

# Coherent Creation of Single Molecules from Single Atoms

Yichao Yu

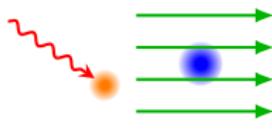
Ni Group/Harvard

Dec. 17, 2020

Simple System

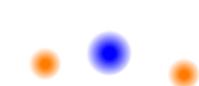


Full Control

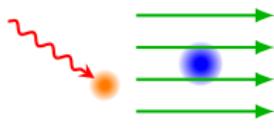


Complex Dynamics

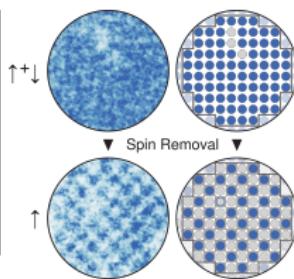
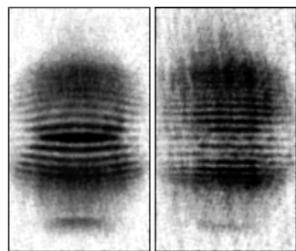
Simple System



Full Control



Complex Dynamics



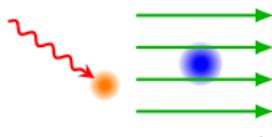
Science 275, 637 (1997)

Nature 545, 462-466 (2017)

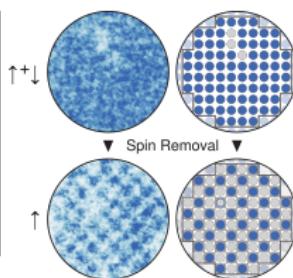
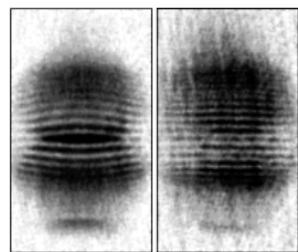
Simple System



Full Control



Complex Dynamics



Science 275, 637 (1997)

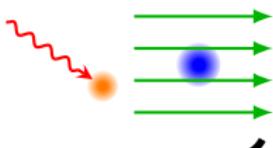
Nature 545, 462-466 (2017)

- ✗ Simple internal structure
- ✗ Weak interaction

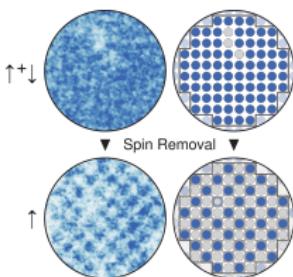
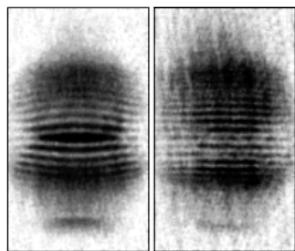
## Simple System



## Full Control



## Complex Dynamics



Science 275, 637 (1997)

Nature 545, 462-466 (2017)

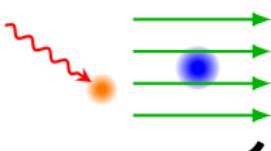
- Strong interaction
- Long coherence time
- Rich internal structure
- Fully controllable

- Simple internal structure
- Weak interaction

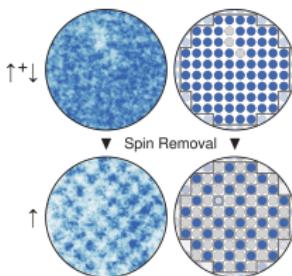
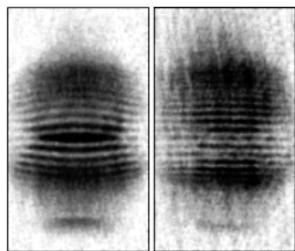
## Simple System



## Full Control



## Complex Dynamics



Science 275, 637 (1997)

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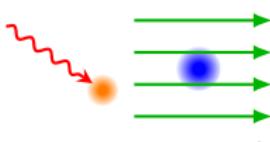
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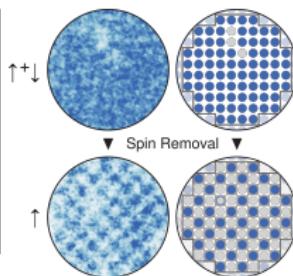
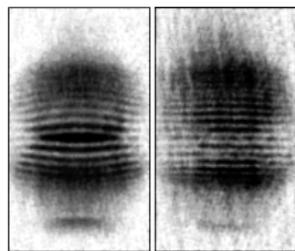
## Simple System



## Full Control



## Complex Dynamics



Science 275, 637 (1997)

Nature 545, 462-466 (2017)

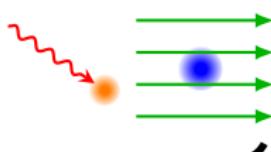
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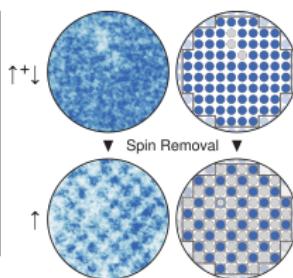
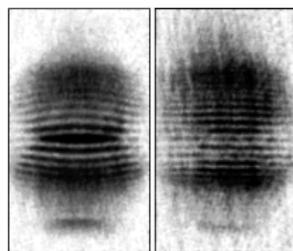
## Simple System



## Full Control



## Complex Dynamics



Science 275, 637 (1997)

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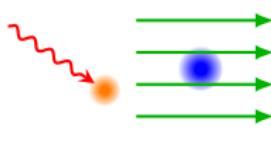
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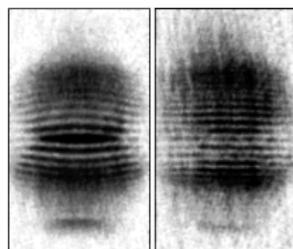
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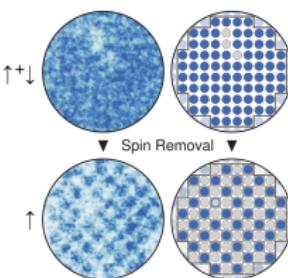
## Full Control



## Complex Dynamics

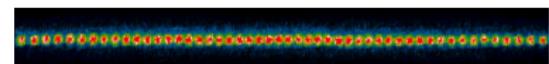


Science 275, 637 (1997)



Nature 545, 462-466 (2017)

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- Long coherence time
- Rich internal structure
- Fully controllable



Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

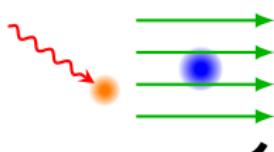
New System?

- Simple internal structure
- Weak interaction

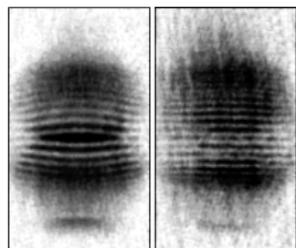
## Simple System



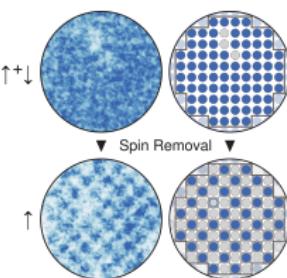
## Full Control



## Complex Dynamics

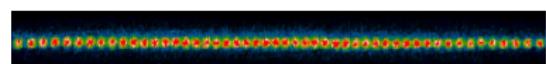


Science 275, 637 (1997)



Nature 545, 462-466 (2017)

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Ions (Photo credit: Monroe group)

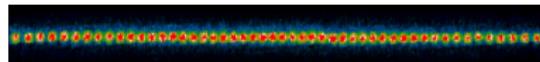


Rydberg Atoms (Photo credit: Lukin group)

## New System!

- Different properties
- New tools and techniques

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



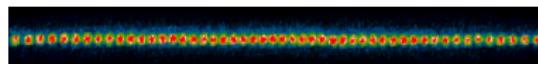
Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

✓ Strong interaction (kHz)

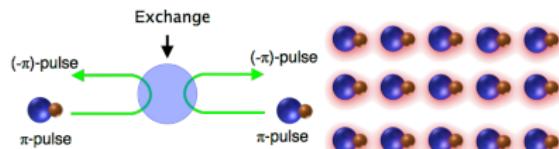
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Ions (Photo credit: Monroe group)

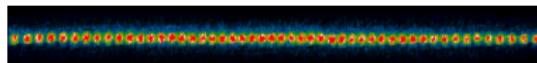


Rydberg Atoms (Photo credit: Lukin group)



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

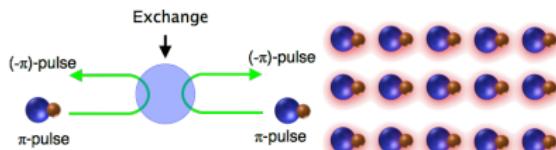
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Ions (Photo credit: Monroe group)

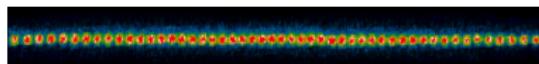


Rydberg Atoms (Photo credit: Lukin group)



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

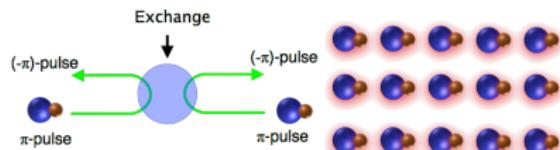
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Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

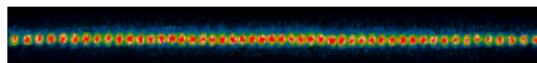


Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

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## Optical tweezers

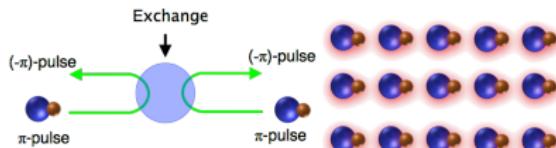
- Single site resolution
- . . .



Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

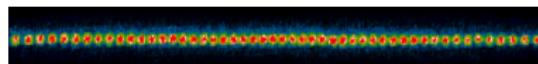


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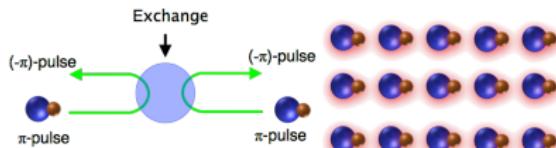
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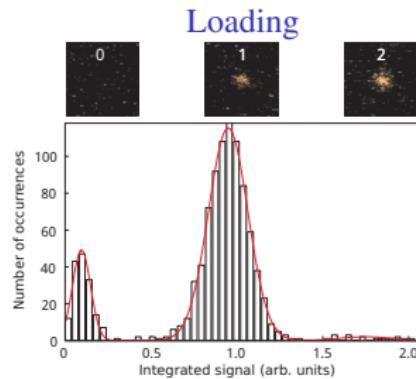


Rydberg Atoms (Photo credit: Lukin group)



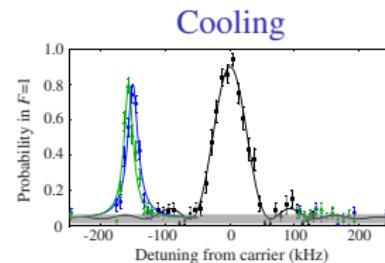
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Nat. Phys. 6, 951 (2010)

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



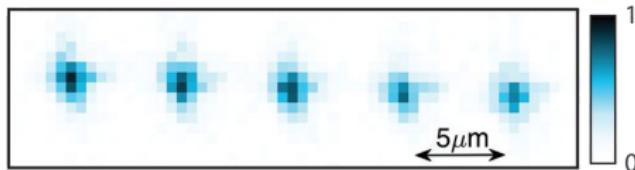
PRX. 2, 041014 (2012)



Science 354, 1024 (2016)

# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

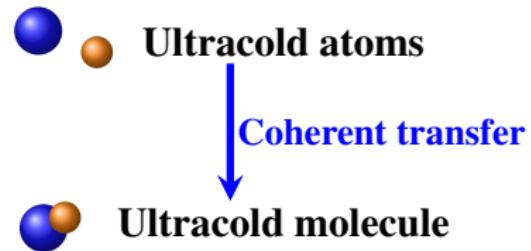
# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

## Assembly



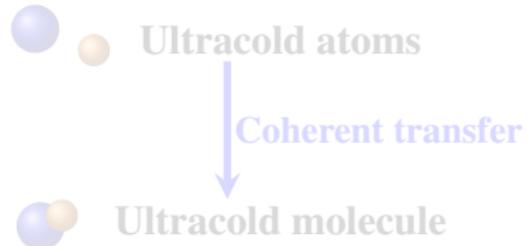
# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

## Assembly



## Challenges

- Temperature in tweezer
- Quantum control

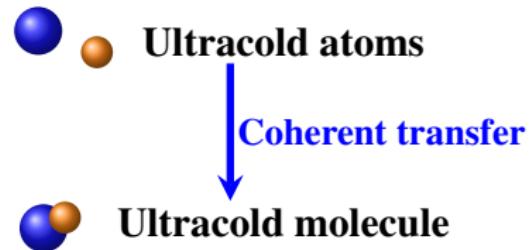
# Ultracold molecules in tweezers

## 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

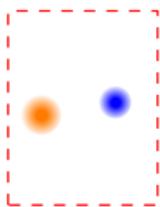
- Atom-atom interaction
- Coherent optical transfer

## 4 Conclusion

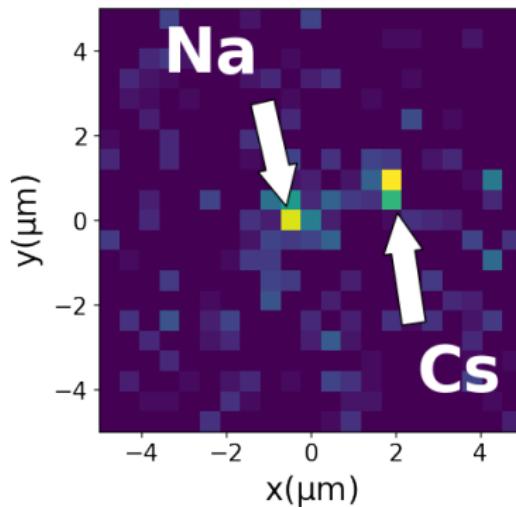
### NaCs molecule

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

## Experiment overview



## Loading

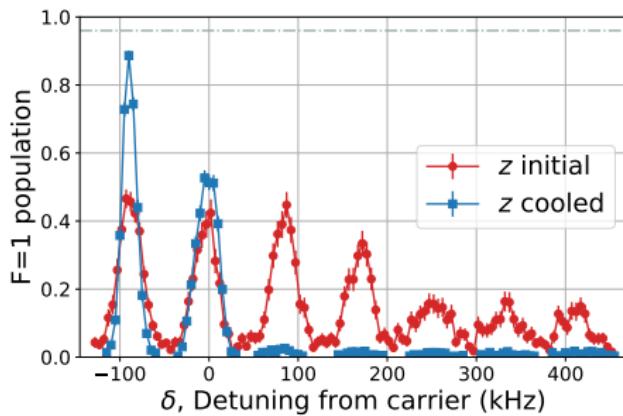


Loading probability per site: 60%  
Post select on initial and final state.

N. R. Hutzler, L. R. Liu, **Y. Yu** et al., New J. Phys. 19, 023007 (2017)  
**Y. Yu** et al., PRX 9, 021039 (2019)

# Experiment overview

## Cooling



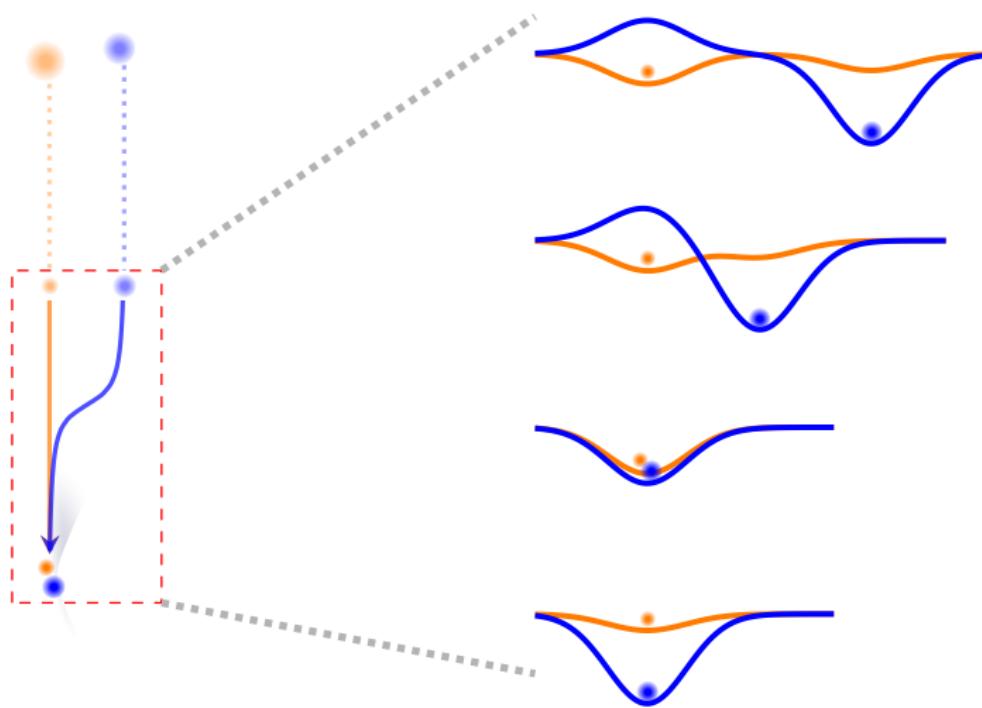
Cs: 96% ground state<sup>1</sup>  
Na: 94% ground state<sup>2</sup>

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

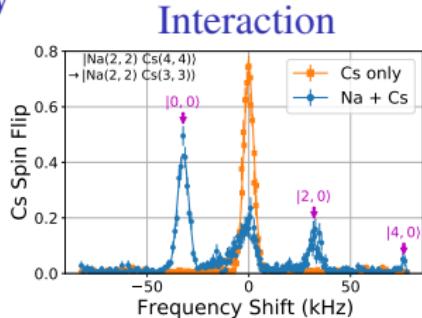
2: **Y. Yu et al.**, PRA 97, 063423 (2018)

## Experiment overview

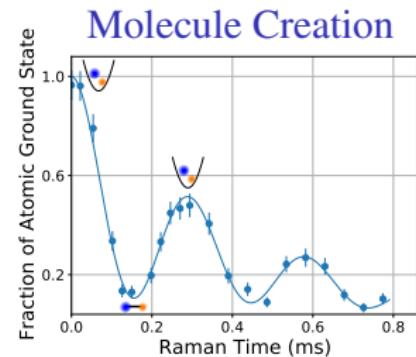
### Merging



# Experiment overview



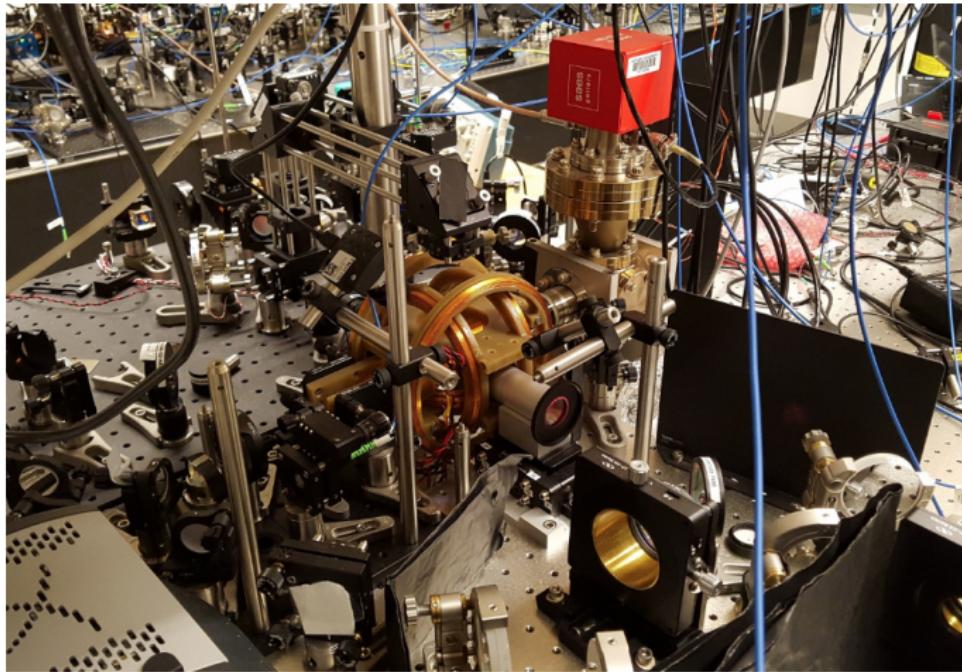
J. D. Hood, Y. Yu et al.,  
Phys. Rev. Research 2, 023108 (2020)

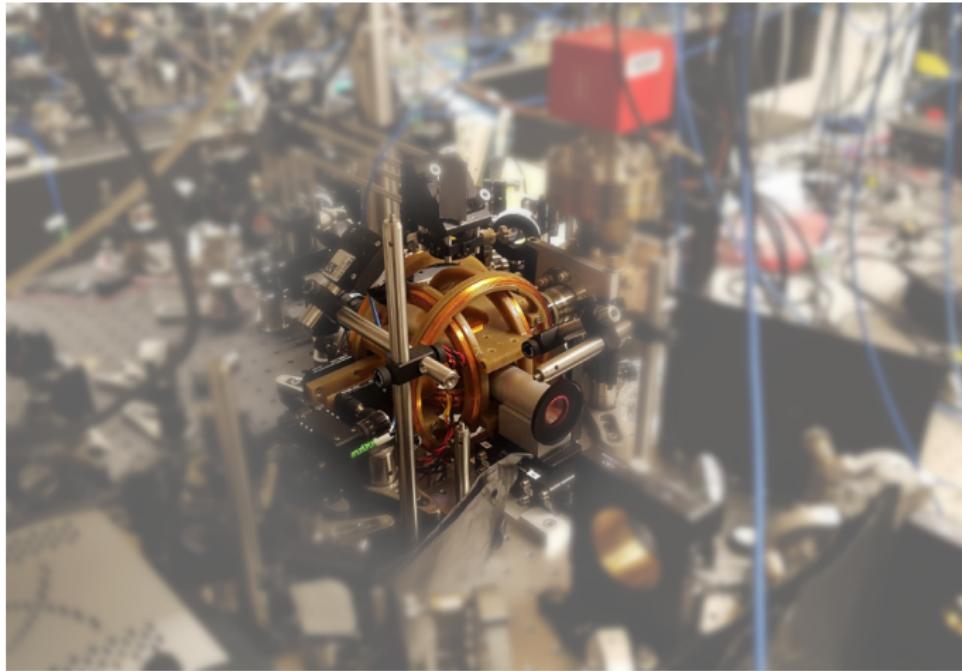


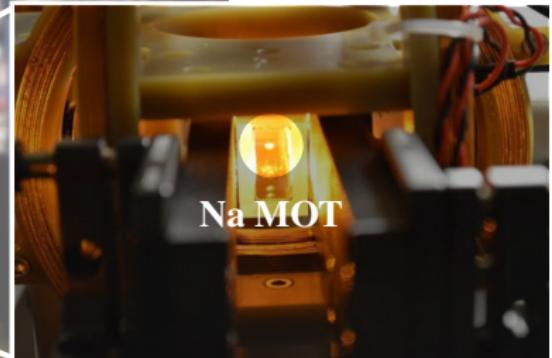
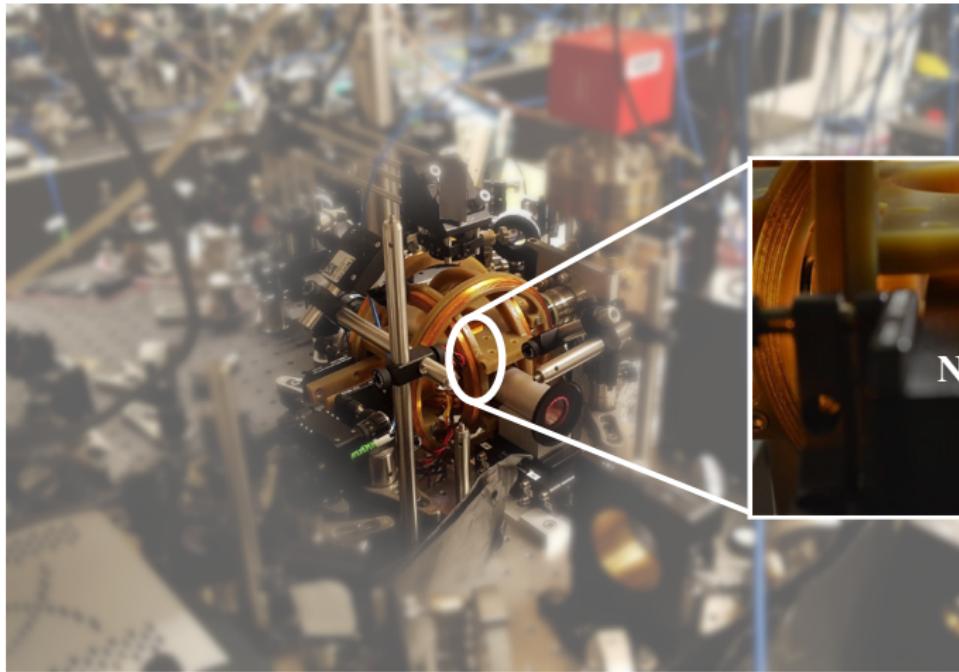
Y. Yu et al., arXiv:2012.09043 (2020)



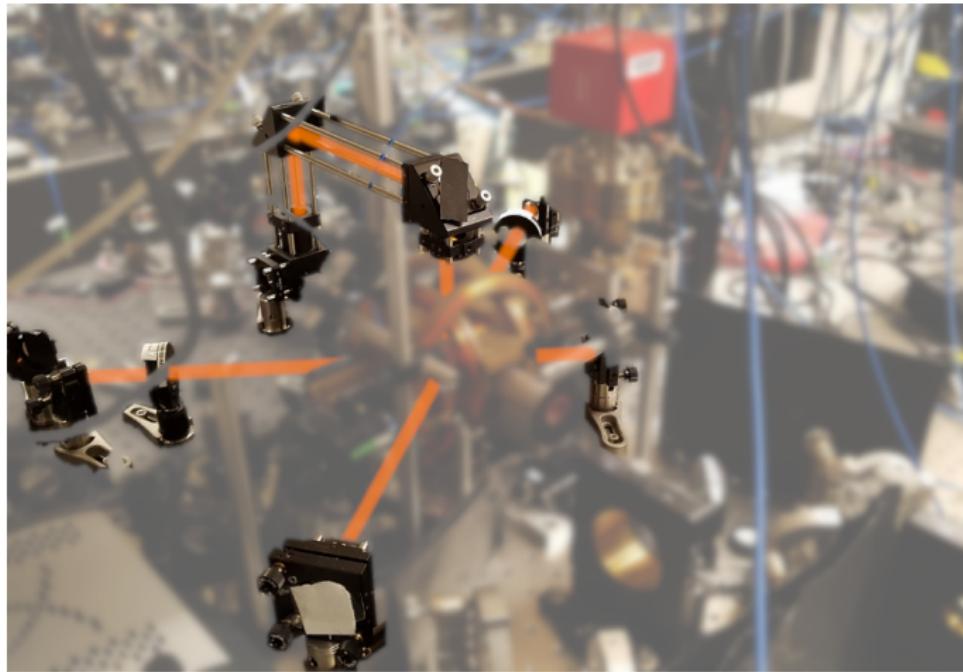
L. R. Liu, J. D. Hood, Y. Yu et al.,  
Science 360, 6391 (2018)



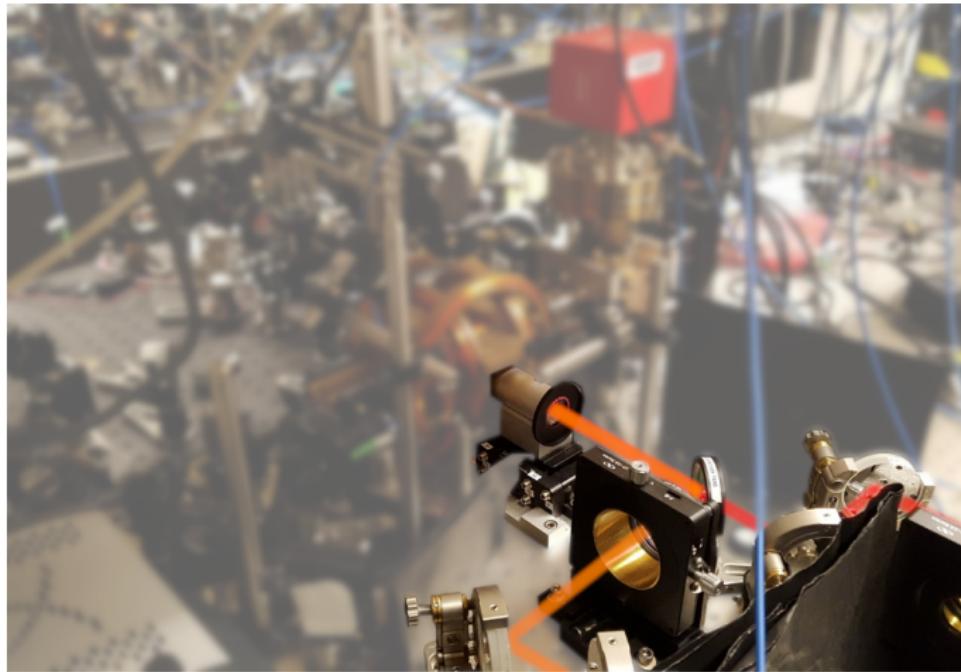




## MOT beam path



## Tweezer and imaging beam path



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- Coherent optical transfer

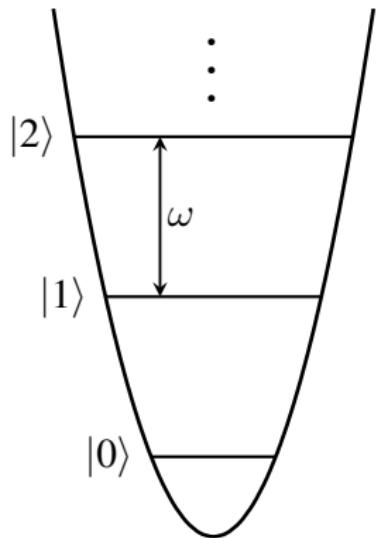
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## Raman sideband cooling

Used for cooling in trap

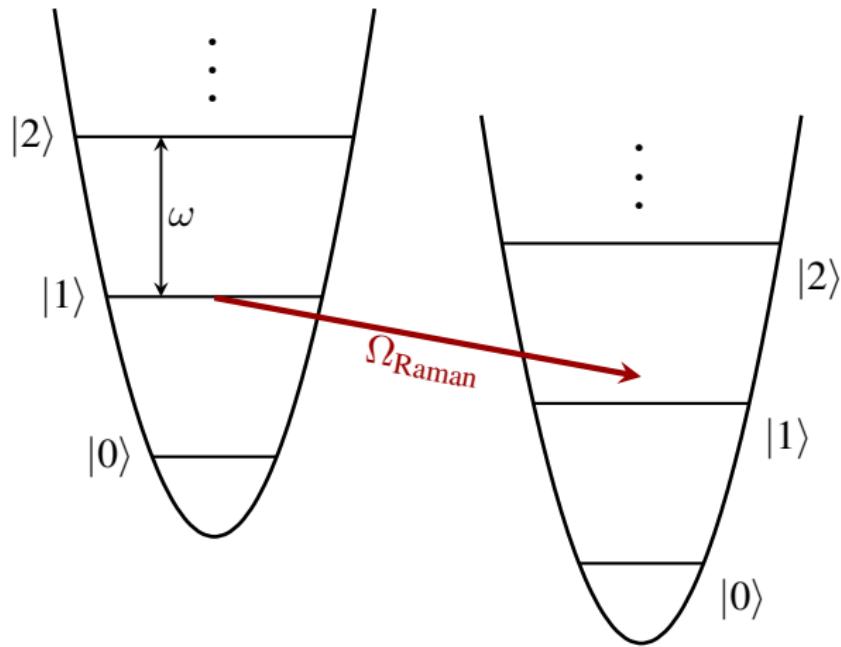
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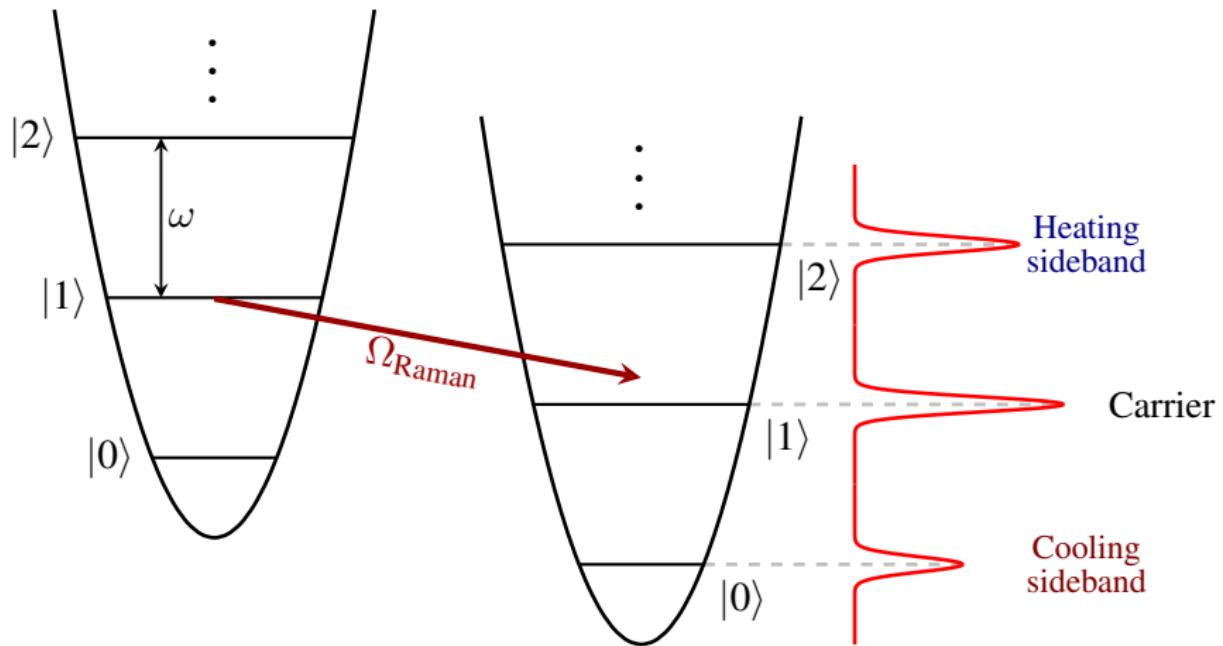
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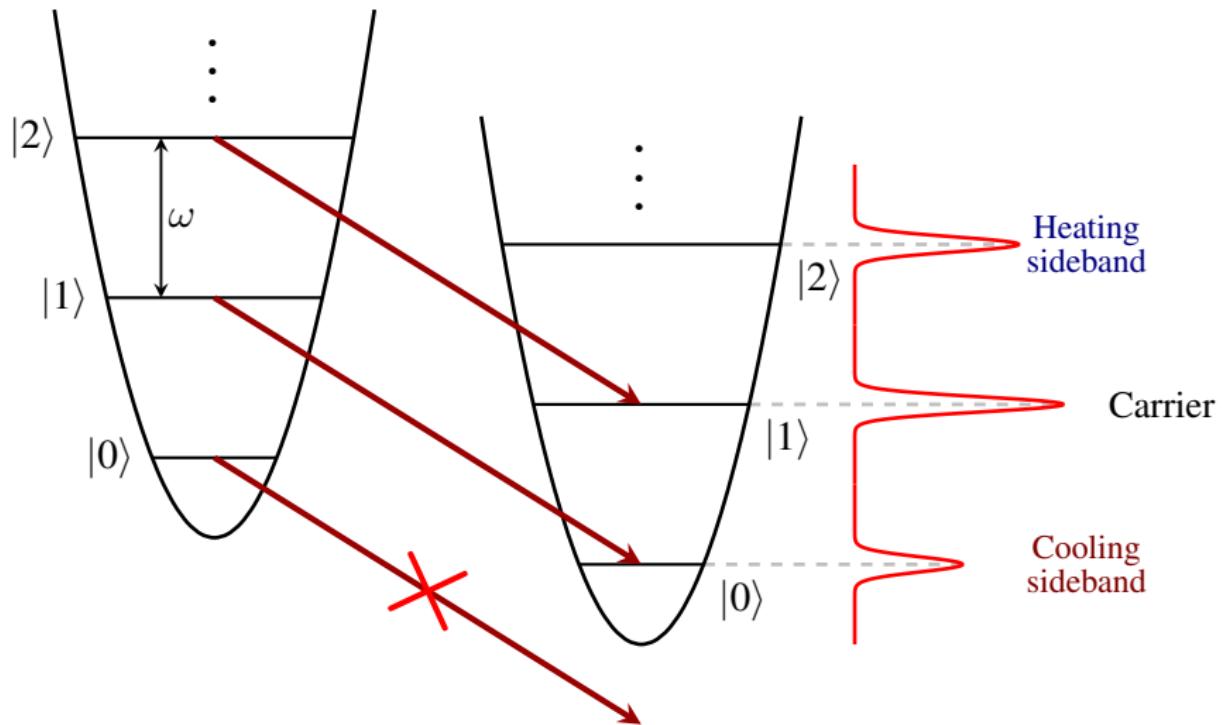
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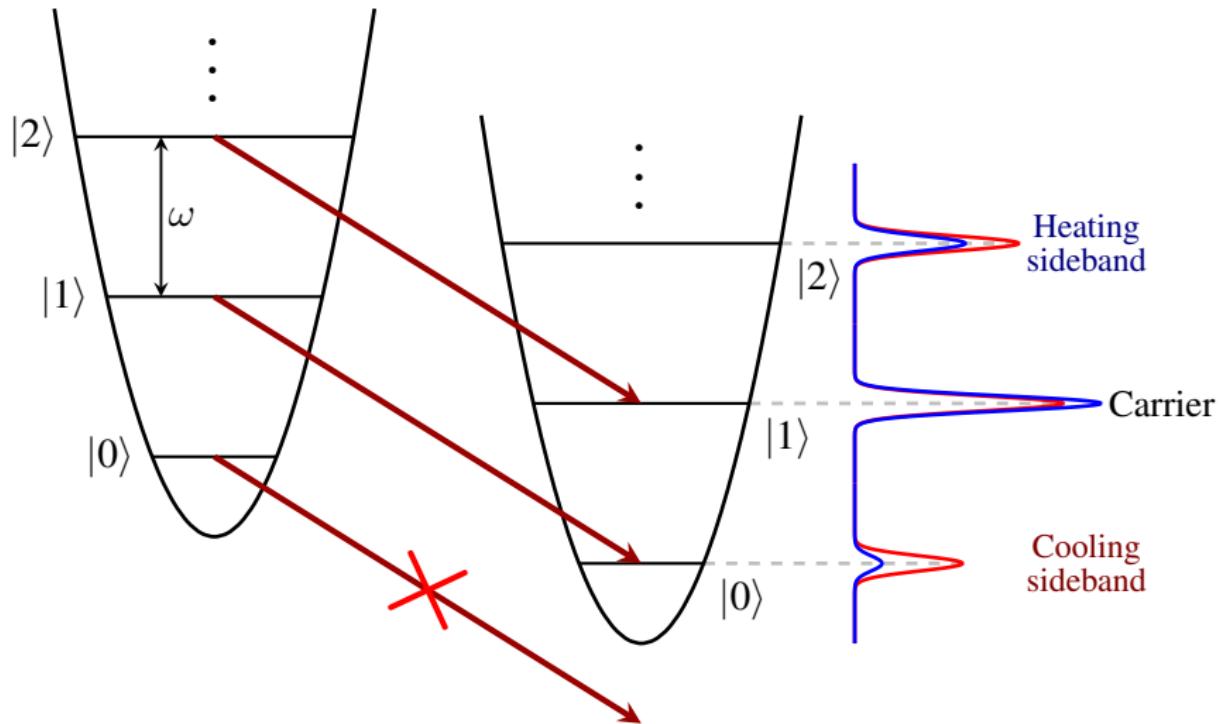
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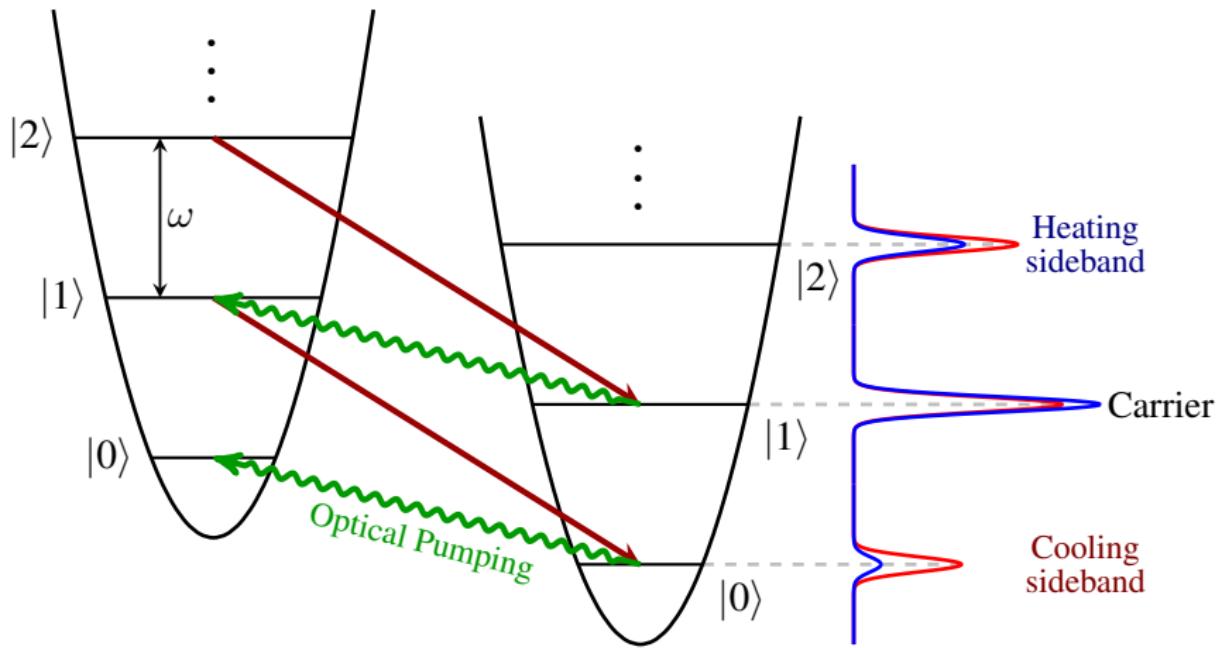
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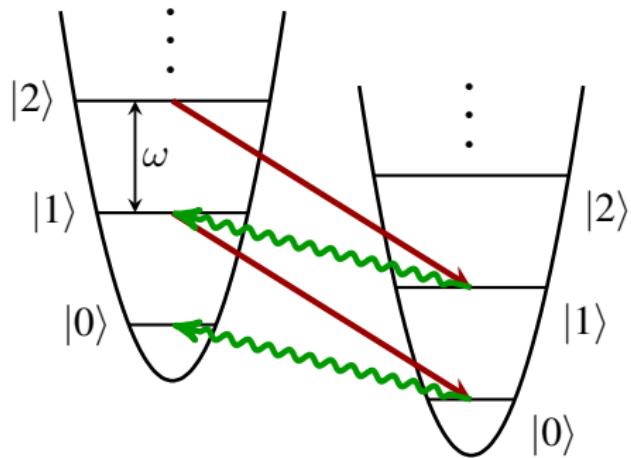


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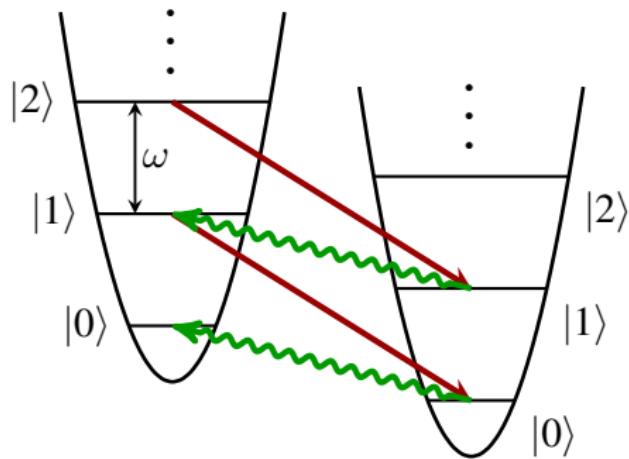
# Raman sideband cooling



## Raman sideband cooling

### Lamb Dicke parameter

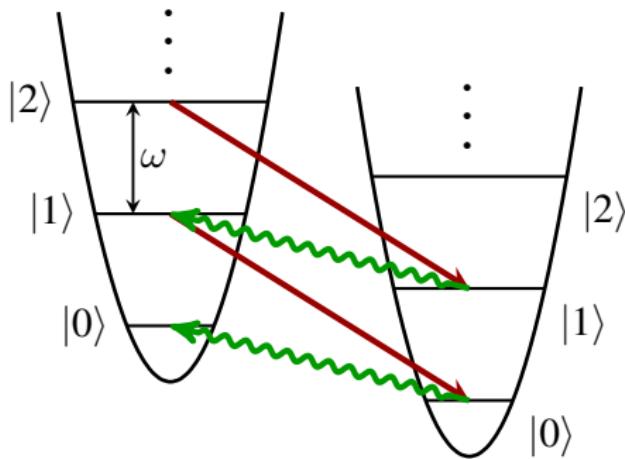
$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$



## Raman sideband cooling

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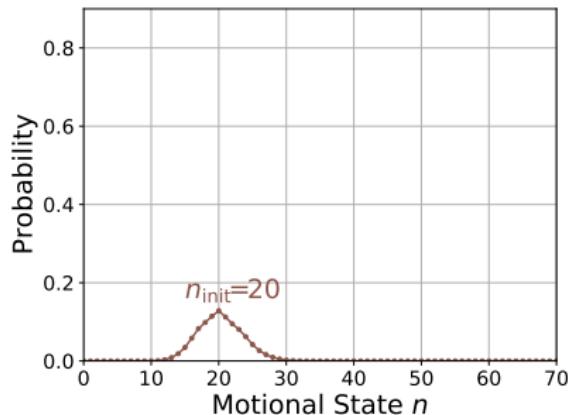
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$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \text{ } \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

## Raman sideband cooling



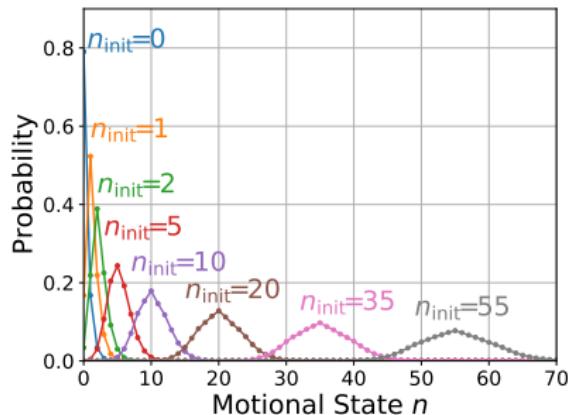
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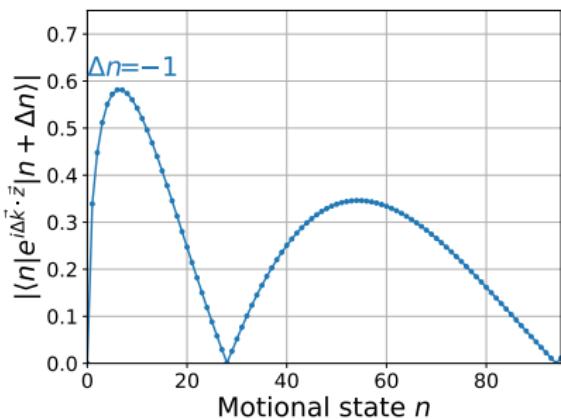
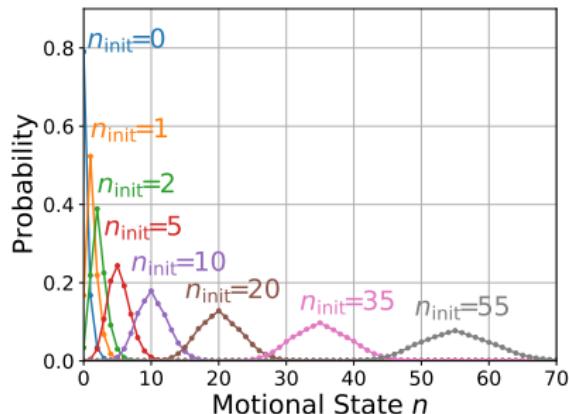
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$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$

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

- Motional state branching
- Coupling “dead zone”

# Raman sideband cooling



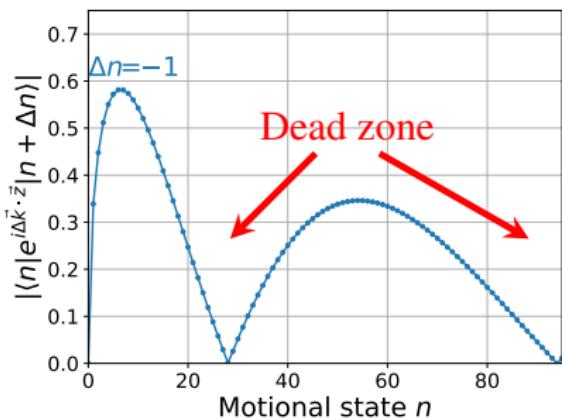
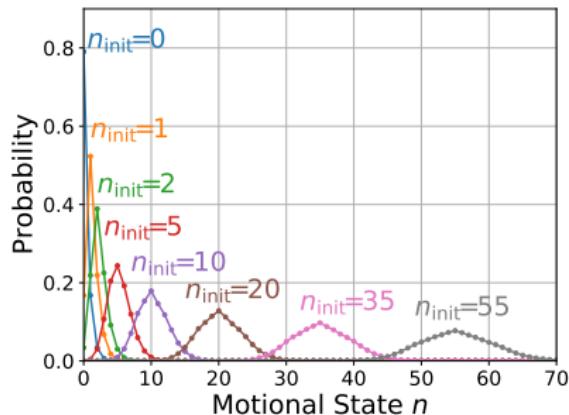
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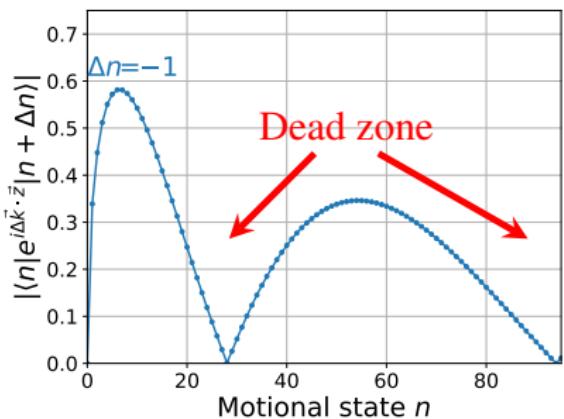
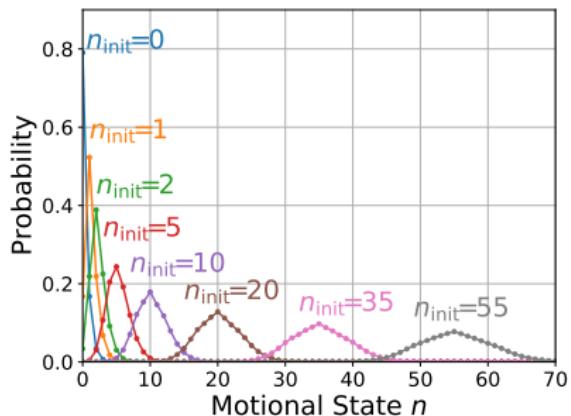
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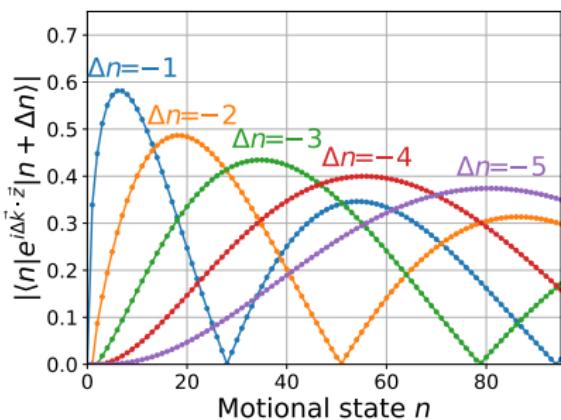
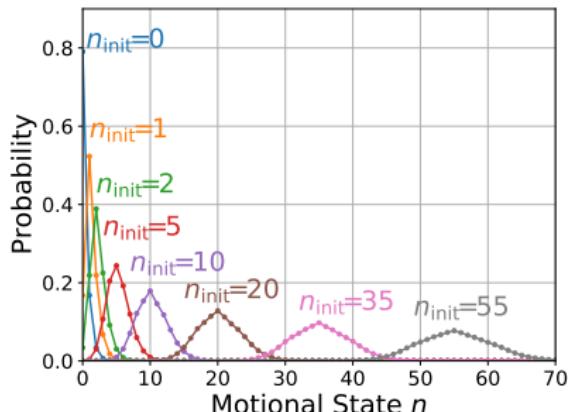
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## Solution

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

# Raman sideband cooling



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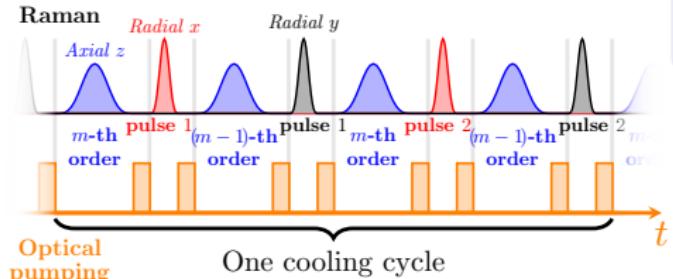
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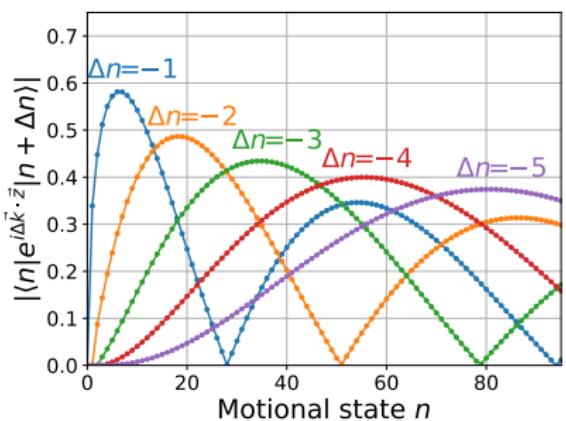


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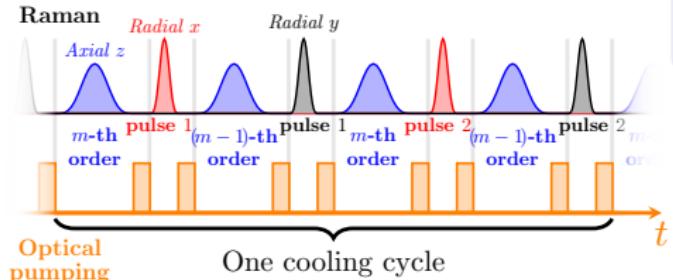
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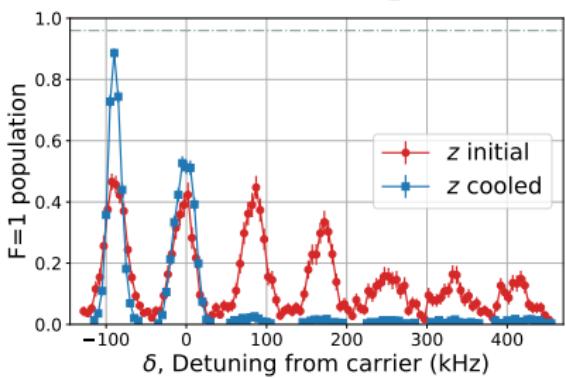
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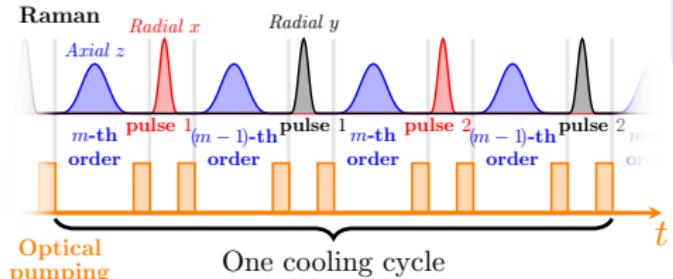
Axial sideband spectrum



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# Raman sideband cooling



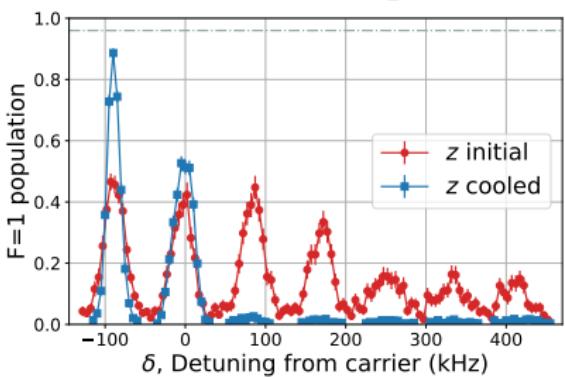
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Axial sideband spectrum



## Solution

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3D ground state: 93.5(7)%

PRA 97, 063423 (2018)

# Outline

## 1 Experiment overview

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- Raman sideband cooling of Na atoms

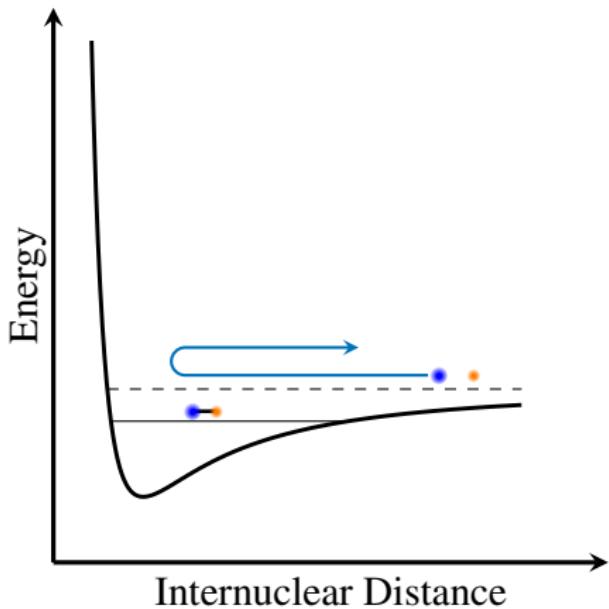
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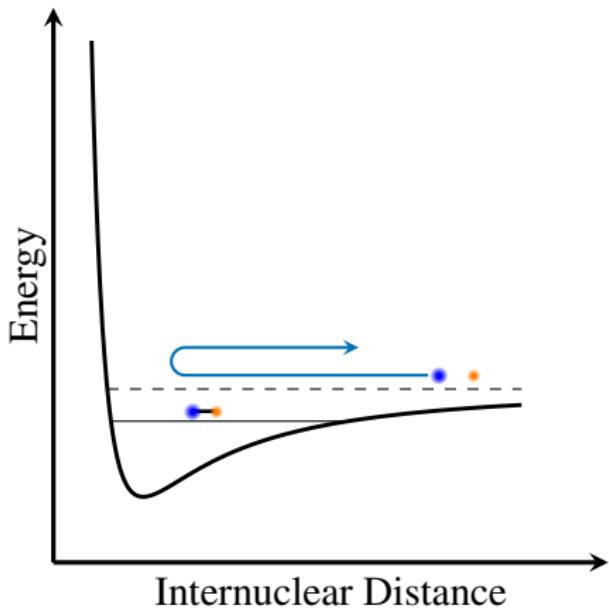
## Scattering length $a$

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



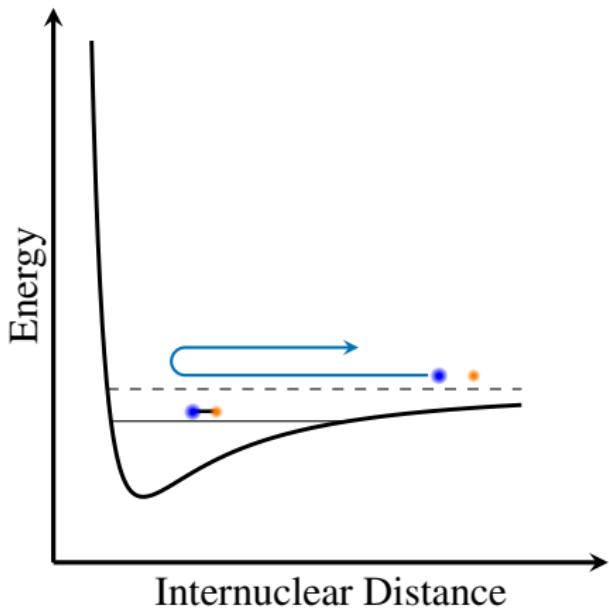
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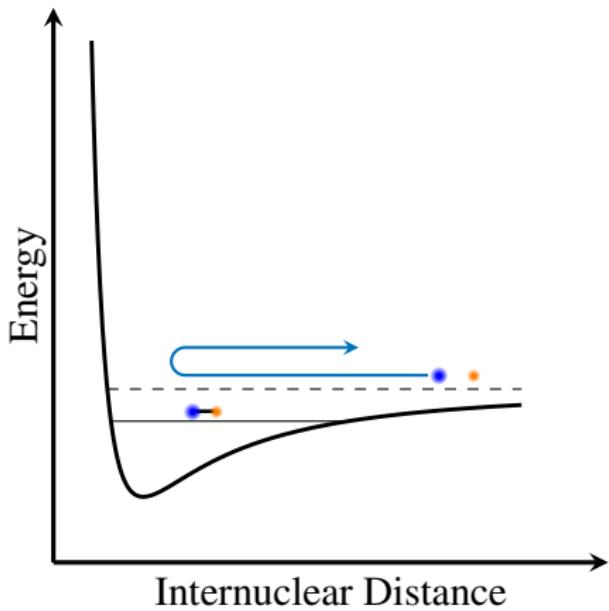
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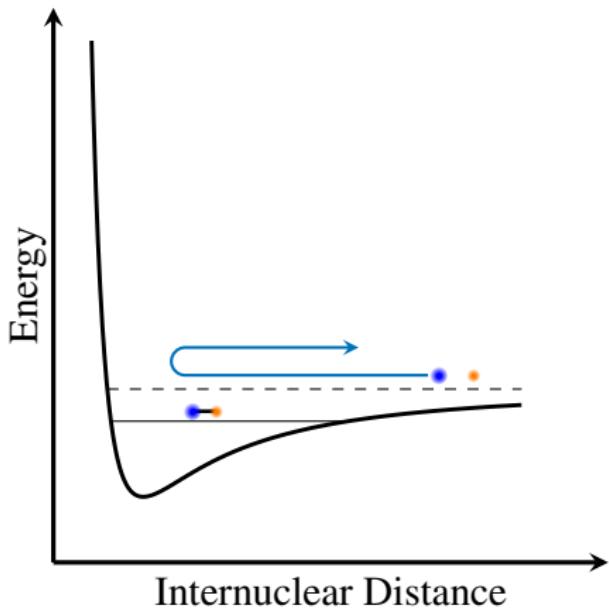
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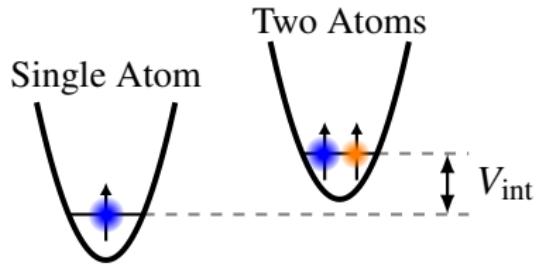


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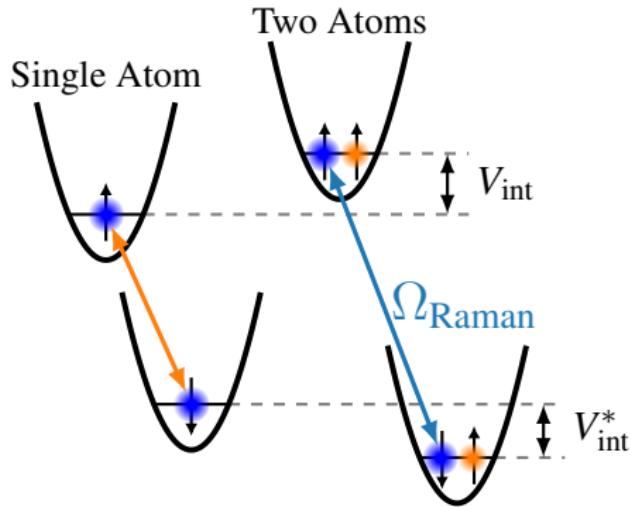
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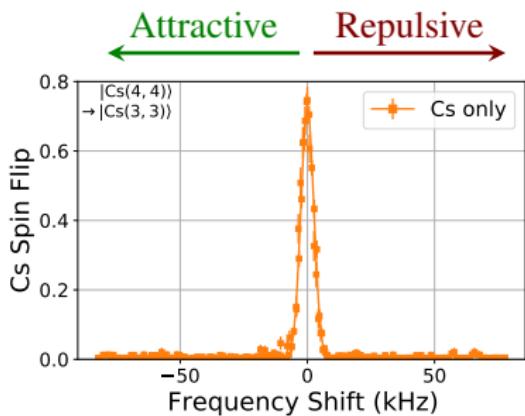
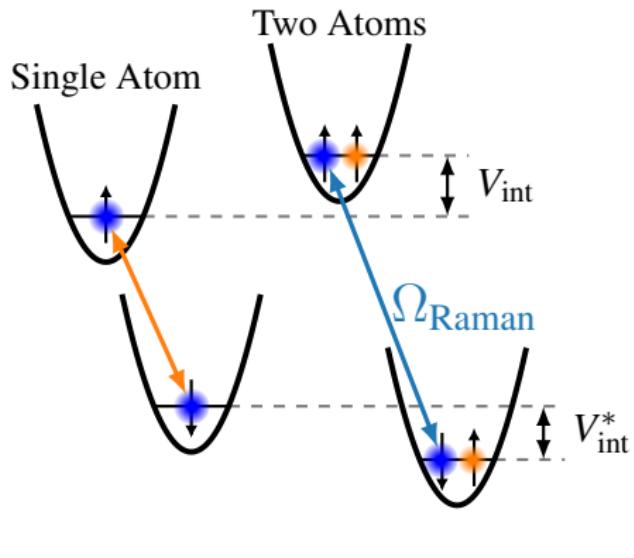
# Interaction shift



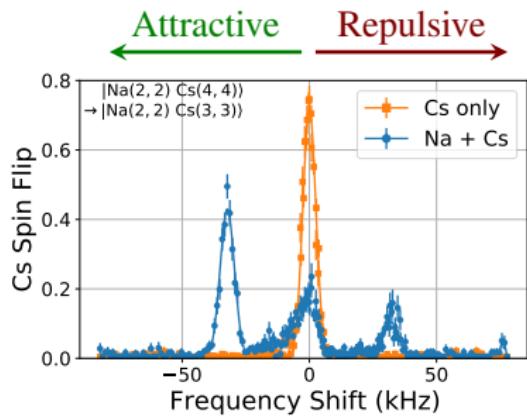
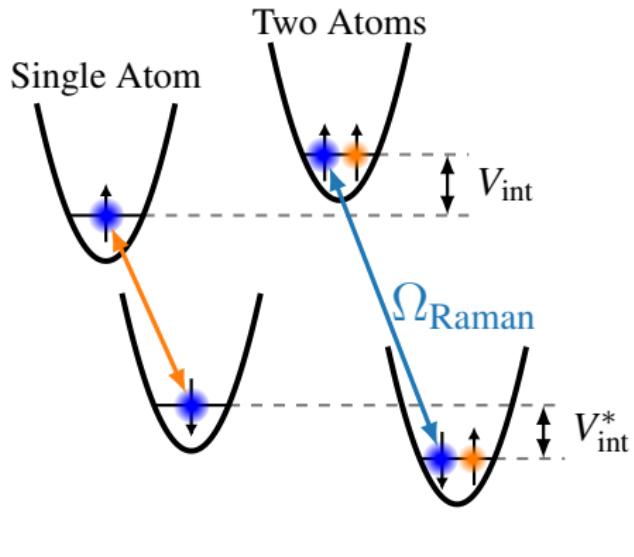
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## Interaction shift



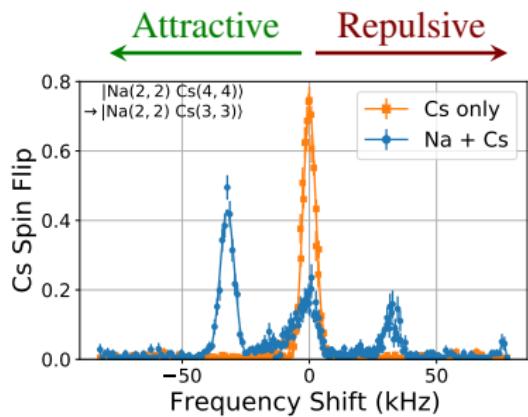
# Interaction shift



## Interaction shift

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

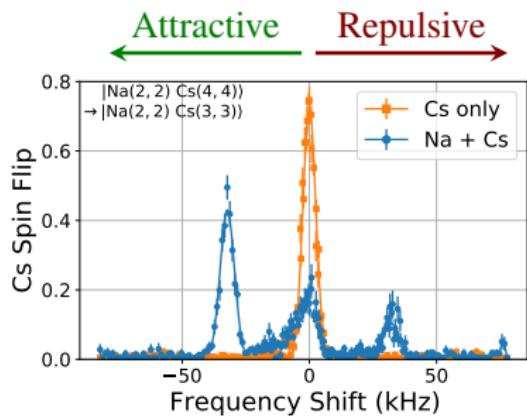
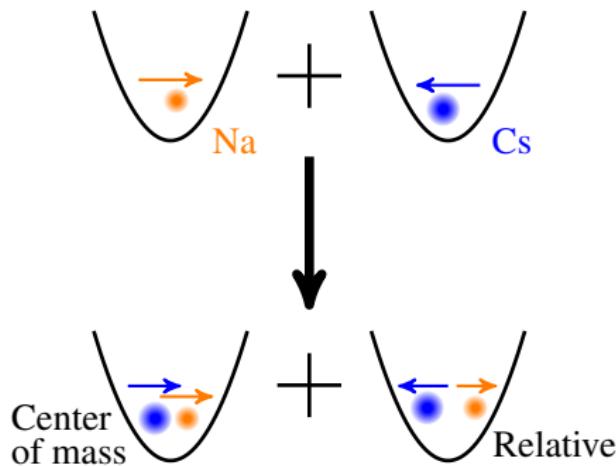
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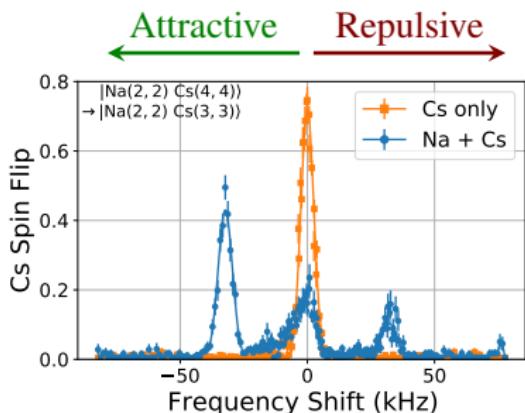
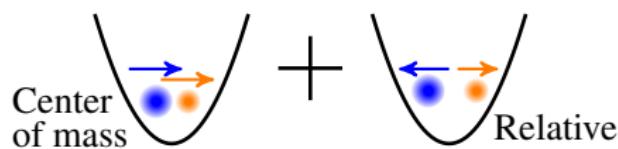
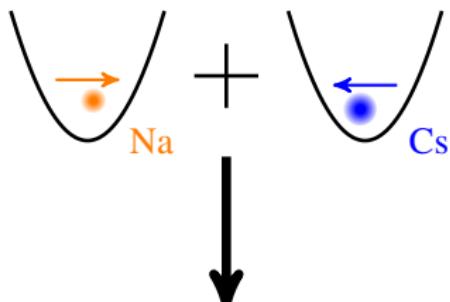
Interaction



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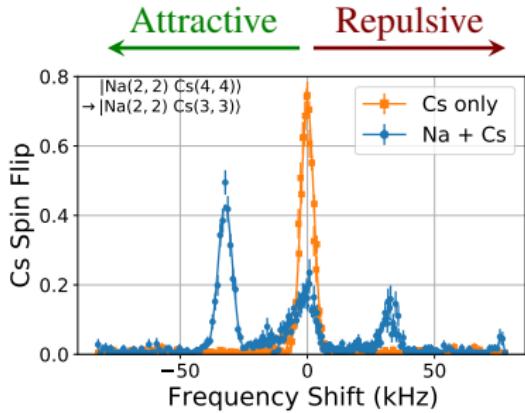
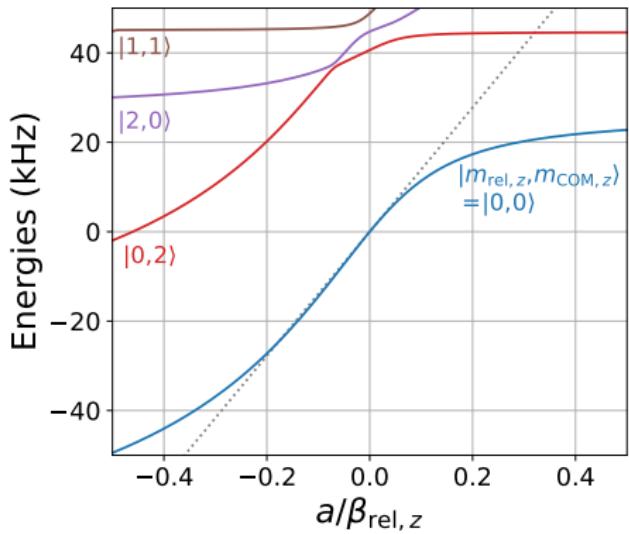
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Interaction



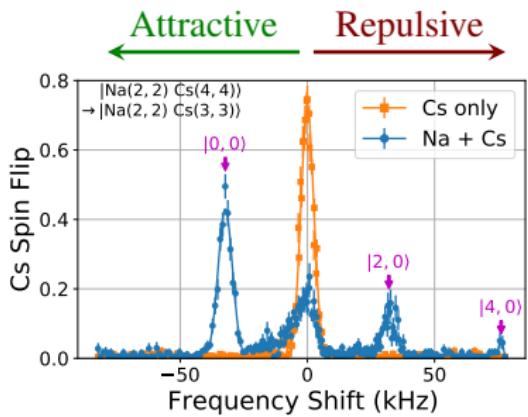
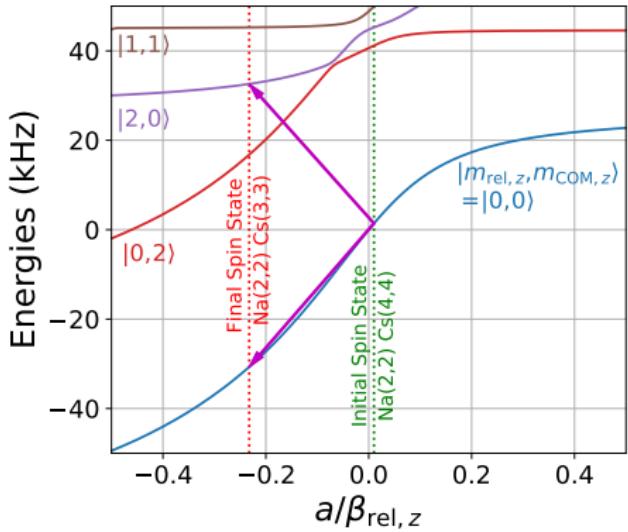
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M \Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu \omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{\text{int}}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z}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

# Interaction shift



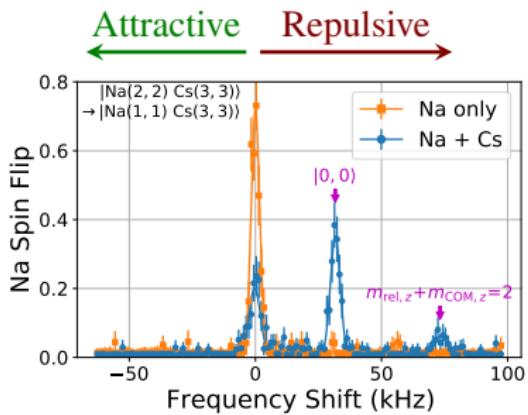
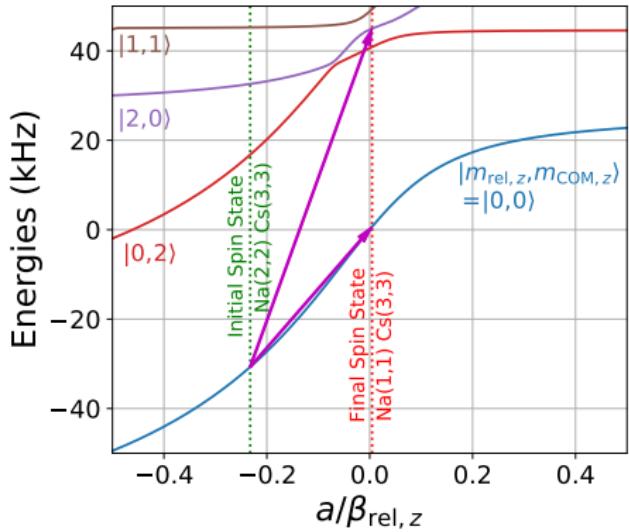
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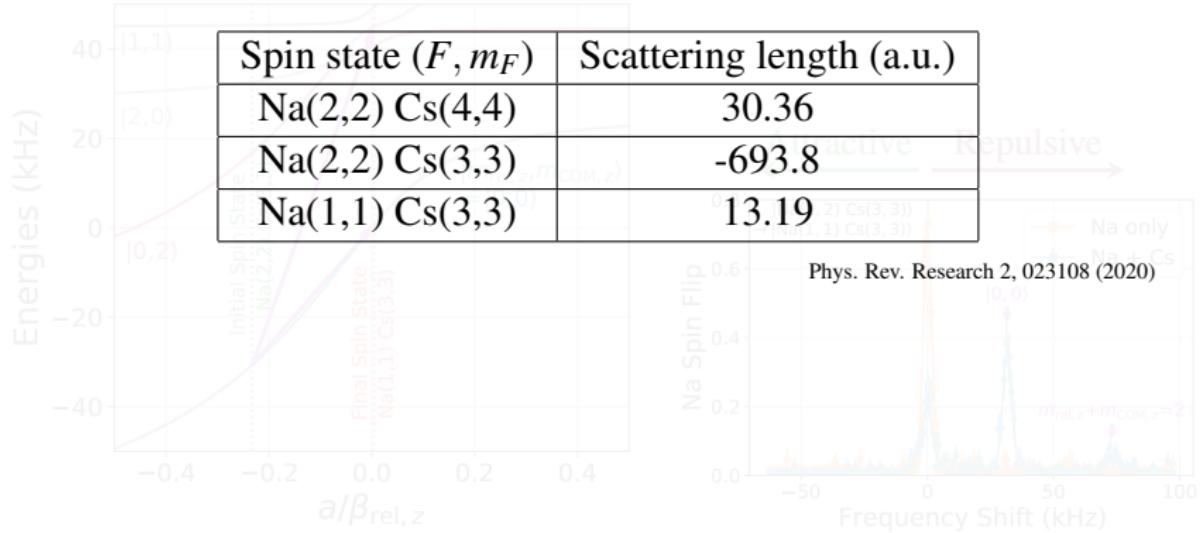
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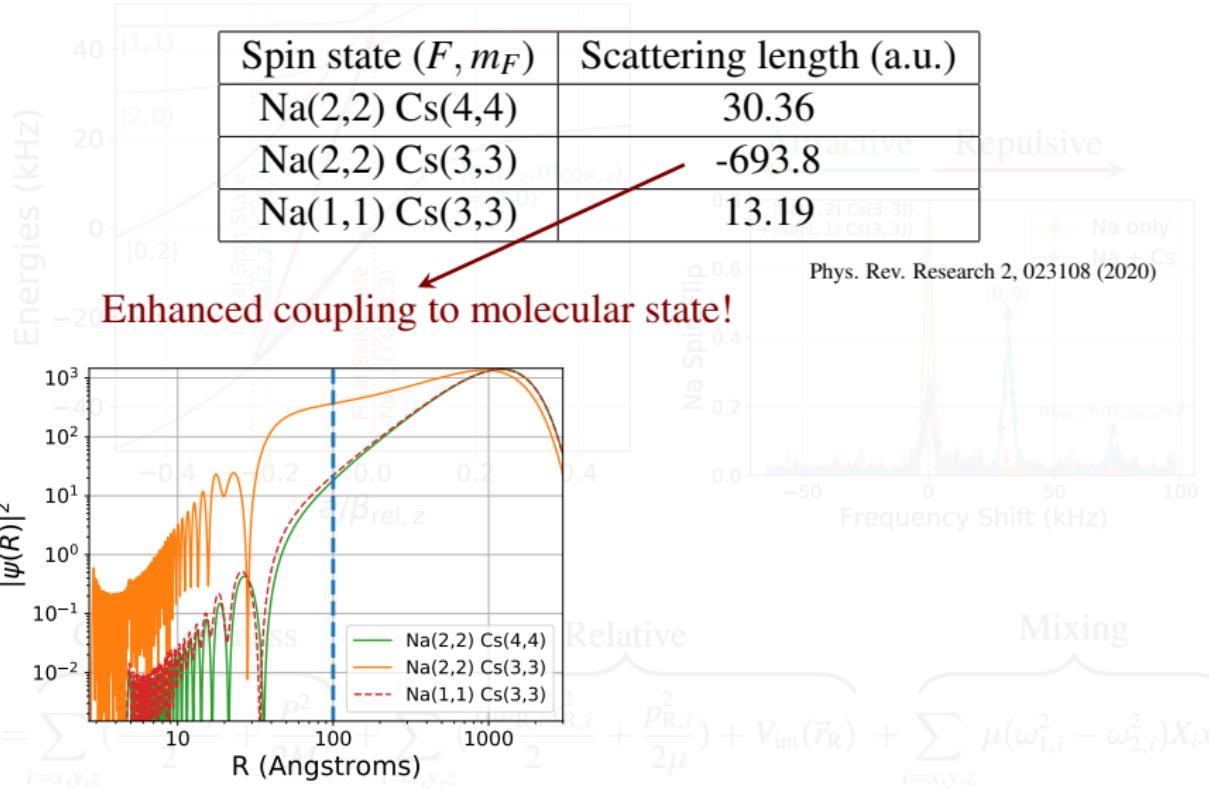
Combined with binding energy measurement on Na(2,2) Cs(4,4)



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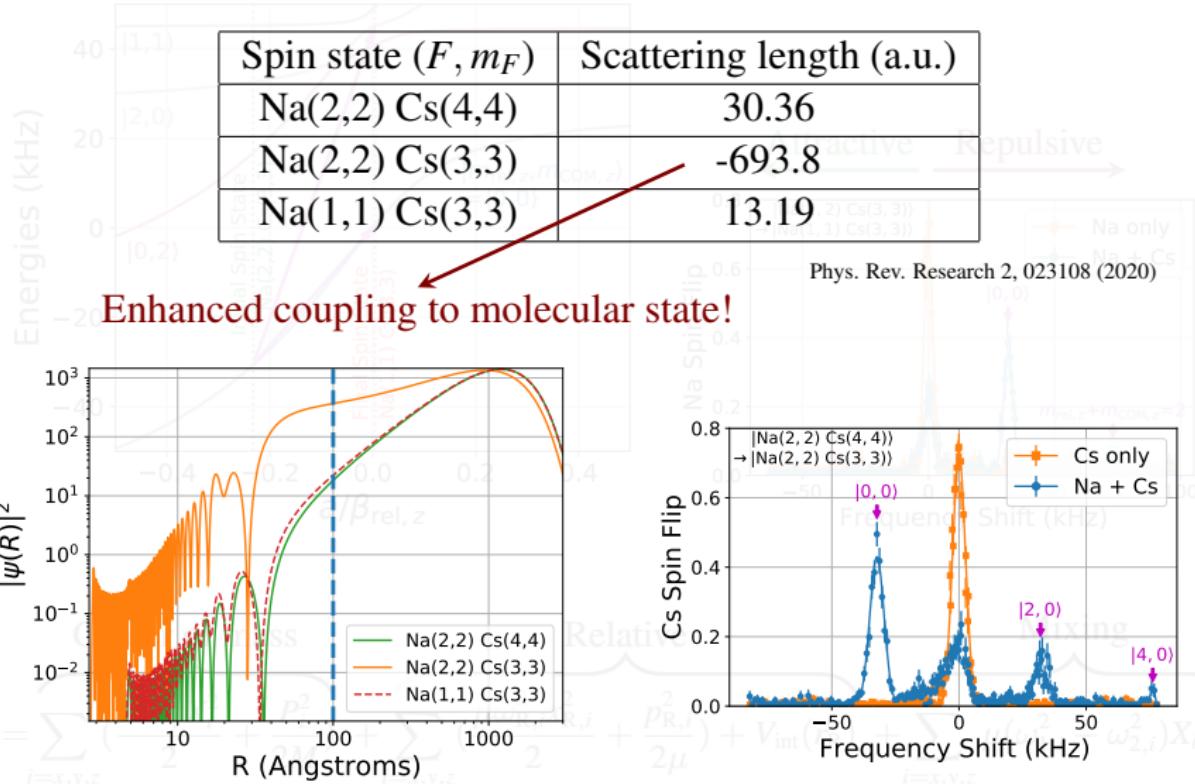
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Combined with binding energy measurement on Na(2,2) Cs(4,4)



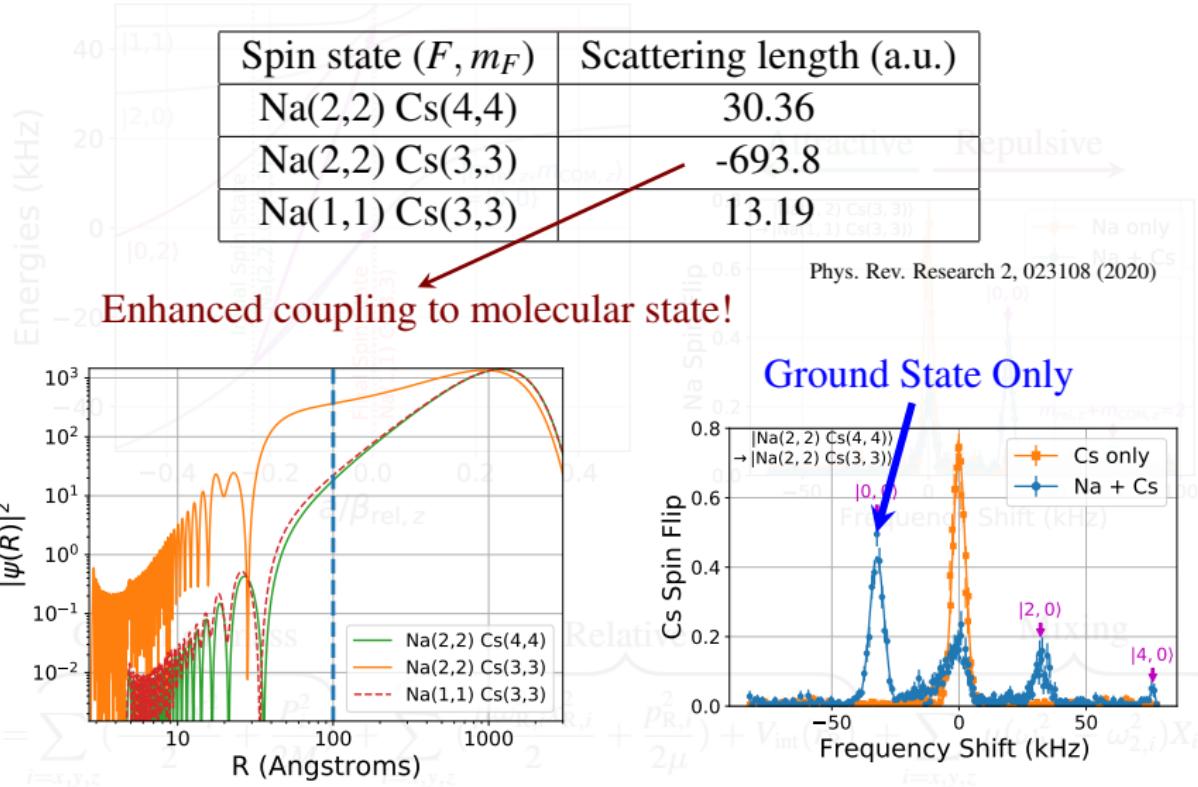
# Interaction shift

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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

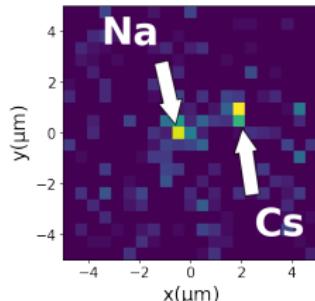
- Raman sideband cooling of Na atoms

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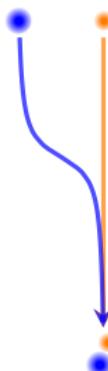
## 4 Conclusion

## Loading

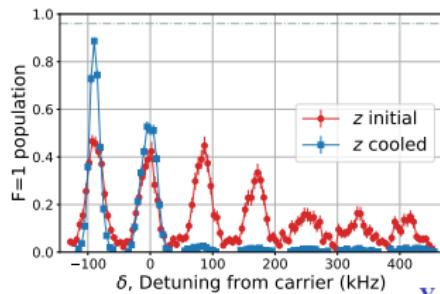


N. R. Hutzler, L. R. Liu, Y. Yu et al.,  
New J. Phys. 19, 023007 (2017)

## Merging

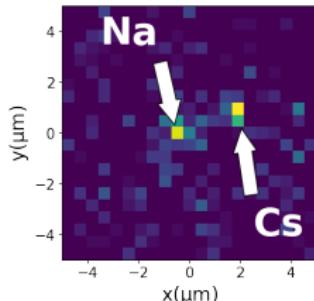


## Cooling



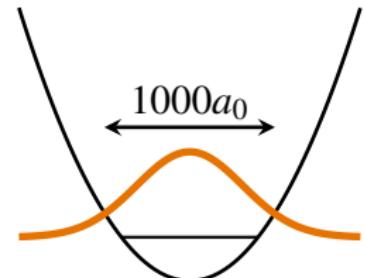
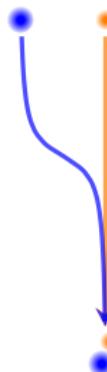
Y. Yu et al., PRX. 9, 021039 (2019)  
Y. Yu et al., PRA. 97, 063423 (2018)

## Loading

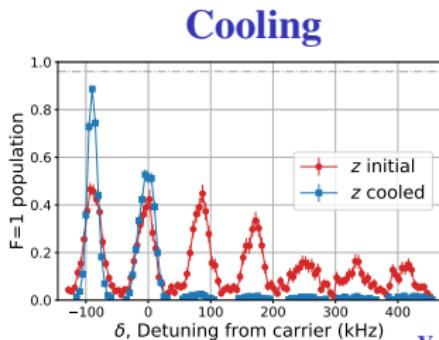


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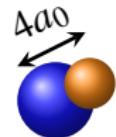


## Atom

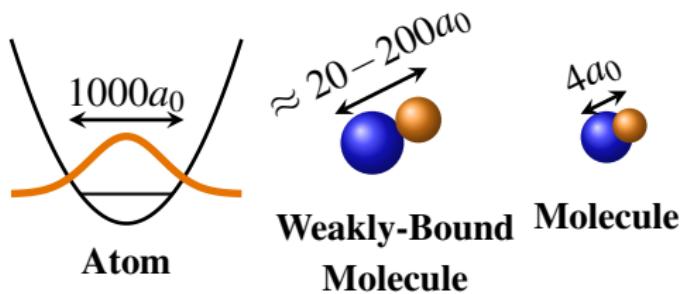


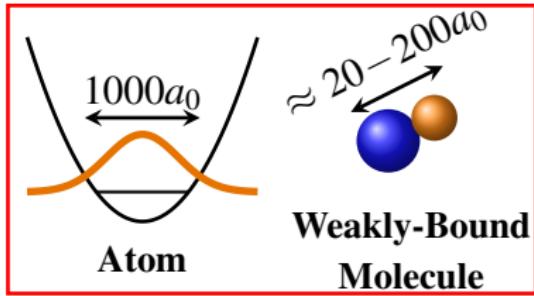
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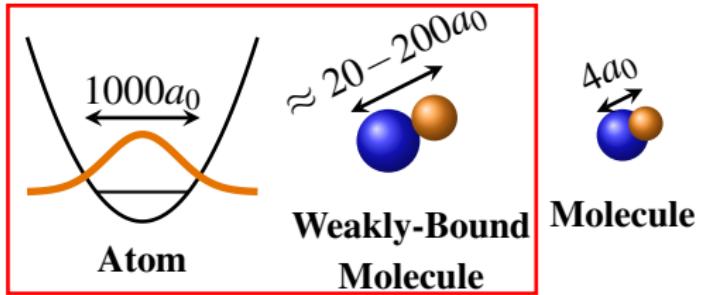


## Molecule

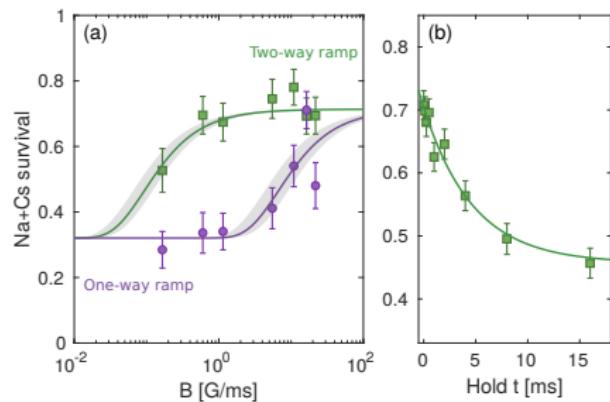




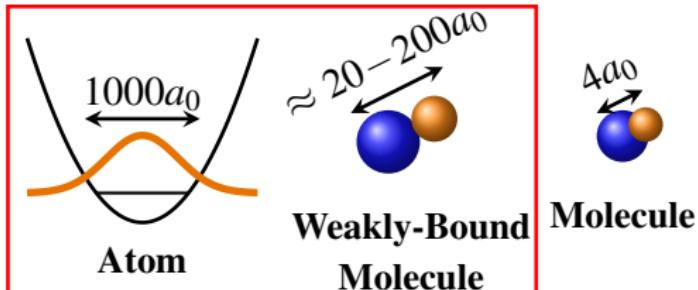
Molecule



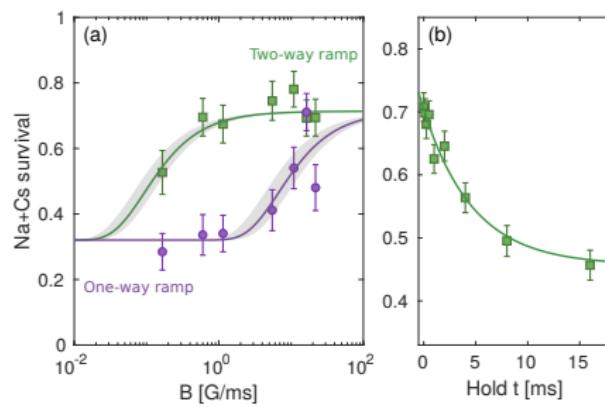
## Feshbach molecule



PRL. 124, 253401 (2020)

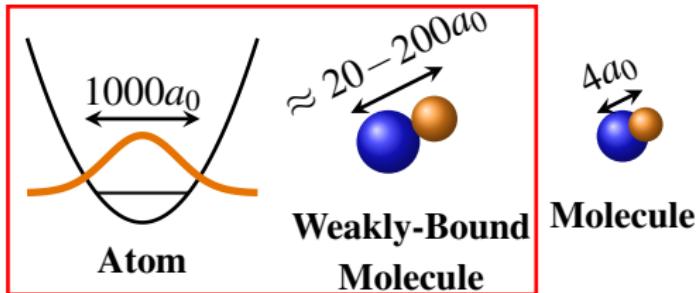


## Feshbach molecule



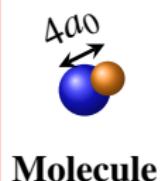
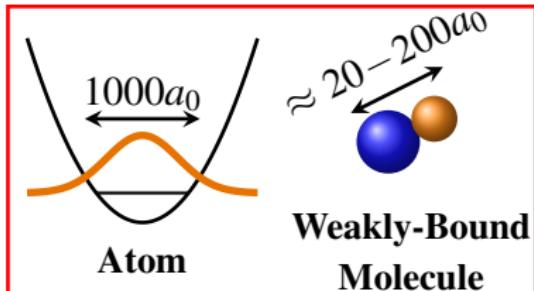
PRL. 124, 253401 (2020)

- Requires Feshbach resonance
- Usually large magnetic field



## Optical transfer

- More general
- Faster

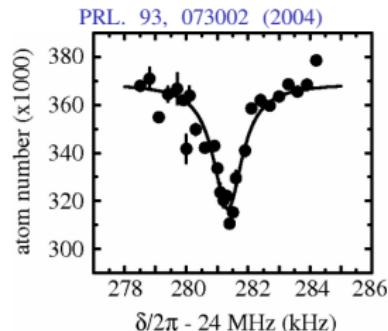


## Optical transfer

- More general
- Faster

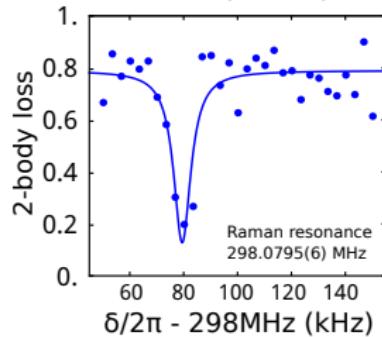
## Previous results

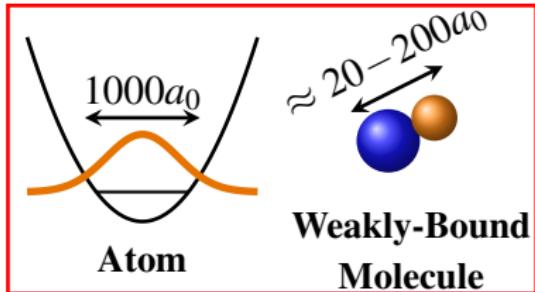
$\text{Rb}_2$  Science 287, 1016 (2000)



$\text{Sr}_2$  PRL. 109, 115302 (2012)

$\text{NaCs}$  Y. Yu et al., PRX. 9, 021039 (2019)





**Molecule**

## Optical transfer

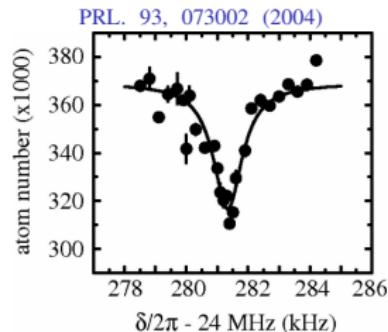
- More general
- Faster

### Limitations so far

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

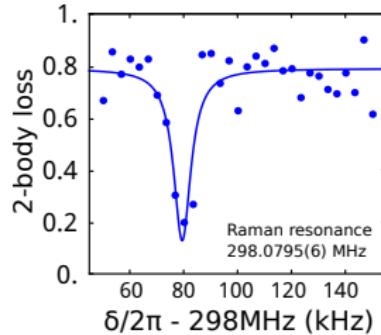
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Rb<sub>2</sub> Science 287, 1016 (2000)

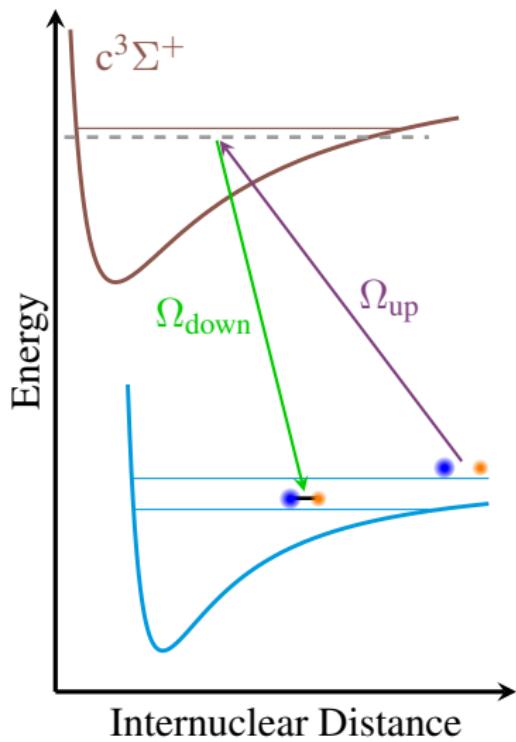


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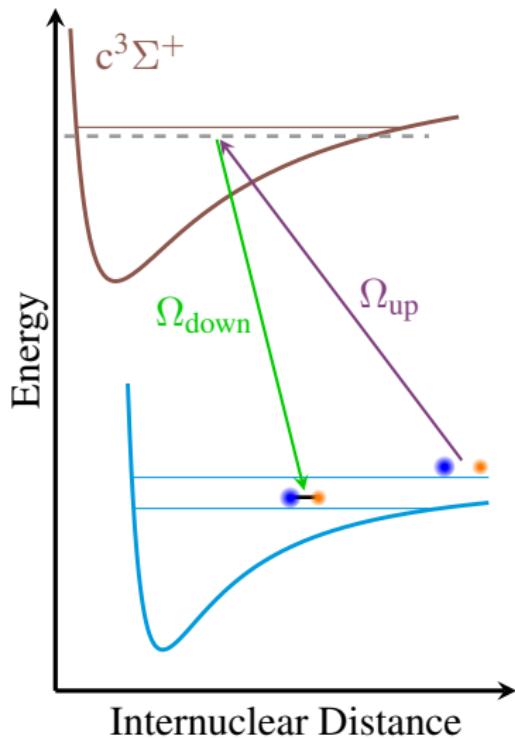
# Raman transfer



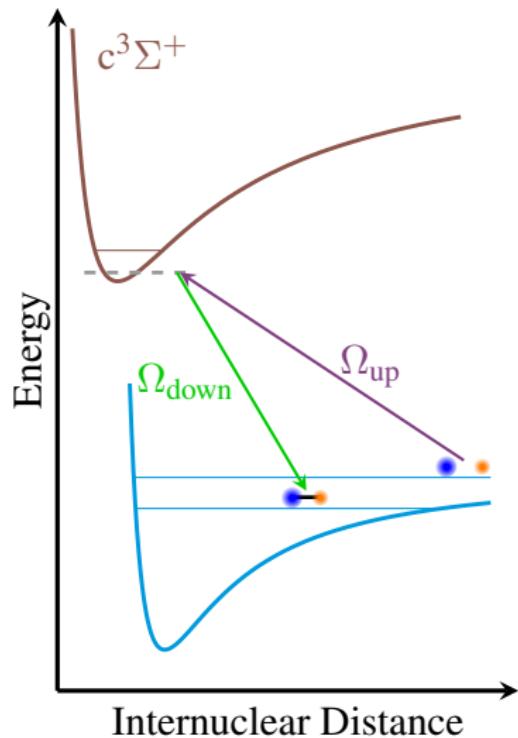
# Raman transfer

## Near threshold states

- Stronger coupling ( $\Omega_{\text{up}}$  and  $\Omega_{\text{down}}$ )
- Closely spaced
- Fast scattering



# Raman transfer



## Near threshold states

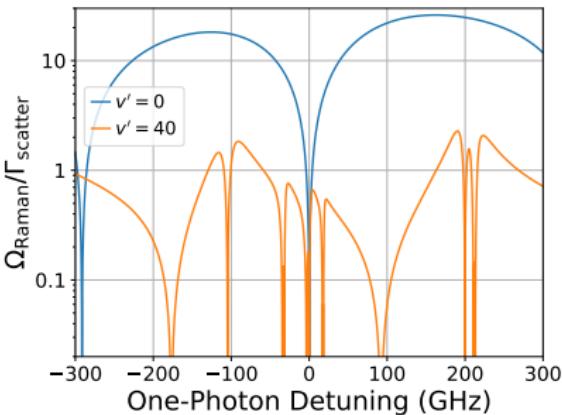
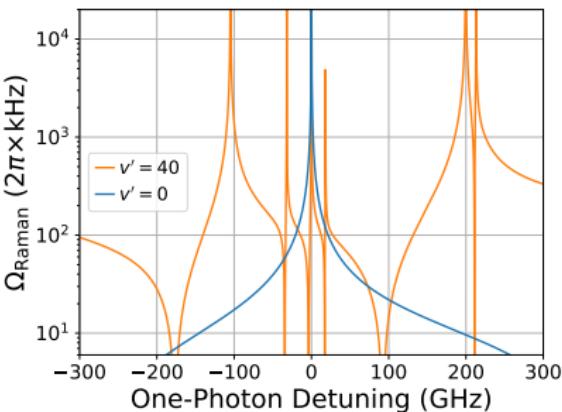
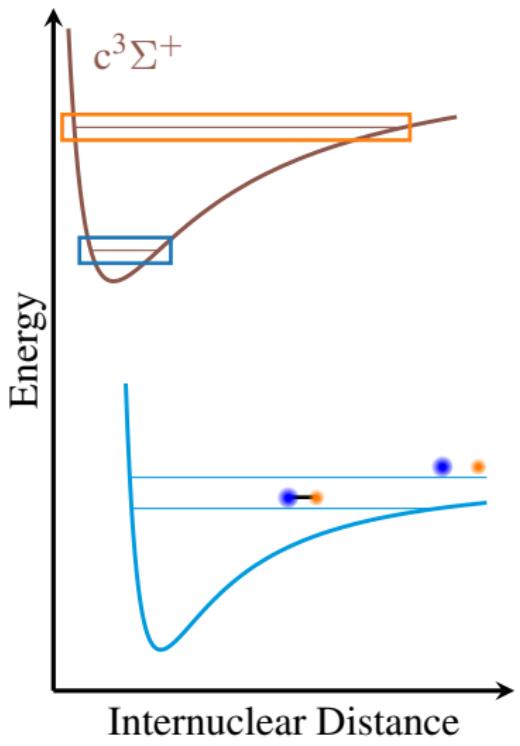
- Stronger coupling ( $\Omega_{\text{up}}$  and  $\Omega_{\text{down}}$ )
- Closely spaced
- Fast scattering

## Deeply bound states

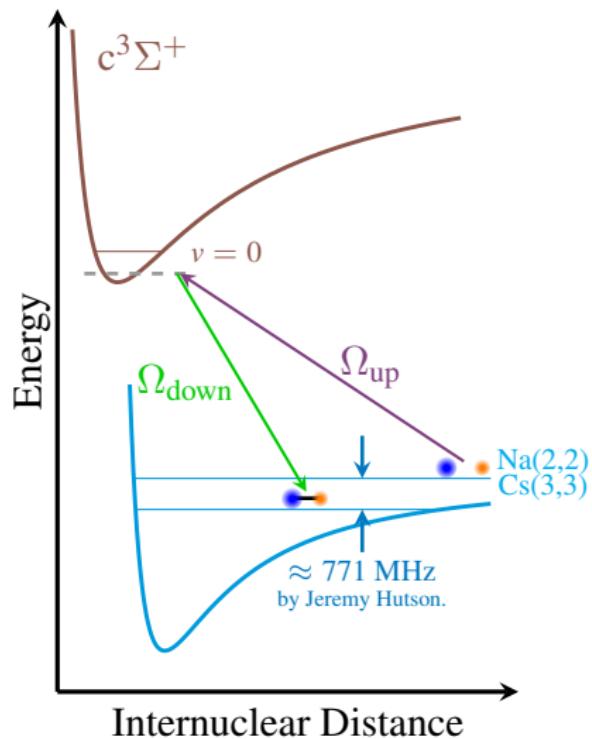
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121 (2017)

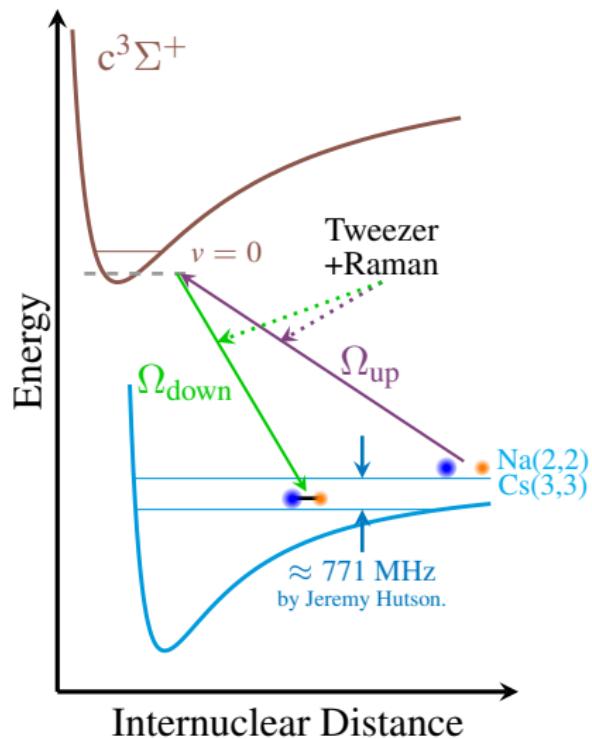
## Raman transfer



# Experiment



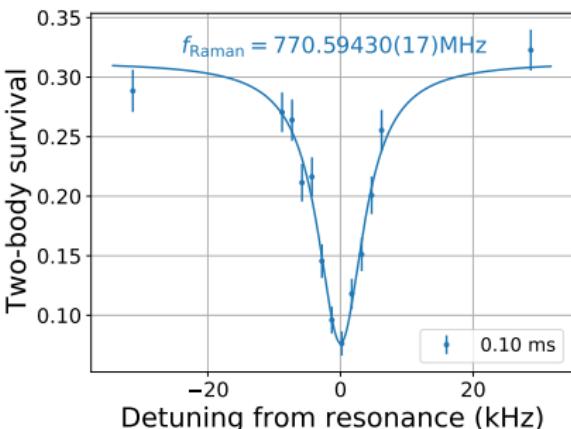
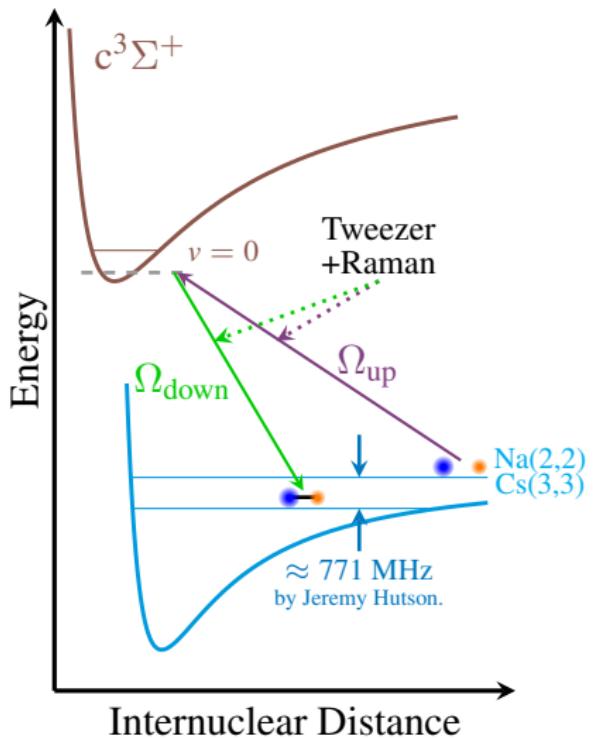
# Experiment



## Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

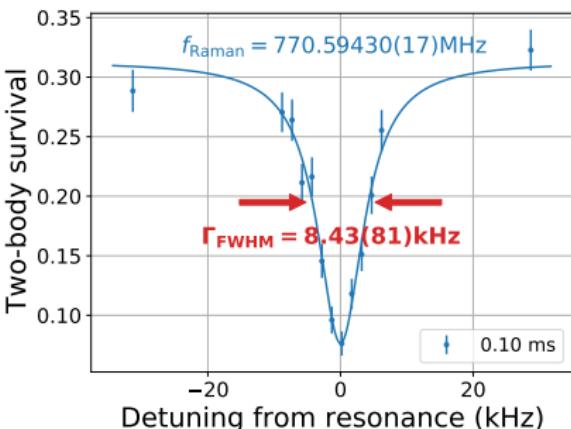
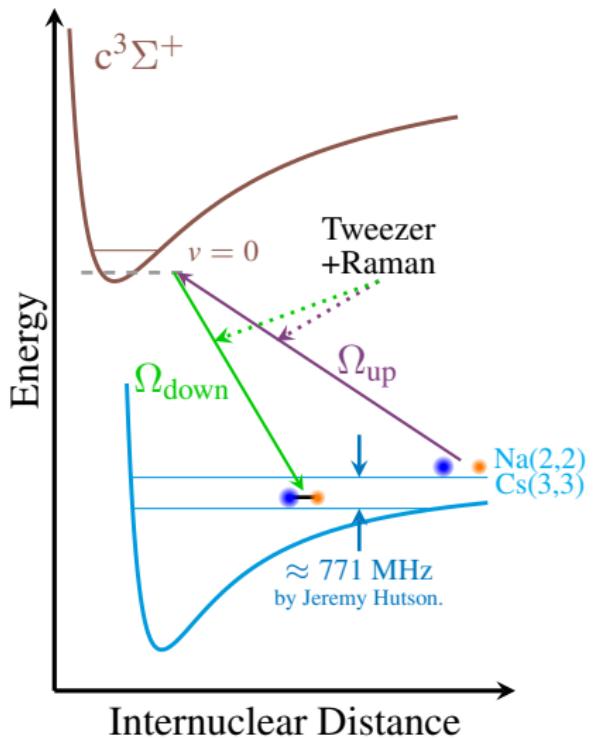
# Experiment



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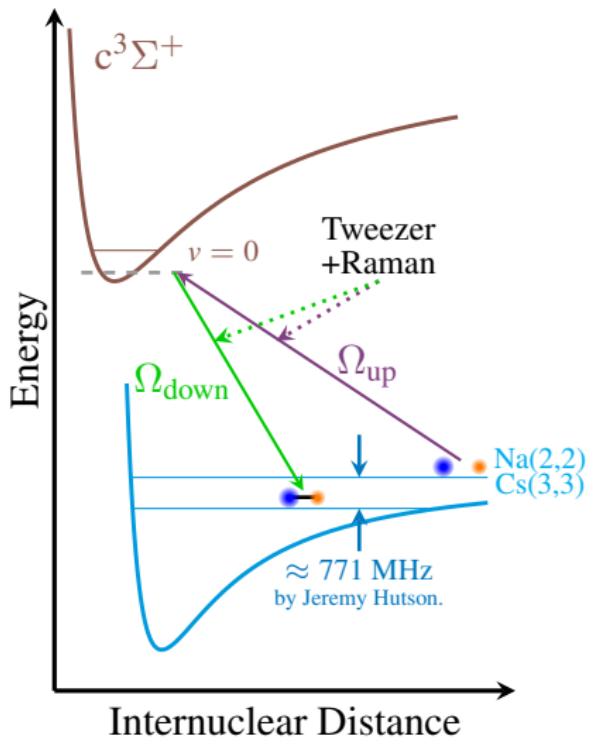
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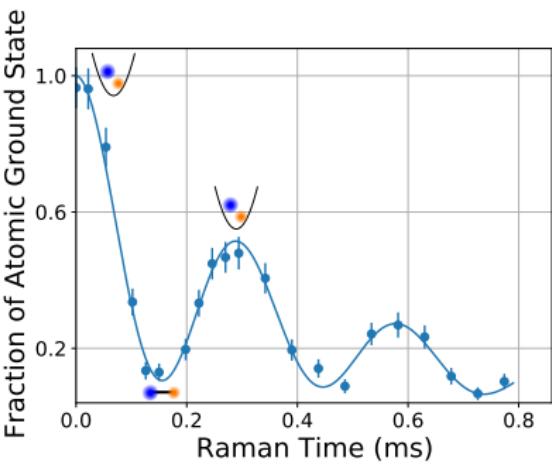
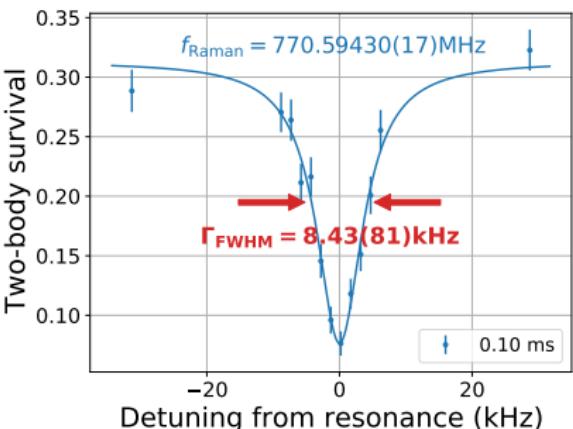
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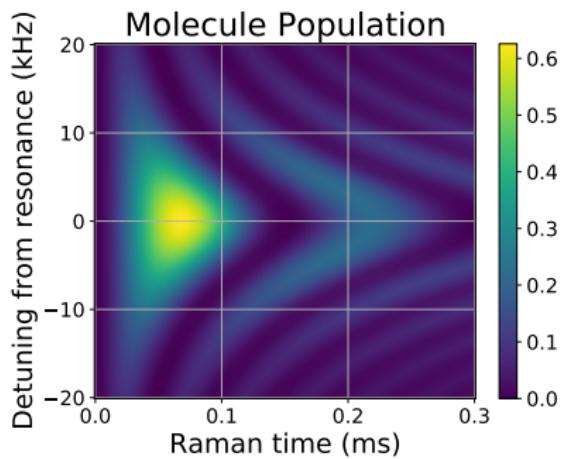
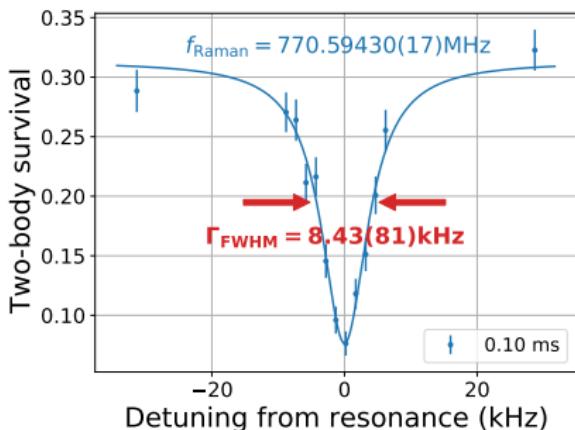


arXiv:2012.09043 (2020)

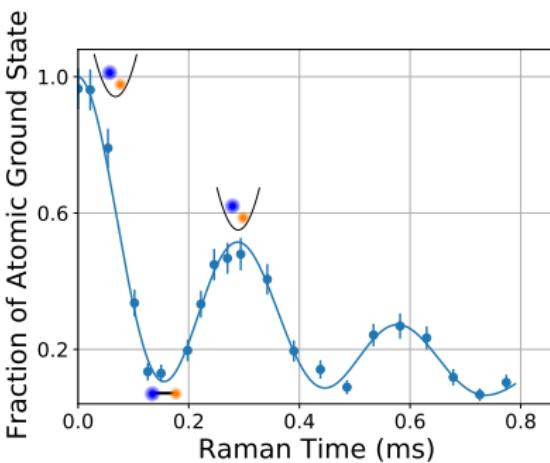


# Experiment

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

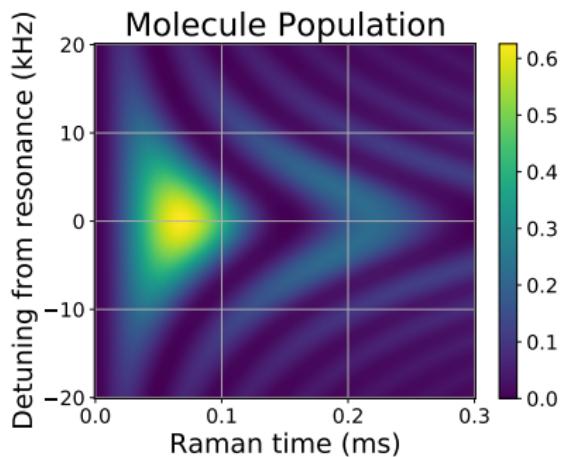
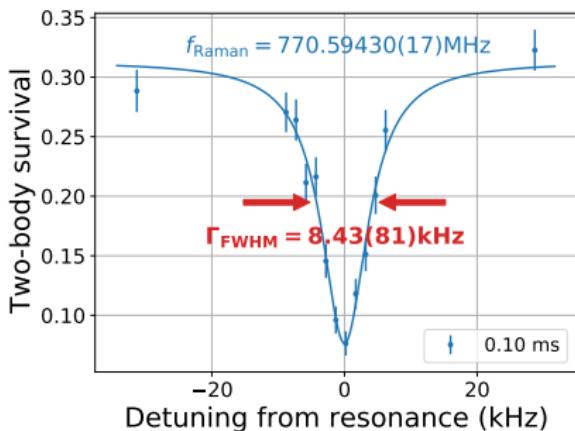


arXiv:2012.09043 (2020)

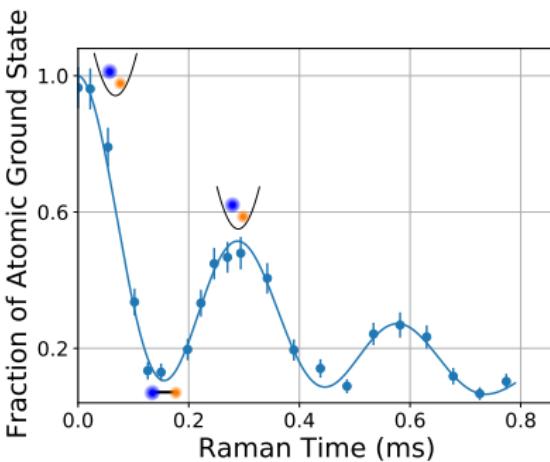


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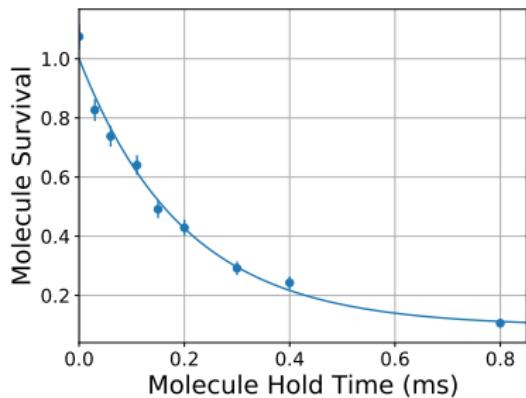


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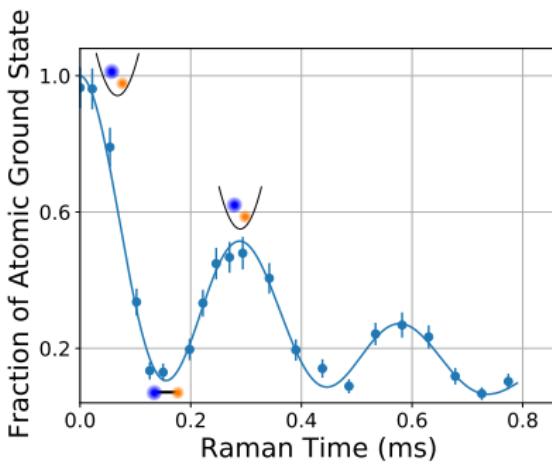
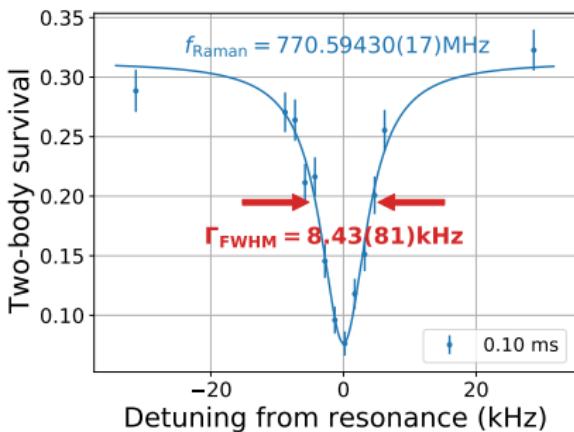


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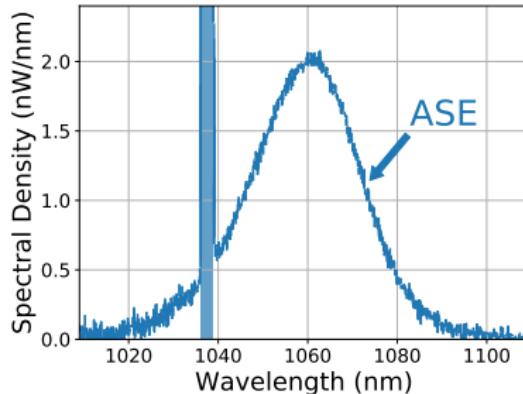
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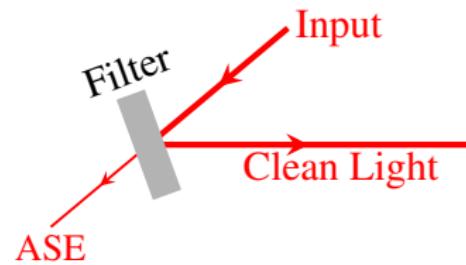
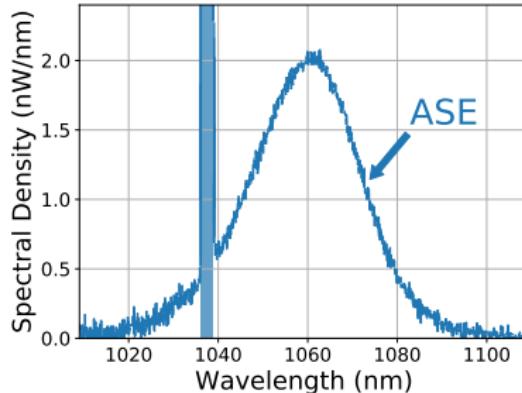
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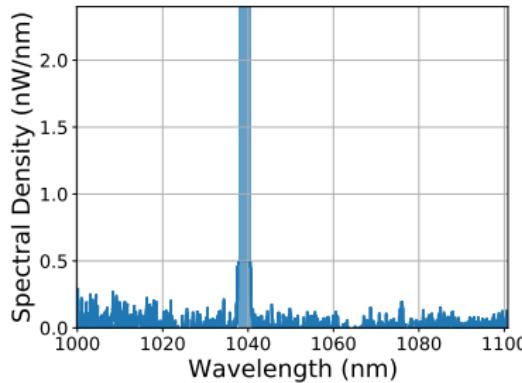
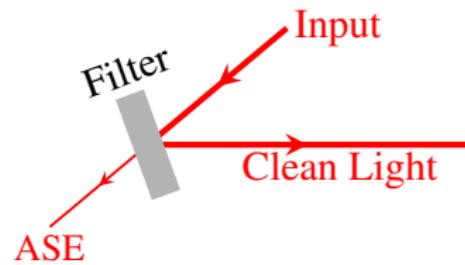
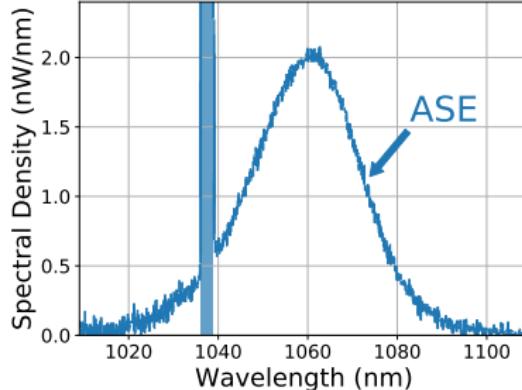
# Amplified Spontaneous Emission (ASE)



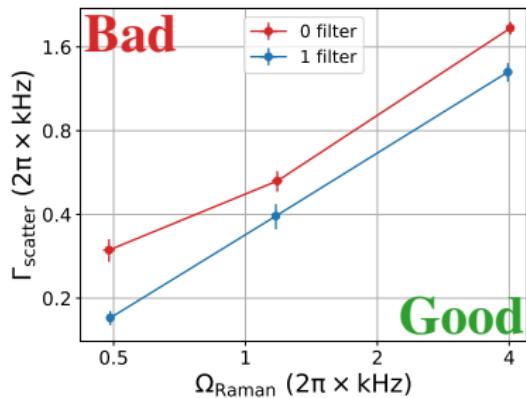
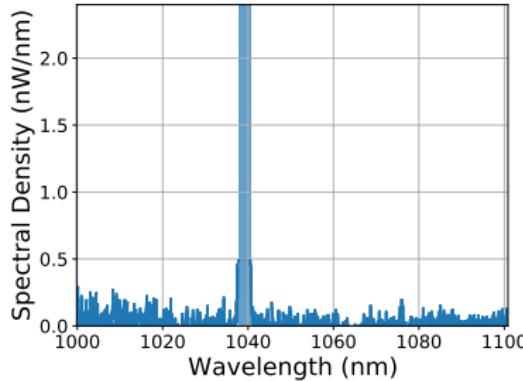
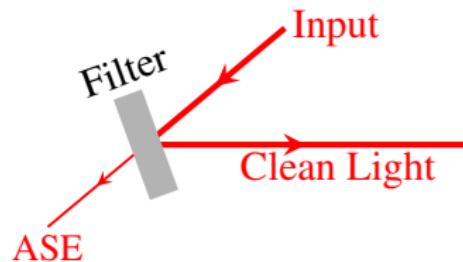
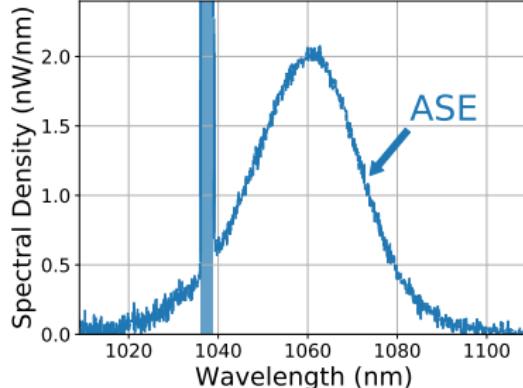
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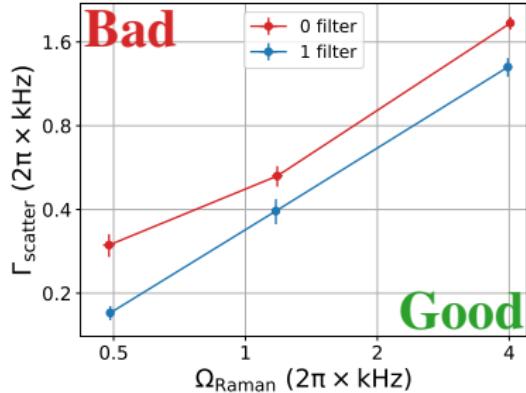
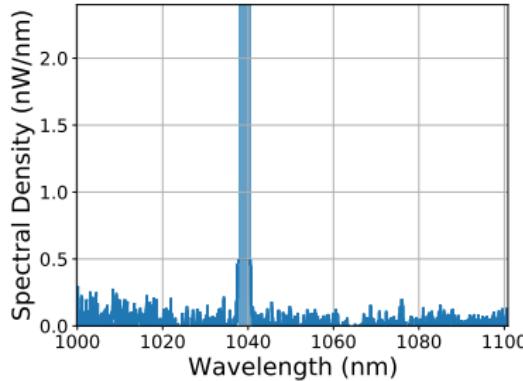
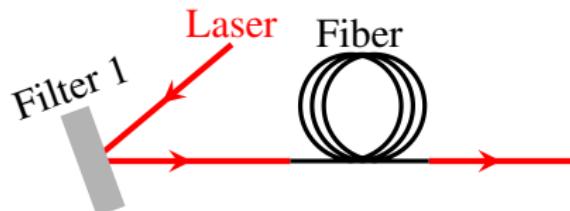
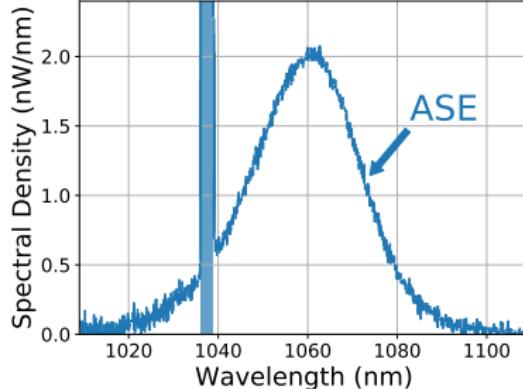
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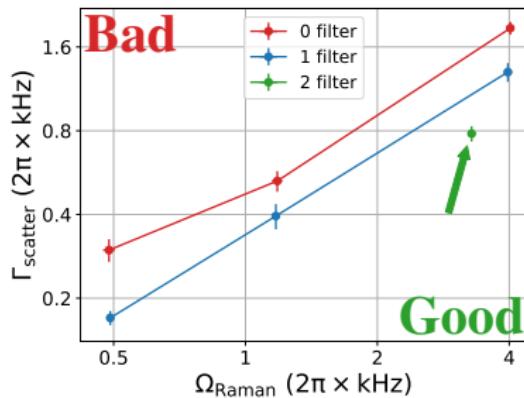
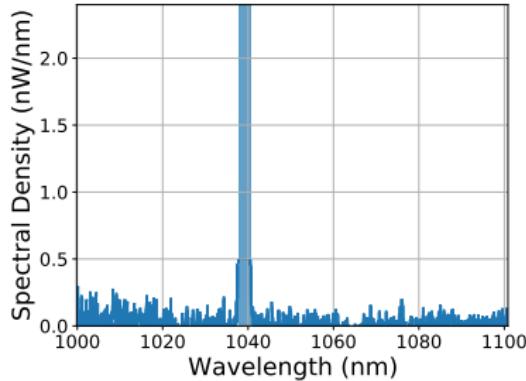
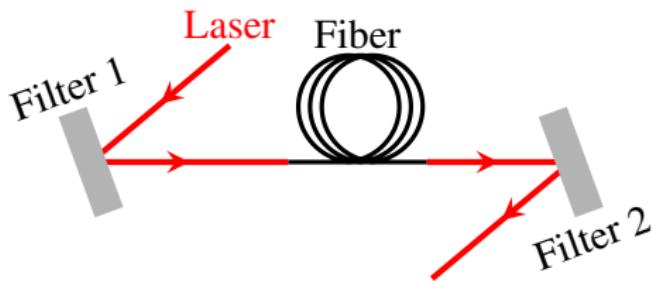
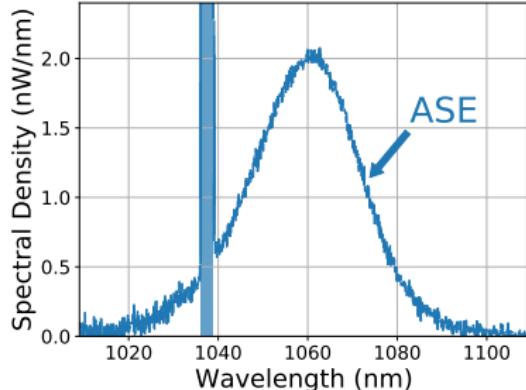
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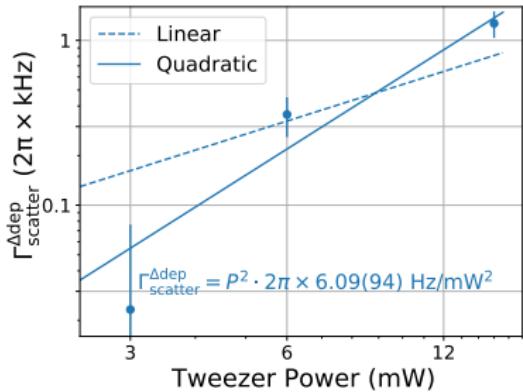


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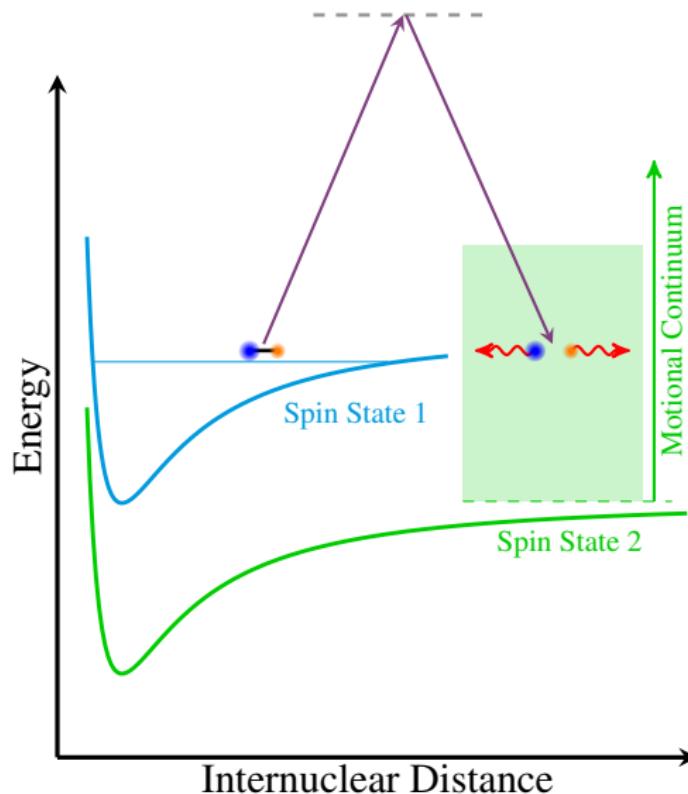
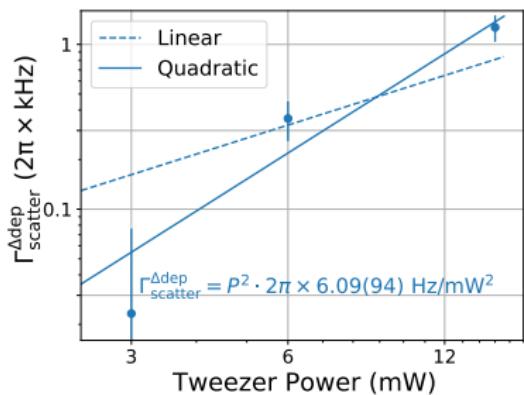
# Two-Photon Scattering

## Detuning Dependent Scattering



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Detuning Dependent Scattering



## Conclusion and outlook

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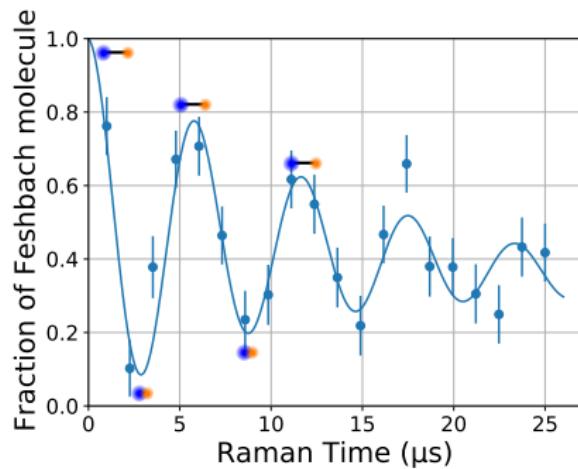
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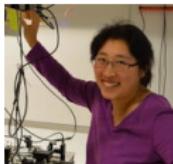
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PI



Kang-Kuen Ni

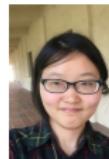
NaCs  
Team



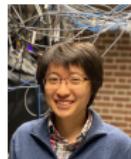
Kenneth  
Wang



Yu  
Wang



Fang  
Fang



Jessie  
Zhang



Lewis  
Picard



William  
Cairncross

KRb  
Team



Lingbang  
Zhu



Mingguang  
Hu



Matthew  
Nichols



Lee Liu  
Postdoc @JILA



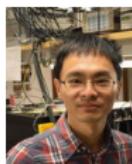
Nick Hutzler  
AP @Caltech



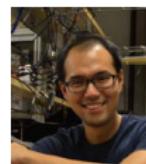
Jonathan Hood  
AP @Purdue



Eliot  
Fenton



Yen-Wei Lin  
Intelon Optics



Yu Liu  
Postdoc @NIST

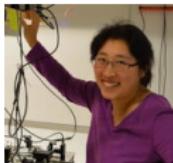


Andrei  
Gheorghe



David Grimes  
Instructor @MIT

PI



Kang-Kuen Ni



NaCs  
Team



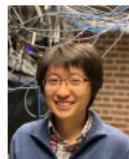
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Wang



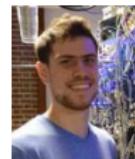
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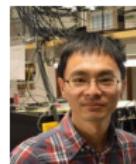
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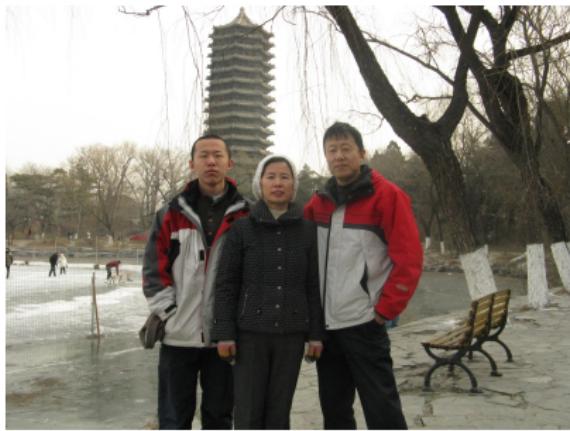
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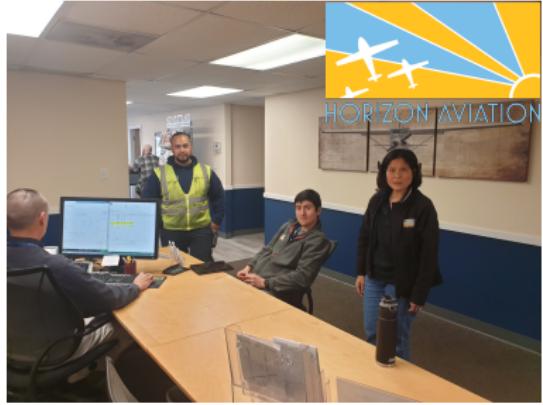
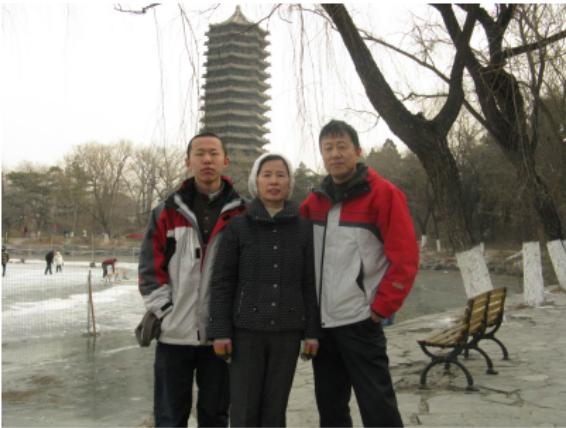
Andrei  
Gheorghe  
Instructor @MIT



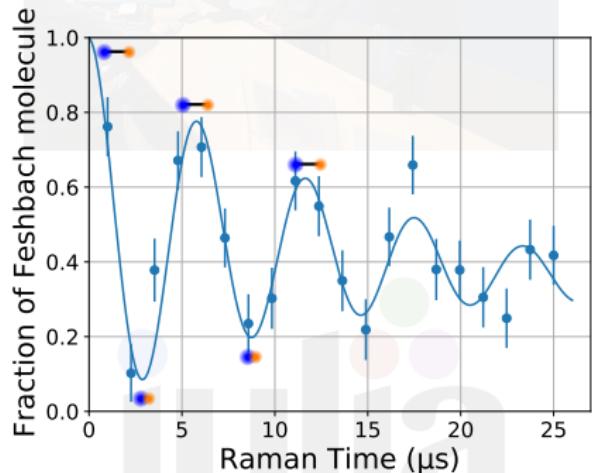
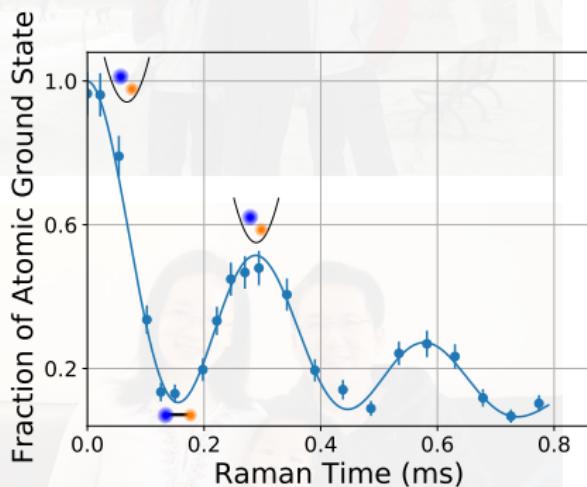
David Grimes  
Instructor @MIT







# Thanks for your attention





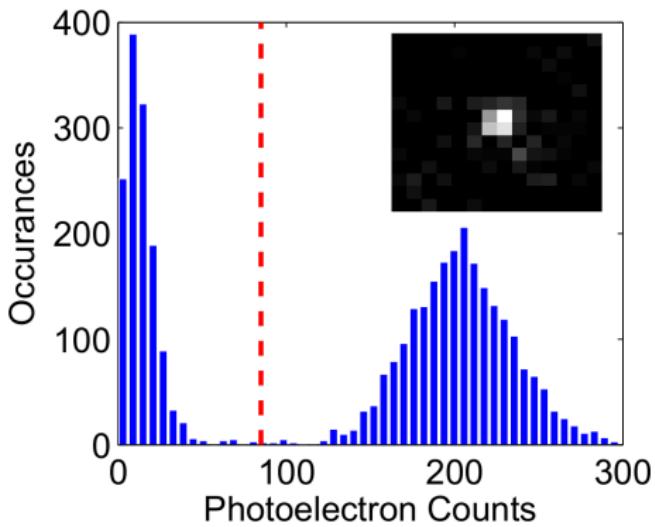


# Single Atom in Tweezer

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

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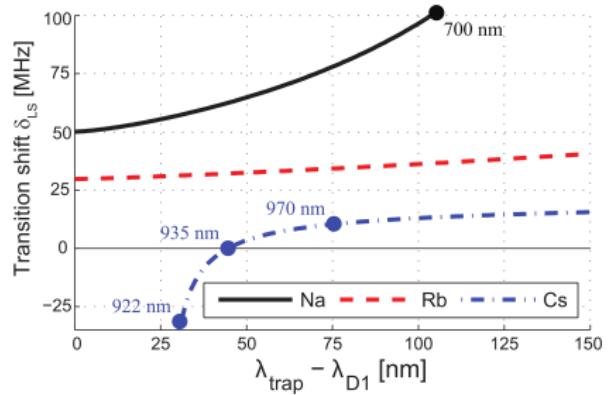
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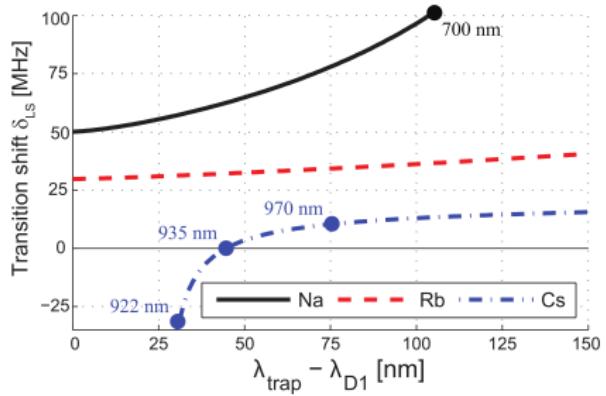
## Issues with Na

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

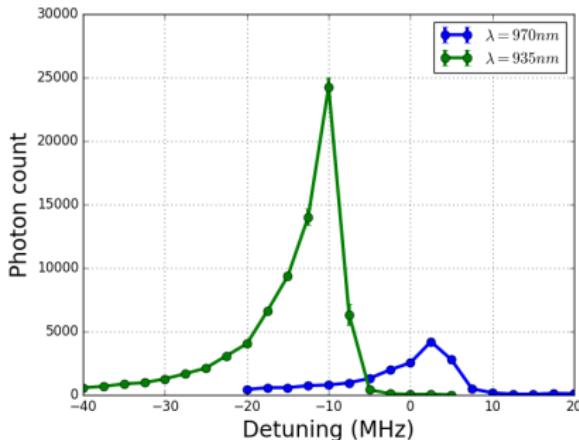
# Real Issue with Na: Light Shift



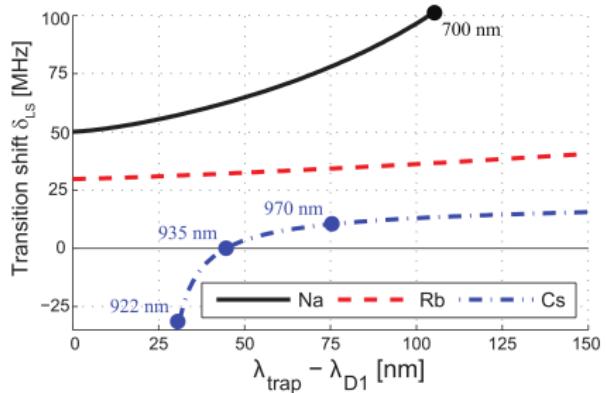
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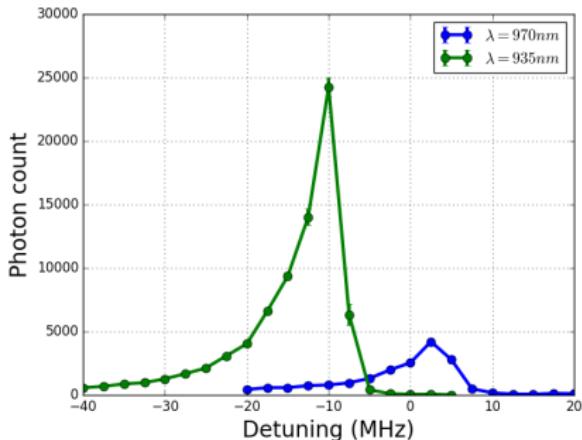
## Cs single atom imaging



## Real Issue with Na: Light Shift



## Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

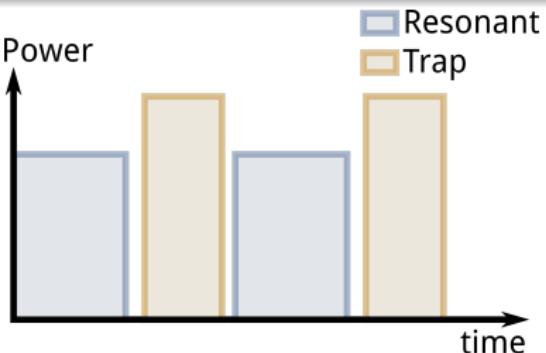
# Real Issue with Na: Light Shift

## Trap modulation

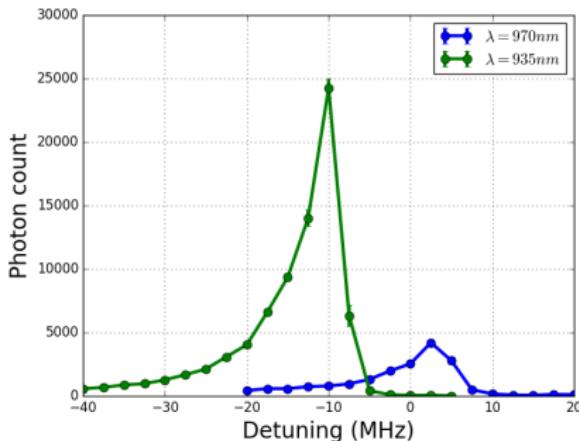
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



## Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

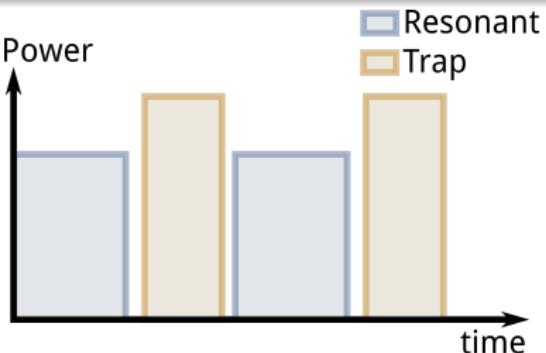
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## Trap modulation

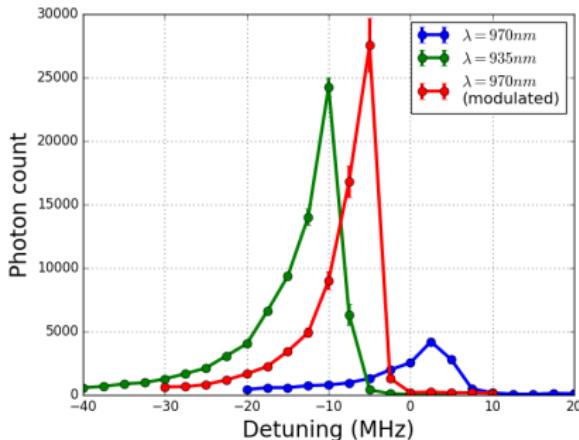
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



## Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

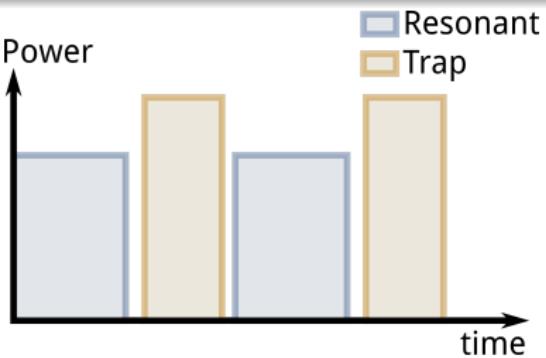
# Real Issue with Na: Light Shift

## Trap modulation

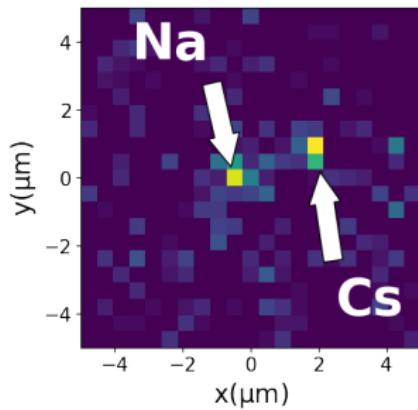
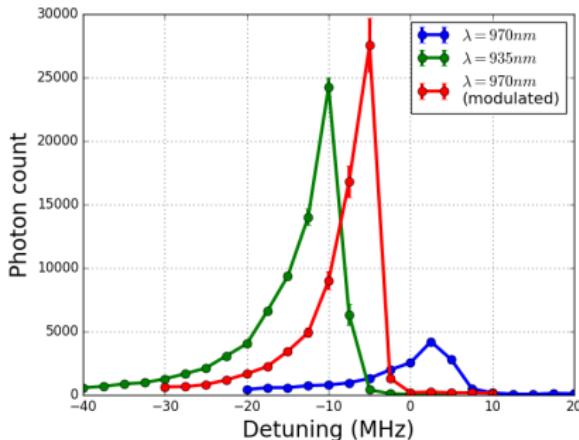
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

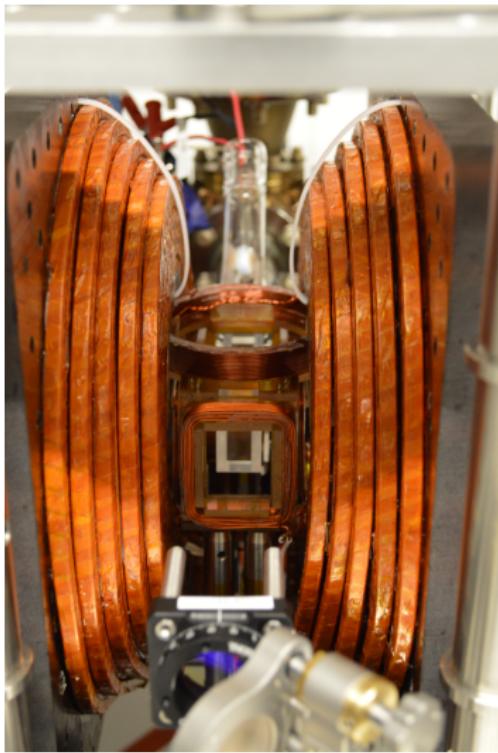
$$\Gamma = 2\pi \times 10 \text{ MHz}$$



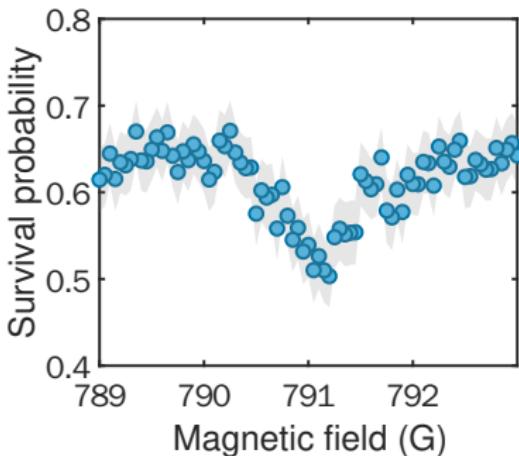
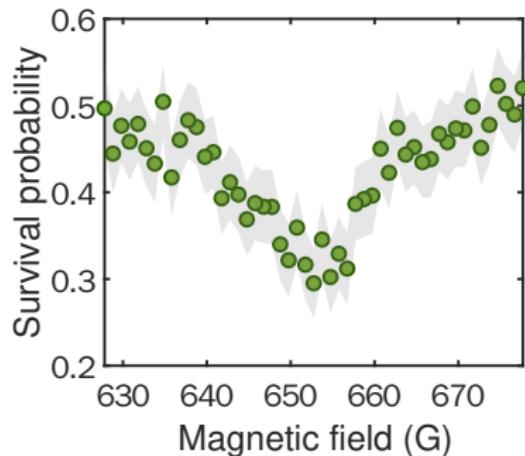
## Cs single atom imaging



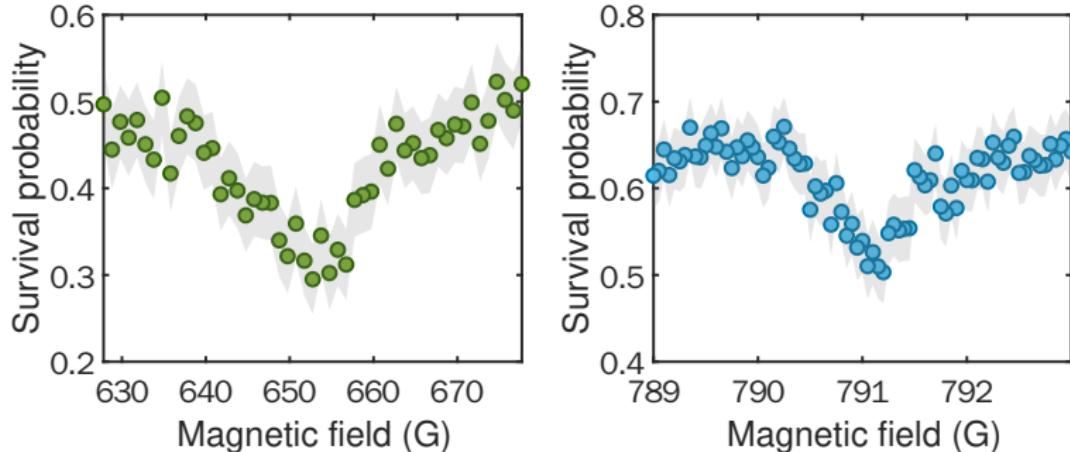
# Na (1, -1) Cs (3, -3) Feshbach resonance



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



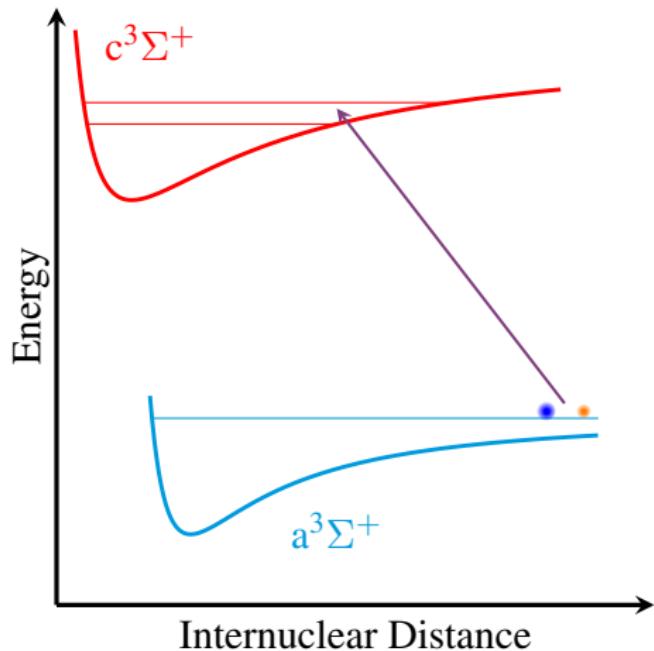
## Na (1, -1) Cs (3, -3) Feshbach resonance



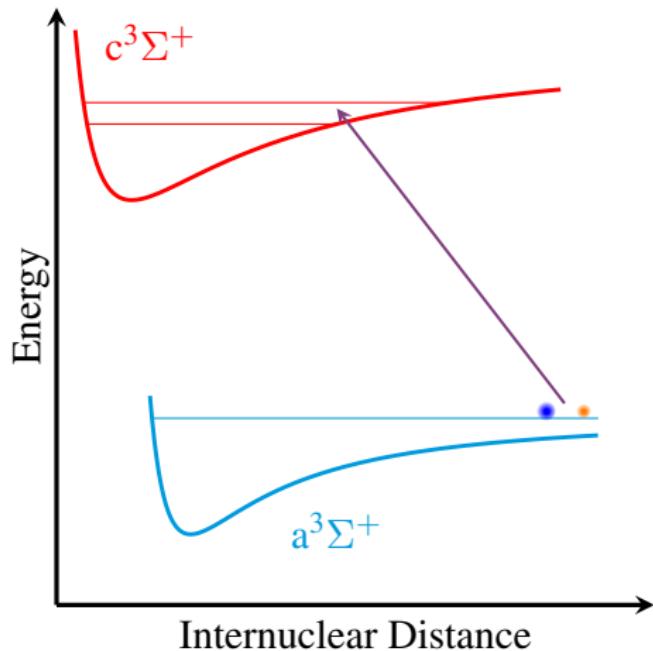
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) <sup>1</sup>	663 G	799 G
Measured	652(3) G	791.2(2) G

<sup>1</sup>In collaboration with Bo Gao

# Photoassociation (PA) Spectroscopy



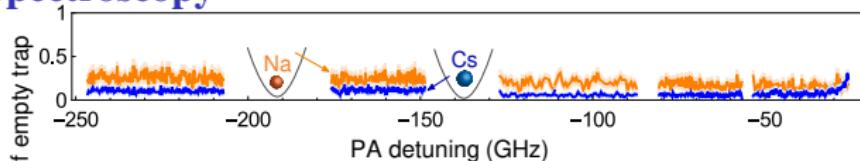
# Photoassociation (PA) Spectroscopy



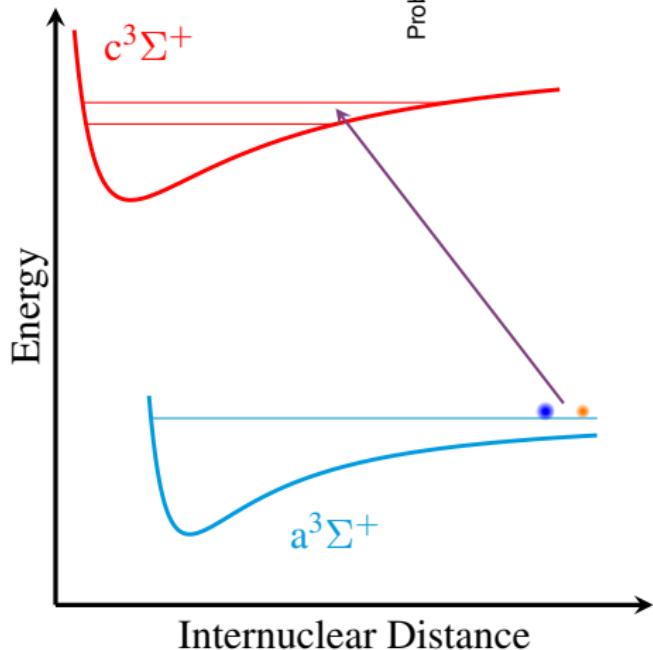
## Single Atom PA

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

# Photoassociation (PA) Spectroscopy



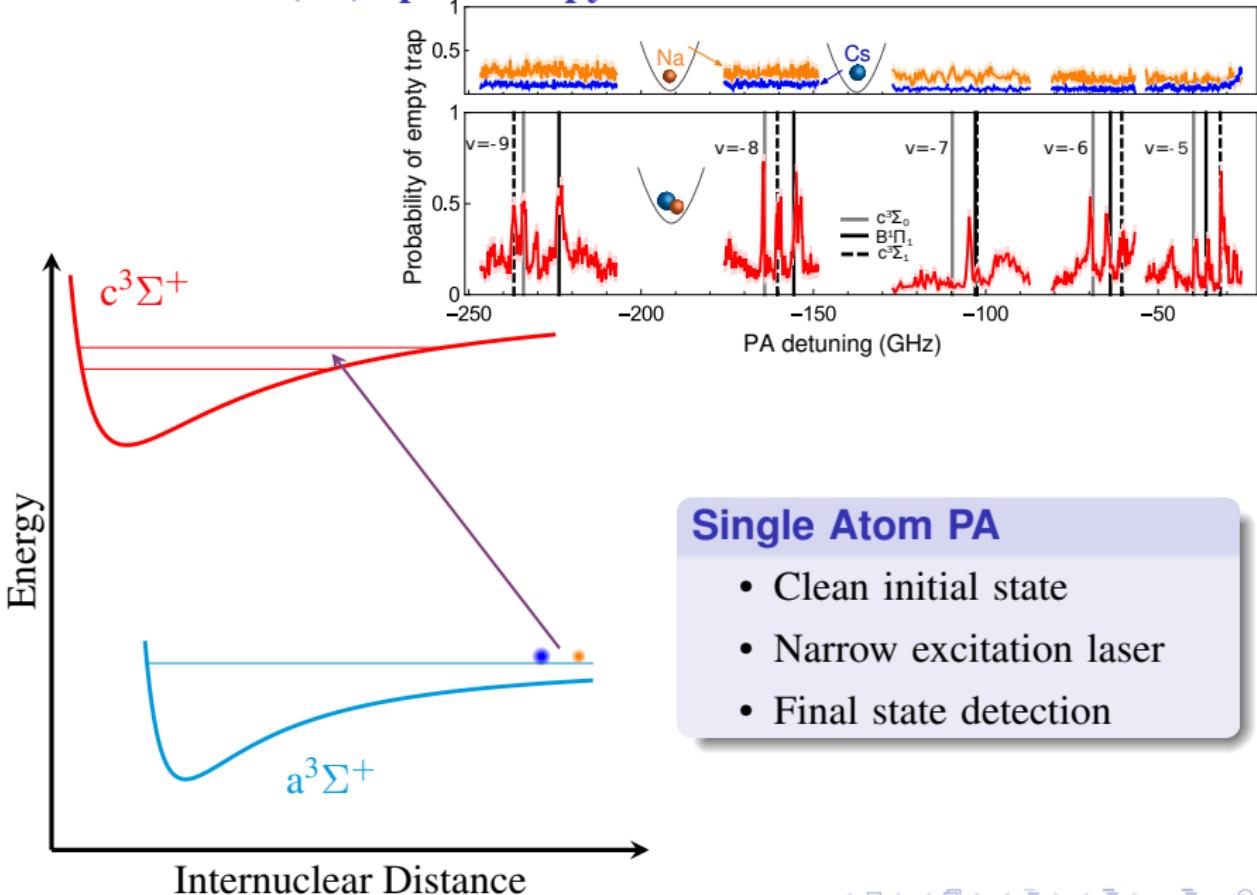
Probability of empty trap



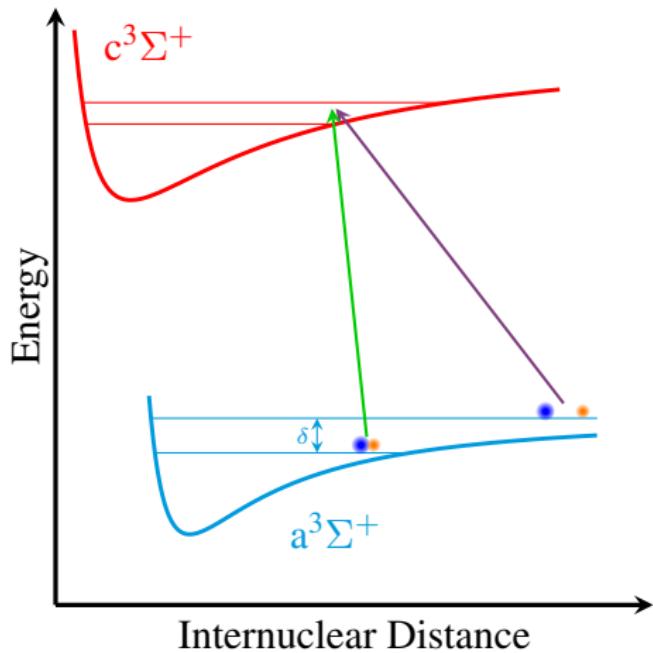
## Single Atom PA

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

# Photoassociation (PA) Spectroscopy



# Electromagnetically Induced Transparency (EIT) Spectroscopy



# Electromagnetically Induced Transparency (EIT) Spectroscopy

