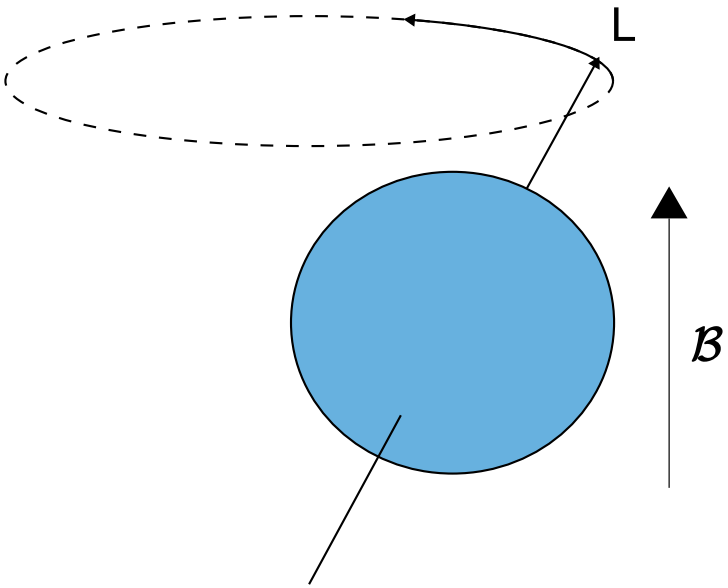
Towards searching for nuclear CP-violating physics using triatomic molecules

Luke Caldwell

2nd March 2026

University College London

Measuring an EDM



$$L = \hbar/2$$

$$\dot{\phi} = \frac{\tau}{L \sin\theta}$$

$$= \frac{\mu\mathcal{B}}{\hbar/2}$$

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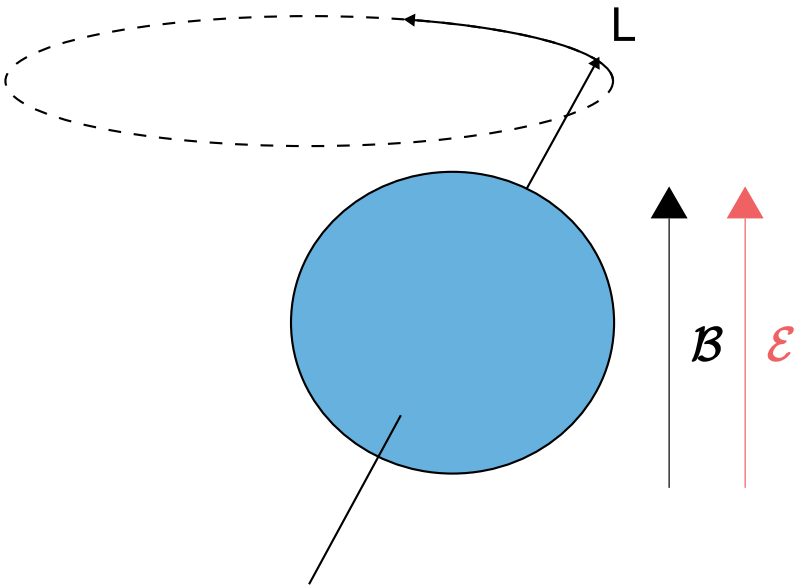

$$L = \hbar/2$$

$$\dot{\phi} = \frac{\tau}{L \sin\theta}$$

$$= \frac{\mu\mathcal{B}}{\hbar/2} + \frac{d\mathcal{E}}{\hbar/2}$$

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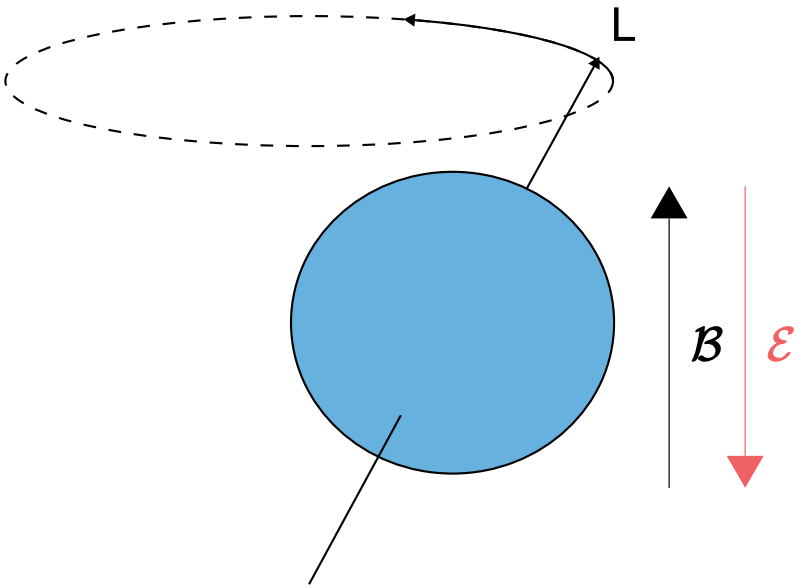

$$L = \hbar/2$$

$$\dot{\phi} = \frac{\tau}{L \sin\theta}$$

$$= \frac{\mu\mathcal{B}}{\hbar/2} - \frac{d\mathcal{E}}{\hbar/2}$$

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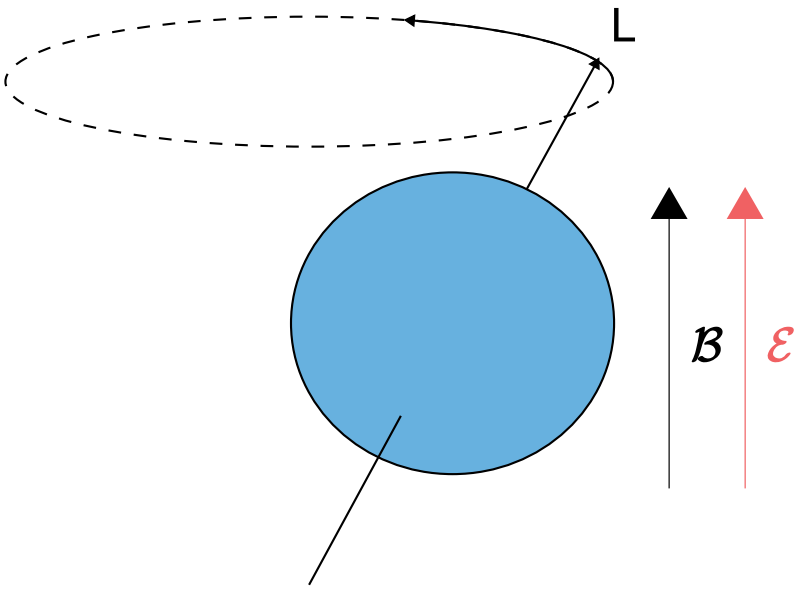

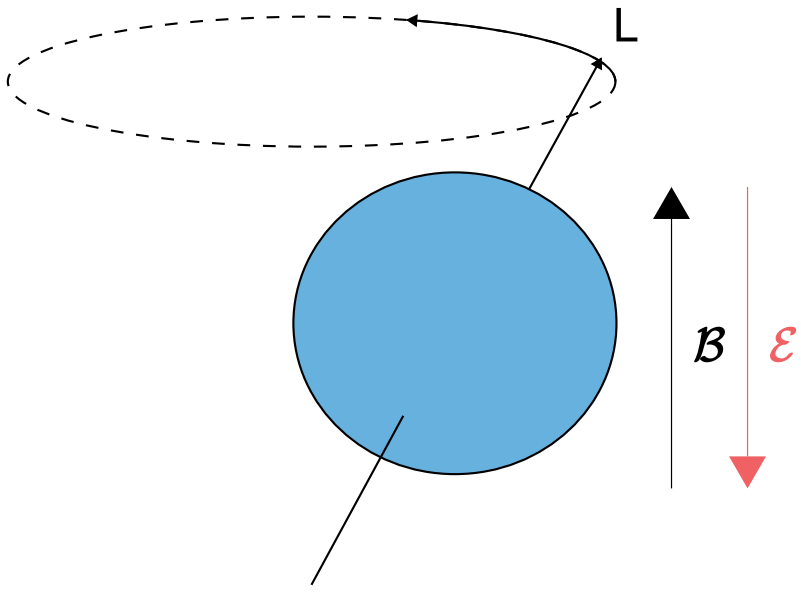

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Measuring the eEDM

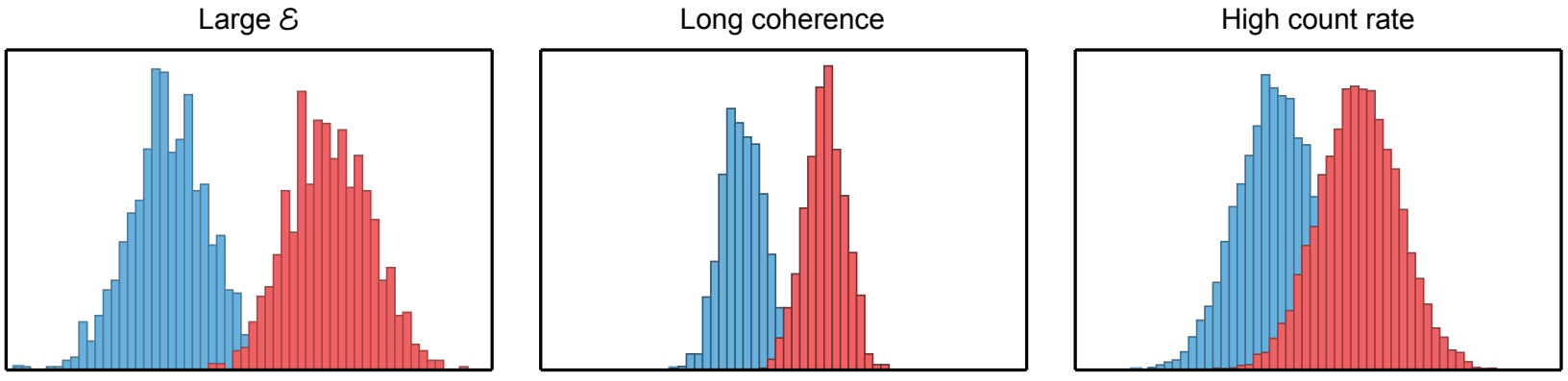
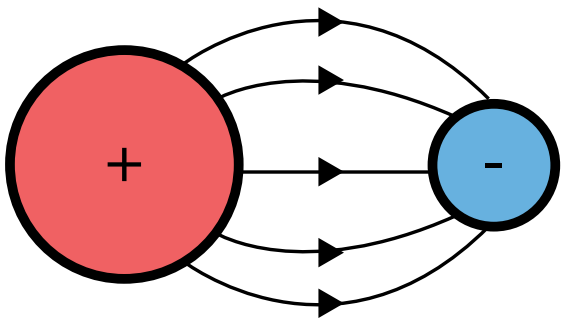


Figure of merit: $\mathcal{E}\sqrt{N\tau}$

Molecules as a laboratory for eEDM search

- Molecules containing heavy atoms can have huge effective internal fields, $\mathcal{E}_{eff} \sim 100\,\mathrm{GV\,cm^{-1}}$
- Similar effects for other CP-violating quantities (e.g. nuclear Schiff or MQM)
- All result in effective EDM of molecule



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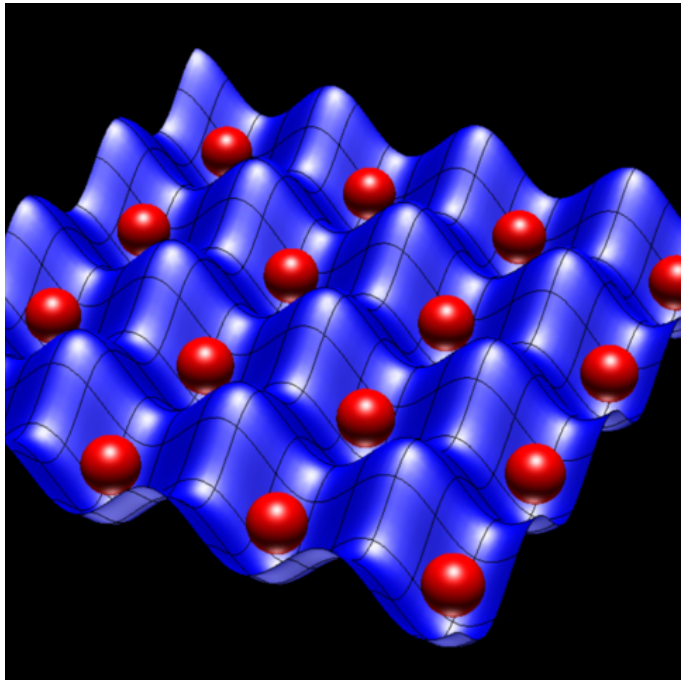

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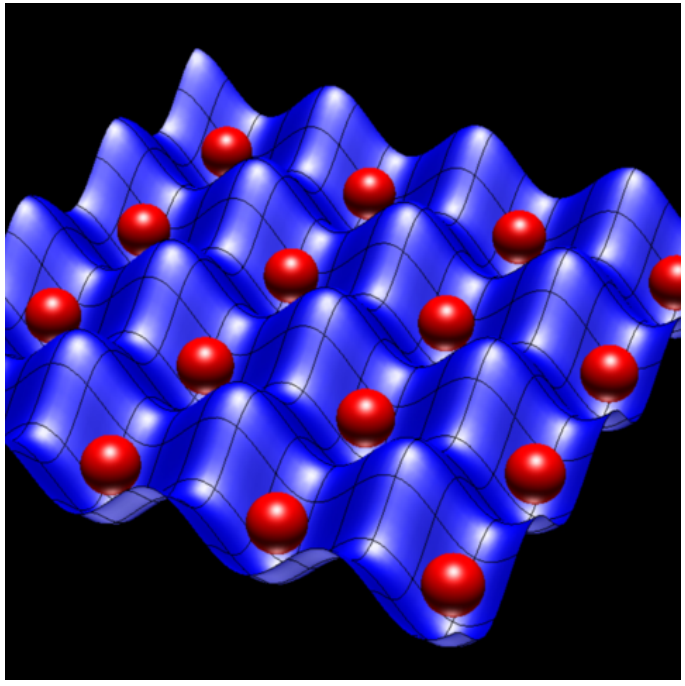

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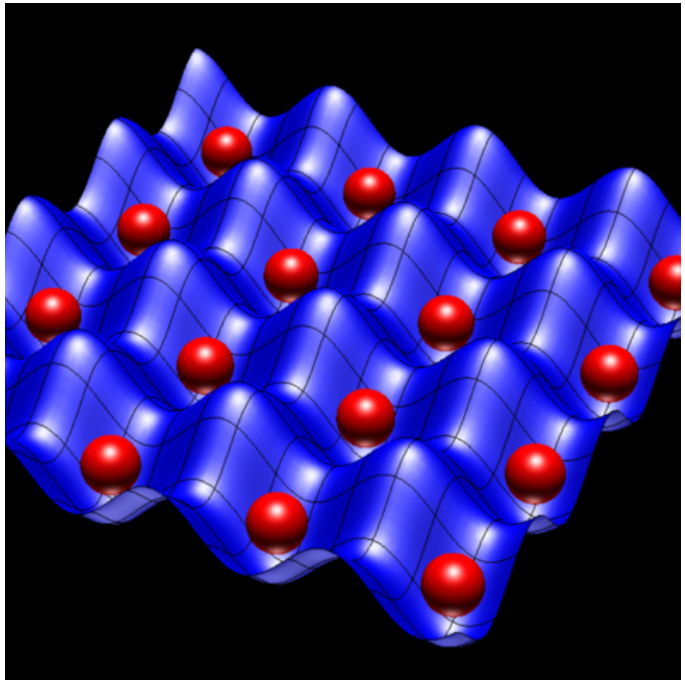
- Molecular beams - big count rates, short coherence times
- Ultracold neutral molecules - best of both worlds?
- Direct laser cooling or association



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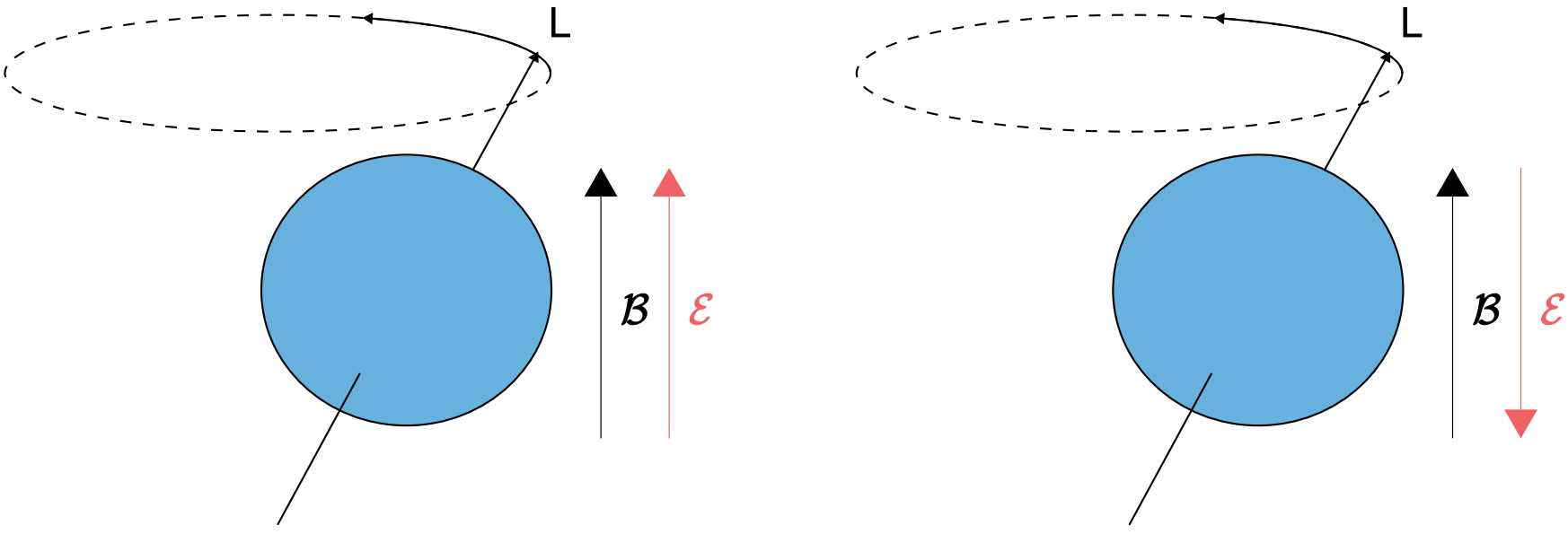
Systematics!

Measuring an EDM



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Magnetic field control is hard

- CGS units: $\frac{\mu}{d} \sim \frac{10^{-20}\,\mathrm{statC{\cdot}cm}}{10^{-39}\,\mathrm{statC{\cdot}cm}} \sim 10^{19}$

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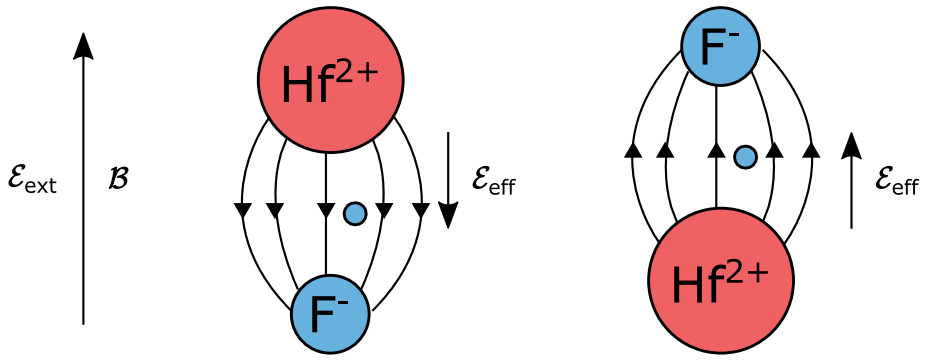
Back of the envelope:

- Searching for effects at μHz level; sensitive to correlated $\mathcal{B}$ fields $\sim 10^{-12}\,\mathrm{G}$
- Earths field fluctuates by $\sim 10^{-5}\,\mathrm{G}$ due to solar wind

Parity Doublets

- ACME, JILA rely on parity doublet states
- Two states: opposite orientation in electric field
- Flip $\mathcal{E}$ without changing external fields
- Simultaneous measurement possible
- Orbital angular momentum



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- Low-field polarisability
- Two states with opposite sensitivity to new physics but equal sensitivity to magnetic fields
- Noise suppression
- Vital feature for JILA/ACME experiments
- Can we have this feature in optically trapped molecules? Laser cooled? Associated?

Polyatomic molecules

- Idea Kozyryev, Hutzler

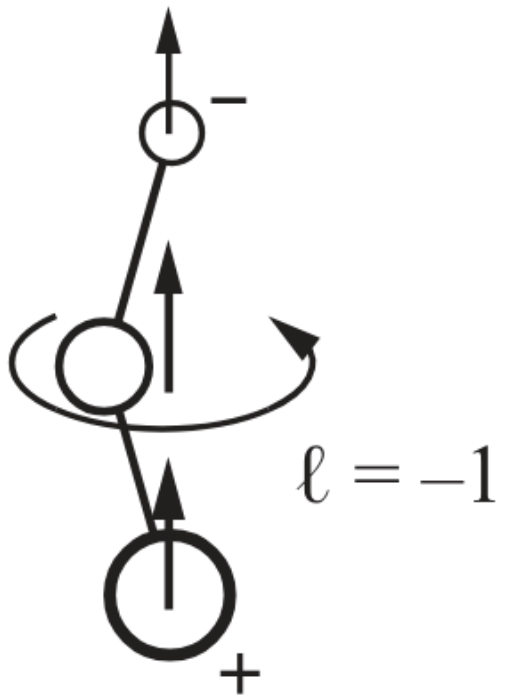

Figure from I. Kozyryev and N. R. Hutzler, Phys. Rev. Lett. **119**, 133002 (2017)

Polyatomic molecules

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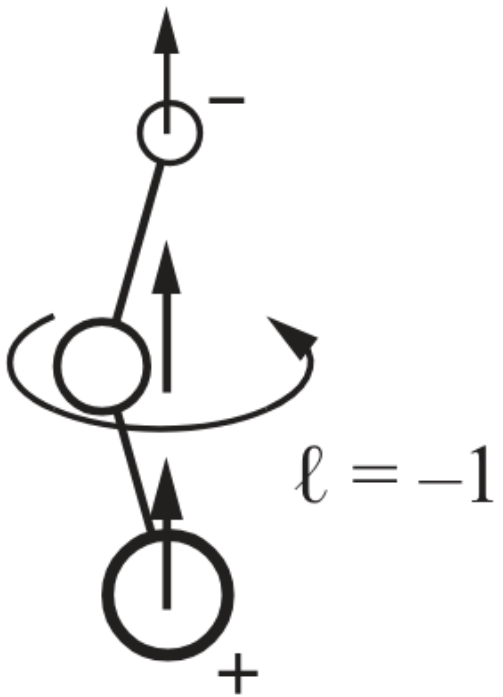
- Rotational angular momentum around ‘internuclear’ axis gives parity doublets



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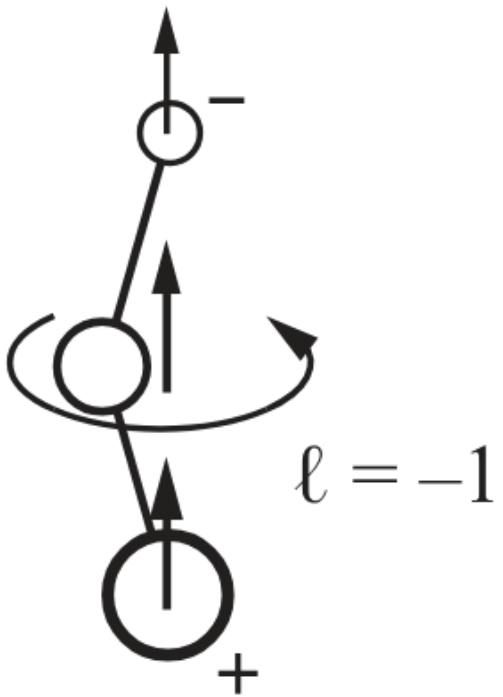

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Polyatomic molecules

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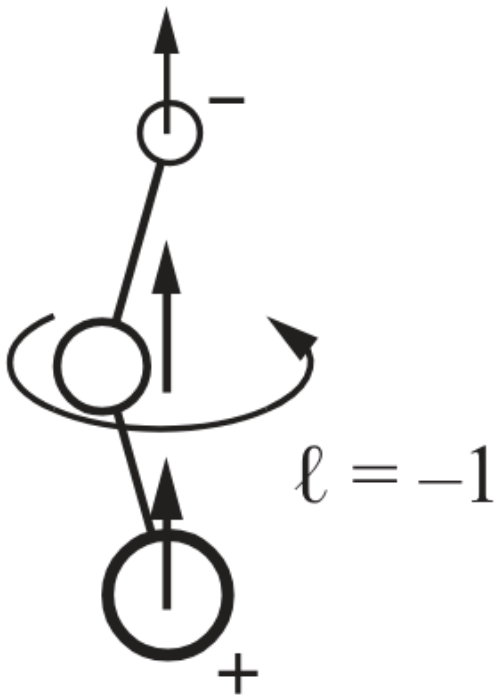
- Rotational angular momentum around ‘internuclear’ axis gives parity doublets
- Linear molecules: bending modes, 100s ms lifetime.
- Non-linear molecules: ground state, essentially infinite lifetime.



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Polyatomic molecules

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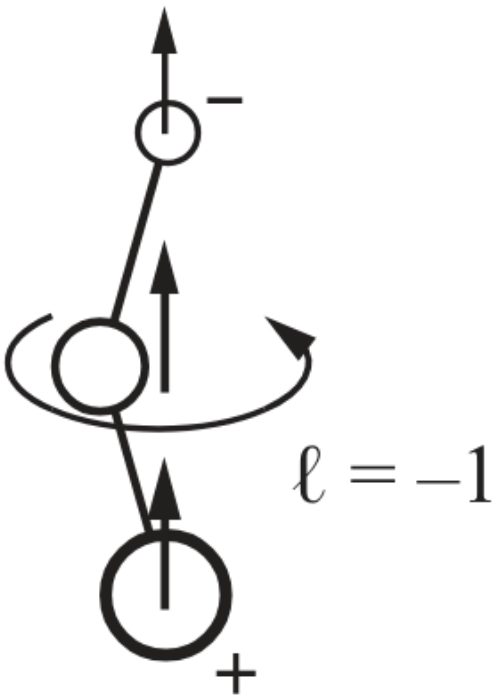
- Rotational angular momentum around 'internuclear' axis gives parity doublets
- Linear molecules: bending modes, 100s ms lifetime. Laser cooling - CaOH, SrOH... YbOH, BaOH, RaOH
- Non-linear molecules: ground state, essentially infinite lifetime.



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Polyatomic molecules

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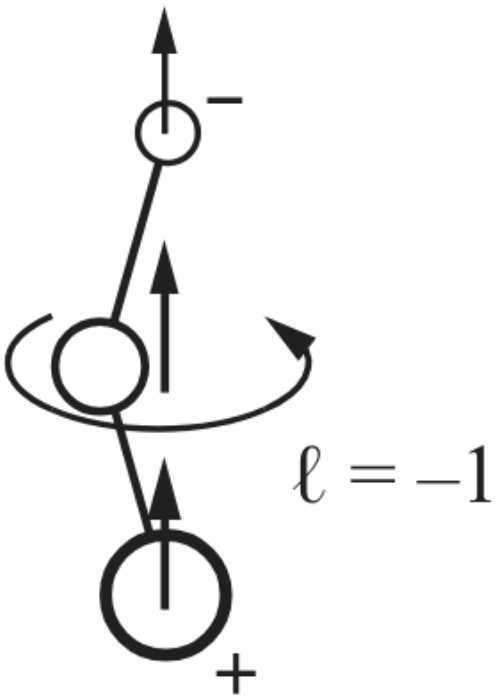
- Rotational angular momentum around 'internuclear' axis gives parity doublets
- Linear molecules: bending modes, 100s ms lifetime. Laser cooling - CaOH, SrOH... YbOH, BaOH, RaOH
- Non-linear molecules: ground state, essentially infinite lifetime. More difficult



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YbCaF

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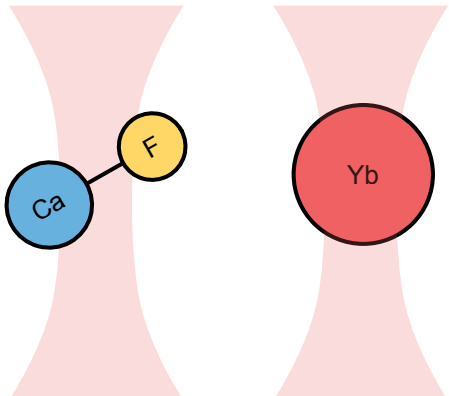
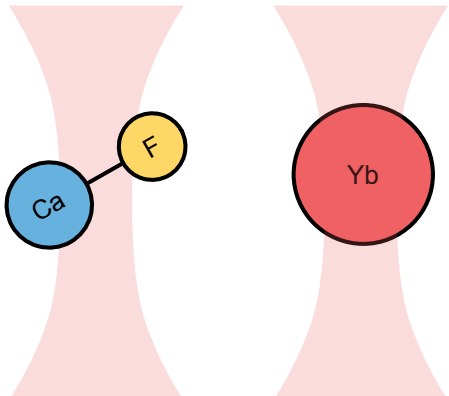
- Ultracold neutral YbCaF molecules
- Yb nucleus is measurement target



YbCaF

- Ultracold neutral YbCaF molecules
- Yb nucleus is measurement target
- Produce from separately laser-cooled species in optical tweezers



Ground state structure

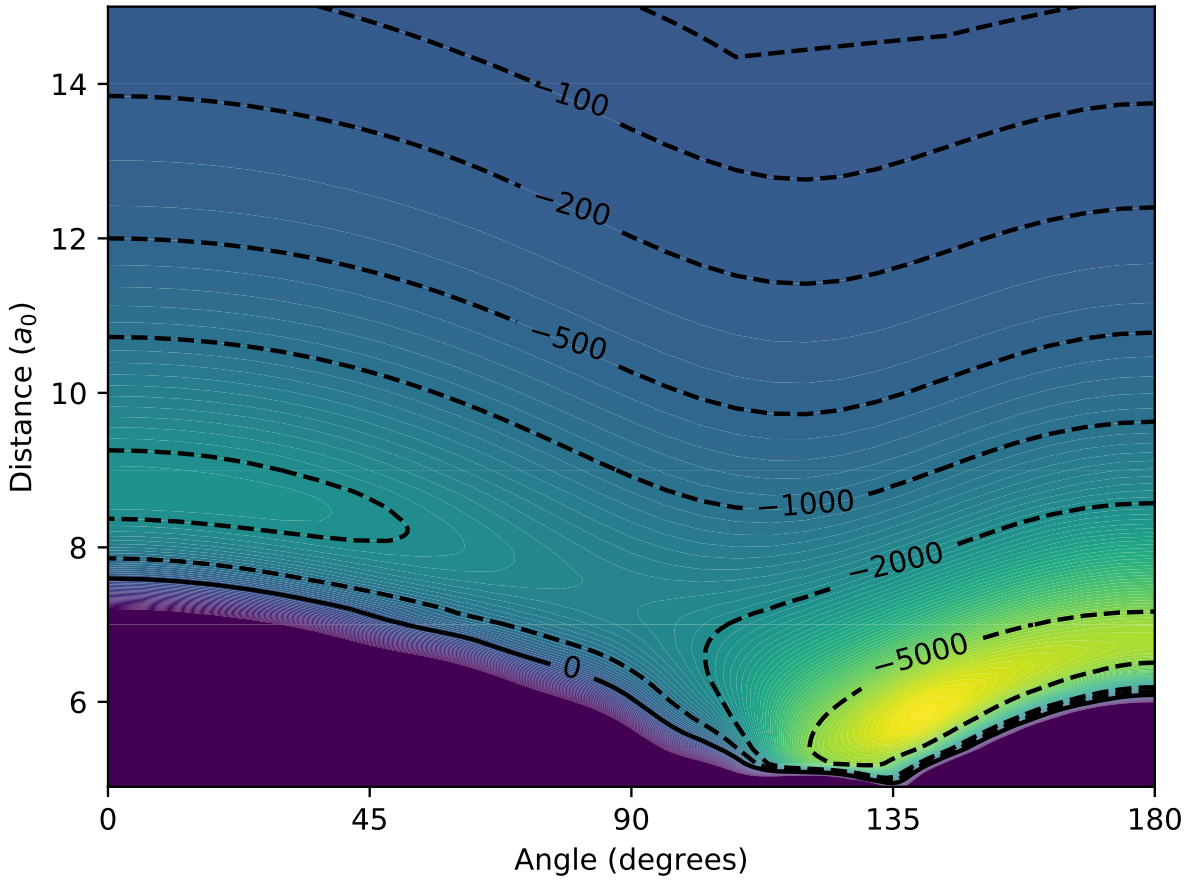


Figure from upcoming preprint Frye et. al

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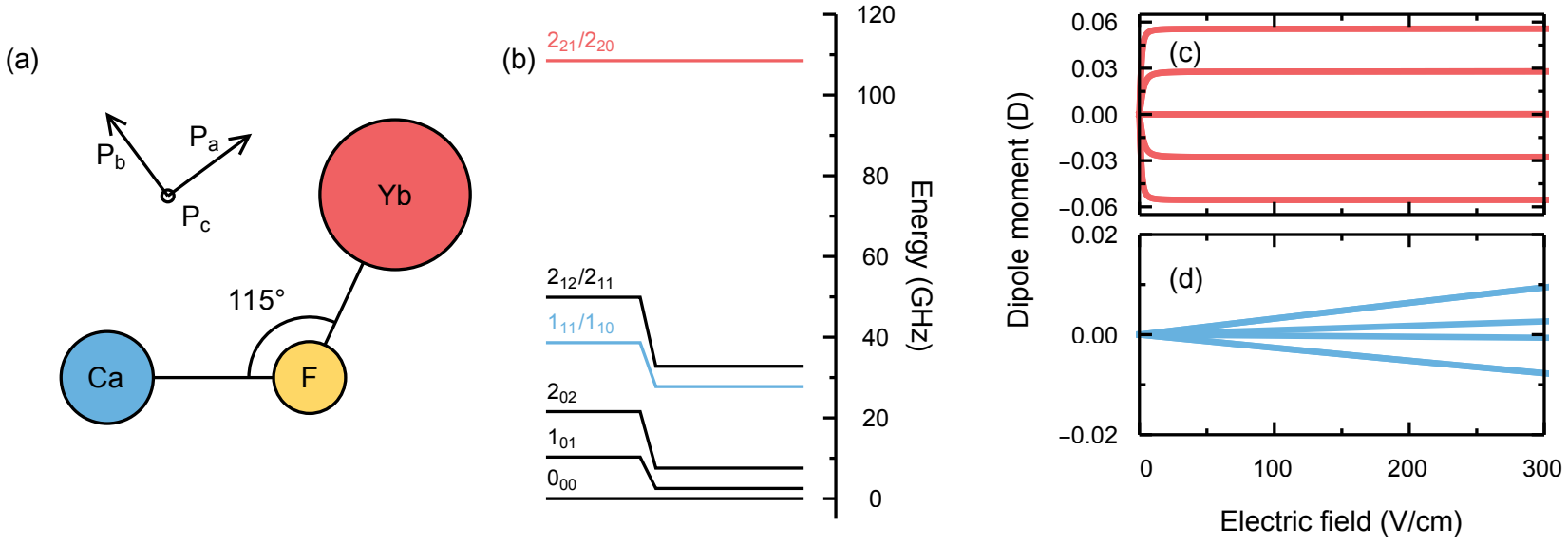
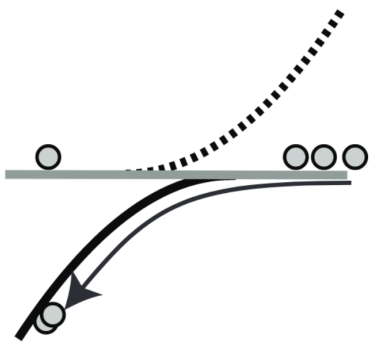

(a)
P_b
P_a
P_c
Yb
115°
Ca
F
(b)
$2_{21}/2_{20}$
$2_{12}/2_{11}$
$1_{11}/1_{10}$
2_{02}
1_{01}
0_{00}
120
100
80
60
40
20
0
Energy (GHz)
(c)
(d)
Dipole moment (D)
0.06
0.03
0.00
−0.03
−0.06
0.02
0.00
−0.02
0
100
200
300
Electric field (V/cm)

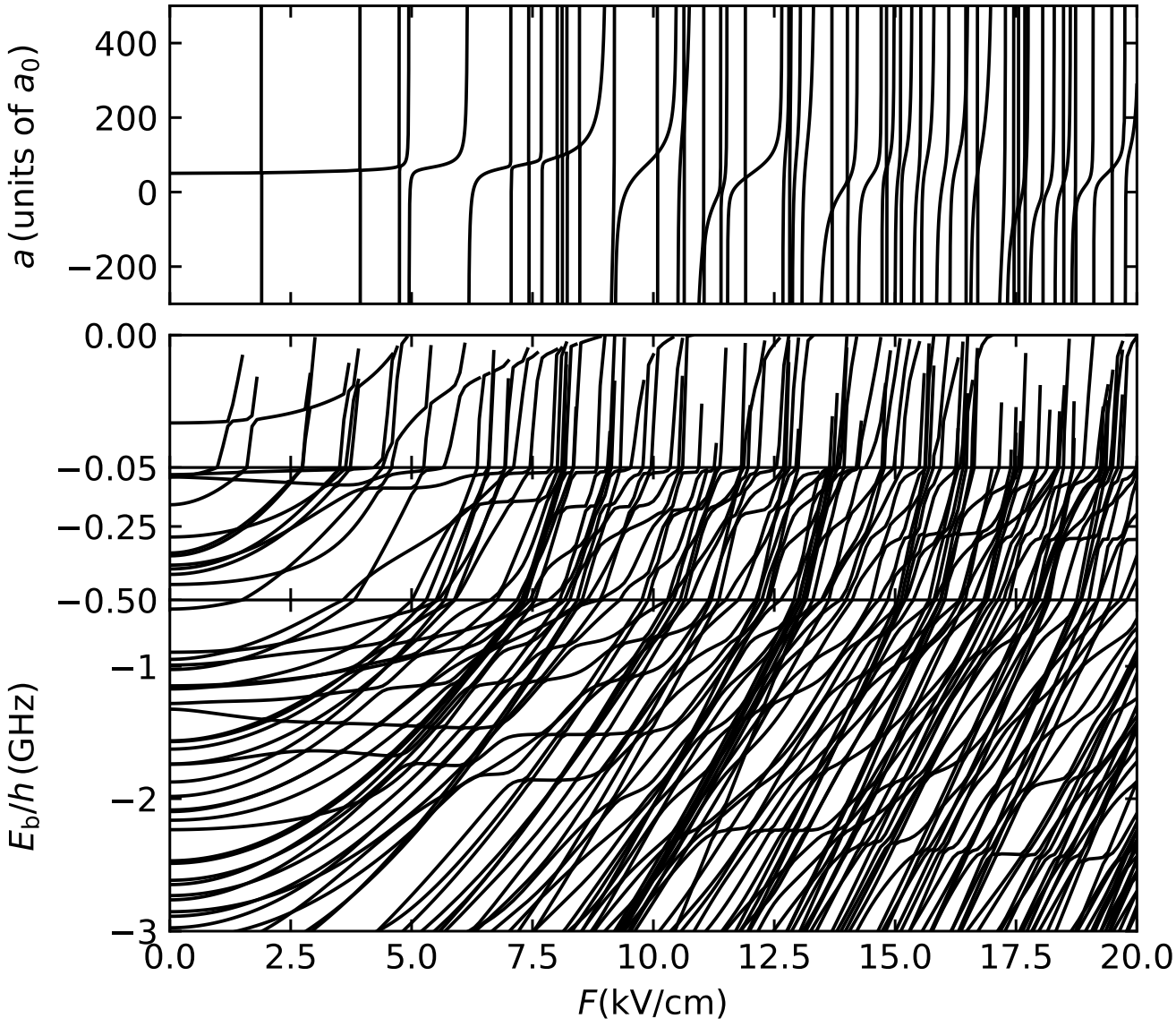
Association

- Ramp (magnetic) field across a Feshbach resonance; avoided crossing between unbound pair and vibrationally excited bound state
- Transfer to ground state with STIRAP, all energy carried away in light field



Association figure from F. Ferlaino et al., in Cold molecules (CRC Press, 2009), pp. 351–386.

Electric field association?



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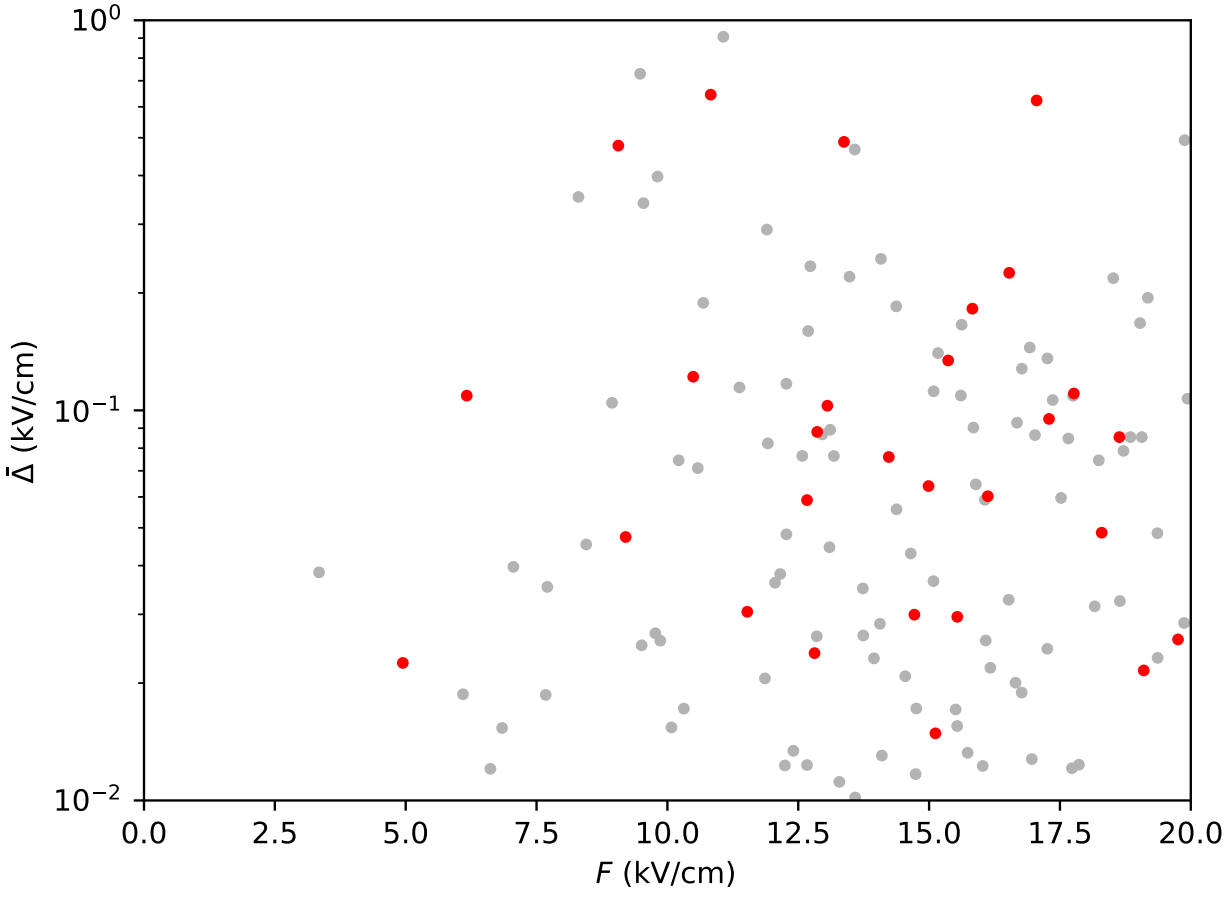

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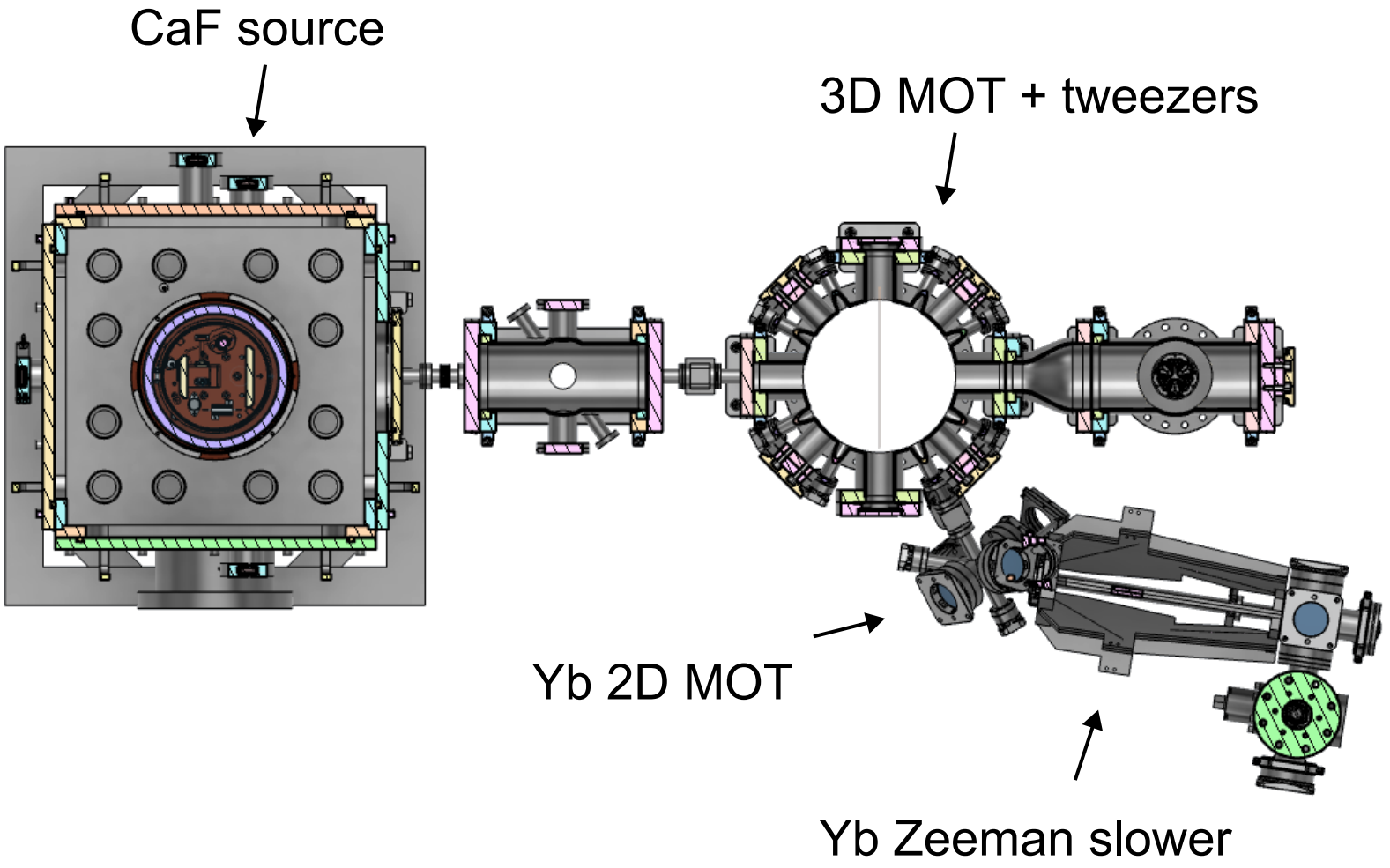
Experimental plans



The YbCaF team

- **Photo** Cathy Darling, Henry Sewell, Ben Fox, Sam Haswell, LC
- **Masters students** Roberto Haubold, Amberley Yang

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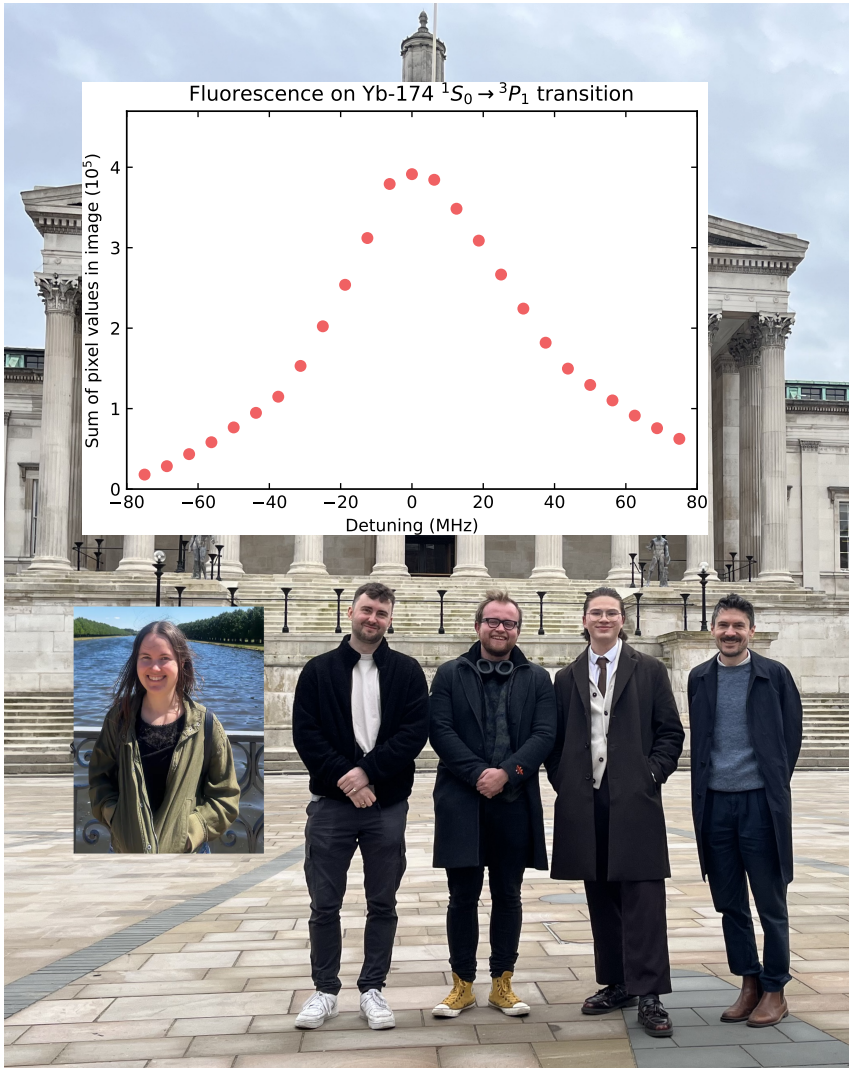

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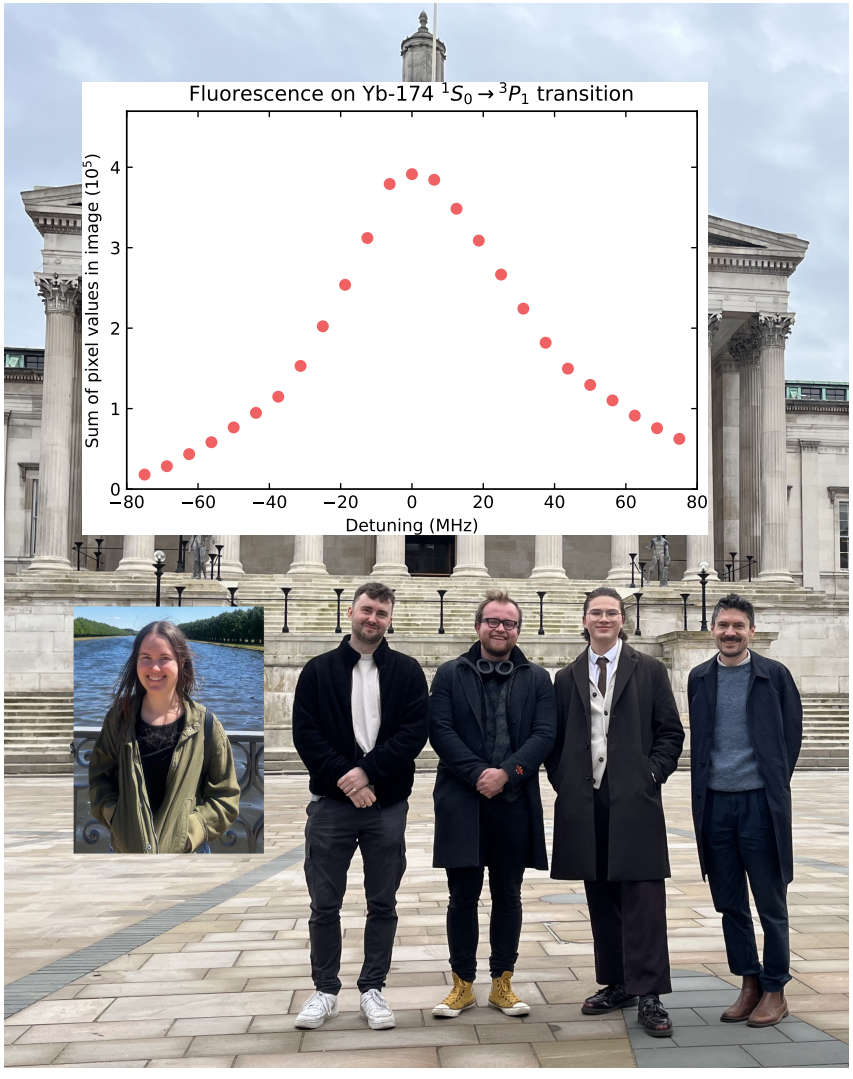

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- **Masters students** Roberto Haubold, Amberley Yang
- **Theory** Michał Tomza, Matthew Frye, Marcin Gronowski, Adam Koza, Konstantin Gaul, Ignacio Aucar Anastasia Borschevsky

Questions?

d_e vs C_S

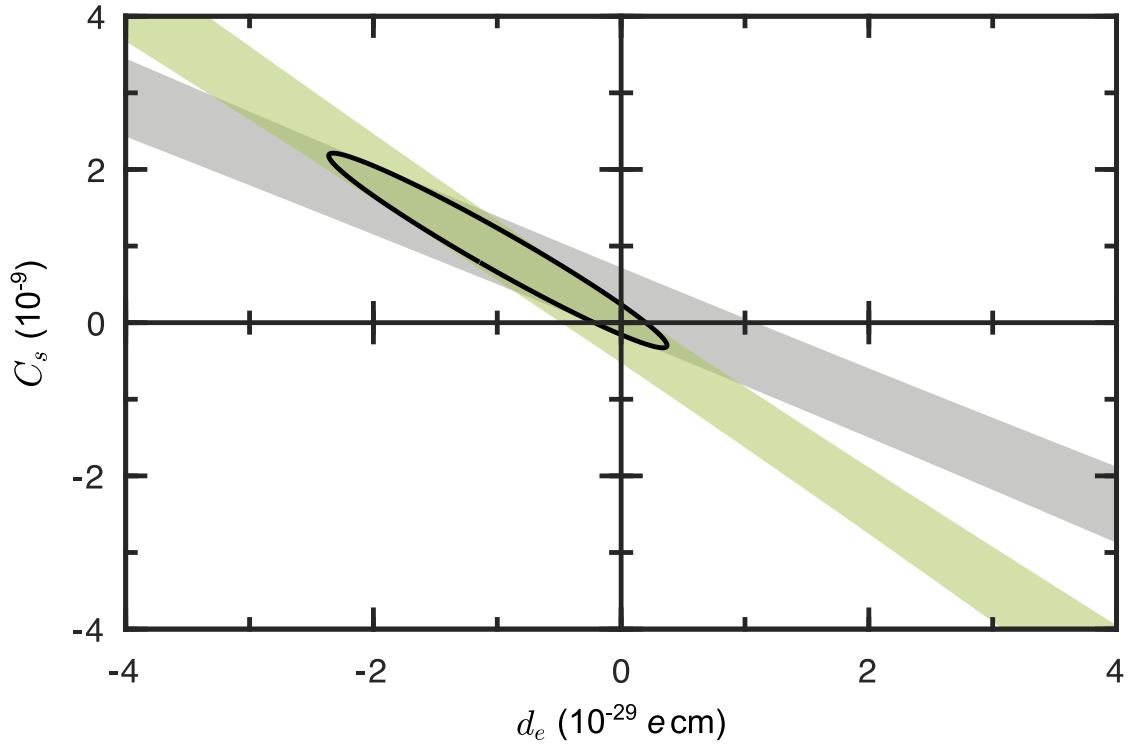
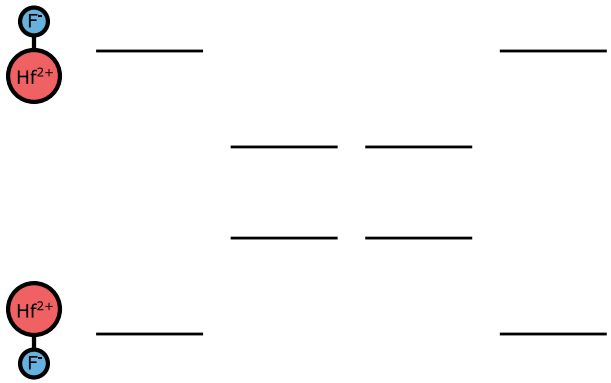


Figure from SM of T. S. Roussy et al., Science **381**, 46–50 (2023)

Spectroscopy in the rotating frame



See A. E. Leanhardt et al., Journal of Molecular Spectroscopy **270**, 1–25 (2011)

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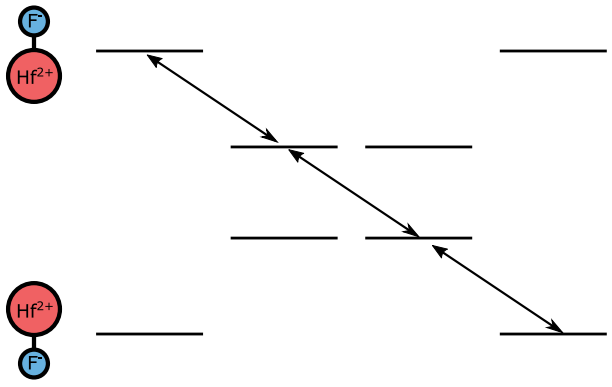

- Rotation mixes neighbouring m_F levels

See A. E. Leanhardt et al., Journal of Molecular Spectroscopy **270**, 1–25 (2011)

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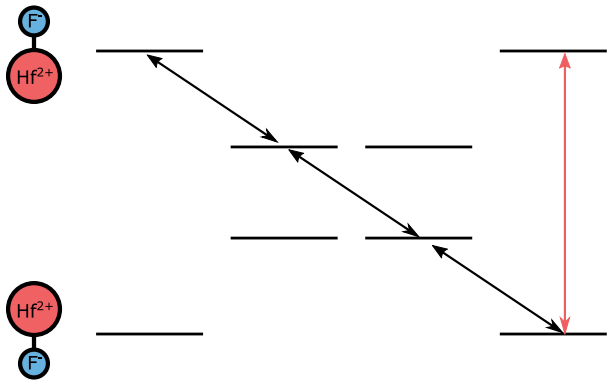

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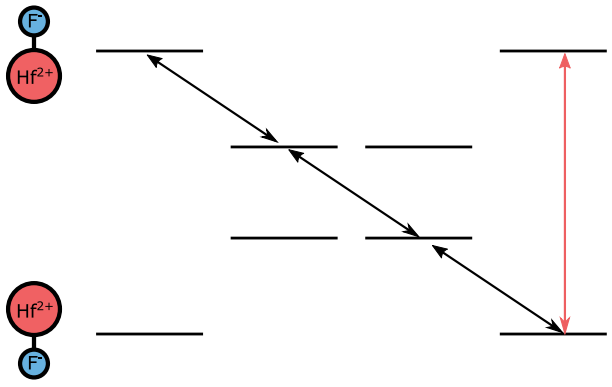

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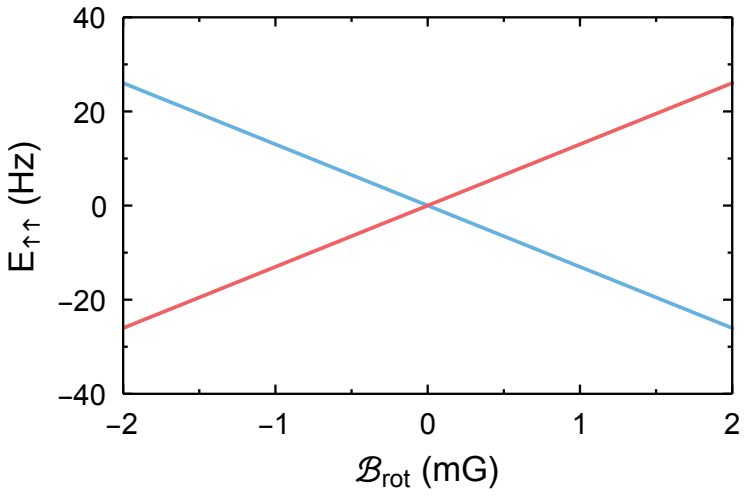

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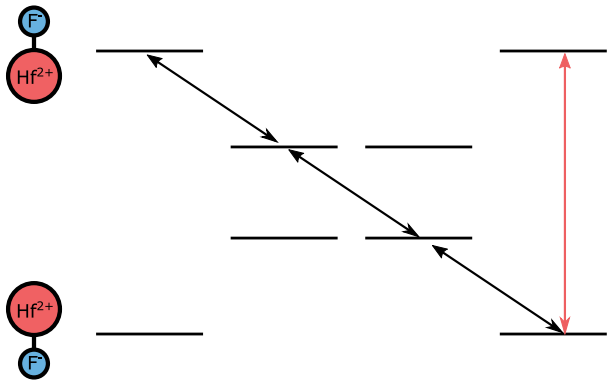

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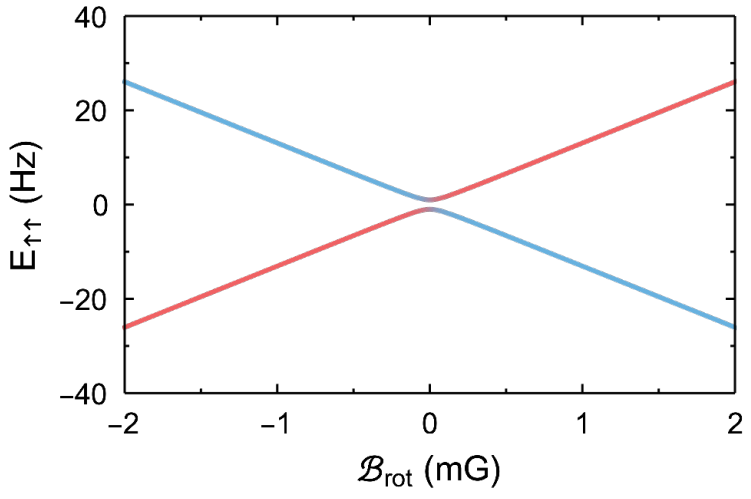

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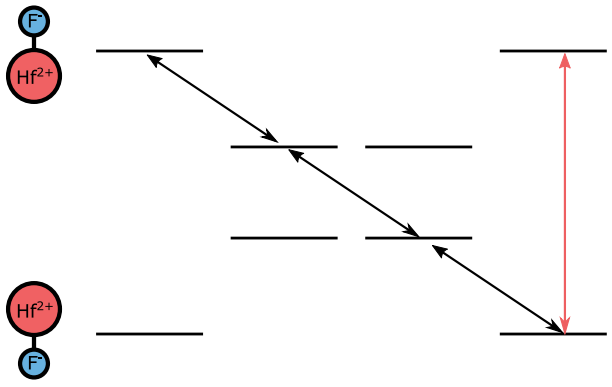

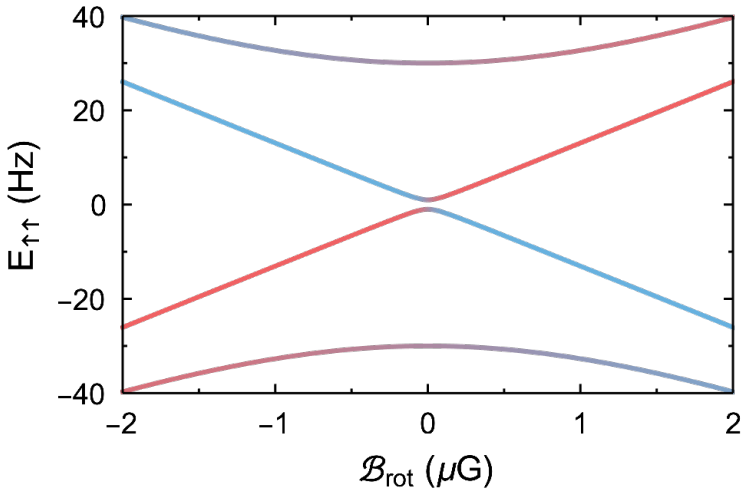

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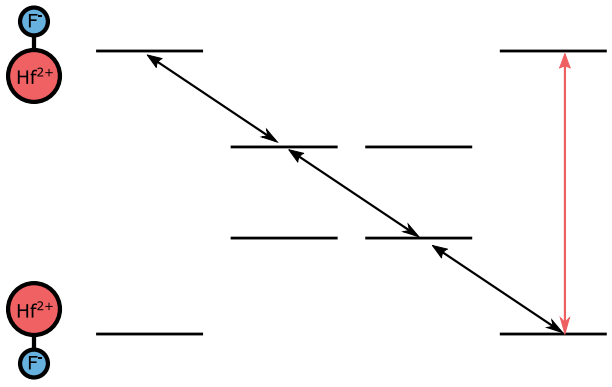

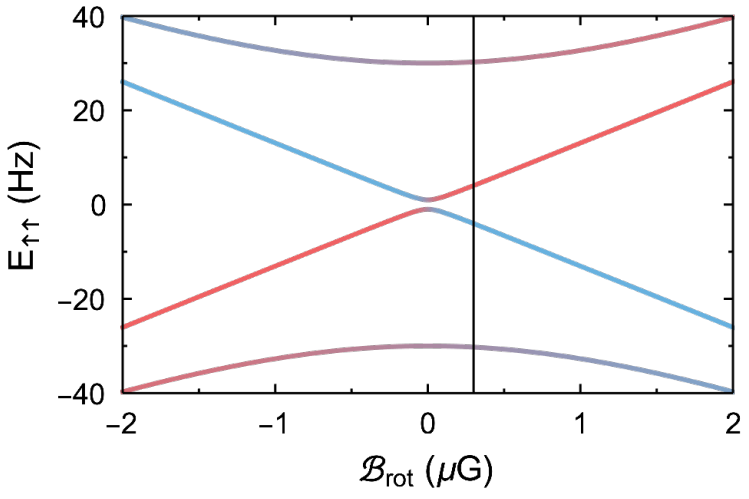

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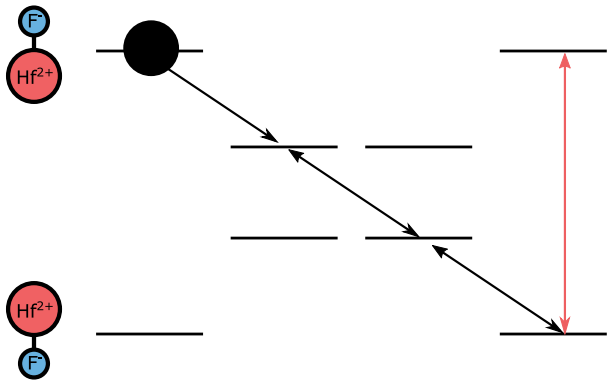
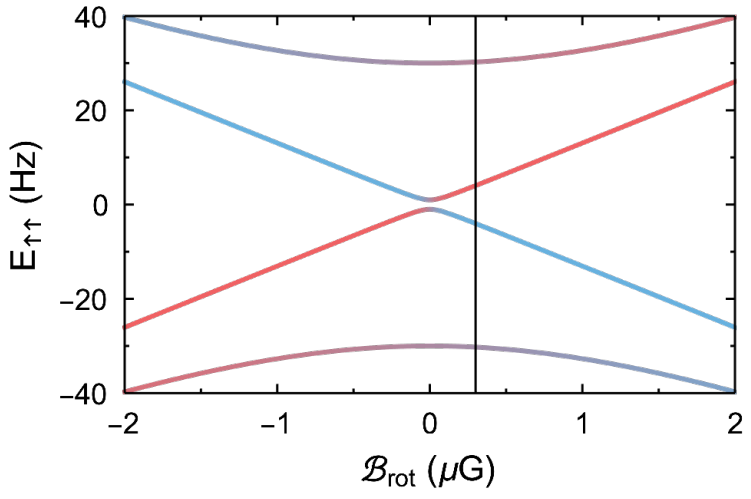
Spectroscopy in the rotating frame

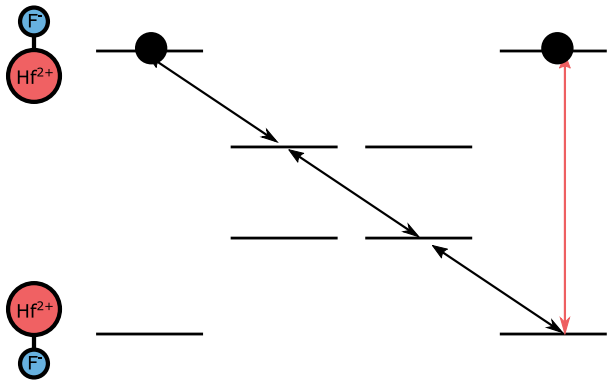


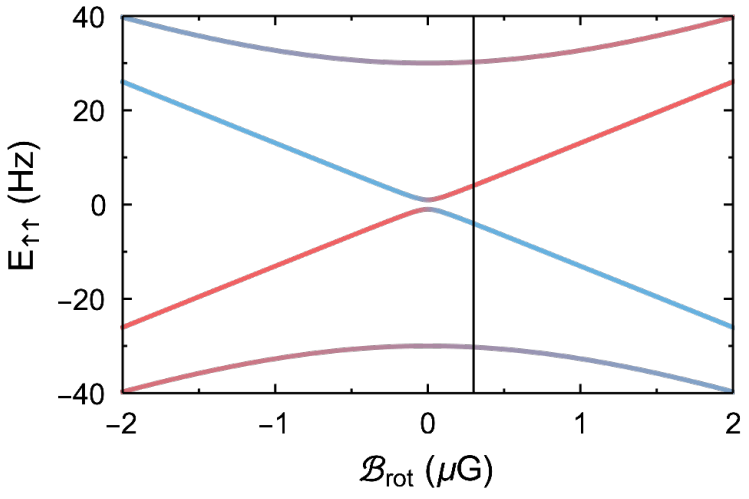
- Rotation mixes neighbouring m_F levels
- Ω-doubling mixes states of same m_F but opposite Ω



See A. E. Leanhardt et al., Journal of Molecular Spectroscopy **270**, 1–25 (2011)

Spectroscopy in the rotating frame





- Rotation mixes neighbouring m_F levels
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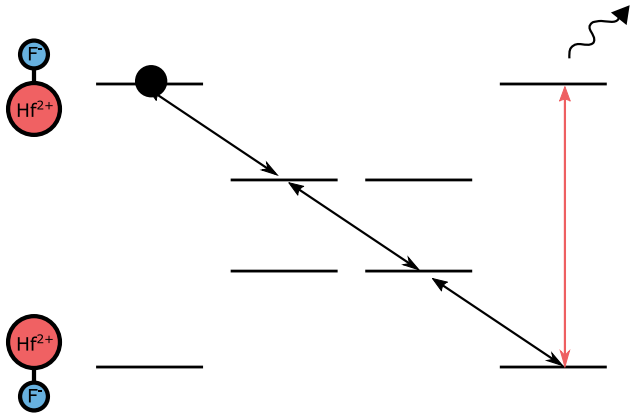
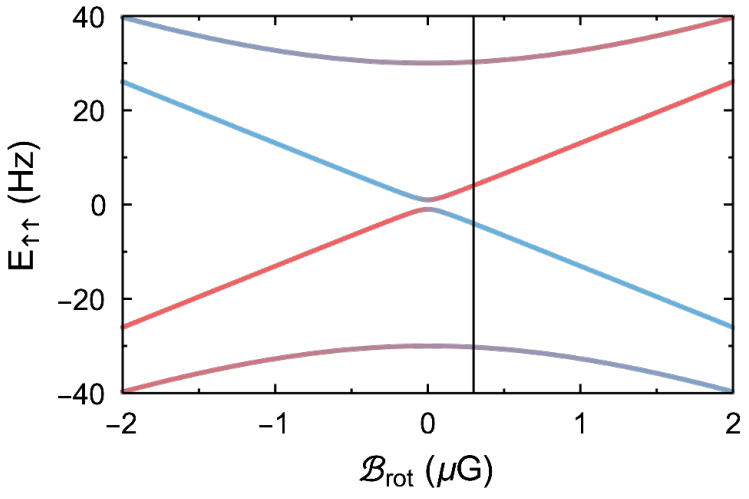
See A. E. Leanhardt et al., Journal of Molecular Spectroscopy **270**, 1–25 (2011)

Spectroscopy in the rotating frame

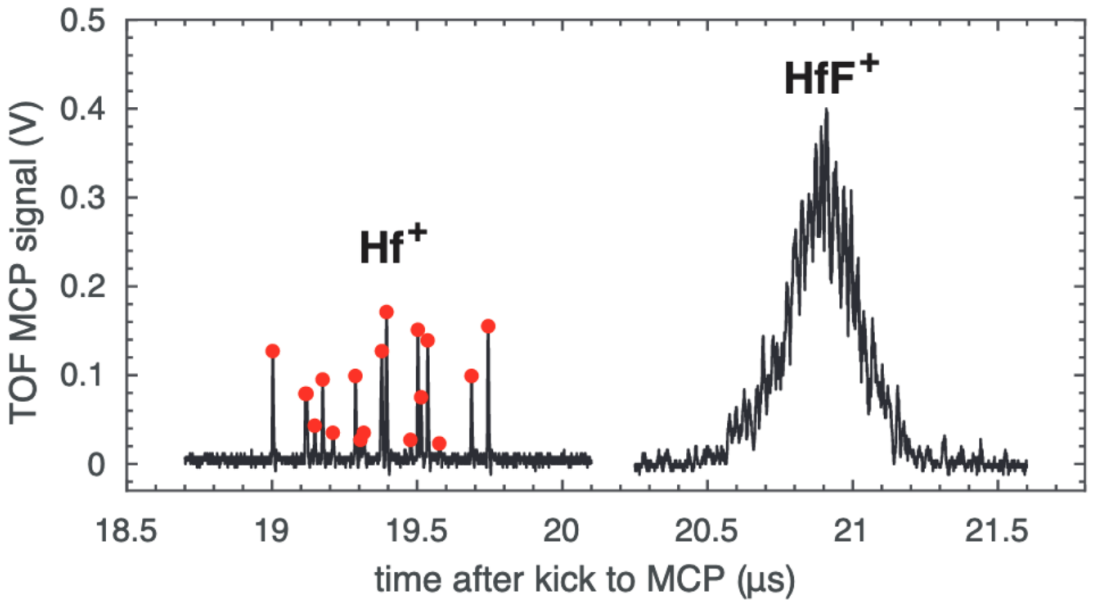




- Rotation mixes neighbouring m_F levels
- Ω-doubling mixes states of same m_F but opposite Ω

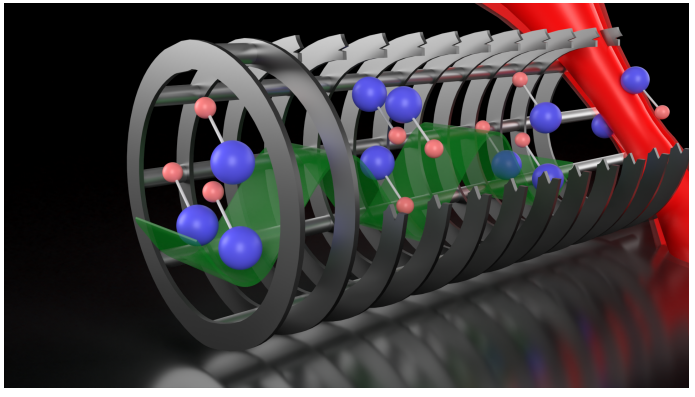
See A. E. Leanhardt et al., Journal of Molecular Spectroscopy **270**, 1–25 (2011)

Readout


0.5
0.4
0.3
0.2
0.1
0
TOF MCP signal (V)
Hf+
HfF+
18.5
19
19.5
20
20.5
21
21.5
time after kick to MCP (μs)

Future – ThF+ bucket brigade

- Switching molecule from HfF+ to ThF+, $\mathcal{E}_{\mathrm{eff}}$ increased from $23\,\mathrm{GV\,cm^{-1}}$ to $36\,\mathrm{GV\,cm^{-1}}$
- Science state is the ground state $\implies$ $10\,\mathrm{s}$ + coherence time (cryogenic environment)
- To take full advantage, we will run many experiments in parallel, in array of moving ion traps



Linear combinations

TABLE VII. Uncertainty budget.

Effect	Section	Correction (μHz)	Uncertainty (μHz)
Magnetic			
Nonreversing $\vec{\mathcal{B}}^0$	VI A 1	0.1	< 0.1
Second harmonic of $\mathcal{E}_{\text{rot}}$ and transverse magnetic field	VI A 3		2.2
Third harmonic of $\mathcal{E}_{\text{rot}}$ and magnetic field gradients	VI A 4		1.5
Higher harmonics of $\mathcal{E}_{\text{rot}}$ and higher-order magnetic field gradients	VI A 4		< 0.1
Ellipticity of $\mathcal{E}_{\text{rot}}$ and magnetic field gradients	VI A 5		1.7
Berry's phase			
Phase modulation due to axial secular motion	VI B 3		3.4
Axial second harmonic with ellipticity of $\mathcal{E}_{\text{rot}}$	VI B 4		1.7
Higher harmonics of $\mathcal{E}_{\text{rot}}$	VI B 5		3.0
Rotation-induced mixing			
Leaking of f^{BR}	VI C 1		0.2
Axial magnetic fields	VI C 2		0.4
Other frequency			
Imperfect overlap of spatial distributions	VI D 2		3.5
Phase			
Improperly characterized imaging contrast	VII B 1		0.7
Swatch position	VII B 2		1.2
Total systematic		0.1	6.9
Statistical			22.8
Total		0.1	23.8

See L. Caldwell et al., Phys. Rev. A **108**, 012804 (2023)