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Measuring the magnetic quadrupole moment of heavy nuclei with diatomic molecules

Chris Ho, SSP 2022

29 Aug 2022

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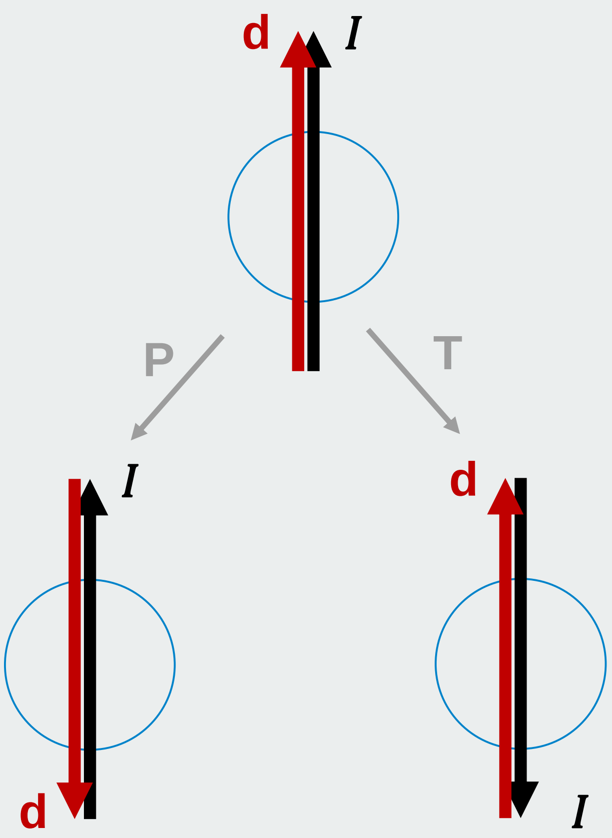
MQMs.. what the heck?

Nuclear EDM (I ≥ 1/2)



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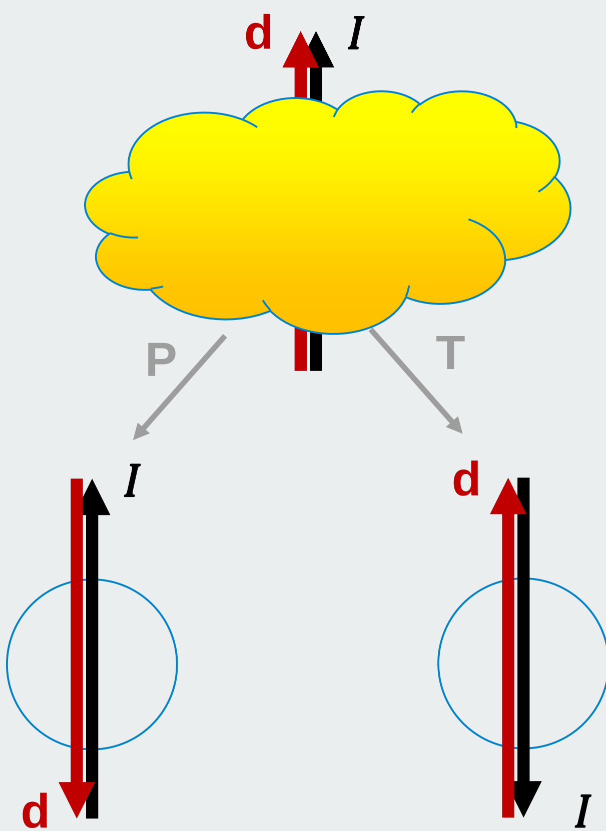

Electrons shield nuclear EDM almost perfectly; remaining moment is called **Schiff moment**

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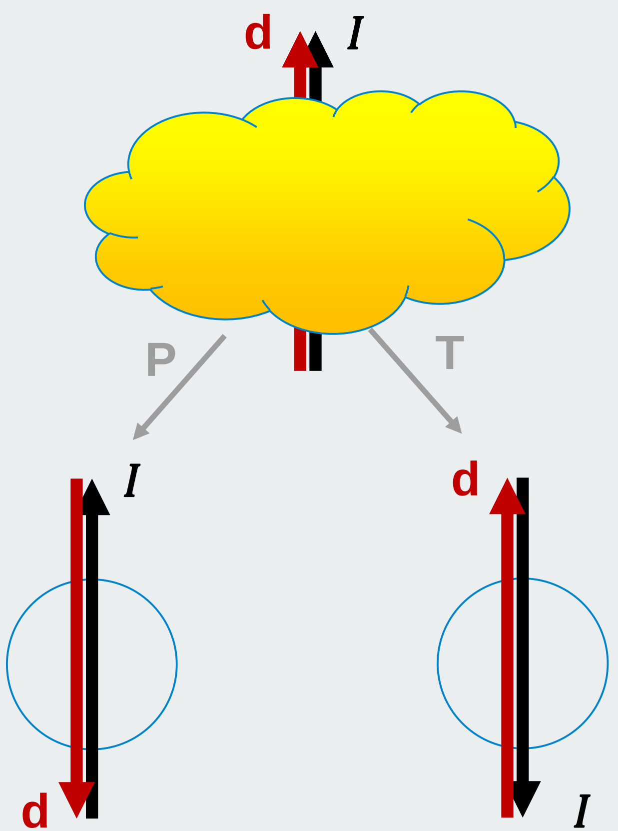
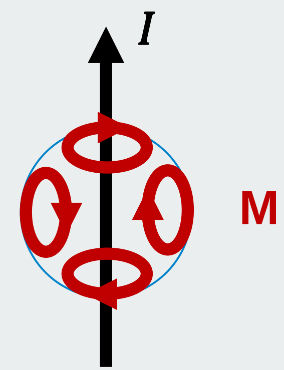
MQMs.. what the heck?

Nuclear ~~EDM~~ Schiff moment (I ≥ 1/2)



Nuclear MQM (I ≥ 1)



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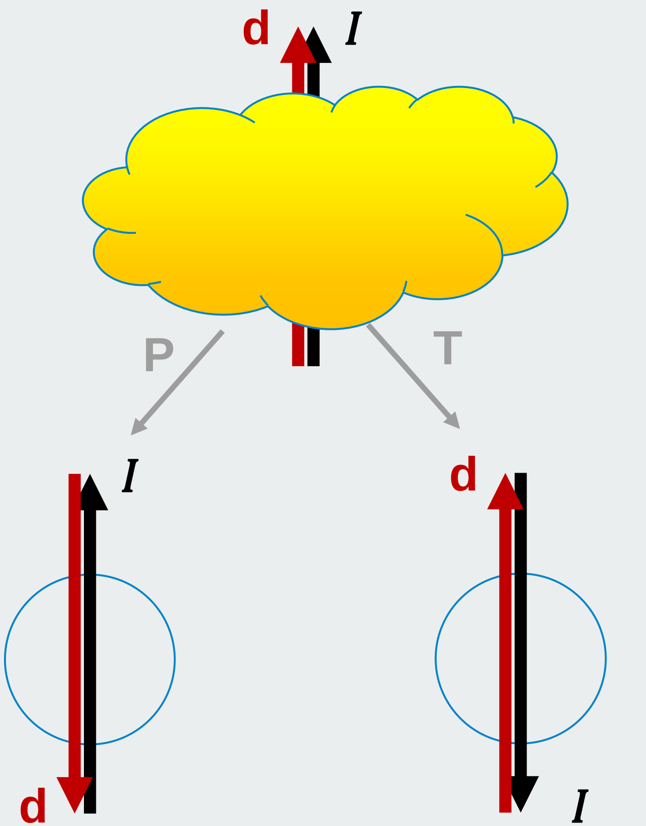

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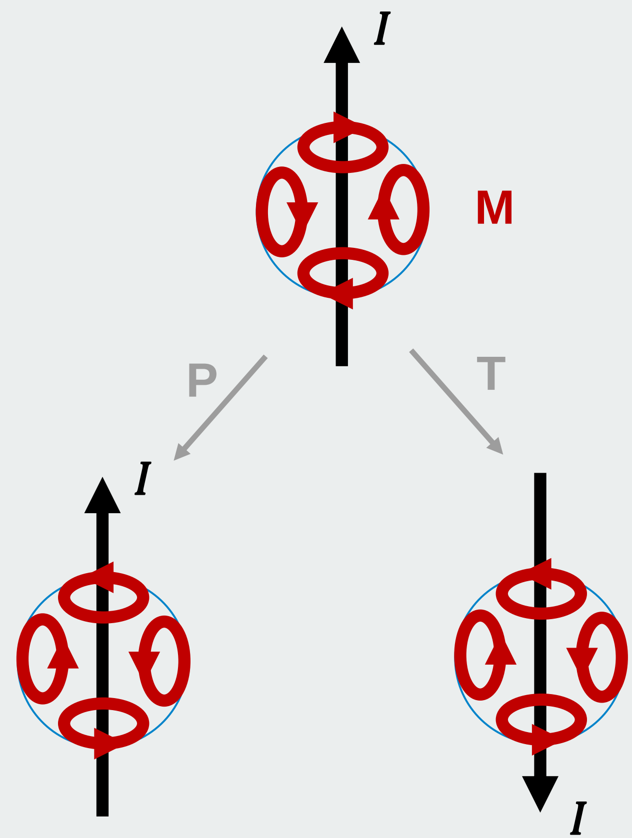

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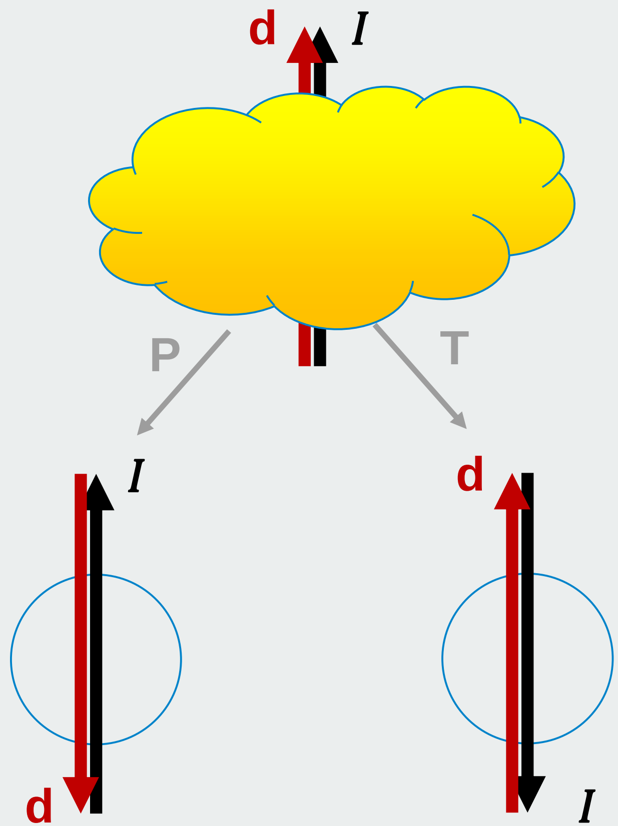

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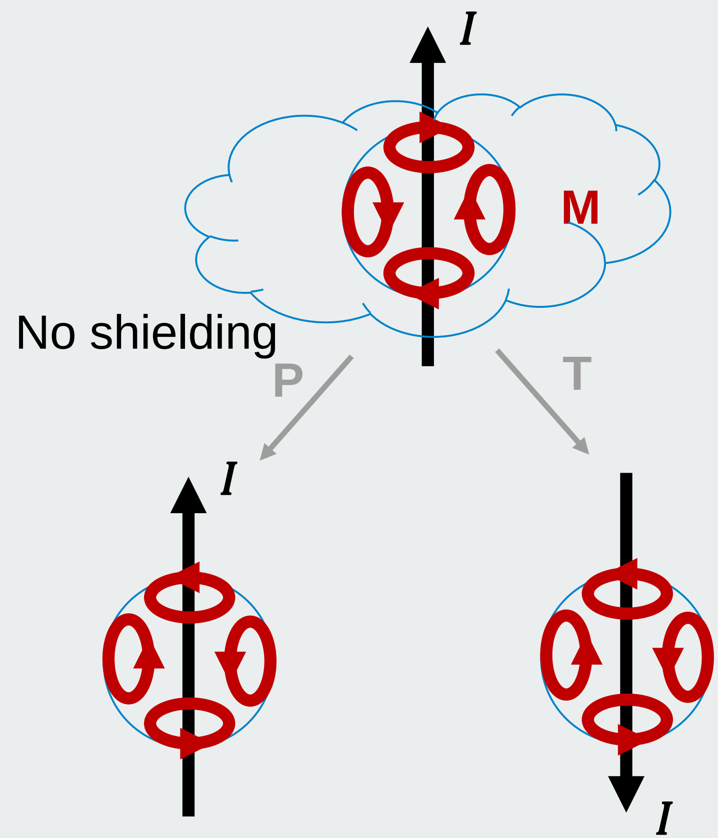

MQMs.. what the heck?

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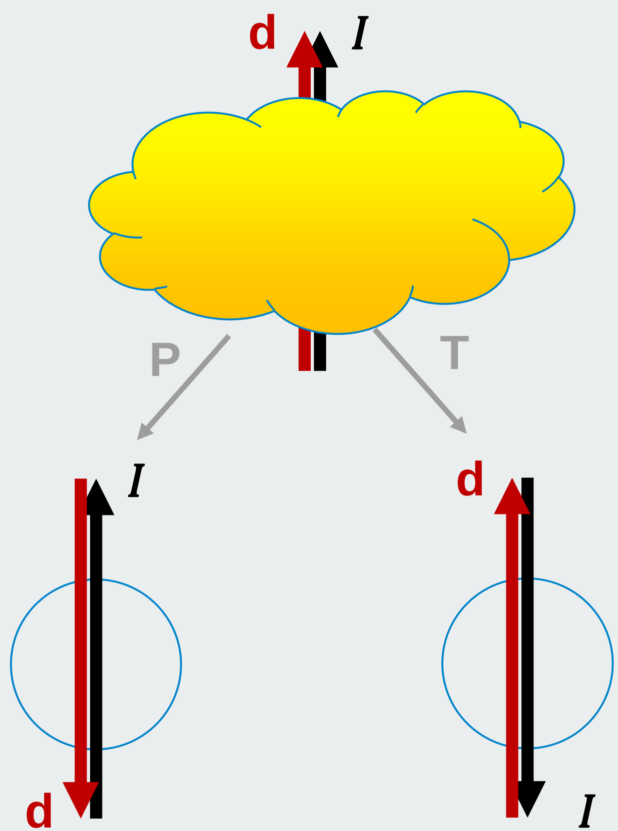

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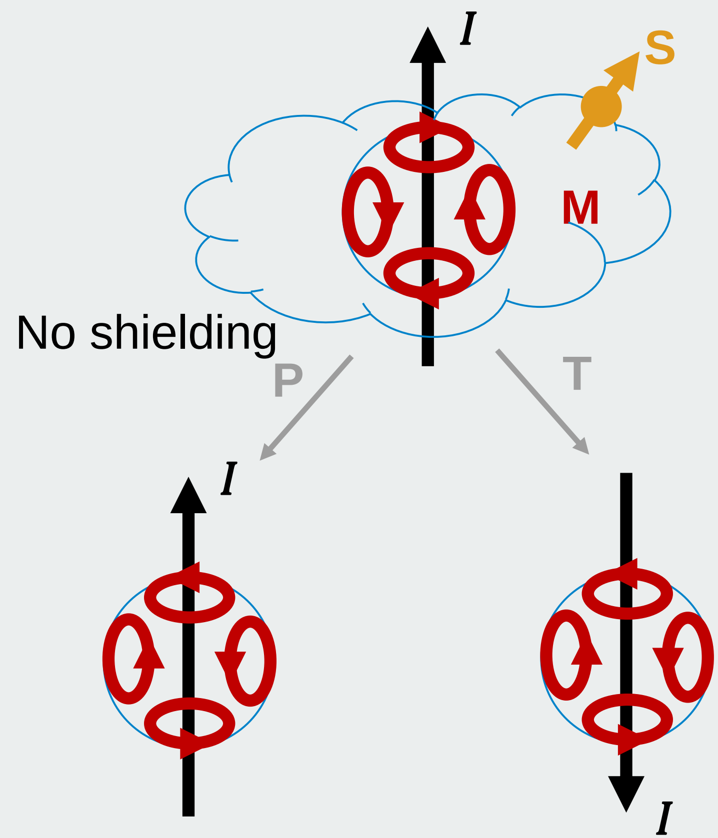

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MQMs.. what the heck?

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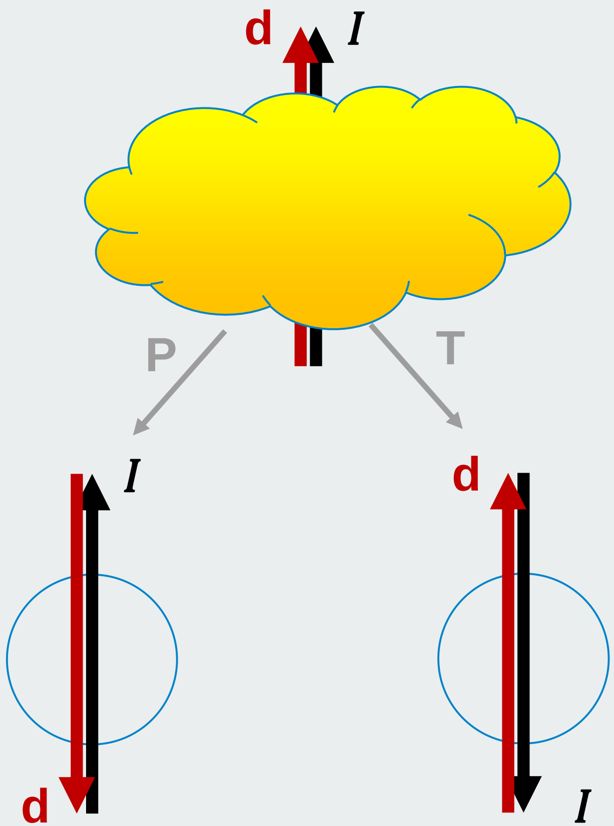

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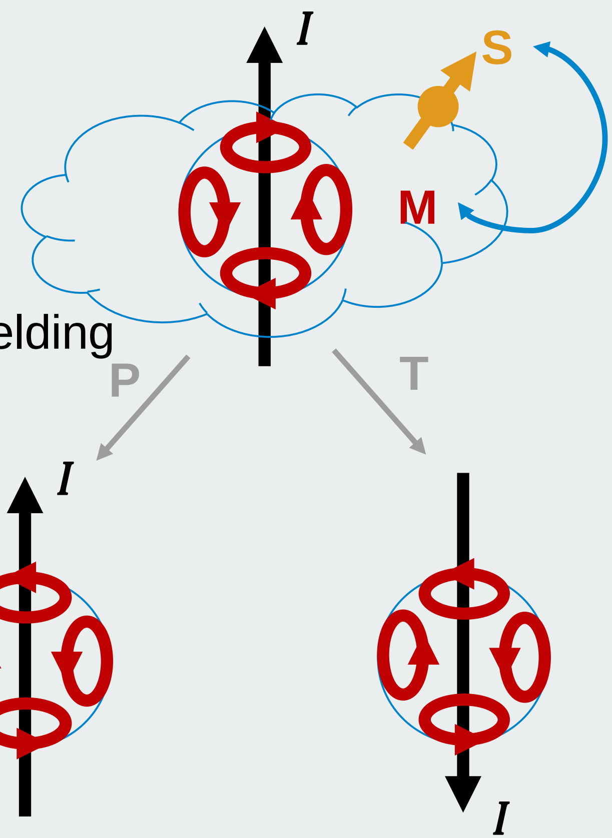

No shielding

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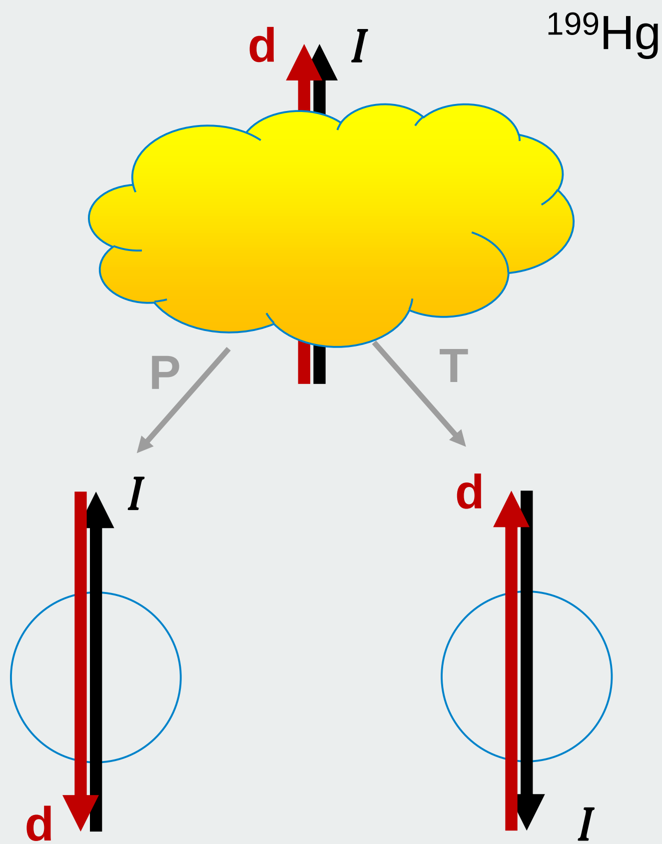
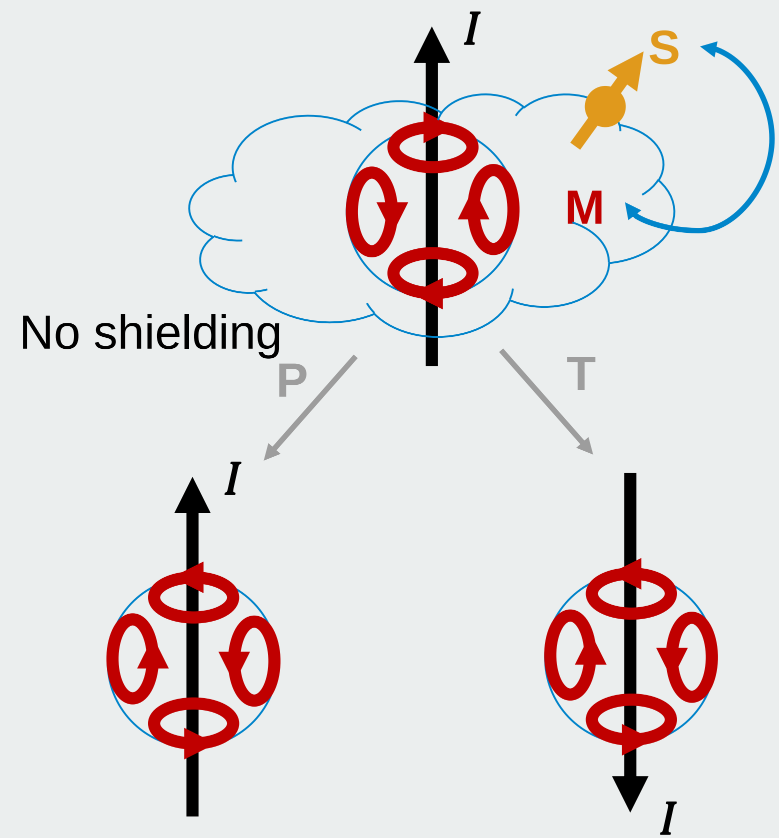
MQMs.. what the heck?

Nuclear ~~EDM~~ Schiff moment (I ≥ 1/2)



Nuclear MQM (I ≥ 1)



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MQMs.. what the heck?

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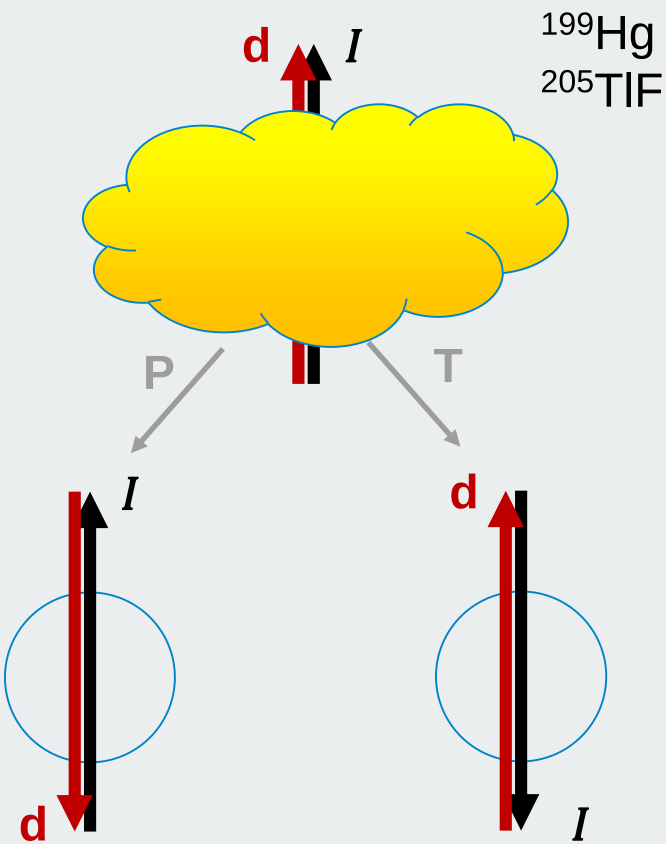

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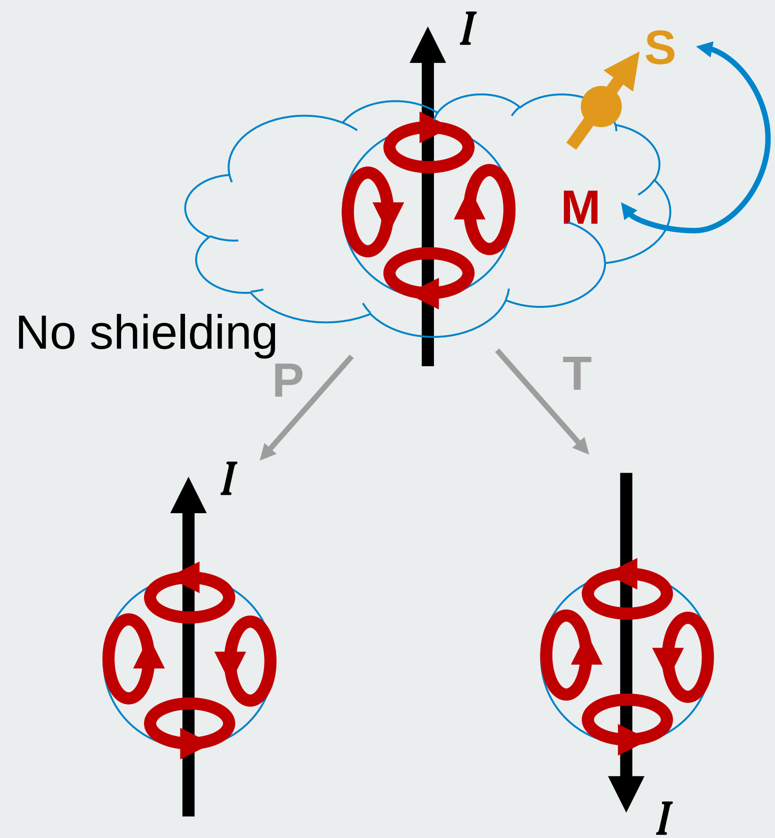

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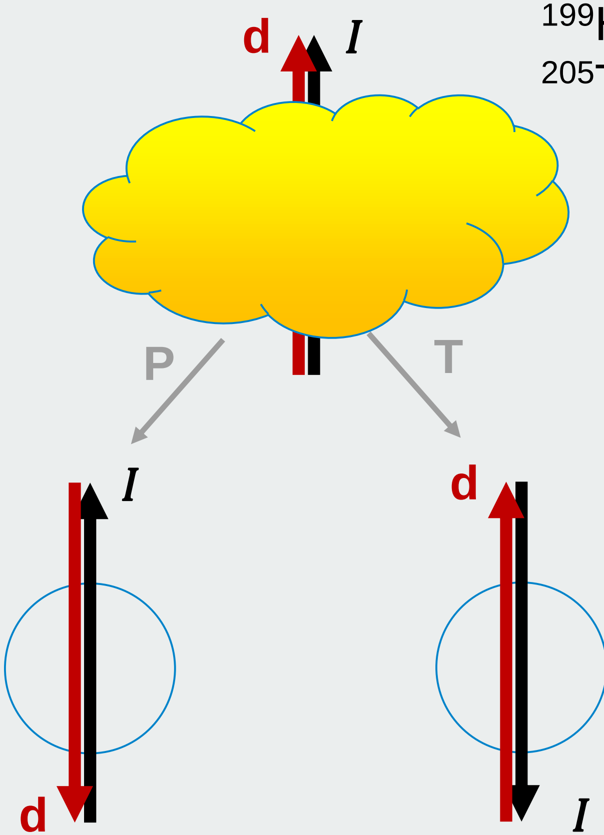
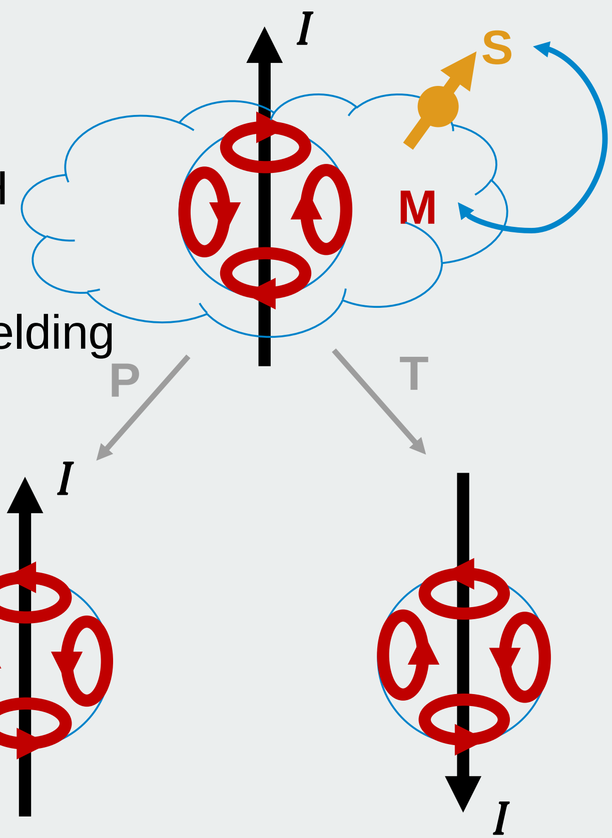


^{199}Hg
^{205}TlF

Nuclear MQM (I ≥ 1)

^{173}YbF
^{229}ThO
^{177}HfF^{+}
^{173}YbOH
^{137}BaF

No shielding



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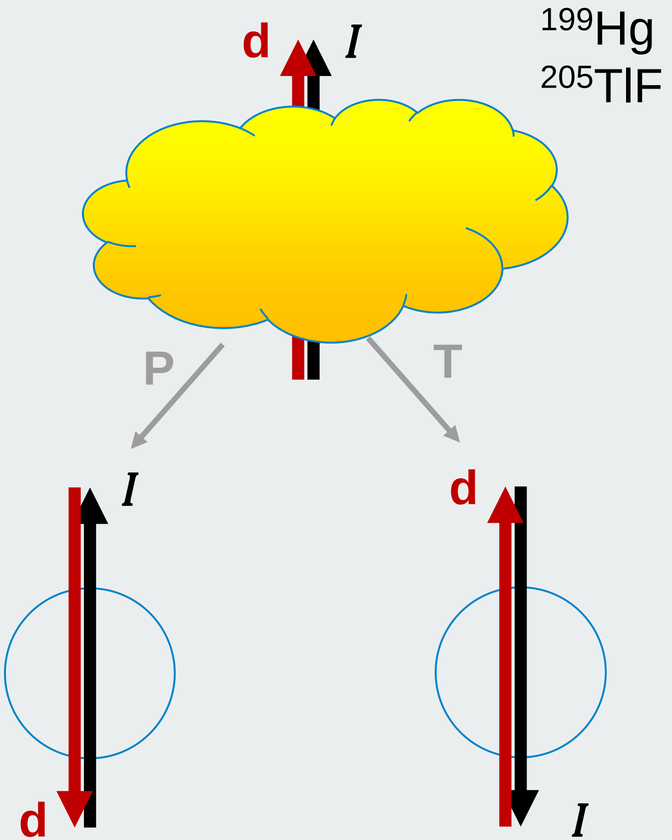
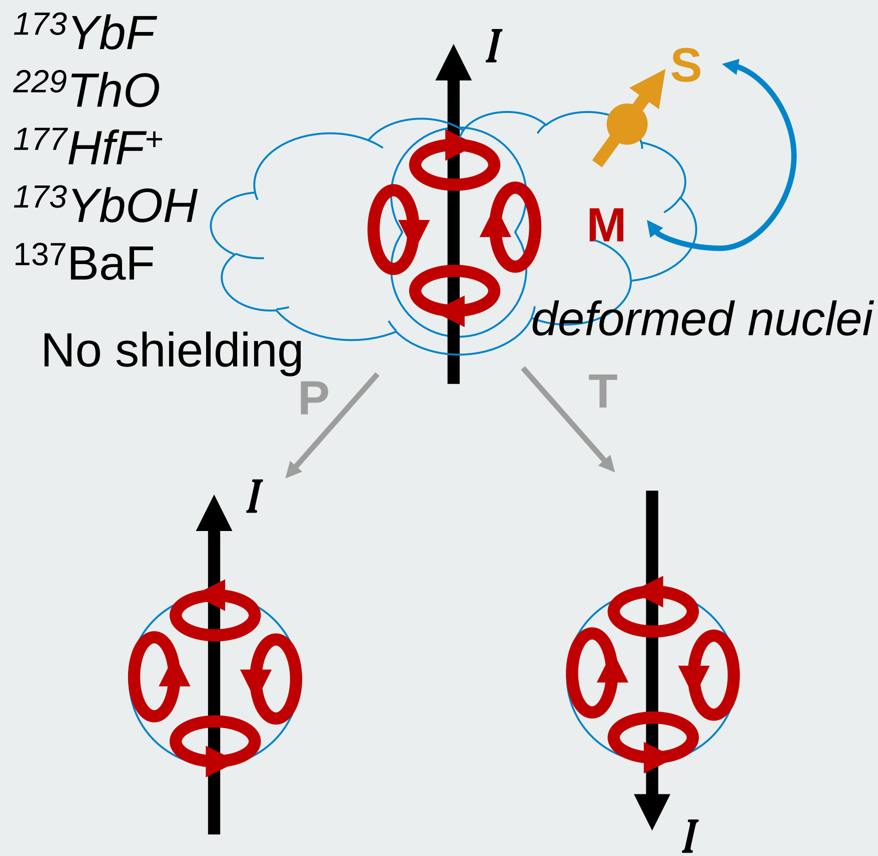
Nuclear ~~EDM~~ Schiff moment (I ≥ 1/2)



Nuclear MQM (I ≥ 1)



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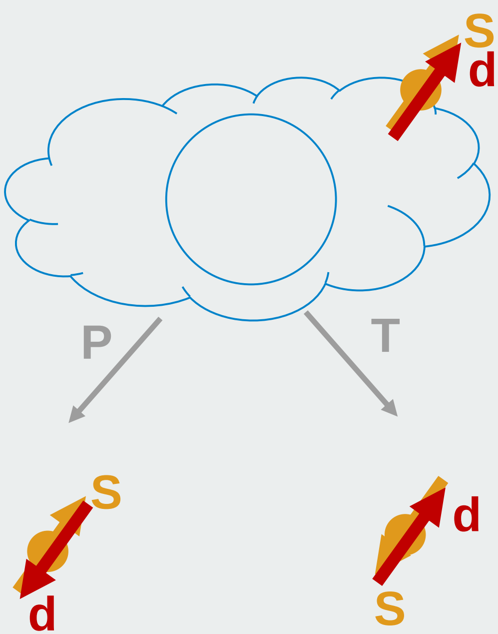
MQMs.. what the heck?

Electron EDM

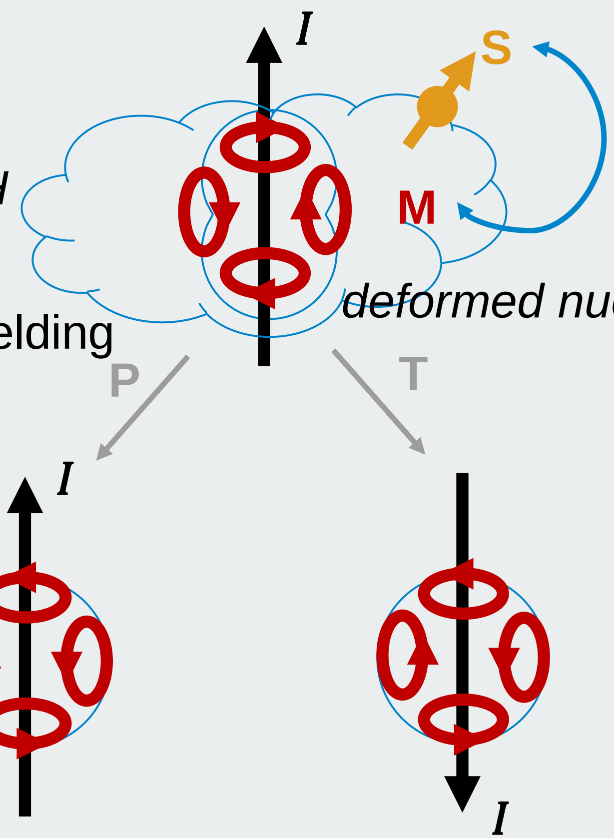


^{174}YbF
^{232}ThO
^{180}HfF^{+}
^{174}YbOH
^{138}BaF

Nuclear MQM (I ≥ 1)

^{173}YbF
^{229}ThO
^{177}HfF^{+}
^{173}YbOH
^{137}BaF

No shielding



deformed nuclei

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

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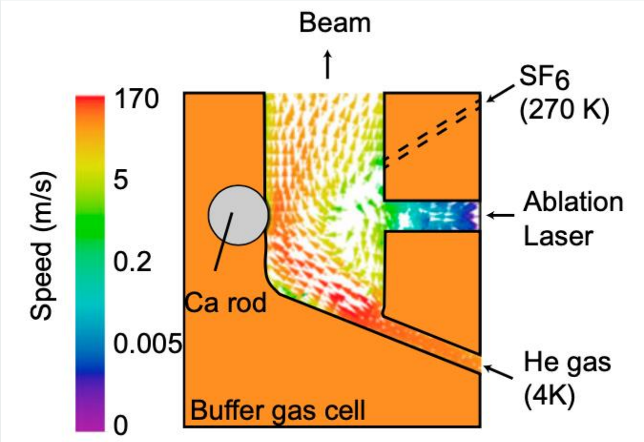
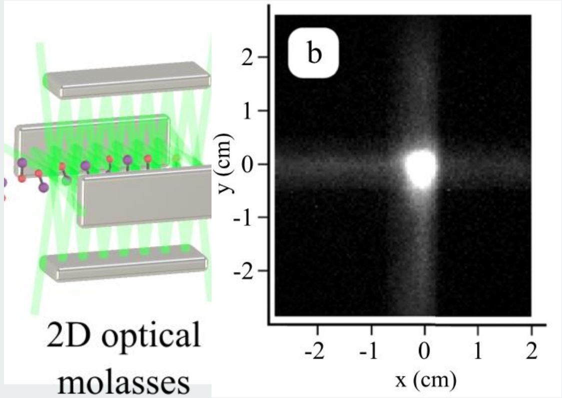
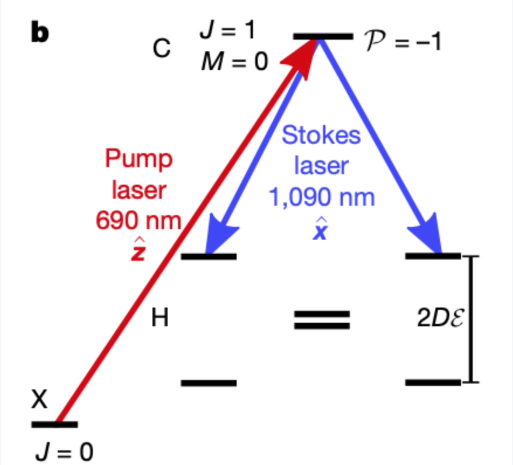
Experimental advances with diatomic molecules



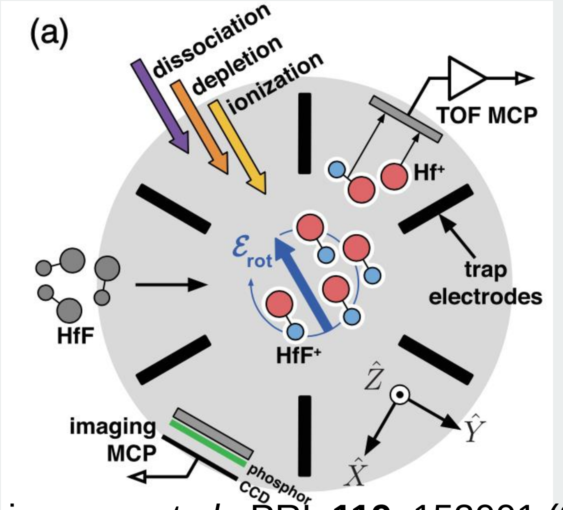
Truppe *et al.*, J. Mod. Opt. **65**, 648 (2018)



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



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



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

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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)

n: unit vector along internuclear axis

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

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

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MQM matrix elements in diatomic molecules

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

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

- 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 **S** and **I** are aligned (i.e. where G is largest) does not give the largest energy shift

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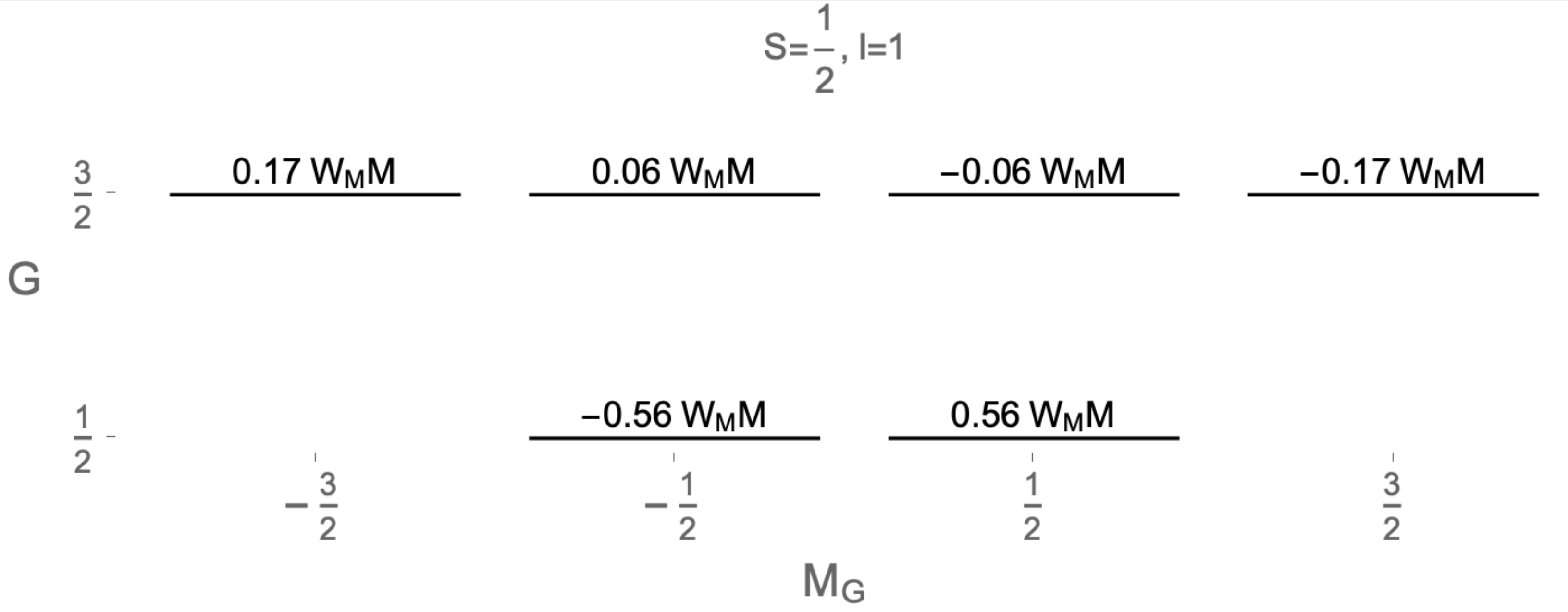
Example I: $\langle H_M \rangle_{mol}$ for S = 1/2, I = 1



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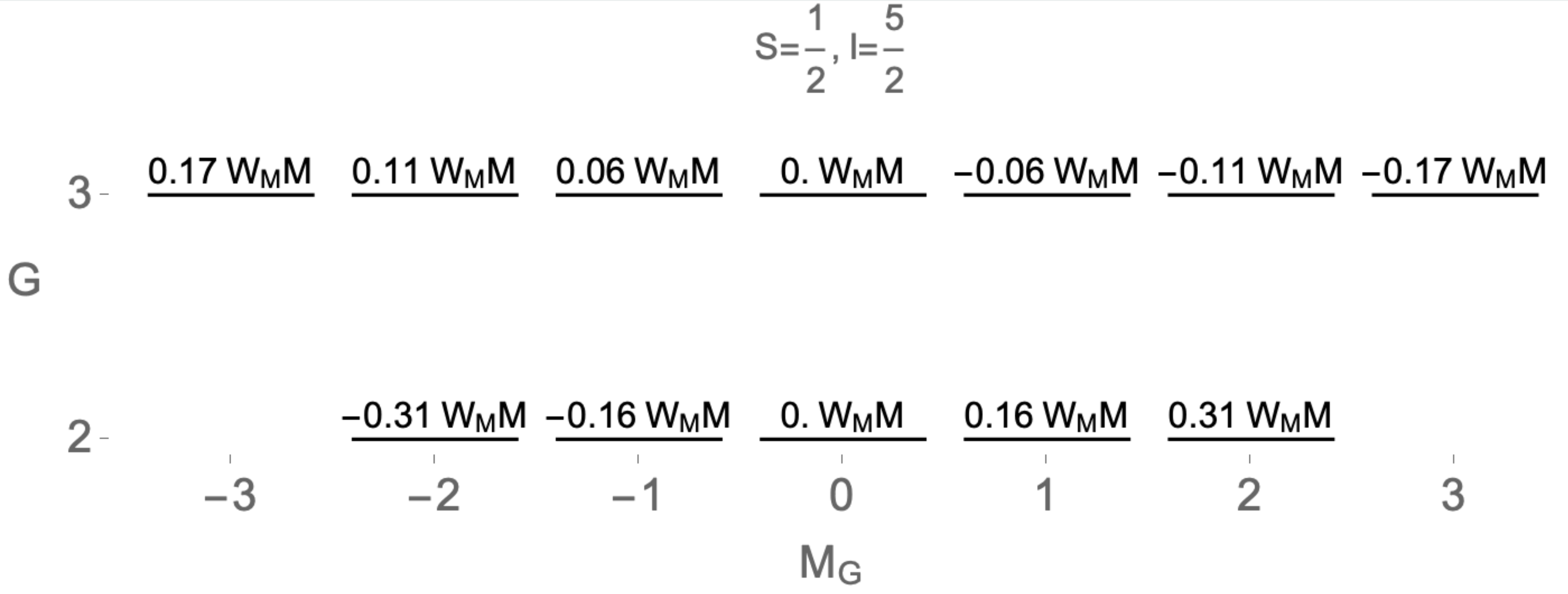
Example II: <H_M>$_{mol}$ for $S = 1/2$, $I = 5/2$ (^{173}YbF)



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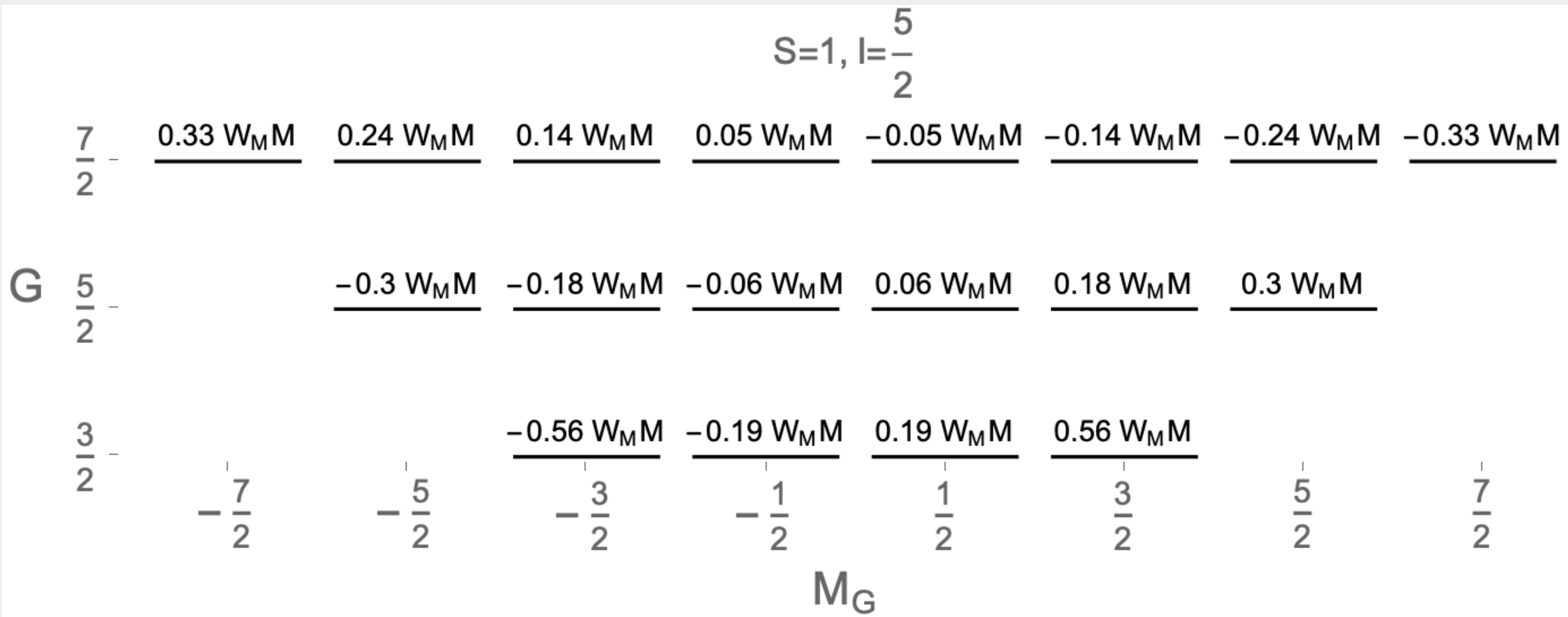
Example III: $\langle H_M \rangle_{mol}$ for S = 1, I = 5/2 (^{229}ThO)



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How to measure the MQM energy shift?

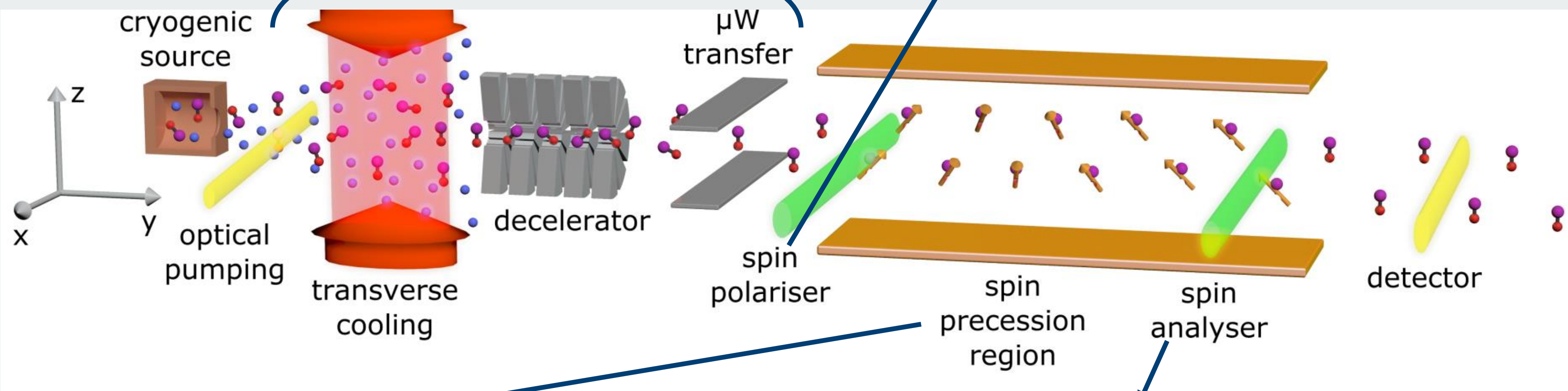
- Use a spin interferometer – just like measuring the electron EDM

state preparation

$$|G, M_G = +G\rangle$$

prepare superposition of stretched states

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



states gain relative phase

$$2\phi = (\Delta_M + \Delta_B)T$$

convert phase difference into population difference

$$\cos\phi\, |G, M_G = +G\rangle + \sin\phi\, |G, M_G = -G\rangle$$

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Preparation of stretched state superposition

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

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

- 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.

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

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

- 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

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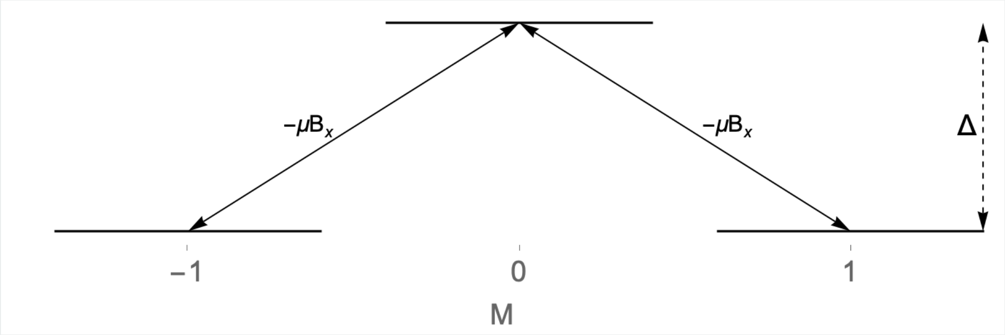
Preparation of stretched state superposition

- 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=±1 states at the effective Rabi frequency $\Omega_{\text{eff}} = |\mu B_x|^2/\Delta$

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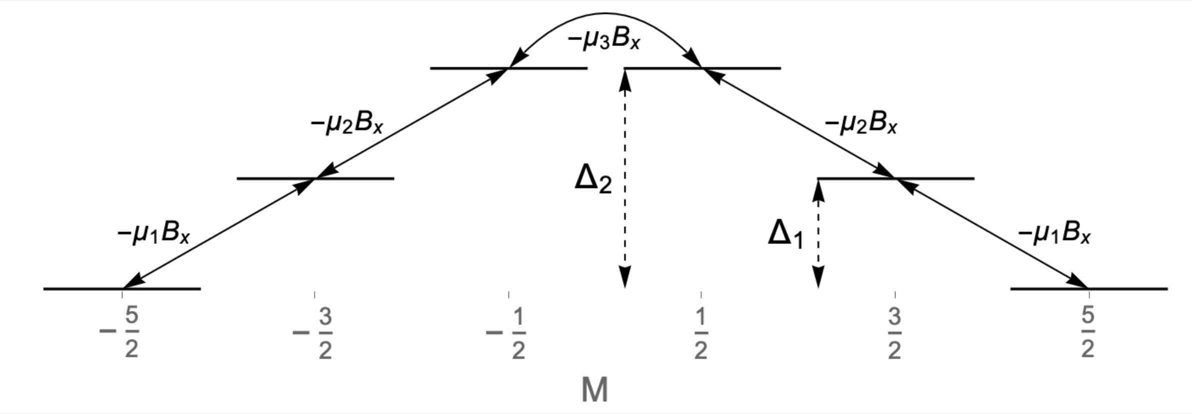
Preparation of stretched state superposition

- This can be generalized to larger stretched states



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

- 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}

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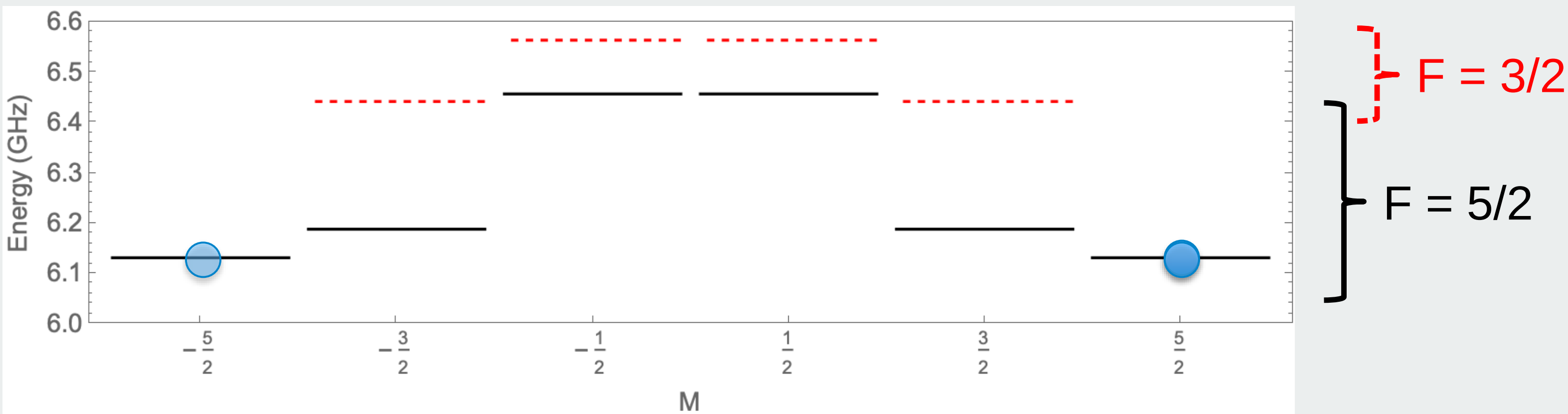
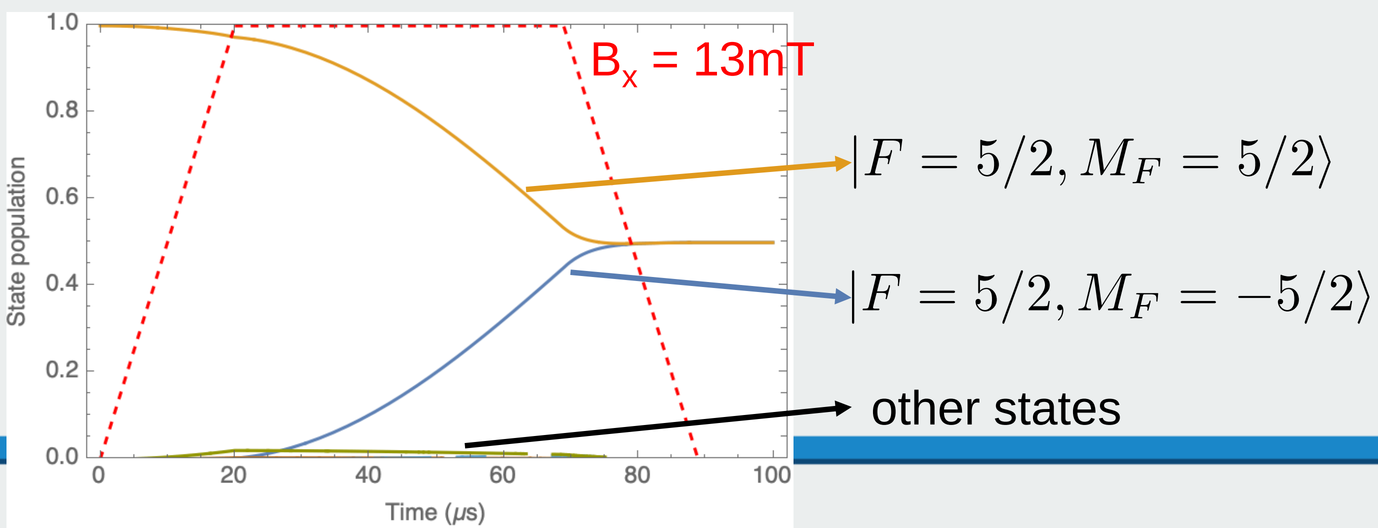
Preparation of stretched state superposition

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





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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 $\sigma_\phi = \dfrac{1}{2\mathcal{C}\sqrt{N}}$
- Given $\mathcal{C} = 0.9$, $N = 2.6 \times 10^{10}$ molecules detected over 100 days of measurement[1], we have $\sigma_\phi = 350$ nrad
- Since $B_x = 13$ mT, we need to be able to limit $B_{y,E} < 0.5$ nT

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

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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, σ_f = 1 mHz/day – proof-of-principle
  - Buffer-gas-cooled beam of YbF, projected σ_f = 20 μHz/day – measurement?
  - Future: optical lattice of YbF, projected σ_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 μHz

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Prospects for nuclear MQM measurements

Species	^{199}Hg (expt.)	^{205}TlF (proj.)	^{173}YbF (proj.)
Nuclear moment	Schiff moment	Schiff moment	MQM
1σ-sensitivity	10 pHz	45 nHz	2 μHz
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, $\theta < 18 \times 10^{-11}$

Hg: Graner *et al.*, PRL **116**, 161601 (2016) | TlF: Grasdijk *et al.*, Q. Sci. Tech. **6**, 044007 (2021) | nEDM: Abel *et al.*, PRL **124**, 081803 (2020)

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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

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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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Acknowledgements

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