Introduction to ultracold molecules

2. Internal state control and trapping molecules

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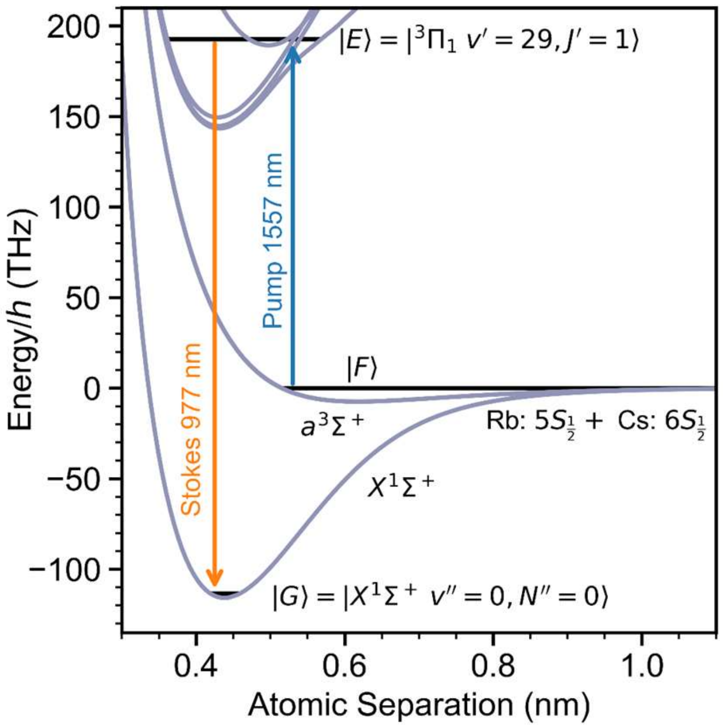
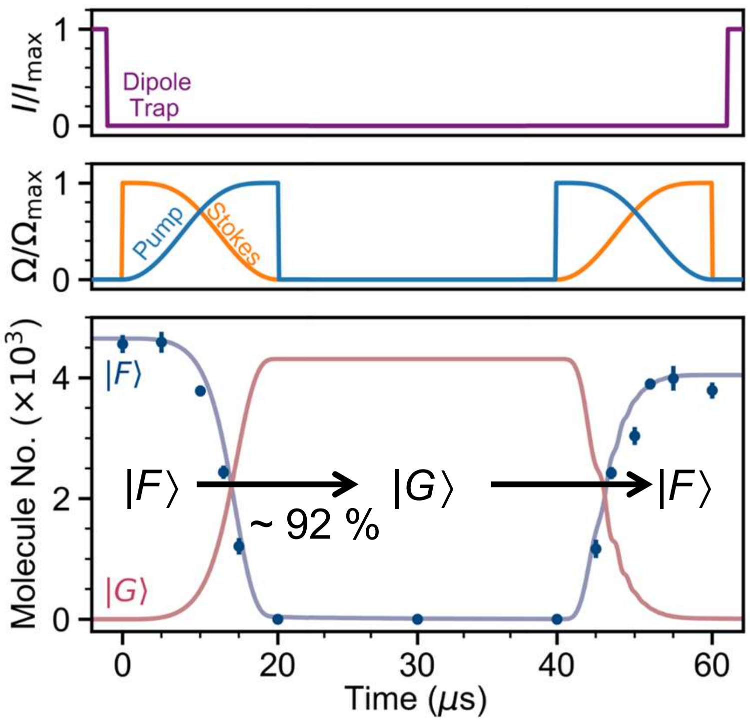
STIRAP to ground state



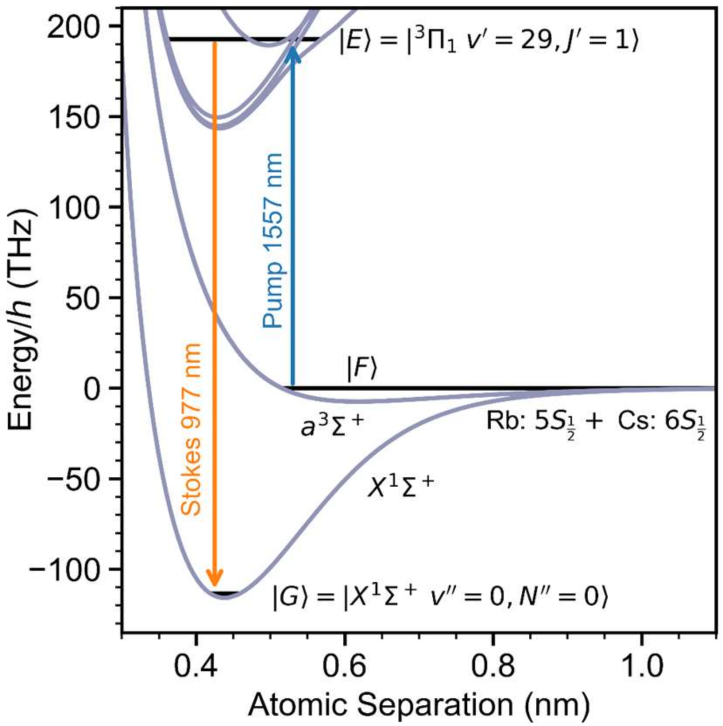


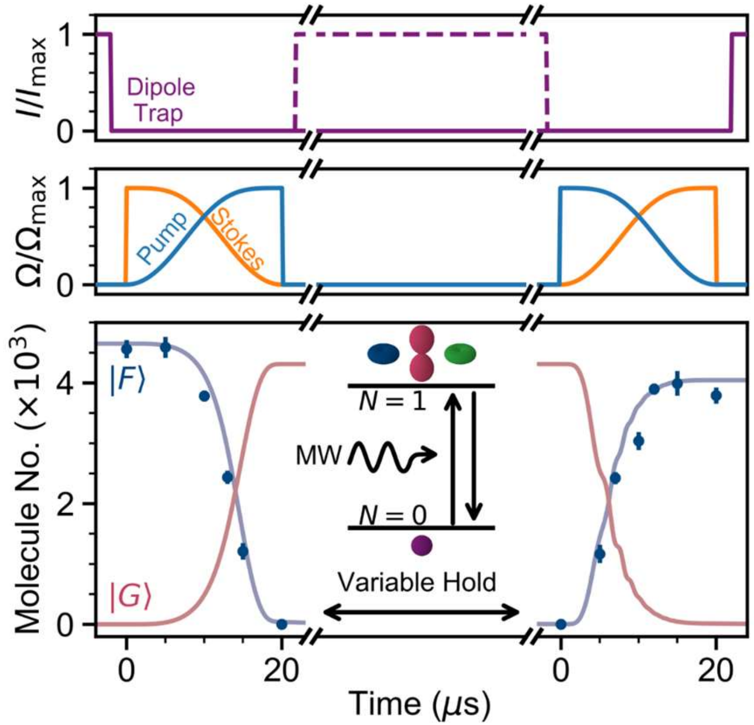
Final Molecule Sample:

N = 4000, T = 1.5 μK

$n_{pk} \approx 2 \times 10^{11}$ cm^{-3}

STIRAP to ground state



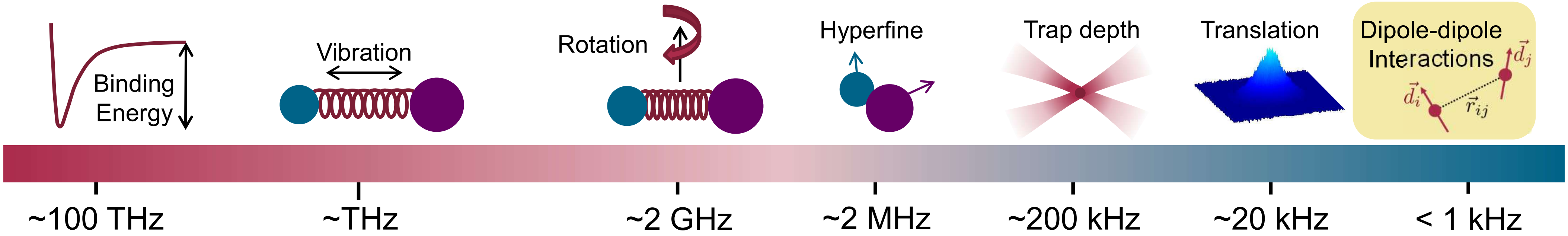


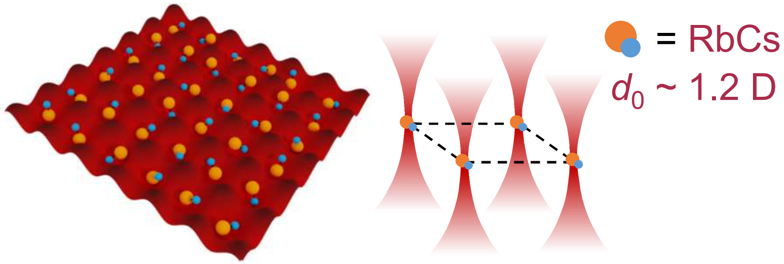
Final Molecule Sample:

N = 4000, T = 1.5 μK

$n_{pk} \approx 2 \times 10^{11}\ cm^{-3}$

Rich internal structure



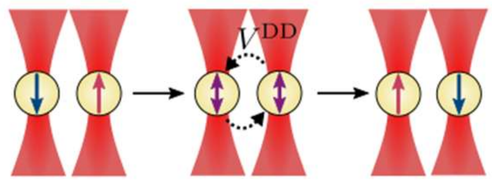


$$H_{XXZ} = \sum_{i \neq j} \left[\frac{J_\perp}{2} (S_i^+ S_j^- + S_i^- S_j^+) + J_z S_i^z S_j^z \right]$$

"Quantum Computation and Quantum Simulation with Ultracold Molecules"
Nature Physics **20**, 730-740 (2024)

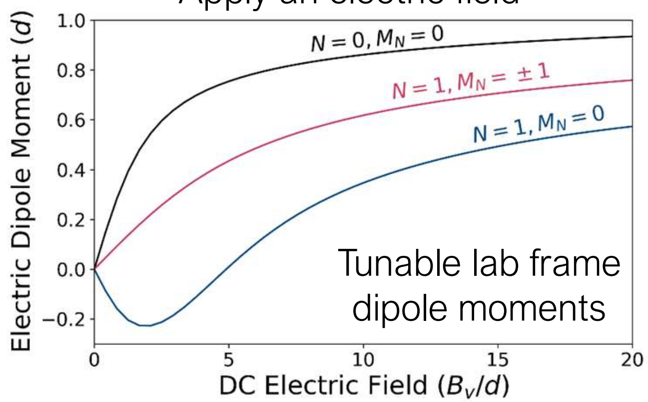
Controllable dipole-dipole interactions

Via transition dipole moment between rotational states



Spin-exchange interactions

Apply an electric field



Or mix states with microwave fields (coherent superpositions / dressing)

Challenge: quantum control of internal and external degrees of freedom of molecules

Controlling the internal state
Microwave spectroscopy,
Ramsey measurements,
Coherence and magic trapping

RbCs hyperfine structure

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The Hamiltonian: $$H = H_{\rm rot} + H_{\rm hf} + H_{\rm Z}$$

$$H_{\rm rot} = B_v \boldsymbol{N}^2 + D_v \boldsymbol{N}^2 \boldsymbol{N}^2$$

Rotation including centrifugal distortion

$$H_{\rm hf} = \sum_{i={\rm Rb,Cs}} \boldsymbol{V}_i \cdot \boldsymbol{Q}_i + \sum_{i={\rm Rb,Cs}} c_i \boldsymbol{N} \cdot \boldsymbol{I}_i + c_3 \boldsymbol{I}_{\rm Rb} \cdot \boldsymbol{T} \cdot \boldsymbol{I}_{\rm Cs} + c_4 \boldsymbol{I}_{\rm Rb} \cdot \boldsymbol{I}_{\rm Cs}$$

Nuclear electric quadrupole interaction

Interaction of nuclear magnetic moment with magnetic field due to rotation

Tensor & scalar interactions between nuclear dipole moments

$$H_{\rm Z} = -g_r \mu_N \boldsymbol{N} \cdot \boldsymbol{B} - \sum_{i={\rm Rb,Cs}} g_i \mu_N \boldsymbol{I}_i \cdot \boldsymbol{B}(1 - \sigma_i)$$

Rotational Zeeman interaction

Nuclear Zeeman interaction

Aldegunde et al., PRA **78**, 033434 (2008).

RbCs hyperfine structure: N=0

Durham University

The Hamiltonian: $$H = H_{\rm rot} + H_{\rm hf} + H_{\rm Z}$$

$$H_{\rm rot} = B_v \boldsymbol{N}^2 + D_v \boldsymbol{N}^2 \boldsymbol{N}^2$$

$$H_{\rm hf} = \sum_{i={\rm Rb,Cs}} \boldsymbol{V}_i \cdot \boldsymbol{Q}_i + \sum_{i={\rm Rb,Cs}} c_i \boldsymbol{N} \cdot \boldsymbol{I}_i + c_3 \boldsymbol{I}_{\rm Rb} \cdot \boldsymbol{T} \cdot \boldsymbol{I}_{\rm Cs} + c_4 \boldsymbol{I}_{\rm Rb} \cdot \boldsymbol{I}_{\rm Cs}$$

$$H_{\rm Z} = -g_r \mu_N \boldsymbol{N} \cdot \boldsymbol{B} - \sum_{i={\rm Rb,Cs}} g_i \mu_N \boldsymbol{I}_i \cdot \boldsymbol{B}(1-\sigma_i)$$

Aldegunde et al., PRA **78**, 033434 (2008).

RbCs hyperfine structure: N=0

$$\boldsymbol{I}_{\mathrm{Rb}} = \frac{3}{2}, \boldsymbol{I}_{\mathrm{Cs}} = \frac{7}{2} \rightarrow (2\boldsymbol{I}_{\mathrm{Rb}}+1)\times(2\boldsymbol{I}_{\mathrm{Cs}}+1) = 32 \text{ states}$$

Zero Field:

$\boldsymbol{I} = 5$

$5c_4$

$\boldsymbol{I} = 4$

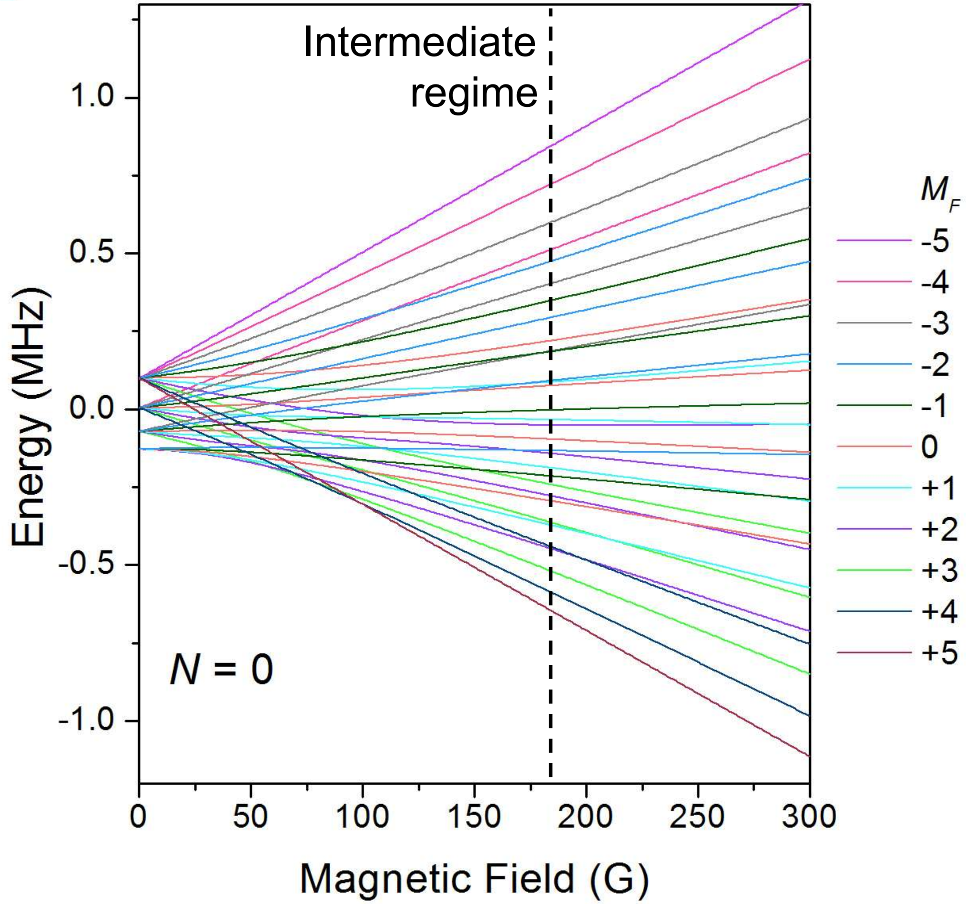
$4c_4$

$\boldsymbol{I} = 3$

$3c_4$

$\boldsymbol{I} = 2$

$c_4 \sim 19$ kHz



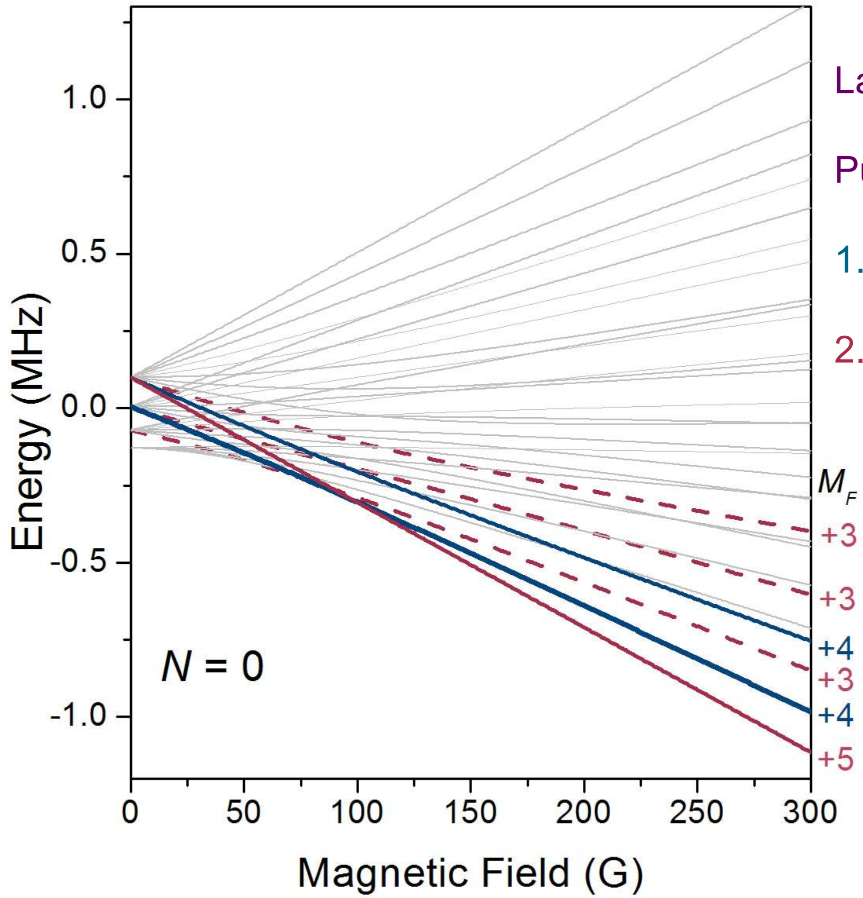
High Field:

Nuclear Zeeman effect dominates

$$E \simeq -(g_I^{\mathrm{Rb}} m_I^{\mathrm{Rb}} + g_I^{\mathrm{Cs}} m_I^{\mathrm{Cs}})\mu_{\mathrm{N}} B$$

Coupling to the Feshbach state

Feshbach states has $M_{\mathrm{TOT}} = +4$ Selection rules limit accessible states:

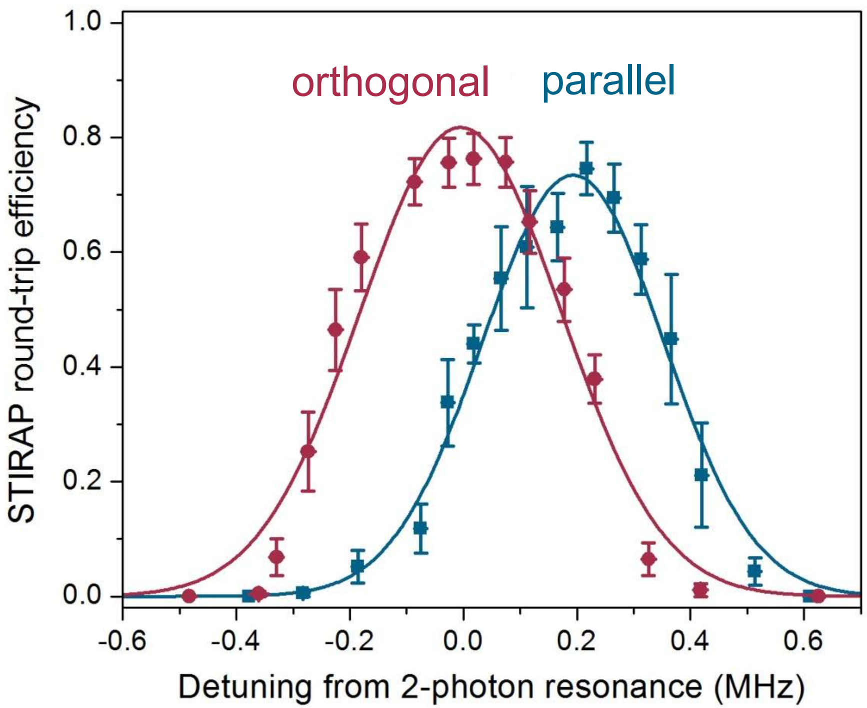


Laser polarisations:

Pump parallel to B and

1. Stokes parallel to B
   → address $M_{\mathrm{F}} = +4$
2. Stokes orthogonal to B
   → address $M_{\mathrm{F}} = +3, +5$

Selection rules save the day…



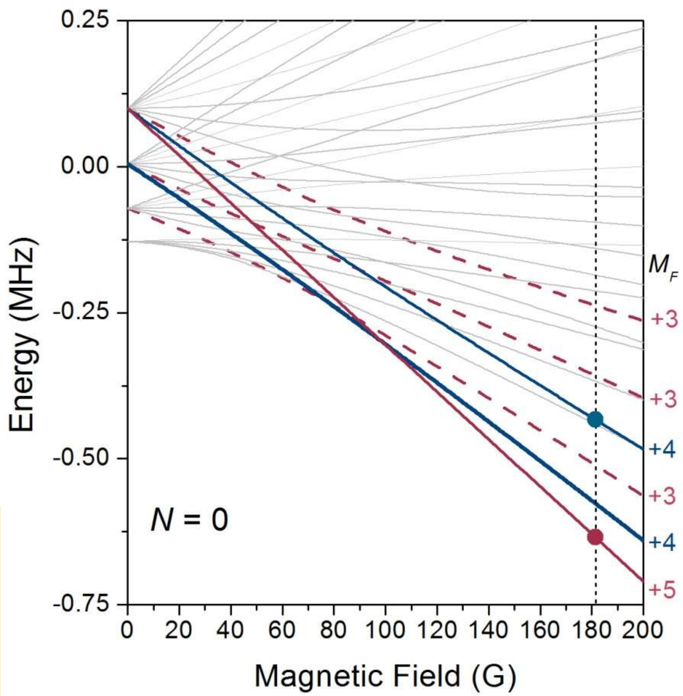
Crucially can transfer to the lowest energy state:

$$|N, m_N, m_I^{\mathrm{Rb}}, m_I^{\mathrm{Cs}}\rangle = |0, 0, +3/2, +7/2\rangle$$

(fully spin-stretched)

Feshbach states has $M_{\mathrm{TOT}} = +4$

From 200 kHz splitting identify two hyperfine states:



Hyperfine structure!

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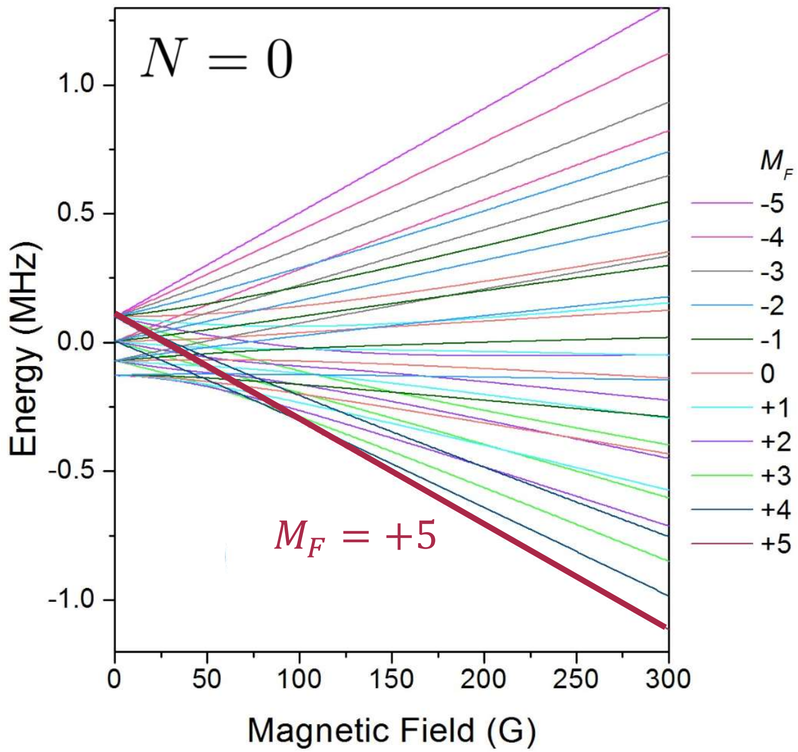
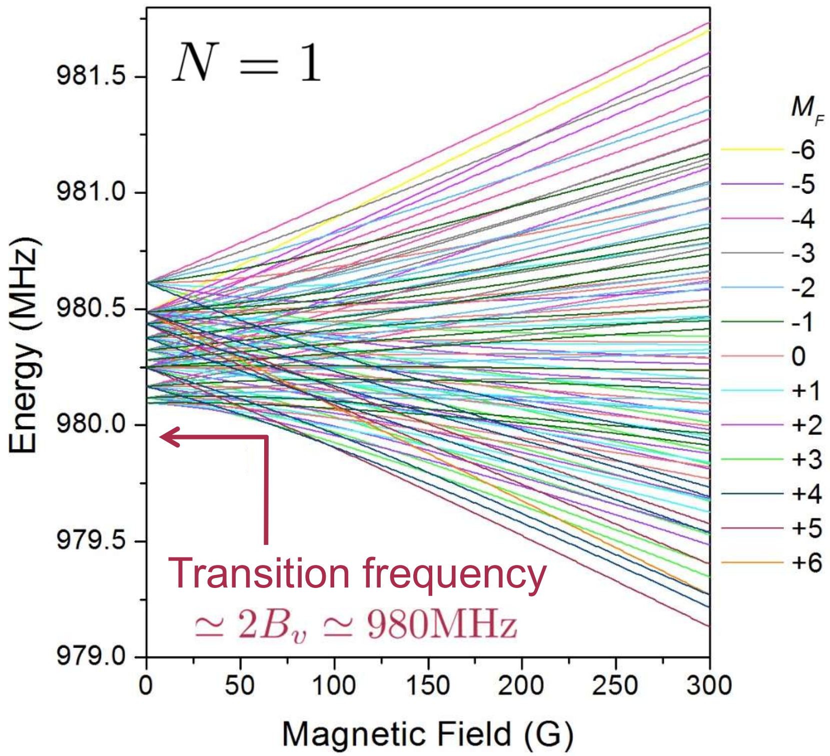
$$\boldsymbol{I}_{\mathrm{Rb}} = \frac{3}{2}, \boldsymbol{I}_{\mathrm{Cs}} = \frac{7}{2}$$

$$\rightarrow (2\boldsymbol{I}_{\mathrm{Rb}}+1)(2\boldsymbol{I}_{\mathrm{Cs}}+1) = 32 \text{ states}$$



$$(2\boldsymbol{N}+1)(2\boldsymbol{I}_{\mathrm{Rb}}+1)(2\boldsymbol{I}_{\mathrm{Cs}}+1) = 96 \text{ states}$$



$$|N, m_N, m_I^{\mathrm{Rb}}, m_I^{\mathrm{Cs}}\rangle = |0, 0, +3/2, +7/2\rangle$$

Aside – comparing molecules

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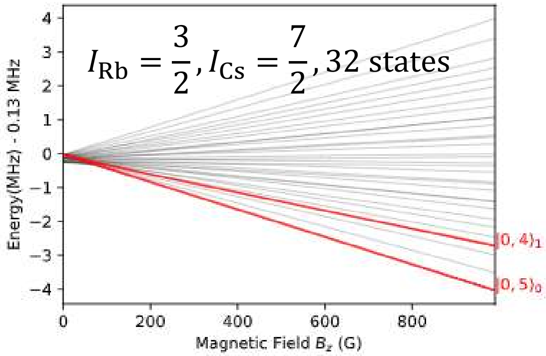
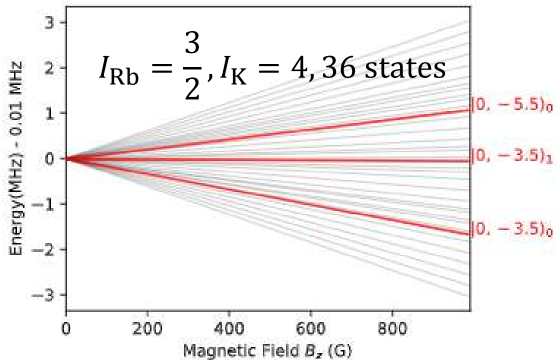
RbCs



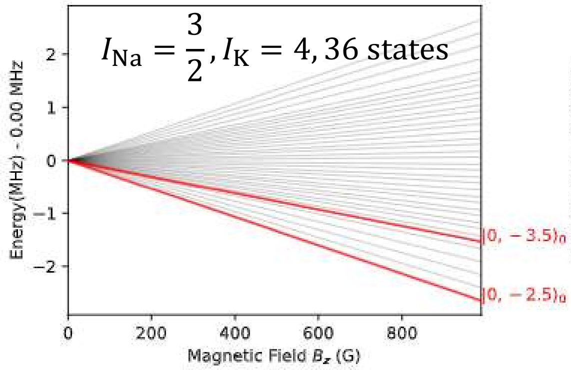
$c_4 = 17\mathrm{kHz}, g_{\mathrm{Rb}} = 1.83, g_{\mathrm{Cs}} = 0.73$

^{40}K^{87}Rb



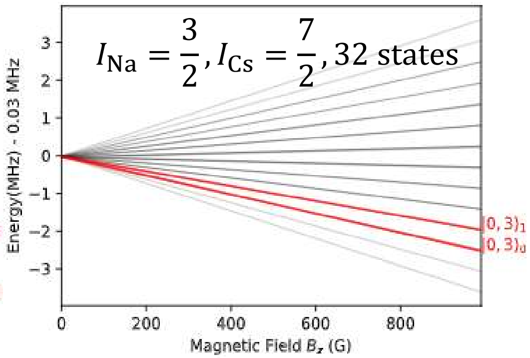
$c_4 = 0.9\mathrm{kHz}, g_{\mathrm{K}} = -0.32, g_{\mathrm{Rb}} = 1.83$

Na^{40}K



$c_4 = -0.45\mathrm{kHz}, g_{\mathrm{Na}} = 1.48, g_{\mathrm{K}} = -0.32$

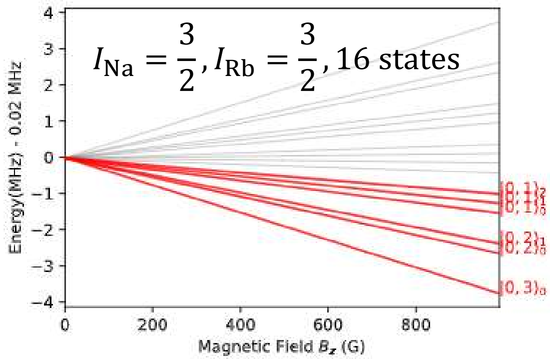
NaCs



$c_4 = 3.9\mathrm{kHz}, g_{\mathrm{Na}} = 1.48, g_{\mathrm{Cs}} = 0.74$

x 2

NaRb



$c_4 = 5.7\mathrm{kHz}, g_{\mathrm{Na}} = 1.48, g_{\mathrm{Rb}} = 1.83$

Tom Hepworth

DU 4th year student + Diatomic-Py

Constants from:

Aldegunde & Hutson, PRA **96**, 042506 (2017)

Microwave spectroscopy

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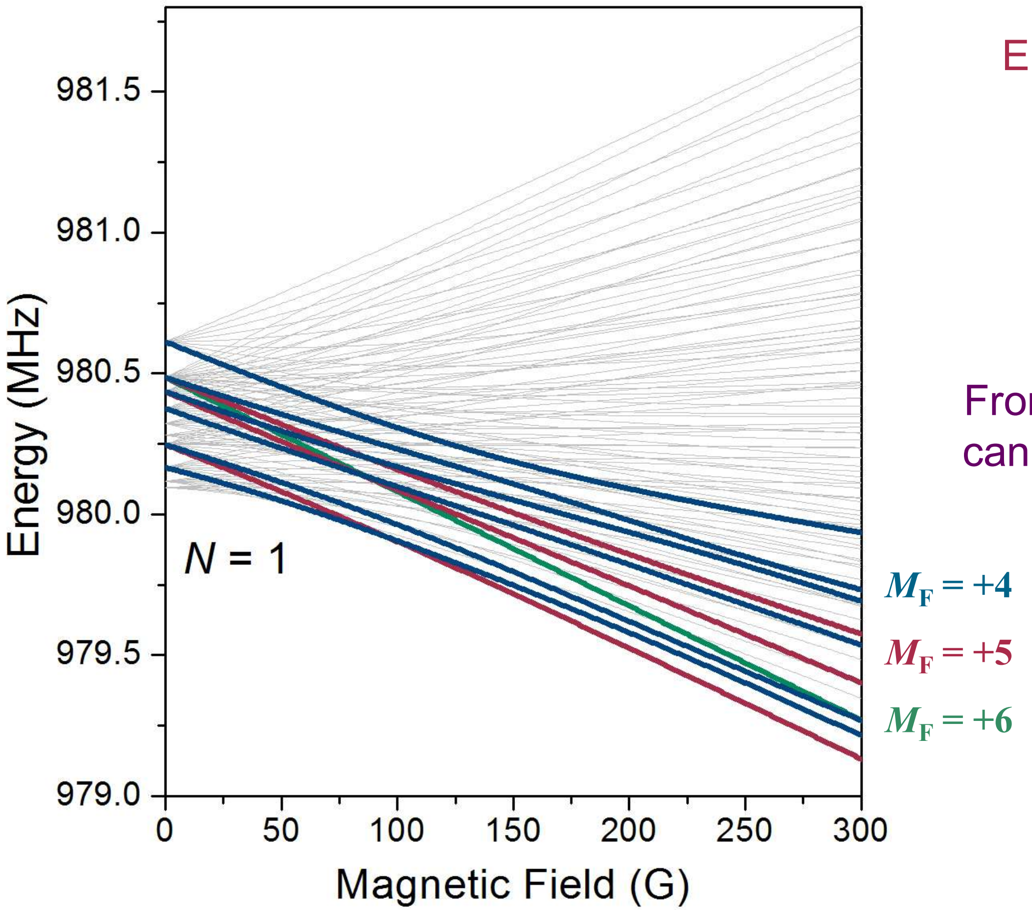
Only certain states are accessible:



Electric dipole selection rules:

$$\Delta M_N = 0, \pm 1$$

$$\Delta m_I^{\mathrm{Rb}} = \Delta m_I^{\mathrm{Cs}} = 0$$

$$\Delta M_F = 0, \pm 1$$

From the $M_F = +5$ ground state we can drive microwave transitions to $M_F = +4, +5$, or $+6$

Expect 10 lines

Rabi oscillations

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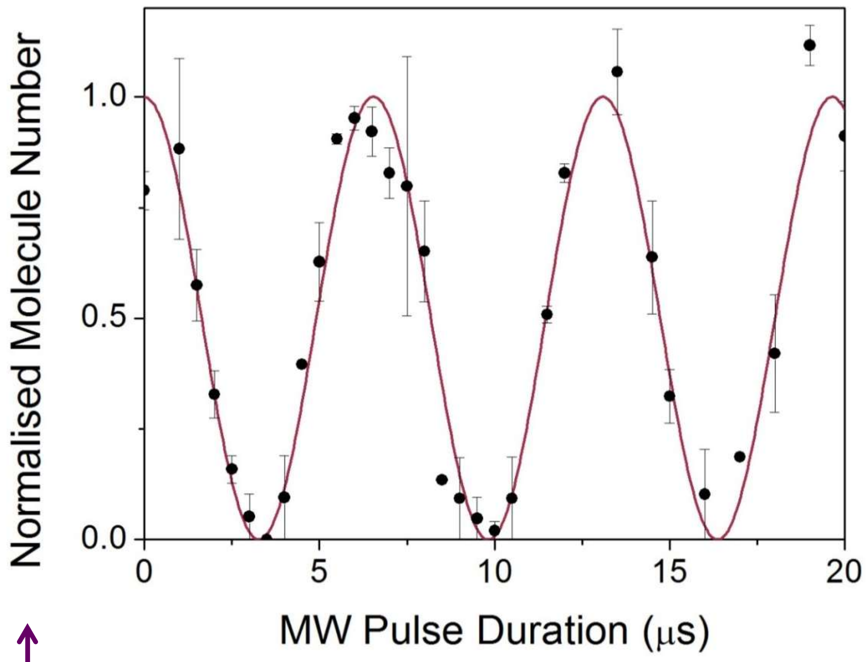
Permanent electric-dipole moment (~1.2 D) allows strong transitions to $N = 1$

➔ Very easy to drive fast Rabi oscillations!



Always measure number in absolute hyperfine & rotational ground state by return STIRAP

For spectroscopy:

Iteratively adjust duration & power to give an approximate π-pulse

➔ Loss feature vs. frequency

Spectroscopy in free space

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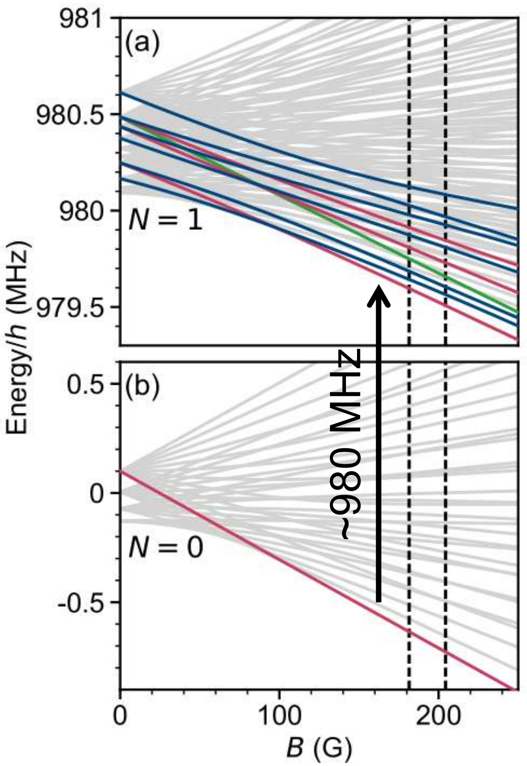
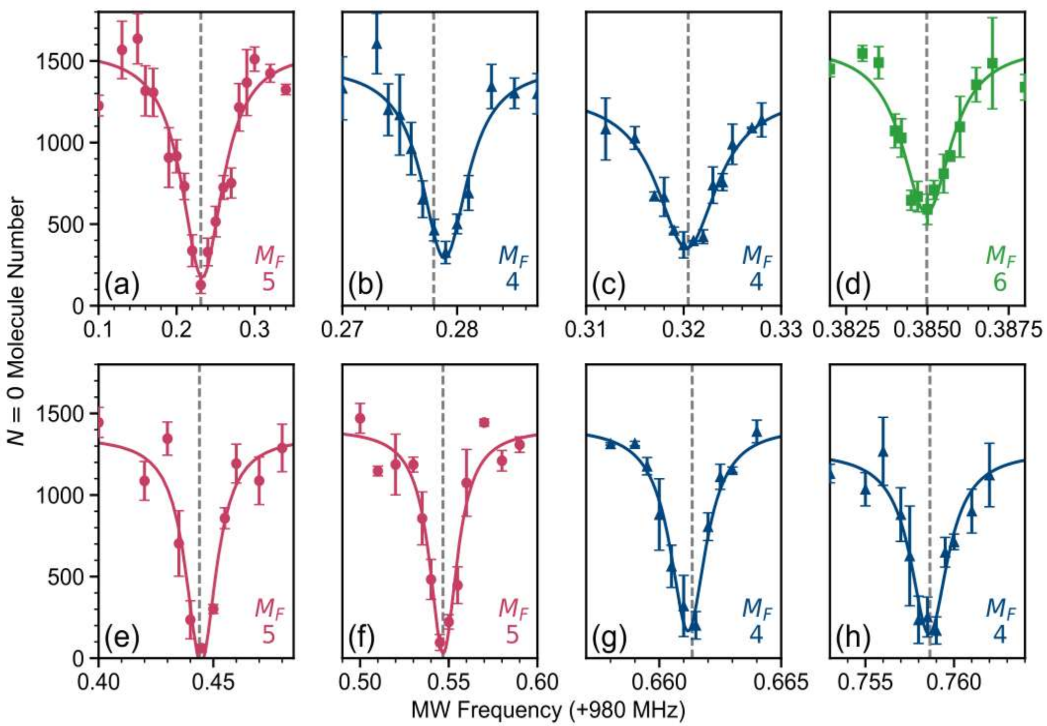


Use microwave pi pulse for spectroscopy



Widths of features are Fourier-transform limited.

Electric dipole selection rules:

$$\Delta M_N = 0, \pm 1, \ \Delta m_I = 0$$

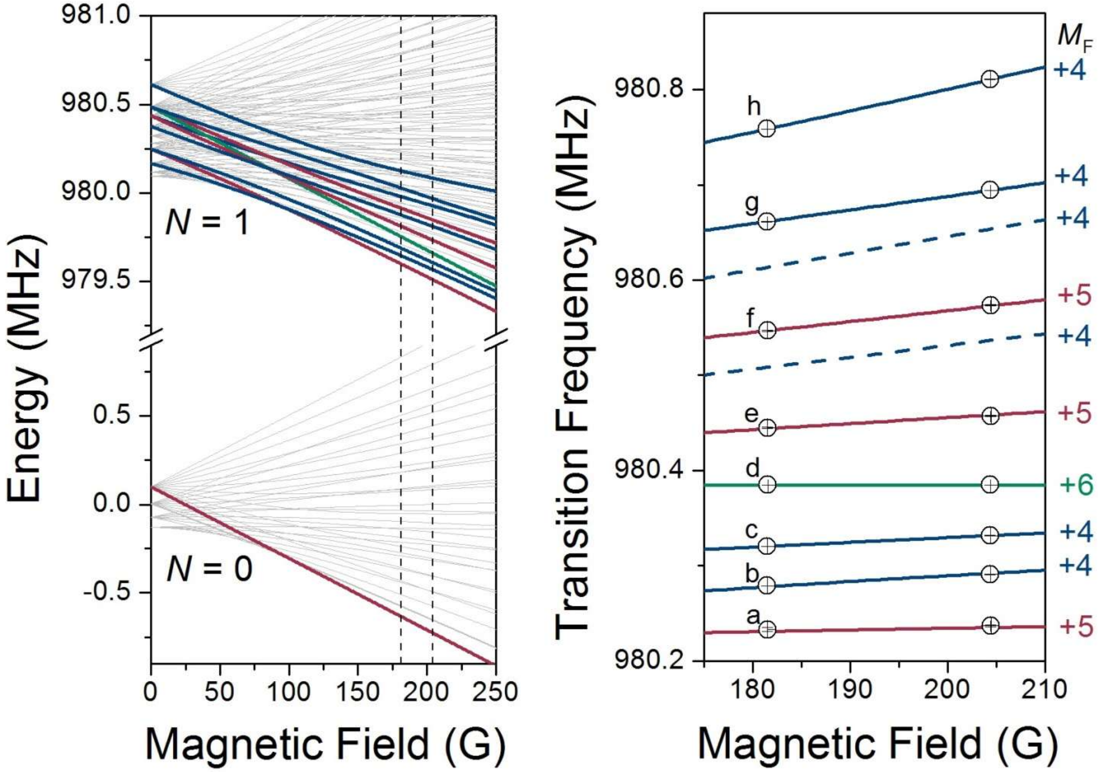
$$\Delta M_F = 0, \pm 1$$

P. D. Gregory *et al.*
PRA **94**, 041403(R) (2016)

Microwave spectroscopy

Used two magnetic fields. Fit hyperfine Hamiltonian to data to determine parameters.

Extract state composition in uncoupled basis to identify fraction that couples to the ground state:



$$B_v = 490.173\ 99(5)\ \text{MHz}$$
$$(eQq)_{\text{Rb}} = -809(1)\ \text{kHz}$$
$$(eQq)_{\text{Cs}} = 59(2)\ \text{kHz}$$
$$c_4 = 19.0(1)\ \text{kHz}$$

Microwave spectroscopy

Durham University

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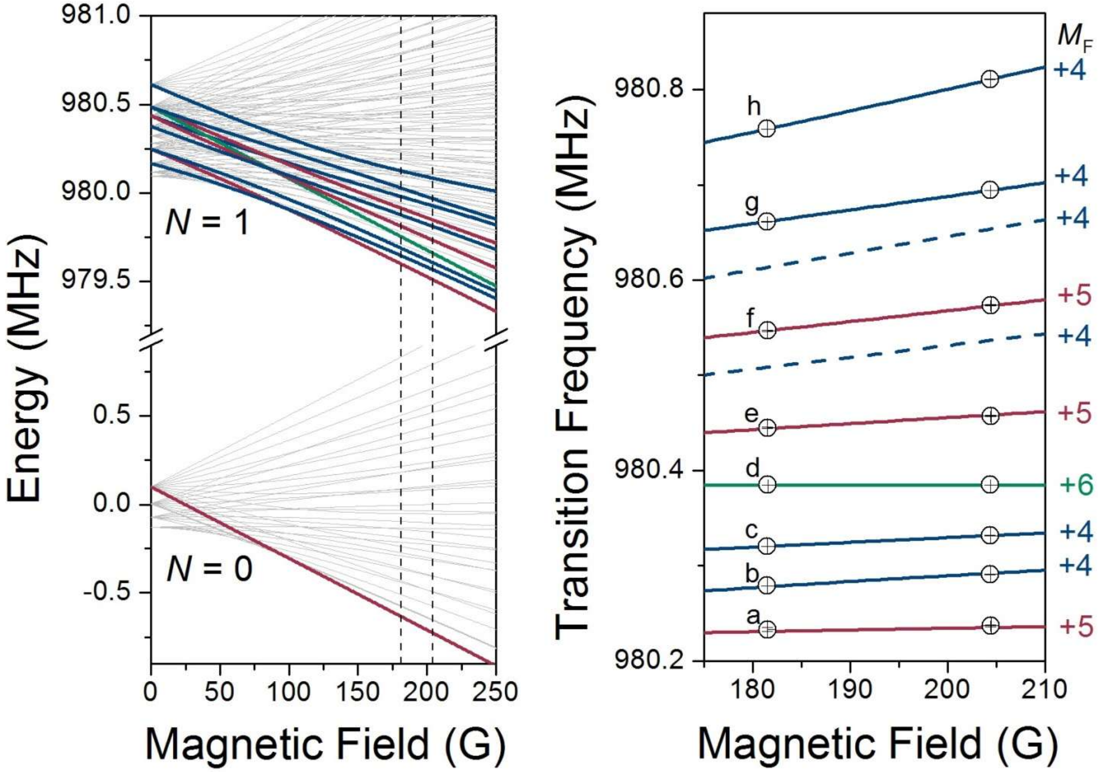


$0.382\,|1,-1,+3/2,+7/2\rangle + \ldots$

$0.208\,|1,-1,+3/2,+7/2\rangle + \ldots$

$0.017\,|1,-1,+3/2,+7/2\rangle + \ldots$

$0.304\,|1,0,+3/2,+7/2\rangle + \ldots$

$0.015\,|1,-1,+3/2,+7/2\rangle + \ldots$

$0.229\,|1,0,+3/2,+7/2\rangle + \ldots$

$|1,+1,+3/2,+7/2\rangle$

$0.687\,|1,-1,+3/2,+7/2\rangle + \ldots$

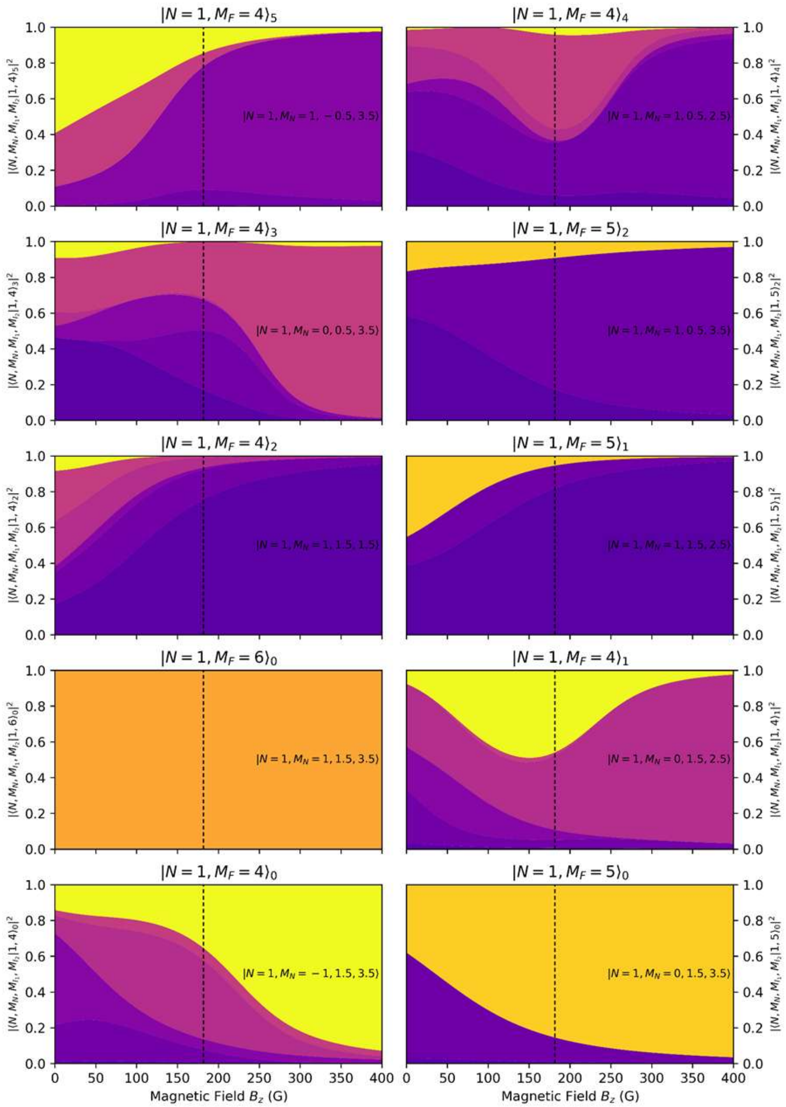
$0.582\,|1,-1,+3/2,+7/2\rangle + \ldots$

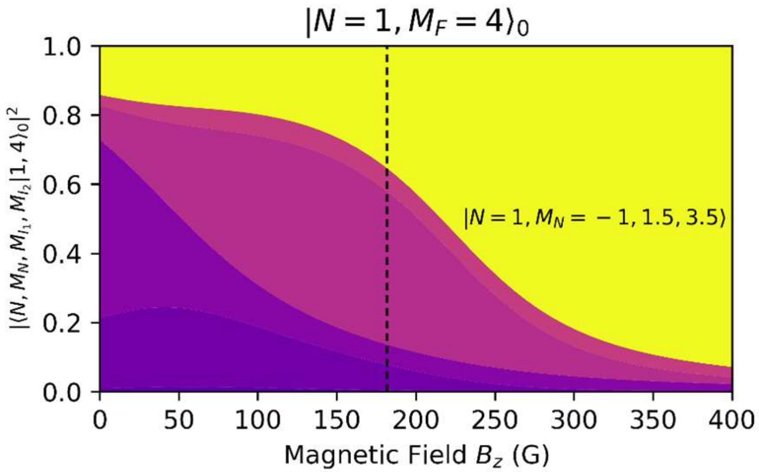
$0.925\,|1,0,+3/2,+7/2\rangle + \ldots$

Square for relative transition strength

Ground state: $|N, m_N, m_I^{\mathrm{Rb}}, m_I^{\mathrm{Cs}}\rangle = |0,0,+3/2,+7/2\rangle$

State composition





Different colours show different components in uncoupled basis

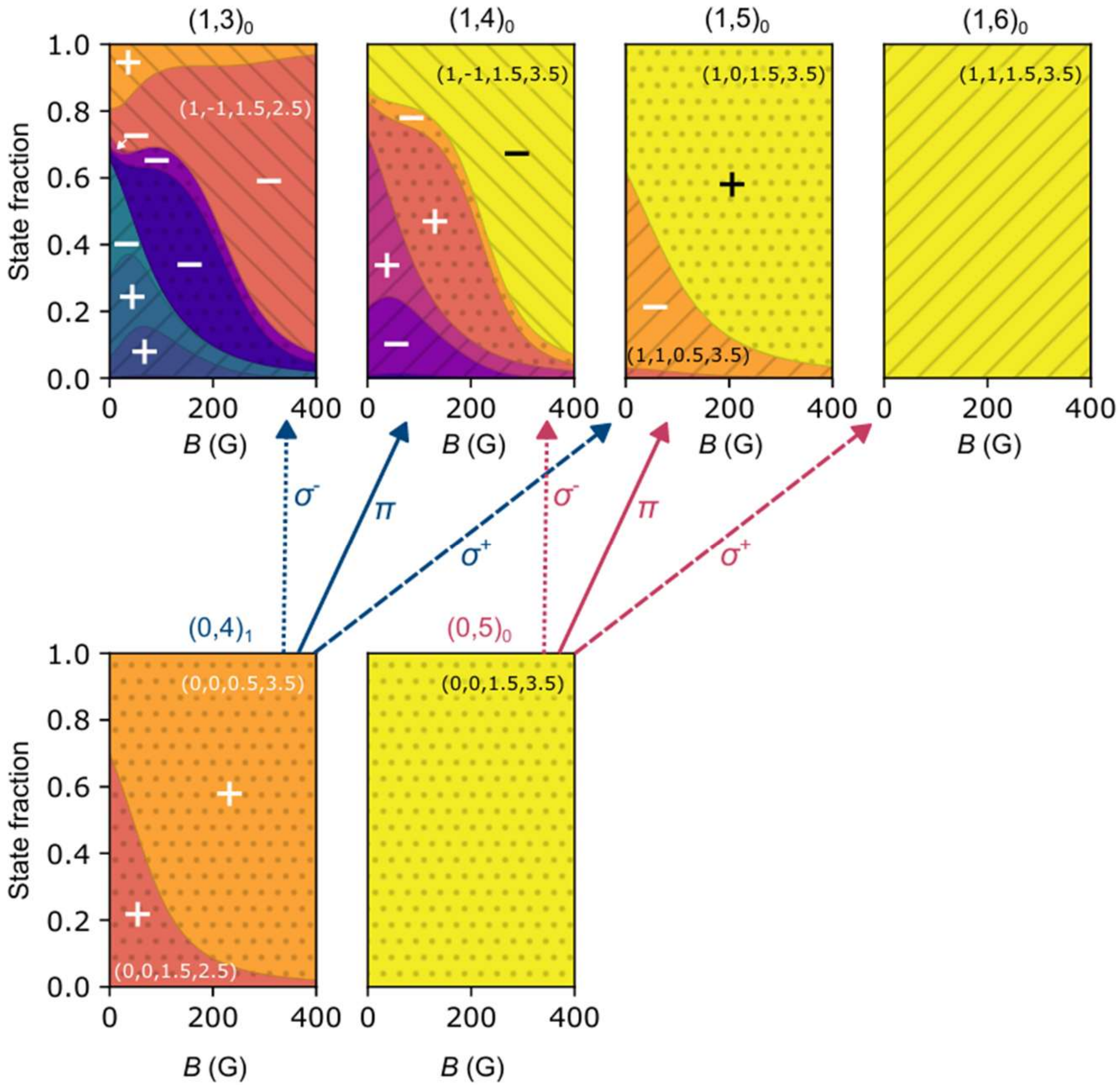
Coupling is to yellow/gold component

Spin stretched state is obvious

At high field see coupling to just three components (as expected) M_N = 0, ±1

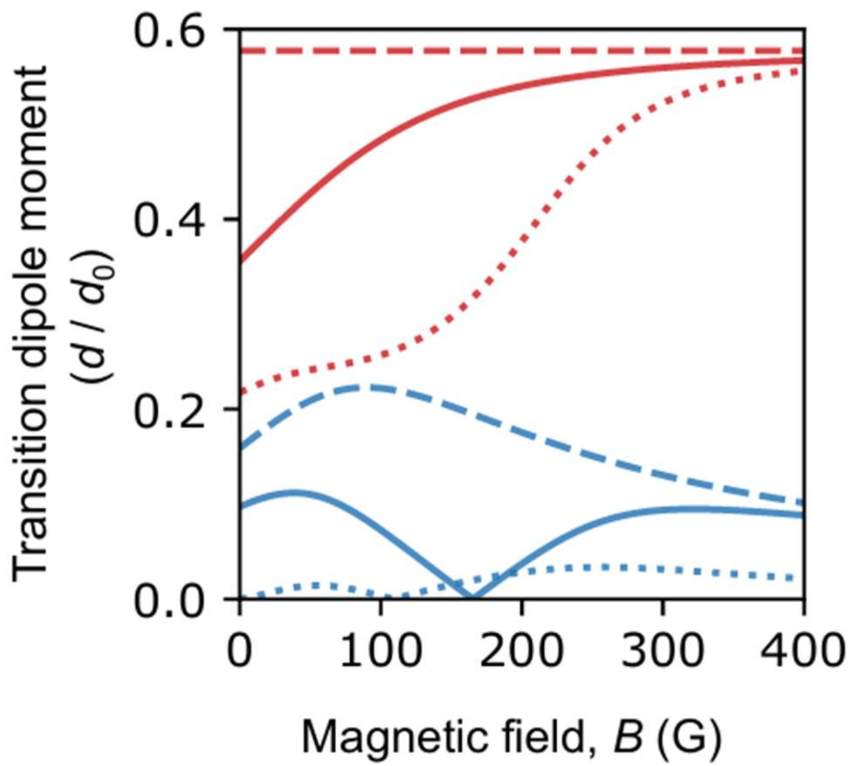
Note – here the magnetic field and choice of state tunes to transition dipole moment = DDI !

Controlling transition strengths



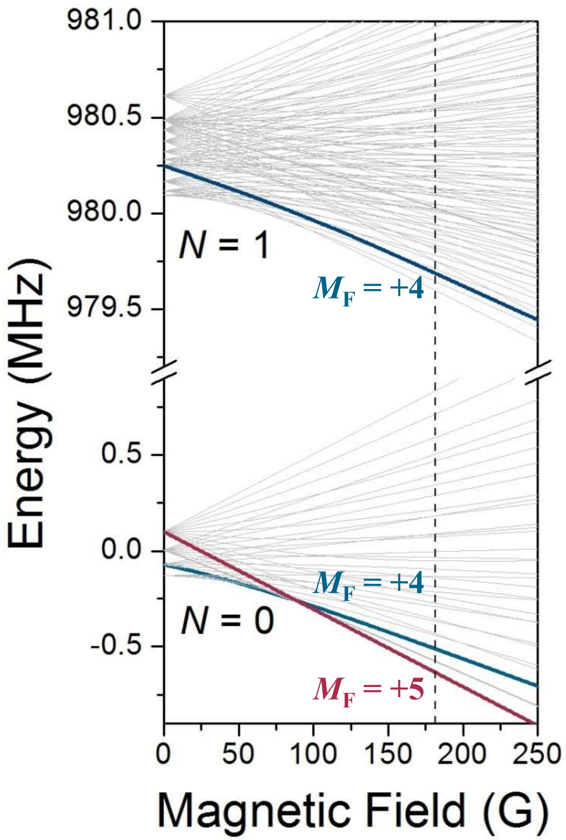
From Hepworth et al., arXiv:2511.03324

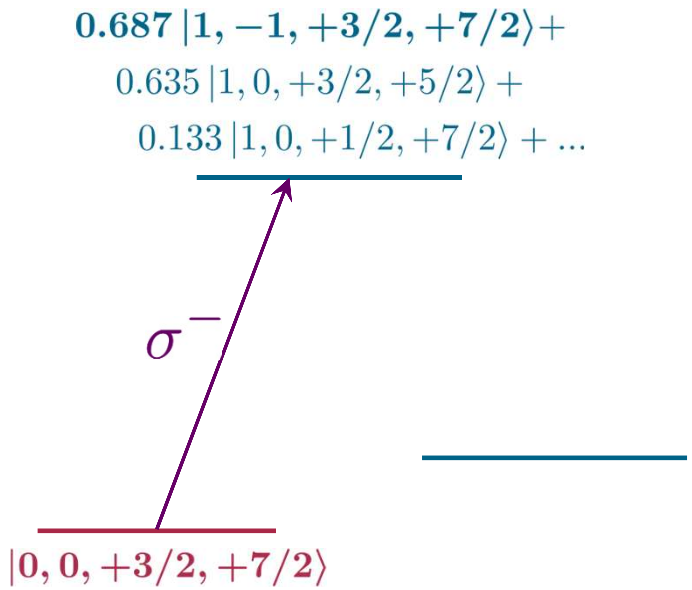
Transitions connect like-colours



Coherent hyperfine state transfer

- Nuclear electric quadrupole coupling in N = 1 strongly mixes the basis functions.
- Can be used to coherently manipulate hyperfine states within N = 0.





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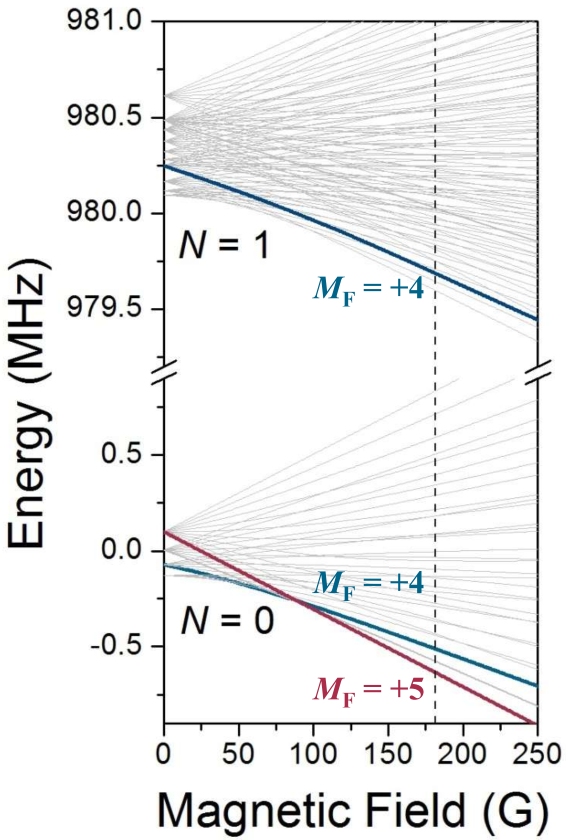

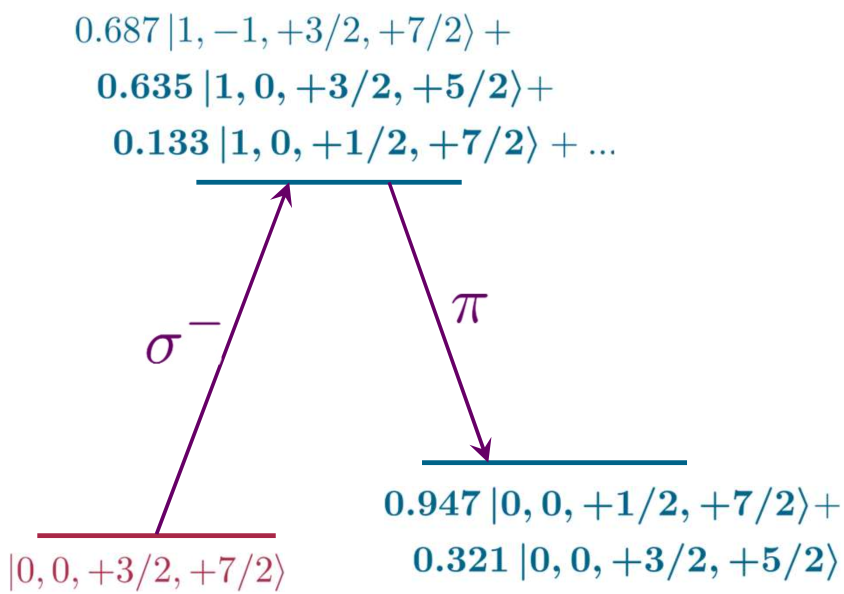

c.f. KRb: *Ospelkaus et al*., PRL **104**, 030402 (2010)
NaK: *Will et al*., PRL **116**, 225306 (2016)

Coherent hyperfine state transfer

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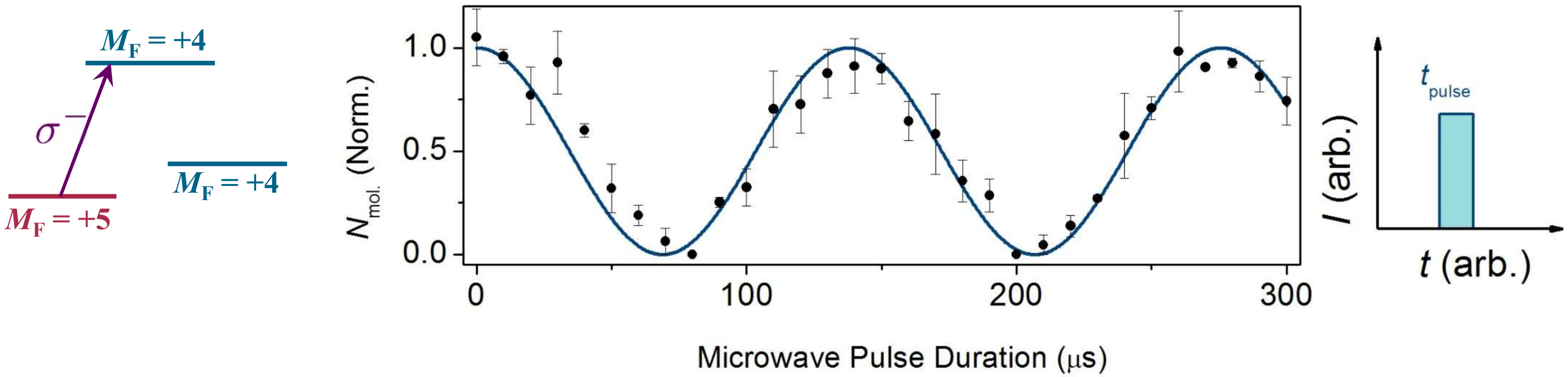

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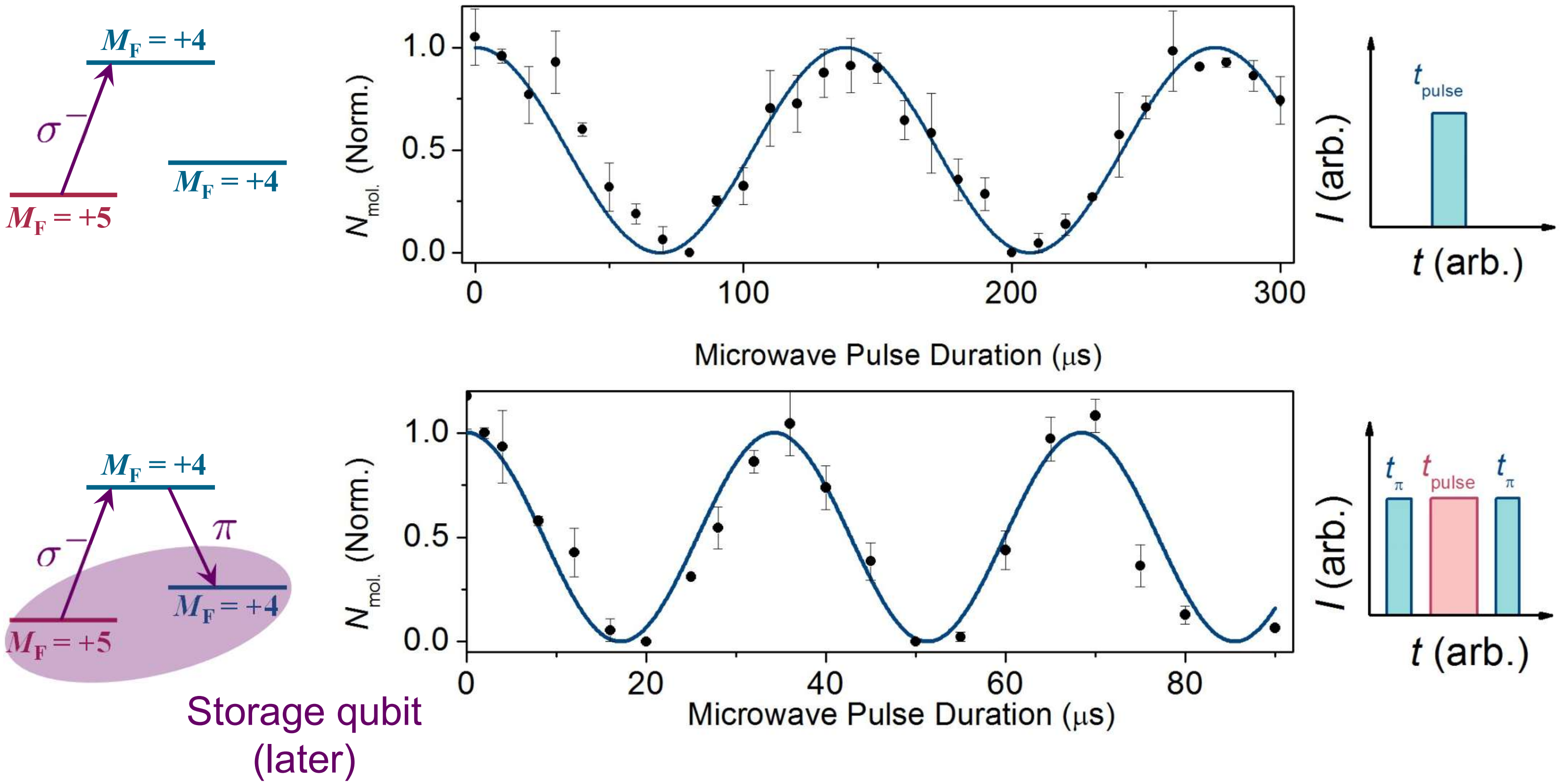
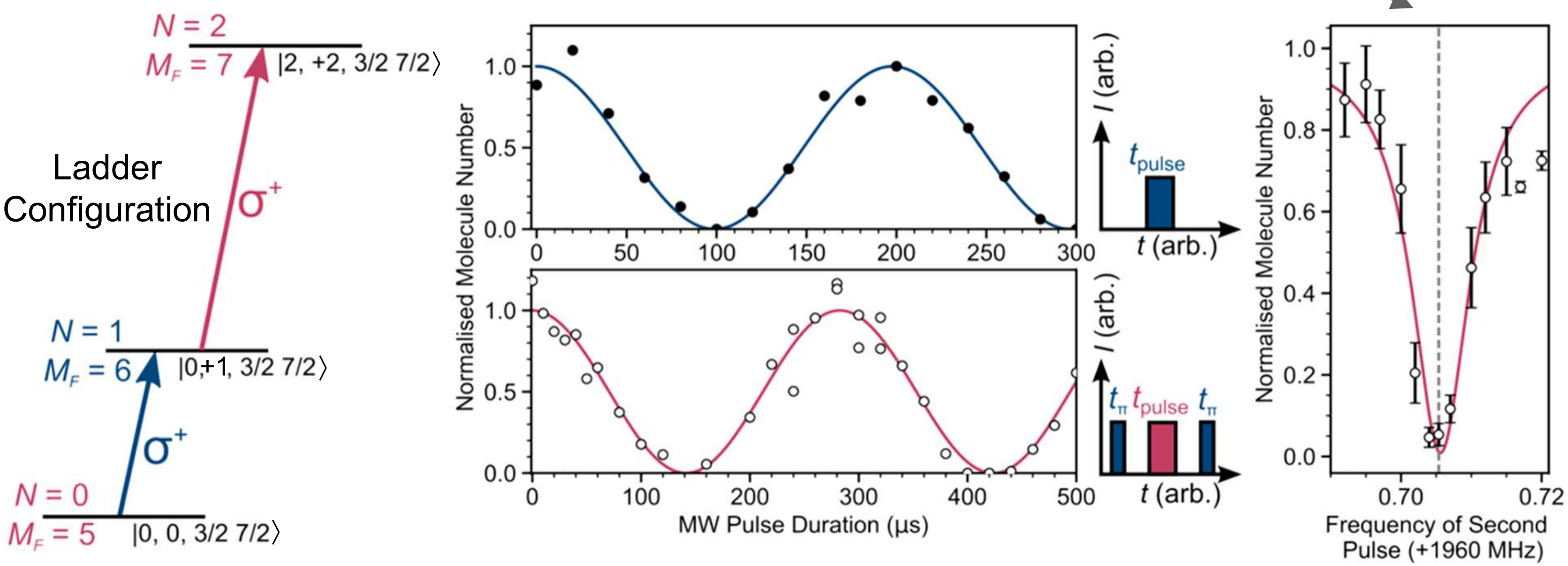


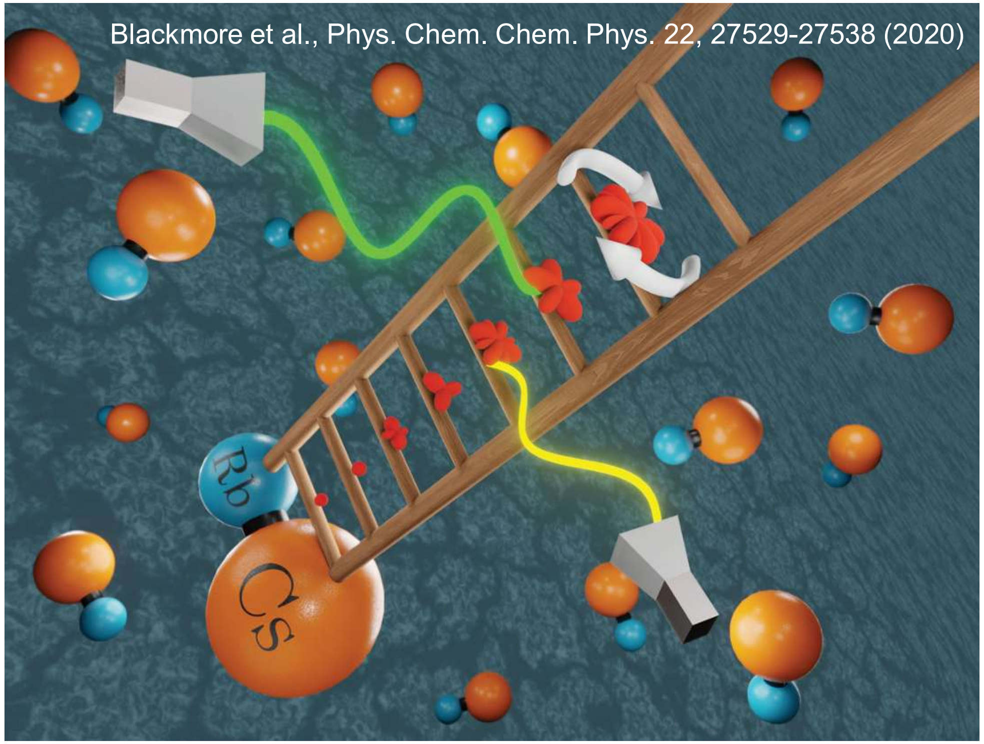
Coherent Microwave State Control

Prediction from spectroscopy

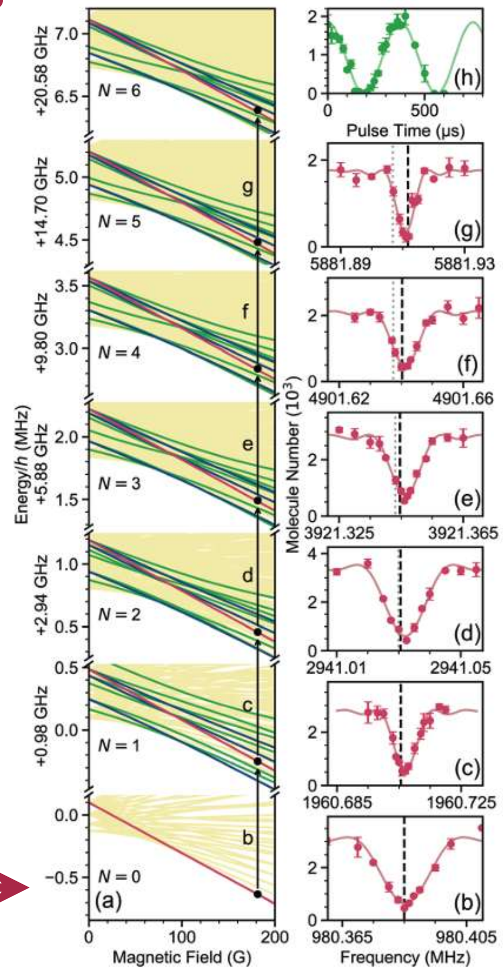


Exploiting the richness of rotation

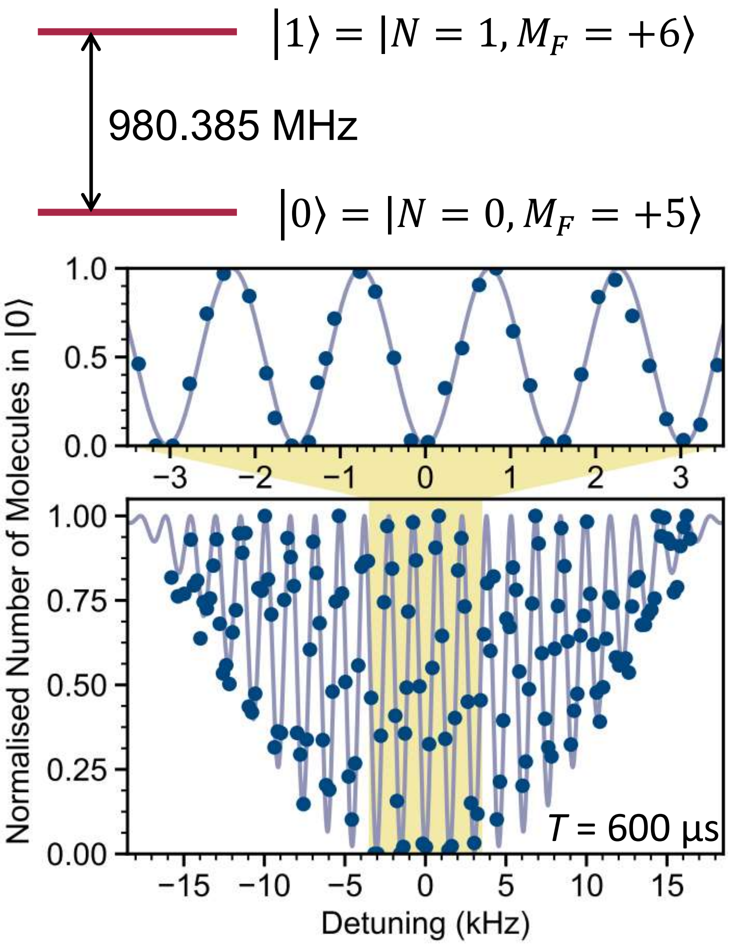
Many opportunities to use rotational states to engineer synthetic dimensions!



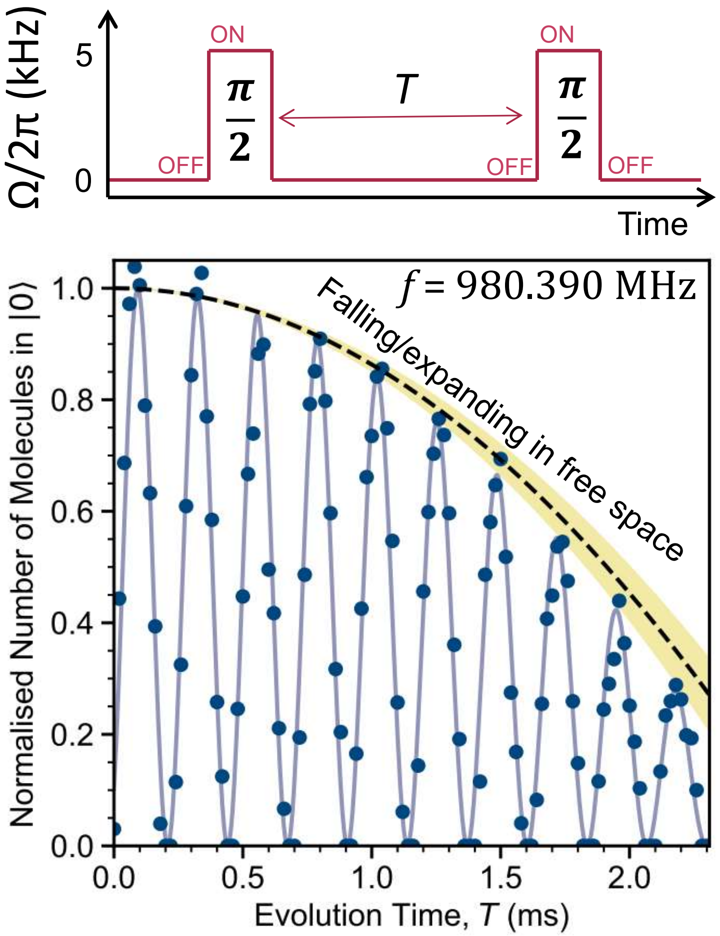
Already identified transitions to $N = 6$ →



Probing coherence with Ramsey spectroscopy



Centre frequency = 980,385,569(8) Hz

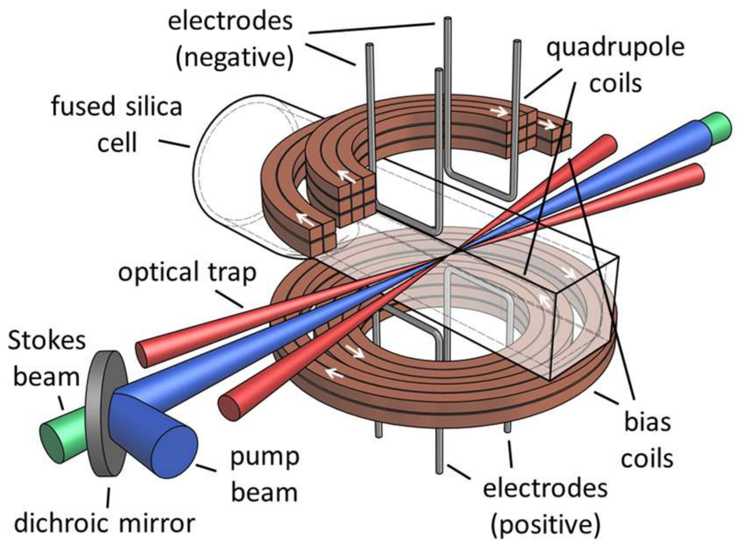


Blackmore *et al*. QST **4**, 014010 (2019)

Trapping molecules

$^1\Sigma$ molecules $\rightarrow$ no electronic magnetic moment = cannot trap magnetically

Simply turn on trap used to prepare atomic mixture



Trap potential:

$$H_{\text{AC Stark}} = \frac{1}{2\epsilon_0 c}\alpha I$$

$\alpha =$ Polarizability tensor

Reminder – dynamic polarizability calculated from sum over allowed transitions

$$\alpha_0(\omega) = \frac{2}{3(2j+1)}\sum_{j'}\frac{(E-E')|\langle j||D||j'\rangle|^2}{\omega^2-(E-E')^2}$$

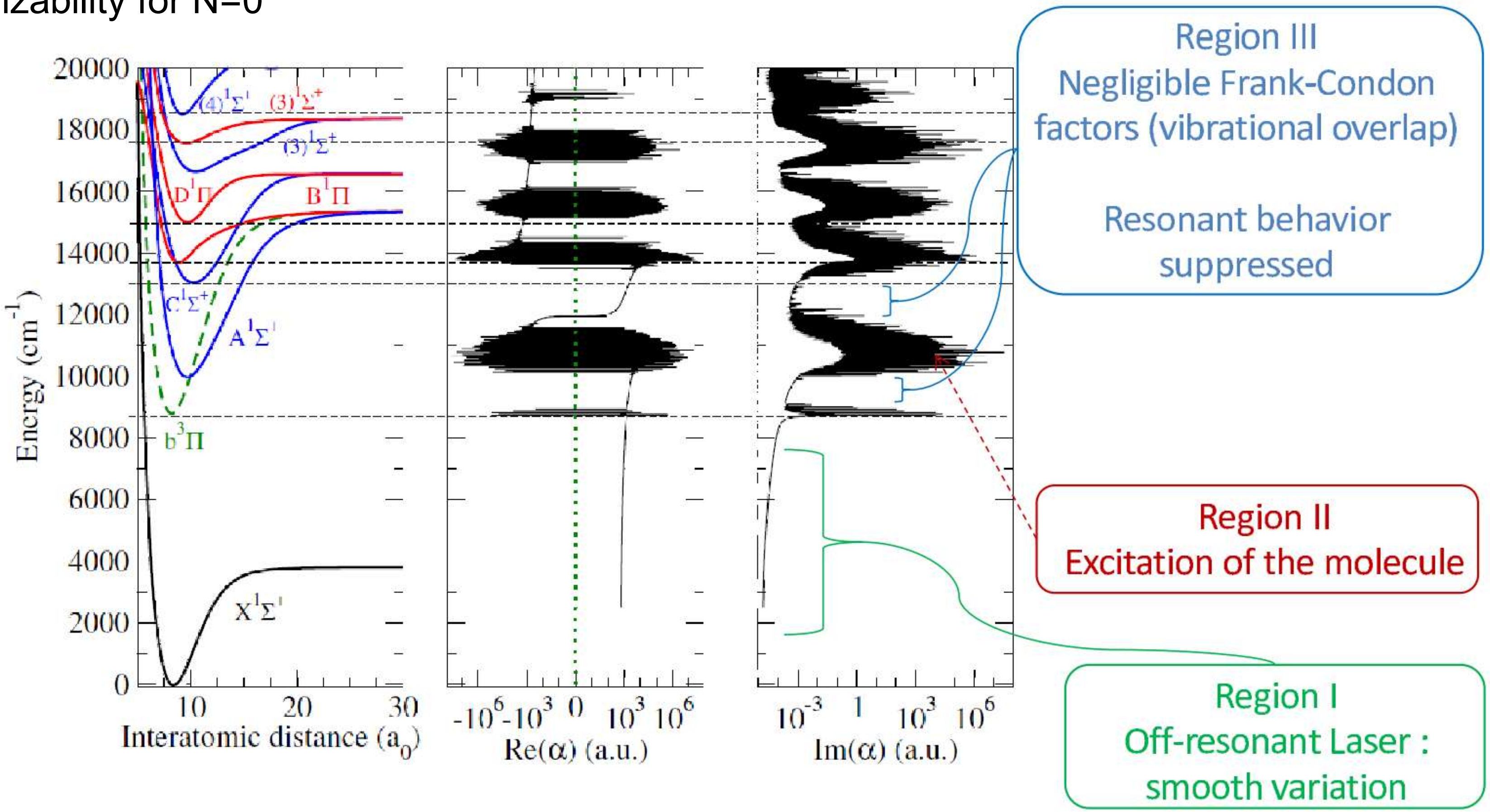
Mitroy *et al.*, *J. Phys. B: At. Mol. Opt. Phys.* **43**, 202001 (2010)

Portal for High-Precision Atomic Data and Computation (University of Delaware)

For red-detuned trapping of ground-state alkalis only need to consider D1 and D2 transition

A little more complicated for molecules…

RbCs polarizability for N=0

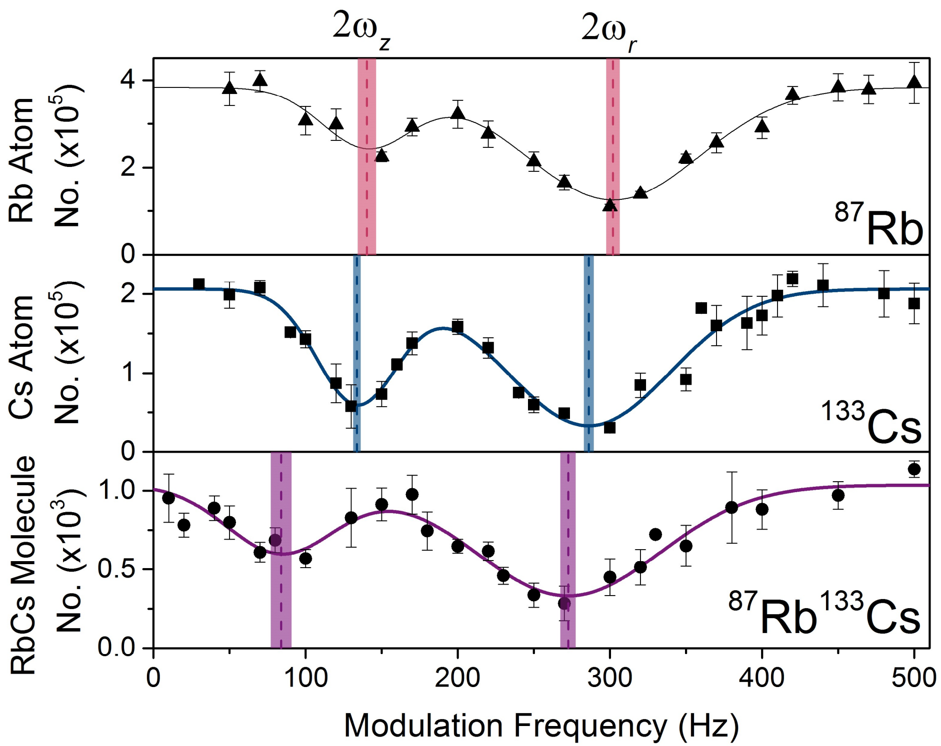


Vexiau *et al*., International Reviews in Physical Chemistry **36**, 709-750 (2017)
arXiv:1707.02168

Measure $N = 0$ polarizability

Durham University

Measure using parametric heating:



Scale known polarizability of Cs:

$$\alpha_{\mathrm{RbCs}} = \left(\frac{\omega_{\mathrm{RbCs}}}{\omega_{\mathrm{Cs}}}\right)^2 \left(\frac{\alpha_{\mathrm{Cs}} m_{\mathrm{RbCs}}}{m_{\mathrm{Cs}}}\right)$$

$$\alpha^{(0)} = 880(10) a_0^3$$

(Theory 870 a_0^3)

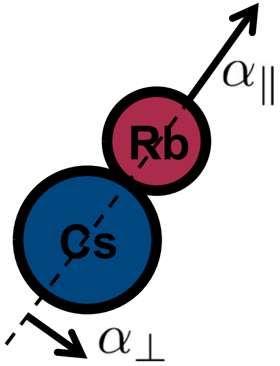
P. D. Gregory *et al.*
PRA **96**, 021402(R) (2017)

Trapping molecules

Durham University

$$H_{\rm AC} = \frac{1}{2\epsilon_0 c}\alpha I$$

$\alpha =$ Polarizability tensor



Polarizability at angle θ to bond axis in molecular frame:

$$\alpha(\theta) = \alpha^{(0)} + \alpha^{(2)} P_2(\cos\theta)$$

$\alpha^{(0)} = \frac{1}{3}(\alpha_{\parallel} + 2\alpha_{\perp})$ Isotropic polarizability – same for all rotational levels

$\alpha^{(2)} = \frac{2}{3}(\alpha_{\parallel} - \alpha_{\perp})$ Anisotropic polarizability – negligible in $N = 0$

– mixes M_N (and M_F) in $N \neq 0$

Results in matrix elements in (N, M_N) basis of the following form

$$\langle 1, M_N' | H_{\rm AC} | 1, M_N \rangle \propto \alpha^{(2)} \begin{pmatrix} 2P_2(\cos\beta) & -\frac{3}{\sqrt{2}}\sin\beta\cos\beta & +\frac{3}{\sqrt{2}}\sin\beta\cos\beta \\ -\frac{3}{\sqrt{2}}\sin\beta\cos\beta & -P_2(\cos\beta) & -\frac{3}{2}\sin^2\beta \\ +\frac{3}{\sqrt{2}}\sin\beta\cos\beta & -\frac{3}{2}\sin^2\beta & -P_2(\cos\beta) \end{pmatrix}$$

Basis

$$\begin{pmatrix} M_N = 0 \\ M_N = +1 \\ M_N = -1 \end{pmatrix}$$

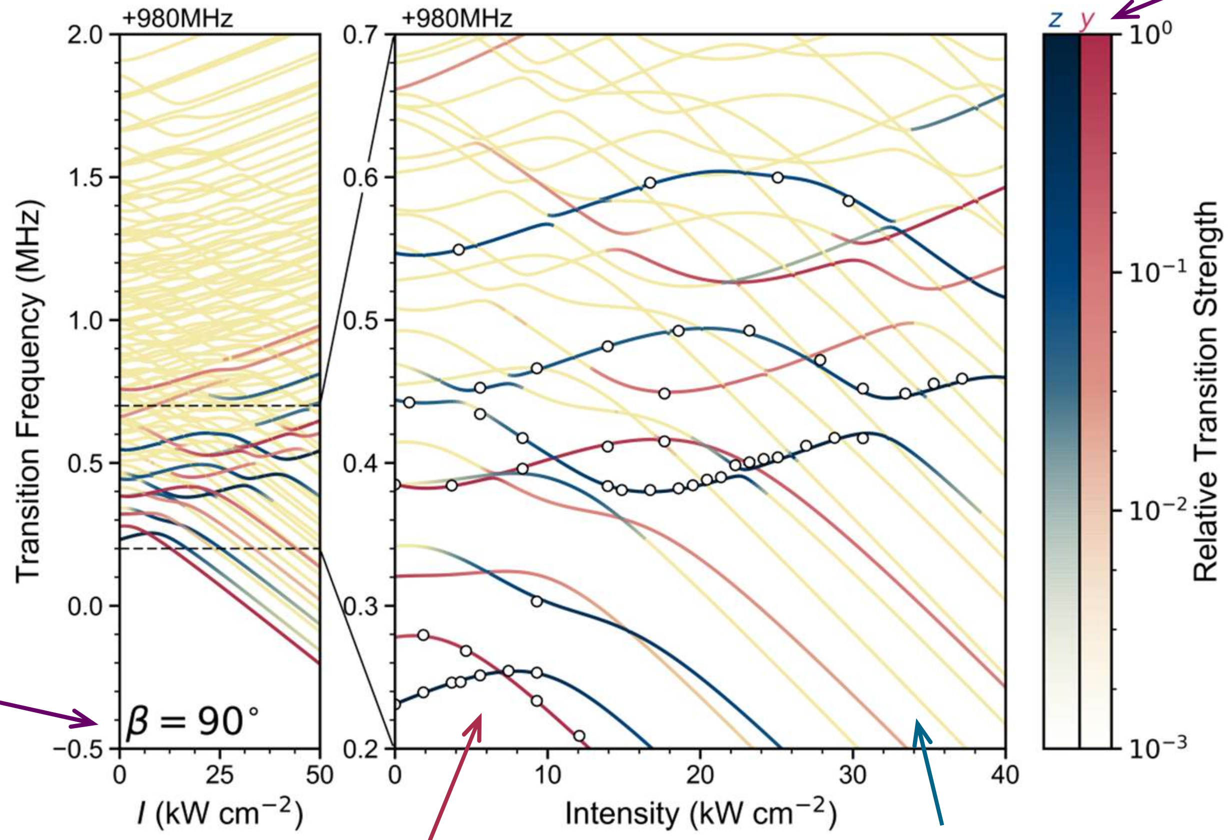
where β is angle between laser polarization and quantisation axis

and $P_2(\cos\beta) = \frac{1}{2}(3\cos^2\beta - 1)$

Gregory et al., PRA **96**, 021402(R) (2017)

AC Stark Effect in $^{87}Rb^{133}Cs$

P. D. Gregory *et al.*
PRA **96**, 021402(R) (2017)



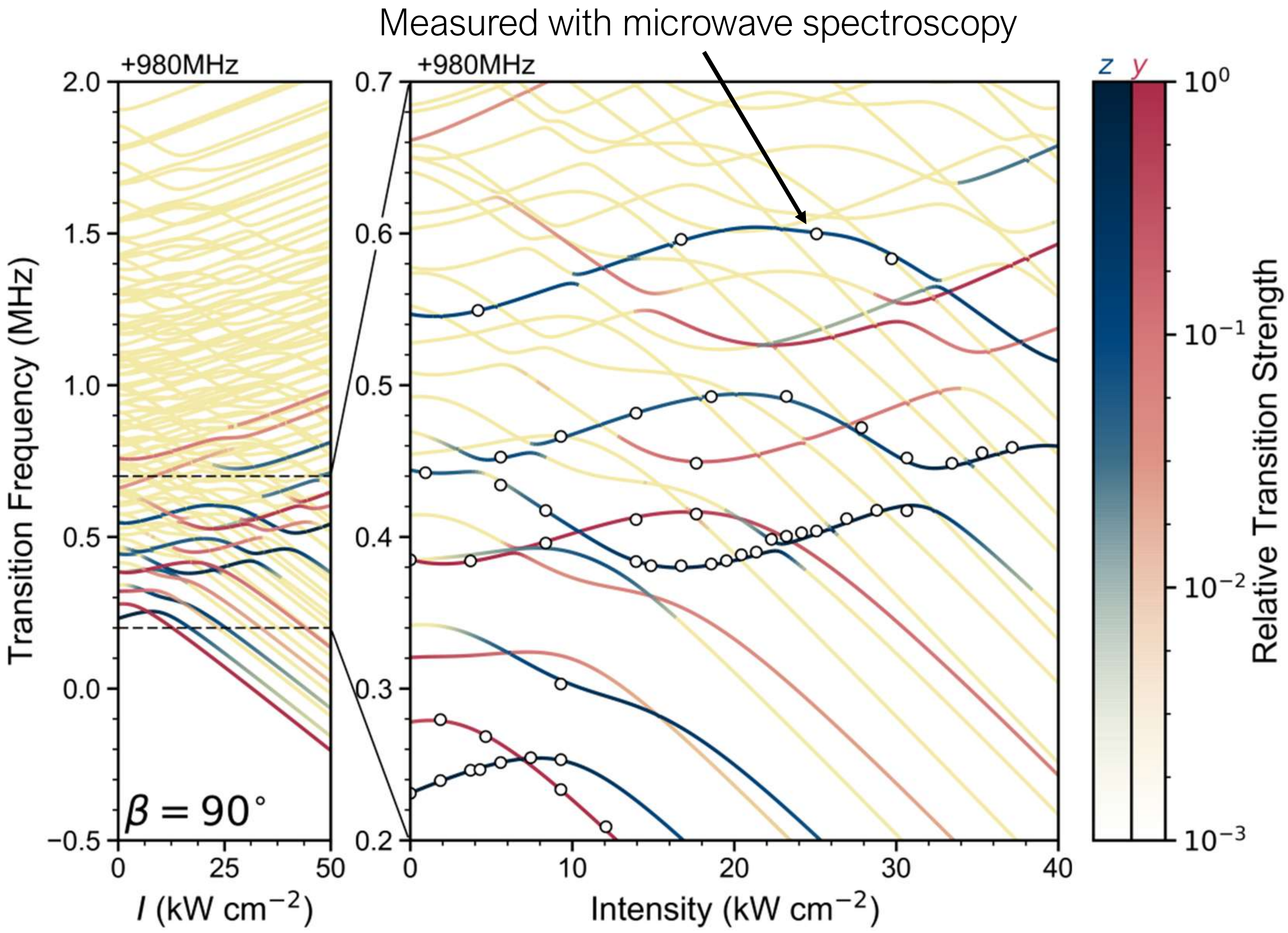
Microwave polarisation

Polarisation angle w.r.t. B_z

Many avoided crossings where $I\alpha^{(2)}$ is comparable to hyperfine couplings and Zeeman splittings

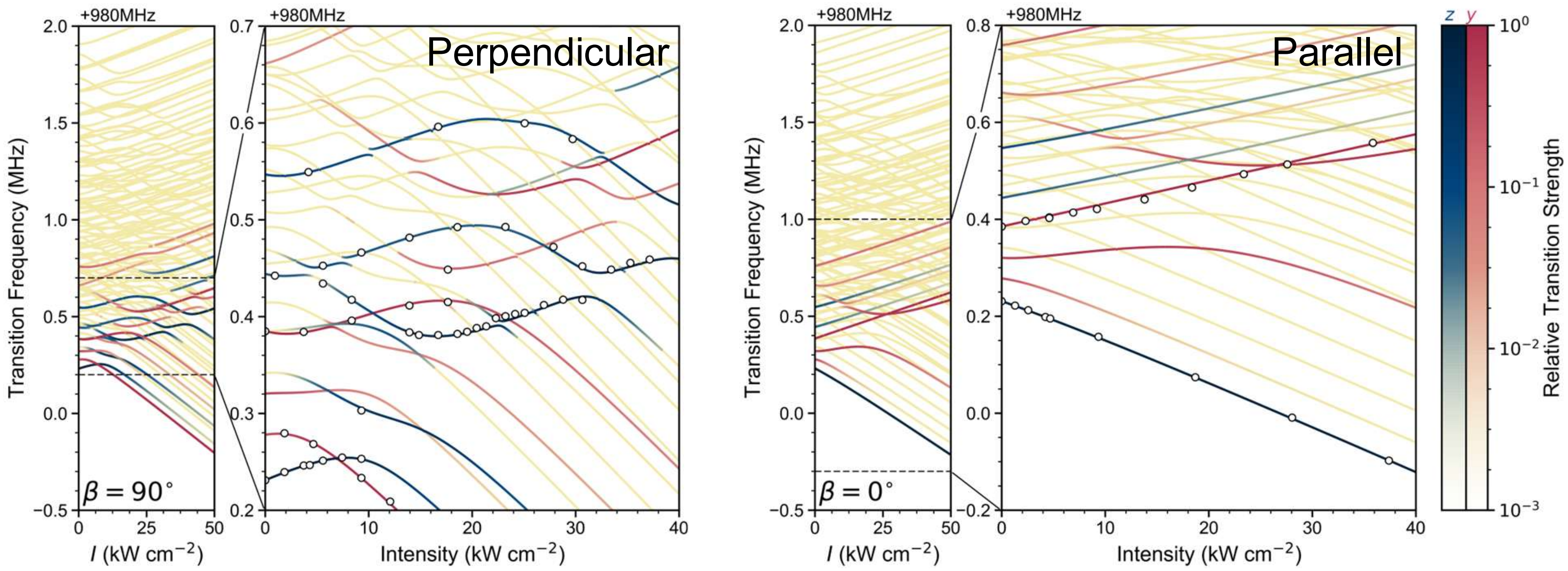
N requantizes along the laser polarisation axis

AC Stark shifts of rotational transitions



Gregory et al., PRA **96**, 021402(R) (2017)

Differential AC Stark shifts in RbCs



Generally, strong intensity dependence leads to rapid dephasing of rotational coherences & suppresses resonant spin-exchange

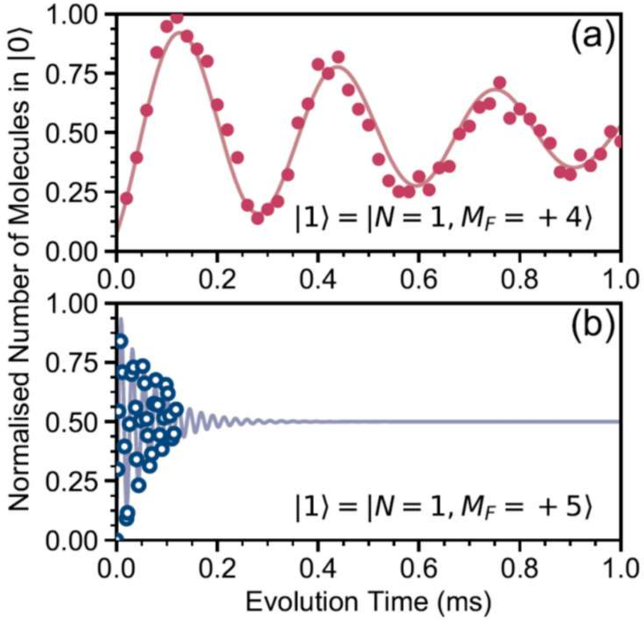
Gregory et al., PRA **96**, 021402(R) (2017)

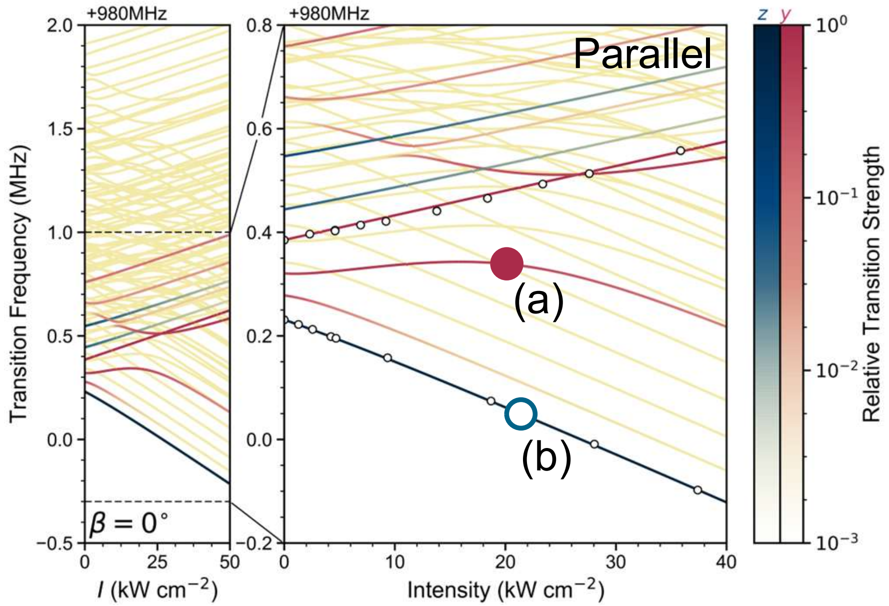
Differential AC Stark shifts in RbCs

Durham University

Blackmore et al., QST 4, 014010 (2019)

Typical Ramsey coherence times ~1ms

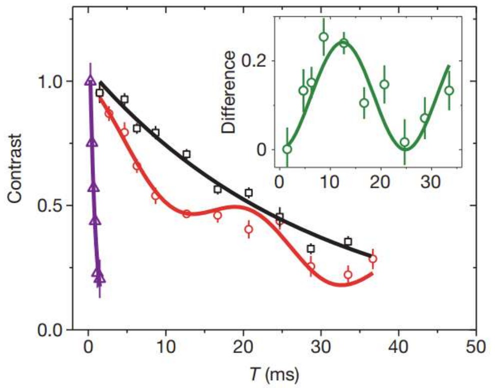




Generally, strong intensity dependence leads to rapid dephasing of rotational coherences & suppresses resonant spin-exchange
Problem for all molecules

Problem common to all molecules

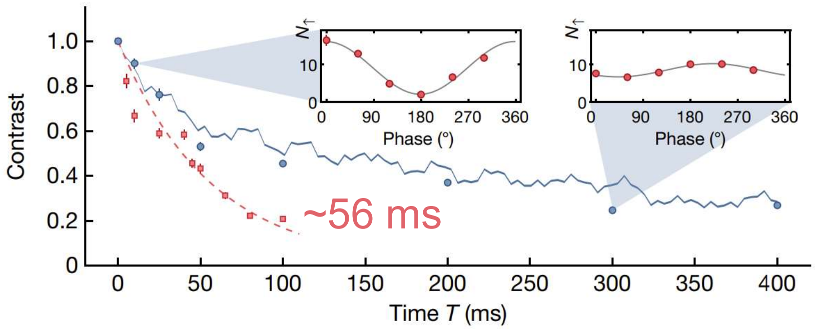
KRb in optical lattice



Single particle dephasing ~ 1 ms

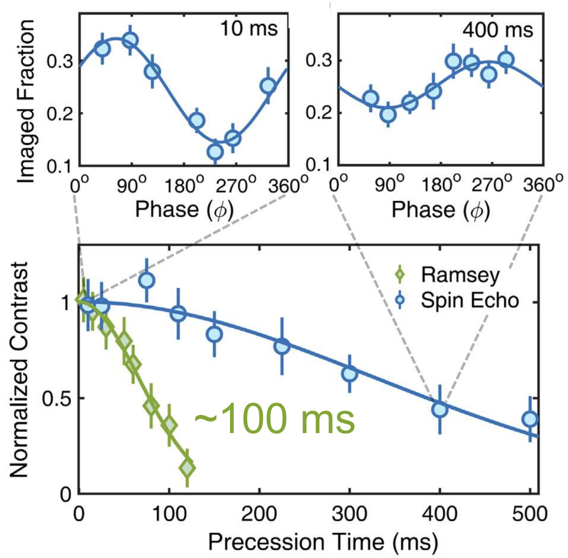
Yan *et al*., Nature 501, 521-525 (2013)

NaRb molecules low filling in near-magic lattice



Christakis *et al*., Nature 614, 64-69 (2023)

Single CaF molecule in magic angle tweezer



Burchesky *et al*., PRL 127,123202 (2021)

NaCs in magic ellipticity tweezer

Park *et al*., PRL 131,183401 (2023)

62(3) ms with single spin-echo pulse

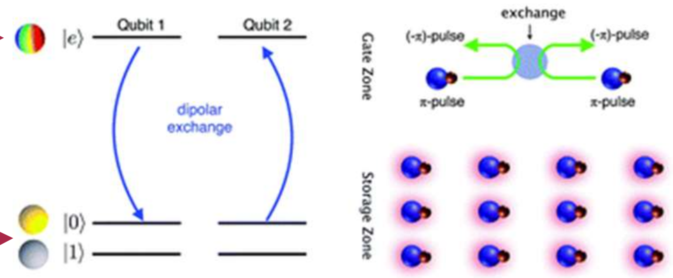
Not a problem in the rotational ground state!
Expect to be able to create long-lived coherent superpositions

A platform for quantum computing

Many proposals… for example:

DeMille, PRL **88**, 067901 (2002).

"Dipolar exchange quantum logic gate with polar molecules"

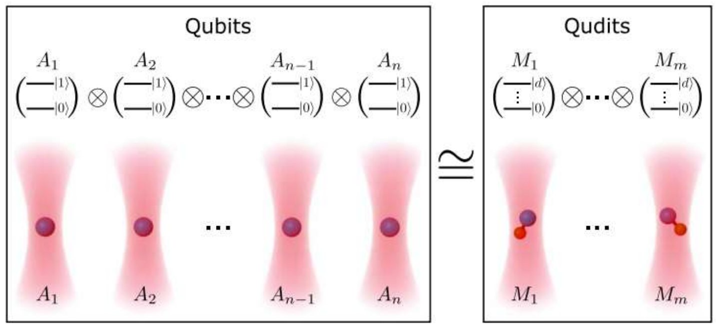


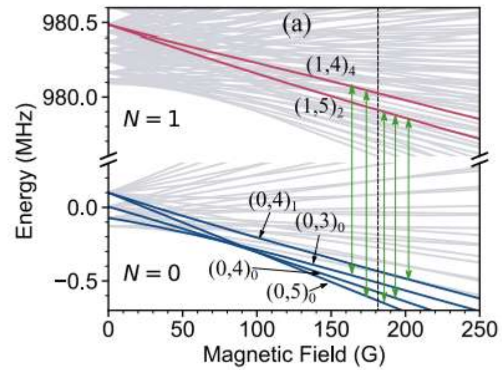
iSWAP gate based on dipolar spin-exchange
Ni *et al.*, Chem. Sci. **9**, 6830 (2018).

Hyperfine states of $N = 0$ used as qudit states (non-interacting)

$N = 1$ state used to introduce controlled spin exchange interactions

"Ultracold molecules as qudits"

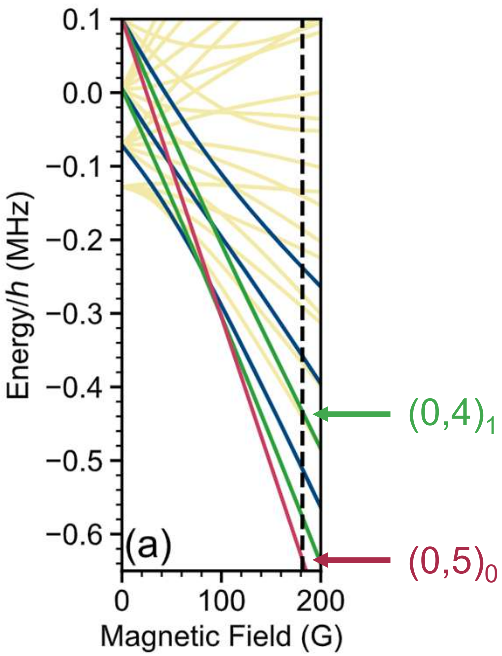




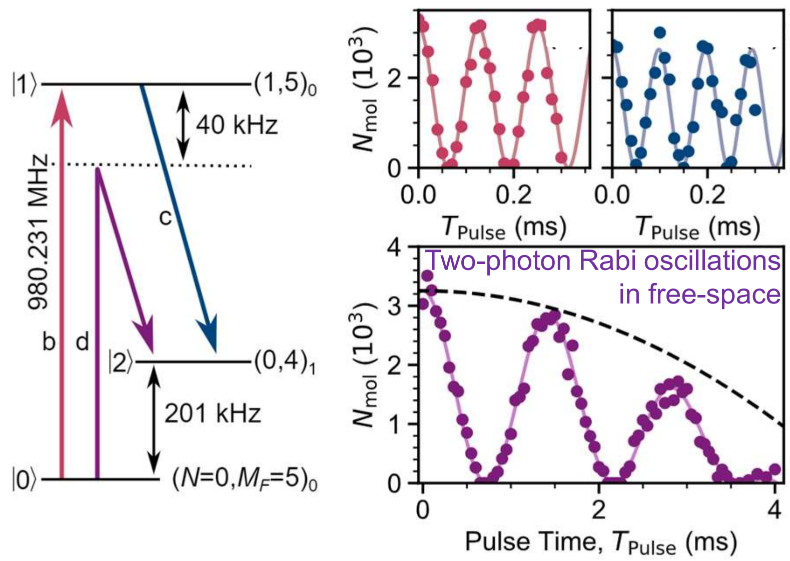
2-photon microwave transitions couple hyperfine levels
Sawant *et al.*, New J. Phys. **22**, 013027 (2020).

RbCs hyperfine coherences

Examine superposition of
$(0,4)_1$ & $(0,5)_0$



Use 2-photon microwave transition to couple states



Blackmore *et al.,* Phys. Chem. Chem. Phys. (2020)

Gregory *et al.* PRA **94**, 041403(R) (2016)

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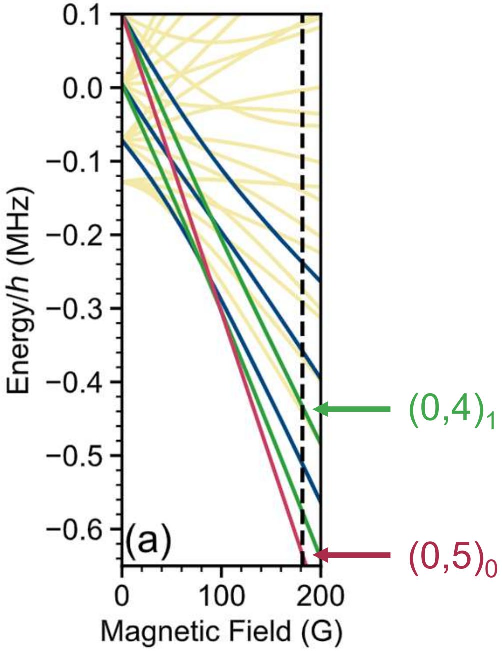
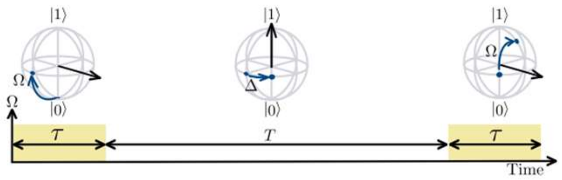


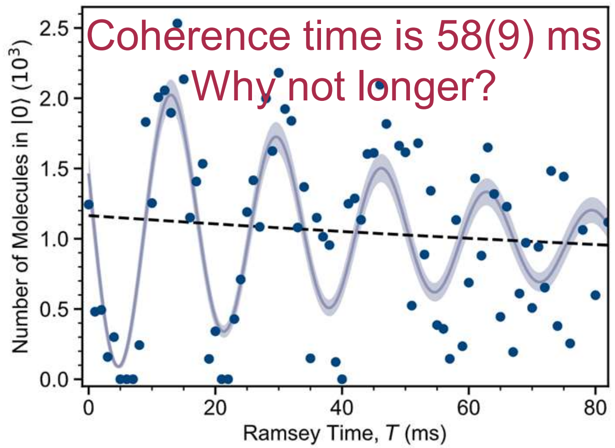
c.f. MIT: 0.7(3)s in NaK *Science* 357, 372-375 (2017)

Expect **long coherence** because:

- Only nuclear magnetic moments ($^1\Sigma$)
- Polarizability should be almost identical

Perform Ramsey measurement:

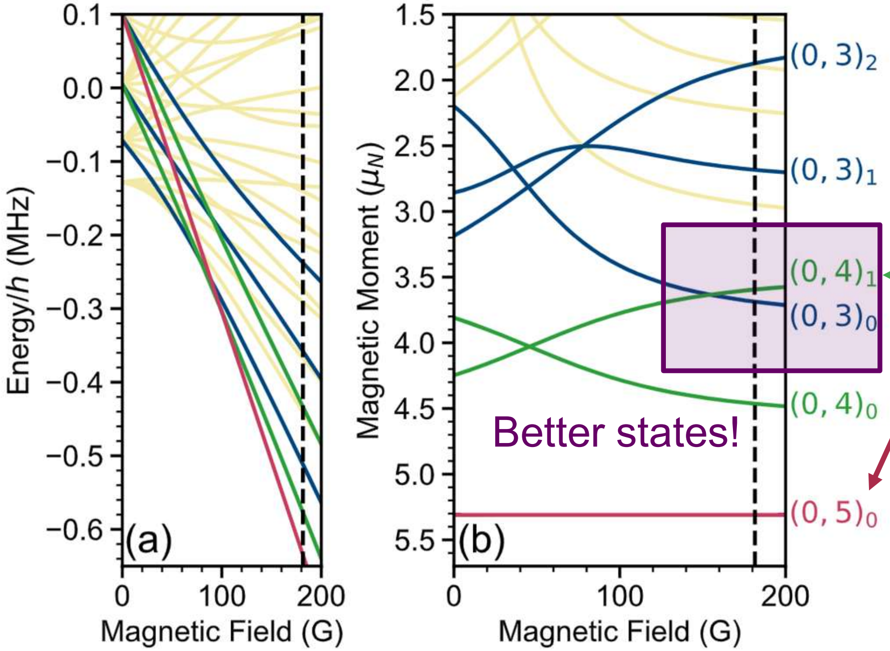


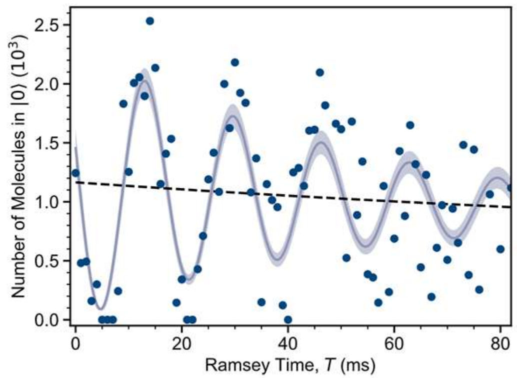


Coherence time is 58(9) ms

Why not longer?

Magnetic decoherence





Ramsey measurements used states $(0,4)_1$ and $(0,5)_0$

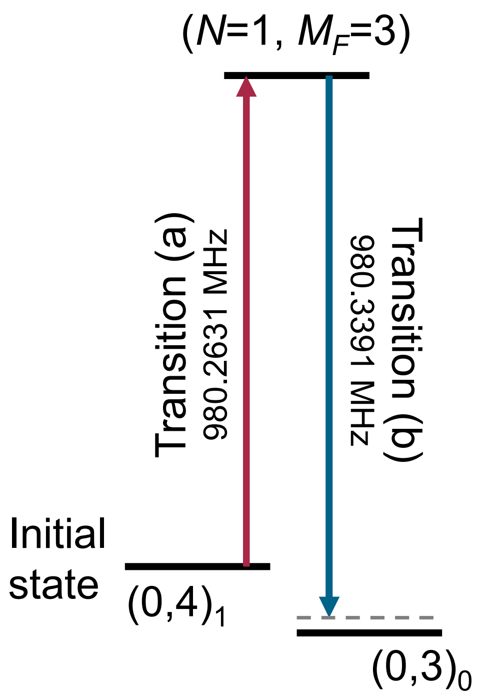
Differential magnetic moment is $1.8\mu_N$ at 181.5 G

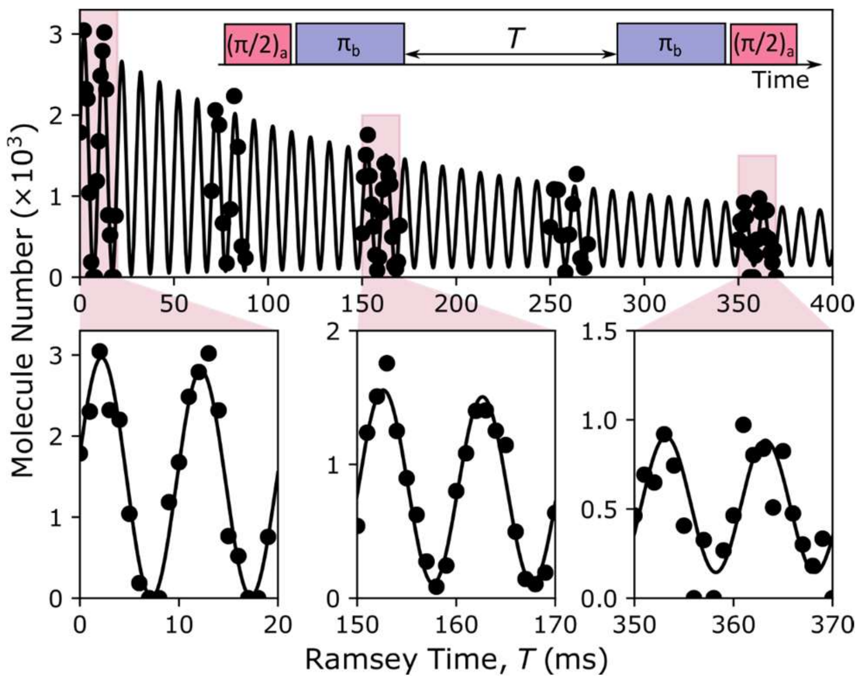
100ms coherence time requires magnetic field to be stable to 7mG

Gregory *et al*. Nat. Phys. **17**, 1149 (2021)

Good ground-state Ramsey

Choose states with same magnetic moment for Ramsey: $(0,4)_1$ and $(0,3)_0$ at 154.5G





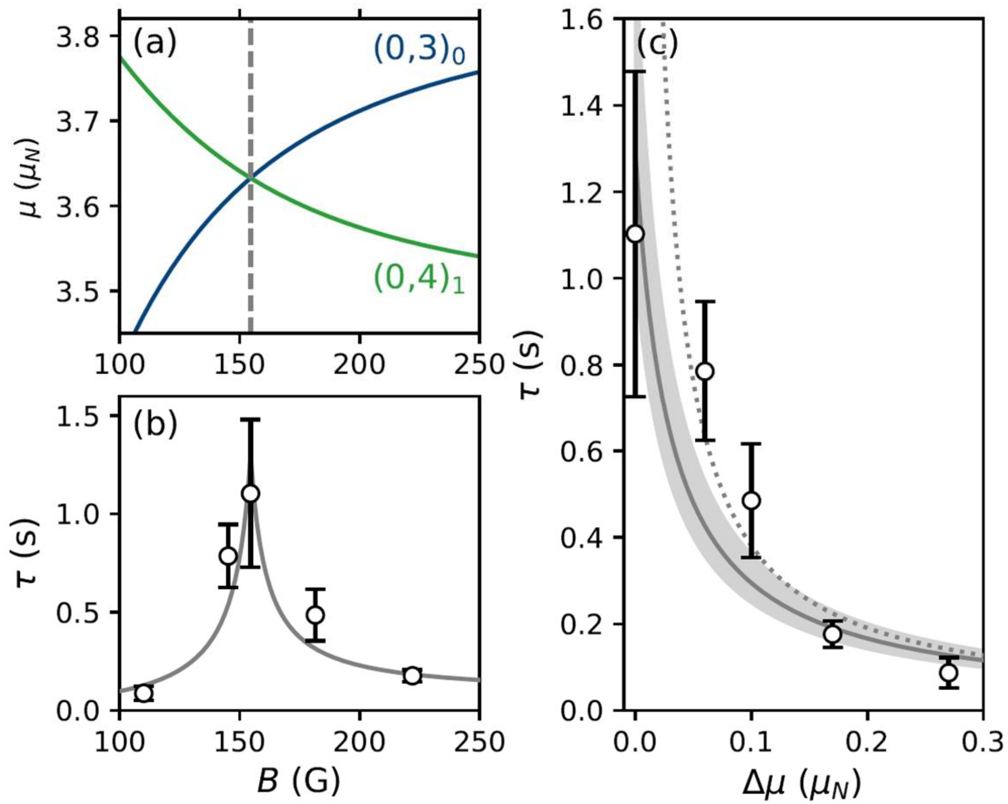
1/e collisional lifetime:
0.33(3) seconds

Coherence time:
1.1(4) seconds

Gregory *et al*. Nat. Phys. **17**, 1149 (2021)

Mapping out magnetic dependence

Difference in magnetic moments causes decoherence due to magnetic field noise



Fit coherence time with:

$$\tau = \left(\frac{\Delta\mu \Delta B}{h} + \frac{1}{T_2^*} \right)^{-1}$$

Consistent with magnetic field noise of 34(5) mG

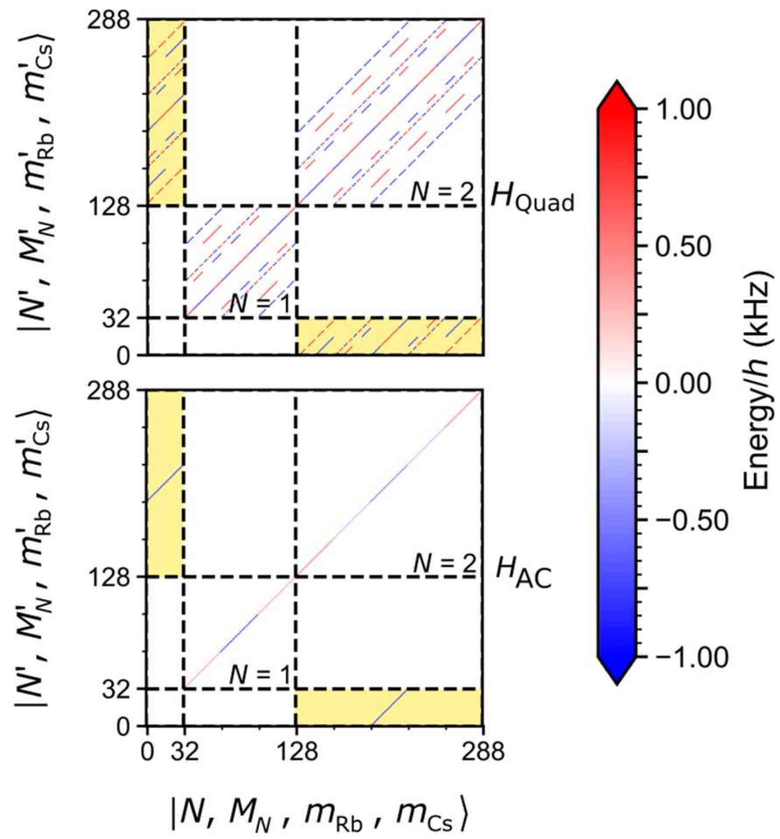
Maximum coherence time:

$$T_2^* = 1.3(4)\,\mathrm{s}$$

No surprises. But what limits coherence at 154.5 G?

Small differential tensor light shifts

Off-diagonal elements in the Hamiltonian mix states with $\Delta N = 2$



Nuclear electric quadrupole interaction

$$H_{\mathrm{quad}} = \sum_{j=\mathrm{Rb,Cs}} e\boldsymbol{Q}_j \cdot \boldsymbol{q}_j,$$

Anisotropic component of polarizability

$$H_{\mathrm{AC}} = -\frac{1}{2}\boldsymbol{E}_{\mathrm{AC}} \cdot \boldsymbol{\alpha} \cdot \boldsymbol{E}_{\mathrm{AC}}$$

Largest contributions are 2nd order terms:

$$\langle 0,0| H_{\mathrm{AC}}^{(2)} |2,0\rangle \langle 2,0| H_{\mathrm{quad}} |0,0\rangle$$

Together produce a small tensor polarizability in the ground state:

$$\Delta\alpha = X\alpha^{(2)} P_2(\cos\beta)$$

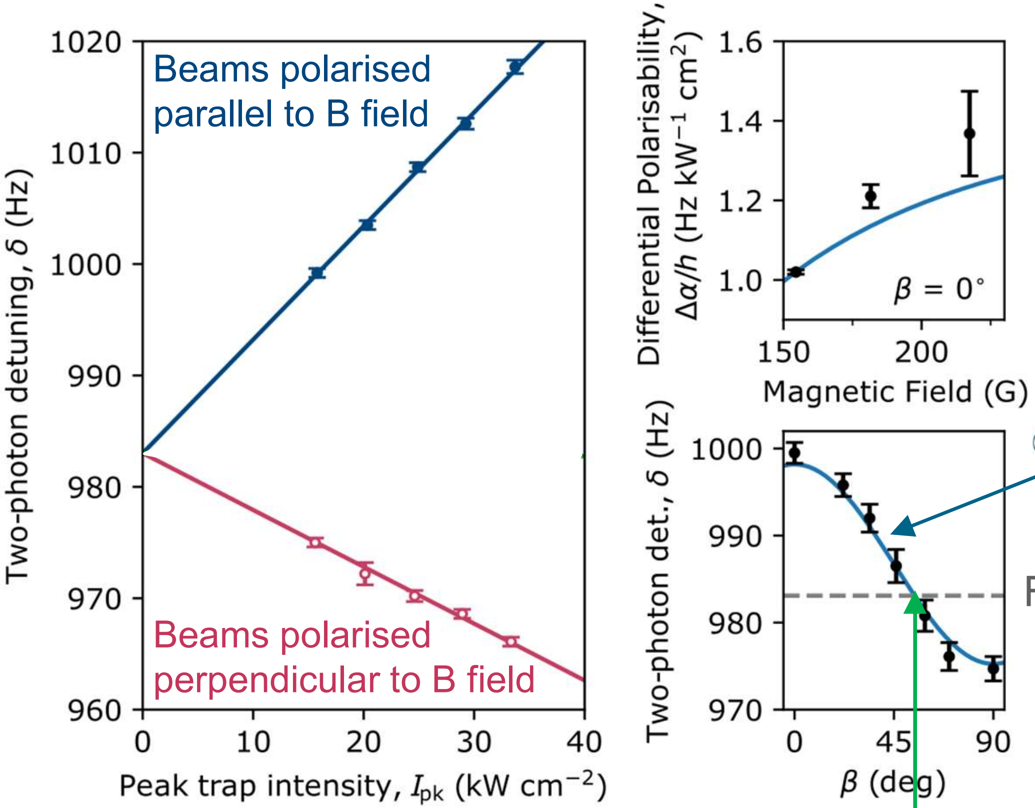
$$X = 3.995... \times 10^{-5}$$

(Quantifies differential admixture of N > 0)

Measuring differential tensor light shifts

Durham University

Use Ramsey to measure energy difference between states as a function of intensity:



$\propto P_2(\cos\beta) = \frac{1}{2}(3\cos^2\beta - 1)$

Free-space value

$= 0$ for $\beta \simeq 55°$

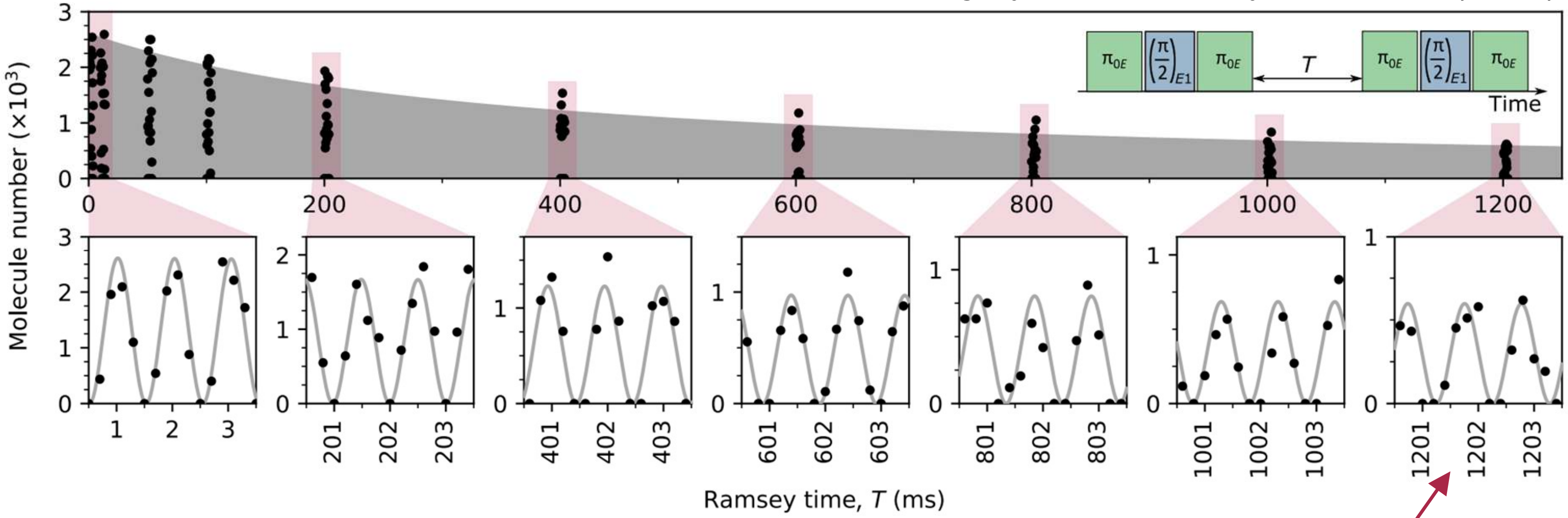
Magic angle trap – no light shifts!

Gregory *et al*. Nat. Phys. **17**, 1149 (2021)

The result

Magic angle trap with a bias field of 154.5 G eliminates differential tensor light shifts AND magnetic sensitivity → Robust Storage Qubits

Gregory *et al*. Nat. Phys. **17**, 1149 (2021)



Coherence time > 6.9 seconds (90% confidence*)

(Estimated limits: ΔB=35 mG → 2000 s & $\Delta\beta$=0.5° → 80 s)

No detectable loss of coherence after 1.2 s

How can we extend this to rotational coherences?

*Feldman & Cousins Phys. Rev. D **57**, 3873 (1998)

Engineering a rotationally magic trap

Require zero anisotropic polarizability

$$\alpha^{(2)} = \frac{2}{3}(\alpha_{||} - \alpha_{\perp})$$

Components come from sum over all allowed electronic transitions with a given symmetry

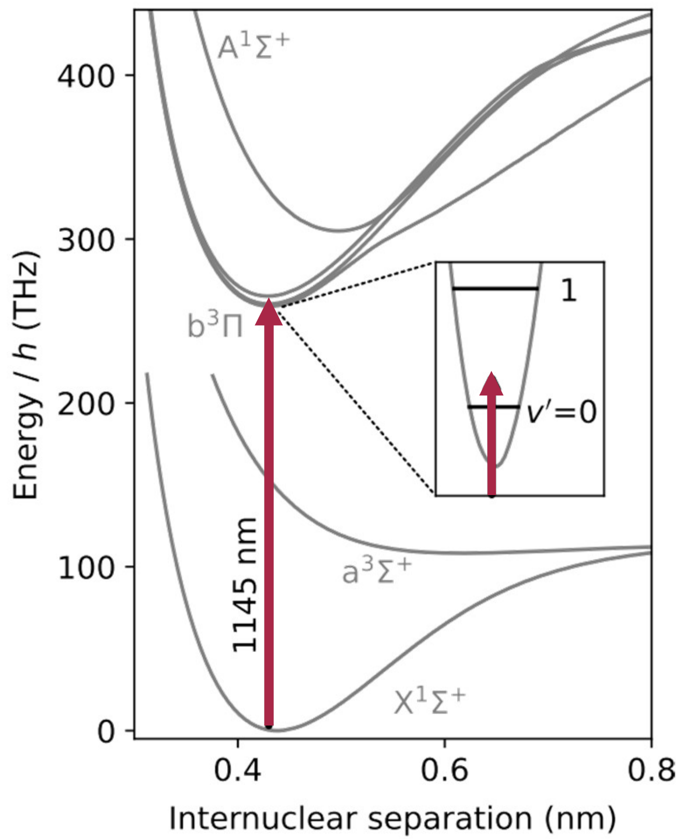
$$\alpha_{||} \rightarrow {}^1\Sigma \qquad \alpha_{\perp} \rightarrow {}^1\Pi$$

Tune one component by trapping close to a transition to a state with a given symmetry

Choose nominally forbidden transition
$X^1\Sigma^+ \rightarrow b^3\Pi_0$ (weakly mixed with $A^1\Sigma^+$)

Tune $\alpha_{||}$ without affecting $\alpha_{\perp}$ so that

$\alpha_{||} = \alpha_{\mathrm{bg},\perp}$ and $\alpha^{(2)} = 0$



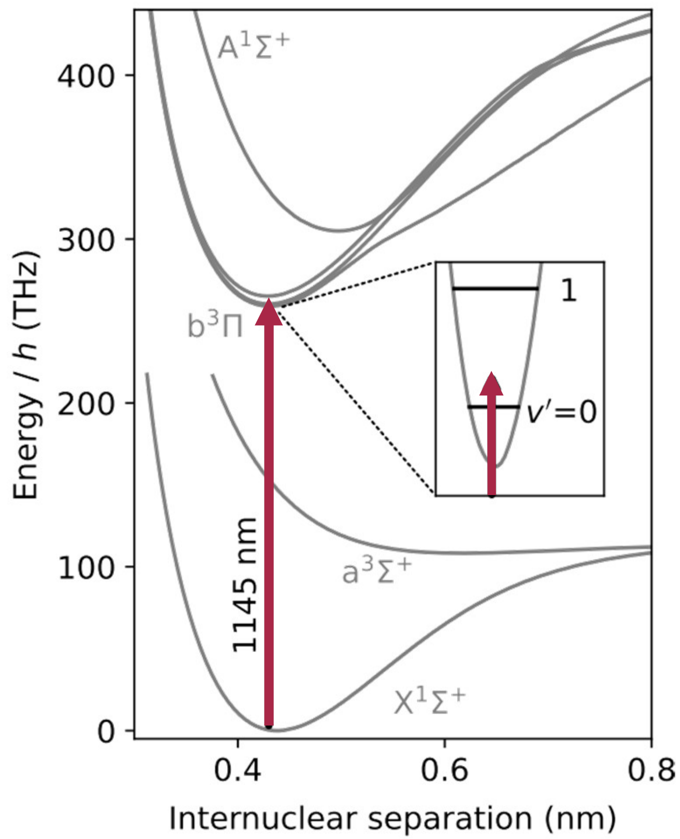
With

Svetlana Kotochigova

Guan, Cornish, and Kotochigova
Phys. Rev. A **103**, 043311 (2021)

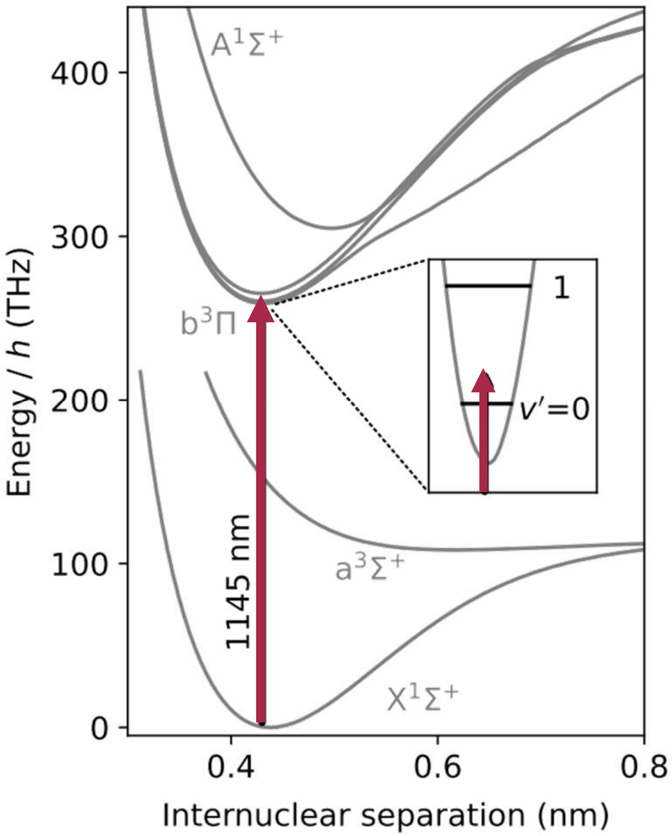
c.f. Bause *et al.*, PRL **125**, 023201 (2020)
He *et al.*, NJP 23, 115003 (2021)

Engineering a rotationally magic trap



Guan, Cornish, and Kotochigova
Phys. Rev. A **103**, 043311 (2021)

Far-detuned magic wavelength between vibrational levels in $b^3\Pi$

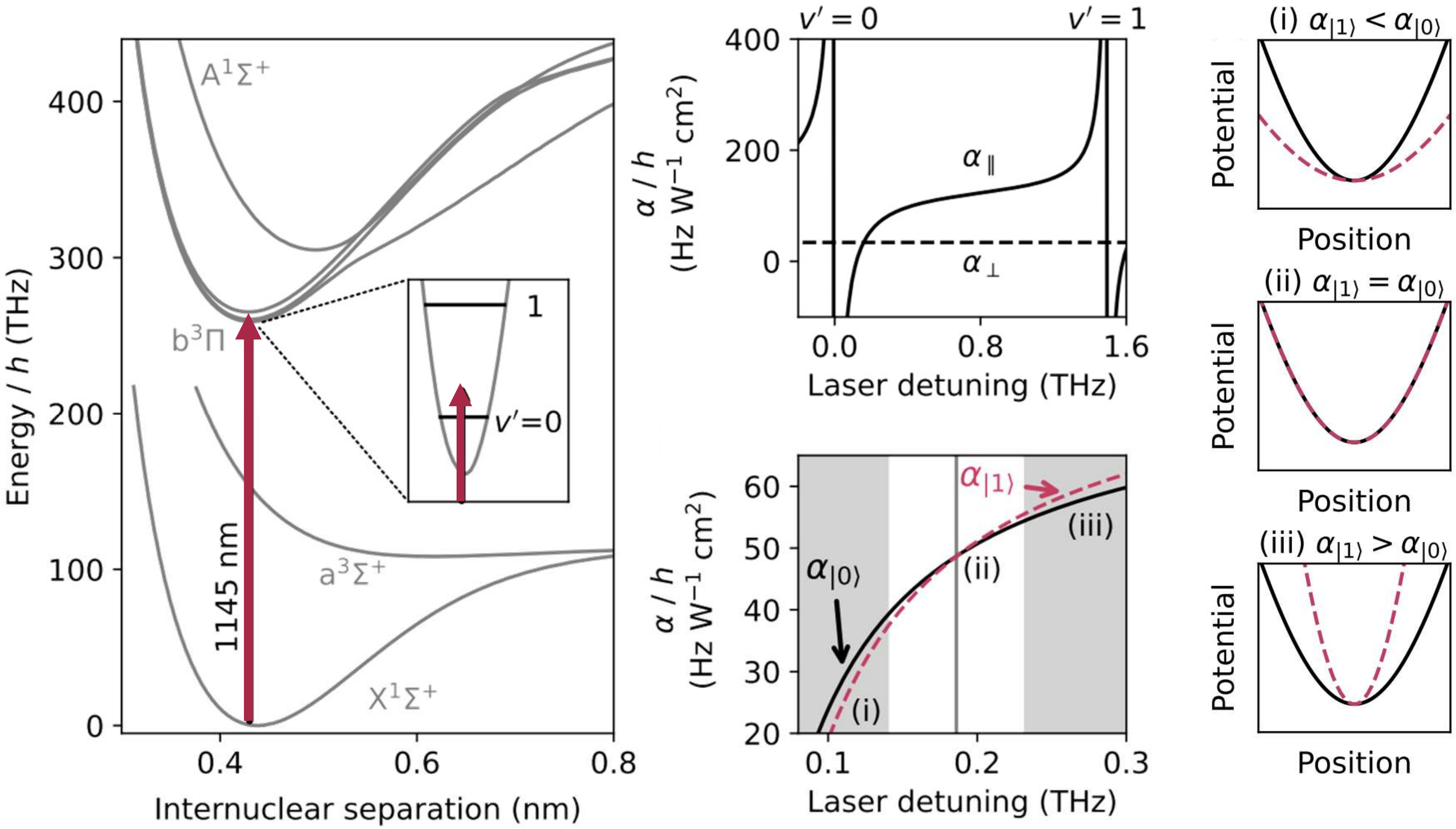


Guan, Cornish, and Kotochigova
Phys. Rev. A **103**, 043311 (2021)

Engineering a rotationally magic trap

Durham University

Far-detuned magic wavelength between vibrational levels in $b^3\Pi$



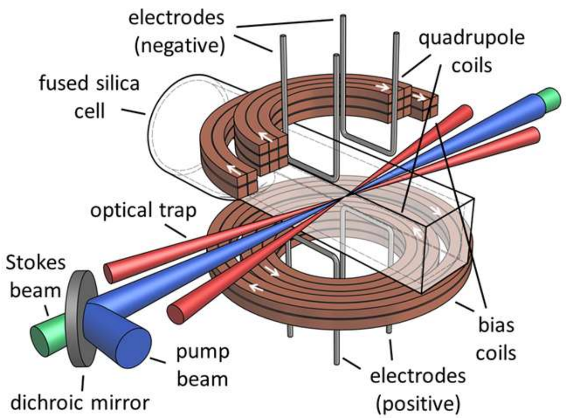
Guan, Cornish, and Kotochigova
Phys. Rev. A **103**, 043311 (2021)

Linewidths: Γ(v' = 0) = 14(1) kHz, Γ(v' = 1) = 8(1) kHz
Magic detuning = 186 GHz !

Coarse spectroscopy

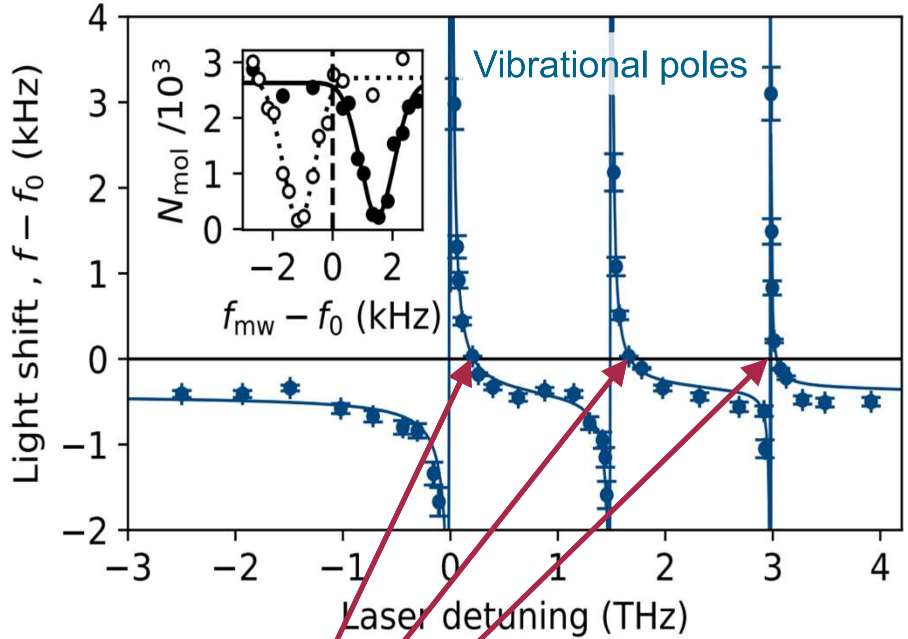
Durham University

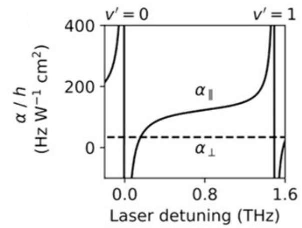
Thermal gas of RbCs molecules in crossed optical dipole trap (with lossy collisions)



Molony et al., PRL **113**, 255301 (2014)

Measure light shift of $(N = 0, M_N = 0) \rightarrow (1,1)$ microwave transition



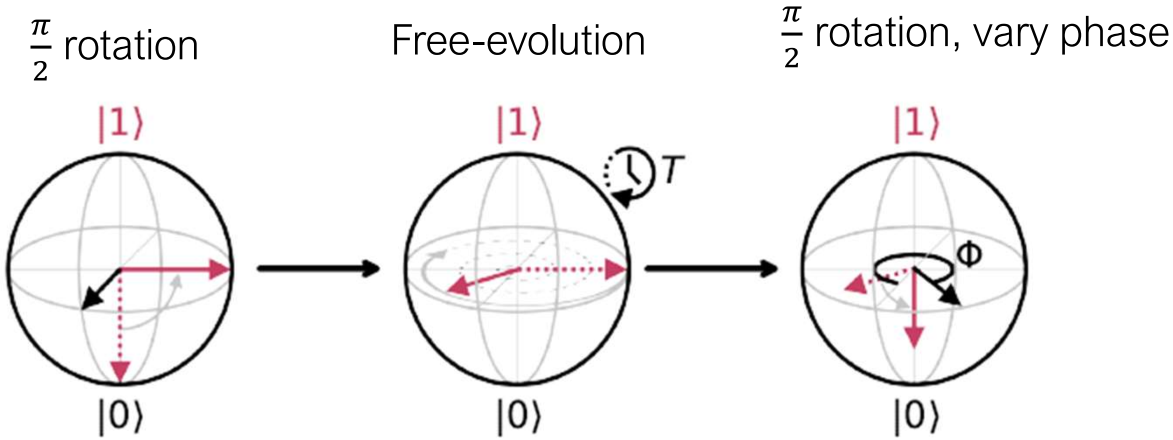


Rotationally magic

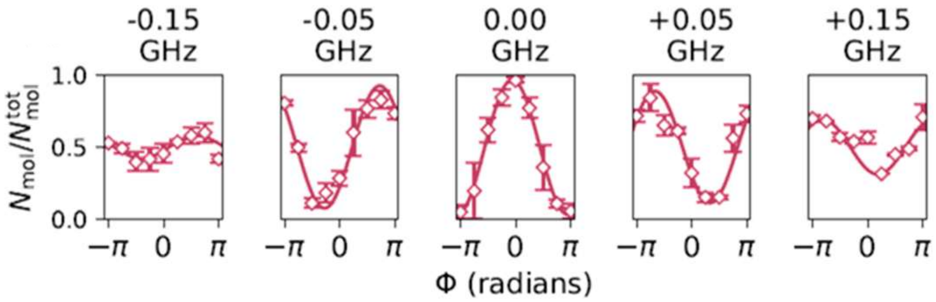
Optimising coherence in the magic trap

Durham University

Perform Ramsey spectroscopy, fixed Ramsey time. Optimise fringe contrast.



Detuning from “magic”



Gregory *et al.*, Nature Physics 20, 415 (2024)

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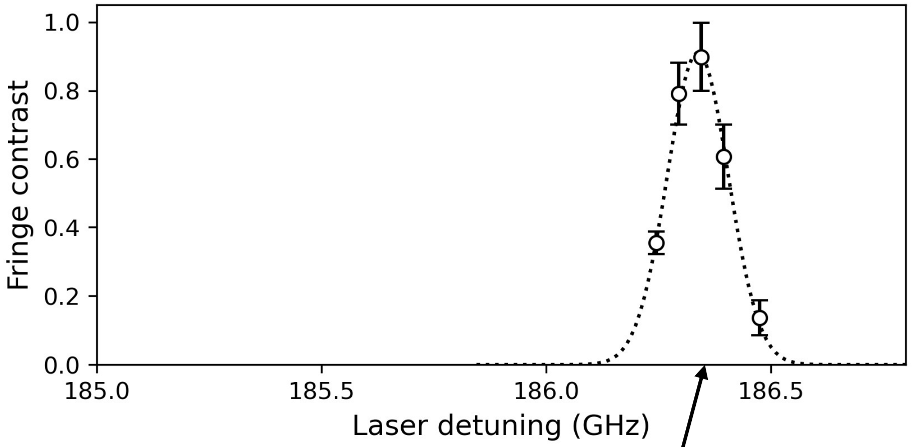
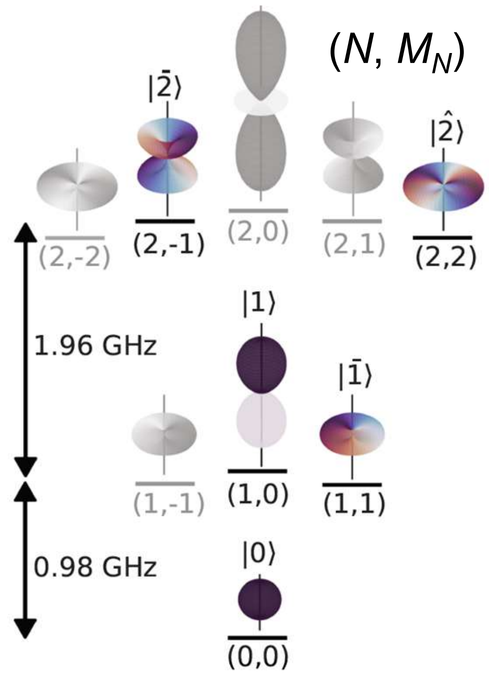


$$\frac{1}{\sqrt{2}}(|0\rangle + |\hat{2}\rangle)$$



Gregory *et al.*, Nature Physics 20, 415 (2024)

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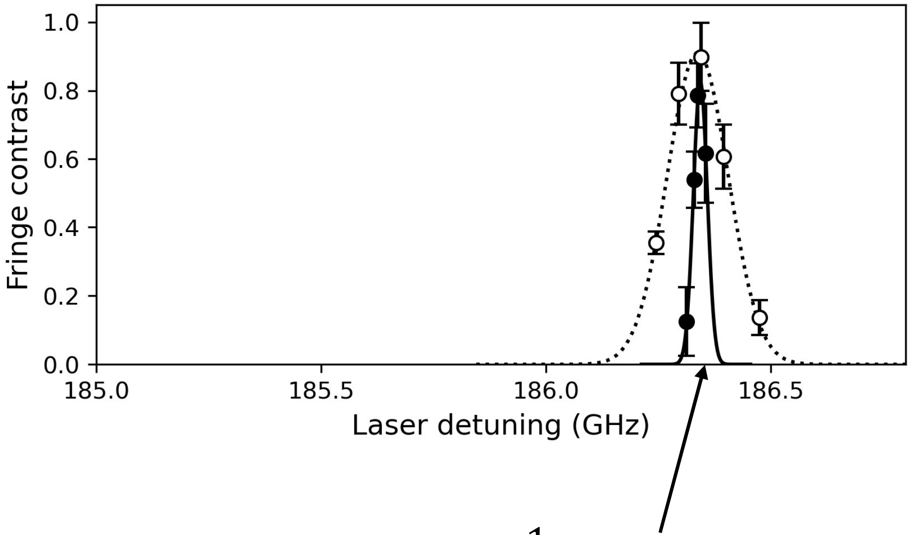

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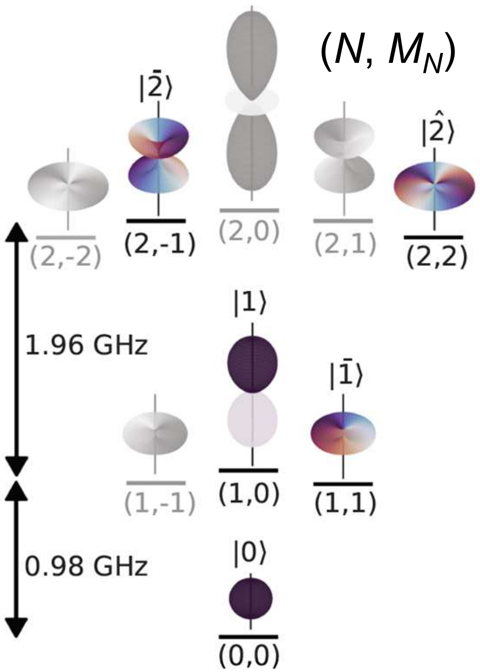

Gregory *et al.*, Nature Physics 20, 415 (2024)

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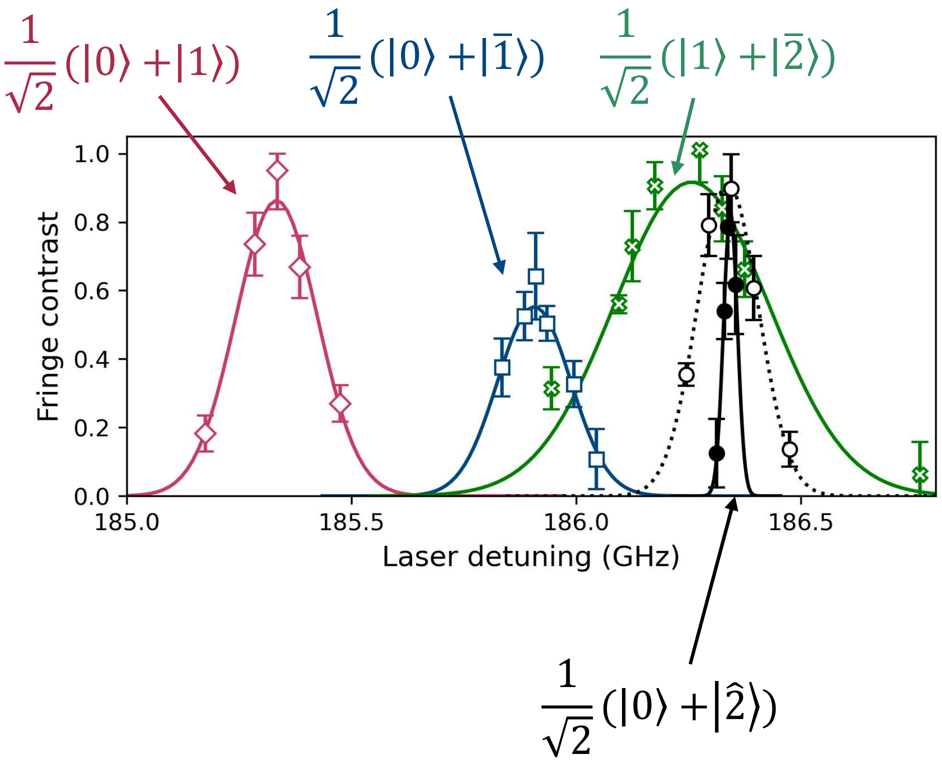
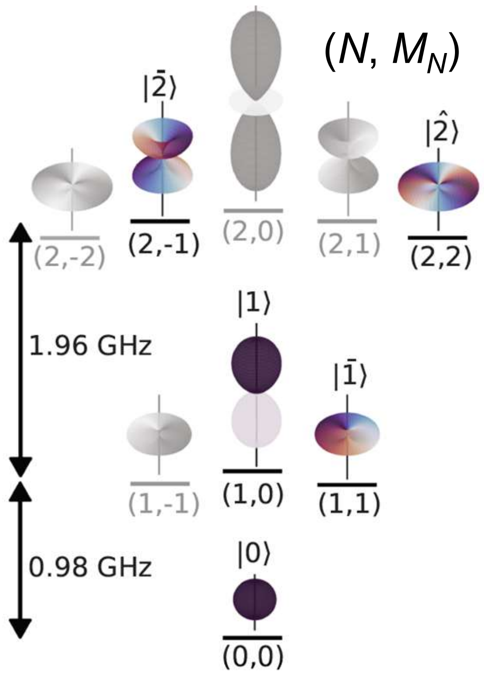


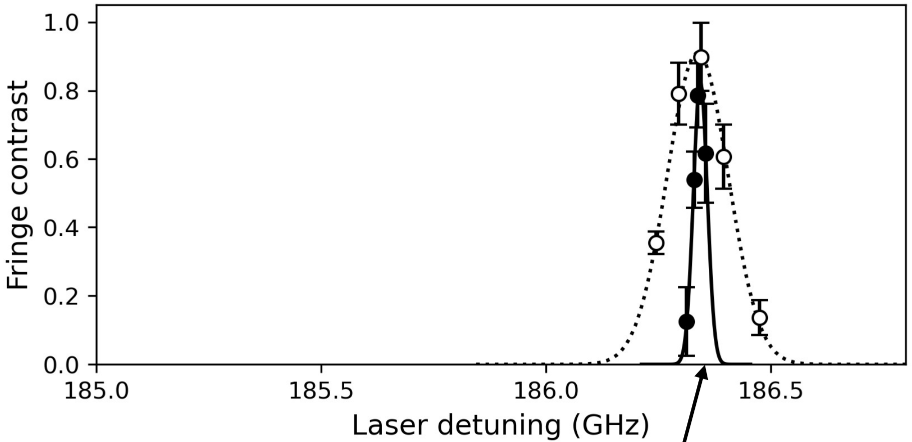


Gregory *et al.*, Nature Physics 20, 415 (2024)

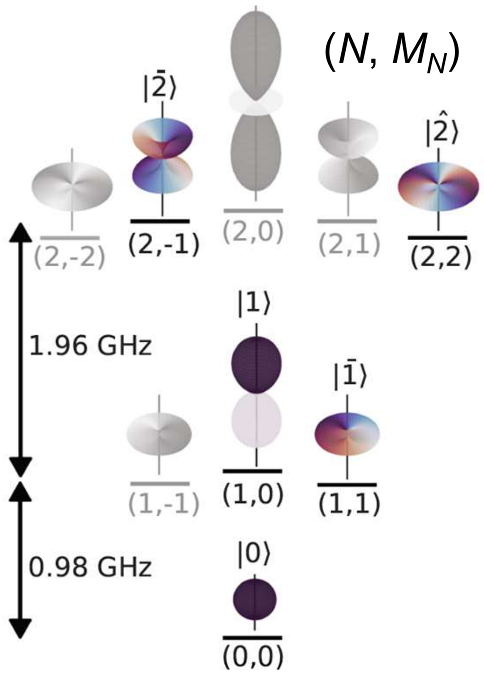
Optimising coherence in the magic trap

Perform Ramsey spectroscopy, fixed Ramsey time. Optimise fringe contrast.

No direct TDM, so no DDI !



$$\frac{1}{\sqrt{2}}(|0\rangle + |\hat{2}\rangle)$$

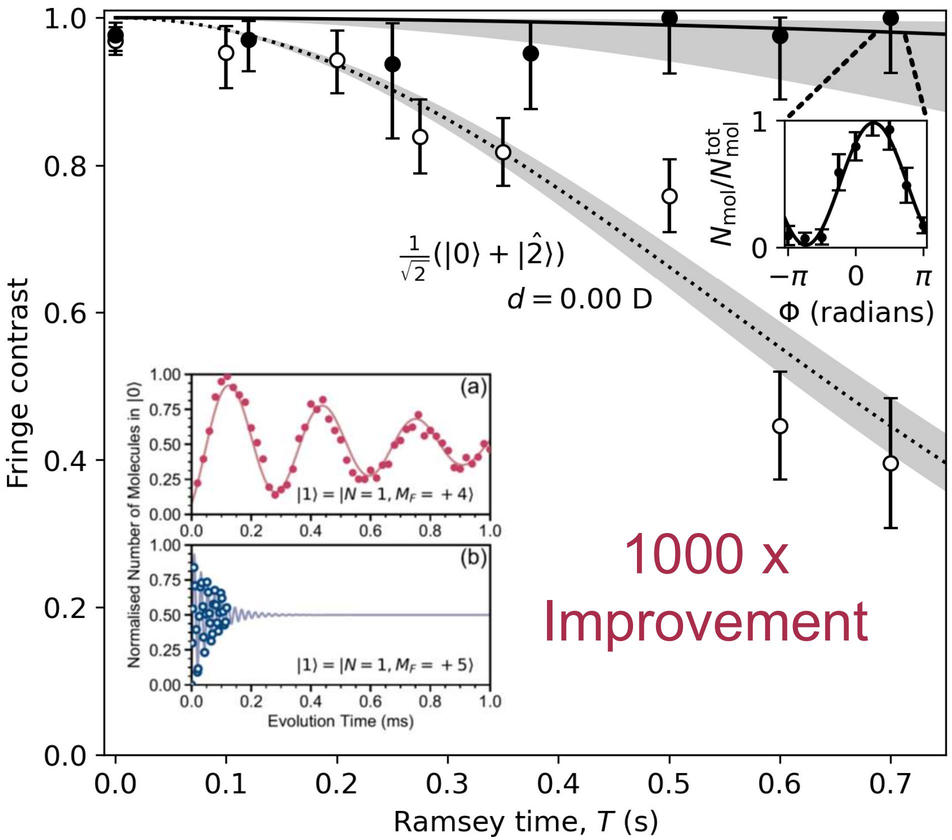


Gregory *et al.*, Nature Physics 20, 415 (2024)

Second-scale rotational coherence

Durham University

Measure coherence of $(0,0) \leftrightarrow (2,2)$ superposition. No TDM, so no DDI !



With spin echo

$T_2 > 1.4$ seconds

(95% confidence level)

Without spin echo

$T_2^* = 0.78(4)$ seconds

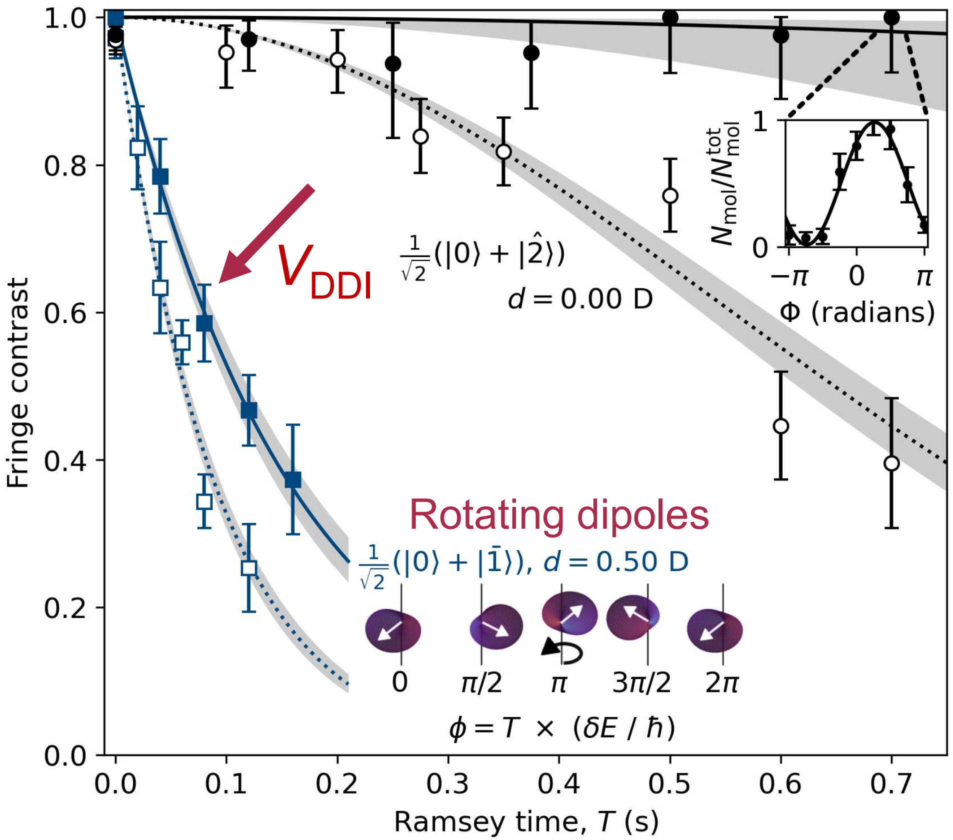
Limited by frequency stability of the trap laser ± 0.76 MHz. (scanning transfer cavity lock)

Long enough to see DDI….

Gregory *et al.*, Nature Physics 20, 415 (2024)

Second-scale rotational coherence

B = 181.5 G. No electric field.



With spin echo

$T_2 > 1.4$ seconds

(95% confidence level)

With interactions (spin echo)

$T_2^{DDI} = 0.157(14)$ seconds

Dephasing due to random thermal distribution of molecules

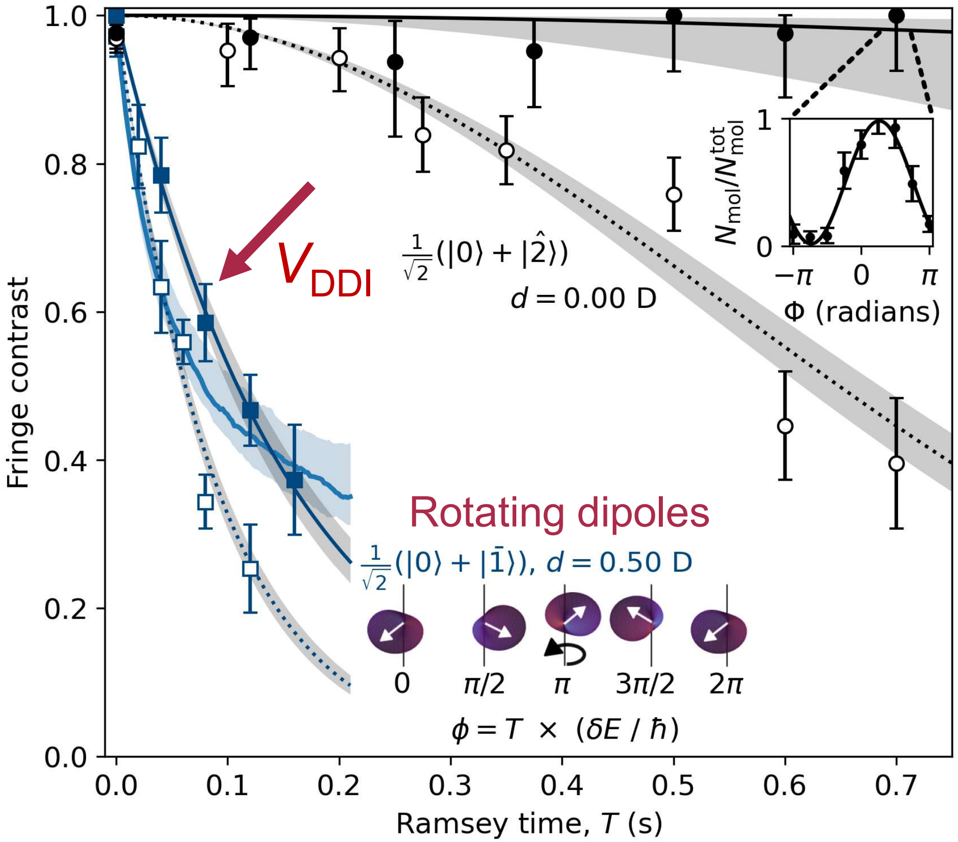
Gregory *et al.*, Nature Physics 20, 415 (2024)

Second-scale rotational coherence

Agreement with Moving Average Cluster Expansion (MACE) simulations

B = 181.5 G. No electric field.

Kaden Hazzard



With spin echo

T_2 > 1.4 seconds

(95% confidence level)

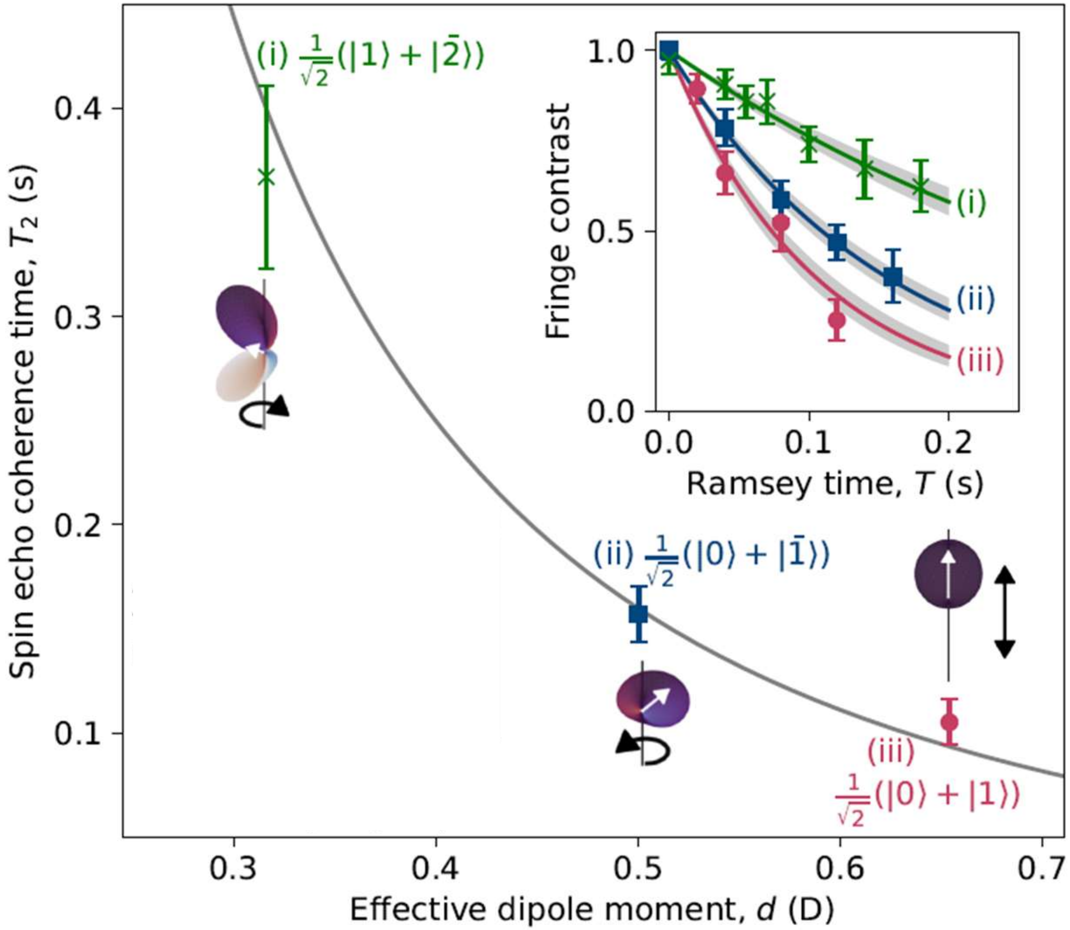
With interactions (spin echo)

$T_2^{DDI} = 0.157(14)$ seconds

Dephasing due to random thermal distribution of molecules

Gregory *et al.*, Nature Physics 20, 415 (2024)

Controllable dipole-dipole interactions



Change superposition
to control size of dipole

$$T_2 \propto 1/V_{DDI}$$

Next step:
Arrays of molecules

Dephasing due to random thermal distribution of molecules

Gregory *et al.*, Nature Physics 20, 415 (2024)

Questions?