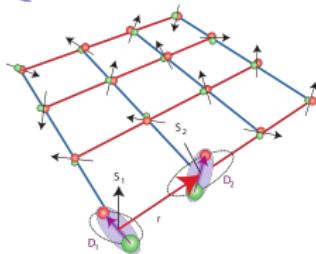


Coherent Association of Single Molecules from Single Atoms

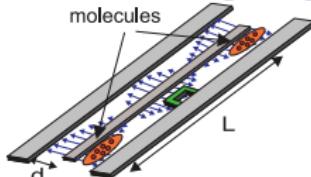
Yichao Yu

Ni Group/Harvard

Quantum Simulation



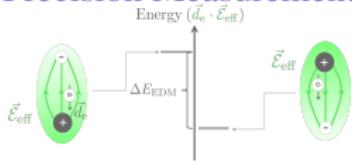
Quantum Computing



PRL. 97, 33003 (2006)

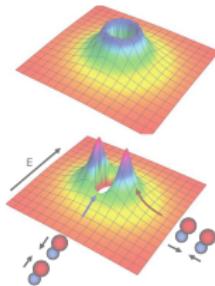
Nat. Phys. 2, 341 (2006)

Precision Measurement



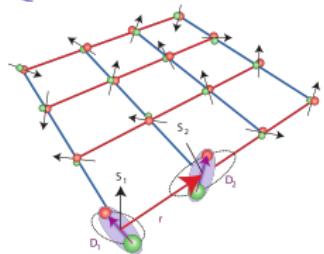
Science 343, 269 (2014)

Quantum Chemistry



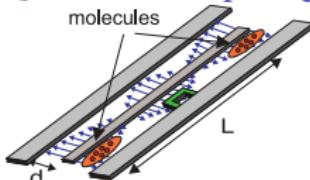
Nature 464, 1324 (2010)

Quantum Simulation



Nat. Phys. 2, 341 (2006)

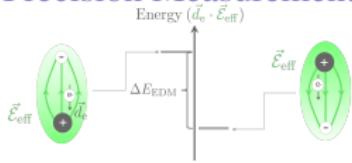
Quantum Computing



PRL. 97, 33003 (2006)

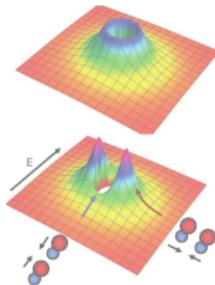
- Full quantum control
- Entanglement
- ...

Precision Measurement



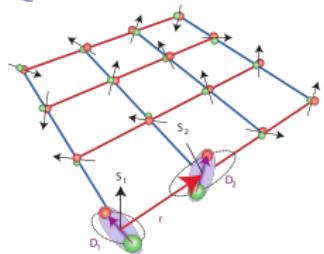
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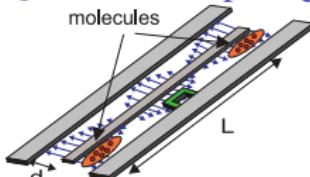
Nature 464, 1324 (2010)

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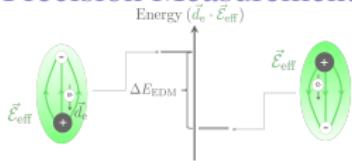
Quantum Computing



PRL. 97, 33003 (2006)

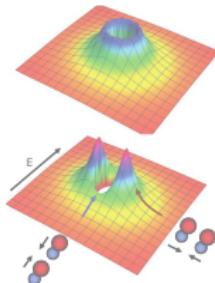
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Precision Measurement



Science 343, 269 (2014)

Quantum Chemistry



Nature 464, 1324 (2010)

New Approach?

Entanglement

Single particle control

Entanglement

i.e. interaction

Single particle control

Entanglement

i.e. interaction

Single particle control

Dipolar molecules

Dipolar molecules

- Strong and tunable interaction
($\approx k\text{Hz}$ at $\approx \mu\text{m}$ distance)
 - ▶ Fast gate operations
 - ▶ Long coherence time
- Rich internal structure
(Electronic, vibrational,
rotational, hyperfine, etc.)

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Single particle control

Optical tweezers

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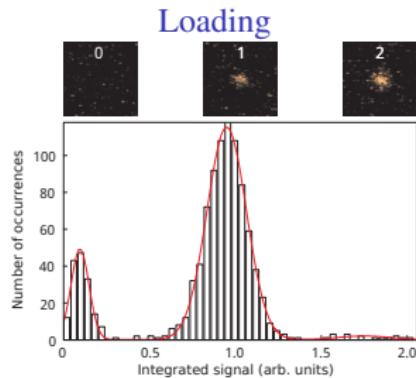
- Single site resolution



Entanglement

i.e. interaction

Dipolar molecules



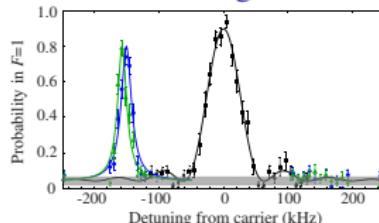
Single particle control

Optical tweezers

- Single site resolution

- ...

Cooling



PRX. 2, 041014 (2012)

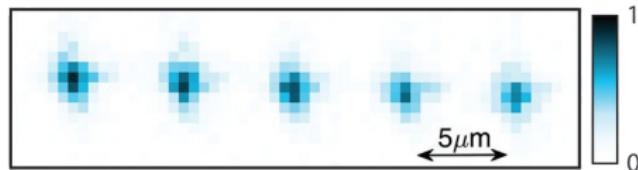
Rearranging



Science 354, 1024 (2016)

Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

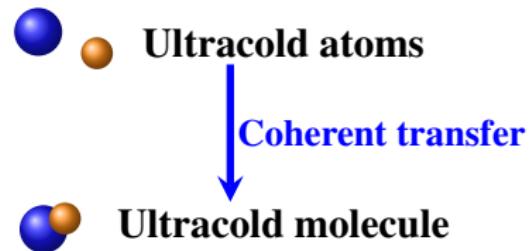
Ultracold molecule in tweezers

Direct cooling



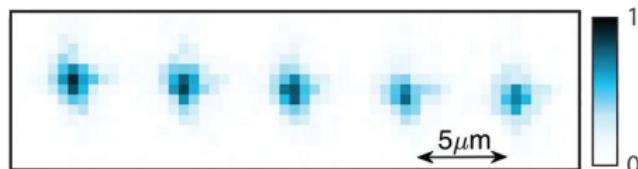
Science 365, 1156 (2019)

Assembly



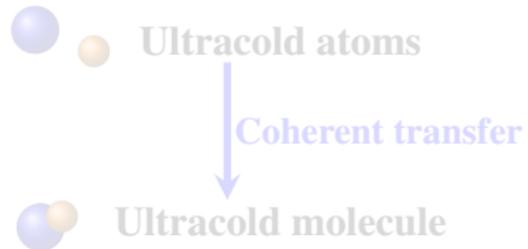
Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezer
- Quantum control

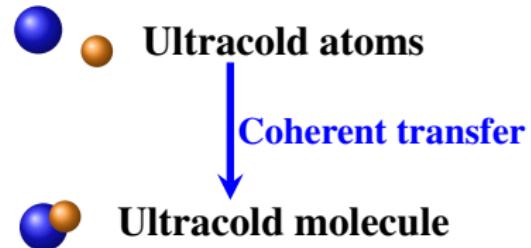
Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

Assembly



Challenges

- Temperature in tweezers
- Quantum control
- Control of atoms
- Coherent creation of molecules

Outline

1 Experiment overview

2 Atom state control

- Raman sideband cooling of Na atoms

3 Molecule creation

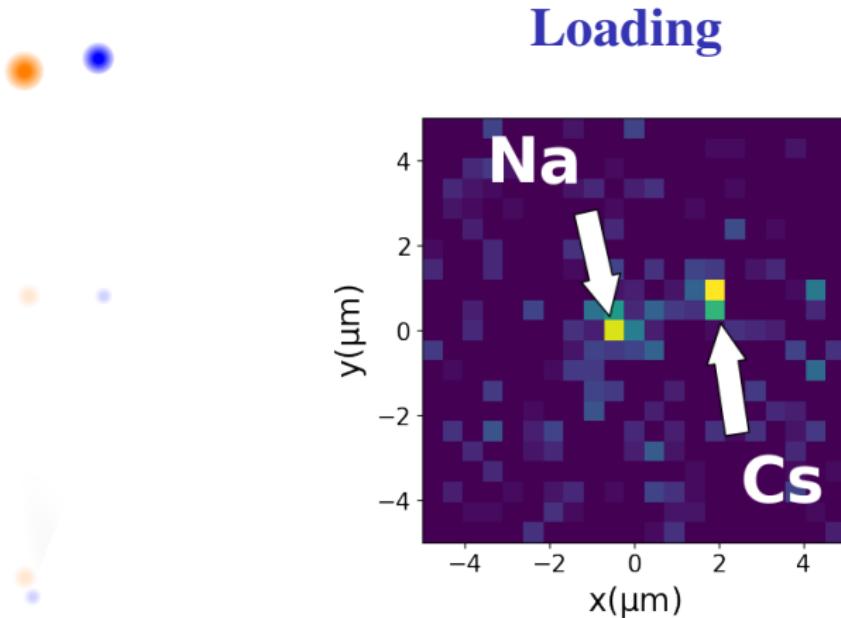
- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

NaCs molecule

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

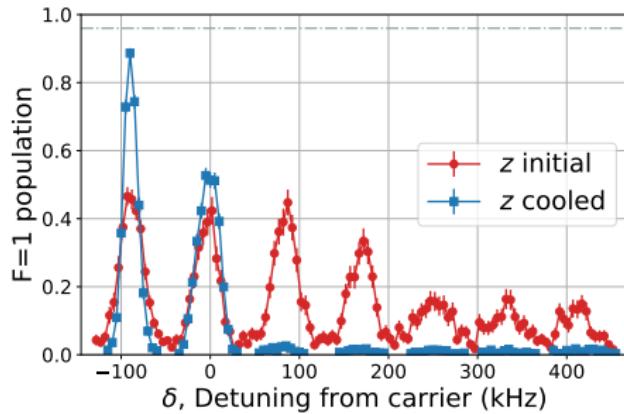
Experiment overview



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

Experiment overview

Cooling



Cs: 96% ground state¹

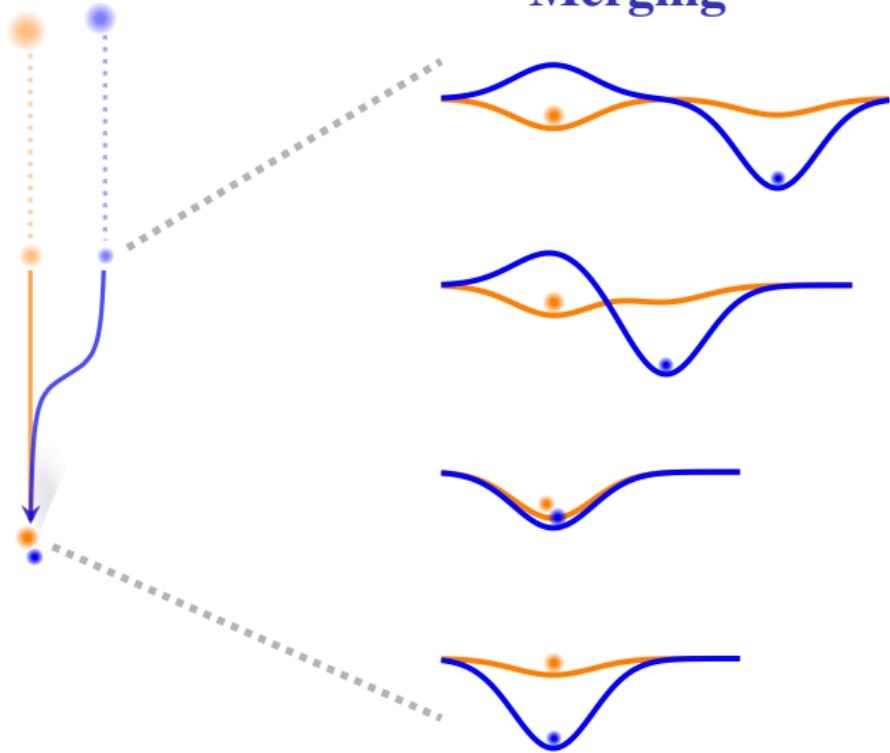
Na: 94% ground state²

¹Y. Yu et al. PRX 9, 021039 (2019)

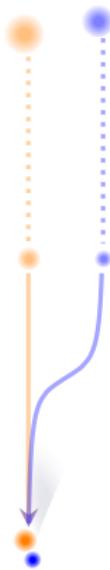
²Y. Yu et al. PRA 97, 063423 (2018)

Experiment overview

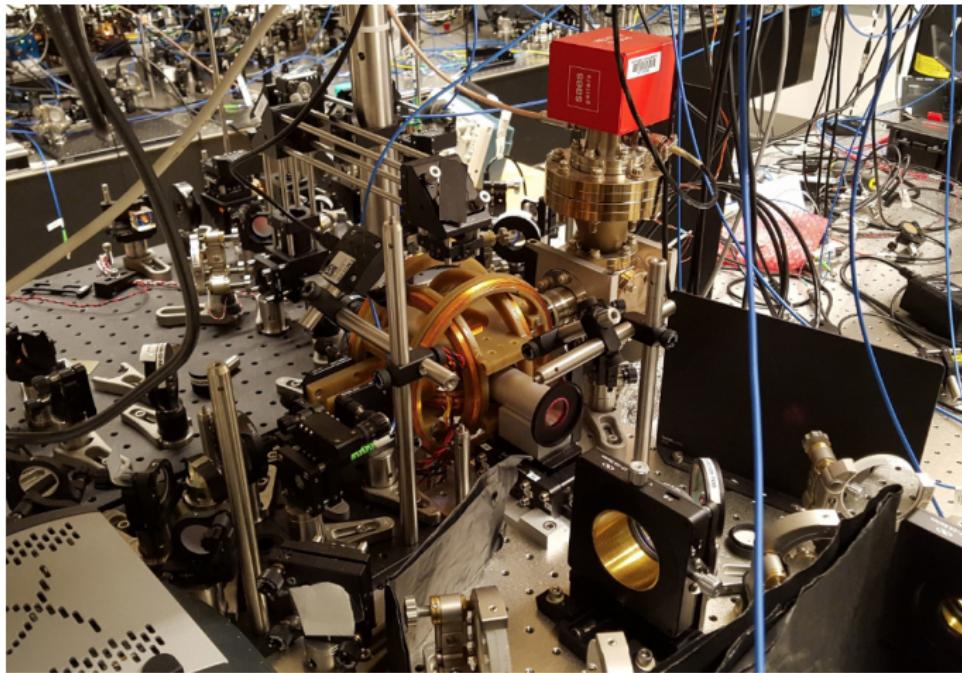
Merging

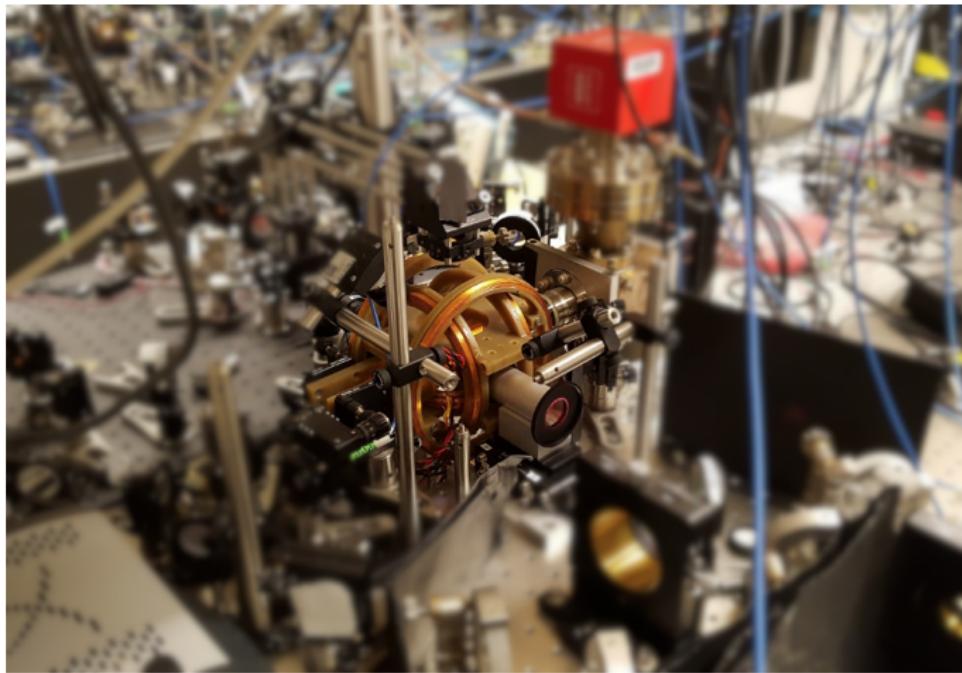


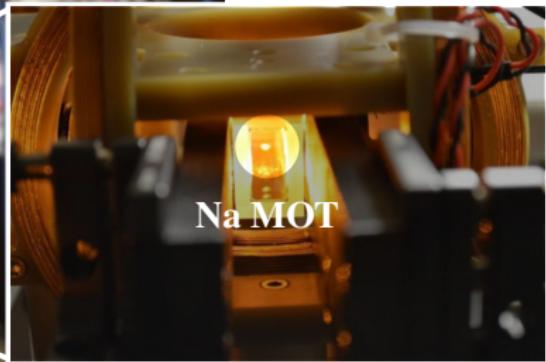
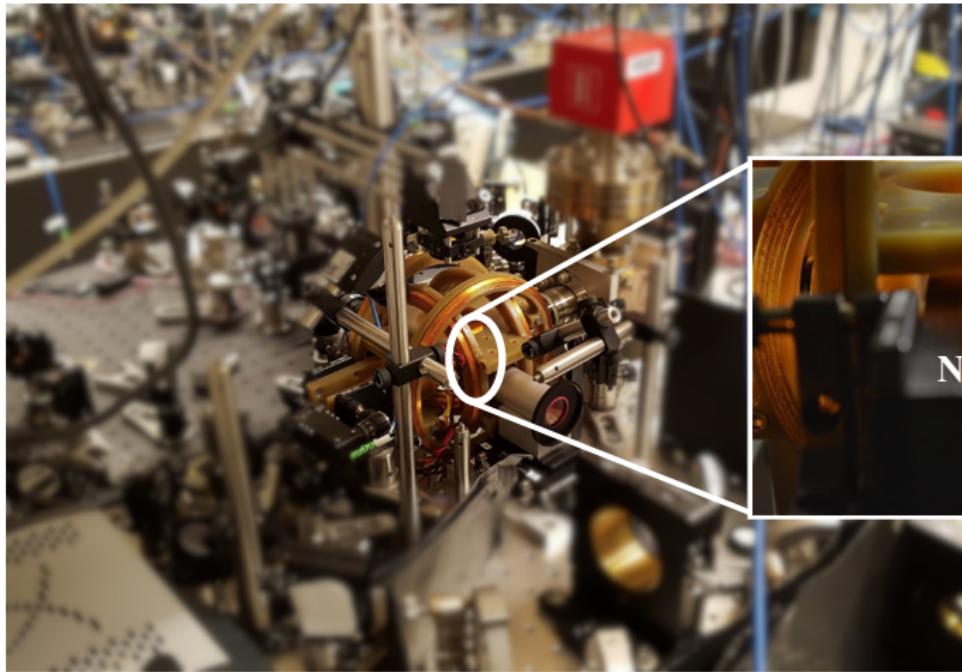
Experiment overview



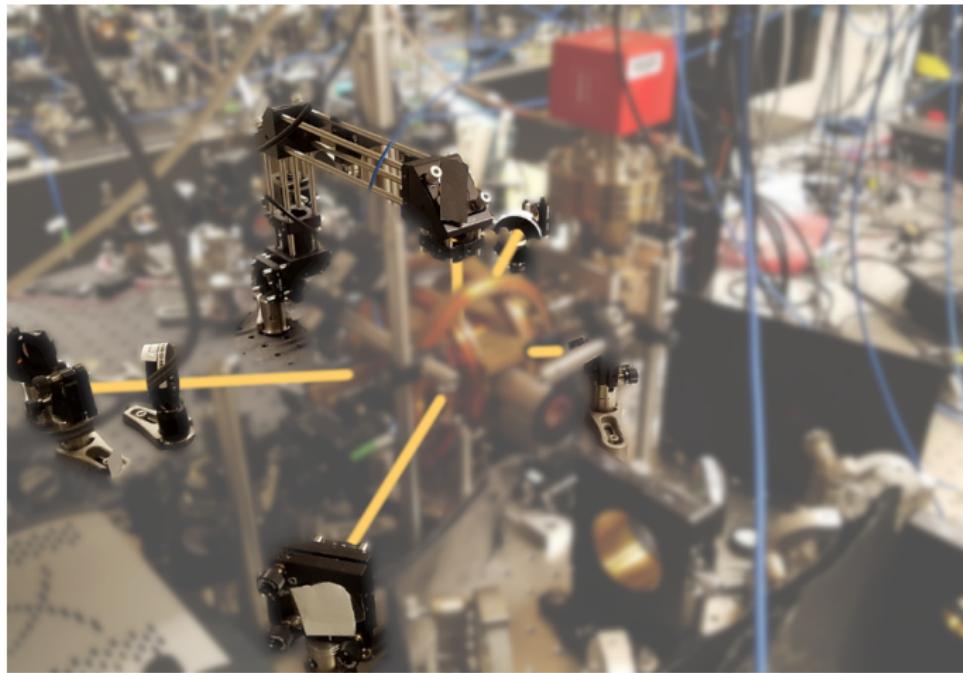
L. R. Liu, J. D. Hood, Y. Yu et al., Science 360, 6391 (2018)



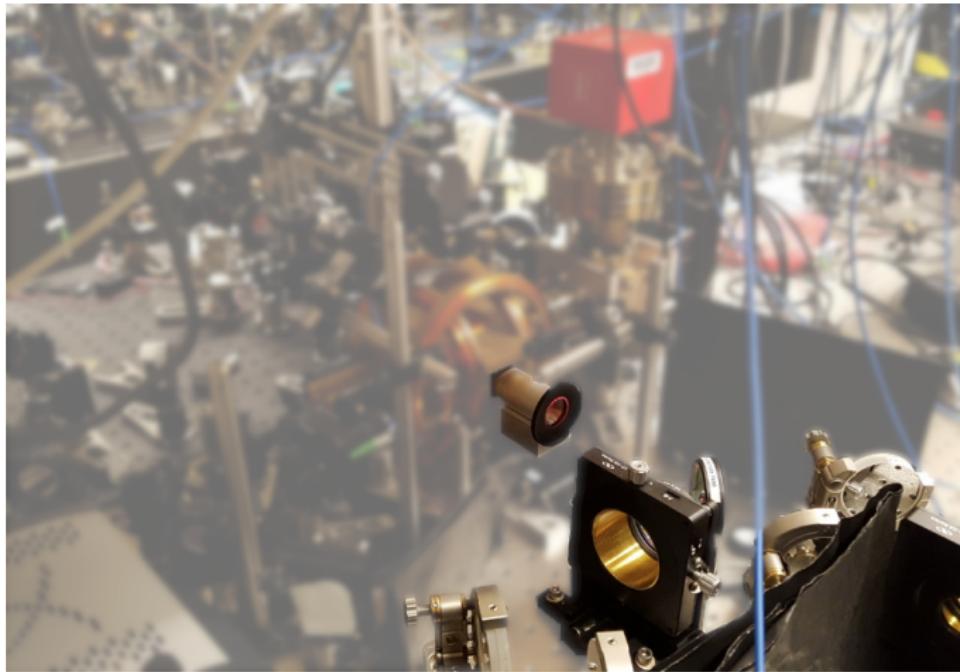




MOT beam path



Tweezer beam path



Outline

1 Experiment overview

2 Atom state control

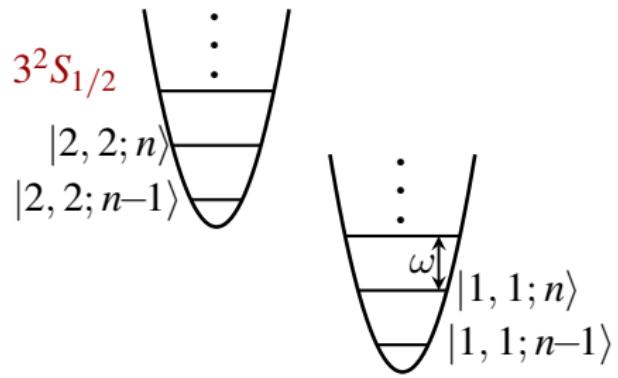
- Raman sideband cooling of Na atoms

3 Molecule creation

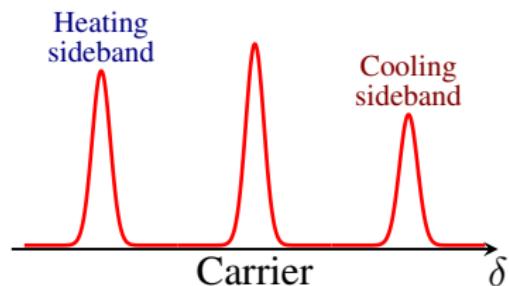
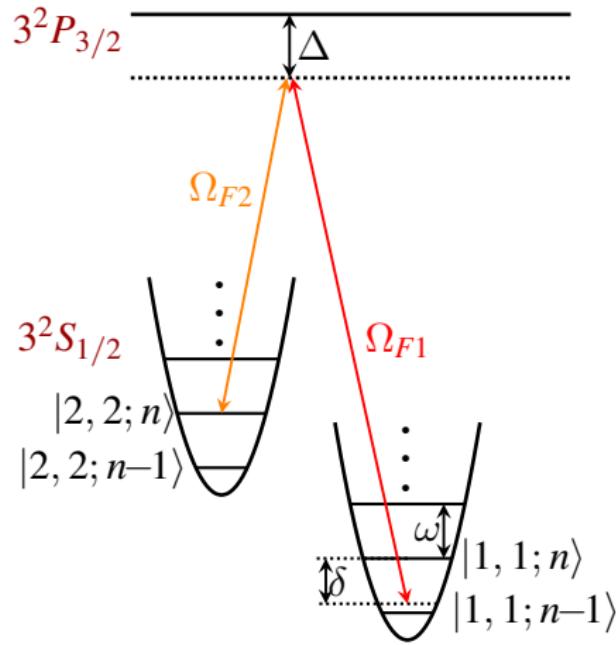
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4 Conclusion

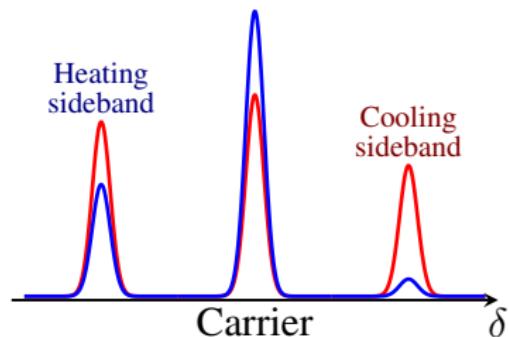
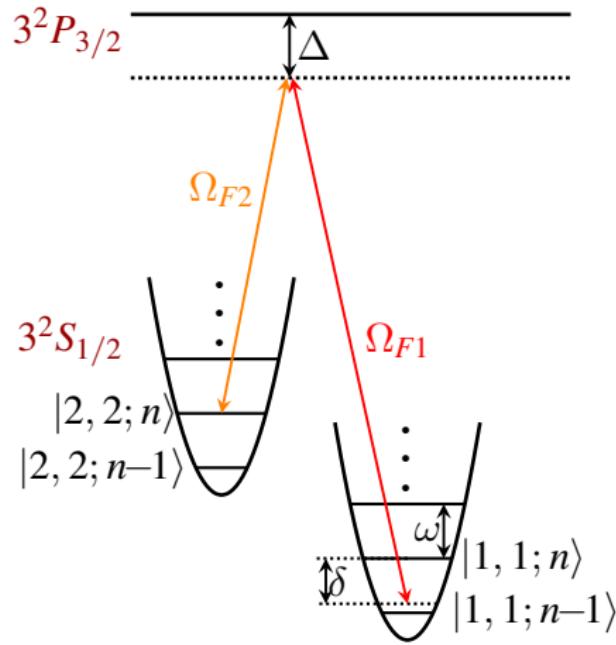
Raman sideband cooling



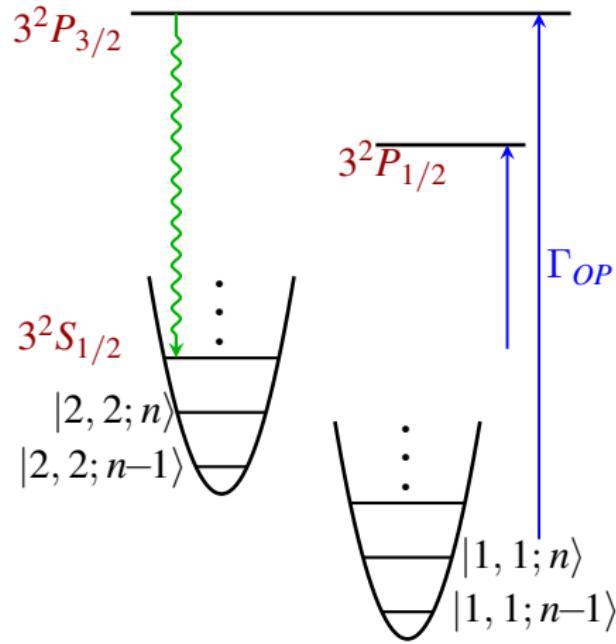
Raman sideband cooling



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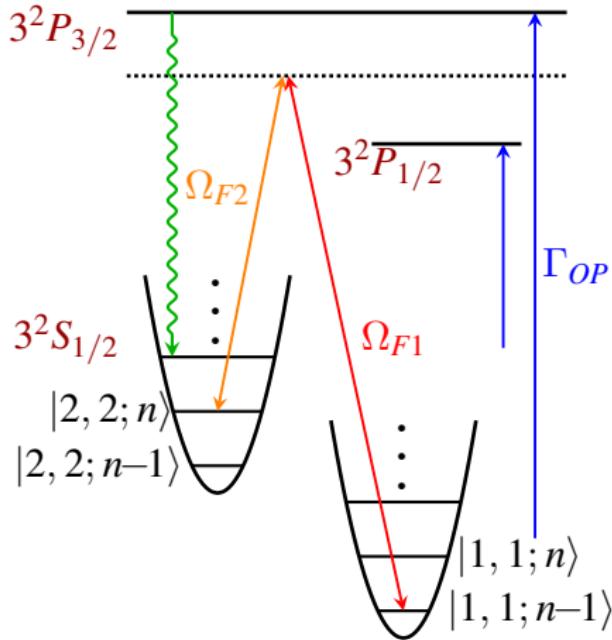
Raman sideband cooling



Raman sideband cooling

Lamb Dicke parameter

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

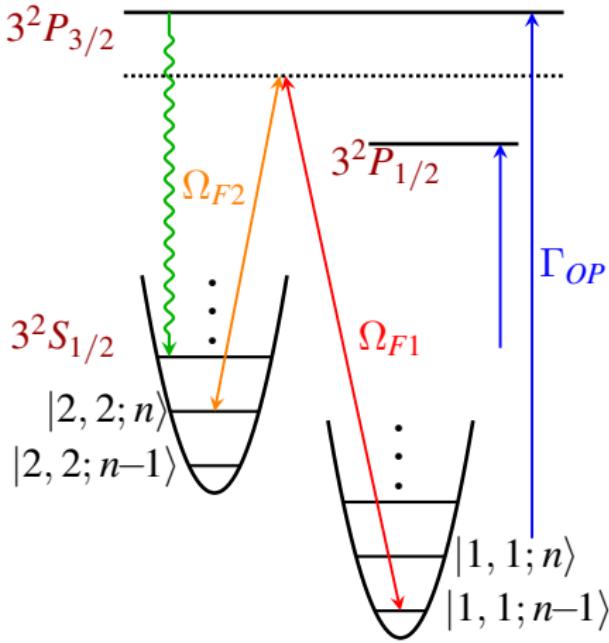


Raman sideband cooling

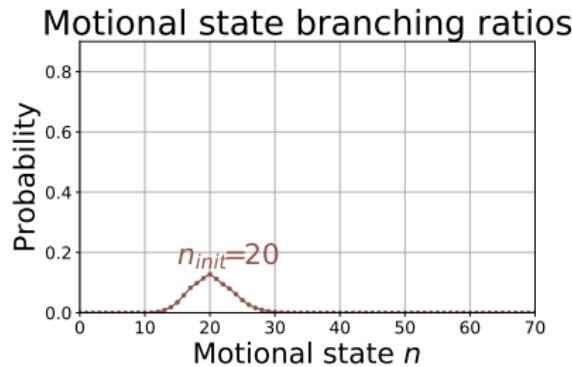
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$$\eta_{Na}^{OP} = 0.55$$



Raman sideband cooling



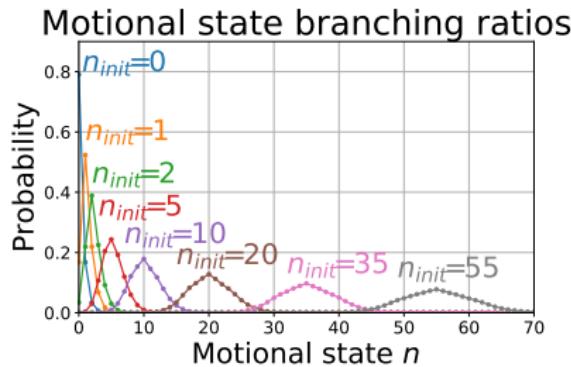
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- Motional state branching
- Coupling “dead zone”

Raman sideband cooling



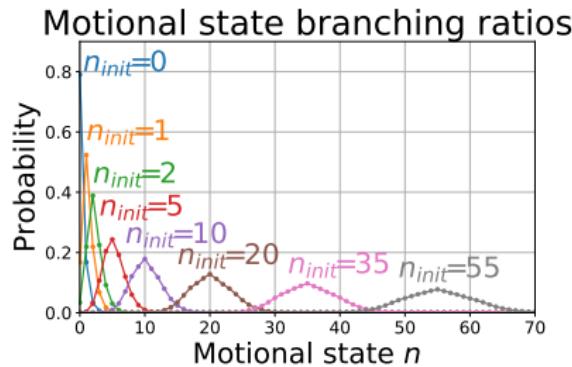
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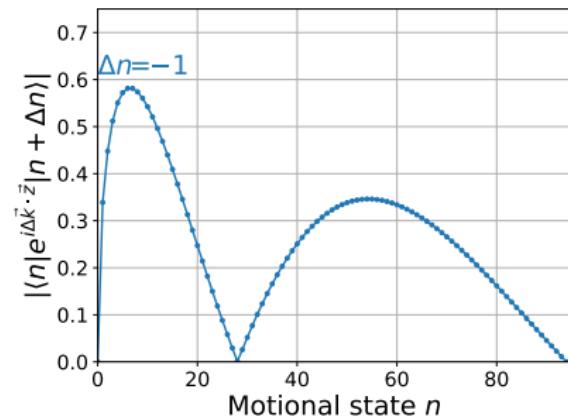


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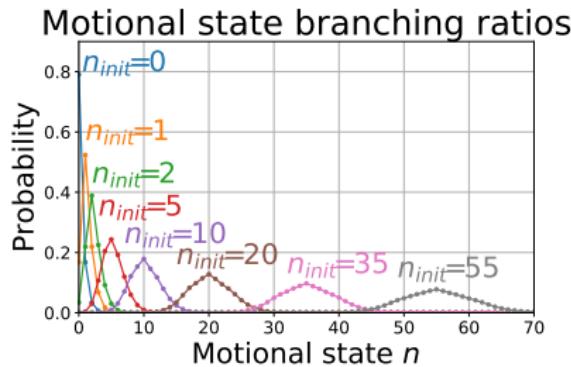
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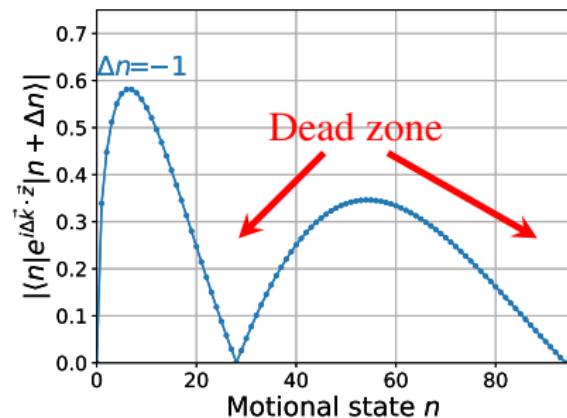


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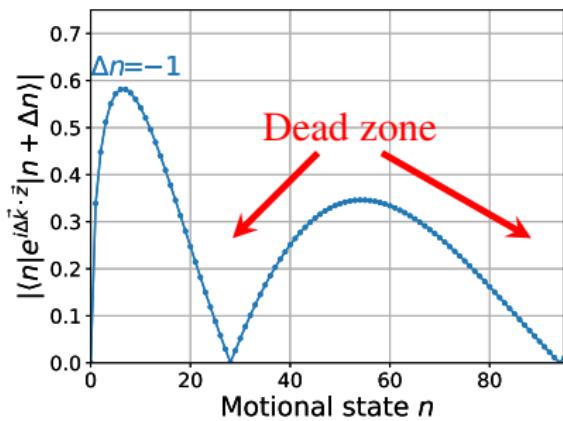
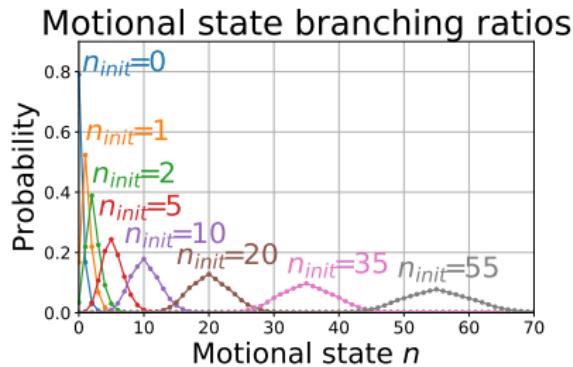
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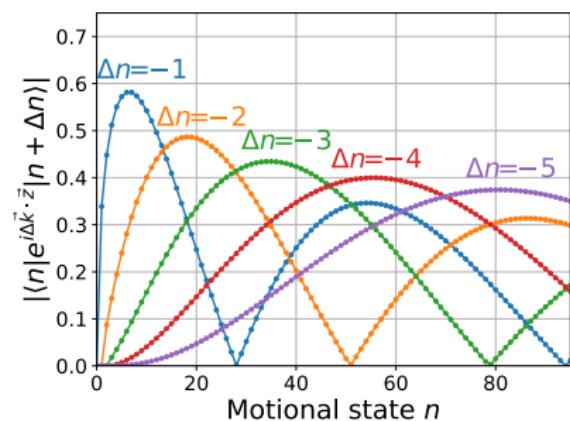
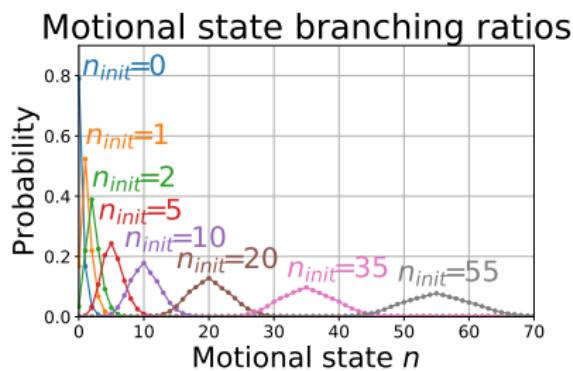
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Solution

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

Raman sideband cooling



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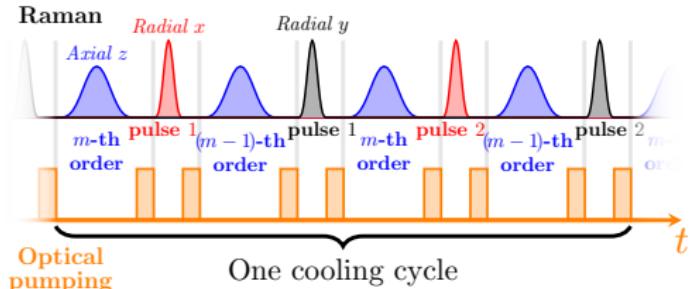
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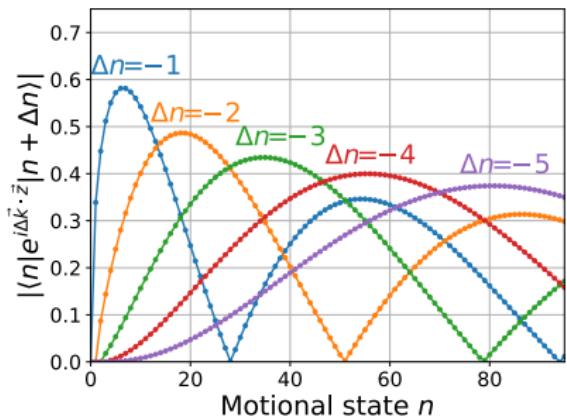
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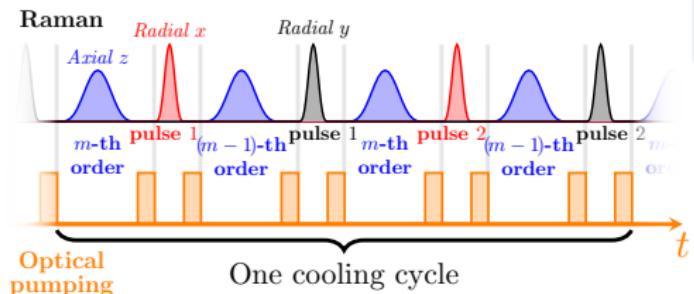
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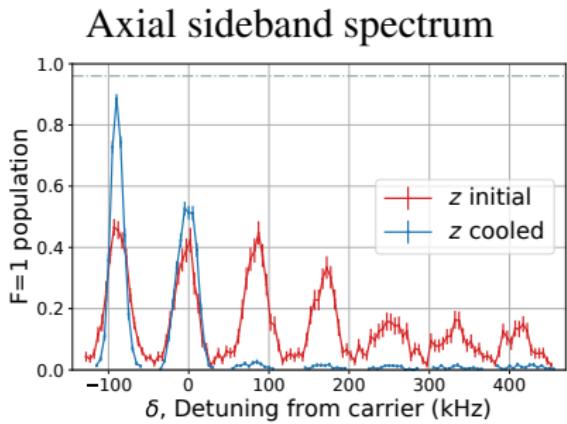
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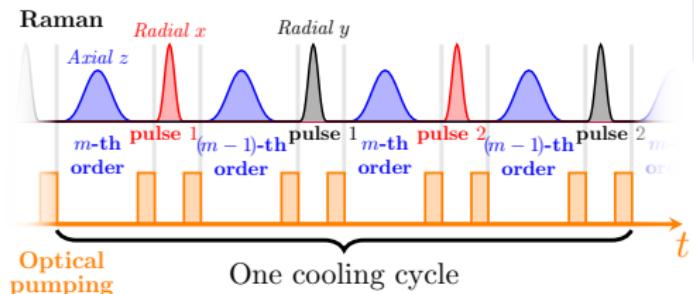
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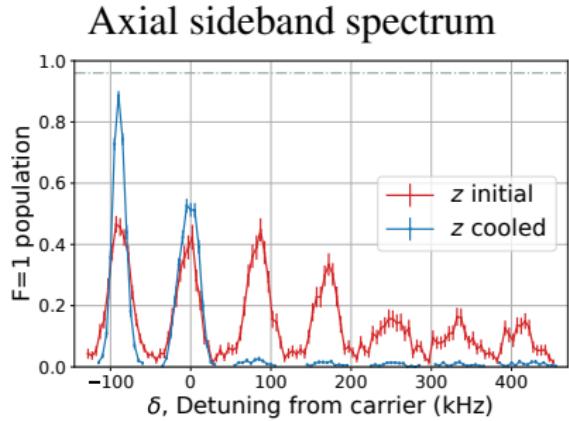
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3D ground state: 93.5(7)%

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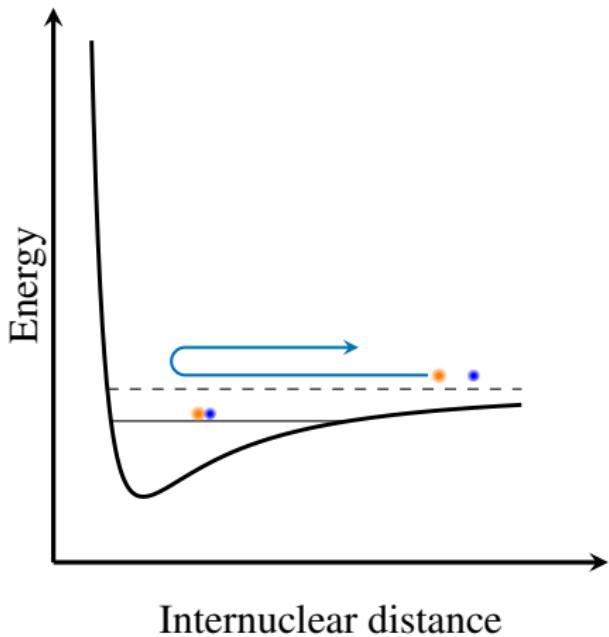
3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

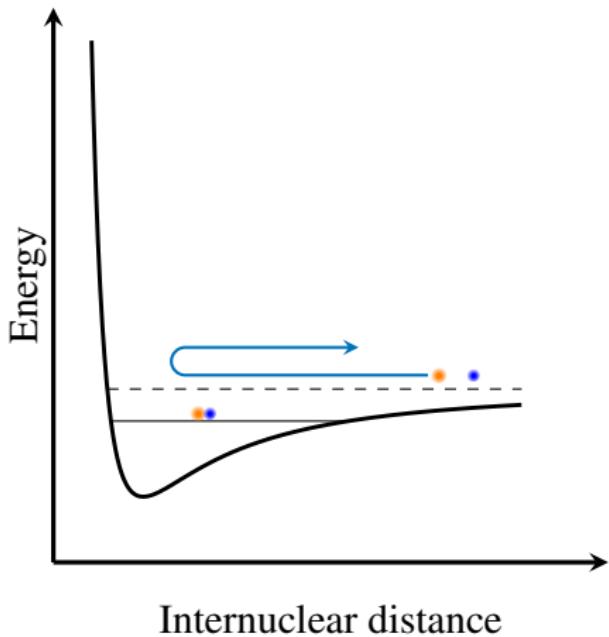
Scattering length a

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



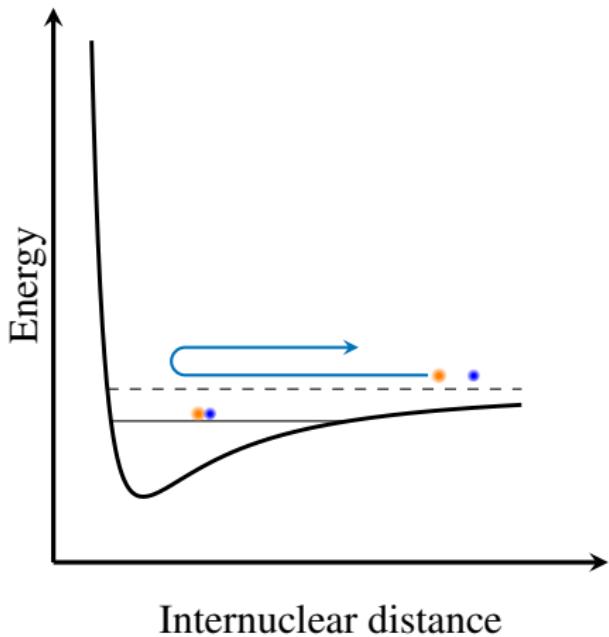
Scattering length a

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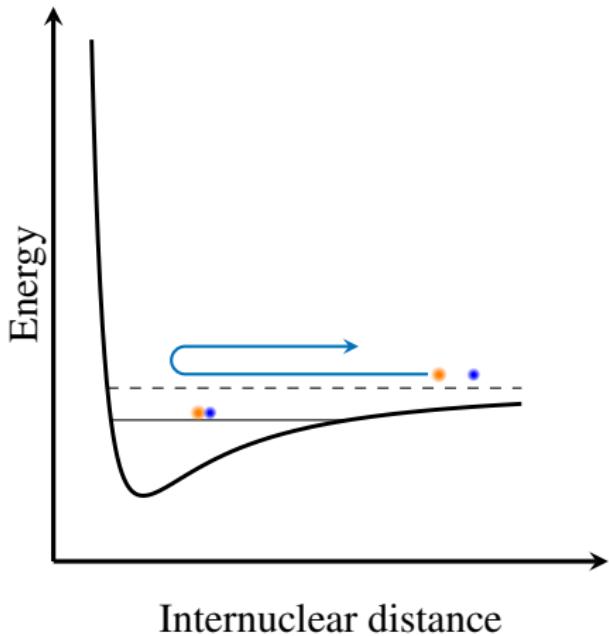
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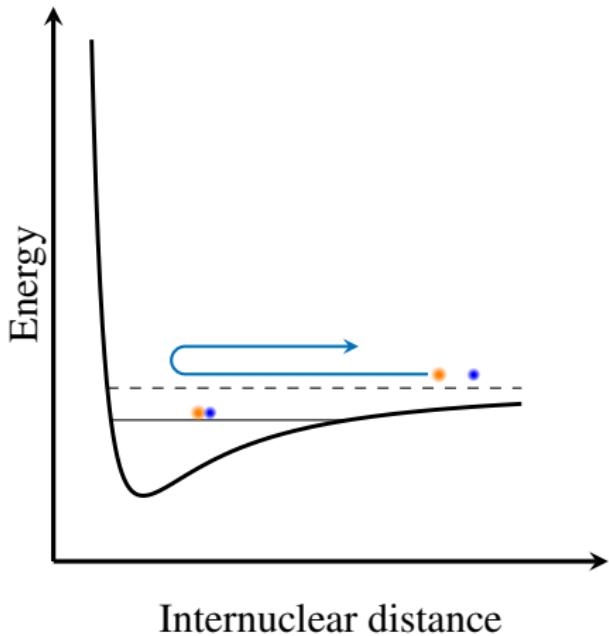
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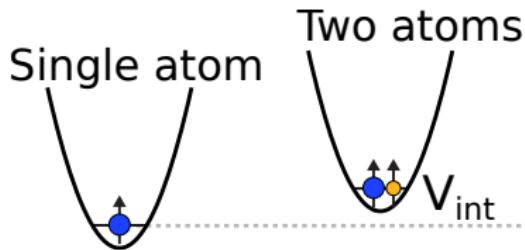


Scattering length a

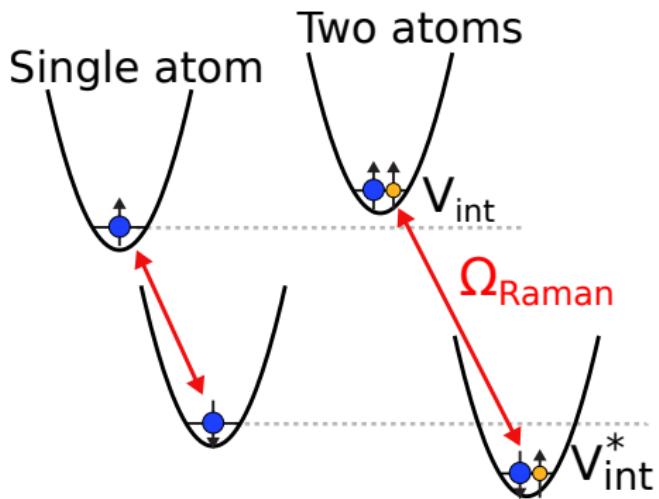
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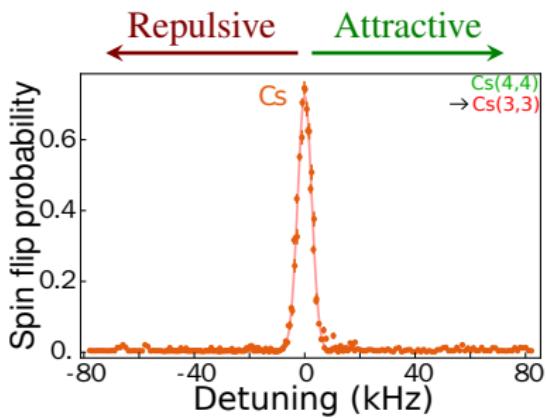
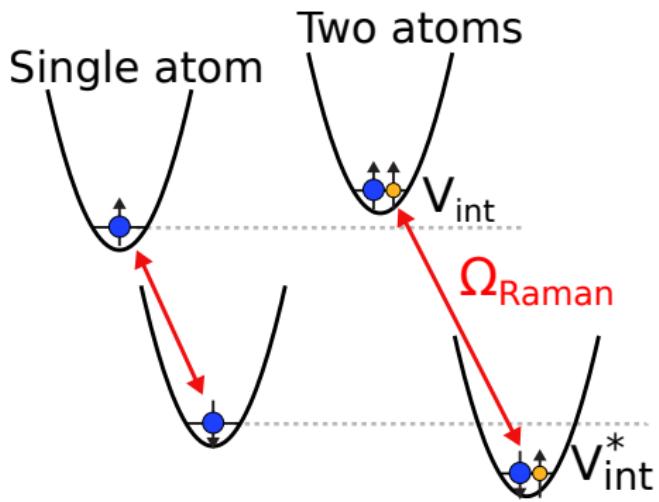
Interaction shift



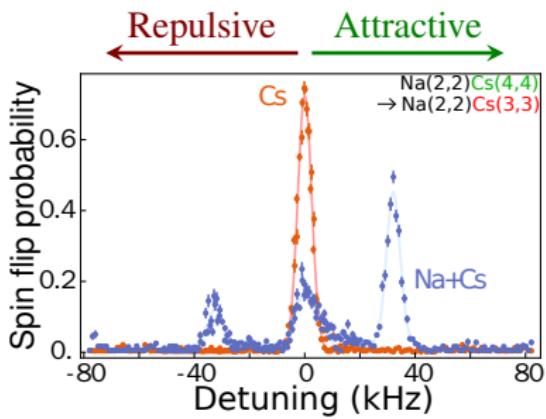
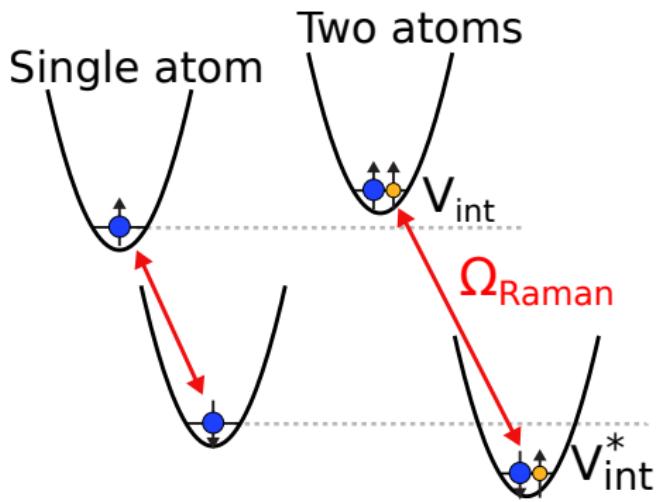
Interaction shift



Interaction shift



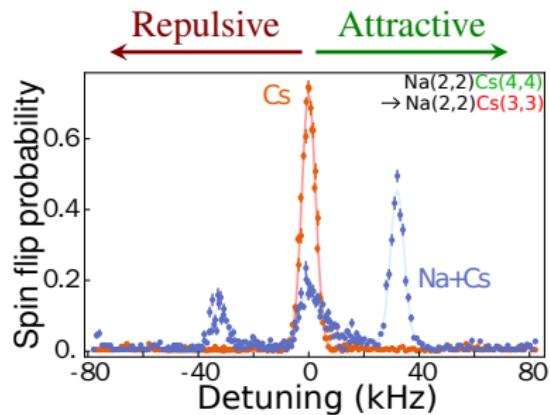
Interaction shift



Interaction shift

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

Interaction



Interaction shift

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

To center of mass
and relative coordinates

$$M = m_1 + m_2$$

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

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

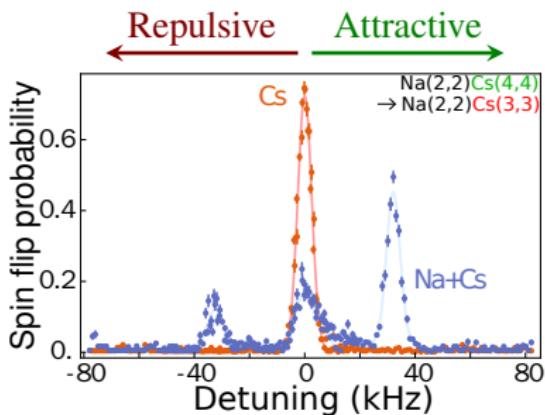
$$\omega_{R,i}^2 = \frac{m_2 \omega_{1,i}^2 + m_1 \omega_{2,i}^2}{m_1 + m_2}$$

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

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

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

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



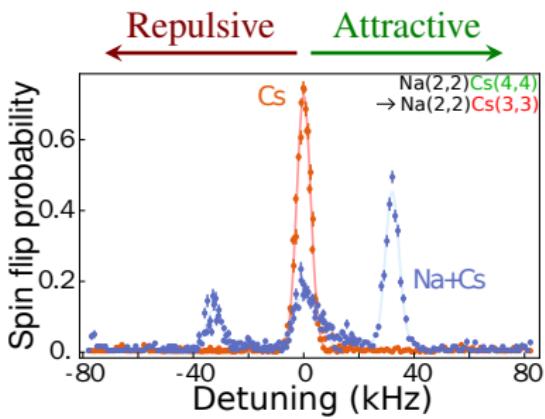
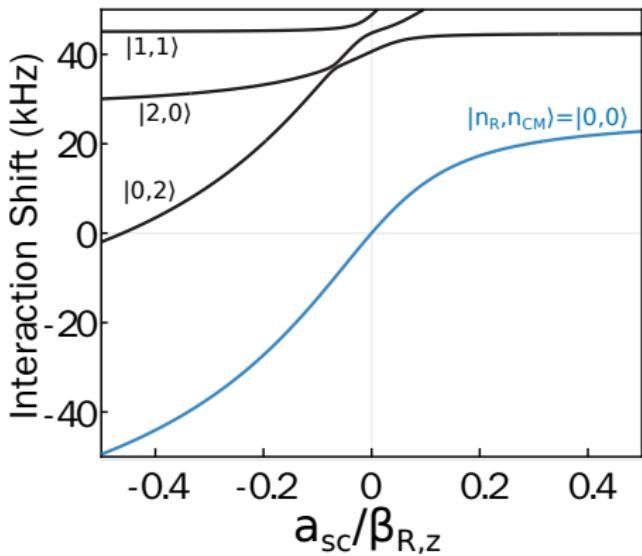
Center of mass

Relative

Mixing

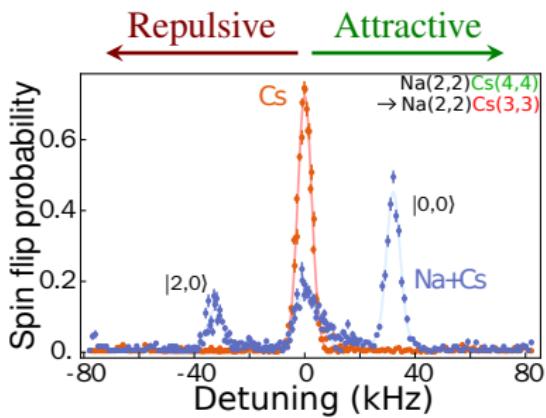
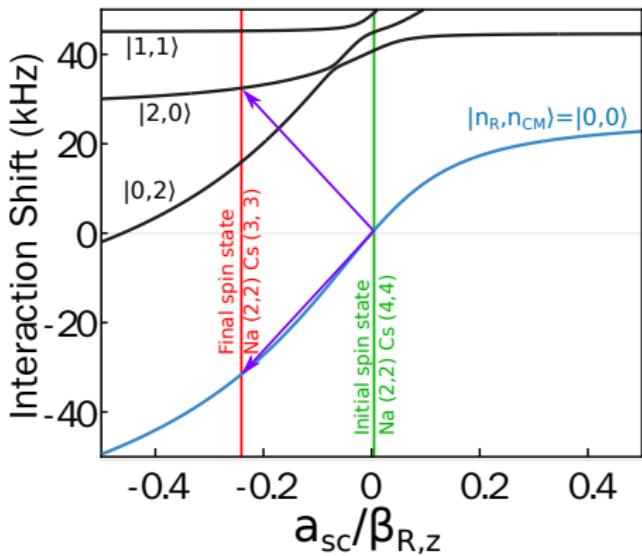
$$H = \overbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)} + \overbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)} + \overbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2)X_i X_{R,i}}$$

Interaction shift



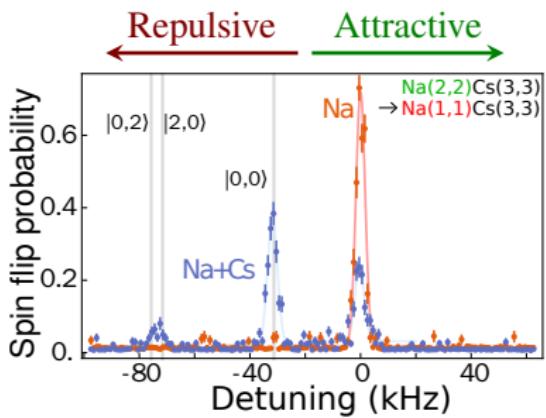
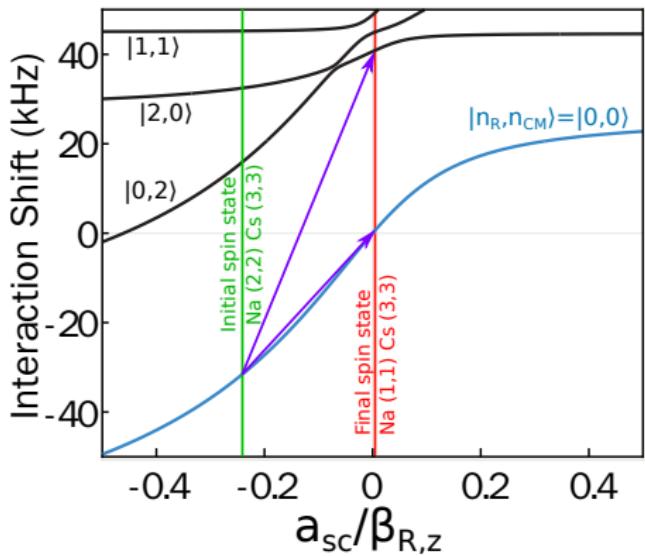
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



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

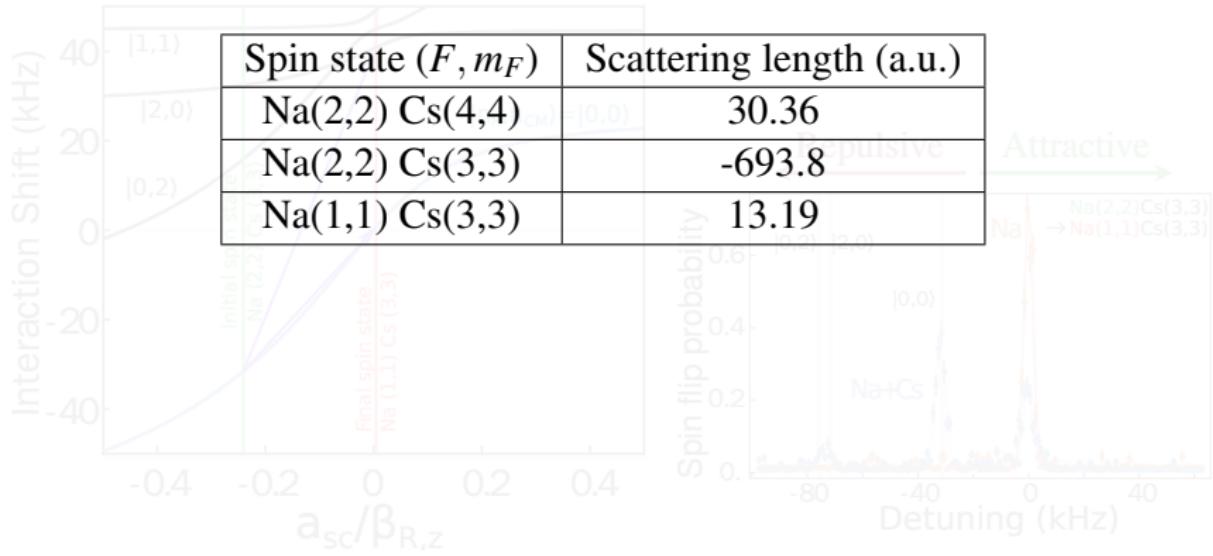
Interaction shift



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

Interaction shift

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



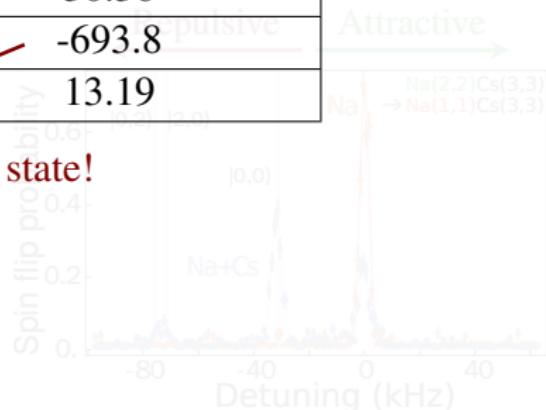
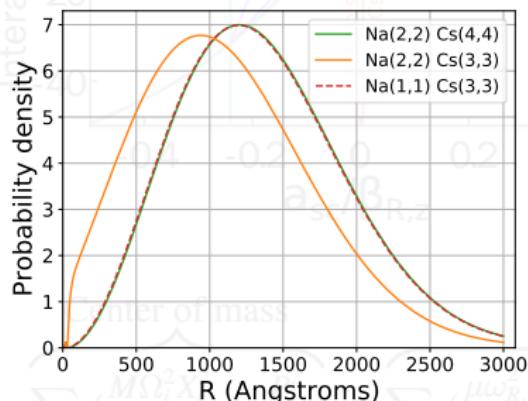
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

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!



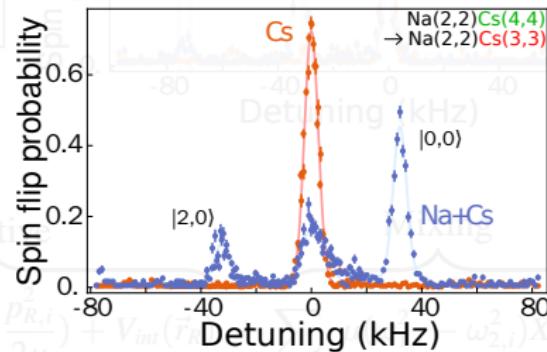
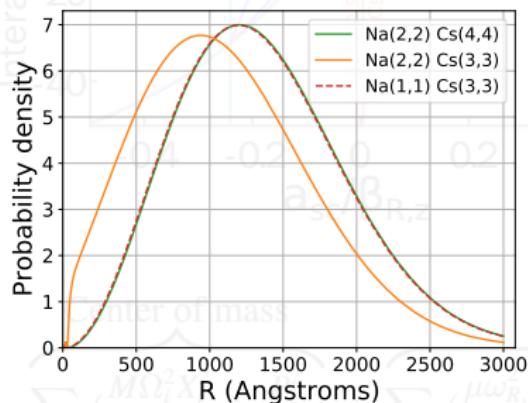
$$H = \sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i} X_{R,i}}{2M} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R) + \sum_{i=x,y,z} \mu (\omega_{1,i}^2 - \omega_{2,i}^2) X_{i,x} X_{R,i}$$

Interaction shift

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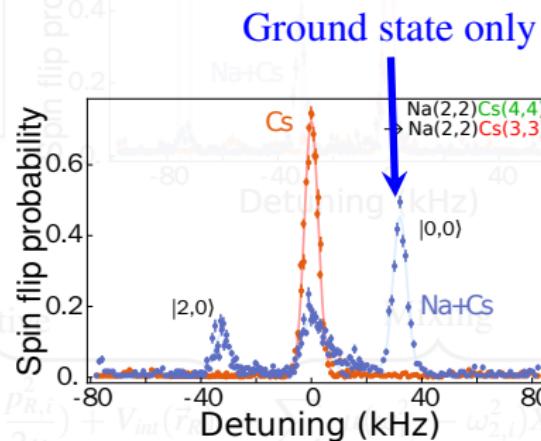
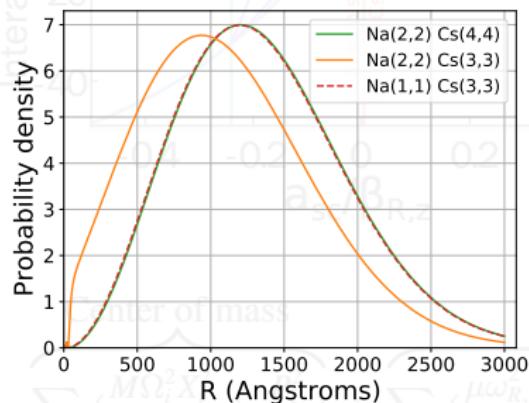


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Enhanced coupling to molecular state!



Outline

1 Experiment overview

2 Atom state control

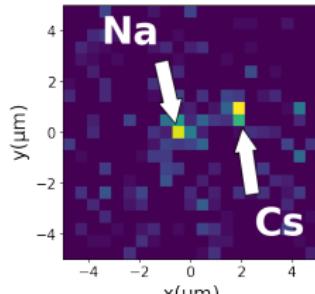
- Raman sideband cooling of Na atoms

3 Molecule creation

- Atom-atom interaction
- Coherent optical transfer

4 Conclusion

Loading

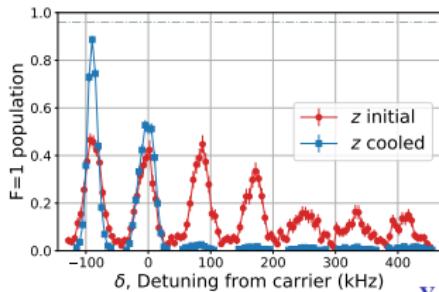


NJP. 19, 023007 (2017)

Merging



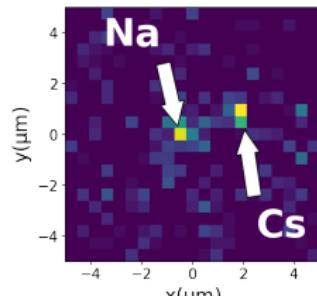
Cooling



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

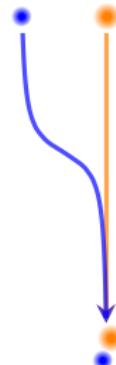
Y. Yu et al. PRA. 97, 063423 (2018)

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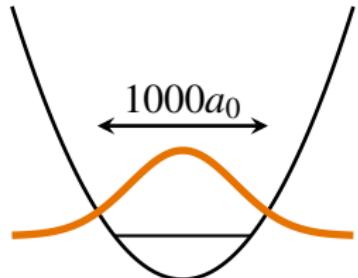


NJP. 19, 023007 (2017)

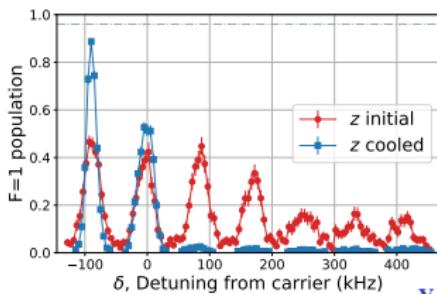
Merging



Atom



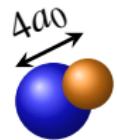
Cooling

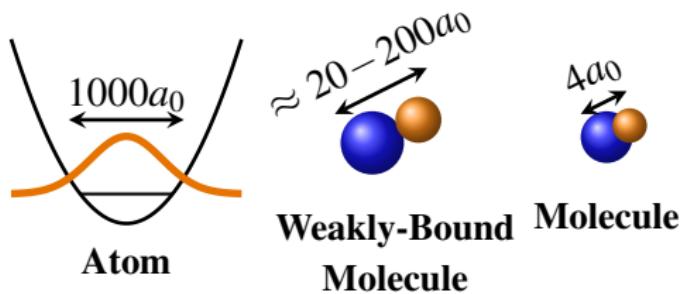


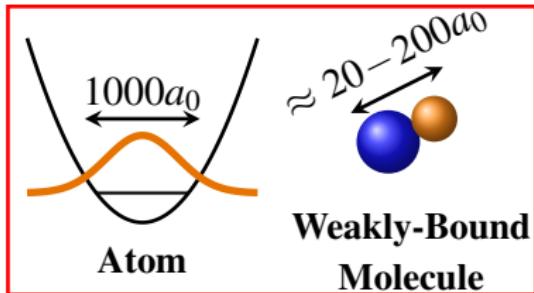
Y. Yu et al. PRA. 97, 063423 (2018)

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

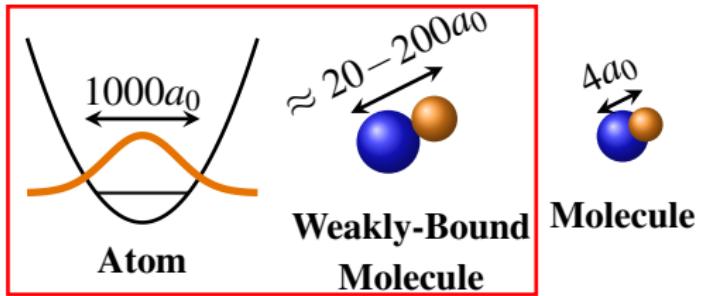
Molecule



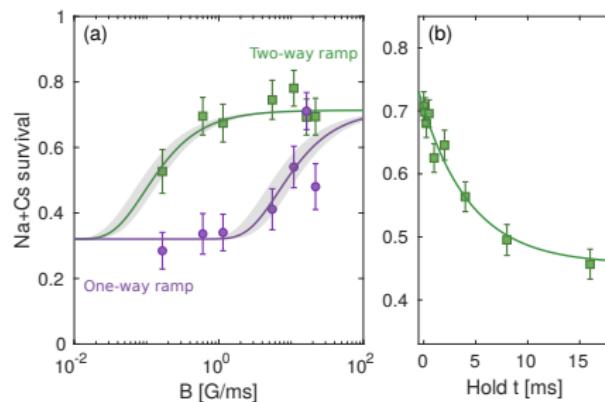




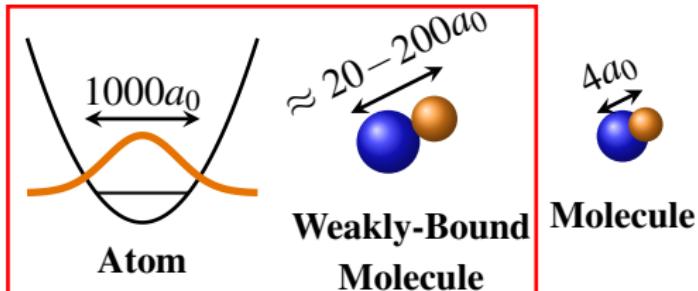
Molecule



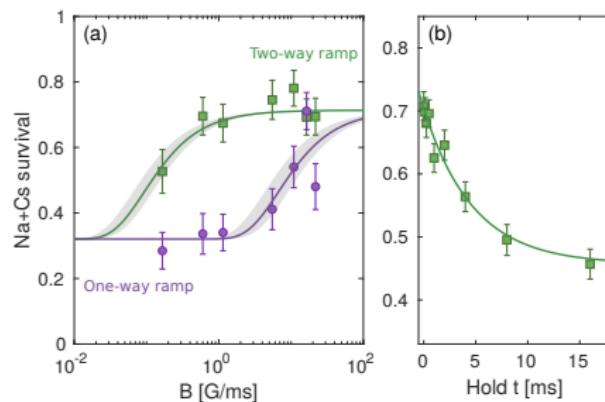
Feshbach molecule



PRL. 124, 253401 (2020)

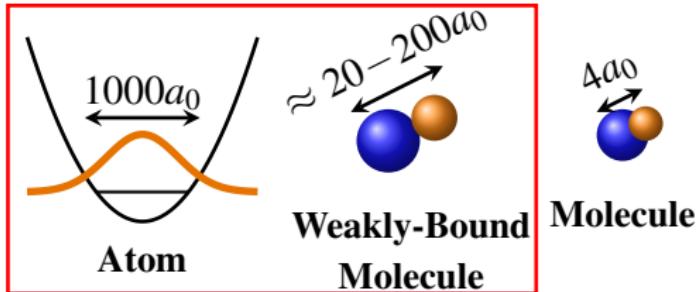


Feshbach molecule



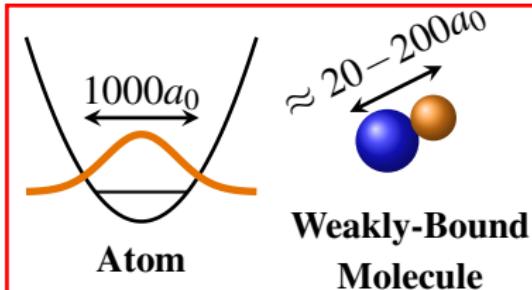
- Requires Feshbach resonance
- Usually large magnetic field

PRL. 124, 253401 (2020)



Optical transfer

- More general
- Faster

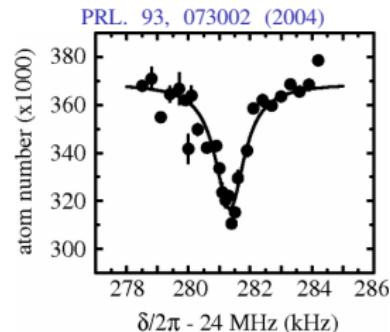


Optical transfer

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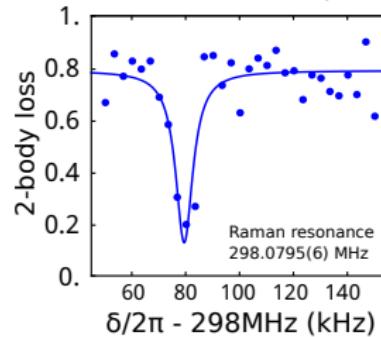
Previous results

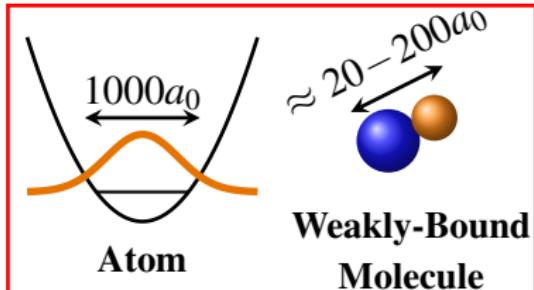
Rb_2 Science 287, 1016 (2000)



Sr_2 PRL. 109, 115302 (2012)

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





Molecule

Optical transfer

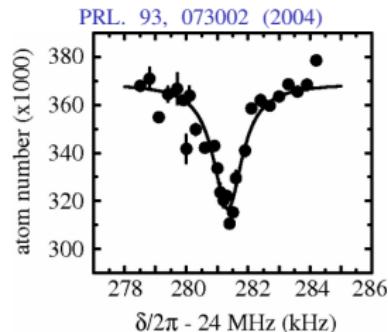
- More general
- Faster

Limitations so far

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

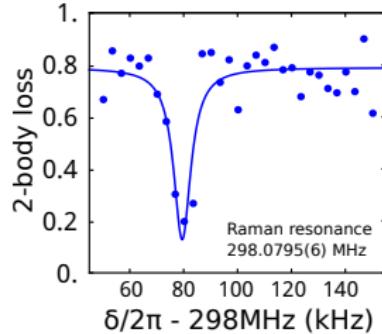
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Rb_2 Science 287, 1016 (2000)

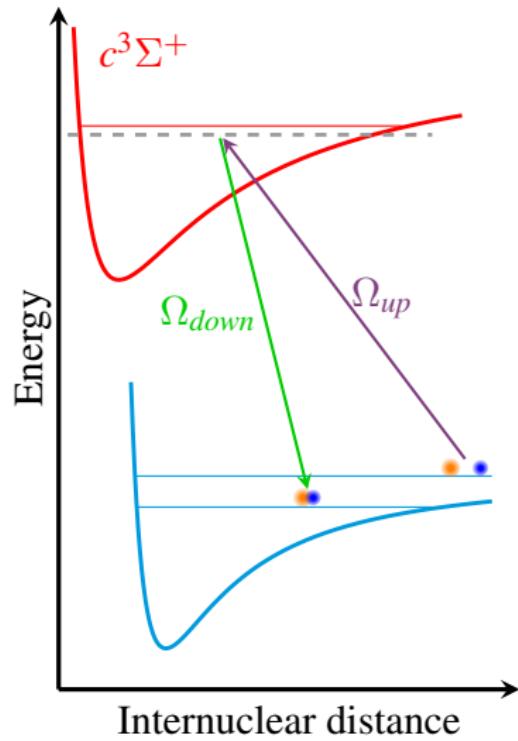


Sr_2 PRL. 109, 115302 (2012)

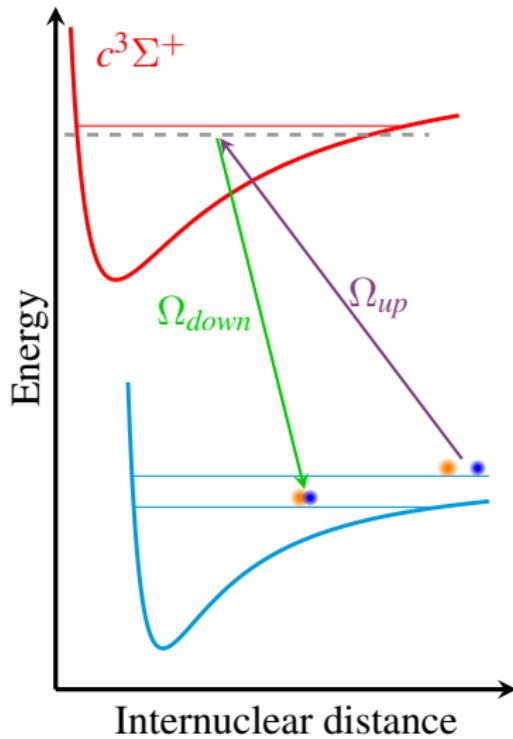
NaCs Y. Yu et al. PRX. 9, 021039 (2019)



Raman transfer



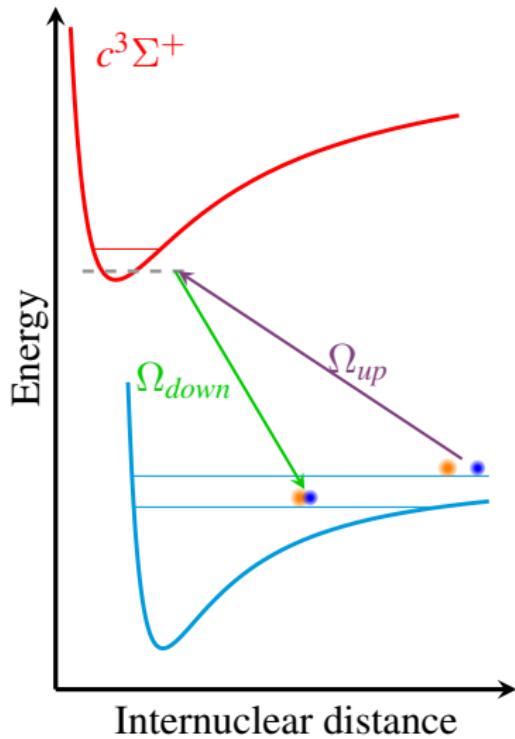
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

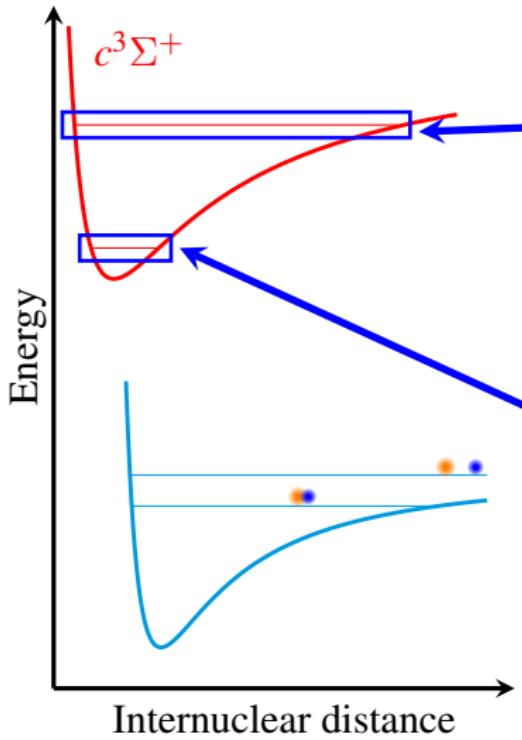
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

Deeply bound states

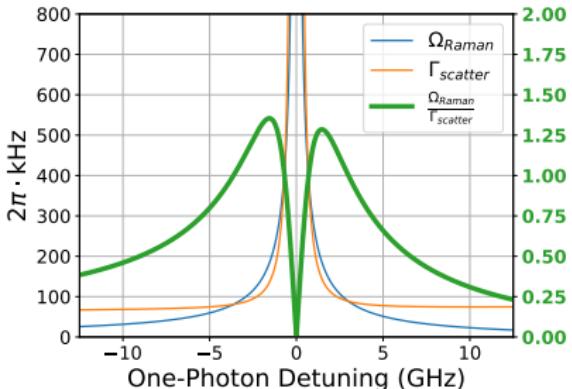
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

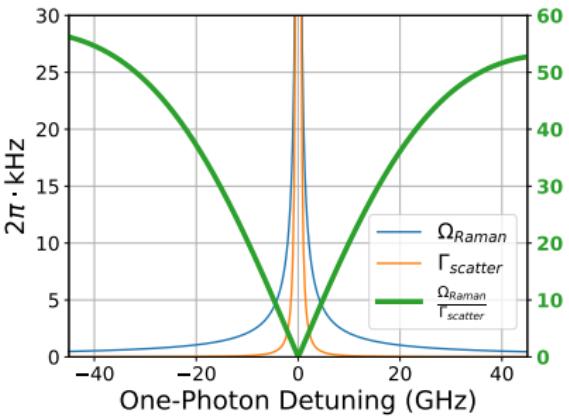
Raman transfer



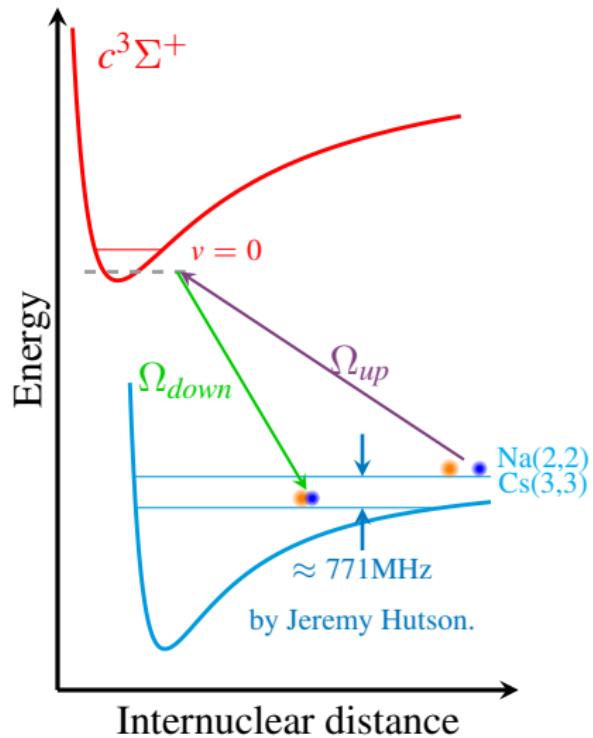
Near threshold states



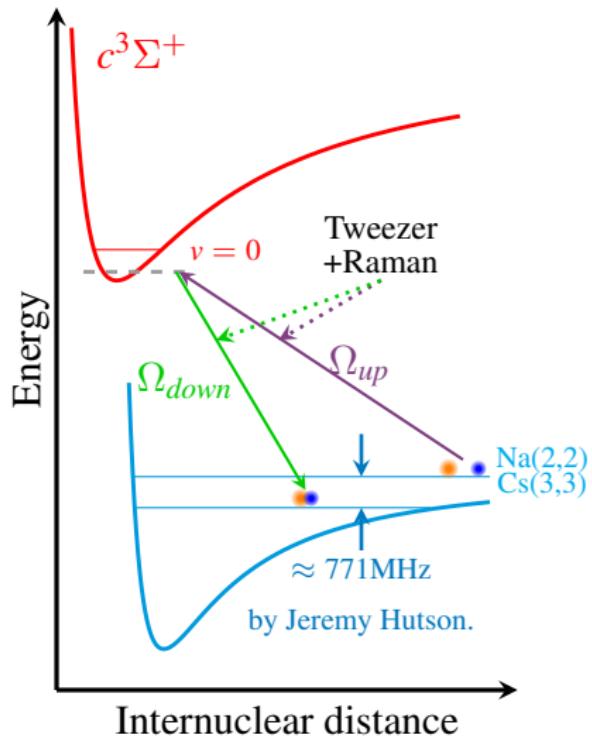
Deeply bound states



Experiment



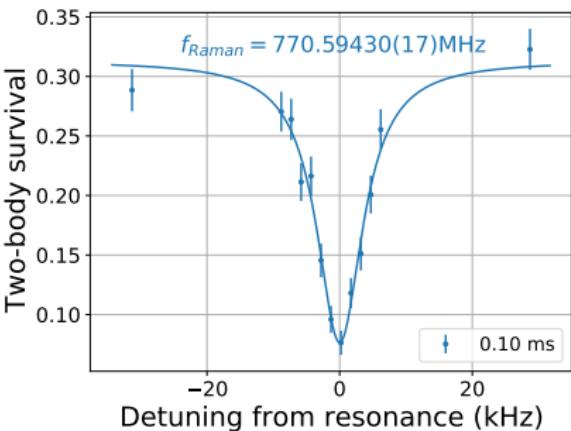
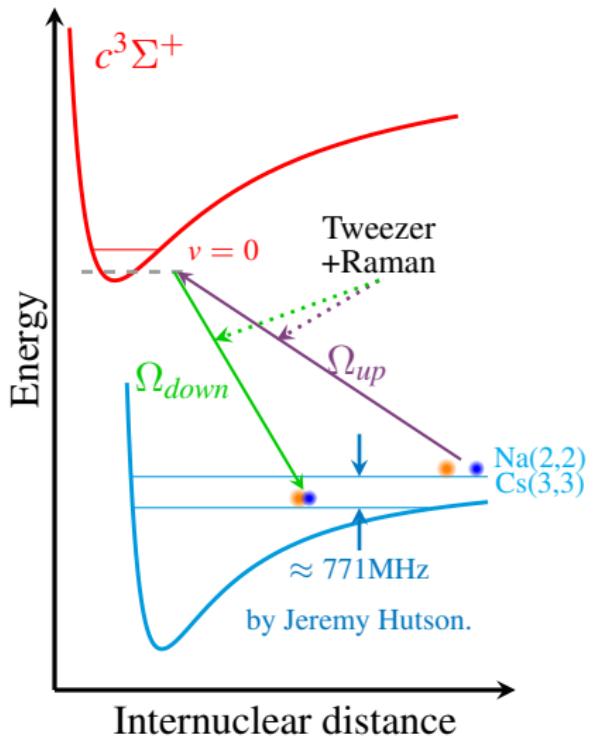
Experiment



Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

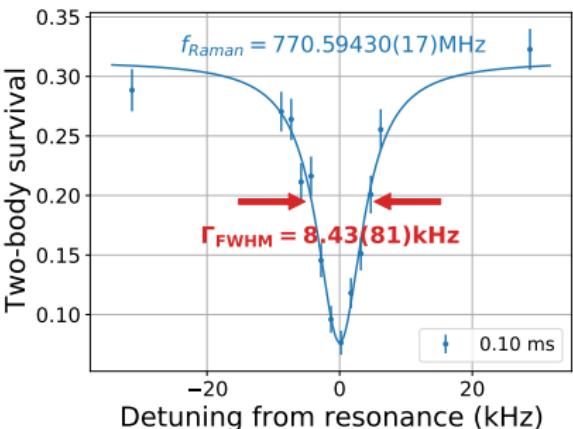
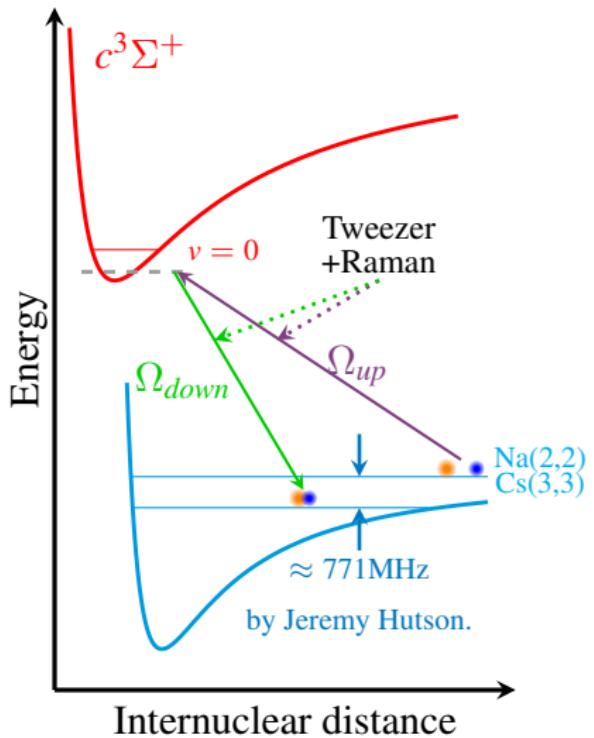
Experiment



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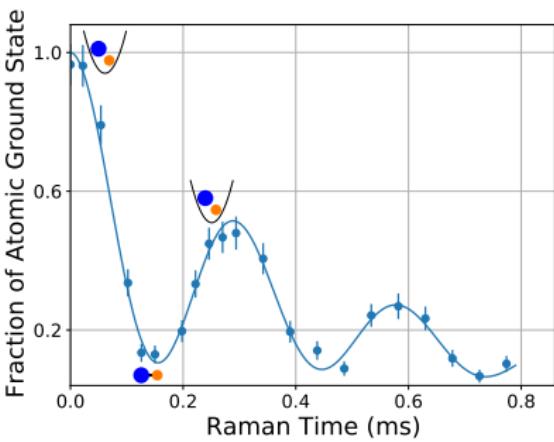
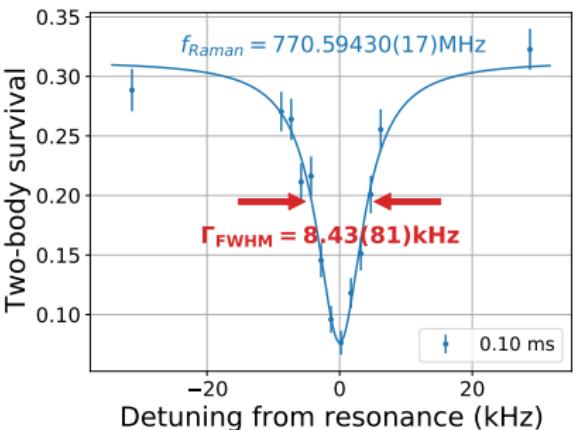
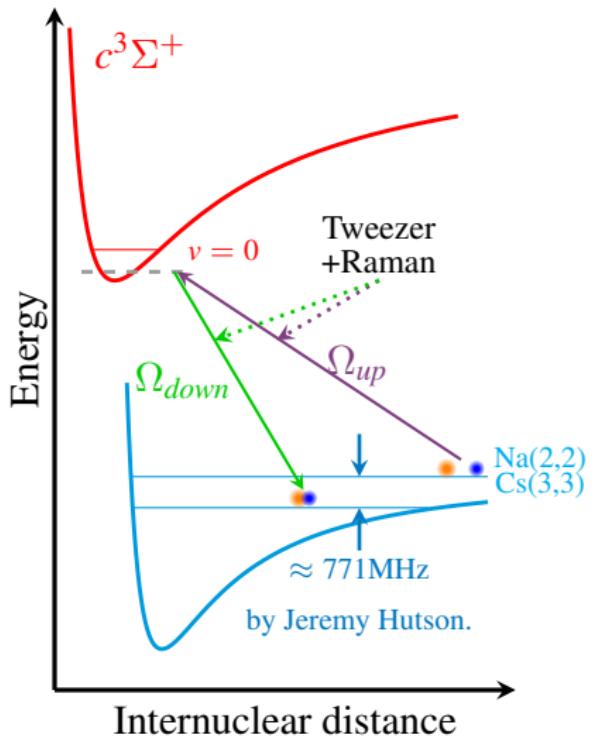
Experiment



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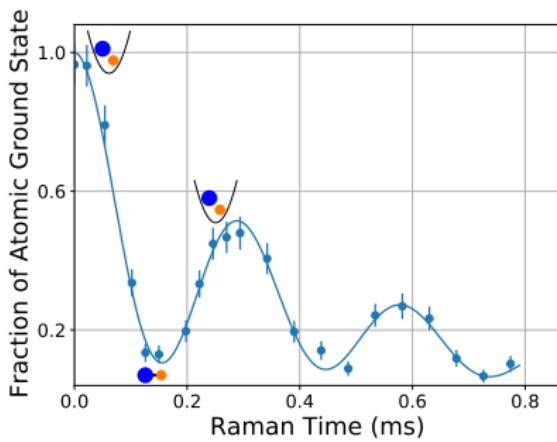
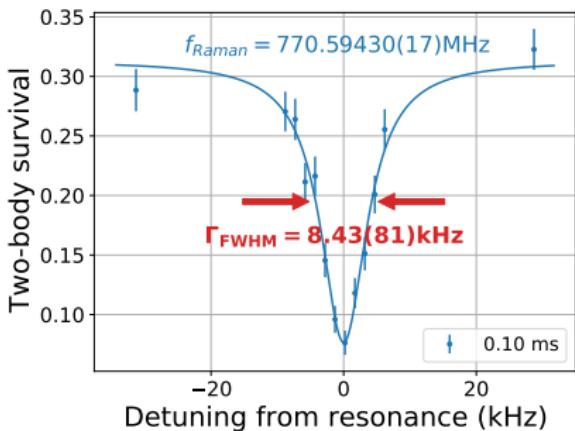
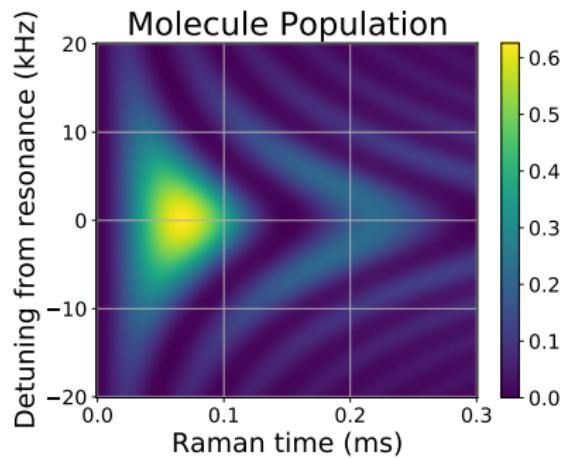
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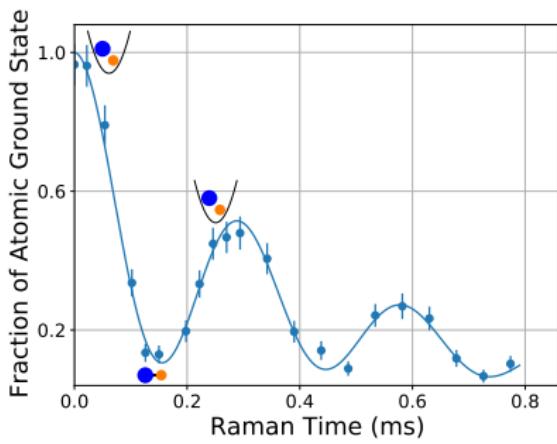
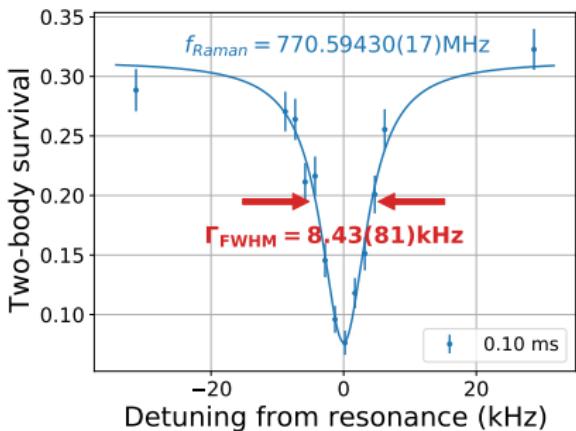
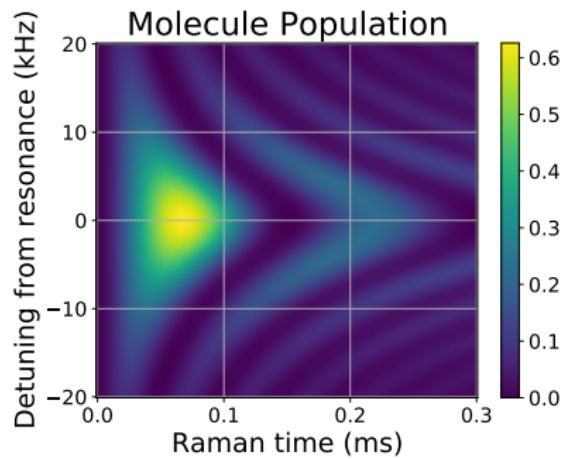
Experiment

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



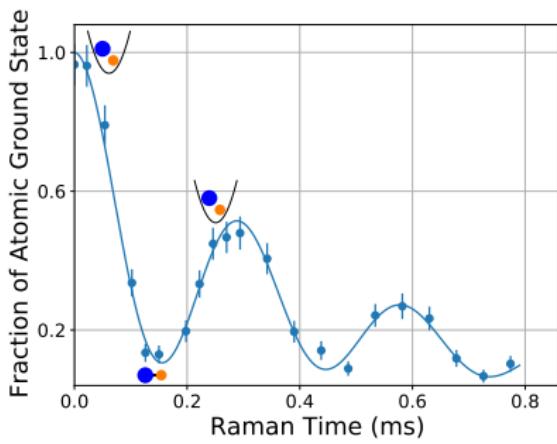
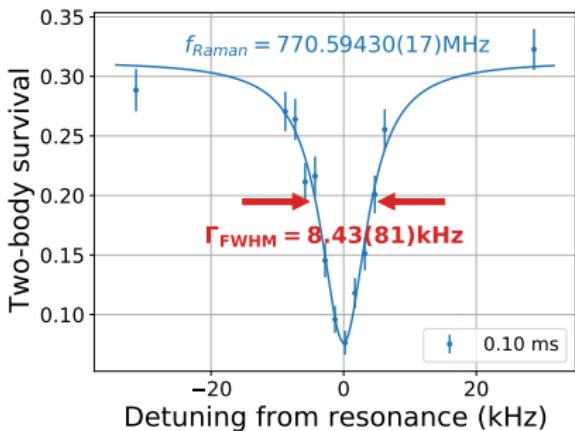
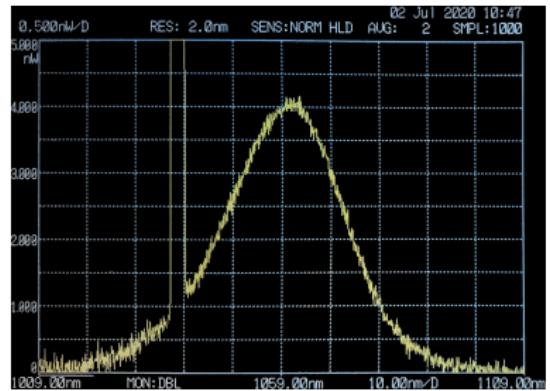
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Conclusion and outlook

- 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
- Working towards fully controlled, strongly interacting tweezer array

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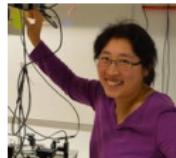
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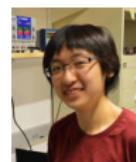
Experiment



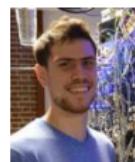
Kang-Kuen Ni



Kenneth
Wang



Jessie
Zhang



Lewis
Picard



William
Cairncross



Lee Liu
Postdoc @JILA



Jonathan Hood
Asstn Prof @Purdue



Nick Hutzler
Asstn Prof @Caltech

Theory



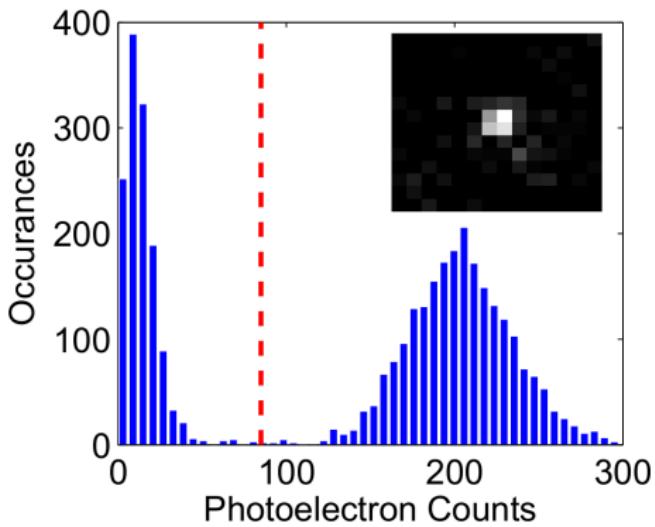
Jeremy Hutson

Single Atom in Tweezer

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

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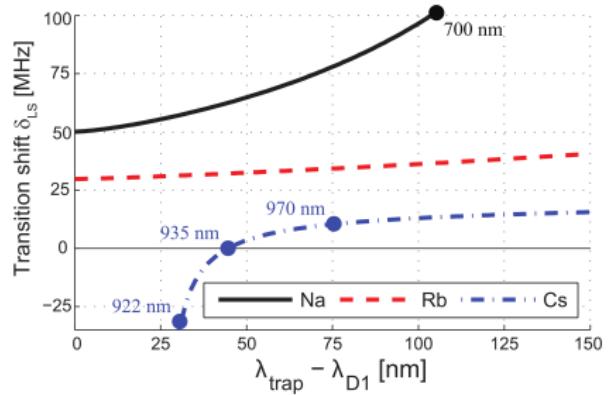
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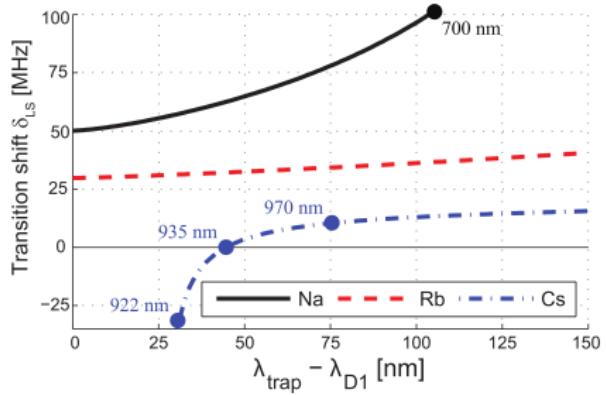
Issues with Na

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

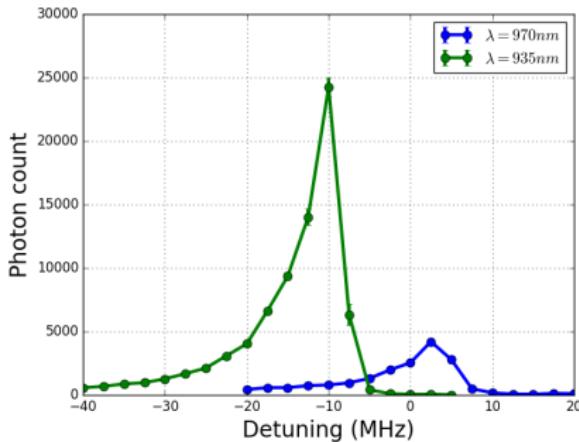
Real Issue with Na: Light Shift



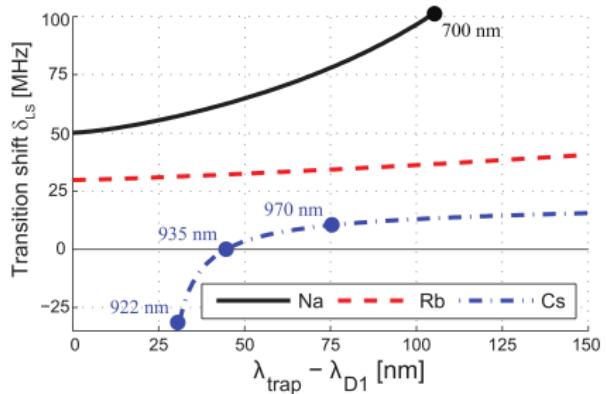
Real Issue with Na: Light Shift



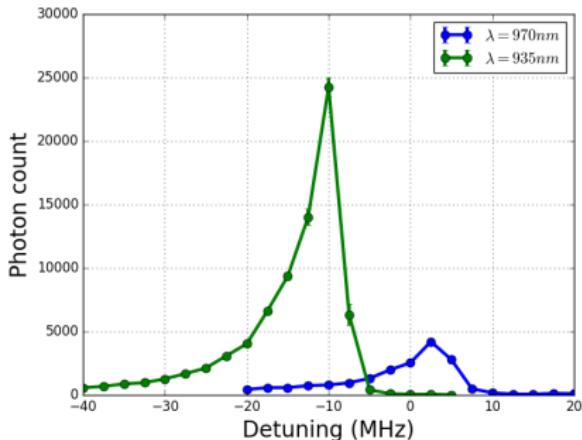
Cs single atom imaging



Real Issue with Na: Light Shift



Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

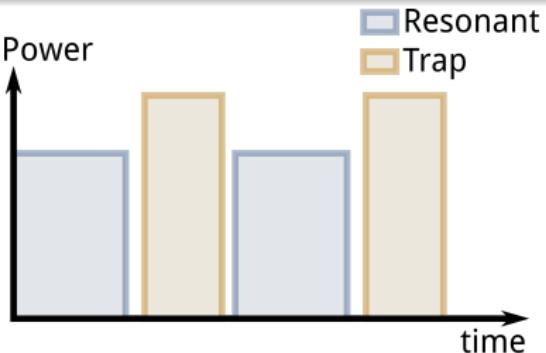
Real Issue with Na: Light Shift

Trap modulation

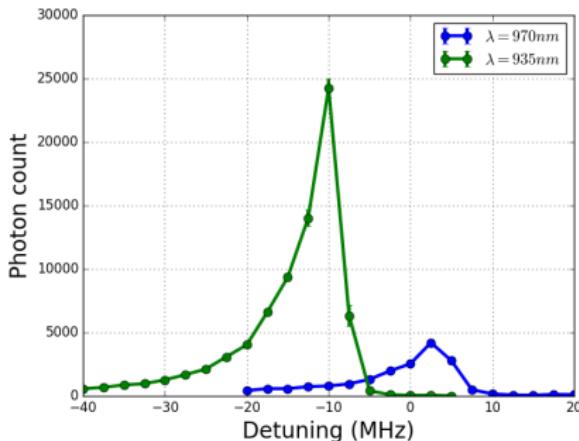
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



Cs single atom imaging



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- No cooling in tweezer

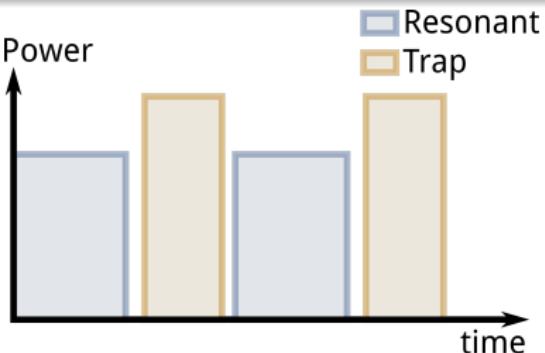
Real Issue with Na: Light Shift

Trap modulation

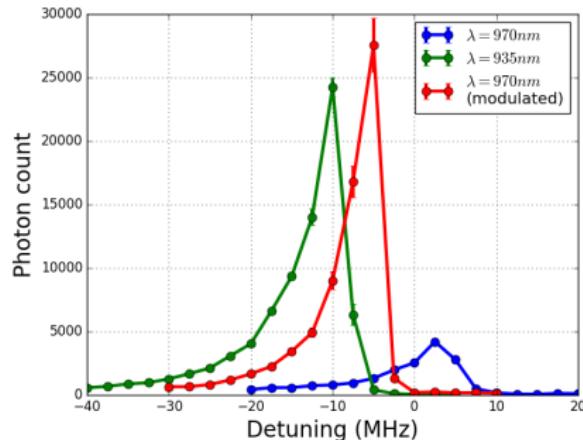
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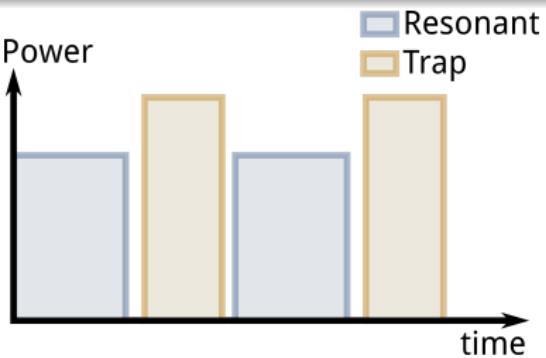
Real Issue with Na: Light Shift

Trap modulation

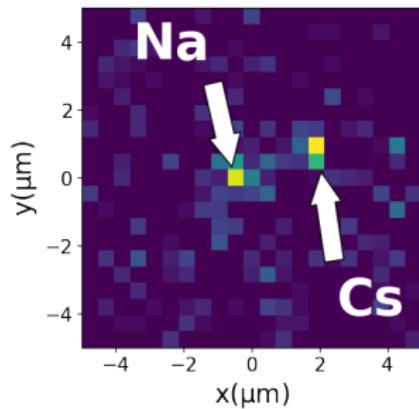
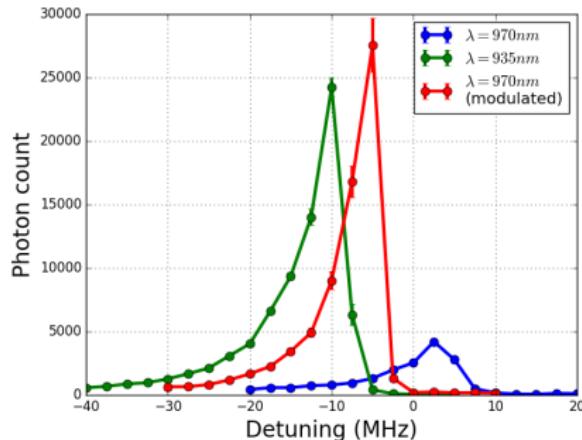
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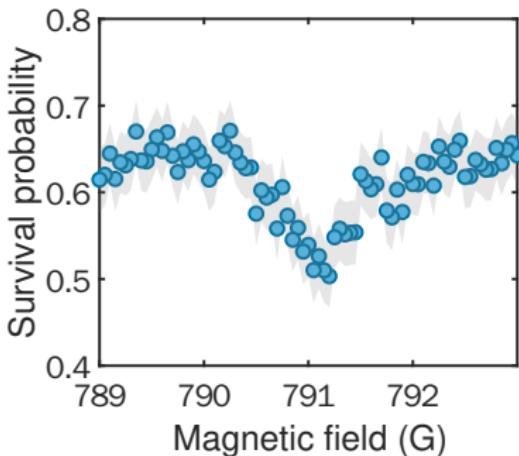
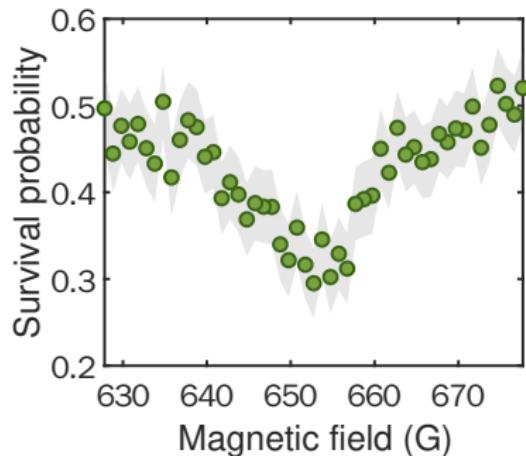
Cs single atom imaging



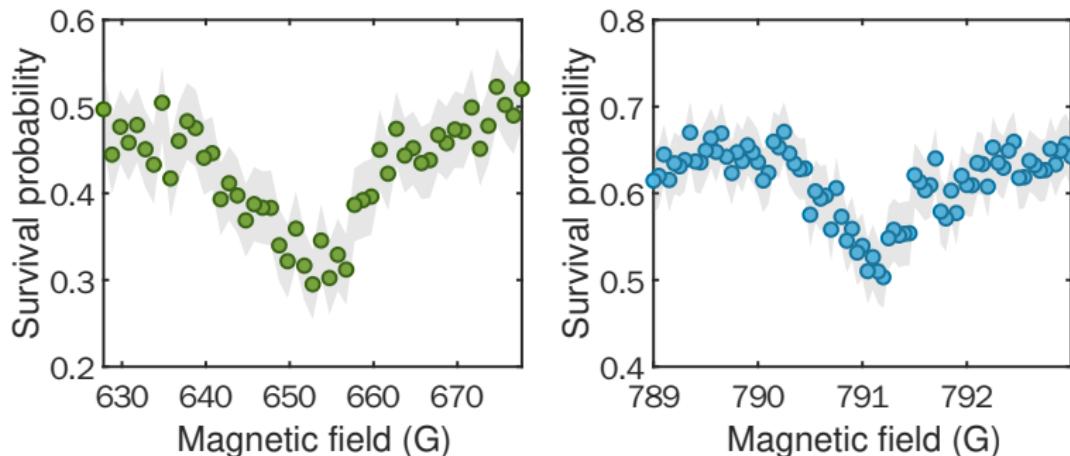
Na (1, -1) Cs (3, -3) Feshbach resonance



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



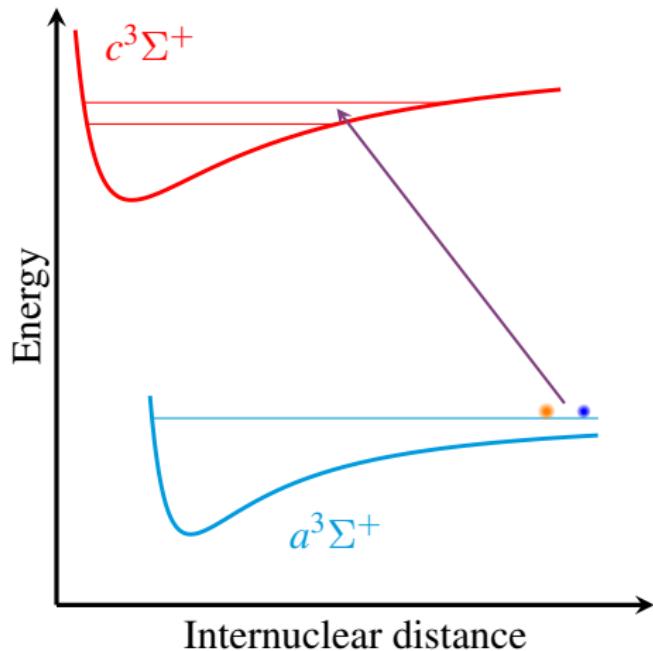
Na (1, -1) Cs (3, -3) Feshbach resonance



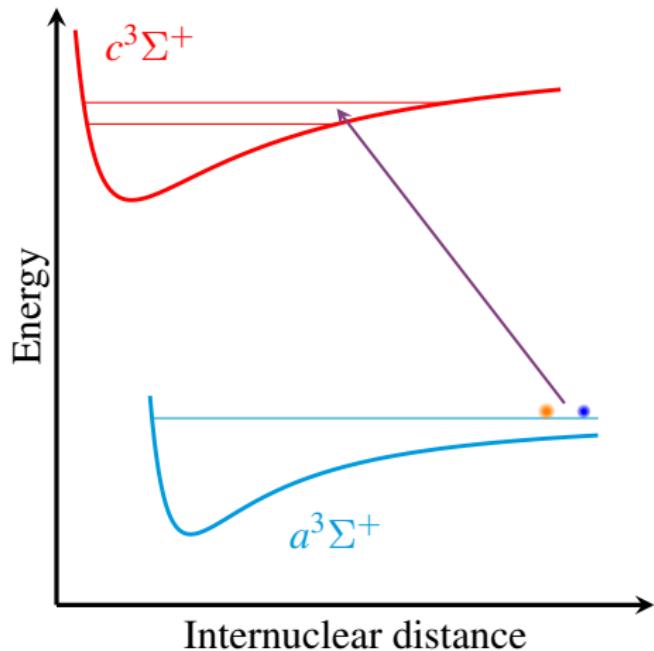
	<i>s</i> -wave	<i>p</i> -wave
Predicted (based on interaction shift) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



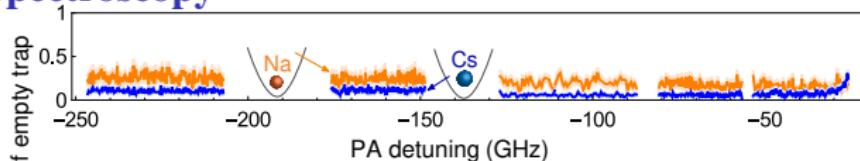
Photoassociation (PA) Spectroscopy



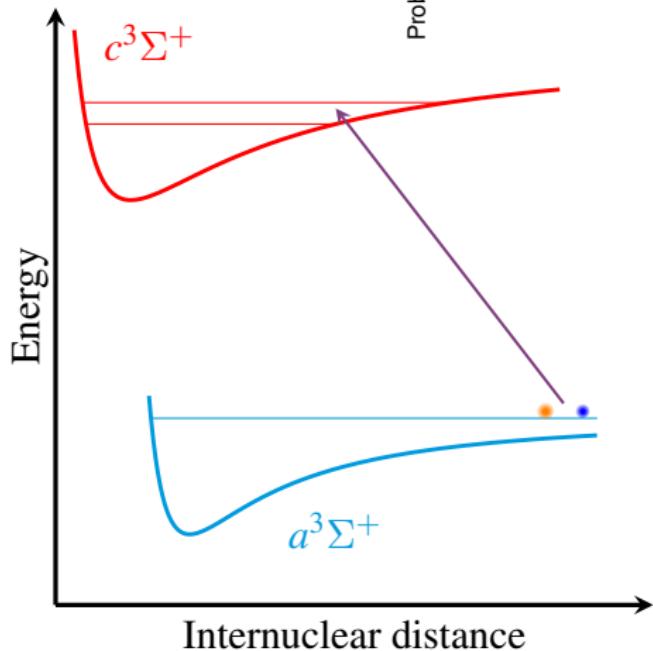
Single Atom PA

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

Photoassociation (PA) Spectroscopy



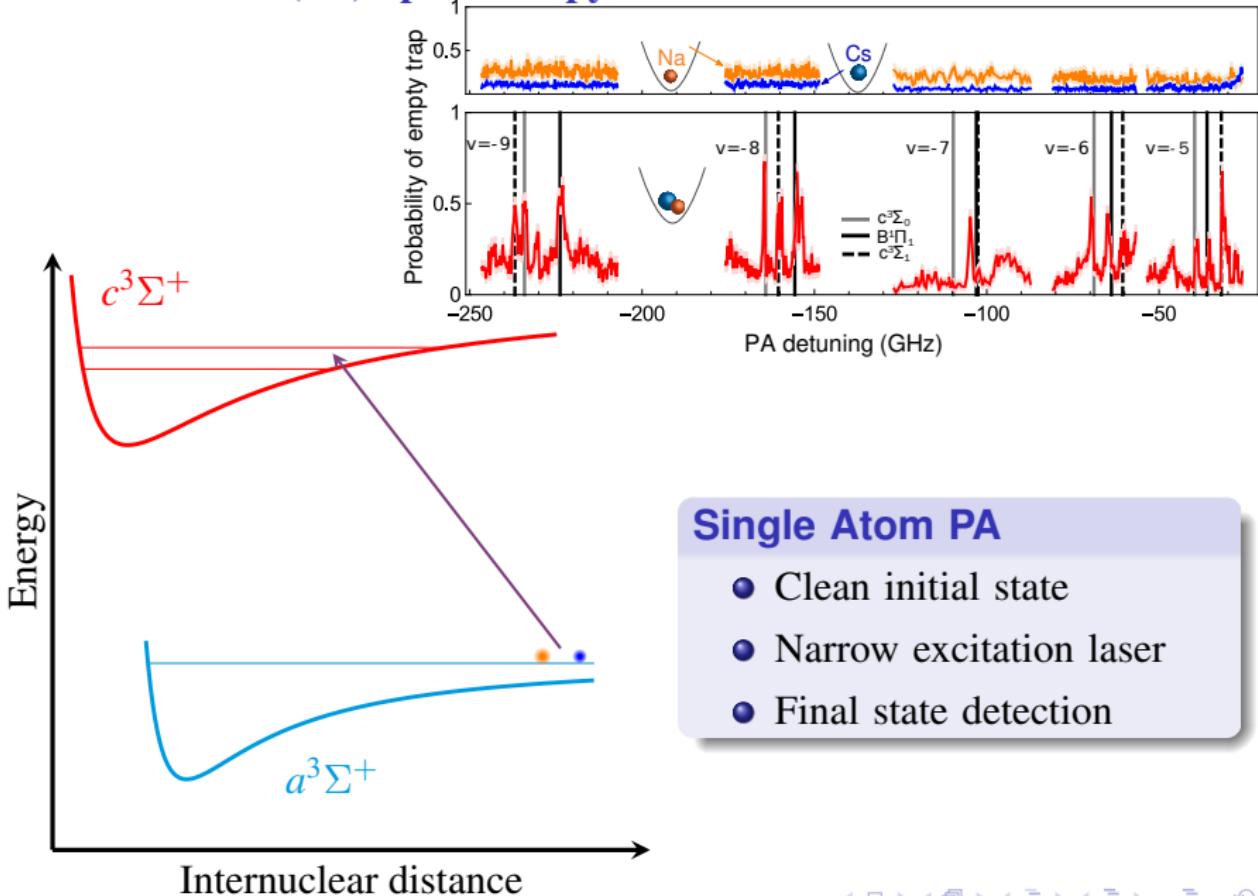
Probability of empty trap



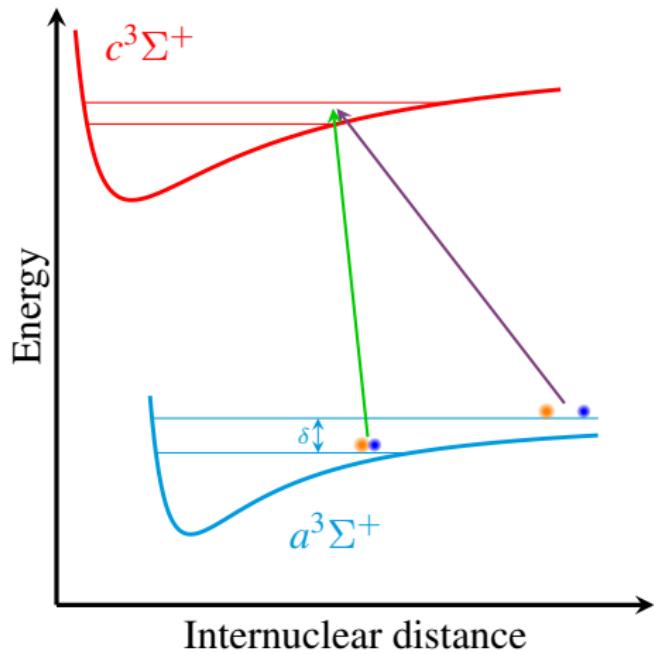
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Photoassociation (PA) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy

