Assembly of ultracold polar molecules in optical tweezers

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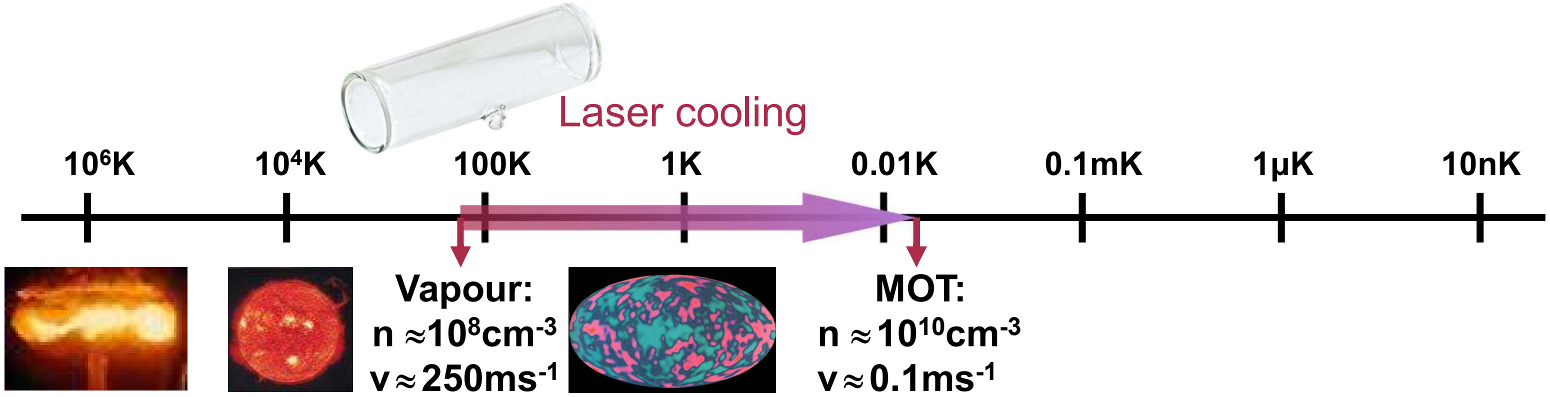
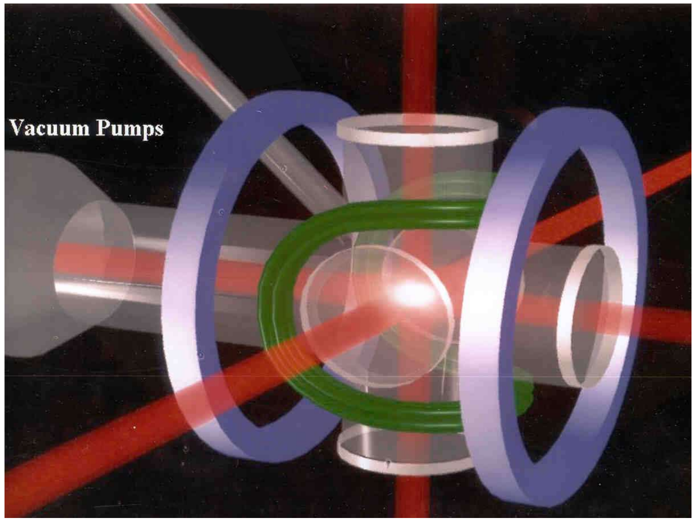
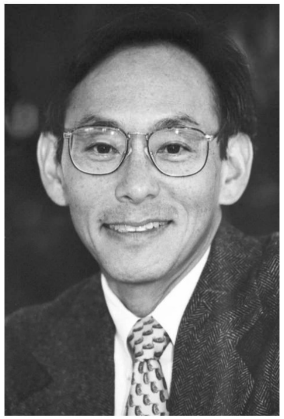
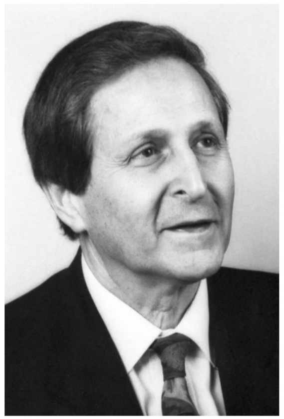
What is ultracold?





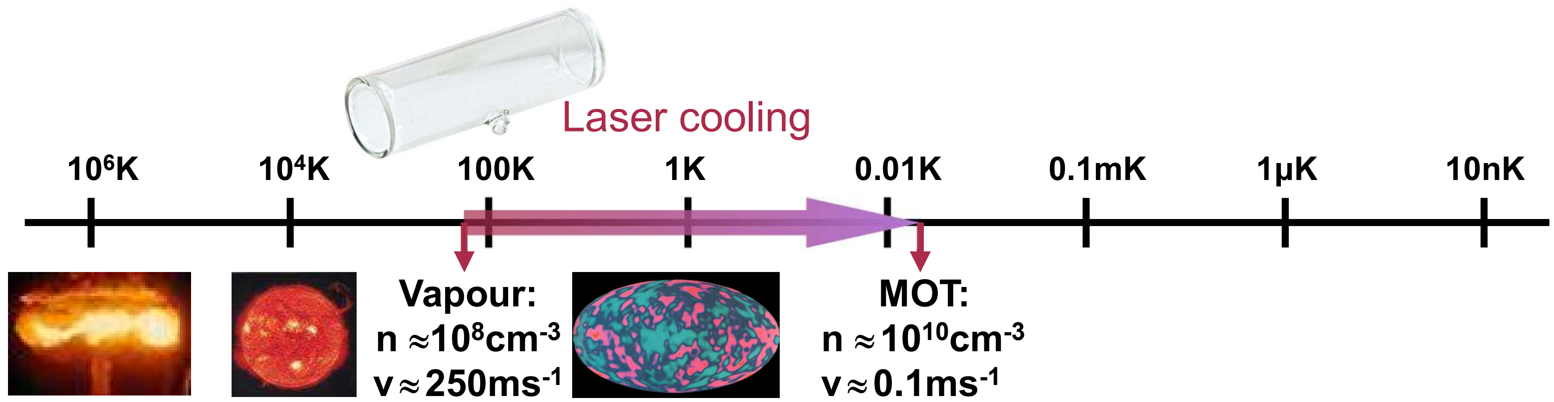
Nobel Prize in Physics 1997


Chu

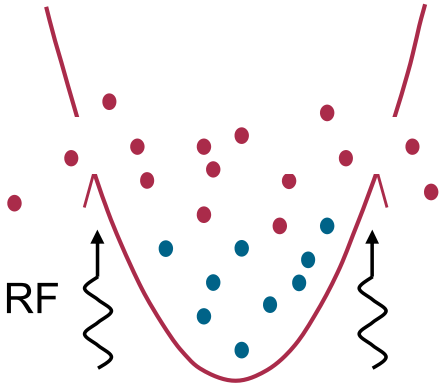

Cohen-Tannoudji

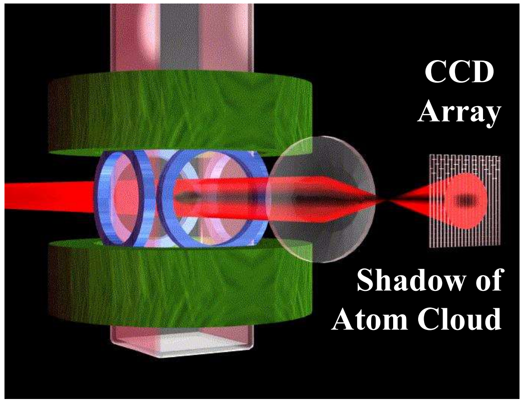

Phillips

What is ultracold?



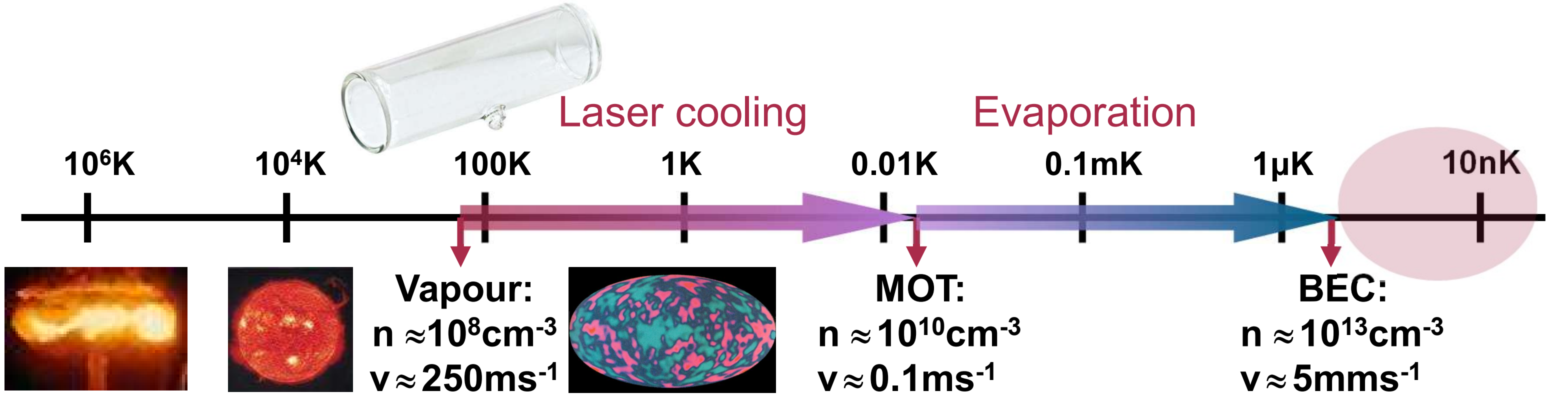
Cold enough to trap and image



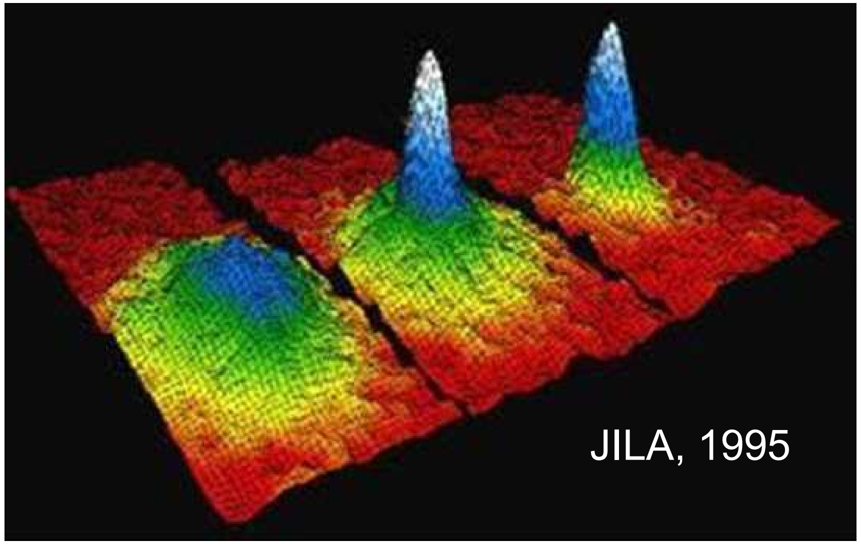


Magnetic trap
(or far-detuned optical trap)

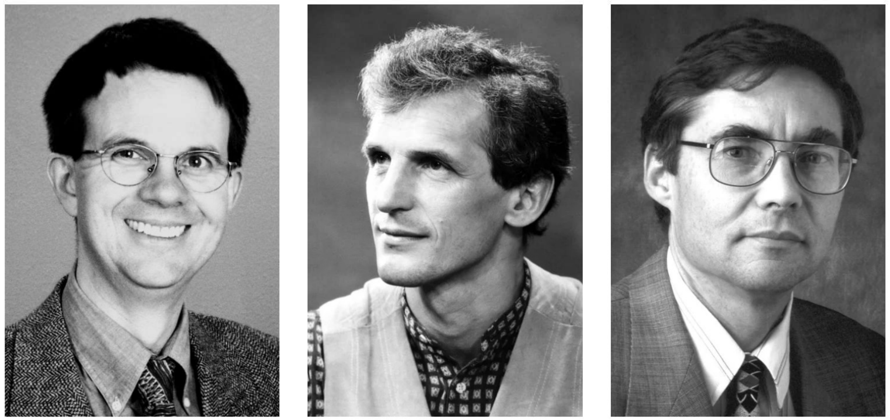
What is ultracold?



Access to Quantum Physics
&
Exquisite Control
&
Long Interrogation



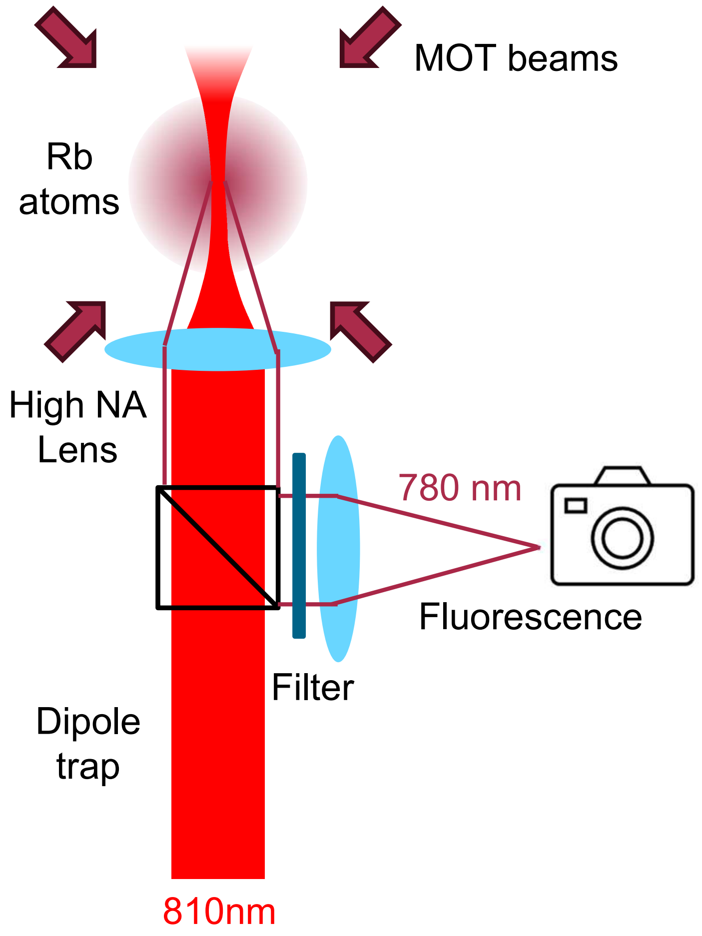
Nobel Prize in Physics 2001



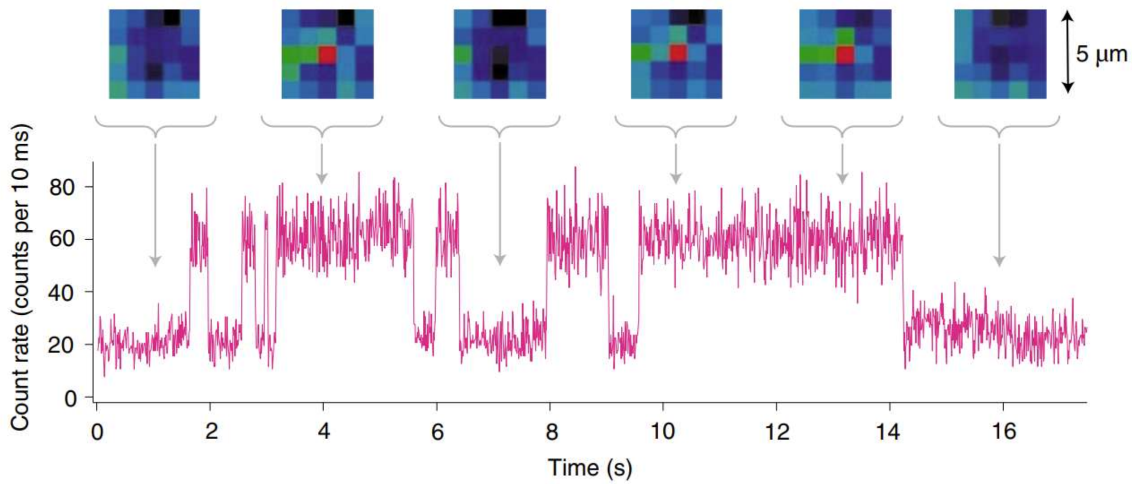
Cornell Ketterle Wieman

Single atom trapping

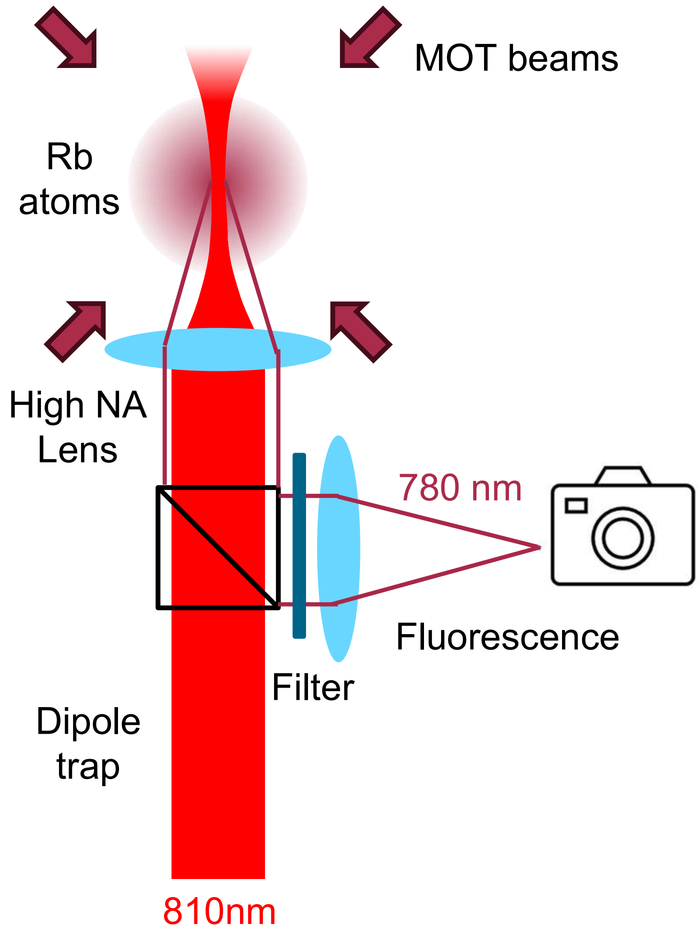
Sub-poissonian loading of single atoms in a microscopic dipole trap
Schlosser…Grangier, Nature **411**, 1024 (2001)



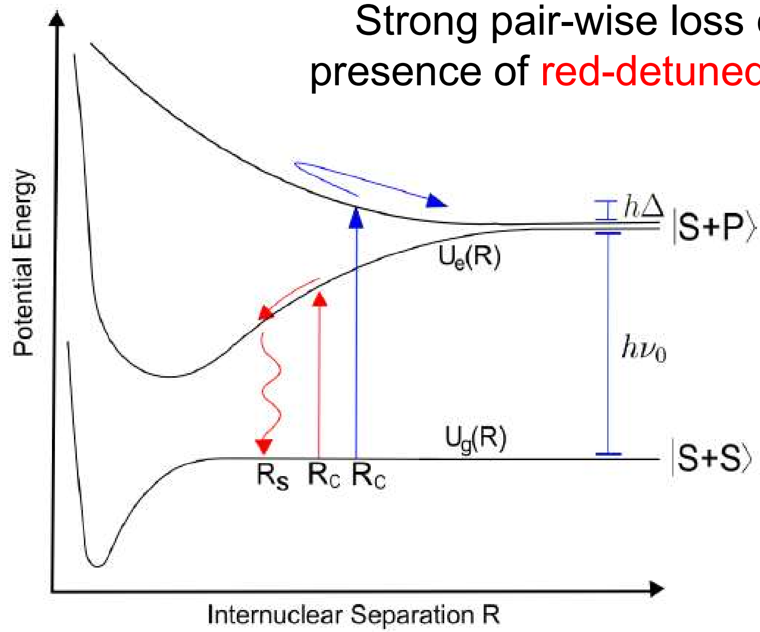
Single atom loading & detection:



Single atom trapping

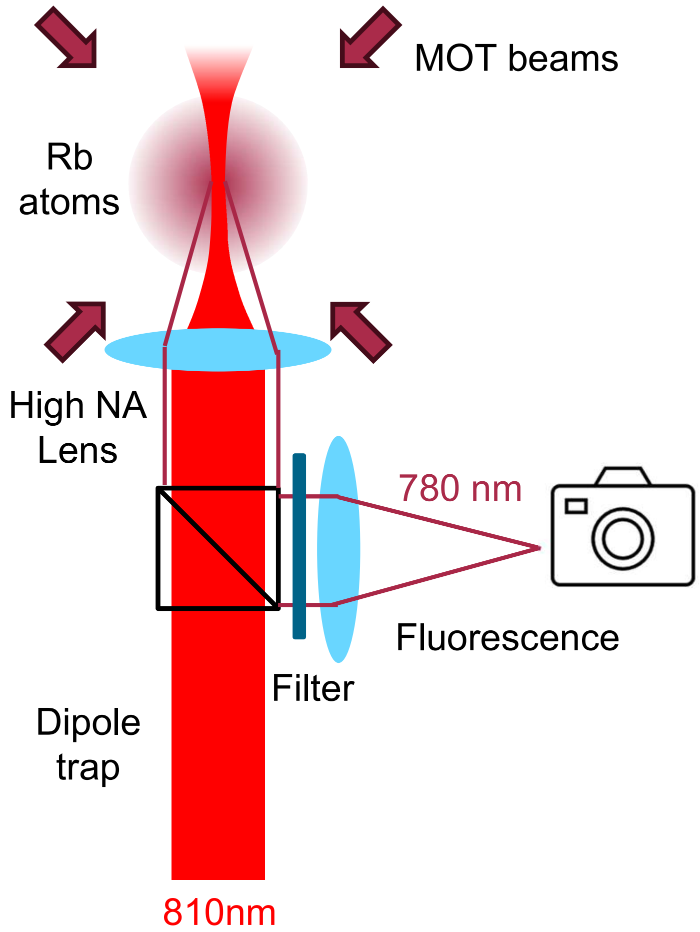


Strong pair-wise loss of atoms in presence of red-detuned cooling light

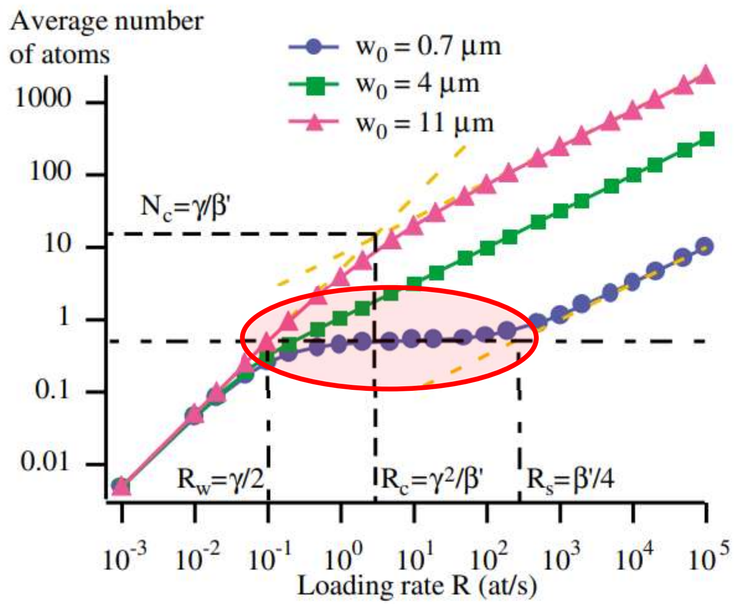


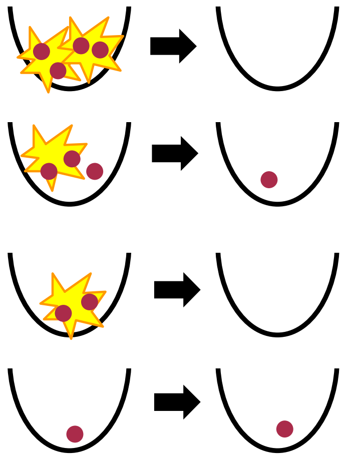
Two-atom collisions and the loading of atoms in microtraps
Andersen, Entropy **16**, 582 (2014)

Single atom trapping



If the trap is small enough leads to **collisional blockade**



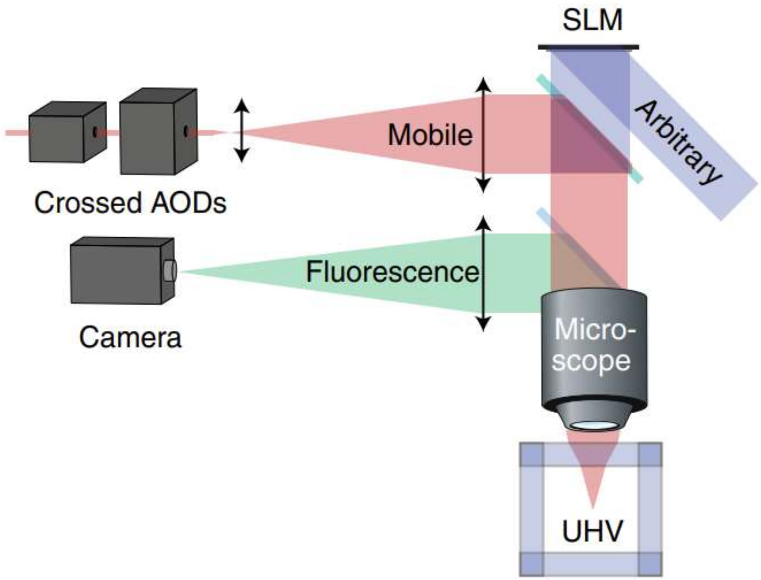


~50% loading probability

Collisional blockade in microscopic optical dipole traps
Grangier, Phys. Rev. Lett. **89**, 023005 (2002)

Now a mature technology…

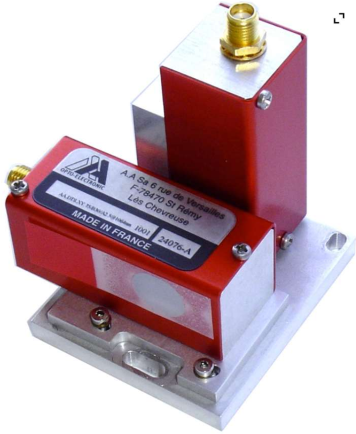
Standard setup:



Kaufman & Ni, Nature Physics **17**, 1324–1333 (2021)

Spatial Light Modulator (SLM)

- Hologram used to generate arbitrary array geometries.
- Aberration correction
- Focussing control

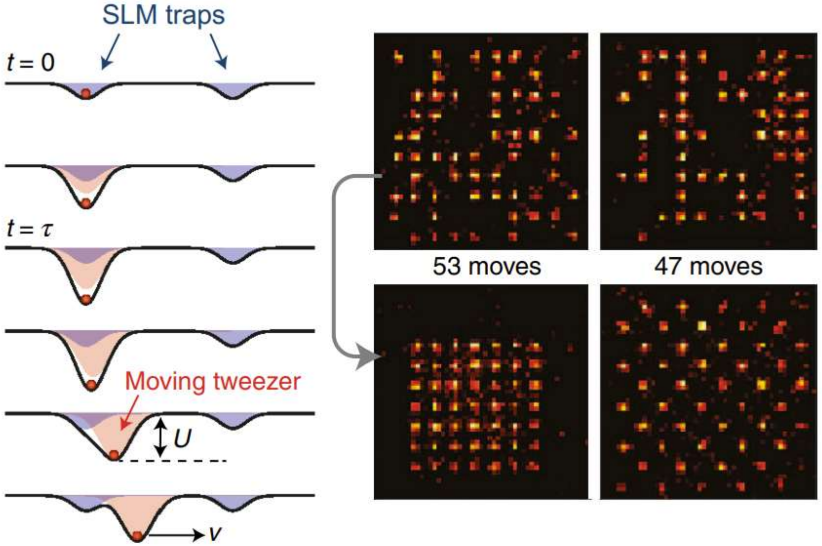


Crossed acousto-optic deflectors

- Each RF tone generates a trap
- 2D arrays
- Dynamic control via programmable arbitrary waveform generators

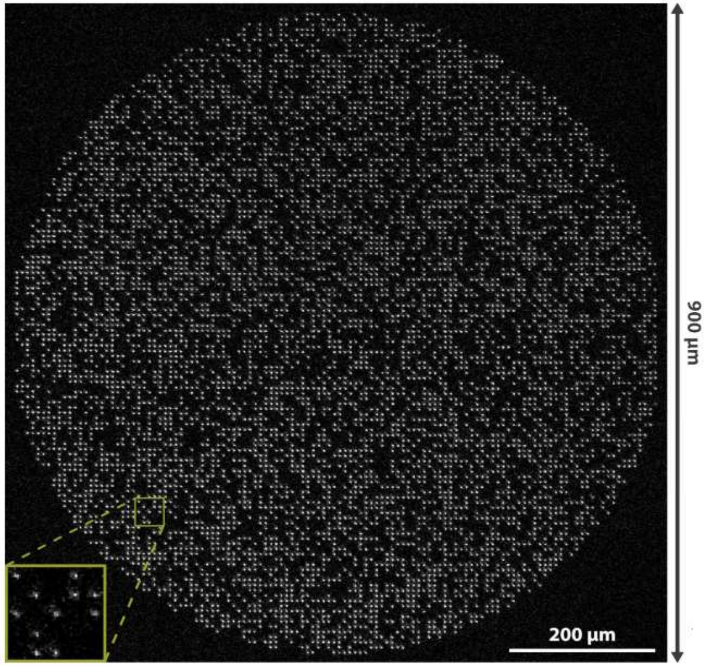
Now a mature technology

Reconfigurable geometries
= defect-free arrays



An atom-by-atom assembler of defect-free arbitrary two-dimensional atomic arrays
Barredo…Browaeys, Science **354**, 1021-1023 (2016)

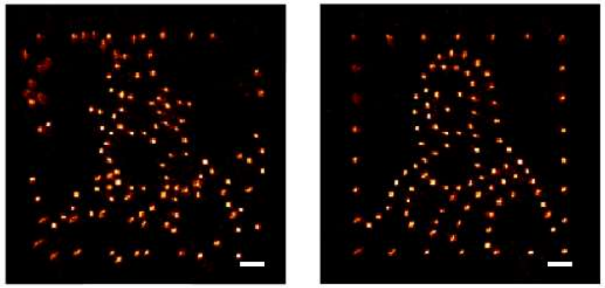
Scalable to large numbers



A tweezer array with 6100 highly coherent atomic qubits
Manetsch…Endres, Nature **647**, 60–67 (2025)

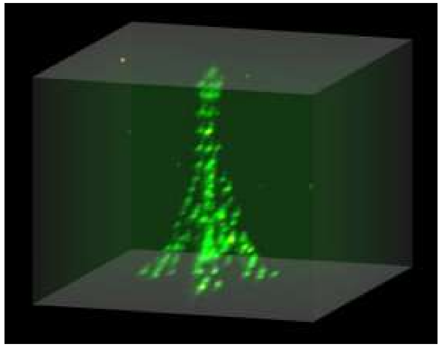
Now a mature technology

Truly arbitrary arrangement



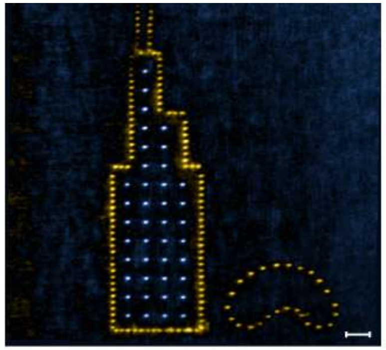
Schymik…Lahaye, PRA **102**, 063107 (2020)

+ Three dimensions



Barredo…Browaeys,
Nature **561**, 79-82 (2018)

+ Two species



Singh… Bernien,
PRX **12**, 011040 (2022)

Now a mature technology

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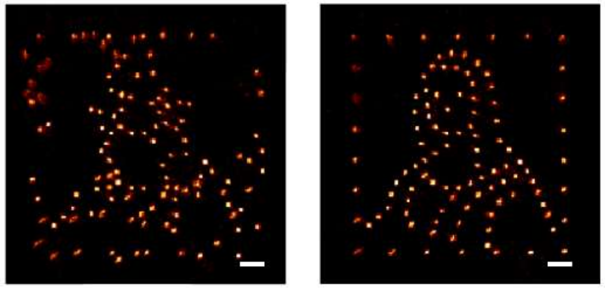

Schymik…Lahaye, PRA **102**, 063107 (2020)

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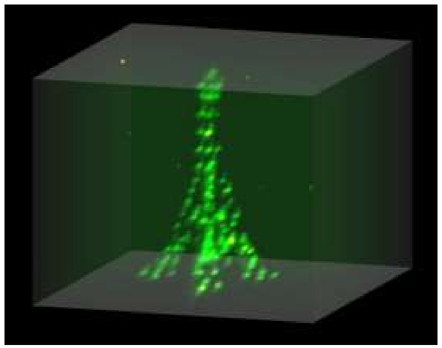

Barredo…Browaeys, Nature **561**, 79-82 (2018)

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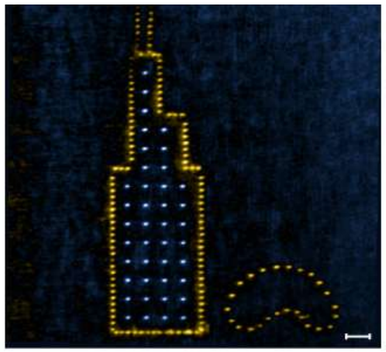
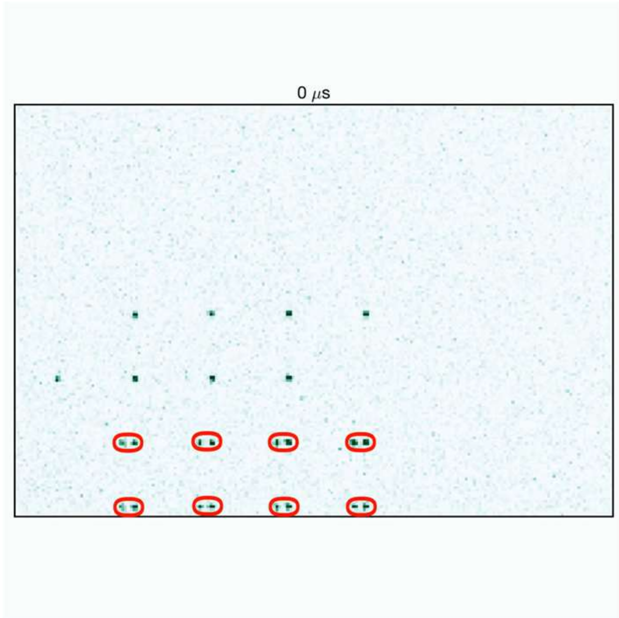


Singh… Bernien, PRX **12**, 011040 (2022)

Real-time dynamic control



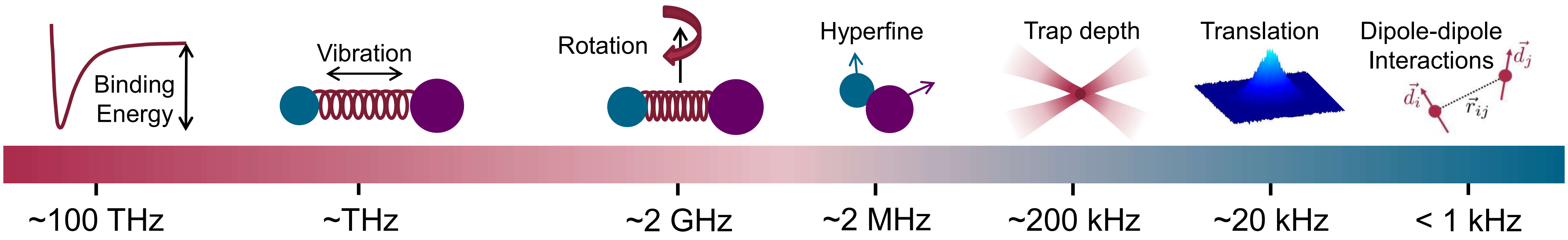
= Laser pulse to make the atoms Interact via Rydberg states

Bluvstein…Lukin, Nature **626**, 58-65 (2024)

Why molecules?

Rich internal structure

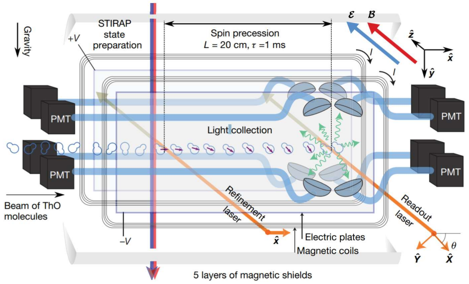
Adapted from Jin/Ye
Physics Today 2011



Why ultracold polar molecules?

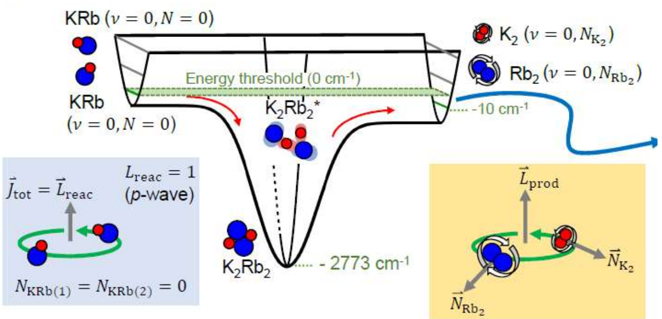
Durham University

Precision measurement



The ACME collaboration, Nature **562**, 355 (2018)

Ultracold Chemistry



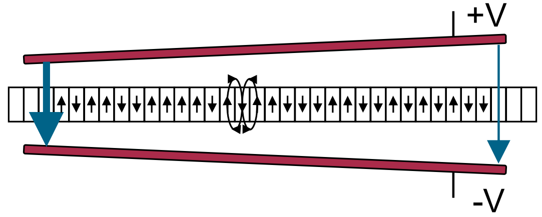
Liu, Nat. Phys. **16**, 1132 (2020)

Achieving full quantum control of molecules is important

Novel quantum fluids

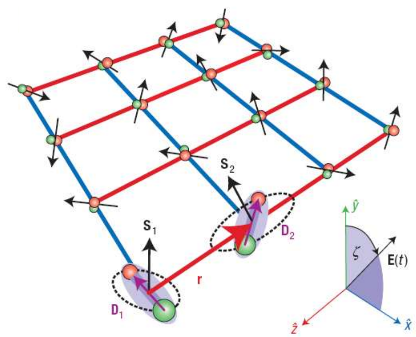
Schmidt, PRR **4**, 013235 (2022)

Quantum computation



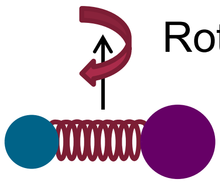
DeMille, PRL **88**, 067901 (2002)

Quantum simulation



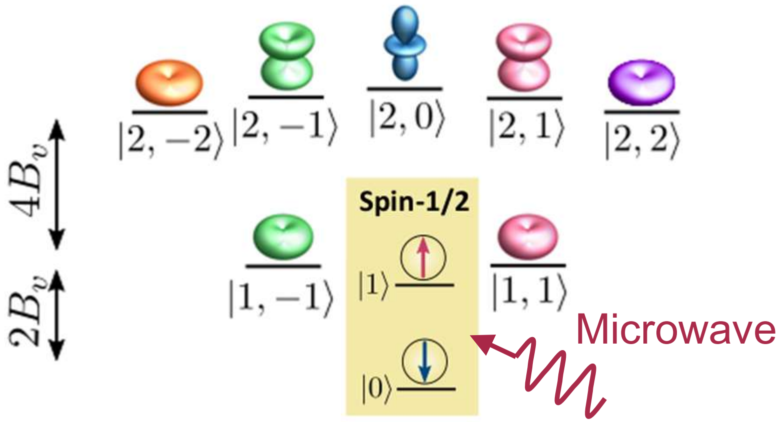
Micheli, Nat. Phys. **2**, 341 (2006)

Rotational states and dipolar interactions



Rotational degree of freedom results in ladder of long-lived states

$$E_N = B_v N(N+1)$$

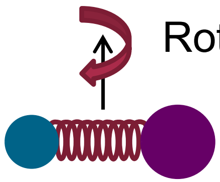


Encode and manipulate spins

Electric dipole transitions connect states with $\Delta N = 1$ and $\Delta M_N = \pm 1$

Rotational states and dipolar interactions

Durham University



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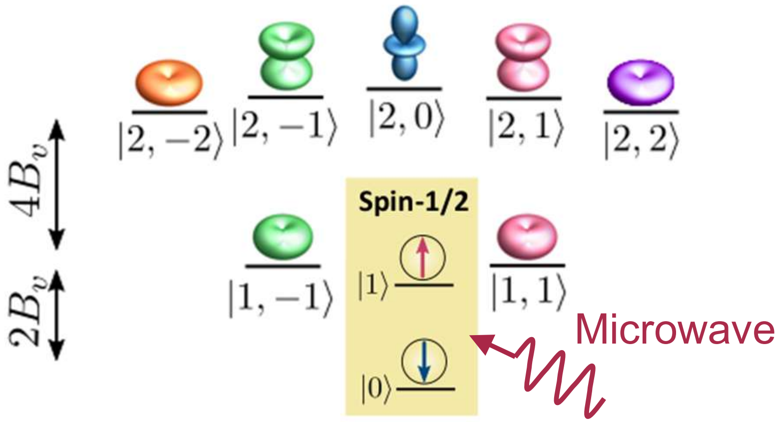


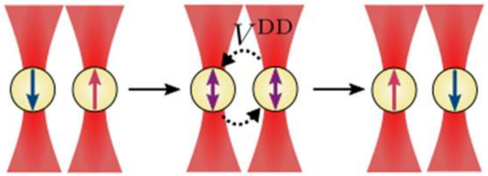
Encode and manipulate spins

Electric dipole transitions connect states with $\Delta N = 1$ and $\Delta M_N = \pm 1$

Electric dipole moment points along internuclear axis
For rotational eigenstates - ***zero in the lab frame!***

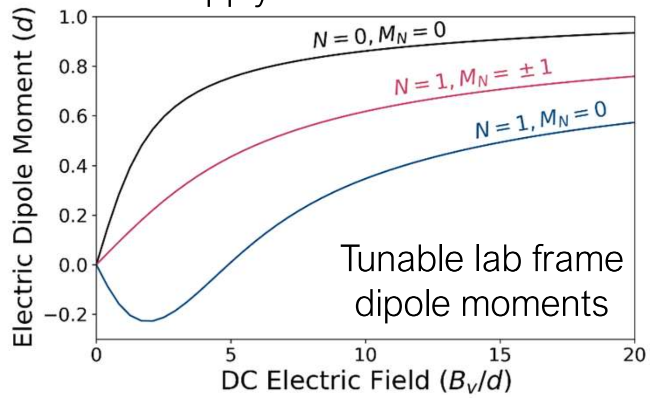
But can engineer dipole-dipole interactions

Via transition dipole moment between rotational states



Spin-exchange interactions

Apply an electric field



Tunable lab frame dipole moments

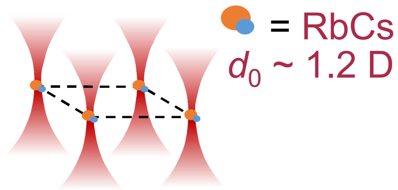
$$\hat{H}_{ij} = \frac{1-3\cos^2\theta_{ij}}{|\mathbf{r}_i - \mathbf{r}_j|^3} \begin{pmatrix} d_\uparrow^2 & 0 & 0 & 0 \\ 0 & d_\downarrow d_\uparrow & d_{\downarrow\uparrow}^2 & 0 \\ 0 & d_{\downarrow\uparrow}^2 & d_\downarrow d_\uparrow & 0 \\ 0 & 0 & 0 & d_\downarrow^2 \end{pmatrix} \begin{matrix} \uparrow\uparrow \\ \uparrow\downarrow \\ \downarrow\uparrow \\ \downarrow\downarrow \end{matrix}$$

(resonant) spin exchange
static dipoles

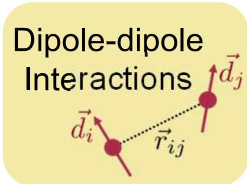
Outline

❶ A (very) brief guide to the association of molecules

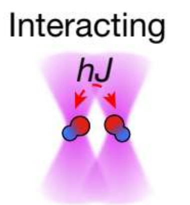
❷ Assembly & control of RbCs molecules in optical tweezers



❸ Engineering long rotational coherences in magic traps



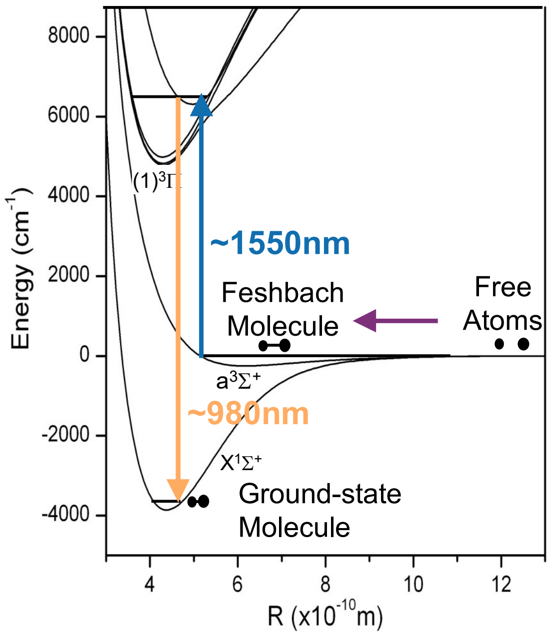
❹ Entanglement of pairs of molecules in magic tweezers



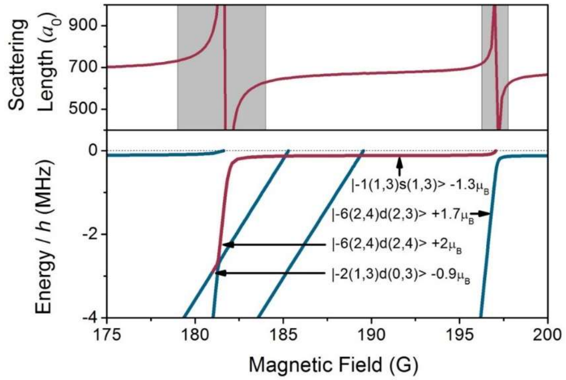
Brief guide to making RbCs molecules (in bulk gases)

Durham University

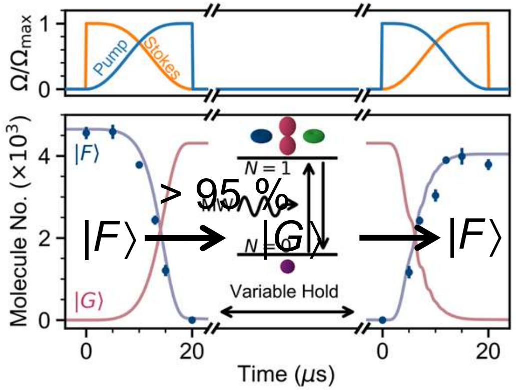
Start with Rb + Cs atoms



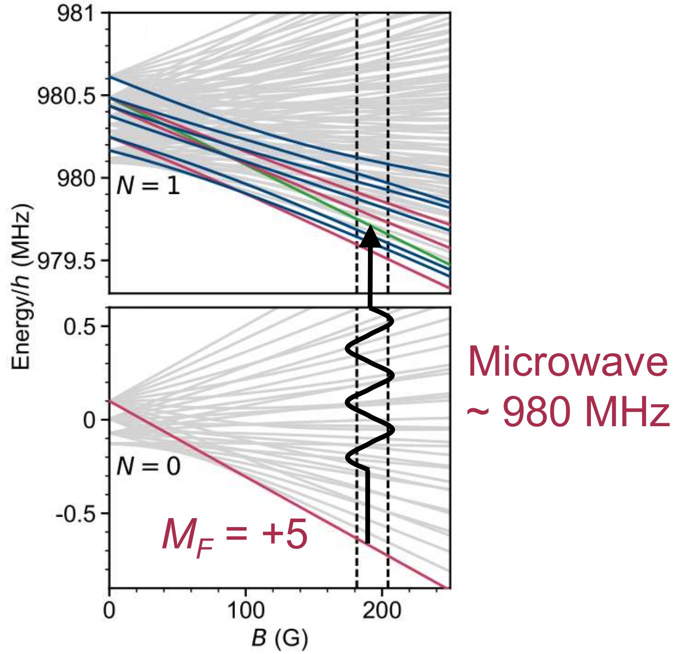
Magnetoassociation



STIRAP at ~181 G



Transfer to the hyperfine ground state

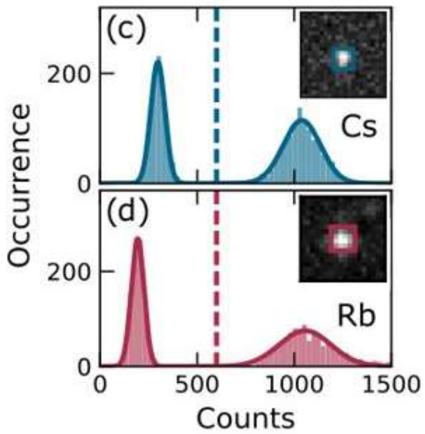


Return STIRAP
= state sensitive
detection

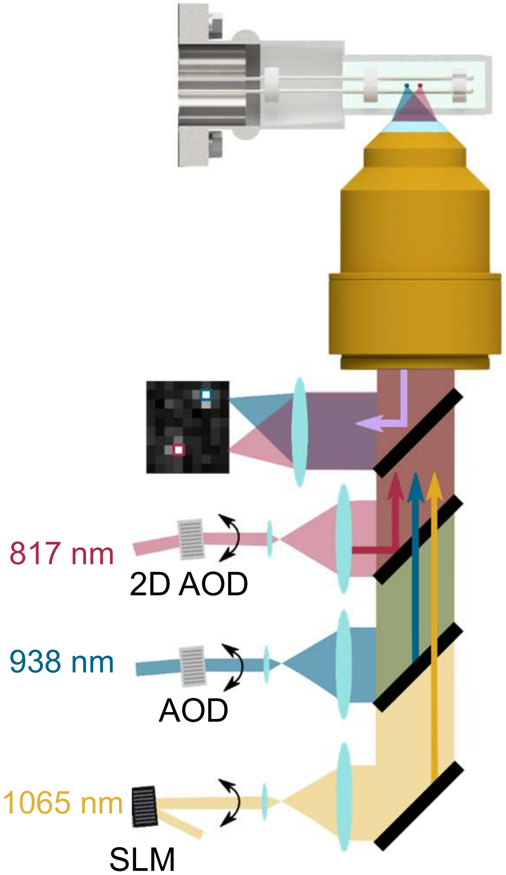
Takekoshi et al., PRL **113**, 205301 (2014)
Molony et al., PRL **113**, 255301 (2014)

Single molecules in optical tweezers

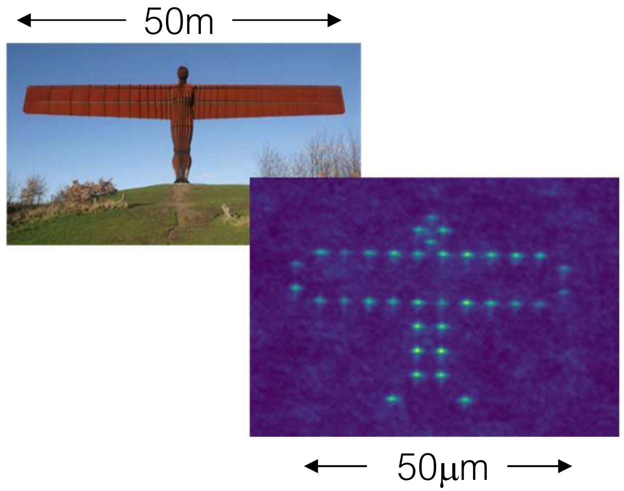
Durham University



Fluorescence imaging to detect tweezer occupation (fidelity >> 99.9%)

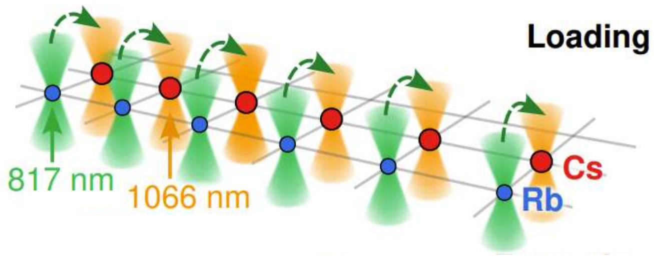


Inspired by experiments in the Ni group at Harvard

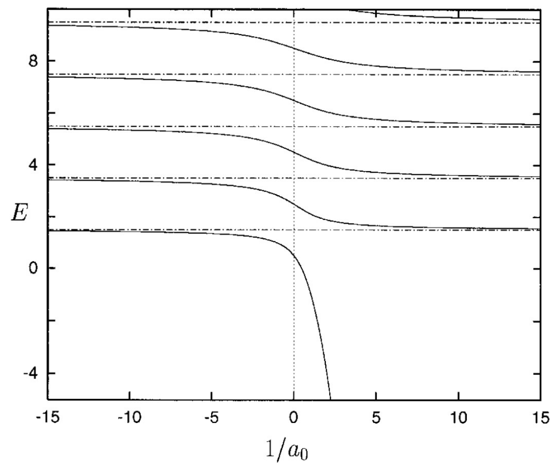


Brooks *et al.*, *NJP* **23** 065002 (2021)
Spence *et al.*, *NJP* **24** 103022 (2022)

Molecular assembly in optical tweezers

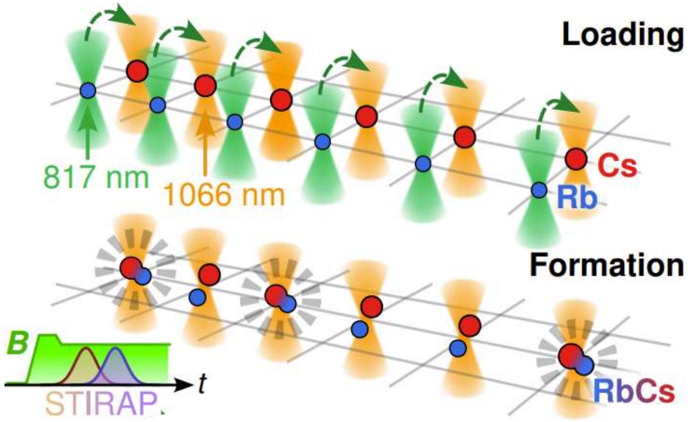


- Load, image, rearrange Rb & Cs
- Raman sideband cooling to 3D ground state
- Merge traps



Busch et al.,
Foundations of Physics
28, 549 (1998)

Molecular assembly in optical tweezers



- Load, image, rearrange Rb & Cs
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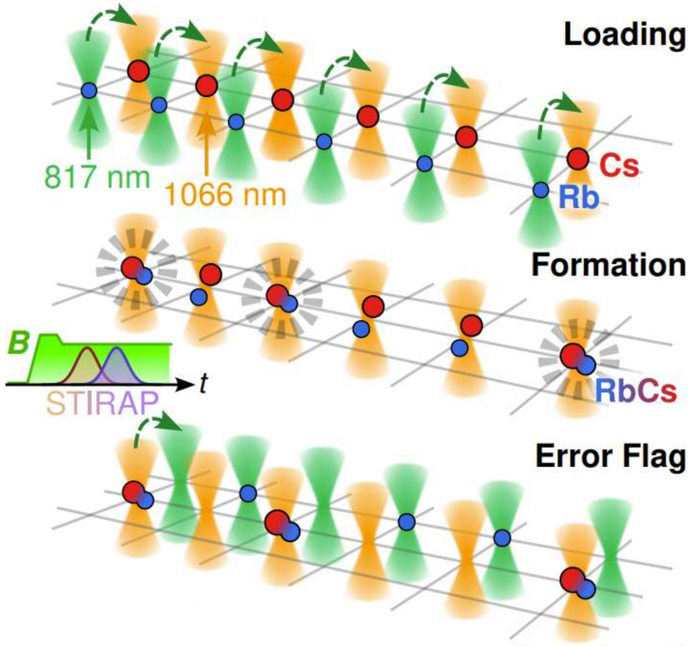
- Magnetoassociation & STIRAP
- Atom pairs remain on sites where formation failed

Molecular assembly in optical tweezers



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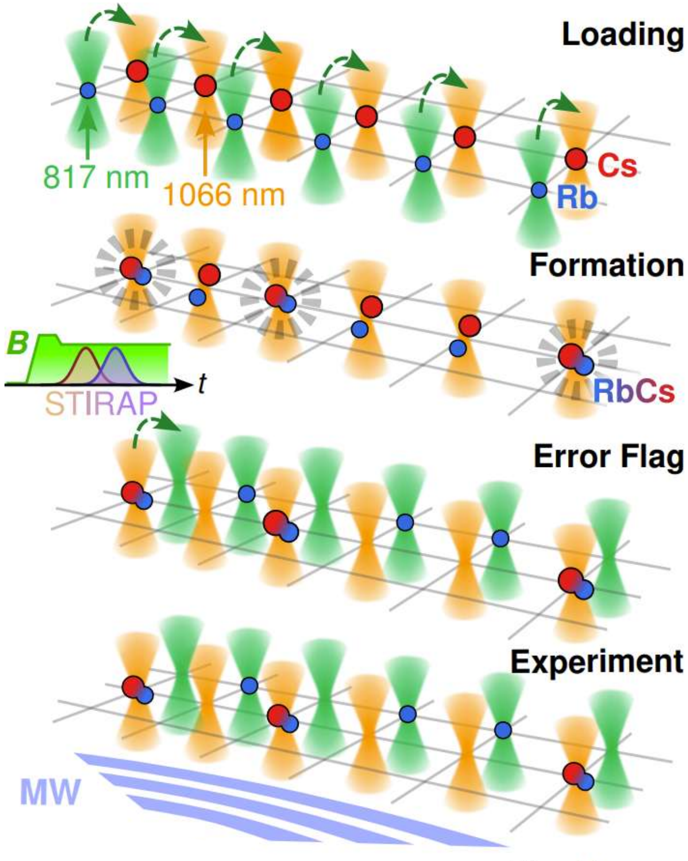
- Pullout Rb using 817nm tweezer
- Blow away Cs

Molecular assembly in optical tweezers



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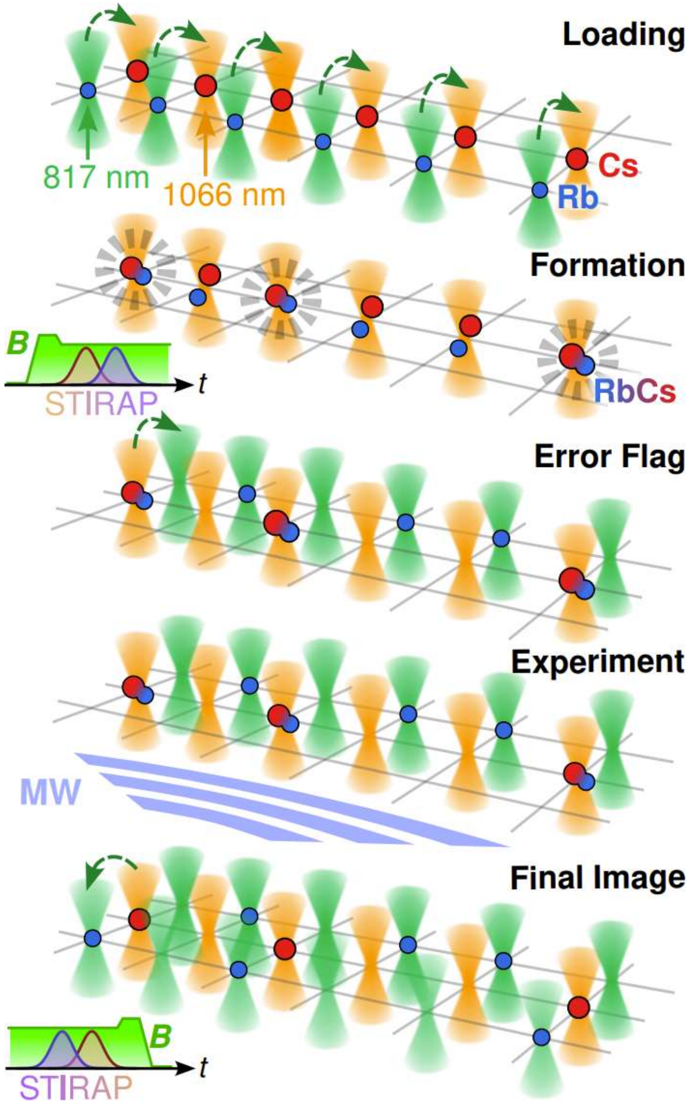
Molecular assembly in optical tweezers



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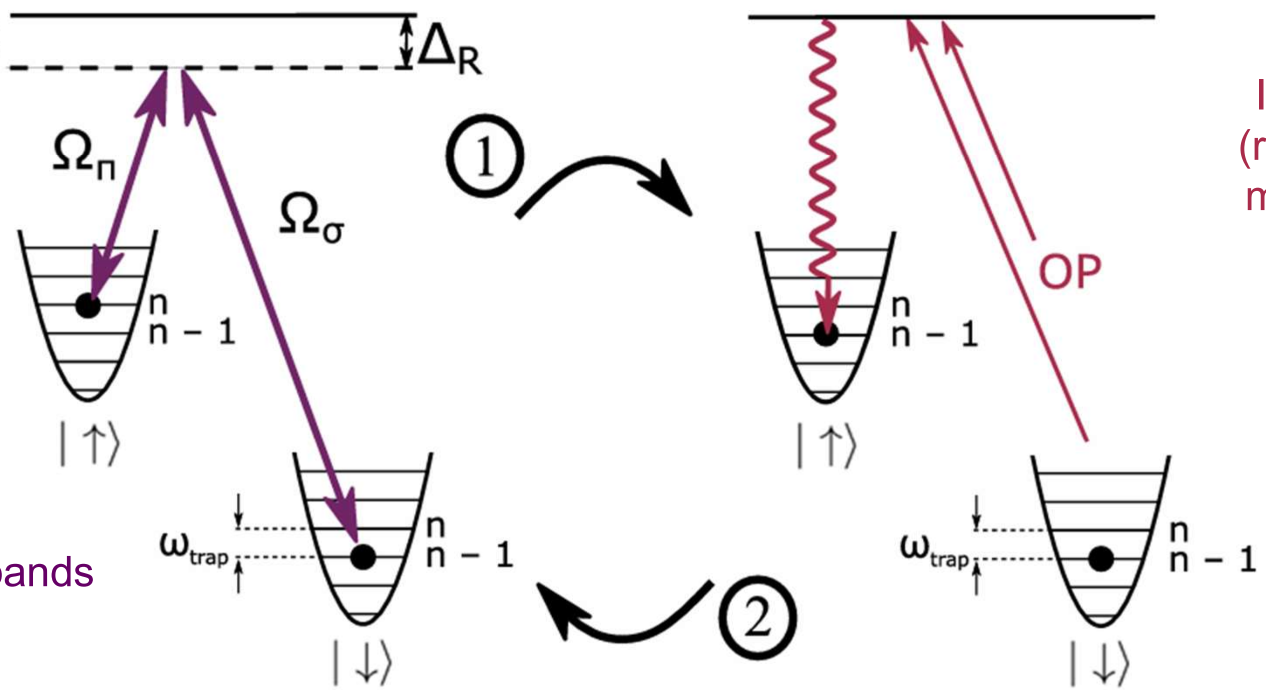
- Perform experiment

- Recover & image atoms
- Post select on recovery of Rb and Cs
- Ignore sites with an error flag

Simultaneous Raman sideband cooling

Raman transition → Optical pumping → Repeat



In Lamb-Dicke regime
(recoil energy less than
motional state spacing)
n does not change

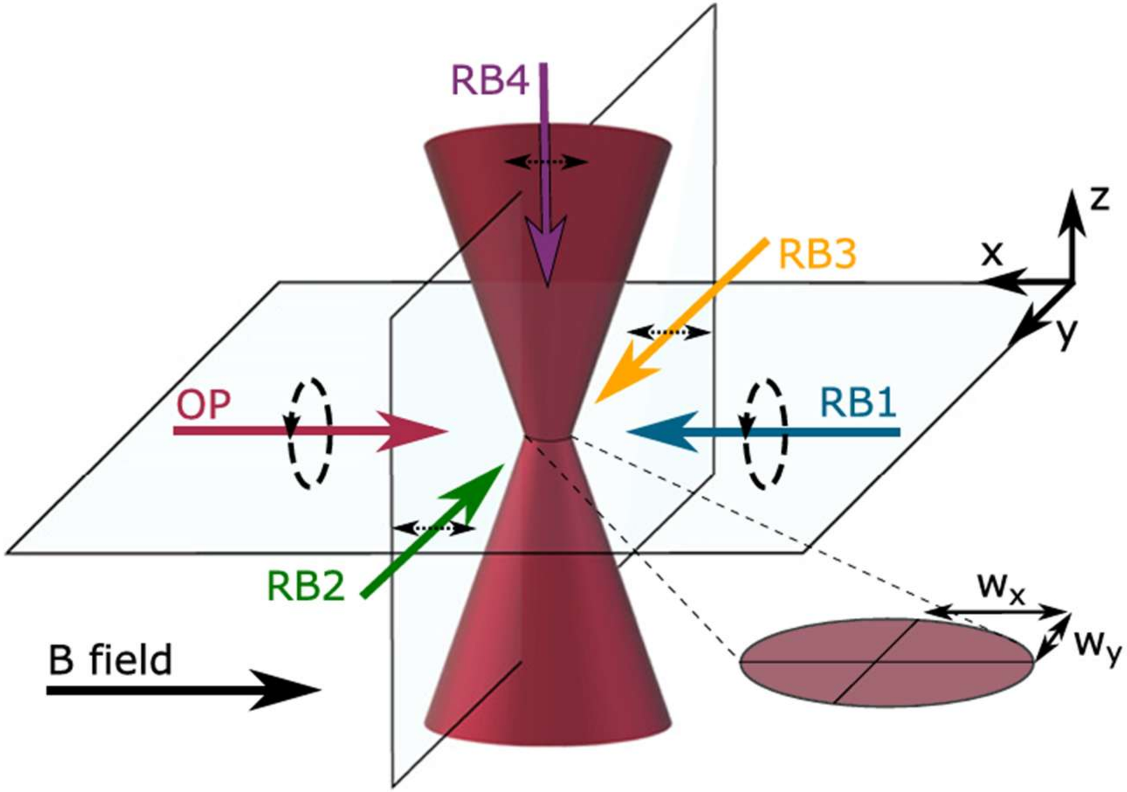
Narrow linewidth
resolves motional sidebands

Spence *et al.*, *NJP* **24** 103022 (2022)

Each cycle removes one quanta of motional energy

Simultaneous Raman sideband cooling

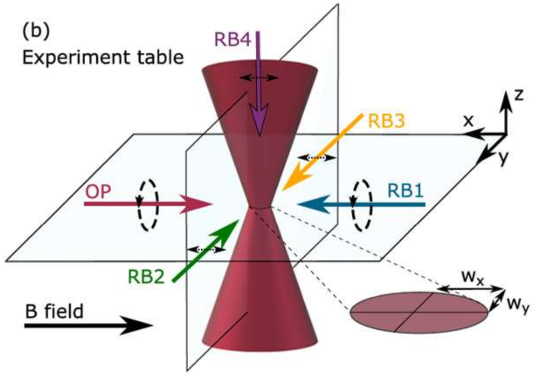
Use three pairs of Raman beams – one for each axis
(RB1 is common to all)

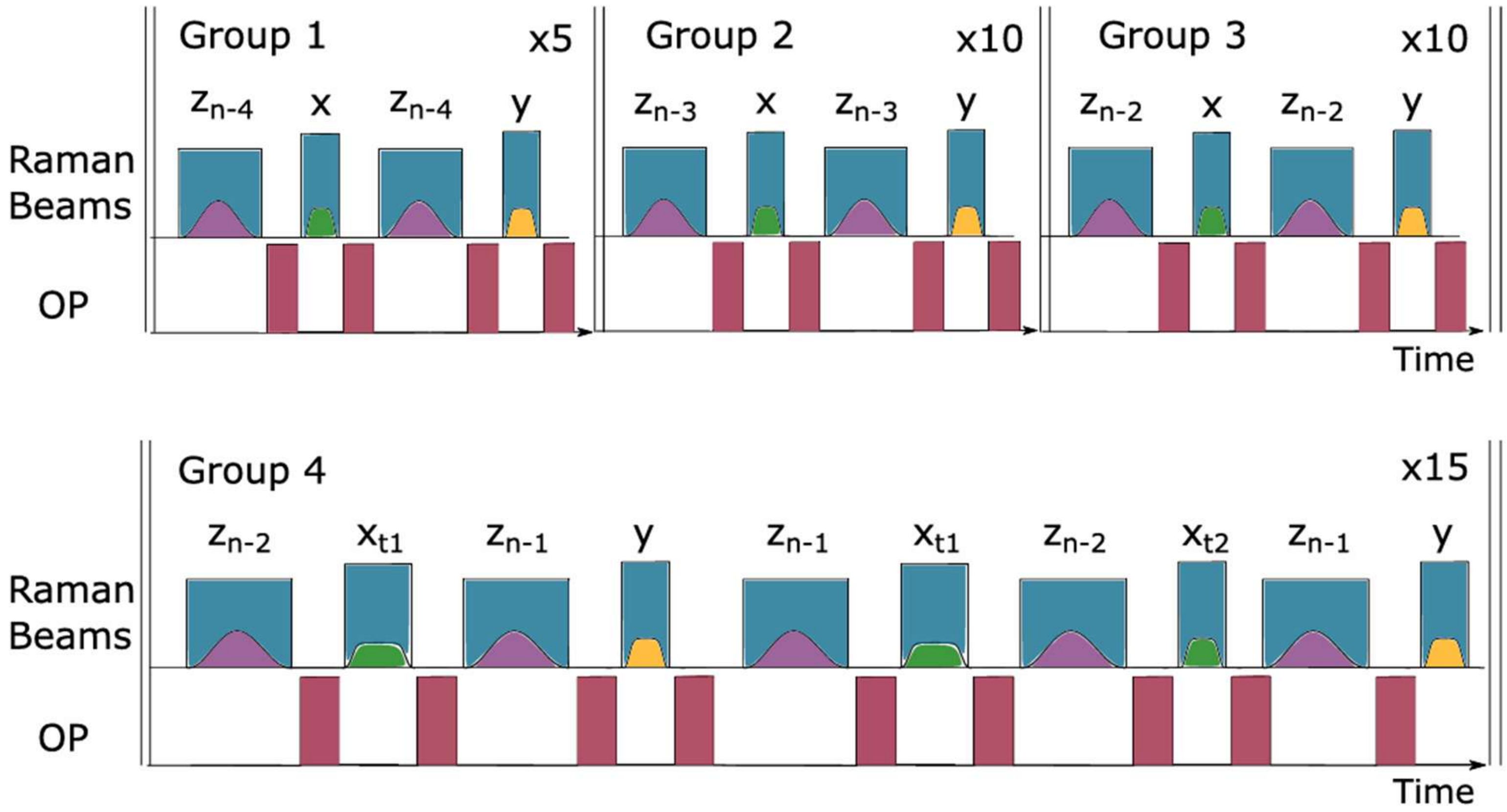


Trap is a little elliptic due to clipping before objective

Simultaneous Raman sideband cooling

Apply a complex pulse sequence interleaving the different axes

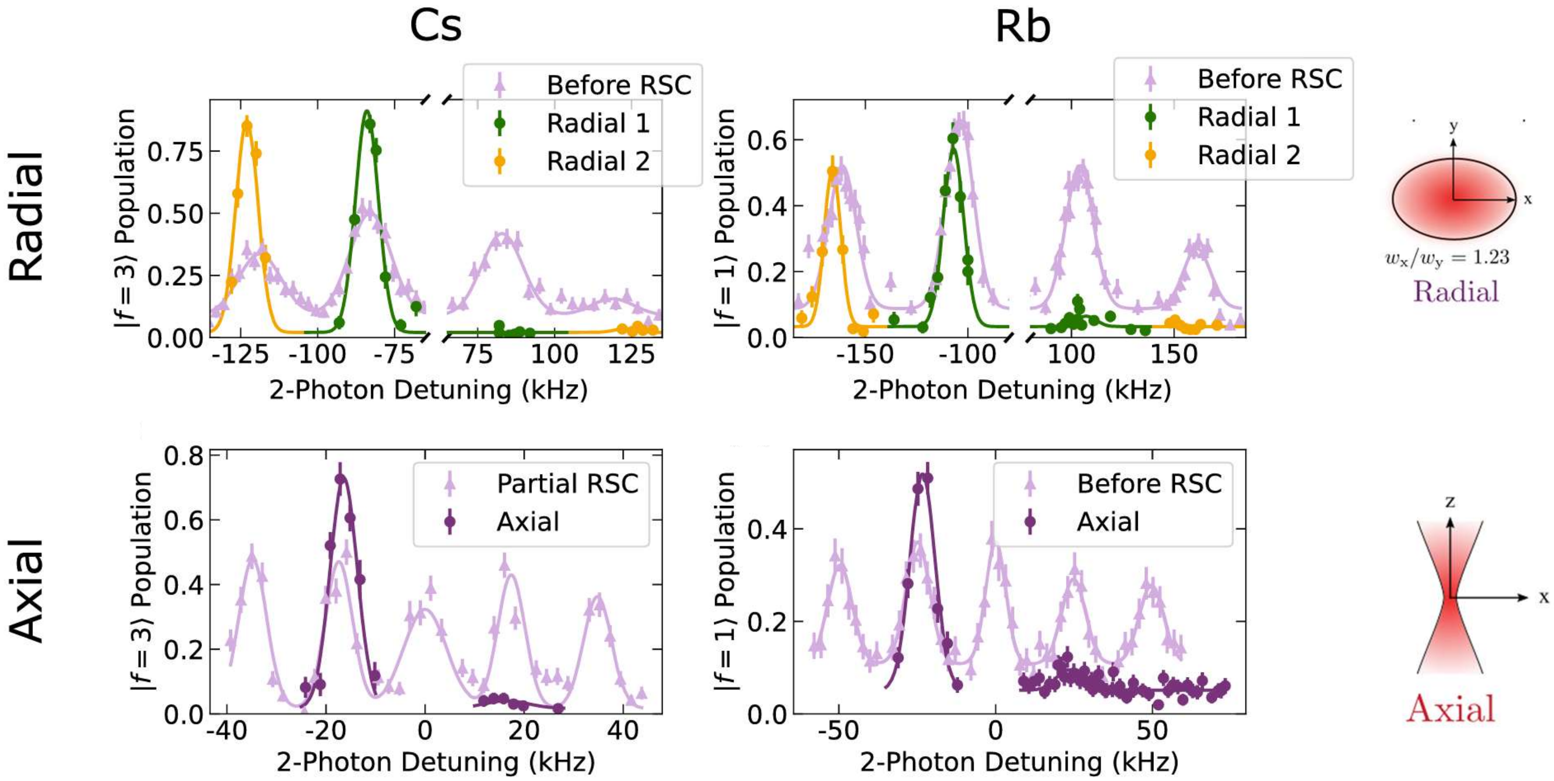




Simultaneous Raman sideband cooling

Compare height of red and blue sidebands (in ground state no red sideband)

After 45 ms of cooling, the probabilities to occupy 3D motional ground state are
86(4)% for Rb and 95(3)% for Cs

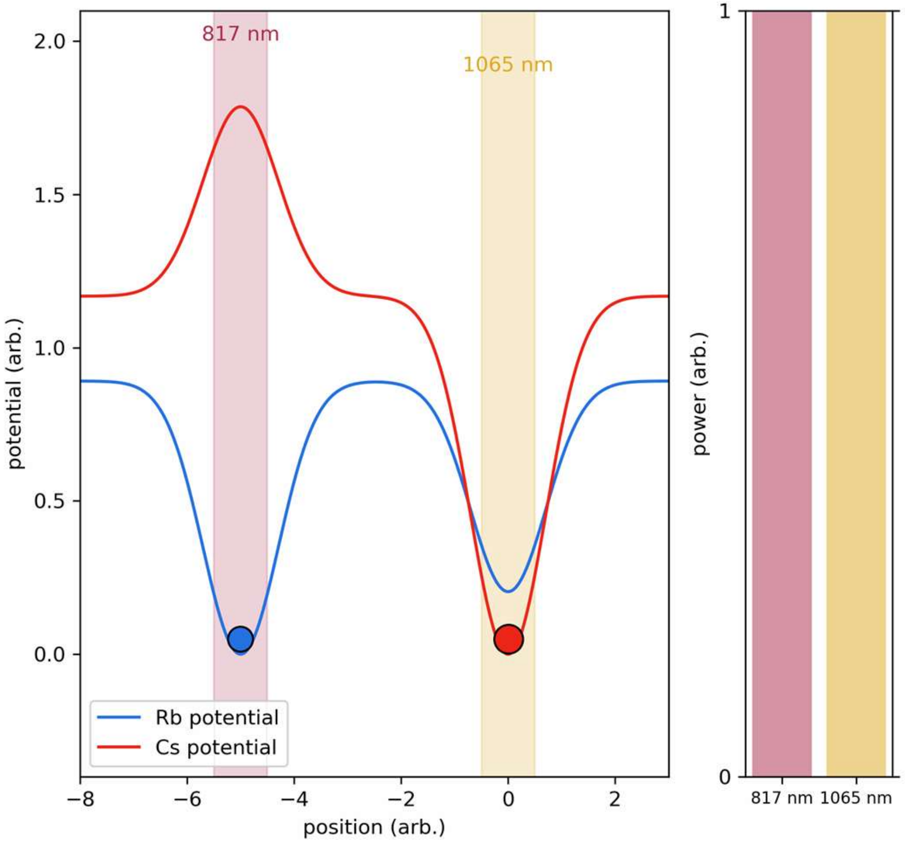


Spence *et al., NJP* **24** 103022 (2022)

Final temperature balance of cooling and heating

Merging Tweezers

Durham University



Merging optimised to maintain high population in ground state of relative motion

Estimate >60% of atom pairs are in (1,1)(3,3) and $n_{rel}=0$

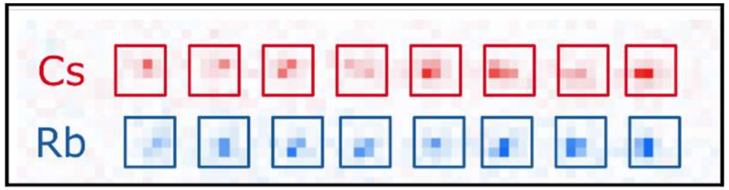
New method of forming molecules

Ruttley & Guttridge *et al.*, PRL **130**, 223401 (2023).

“Mergoassociation”

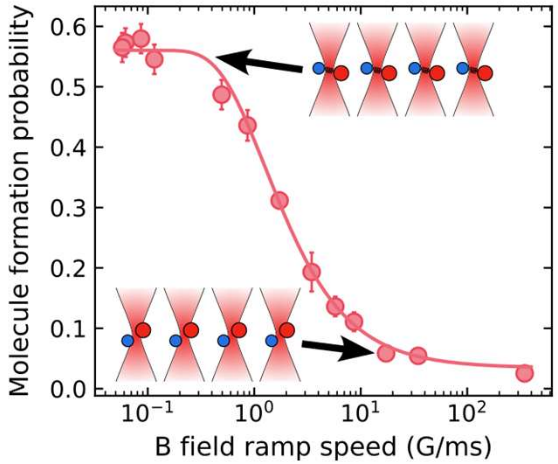
Formation of arrays of molecules

Typically load 8-12 tweezers



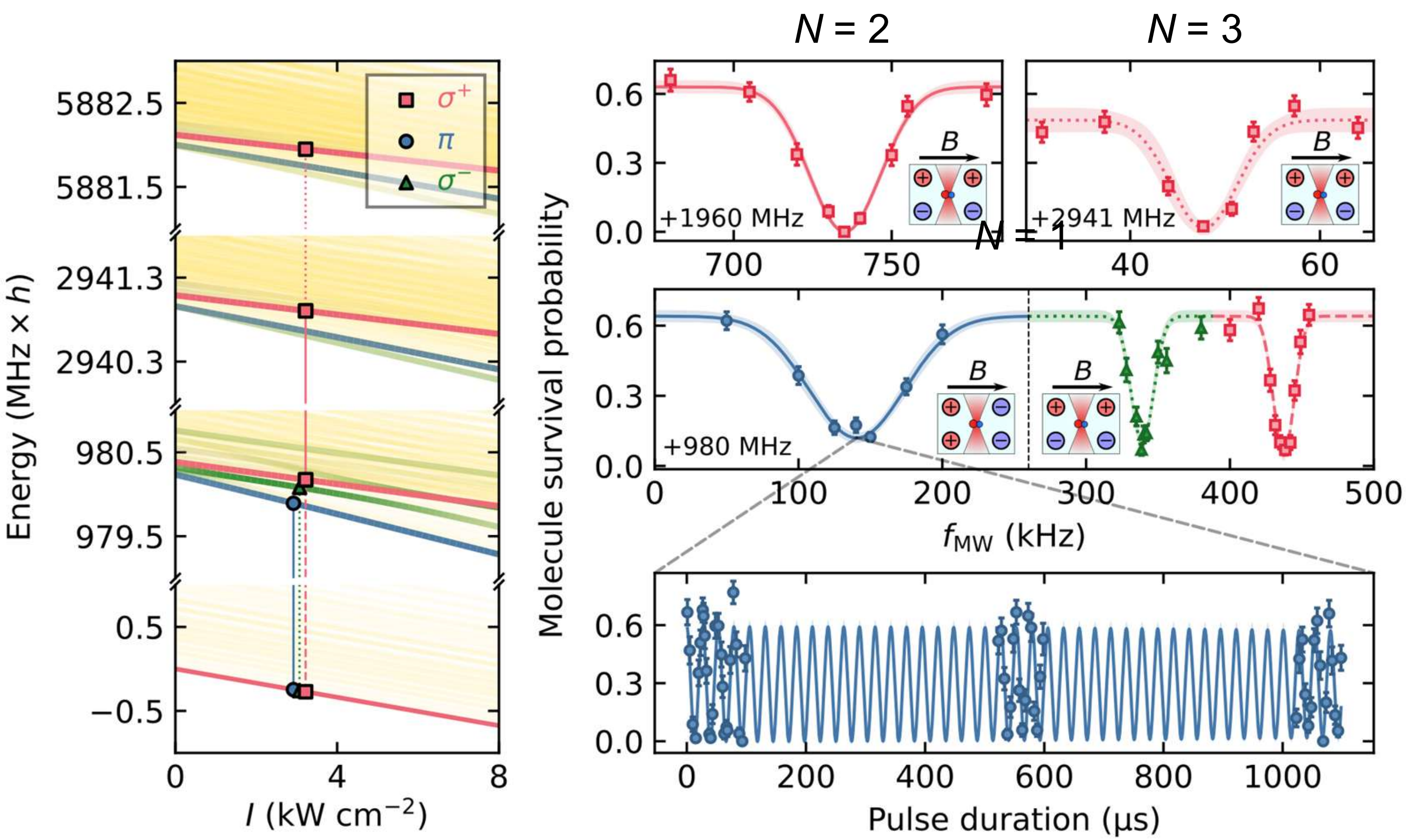
Use 1D rearrangement of Rb and Cs

Molecule formation budget	
Atom pair state preparation	0.93(3)
Relative motional ground state occupancy after merging	0.56(5)
Magnetoassociation	>0.99



Maddox, Mortlock et al., PRL 133, 253202 (2024)

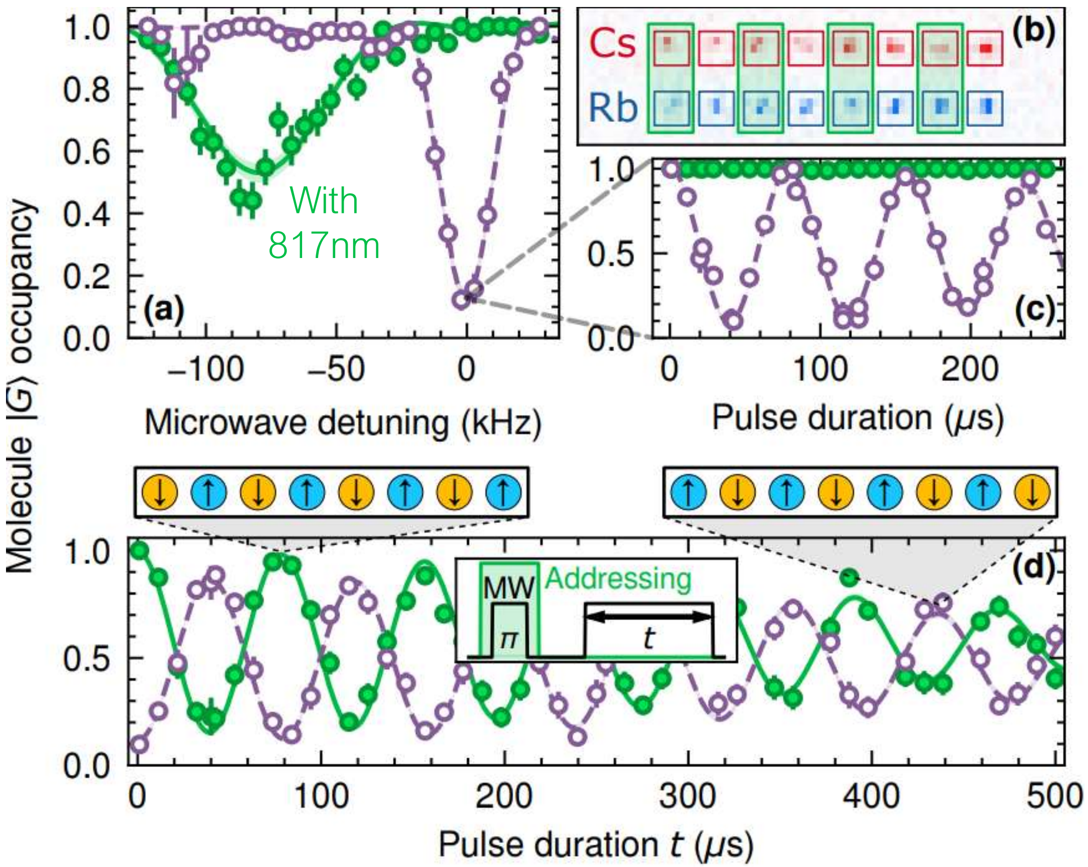
MW Spectroscopy



Builds upon spectroscopy in bulk gases: Gregory et al. Phys. Rev. A 94, 041403(R) (2016)

Microwave transitions & single-site addressing

Add 817nm tweezer to AC Stark shift sites off resonance, global microwave pulse addresses remaining sites

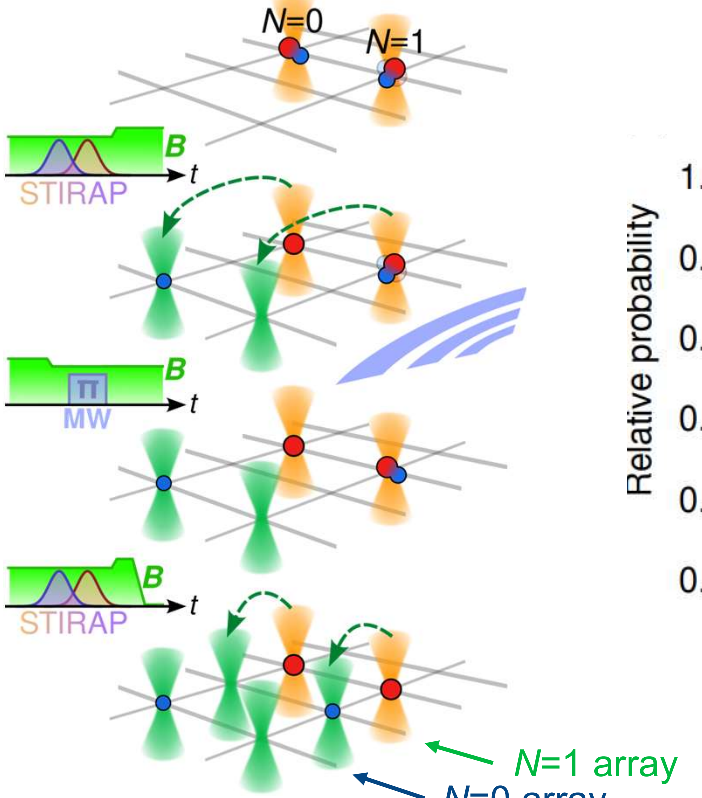


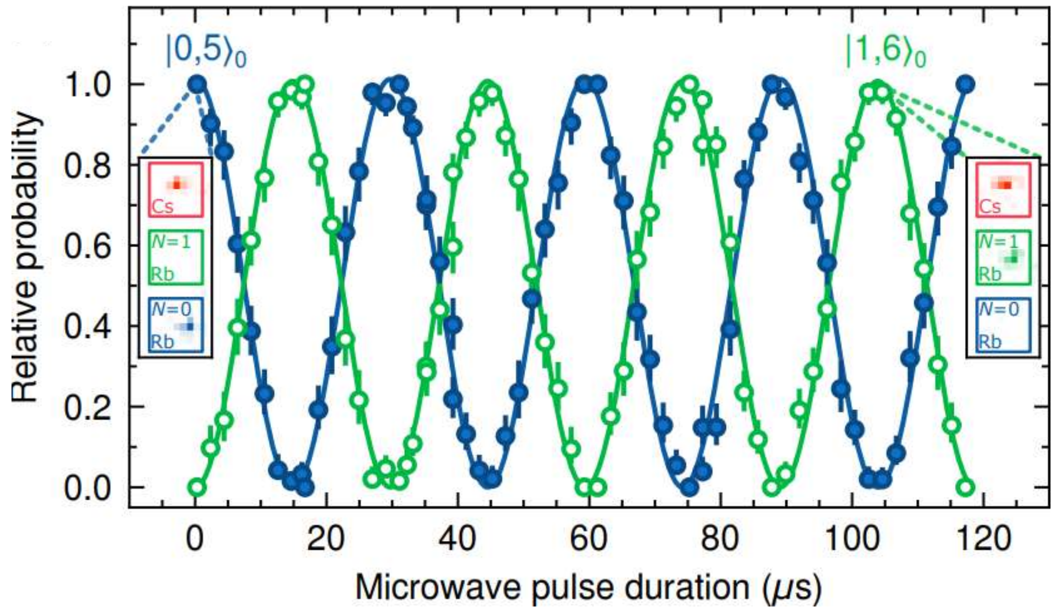
Ruttley et al., PRX Quantum 5, 020333 (2024)
see also Picard et al., PRX Quantum 5, 020344 (2024)

Microwave transition with multi-state readout

Multistate readout key to microwave measurements

Mitigates leakage errors and loss/formation errors that would otherwise look like excitation to $N=1$



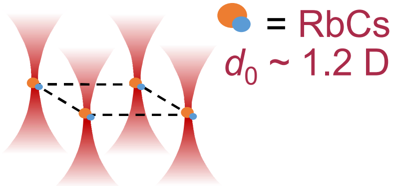


Ruttley et al., PRX Quantum 5, 020333 (2024)
see also Picard et al., PRX Quantum 5, 020344 (2024)

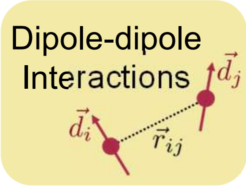
Outline

❶ A (very) brief guide to the association of molecules

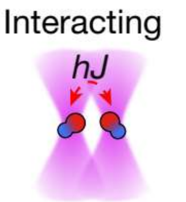
❷ Assembly & control of RbCs molecules in optical tweezers



❸ Engineering long rotational coherences in magic traps



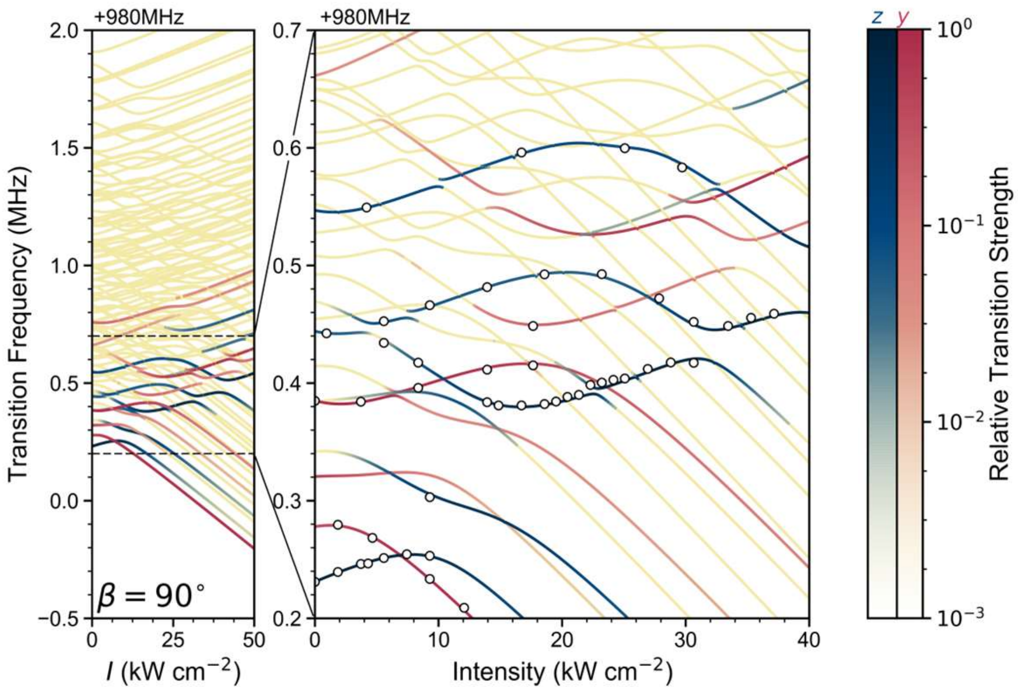
❹ Entanglement of pairs of molecules in magic tweezers



Overcoming differential AC Stark shifts

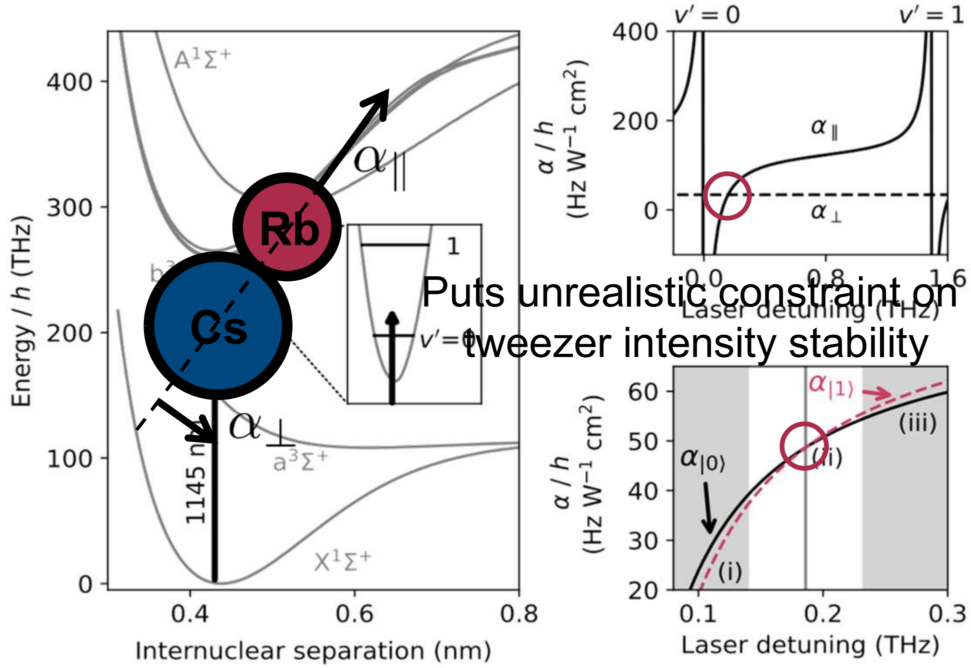
Resonant spin-exchange requires the elimination of differential AC Stark shifts

But anisotropic polarizability a problem:



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

Solution: a magic-wavelength trap

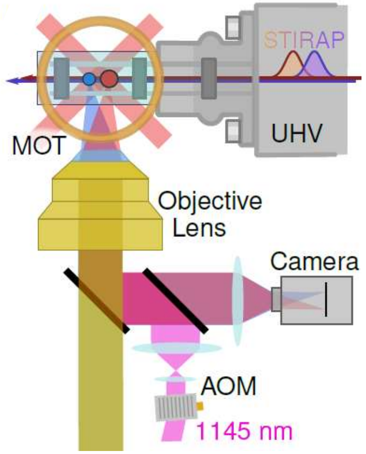


Puts unrealistic constraint on tweezer intensity stability

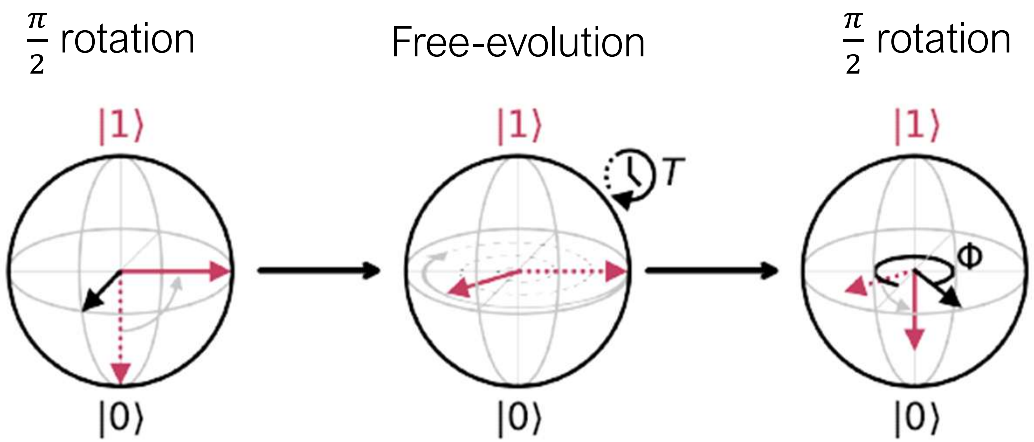
Guan, Cornish & Kotochigova PRA **103**, 043311 (2021)
Gregory *et al.*, Nature Physics 20, 415 (2024)

Magic-wavelength tweezers

Added magic tweezer into imaging path – waist 1.87(5) μm. Laser now locked to ULE cavity.
Find magic detuning on $(N=0, M_N=0) \rightarrow (1,1)$ transition using Ramsey spectroscopy.



Use a Ramsey sequence to precisely measure AC Stark shifts



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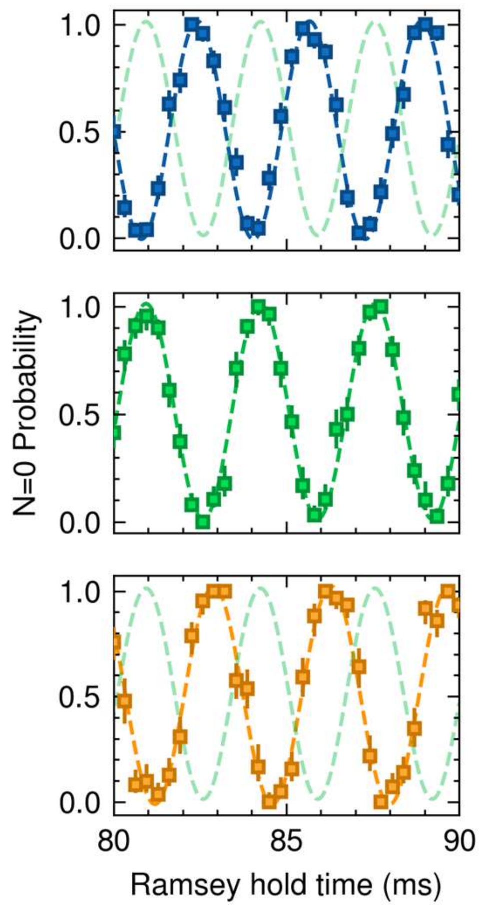
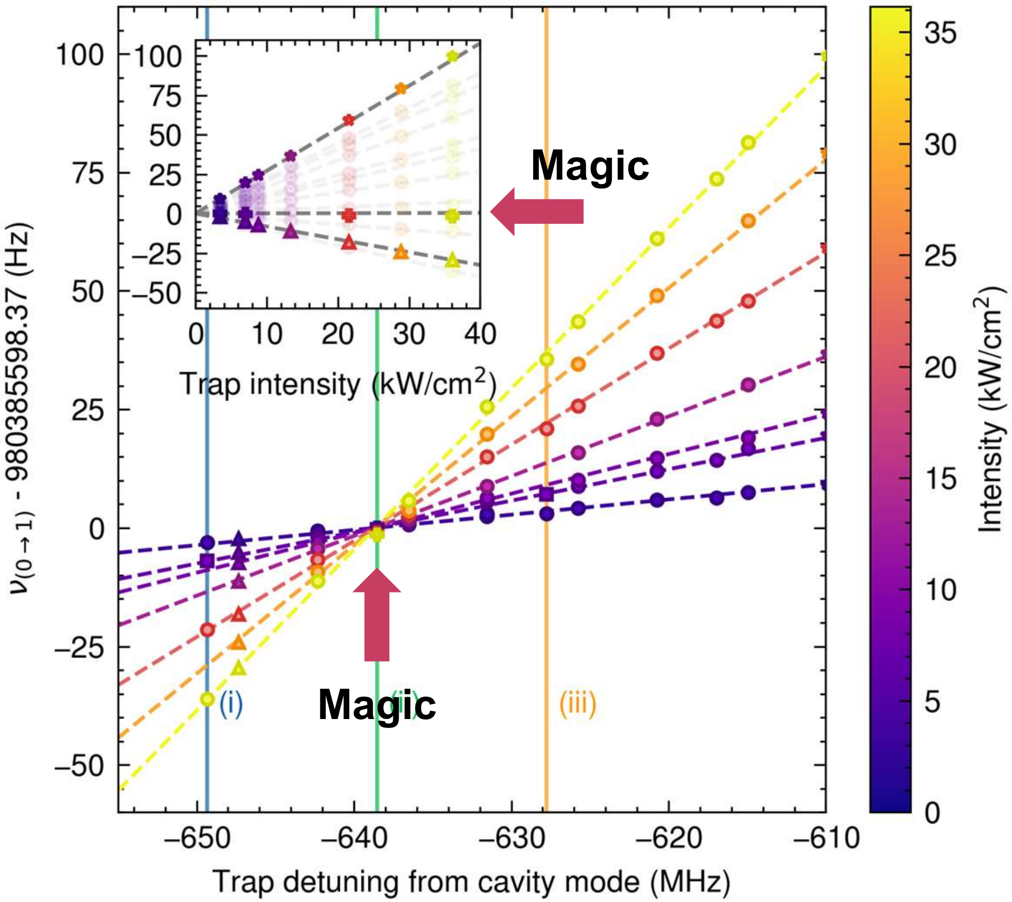
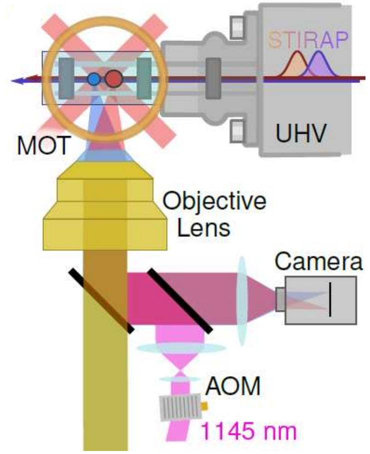






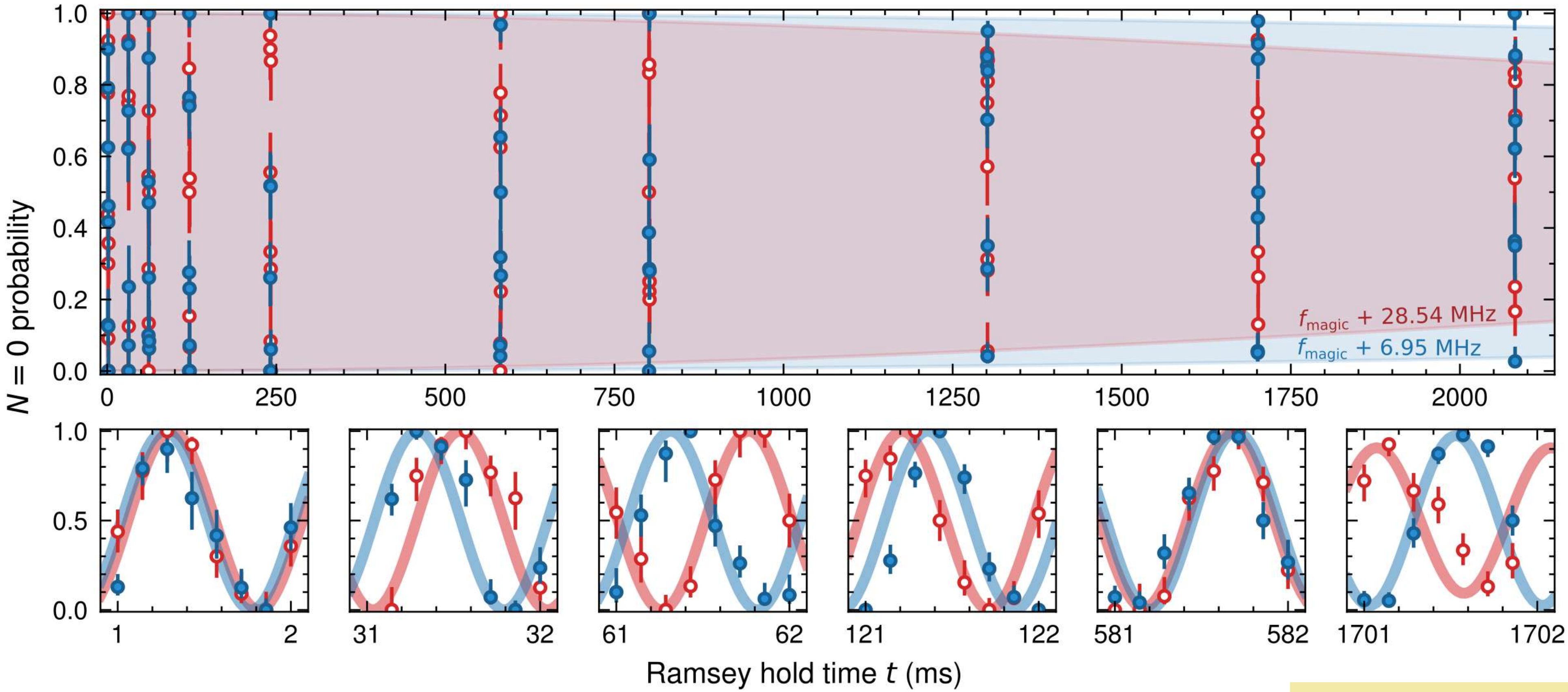
Sensitivity
93.9(3) mHz / MHz/(kW/cm²)

Trap detuning and intensity

Typical coherence assuming 1 MHz detuning, 4 kW/cm² and 4% intensity variation:
~66 seconds!

What about rotational coherence?

Two tweezers 8 μm apart so no interactions. Depth = 4 μK (3.6kW/cm^2). Lifetime ~ 10(2) s.
(Blue f_{magic}+6.95 MHz, Red f_{magic} +28.4 MHz) → ~6 Hz difference in transition frequencies



No spin echo. No dynamic decoupling. Just magic!

Measurement time > 2 seconds!

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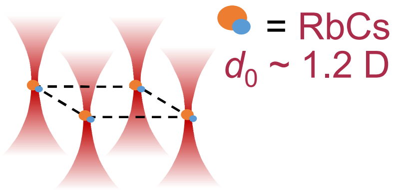
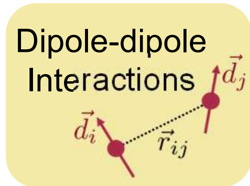


➌ Engineering long rotational coherences in magic traps



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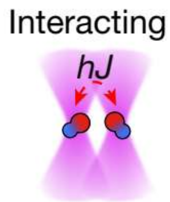
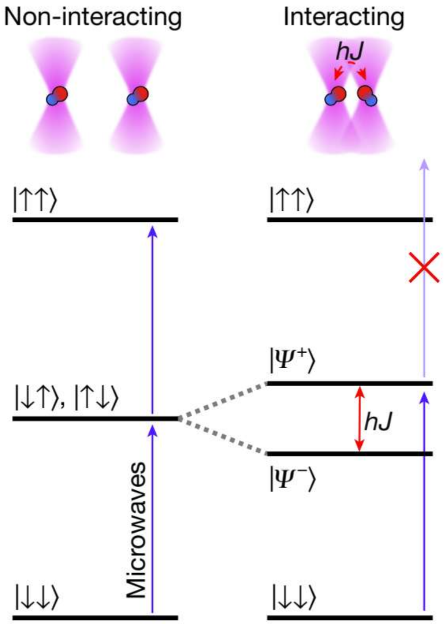


What about interactions?

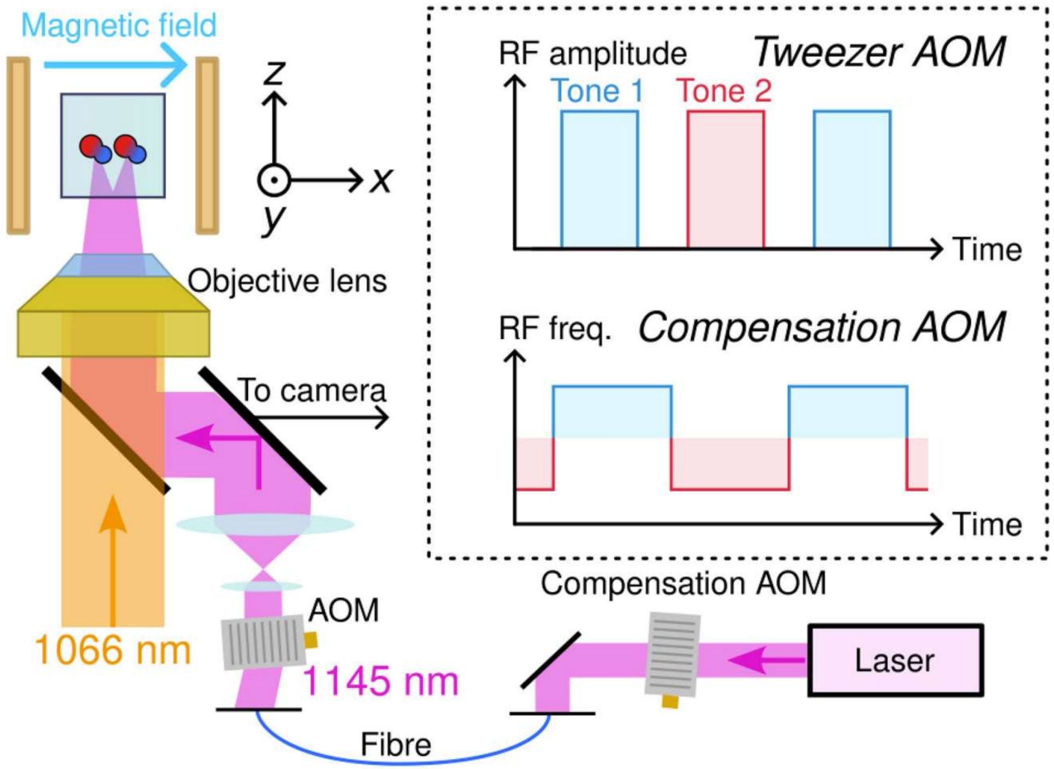
$$H_{\mathrm{DDI}} = \frac{J}{2}(\hat{s}_1^+ \hat{s}_2^- + \hat{s}_1^- \hat{s}_2^+)$$

But for ~3 μm separation and $(0,0) \rightarrow (1,1)$ transition, expect $J \simeq 5.6\,\mathrm{Hz}$

Need to make both tweezers magic! Requires some trickery…



Chop tweezers on & off. Use 2nd AOM to compensate detuning.



Ruttley et al., Nature **637**, 821 (2025)

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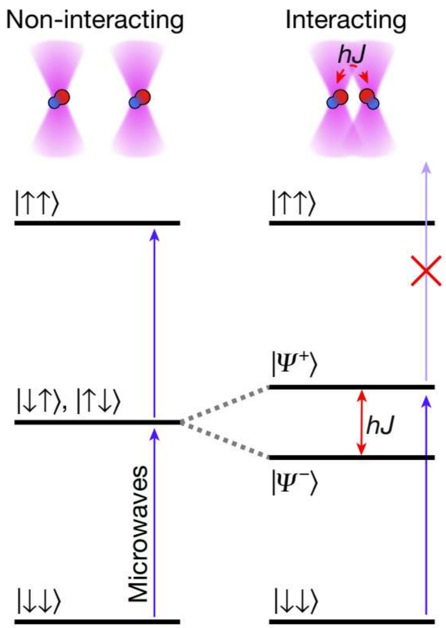
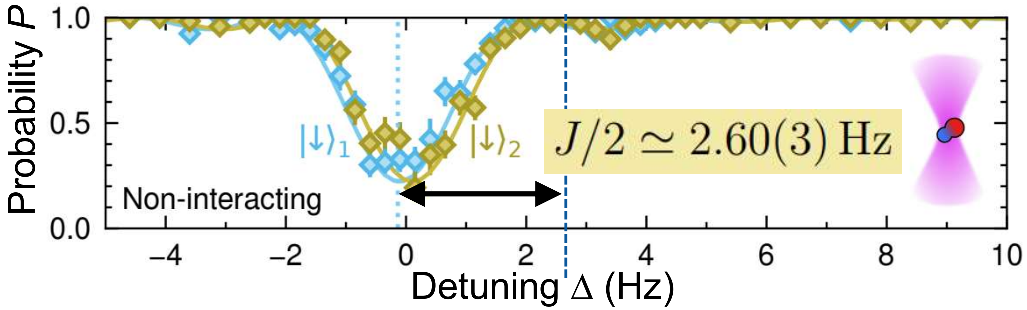
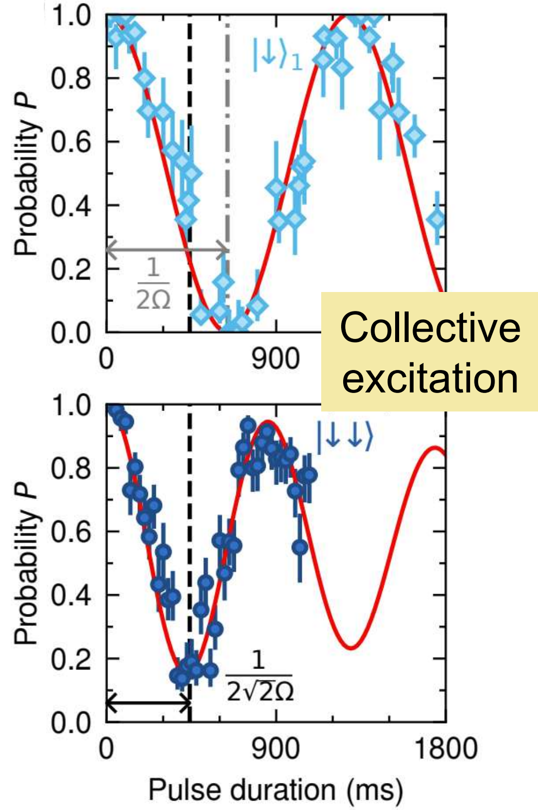






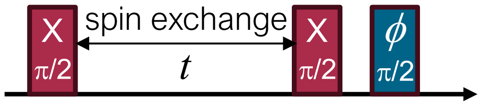
Ruttley et al., Nature **637**, 821 (2025)

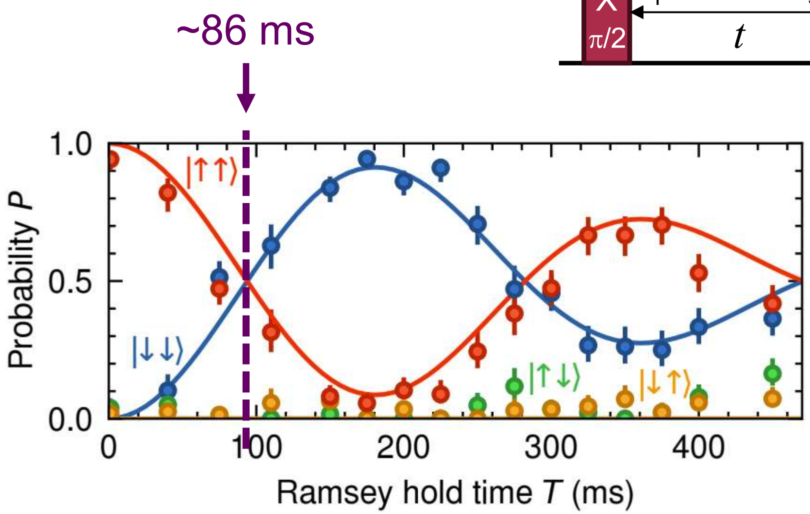
Entanglement via spin-exchange interactions

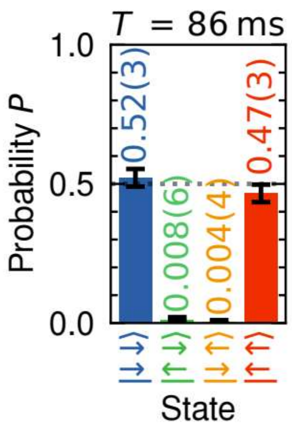
Durham University

Spin-exchange interaction in Ramsey sequence $|\Psi(t)\rangle = ie^{-2\pi i \frac{Jt}{4}} \left[\sin\left(2\pi \frac{Jt}{4}\right)|\downarrow\downarrow\rangle + i\cos\left(2\pi\frac{Jt}{4}\right)|\uparrow\uparrow\rangle\right]$

Evolve for $\dfrac{1}{2J}$ to produce maximally entangled state $\frac{1}{\sqrt{2}}(|\downarrow\downarrow\rangle + i\,|\uparrow\uparrow\rangle)$ (- equivalent to Bell state $|\Phi^{\pm}\rangle$)

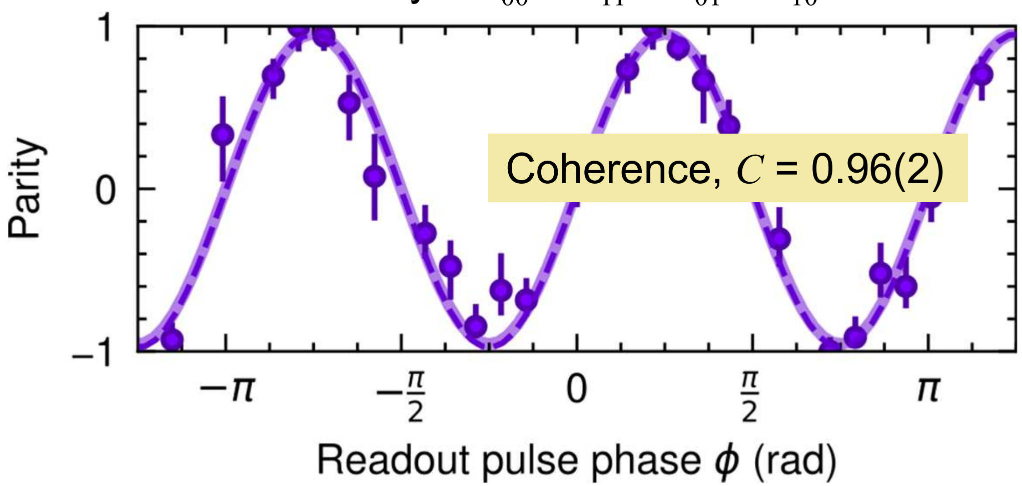






Measure coherence with parity oscillations

Parity $= P_{00} + P_{11} - P_{01} - P_{10}$



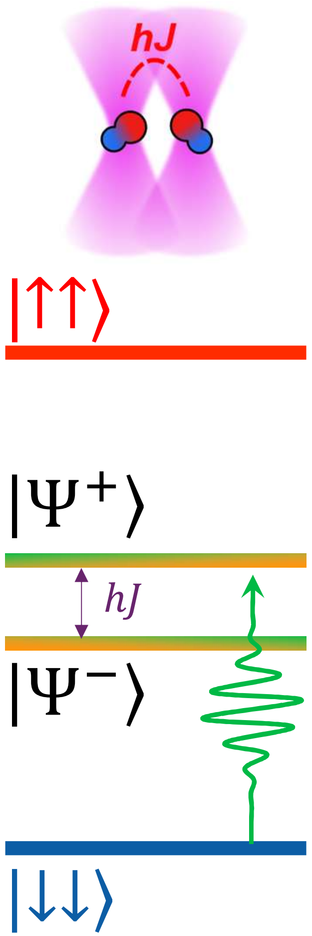
$\text{Fidelity} = \frac{1}{2}(C + P_{|\downarrow\downarrow\rangle} + P_{|\uparrow\uparrow\rangle}) = 0.976^{+0.014}_{-0.016}$

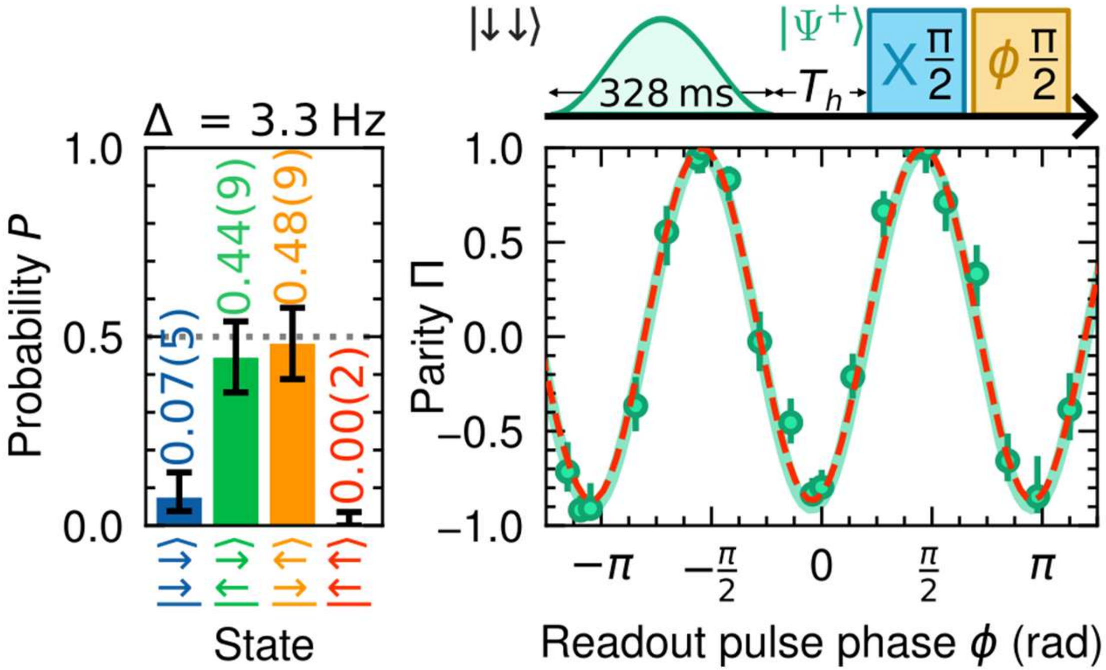
Ruttley et al., Nature **637**, 821 (2025)

(Automatically corrected for loss; otherwise $0.924^{+0.013}_{-0.016}$)

Direct microwave entanglement

Durham University





$$\mathcal{F} = (P_{\downarrow\uparrow} + P_{\uparrow\downarrow} + C)/2$$

$$\mathcal{F} = 0.76^{+0.03}_{-0.04}$$

Limited by detectable leakage/erasure errors

Correct for these errors

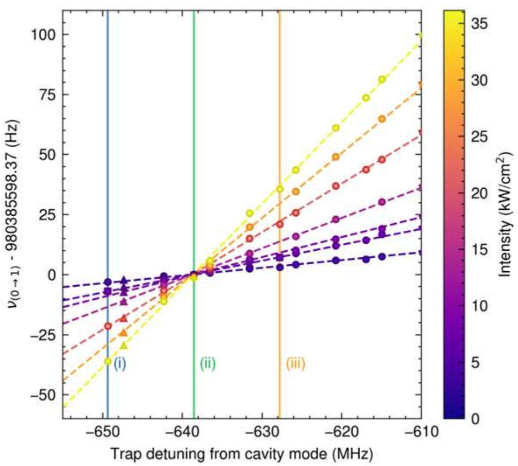
$$\mathcal{F} = 0.93^{+0.03}_{-0.05}$$

Limited by molecule temperature

Summary: magic trapping and control of single molecules

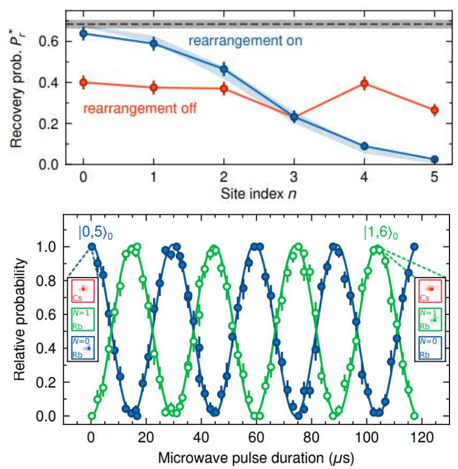
Coherence time = many seconds

Magic trapping and long rotational coherence



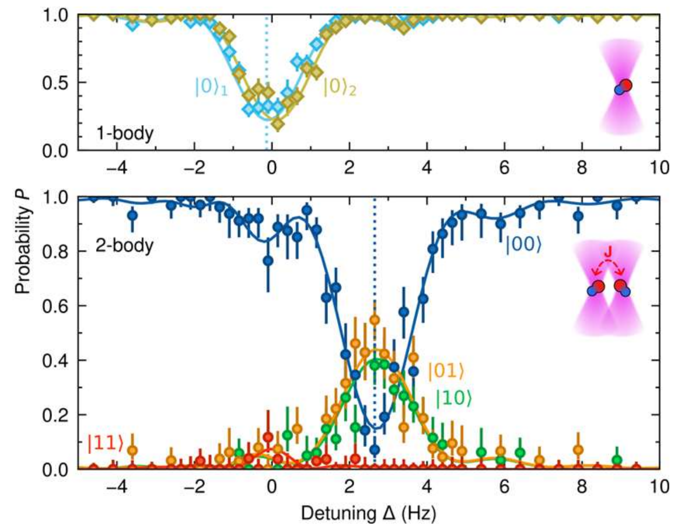
Gregory et al., Nature Physics (2024)

Tweezer arrays of RbCs molecules

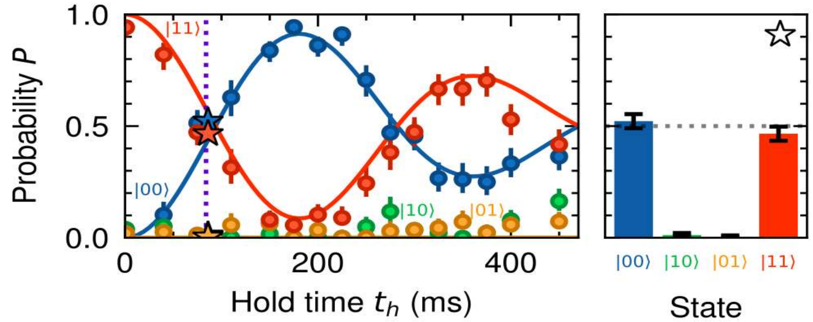


Ruttley et al., PRX Quantum (2024)

Spin exchange interactions



Controlled entanglement

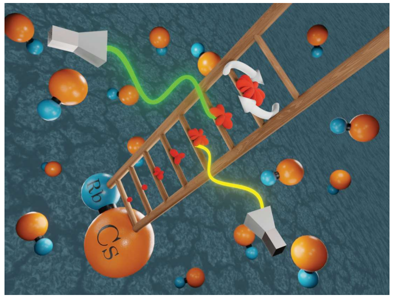


Ruttley et al., Nature (2025)

Outlook

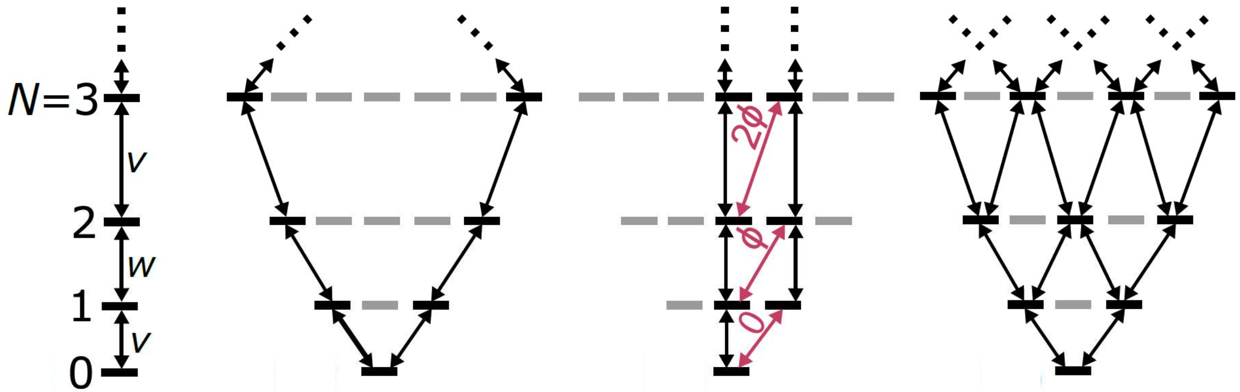
(things we are working on currently)

Outlook: synthetic dimensions



Advantages of synthetic dimensions:

- Increase dimensionality of the system
- Site specific tunnelling controlled with microwave fields
- Geometry easily reconfigured
- Single site detection trivial



Su-Schrieffer-Heeger model

1D chain with periodic boundary conditions

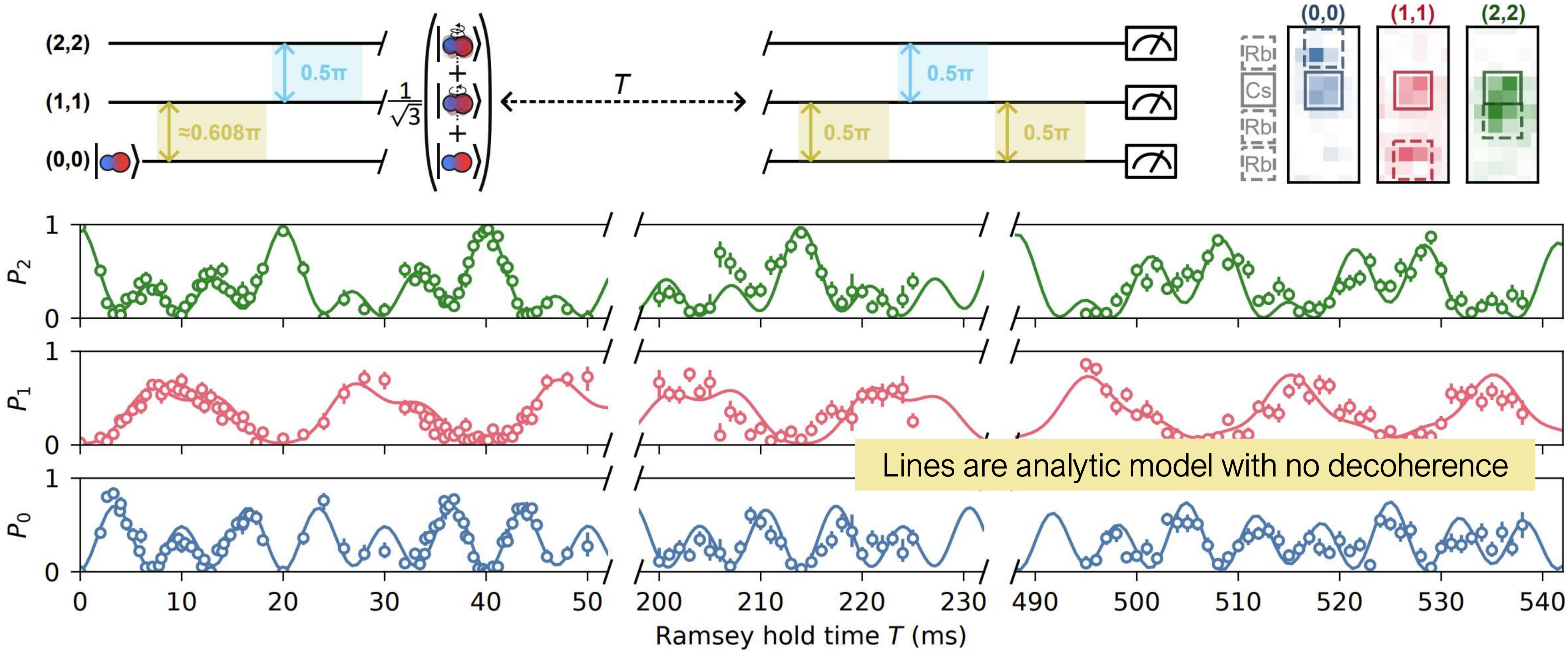
Simulating gauge fields

2D square lattice

"Synthetic dimensions in ultracold polar molecules"
Sundar, Gadway, Hazzard, Scientific Reports **8**, 3422 (2018)

Outlook: extend magic trapping to more levels

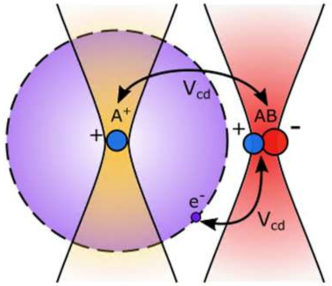
Prepare coherent superposition of 3 **rotational states**, evolve, close Ramsey interferometer. Multistate readout.



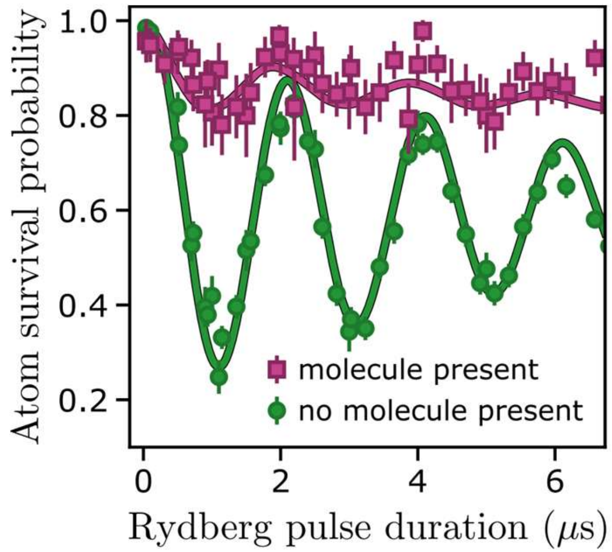
Next: interacting spin-1 system OR mapping to spin ½ hard-core bosons / t-J models:
Homeier et al., PRL **132**, 230401 (2024); Wellnitz et al., arXiv:2409.05109; Qiao et al., arXiv:2501.08233

Outlook: combining Rydberg atoms and molecules

Rydberg-blockade due to charge-dipole interaction with molecule



~300 nm separation!



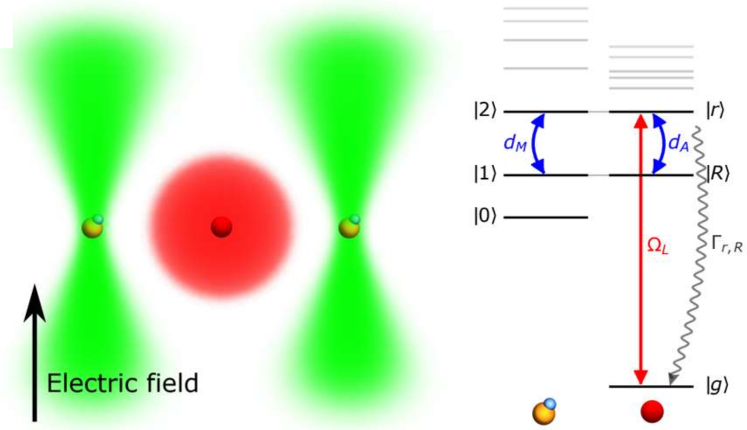
Guttridge & Ruttley *et al.*, PRL **131**, 013401 (2023).

Resonant dipole-dipole interaction:

Non-destructive detection & Rydberg mediated gates

Rydberg spacing matches rotational splitting – resonant dipole-dipole interaction ∝ d_M x d_A

MHz rather than kHz interactions at ~ 1 micron



Zhang & Tarbutt, PRX Quantum **3**, 030339 (2022).
See also Wang et al., PRX Quantum **3**, 030339 (2022).

Acknowledgements

Durham University

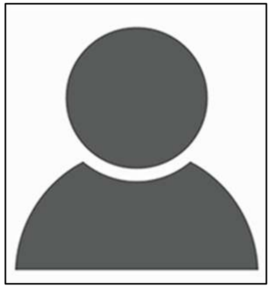
These experiments are a big team effort:

Philip Gregory

Alex Guttridge

Royal Society University Research Fellows

You?



Postdocs wanted

Theory Collaborators:
Jeremy Hutson (everything molecule related)
Svetlana Kotochigova (magic trapping)
Rosario Gonzalez-Ferez (Rydberg molecules)
Hossein Sadeghpour (Rydberg molecules)
Kaden Hazzard (DDI & synthetic dimensions)
Matt Eiles (Rydbery molecules)

erc
European Research Council

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UKRI Engineering and Physical Sciences Research Council

Thank you