

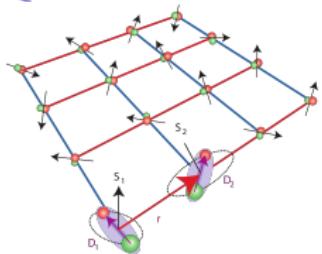
Building Single Molecules from Single Atoms

Yichao Yu

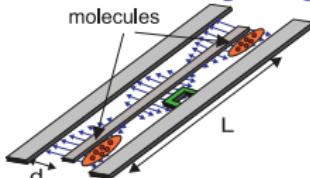
Ni Group/Harvard

Jul. 2020

Quantum Simulation



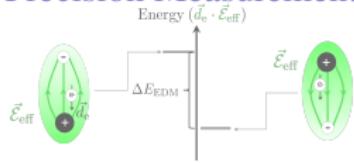
Quantum Computing



PRL. 97, 33003 (2006)

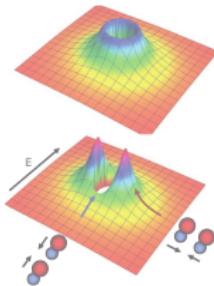
Nat. Phys. 2, 341 (2006)

Precision Measurement



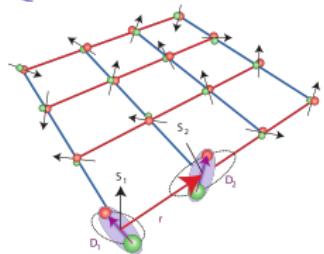
Science 343, 269 (2014)

Quantum Chemistry



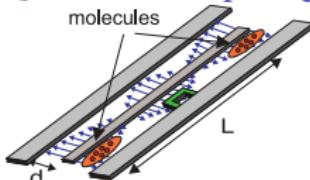
Nature 464, 1324 (2010)

Quantum Simulation



Nat. Phys. 2, 341 (2006)

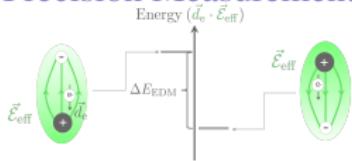
Quantum Computing



PRL. 97, 33003 (2006)

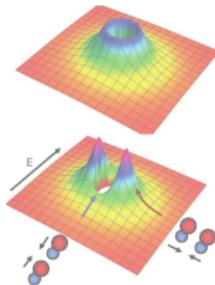
- Full quantum control
- Entanglement
- ...

Precision Measurement



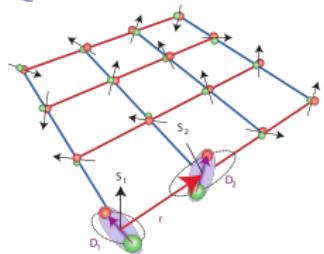
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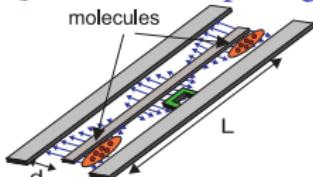
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Quantum Simulation



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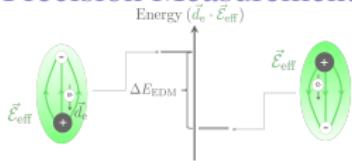
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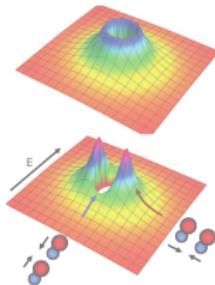
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Precision Measurement



Science 343, 269 (2014)

Quantum Chemistry



Nature 464, 1324 (2010)

New Approach?

Entanglement

Single particle control

Entanglement

i.e. interaction

Single particle control

Entanglement

i.e. interaction

Single particle control

Dipolar molecules

Dipolar molecules

- Strong and tunable interaction
($\approx k\text{Hz}$ at $\approx \mu\text{m}$ distance)
 - ▶ Fast gate operations
 - ▶ Long coherence time
- Rich internal structure
(Electronic, vibrational,
rotational, hyperfine, etc.)

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Single particle control

Optical tweezers

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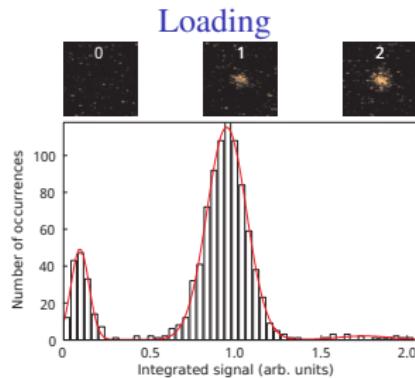
- Single site resolution



Entanglement

i.e. interaction

Dipolar molecules



Nat. Phys. 6, 951 (2010)

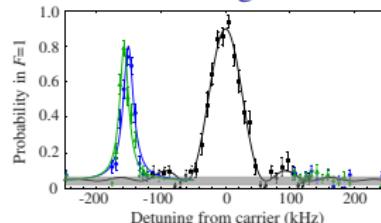
Single particle control

Optical tweezers

- Single site resolution

- ...

Cooling



PRX. 2, 041014 (2012)

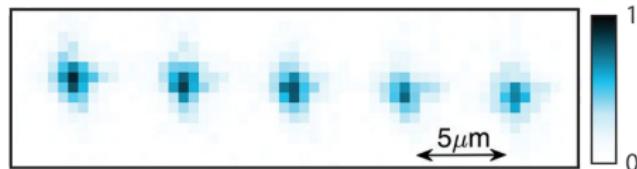
Rearranging



Science 354, 1024 (2016)

Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

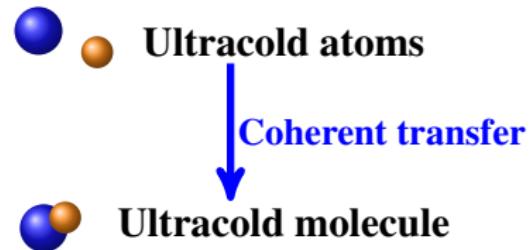
Ultracold molecule in tweezers

Direct cooling



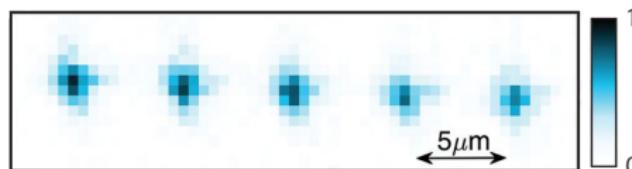
Science 365, 1156 (2019)

Assembly



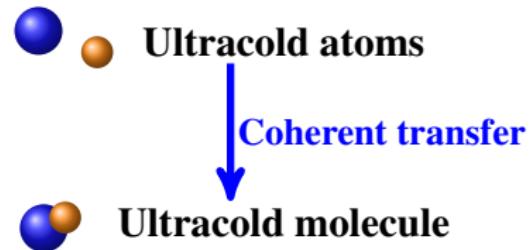
Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezer
- Quantum control

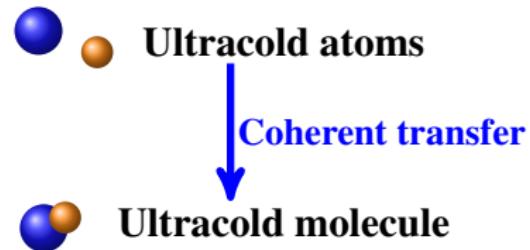
Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezers
- Quantum control
- Control of atoms
- Coherent creation of molecules

Outline

1 Experiment overview

2 Atom state control

- Raman sideband cooling of Na atoms

3 Atom-atom interaction

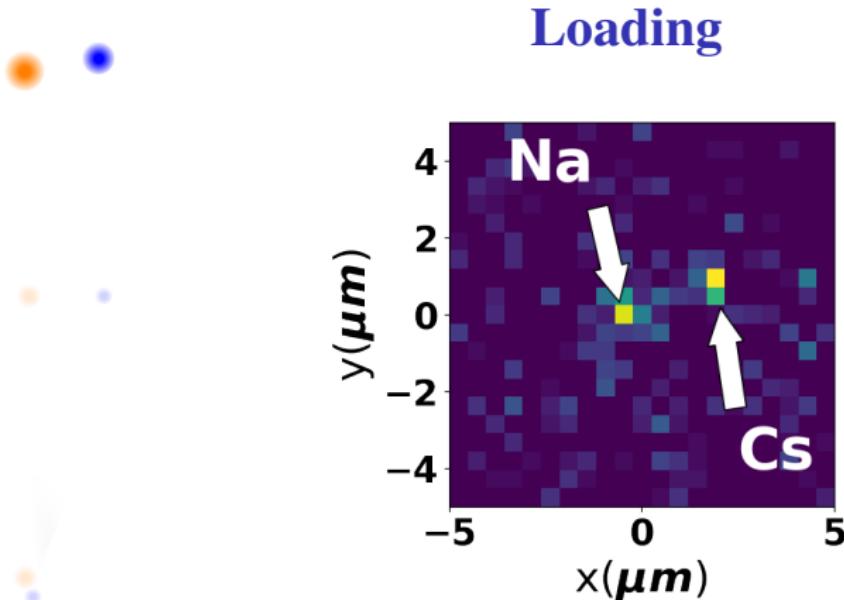
4 Optical molecule creation

5 Conclusion

NaCs molecule

- Bi-alkali
- Large dipole moment: 4.6D

Experiment overview

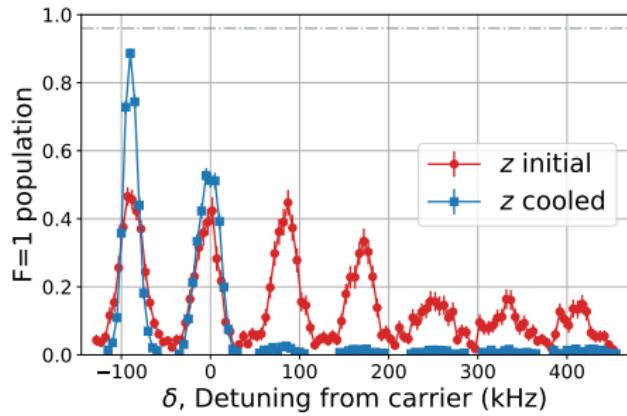


Loading probability per site: 60%
Post select on initial and final state.

Experiment overview



Cooling

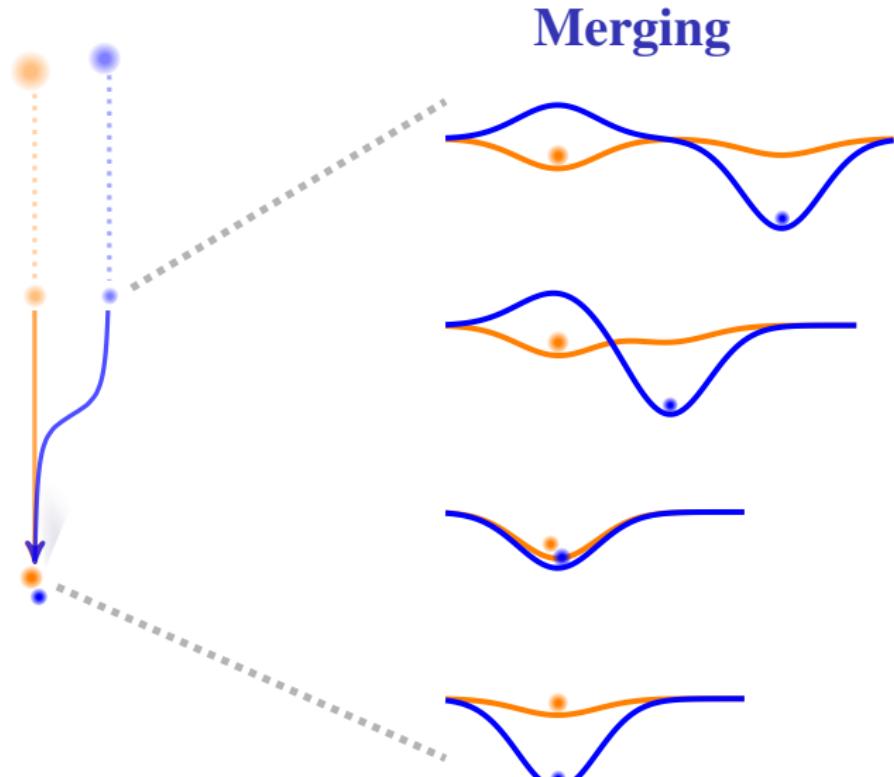


Cs: 96% ground state¹
Na: 94% ground state²

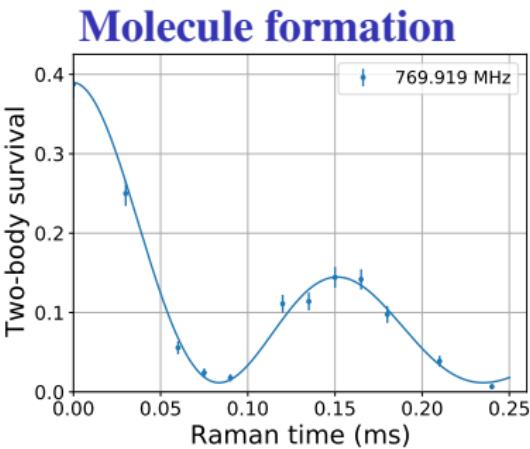
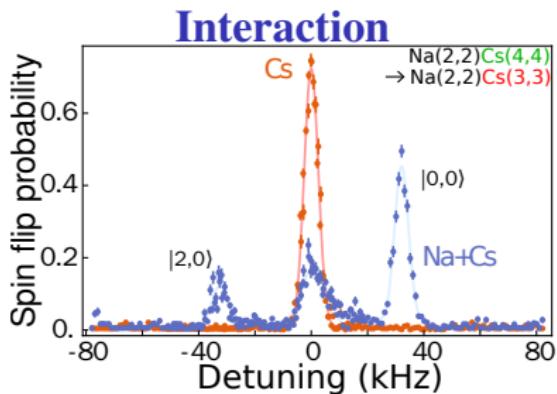
¹PRX 9, 021039

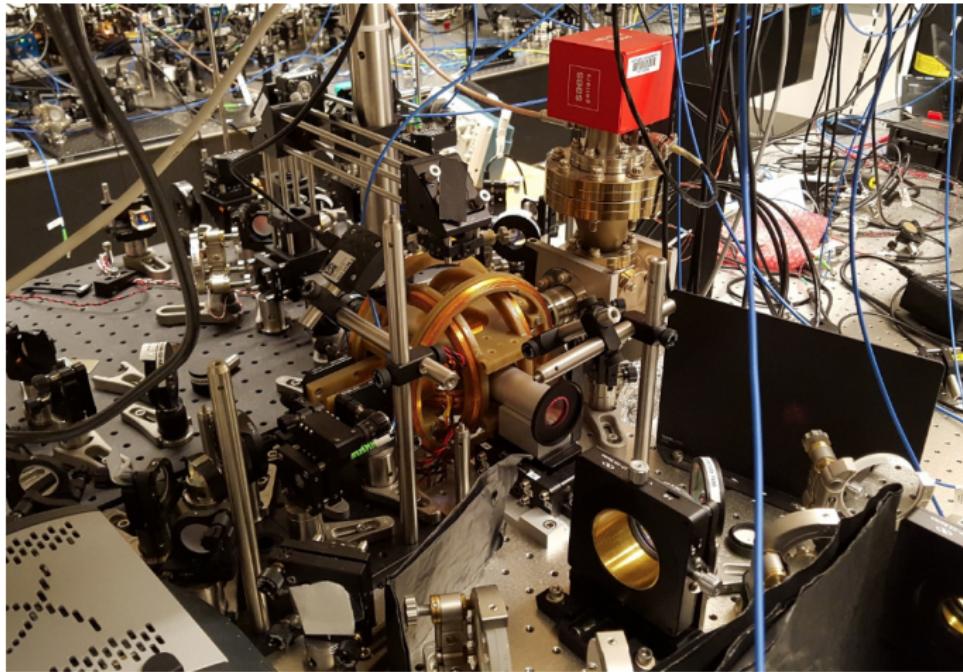
²PRA 97, 063423

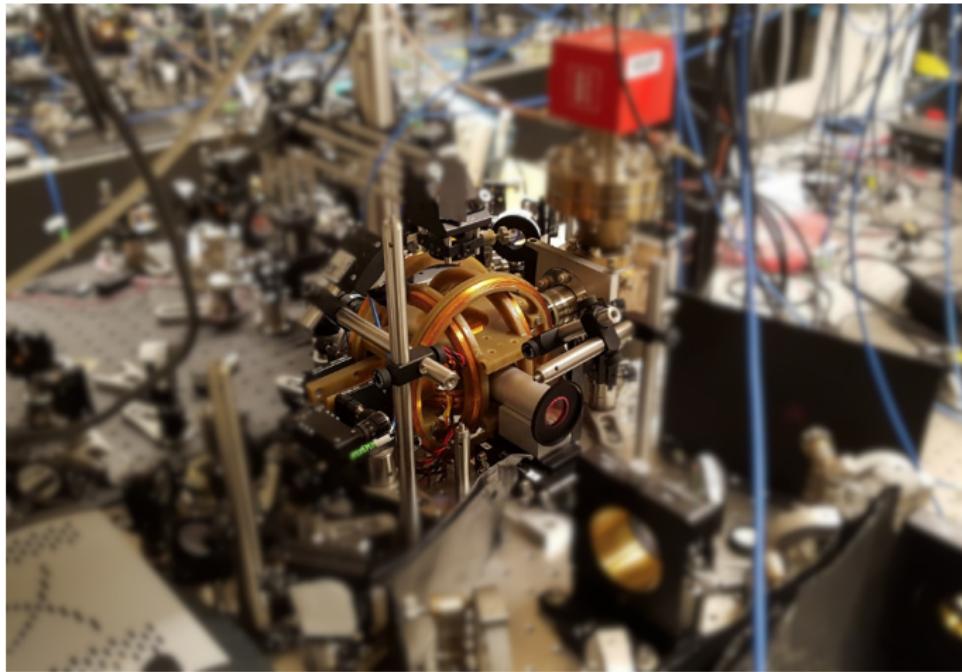
Experiment overview

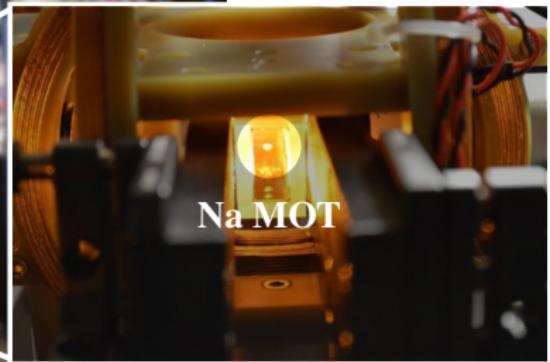
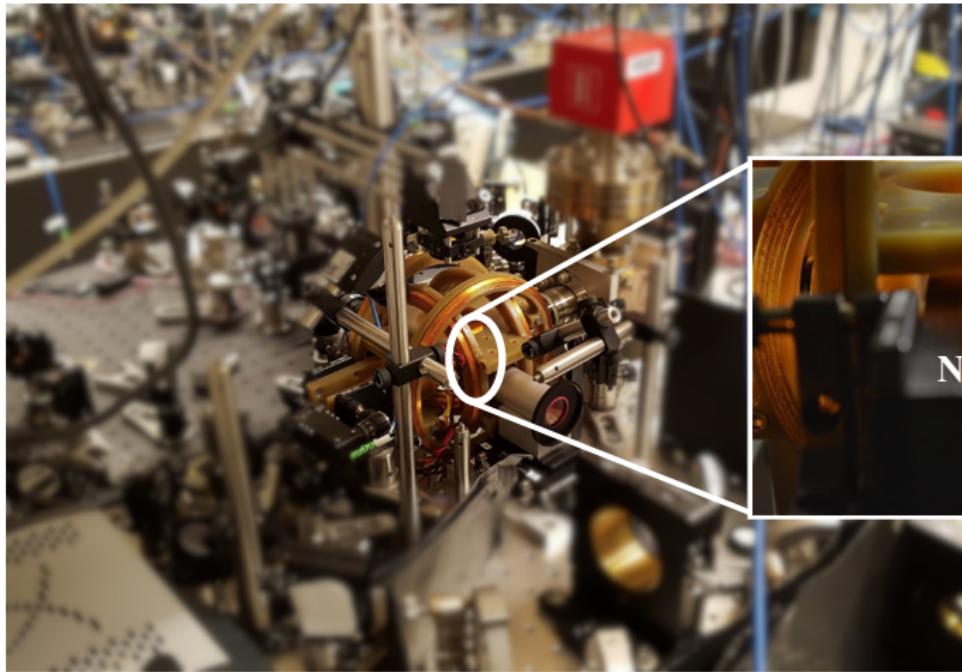


Experiment overview

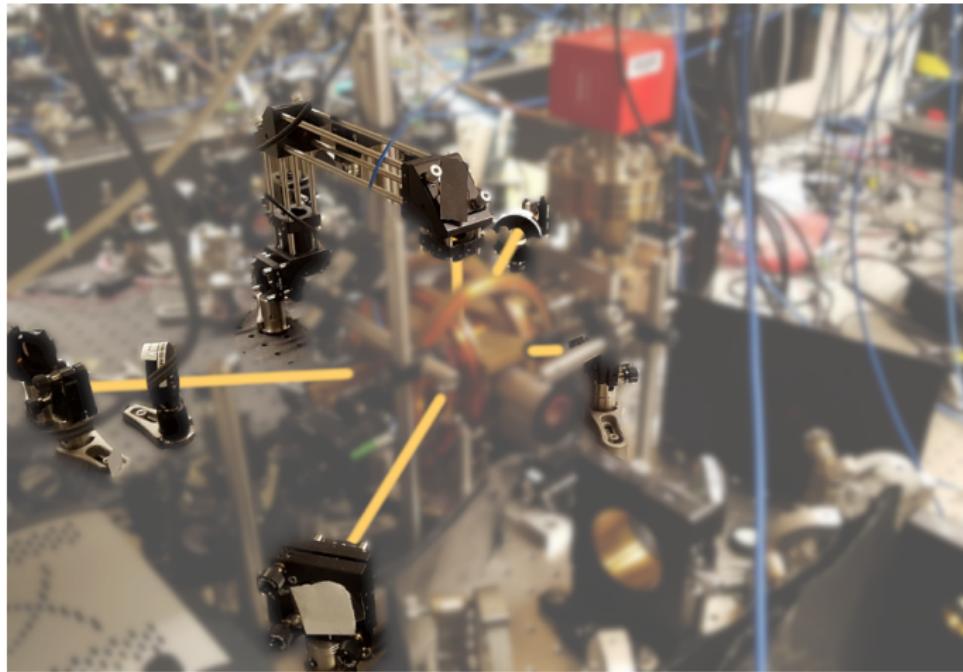




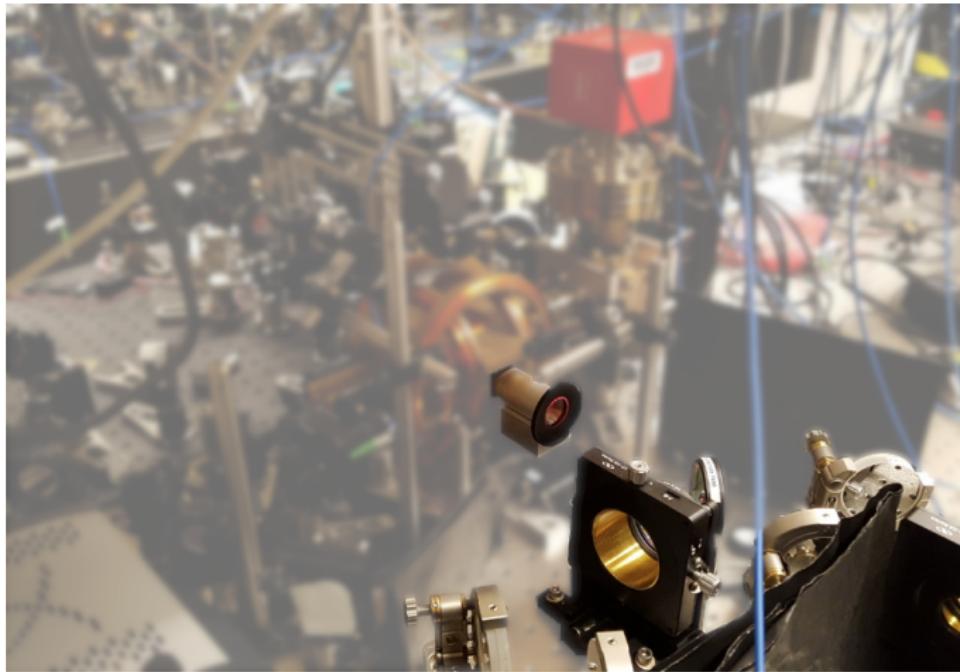




MOT beam path



Tweezer beam path



Outline

1 Experiment overview

2 Atom state control

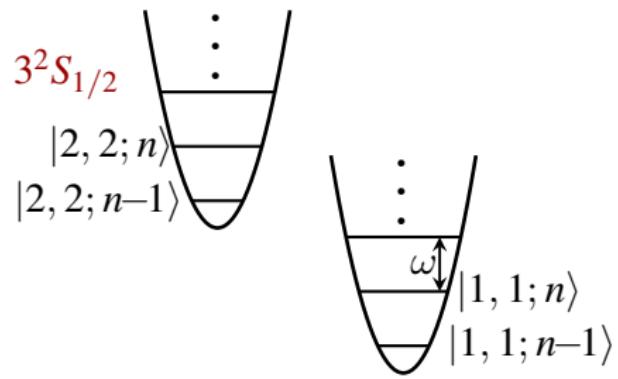
- Raman sideband cooling of Na atoms

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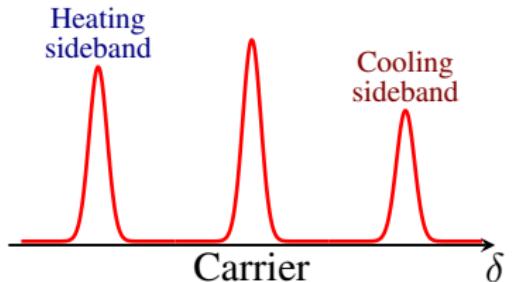
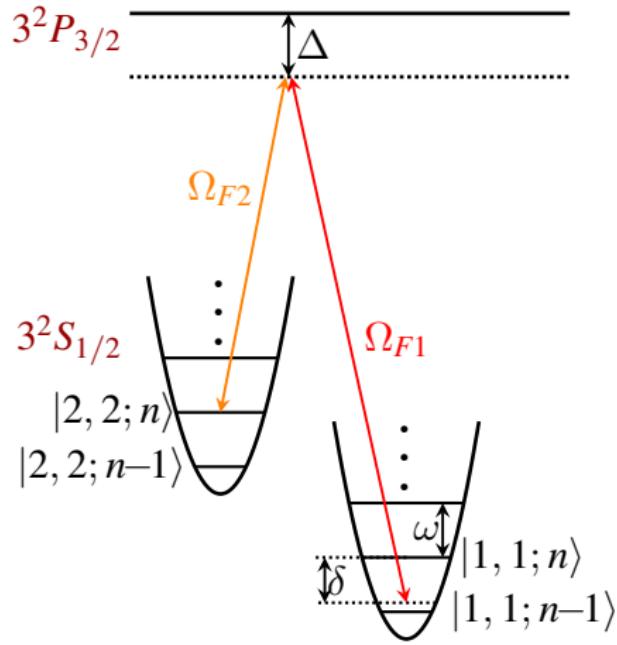
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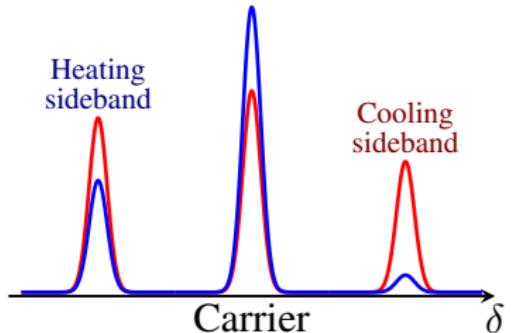
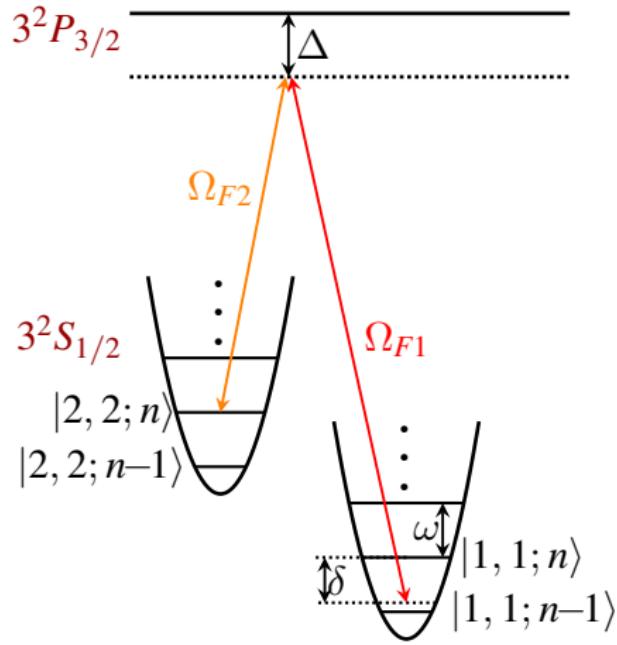
Raman sideband cooling



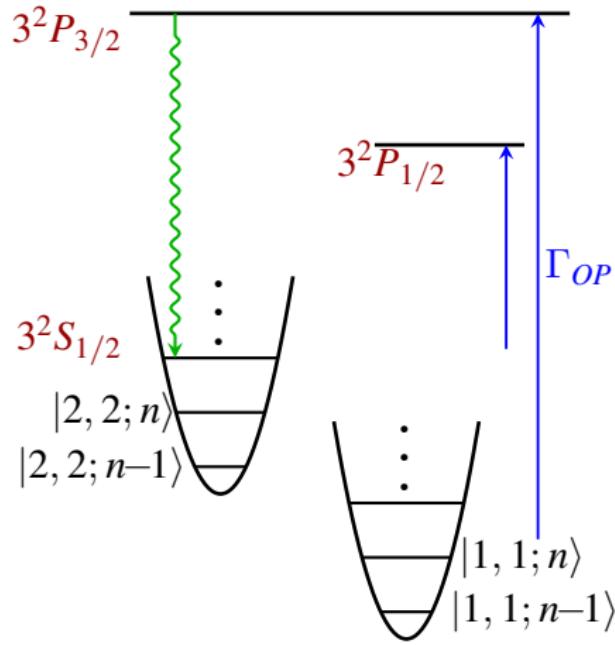
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Raman sideband cooling



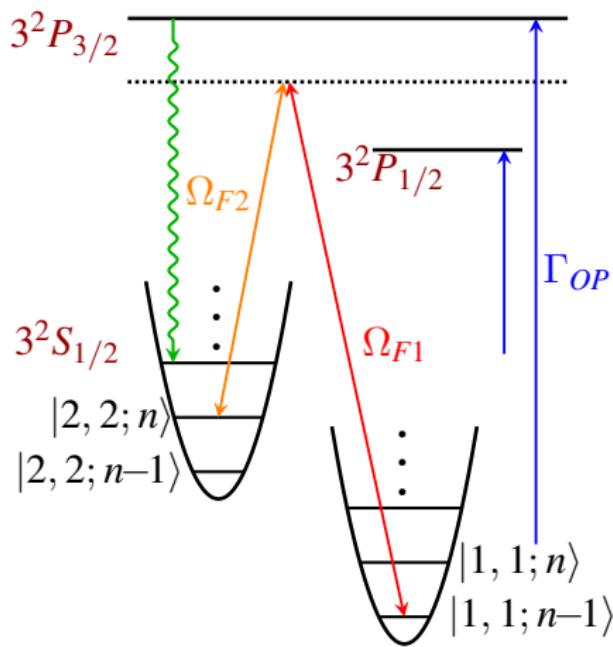
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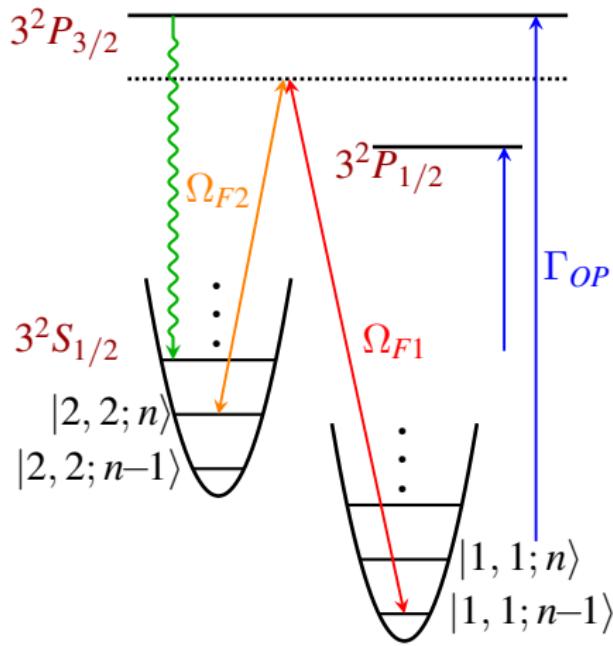
Raman sideband cooling

Lamb Dicke parameter

$$\eta \equiv kz_0 = \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{recoil}}{\omega_{trap}}}$$



Raman sideband cooling

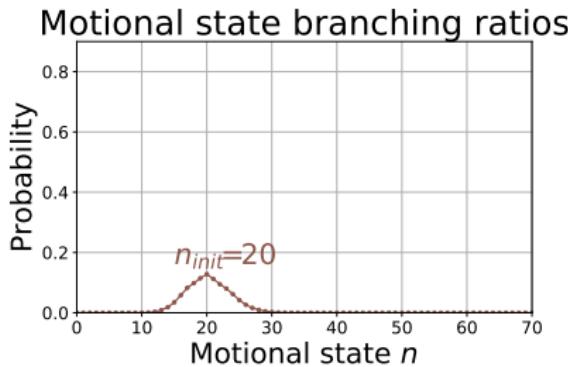


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$$\eta_{Na}^{OP} = 0.55$$

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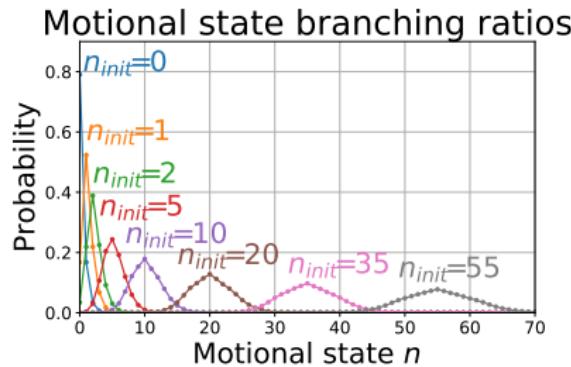
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- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



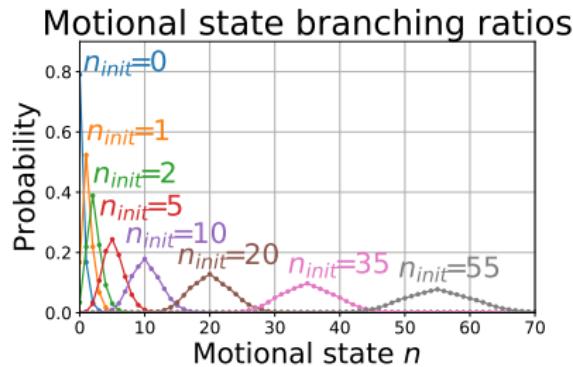
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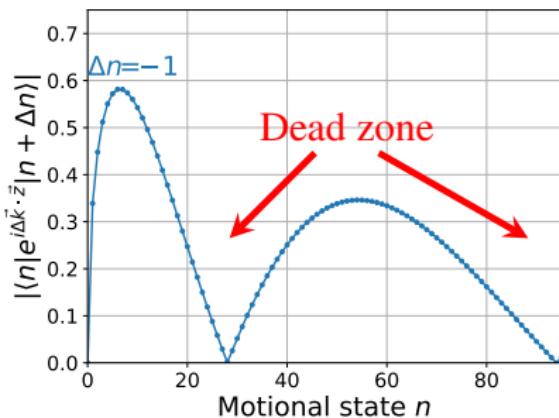


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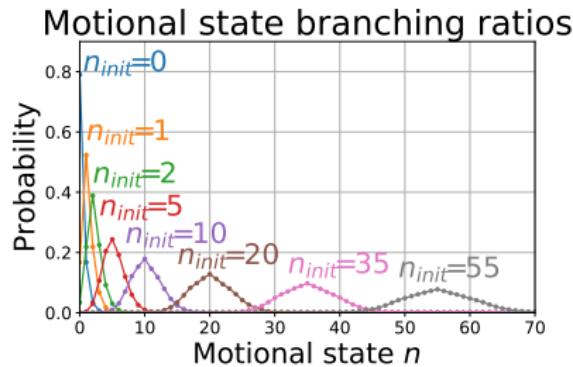
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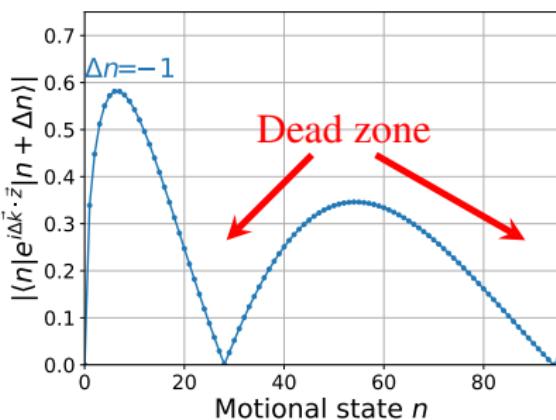


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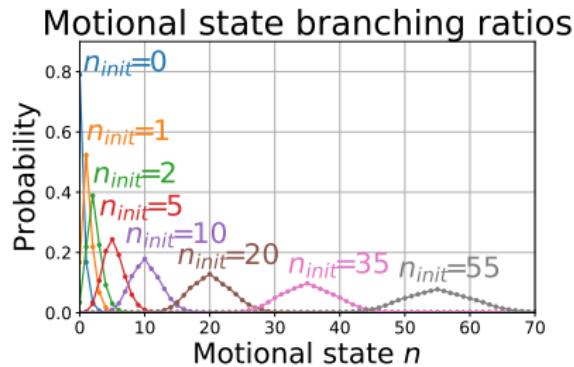
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Solution

Use higher order sidebands.

Raman sideband cooling

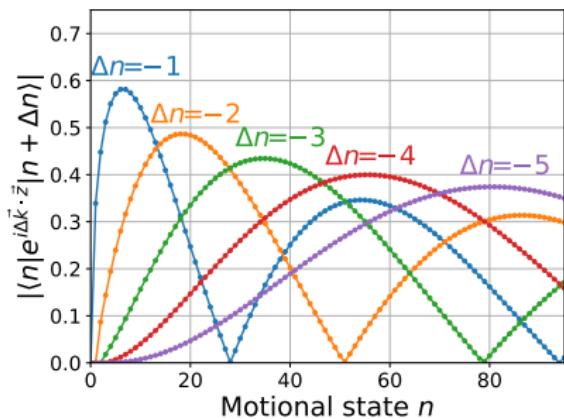


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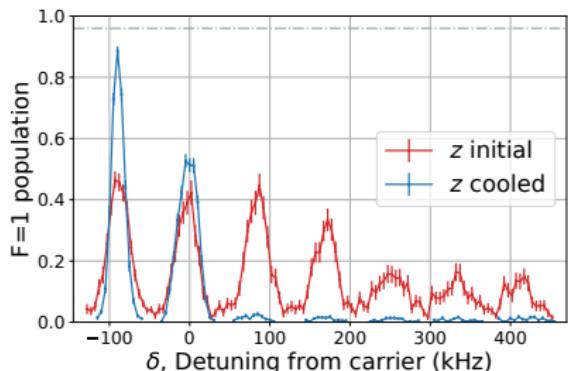


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Axial sideband spectrum

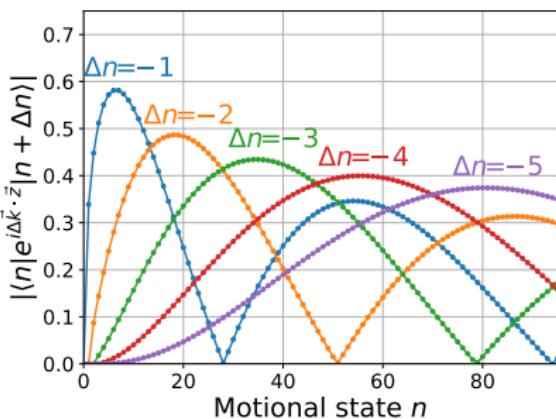


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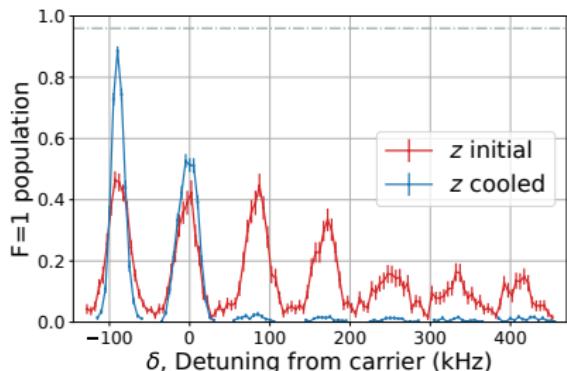


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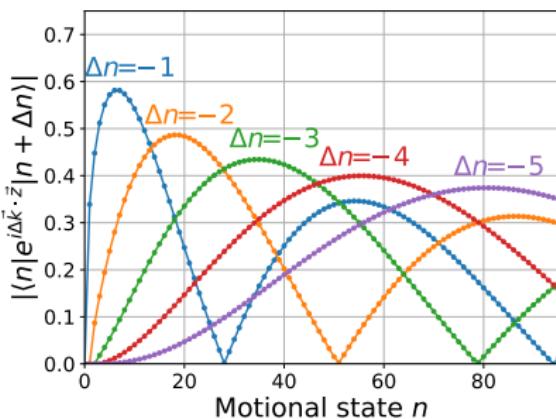


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3D ground state: 93.5(7)%

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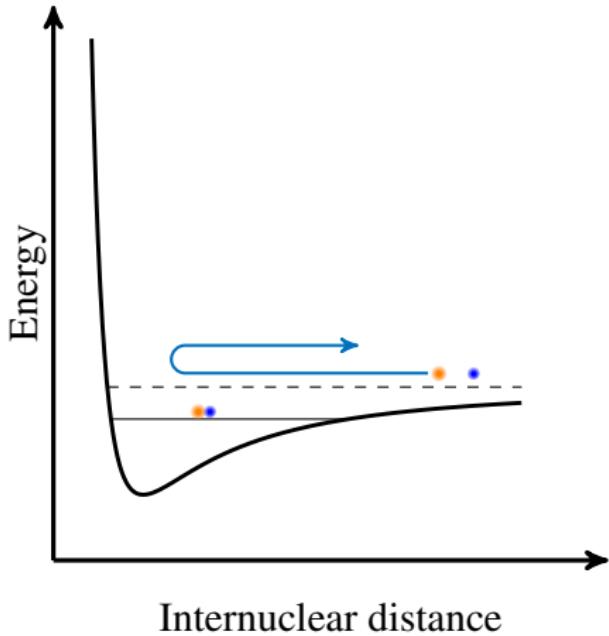
3 Atom-atom interaction

4 Optical molecule creation

5 Conclusion

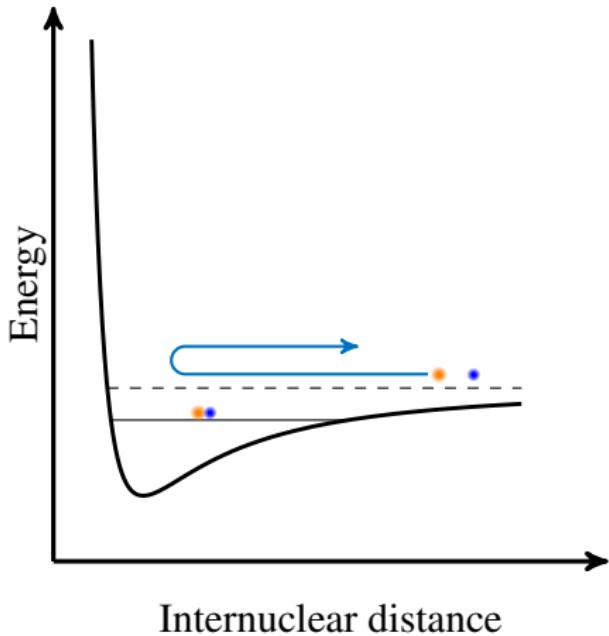
Scattering length a

- Binding energy
- Molecular potential
- Feshbach resonance
- Molecule formation
- ⋮



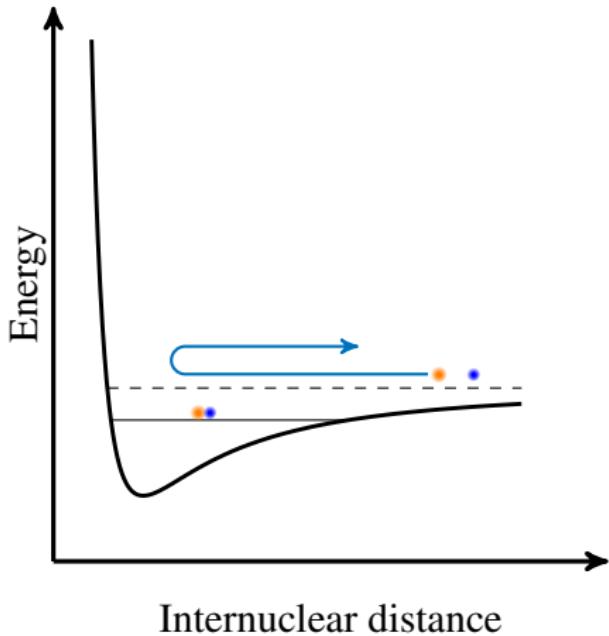
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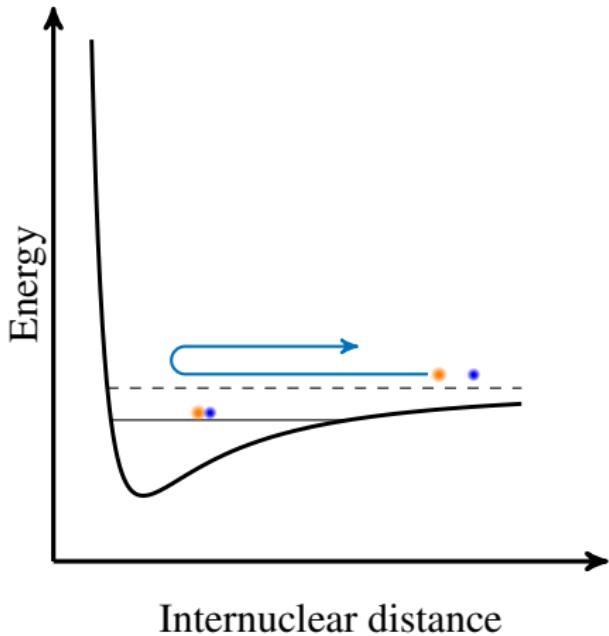
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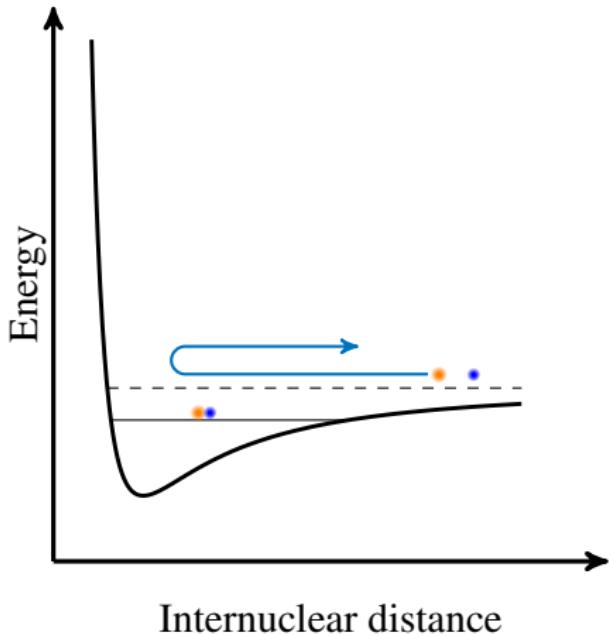
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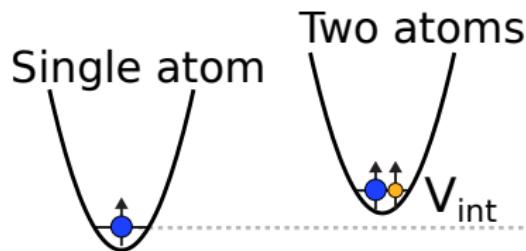


Scattering length a

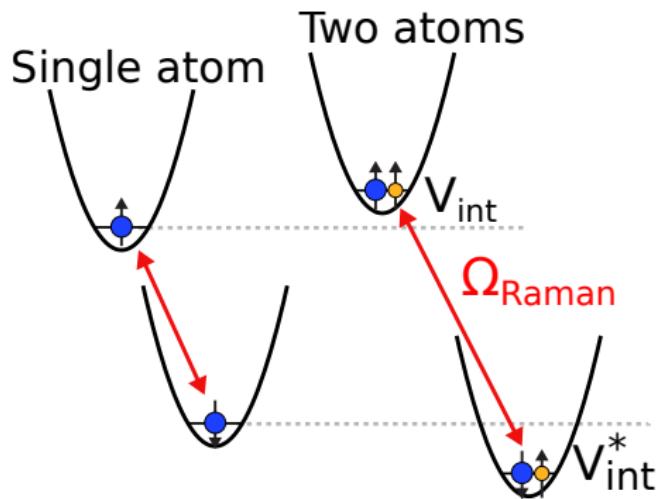
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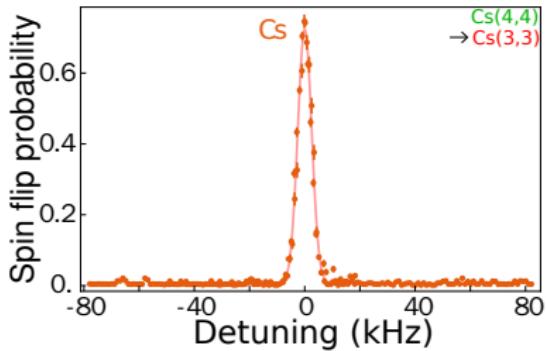
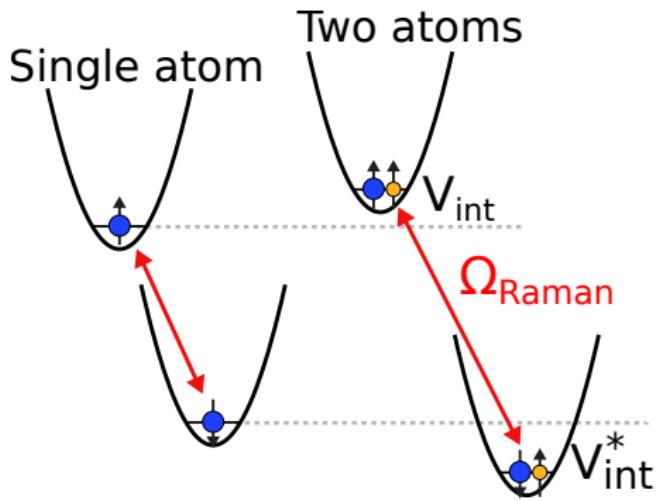
Interaction shift



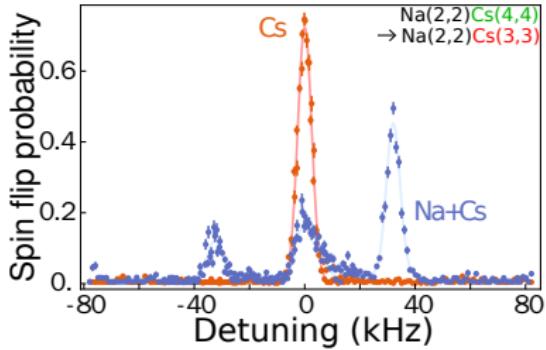
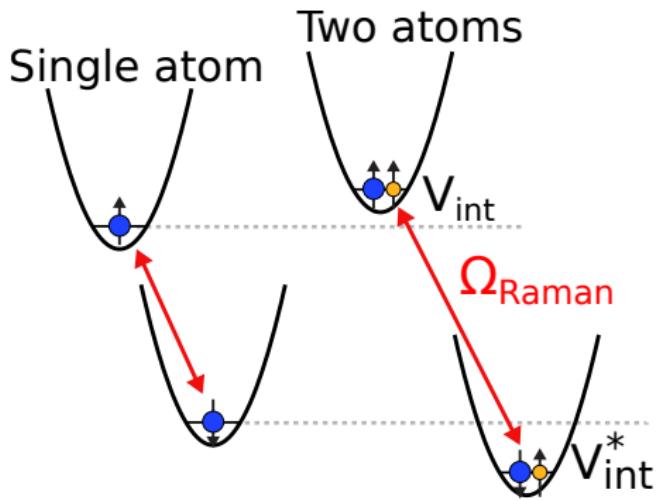
Interaction shift



Interaction shift



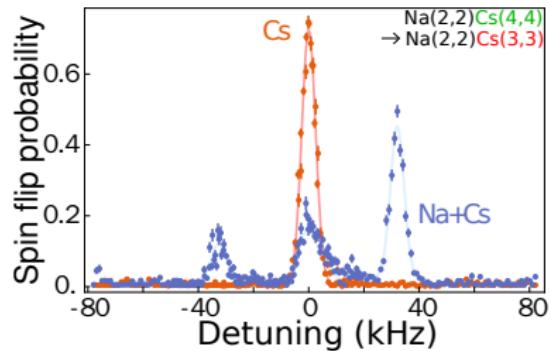
Interaction shift



Interaction shift

$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{m_1 \omega_{1,i}^2 x_{1,i}^2}{2} + \frac{p_{1,i}^2}{2m_1} \right)}_{\text{Na}} + \underbrace{\sum_{i=x,y,z} \left(\frac{m_2 \omega_{2,i}^2 x_{2,i}^2}{2} + \frac{p_{2,i}^2}{2m_2} \right)}_{\text{Cs}} + V_{int}(\vec{r}_1 - \vec{r}_2)$$

Interaction



Interaction shift

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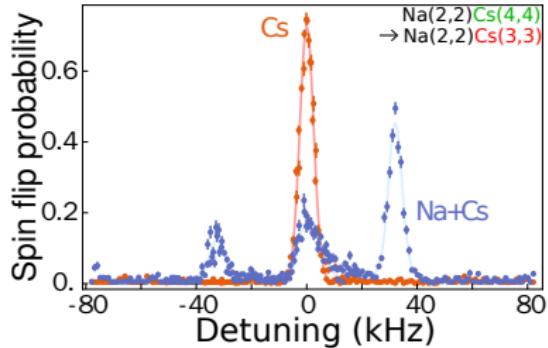
To center of mass
and relative coordinates

$$M = m_1 + m_2 \quad \mu = \frac{m_1 m_2}{m_1 + m_2}$$

$$\Omega_i^2 = \frac{m_1\omega_{1,i}^2 + m_2\omega_{2,i}^2}{m_1 + m_2} \quad \omega_{R,i}^2 = \frac{m_2\omega_{1,i}^2 + m_1\omega_{2,i}^2}{m_1 + m_2}$$

$$X_i = \frac{m_1 x_{1,i} + m_2 x_{2,i}}{m_1 + m_2} \quad x_{R,i} = x_{1,i} - x_{2,i}$$

$$P_i = p_{1,i} + p_{2,i} \quad p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$



Center of mass

$$H = \overbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}$$

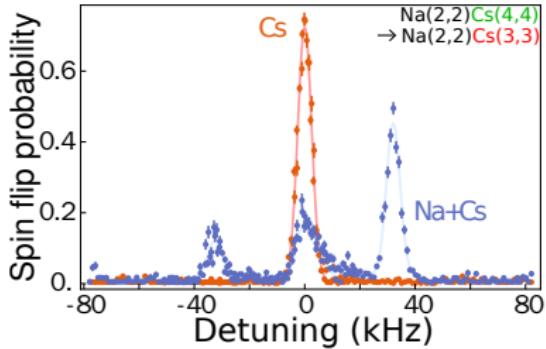
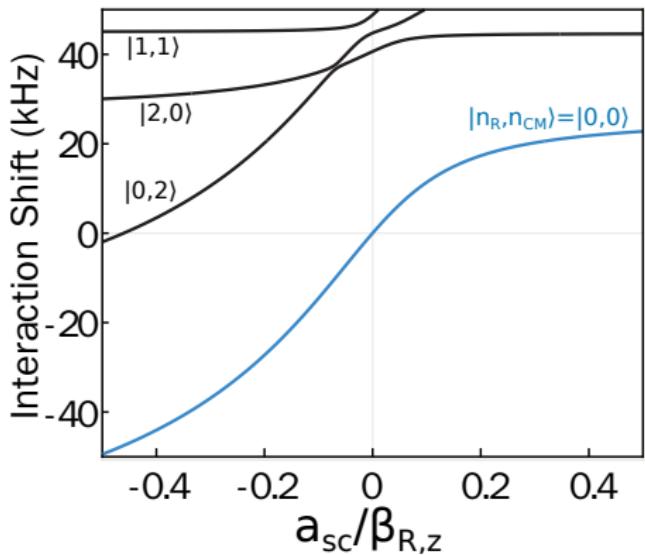
Relative

$$+ \underbrace{\sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right)}_{V_{int}(\vec{r}_R)} + V_{ext}(\vec{r}_R)$$

Mixing

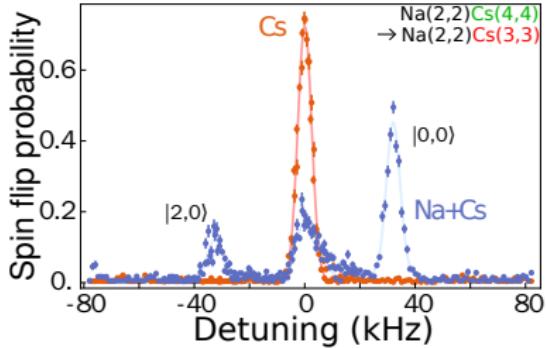
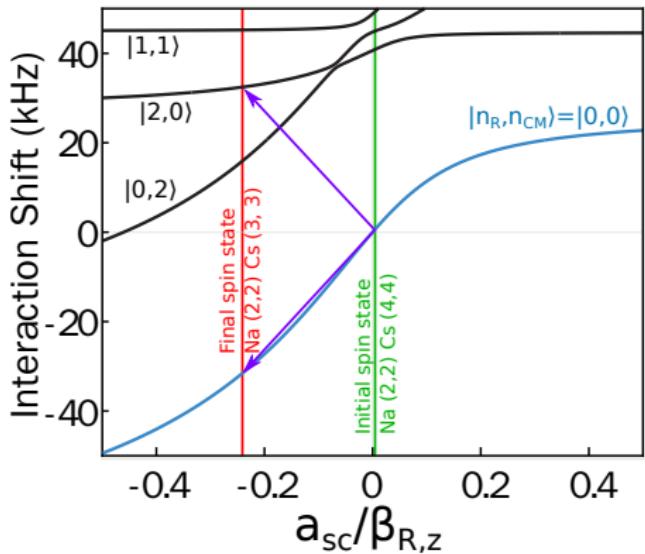
$$+ \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i x_{R,i}}$$

Interaction shift



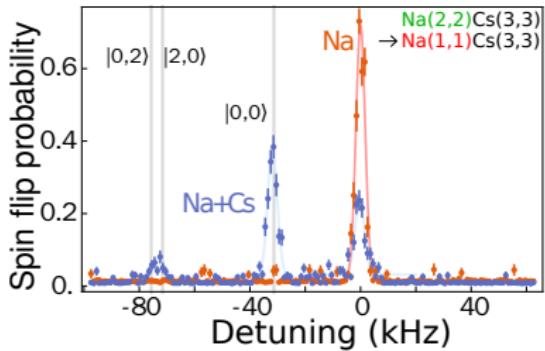
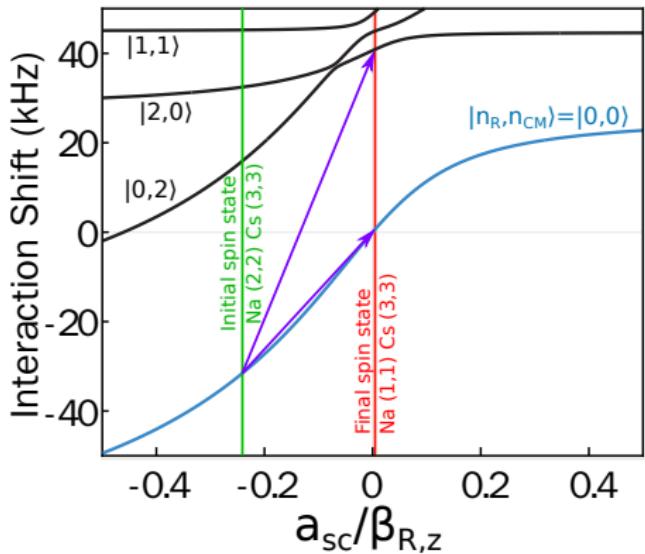
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i X_{R,i}}_{\text{Mixing}}$$

Interaction shift



$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i X_{R,i}}_{\text{Mixing}}$$

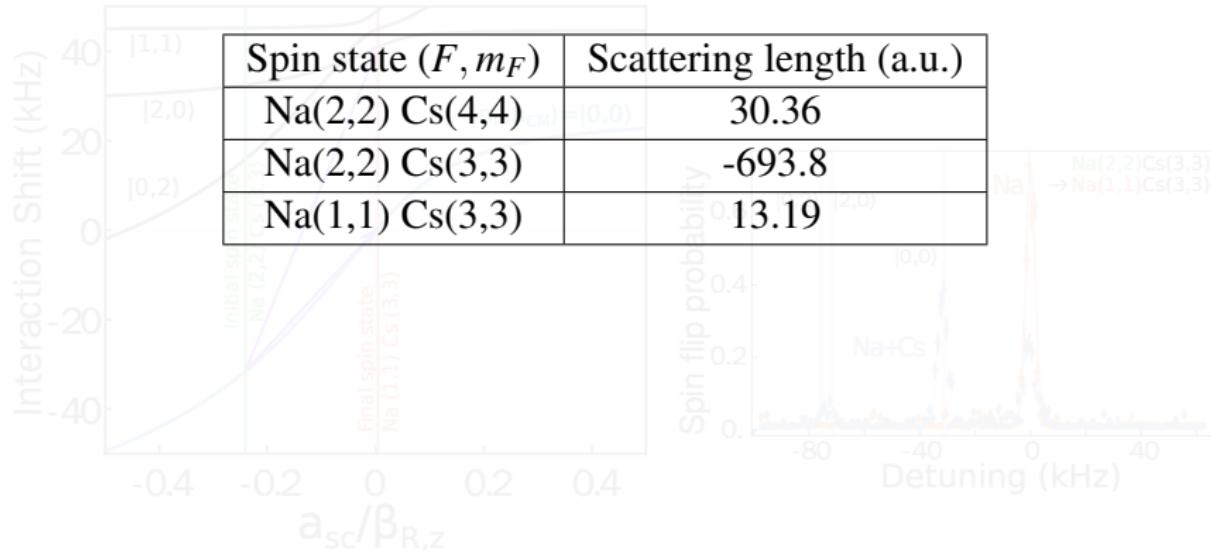
Interaction shift



$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i X_{R,i}}_{\text{Mixing}}$$

Interaction shift

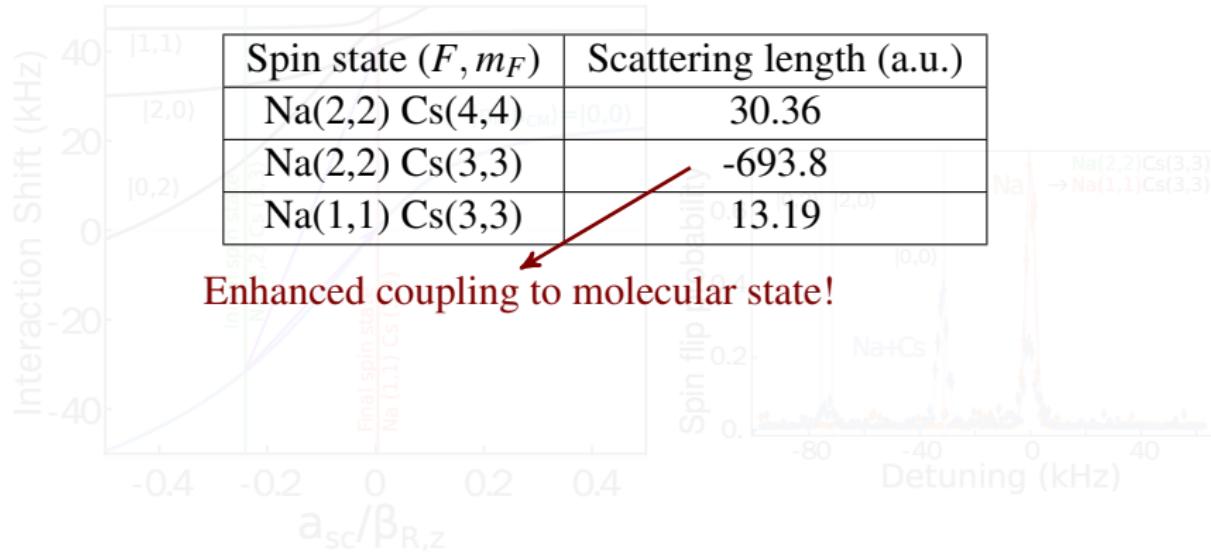
Combined with binding energy measurement on Na(2,2) Cs(4,4)



$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i X_{R,i}}_{\text{Mixing}}$$

Interaction shift

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Outline

1 Experiment overview

2 Atom state control

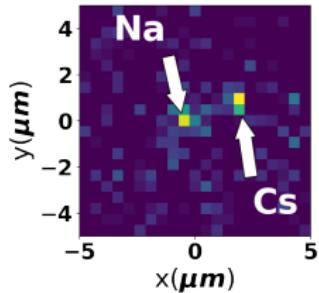
- Raman sideband cooling of Na atoms

3 Atom-atom interaction

4 Optical molecule creation

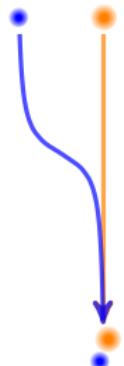
5 Conclusion

Loading

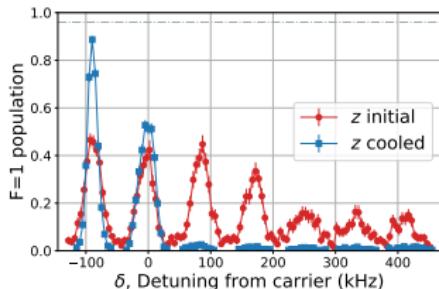


NJP. 19, 023007 (2017)

Merging



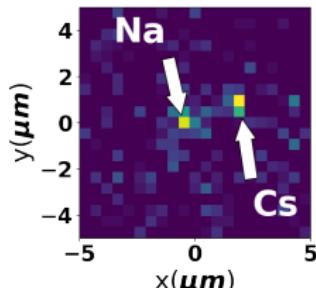
Cooling



PRA. 97, 063423 (2018)

PRX. 9, 021039 (2019)

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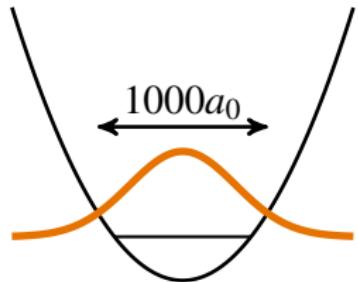


NJP. 19, 023007 (2017)

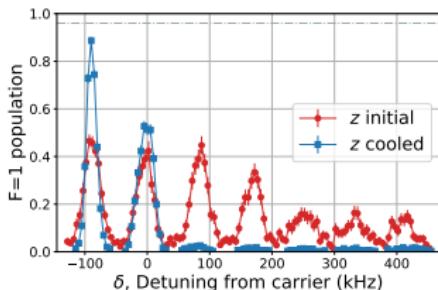
Merging



Atom



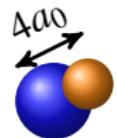
Cooling

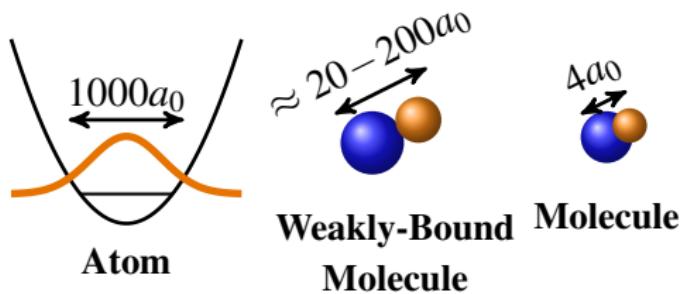


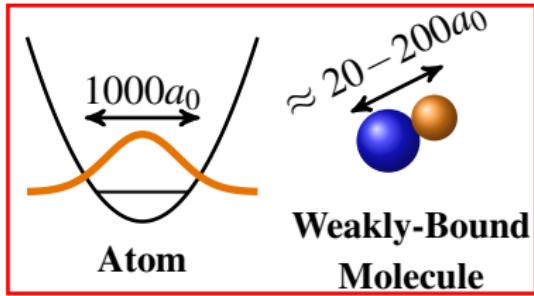
PRA. 97, 063423 (2018)

PRX. 9, 021039 (2019)

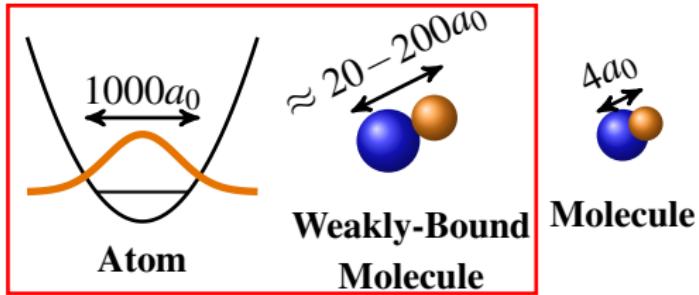
Molecule



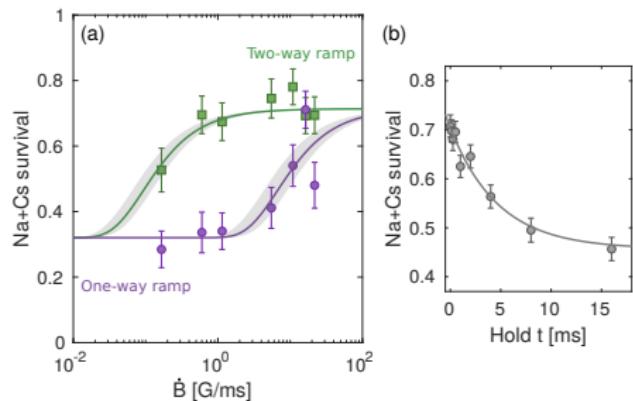




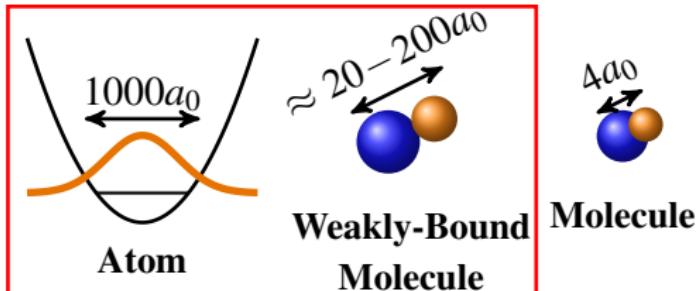
Molecule



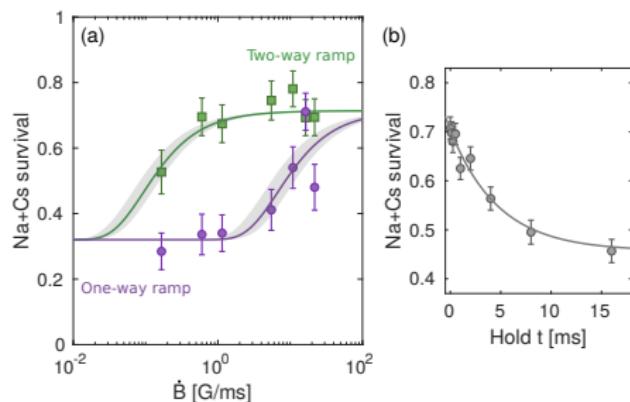
Feshbach molecule



PRL. 124, 253401 (2020)

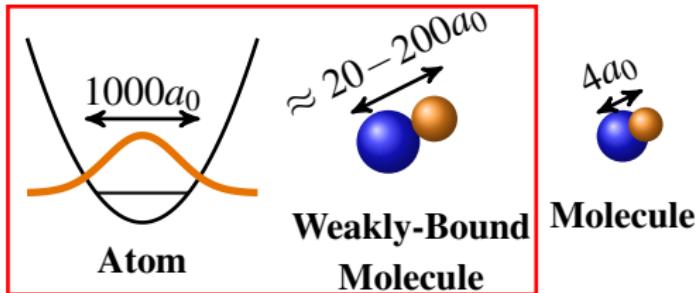


Feshbach molecule



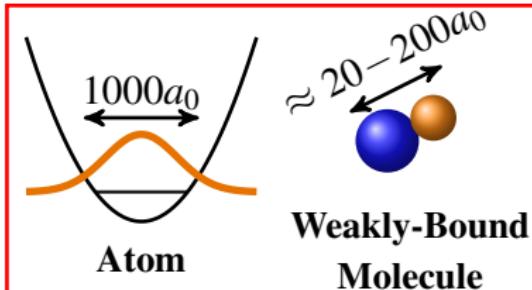
- Requires Feshbach resonance
- Usually large magnetic field

PRL. 124, 253401 (2020)



Optical transfer

- More general
- Faster

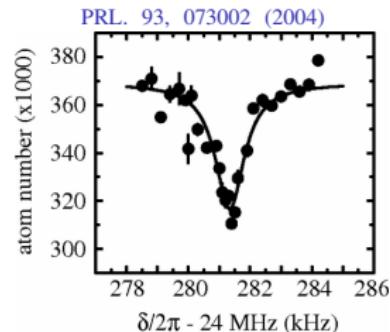


Optical transfer

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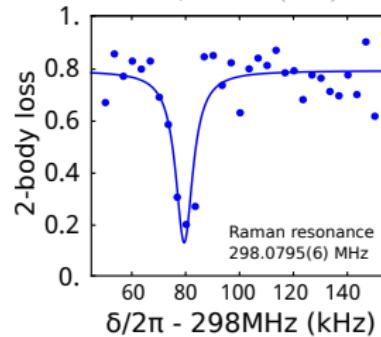
Previous results

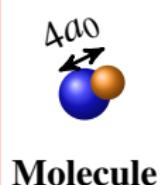
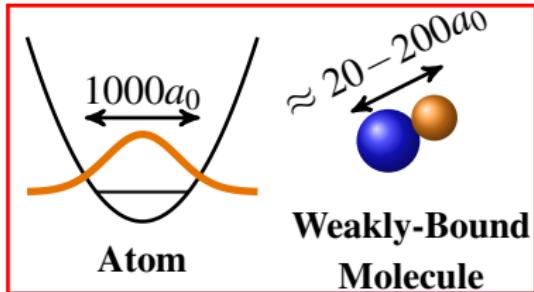
Rb_2 Science 287, 1016 (2000)



Sr_2 PRL. 109, 115302 (2012)

NaCs PRX. 9, 021039 (2019)





Optical transfer

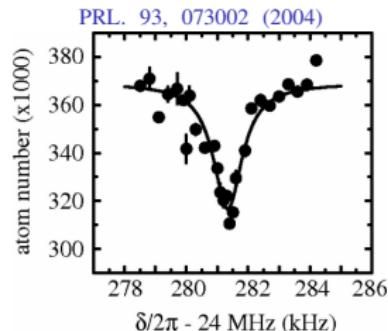
- More general
- Faster

Limitations so far

- Incoherent due to scattering
- Rely on narrow line optical transition

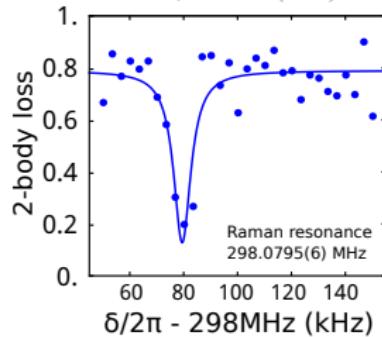
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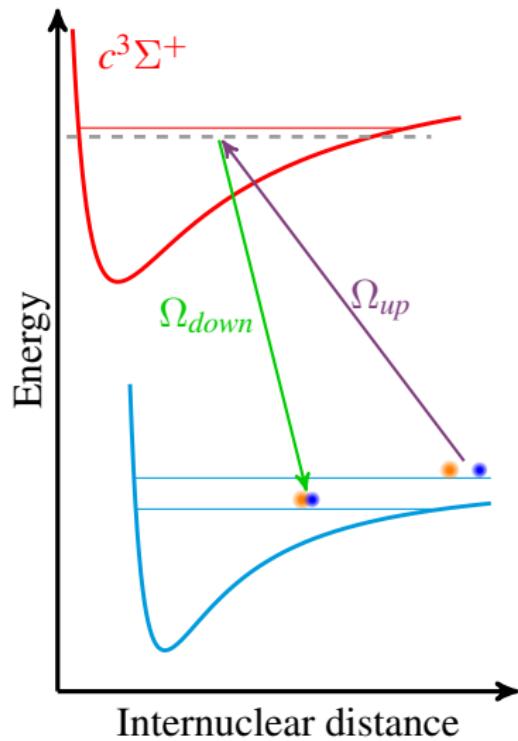


Sr_2 PRL. 109, 115302 (2012)

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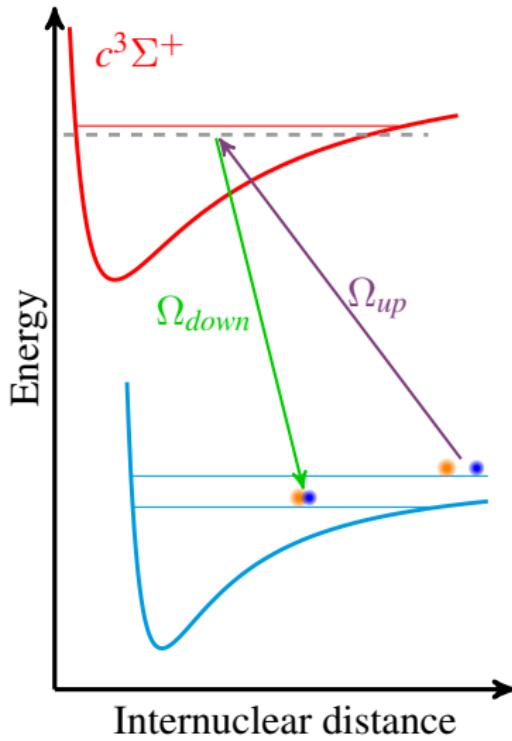
Raman transfer



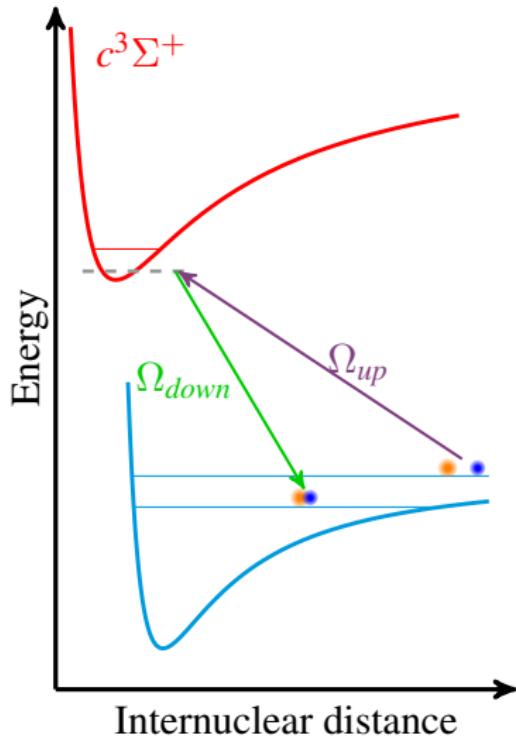
Raman transfer

Near threshold states

- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering



Raman transfer



Near threshold states

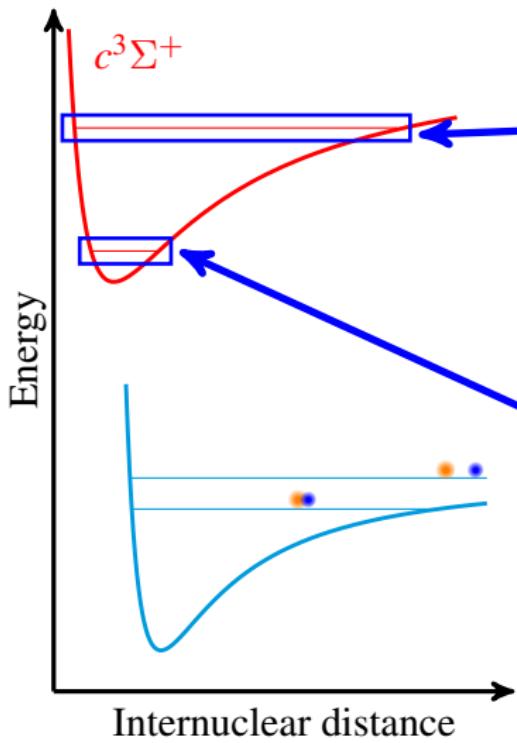
- Stronger coupling (Ω_{up} and Ω_{down})
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Deeply bound states

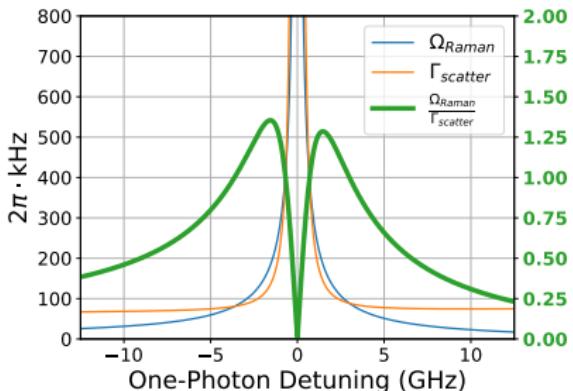
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

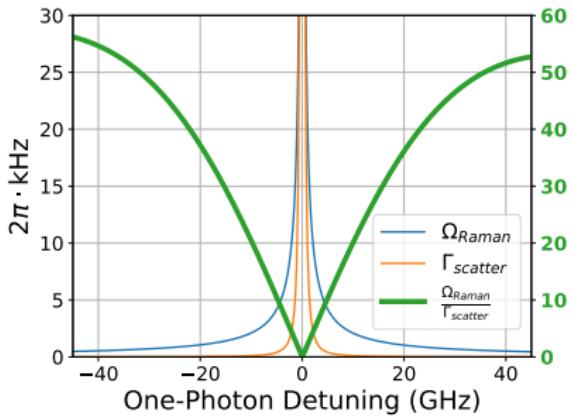
Raman transfer



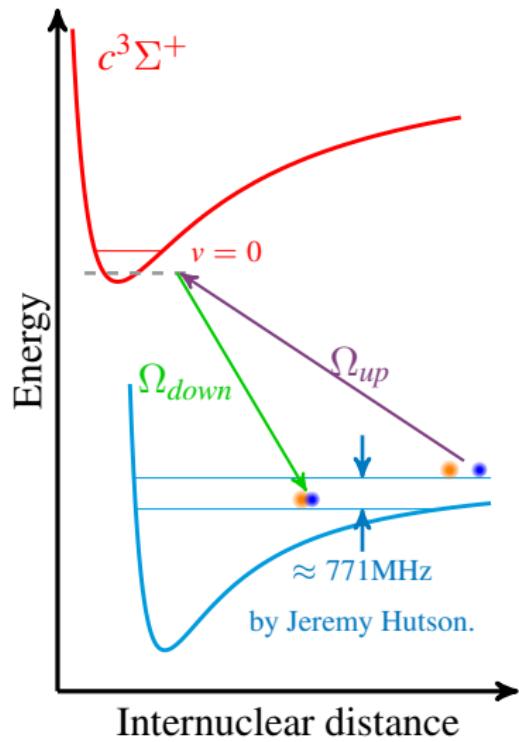
Near threshold states



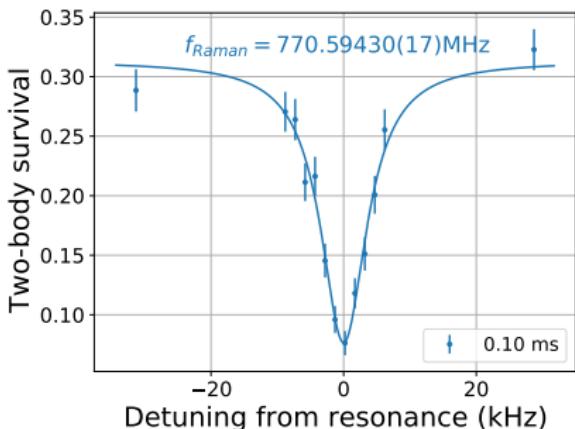
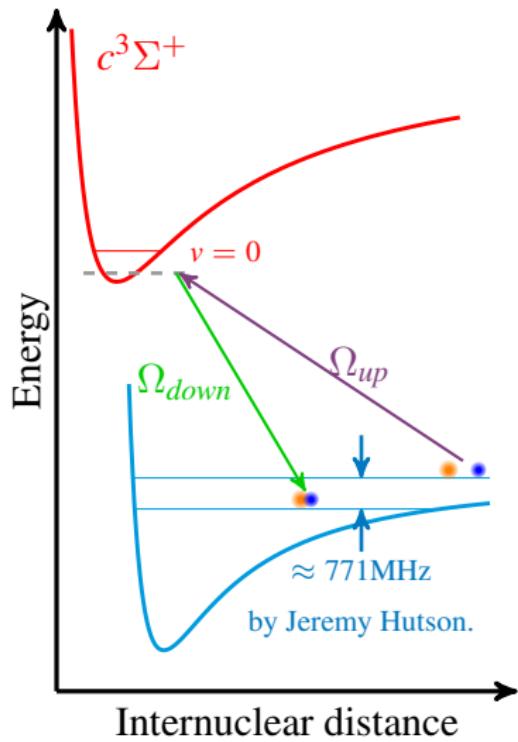
Deeply bound states



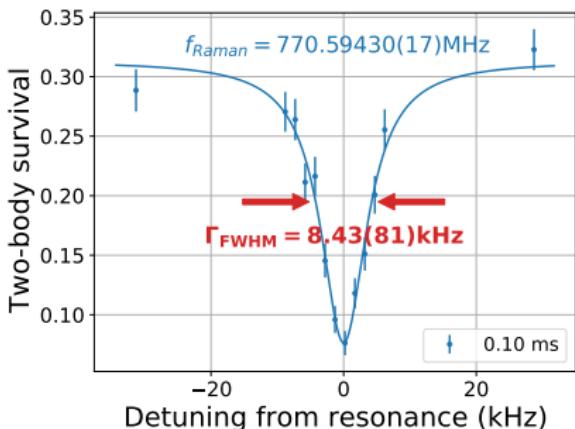
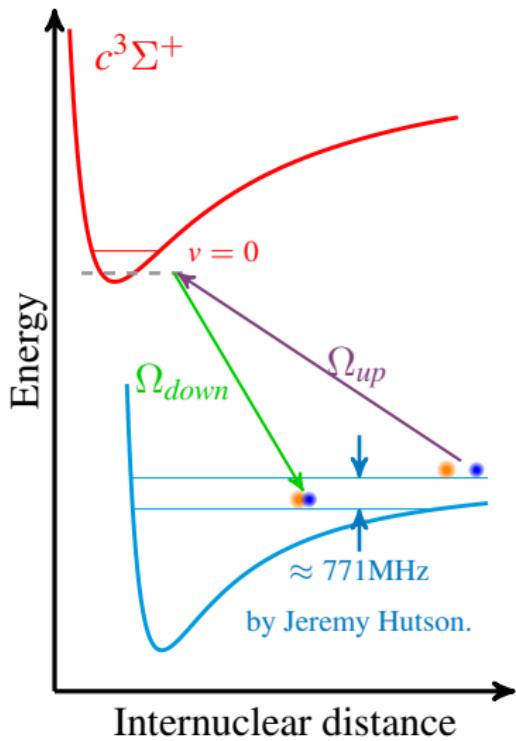
Experiment



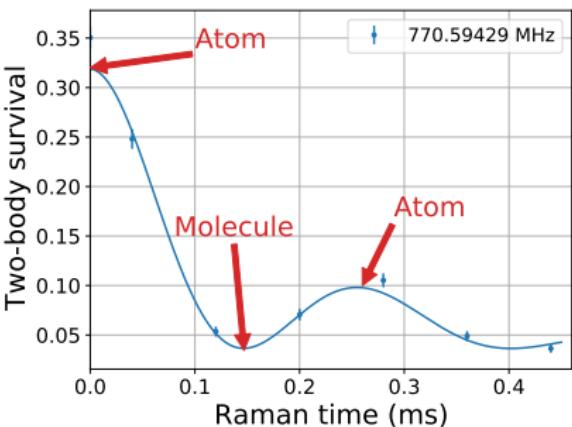
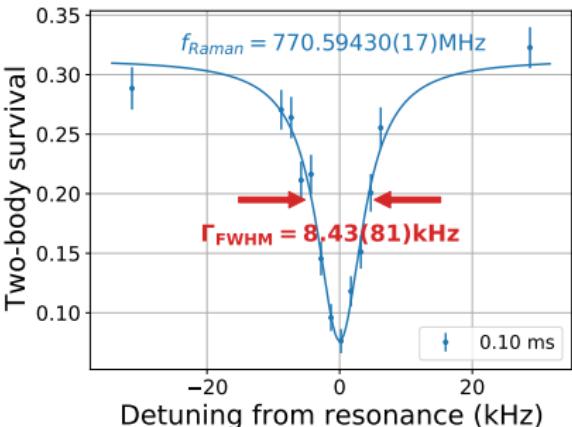
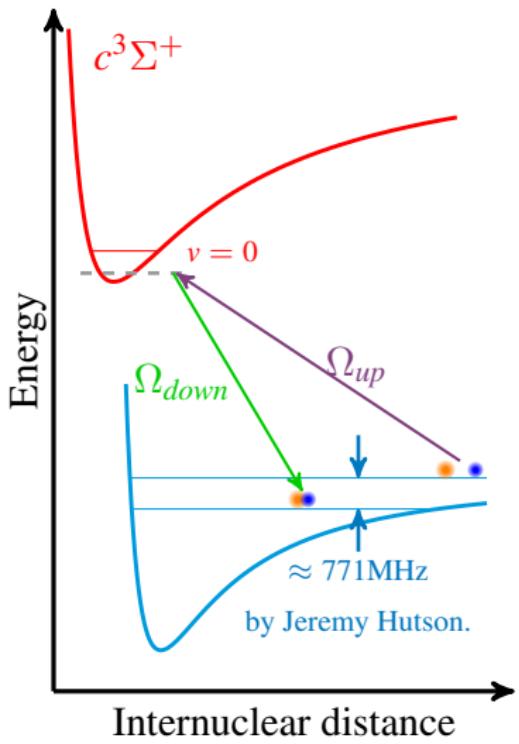
Experiment



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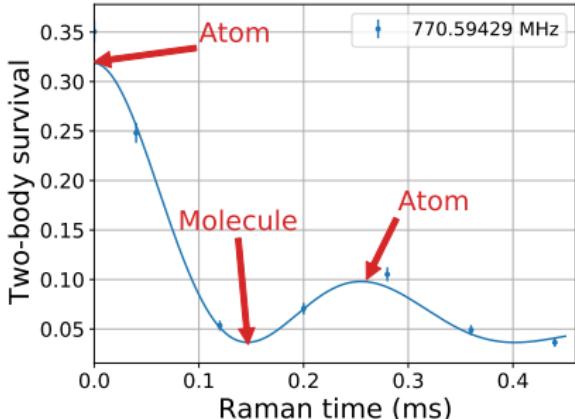
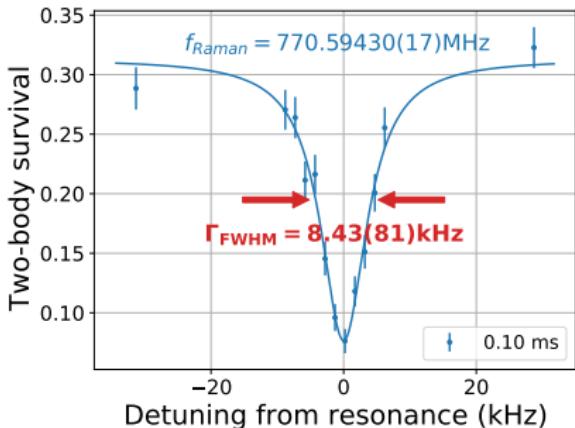
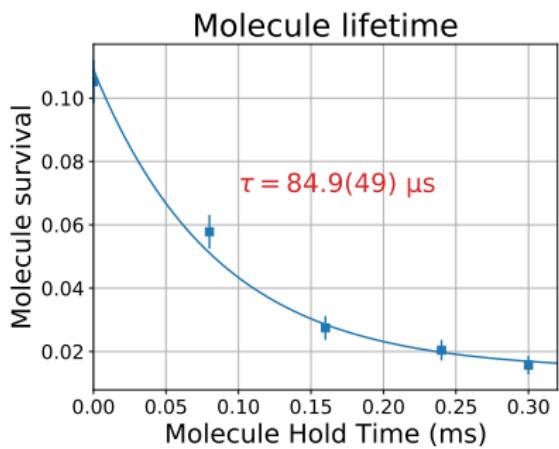


Experiment



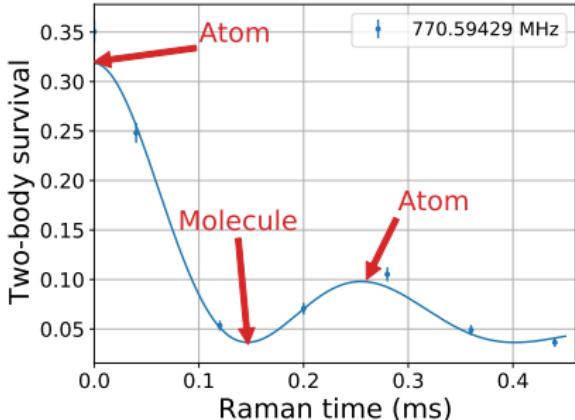
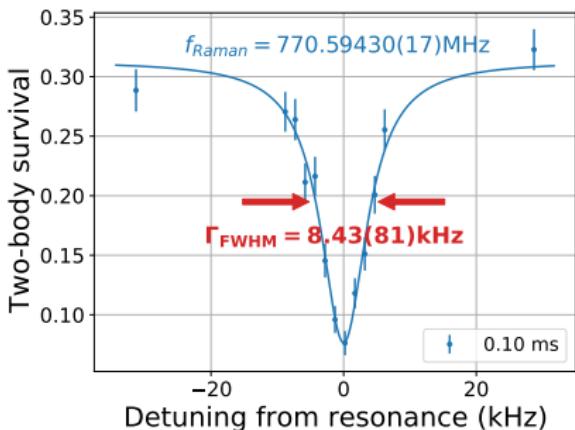
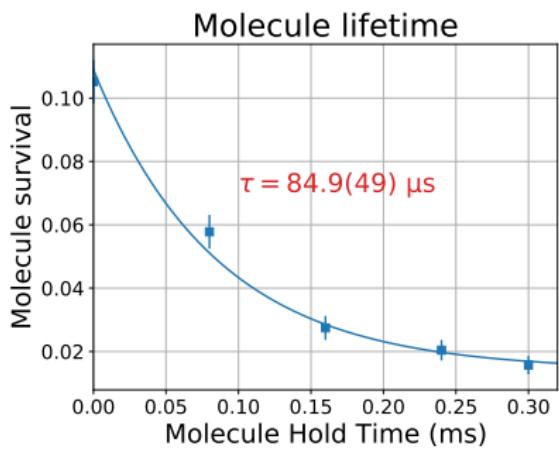
Experiment

- Transferred 50% of ground state atom to molecule.
- Single molecule spin state
- > 50% of molecule in motional ground state.
- Improving signal



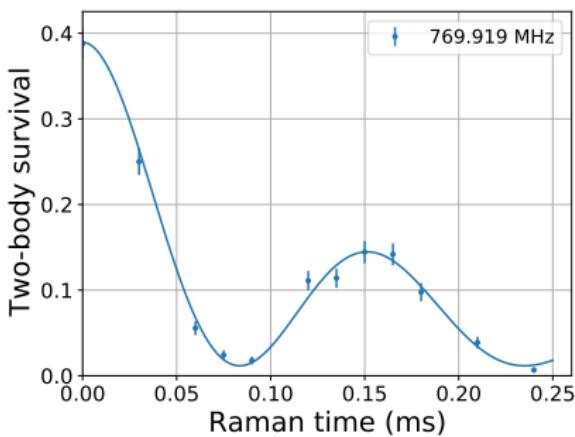
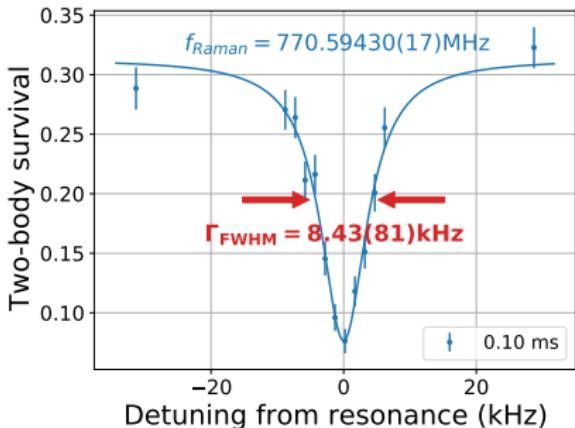
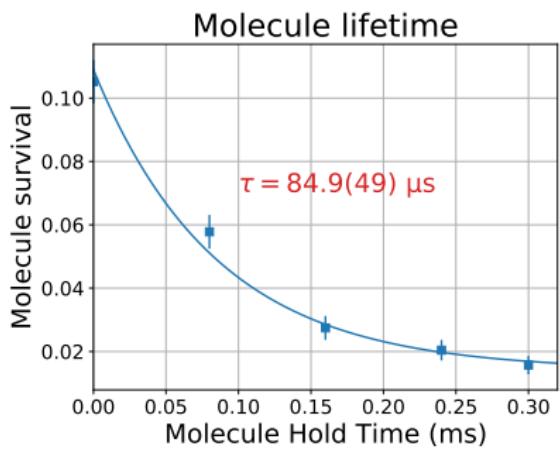
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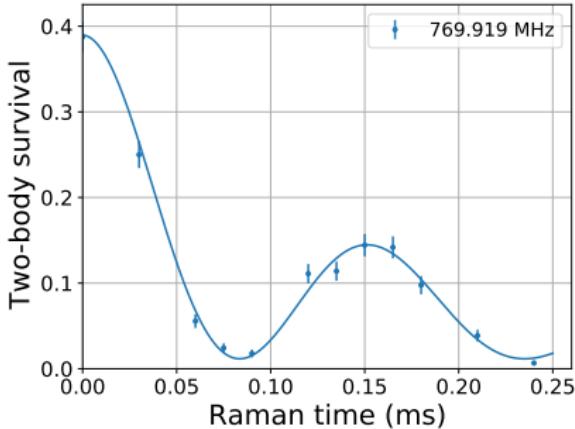
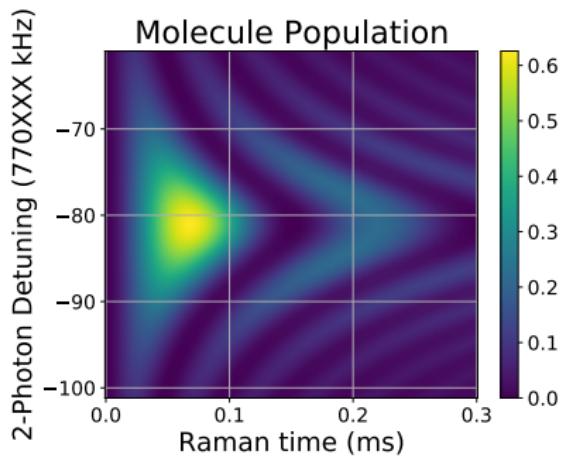
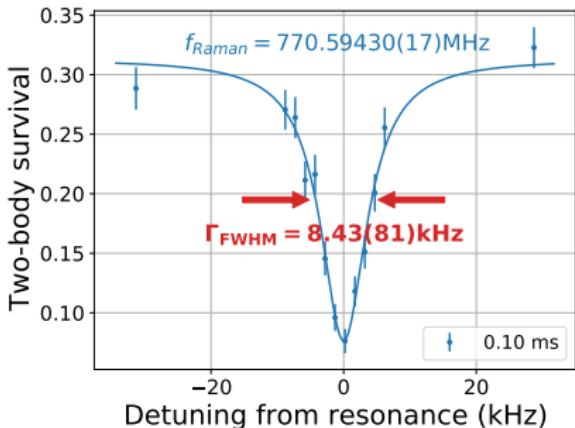
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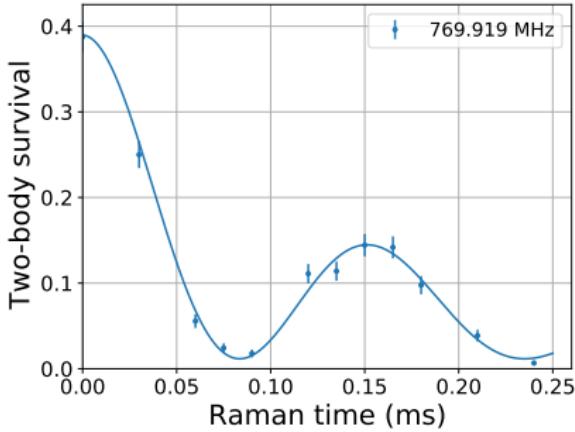
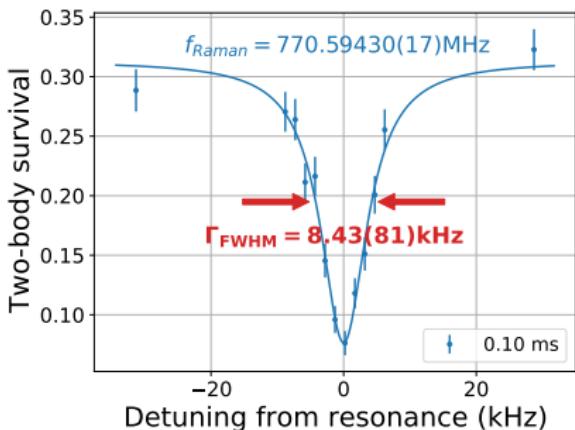
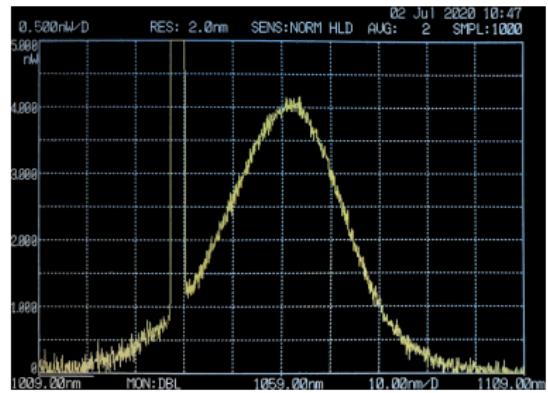
Experiment

- Transferred 63% of ground state atom to molecule.
- Single molecule spin state
- > 50% of molecule in motional ground state.
- Improving signal



Experiment

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Conclusion and outlook

- Full quantum control of atoms in optical tweezers
- Coherent all-optical creation of single molecule
- Working towards fully controlled, strongly interacting tweezer array

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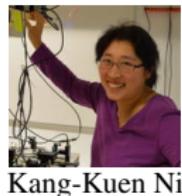
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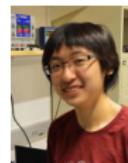
Experiment



Kang-Kuen Ni



Kenneth
Wang



Jessie
Zhang



Lewis
Picard



William
Cairncross



Lee Liu
Postdoc @JILA



Jonathan Hood
Assnt Prof @Purdue



Nick Hutzler
Assnt Prof @Caltech

Theory



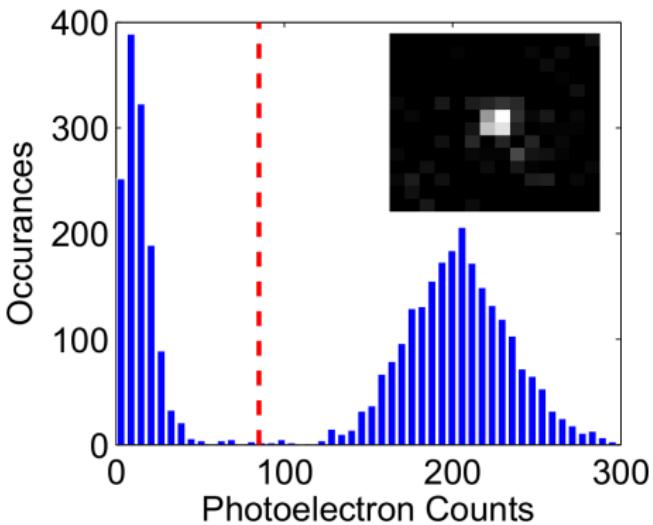
Jeremy Hutson

Single Atom in Tweezer

- Previously done with Rb
- Works for Cs
- Doesn't work for Na

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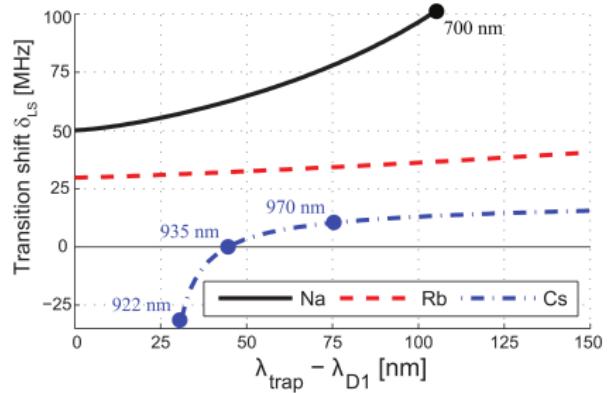
Single Atom in Tweezer

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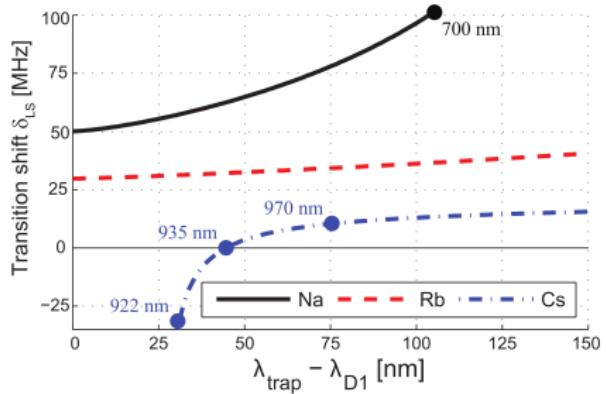
Issues with Na

- Low vapor pressure
- Broad linewidth
- Low mass
- Small hyperfine structure

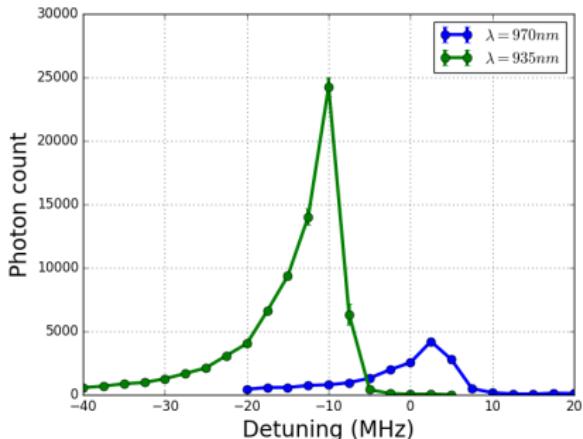
Real Issue with Na: Light Shift



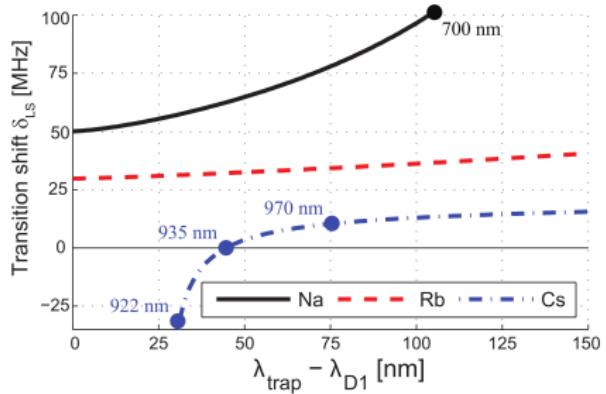
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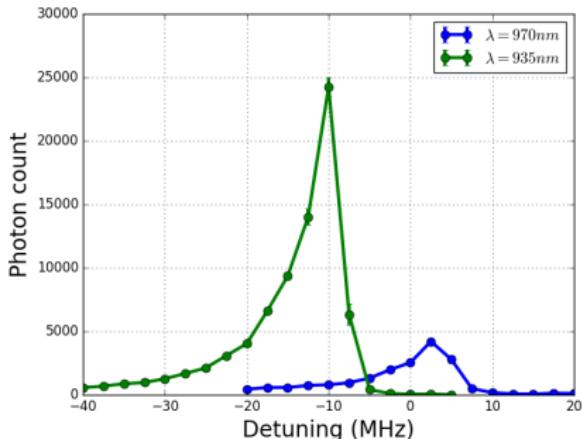
Cs single atom imaging



Real Issue with Na: Light Shift



Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

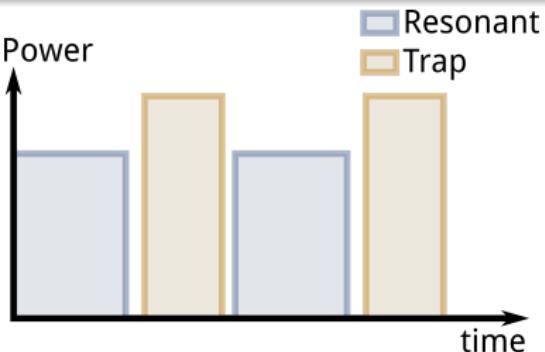
Real Issue with Na: Light Shift

Trap modulation

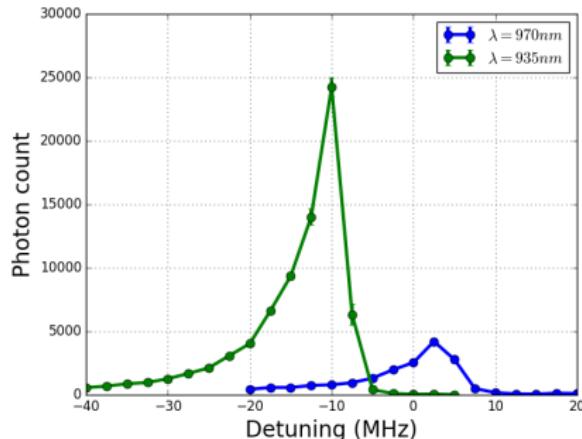
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

$$f_{trap} = 100 \sim 500 \text{ kHz}$$

$$\Gamma = 2\pi \times 10 \text{ MHz}$$



Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

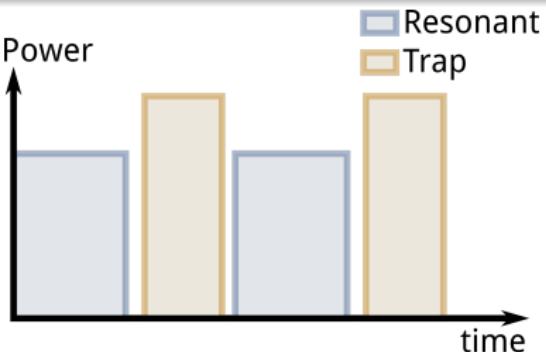
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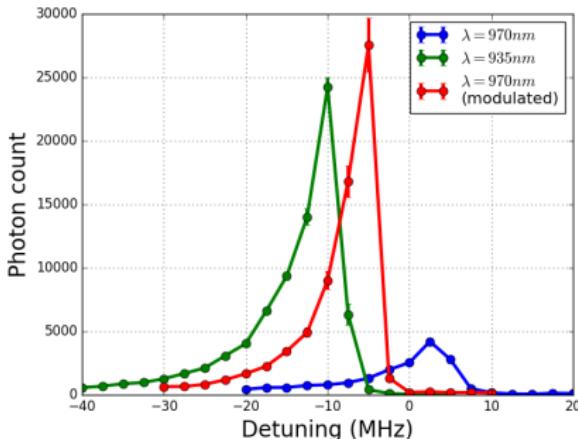
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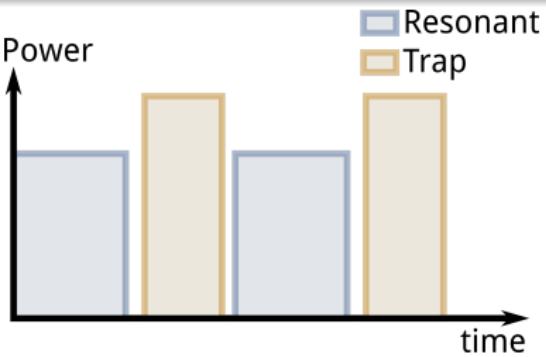
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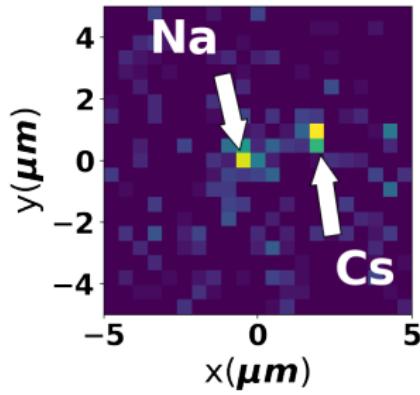
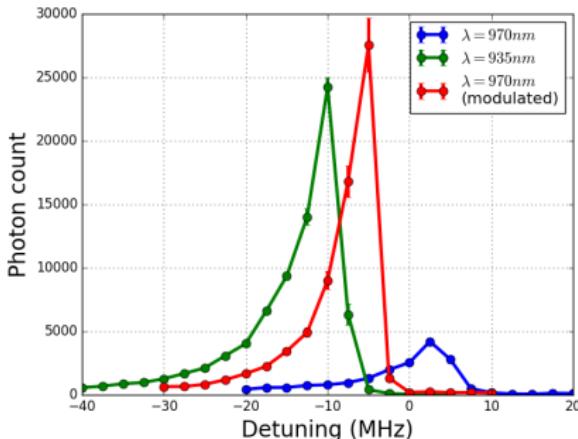
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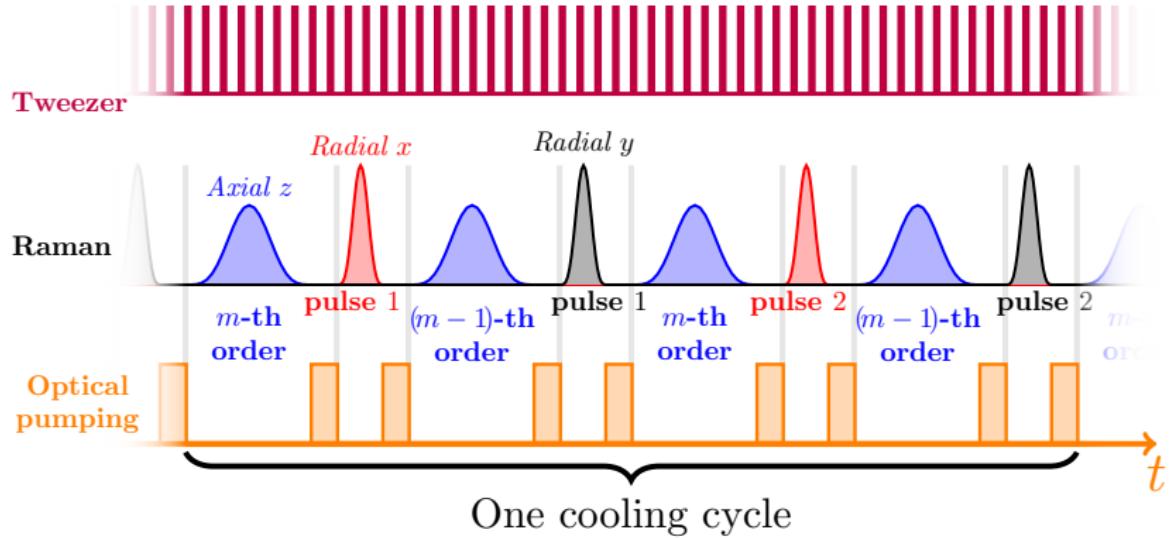
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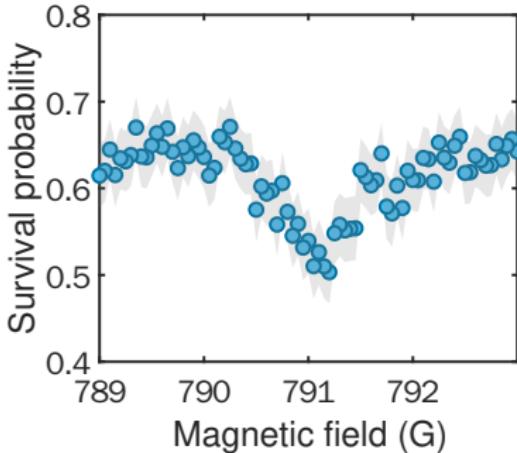
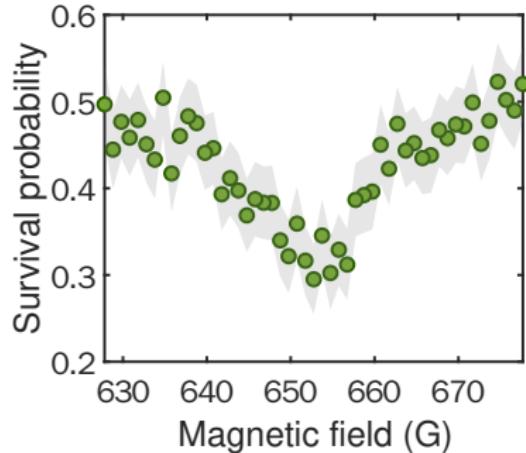
Na Raman sideband cooling sequence



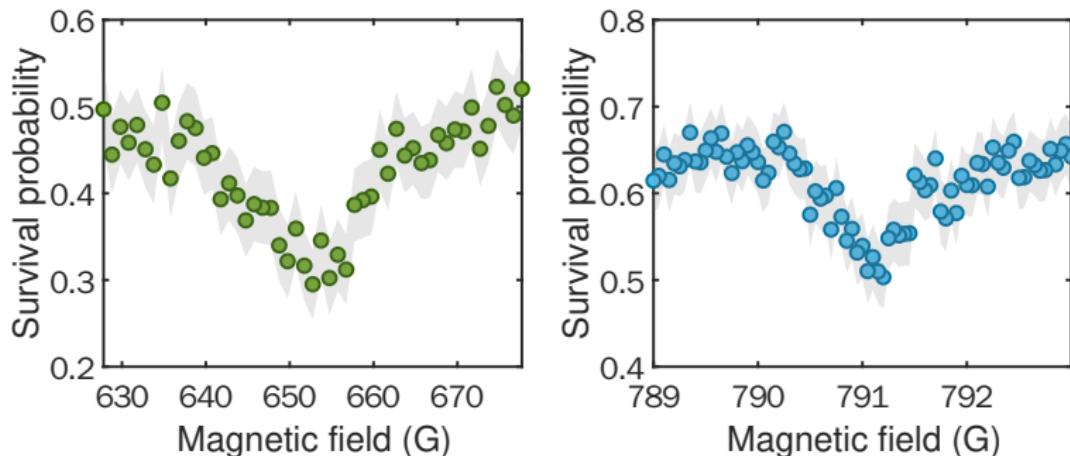
Na (1, -1) Cs (3, -3) Feshbach resonance



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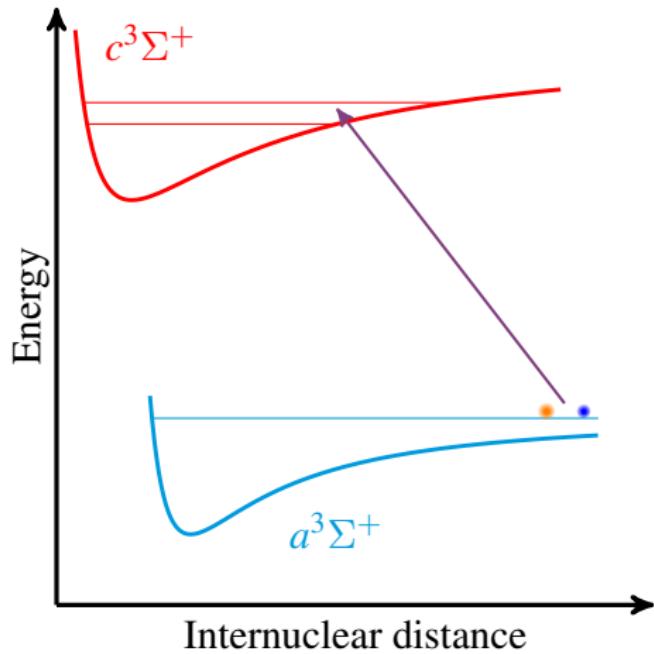
Na (1, -1) Cs (3, -3) Feshbach resonance



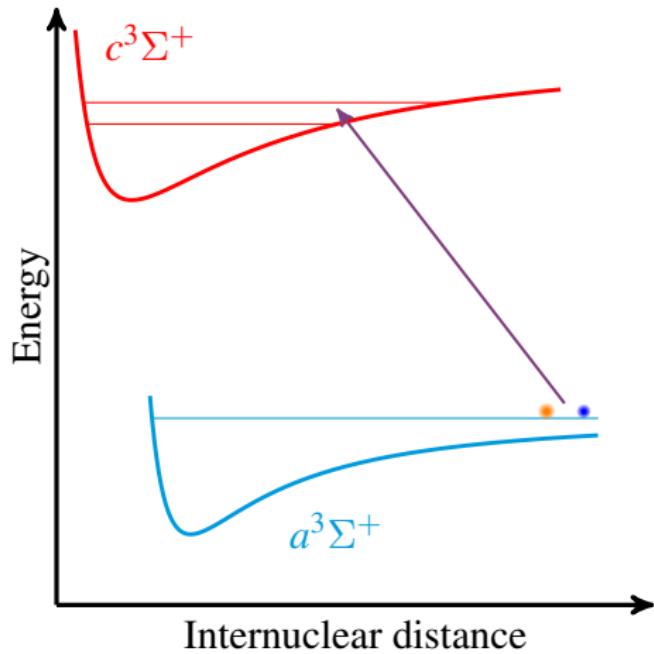
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



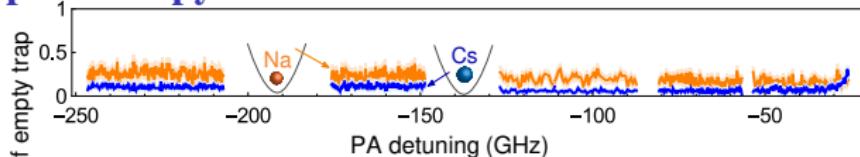
Photoassociation (PA) Spectroscopy



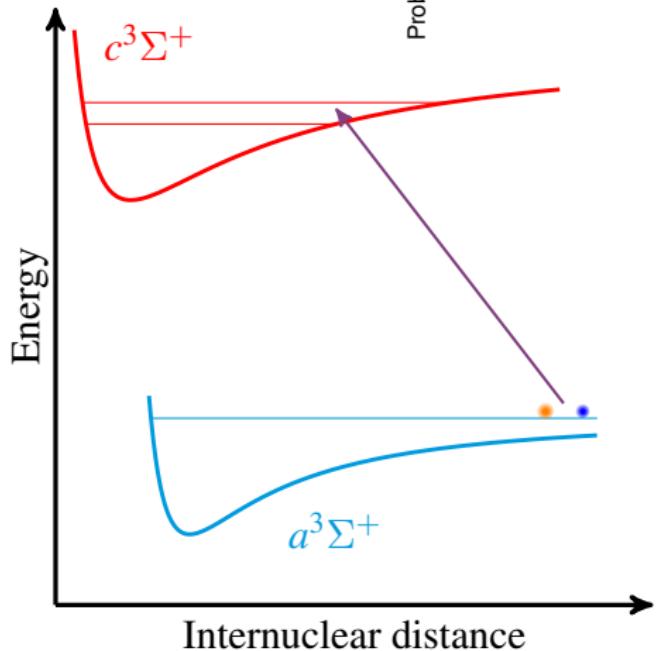
Single Atom PA

- Clean initial state
- Narrow excitation laser
- Final state detection

Photoassociation (PA) Spectroscopy



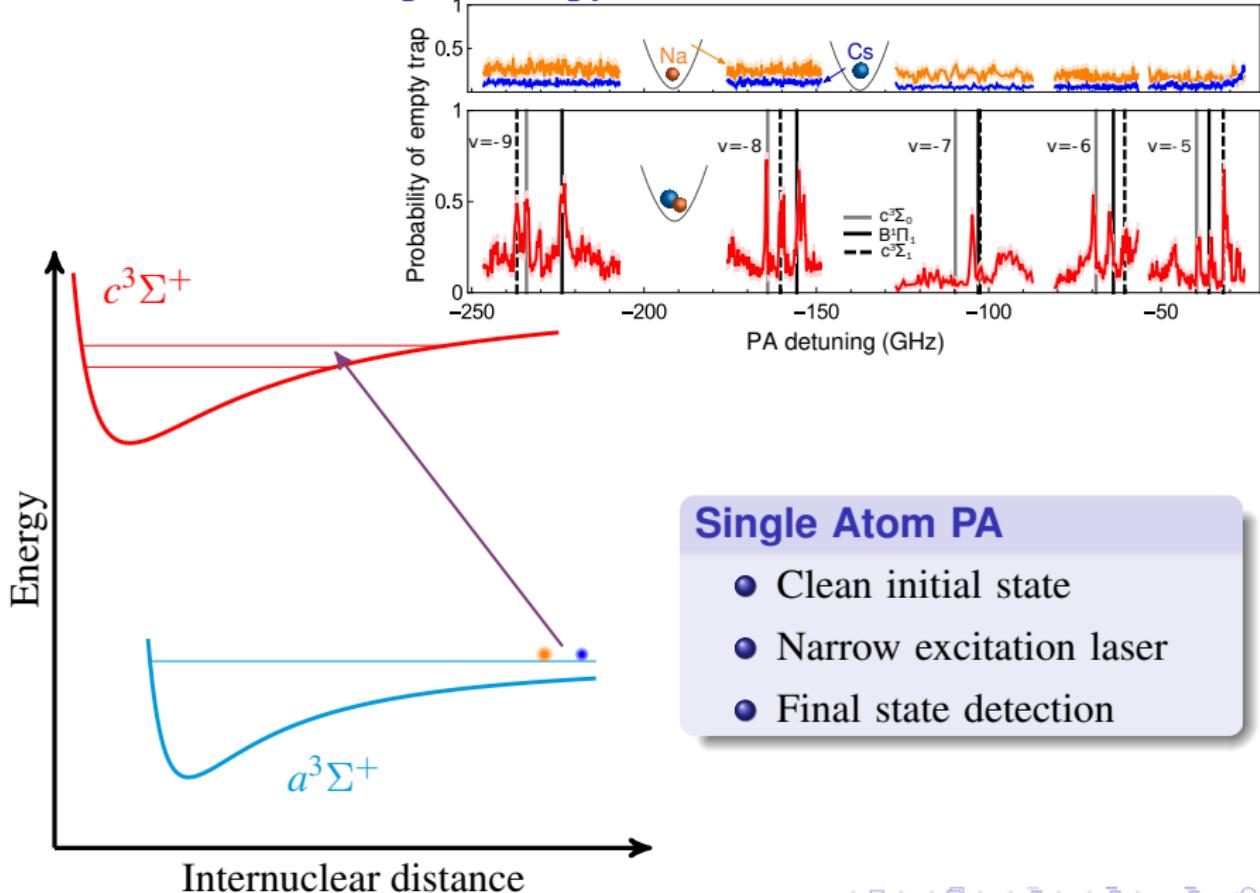
Probability of empty trap



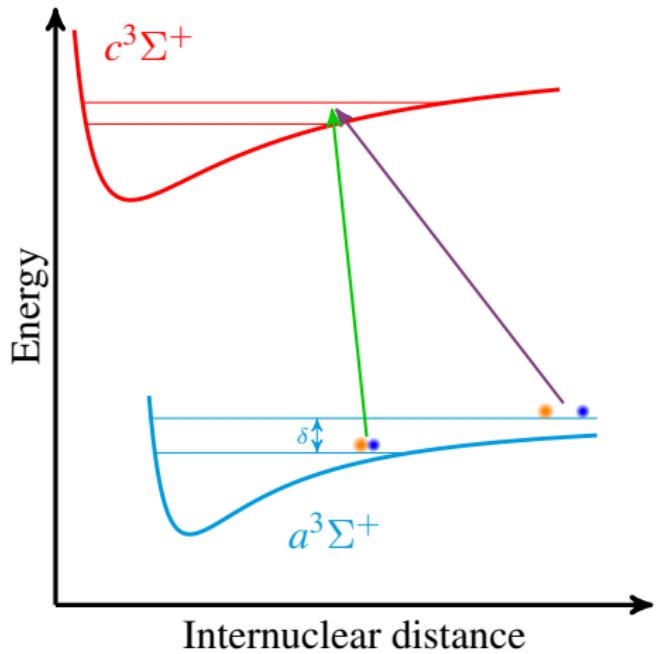
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Photoassociation (PA) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy

