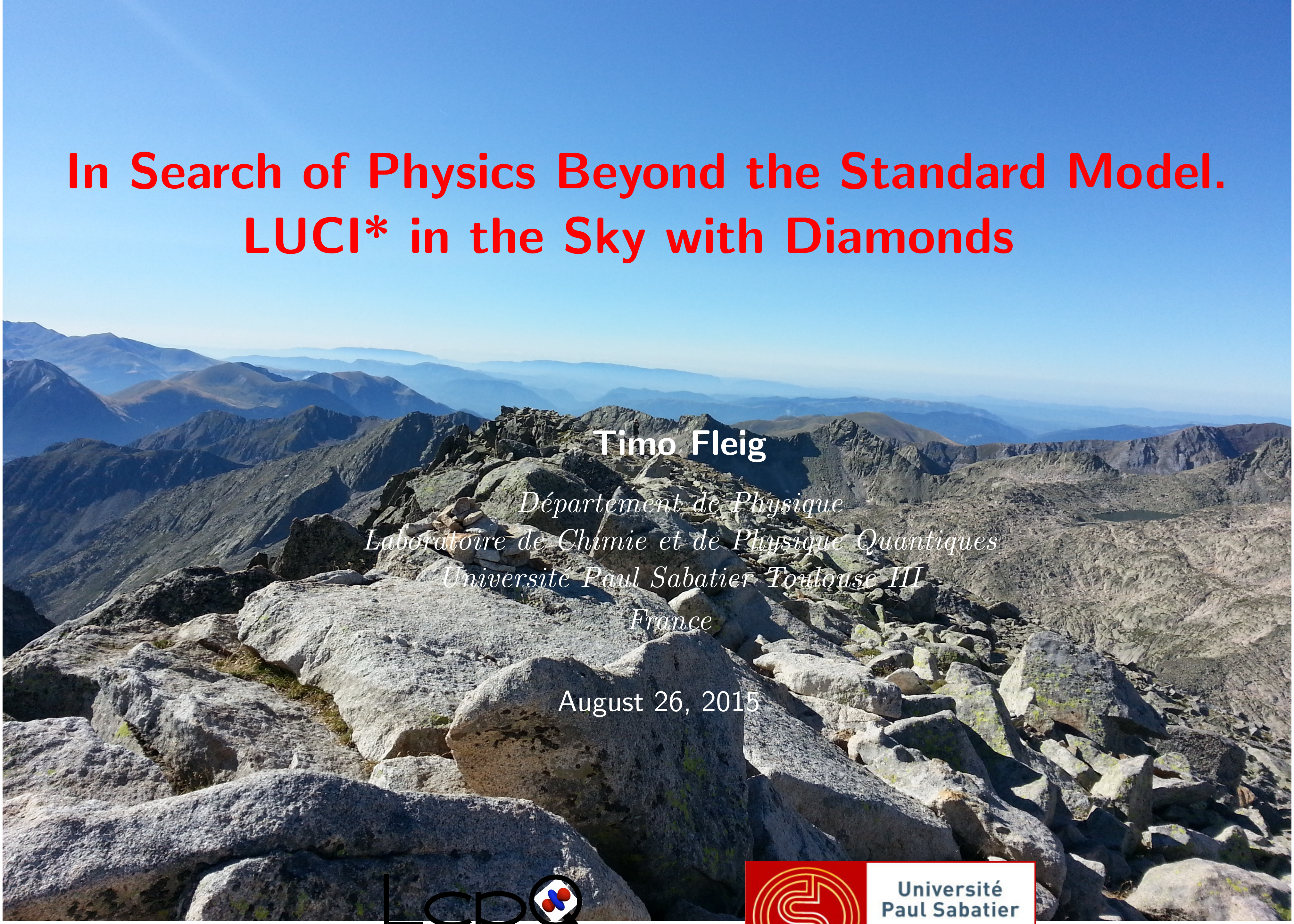


In Search of Physics Beyond the Standard Model. LUCI* in the Sky with Diamonds

Timo Fleig

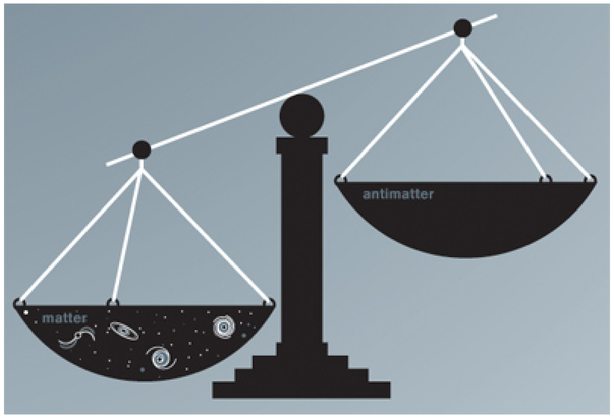
Département de Physique
Laboratoire de Chimie et de Physique Quantiques
Université Paul Sabatier Toulouse III
France

August 26, 2015

Laboratoire de Chimie et Physique Quantiques

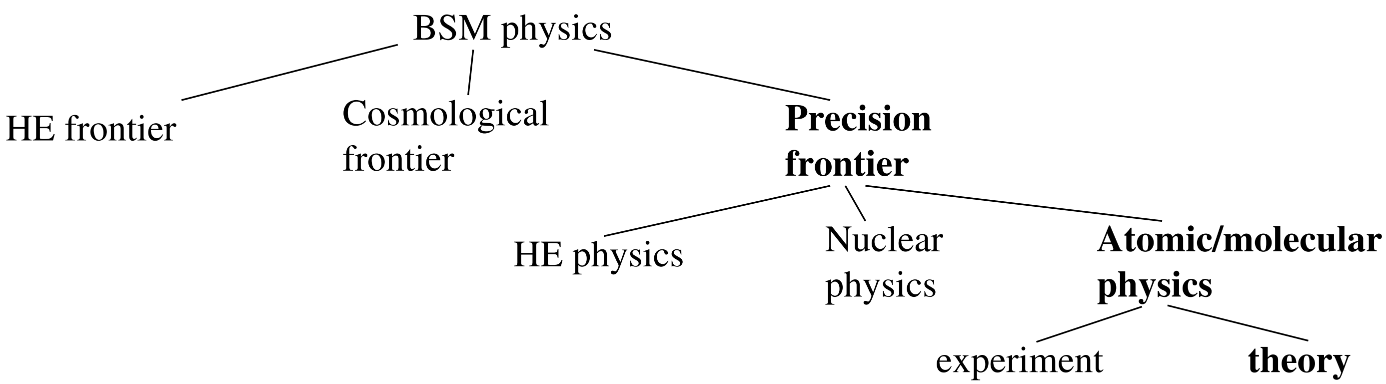
Université Paul Sabatier
TOULOUSE III

Open Questions at Large Scale and at Small Scale



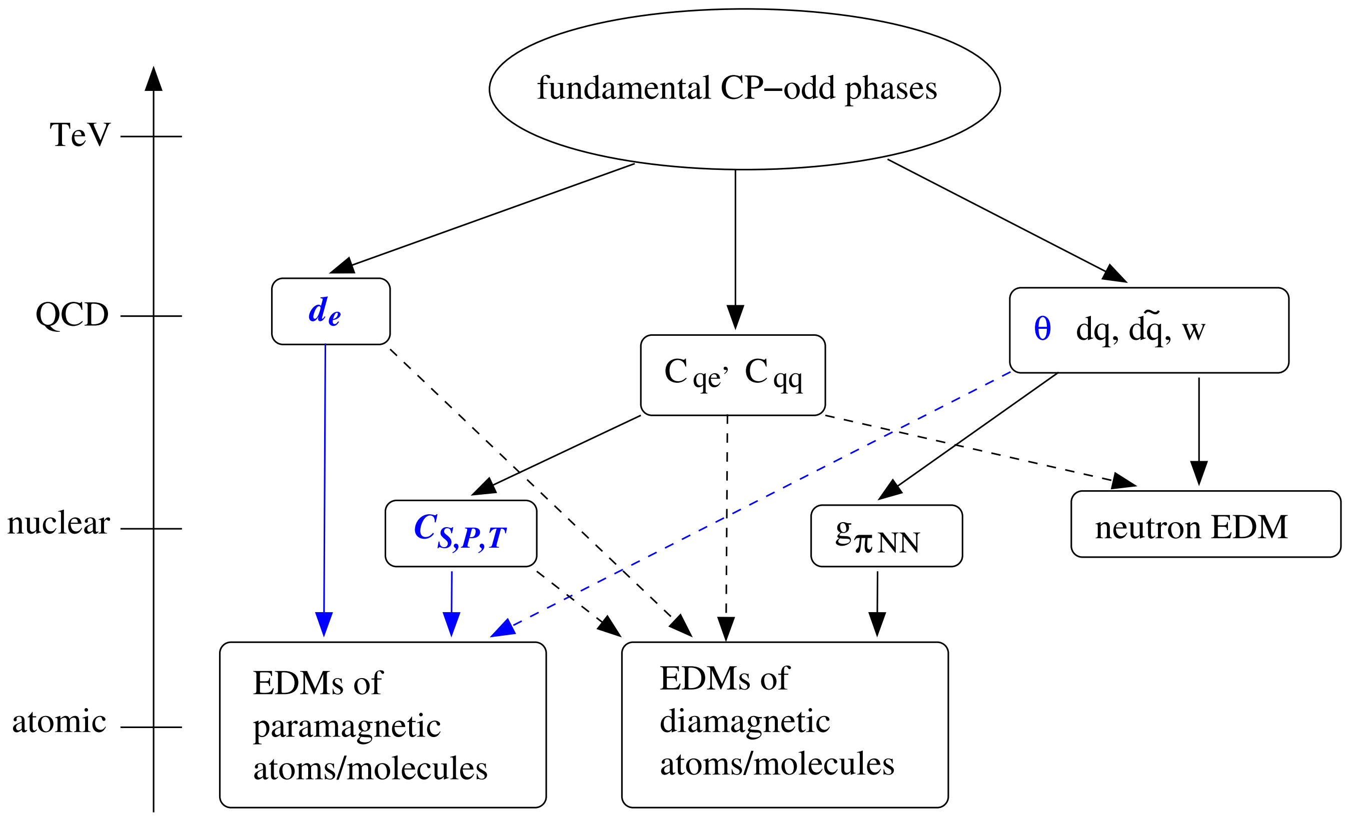
- **Matter-antimatter asymmetry** of the universe[1]
- Nature of **cold dark matter**
- Degree of $\mathcal{CP}$ **violation** in nature[2]

- Detection/constraint of **EDMs** as a powerful probe of possible explanations/consequences[3]



[1] M. Dine, A. Kusenko, *Rev. Mod. Phys.* **76** (2004) *1*
[2] G. C. Branco, R. G. Felipe, F. R. Joaquim, *Rev. Mod. Phys.* **84** (2012) *515*
[3] J. Engel, M. J. Ramsey-Musolf, U. van Kolck, *Prog. Part. Nuc. Phys.* **71** (2013) *21*

Electric Dipole Moments and Their Source Tree[4]



d_e: electron EDM

($\mathcal{P}$ and $\mathcal{T}$)-violating electron-nucleon interaction

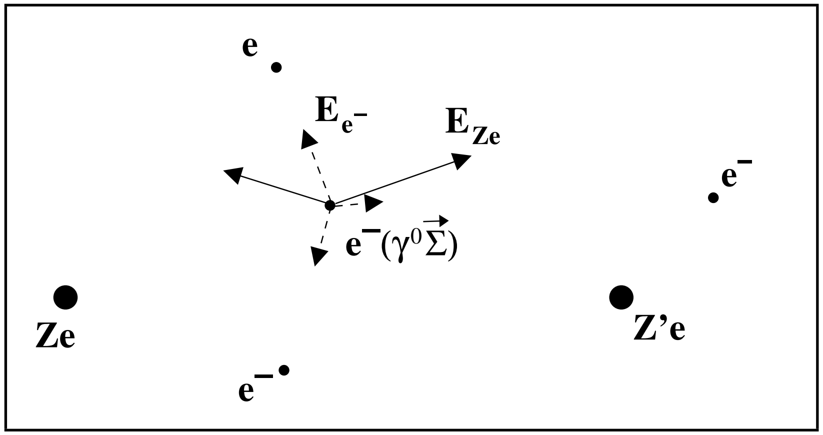
($\mathcal{P}$ and $\mathcal{T}$)-violating NMQM interaction

- EDMs are low-energy physics probes of high-energy physics symmetry breaking

[4] M. Pospelov, A. Ritz, "Electric dipole moments as probes of new physics", *Ann. Phys.* **318** (2005) *119*

Electron EDM Interaction

$$d_e = \frac{\Delta\epsilon}{E_{\text{eff}}} \quad \begin{matrix}\text{(Experiment)}\\ \text{(Theory)}\end{matrix}$$



Single-particle $\mathcal{P}$- and $\mathcal{T}$-odd eEDM Hamiltonian[5]:

$$\hat{H}_{\text{EDM}} = -\frac{d_e}{4}\gamma^0\gamma^5\left(\gamma^\mu\gamma^\nu - \gamma^\nu\gamma^\mu\right)F_{\mu\nu} = -d_e\gamma^0\left[\boldsymbol{\Sigma}\cdot\mathbf{E} + \imath\boldsymbol{\alpha}\cdot\mathbf{B}\right]$$

Internal electric field contributions

$$\mathbf{E}_{\text{int}}(i) = \sum_{A=1}^{N}\frac{Ze\left(\vec{r}_i - \vec{r}_A\right)}{||\vec{r}_i - \vec{r}_A||^3} - \sum_{j=1}^{n}\frac{e\left(\vec{r}_i - \vec{r}_j\right)}{||\vec{r}_i - \vec{r}_j||^3}$$

Expectation value in many-body system in accord with stratagem II[6]

$$-\left\langle \sum_{j=1}^{n}\gamma_j^0\,\boldsymbol{\Sigma}_j\cdot\mathbf{E}_j \right\rangle_{\psi^{(0)}} \approx \frac{2\imath c}{e\hbar}\left\langle \sum_{j=1}^{n}\gamma_j^0\gamma_j^5\,\vec{p}_j^{\;2} \right\rangle_{\psi^{(0)}} := E_{\text{eff}}$$

[5] E. Salpeter, *Phys Rev* **112** (1958) *1642*

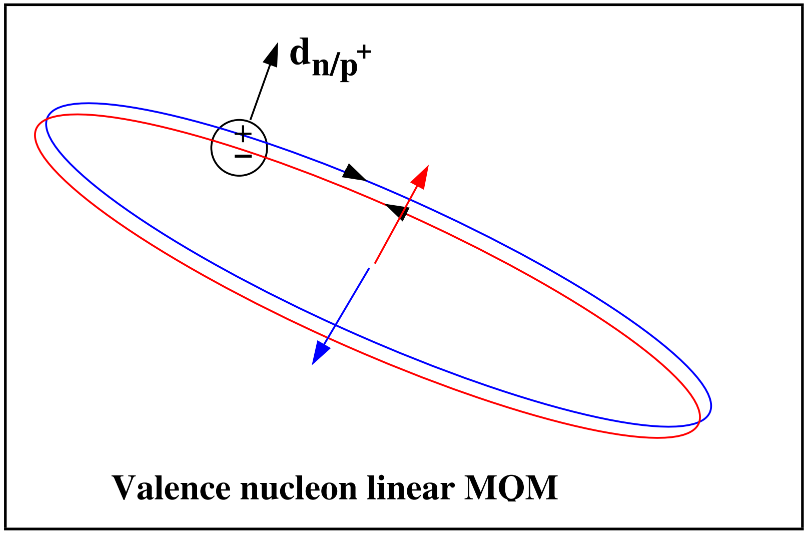
[6] E. Lindroth, E. Lynn, P.G.H. Sandars, *J Phys B: At Mol Opt Phys* **22** (1989) *559*

Nuclear Magnetic Quadrupole Moment Interaction[7]

Effective molecular Hamiltonian[8]
for linear molecule along $\mathbf{n}$:

$$\hat{H}^{\text{eff}}_{MQM} = -\frac{W_M\, M}{2I(2I-1)}\, \mathbf{J_e}\, \hat{\mathbf{T}}\, \mathbf{n}$$

W_M: MQM-electron-magnetic-field interaction constant



with the components of the nuclear MQM

$$M_{i,k} = \frac{3M}{2I(2I-1)}\, T_{i,k} \qquad T_{i,k} = I_i I_k + I_k I_i - \frac{2}{3}\, \delta_{i,k}\, I(I+1),$$

$$W_M \propto \left(\frac{\boldsymbol{\alpha}\times\mathbf{r}}{r^5}\right)_3 x_3$$

Calculation[9] via electric-field gradient with the help of

$$\left(\frac{\boldsymbol{\alpha}\times\mathbf{r}}{r^5}\right)_3 x_3 = \alpha_1\, \frac{x_2 x_3}{r^5} - \alpha_2\, \frac{x_1 x_3}{r^5} \qquad \iiint_{\mathcal{V}} \frac{x_i x_j}{r^5}\, d^3x = \frac{1}{3} \iiint_{\mathcal{V}} \frac{\partial}{\partial x_i} \frac{x_j}{r^3}\, d^3x$$

[7] I.B. Khriplovich, *Sov. Phys. JETP* **44** (1976) *25*; O.P. Sushkov, V.V. Flambaum, I.B. Khriplovich, *Sov. Phys. JETP* **60** (1984) *873*
[8] V.V. Flambaum, D. DeMille, M.G. Kozlov, *Phys Rev Lett*, **113** (2014) *103003*
[9] TF, M.K. Nayak, M.G. Kozlov, *Phys Rev*, to be submitted

Correlated Wavefunction Theory for $\mathcal{P}$,$\mathcal{T}$-odd Properties

- Dirac-Coulomb Hamiltonian operator

$$\hat{H}^{DC} = \sum_A \sum_i \left[c(\vec{\alpha}\cdot\vec{p})_i + \beta_i m_0 c^2 + V_{iA}\right] + \sum_{i,j>i} \frac{1}{r_{ij}} \mathbb{1}_4 + \sum_{A,B>A} V_{AB}$$

- All-electron Dirac-Coulomb Hartree-Fock (DCHF) calculation

  set of time-reversal paired 4-spinors $\hat{K}\varphi_i = \varphi_{\bar{i}}$ and $\hat{K}\varphi_{\bar{i}} = -\varphi_i$

- Expansion and variation[10] in n-electron sector of Fock space

$$|\psi_k\rangle = \sum_{I=1}^{\dim\mathcal{F}^{\rm t}({\rm M,n})} c_{kI} \, (\mathcal{S}\overline{\mathcal{T}})_I \,|\ \rangle$$

unbarred (Kramers up) string $\mathcal{S} = a_i^\dagger a_j^\dagger a_k^\dagger \ldots$

barred (Kramers down) string $\overline{\mathcal{S}} = a_{\bar{l}}^\dagger a_{\bar{m}}^\dagger a_{\bar{n}}^\dagger \ldots$

Expectation values over relativistic Configuration Interaction wavefunctions[11]

$$\left\langle \hat{H}' \right\rangle_{\psi_k^{(0)}} = \sum_{I,J=1}^{\dim\mathcal{F}^{\rm t}({\rm M,n})} c_{kI}^* c_{kJ} \, \langle\ | (\mathcal{S}\overline{\mathcal{T}})_I^\dagger \left| \hat{H}' \right| (\mathcal{S}\overline{\mathcal{T}})_J |\ \rangle$$

---

[10] S. Knecht, H.J.Aa. Jensen, T.F., *J Chem Phys* **132** (2010) *014108*

[11] S. Knecht, Dissertation, HHU Düsseldorf 2009

$\mathcal{P},\mathcal{T}$-odd Properties as Expectation Values

Interaction constants for n-electron system

- Electron eEDM interaction constant

$$W_d := \frac{2\imath c}{\Omega\, e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \vec{p}_j^{\ 2} \right\rangle_{\psi_k^{(0)}} \qquad \left\langle \hat{H}_{\text{eEDM}} \right\rangle = d_e\, \Omega\, W_d$$

- S-PS electron-nucleon interaction constant

$$W_{\mathcal{P},\mathcal{T}} := \frac{\imath}{\Omega} \frac{G_F}{\sqrt{2}}\, Z \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \rho_N(\vec{r}_j) \right\rangle_{\psi_k^{(0)}} \qquad \left\langle \hat{H}_{\text{e-nSPS}} \right\rangle = k_s\, \Omega\, W_{\mathcal{P},\mathcal{T}}$$

- Nuclear magnetic quadrupole - electronic magnetic field interaction

$$W_M = \frac{3}{2\Omega} \left\langle \sum_{j=1}^{n} \left(\frac{\boldsymbol{\alpha}_j \times \mathbf{r}_{jA}}{r_{jA}^5} \right)_z (r_{jA})_z \right\rangle_{\psi_k^{(0)}}$$

Four-Spinor Based Generalized-Active-Space CI[12]

TaN sample system wavefunction parameterization

		# of Kramers pairs	accumulated # of electrons min.	max.
V	*Virtual*	110	18	18
IV	*Ta: 6p,7s,7p,π* *Ta: 6s, 5dδ*	K	18–q	18
III	*N: 2p (Ta: d)*	3	16–p	16
II	*N: 2s (Ta: d)*	1	10–n	10
I	*Ta: 5s, 5p*	4	8–m	8
	Frozen core	(31)		

GAS-extended projection manifold

$$\left\langle \mu^{N}_{\rm GASCI} \right| = \left\langle \mu^{\text{particle space}}_{\text{hole space}} \right|$$

Selected sub-sets of higher excitations in projection manifold:

$$\left\langle \mu^{T} \right| \in \left\{ \left\langle \mu^{IV^1,V^2}_{III^3} \right|, \ldots, \left\langle \mu^{IV^1,V^2}_{II^2,III^1} \right| \right\}$$

$$\left\langle \mu^{Q} \right| \in \left\{ \left\langle \mu^{IV^4}_{III^3,IV^1} \right|, \ldots, \left\langle \mu^{IV^2,V^2}_{III^3,IV^1} \right|, \ldots, \left\langle \mu^{IV^2,V^2}_{II^2,III^1,IV^1} \right| \right\}$$

$$\left\langle \mu^{5} \right| \in \left\{ \left\langle \mu^{IV^5}_{III^3,IV^2} \right|, \ldots, \left\langle \mu^{IV^3,V^2}_{III^3,IV^2} \right|, \ldots, \left\langle \mu^{IV^3,V^2}_{II^2,III^1,IV^2} \right|, \ldots, \left\langle \mu^{IV^3,V^2}_{I^1,II^1,III^1,IV^2} \right| \right\}$$

[12] TF, J. Olsen, L. Visscher, *J Chem Phys* **119** (2003) *2963* , S. Knecht, H.J.Aa. Jensen, TF, *J Chem Phys* **132** (2010) *014108* , J. Olsen, *J Chem Phys* **113** (2000) *7140*

Search for the Electron EDM

Why molecules?

Be an atom in a parity eigenstate $\hat{\mathcal{P}}\,|\psi_p\rangle = \prod_{i=1}^{n} \hat{p}(i)\,\hat{\mathcal{A}}\,|\varphi_a(1)\ldots\varphi_m(n)\rangle$. Then

$$\left\langle \psi_p | \hat{H}_{\mathrm{EDM}} | \psi_p \right\rangle = \left\langle \psi_p | \hat{\mathcal{P}}^\dagger \hat{\mathcal{P}} \hat{H}_{\mathrm{EDM}} \hat{\mathcal{P}}^\dagger \hat{\mathcal{P}} | \psi_p \right\rangle = -p^2 \left\langle \psi_p | \hat{H}_{\mathrm{EDM}} | \psi_p \right\rangle$$
$$= -\left\langle \psi_p | \hat{H}_{\mathrm{EDM}} | \psi_p \right\rangle = 0$$

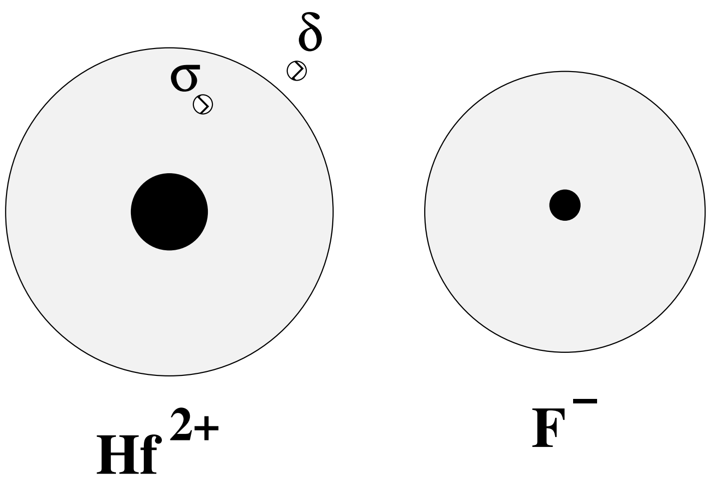
Parity eigenstates need to be mixed (polarization).

1. A perturbing laboratory $\mathbf{E}$ field is required to mix parity eigenstates.
   Tl experiment[13] $E_{\mathrm{eff}} \approx 0.05 \left[\frac{\mathrm{GV}}{\mathrm{cm}}\right]$
2. Molecular fields:
   YbF[14]: $E_{\mathrm{eff}} \approx 26 \left[\frac{\mathrm{GV}}{\mathrm{cm}}\right]$, HgF[15]: $E_{\mathrm{eff}} \approx 100 \left[\frac{\mathrm{GV}}{\mathrm{cm}}\right]$,

[13] V.V. Flambaum, *Sov J Nucl Phys* **24** (1976) *199*
[14] D.M. Kara, I.J. Smallman, J.J. Hudson, B.E. Sauer, M.R. Tarbutt, E.A. Hinds, *New J Phys* **14** (2012) *103051*
[15] Dmitriev et al., *Phys Lett* **167A** (1992) *280*

The eEDM in a molecular framework

$^3\Delta$ molecules[16]



- One heavy nucleus (relativistic effect)
- One "science" electron (σ^1)
  one "spectroscopy" electron (δ^1)
- Large $E_{\rm eff}$ for σ^1 electron

- Deeply bound and strongly polar molecules (fluorides, oxides, (nitrides))
- Small Λ (Ω)-doublet splitting[17] (optimal polarization)
- Small reduced mass (one heavy, one light atom)
- $\Omega = 1$ component preferred (small magnetic moment)

  $\Rightarrow$ Low-lying $^3\Delta_1$ as "science" state

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[16] E. Meyer, J. Bohn, D.A. Deskevich, *Phys Rev A* **73** (2006) *062108*

[17] TF, C.M. Marian, *J Mol Spectrosc* **178** (1996) *1*

ThO

Experiment: ACME Collaboration, Yale/Harvard, (DeMille/Doyle/Gabrielse groups)

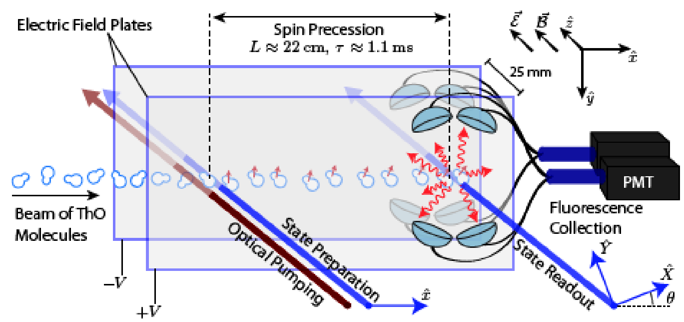
Order of Magnitude Smaller Limit on the Electric Dipole Moment of the Electron

The ACME Collaboration*: J. Baron[1], W. C. Campbell[2], D. DeMille[3], J. M. Doyle[1], G. Gabrielse[1], Y. V. Gurevich[1,**], P. W. Hess[1], N. R. Hutzler[1], E. Kirilov[3,#], I. Kozyryev[3,†], B. R. O'Leary[3], C. D. Panda[1], M. F. Parsons[1], E. S. Petrik[1], B. Spaun[1], A. C. Vutha[4], and A. D. West[3]

The Standard Model (SM) of particle physics fails to explain dark matter and why matter survived annihilation with antimatter following the Big Bang. Extensions to the SM, such as weak-scale Supersymmetry, may explain one or both of these phenomena by positing the existence of new particles and interactions that are asymmetric under time-reversal (T). These theories nearly always predict a small, yet potentially measurable (10^{-27}-10^{-30} e cm) electron electric dipole moment (EDM, d_e), which is an asymmetric charge distribution along the spin ($\vec{S}$). The EDM is also asymmetric under T. Using the polar molecule thorium monoxide (ThO), we measure $d_e = (-2.1 \pm 3.7_{\rm stat} \pm 2.5_{\rm syst}) \times 10^{-29}$ e cm. This corresponds to an upper limit of $|d_e| < 8.7 \times 10^{-29}$ e cm with 90 percent confidence, an order of magnitude improvement in sensitivity compared to the previous best limits. Our result constrains T-violating physics at the TeV energy scale.

The exceptionally high internal effective electric field ($\mathcal{E}_{\rm eff}$) of heavy neutral atoms and molecules can be used to precisely probe

is prepared using optical pumping and state preparation lasers. Parallel electric ($\vec{\mathcal{E}}$) and magnetic ($\vec{\mathcal{B}}$) fields exert torques on the electric and magnetic dipole moments, causing the spin vector to precess in the xy plane. The precession angle is measured with a readout laser and fluorescence detection. A change in this angle as $\vec{\mathcal{E}}_{\rm eff}$ is reversed is proportional to d_e.



Science **6168** (2014) *269*

Theory	$E_{\rm eff}[\frac{\rm GV}{\rm cm}]$	$W_{P,T}$[kHz]
2c-CCSD(T)[18]	81.5	112
4c-MR-CISD[19]	75.2	105

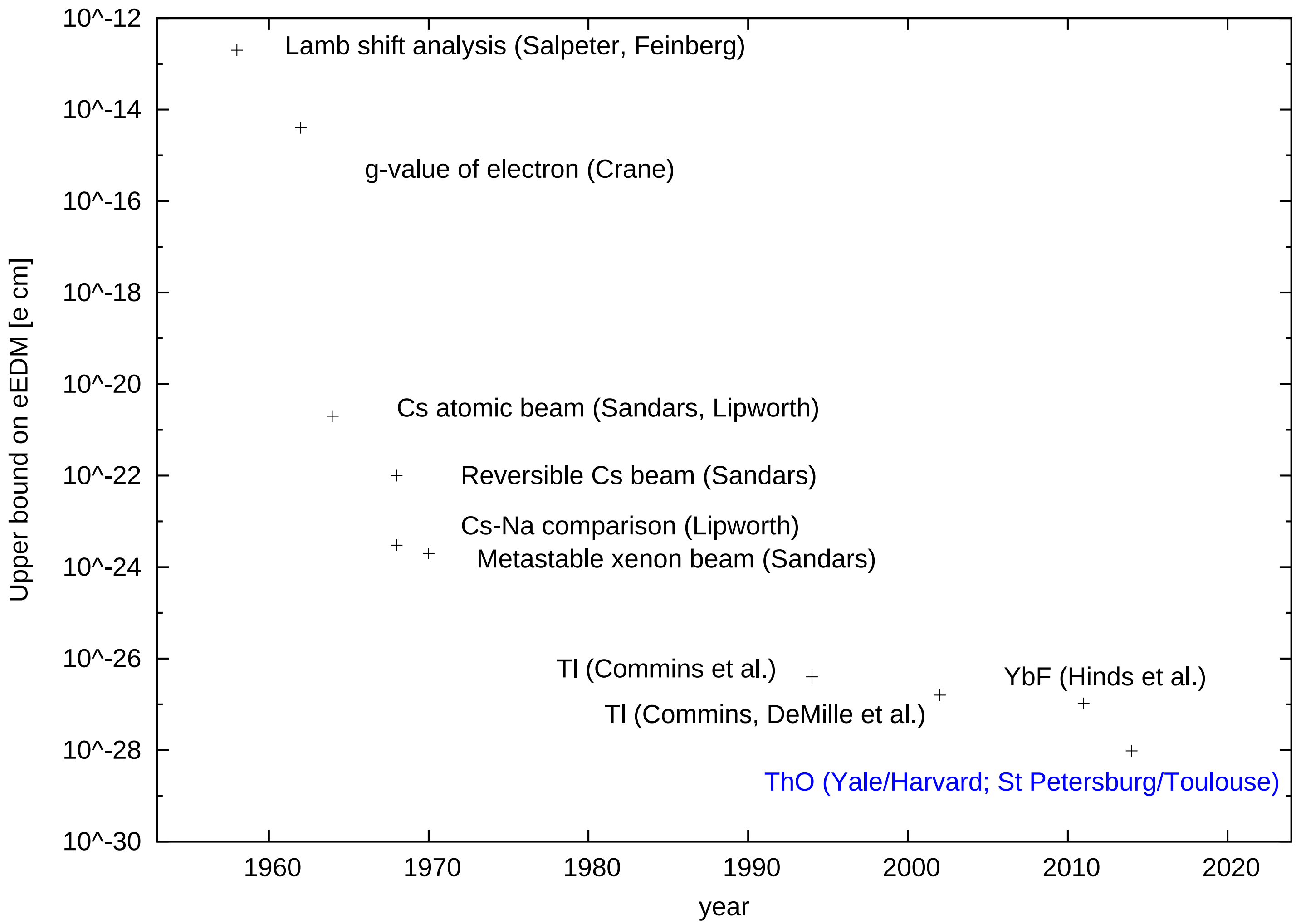
Variation due to spinor choice **0.2%**

Augmentation of virtual space: **further slight decrease**

[18] L. Skripnikov, A.V. Titov, *J Chem Phys* **142** (2015) *024301*

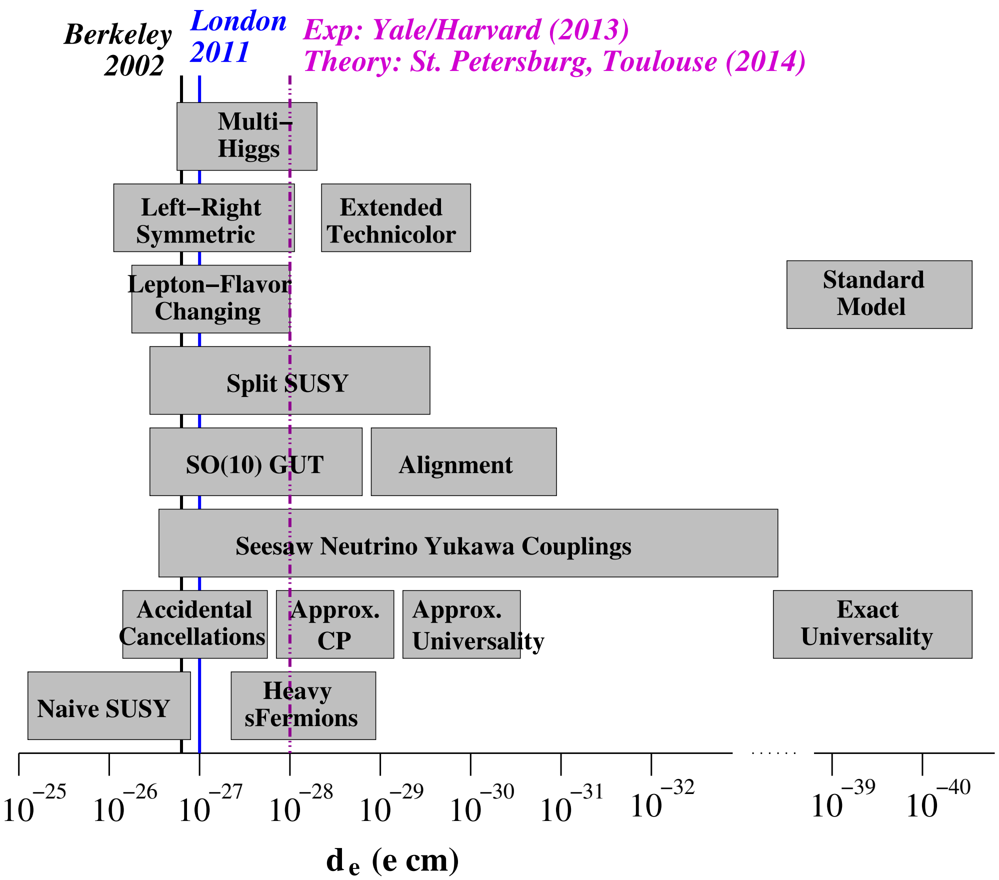
[19] TF, M.K. Nayak, *J Mol Spectrosc* **300** (2014) *16*; M. Denis, TF (2015)

Historical Development of eEDM Upper Bound[20]



[20] Sandars (1975), Commins, DeMille (2008)

eEDM Constraint on Beyond-Standard-Model Theories[21]



Model	$\lvert d_e\rvert[e\cdot cm]$
Standard model	$< 10^{-38}$
Left-right symmetric	$10^{-28}\ldots 10^{-26}$
Lepton-flavor changing	$10^{-29}\ldots 10^{-26}$
Multi-Higgs	$10^{-28}\ldots 10^{-27}$
Supersymmetric	$\leq 10^{-25}$
Experimental limit (Tl)[22]	$< 1.6\cdot 10^{-27}$
Experimental limit (YbF)[23]	$< 10.5\cdot 10^{-28}$
Experimental limit (ThO)[24]	$< 9.6\cdot 10^{-29}$

[21] Courtesy: DeMille (2005), Huliyar (2009)

[22] B.C. Regan, E.D. Commins, C.J. Schmidt, D.P. DeMille, *Phys Rev Lett* **88** (2002) *071805/1*

[23] J.J. Hudson, D.M. Kara, I.J. Smallman, B.E. Sauer, M.R. Tarbutt, E.A. Hinds, *Nature* **473** (2011) *493*

[24] D. DeMille, ICAP 2014, Washington D.C., ACME Collaboration, *Science* **6168** (2014) *269*, TF and M. K. Nayak, *J. Mol. Spectrosc.* **300** (2014) *16*, L. V. Skripnikov, A. N. Petrov, A. V. Titov, *J. Chem. Phys.* **139** (2013) *221103*, L. V. Skripnikov, A. V. Titov, *J. Chem. Phys.* **142** (2015) *024301*

Molecular (cat)ions

HfF^+/ThF^+

Experiment: JILA, Boulder, Colorado (Cornell group)

EDM Studies in Molecular Ions

as opposed to neutral molecules[25]

- Valence isoelectronic with neutral contenders (ThO, WC, *et al.*)
- Sufficiently large value of E_{eff}
  Hope for very large value[26] in ThF^+ due to $Z = 90$
- Use of ion traps and rotating electric fields
  $\Rightarrow$ Long interrogation times
- A related point:
  HfF^+ electronic ground state: ${}^1\Sigma_0^+$
  ThF^+ electronic ground state[27]: ${}^3\Delta_1$ **or** ${}^1\Sigma_0^+$

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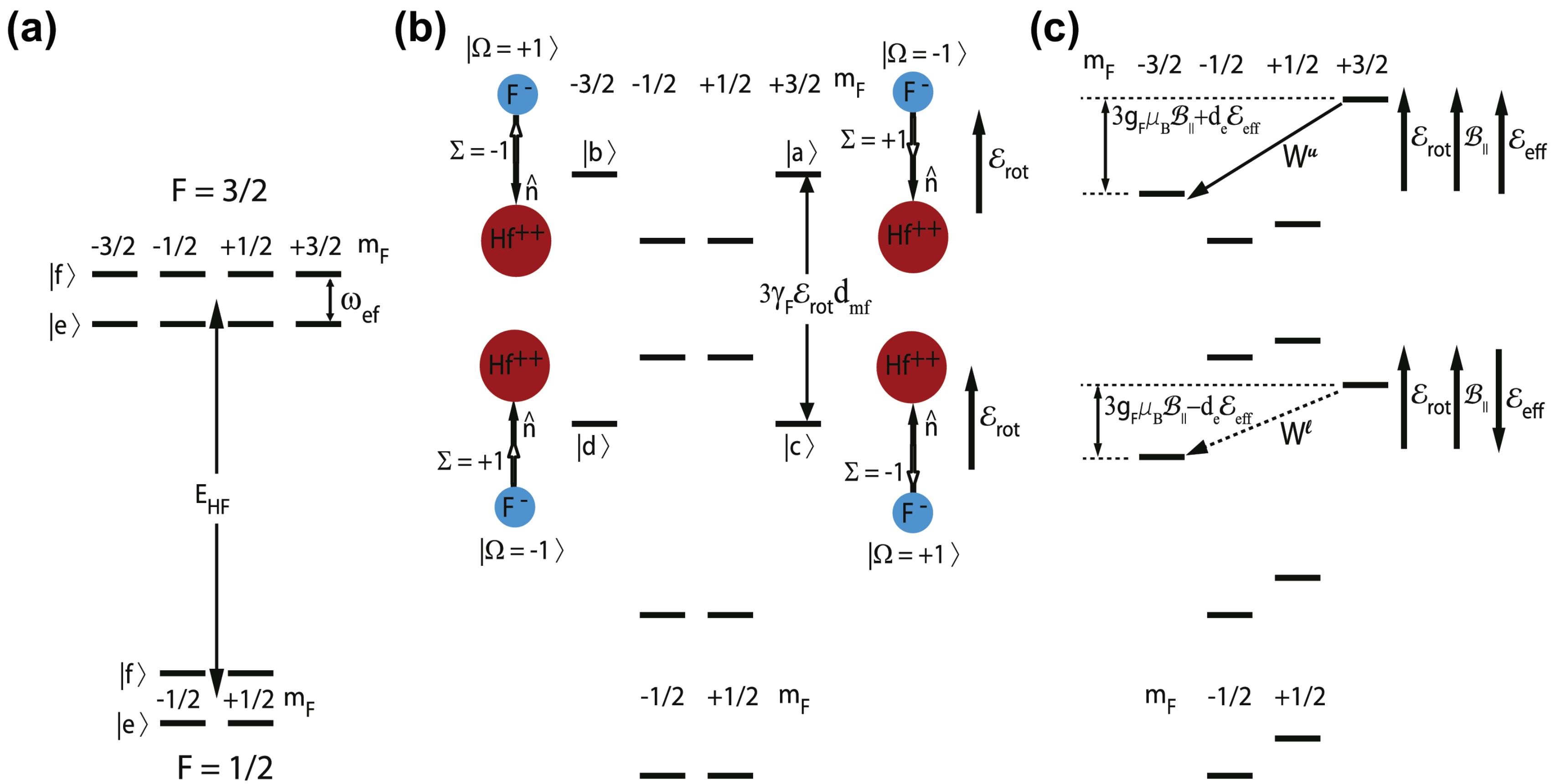
[25] H. Loh, K.C. Cossel, M.C. Grau, K.-K. Ni, E.R. Meyer, J.L. Bohn, J. Ye, E.A. Cornell, *Science* **342** (2013) *1220*
A.E. Leanhardt, J.L. Bohn, H. Loh, P. Maletinsky, E.R. Meyer, L.C. Sinclair, R.P. Stutz, E.A. Cornell, *J Mol Spectrosc* **270** (2011) *1*

[26] E.R. Meyer, J.L. Bohn, *Phys Rev A* **78** (2008) *010502(R)*

[27] M. Denis, M.N. Pedersen, H.J.Aa. Jensen, A.S.P. Gomes, M.K. Nayak, S. Knecht, TF, *New J Phys* (2015) **7** (2015) 043005
B. Barker, I.O. Antonov, M.C. Heaven, K.A. Peterson, *J Chem Phys* **136** (2012) *104305*

The eEDM in a molecular framework

A Proposed Measurement[28] on HfF$^+$



$$W^u(B) - W^u(-B) = 2d_e E_{\rm eff}$$

[28] A.E. Leanhardt, J.L. Bohn, H. Loh, P. Maletinsky, E.R. Meyer, L.C. Sinclair, R.P. Stutz, E.A. Cornell, *J Mol Spectrosc* **270** (2011) *1*

Molecular Wavefunctions from CC and CI

	Model	Th spinor distribution on spaces				
		4f5s5p	5d	6s6p	5f6d7s	7p7d8s8p6f
IHFSCC	$\mathcal{I}^{\mathcal{CC}}$	frozen	frozen	Q	P_m	P_i
	$\mathcal{II}^{\mathcal{CC}}$	frozen	Q	Q	P_m	P_i
	$\mathcal{III}^{\mathcal{CC}}$	Q	Q	Q	P_m	P_i
MRCI	$\mathcal{I}^{\mathcal{CI}}$	frozen	$Q-S$	$Q-S$	P_m	$Q-SD$
	$\mathcal{II}^{\mathcal{CI}}$	frozen	$Q-SD$	$Q-SD$	P_m	$Q-SD$

Model	Th $6s$,$6p$ F $2s$,$2p$	Th $7s$,$6d\delta$	Th $6d\pi$	Th $6d\sigma$,$7p\pi$	Th $7p\sigma$,$8s$	< 10 a.u
$\mathcal{III}^{\mathcal{CI},3}$	$Q-SD$	P_m	$Q-SD$	$Q-SD$	$Q-SD$	$Q-SD$
$\mathcal{III}^{\mathcal{CI}+T,3}$	$Q-SD$	P_m	$Q-SDT$	$Q-SDT$	$Q-SDT$	$Q-SDT$
$\mathcal{III}^{\mathcal{CI},5}$	$Q-SD$	P_m	P_m	$Q-SD$	$Q-SD$	$Q-SD$
$\mathcal{III}^{\mathcal{CI},8}$	$Q-SD$	P_m	P_m	P_m	$Q-SD$	$Q-SD$
$\mathcal{III}^{\mathcal{CI},10}$	$Q-SD$	P_m	P_m	P_m	P_m	$Q-SD$
$\mathcal{IV}^{\mathcal{CI}}$	frozen	P_m	P_m	P_m	P_m	$Q-SD$

Low-Lying Electronic States[29] of ThF^+

Method	Model[a]	Hamiltonian	Electronic state energy $^1\Sigma^+_{0+}$	$^3\Delta_1$	$^3\Delta_2$	$^3\Delta_3$
IHFSCC	$\mathcal{II}^{\mathcal{CC}}$	2c	42	0.00	1062	3146
	$\mathcal{III}^{\mathcal{CC},\dagger}$	2c	15	0	1062	3149
	$\mathcal{III}^{\mathcal{CC},\ddagger}$	2c	191	0	1048	3157
	$\boldsymbol{\mathcal{III}^{\mathcal{CC},*}}$	**2c**	**319**	**0**	**1039**	**3162**
MRCI	$\mathcal{I}^{\mathcal{CI}}$	2c	854	0	1154	3189
	$\mathcal{II}^{\mathcal{CI}}$	2c	630	0	1167	2986
	$\boldsymbol{\mathcal{III}^{\mathcal{CI},10}}$	**4c**	**538**	**0**	**1155**	**3012**
CCSD(T)+SO[b]			501	0	890	2157
CCSDT+SO[b]			143	0	890	2157
CCSDT(Q)+SO[b]			0	66	955	2223
Experiment (Heaven et al.)[b]			0	315.0(5)	1052.5(5)	3150(15)
Experiment (Cornell et al.)[c]			314.0(2)	0		

Spinor-based correlation methods yield **similar results**

Orbital-based perturbative methods **underestimate** $^3\Delta$ **splittings**

[29][a] M. Denis, M.N. Pedersen, H.J.Aa. Jensen, A.S.P. Gomes, M.K. Nayak, S. Knecht, TF, *New J Phys* **7** (2015) *043005*

[b] B. Barker, I.O. Antonov, M.C. Heaven, K.A. Peterson, *J Chem Phys* **136** (2012) *104305*

[c] D.N. Gresh, K.C. Cossel, Y. Zhou, J. Ye, E.A. Cornell, (2015) *unpublished manuscript*

(Four-)Spinors vs. Orbitals

The electronic ground state of of ThF^+

Hypothesis: Orbital-based correlation methods underestimate the splitting $\Delta\varepsilon_{\delta_{5/2}-\delta_{3/2}} = 2166\ \mathrm{cm}^{-1}$.

Configurational composition of ${}^3\Delta$ multiplet states (from MR_{10}-CISD(18) model)

${}^3\Delta_1$	$89\%\ \sigma^1_{-1/2}\,\delta^1_{3/2}$
${}^3\Delta_2$	$61\%\ \sigma^1_{1/2}\,\delta^1_{3/2}$, $28\%\ \sigma^1_{-1/2}\,\delta^1_{5/2}$
${}^3\Delta_3$	$89\%\ \sigma^1_{1/2}\,\delta^1_{5/2}$

Orbital-based methods underestimate term splittings

Error is large for ${}^3\Delta_2$-${}^3\Delta_3$ splitting
Error is smaller for ${}^3\Delta_1$-${}^3\Delta_2$ splitting

Suggested explanation for differing ground-state predictions

^{19}F Magnetic Hyperfine Interaction in ThF^+ and HfF^+ $(\Omega = 1)$

Magnetic hyperfine interaction constant:

$$A_{||} = \frac{\mu_F}{I\Omega} \left\langle \sum_{i=1}^{n} \left(\frac{\vec{\alpha}_i \times \vec{r}_{iF}}{r_{iF}^3} \right)_z \right\rangle_\psi$$

System	Model	$A_{\vert\vert}$[MHz]	spinor character	R_e [a.u.]
ThF^+	MR_{10}-CISD(20)	**8.9**	0.001 p_z(F)	3.75
([30])	MR_{10}-CISD(18)	4.3		
HfF^+ ([31])	MR_6-CISD(20)	**45.3**	0.001 p_z(F)	3.41

- Unpaired electrons localized on heavy atom
- Correlation of $1s$ (F) electrons of crucial importance
- $A_{||}$ for ThF^+ very small due to long internuclear distance

[30] M. Denis, M.N. Pedersen, H.J.Aa. Jensen, A.S.P. Gomes, M.K. Nayak, S. Knecht, TF, *New J Phys* (2015) **7** (2015) 043005

[31] TF and M.K. Nayak, *Phys Rev A* **88** (2013) *032514*

$\mathcal{P},\mathcal{T}$-Odd Interactions in ThF$^+$ $(\Omega = 1)$

Basis Sets

Basis set	$T_v[\text{cm}^{-1}]$	$E_{\text{eff}}[\frac{\text{GV}}{\text{cm}}]$	$A_{\vert\vert}$[MHz]	$W_{P,T}$[kHz]
DZ	378	37.8	1824	51.90
TZ'	787	36.9	1836	50.73
QZ'	877	36.9	1830	50.77

Vertical excitation energy for $\Omega = 0^+$, electron EDM effective electric field, magnetic hyperfine interaction constant, and scalar-pseudoscalar electron-nucleon interaction constant for $\Omega = 1$ at an internuclear distance of $R = 3.779\ a_0$ using basis sets with increasing cardinal number and the wavefunction model $\mathcal{III}^{\mathcal{CI},5}$.

Scalar-pseudoscalar electron-nucleon interaction constant:

$$W_{P,T} = \frac{\imath}{\Omega}\frac{G_F}{\sqrt{2}} Z \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \rho_N(\vec{r}_j) \right\rangle_\psi$$

The eEDM in ThF^+ ($\Omega = 1$)

Active 4-Spinor Spaces

CI model(TZ basis)	$T_v[\mathrm{cm}^{-1}]$	$E_{\mathrm{eff}}[\frac{\mathrm{GV}}{\mathrm{cm}}]$	$A_{\|\|}$[MHz]	$W_{P,T}$[kHz]
$\mathcal{IV}^{\mathcal{CI}}$	274	35.4	1749	49.44
$\mathcal{III}^{\mathcal{CI},3}$	1029	47.5	1842	65.78
$\mathcal{III}^{\mathcal{CI},5}$	787	36.9	1836	50.73
$\mathcal{III}^{\mathcal{CI},6}$	709	36.2	1836	49.90
$\mathcal{III}^{\mathcal{CI},8}$	598	35.6	1834	49.04
$\boldsymbol{\mathcal{III}^{\mathcal{CI},10}}$	538	**35.2**	**1833**	**48.35**
$\mathcal{III}^{\mathcal{CI},12}$		35.1	1832	

Vertical excitation energy for $\Omega = 0^+$, electron EDM effective electric field, magnetic hyperfine interaction constant, and scalar-pseudoscalar electron-nucleon interaction constant for $\Omega = 1$ at an internuclear distance of $R = 3.779\ a_0$ using the TZ' basis set, varying number of correlated electrons and varying active spinor spaces.

- Large active space $\Rightarrow$ shifts electron density from Th(s) to Th(p) and Th(d), reducing E_{eff}.

The eEDM in ThF^+ $(\Omega = 1)$

Higher Excitations

CI model(DZ basis)	$T_v[\mathrm{cm}^{-1}]$	$E_{\mathrm{eff}}[\frac{\mathrm{GV}}{\mathrm{cm}}]$	$A_{\|\|}$[MHz]	$W_{P,T}$[kHz]
$\mathcal{III}^{\mathcal{CI},3}$	654	47.0	1830	64.92
$\mathcal{III}^{\mathcal{CI},10}$	88	37.1	1832	51.06
$\mathcal{III}^{\mathcal{CI}+T,3}$	247	35.4	1834	48.64

Vertical excitation energy for $\Omega = 0^+$, electron EDM effective electric field, magnetic hyperfine interaction constant, and scalar-pseudoscalar electron-nucleon interaction constant for $\Omega = 1$ at an internuclear distance of $R = 3.779\ a_0$ using the DZ basis set and varying maximum excitation rank.

- Active space accounts for important higher excitations

ThF^+

Static Molecular Electric Dipole Moment

${}^M\Lambda_\Omega$ State	T_v [cm^{-1}]	$\left\langle {}^M\Lambda_\Omega \vert \hat{D}_z \vert {}^M\Lambda_\Omega \right\rangle$ [D]
${}^1\Sigma_0^+$	630	3.941
${}^3\Delta_1$	0	4.029
${}^3\Delta_2$	1167	3.970
${}^3\Delta_3$	2986	4.034

Molecular static electric dipole moments $\left\langle {}^M\Lambda_\Omega \vert \hat{D}_z \vert {}^M\Lambda_\Omega \right\rangle$, with $\hat{\vec{D}}$ the electric dipole moment operator, using the TZ basis set and the CI model $\mathcal{II}^{\mathcal{CI}}$. The origin is at the center of mass, and the internuclear distance is $R = 3.779$ $[a_0]$ (F nucleus at $z\vec{e}_z$ with $z < 0$).

- Very large center-of-mass dipole moment
  Effectively polarizable, suggest large value of E_{eff}

ThF^+

Electric Transition Dipole Moments

${}^{M}\Lambda_{\Omega}$ State	T_v [cm^{-1}]	${}^{1}\Sigma_0^+$	${}^{3}\Delta_1$	${}^{3}\Delta_2$	${}^{3}\Delta_3$	${}^{1}\Sigma_0({}^{3}\Pi_0)$	${}^{3}\Pi_0$	${}^{1,3}\Pi_1({}^{3}\Sigma_1)$	${}^{3}\Pi_0({}^{1}\Sigma_0)$
${}^{1}\Sigma_0^+$	274	-4.004							
${}^{3}\Delta_1$	0	0.012	-4.075						
${}^{3}\Delta_2$	724	0.000	0.070	-4.022					
${}^{3}\Delta_3$	2198	0.000	0.000	0.052	-4.075				
${}^{1}\Sigma_0({}^{3}\Pi_0)$	6344	0.439	0.455	0.000	0.000	-3.752			
${}^{3}\Pi_0$	6528	0.000	0.571	0.000	0.000	0.000	-2.116		
${}^{1,3}\Pi_1({}^{3}\Sigma_1)$	6639	0.868	0.142	0.218	0.000	0.197	0.000	-2.375	
${}^{3}\Pi_0({}^{1}\Sigma_0)$	6747	0.003	0.391	0.000	0.000	0.929	0.000	0.094	-2.717
${}^{1,3}\Delta_2({}^{3}\Pi_2)$	7008	0.000	0.473	0.334	0.298	0.000	0.000	0.529	0.000
${}^{3}\Sigma_1$	7490	0.226	0.069	0.221	0.000	0.136	0.197	0.451	0.145
${}^{1,3}\Pi_1$	7918	0.667	0.052	0.801	0.000	0.011	0.064	0.107	0.043
${}^{3}\Phi_2({}^{3}\Pi_2)$	8245	0.000	1.338	0.234	0.272	0.000	0.000	0.134	0.000

Electric transition dipole moments $\left|\left|\left\langle {}^{M}\Lambda'_{\Omega}|\hat{\vec{D}}|{}^{M}\Lambda_{\Omega}\right\rangle\right|\right|$, with $\hat{\vec{D}}$ the electric dipole moment operator, and vertical transition energies for low-lying electronic states in [D] units using the TZ' basis set and the CI model $\mathcal{IV}^{\mathcal{CI}}$. The origin is at the center of mass, and the internuclear distance is $R = 3.779$ $[a_0]$. $\left({}^{M}\Lambda_{\Omega}\right)$ denotes a term contributing at least 10% to the state. ^{1,3} denotes cases where Λ-S coupling breaks down significantly according to the analysis of our spinor-based ω-ω coupled wavefunctions.

HfF$^+$ and ThF$^+$: E_{eff} in the $\Omega = 1$ science state[32]

HfF$^+$		ThF$^+$	
Model	E_{eff} $\left[\frac{\text{GV}}{\text{cm}}\right]$	Model	E_{eff} $\left[\frac{\text{GV}}{\text{cm}}\right]$
CAS-CI(10)	24.1		
MR-CISD(10)	22.4		
MR-CISD(20)	23.3	MR_3-CISD(18)	47.5
MR-CISD+T(20)	23.7	MR_6-CISD(18)	36.2
MR-CISD(34)	22.9	MR_{10}-CISD(18)	35.2
MR-CISD(34)+T	23.3	MR_3-CISDT(18)	35.4
Estimate, Meyer et al.[33]	≈ 30	Meyer et al.	≈ 90
20 e^- corr., Titov et al.[34]	24.2	38 e^- corr., Titov et al.[35]	≈ 37.3

(HfF$^+$)

Similar results with various methods
System currently under exp. study

(ThF$^+$)

Meyer's model inaccurate
CC and CI approaches yield similar results

[32] TF and M.K. Nayak, *Phys Rev A* **88** (2013) *032514*
M. Denis, M. K. Nørby, H. J. Aa. Jensen, A. S. P. Gomes, M.K. Nayak, S. Knecht, TF, *New J Phys* **7** (2015) *043005*
[33] E.R. Meyer, J.L. Bohn, *Phys Rev A* **78** (2008) *010502(R)*
[34] A.N. Petrov, N.S. Mosyagin, T.A. Isaev, A.V. Titov, *Phys Rev A* **76** (2007) *030501(R)*
[35] L. V. Skripnikov, A.V. Titov, *arXiv:1503.01001v1* (2015)

Nuclear Magnetic Quadrupole Moment
TaN

Constraining $\mathcal{P}$,$\mathcal{T}$-violating hadron physics

- Nuclear MQM has two possible sources[36]:
  1. Intranuclear $\mathcal{P}$-,$\mathcal{T}$-odd interactions, described by QCD $(\mathcal{CP})$-violating parameter[37] $\tilde{\Theta}$,
  $$M_0^{p,n}(\tilde{\Theta}) \approx 2 \times 10^{-29}\,\tilde{\Theta}\,e\,\text{cm}^2$$
  M: valence nucleon MQM
  2. Neutron/proton EDM (order of magnitude smaller)

- MQM is enhanced in non-spherical (deformed) nuclei[38]

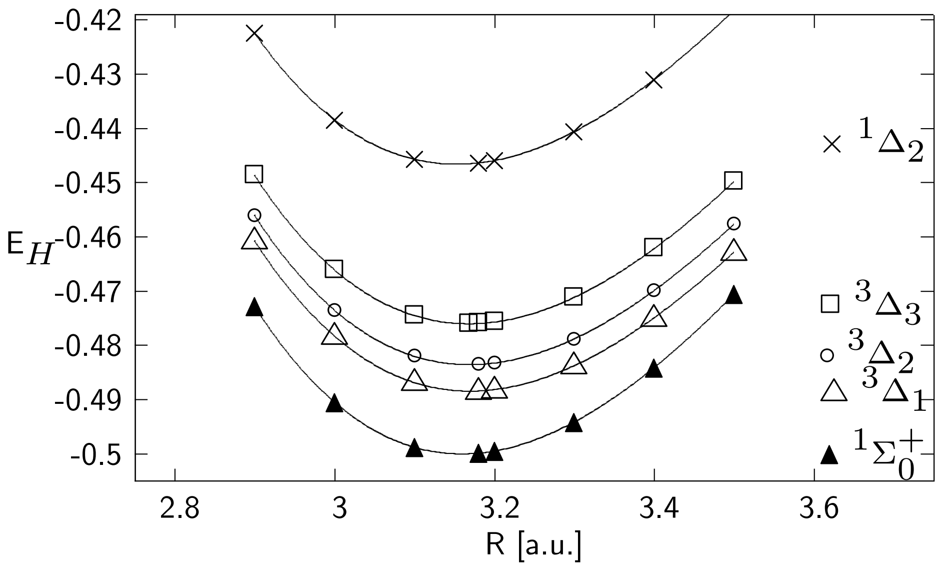
- Enhancement[41] of ≈ 12 in ^{181}Ta, compared to $M_0^{p,n}$

- TaN is a "$^3\Delta$ molecule", experiments planned at ACME (Yale/Harvard)

[36] V. V. Flambaum, D. DeMille, M. G. Kozlov, *Phys Rev Lett* **113** (2014) *103003*
[37] R. J. Crewther, P. Di Vecchia, G. Veneziano, E. Witten, *Phys Lett* **88B** (1979) *123*
[38] V. V. Flambaum, *Phys Lett B* **320** (1994) *211*

TaN, Spectroscopic properties

State	Model	R_e [a.u.]	ω_e [cm^{-1}]	B_e [cm^{-1}]	T_e [cm^{-1}]
	av. DCHF	3.115	1163	0.477	0
$^1\Sigma_0^+$	MR$_{12}$-CISD(10)	3.160	1161	0.464	0
	Exp.[39]	3.181	1070		0.0
$^3\Delta_1$	MR$_{12}$-CISD(10)	3.170	1116	0.461	2526
	Exp.[39]	3.196			2827.2917
$^3\Delta_2$	MR$_{12}$-CISD(10)	3.169	1117	0.461	3618
$^3\Delta_3$	MR$_{12}$-CISD(10)	3.168	1119	0.462	5276
$^1\Delta_2$	MR$_{12}$-CISD(10)	3.153	1123	0.466	11729



- Strongly bound molecule
- Low-lying “science state” $^3\Delta_1$ (long lifetime)
- Large mol. dipole moment

[39] M. Zhou, L. Andrews, *J Phys Chem A* **102** (1998) *9061*; R. S. Ram, J. Liévin, P. F. Bernath, *J Mol Spectrosc* **215** (2002) *275*

TaN, Spectroscopic properties

${}^M\Lambda_\Omega$ State	${}^1\Sigma_0^+$	${}^3\Delta_1$	${}^3\Delta_2$	${}^3\Delta_3$	${}^1\Delta_2$
${}^1\Sigma_0^+$	-3.515				
${}^3\Delta_1$	0.028	-4.809			
${}^3\Delta_2$	0.000	0.085	-4.775		
${}^3\Delta_3$	0.000	0.000	0.087	-4.776	
${}^1\Delta_2$	0.000	0.139	0.114	0.164	-4.000

Molecular static electric dipole moments $\left\langle {}^M\Lambda_\Omega|\hat{D}_z|{}^M\Lambda_\Omega\right\rangle$, transition dipole moments $\left\|\left\langle {}^M\Lambda'_\Omega|\hat{\vec{D}}|{}^M\Lambda_\Omega\right\rangle\right\|$, with $\hat{\vec{D}}$ the electric dipole moment operator (both in [D] units) at $R = 3.1806$ a$_0$, using the model MR$_{12}^{+T}$-CISD(10)

- Large molecular electric dipole moment in ${}^3\Delta_1$ science state
- $\approx 3\%$ non-Δ character of science state
  Transition to ${}^1\Sigma_0^+$ borrows intensity via ${}^3\Delta_1 - {}^1\Pi_1$ and other second-order spin-orbit couplings

Molecular Nuclear Magnetic Quadrupole Moment

Results for ^{181}TaN, $\Omega = 1$

Cutoff/CI Model	E_{eff} $\left[\frac{\text{GV}}{\text{cm}}\right]$	$A_{\vert\vert}$ [MHz]	$W_{P,T}$ [kHz]	W_M $\left[\frac{10^{33}\text{Hz}}{e\,\text{cm}^2}\right]$
vTZ-30a.u./MR_{12}-CISD(10)	30.1	−3104	27.4	0.633
vTZ-30a.u./MR_{12}-CISDT(10)	29.7	−3092	27.1	0.626
vTZ-30a.u./MR_{12}-CISD(18)	33.6	−3059	30.5	0.718
vTZ-30a.u./MR_{12}^{+T}-CISD(10)	31.4	−3067	28.7	0.645
vTZ-30a.u./MR_{12}^{+T}-CISD(18)	35.1	−3025	31.9	0.737
vTZ-30a.u./MR'_{12}-CISD(26)	34.2	−3237	31.1	0.732
Skripnikov et al.[40]	34.9	−3132	31	1.08
Flambaum *et al.*[41]	25(YbF)			≈ 1

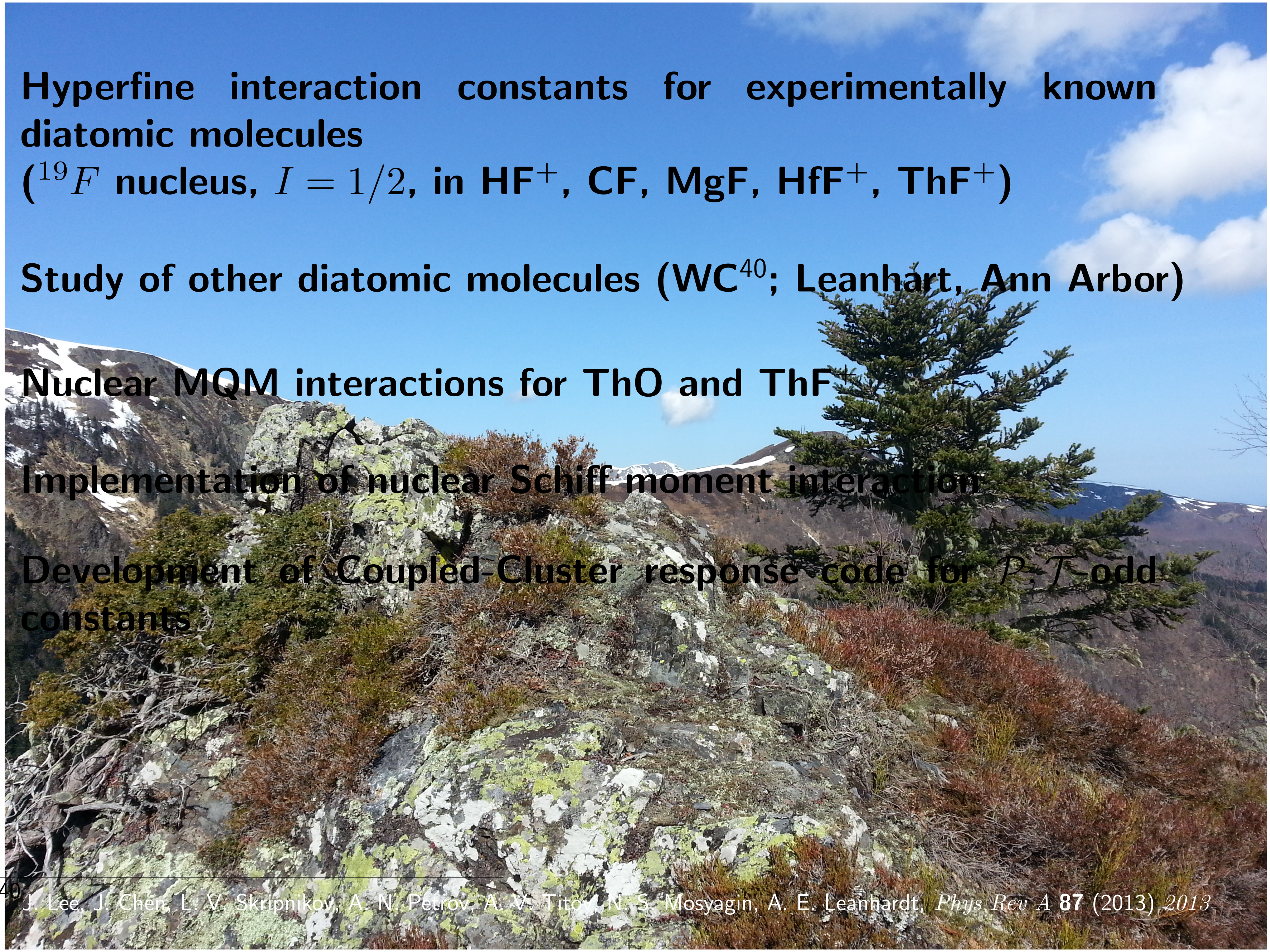
$\mu(^{181}\text{Ta}) = 2.35\mu_N \qquad I = \frac{7}{2}$

- EDM effective field (and $W_{P,T}$) sufficiently large
- NMQM interaction constant W_M smaller than reference values

[40] N. S. Mosyagin, M. G. Kozlov, A. V. Titov, *J Phys B* **31** (1998) *L763*

[41] V. V. Flambaum, D. DeMille, M. G. Kozlov, *Phys Rev Lett* **113** (2014) *103003*

Outlook

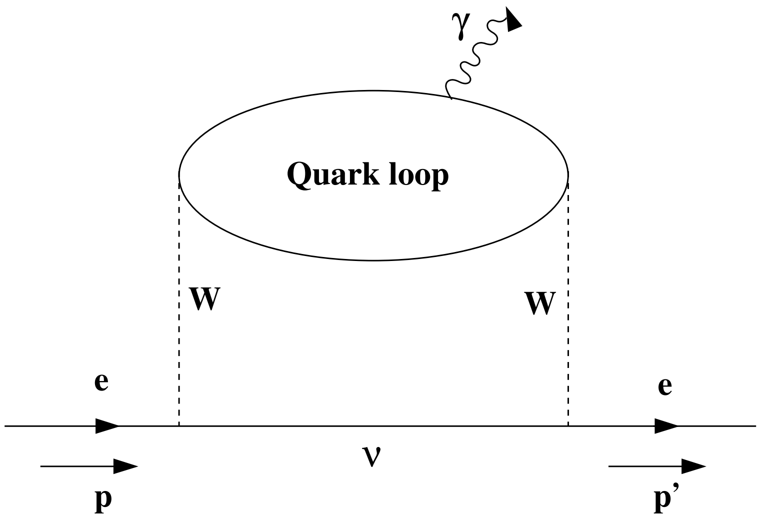


- Hyperfine interaction constants for experimentally known diatomic molecules (^{19}F nucleus, $I = 1/2$, in HF^+, CF, MgF, HfF^+, ThF^+)
- Study of other diatomic molecules (WC[40]; Leanhart, Ann Arbor)
- Nuclear MQM interactions for ThO and ThF^+
- Implementation of nuclear Schiff moment interaction
- Development of Coupled-Cluster response code for $\mathcal{P},\mathcal{T}$-odd constants

[40] J. Lee, J. Chen, L. V. Skripnikov, A. N. Petrov, A. V. Titov, N. S. Mosyagin, A. E. Leanhardt, *Phys Rev A* **87** (2013) *2013*

The induced fermion EDM

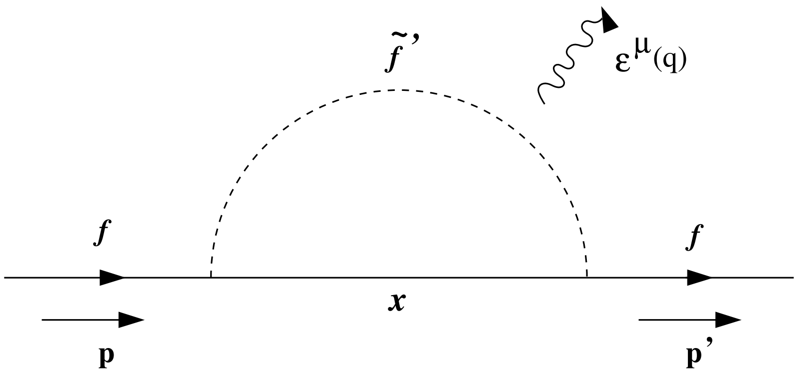
Standard Model Picture[7]



- Three-loop $\mathcal{CP}$-odd contributions zero in the absence of gluonic corrections[8]

$d_e^{SM} \leq 10^{-38}\, e$ cm

BSM Picture



χ: chargino, neutralino

$\tilde{f}'_j$: supersymmetry (s)-fermion

$\epsilon^\mu(q)$: photon

- MSSM ("naïve SUSY") prediction[9]:
  $d_e \leq 10^{-27}\, e$ cm

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[7] E.D. Commins, *Adv At Mol Opt Phys* **40** (1998) *1*

[8] M. Pospelov, I.B. Khriplovich, *Sov J Nuc Phys* **53** (1991) *638*

[9] J. Ellis, J.S. Lee, A. Pilaftsis, *J High Energy Phys* **10** (2008) *049*