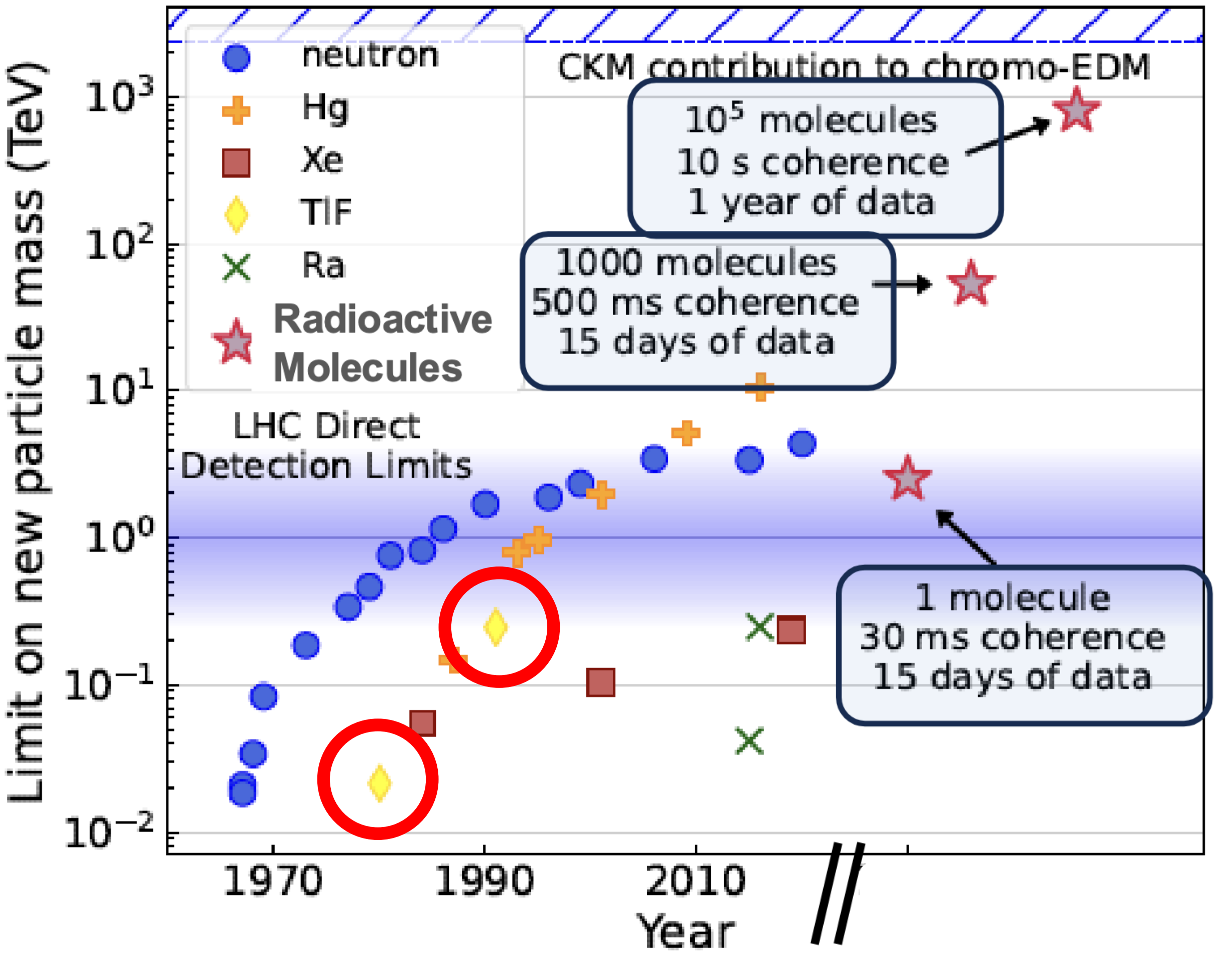
Progress towards a measurement of the Schiff moment of ^{205}Tl in TlF Molecules with CeNTREX

CeNTREX

Cold molecule **N**uclear **T**ime **R**eversal **EX**periment

Dave Kawall, University of Massachusetts Amherst

Progress towards a measurement of the Schiff moment of ^{205}Tl in TlF Molecules with CeNTREX



From Ronald's talk

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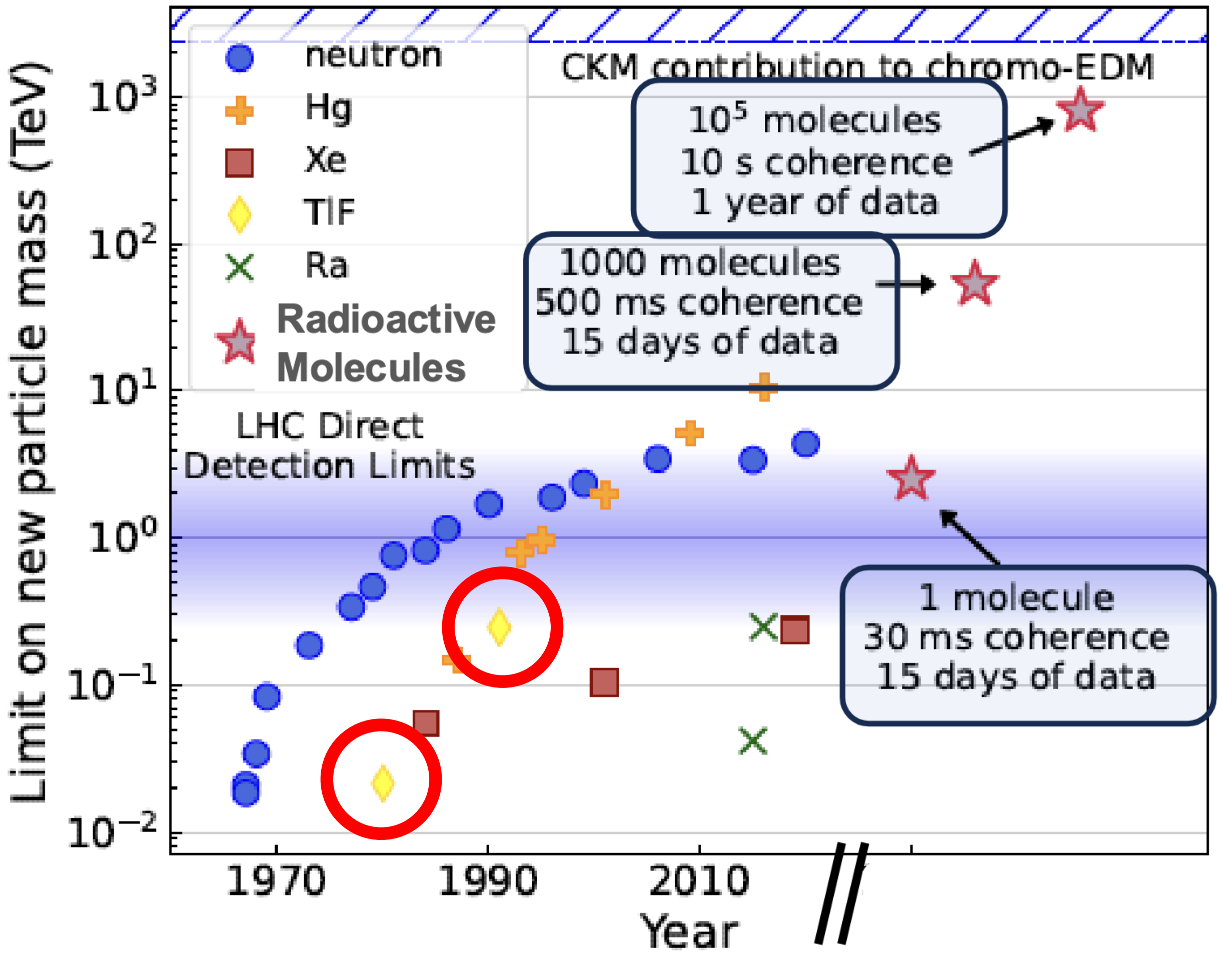

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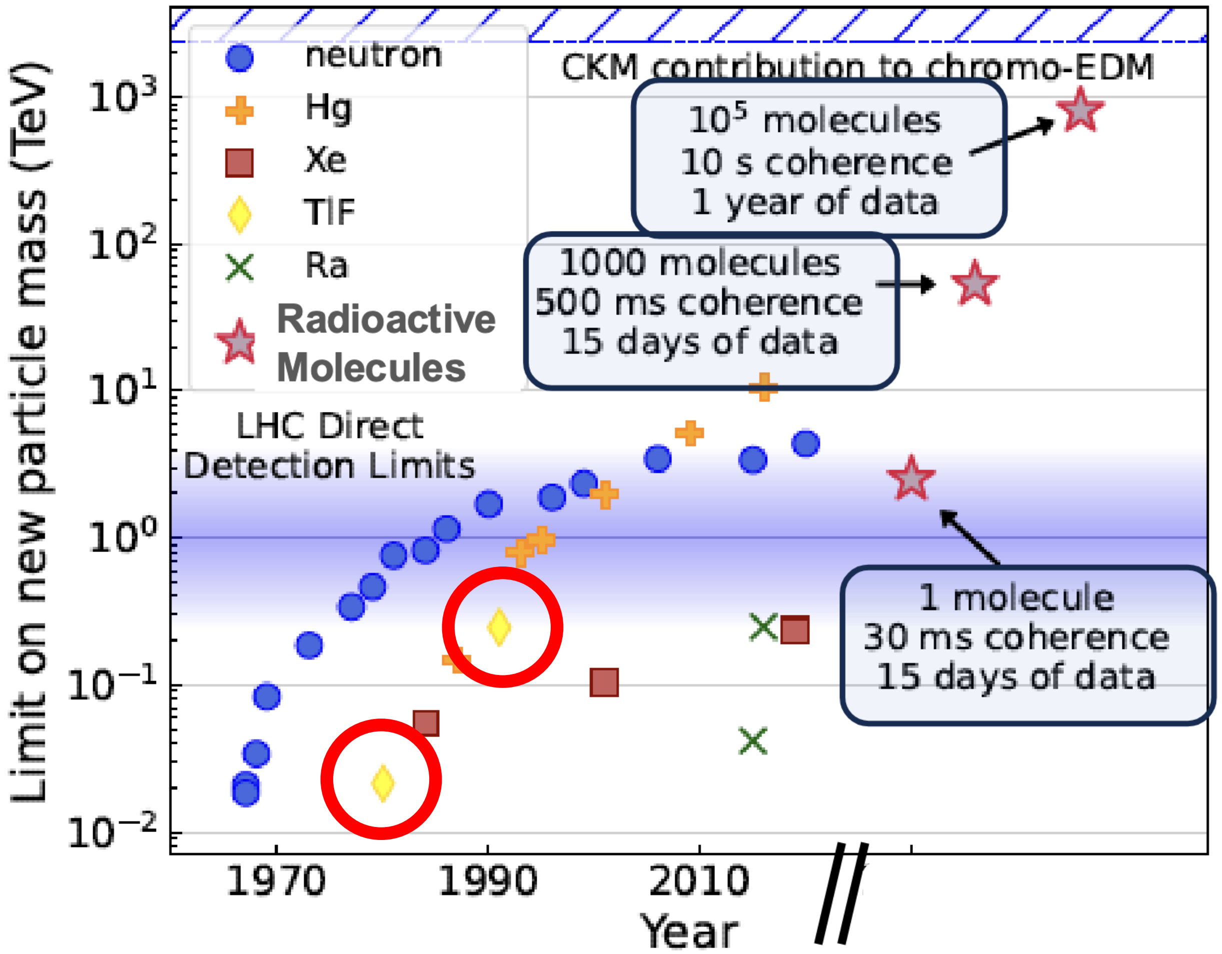

From Ronald's talk

CeNTREX Team

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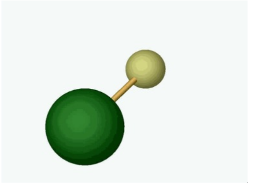

Group

Research Scientist

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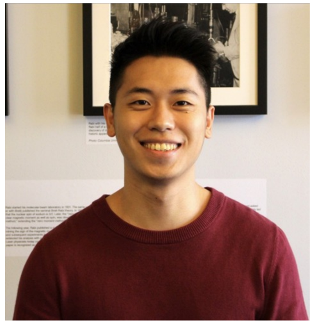

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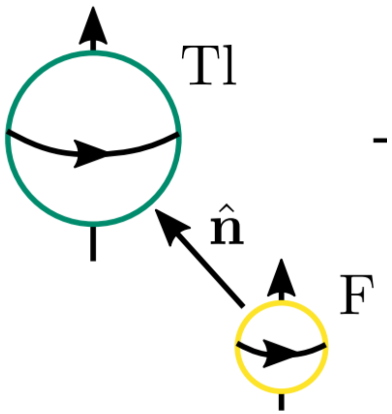
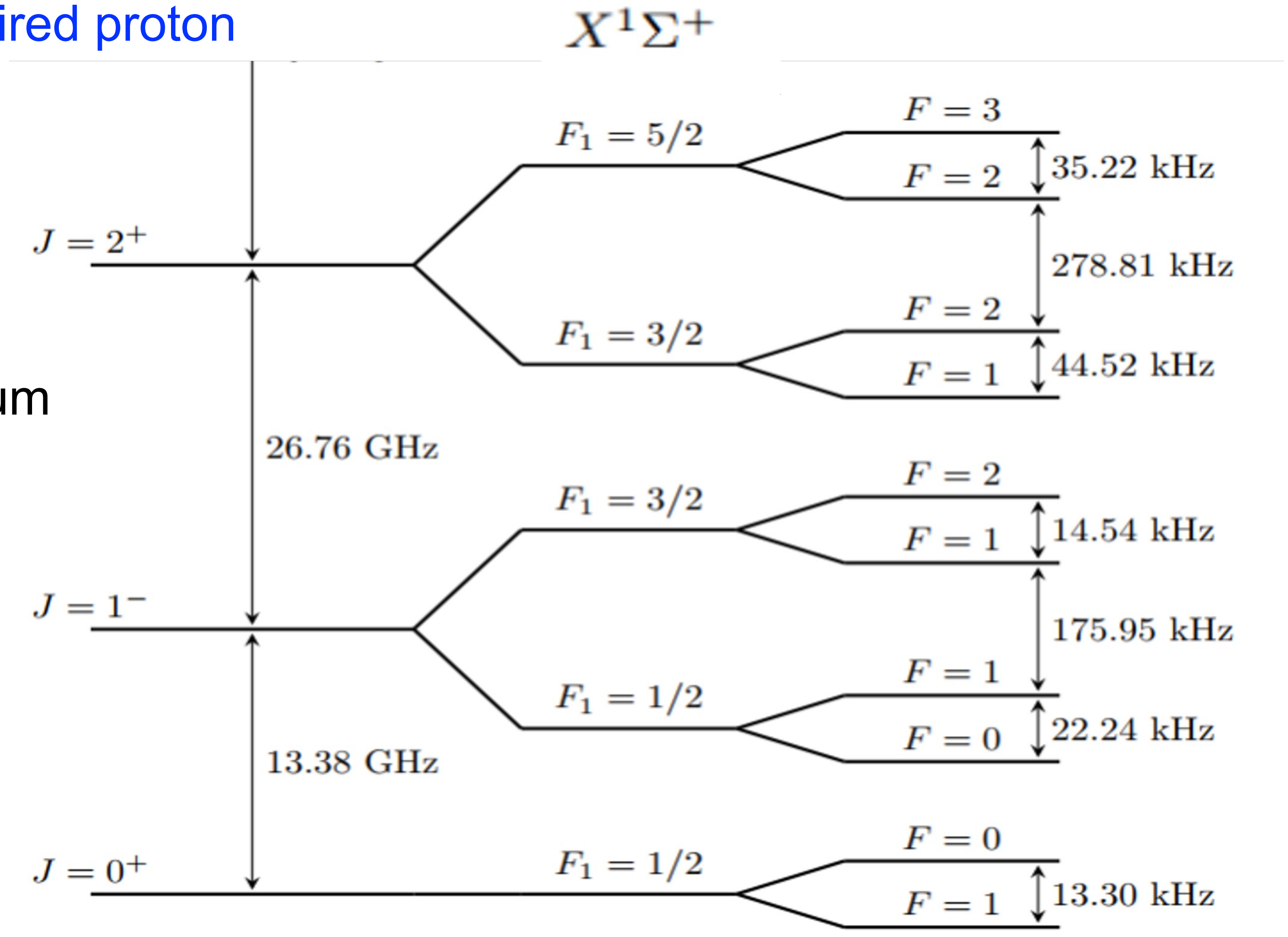
Introduction to Thallium Fluoride

- ^{205}Tl : 81 protons, 124 neutrons, so one unpaired proton
- ^{205}Tl : nuclear spin $I_1 = ½$
- ^{19}F : nuclear spin $I_2 = ½$

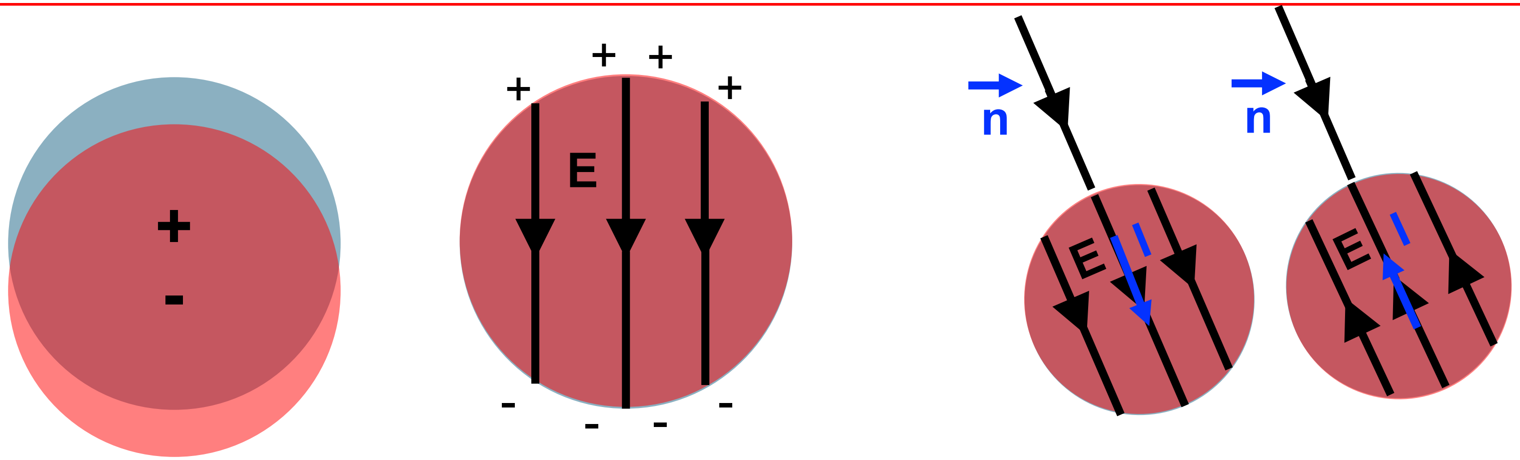
- Molecular ground state: X $^1\Sigma\ (v = 0)$
- No electronic spin or orbital angular momentum
- Rotation **J** and two nuclear spins, $\mathbf{I_1}$ and $\mathbf{I_2}$
- Couple $\mathbf{F_1} = \mathbf{J} + \mathbf{I_1}$, and $\mathbf{F} = \mathbf{F_1} + \mathbf{I_2}$

- Each rotational level J has
  4 x (2J+1) magnetic sublevels





Schiff Moment: Simple-minded view



Surface charge cos(θ)

- Electric field due to nuclear EDM distribution
- Electrons spend some time inside nucleus
  - Energies are shifted if gradient in electron density
- Larger nuclear charge → electron spends more time inside nucleus: $Schiff\ moment \propto Z^2$
- ^{205}Tl nucleus has unpaired proton
- Sensitive to p EDM; previous hadronic EDMs mostly sensitive to n EDM

Effective Hamiltonian Ground State Thallium Fluoride

$$\mathcal{H}_{\mathrm{TlF}} = \mathcal{H}_{\mathrm{rot}} + \mathcal{H}_{\mathrm{sr}} + \mathcal{H}_{\mathrm{ss}} + \mathcal{H}_{\mathrm{S}},$$

$$\mathcal{H}_{\mathrm{rot}} = B\mathbf{J}^2,$$

$$\mathcal{H}_{\mathrm{sr}} = c_1(\mathbf{I}_1 \cdot \mathbf{J}) + c_2(\mathbf{I}_2 \cdot \mathbf{J}),$$

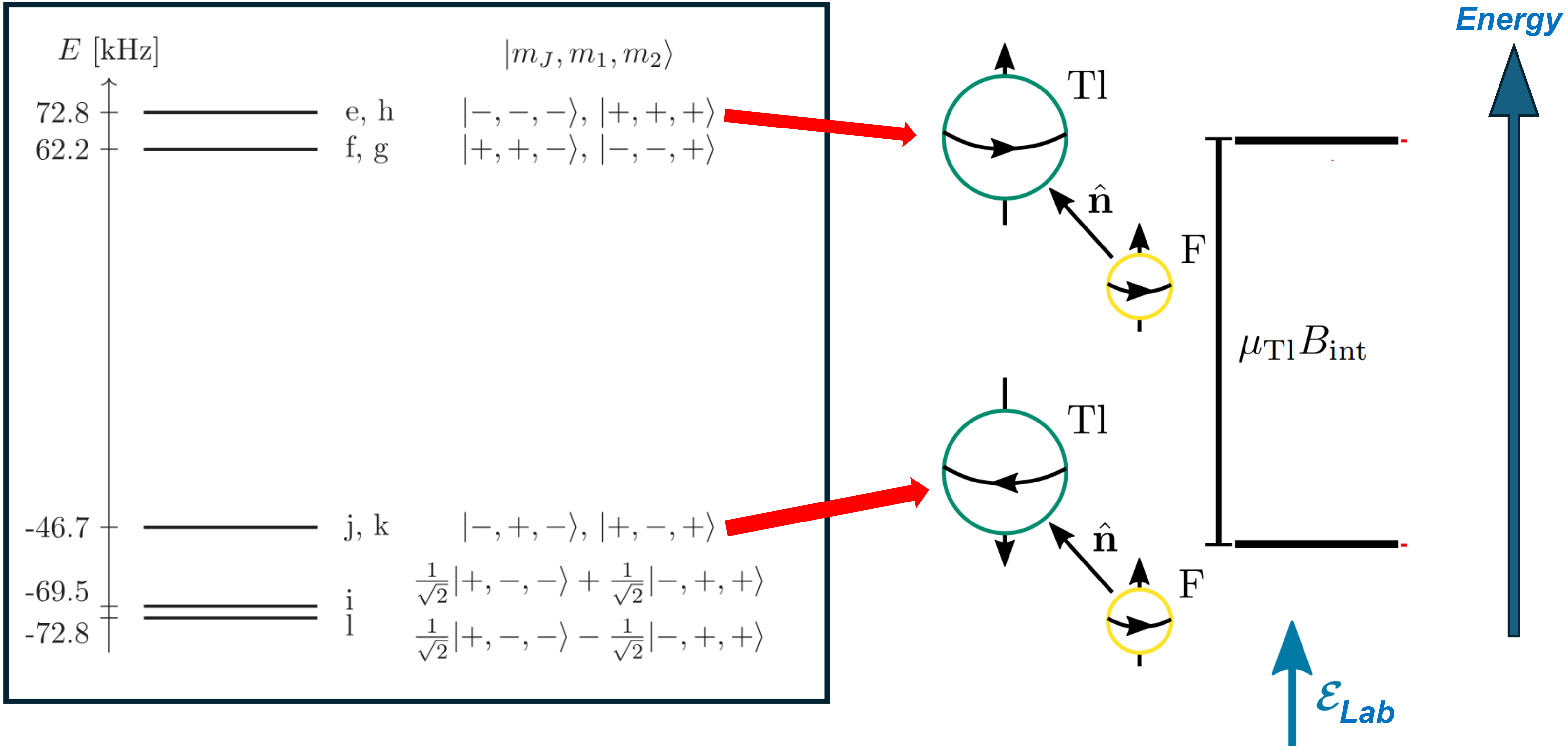
$$\mathcal{H}_{\mathrm{ss}} = c_3 T^2(\mathbf{C}) \cdot T^2(\mathbf{I}_1, \mathbf{I}_2) + c_4(\mathbf{I}_1 \cdot \mathbf{I}_2),$$

$$\mathcal{H}_{\mathrm{S}} = -\mu_e \cdot \boldsymbol{\mathcal{E}},$$

B	$=$	6.66733	GHz	μ_e	$=$	2.1285(4)	MHz/V/cm
c_1	$=$	126.03(12)	kHz	c_2	$=$	17.89(15)	kHz
c_3	$=$	0.70(3)	kHz	c_4	$=$	$-13.30(72)$	kHz

- Interesting feature: spin-rotation term behaves like internal magnetic field

^{205}TlF ground state energy levels at high electric field



- Rotation creates internal magnetic field along z

Effect of Schiff Moment in Thallium Fluoride

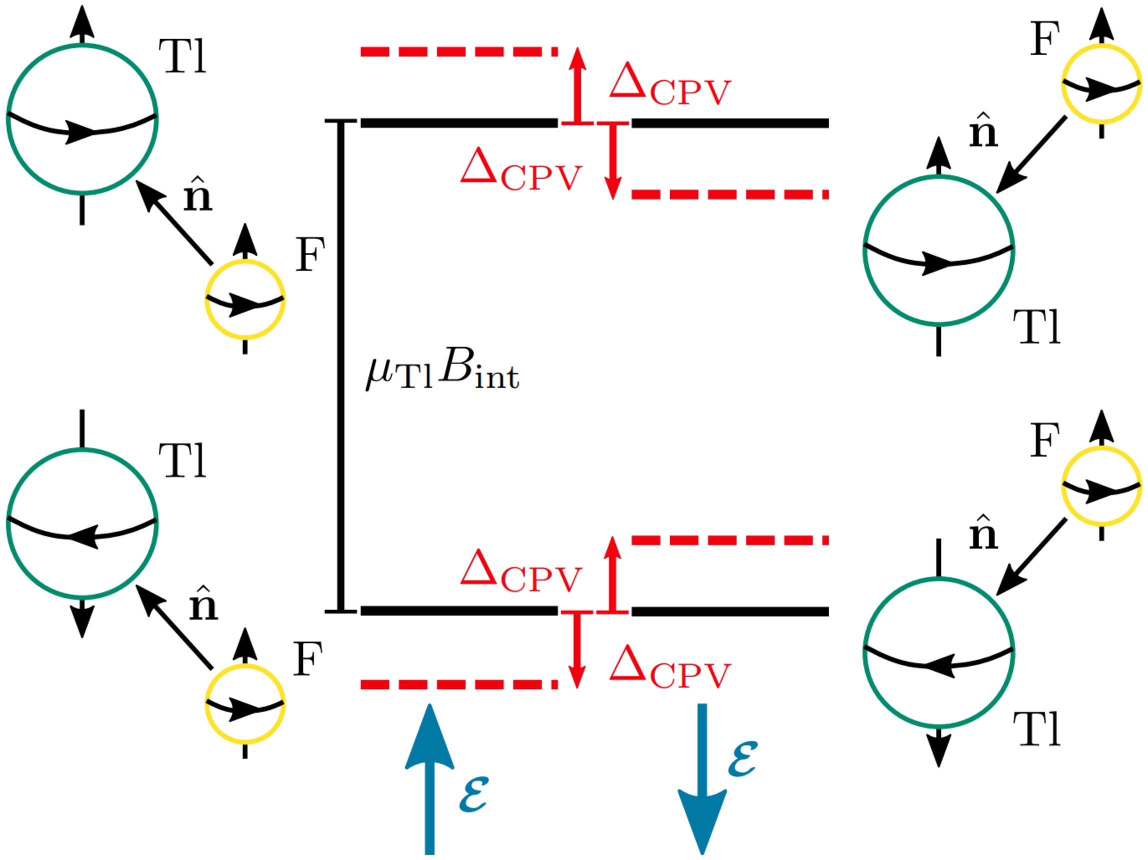
CP-Violating Effective Hamiltonian:

$$H_{\text{CPV}} = W_S S \frac{\boldsymbol{I}}{I} \cdot \hat{n}$$

- W_S = intramolecular energy associated with NSM
- S = Schiff moment
- $\boldsymbol{I}$ = nuclear spin = 1/2

Energy shift due to CPV:

$$\Delta_{\text{CPV}} = W_S S \mathcal{P}$$



- $\mathcal{P}$ =degree of polarization w.r.t $\boldsymbol{\mathcal{E}}$ ($\mathcal{P} = 0.547$ @ 30 kV/cm)
- $W_S = (1.8 \pm 0.2) \times 10^6\ Hz\,/(e\,fm^3)$ (M. Hubert, Timo Fleig, Phys Rev A **106**, 022817 (2022))
- $S(^{205}\text{Tl})$ = $(3.9 \pm 6.8) \times 10^{-11}$ *e* fm^3 ⟷ $(1.4 \pm 2.4) \times 10^{-4}$ Hz. (D. Cho et al, Phys Rev A **44**, 2783 (1991))

Schiff Moment in Thallium has many possible sources

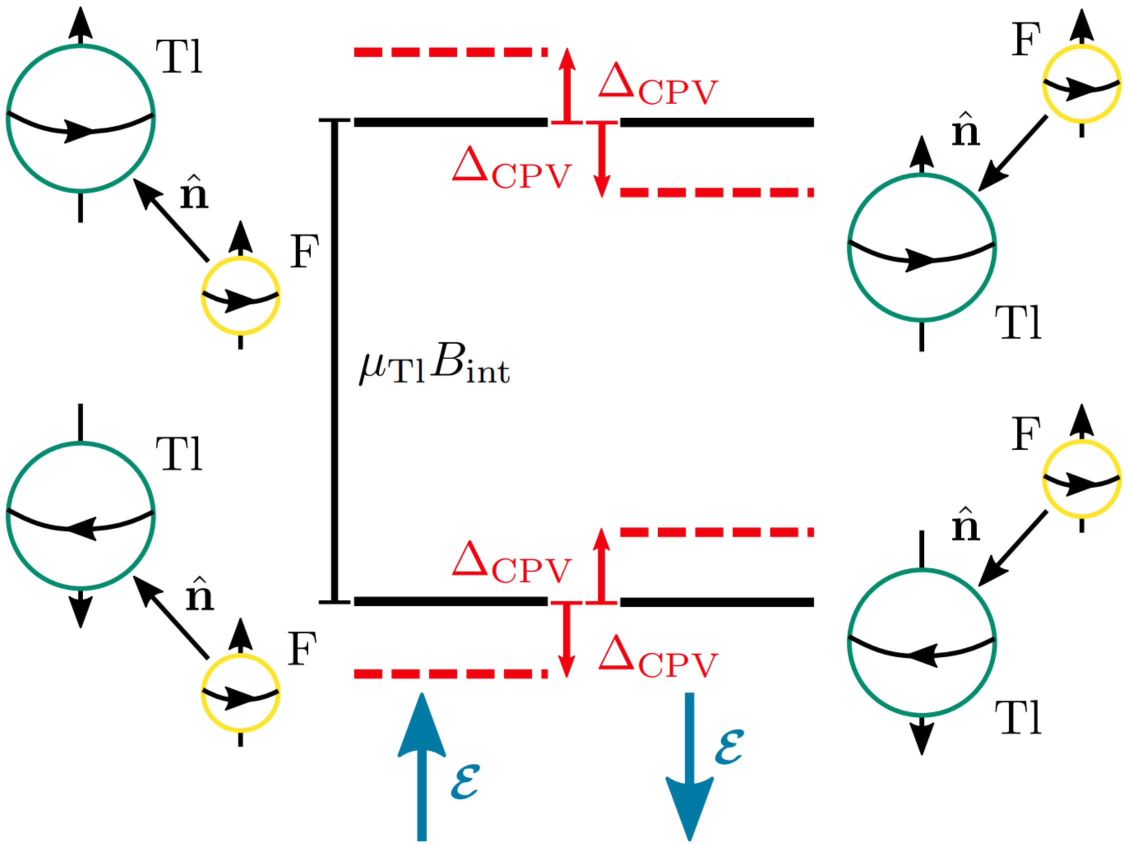
Extract S from $\Delta_{\text{CPV}} = W_S S \mathcal{P}$

$S(^{205}\text{Tl}) \simeq (13 g\bar{g}_0 - 0.04 g\bar{g}_1 - 0.27 g\bar{g}_2)\, e\ \text{fm}^3$

$S(^{205}\text{Tl}) \simeq 0.027\bar{\theta}\ e\ \text{fm}^3$

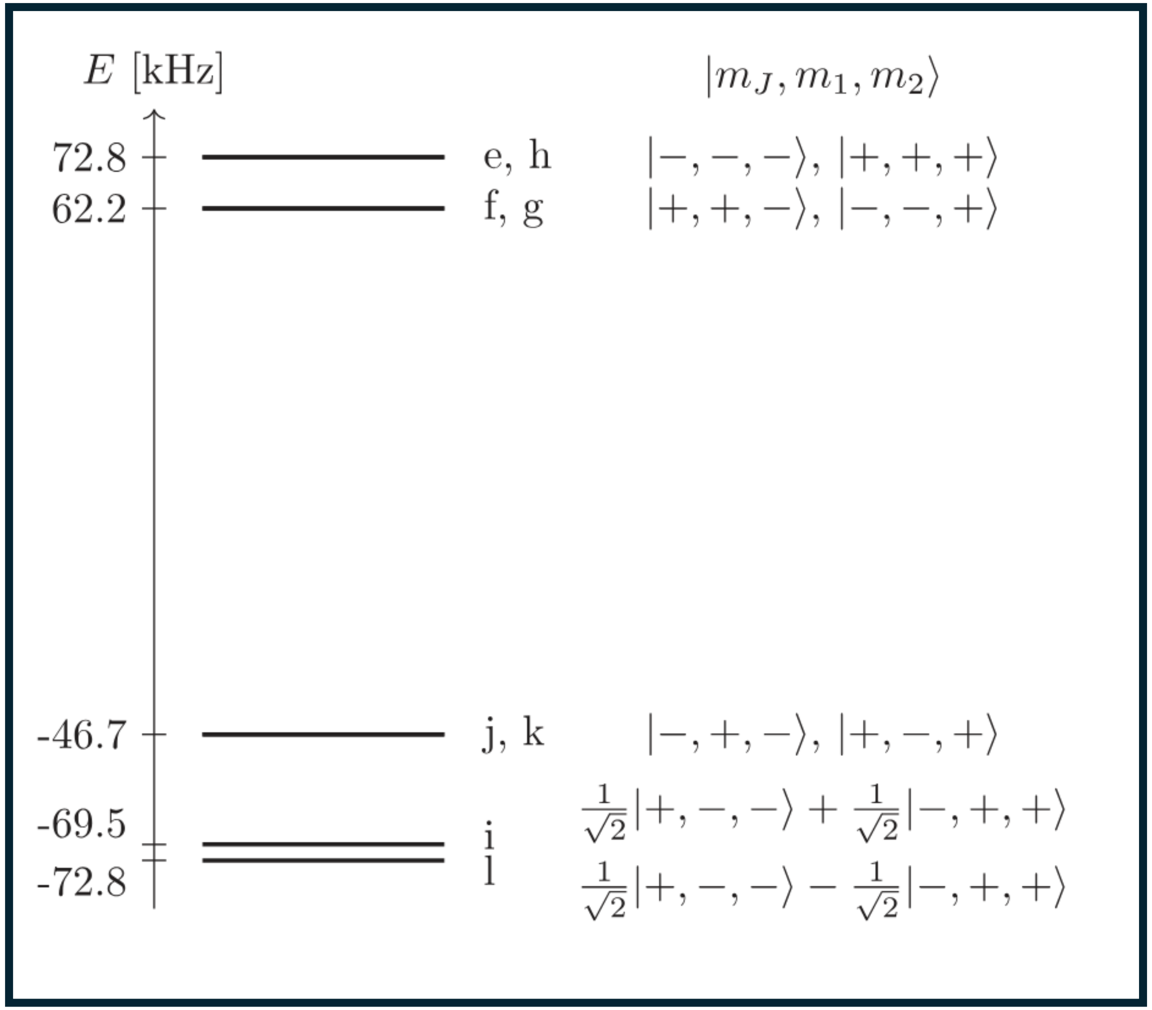
$S(^{205}\text{Tl}) \simeq (12\tilde{d}_d + 9\tilde{d}_u) e\ \text{fm}^2$

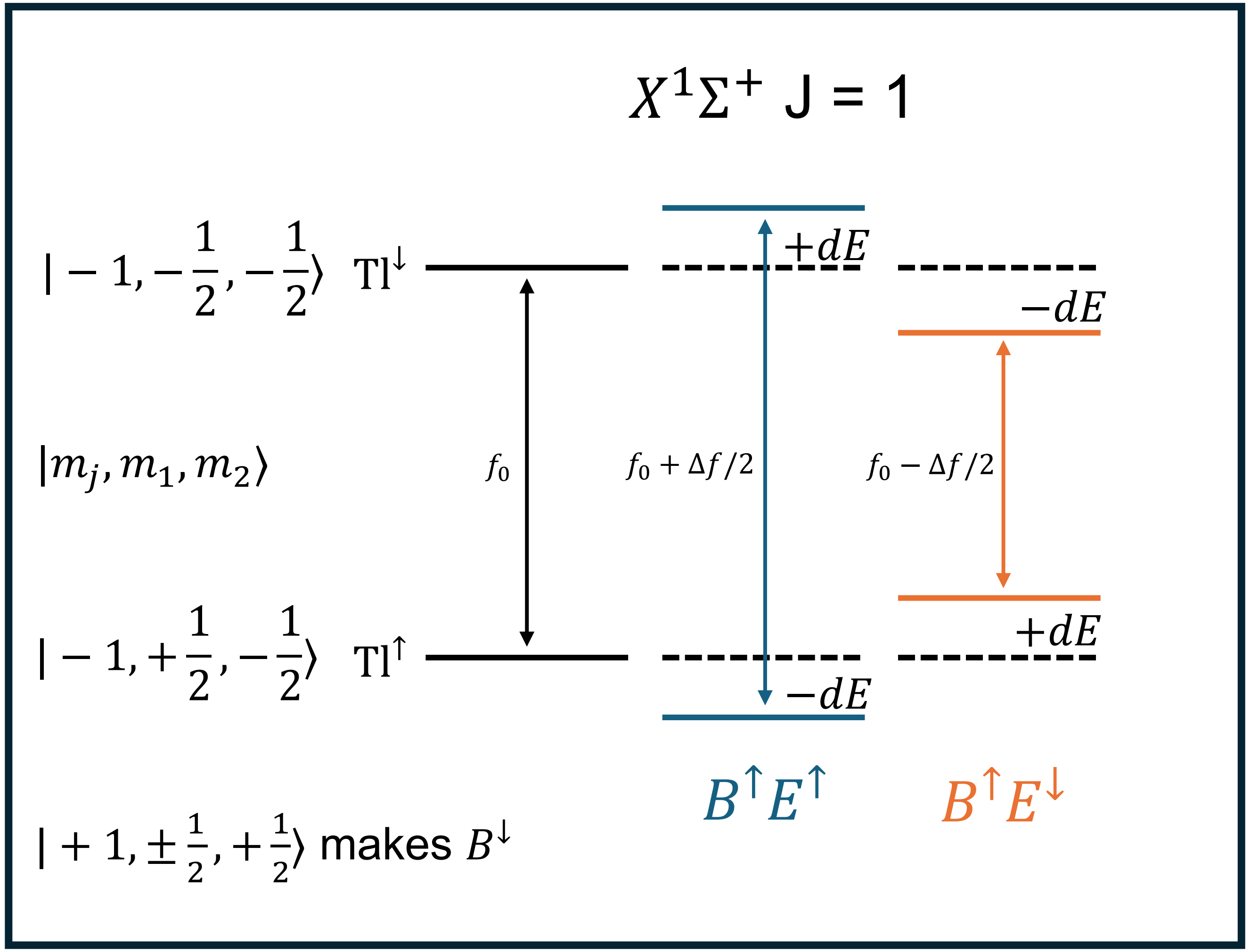
$S(^{205}\text{Tl}) \simeq 0.4 d_p\ \text{fm}^2$



V.V. Flambaum and V.A. Dzuba, Cho et al, Phys Rev A **101**, 042504 (2020)

^{205}TlF ground state energy levels at high electric field



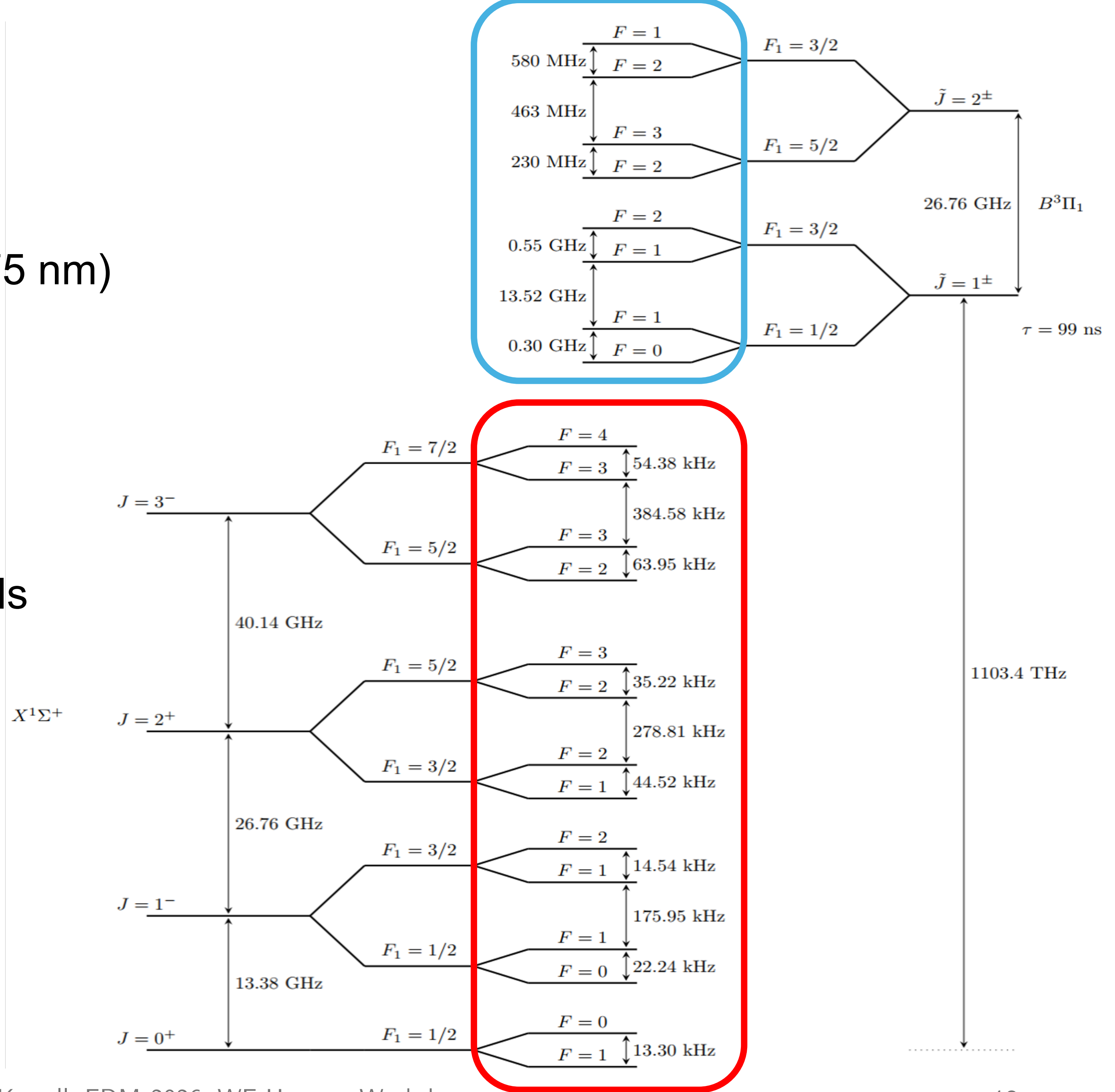


- EDM search uses e+j and/or h+k states in which Tl spin flips
- (e+j) and (h+k) have opposite signs of internal B field
- Need molecules in J=1, high electric field

^{205}TlF Energy Levels

Zero electric field

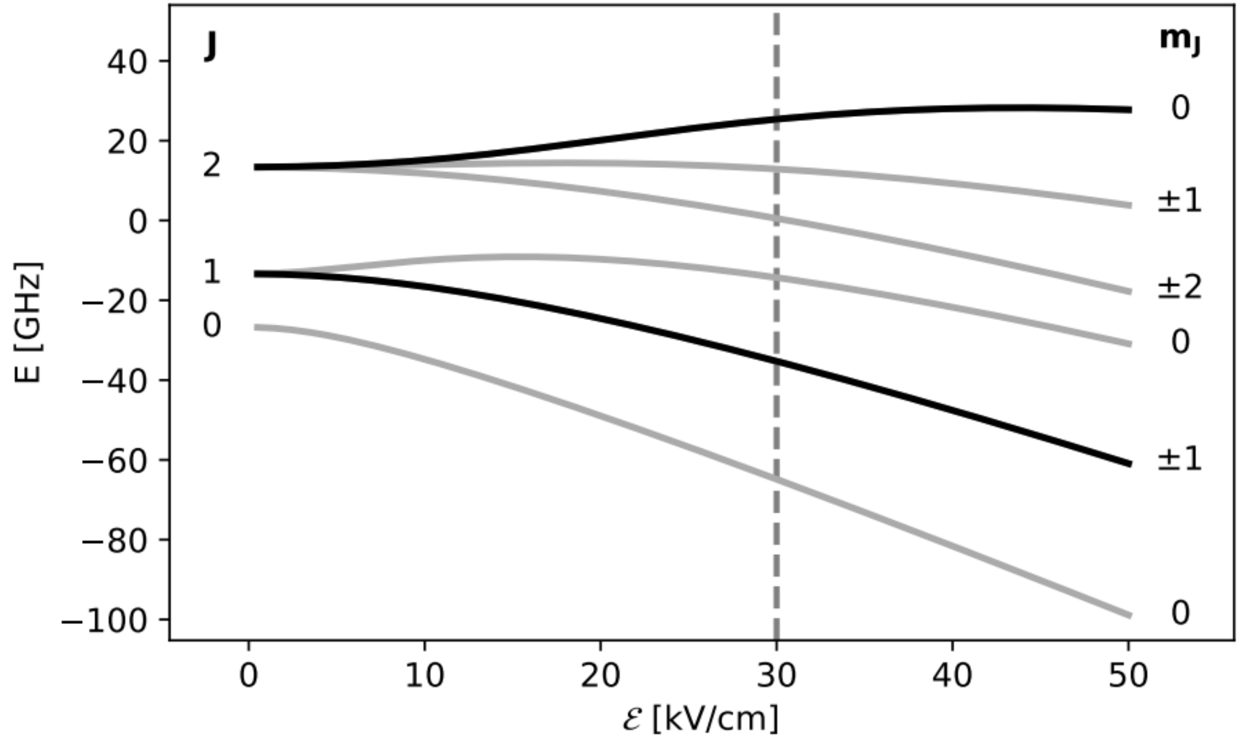
- Laser transitions are UV (1100 THz, 271.75 nm)
  - X ground state hfs ≤ 100 kHz
  - B excited state hfs ≥ 100 MHz
  - $\Gamma = 2\pi \times 1.6$ MHz
  - Dark states; $n_g \gg n_e$
- Use microwaves to change rotational levels



Ground State Energy Levels in Electric Field

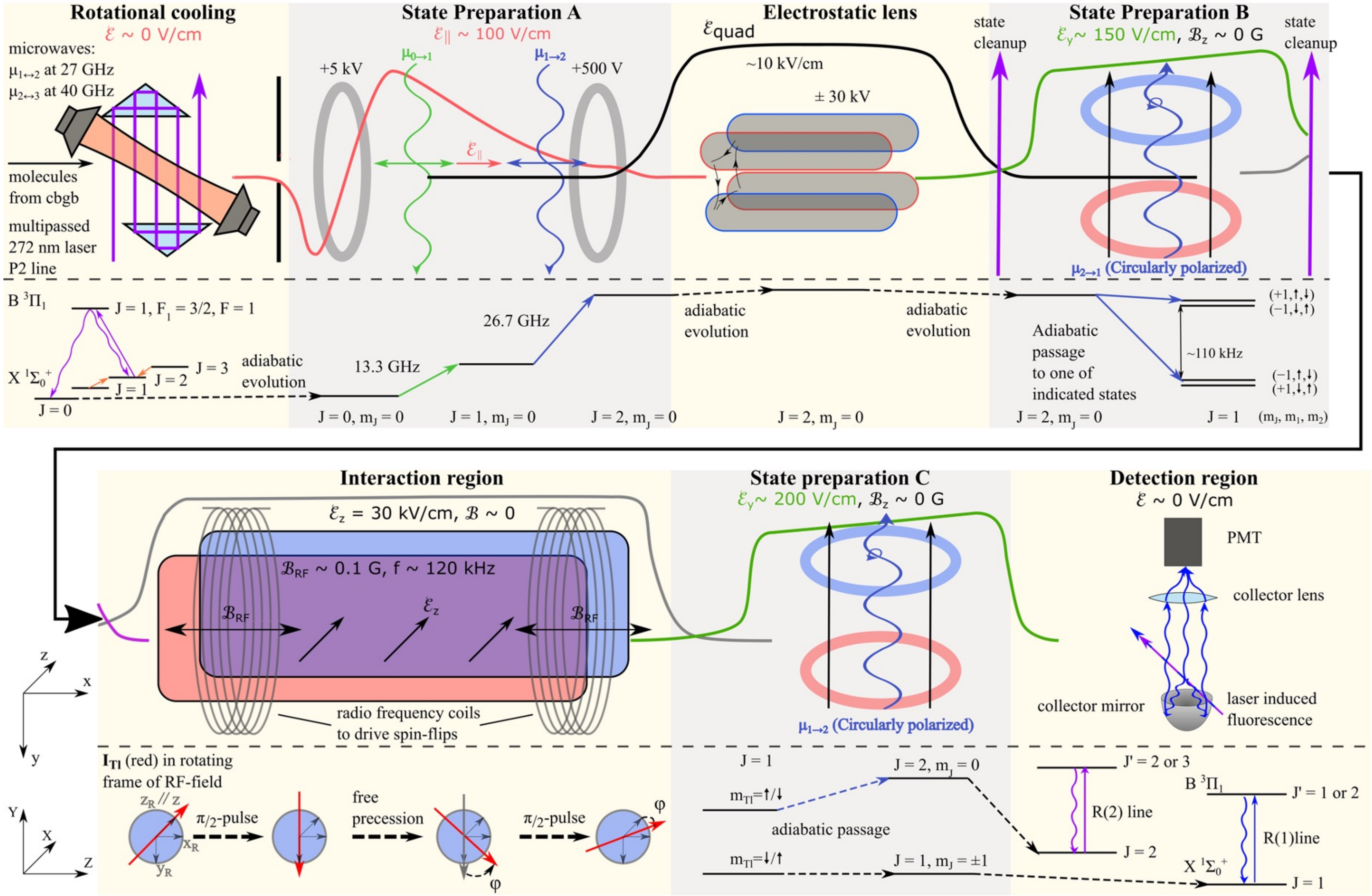
$\Delta_{\mathrm{CPV}} = W_S S \mathcal{P}$

- $\mathcal{P}$ larger for smaller J
  - $\mathcal{P}$ arises from mixing between states with different parity, i.e. different J
  - J states closer together for smaller J
- systematics supressed with strong spin-rotation
  - requires $m_J \neq 0$
  - ‘effective’ intra-molecular magnetic field $\mathcal{B}_{\mathrm{int}}$
  - no external magnetic field required

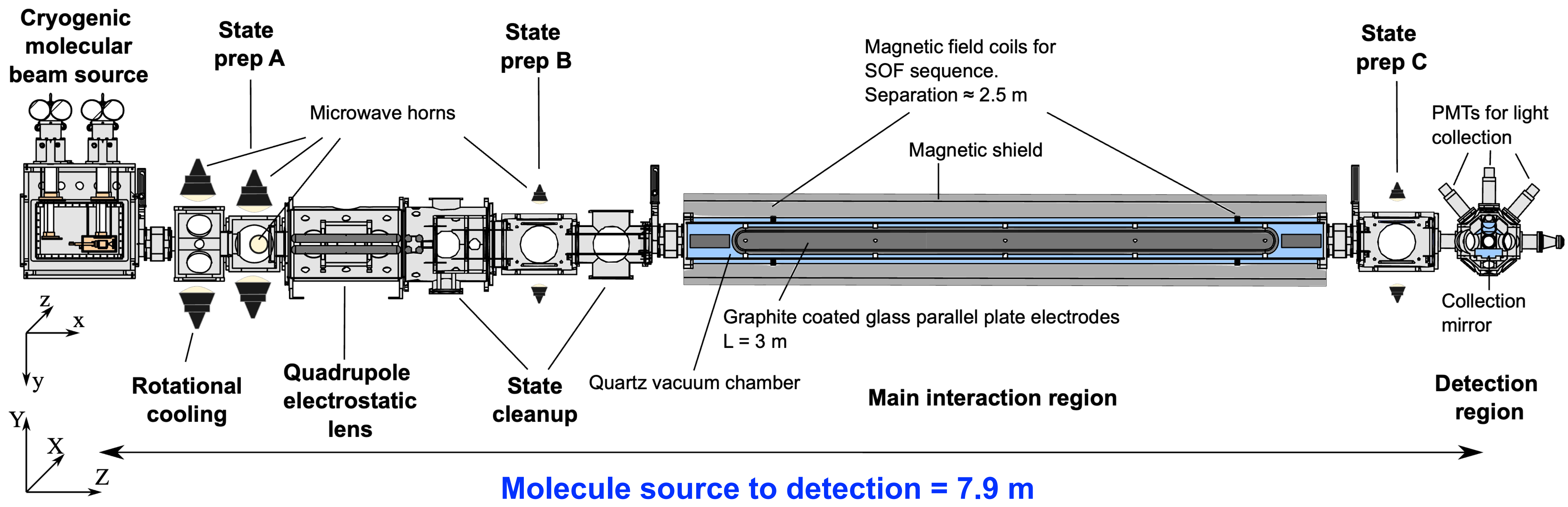


- Schiff Moment measurement performed in $\left|\tilde{J} = 1, m_J = \pm 1\right\rangle$

Experiment Overview



Experiment Overview: Beamline

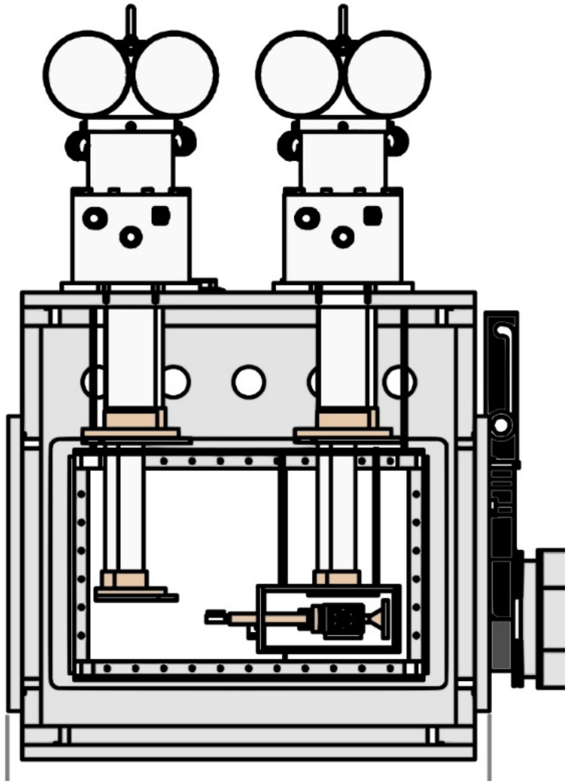


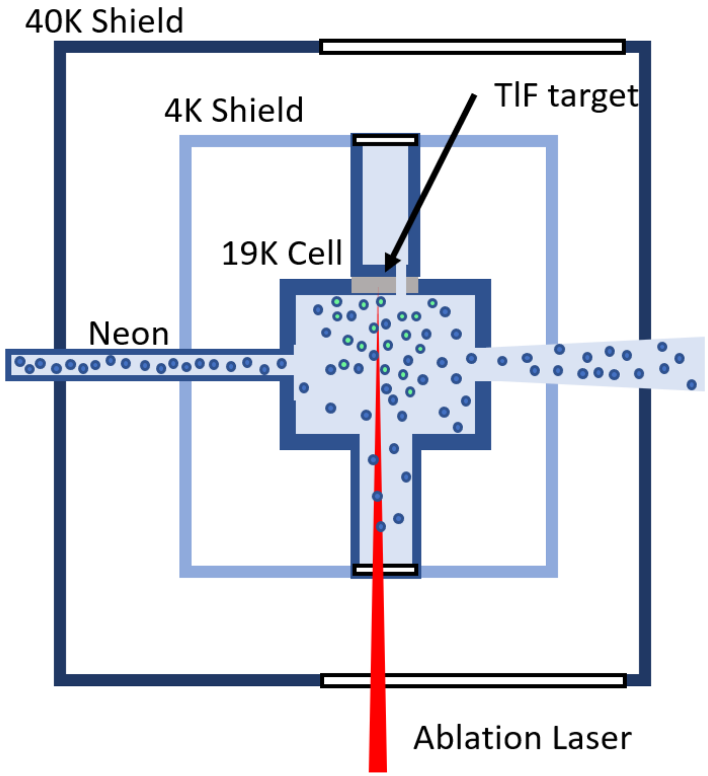
Experiment Overview: Beamline

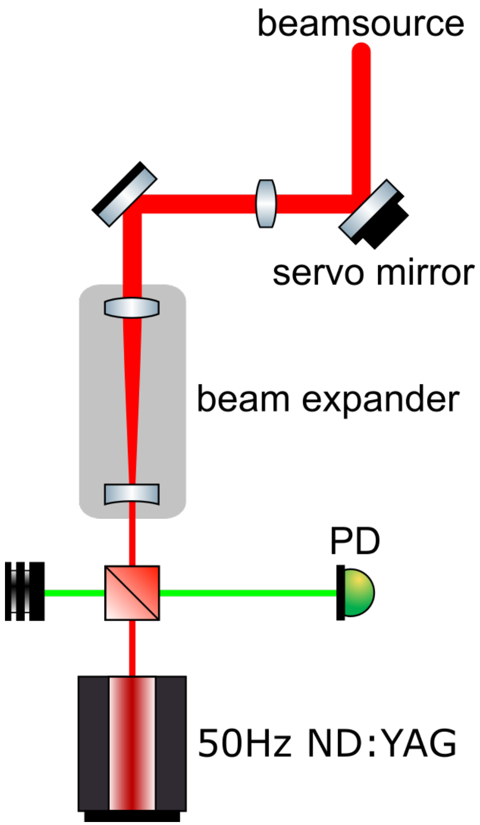
Still Officially Tabletop!

Longest table: Taj Falaknuma Palace
33 meters long = 4 x CeNTREX !
Seats 100

Cryogenic Buffer Gas Beam Source of TlF

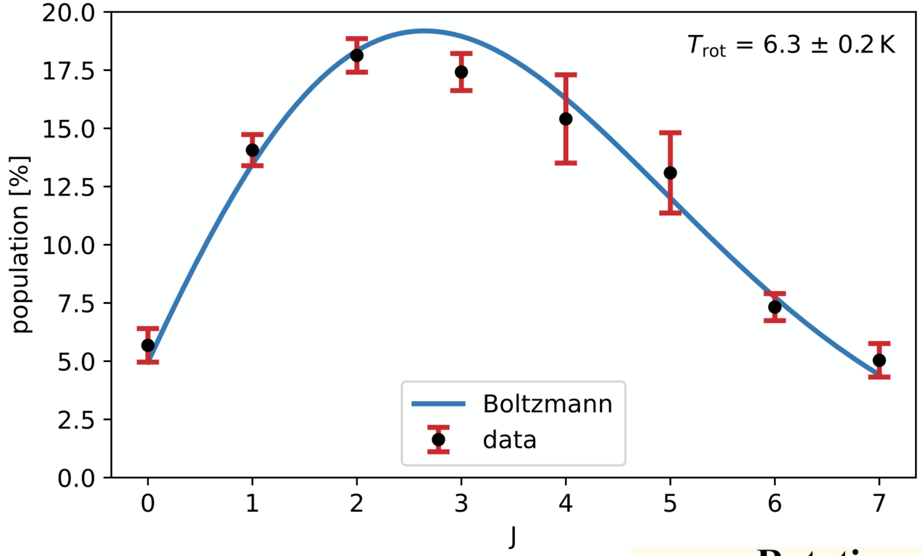




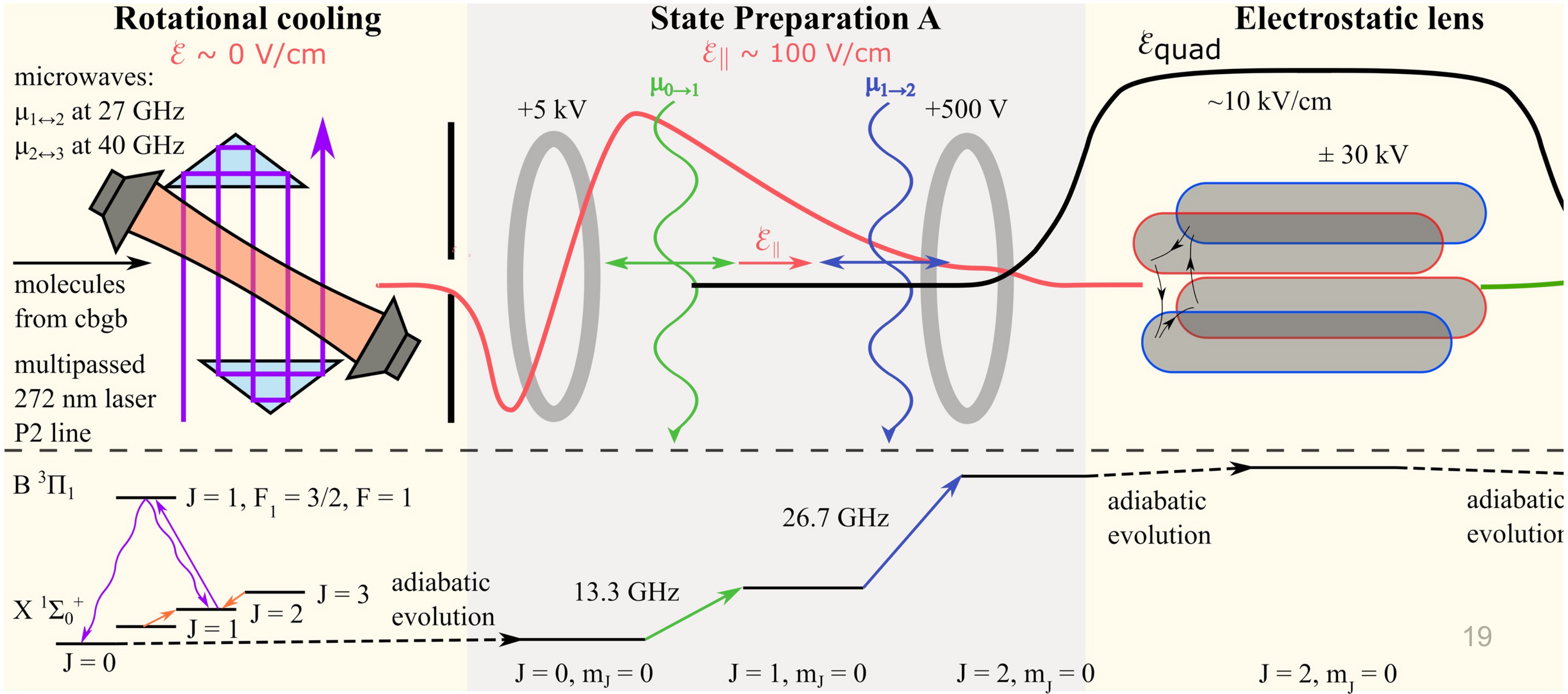


- Cold cell @19K cools down neon buffer gas
- YAG pulse ablates TlF target at 50 Hz
- Collisional cooling + supersonic expansion creates cold TlF beam
- 6.3(2) K rotational temperature, 184(16) m/s forward velocity, $5{\times}10^{12}$ molecules/state/sr/s

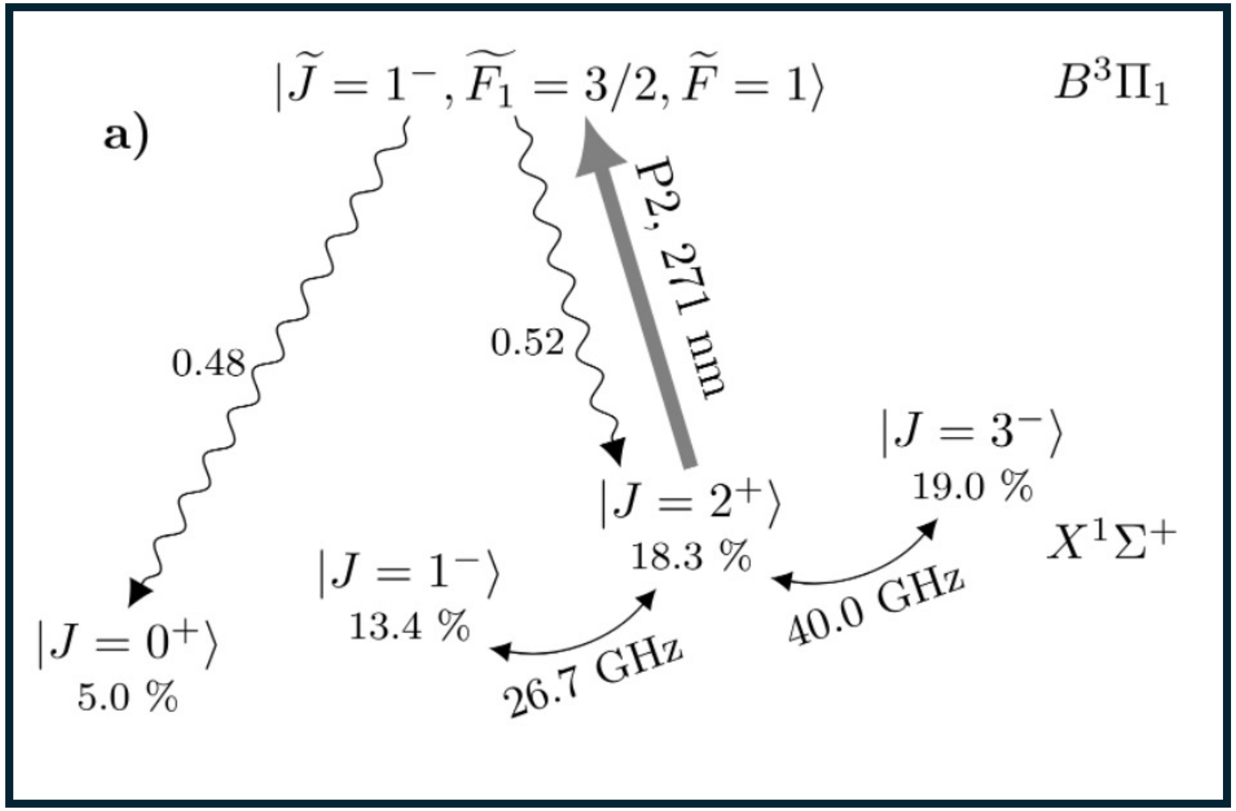
Rotational Cooling + Electrostatic Lens: Increase #molecules for measurement

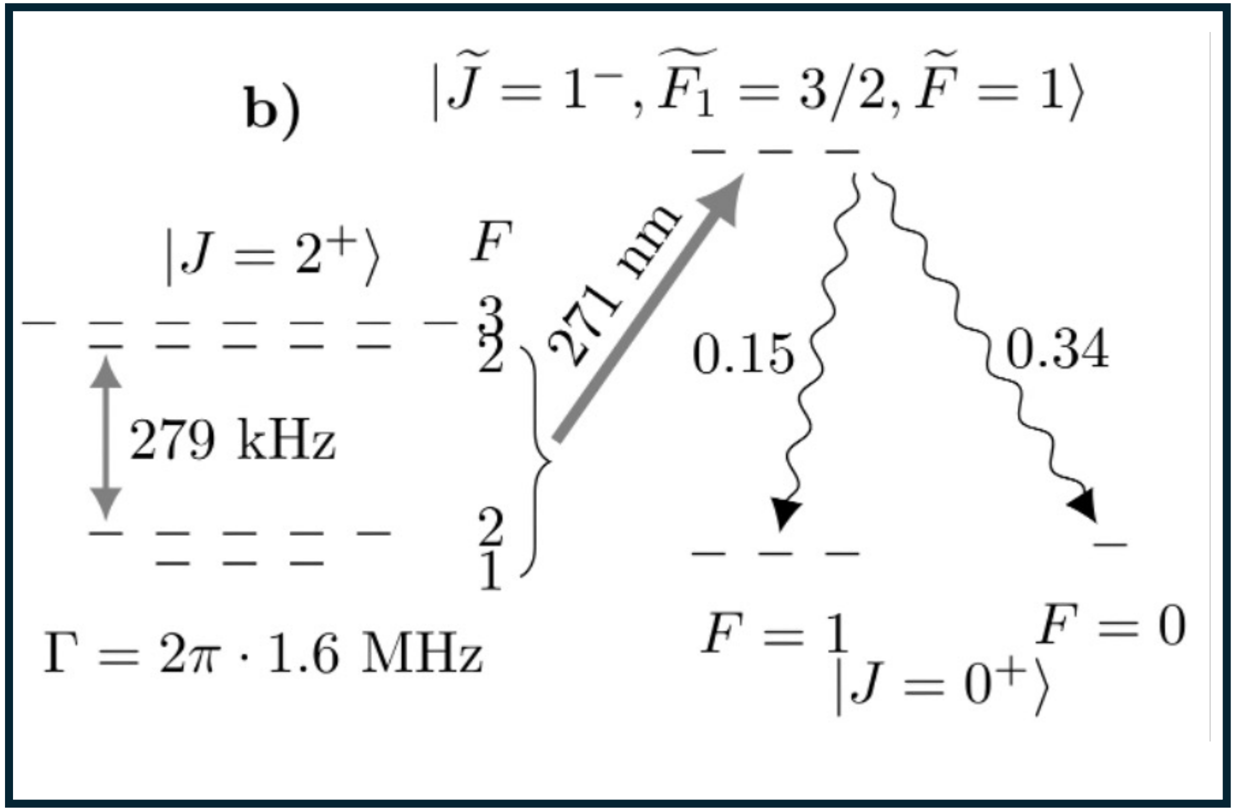


- $T_{rot} = 6.3 \pm 0.2$ K
- ≈56% in lowest 4 rotational states
- Rotational cooling: transfer J=3, 2, 1 into J=0
- Electrostatic focusing



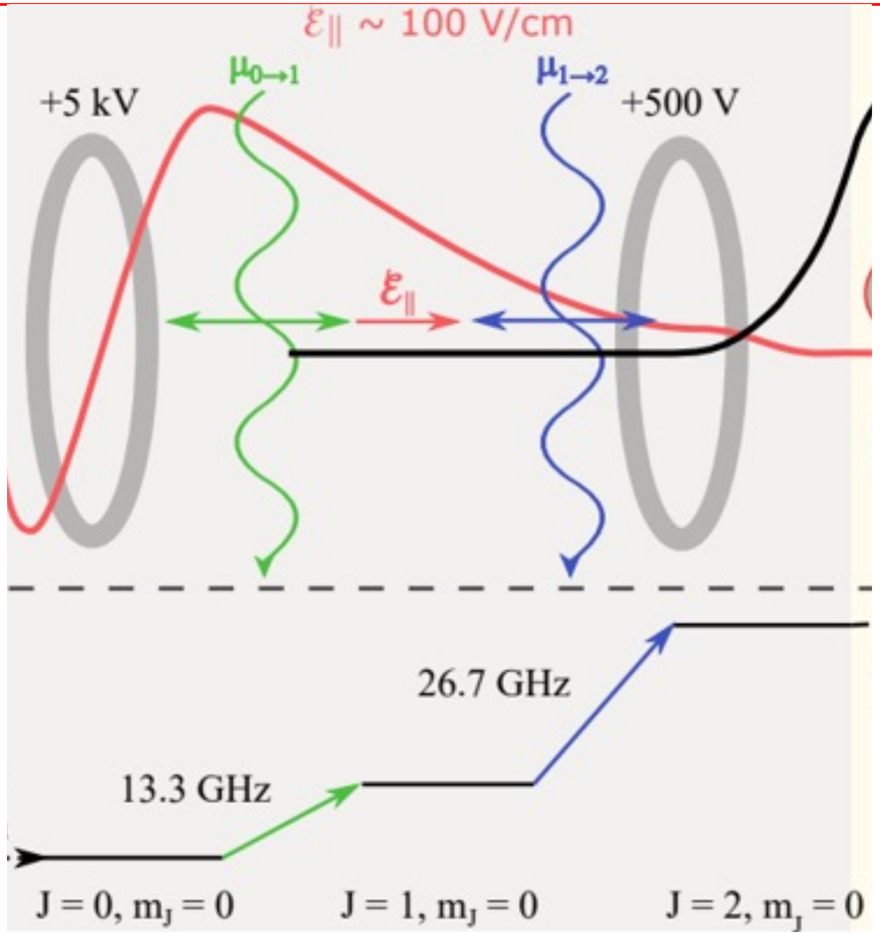
Rotational Cooling: Transfer population from J=3, 2, 1 to J=0

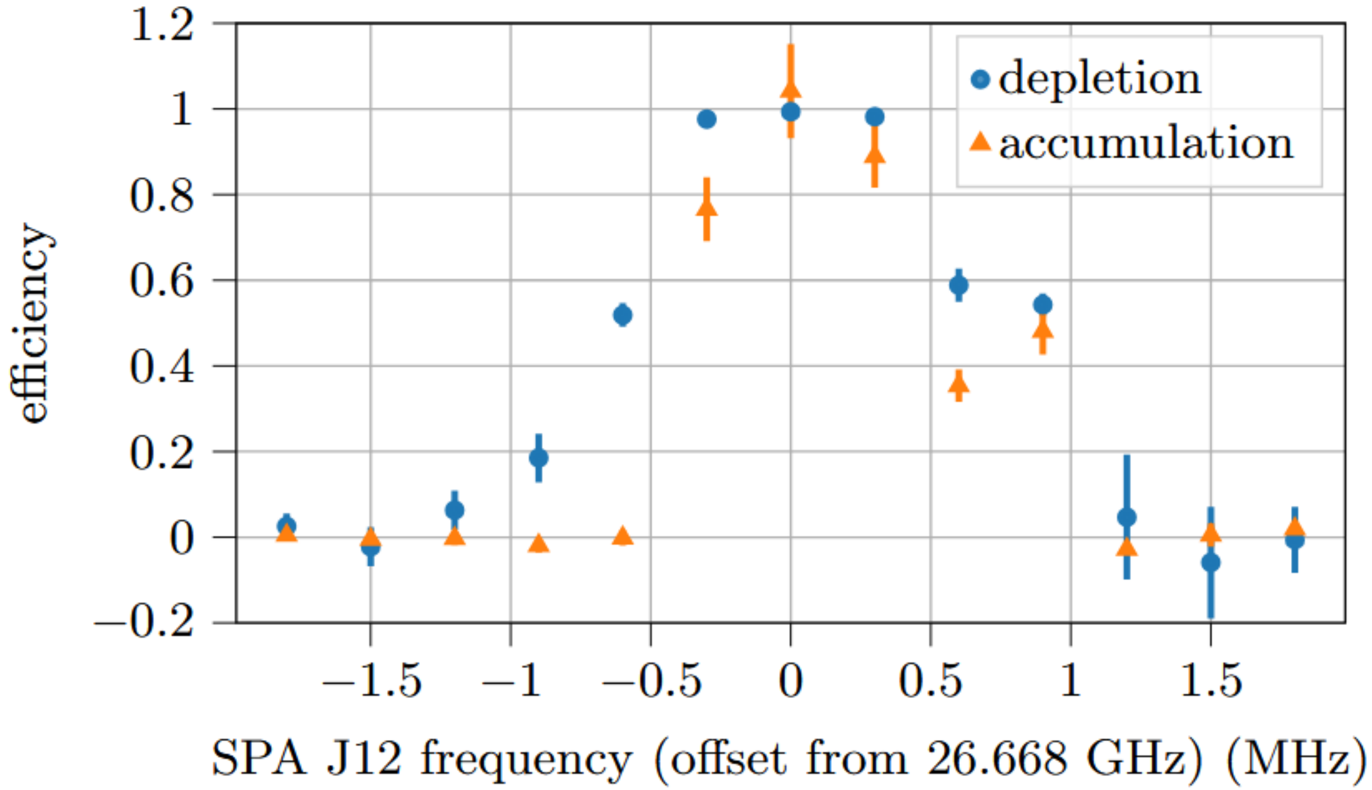




- Gain of 20.1(4) achieved (@ 6.3 K)
- Full rotational cooling in $J = 1, 2, 3$ would give ≈ 29 gain
- Limited by laser power; currently 90 mW @ 271 nm

State Preparation A: Transfer from J=0,m$_J$=0 to J=2,m$_J$=0 for electrostatic lens





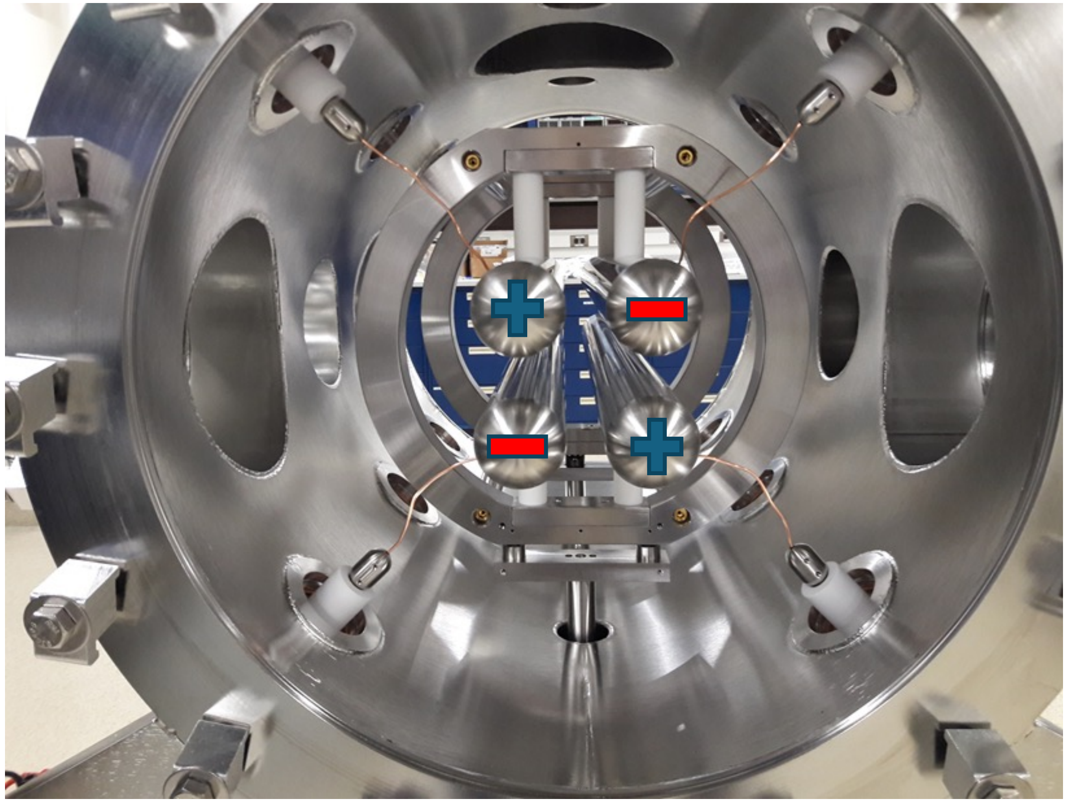
Adiabatic passage from $J = 0, I_{tot} = 0 \rightarrow J = 1 \rightarrow J = 2, I_{tot} = 0$

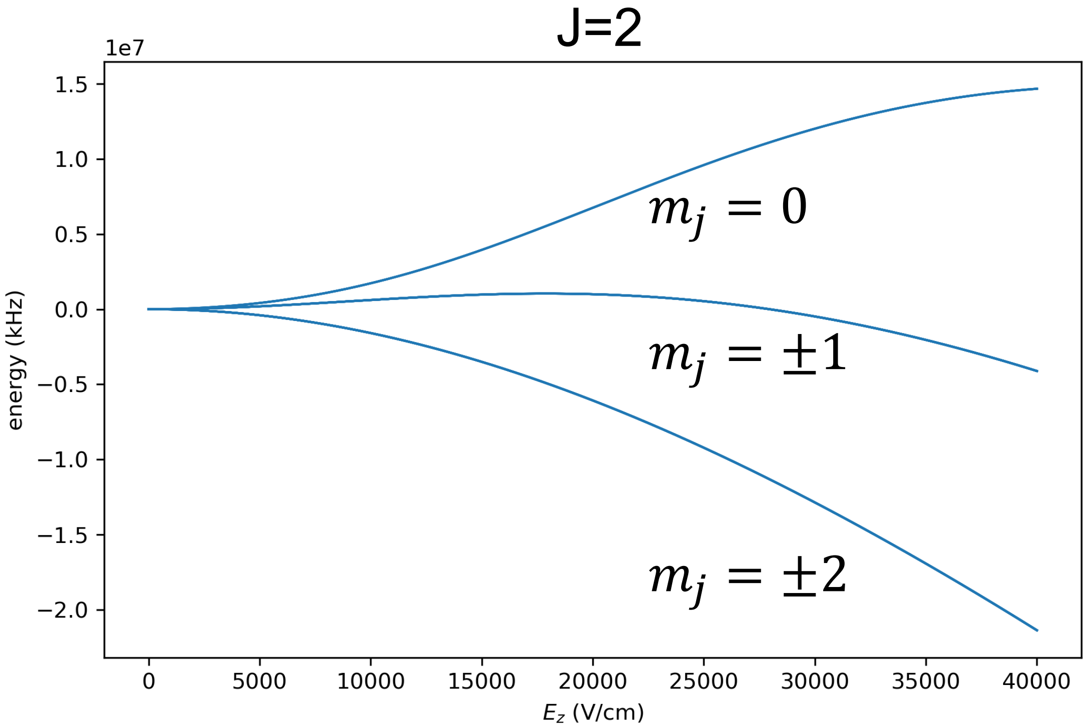
- Microwave driven with Gaussian focusing horns to localize fields
- Spatially-varying DC electric field yields Stark shift

Efficiency

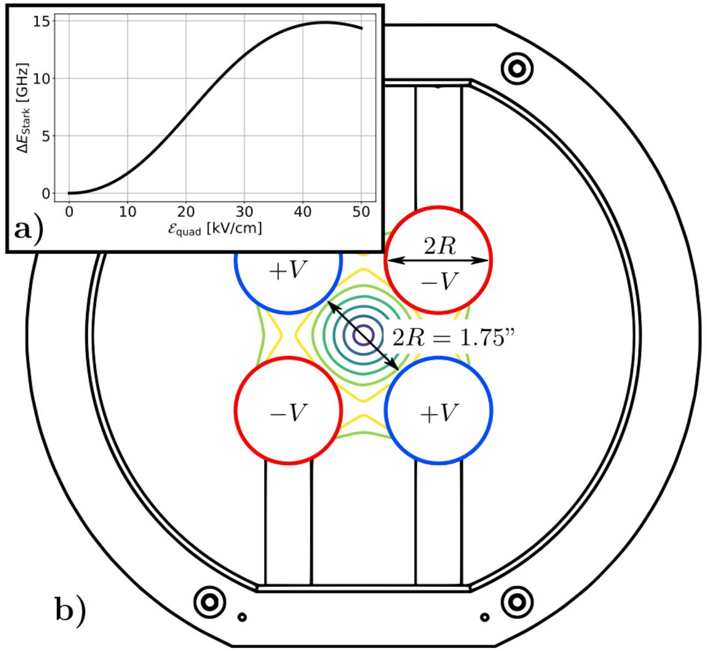
- $J = 0 \rightarrow J = 1 \Rightarrow 88(11)\%$
- $J = 1 \rightarrow J = 2 \Rightarrow 104(11)\%$

Electrostatic Focusing

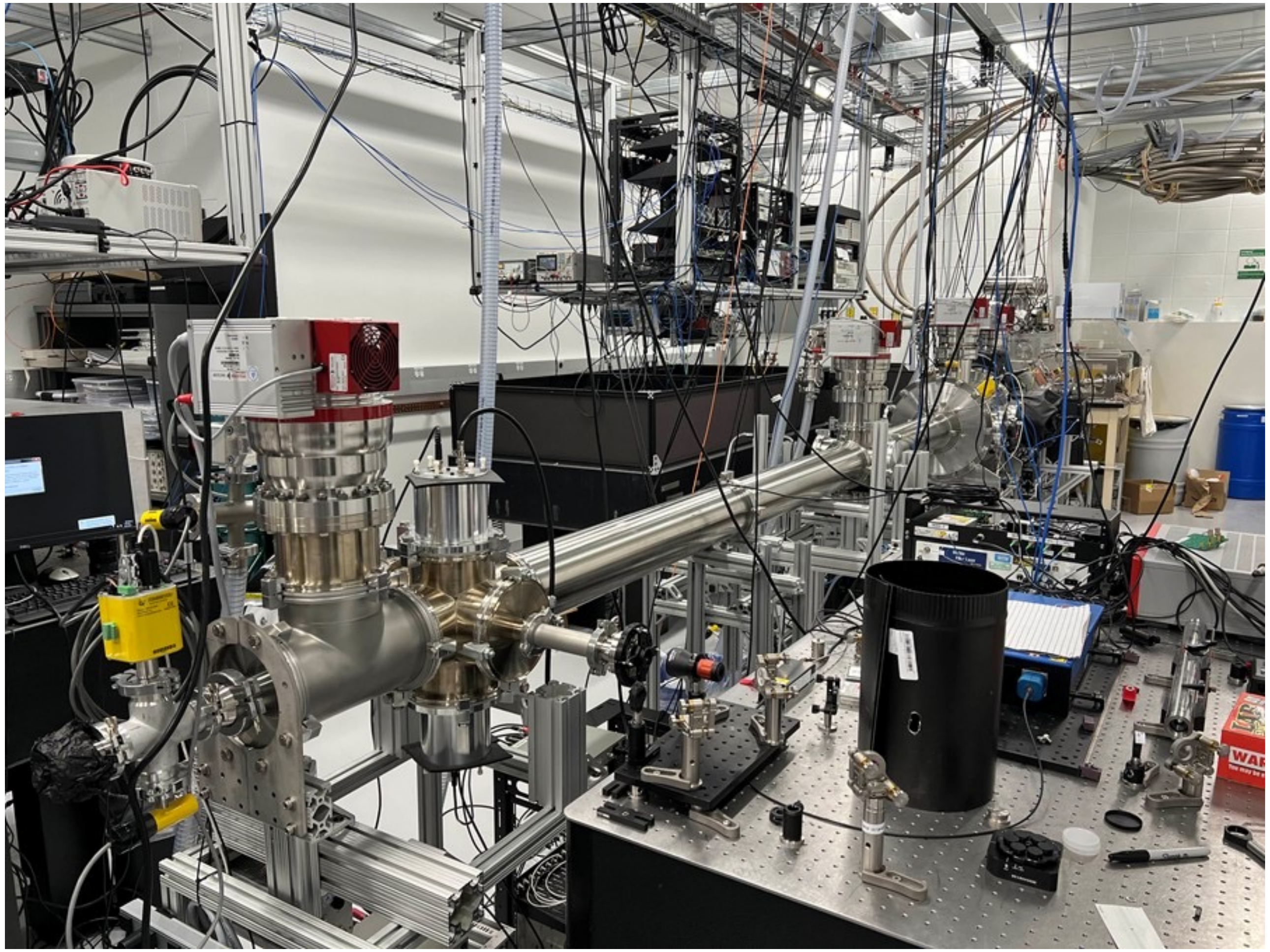


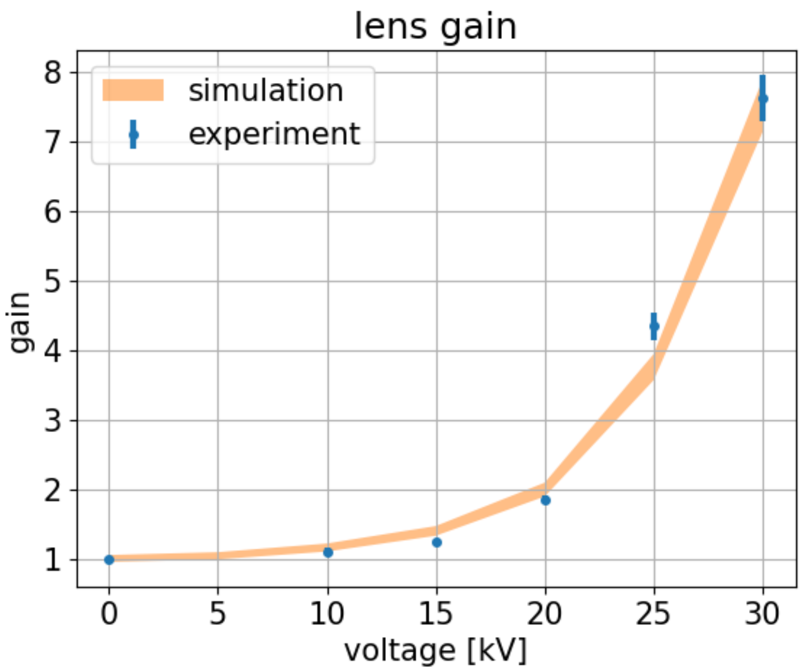


- 4 rods, 60 cm long, rod diameter = separation=1.75”
- Quadrupole config makes linear radial electric field
- ≈ quadratic Stark shift of low-field seeking state $J = 2, m_J = 0$
- Looks like harmonic potential, analogous to thick lens
- Measured gain at 1.26 m from lens of ≈ 7.5(7)
- At full scale expect gain in lensing state of 23 {lens in/out}



Electrostatic Focusing





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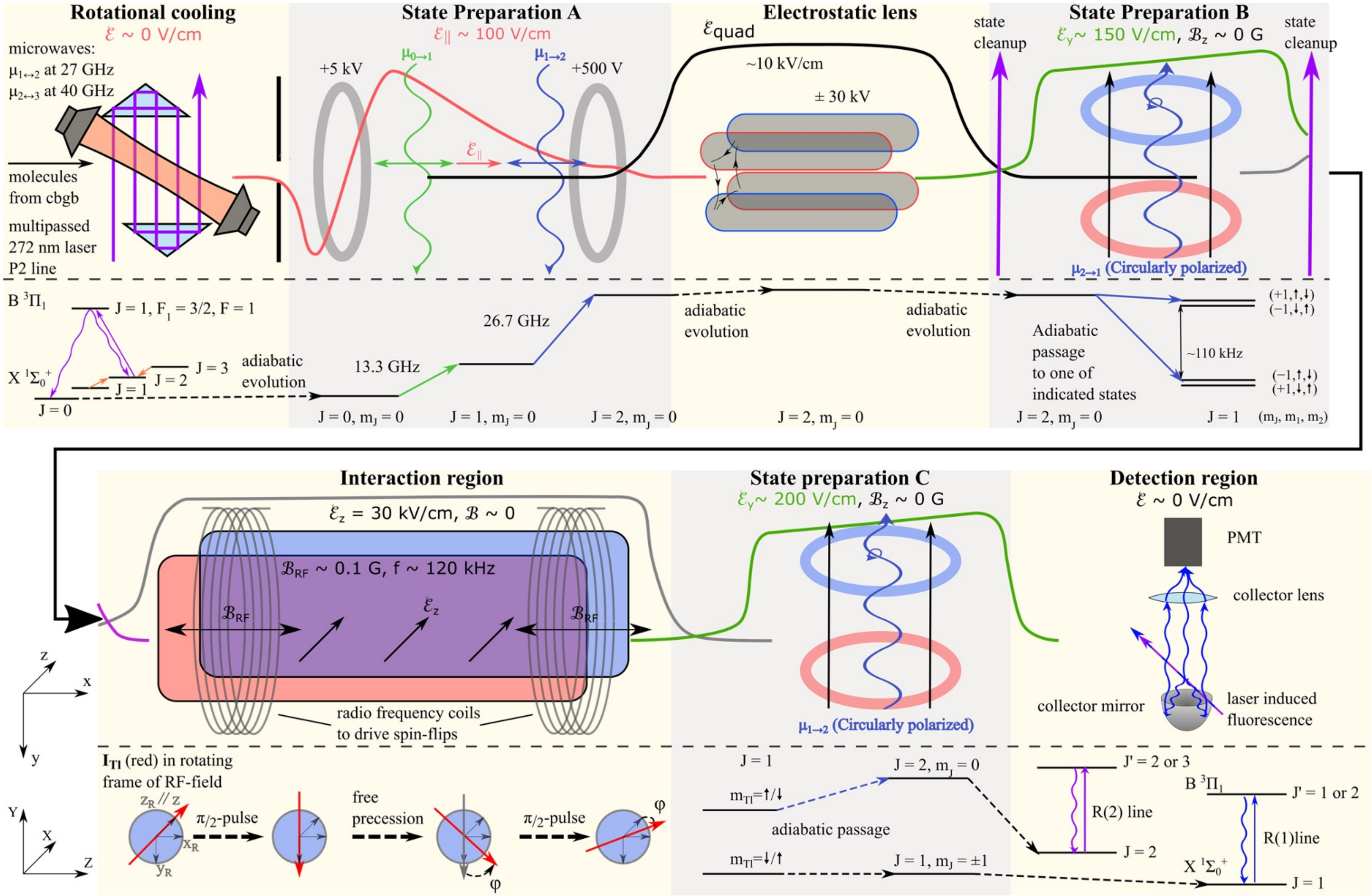
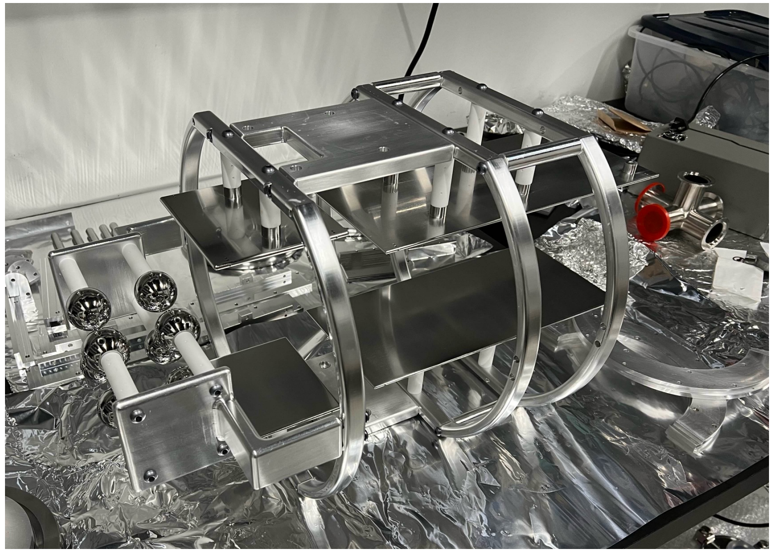
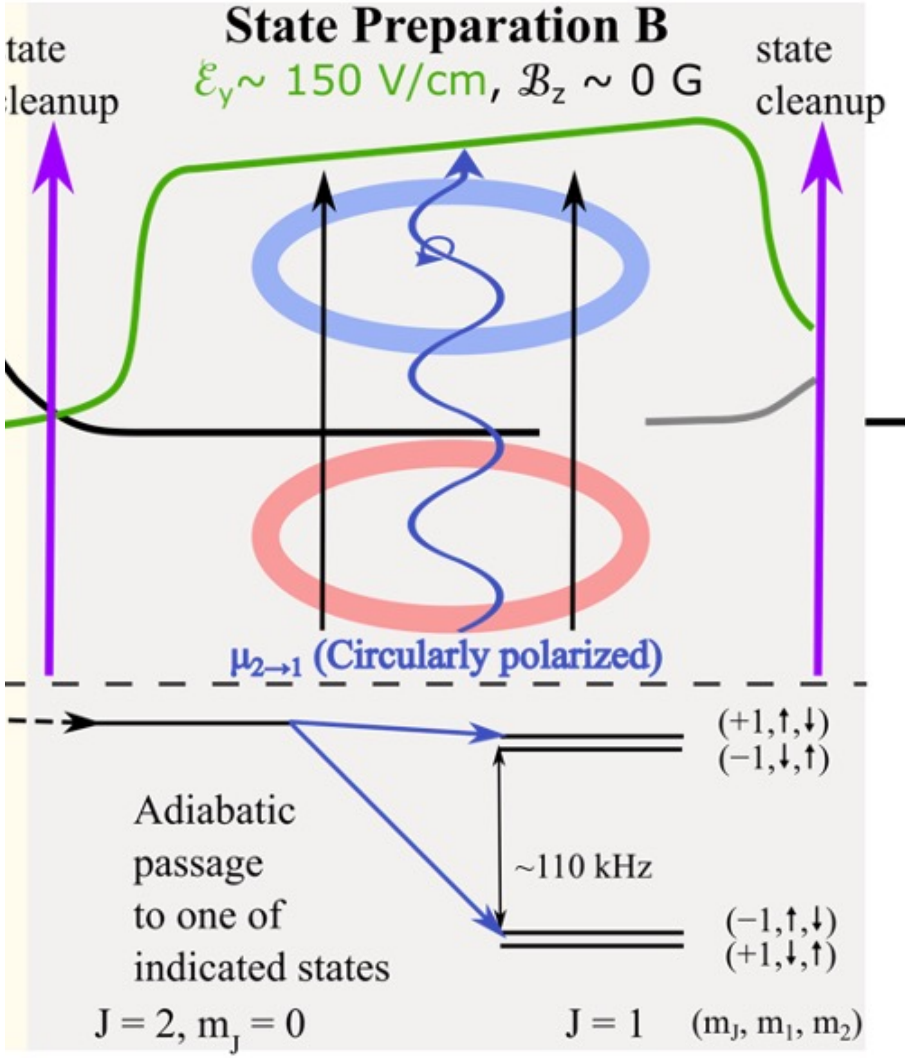
Experiment Overview



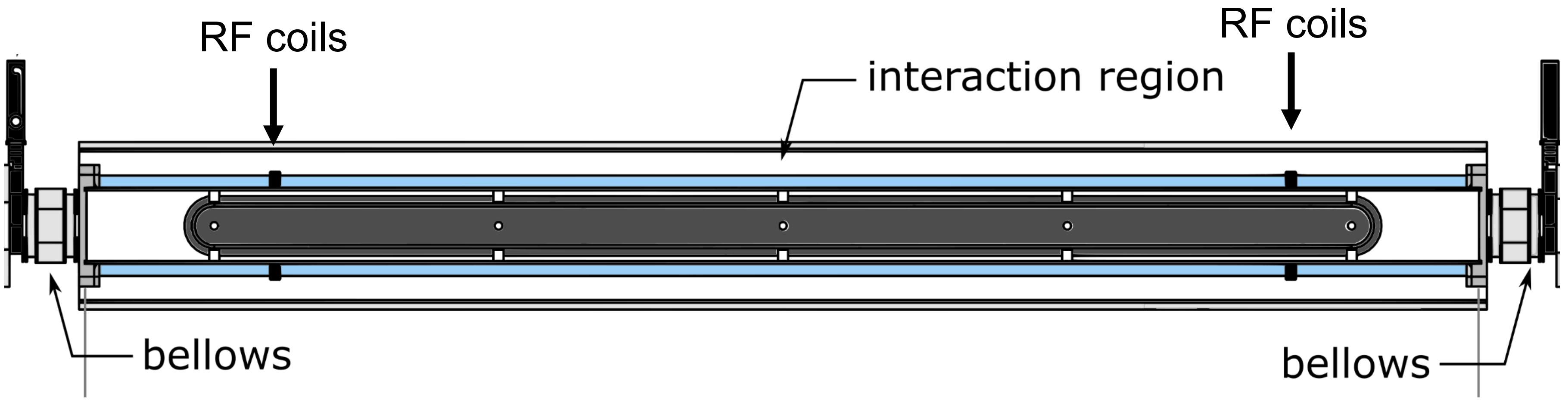
State Preparation B and State Cleanup

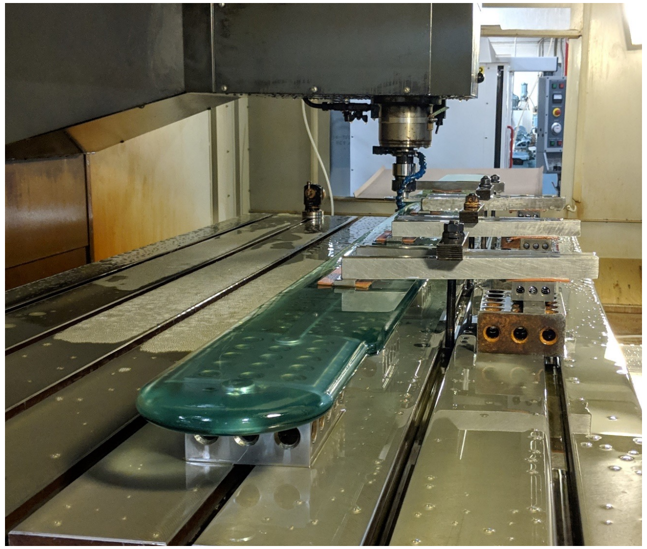
- Transfer from lens state to SM measurement state
  - $|J = 2, m_J = 0\rangle \rightarrow |J = 1, m_J = \pm 1\rangle$
- Microwave driven adiabatic passage
  - Analogous to SPA, except circular polarization
- Lasers remove remaining population in $J = 1$ & $J = 2$
  - Reduces potential systematics

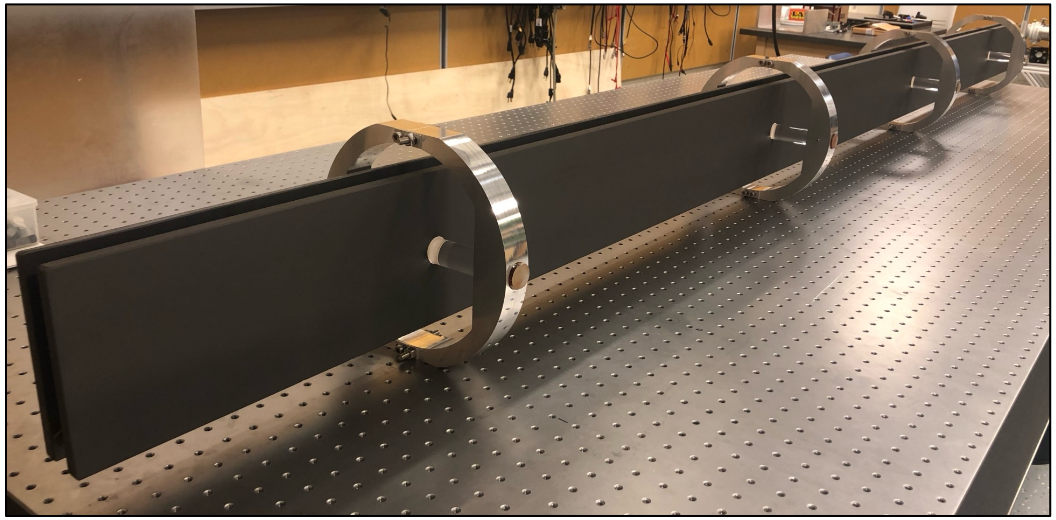




Interaction Region



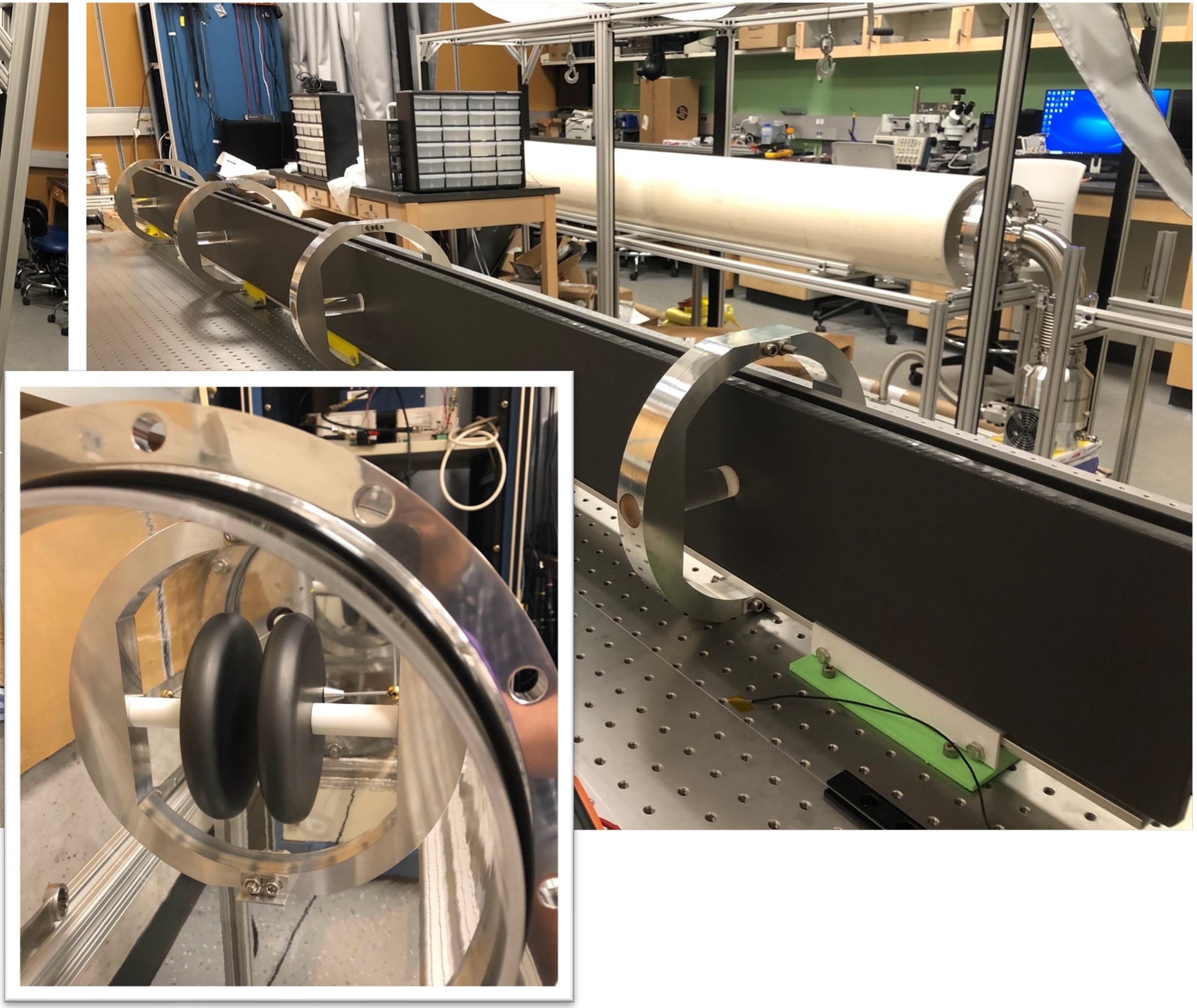




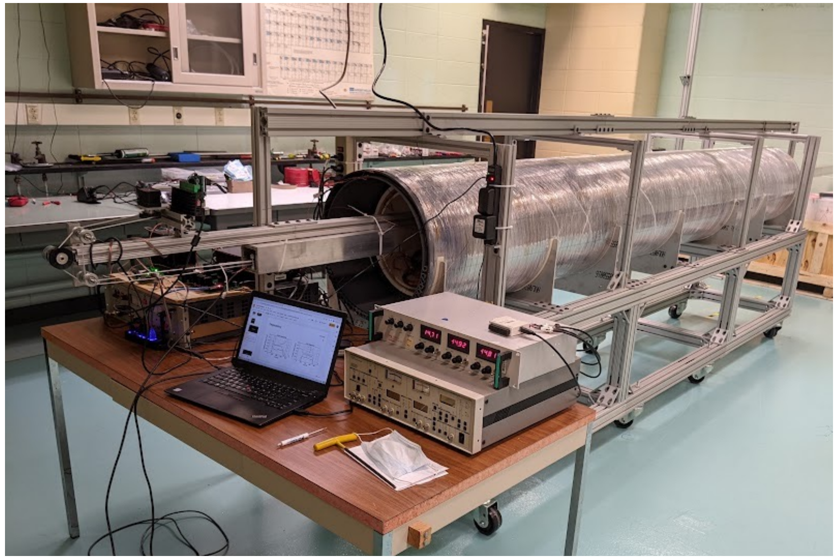
- Try to minimize magnetic Johnson noise
  - 3.0 m long glass machined with Rogowski profile
  - Coated with conductive graphite coating (Aquadag)
  - 2 cm apart, +30 kV or -30 kV on each plate
  - 3.5 m long, 25 cm diam. quartz vacuum chamber
- Ramsey SOF sequence

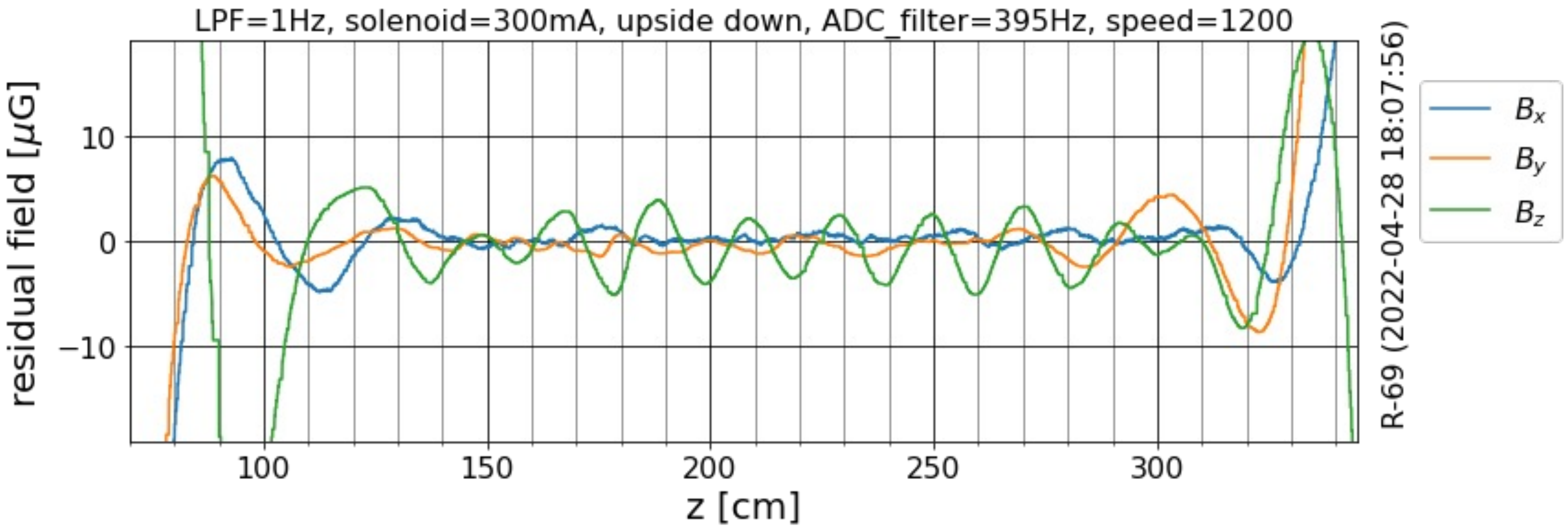
Interaction Region



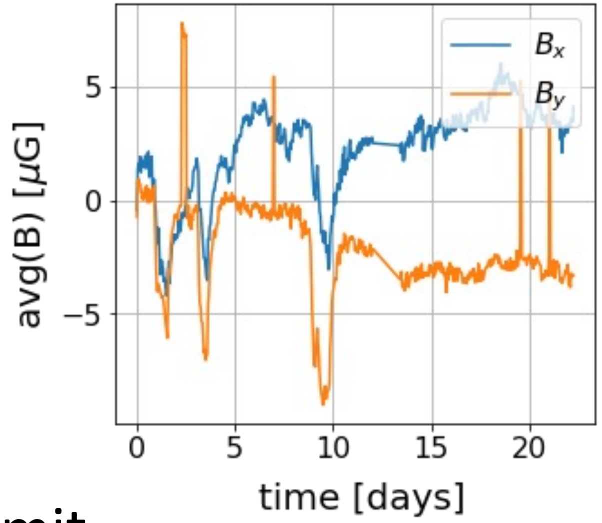


Magnetic Shielding of Interaction Region

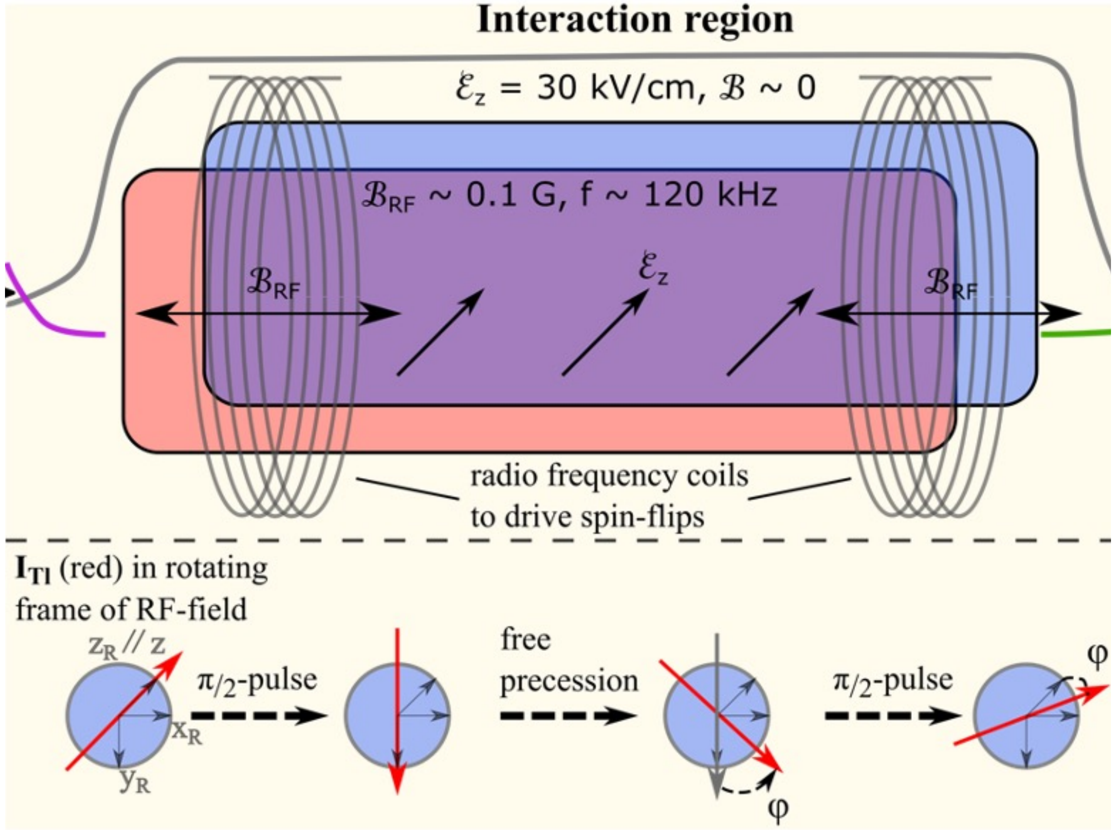




- Spin-rotation generates internal magnetic field
  - Must shield external magnetic field
- 4 shielding layers: 3 layers Metglass, 1 layer mu-metal
- 80 shim coils
- Achieved <10 μG over the interaction region
  - Required to keep anticipated systematics below the statistical limit



Ramsey Separated Oscillatory Fields



(1) Create Tl nucleus ↑/↓ superposition with RF field $\left(\frac{\pi}{2}\text{pulse}\right)$

(2) Free spin precession for time T, accumulate phase difference:

$$\phi \simeq \left(-\mu_{Tl} B_{int}\,\mathrm{sgn}(\mathrm{m_J}) + C_s m_{I_z} + 2W_s SP\mathrm{sgn}(E_{MI})\right) T/\hbar$$

(3) After second RF $\pi/2$ pulse, probability of ↑→↓ :

$$P_{\uparrow\to\downarrow} = \sin^2\left(\frac{1}{2}\Omega_{RF}\tau\right)\cos^2\frac{1}{2}(\phi_{CPV} + \phi_{SOF})$$

where $\phi_{CPV} = 2W_s SP\mathrm{sgn}(\mathrm{E_{MI}})\mathrm{T}/\hbar = 2\Delta_{CPV}T/\hbar$ and $\phi_{SOF} = \pm\pi/2$

(4) Signal: $S_{SOF} = P_{\uparrow\to\downarrow}(\phi_{SOF} = +\pi/2) - P_{\uparrow\to\downarrow}(\phi_{SOF} = -\pi/2)$

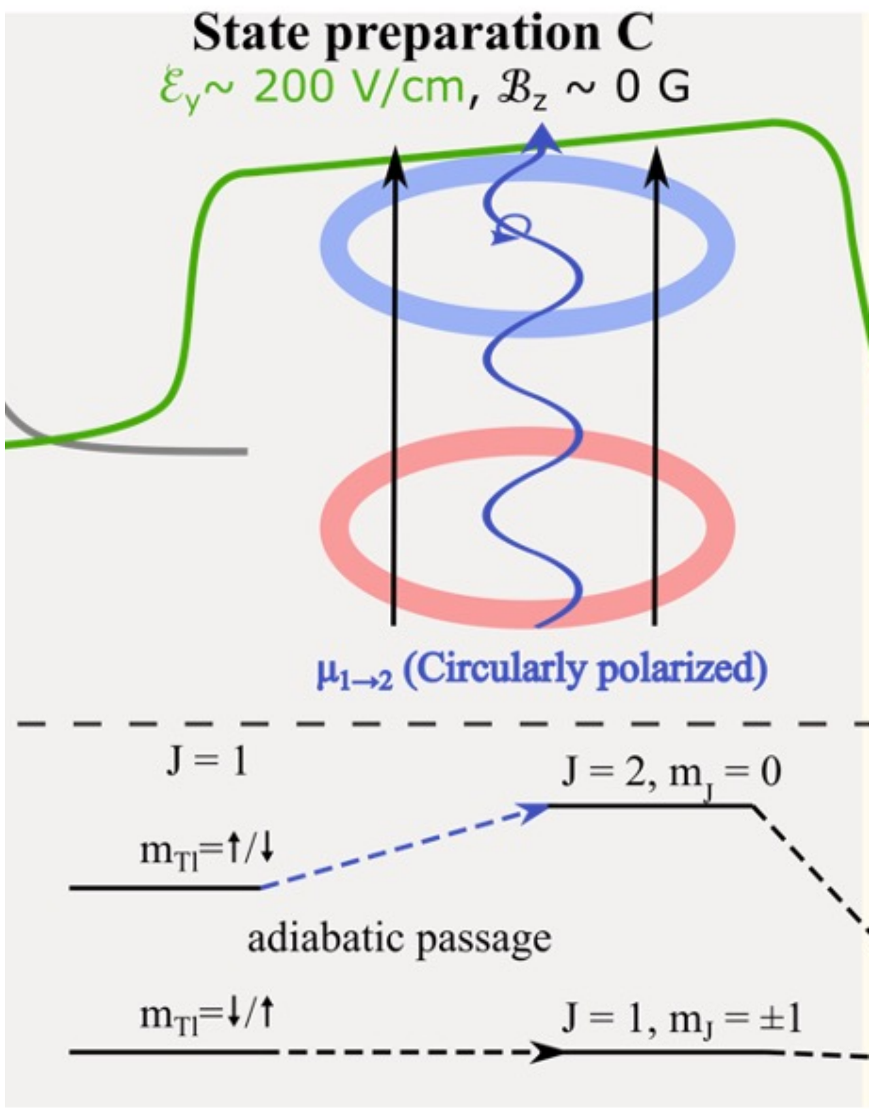
State Preparation C: Detection of Tl Nuclear Spin State

- After interaction region molecules in:

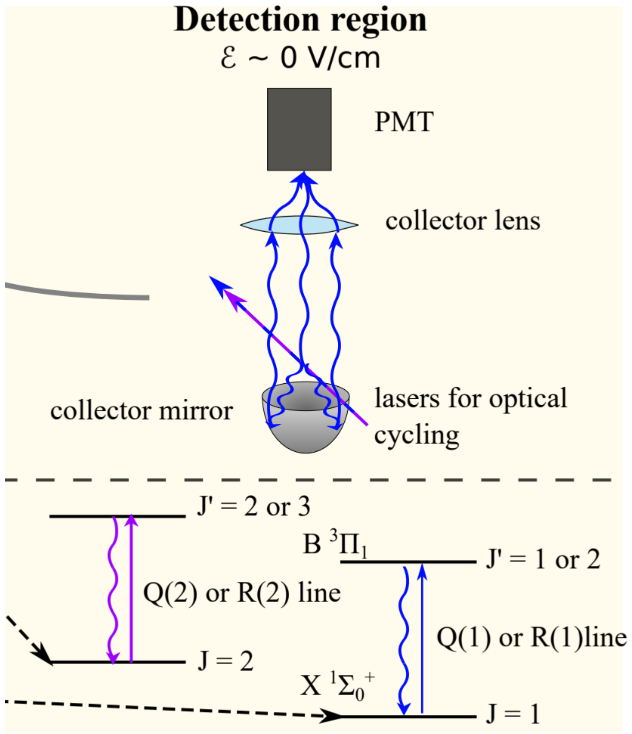
$\left|J = 1, m_J, m_{I1} = +1/2, m_2\right\rangle$ and $\left|J = 1, m_J, m_{I1} = -1/2, m_2\right\rangle$

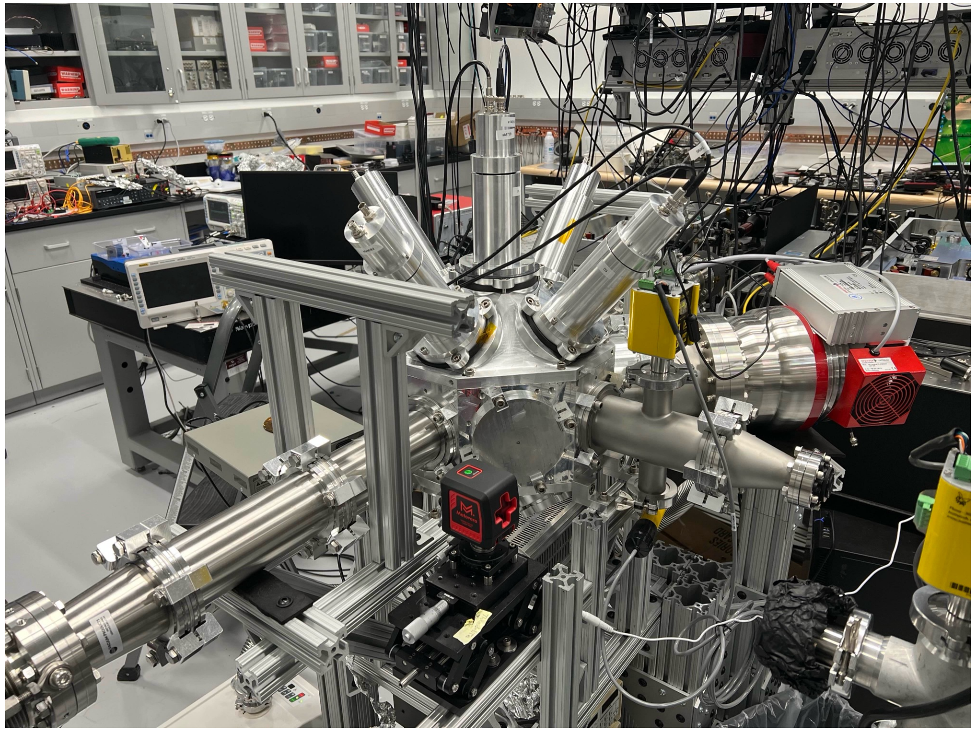
- Thallium ↑ & ↓ population in different hyperfine levels

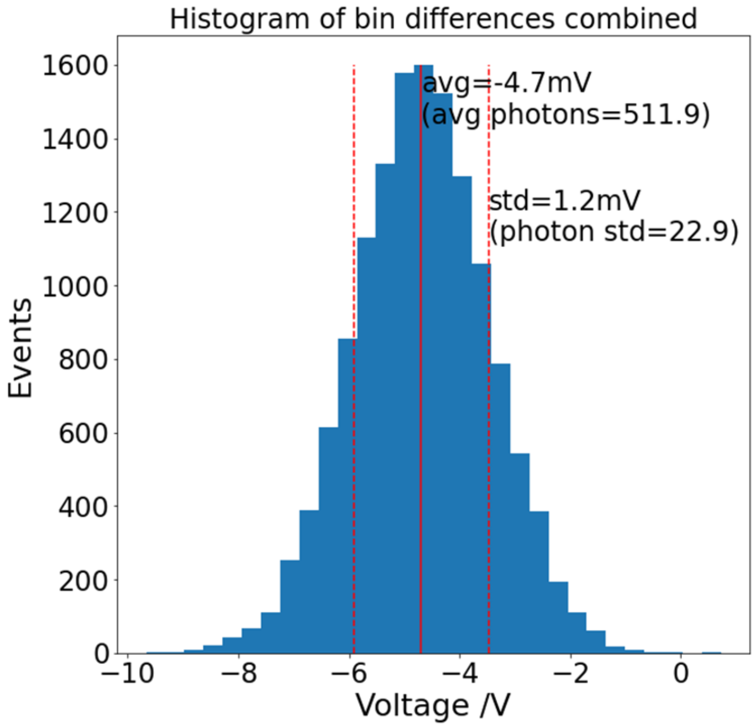
  < 200 kHz apart
- HF splitting ≤ natural linewidth Γ = 1.6 MHz
  - Can't resolve ↑ & ↓ with optical readout
- Move ↑ or ↓ to different rotational states
- Microwave driven adiabatic passage like State Prep. B



Detection of Tl Nuclear Spin State

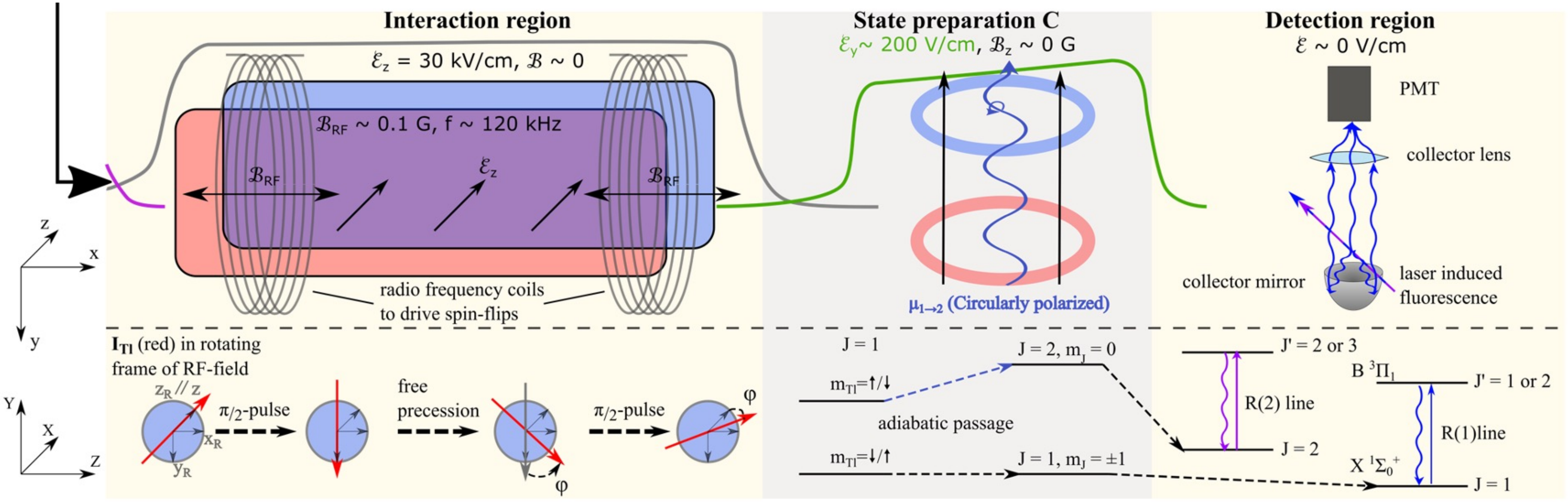






- Laser induced fluorescence to read out molecules
- Use two UV lasers at 271.8 nm (offset by relevant rotational splittings)
- Excite and detect at same wavelength so must reduce scattered light
- R(1) and R(2) transition to read out ↑ & ↓ populations
  - Rapidly switch between transitions in a single pulse using AOMs

Detection of Phase Shift sensitive to CP-Violation



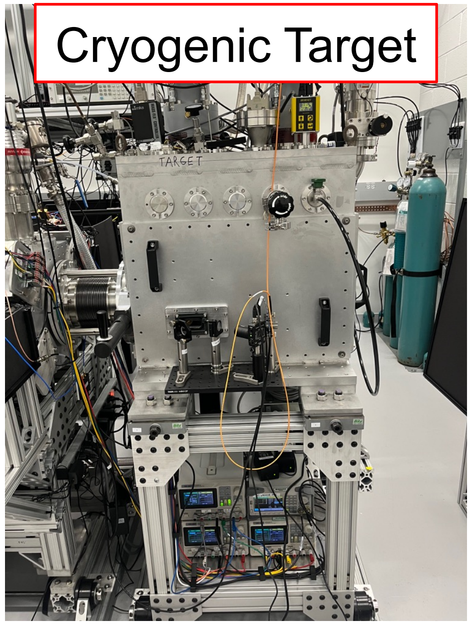
- Construct asymmetry signal

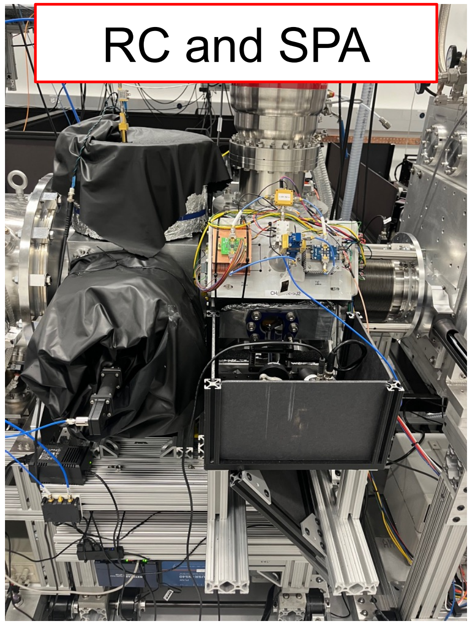
$$A = \frac{S_\uparrow - S_\downarrow}{S_\uparrow + S_\downarrow}$$

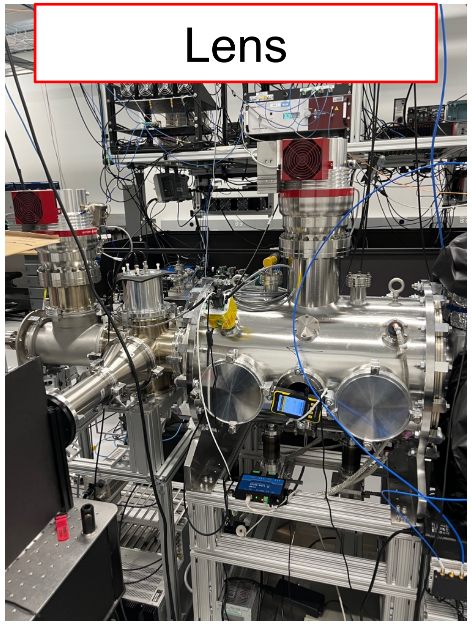
- With SOF on resonance

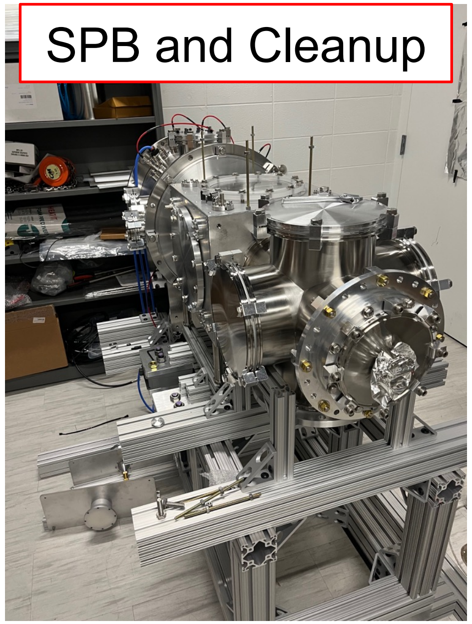
$$A \simeq 1 - 2\sin^2 \Omega_{RF}\tau \cos^2 \frac{1}{2}(\phi_{CPV} + \phi_{SOF})$$

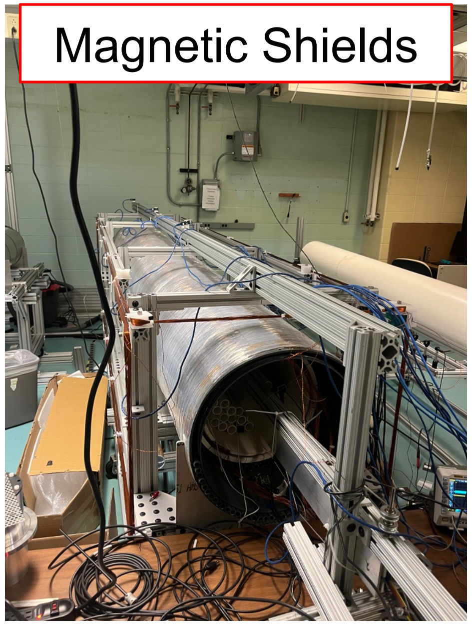
- With $\phi_{SOF} = \pm\pi/2$ and $\Omega_{RF}\tau = \pi/2$: $\quad A \simeq sgn(\phi_{SOF})\sin\phi_{CPV} \sim \pm\phi_{CPV}$

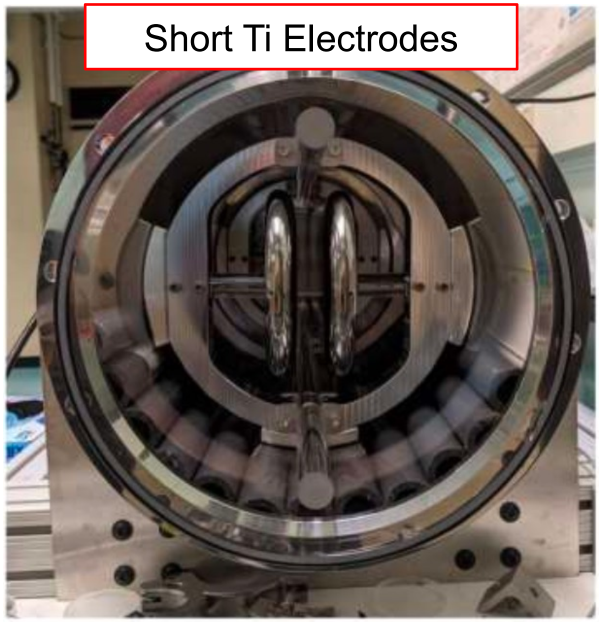

Cryogenic Target


RC and SPA

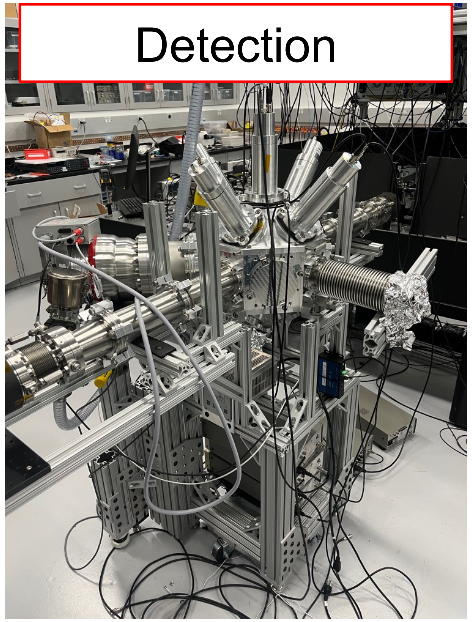

Lens


SPB and Cleanup


Magnetic Shields


Short Ti Electrodes


Long Electrode Prep


Detection

^{205}TlF ground state energy levels at high electric field

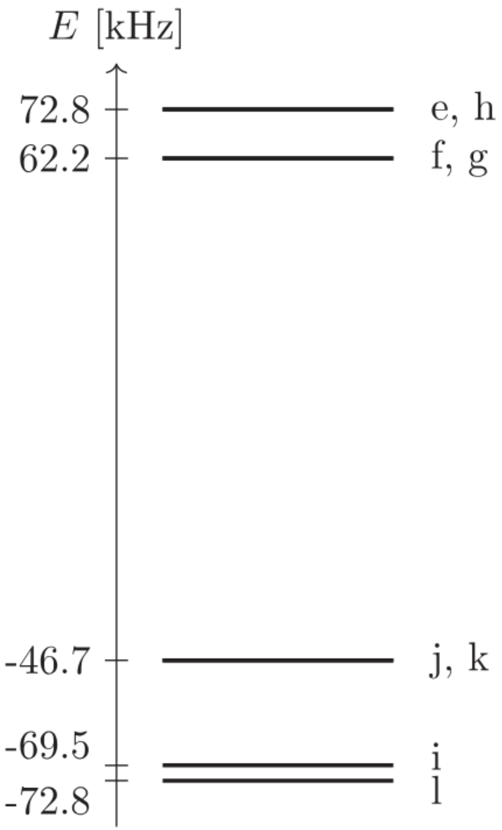


Table 3. All non-degenerate pairs of $|m_J| = 1$ states in the $\widetilde{J} = 1$ manifold at $\mathcal{E}_{MI} = 30$ kV cm^{-1} that do not involve the states $|i\rangle$ and $|l\rangle$ or a flip of m_J. The quantum numbers given are that of the largest decoupled-basis component. State labels l are as in [32] and figure 11. The quantities $df_0/d\mathcal{B}_z$ and $df_0/d\mathcal{E}_z$ give the slope of the resonance frequency with respect to the external magnetic field and electric field, respectively. Shift $\mathcal{B}_{mot}$ indicates the resonance frequency shift, due to the motional field that accompanies $\mathcal{E}$ reversal, with respect to a stray field component $\mathcal{B}_y$. S denotes the sensitivity to the NSM relative to the maximum possible value; it is given by $|\langle I_{1,z}\rangle_1 - \langle I_{1,z}\rangle_2|$ for the transition between states 1 and 2. f_0 indicates the transition frequency between the two states. $|\langle\cdot|\mathcal{H}_Z|\cdot\rangle|$ indicates the magnitude of the transition dipole moment between states 1 and 2. All shifts are calculated from diagonalization of the ground-state Hamiltonian (equation (7)).

	State 1				State 2				f_0	$df_0/d\mathcal{B}_z$	$df_0/d\mathcal{E}_z$	Shift $\mathcal{B}_{mot}$	S	$\lvert\langle\cdot\rvert\mathcal{H}_Z\lvert\cdot\rangle\rvert$ [kHz]	
What flips?	l	m_J	m_{I_1}	m_{I_2}	l	m_J	m_{I_1}	m_{I_2}	kHz	[mHz/μG]	[mHz/(V/cm)]	[mHz/μG]	-	x, y	z
m_{I_1}	e	−	−	−	j	−	+	−	119.52	+2.49	−31.50	$+4.66 \times 10^{-5}$	0.95	1.33	0.00
	h	+	+	+	k	+	−	+		−2.49		$+5.22 \times 10^{-5}$			
m_{I_1}, m_{I_2}	f	+	+	−	k	+	−	+	108.92	+1.52	−3.57	-1.17×10^{-4}	0.99	0.00	0.09
	g	−	−	+	j	−	+	−		−1.52		-1.23×10^{-4}			
m_{I_2}	e	−	−	−	g	−	−	+	10.59	+4.00	−27.93	$+1.69 \times 10^{-4}$	0.04	1.88	0.00
	h	+	+	+	f	+	+	−		−4.00					

- EDM search uses e+j and/or h+k states in which Tl spin flips
- (e+j) and (h+k) have opposite signs of internal B field
- Need molecules in J=1, high electric field

Projected sensitivity

	^{205}TlF (projected)	^{199}Hg	^{129}Xe	^{225}Ra	^{171}Yb	Neutron
Energy shift $\delta\nu$ (Hz)	1×10^{-7}	7×10^{-12}	5×10^{-10}	4×10^{-3}	5×10^{-7}	3×10^{-8}
δS (e fm^3) or δd (e cm)	3×10^{-14}	1×10^{-13}	2×10^{-10}	3×10^{-6}	8×10^{-10}	1×10^{-26}
$\delta\bar{\theta}_{QCD}$	1×10^{-12}	6×10^{-11}	3×10^{-8}	3×10^{-6}	–	8×10^{-11}
quark c-EDMs $\alpha\tilde{d}_u + \beta\tilde{d}_d$	$0.8\tilde{d}_d + 0.6\tilde{d}_u$	$\tilde{d}_d$	$\tilde{d}_d$	$0.7\tilde{d}_u - 0.7\tilde{d}_d$	–	–
$\delta(\alpha\tilde{d}_u + \beta\tilde{d}_d)$ (cm^{-1})	3×10^{-28}	2×10^{-27}	3×10^{-24}	3×10^{-23}	–	1×10^{-26}
NSM or EDM in terms of d_p or d_n	d_p	$d_n + 0.1d_p$	$d_n + 0.2d_p$	–	–	d_n
δ in nucleon EDM (e cm)	8×10^{-27}	1×10^{-26}	3×10^{-23}	–	–	1×10^{-26}

values indicated with – not found in literature

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Current Status

- State Preparation B and C nearing completion
- Small (1m) interaction region with Ti electrodes ready to go
- Alignment of beamline is challenging!
- Coating and alignment of 3.0 m long electrodes over next few months

- First science data in 2026 ?