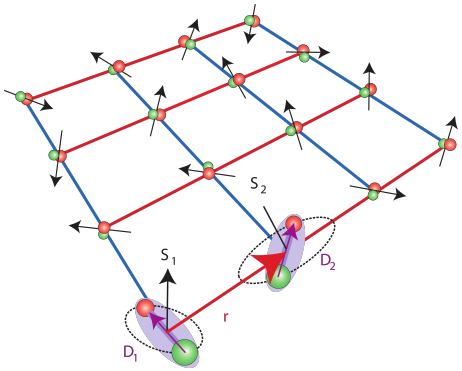
Building Single Molecules from Single Atoms

Yichao Yu

Ni Group/Harvard

Sep. 4, 2020

Quantum Simulation



Nat. Phys. 2, 341 (2006)

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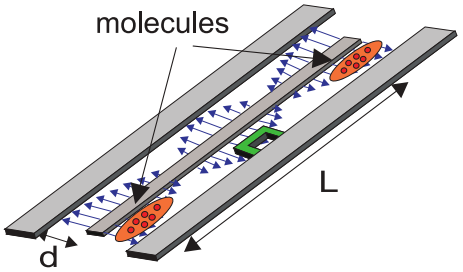
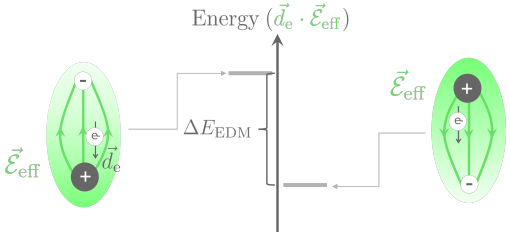


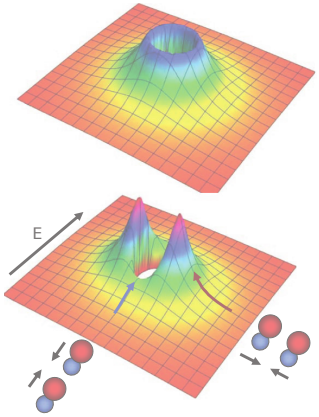
PRL. 97, 33003 (2006)

Precision Measurement



Science 343, 269 (2014)

Quantum Chemistry



Nature 464, 1324 (2010)

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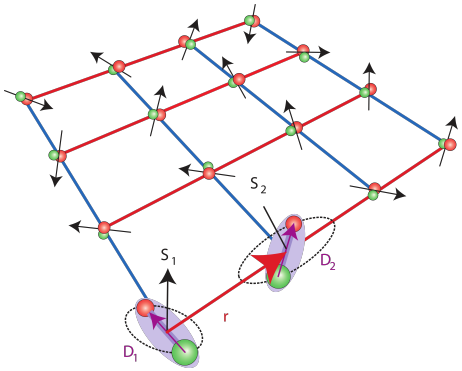

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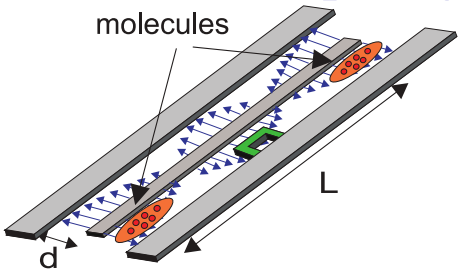

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- Full quantum control
- Entanglement
- ...

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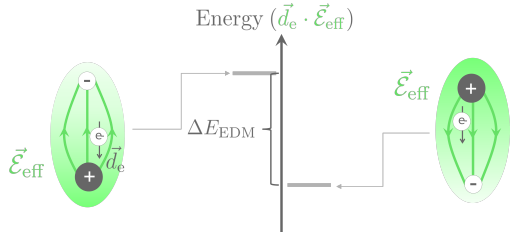

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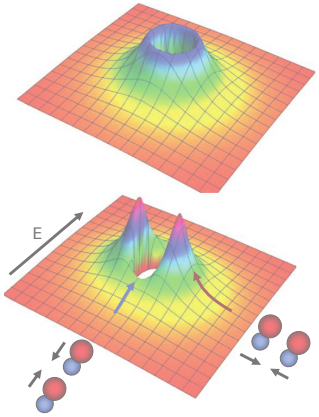

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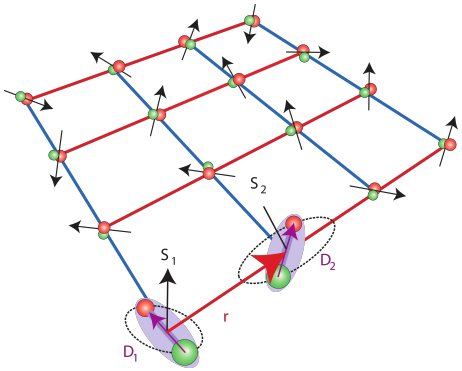

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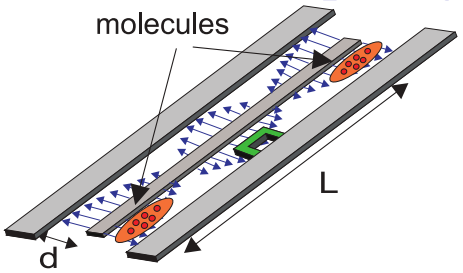

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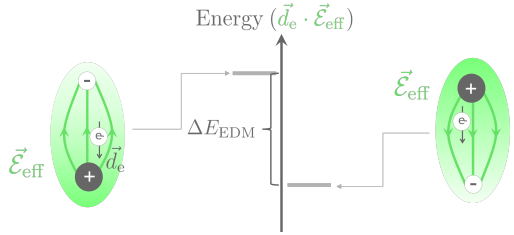

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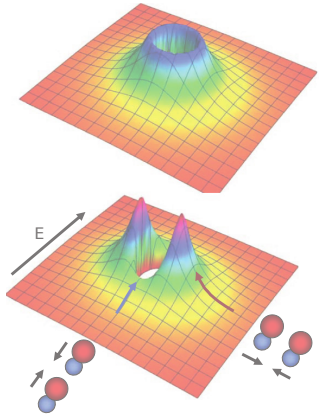

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New Approach?

Entanglement

Single particle control

Entanglement

i.e. interaction

Entanglement

i.e. interaction

Dipolar molecules

Entanglement

i.e. interaction

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.)

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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

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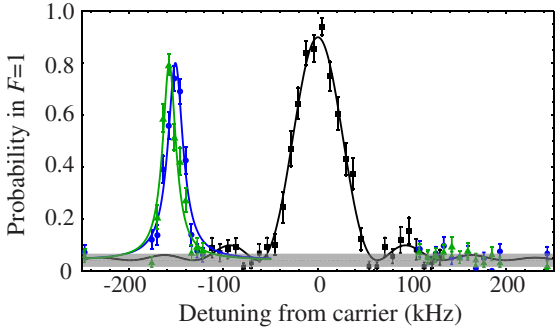
Cooling



PRX. 2, 041014 (2012)

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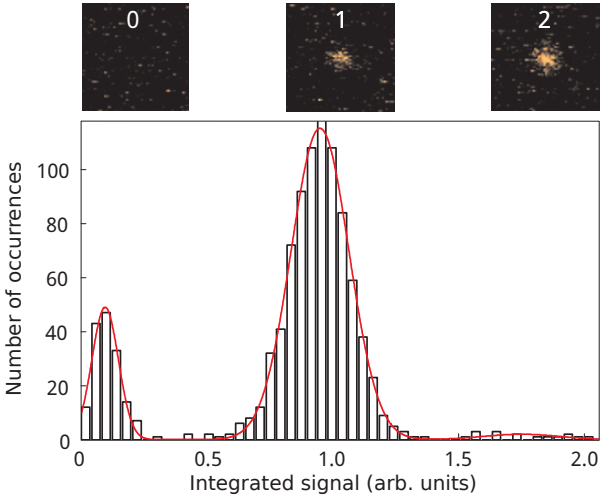
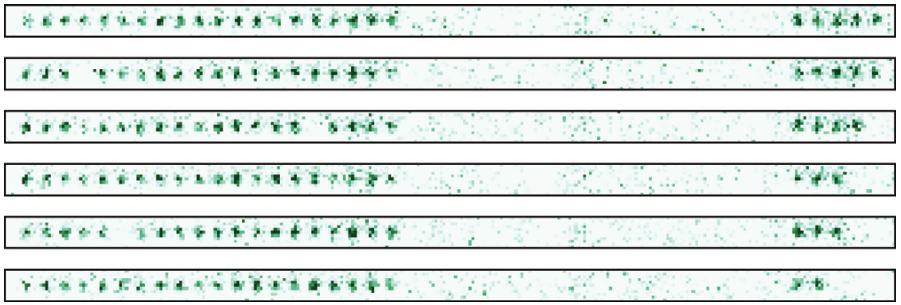


Nat. Phys. 6, 951 (2010)

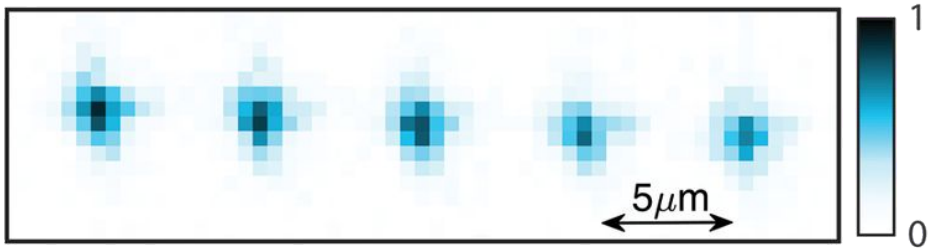
Rearranging



Science 354, 1024 (2016)

Ultracold molecule in tweezers

Direct cooling



Science 365, 1156 (2019)

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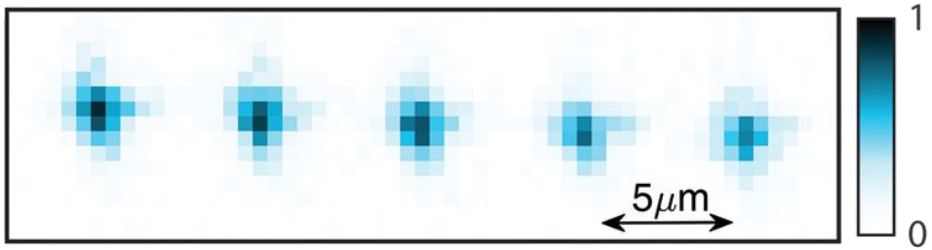
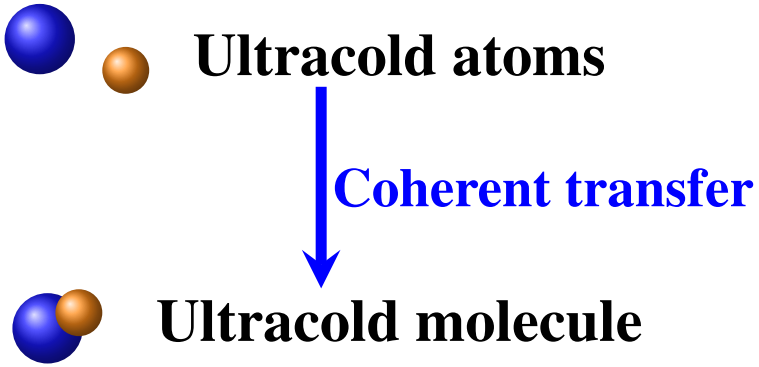


Science 365, 1156 (2019)

Assembly



Ultracold molecule in tweezers

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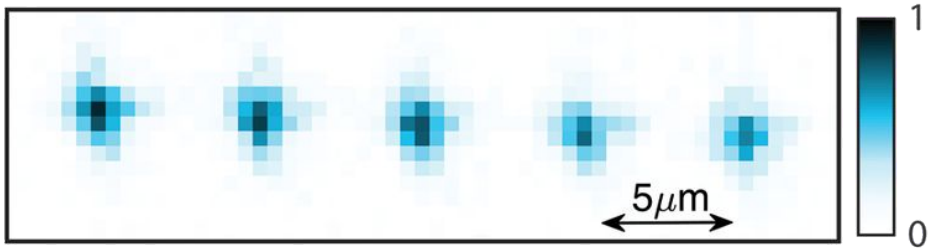


Science 365, 1156 (2019)

Assembly

Challenges

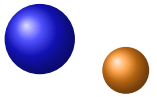
- Temperature in tweezer
- Quantum control

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

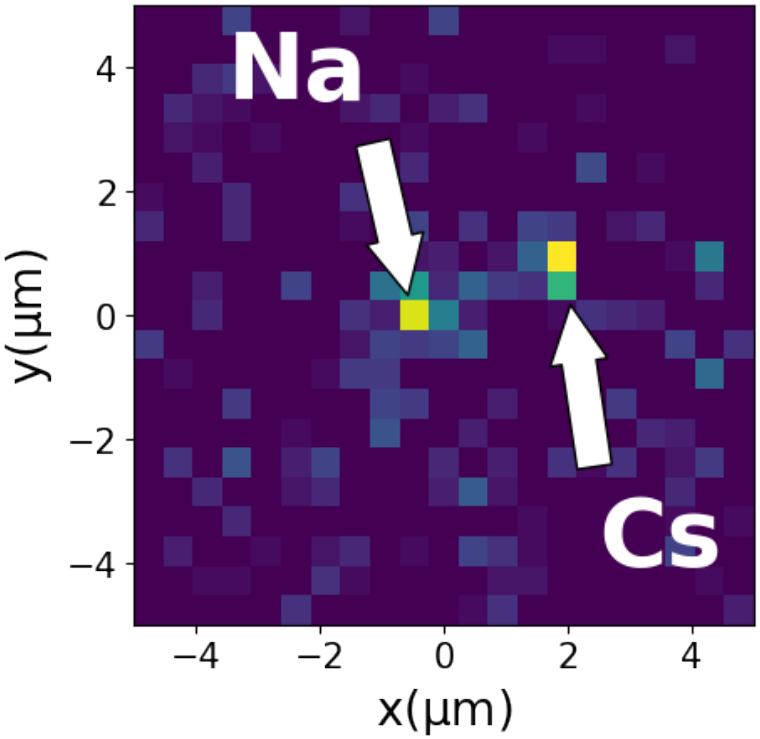
1. **Experiment overview**
2. **Atom state control**
   - Raman sideband cooling of Na atoms
3. **Molecule creation**
   - Atom-atom interaction
   - Coherent optical transfer
4. **Conclusion**

NaCs molecule

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

Experiment overview

Loading



Loading probability per site: 60%
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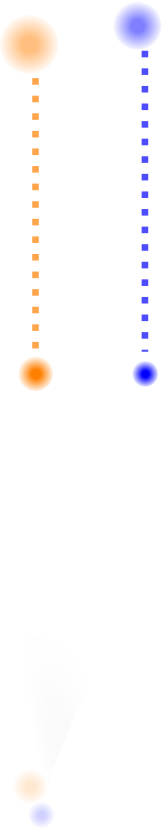
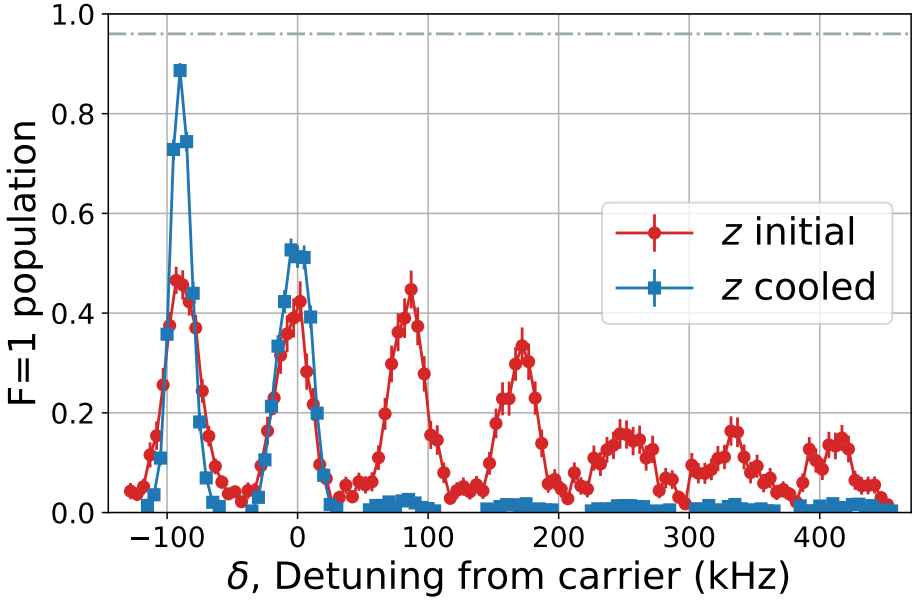
Experiment overview



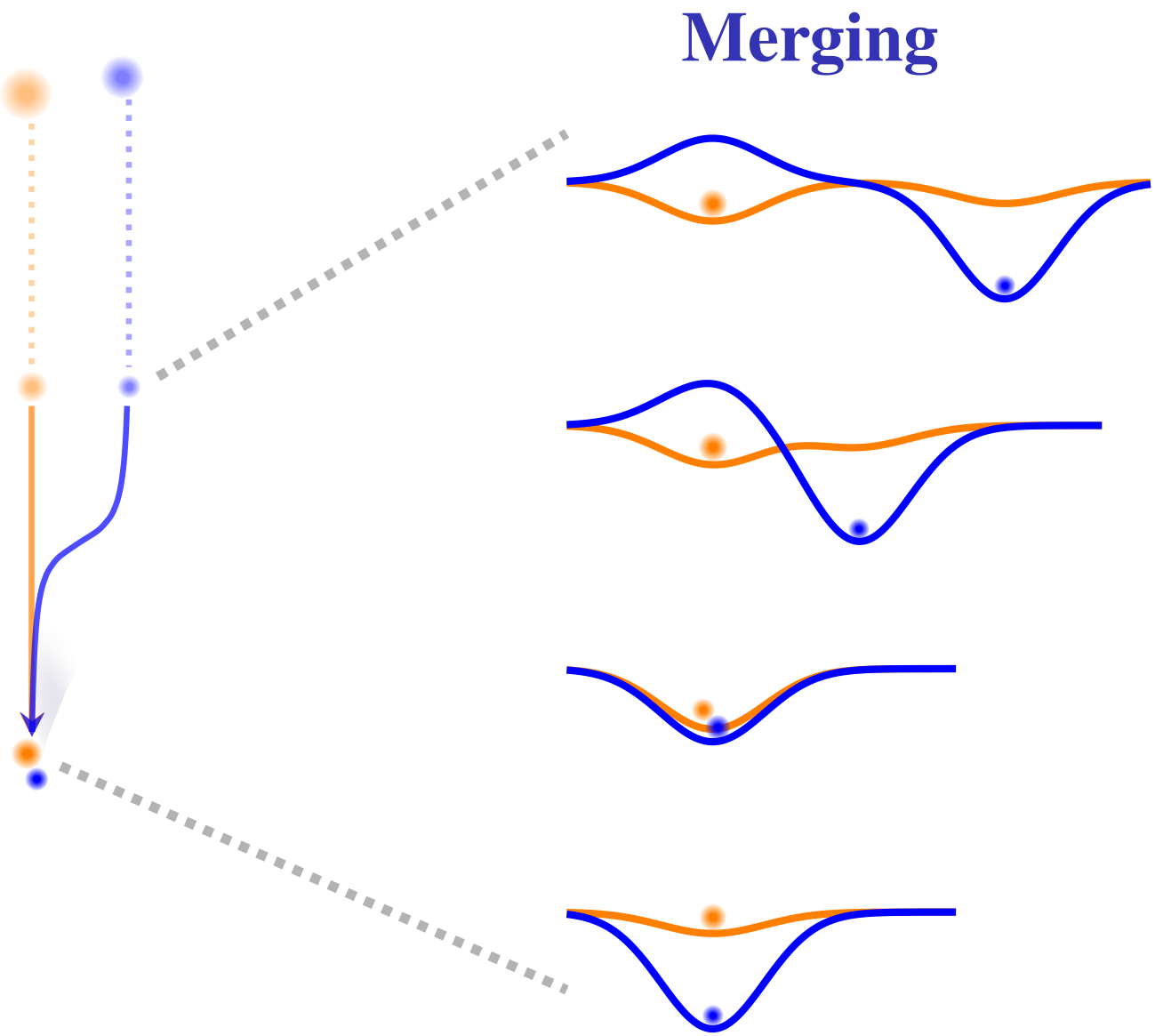
Cooling



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



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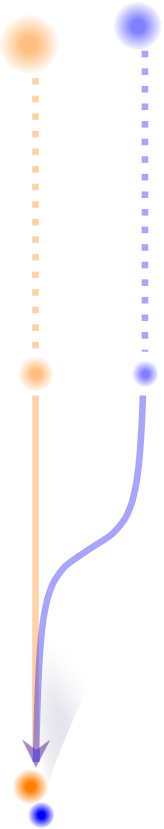
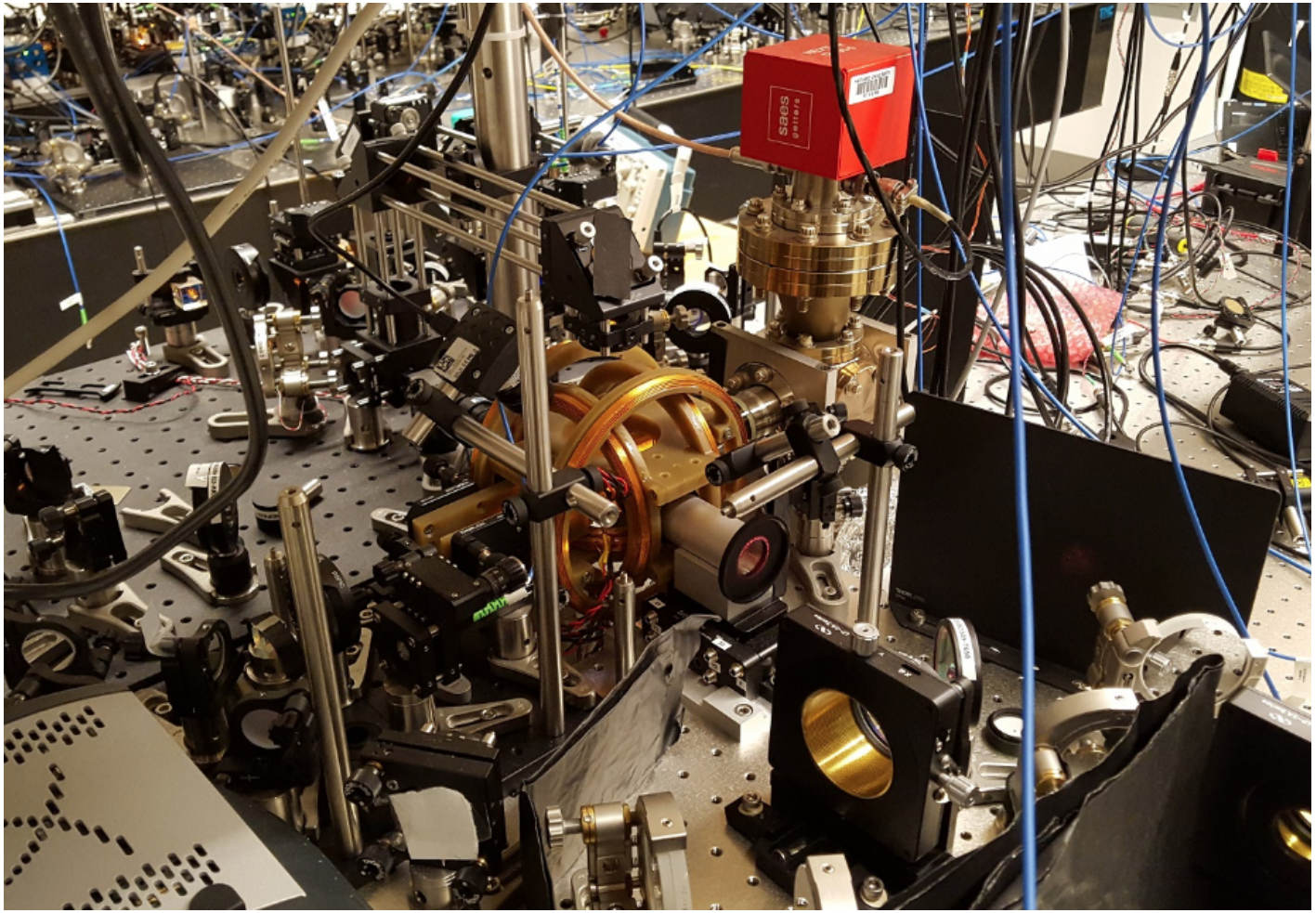


L. R. Liu, J. D. Hood, **Y. Yu** et al., Science 360, 6391 (2018)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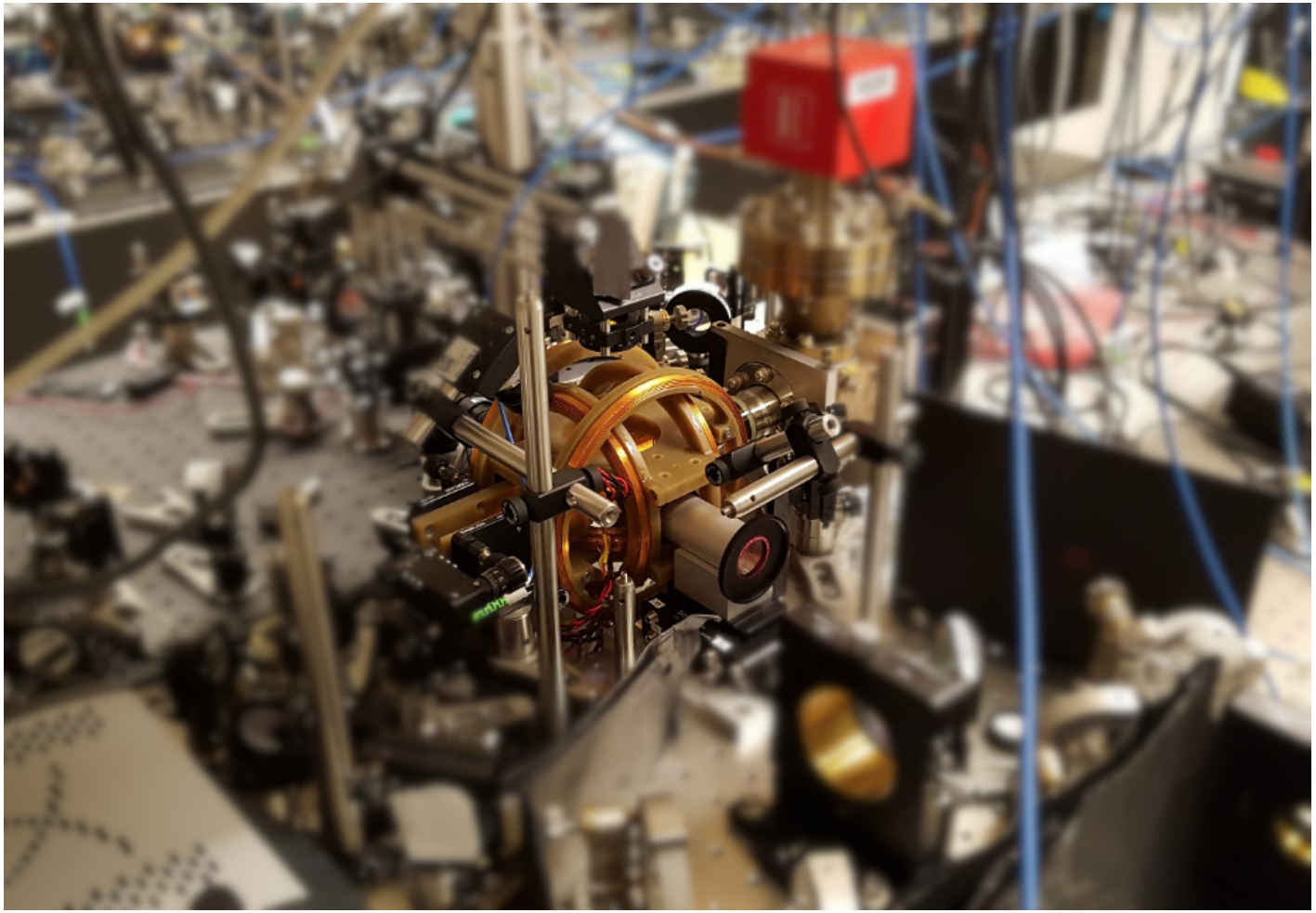
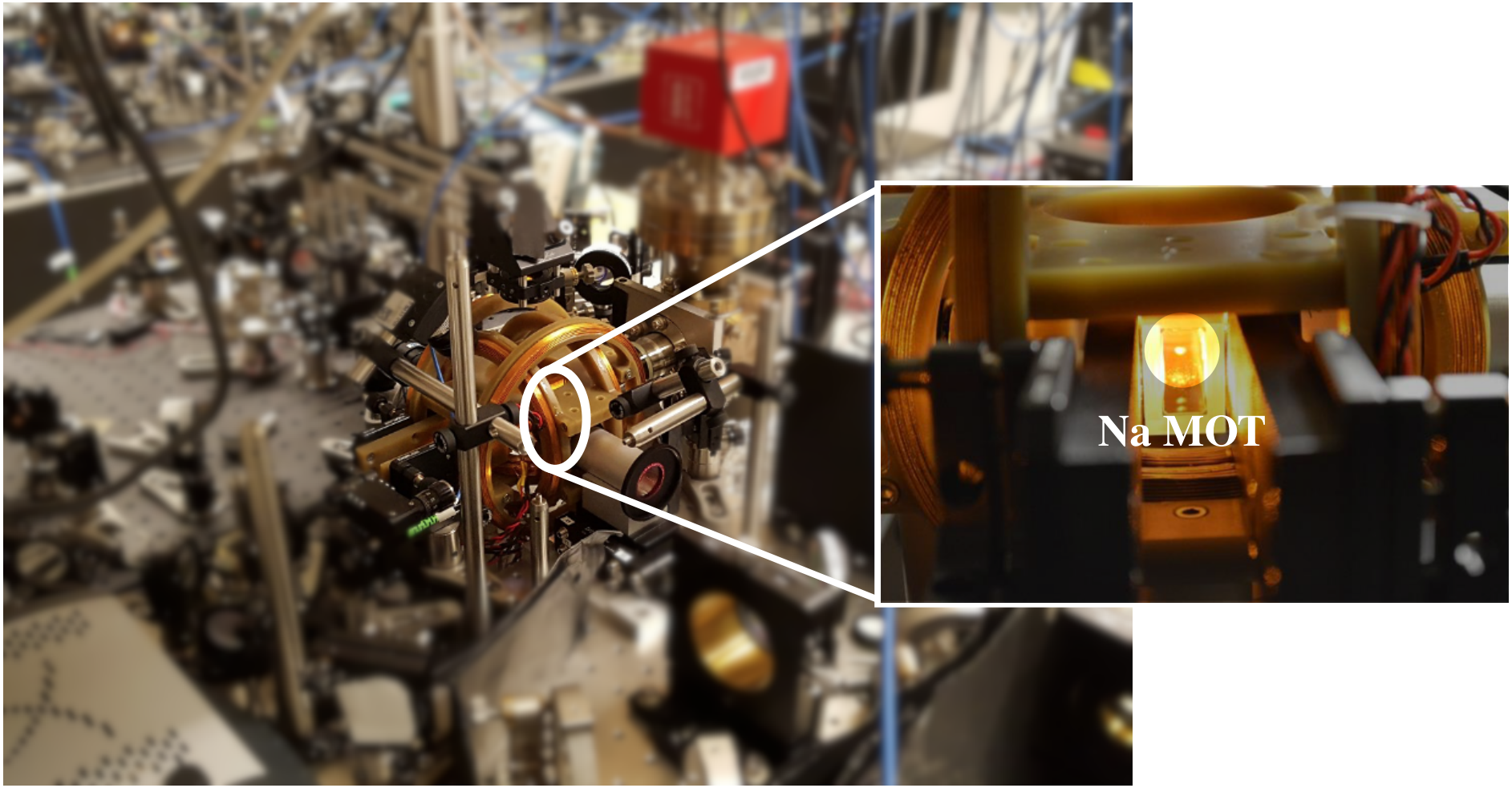
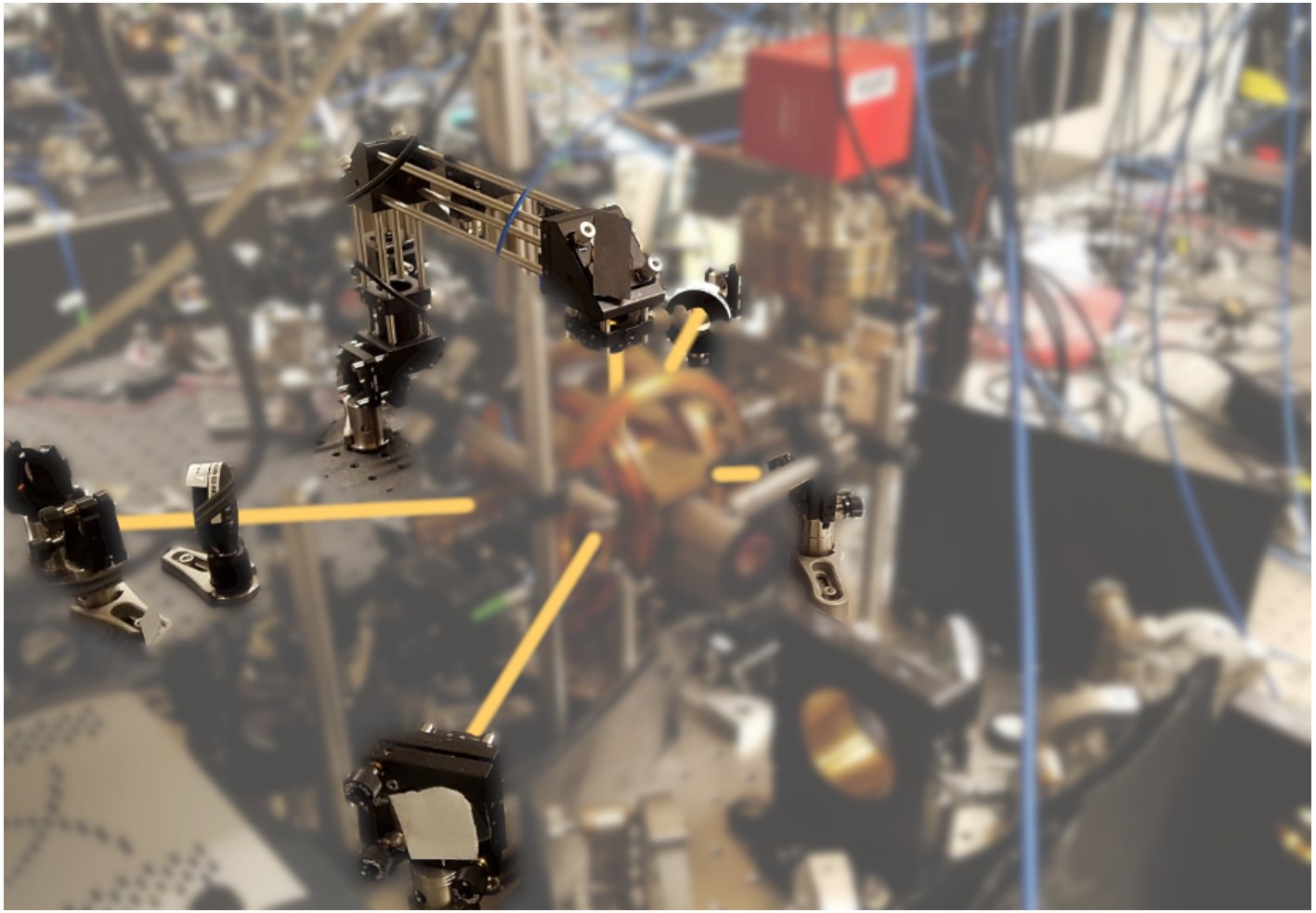
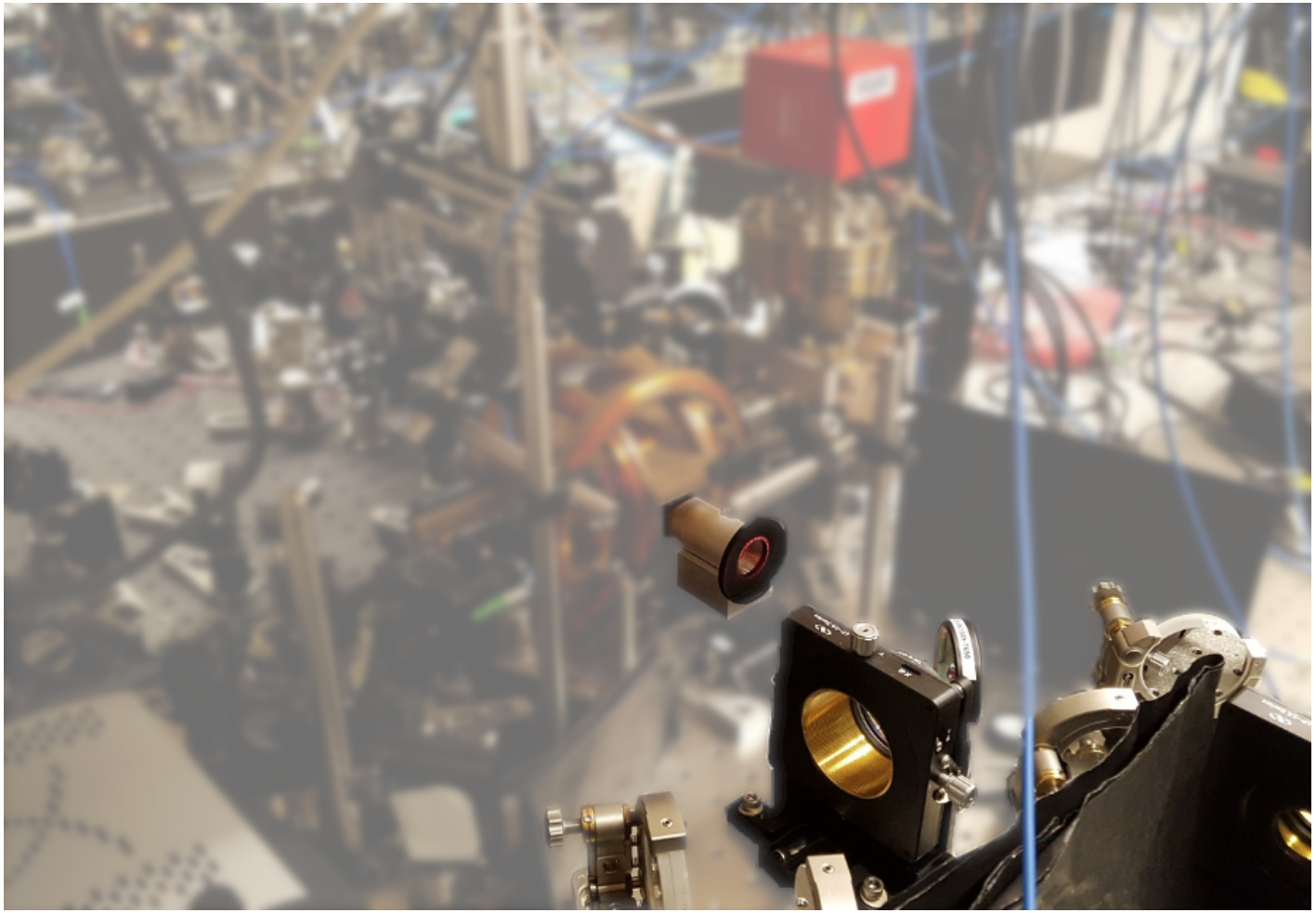



Na MOT

MOT beam path



Tweezer beam path



Outline

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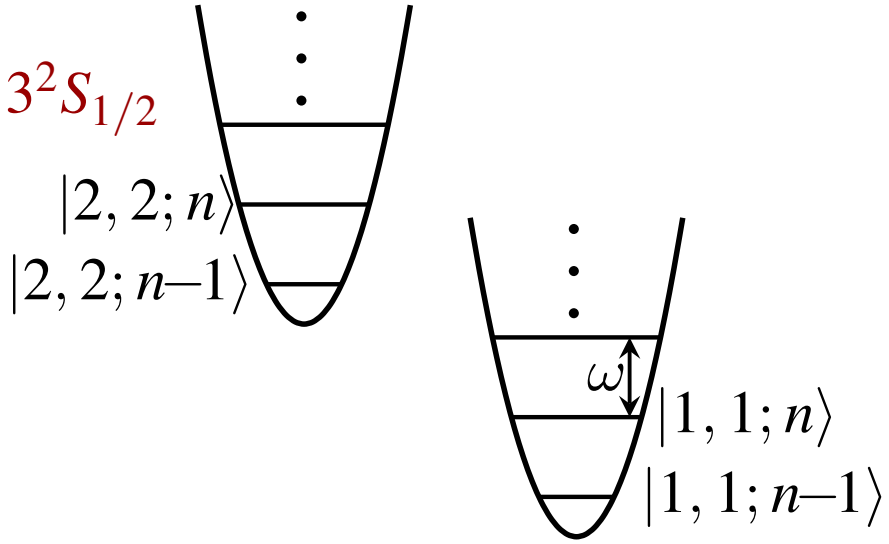
Raman sideband cooling



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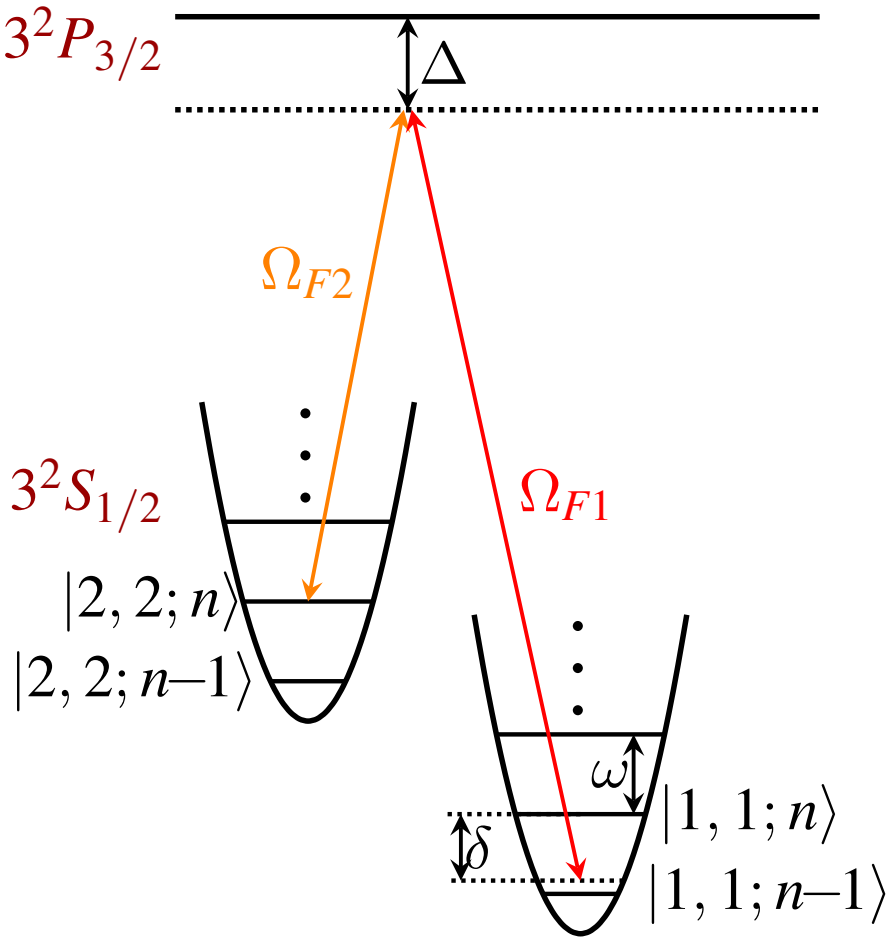

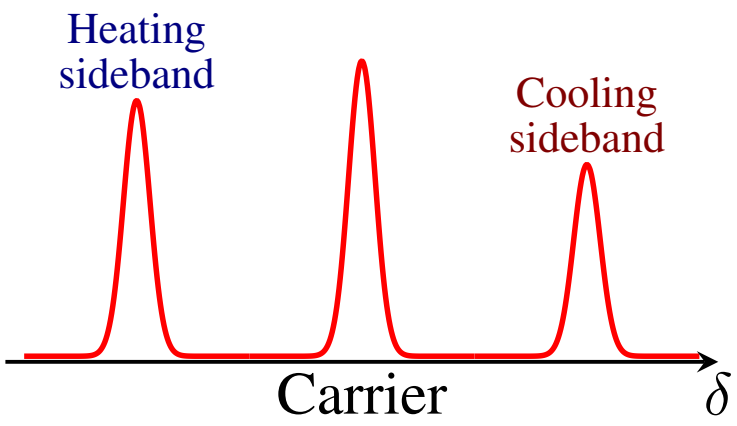

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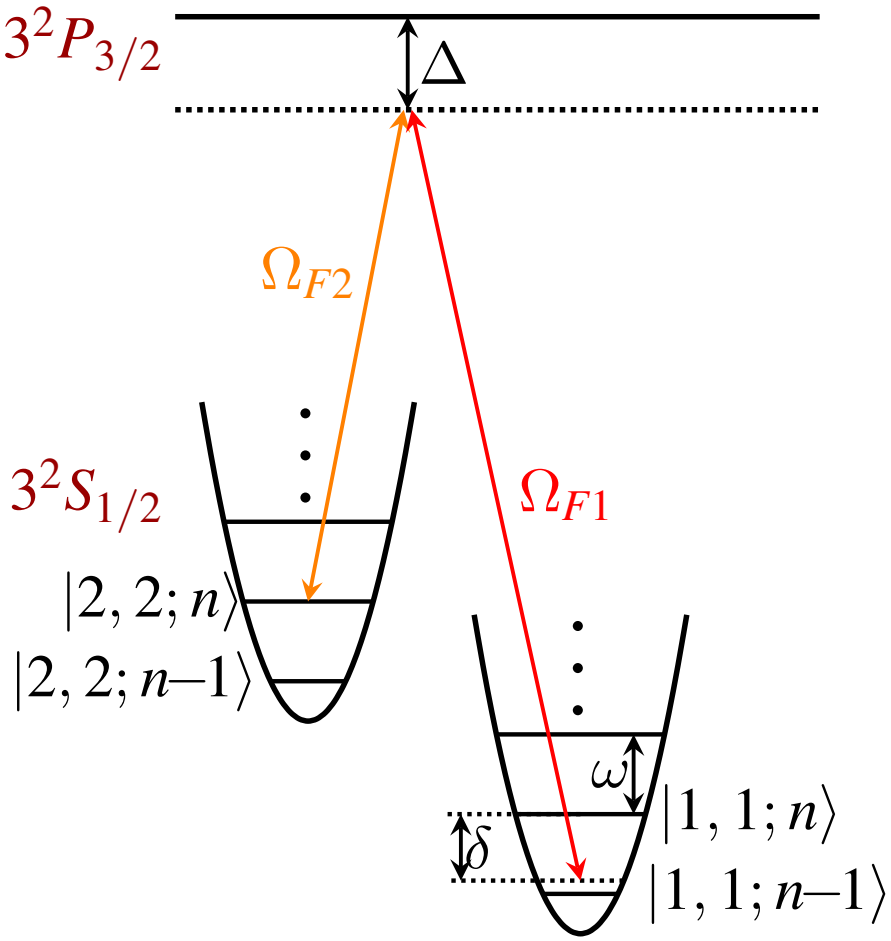

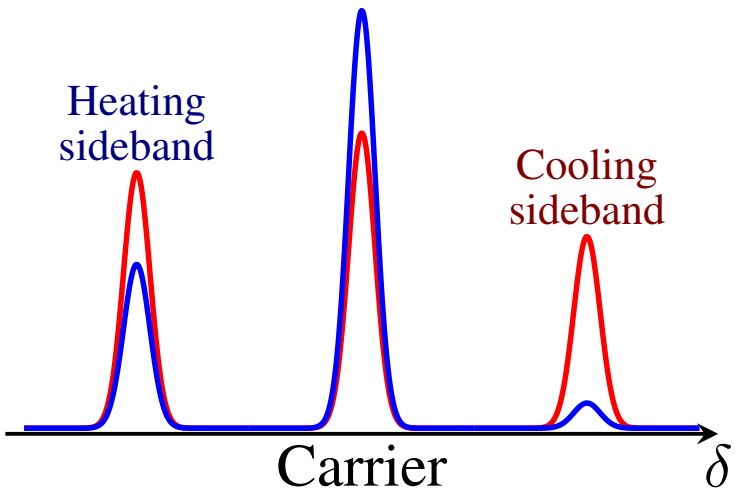

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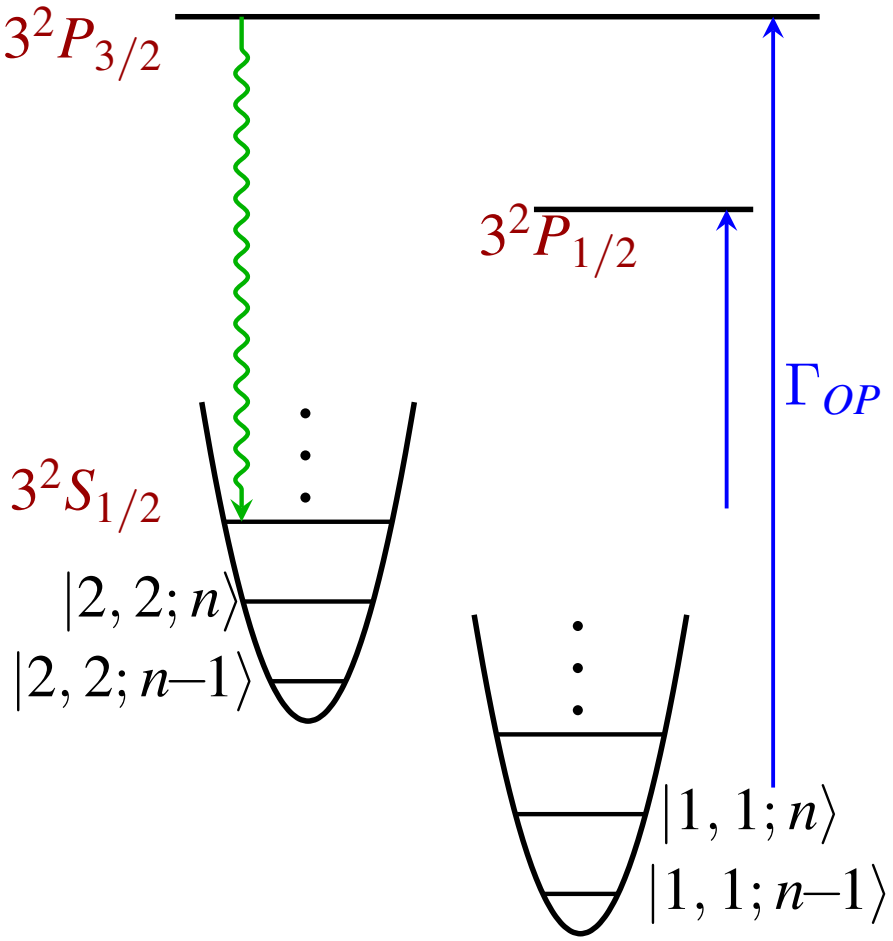

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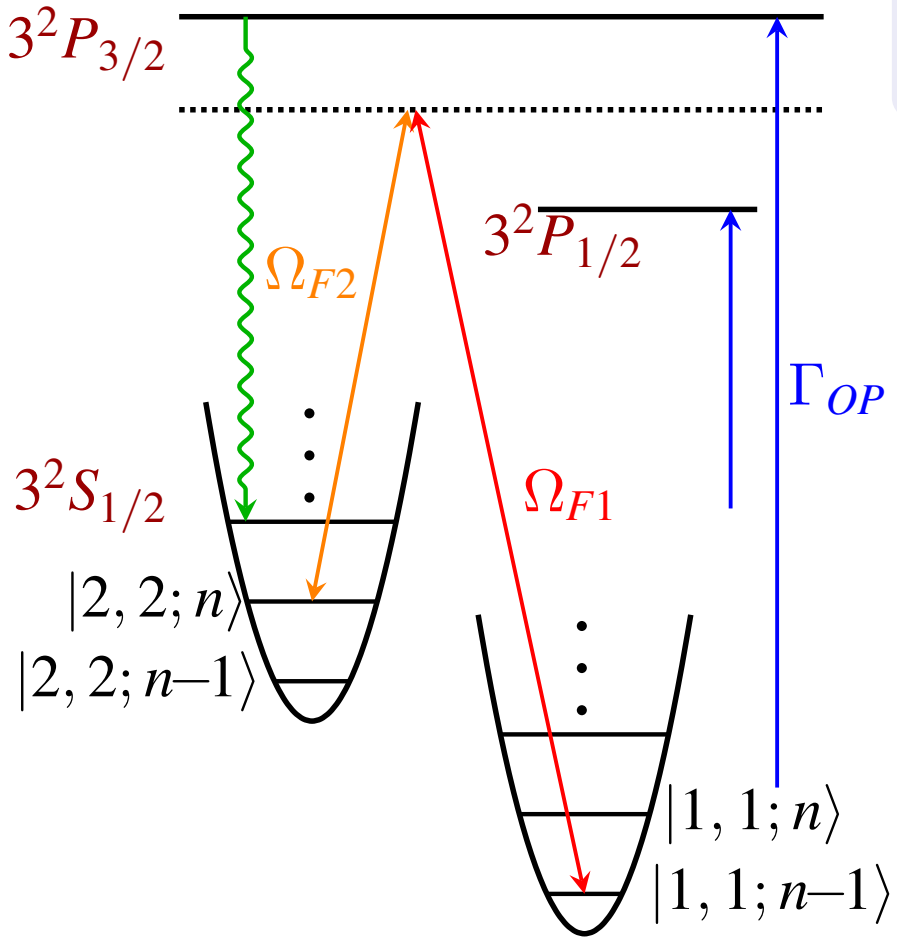
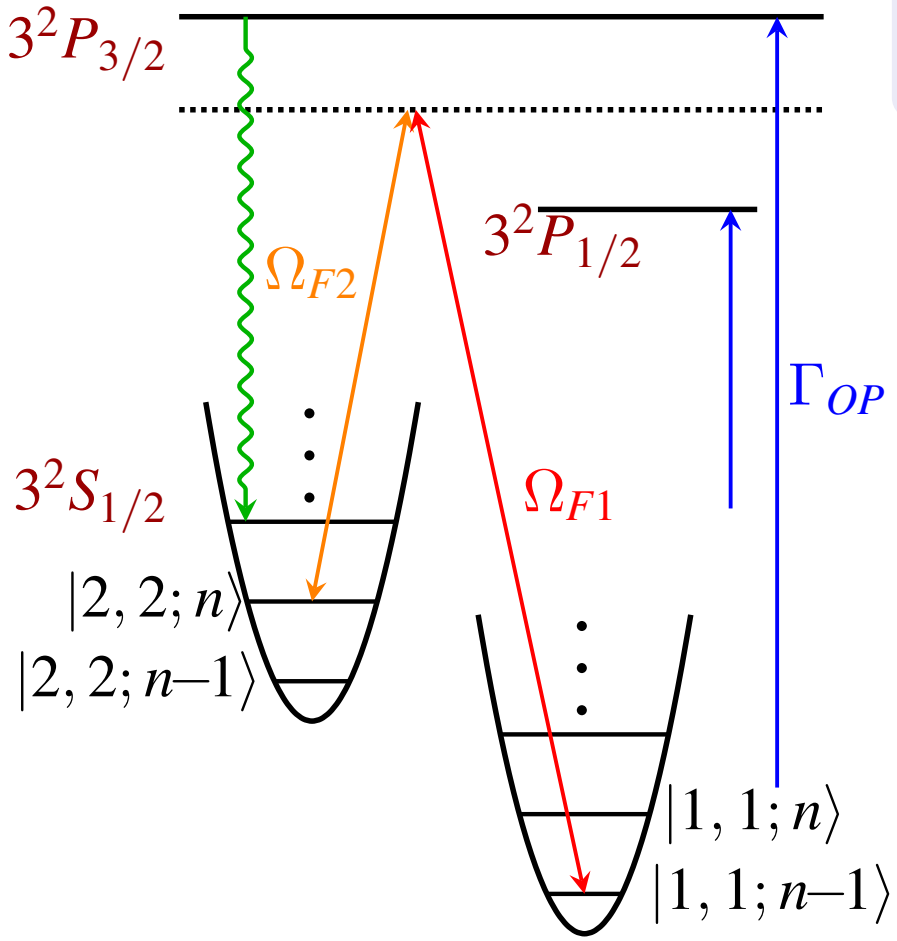


Lamb Dicke parameter

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

Raman sideband cooling



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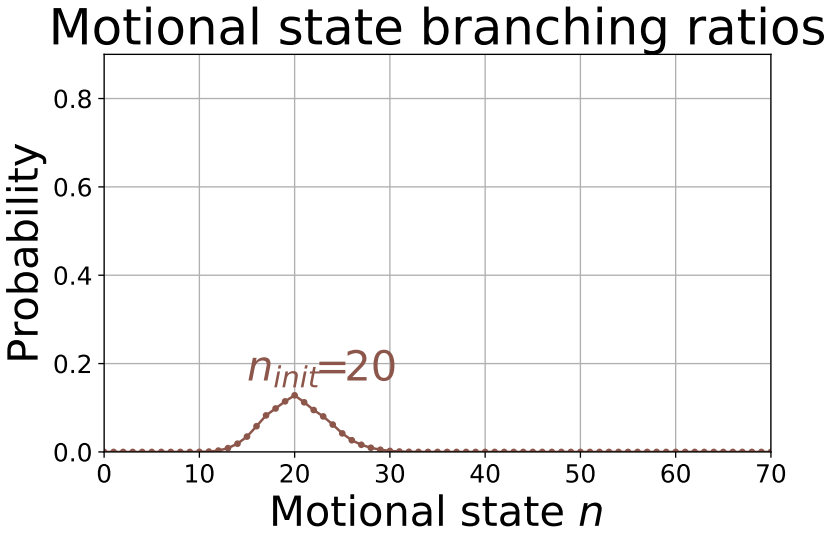

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- Motional state branching
- Coupling "dead zone"

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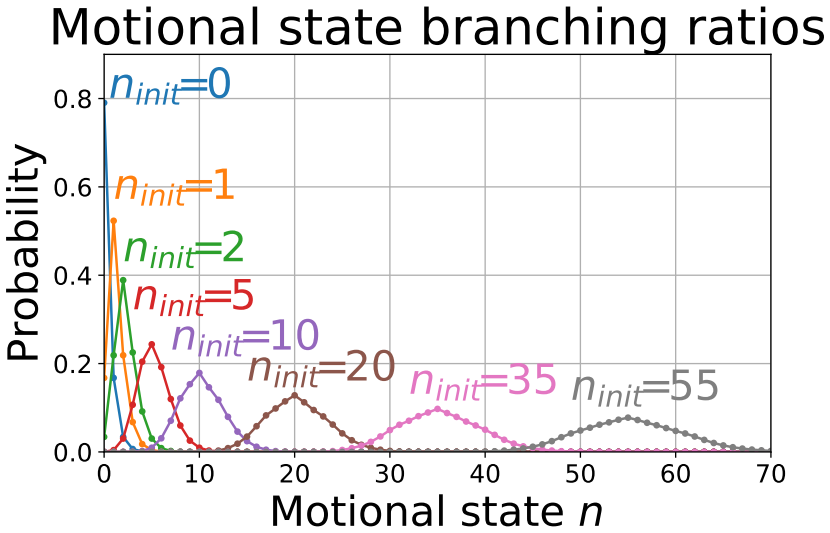

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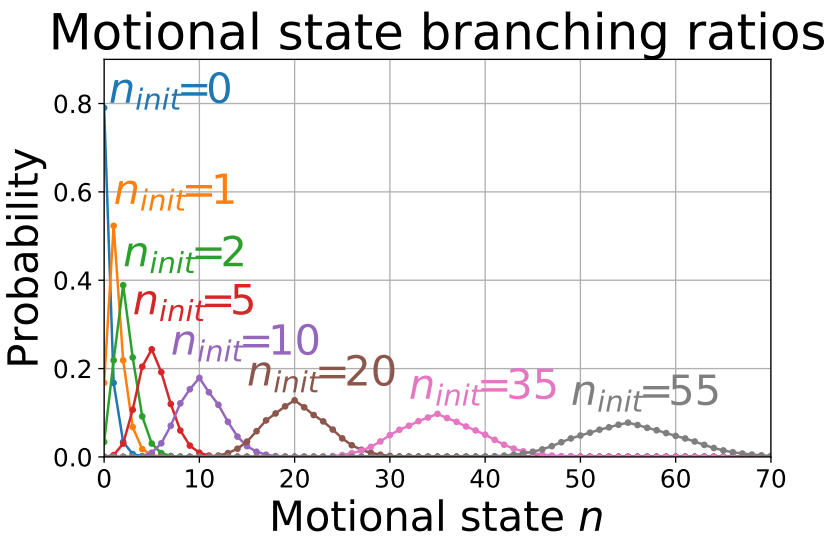

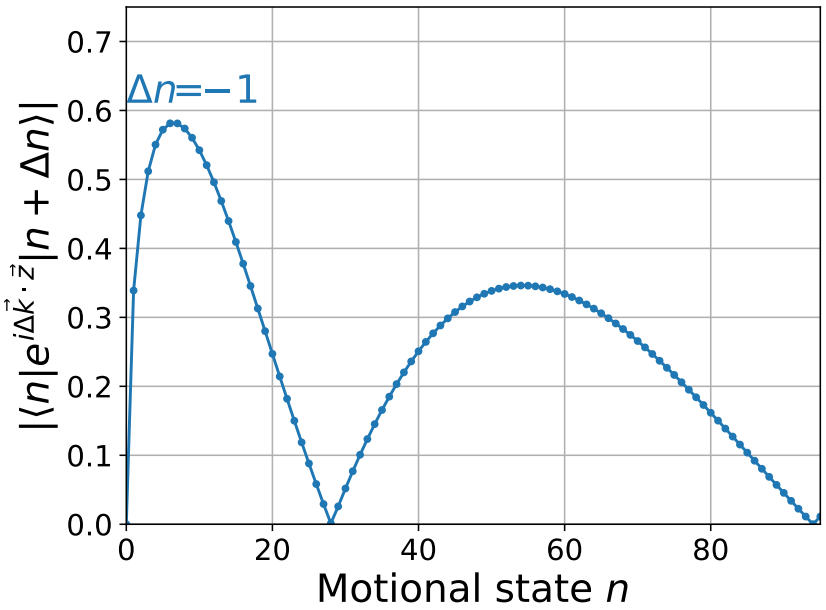

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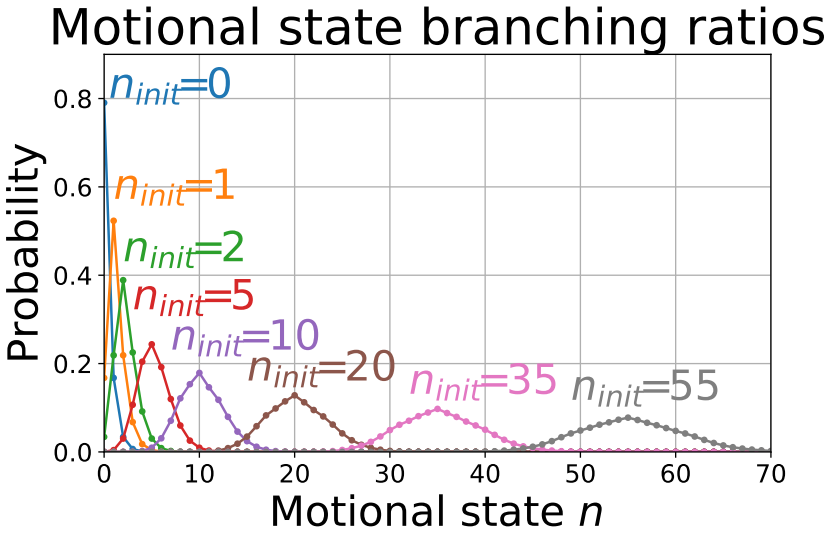

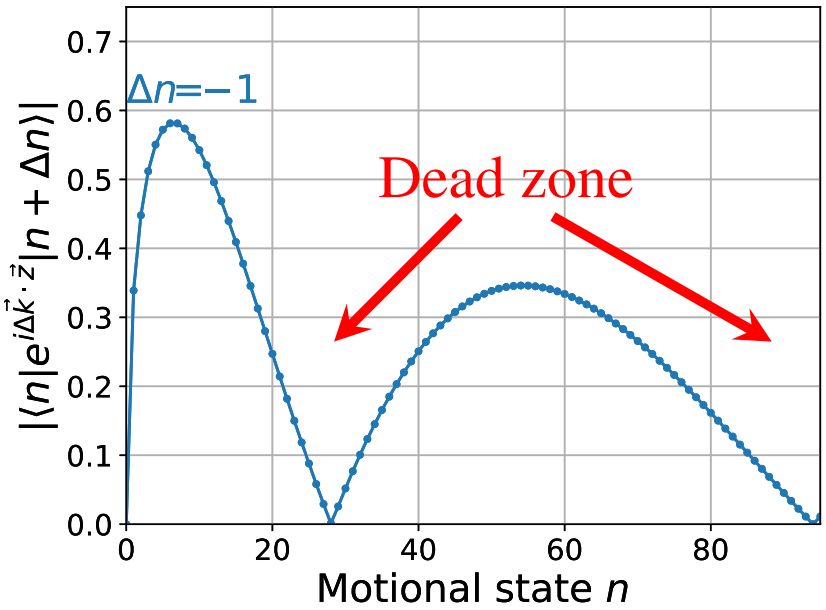

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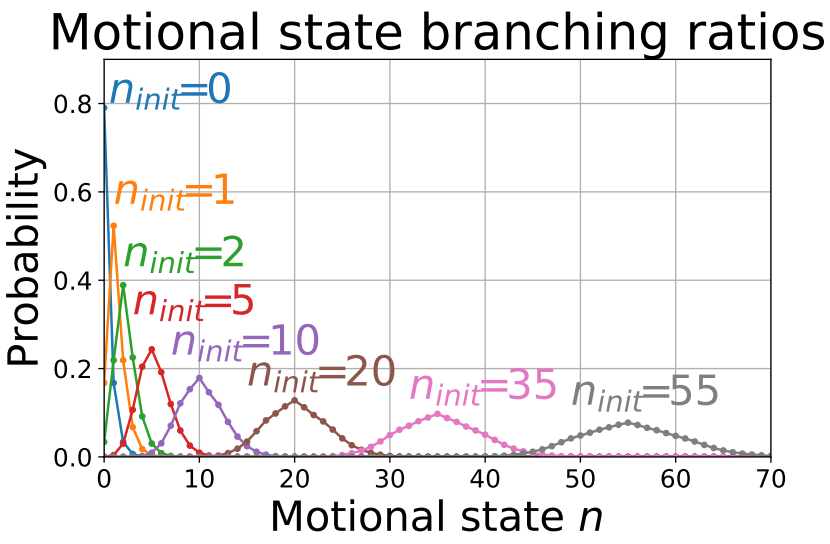

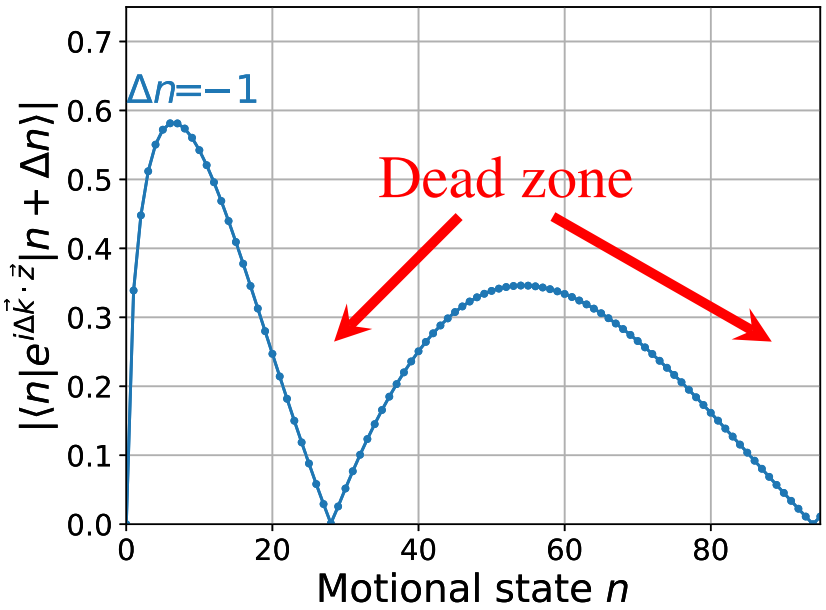

Lamb Dicke parameter

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

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

- Motional state branching
- Coupling "dead zone"

Solution

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

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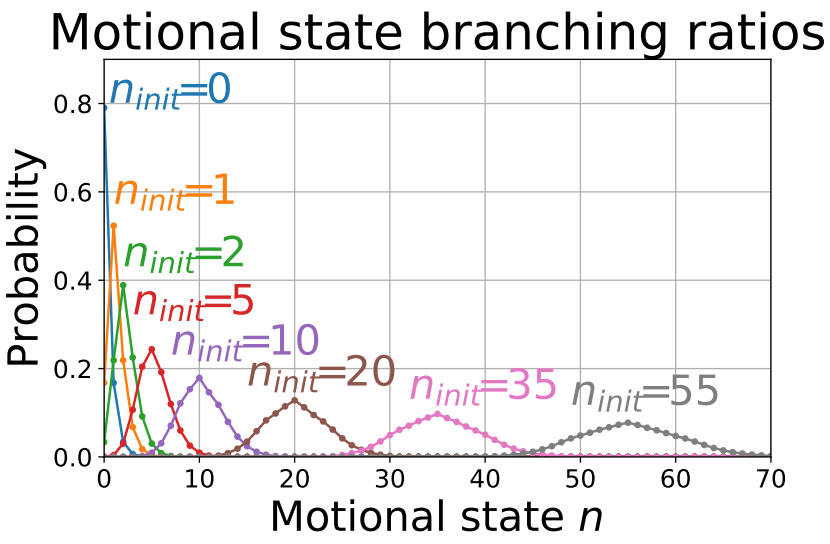

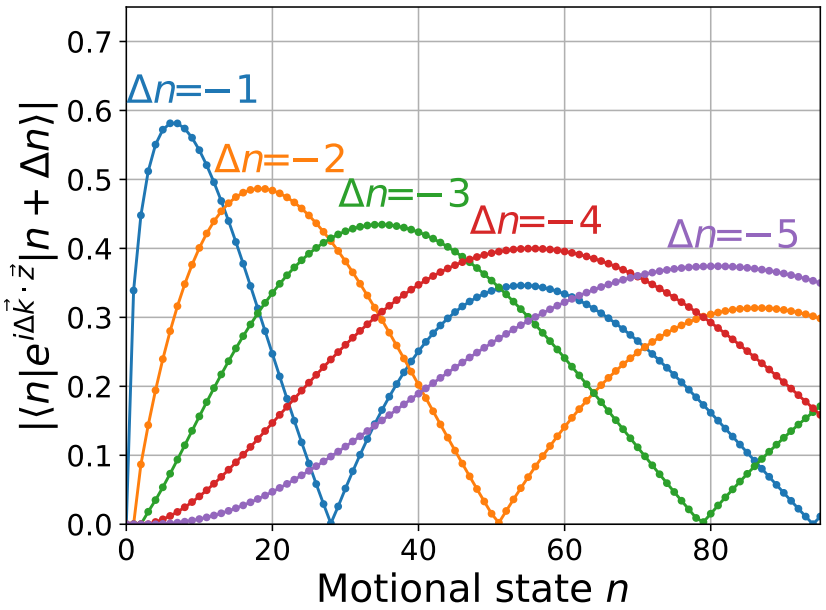

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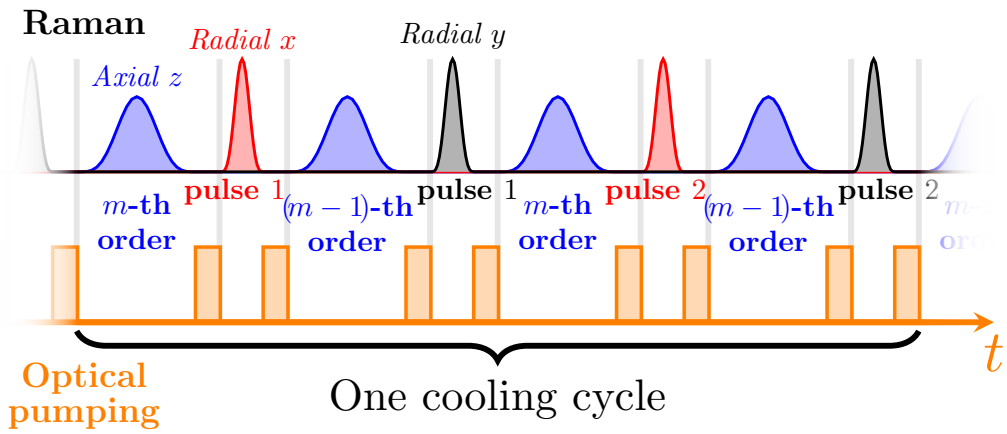

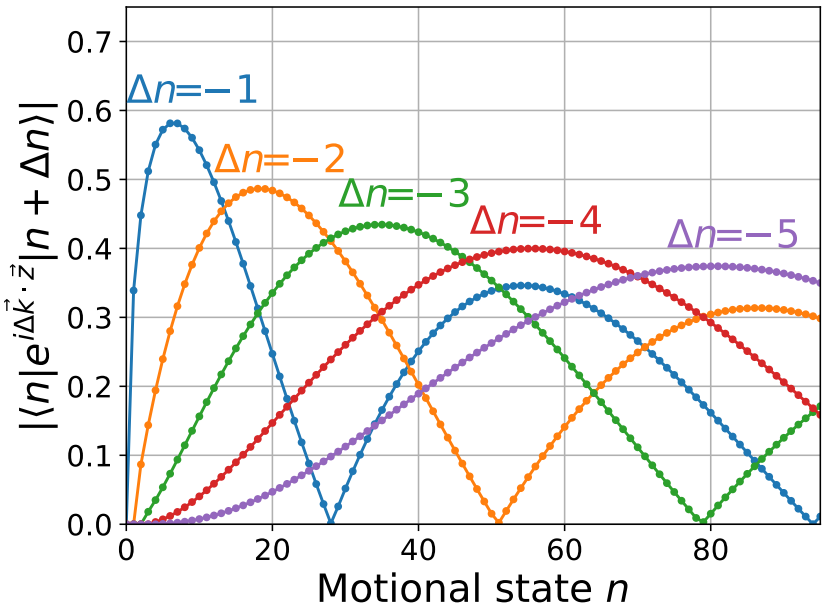

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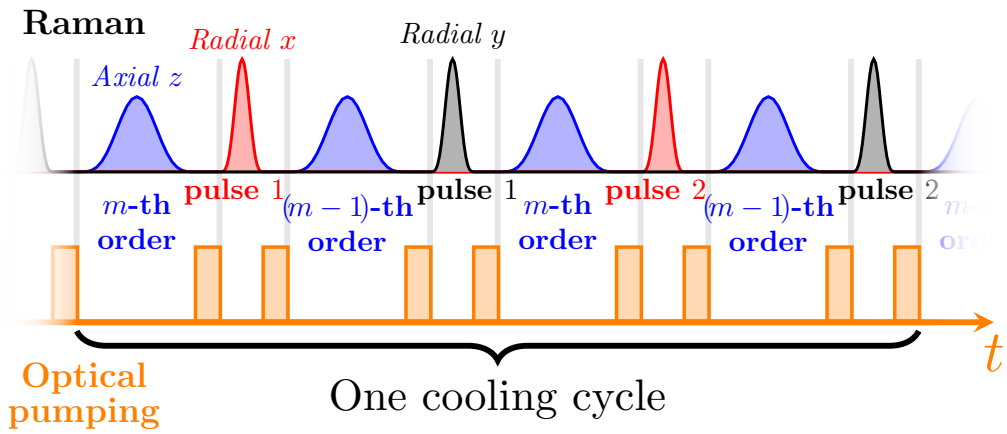
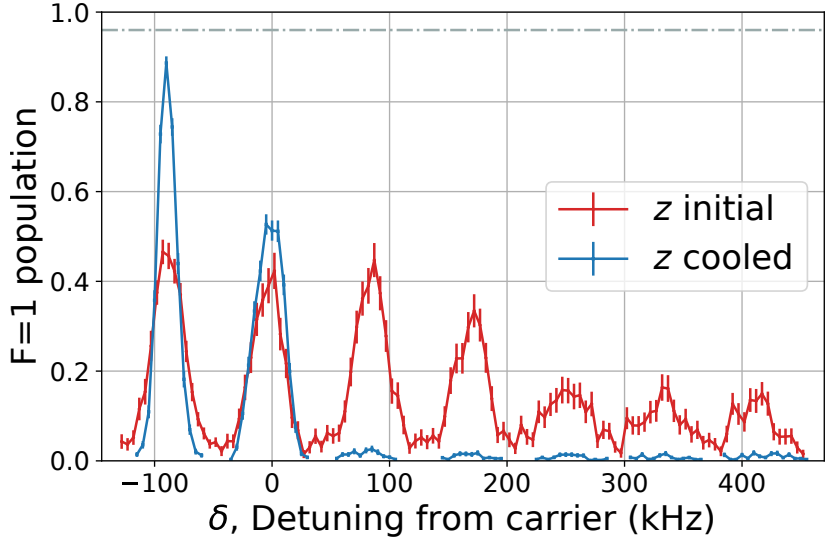


Axial sideband spectrum



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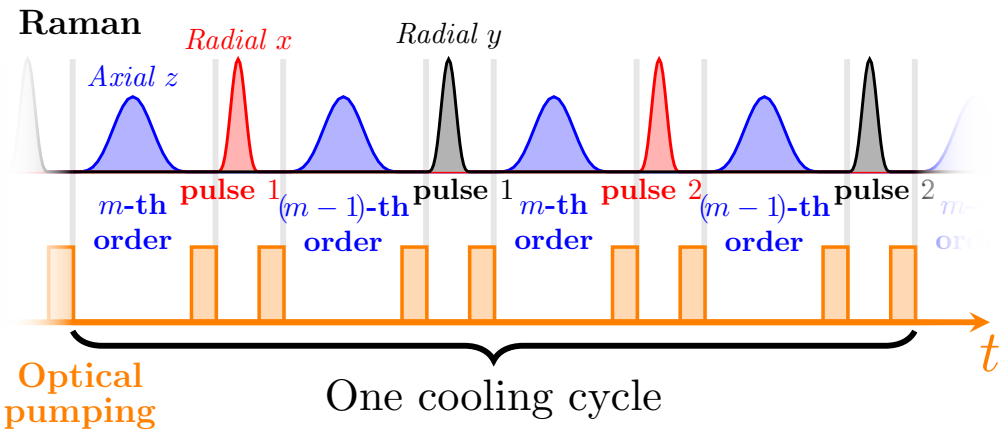

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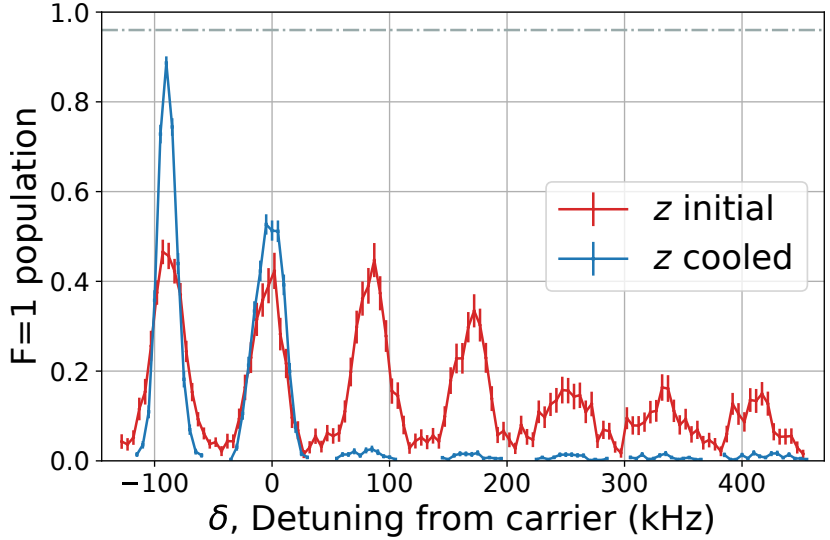

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3D ground state: 93.5(7)%

Outline

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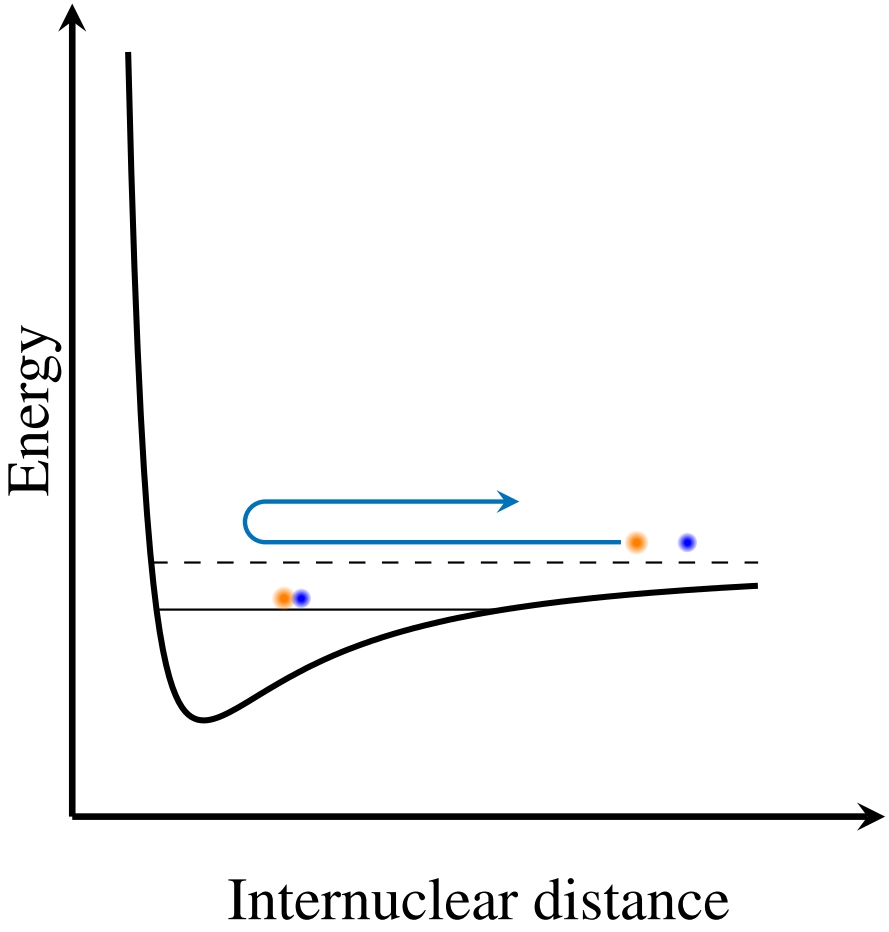
Scattering length a

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

⋮



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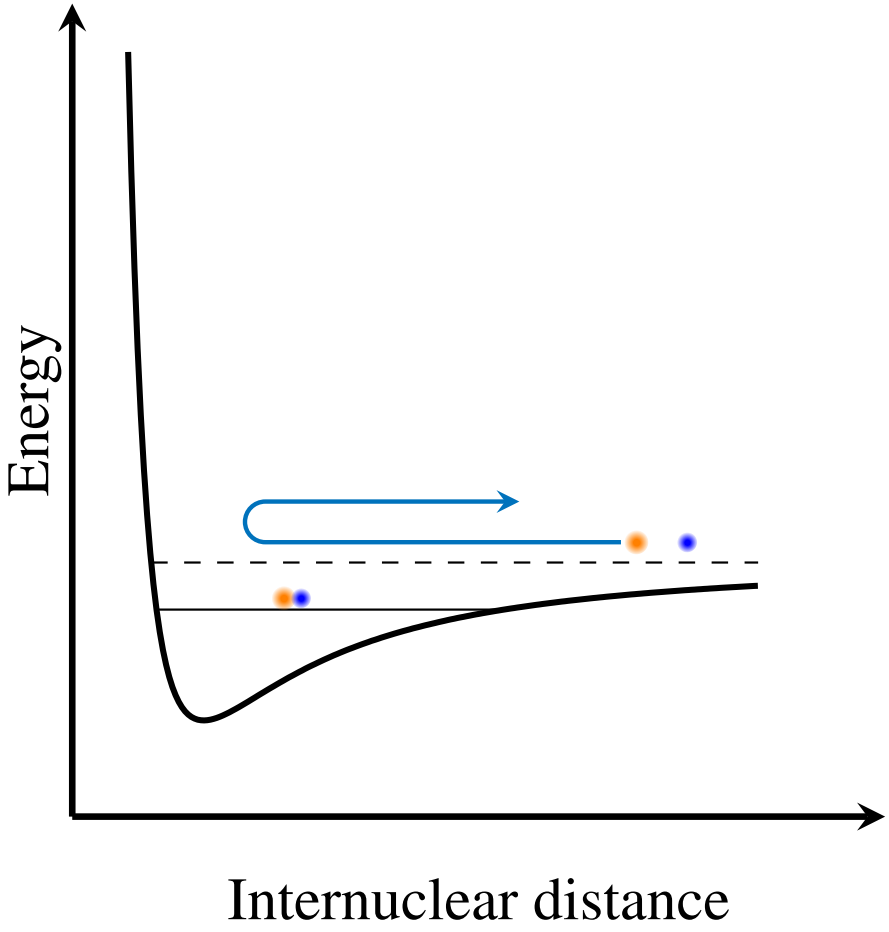

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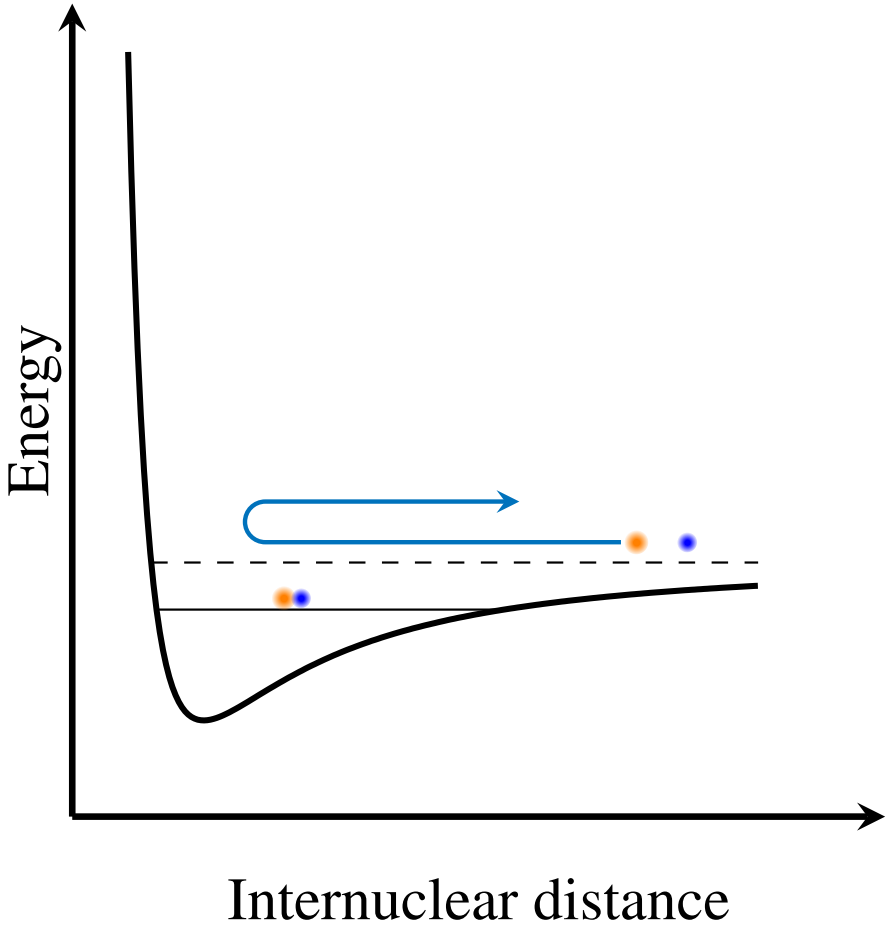

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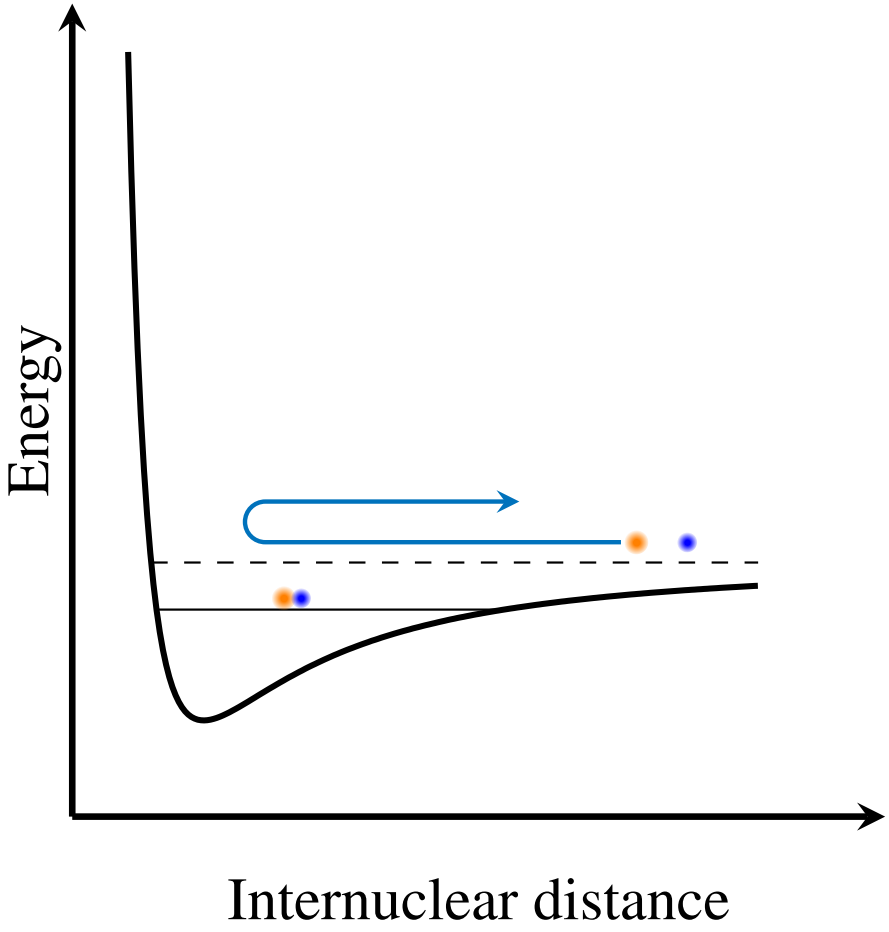

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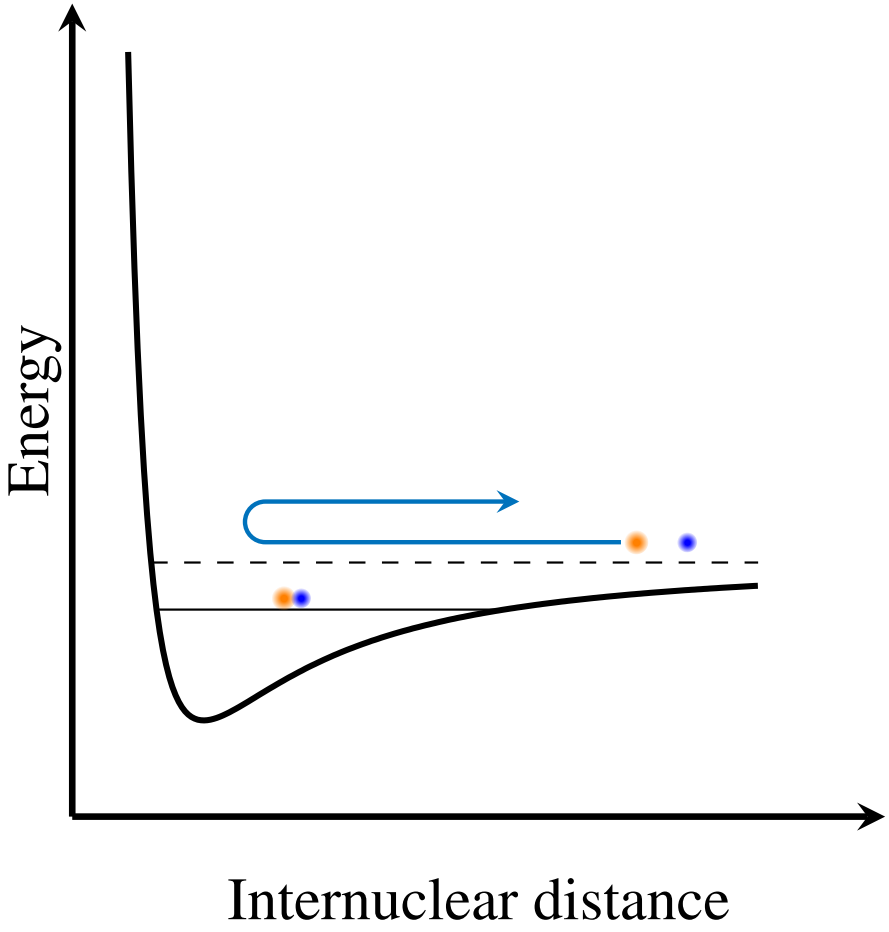
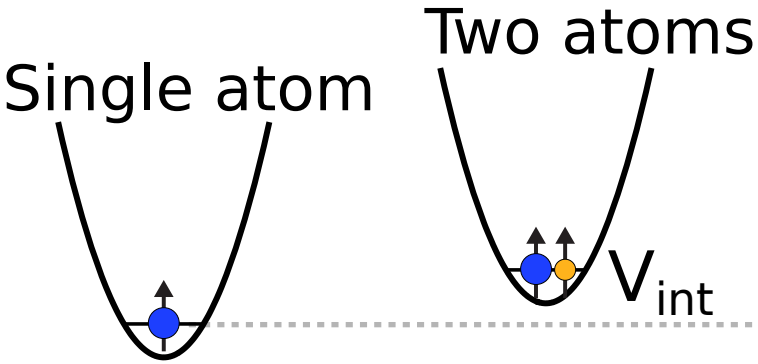


Interaction shift



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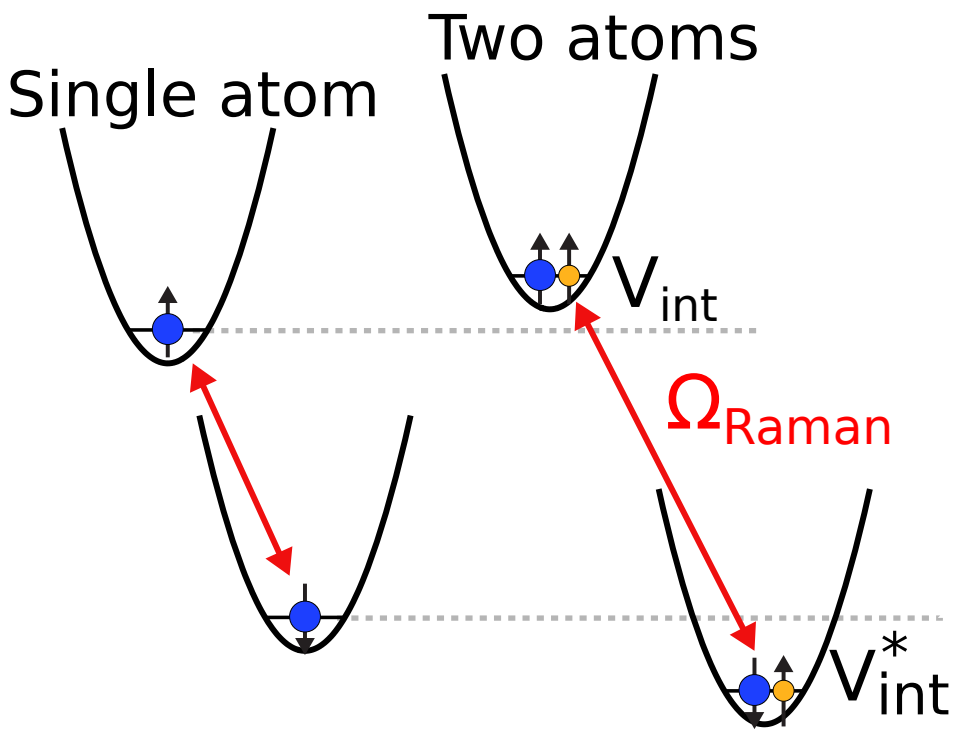

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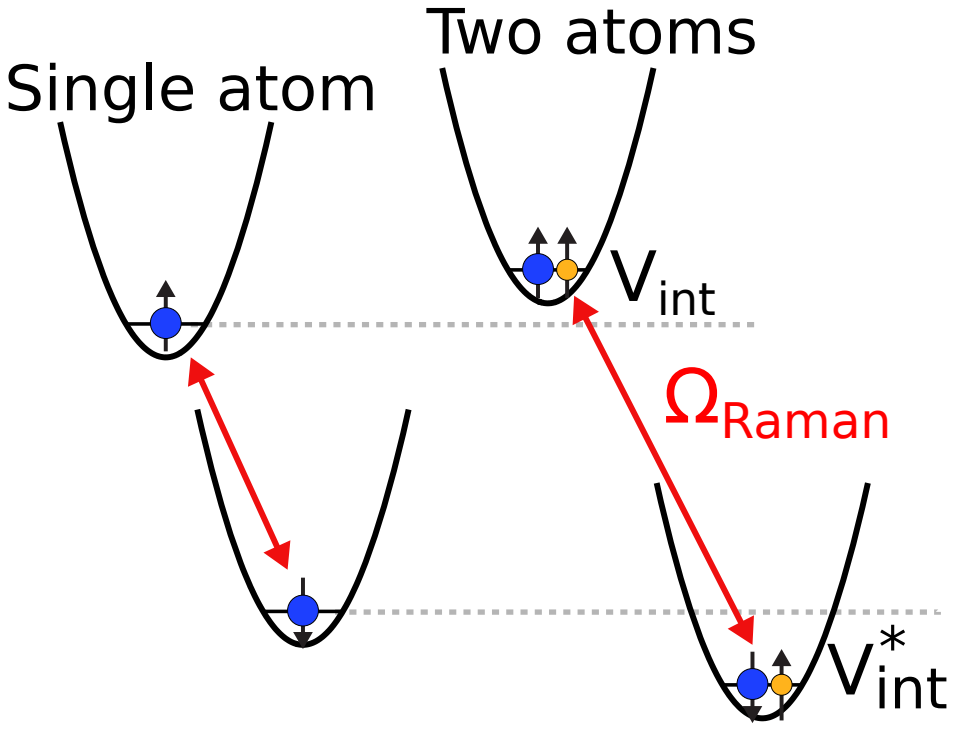

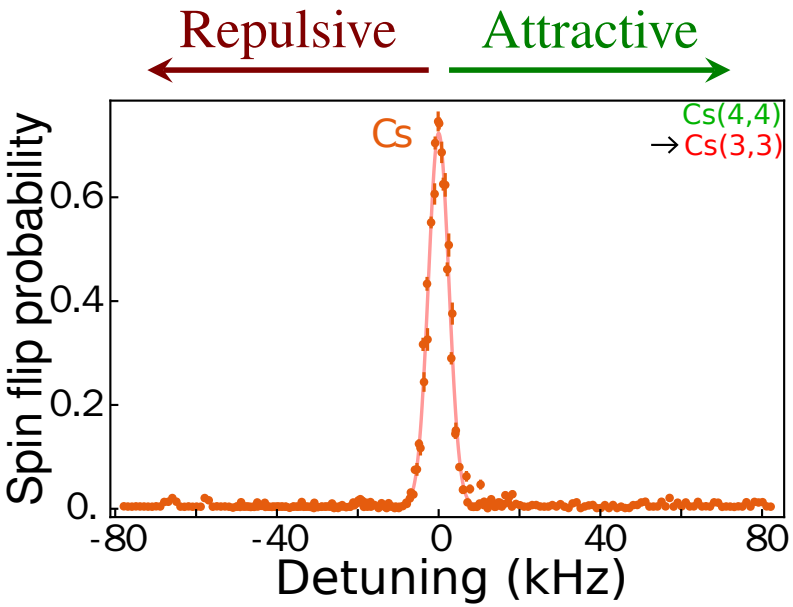

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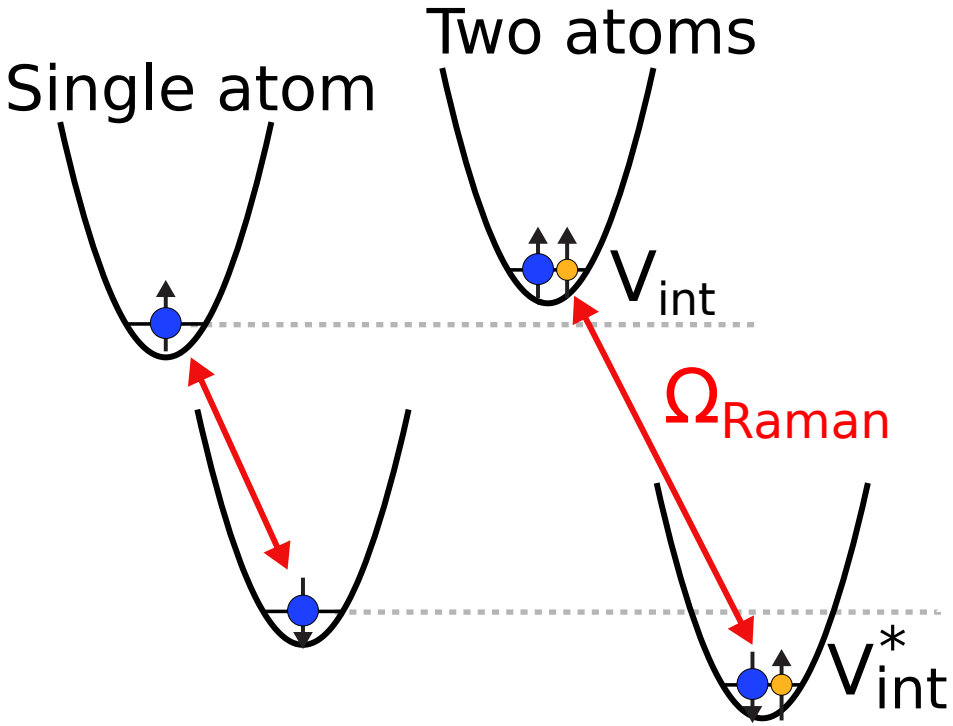
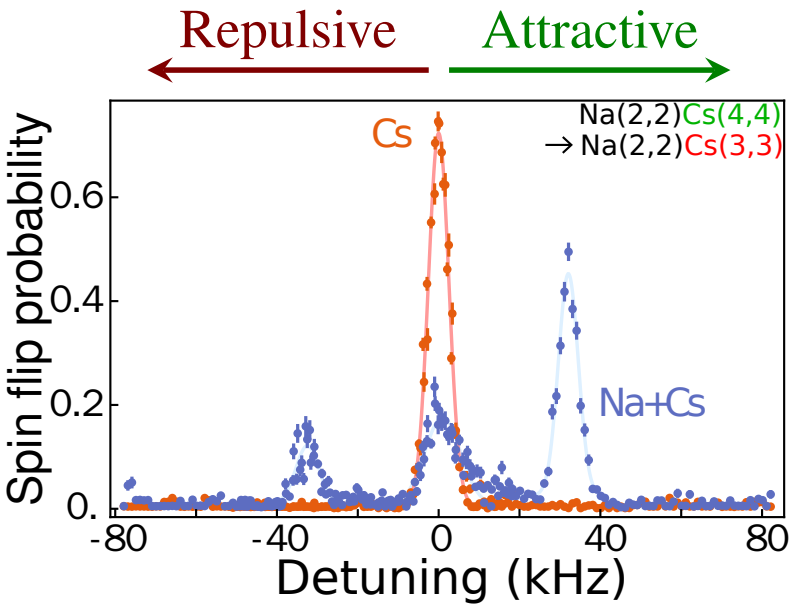
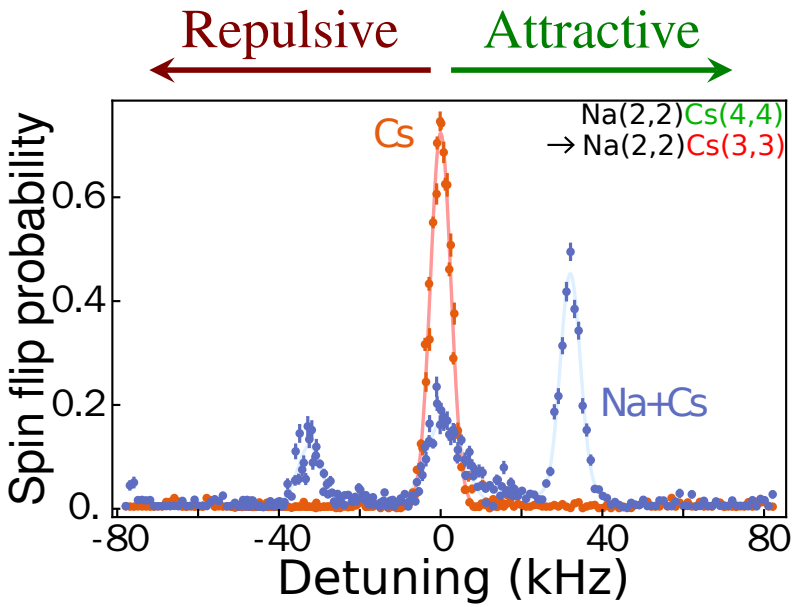




Interaction shift

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



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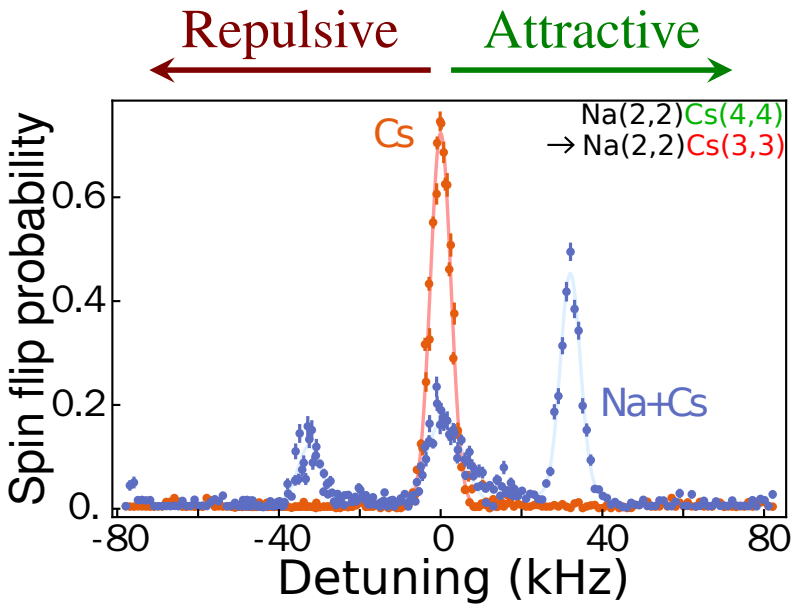
To center of mass and relative coordinates

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

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

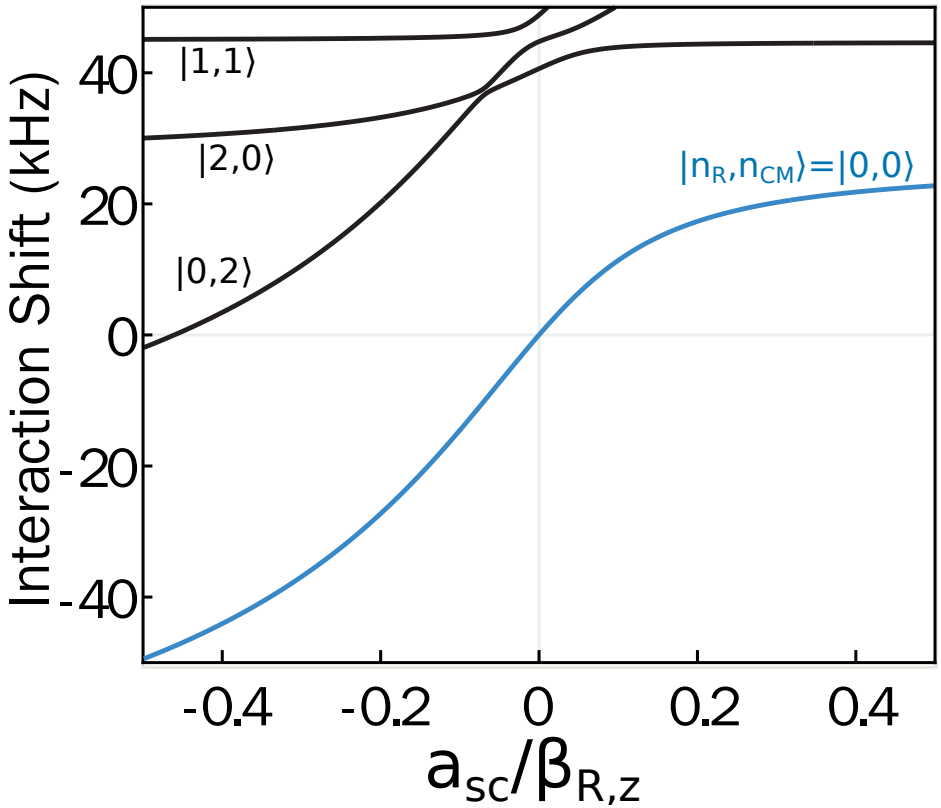
$$X_i = \frac{m_1 x_{1,i} + m_2 x_{2,i}}{m_1 + m_2} \qquad x_{R,i} = x_{1,i} - x_{2,i}$$

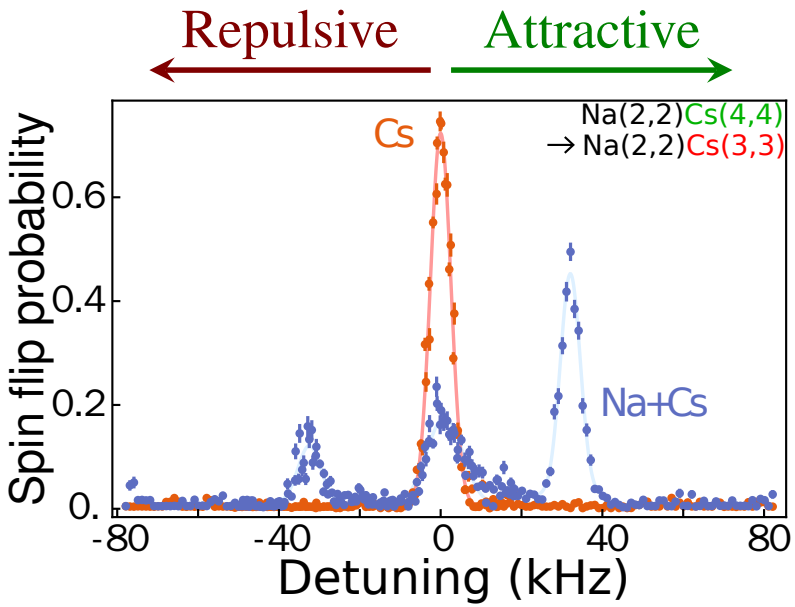
$$P_i = p_{1,i} + p_{2,i} \qquad p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$



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

Interaction shift





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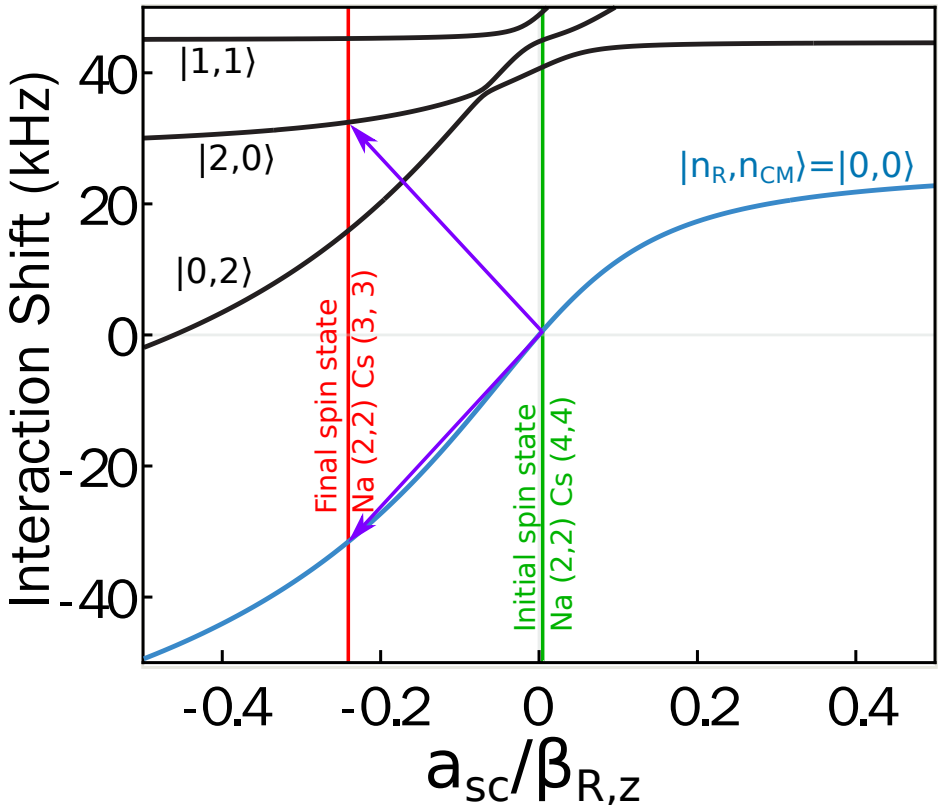

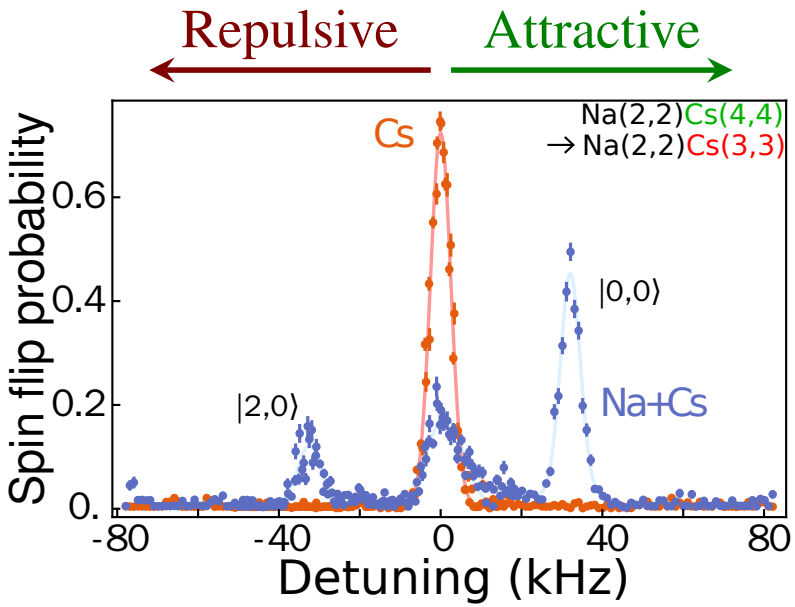

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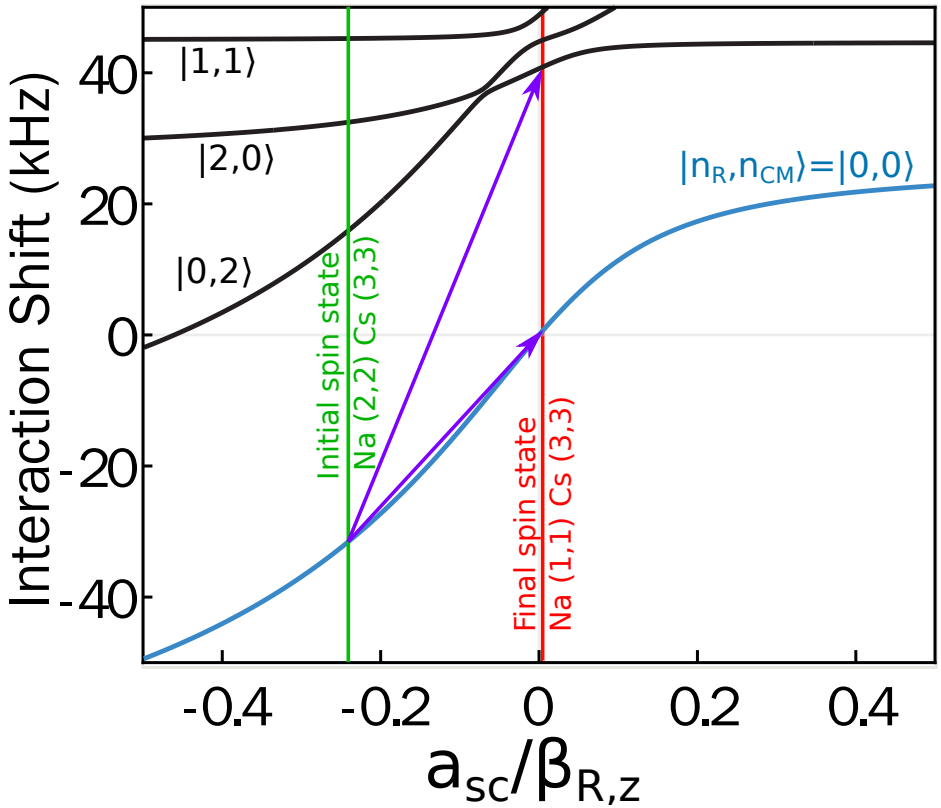

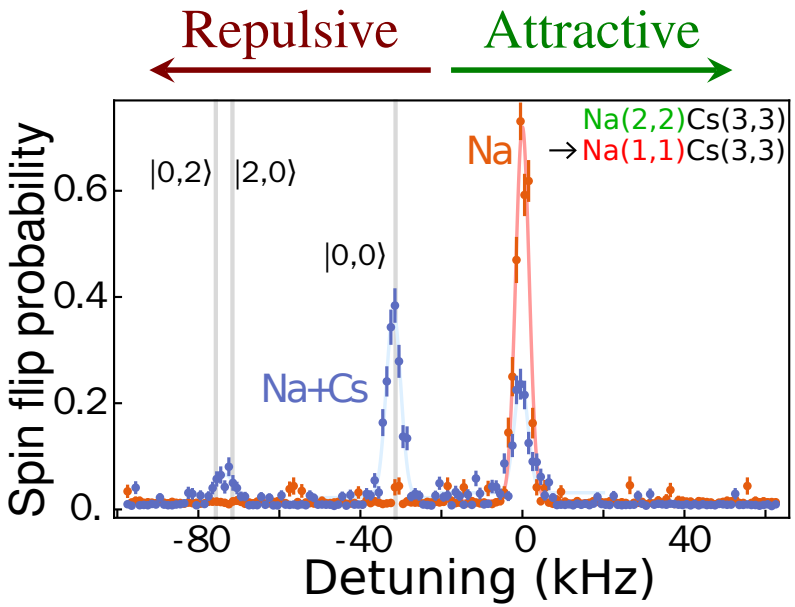

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Interaction shift

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

Spin state (F, m_F)	Scattering length (a.u.)
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Na(2,2) Cs(3,3)	-693.8
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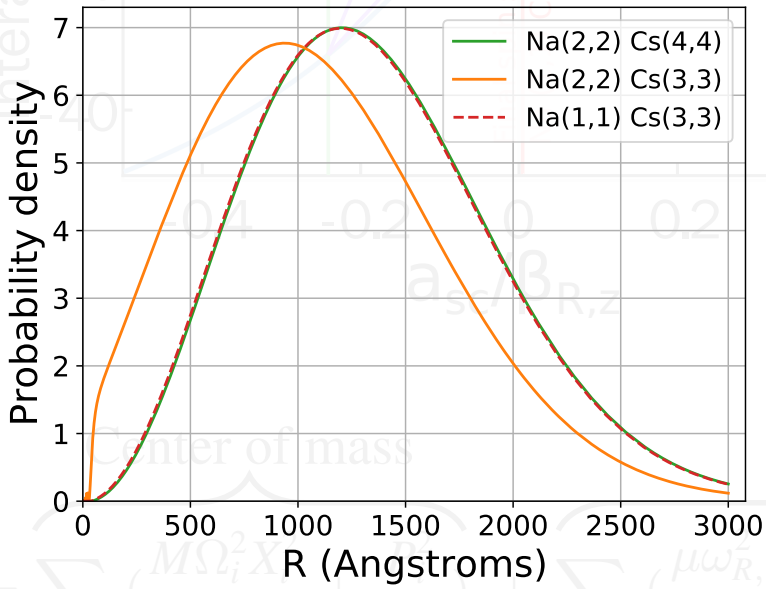
Enhanced coupling to molecular state!



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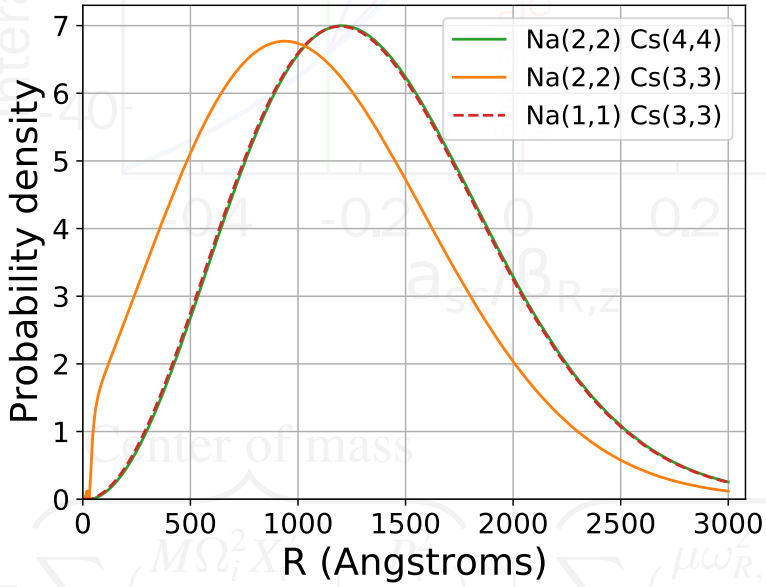

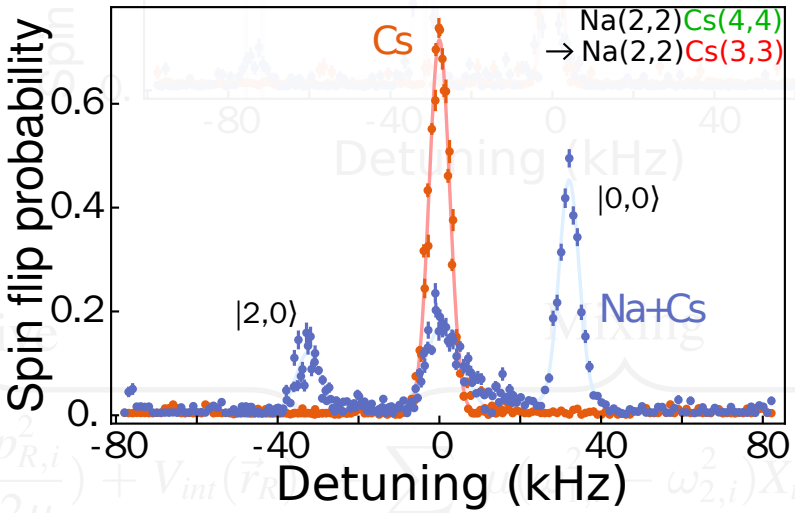

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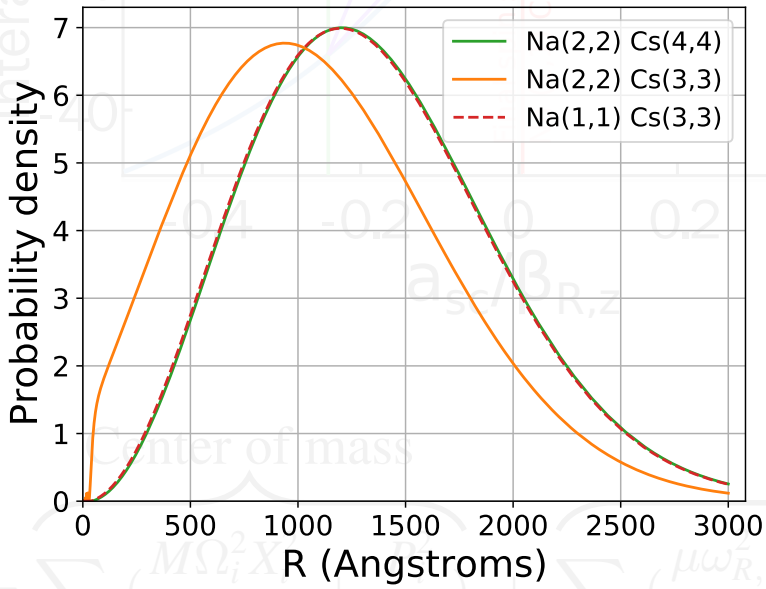
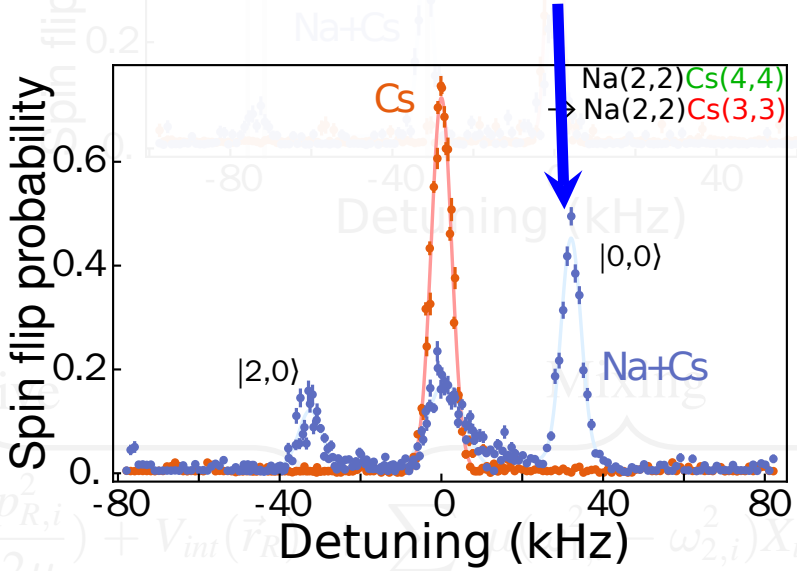


Ground state only



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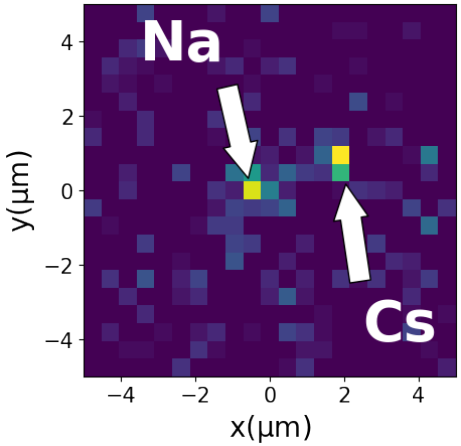
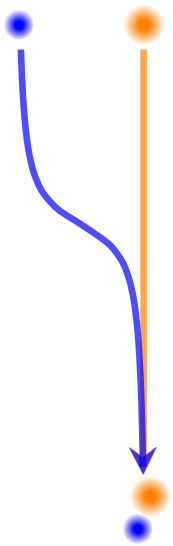
Loading

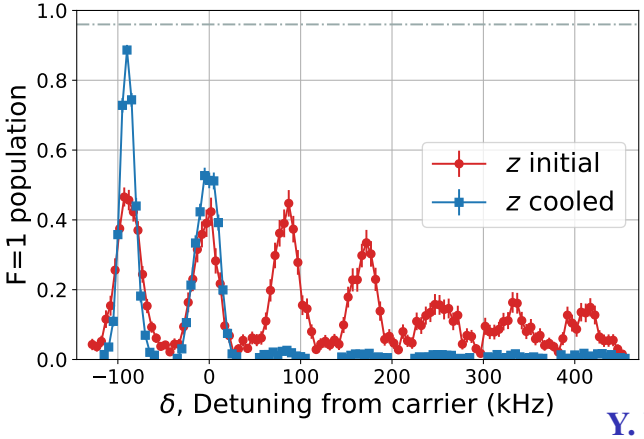


NJP. 19, 023007 (2017)

Merging



Cooling



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

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

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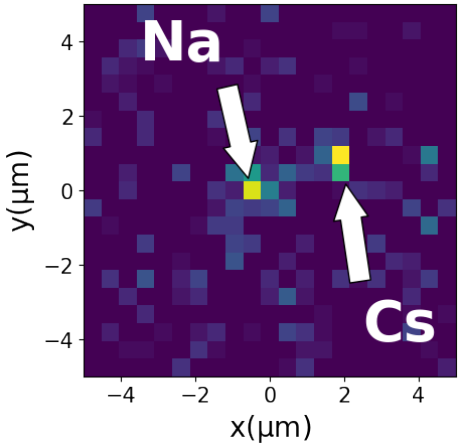
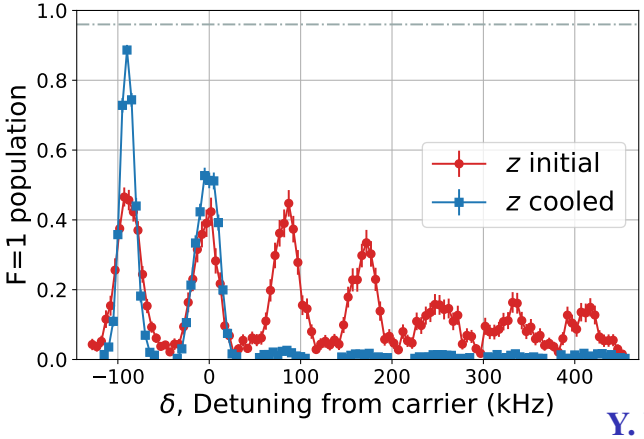


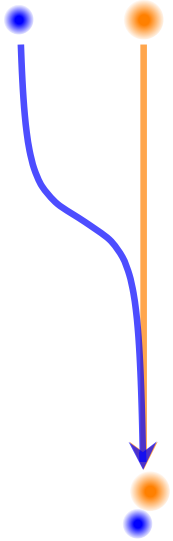
NJP. 19, 023007 (2017)

Cooling

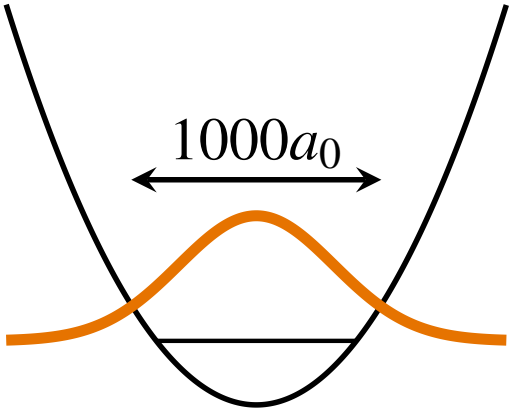


Y. Yu et al. PRA. 97, 063423 (2018)

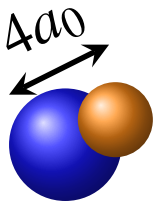
Merging



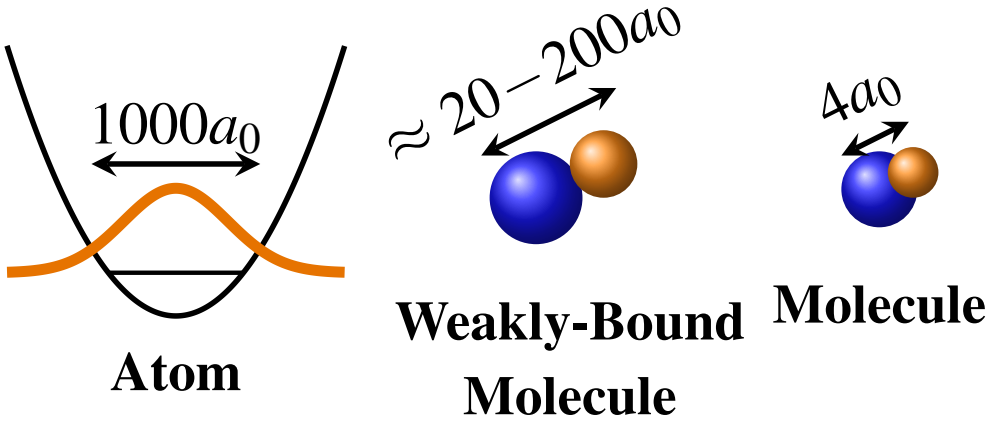
Y. Yu et al. PRX. 9, 021039 (2019)

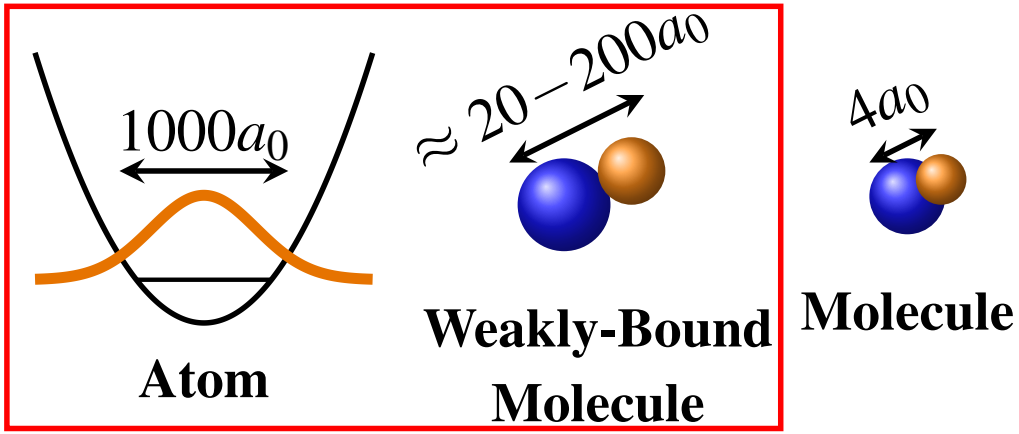


Atom



Molecule


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


$1000a_0$
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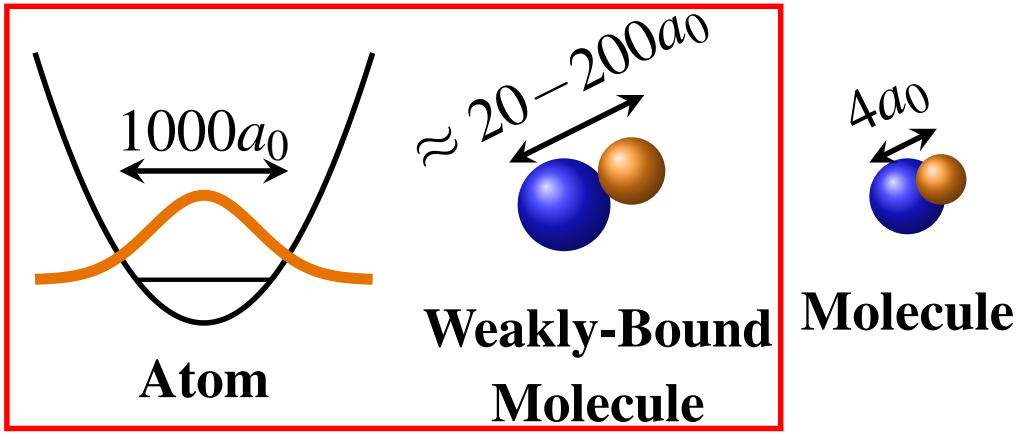
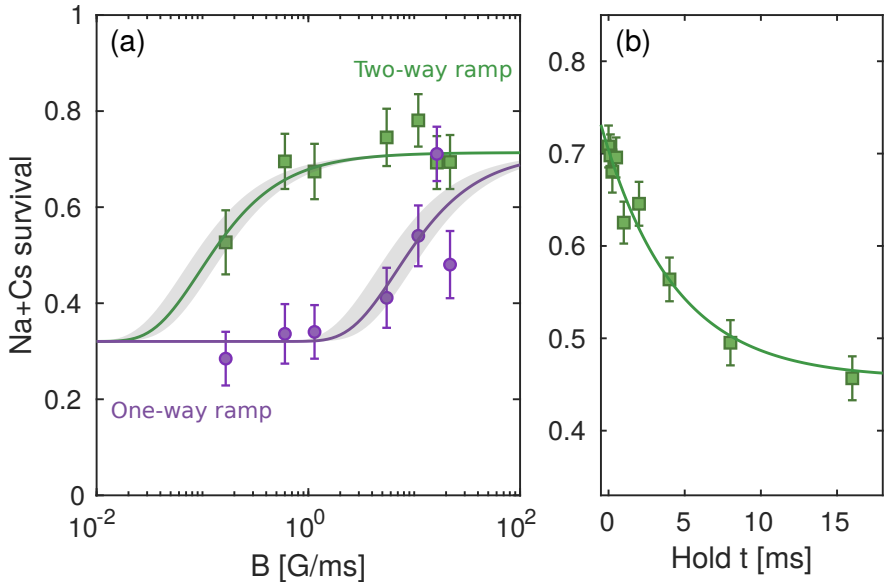
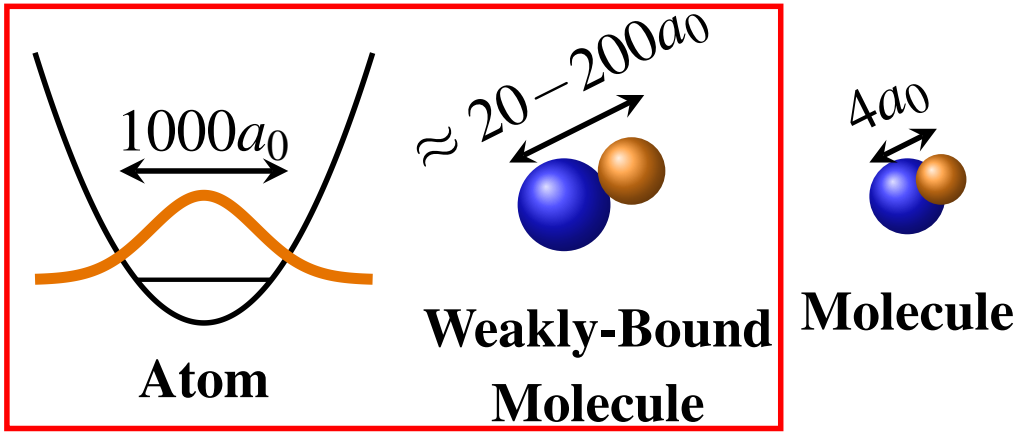


Feshbach molecule



PRL. 124, 253401 (2020)



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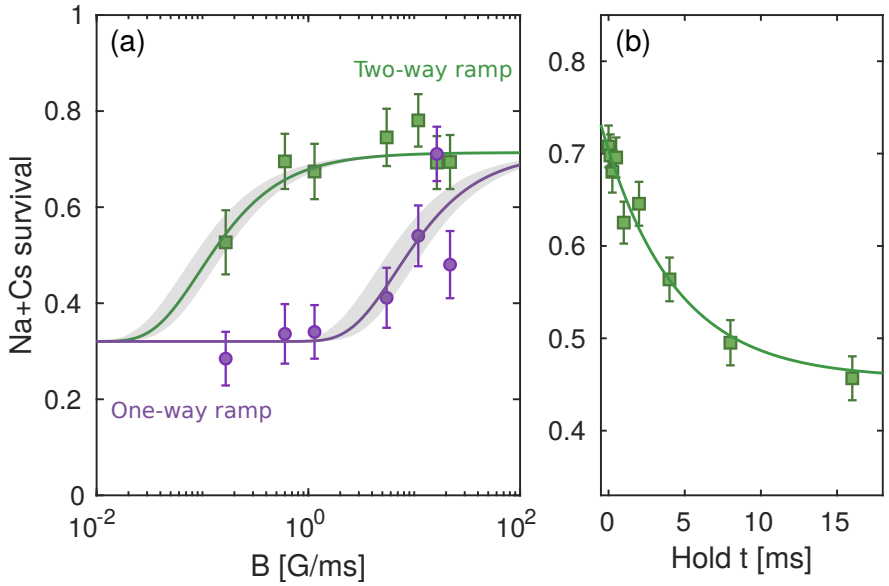
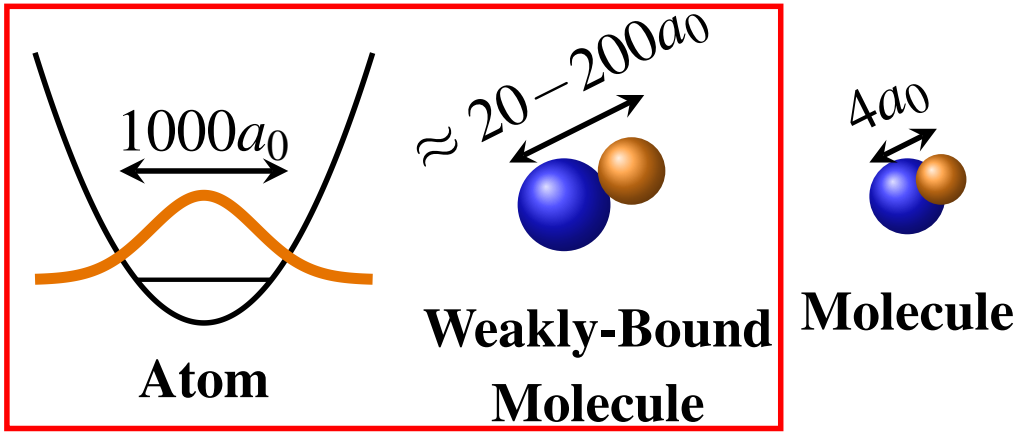


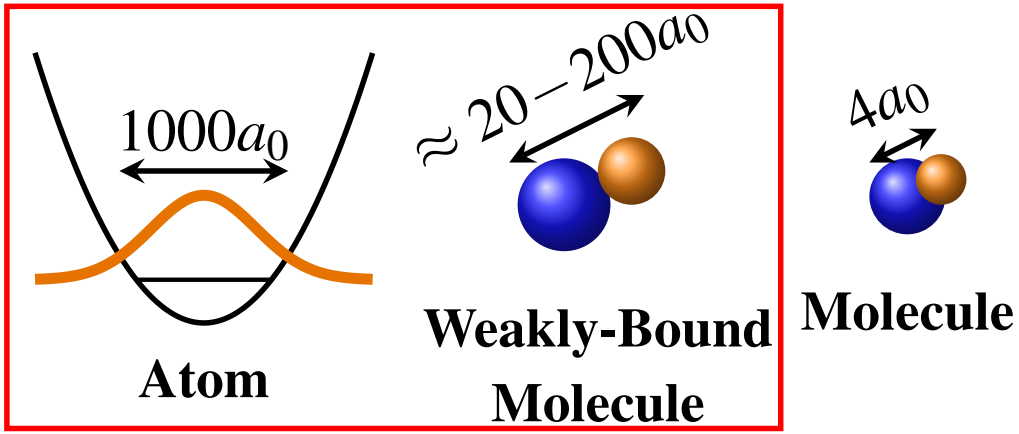
PRL. 124, 253401 (2020)

- Requires Feshbach resonance
- Usually large magnetic field



Optical transfer

- More general
- Faster



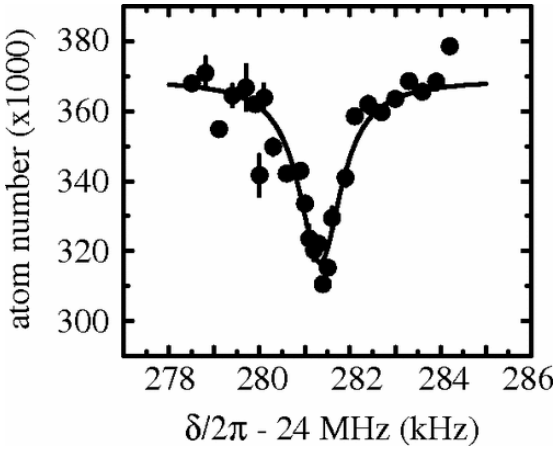
Optical transfer

- More general
- Faster

Previous results

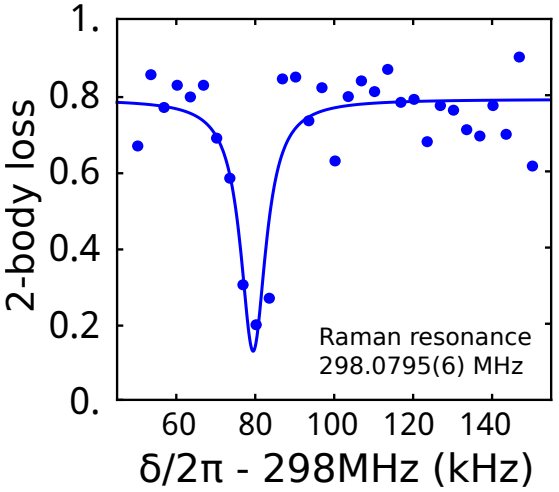
Rb_2 Science 287, 1016 (2000)

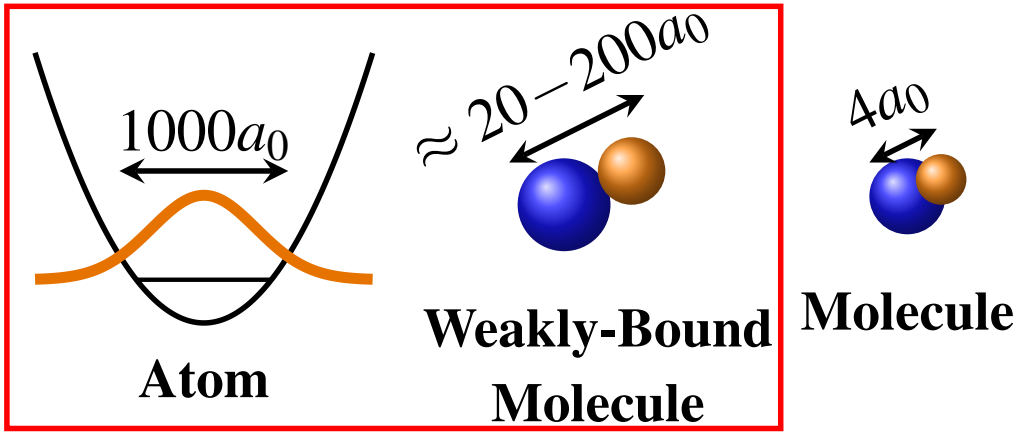
PRL. 93, 073002 (2004)



Sr_2 PRL. 109, 115302 (2012)

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





Optical transfer

- More general
- Faster

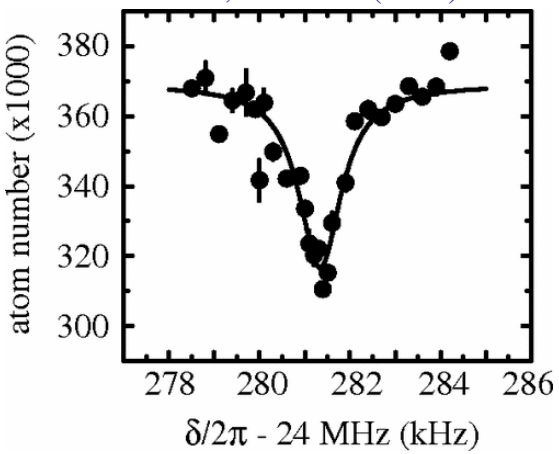
Limitations so far

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

Previous results

Rb_2 Science 287, 1016 (2000)

PRL. 93, 073002 (2004)



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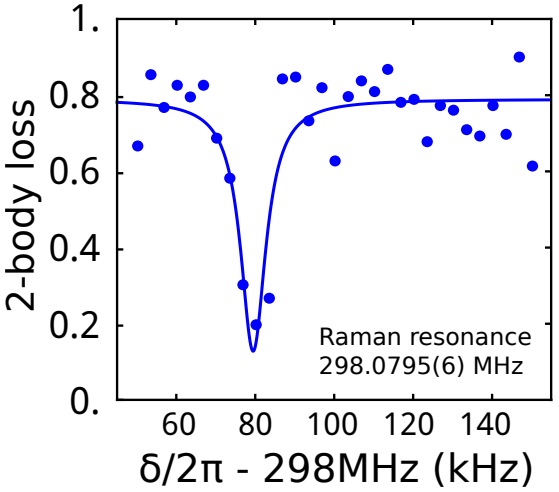
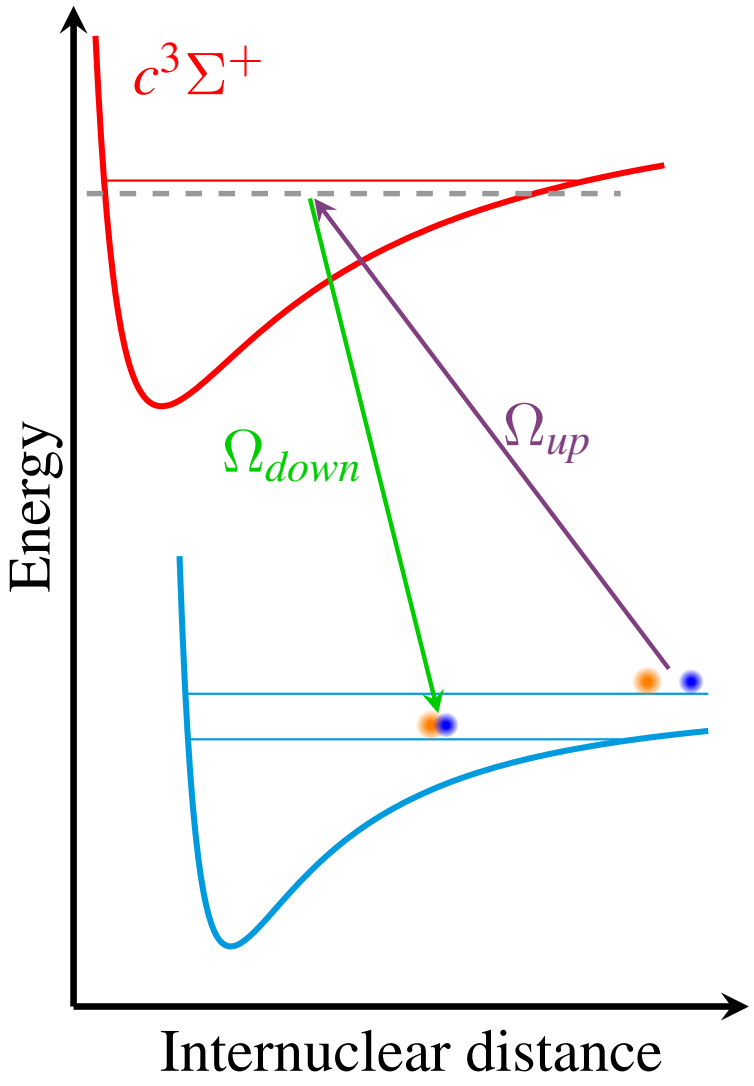
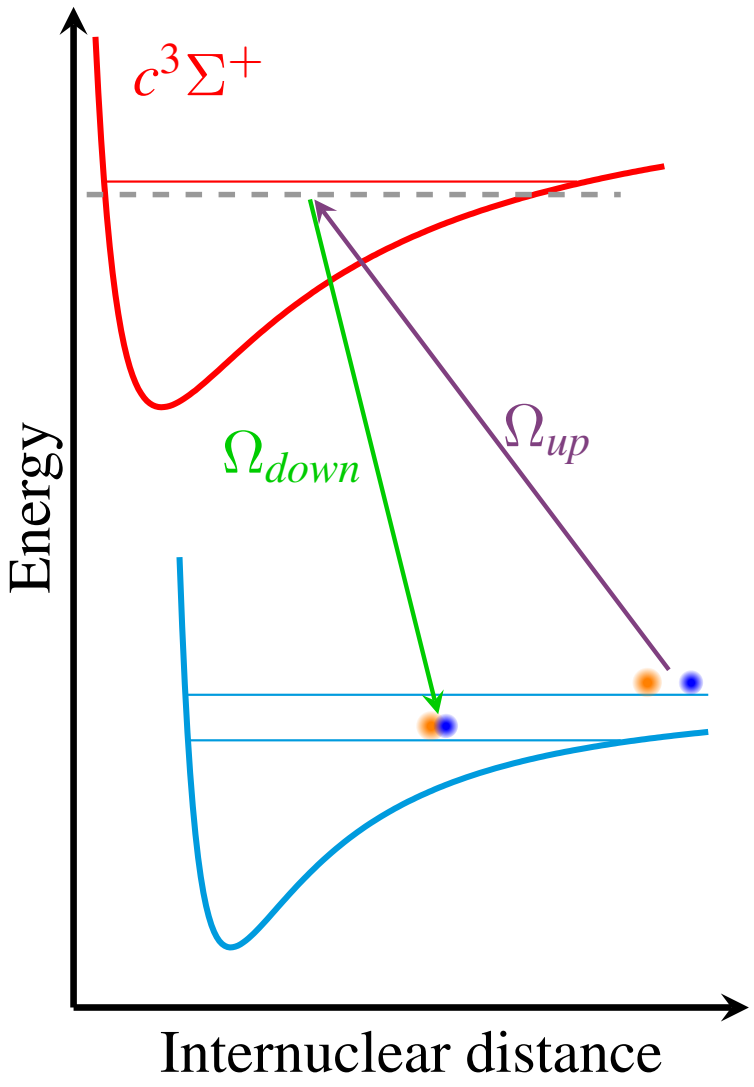


Raman transfer



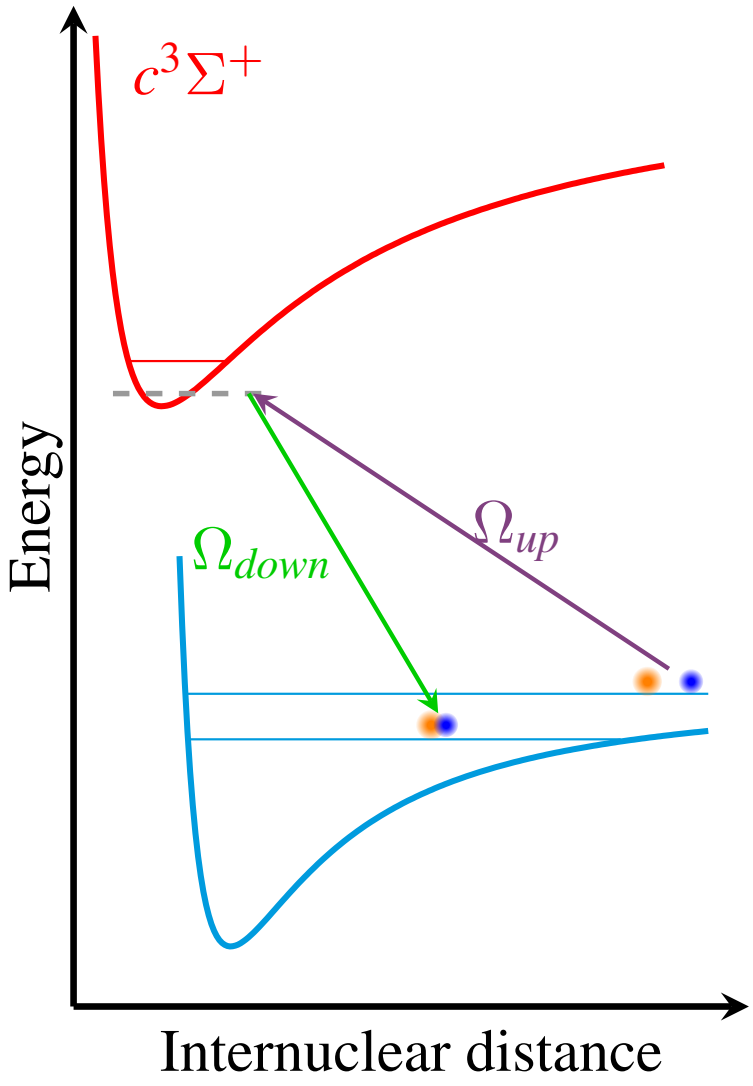
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

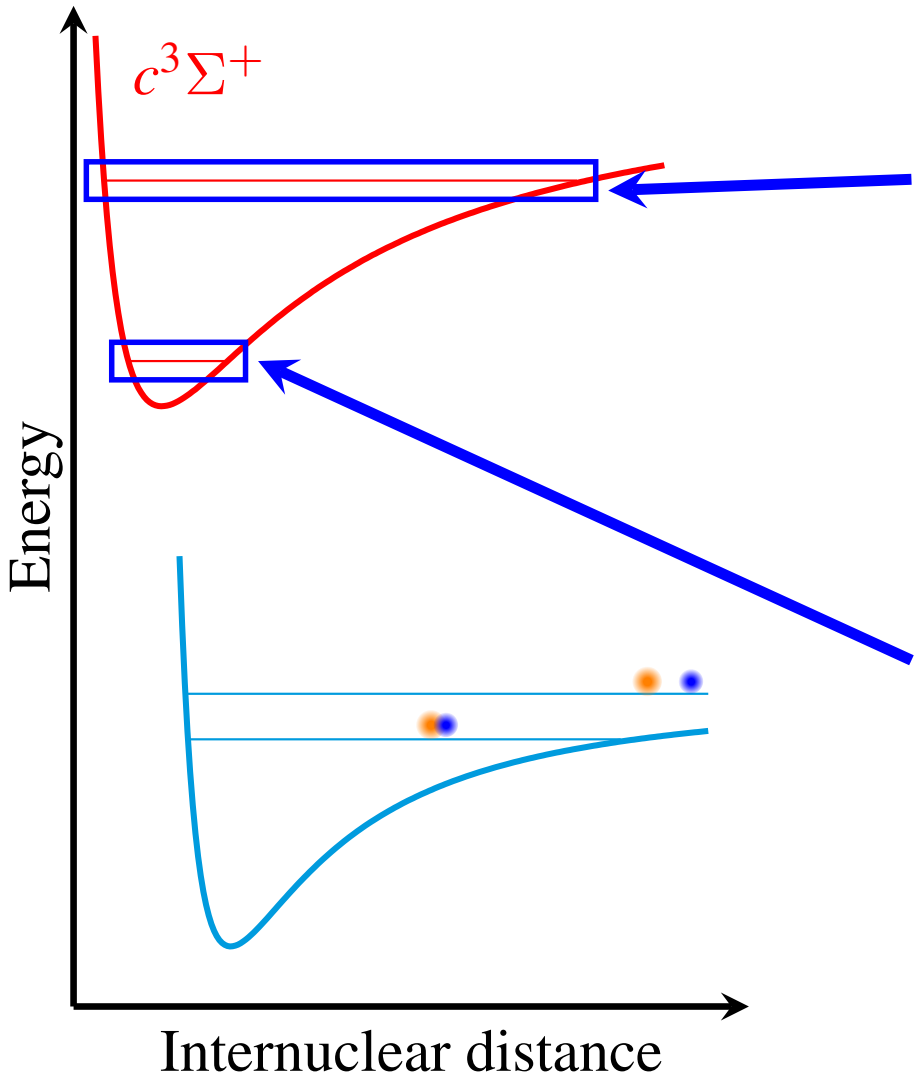
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

Deeply bound states

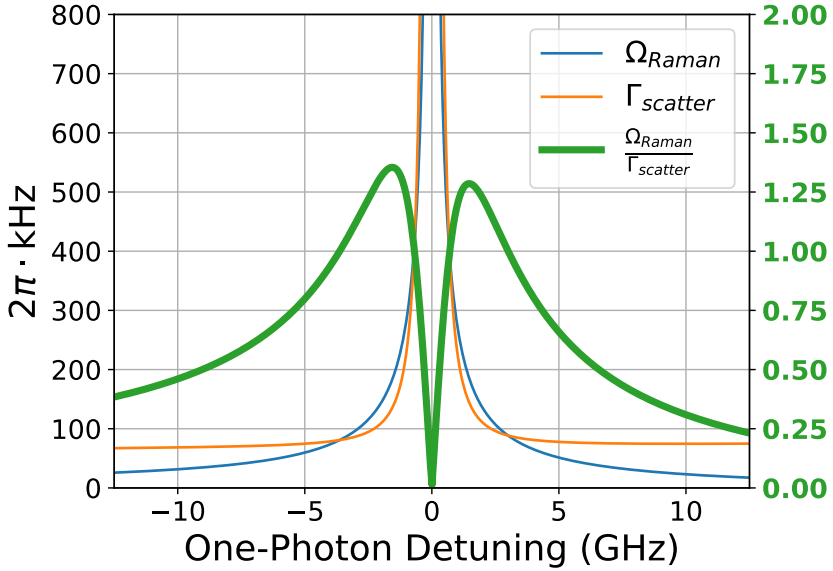
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

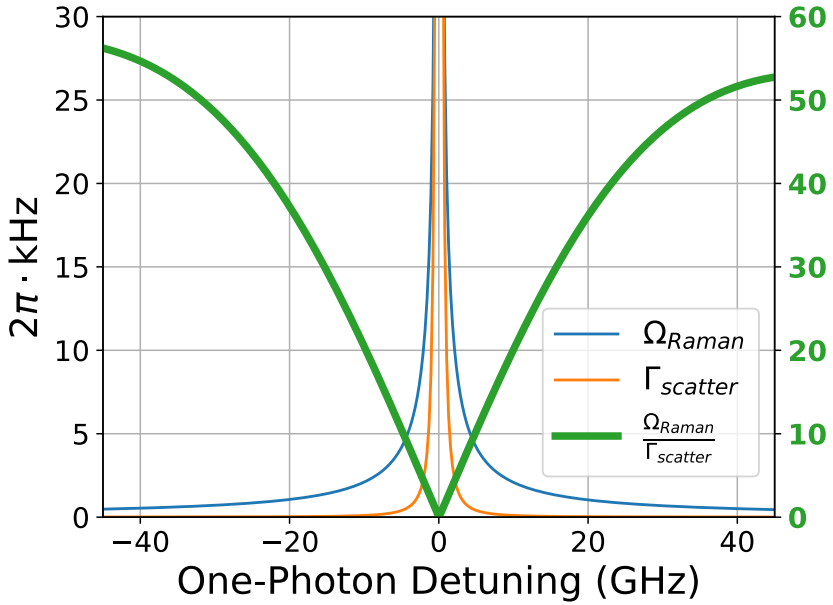
Raman transfer



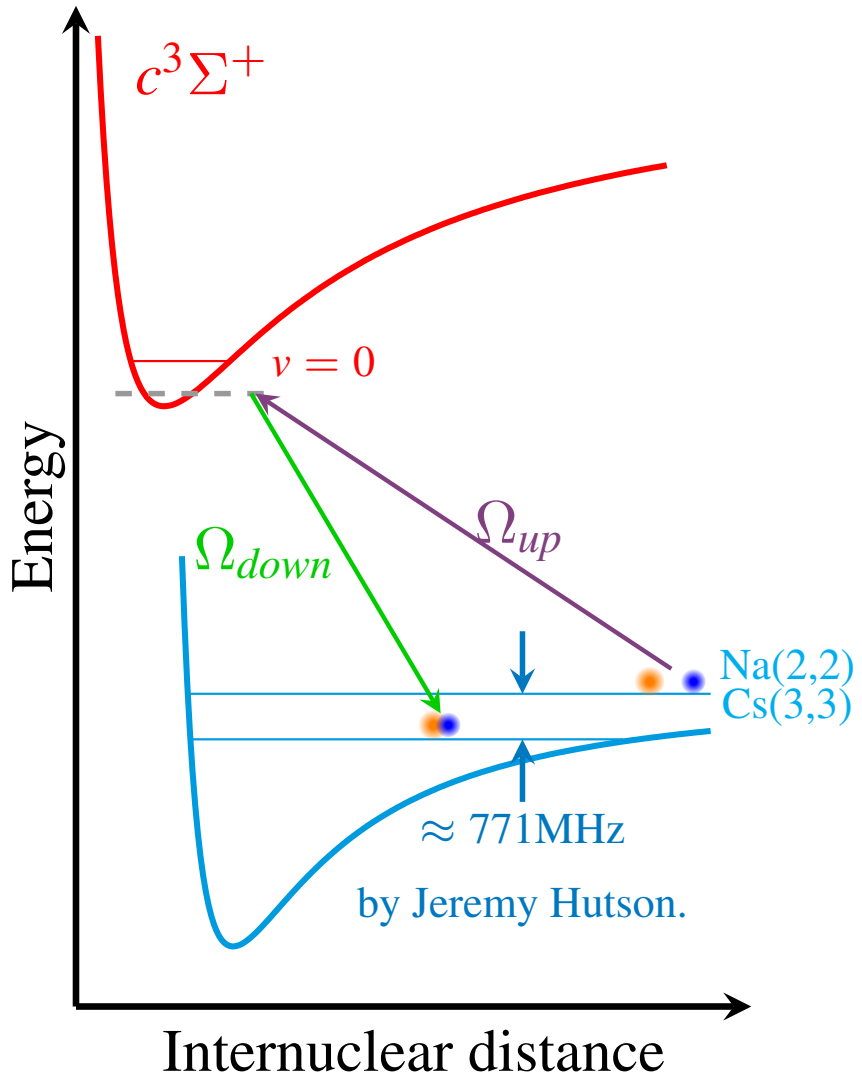
Near threshold states



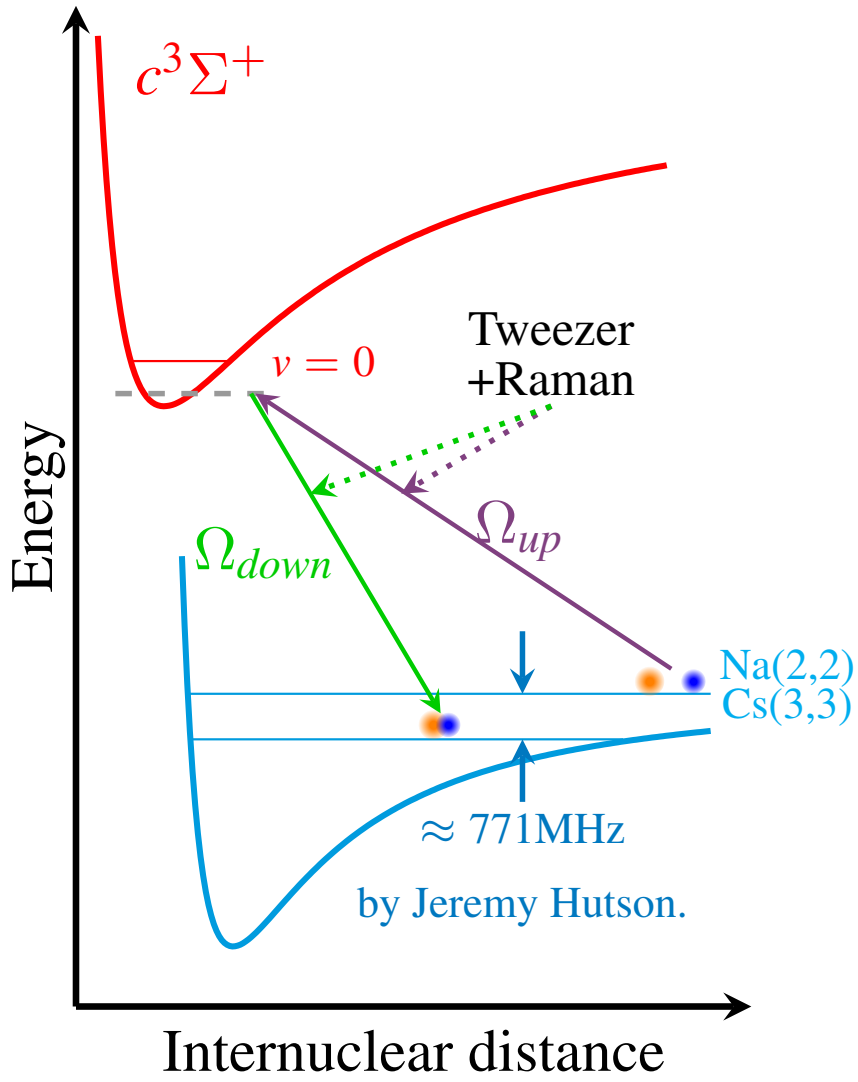
Deeply bound states



Experiment


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

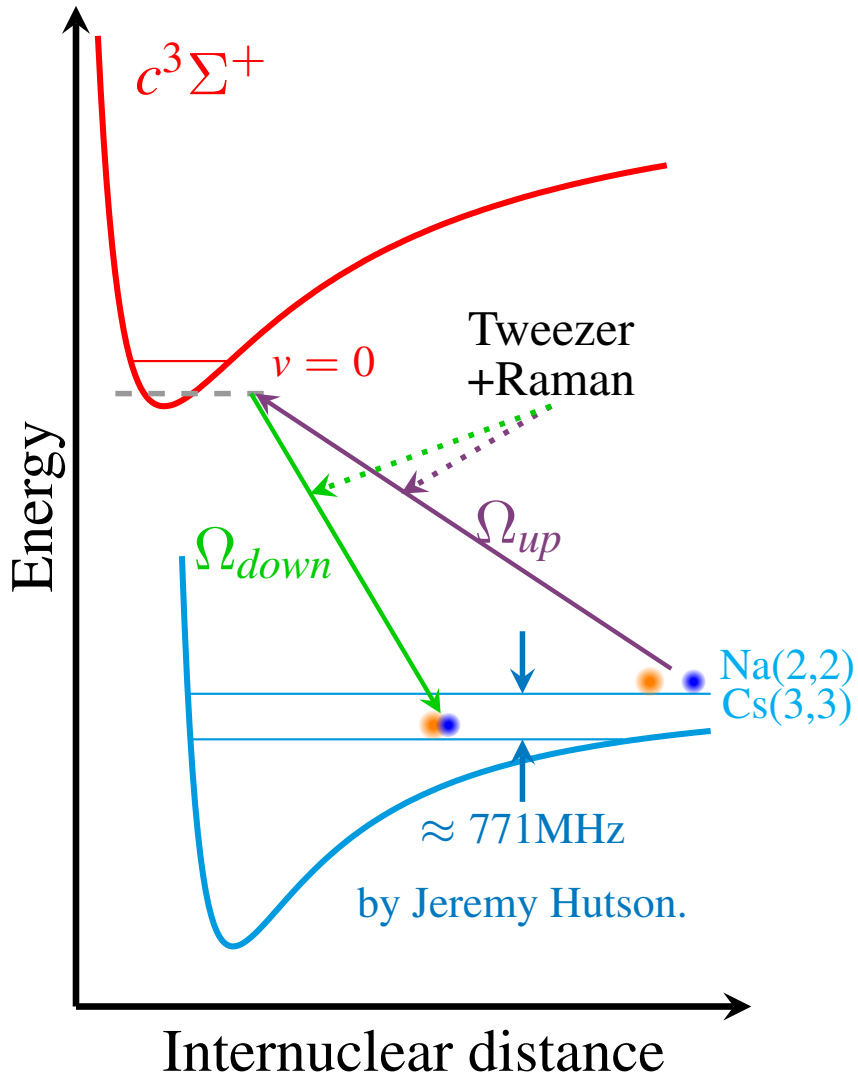
Experiment

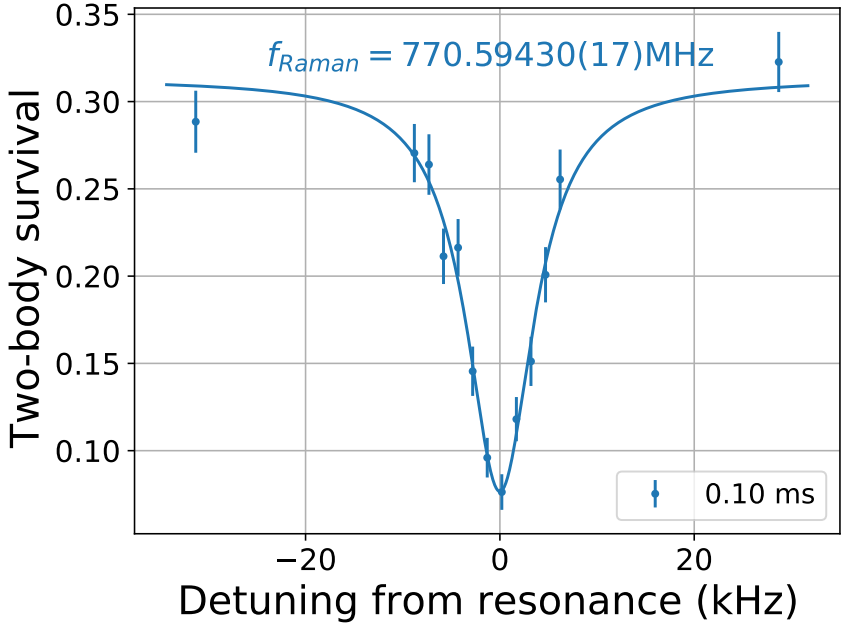


Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

Experiment





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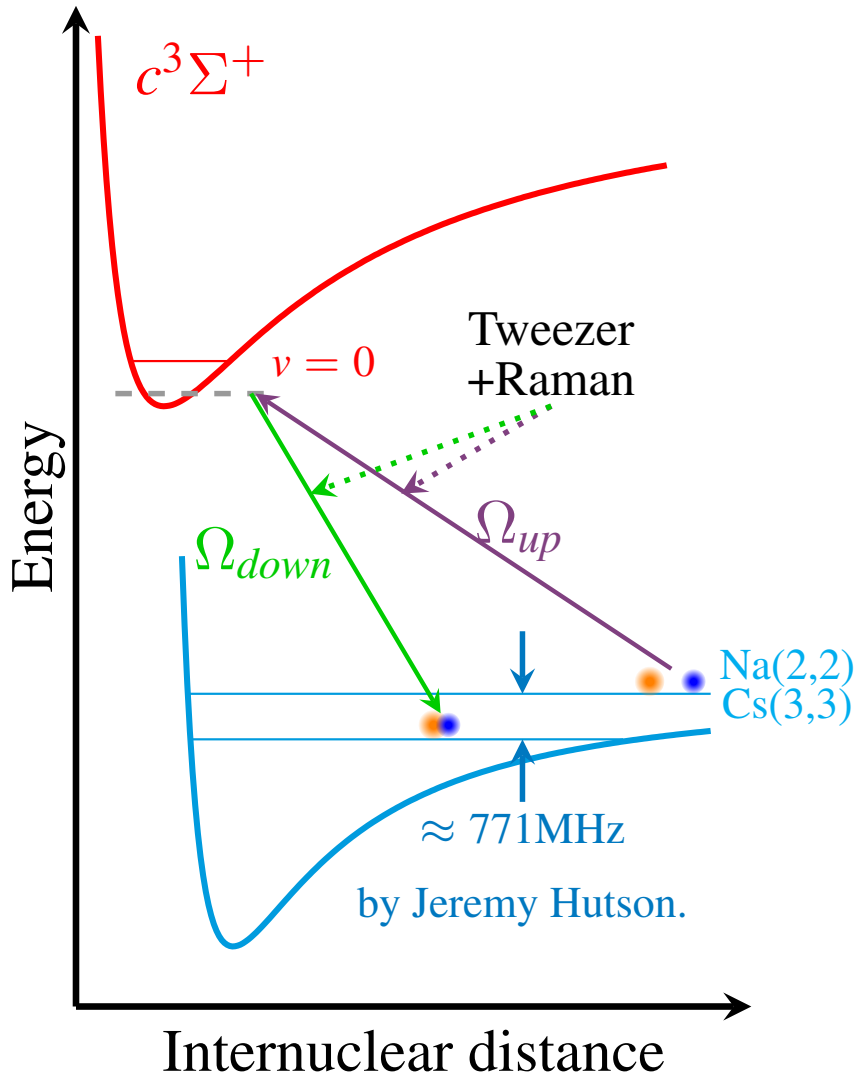

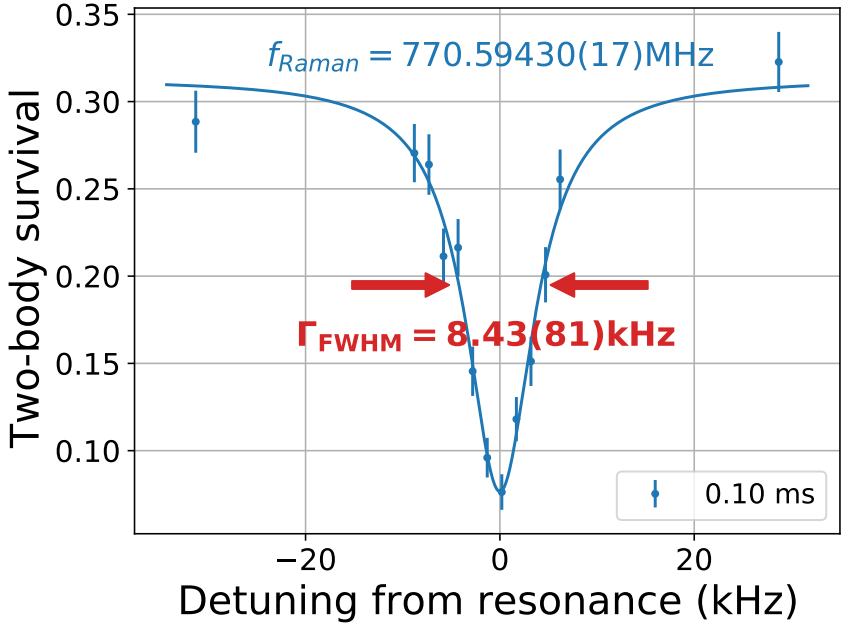

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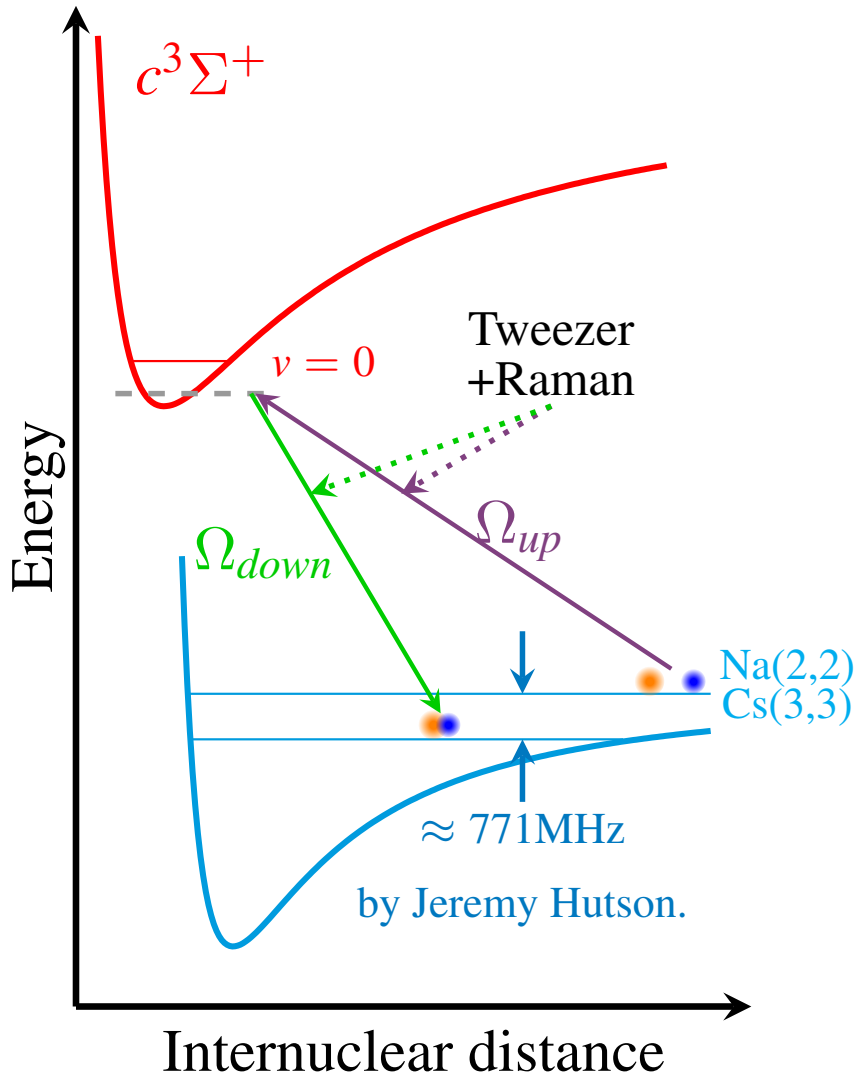
$c^3\Sigma^+$
$v = 0$
Tweezer
+Raman
Ω_{up}
Ω_{down}
Na(2,2)
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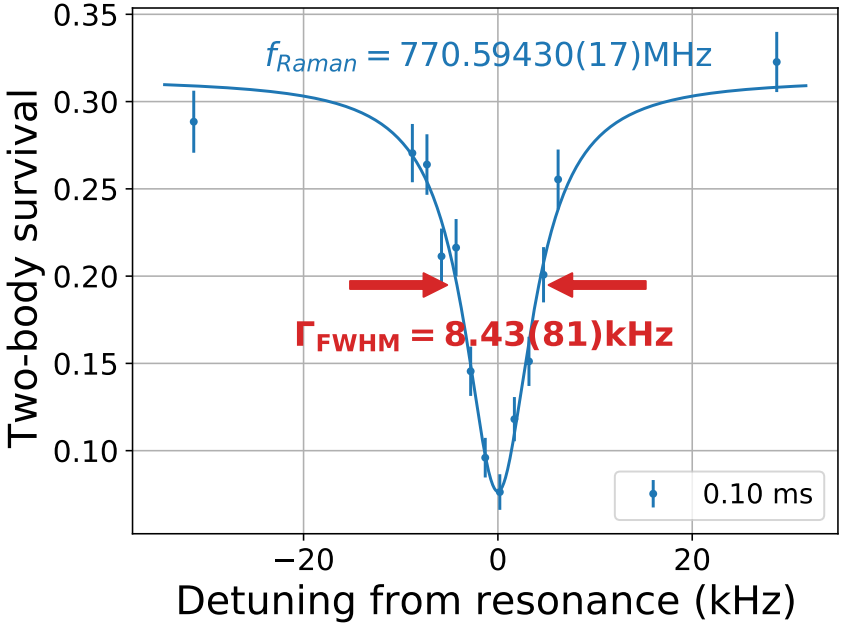
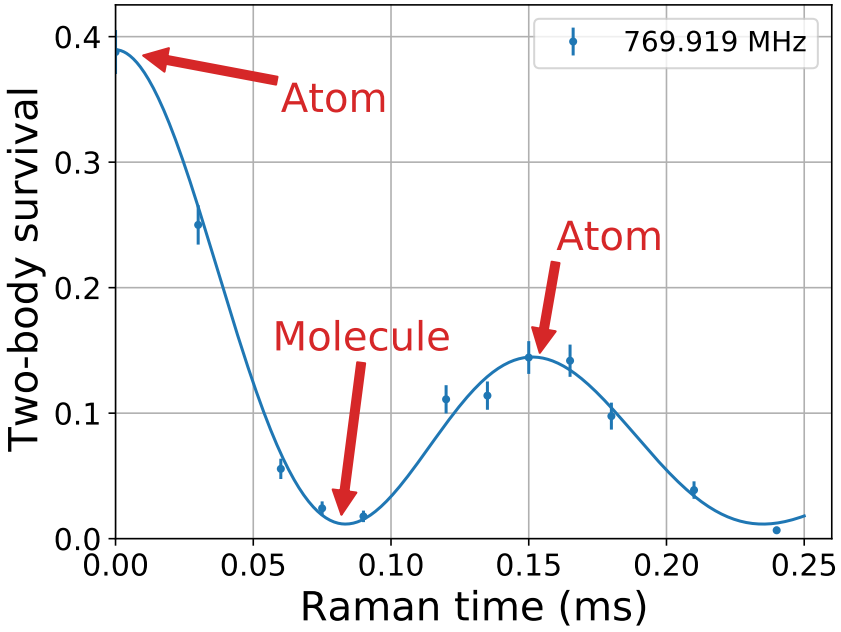
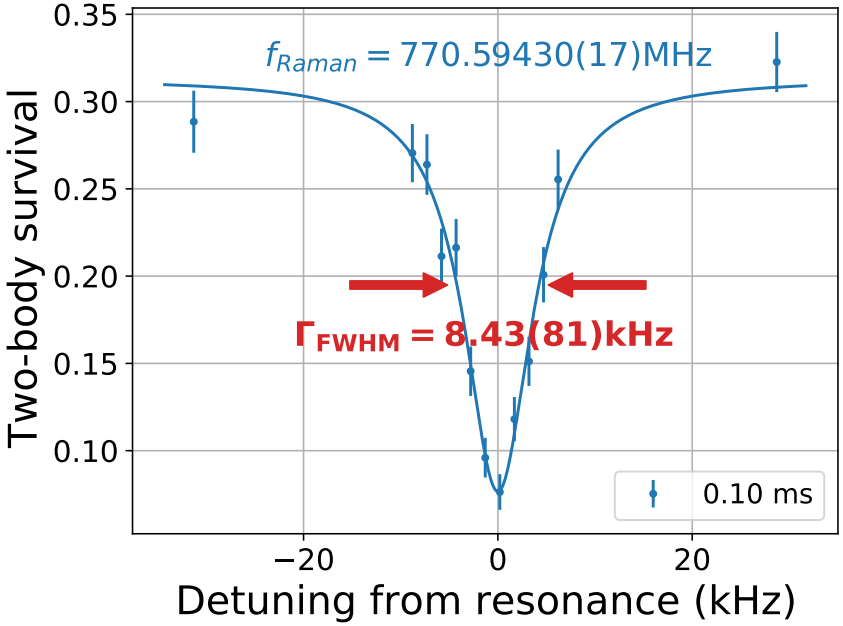
Energy
Internuclear distance

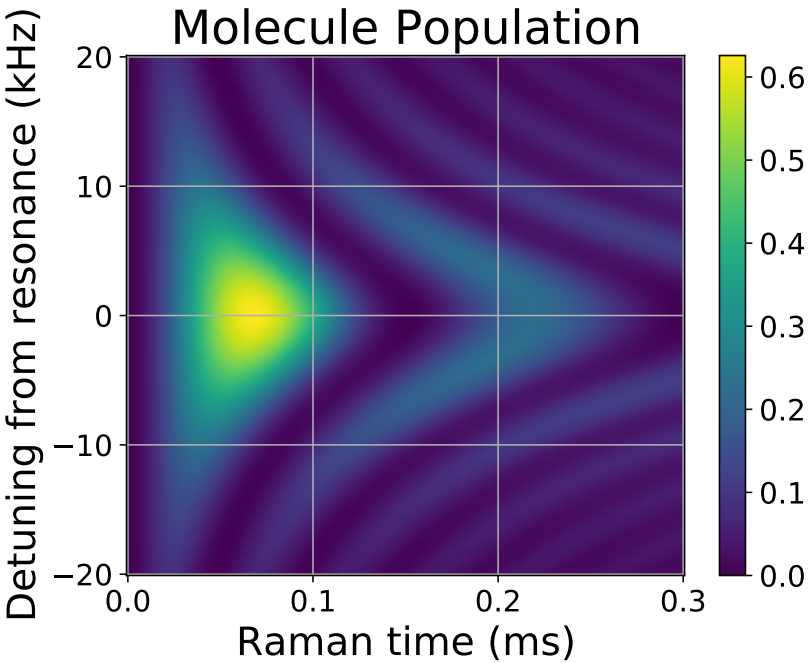

f_{Raman} = 770.59430(17)MHz
Γ_{FWHM} = 8.43(81)kHz
0.10 ms
Two-body survival
Detuning from resonance (kHz)

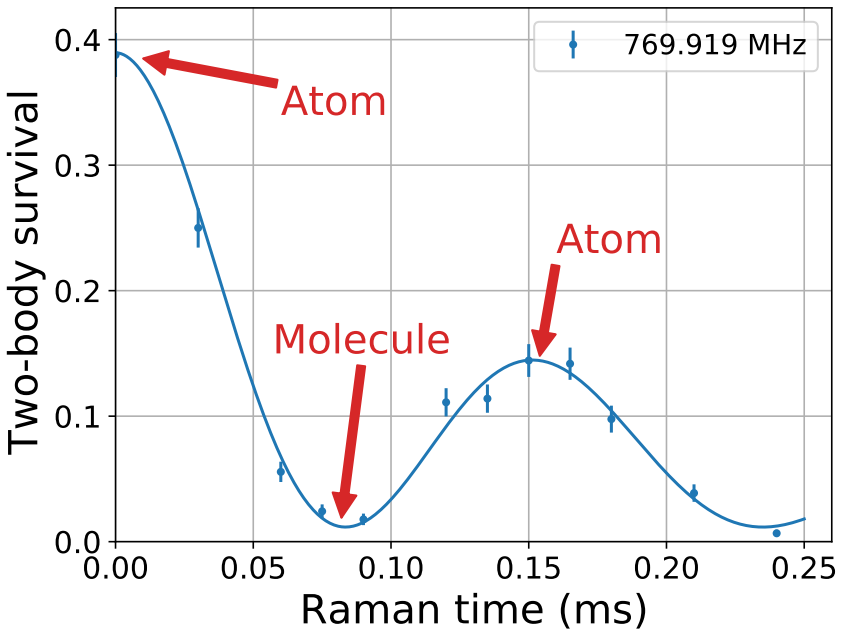

769.919 MHz
Atom
Atom
Molecule
Two-body survival
Raman time (ms)

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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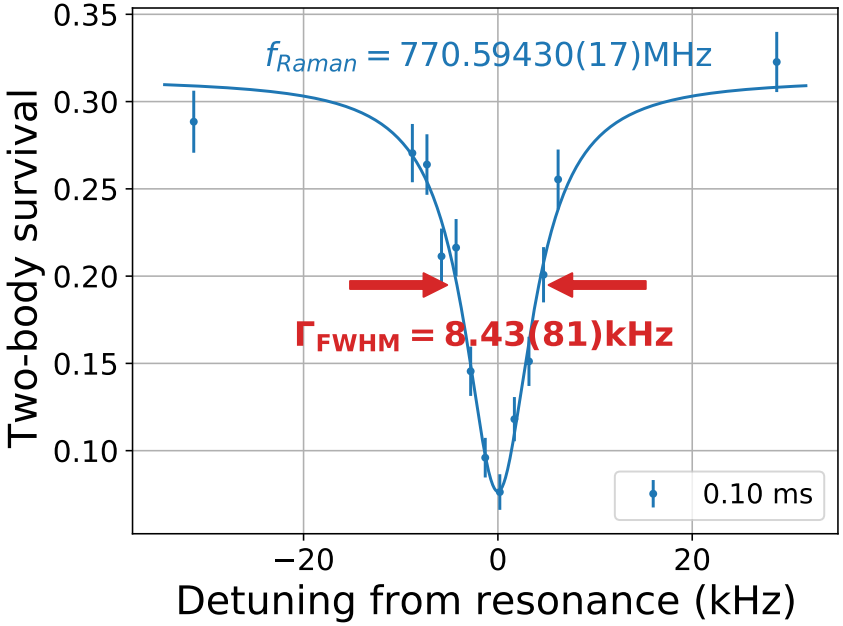

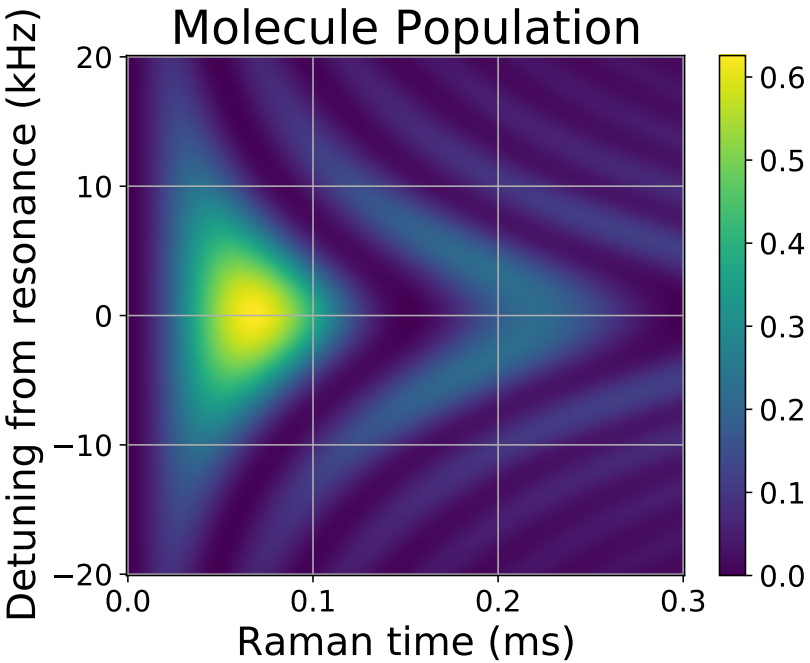

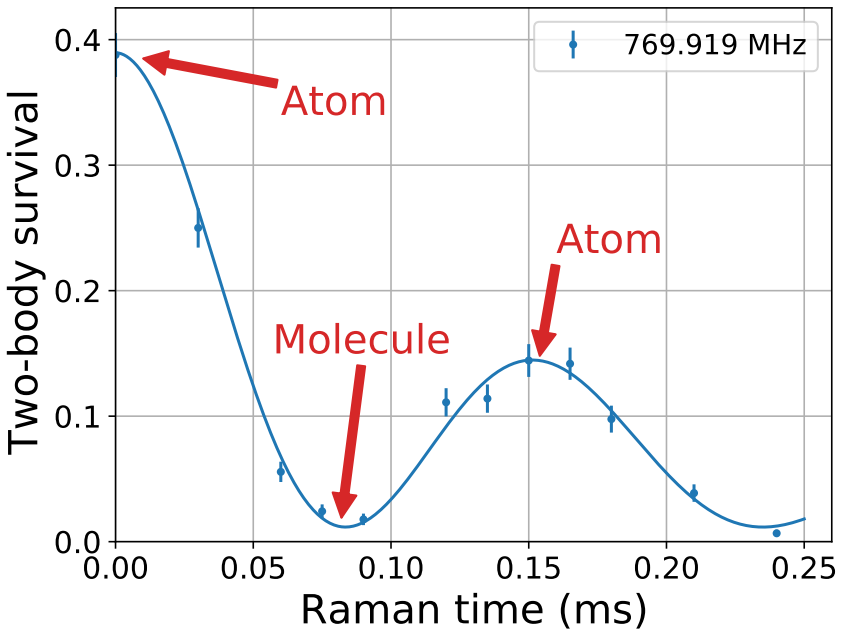

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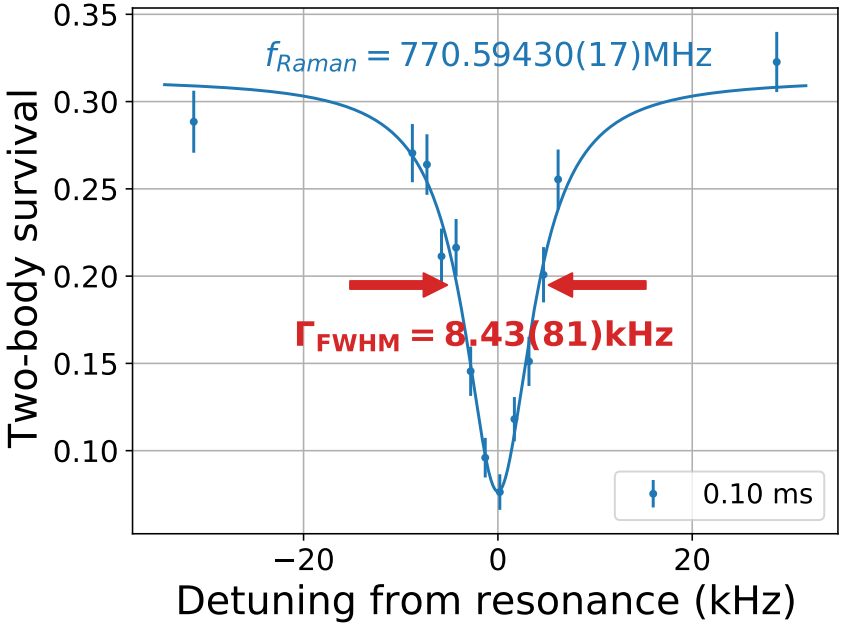

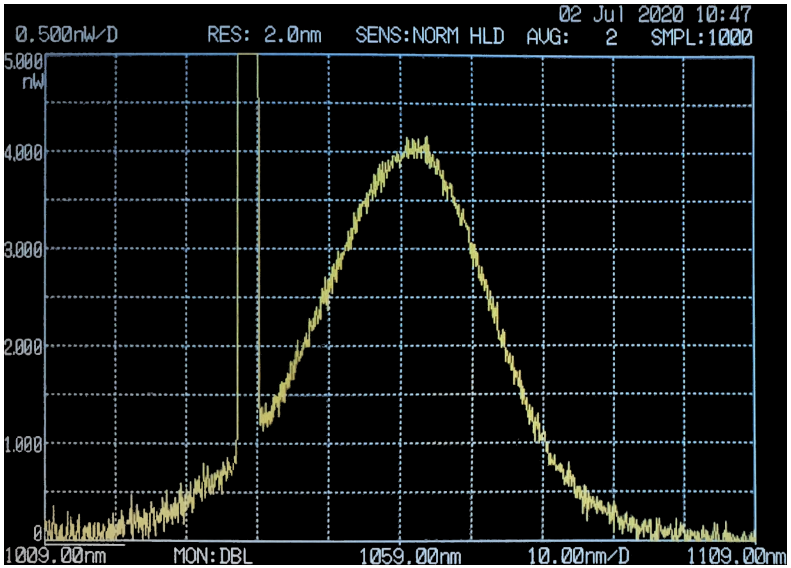

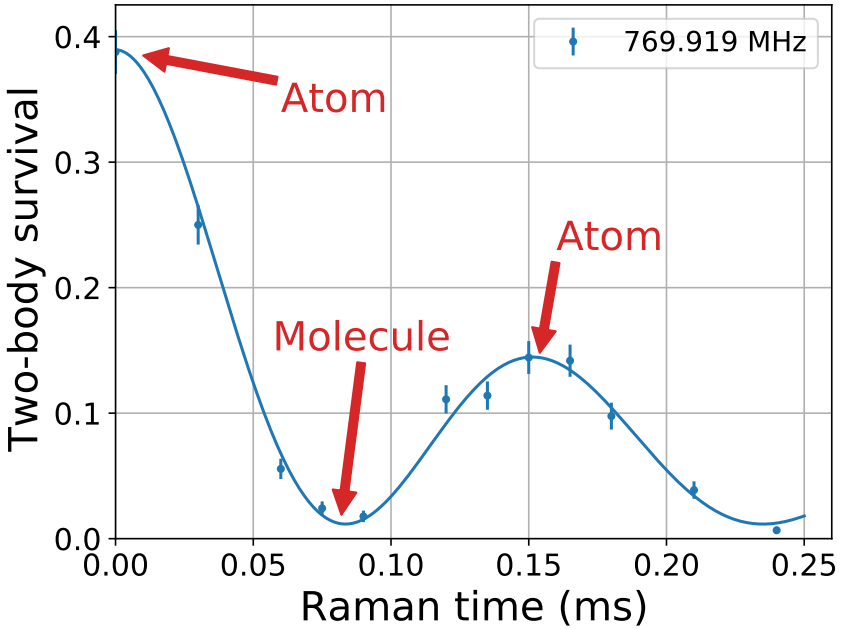

Conclusion and outlook

- 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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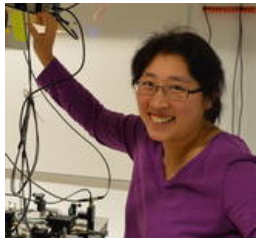
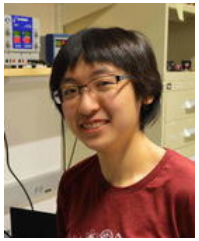
Experiment



Kang-Kuen Ni

Kenneth Wang



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Lewis Picard

William Cairncross

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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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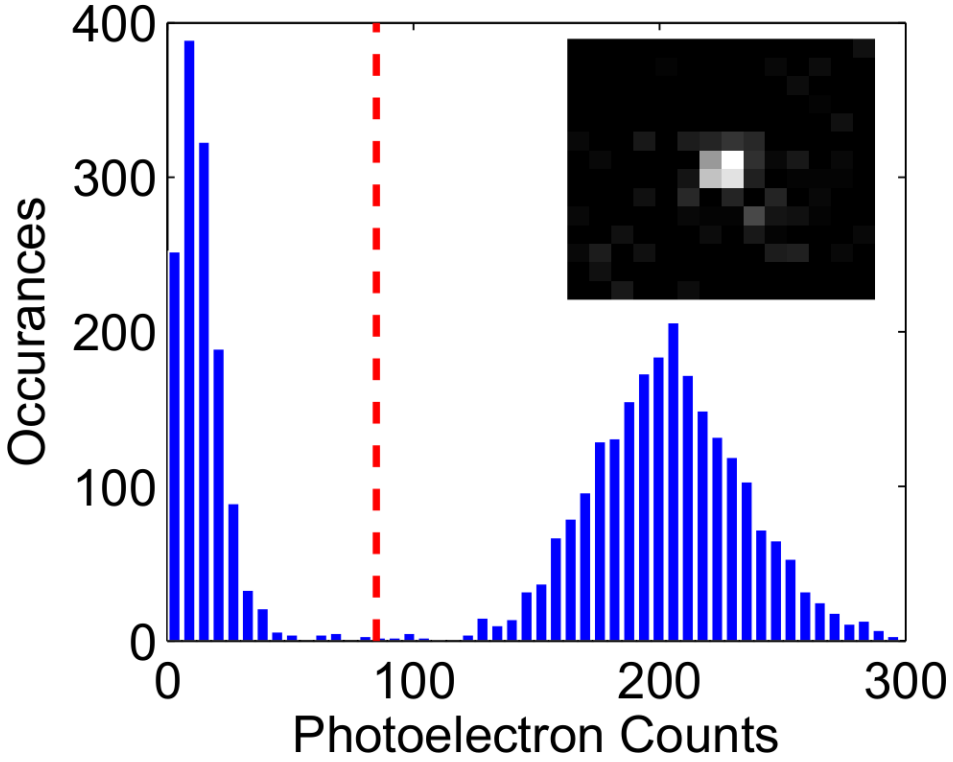

Single Atom in Tweezer

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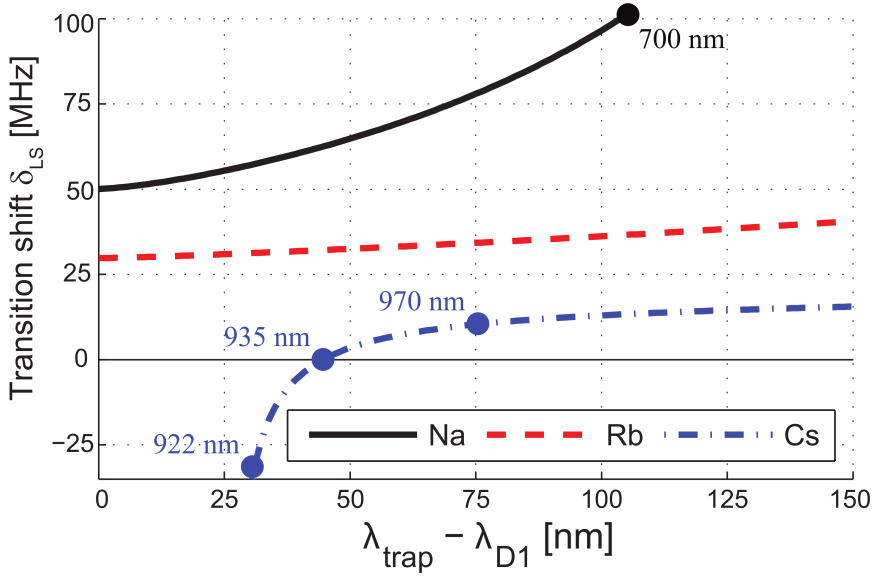
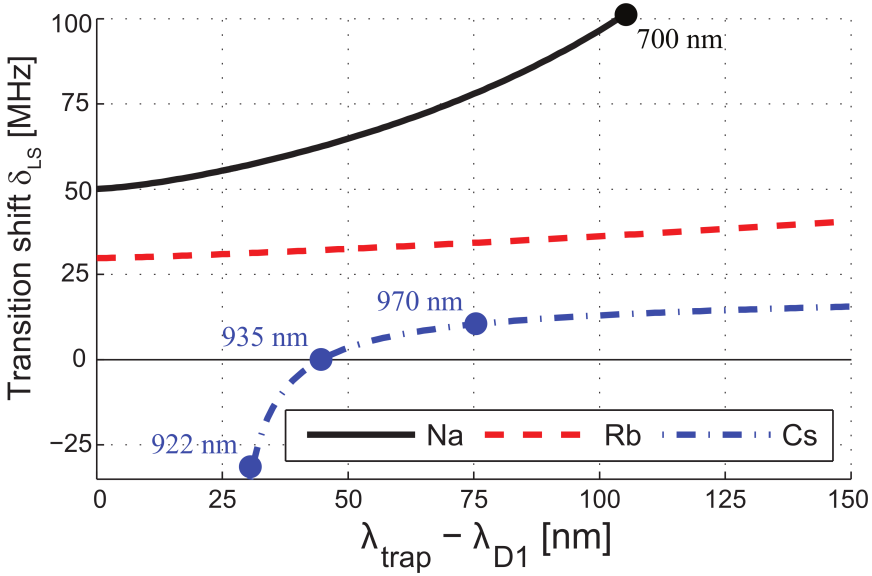
Issues with Na

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

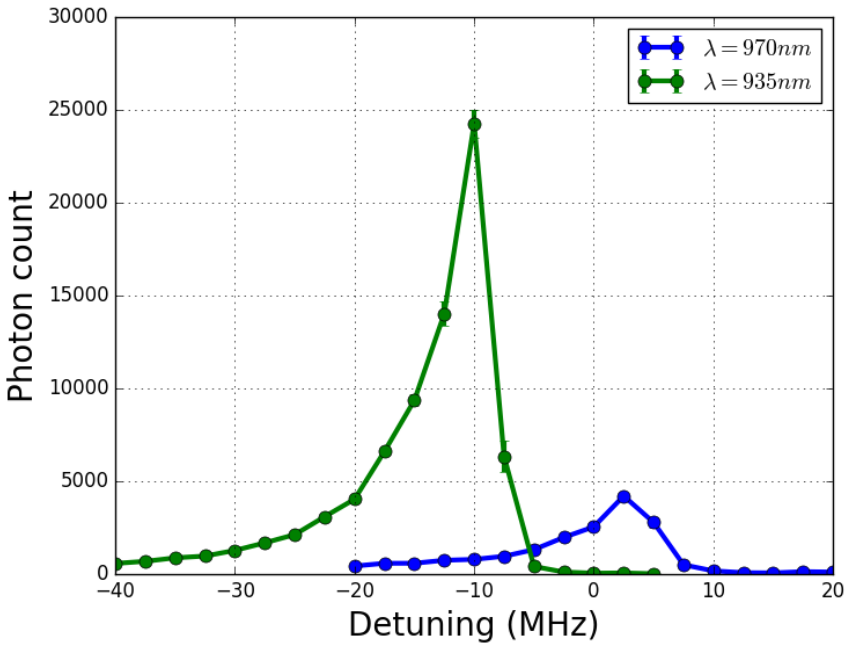
Real Issue with Na: Light Shift



Real Issue with Na: Light Shift



Cs single atom imaging



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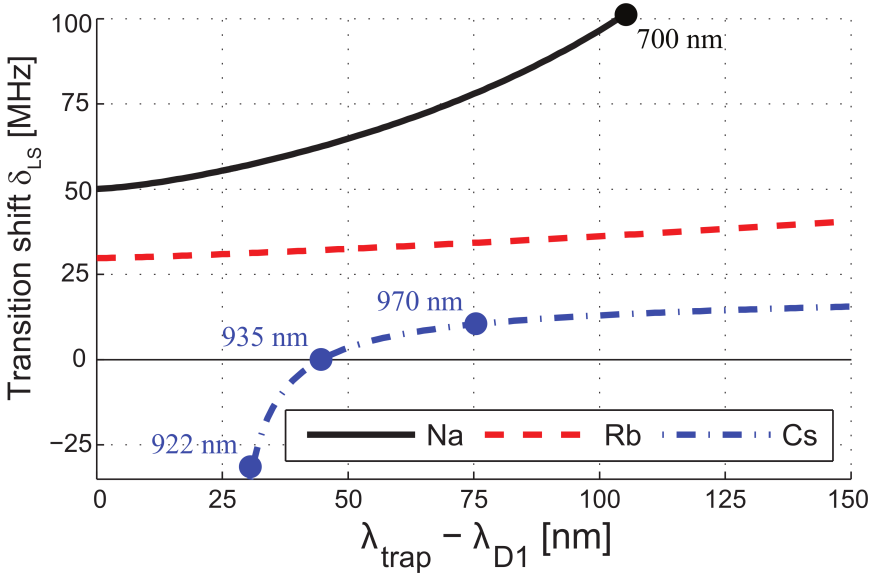

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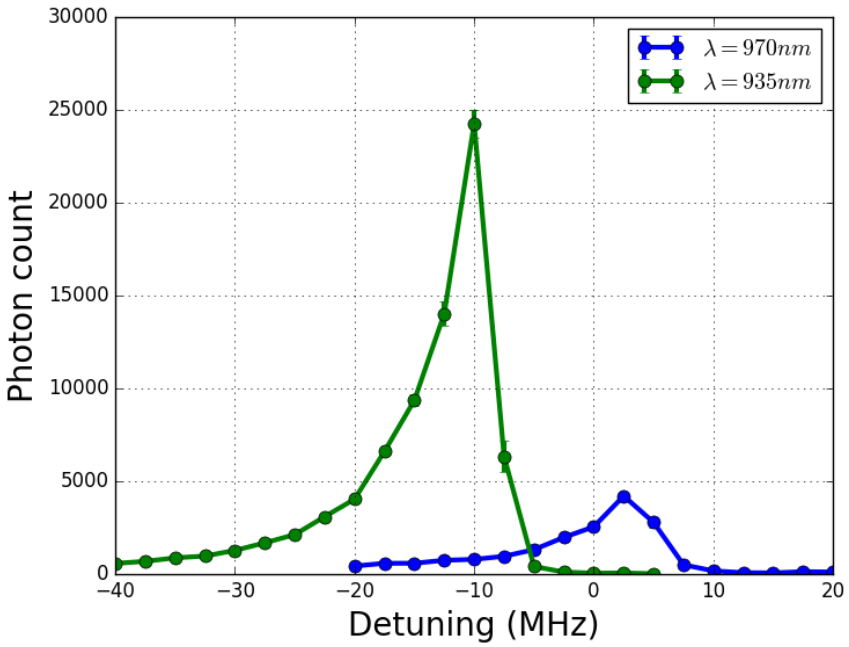


- Low imaging signal
- No cooling in tweezer

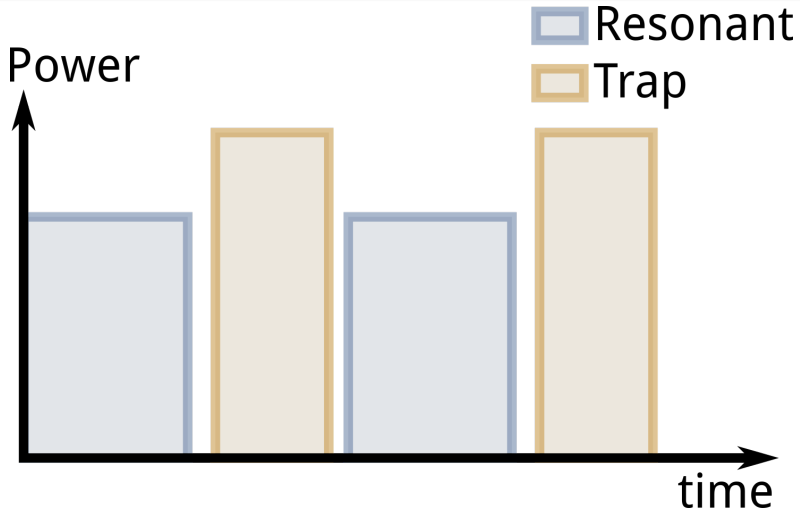
Real Issue with Na: Light Shift

Trap modulation

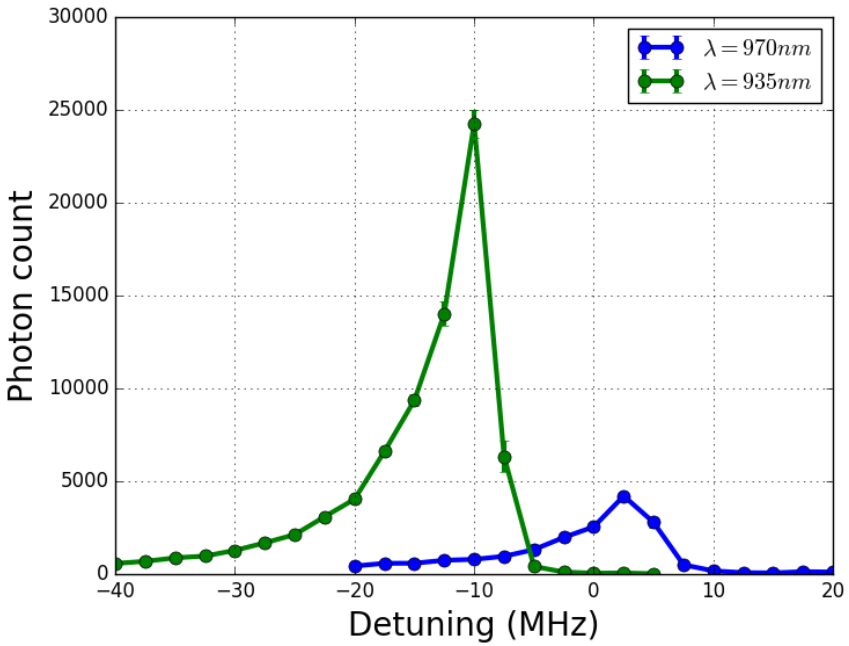
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



Cs single atom imaging



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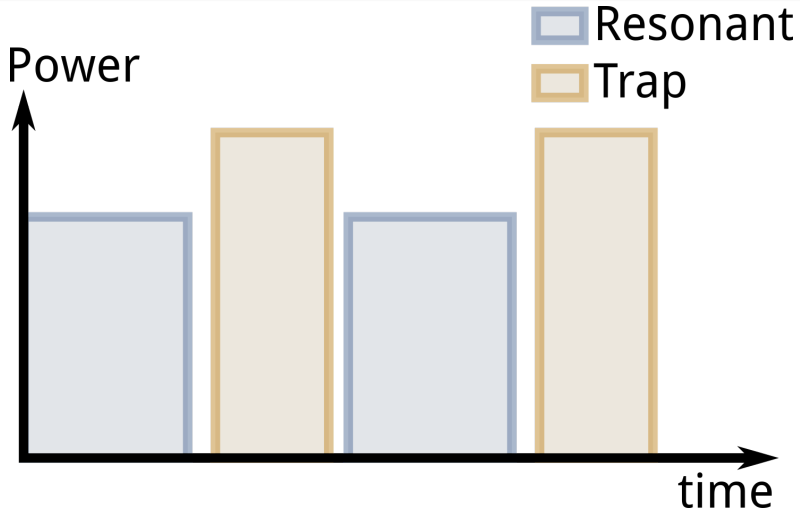

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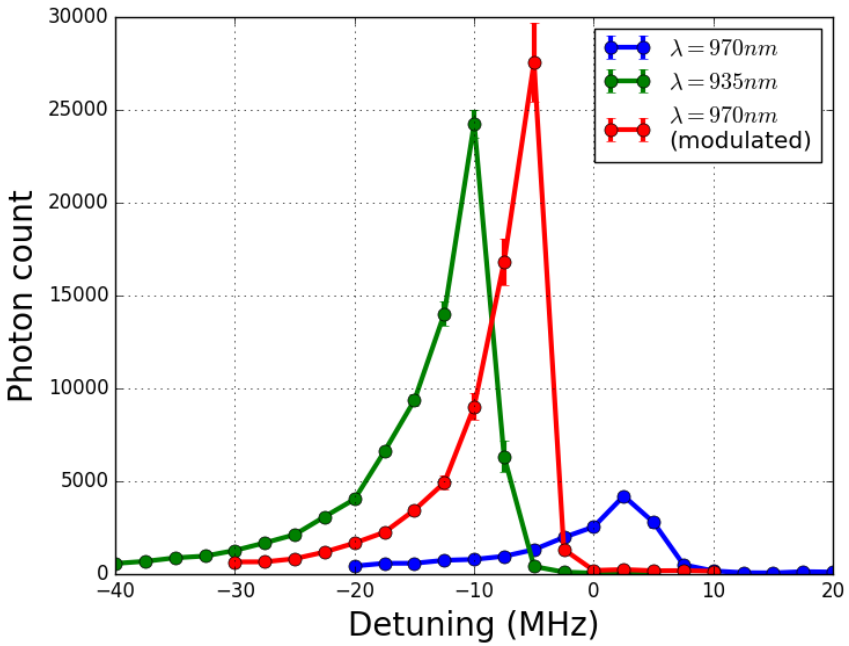

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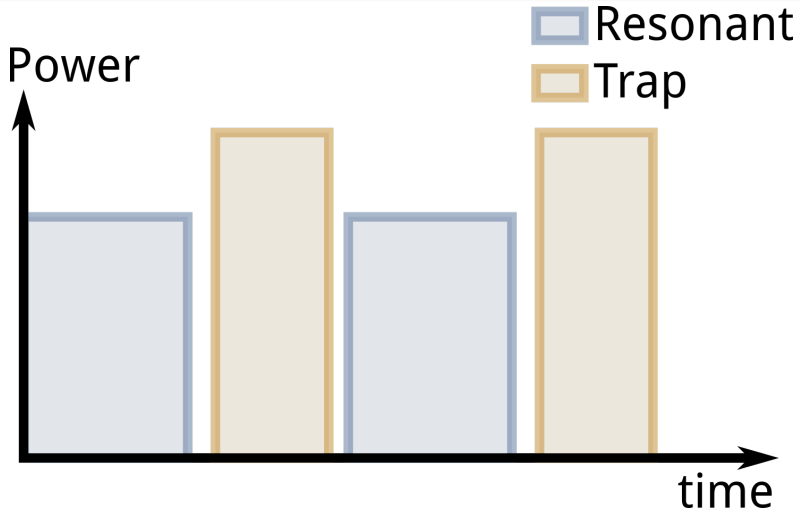

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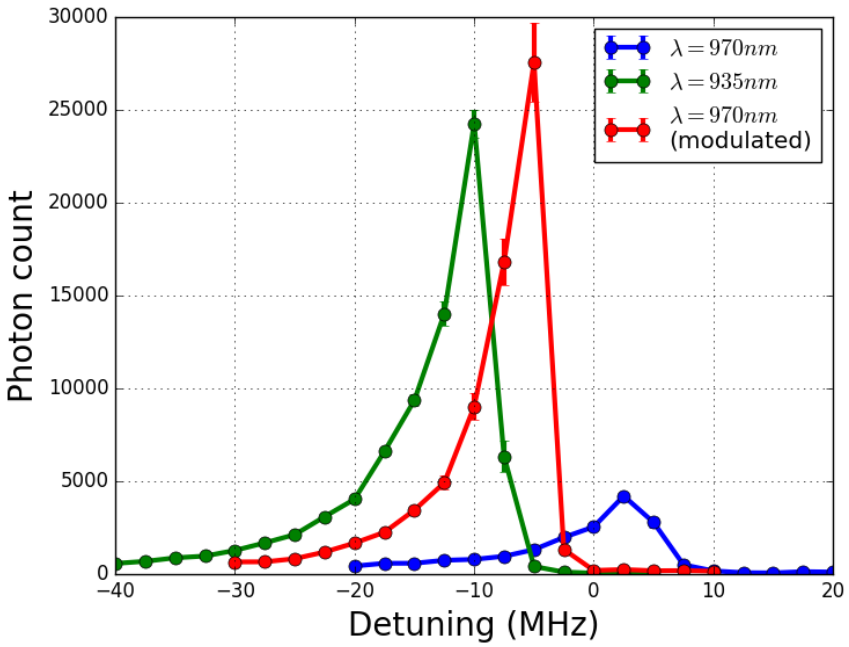
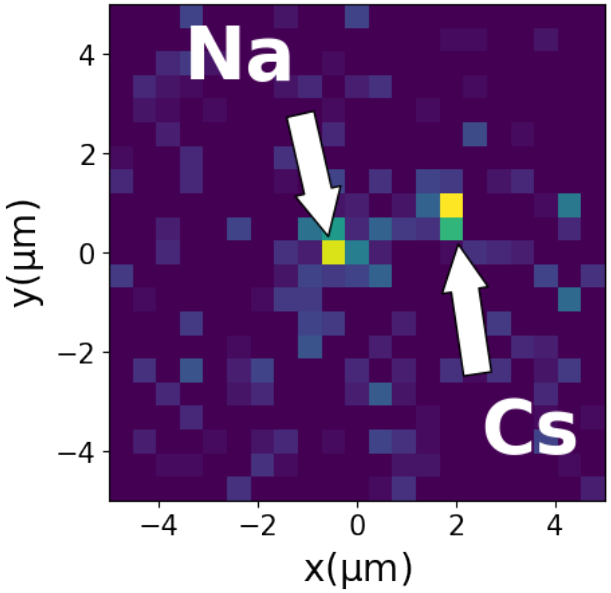
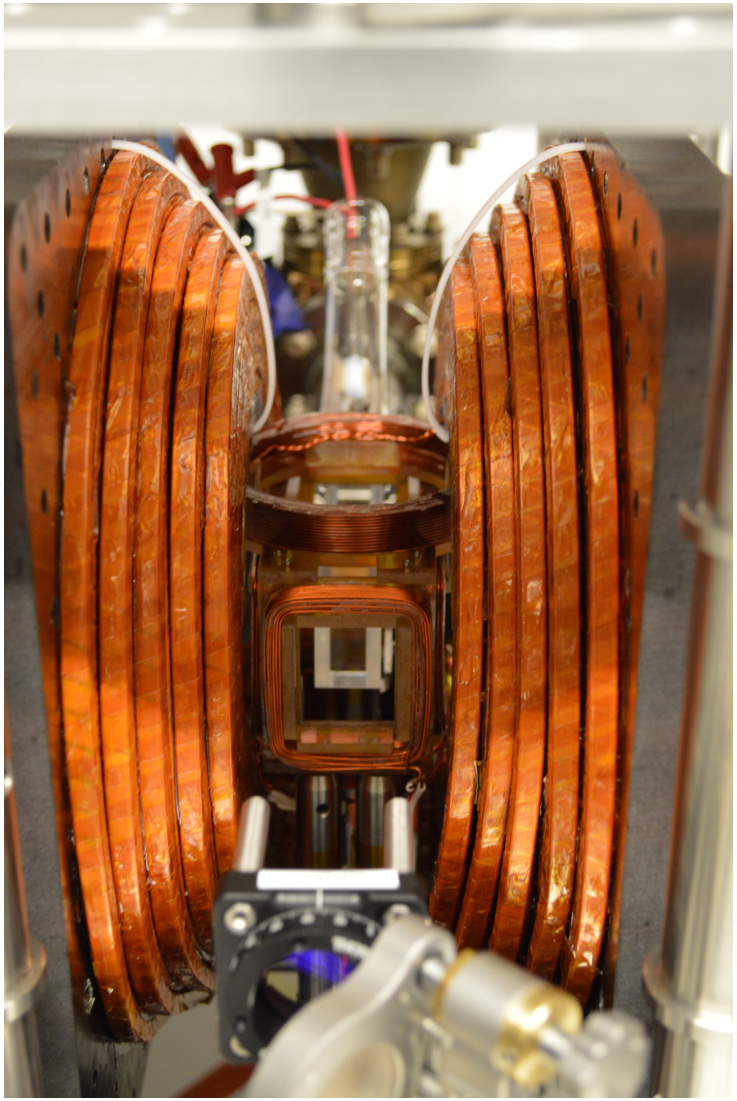




Na (1, -1) Cs (3, -3) Feshbach resonance



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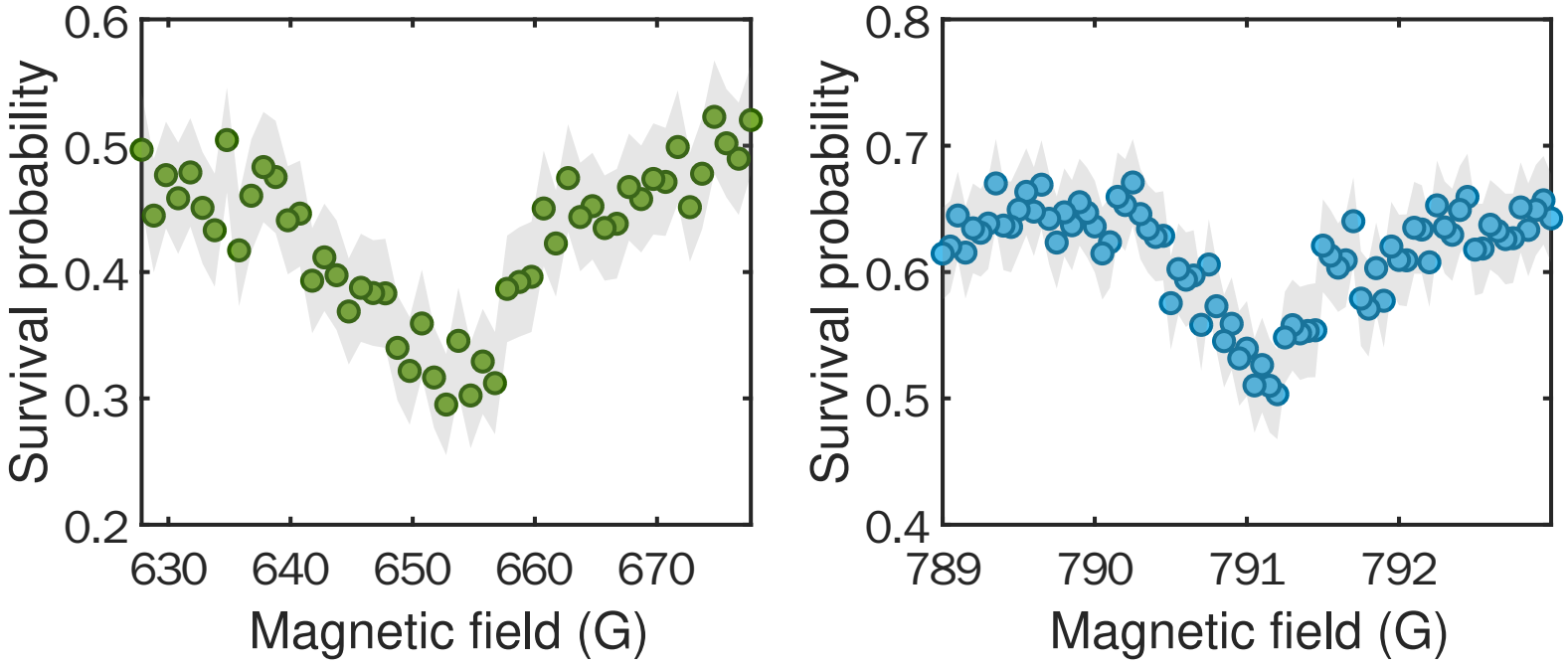

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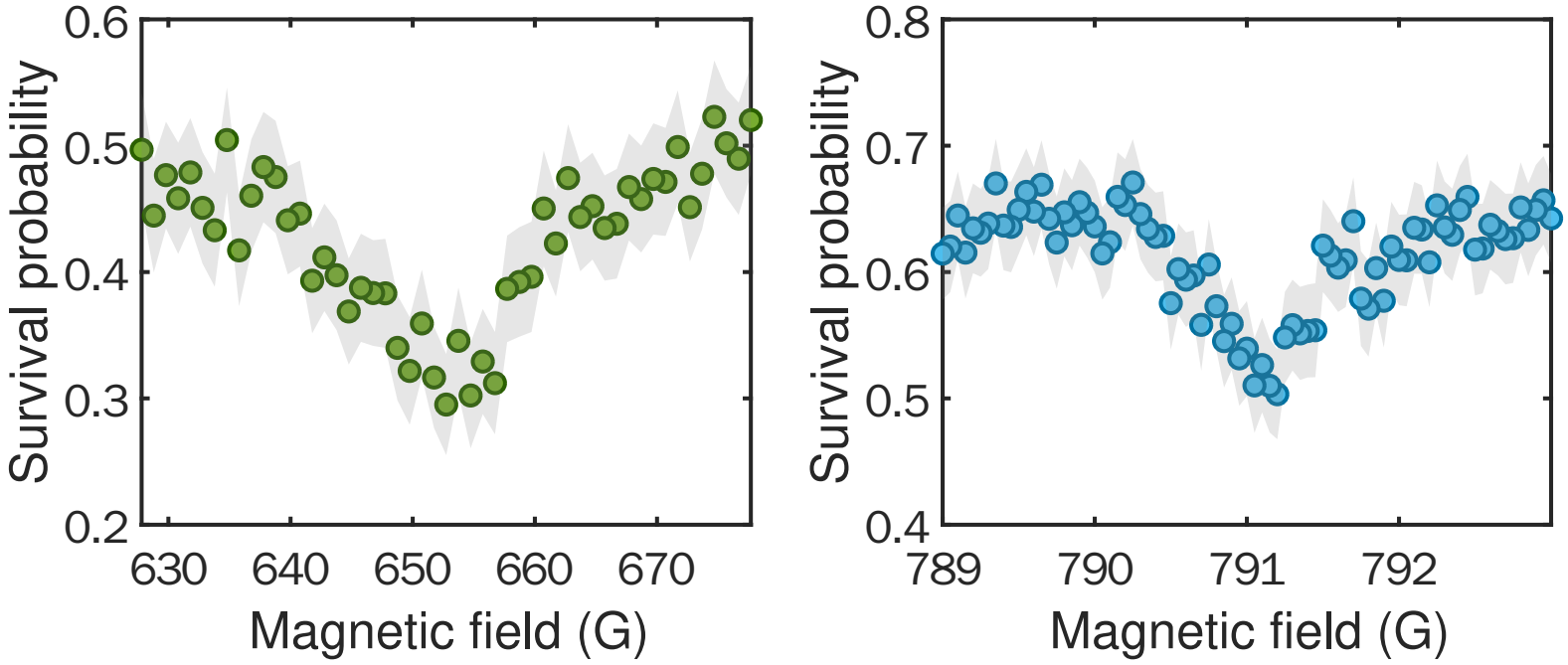
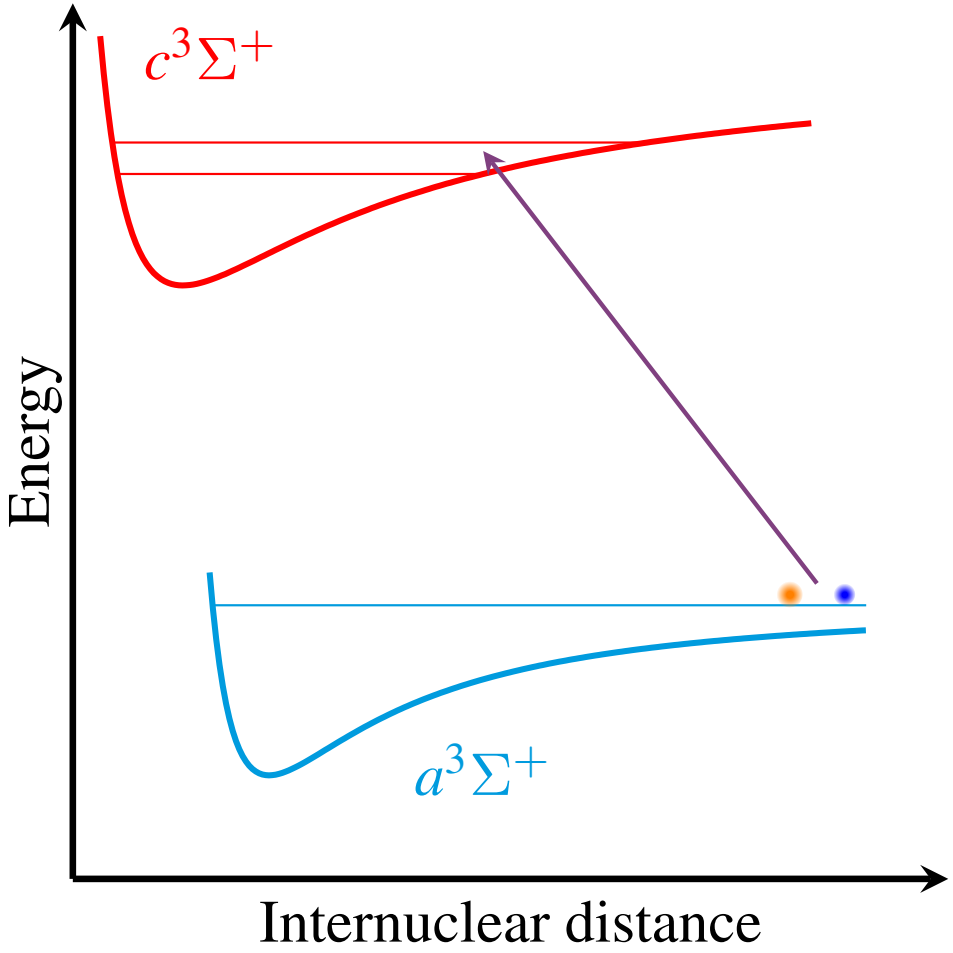


	s-wave	p-wave
Predicted (based on interaction shift)[1]	663 G	799 G
Measured	652(3) G	791.2(2) G

[1] In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



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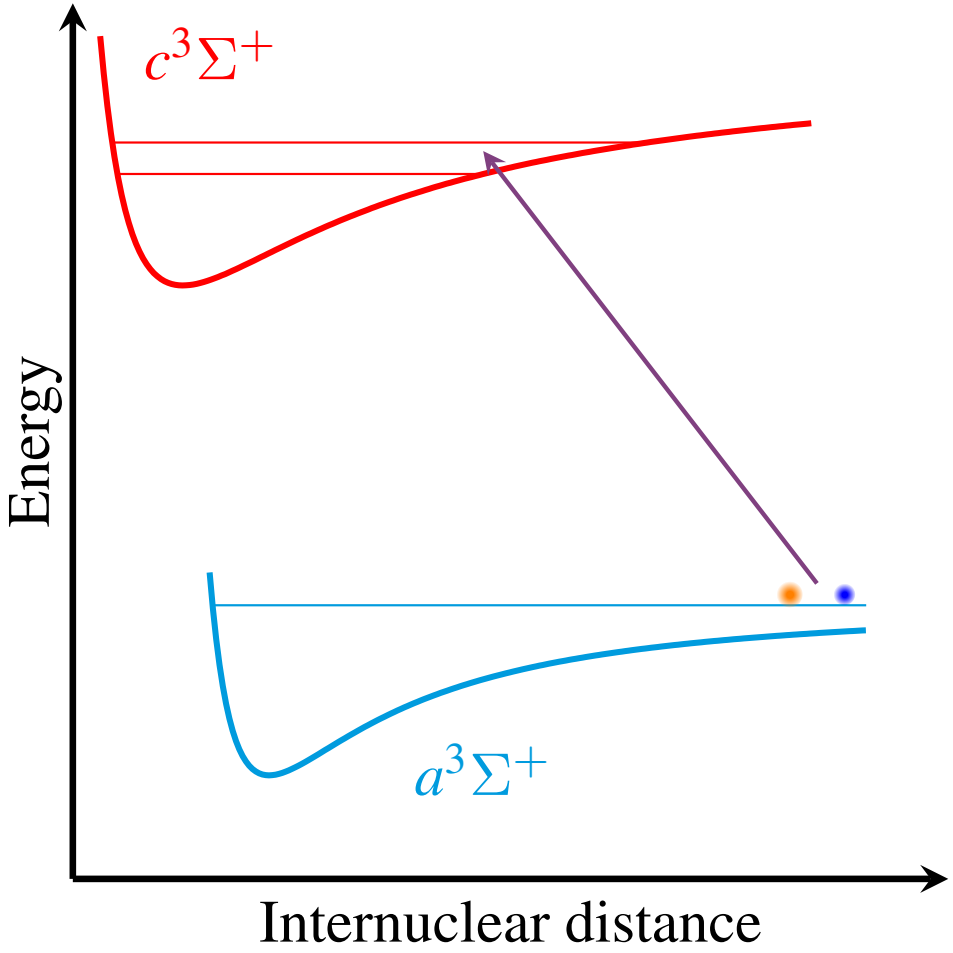

Single Atom PA

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

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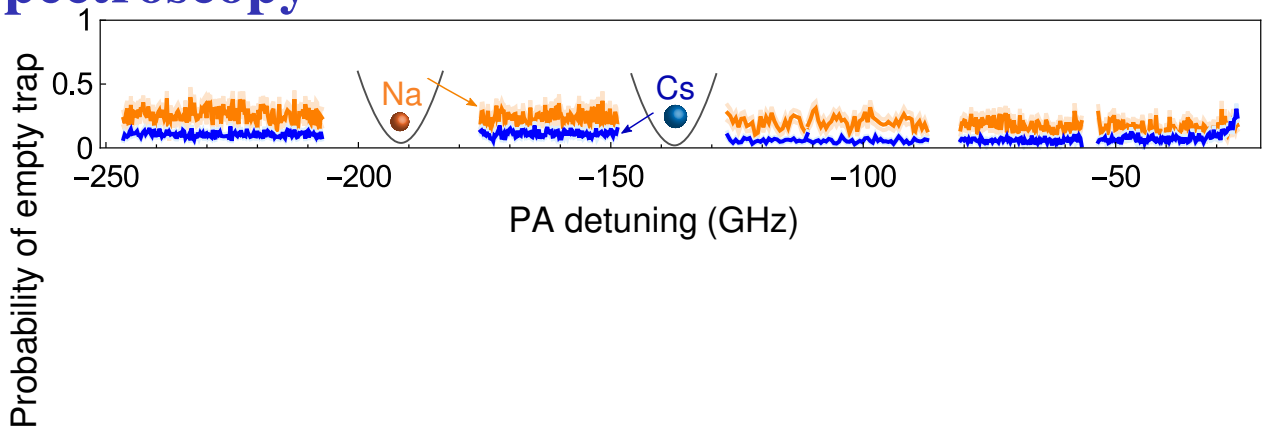

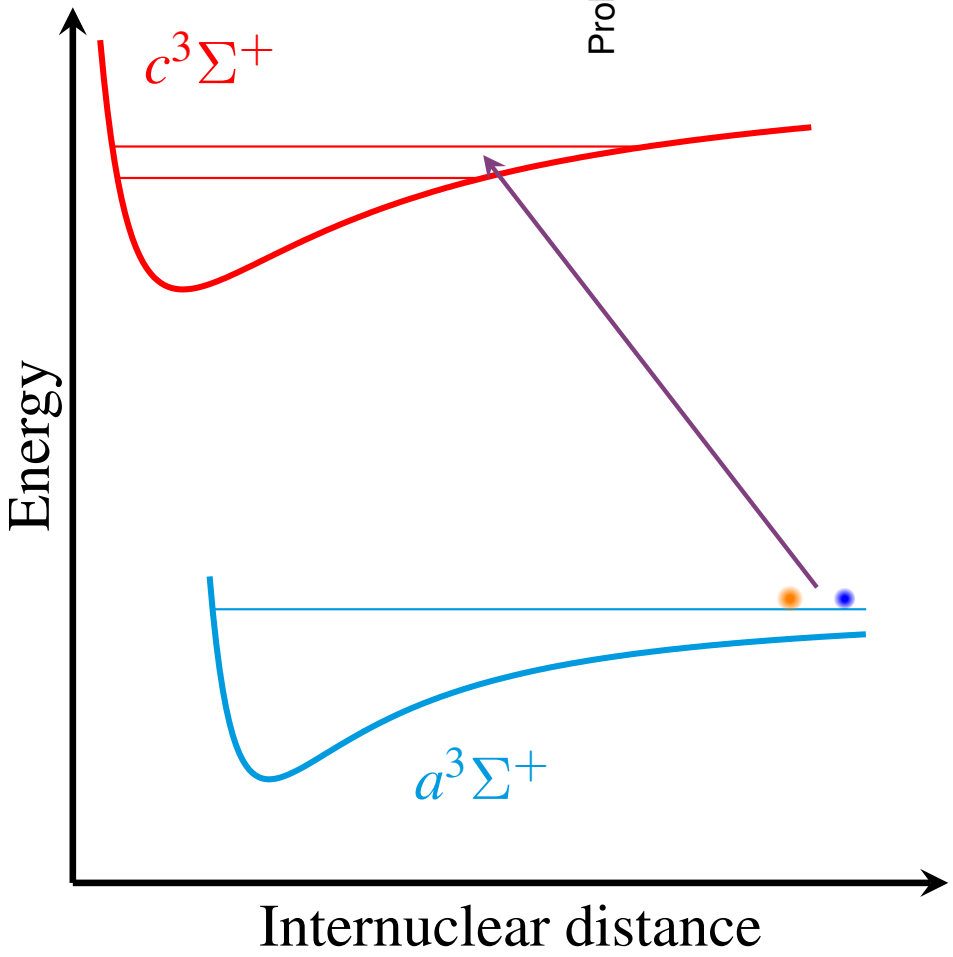

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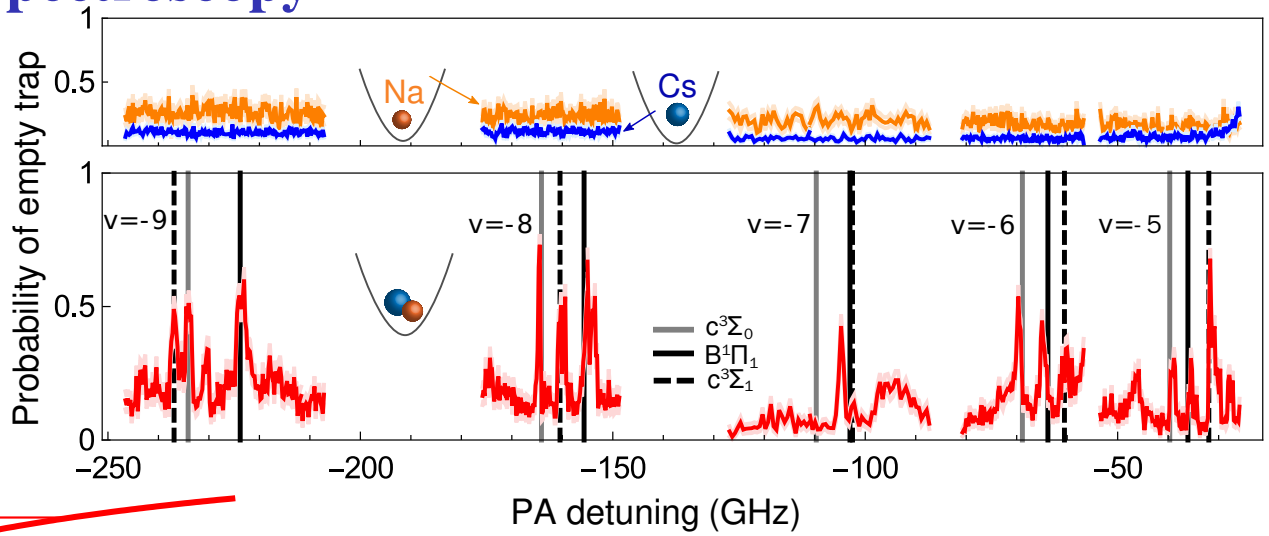

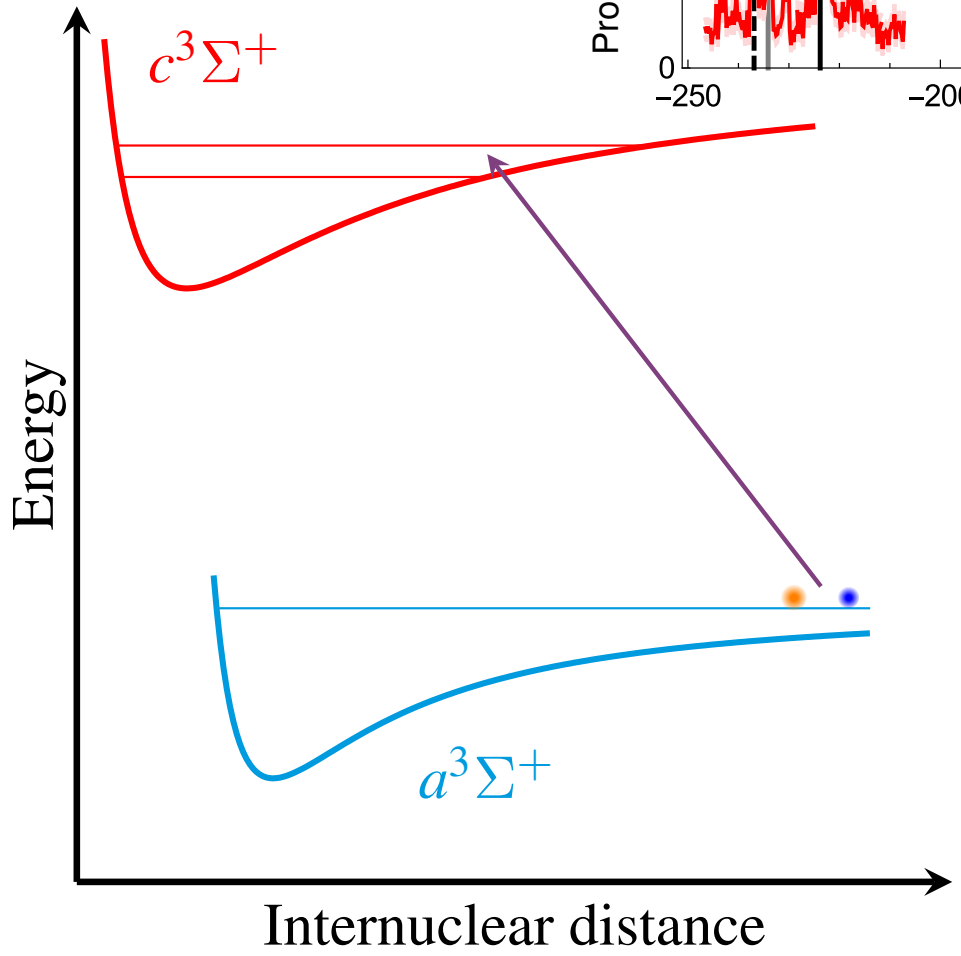

Single Atom PA

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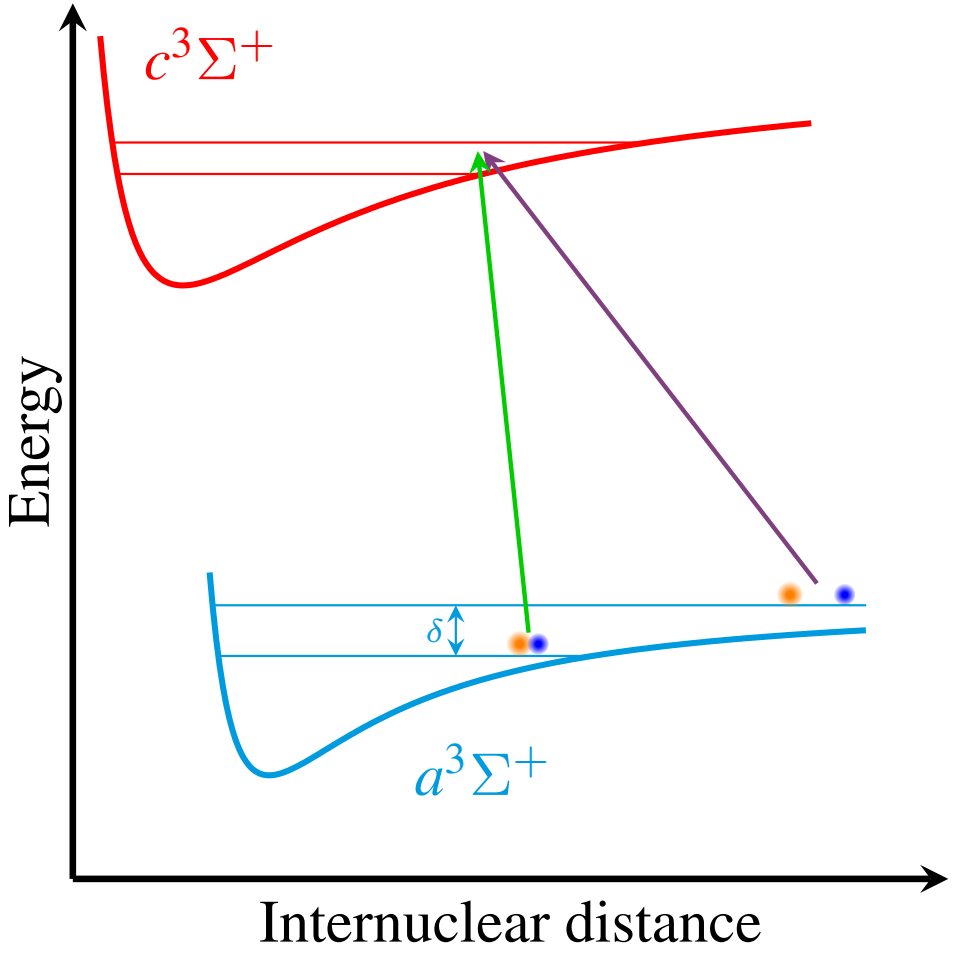
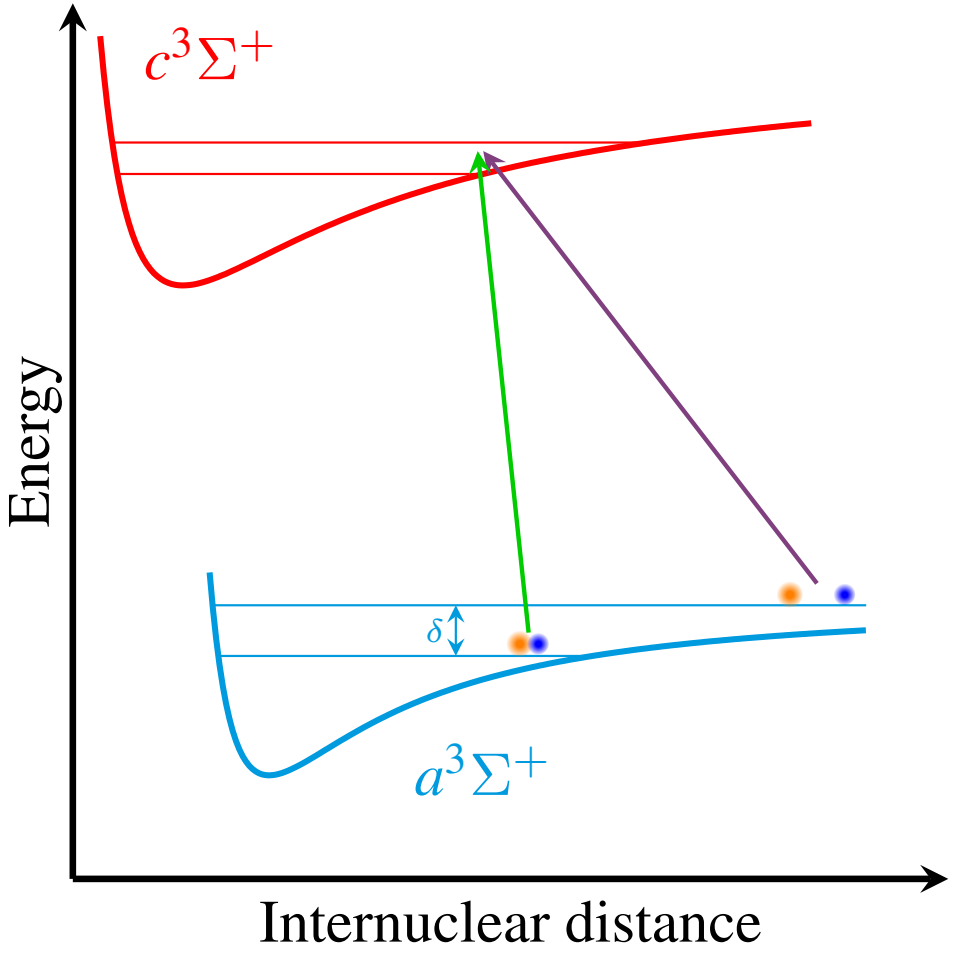
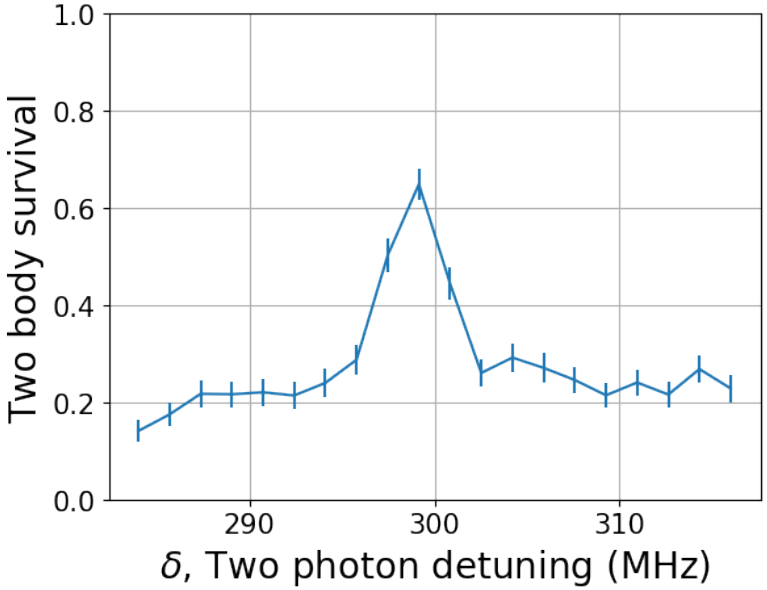
Electromagnetically Induced Transparency (EIT) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy