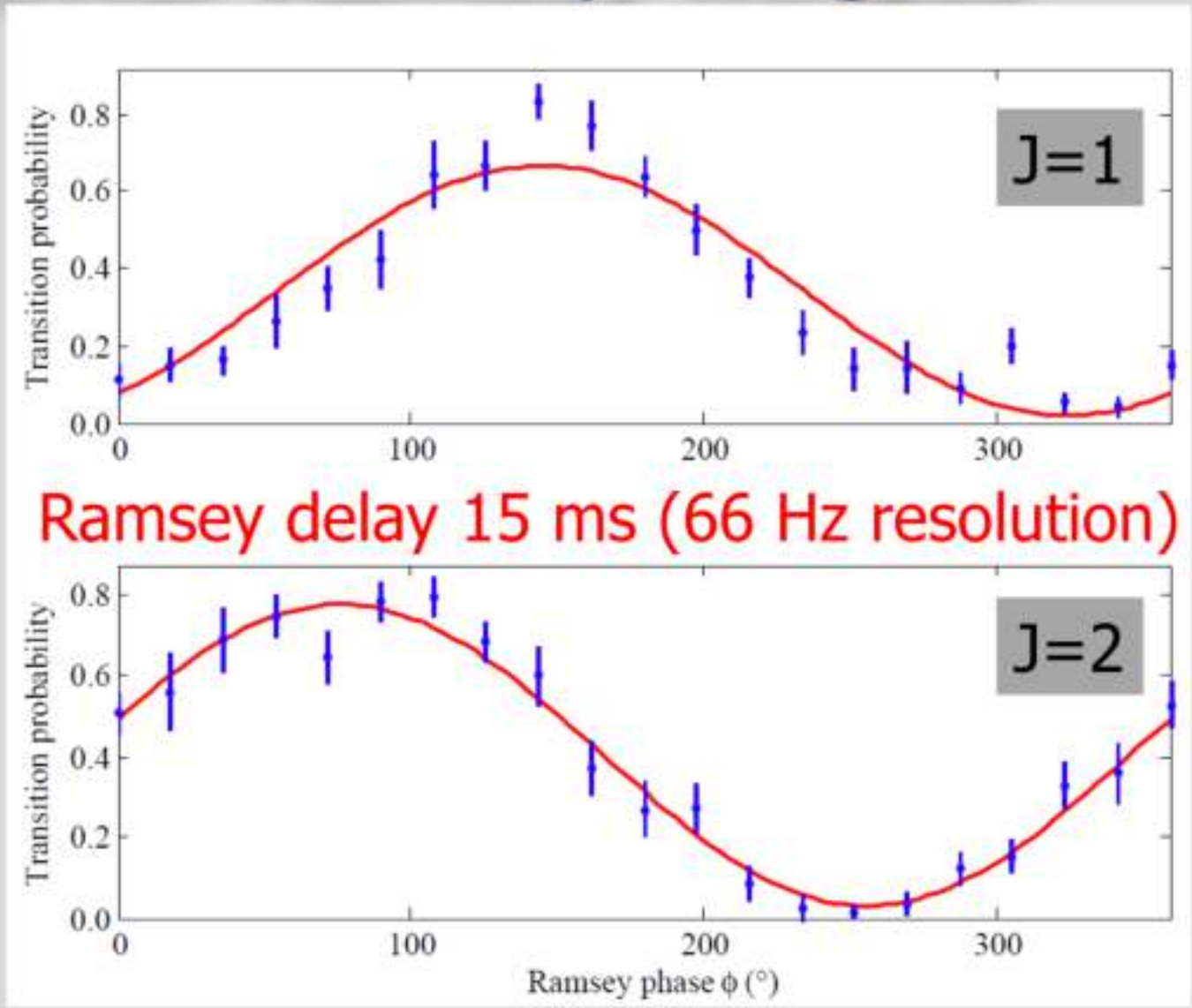
coherent manipulation
carrier Ramsey fringes

quantum logic
on Ca^+, $|0\rangle$ or $|1\rangle$?



$\pi/2$ - delay - $\pi/2$

C.W. Chou et al.,
Nature 545, 203
(2017)



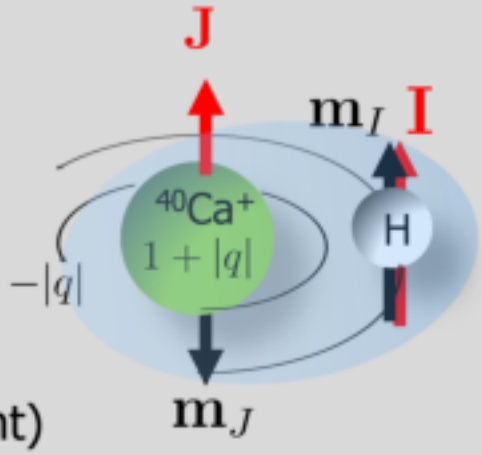
a fine fingerprint

(thanks to Heather Lewandowski !)

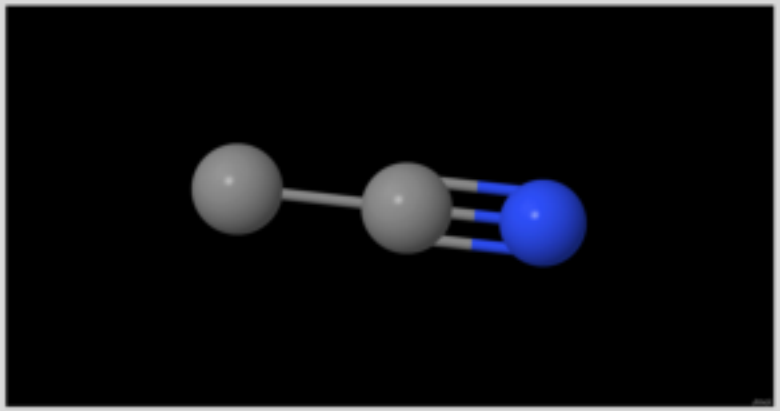
general spin-rotational structure for fixed J:

nucleus-nucleus: ≈ 1-100 kHz

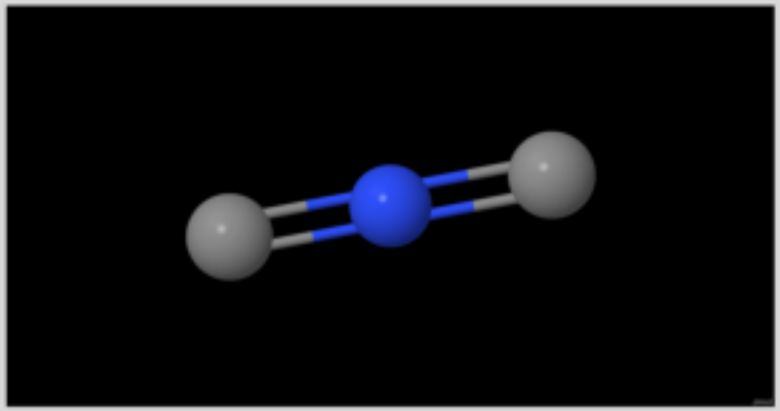
rotational magnetic moment-nucleus: ≈ 1-100 kHz



- energy splittings accessible with cw-Raman beams
- structure provides an alternative molecular fingerprint
- distinguishes isomers (same nuclei in different arrangement)



C_2N^+



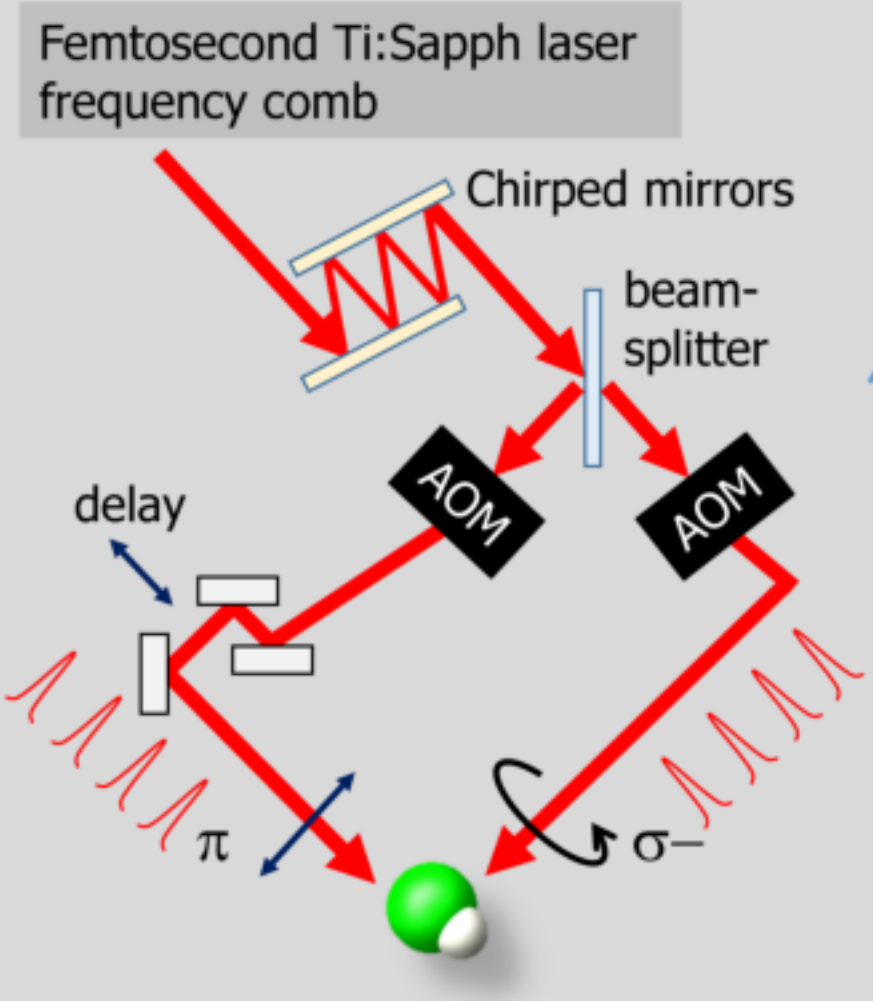
CCN^+ d ≈ 3 D I_N=1 Σ ground state

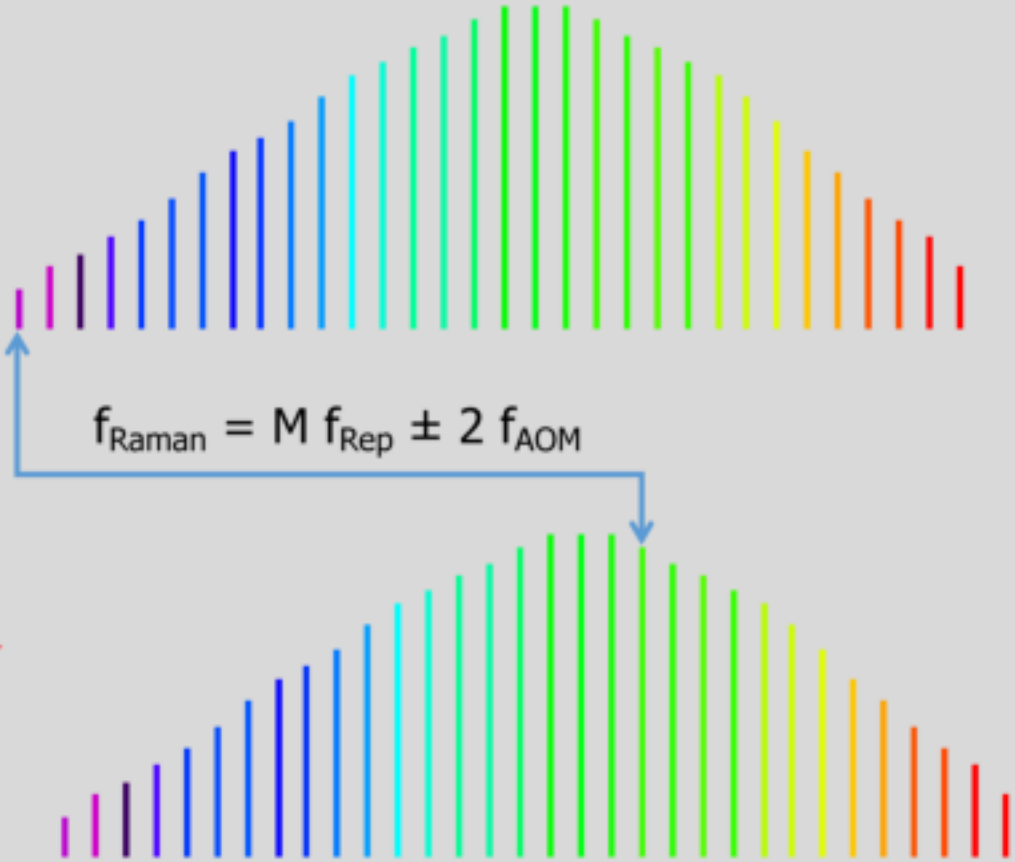
CNC^+ d = 0 I_N =1 Σ ground state

"predicted to be one of the most abundant molecular ions in the interstellar medium" *

*Hartquist and Dalgamo in "Giant molecular clouds in the galaxy", Pergamon (1980).

THz rotational transitions with a frequency comb





S. Ding and D. Matsukevich, New J. Phys. **14** 023028 (2012)
D. Leibfried, New J. Phys. **14** 023029 (2012)

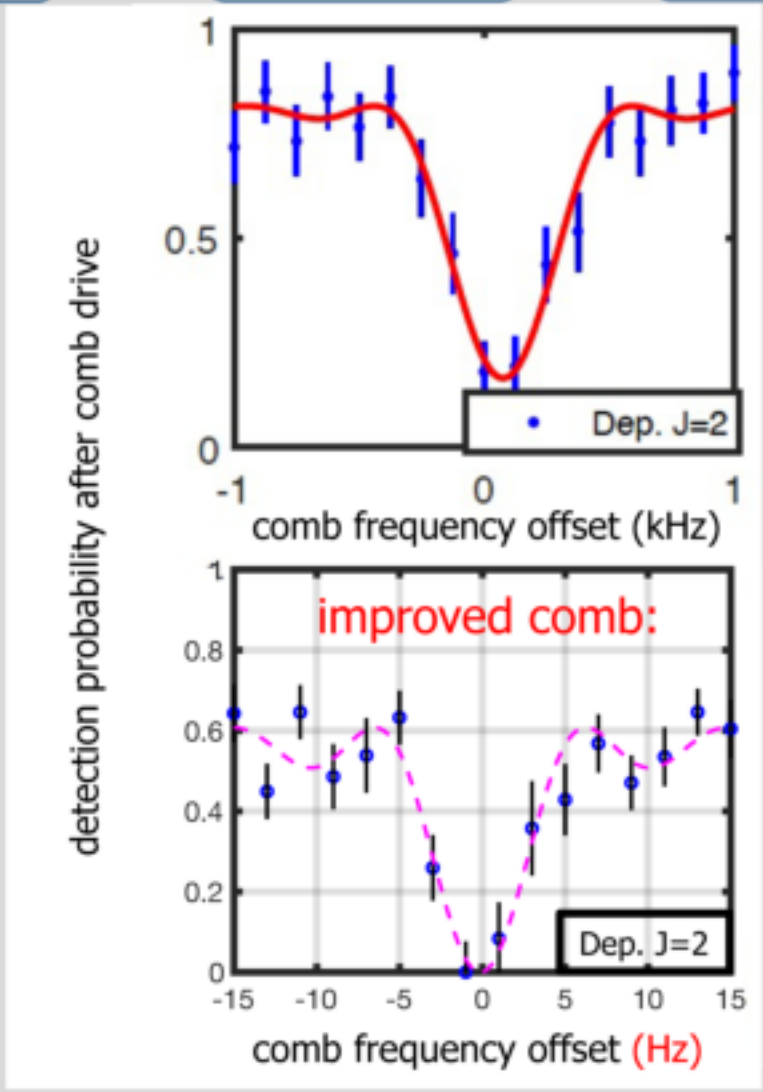
THz rotational transitions

State preparation → Comb pulse → State detection

example transition
J=2 to J=4
1.6 ms probe time

transition frequency
1.992 911 000 14 THz
± 20 Hz
transform limited line
FWHM ≈ 250 Hz

line center uncertainty
≈ 20 Hz



news from the lab:
J=2 to J=4
128 ms probe time

transform limited line
FWHM ≈ 5 Hz, lifetime limited coherence (?)

line center uncertainty
≈ 0.5 Hz (≈ 2.5×10^{-13} resolution)

C.W. Chou et al., Science **367**, 1458 (2020)

rotational transition frequencies

B-field free centroid of spin-rotation splitting: $E_J = h \sum_{k=1,2,\ldots} C_k J^k (J+1)^k$

J''	J'	$f_{J'',J'}$ (THz)	Statistical uncertainty $\delta f_{J'',J'}$ (Hz)	$cf_{J'',J'}$ (THz)	$\delta cf_{J'',J'}$ (kHz)
1	3	1.424 204 460 565	14	1.424 204 457 7	2.4
2	4	1.992 911 000 121	16	1.992 910 990 8	3.3
3	5	2.560 643 630 446	20	2.560 643 614 2	3.7
4	6	3.127 125 998 610	63	3.127 125 974 8	4.5

k	Experimental C_k (Hz)	Ab initio C_k (Hz)	Comments
1	1.42 501 777 9 (17) $\times 10^{11}$	1.427 (11) $\times 10^{11}$	B_R (rotational constant)
2	−5.81217 (19) $\times 10^{6}$	−5.831 (19) $\times 10^{6}$	$-D_R$ (centrifugal correction)
3	222.9 (7.2)	222.6 (0.6)	H_R (second centrifugal correction)
4	−0.021 (88)	−0.0158 (4)	Third centrifugal correction

C.W. Chou et al., Science **367**, 1458 (2020)

dipole moment measurement

terizing its electronic structure and radiative energy-transfer processes. For a large number of neutral molecules, precise measurements (0.01 %) of dipole moments have been carried out exclusively through use of the Stark effect. Because charged molecules are accelerated in an electric field, observation of the Stark effect becomes impractical, and experimental determination of the electric dipole of an ion has not yet been made, although the dipole derivative of

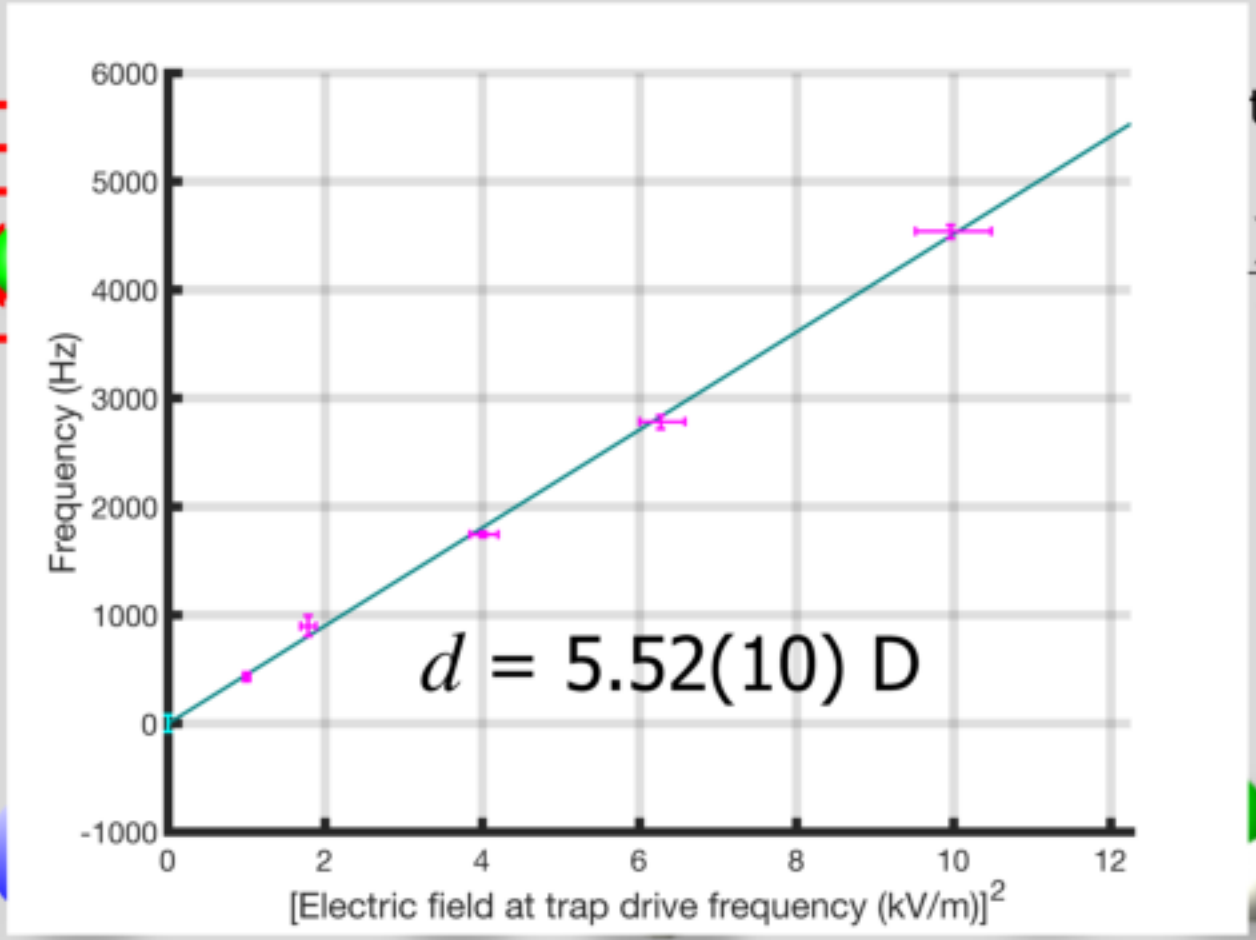
K. B. Laughlin *et al.*, Phil. Trans. R. Soc. Lond. A **234**, 109 (1988)

measured ArH^+ dipole moment by essentially backing it out of rotational magnetic moments for ArH^+ and ArD^+ isotopomeres

(3.0 $\pm$ 0.6) D

dipole moment measurement

static E- field



tic Stark effect

$$\frac{) - 3m_J}{-1)(2J+3)}$$

determine E_{rf} from Ca^+ mm-sideband/carrier Rabi frequency ratio

exchange Ca^+ and CaH^+ positions

measure CaH^+ $|0,-1/2,-\rangle \leftrightarrow |2,-3/2,-\rangle$ rotational transition frequency

entangle CaH^+ and Ca^+

Ca^+ CaH^+ motion

$$|S\rangle|-5/2\rangle|n=0\rangle$$

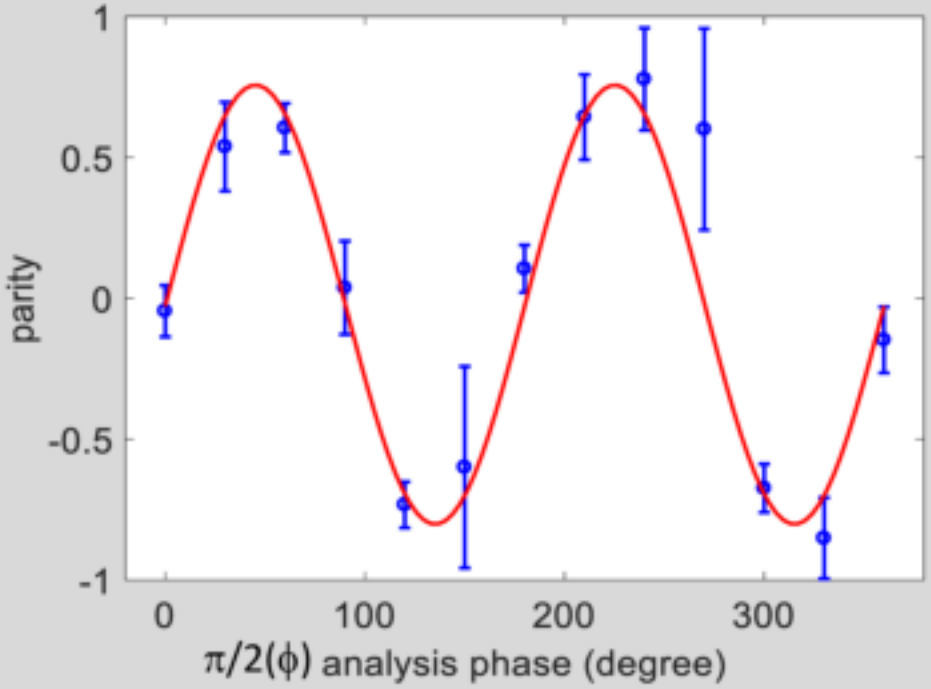
prepare system in pure state as before

$\pi/2$ CaH^+-motion blue sideband

$$1/\sqrt{2}|S\rangle(|-5/2\rangle|n=0\rangle+|-3/2\rangle|n=1\rangle)$$

π Ca^+-motion red sideband

$$1/\sqrt{2}(|S\rangle|-5/2\rangle+|D\rangle|-3/2\rangle)|n=0\rangle$$



electronic state of atom entangled with rotational state
fidelity F ≈ 0.87

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

summary

- quantum logic provides general methods to enable quantum coherent manipulation of charged particles
- can prepare and manipulate quantum states and detect state changes of single molecular ions with high fidelity
- suitable for singly charged polyatomic molecular ions with mass up to approximately 500 amu (q/m within factor 3).
- quantum control of spin-rotational transitions, entangle atom and molecule, hopefully soon: vibrational transitions
- special purpose quantum machine that runs "algorithms" to learn about real molecules, one (qu)bit at a time