

Coherent Creation of Single Molecules from Single Atoms

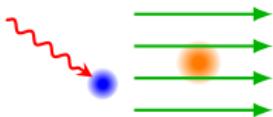
Yichao Yu

Ni Group/Harvard

Simple System



Full Control



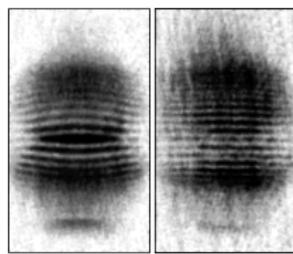
Complex Dynamic

Simple System

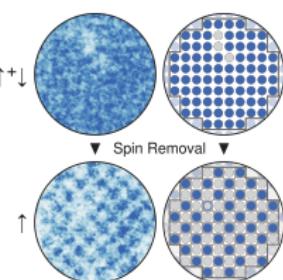
Full Control



Complex Dynamic



Science 275, 637 (1997)



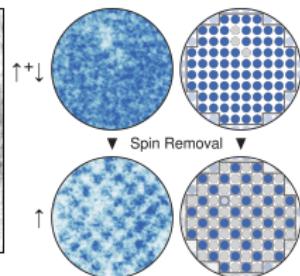
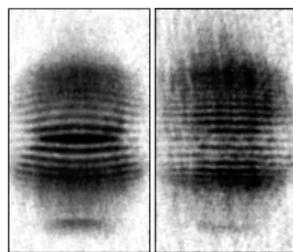
Nature 545, 462-466 (2017)

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Complex Dynamic



Science 275, 637 (1997)

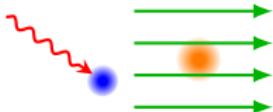
Nature 545, 462-466 (2017)

- ✗ Simple internal structure
- ✗ Weak interaction

Simple System



Full Control

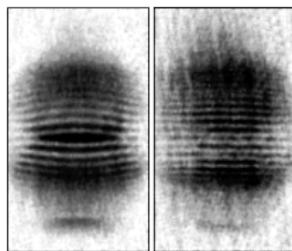


Strong interaction

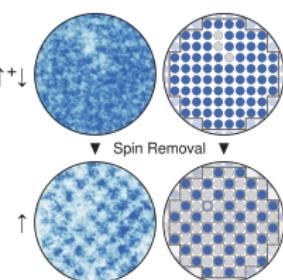
Rich internal structure

Long coherence time

Fully controllable



Science 275, 637 (1997)



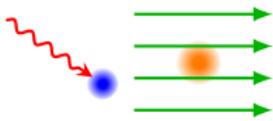
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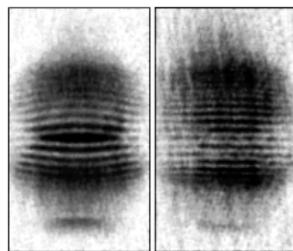
Simple System



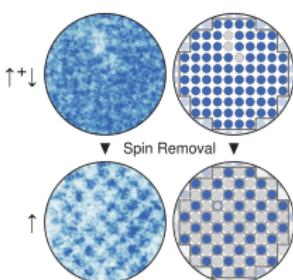
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Complex Dynamic



Science 275, 637 (1997)



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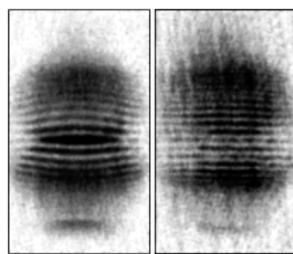
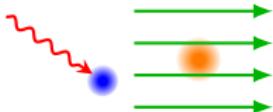
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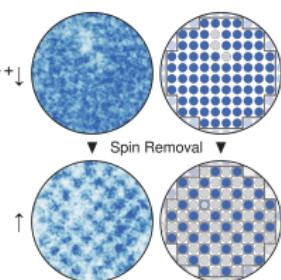
Simple System



Full Control



Science 275, 637 (1997)



Nature 545, 462-466 (2017)

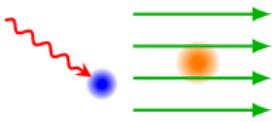
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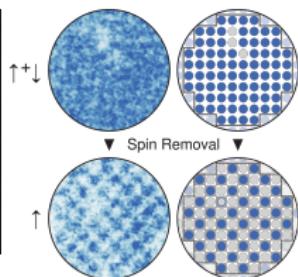
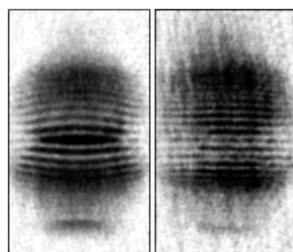
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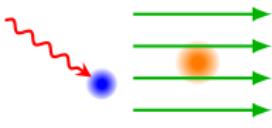
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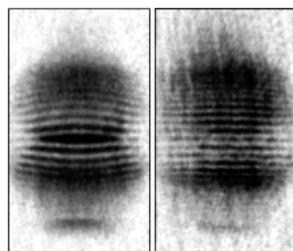
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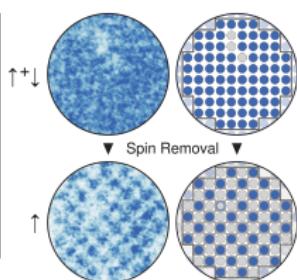
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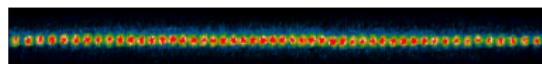


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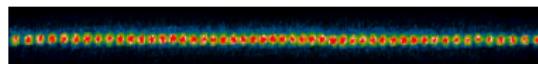
Ions (Photo Credit Monroe Group)



Rydberg Atoms (Photo Credit Lukin Group)

- Simple internal structure
- Weak interaction

- Strong interaction
- Rich internal structure
- Long coherence time
- Fully controllable



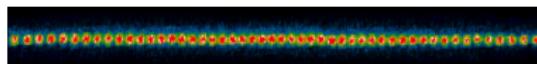
Ions (Photo Credit Monroe Group)



Rydberg Atoms (Photo Credit Lukin Group)

✓ Strong interaction (kHz)

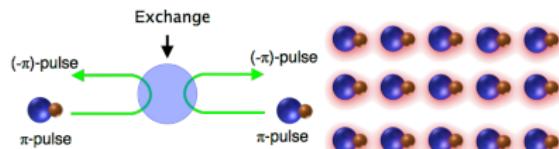
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Ions (Photo Credit Monroe Group)



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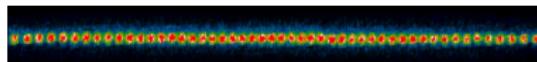
Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

✓ Strong interaction (kHz)

□ Rich internal structure

✓ Long coherence time

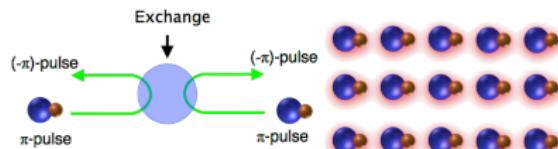
□ Fully controllable



Ions (Photo Credit Monroe Group)

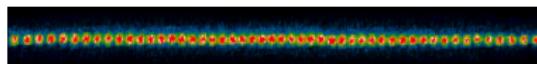


Rydberg Atoms (Photo Credit Lukin Group)



Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

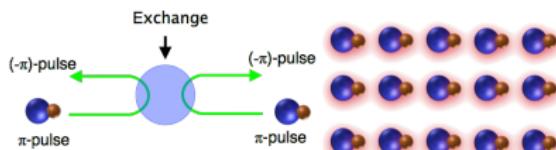
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Ions (Photo Credit Monroe Group)



Rydberg Atoms (Photo Credit Lukin Group)

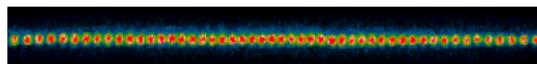


Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

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Optical tweezers

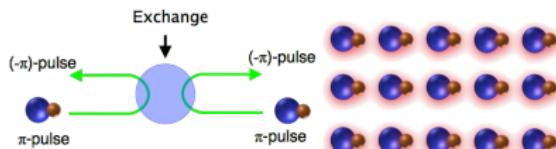
- Single site resolution
- . . .



Ions (Photo Credit Monroe Group)

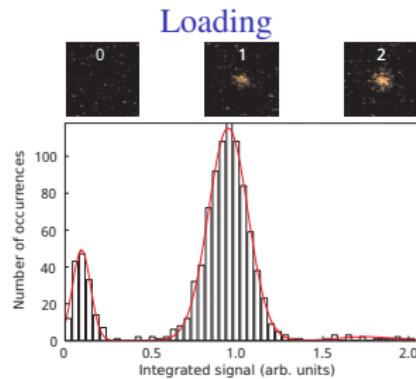


Rydberg Atoms (Photo Credit Lukin Group)



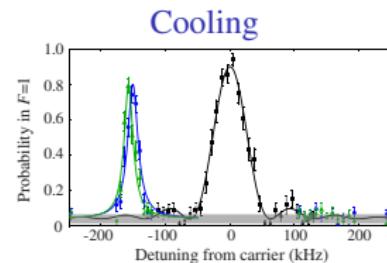
Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

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Nat. Phys. 6, 951 (2010)

- ## Optical tweezers
- Single site resolution
 - . . .



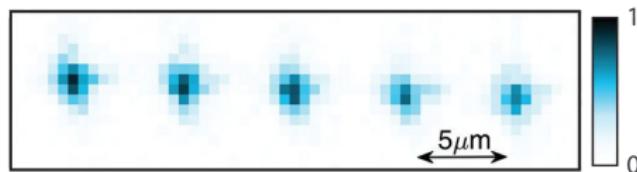
PRX. 2, 041014 (2012)



Science 354, 1024 (2016)

Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

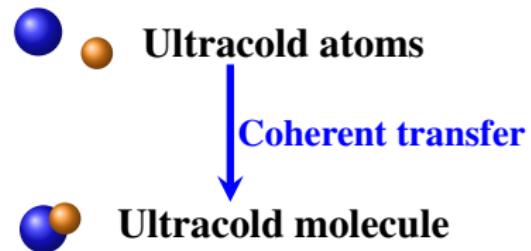
Ultracold molecules in tweezers

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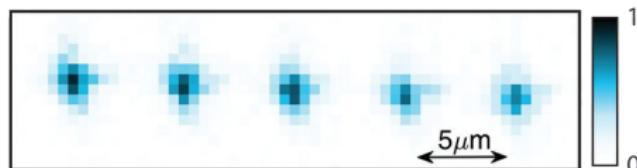
Science 365, 1156 (2019)

Assembly



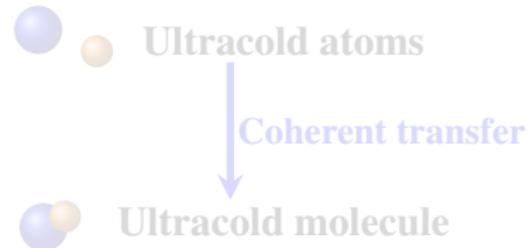
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezer
- Quantum control

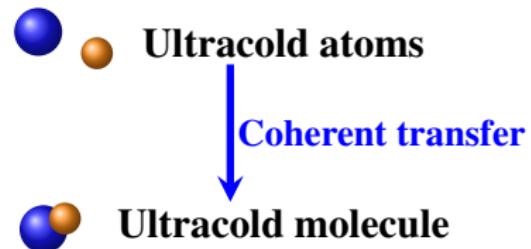
Ultracold molecules in tweezers

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 Molecule creation

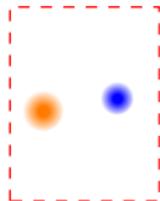
- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

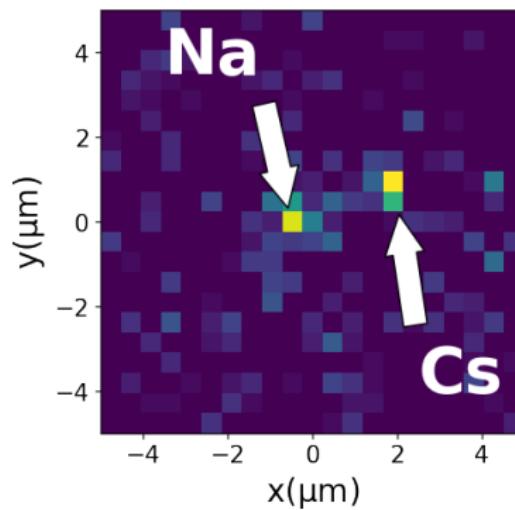
NaCs molecule

- Bi-alkali (easy to control)
- Large dipole moment: 4.6 D

Experiment overview



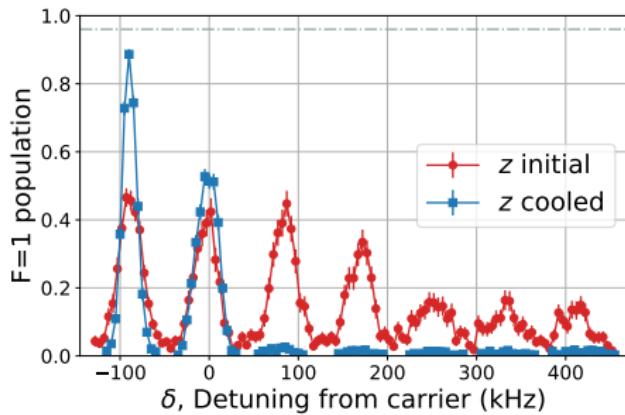
Loading



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

Experiment overview

Cooling



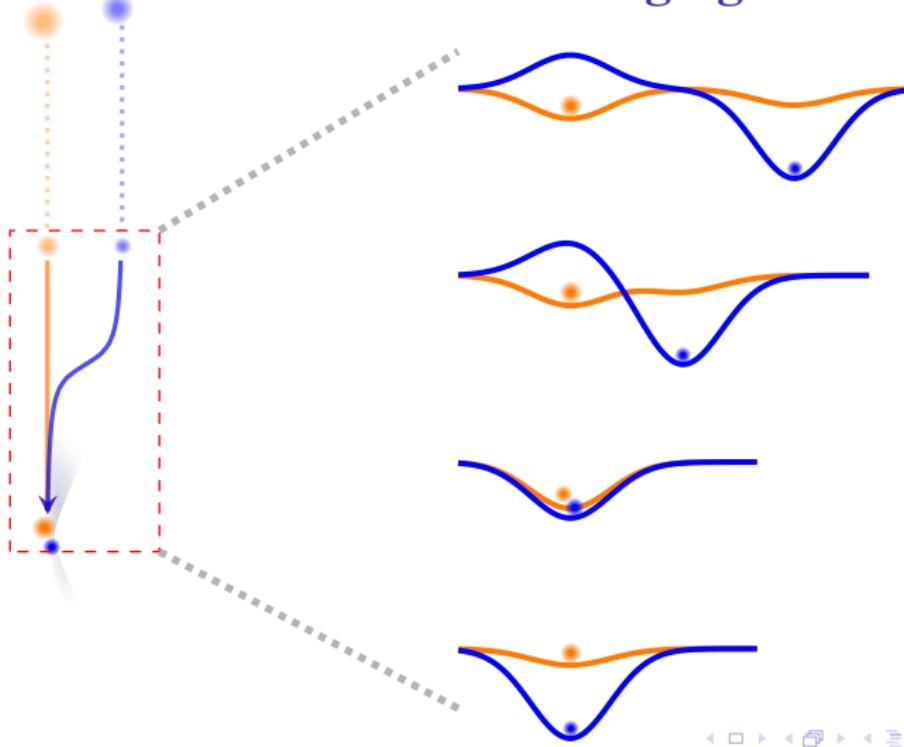
Cs: 96% ground state¹
Na: 94% ground state²

¹Y. Yu et al. PRX 9, 021039 (2019)

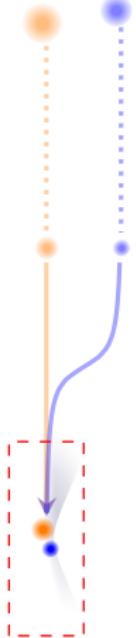
²Y. Yu et al. PRA 97, 063423 (2018)

Experiment overview

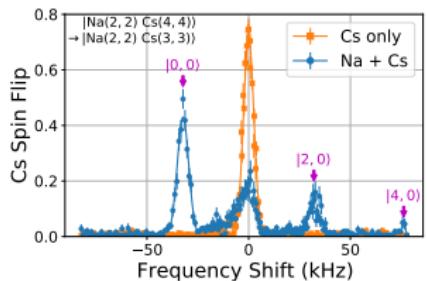
Merging



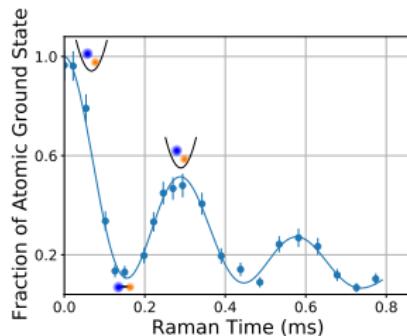
Experiment overview



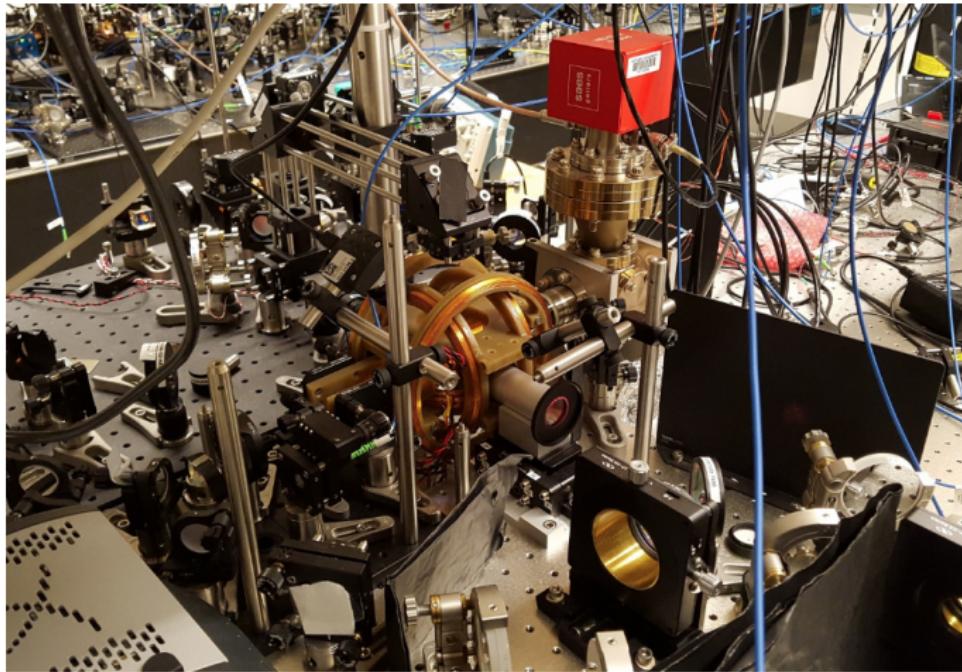
Interaction

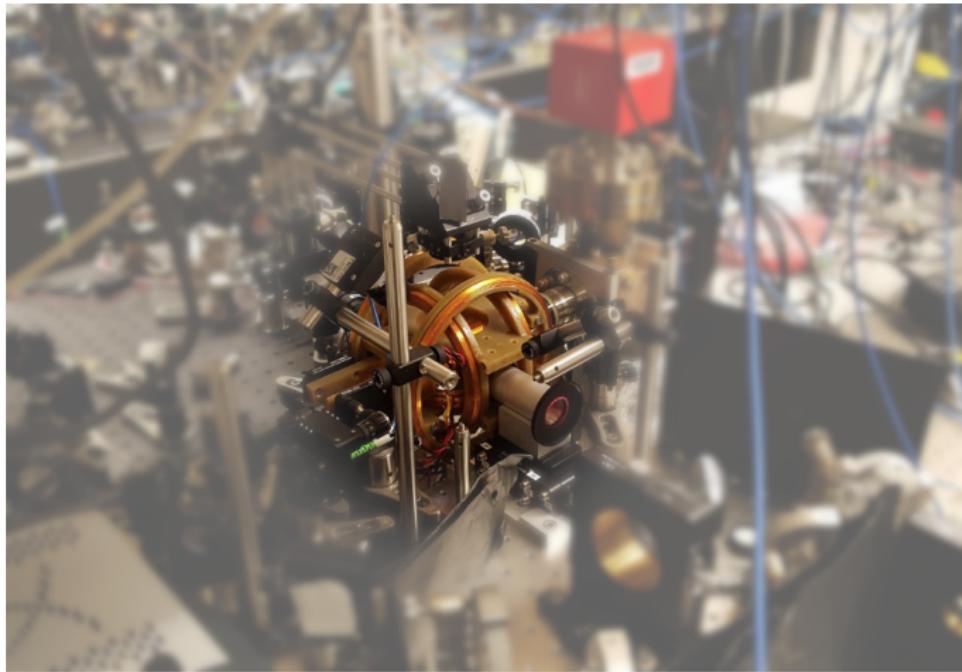


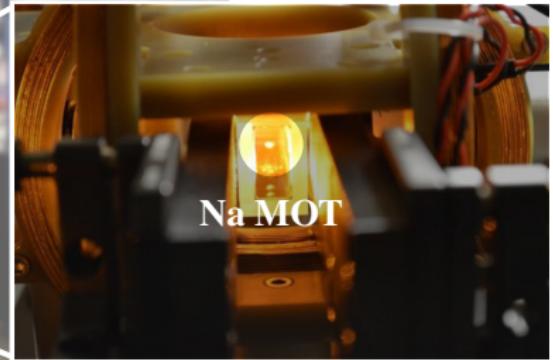
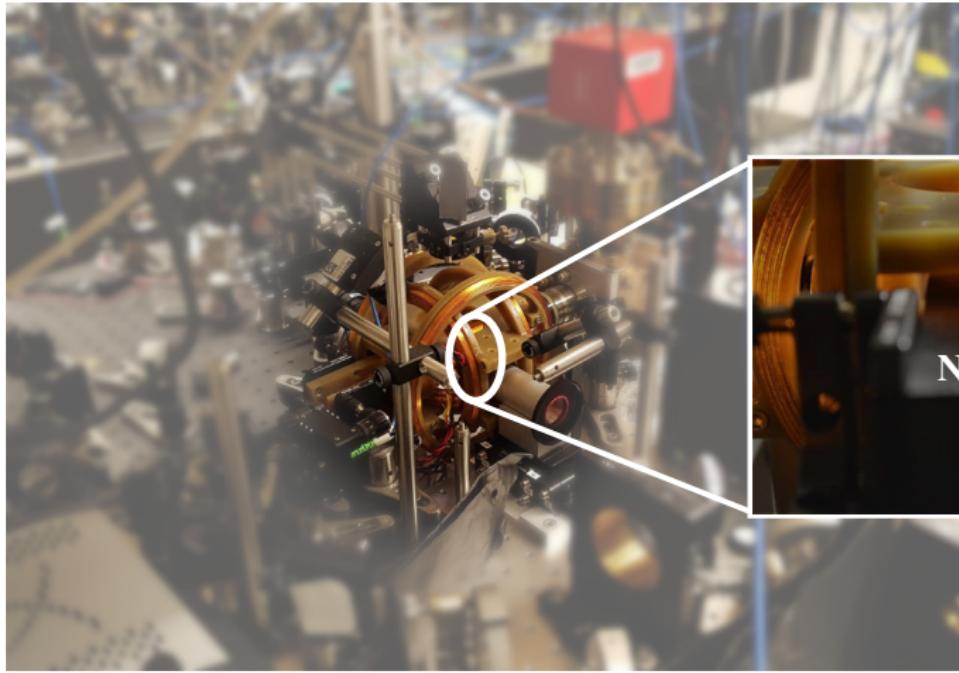
Molecule Creation



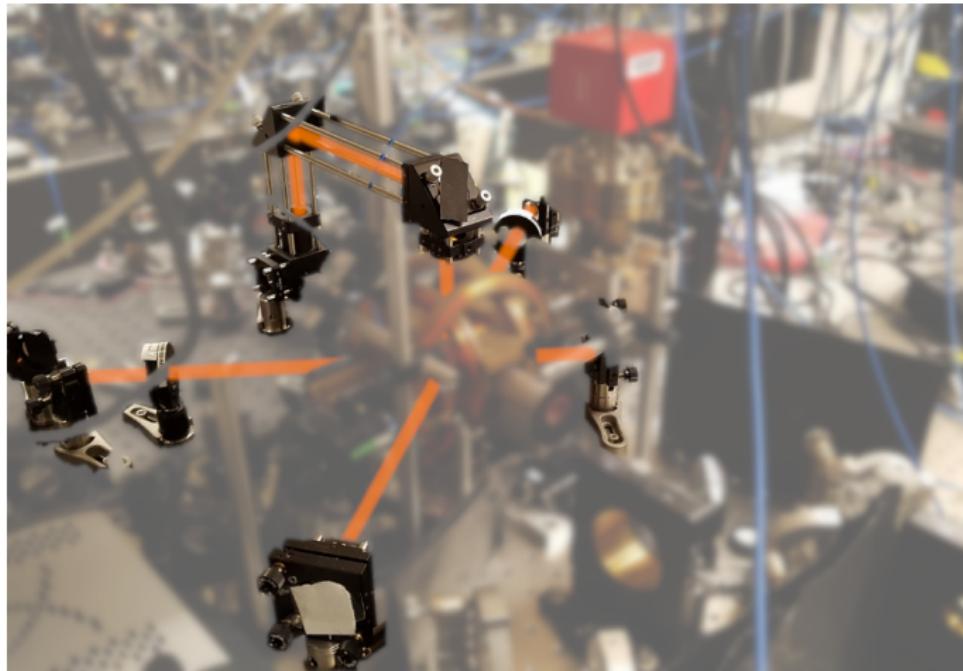
L. R. Liu, J. D. Hood, Y. Yu et al.,
Science 360, 6391 (2018)



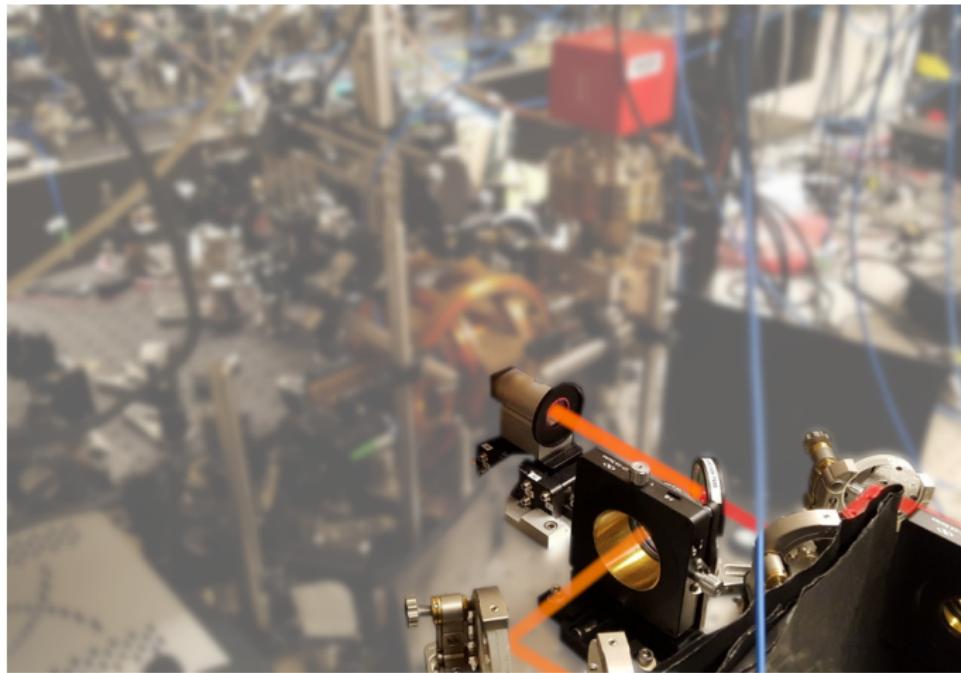




MOT beam path



Tweezer and imaging beam path



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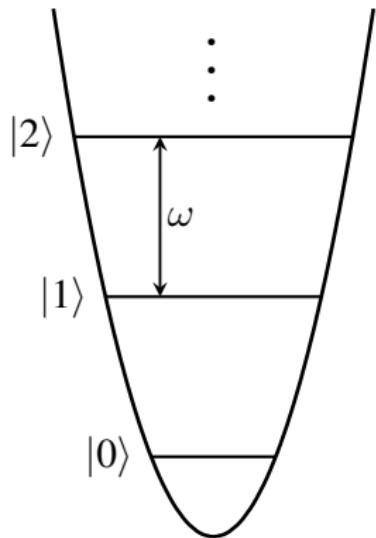
4 Conclusion

Raman sideband cooling

Used for cooling in trap

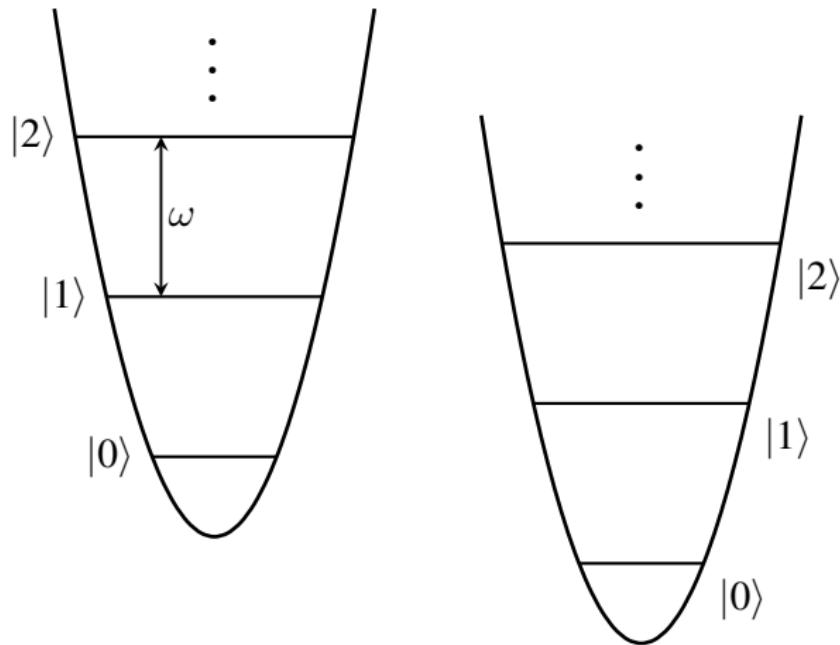
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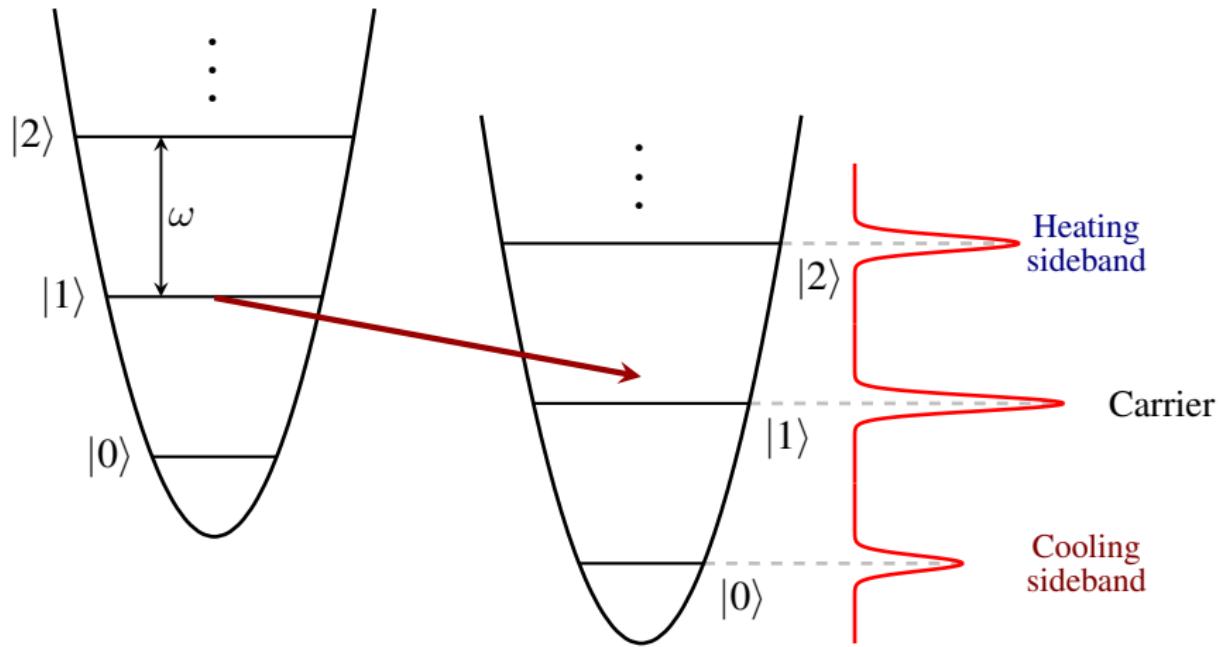
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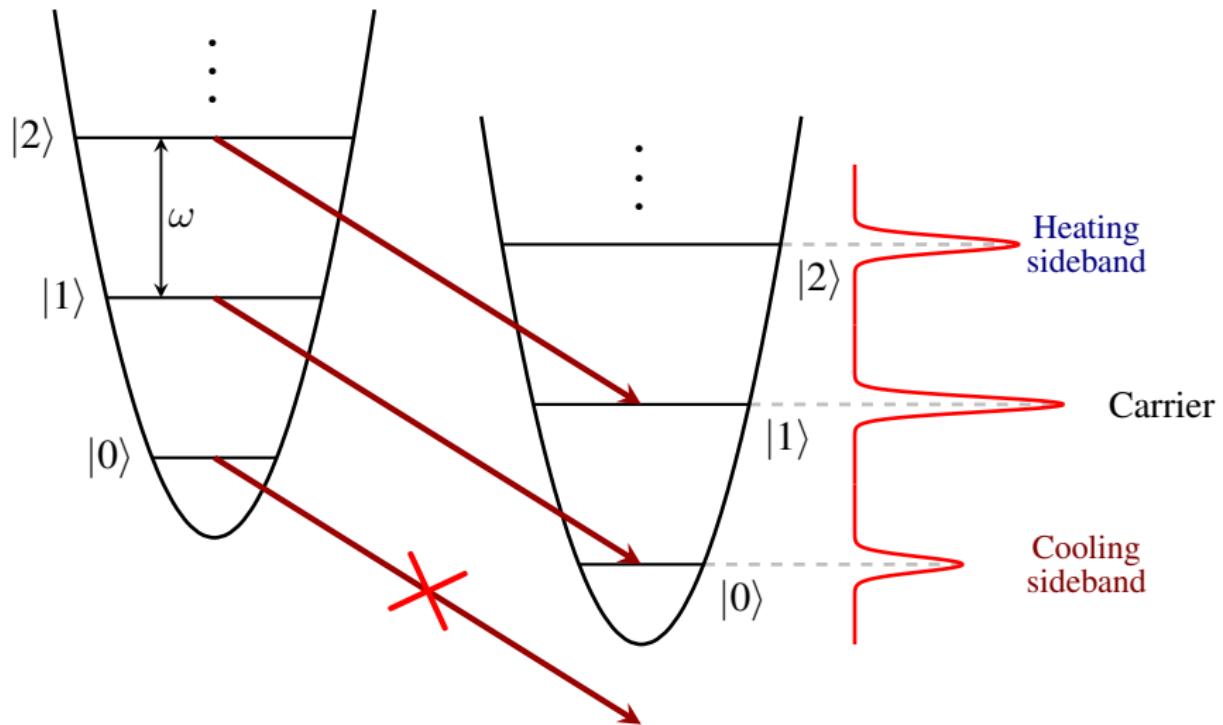
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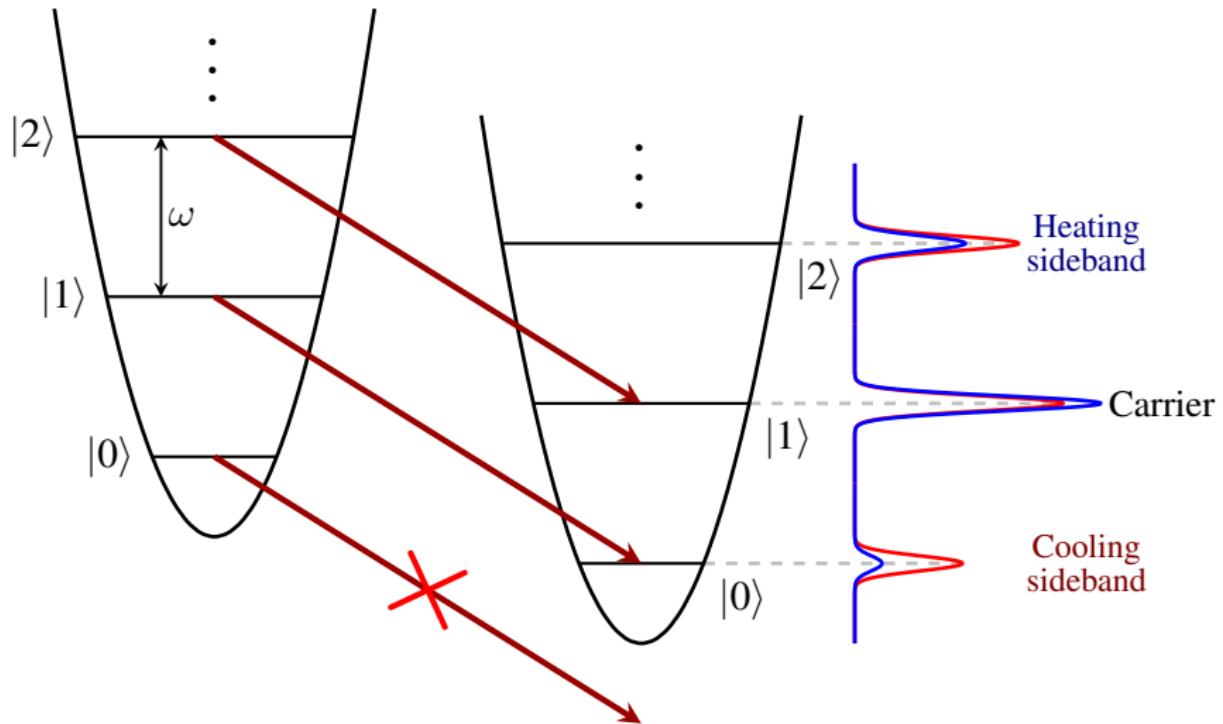
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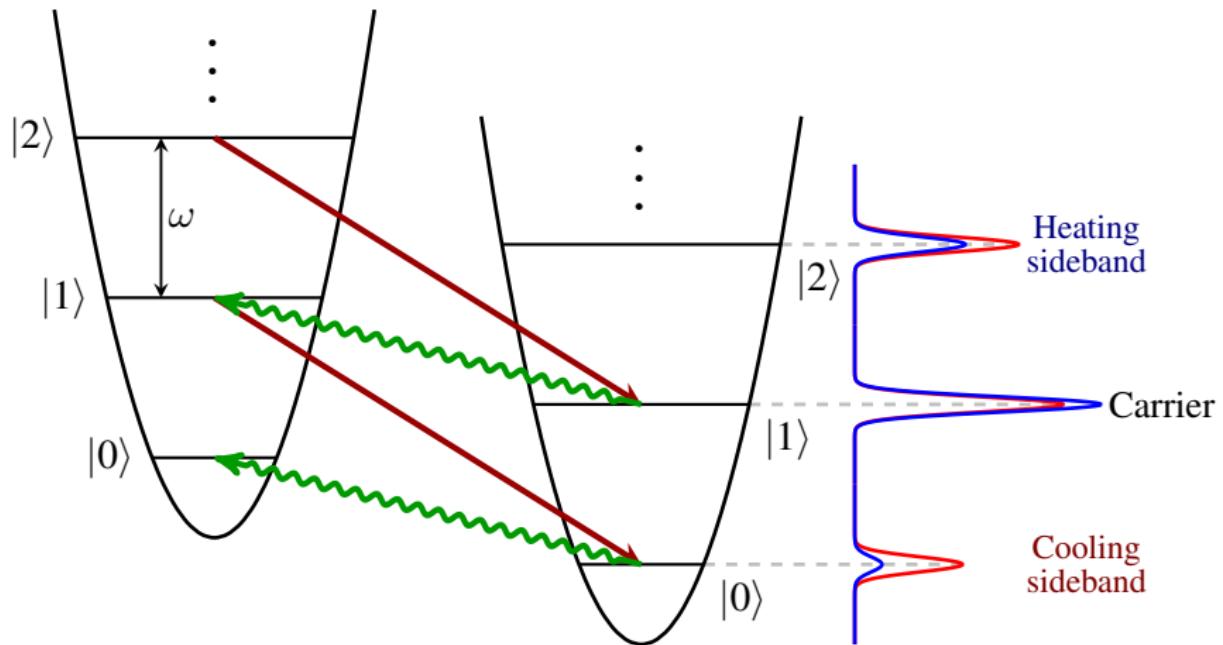
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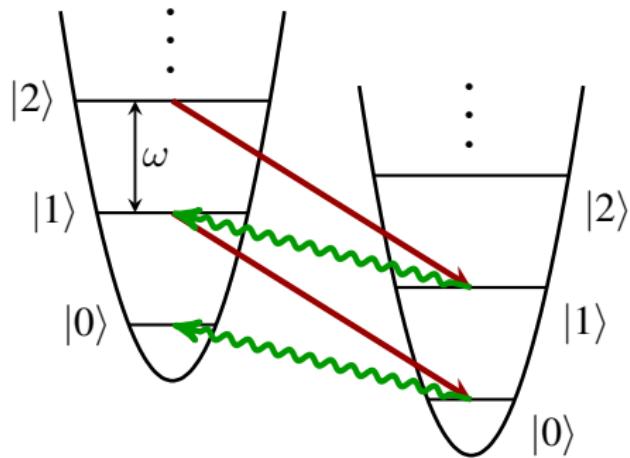


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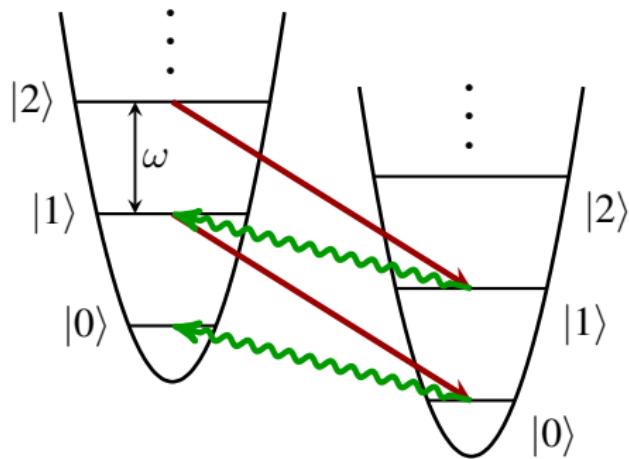
Raman sideband cooling



Raman sideband cooling

Lamb Dicke parameter

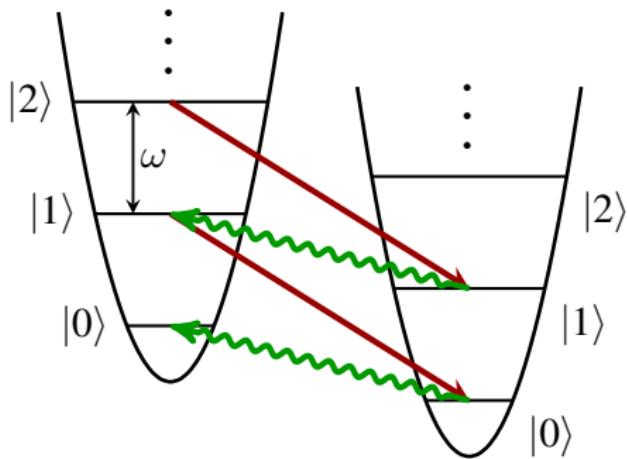
$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$



Raman sideband cooling

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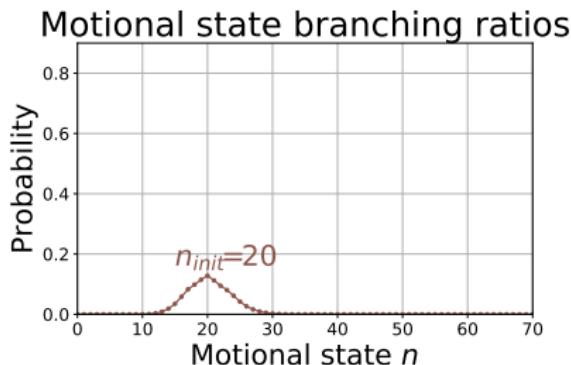
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$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



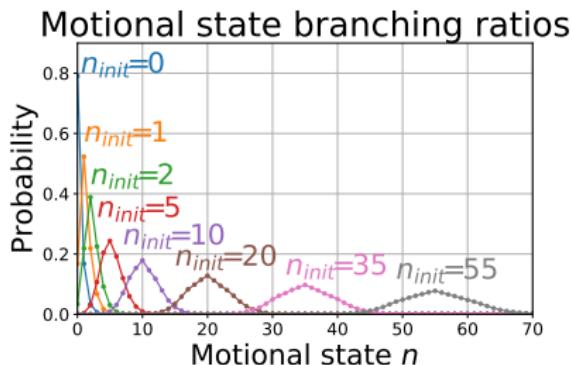
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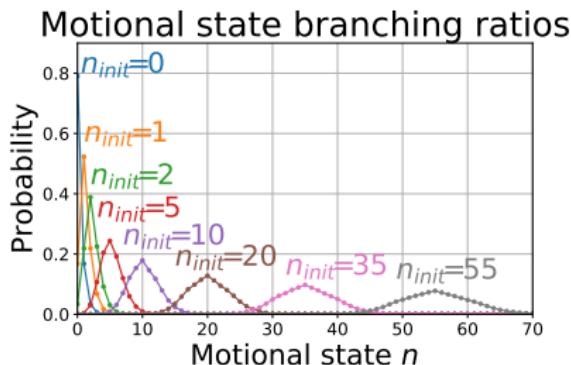
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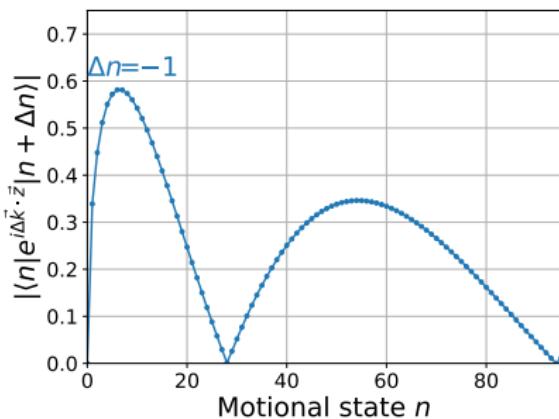


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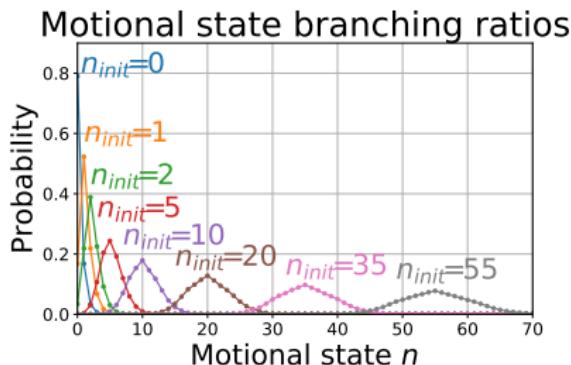
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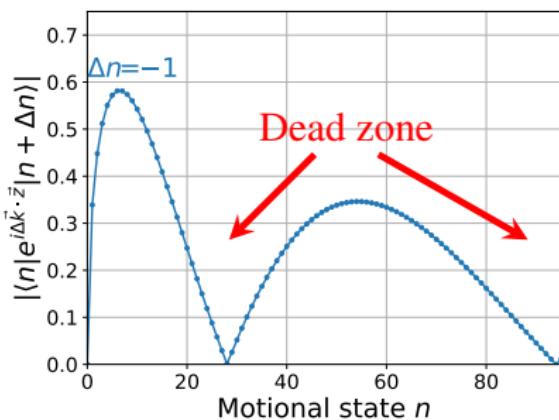


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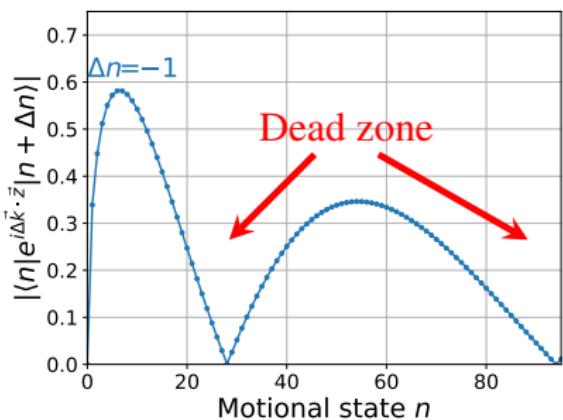
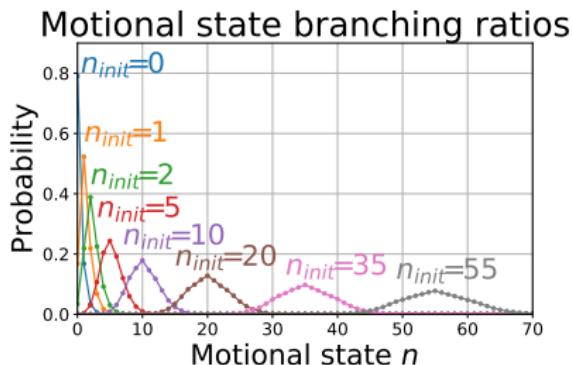
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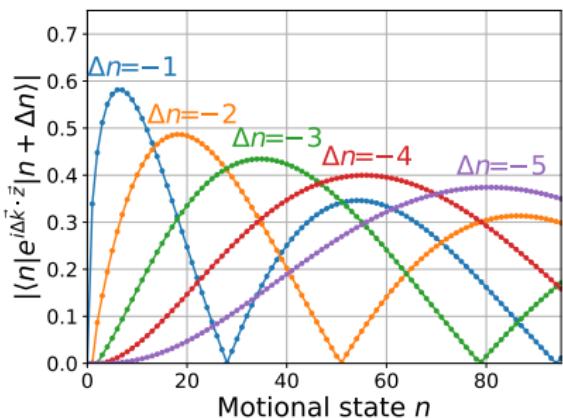
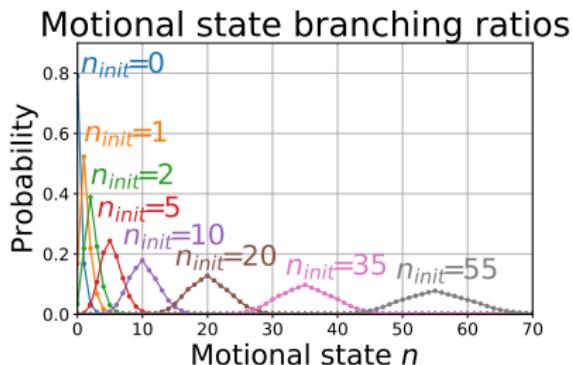
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Solution

- Use higher order sidebands.
- Simulation-guided optimization.

Raman sideband cooling



Lamb Dicke parameter

$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$

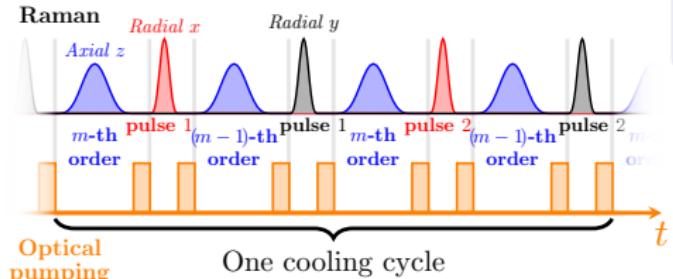
$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \text{ } \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

Solution

- Use higher order sidebands.
- Simulation-guided optimization.

Raman sideband cooling

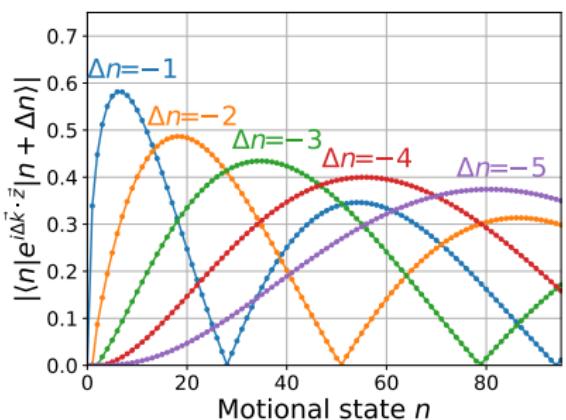


Lamb Dicke parameter

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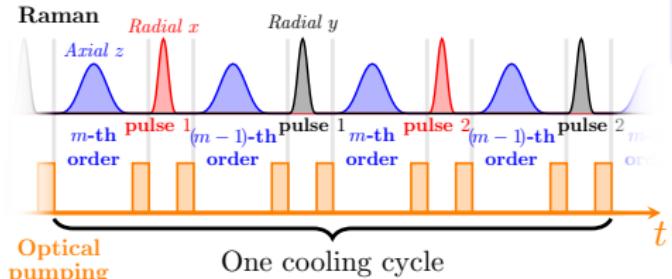
- Motional state branching
- Coupling “dead zone”



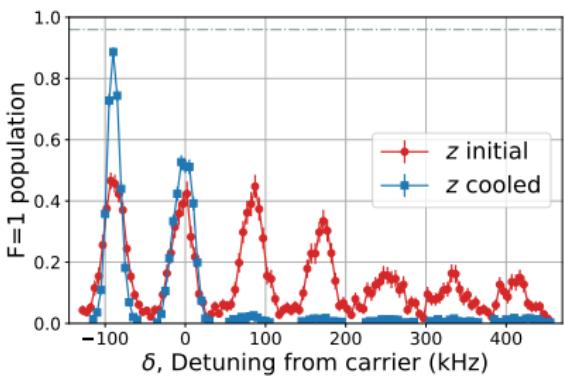
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Raman sideband cooling



Axial sideband spectrum



Lamb Dicke parameter

$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$

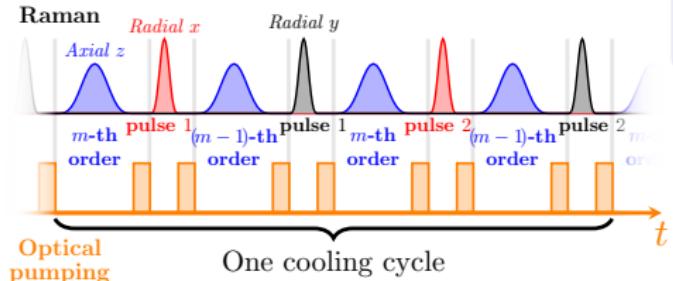
$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

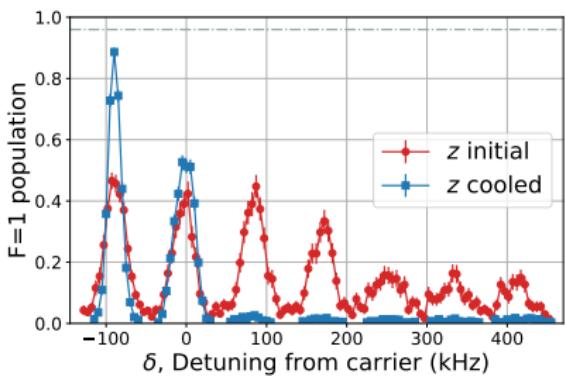
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Axial sideband spectrum



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- Motional state branching
- Coupling “dead zone”

Solution

- Use higher order sidebands.
- Simulation-guided optimization.

3D ground state: 93.5(7)%

Outline

1 Experiment overview

2 Atom state control

- Raman sideband cooling of Na atoms

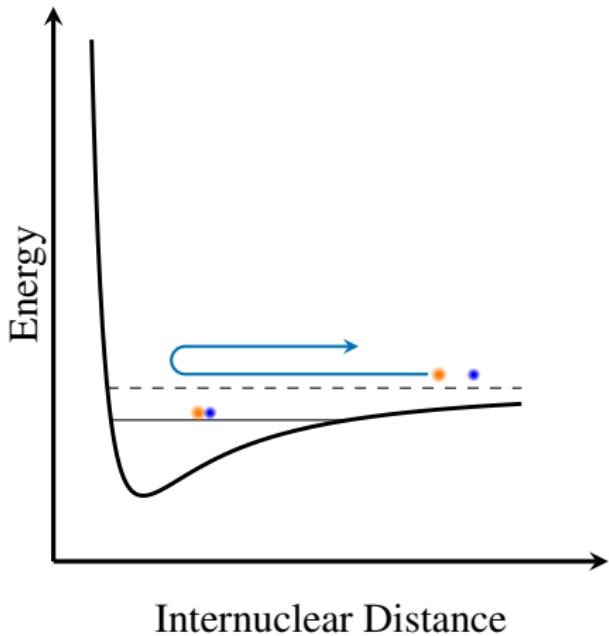
3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

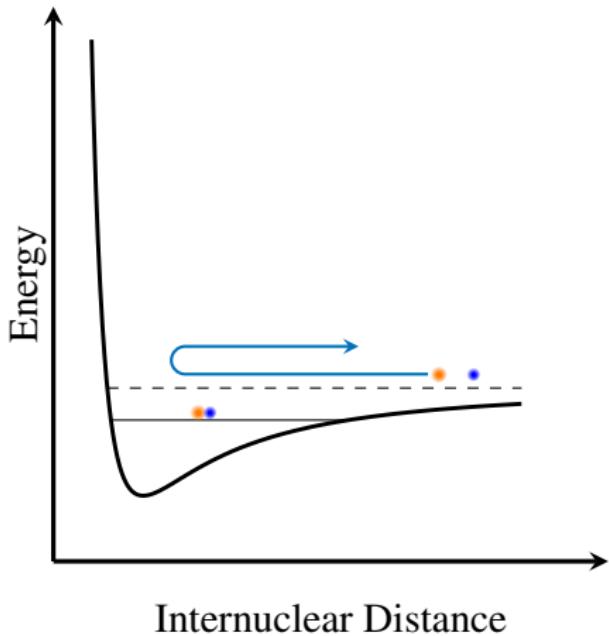
Scattering length a

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



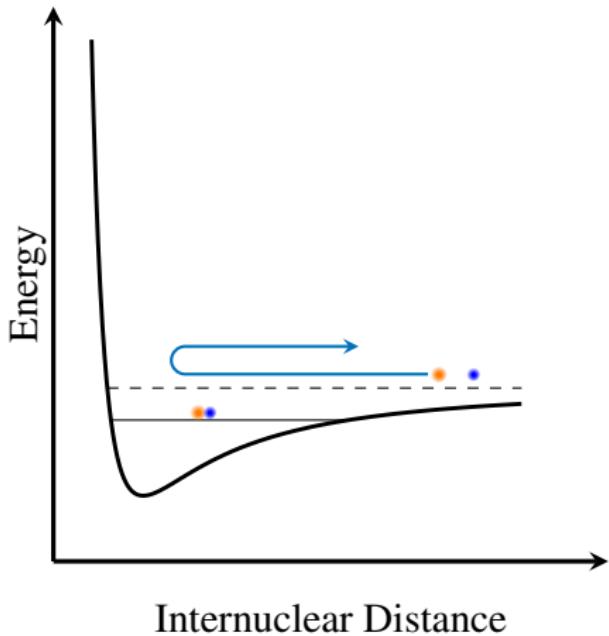
Scattering length a

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



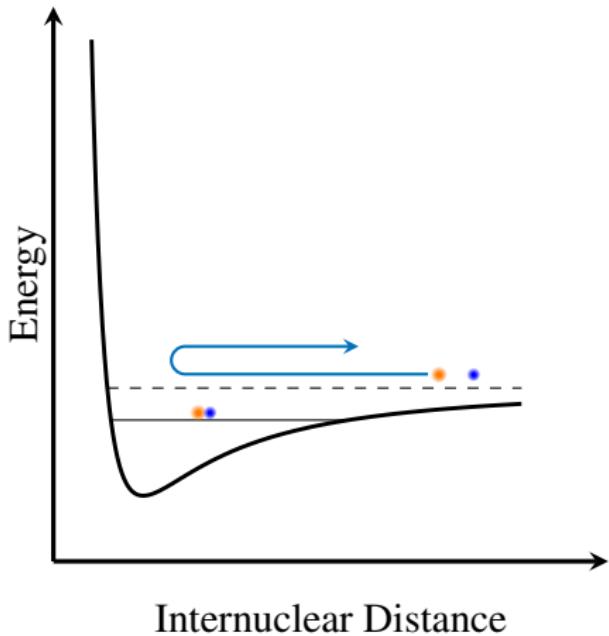
Scattering length a

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



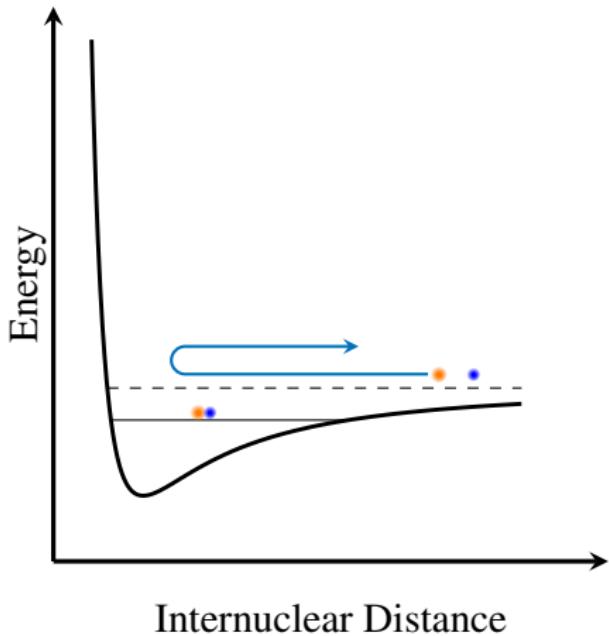
Scattering length a

- Binding energy
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- ⋮

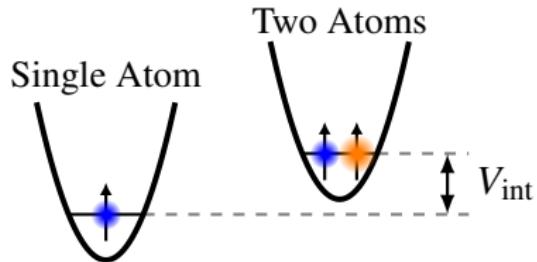


Scattering length a

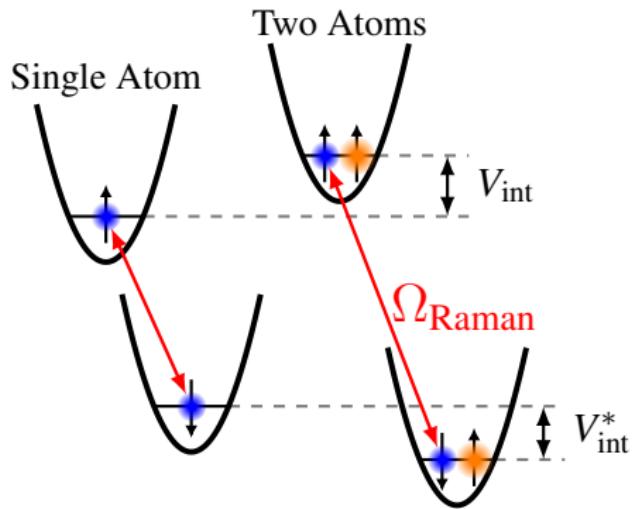
- Binding energy
- Molecular potential
- Molecule formation
- Feshbach resonance
- :



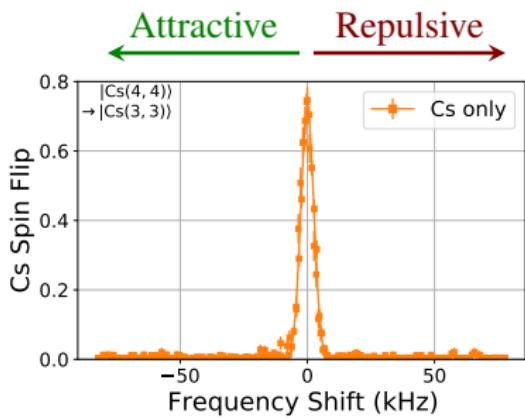
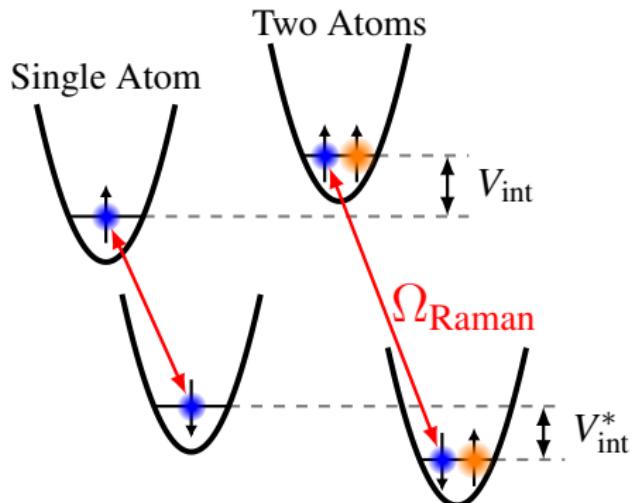
Interaction shift



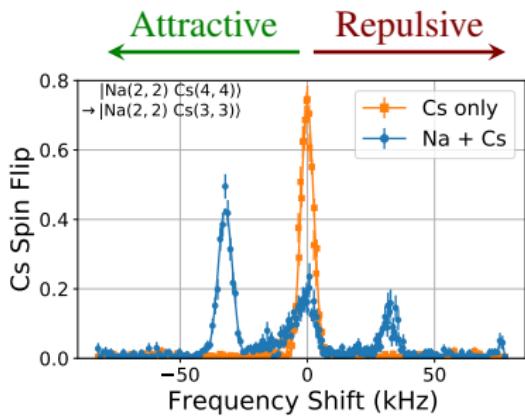
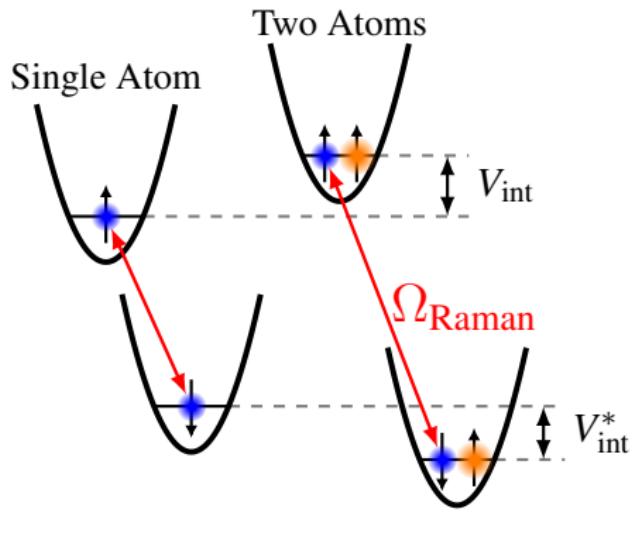
Interaction shift



Interaction shift

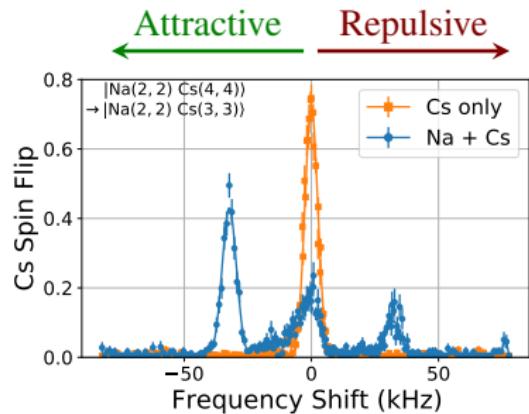


Interaction shift



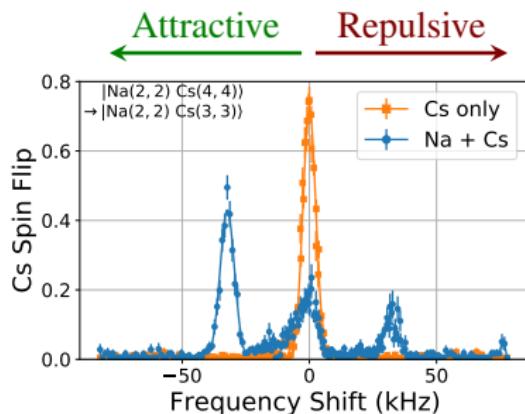
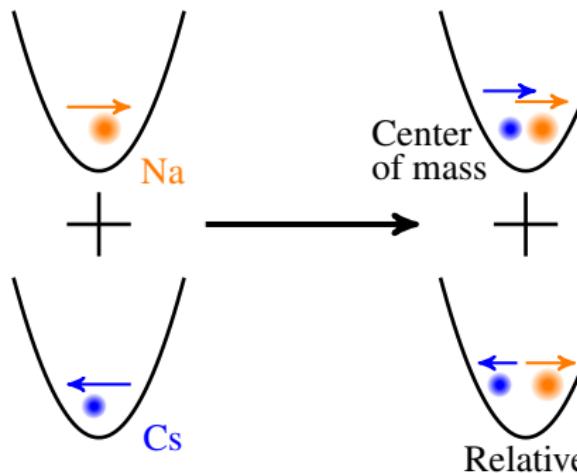
Interaction shift

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



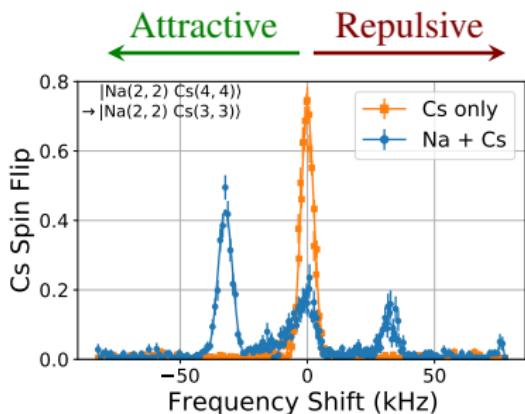
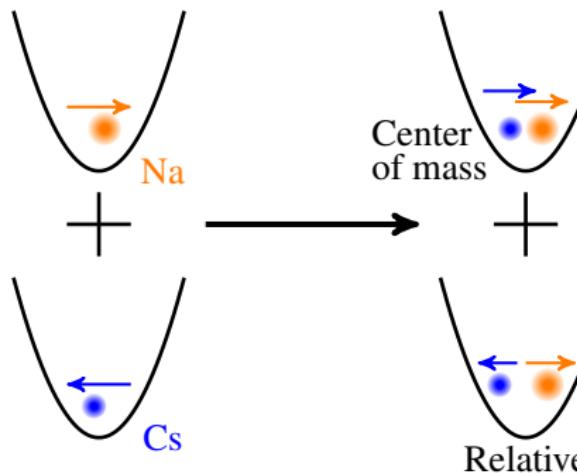
Interaction shift

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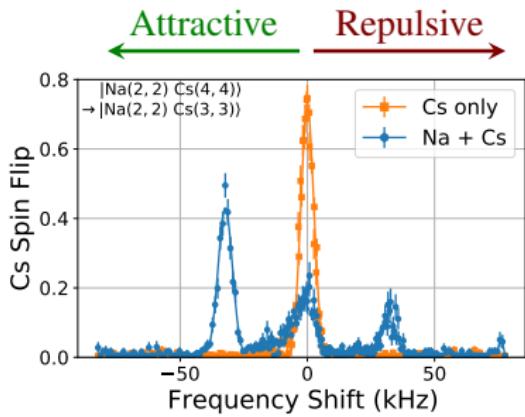
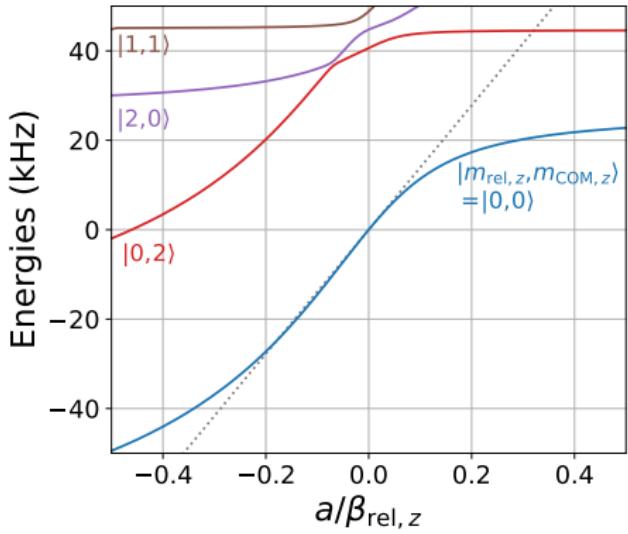
Interaction shift

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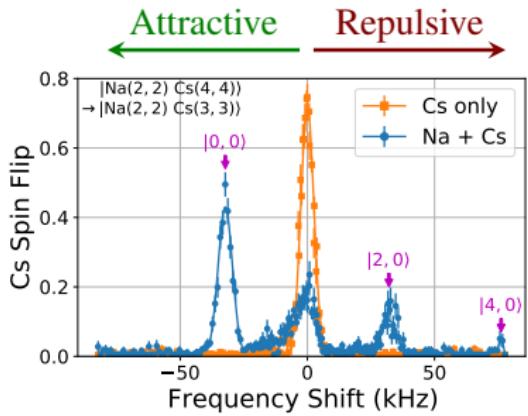
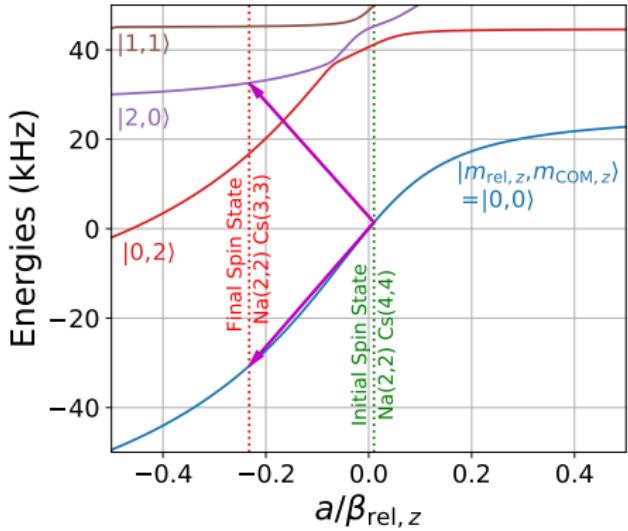
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



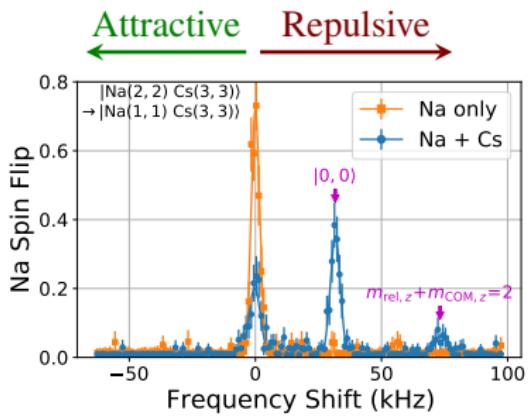
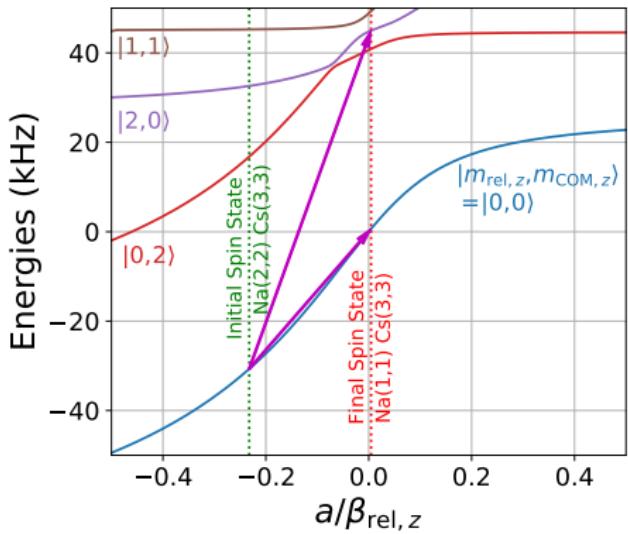
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Interaction shift



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

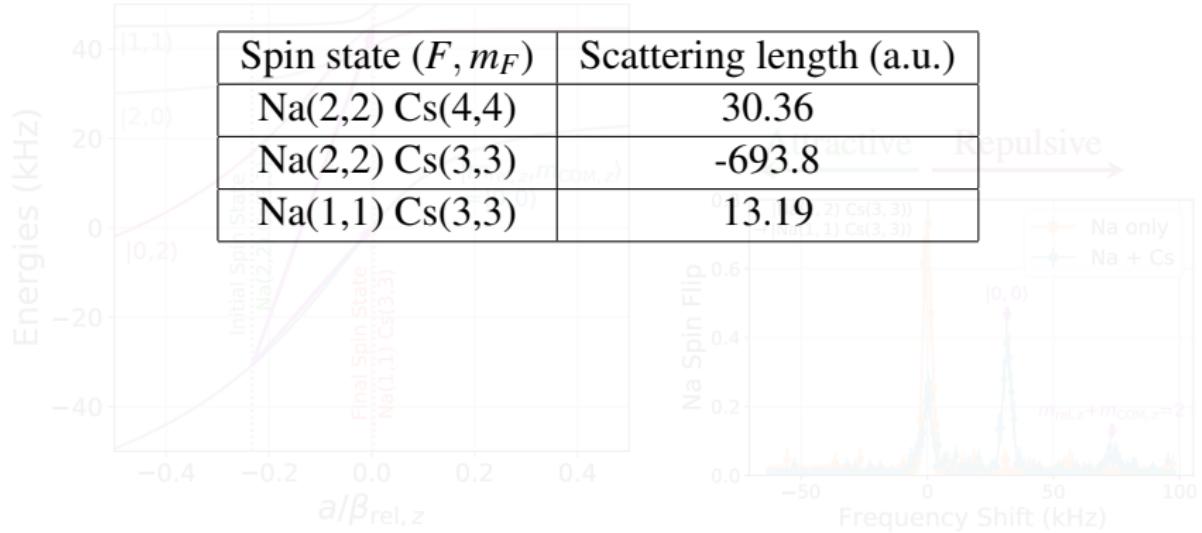
Interaction shift



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

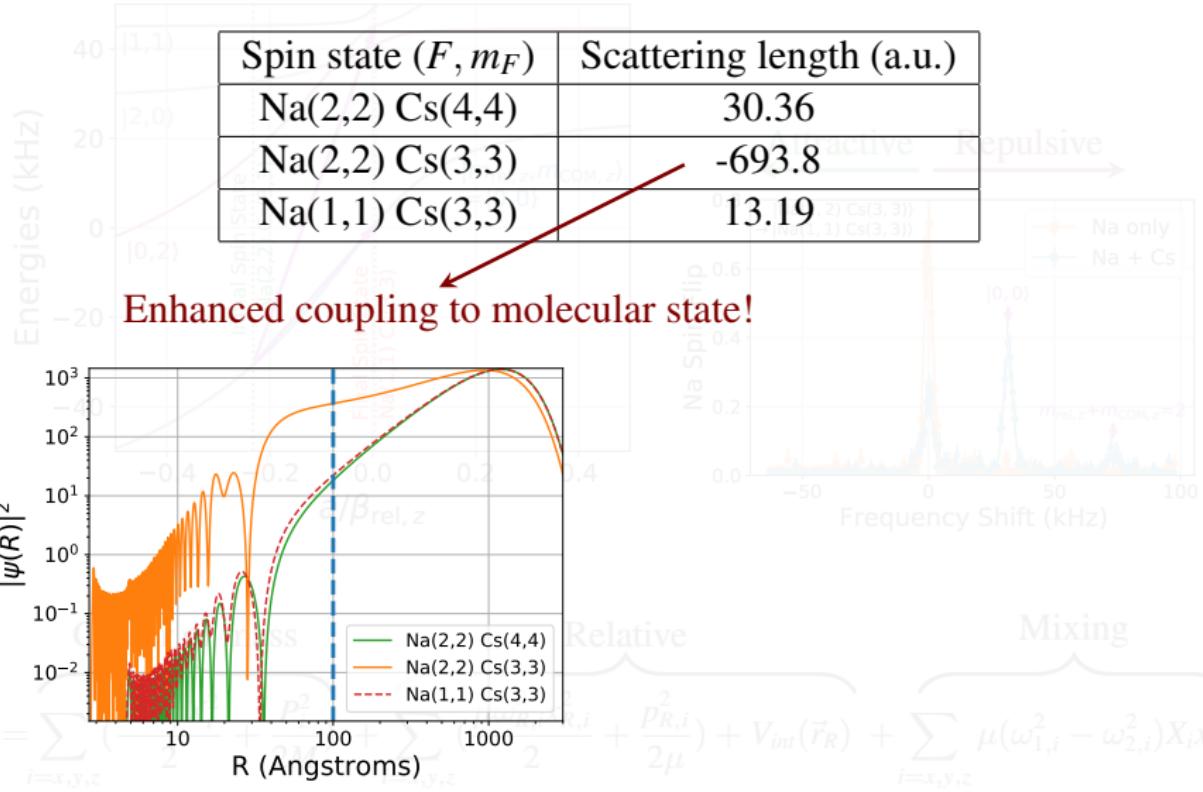
Interaction shift

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



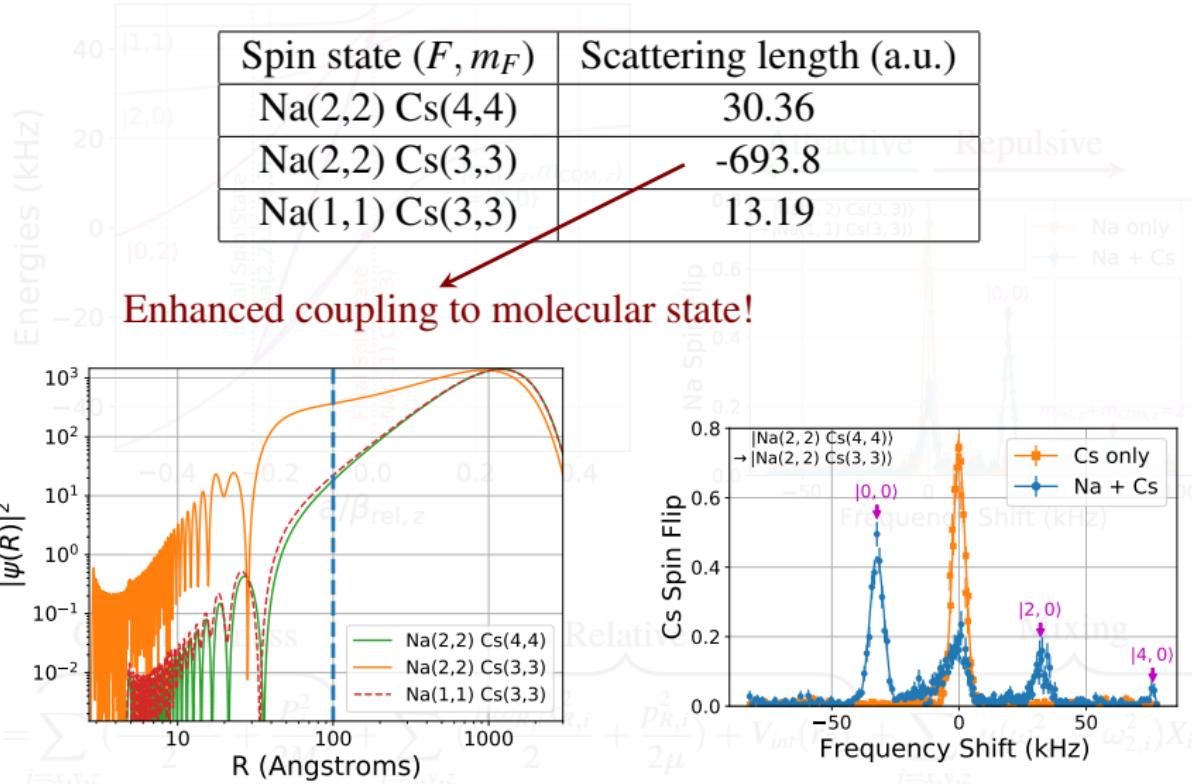
Interaction shift

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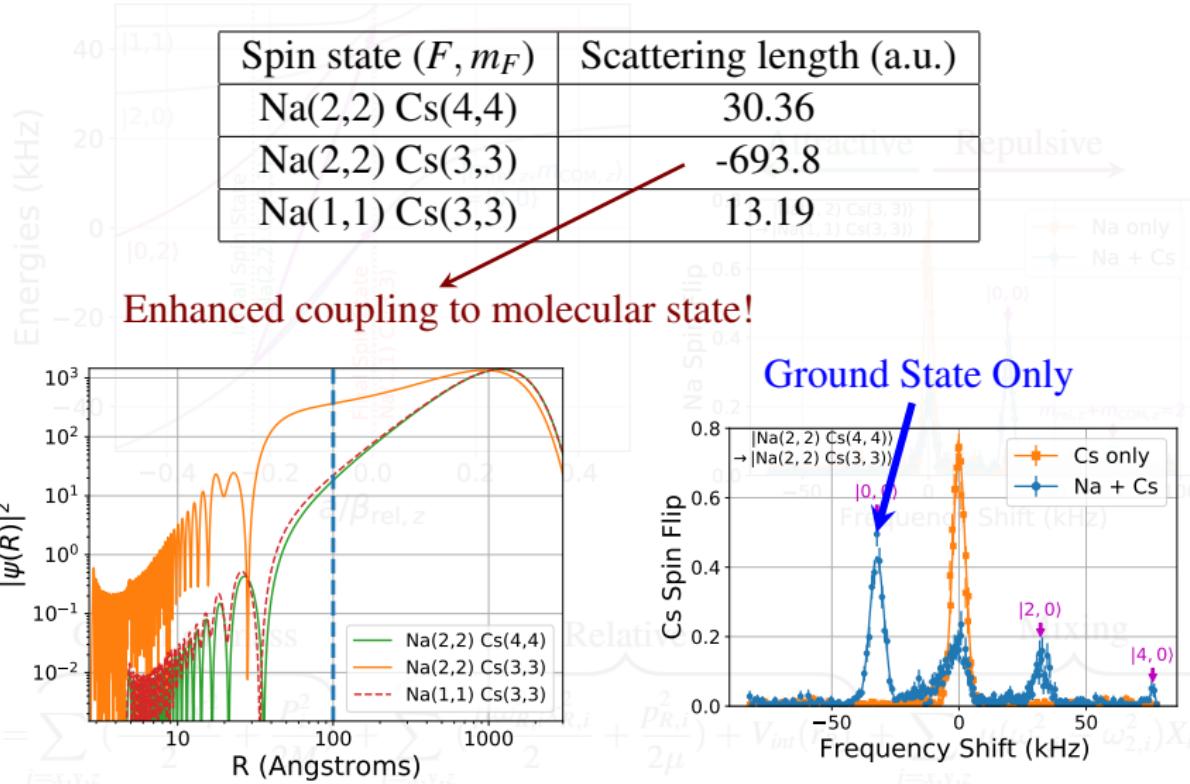
Interaction shift

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Interaction shift

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



Outline

1 Experiment overview

2 Atom state control

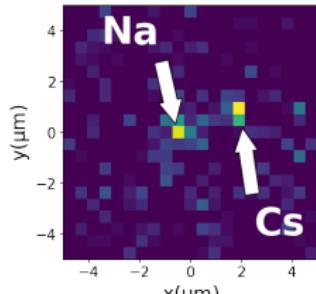
- Raman sideband cooling of Na atoms

3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

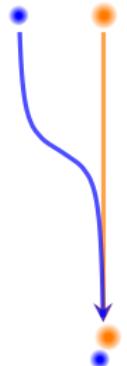
4 Conclusion

Loading

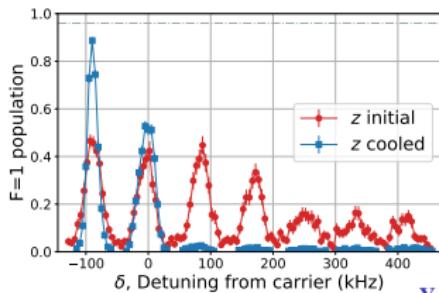


NJP. 19, 023007 (2017)

Merging



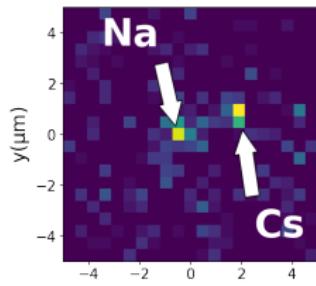
Cooling



Y. Yu et al. PRX. 9, 021039 (2019)

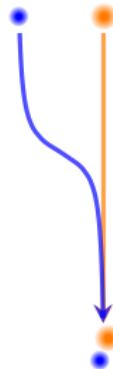
Y. Yu et al. PRA. 97, 063423 (2018)

Loading

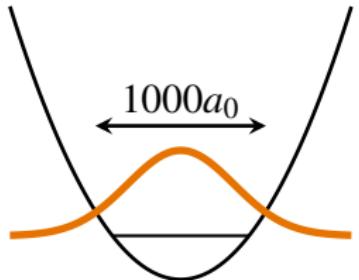


NJP. 19, 023007 (2017)

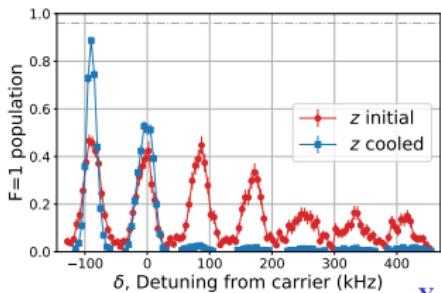
Merging



Atom



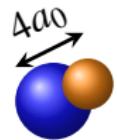
Cooling

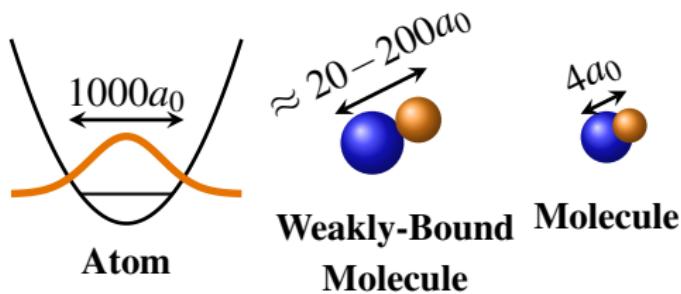


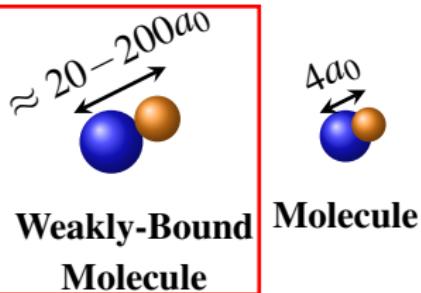
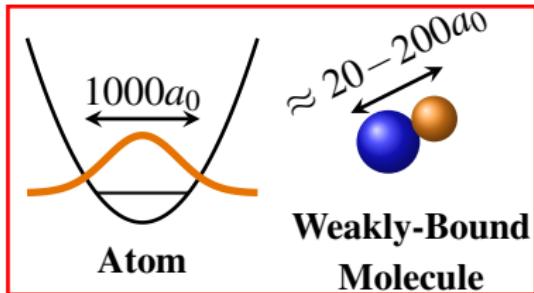
Y. Yu et al. PRA. 97, 063423 (2018)

Y. Yu et al. PRX. 9, 021039 (2019)

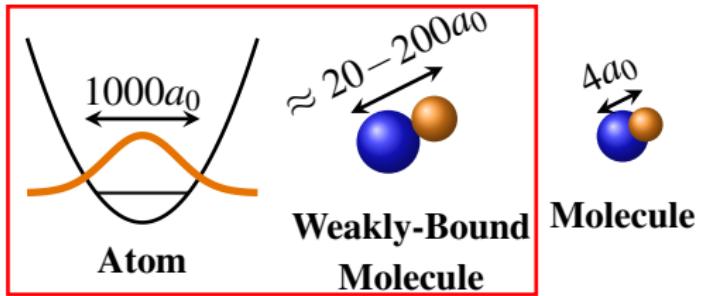
Molecule



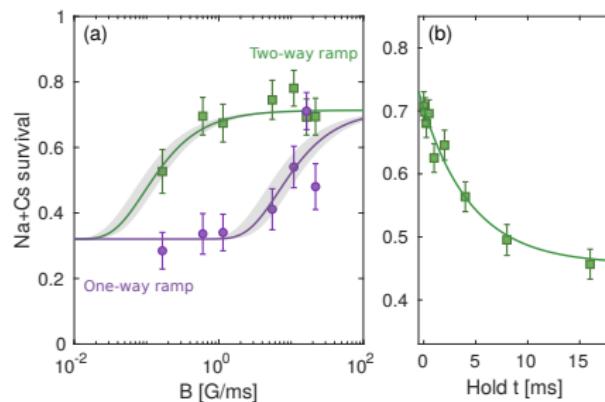




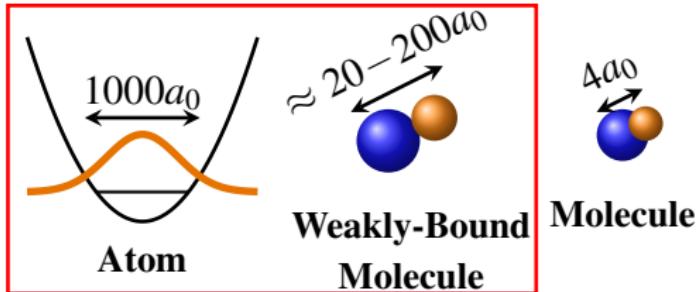
Molecule



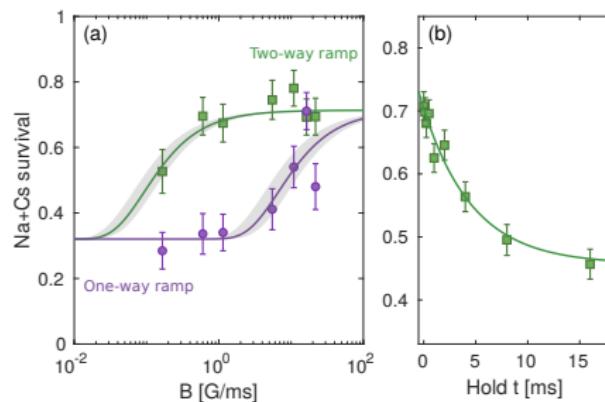
Feshbach molecule



PRL. 124, 253401 (2020)

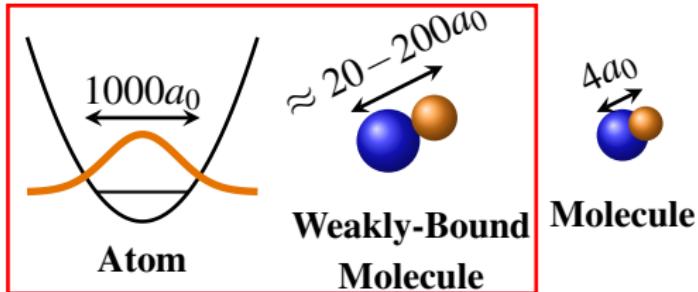


Feshbach molecule



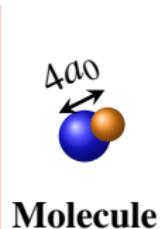
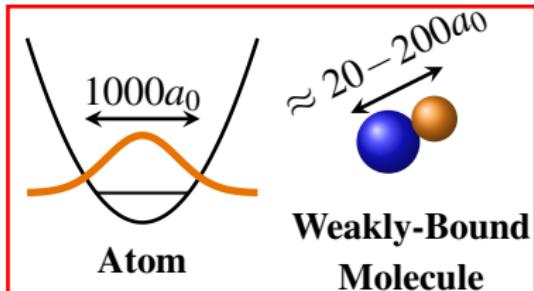
- Requires Feshbach resonance
- Usually large magnetic field

PRL. 124, 253401 (2020)



Optical transfer

- More general
- Faster

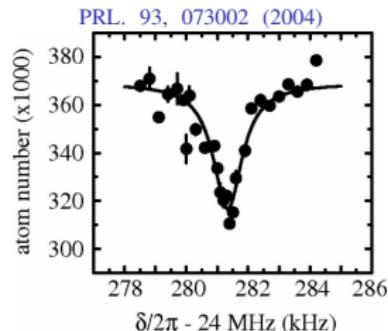


Optical transfer

- More general
- Faster

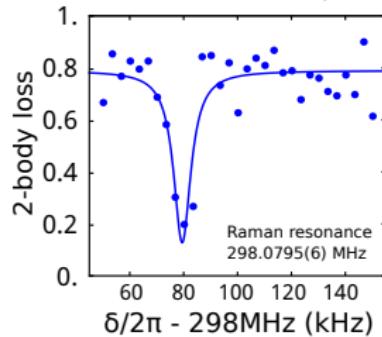
Previous results

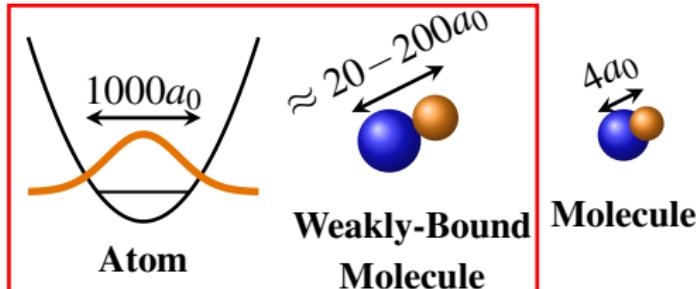
Rb₂ Science 287, 1016 (2000)



Sr₂ PRL. 109, 115302 (2012)

NaCs Y. Yu et al. PRX. 9, 021039 (2019)





Optical transfer

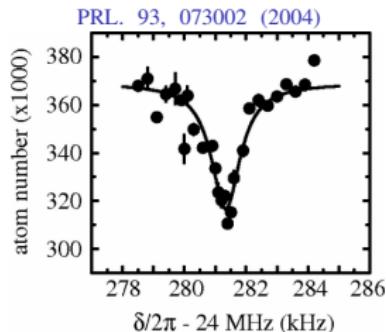
- More general
 - Faster

Limitations so far

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

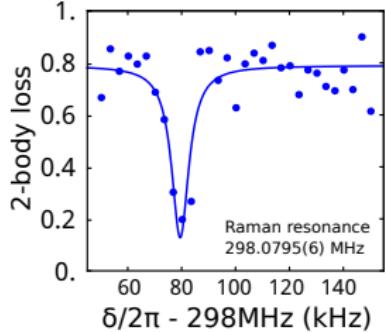
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Rb₂ Science 287, 1016 (2000)

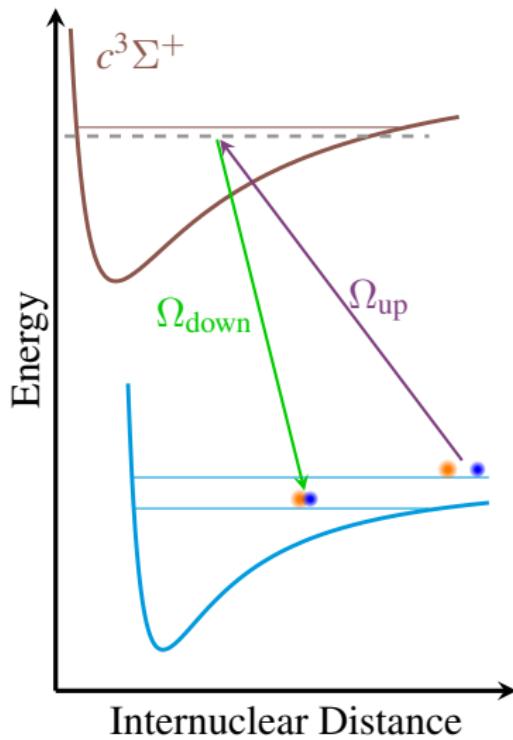


Sr₂ PRL. 109, 115302 (2012)

NaCs Y. Yu et al. PRX. 9, 021039 (2019)



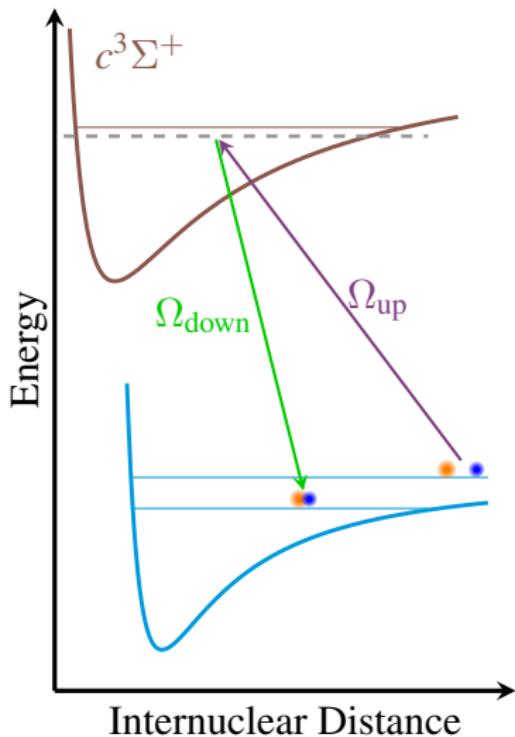
Raman transfer



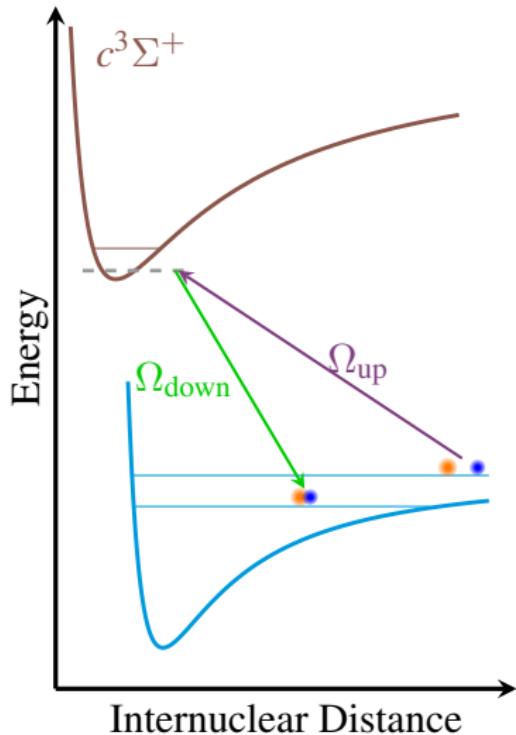
Raman transfer

Near threshold states

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



Raman transfer



Near threshold states

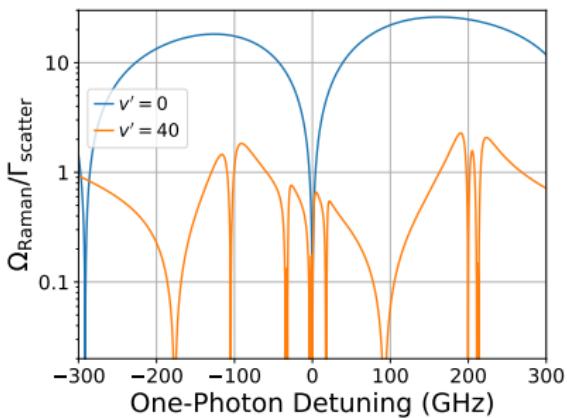
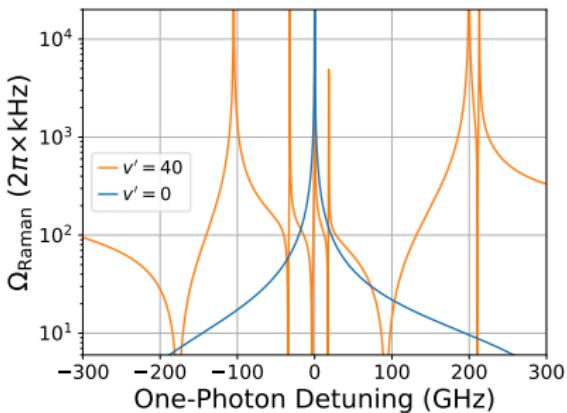
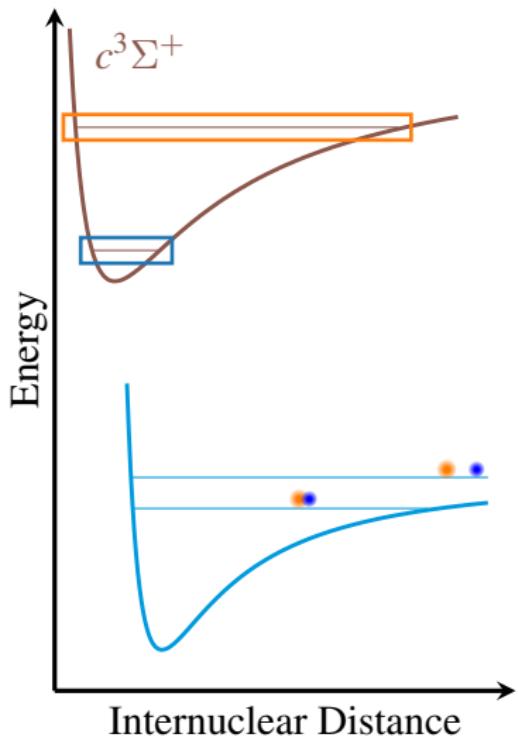
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

Deeply bound states

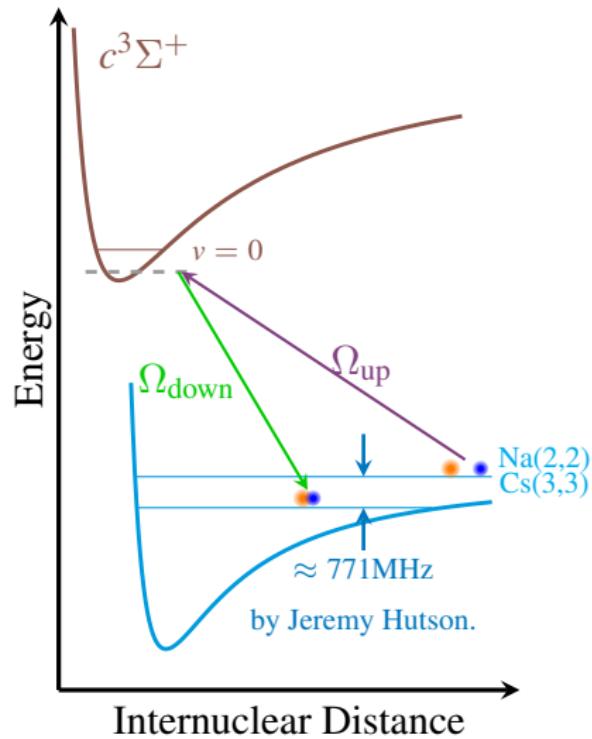
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

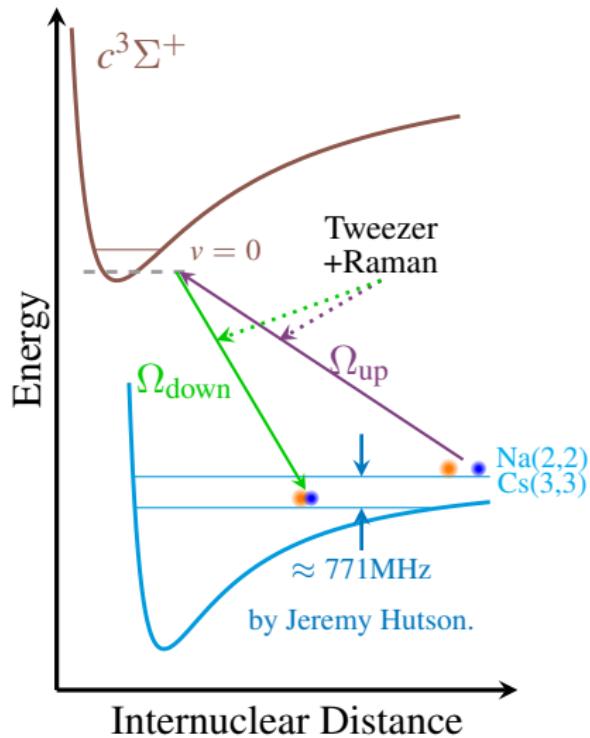
Raman transfer



Experiment



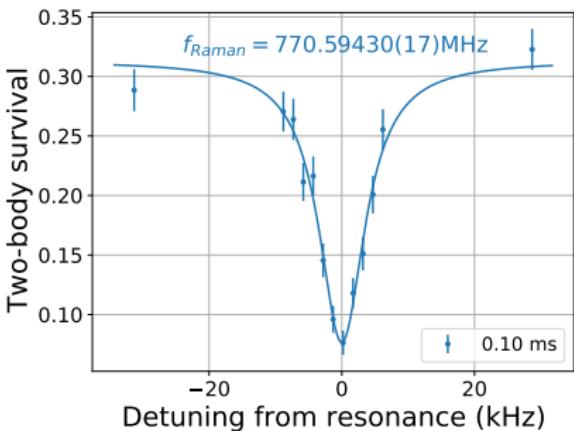
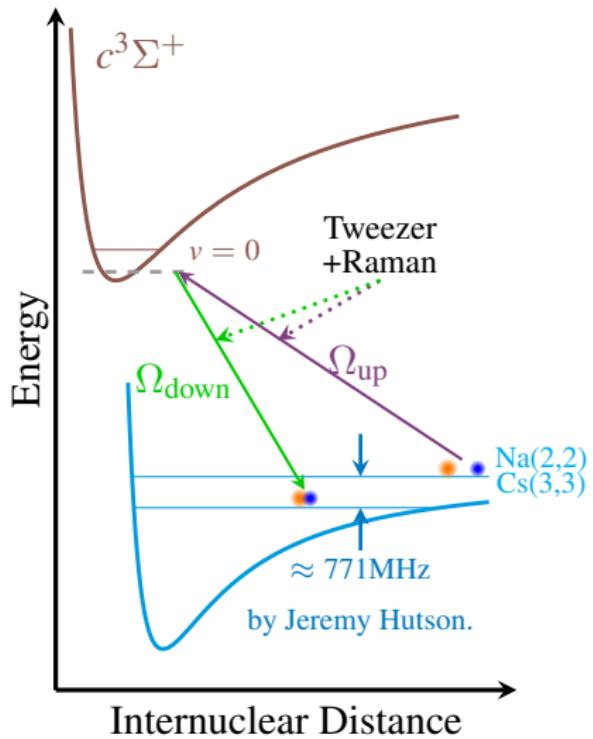
Experiment



Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

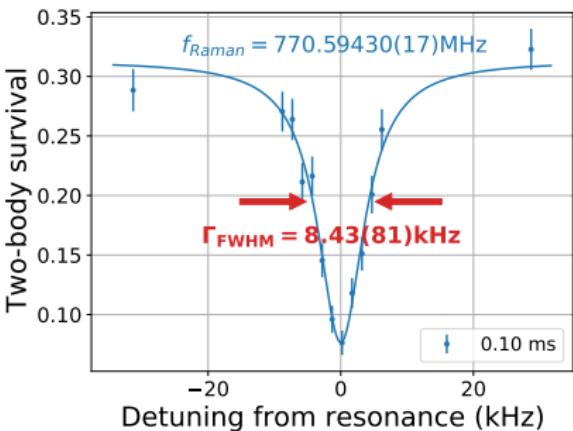
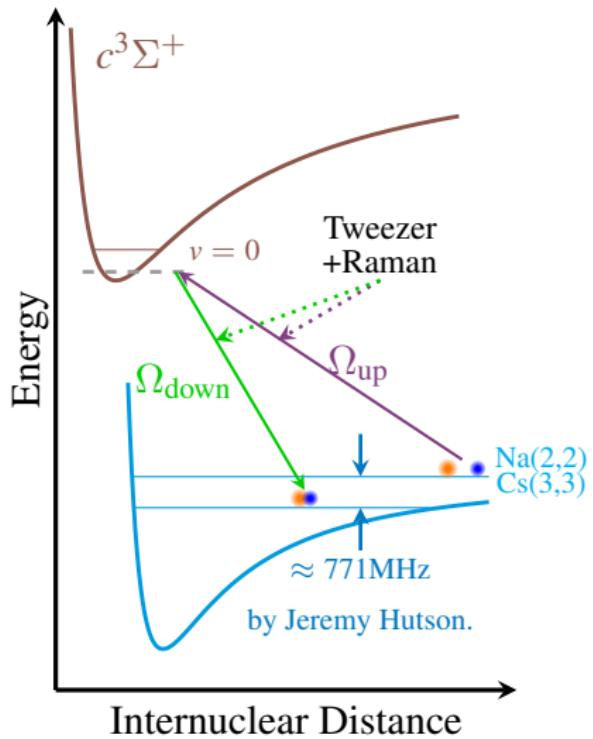
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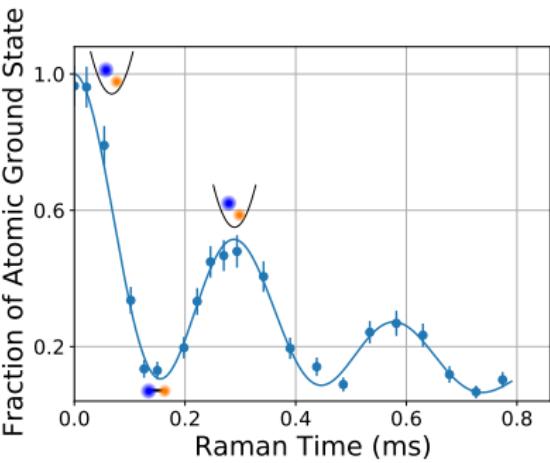
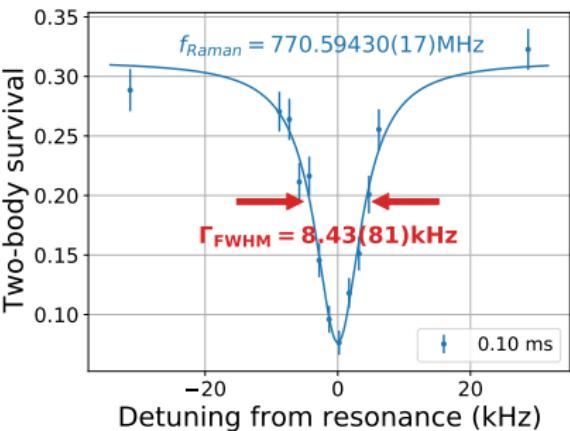
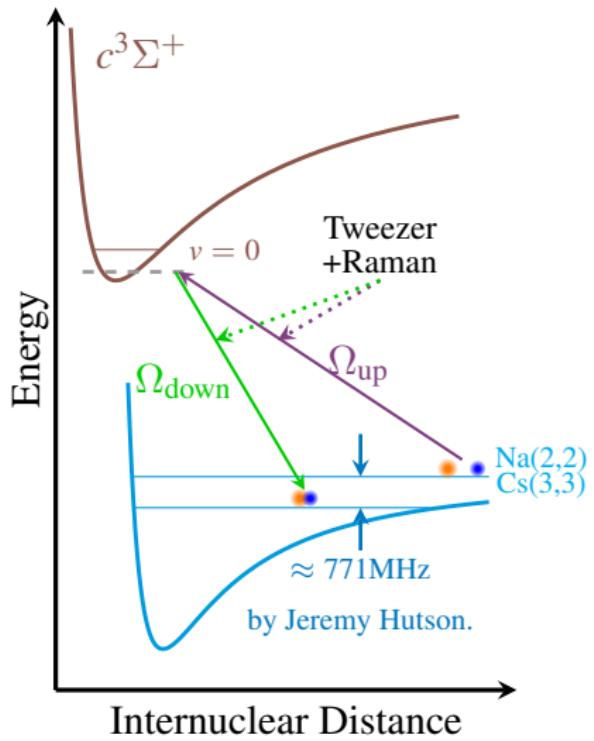
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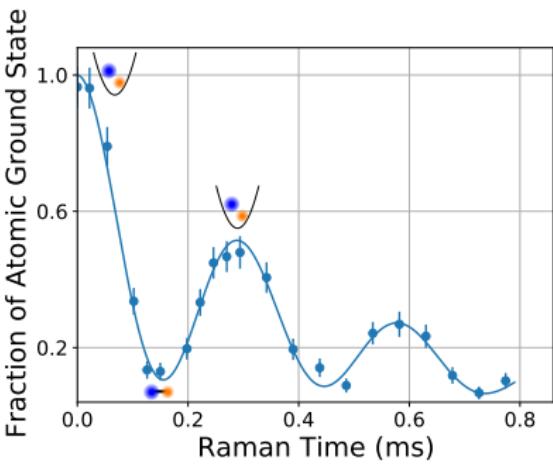
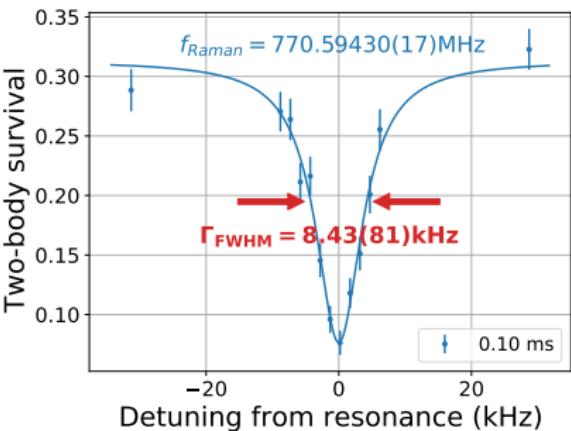
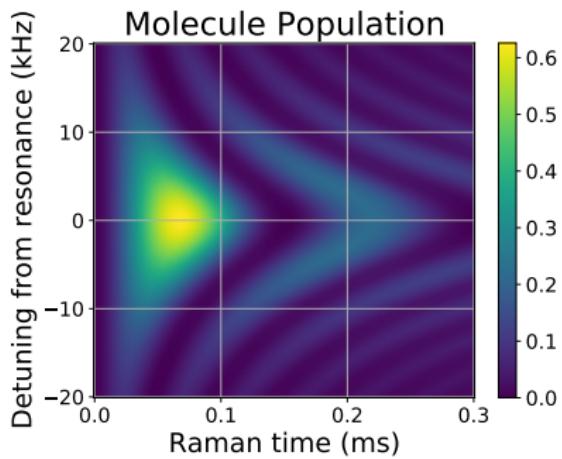
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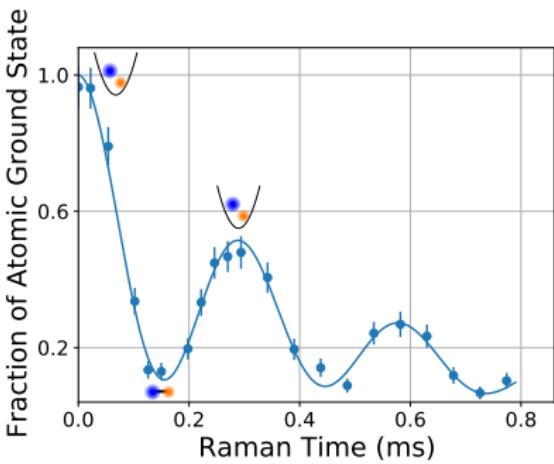
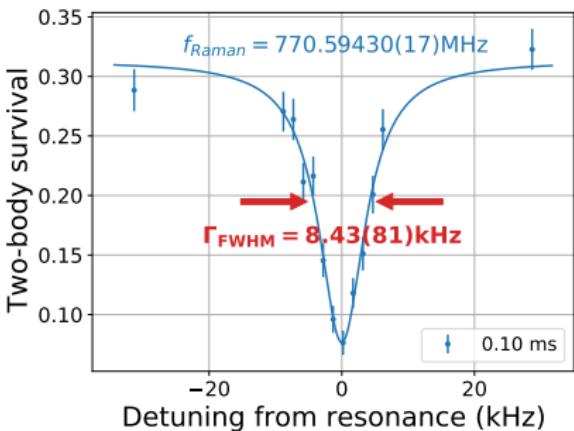
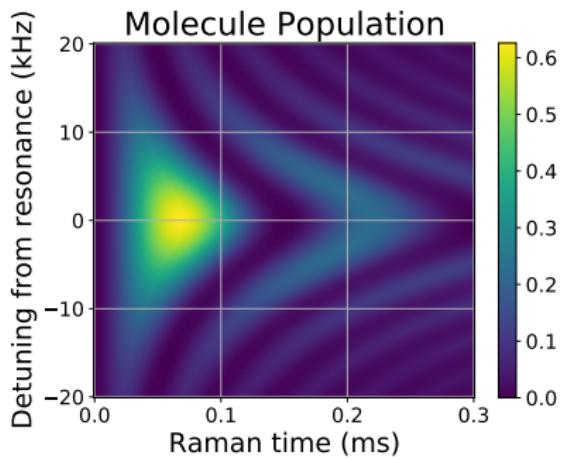
Experiment

- Transferred 63% of ground state atom to molecule.
- Single molecule spin state
- >50% of molecule in motional ground state.
- Limited by molecule lifetime



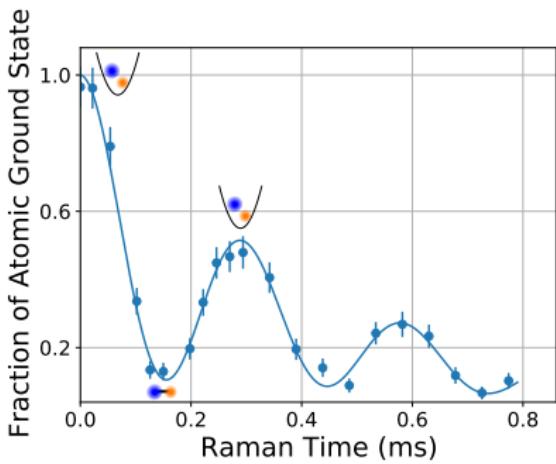
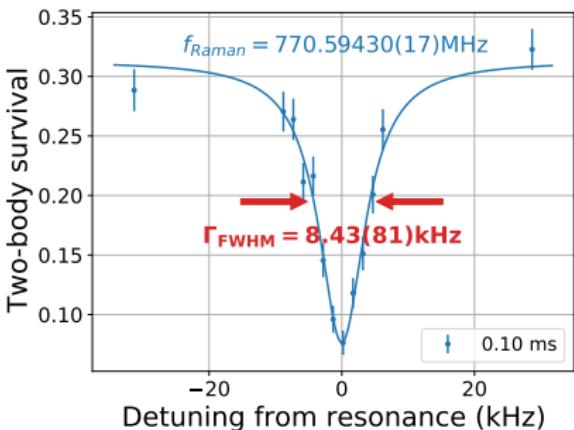
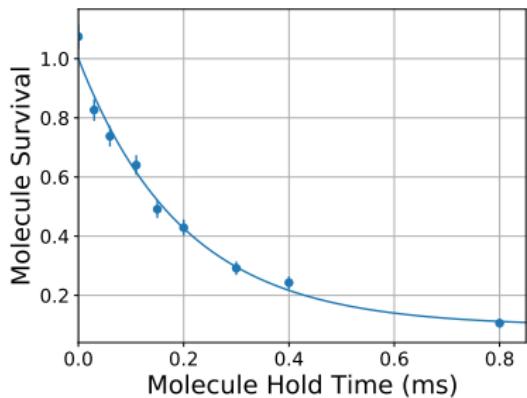
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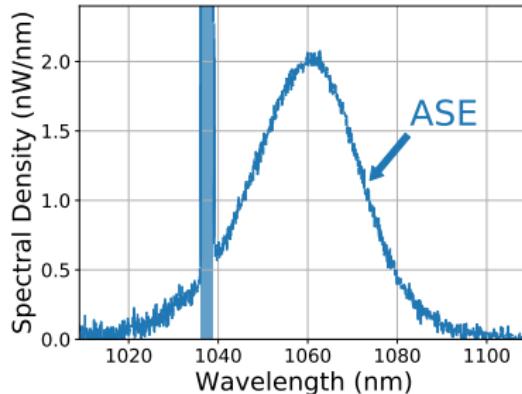


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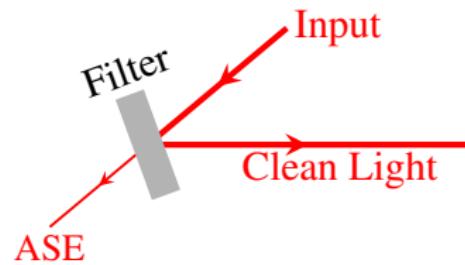
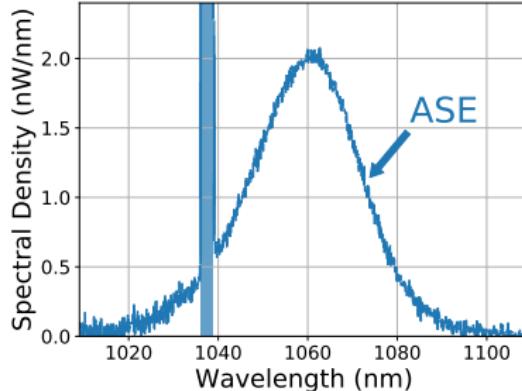
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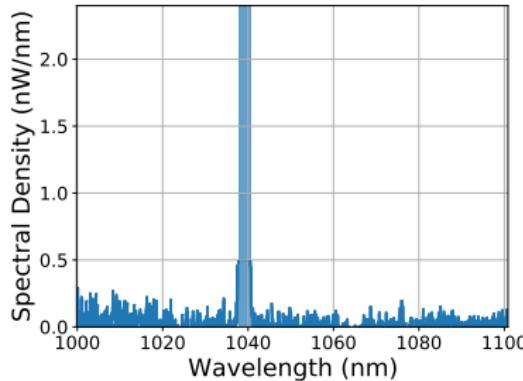
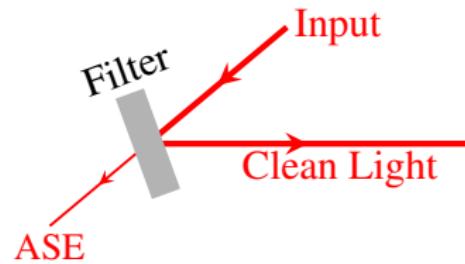
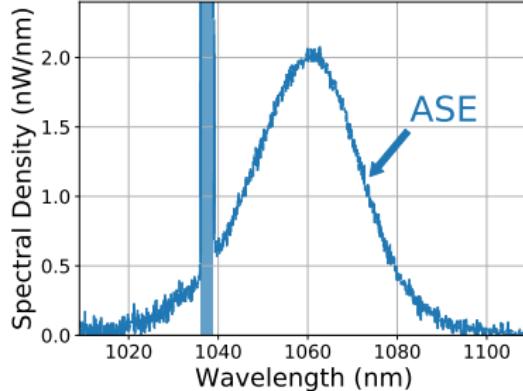
Amplified Spontaneous Emission (ASE)



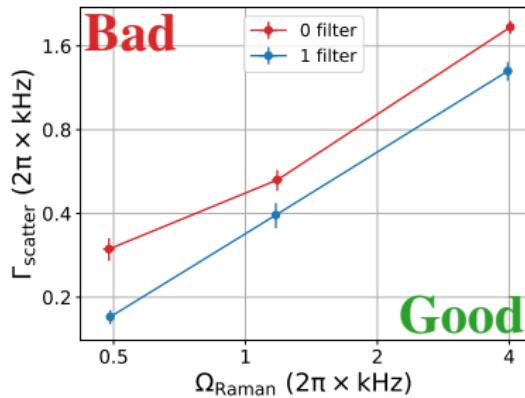
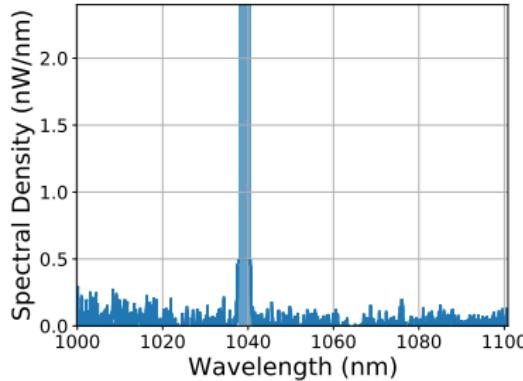
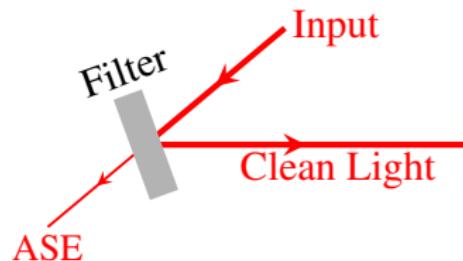
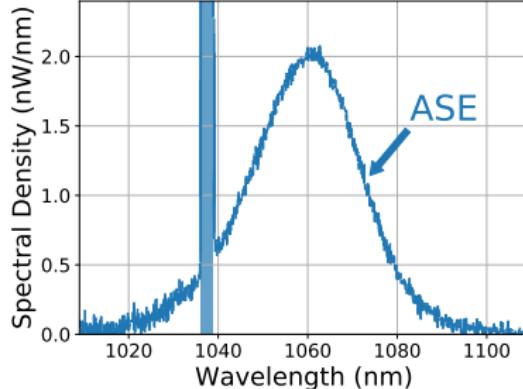
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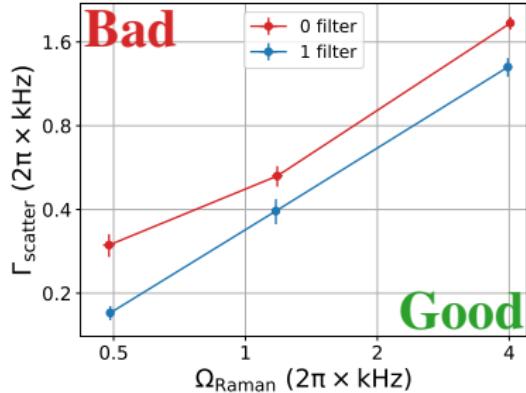
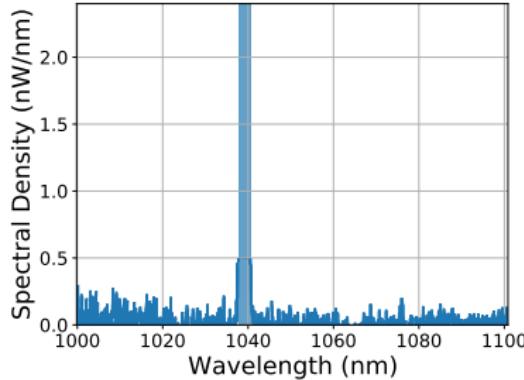
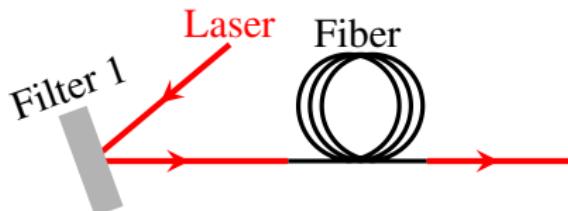
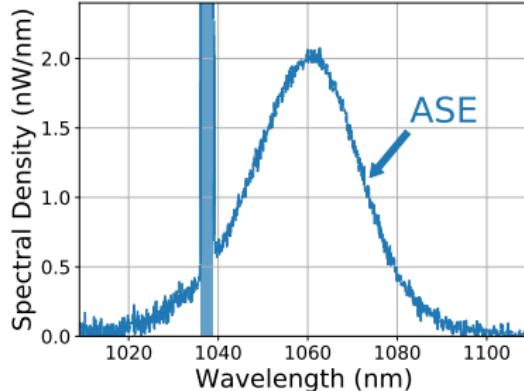
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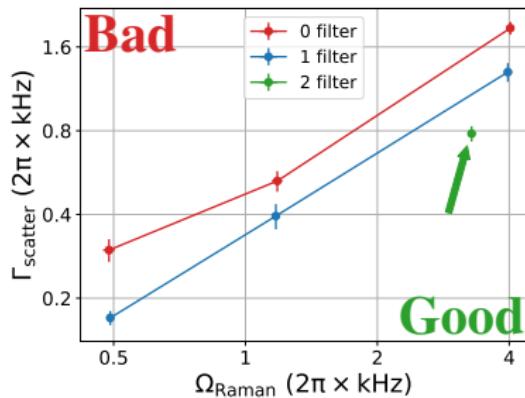
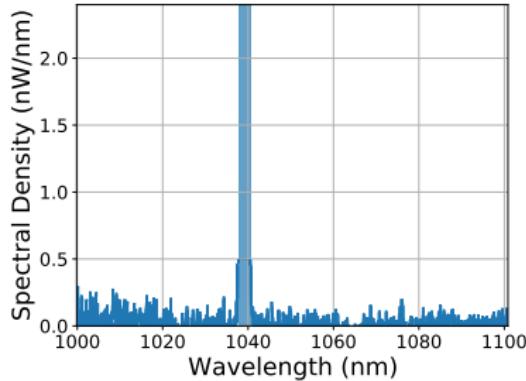
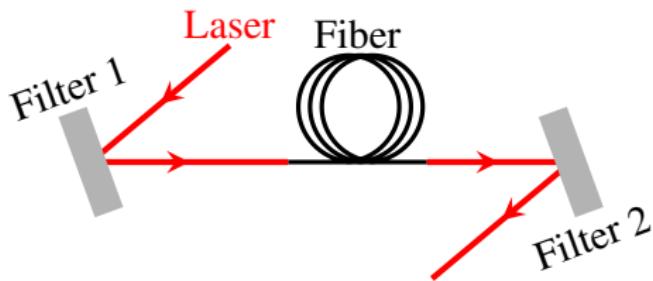
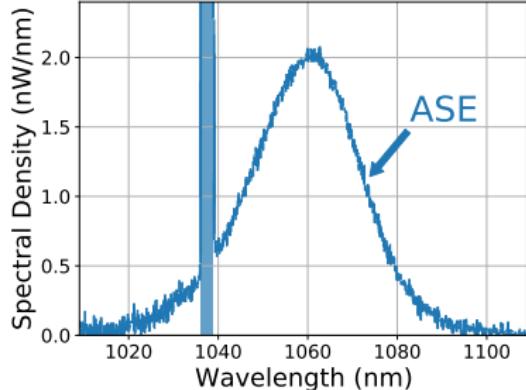
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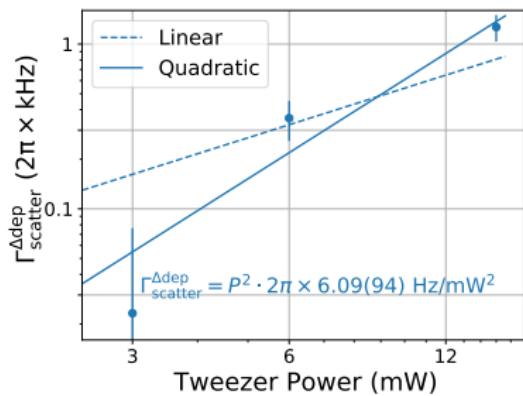


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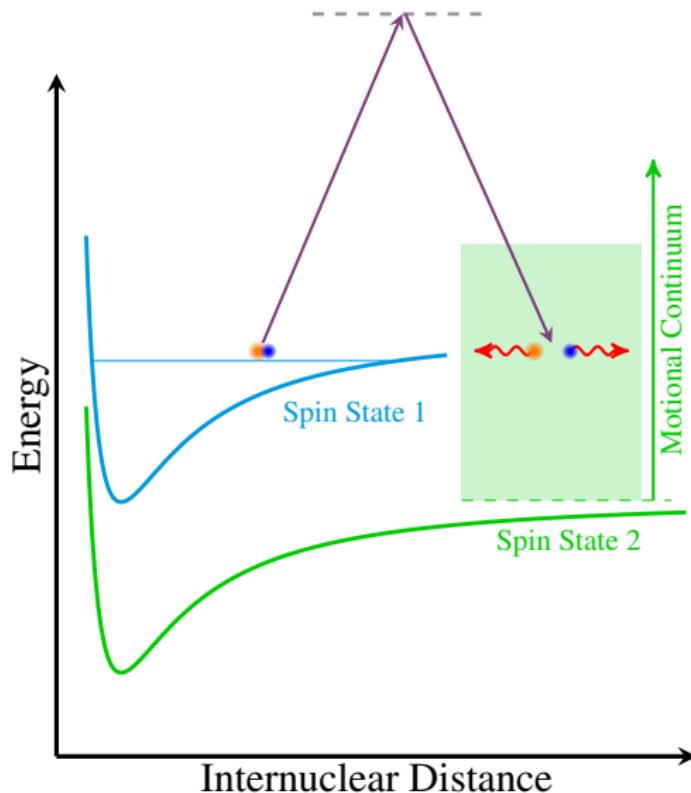
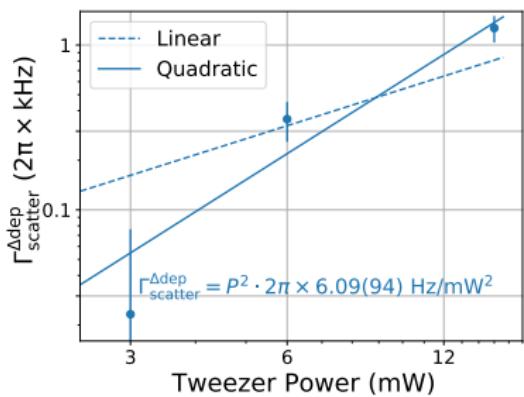
Two-Photon Scattering

Detuning Dependent Scattering



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Conclusion and outlook

- New quantum platform based on ultracold molecules in tweezers
- Full quantum control of atoms in optical tweezers
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- Coherent all-optical creation of single molecule
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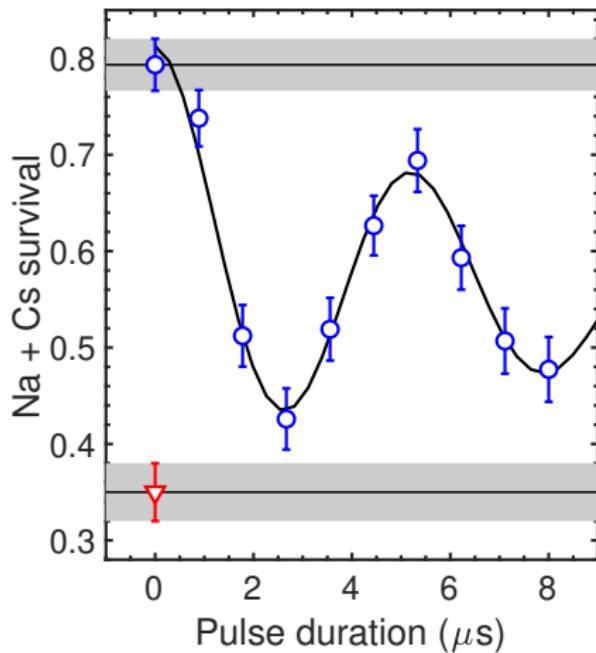
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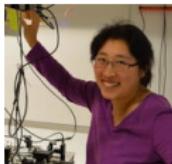
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PI



Kang-Kuen Ni

NaCs
Team



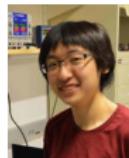
Kenneth
Wang



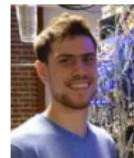
Yu
Wang



Fang
Fang



Jessie
Zhang



Lewis
Picard



William
Cairncross

KRb
Team



Lingbang
Zhu



Mingguang
Hu



Matthew
Nichols



Lee Liu
Postdoc @JILA



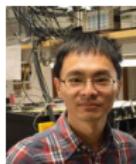
Nick Hutzler
AP @Caltech



Jonathan Hood
AP @Purdue



Eliot
Fenton



Yen-Wei Lin
Intelon Optics



Yu Liu
Postdoc @NIST

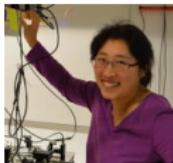


Andrei
Gheorghe
Instructor @MIT



David Grimes
Instructor @MIT

PI



Kang-Kuen Ni



NaCs
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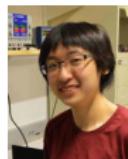
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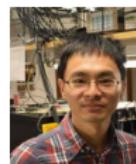
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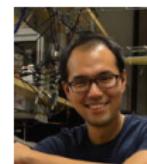
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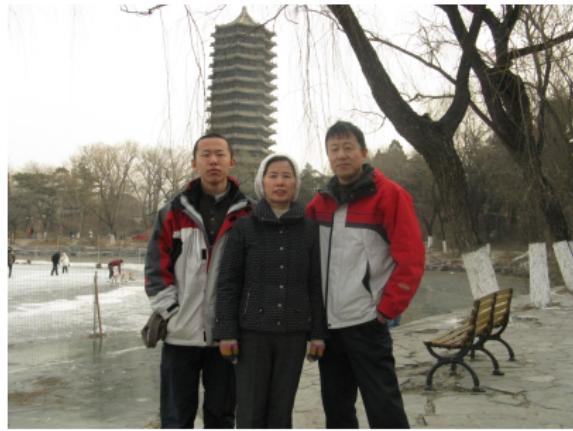
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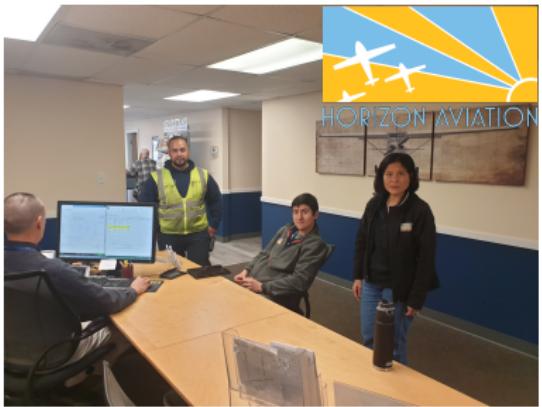
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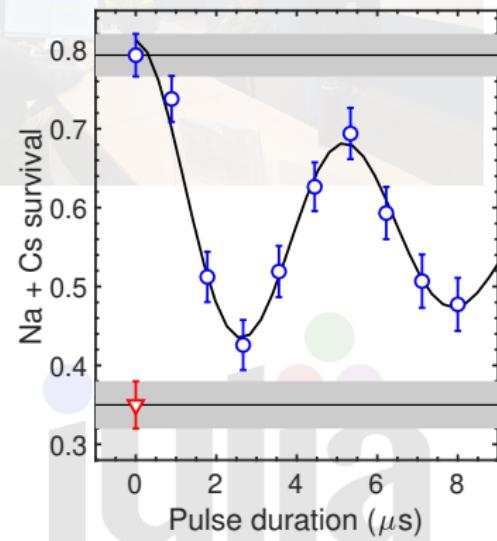
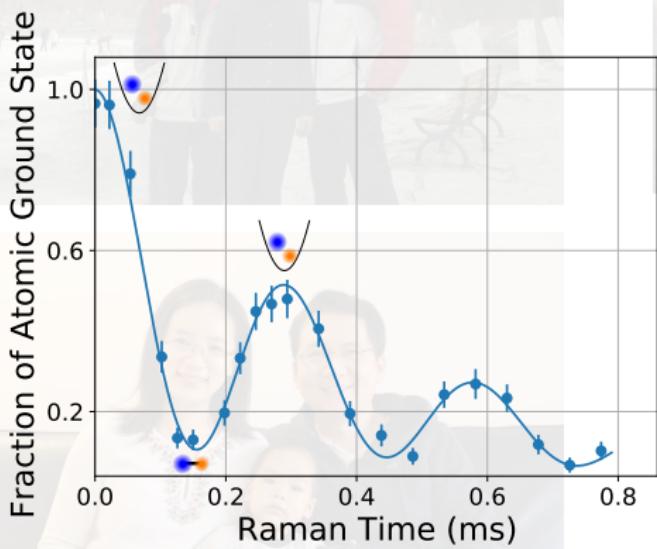
David Grimes
Instructor @MIT







Thanks for your attention

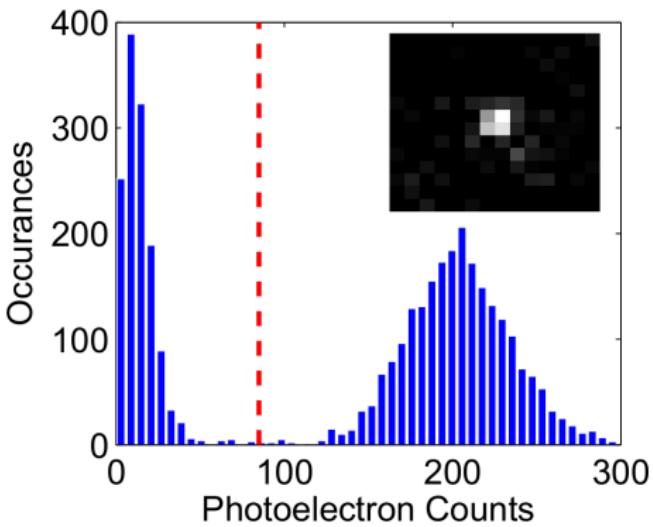


Single Atom in Tweezer

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

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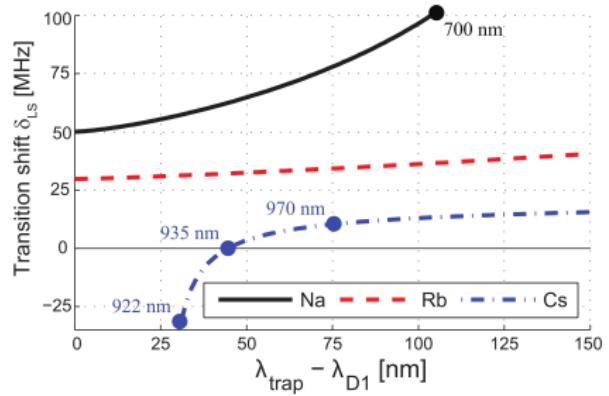
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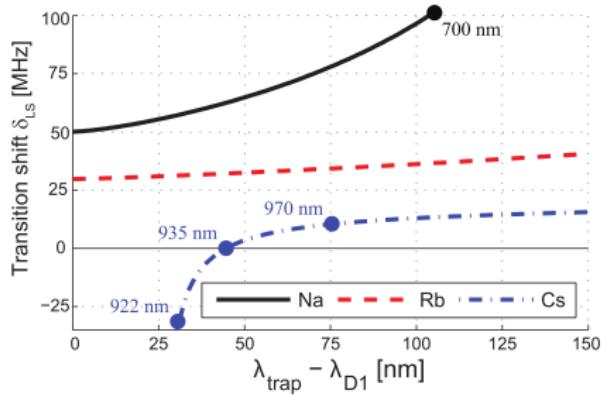
Issues with Na

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

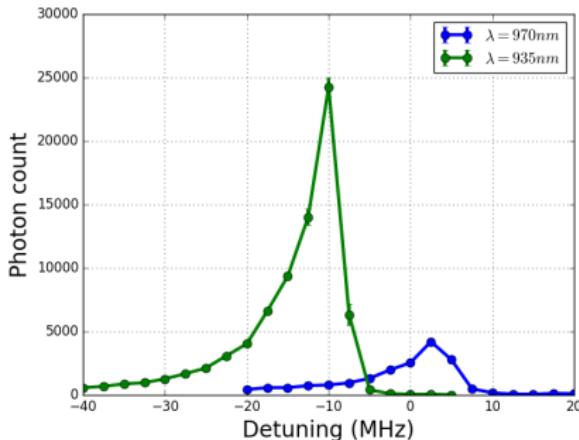
Real Issue with Na: Light Shift



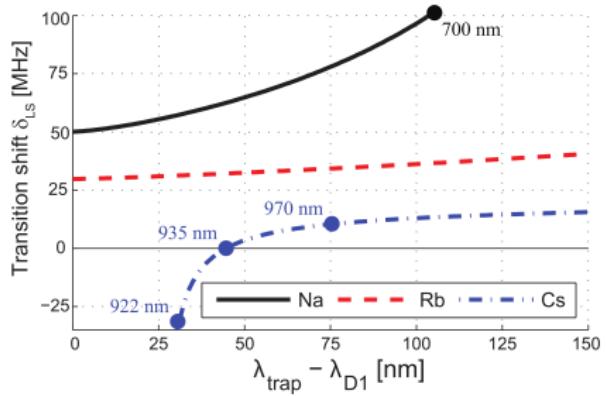
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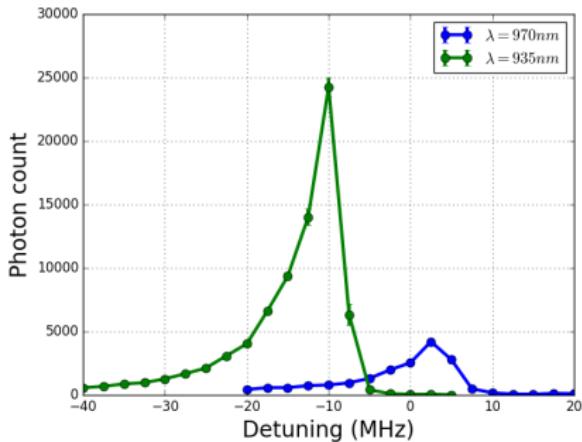
Cs single atom imaging



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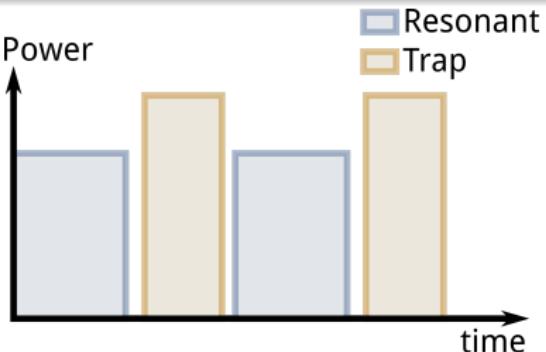
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Trap modulation

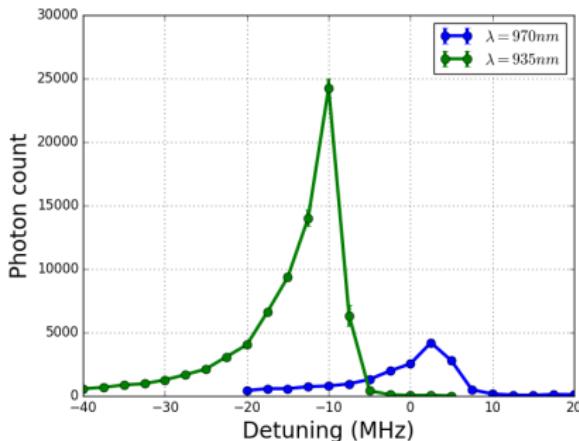
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



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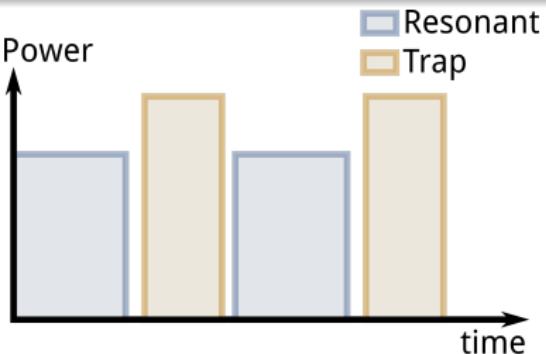
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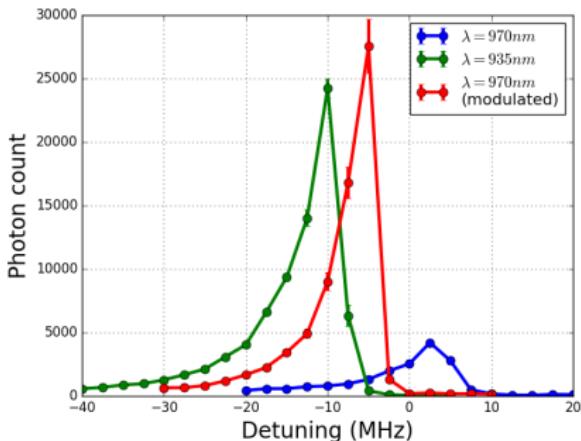
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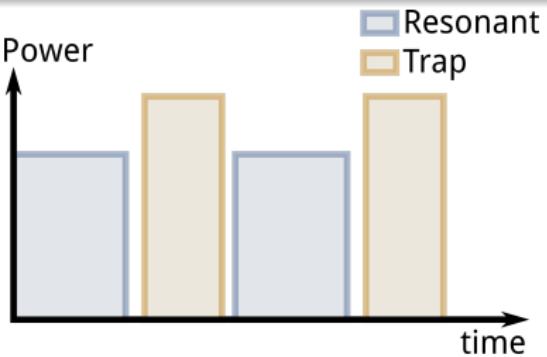
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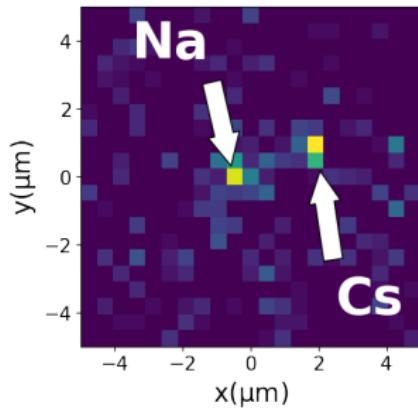
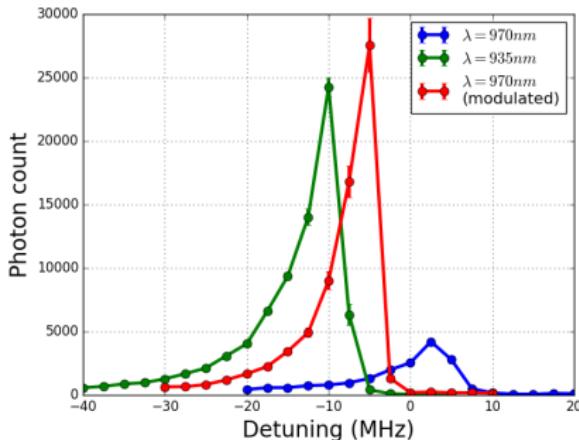
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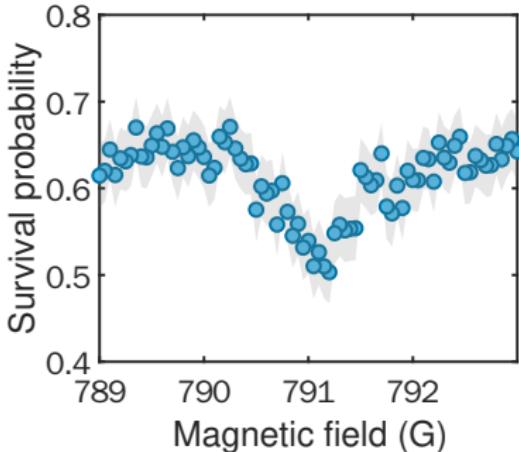
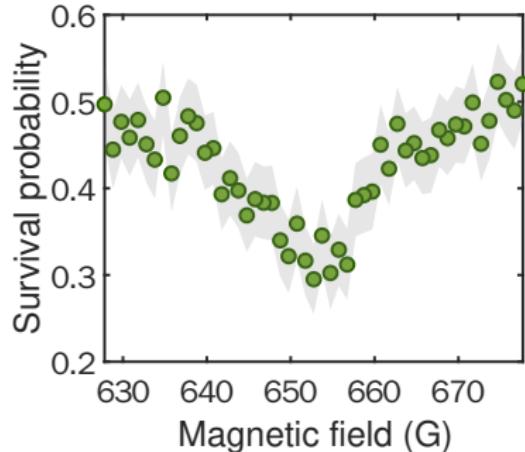
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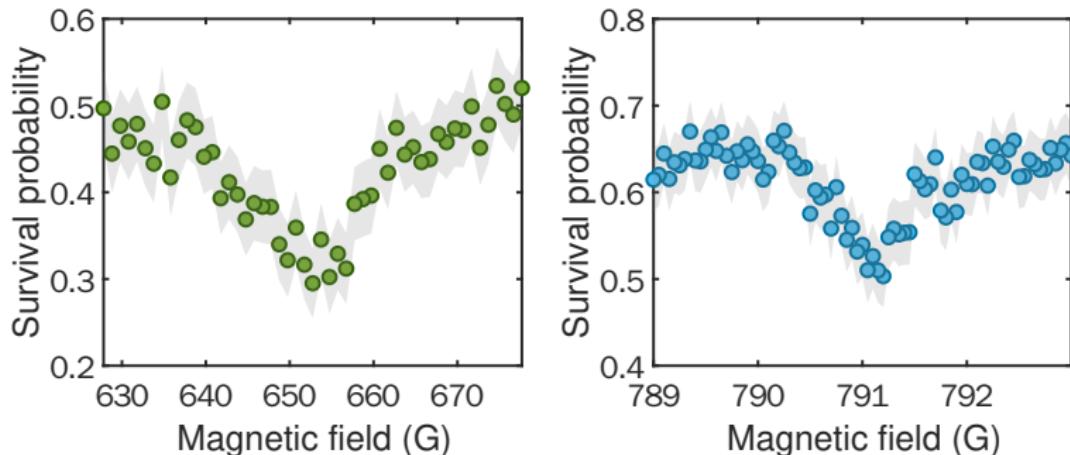
Na (1, -1) Cs (3, -3) Feshbach resonance



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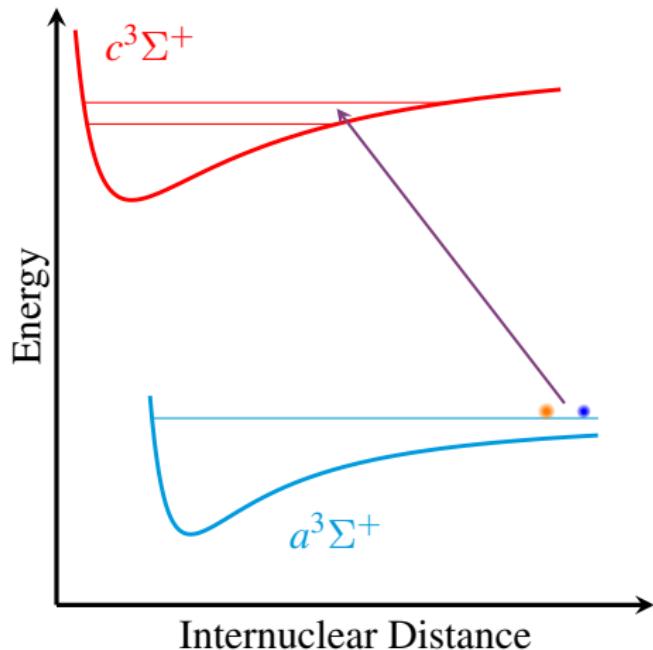
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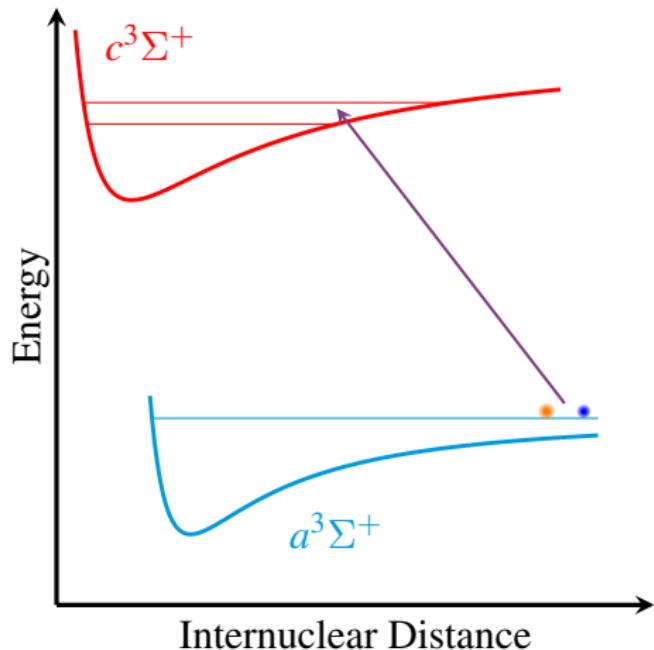
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



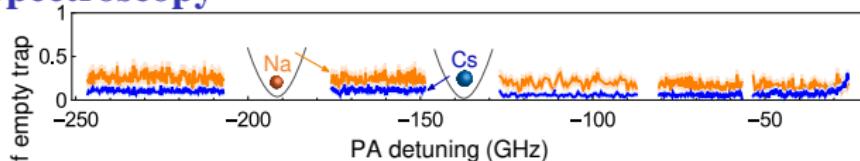
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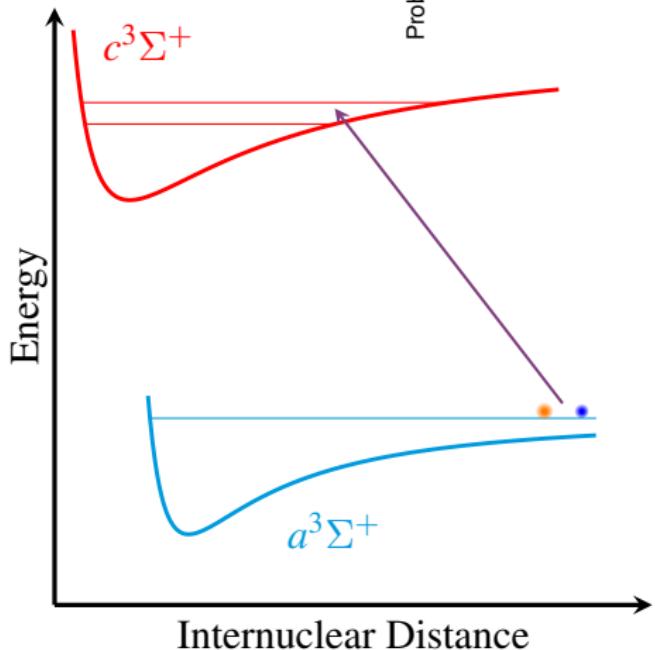
Single Atom PA

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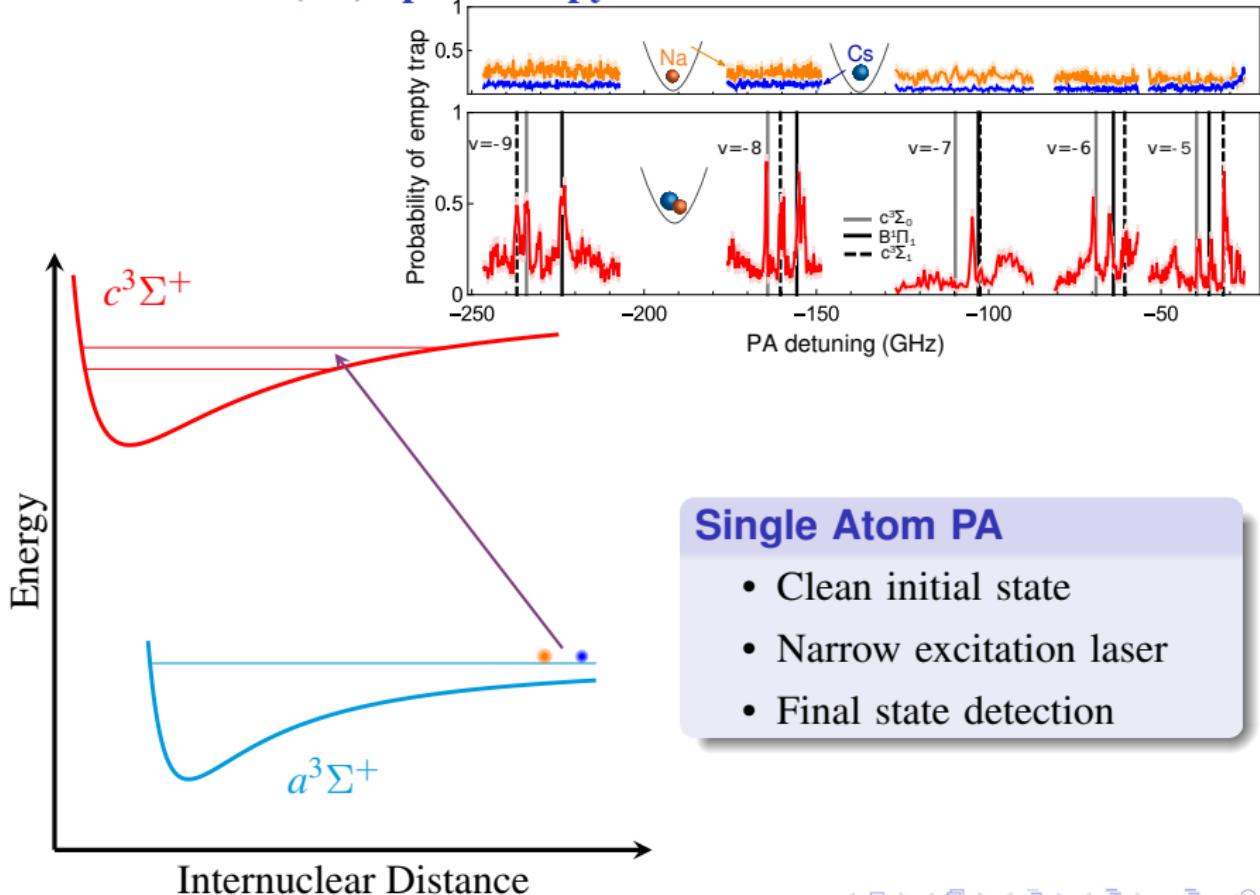
Probability of empty trap



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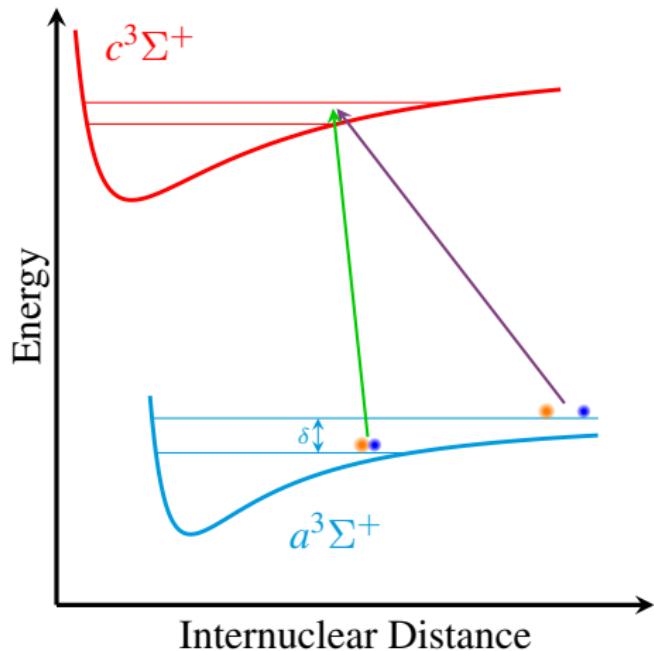
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Electromagnetically Induced Transparency (EIT) Spectroscopy



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