

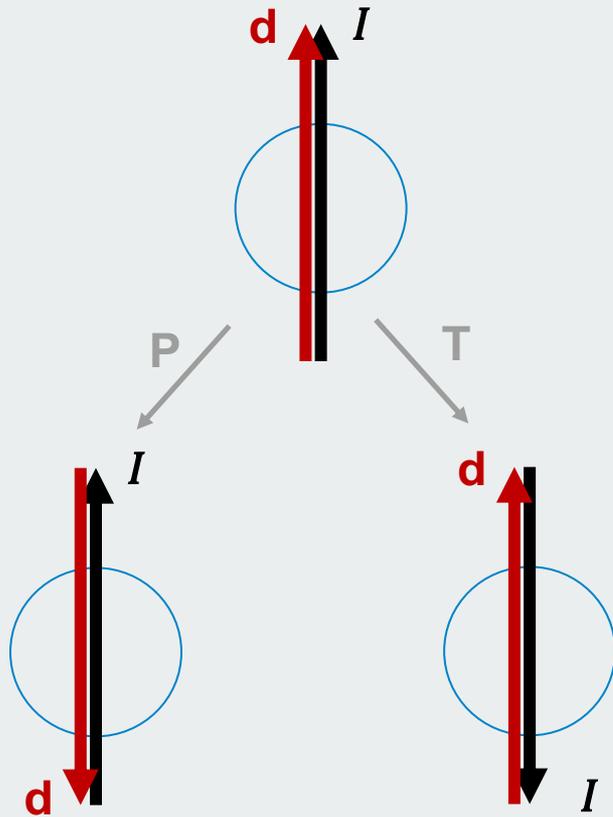
Measuring the magnetic quadrupole moment of heavy nuclei with diatomic molecules

Chris Ho, SSP 2022

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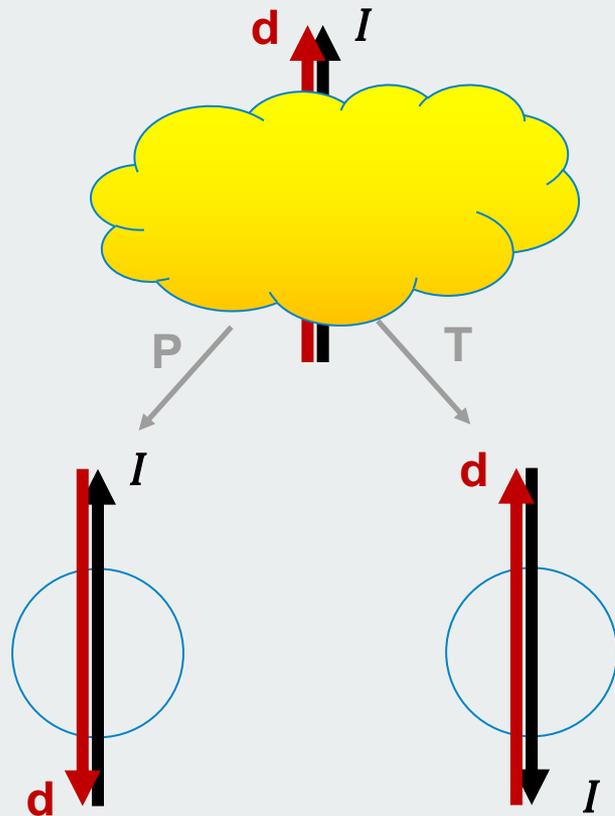
MQMs.. what the heck?

Nuclear EDM ($I \geq 1/2$)



MQMs.. what the heck?

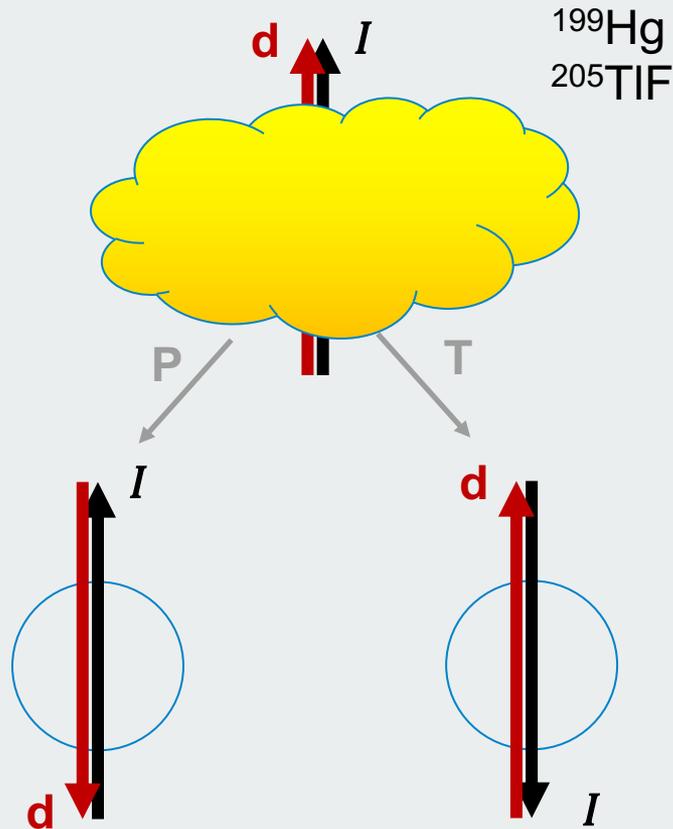
Nuclear EDM ($I \geq 1/2$)



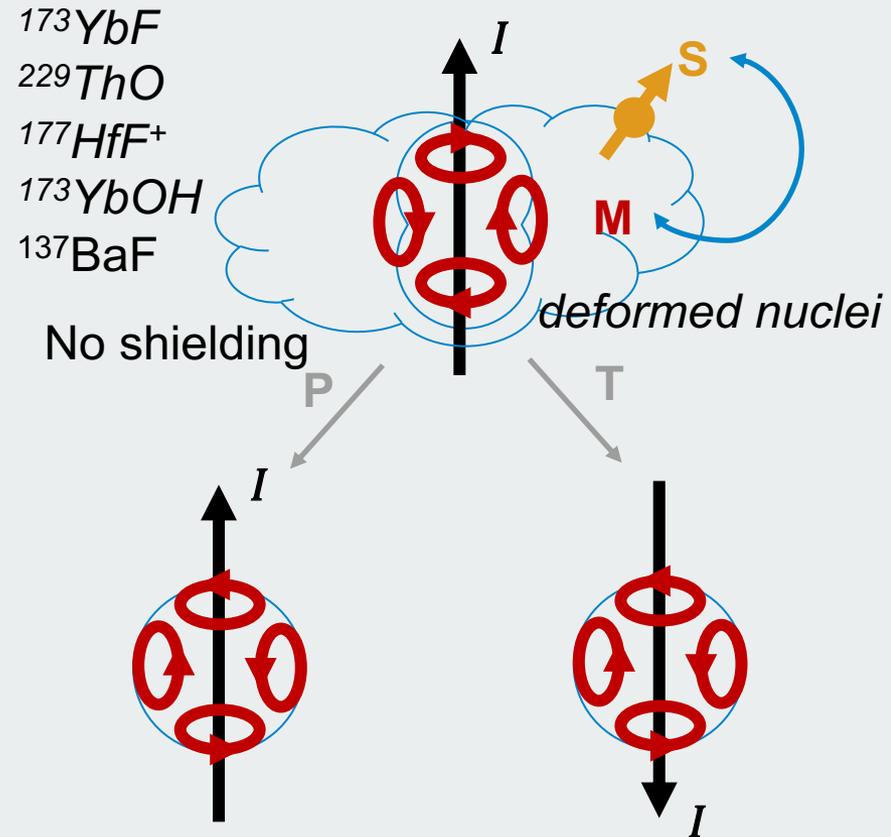
Electrons shield nuclear
EDM almost perfectly;
remaining moment is called
Schiff moment

MQMs.. what the heck?

Nuclear EDM Schiff moment ($I \geq 1/2$)

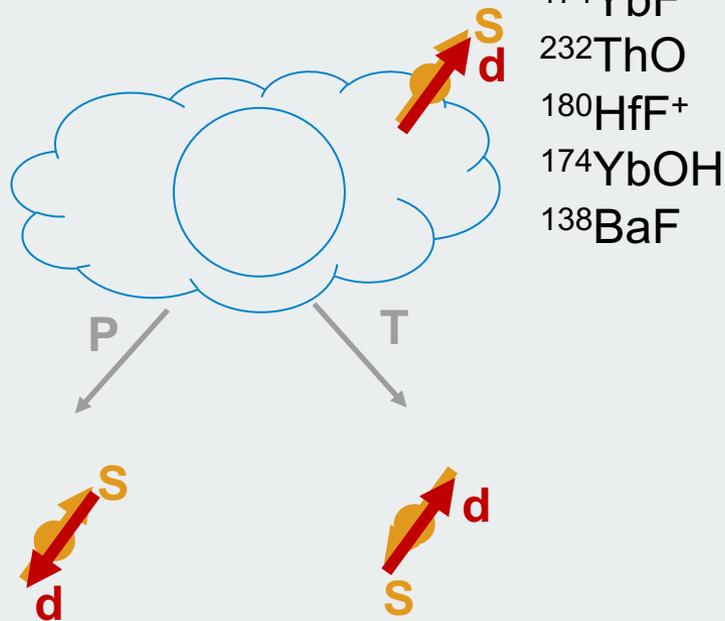


Nuclear MQM ($I \geq 1$)

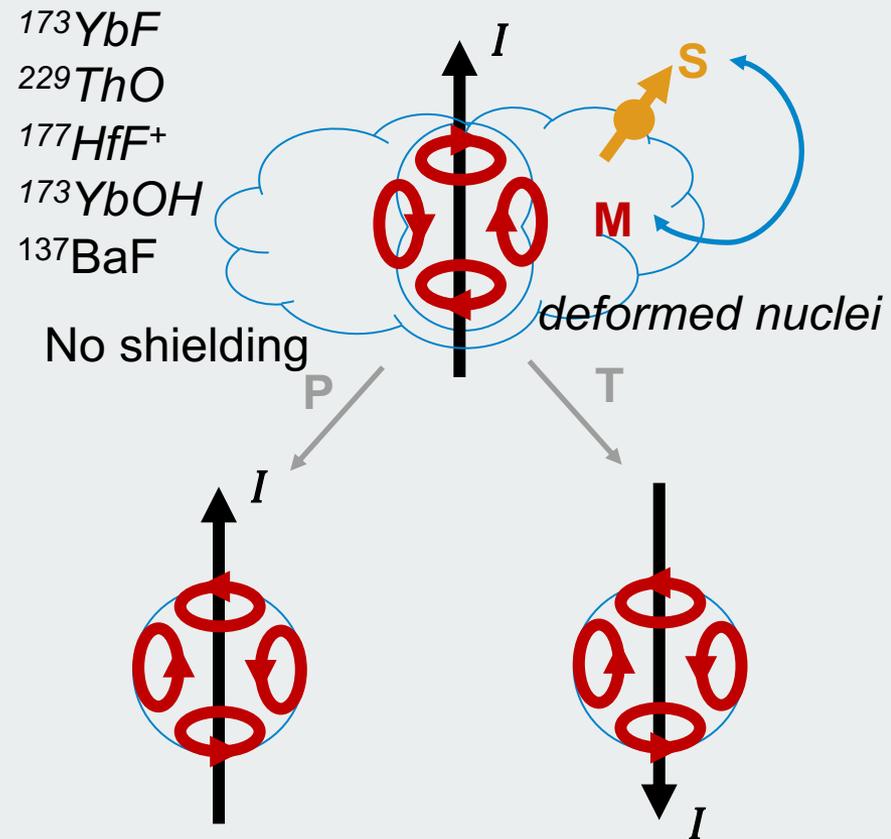


MQMs.. what the heck?

Electron EDM



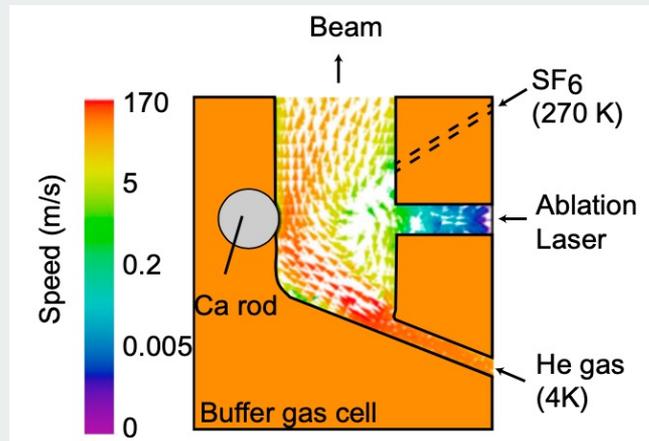
Nuclear MQM ($I \geq 1$)



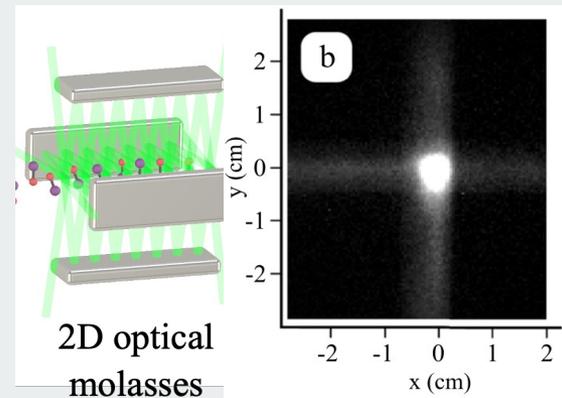
Motivation - summary

- Nuclear MQMs are P,T-violating moments sensitive to hadronic CP-violating parameters
 - Measuring such moments can help discover or constrain new CP-violating physics in theories beyond the Standard Model
 - Only previous measurement of a nuclear MQM was in ^{133}Cs :
 - Murthy *et al.*, PRL **63** (9), 965 (1989)
- Best measurements of these parameters set by ^{199}Hg and neutron EDM
 - Graner *et al.*, PRL **116**, 161601 (2016)
 - Abel *et al.*, PRL **124**, 081803 (2020)
- MQMs of heavy, deformed nuclei suffer no electron shielding and have collective enhancement from single nucleon MQM
- Diatomic molecules provide further enhancement of MQM interaction due to relativistic effects and ease of full polarization
- Suitable molecules (though of different isotopes) already used in electron EDM experiments

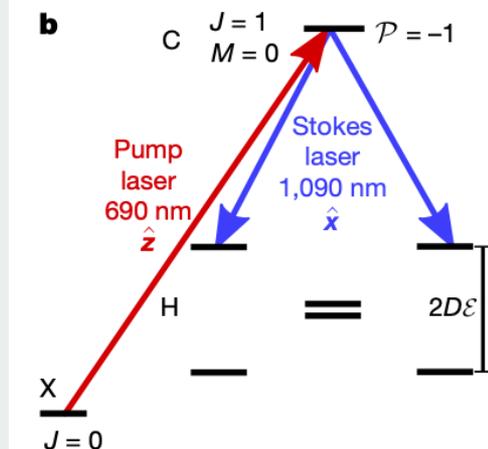
Experimental advances with diatomic molecules



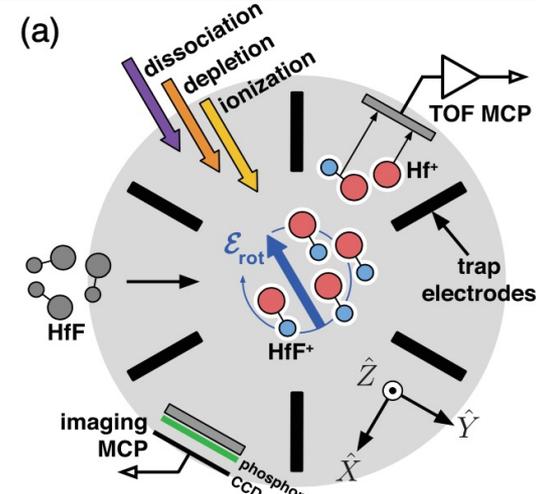
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Alauze *et al.*, *Q. Sci. Technol.* **6**, 044005 (2021)



ACME Collaboration, *Nature* **562**, 355 (2018)



Cairncross *et al.*, *PRL* **119**, 153001 (2017)

The effective MQM Hamiltonian

- The classic paper by Sushkov, Flambaum, Khriplovich (JETP **60** (5), 873 (1984)) gives

$$\mathcal{H}_M = -\frac{W_M M}{2I(2I-1)} \mathbf{S} \cdot \hat{\mathbf{T}} \cdot \mathbf{n}$$
$$T_{i,k} = I_i I_k + I_k I_i - \frac{2}{3} \delta_{i,k} I(I+1)$$

W_M : interaction parameter (relativistic enhancement)

M : nuclear MQM (collective enhancement)

\mathbf{n} : unit vector along internuclear axis

- We can rewrite this (in spherical tensor notation) as

$$\mathcal{H}_M = \frac{W_M M}{2I(2I-1)} \sqrt{\frac{20}{3}} T^{(1)}(\mathbf{S}, T^{(2)}(\mathbf{I}, \mathbf{I})) \cdot \mathbf{n} \equiv \mathbf{M} \cdot \mathbf{n}$$

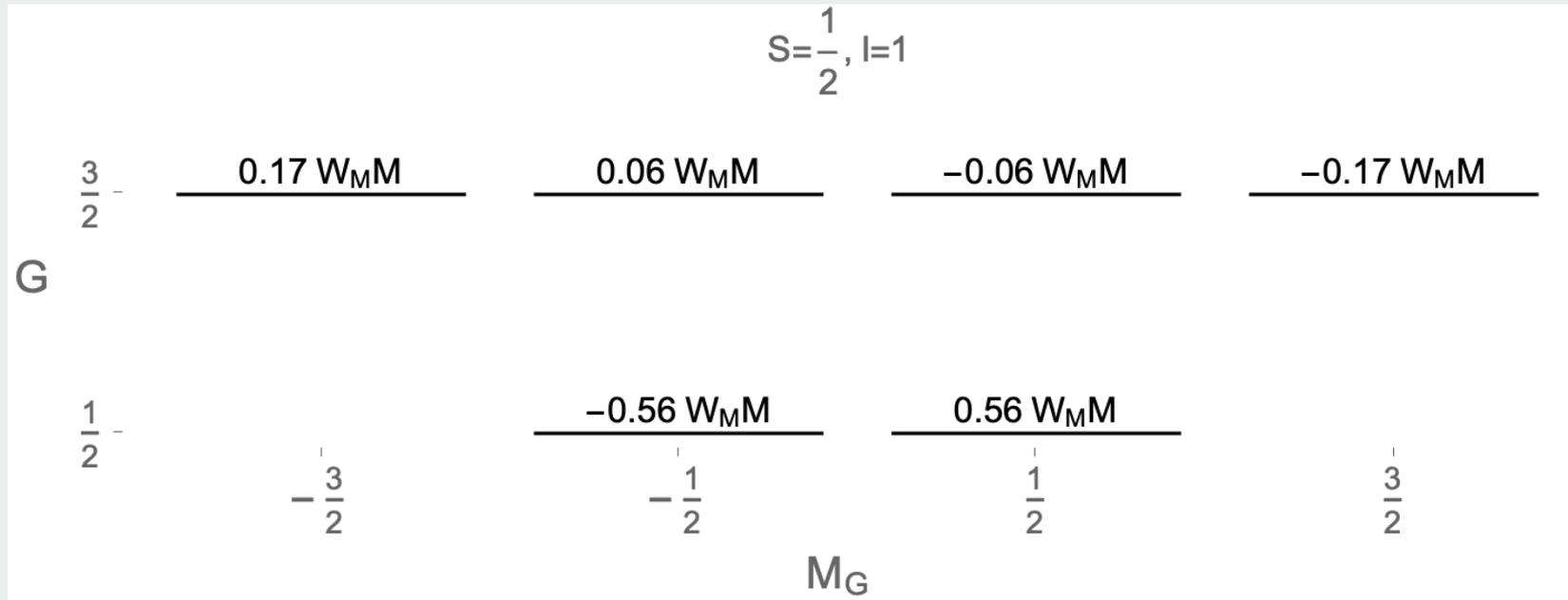
MQM matrix elements in diatomic molecules

- Consider a diatomic molecule with electron spin \mathbf{S} , nuclear spin \mathbf{I} , total spin $\mathbf{G} = \mathbf{S} + \mathbf{I}$ and rotational angular momentum \mathbf{N}
- MQM energy shift is given by

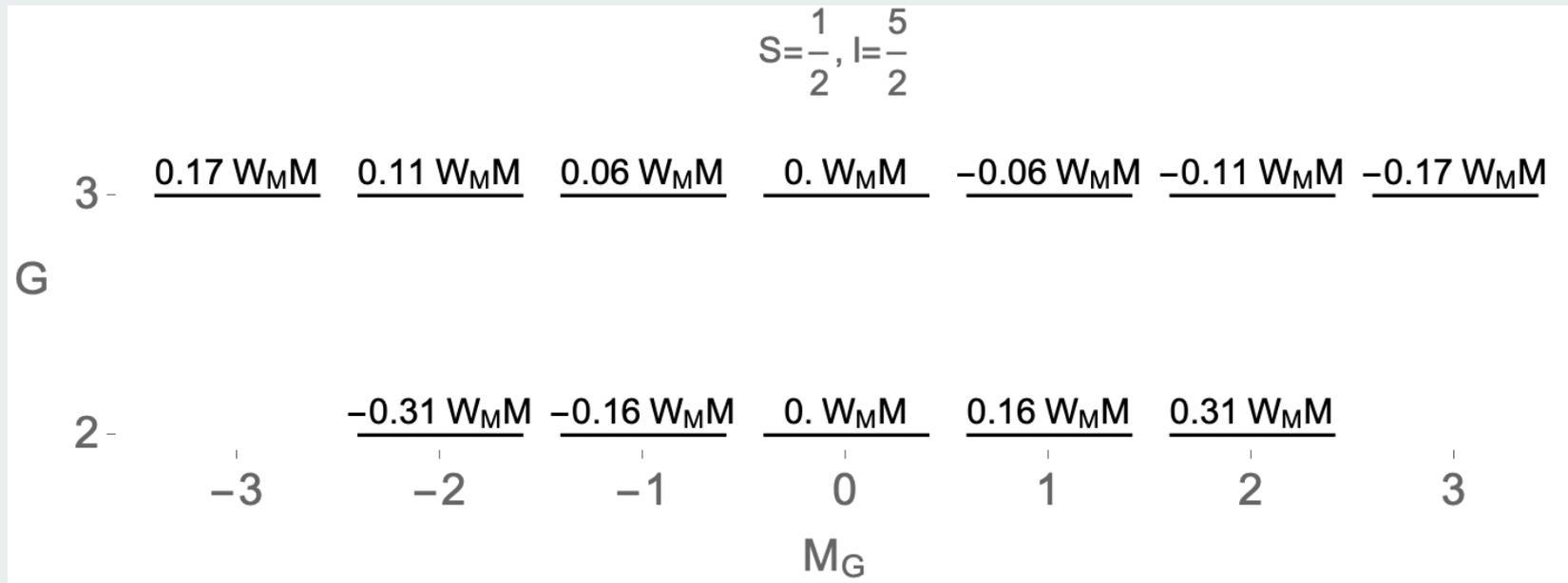
$$\begin{aligned}\Delta_M &= \langle (S, I)G, M_G; N, M_N | \mathcal{H}_M | (S, I)G, M_G; N', M_N \rangle \\ &= \langle (S, I)G, M_G | \mathbf{M} | (S, I)G, M_G \rangle \langle N, M_N | \mathbf{n} | N', M_N \rangle \\ &= \langle \mathcal{H}_M \rangle_{\text{mol}} \mathcal{P}\end{aligned}$$

- The measured MQM energy shift in the lab is the product of the energy shift in the molecule-fixed frame and a polarization factor
- The state where \mathbf{S} and \mathbf{I} are aligned (i.e. where G is largest) does not give the largest energy shift

Example I: $\langle H_M \rangle_{\text{mol}}$ for $S = 1/2, I = 1$



Example II: $\langle H_M \rangle_{\text{mol}}$ for $S = 1/2, I = 5/2$ (^{173}YbF)

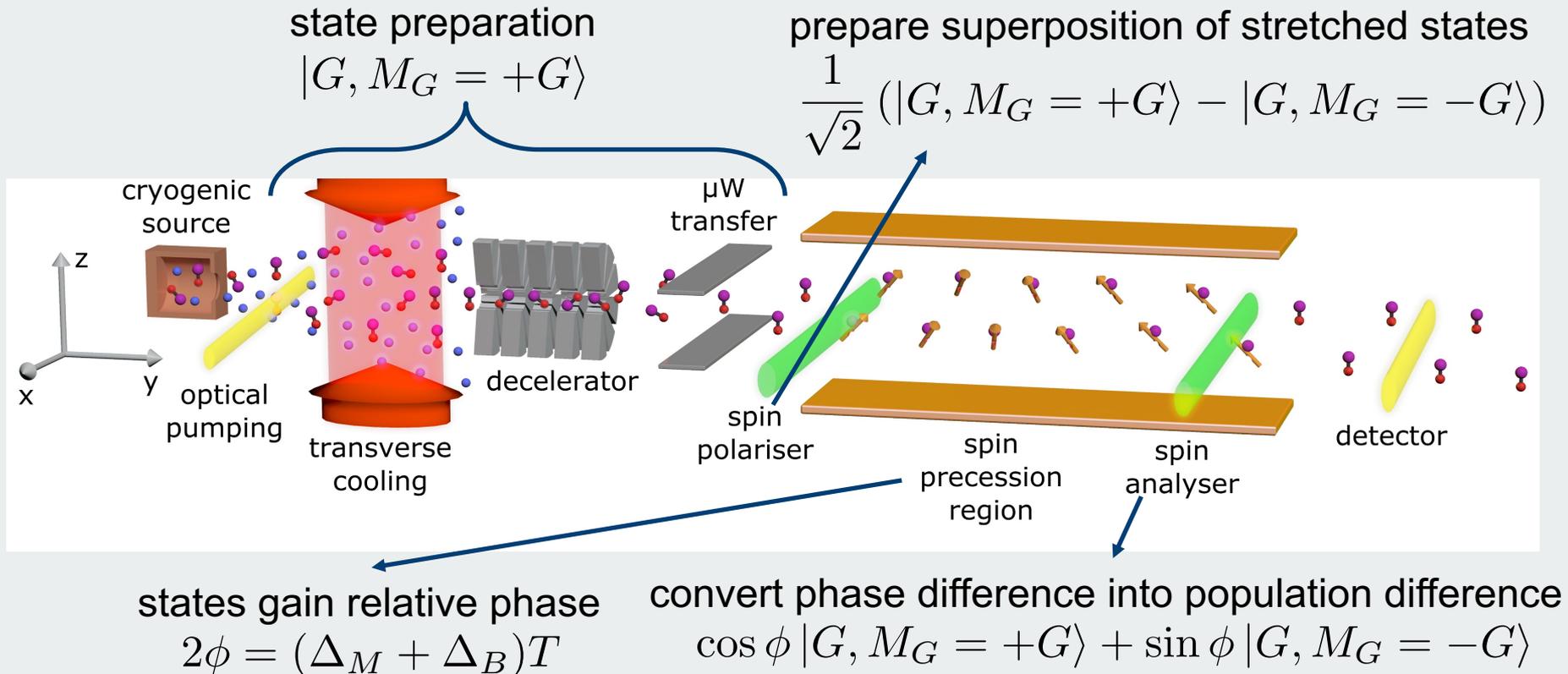


Example III: $\langle H_M \rangle_{\text{mol}}$ for $S = 1, I = 5/2$ (^{229}ThO)

		$S=1, I=\frac{5}{2}$							
	$\frac{7}{2}$	<u>$0.33 W_M M$</u>	<u>$0.24 W_M M$</u>	<u>$0.14 W_M M$</u>	<u>$0.05 W_M M$</u>	<u>$-0.05 W_M M$</u>	<u>$-0.14 W_M M$</u>	<u>$-0.24 W_M M$</u>	<u>$-0.33 W_M M$</u>
G	$\frac{5}{2}$		<u>$-0.3 W_M M$</u>	<u>$-0.18 W_M M$</u>	<u>$-0.06 W_M M$</u>	<u>$0.06 W_M M$</u>	<u>$0.18 W_M M$</u>	<u>$0.3 W_M M$</u>	
	$\frac{3}{2}$			<u>$-0.56 W_M M$</u>	<u>$-0.19 W_M M$</u>	<u>$0.19 W_M M$</u>	<u>$0.56 W_M M$</u>		
		$-\frac{7}{2}$	$-\frac{5}{2}$	$-\frac{3}{2}$	$-\frac{1}{2}$	$\frac{1}{2}$	$\frac{3}{2}$	$\frac{5}{2}$	$\frac{7}{2}$
		M_G							

How to measure the MQM energy shift?

- Use a spin interferometer – just like measuring the electron EDM



Preparation of stretched state superposition

- The maximum sensitivity to the MQM energy shift is obtained when the stretched superposition state is prepared:

$$\frac{1}{\sqrt{2}} (|G, M_G = +G\rangle - |G, M_G = -G\rangle)$$

- Not trivial for states with angular momentum greater than 1 as the state can't be prepared as a pure M state in another Cartesian basis, e.g.

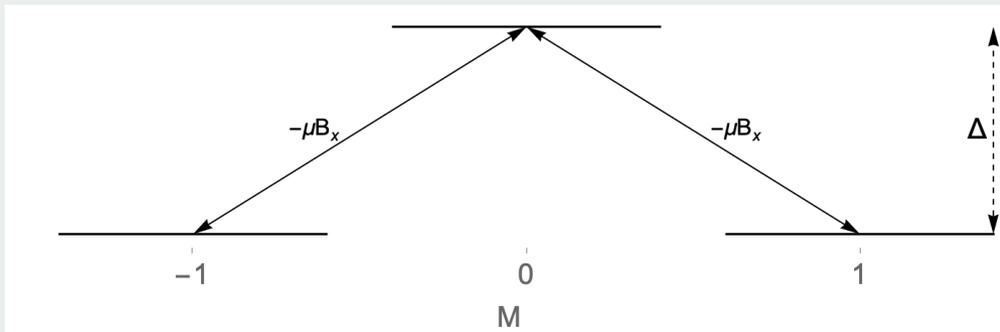
$$|M = 0\rangle_x = \frac{1}{\sqrt{2}} (|M = 1\rangle_z - |M = -1\rangle_z)$$

$$|M = -\frac{1}{2}\rangle_x = \frac{1}{\sqrt{2}} \left(|M = \frac{1}{2}\rangle_z + |M = -\frac{1}{2}\rangle_z \right)$$

- Possibly can use a series of coherent rf pulses to drive successive transitions, e.g. starting from M=0, but does not work for half-integer M

Preparation of stretched state superposition

- A general method: apply a magnetic field perpendicular to a strong static electric field (which generates a tensor Stark shift Δ)



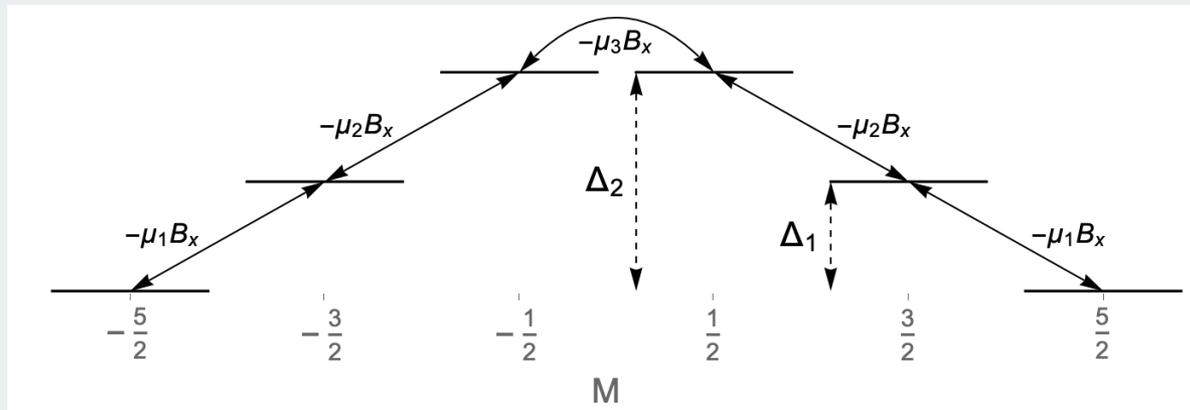
$$\mathcal{H} = \begin{pmatrix} 0 & -\frac{\mu B_x}{\sqrt{2}} & 0 \\ -\frac{\mu B_x}{\sqrt{2}} & \Delta & -\frac{\mu B_x}{\sqrt{2}} \\ 0 & -\frac{\mu B_x}{\sqrt{2}} & 0 \end{pmatrix}$$

- The same Hamiltonian one gets from a Raman transition with zero two-photon detuning in the rotating frame
- So long as $|\Delta| \gg |\mu B_x|$, we can adiabatically eliminate the intermediate state to get an effective two-level Hamiltonian:

$$\mathcal{H}_{\text{eff}} = -\frac{|\mu B_x|^2}{2\Delta} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}$$
- This allows driving of Rabi oscillations between the $M=\pm 1$ states at the effective Rabi frequency $\Omega_{\text{eff}} = |\mu B_x|^2 / \Delta$

Preparation of stretched state superposition

- This can be generalized to larger stretched states

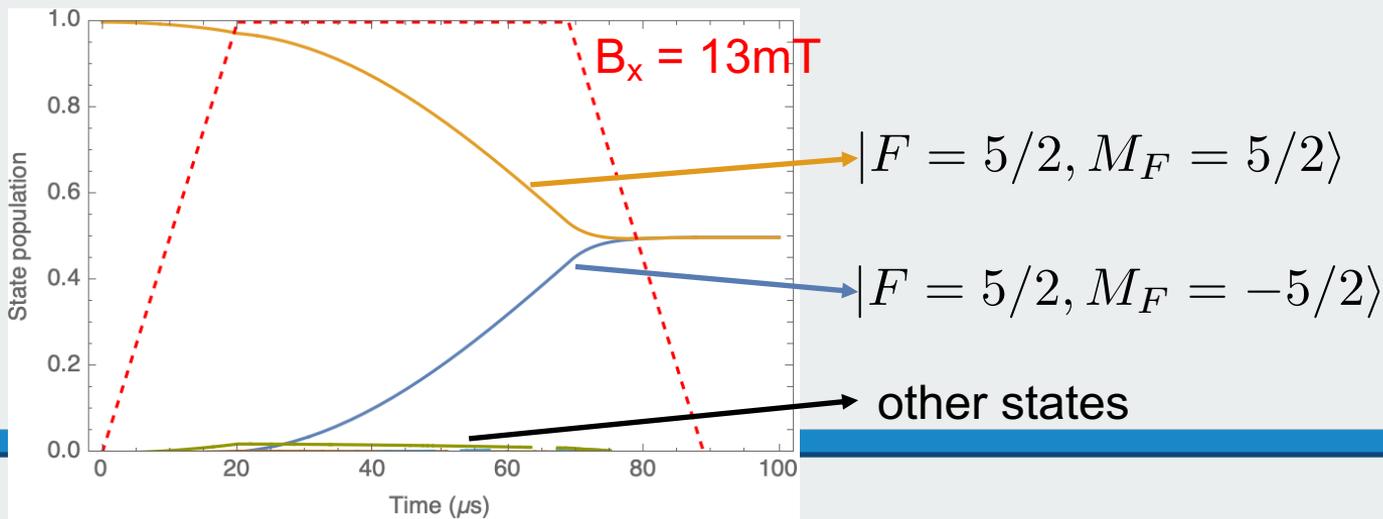
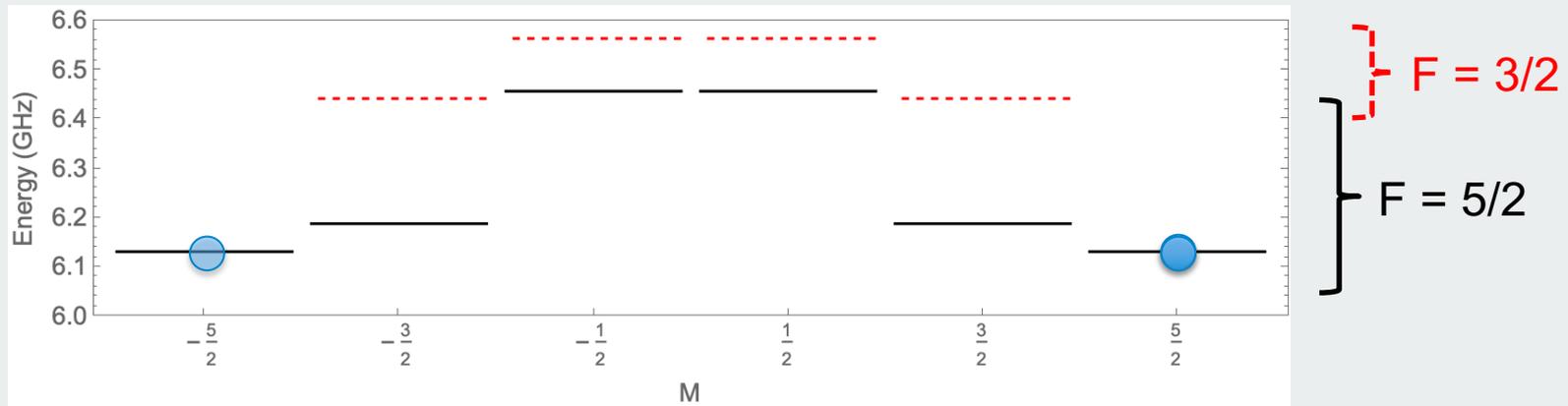


$$\mathcal{H}_{\text{eff}} = \begin{pmatrix} -\frac{|\mu_1 B_x|^2}{2\Delta_1} & \frac{|\mu_1 B_x|^2 |\mu_2 B_x|^2 \mu_3 B_x}{4\sqrt{2}\Delta_1^2 \Delta_2^2} \\ \frac{|\mu_1 B_x|^2 |\mu_2 B_x|^2 \mu_3 B_x}{4\sqrt{2}\Delta_1^2 \Delta_2^2} & -\frac{|\mu_1 B_x|^2}{2\Delta_1} \end{pmatrix}$$

- which is valid for $|\Delta_1| \gg |\mu_1 B_x|, |\mu_2 B_x|, |\Delta_2| \gg |\mu_2 B_x|, |\mu_3 B_x|$
- This still works for the weaker condition $|\Delta_1| \gg |\mu_1 B_x|$, just with a different Ω_{eff}

Preparation of stretched state superposition

- Numerical example: ^{173}YbF , $N=0$, $G=2$ manifold at $E=20\text{kV/cm}$



Systematics?

- A small B_y would cause an effective interferometer phase of $\phi = B_y/B_x$
- The part of B_y which correlates with E-field direction, $B_{y,E}$, leads to a phase that correlates with E, $\phi_E = B_{y,E}/B_x$, i.e. a systematic error
- Phase sensitivity given by $\sigma_\phi = \frac{1}{2C\sqrt{N}}$
- Given $C = 0.9$, $N = 2.6 \times 10^{10}$ molecules detected over 100 days of measurement¹, we have $\sigma_\phi = 350$ nrad
- Since $B_x = 13$ mT, we need to be able to limit $B_{y,E} < 0.5$ nT

Prospects for nuclear MQM measurements

- Can convert phase sensitivity into a frequency: $\sigma_f = \sigma_\phi / (2\pi T)$
- At Imperial (electron EDM experiments)
 - Supersonic beam of YbF, $\sigma_f = 1$ mHz/day – proof-of-principle
 - Buffer-gas-cooled beam of YbF, projected $\sigma_f = 20$ μ Hz/day – measurement?
 - Future: optical lattice of YbF, projected $\sigma_f = 90$ nHz/day – measurement in the (far) future?
- Assuming 100 days of measurement, with buffer-gas-cooled beam, can get to statistical sensitivity of 2 μ Hz

Prospects for nuclear MQM measurements

Species	¹⁹⁹ Hg (expt.)	²⁰⁵ TlF (proj.)	¹⁷³ YbF (proj.)
Nuclear moment	Schiff moment	Schiff moment	MQM
1 σ -sensitivity	10 pHz	45 nHz	2 μ Hz
QCD θ -term constraint*	6.2×10^{-11}	0.33×10^{-11}	3.0×10^{-11}
Proton EDM constraint	5.1×10^{-25} e cm	0.23×10^{-25} e cm	0.77×10^{-25} e cm
Neutron EDM constraint*	5.1×10^{-26} e cm	–	4.3×10^{-26} e cm
\bar{g}_0 constraint	1.0×10^{-12}	0.05×10^{-12}	0.8×10^{-12}
\bar{g}_1 constraint	3.3×10^{-12}	1.7×10^{-12}	0.15×10^{-12}
\bar{g}_2 constraint	8.1×10^{-13}	0.24×10^{-13}	4.0×10^{-13}

*Direct neutron EDM measurement: $|d_n| < 2.2 \times 10^{-26}$ e cm, $\theta < 18 \times 10^{-11}$

Conclusions

- Nuclear MQM measurements in diatomic molecules are promising low-energy precision searches for new CPV physics in the hadronic sector
- Current electron EDM experiments can be converted into nuclear MQM measurements
- The required sensitivity of experiments with diatomic molecules is much lower than atomic/direct neutron experiments
- State which gives maximal sensitivity to MQM is not the state where nuclear and electron spins are aligned
- Higher-order couplings required to create superposition of stretched states, e.g. using a perpendicular B field

References

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MQM effective Hamiltonian

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Collective enhancement of MQMs in heavy deformed nuclei

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Calculation of W_M for YbF

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