

Coherent Creation of Single Molecules from Single Atoms

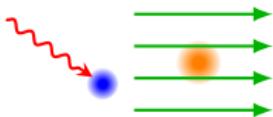
Yichao Yu

Ni Group/Harvard

Simple System



Full Control



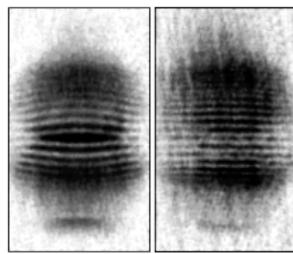
Complex Dynamic

Simple System

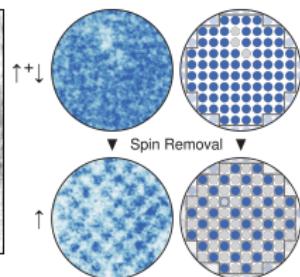
Full Control



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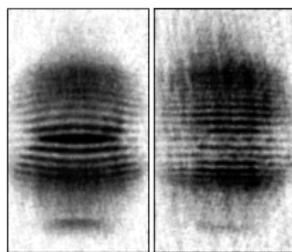
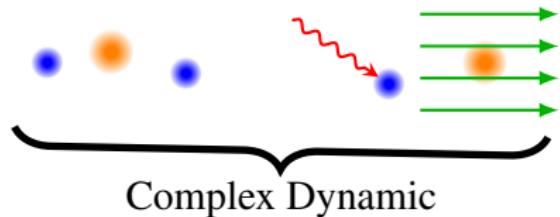
Ketterle et al.



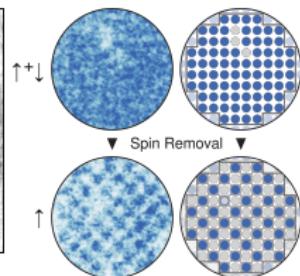
Greiner et al.

Simple System

Full Control



Ketterle et al.



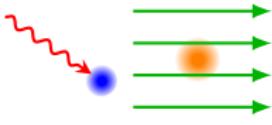
Greiner et al.

- ✗ Simple internal structure
- ✗ Weak interaction

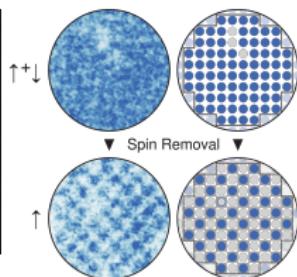
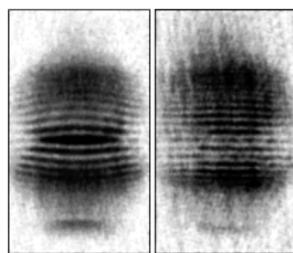
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Complex Dynamic



Ketterle et al.

Greiner et al.

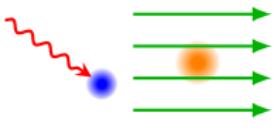
- Strong interaction
- Rich internal structure
- Long coherence time
- Fully controllable

- Simple internal structure
- Weak interaction

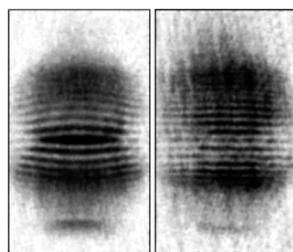
Simple System



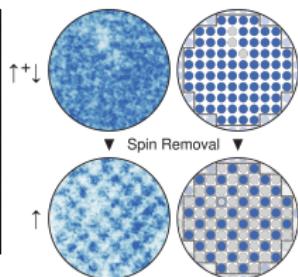
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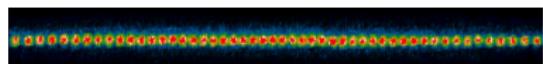


Ketterle et al.



Greiner et al.

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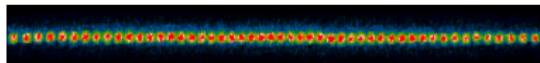
Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)

- Simple internal structure
- Weak interaction

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



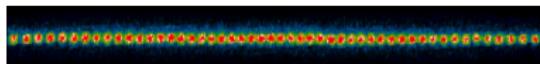
Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)

✓ Strong interaction (kHz)

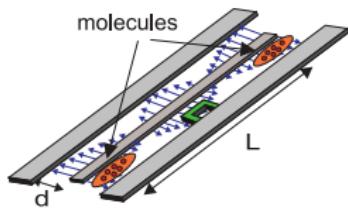
- Rich internal structure
- Long coherence time
- Fully controllable



Ions (Monroe et al.)



Rydberg Atoms (Lukin et al.)



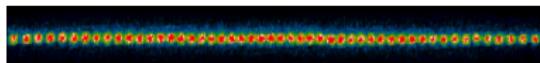
Dipolar Molecule (PRL. 97, 33003 (2006))

✓ Strong interaction (kHz)

□ Rich internal structure

✓ Long coherence time

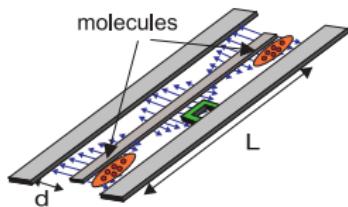
□ Fully controllable



Ions (Monroe et al.)

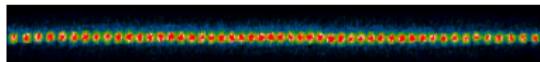


Rydberg Atoms (Lukin et al.)



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

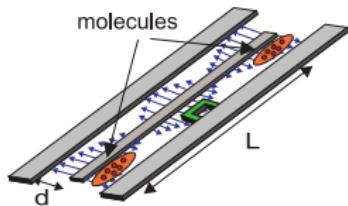
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Ions (Monroe et al.)



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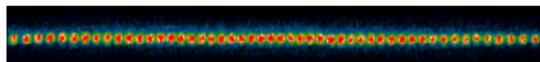
Dipolar Molecule (PRL. 97, 33003 (2006))

- ✓ Strong interaction (kHz)
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Optical tweezers

- Single site resolution

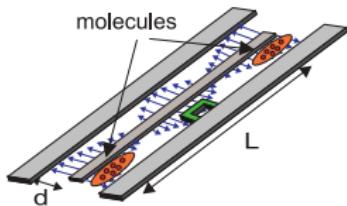
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Ions (Monroe et al.)

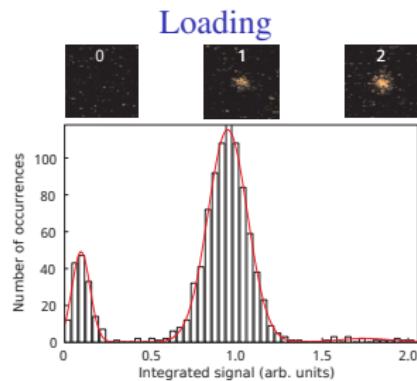


Rydberg Atoms (Lukin et al.)



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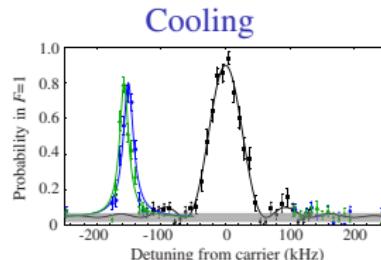
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Nat. Phys. 6, 951 (2010)

Optical tweezers

- Single site resolution
- ...



PRX. 2, 041014 (2012)

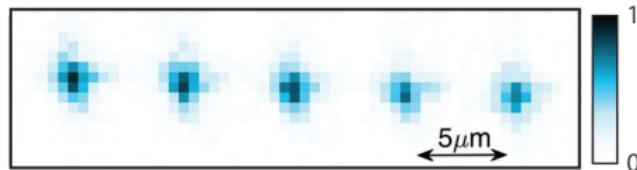
Rearranging



Science 354, 1024 (2016)

Ultracold molecules in tweezers

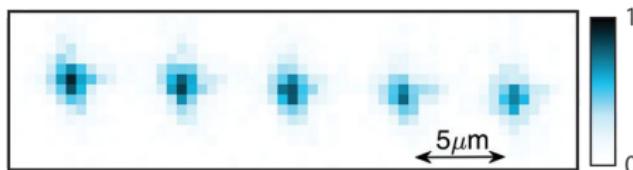
Direct cooling



Science 365, 1156 (2019)

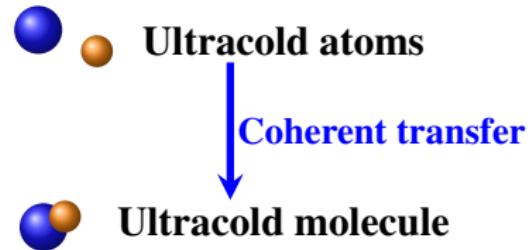
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



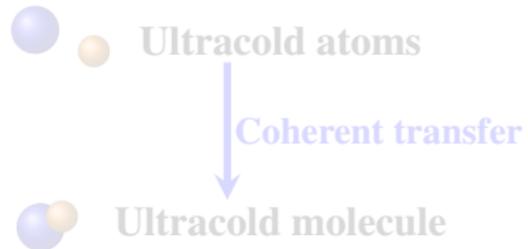
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezer
- Quantum control

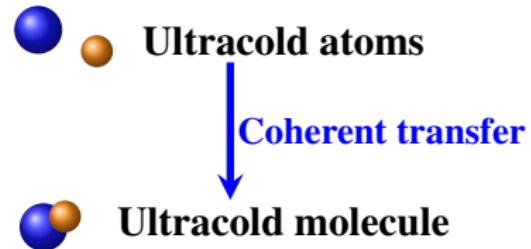
Ultracold molecules in tweezers

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

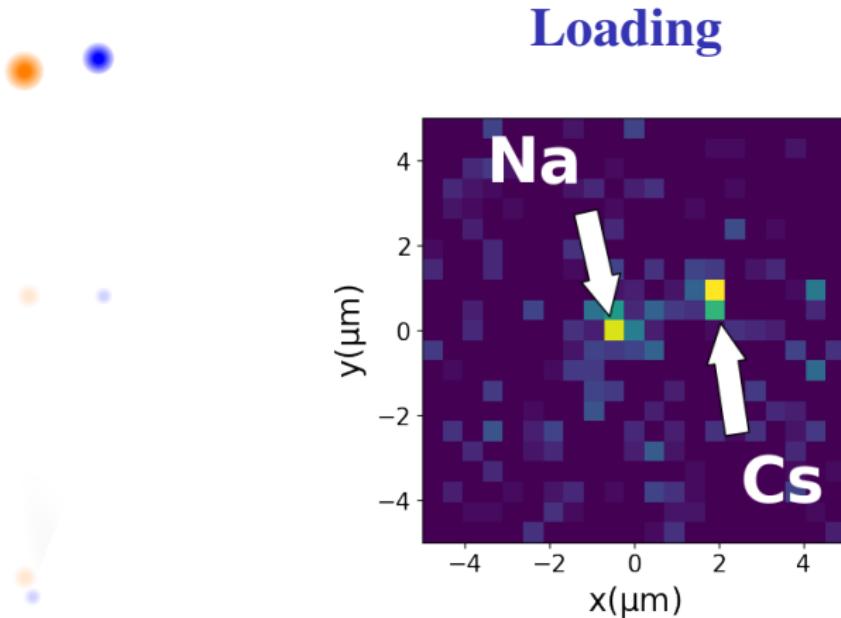
- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

NaCs molecule

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

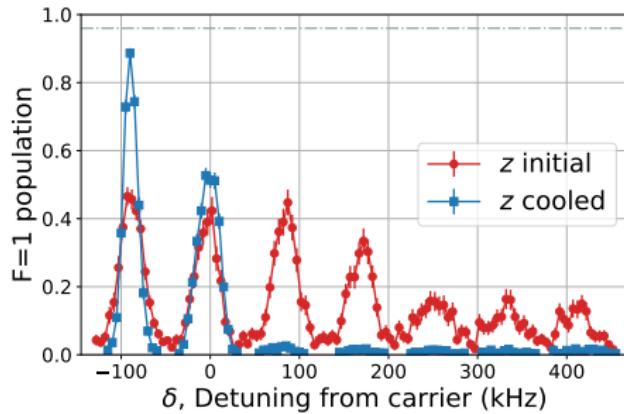
Experiment overview



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

Experiment overview

Cooling



Cs: 96% ground state¹

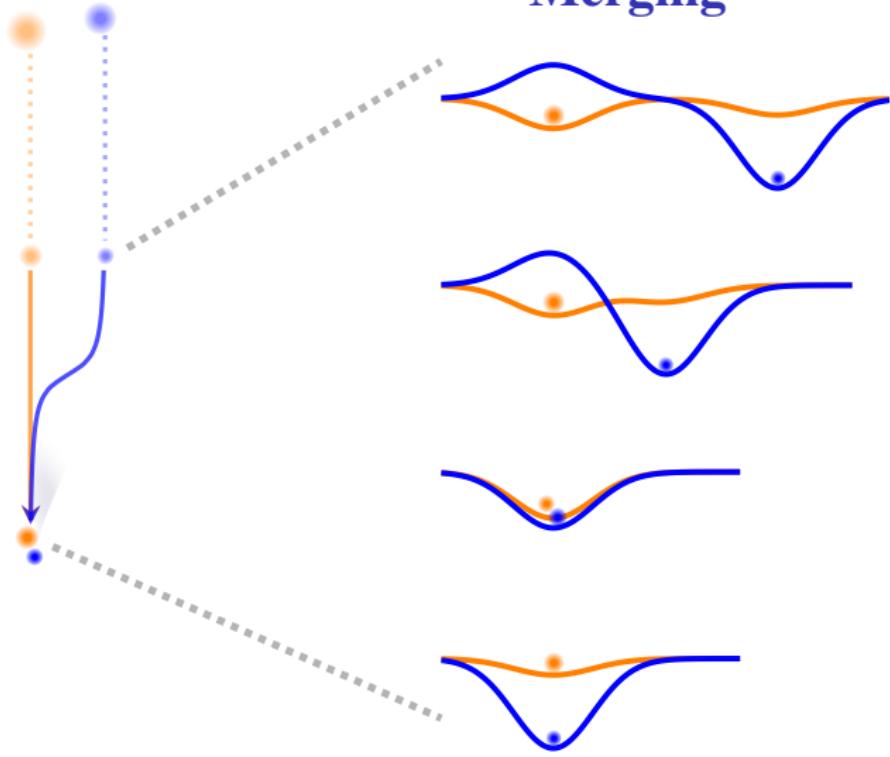
Na: 94% ground state²

¹Y. Yu et al. PRX 9, 021039 (2019)

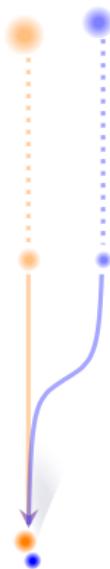
²Y. Yu et al. PRA 97, 063423 (2018)

Experiment overview

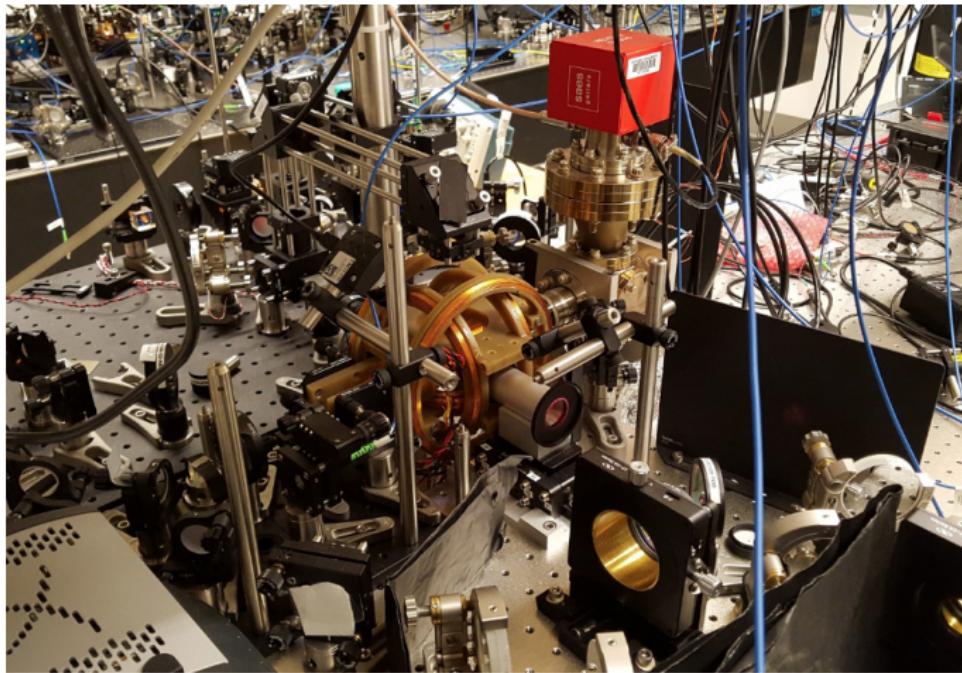
Merging

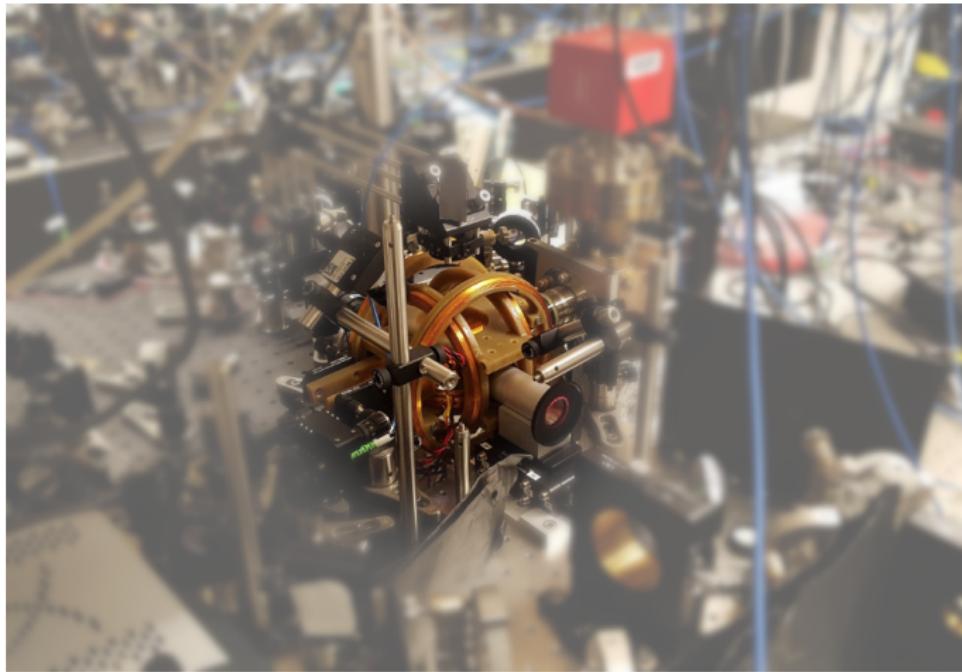


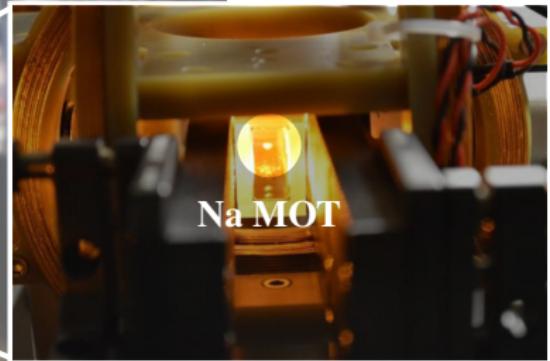
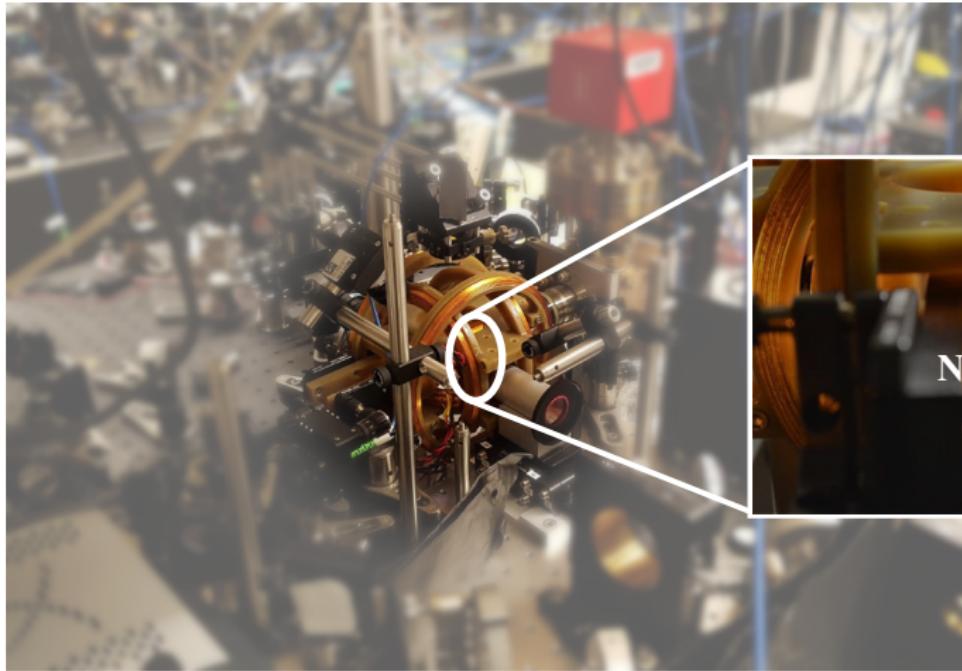
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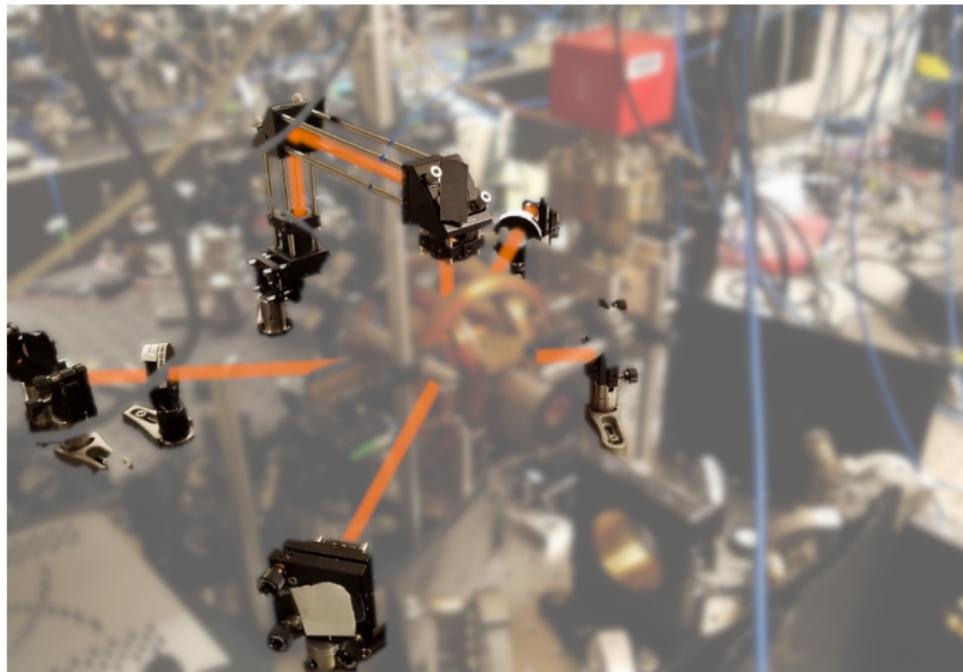
L. R. Liu, J. D. Hood, **Y. Yu** et al., *Science* 360, 6391 (2018)



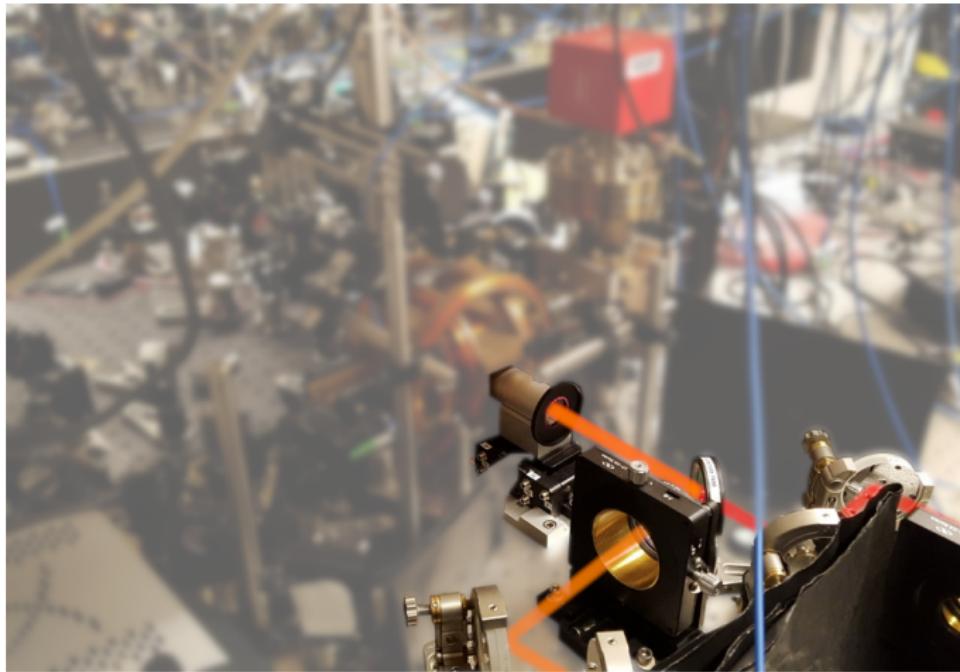




MOT beam path



Tweezer beam path



Outline

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2 Atom state control

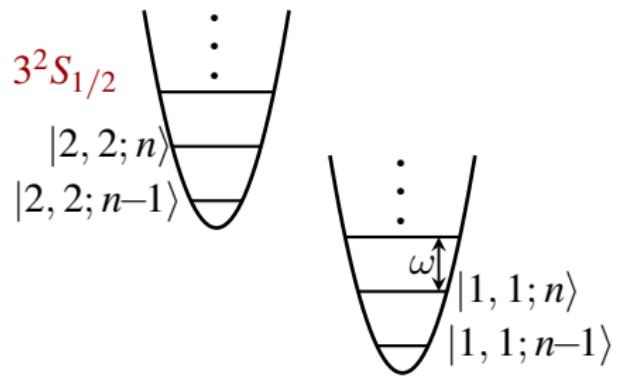
- Raman sideband cooling of Na atoms

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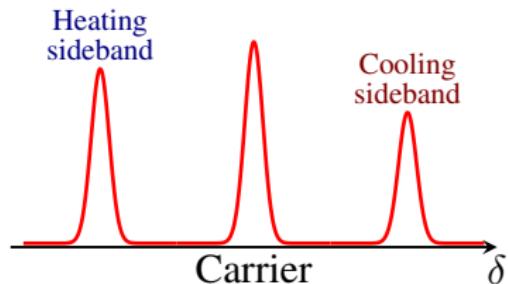
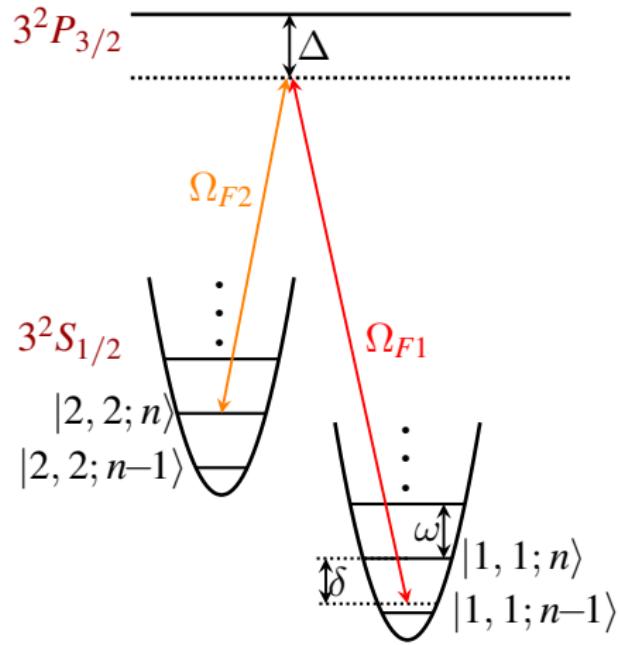
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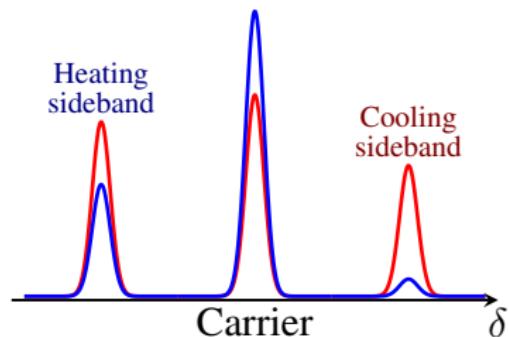
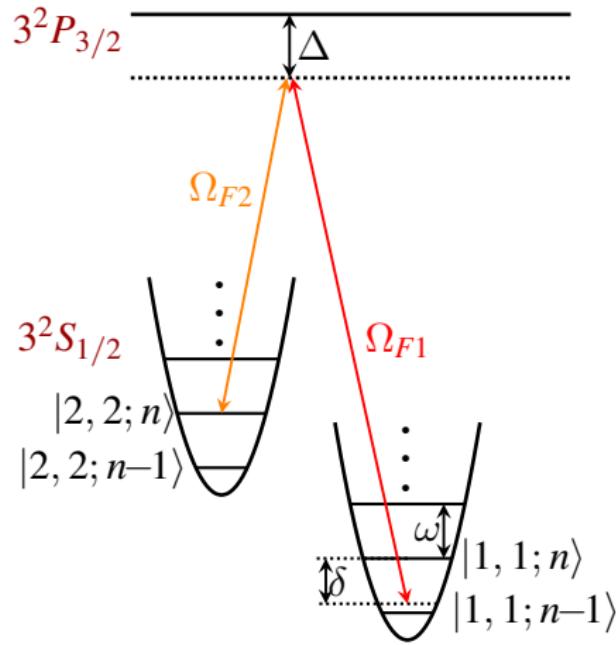
Raman sideband cooling



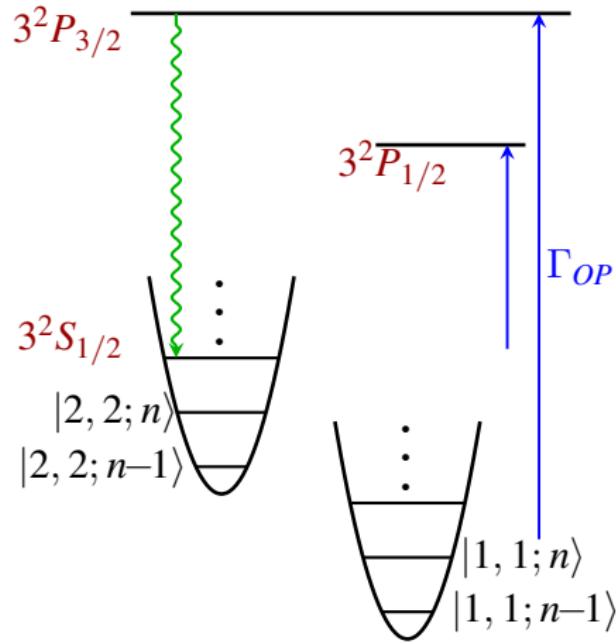
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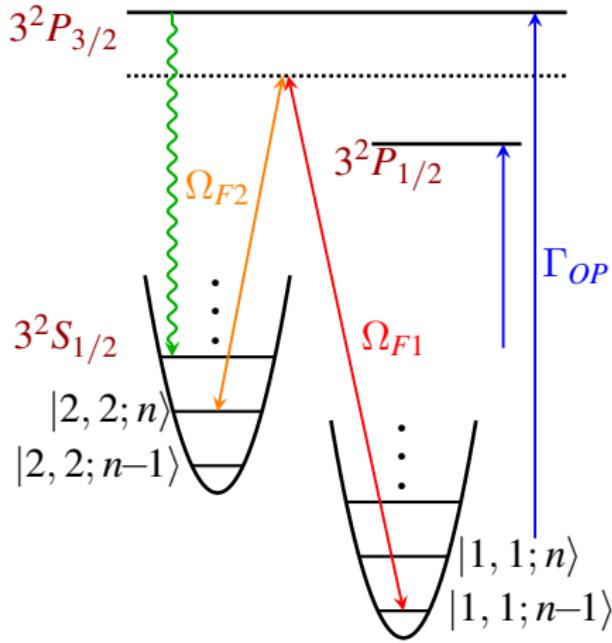
Raman sideband cooling



Raman sideband cooling

Lamb Dicke parameter

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

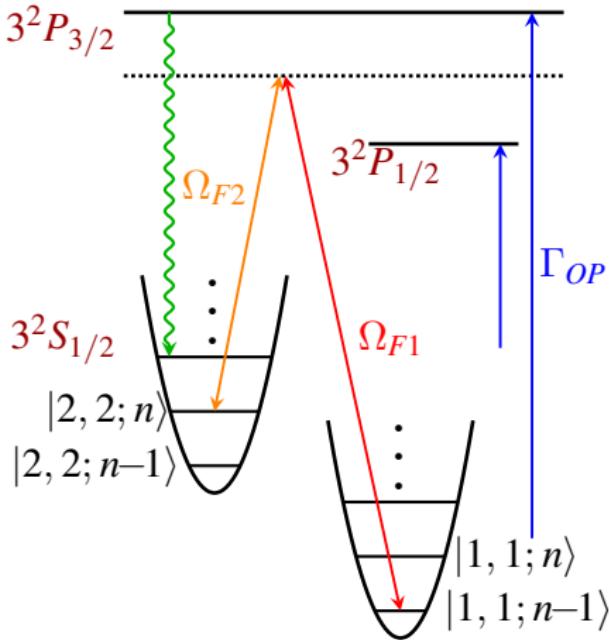


Raman sideband cooling

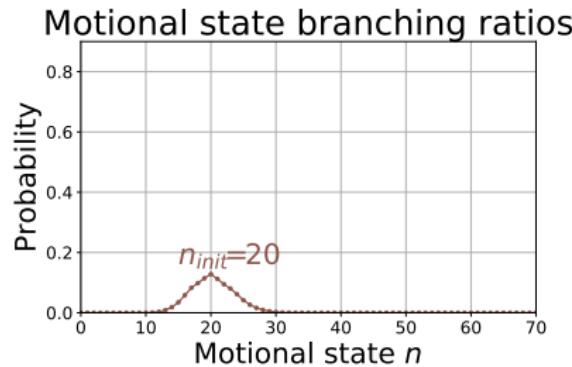
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$$\eta_{Na}^{OP} = 0.55$$



Raman sideband cooling



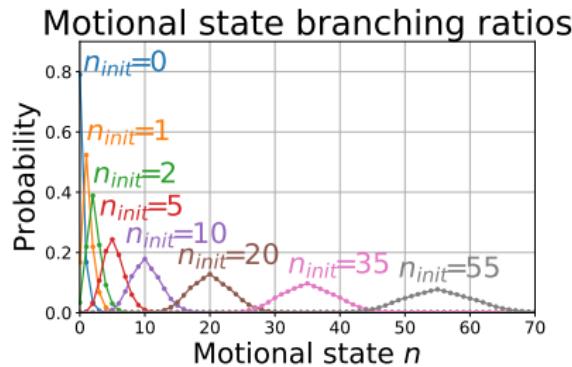
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- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



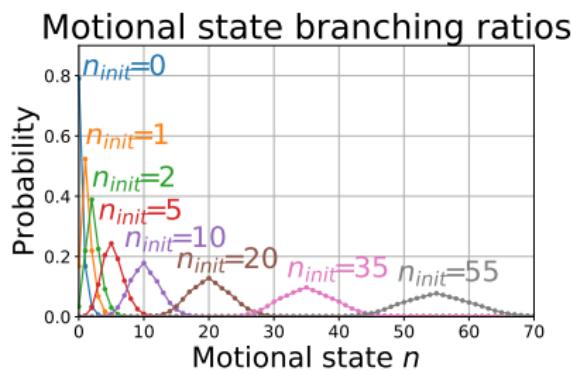
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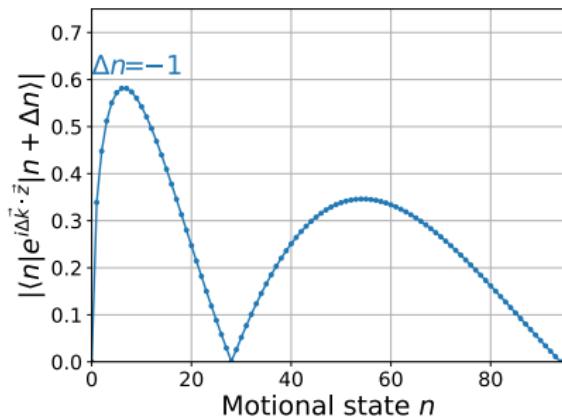


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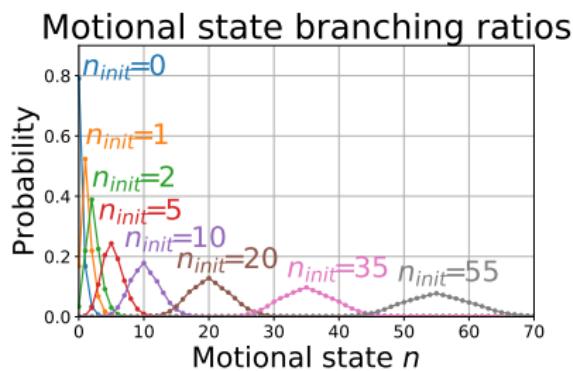
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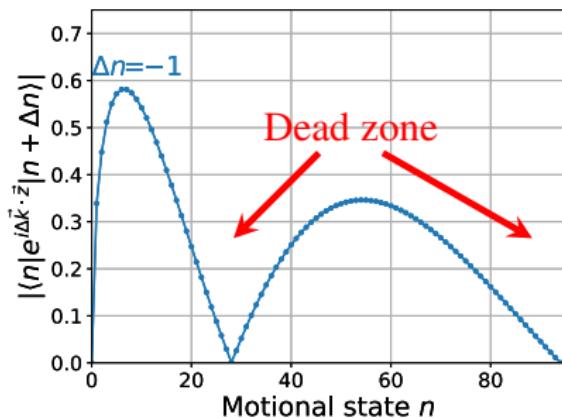


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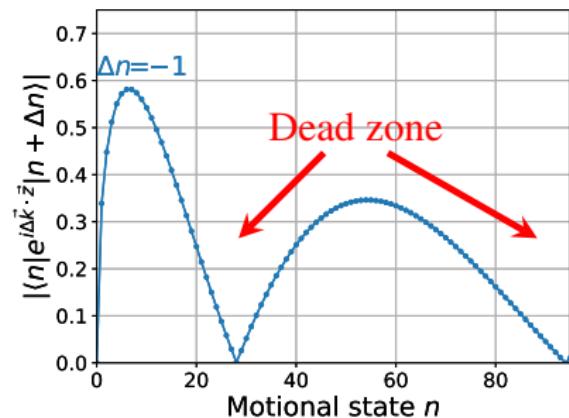
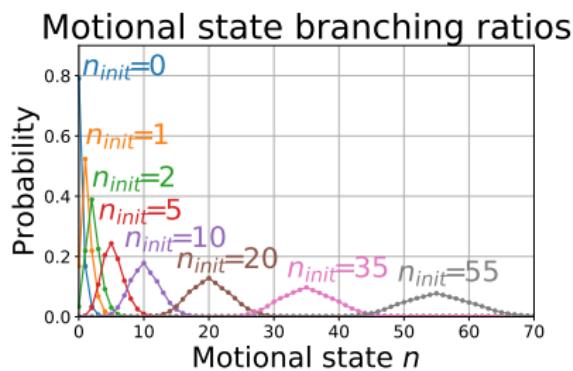
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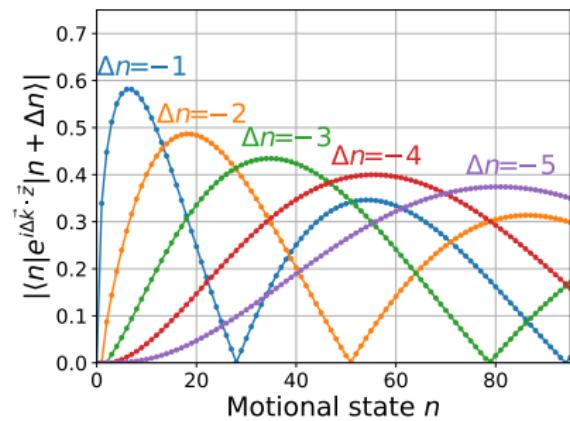
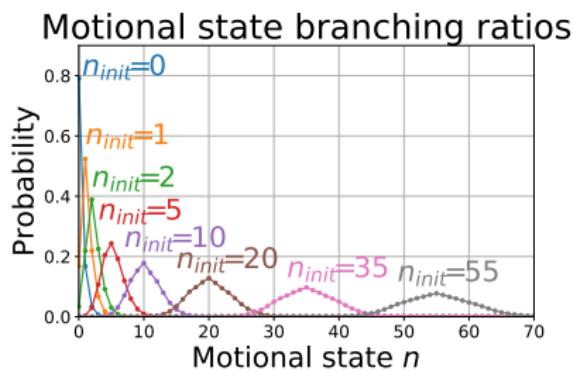
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Solution

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

Raman sideband cooling



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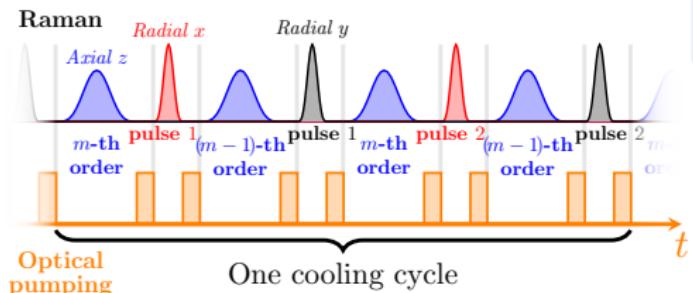
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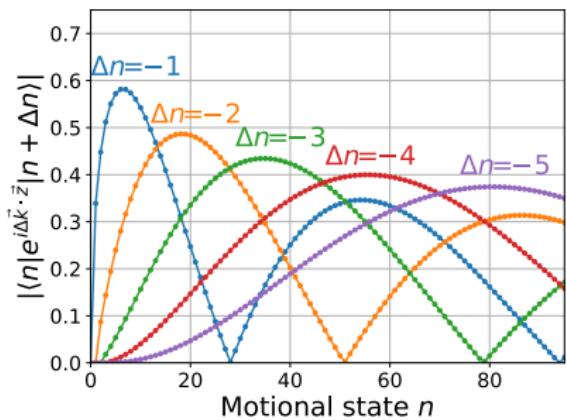
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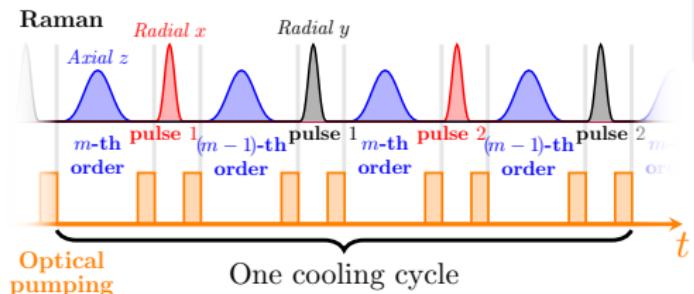
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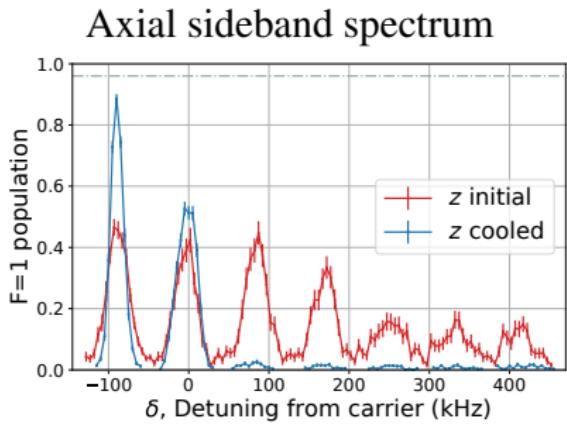
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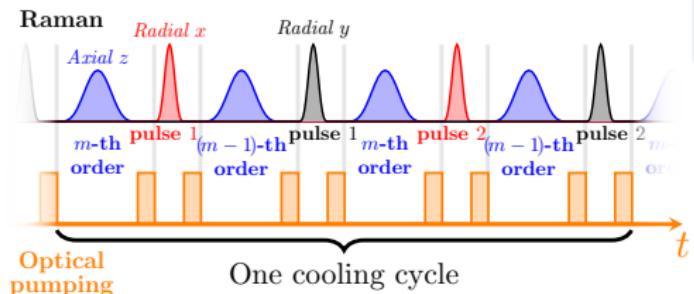
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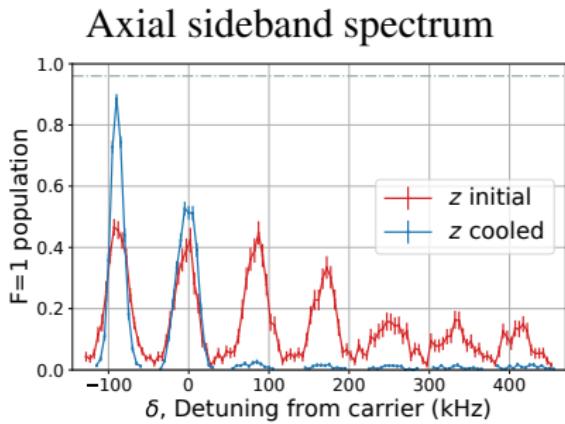
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3D ground state: 93.5(7)%

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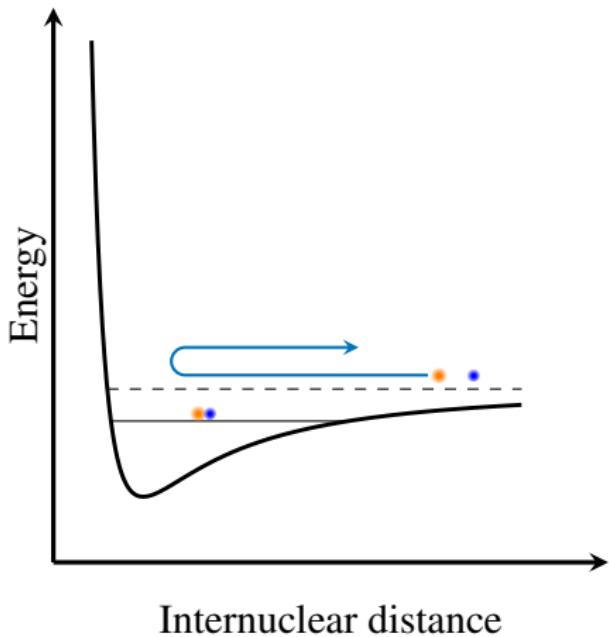
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- Coherent optical transfer

4 Conclusion

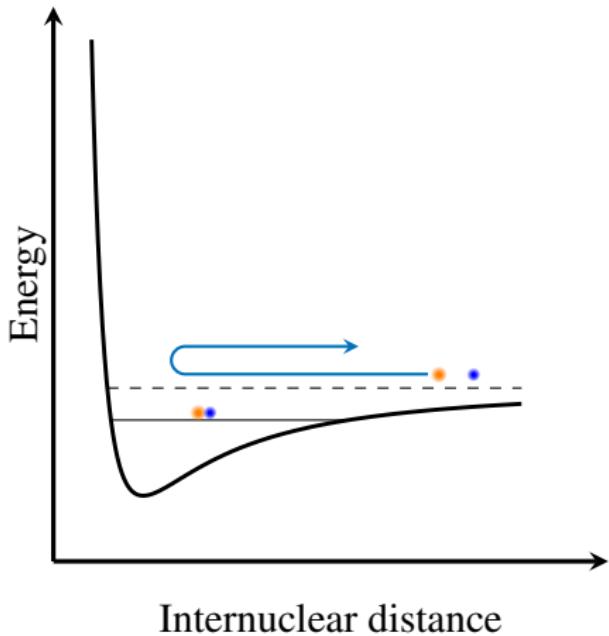
Scattering length a

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



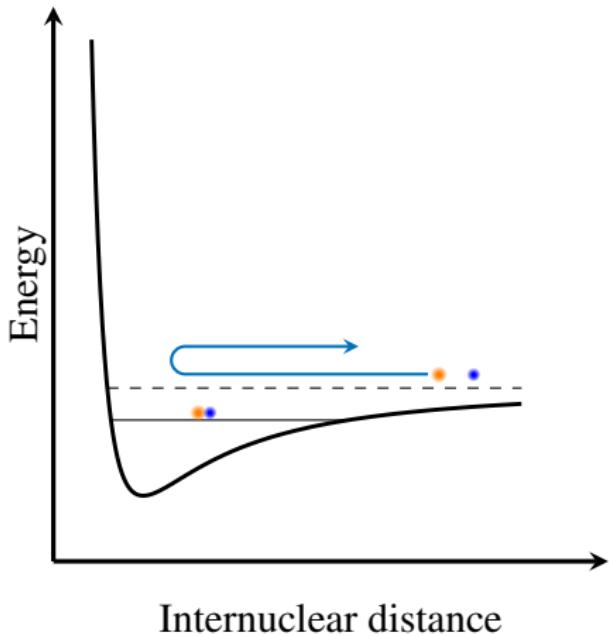
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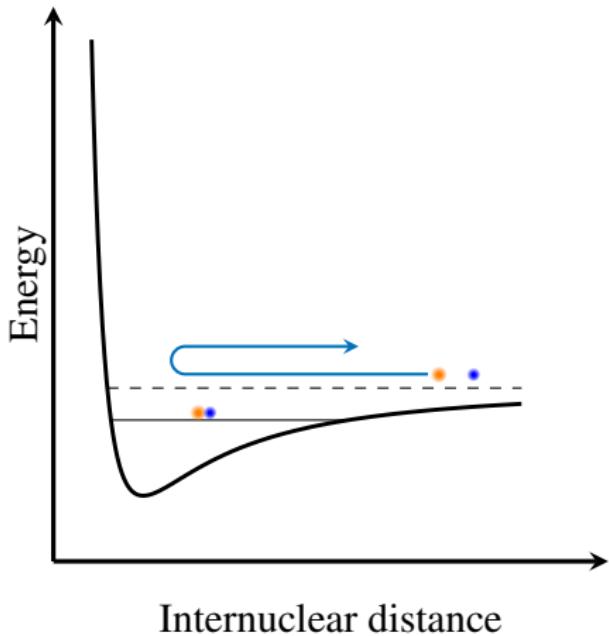
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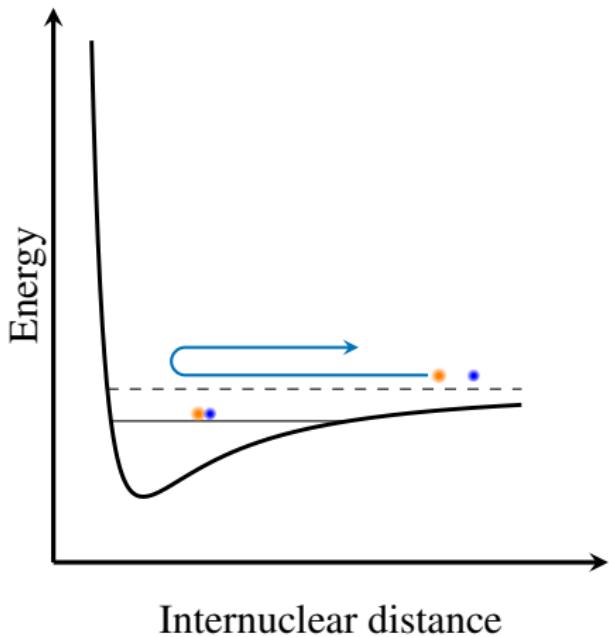
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- Feshbach resonance
- ⋮

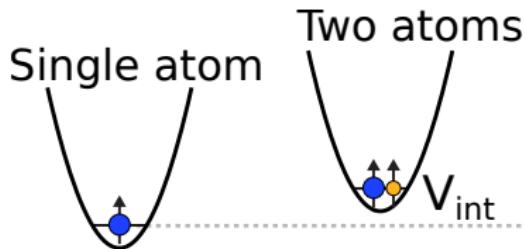


Scattering length a

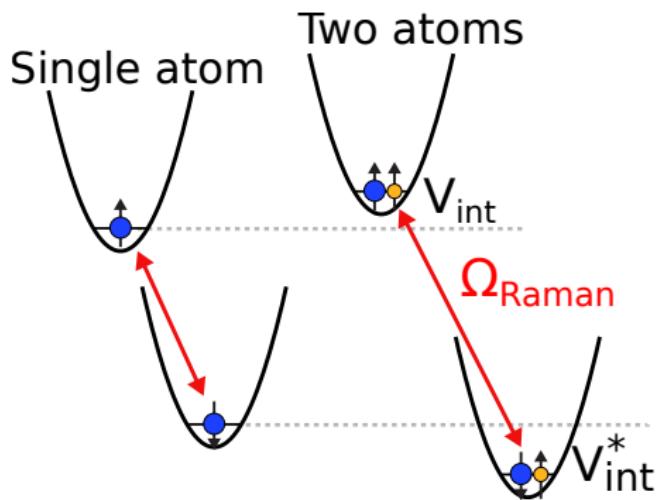
- Binding energy
- Molecular potential
- Molecule formation
- Feshbach resonance
- :



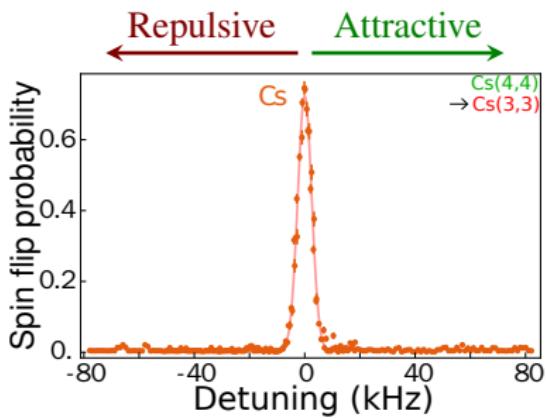
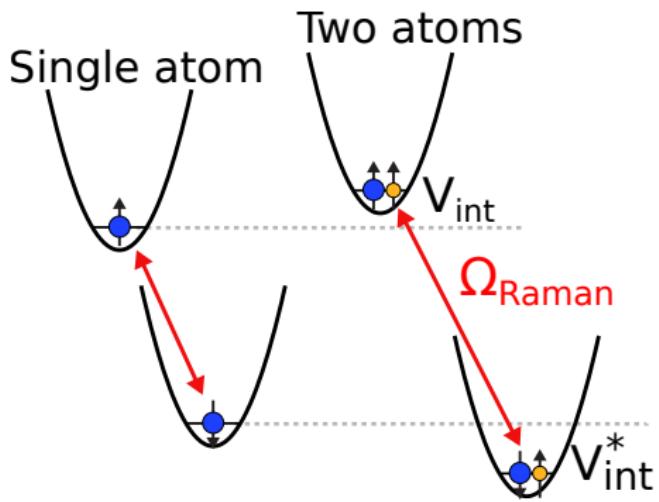
Interaction shift



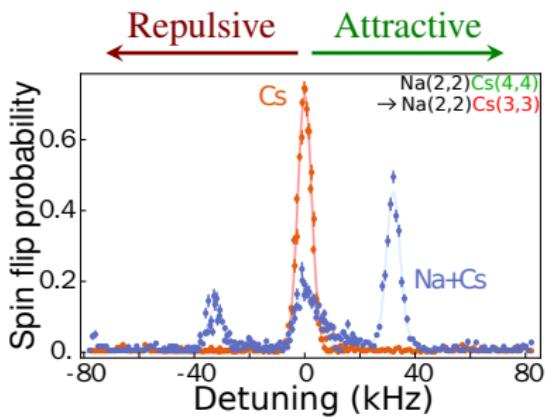
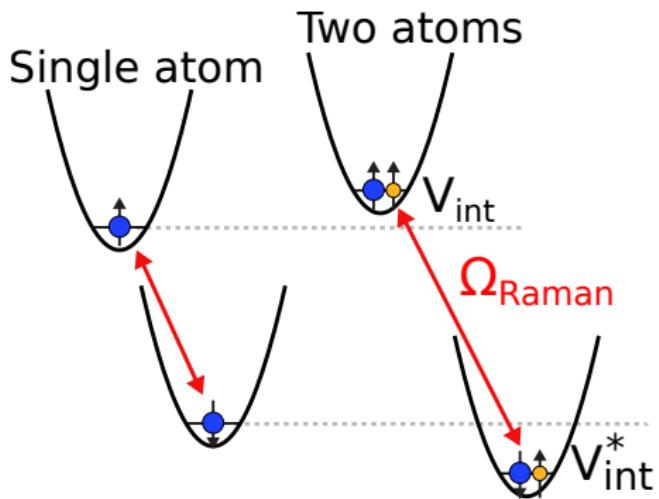
Interaction shift



Interaction shift



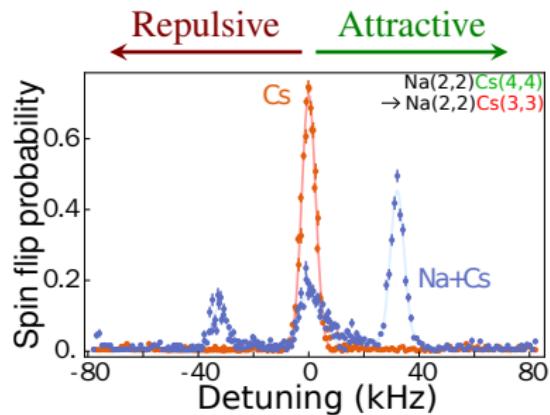
Interaction shift



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

$$M = m_1 + m_2$$

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

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

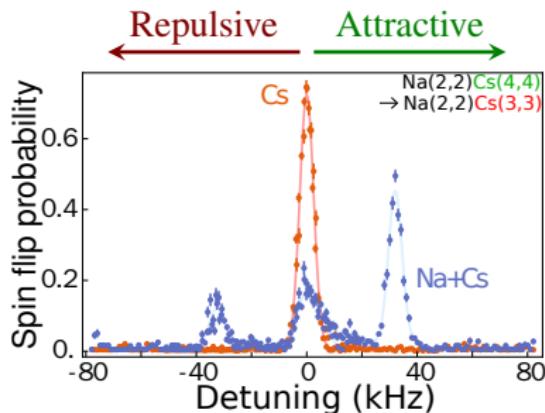
$$\omega_{R,i}^2 = \frac{m_2 \omega_{1,i}^2 + m_1 \omega_{2,i}^2}{m_1 + m_2}$$

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

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

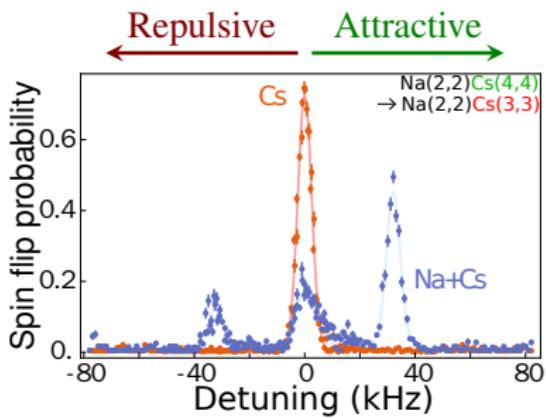
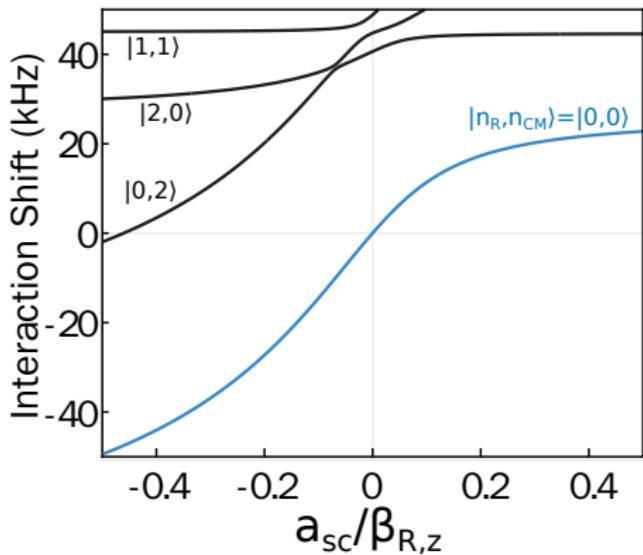
$$P_i = p_{1,i} + p_{2,i}$$

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



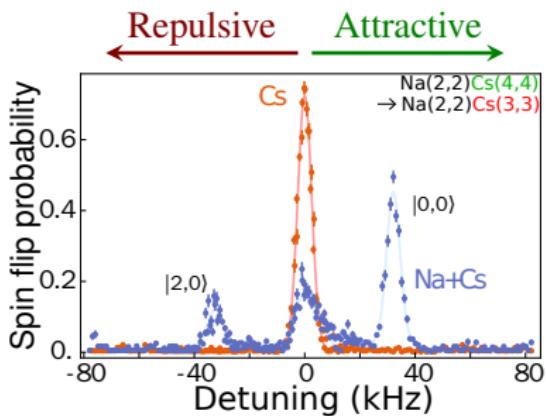
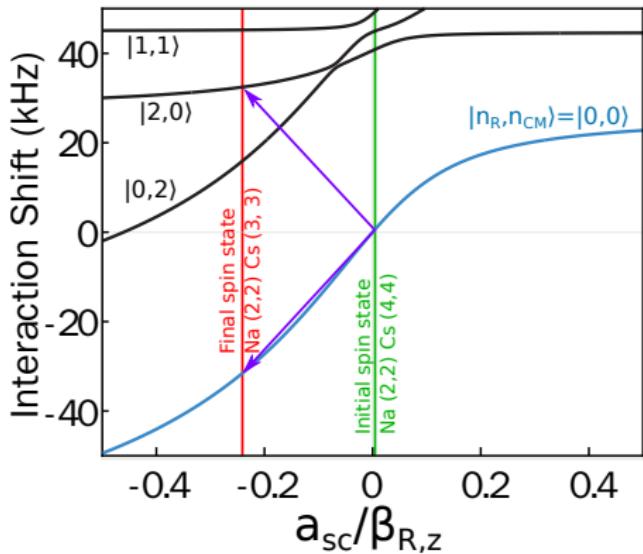
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M \Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu (\omega_{1,i}^2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

Interaction shift



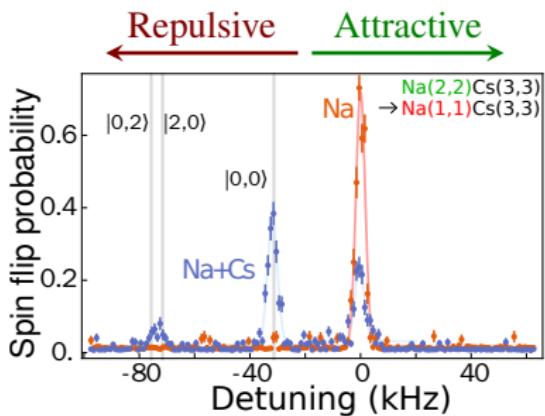
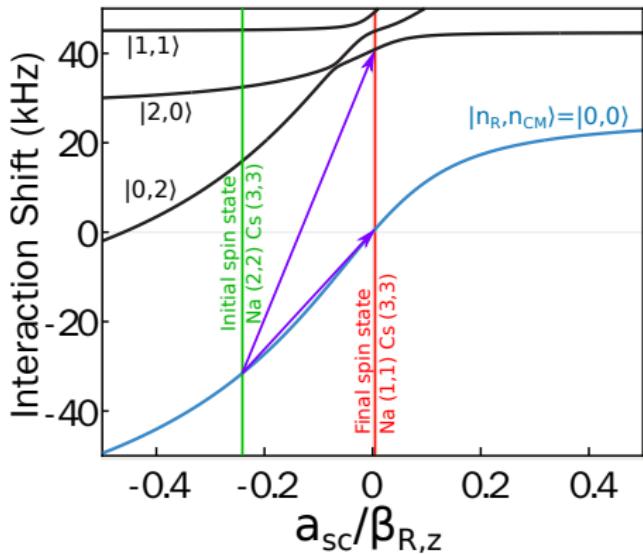
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



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

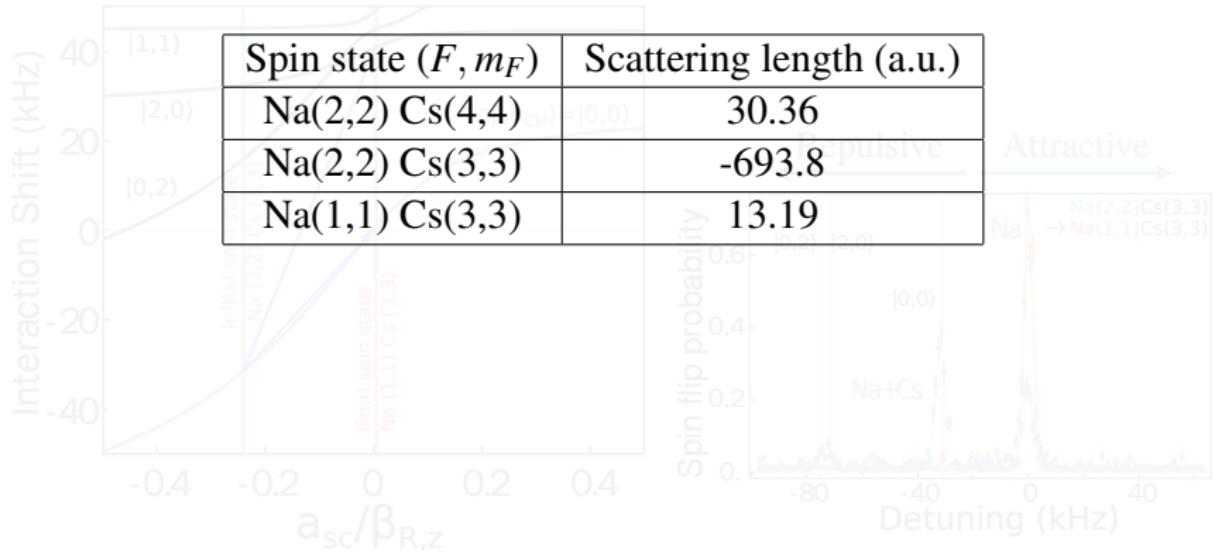
Interaction shift



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

Interaction shift

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



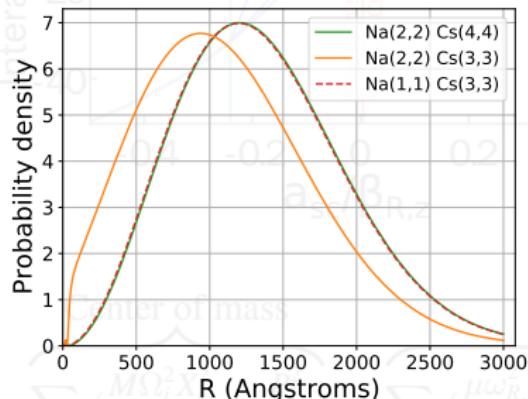
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

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!



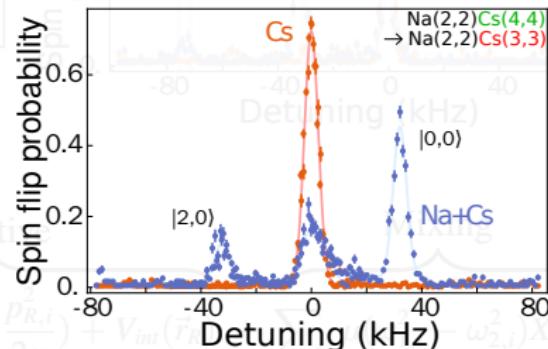
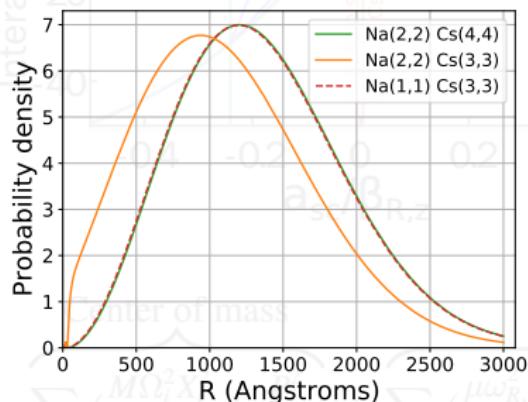
$$H = \sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i} X_{R,i}}{2M} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R) + \sum_{i=x,y,z} \mu (\omega_{1,i}^2 - \omega_{2,i}^2) X_i x_{R,i}$$

Interaction shift

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Enhanced coupling to molecular state!

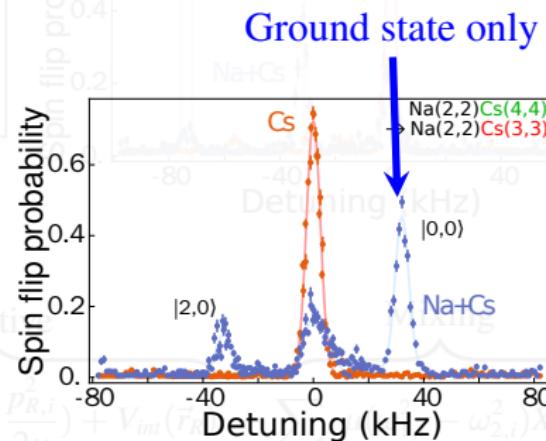
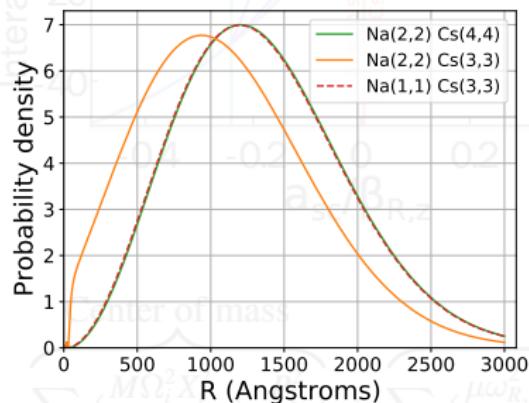


Interaction shift

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Enhanced coupling to molecular state!



Outline

1 Experiment overview

2 Atom state control

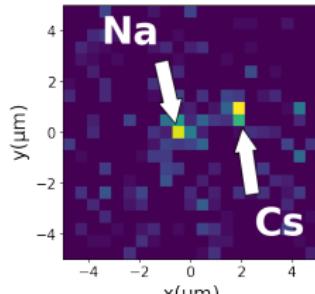
- Raman sideband cooling of Na atoms

3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

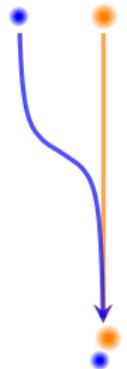
4 Conclusion

Loading

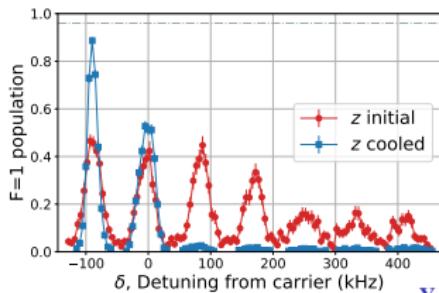


NJP. 19, 023007 (2017)

Merging



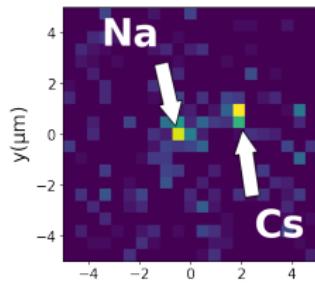
Cooling



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

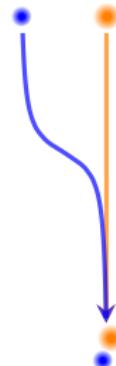
Y. Yu et al. PRA. 97, 063423 (2018)

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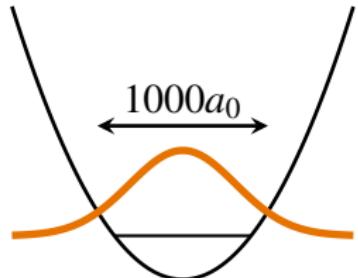


NJP. 19, 023007 (2017)

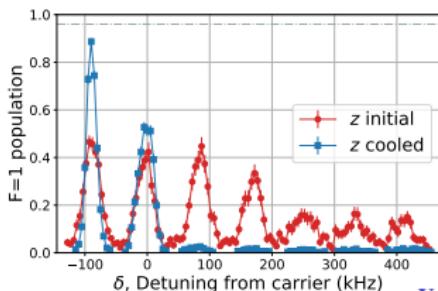
Merging



Atom



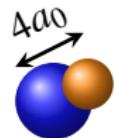
Cooling

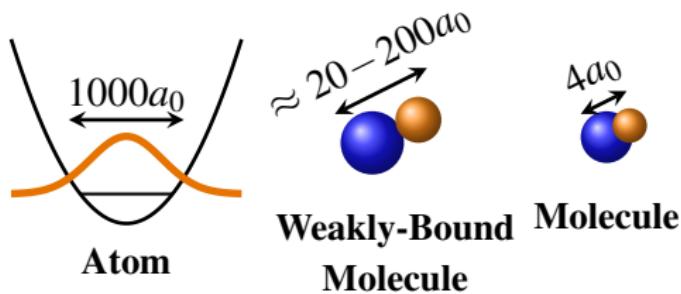


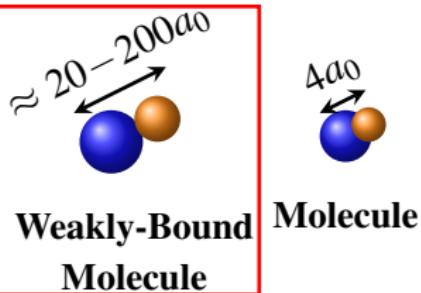
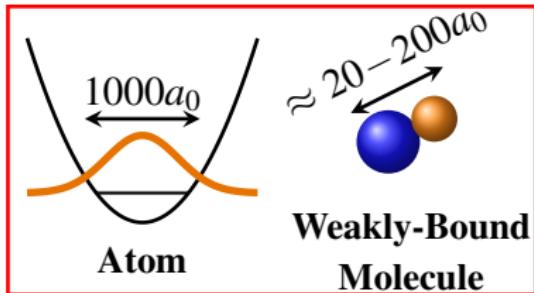
Y. Yu et al. PRA. 97, 063423 (2018)

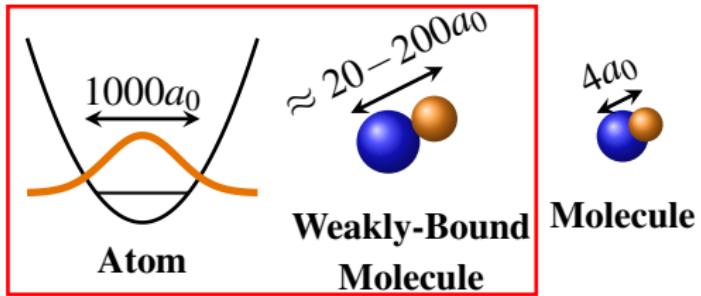
Y. Yu et al. PRX. 9, 021039 (2019)

Molecule

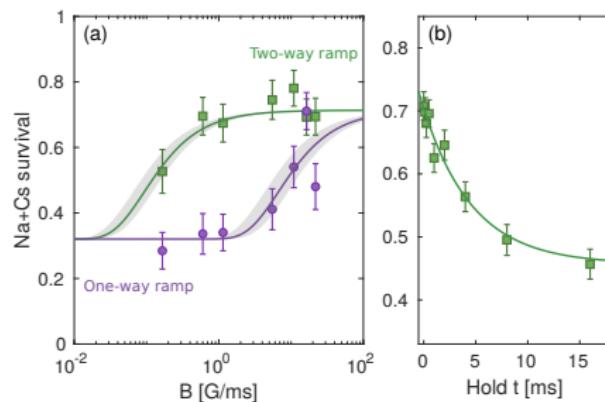




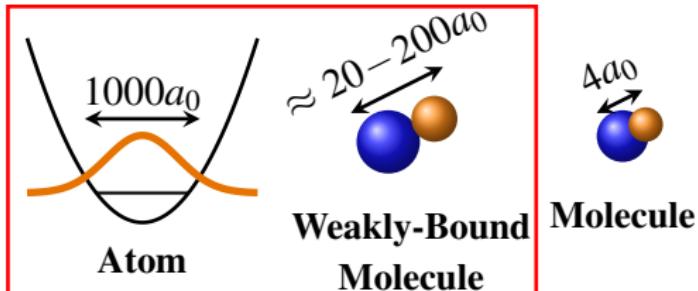




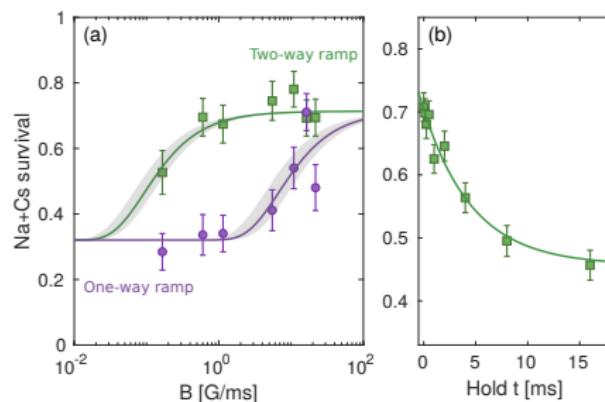
Feshbach molecule



PRL. 124, 253401 (2020)

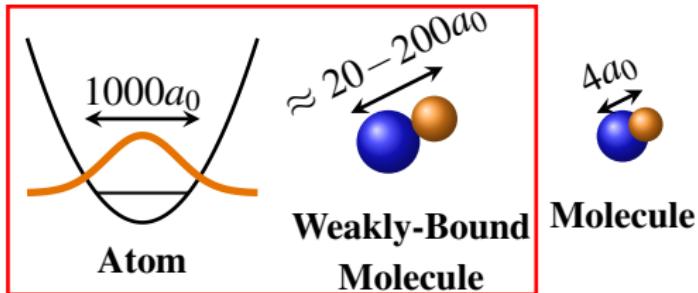


Feshbach molecule



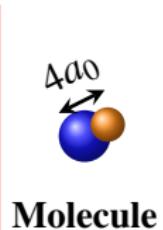
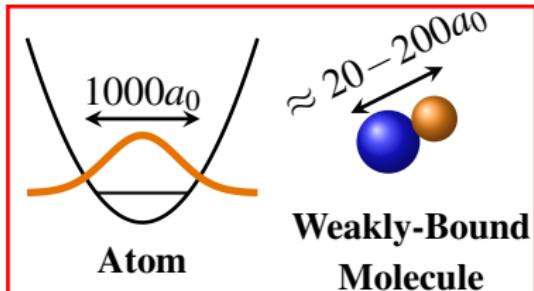
- Requires Feshbach resonance
- Usually large magnetic field

PRL. 124, 253401 (2020)



Optical transfer

- More general
- Faster

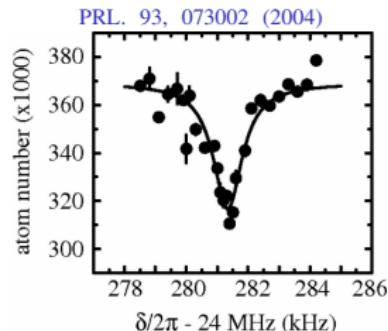


Optical transfer

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- Faster

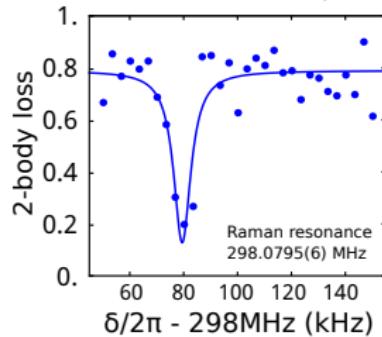
Previous results

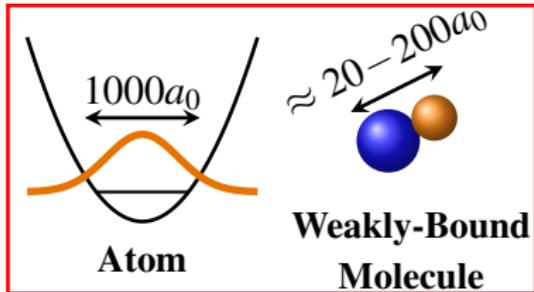
Rb₂ Science 287, 1016 (2000)



Sr₂ PRL. 109, 115302 (2012)

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





Optical transfer

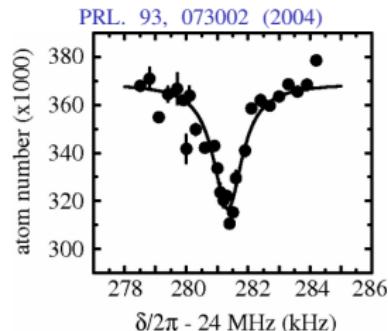
- More general
- Faster

Limitations so far

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

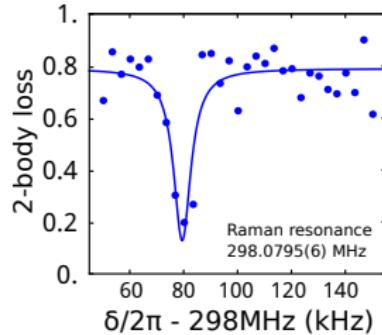
Previous results

Rb_2 Science 287, 1016 (2000)

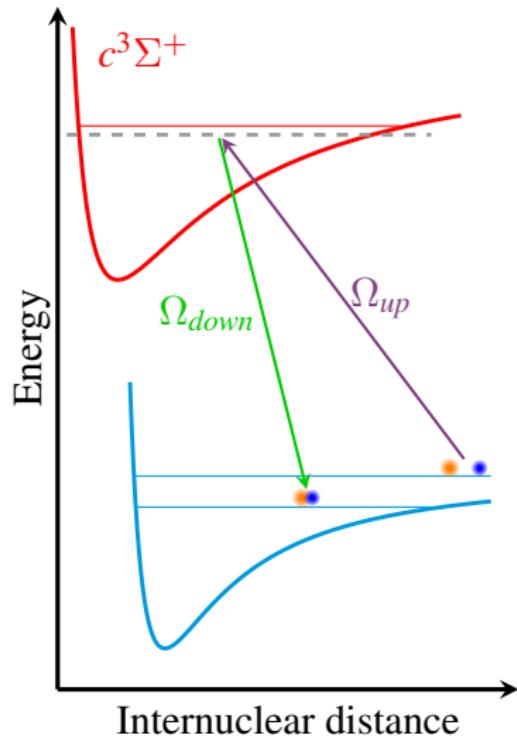


Sr_2 PRL. 109, 115302 (2012)

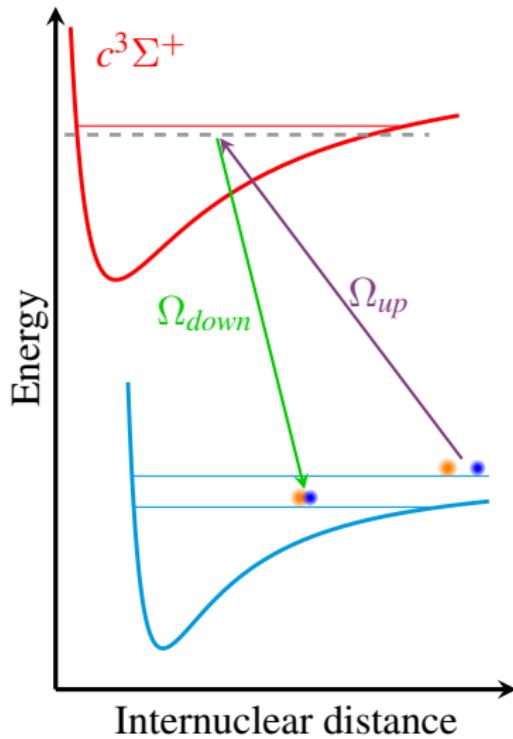
NaCs Y. Yu et al. PRX. 9, 021039 (2019)



Raman transfer



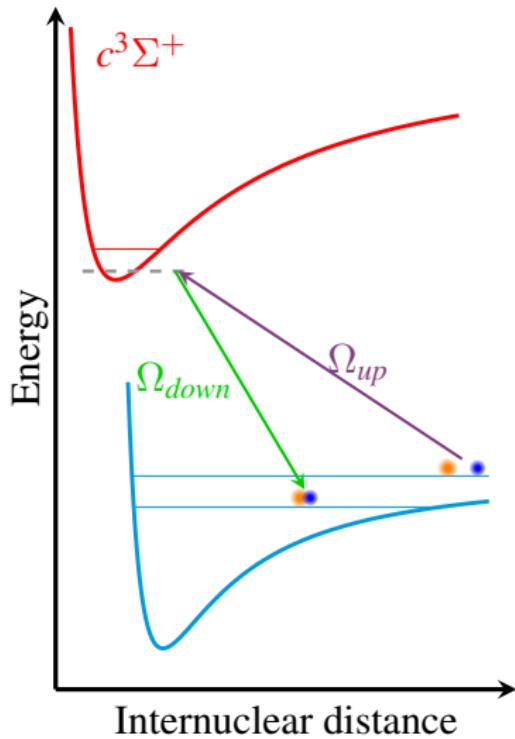
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

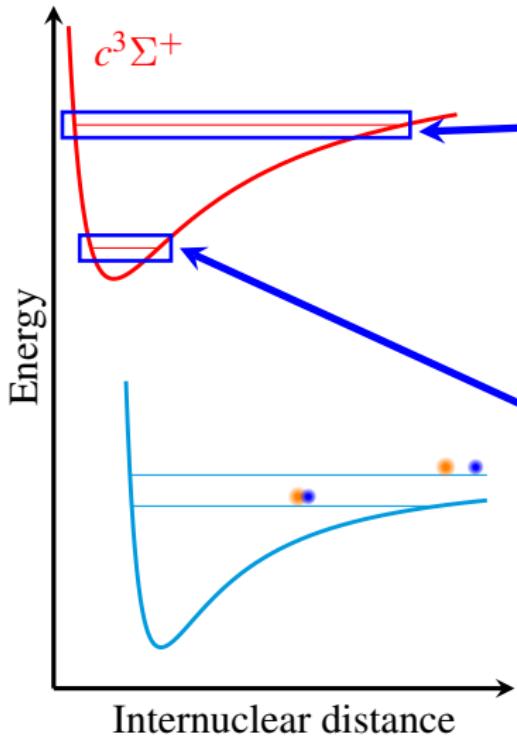
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

Deeply bound states

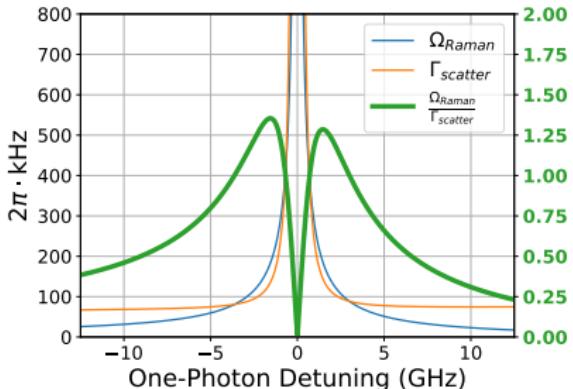
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

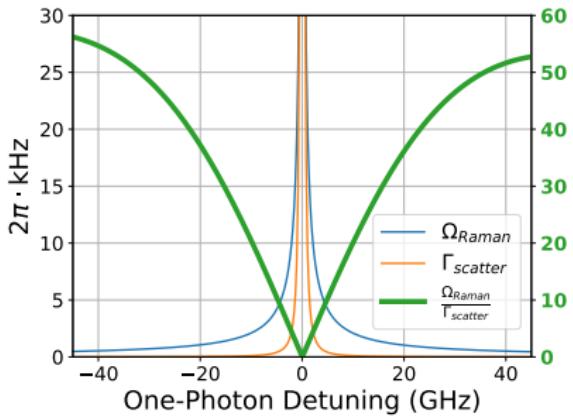
Raman transfer



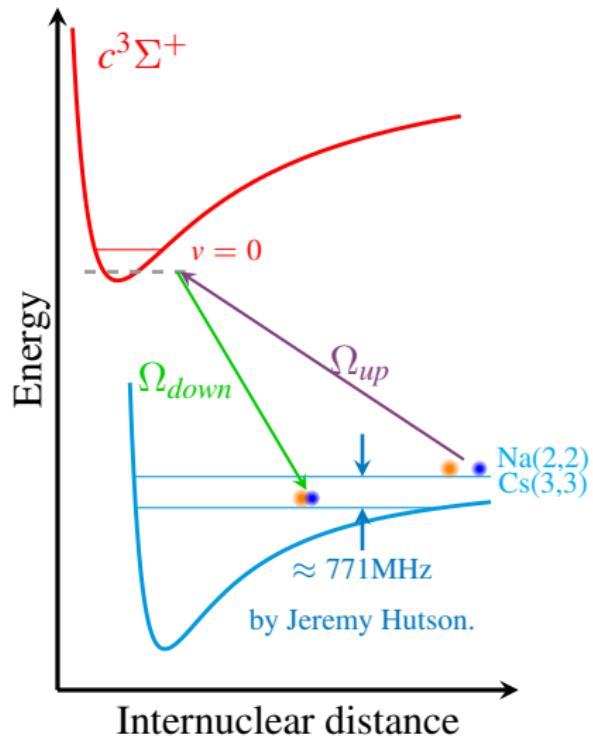
Near threshold states



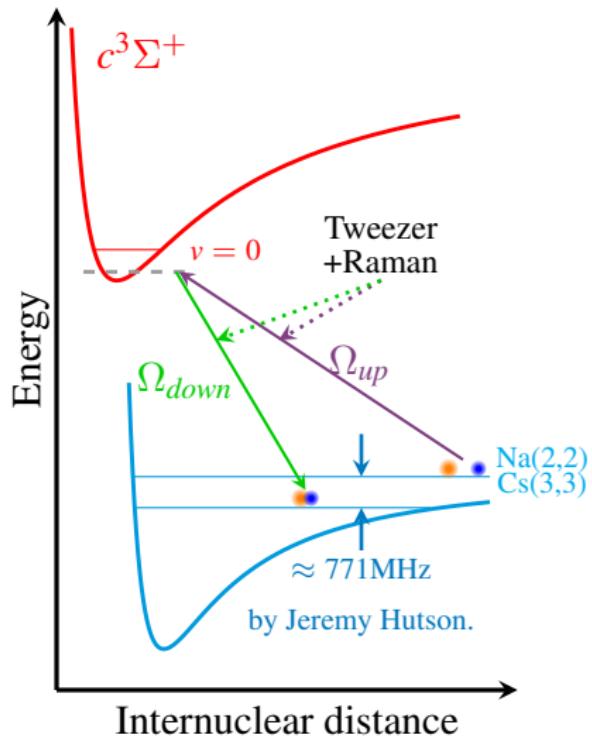
Deeply bound states



Experiment



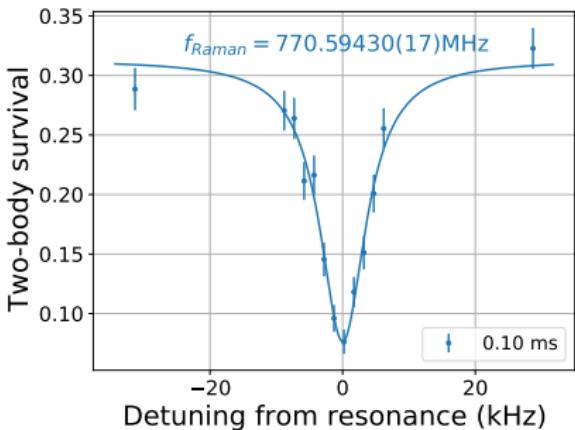
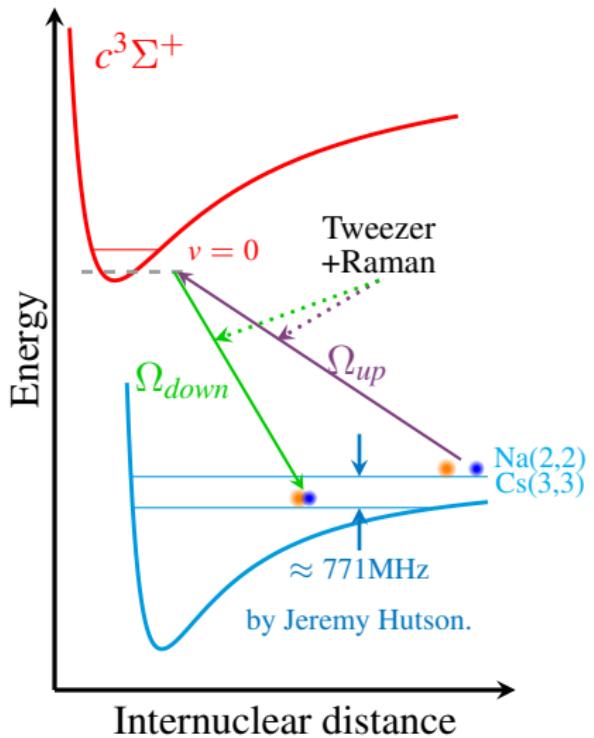
Experiment



Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

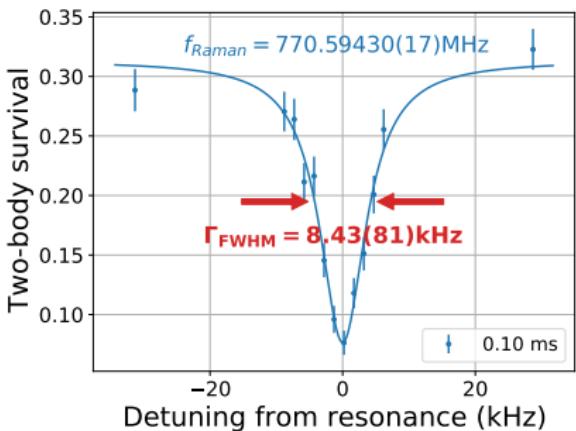
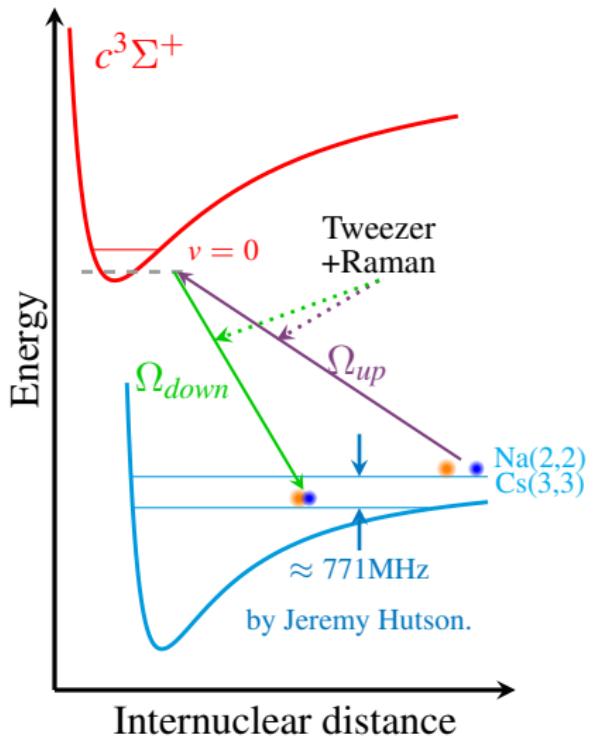
Experiment



Tweezer as Raman beam

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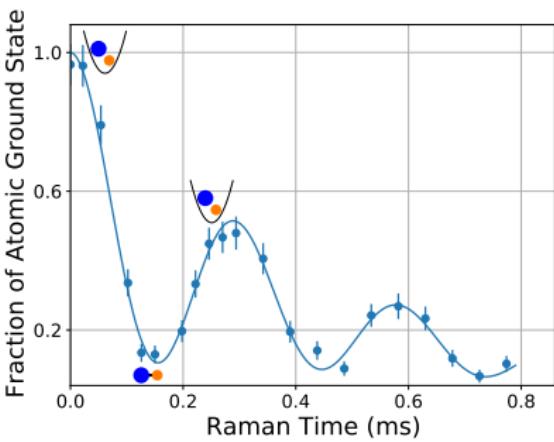
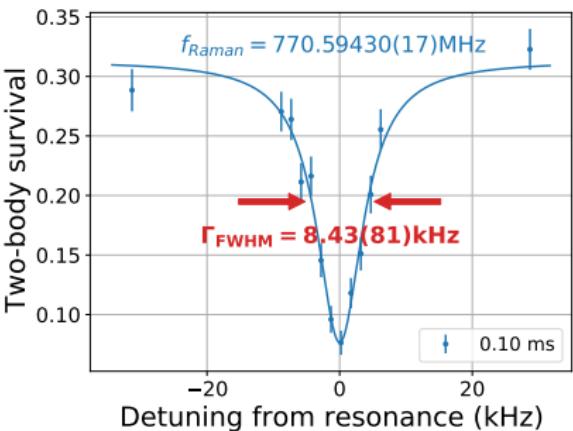
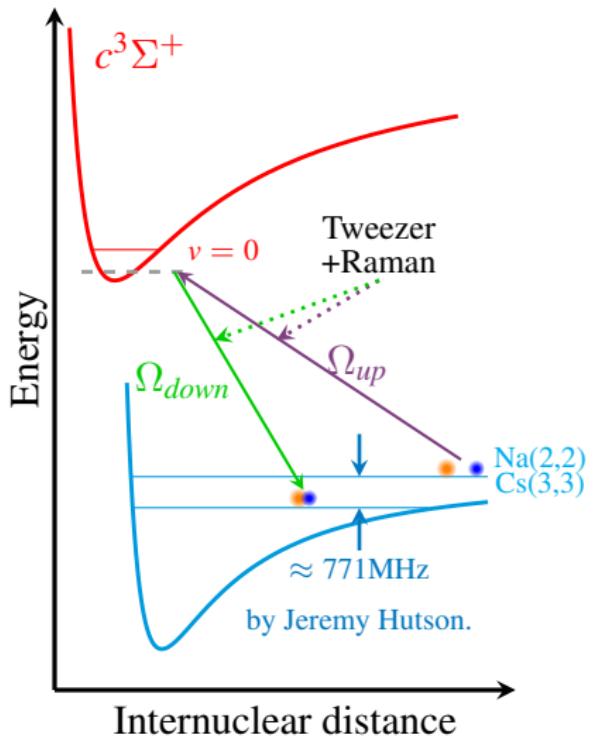
Experiment



Tweezer as Raman beam

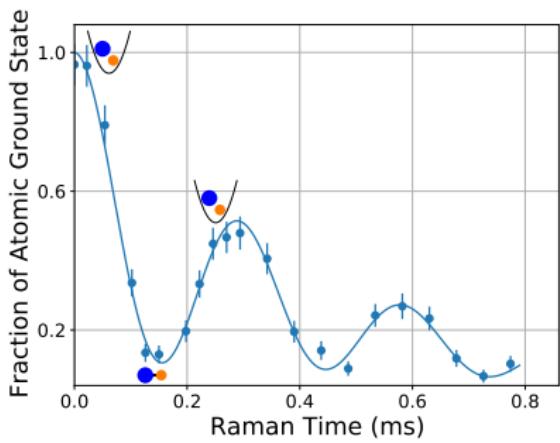
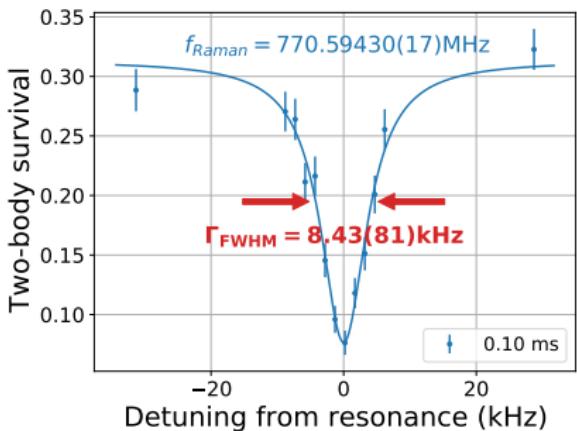
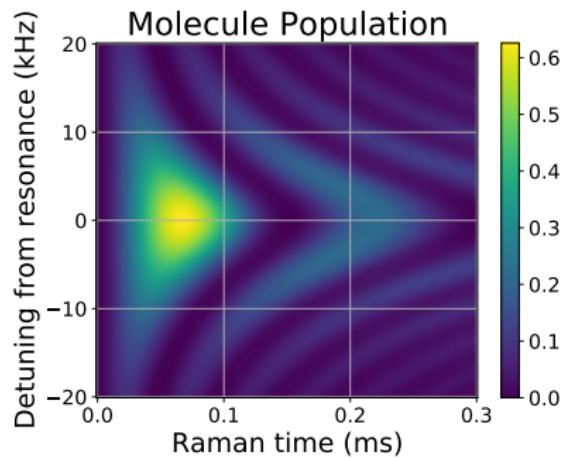
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Experiment



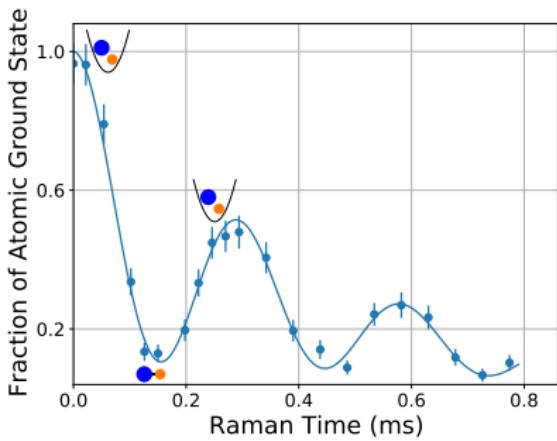
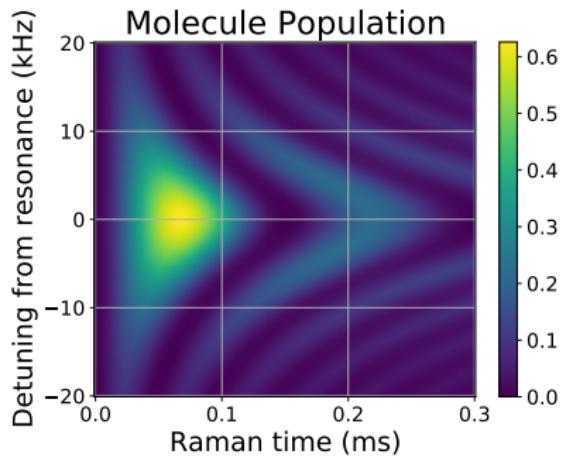
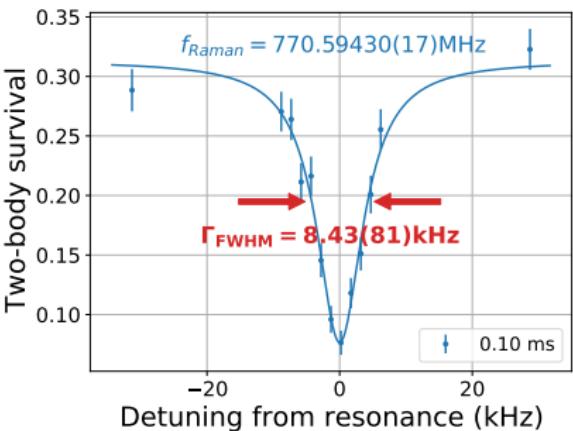
Experiment

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



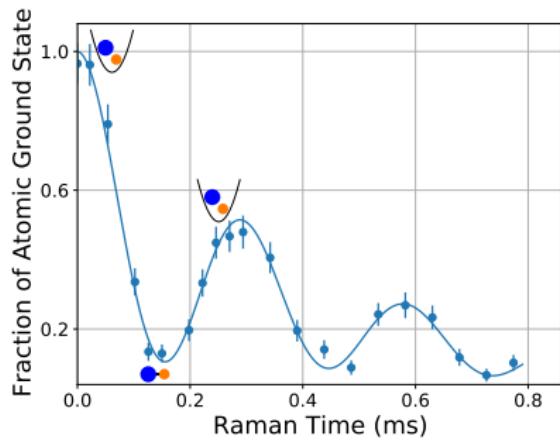
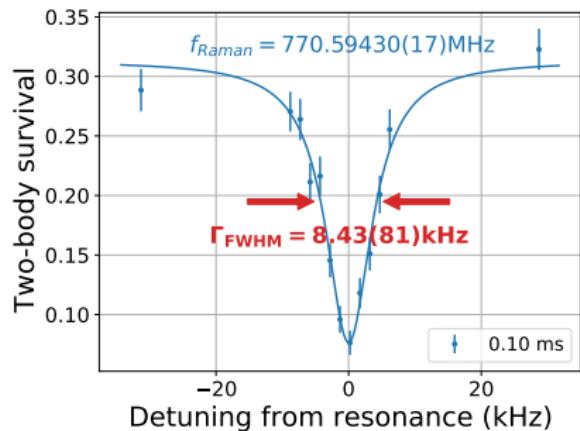
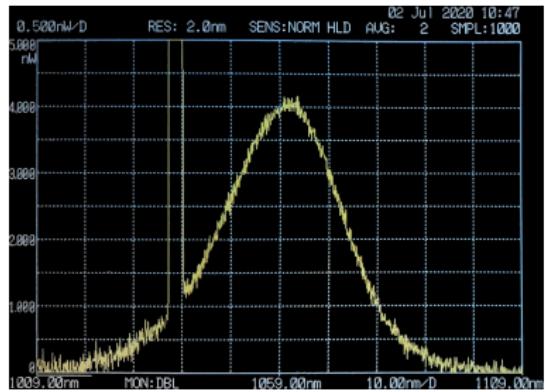
Experiment

- Transferred 63% of ground state atom to molecule.
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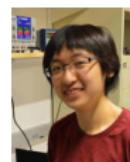
Experiment



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



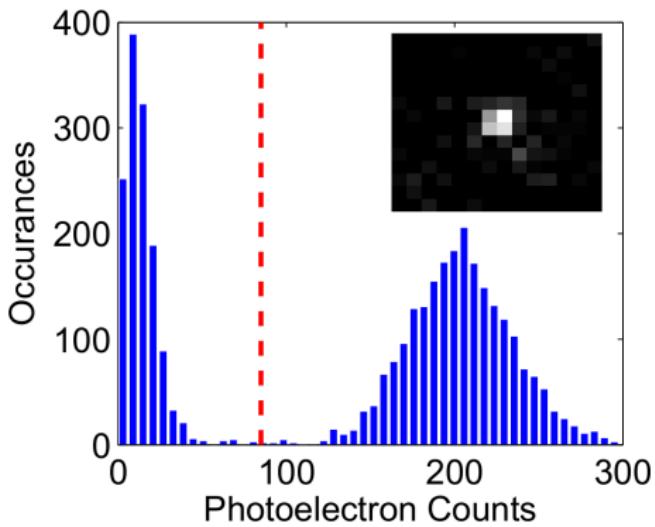
Jeremy Hutson

Single Atom in Tweezer

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

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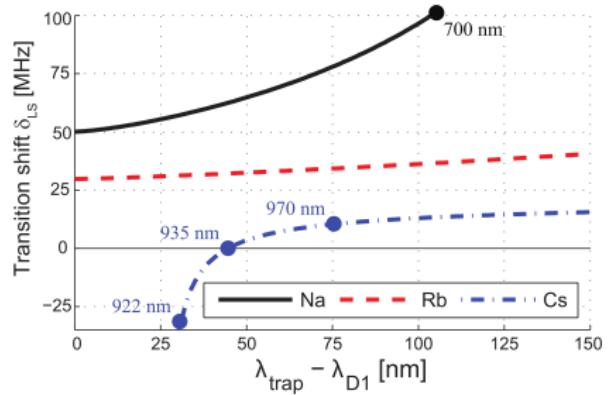
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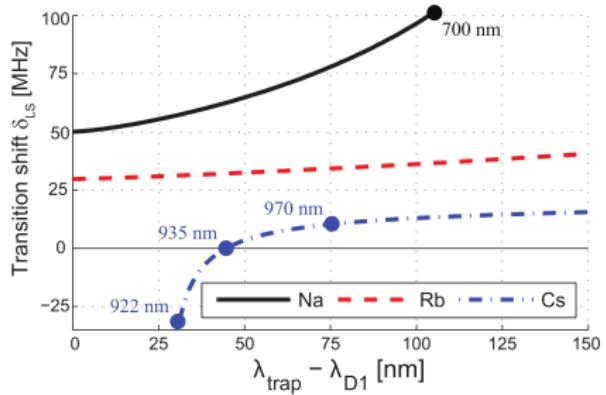
Issues with Na

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

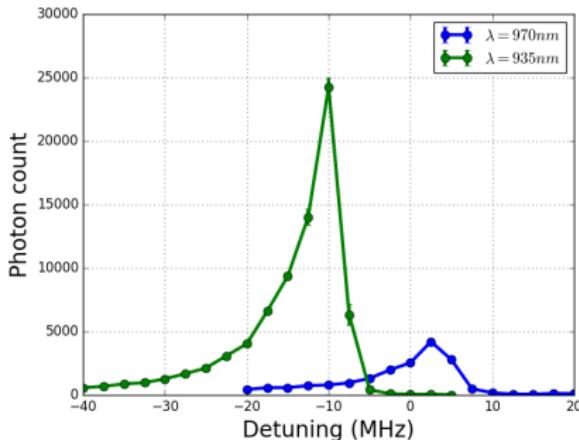
Real Issue with Na: Light Shift



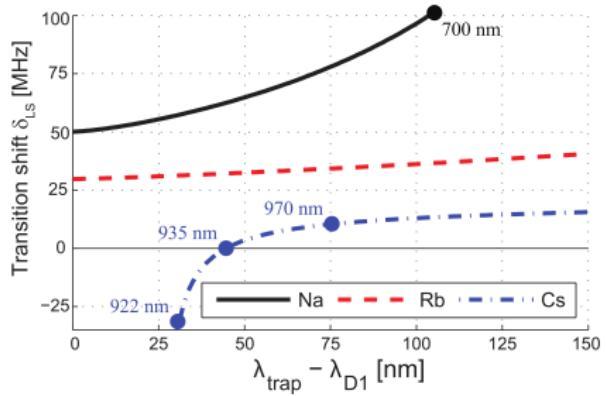
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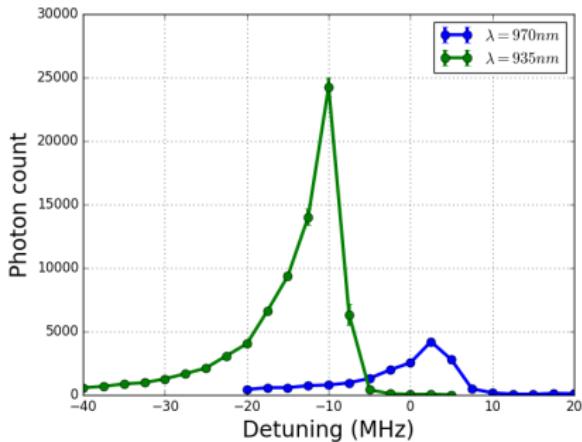
Cs single atom imaging



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Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

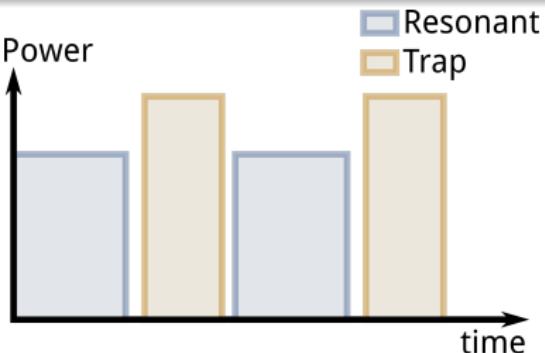
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Trap modulation

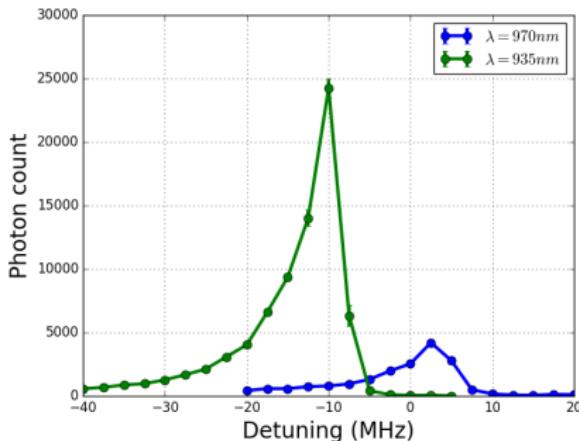
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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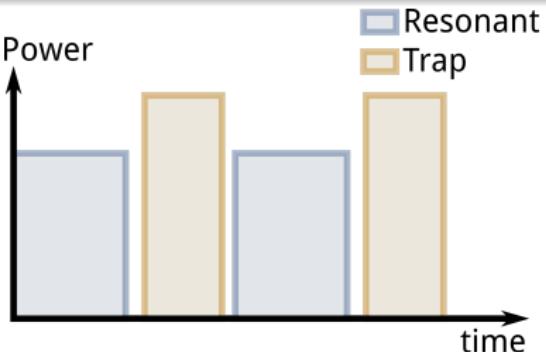
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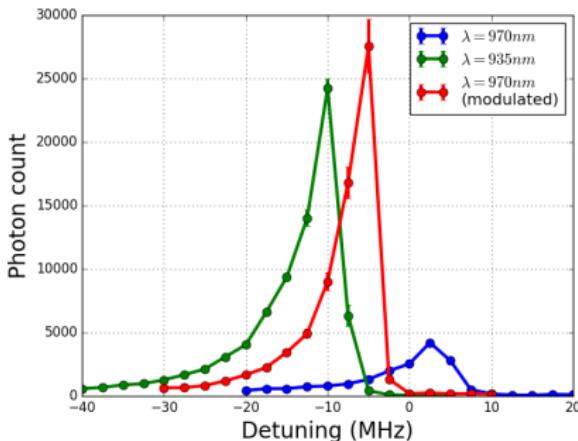
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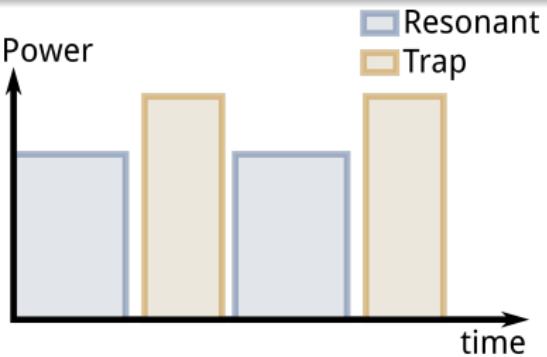
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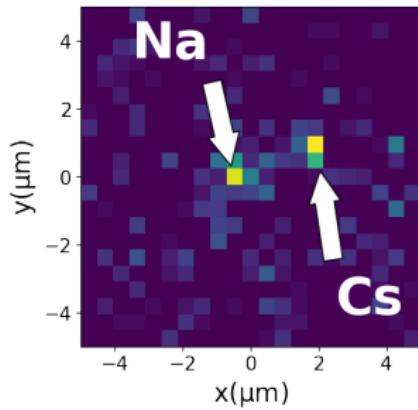
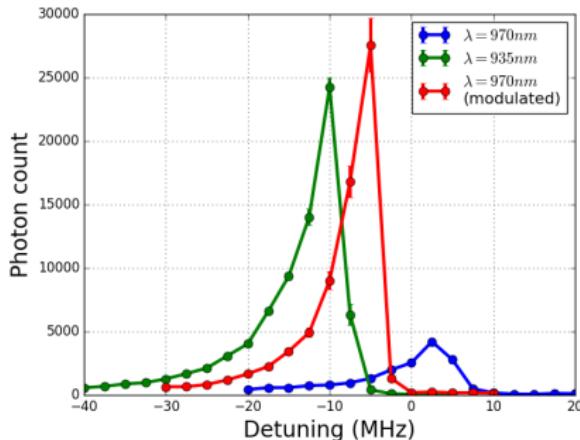
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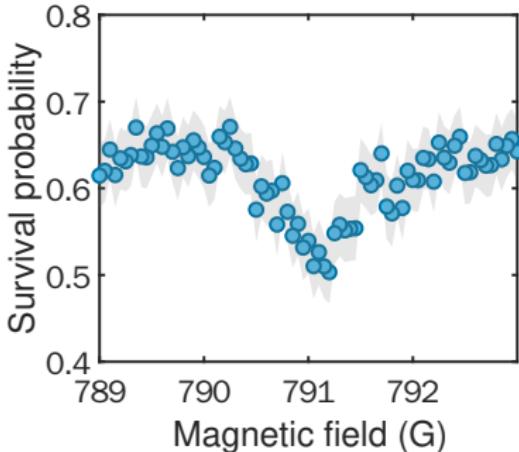
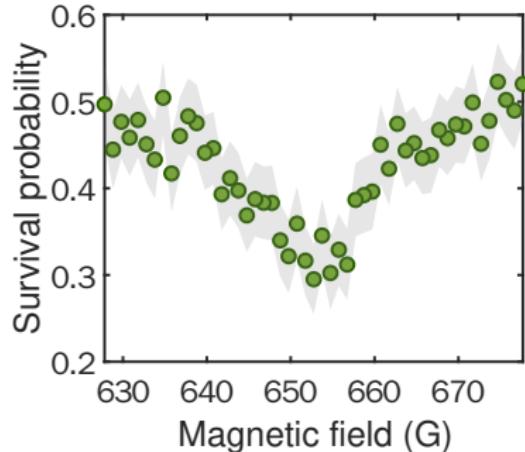
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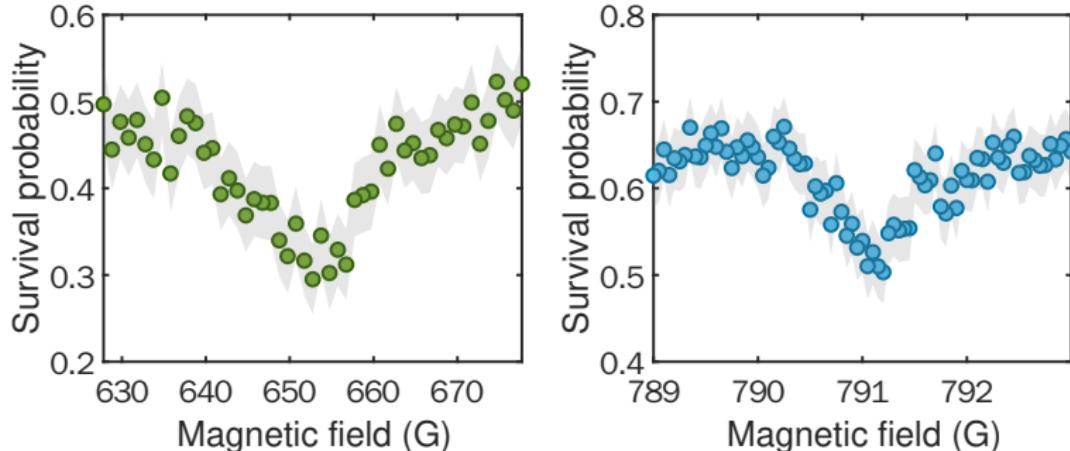
Na (1, -1) Cs (3, -3) Feshbach resonance



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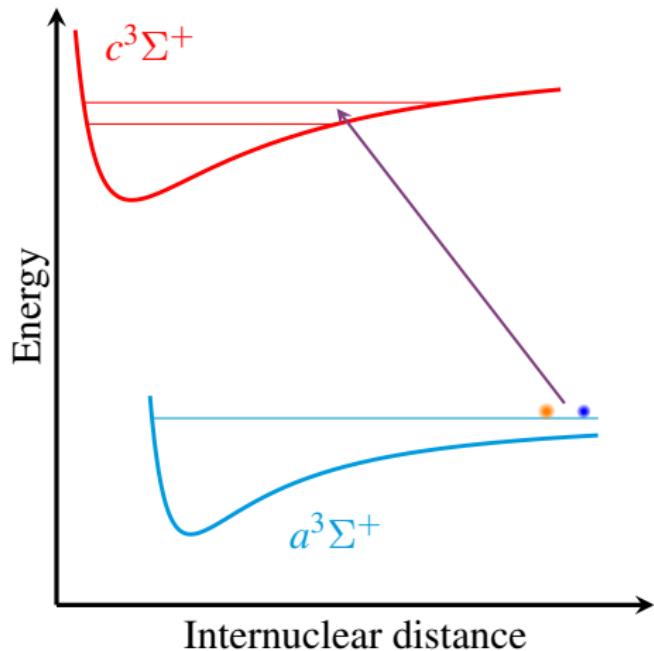
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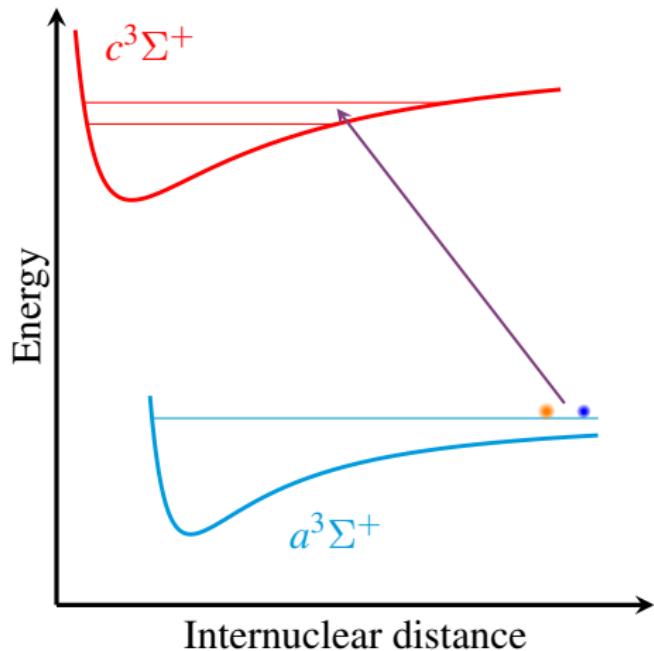
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



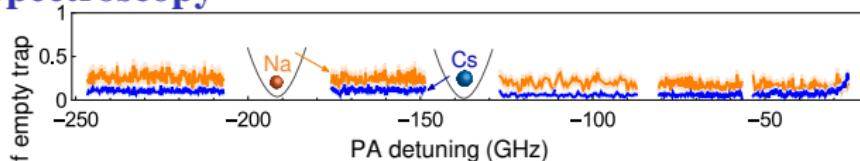
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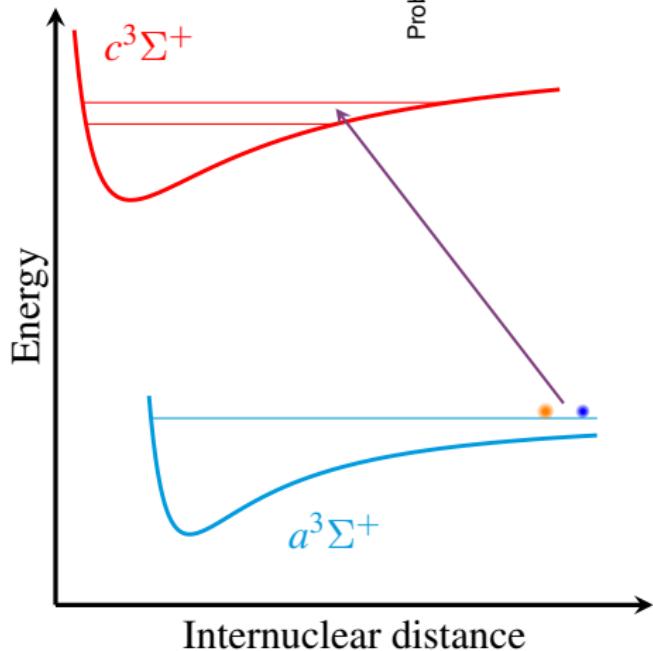
Single Atom PA

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

Photoassociation (PA) Spectroscopy



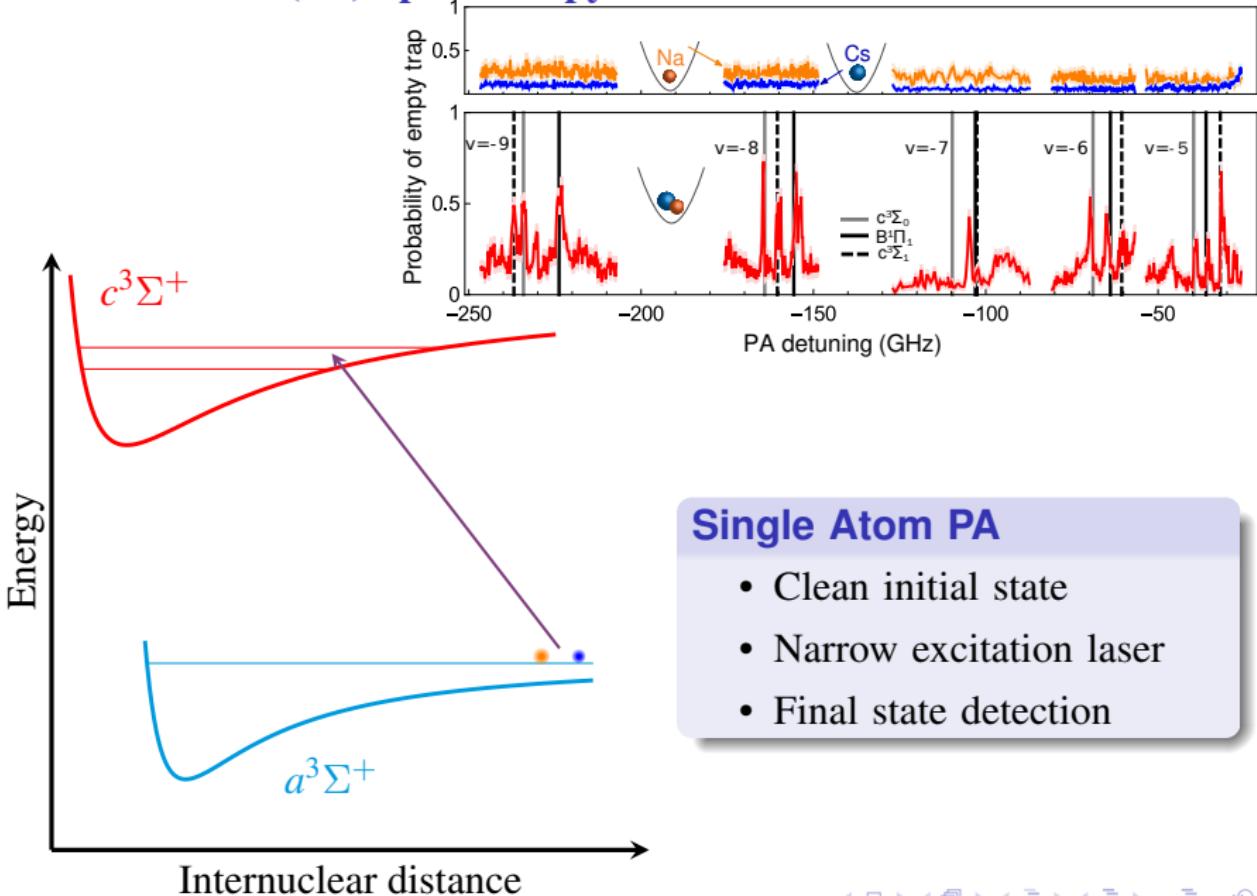
Probability of empty trap



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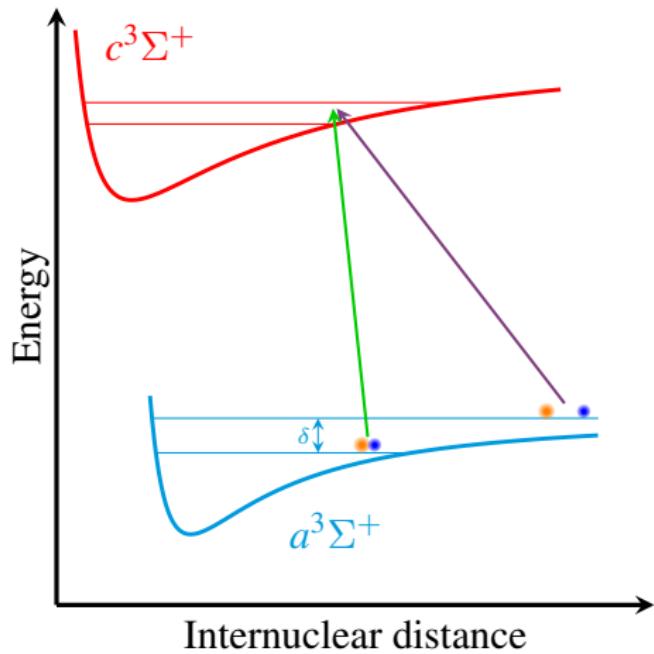
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Electromagnetically Induced Transparency (EIT) Spectroscopy



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