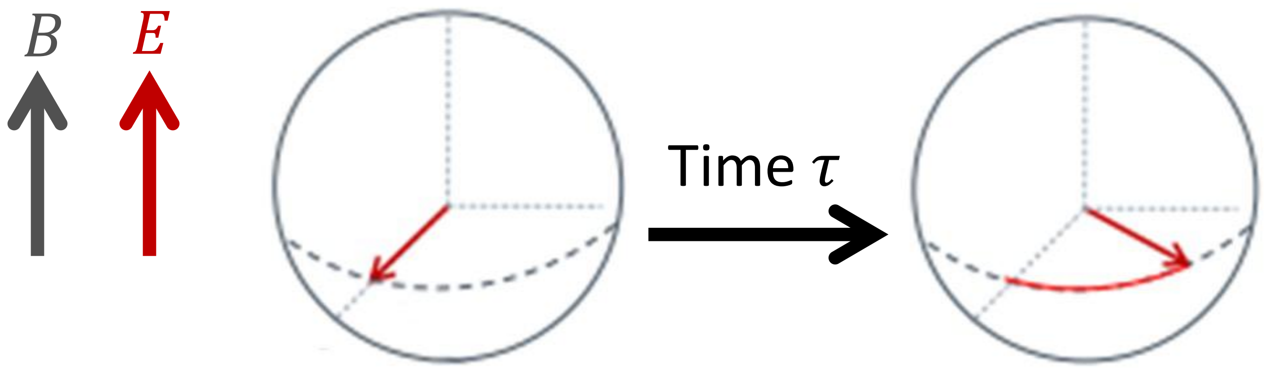
Laser-cooled polyatomic molecules for CP-violation searches

Zack Lasner
Doyle group, Harvard University

Kobayashi-Maskawa Institute, 03/03/2023

EDM measurement scheme

$$H = -\mu \cdot B - d \cdot E$$



$$\phi_+ = \mu B\tau + dE\tau$$

EDM measurement scheme

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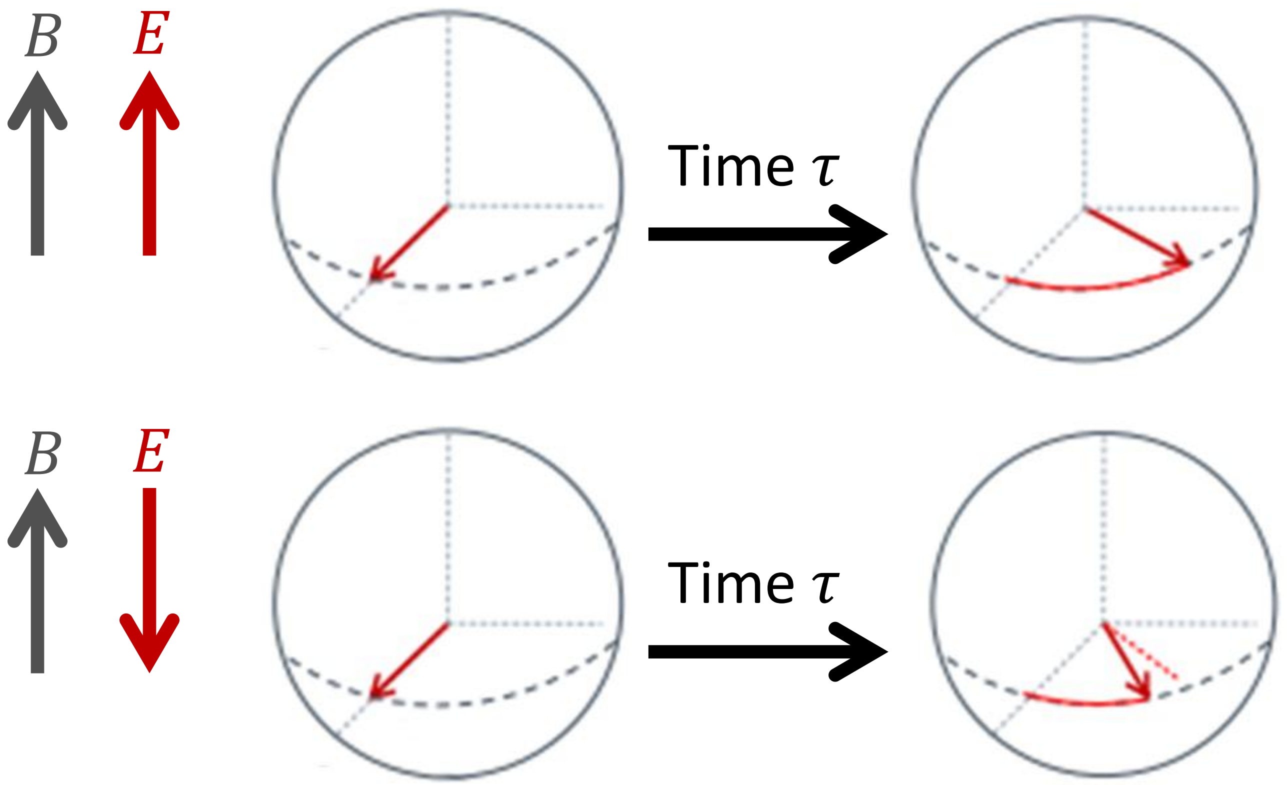

$$\phi_+ = \mu B\tau + dE\tau$$

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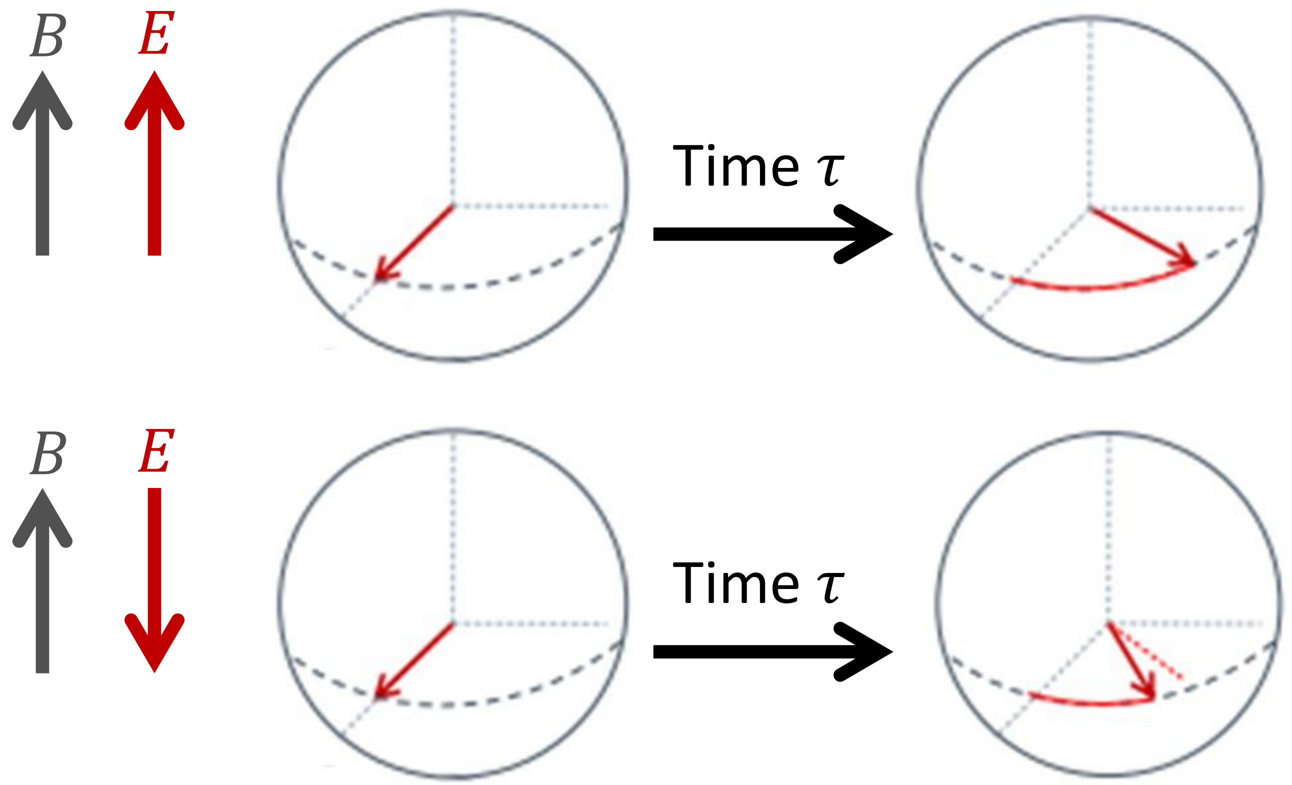

$$\phi_{+} = \mu B\tau + dE\tau$$

$$\phi_{-} = \mu B\tau - dE\tau$$

$$d \propto \phi_{+} - \phi_{-}$$

EDM measurement scheme

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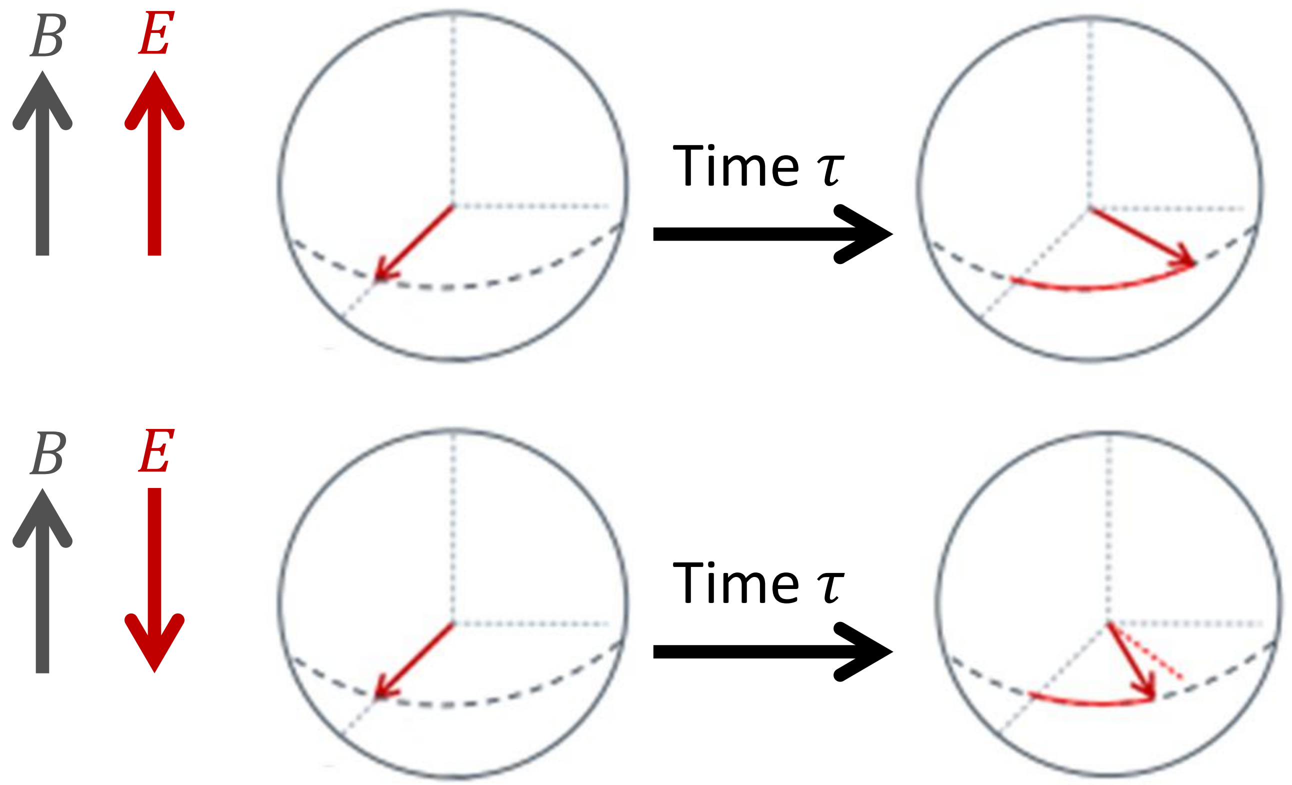


$$\phi_+ = \mu B\tau + dE\tau$$

$$\phi_- = \mu B\tau - dE\tau$$

Figure of merit:
$\frac{1}{\Delta d} \propto E\tau\sqrt{N}$

E = electric field
τ = precession time
N = number of repetitions

$$d \propto \phi_+ - \phi_-$$

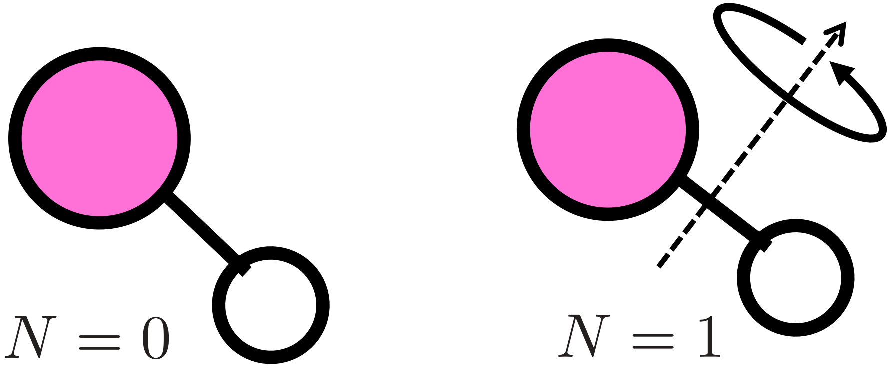
The polyatomic advantage:

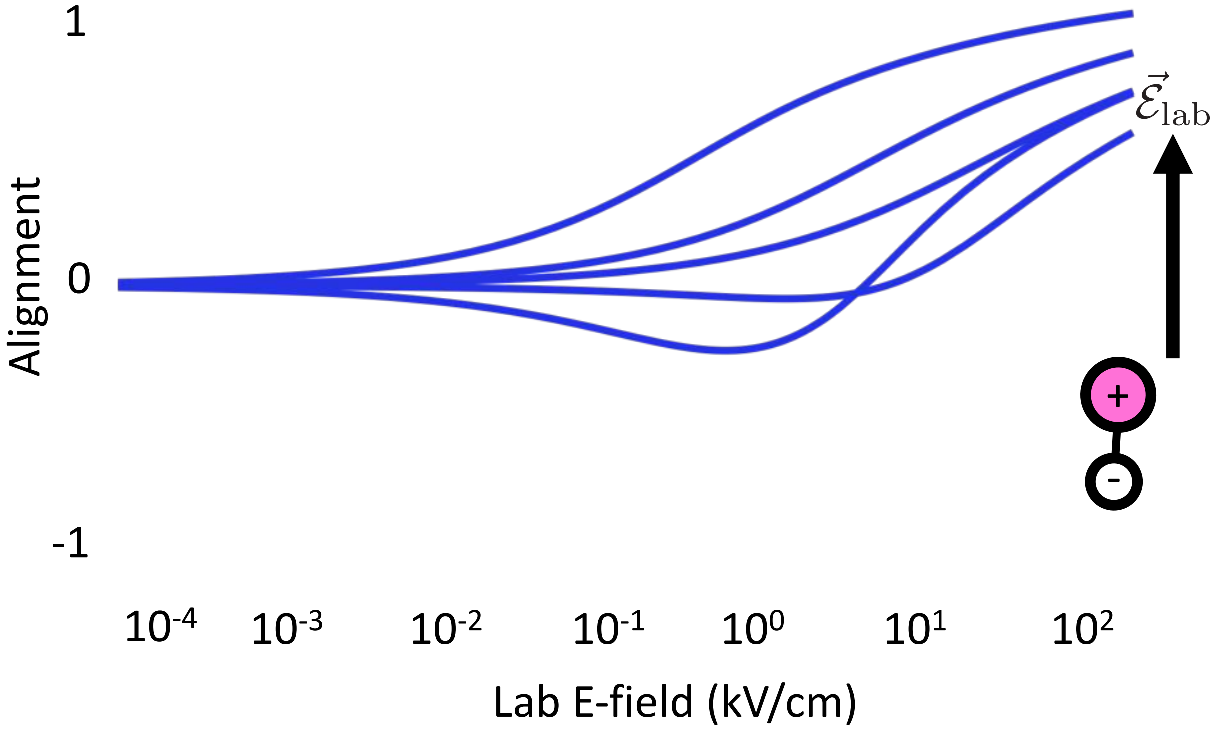
Figure of merit:
$$\frac{1}{\Delta d} \propto E\tau\sqrt{N}$$

Parity doubling + trapping + high density

- Large E → fully polarized → "small" splitting between $P = \pm 1$
- Large τ → trapped → ions or ultracold
- Large N → neutral

Diatomic Molecule – Rotation





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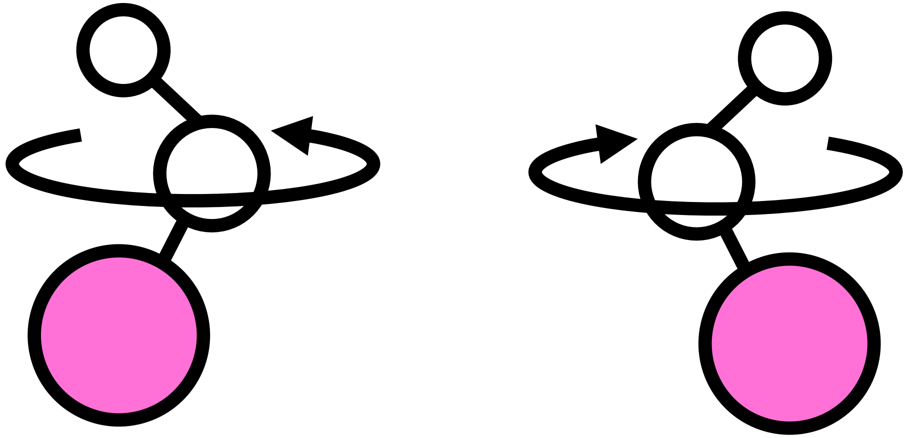
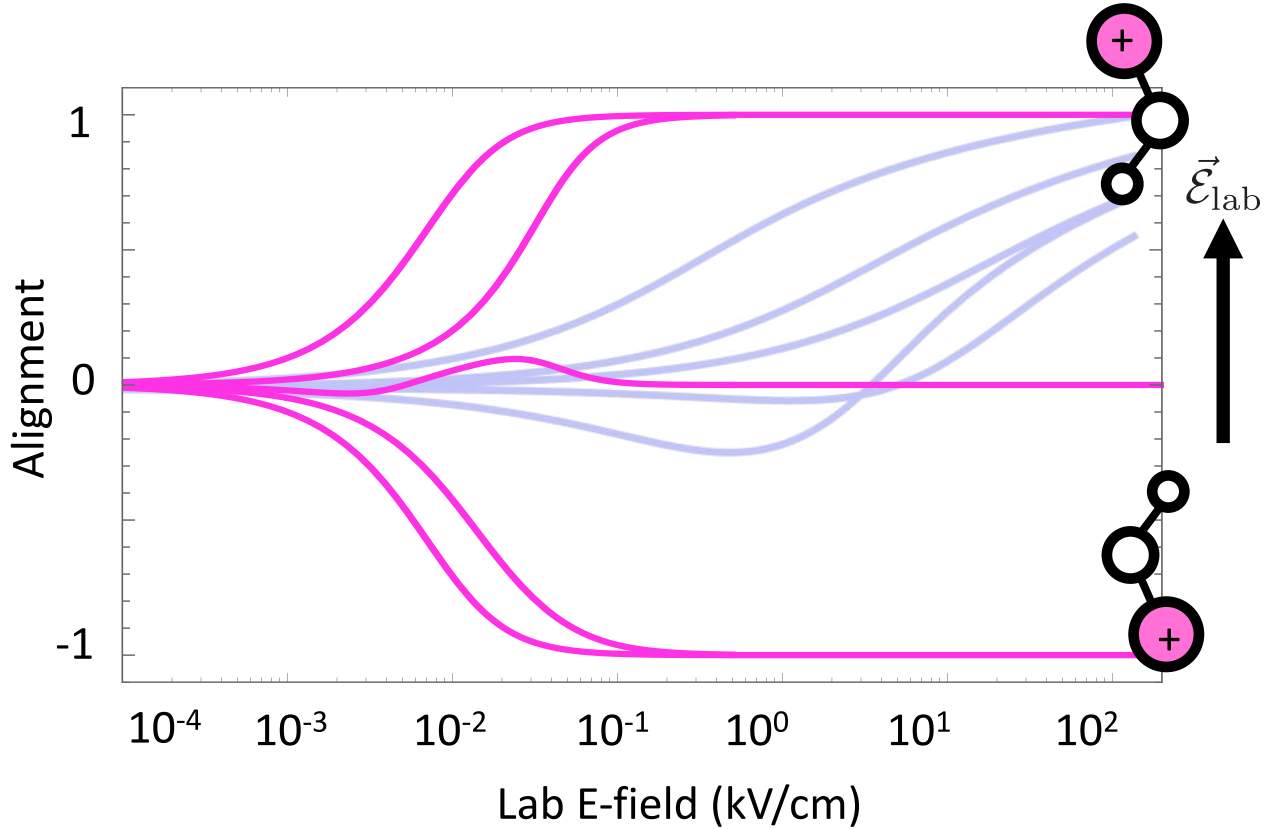
Polyatomic Molecule – Bending Mode



$\ell = +1$ $\ell = -1$



The polyatomic advantage:

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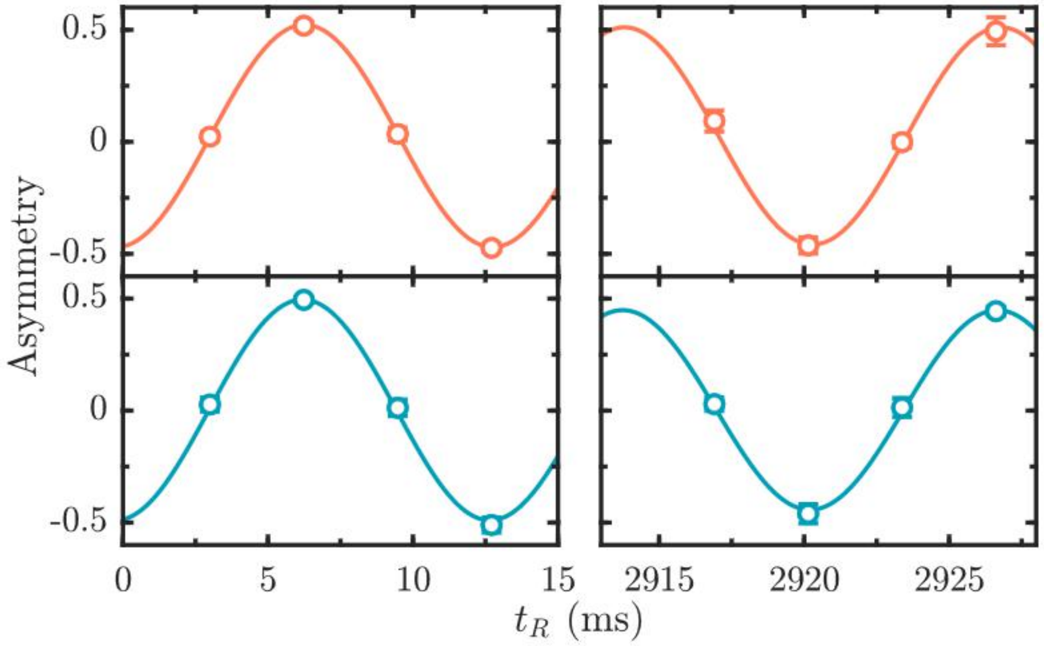
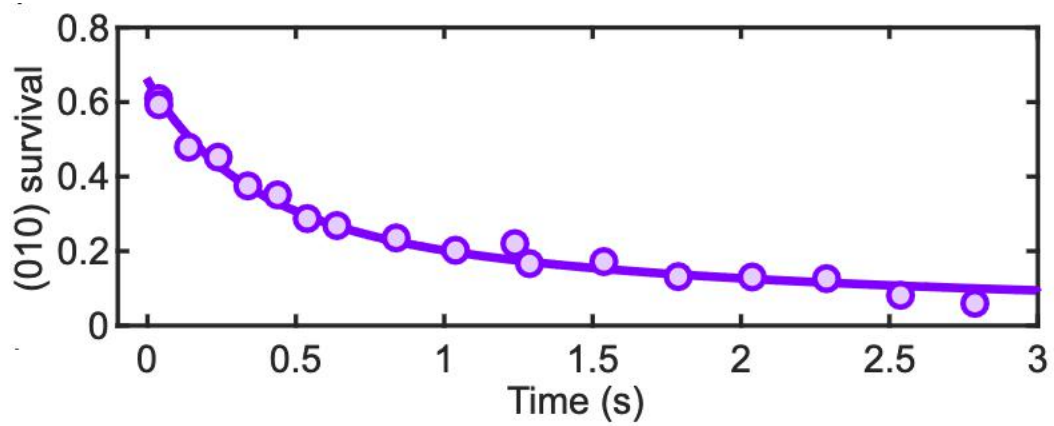


arxiv:2212.11837 [JILA]



arxiv:2208.13762 [CaOH, accepted PRL]

The polyatomic advantage:

Parity doubling + trapping + high density

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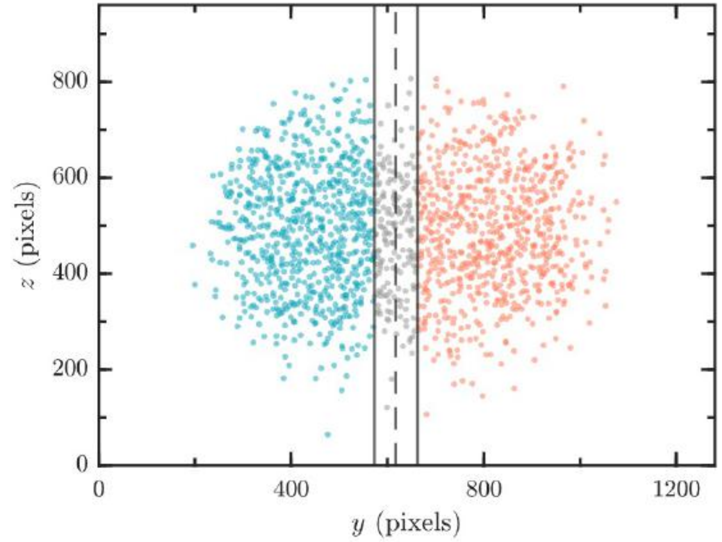
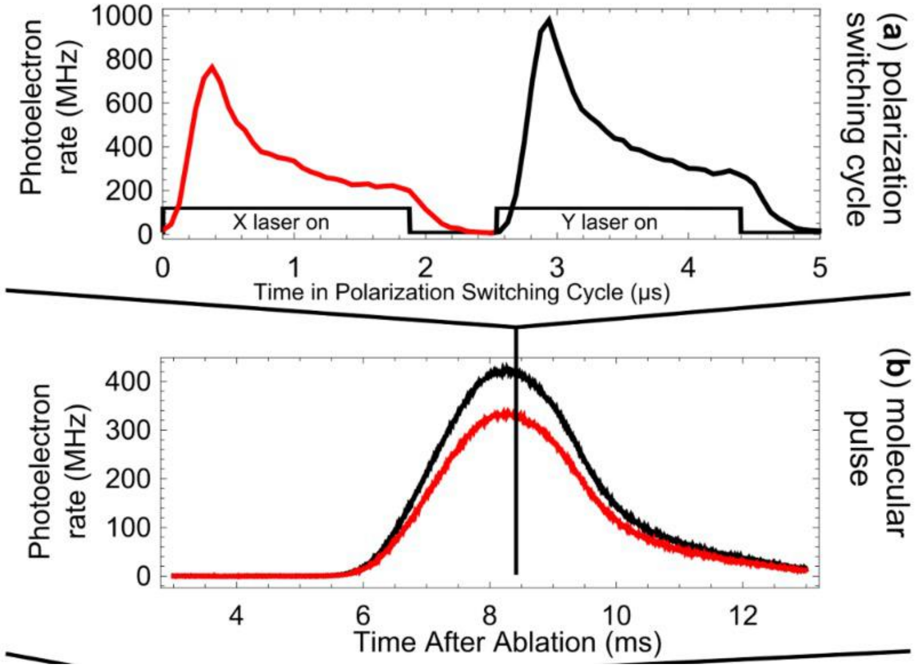


arxiv:2212.11837
JILA: hundreds per shot



https://www.nature.com/articles/s41586-018-0599-8
~300,000 per pulse [ACME III will be much higher]

The polyatomic advantage:

Figure of merit:
$$\frac{1}{\Delta d} \propto E\tau\sqrt{N}$$

Parity doubling + trapping + high density

- Large E → fully polarized → "small" splitting between $P = \pm 1$
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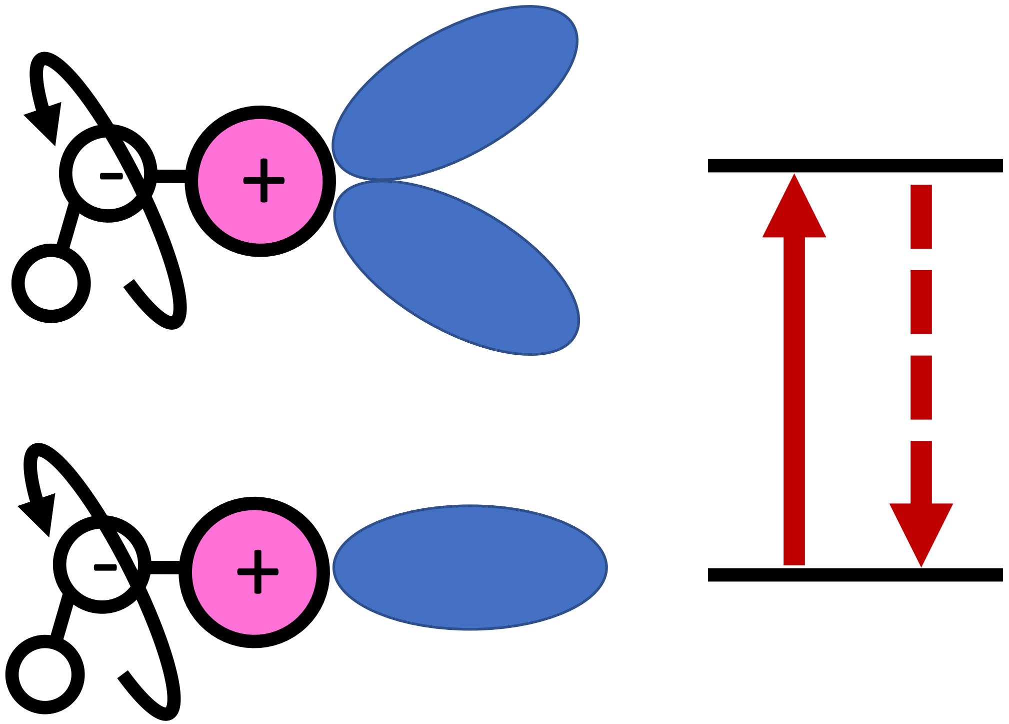
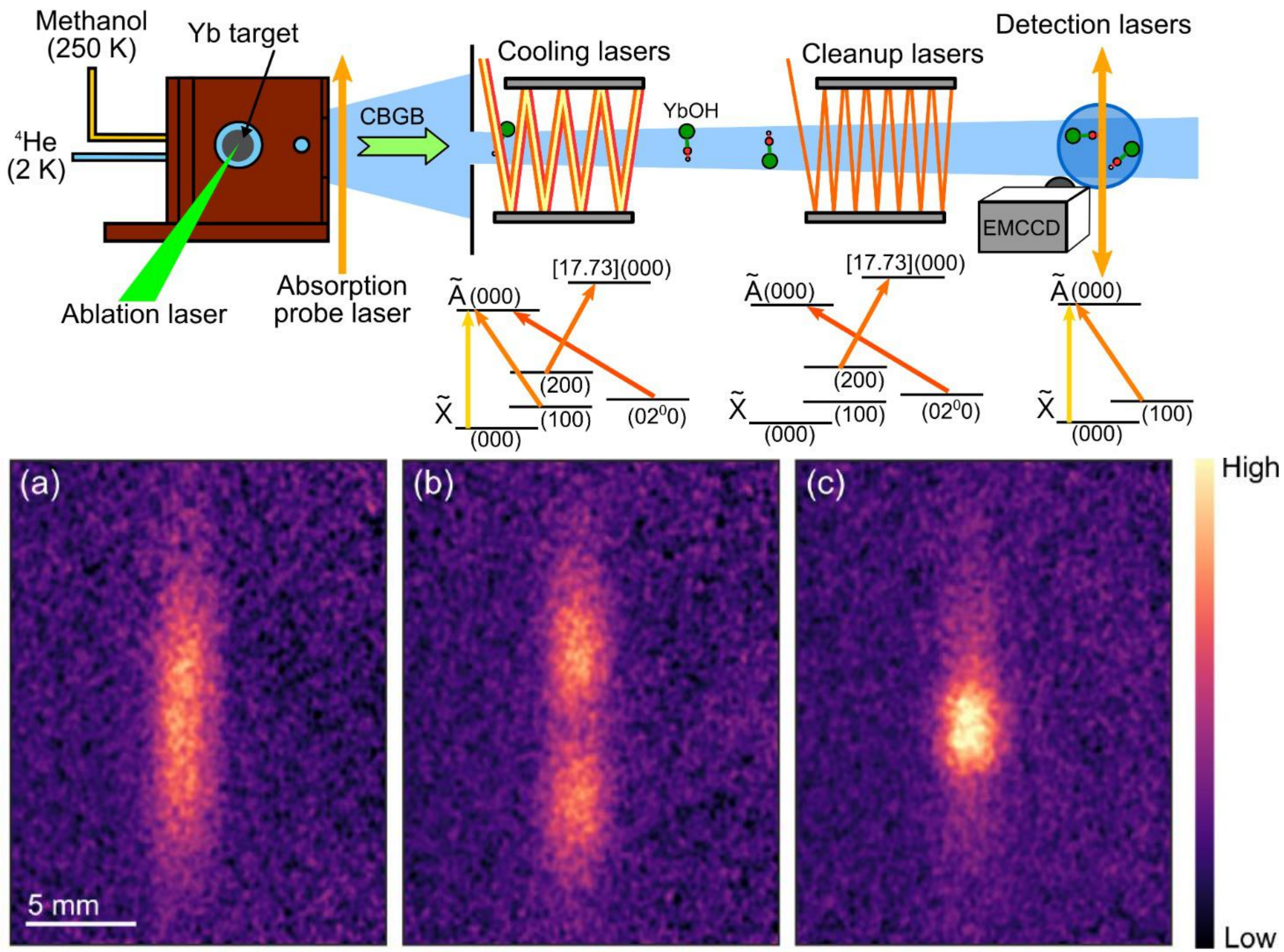


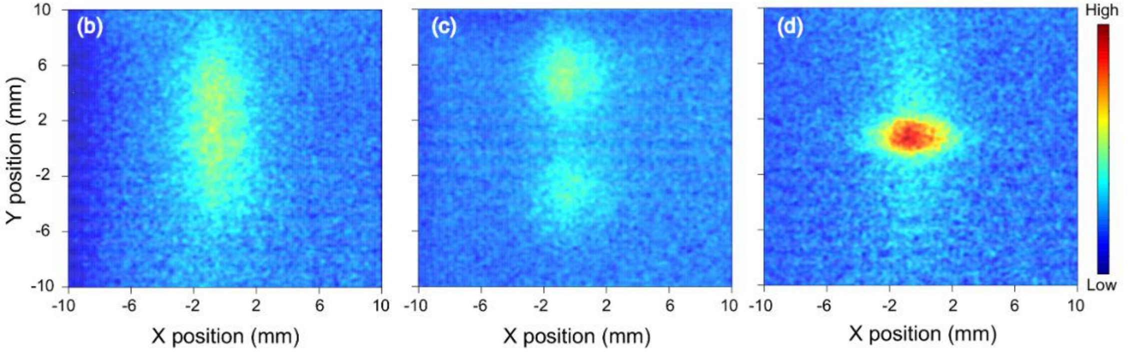
- Parity doublet achieved by nuclear motion, laser cooling achieved by isolated electronic excitation
- Unlike diatomics, the mechanism for the parity doublet is isolated from the laser cooling process

Previous work in polyatomic laser cooling

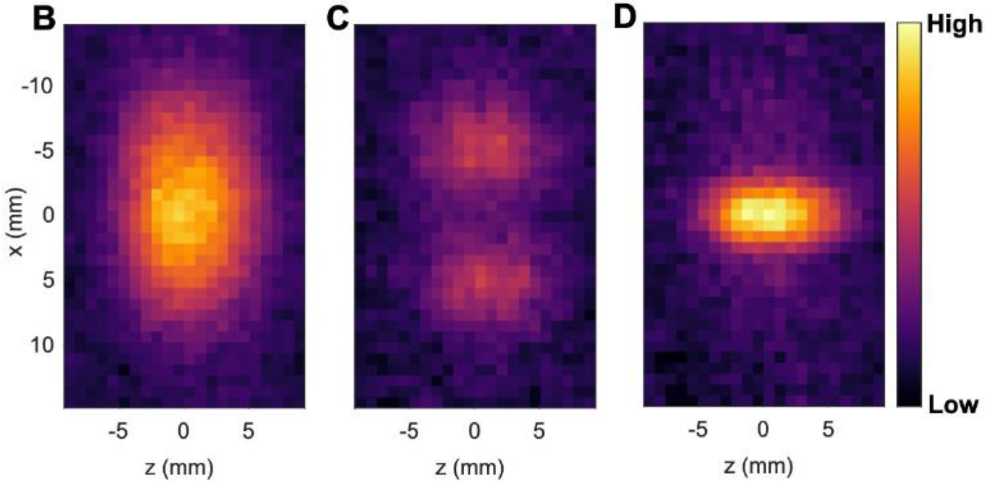
YbOH: NJP **22** 022003 (2020)



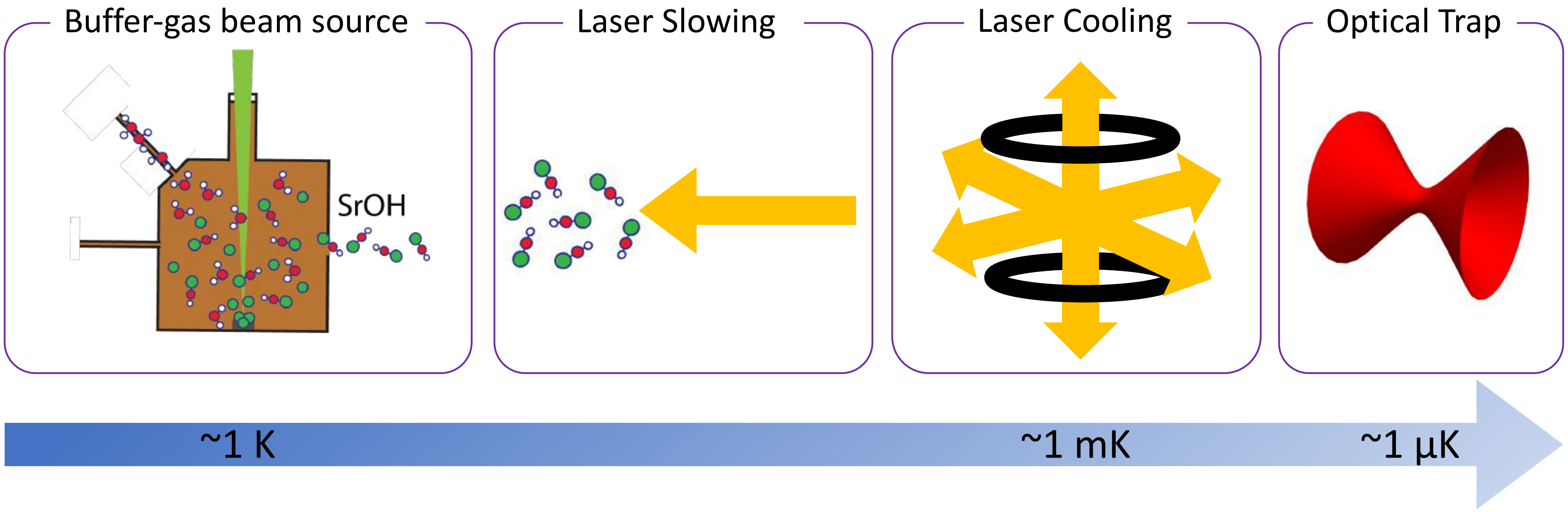
SrOH: PRL **118** 173201 (2018)



$CaOCH_3$: Science **369** 1366 (2020)

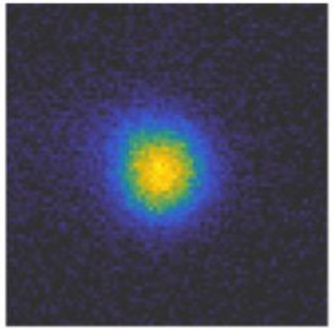


Pathway to trapped, ultracold molecules

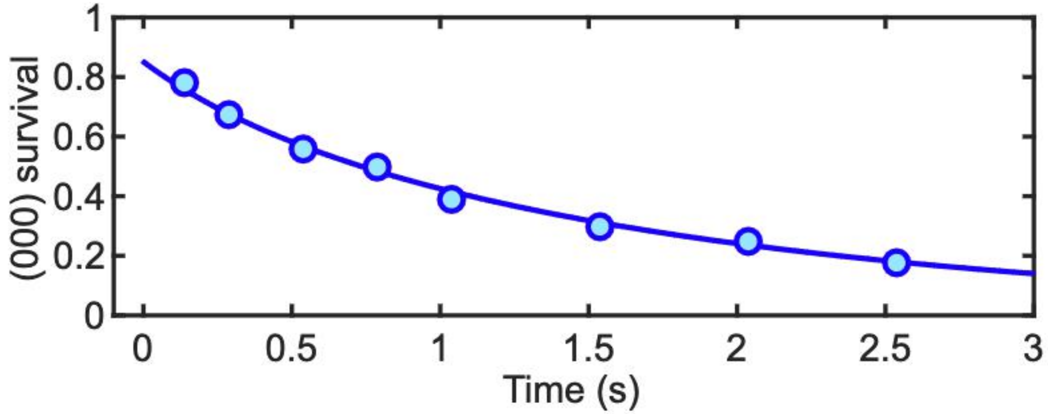


CaOH: State of the art in polyatomic laser cooling

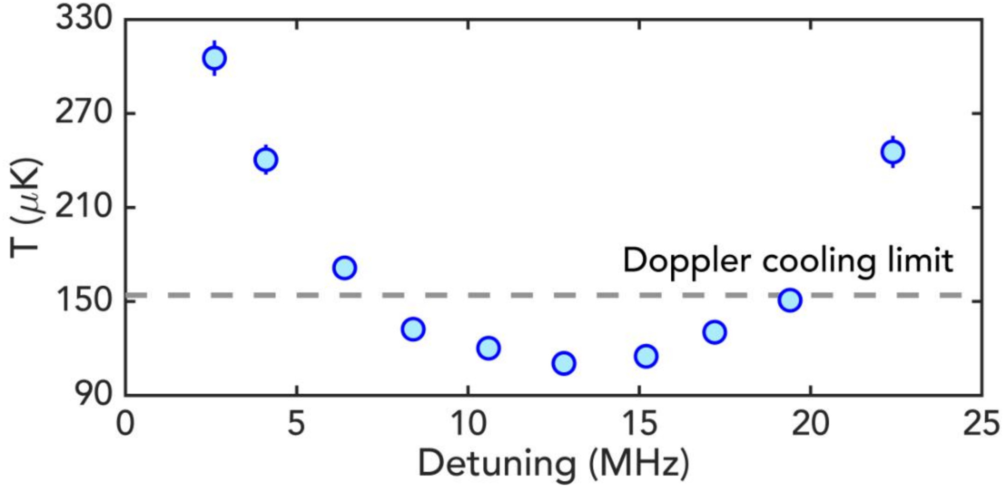
Magneto-optical trapping



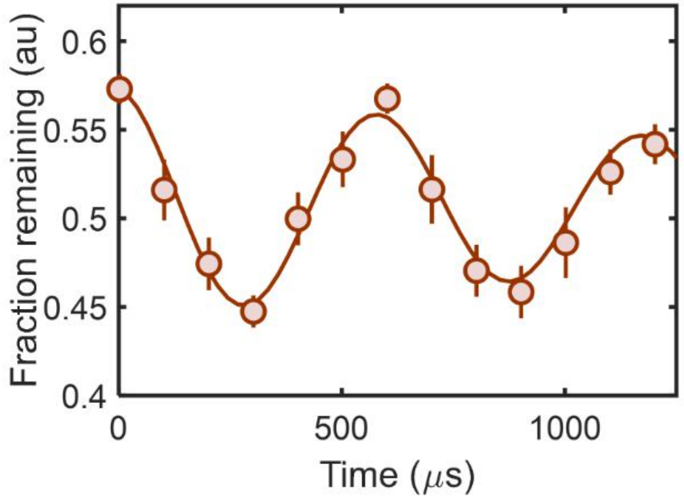
Long lifetimes in optical trap



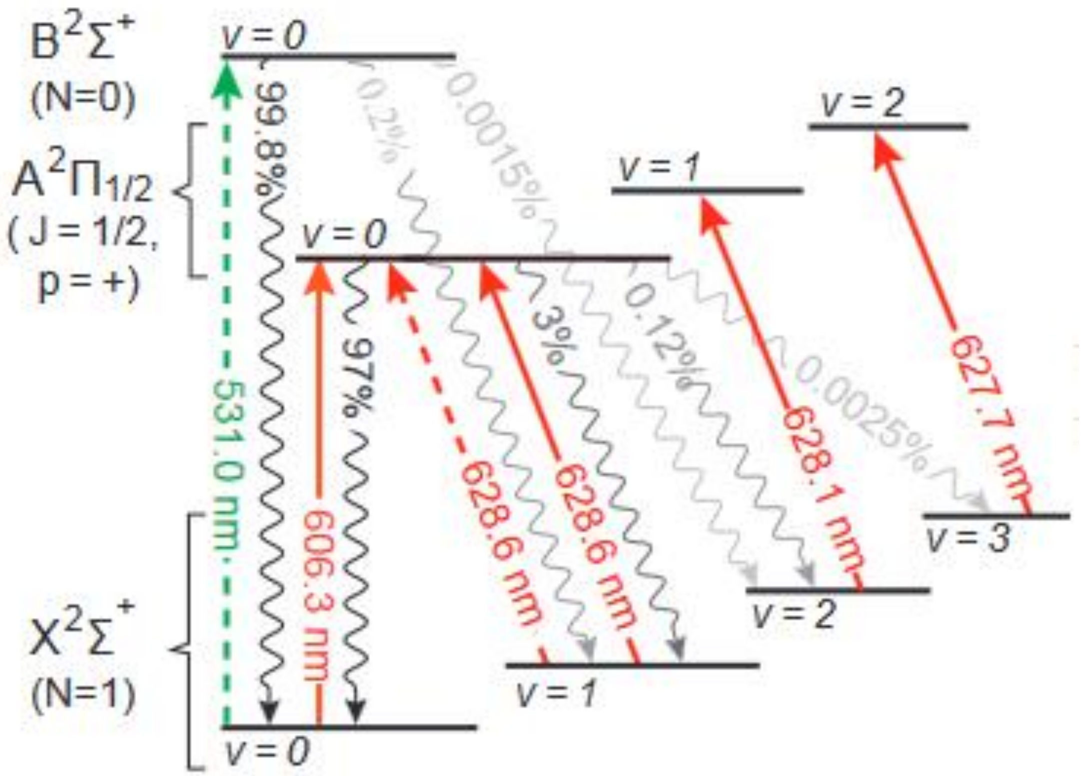
Sub-Doppler cooling



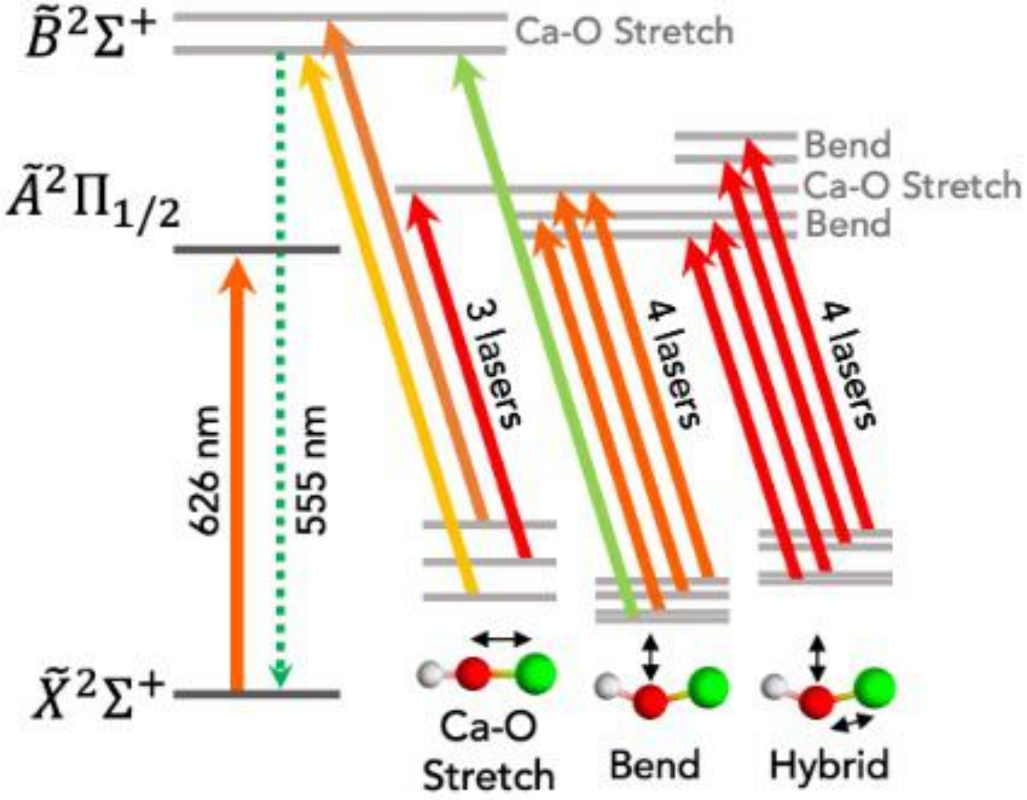
Spin precession in eEDM-sensitive states



The “hard” part: vibrational closure

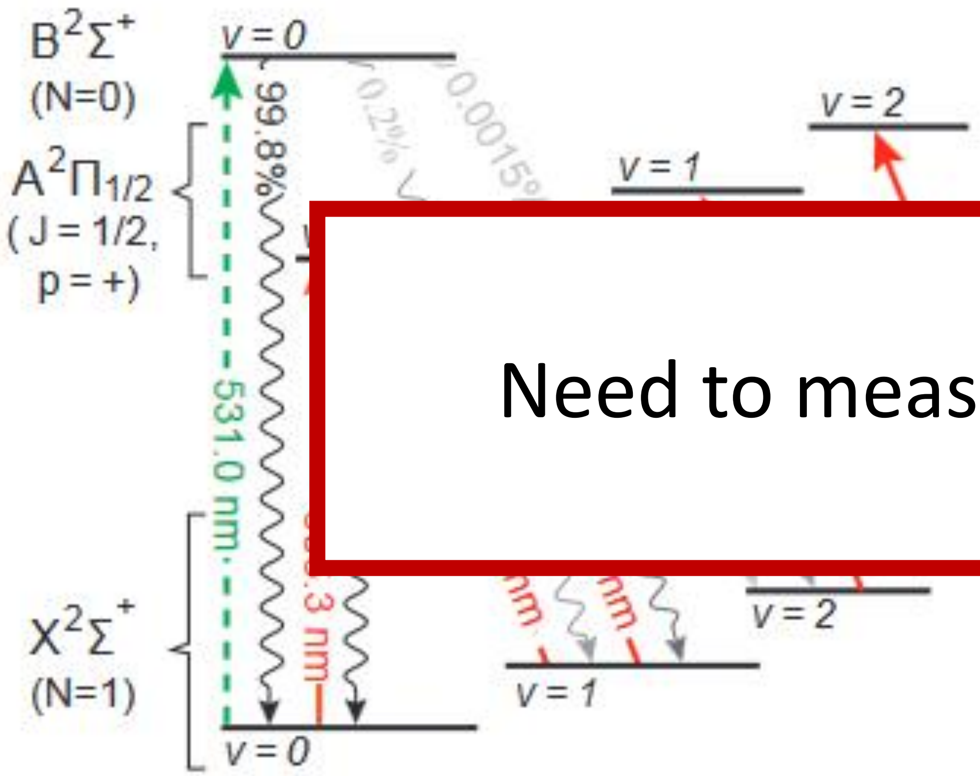


First CaF MOT (Tarbutt group), very similar to SrF MOT scheme

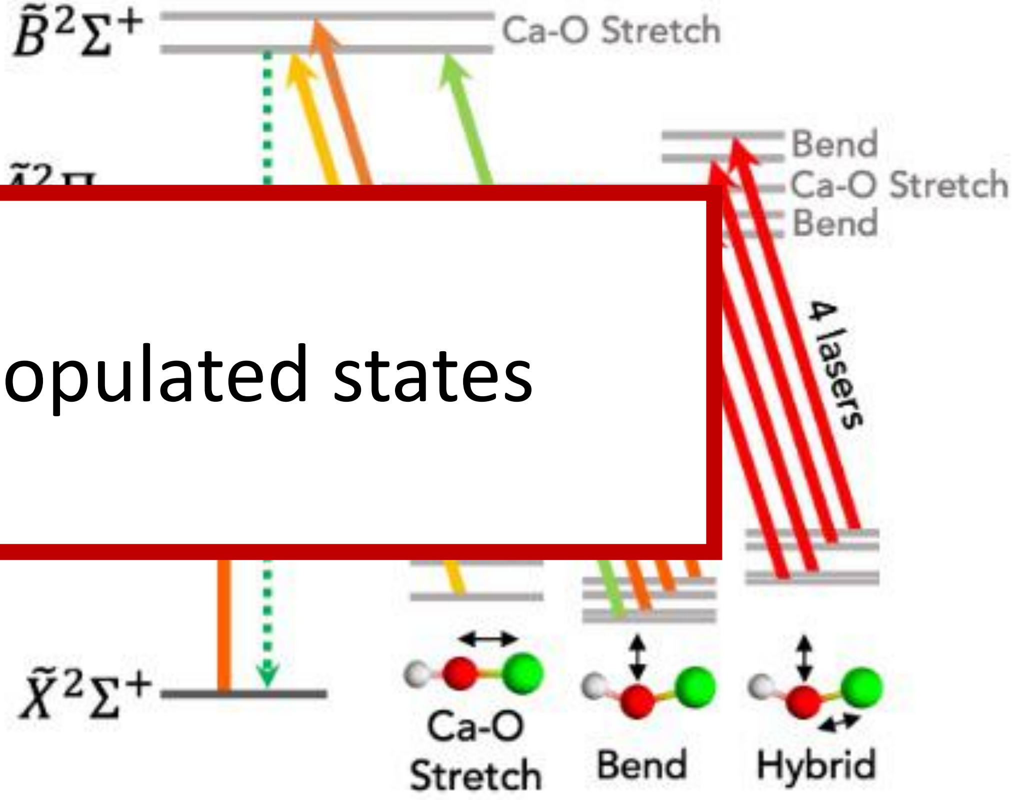


CaOH MOT, involves more bend and hybrid stretch+bend modes than simple models or extrapolations

The “hard” part: vibrational closure



Need to measure the populated states



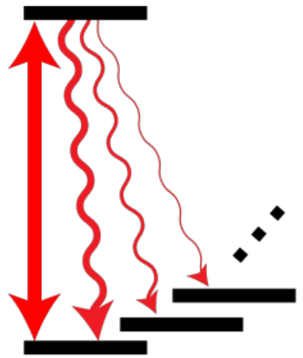
First CaF MOT (Tarbutt group), very similar to SrF MOT scheme

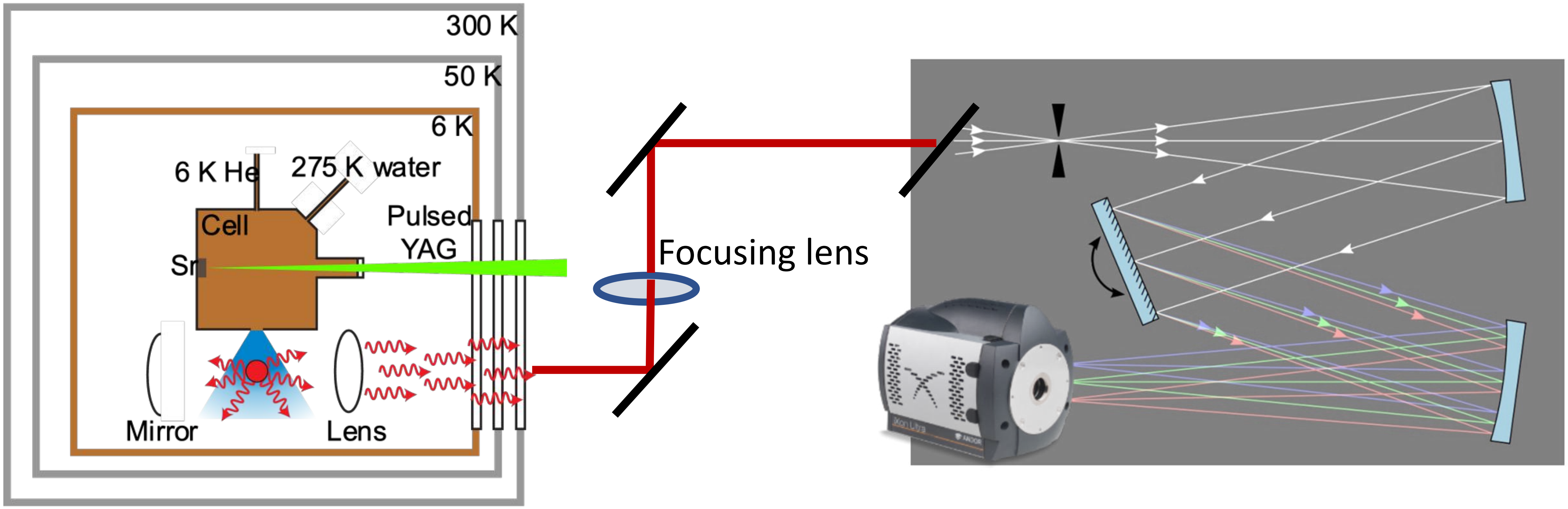
CaOH MOT, involves more bend and hybrid stretch+bend modes than simple models or extrapolations

Vibrational branching ratios of SrOH

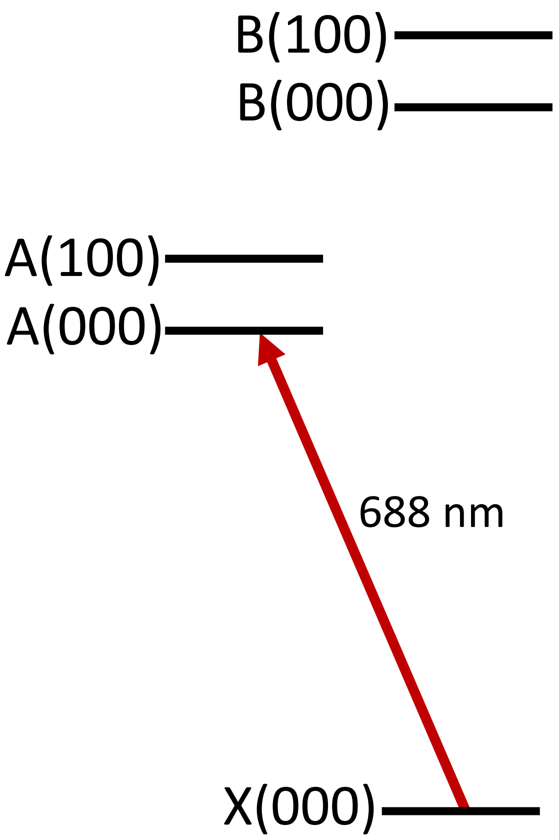
Optically cycle to get many photons per molecule

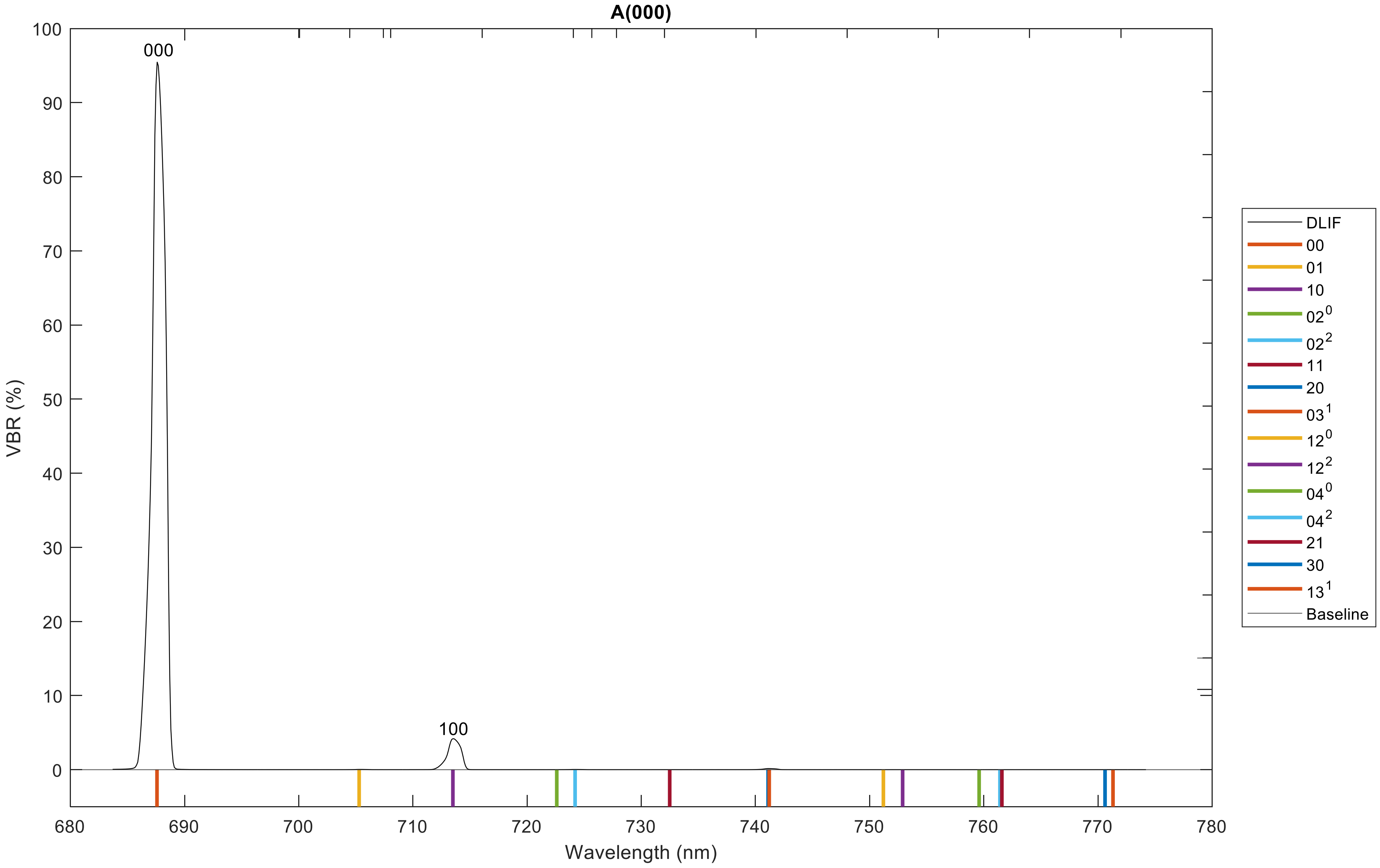
Disperse fluorescence on grating, observe wavelengths of decays



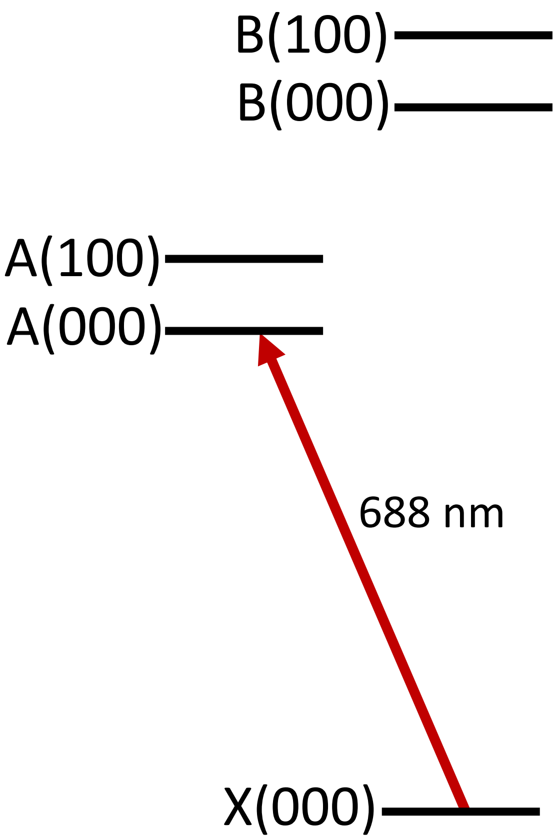


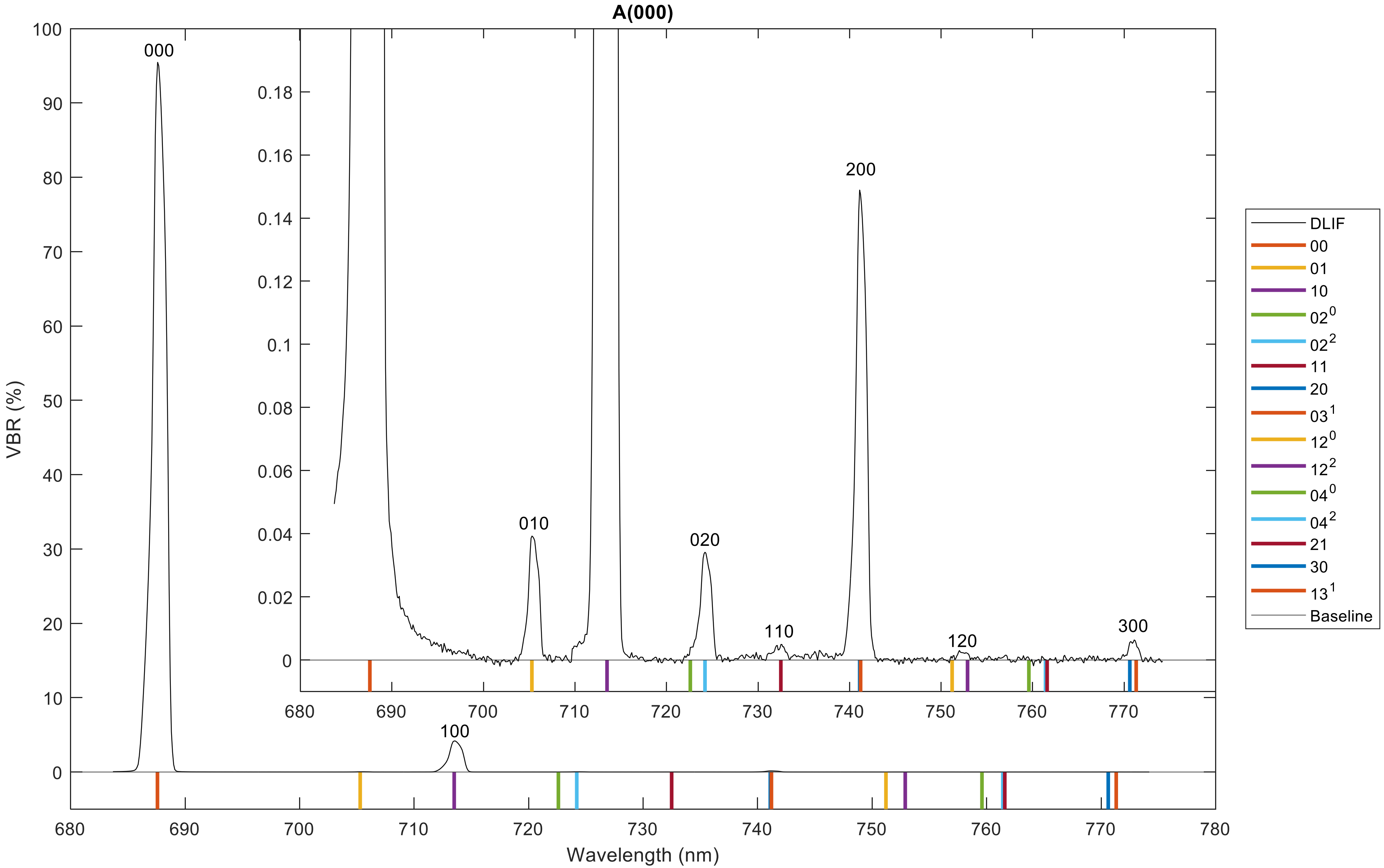
Vibrational branching ratios



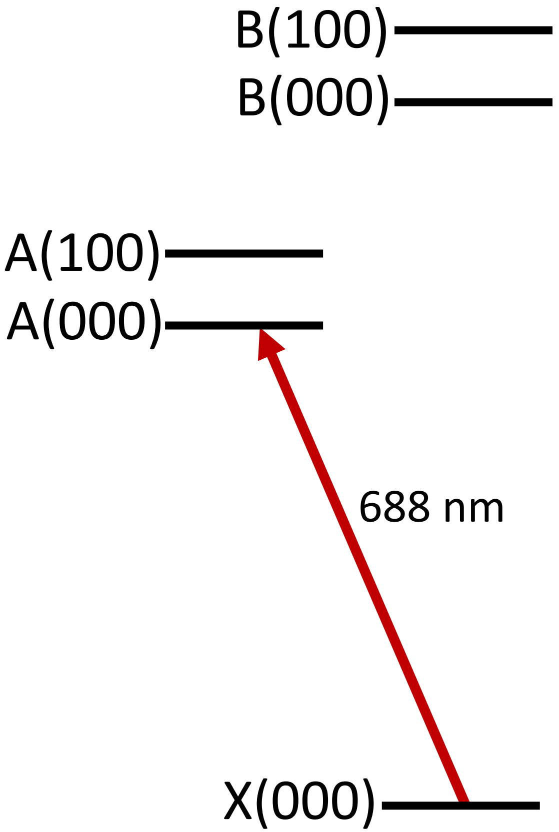


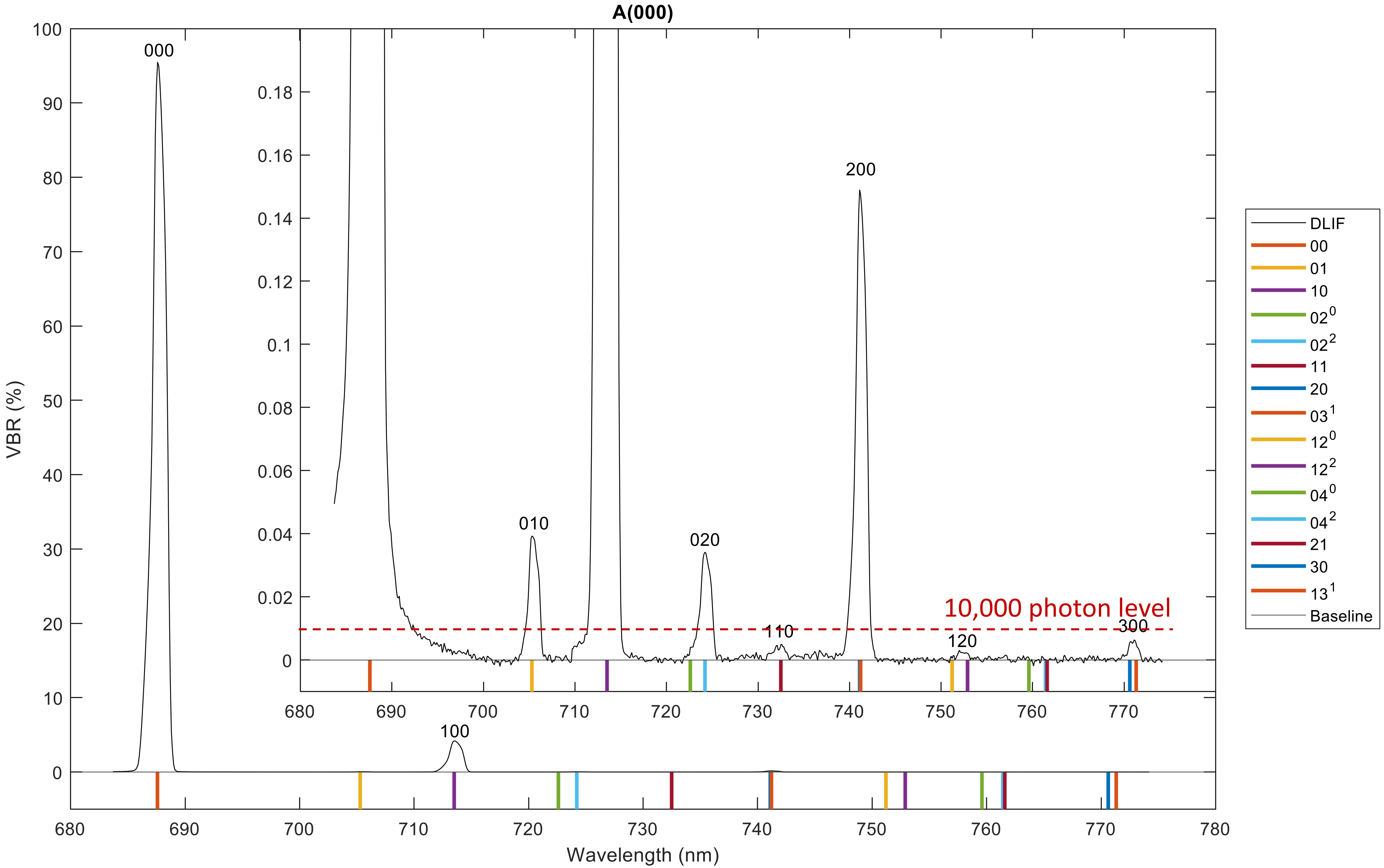
Vibrational branching ratios



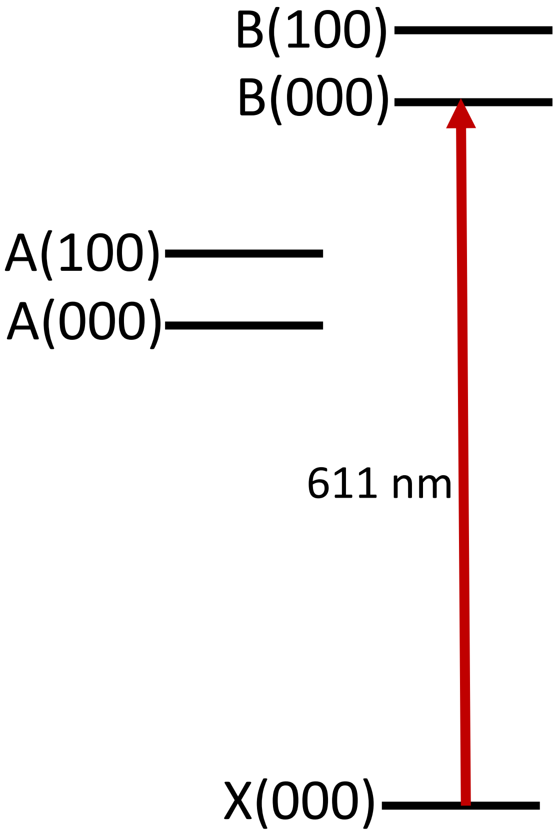


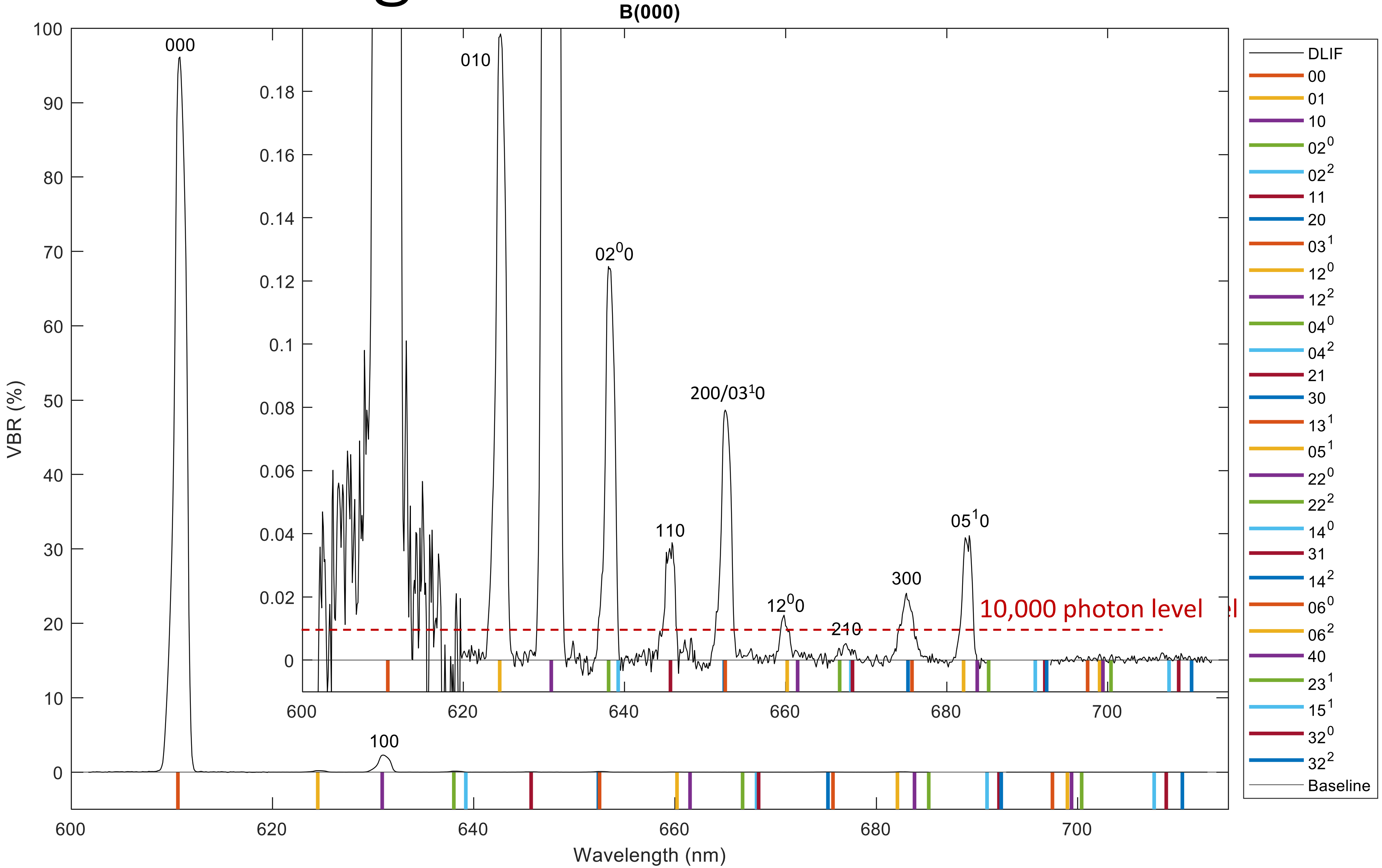
Vibrational branching ratios



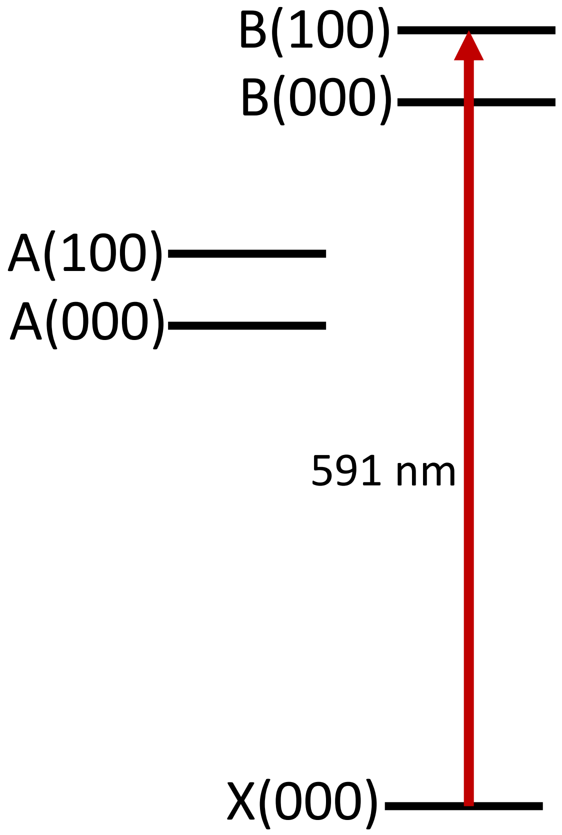


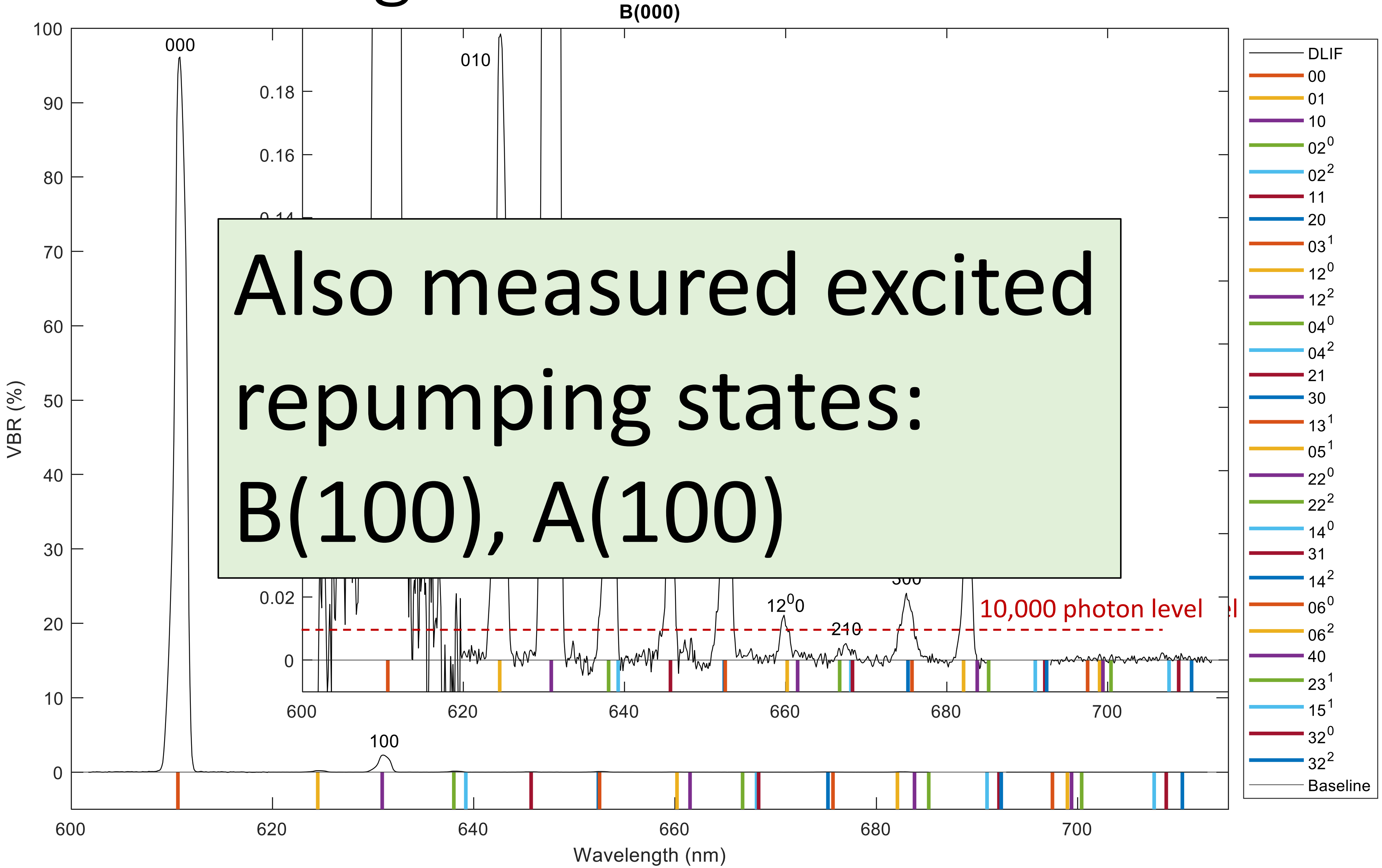
Vibrational branching ratios





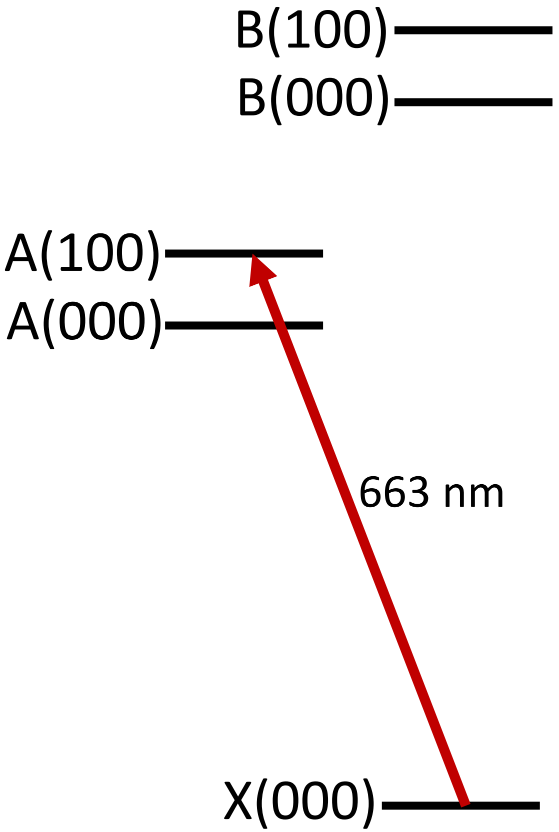
Vibrational branching ratios

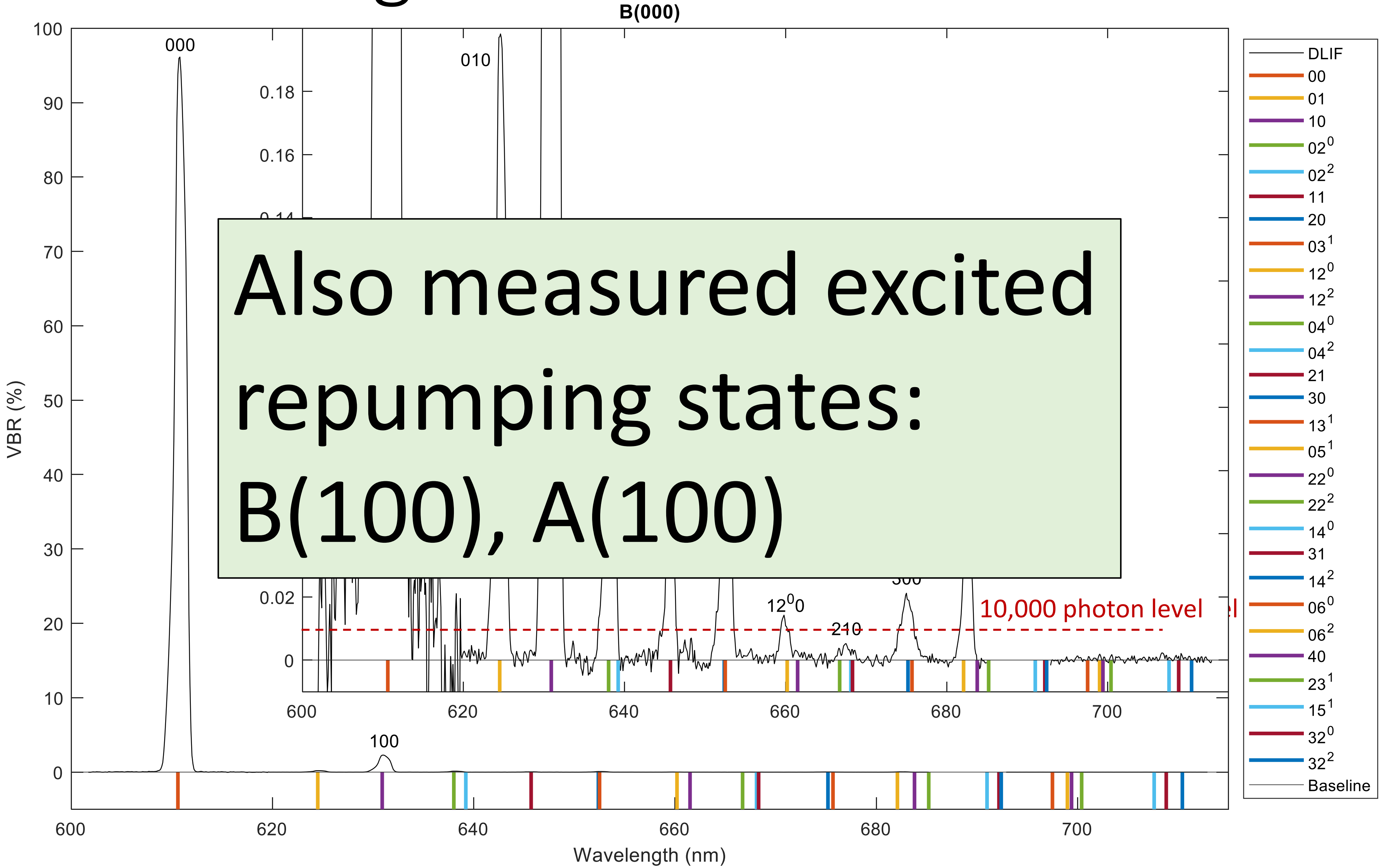




Also measured excited repumping states: B(100), A(100)

Vibrational branching ratios





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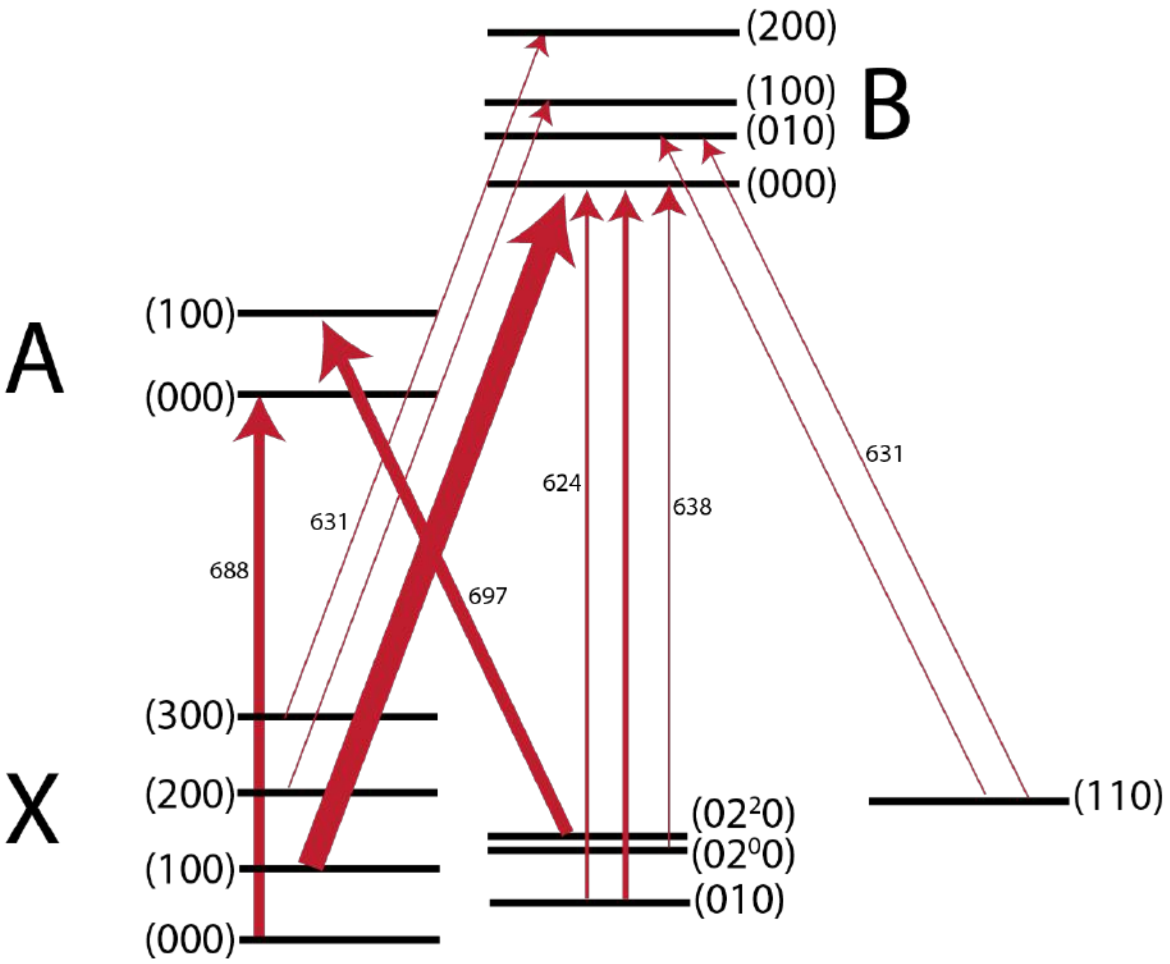
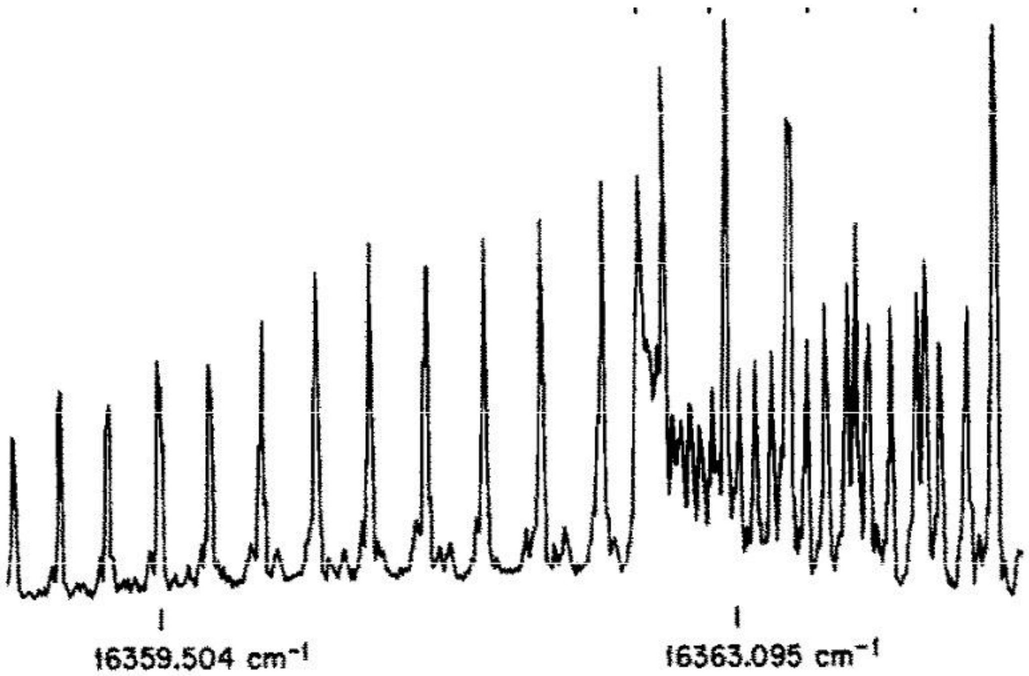
Laser cooling scheme

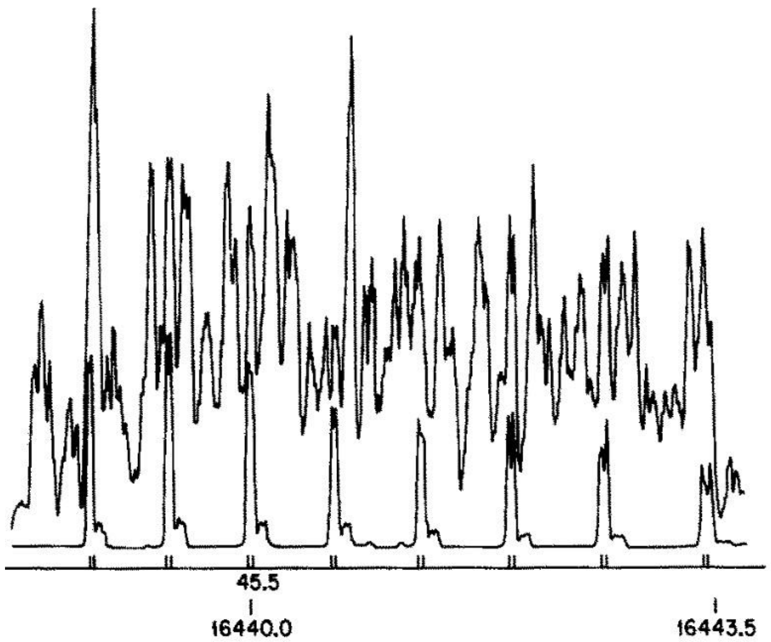


Laser	Ground	Excited	λ (nm)	# photons
1	(000)	A(000)	687.6	23
2	(100)	B(000)	630.9	380
3	(200)	B(100)	630.5	840
4+5	(010)	B(000)	624.5	1300
6	(02^20)	A(100)	696.8	2000
7+8	(110)	B(010)	629.4	3700
9	(02^00)	B(000)	638.0	7200
10	(300)	B(200)	630.0	16000

- Observed B(010) laser cooling state
- Found X(110) manifold, including laser cooling transition
- Looking for B(200) with pulsed dye laser spectroscopy; will use to populate X(300) and search for repumping transition
- All other lines were already known

What “real spectroscopy” looks like





Example: Selected regions of X(000)-B(000) of SrOH spectrum, with assigned lines

J. Mol. Spec. **97** 37 (1983)

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TABLE I

Observed and Calculated Frequencies (in cm^{-1}) of (0, 0, 0)–(0, 0, 0) $B^2\Sigma^+-X^2\Sigma^+$ of SrOH

Line		Frequency	Δν
P_1	(1.5)	16377.002	-0.004
	(2.5)	76.451	0.010
	(3.5)	75.886	0.003
	(4.5)	75.332	0.002
	(5.5)	74.790	0.007
	(6.5)	74.267	0.024
	(7.5)	73.729	0.021
	(8.5)	73.166	-0.014
	(9.5)	72.644	-0.013
	(10.5)	72.141	0.000
	(11.5)	71.633	0.002
	(13.5)	70.633	0.004
	(14.5)	70.131	-0.006
	(15.5)	69.639	-0.012
	(16.5)	69.171	-0.001
	(17.5)	68.693	-0.005
	(18.5)	68.232	0.001
	(19.5)	67.773	0.004
	(20.5)	67.310	-0.004
	(21.5)	66.854	-0.011
	(22.5)	66.443	0.020
	(23.5)	65.989	0.003
	(24.5)	65.559	0.004
	(25.5)	65.135	0.004
	(26.5)	64.712	-0.001
	(27.5)	64.295	-0.006
	(28.5)	63.887	-0.008
	(29.5)	63.498	0.003
	(30.5)	63.095	-0.007
	(31.5)	62.712	-0.003
	(32.5)	62.332	-0.001
	(33.5)	61.950	-0.009
	(34.5)	61.596	0.006
	(35.5)	61.230	0.003
	(36.5)	60.868	-0.003
	(37.5)	60.515	-0.006
	(38.5)	60.169	-0.008
	(39.5)	59.840	0.001
	(40.5)	59.504	-0.004
	(41.5)	59.175	-0.008
	(42.5)	58.864	0.000
	(43.5)	58.551	0.000
	(44.4)	58.242	-0.002
	(45.5)	57.945	0.001
	(46.5)	57.651	0.001
	(47.5	57.368	0.006
	(48.5)	57.081	0.000
	(49.5)	56.800	-0.005
	(50.5)	56.546	0.010
	(51.5)	56.283	0.010
	(52.5)	56.013	-0.004
	(53.5)	55.765	-0.001
	(54.5)	55.520	-0.002
	(55.5)	55.290	0.005
	(56.5)	55.059	0.006
	(57.5)	54.823	-0.005
	(58.5)	54.616	0.007
	(59.5)	54.399	0.003
	(60.5)	54.190	-0.000
	(61.5)	53.995	0.005
	(62.5)	53.791	-0.005
	(63.5)	53.618	0.009
	(64.5)	53.424	-0.004
	(65.5)	53.254	0.001
	(66.5)	53.085	0.001
	(67.5)	52.922	0.000
	(68.5)	52.771	0.005
	(69.5)	52.612	-0.004

Line		Frequency	Δν
P_1	(70.5)	16352.471	-0.002
	(71.5)	52.332	-0.004
	(72.5)	52.202	-0.003
	(73.5)	52.088	0.007
	(74.5)	51.973	0.010
	(75.5)	51.856	0.005
	(76.5)	51.741	-0.005
	(77.5)	51.644	-0.003
	(78.5)	51.522	-0.002
	(79.5)	51.472	0.004
	(80.5)	51.390	0.003
	(81.5)	51.310	-0.004
	(82.5)	51.239	-0.007
	(83.5)	51.182	-0.003
	(84.5)	51.130	-0.001
	(85.5)	51.078	-0.004
	(86.5)	51.041	0.001
R_1	(0.5)	16377.943	0.006
	(1.5)	78.375	0.000
	(2.5)	78.831	0.012
	(3.5)	79.268	-0.001
	(4.5)	79.721	-0.004
	(5.5)	80.176	-0.011
	(6.5)	80.644	-0.011
	(7.5)	81.126	-0.004
	(8.5)	81.638	0.028
	(9.5)	82.100	0.004
	(10.5)	82.593	0.005
	(11.5)	83.083	-0.003
	(12.5)	83.592	0.002
	(13.5)	84.095	-0.005
	(14.5)	84.614	-0.003
	(15.5)	85.138	-0.001
	(16.5)	85.668	0.001
	(17.5)	86.219	0.018
	(18.5)	86.752	0.012
	(19.5)	87.287	0.001
	(20.5)	87.836	-0.002
	(21.5)	88.389	-0.007
	(22.5)	88.957	-0.002
	(23.5)	89.532	0.003
	(24.5)	90.104	-0.000
	(25.5)	90.684	-0.002
	(26.5)	91.276	0.003
	(27.5)	91.860	-0.006
	(28.5)	92.461	-0.004
	(29.5)	93.077	0.007
	(30.5)	93.691	0.010
	(31.5)	94.292	-0.006
	(32.5)	94.918	-0.002
	(33.5)	95.554	0.005
	(34.5)	96.193	0.010
	(35.5)	96.829	0.006
	(36.5)	97.461	-0.008
	(37.5)	98.137	0.016
	(38.5)	98.781	0.003
	(39.5)	99.438	-0.004
	(40.5)	16400.107	-0.004
	(41.5)	00.779	-0.007
	(42.5)	01.470	0.003
	(43.5)	02.152	-0.001
	(44.5)	02.851	0.005
	(45.5)	03.556	0.012
	(46.5)	04.227	-0.021
	(47.5)	04.954	-0.003
	(48.5)	05.667	-0.006
	(49.5)	06.380	-0.014

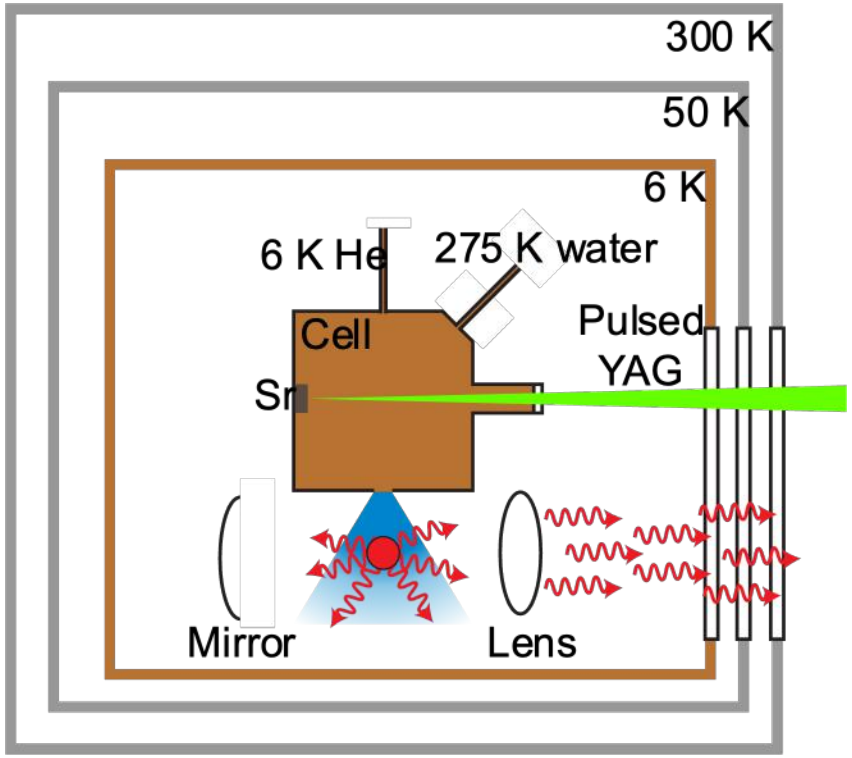
ANALYSIS OF $B^2\Sigma^+-X^2\Sigma^+$ OF SrOH AND SrOD 43

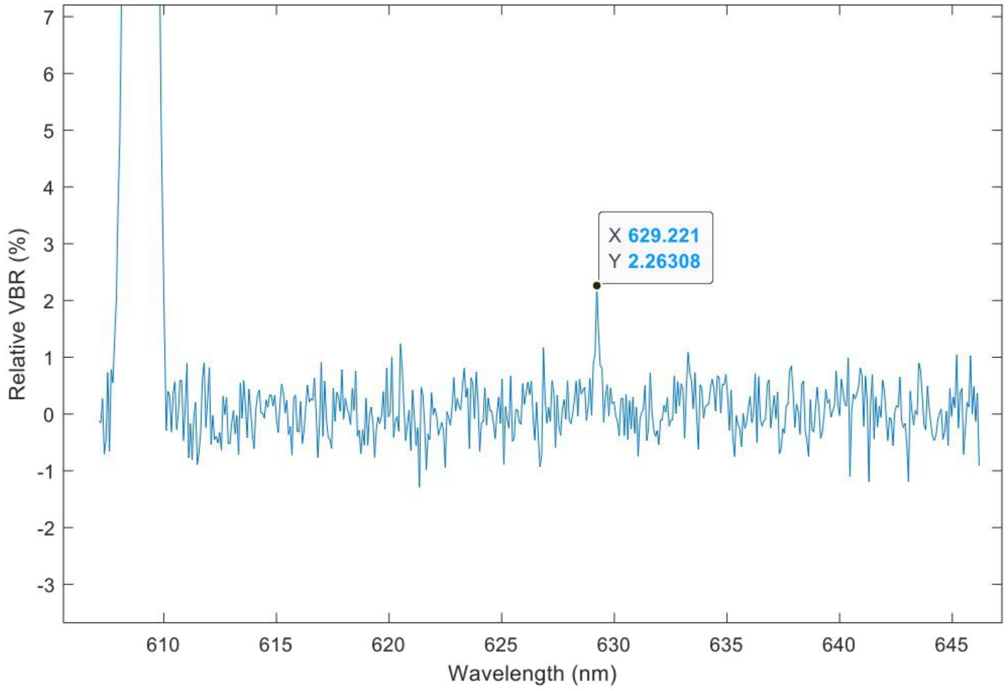
TABLE I—*Continued*

Line		Frequency	Δν
R_1	(50.5)	16407.116	-0.005
	(51.5)	07.883	0.029
	(52.5)	08.596	0.004
	(53.5)	09.339	0.003
	(54.5)	10.088	0.002
	(55.5)	10.838	-0.004
	(56.5)	11.605	0.002
	(57.5)	12.369	-0.001
	(58.5)	13.136	-0.006
	(59.5)	13.900	-0.021
	(60.5)	14.721	0.016
	(61.5)	15.475	-0.019
	(62.5)	16.283	-0.007
	(63.5)	17.084	-0.007
	(64.5)	17.896	-0.001
	(65.5)	18.724	0.015
	(66.5)	19.520	-0.007
	(67.5)	20.349	-0.002
	(68.5)	21.187	0.007
Q_2	(0.5)	16378.179	0.025
P_2	(1.5)	16376.659	-0.000
	(2.5)	76.245	-0.000
	(3.5)	75.841	0.003
	(4.5)	75.430	-0.006
	(5.5)	75.039	-0.001
	(6.5)	74.653	0.002
	(7.5)	74.267	-0.000
	(8.5)	73.911	0.021
	(9.5)	73.525	0.006
	(10.5)	73.166	0.013
	(11.5)	72.805	0.011
	(12.5)	72.431	-0.010
	(13.5)	72.102	0.008
	(14.5)	71.757	0.003
	(15.5)	71.408	-0.011
	(16.5)	71.079	-0.012
	(17.5)	70.767	-0.001
	(18.5)	70.449	-0.003
	(19.5)	70.131	-0.011
	(20.5)	69.833	-0.005
	(21.5)	69.538	-0.002
	(22.5)	69.253	0.005
	(23.5)	68.965	0.002
	(24.5)	68.693	0.009
	(25.5)	68.404	-0.006
	(26.5)	68.143	-0.000
	(27.5)	67.886	0.004
	(28.5)	67.624	-0.004
	(29.5)	67.369	-0.010
	(30.5)	67.133	-0.004
	(31.5)	66.888	-0.013
	(32.5)	66.653	-0.018
	(33.5)	66.443	-0.004
	(34.5)	66.227	-0.003
	(35.5)	66.017	-0.001
	(36.5)	65.811	-0.002
	(37.5)	65.618	0.004
	(38.5)	65.417	-0.005
	(39.5)	65.243	0.008
	(40.5)	65.051	-0.004
	(41.5)	64.878	-0.003
	(42.5)	64.712	-0.001
	(43.5)	64.555	0.003
	(44.5)	64.392	0.004
	(45.5)	64.250	0.003
	(46.5)	64.106	0.002
	(47.5)	63.965	-0.003
	(48.5)	63.834	-0.003
	(49.5)	63.715	0.002

Line		Frequency	Δν
P_2	(50.5)	16363.596	0.000
	(51.5)	63.498	0.014
	(52.5)	63.389	0.010
	(53.5)	63.283	0.003
	(54.5)	63.182	-0.005
	(55.5)	63.095	-0.005
	(56.5)	63.019	-0.001
	(57.5)	62.947	0.001
	(58.5)	62.878	-0.001
	(59.5)	62.820	0.003
	(60.5)	62.772	0.010
	(61.5)	62.712	-0.001
R_2	(0.5)	16378.743	0.006
	(1.5)	79.330	0.004
	(2.5)	79.921	-0.000
	(3.5)	80.521	-0.001
	(4.5)	81.126	-0.004
	(5.5)	81.753	0.010
	(6.5)	82.371	0.009
	(7.5)	82.983	-0.004
	(8.5)	83.592	-0.026
	(9.5)	84.247	-0.008
	(10.5)	84.899	0.001
	(11.5)	85.547	-0.000
	(12.5)	86.219	0.016
	(13.5)	86.866	0.002
	(14.5)	87.519	-0.012
	(15.5)	88.195	-0.009
	(16.5)	88.877	-0.006
	(17.5)	89.566	-0.001
	(18.5)	90.253	-0.005
	(19.5)	90.953	-0.002
	(20.5)	91.645	-0.013
	(21.5)	92.363	-0.003
	(22.5)	93.077	-0.004
	(23.5)	93.814	0.013
	(24.5)	94.518	-0.010
	(25.5)	95.247	-0.013
	(26.5)	95.990	-0.008
	(27.5)	96.747	0.005
	(28.5)	97.477	-0.015
	(29.5)	98.246	-0.002
	(30.5)	99.006	-0.004
	(31.5)	99.773	-0.004
	(32.5)	16400.548	-0.002
	(33.5)	01.322	-0.008
	(34.5)	02.109	-0.006
	(35.5)	02.898	-0.008
	(36.5)	03.712	0.010
	(37.5)	04.525	0.020
	(38.5)	05.323	0.010
	(39.5)	06.125	-0.003
	(40.5)	06.939	-0.009
	(41.5)	07.771	-0.002
	(42.5)	08.596	-0.009
	(43.5)	09.448	0.006
	(44.5)	10.287	0.001
	(45.5)	11.141	0.007
	(46.5)	11.981	-0.008
	(47.5)	12.852	0.002
	(48.5)	13.711	-0.005
	(49.5)	14.600	0.012
	(50.5)	15.475	0.010
	(51.5)	16.348	-0.001
	(52.5)	17.233	-0.005
	(53.5)	18.129	-0.004
	(54.5)	19.031	-0.002
	(55.5)	19.924	-0.016
	(56.5)	20.858	0.007
	(57.5)	21.772	0.003
	(58.5)	22.726	0.034

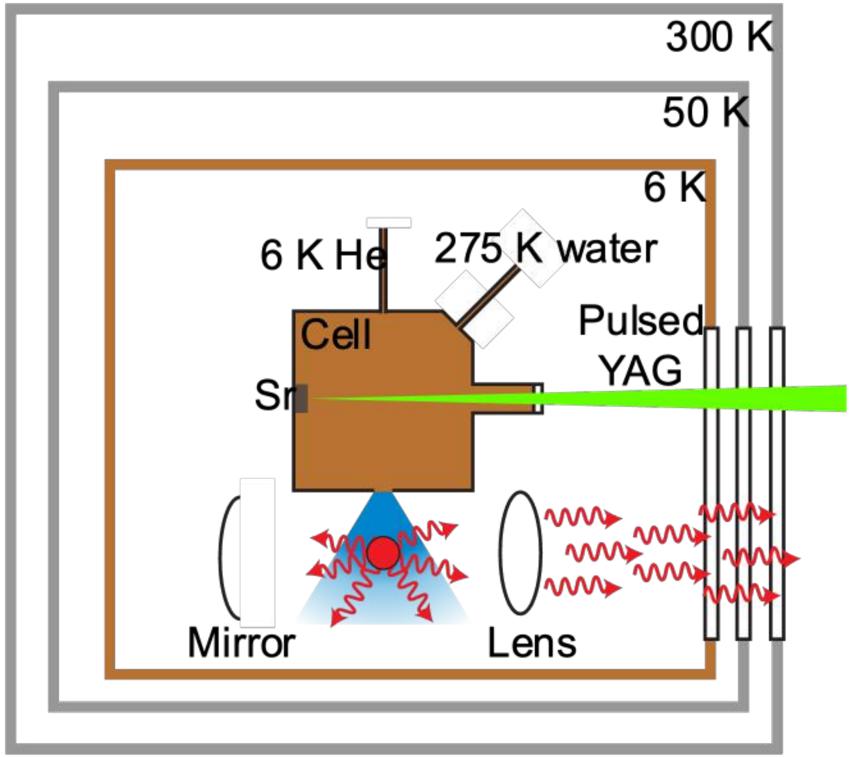
What *our* spectroscopy looks like: X(110)-B(010)

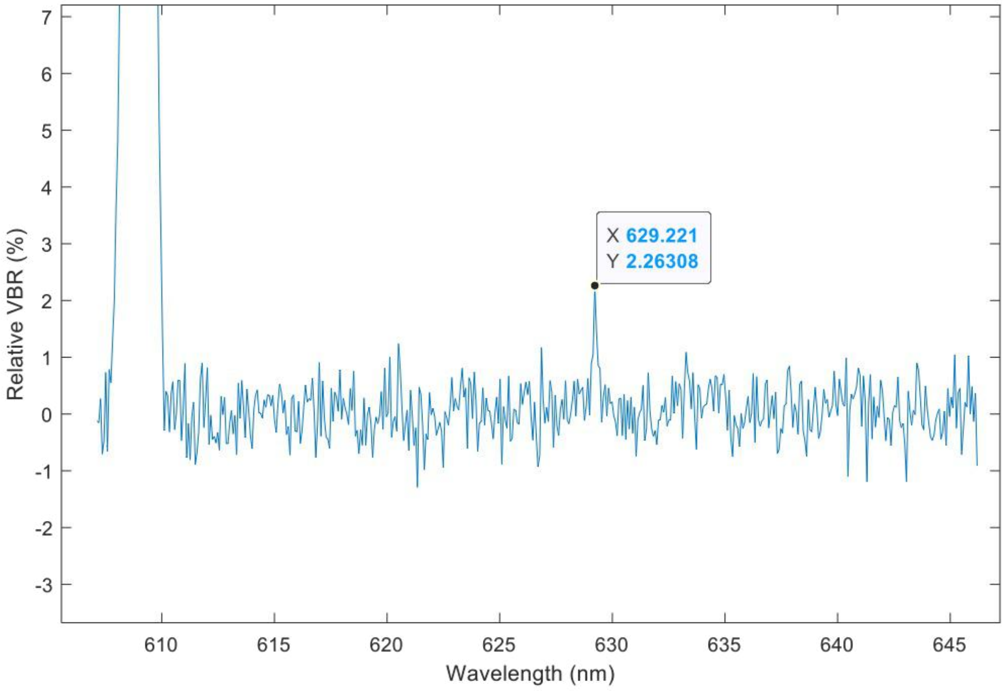




First excite X(000)-B(010) at 596 nm, observe decays to X(010) at 609 nm and X(110) at 629 nm

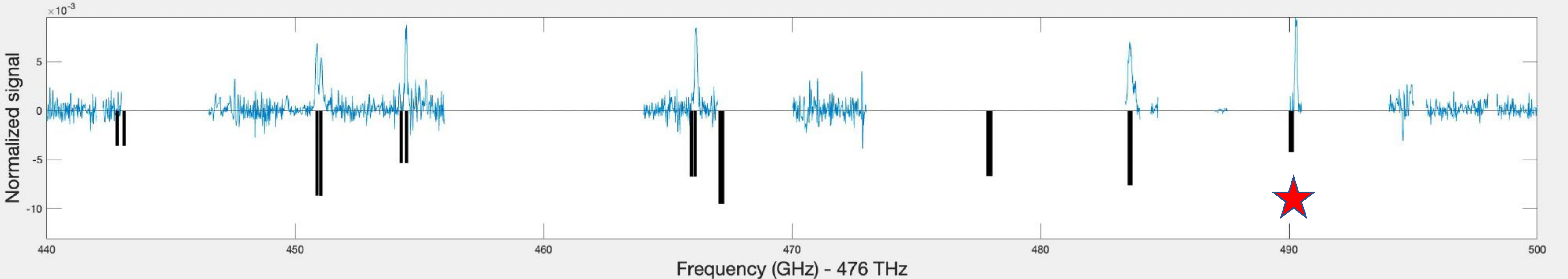
What *our* spectroscopy looks like: X(110)-B(010)



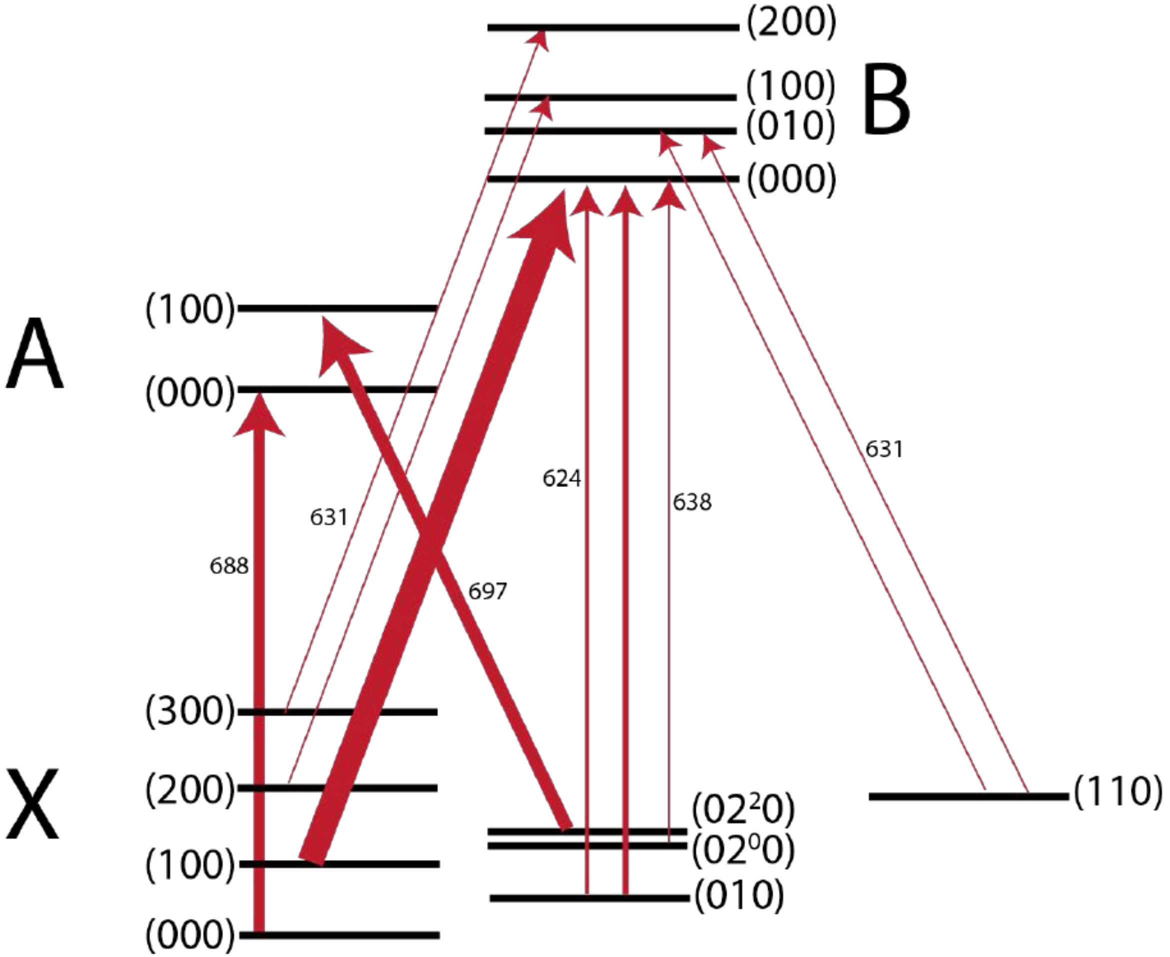


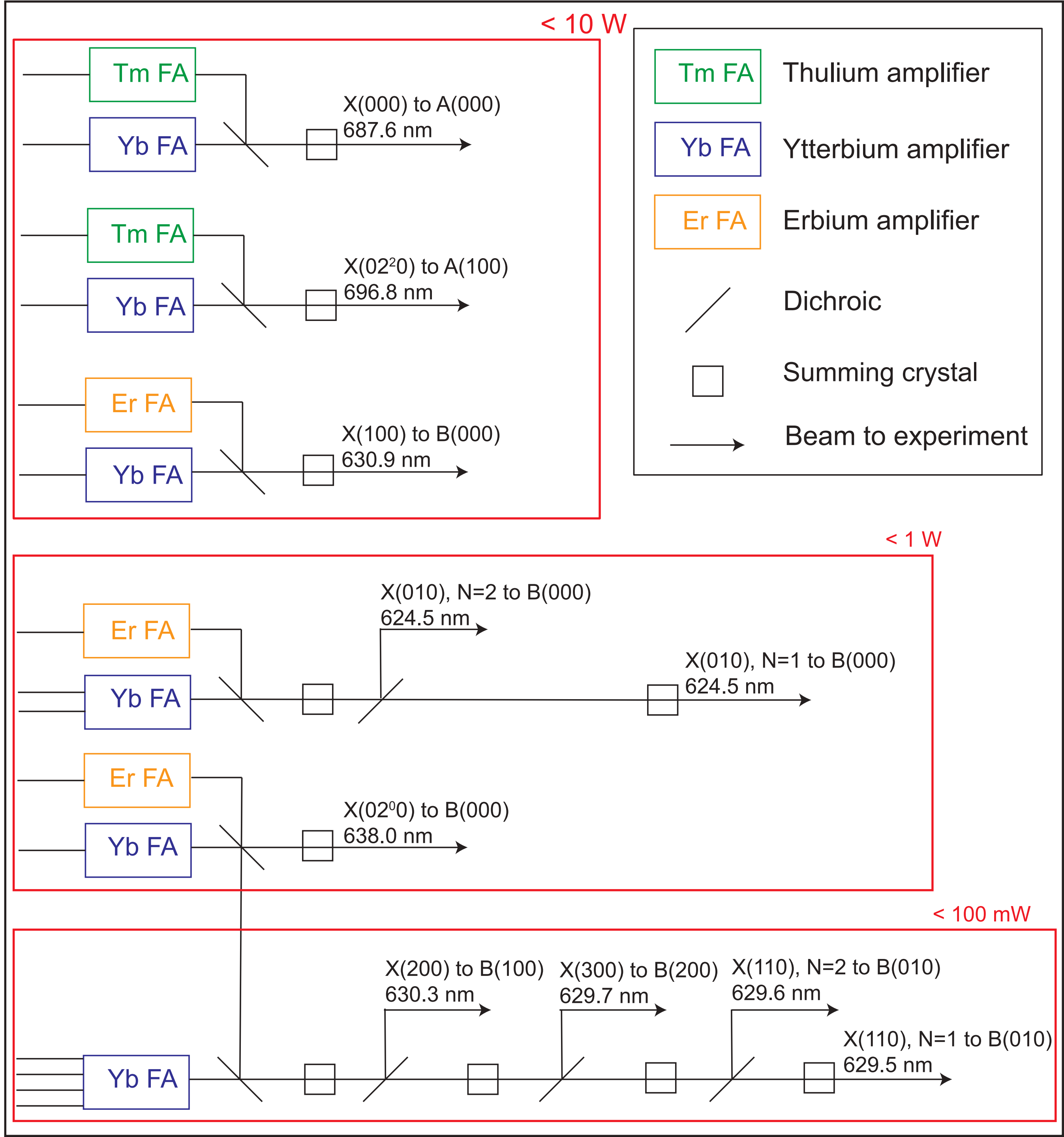
First excite X(000)-B(010) at 596 nm, observe decays to X(010) at 609 nm and X(110) at 629 nm

Scan excitation laser around X(110) decay wavelength

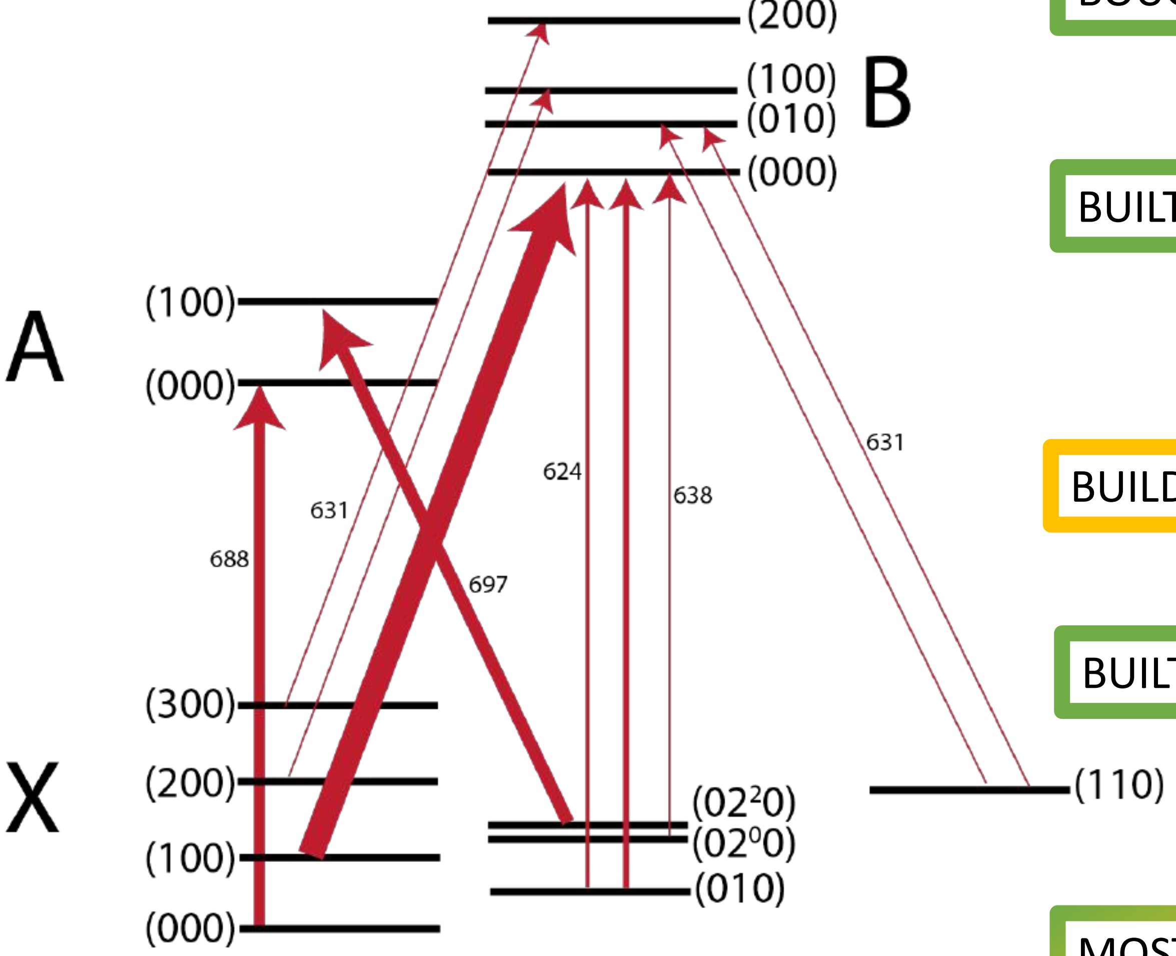


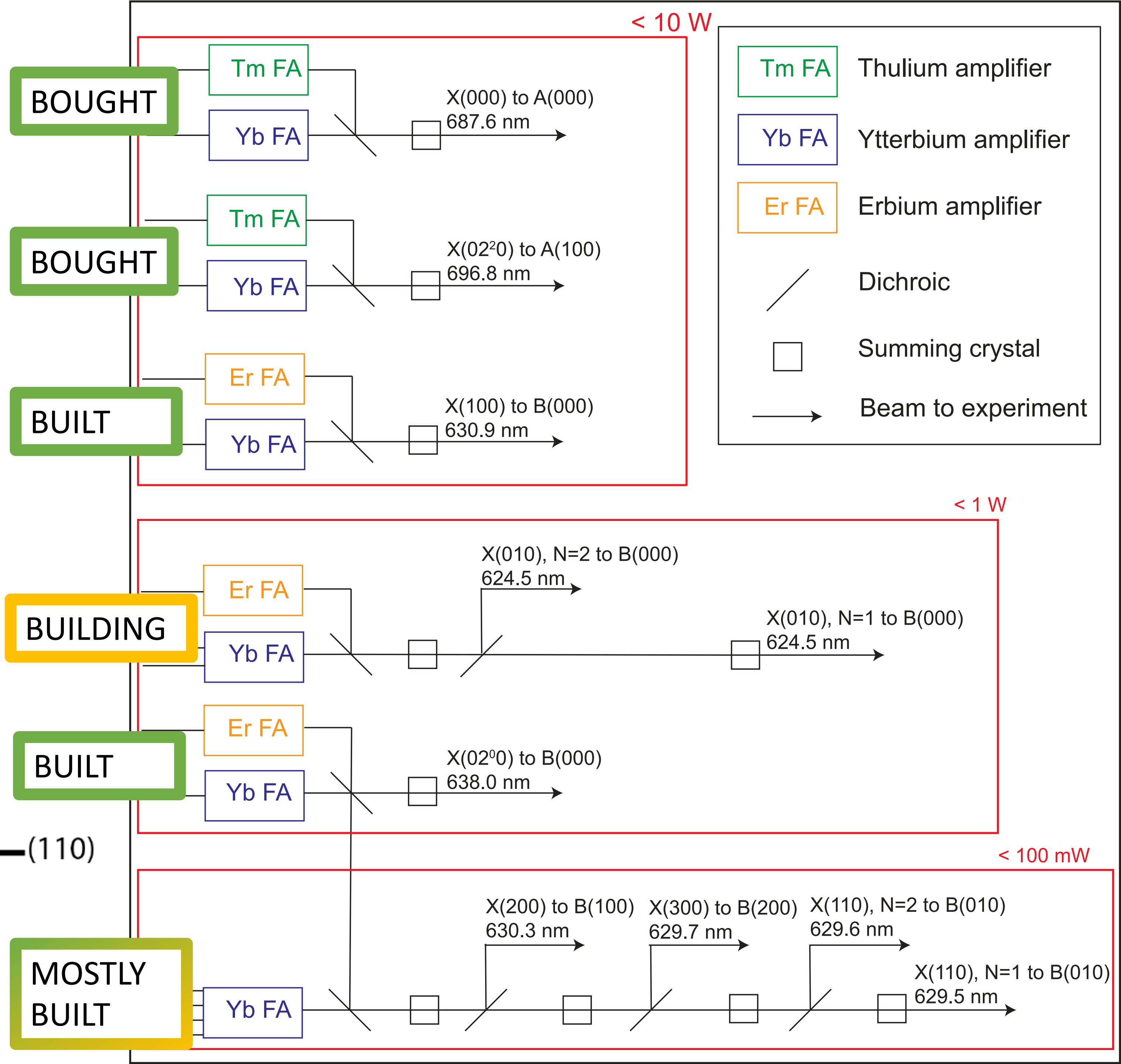
Laser cooling Implementation





Laser cooling Implementation





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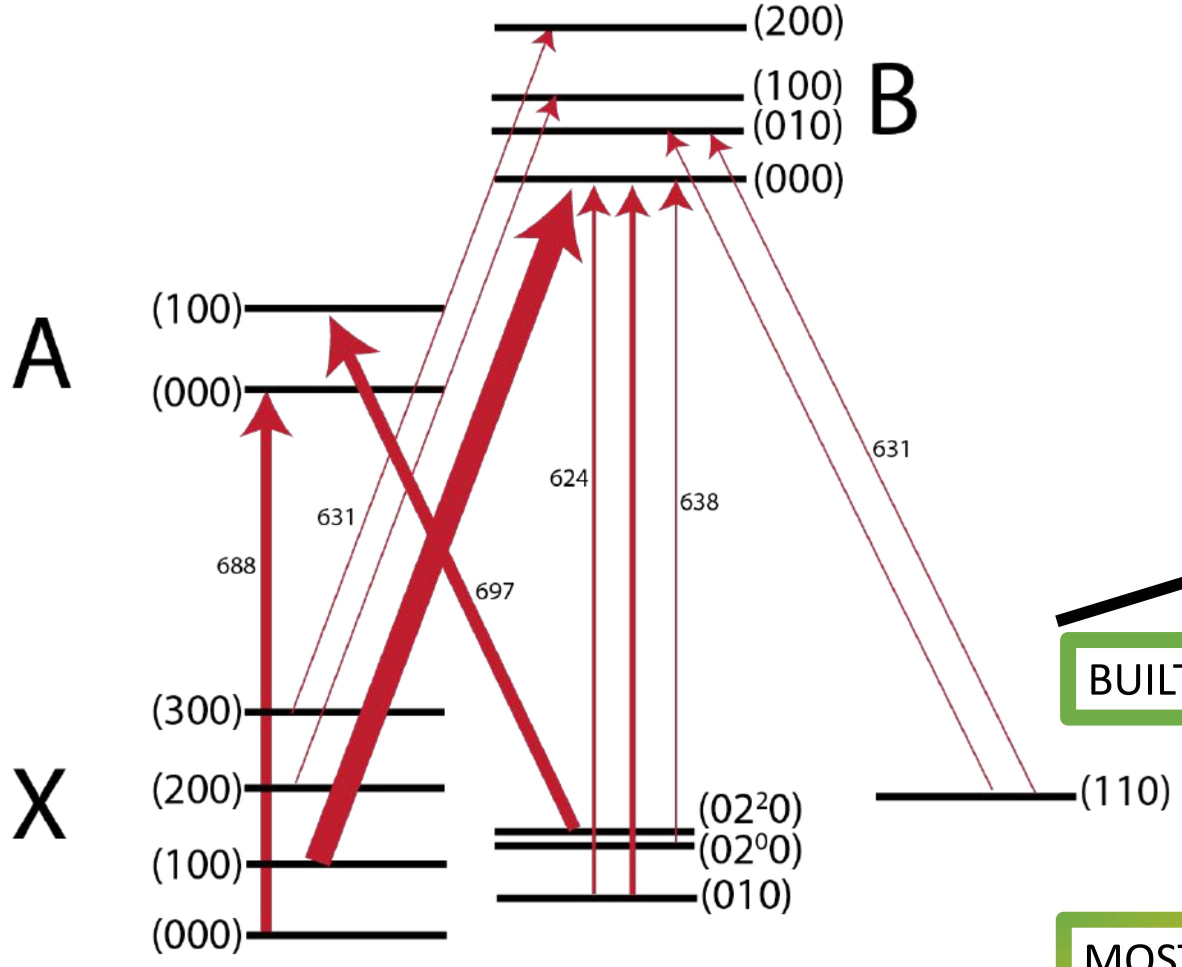
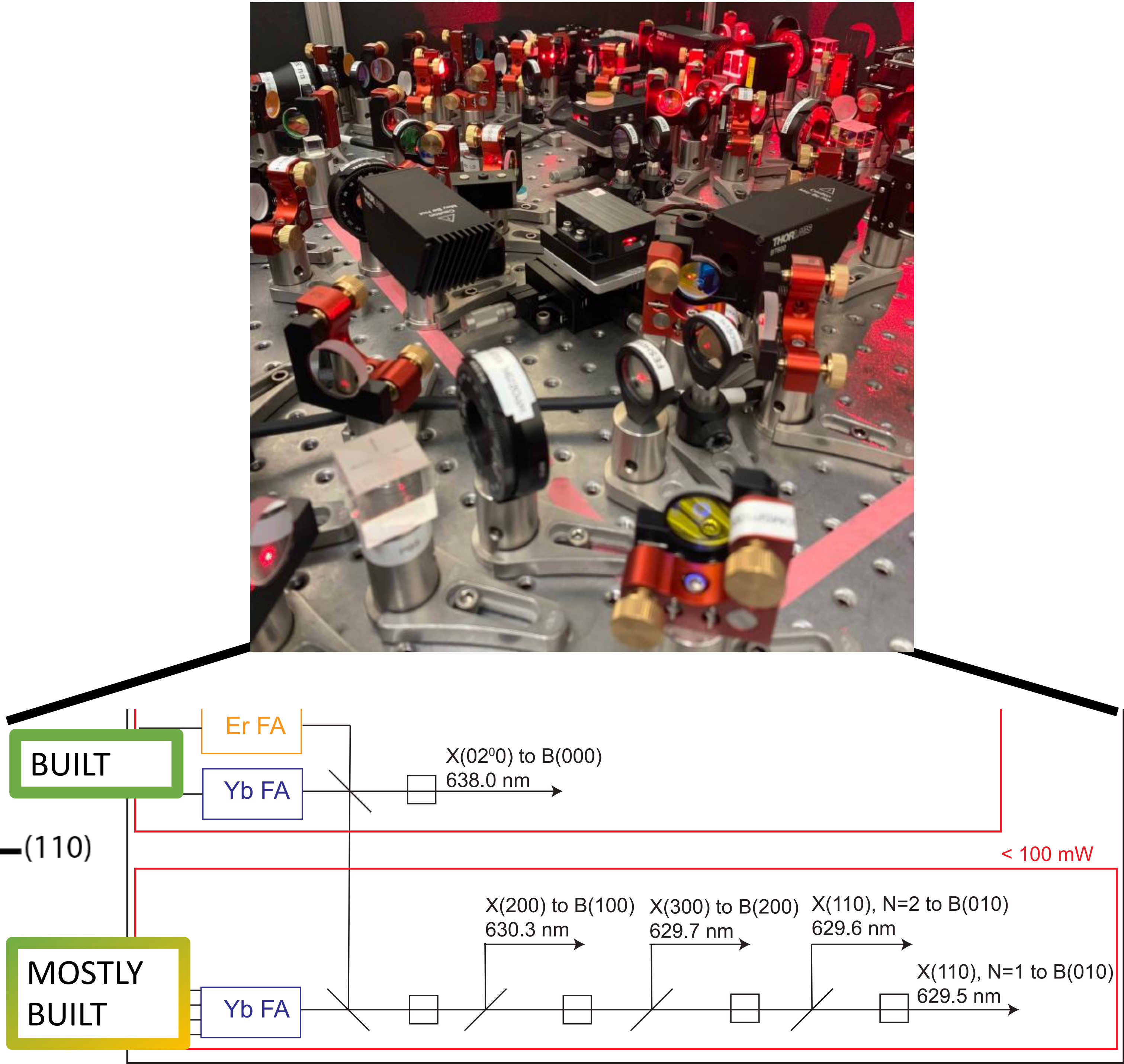
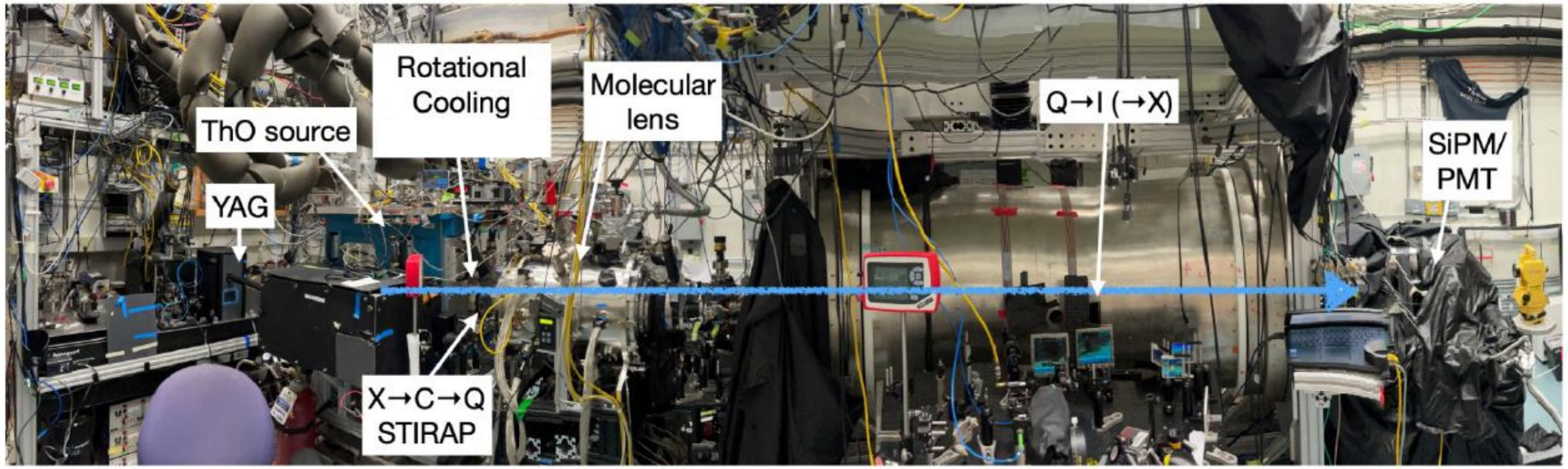


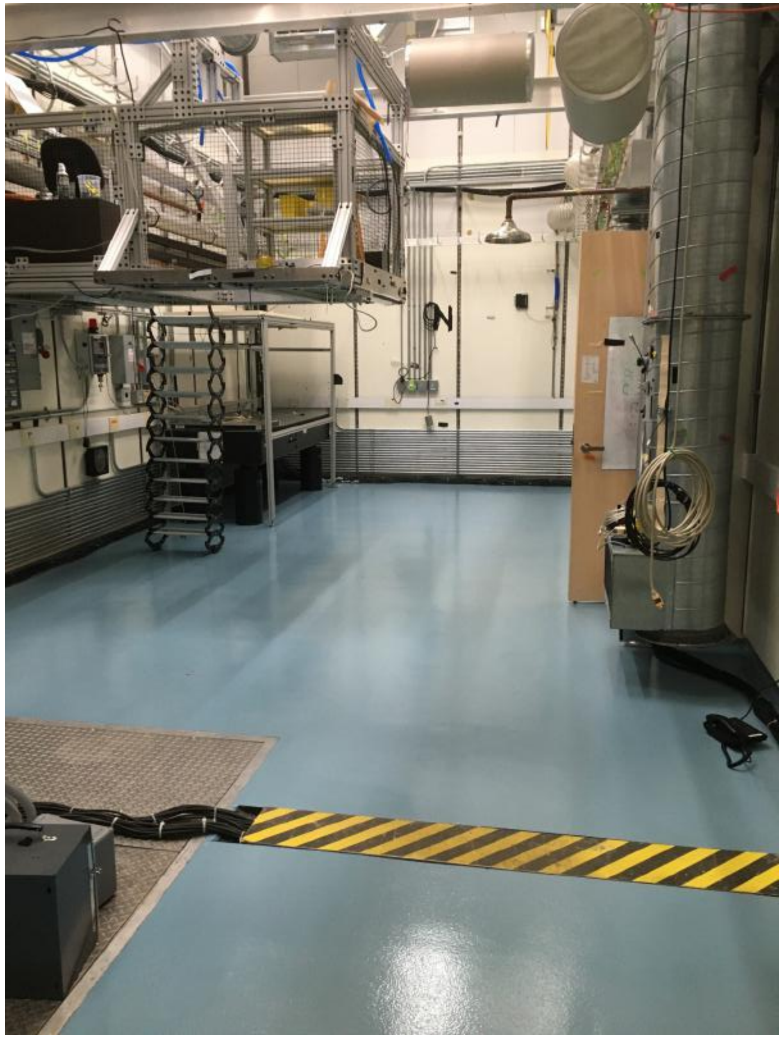


Building the lab



~September 2022: "our" lab was the ACME lab at Harvard [photo 2021]

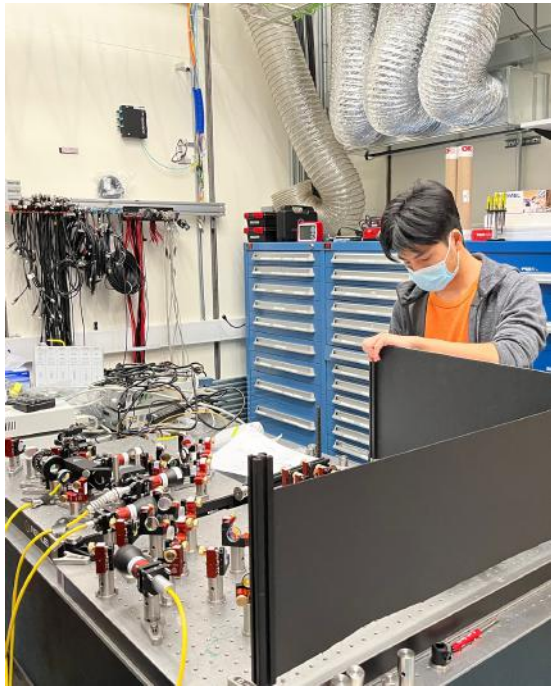
ACME has since moved equipment for ACME III to Northwestern to make an improved measurement, and select equipment has been relocated to a new lab space at Harvard for parallel development/test work



~October 2022: lab cleaned out

Building the lab

Beam source/beam line/MOT
table, control station



High-power
laser table

Adjoining “quiet” optics room
(wavemeter, lasers <1 W)

Building the lab

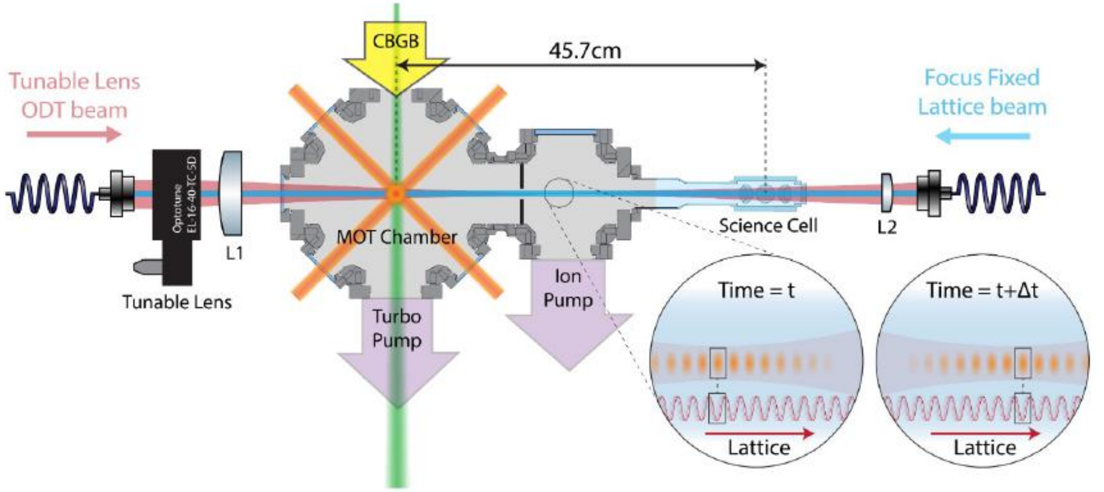
Beam source/beam line/MOT table, control station

Near-term prospects

- We expect a MOT of SrOH this year
- Past experience:

  in a MOT, everything is an atom

- Will optically trap, transfer to science chamber (done already for CaF), perform spin precession (similar to work in CaOH)
- Anticipate competitive eEDM sensitivity to current published limit (ACME II) due to long coherence times and large trap numbers (thousands)



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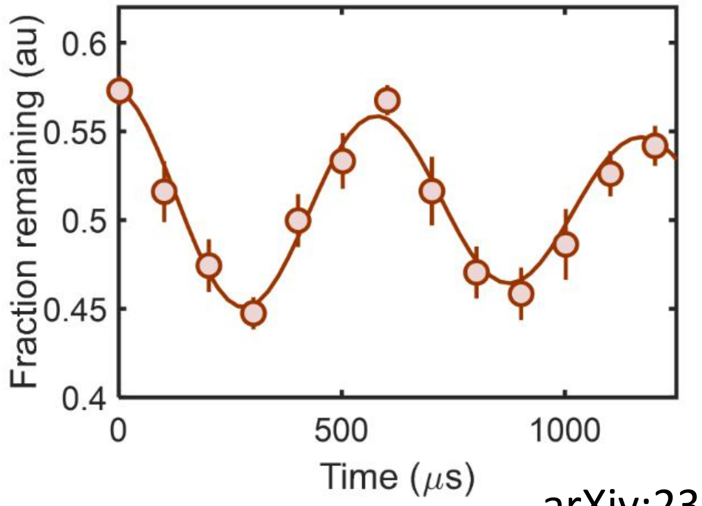
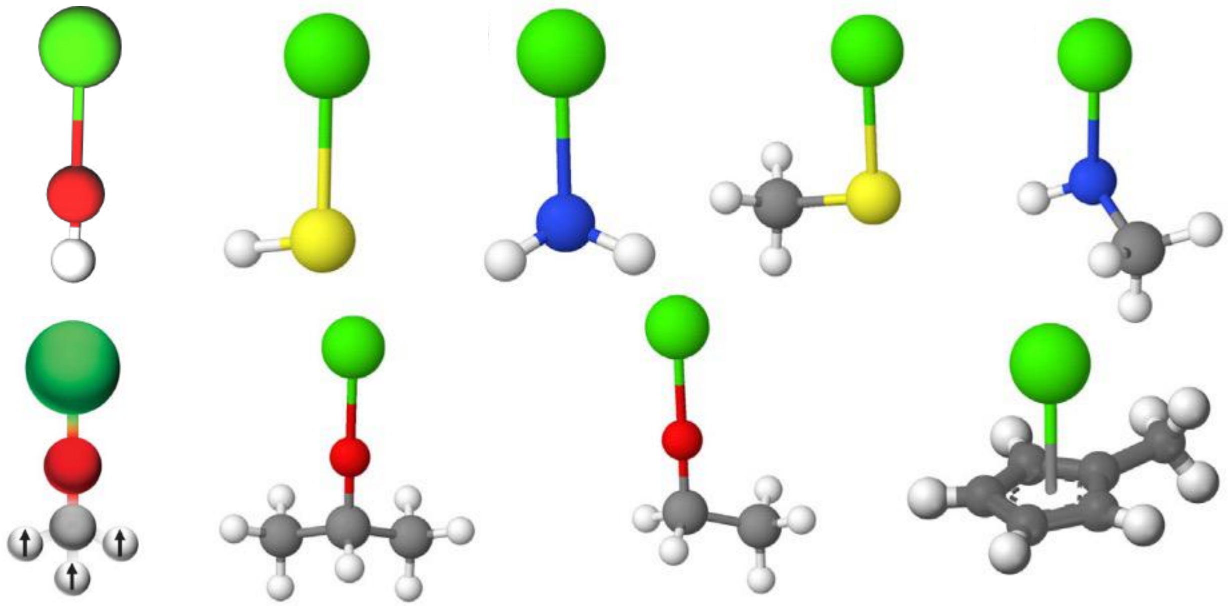


arXiv:2301.08656

Long-term prospects

Scheme discussed in this talk applies to a versatile choice of *metal* (Ca, Sr, Yb, Ra) and *ligand* (F, OH, SH, OCH_3)



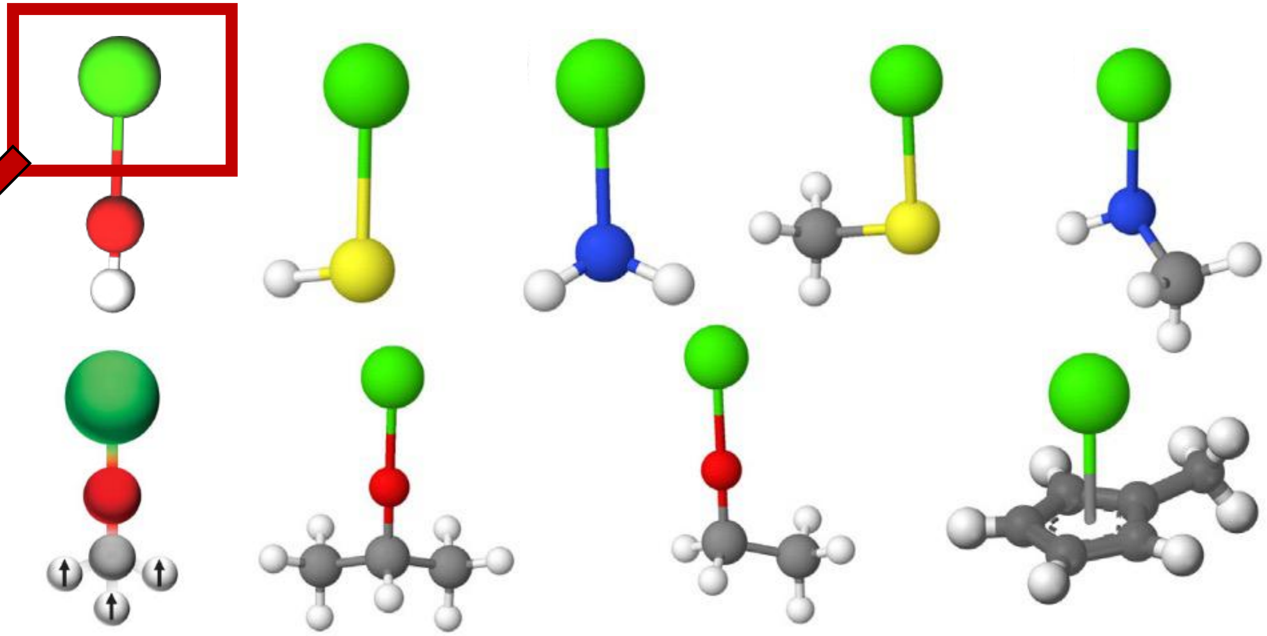
Molecule	E_{eff} [GV/cm]	# in trap for ACME II level
CaOH	0.30	100,000
SrOH	2.2	2,000
BaOH	6.8	200
YbOH	23.6	20
RaOH	56.9	3

Parameter	Value
Loss (s^{-1})	1 / 0.750
Decoherence (s^{-1})	1 / 10
Dead time (s)	0.010
Contrast	1
Integration time (hours)	168 (24*7)

Collaborators at Caltech developing studies of RaOH, presently challenging due to low available numbers and radioactivity

We have a concrete pathway for trapped, ultracold YbOH requiring much more spectroscopy

Long-term prospects



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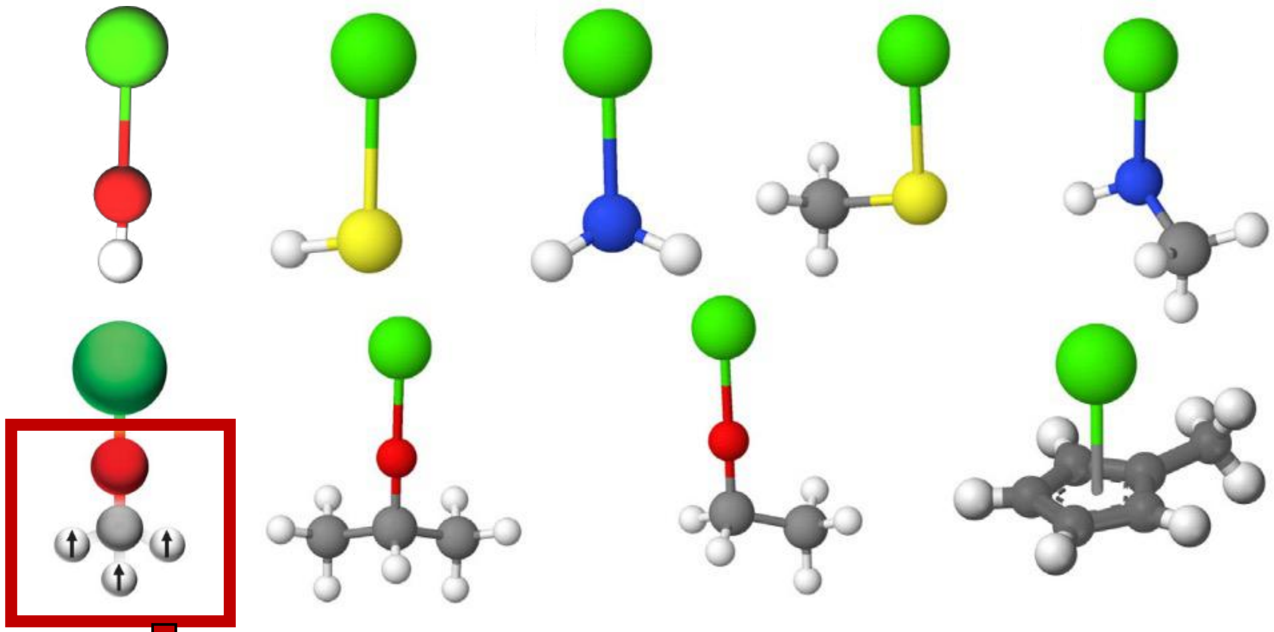

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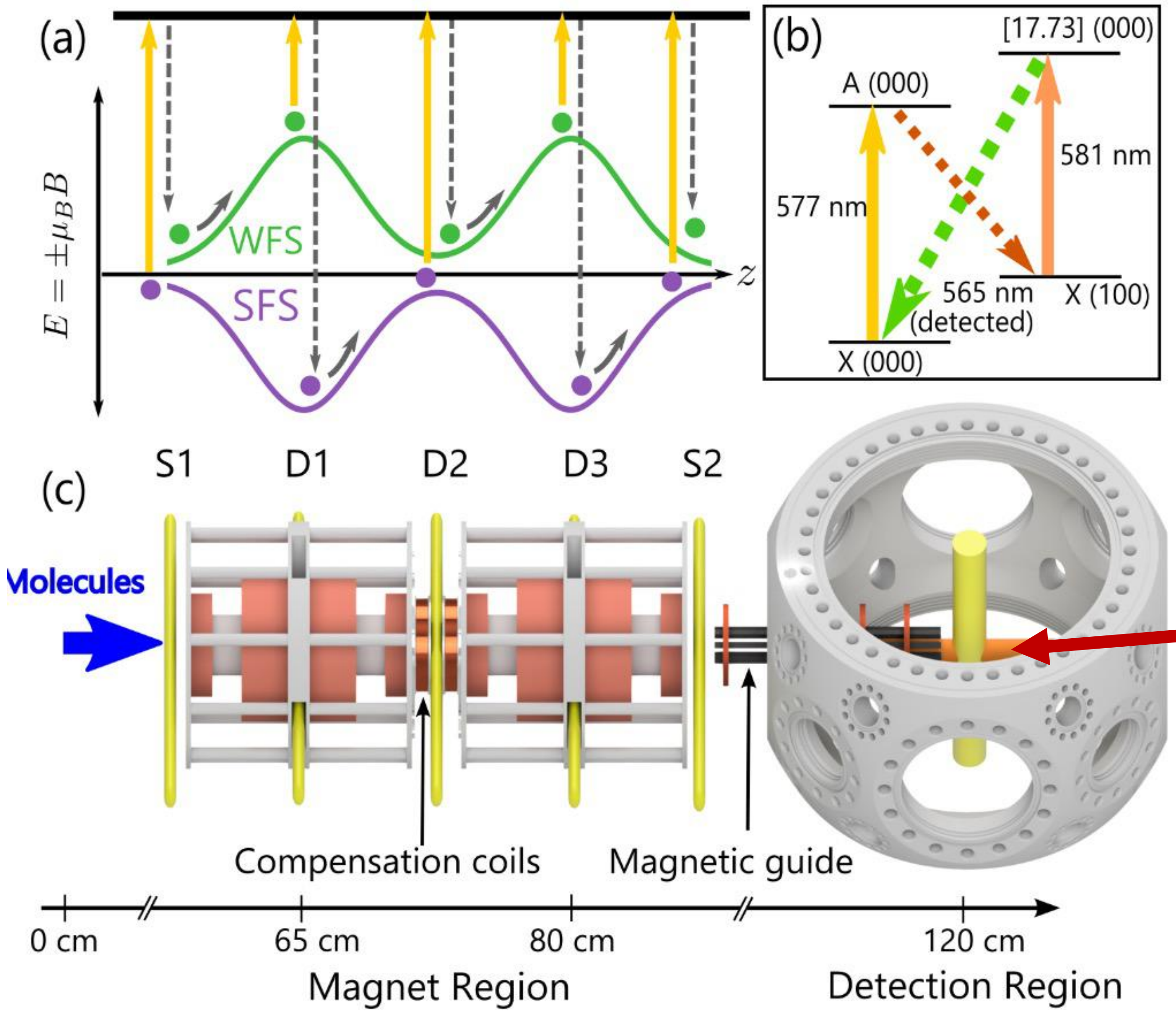
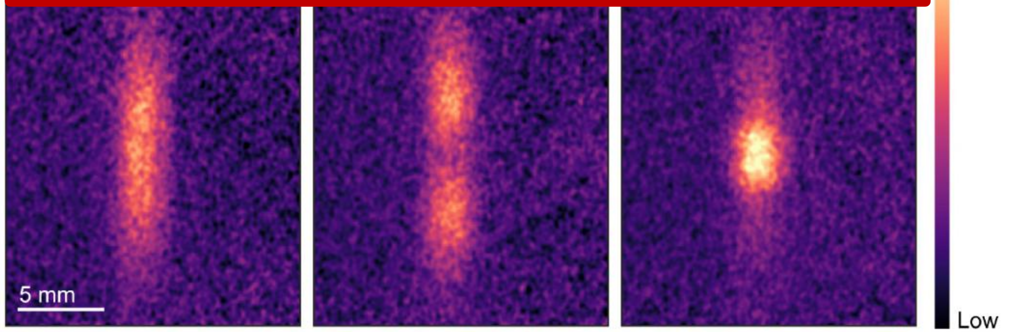
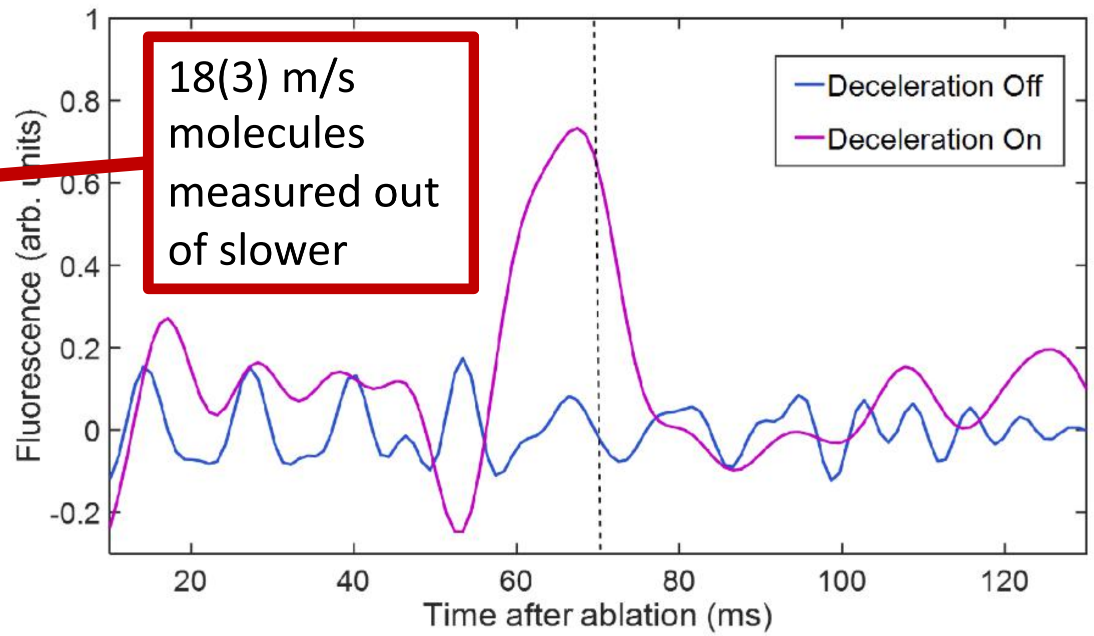
We have a concrete pathway for trapped, ultracold YbOH requiring much more spectroscopy

State of the art in YbOH: slowing + cooling



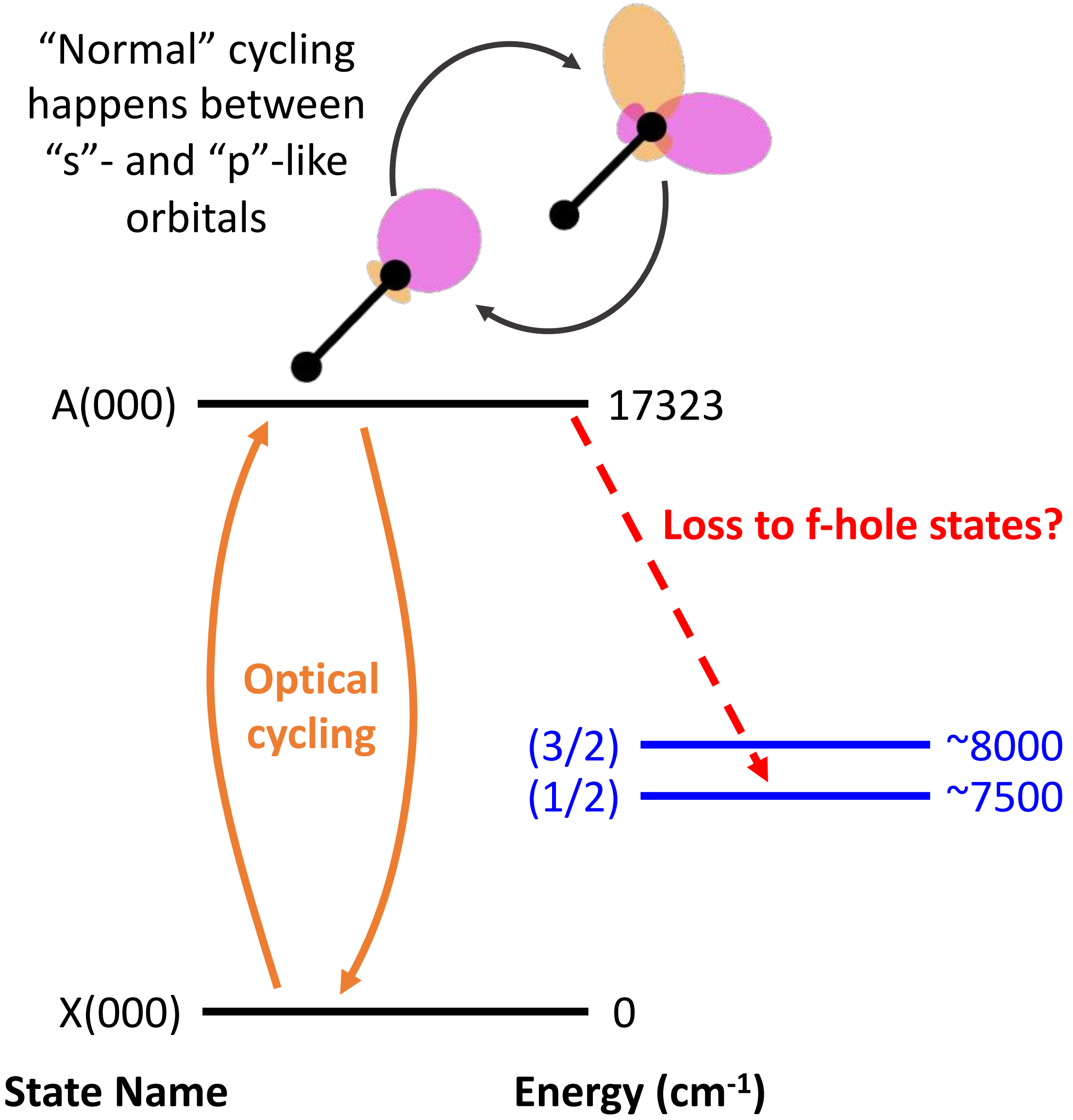
Previous laser cooling/heating demonstrated





Extra challenge in heavy species: YbOH

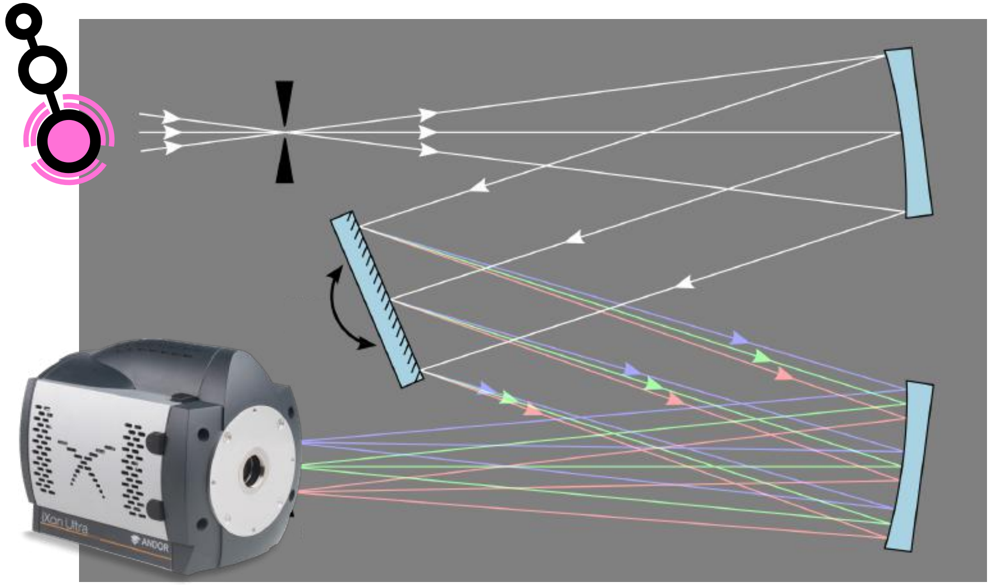
- Yb configuration ~ [Xe] $4f^{14}\,6s^2$
- After bonding to OH, ground state is like $4f^{14}6s^1$ and excited states involve excitations of the $6s^1$ electron
- **But**, excitations involving the $4f^{13}6s^2$ configuration also exist and can be between the X and A states
  - Colloquially: "f-hole states"

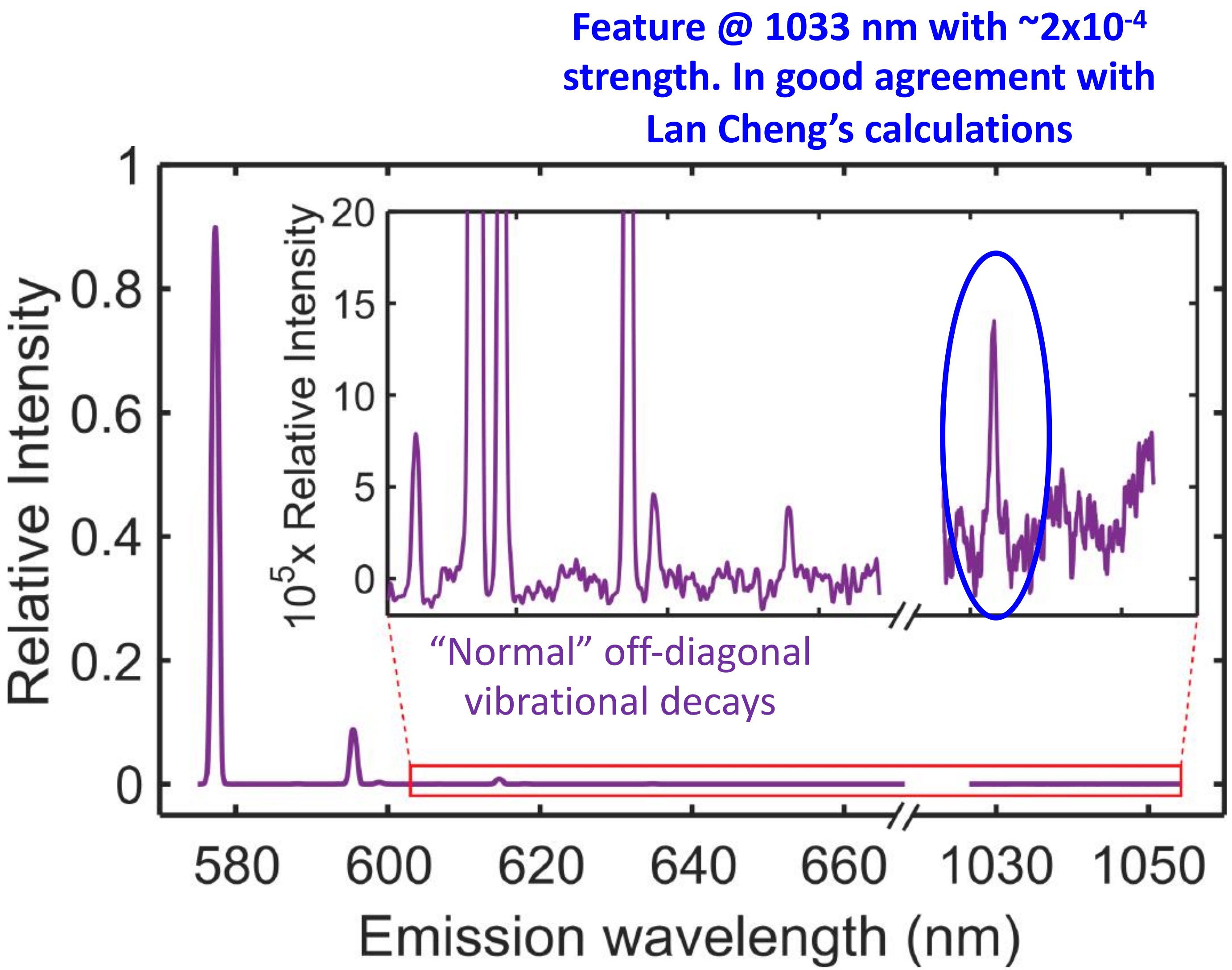


See Cheng, Steimle, Tarbutt group paper on these states in YbF: 10.1016/j.jms.2022.111625

Direct Observation of Decay to “4f^{13}” Level

Look for decay to metastable electronic states in YbOH in the “usual” way, analogous to vibrationally excited states



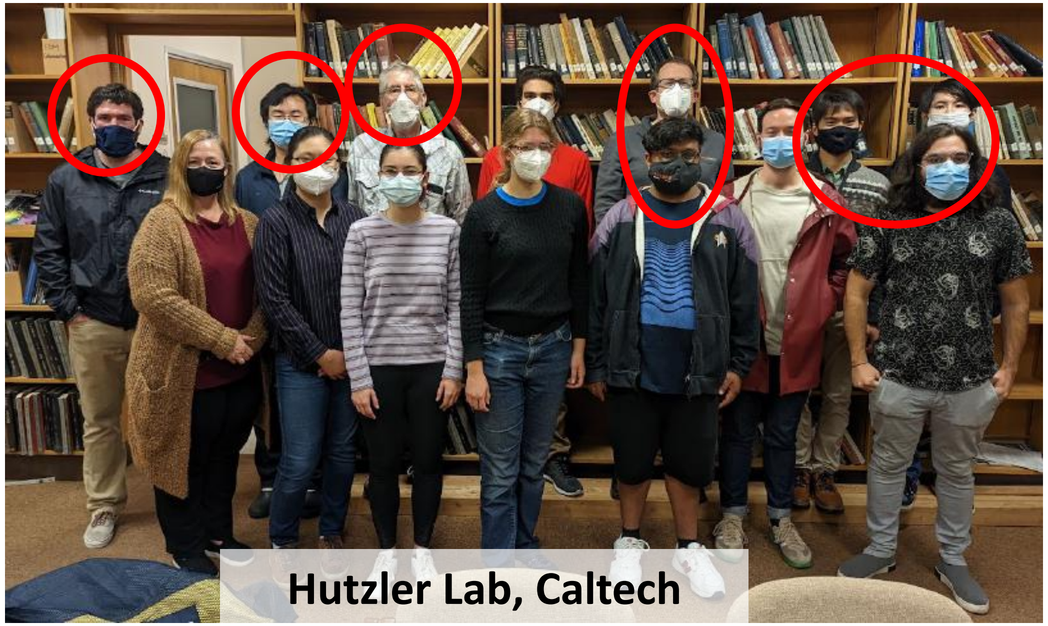


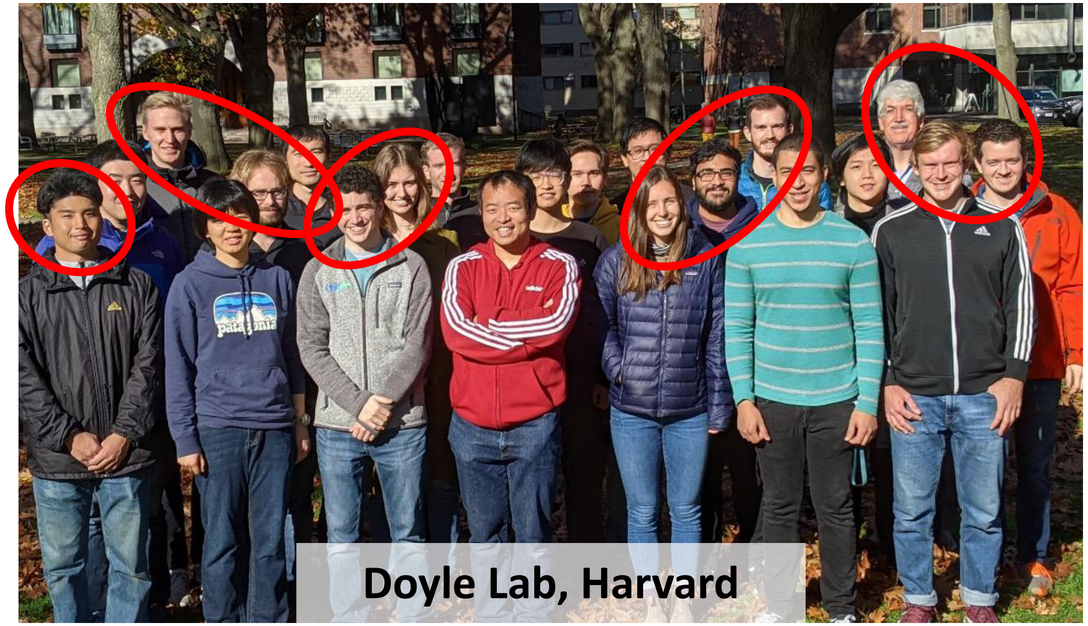
Implications for photon cycling in YbOH

- Combine high-resolution vibrational branching and metastable electronic state branching (which has its own vibrational states) to project a "photon budget" for YbOH
- Estimate ~4000 photons to load MOT from slowed beam of YbOH
  - Based on MOT loading measurements of CaOH (and scaling by mass, etc.)

State	VBR (%)	Cumulative	Photons @ 1/e loss	Notes
000	89.3654	0.893654	9.40	Found at high resolution
100	9.1024	0.984677	65.26	Found at high resolution
200	0.9132	0.993810	161.55	Found at high resolution
02^00	0.3347	0.997157	351.75	Found at high resolution
300	0.0669	0.997826	460.08	Found at high resolution
12^00	0.0550	0.998376	615.77	
010, N=1	0.0540	0.998916	922.15	Found at high resolution
f13(3/2) 000	0.0200	0.999115	1130.21	Observed in direct decay measurement
010, N=2	0.0180	0.999295	1418.55	Found at high resolution
02^20	0.0160	0.999455	1834.60	
110, N=1	0.0100	0.999555	2246.38	
f13(3/2) 100	0.0100	0.999655	2895.66	
f13(1/2) 000	0.0067	0.999721	3586.79	
22^00	0.0050	0.999771	4369.82	
400	0.0045	0.999816	5438.34	
12^20	0.0034	0.999850	6670.75	
110, N=2	0.0033	0.999883	8576.14	
f13(1/2) 100	0.0033	0.999917	12000.42	
f13(3/2) 200	0.0033	0.999950	19976.75	

Technical complication repumping "f-hole states": well-understood excited states have weak coupling, so large laser powers or analysis of new excited state needed





Tim Steimle
ASU/Caltech

Amar Vutha
Toronto

Summary:

- Polyatomic molecules offer “modular” structure to achieve laser cooling and parity doublets
- We’re on track for an SrOH trap within the year, and a competitive eEDM measurement soon after (+2 years?)
- More spectroscopy is needed for heavy molecules like YbOH with larger E_{eff}, or molecules with longer-lived science states like $MOCH_3$ (M=Sr, Yb, Ra)
- No “show-stoppers” to future work with these harder molecules!