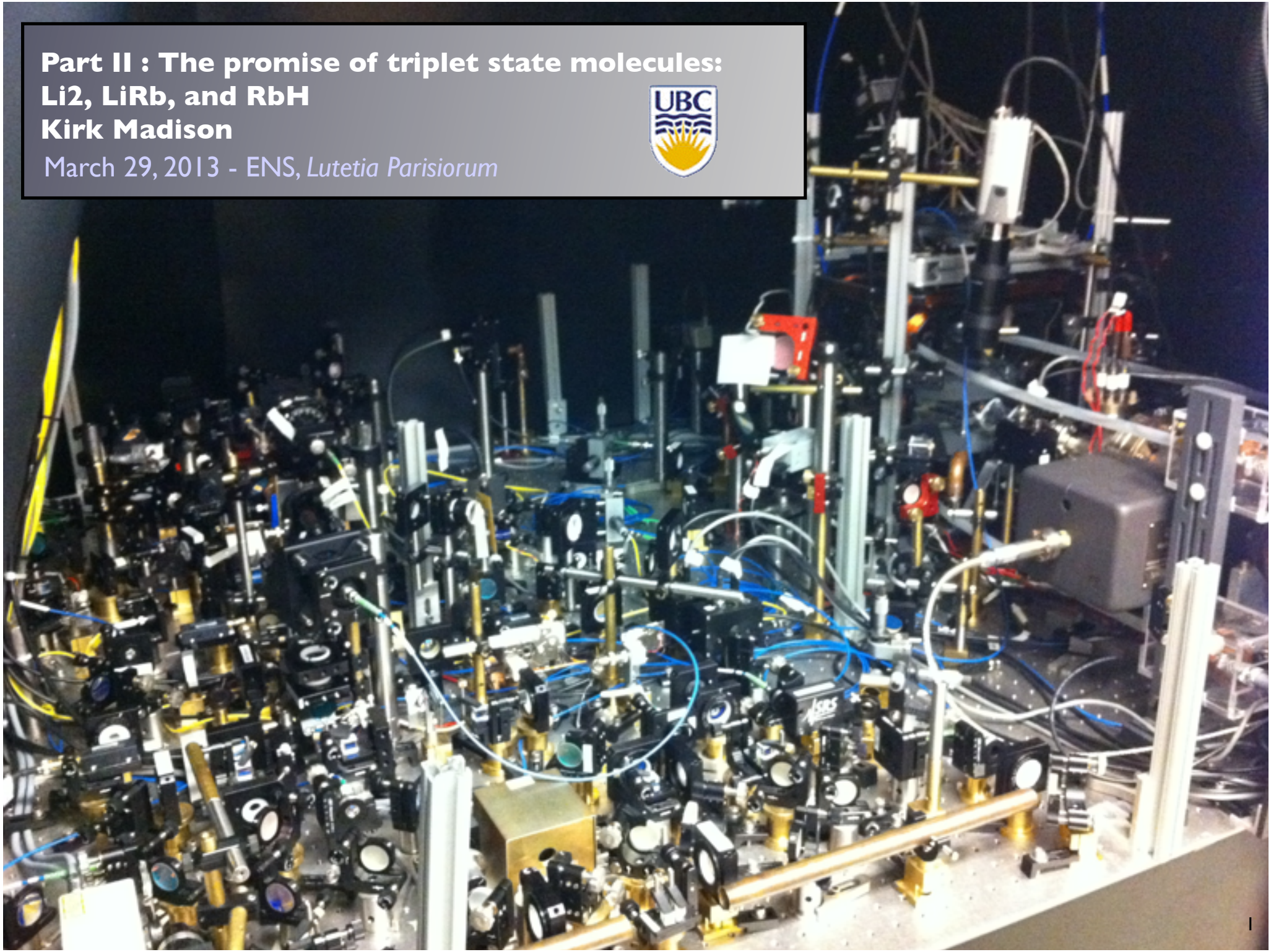


Part II : The promise of triplet state molecules: Li2, LiRb, and RbH

Kirk Madison

March 29, 2013 - ENS, *Lutetia Parisiorum*

Li+Rb mixtures

Will Gunton

Mariusz Semczuk

Theory support for Li_2 from Xuan Li[1] and Nike Dattani[2]

[1]Chemical Science Division, Lawrence Berkeley National Laboratory, Berkeley, USA
[2]Department of Chemistry, University of Oxford, Oxford, UK

CRUCS Canadian Centre for Research on Ultra-Cold Systems

NSERC CRSNG

UBC British Columbia

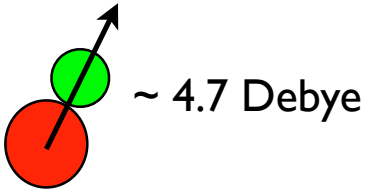
CFI

Peter Wall Institute for Advanced Studies

CIFAR CANADIAN INSTITUTE for ADVANCED RESEARCH

Long term goal: production of dense, ultracold ensembles of polar molecules

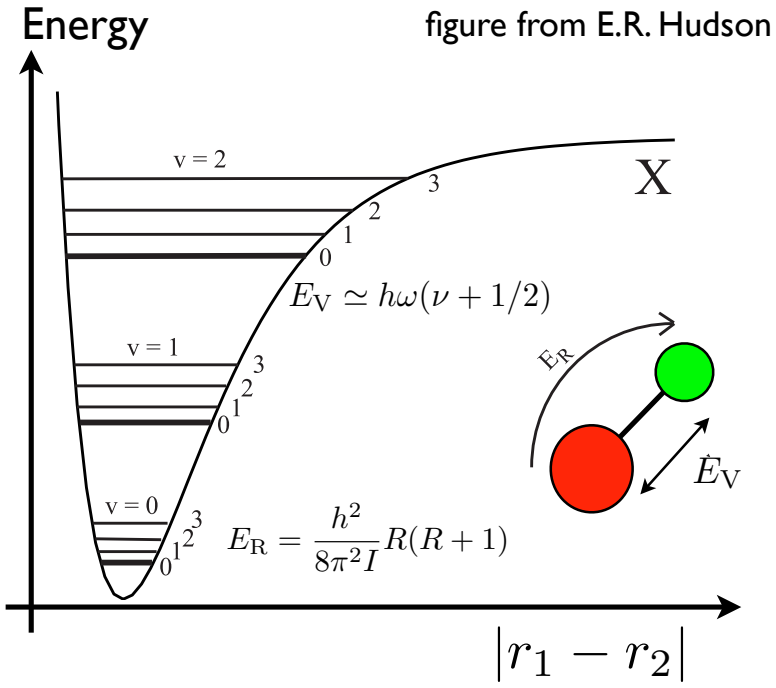
RbLi is a polar molecule



Polar molecules: scientific motivation

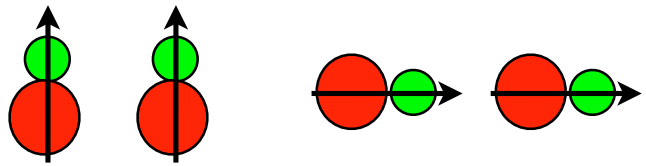
Additional **features** not available with atoms:

Rich internal structure:
- *rotational, vibrational*



Dipolar interactions:
- *large and long range* $\sim 1/r^3$
- *angular dependence (anisotropic)*

$$U_{\mathrm{dd}}^{\mathrm{electric}} = \frac{1}{4\pi\epsilon_0} \frac{\mathbf{d_1} \cdot \mathbf{d_2} - 3(\mathbf{d_1} \cdot \hat{r})(\mathbf{d_2} \cdot \hat{r})}{r^3}$$



... and zero at the 'magic angle'

Polar molecules: scientific motivation

Dipolar interactions - *how strong?*

* fully polarized

Inter-particle interactions between RbLi dimers (4.7 Debye)*

~1000x larger than mean field interaction in a BEC

~10,000x larger than magnetic dipolar interactions in Cr

Dipolar interactions - *at what range?*

532 nm optical lattice

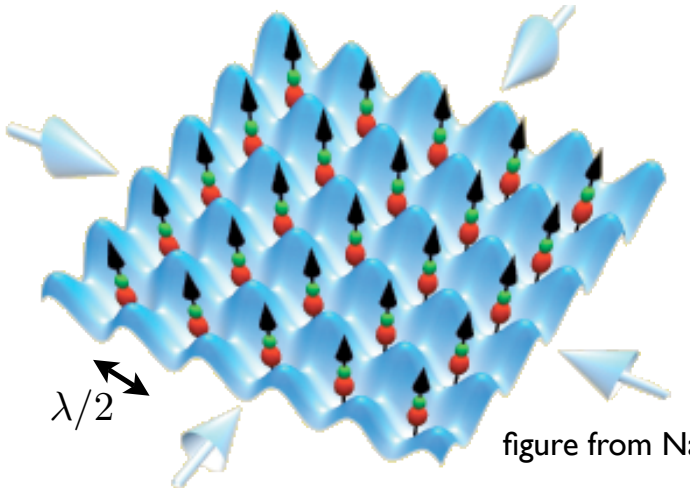
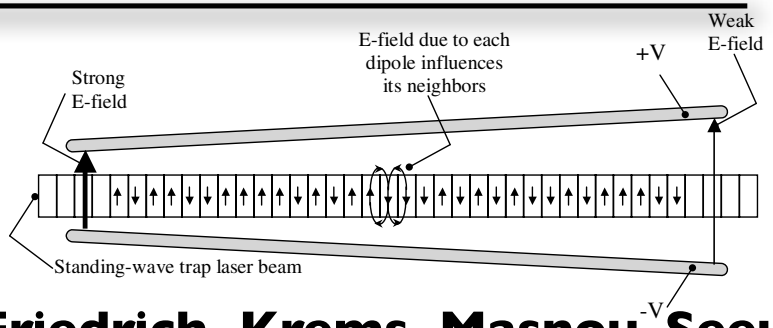


figure from Nature **2**, 341 (2006)

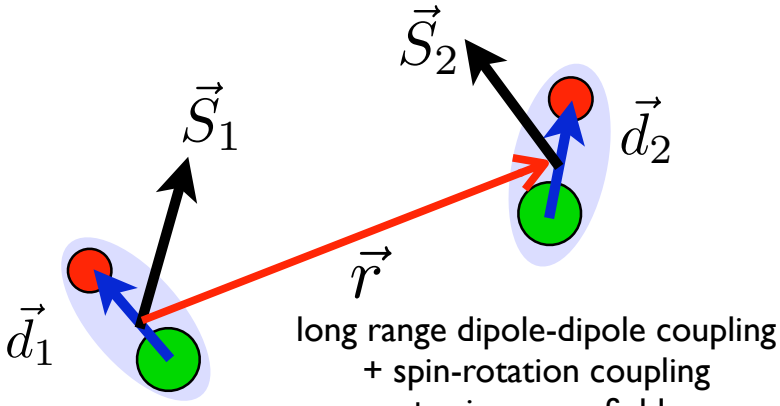
$$U_{\mathrm{dd}} \sim \left.\frac{(4.7\ \mathrm{Debye})^2}{4\pi\epsilon_0 r^3}\right|_{r=0.266\mu\mathrm{m}} = k_{\mathrm{B}} \times 8.5\ \mu\mathrm{K} = h \times 177\ \mathrm{kHz}$$

Polar molecules: scientific motivation

few and many body QM

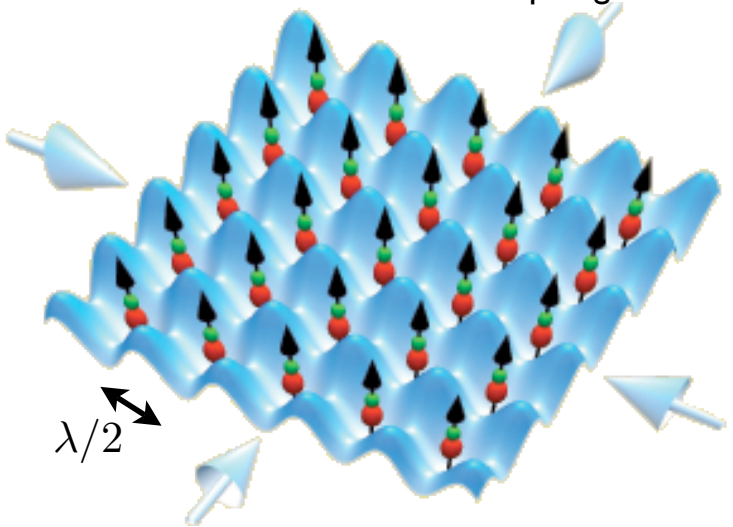


- **2002, Quantum logic gates [DeMille]**
- **2004, Quo vadis, cold molecules? [Doyle, Friedrich, Krems, Masnou-Seeuws]**
- **2005, Ultracold chemistry - exotic few body QM [Krems]**
  "Inelastic collisions and chemical reactions of cold molecules in external fields"
- **2006 Exotic many body quantum mechanics [Micheli, Brennen, Zoller]**
  with polar molecules in an optical lattice dressed with a microwave field, you can realize just about any spin lattice model : 1D xyz, 2D Ising, 3D Heisenberg, Kitaev model, etc...*



long range dipole-dipole coupling
+ spin-rotation coupling
+ microwave field
= effective spin-spin coupling

* topological order



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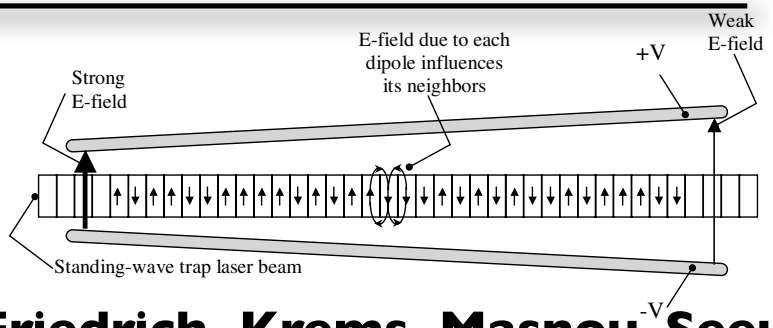

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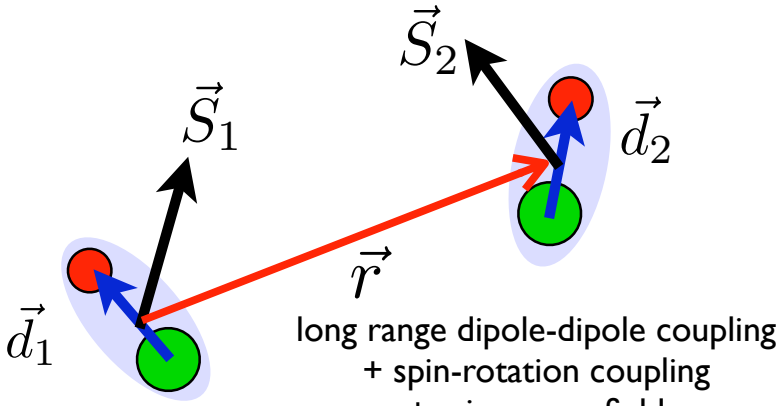

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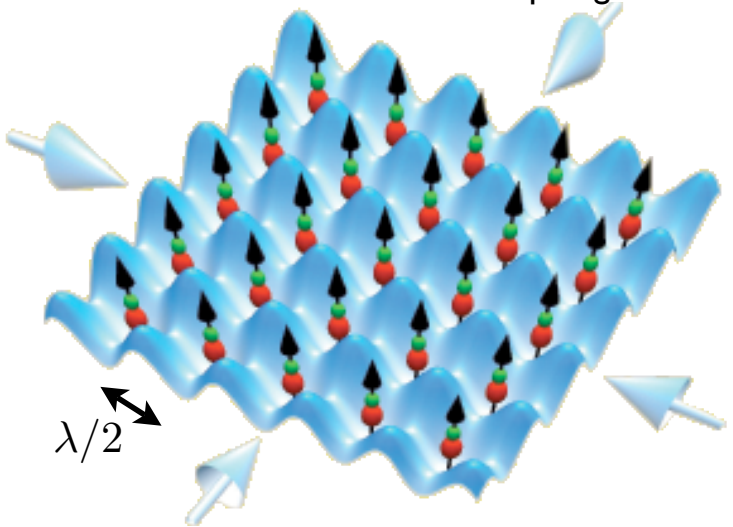

for this, you need paramagnetic, polar molecules !

Quantum degenerate polar molecules from cold atoms: state of the art

***2004 Proposal**: Efficient conversion of ultracold Feshbach-resonance-related polar molecules into ultracold ground state molecules [**Stwalley**],*
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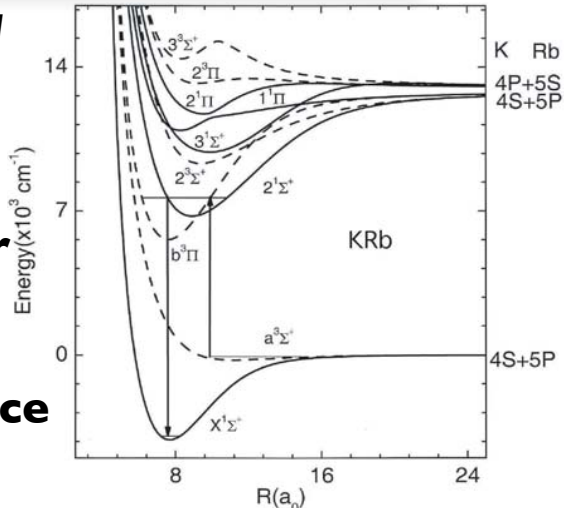

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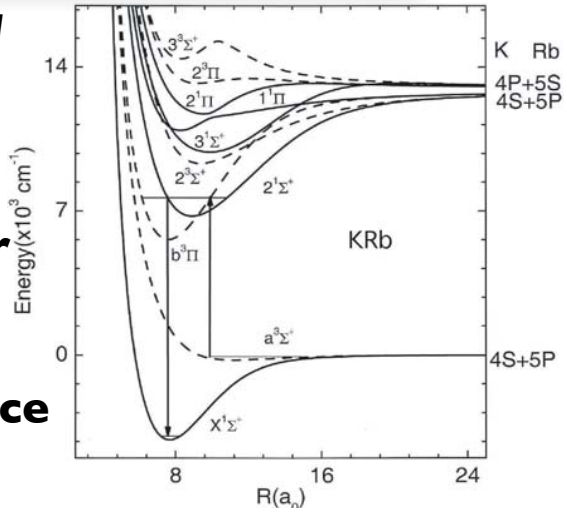

Limitations of KRb ?

- **chemically reactive** KRb + KRb -> K_2 + Rb_2

  solution: other alkali mixtures are chemically stable
  NaK, NaRb, NaCs, KCs, RbCs (endoergic - stable)
  LiNa, LiK, LiRb, LiCs and KRb (exoergic - unstable)

- **Zero spin**

  solution : ultra-cold paramagnetic polar molecules made from alkaline-earth or rare earth + alkali atoms : SrLi, YbLi

Dipole moments (Debye)

X \ Y	Na	K	Rb	Cs
Li	0.53 (0.45[a])	3.50 (3.41[b])	4.13 (4.01[c])	5.48
Na		2.75 (2.73[d])	3.33 (3.05[c])	4.60 (4.57[e])
K			0.64	1.92
Rb				1.26

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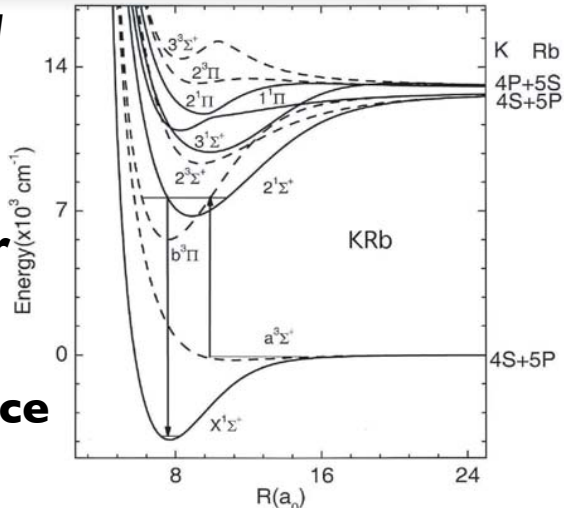

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what about the triplet state of a bi-alkali molecule?

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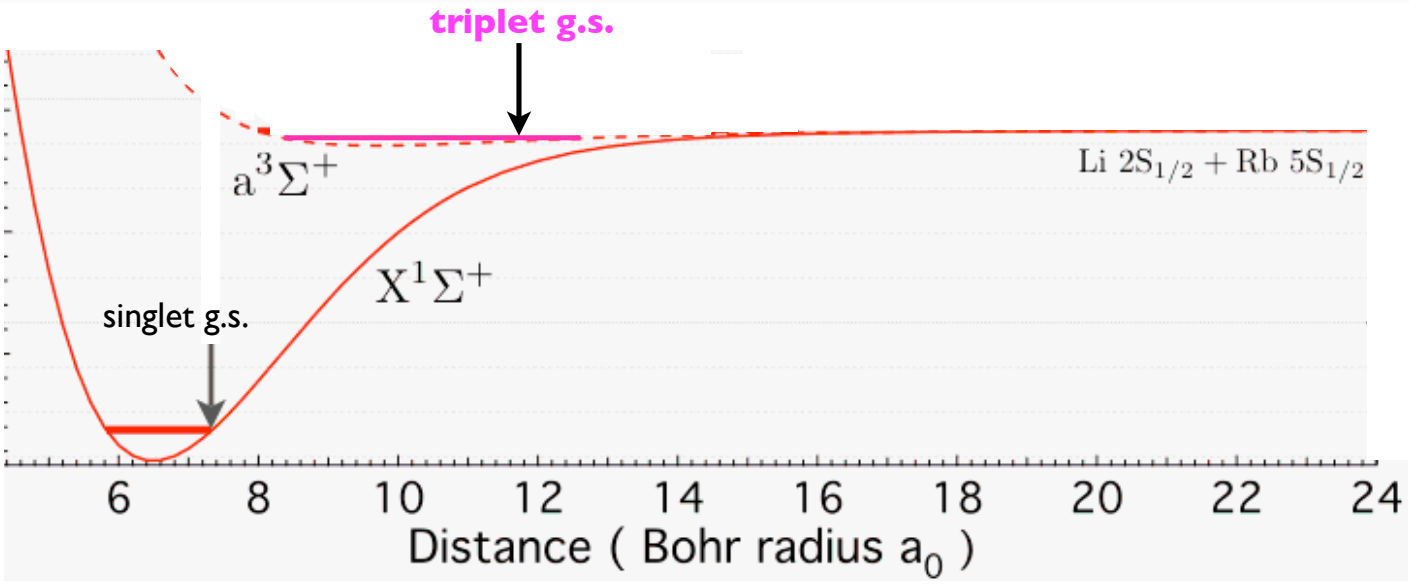

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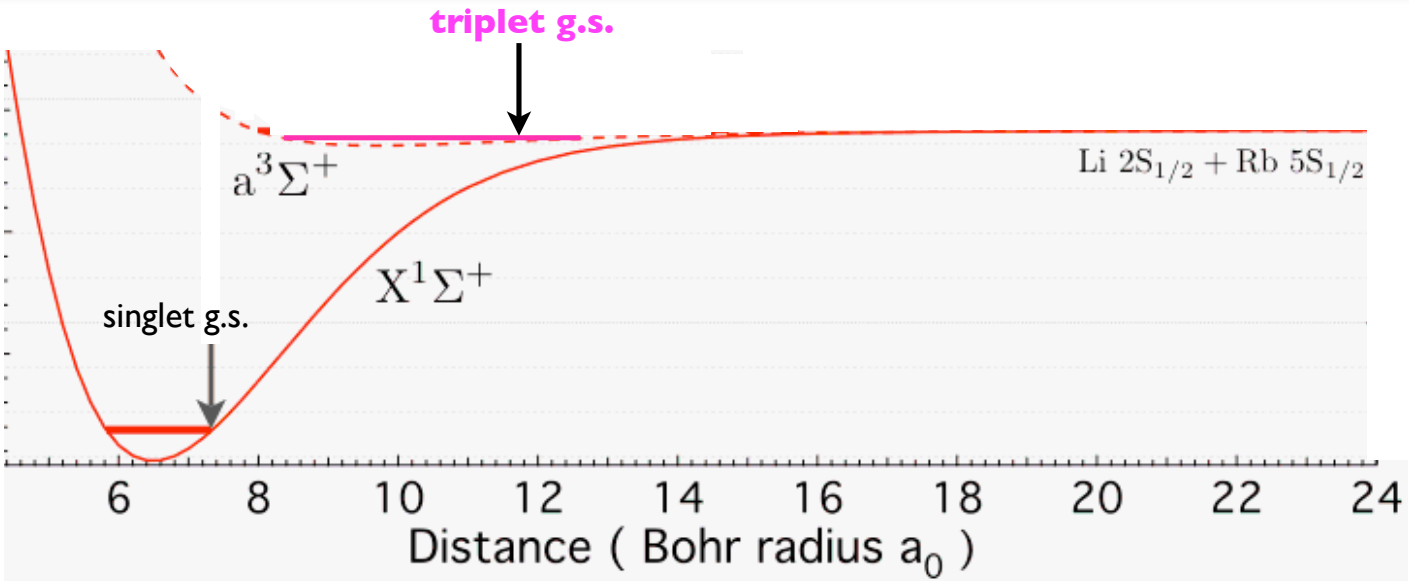

✓ **paramagnetic**

magnetically tunable collisions (FRs)

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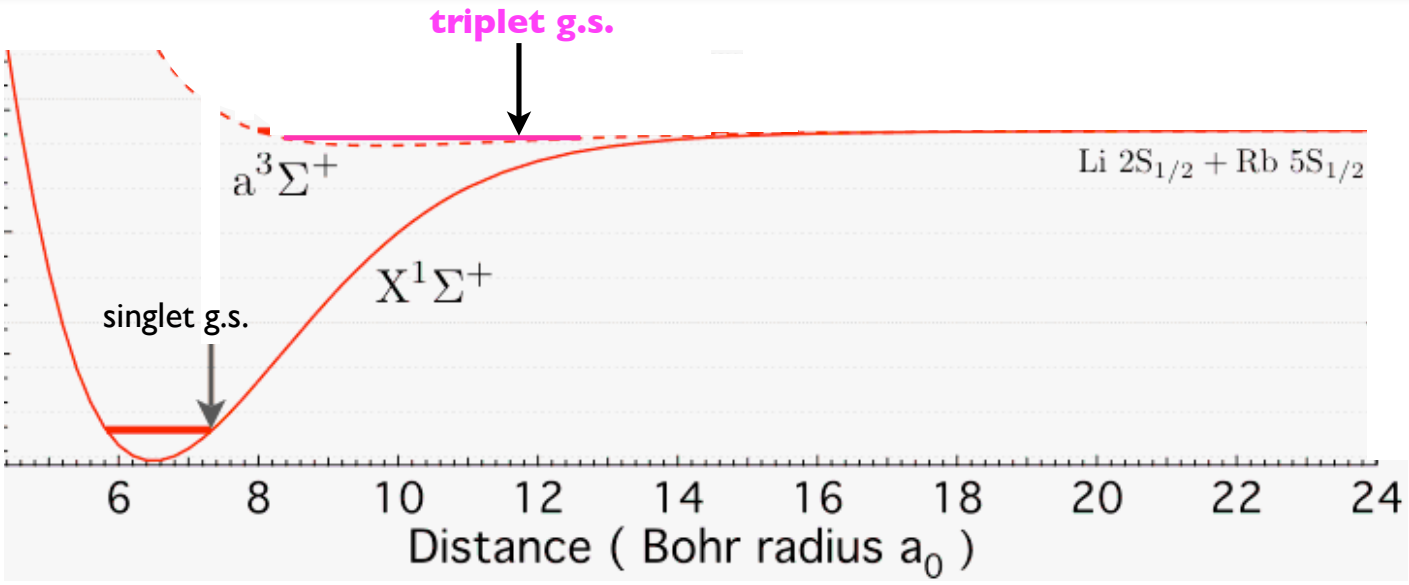

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✓ **somewhat polar (triplet DM ~1/10 that of singlet)**

DM of triplet state LiRb is about 0.4 Debye
(DM of singlet state KRb is about 0.5 Debye)

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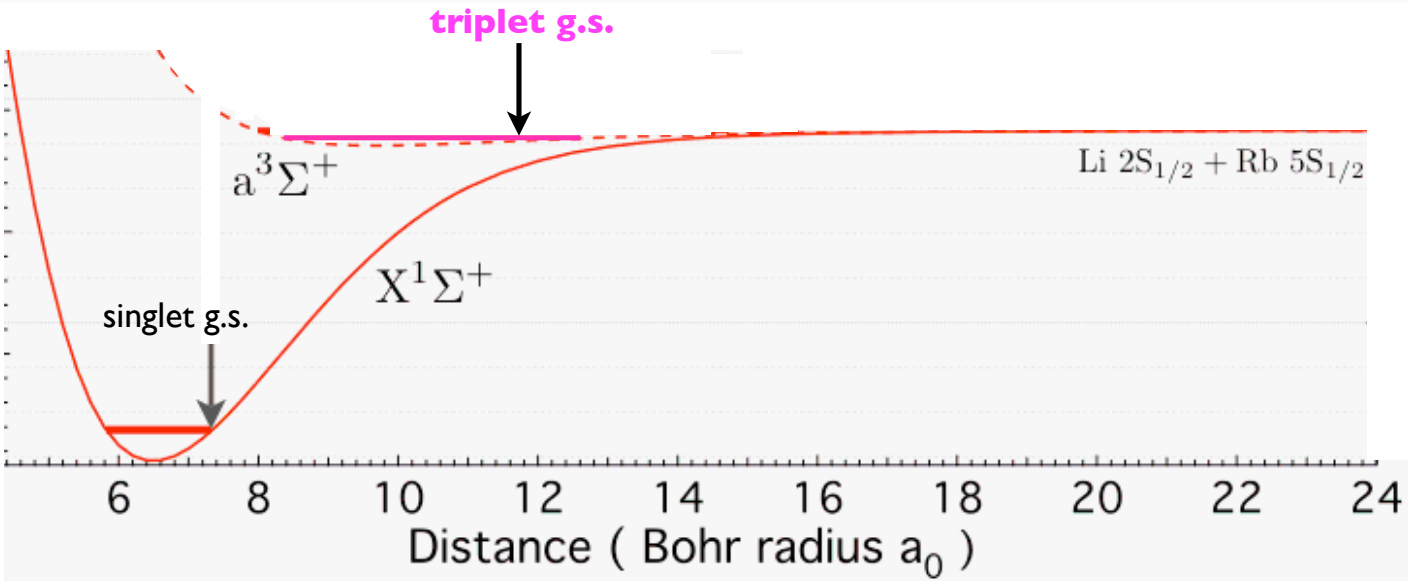

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??? Is it stable ???

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Is the lowest triplet state stable?

- **spin relaxation collisions (triplet to singlet coupling) ?**

- **chemical reactivity ?**

by some magic*, is the triplet state stable?

** or suitably physical reasons*

Is the lowest triplet state stable?

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  "Spin-orbit couplings are small in light systems. For Li_2, one wouldn't expect fast relaxation. ... but will be more important in heavy systems (like LiRb), but how important?"

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Cold Controlled Chemistry, PCCP **10**, 4079 (2008). [Krems]

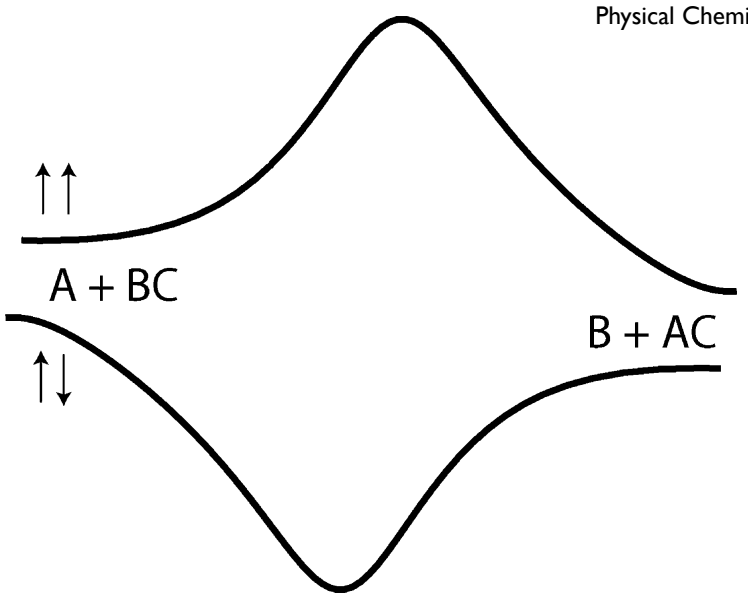

Fig. 4 Schematic illustration of minimum energy profiles for an $A(^2S) + BC(^2\Sigma)$ chemical reaction in the singlet-spin (lower curve) and triplet-spin (upper curve) electronic states. Electric fields may induce non-adiabatic transitions between the different spin states and modify the reaction mechanism.

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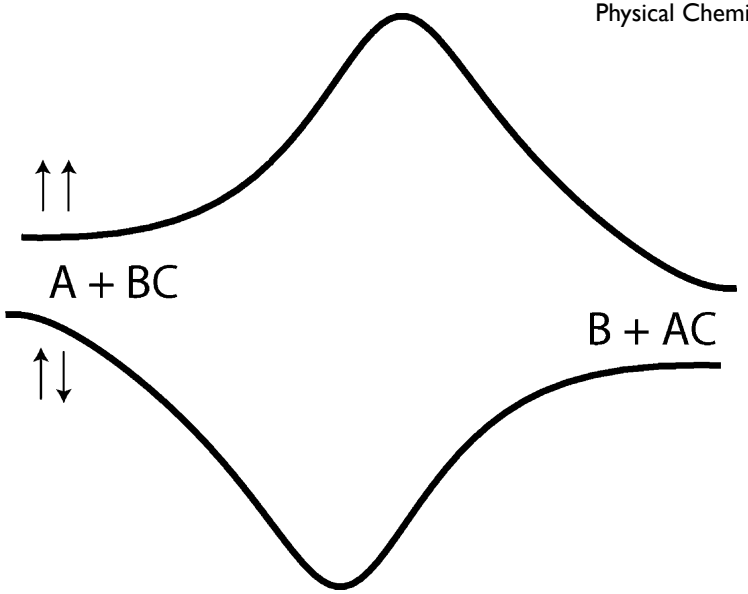
Physical Chemistry Chemical Physics



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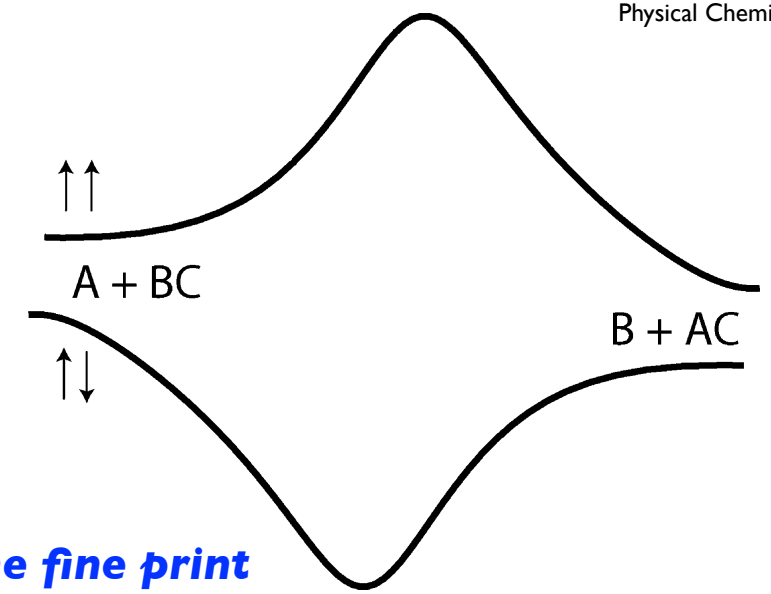

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the fine print

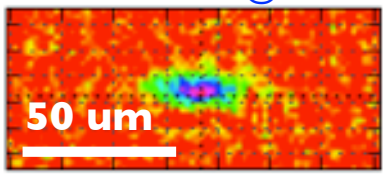
"... non-adiabatic coupling ... may be induced by the spin-rotation interaction and the magnetic dipole–dipole interaction. The latter is negligibly small and ... transitions are determined by the spin-rotation interaction in the open-shell molecule. The spin-rotation interaction can be effectively manipulated with an external electric field."

Experimental goals and results

Li_2

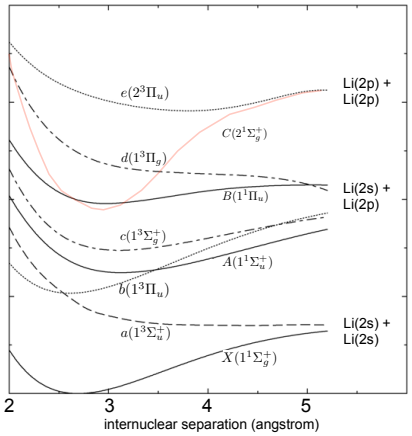
✓ **Feshbach resonances and FR molecule production**

BEC of Li dimers @ 300 nK



✓ **excited triplet spectroscopy (1-color PA)**

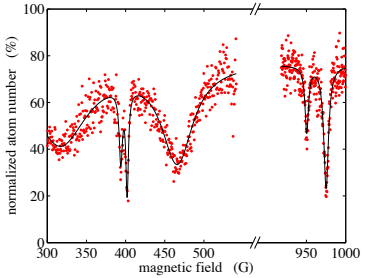
✓ **lowest triplet spectroscopy (2-color PA)**



STIRAP production of triplet Li_2 from FR molecules

LiRb

✓ **Feshbach resonances and FR molecule production**



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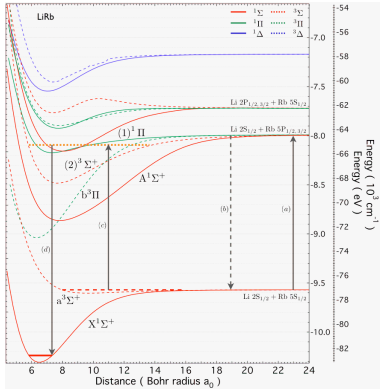

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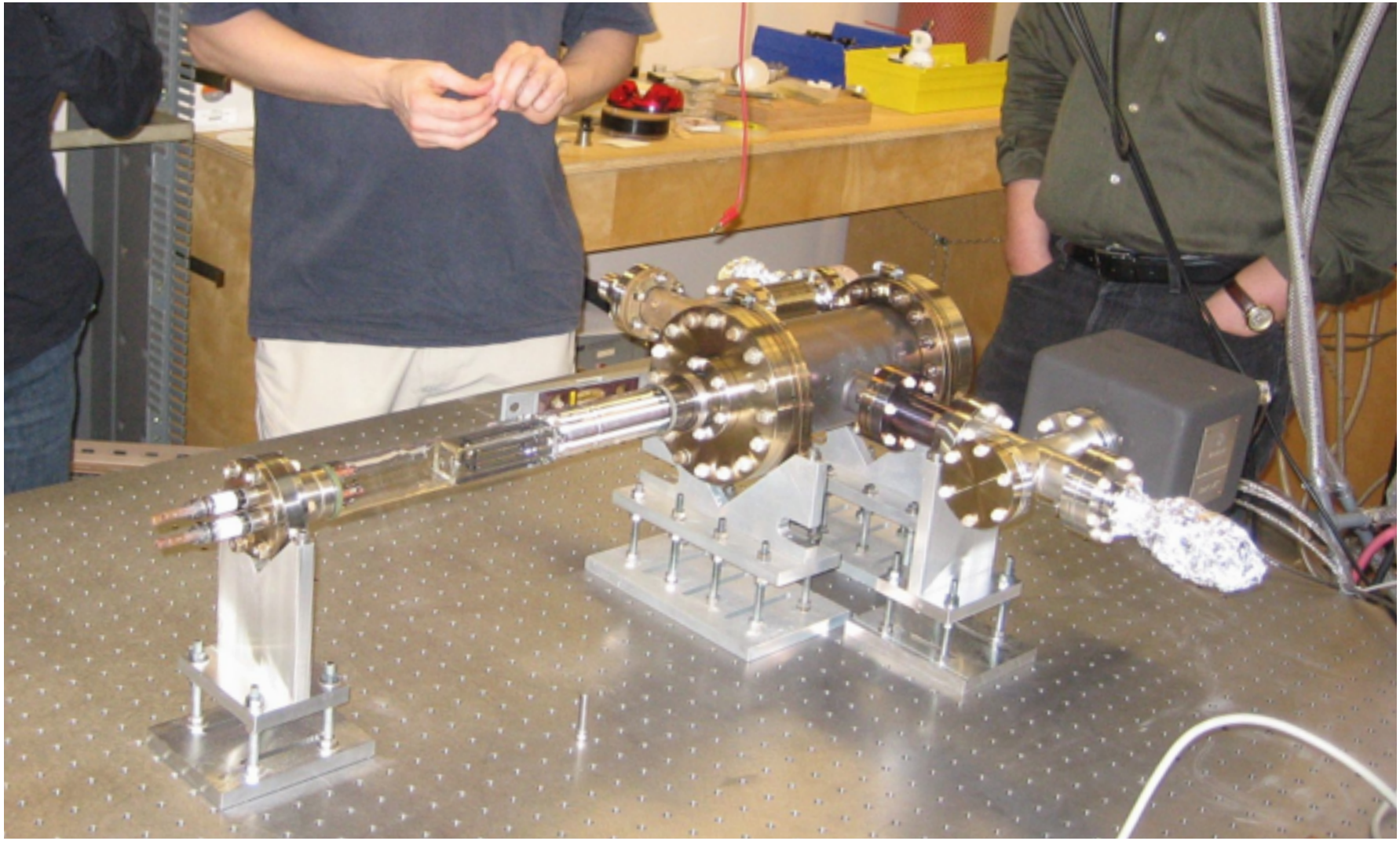
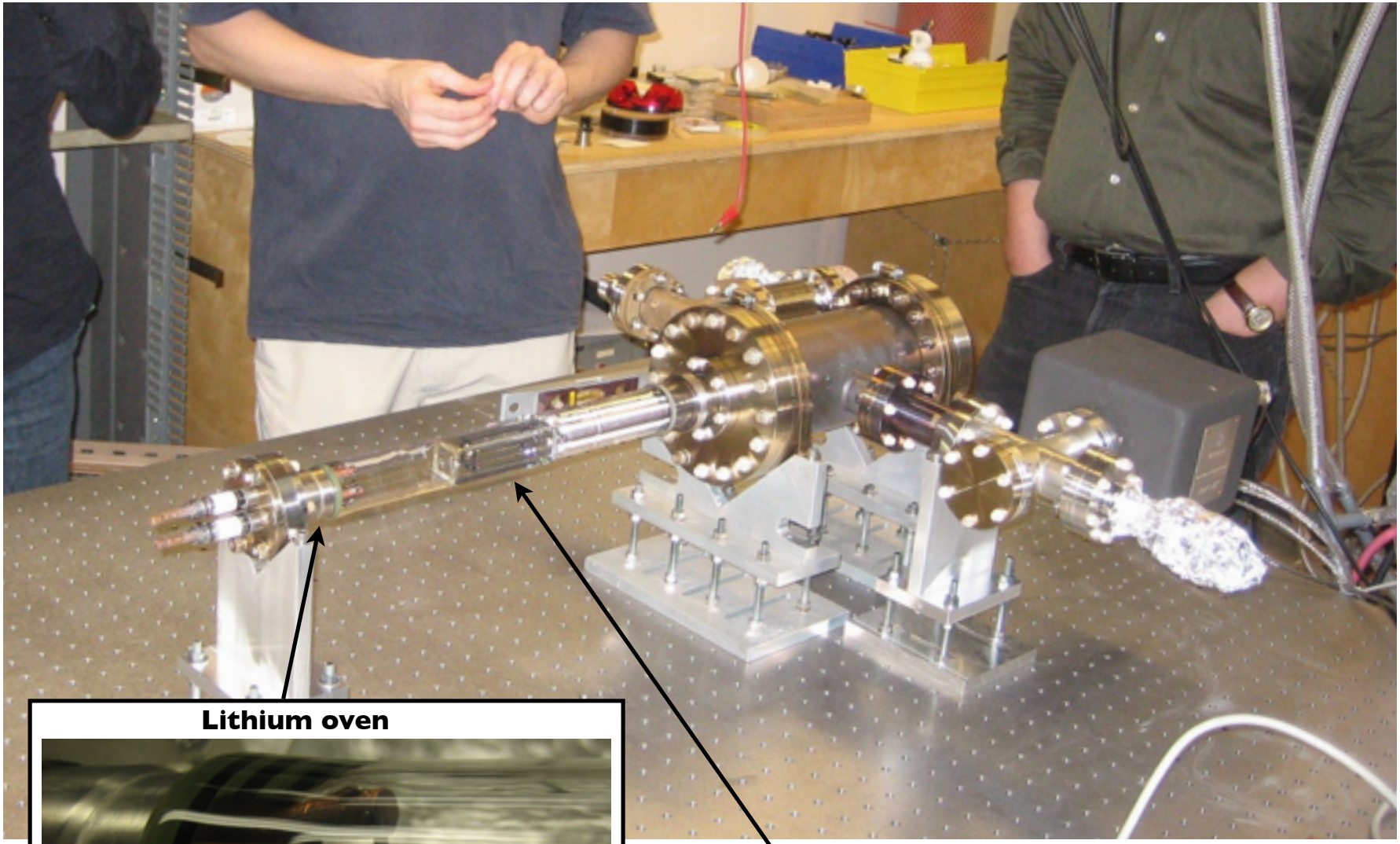


ultra-cold Li+Rb mixtures



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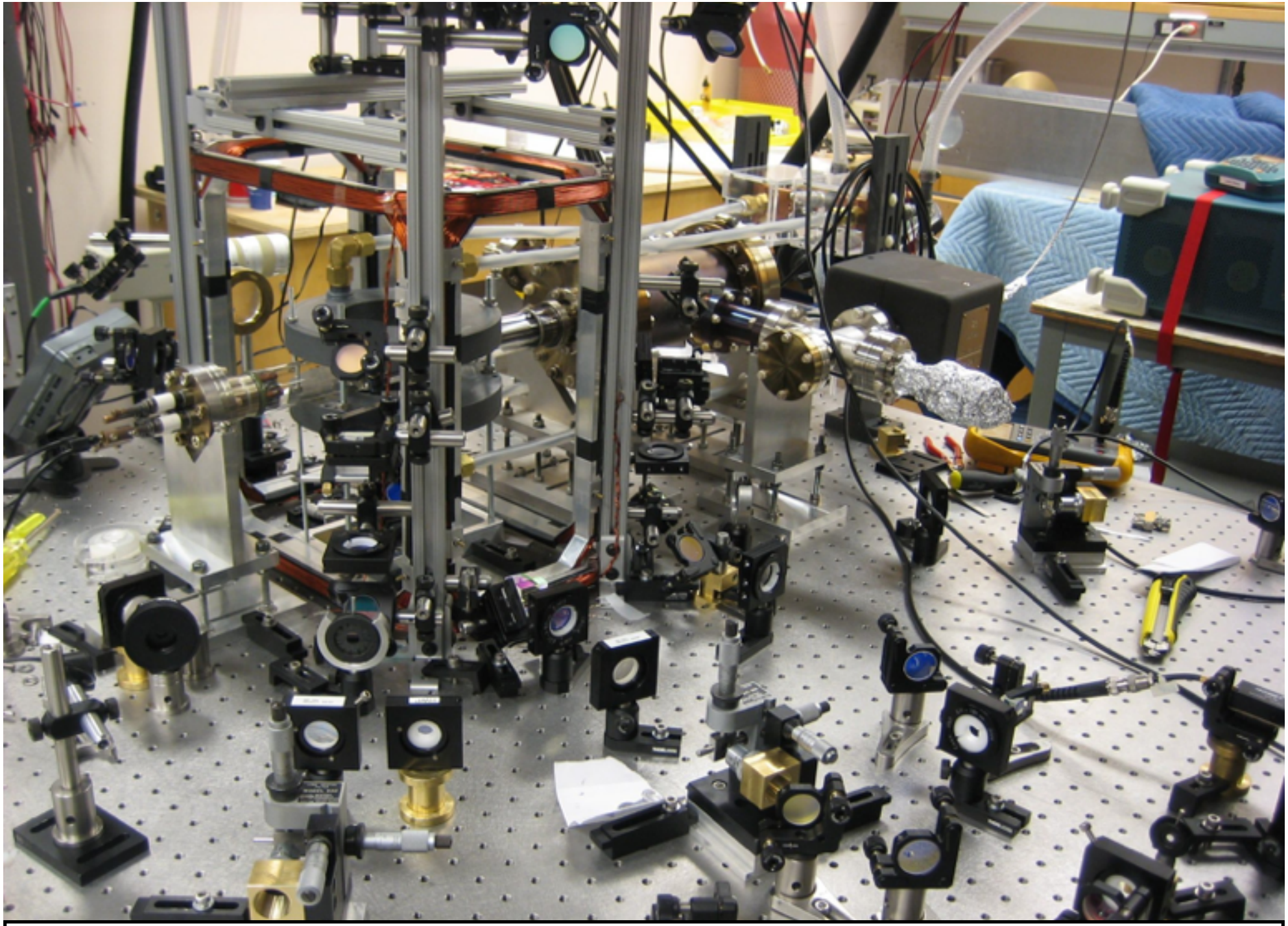

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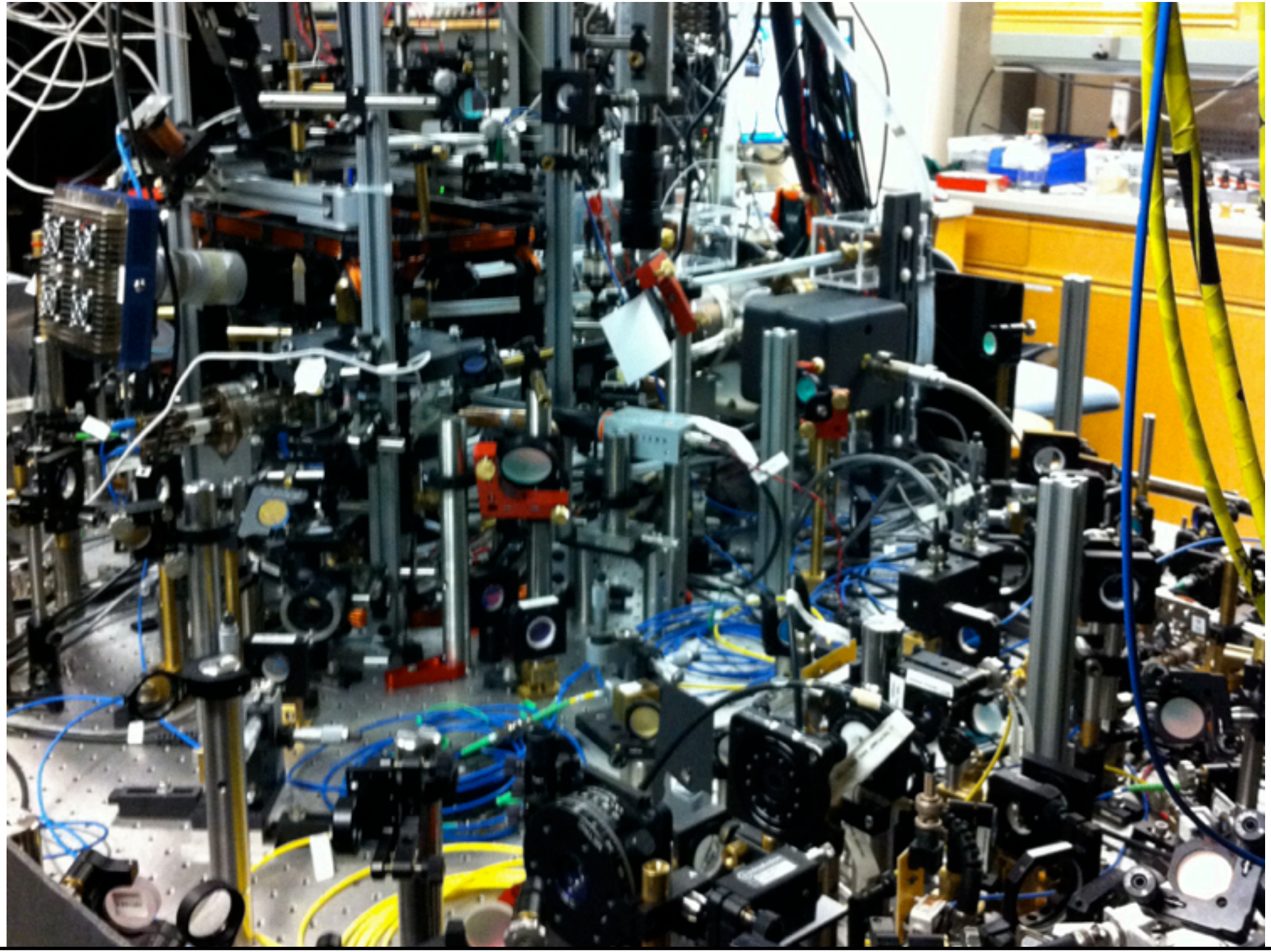

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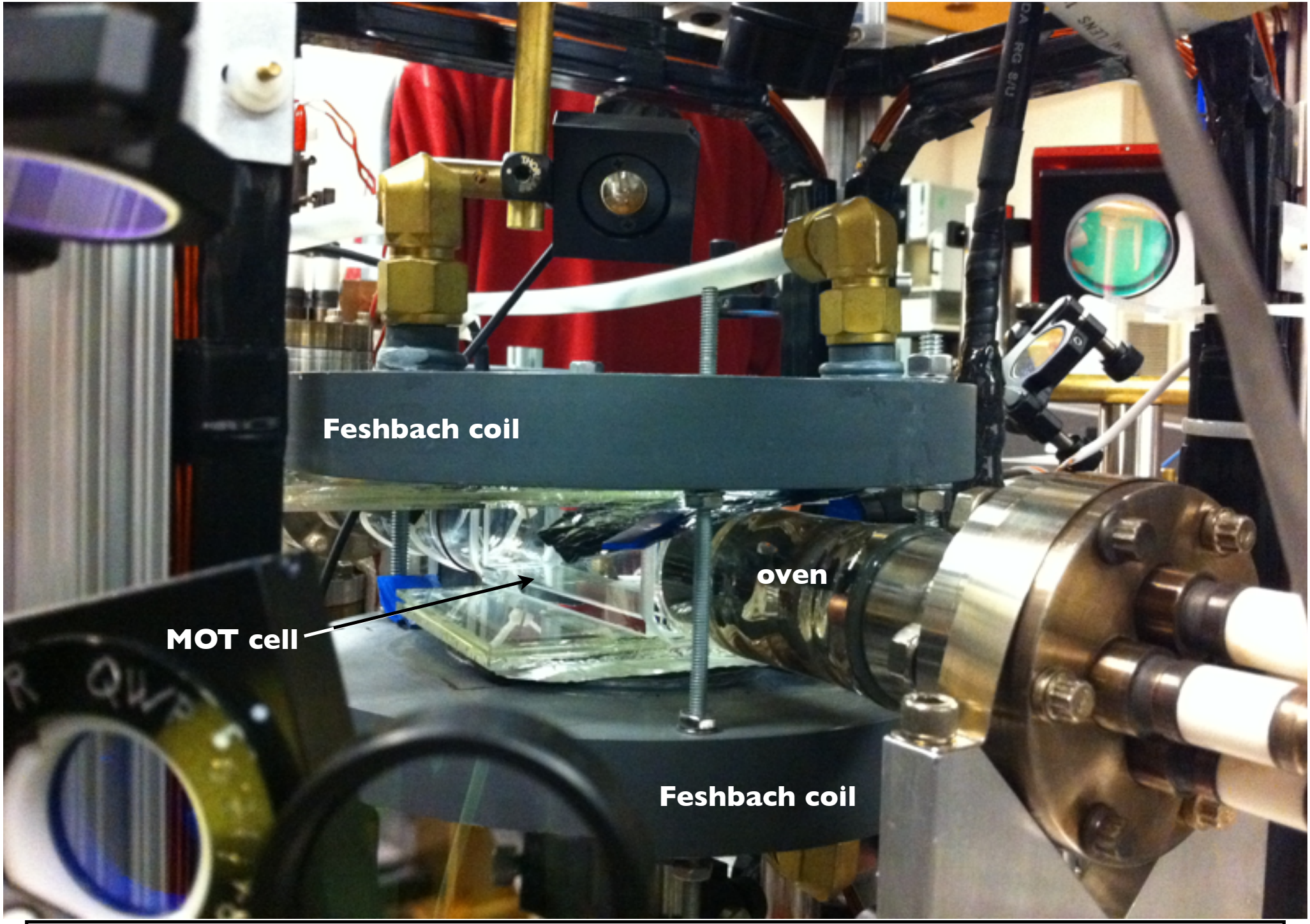

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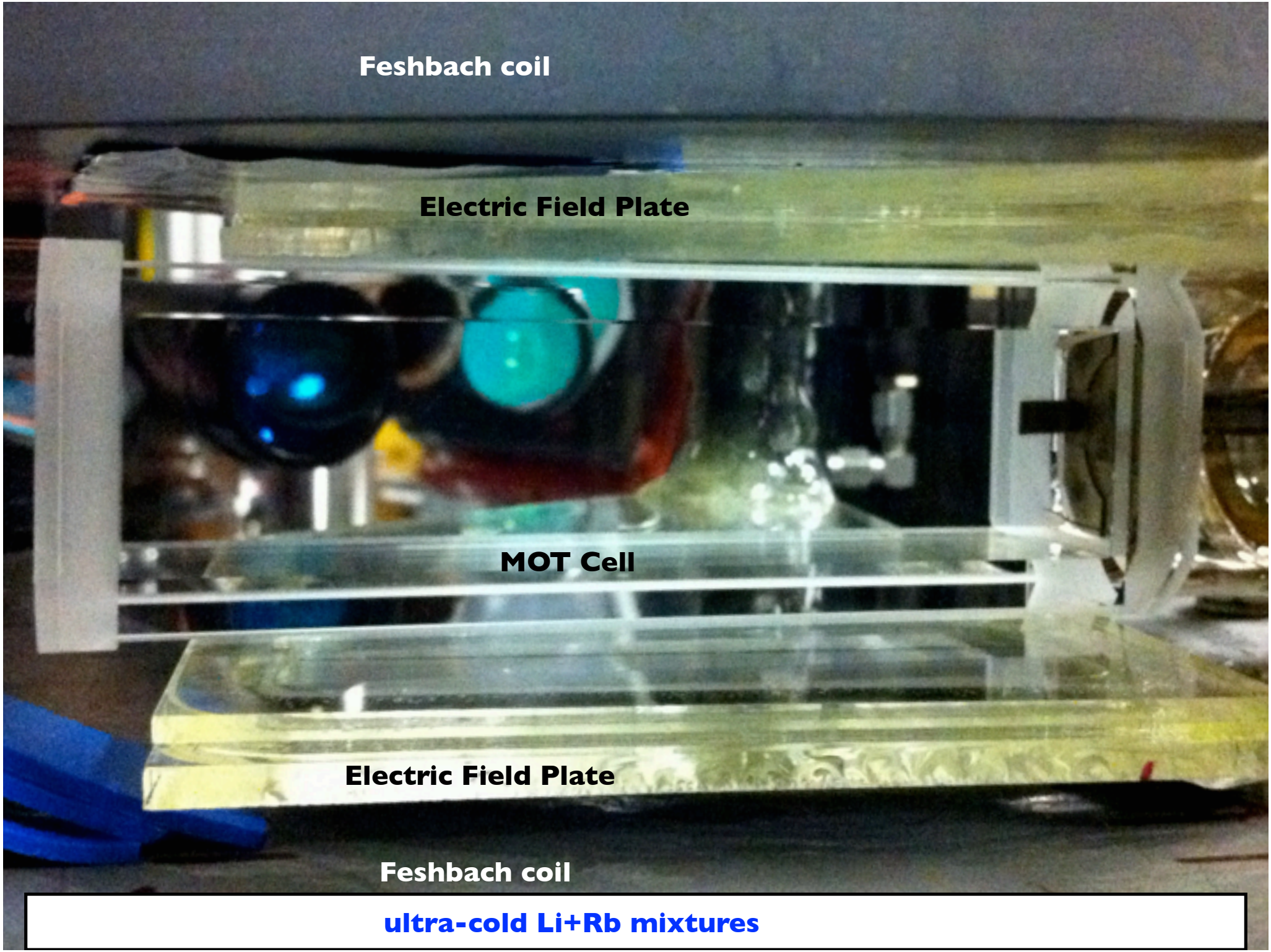

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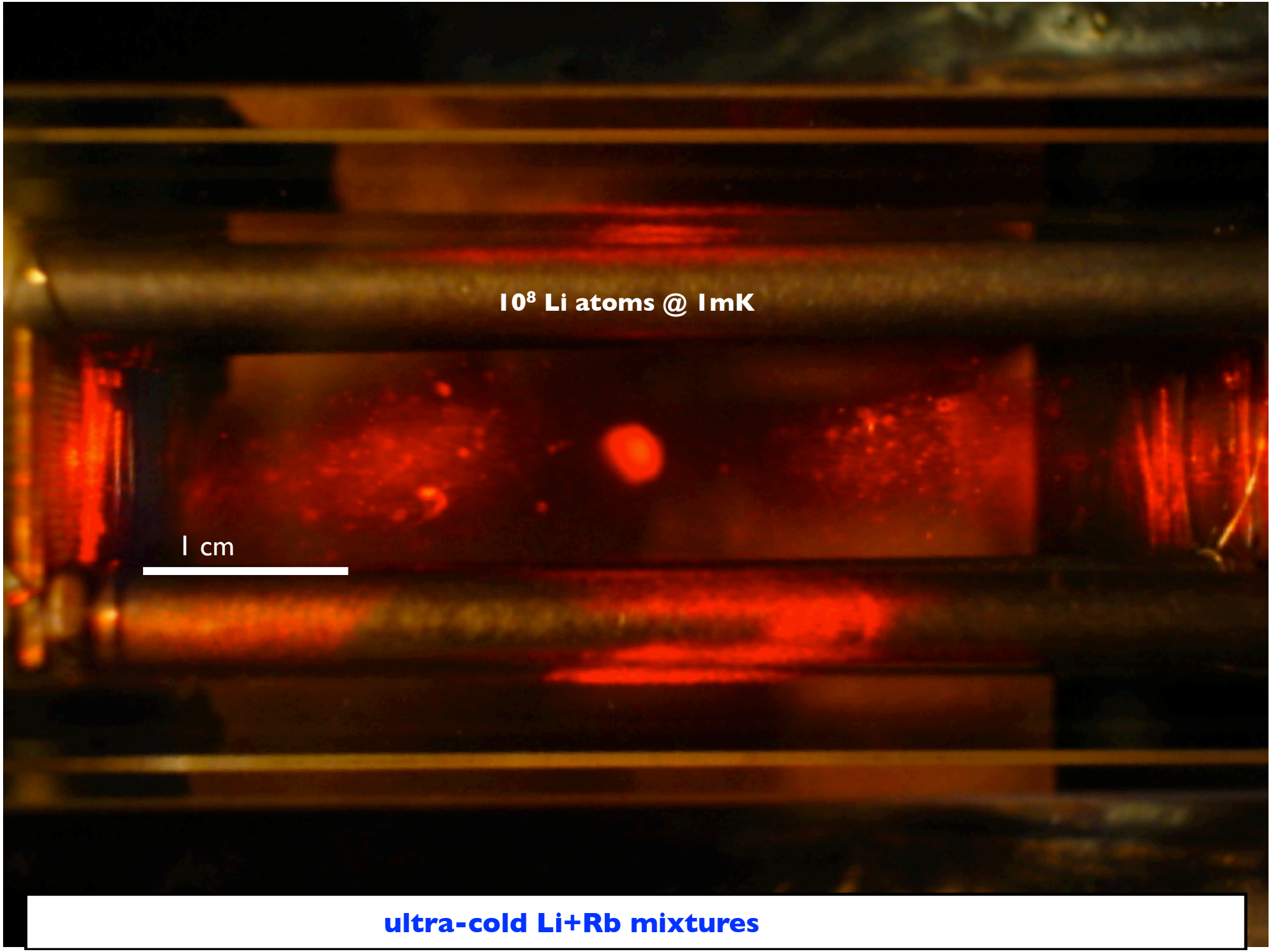

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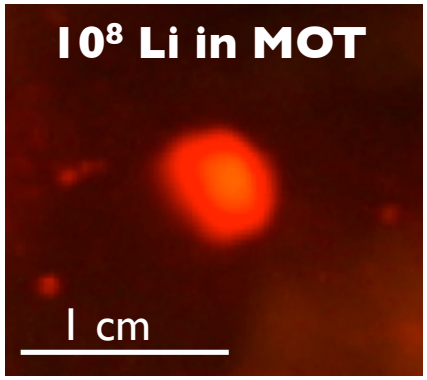
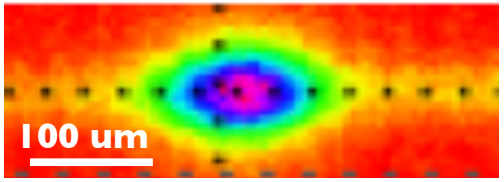
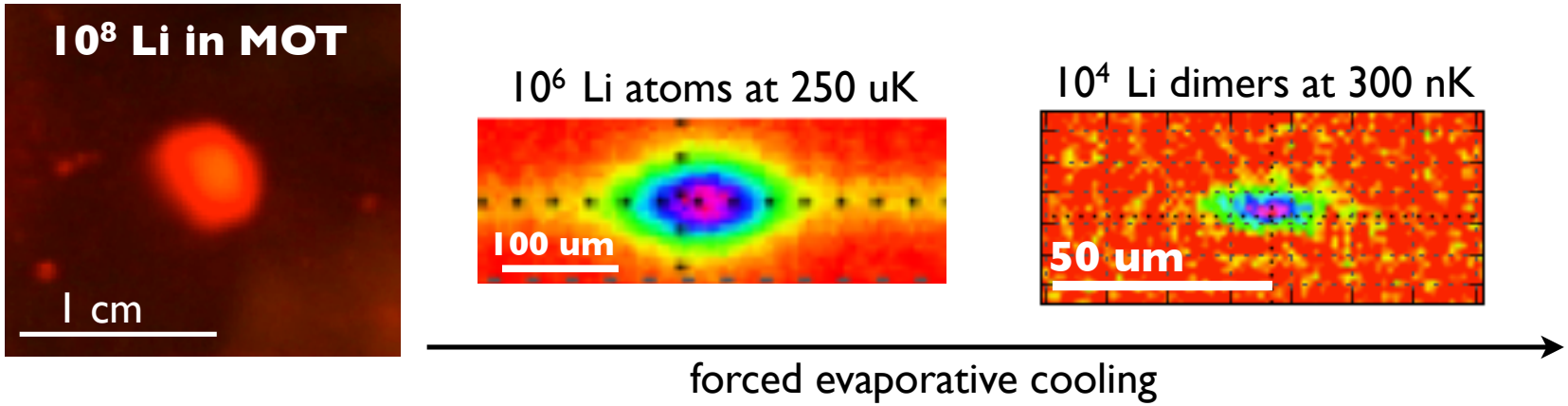


10^6 Li atoms at 250 uK

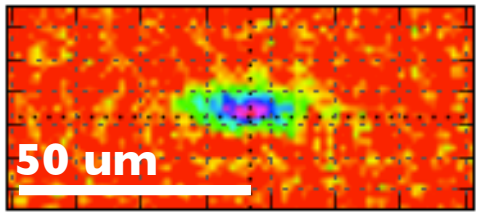


transfer to an ODT

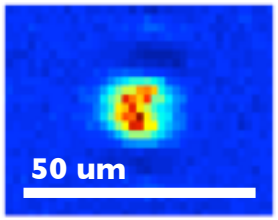

10^8 Li in MOT
1 cm
10^6 Li atoms at 250 uK
100 um
10^4 Li dimers at 300 nK
50 um
forced evaporative cooling

Only Lithium: 10^4 Li dimers, $T \sim 300$nK, $T/T_c \sim 0.7$

10^4 Li dimers at 300 nK

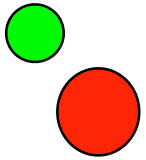


Only Rubidium: 2×10^4 atoms, $T \sim 500$nK, $T/T_c \sim 2$

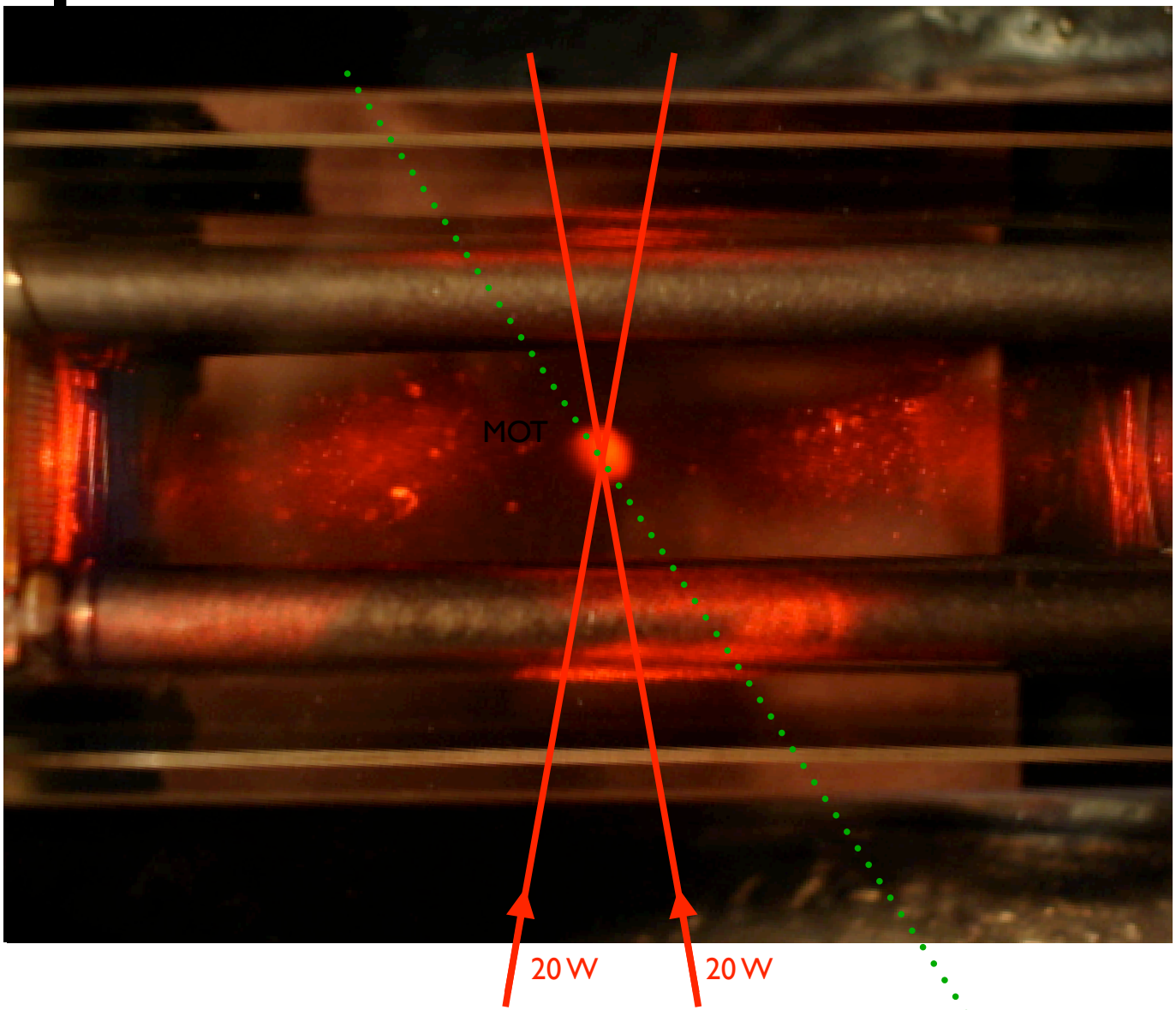


Mixture @ $T \sim 2$ uK, 10^4 Li, $T/T_F \sim 0.3$, 10^4 Rb, $T/T_c \sim 3$

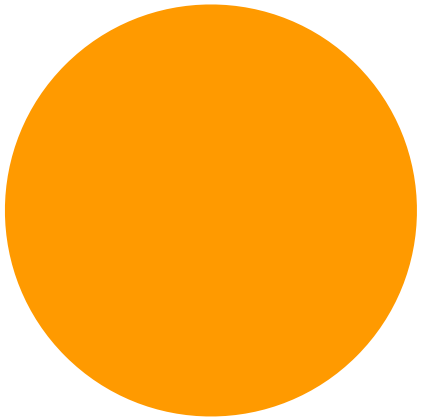
observing Rb+Li Feshbach resonances



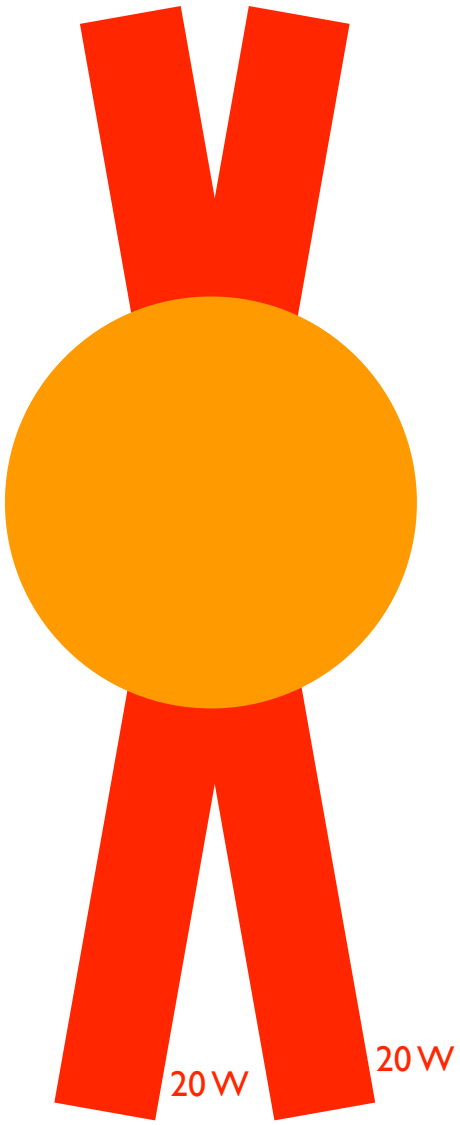
Trap Schematic



Load Lithium MOT

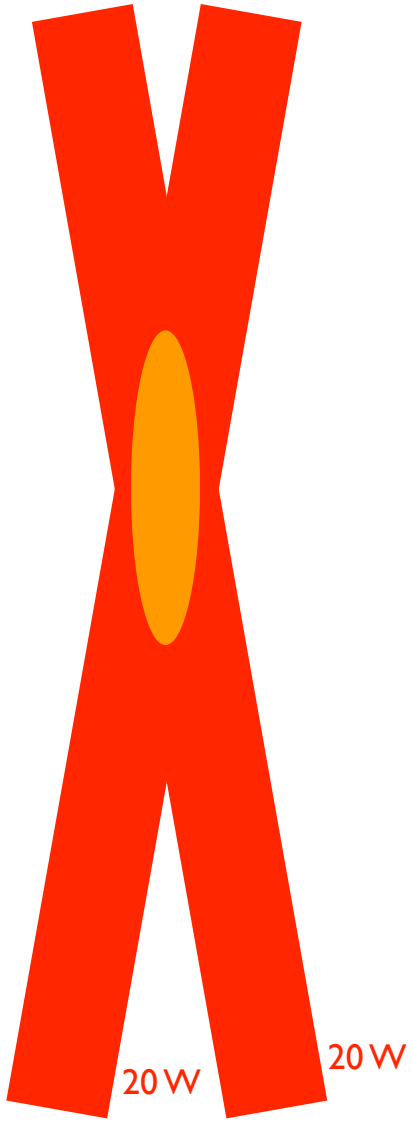


Transfer Li from MOT to crossed trap



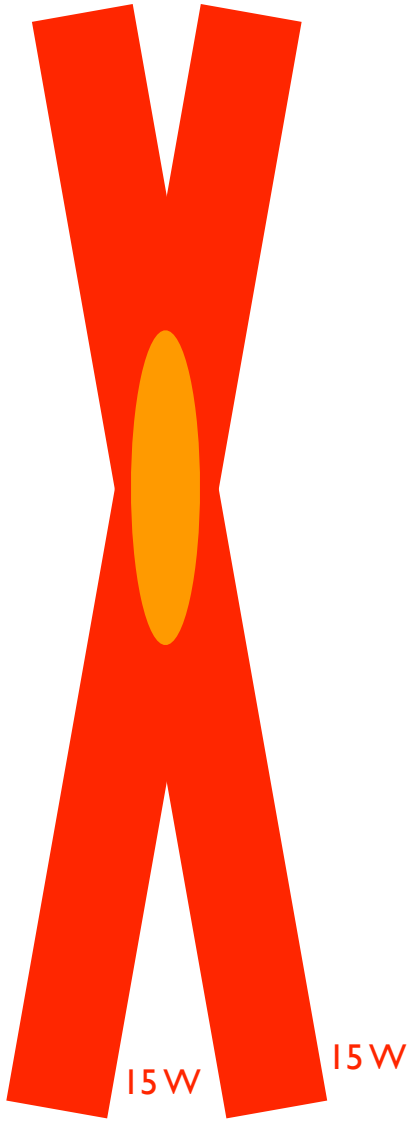
Crossed ODT

Transfer Li from MOT to crossed trap



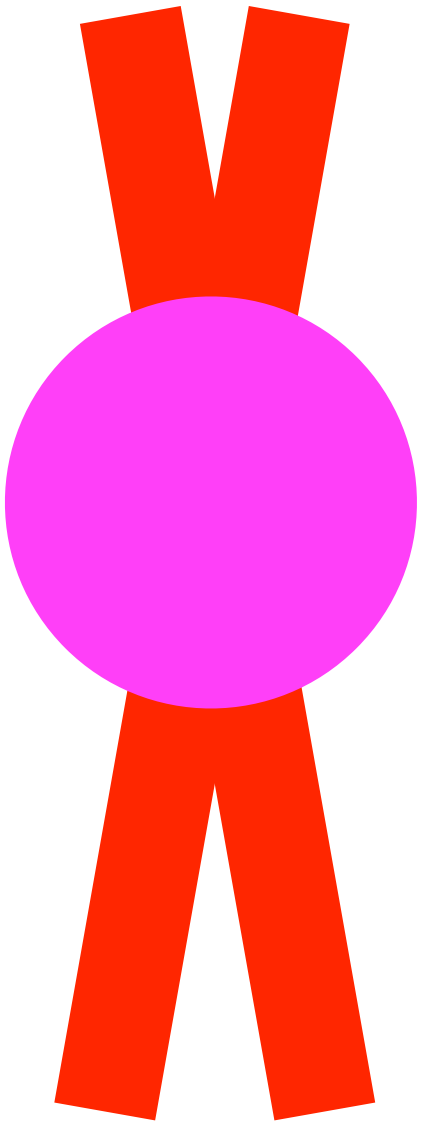
Crossed ODT

Evaporate Li from crossed trap



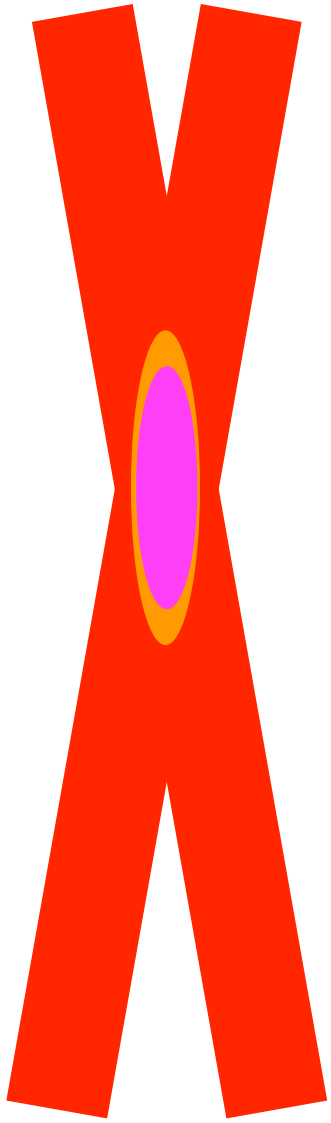
Crossed ODT

Load Rb MOT while holding Li



Crossed ODT

Transfer Rb to crossed trap

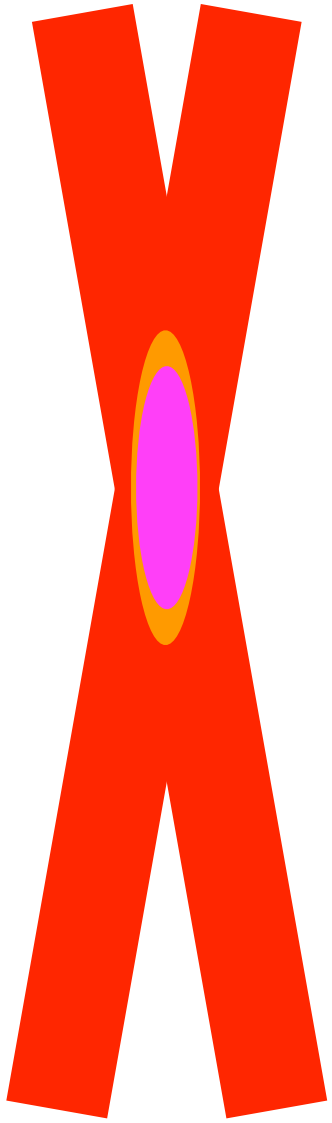


Crossed ODT

Rb MOT turned off:

In trap: Rubidium and Lithium

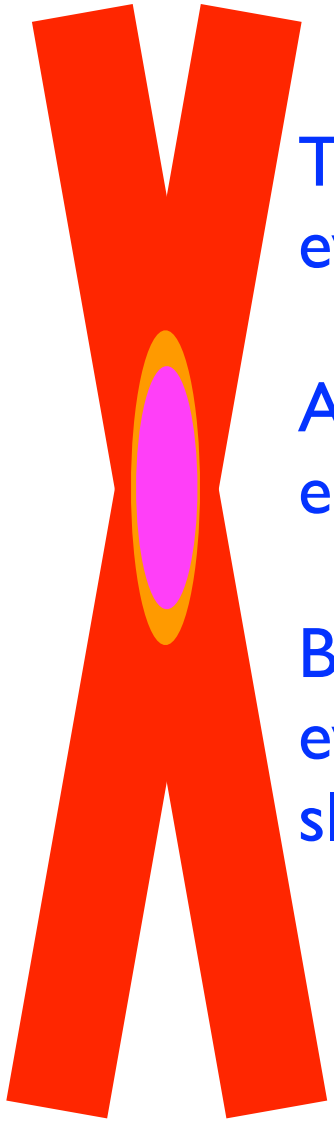
Both Rb and Li in trap: *trap depths different*



Trap for Li half as deep for Rb

Crossed ODT

Simultaneous evaporation of Li and Rb



Crossed ODT

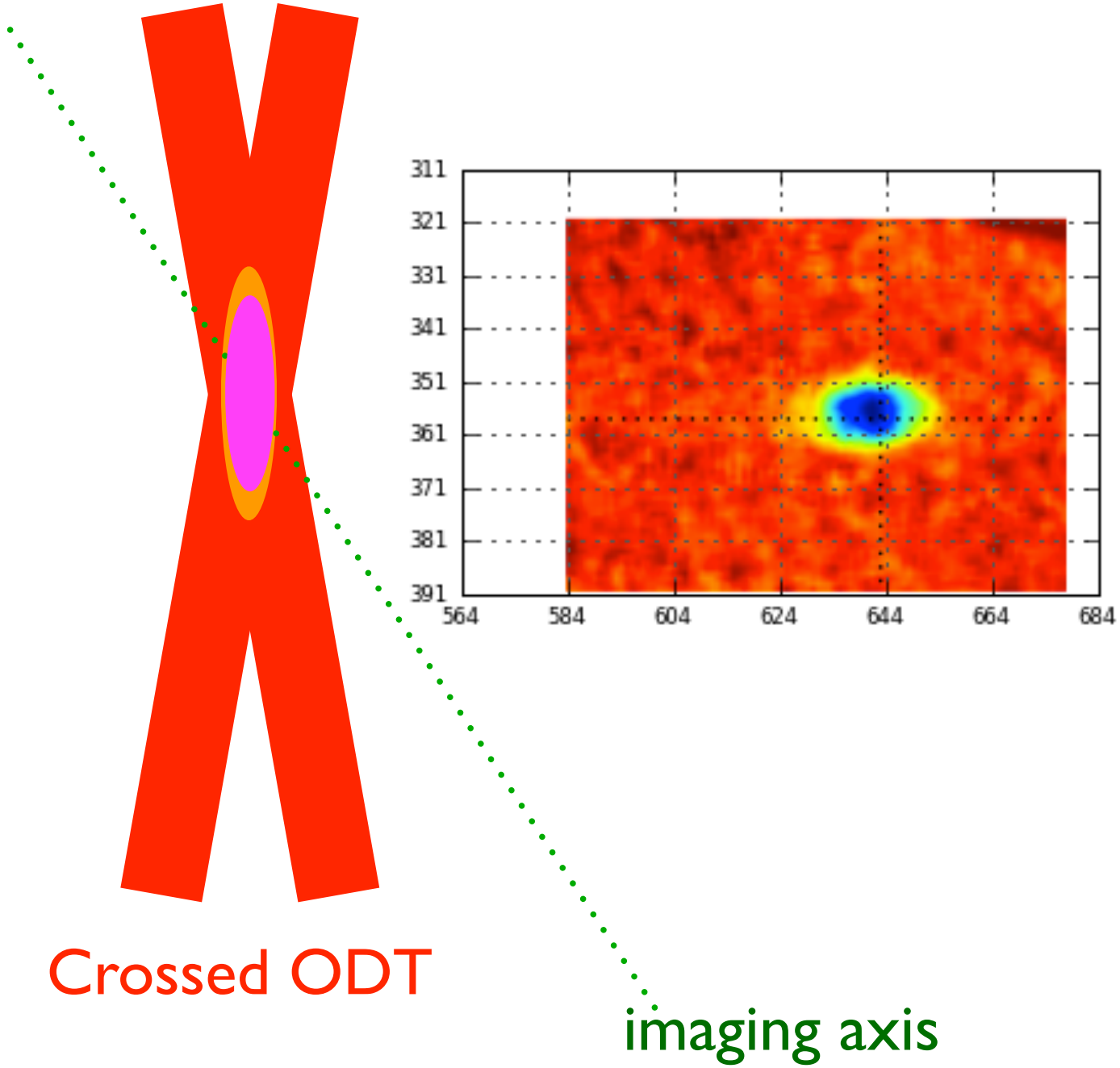
Trap power lowered to force evaporation losses

A) If Li+Rb reach thermal equilibrium, Li leaves trap quickly

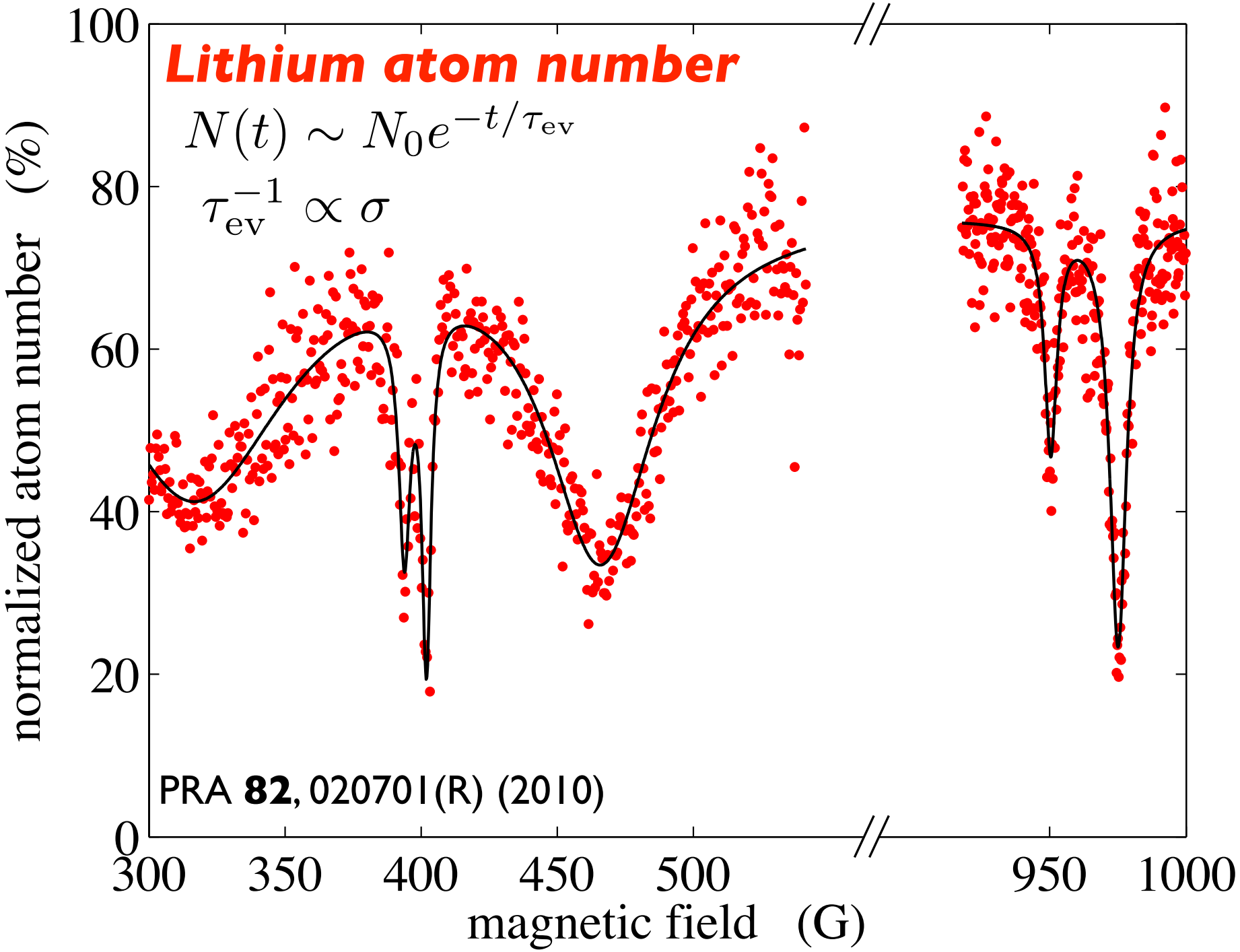
B) If Li+Rb decoupled, Li evaporatively cools and leaves trap slowly

Trap for Li half as deep for Rb

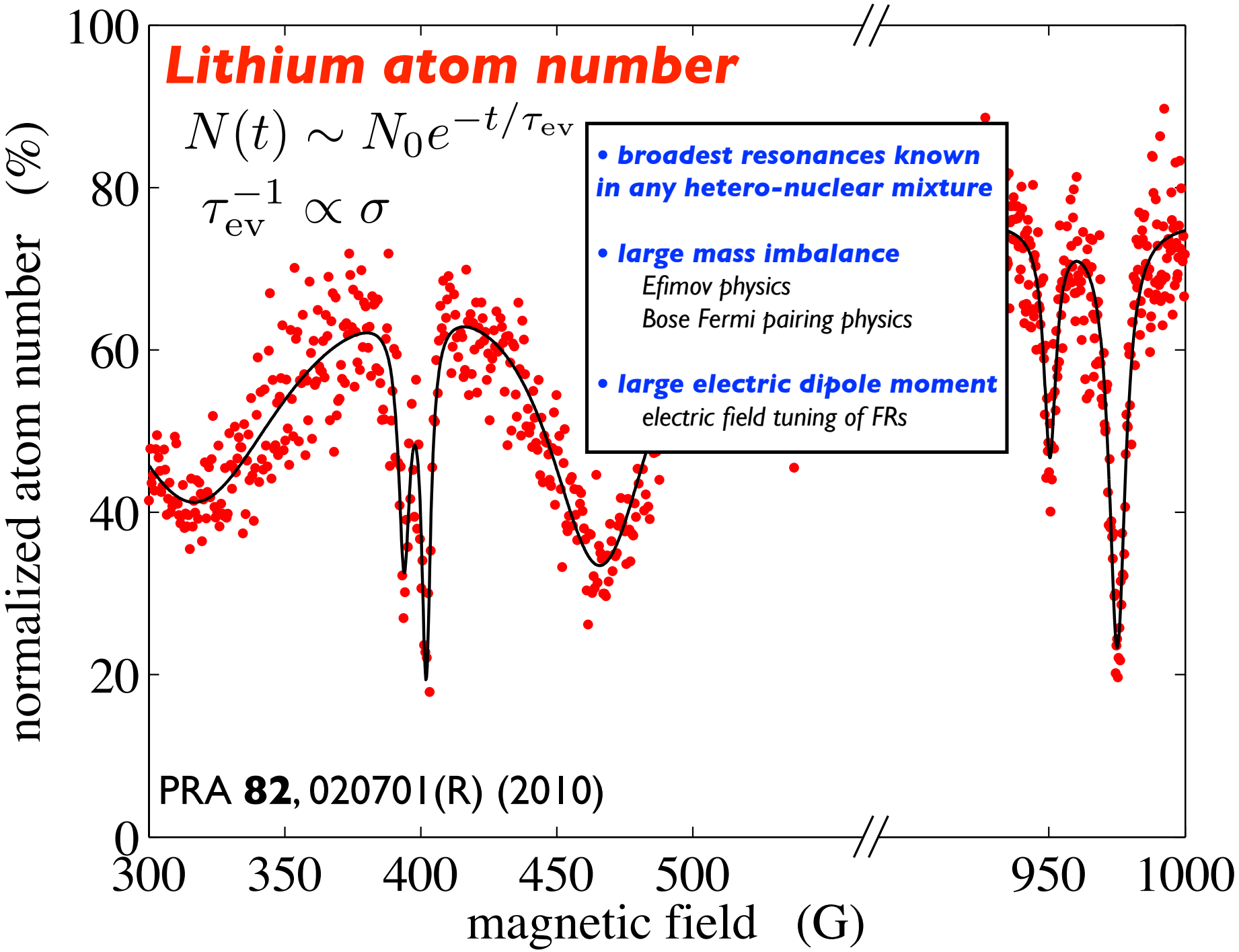
Image Li or Rb



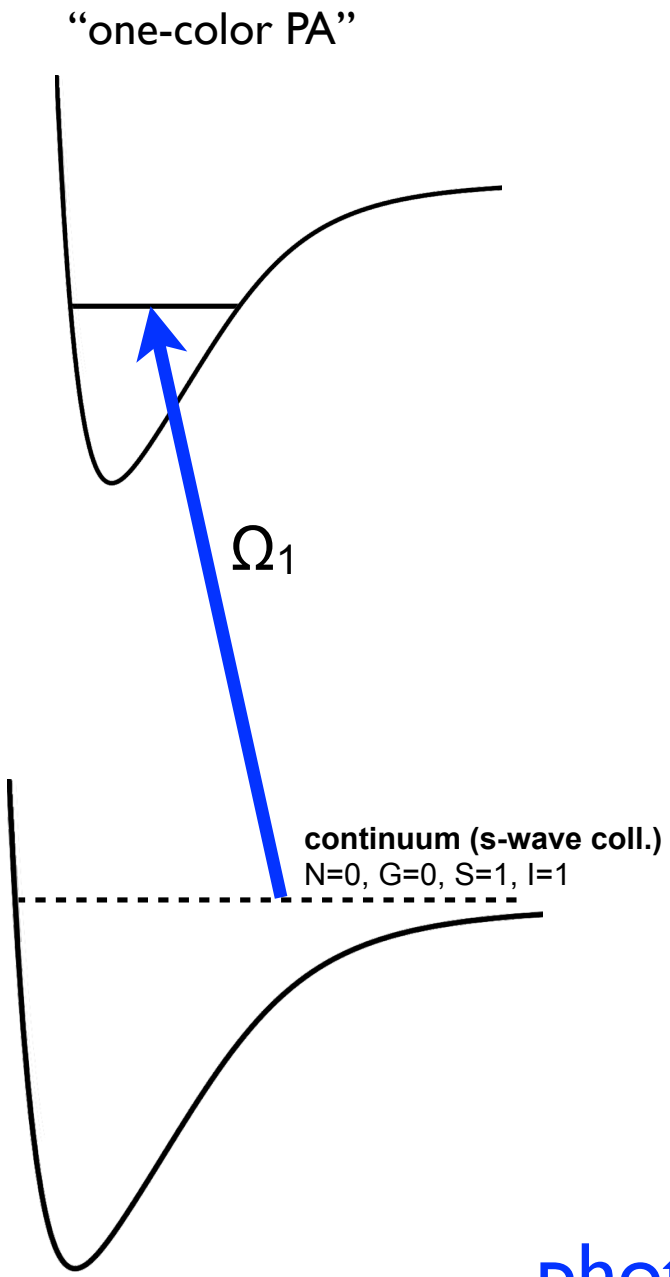
Feshbach resonances in ^{6}Li+^{85}Rb mixtures



Feshbach resonances in ^{6}Li+^{85}Rb mixtures



For photo-association, just add additional light



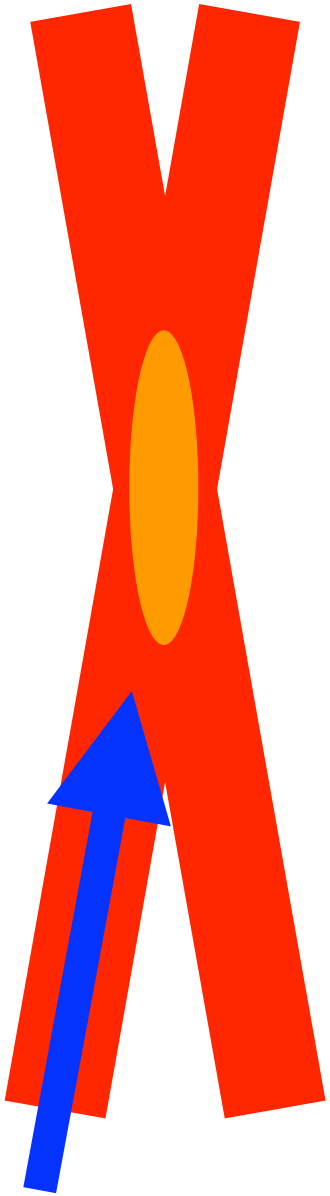
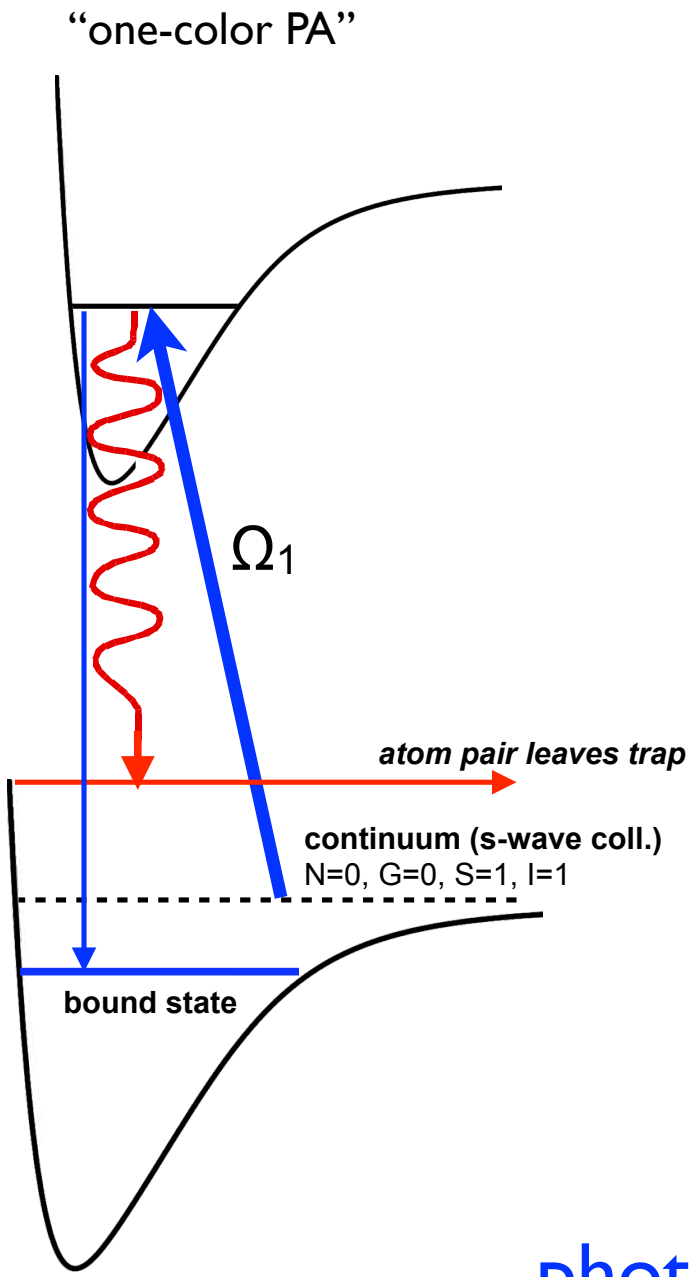


photo-association light

For photo-association, just add additional light



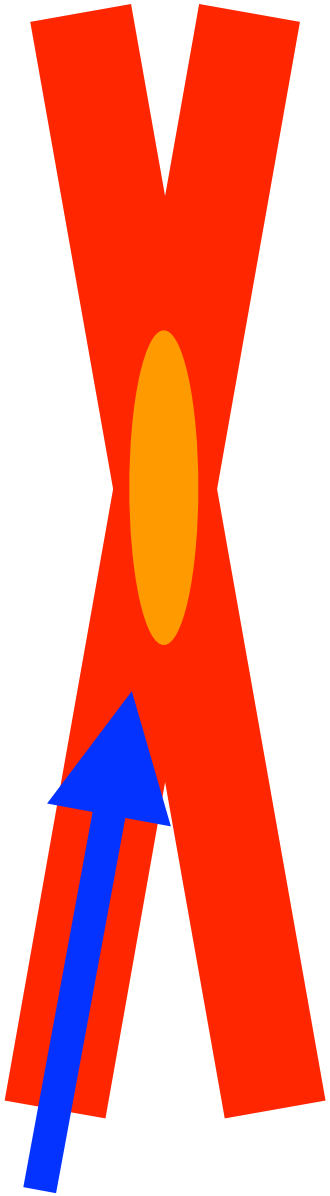


photo-association light

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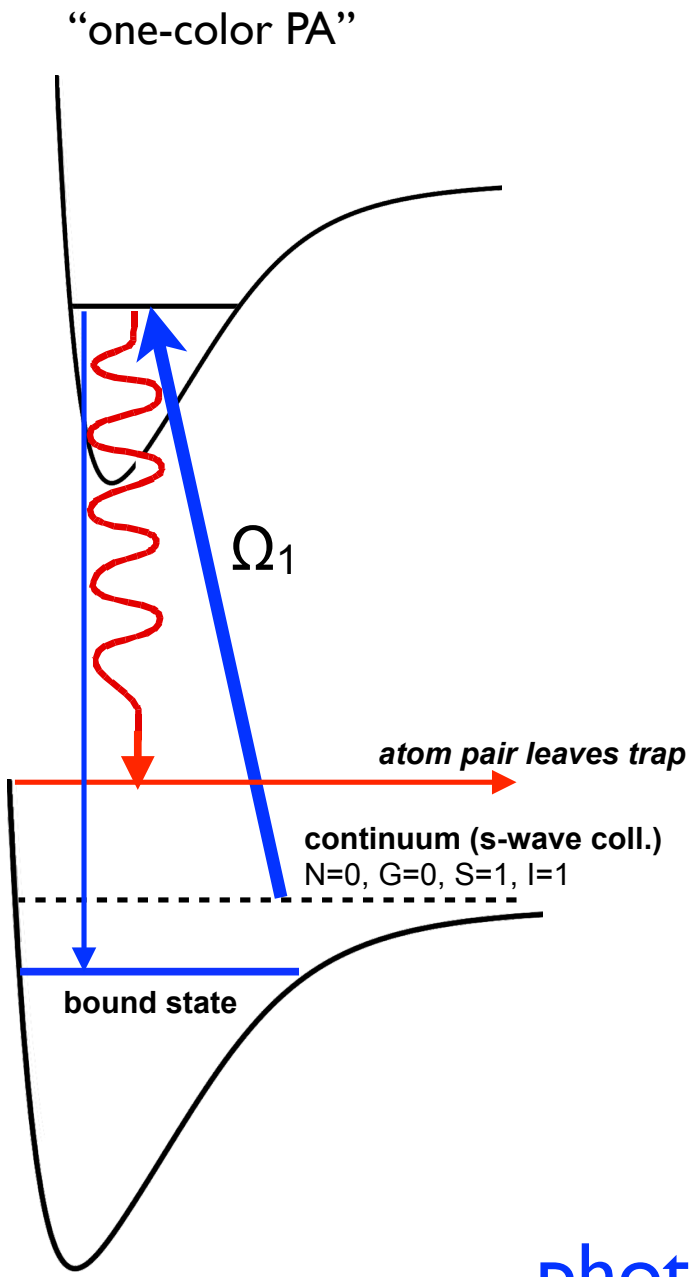


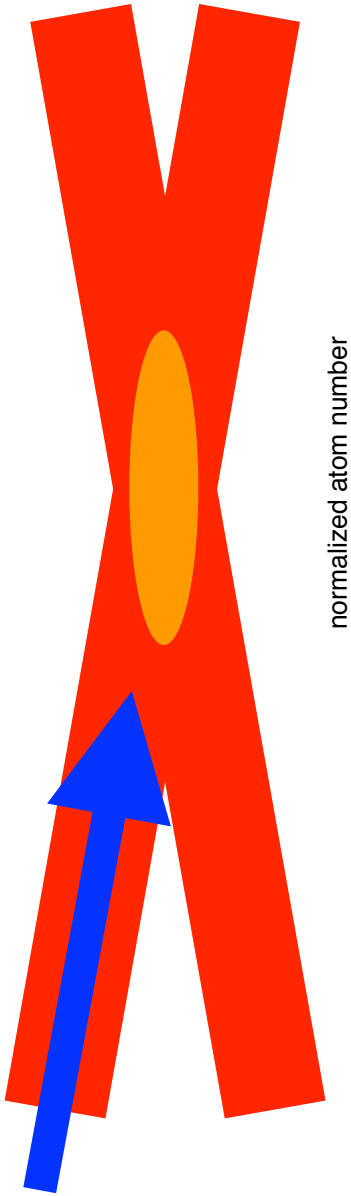
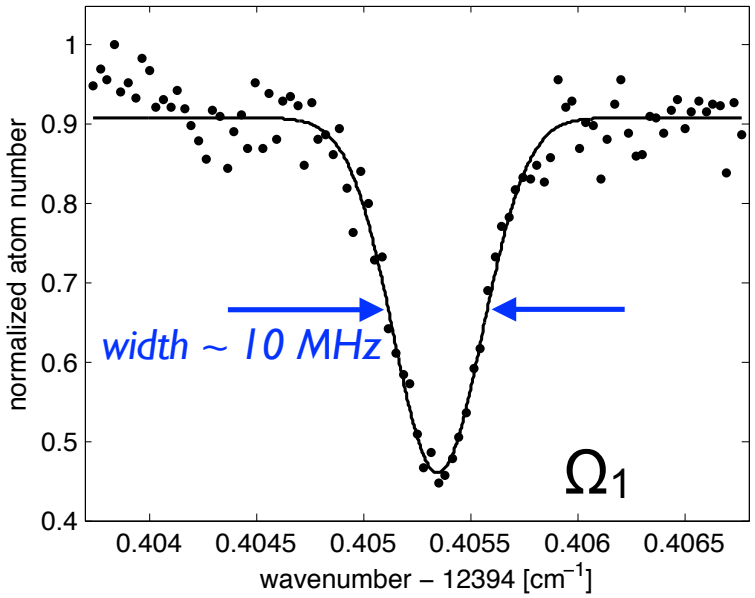


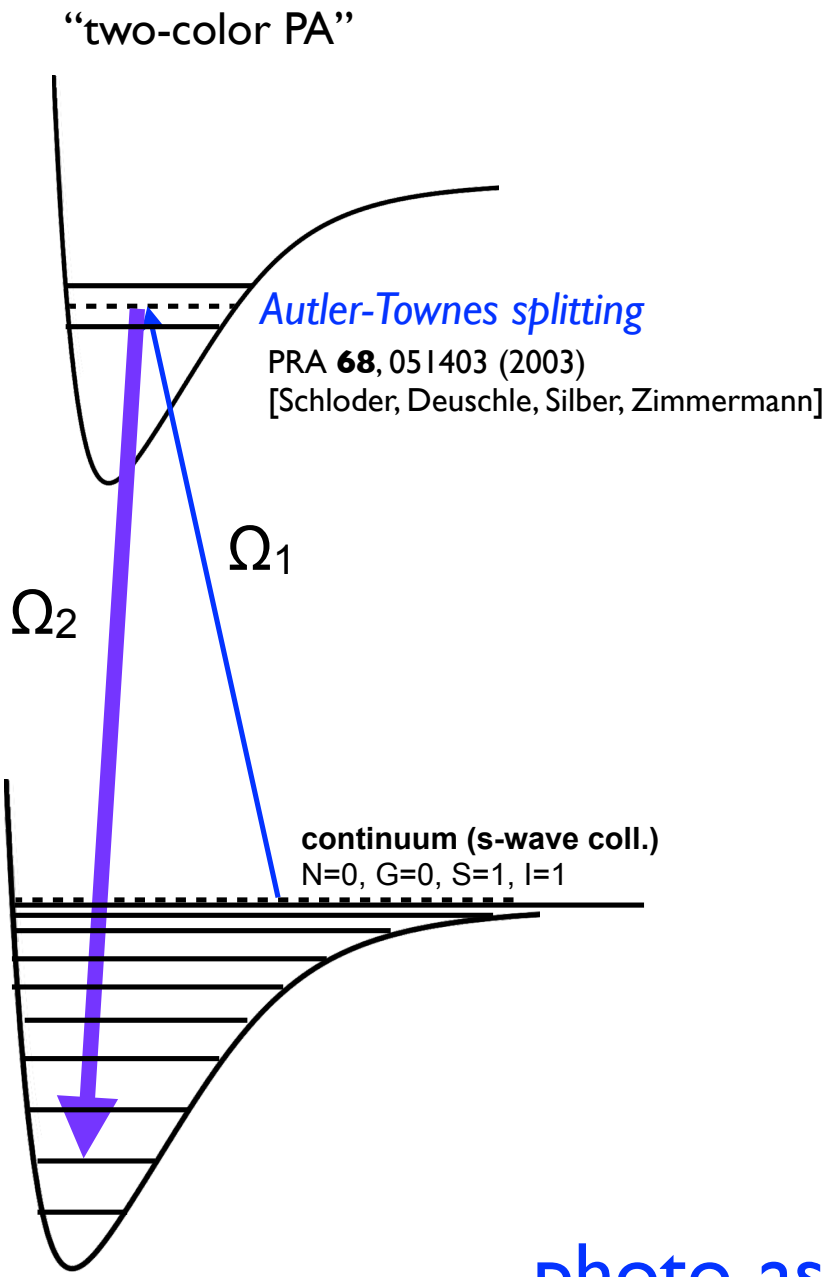
photo-association light

atom loss



width set by excited state lifetime

For photo-association, just add additional light



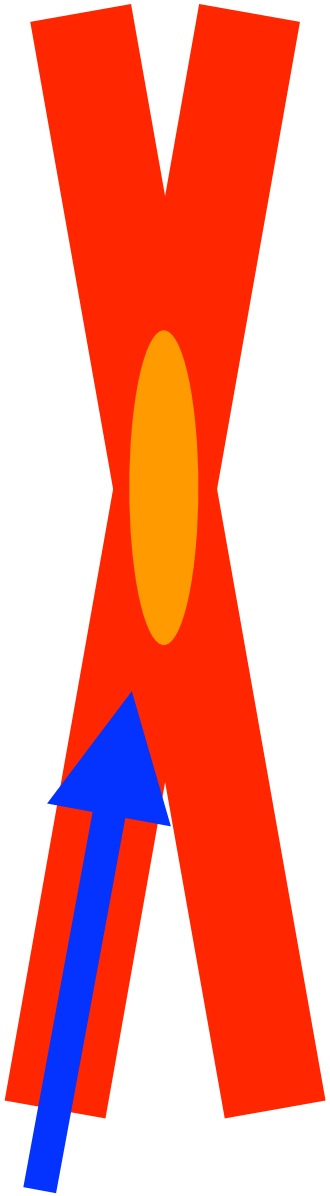
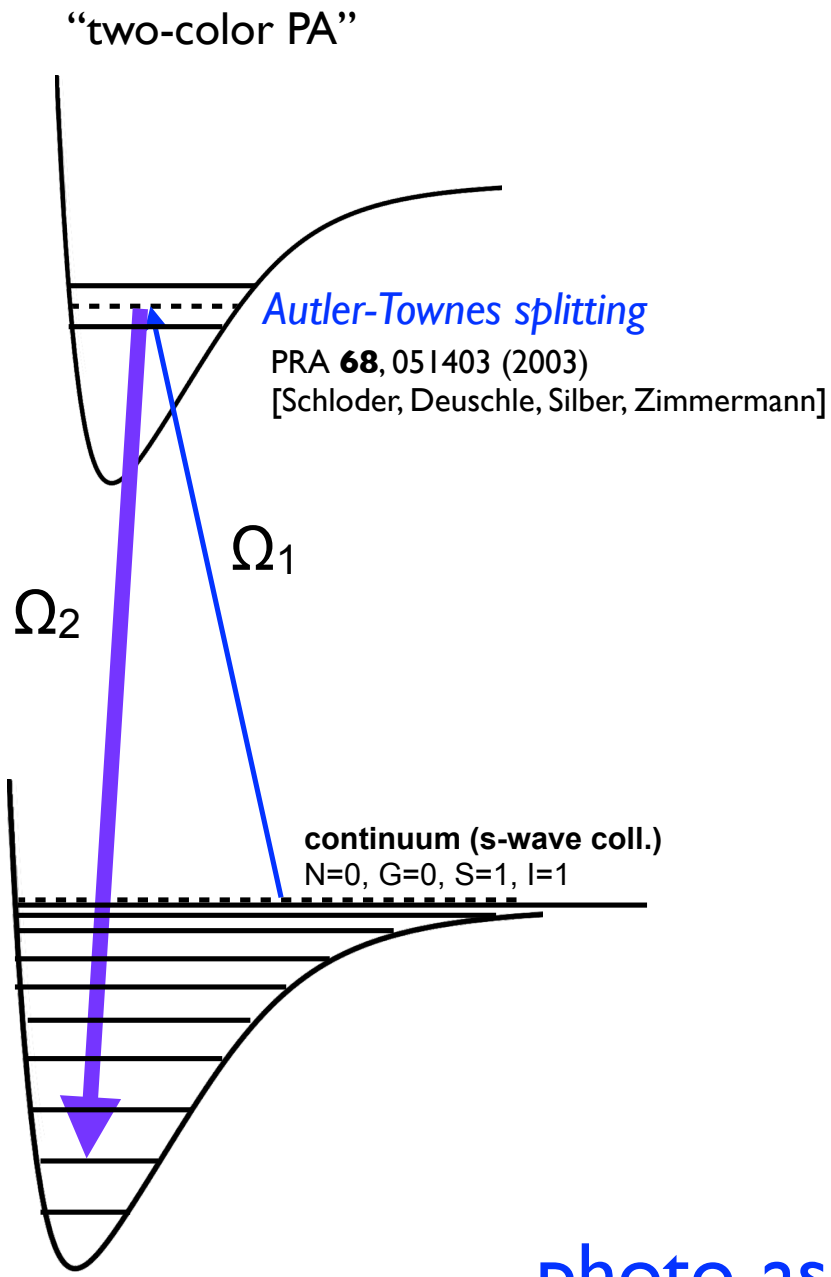
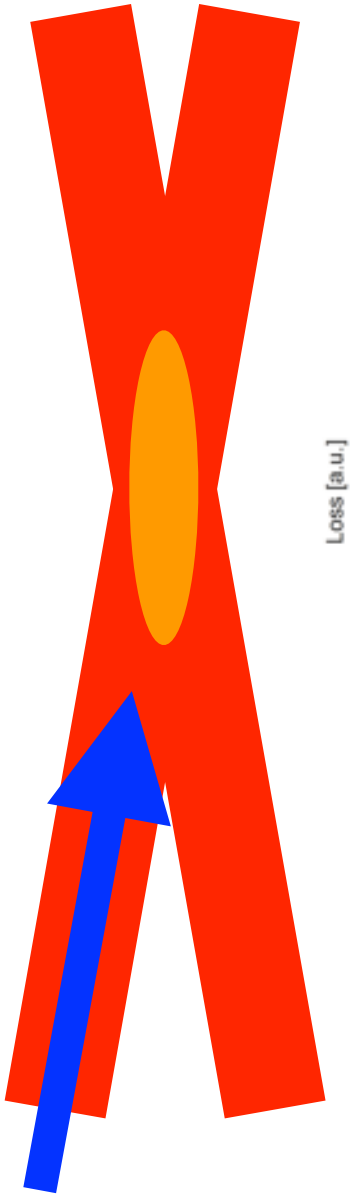


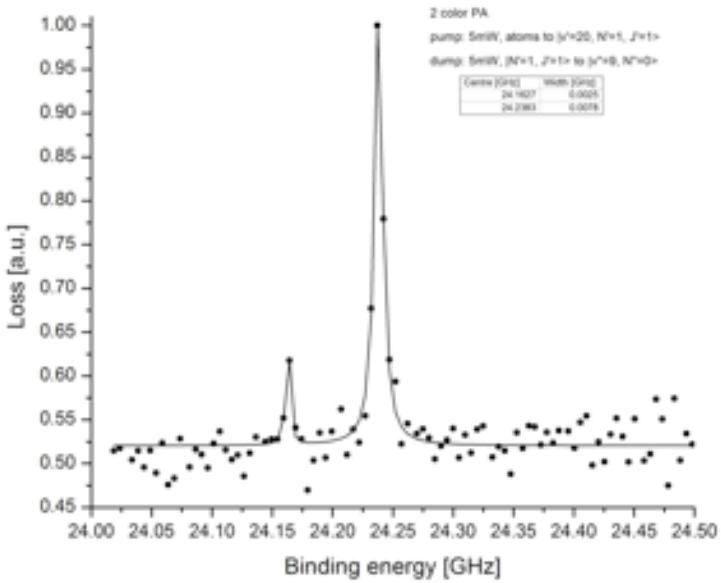
photo-association light

For photo-association, just add additional light





atom loss suppressed

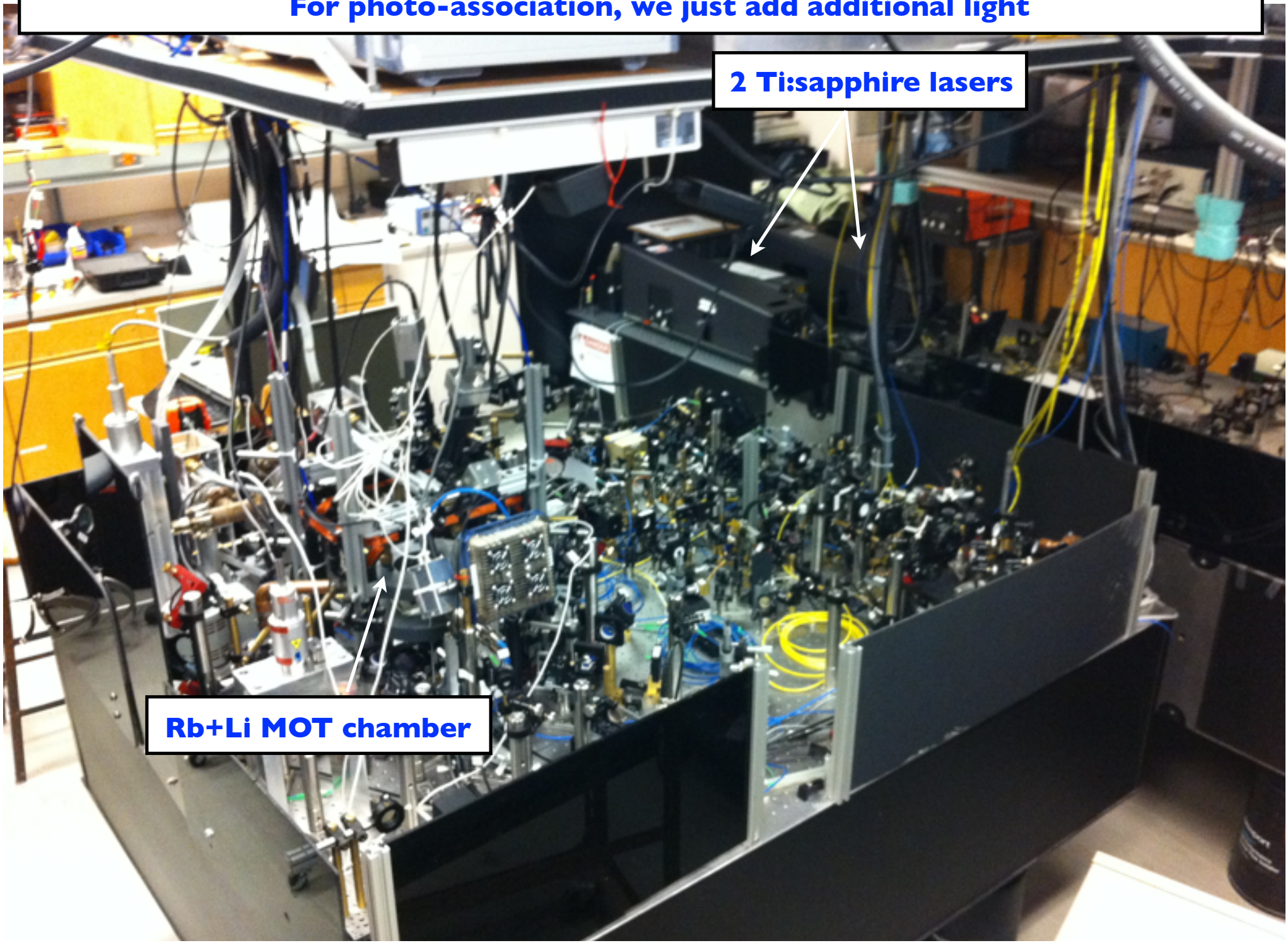


$\Omega_2 - \Omega_1$

width set by coupling induced by Ω_2

photo-association light

For photo-association, we just add additional light



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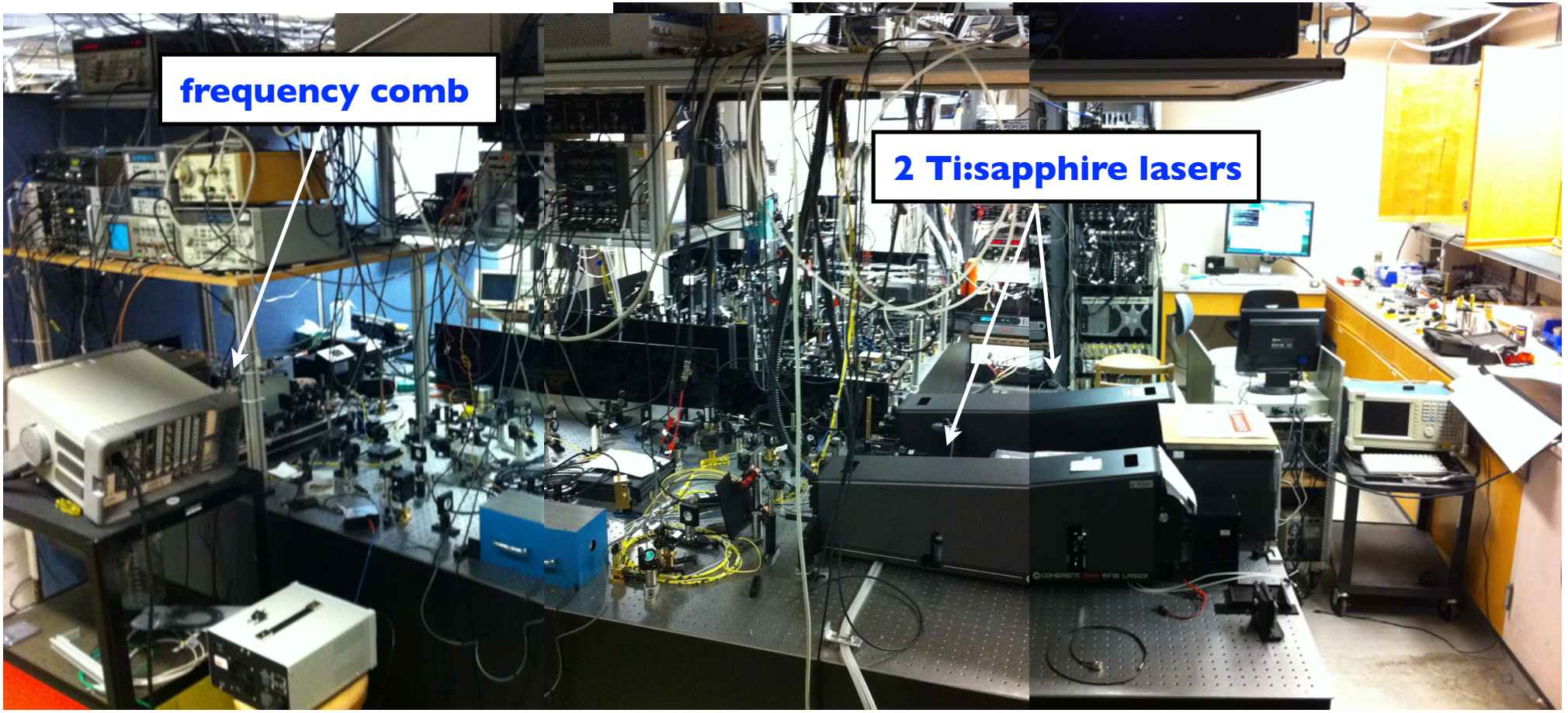
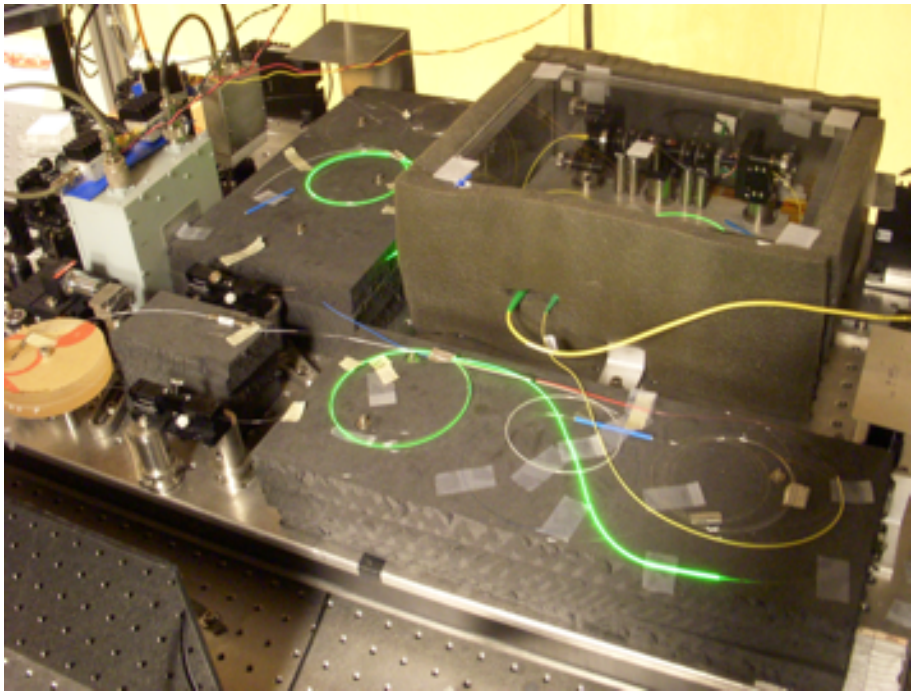
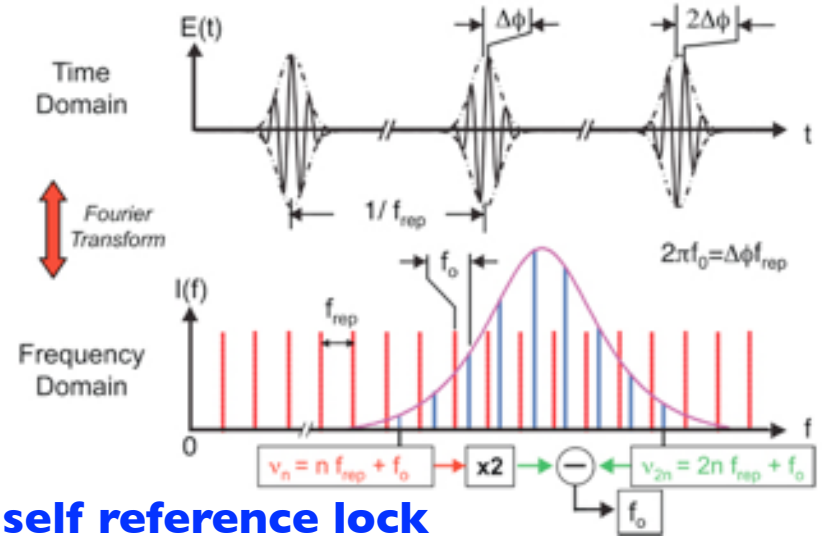


Photo-association (PA) laser system

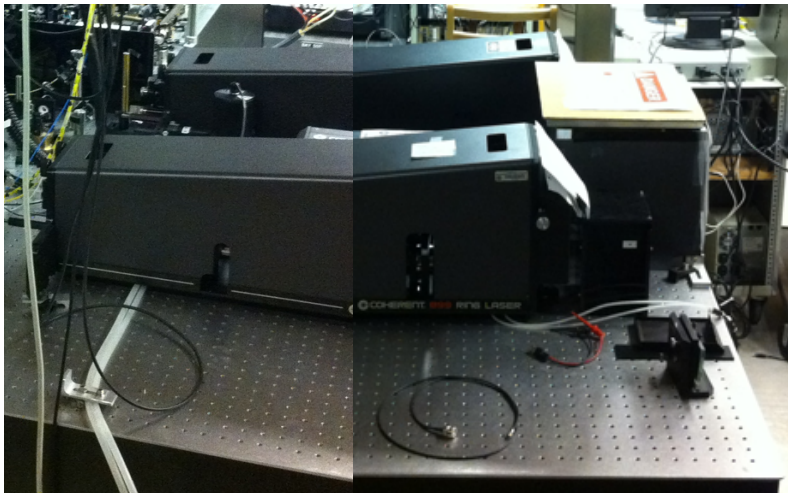
frequency comb





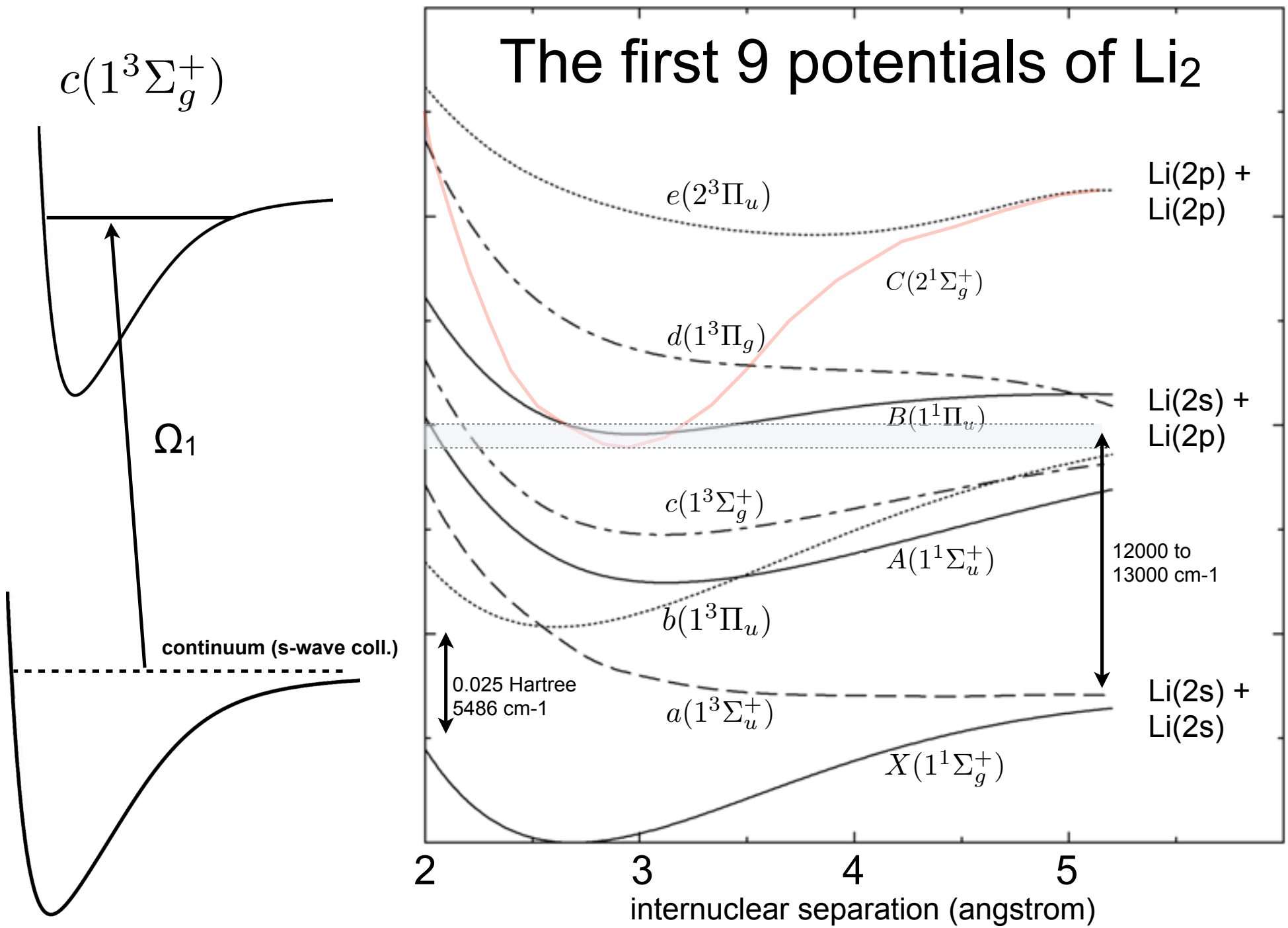
self reference lock

2 Ti:sapphire lasers



When locked to comb:
1) uncertainty on frequency difference < 10 kHz.
2) line width of each Ti:sapphire: ~ 100 kHz
(verified by an independent heterodyne measurement)

1-color PA spectroscopy results



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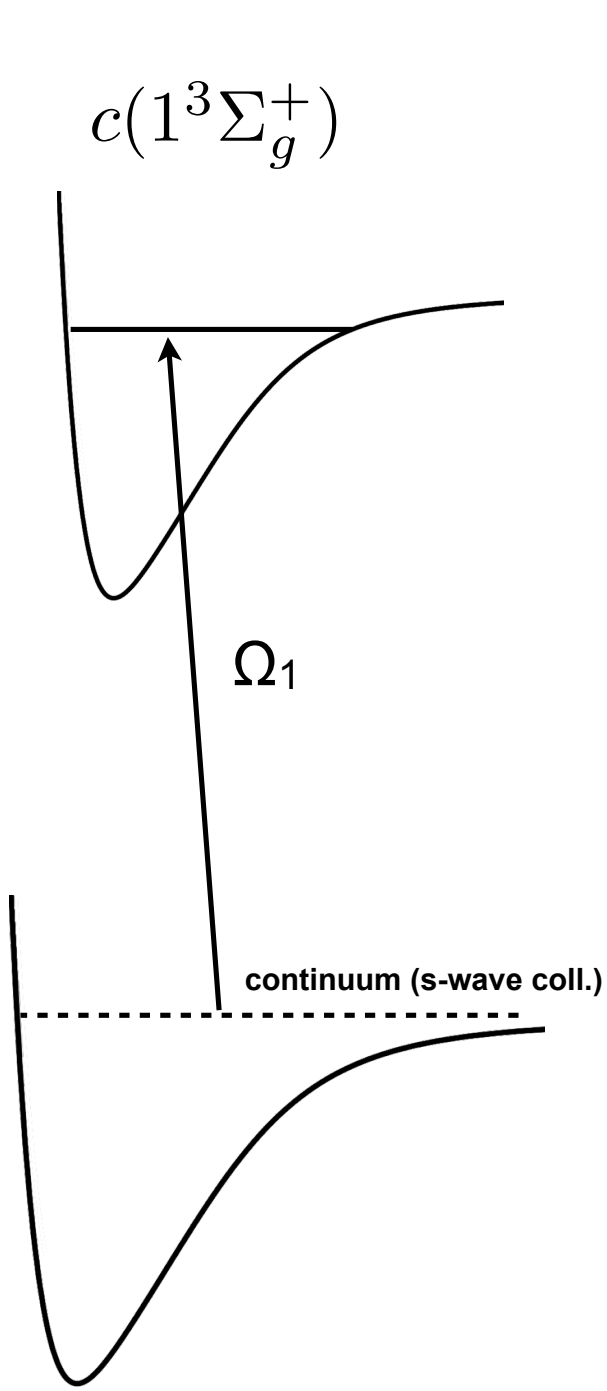

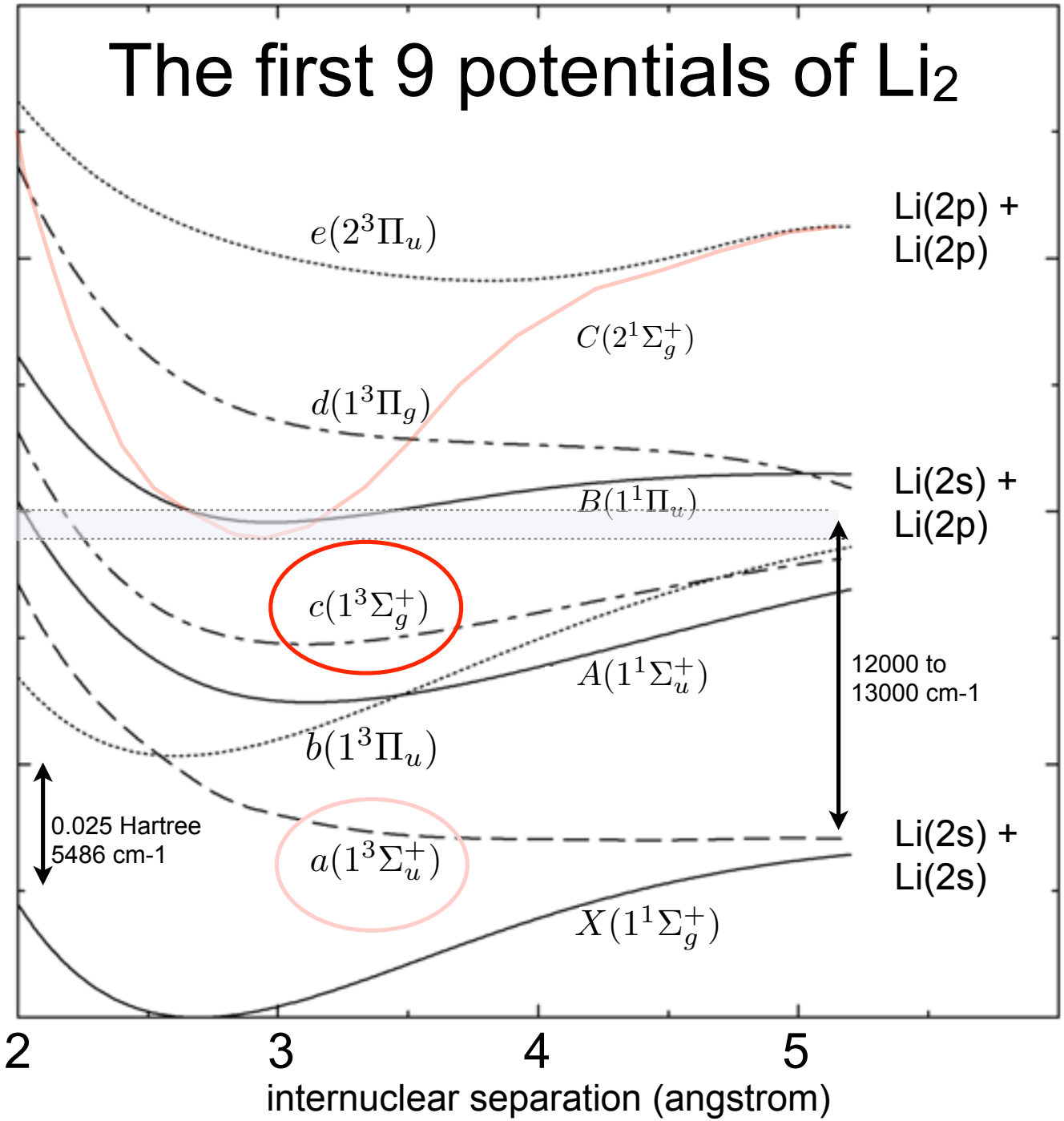

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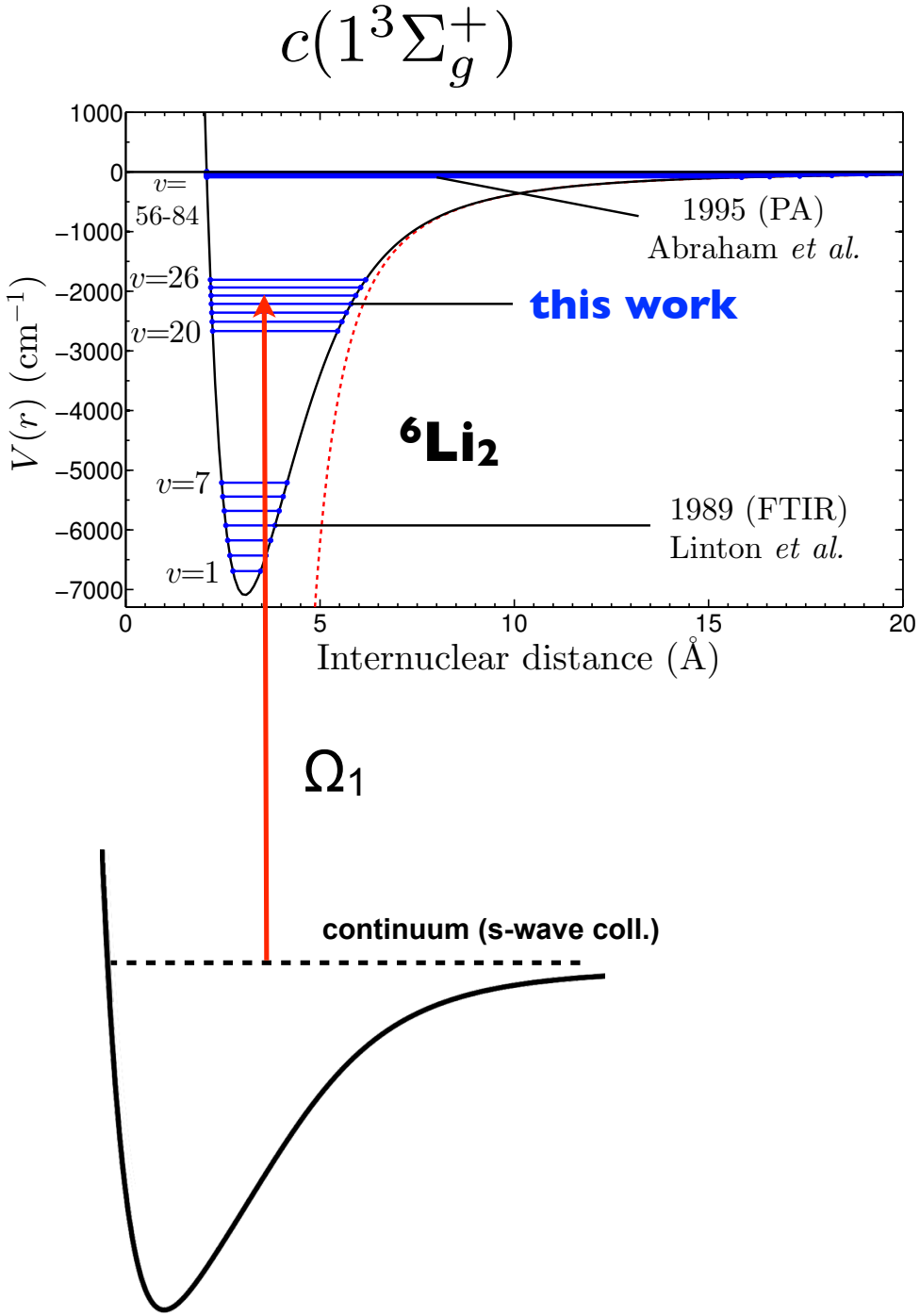

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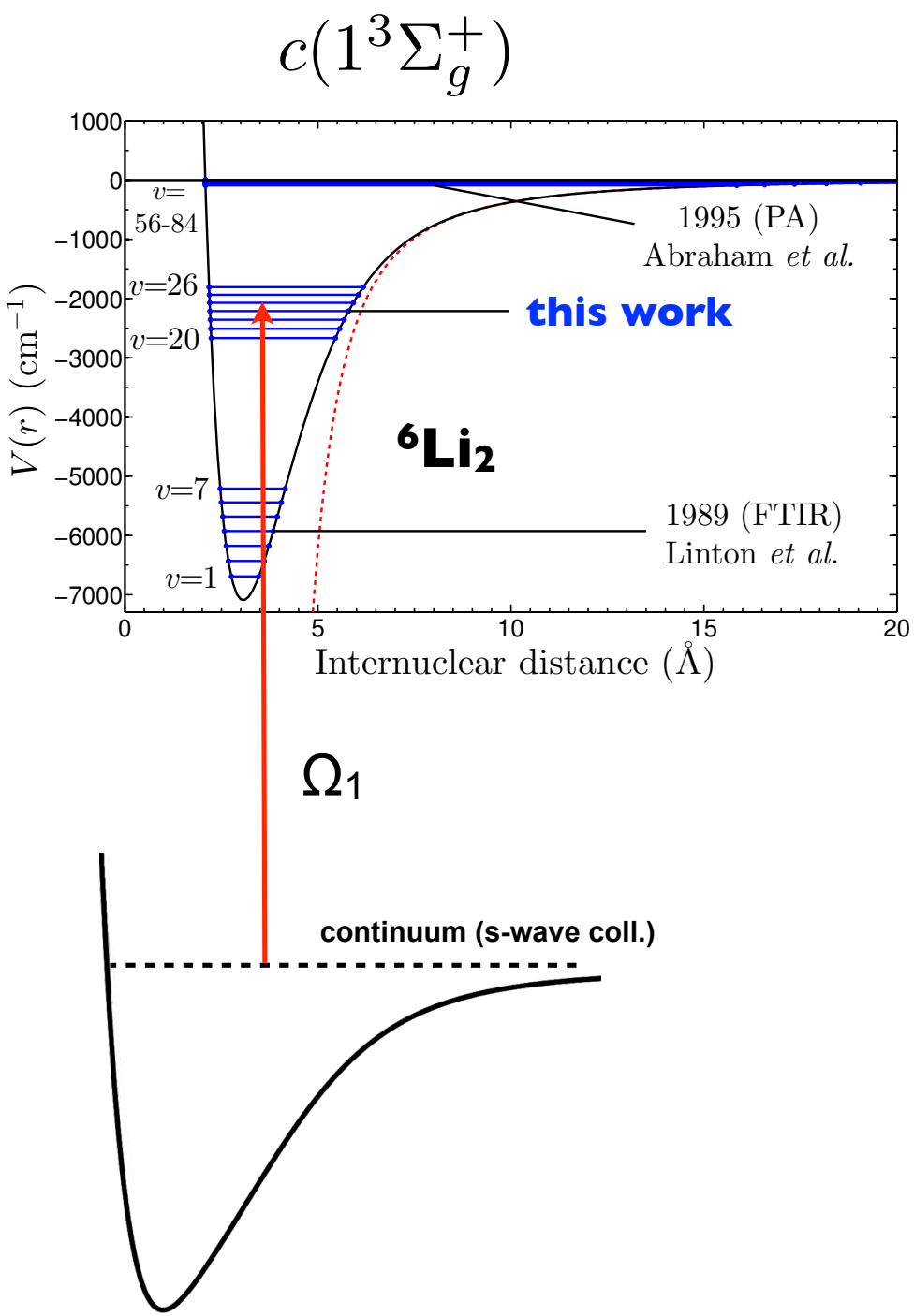
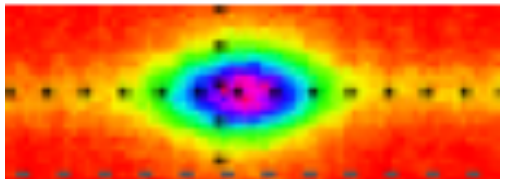


0) To minimize broadening and systematic shifts, we evaporate to very low trap powers and photo-associate a 2-component quantum degenerate Fermi gas ($T/T_F \sim 0.4$)



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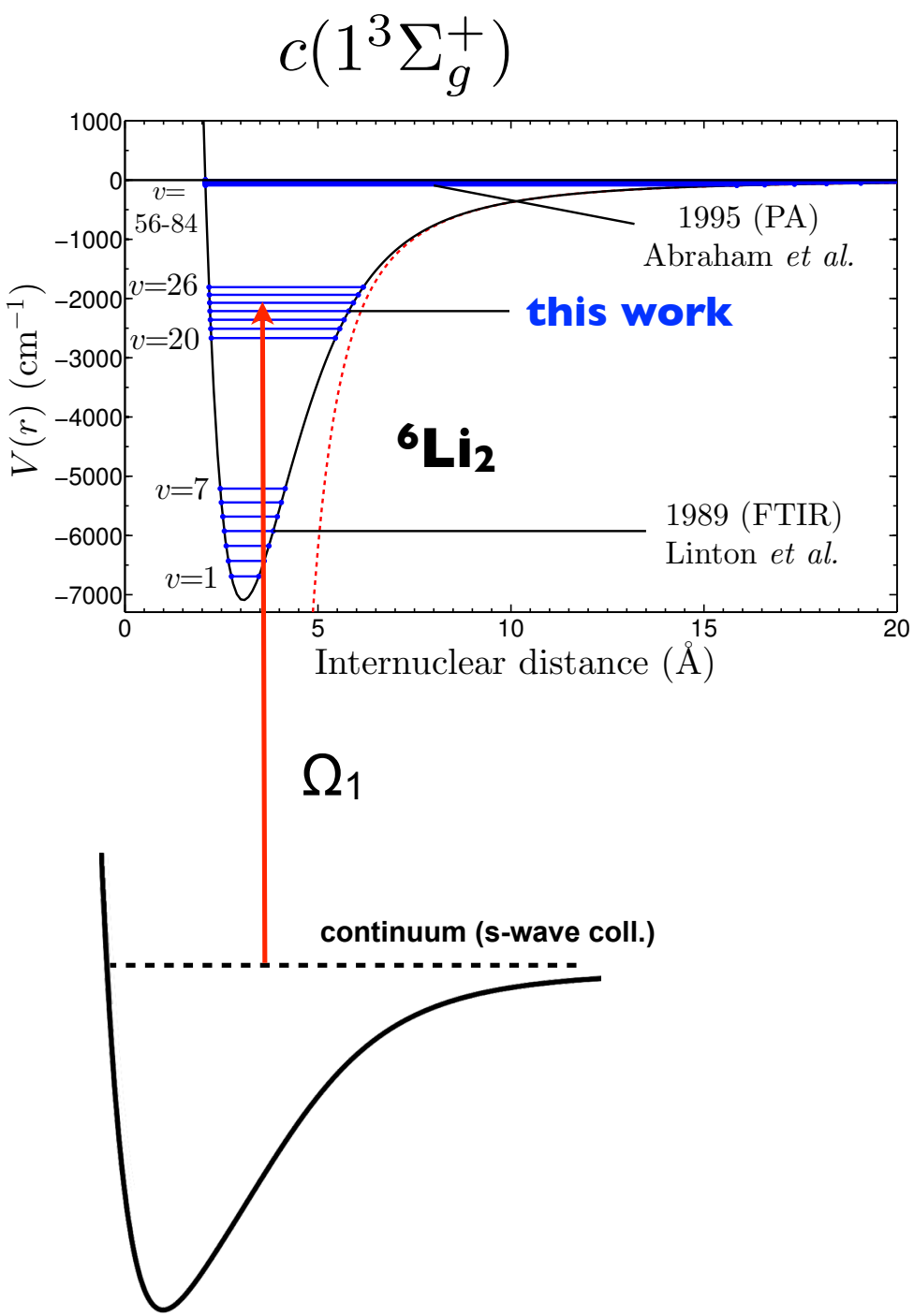

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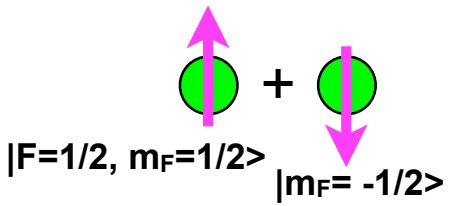
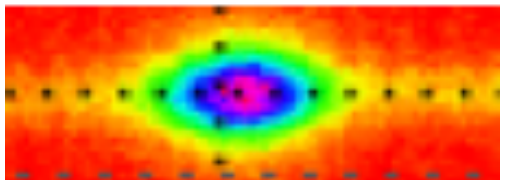
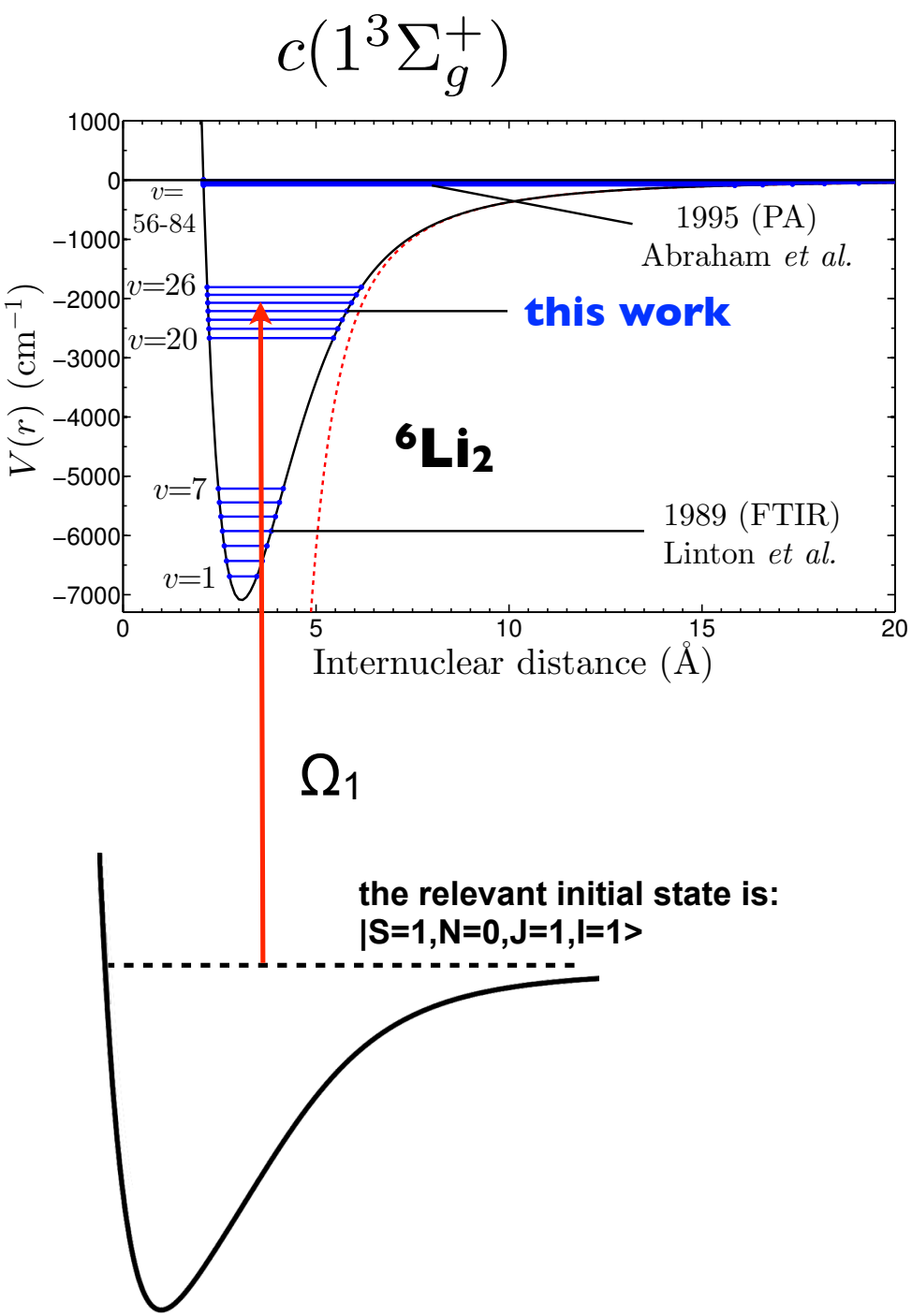


Initial state:
S-wave collision: N=0
Total spin = 0 (G = S+I = 0)
electronic spin = 0, 1



1-color PA spectroscopy results



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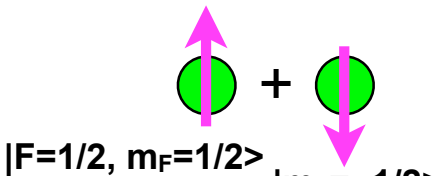
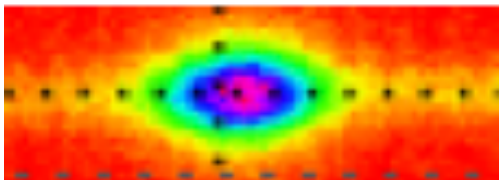


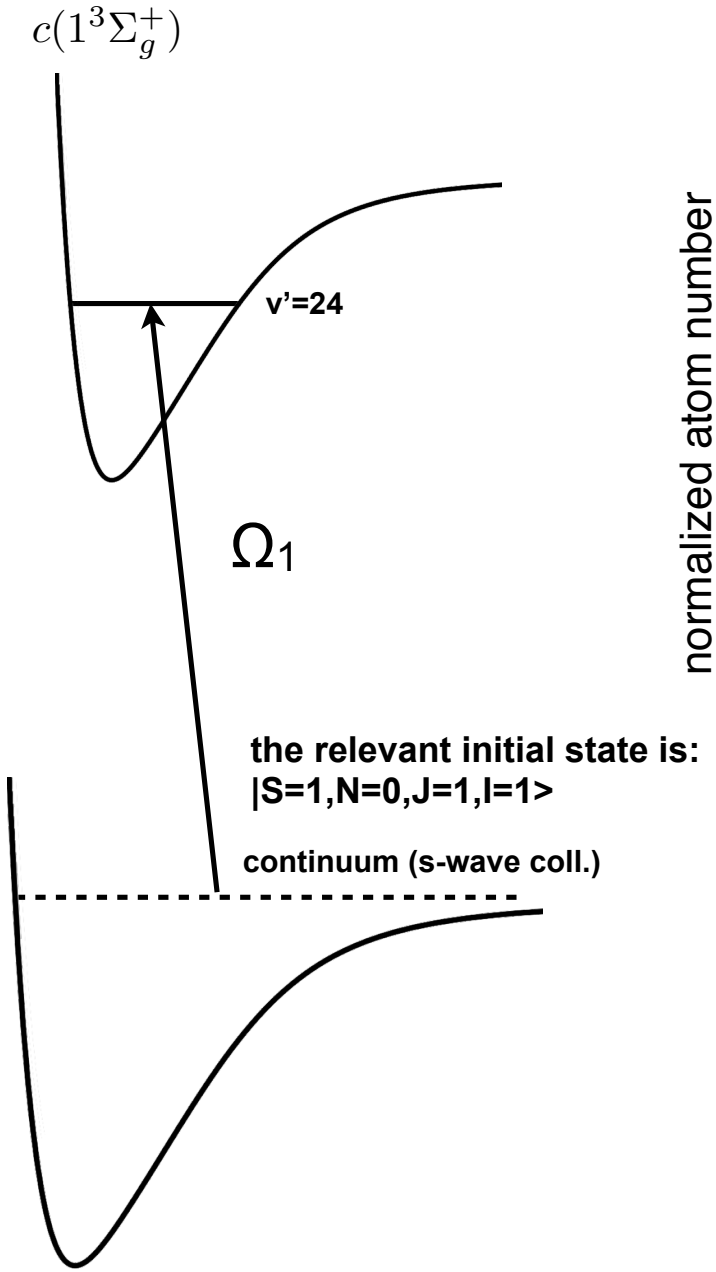
Initial state:
S-wave collision: N=0
Total spin = 0 (G = S+I = 0)
electronic spin = 0, 1

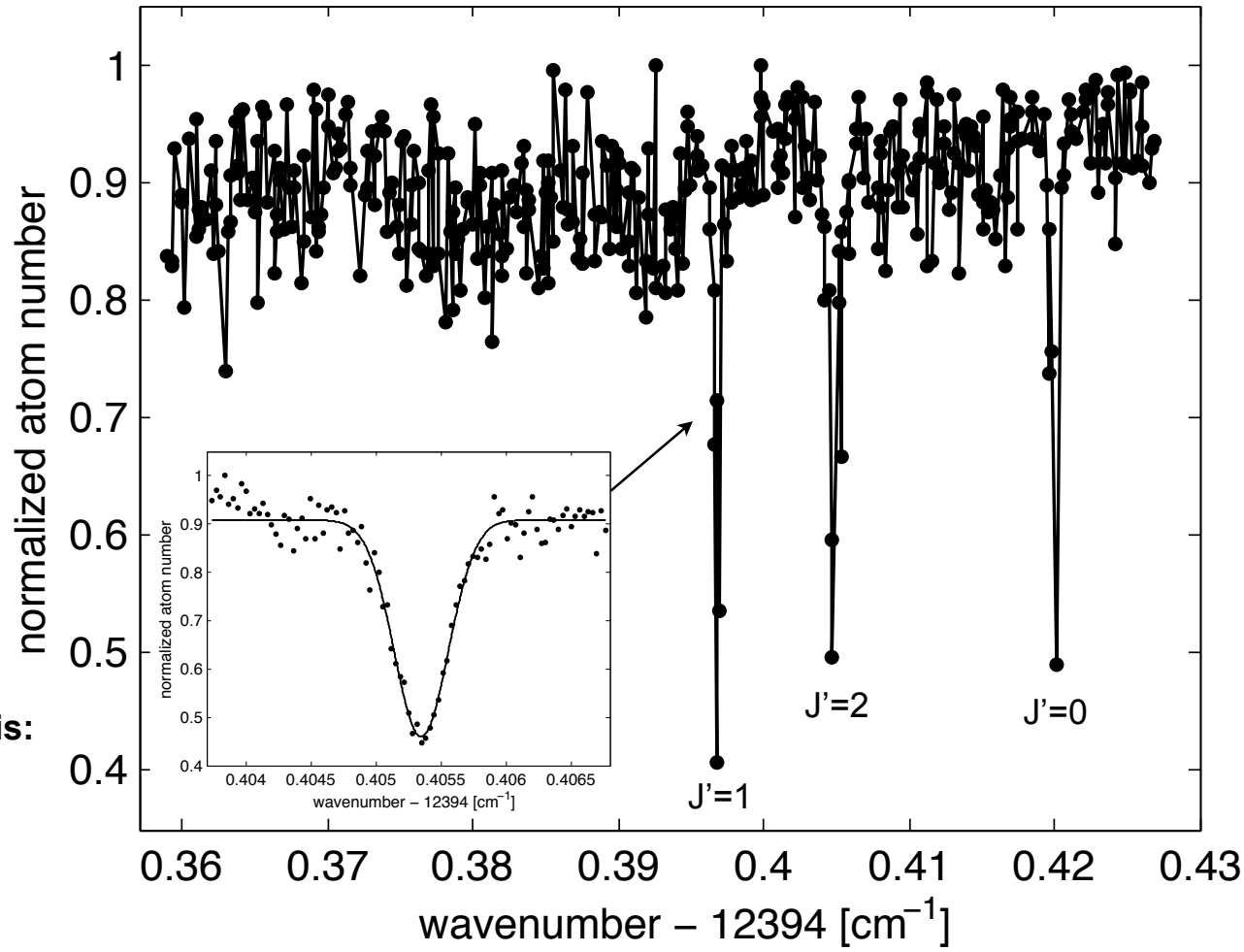
two states are:
|S=0,N=0,J=0,I=0>
|S=1,N=0,J=1,I=1>



1-color PA spectroscopy results

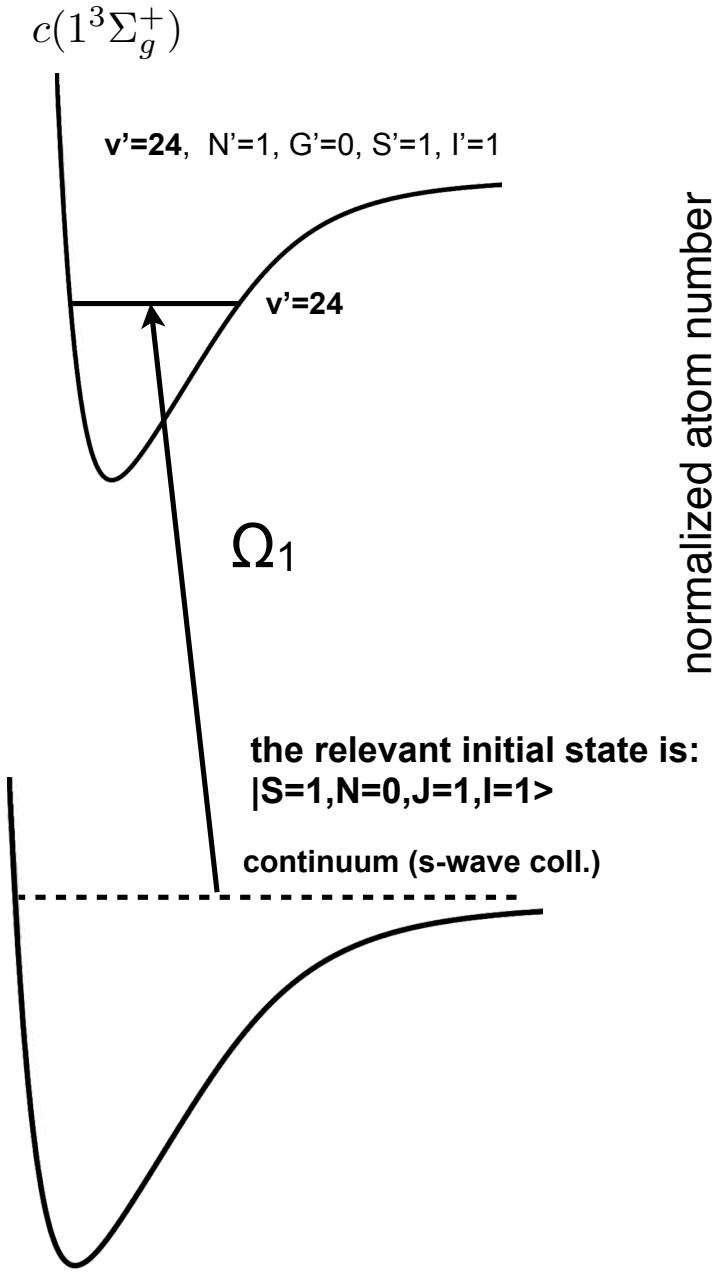
We resolve three features for each vibrational level

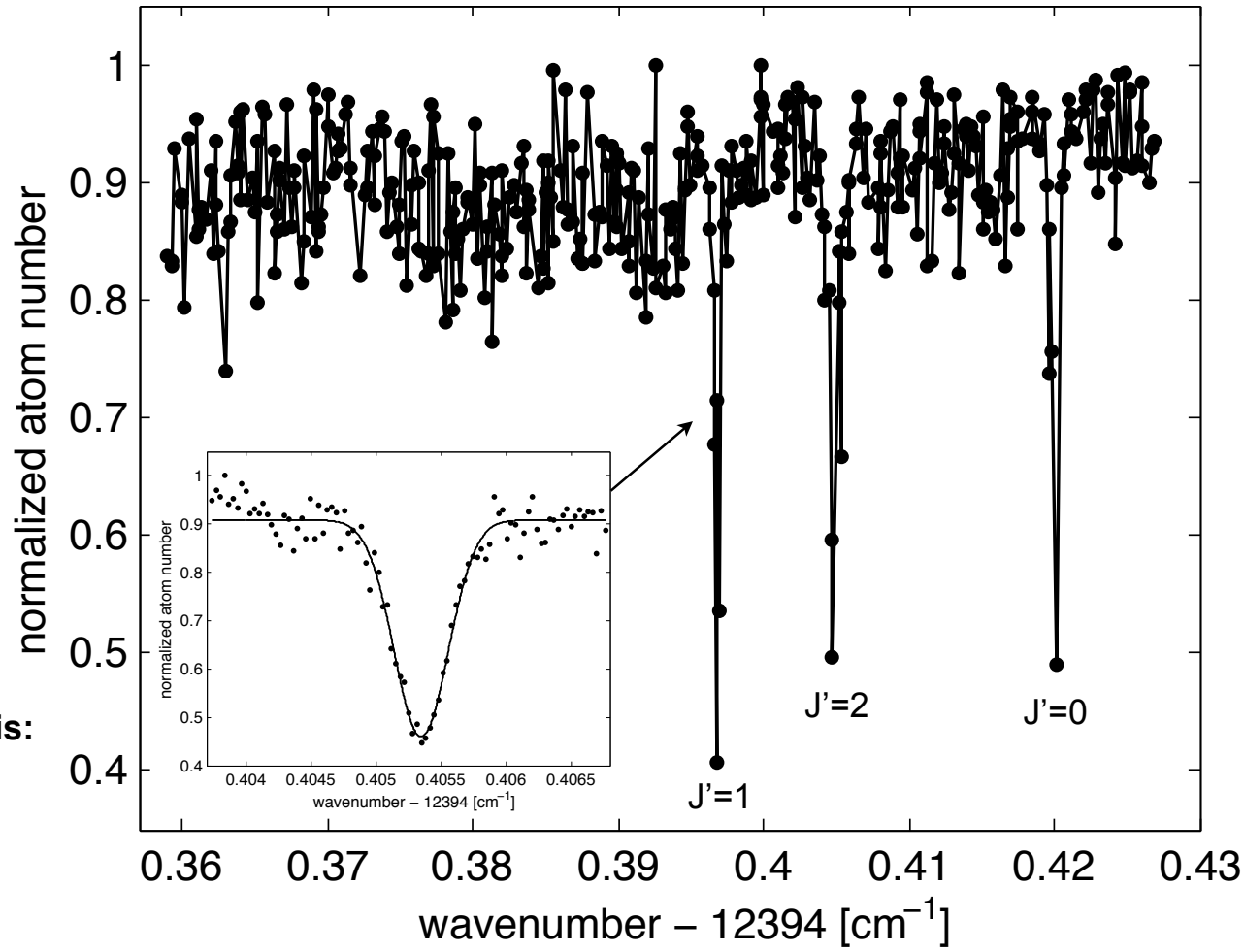




1-color PA spectroscopy results

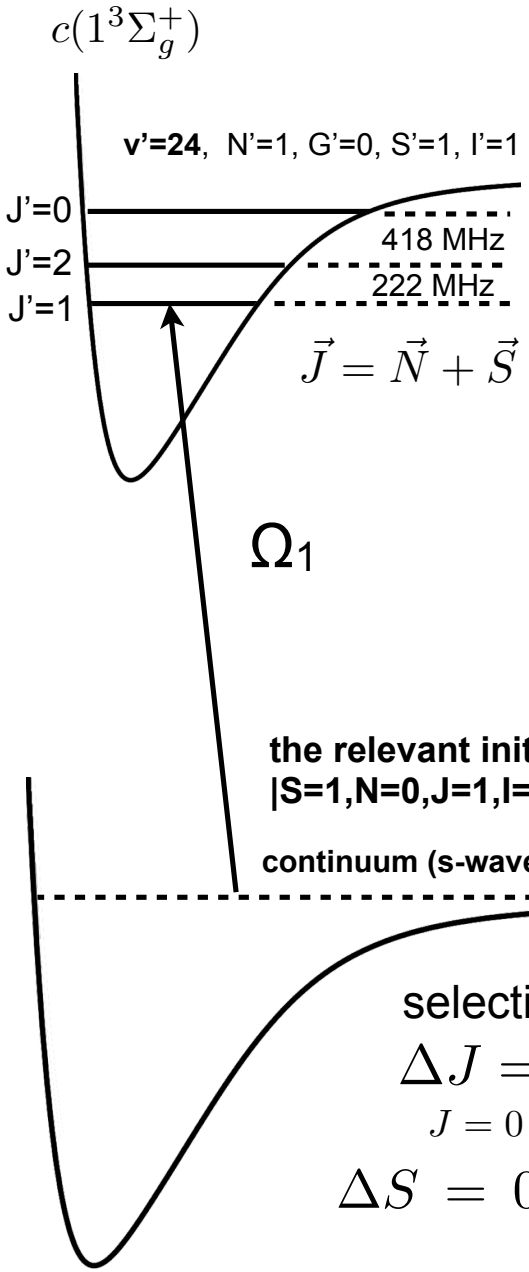
We resolve three features for each vibrational level





1-color PA spectroscopy results

We resolve three features for each vibrational level split by spin-spin and spin-rotation coupling

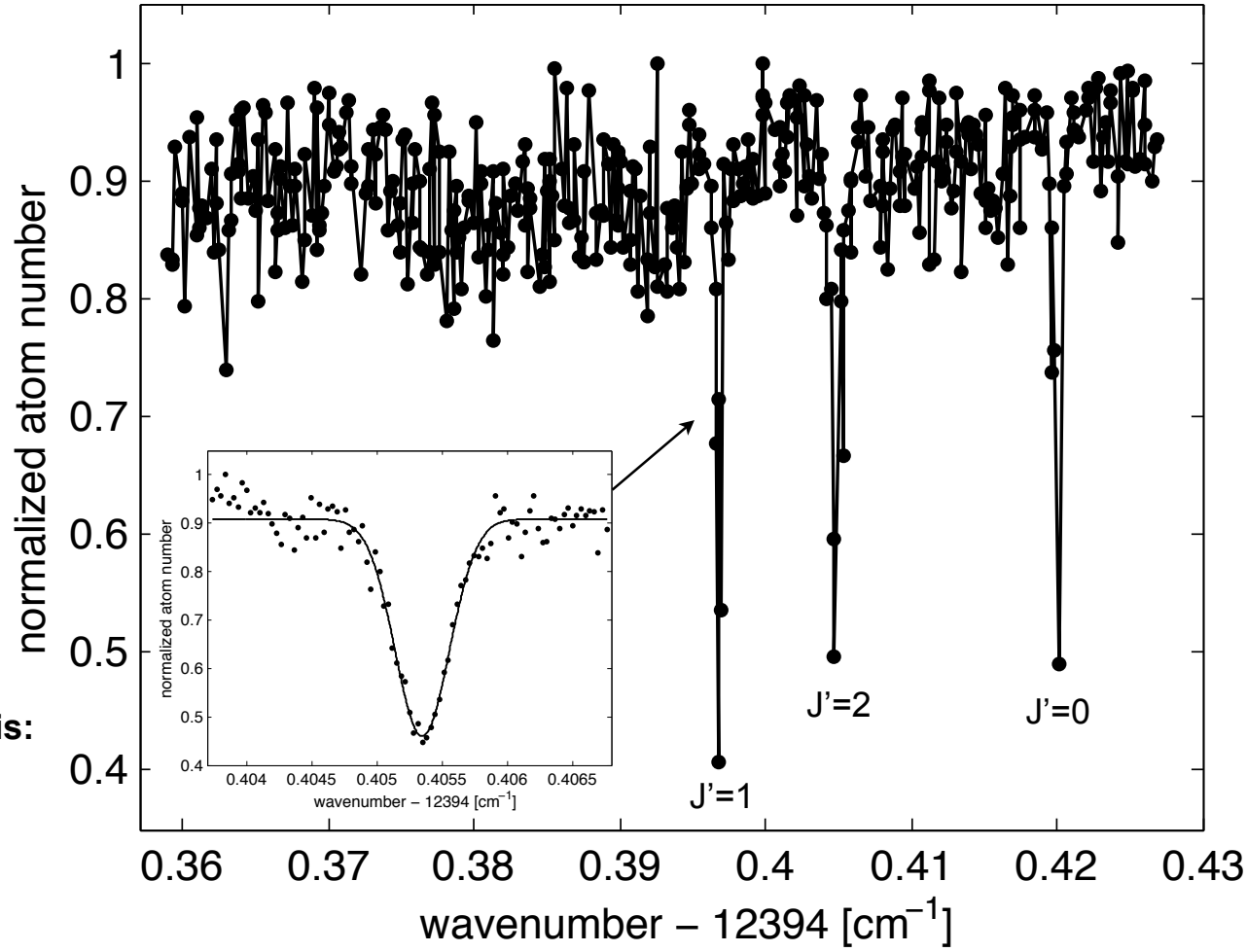


selection rules

$$\Delta J = \pm 1, 0$$

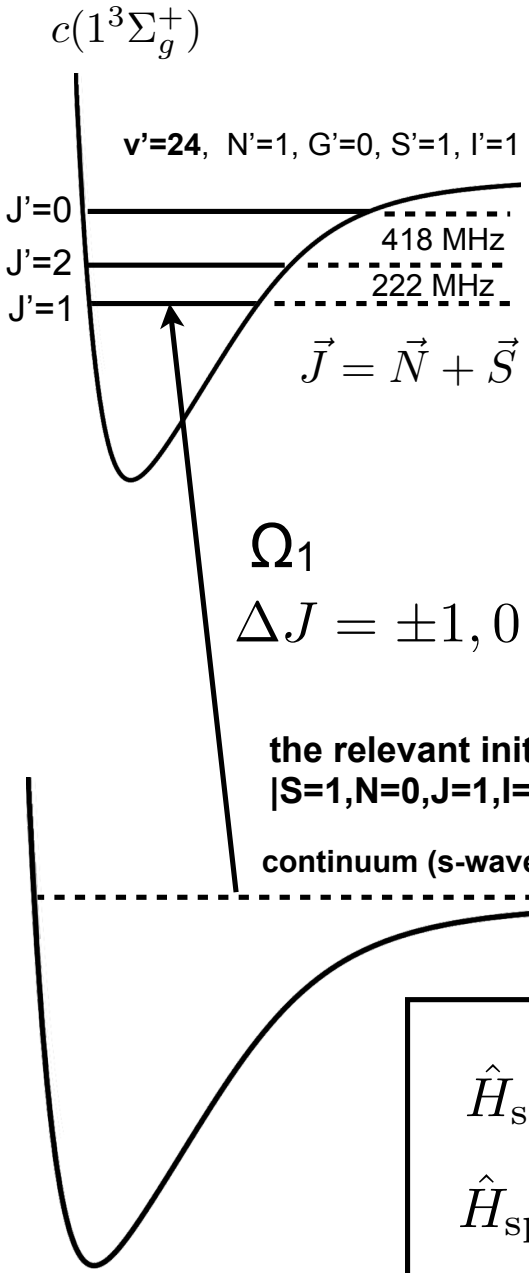
$$J = 0 \nleftrightarrow J = 0$$

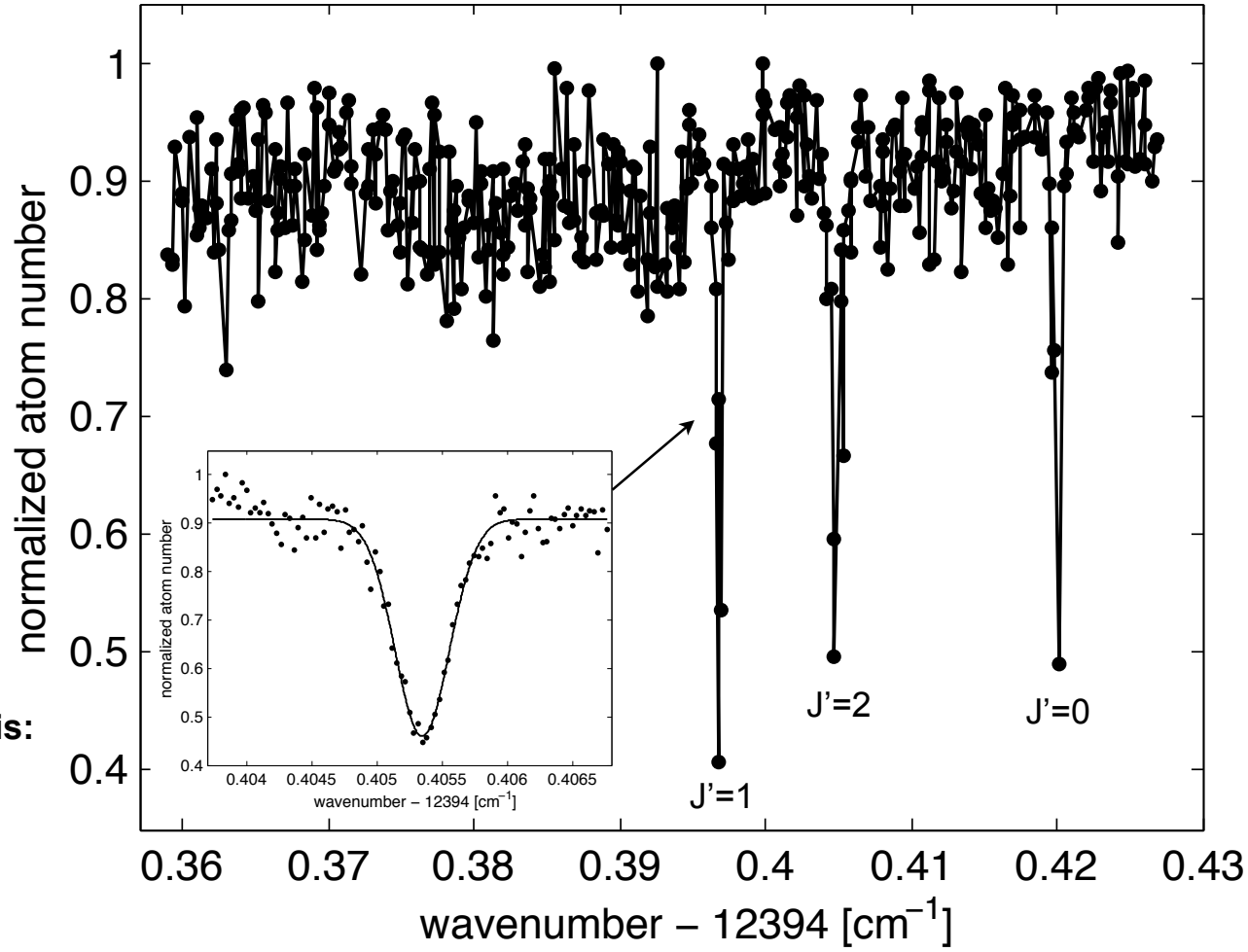
$$\Delta S = 0 \quad \Delta N = \pm 1$$



1-color PA spectroscopy results

We resolve three features for each vibrational level split by spin-spin and spin-rotation coupling



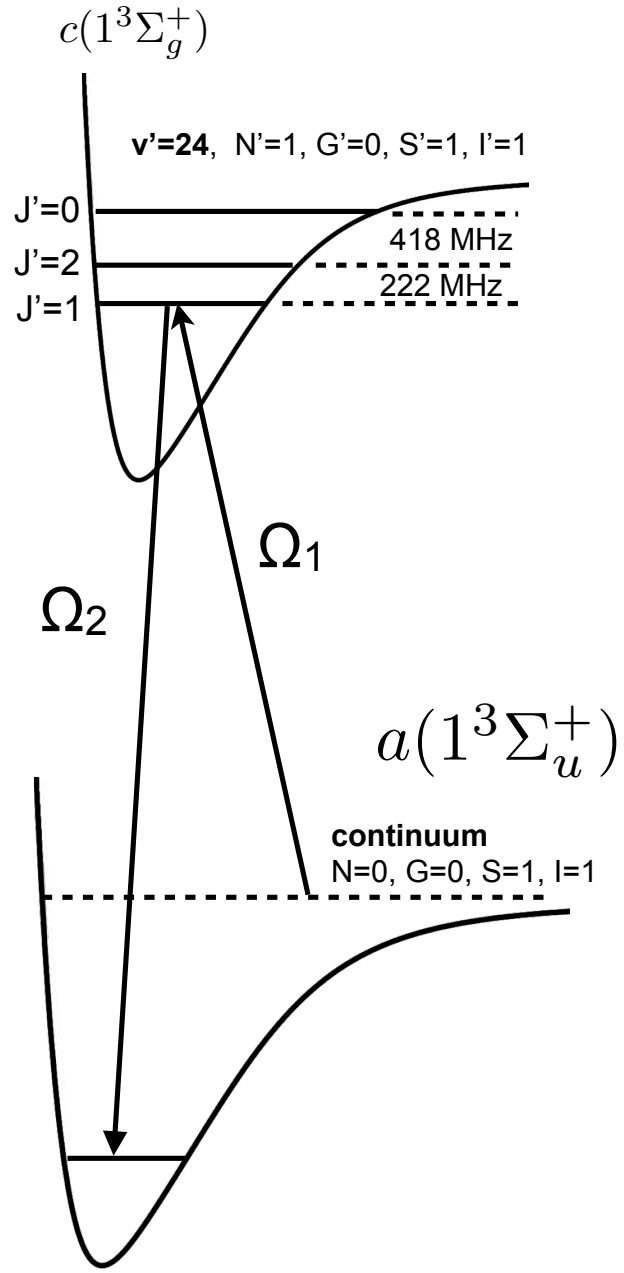


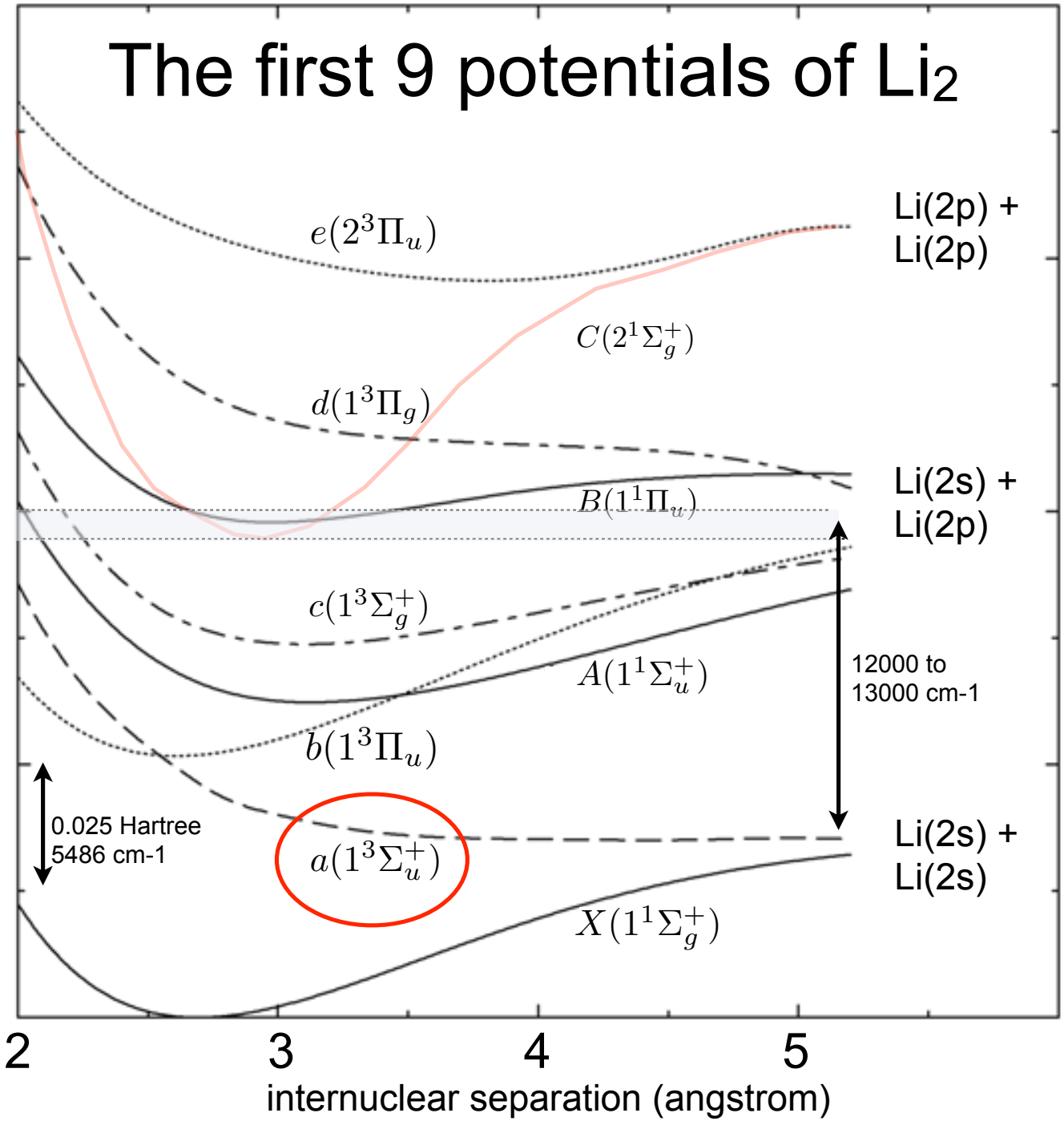
$$\hat{H}_{\mathrm{spin-spin}} = 2\lambda_v[\hat{S}_z^2 - \hat{S}^2/3]$$

$$\hat{H}_{\mathrm{spin-rot}} = \gamma_v \vec{N} \cdot \vec{S}$$

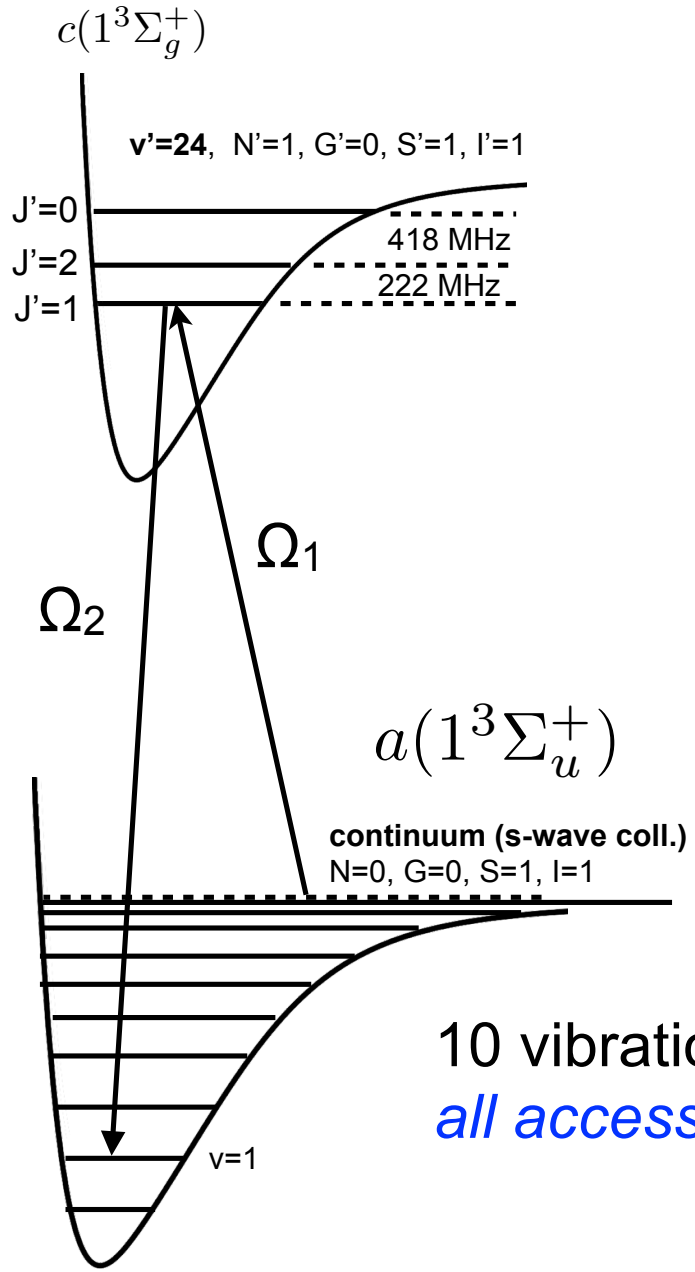
v'	λ_v (MHz)	γ_v (MHz)
20	-348.2	-14.5
21	-339.4	-14.5
22	-331.1	-14.7
23	-321.7	-14.2
24	-312.2	-14.4
25	-309.6	-14.0
26	-294.3	-14.3

2-color PA spectroscopy : ground electronic state spectroscopy





2-color PA spectroscopy : ground electronic state spectroscopy



Lowest triplet state Li_2

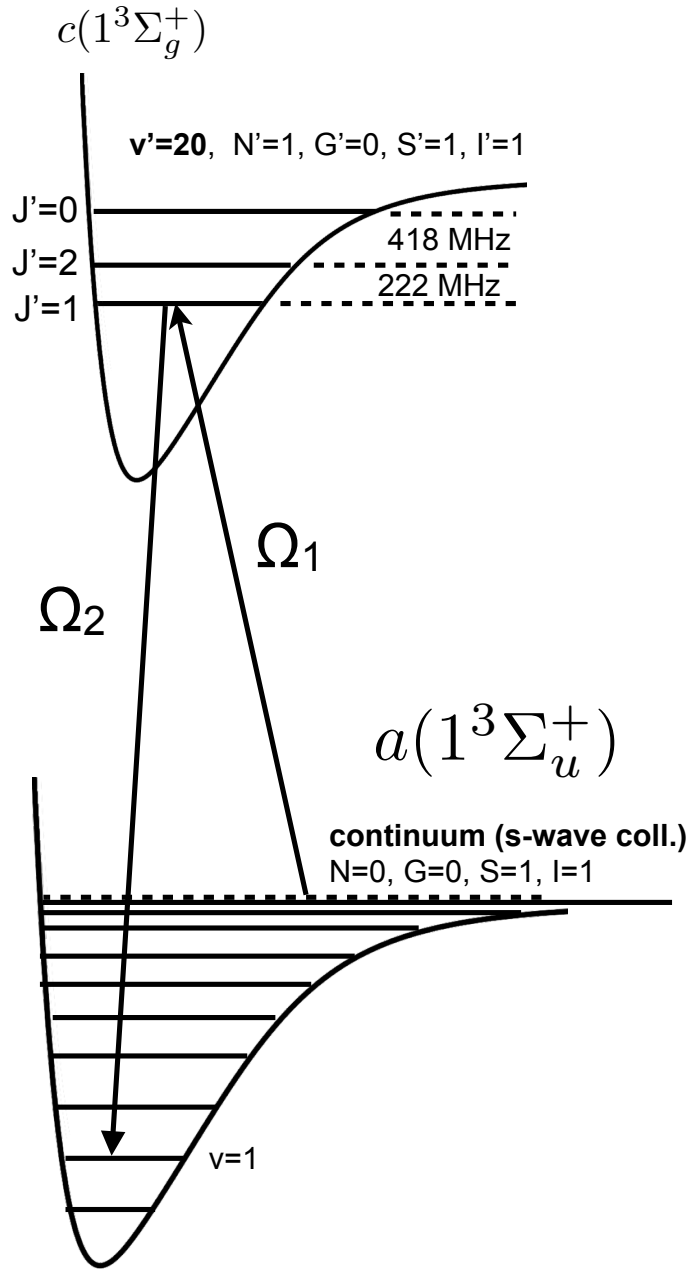
Motivations:

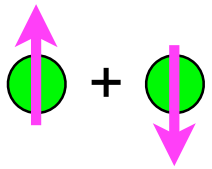
1) Technical: we can access with our lasers.
2) It has a magnetic moment (Molecular FRs !)
3) Make a BEC of ground state molecules
4) Measure the "Spin blockade"
5) Study collision properties of ultra-cold super-rotors (collaboration with Valery Milner)
6) Stepping stone for making triplet LiRb

10 vibrational levels

all accessible with our laser system

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned

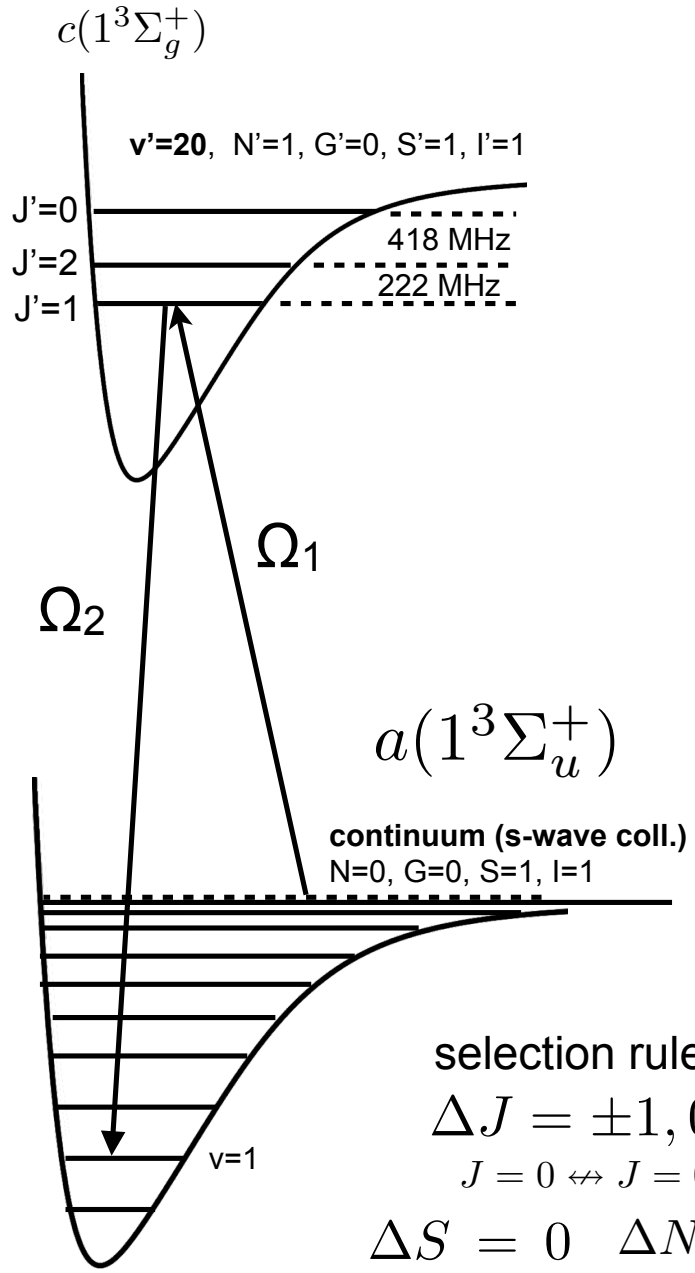


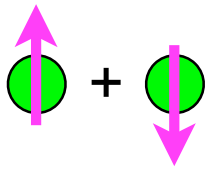


the relevant initial state is:
|S=1,N=0,J=1,I=1>

the intermediate state is:
|S=1,N=1,J=1,I=1>

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned





the relevant initial state is:
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the intermediate state is:
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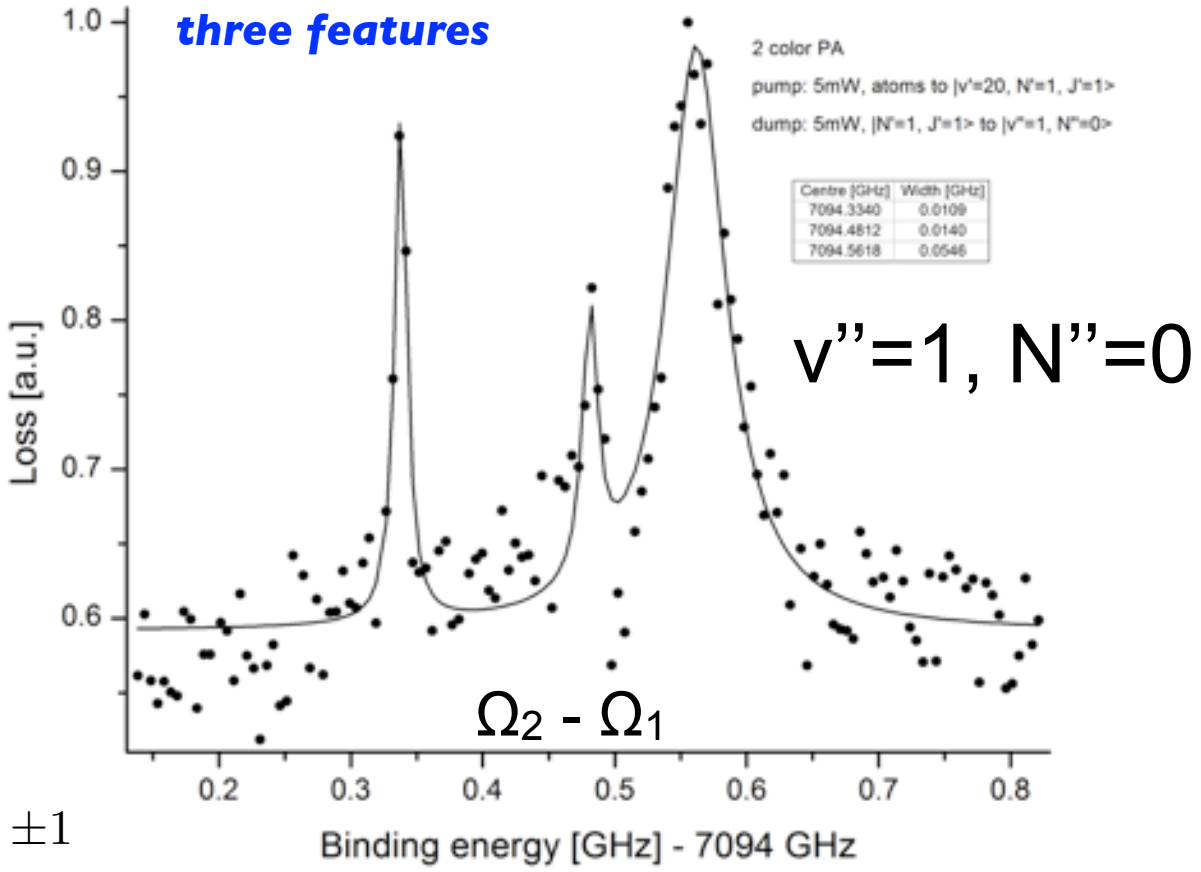
selection rules

$$\Delta J = \pm 1, 0$$

$$J = 0 \nleftrightarrow J = 0$$

$$\Delta S = 0 \quad \Delta N = \pm 1$$



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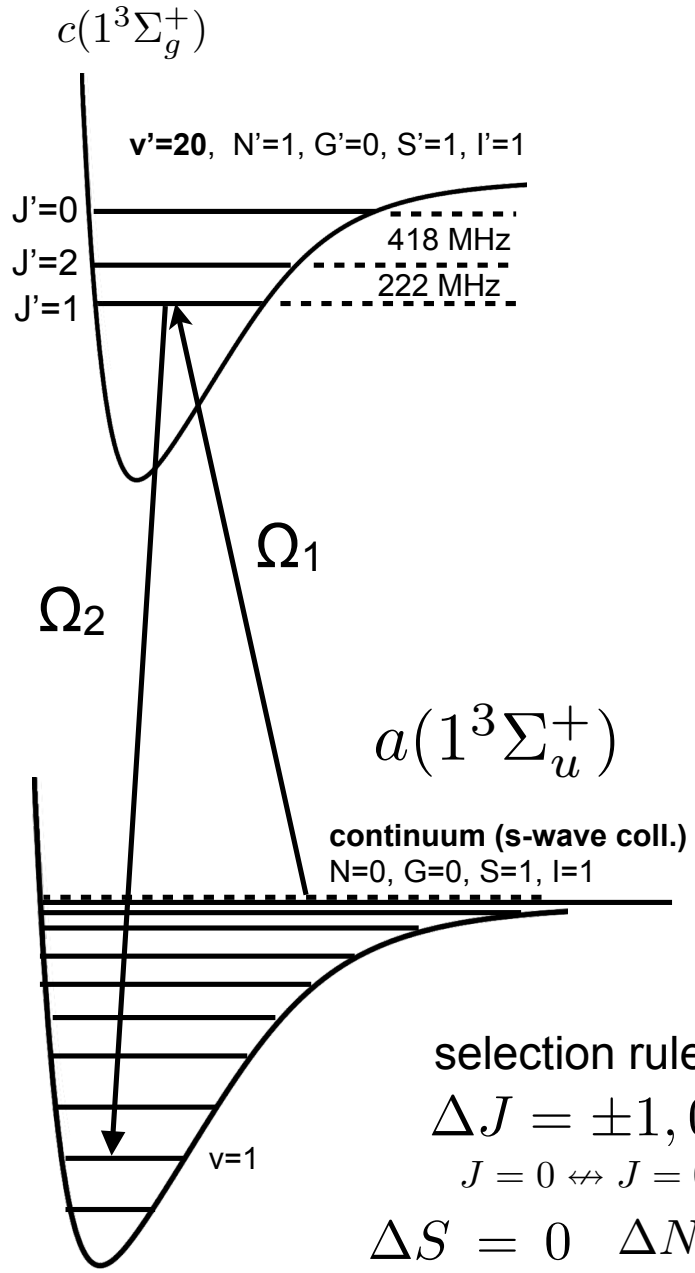

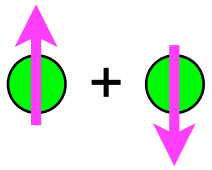

the relevant initial state is:
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the intermediate state is:
|S=1,N=1,J=1,I=1>

the final state is: *only one value of J*
|S=1,N=0,J=1,I=1>

selection rules

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$$J = 0 \nleftrightarrow J = 0$$

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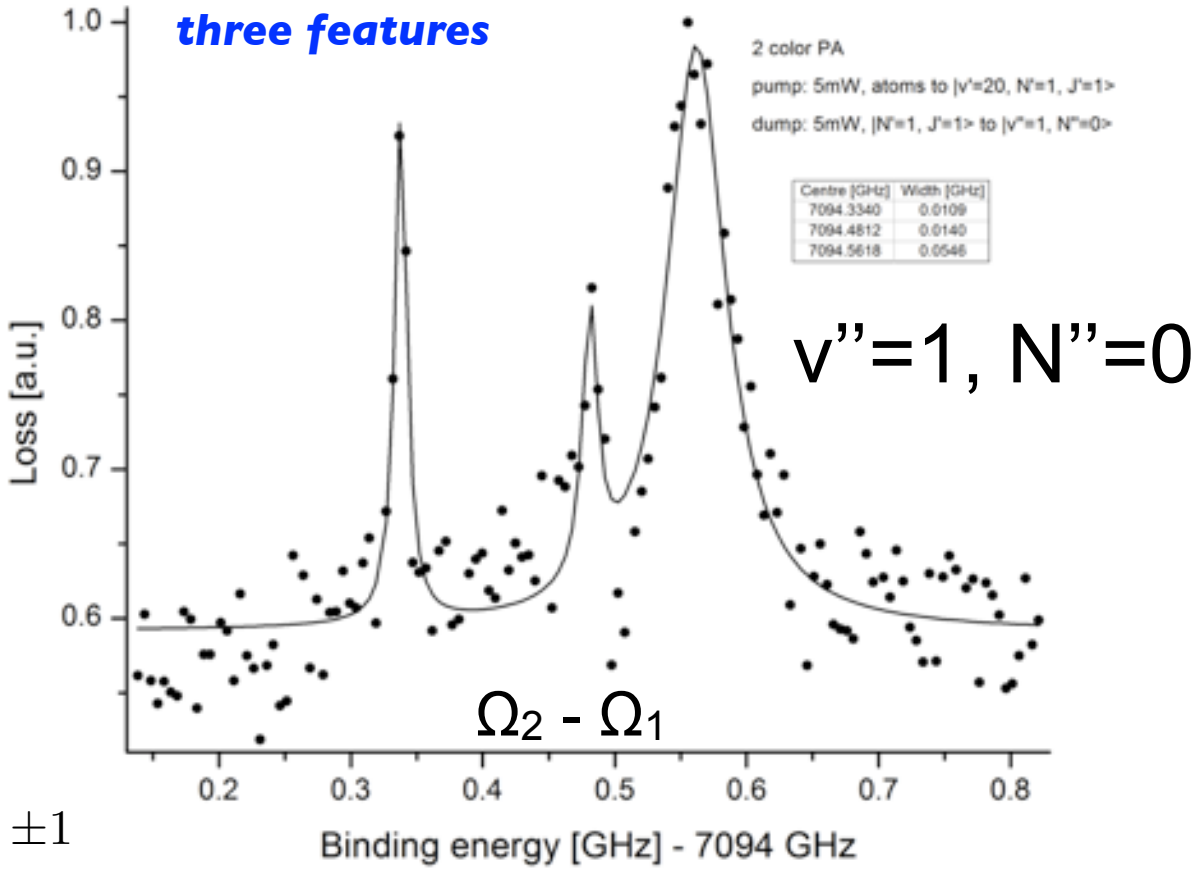

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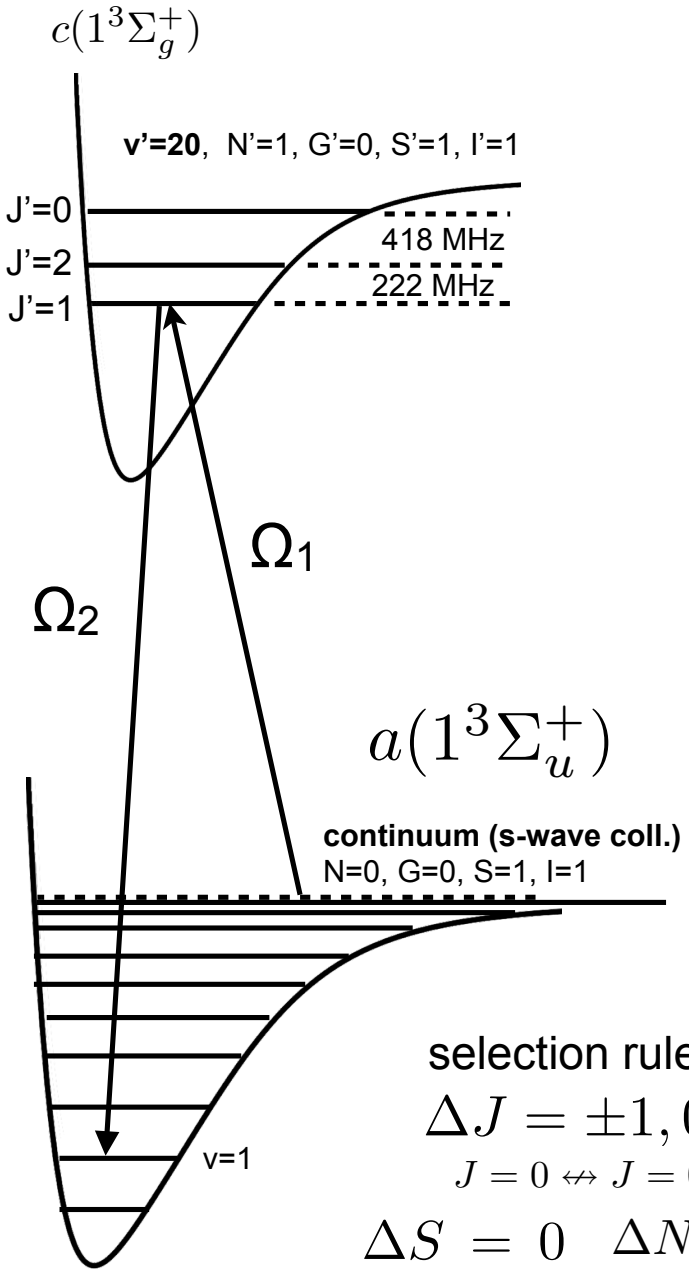

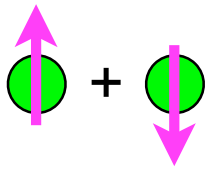

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the final state is:
|S=1,N=0,J=1,I=1>

only one value of J

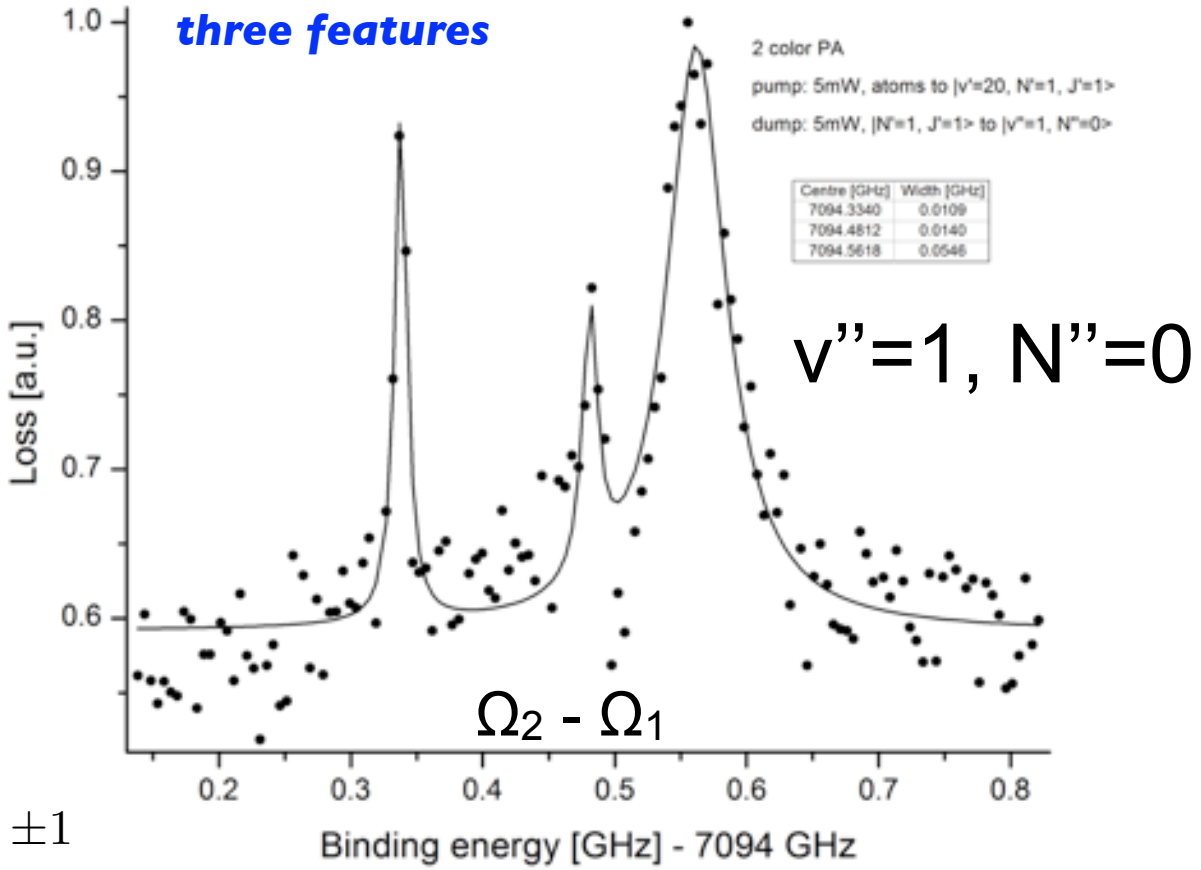
F = I + S = 0, 1, 2 (expect three hyperfine levels)

selection rules

$$\Delta J = \pm 1, 0$$

$$J = 0 \nleftrightarrow J = 0$$

$$\Delta S = 0 \quad \Delta N = \pm 1$$



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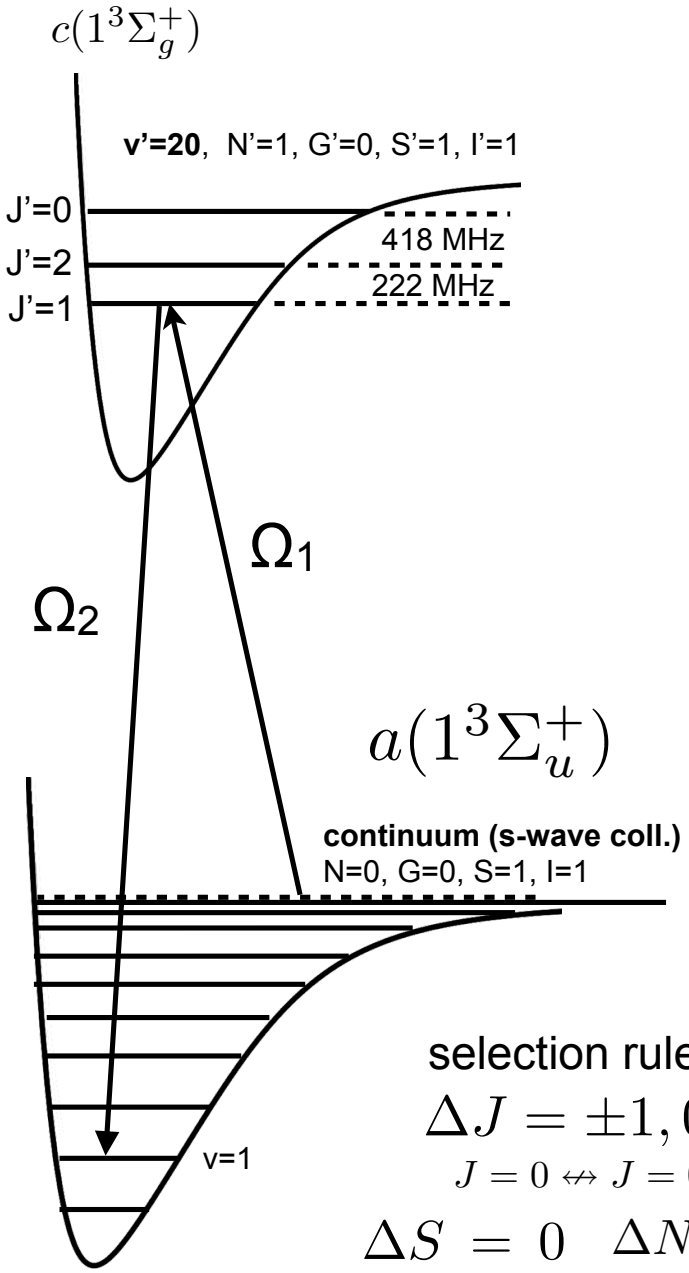

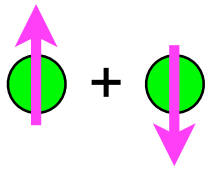

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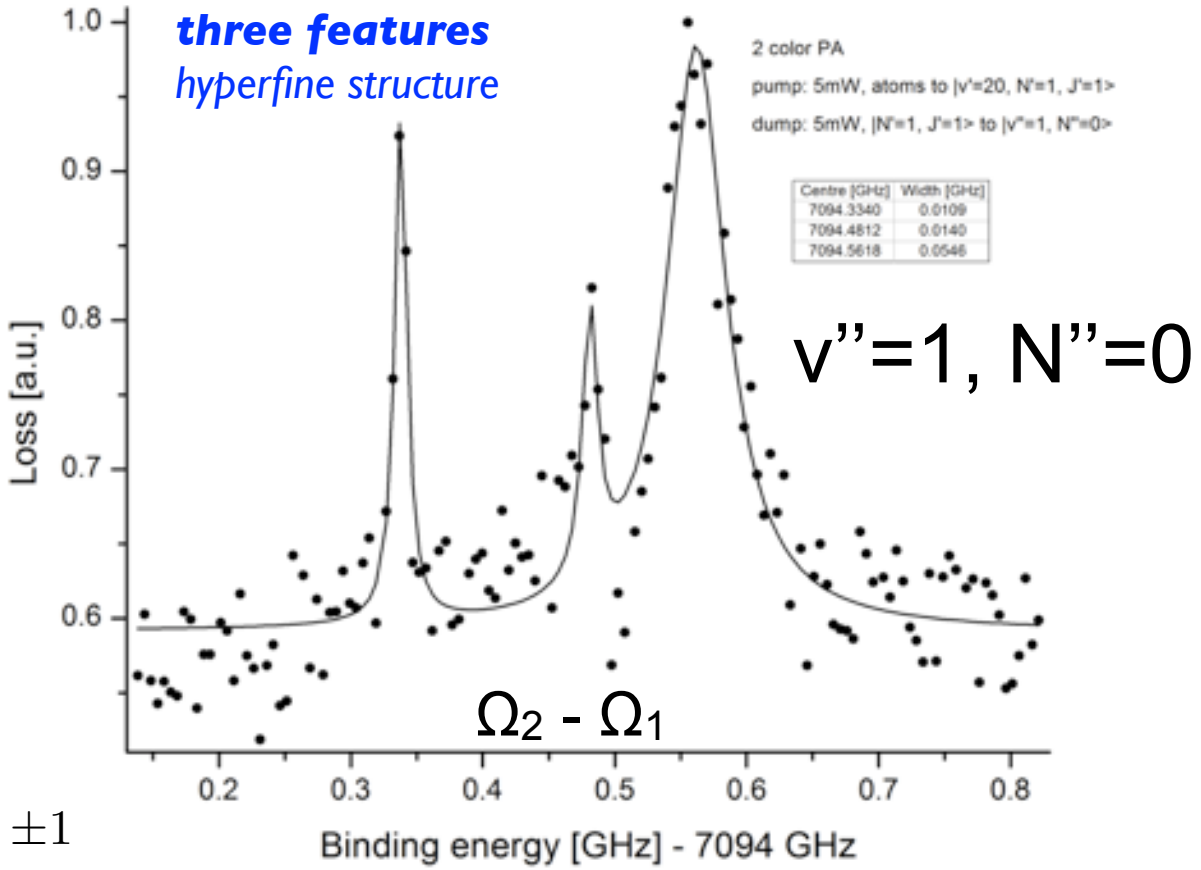

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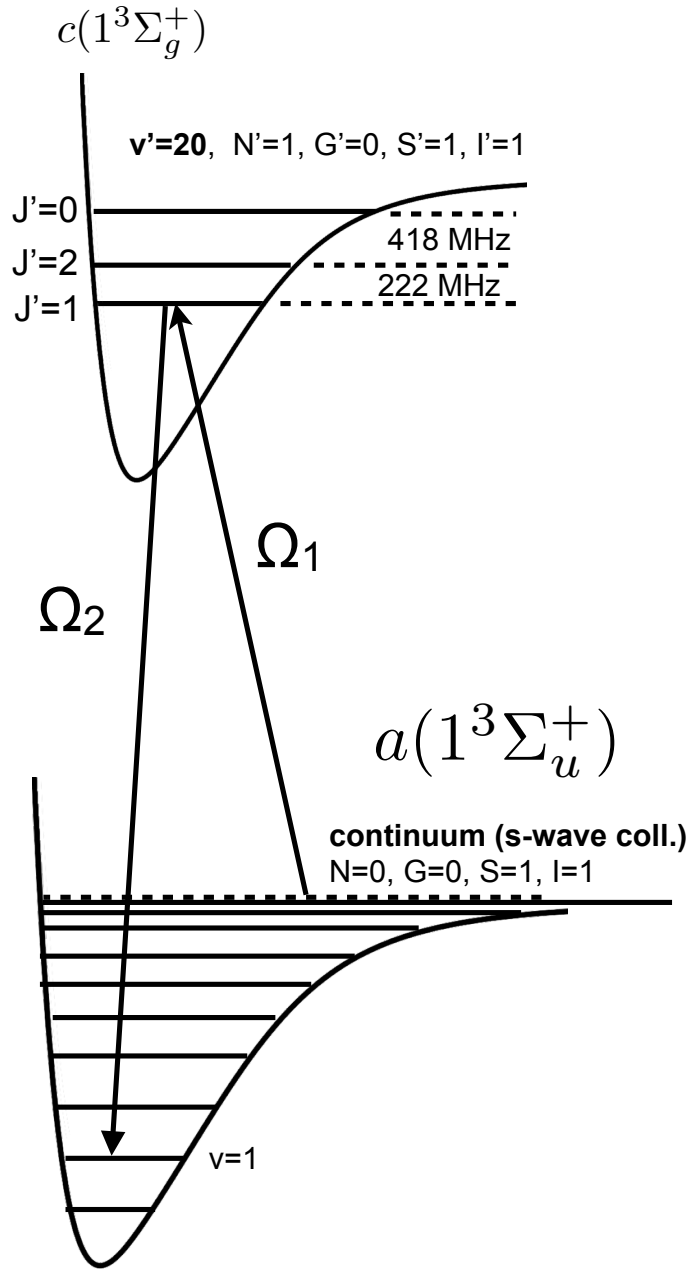

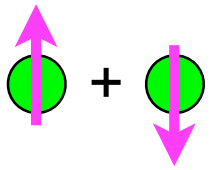

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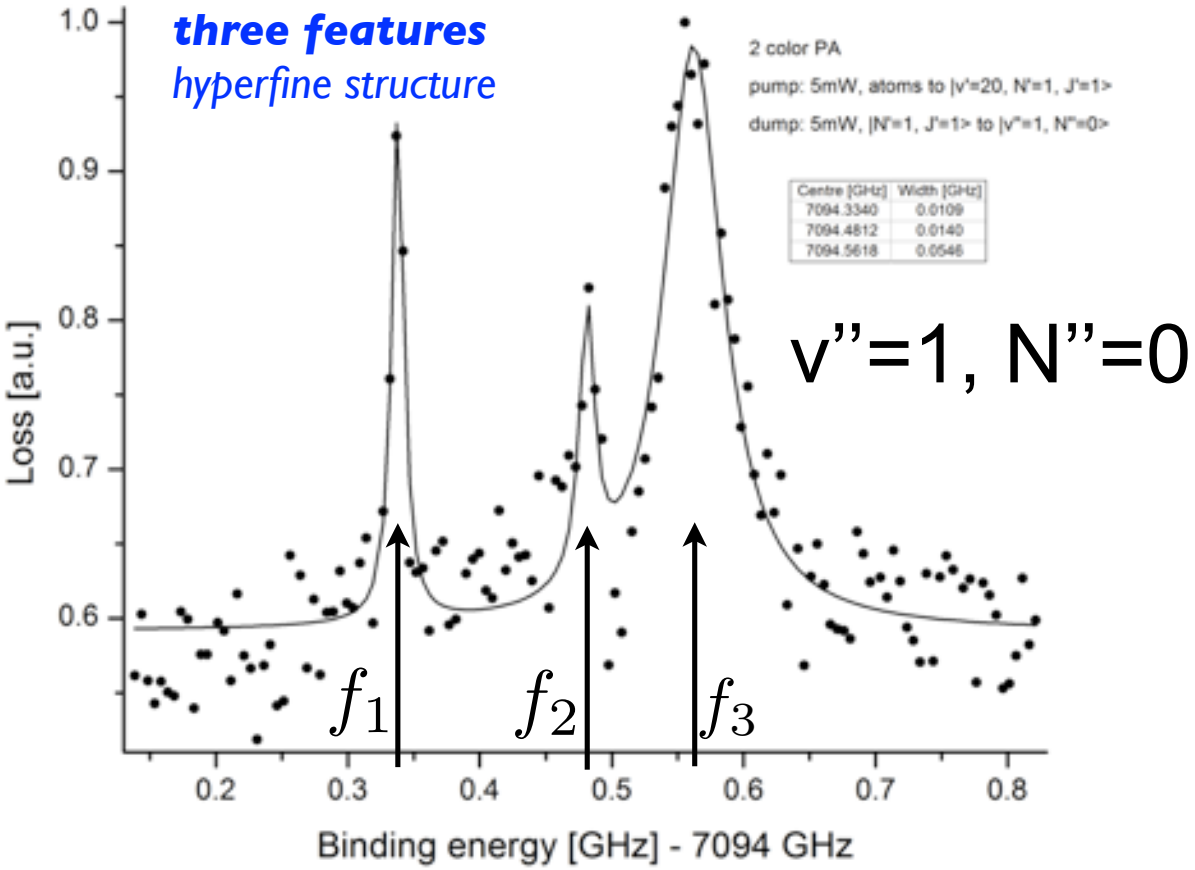
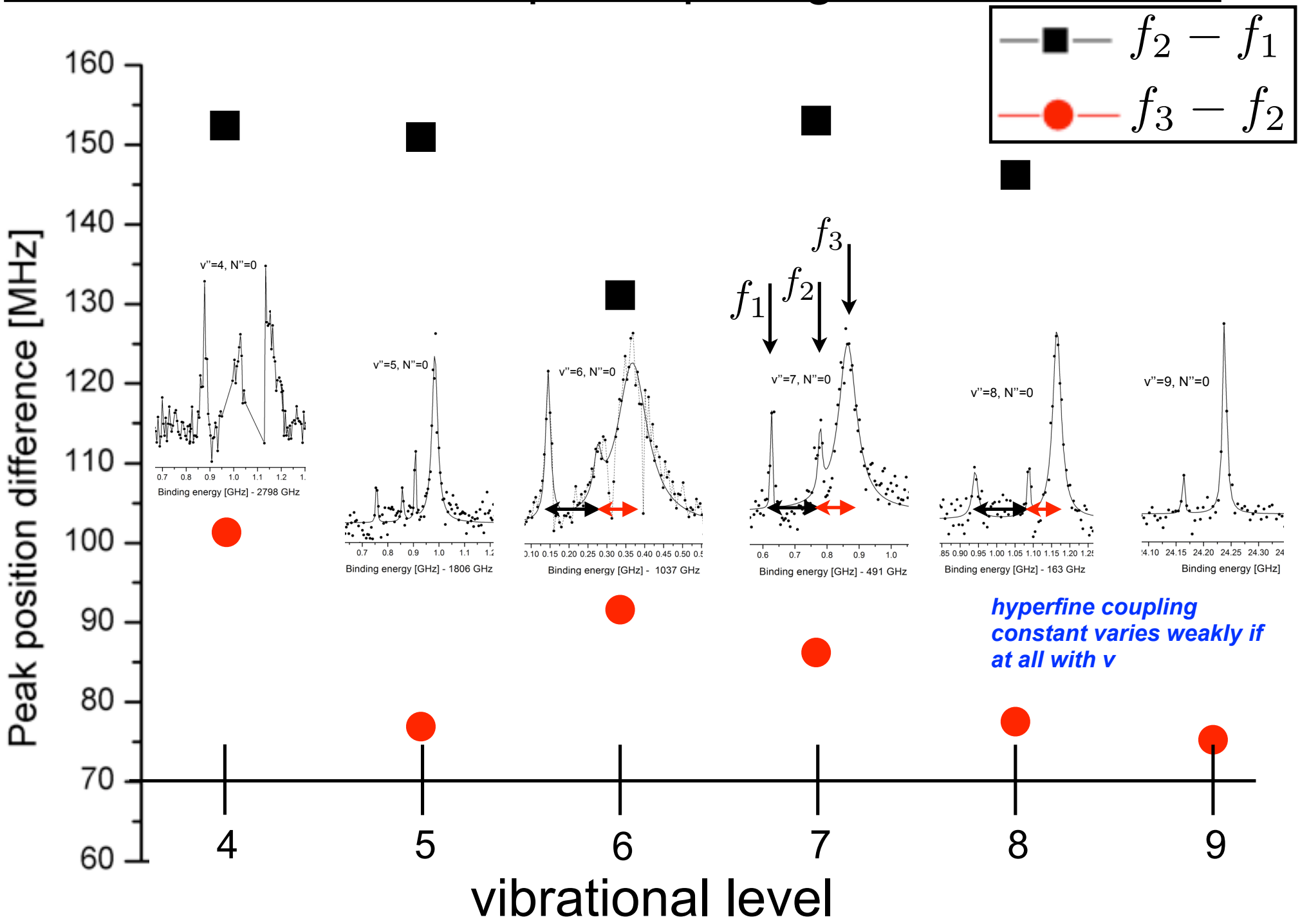
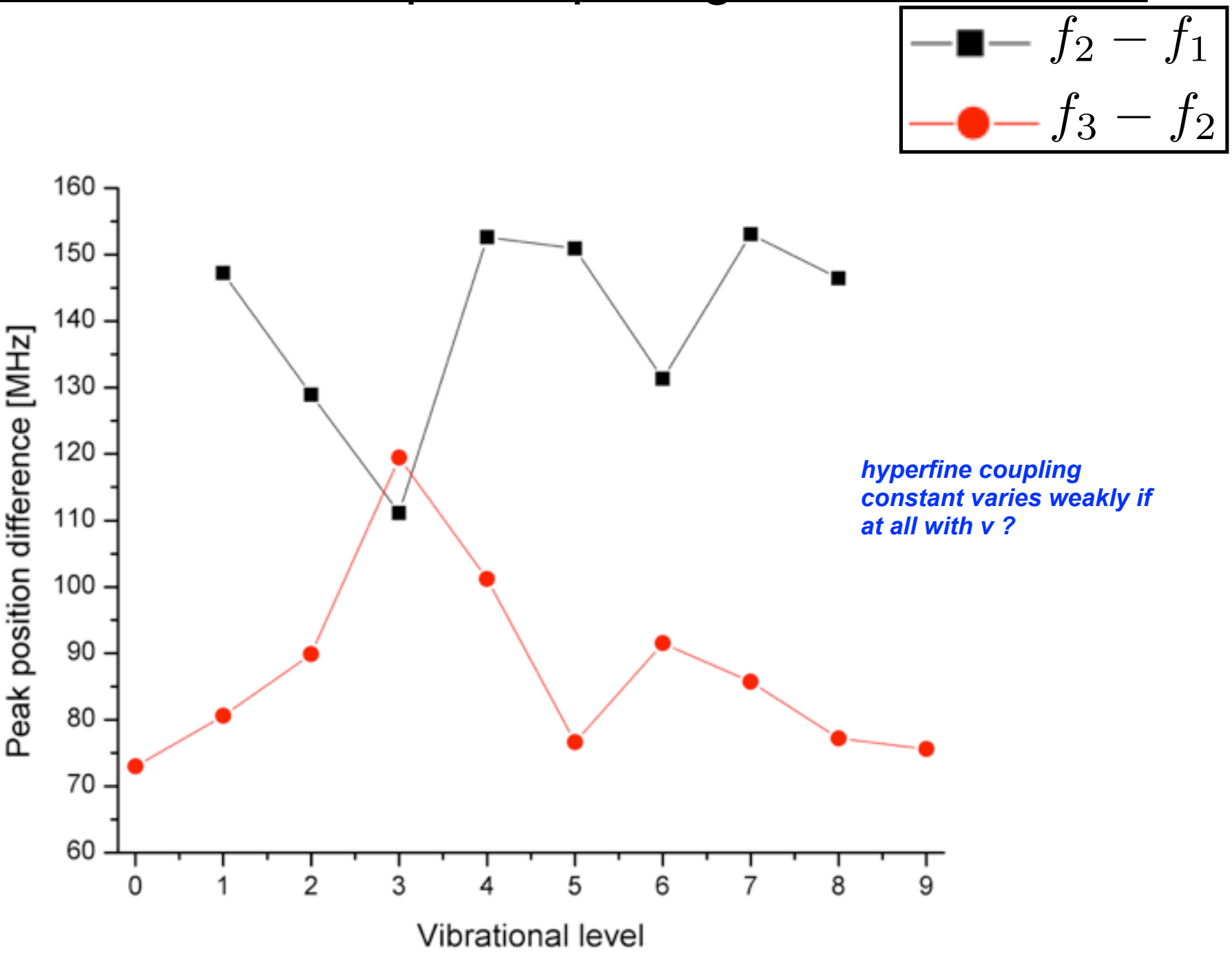


N=0 peak splittings

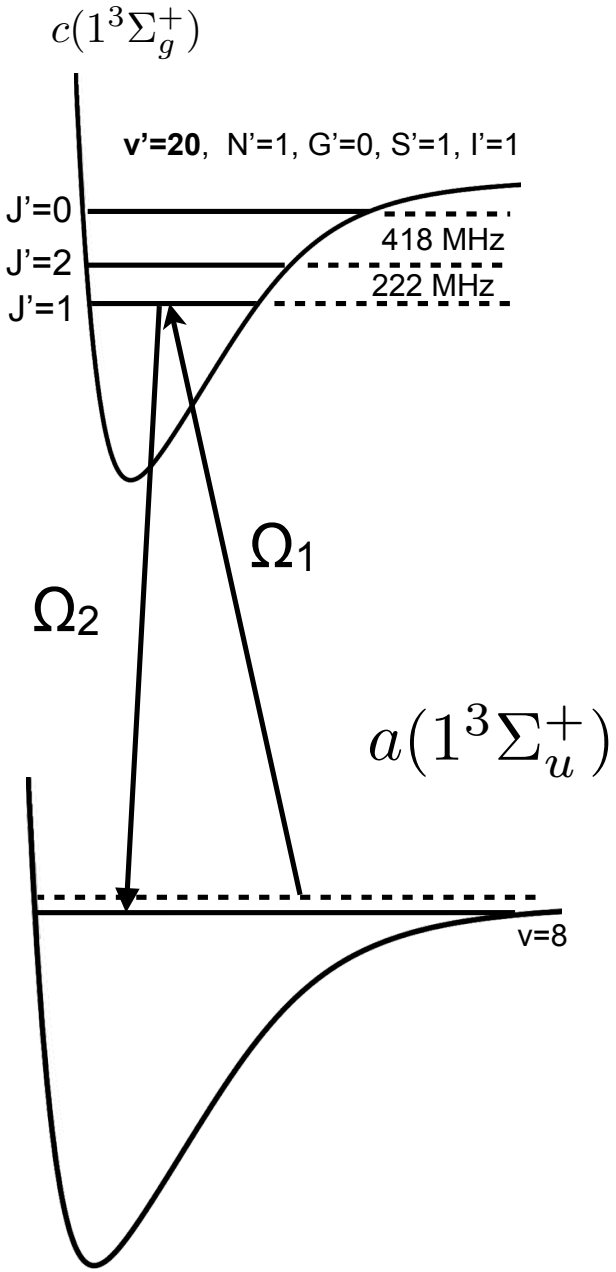


N=0 peak splittings

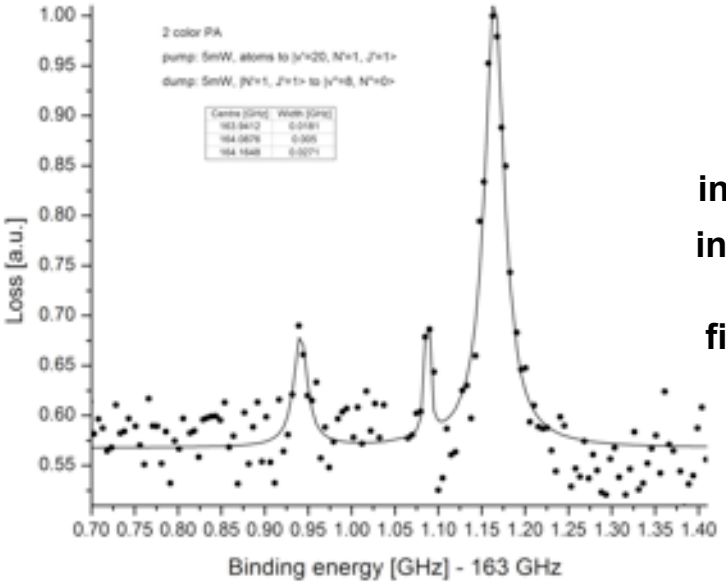


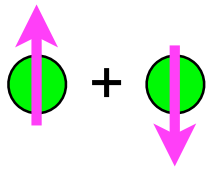
hyperfine coupling constant varies weakly if at all with v ?

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned



v''=8, N''=0





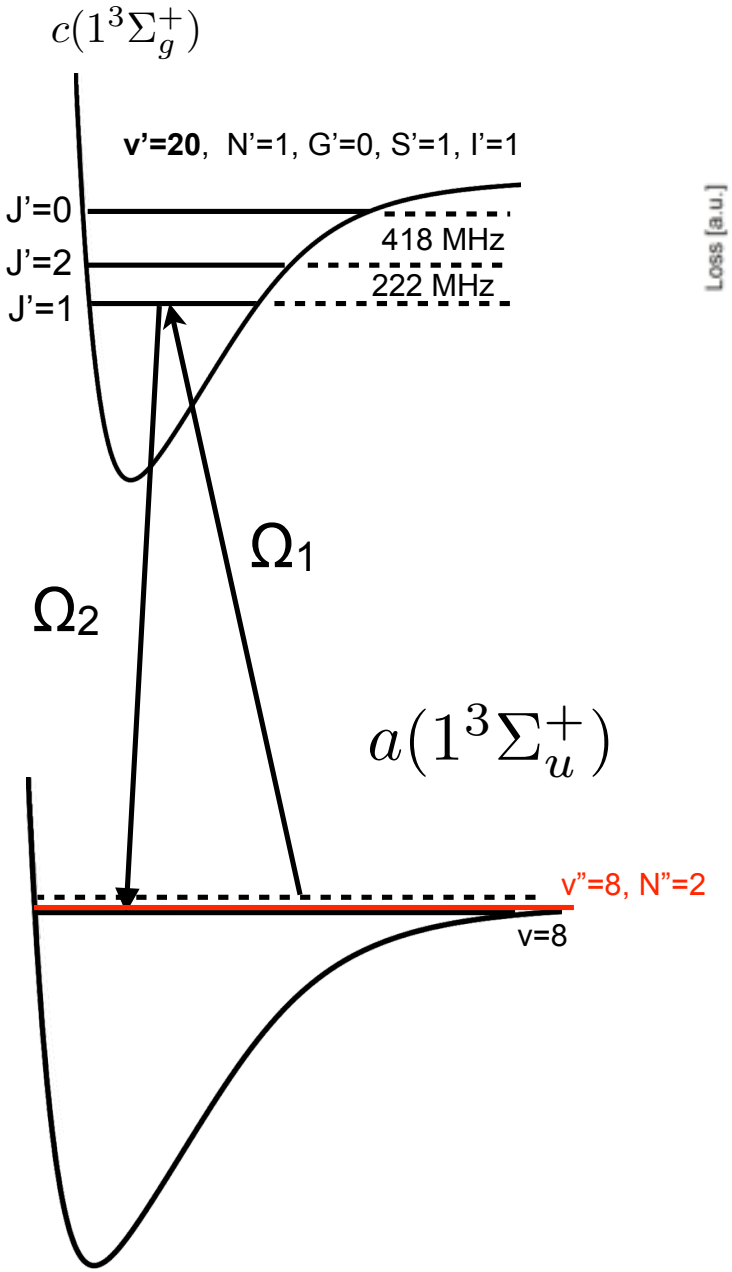
initial state |S=1,N=0,J=1,I=1>

intermediate: |S=1,N=1,J=1,I=1>

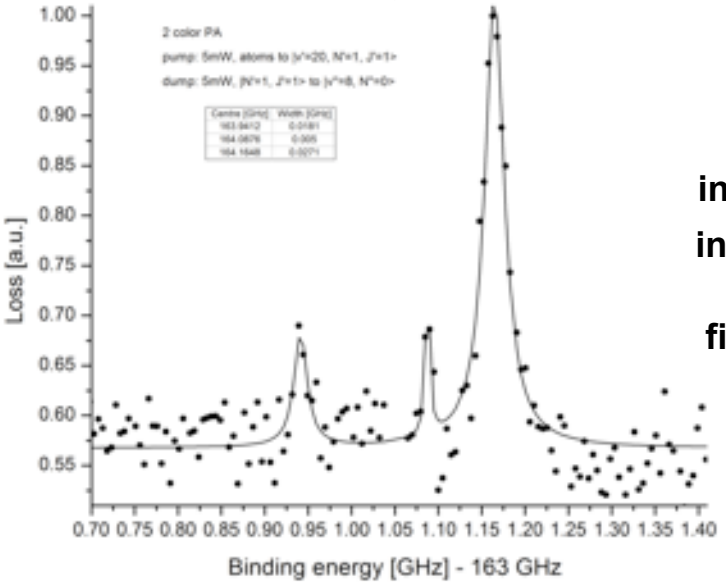
final: |S=1,N=0,J=1,I=1>

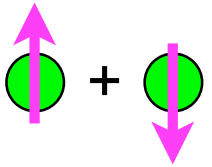
three features

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned



v"=8, N"=0





initial state |S=1,N=0,J=1,I=1>

intermediate: |S=1,N=1,J=1,I=1>

final: |S=1,N=0,J=1,I=1>

three features

v"=8, N"=2

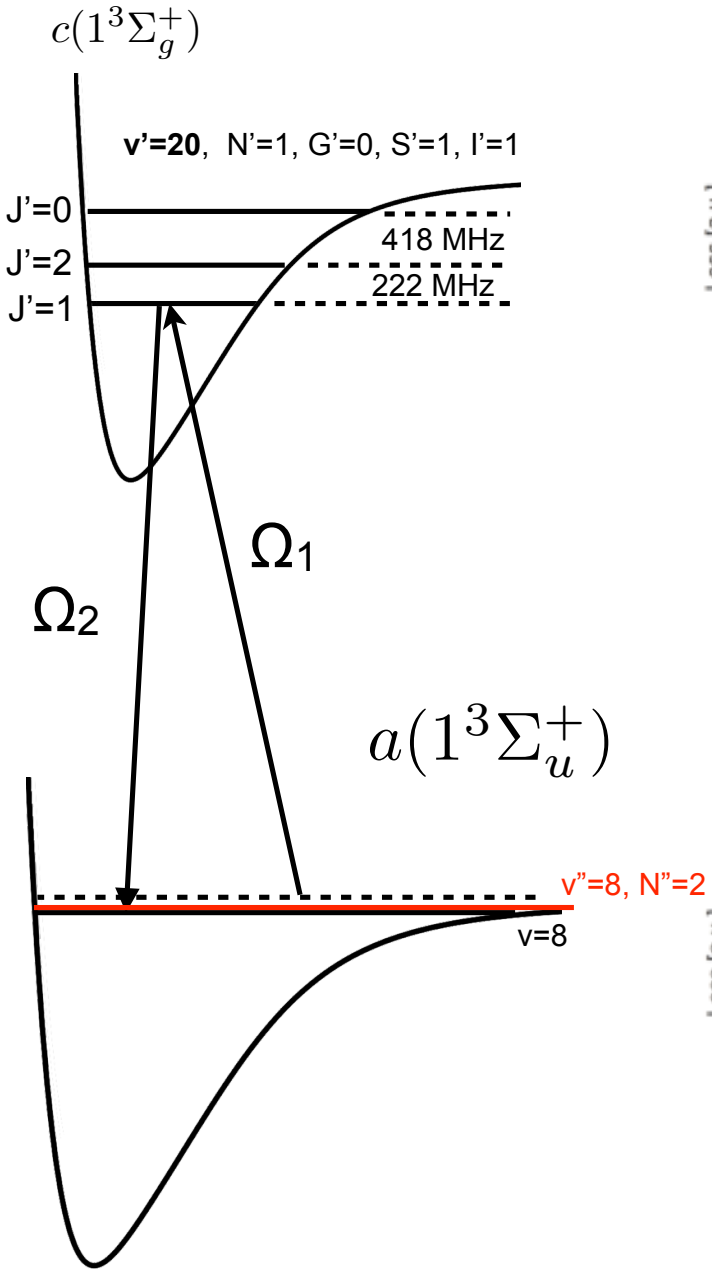
final state:
|S=1,N=2,J=1, I=1>

OR

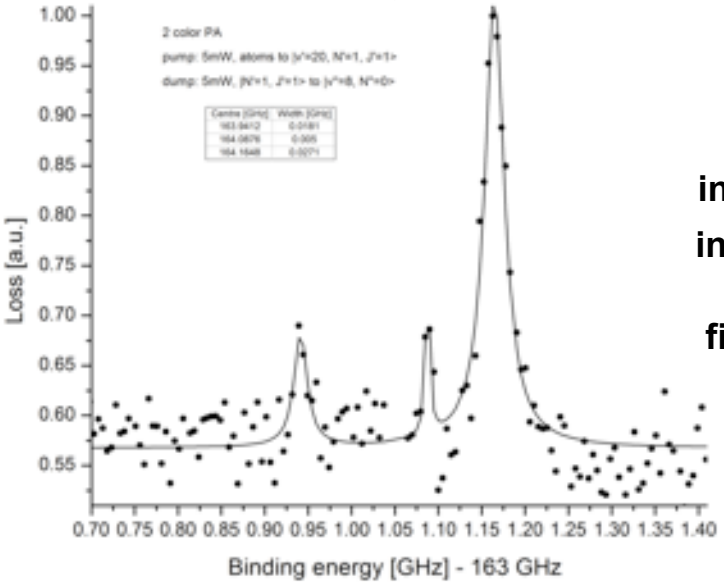
|S=1,N=2,J=2, I=1>

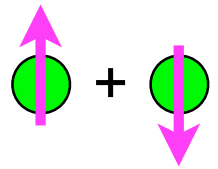
six features ?

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned



v"=8, N"=0





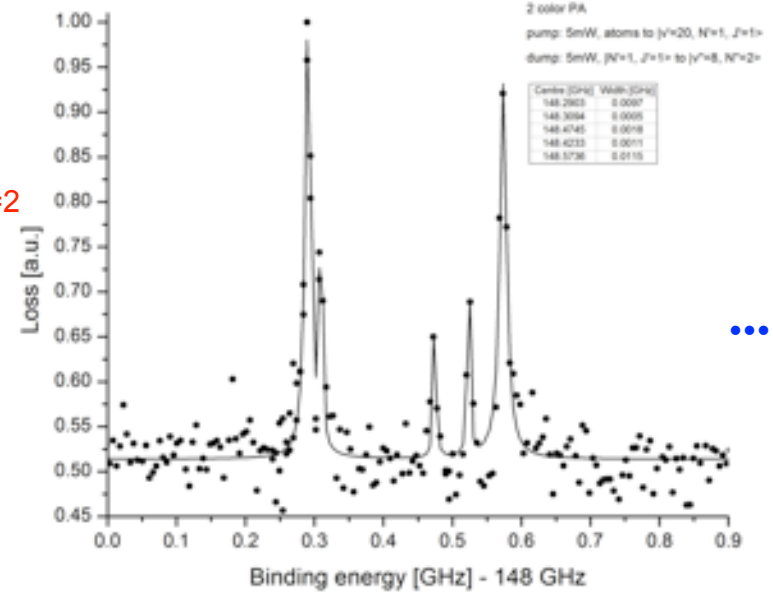
initial state |S=1,N=0,J=1,I=1>

intermediate: |S=1,N=1,J=1,I=1>

final: |S=1,N=0,J=1,I=1>

three features

v"=8, N"=2



final state:
|S=1,N=2,J=1, I=1>

OR

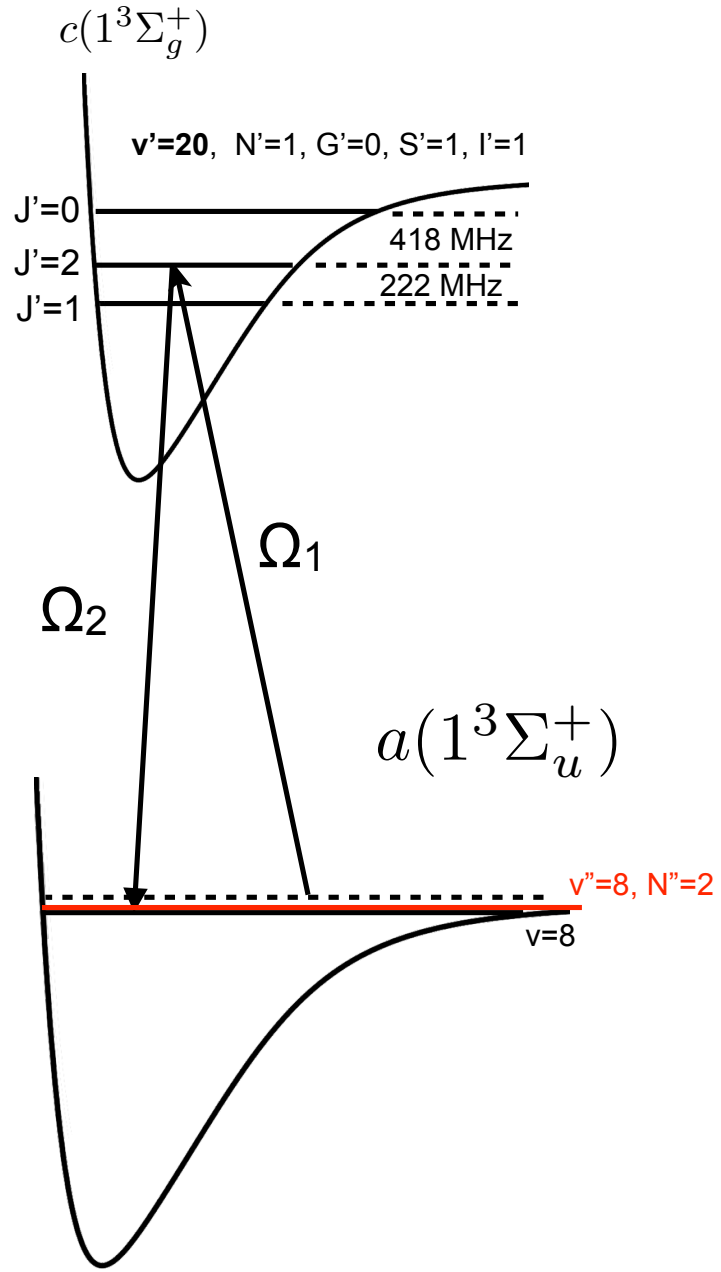
|S=1,N=2,J=2, I=1>

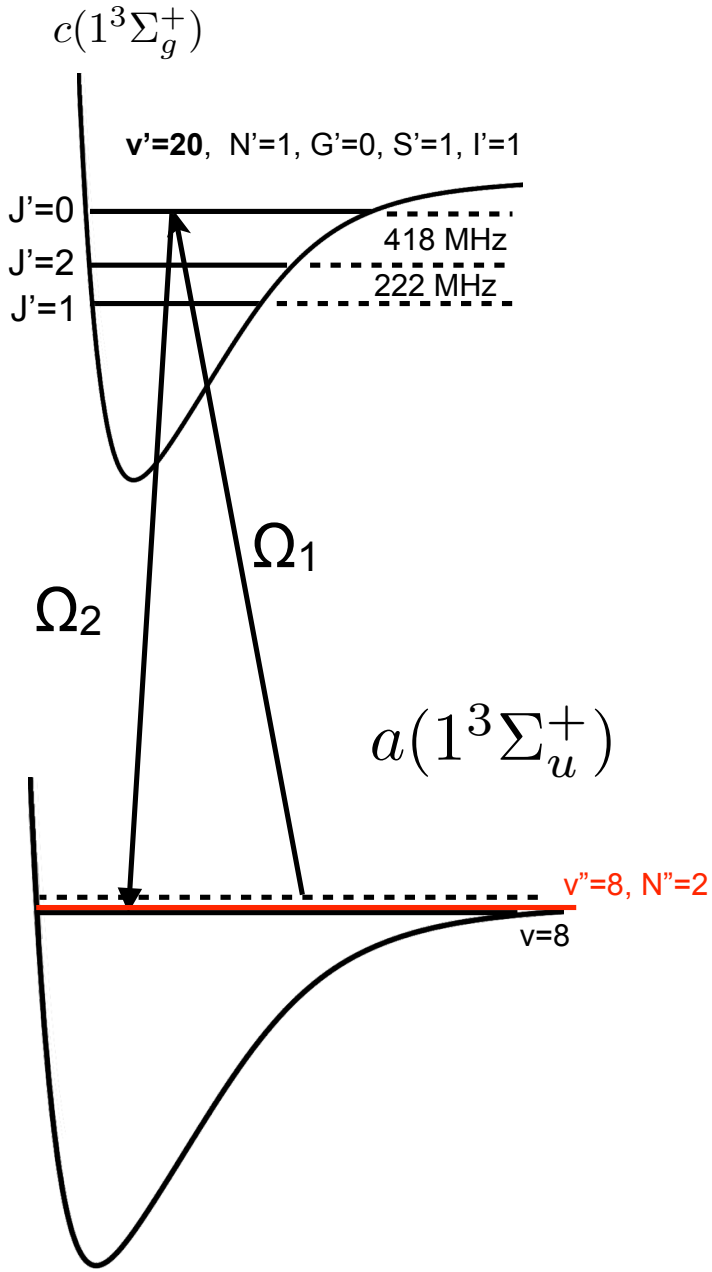
six features ?

... we see five features

2-color PA spectroscopy results : here Ω_1 is fixed and Ω_2 is scanned

we have 2 other choices of intermediate state





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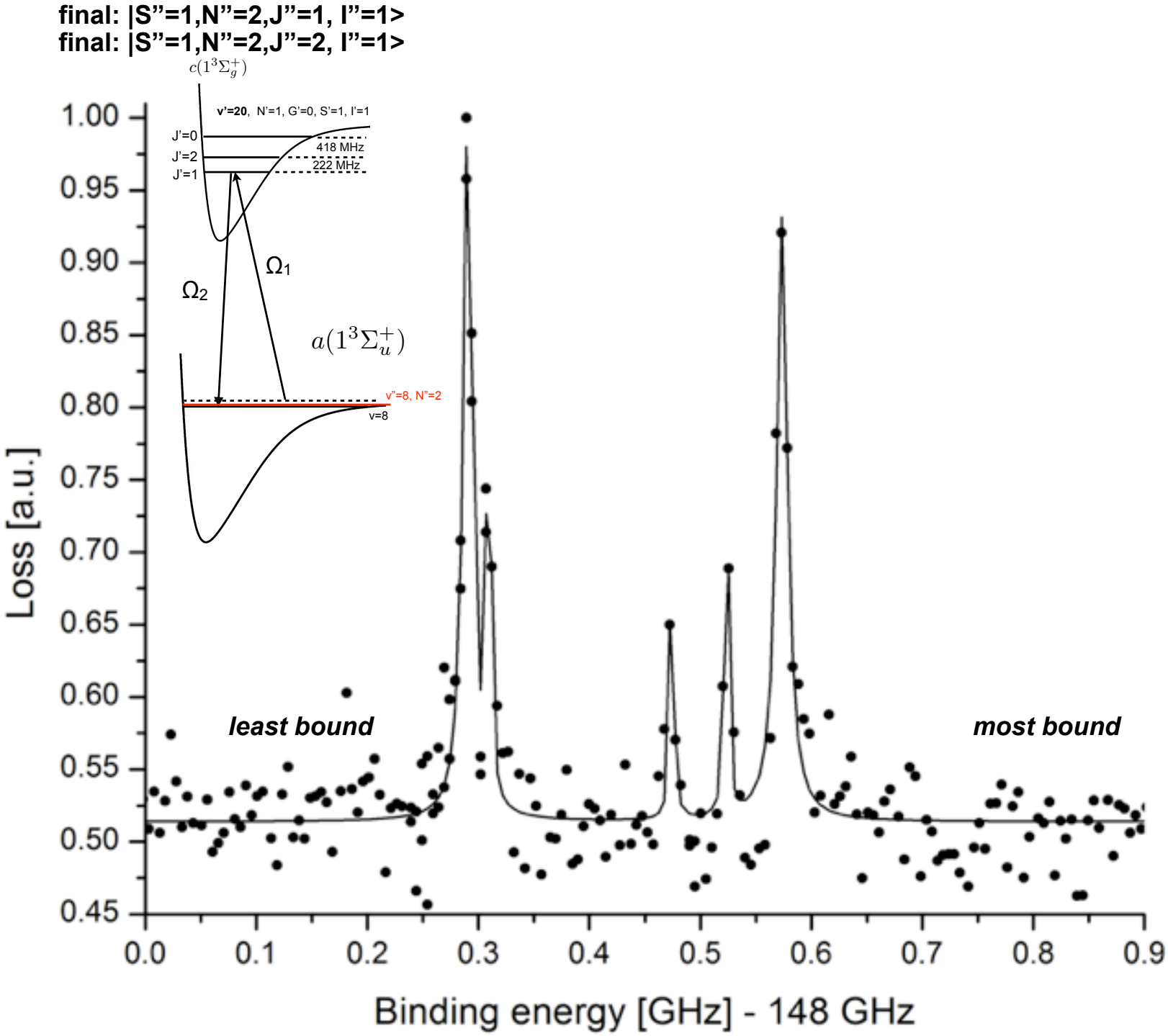

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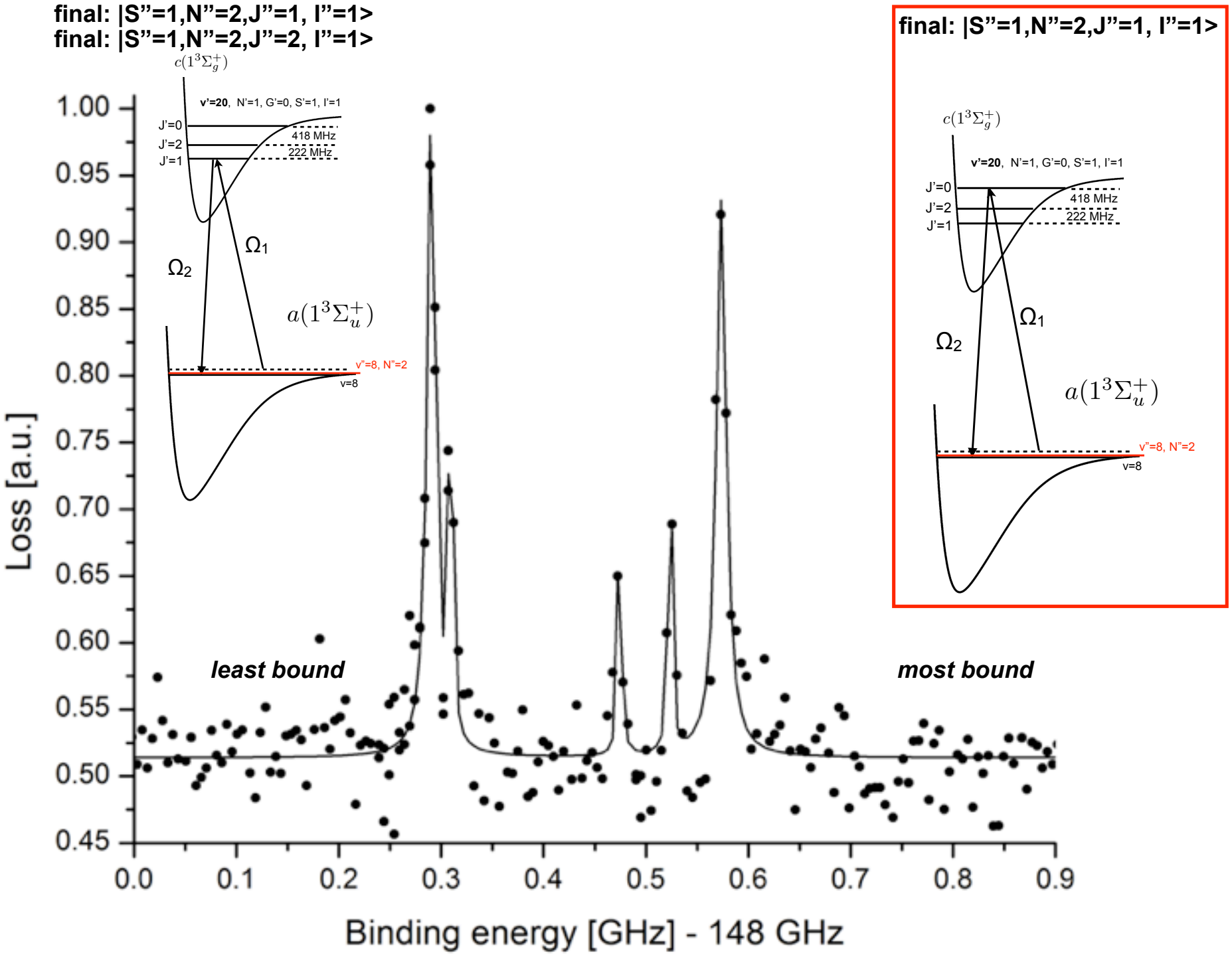

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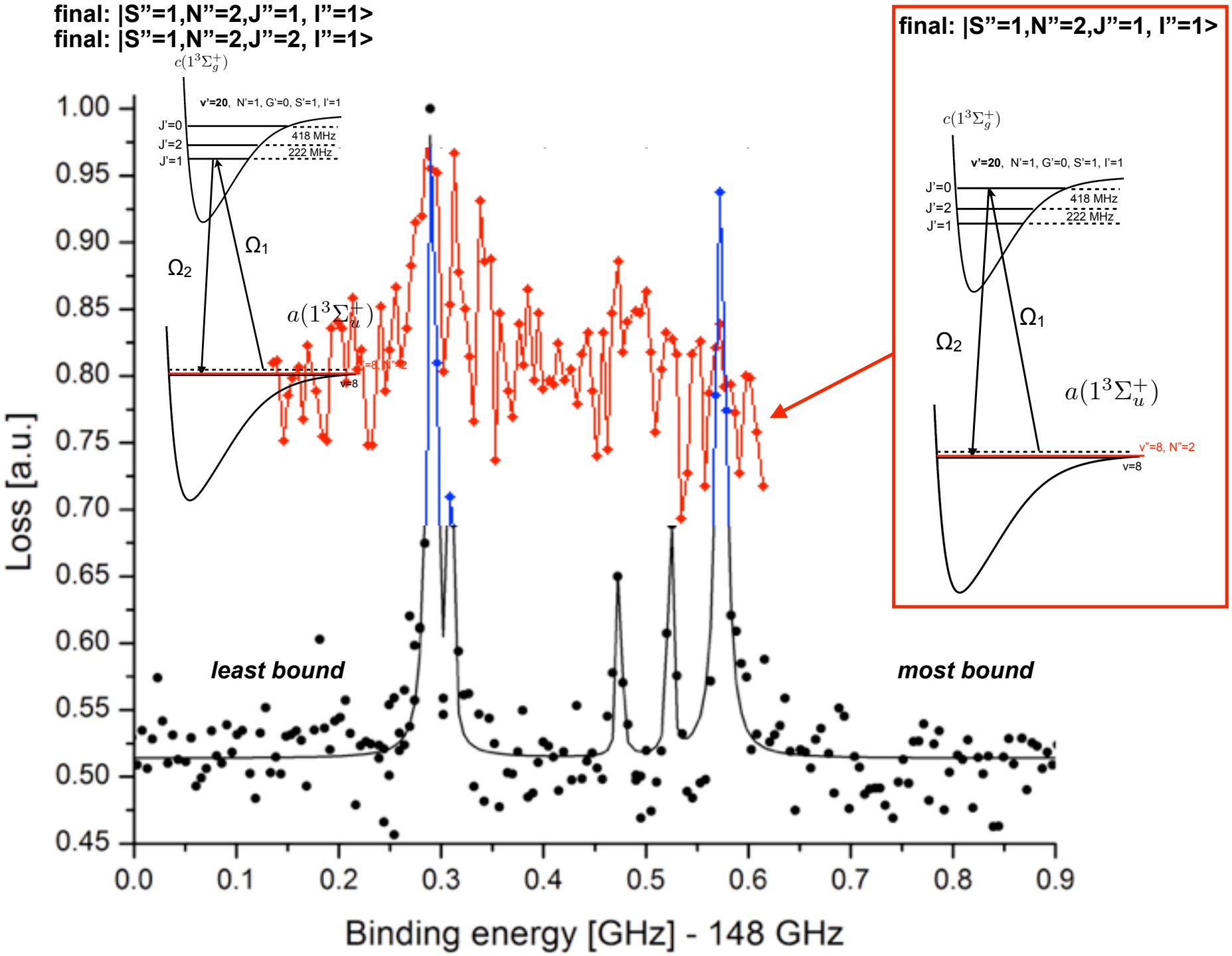

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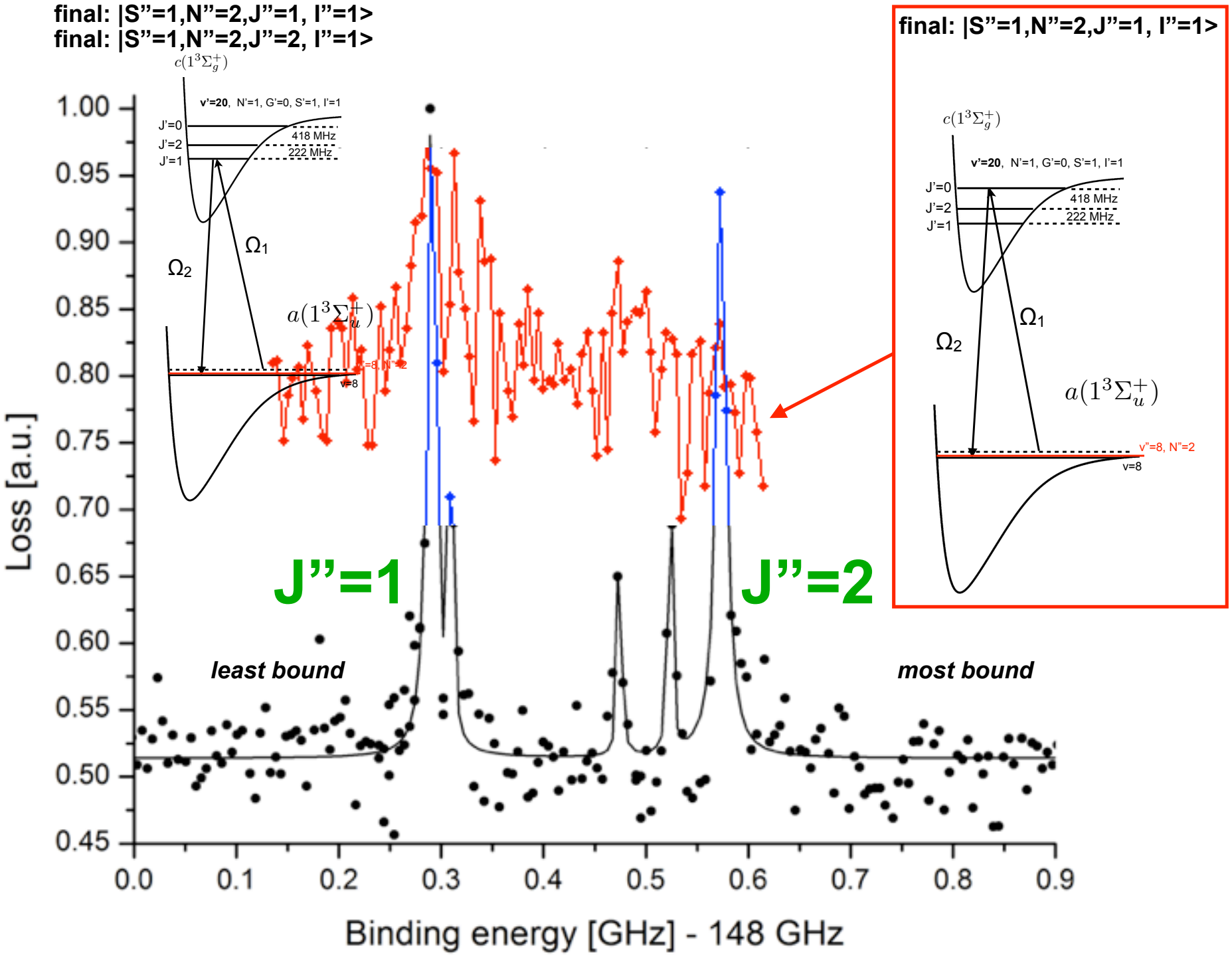

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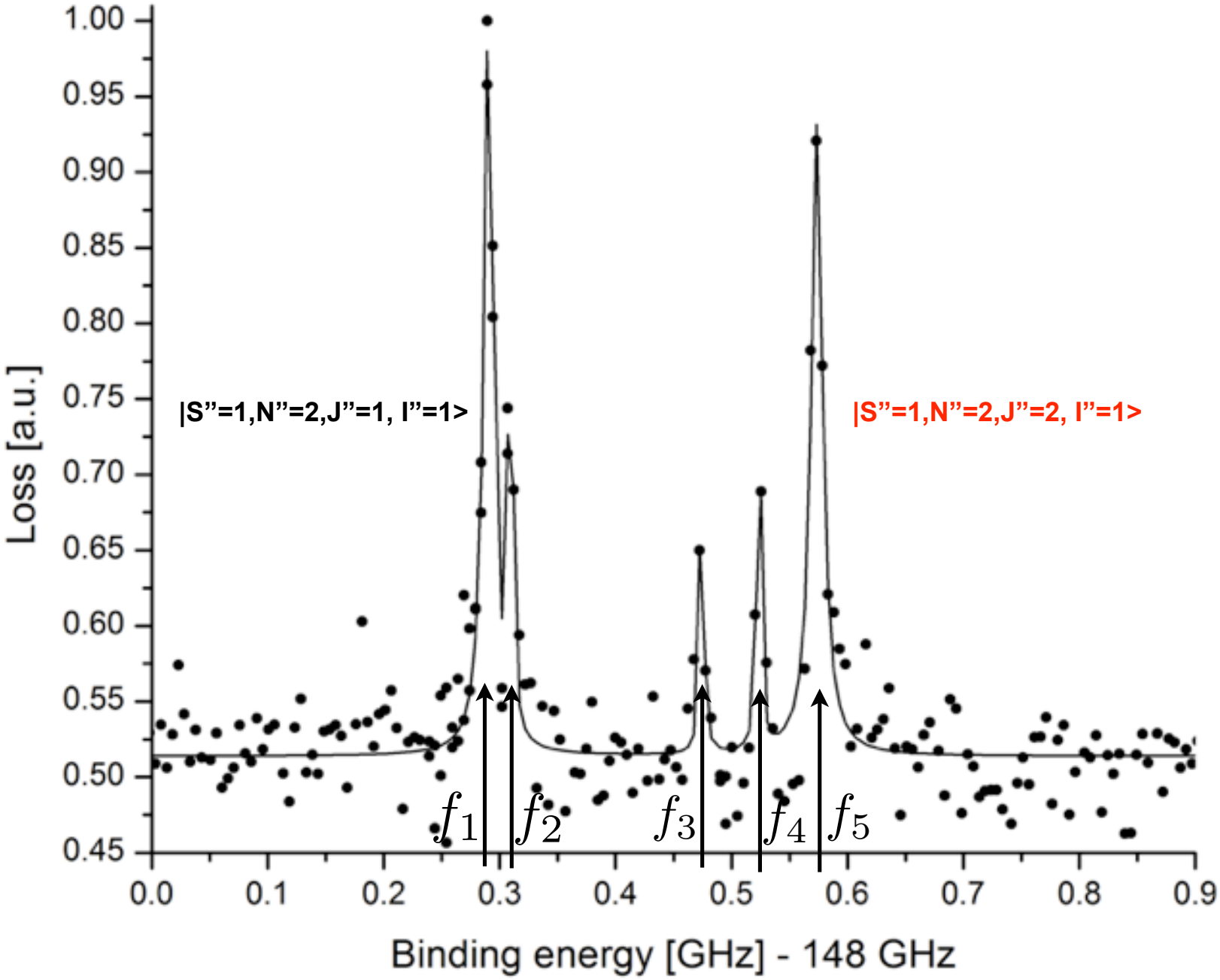
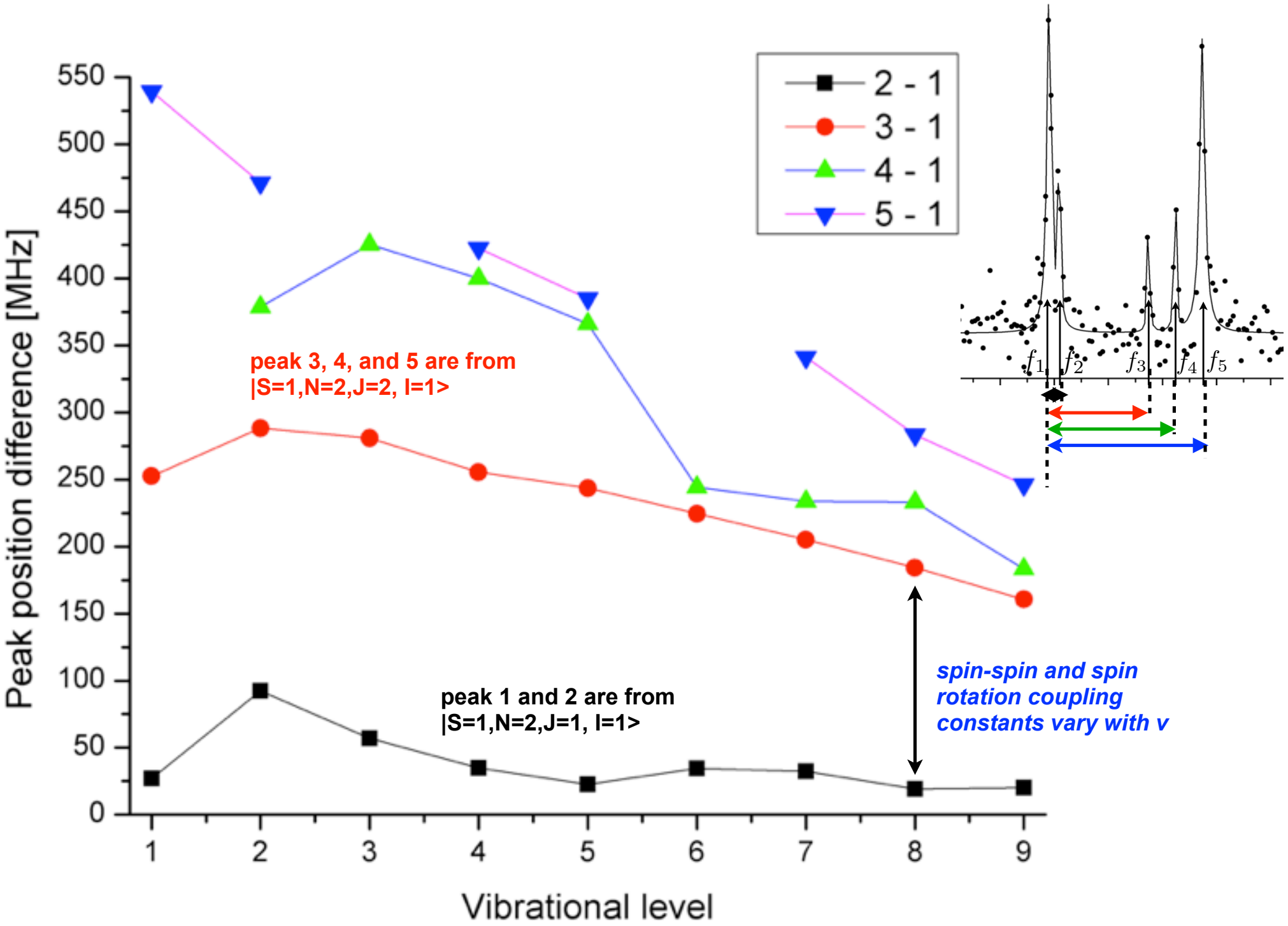
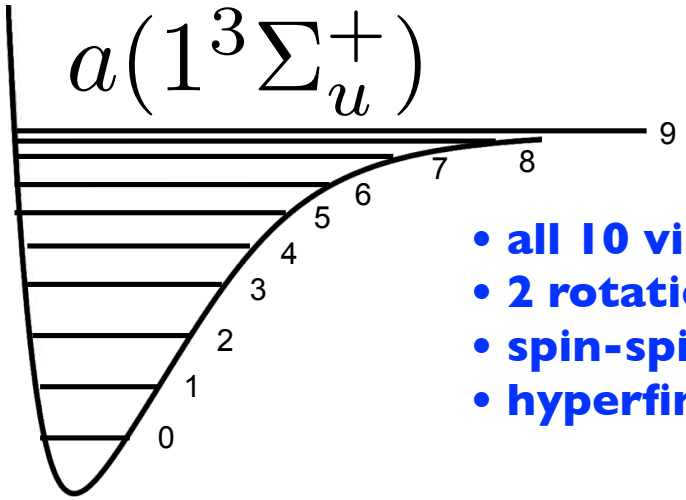



2 - 1
3 - 1
4 - 1
5 - 1
peak 3, 4, and 5 are from
|S=1,N=2,J=2, I=1>
peak 1 and 2 are from
|S=1,N=2,J=1, I=1>
spin-spin and spin
rotation coupling
constants vary with v
f_1 f_2 f_3 f_4 f_5
Peak position difference [MHz]
Vibrational level
0
50
100
150
200
250
300
350
400
450
500
550
1
2
3
4
5
6
7
8
9

Triplet ground state Li_2

Full characterization of the bound levels of the ground triplet state of $^6Li_2$



- **all 10 vibrational levels found**
- **2 rotational levels for each vibrational level (N"=0, N"=2)**
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- **hyperfine structure resolved**

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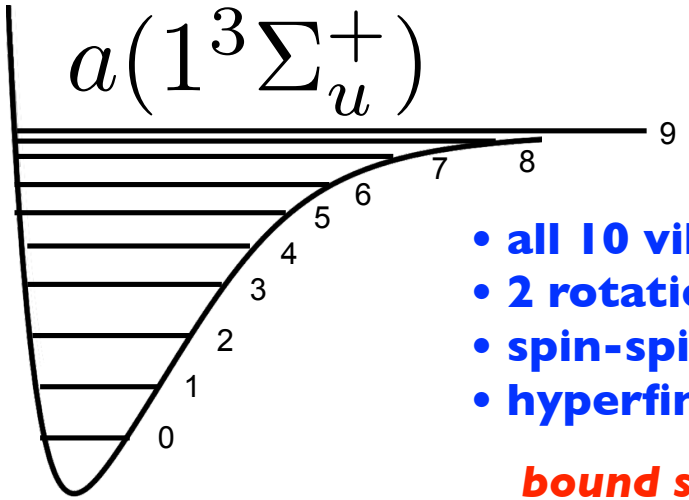

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bound state energy resolution < 500 kHz (< 100 kHz)

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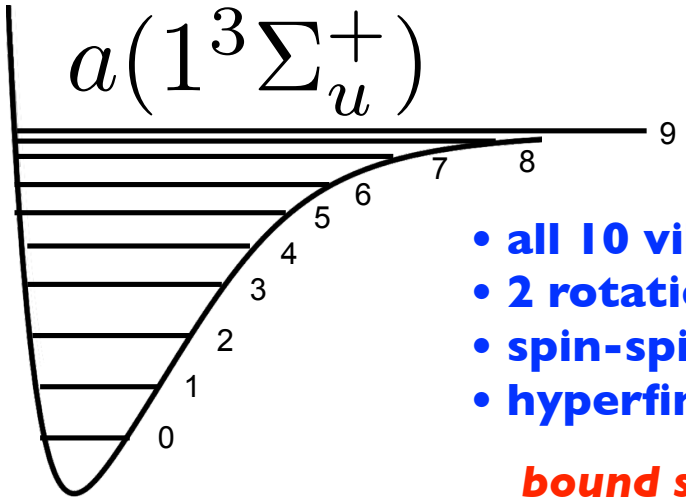

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the ground triplet state of Li is now the most accurately known molecular potential.

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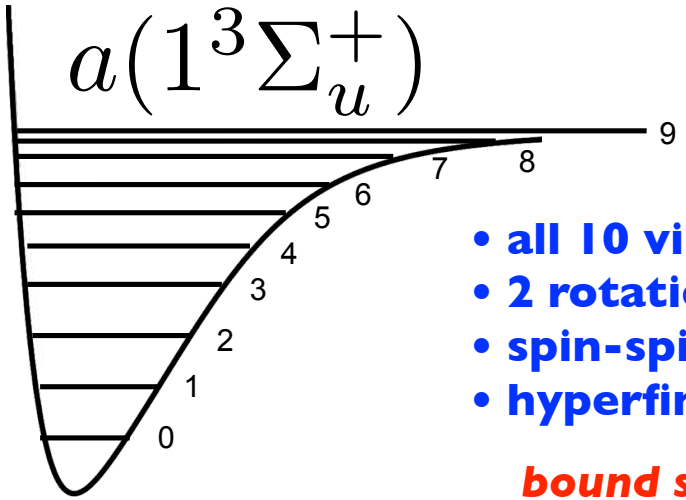

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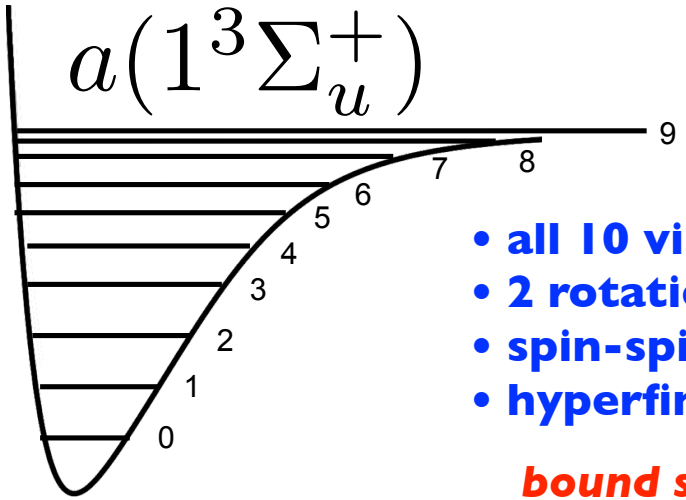
the ground triplet state of Li is now the most accurately known molecular potential.

Is this useful?
It's the simplest molecule after to H_2

Triplet ground state Li_2

Full characterization of the bound levels of the ground triplet state of $^6Li_2$



- **all 10 vibrational levels found**
- **2 rotational levels for each vibrational level (N"=0, N"=2)**
- **spin-spin and spin-rotation splitting of J levels observed**
- **hyperfine structure resolved**

bound state energy resolution < 500 kHz (< 100 kHz)

the ground triplet state of Li is now the most accurately known molecular potential.

Is this useful?
It's the simplest molecule after to H_2

Next step: STIRAP transfer FR molecules into these states and measure their lifetime

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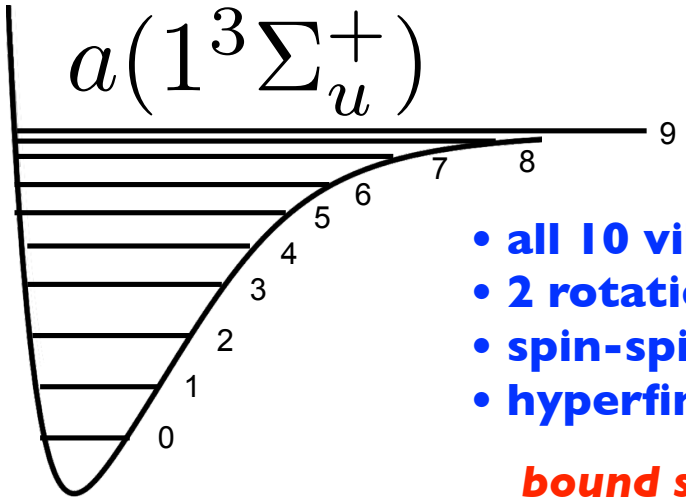

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