

# Building Single Molecules from Single Atoms

Yichao Yu

Lee Liu, Kenneth Wang, Lewis Picard, Jonathan Hood  
Jessie T. Zhang, Eliot Fenton, Yen-Wei Lin

Ni Group/Harvard

Sep 5, 2019

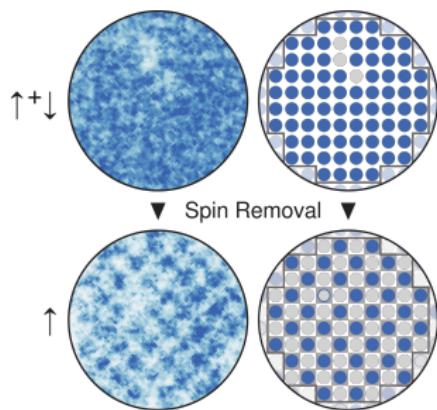
# Atom

- Laser cooling/trapping
  - Internal state control
  - High fidelity imagining
- ⋮

# From Atom to Molecule

## Atom

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- Internal state control
- High fidelity imagining
- ⋮



Nature 545, 462-466 (2017)

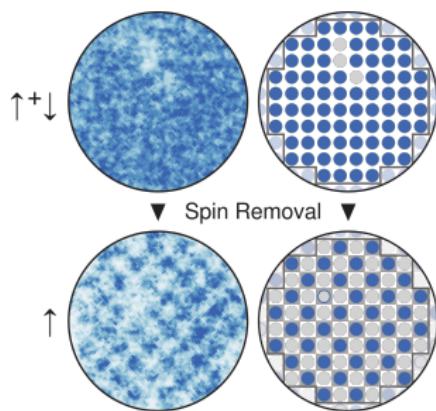
# From Atom to Molecule

## Atom

- Laser cooling/trapping
- Internal state control
- High fidelity imagining
- ⋮

## Molecule

- Strong interaction
- Rich internal structure



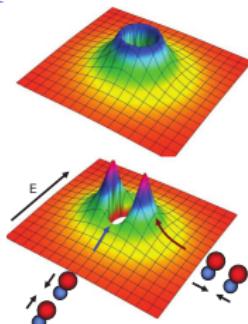
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# From Atom to Molecule

## Atom

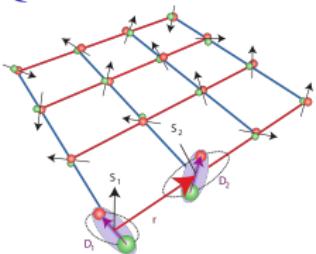
- Laser cooling/trapping
- Internal state control
- High fidelity imagining

## Quantum Chemistry



Nature 464, 1324 (2010)

## Quantum Simulation

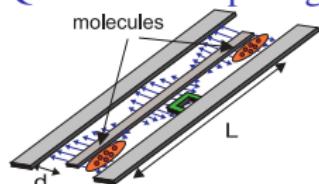


Nat. Phys. 2, 341 (2006)

## Molecule

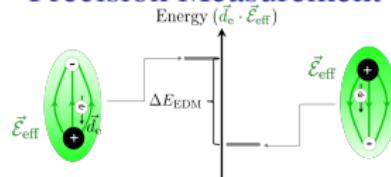
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- Rich internal structure

## Quantum Computing



Phys. Rev. Lett. 97, 33003 (2006)

## Precision Measurement

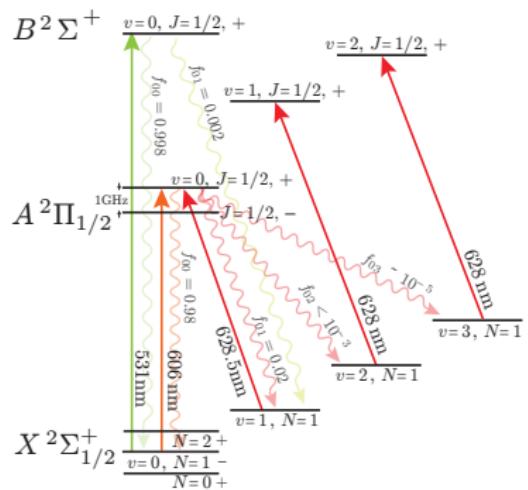


Science 343, p. 269-272 (2014)

# Path to Ultracold Molecules

## Direct molecule cooling

“Diagonal molecules”:  
CaF, SrF, YO, . . .

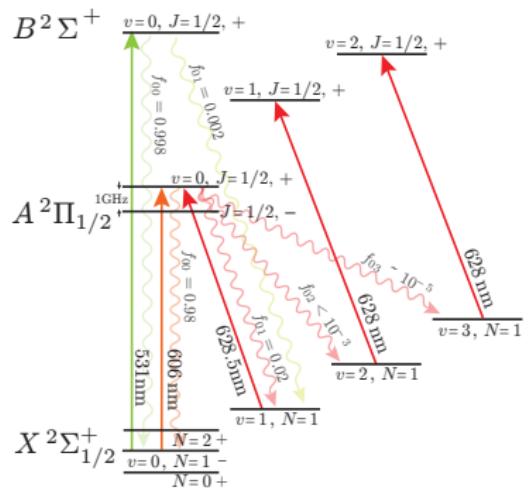


Phys. Rev. Lett. 119, 103201 (2017)

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## Direct molecule cooling

“Diagonal molecules”:  
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Phys. Rev. Lett. 119, 103201 (2017)

## Making molecule from atoms

Bi-Alkali/Alkaline Earth:  
KRb, RbCs, NaK, LiNa, . . .

- ➊ Cold gas of atoms
- ➋ Feshbach association
- ➌ STIRAP

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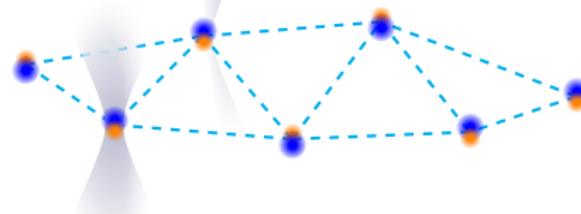
## Assemble molecule in tweezers

- ① Atoms in tweezers
- ② Optical association

## Making molecule from atoms

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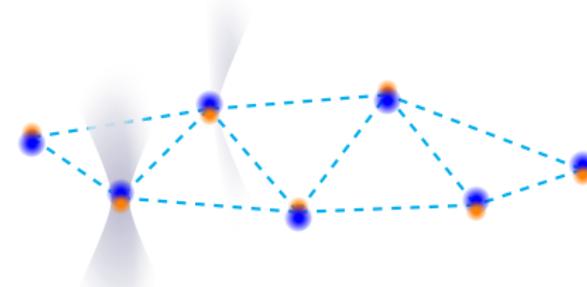
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## Assemble molecule in tweezers

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- Single site detection
- Single site addressing
- Control loading and interaction by rearranging.



## Making molecule from atoms

Bi-Alkali/Alkaline Earth:  
KRb, RbCs, NaK, LiNa, ...

- ① Cold gas of atoms
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# Outline

1 System Overview

2 Trapping and Cooling of Atoms

3 Atom-Atom Interaction and Molecule Formation

# Steps



Loading



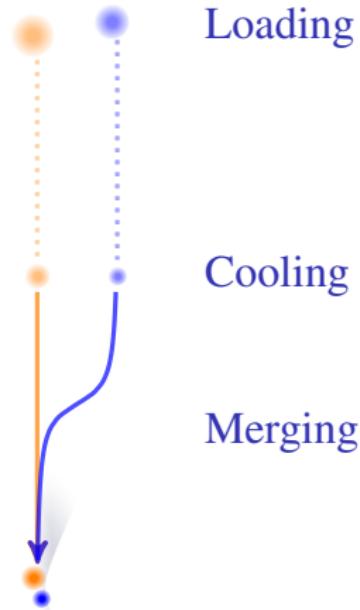
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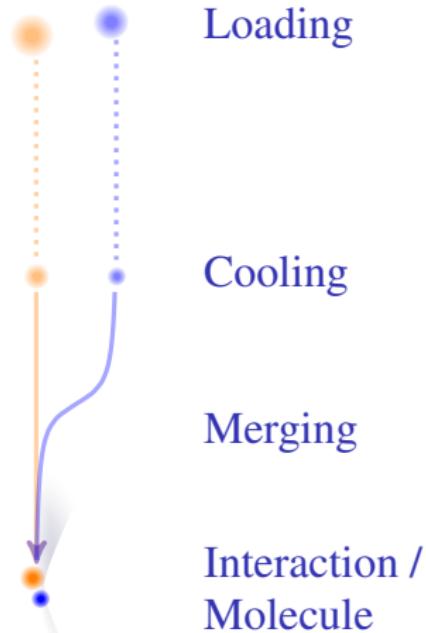
Loading

Cooling

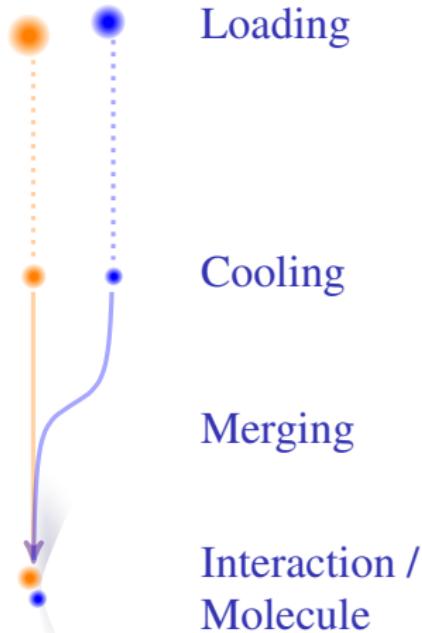
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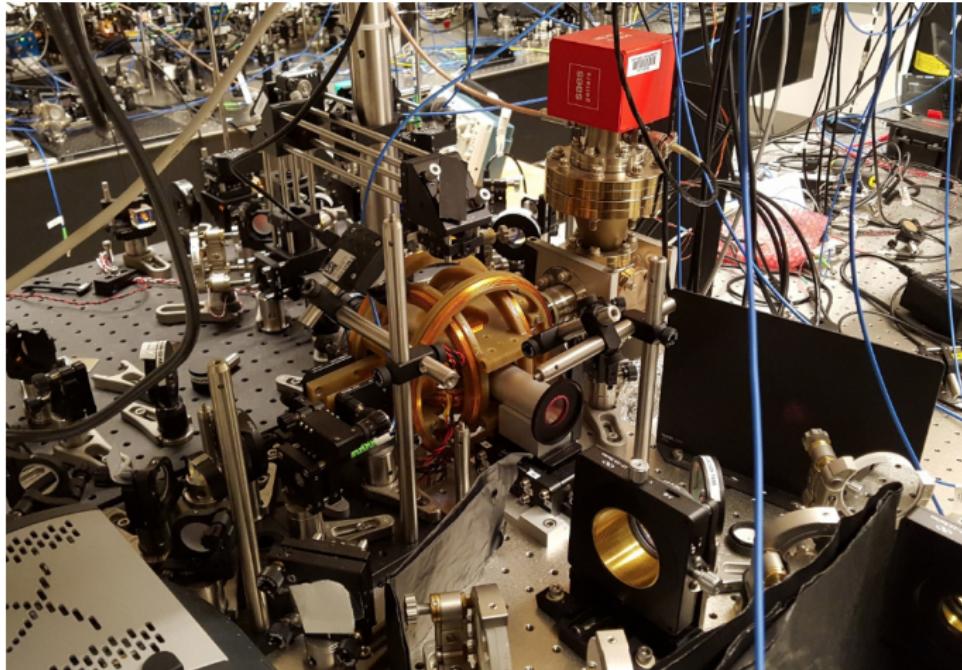
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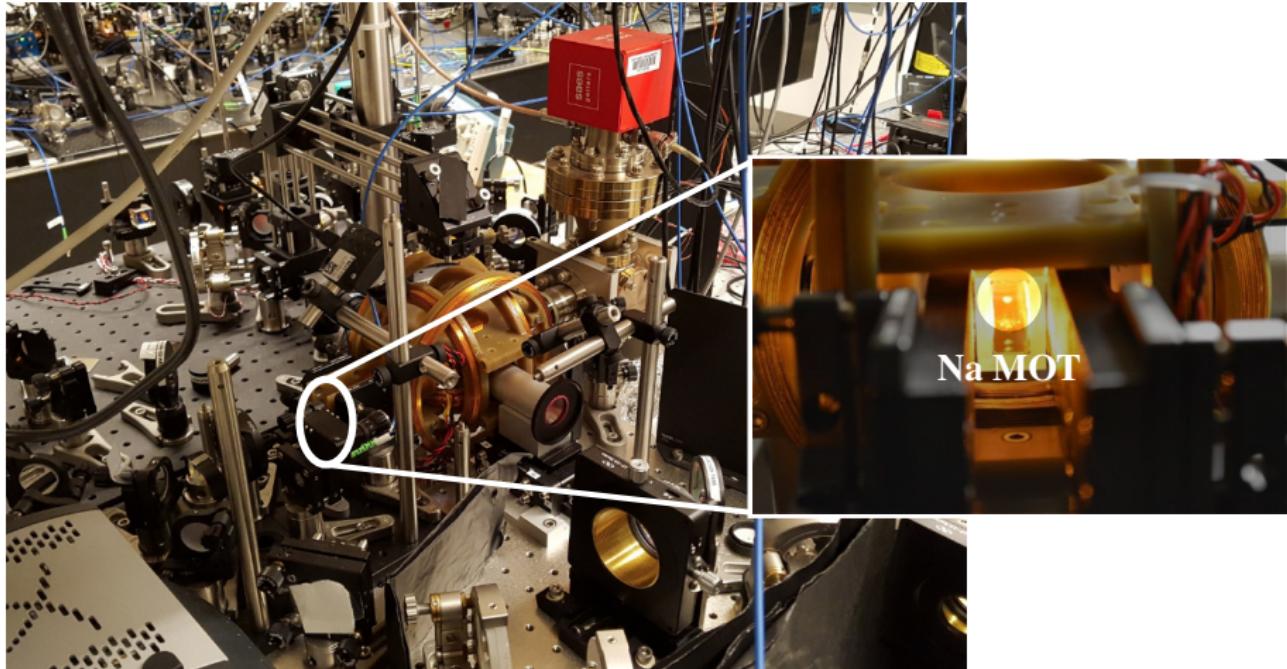
Merging

Interaction /  
Molecule



Science 360 6391, 2018



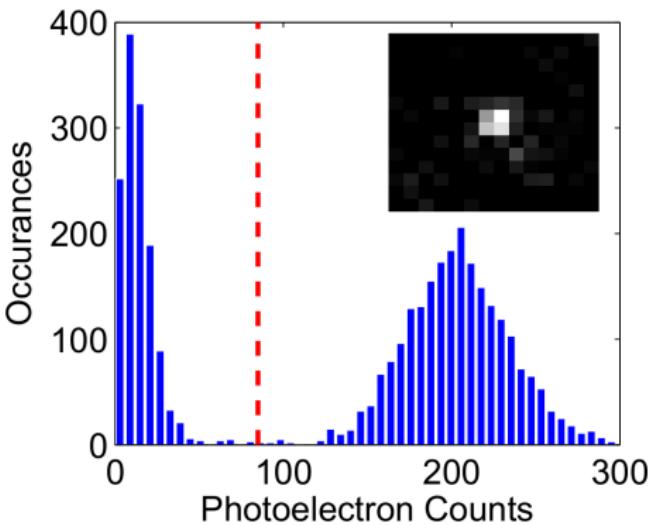


# Single Atom in Tweezer

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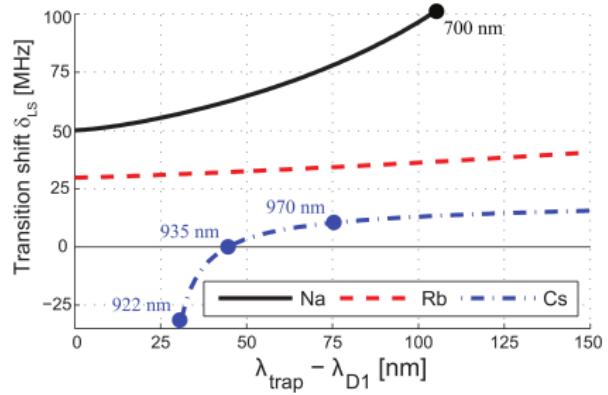
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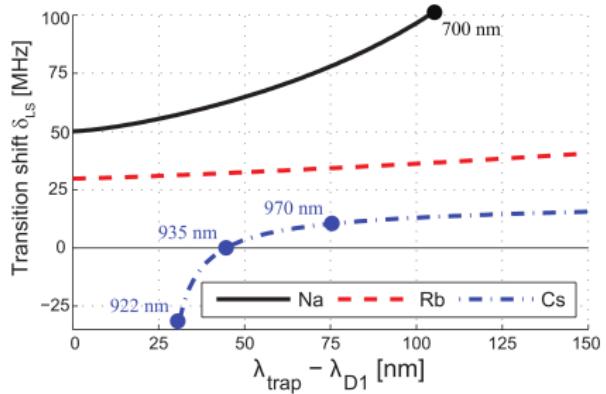
## Issues with Na

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

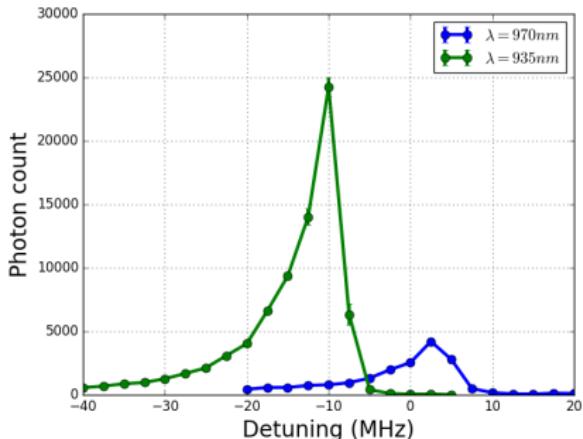
## Real Issue with Na: Light Shift



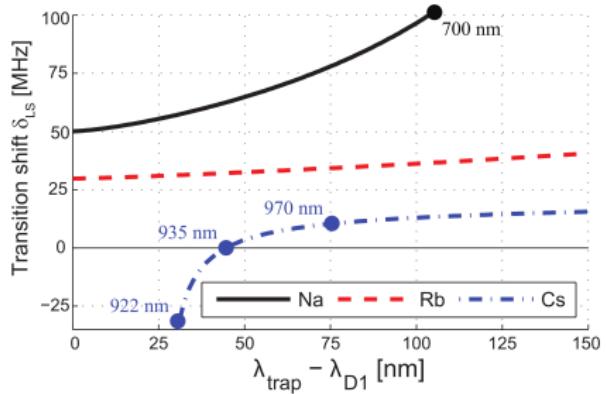
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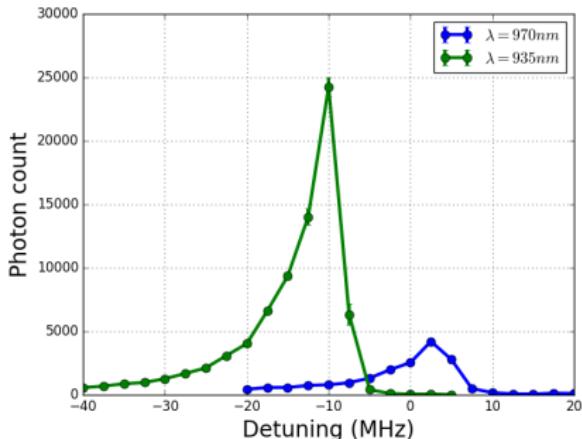
## Cs single atom imaging



## Real Issue with Na: Light Shift



## Cs single atom imaging



- Low imaging signal
- No cooling in tweezer

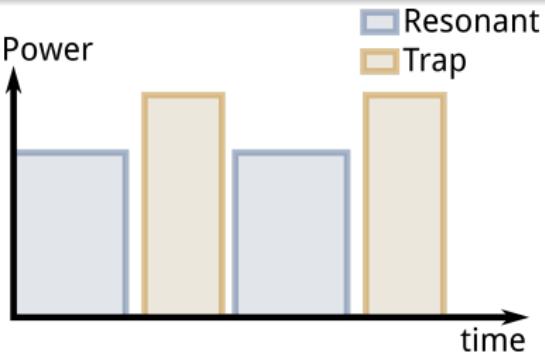
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## Trap modulation

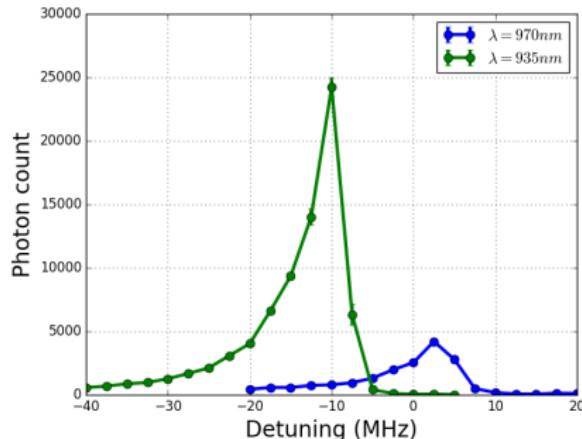
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



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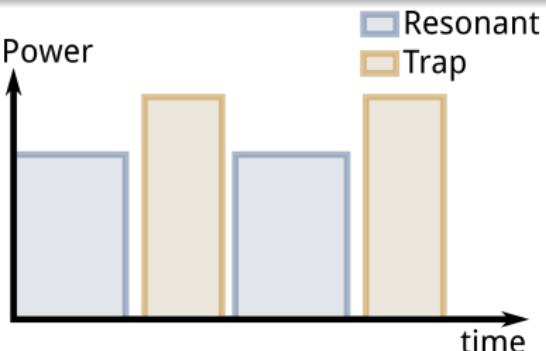
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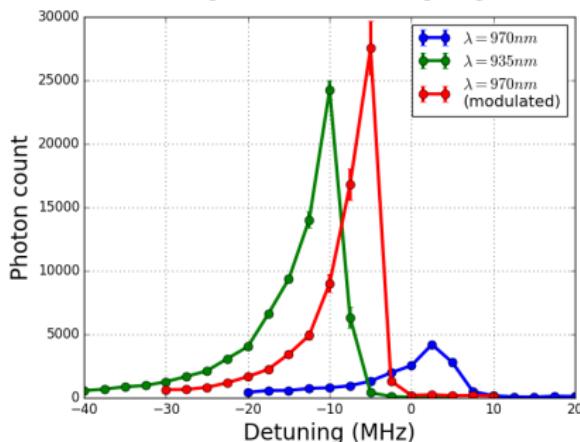
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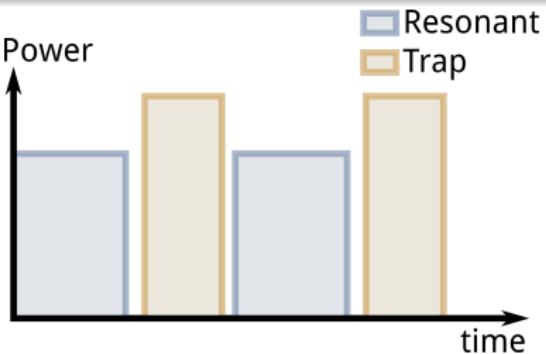
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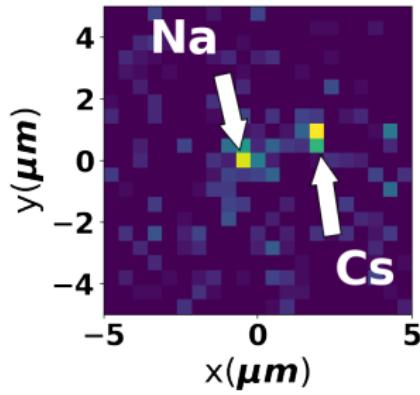
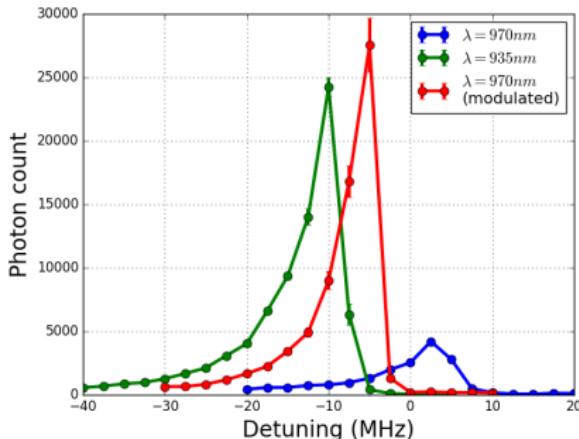
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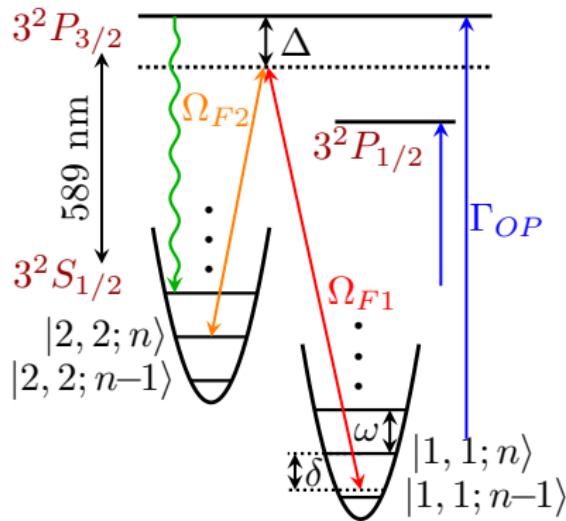
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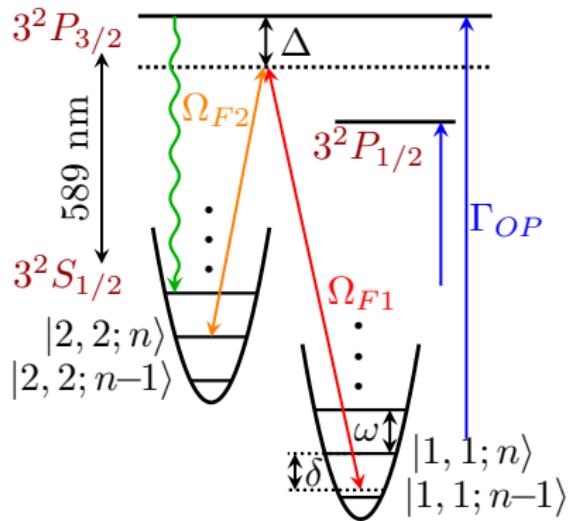
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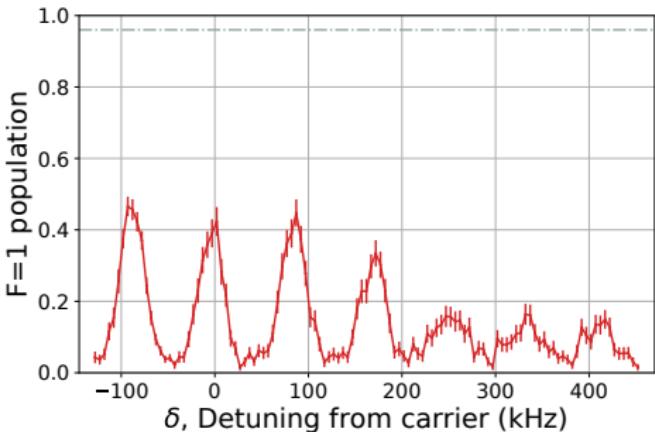
# Raman Sideband Cooling



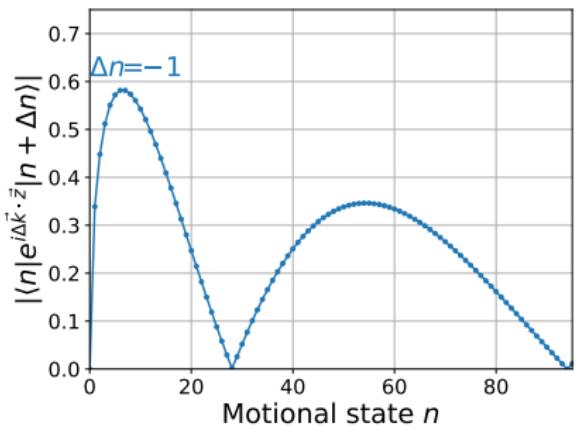
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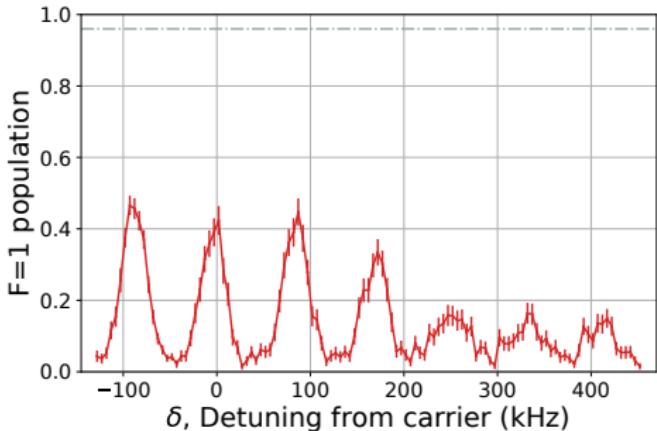
Axial sideband spectrum



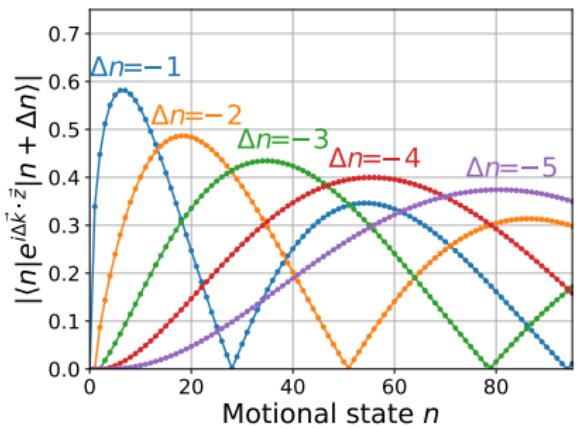
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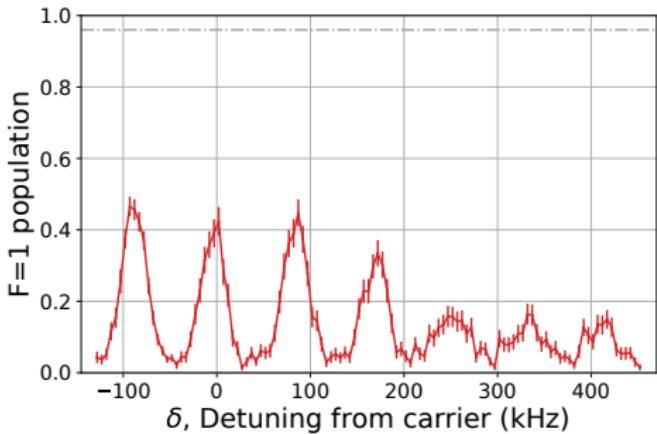
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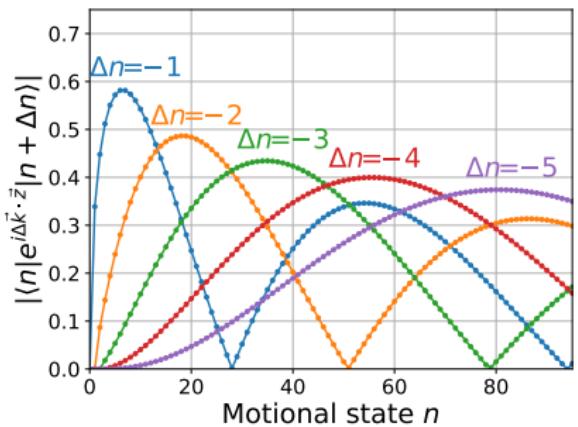
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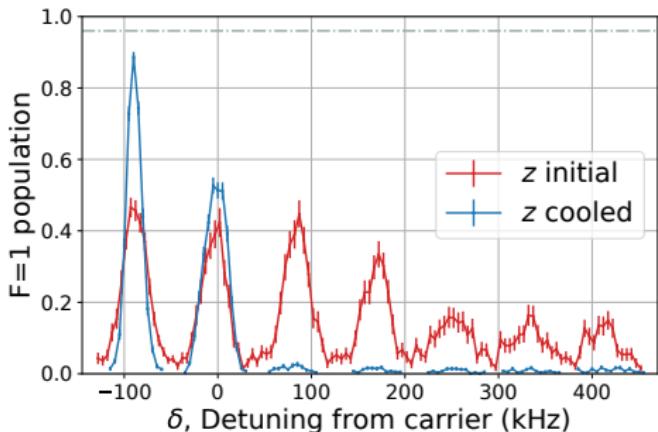
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# Raman Sideband Cooling



Axial sideband spectrum



3D ground state: 93.5(7)%

# Outline

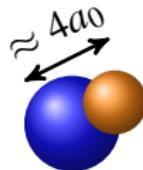
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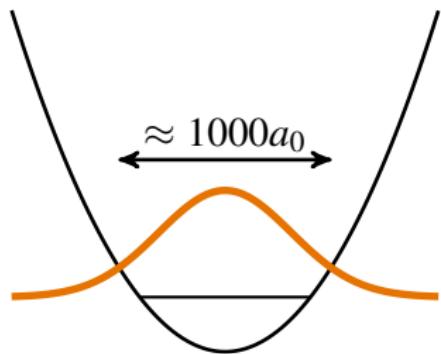
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# Optical Transfer to Molecular State

Binding energy  
 $\approx 150\text{THz}$



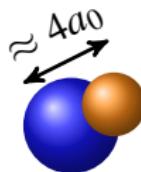
Molecule



Atoms

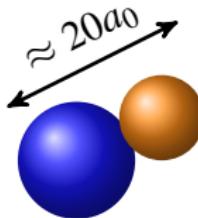
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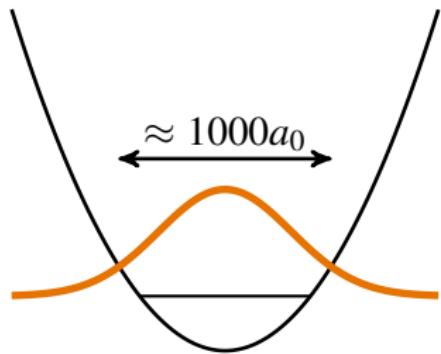


Molecule

Binding energy  
 $\approx 300\text{MHz}$

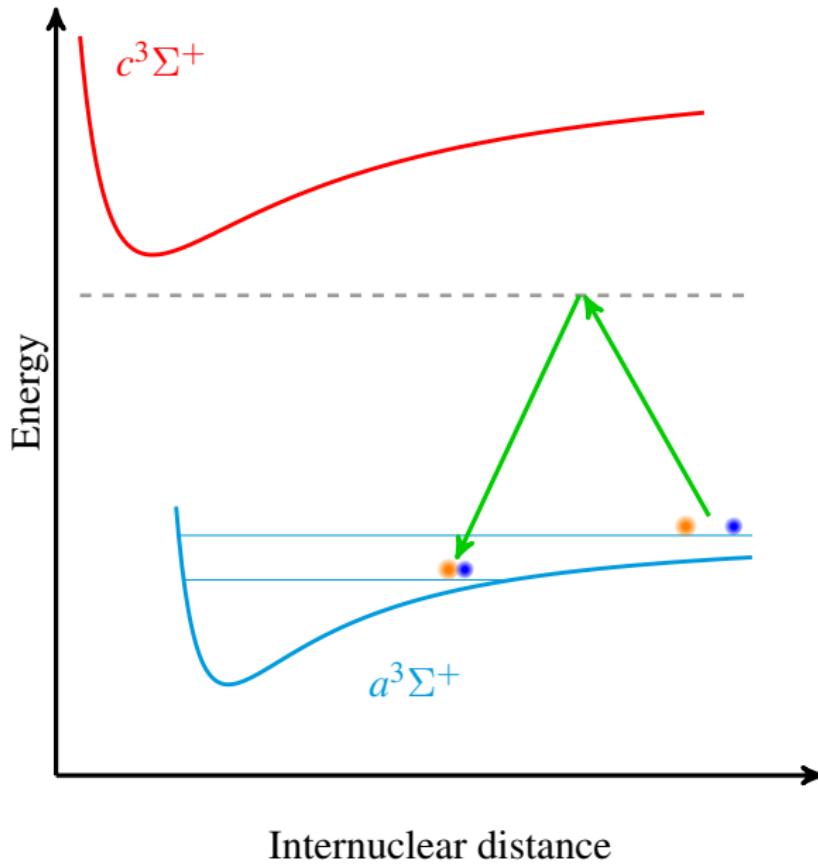


Weakly-Bound  
Molecule

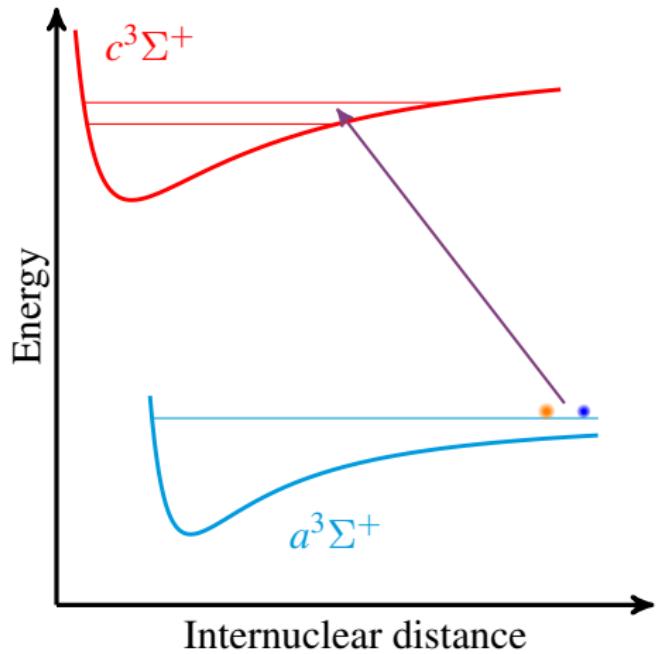


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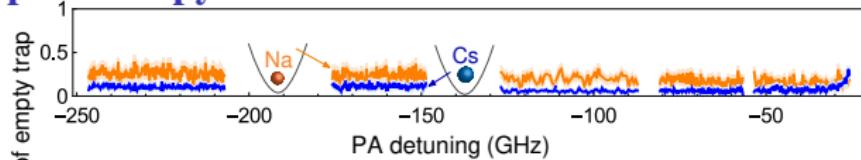
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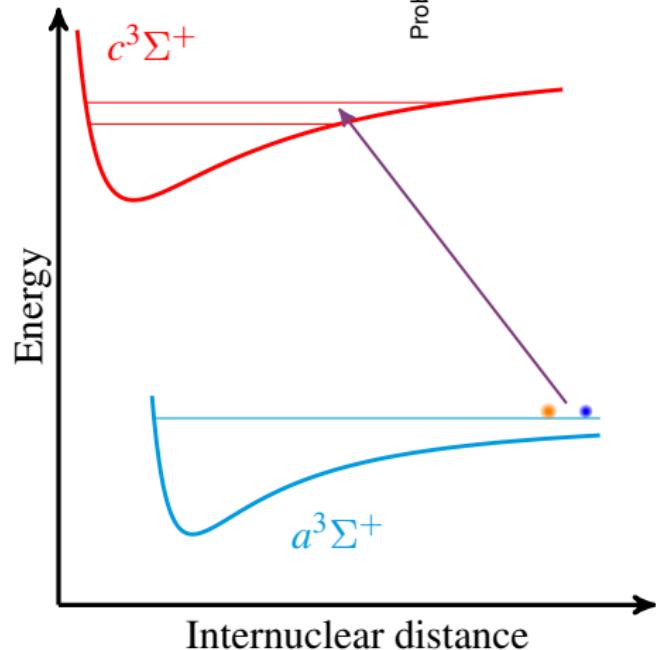
# Photoassociation (PA) Spectroscopy



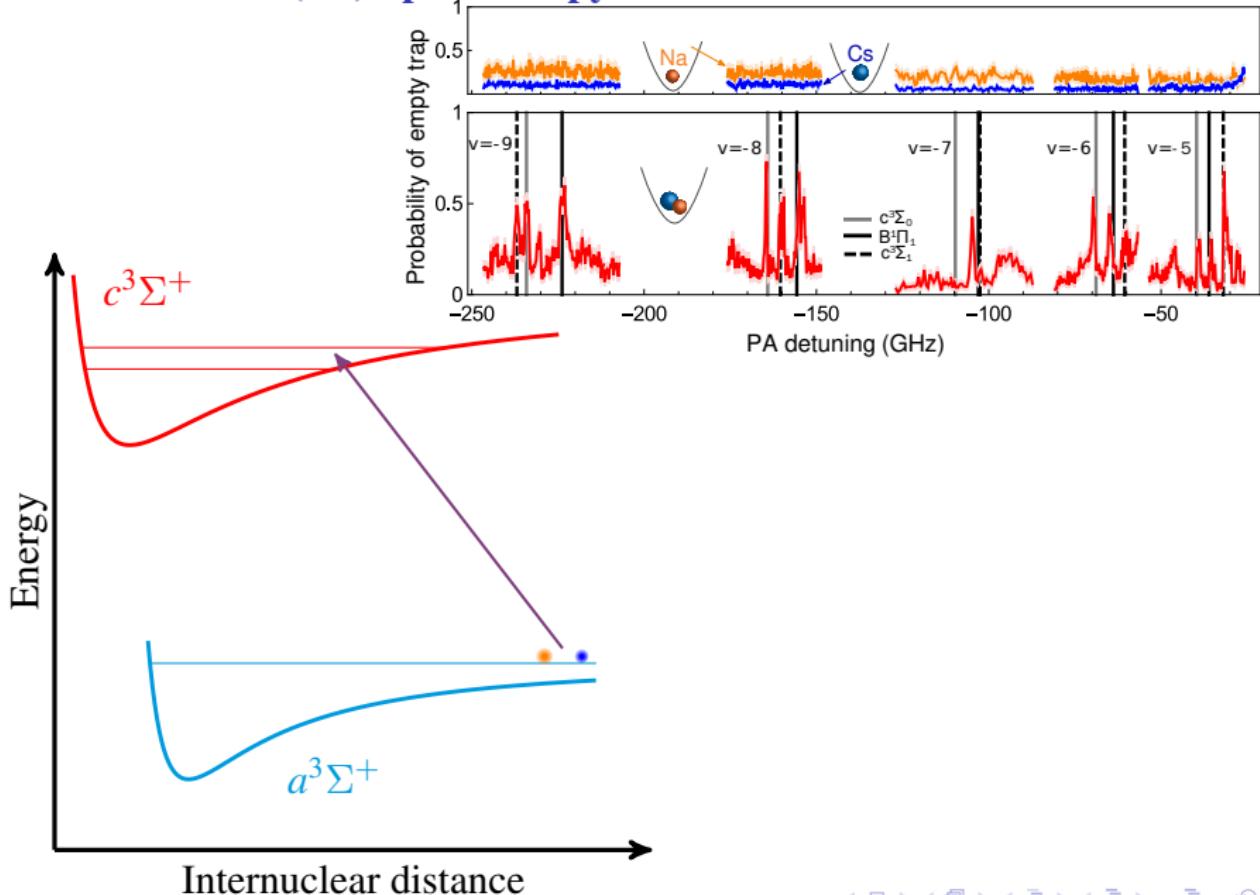
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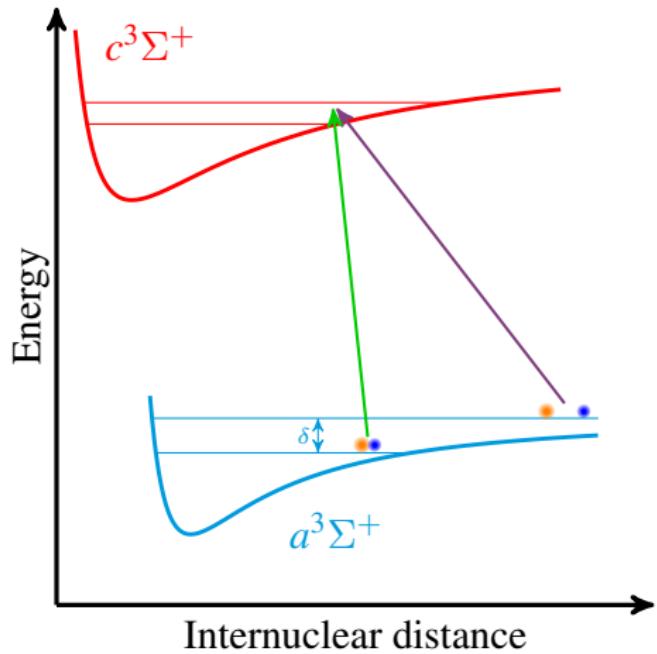
Probability of empty trap



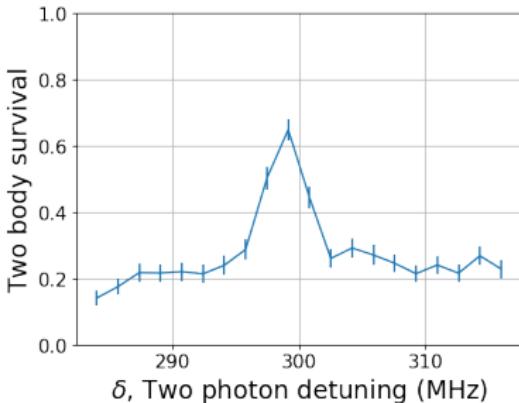
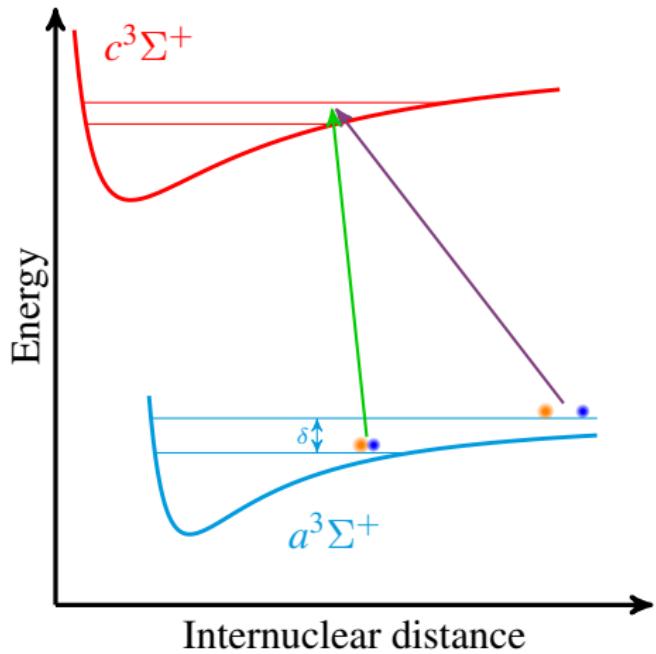
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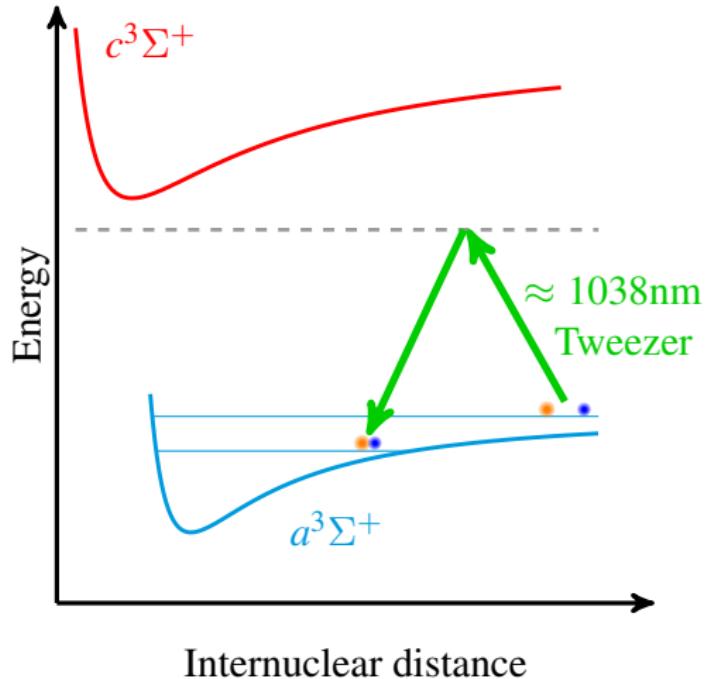
# Electromagnetically Induced Transparency (EIT) Spectroscopy



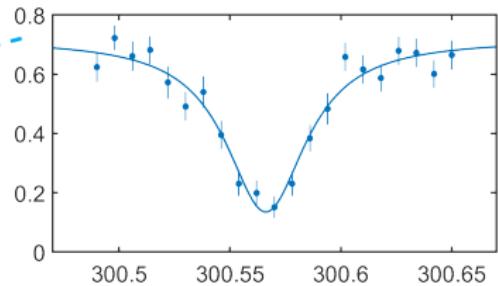
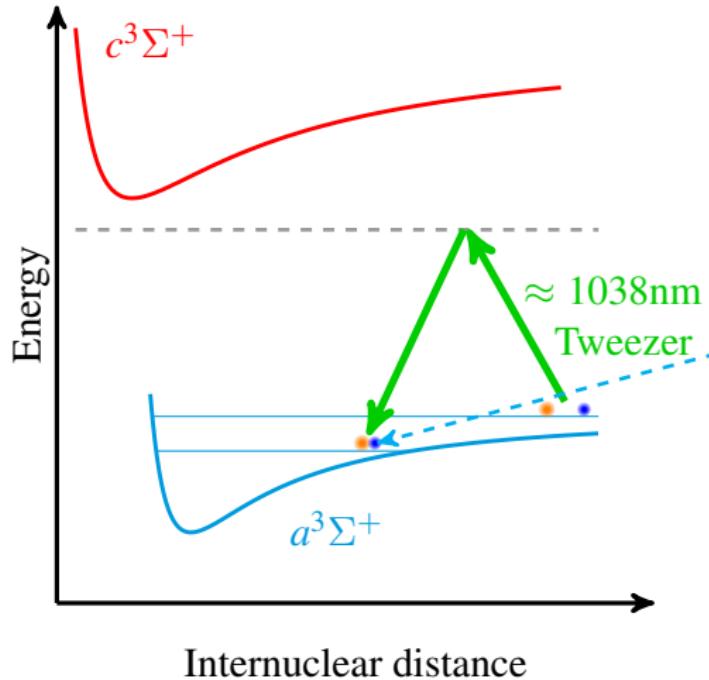
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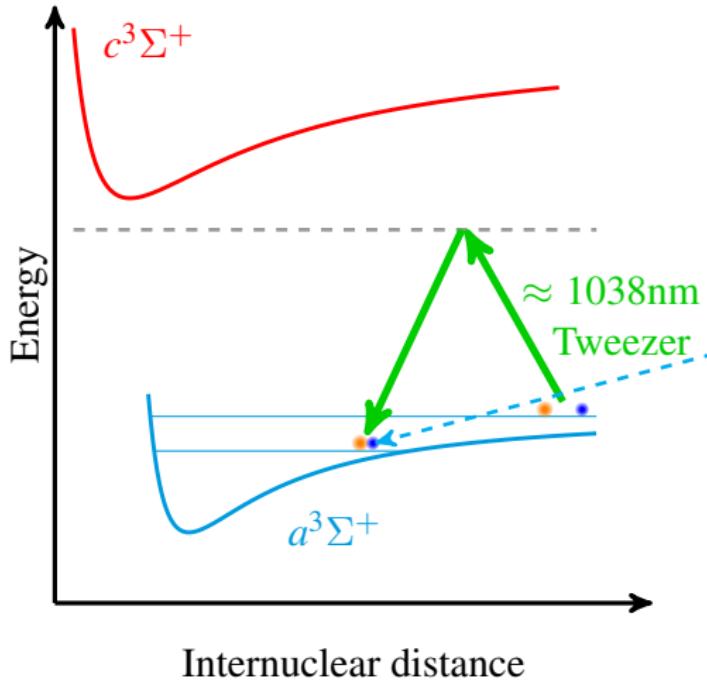
# Optical Transfer to Weakly-Bound Molecular



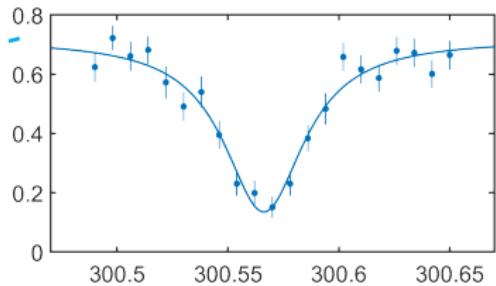
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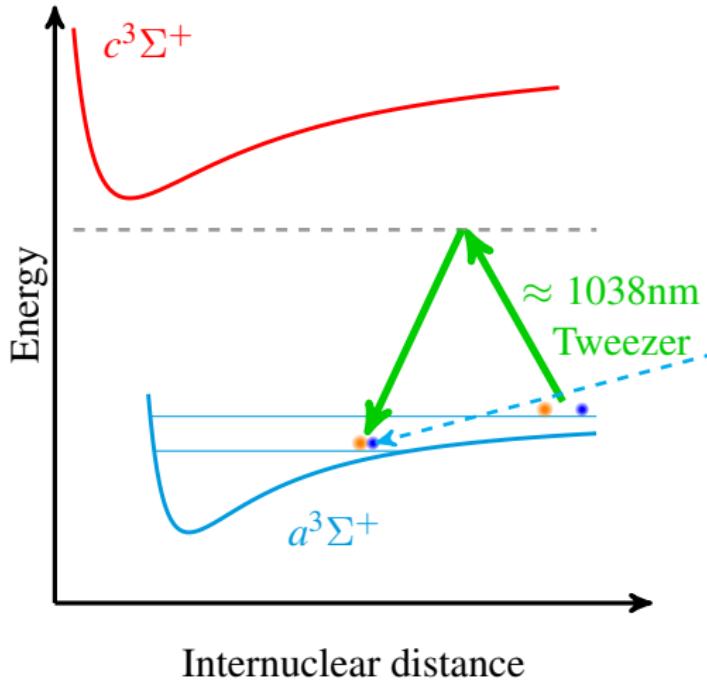
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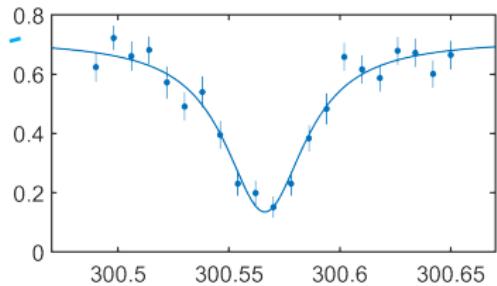


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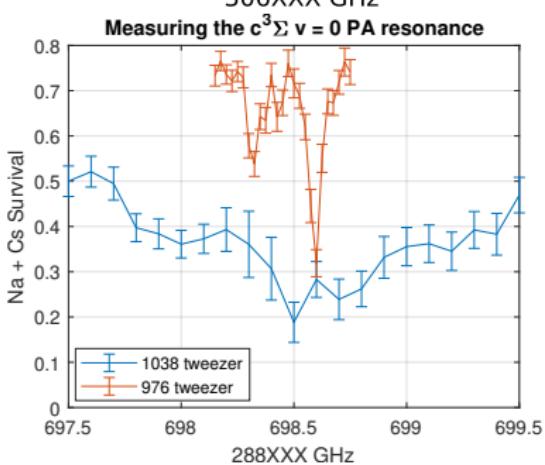
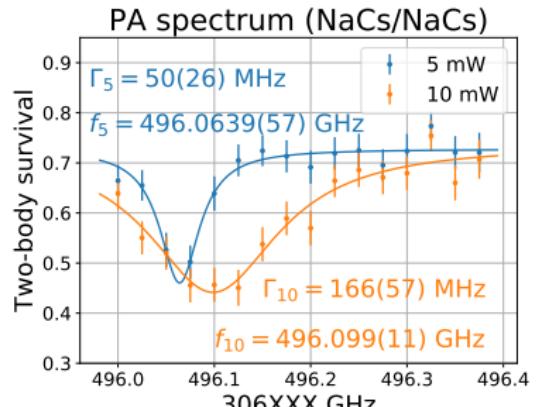


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$\Gamma_e$  is  $\approx 50 \sim 100$ x theory value.

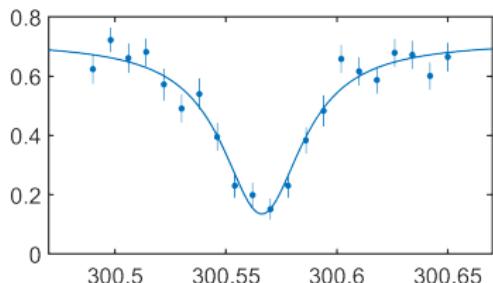


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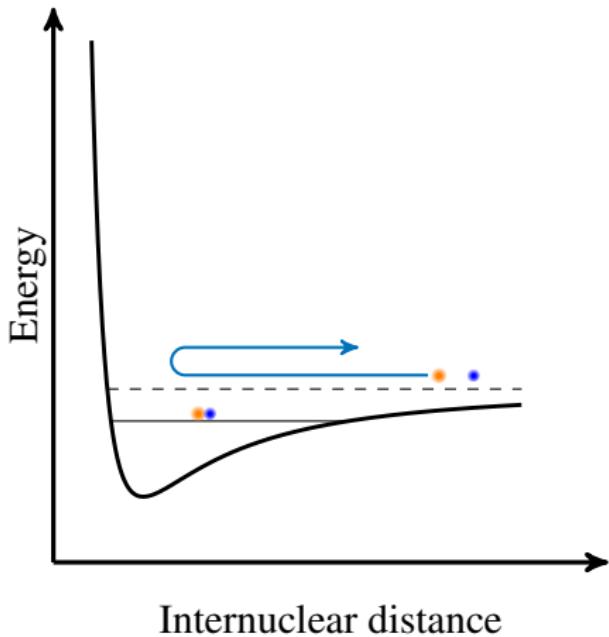
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## Scattering length $a$

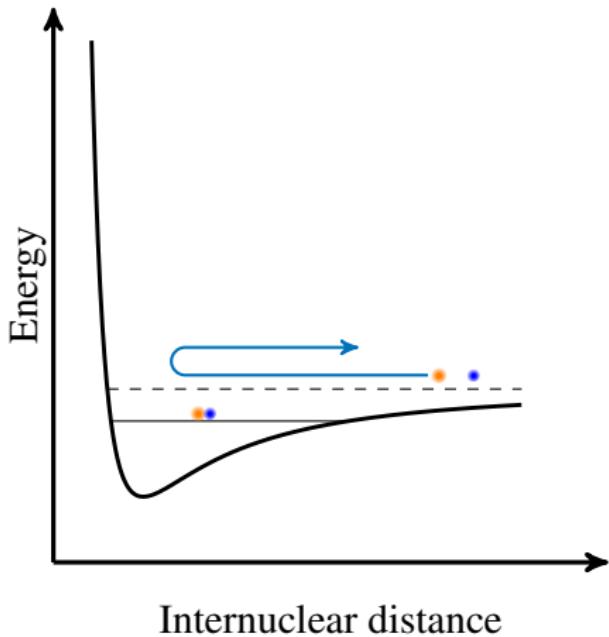
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  - Molecular potential
  - Feshbach resonance
  - Molecule formation

⋮



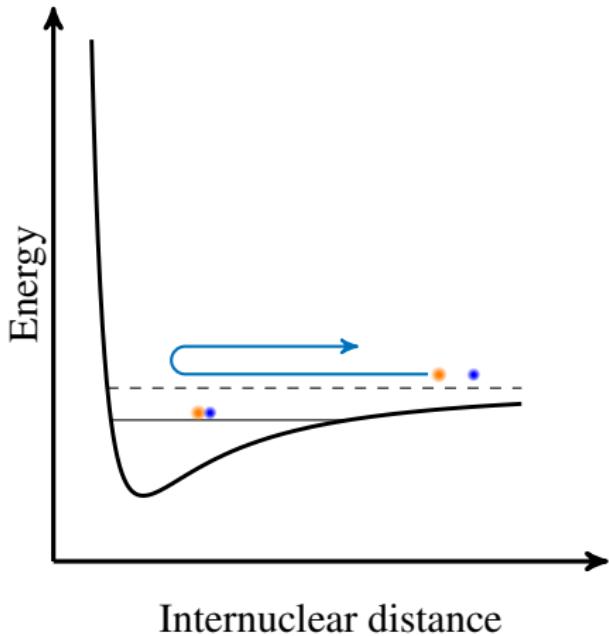
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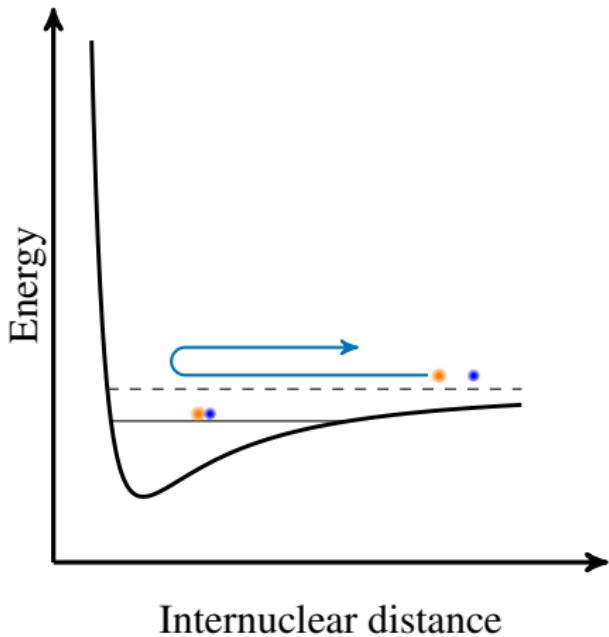
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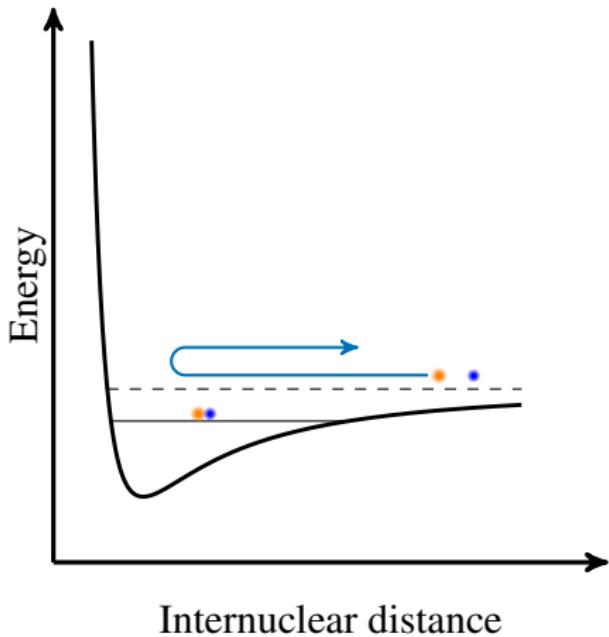
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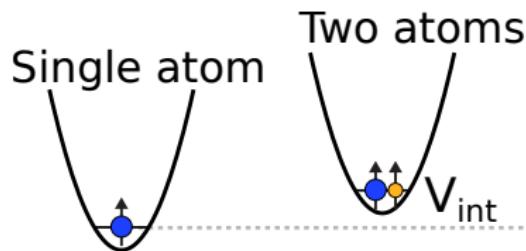


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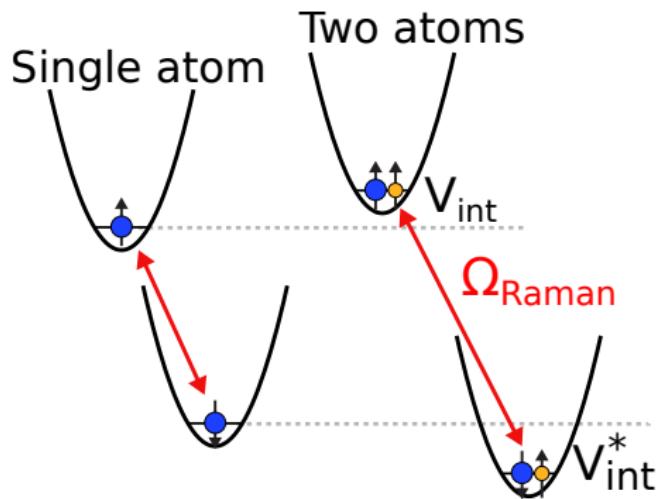
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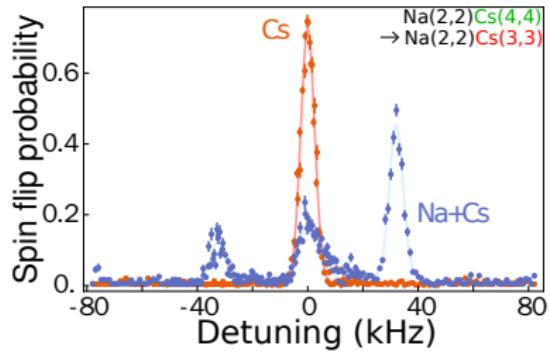
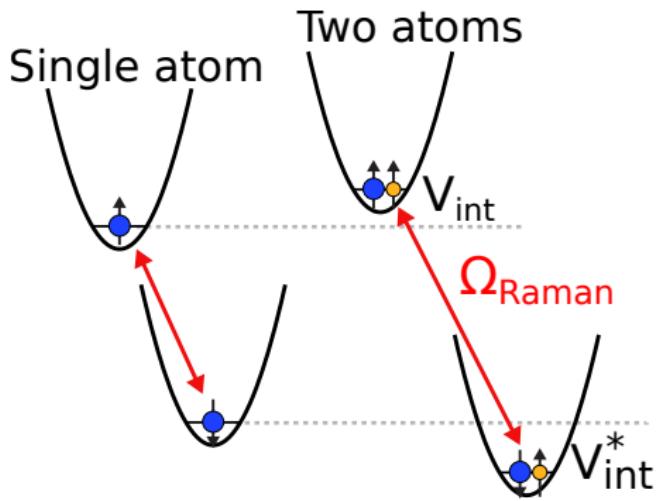
## Interaction shift



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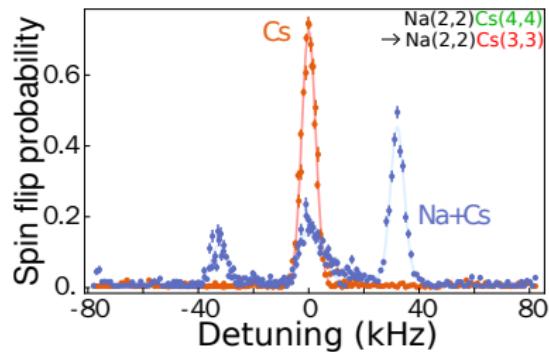
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$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{m_1 \omega_{1,i}^2 x_{1,i}^2}{2} + \frac{p_{1,i}^2}{2m_1} \right)}_{\text{Na}} + \underbrace{\sum_{i=x,y,z} \left( \frac{m_2 \omega_{2,i}^2 x_{2,i}^2}{2} + \frac{p_{2,i}^2}{2m_2} \right)}_{\text{Cs}} +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

Interaction



## Interaction shift

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

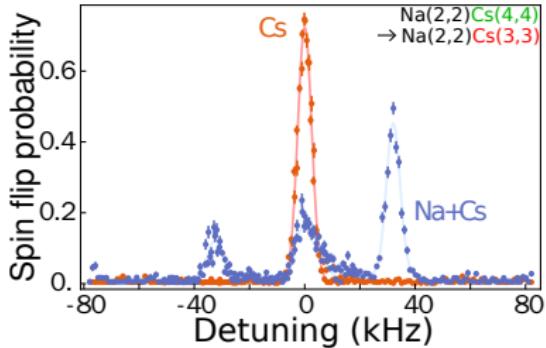
To center of mass  
and relative coordinates

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

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

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

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



### Center of mass

$$H = \overbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}$$

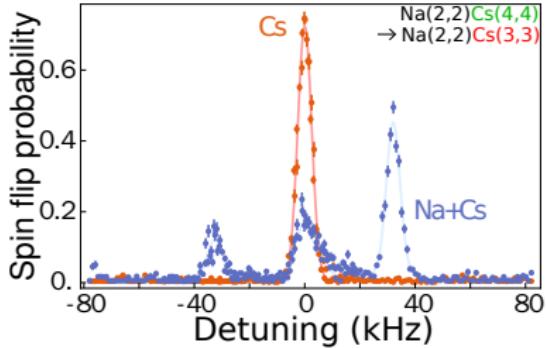
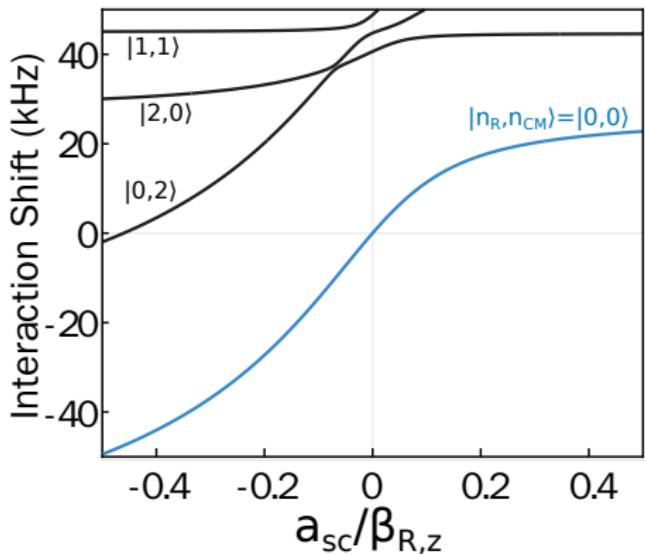
## Relative

$$+ \underbrace{\sum_{i=x,y,z} \left( \frac{\mu \omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right)}_{V_{int}(\vec{r}_R)} + V_{ext}(\vec{r}_R)$$

Mixing

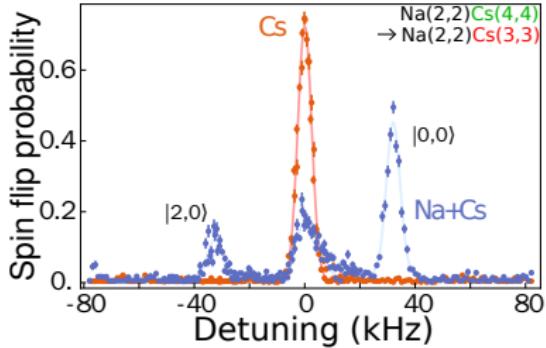
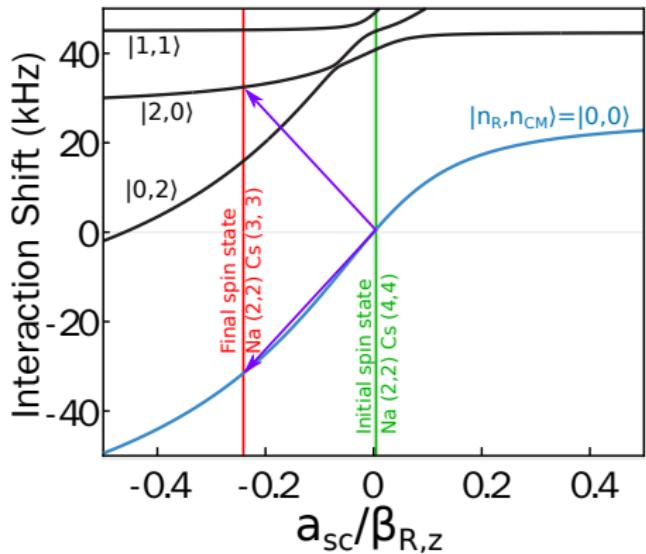
$$+ \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i x_{R,i}}$$

# Interaction shift



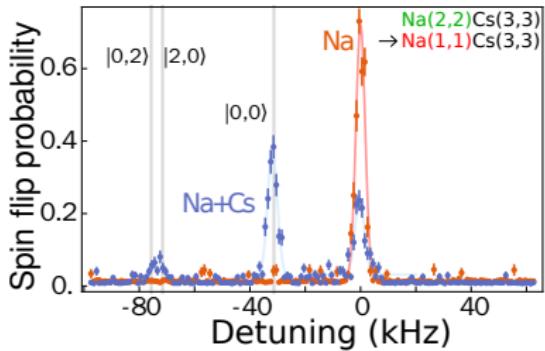
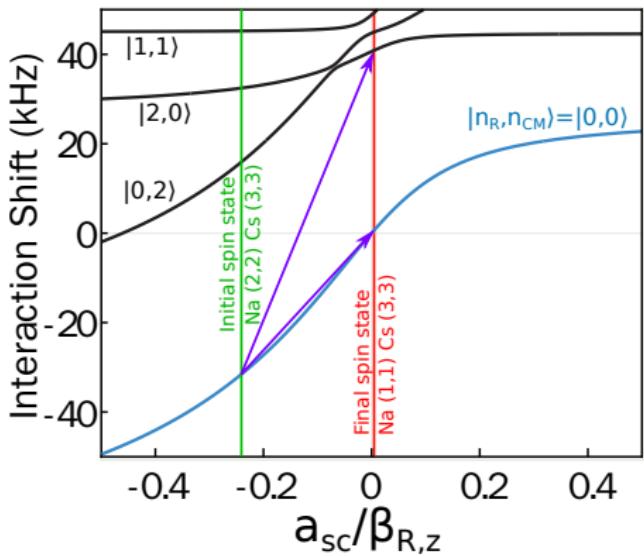
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

# Interaction shift



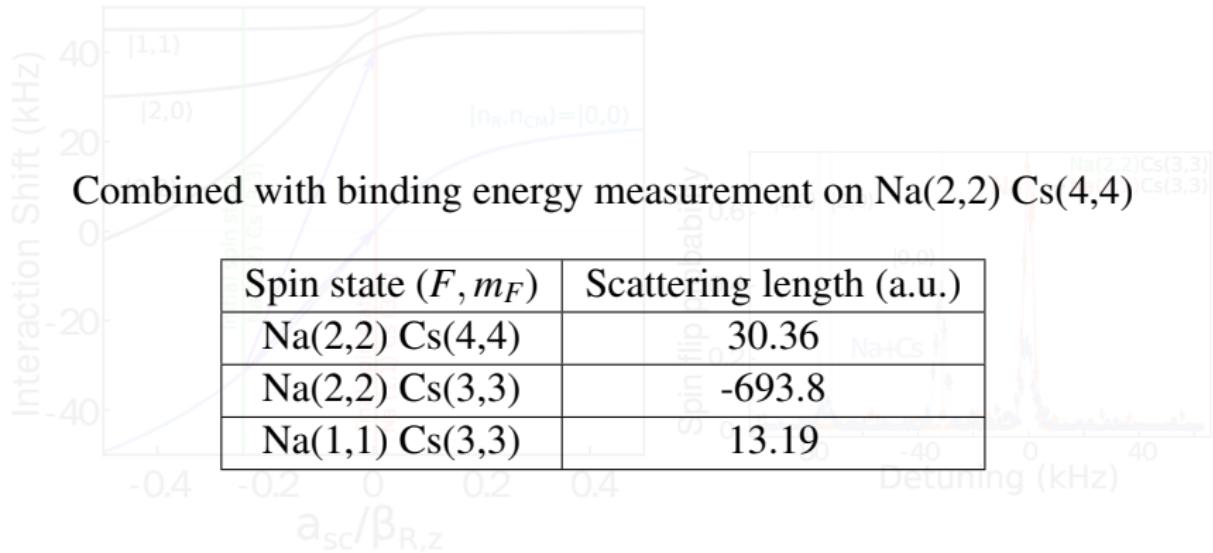
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

## Interaction shift



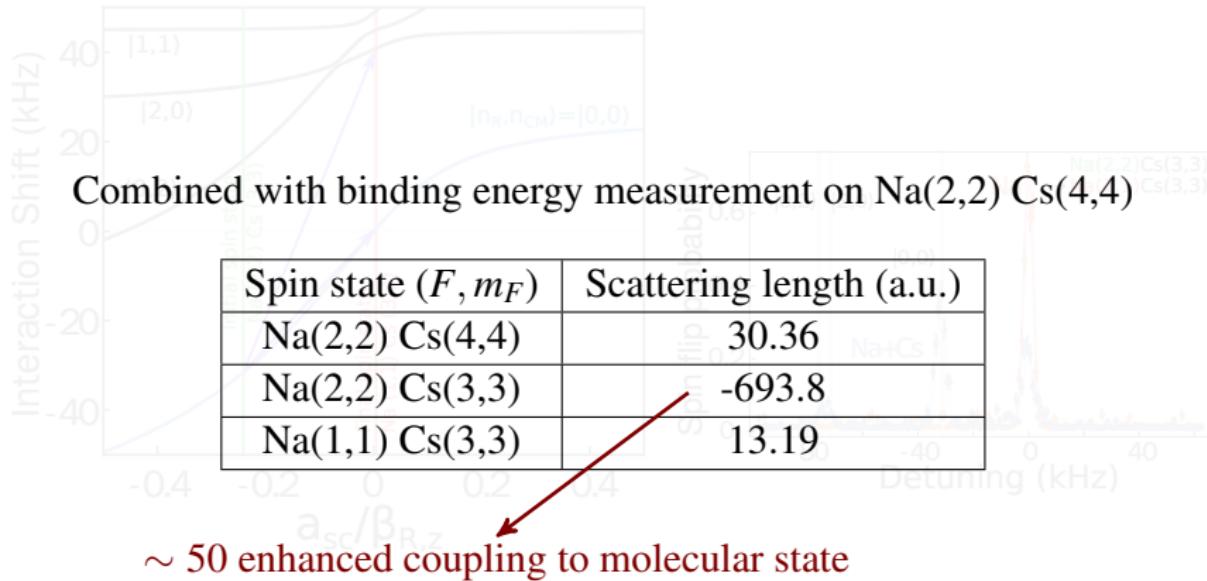
$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

# Interaction shift



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

# Interaction shift

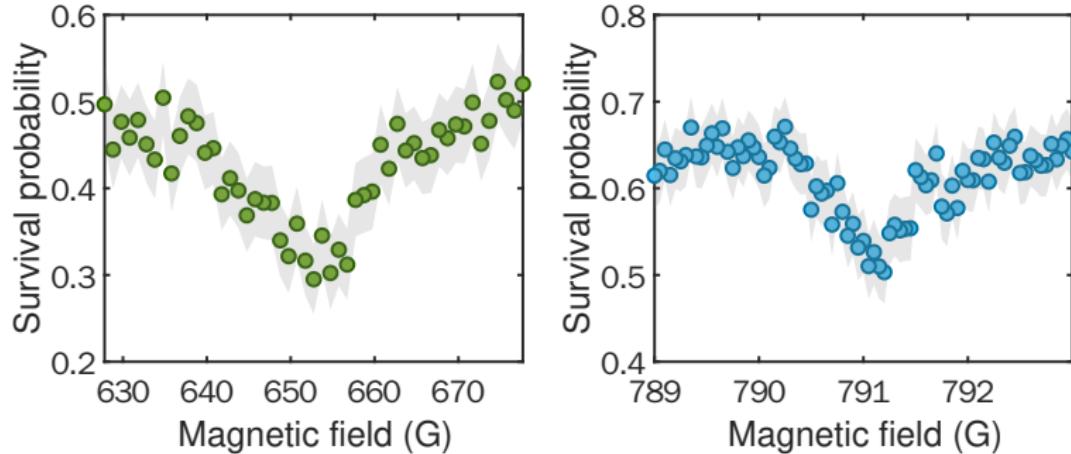


$$H = \underbrace{\sum_{i=x,y,z} \left( \frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left( \frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

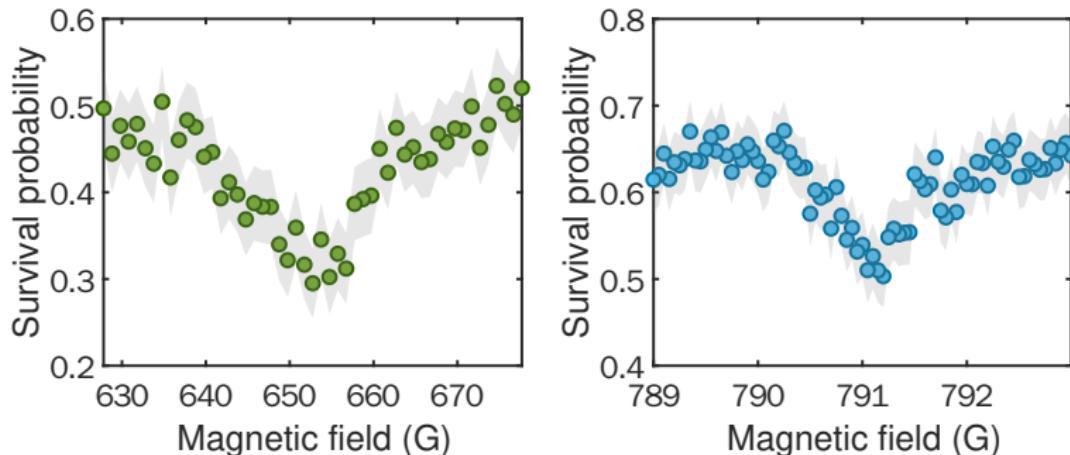
## Na (1, -1) Cs (3, -3) Feshbach resonance



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



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



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

<sup>1</sup>In collaboration with Bo Gao

## Summary

- A single Na and Cs atom prepared in the motional ground state of the same optical tweezer.
- Photoassociation and Raman transfer to weakly-bound state.
- Characterized Na-Cs scattering and observed first Feshbach resonances.



