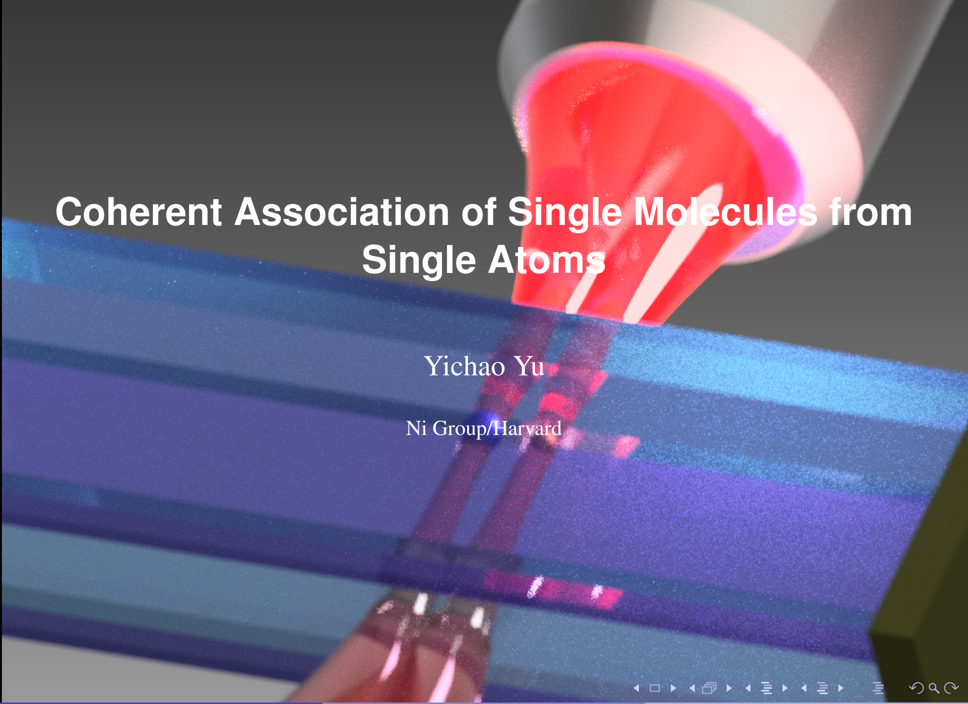
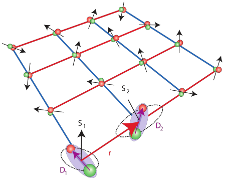


Coherent Association of Single Molecules from Single Atoms

Yichao Yu

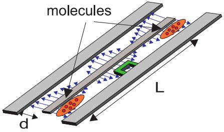
Ni Group/Harvard

Quantum Simulation



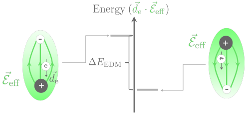
Nat. Phys. 2, 341 (2006)

Quantum Computing



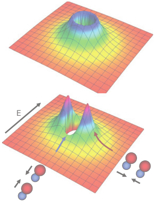
PRL. 97, 33003 (2006)

Precision Measurement



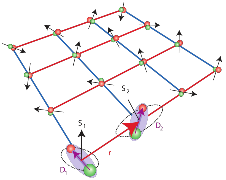
Science 343, 269 (2014)

Quantum Chemistry



Nature 464, 1324 (2010)

Quantum Simulation



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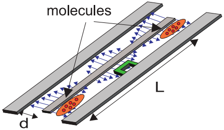
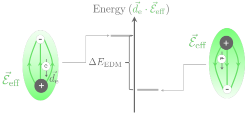


PRL. 97, 33003 (2006)

- Full quantum control
- Entanglement
- ...

Precision Measurement



Science 343, 269 (2014)

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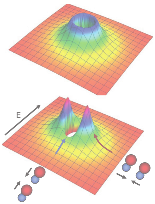

Nature 464, 1324 (2010)

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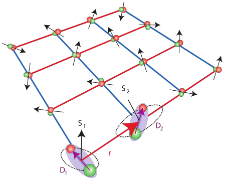

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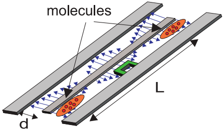

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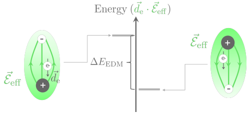

Science 343, 269 (2014)

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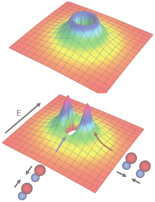

Nature 464, 1324 (2010)

New Approach?

Entanglement

Single particle control

Entanglement

i.e. interaction

Single particle control

Entanglement

i.e. interaction

Single particle control

Dipolar molecules

Entanglement

i.e. interaction

Single particle control

Dipolar molecules

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

Entanglement
i.e. interaction

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Single particle control

Optical tweezers

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Single particle control

Optical tweezers

- Single site resolution
- . . .

Entanglement

i.e. interaction

Dipolar molecules

Single particle control

Optical tweezers

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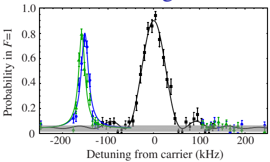
- . . .

Cooling



PRX. 2, 041014 (2012)

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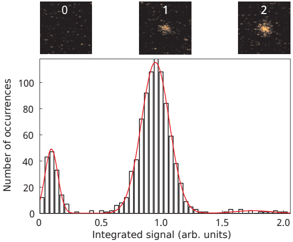
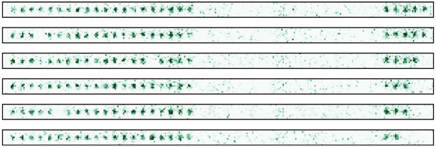


Nat. Phys. 6, 951 (2010)

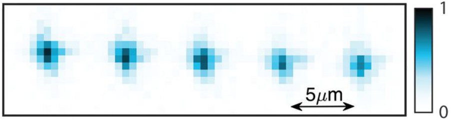
Rearranging



Science 354, 1024 (2016)

Ultracold molecule in tweezers

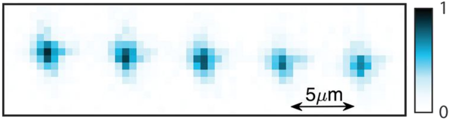
Direct cooling



Science 365, 1156 (2019)

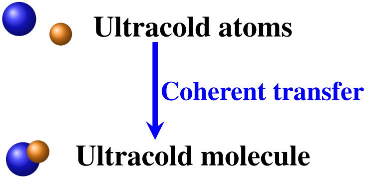
Ultracold molecule in tweezers

Direct cooling



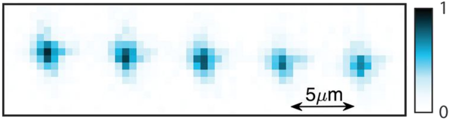
Science 365, 1156 (2019)

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

Ultracold atoms

Coherent transfer

Ultracold molecule

Challenges

- Temperature in tweezer
- Quantum control

- Control of atoms
- Coherent creation of molecules

Outline

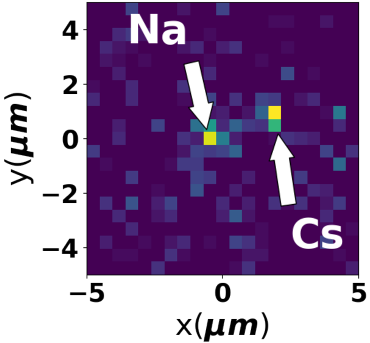
1. **Experiment overview**
2. **Atom state control**
   - Raman sideband cooling of Na atoms
3. **Molecule creation**
   - Atom-atom interaction
   - Coherent optical transfer
4. **Conclusion**

NaCs molecule

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

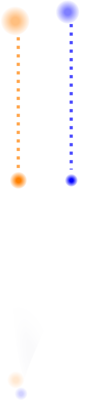
Experiment overview

Loading

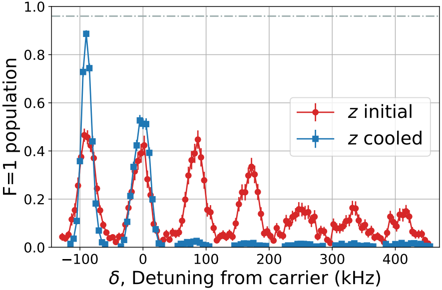


Loading probability per site: 60%
Post select on initial and final state.

Experiment overview



Cooling

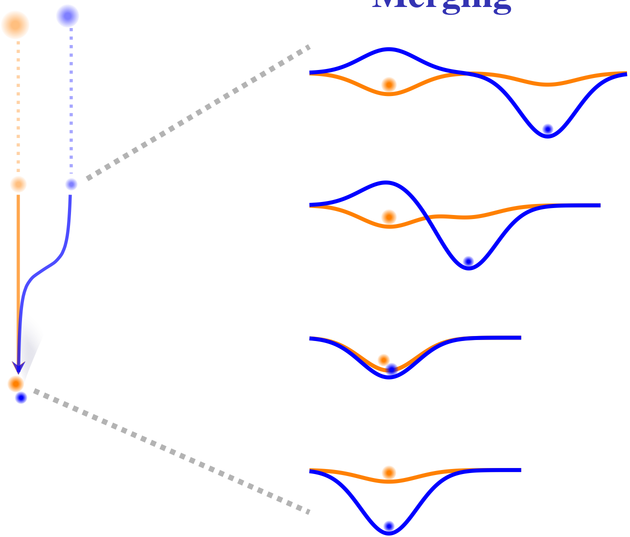


Cs: 96% ground state[1]
Na: 94% ground state[2]

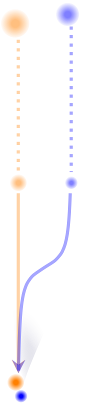
[1]**Y. Yu** et al. PRX 9, 021039 (2019)
[2]**Y. Yu** et al. PRA 97, 063423 (2018)

Experiment overview

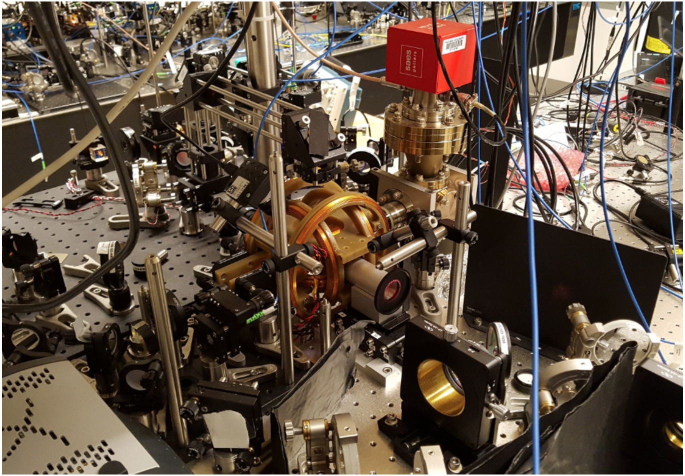
Merging



Experiment overview



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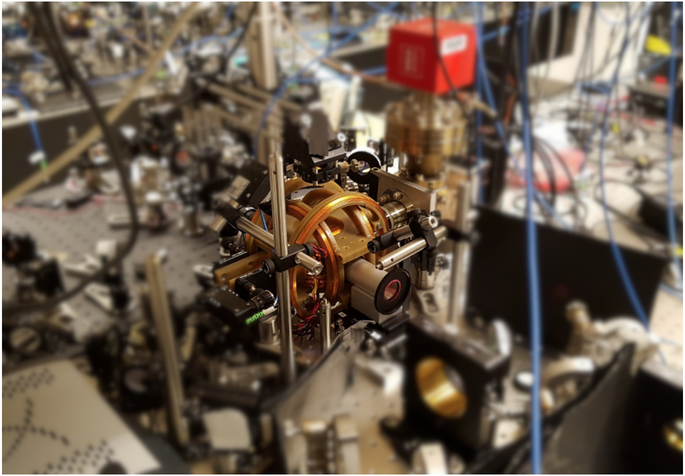
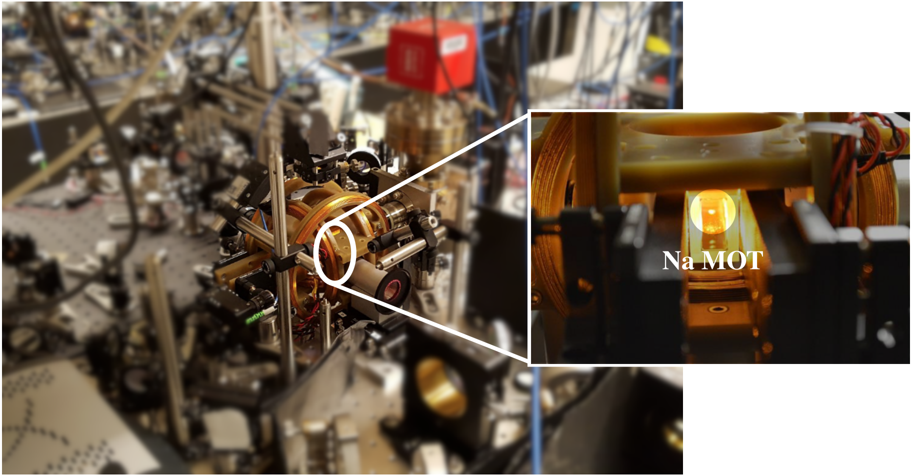
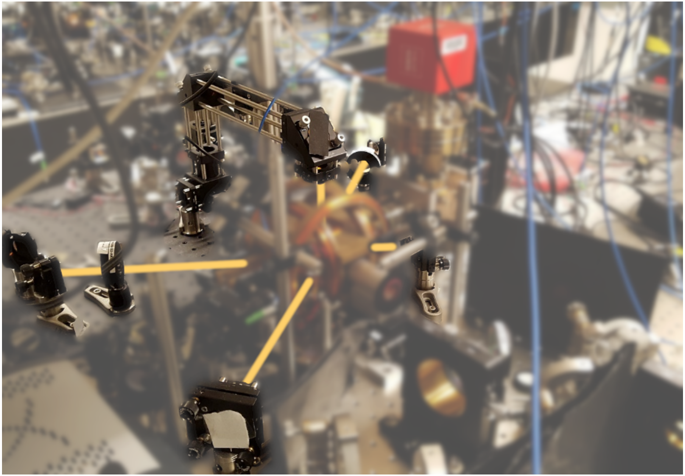
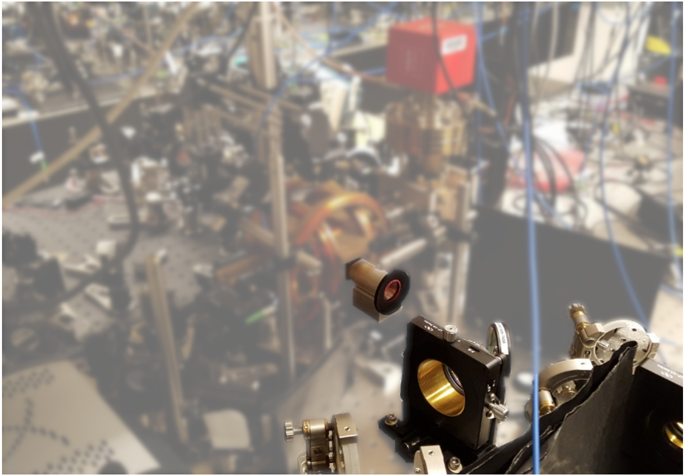



Na MOT

MOT beam path



Tweezer beam path



Outline

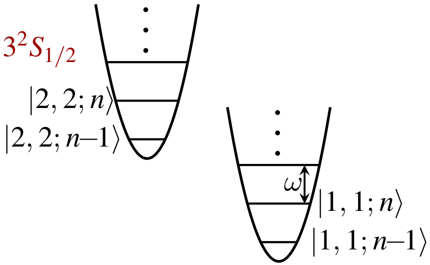
1. Experiment overview

2. **Atom state control**
   - Raman sideband cooling of Na atoms

3. Molecule creation
   - Atom-atom interaction
   - Coherent optical transfer

4. Conclusion

Raman sideband cooling



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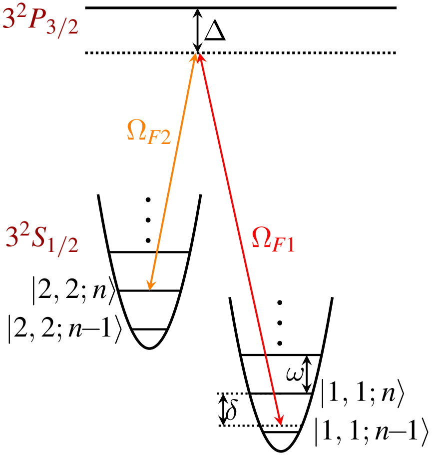

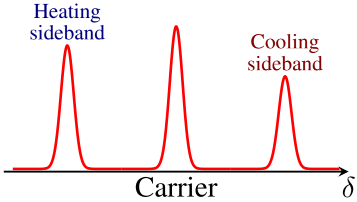

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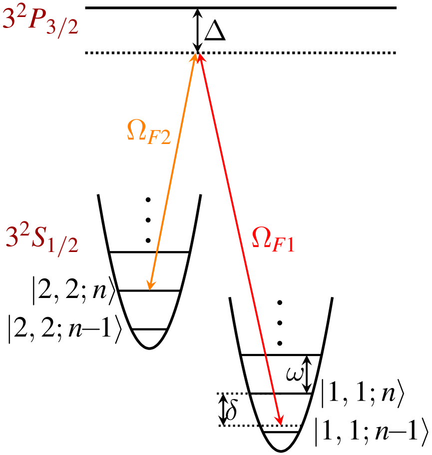

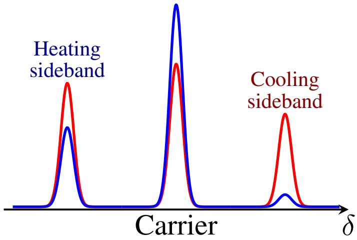

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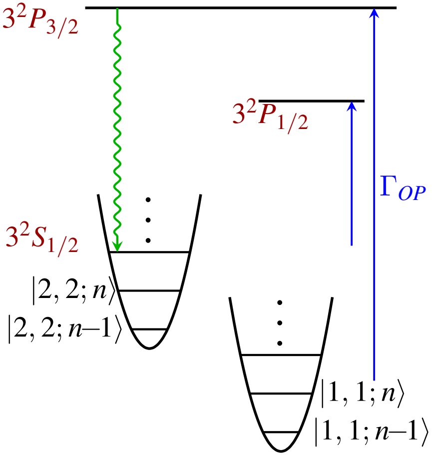

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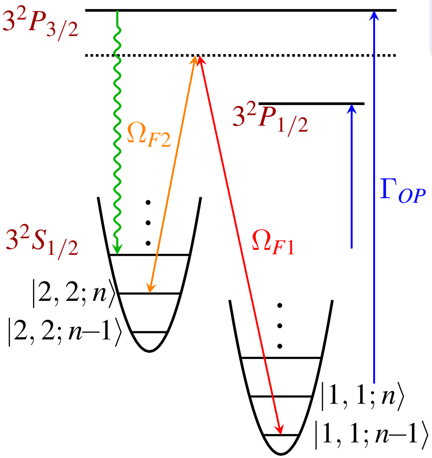
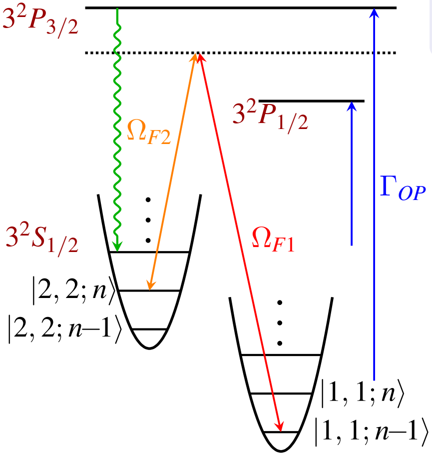


Lamb Dicke parameter

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

Raman sideband cooling



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

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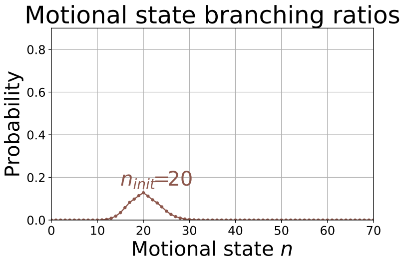

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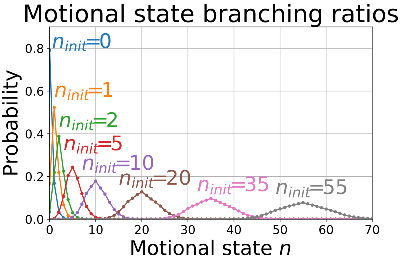

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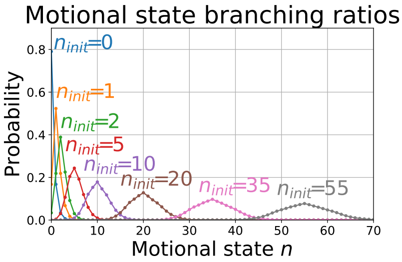

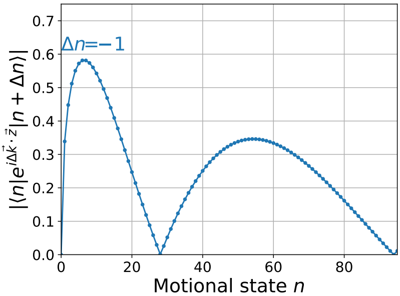

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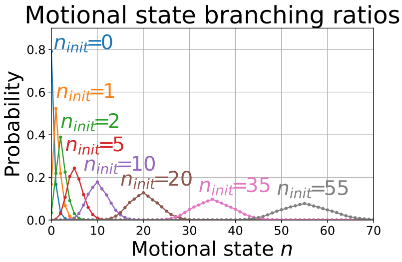

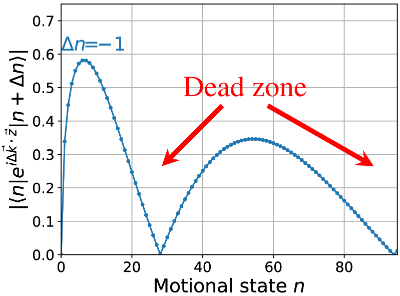

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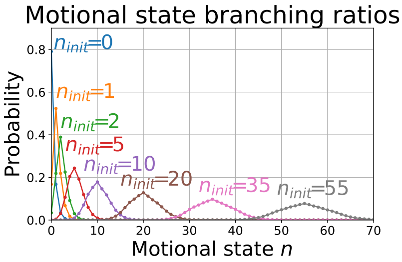

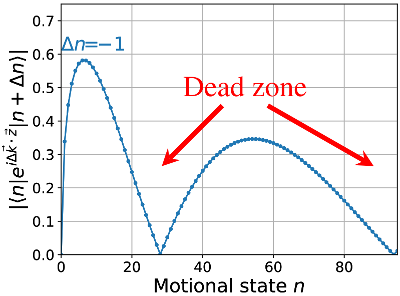

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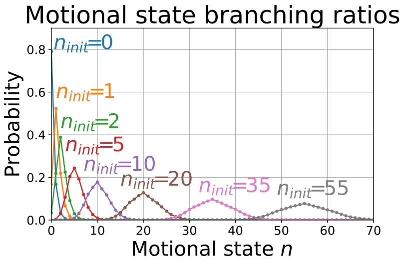
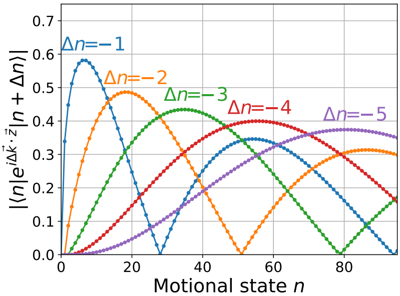
Solution

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

Raman sideband cooling





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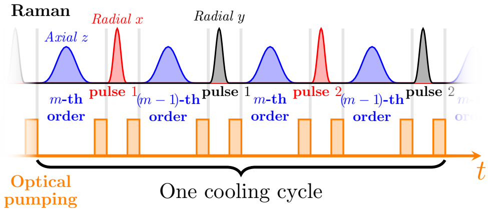

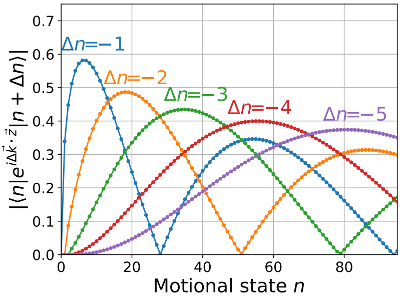

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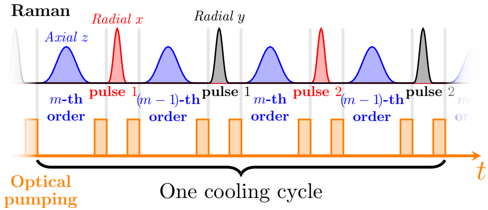

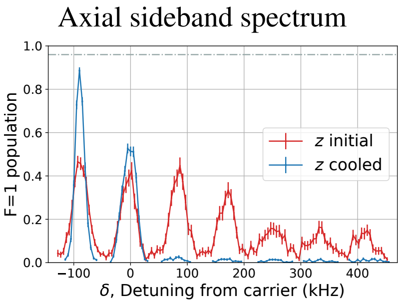

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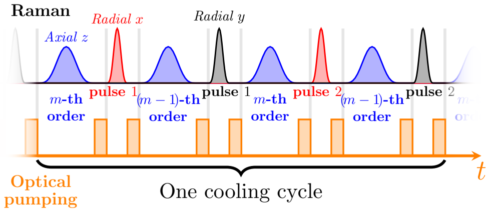

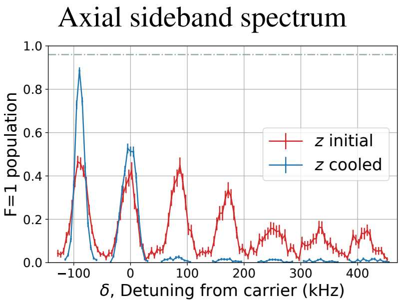

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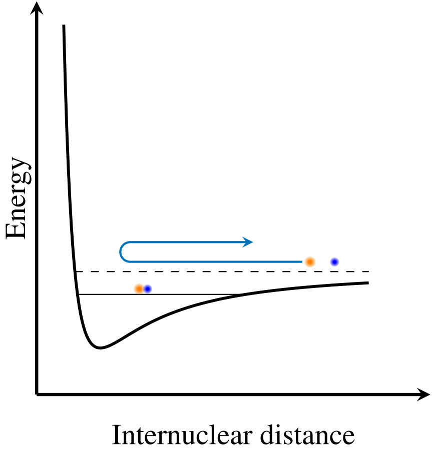
3D ground state: $93.5(7)\%$

Outline

Scattering length a

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

$\vdots$



Scattering length a

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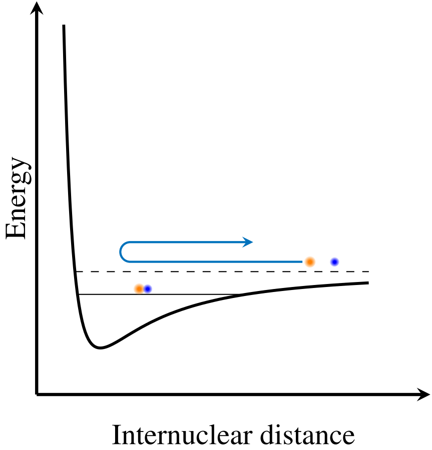
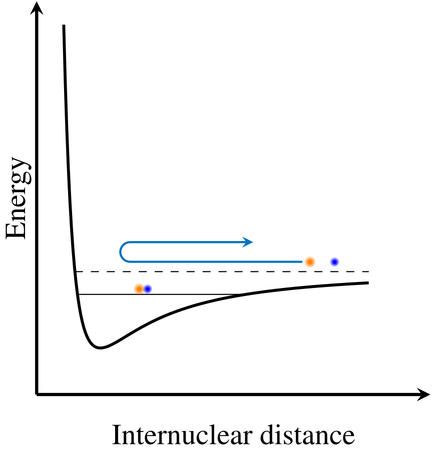
- Feshbach resonance
- ⋮



Scattering length a

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

⋮



Scattering length a

- Binding energy
- Molecular potential
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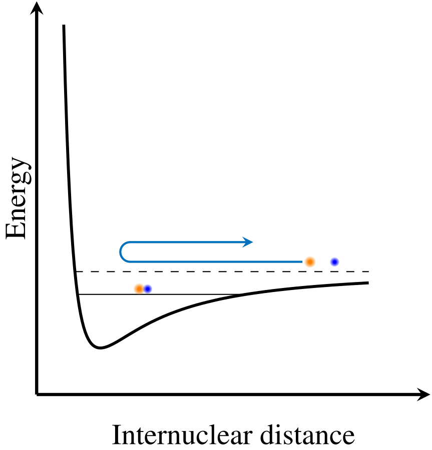
- Feshbach resonance

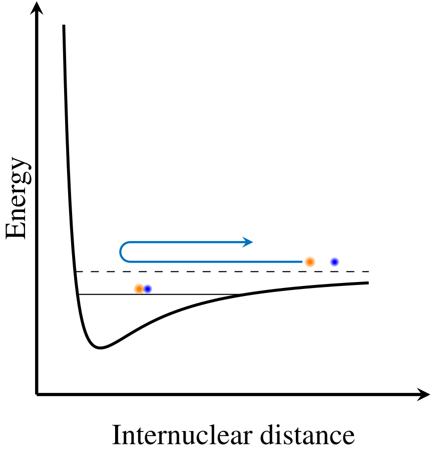
⋮



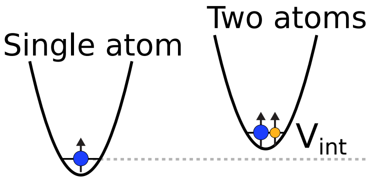
Scattering length a

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

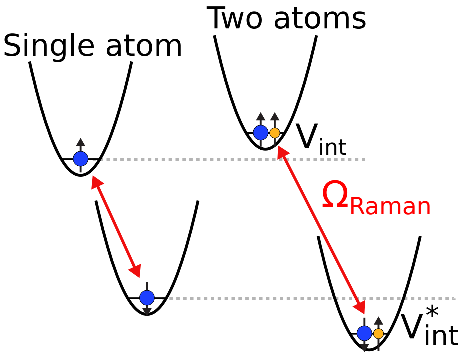
$\vdots$



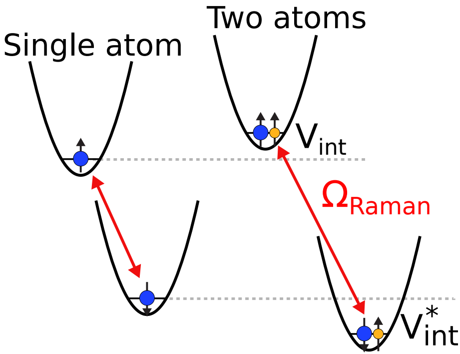
Interaction shift

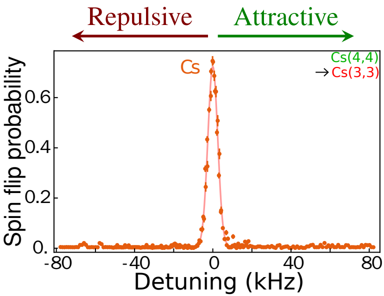


Interaction shift

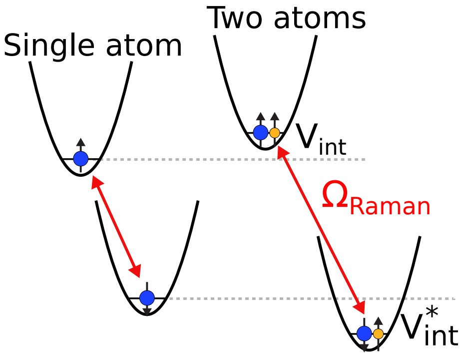


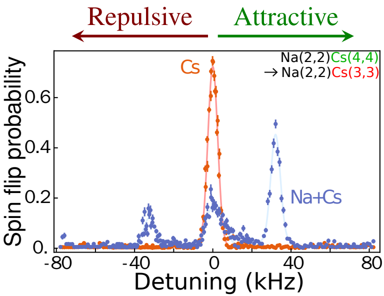
Interaction shift





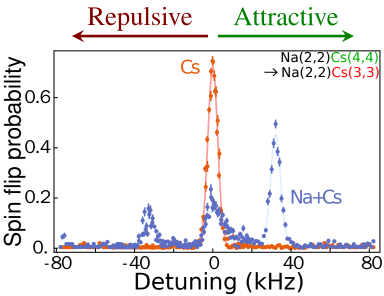
Interaction shift





Interaction shift

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



Interaction shift

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

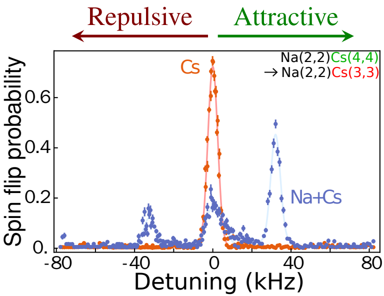
To center of mass
and relative coordinates

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

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

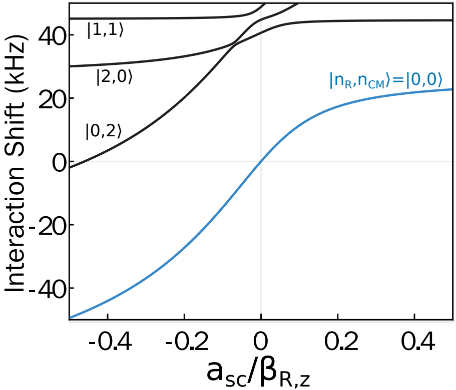
$$X_i = \frac{m_1 x_{1,i} + m_2 x_{2,i}}{m_1 + m_2} \qquad x_{R,i} = x_{1,i} - x_{2,i}$$

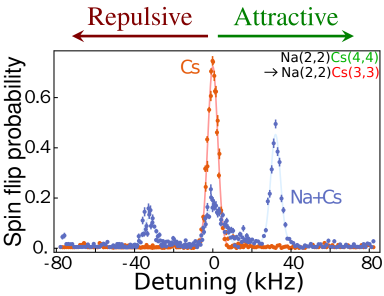
$$P_i = p_{1,i} + p_{2,i} \qquad p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$



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

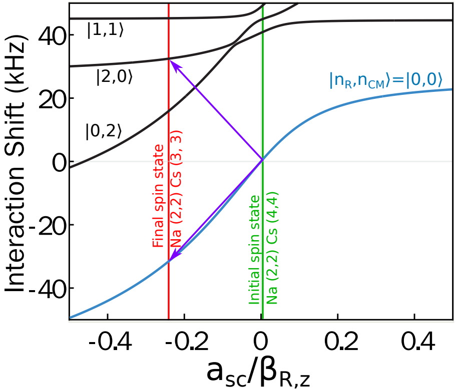
Interaction shift

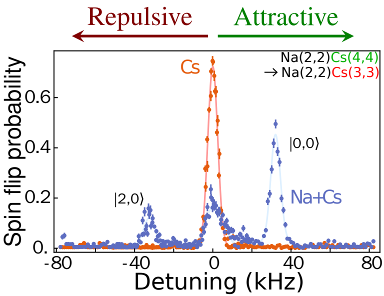




$$H = \overbrace{\sum_{i=x,y,z}\left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M}\right)}^{\text{Center of mass}} + \overbrace{\sum_{i=x,y,z}\left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu}\right) + V_{int}(\vec{r}_R)}^{\text{Relative}} + \overbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

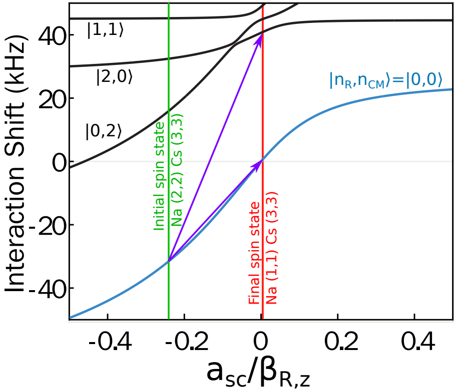
Interaction shift

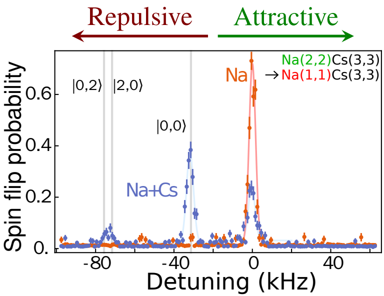




$$H = \overbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M}\right)}^{\text{Center of mass}} + \overbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu}\right) + V_{int}(\vec{r}_R)}^{\text{Relative}} + \overbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



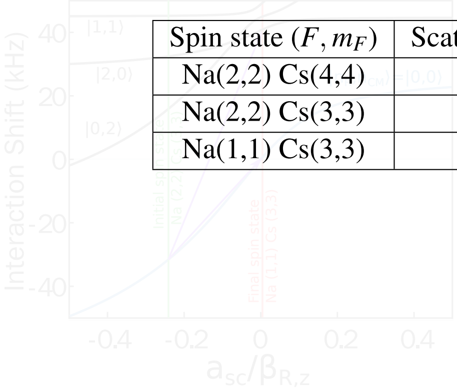


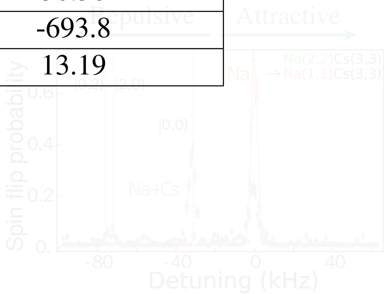
$$H = \overbrace{\sum_{i=x,y,z}\left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M}\right)}^{\text{Center of mass}} + \overbrace{\sum_{i=x,y,z}\left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu}\right) + V_{int}(\vec{r}_R)}^{\text{Relative}} + \overbrace{\sum_{i=x,y,z}\mu(\omega_{1,i}^2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

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





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

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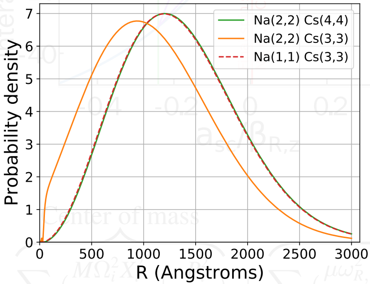
Enhanced coupling to molecular state!



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
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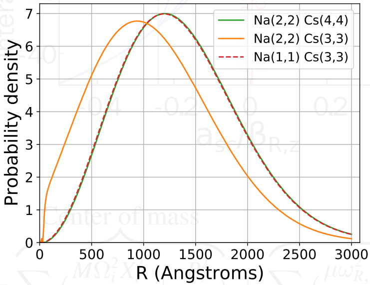
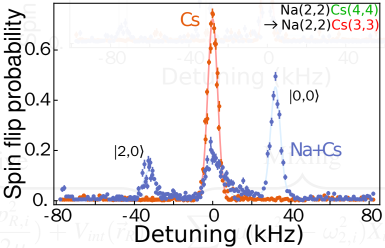
Enhanced coupling to molecular state!





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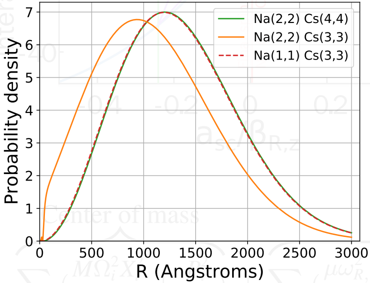
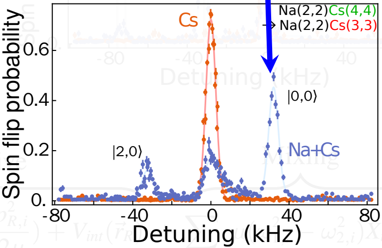
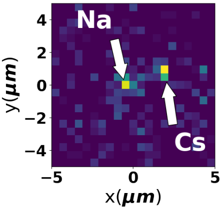


Ground state only



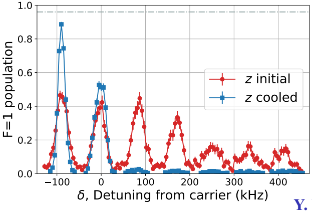
Outline

Loading



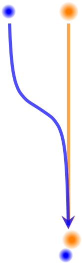
NJP. 19, 023007 (2017)

Cooling



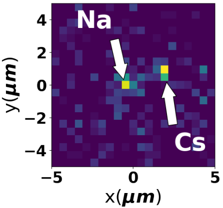
Y. Yu et al. PRA. 97, 063423 (2018)

Merging



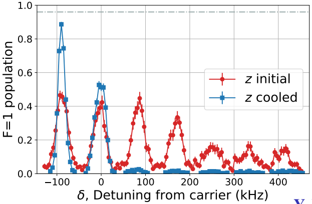
Y. Yu et al. PRX. 9, 021039 (2019)

Loading



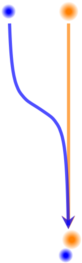
NJP. 19, 023007 (2017)

Cooling

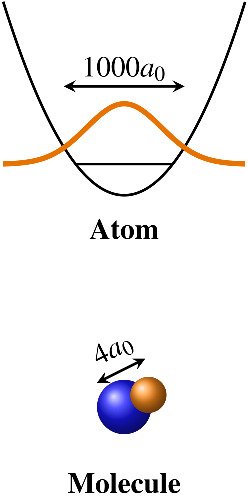


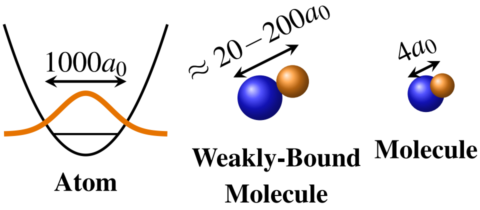
Y. Yu et al. PRA. 97, 063423 (2018)

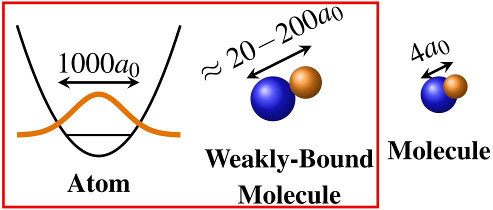
Merging

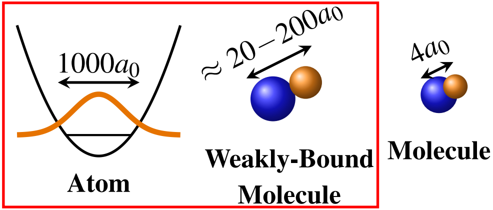


Y. Yu et al. PRX. 9, 021039 (2019)

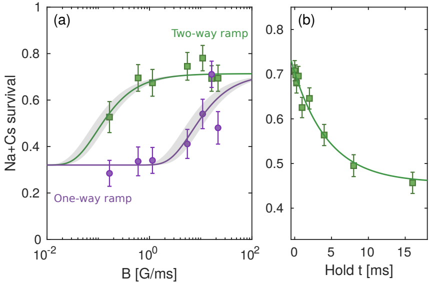



$1000a_0$
Atom
$\approx 20-200a_0$
Weakly-Bound Molecule
$4a_0$
Molecule

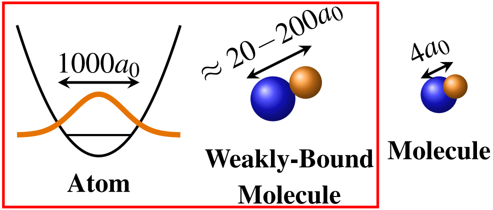

$1000a_0$
Atom
$\approx 20-200a_0$
Weakly-Bound Molecule
$4a_0$
Molecule



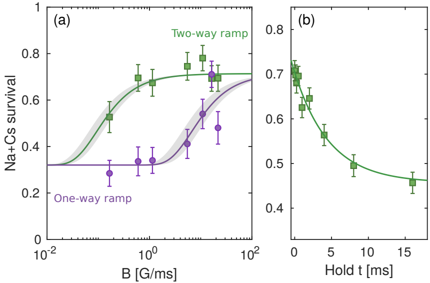
Feshbach molecule



PRL. 124, 253401 (2020)

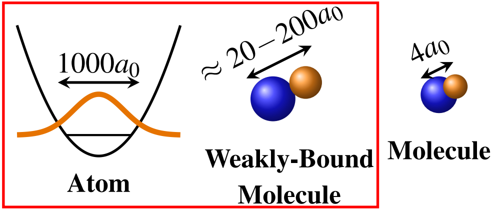


Feshbach molecule



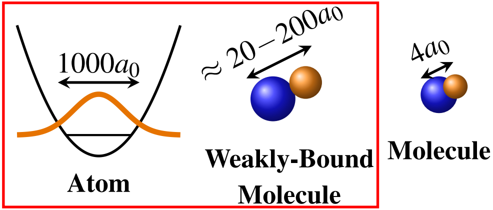
PRL. 124, 253401 (2020)

- Requires Feshbach resonance
- Usually large magnetic field



Optical transfer

- More general
- Faster



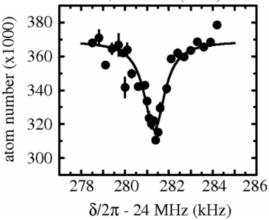
Optical transfer

- More general
- Faster

Previous results

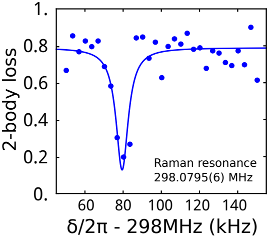
Rb_2 Science 287, 1016 (2000)

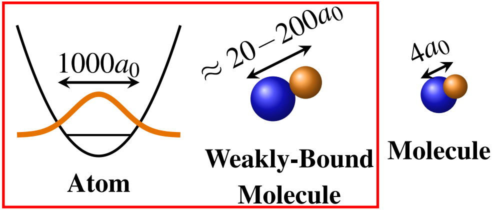
PRL. 93, 073002 (2004)



Sr_2 PRL. 109, 115302 (2012)

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





Optical transfer

- More general
- Faster

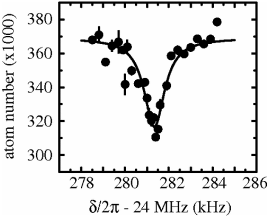
Limitations so far

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

Previous results

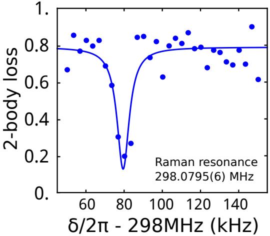
Rb_2 Science 287, 1016 (2000)

PRL. 93, 073002 (2004)

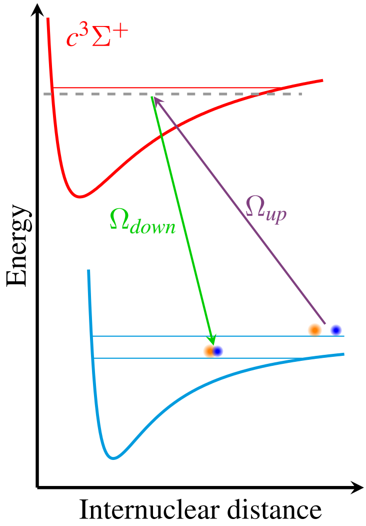


Sr_2 PRL. 109, 115302 (2012)

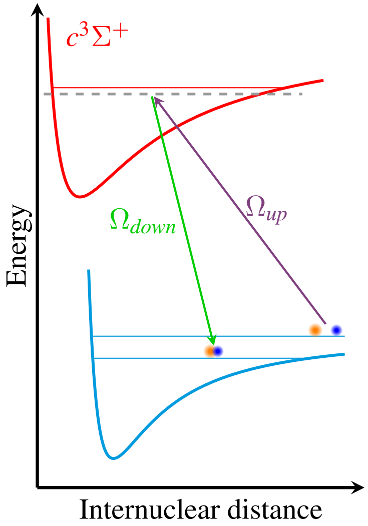
NaCs **Y. Yu** et al. PRX. 9, 021039 (2019)



Raman transfer



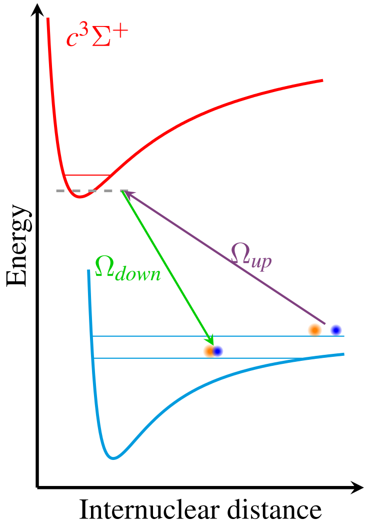
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

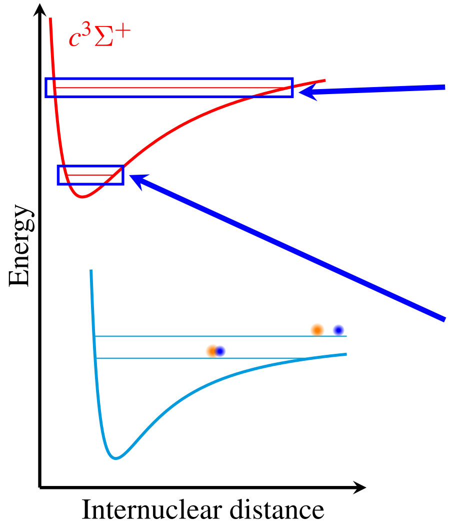
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

Deeply bound states

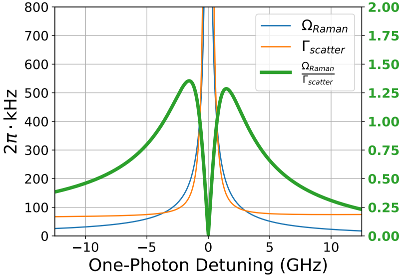
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

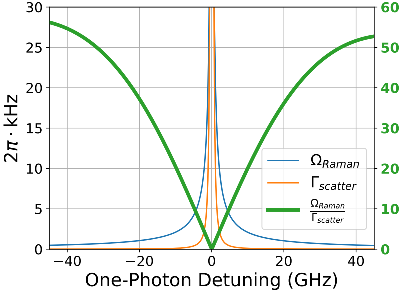
Raman transfer



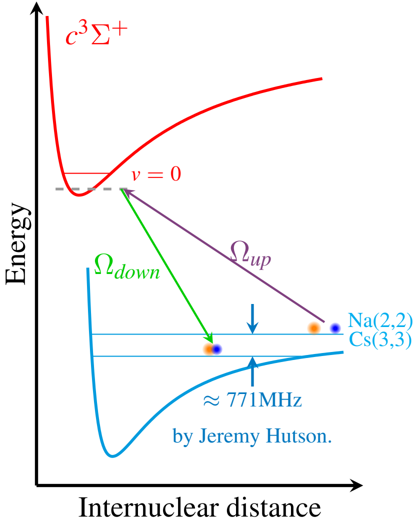
Near threshold states



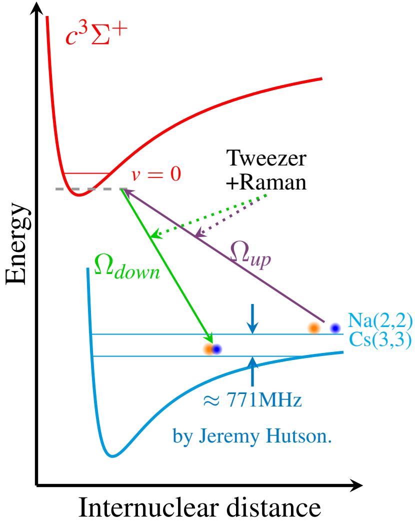
Deeply bound states



Experiment


$c^3\Sigma^+$
$v = 0$
Ω_{up}
Ω_{down}
Energy
Na(2,2)
Cs(3,3)
$\approx$ 771MHz
by Jeremy Hutson.
Internuclear distance

Experiment



Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

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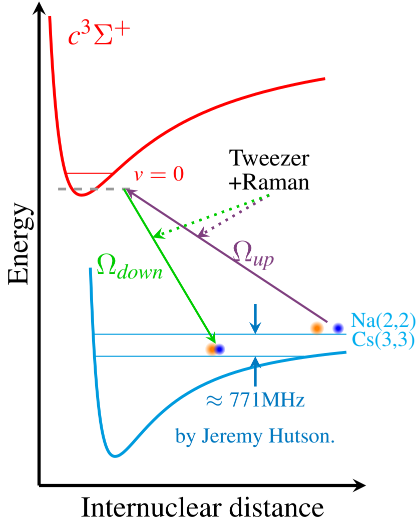

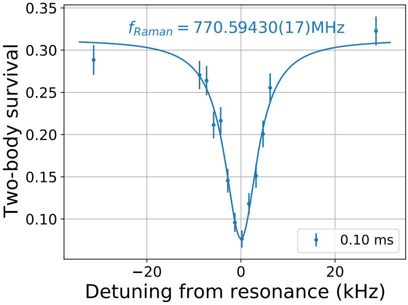

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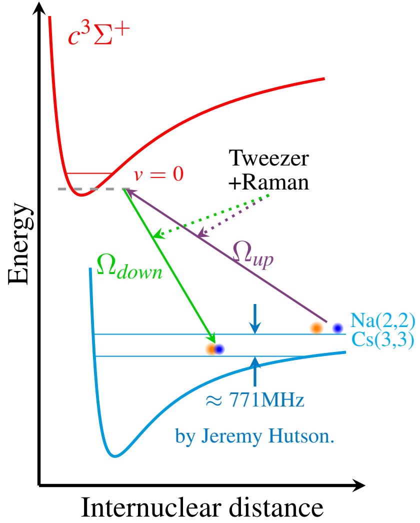

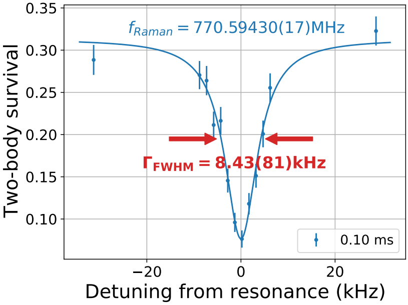

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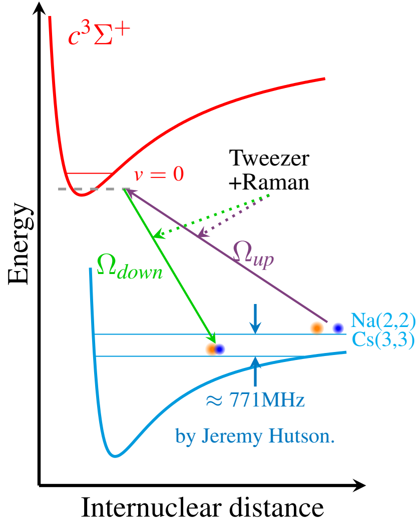
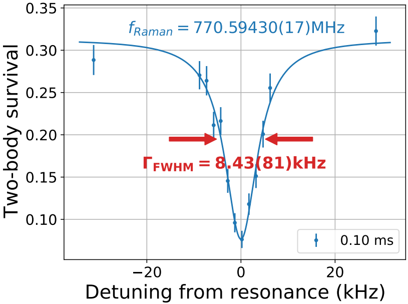
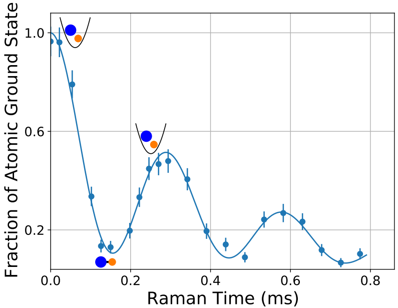


$c^3\Sigma^+$
$v = 0$
Tweezer
+Raman
Ω_{up}
Ω_{down}
Energy
Na(2,2)
Cs(3,3)
≈ 771MHz
by Jeremy Hutson.
Internuclear distance



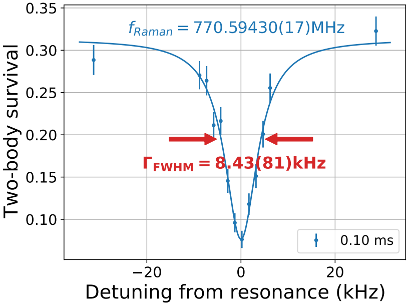
f_{Raman} = 770.59430(17)MHz
Γ_{FWHM} = 8.43(81)kHz
0.10 ms
Two-body survival
0.35
0.30
0.25
0.20
0.15
0.10
−20
0
20
Detuning from resonance (kHz)

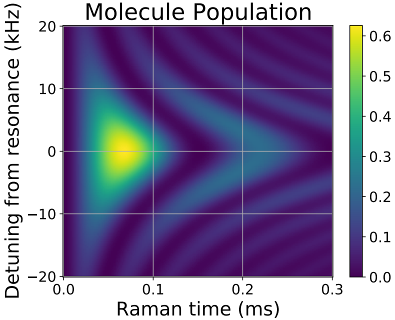


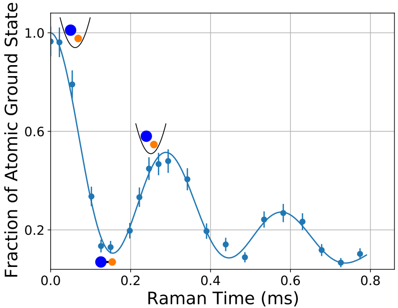
Fraction of Atomic Ground State
1.0
0.6
0.2
0.0
0.2
0.4
0.6
0.8
Raman Time (ms)

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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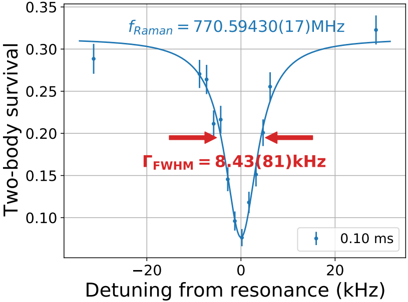

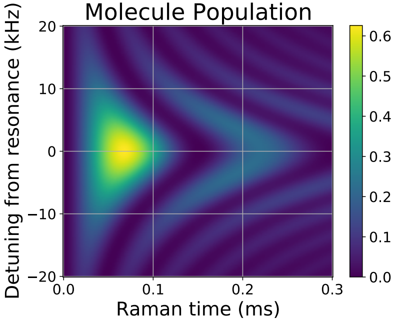

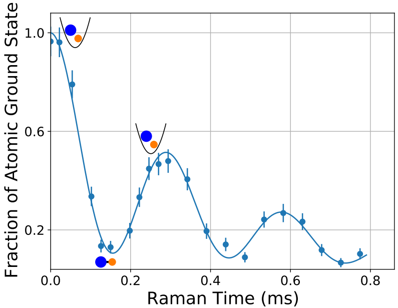

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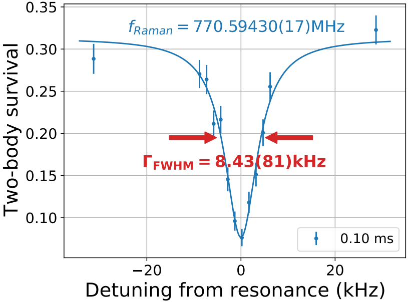

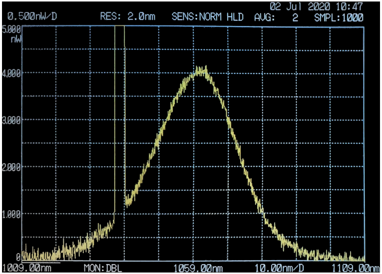

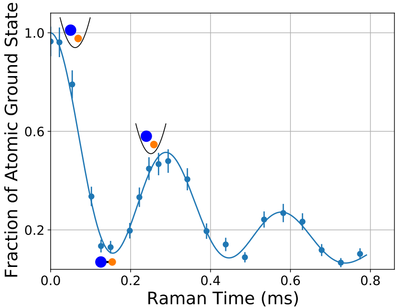

Conclusion and outlook

- New quatum 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 molecucle
- Working towards fully controlled, strongly interacting tweezer array

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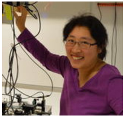
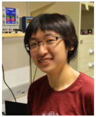
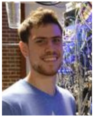


Kang-Kuen Ni

Kenneth Wang



Jessie Zhang

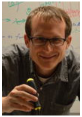


Lewis Picard

William Cairncross

Lee Liu
Postdoc @JILA

Jonathan Hood
Assnt Prof @Purdue



Nick Hutzler
Assnt 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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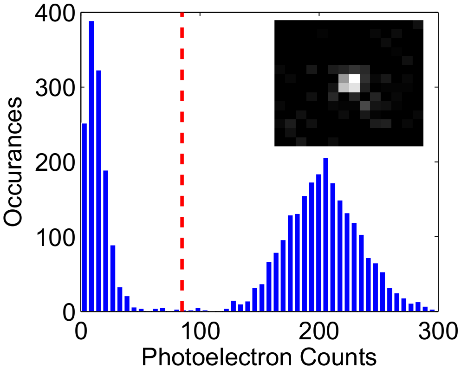

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Single Atom in Tweezer

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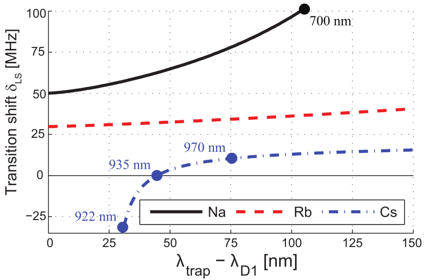
Issues with Na

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

Real Issue with Na: Light Shift



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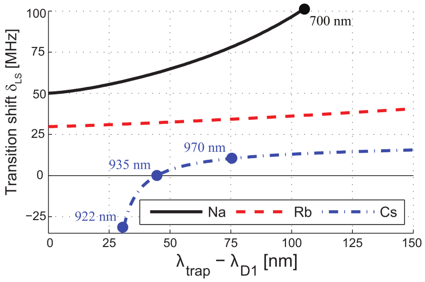
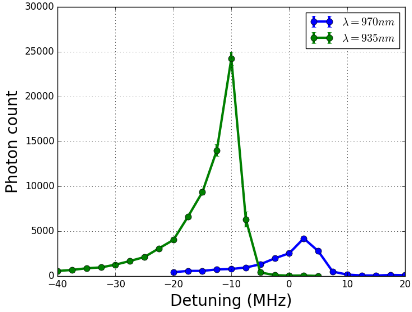


Cs single atom imaging



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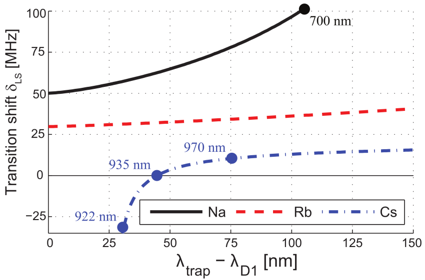

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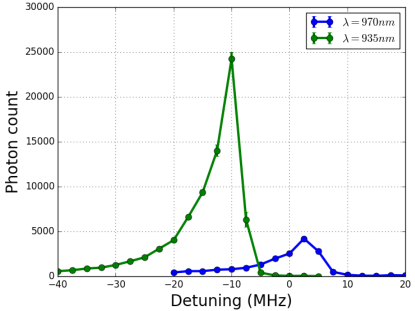


- Low imaging signal
- No cooling in tweezer

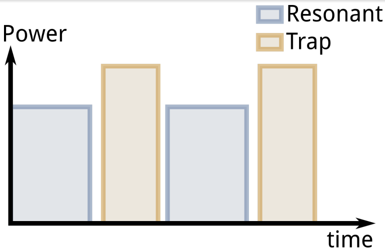
Real Issue with Na: Light Shift

Trap modulation

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

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

$\Gamma = 2\pi \times 10$ MHz



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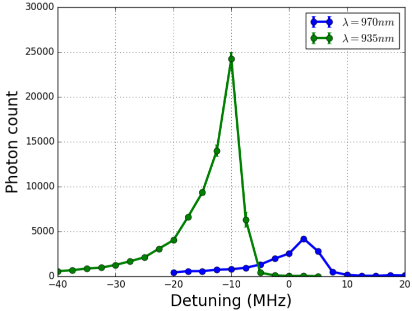

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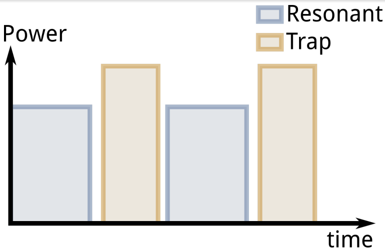

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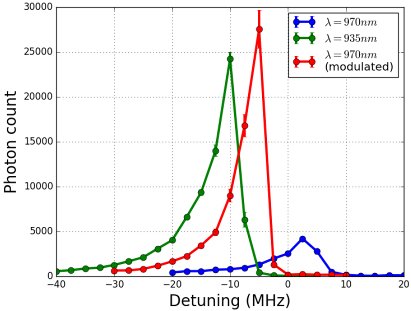

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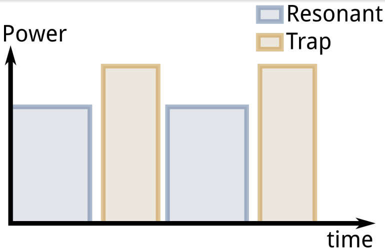

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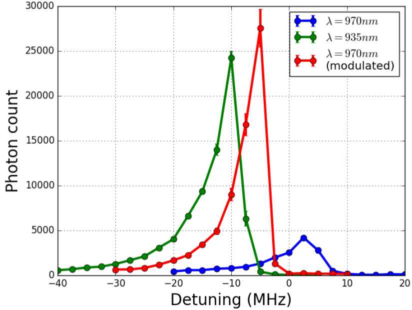
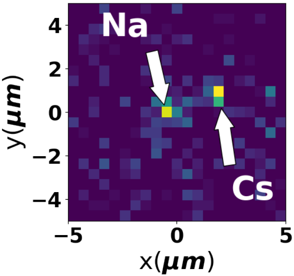
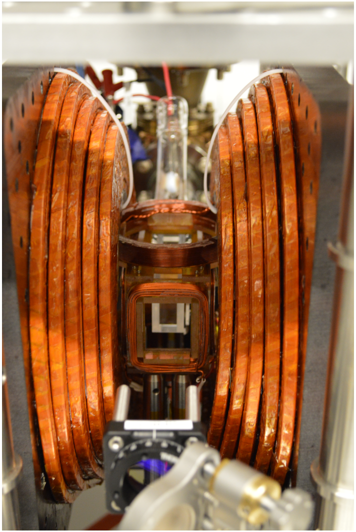




Na (1, -1) Cs (3, -3) Feshbach resonance



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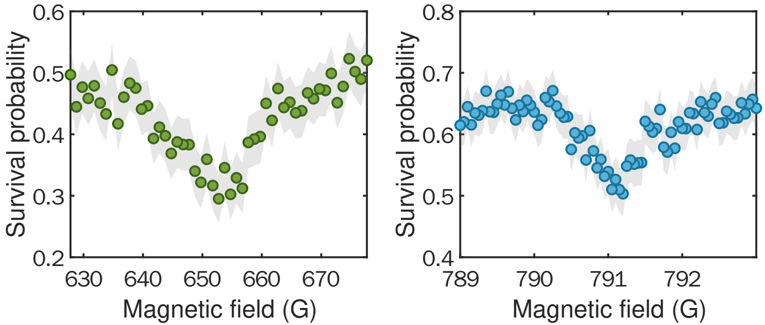

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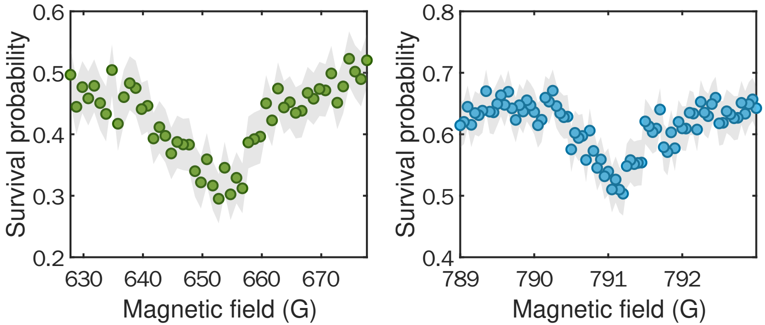
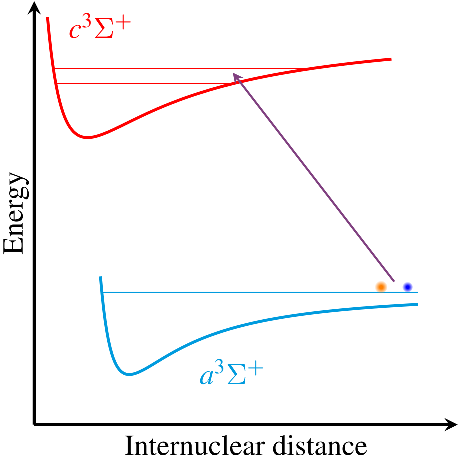


	s-wave	*p*-wave
Predicted (based on interaction shift)[1]	663 G	799 G
Measured	652(3) G	791.2(2) G

[1] In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



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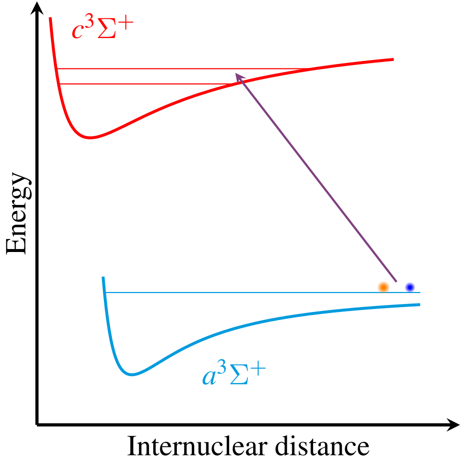
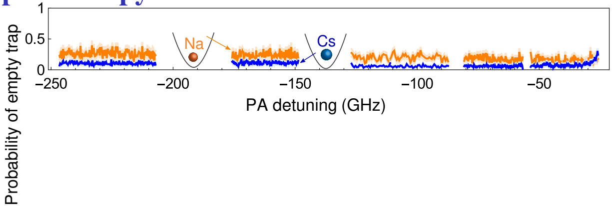
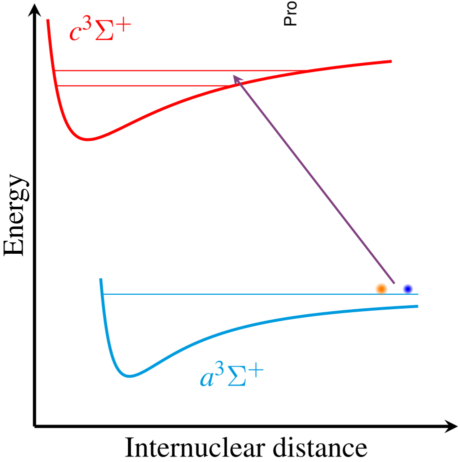


Single Atom PA

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

Photoassociation (PA) Spectroscopy





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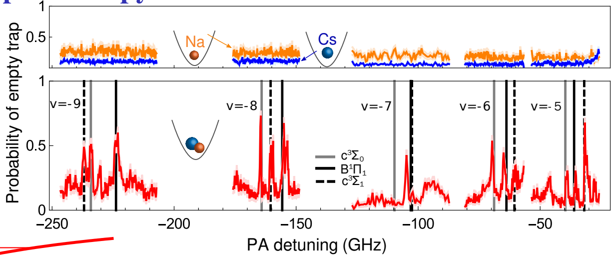

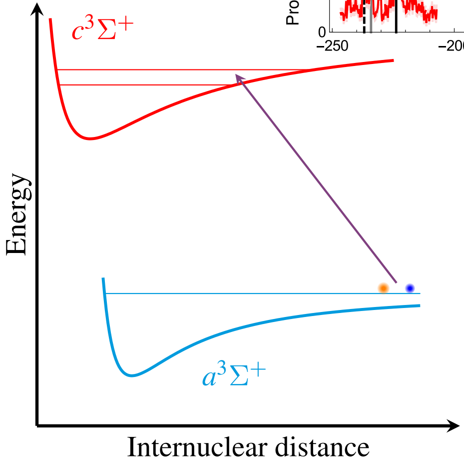

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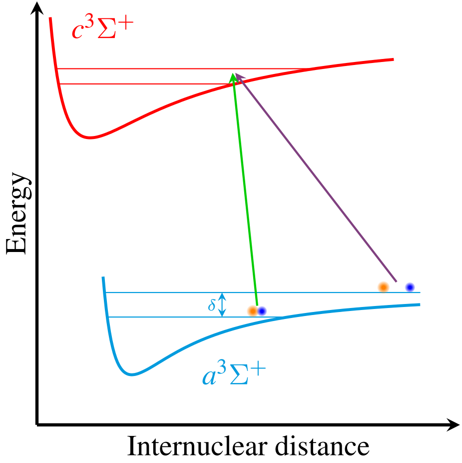
Electromagnetically Induced Transparency (EIT) Spectroscopy



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