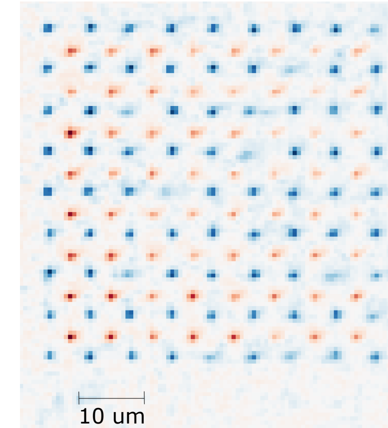
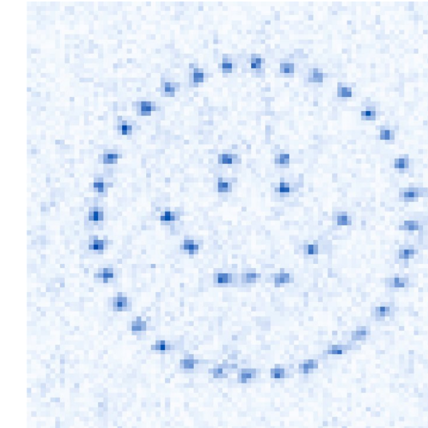
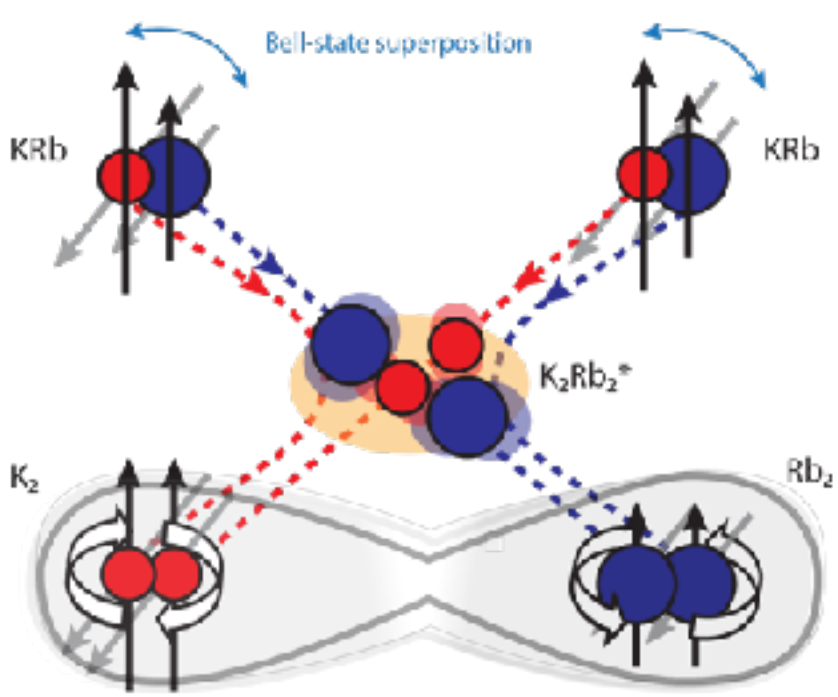


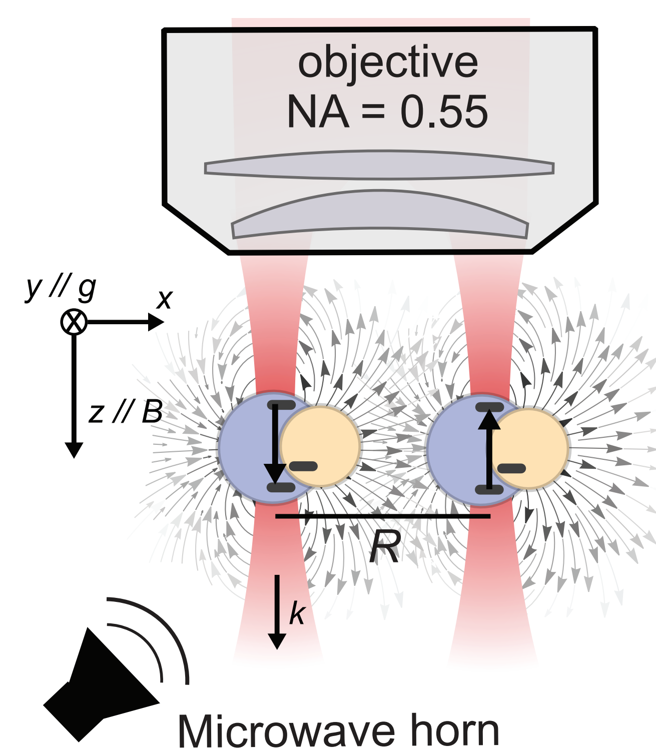


Many-Body Physics/QIS:

Entanglement and Logic Gate between Molecules



Kang-Kuen Ni
Harvard University



Funding: AFOSR, NSF, DOE, Moore Foundation, MURI-AFOSR, IARPA, LBNL

Why Ultracold Molecules?

Physica Scripta. Vol. T70, 34–41, 1997

Testing Time Reversal Symmetry Using Molecules

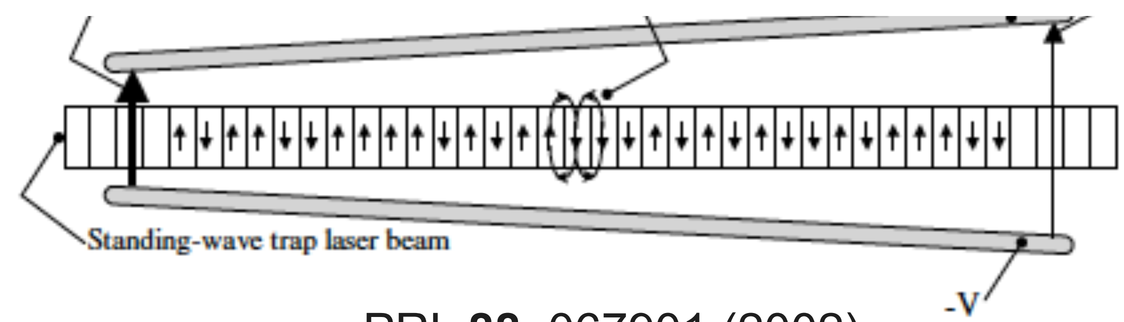
E. A. Hinds

Physics Department, University of Sussex, Falmer, Brighton BN1 9QH, U.K.

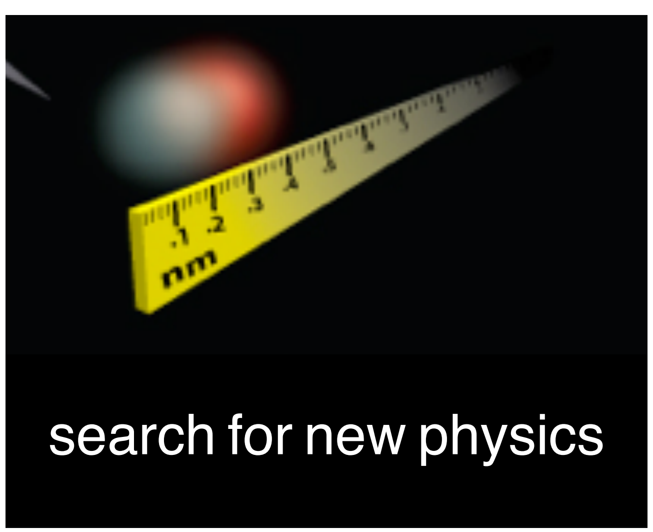
Received February 16, 1996; accepted April 30, 1996

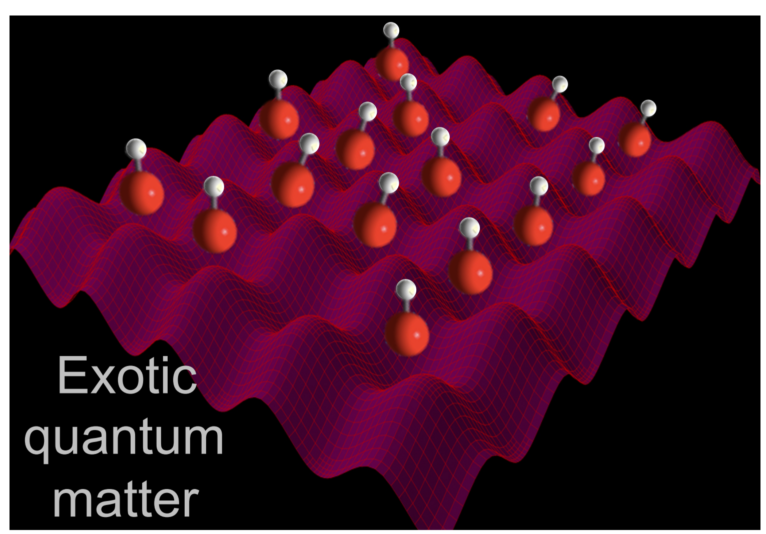
Quantum Computation with Trapped Polar Molecules

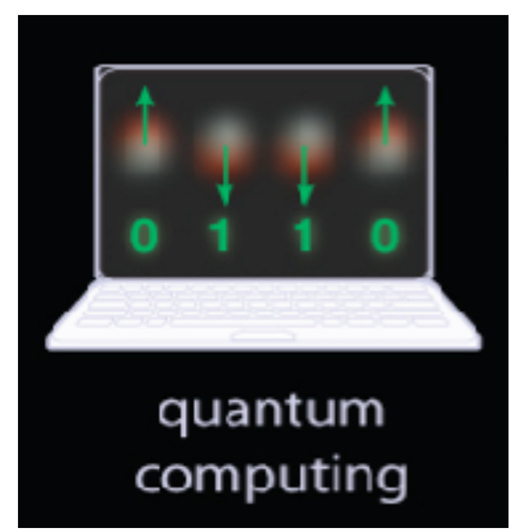
D. DeMille

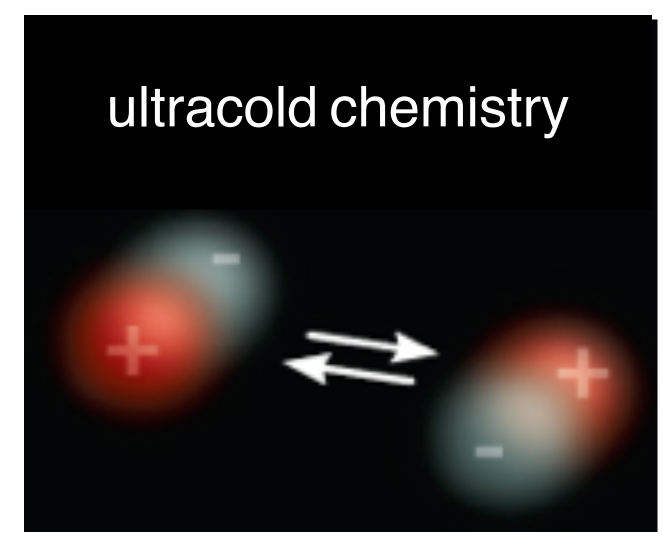


PRL **88**, 067901 (2002)









single-body physics

two- to many-body physics

two- to few-body physics

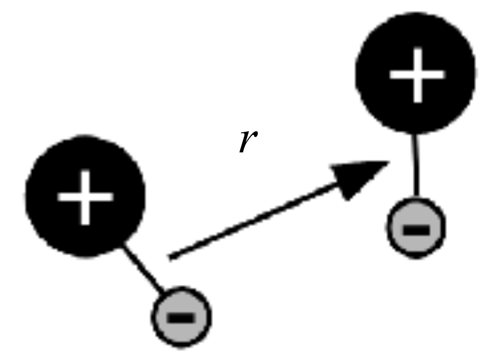
Molecular Resources

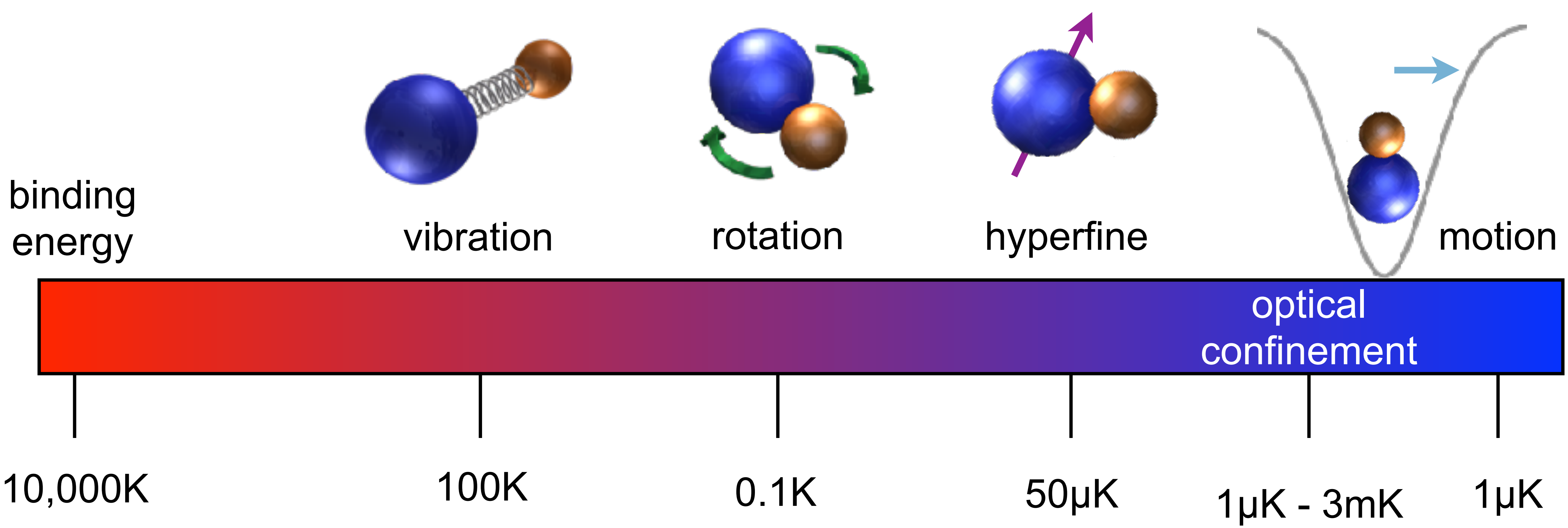
- Rich internal degrees of freedom
- Intrinsic, tunable electric dipolar interactions

complex, but intrinsically coherent *qubits*
to tailor to multiple requirements

Dipole-dipole interaction

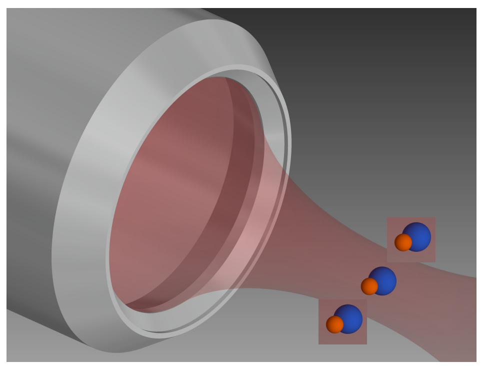
$$\hat{H}_{DD} = \frac{1}{4\pi\epsilon_0 r^3}\left[\hat{d}_1 \cdot \hat{d}_2 - 3(\hat{d}_1 \cdot \hat{e}_r)(\hat{d}_2 \cdot \hat{e}_r)\right]$$

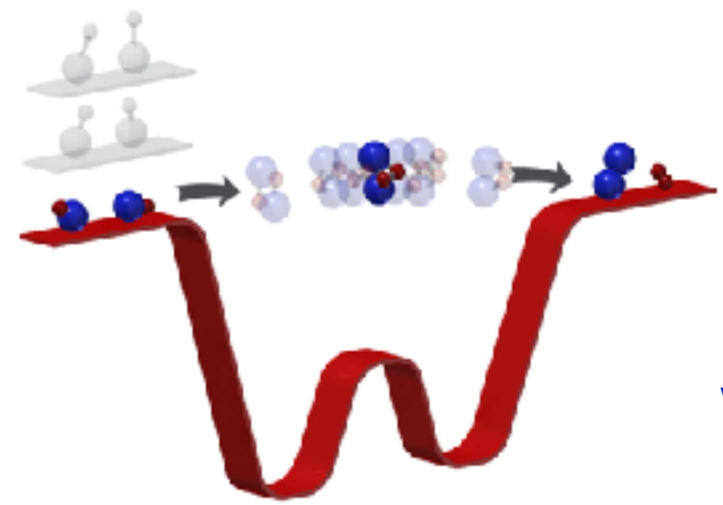




Entangling Physical System

Molecules in optical tweezer arrays





iSWAP gate with molecules

- via well-controlled interactions
- New approaches?

Dipolar interaction between molecules

Take advantage of chemical reactions?

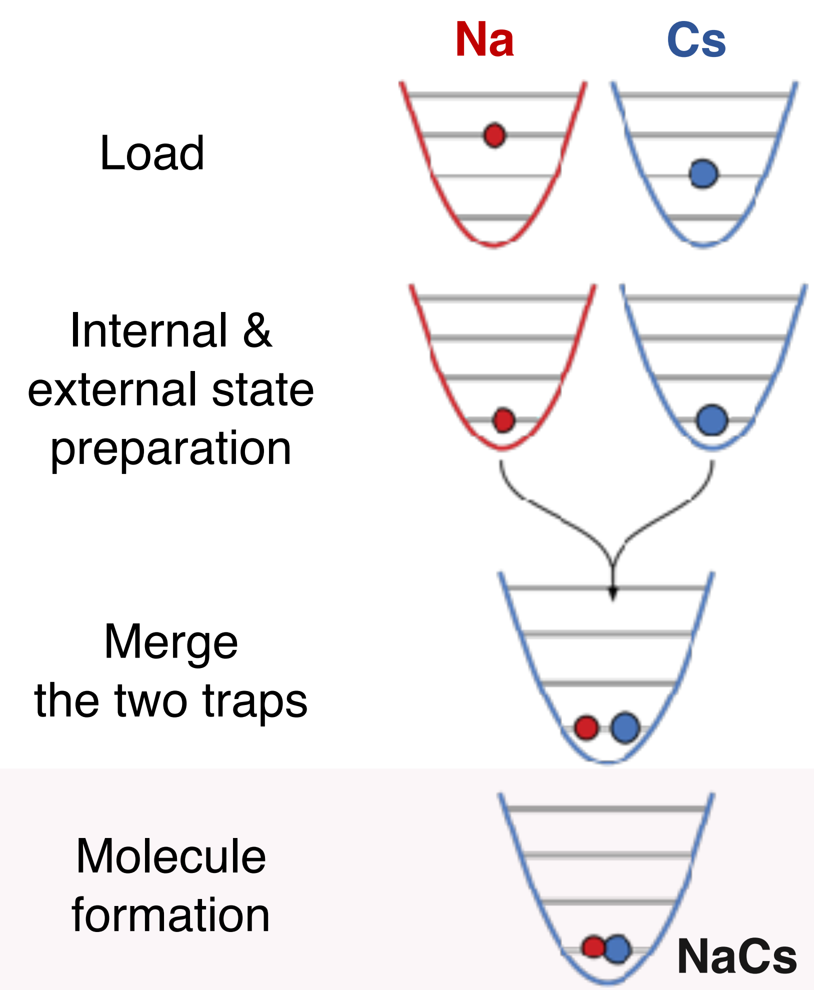
Two approaches to

Programmable arrays of *single* molecules

Science **360**, 900 (2018)

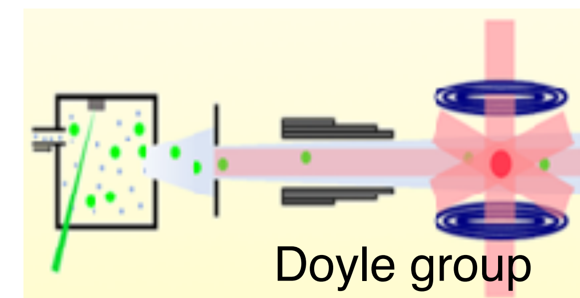
- single particle control and detection
- single quantum state preparation
- rovibrational ground state
- motional ground state

Single molecule association from laser-cooled atoms

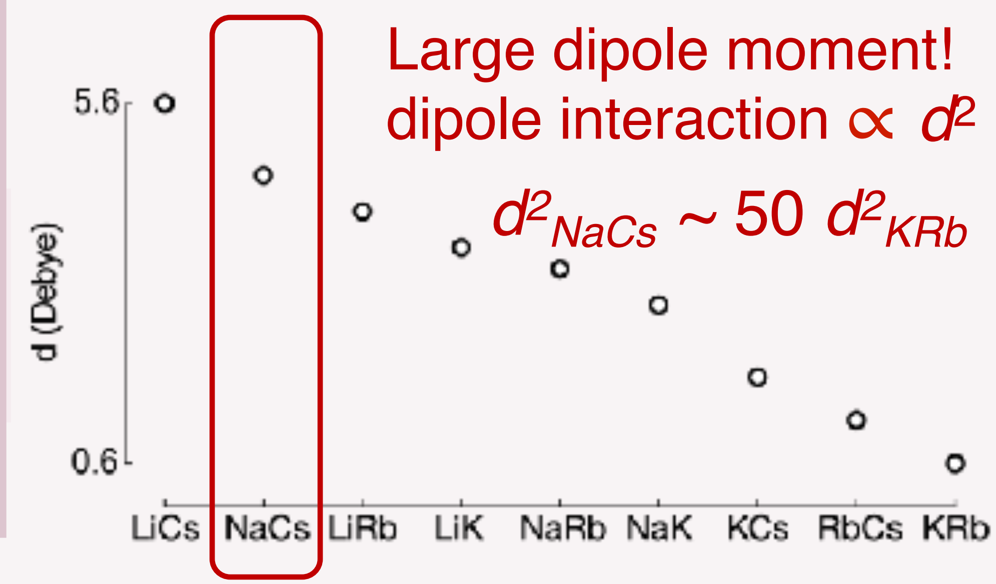


Another approach

Direct laser cooling



CaF tweezer array
Doyle @ Harvard
Cheuk @ Princeton

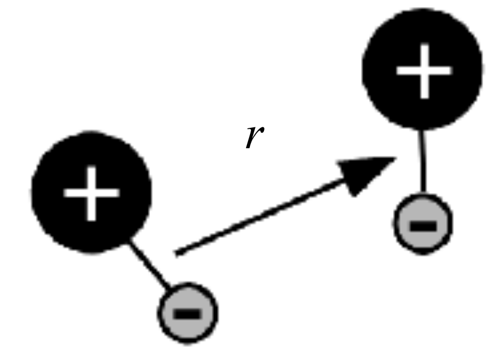


Large dipole moment!
dipole interaction $\propto d^2$

$d^2_{NaCs} \sim 50\ d^2_{KRb}$

Related Molecule work in Tweezers: Doyle (CaF, CaOH), Cheuk (CaF), Cornish (RbCs), Wuhan group (Rb_2*)
Other approaches: molecular ions and quantum gas microscope of molecules
Pioneering work: JILA, Innsbruck, …

Resonant Dipolar Exchange



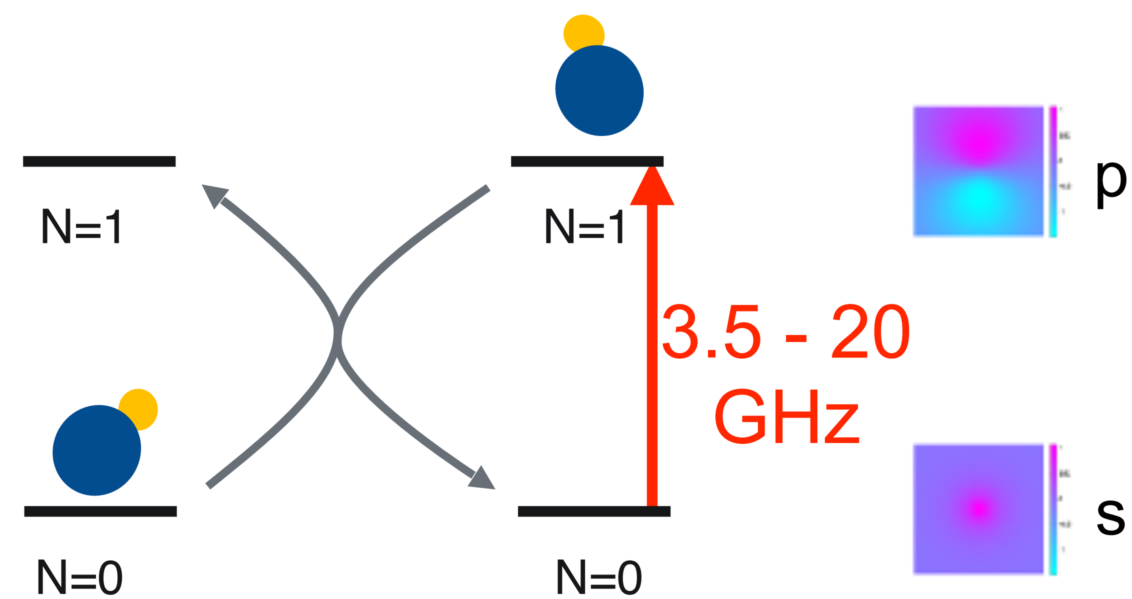
in the absence of a laboratory
polarizing electric field

$$\hat{H}_{DD} = \frac{J_\perp}{2} \sum_{i>j} V_{dd}(r_i - r_j)(S_i^+ S_j^- + S_i^- S_j^+)$$

Rey, Gorshkov et al., PRL 107, 115301 (2011)

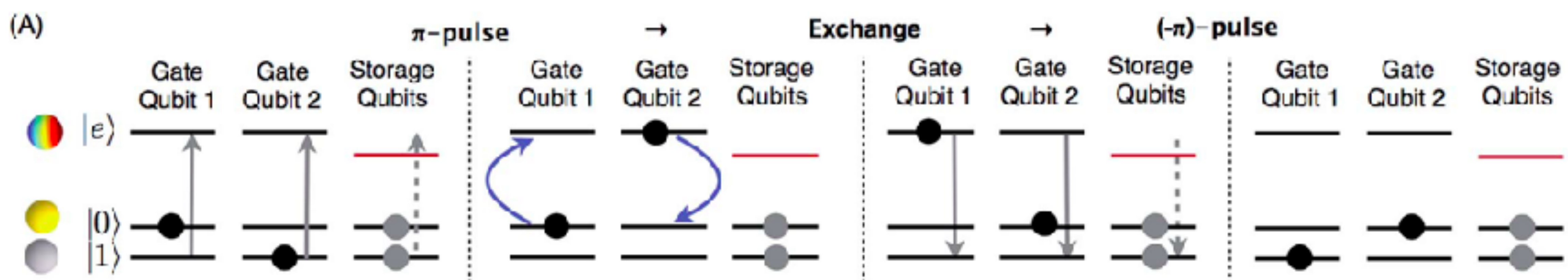
(JILA) Jin & Ye & Rey, Yan et al., Nature (2013)

Bakr, Cheuk, Doyle groups (2023)



Pioneering work with Rydberg atoms & Magnetic atoms: Browaeys, Pfau, Laburthe-Tolra, Ferlaino, Lev…

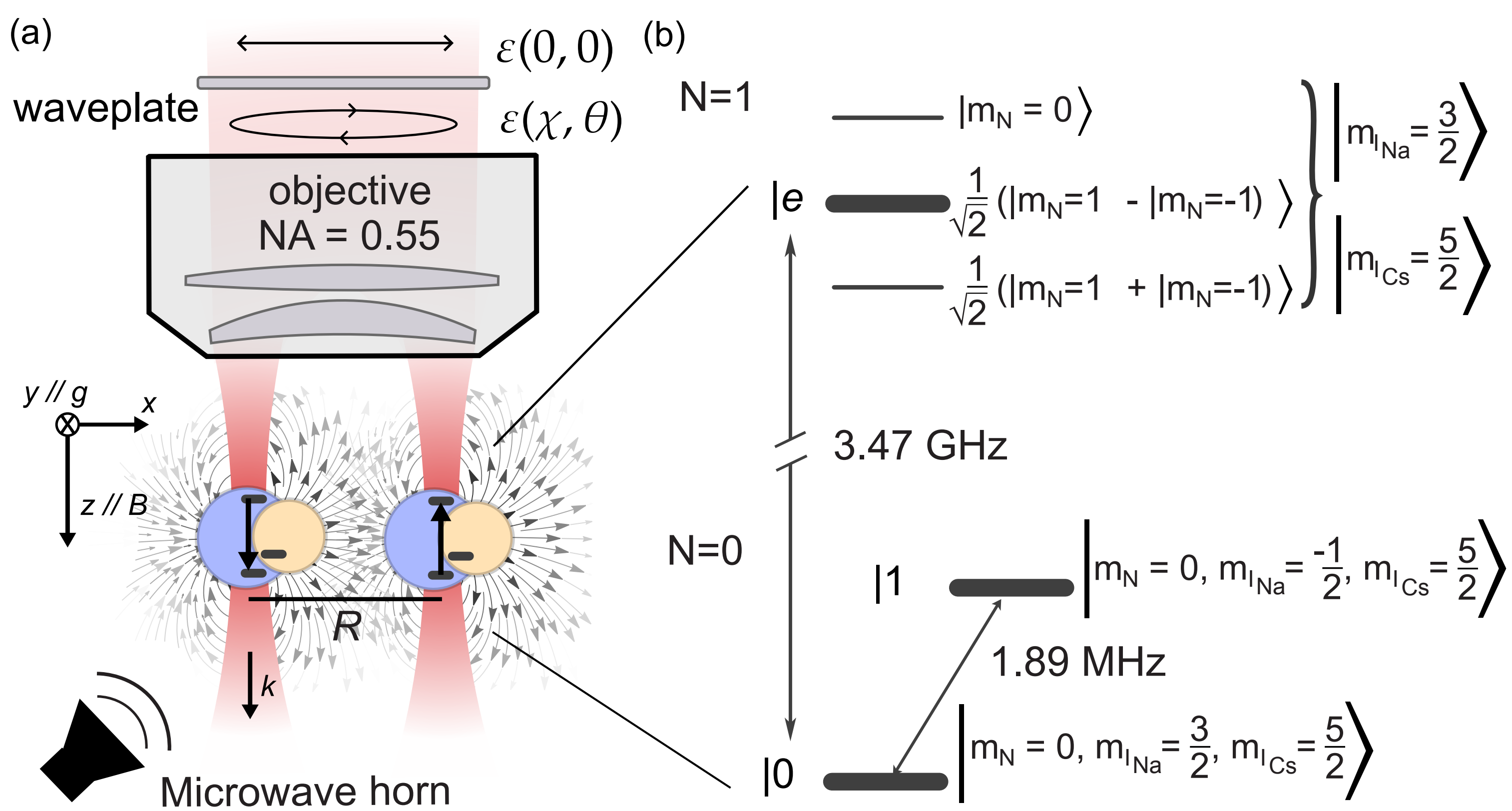
iSWAP logic gate



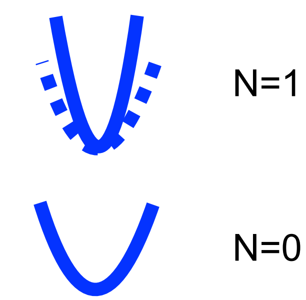
Ni, Rosenband, Grimes, Chemical Science **9**, 6830-6838 (2018)

Related work: Hudson, Campbell, PRA **98**, 040302 (2018)

NaCs Molecular Structure

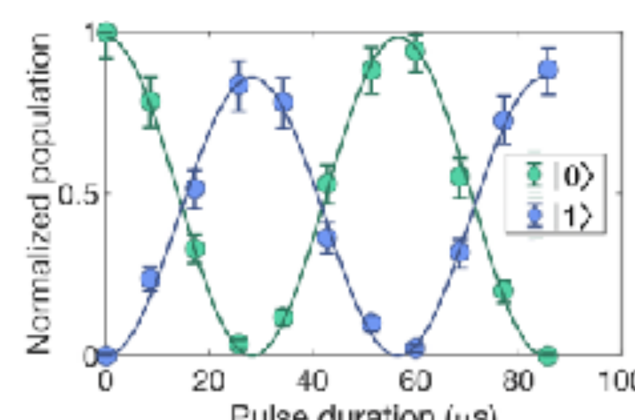


- 3 relevant levels $|0\rangle$, $|1\rangle$, $|e\rangle$
- “Magic” ellipticity trapping



Park*, Picard* et al, PRL(2023)

- Multi-state readout



Picard*, Pantenotte*, Park* et al, PRX Quantum (2024)

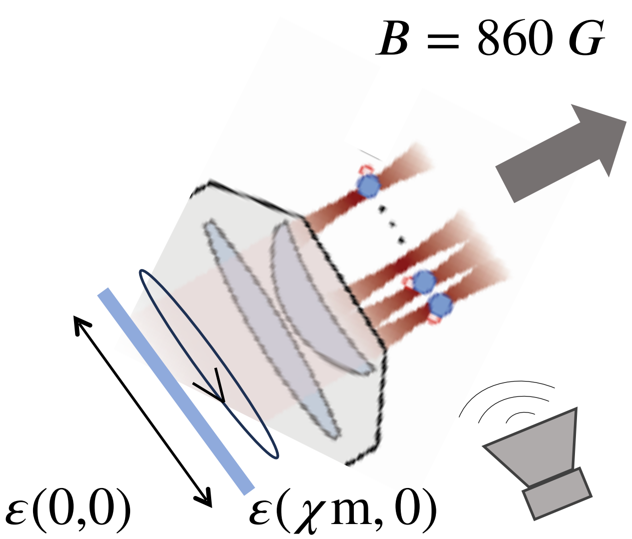
Annie Park

Lewis Picard

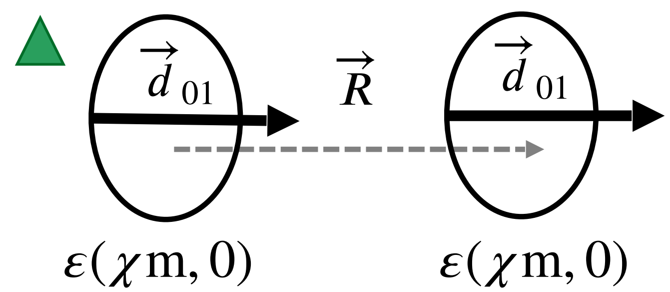
Gabriel Patenotte

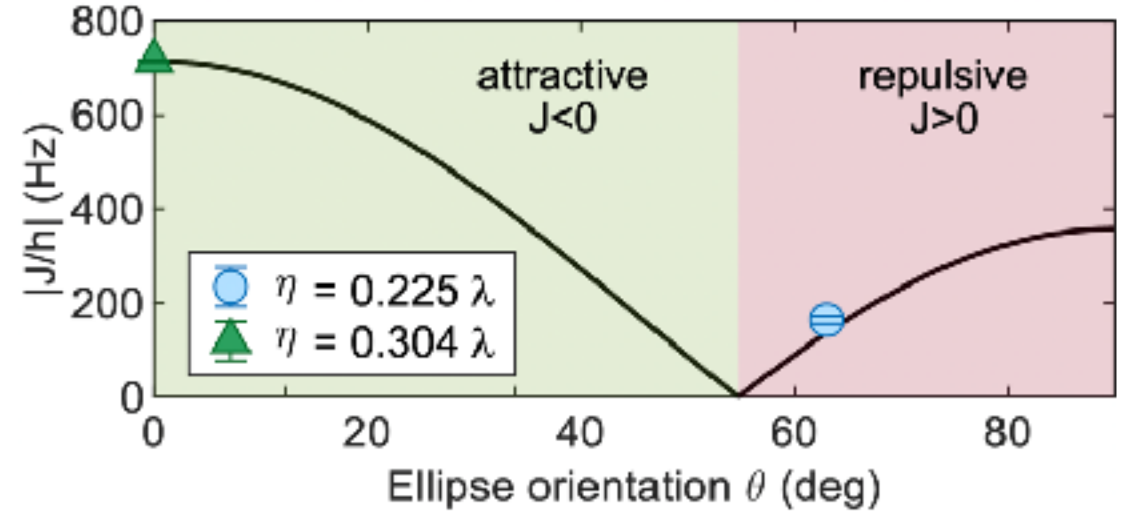
Sam Gebretsadkan

Tunable Dipolar Interaction

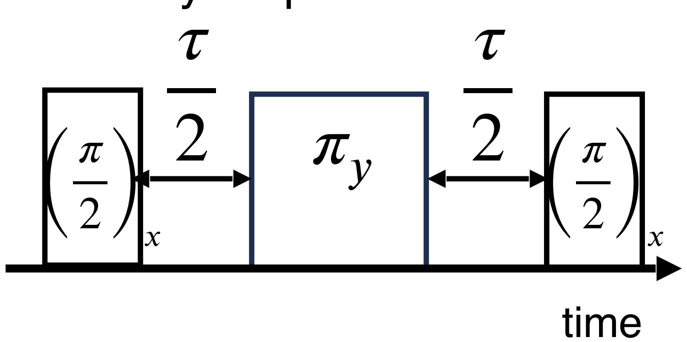


Tune interaction to max,
and bring molecules close



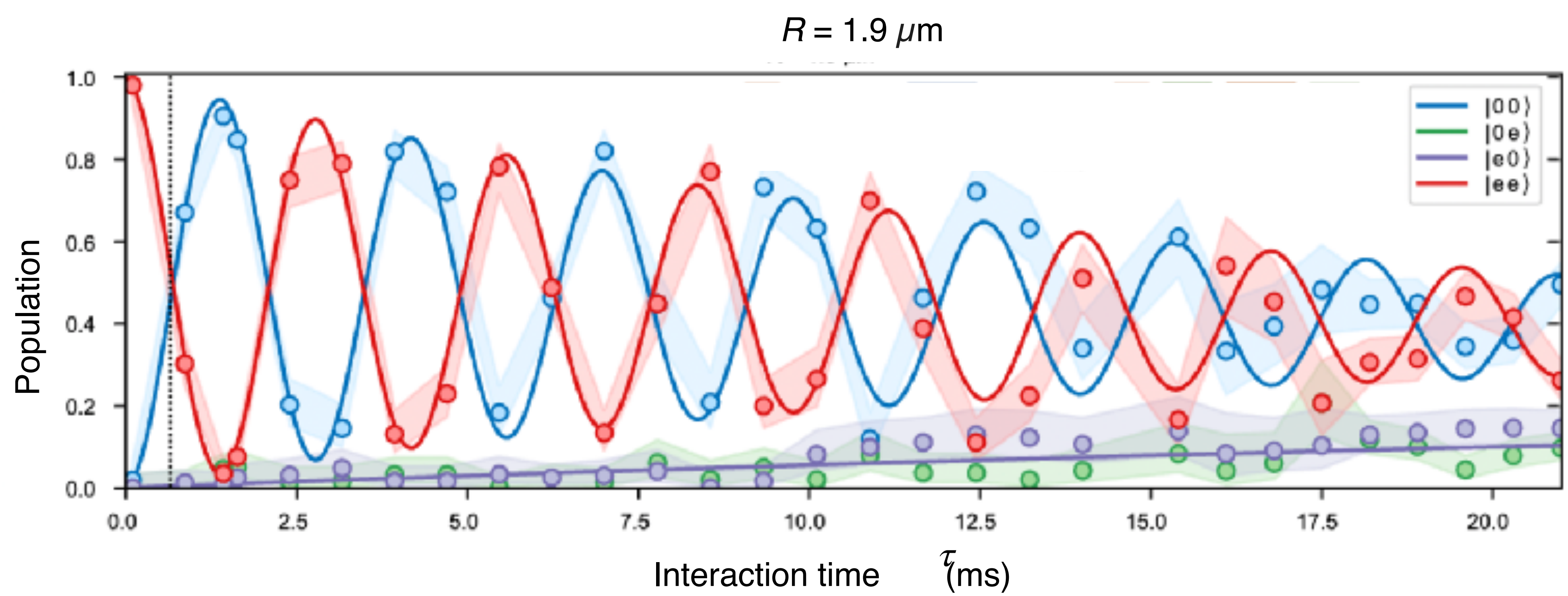


Ramsey sequence



Final state:

$$|\psi(\tau)\rangle = \tfrac{1}{2}\left(1 - e^{-i\frac{J\tau}{2\hbar}}\right)|00\rangle + \tfrac{1}{2}\left(1 + e^{-i\frac{J\tau}{2\hbar}}\right)|ee\rangle$$

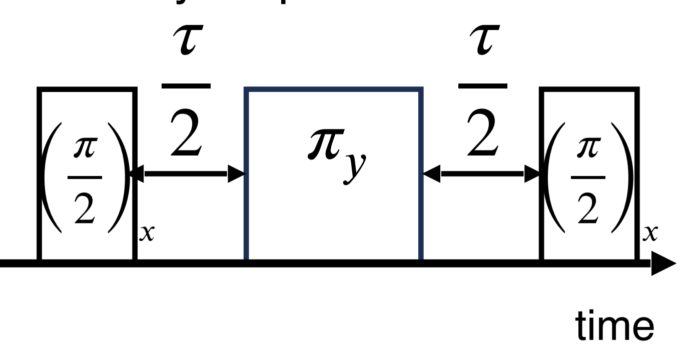


Solid lines: phenomenological Master equation fit

Picard*, Park*, Patenotte, Gebretsadkan, Wellnitz, Rey, Ni arXiv:2406.15345 (2024)

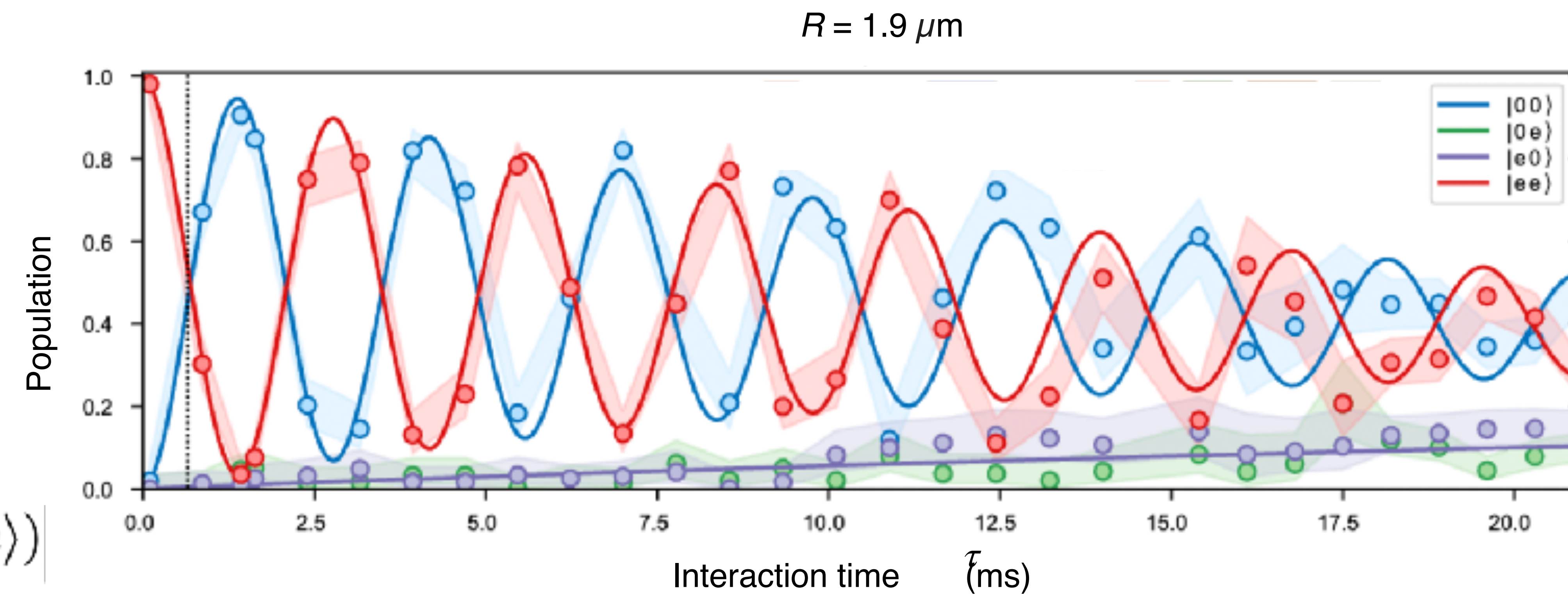
Sub-millisecond Bell-state Creation

Ramsey sequence



At $\tau = h/2J$ **=664μs**

$$|\psi(\tau)\rangle = \frac{1}{\sqrt{2}}(|00\rangle - i\,|ee\rangle)$$

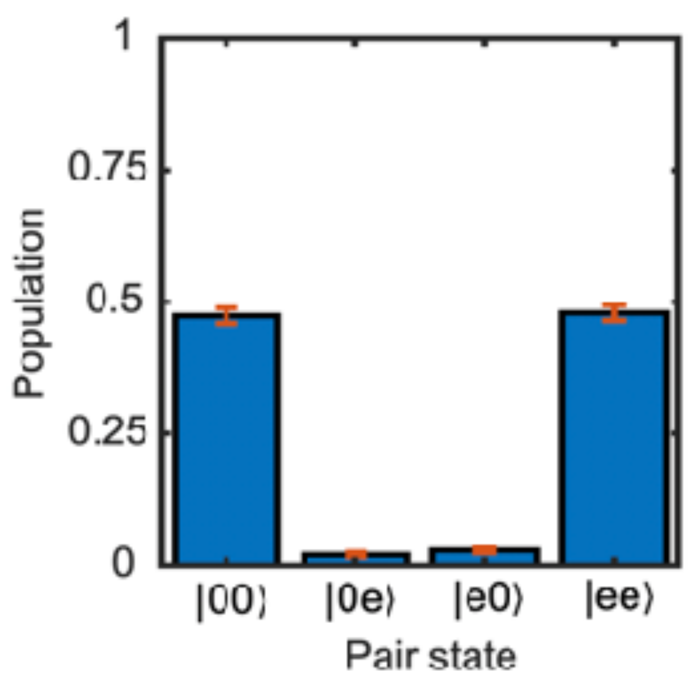


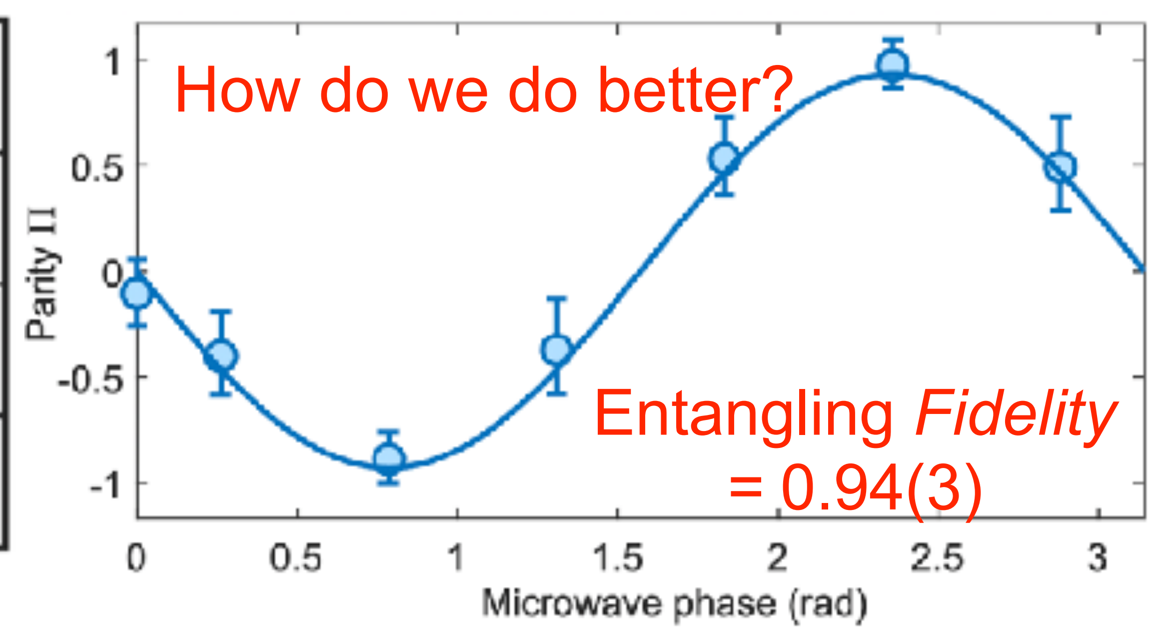
Bell state fidelity

$$F = \frac{1}{2}\left(C + P_{00} + P_{ee}\right)$$

1. Population measurement
2. Parity Oscillation

$$\Pi = P_{00} + P_{ee} - \left(P_{0e} + P_{e0}\right) = C\sin(2\theta)$$





Previous work:

Holland et al., Science **382**, 1143 (2023)
Bao et al., Science **382**, 1138 (2023)

Picard*, Park*, Patenotte, Gebretsadkan, Wellnitz, Rey, Ni arXiv:2406.15345 (2024)

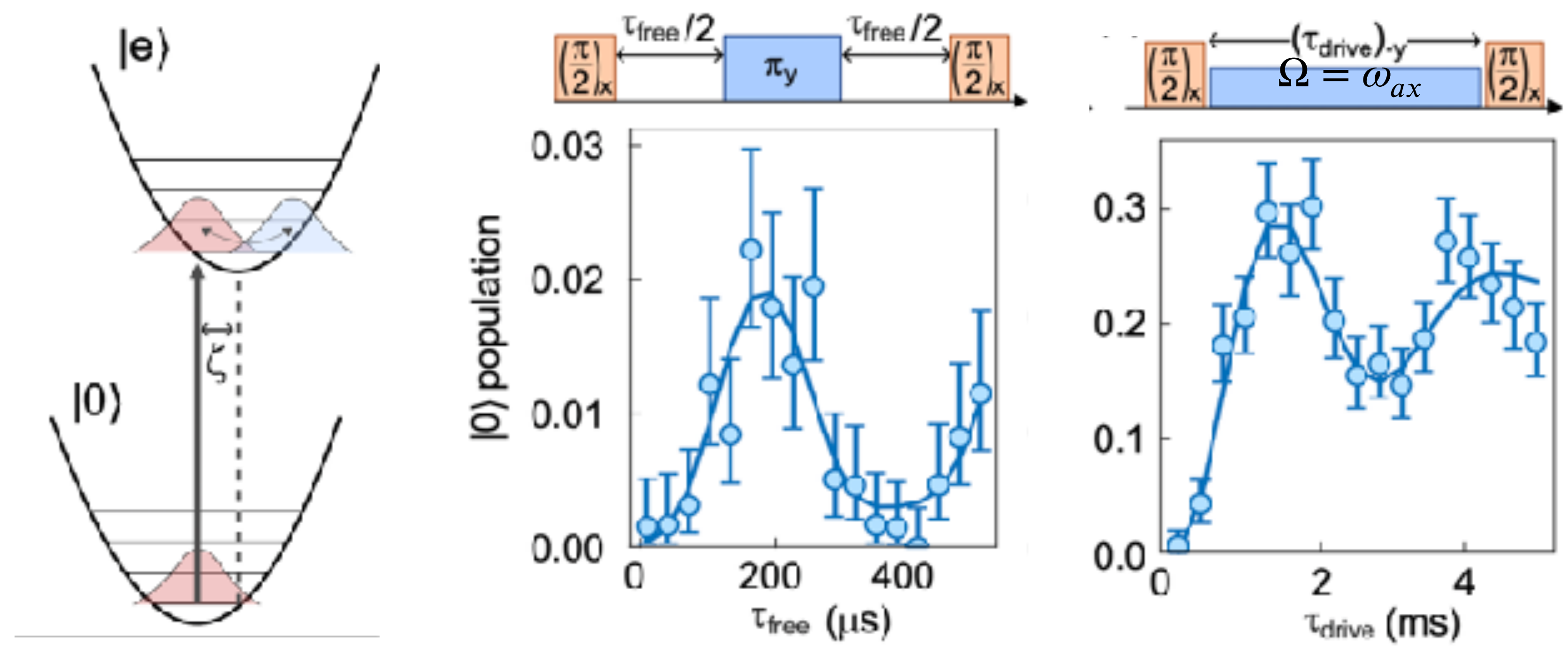
leveraging Motion-Rotation Coupling

David Wellnitz

Ana Maria Rey

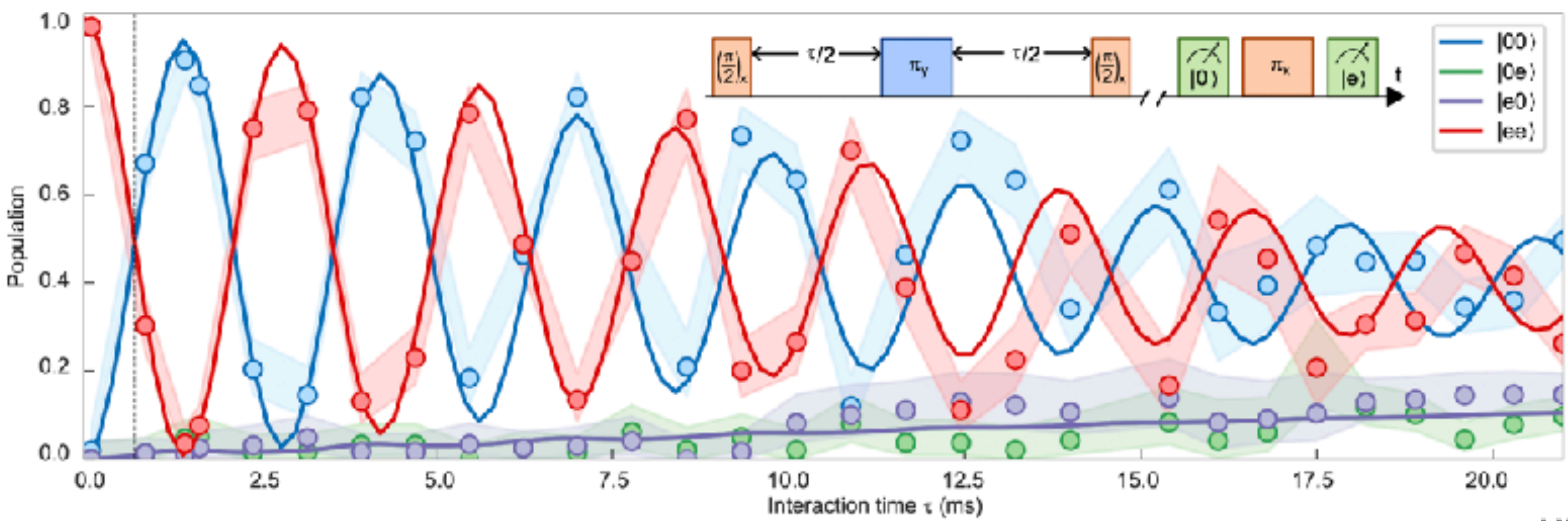
Determine temperature (T) of molecules, which estimates noise in separation R
→ *the **main** limit on* two-particle decoherence

Single-particle measurements to estimate ζ and T



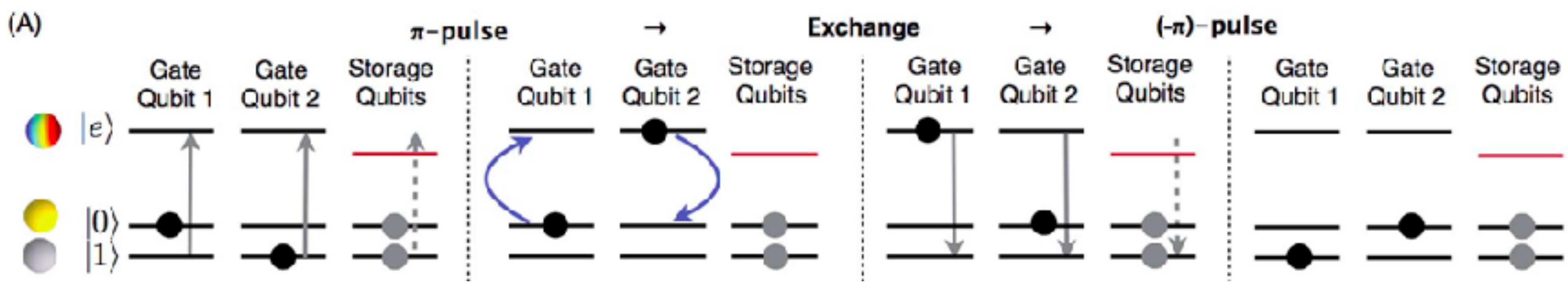
$\zeta \sim 5$ nm, $\hbar\omega_{\mathrm{ax}}/(k_B T) = 0.42(6)$ → Axial ground state fraction: 34(4) %

Solid lines:
Theoretical simulation
assuming parameters found above + single particle decoherence chosen to match the data



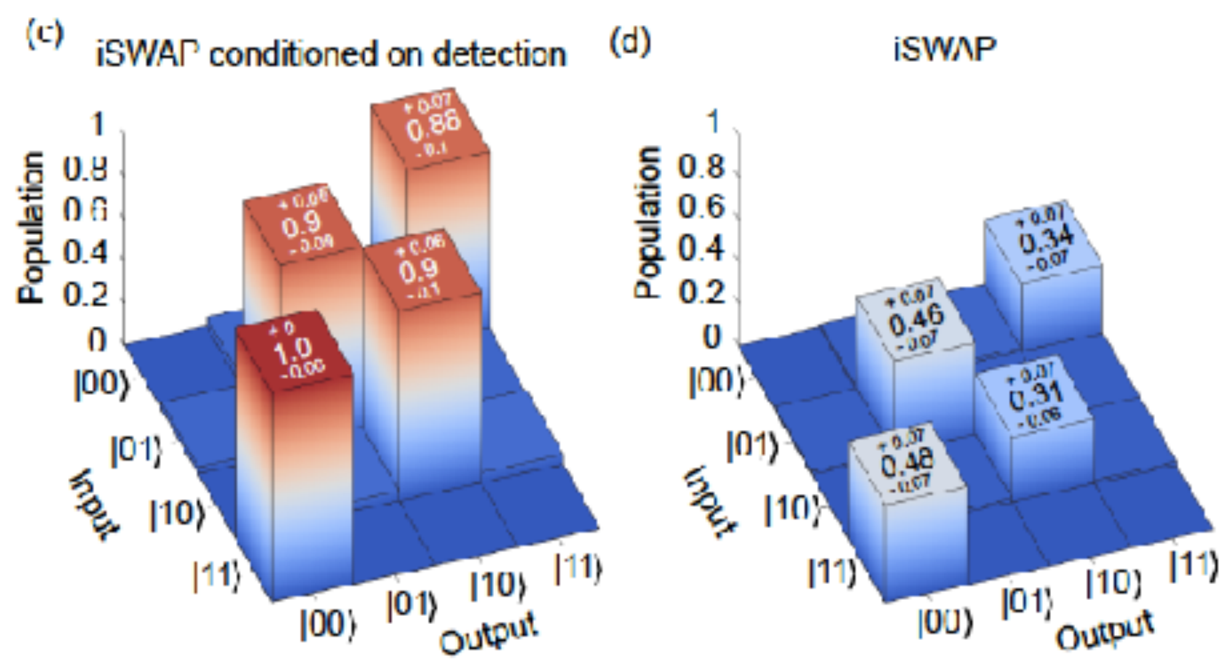
Future:
Infidelity ~ 2x10^{-4} by achieving 80 % ground state fraction + fixing astigmatism

iSWAP gate



- Encode qubits in a pair of hyperfine states
- Switch on/off interaction by driving between |1⟩ and |e⟩ via nuclear quadruple coupling (the current limiting factor)
- Total gate time 960 μs

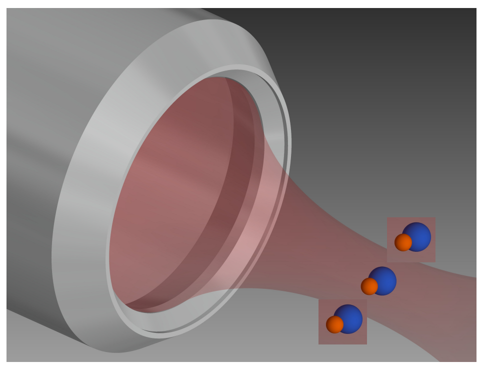
Logic output truth table

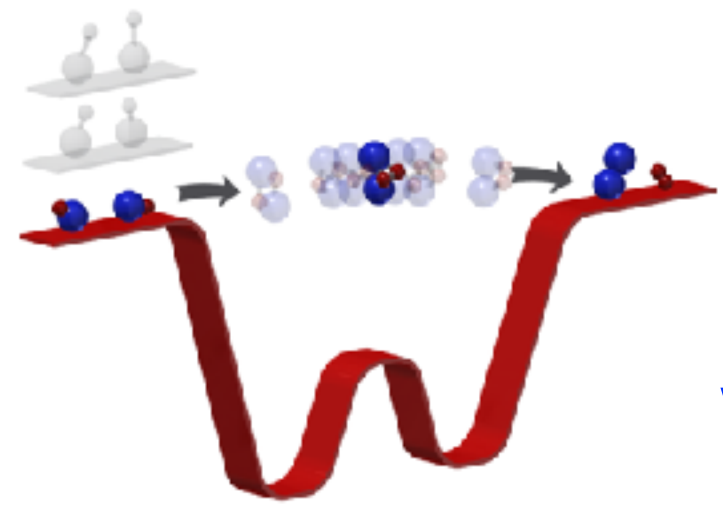


Leveraging the intrinsic molecular properties gives rise to highly coherent interactions. Quantum state preparation of molecules remains a key challenge.

Entangling Physical System

Molecules in optical tweezer arrays





iSWAP gate with molecules

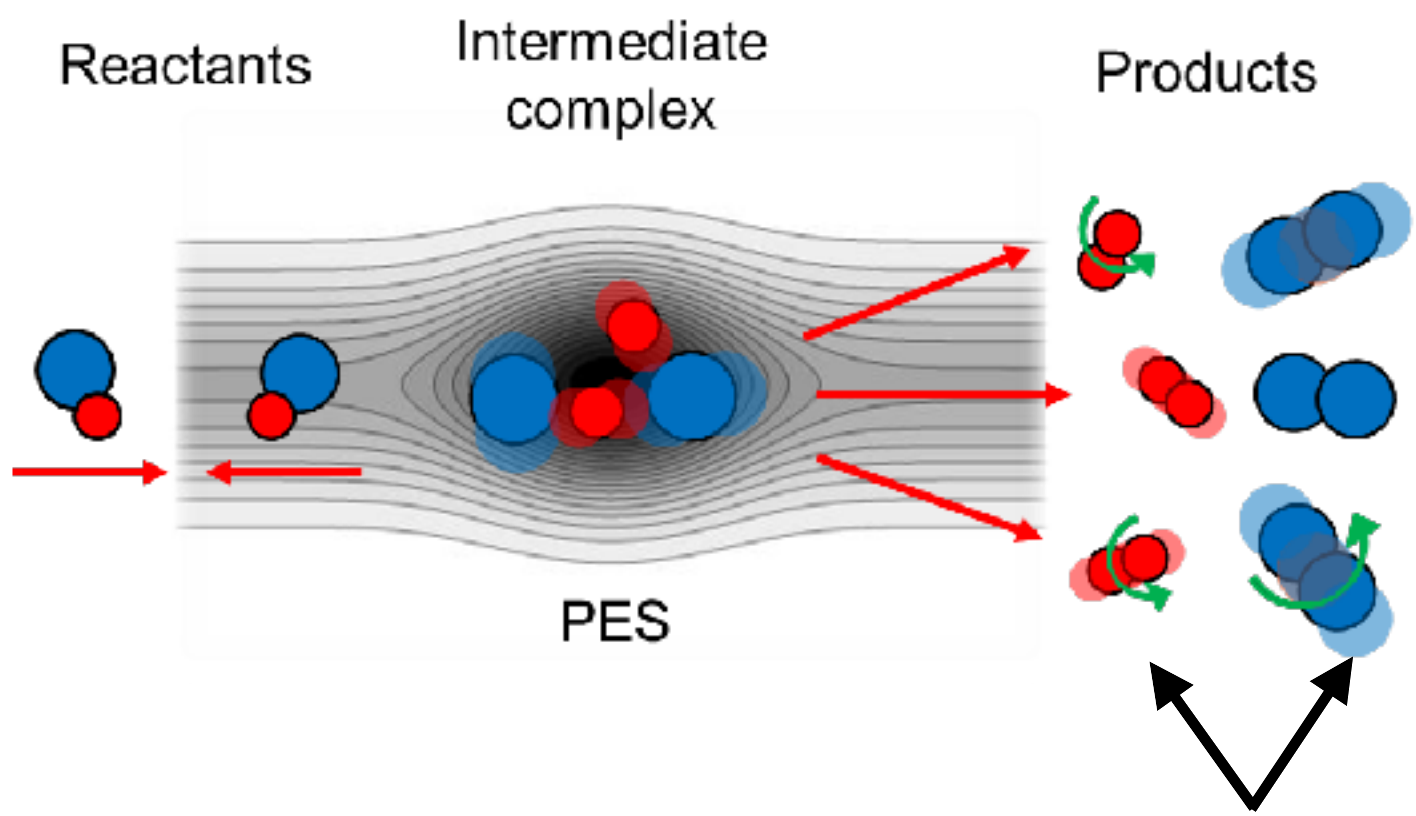
- via well-controlled interactions

Dipolar interaction between molecules

- New approaches?

Take advantage of chemical reactions?

Harnessing Chemical Reaction for Quantum Science?

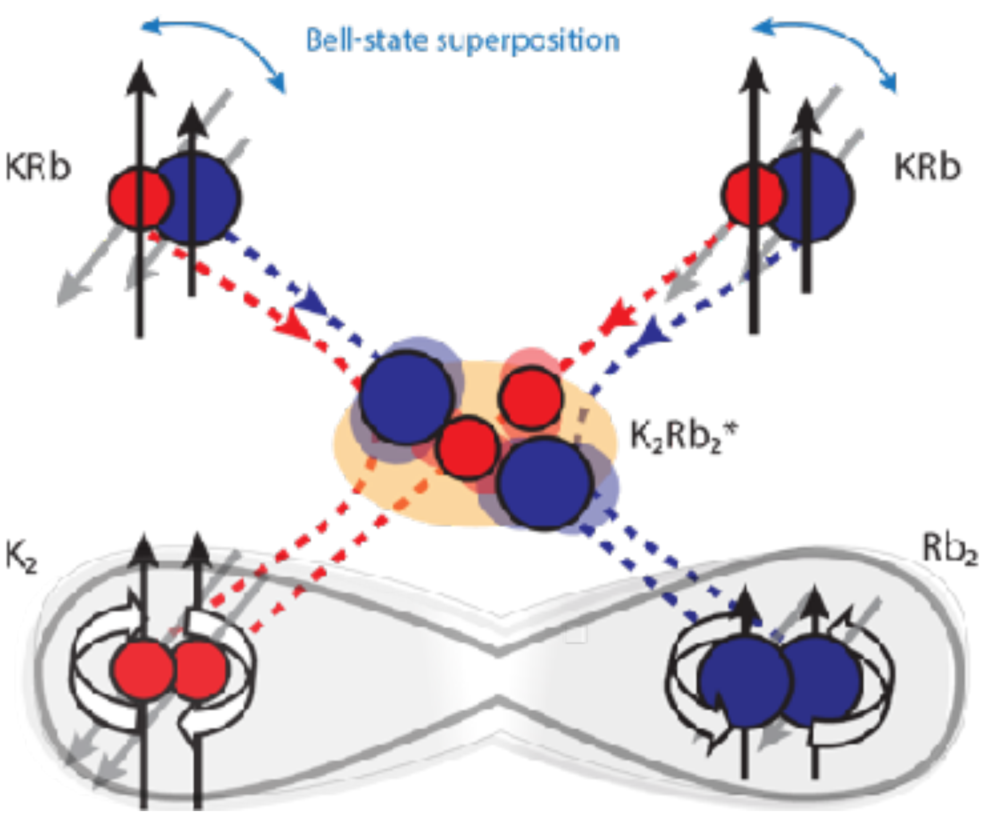


Entangled pairs??

Idea

Prepare entangled spins within each molecules, then use chemical reaction to rearrange the atoms into separate, entangled molecules

Questions:
Can coherence (phase) be maintained throughout a reaction?




Yi-Xiang Liu


Lingbang Zhu


Jeshurun Luke


Mark Babin


Arfor Houwman

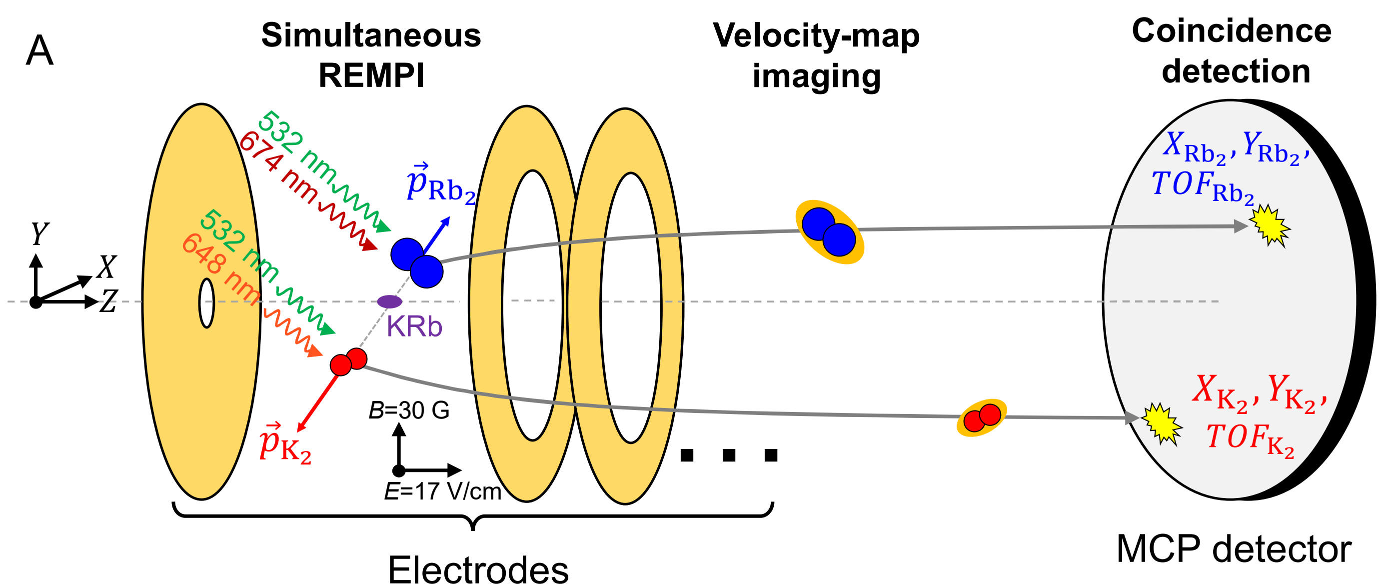
Science **384**, 1117 (2024)

What we need?

- ☑ Molecules in a *single* quantum state
- ☑ Chemical Reactions!
- ☑ State detection of products
- ☐ Coincidence detection of products from the same event
  - ☐ Avoid chaotic dynamics?

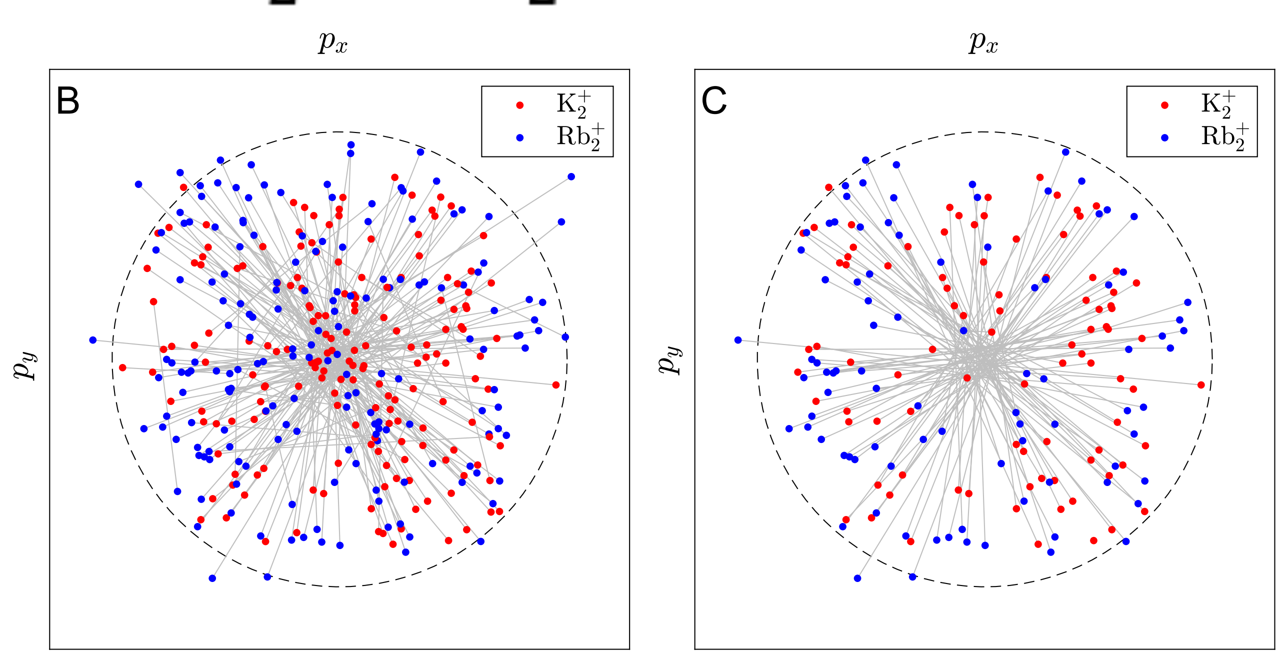
    Look in the nuclear spin degree of freedom?!
- ☐ Check to see coherence survives reactions

Coincidence detection of products



Momentum conservation constrains

$$\vec{P}_{K_2} + \vec{P}_{Rb_2} = \vec{P}_{KRb} + \vec{P}_{KRb} \rightarrow 0$$



- Identify product pairs created in the same reaction event
- Each K_2, Rb_2 rotation state pair corresponding to a possible outcome of the reaction

What we need?

- ☑ Molecules in a *single* quantum state
- ☑ Chemical Reactions!
- ☑ State detection of products
- ☑ Coincidence detection of products from the same event
- ☑ Avoid chaotic dynamics?

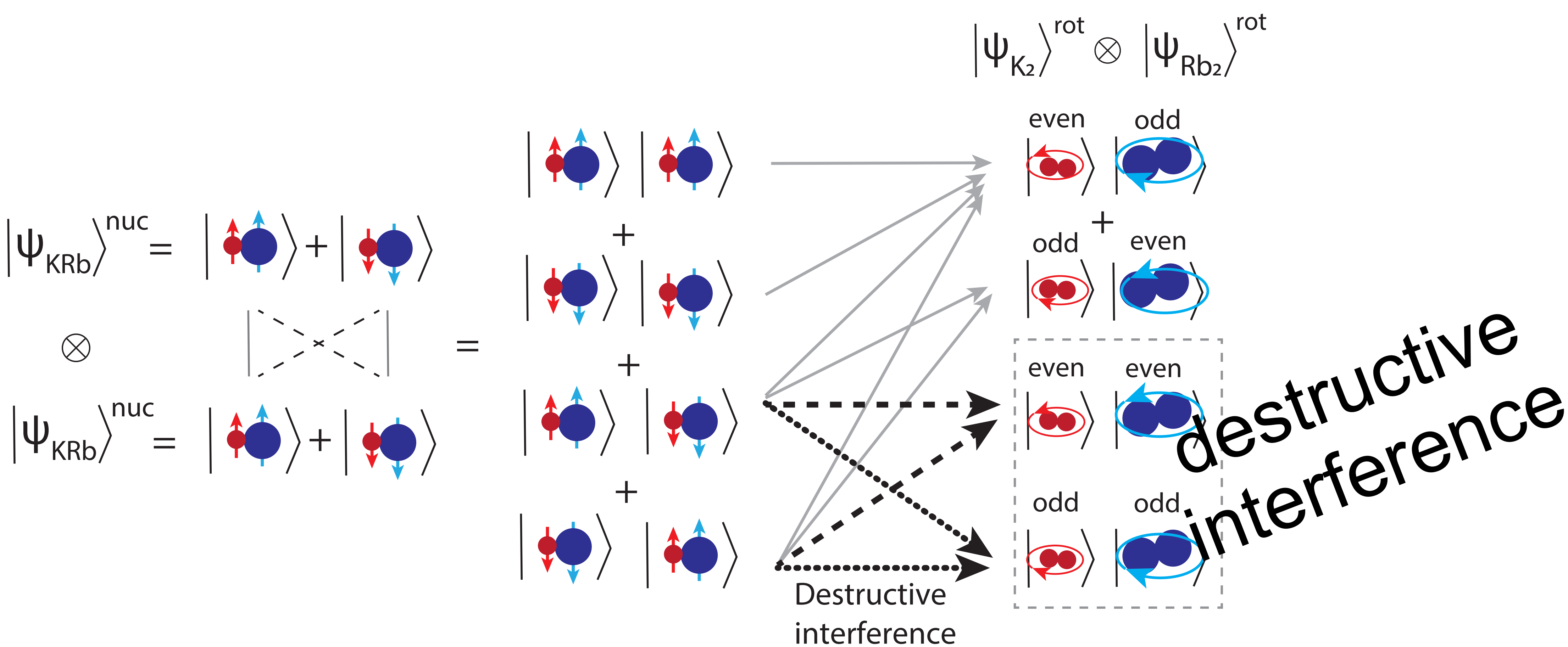
  Look in the nuclear spin degree of freedom?!
- ☐ Check to see coherence survives reactions

Assume coherence survives reactions

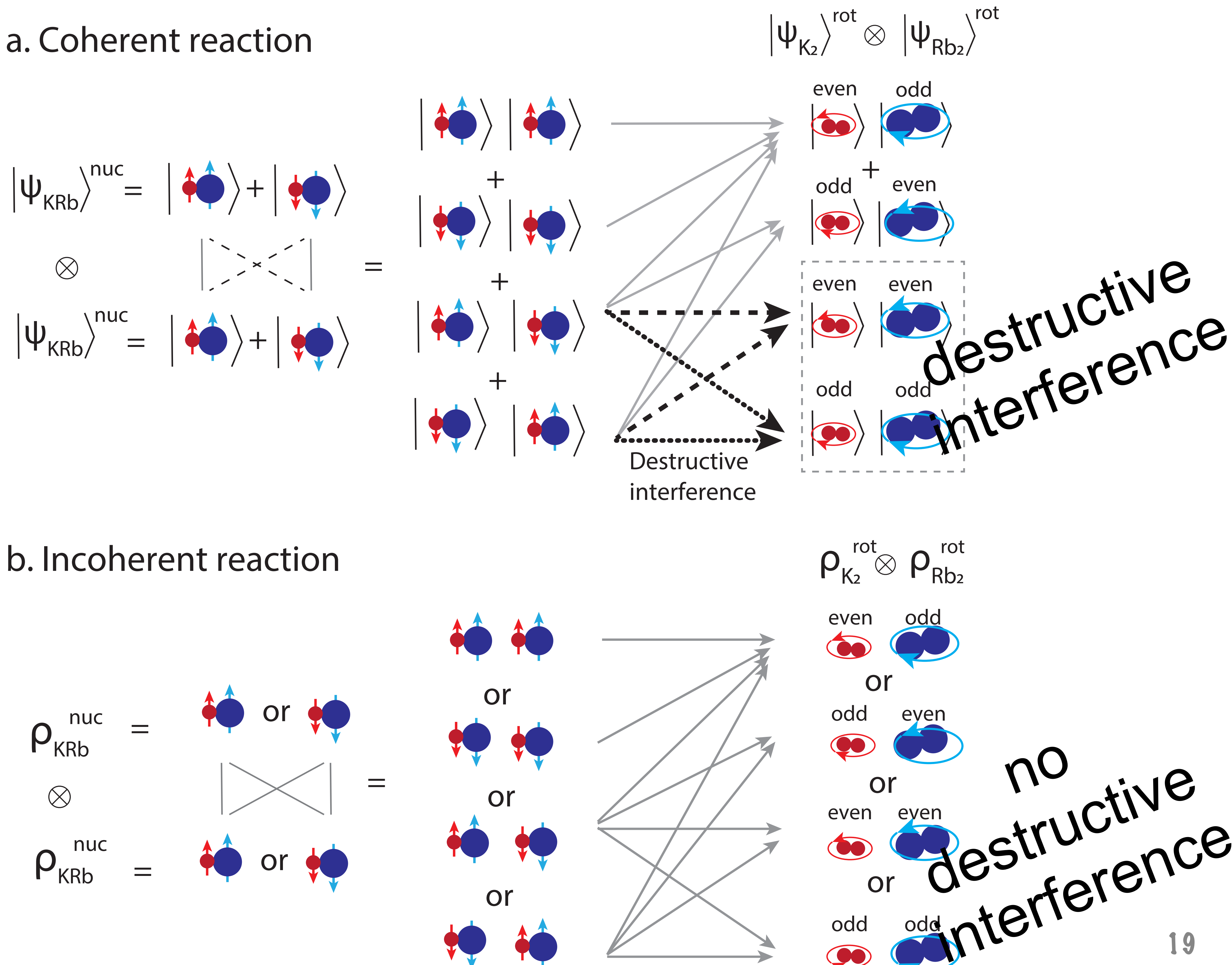
$$\frac{1}{\sqrt{2}}\Big(|\uparrow\mathrm{K}\uparrow\mathrm{Rb}\rangle+|\downarrow\mathrm{K}\downarrow\mathrm{Rb}\rangle\Big)_A\otimes\frac{1}{\sqrt{2}}\Big(|\uparrow\mathrm{K}\uparrow\mathrm{Rb}\rangle+|\downarrow\mathrm{K}\downarrow\mathrm{Rb}\rangle\Big)_B$$

$$\frac{1}{2}\Big(|\uparrow\mathrm{K_A}\uparrow\mathrm{K_B};\uparrow\mathrm{Rb_A}\uparrow\mathrm{Rb_B}\rangle+\underline{|\uparrow\mathrm{K_A}\downarrow\mathrm{K_B};\uparrow\mathrm{Rb_A}\downarrow\mathrm{Rb_B}\rangle+|\downarrow\mathrm{K_A}\uparrow\mathrm{K_B};\downarrow\mathrm{Rb_A}\uparrow\mathrm{Rb_B}\rangle}+|\downarrow\mathrm{K_A}\downarrow\mathrm{K_B};\downarrow\mathrm{Rb_A}\downarrow\mathrm{Rb_B}\rangle\Big)$$

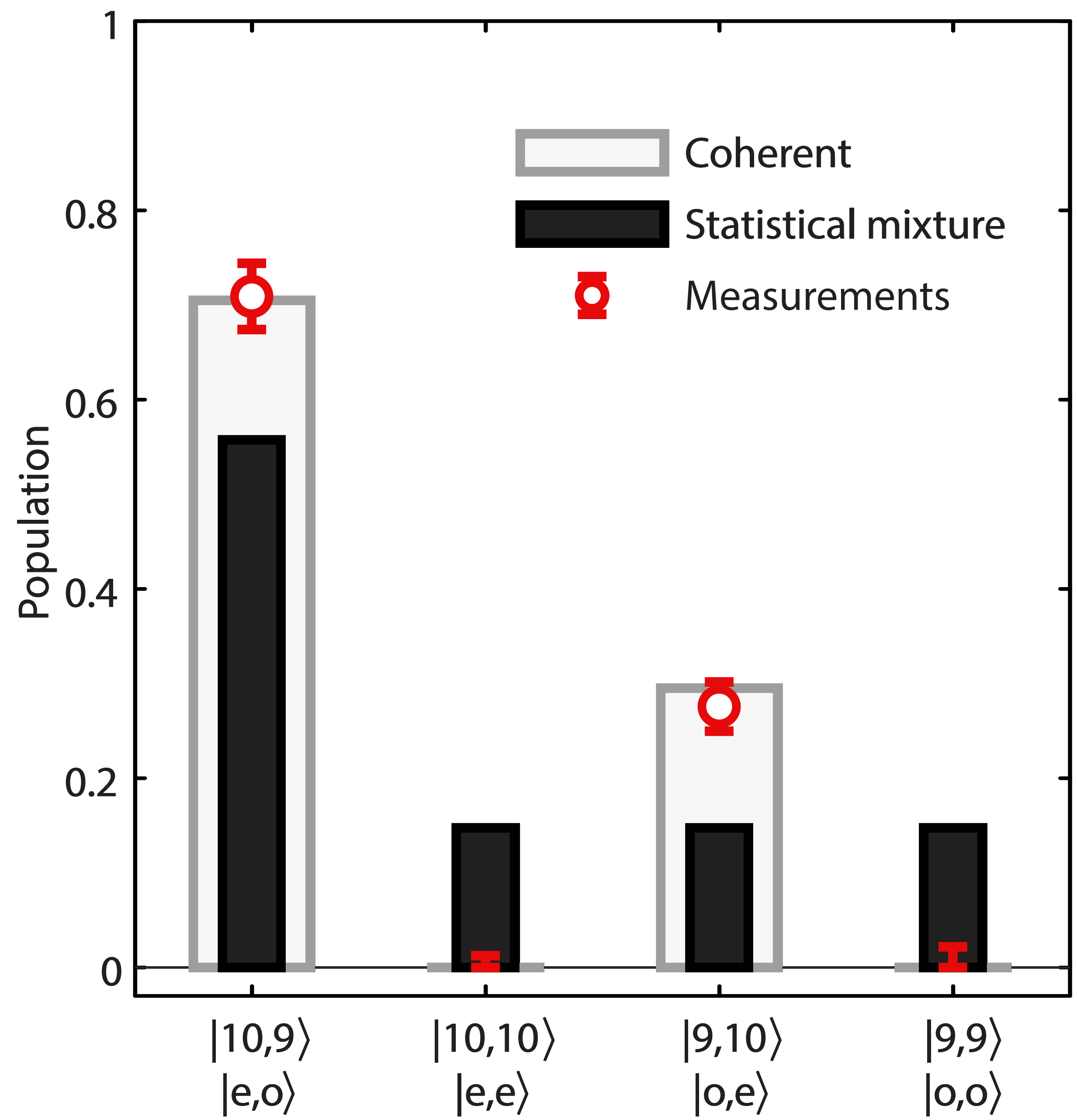
need to be properly symmetrized because indistinguishability



Coherence vs Incoherence in reactions



Let's check!



Science 384, 1117 (2024)

Coherence is [0.9014, 1] (95% confidence level)

What we need?

☑ Check to see coherence survives reactions

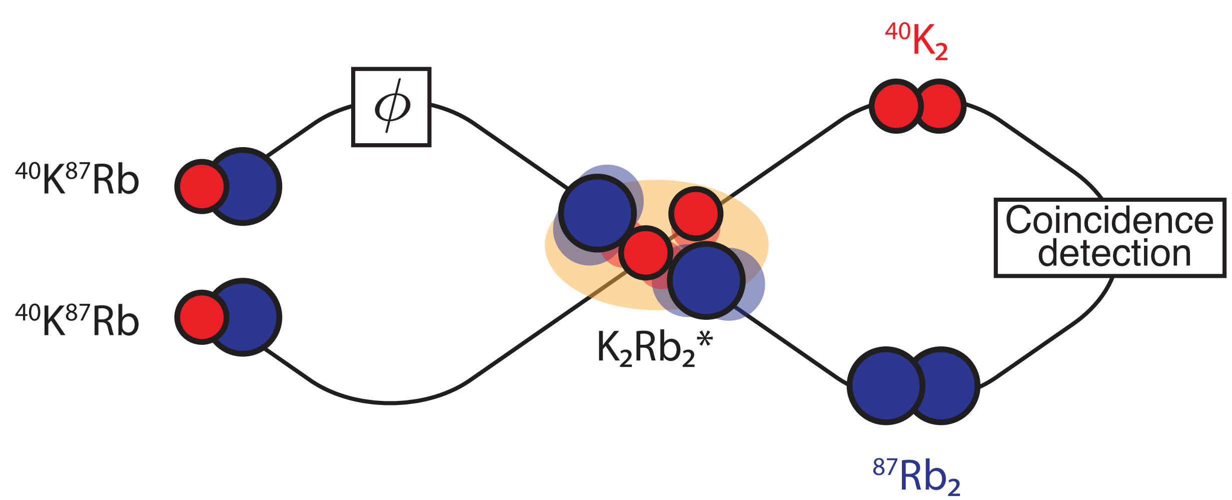
This *implies* the products are entangled

Open Questions

* Entangled products need to be rigorously shown via a direct phase measurement, but we have only measured populations

* What would break the phase coherence of the reactions? A new way to scrutinize the short-range physics and the role of nuclear spins in reactions. How general is it?

* How do we measure and alter the phase directly?

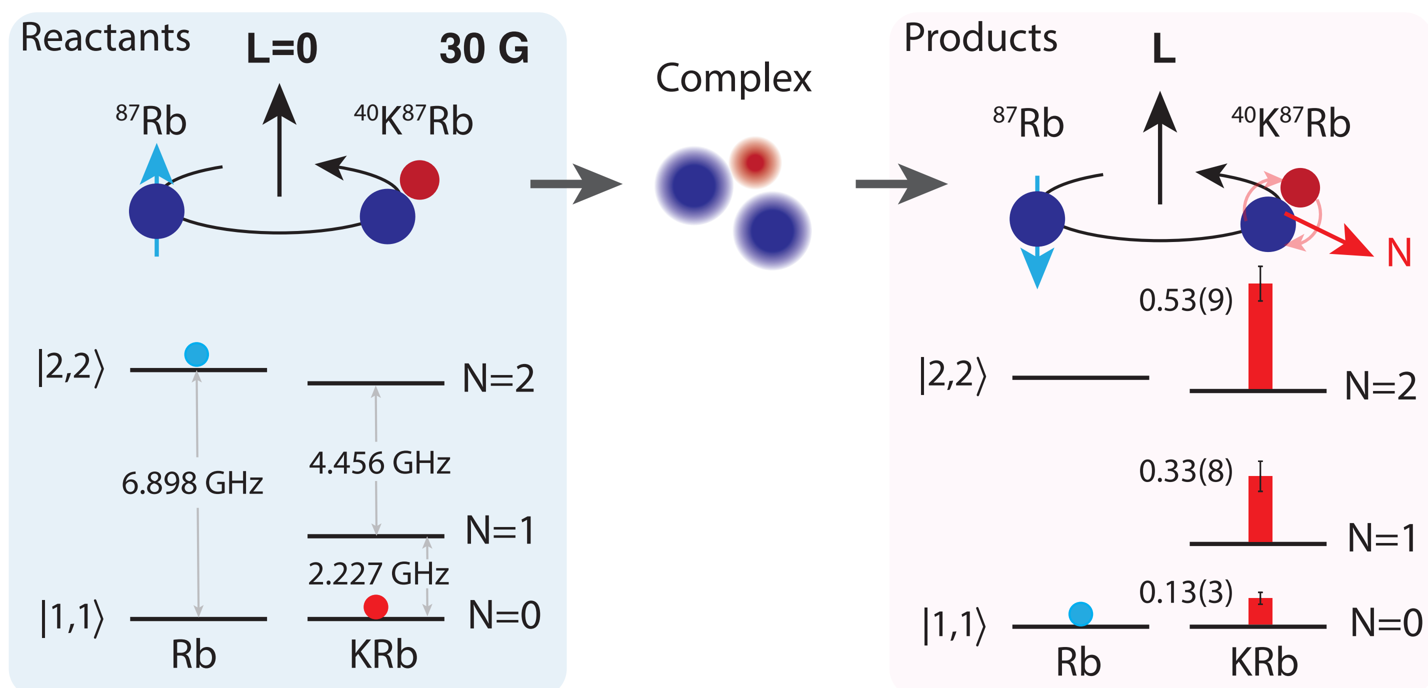


reaction interferometer

Luke et al.,
Faraday Discussion 2024

Hyperfine-to-Rotational energy transfer in ultracold atom-molecule collisions

YX Liu*, LB Zhu* et al., in collaboration with Tscherbul, Tomza, Bohn (arXiv:2407.08891)



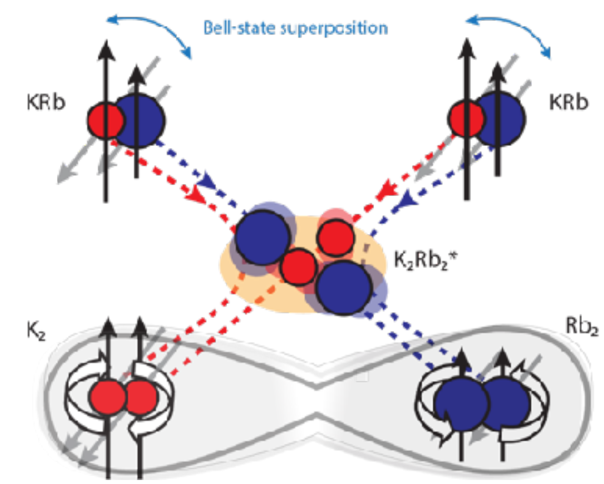
- Spin-dependent interaction is important in KRb_2^* complex ($\times 10^5$ long-lived complex a consequence?)

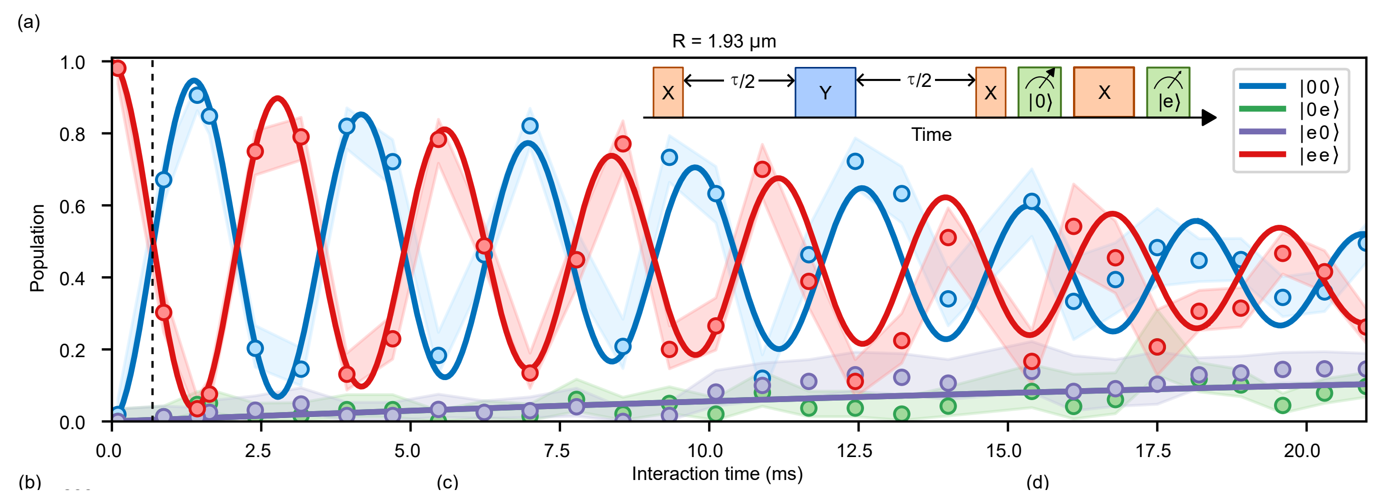
- Conical intersection lies below the collision energy!

- "Statistical" branching ratio cannot be explained by tour-de-force quantum scattering calculation assuming Rigid Rotor of KRb. Considering vibration will be important for a quantitative agreement!!

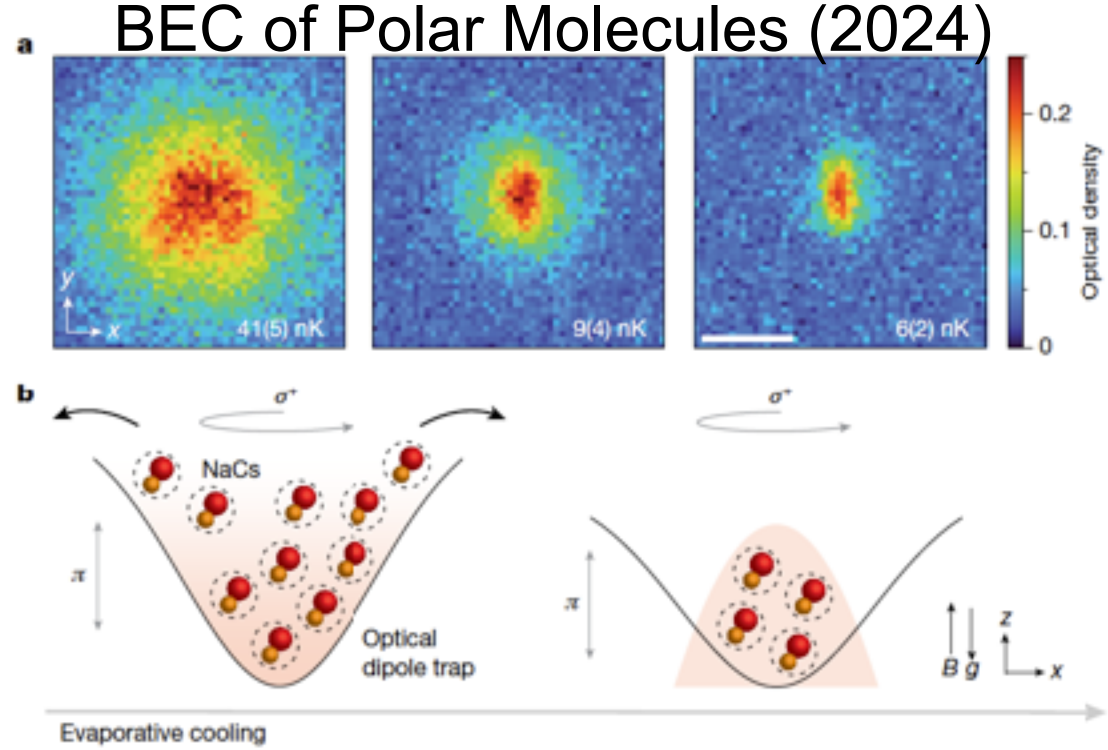
Summary and Outlook

* Individual molecules under control!
* Fast, coherent dipolar interaction in a “programmable” tweezer array of molecules
* iSWAP gate with molecules demonstrated!
* entanglement fidelity limited by thermal motion -> a path forward
* Phase coherence is preserved for nuclear spins in $KRb + KRb \rightarrow K_2 + Rb_2$ reactions. Is it general?
* Harnessing molecular resources for quantum science





Sebastian Will Group (Columbia)
BEC of Polar Molecules (2024)



Thank you!

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Will Cairncross (Atom Computing)
Yichao Yu (Duke)
Jon Hood (Purdue)
Ming-Guang Hu (QuEra Computing)
Yu Liu (NIST)
Matt Nichols (Apple)
Lee Liu (JILA)
Nick Hutzler (Caltech)
Yen-Wei Lin (Intelon Optics)
David Grimes (MIT)

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Guildo Pupillo
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Hossein Sadeghpour

Tijs Karman

Timur Tscherbul

Michal Tomza

Goulven Quemener

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