

Coherent Creation of Single Molecules from Single Atoms

Yichao Yu

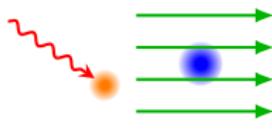
Ni Group/Harvard

Dec. 17, 2020

Simple System

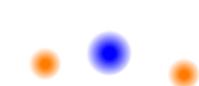


Full Control

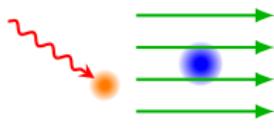


Complex Dynamics

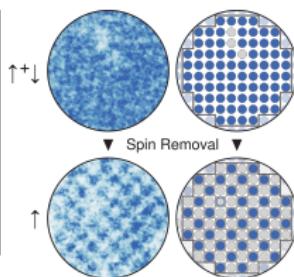
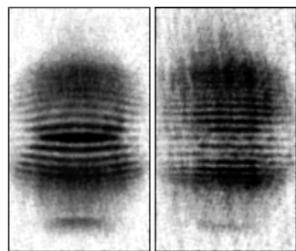
Simple System



Full Control



Complex Dynamics



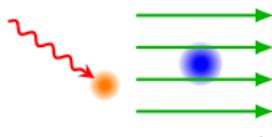
Science 275, 637 (1997)

Nature 545, 462-466 (2017)

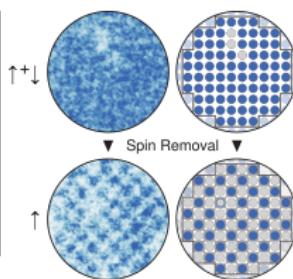
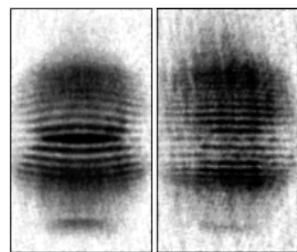
Simple System



Full Control



Complex Dynamics



Science 275, 637 (1997)

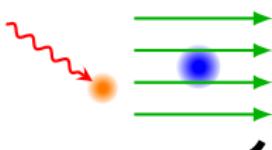
Nature 545, 462-466 (2017)

- ✗ Simple internal structure
- ✗ Weak interaction

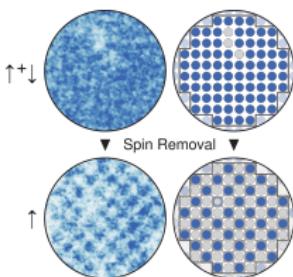
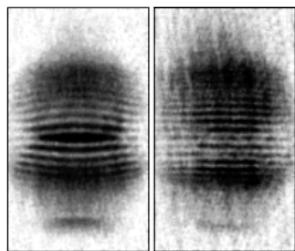
Simple System



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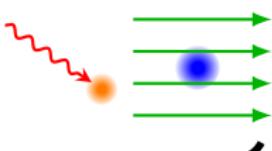
- Strong interaction
- Long coherence time
- Rich internal structure
- Fully controllable

- Simple internal structure
- Weak interaction

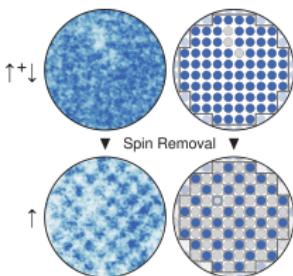
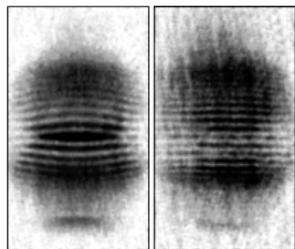
Simple System



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Complex Dynamics



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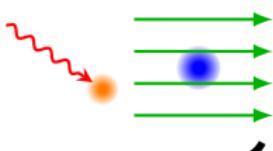
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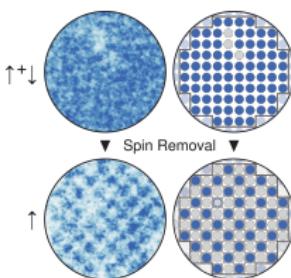
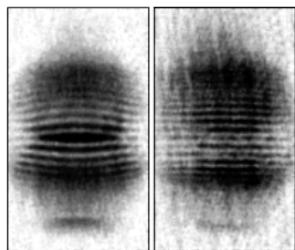
Simple System



Full Control



Complex Dynamics



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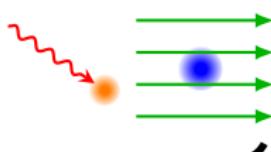
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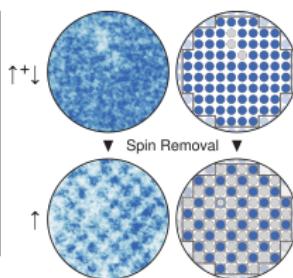
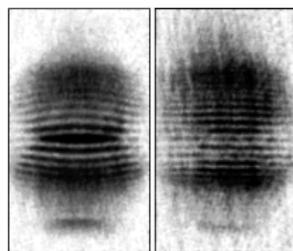
Simple System



Full Control



Complex Dynamics



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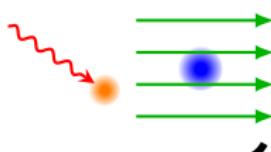
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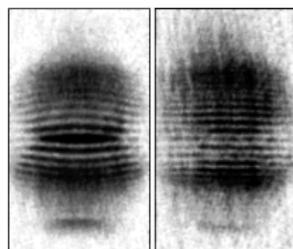
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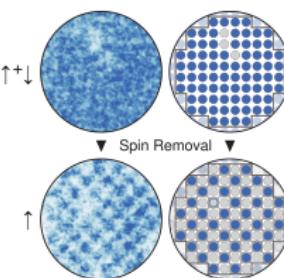
Full Control



Complex Dynamics

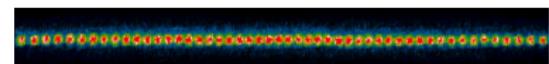


Science 275, 637 (1997)



Nature 545, 462-466 (2017)

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Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

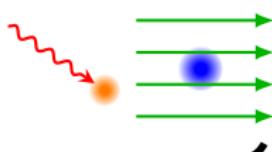
New System?

- Simple internal structure
- Weak interaction

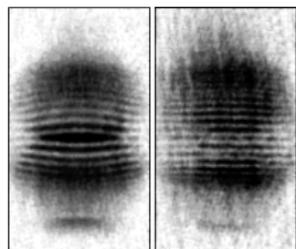
Simple System



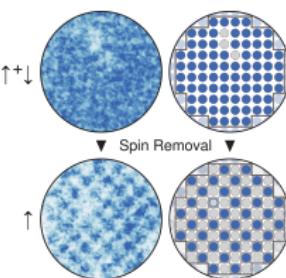
Full Control



Complex Dynamics

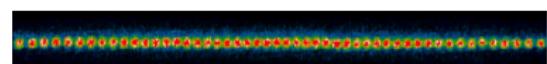


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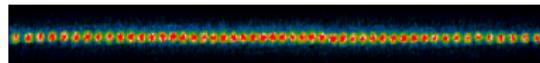


Rydberg Atoms (Photo credit: Lukin group)

New System!

- Different properties
- New tools and techniques

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



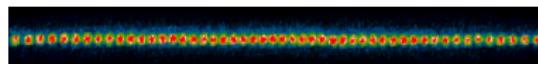
Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

✓ Strong interaction (kHz)

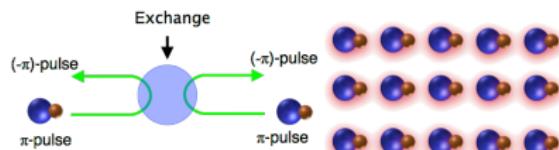
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Ions (Photo credit: Monroe group)

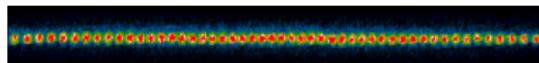


Rydberg Atoms (Photo credit: Lukin group)



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

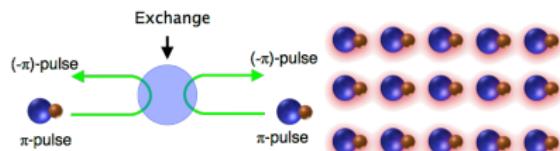
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Ions (Photo credit: Monroe group)

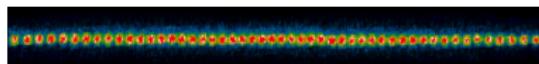


Rydberg Atoms (Photo credit: Lukin group)



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

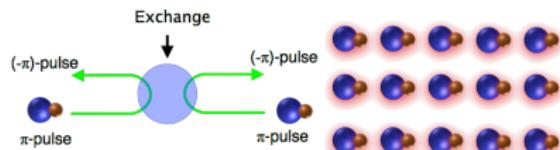
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Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

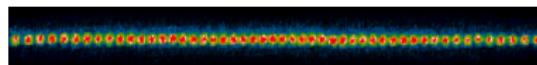


Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

- ✓ Strong interaction (kHz)
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Optical tweezers

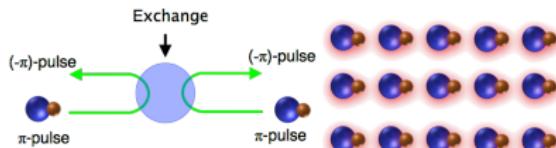
- Single site resolution
- . . .



Ions (Photo credit: Monroe group)



Rydberg Atoms (Photo credit: Lukin group)

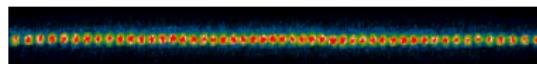


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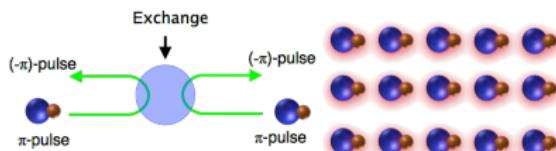
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Ions (Photo credit: Monroe group)

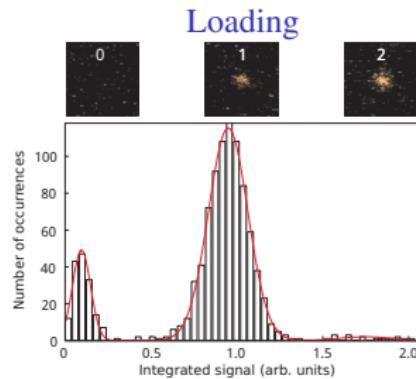


Rydberg Atoms (Photo credit: Lukin group)



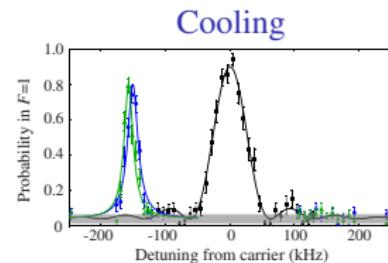
Dipolar Molecule (Chemical Science 9, 6830 - 6838 (2018))

- ✓ Strong interaction (kHz)
- ✓ Long coherence time
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Nat. Phys. 6, 951 (2010)

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



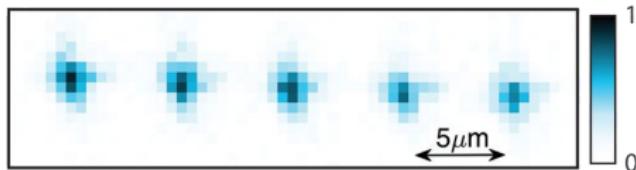
PRX. 2, 041014 (2012)



Science 354, 1024 (2016)

Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

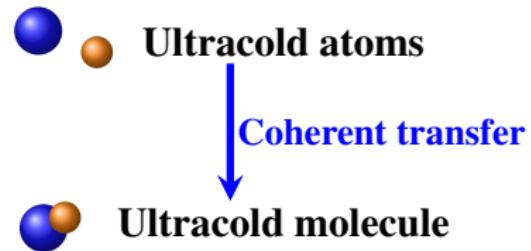
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



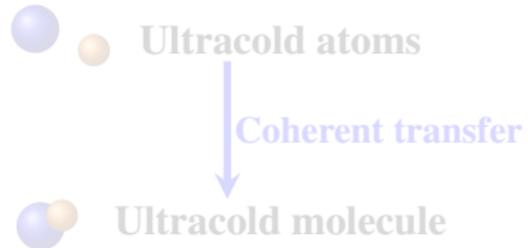
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezer
- Quantum control

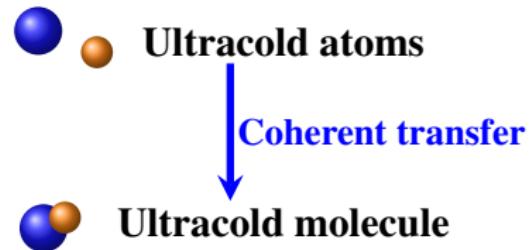
Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in 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

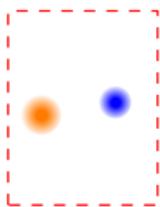
- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

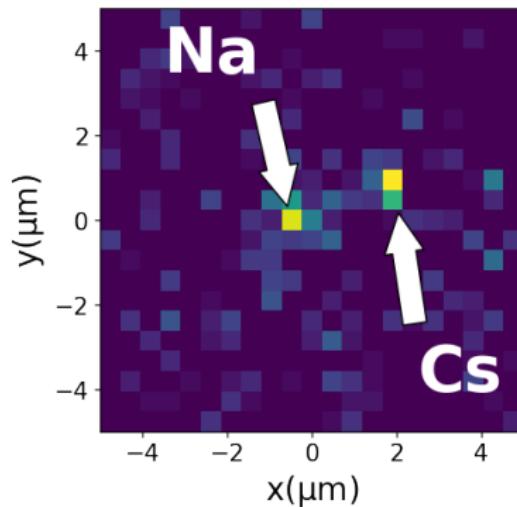
NaCs molecule

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

Experiment overview



Loading

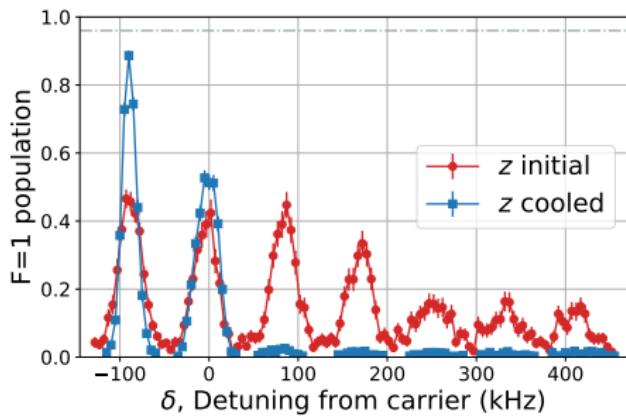


Loading probability per site: 60%
Post select on initial and final state.

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

Experiment overview

Cooling



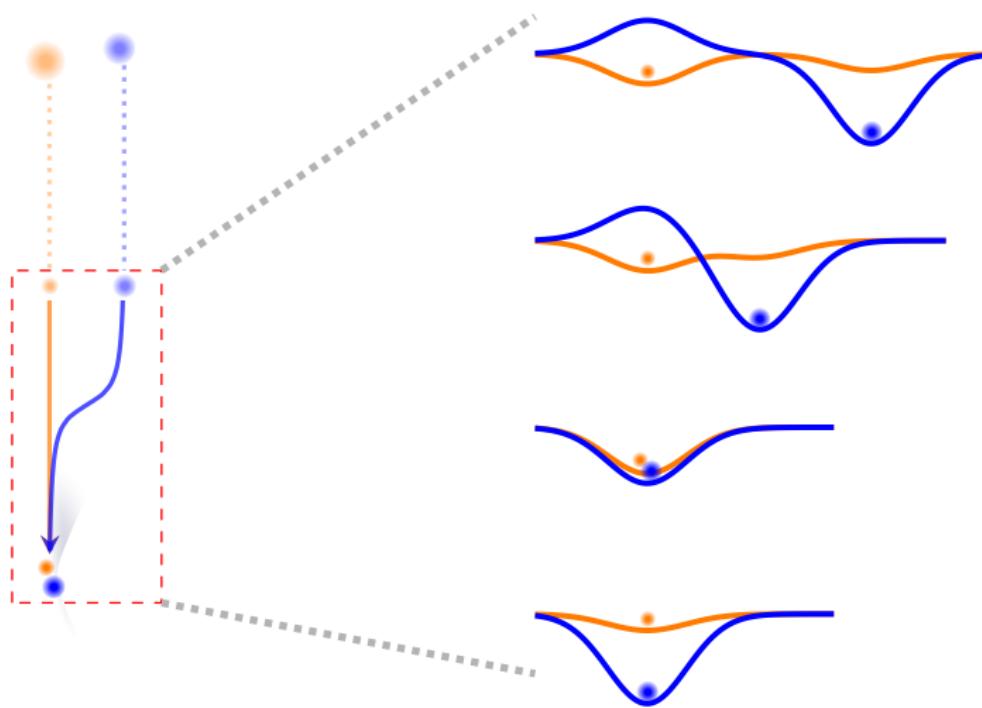
Cs: 96% ground state¹
Na: 94% ground state²

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

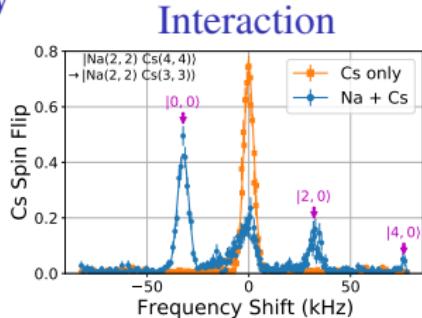
2: **Y. Yu et al.**, PRA 97, 063423 (2018)

Experiment overview

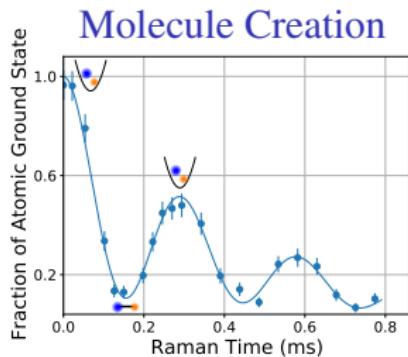
Merging



Experiment overview



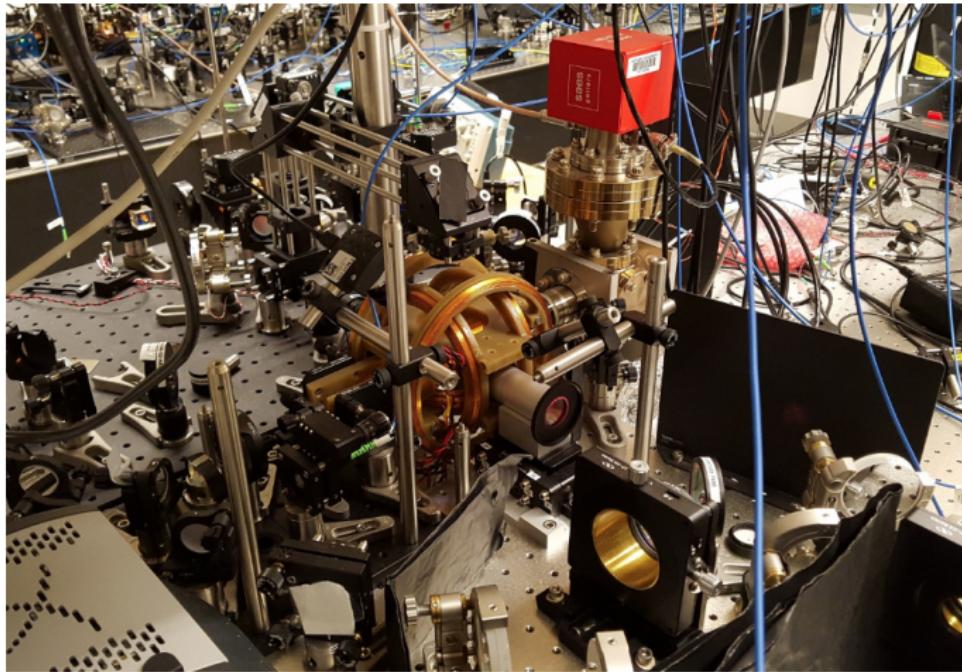
J. D. Hood, Y. Yu et al.,
Phys. Rev. Research 2, 023108 (2020)

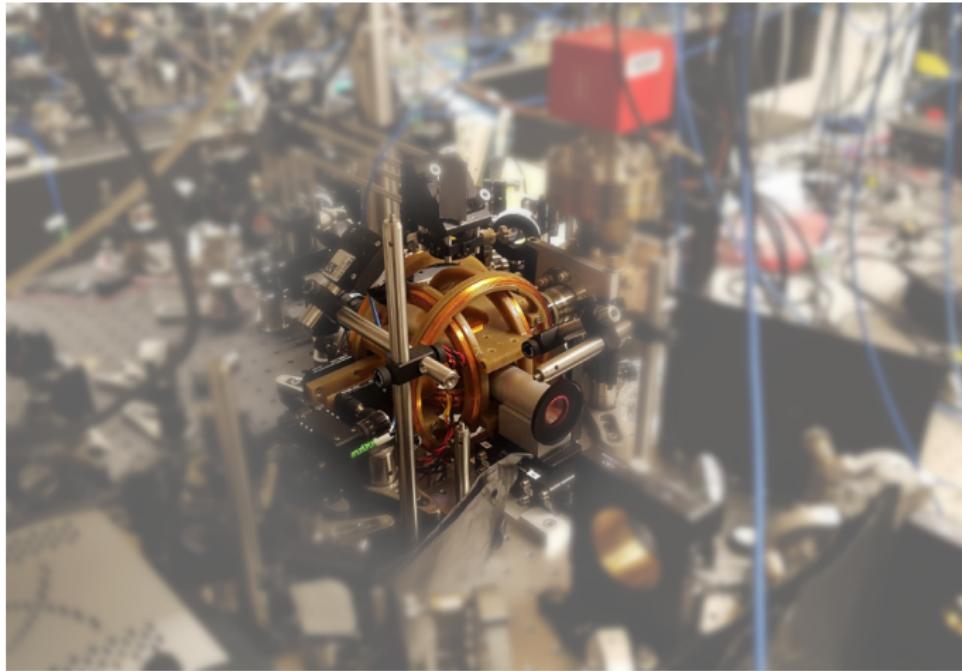


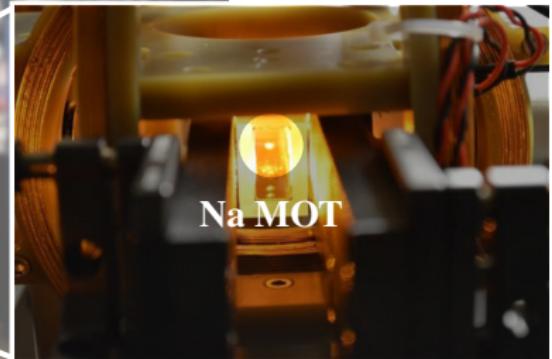
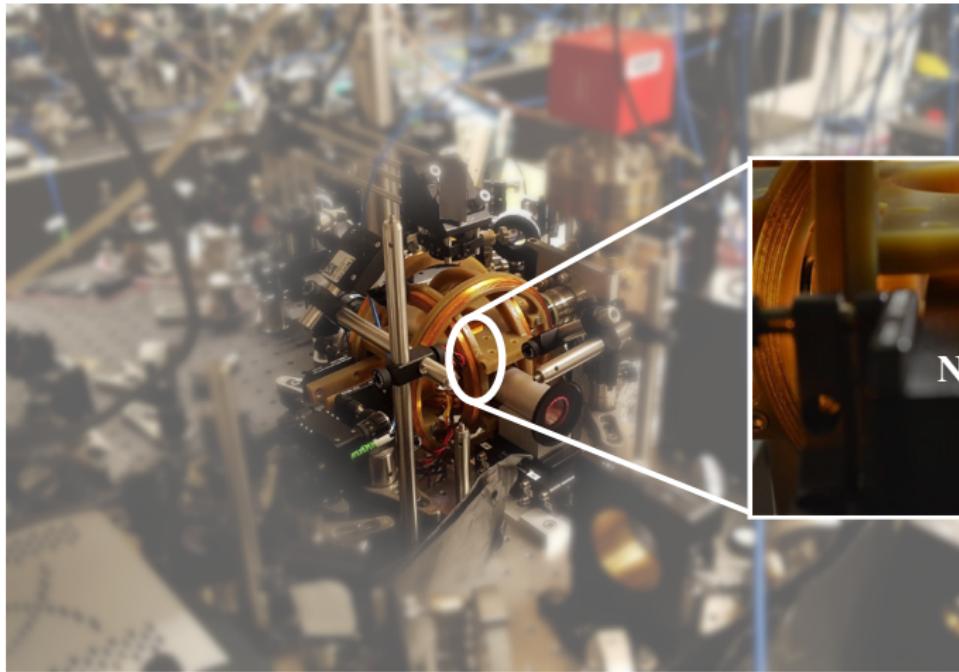
Y. Yu et al., arXiv:2012.09043 (2020)



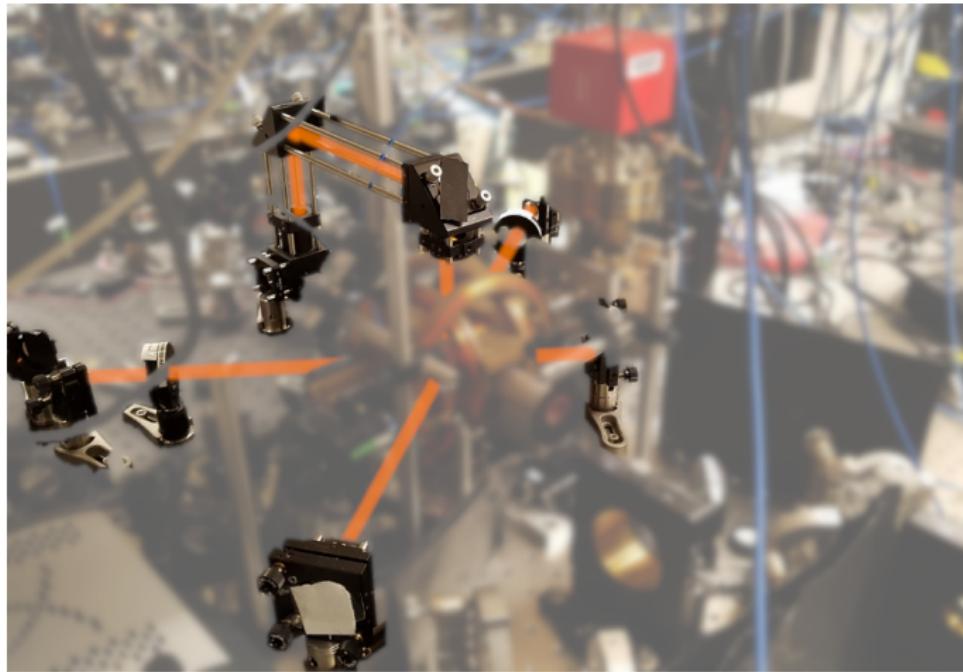
L. R. Liu, J. D. Hood, Y. Yu et al.,
Science 360, 6391 (2018)



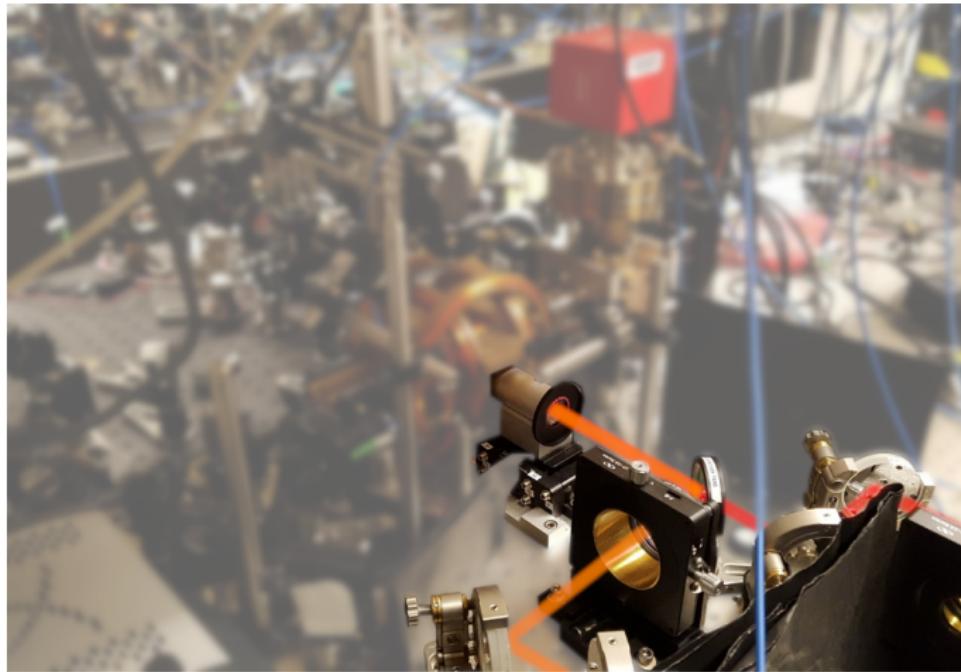




MOT beam path



Tweezer and imaging beam path



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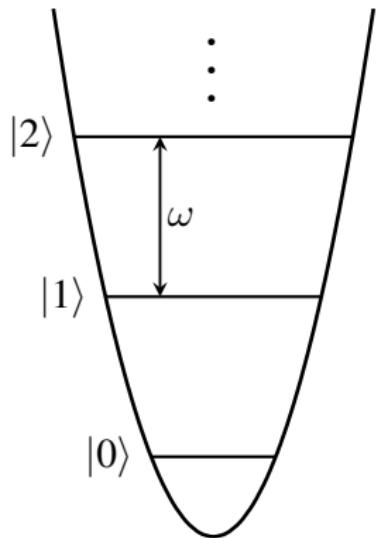
4 Conclusion

Raman sideband cooling

Used for cooling in trap

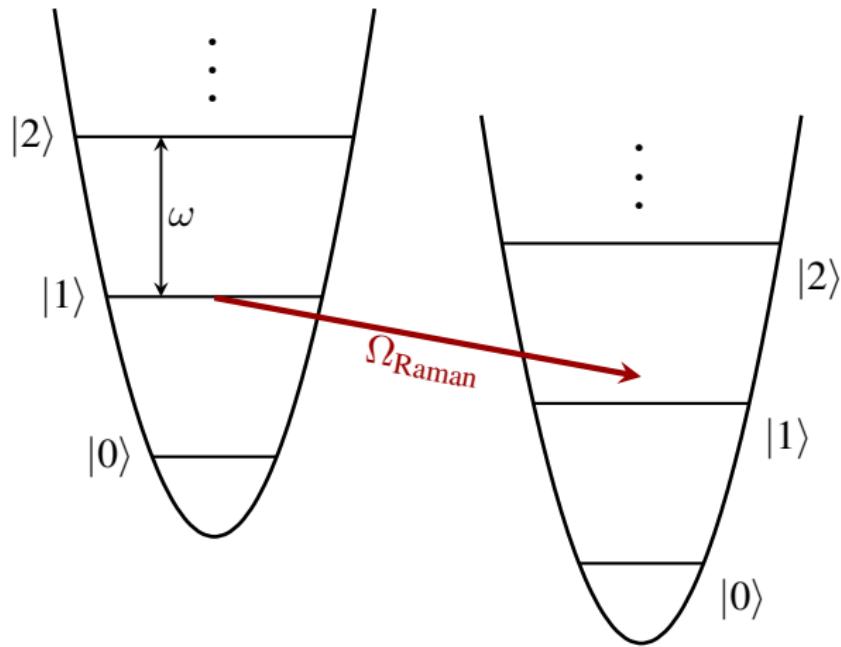
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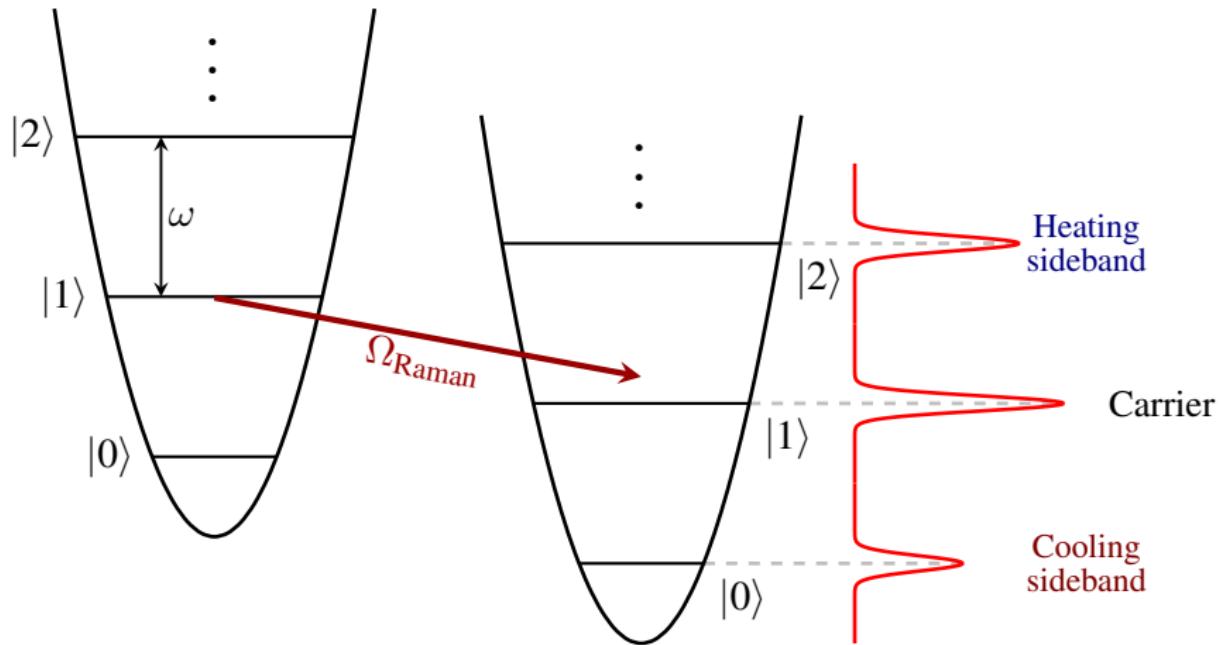
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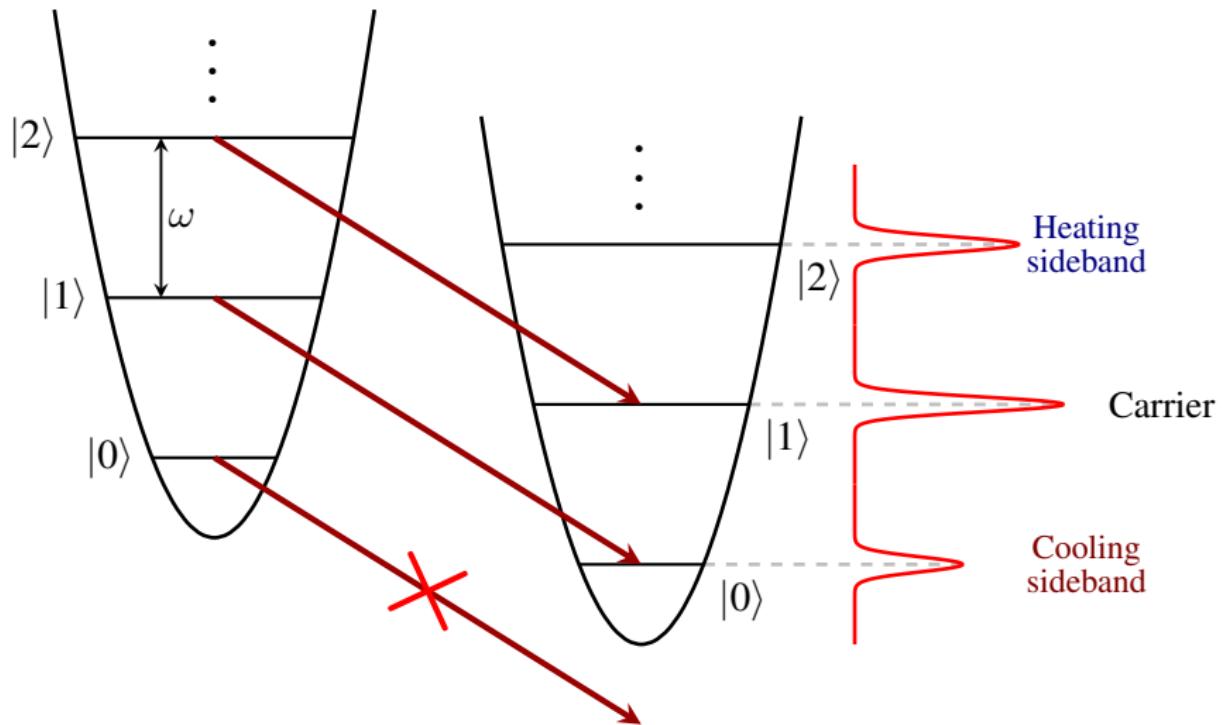
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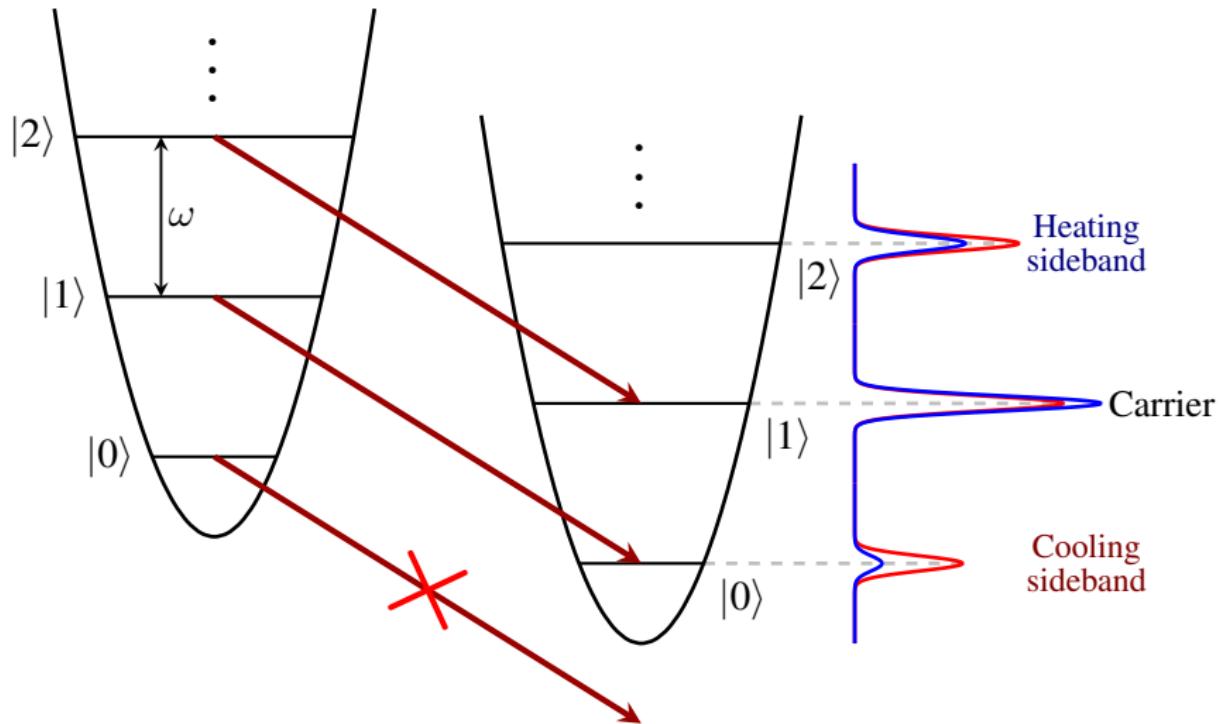
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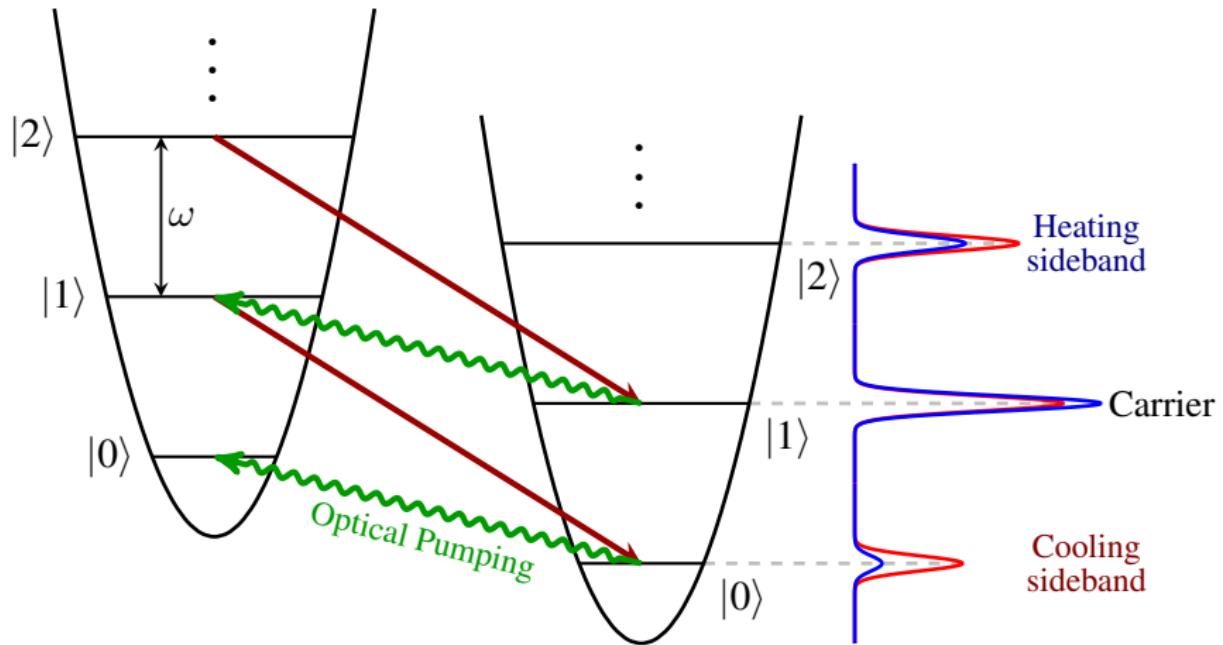
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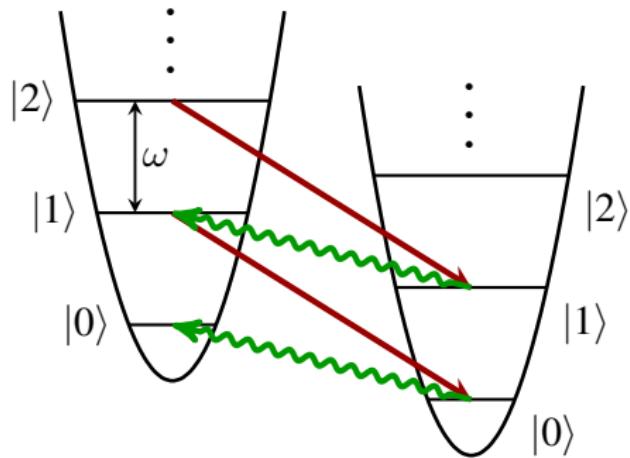


Raman sideband cooling

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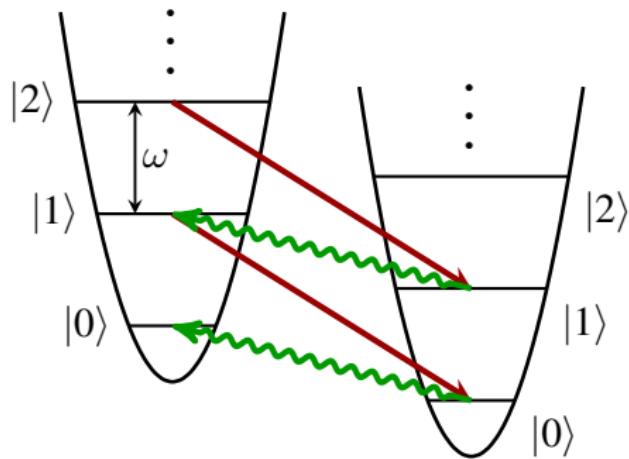
Raman sideband cooling



Raman sideband cooling

Lamb Dicke parameter

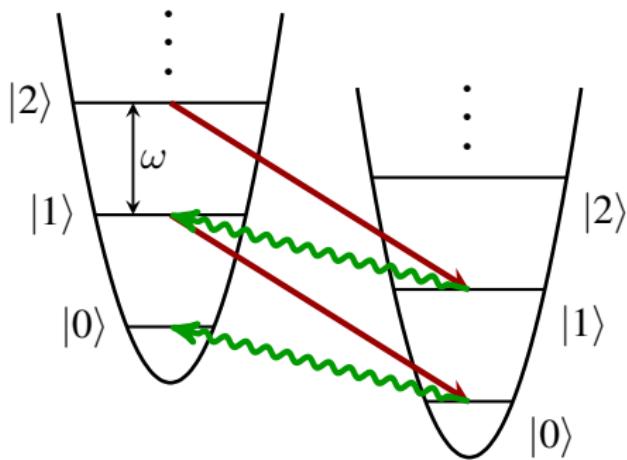
$$\eta \equiv \frac{2\pi z_0}{\lambda} = \sqrt{\frac{\omega_{\text{recoil}}}{\omega_{\text{trap}}}} = \frac{\pi}{\lambda} \sqrt{\frac{2\hbar}{m\omega}}$$



Raman sideband cooling

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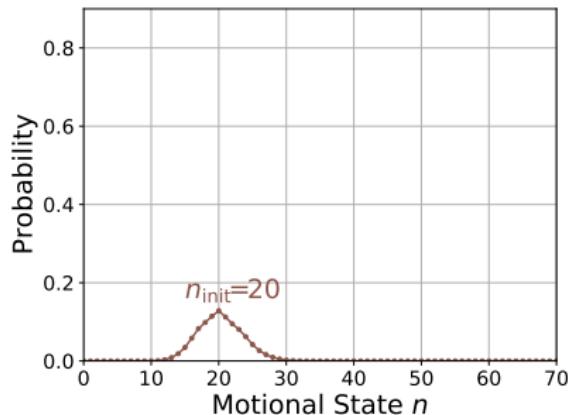


$$\eta_{\text{Na}}^{\text{OP}} = 0.55$$

$$T_{\text{init}} = 80 \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



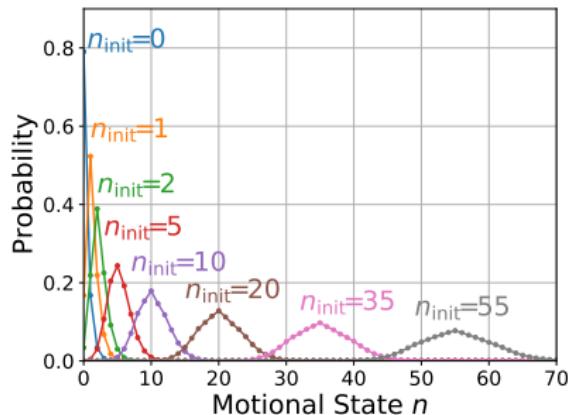
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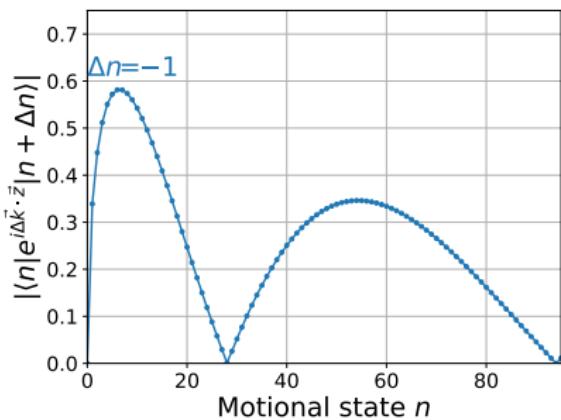
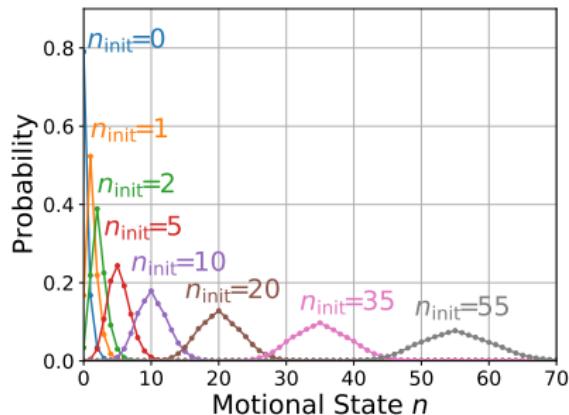
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$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \text{ } \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



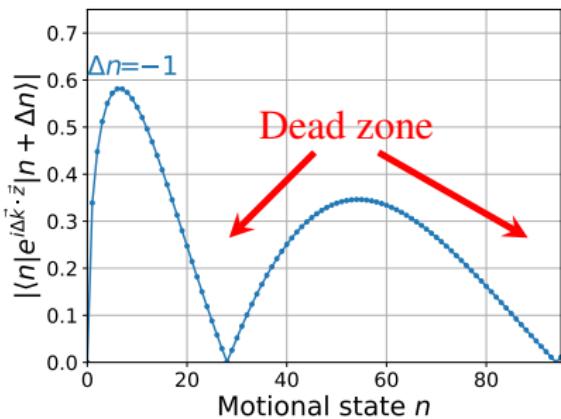
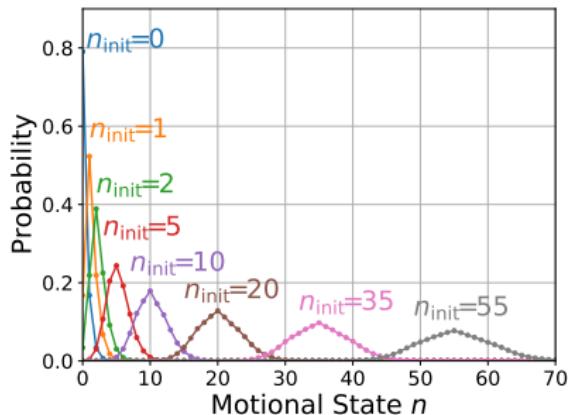
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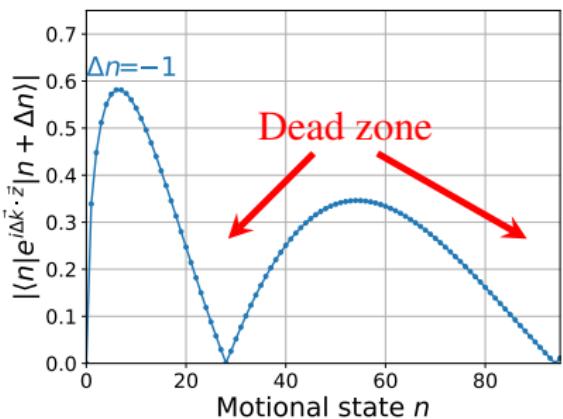
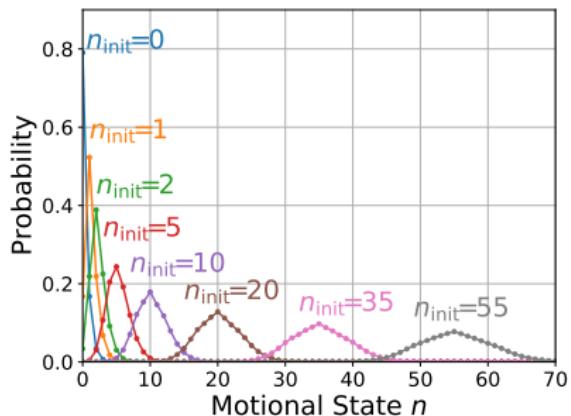
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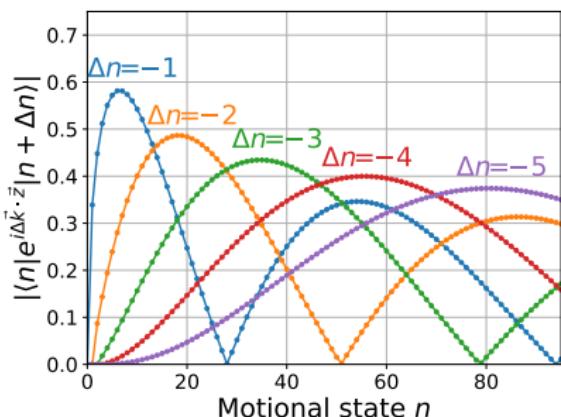
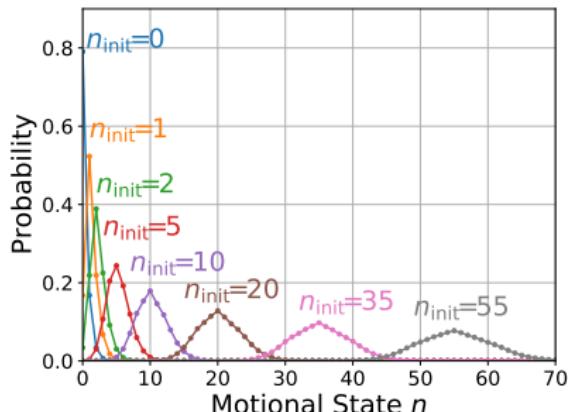
$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \mu\text{K}$$

- Motional state branching
- Coupling “dead zone”

Solution

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

Raman sideband cooling



Lamb Dicke parameter

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

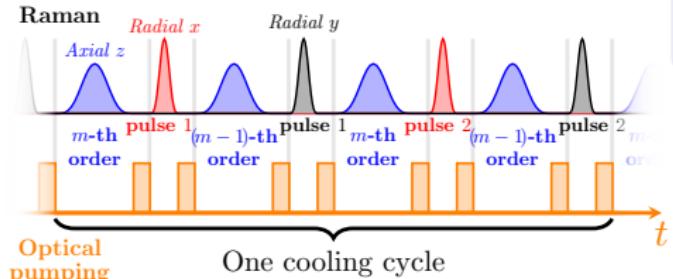
$$\eta_{\text{Na}}^{\text{OP}} = 0.55 \quad T_{\text{init}} = 80 \mu\text{K}$$

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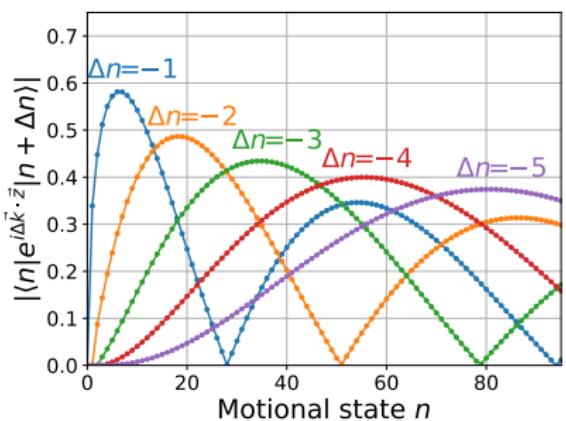


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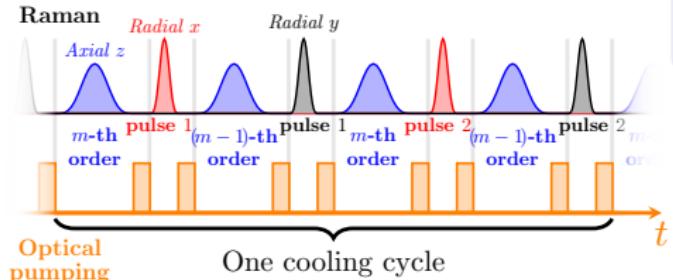
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Raman sideband cooling



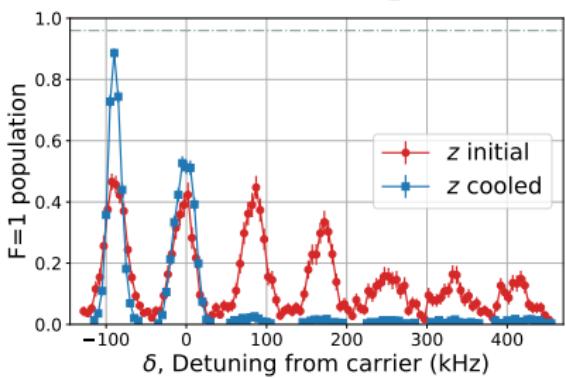
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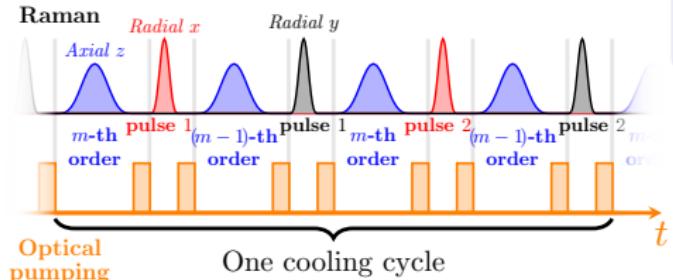
Axial sideband spectrum



Solution

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

Raman sideband cooling



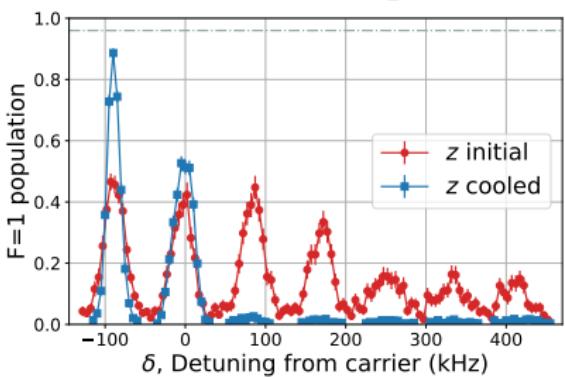
Lamb Dicke parameter

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Axial sideband spectrum



Solution

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

3D ground state: 93.5(7)%

PRA 97, 063423 (2018)

Outline

1 Experiment overview

2 Atom state control

- Raman sideband cooling of Na atoms

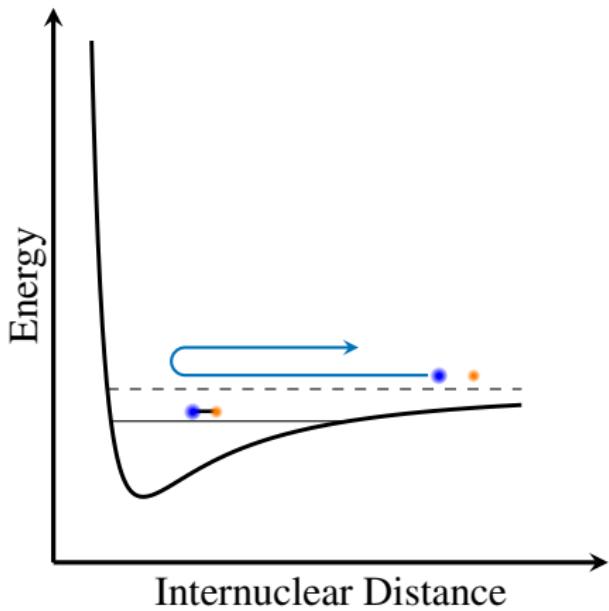
3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

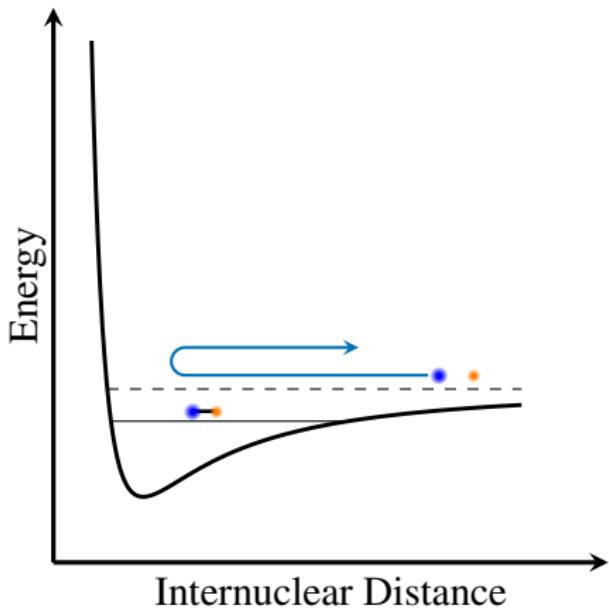
Scattering length a

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



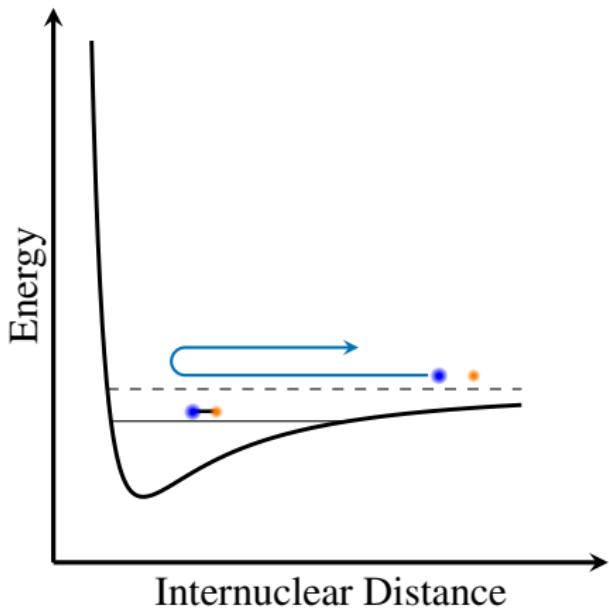
Scattering length a

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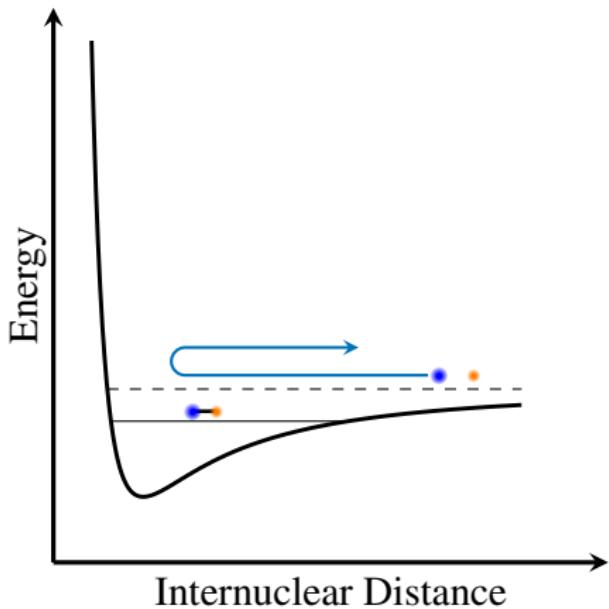
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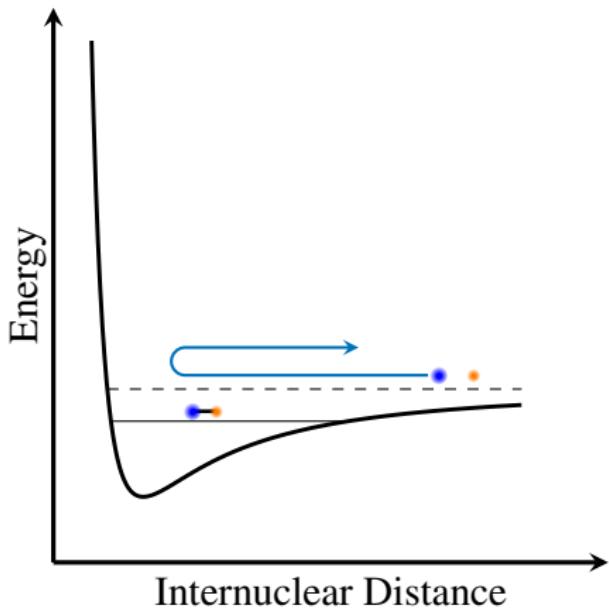
Scattering length a

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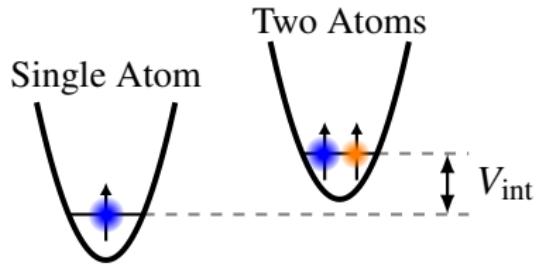


Scattering length a

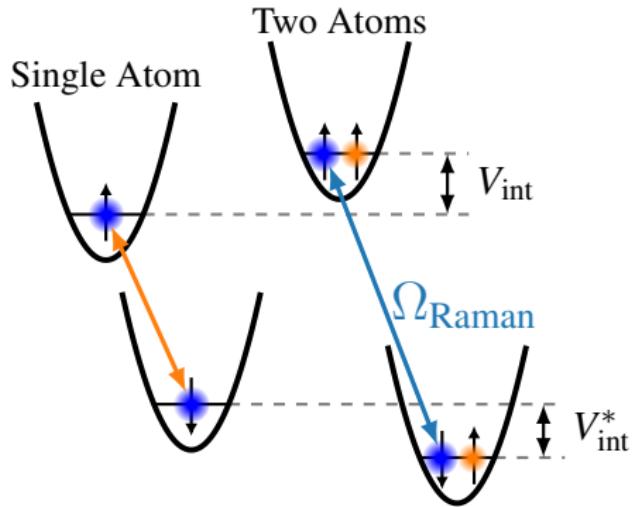
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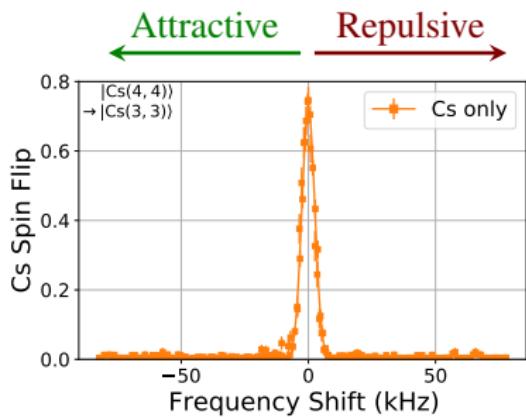
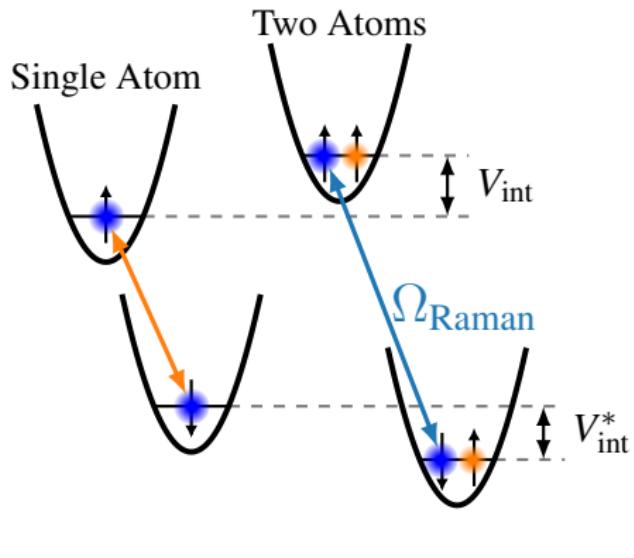
Interaction shift



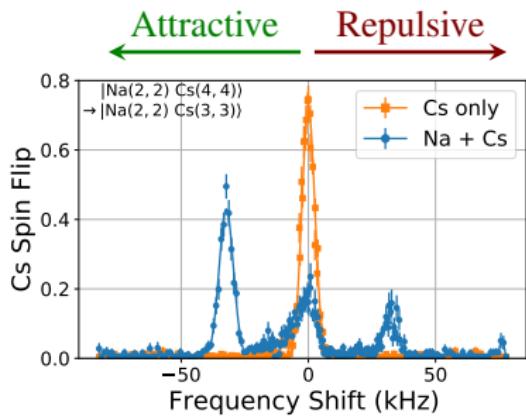
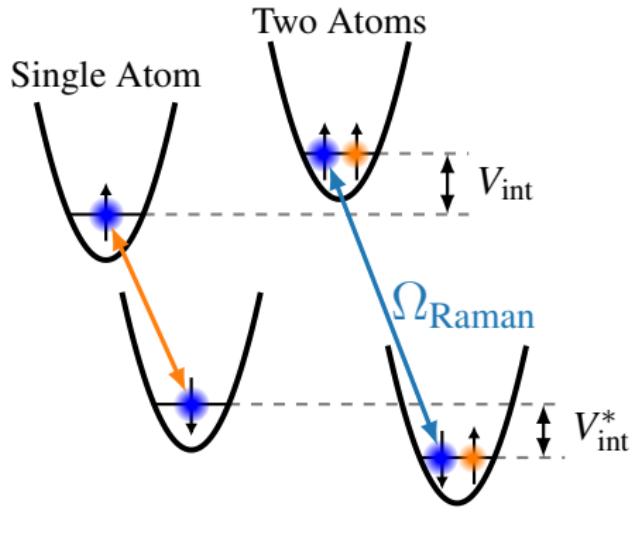
Interaction shift



Interaction shift



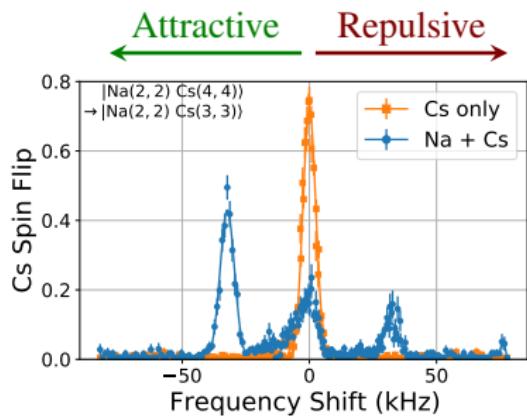
Interaction shift



Interaction shift

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

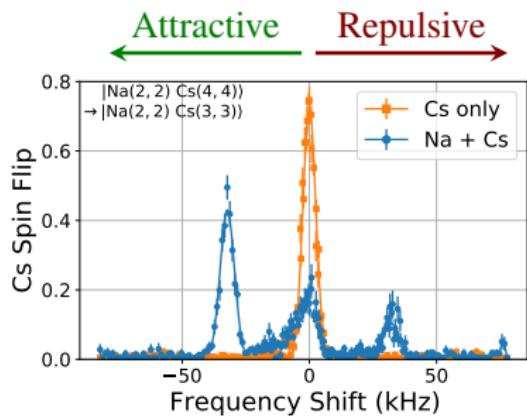
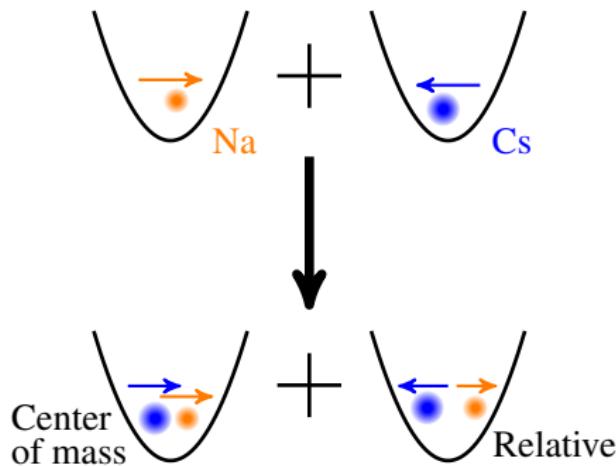
Interaction



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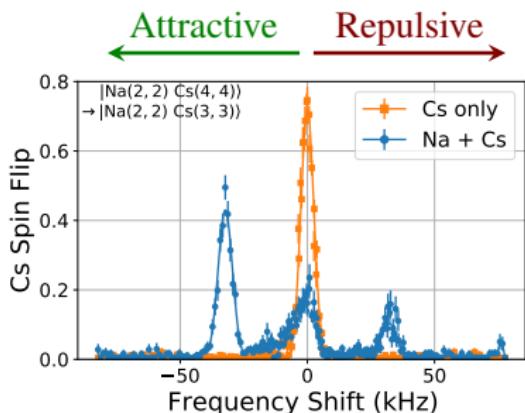
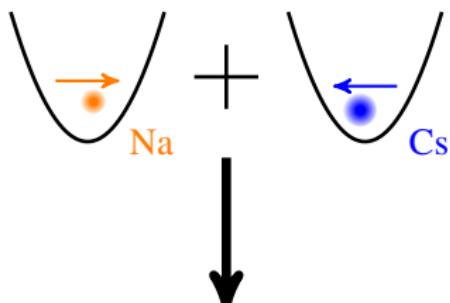
Interaction



Interaction shift

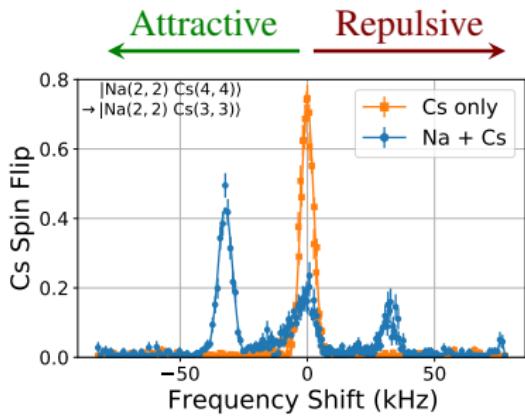
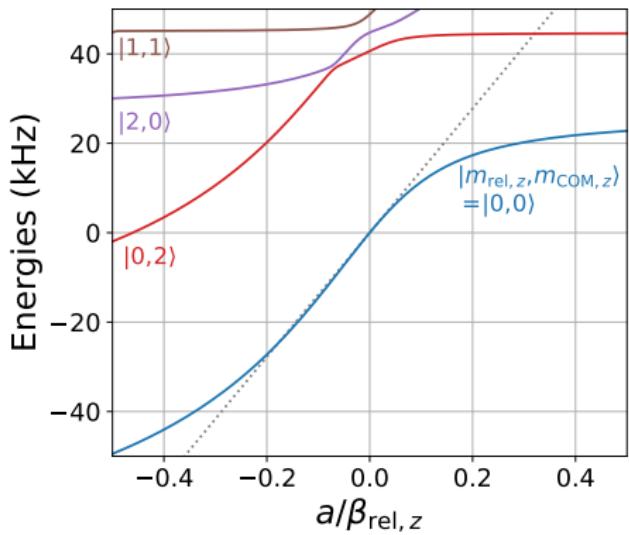
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Interaction



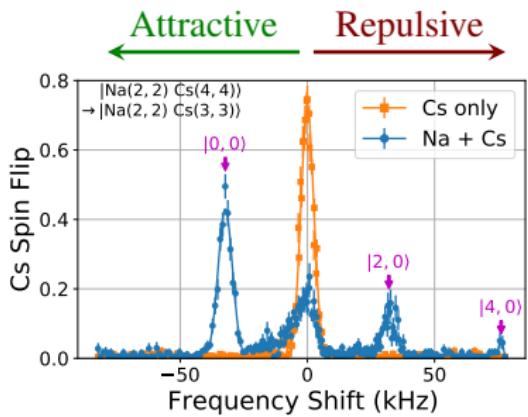
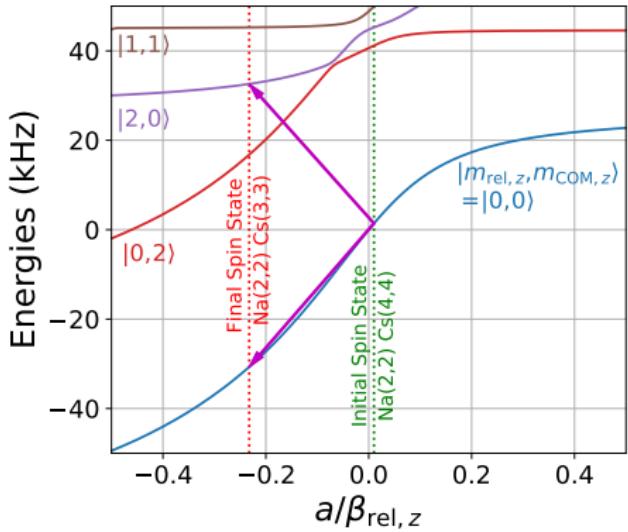
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M \Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{\text{int}}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z}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

Interaction shift



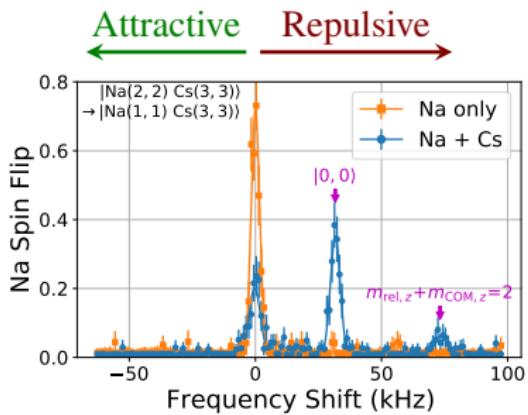
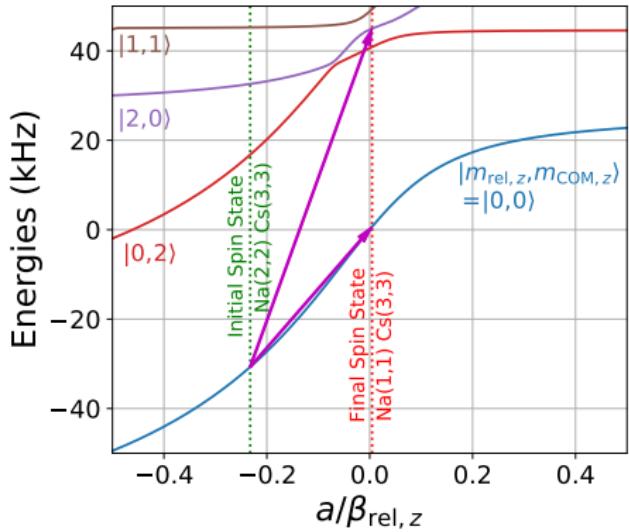
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Interaction shift



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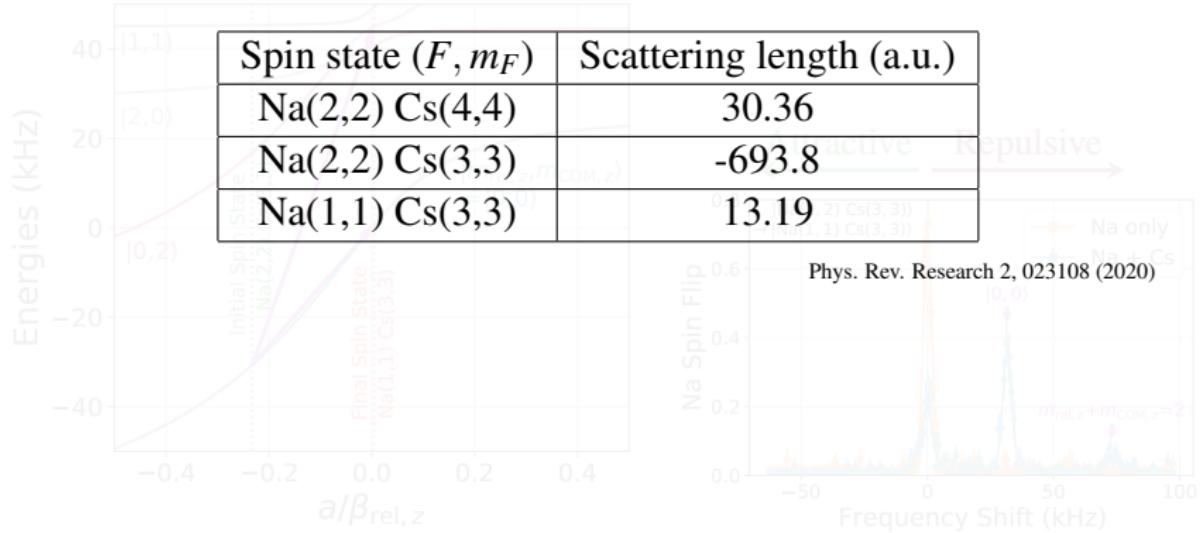
Interaction shift



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Interaction shift

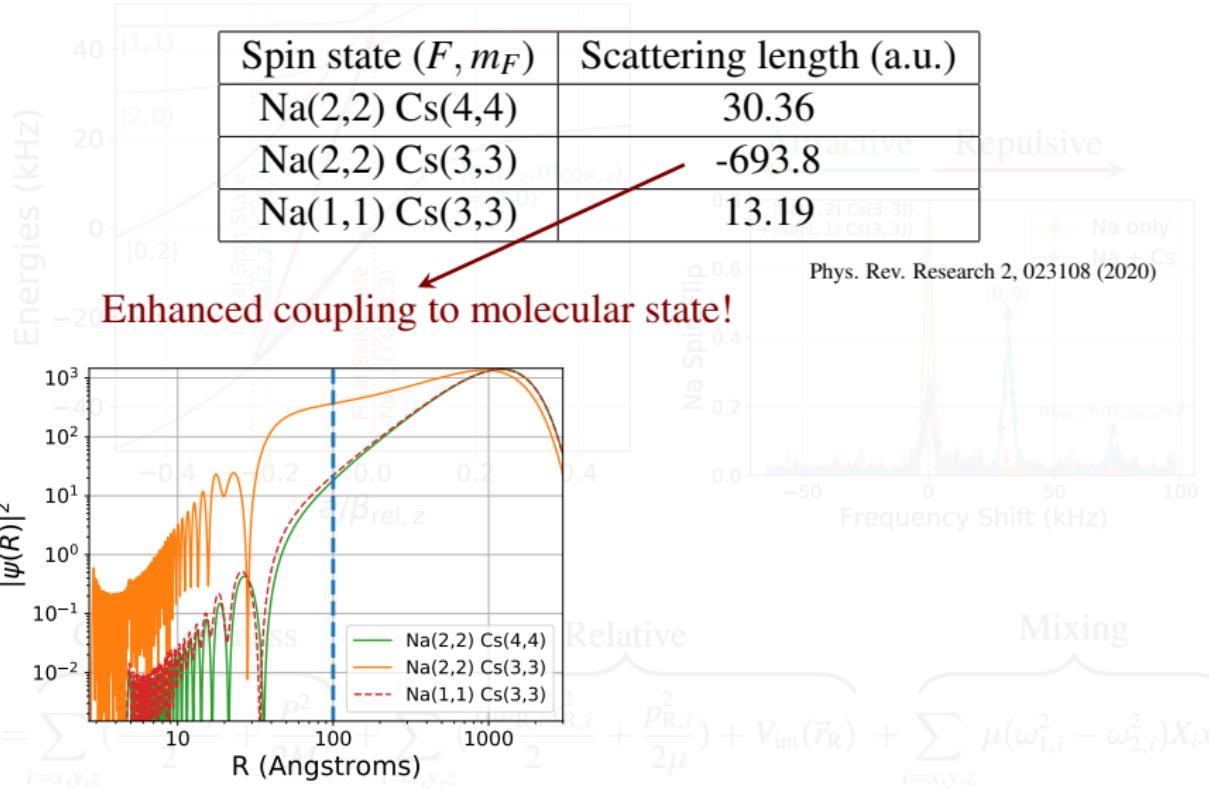
Combined with binding energy measurement on Na(2,2) Cs(4,4)



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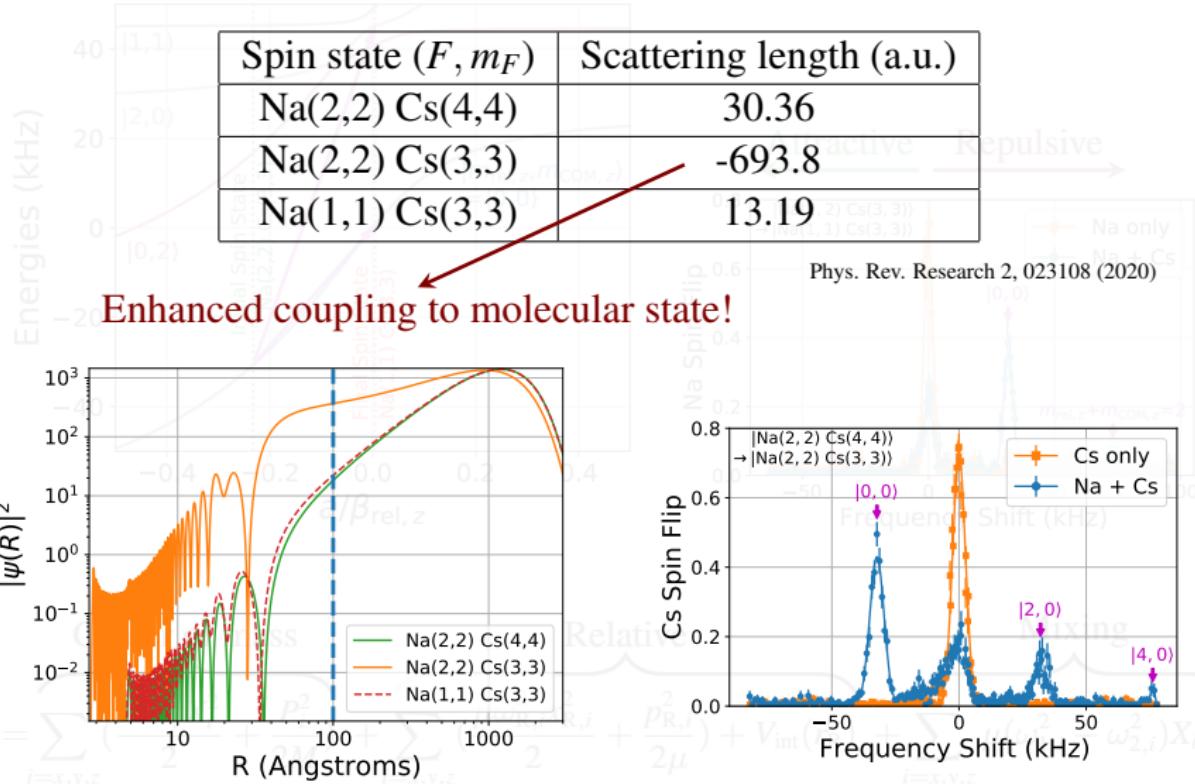
Interaction shift

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Interaction shift

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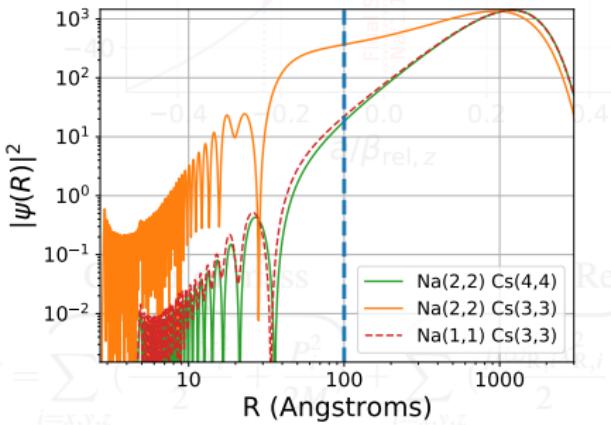


Interaction shift

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

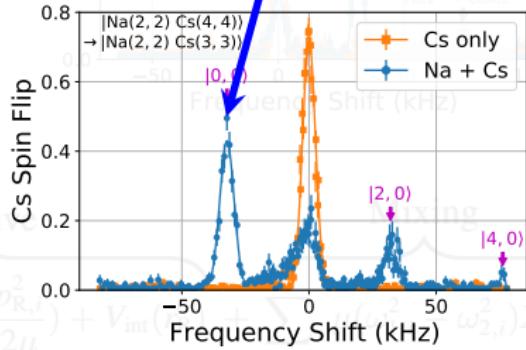
Spin state (F, m_F)	Scattering length (a.u.)
Na(2,2) Cs(4,4)	30.36
Na(2,2) Cs(3,3)	-693.8
Na(1,1) Cs(3,3)	13.19

Enhanced coupling to molecular state!



Phys. Rev. Research 2, 023108 (2020)

Ground State Only



Outline

1 Experiment overview

2 Atom state control

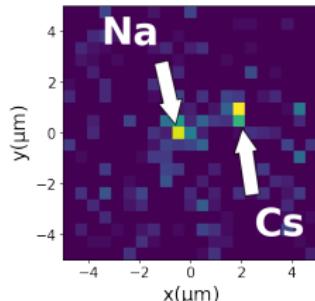
- Raman sideband cooling of Na atoms

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- Coherent optical transfer

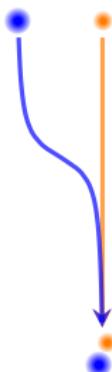
4 Conclusion

Loading

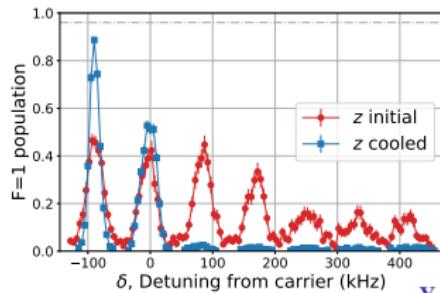


N. R. Hutzler, L. R. Liu, Y. Yu et al.,
New J. Phys. 19, 023007 (2017)

Merging

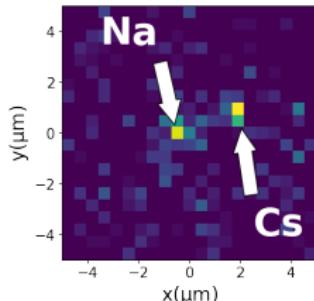


Cooling



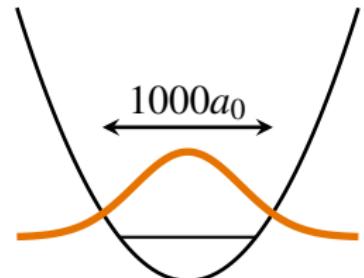
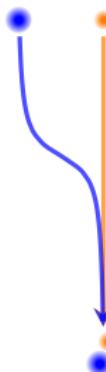
Y. Yu et al., PRX. 9, 021039 (2019)
Y. Yu et al., PRA. 97, 063423 (2018)

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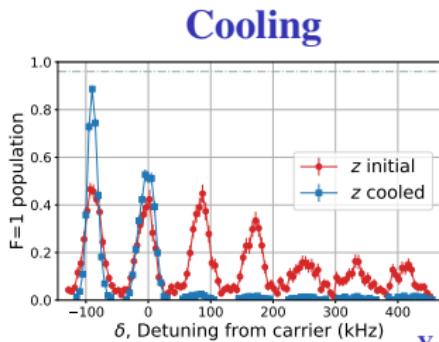


N. R. Hutzler, L. R. Liu, Y. Yu et al.,
New J. Phys. 19, 023007 (2017)

Merging

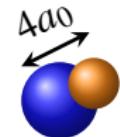


Atom

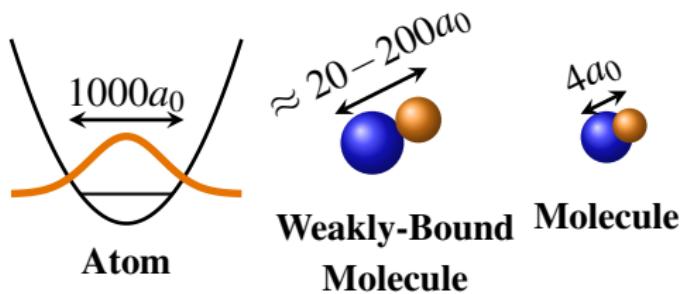


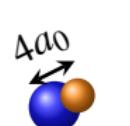
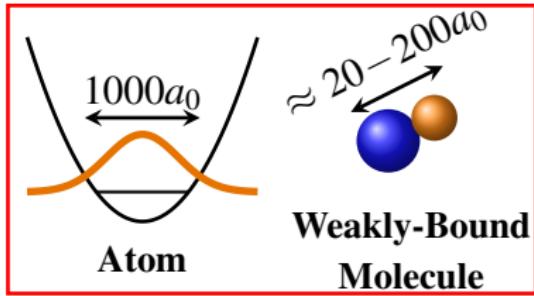
Y. Yu et al., PRA. 97, 063423 (2018)

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

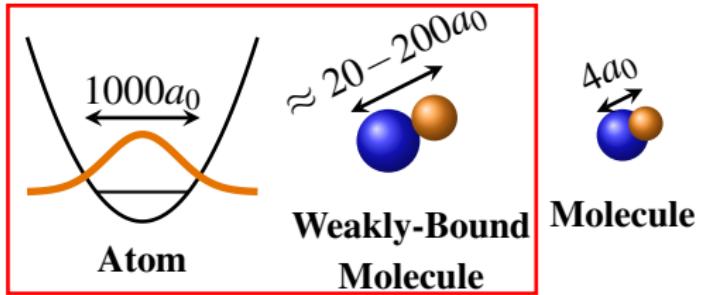


Molecule

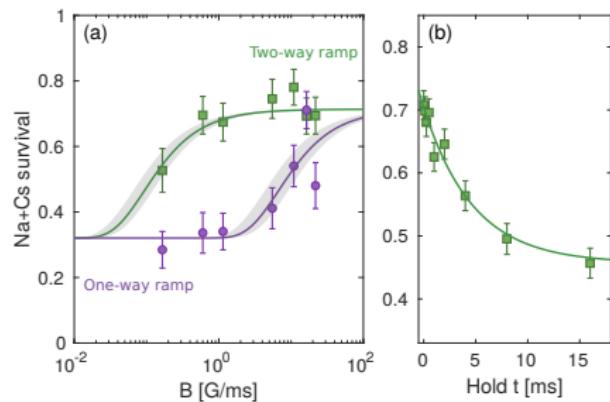




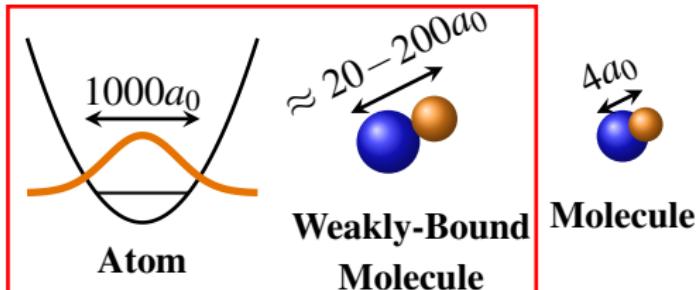
Molecule



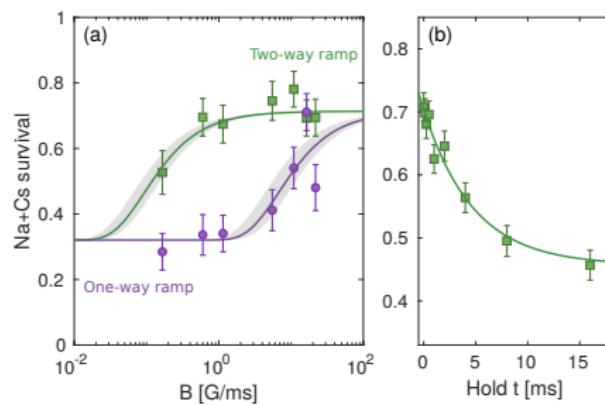
Feshbach molecule



PRL. 124, 253401 (2020)

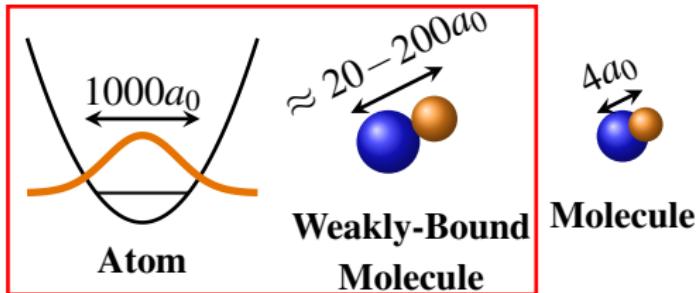


Feshbach molecule



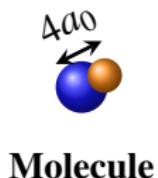
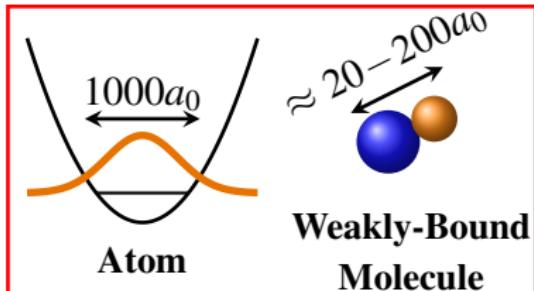
PRL. 124, 253401 (2020)

- Requires Feshbach resonance
- Usually large magnetic field



Optical transfer

- More general
- Faster

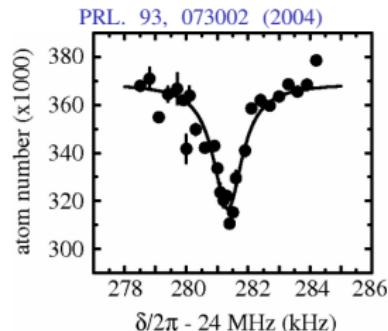


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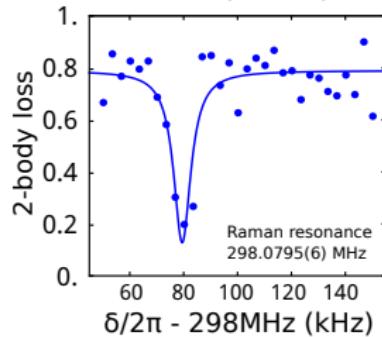
Previous results

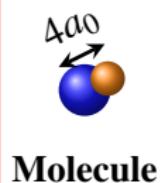
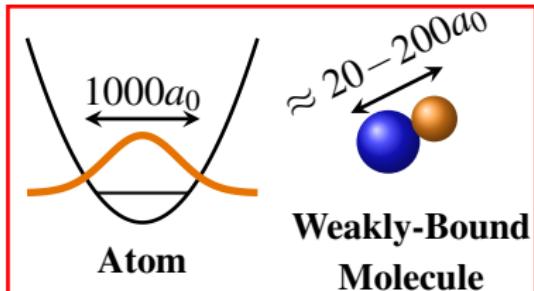
Rb_2 Science 287, 1016 (2000)



Sr_2 PRL. 109, 115302 (2012)

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





Optical transfer

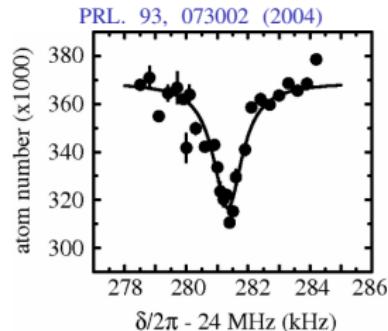
- More general
- Faster

Limitations so far

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

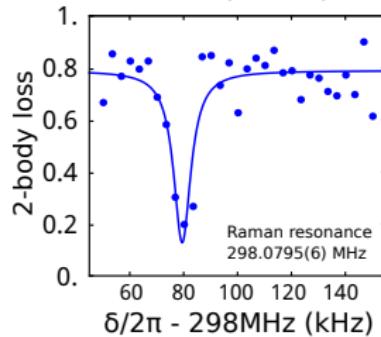
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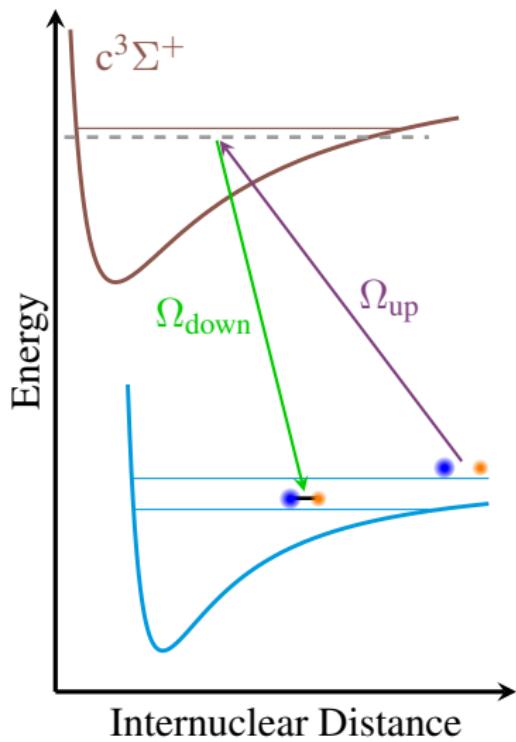


Sr_2 PRL. 109, 115302 (2012)

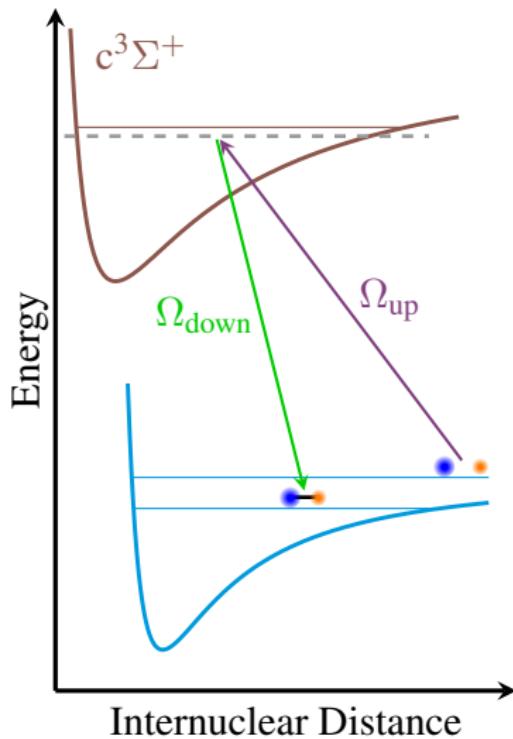
NaCs Y. Yu et al., PRX. 9, 021039 (2019)



Raman transfer



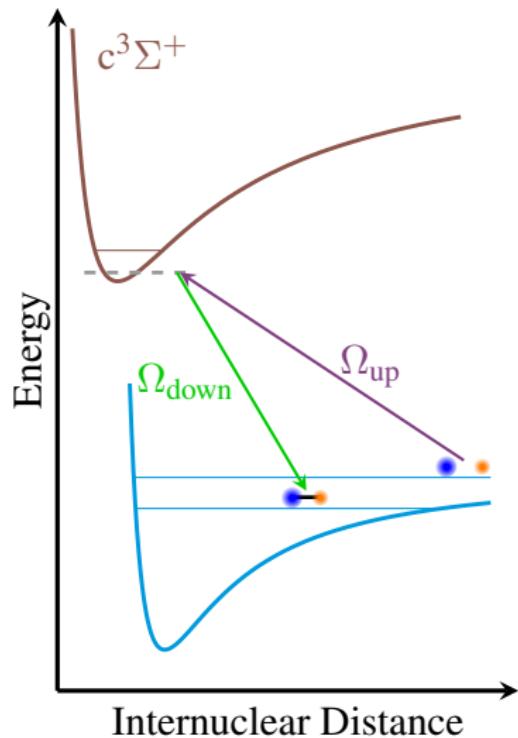
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

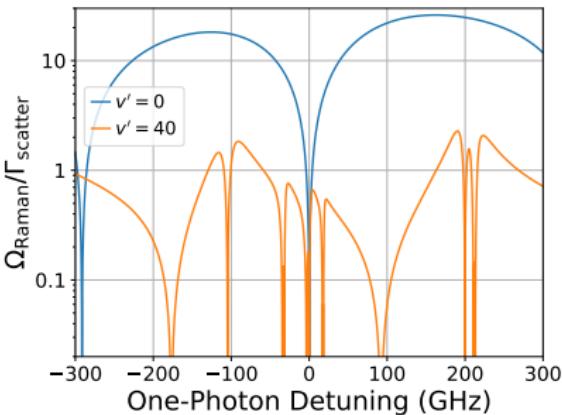
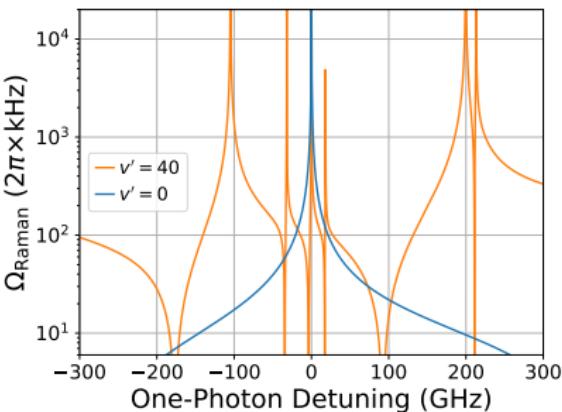
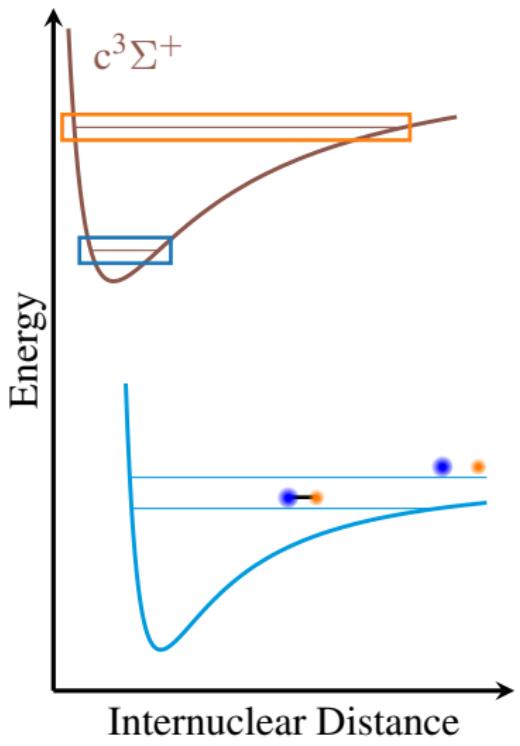
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- Closely spaced
- Fast scattering

Deeply bound states

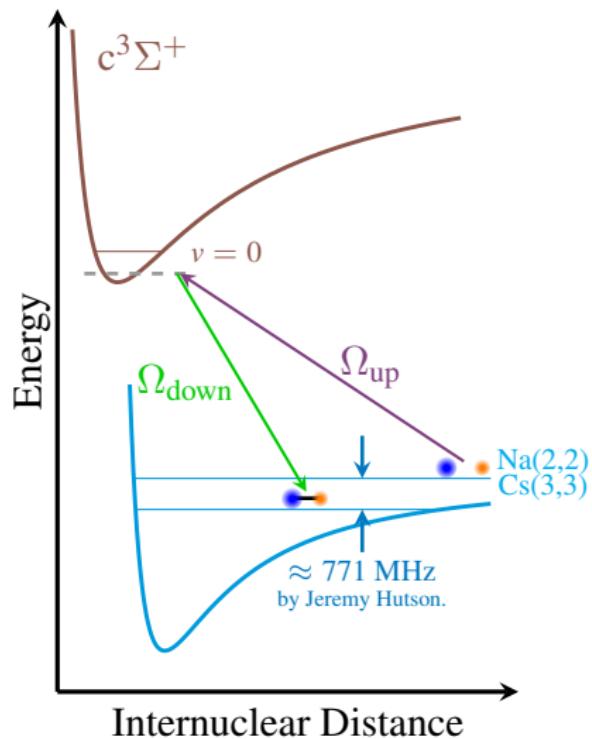
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121 (2017)

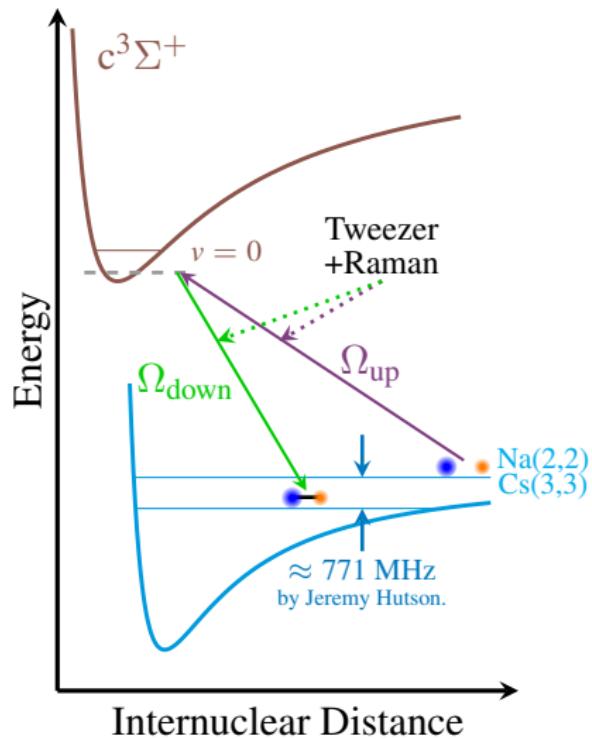
Raman transfer



Experiment



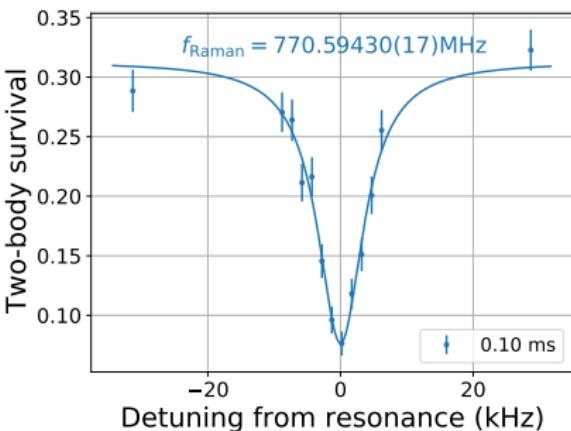
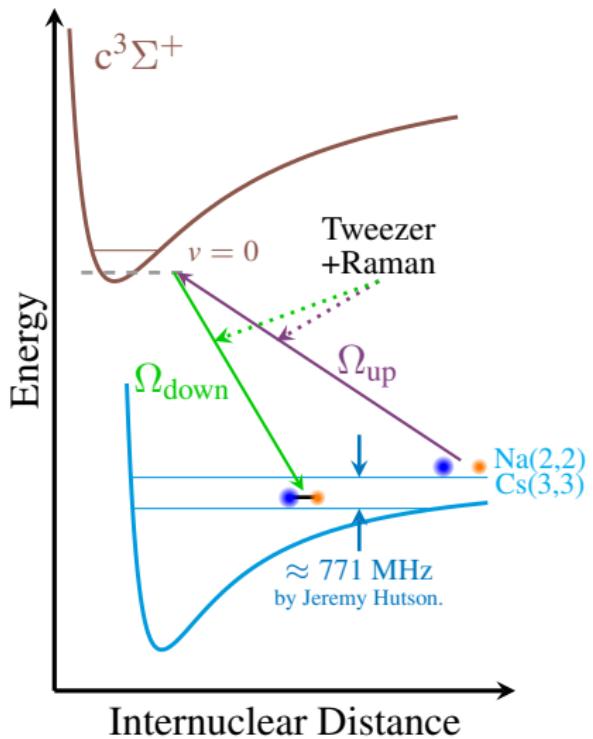
Experiment



Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

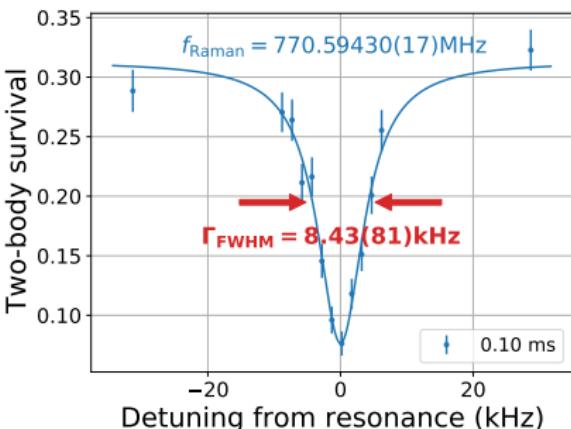
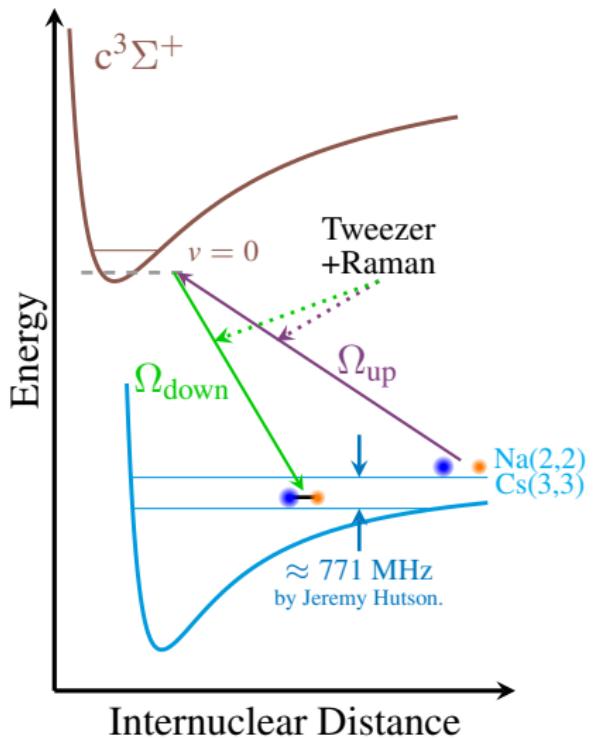
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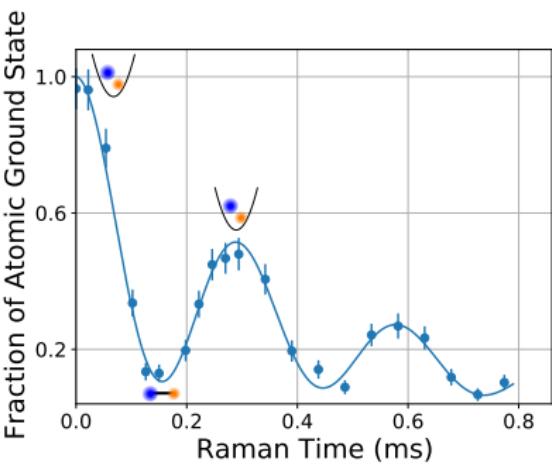
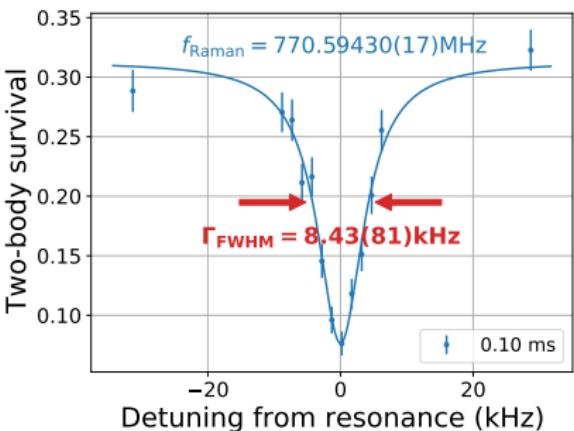
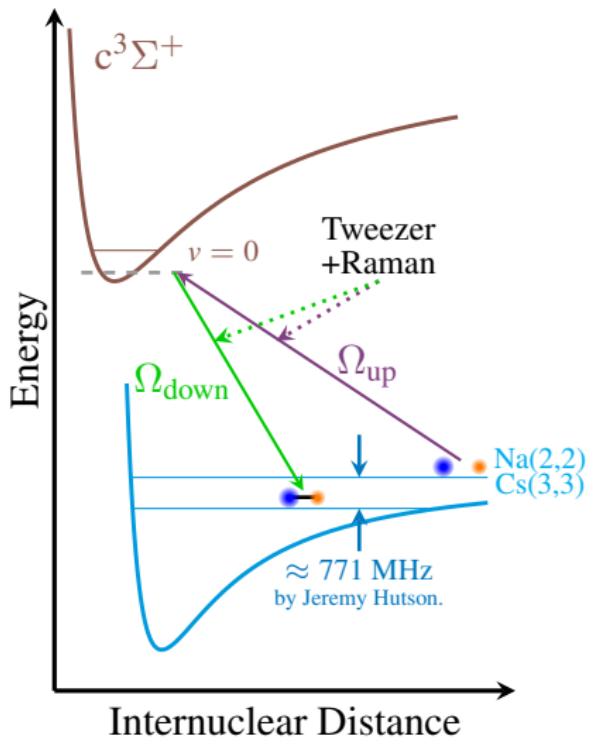
Experiment



Tweezer as Raman beam

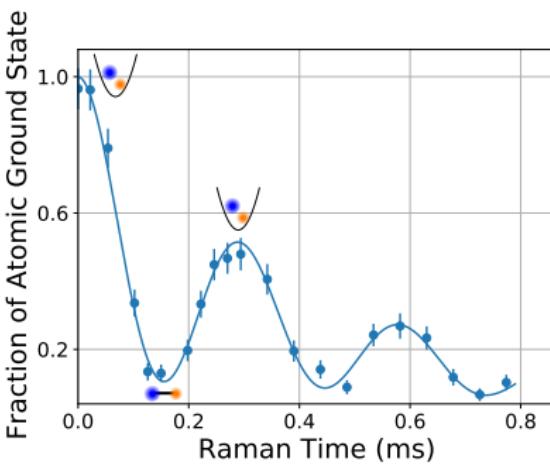
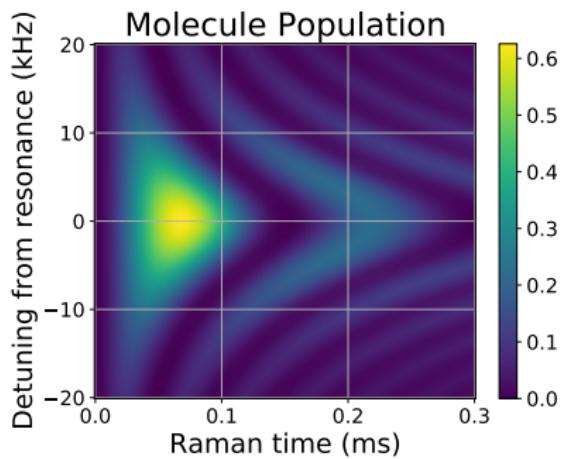
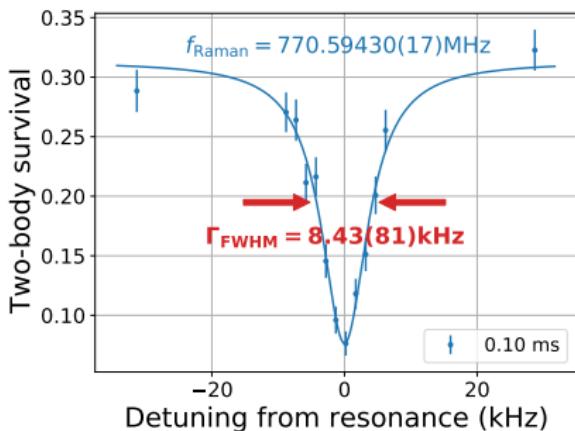
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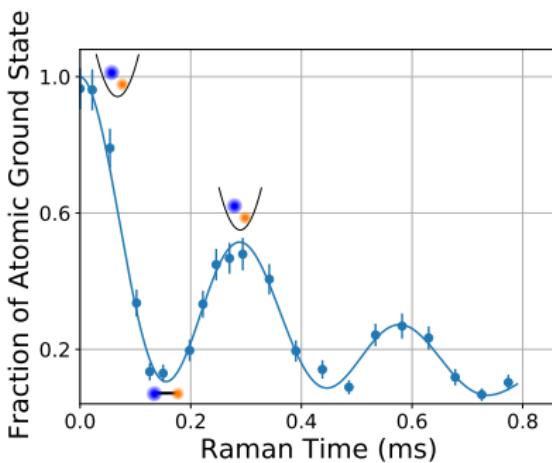
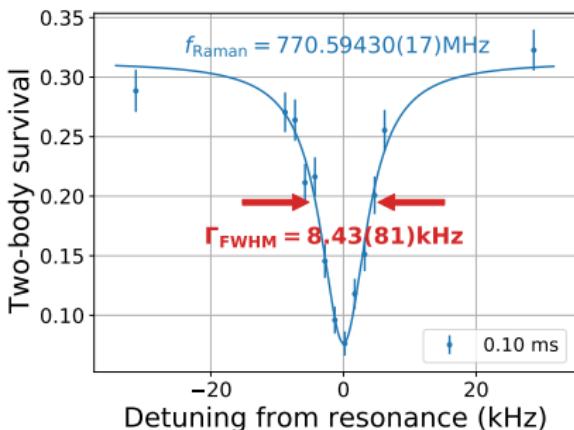
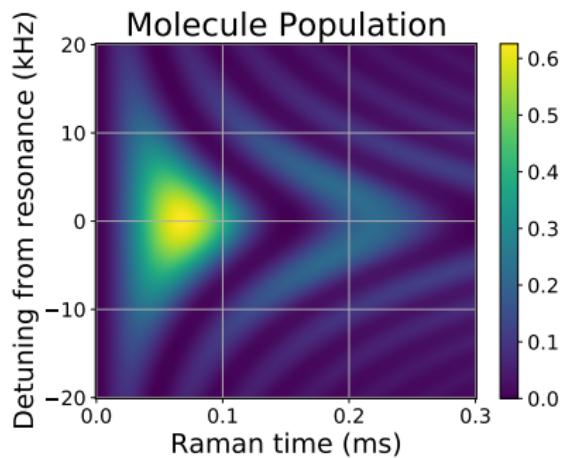
Experiment

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



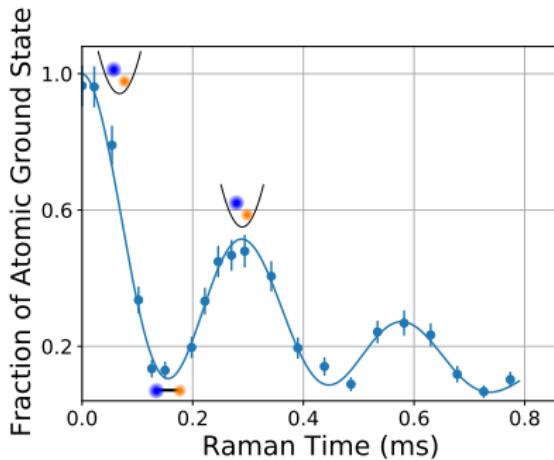
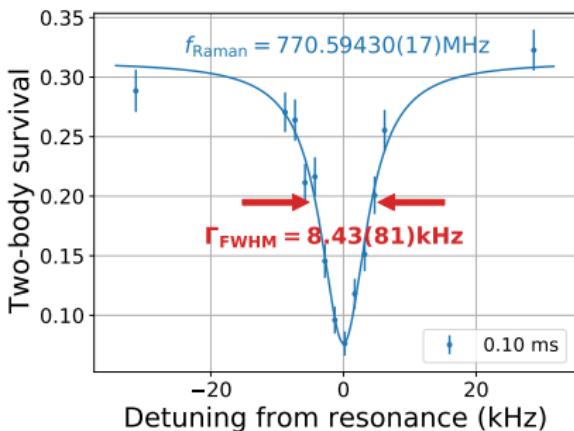
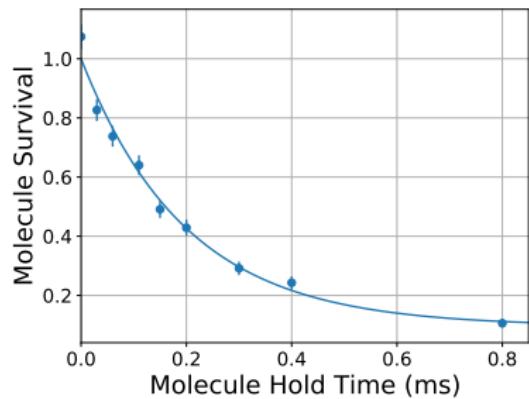
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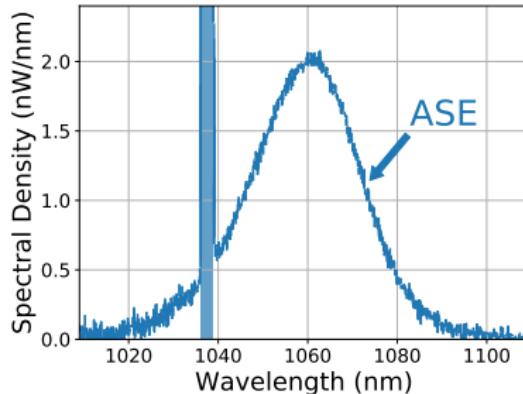


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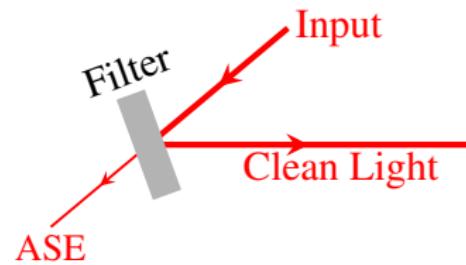
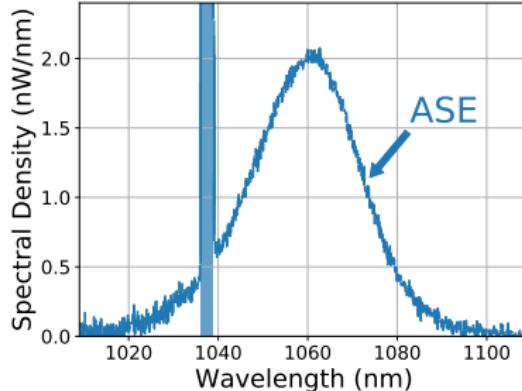
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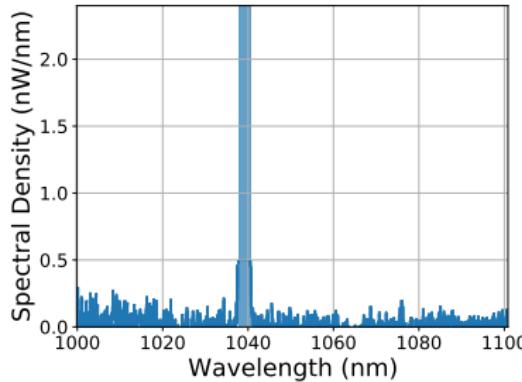
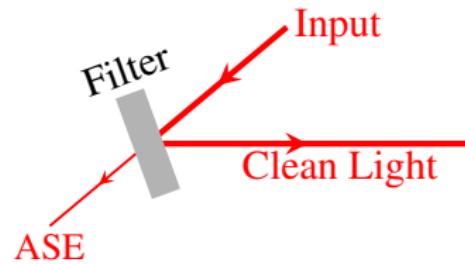
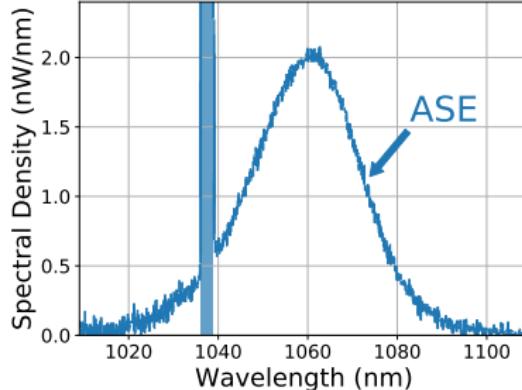
Amplified Spontaneous Emission (ASE)



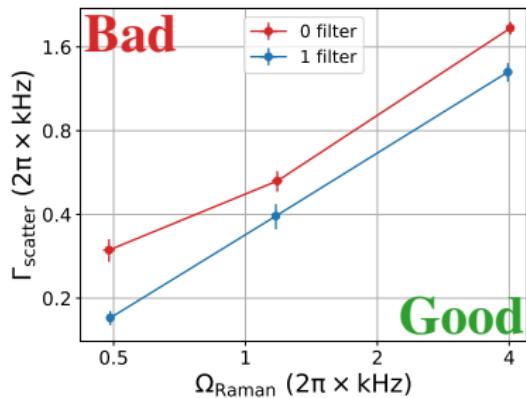
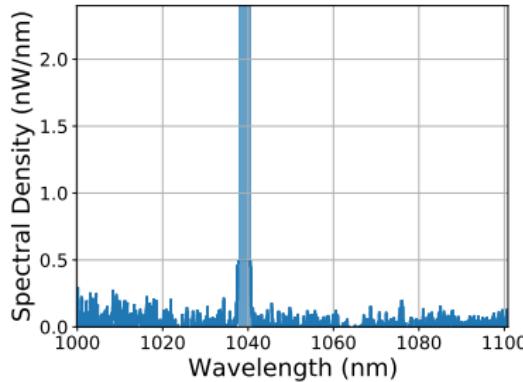
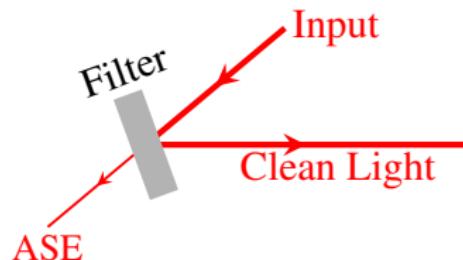
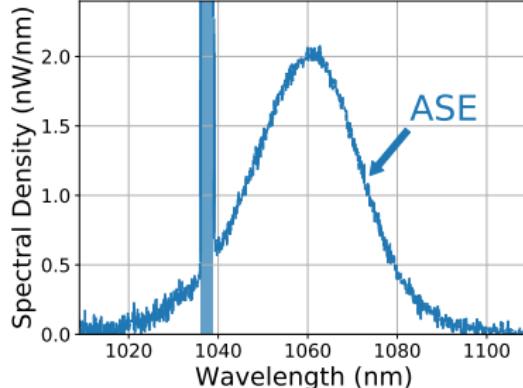
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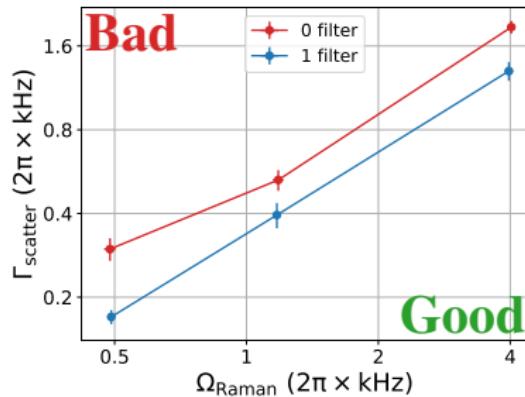
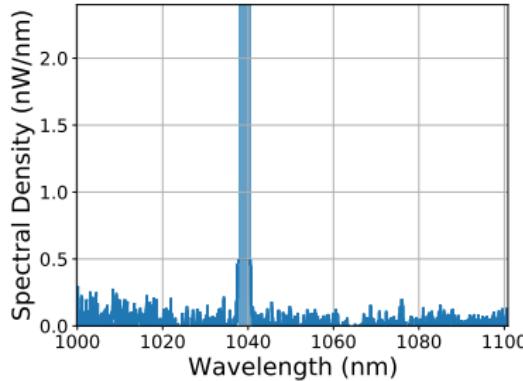
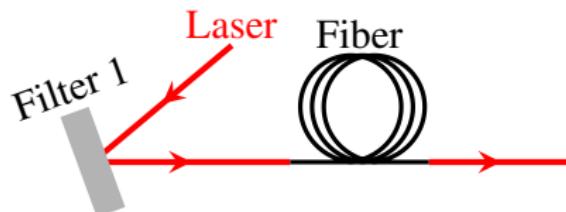
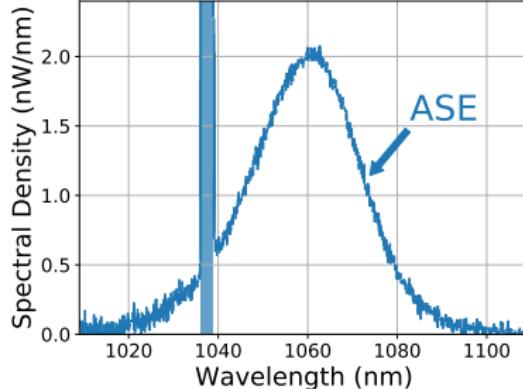
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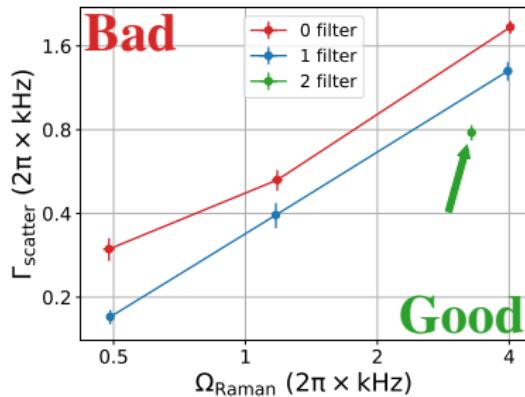
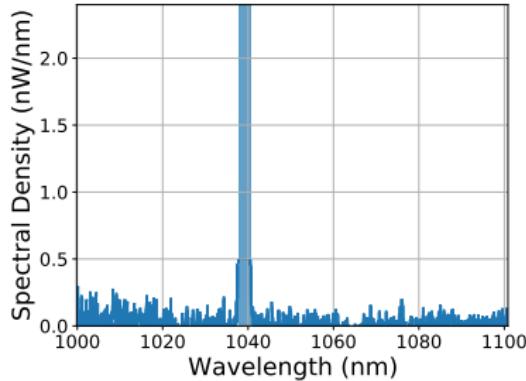
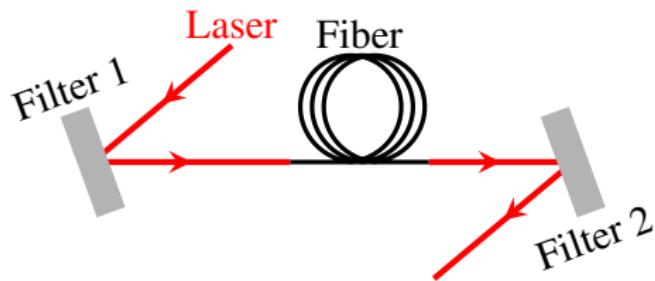
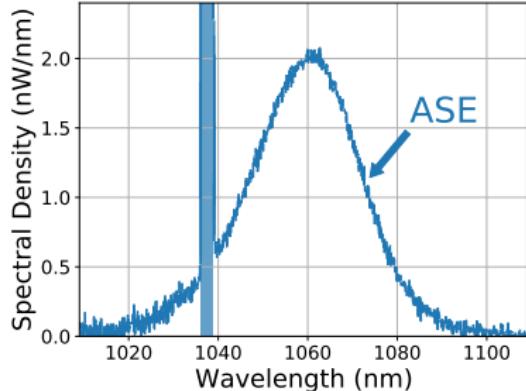
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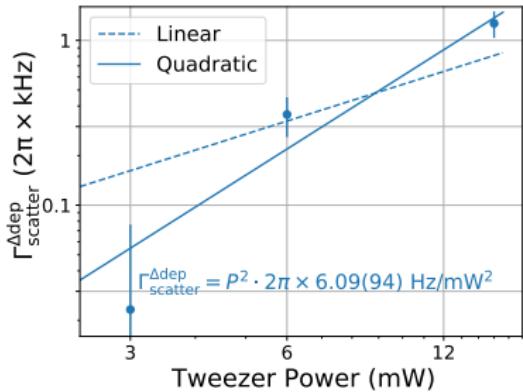


Amplified Spontaneous Emission (ASE)



Two-Photon Scattering

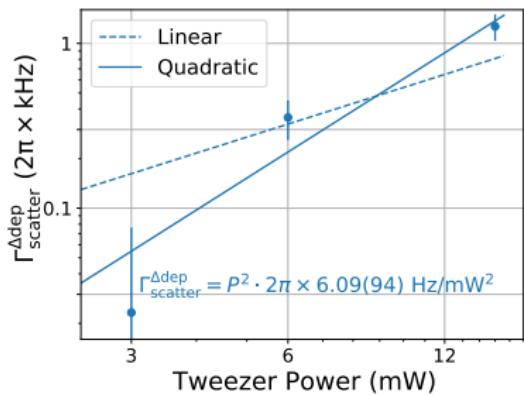
Detuning Dependent Scattering



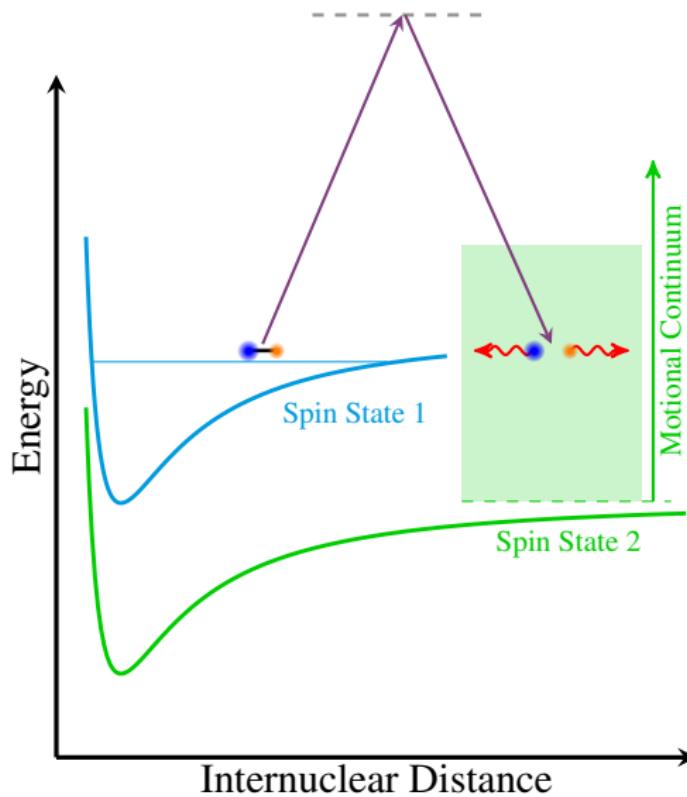
arXiv:2012.09043 (2020)

Two-Photon Scattering

Detuning Dependent Scattering



arXiv:2012.09043 (2020)



Conclusion and outlook

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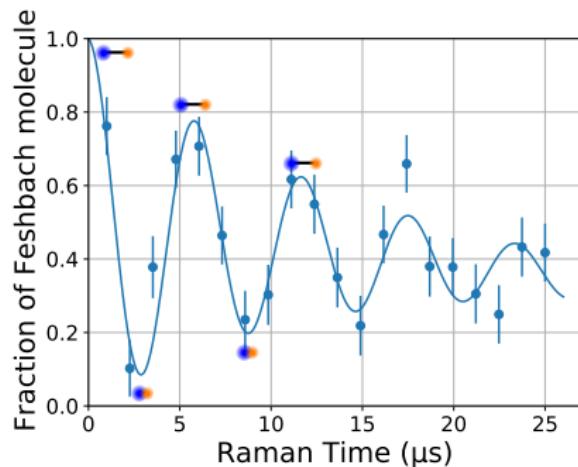
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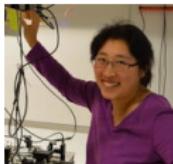
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PI



Kang-Kuen Ni

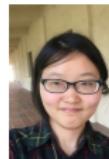
NaCs
Team



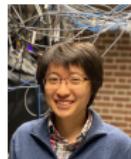
Kenneth
Wang



Yu
Wang



Fang
Fang



Jessie
Zhang



Lewis
Picard



William
Cairncross

KRb
Team



Lingbang
Zhu



Mingguang
Hu



Matthew
Nichols



Lee Liu
Postdoc @JILA



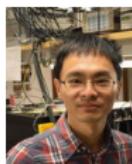
Nick Hutzler
AP @Caltech



Jonathan Hood
AP @Purdue



Eliot
Fenton



Yen-Wei Lin
Intelon Optics



Yu Liu
Postdoc @NIST

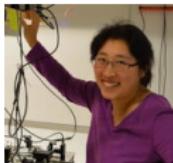


Andrei
Gheorghe



David Grimes
Instructor @MIT

PI



Kang-Kuen Ni



NaCs
Team



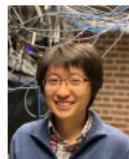
Kenneth
Wang



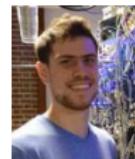
Yu
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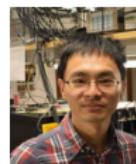
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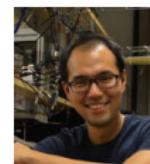
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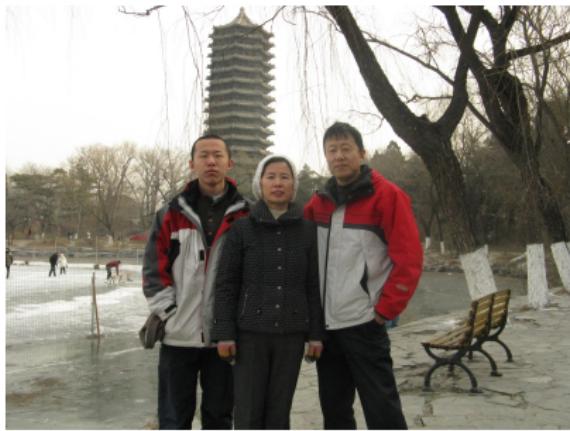
Yu Liu
Postdoc @NIST



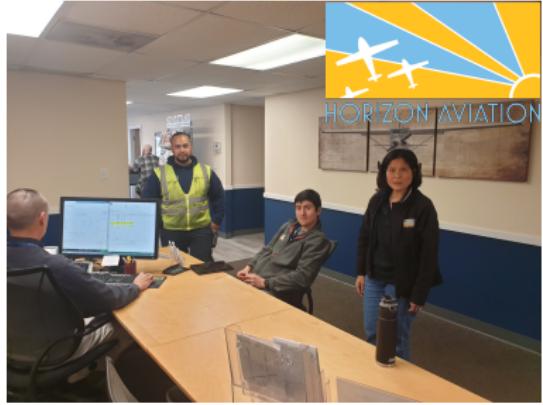
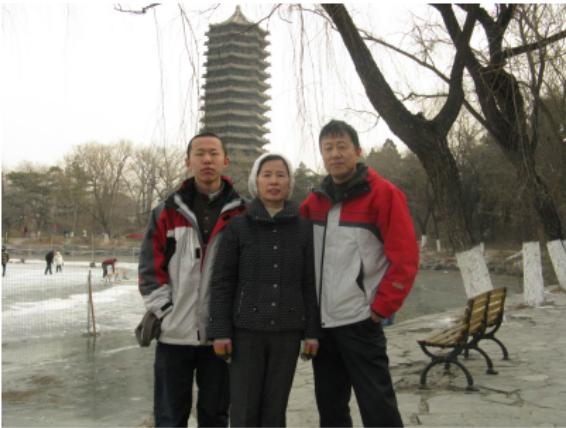
Andrei
Gheorghe



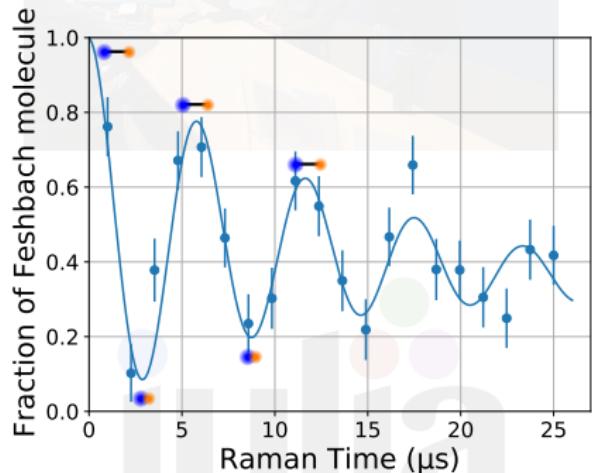
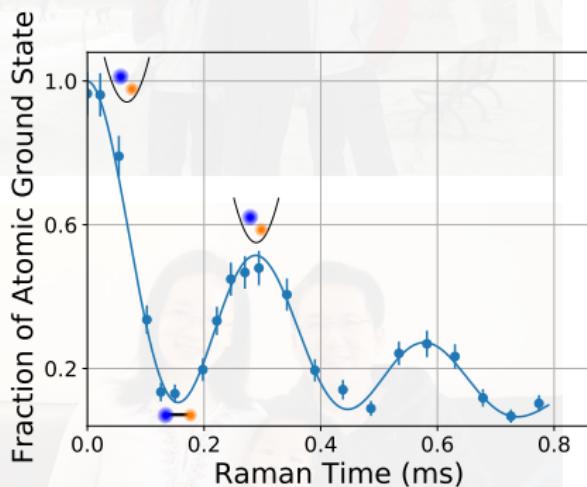
David Grimes
Instructor @MIT







Thanks for your attention

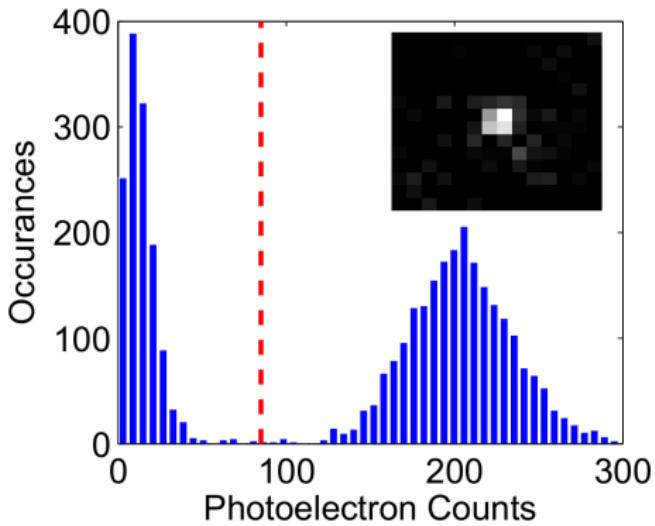


Single Atom in Tweezer

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

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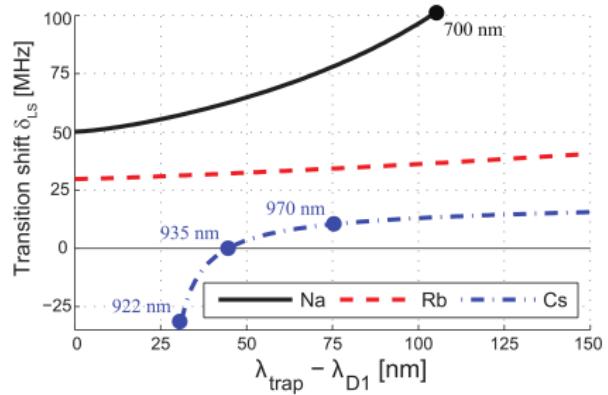
Single Atom in Tweezer

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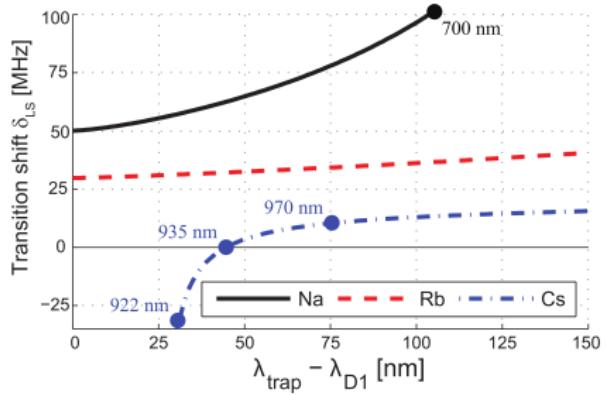
Issues with Na

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

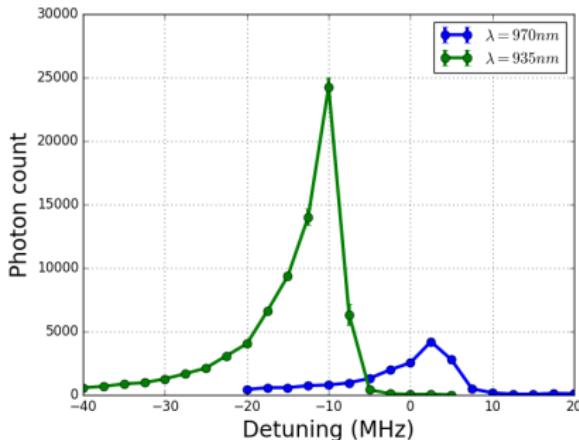
Real Issue with Na: Light Shift



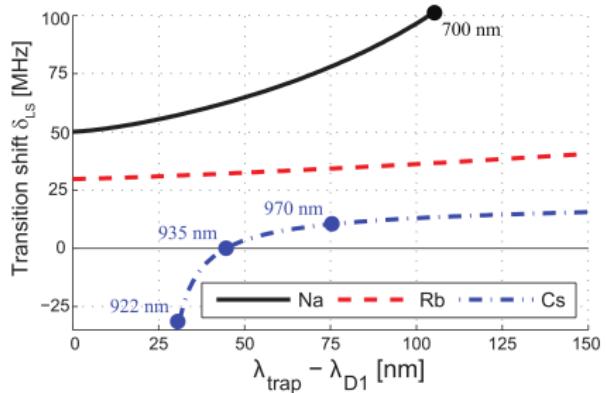
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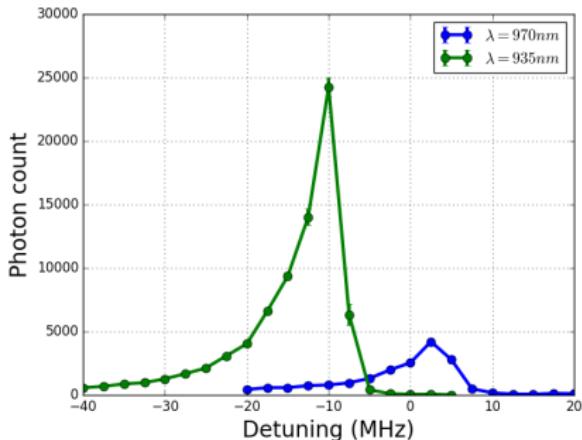
Cs single atom imaging



Real Issue with Na: Light Shift



Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

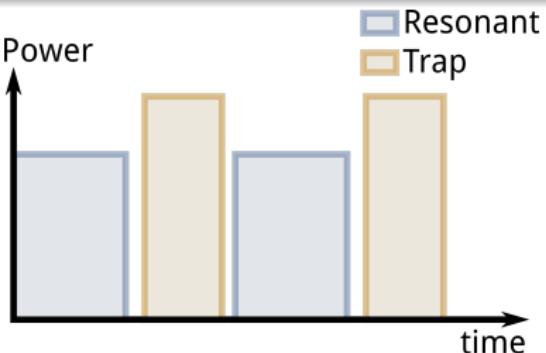
Real Issue with Na: Light Shift

Trap modulation

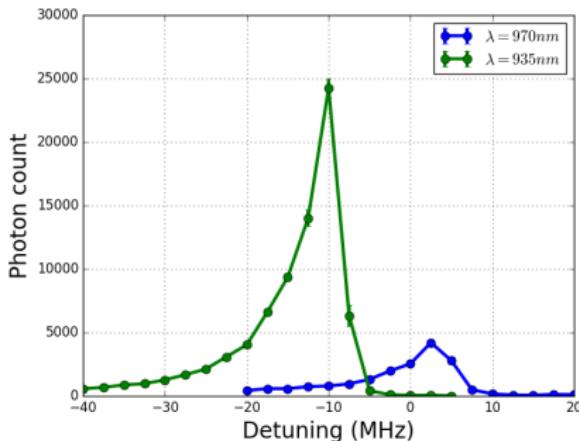
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



Cs single atom imaging



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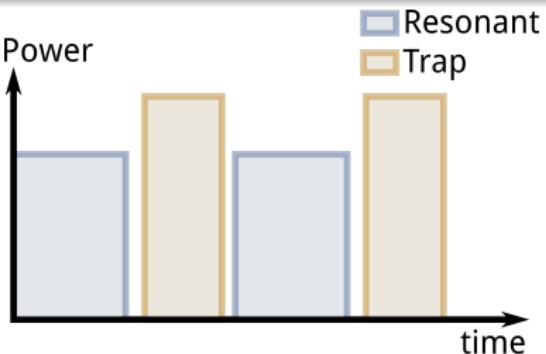
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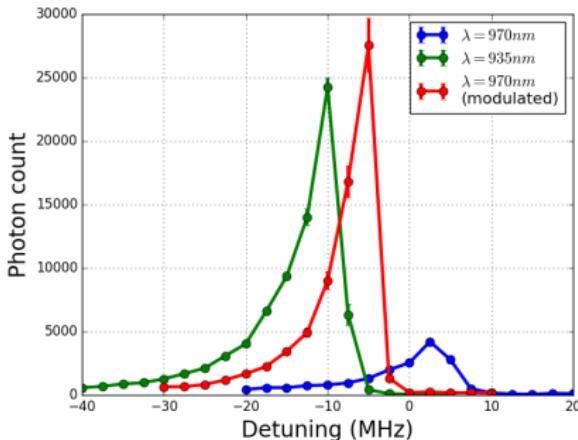
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Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

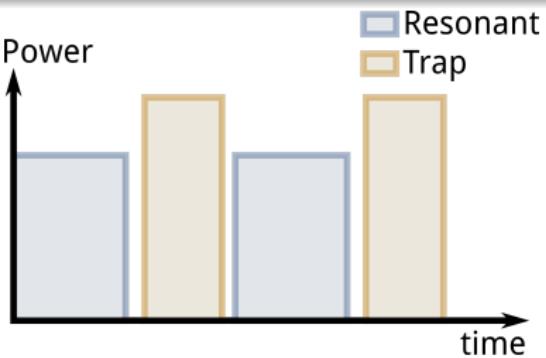
Real Issue with Na: Light Shift

Trap modulation

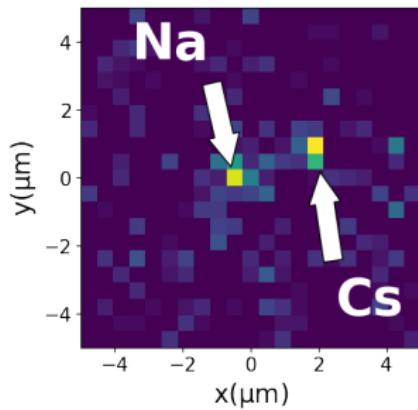
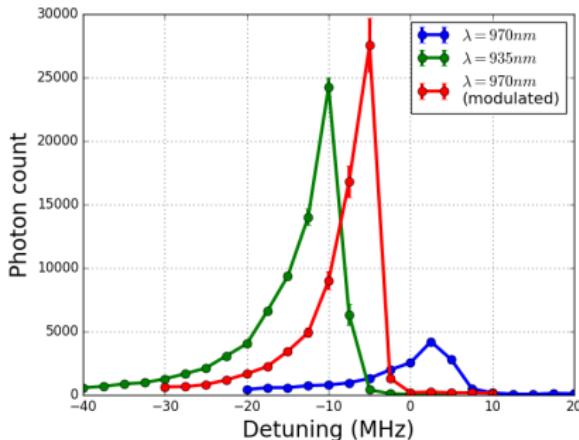
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

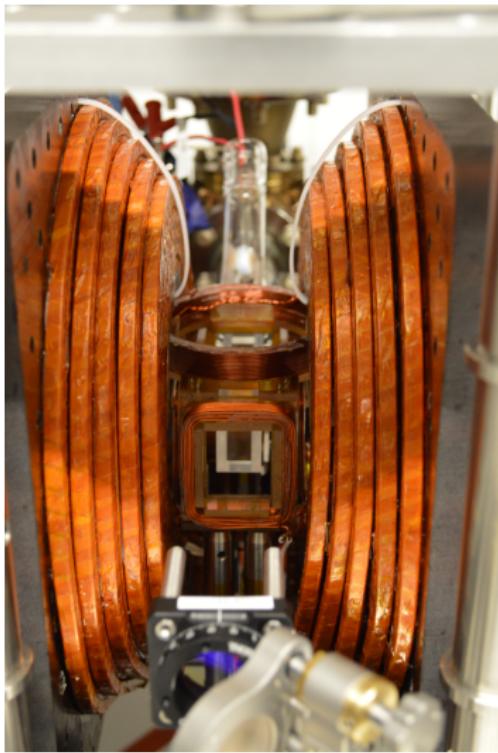
$$\Gamma = 2\pi \times 10 \text{ MHz}$$



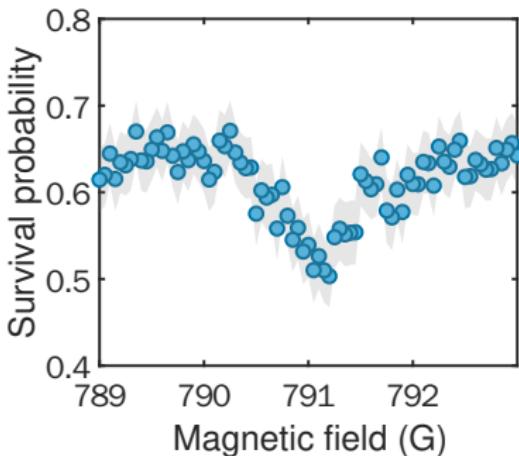
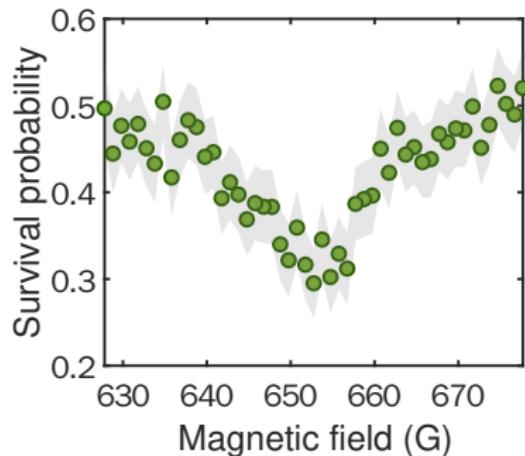
Cs single atom imaging



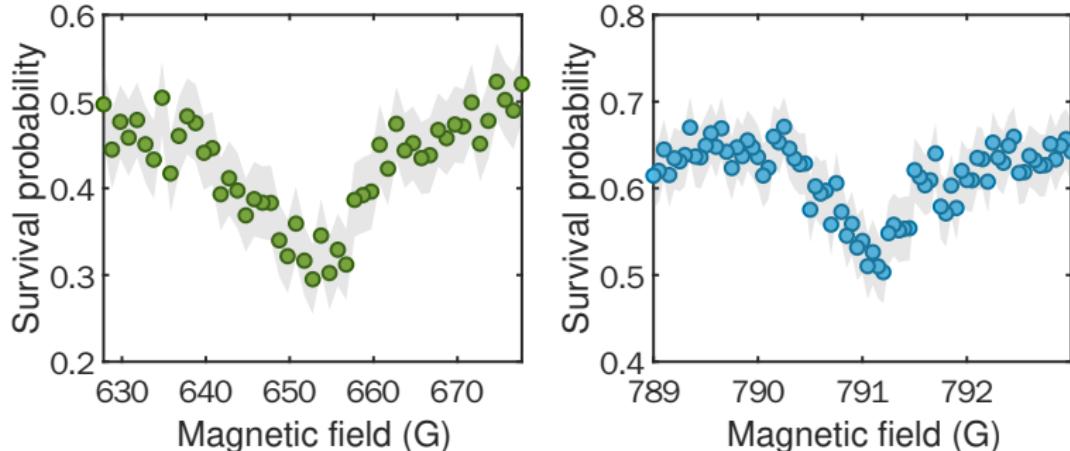
Na (1, -1) Cs (3, -3) Feshbach resonance



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



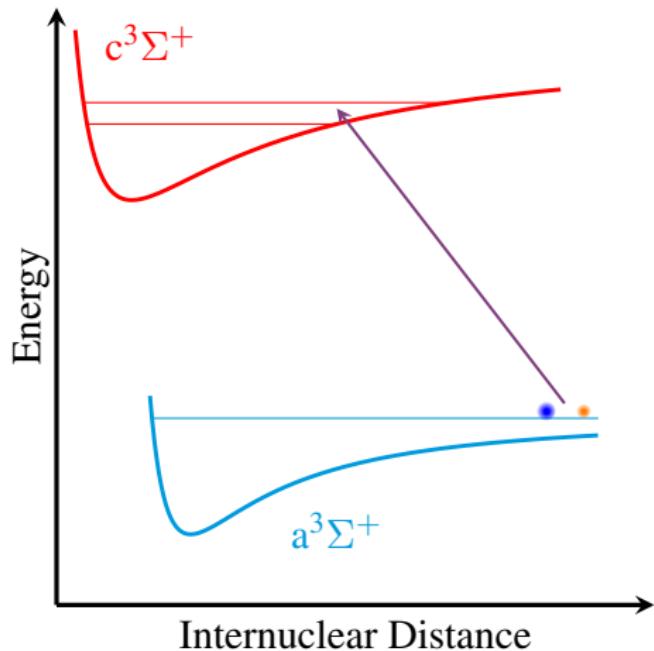
Na (1, -1) Cs (3, -3) Feshbach resonance



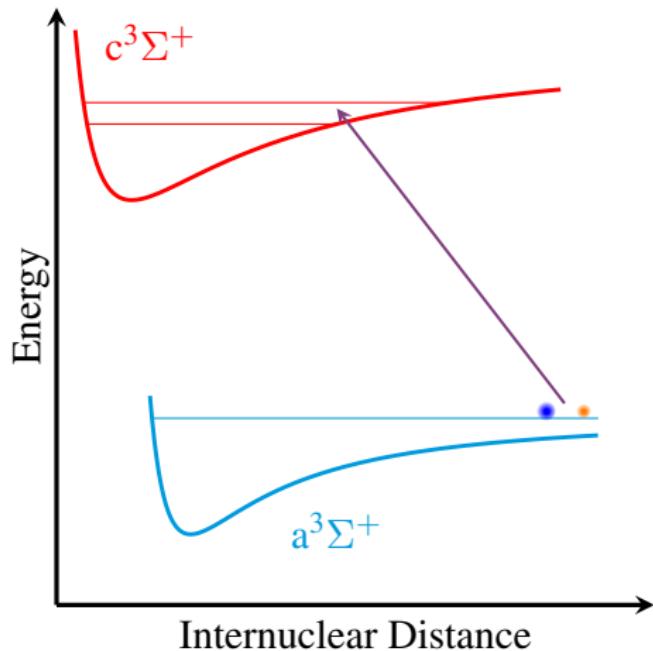
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



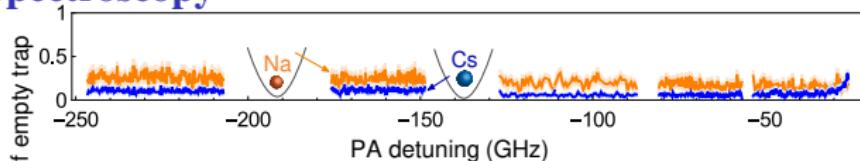
Photoassociation (PA) Spectroscopy



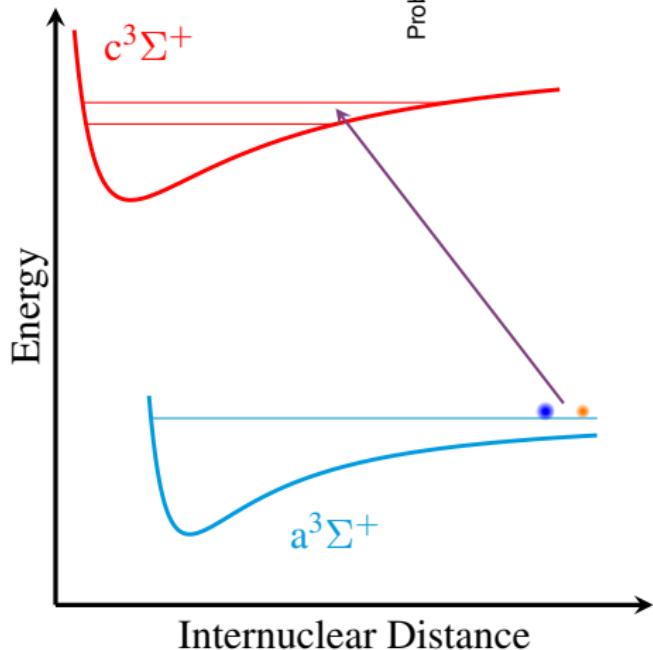
Single Atom PA

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

Photoassociation (PA) Spectroscopy



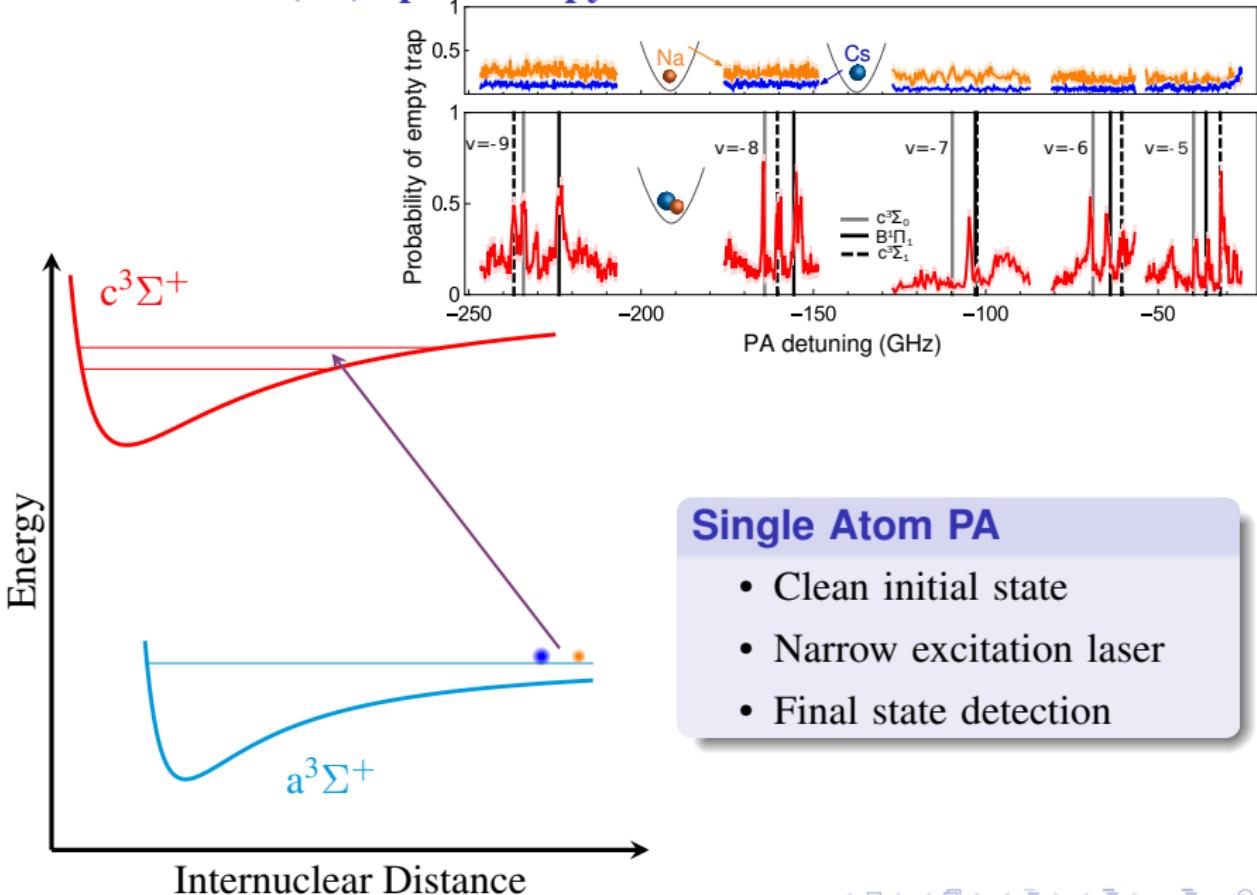
Probability of empty trap



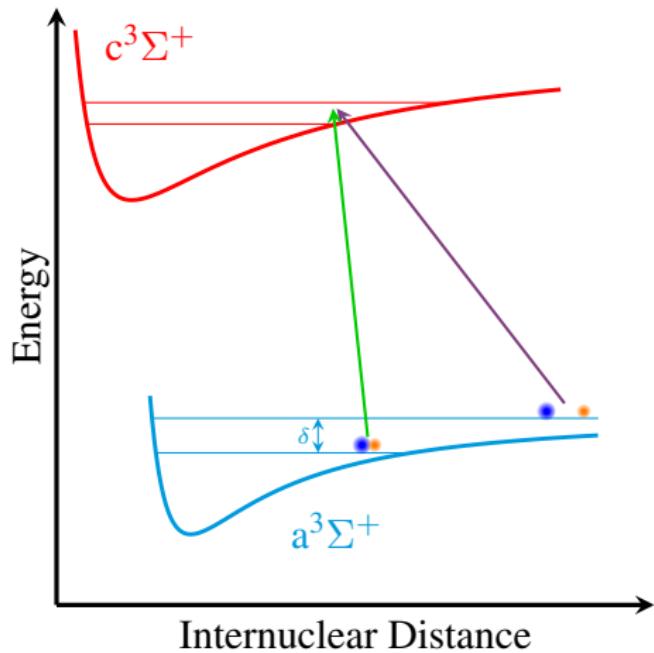
Single Atom PA

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

Photoassociation (PA) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy

