

# Coherent Creation of Single Molecules from Single Atoms

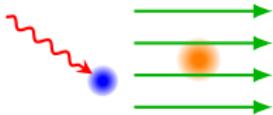
Yichao Yu

Ni Group/Harvard

Simple System



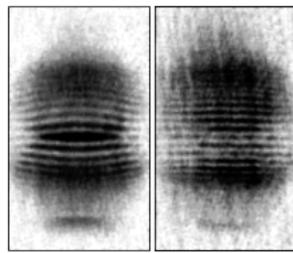
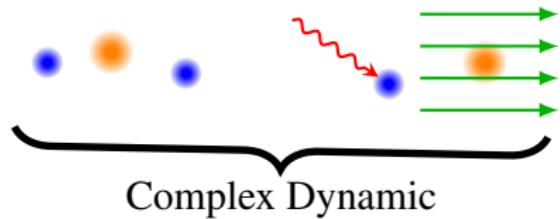
Full Control



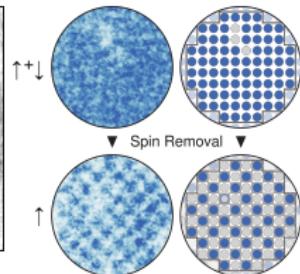
Complex Dynamic

Simple System

Full Control



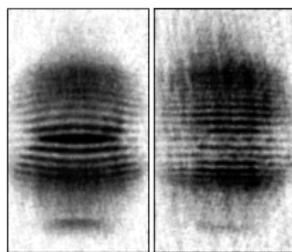
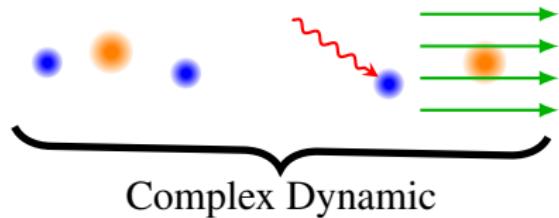
Ketterle et al.



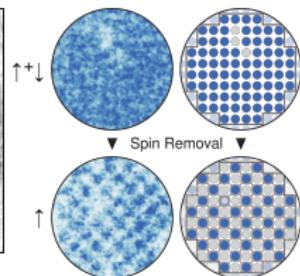
Greiner et al.

Simple System

Full Control



Ketterle et al.



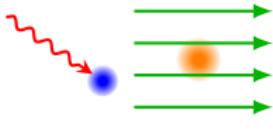
Greiner et al.

- ✗ Simple internal structure
- ✗ Weak interaction

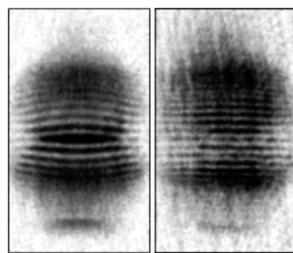
## Simple System



## Full Control

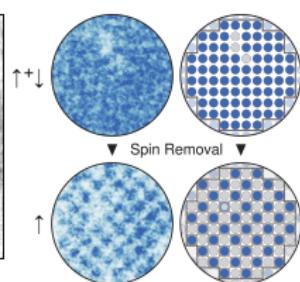


## Complex Dynamic



Ketterle et al.

Greiner et al.



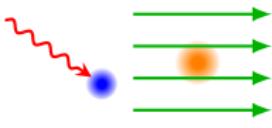
- Strong interaction
- Rich internal structure
- Long coherence time
- Fully controllable

- Simple internal structure
- Weak interaction

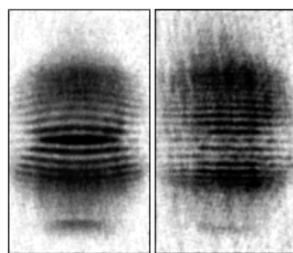
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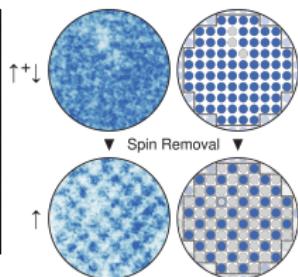
## Full Control



## Complex Dynamic

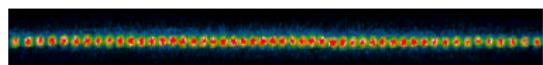


Ketterle et al.



Greiner et al.

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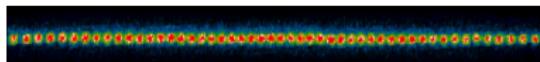
Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)

- Simple internal structure
- Weak interaction

- Strong interaction
- Rich internal structure
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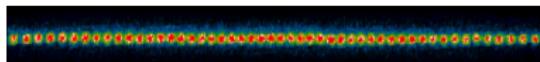
Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)

## ✓ Strong interaction (kHz)

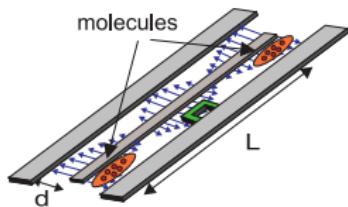
- Rich internal structure
- Long coherence time
- Fully controllable**



Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)



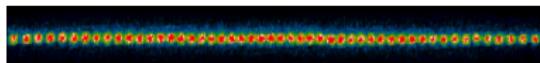
Dipolar Molecule (PRL. 97, 33003 (2006))

✓ Strong interaction (kHz)

□ Rich internal structure

✓ Long coherence time

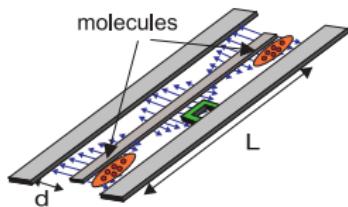
□ Fully controllable



Ions (Monroe et al.)

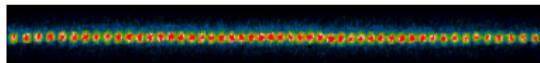


Rydberg Atoms (Lukin et al.)



Dipolar Molecule (PRL. 97, 33003 (2006))

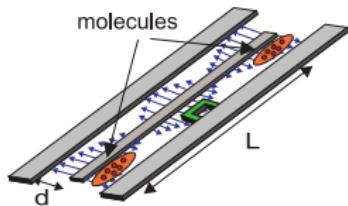
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Ions (Monroe et al.)



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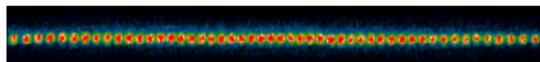
Dipolar Molecule (PRL. 97, 33003 (2006))

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## Optical tweezers

- Single site resolution

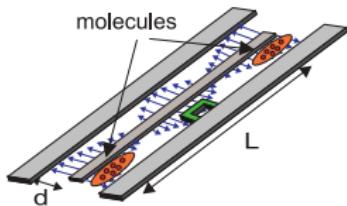
... . . .



Ions (Monroe et al.)

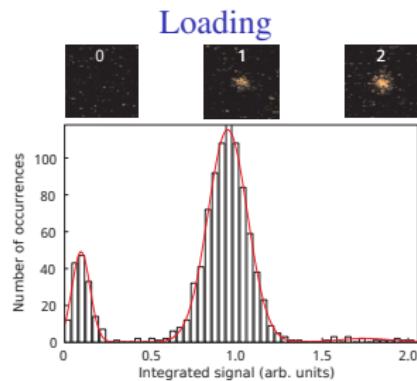


Rydberg Atoms (Lukin et al.)



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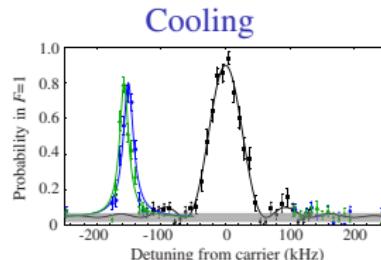
- ✓ Strong interaction (kHz)
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- ✓ Long coherence time
- ✓ Fully controllable



Nat. Phys. 6, 951 (2010)

## Optical tweezers

- Single site resolution
- ...



PRX. 2, 041014 (2012)

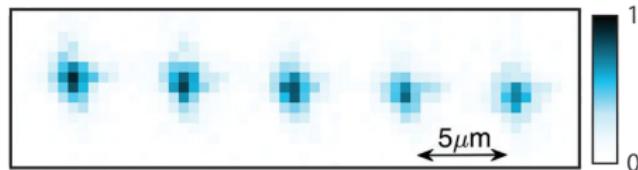
## Rearranging



Science 354, 1024 (2016)

# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

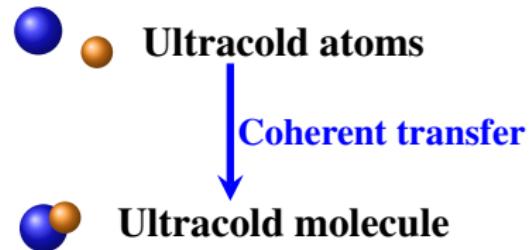
# Ultracold molecules in tweezers

## Direct cooling



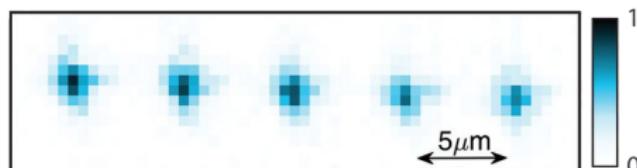
Science 365, 1156 (2019)

## Assembly



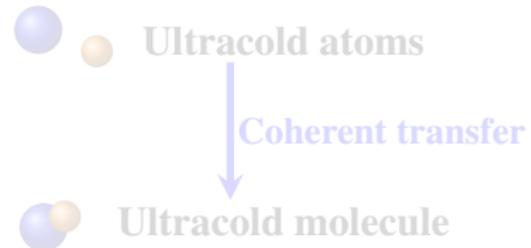
# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

## Assembly



## Challenges

- Temperature in tweezer
- Quantum control

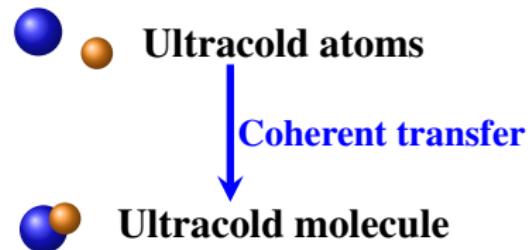
# Ultracold molecules in tweezers

## Direct cooling



Science 365, 1156 (2019)

## Assembly



## Challenges

- Temperature in tweezer
- 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

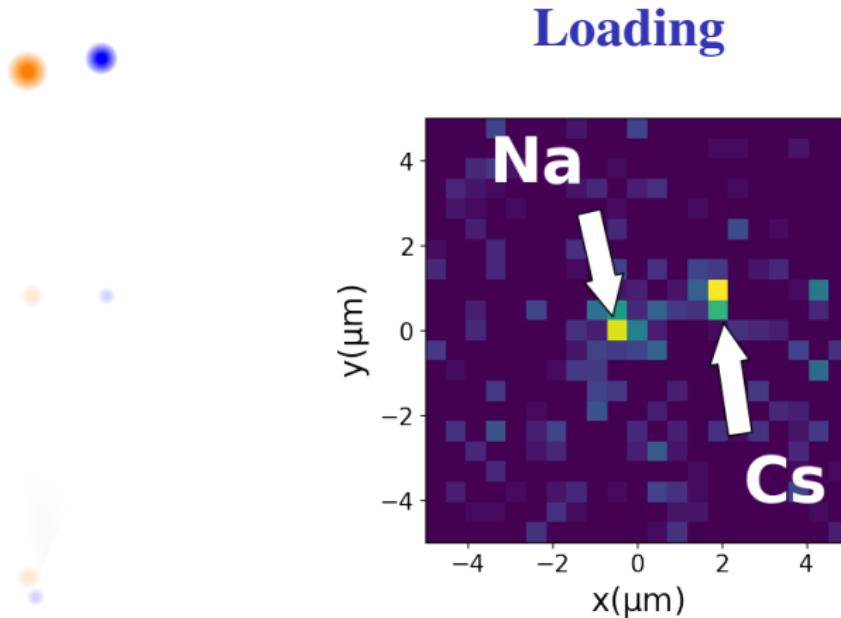
- Atom-atom interaction
- Coherent optical transfer

## 4 Conclusion

### NaCs molecule

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

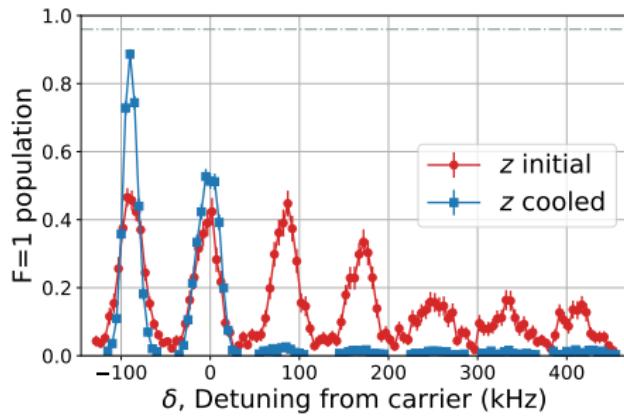
## Experiment overview



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

# Experiment overview

## Cooling



Cs: 96% ground state<sup>1</sup>

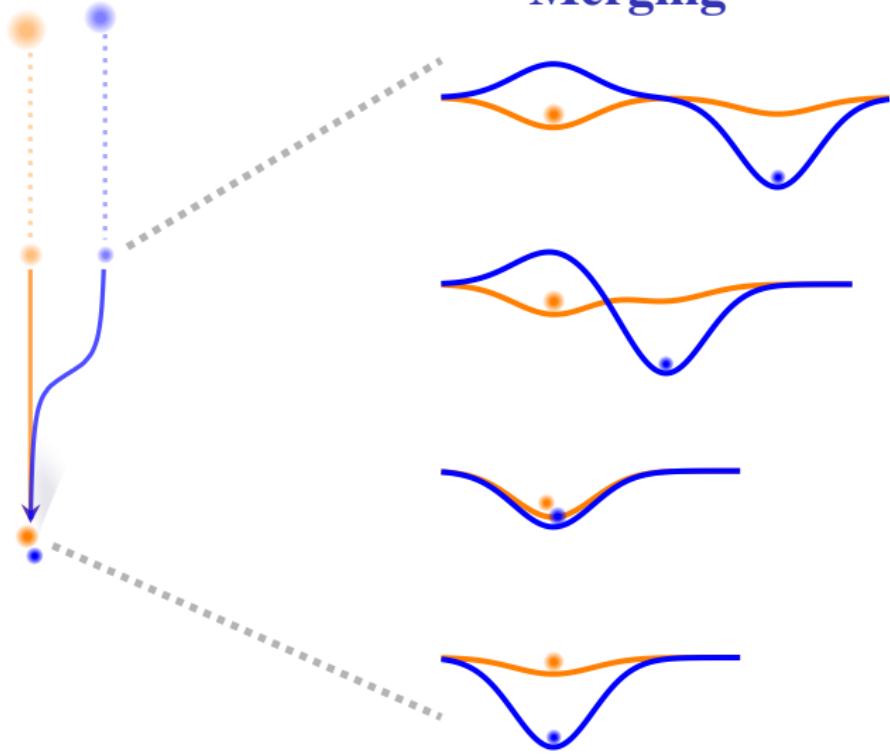
Na: 94% ground state<sup>2</sup>

<sup>1</sup>Y. Yu et al. PRX 9, 021039 (2019)

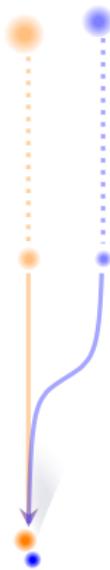
<sup>2</sup>Y. Yu et al. PRA 97, 063423 (2018)

## Experiment overview

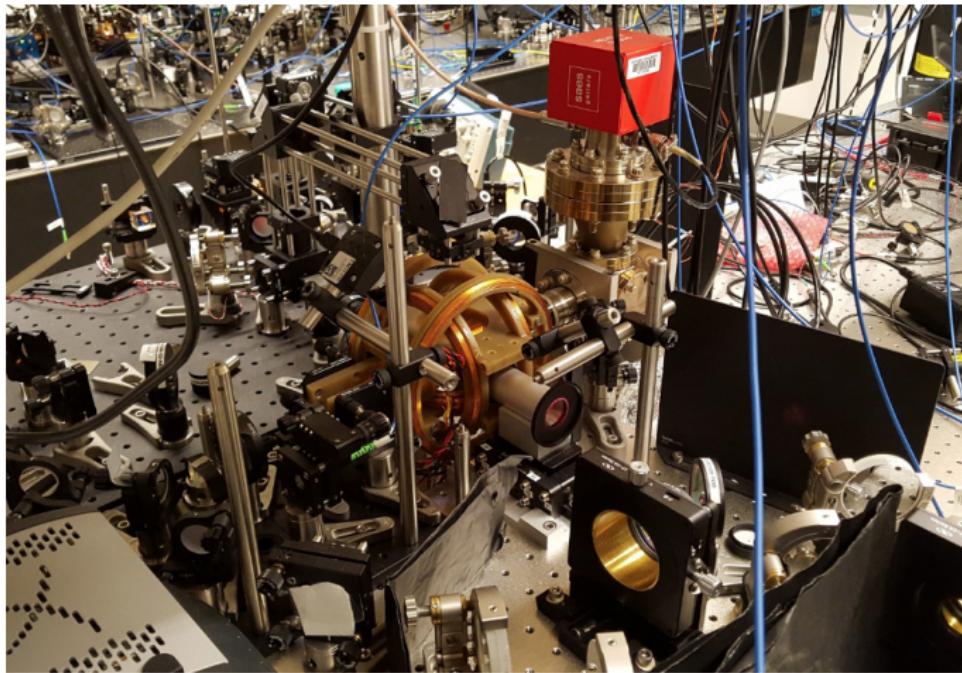
### Merging

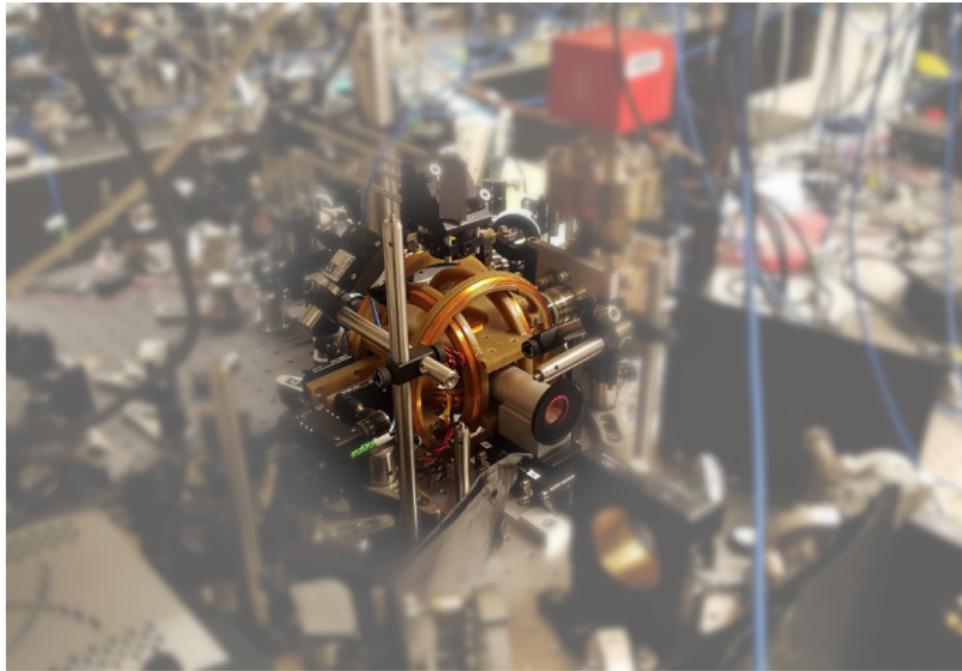


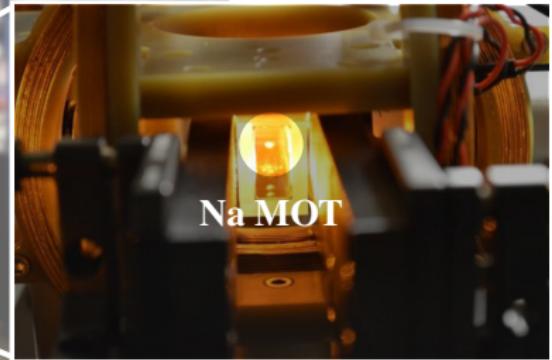
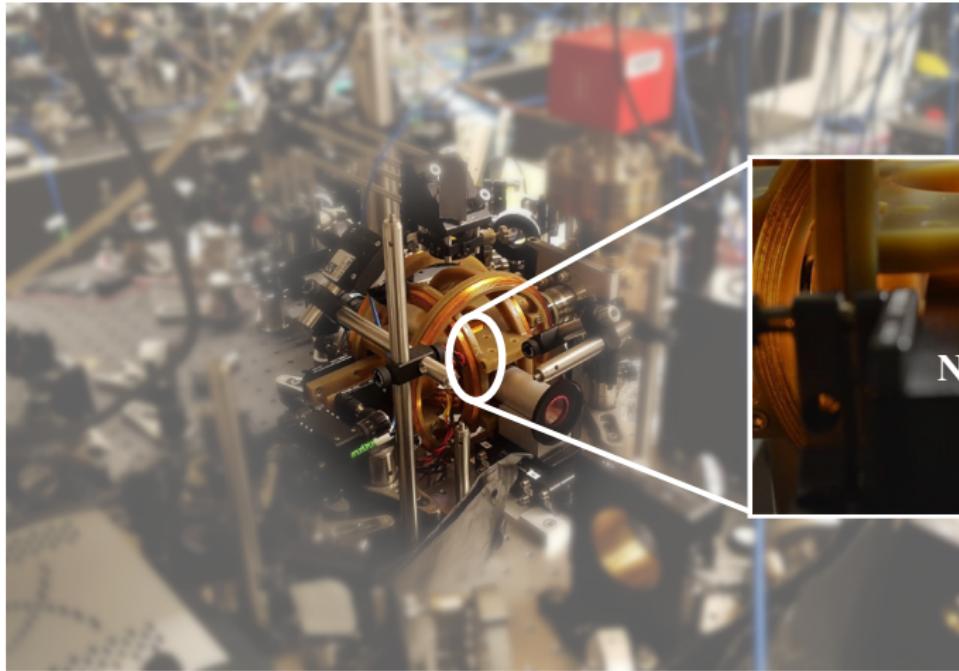
# Experiment overview



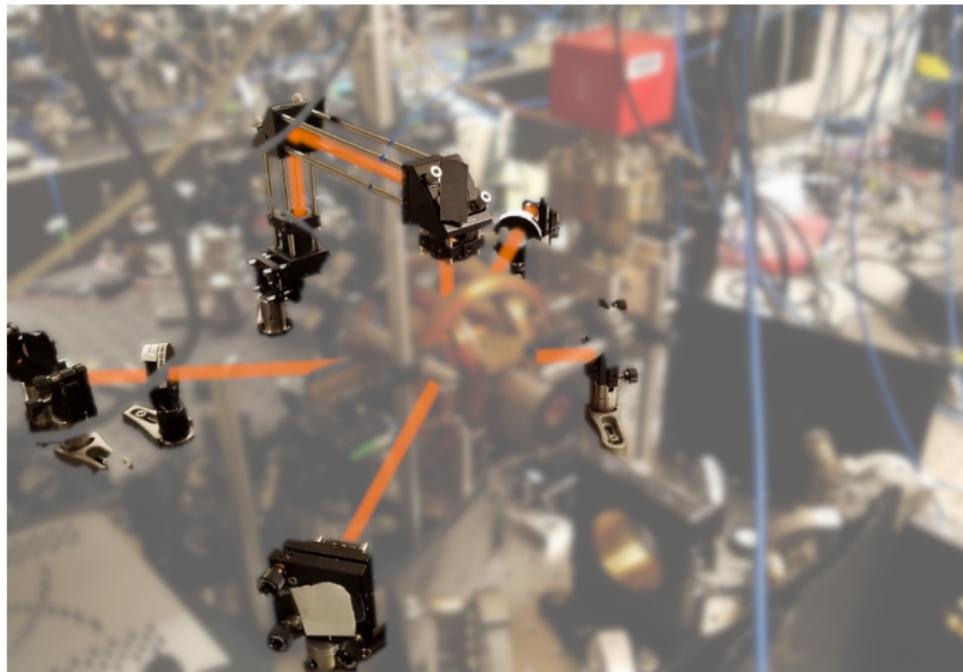
L. R. Liu, J. D. Hood, Y. Yu et al., Science 360, 6391 (2018)



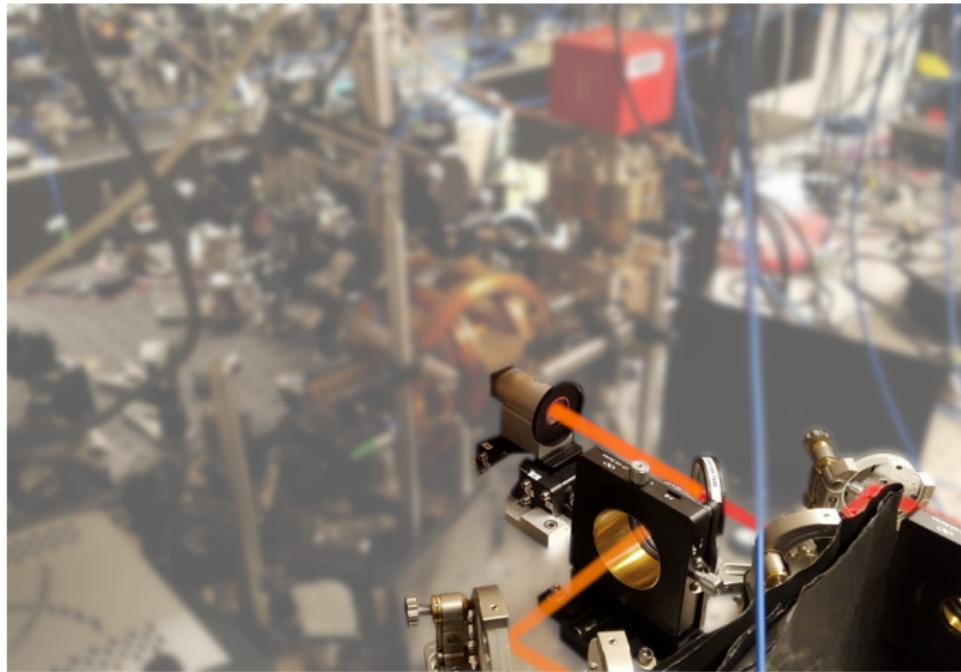




## MOT beam path



## Tweezer and imaging beam path



# Outline

## 1 Experiment overview

## 2 Atom state control

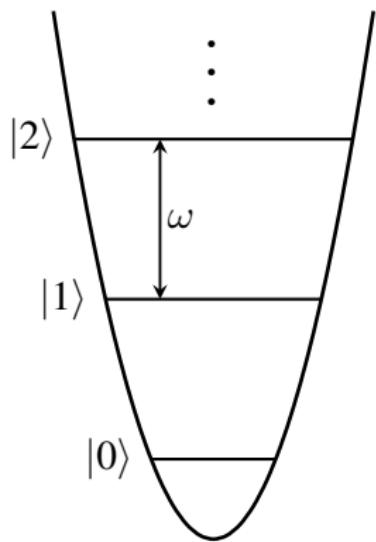
- Raman sideband cooling of Na atoms

## 3 Molecule creation

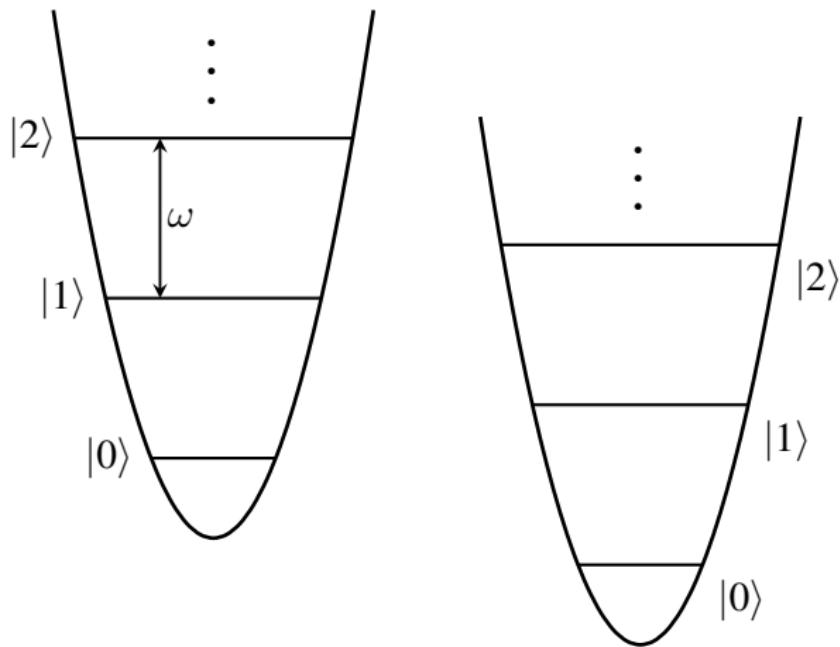
- Atom-atom interaction
- Coherent optical transfer

## 4 Conclusion

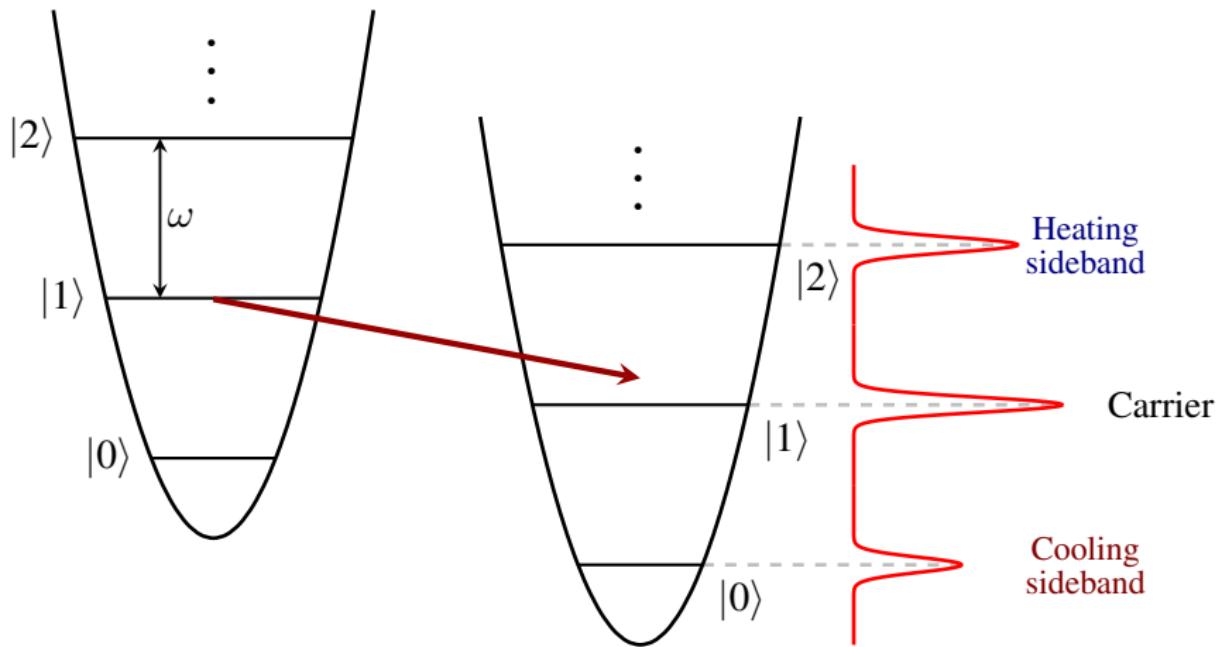
## Raman sideband cooling



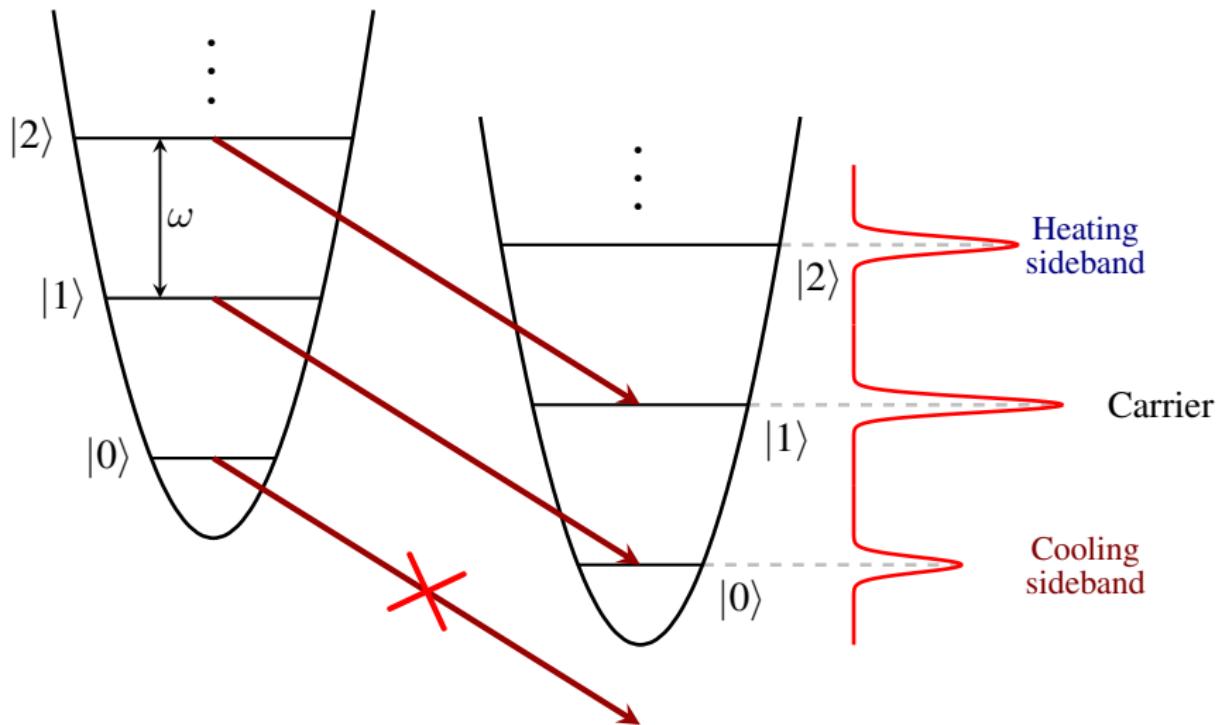
## Raman sideband cooling



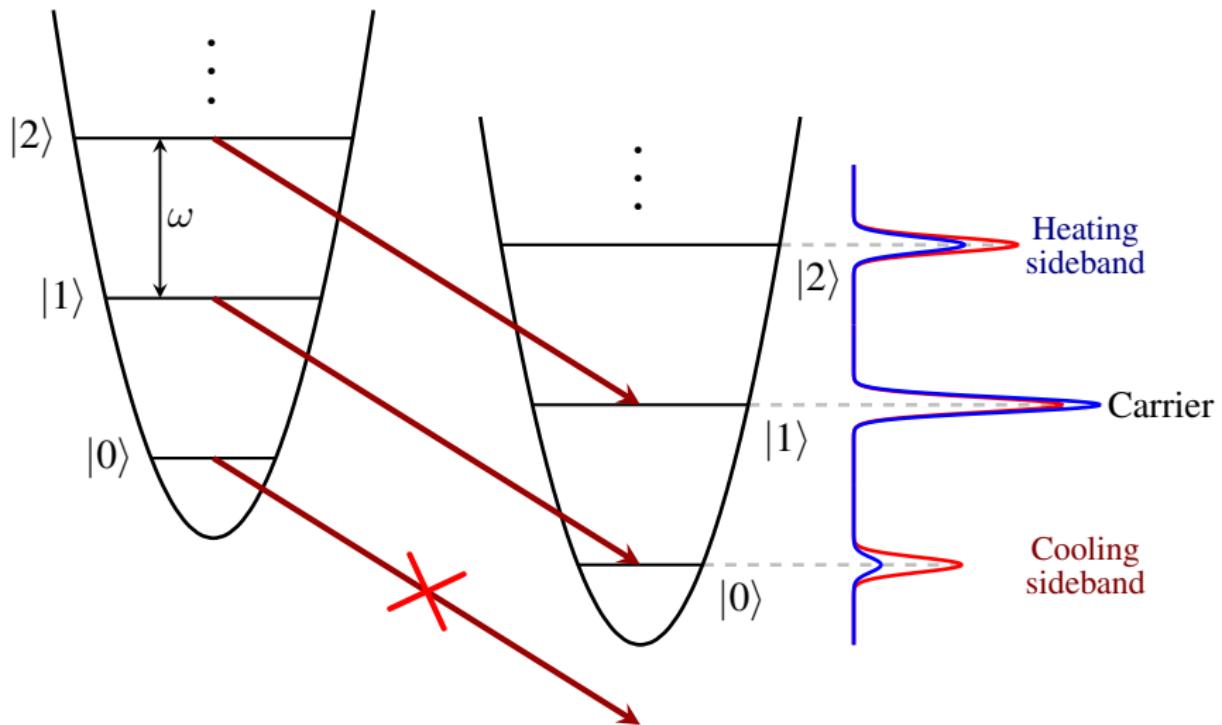
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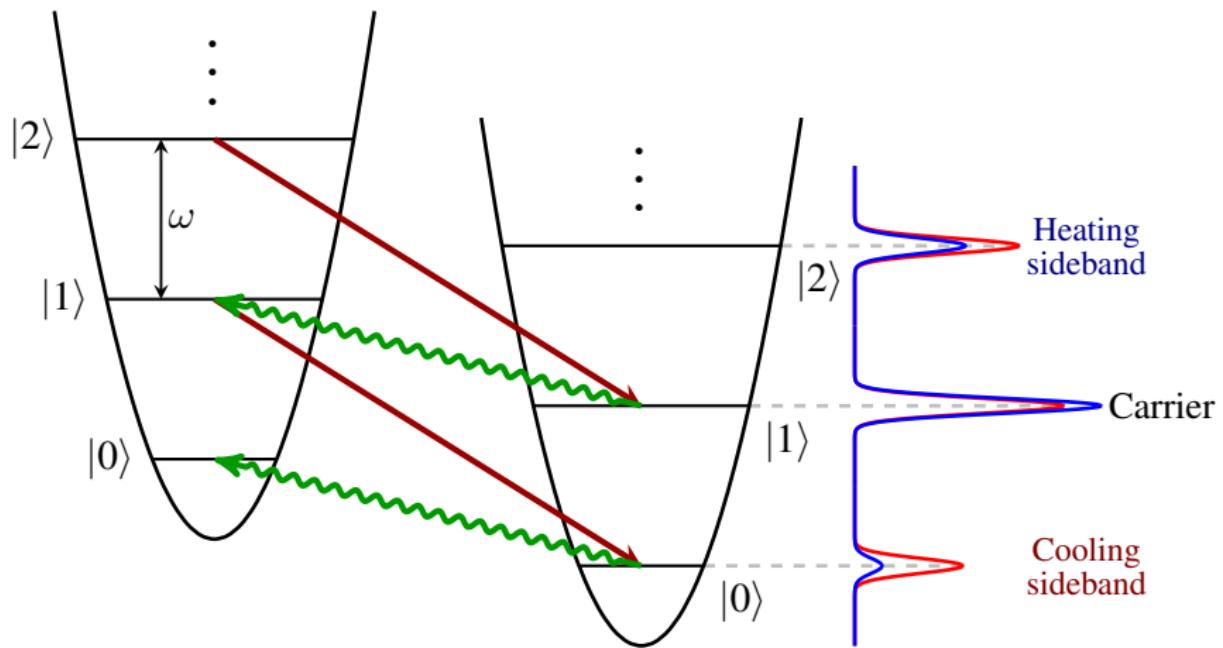
## Raman sideband cooling



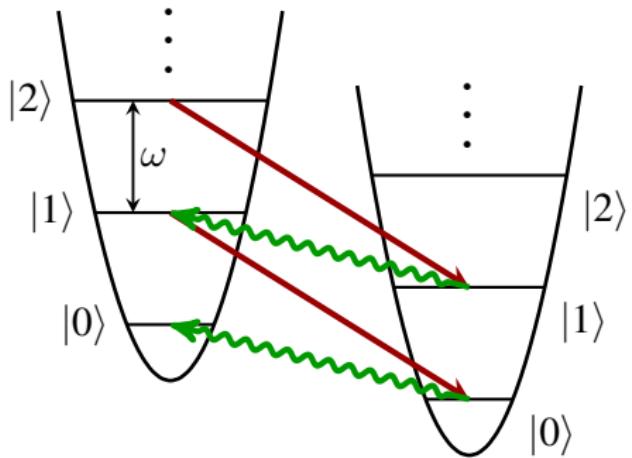
## Raman sideband cooling



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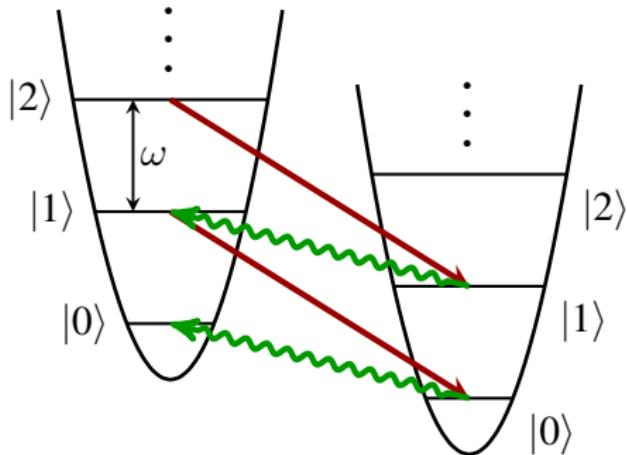
## Raman sideband cooling



## Raman sideband cooling

### Lamb Dicke parameter

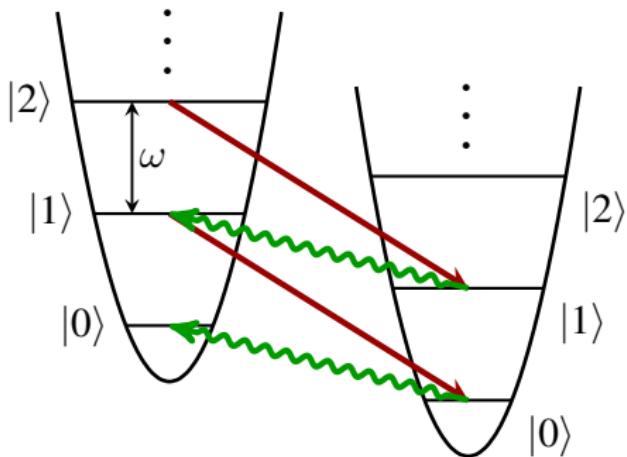
$$\eta \equiv kz_0 = \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}}$$



## Raman sideband cooling

### Lamb Dicke parameter

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

- Motional state branching
- Coupling “dead zone”

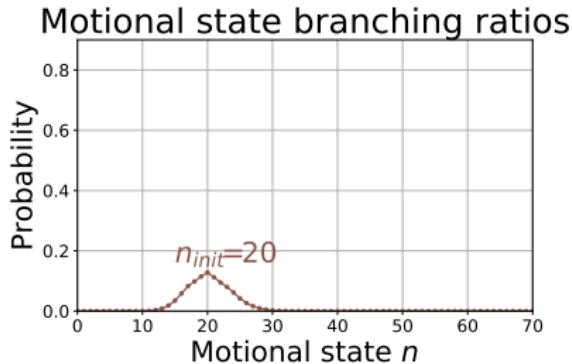
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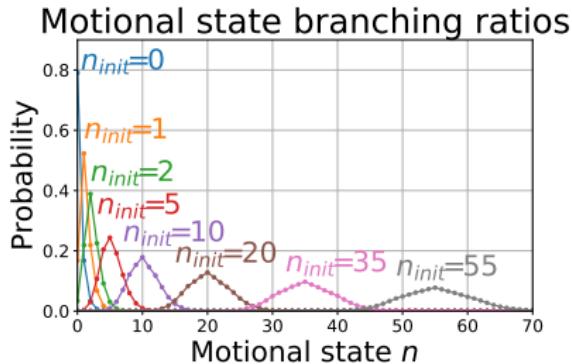
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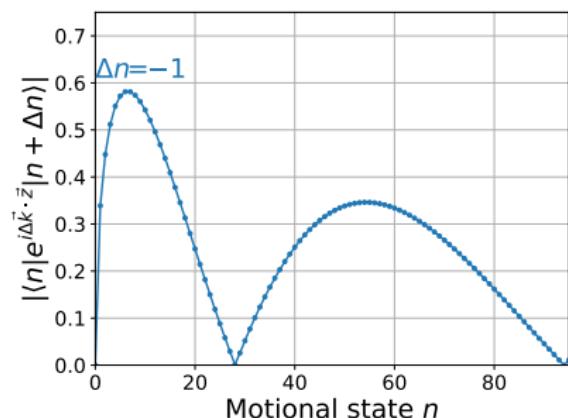
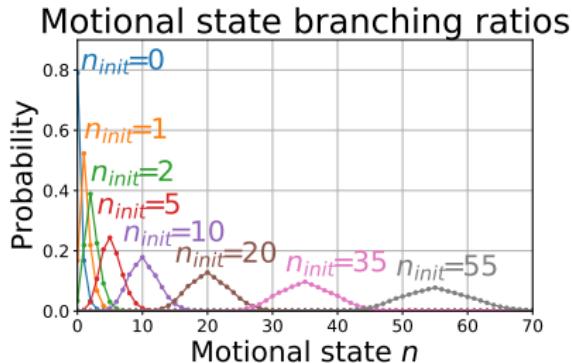
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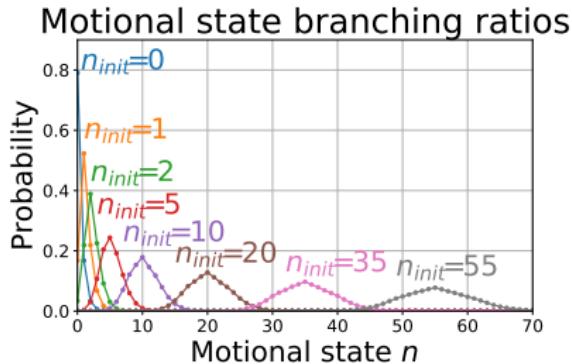
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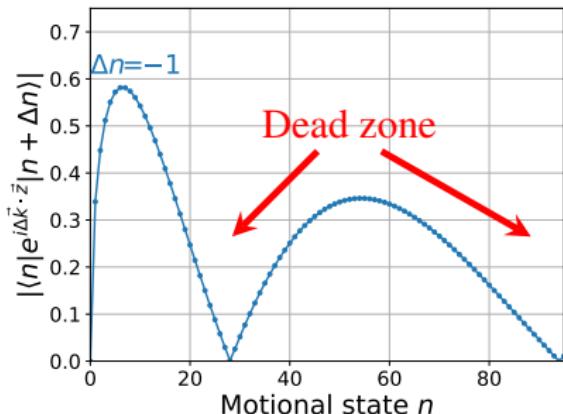


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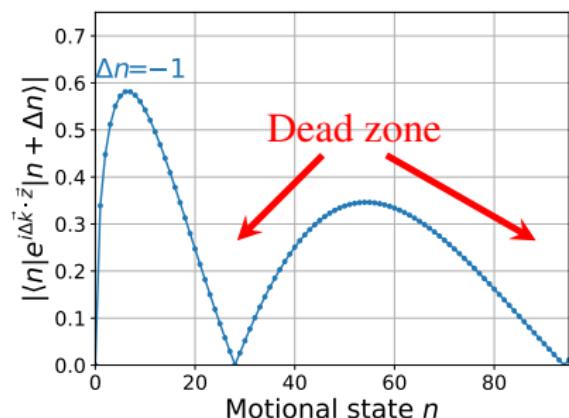
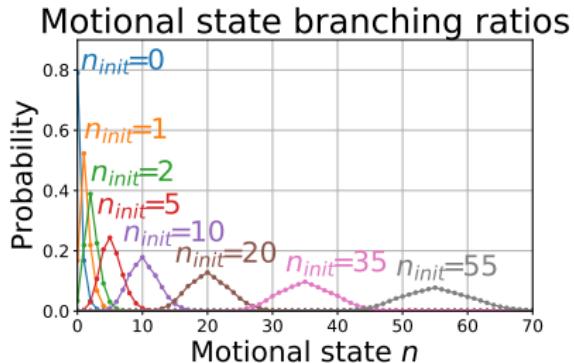
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## Raman sideband cooling



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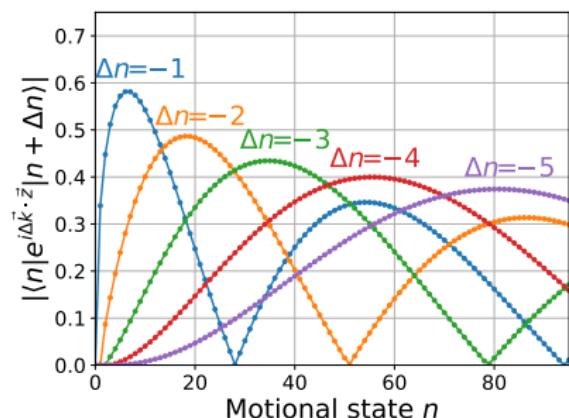
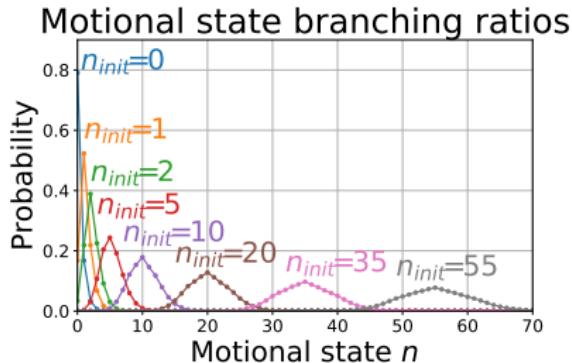
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## Solution

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

## Raman sideband cooling



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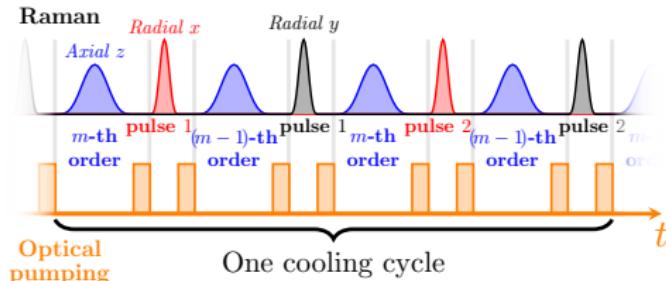
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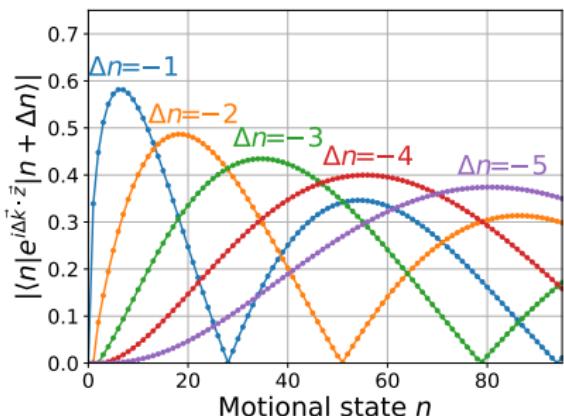
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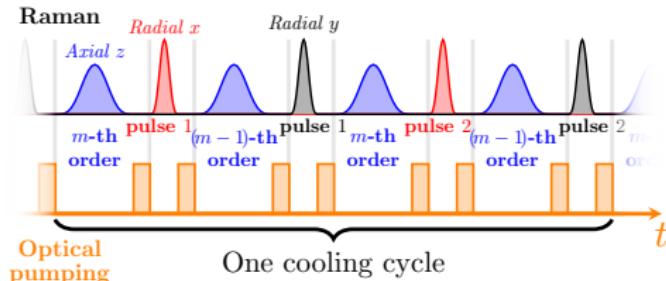
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# Raman sideband cooling



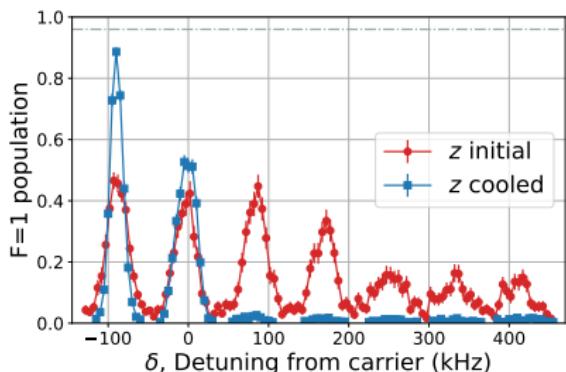
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- Motional state branching
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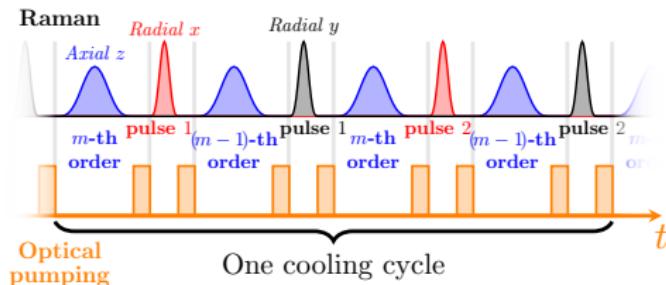
Axial sideband spectrum



## Solution

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

# Raman sideband cooling



## Lamb Dicke parameter

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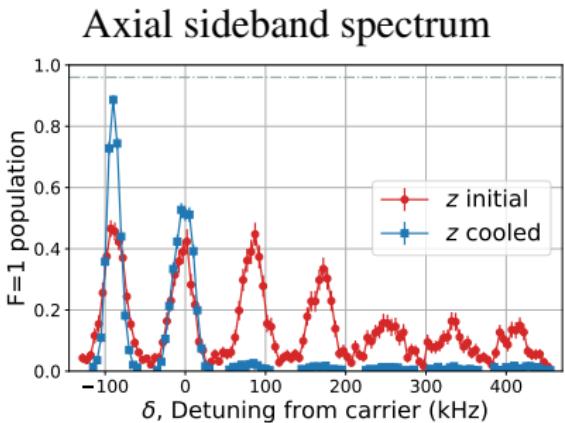
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- Motional state branching
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## Solution

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3D ground state: 93.5(7)%



# Outline

## 1 Experiment overview

## 2 Atom state control

- Raman sideband cooling of Na atoms

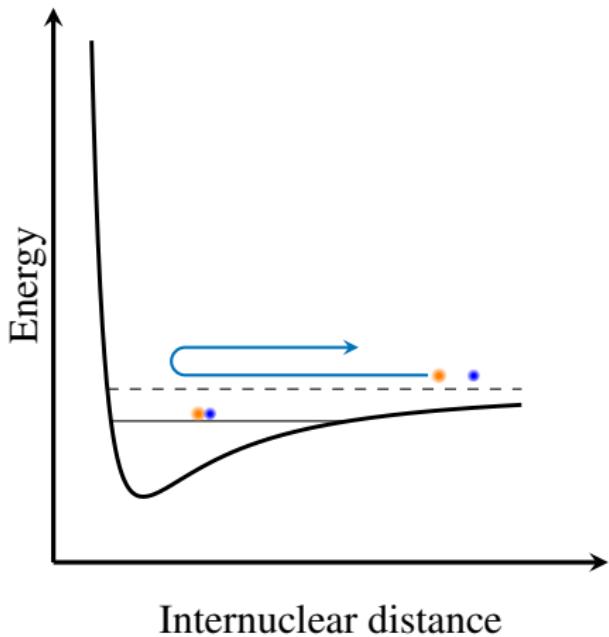
## 3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

## 4 Conclusion

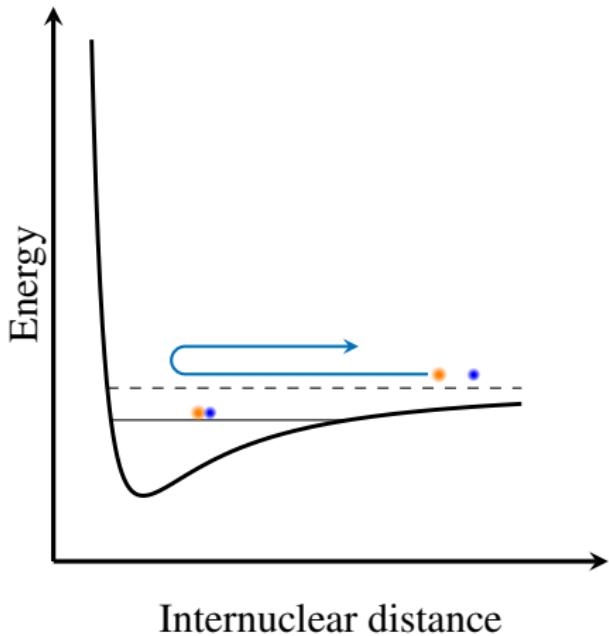
## Scattering length $a$

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



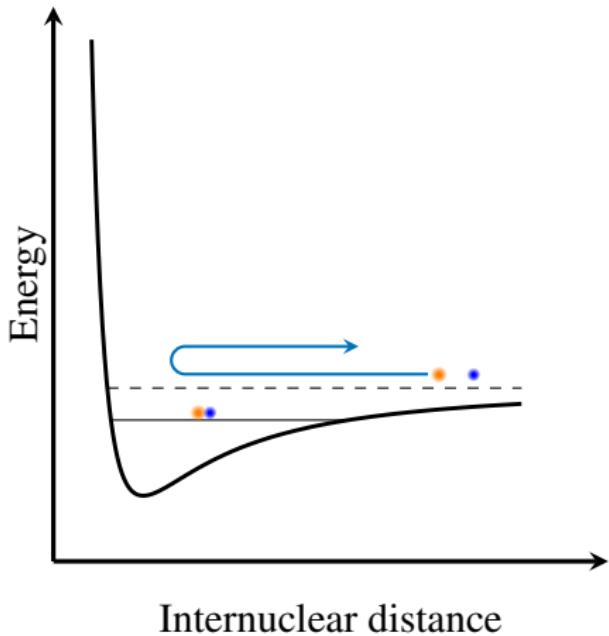
## Scattering length $a$

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



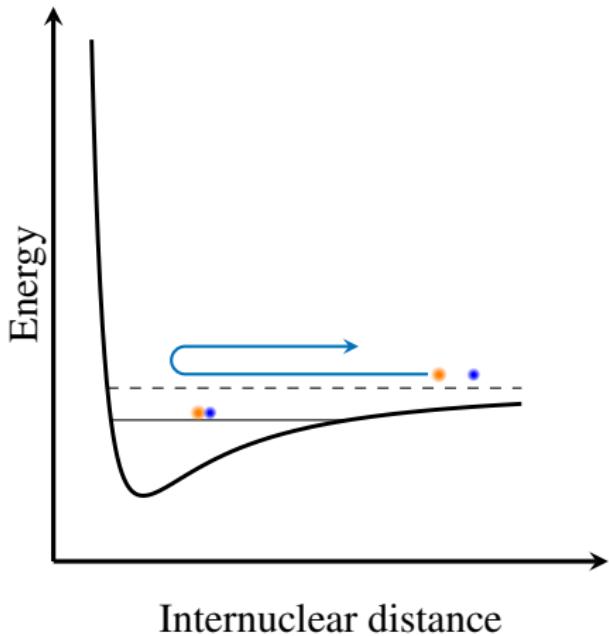
## Scattering length $a$

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



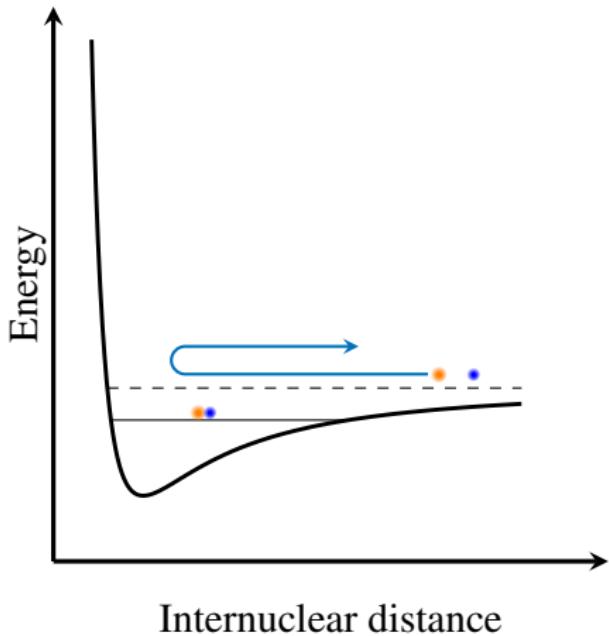
## Scattering length $a$

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

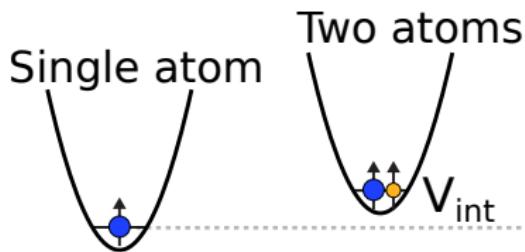


## Scattering length $a$

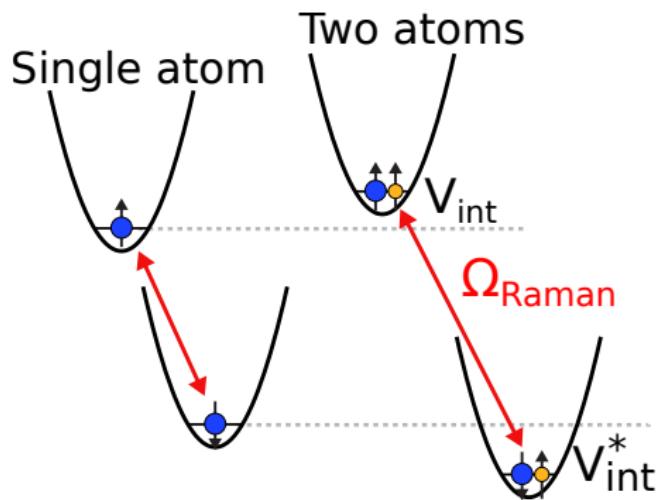
- Binding energy
- Molecular potential
- Molecule formation
- Feshbach resonance
- :



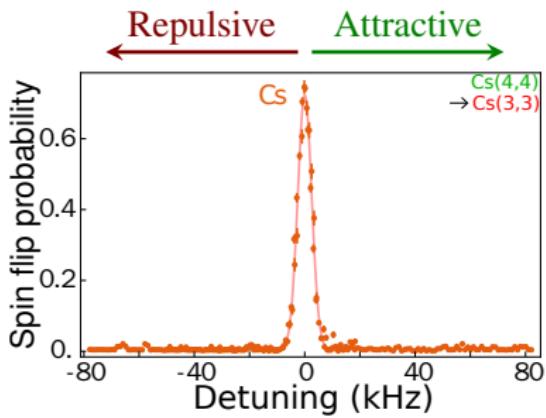
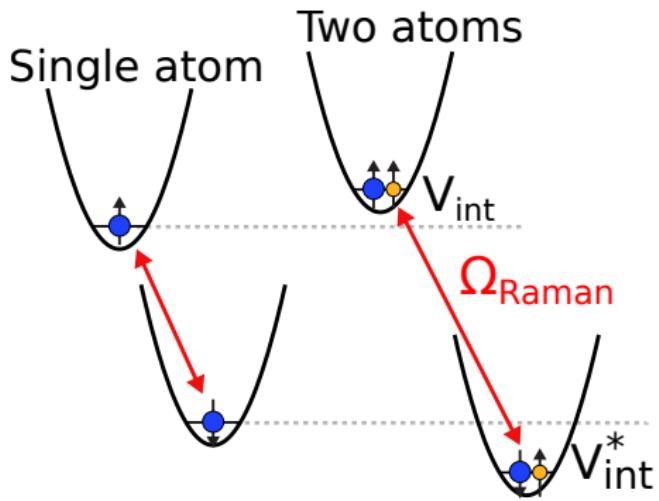
## Interaction shift



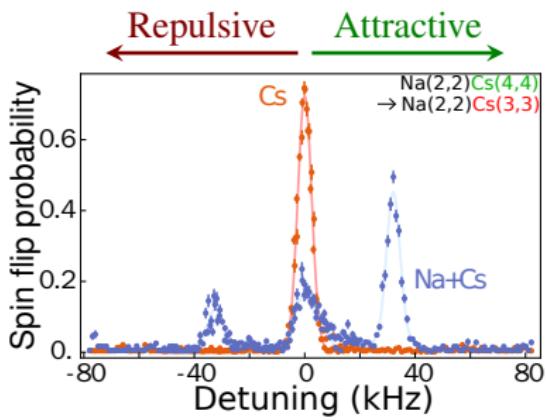
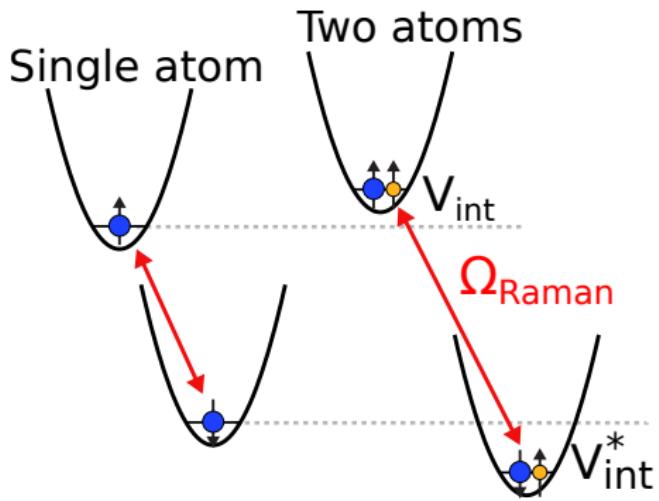
## Interaction shift



## Interaction shift



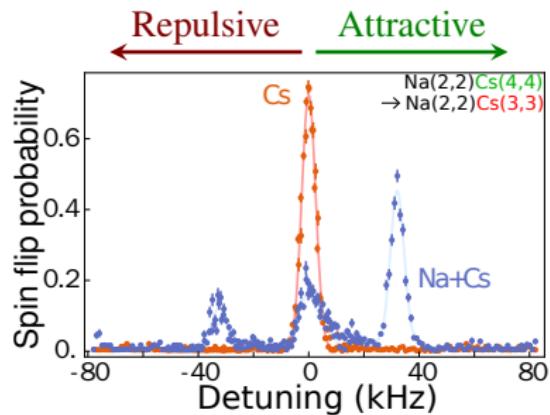
## Interaction shift



## Interaction shift

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

Interaction



## Interaction shift

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

To center of mass  
and relative coordinates

$$M = m_1 + m_2$$

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

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

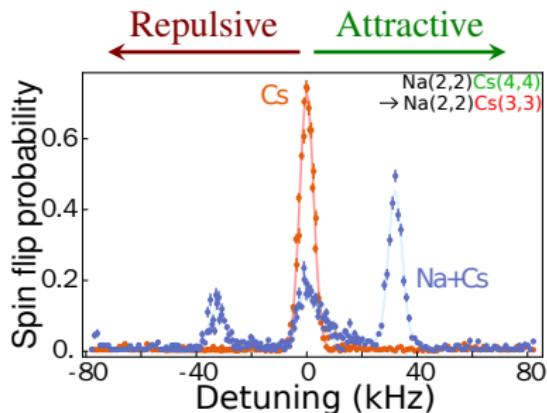
$$\omega_{R,i}^2 = \frac{m_2 \omega_{1,i}^2 + m_1 \omega_{2,i}^2}{m_1 + m_2}$$

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

$$x_{R,i} = x_{1,i} - x_{2,i}$$

$$P_i = p_{1,i} + p_{2,i}$$

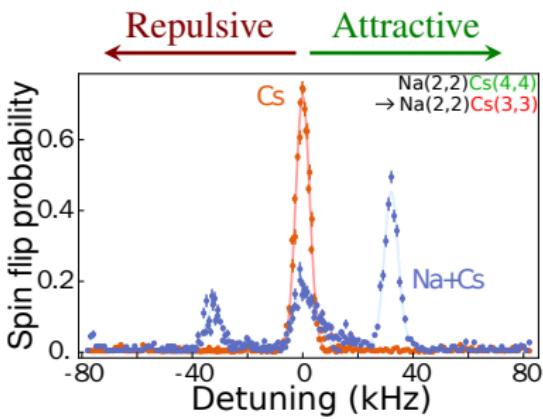
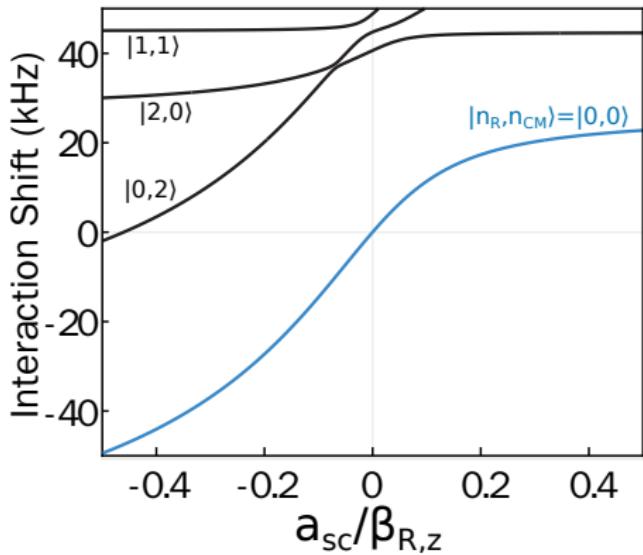
$$p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$



Center of mass

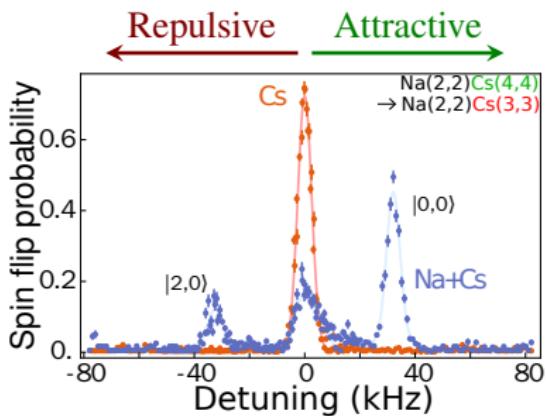
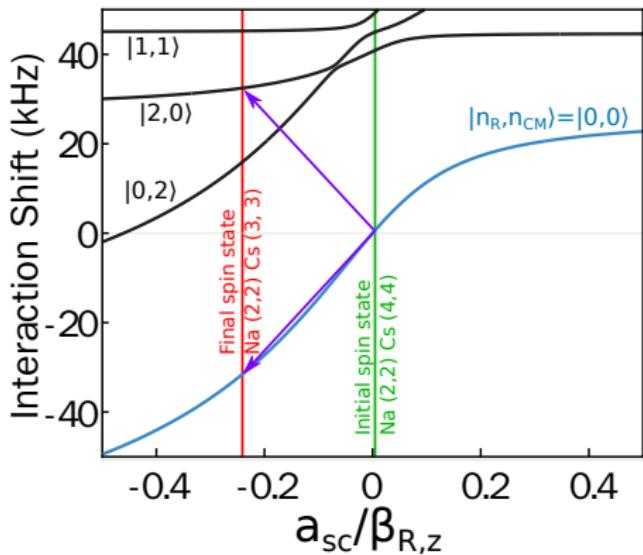
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M \Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu \omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu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

# Interaction shift



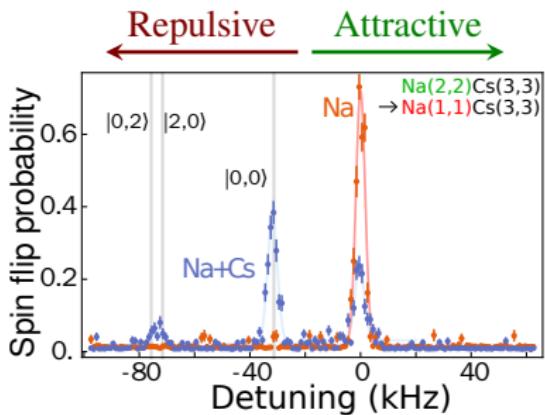
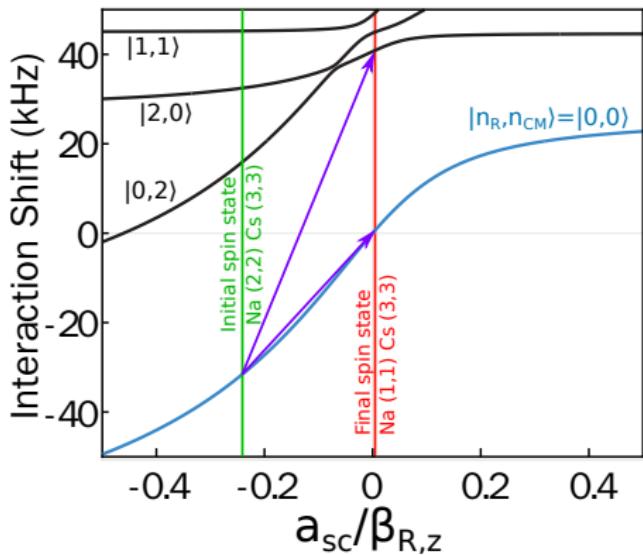
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

# Interaction shift



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

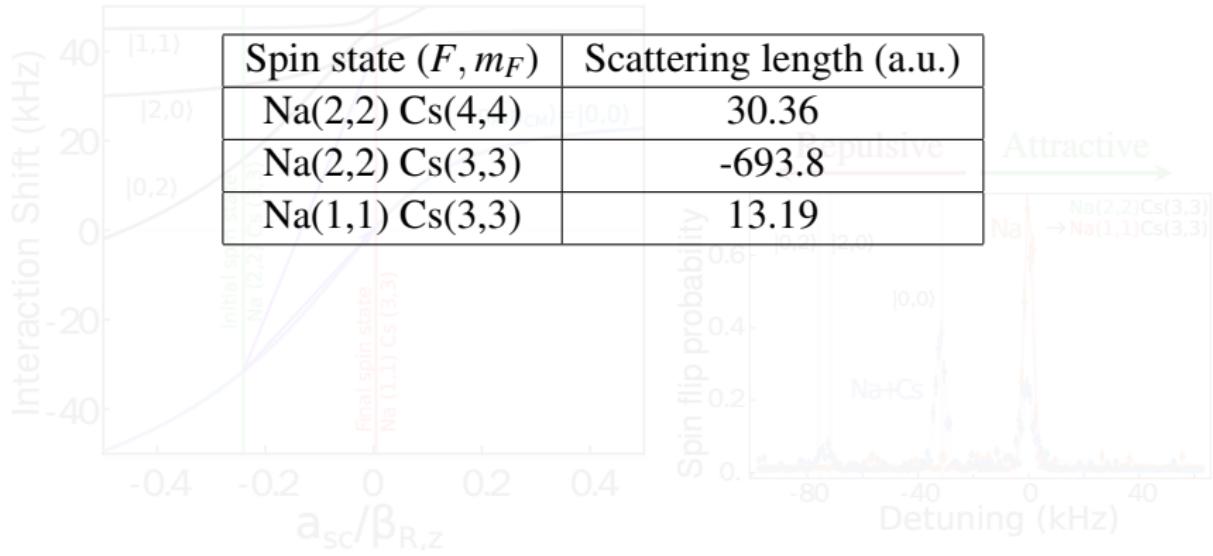
# Interaction shift



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

# Interaction shift

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



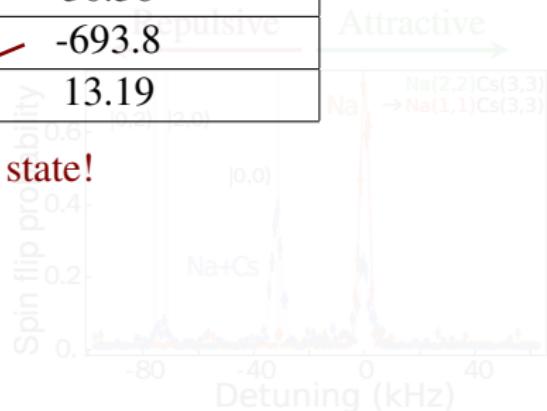
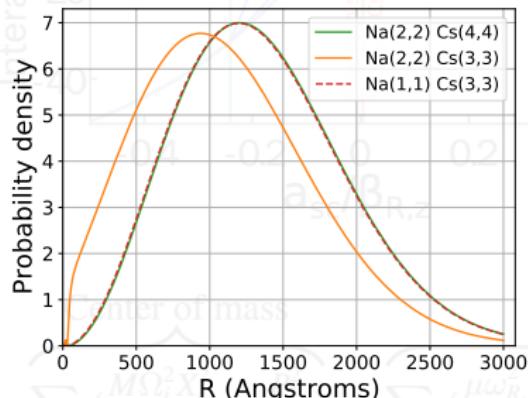
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

# Interaction shift

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

Spin state ( $F, m_F$ )	Scattering length (a.u.)
Na(2,2) Cs(4,4)	30.36
Na(2,2) Cs(3,3)	-693.8
Na(1,1) Cs(3,3)	13.19

Enhanced coupling to molecular state!



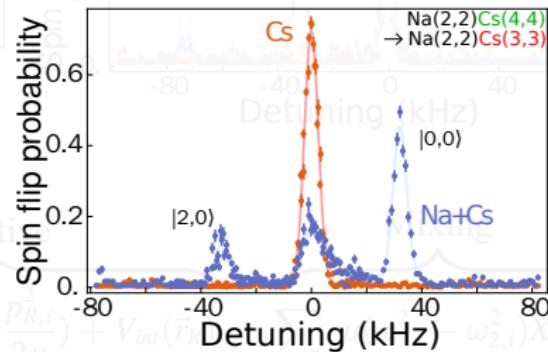
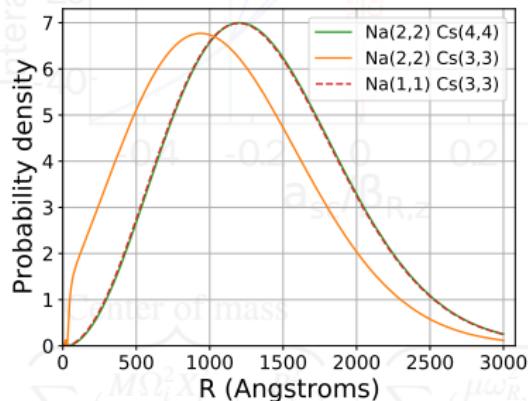
$$H = \sum_{i=x,y,z} \left( \frac{\mu \omega_{R,i} X_{R,i}}{2M} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R) + \sum_{i=x,y,z} \mu (\omega_{1,i}^2 - \omega_{2,i}^2) X_i X_{R,i}$$

# Interaction shift

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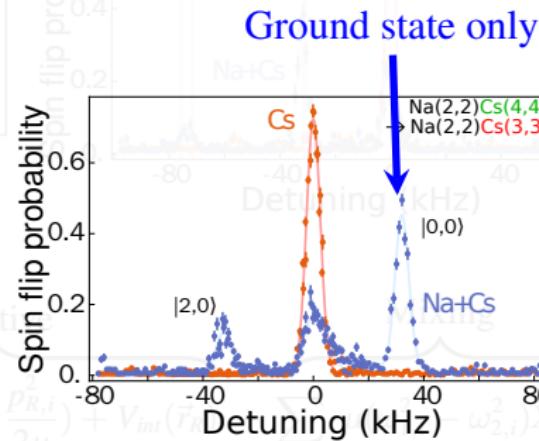
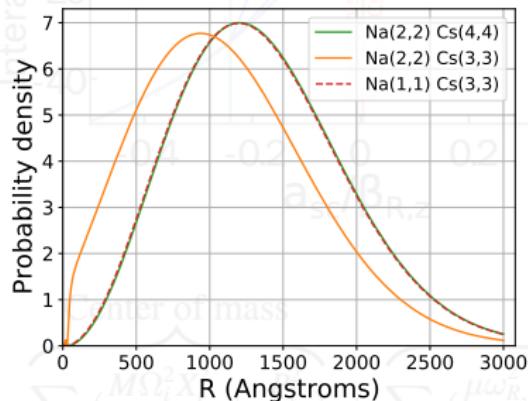


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Enhanced coupling to molecular state!



# Outline

## 1 Experiment overview

## 2 Atom state control

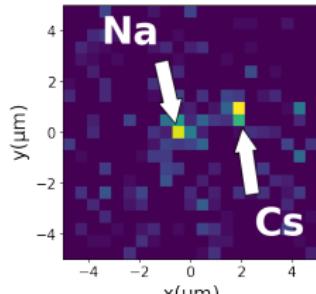
- Raman sideband cooling of Na atoms

## 3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

## 4 Conclusion

## Loading

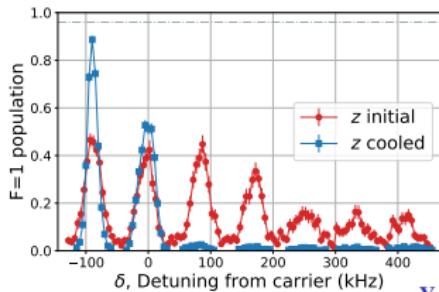


NJP. 19, 023007 (2017)

## Merging



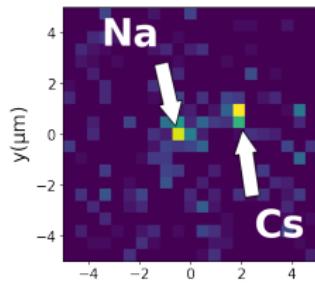
## Cooling



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

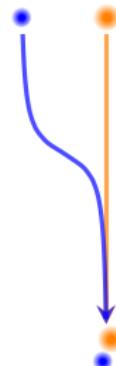
Y. Yu et al. PRA. 97, 063423 (2018)

## Loading

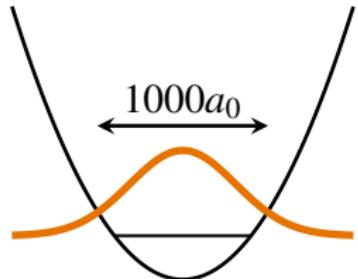


NJP. 19, 023007 (2017)

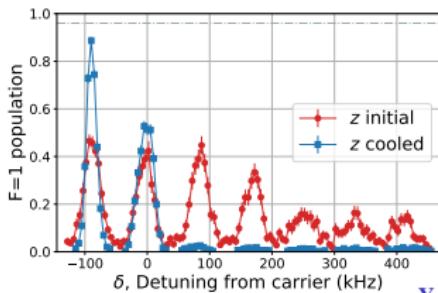
## Merging



## Atom



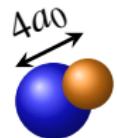
## Cooling

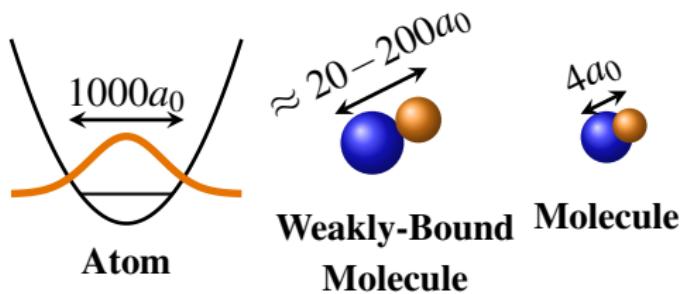


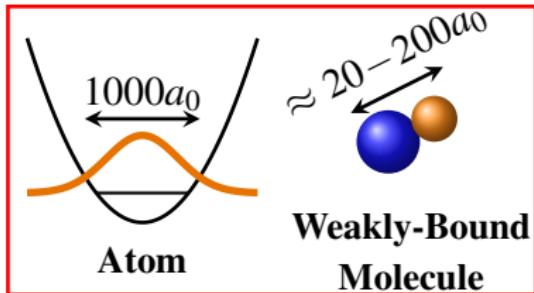
Y. Yu et al. PRA. 97, 063423 (2018)

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

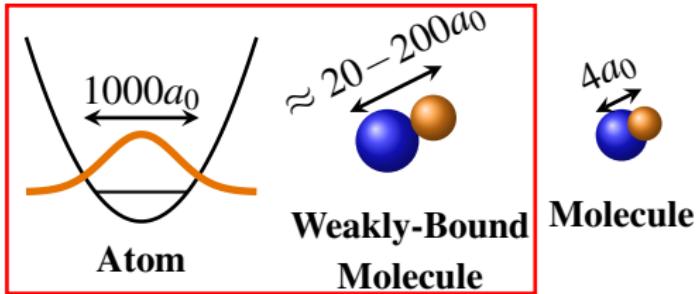
## Molecule



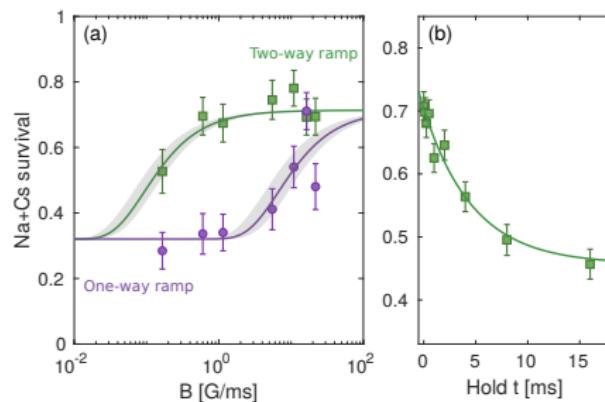




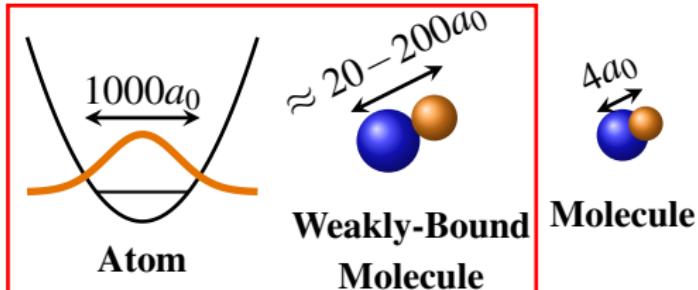
**Molecule**



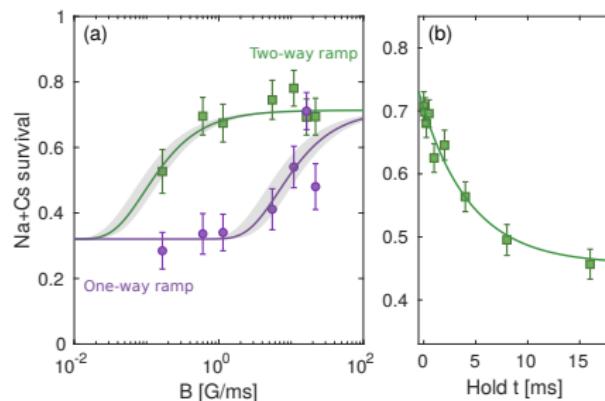
## Feshbach molecule



PRL. 124, 253401 (2020)

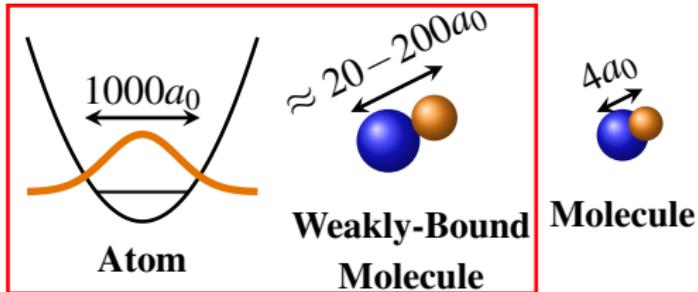


## Feshbach molecule



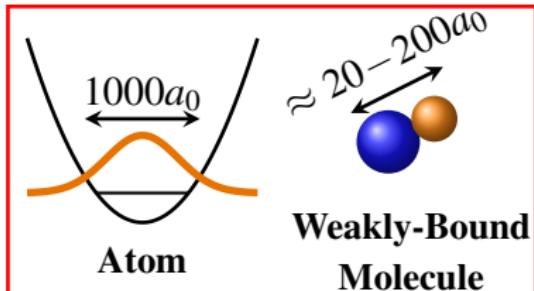
- Requires Feshbach resonance
- Usually large magnetic field

PRL. 124, 253401 (2020)



## Optical transfer

- More general
- Faster

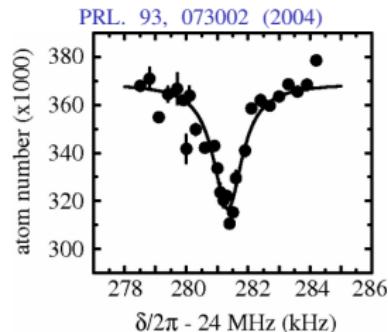


## Optical transfer

- More general
- Faster

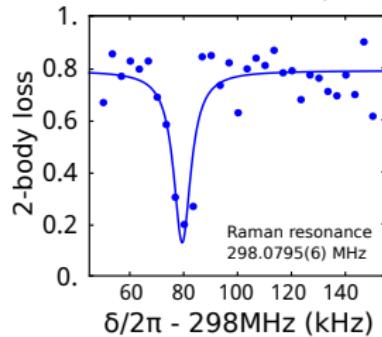
## Previous results

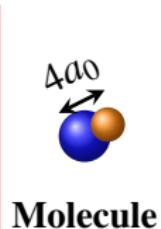
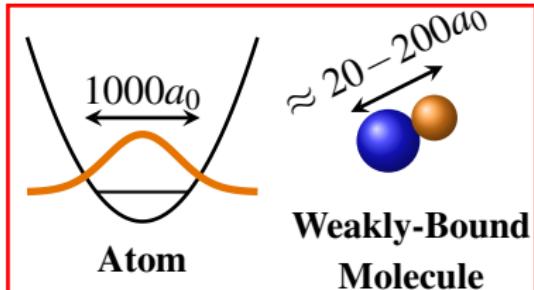
$\text{Rb}_2$  Science 287, 1016 (2000)



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

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





## Optical transfer

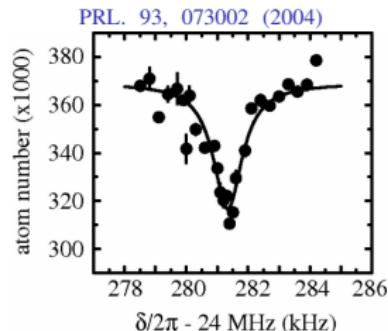
- More general
- Faster

### Limitations so far

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

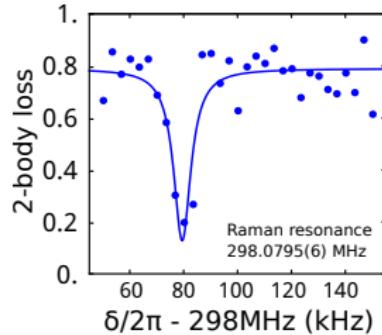
## Previous results

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

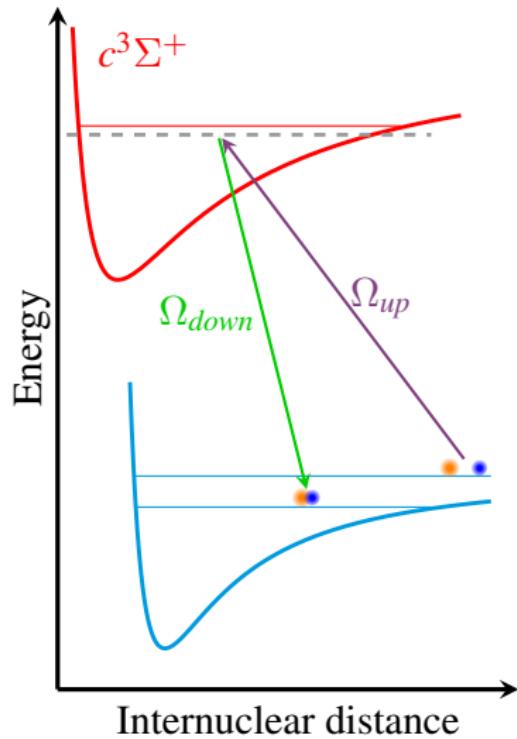


$\text{Sr}_2$  PRL. 109, 115302 (2012)

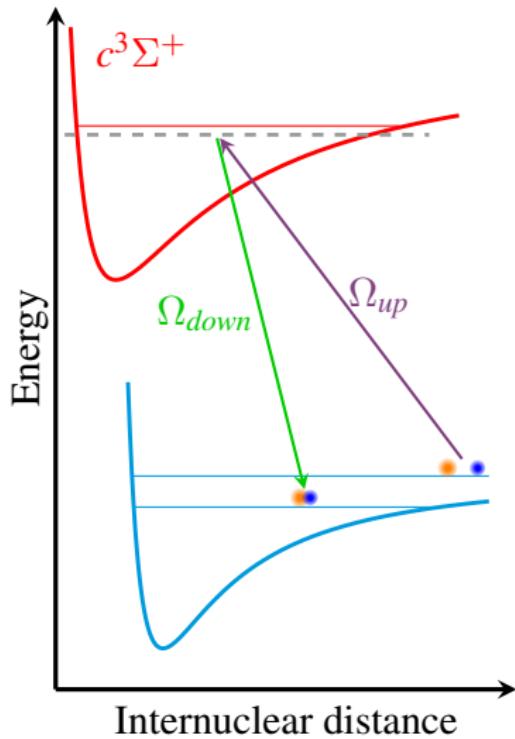
$\text{NaCs}$  Y. Yu et al. PRX. 9, 021039 (2019)



# Raman transfer



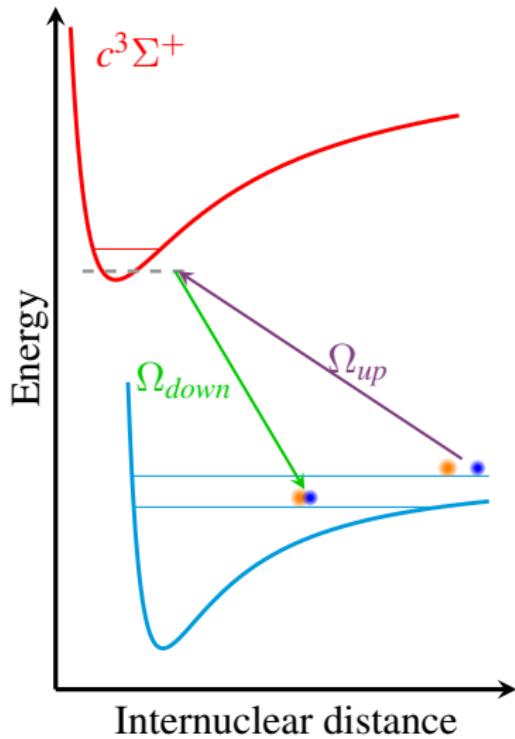
# Raman transfer



## Near threshold states

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

# Raman transfer



## Near threshold states

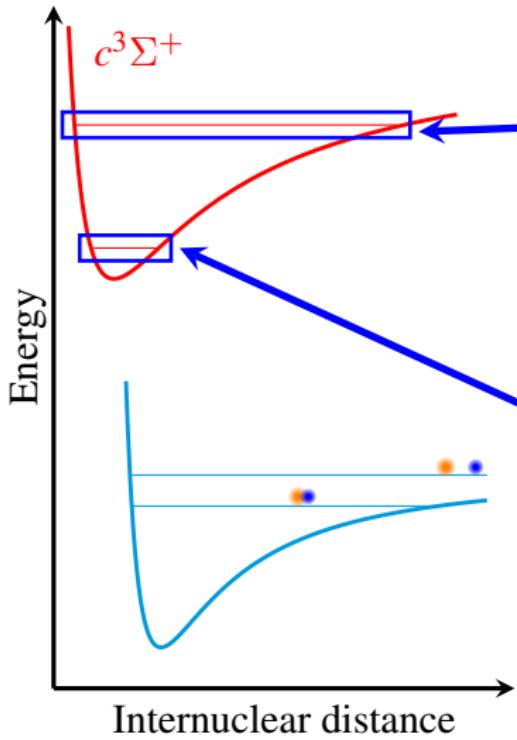
- Stronger coupling ( $\Omega_{up}$  and  $\Omega_{down}$ )
- Closely spaced
- Fast scattering

## Deeply bound states

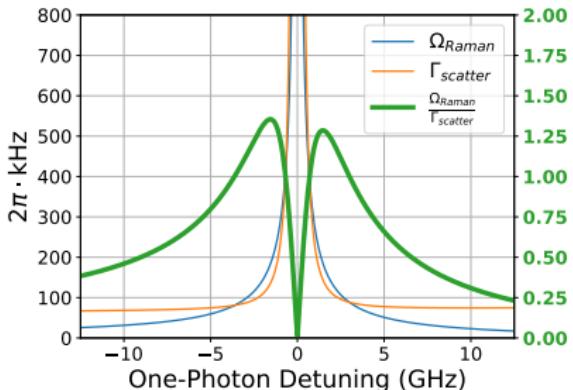
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

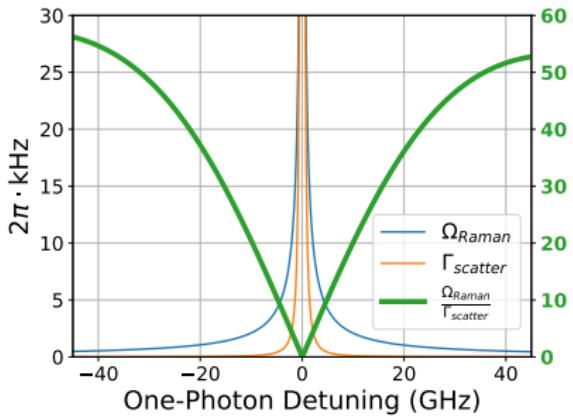
## Raman transfer



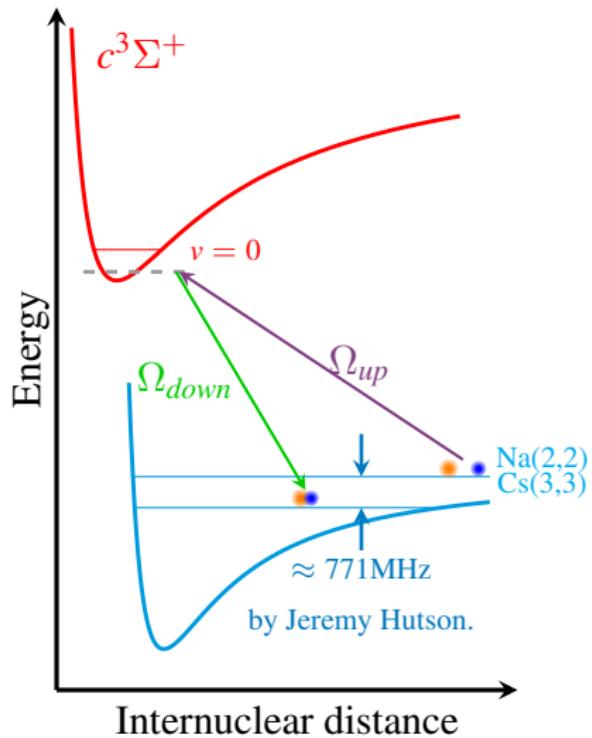
## Near threshold states



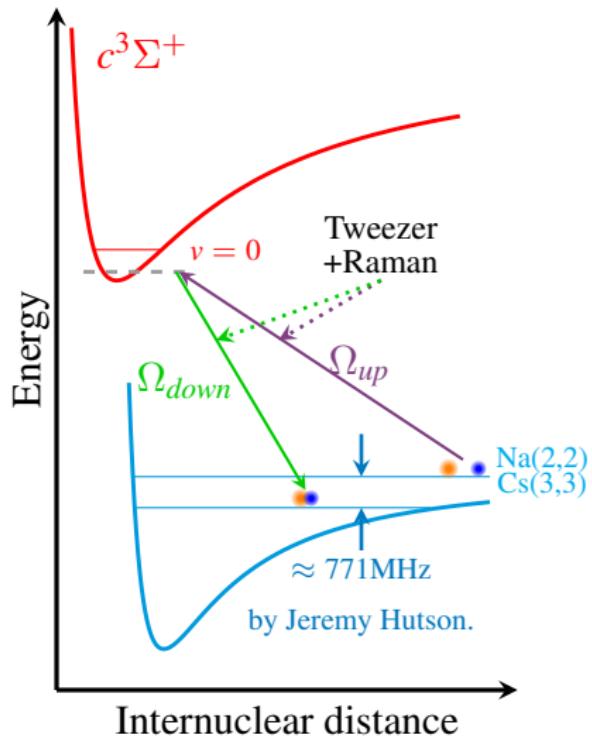
## Deeply bound states



# Experiment



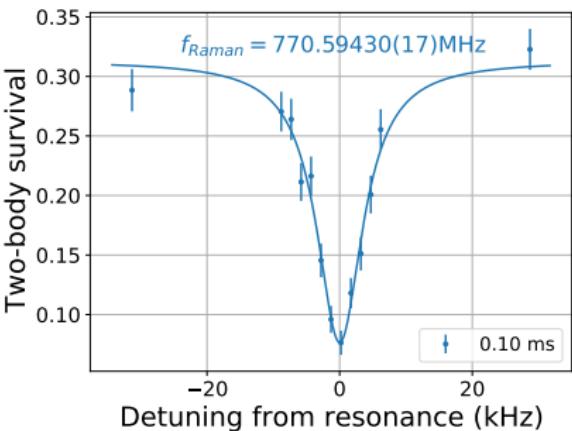
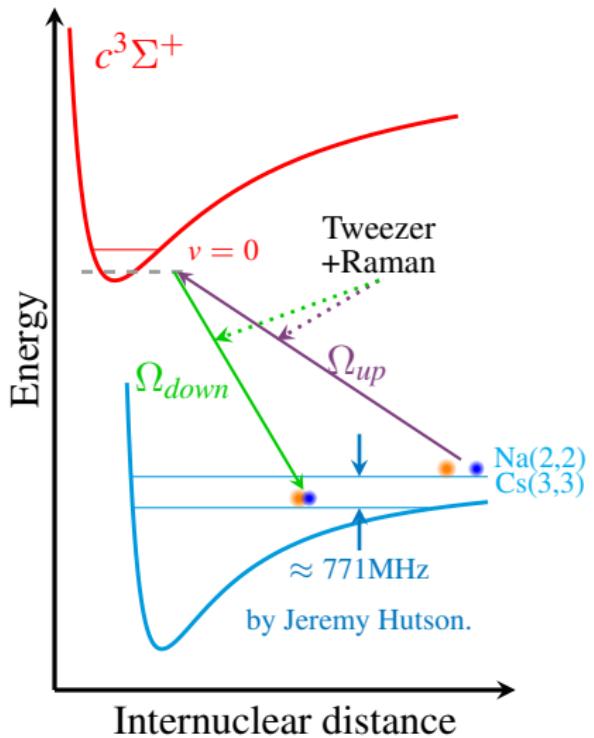
# Experiment



## Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

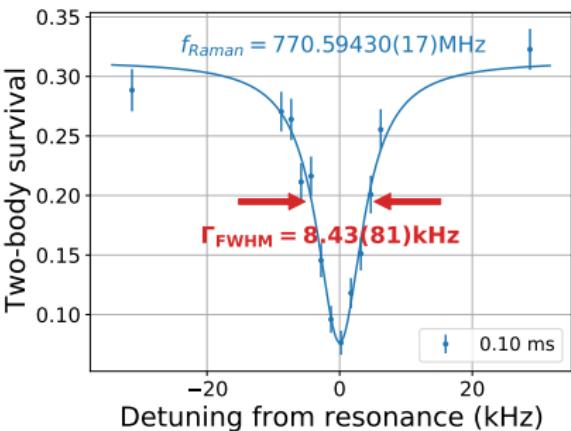
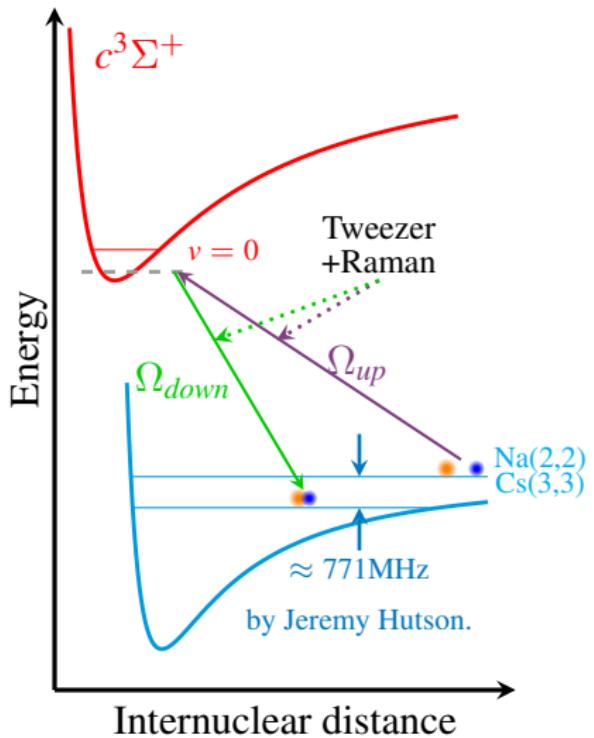
# Experiment



## Tweezer as Raman beam

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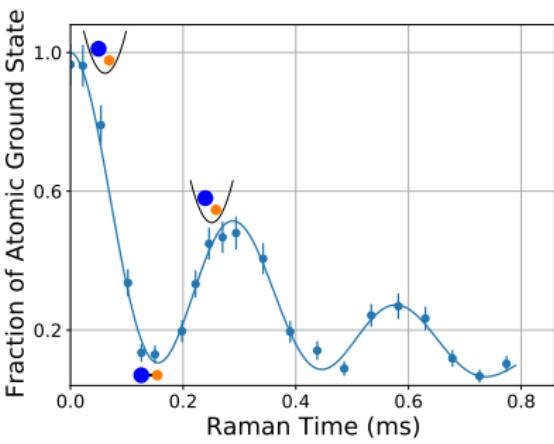
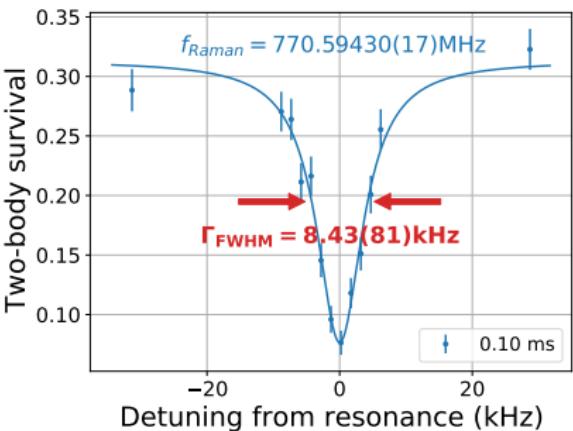
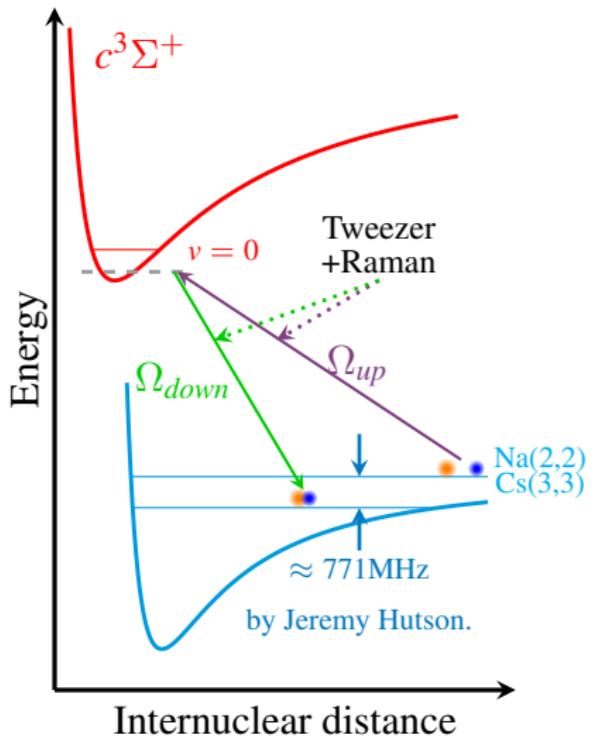
# Experiment



## Tweezer as Raman beam

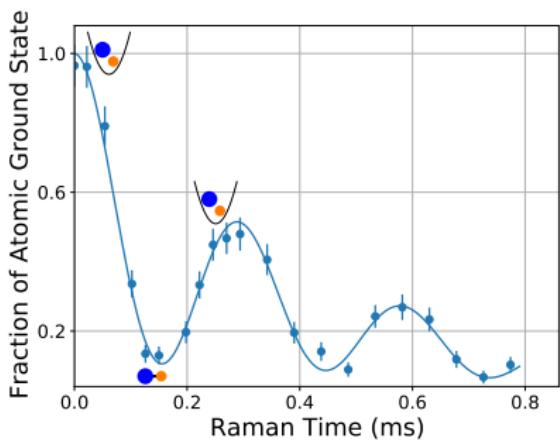
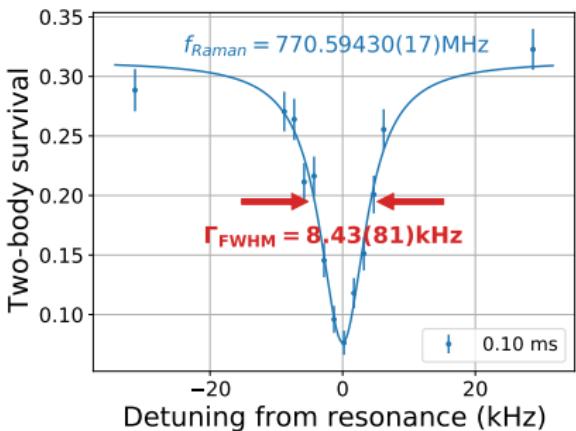
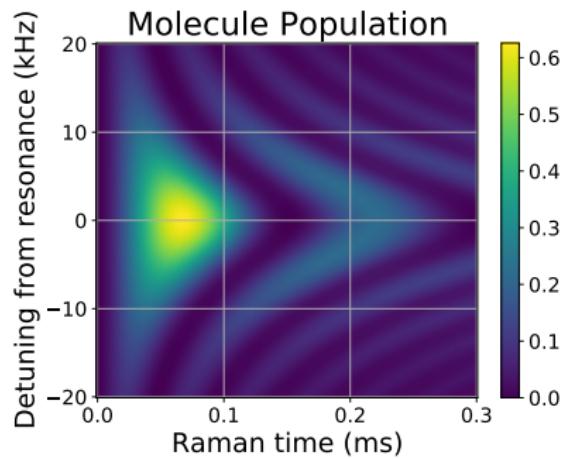
- Higher Raman Rabi frequency
- Lower scattering from other sources

# Experiment



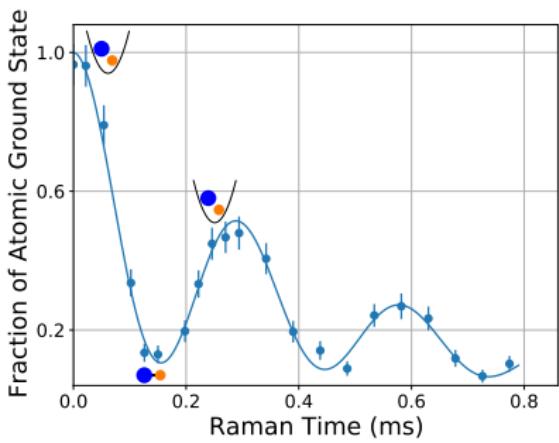
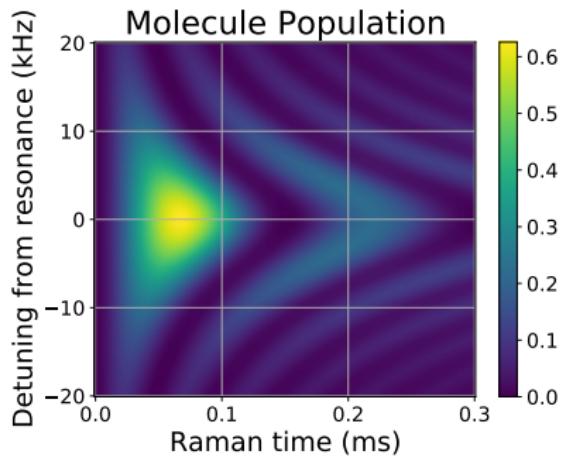
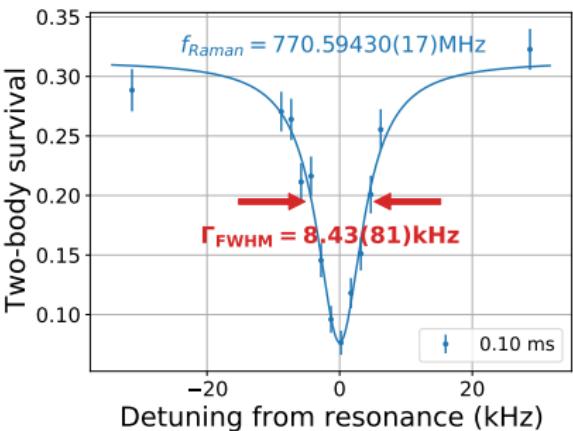
# Experiment

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



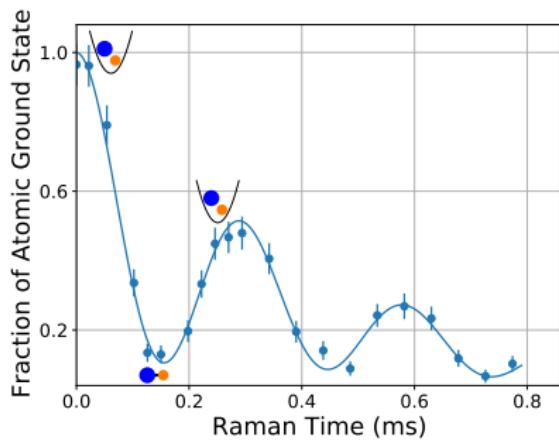
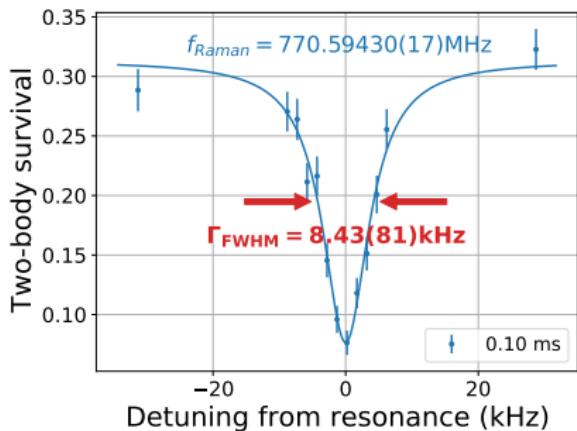
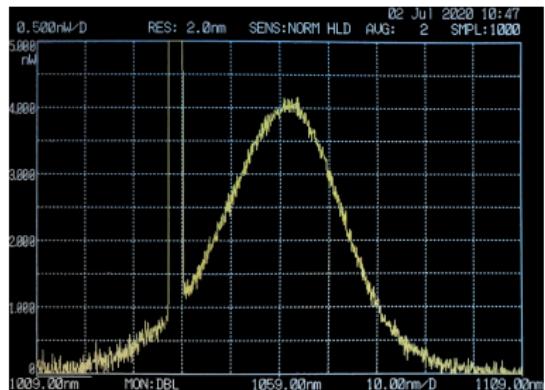
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- Full quantum control of atoms in optical tweezers
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- Coherent all-optical creation of single molecule
- Working towards fully controlled, strongly interacting tweezer array

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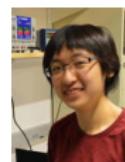
### Experiment



Kang-Kuen Ni



Kenneth  
Wang



Jessie  
Zhang



Lewis  
Picard



William  
Cairncross



Lee Liu  
Postdoc @JILA



Jonathan Hood  
Asstn Prof @Purdue



Nick Hutzler  
Asstn Prof @Caltech

### Theory



Jeremy Hutson



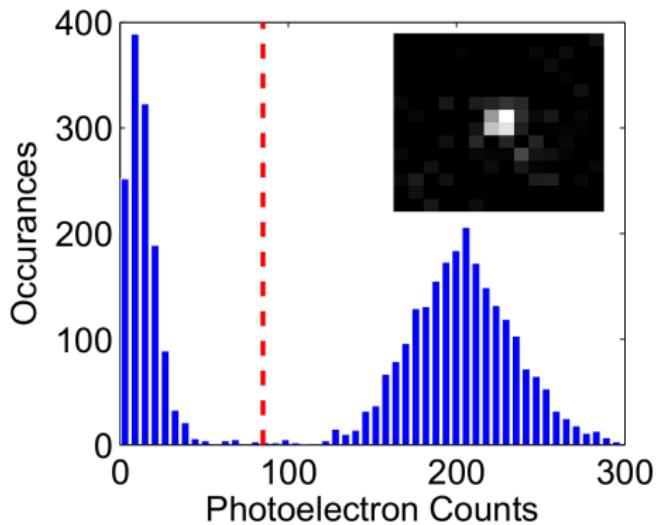


## Single Atom in Tweezer

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

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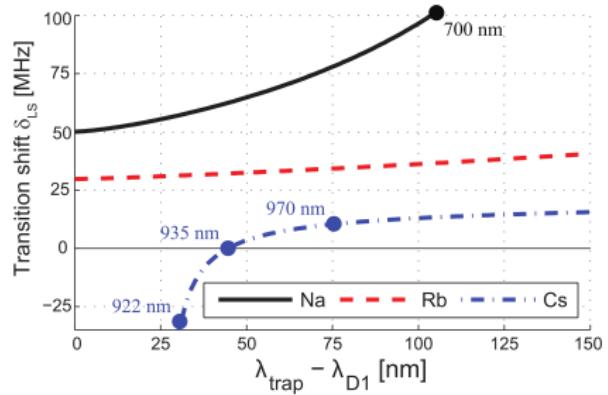
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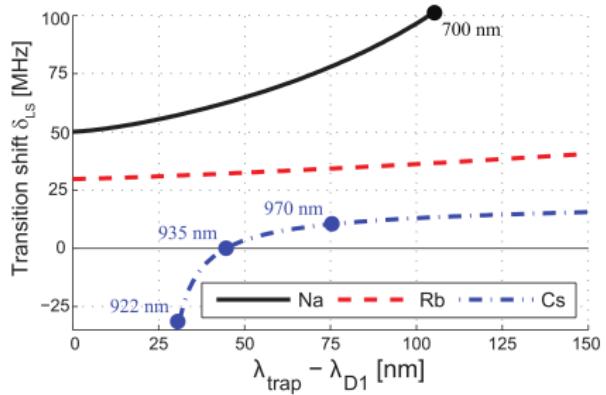
## Issues with Na

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

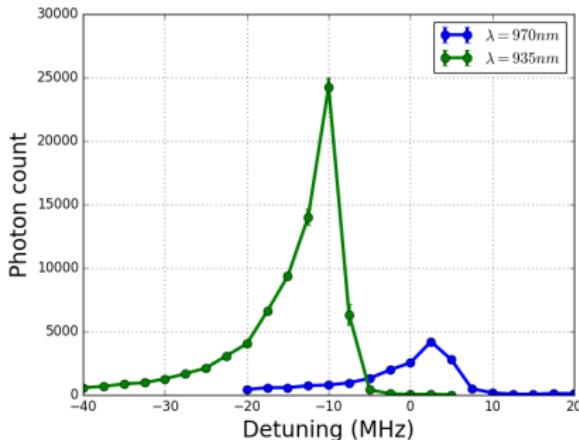
## Real Issue with Na: Light Shift



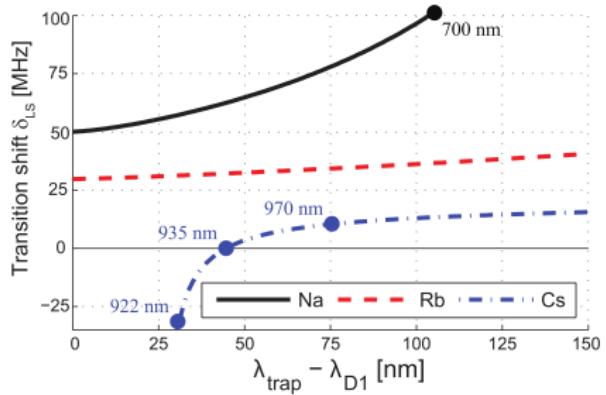
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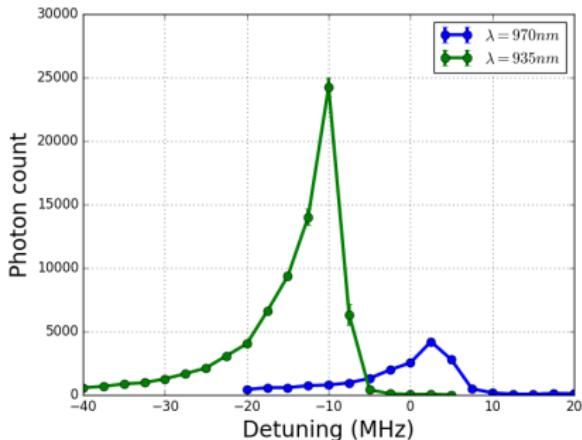
## Cs single atom imaging



## Real Issue with Na: Light Shift



## Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

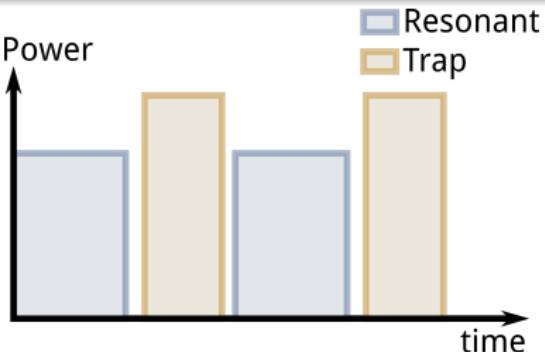
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### Trap modulation

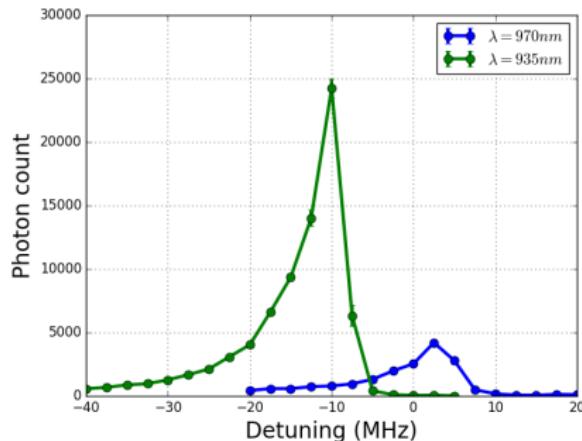
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



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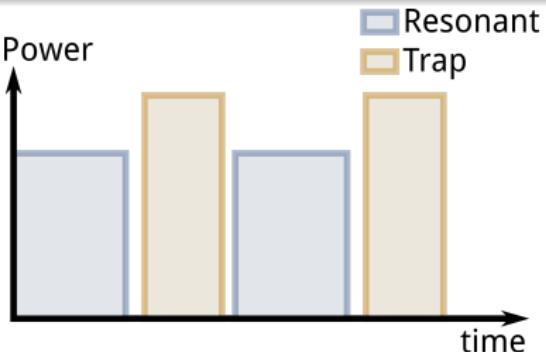
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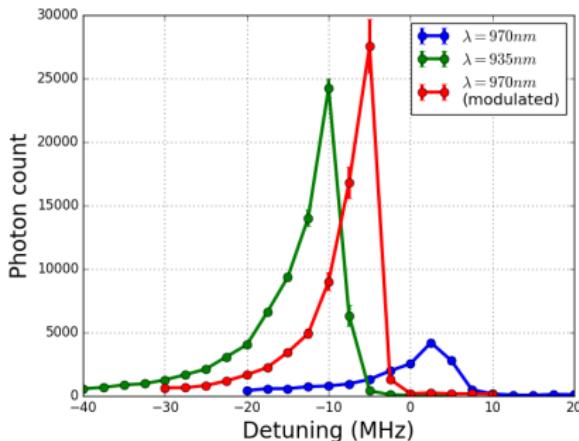
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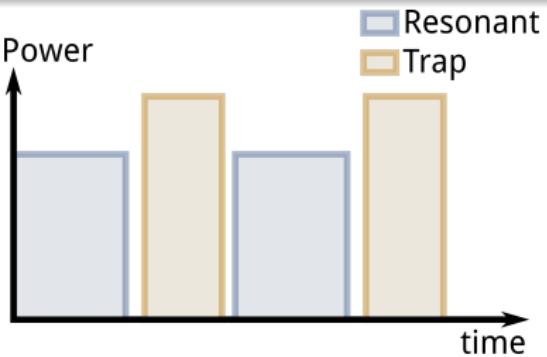
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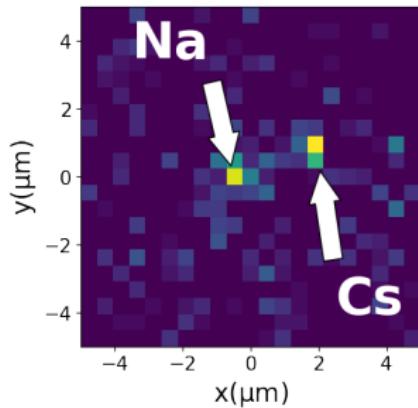
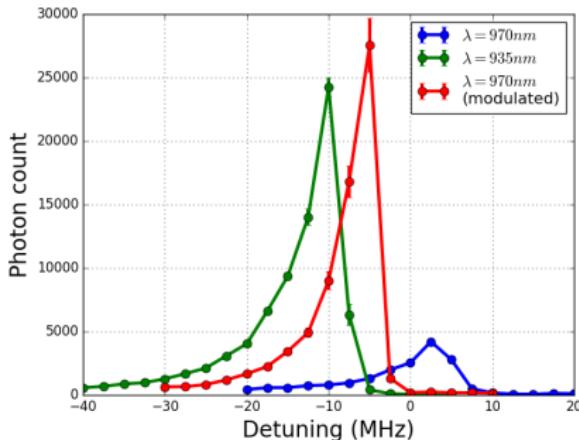
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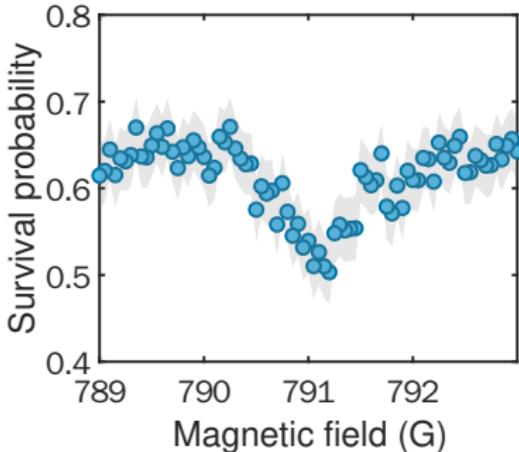
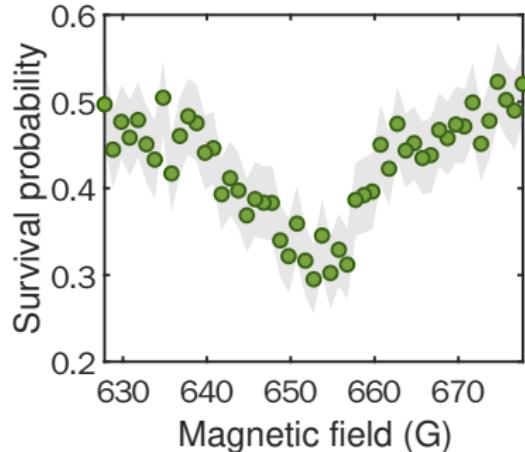
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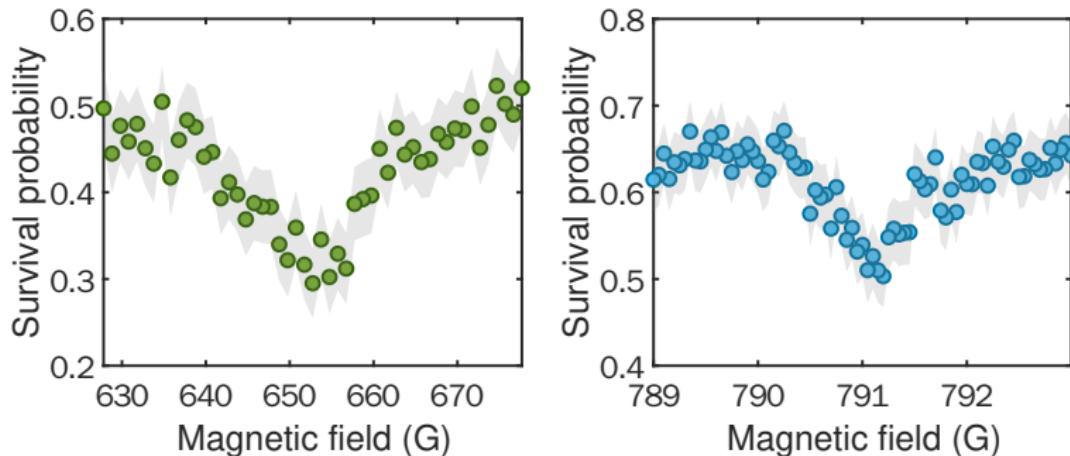
## Na (1, -1) Cs (3, -3) Feshbach resonance



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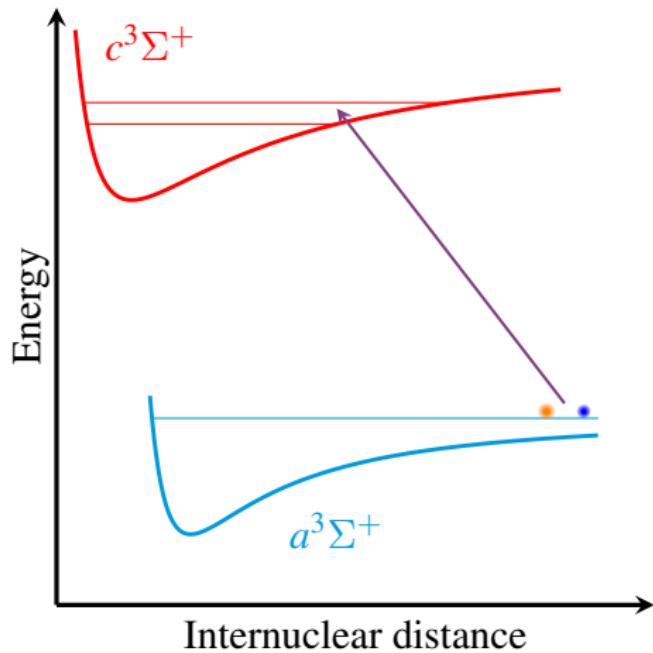
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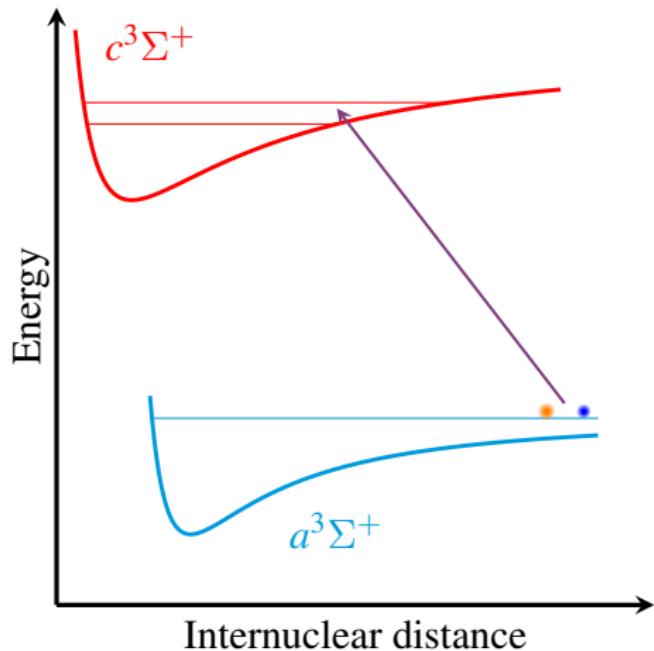
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) <sup>1</sup>	663 G	799 G
Measured	652(3) G	791.2(2) G

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

# Photoassociation (PA) Spectroscopy



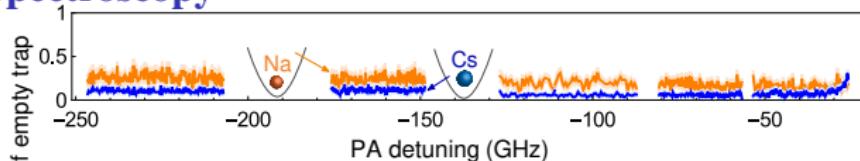
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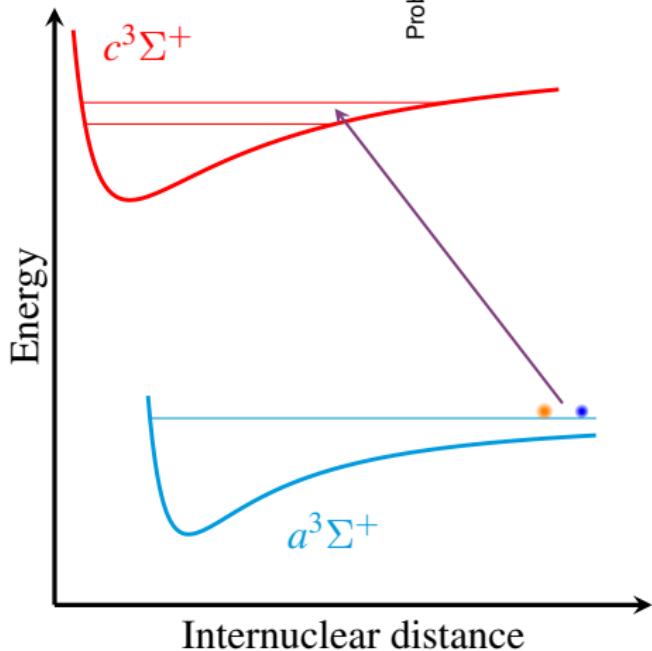
## Single Atom PA

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

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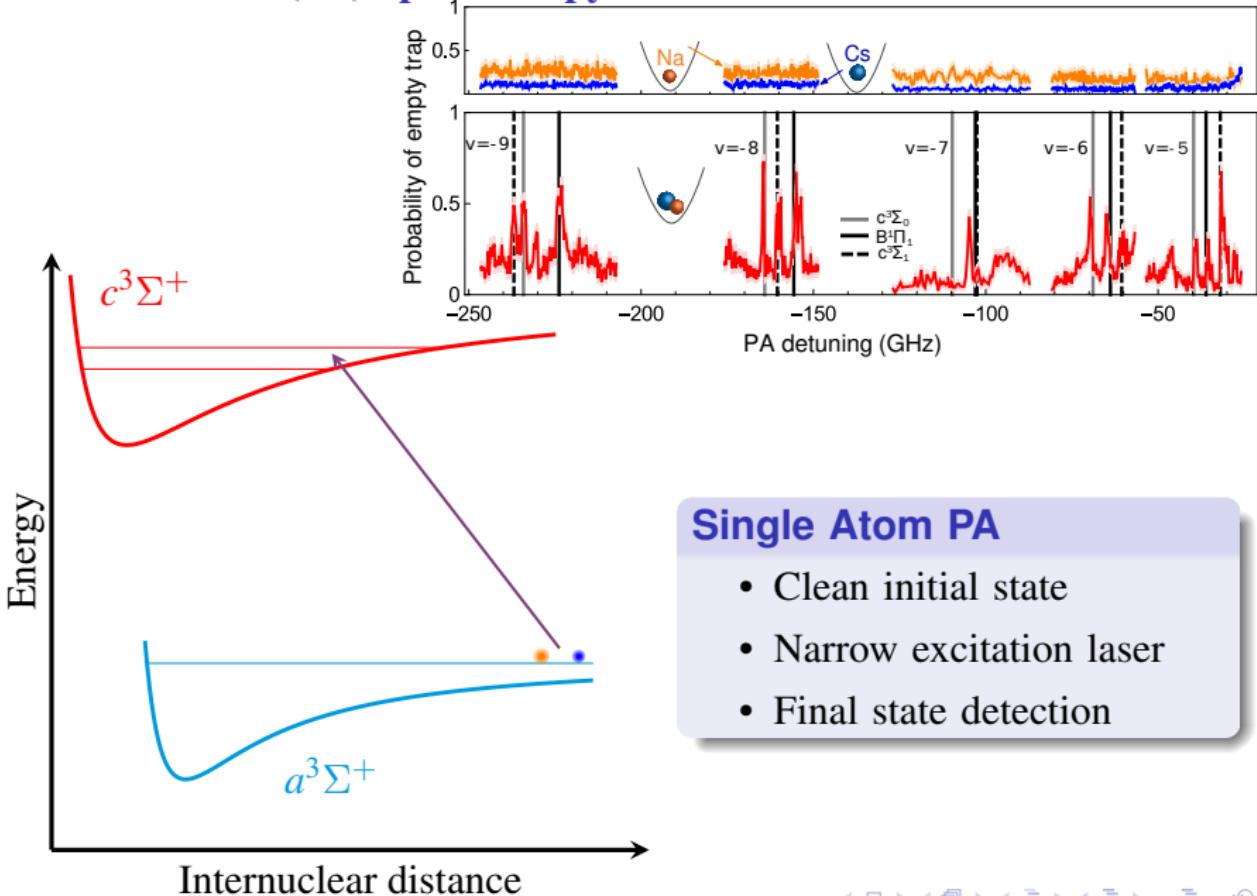
Probability of empty trap



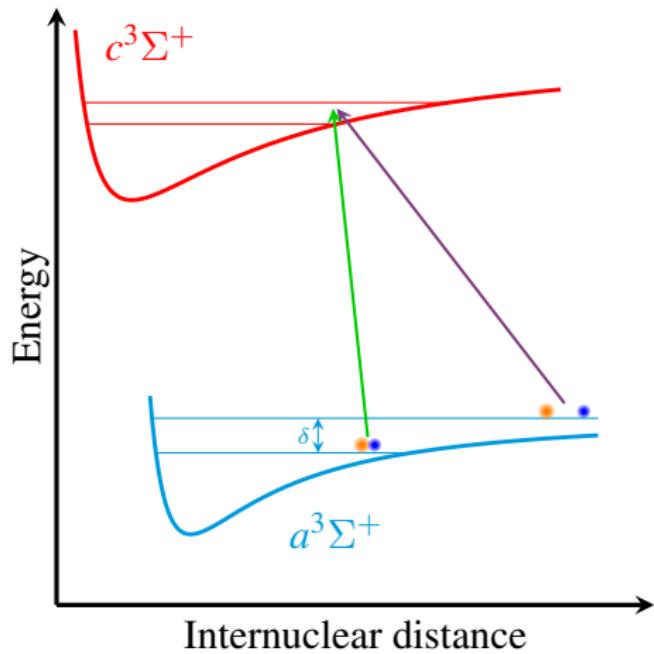
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# Electromagnetically Induced Transparency (EIT) Spectroscopy



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