

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

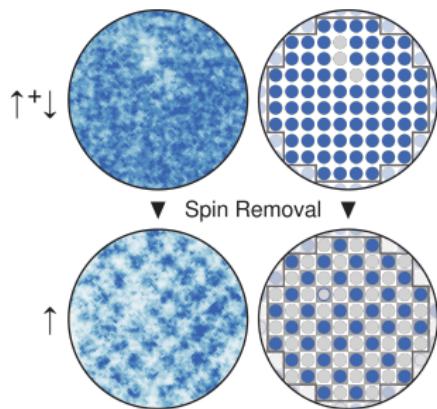
Atom

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

From Atom to Molecule

Atom

- Laser cooling/trapping
- Internal state control
- High fidelity imagining
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Nature 545, 462-466 (2017)

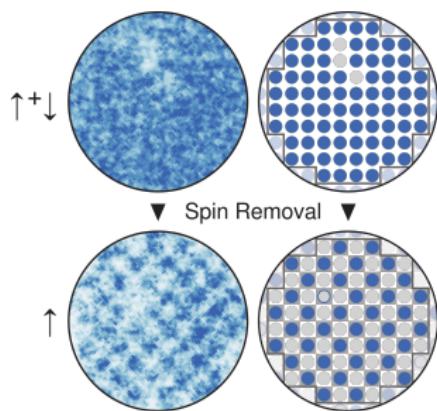
From Atom to Molecule

Atom

- Laser cooling/trapping
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Molecule

- Strong interaction
- Rich internal structure



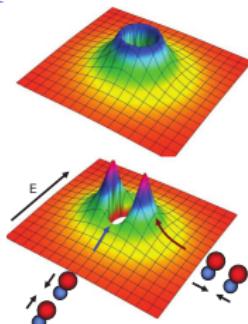
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From Atom to Molecule

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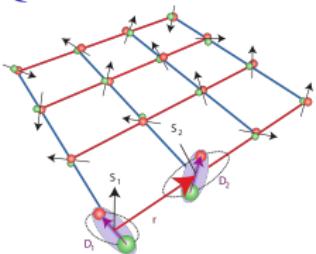
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Quantum Chemistry



Nature 464, 1324 (2010)

Quantum Simulation

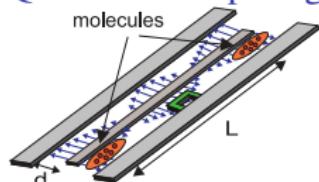


Nat. Phys. 2, 341 (2006)

Molecule

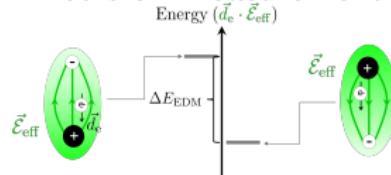
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Quantum Computing



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

Precision Measurement

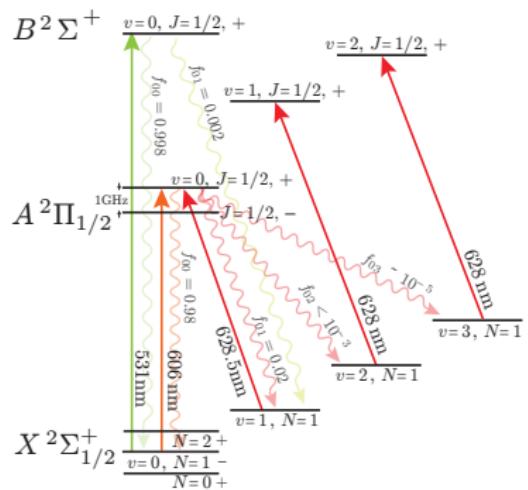


Science 343, p. 269-272 (2014)

Path to Ultracold Molecules

Direct molecule cooling

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

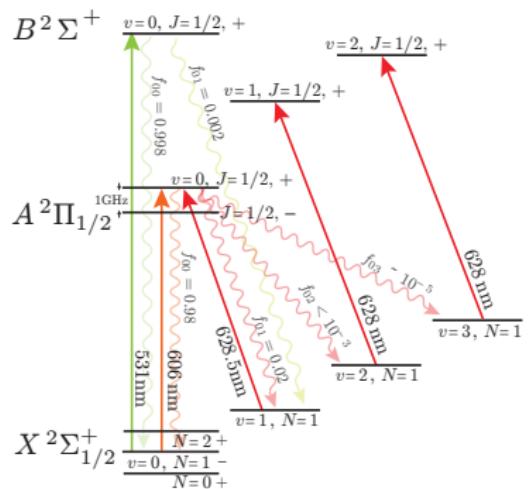


Phys. Rev. Lett. 119, 103201 (2017)

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Making molecule from atoms

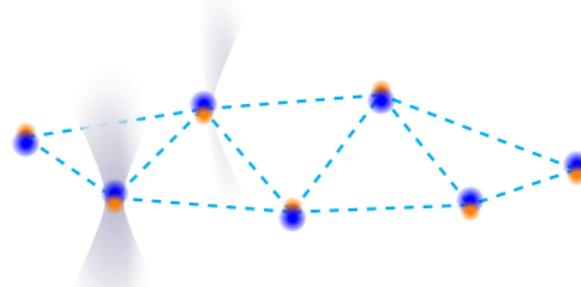
Bi-Alkali/Alkaline Earth:
KRb, RbCs, NaK, LiNa, . . .

- 1 Cold gas of atoms
- 2 Feshbach association
- 3 STIRAP

Path to Ultracold Molecules

Assemble molecule in tweezers

- ① Atoms in tweezers
- ② Optical association



Making molecule from atoms

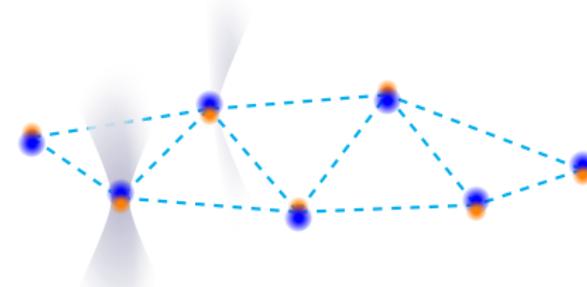
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Assemble molecule in tweezers

- ① Atoms 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



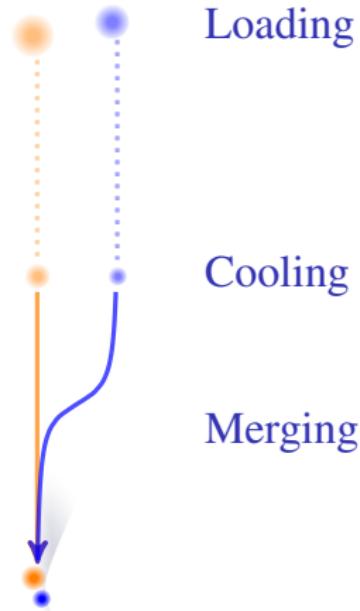
Steps



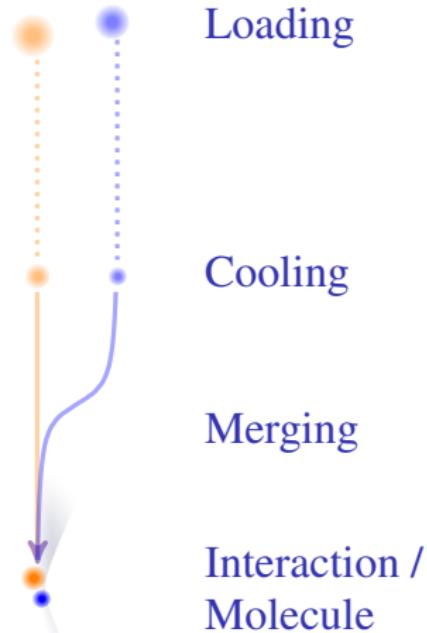
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Cooling

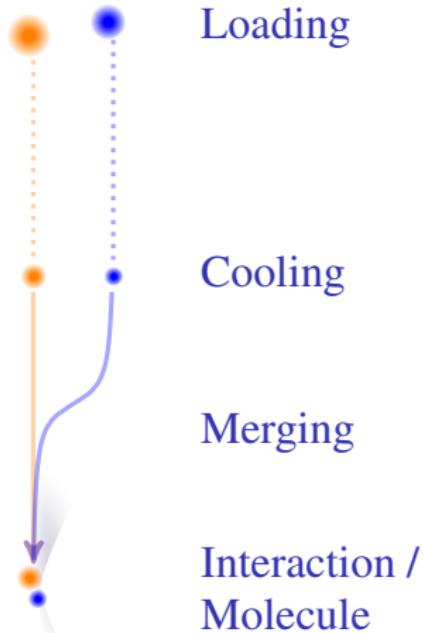
Steps



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Steps



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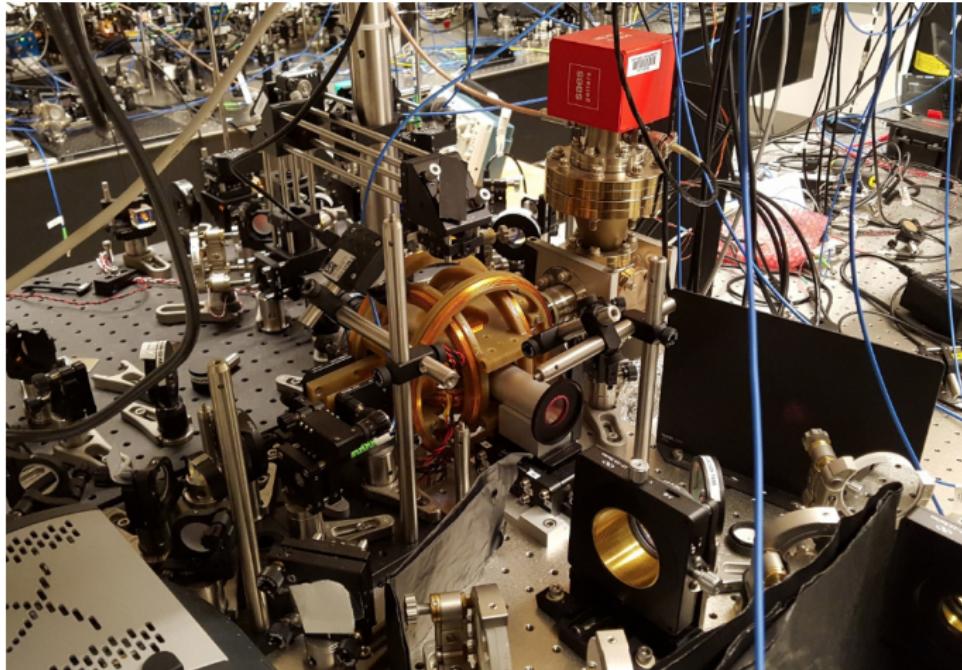
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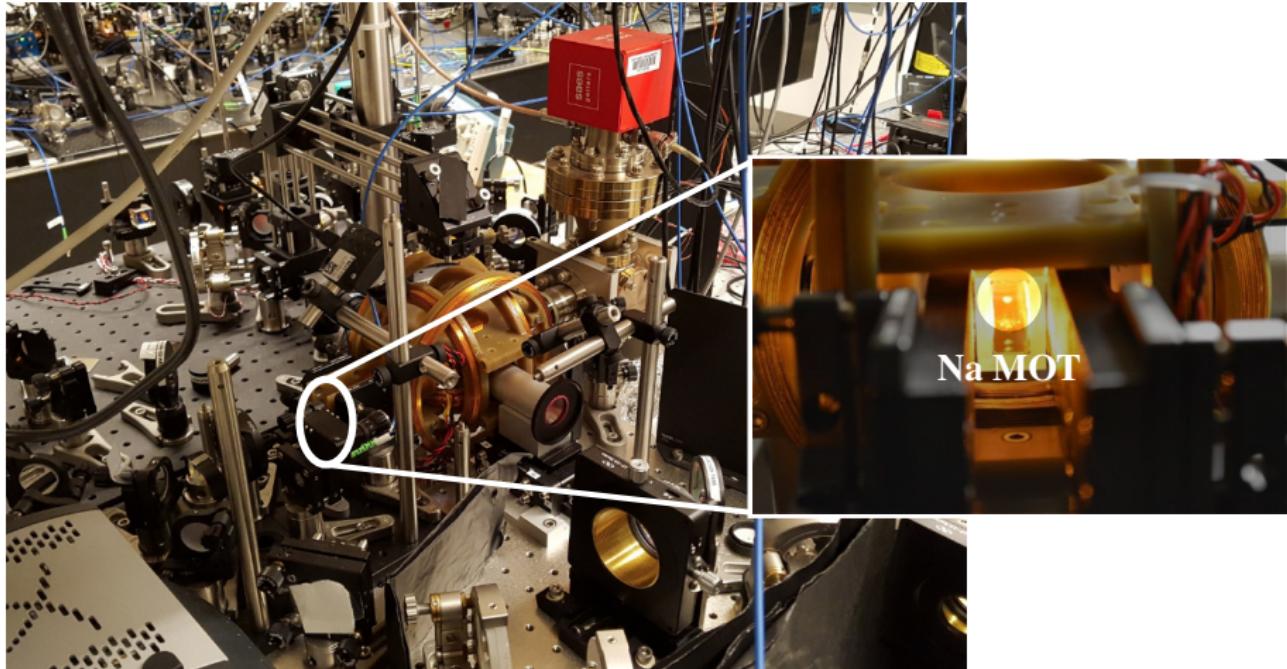
Merging

Interaction /
Molecule



Science 360 6391, 2018



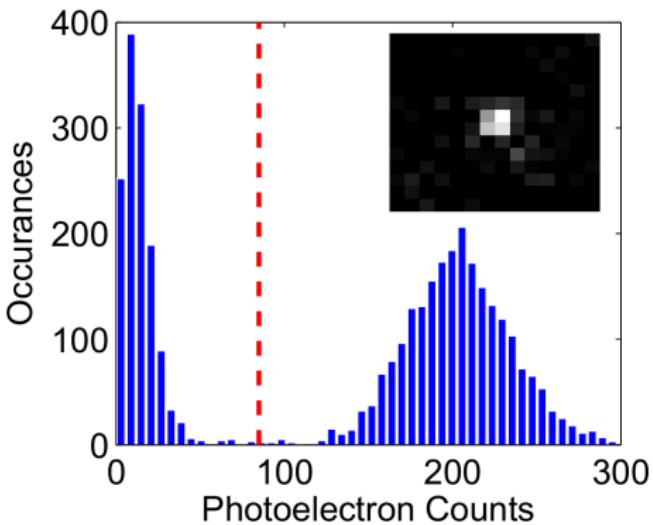


Single Atom in Tweezer

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

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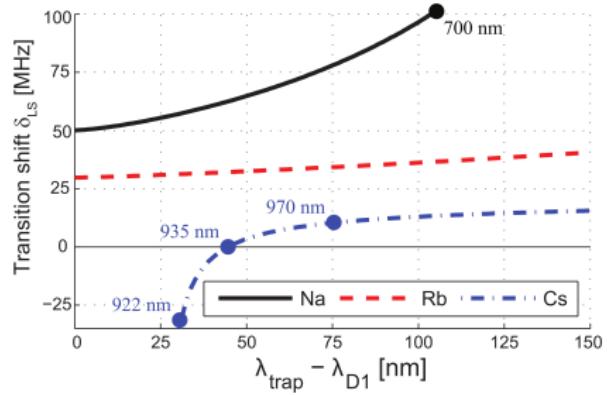
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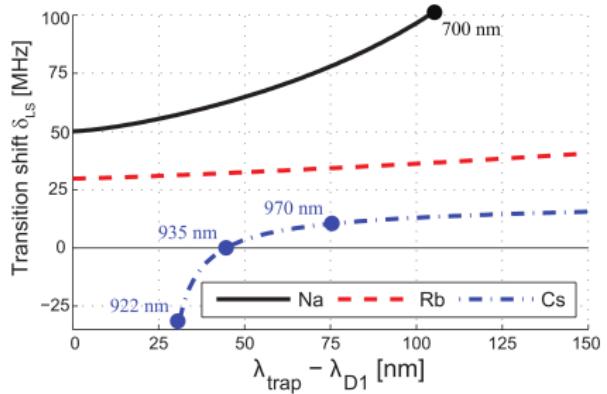
Issues with Na

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

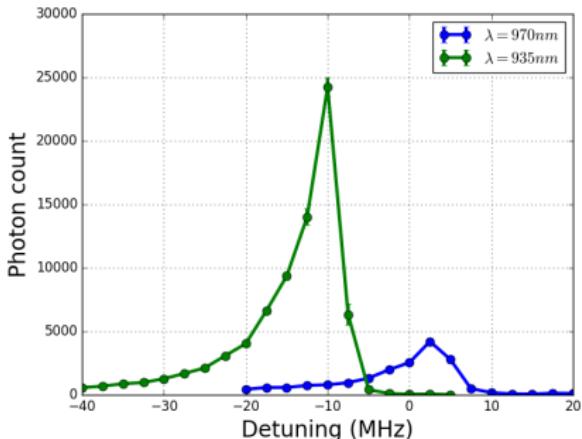
Real Issue with Na: Light Shift



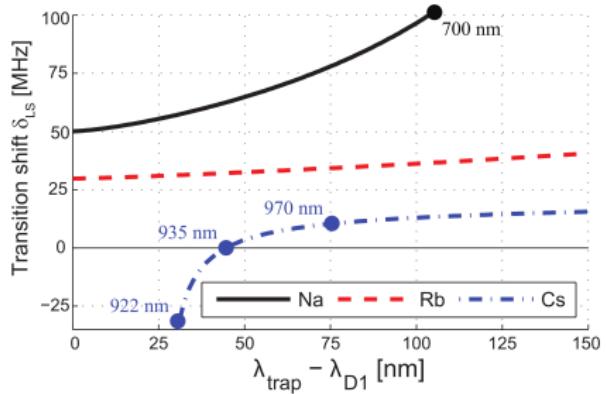
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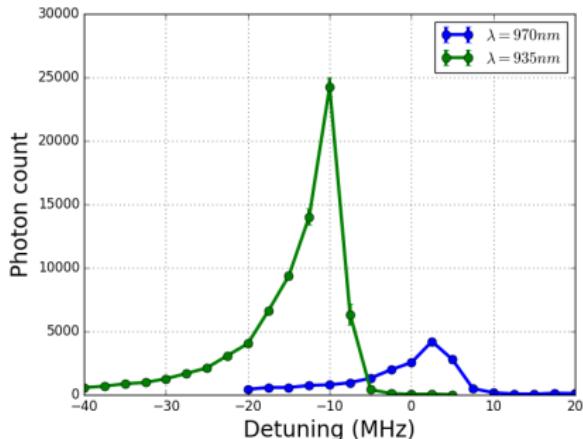
Cs single atom imaging



Real Issue with Na: Light Shift



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- Low imaging signal
- No cooling in tweezer

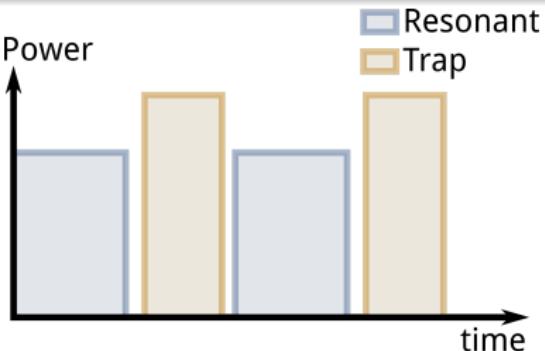
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Trap modulation

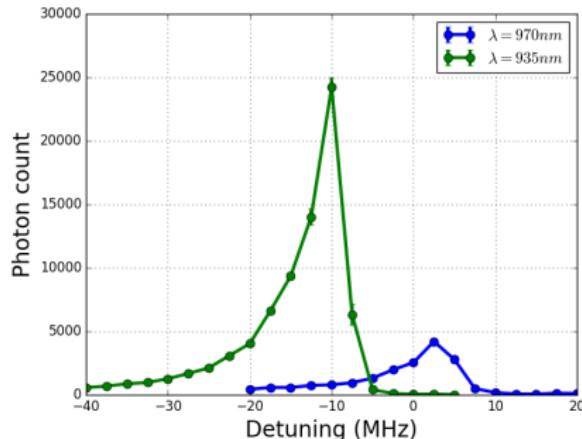
Alternate between trap and resonant (cooling and imaging) light at 2.5 MHz

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

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



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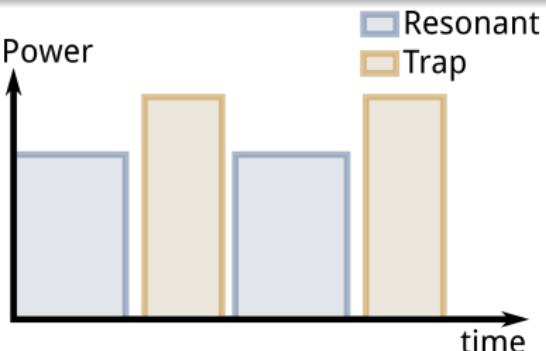
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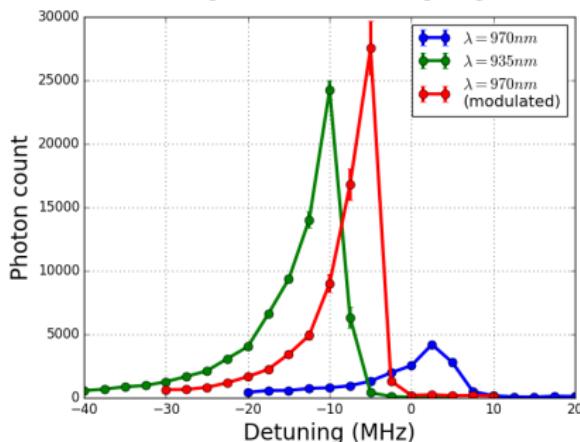
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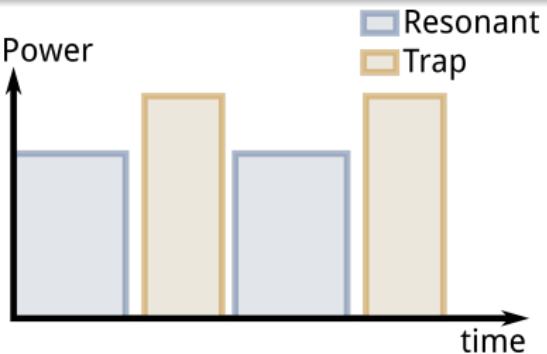
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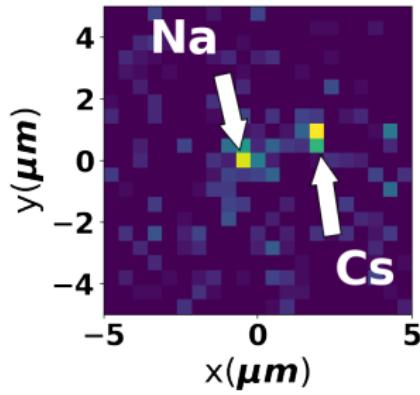
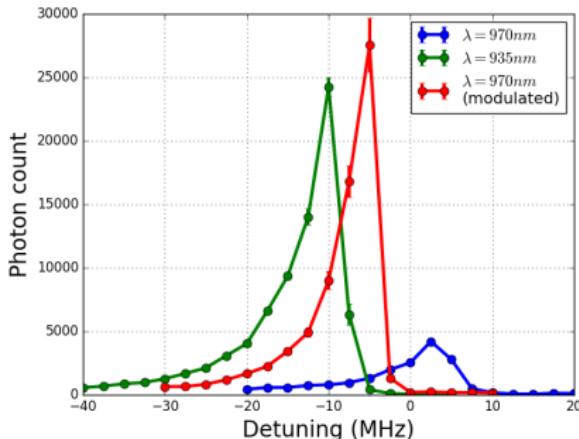
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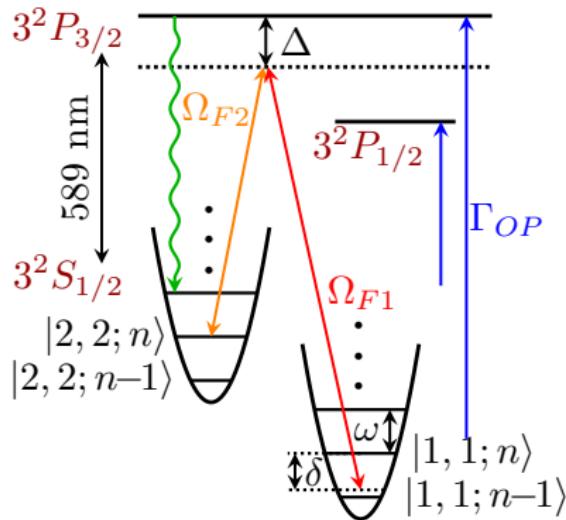
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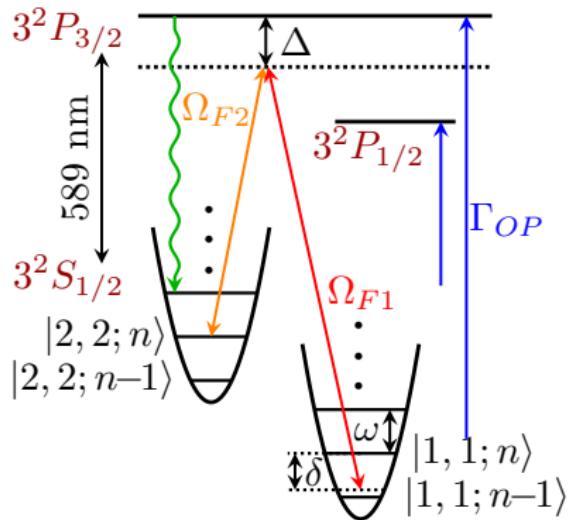
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Raman Sideband Cooling



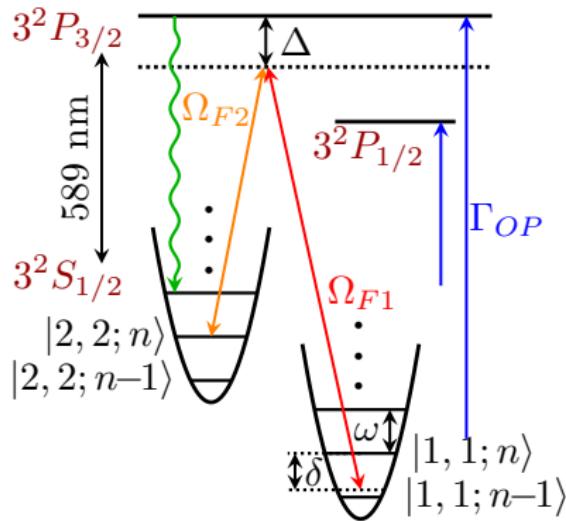
Raman Sideband Cooling



Large Lamb Dicke parameter

- Large recoil heating
- Many sideband orders
- Coupling “dead zone”

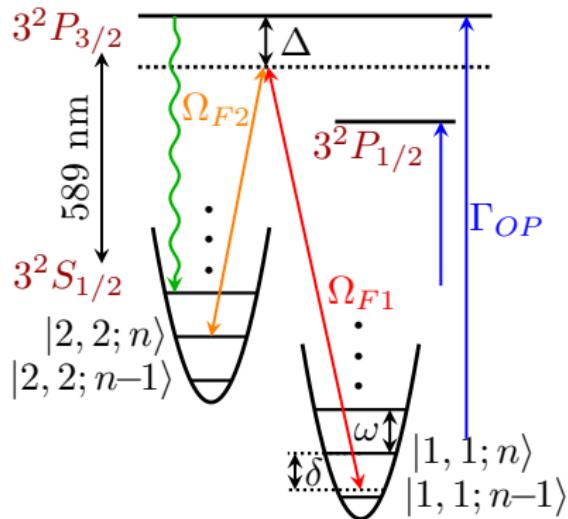
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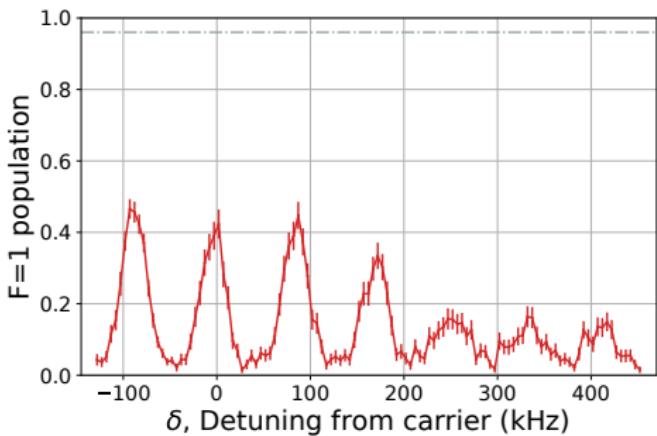
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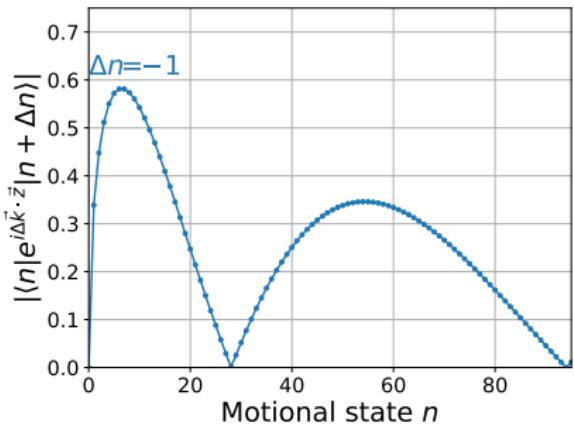
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Axial sideband spectrum

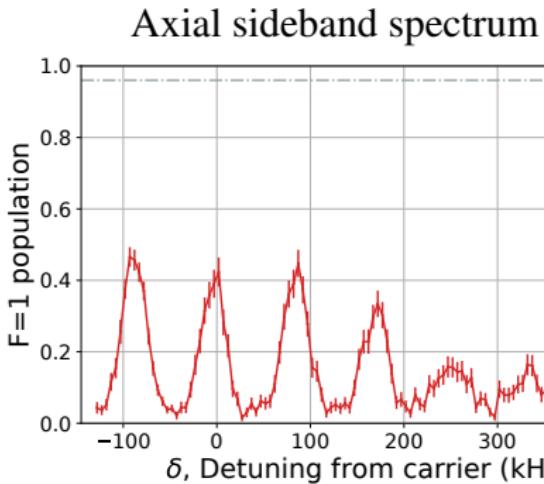


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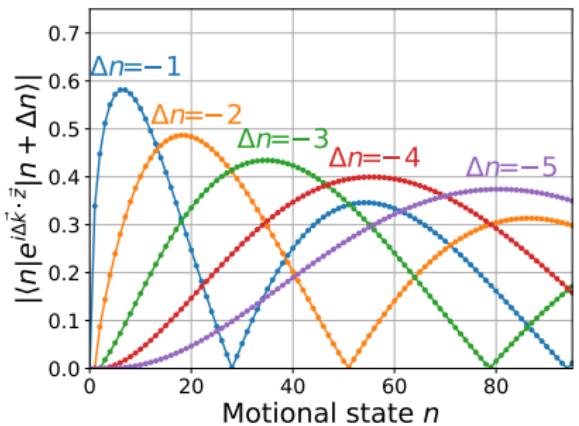


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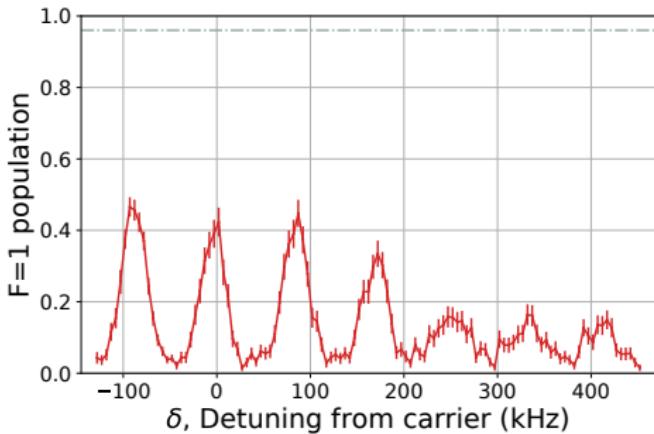
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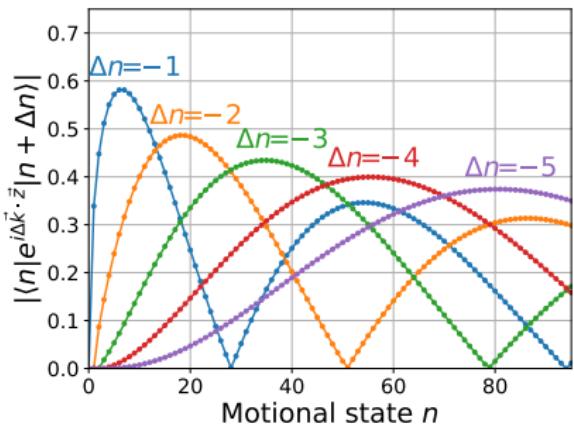
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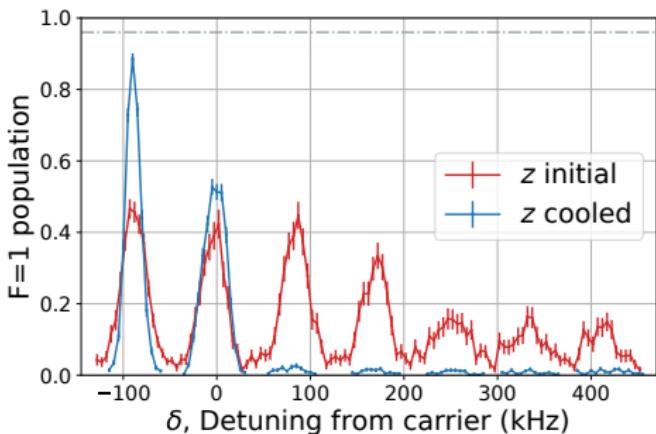


3D ground state: 93.5(7)%

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Axial sideband spectrum



Outline

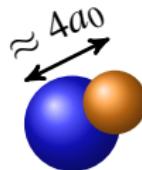
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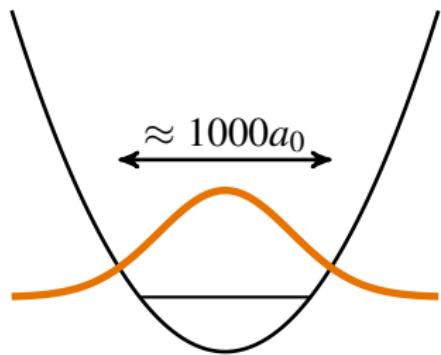
3 Atom-Atom Interaction and Molecule Formation

Optical Transfer to Molecular State

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



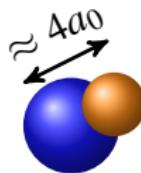
Molecule



Atoms

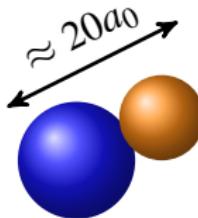
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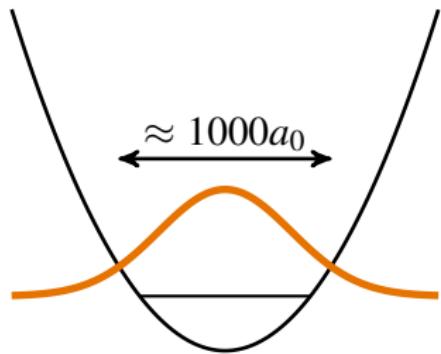


Molecule

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

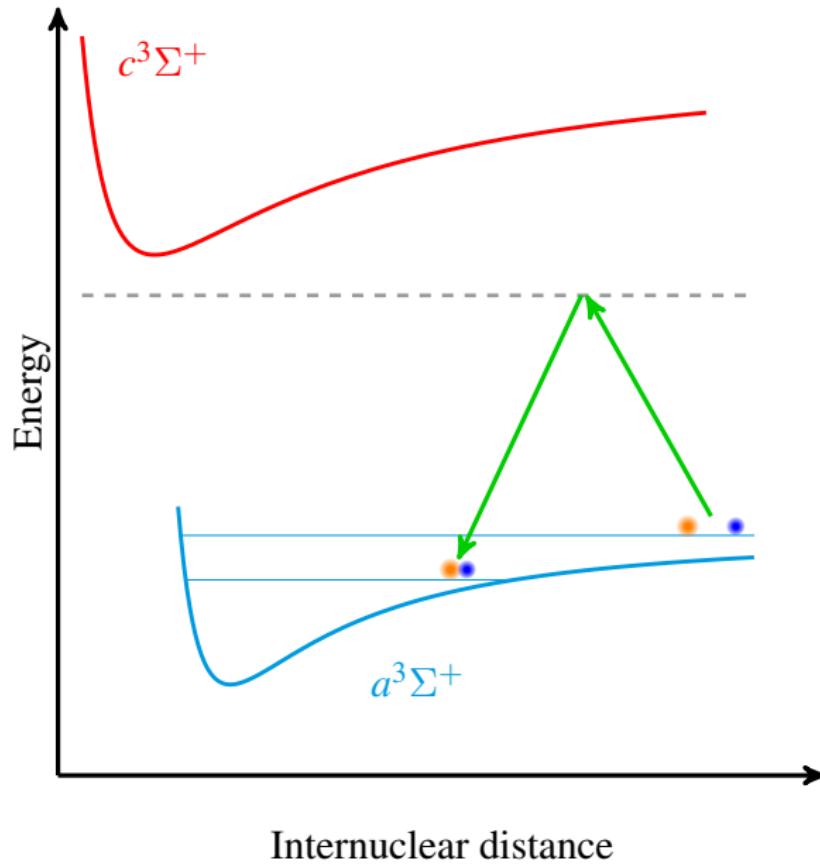


Weakly-Bound
Molecule

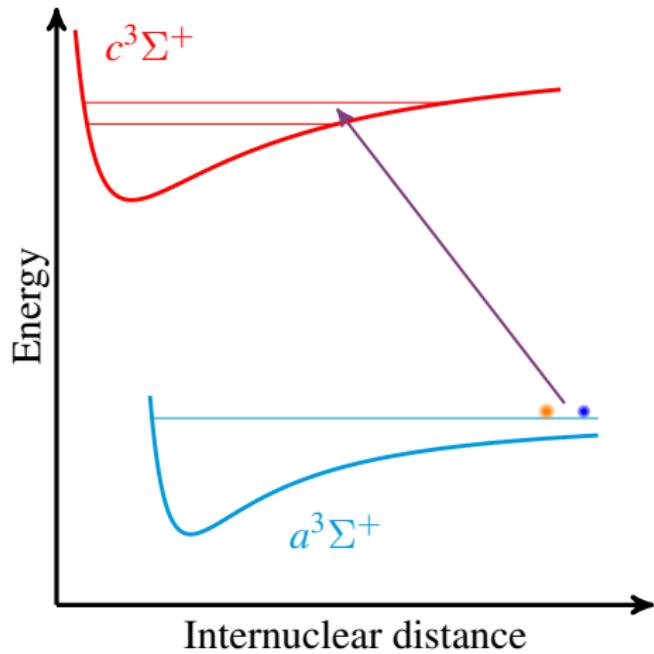


Atoms

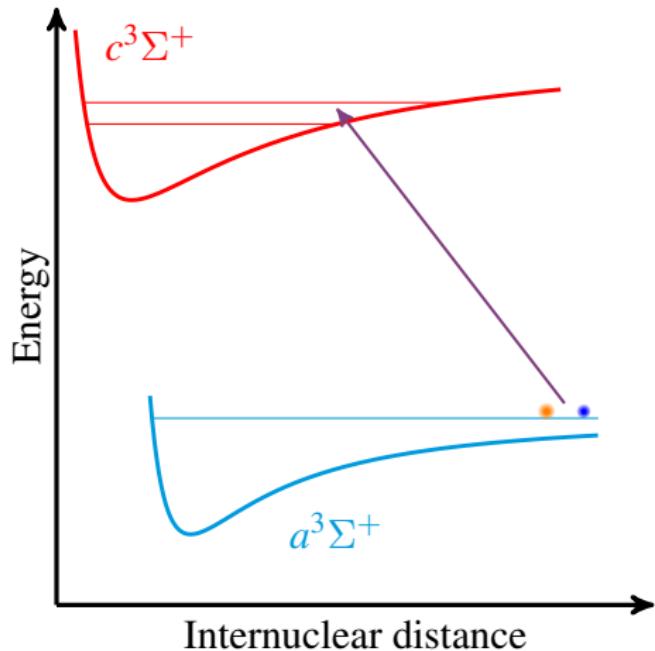
Optical Transfer to Molecular State



Photoassociation (PA) Spectroscopy



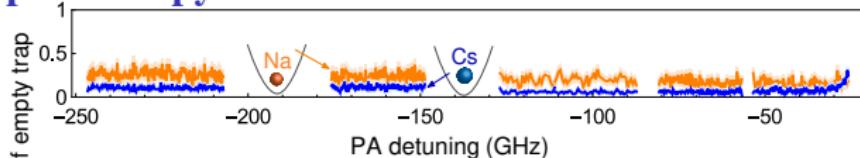
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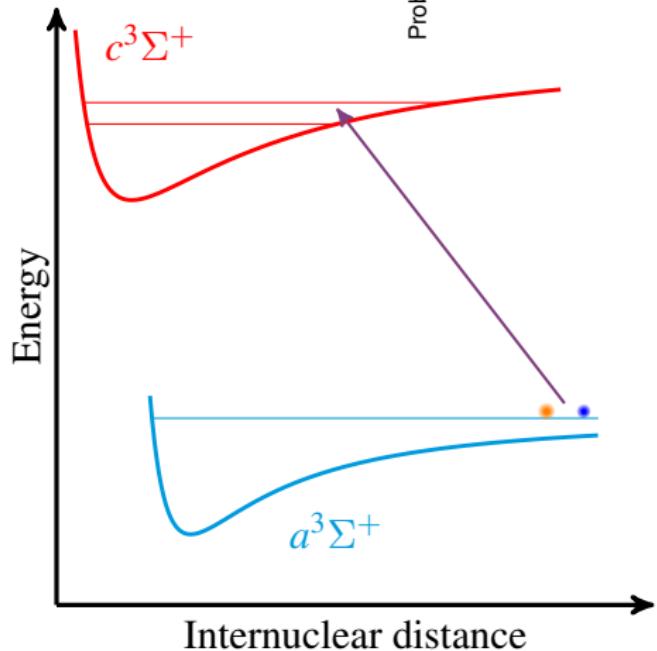
Single Atom PA

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

Photoassociation (PA) Spectroscopy



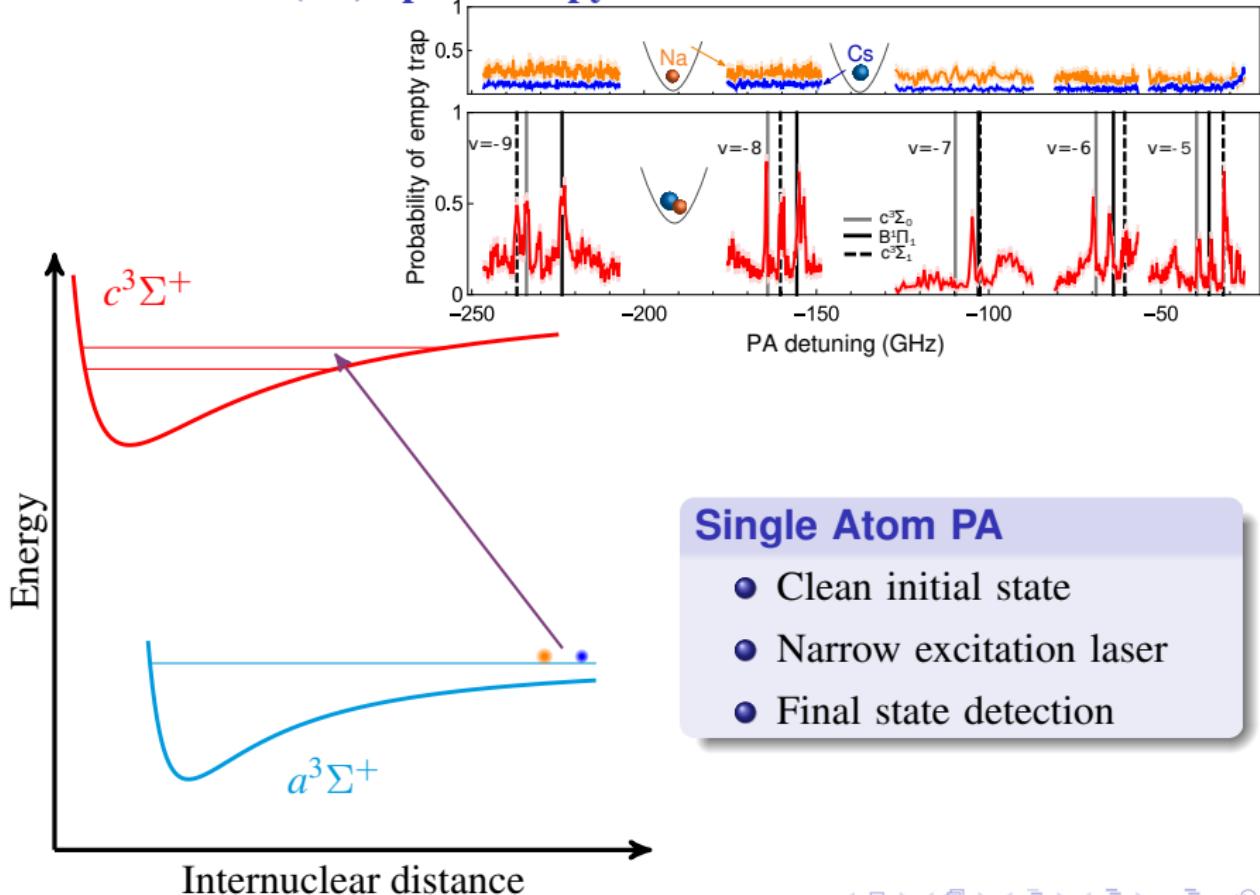
Probability of empty trap



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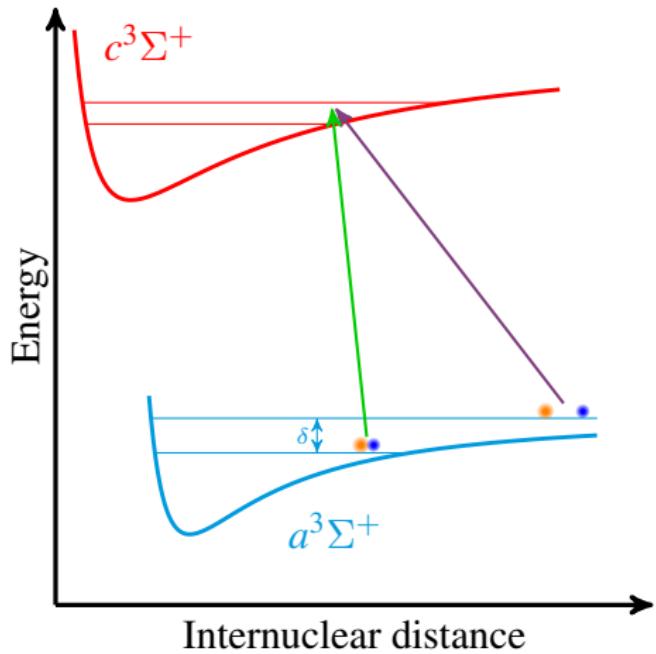
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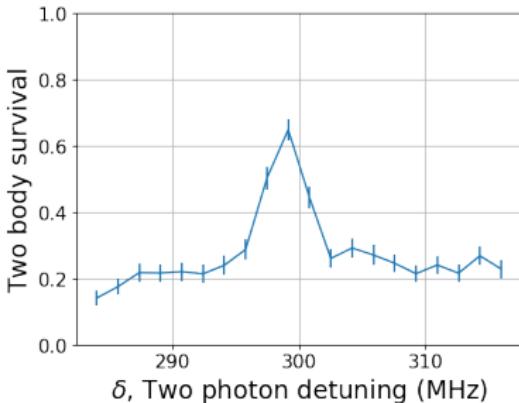
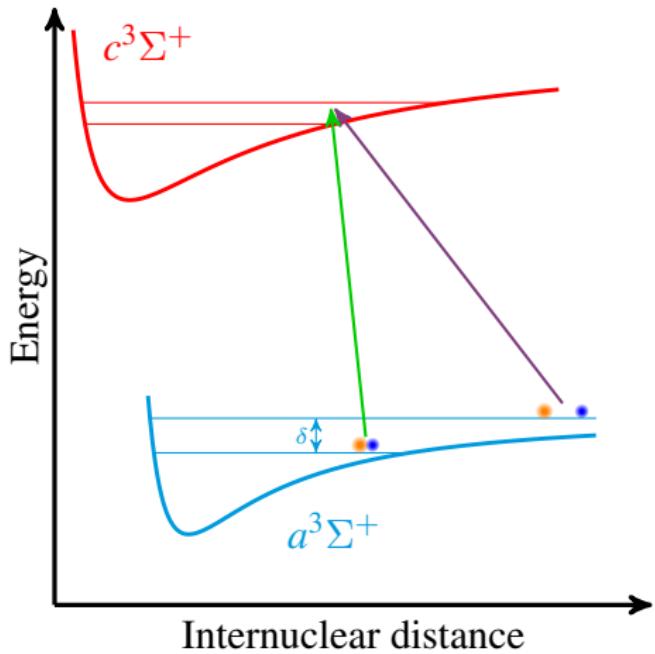
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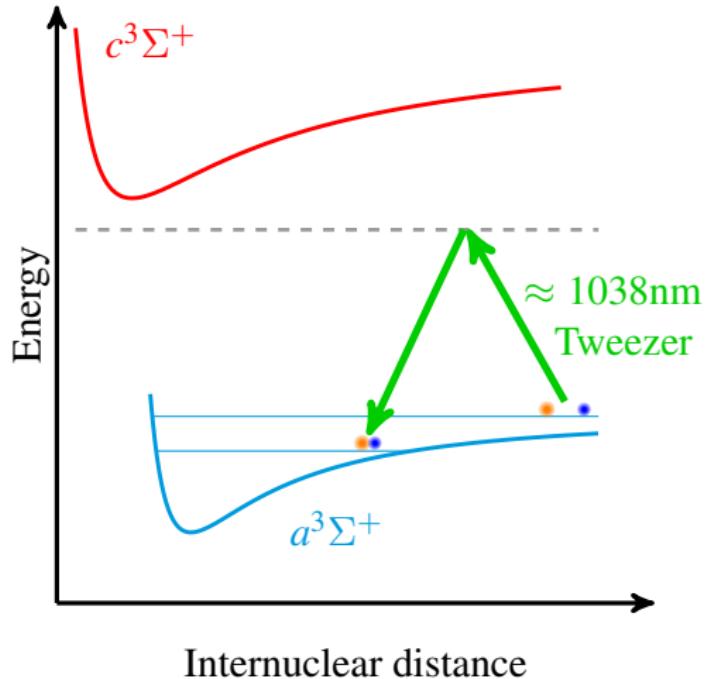
Electromagnetically Induced Transparency (EIT) Spectroscopy



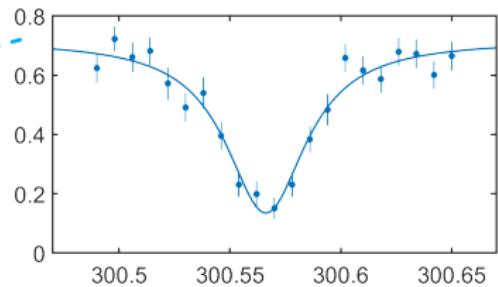
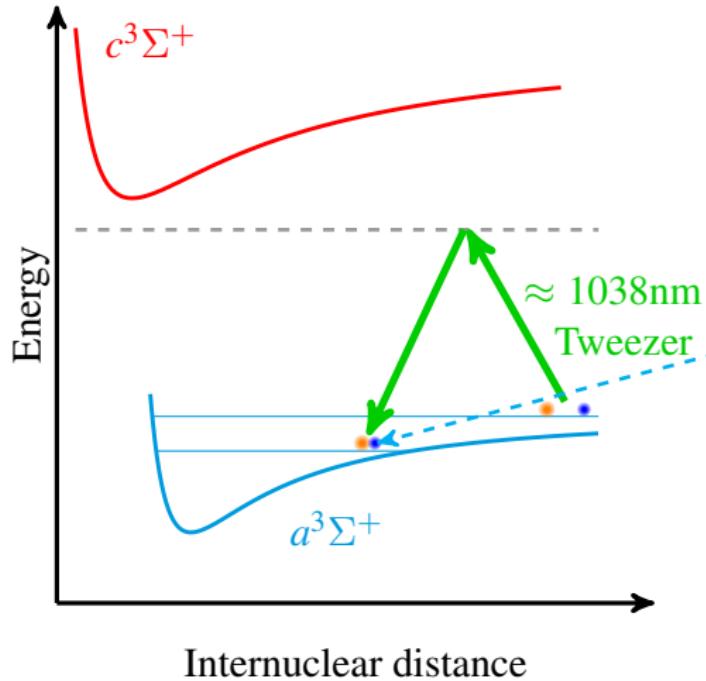
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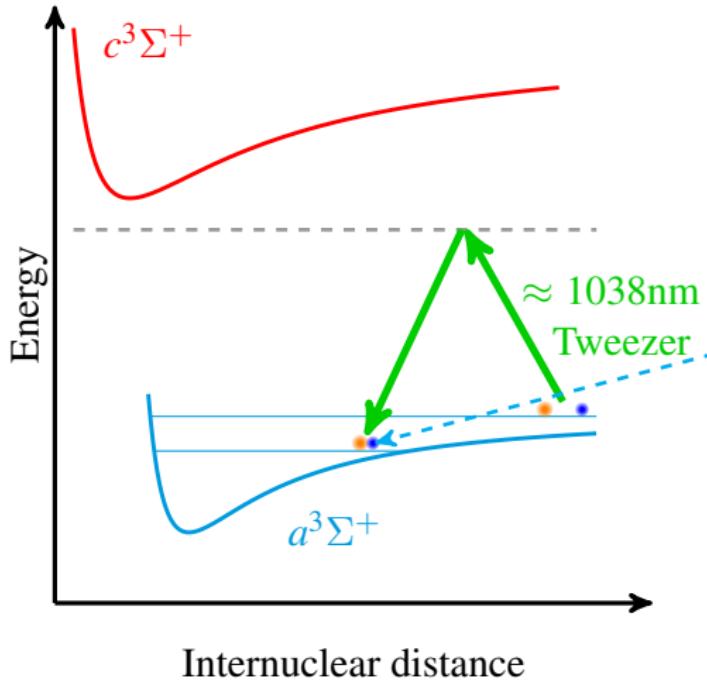
Optical Transfer to Weakly-Bound Molecular



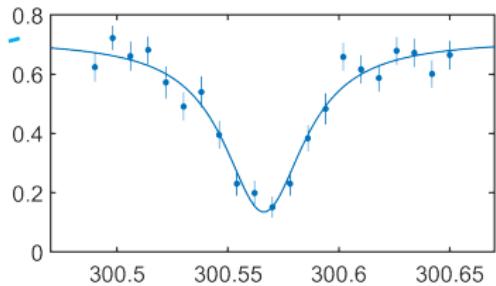
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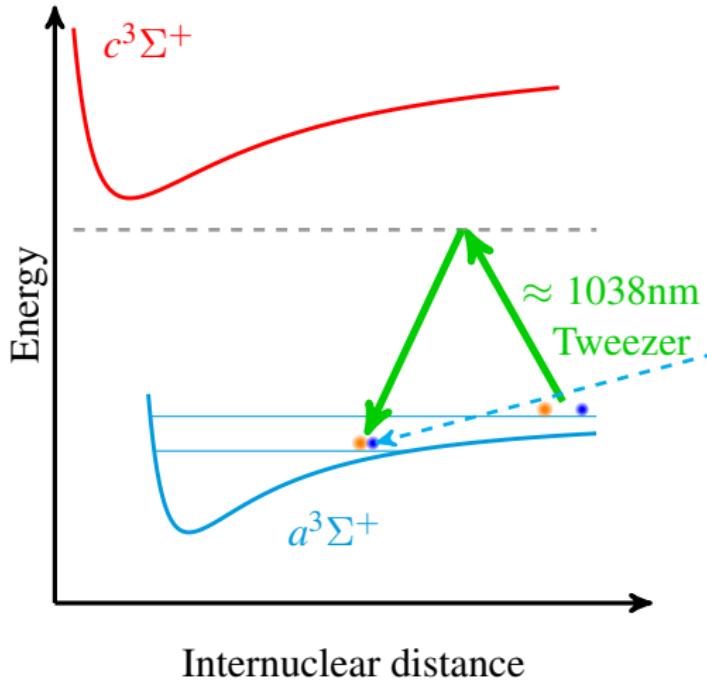
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But no Rabi oscillation

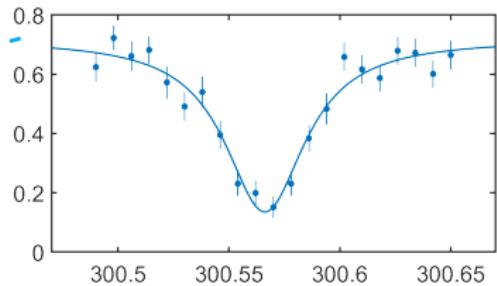


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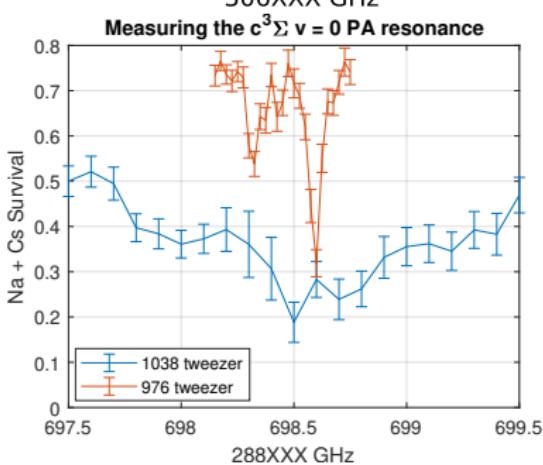
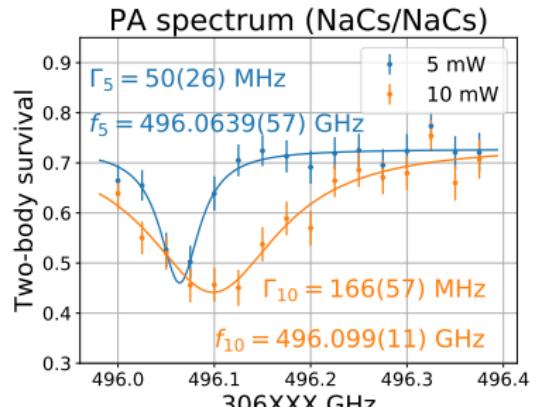


But no Rabi oscillation

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

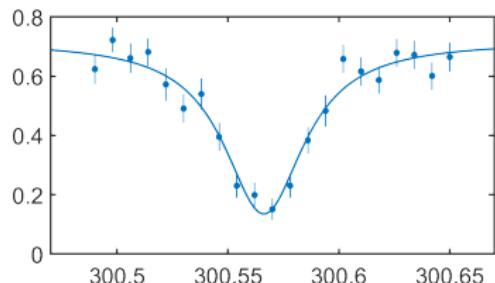


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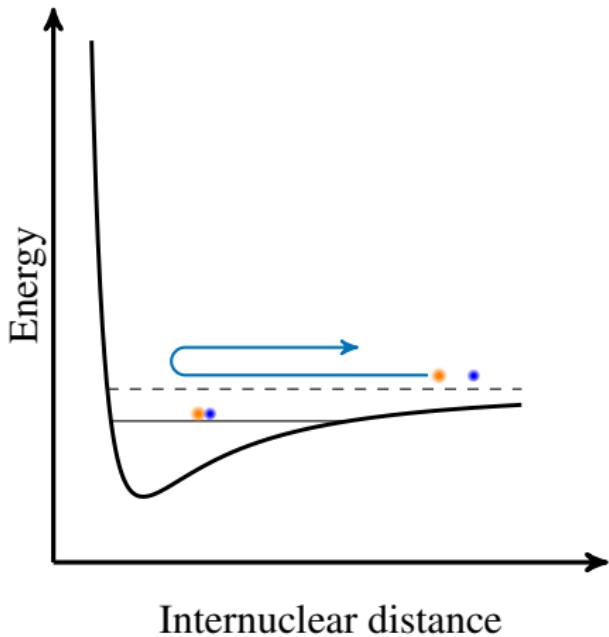
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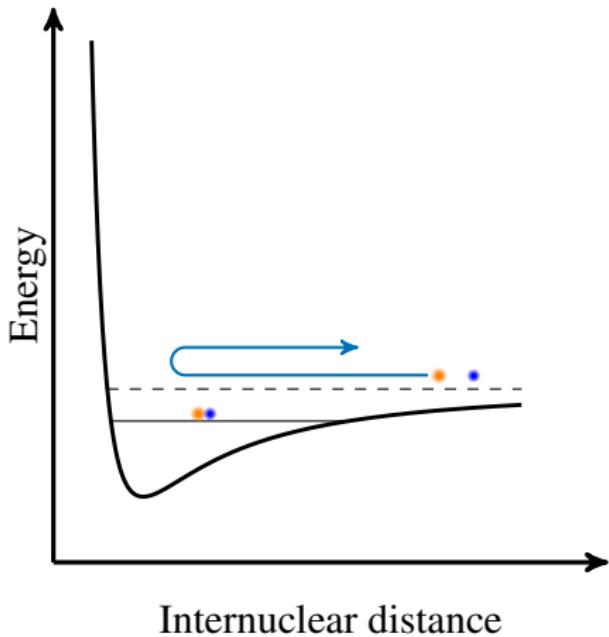
Scattering length a

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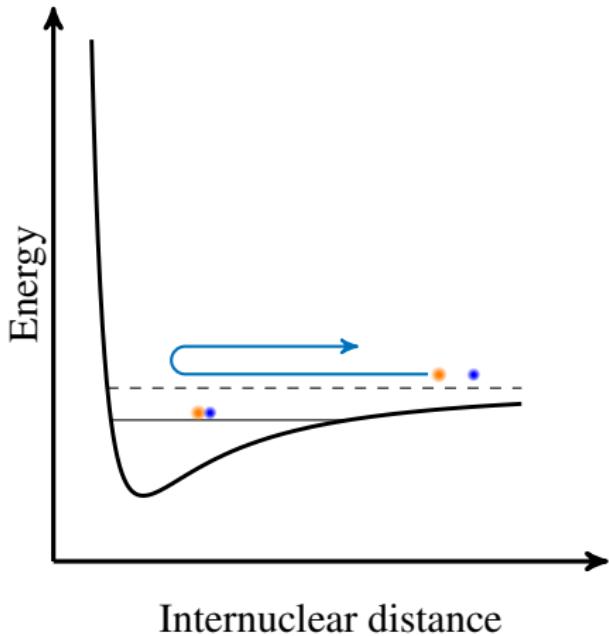
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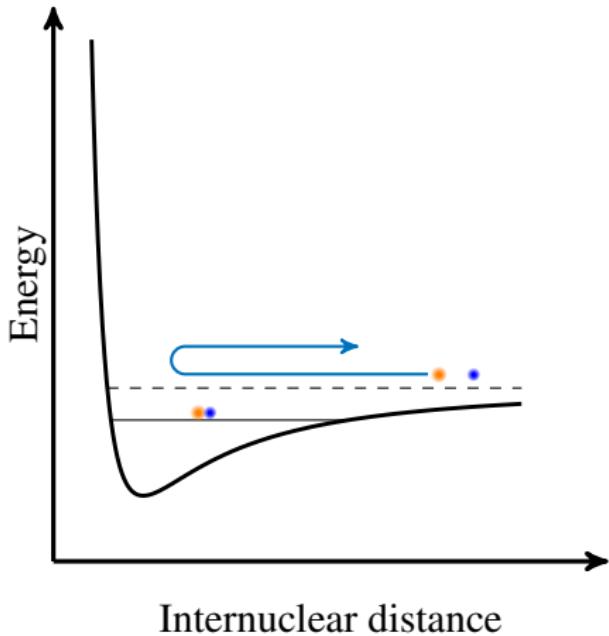
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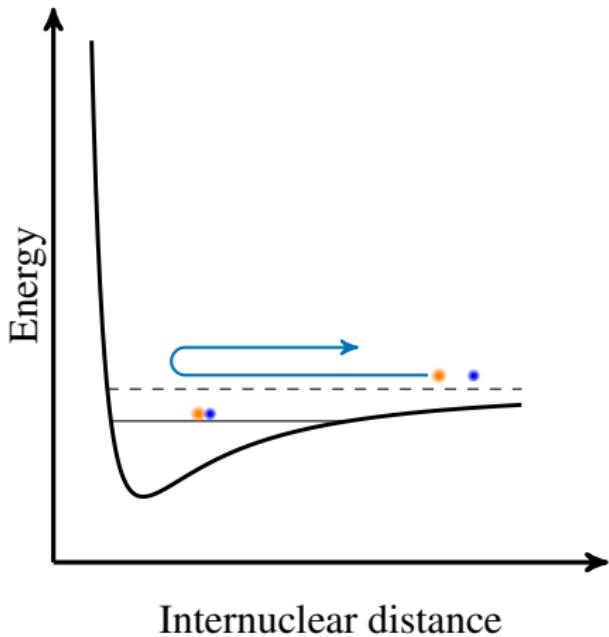
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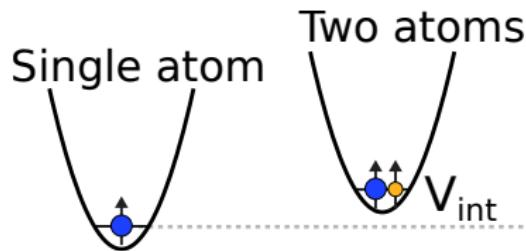


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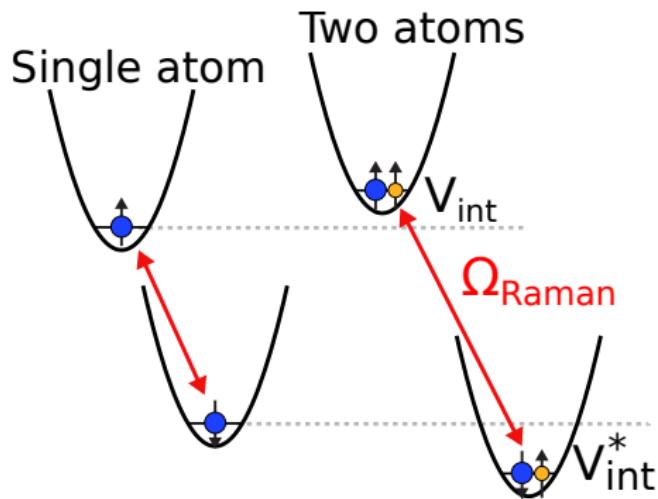
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- ⋮



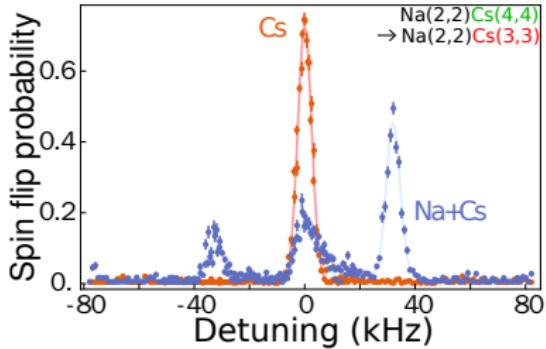
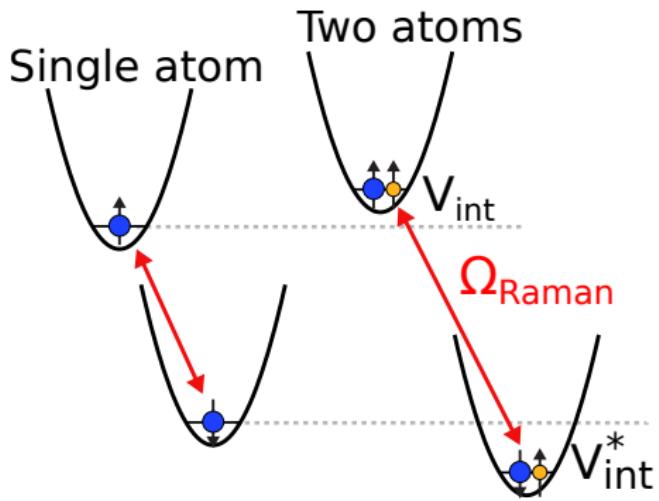
Interaction shift



Interaction shift



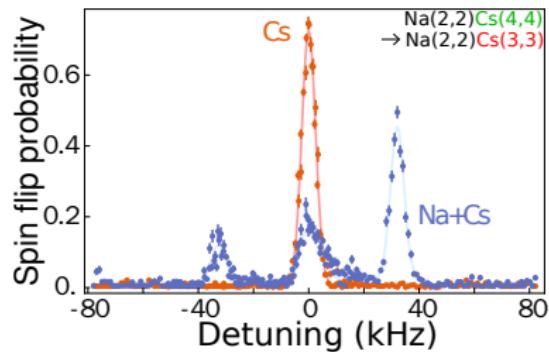
Interaction shift



Interaction shift

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

Interaction



Interaction shift

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

Interaction

To center of mass
and relative coordinates

$$M = m_1 + m_2$$

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

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

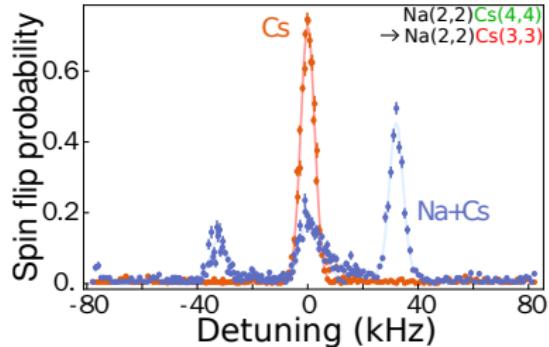
$$\omega_{R,i}^2 = \frac{m_2 \omega_{1,i}^2 + m_1 \omega_{2,i}^2}{m_1 + m_2}$$

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

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

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

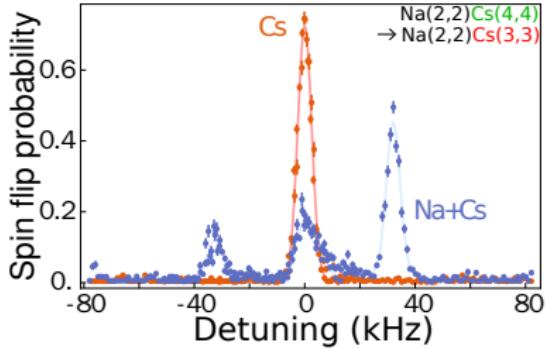
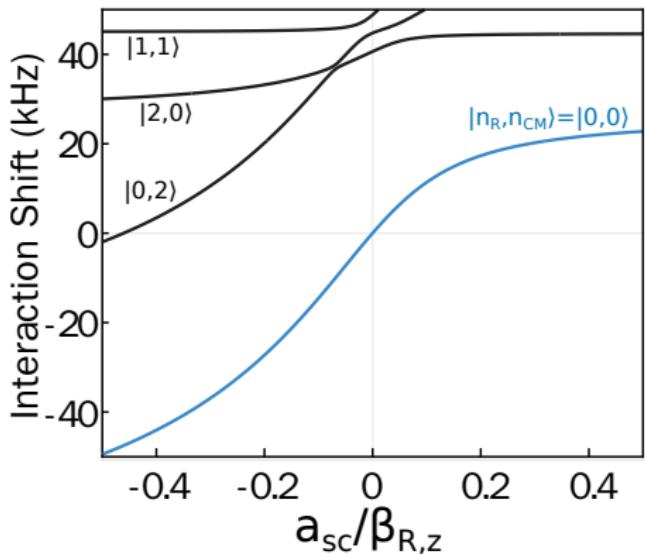
$$p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$



Center of mass

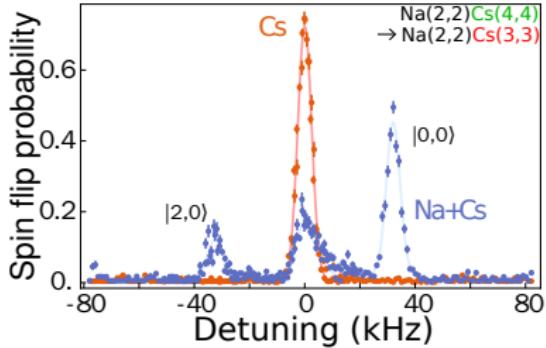
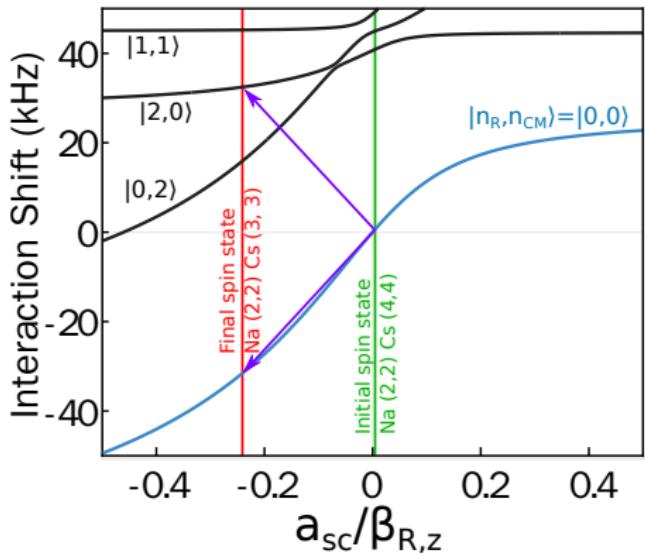
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M \Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu \omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu (\omega_{1,i}^2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

Interaction shift



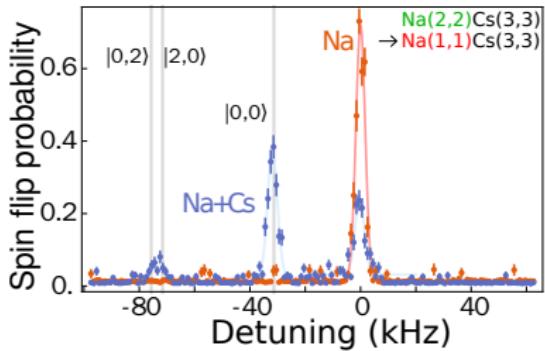
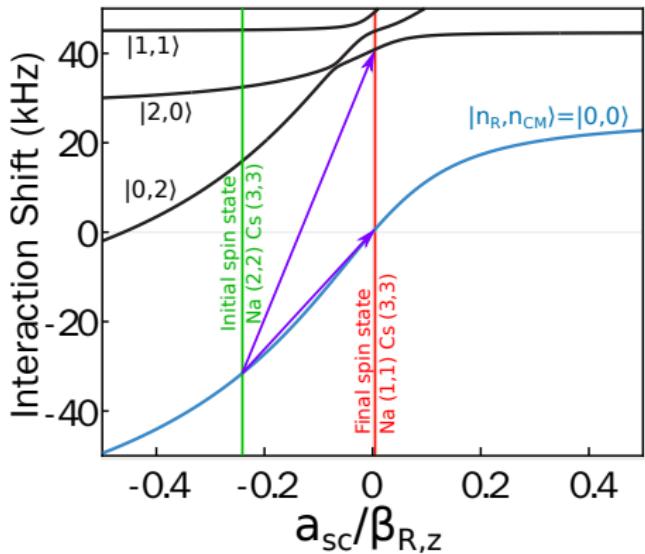
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



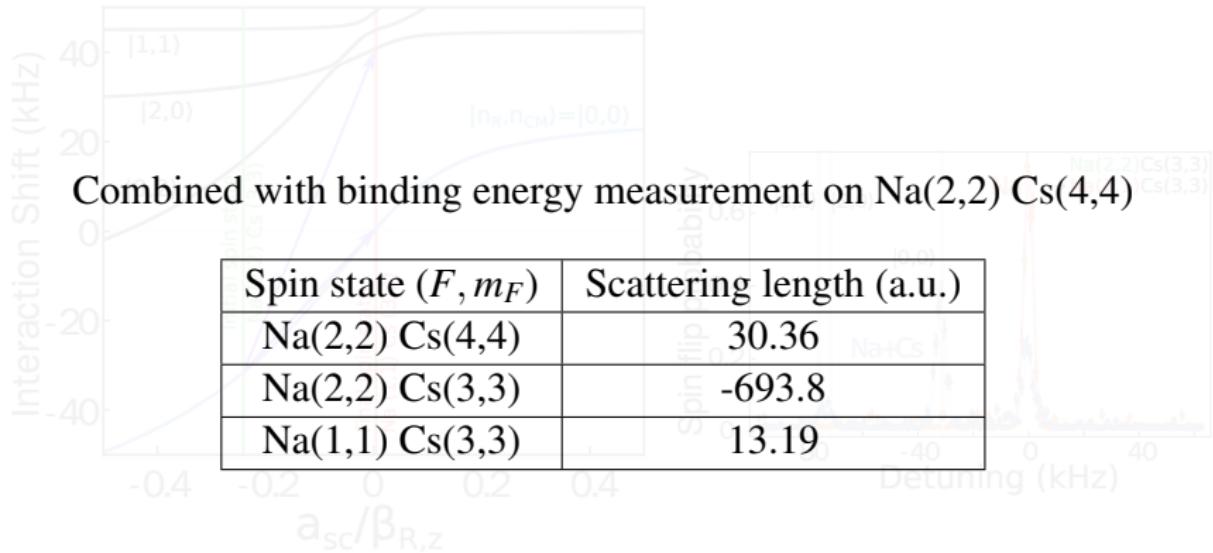
$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

Interaction shift



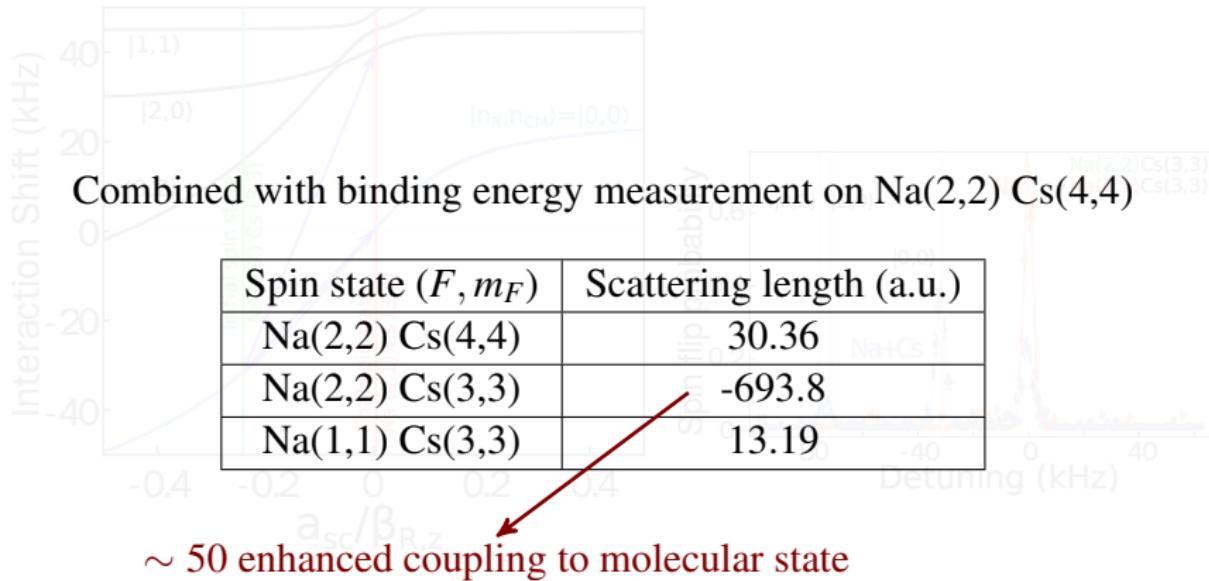
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Interaction shift



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Interaction shift

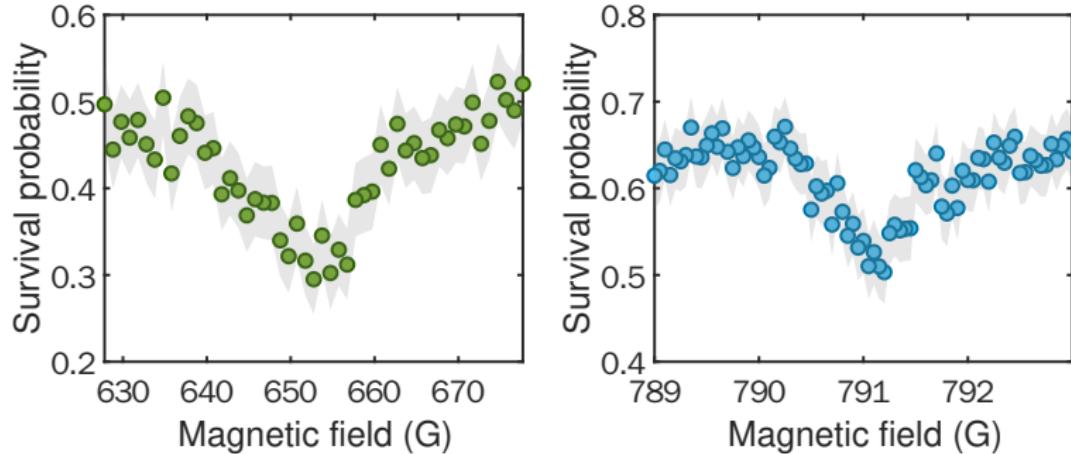


$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M} \right)}_{\text{Center of mass}} + \underbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 X_{R,i}^2}{2} + \frac{P_{R,i}^2}{2\mu} \right) + V_{int}(\vec{r}_R)}_{\text{Relative}} + \underbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2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

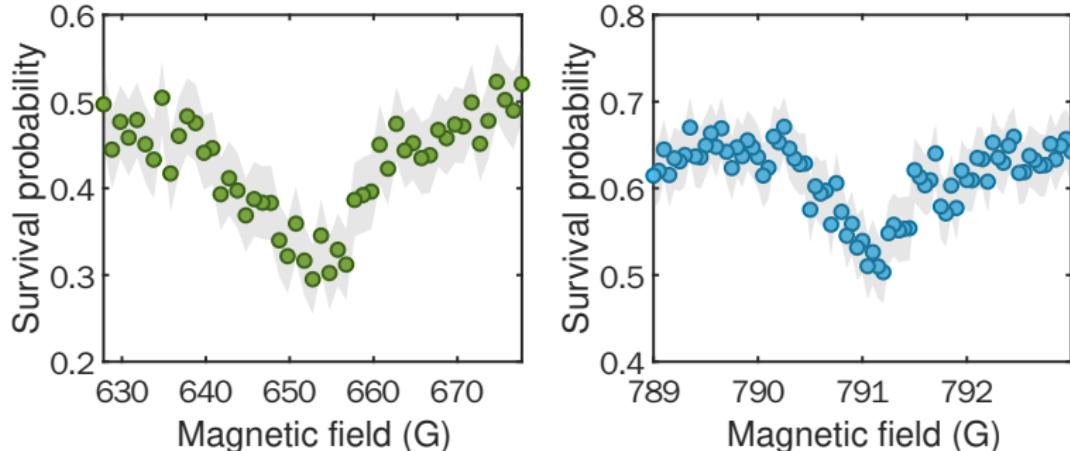
Na (1, -1) Cs (3, -3) Feshbach resonance



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) ¹	663 G	799 G
Measured	652(3) G	791.2(2) G

¹In 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.

