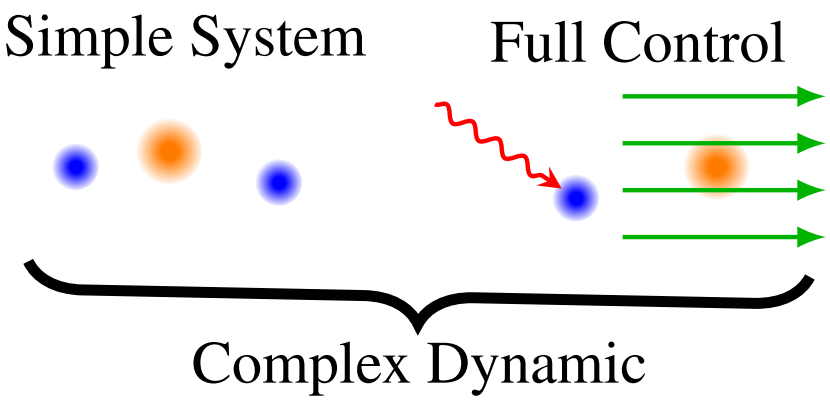
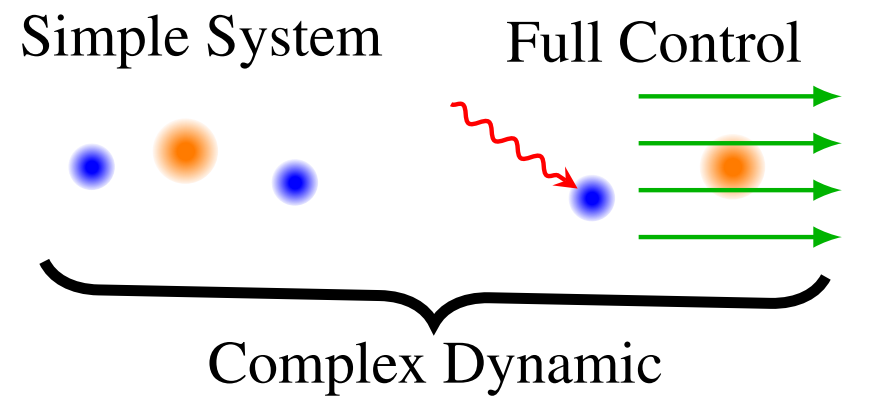
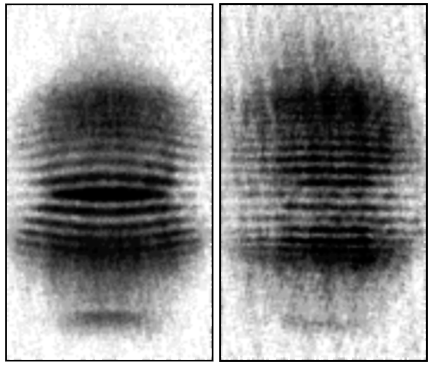
Coherent Creation of Single Molecules from Single Atoms

Yichao Yu

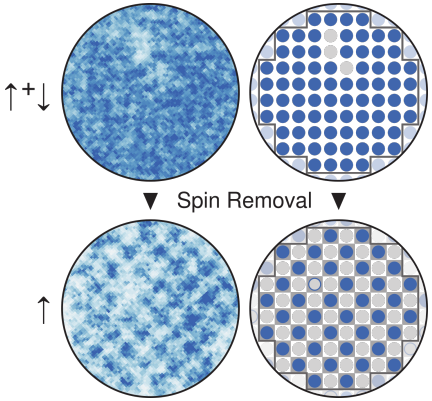
Ni Group/Harvard


Simple System
Full Control
Complex Dynamic

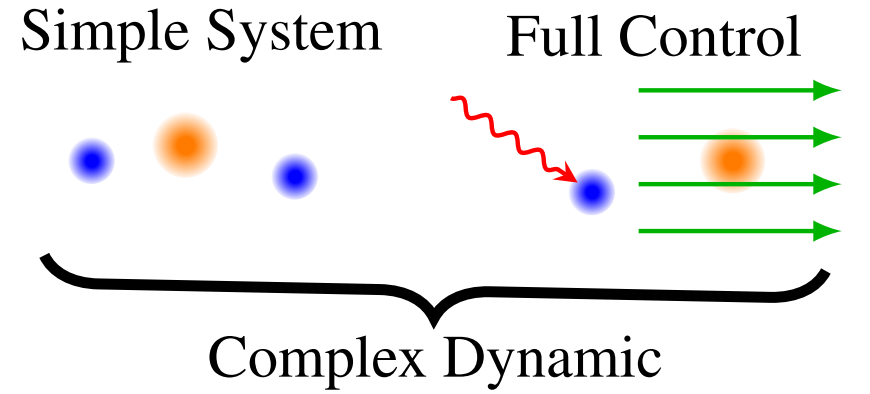


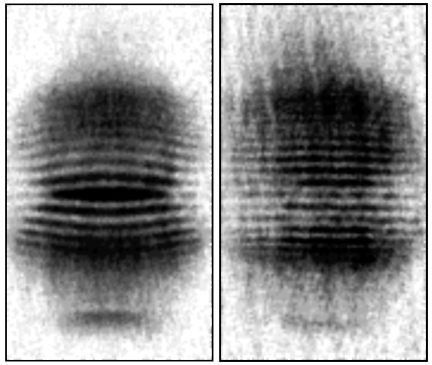


Ketterle et al.



Greiner et al.





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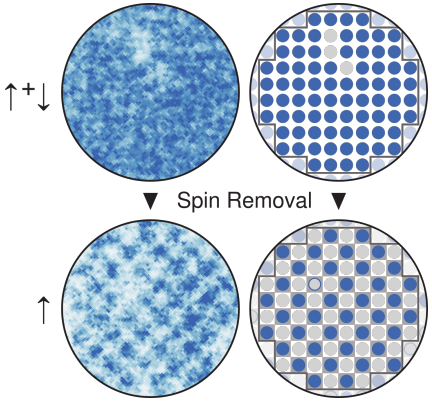
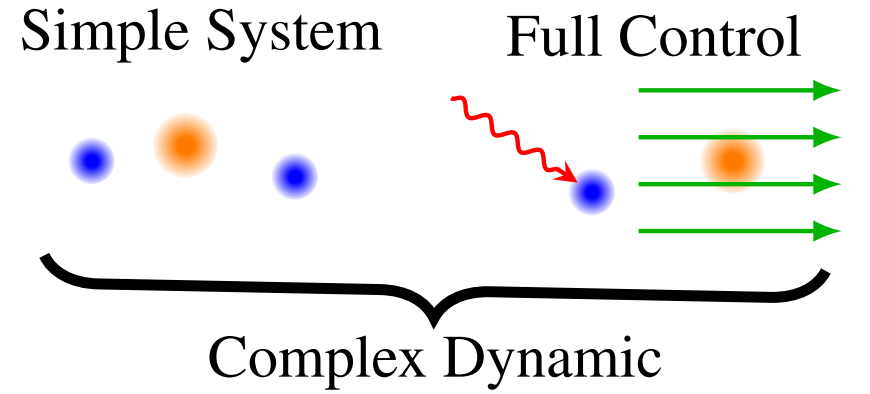
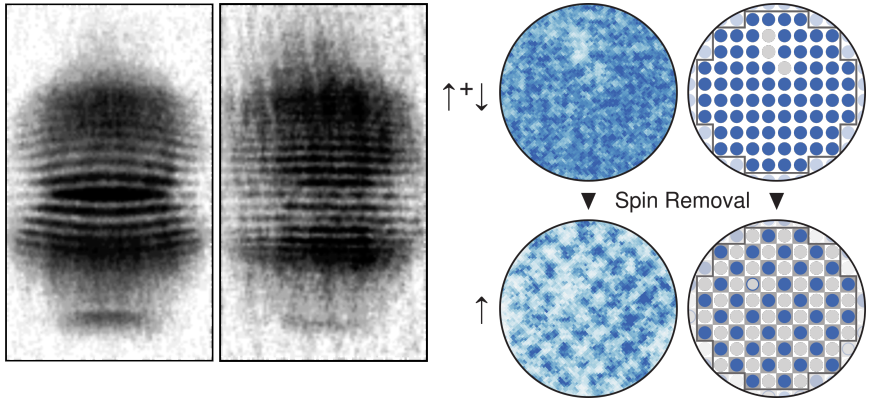


Greiner et al.

- × Simple internal structure
- × Weak interaction





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

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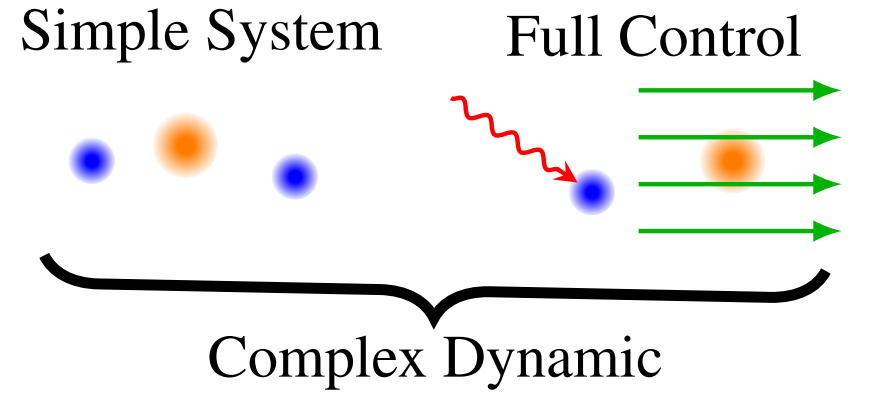

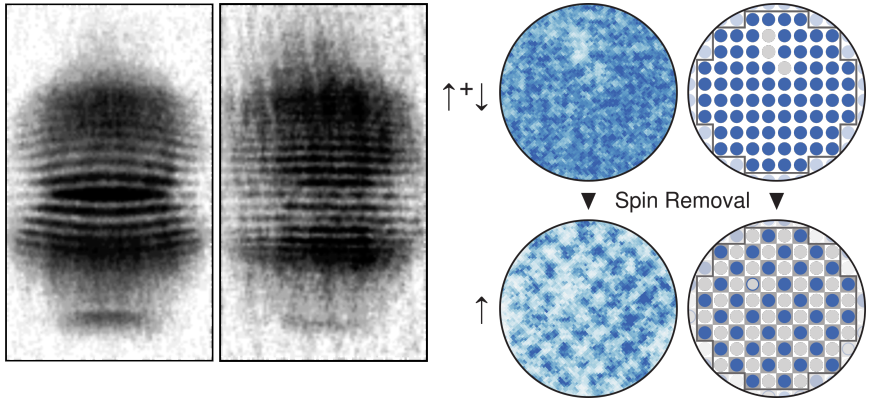

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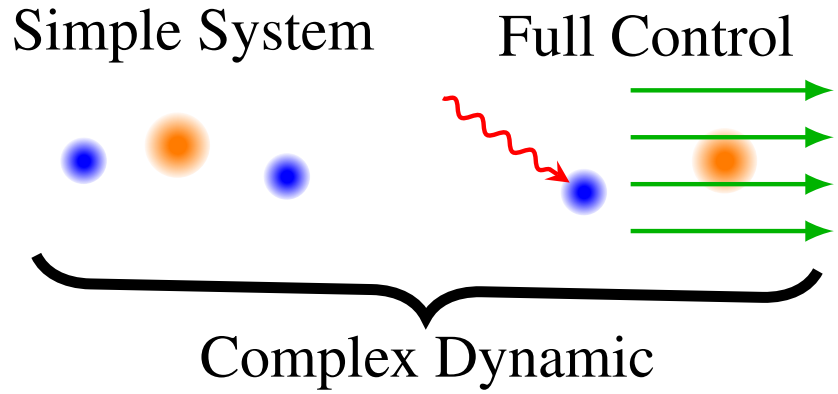

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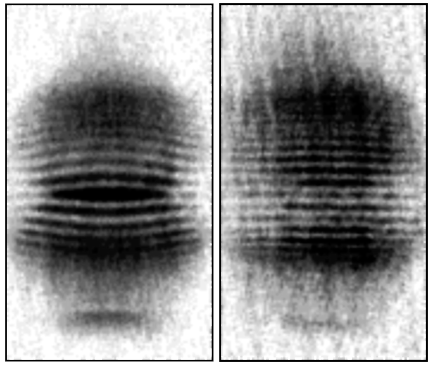

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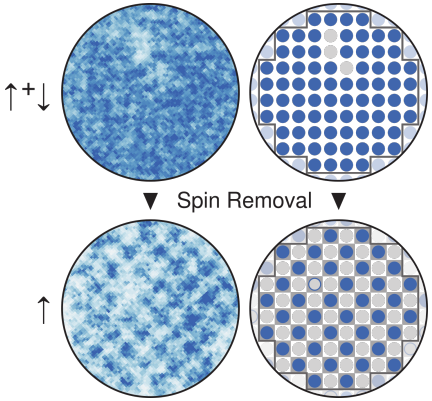

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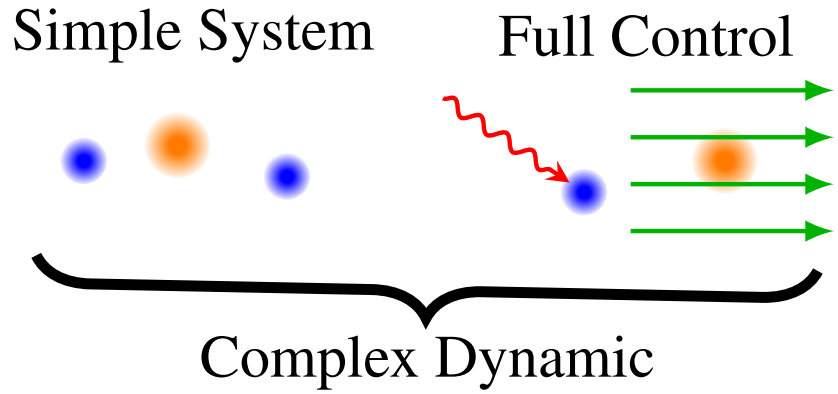

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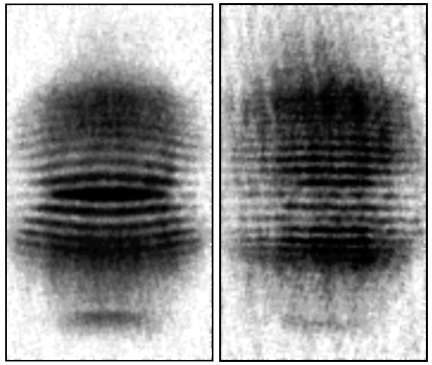

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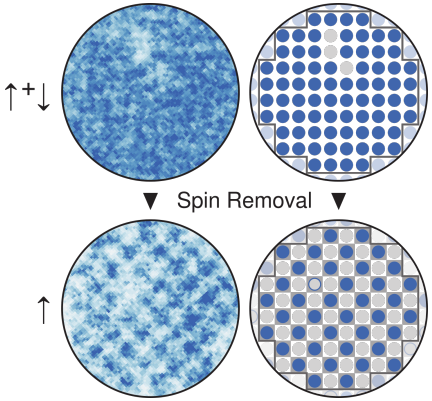

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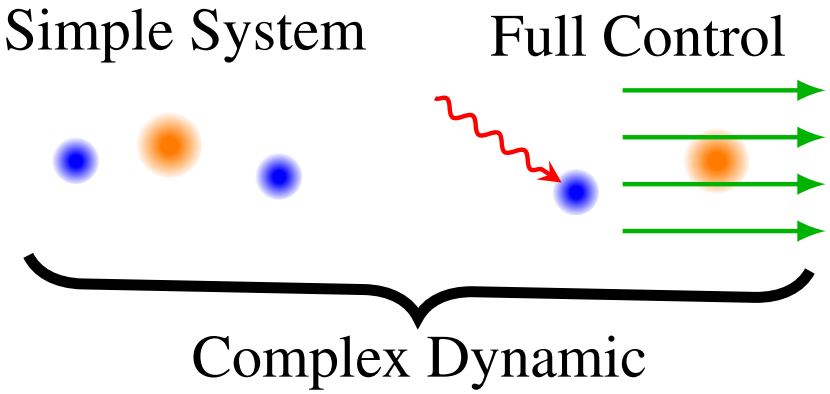

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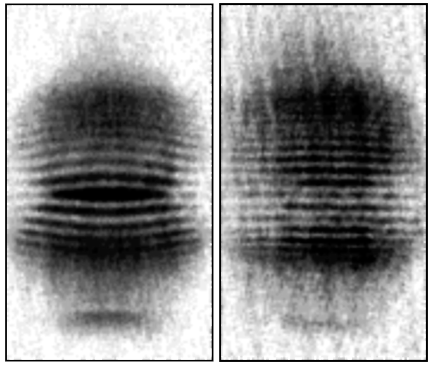

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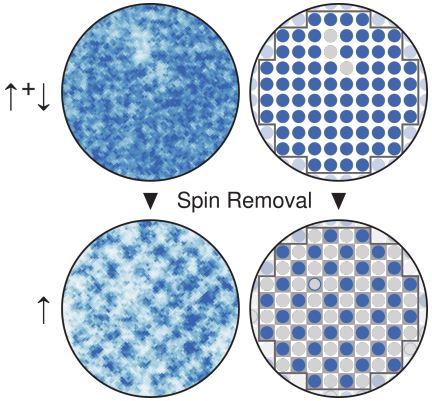

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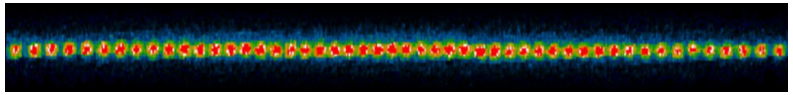

Ions (Monroe et al.)

Rydberg Atoms (Lukin et al.)

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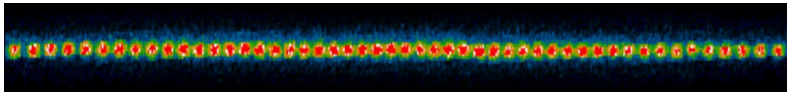

Ions (Monroe et al.)

Rydberg Atoms (Lukin et al.)

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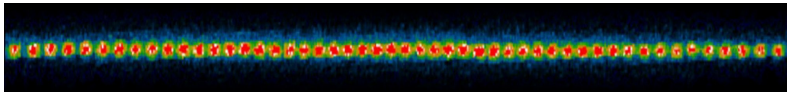

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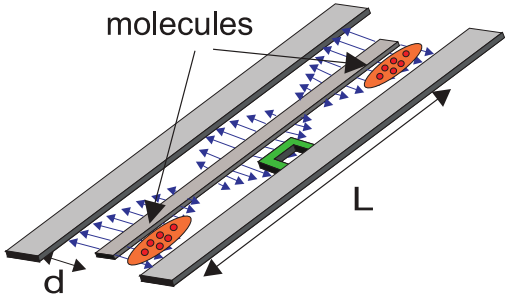

Dipolar Molecule (PRL. 97, 33003 (2006))

- ✓ **Strong interaction** (kHz)
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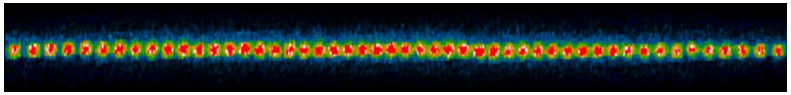

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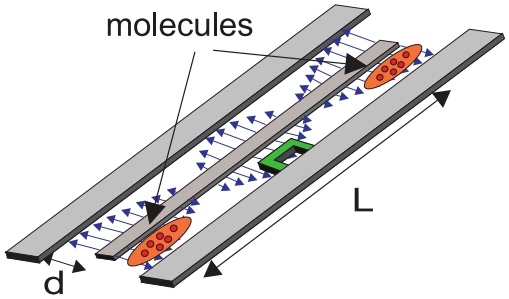

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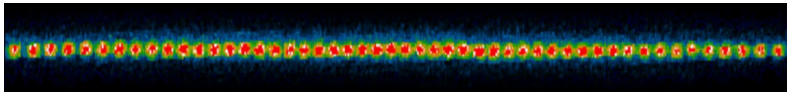

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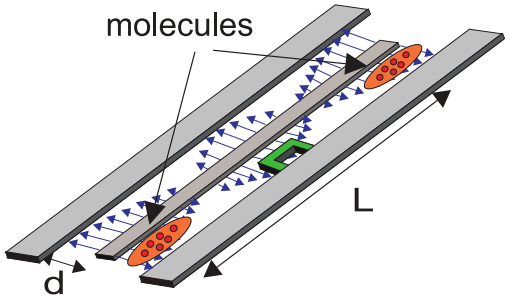

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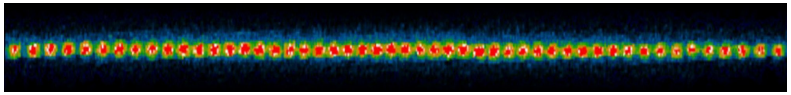

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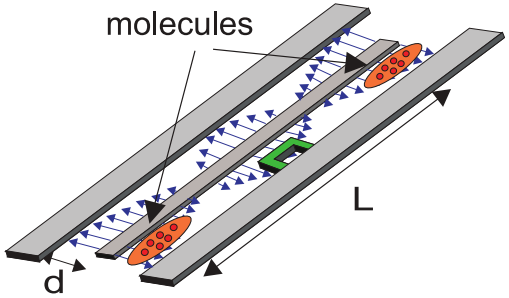

Dipolar Molecule (PRL. 97, 33003 (2006))

Optical tweezers

- Single site resolution
- . . .

- ✓ **Strong interaction** (kHz)
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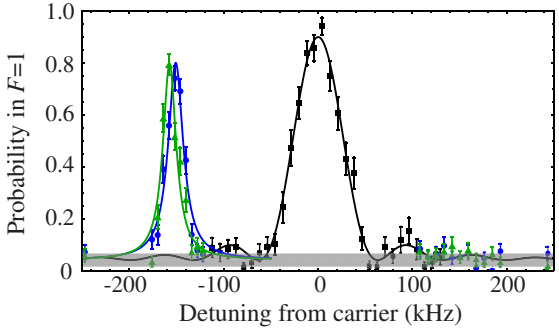
- ✓ **Fully controllable**

Optical tweezers

- Single site resolution
- . . .

Cooling



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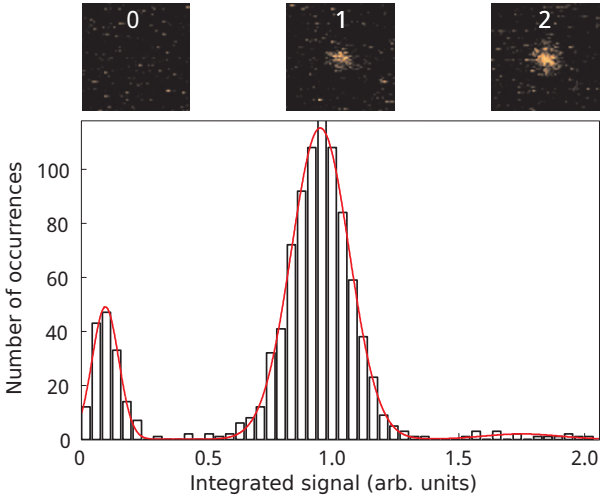
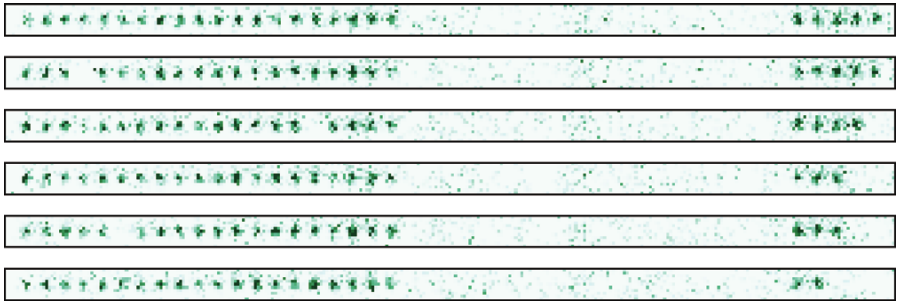


Nat. Phys. 6, 951 (2010)

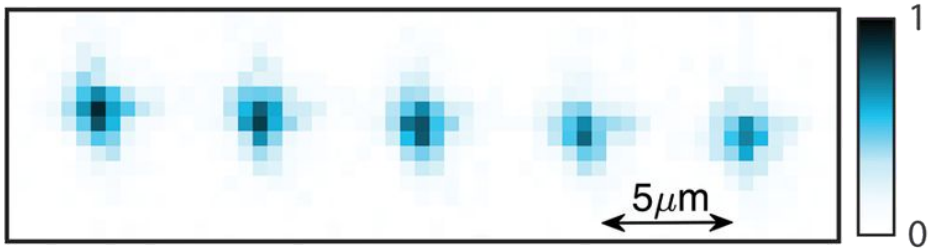
Rearranging



Science 354, 1024 (2016)

Ultracold molecules in tweezers

Direct cooling



Science 365, 1156 (2019)

Ultracold molecules in tweezers

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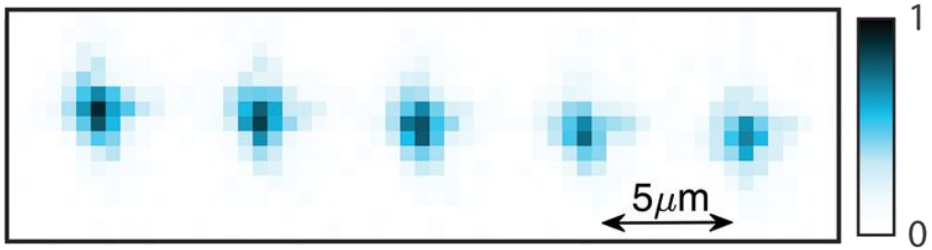
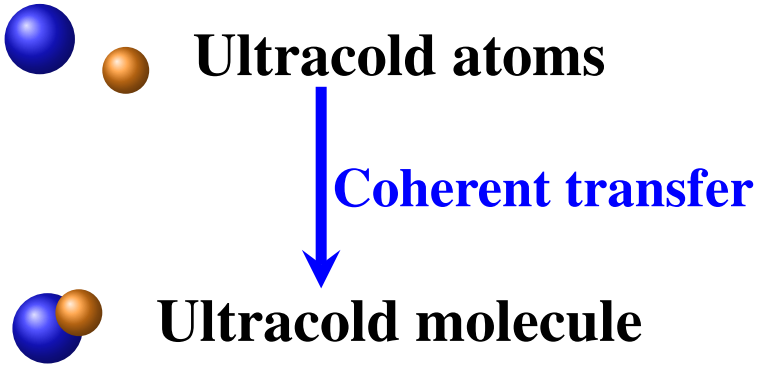


Science 365, 1156 (2019)

Assembly



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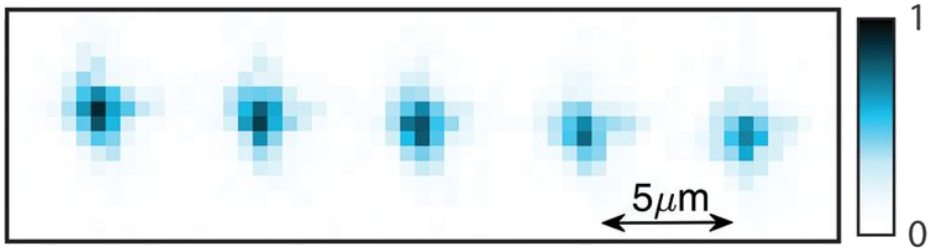

Science 365, 1156 (2019)

Assembly

Challenges

- Temperature in tweezer
- Quantum control

Ultracold molecules in tweezers

Direct cooling

Science 365, 1156 (2019)

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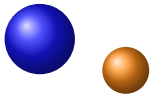


Ultracold atoms

Coherent transfer

Ultracold molecule

Challenges

- Temperature in tweezer
- Quantum control

- Control of atoms
- Coherent creation of molecules

Outline

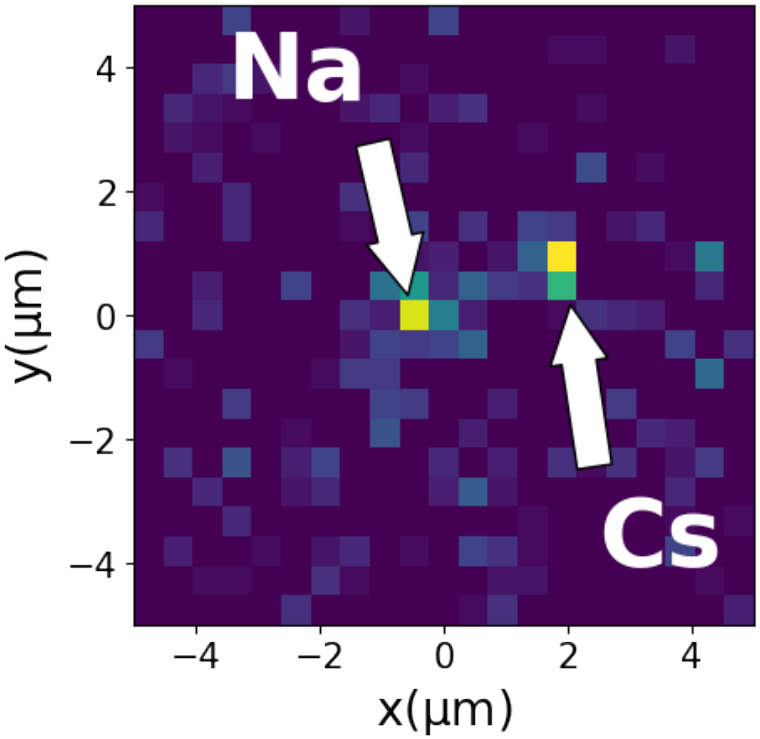
1. **Experiment overview**
2. **Atom state control**
   - Raman sideband cooling of Na atoms
3. **Molecule creation**
   - Atom-atom interaction
   - Coherent optical transfer
4. **Conclusion**

NaCs molecule

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

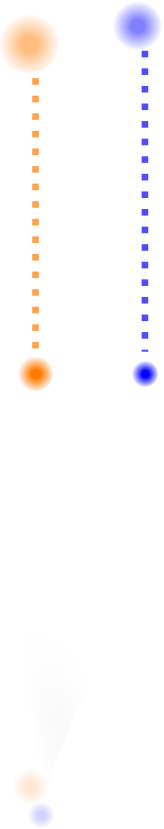
Experiment overview

Loading

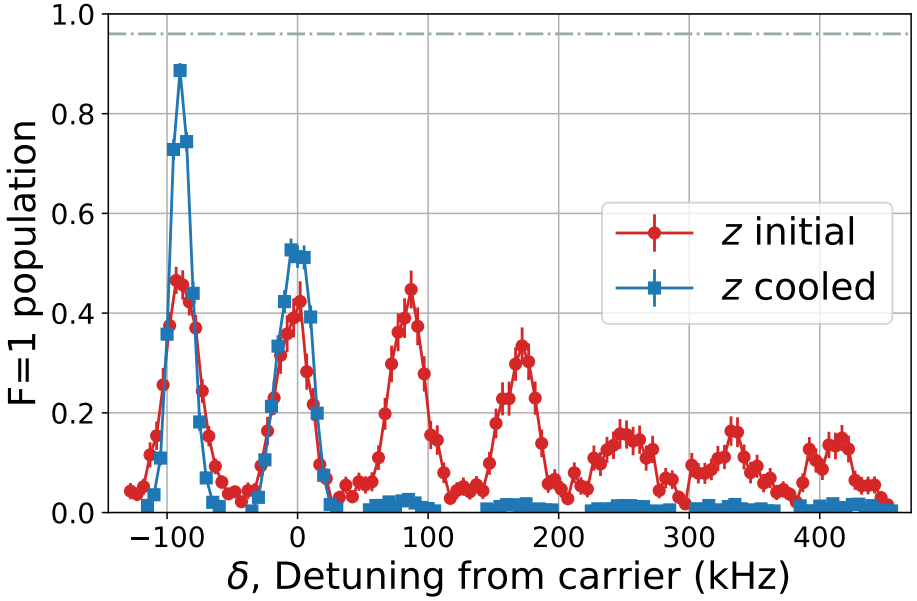


Loading probability per site: 60%
Post select on initial and final state.

Experiment overview



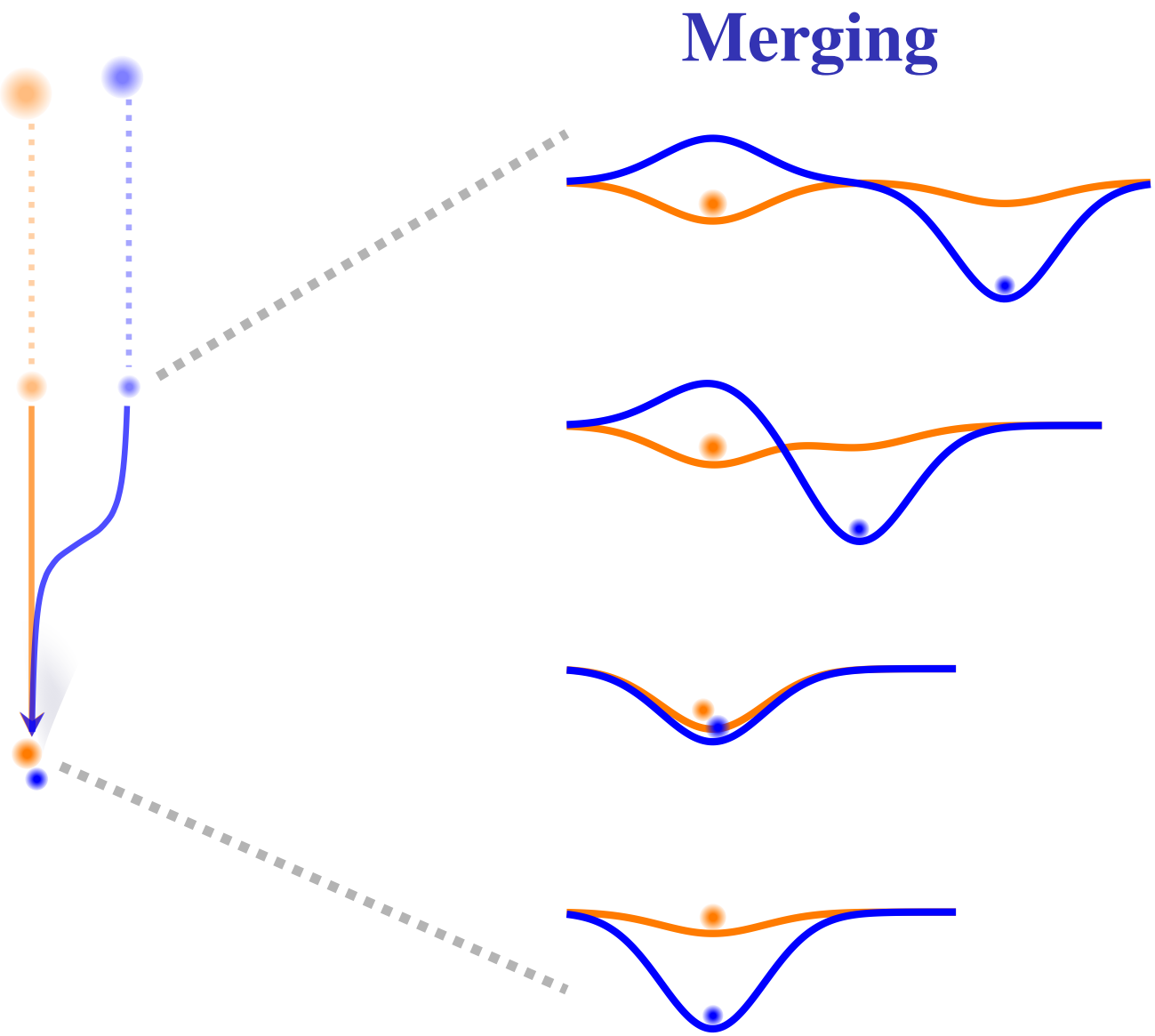
Cooling



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



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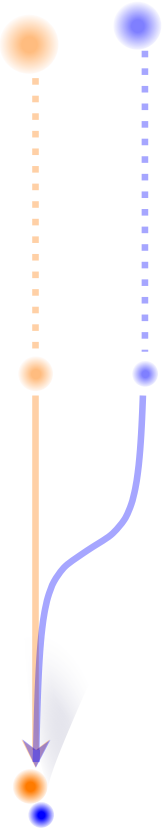
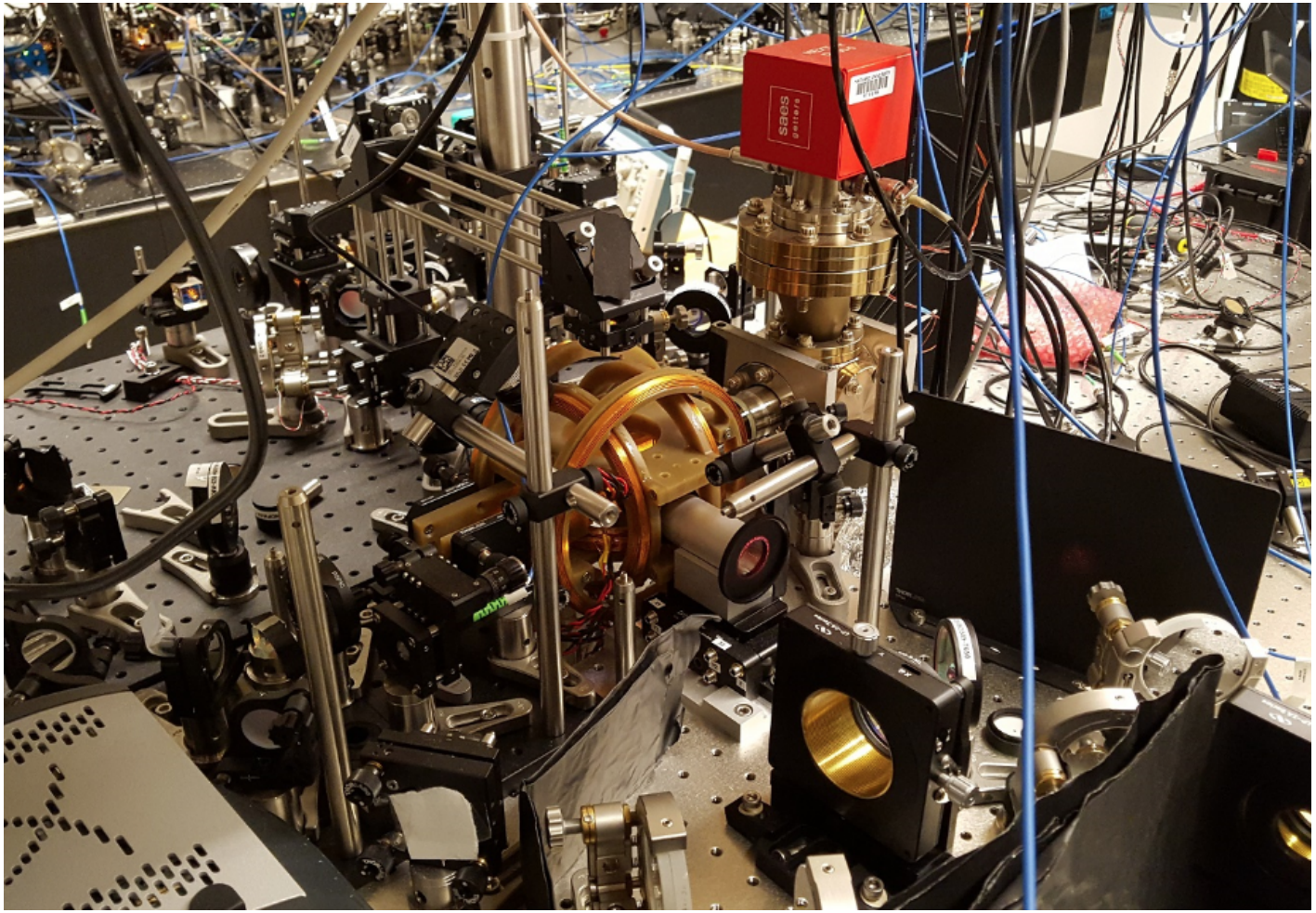


L. R. Liu, J. D. Hood, **Y. Yu** et al., Science 360, 6391 (2018)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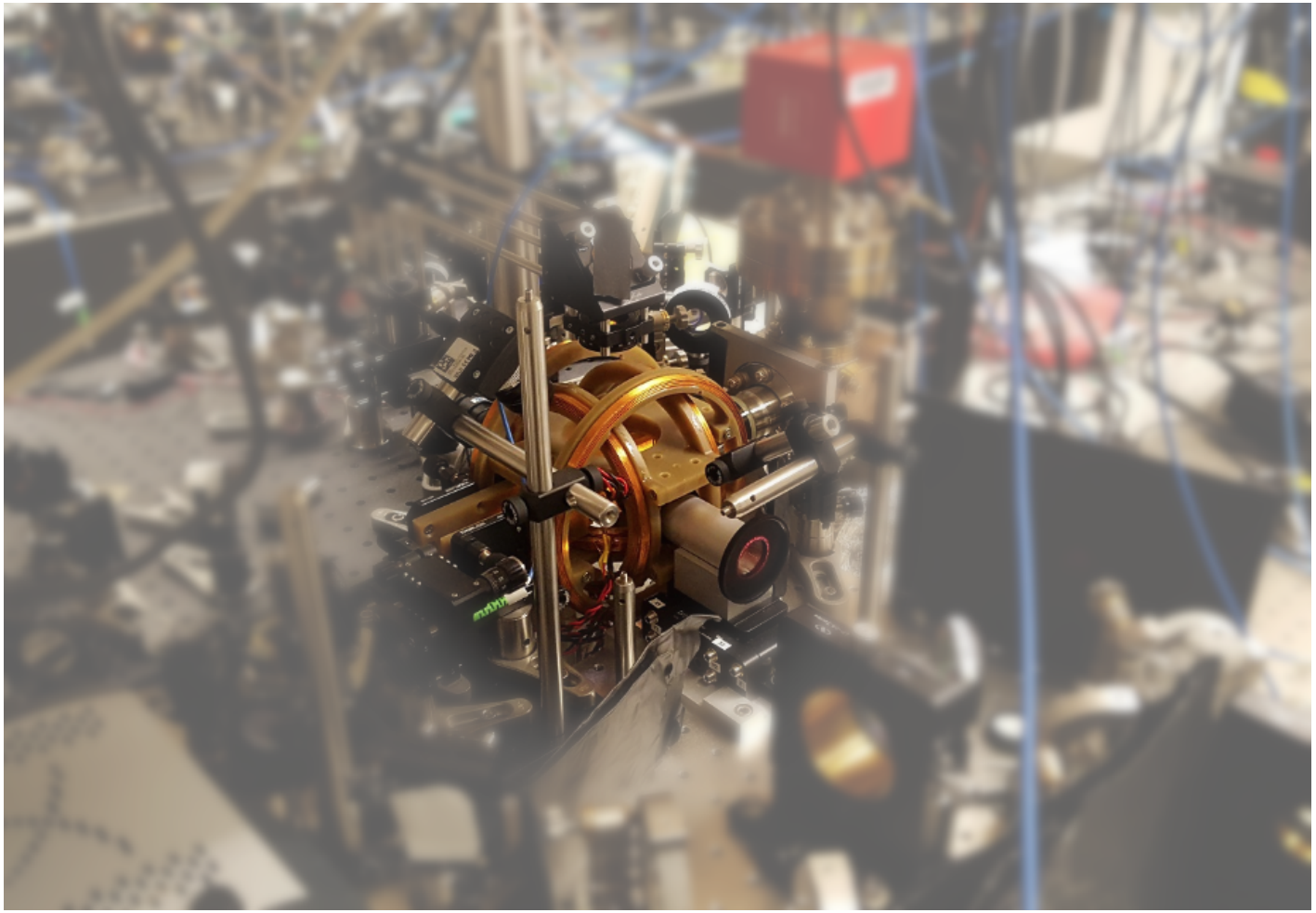
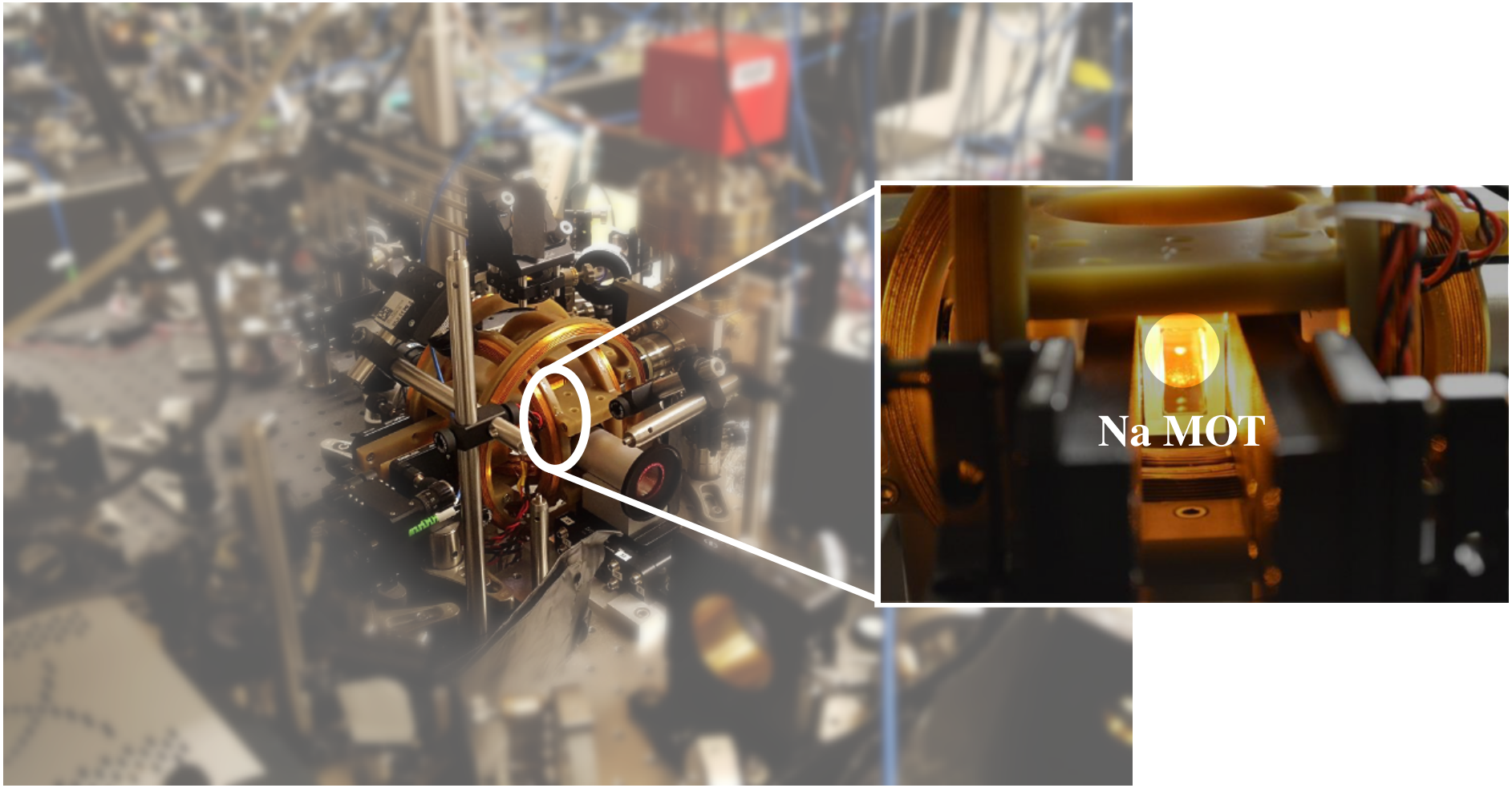
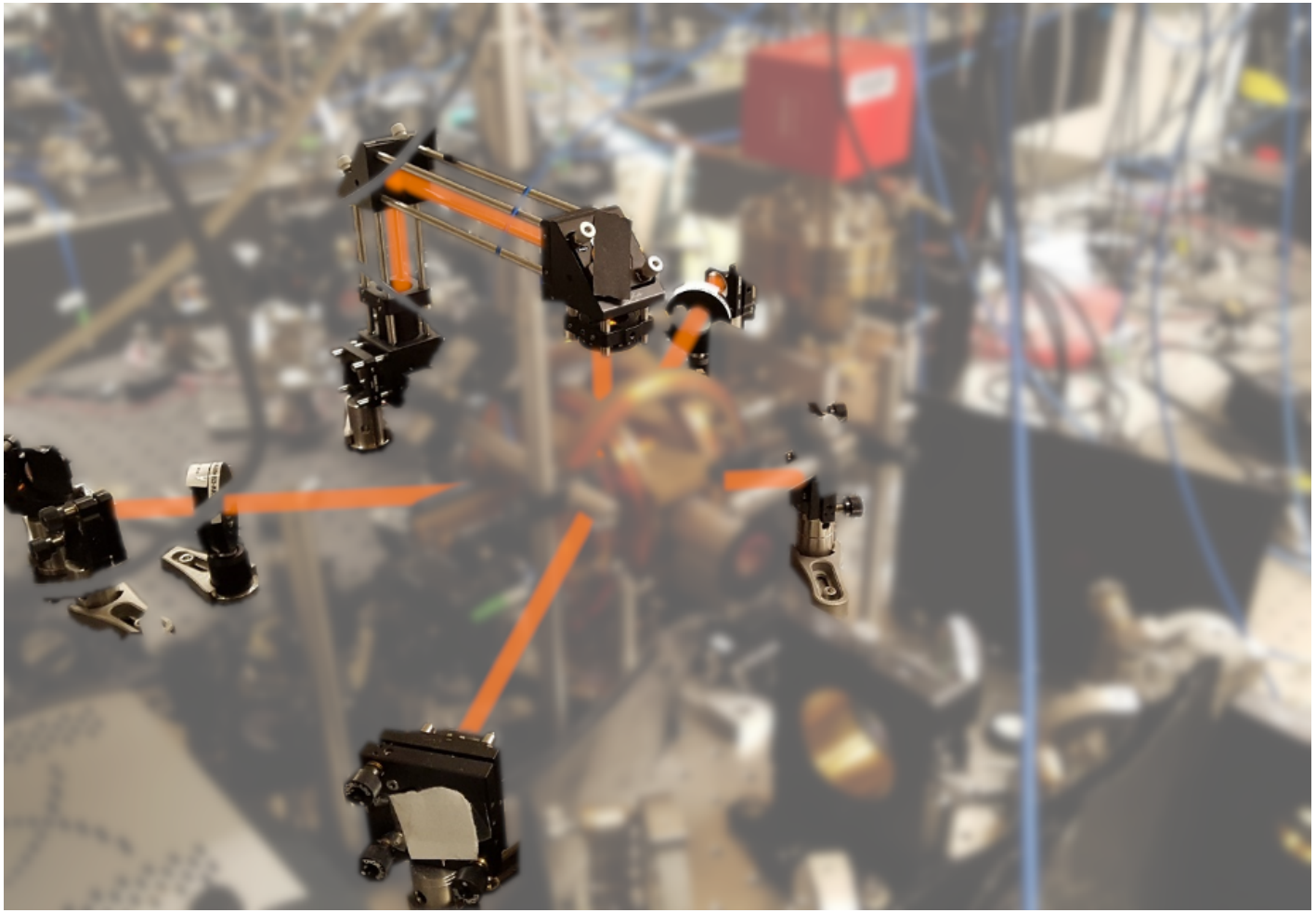
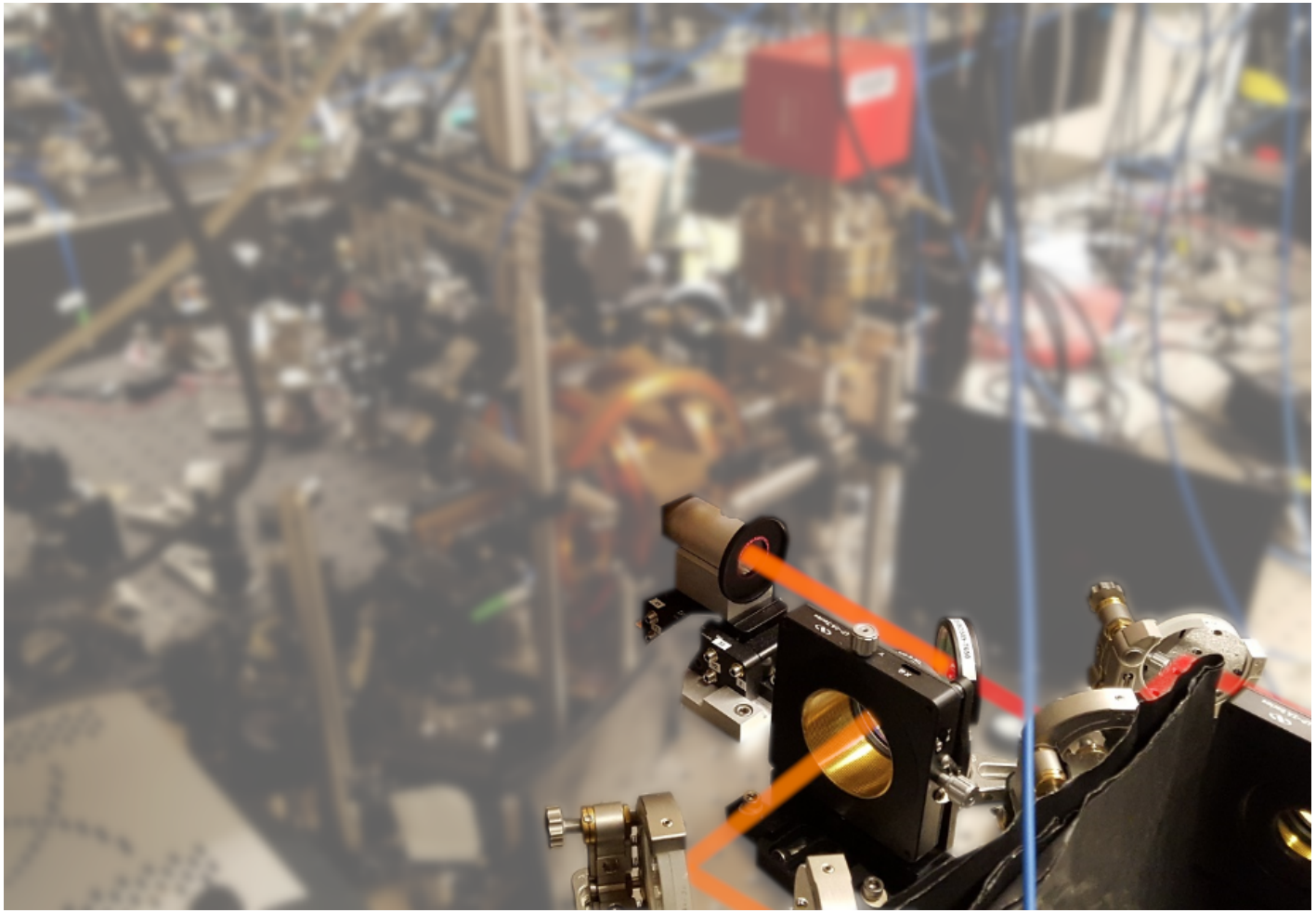



Na MOT

MOT beam path



Tweezer and imaging beam path



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Raman sideband cooling

Used for cooling in trap

Raman sideband cooling

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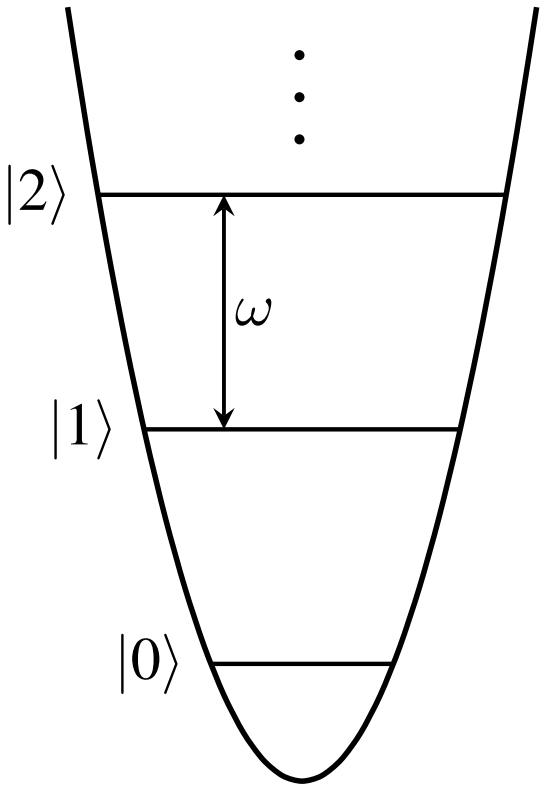

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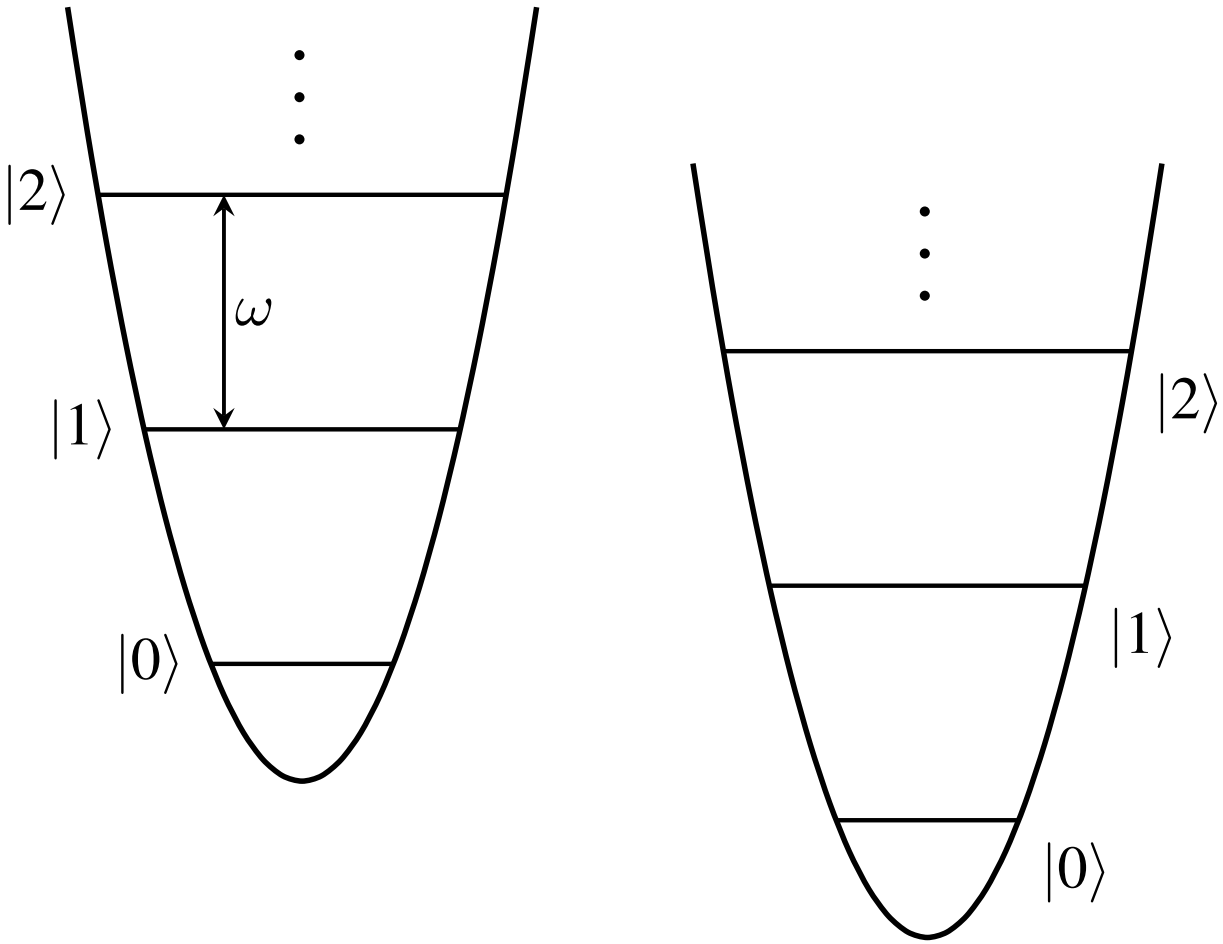

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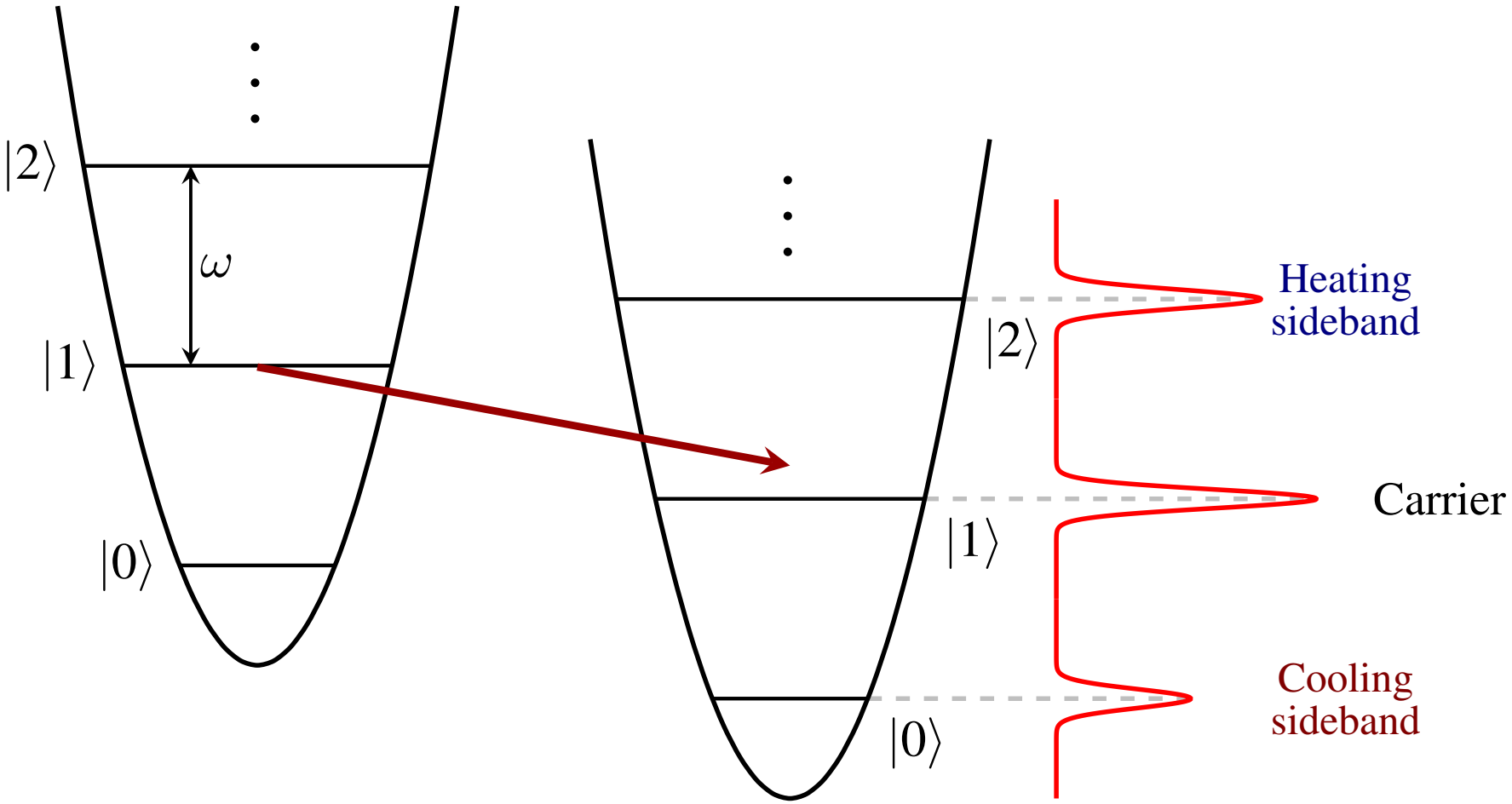

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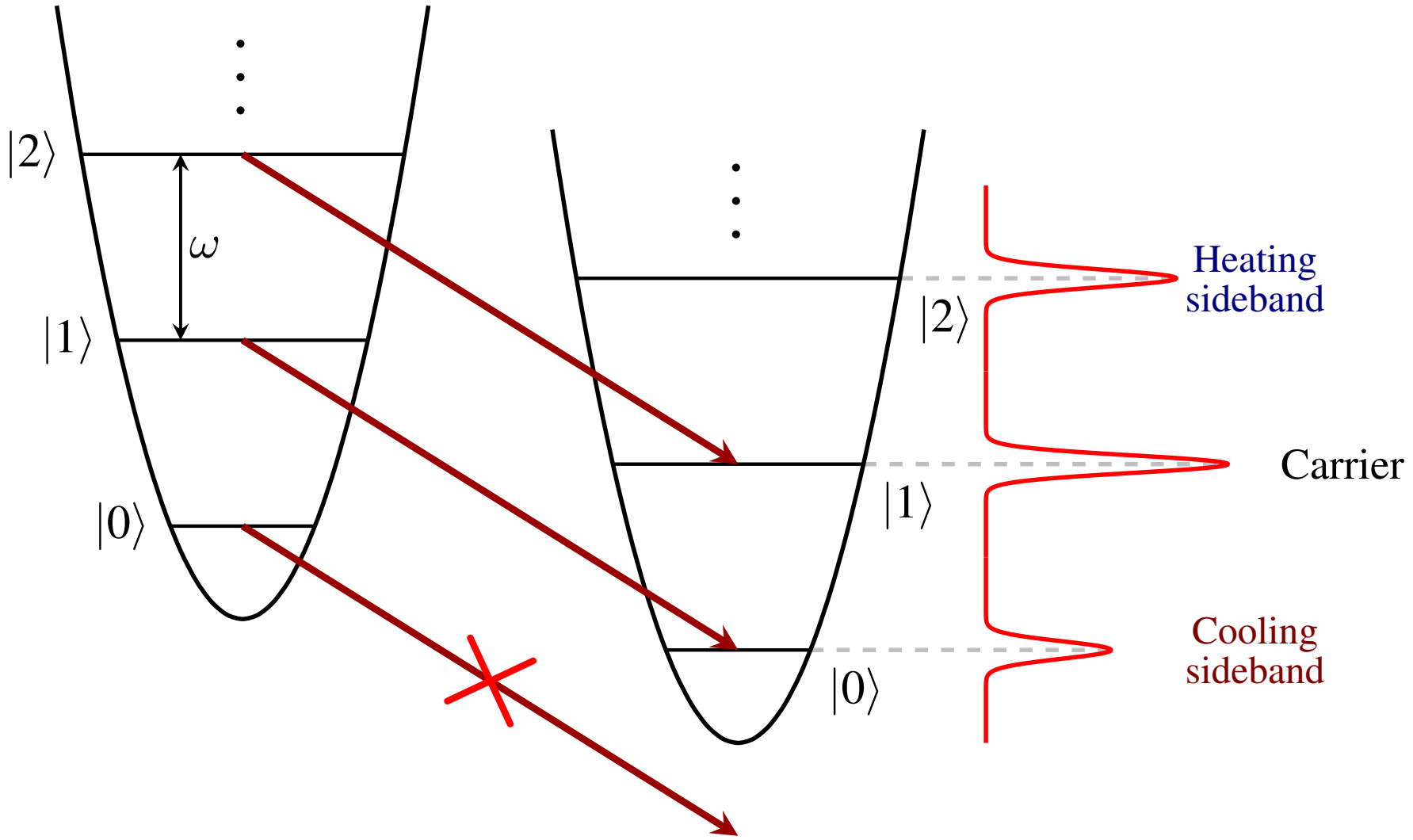

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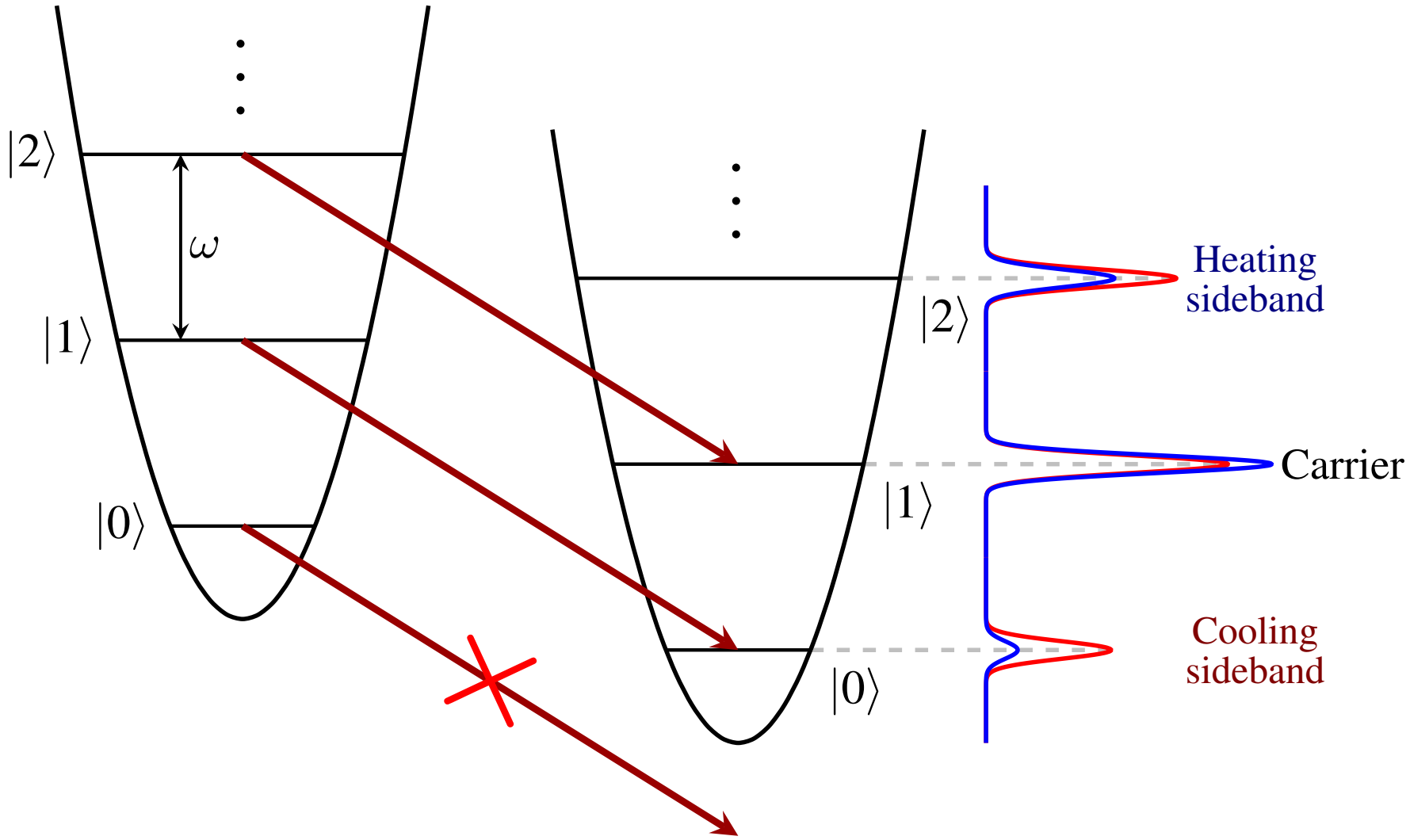

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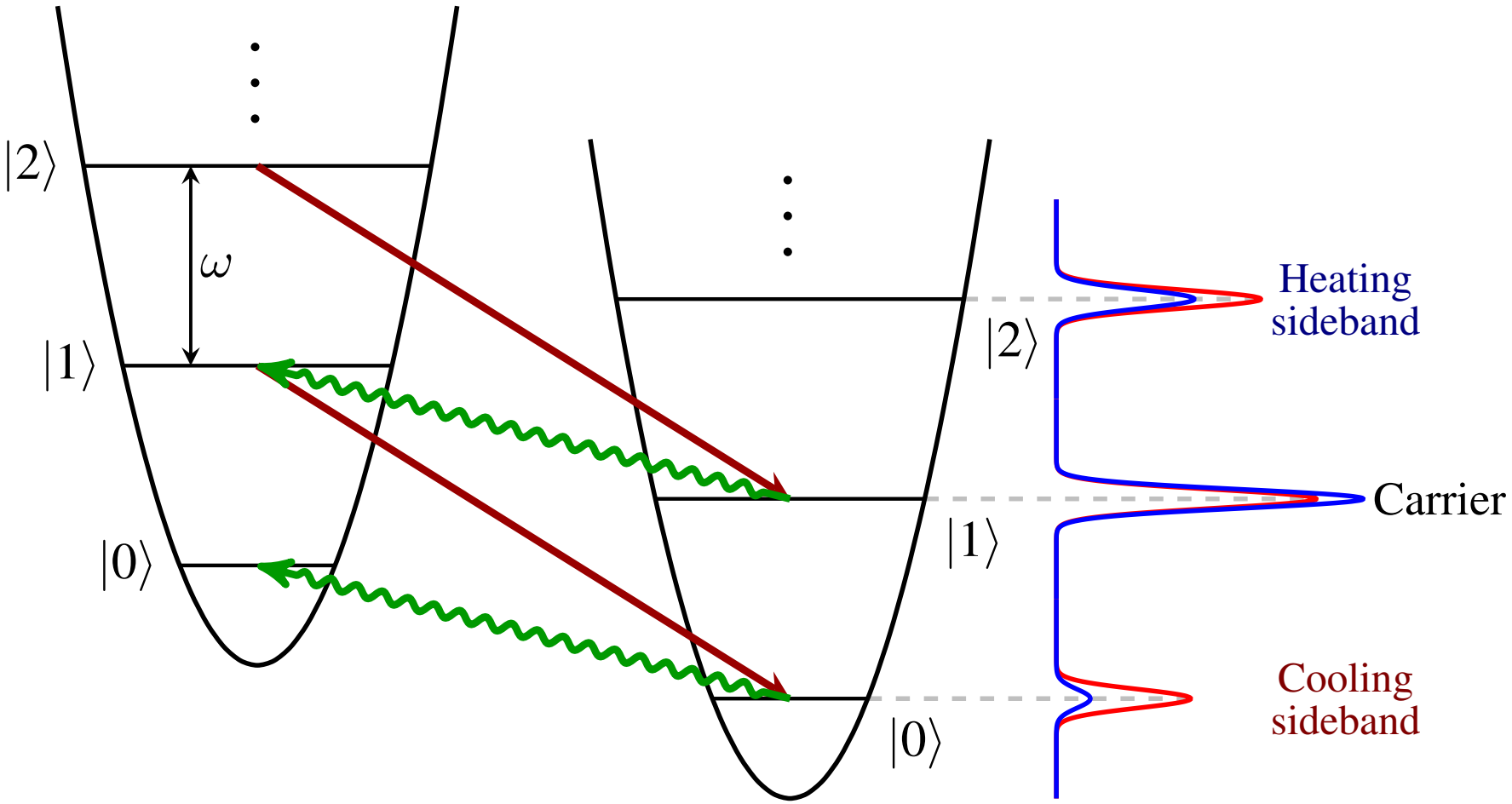

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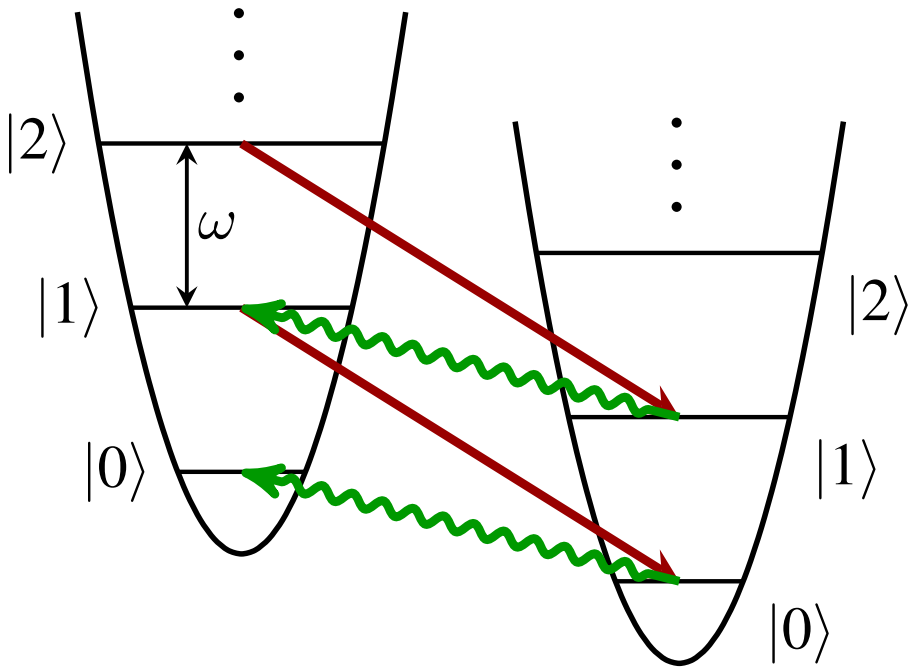

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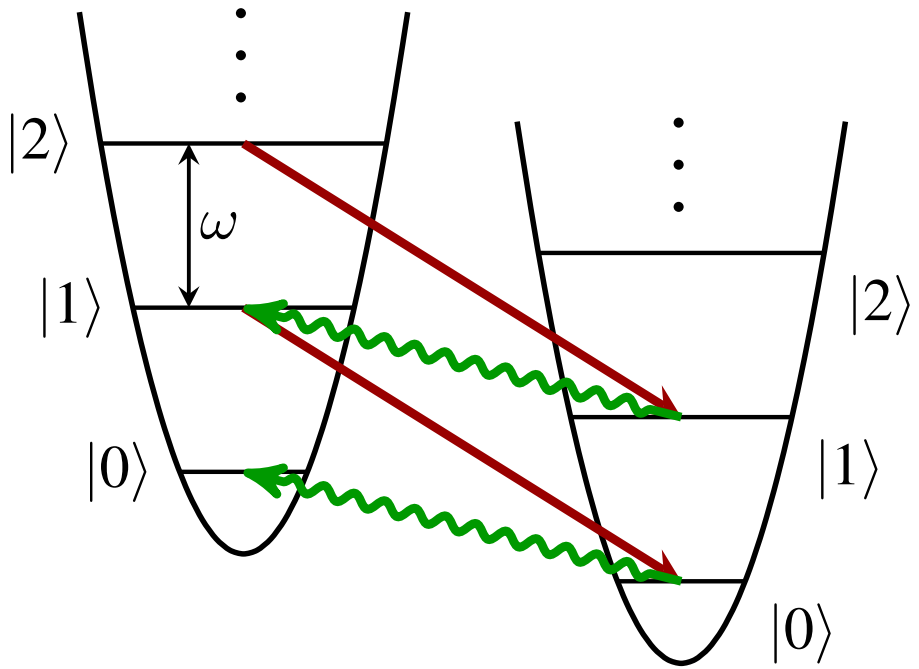

Lamb Dicke parameter

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

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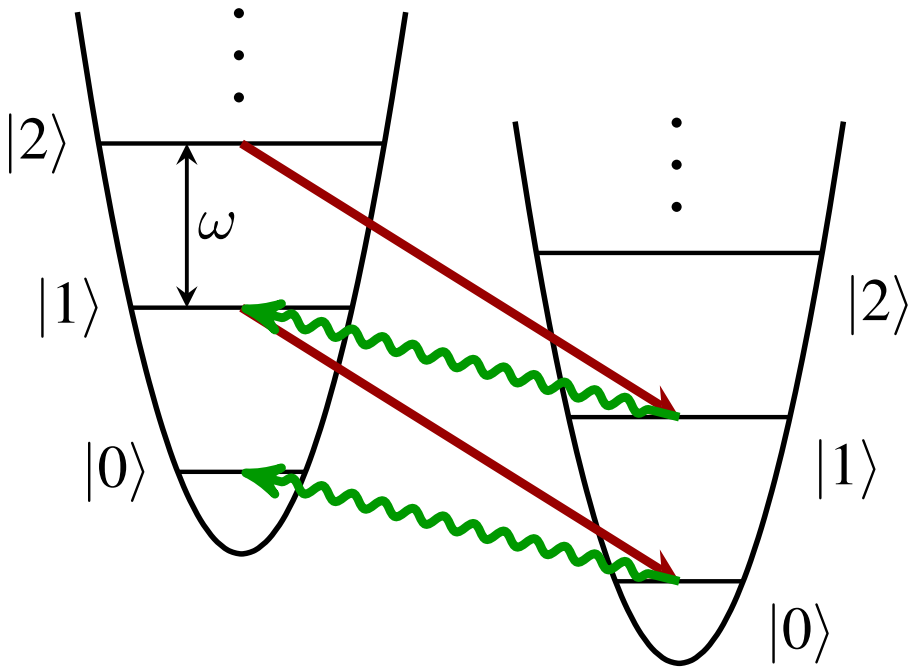

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- Motional state branching
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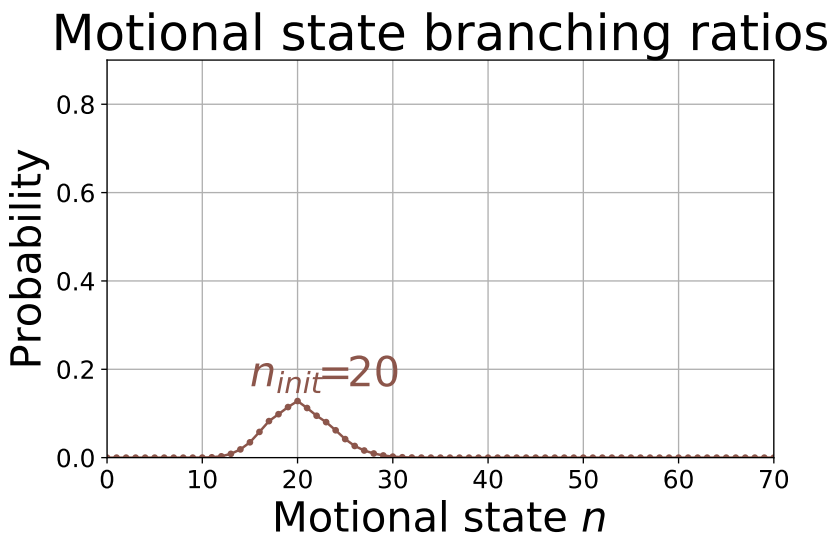

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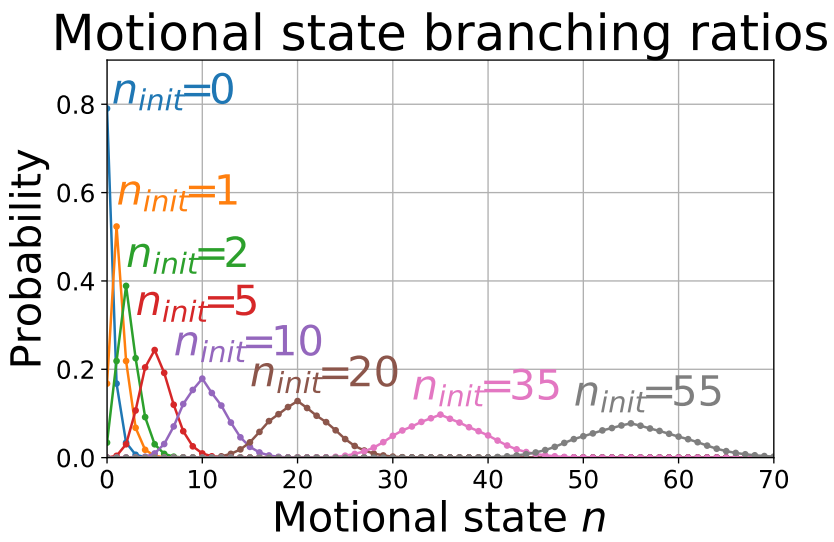

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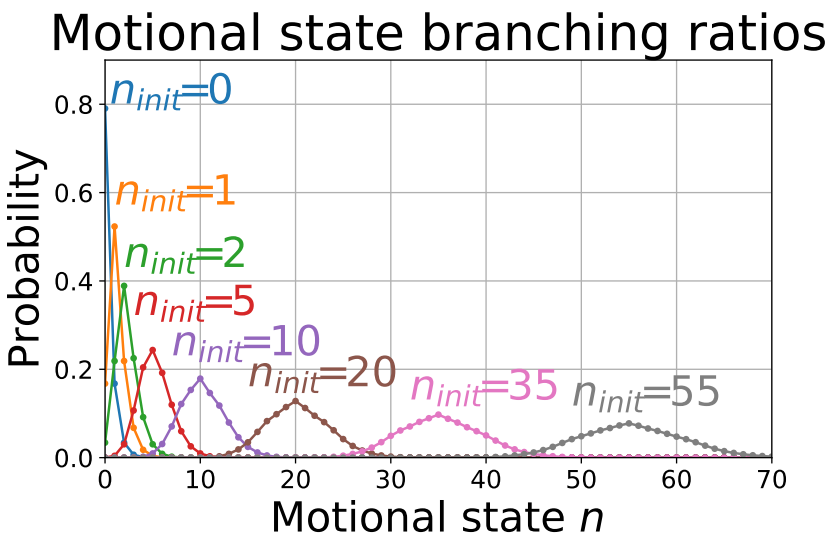

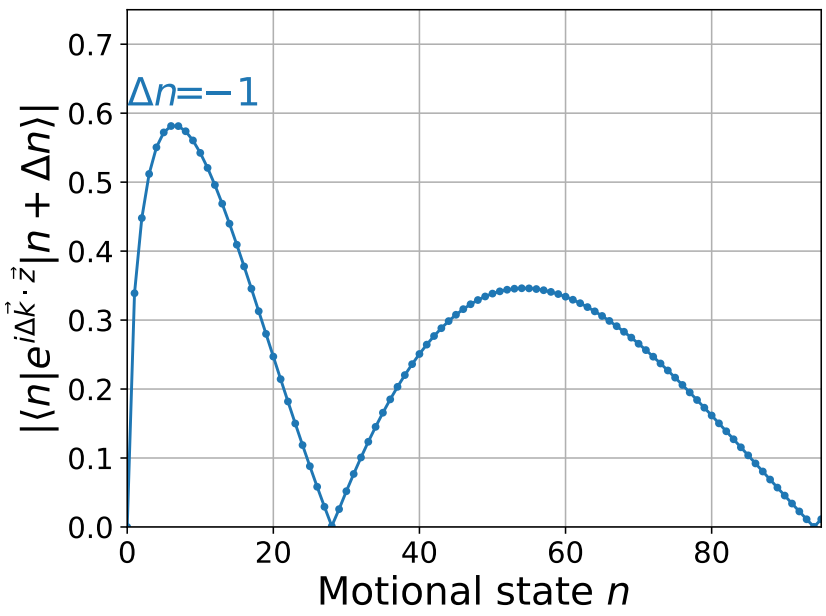

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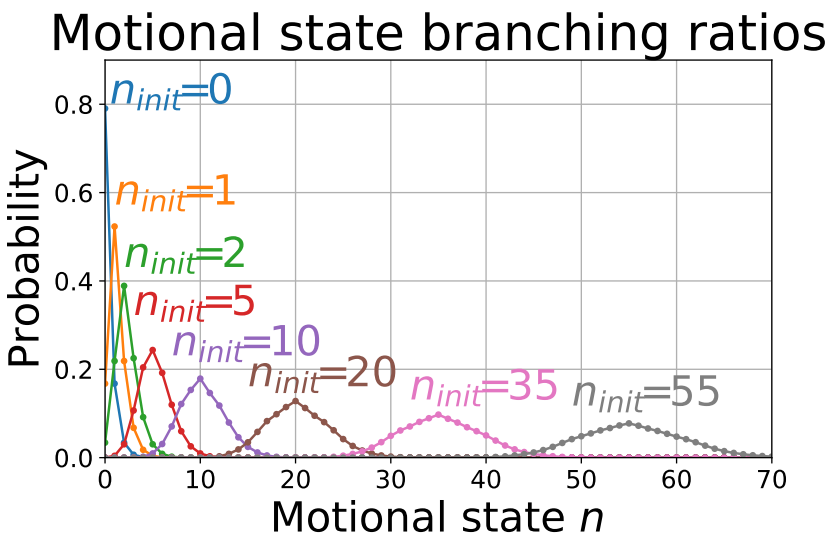

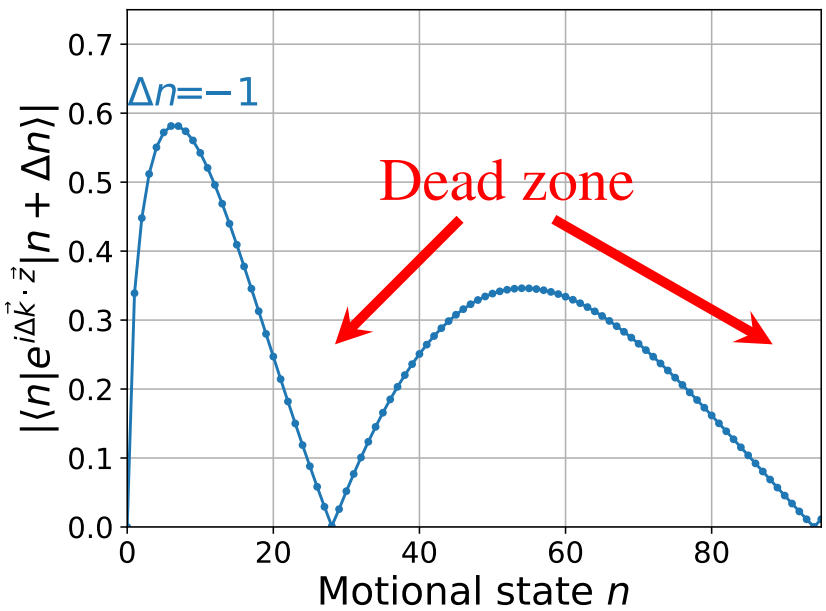

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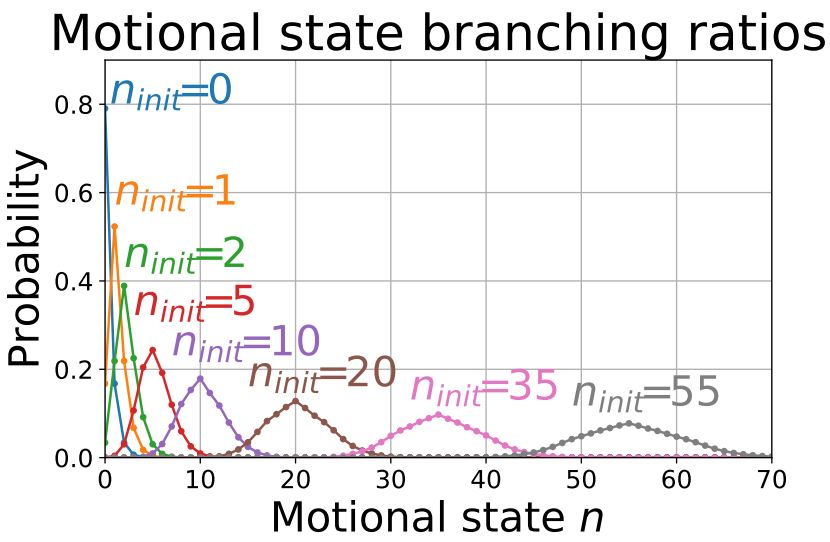

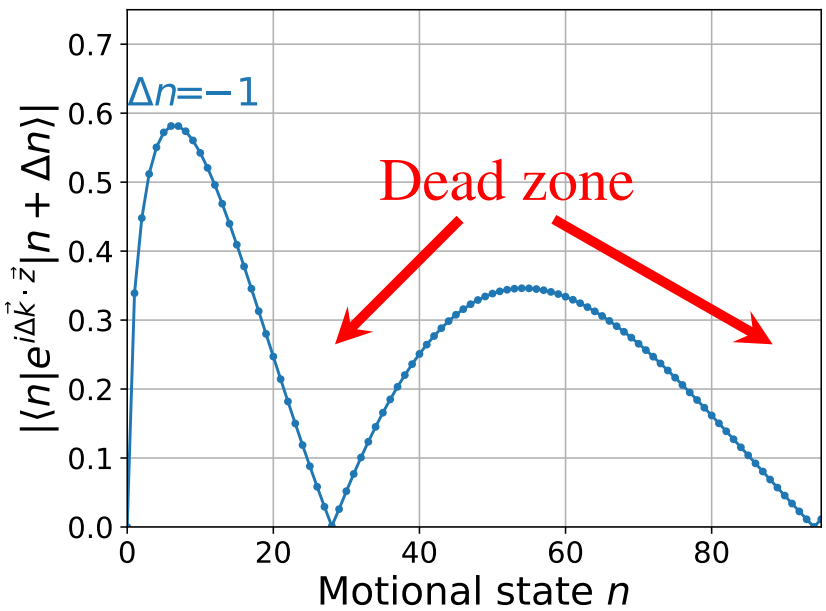

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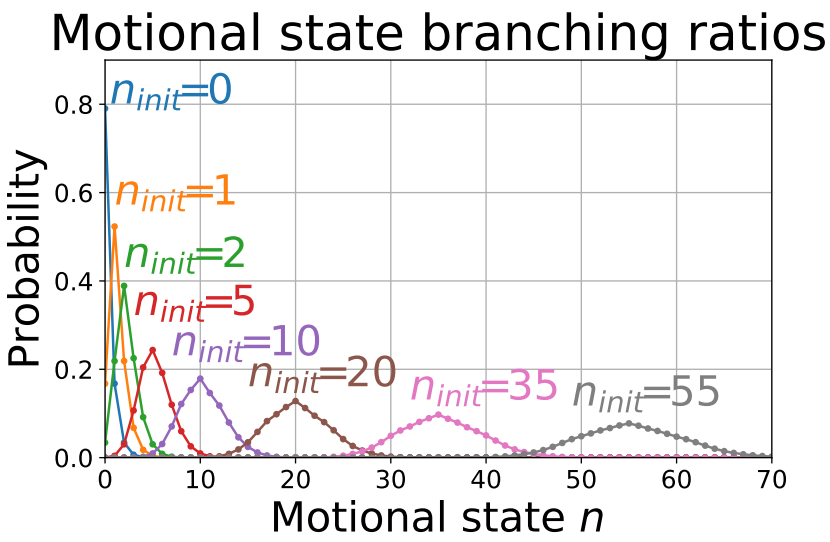

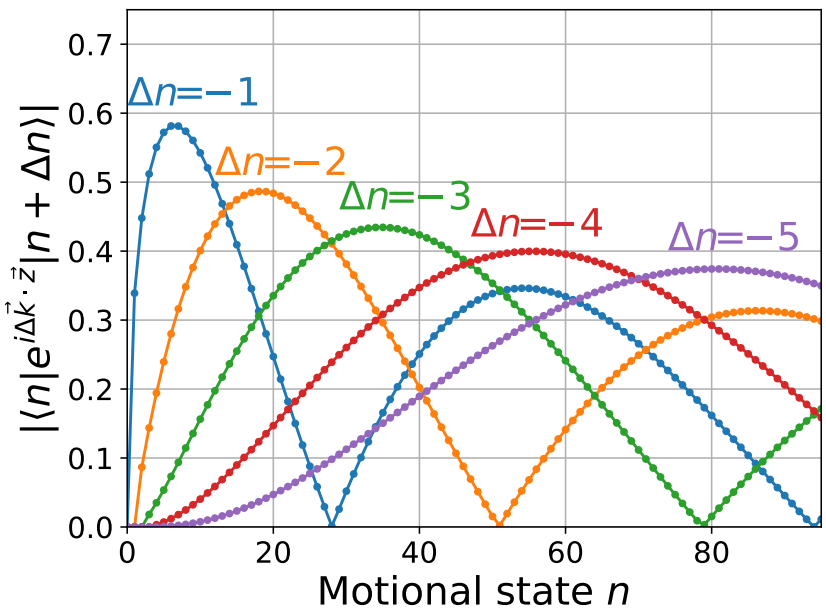

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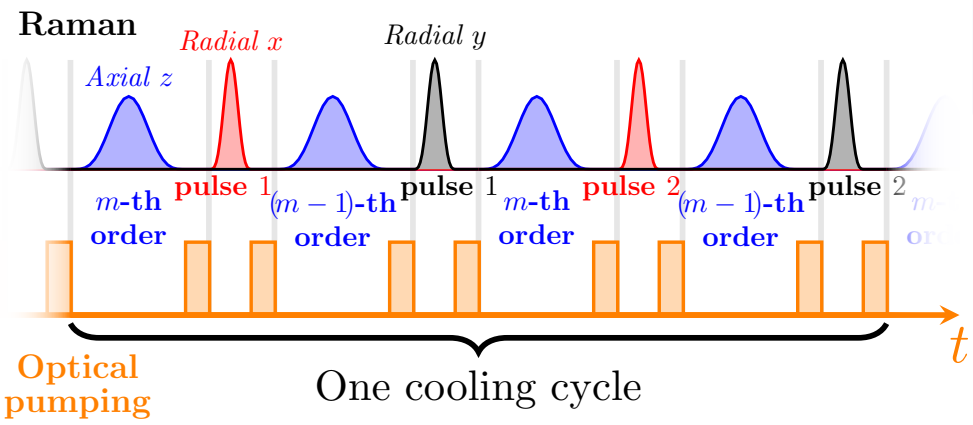

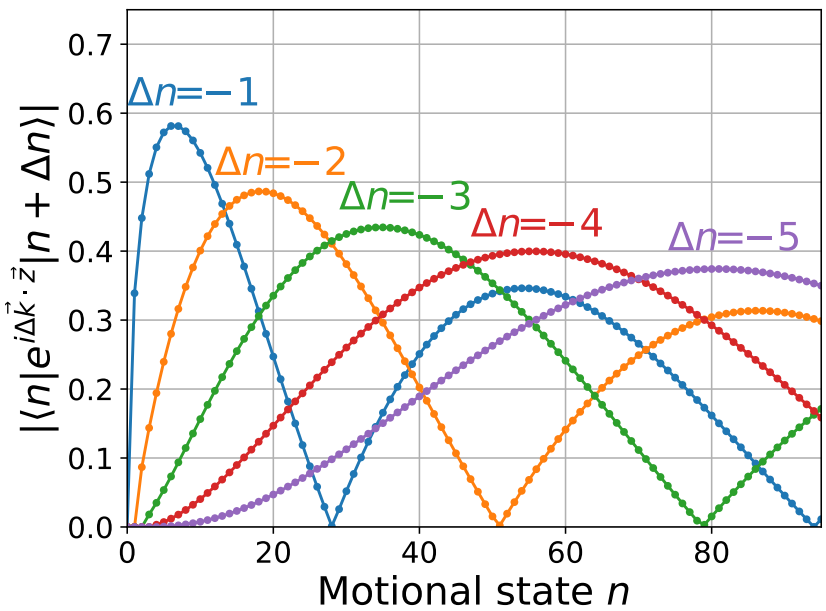

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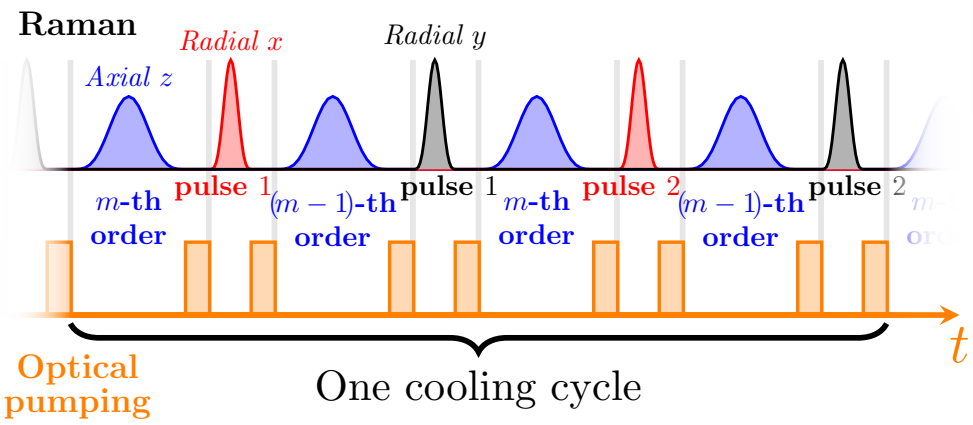
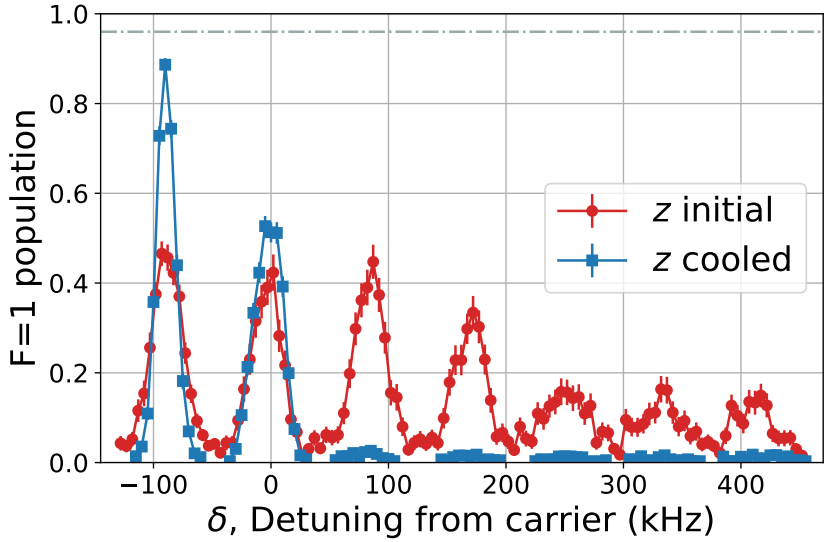


Axial sideband spectrum



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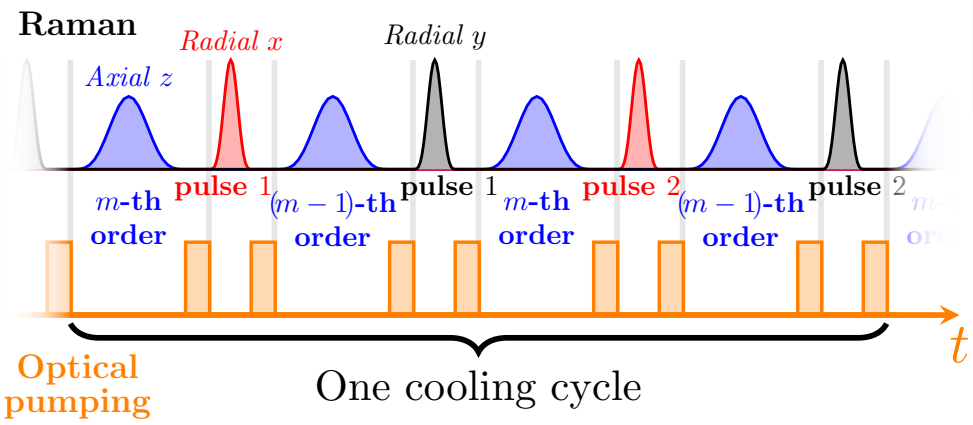

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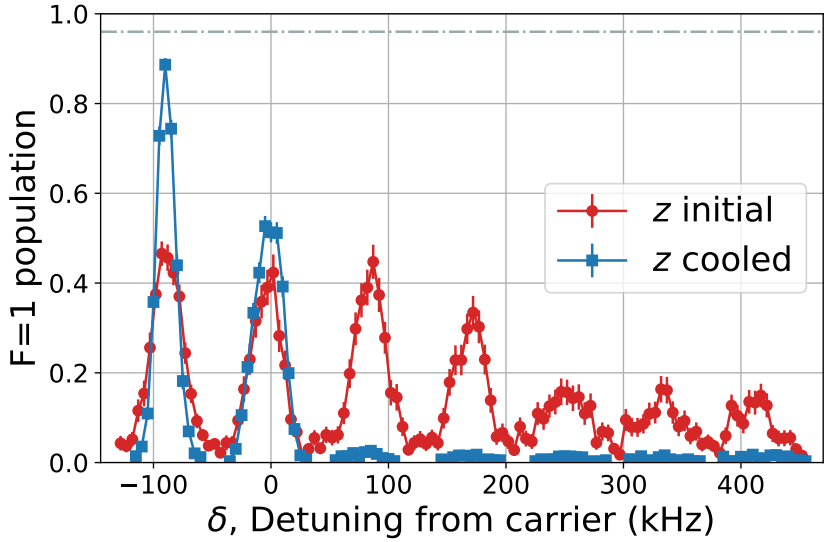

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3D ground state: 93.5(7)%

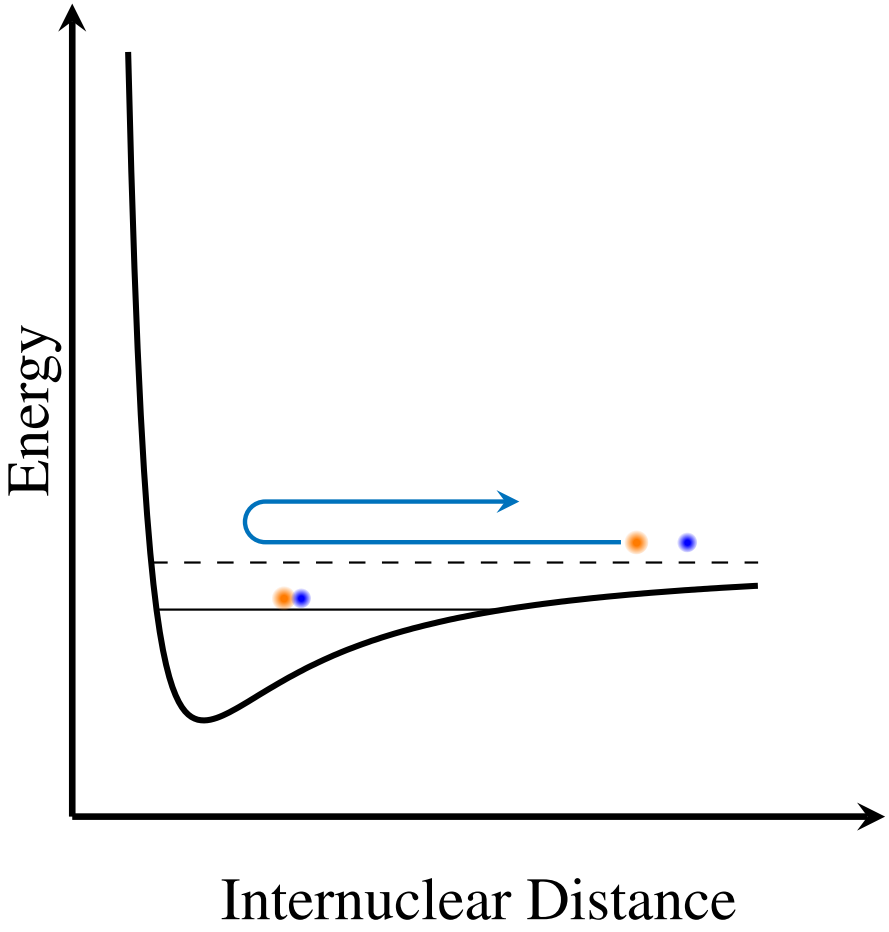
Outline

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

Scattering length a

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

⋮



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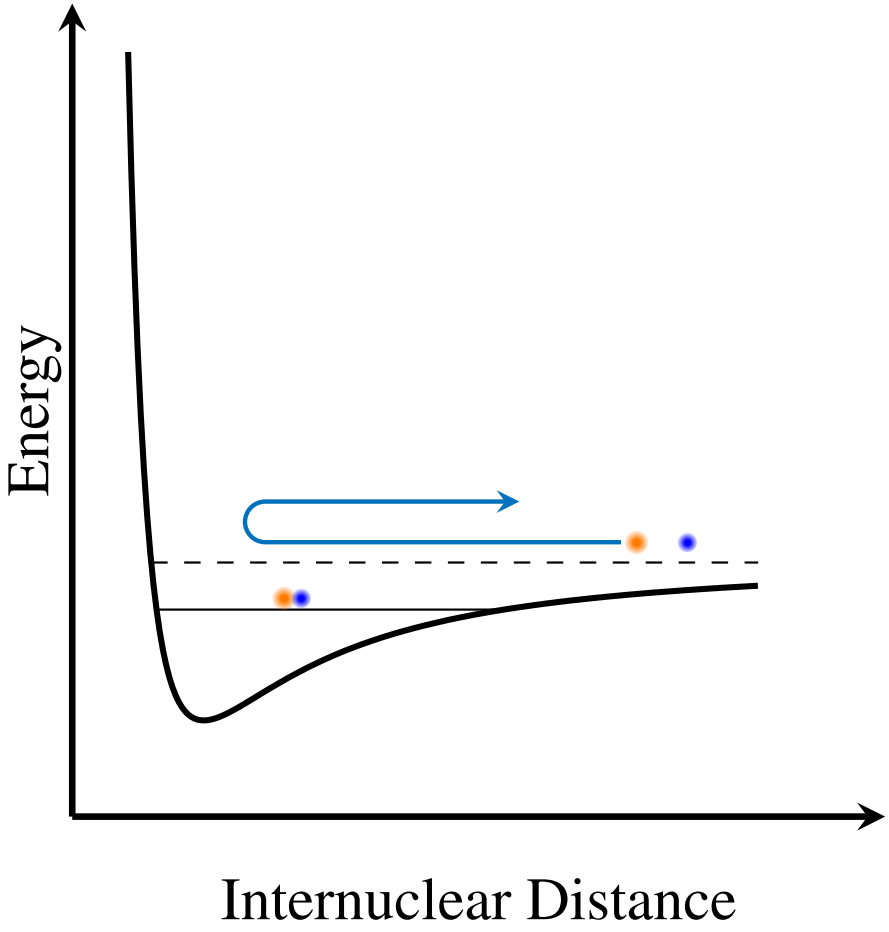

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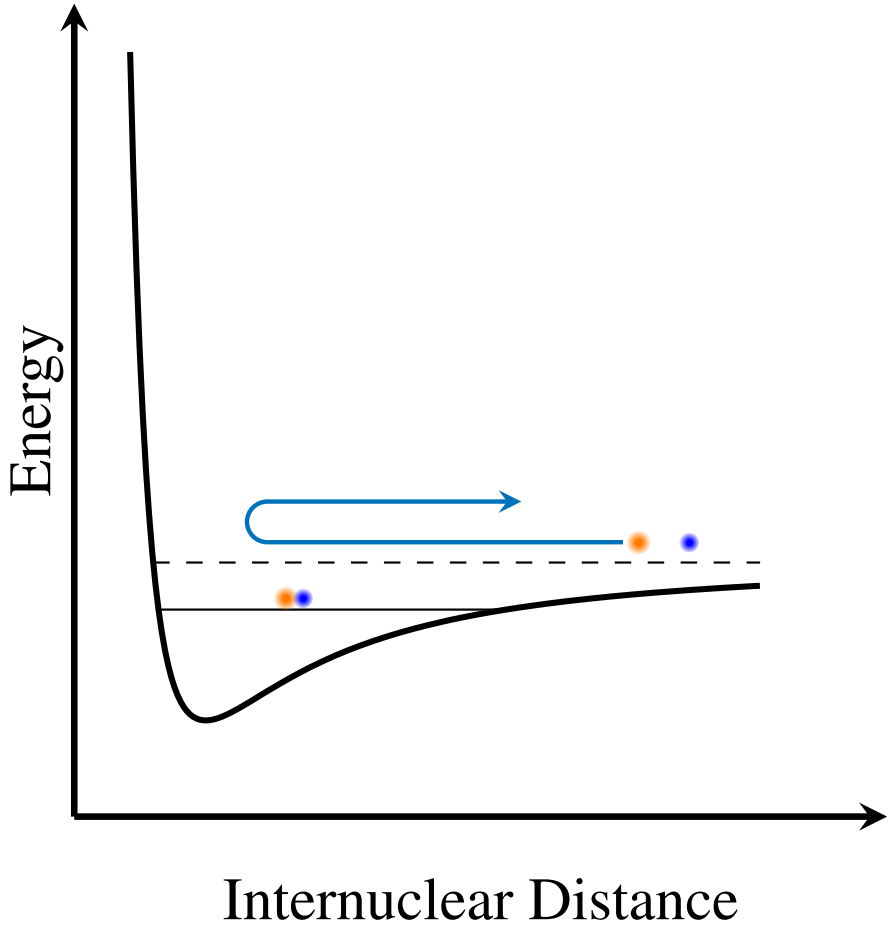

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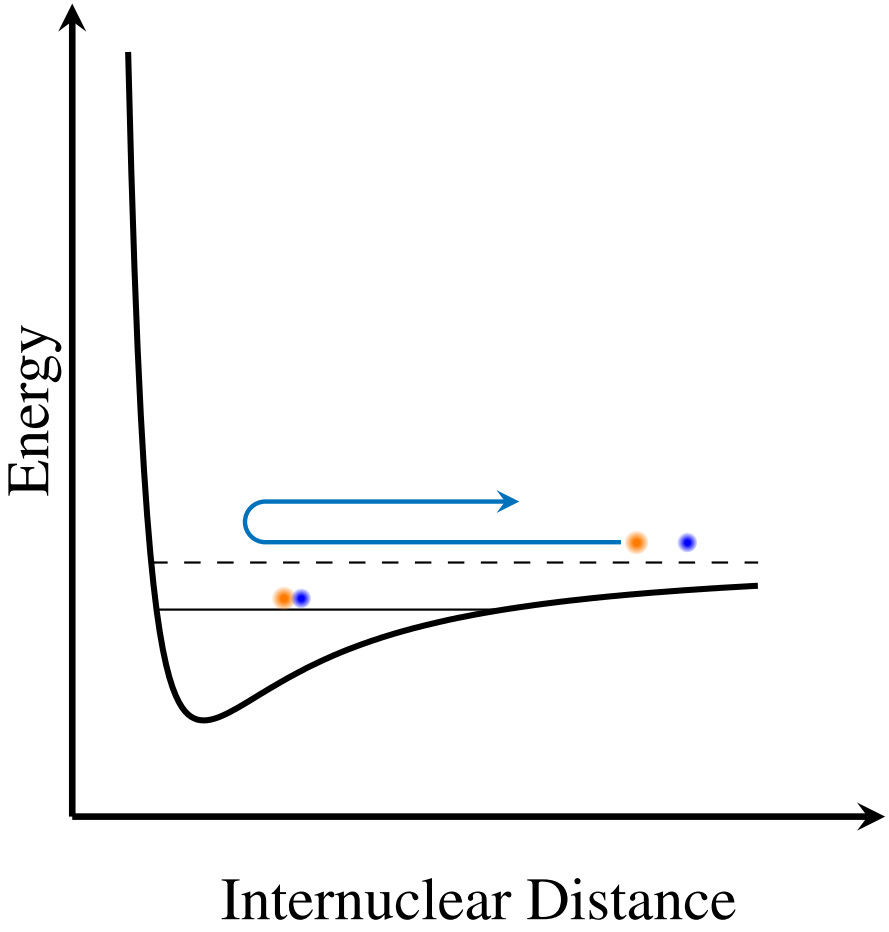

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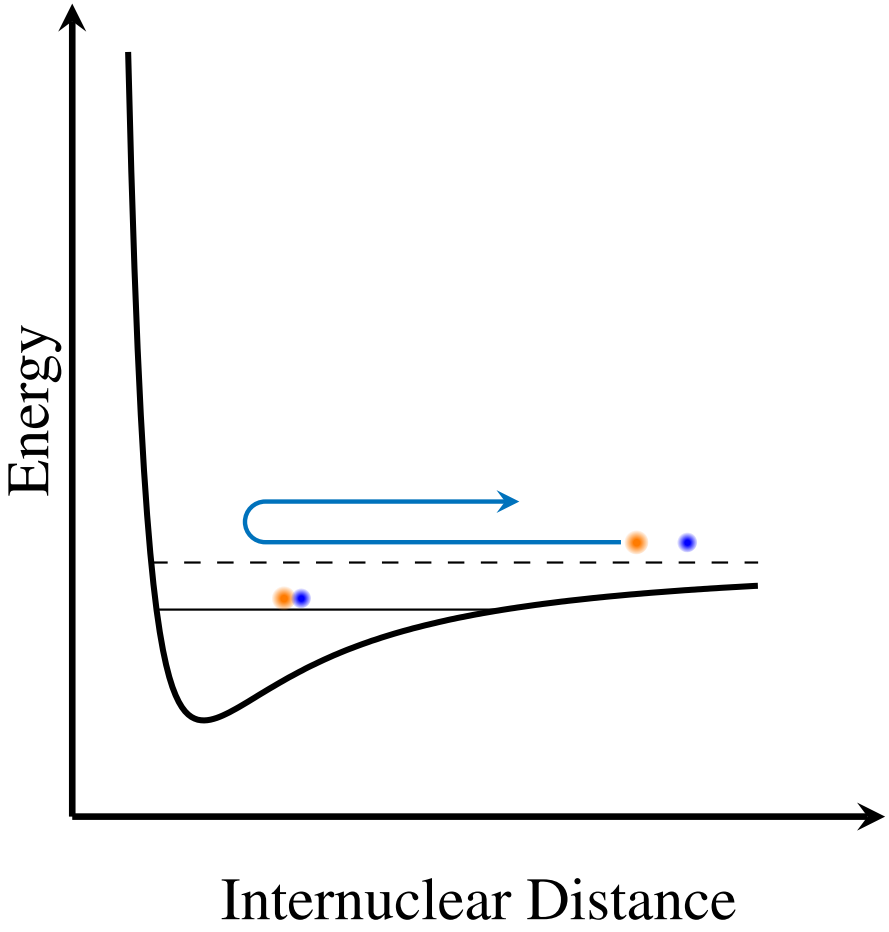
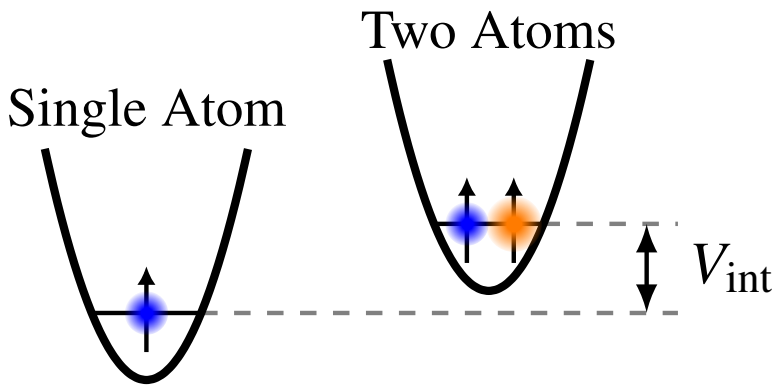


Interaction shift



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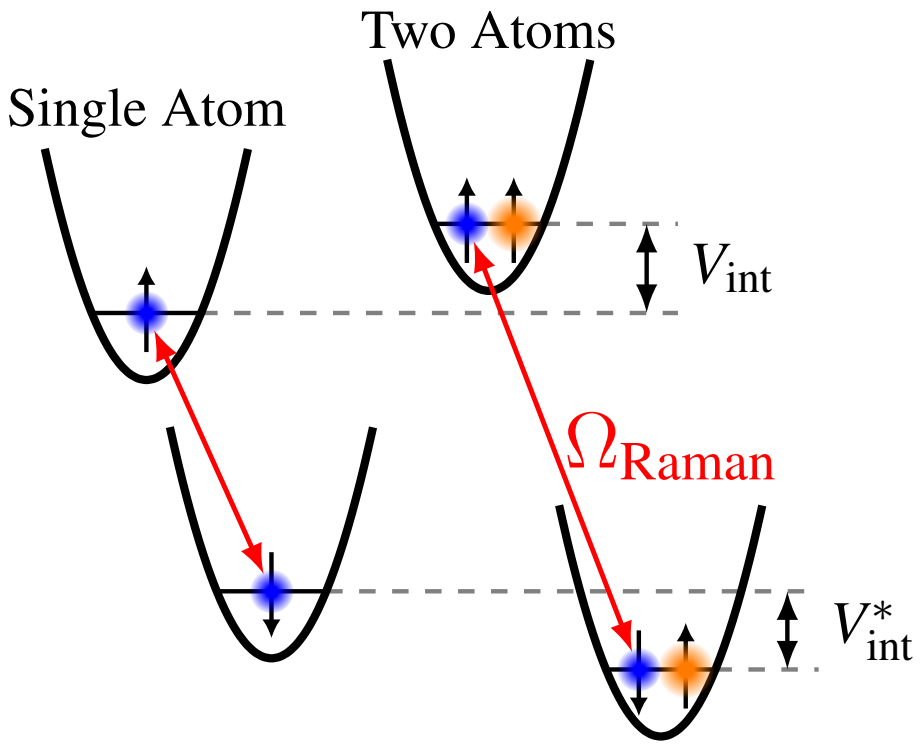

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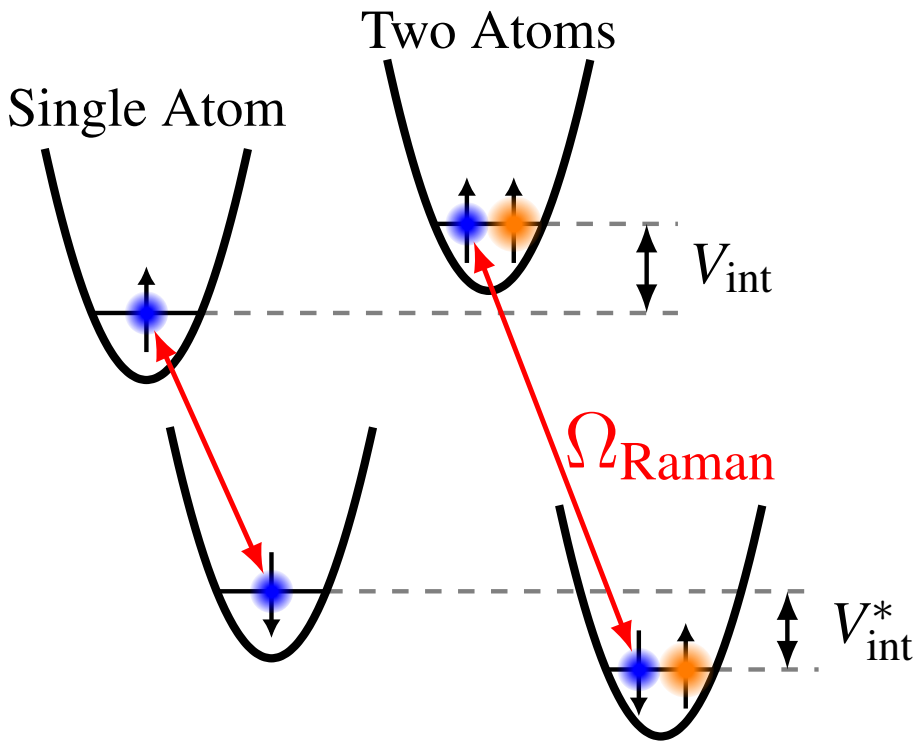

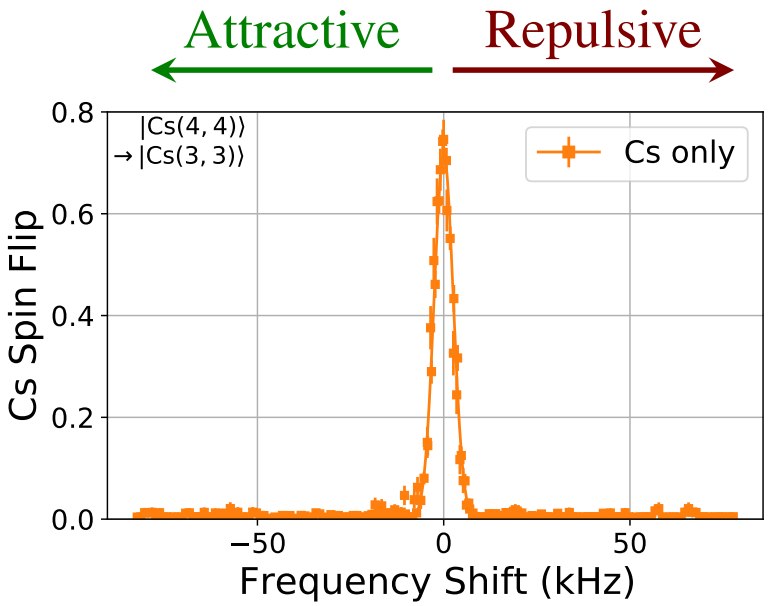

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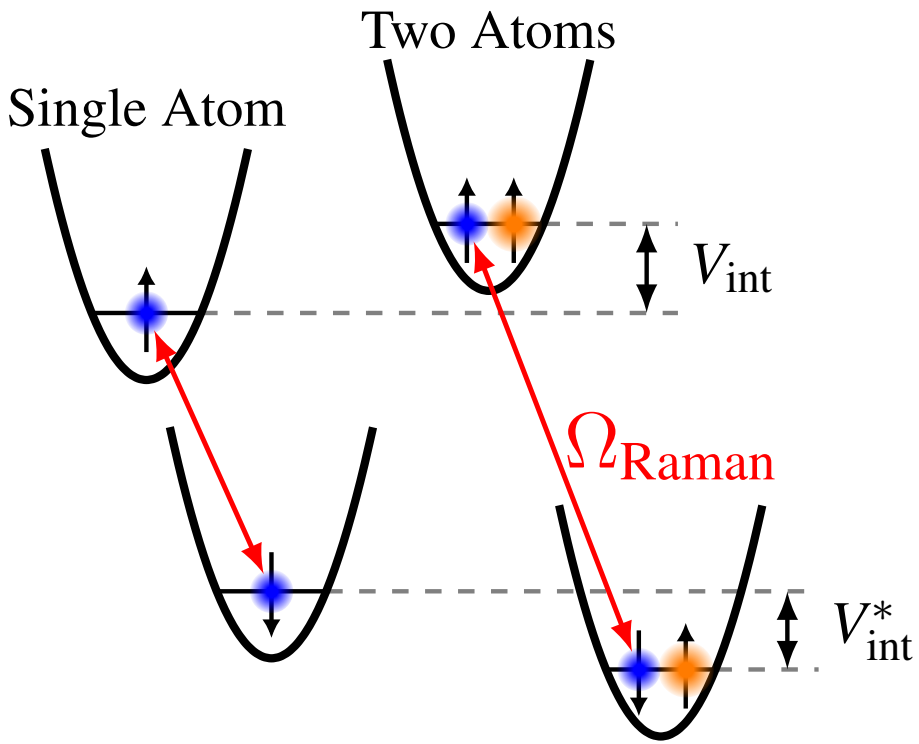

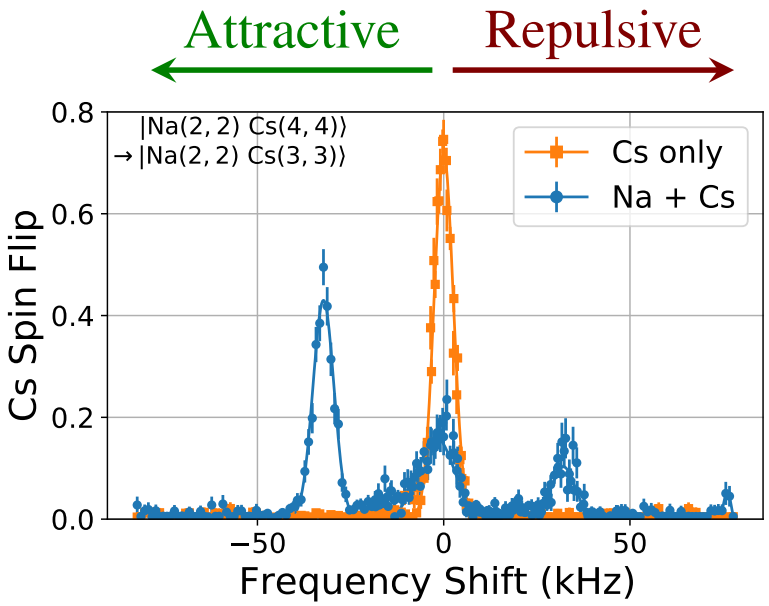

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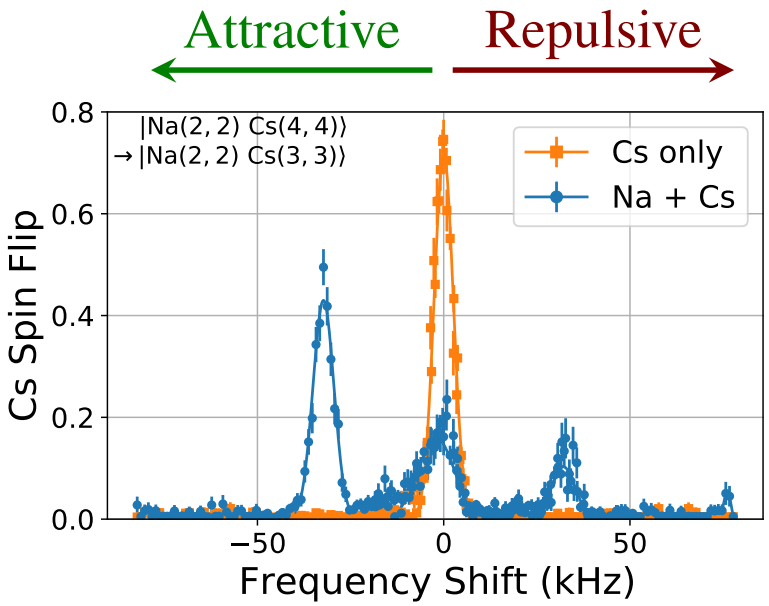
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{m_1\omega_{1,i}^2 x_{1,i}^2}{2} + \frac{p_{1,i}^2}{2m_1}\right)}_{\text{Na}} + \underbrace{\sum_{i=x,y,z} \left(\frac{m_2\omega_{2,i}^2 x_{2,i}^2}{2} + \frac{p_{2,i}^2}{2m_2}\right)}_{\text{Cs}} + \underbrace{V_{int}(\vec{r}_1 - \vec{r}_2)}_{\text{Interaction}}$$



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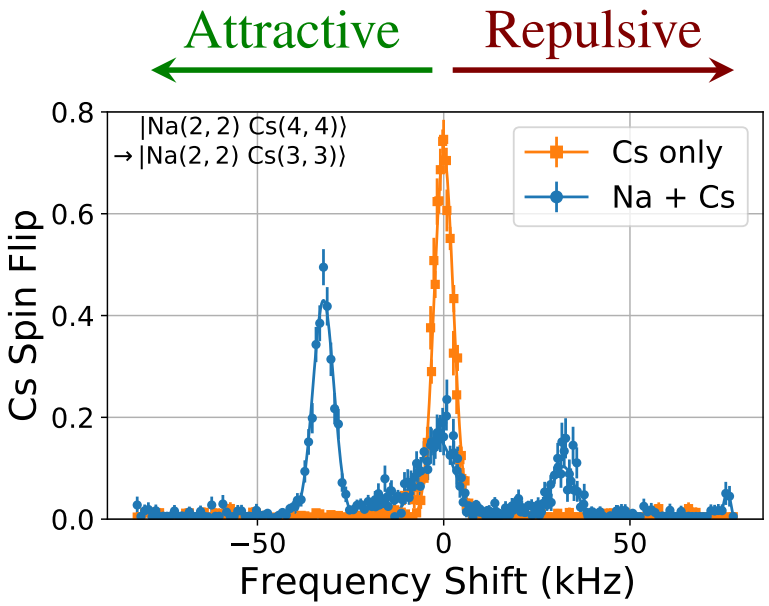
To center of mass and relative coordinates

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

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

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

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



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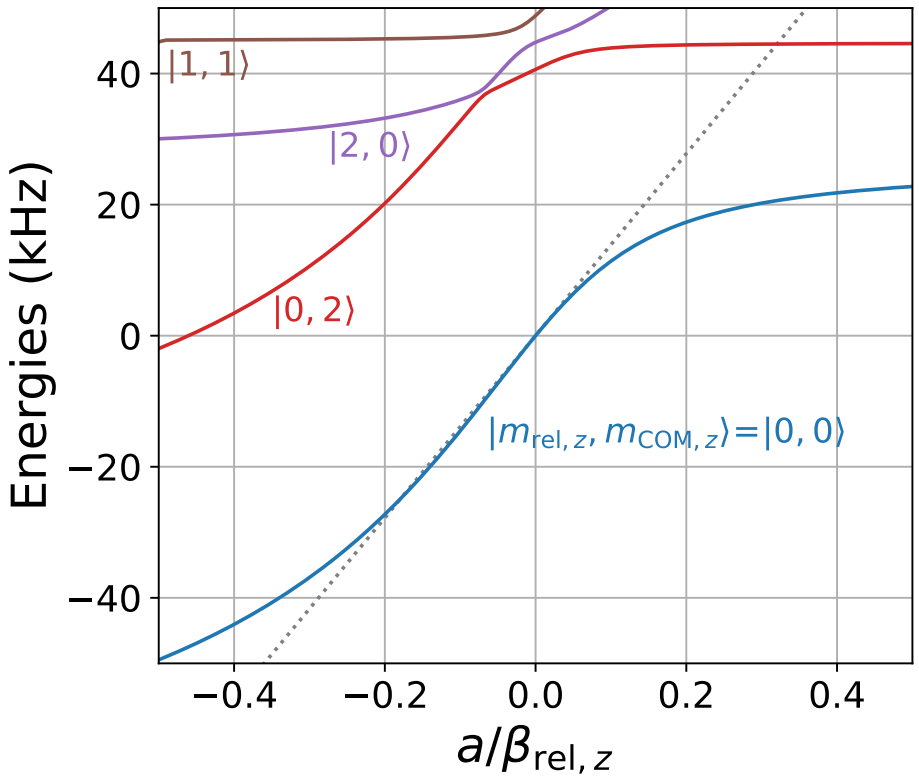

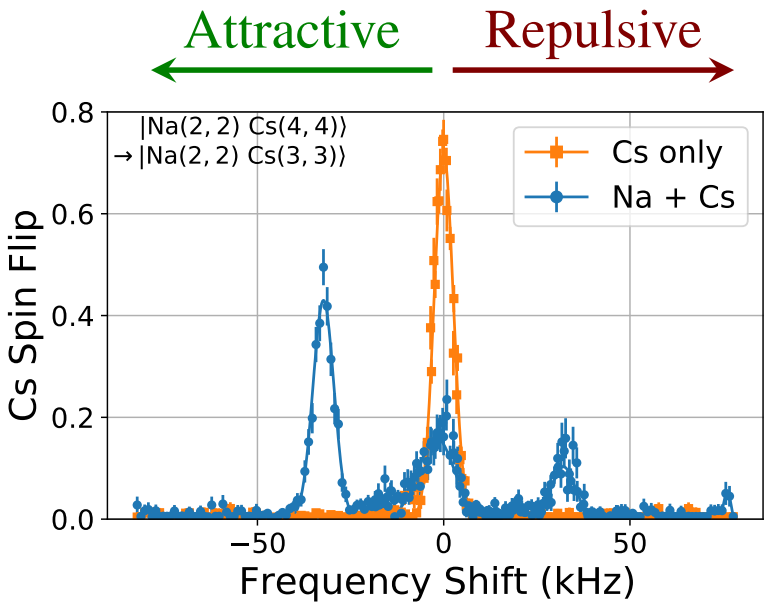

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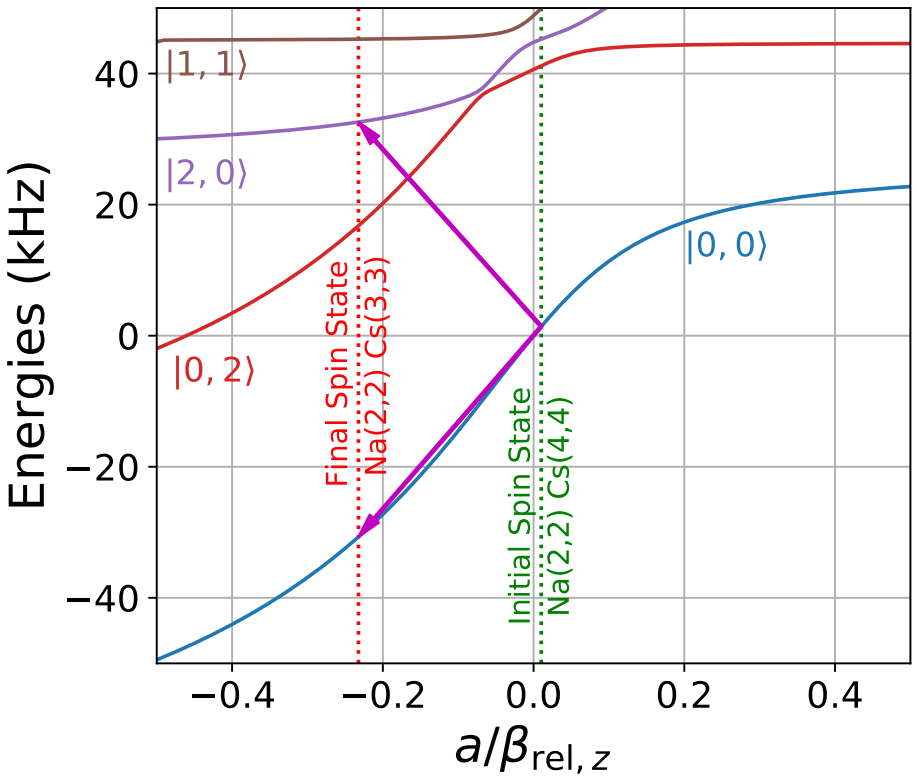
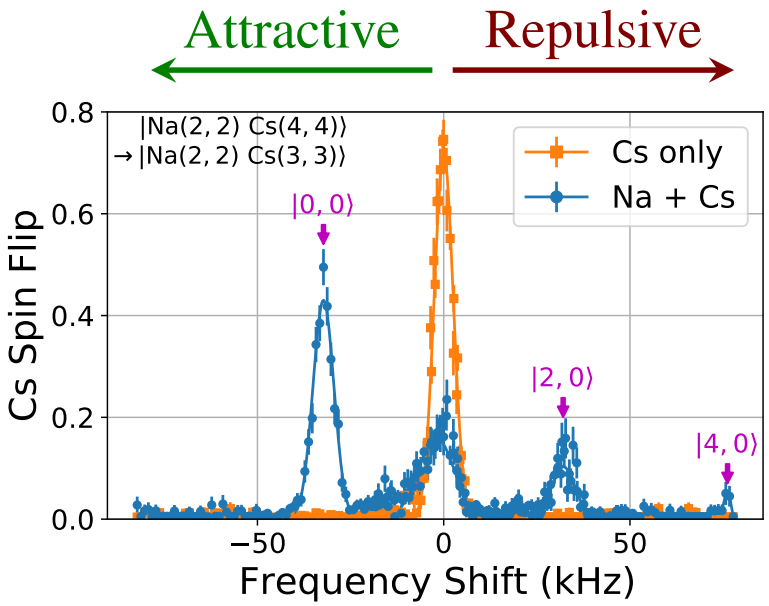
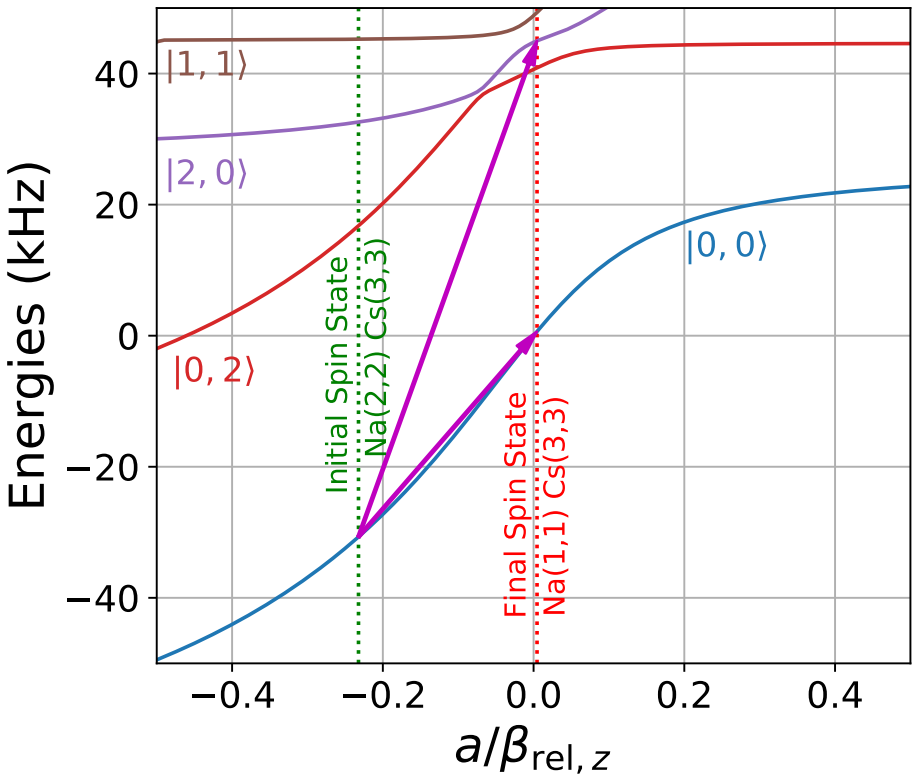
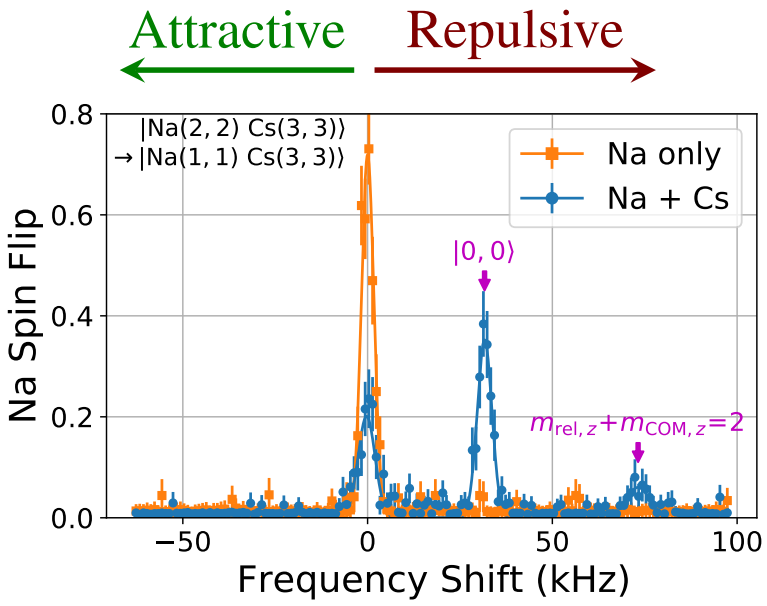




$$H = \overbrace{\sum_{i=x,y,z}\left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M}\right)}^{\text{Center of mass}} + \overbrace{\sum_{i=x,y,z}\left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu}\right) + V_{int}(\vec{r}_R)}^{\text{Relative}} + \overbrace{\sum_{i=x,y,z}\mu(\omega_{1,i}^2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

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

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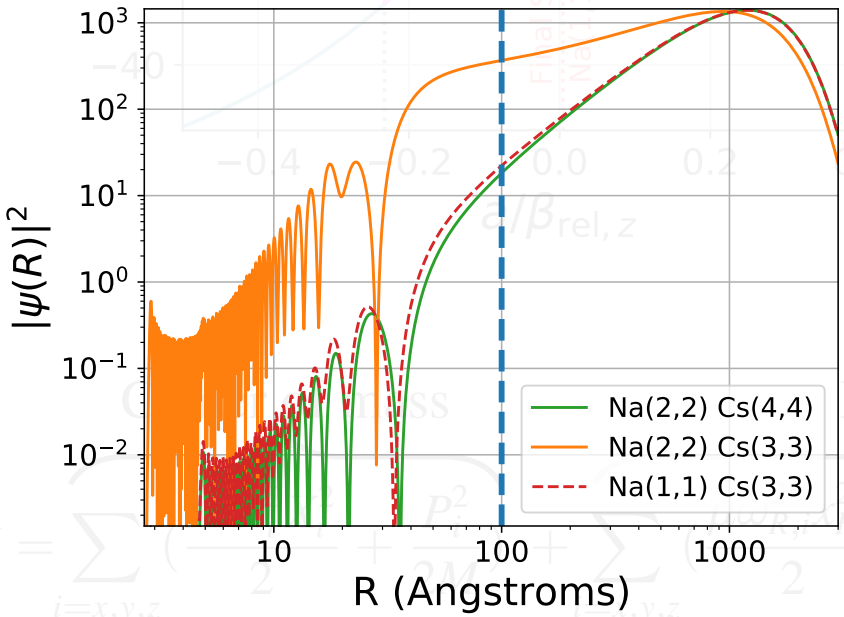
Enhanced coupling to molecular state!



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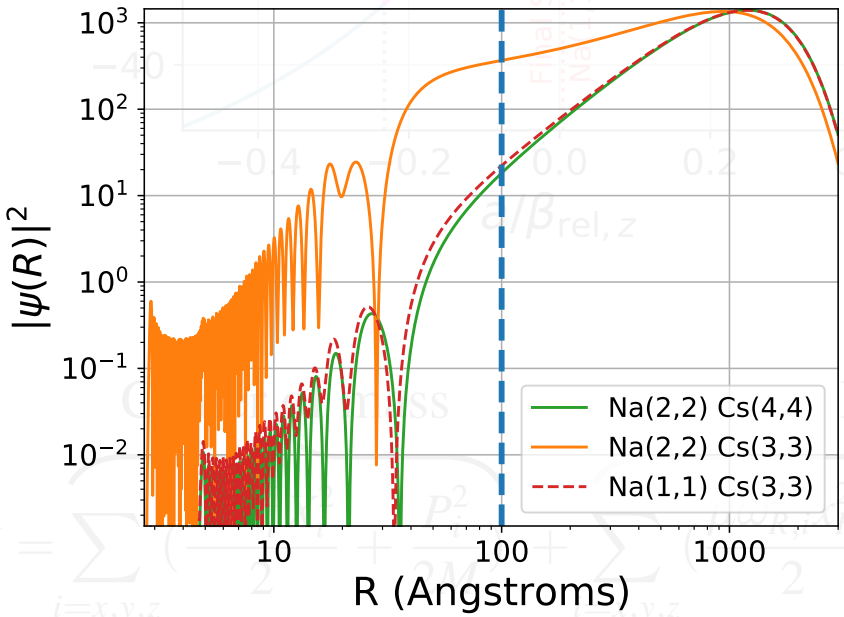

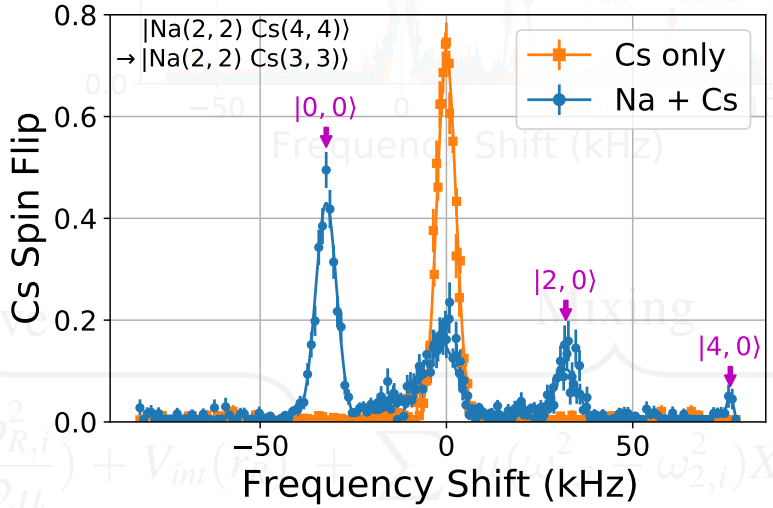

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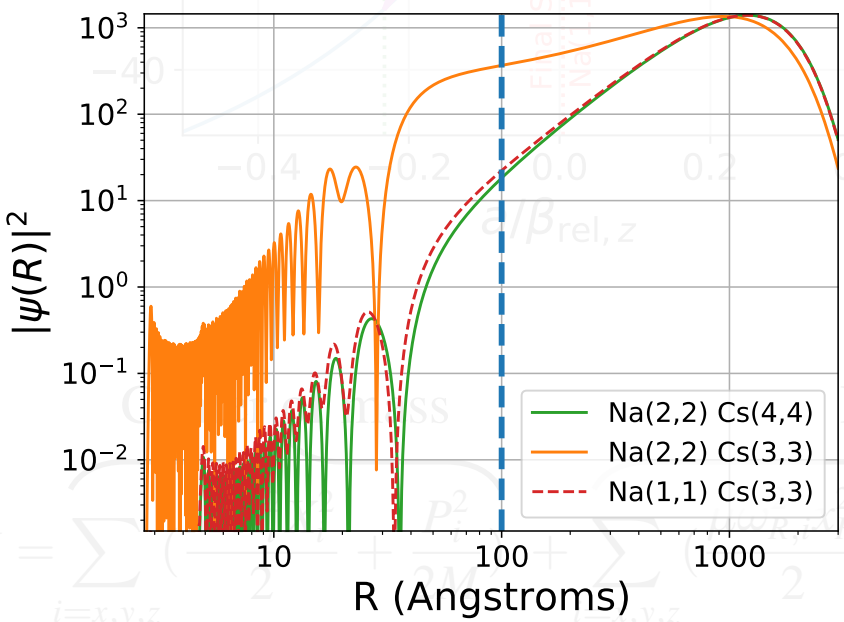
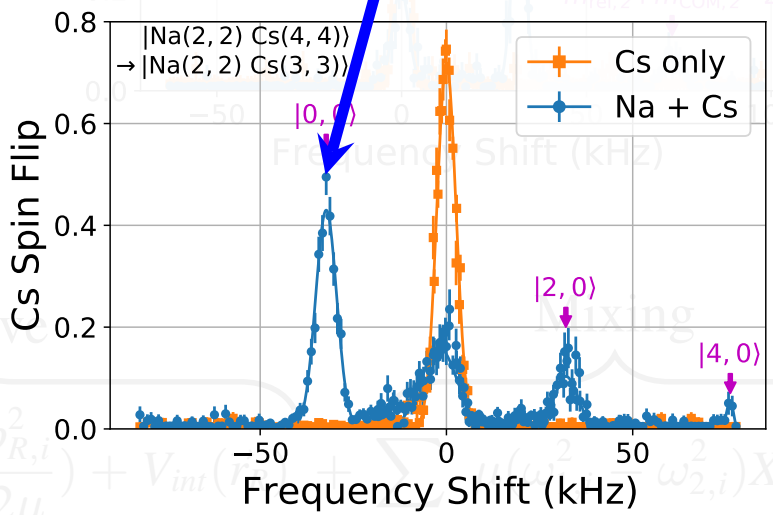


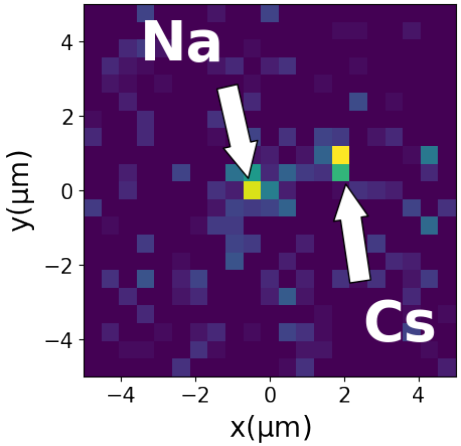
Ground State Only



Outline

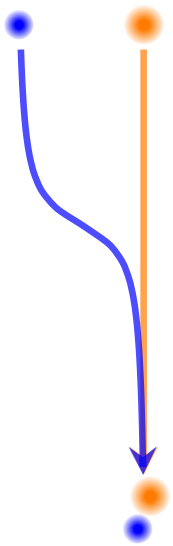
1. Experiment overview
2. Atom state control
   - Raman sideband cooling of Na atoms
3. **Molecule creation**
   - Atom-atom interaction
   - Coherent optical transfer
4. Conclusion

Loading

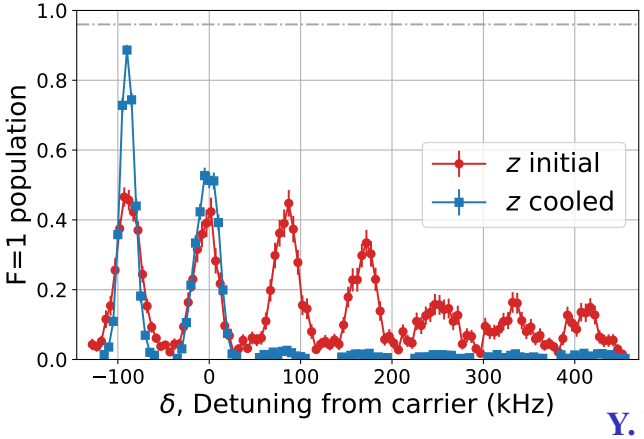


NJP. 19, 023007 (2017)

Merging



Cooling



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

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

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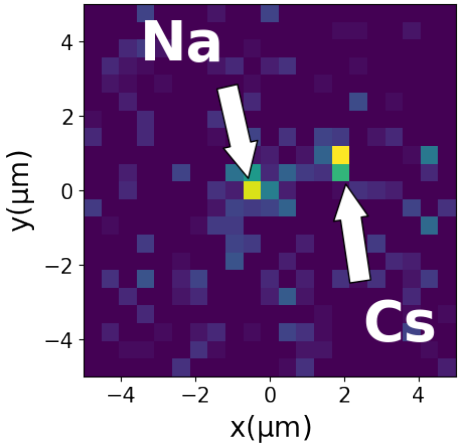
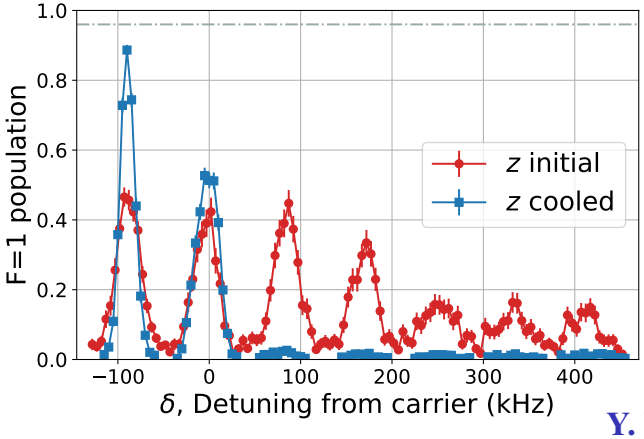


NJP. 19, 023007 (2017)

Cooling



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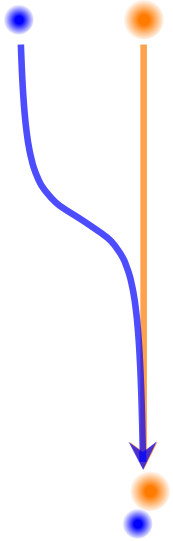

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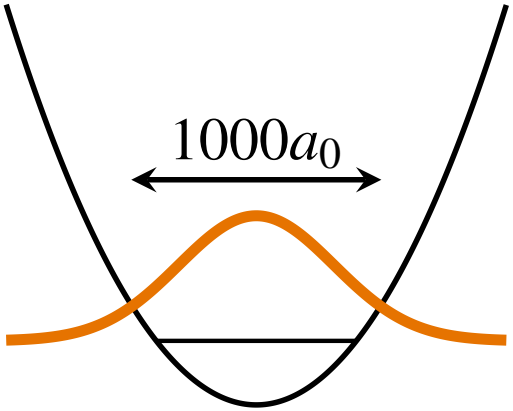
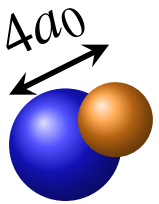
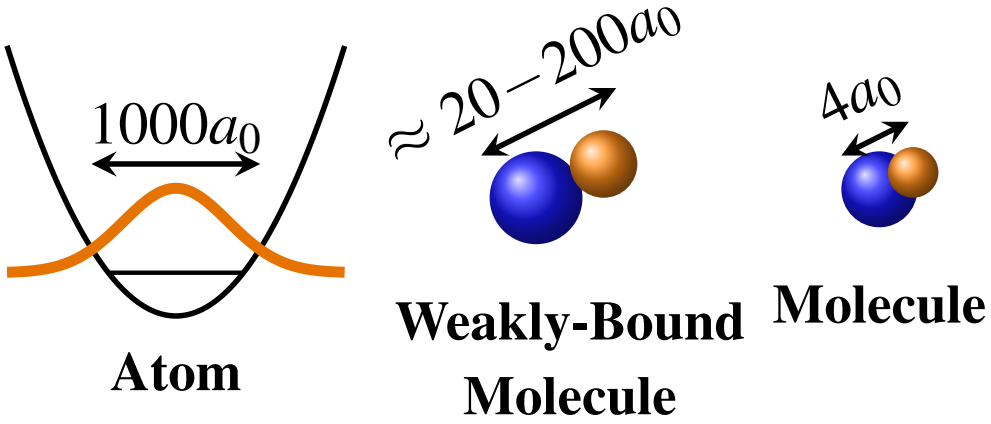
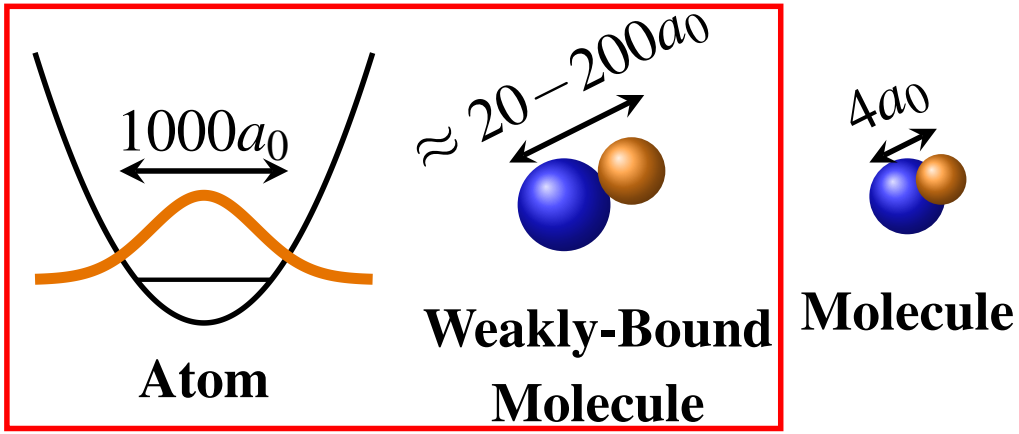


Atom



Molecule


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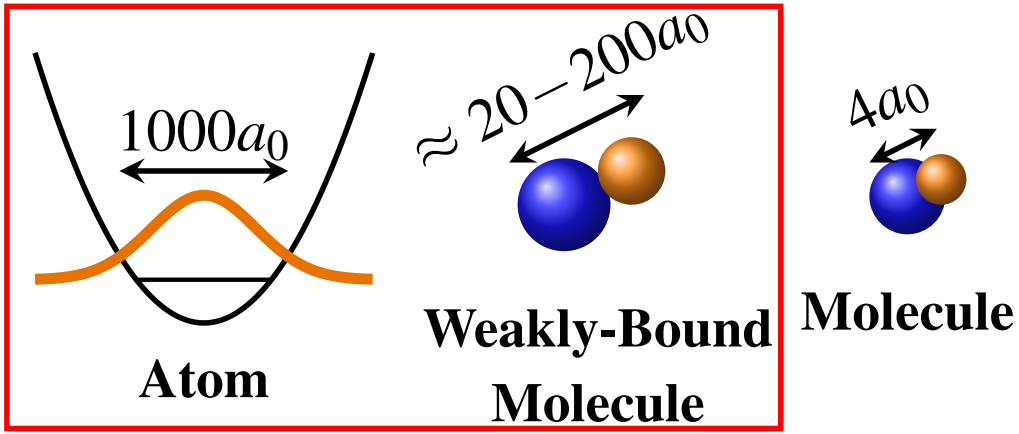
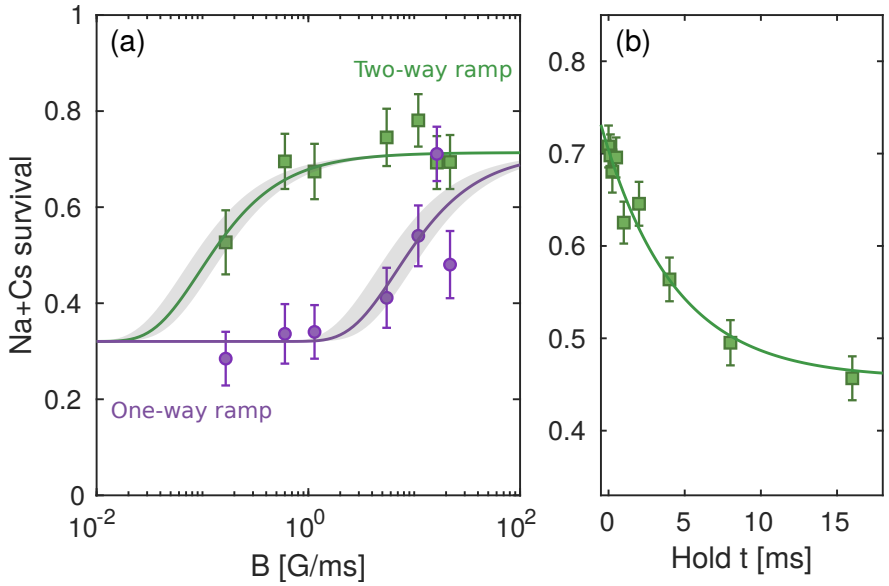
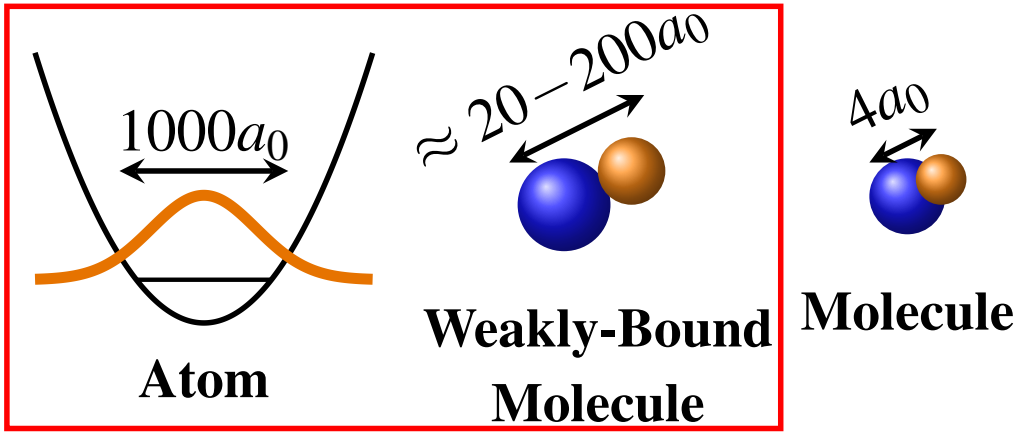


Feshbach molecule



PRL. 124, 253401 (2020)



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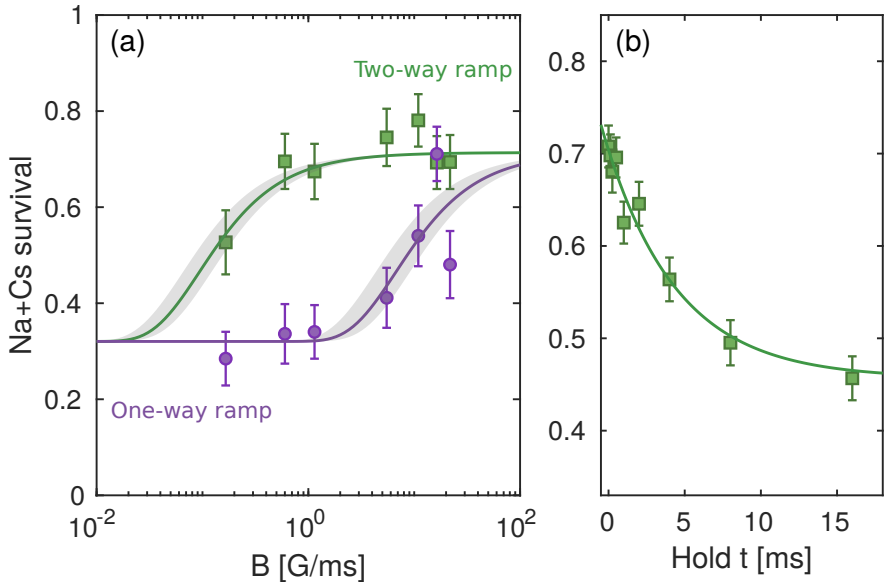
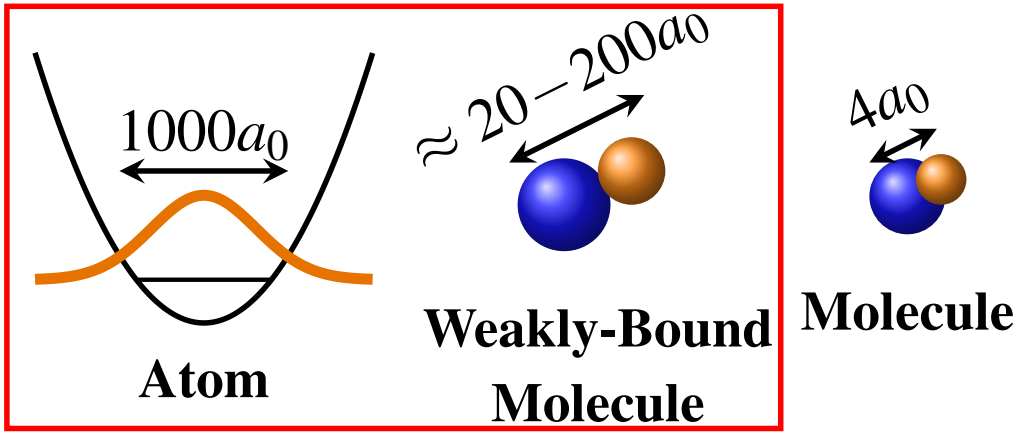


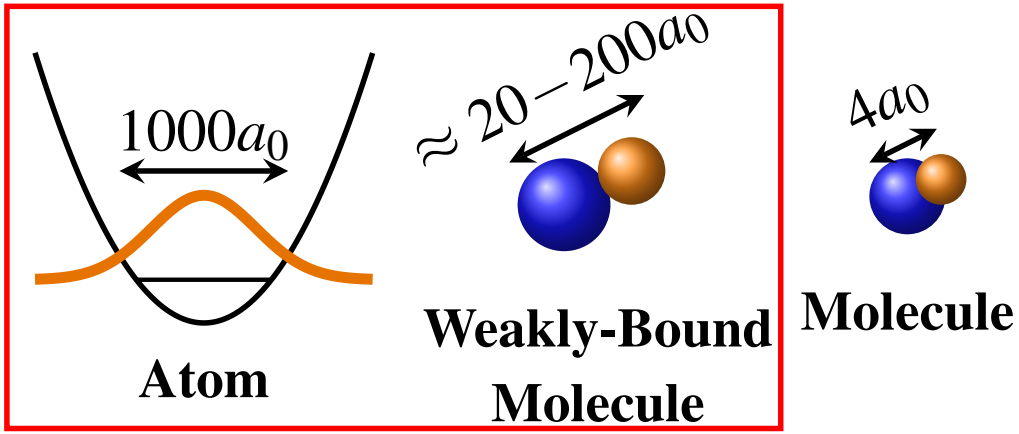
PRL. 124, 253401 (2020)

- Requires Feshbach resonance
- Usually large magnetic field



Optical transfer

- More general
- Faster



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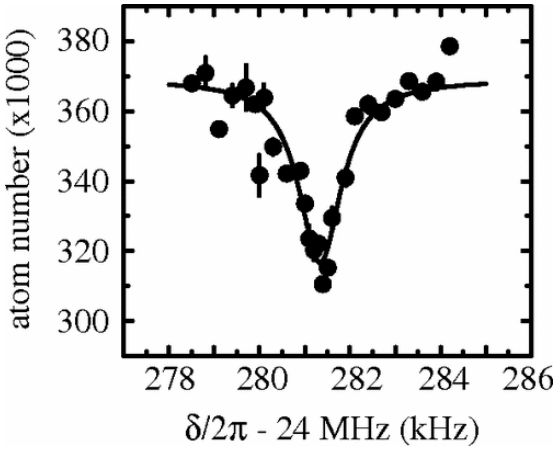
Previous results

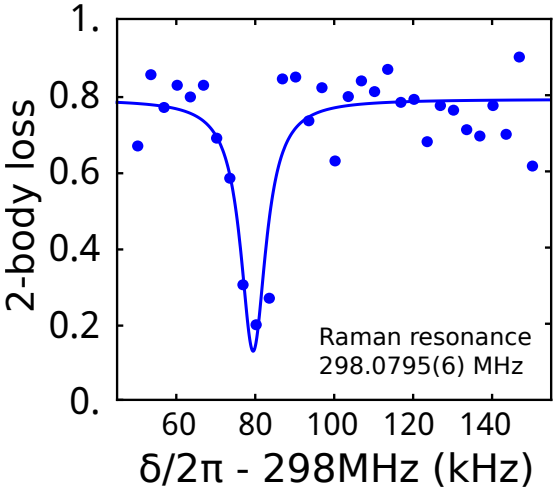
Rb_2 Science 287, 1016 (2000)

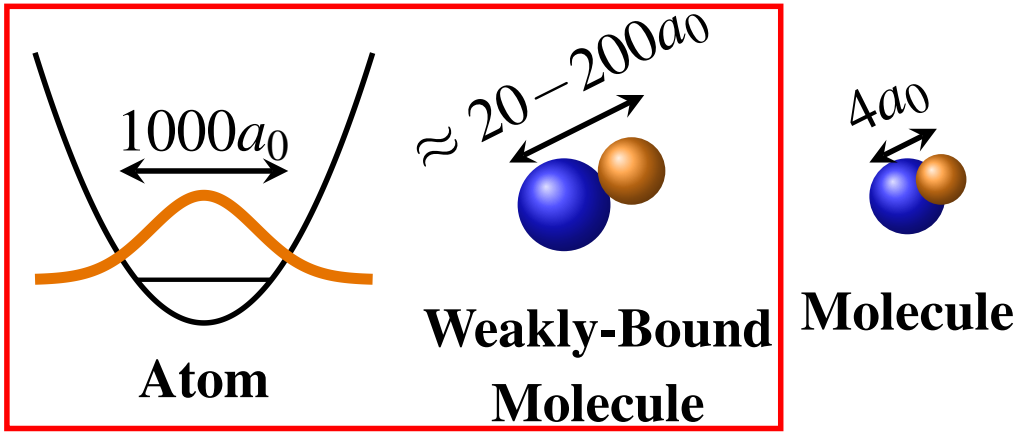
PRL. 93, 073002 (2004)



Sr_2 PRL. 109, 115302 (2012)

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





Optical transfer

- More general
- Faster

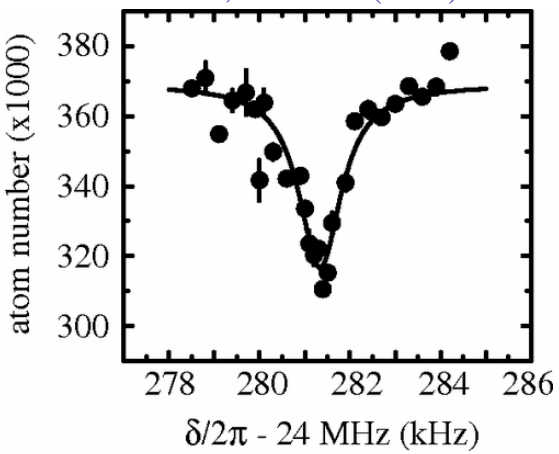
Limitations so far

- 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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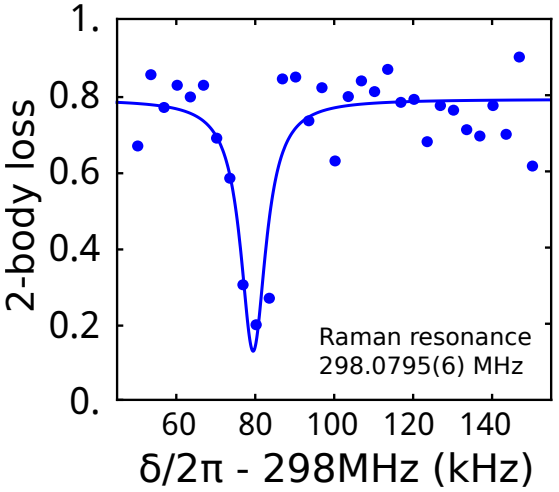
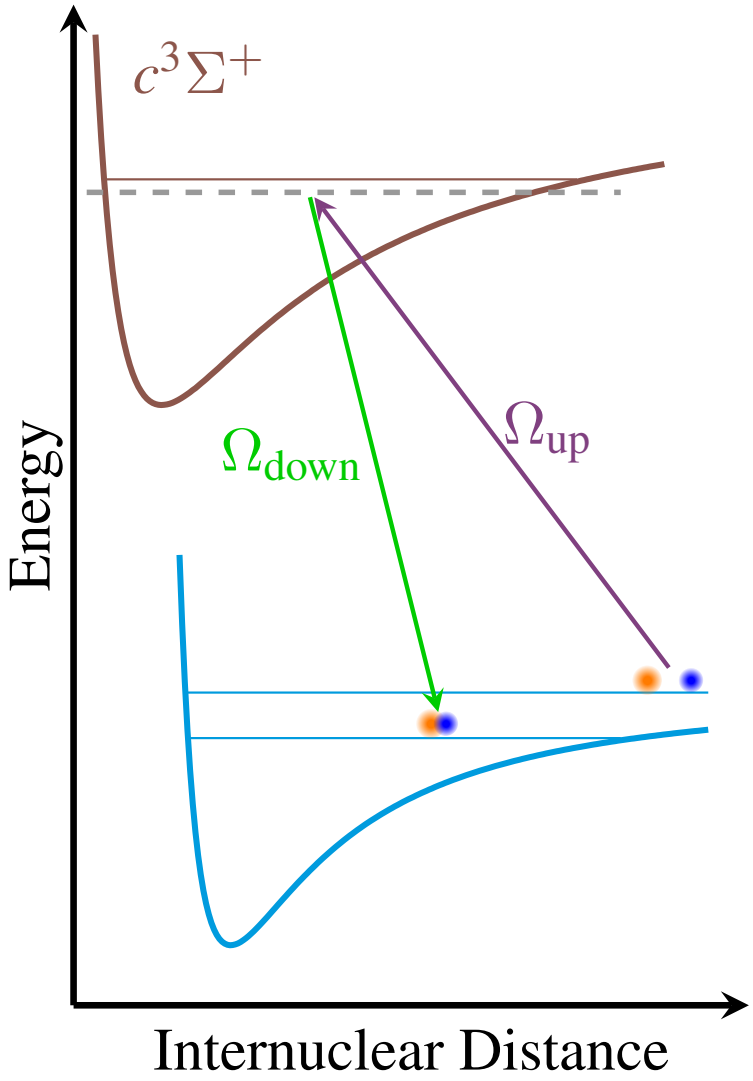
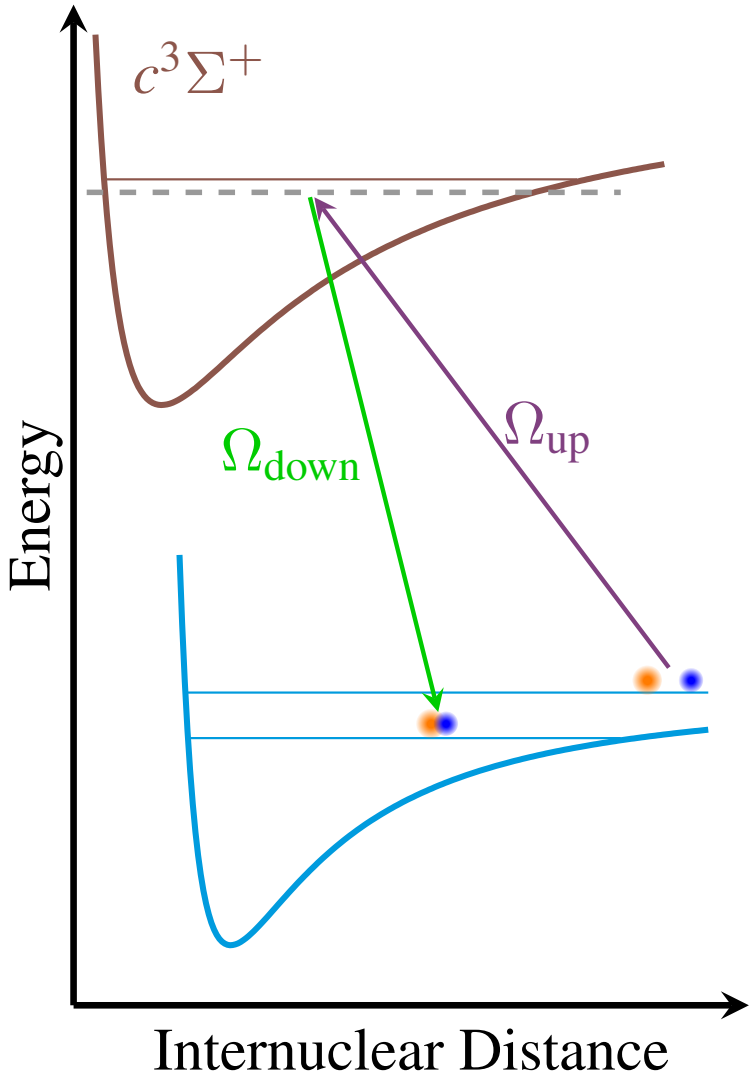


Raman transfer



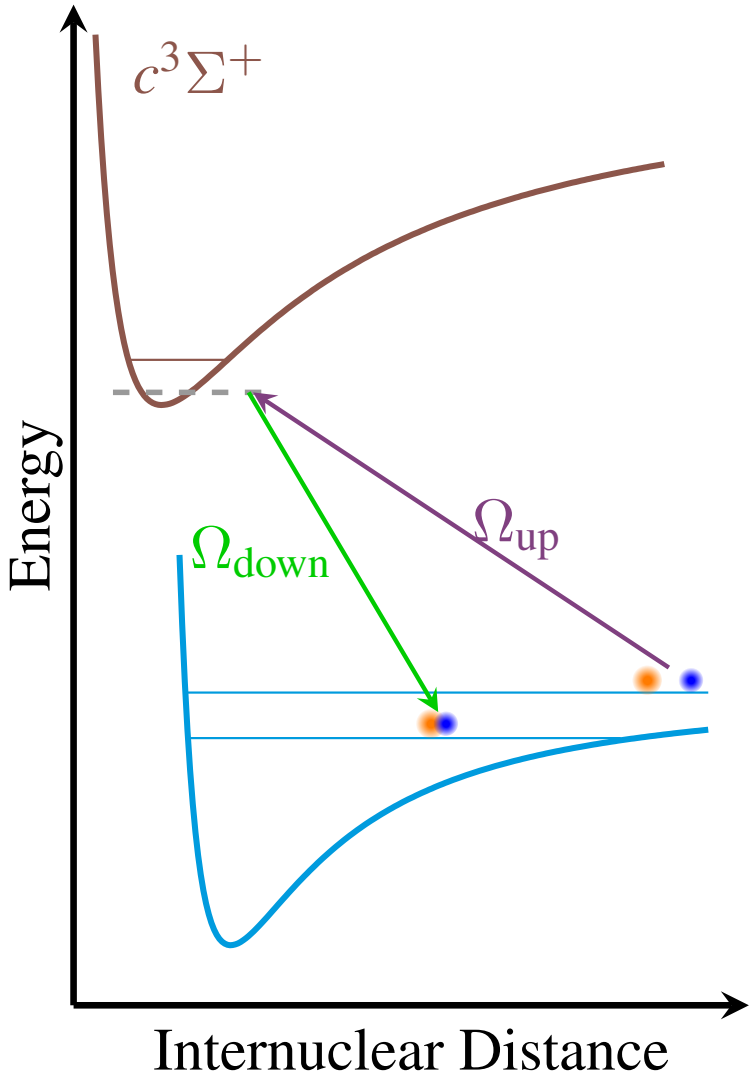
Raman transfer



Near threshold states

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

Raman transfer



Near threshold states

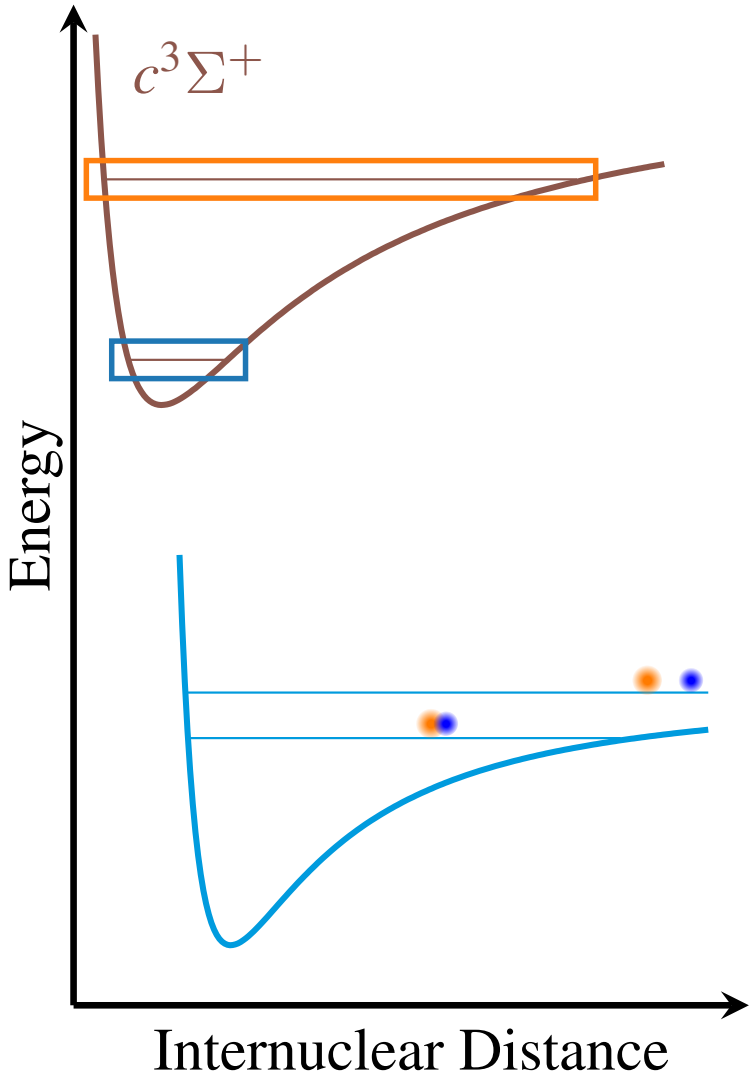
- Stronger coupling (Ω_{up} and Ω_{down})
- Closely spaced
- Fast scattering

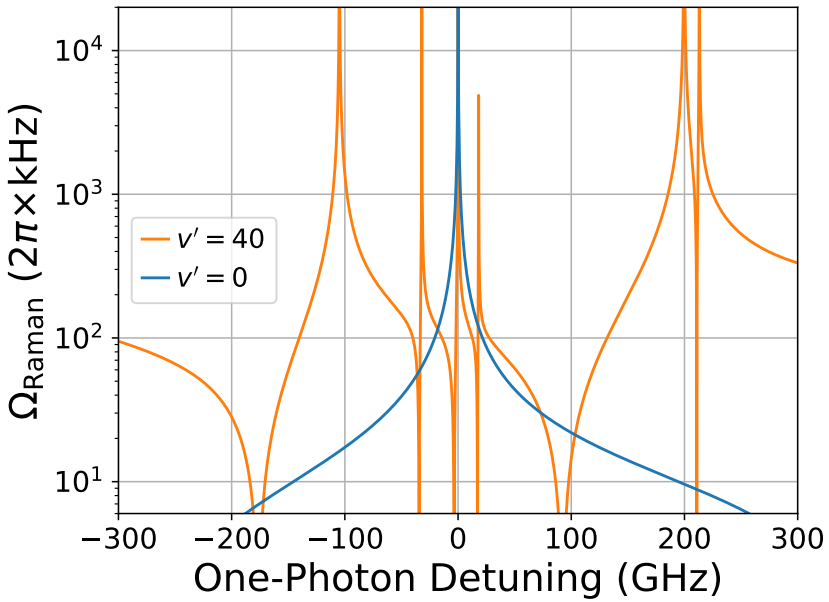
Deeply bound states

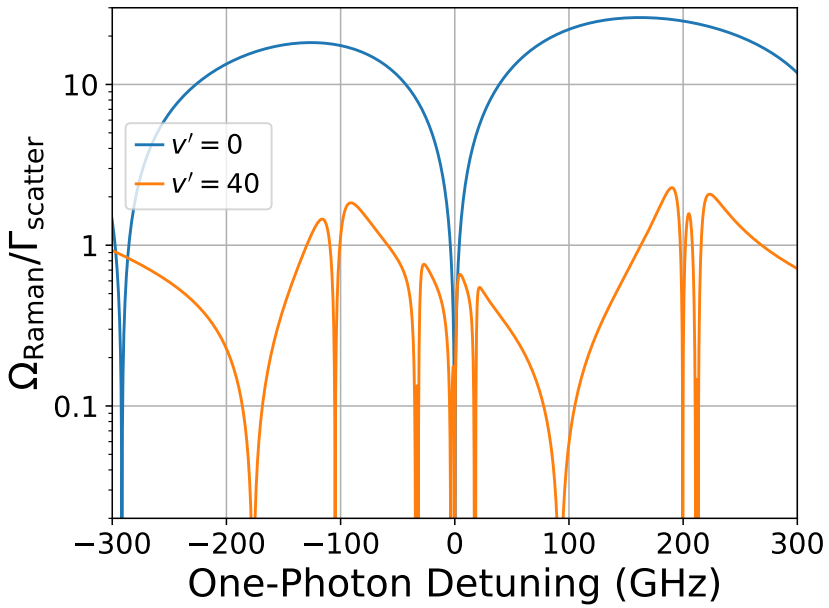
- Weaker coupling
- Sparsely spaced
- Allow larger detuning
- Slower scattering

arXiv:1701.03121(2017)

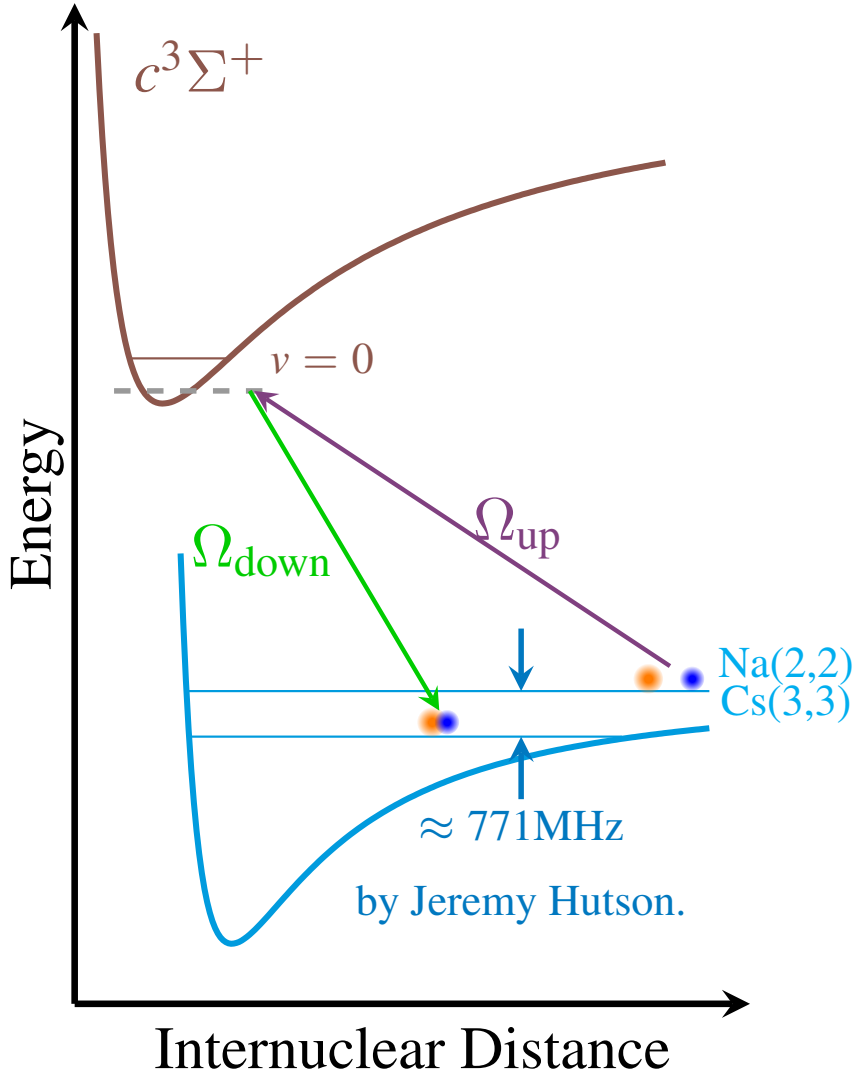
Raman transfer



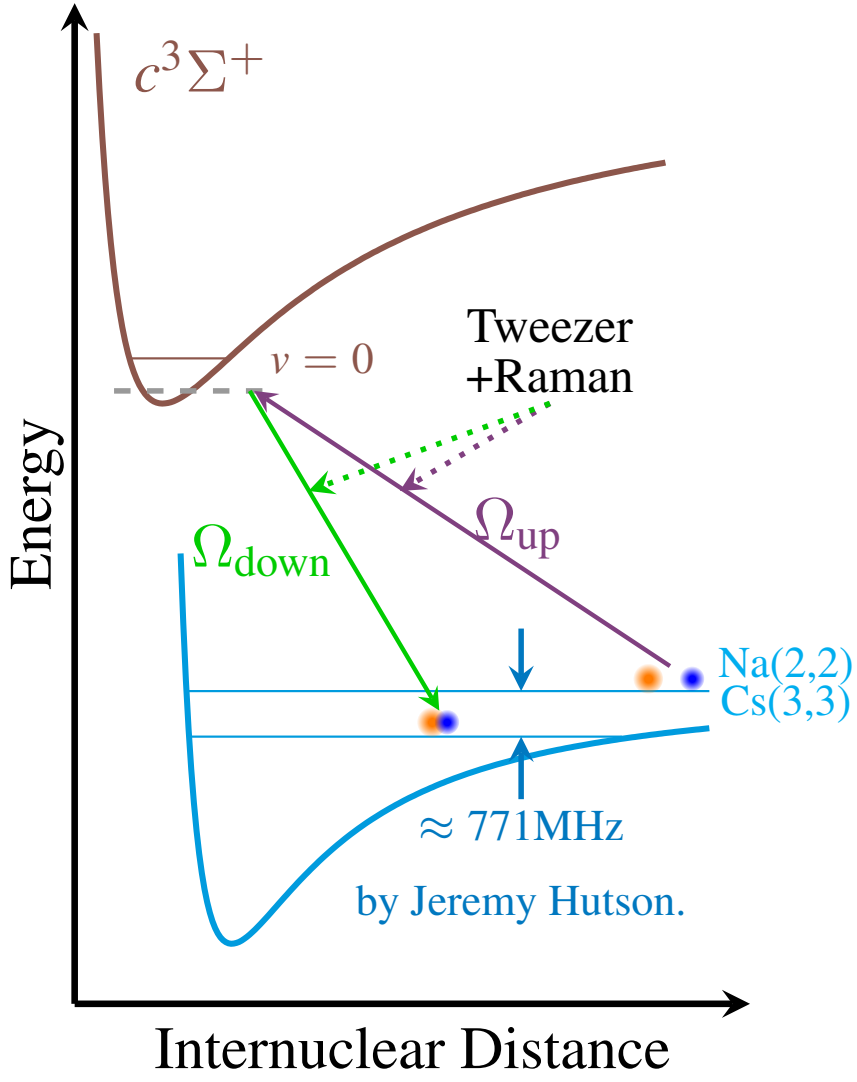




Experiment


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

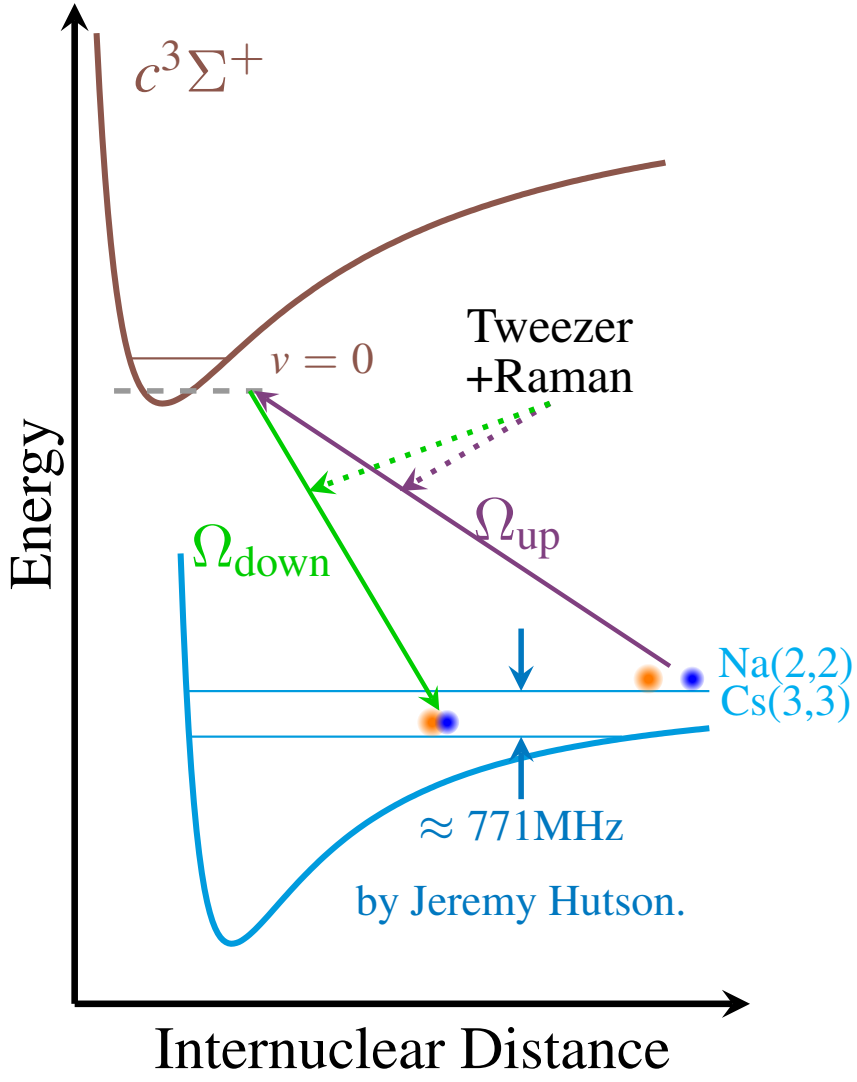
Experiment

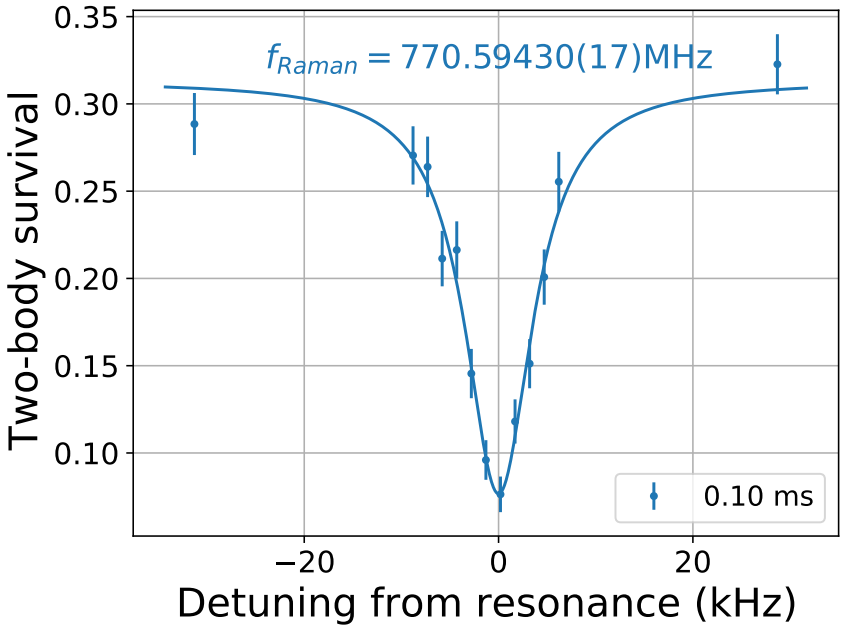


Tweezer as Raman beam

- Higher Raman Rabi frequency
- Lower scattering from other sources

Experiment





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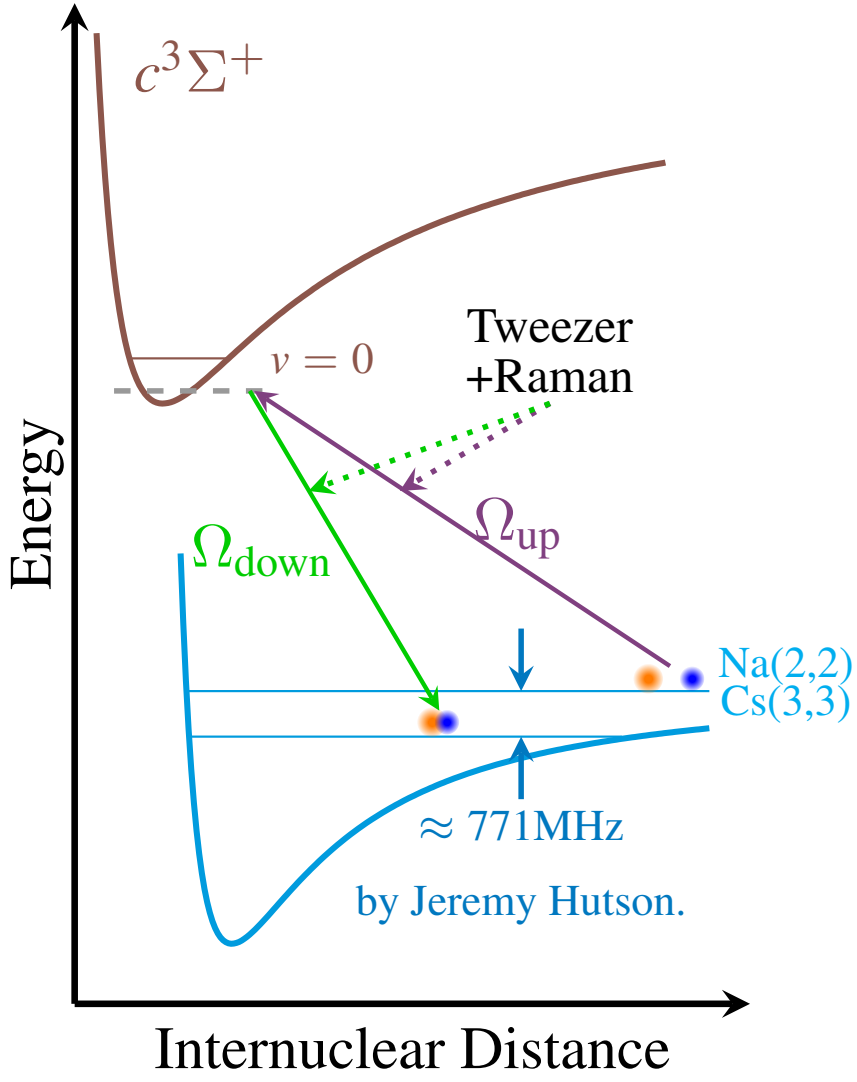

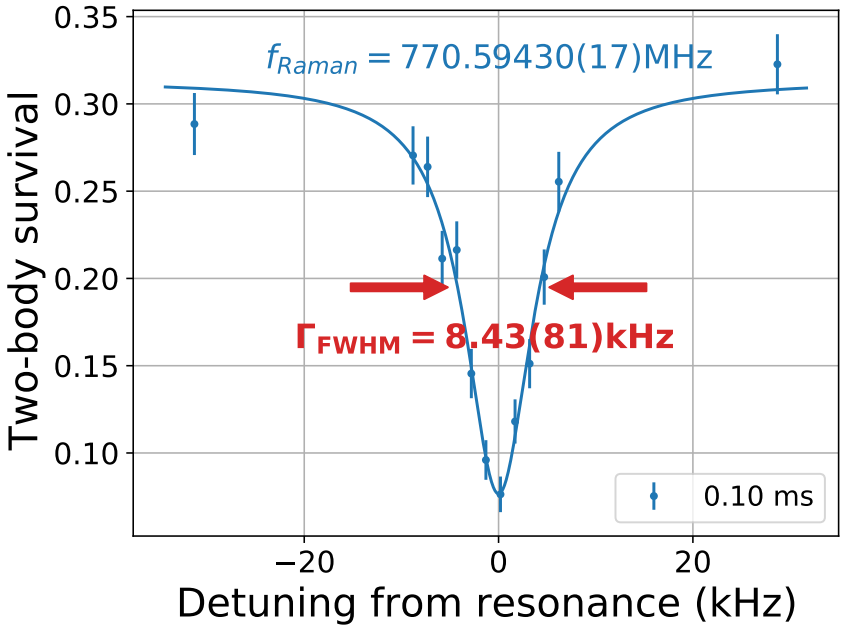

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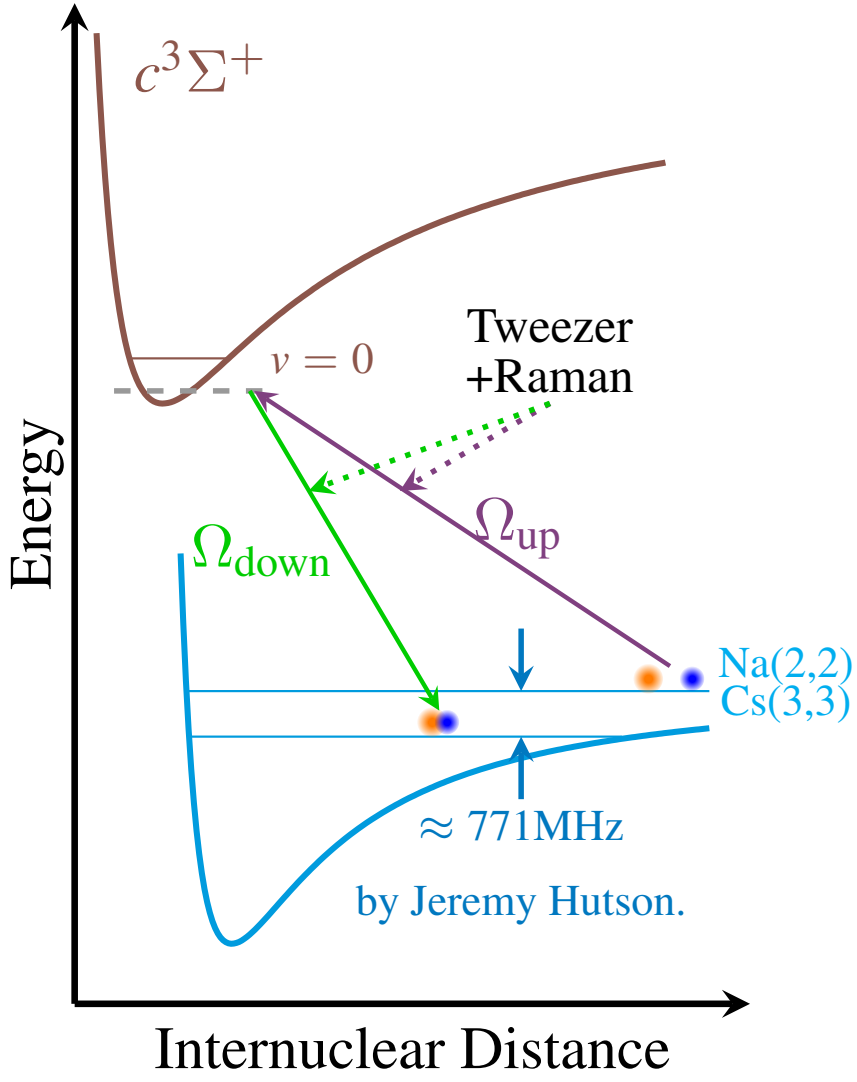
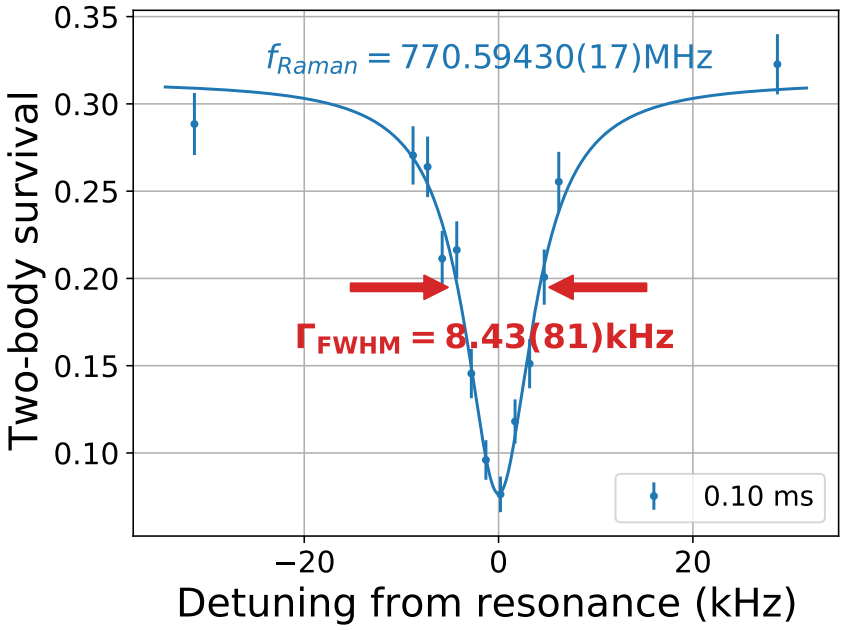
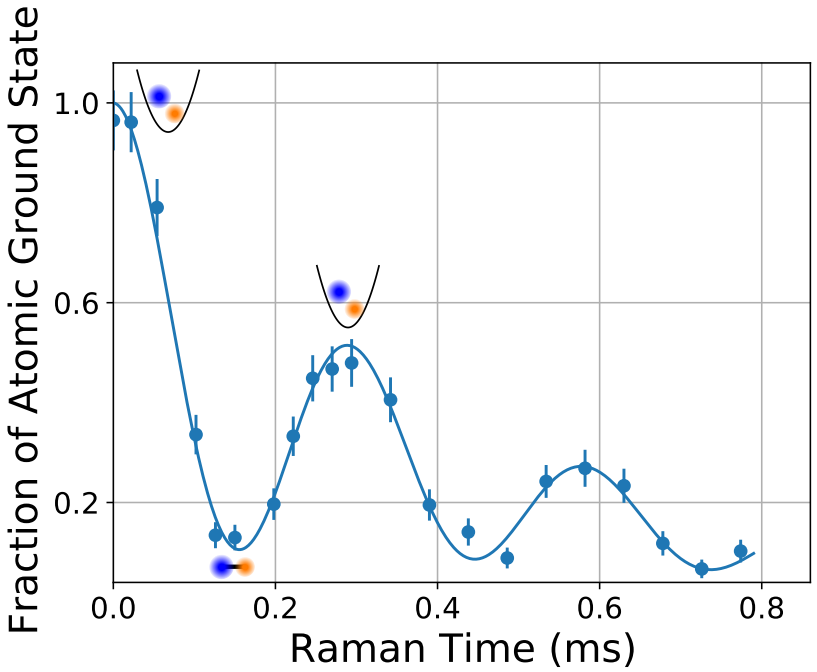
Experiment

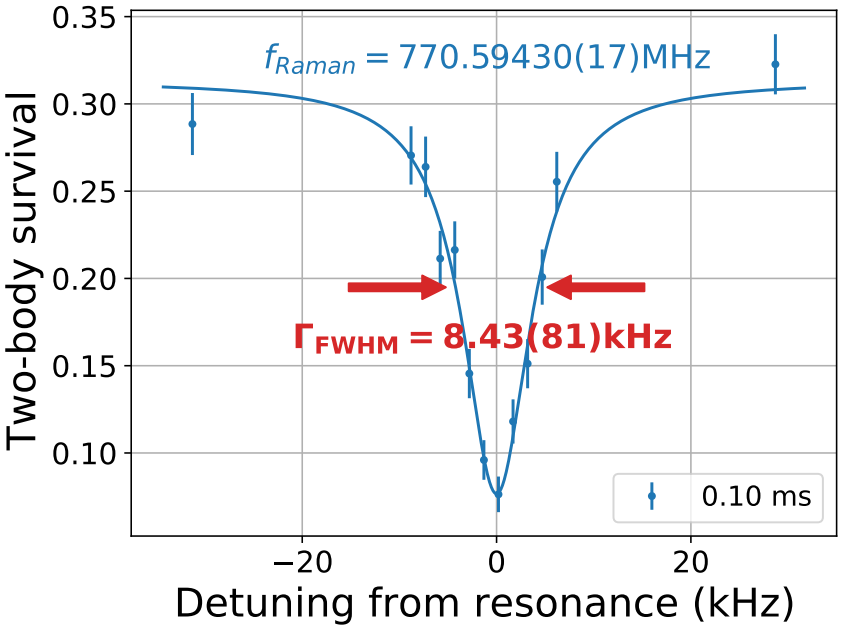

$c^3\Sigma^+$
$v = 0$
Tweezer
+Raman
Ω_{up}
Ω_{down}
Na(2,2)
Cs(3,3)
≈ 771MHz
by Jeremy Hutson.
Energy
Internuclear Distance

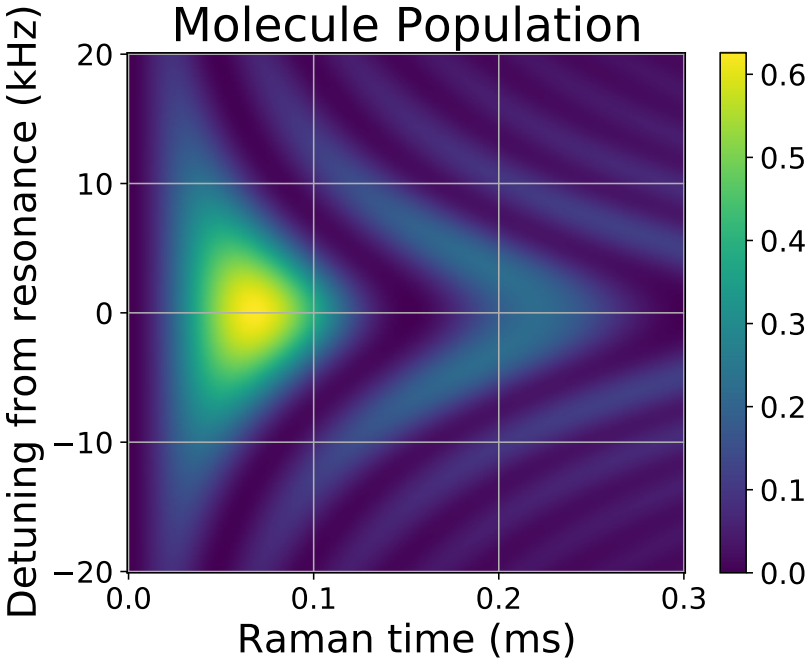

f_{Raman} = 770.59430(17)MHz
Γ_{FWHM} = 8.43(81)kHz
0.10 ms
Two-body survival
Detuning from resonance (kHz)
0.35
0.30
0.25
0.20
0.15
0.10
−20
0
20

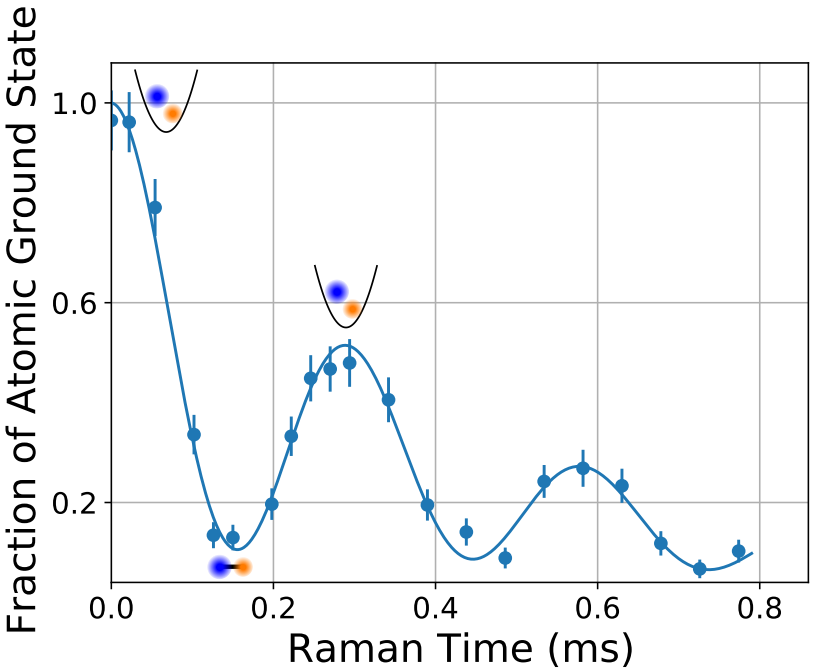

Fraction of Atomic Ground State
Raman Time (ms)
1.0
0.6
0.2
0.0
0.2
0.4
0.6
0.8

Experiment

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







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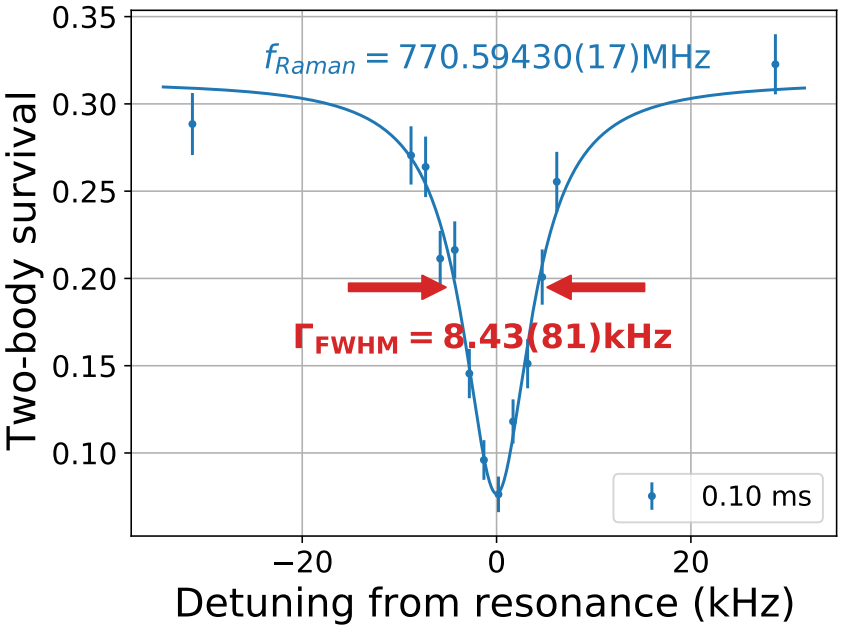

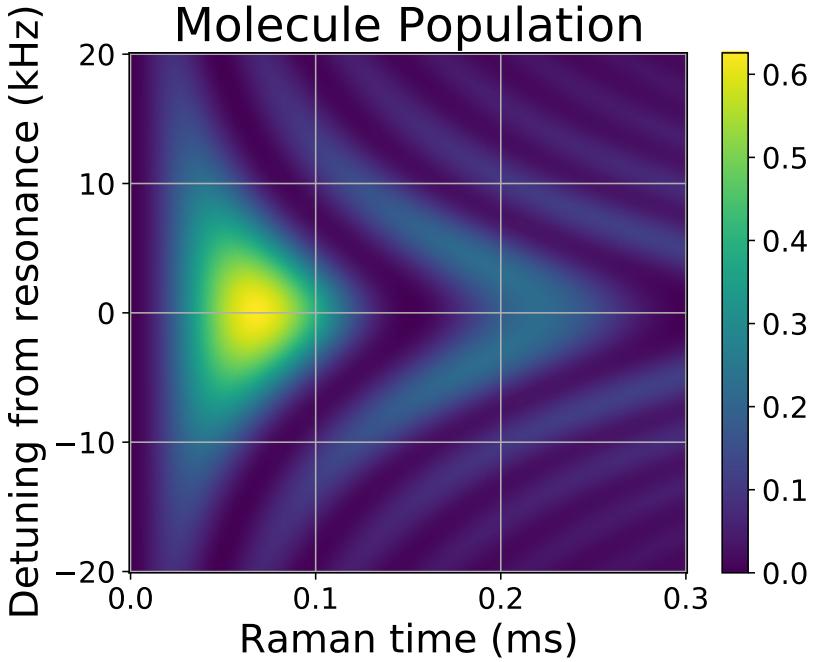

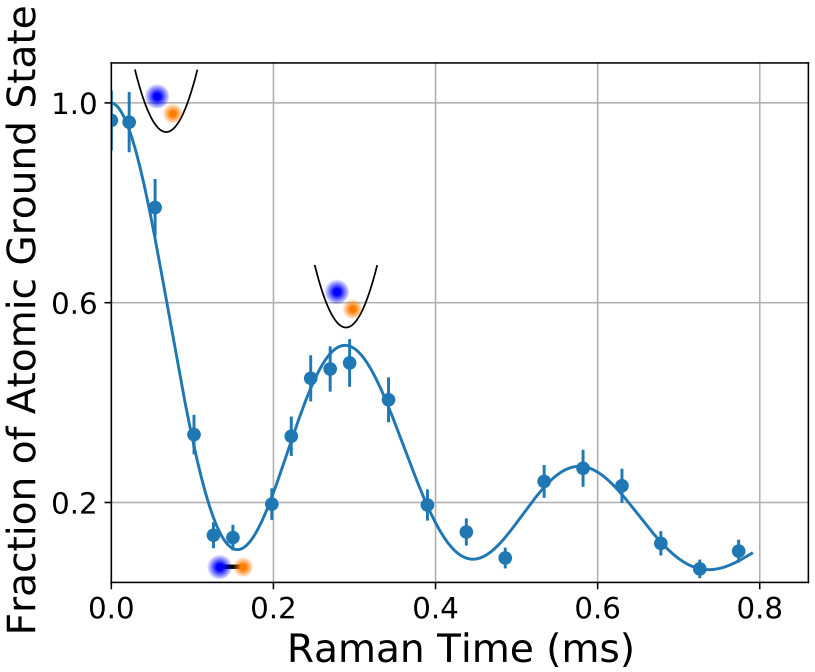

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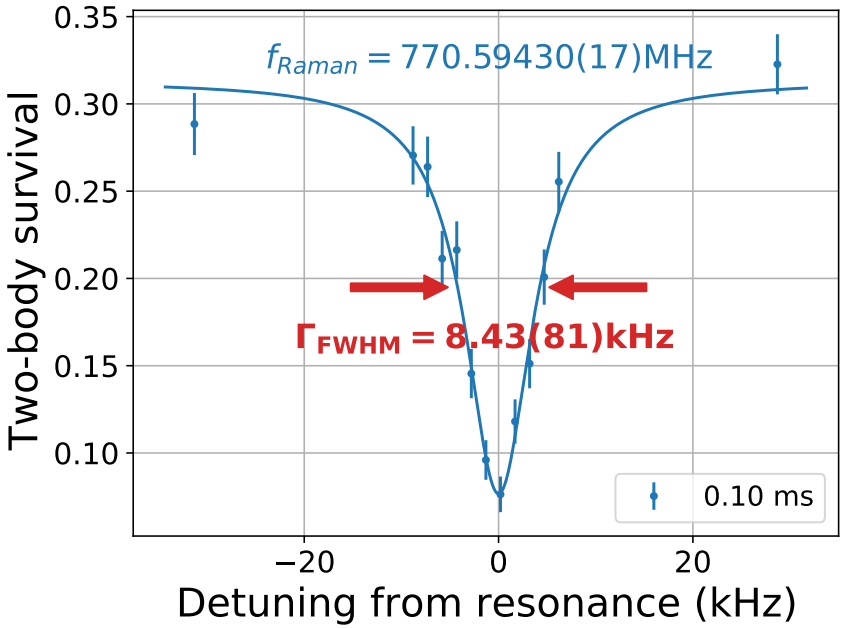
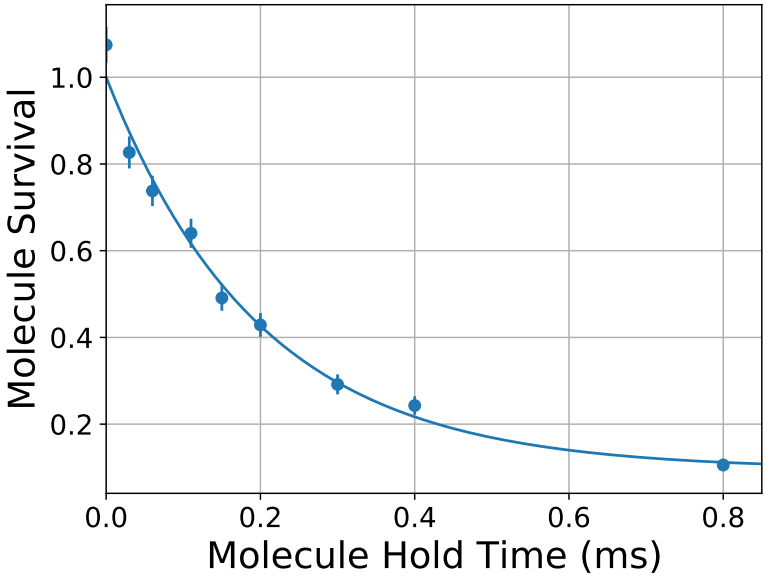
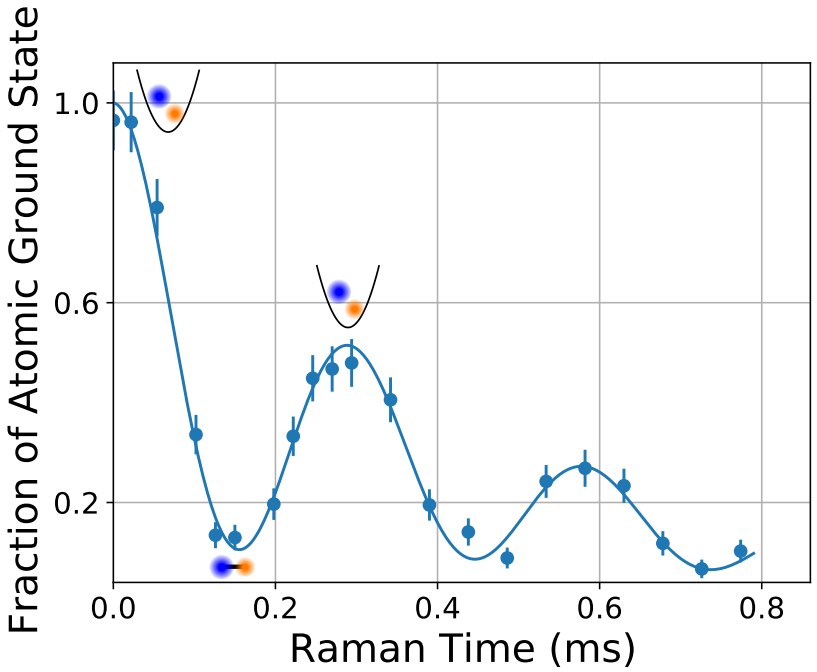
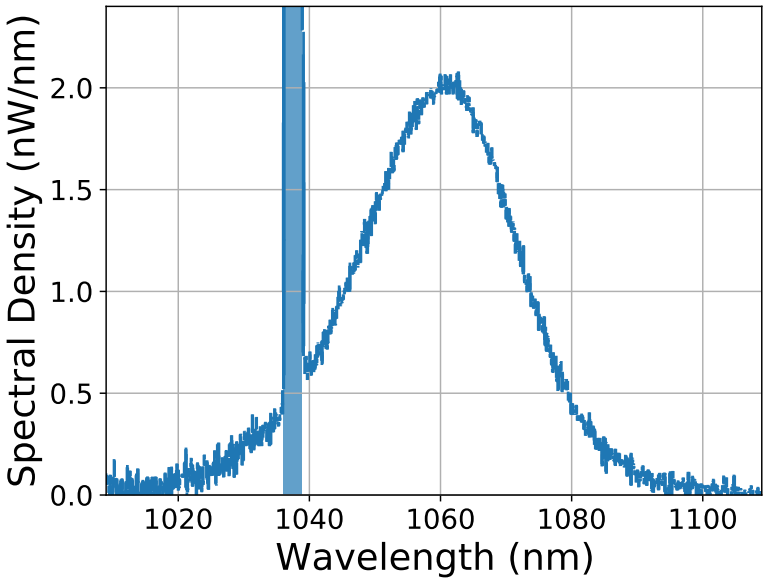






Amplified Spontaneous Emission (ASE)



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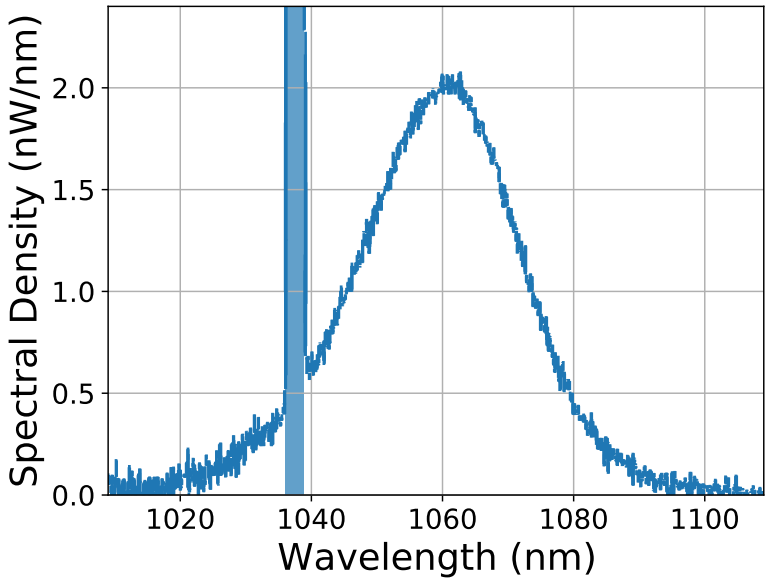

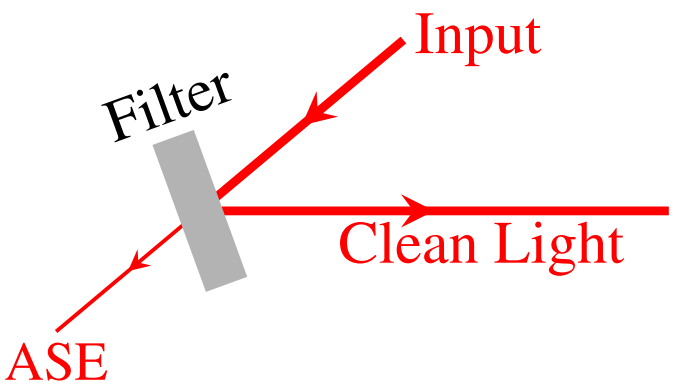

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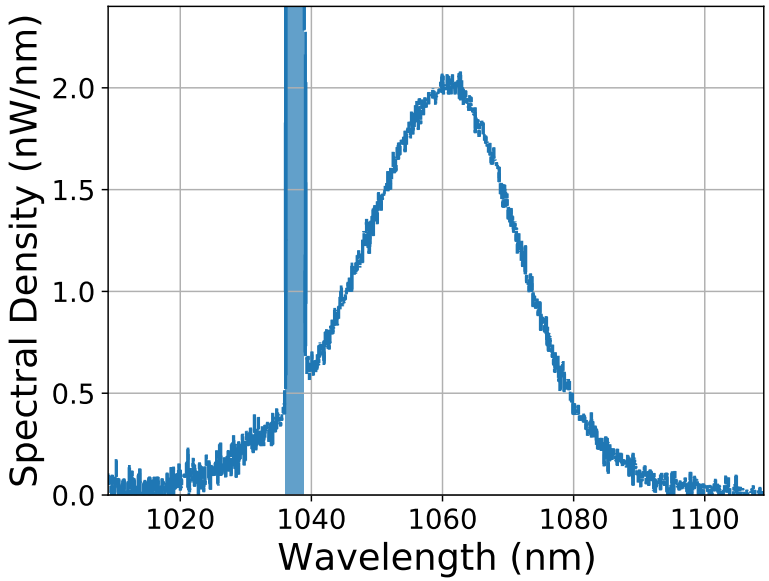

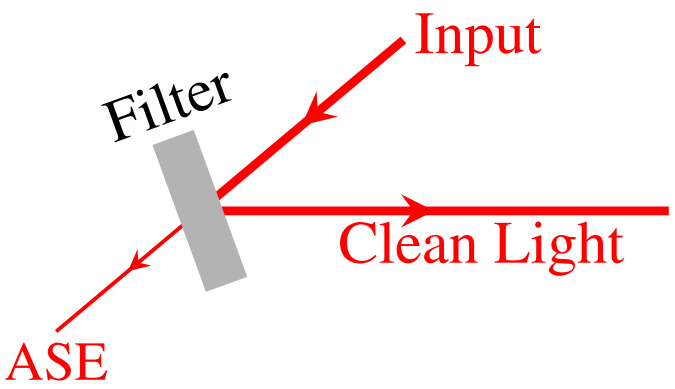

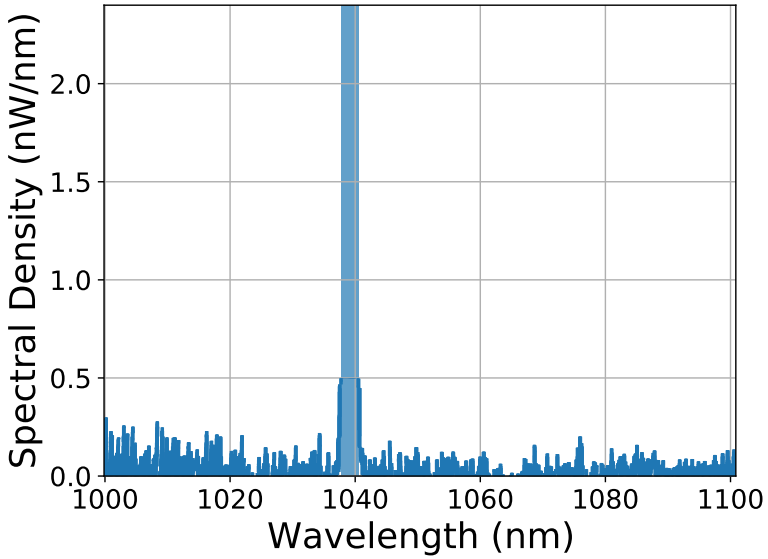

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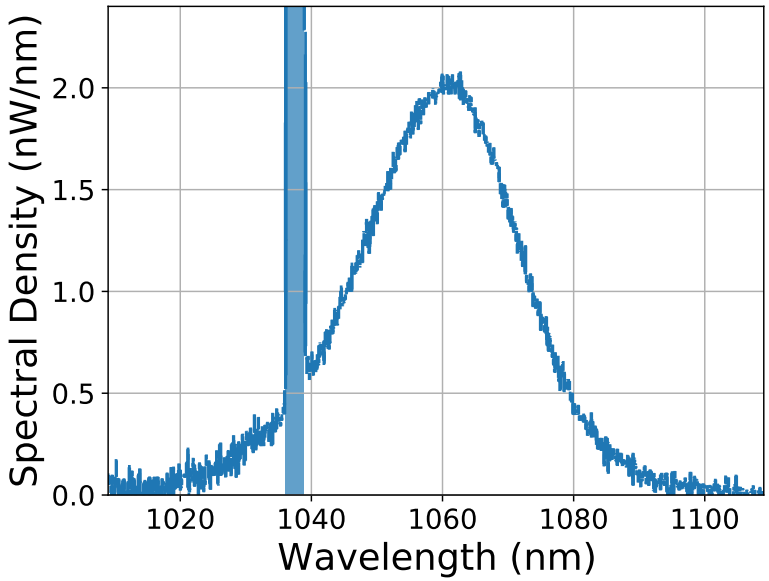

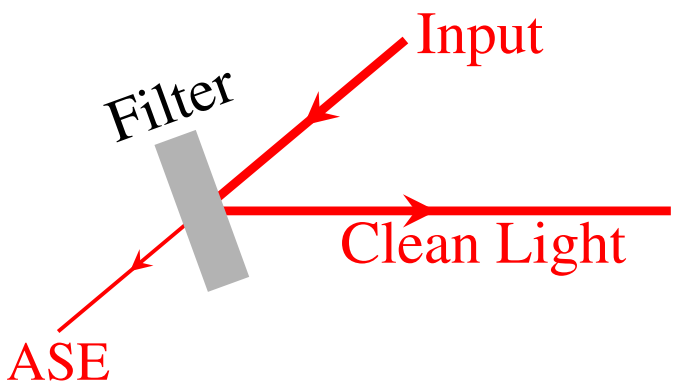

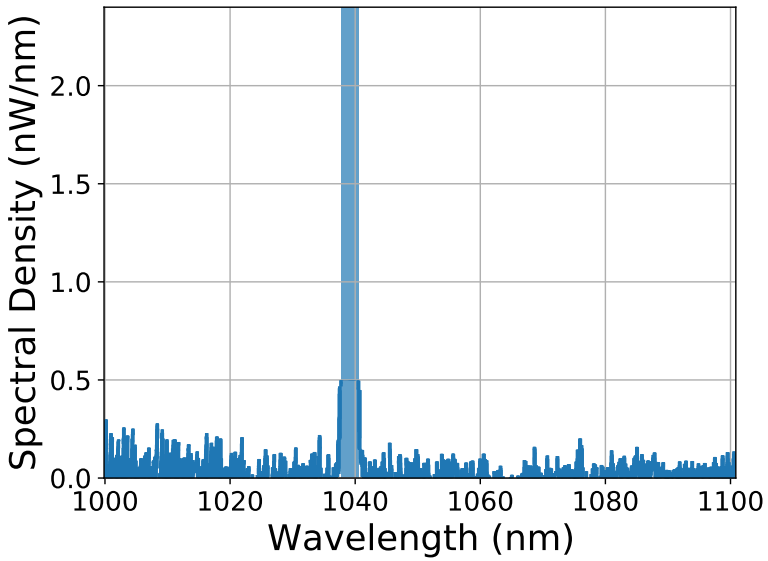

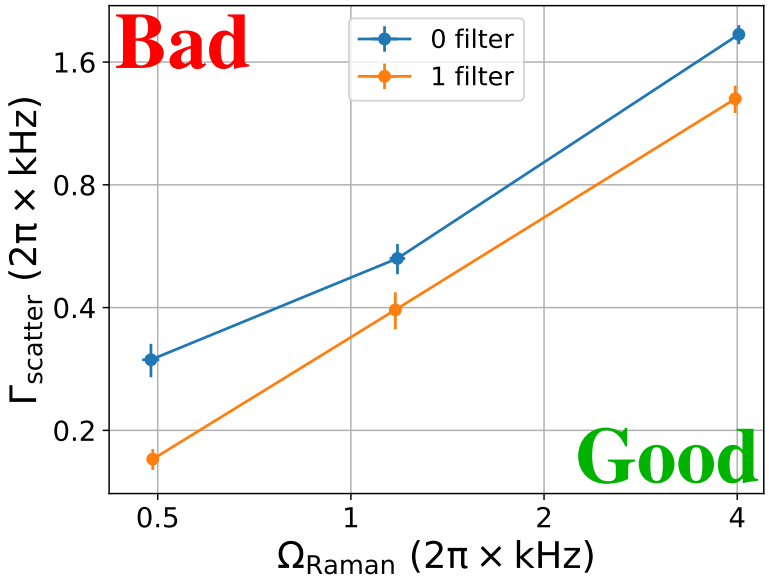

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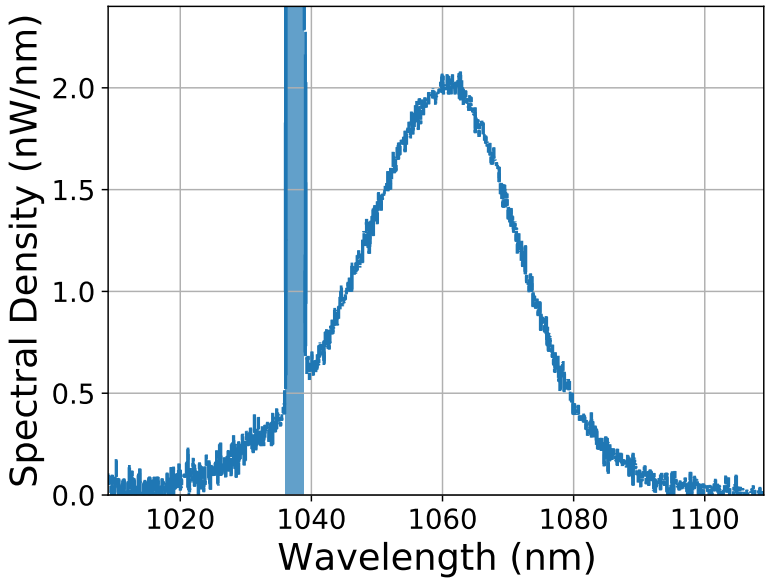

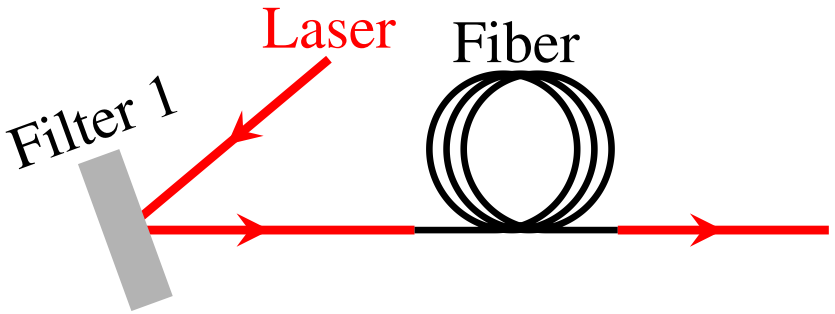

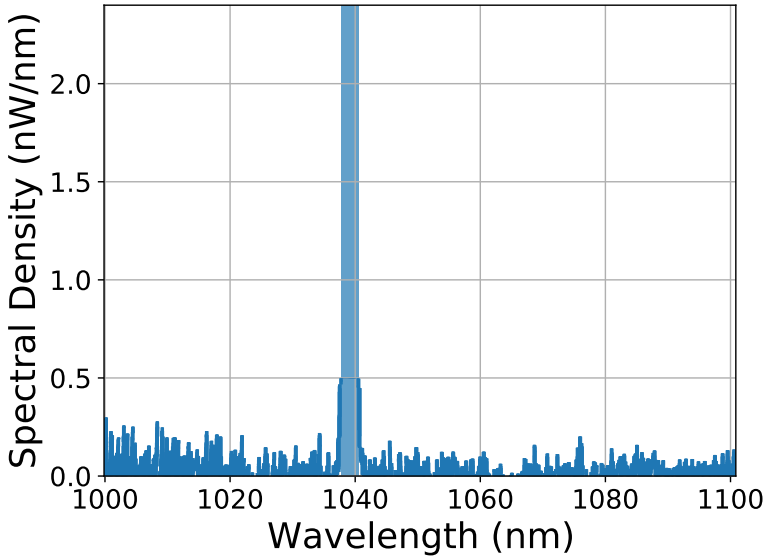

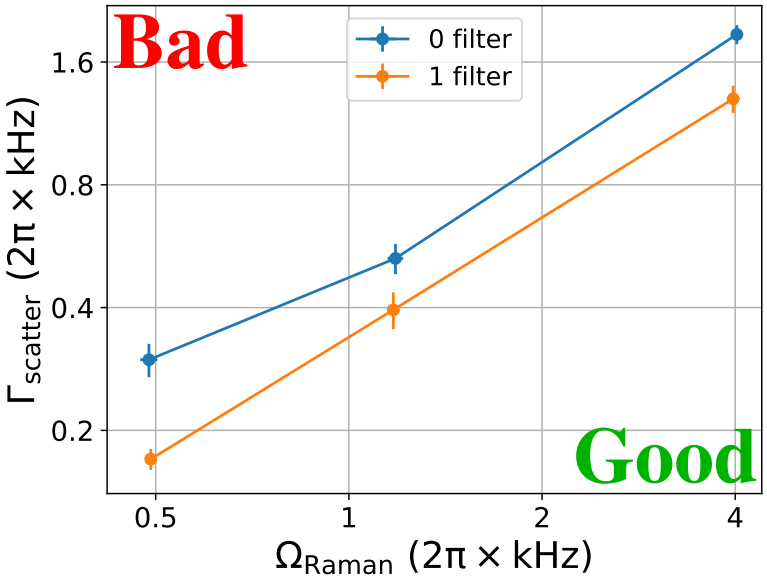

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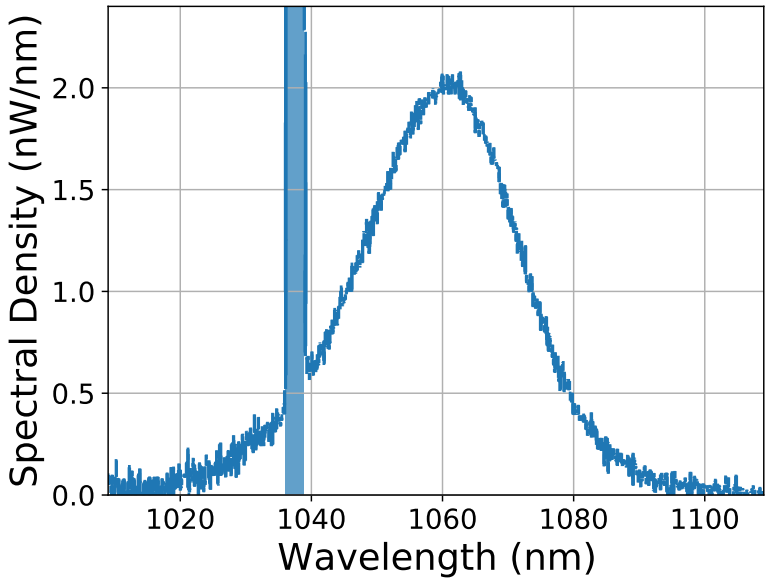
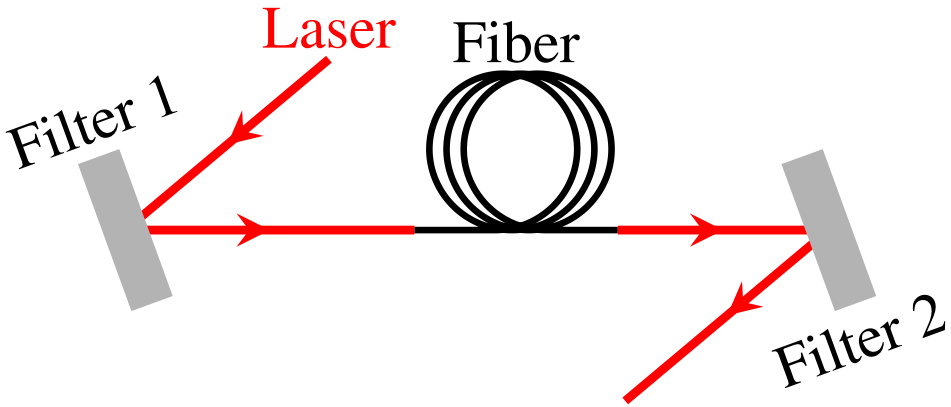
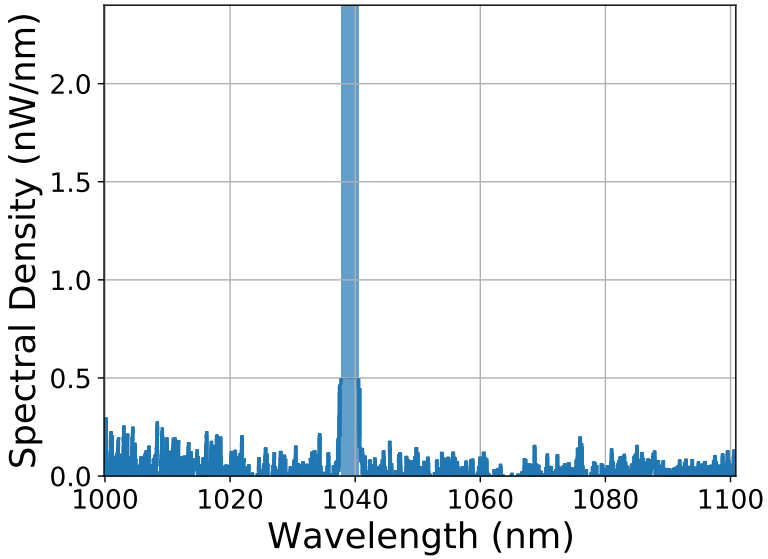
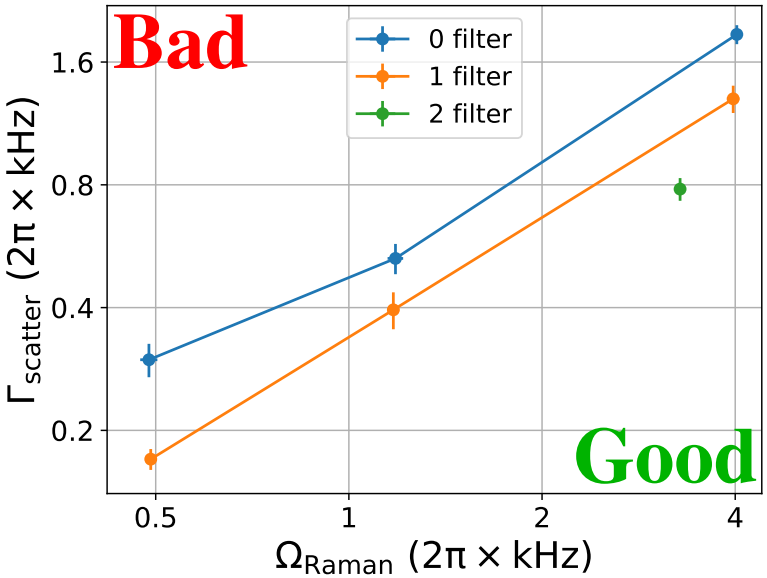
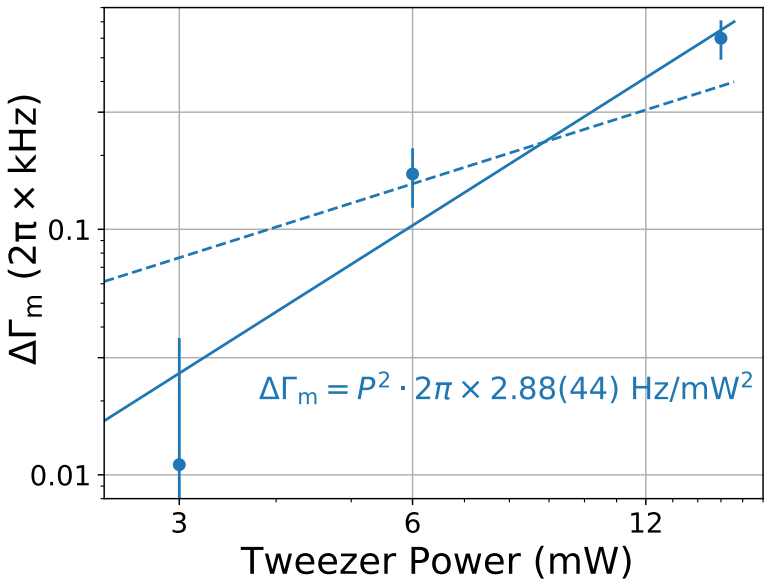








Two-Photon Scattering



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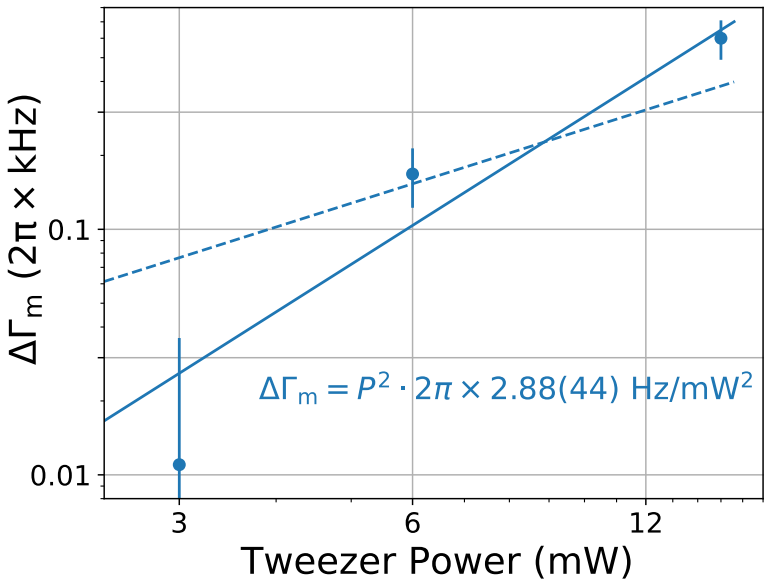

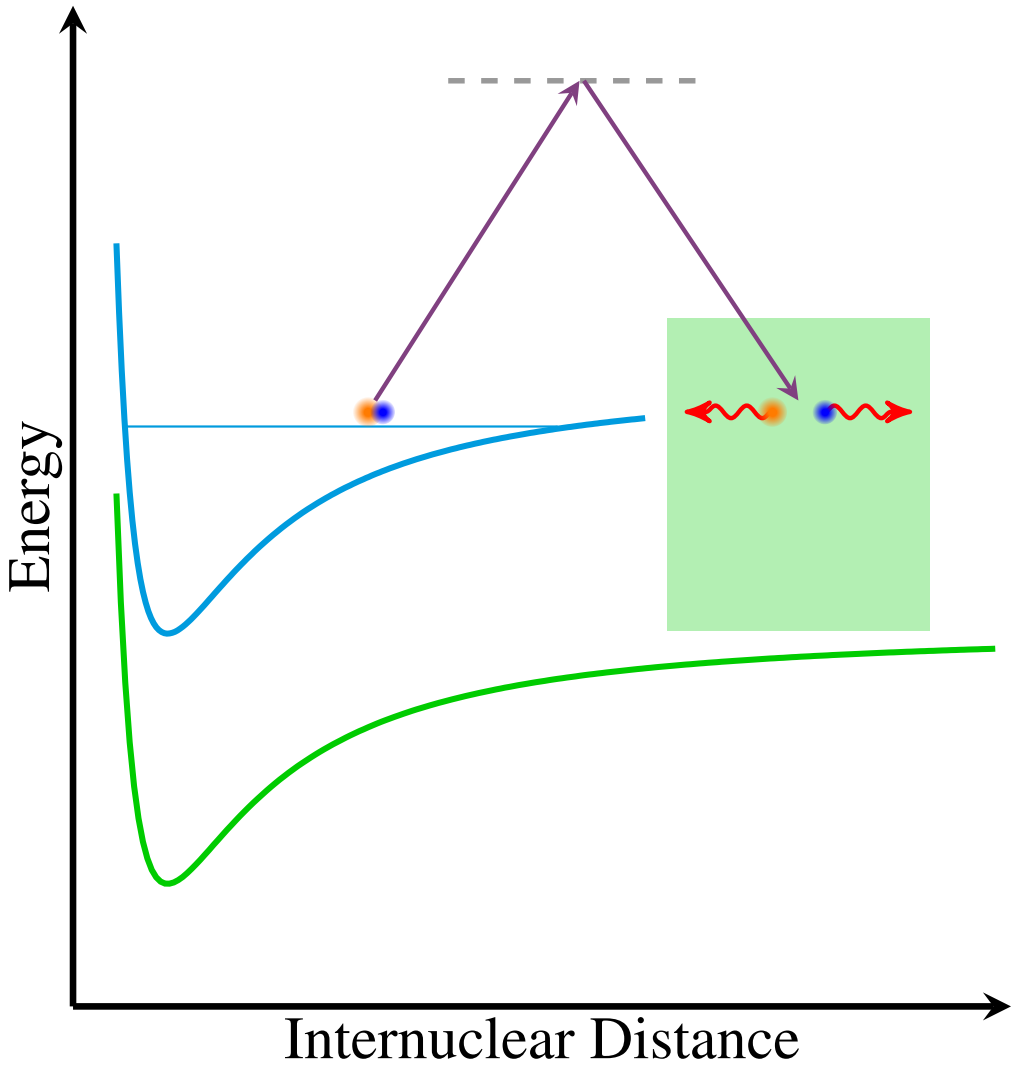

Conclusion and outlook

- 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
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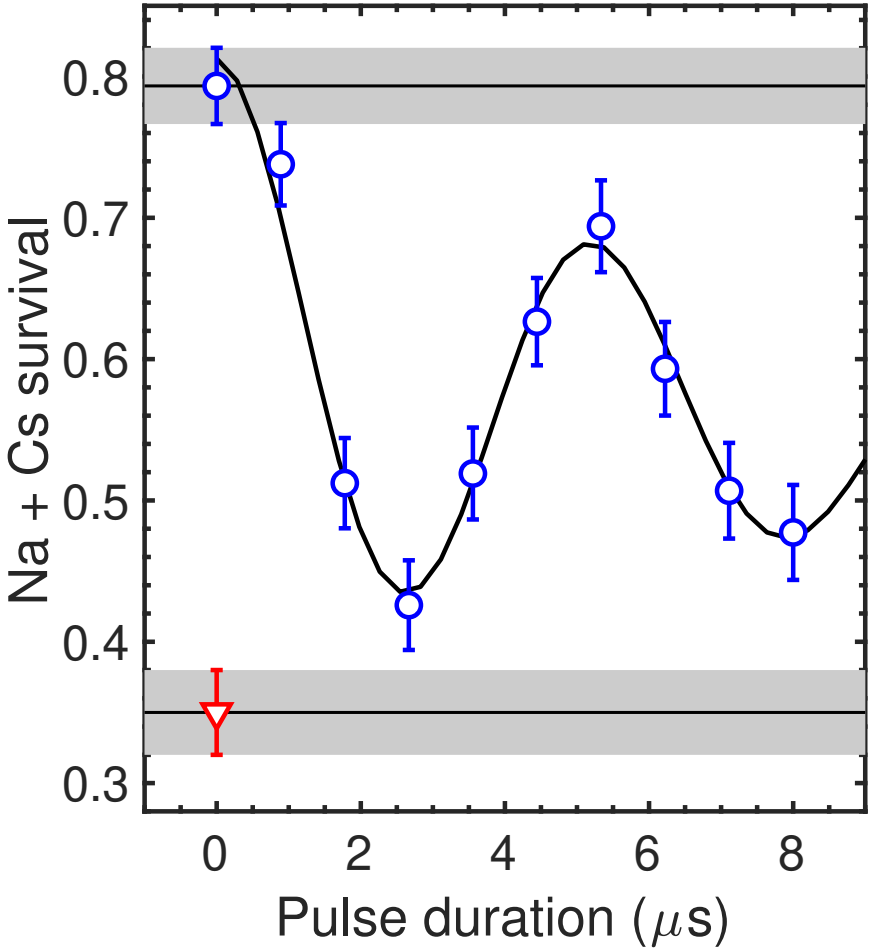
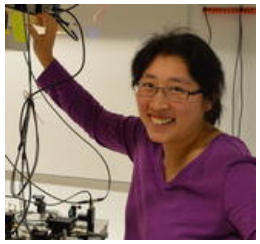


PI



Kang-Kuen Ni

NaCs Team

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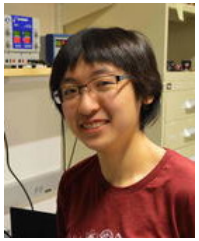
Yu Wang

Fang Fang



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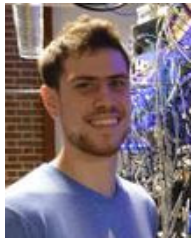

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William Cairncross

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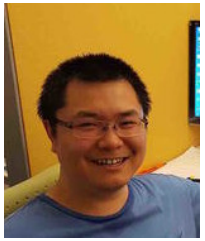

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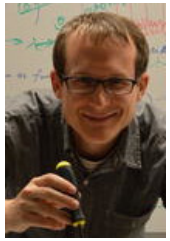
Postdoc @JILA



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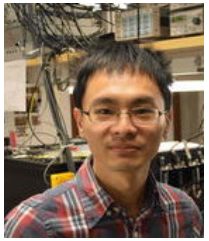

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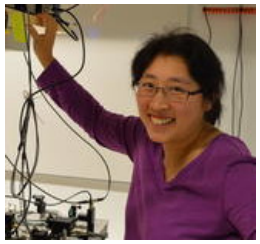

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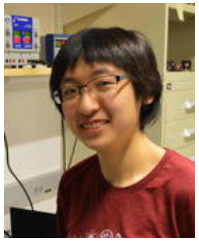

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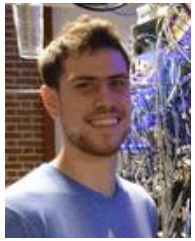

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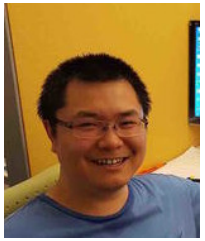

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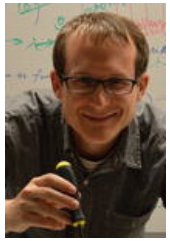

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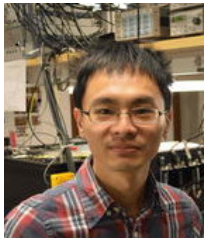

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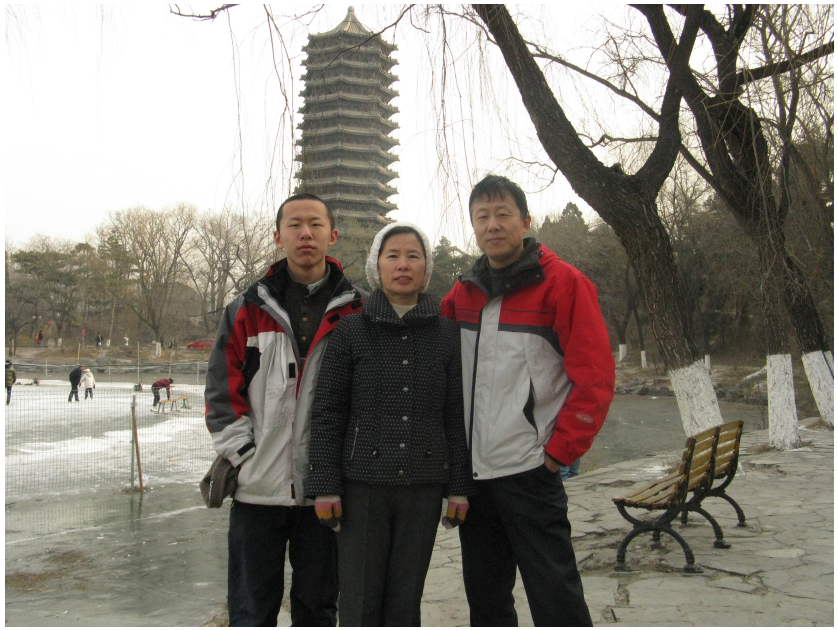
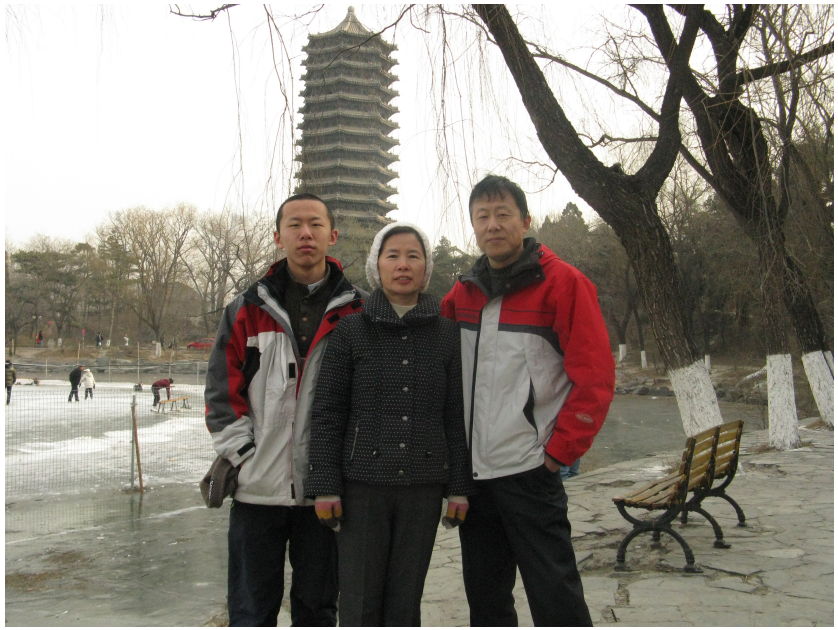
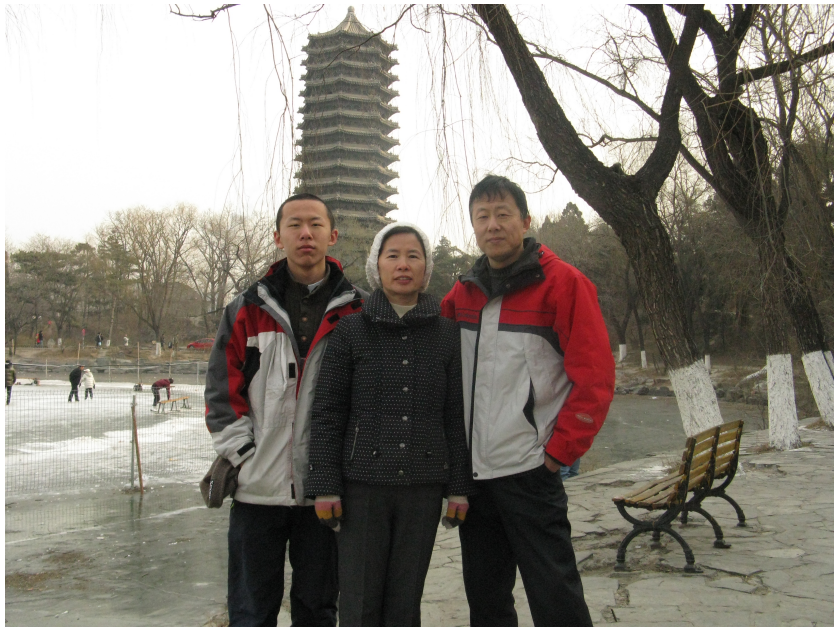
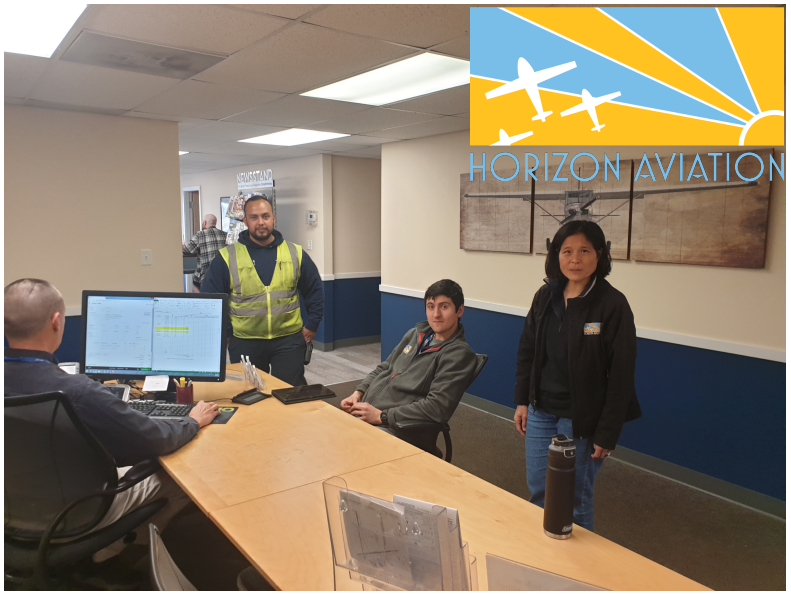


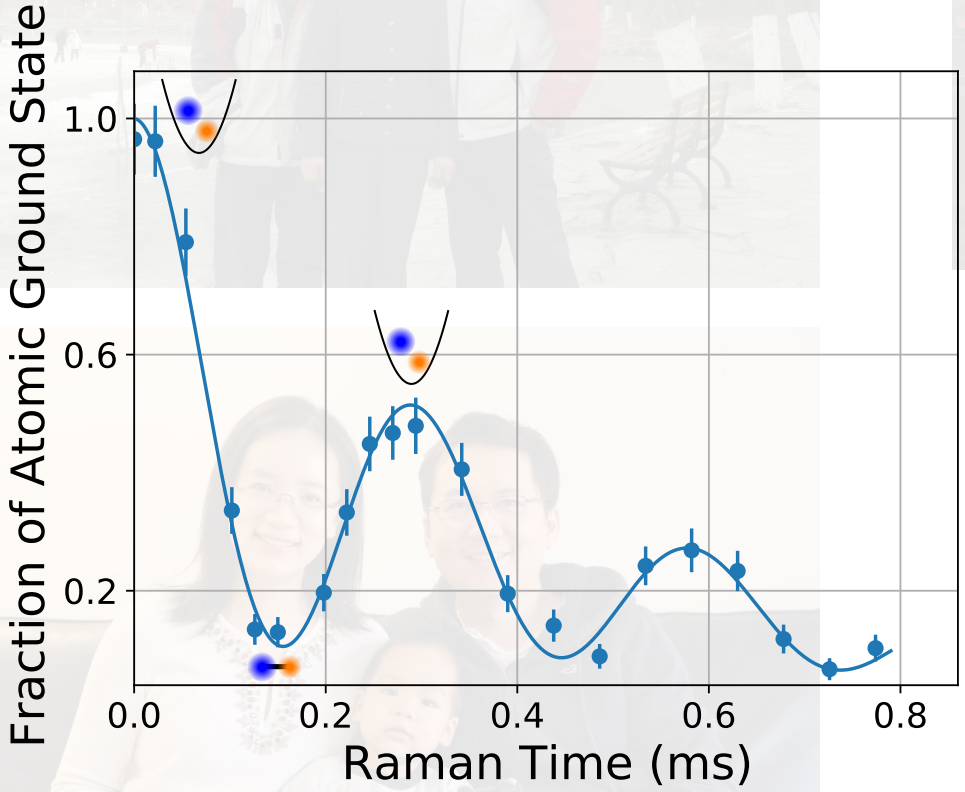


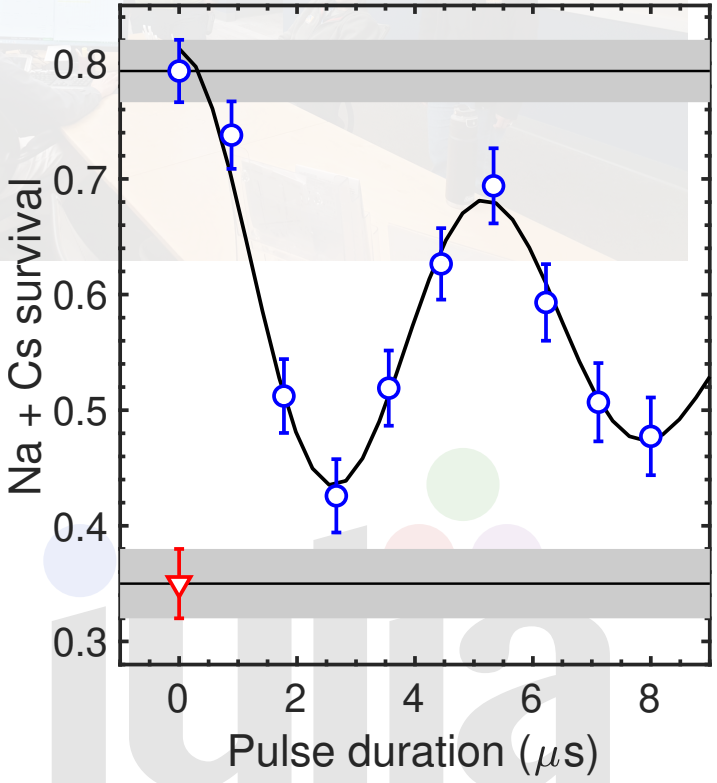




julia

Thanks for your attention





Single Atom in Tweezer

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- Works for Cs
- Doesn't work for Na

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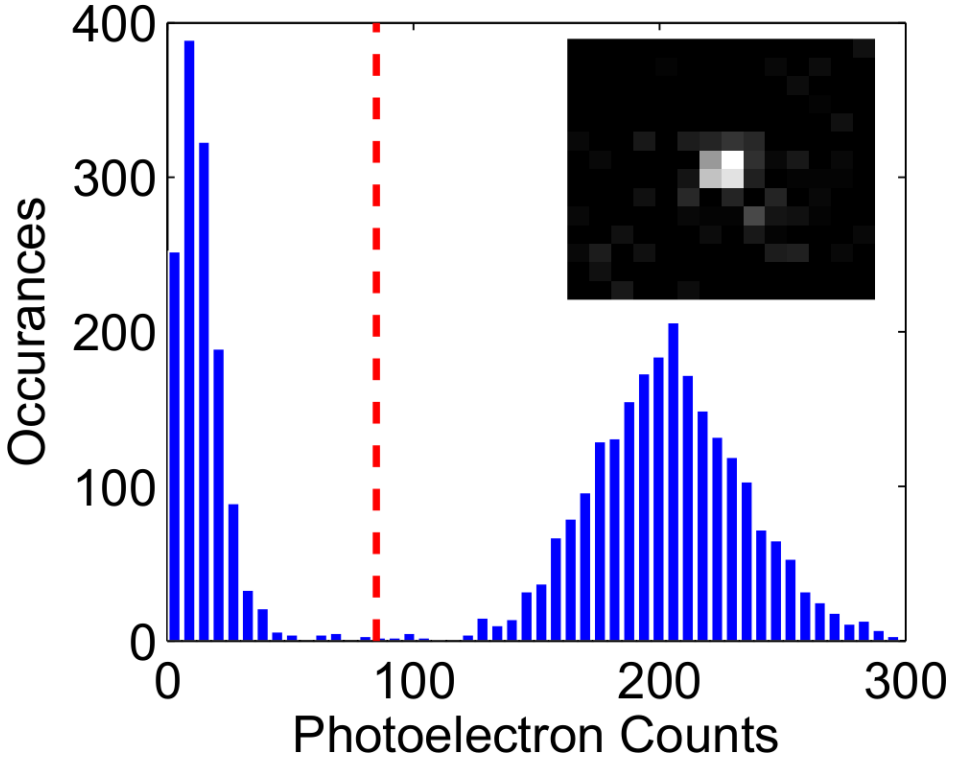

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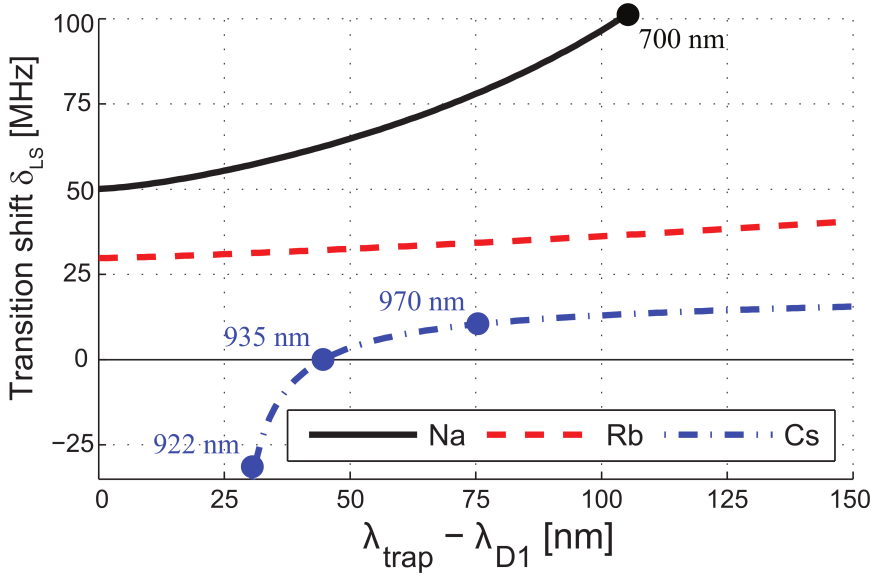
Issues with Na

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

Real Issue with Na: Light Shift



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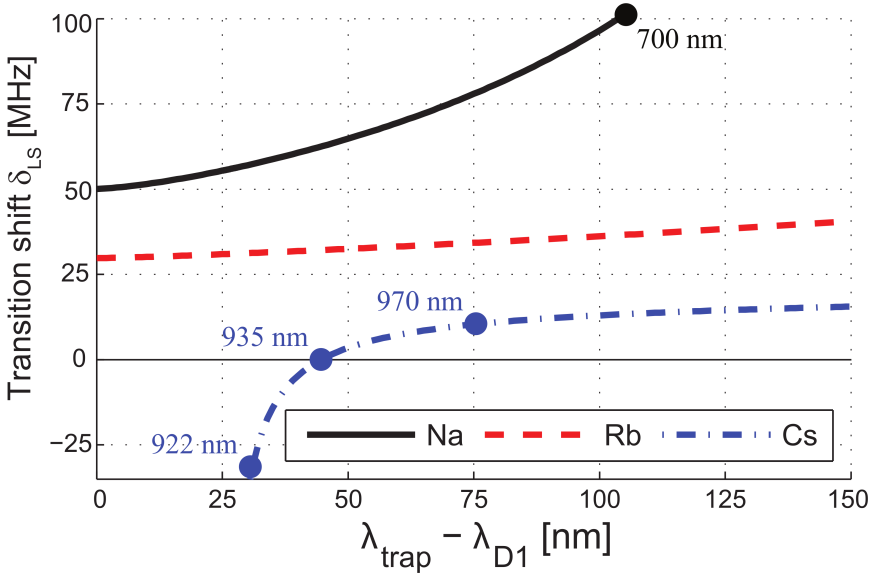
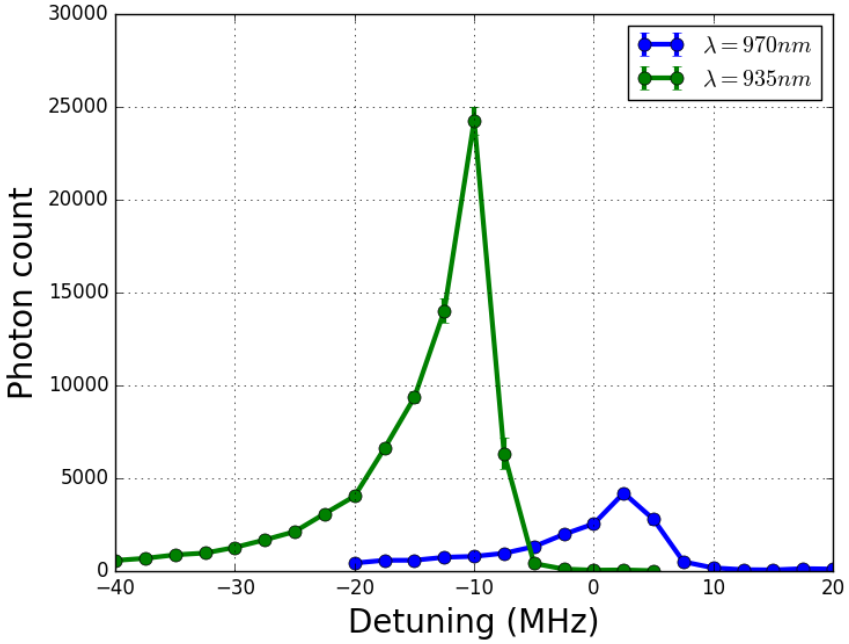


Cs single atom imaging



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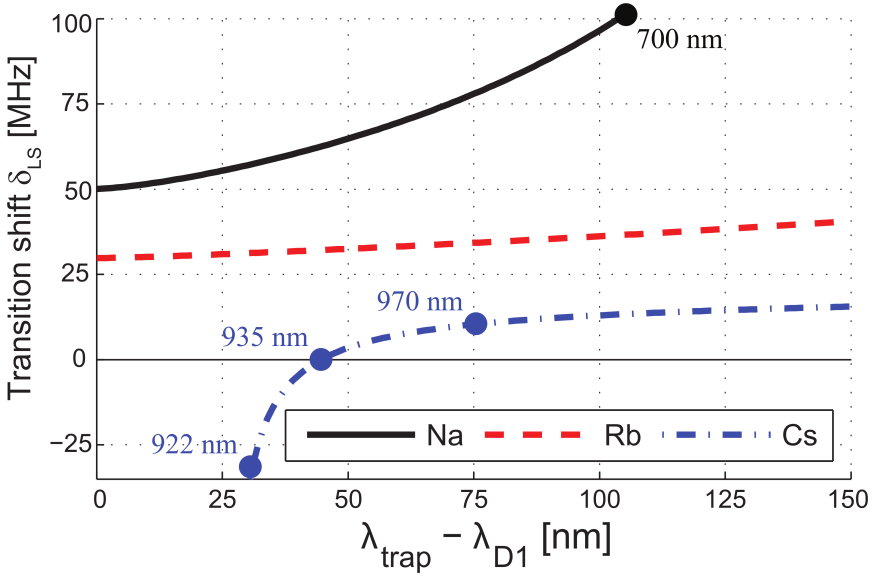

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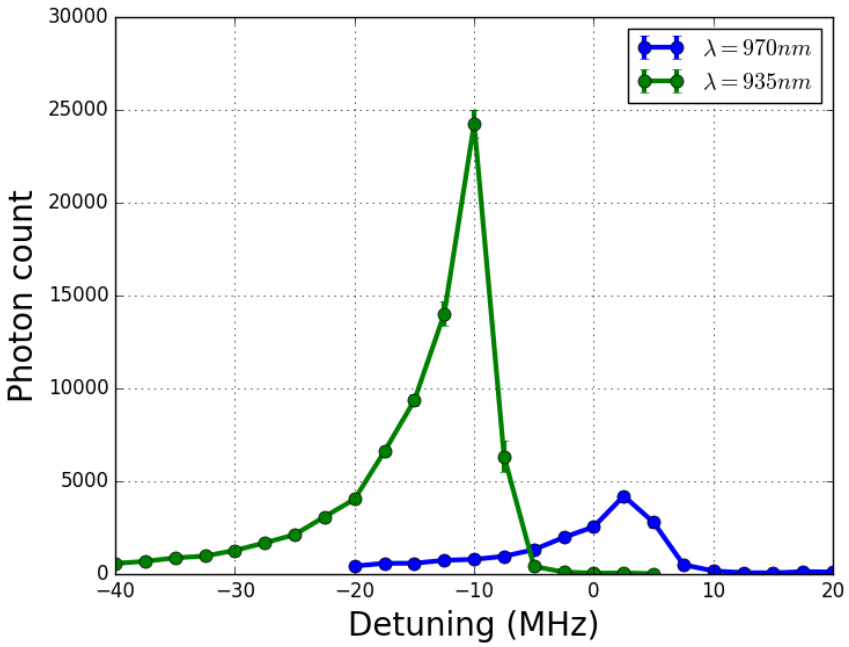


- Low imaging signal
- No cooling in tweezer

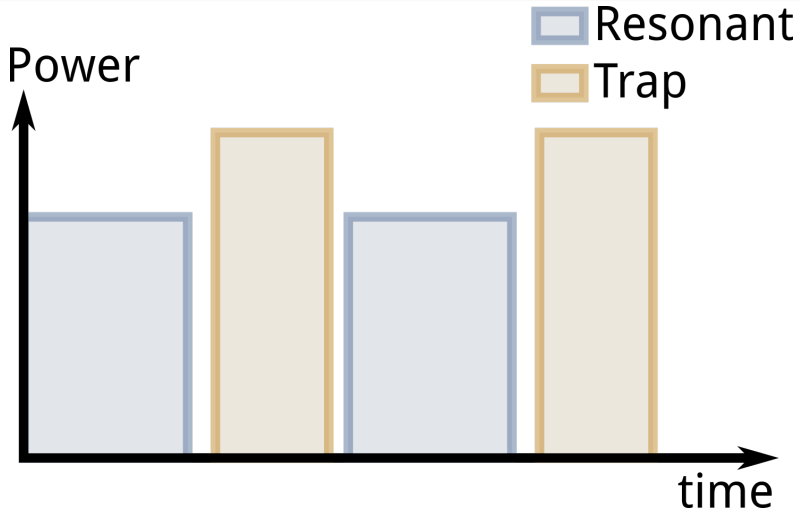
Real Issue with Na: Light Shift

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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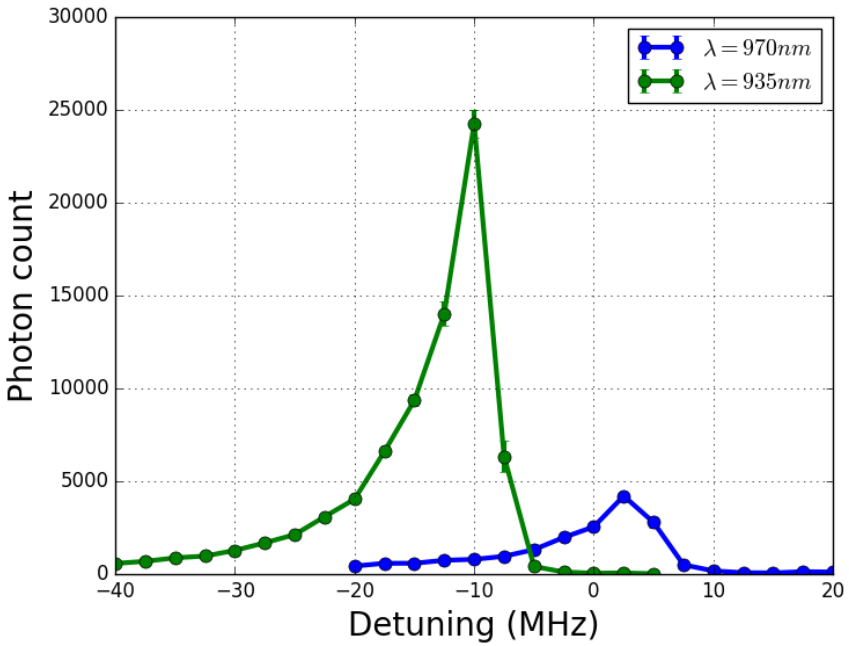

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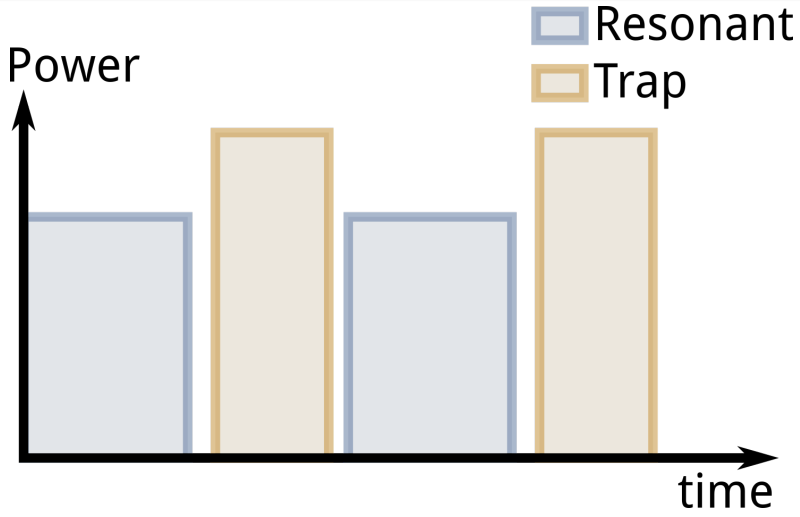

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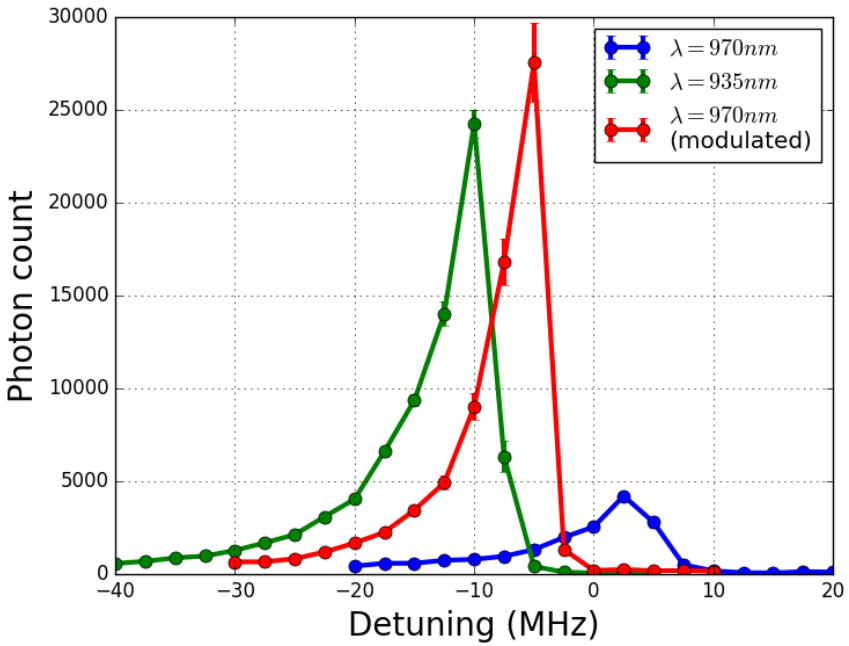

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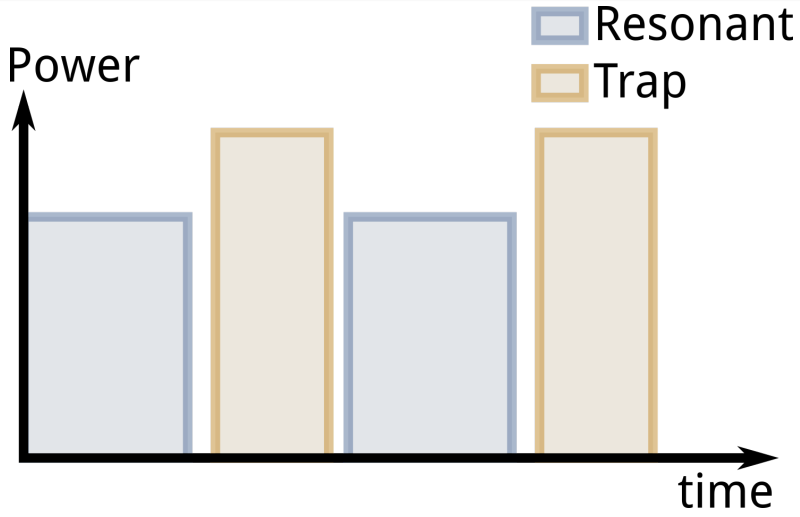

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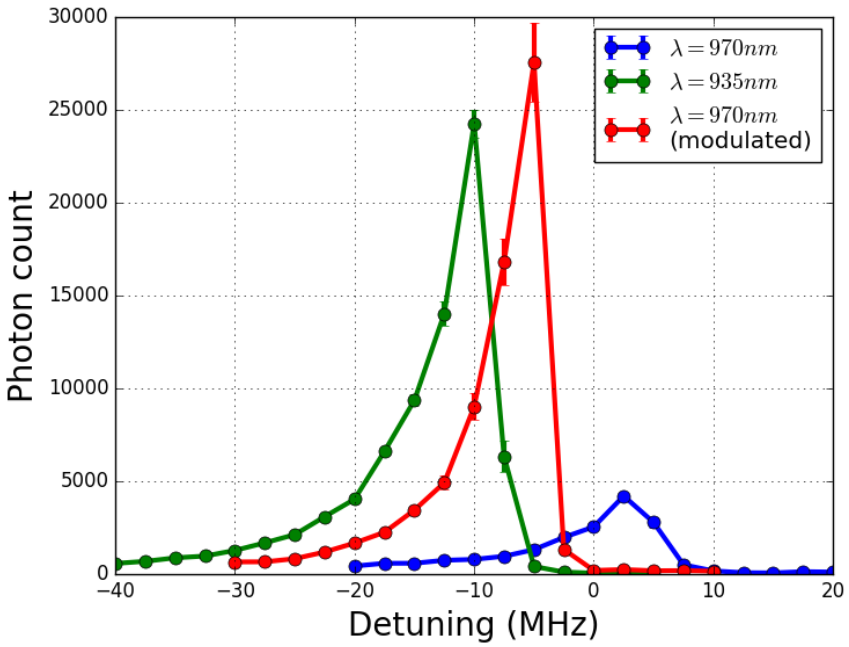
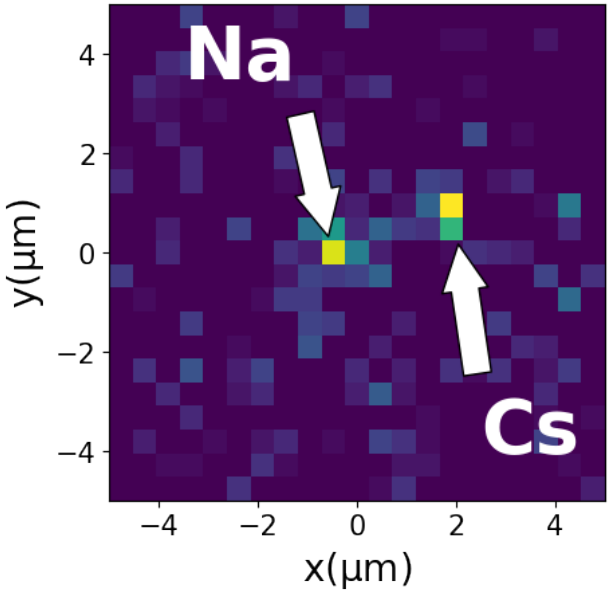
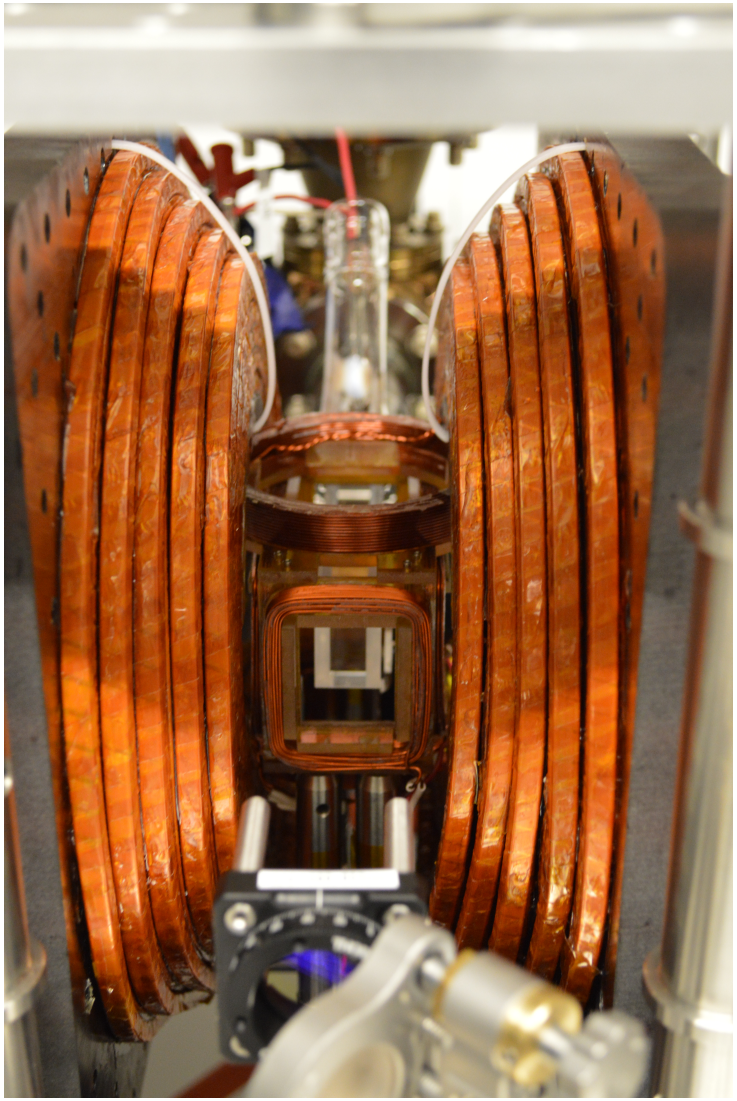




Na (1, -1) Cs (3, -3) Feshbach resonance



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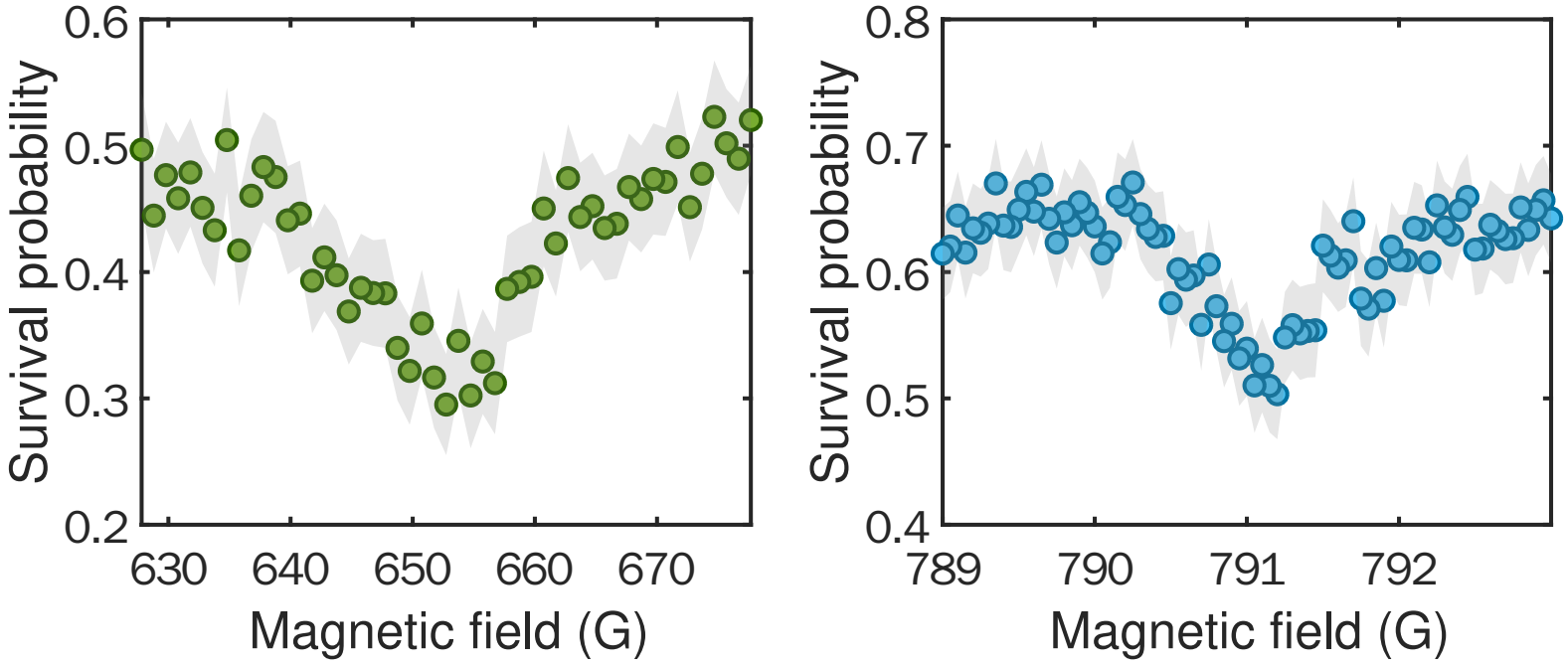

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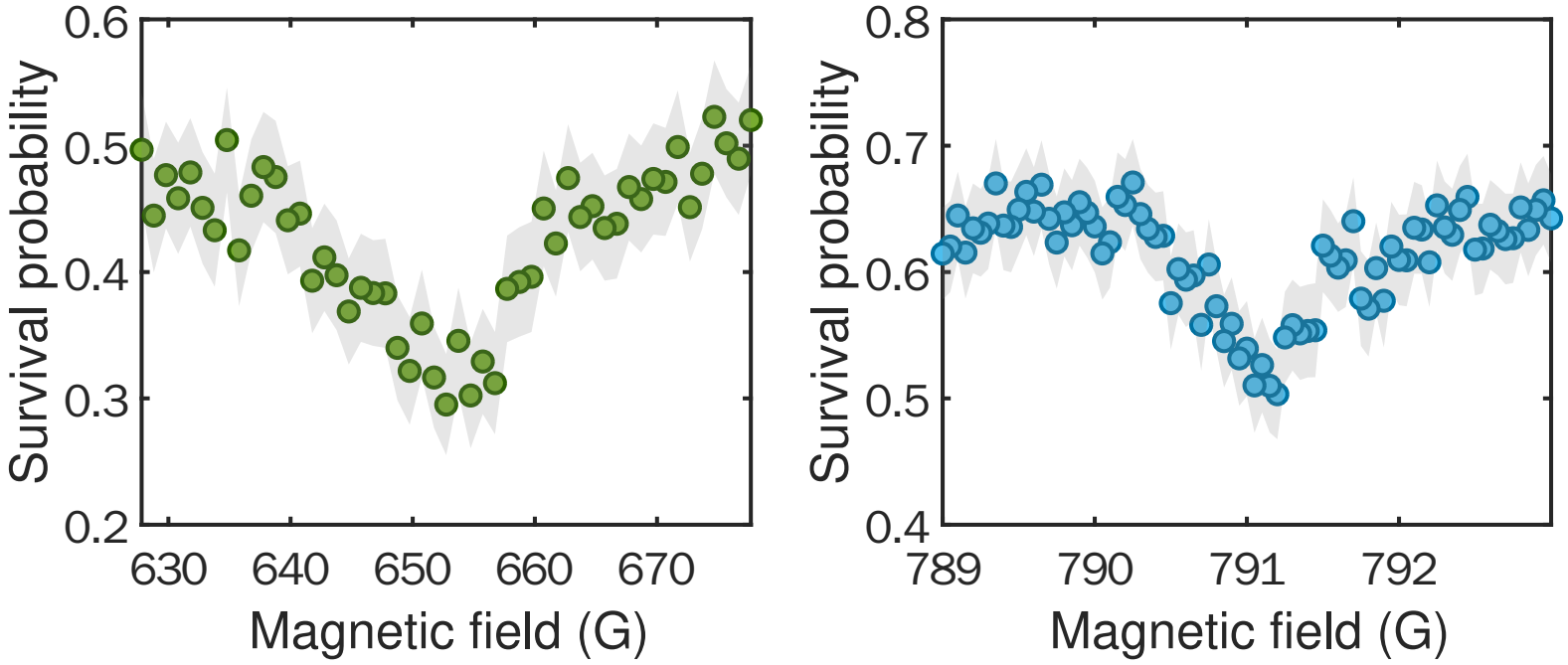
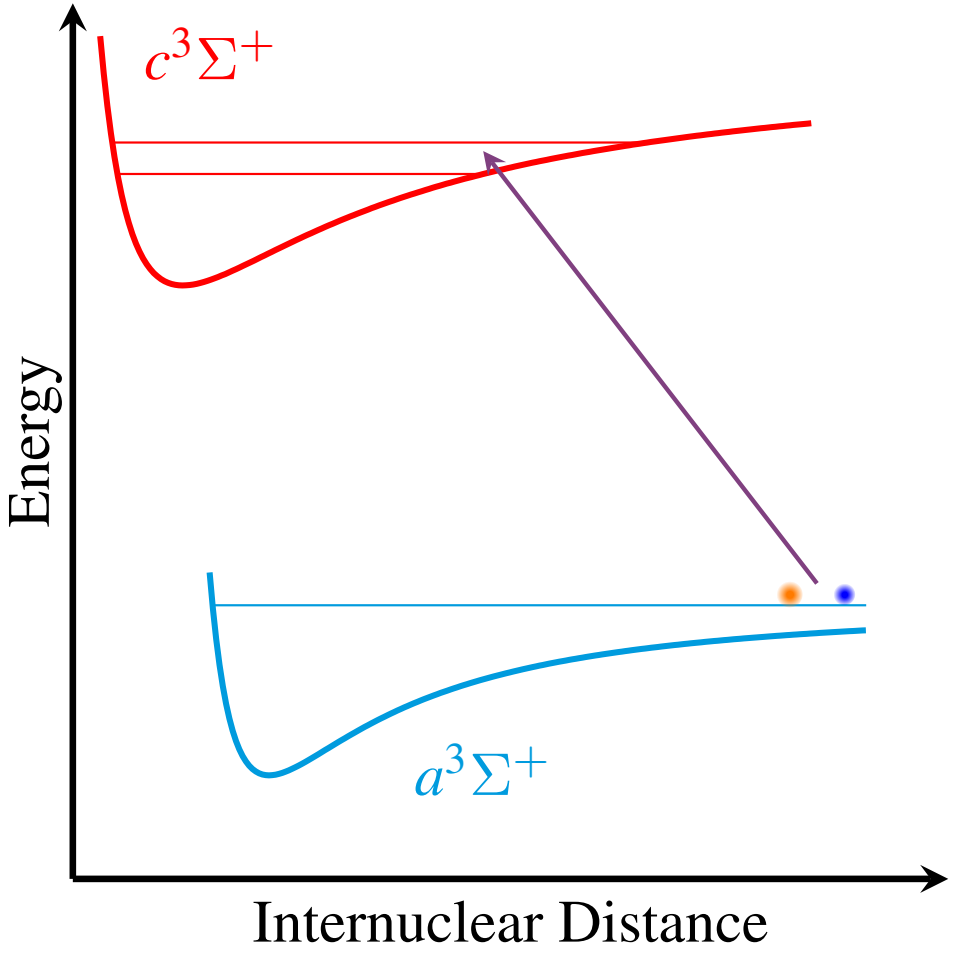


	s-wave	p-wave
Predicted (based on interaction shift)[1]	663 G	799 G
Measured	652(3) G	791.2(2) G

[1] In collaboration with Bo Gao

Photoassociation (PA) Spectroscopy



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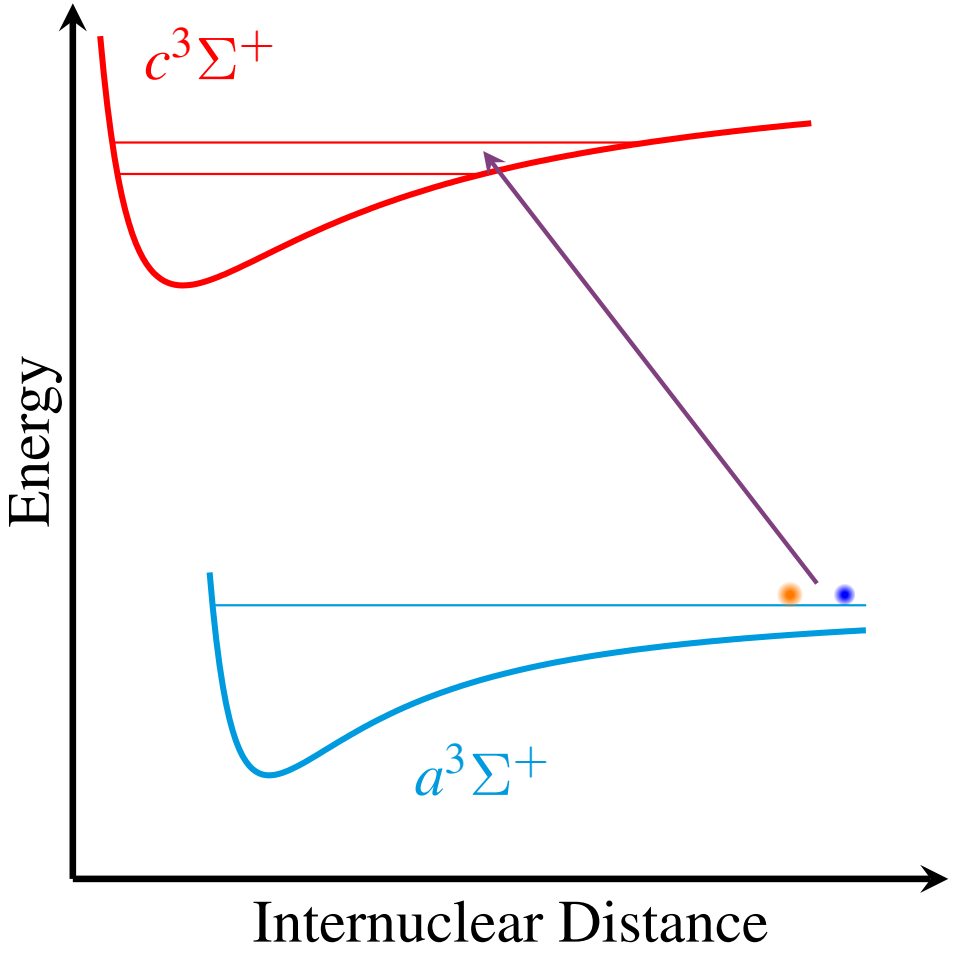

Single Atom PA

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

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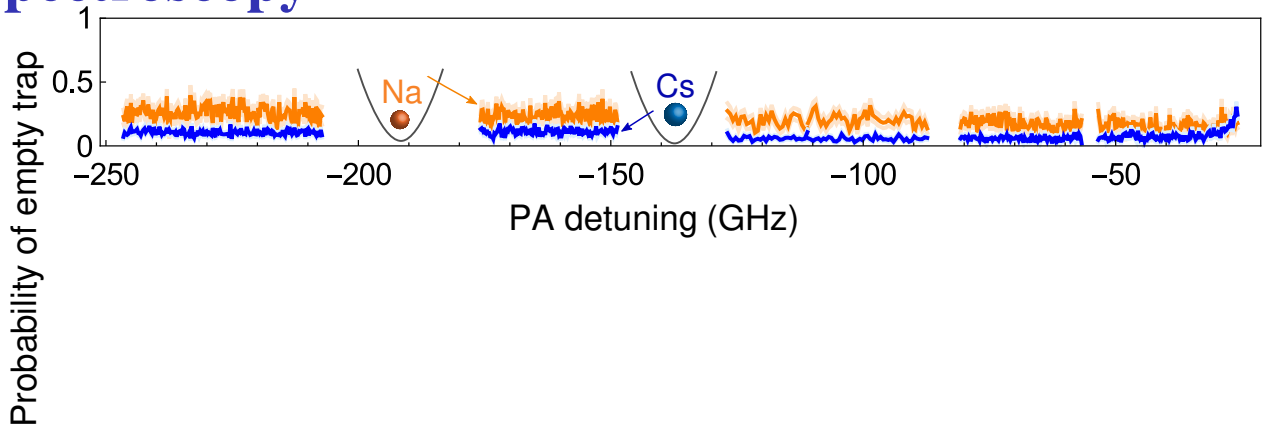

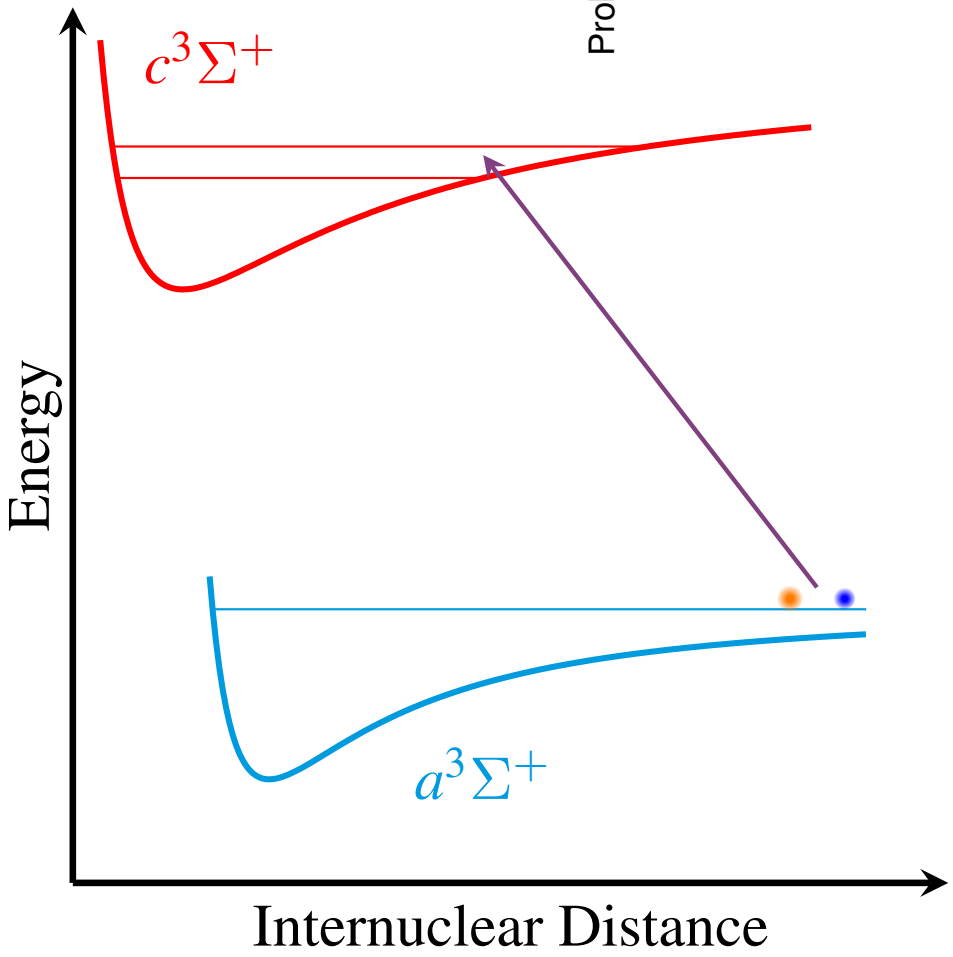

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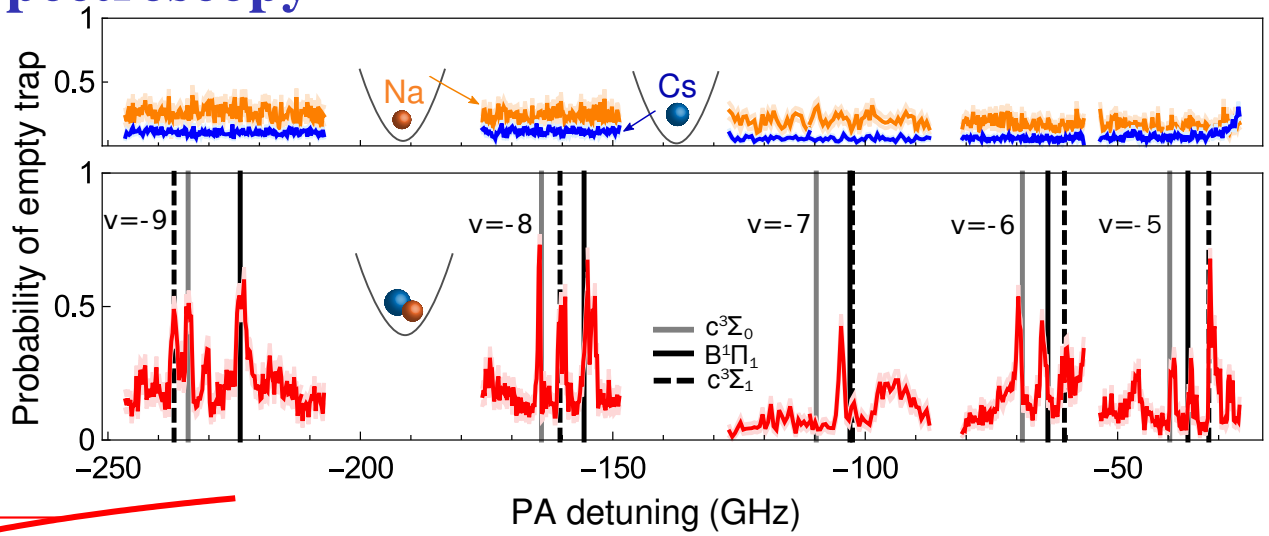

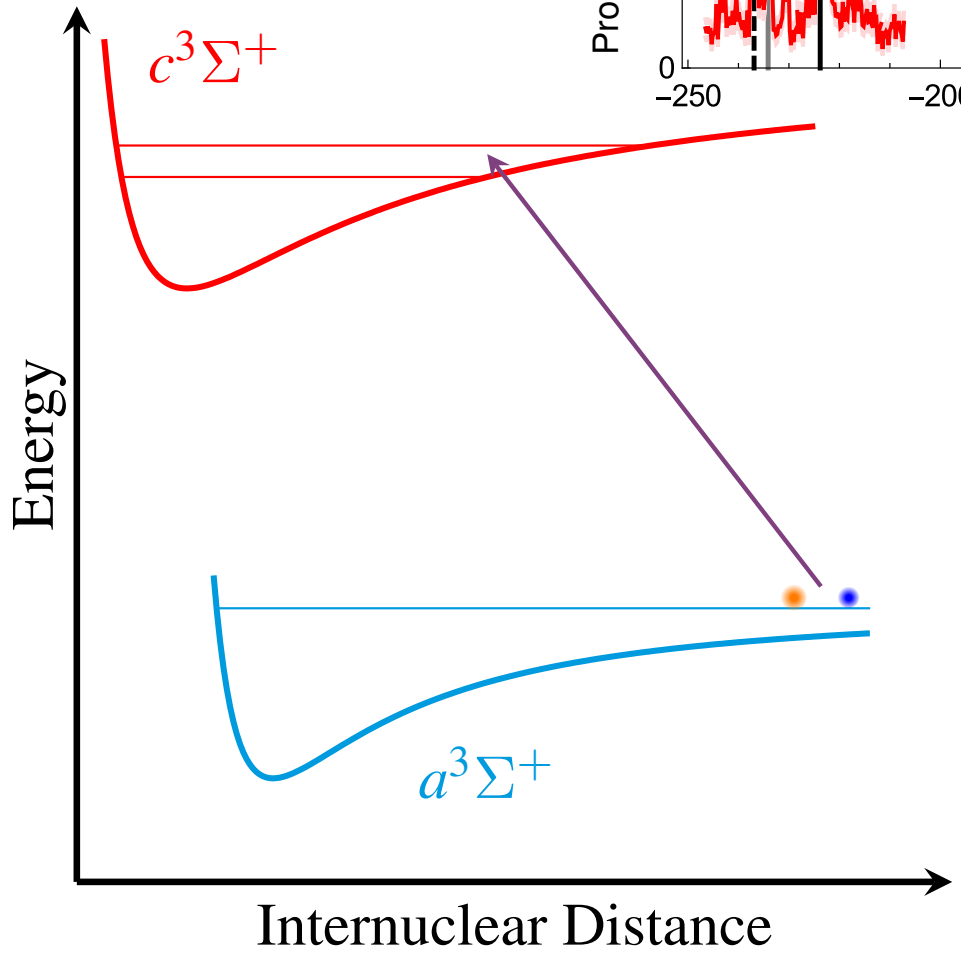

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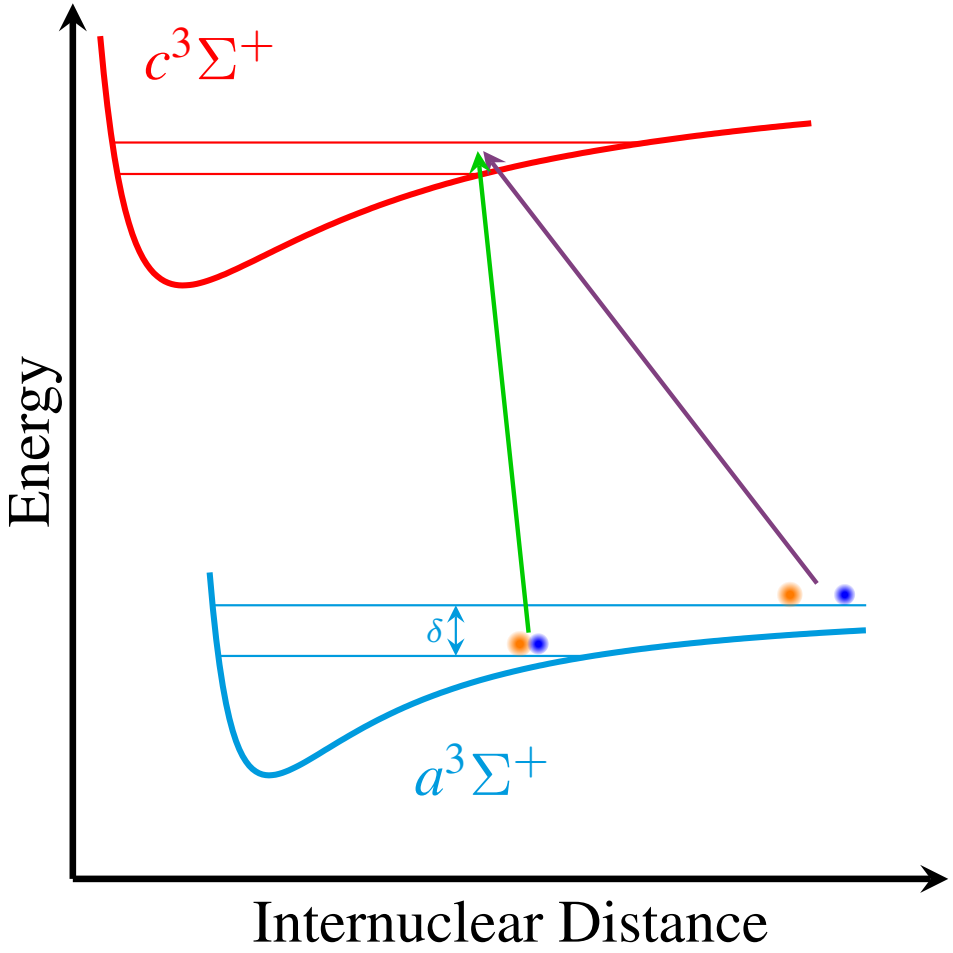
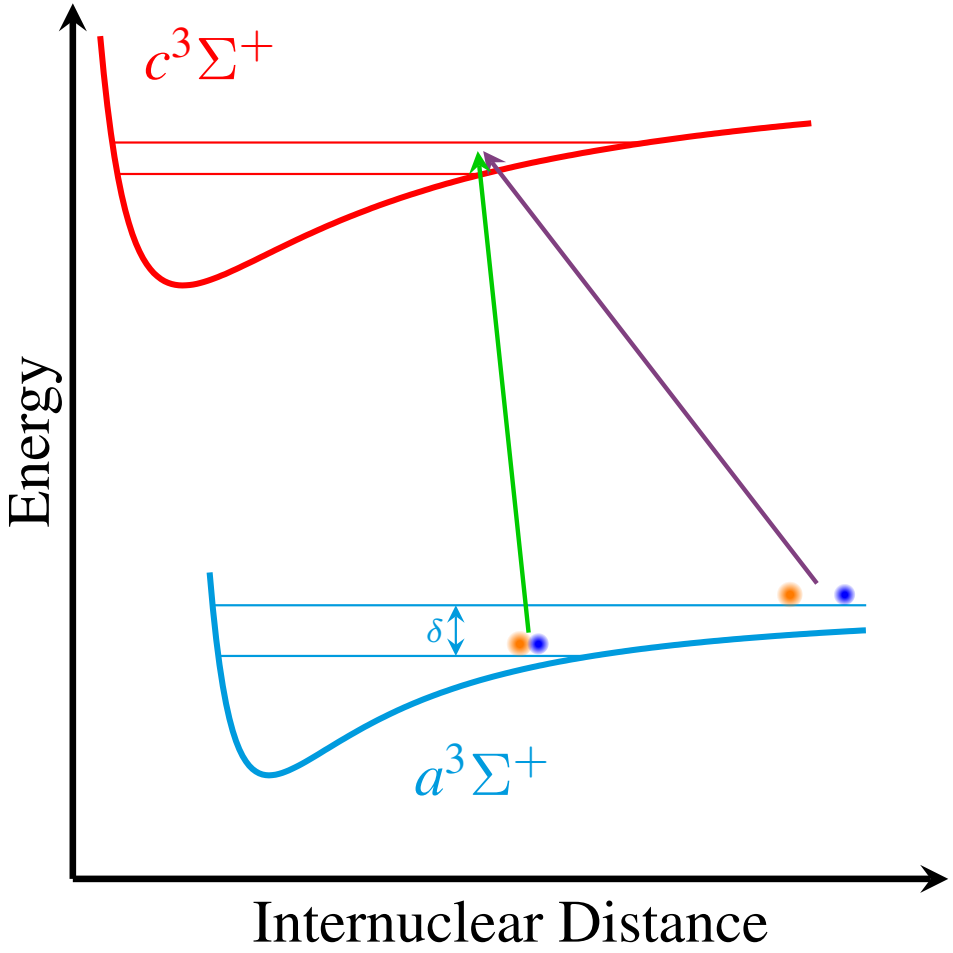
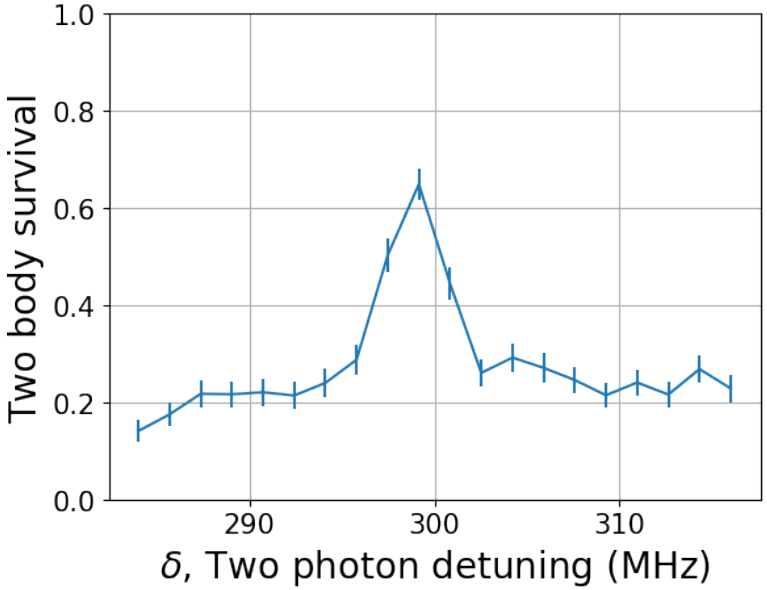
Electromagnetically Induced Transparency (EIT) Spectroscopy



Electromagnetically Induced Transparency (EIT) Spectroscopy