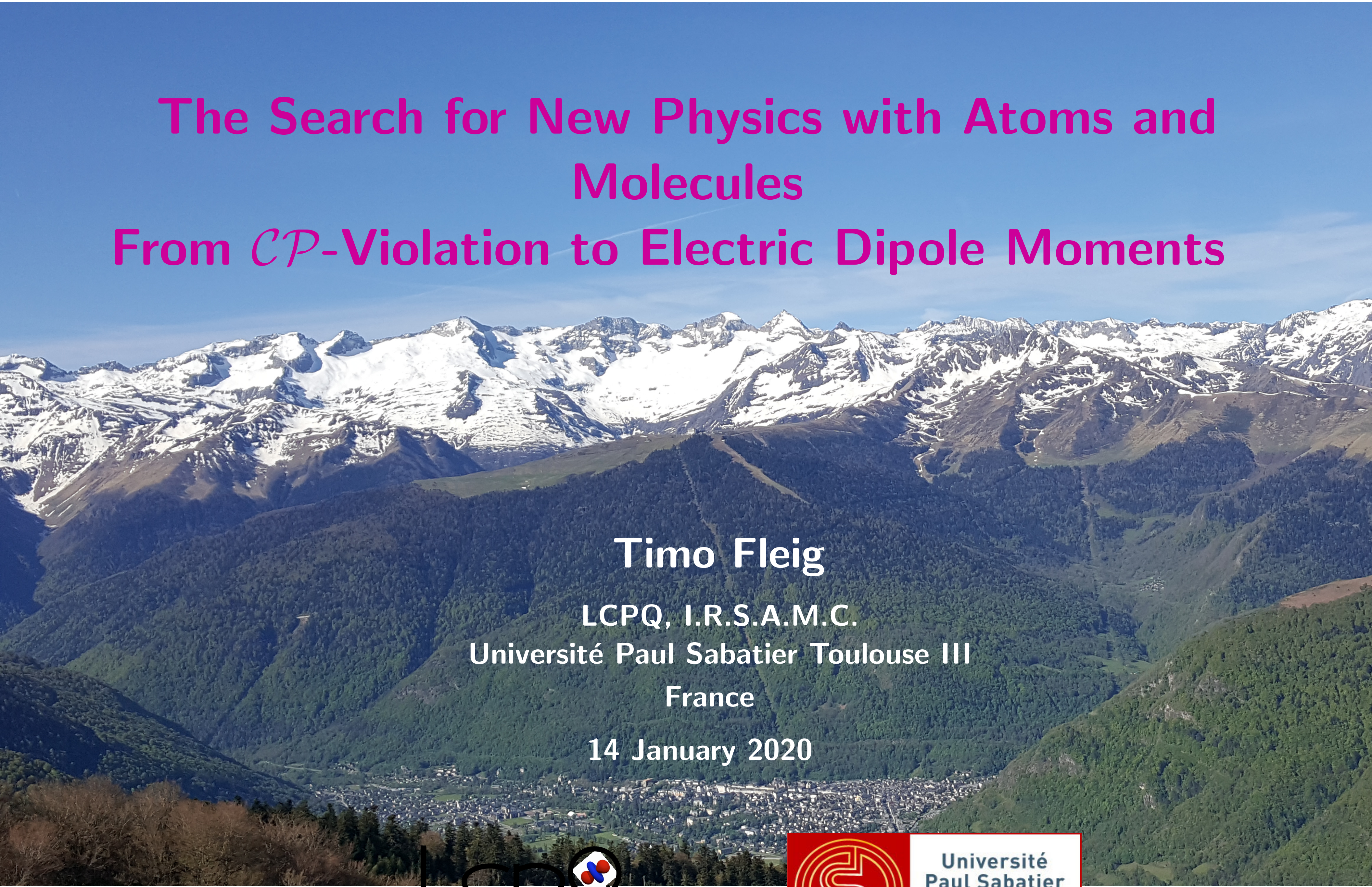

The Search for New Physics with Atoms and Molecules
From $\mathcal{CP}$-Violation to Electric Dipole Moments

Timo Fleig

LCPQ, I.R.S.A.M.C.
Université Paul Sabatier Toulouse III
France

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Laboratoire de Chimie et Physique Quantiques

Université Paul Sabatier TOULOUSE III

Outline

Problems (in a Nutshell)

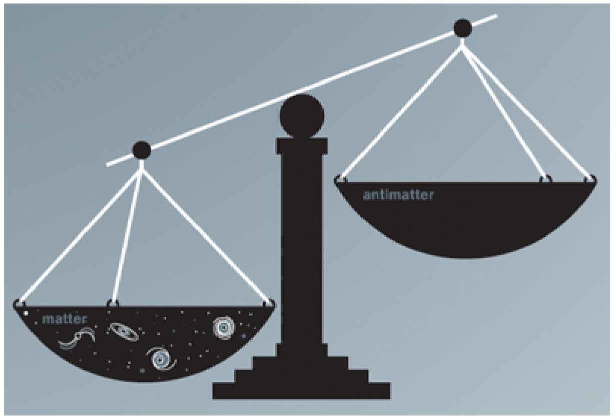
Relevant **particle physics**

Relevant **atomic physics**

Science (1) : Search for the **Electron EDM**

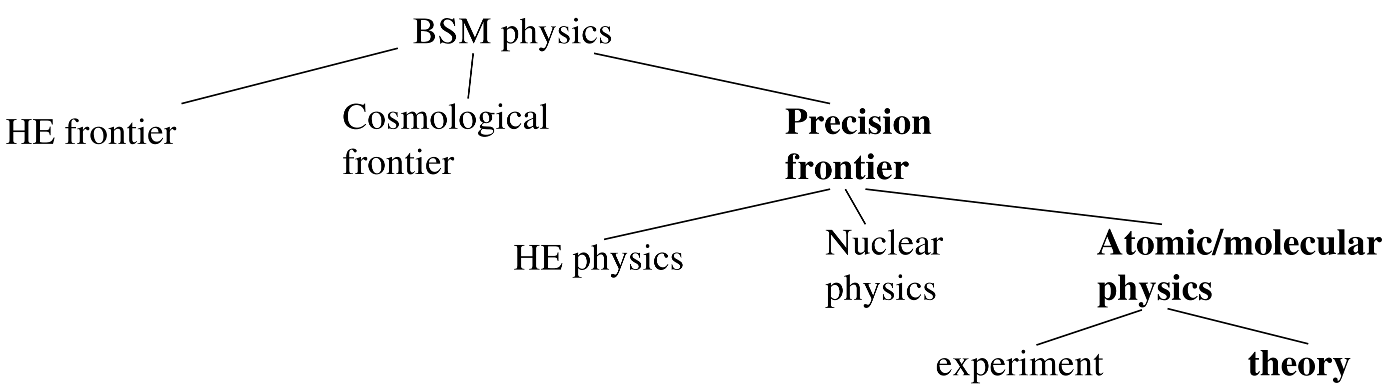
Science (2) : Atomic EDMs and **stronger constraints**

Questions Begging an Answer



- **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[3]

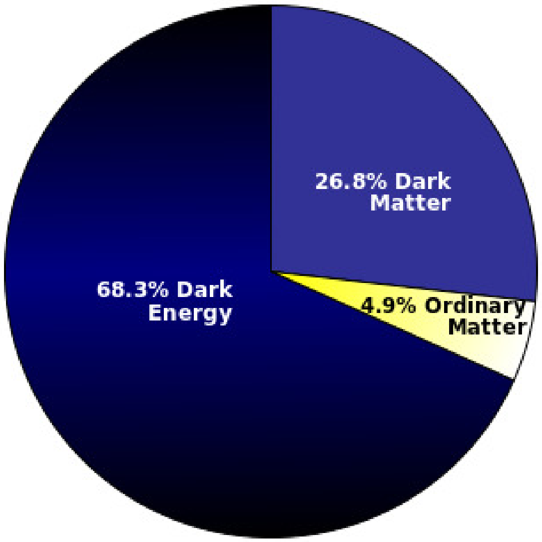


[1] M. Dine, A. Kusenko, *Rev. Mod. Phys.* **76** (2004) *1*

[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*
M. Safronova, D. Budker, D. DeMille, D.F. Jackson Kimball, A. Derevianko, C.W. Clark, *Rev. Mod. Phys.* **90** (2018) *025008*
T.E. Chupp, P. Fierlinger, M.J. Ramsey-Musolf, J.T. Singh, *Rev. Mod. Phys.* **91** (2019) *015001*

Energy Content and Baryon Asymmetry of the Universe (BAU)[4]



Dark energy: Why accelerated expansion? Cosmological constant?

Dark matter: Particle (LSP, axion)? Modification of gravity?

Ordinary matter: Existence contradicts SM prediction!

Evidence for the BAU:

$$Y_B = \frac{n_B - \overline{n}_B}{S} \approx \frac{n_B}{S} = \begin{cases} (7.3 \pm 2.5) \times 10^{-11} & \text{Big-Bang Nucleosynthesis (BBN)}^5 \\ (9.2 \pm 1.1) \times 10^{-11} & \text{(WMAP, exp.)}^6 \\ (8.59 \pm 0.11) \times 10^{-11} & \text{(Planck, exp.)}^7 \end{cases}$$

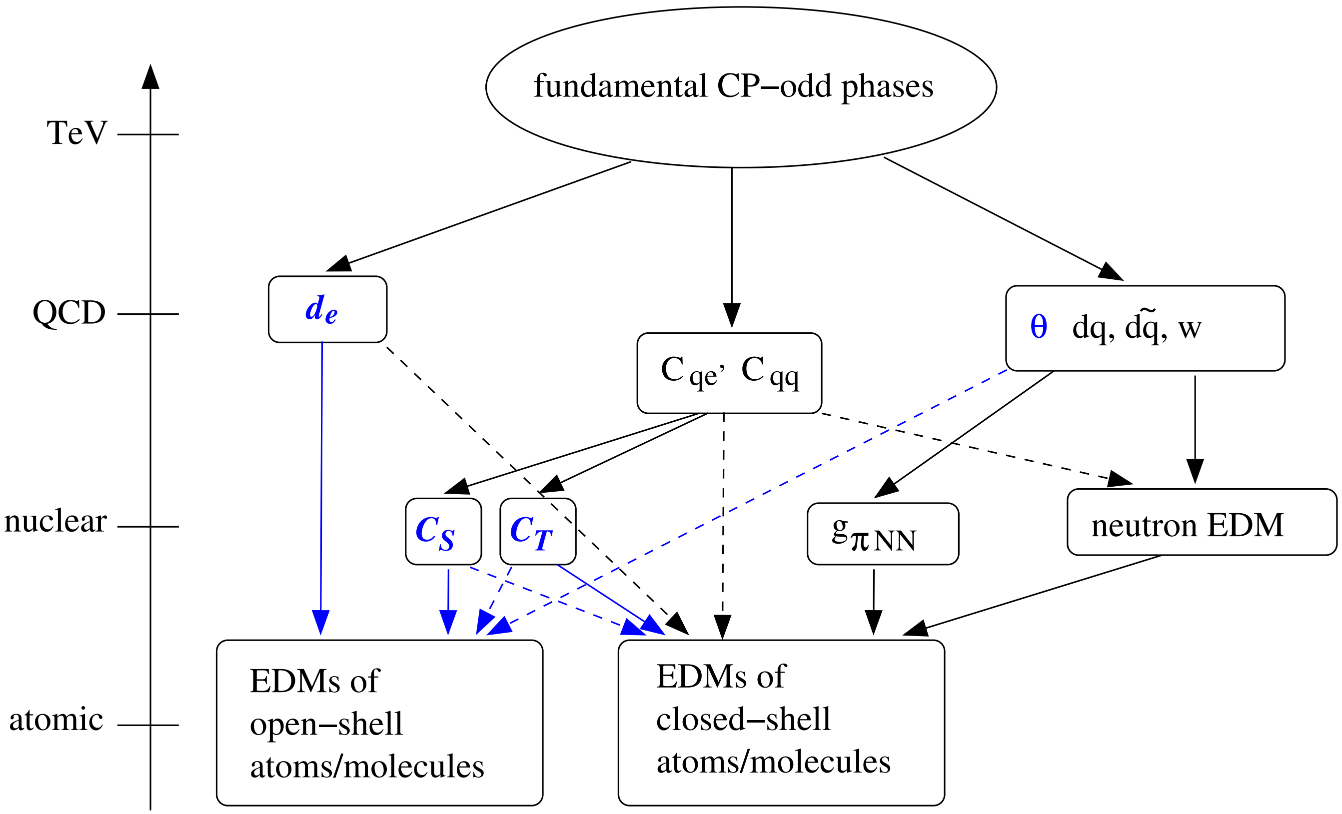
[4] G. A. White, *A Pedagogical Introduction to Electroweak Baryogenesis* **Morgan & Clay** (2016) *1*

[5] S. Eidelman *et al.*, *Rev. Part. Phys. Phys. Lett. B* **592** (2004) *1*

[6] D. N. Spergel *et al.*, *Astron. J. Suppl.* **148** (2003) *175*

[7] P. A. R. Ade *et al.*, *Astron. Astrophys.* **571** (2013) *A16*

Electric Dipole Moments and Their Source Tree[8]



$d_{e,q}$: (electron, quark) EDM

$C_{S,P,T}$: Coupling constants of $\mathcal{CP}$-odd electron-nucleon interaction

θ: QCD "theta" term

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

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

Fundamental Discrete Symmetries

(P) Violation

The fall of $\mathcal{P}$ invariance[9]; measuring helicity $\boldsymbol{\sigma} \cdot \hat{\mathbf{p}}$ in weak decays

$$\underset{\left(\substack{s_\mu \\ \rightarrow}\right)}{\mu^+} \longleftarrow \underset{s_\pi = 0}{\pi^+} \longrightarrow \underset{\left(\substack{s_\nu \\ \leftarrow}\right)}{\nu_\mu}$$

left-handed μ^+ left-handed ν_μ

$$\xrightarrow{\hat{\mathcal{P}}}$$

$$\underset{\left(\substack{s_\nu \\ \leftarrow}\right)}{\nu_\mu} \longleftarrow \underset{s_\pi = 0}{\pi^+} \longrightarrow \underset{\left(\substack{s_\mu \\ \rightarrow}\right)}{\mu^+}$$

right-handed ν_μ right-handed μ^+

$$\underset{\left(\substack{s_\mu \\ \leftarrow}\right)}{\mu^-} \longleftarrow \underset{s_\pi = 0}{\pi^-} \longrightarrow \underset{\left(\substack{s_{\overline{\nu}} \\ \rightarrow}\right)}{\overline{\nu}_\mu}$$

right-handed μ^- right-handed $\overline{\nu}_\mu$

$$\xrightarrow{\hat{\mathcal{P}}}$$

$$\underset{\left(\substack{s_{\overline{\nu}} \\ \rightarrow}\right)}{\overline{\nu}_\mu} \longleftarrow \underset{s_\pi = 0}{\pi^-} \longrightarrow \underset{\left(\substack{s_\mu \\ \leftarrow}\right)}{\mu^-}$$

left-handed $\overline{\nu}_\mu$ left-handed μ^-

right-handed μ^+ or left-handed μ^- never observed

$\Rightarrow$ right-handed ν_μ and left-handed $\overline{\nu}_\mu$ do not exist.

$\Rightarrow$ $\mathcal{P}$ *maximally violated* in weak processes.

[9] C. S. Wu et al., *Phys Rev* **105** (1957) *254*
G. Backenstoss et al., *Phys Rev Lett* **6** (1961) *415*
M. Bardon et al., *Phys Rev Lett* **7** (1961) *23*

Fundamental Discrete Symmetries

(CP) Conservation

Same weak decays under $(\hat{\mathcal{C}}\hat{\mathcal{P}})$ transformation:

$$\underset{\text{left-handed } \mu^+}{\mu^+ \binom{s_\mu}{\rightarrow}} \longleftarrow \underset{s_\pi = 0}{\pi^+} \longrightarrow \underset{\text{left-handed } \nu_\mu}{\nu_\mu \binom{s_\nu}{\leftarrow}}$$

$$\xrightarrow{\hat{\mathcal{C}}\hat{\mathcal{P}}}$$

$$\underset{\text{right-handed } \overline{\nu}_\mu}{\overline{\nu}_\mu \binom{s_{\overline{\nu}}}{\leftarrow}} \longleftarrow \underset{s_\pi = 0}{\pi^-} \longrightarrow \underset{\text{right-handed } \mu^-}{\mu^- \binom{s_\mu}{\rightarrow}}$$

$$\underset{\text{right-handed } \mu^-}{\mu^- \binom{s_\mu}{\leftarrow}} \longleftarrow \underset{s_\pi = 0}{\pi^-} \longrightarrow \underset{\text{right-handed } \overline{\nu}_\mu}{\overline{\nu}_\mu \binom{s_{\overline{\nu}}}{\rightarrow}}$$

$$\xrightarrow{\hat{\mathcal{C}}\hat{\mathcal{P}}}$$

$$\underset{\text{left-handed } \nu_\mu}{\nu_\mu \binom{s_\nu}{\rightarrow}} \longleftarrow \underset{s_\pi = 0}{\pi^+} \longrightarrow \underset{\text{left-handed } \mu^+}{\mu^+ \binom{s_\mu}{\leftarrow}}$$

The world is back to normal under $(\hat{\mathcal{C}}\hat{\mathcal{P}})$.
Perhaps it is $(\mathcal{C}\mathcal{P})$ that is always conserved ?

Fundamental Discrete Symmetries

The fall of $(\mathcal{C}\mathcal{P})$ invariance[10]

Weak K-meson decays under $(\mathcal{C}\mathcal{P})$:

$$(\hat{\mathcal{C}}\hat{\mathcal{P}})\ |K_1\rangle = (\hat{\mathcal{C}}\hat{\mathcal{P}})\frac{1}{\sqrt{2}}\left(\left|K^0\right\rangle - \left|\overline{K}^0\right\rangle\right) = +1\,|K_1\rangle \qquad \tau_1 \approx 10^{-10}[s]$$

$$(\hat{\mathcal{C}}\hat{\mathcal{P}})\ |K_2\rangle = (\hat{\mathcal{C}}\hat{\mathcal{P}})\frac{1}{\sqrt{2}}\left(\left|K^0\right\rangle + \left|\overline{K}^0\right\rangle\right) = -1\,|K_2\rangle \qquad \tau_2 \approx 5\times 10^{-8}[s]$$

$$(\hat{\mathcal{C}}\hat{\mathcal{P}})\ \left|\pi^+\pi^-\right\rangle = (-1)^2\,\hat{\mathcal{C}}\ \left|\pi^+\pi^-\right\rangle = +1\left|\pi^+\pi^-\right\rangle$$

$$(\hat{\mathcal{C}}\hat{\mathcal{P}})\ \left|\pi^+\pi^-\pi^0\right\rangle = (-1)^3\,\hat{\mathcal{C}}\ \left|\pi^+\pi^-\pi^0\right\rangle = -1\left|\pi^+\pi^-\pi^0\right\rangle$$

However, in 0.2% of decays: $|K_2\rangle \longrightarrow |\pi^+\pi^-\rangle \quad \Rightarrow (\hat{\mathcal{C}}\hat{\mathcal{P}})$-**nonconservation**

Therefore: $|K_L\rangle = \frac{1}{\sqrt{1+\varepsilon}}\left(|K_2\rangle + \varepsilon\,|K_1\rangle\right) \qquad \varepsilon \approx 2.3\times 10^{-3}$

10 J. H. Christenson et al., *Phys Rev Lett* **13** (1964) *138*

Fundamental Discrete Symmetries

$(\mathcal{CP})$-Violation and Matter-Antimatter Asymmetry[11]

In 39% of events K_L decays differently:

$$K_L \longrightarrow \begin{cases} \pi^+ + e^- + \overline{\nu}_e & (-) \\ \pi^- + e^+ + \nu_e & (+) \end{cases}$$

N^+: decay into e^+
N^-: decay into e^-

$$\delta = \frac{N^+ - N^-}{N^+ + N^-} \approx 3 \times 10^{-3}$$

$(\hat{\mathcal{C}}\hat{\mathcal{P}})\,(\pm) \rightarrow (\mp)$
$|K_L\rangle$ is not $(\hat{\mathcal{C}}\hat{\mathcal{P}})$ eigenstate.

$(\hat{\mathcal{C}}\hat{\mathcal{P}})$-violation
$\Rightarrow N = N_e - N_{\overline{e}} \neq 0$

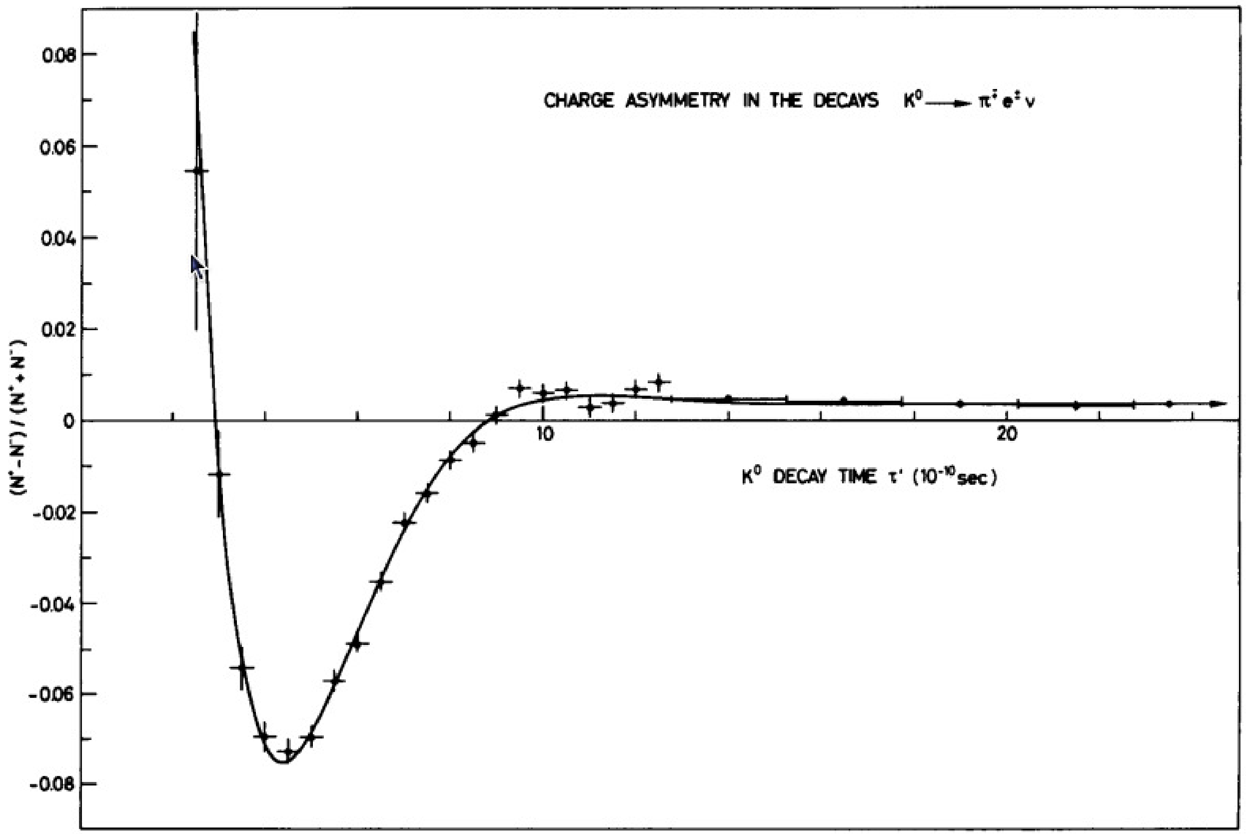


Fig. 1. The charge asymmetry as a function of the reconstructed decay time τ' for the K_{e3} decays. The experimental data are compared to the best fit as indicated by the solid line.

[11] S. Gjesdal et al., *Phys Lett* **52B** (1974) *113*
F. Wilczek (1980)

Fundamental Discrete Symmetries

$(\mathcal{CP})$-Violation in the Standard Model[12]

CKM quark-generation mixing matrix for charged weak interactions among quarks:

$$\begin{pmatrix} d' \\ s' \\ b' \end{pmatrix} = \begin{pmatrix} c_1 & s_1 c_3 & s_1 s_3 \\ -s_1 c_2 & c_1 c_2 c_3 - s_2 s_3\, e^{\imath\delta} & c_1 c_2 s_3 + s_2 c_3\, e^{\imath\delta} \\ -s_1 s_2 & c_1 s_2 c_3 + c_2 s_3\, e^{\imath\delta} & c_1 s_2 s_3 - c_2 c_3\, e^{\imath\delta} \end{pmatrix} \begin{pmatrix} d \\ s \\ b \end{pmatrix}$$

includes complex phase with $(\mathcal{CP})$-violating "δ parameter".

Cosmology:

A Matter-Antimatter Universe?[13] $\longrightarrow$ ruled out.

Leptogenesis[14]

Electroweak baryogenesis[15]

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[12] C. Cabibbo, *Phys Rev Lett* **10** (1963) *531*
M. Kobayashi, K. Maskawa, *Prog Theor Phys* **49** (1973) *652*

[13] A.G. Cohen, A. De Rújula, S.L. Glashow, *Astrophys J* **495** (1998) *539*

[14] S. Davidson, E. Nardi, Y. Nir, *Phys Rep* **466** (2008) *105*

[15] D.E. Morrissey, M.J. Ramsey-Musolf, *New J Phys* **14** (2012) *125003*

$(\mathcal{C}\mathcal{P})$-Violation and the $(\mathcal{C}\mathcal{P}\mathcal{T})$ Theorem[16]

$$(\not{\mathbf{C}}\not{\mathbf{P}}) \xrightarrow{\mathbf{CPT}} (\not{\mathbf{T}})$$

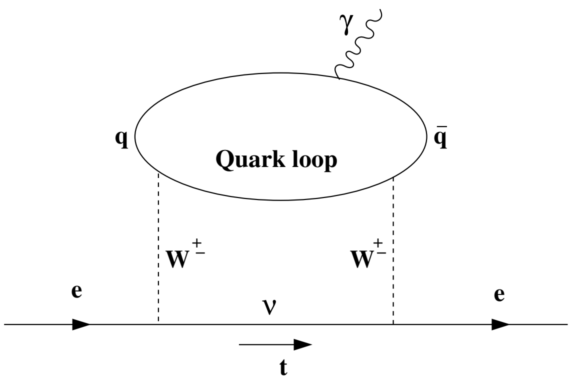
EDMs violate $\mathcal{T}$ symmetry

(at least in a minute ...)

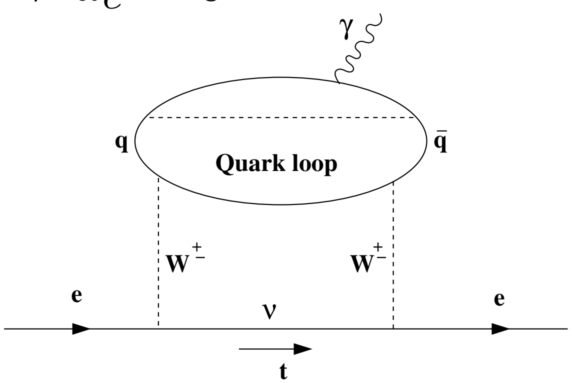
[16] W. Pauli, *Niels Bohr and the Development of Physics* (1955) *30*

A Lonely Electron in the Universe

Standard-Model Prediction of the Electron EDM

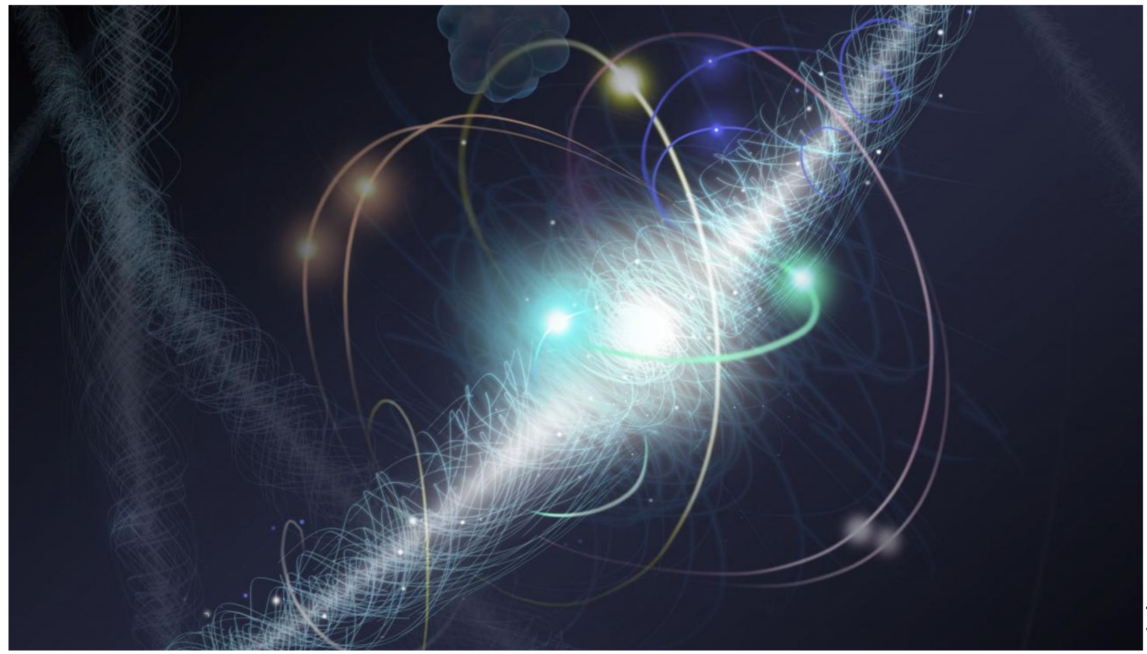


Summed two-loop diagrams[16]
$\Rightarrow d_e = 0$



Summed three-loop diagrams[16]
$\Rightarrow d_e = 0$

Interactions with virtual particles including $\mathcal{CP}$-violation
$\Rightarrow$ fermion EDM


[17]

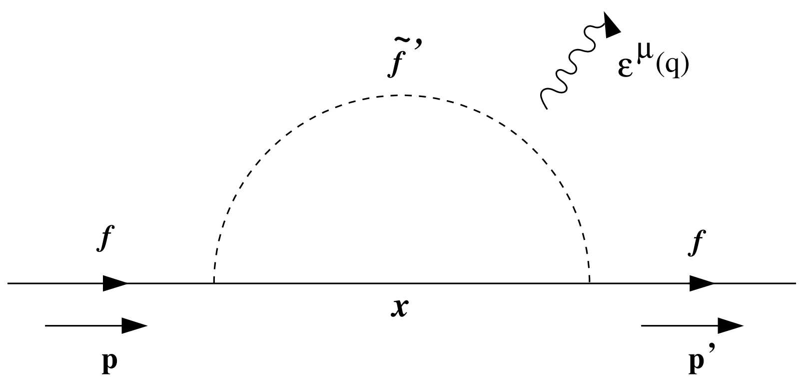
The SM eEDM is extremely small: $d_e < 10^{-38}\, e$cm

[16] M.É. Pospelov, I.B. Khriplovich, *Yad. Fiz.* **53** (1991) *1030*

[17] https://www.pourlascience.fr/sd/physique-particules/lelectron-met-a-mal-des-theories-au-dela-du-modele-standard-15089.php

The induced fermion EDM

Beyond the Standard Model



χ: chargino, neutralino

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

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

Chargino ($\tilde{\chi}^{\pm}_{1,2}$), neutralino ($\tilde{\chi}^{0}_{1,2,3,4}$) or gluino ($\tilde{g}^a$) fermion/sfermion interaction Lagrangian:

$$\mathcal{L}_{\chi f \tilde{f}'} = g^{\chi f \tilde{f}'_j}_{Lij} \left(\overline{\chi}_i P_L f\right) \tilde{f}'^{*}_j + g^{\chi f \tilde{f}'_j}_{Rij} \left(\overline{\chi}_i P_R f\right) \tilde{f}'^{*}_j + h.c.$$

One-loop fermion EDM:[19]

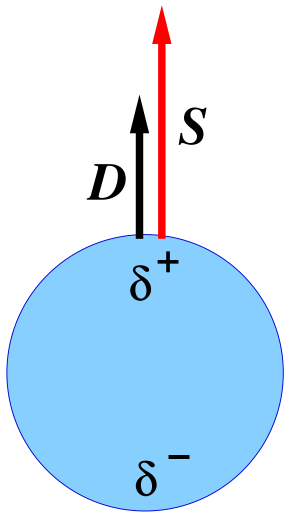
$$\left(\frac{d^E_f}{e}\right)^{\chi} = \frac{m_{\chi_i}}{16\pi^2 m^2_{\tilde{f}'_j}} \mathcal{I}m\left[\left(g^{\chi f \tilde{f}'_j}_{Rij}\right)^{*} g^{\chi f \tilde{f}'_j}_{Lij}\right] \left[Q_\chi A\left(\frac{m_{\chi_i}}{m^2_{\tilde{f}'_j}}\right) + Q_{\tilde{f}'_j} B\left(\frac{m_{\chi_i}}{m^2_{\tilde{f}'_j}}\right)\right]$$

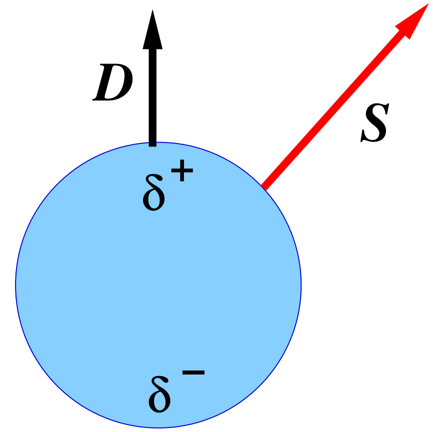
MSSM ("naïve SUSY") prediction:

$d_e \leq 10^{-27}\, e$ cm

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

The Fermion Electric Dipole Moment (fEDM)

Why 

? and not 

?

Sets of valid quantum numbers for fermion state:

$|C, T, U, \ldots, s, m_s\rangle$ $\quad$ $|C, T, U, \ldots, s, m_s, m_{\text{EDM}}\rangle$

The many-fermion state could be written:

$$|C(1) = C(2), T(1) = T(2), \ldots, s(1) = s(2), m_s(1) = m_s(2), m_{\text{EDM}}(1) \neq m_{\text{EDM}}(2)\rangle$$

leads to an internal contradiction (fermions would not be fermions) !

The Fermion EDM

Hamiltonian in Electromagnetic Field

Classical electromagnetism:

$\varepsilon_{\rm dip} = -\mathbf{D}\cdot\mathbf{E}$

Fermion EDM vector operator $\hat{\mathbf{d}} \propto \mathbf{\Sigma} = \begin{pmatrix} \boldsymbol{\sigma} & \mathbf{0} \\ \mathbf{0} & \boldsymbol{\sigma} \end{pmatrix}$ and so[20]

$\hat{H}_{\sf EDM} = -d_f\,\gamma^0\,\mathbf{\Sigma}\cdot\mathbf{E}$

The proportionality constant d_f is the fermion EDM.

Dirac matrix γ^0 ensures that $\left\langle \hat{H} \right\rangle$ is a Lorentz scalar (I'll come back to that)

Energy $\left\langle \hat{H} \right\rangle$ *violates* space-inversion ($\mathcal{P}$) and time-reversal ($\mathcal{T}$) symmetries:

$(\gamma^0)^{-1}\;\gamma^0\,\mathbf{\Sigma}\;\gamma^0 = \gamma^0\,\mathbf{\Sigma}$ $\qquad \mathcal{P}^{-1}\;\mathbf{E}\;\mathcal{P} = -\mathbf{E}$

$\left(\imath\gamma^0\gamma^5\gamma^2\,\hat{K}_0\right)^{-1}\gamma^0\,\mathbf{\Sigma}\;\imath\gamma^0\gamma^5\gamma^2\,\hat{K}_0 = -\gamma^0\,\mathbf{\Sigma}$ $\qquad \mathcal{T}^{-1}\;\mathbf{E}\;\mathcal{T} = \mathbf{E}$

This energy $\left\langle \hat{H} \right\rangle$ is a $\mathcal{T}$-odd *pseudo*scalar.

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

Atomic EDM

Schiff's Theorem

"The electric dipole moment of a bound-state atom composed of particles with non-zero electric dipole moments is zero in non-relativistic approximation."[21]

Consider the expectation value in eigenstate $\psi^{(0)}$ (incl. E_{ext})

$$\varepsilon_{\text{EDM}} = \left\langle -d_e\gamma^0\mathbf{\Sigma}\cdot\mathbf{E}\right\rangle_{\psi^{(0)}} = \left\langle -d_e\mathbf{\Sigma}\cdot\mathbf{E}\right\rangle_{\psi^{(0)}} + \left\langle d_e(\mathbb{1}_4-\gamma^0)\mathbf{\Sigma}\cdot\mathbf{E}\right\rangle_{\psi^{(0)}}$$

In the non-rel. limit $\gamma^0 \overset{\text{nrlimit}}{\longrightarrow} \mathbb{1}_4$ and so we consider

$$\left\langle -d_e\mathbf{\Sigma}\cdot\mathbf{E}\right\rangle_{\psi^{(0)}} = \frac{-d_e}{-e}\left\langle \mathbf{\Sigma}\cdot(\nabla_{\mathbf{x}}\,e\phi)\right\rangle_{\psi^{(0)}} = \frac{\imath d_e}{e\hbar}\left\langle [\mathbf{\Sigma}\cdot\mathbf{p}, e\phi\mathbb{1}_4]\right\rangle_{\psi^{(0)}}$$

$$= \frac{\imath d_e}{e\hbar}\left\langle \left[\mathbf{\Sigma}\cdot\mathbf{p}, c\boldsymbol{\alpha}\cdot\mathbf{p}+\gamma^0 m_0c^2-\hat{H}^{(0)}\right]\right\rangle_{\psi^{(0)}}$$

Since $\hat{H}^{(0)}\left|\psi^{(0)}\right\rangle = E^{(0)}\left|\psi^{(0)}\right\rangle$ all commutators vanish, and so

$\left\langle -d_e\mathbf{\Sigma}\cdot\mathbf{E}\right\rangle_{\psi^{(0)}} = 0 \quad \square.$

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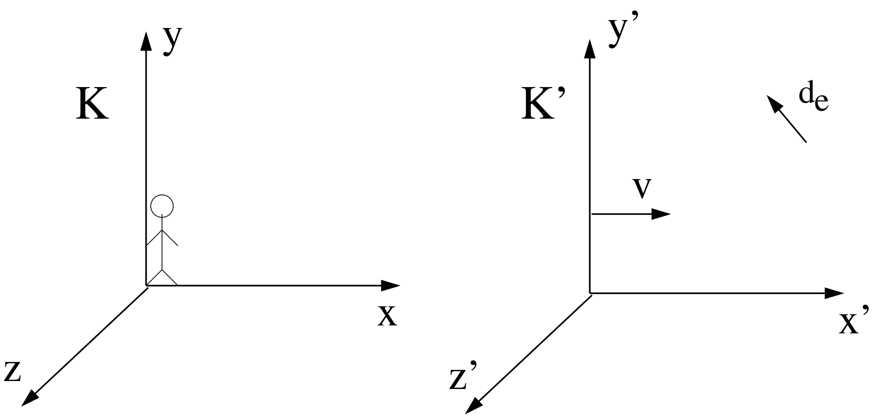
[21] L.I. Schiff, *Phys Rev* **132** (1963) *2194*

Atomic EDM

Evading Schiff's Theorem by Special Relativity[22]



Length contraction for collinear movement:

$$\mathbf{d}_e(K) = \frac{\mathbf{d}_e(K')}{\gamma} = \mathbf{d}_e(K') \left(1 - \frac{\gamma}{1+\gamma}\frac{v^2}{c^2}\right)$$

... and for general movement:

$$\mathbf{d}_e(K) = \mathbf{d}_e(K') - \frac{\gamma}{1+\gamma}\frac{\mathbf{v}}{c}\left(\mathbf{d}_e(K') \cdot \frac{\mathbf{v}}{c}\right)$$

The dipole energy in K then is

$$\varepsilon_{\text{dip}} = -\mathbf{d}_e(K) \cdot \mathbf{E} = -\mathbf{d}_e(K') \cdot \left[\mathbf{E} - \frac{\gamma}{1+\gamma}\frac{\mathbf{v}}{c}\left(\frac{\mathbf{v}}{c} \cdot \mathbf{E}\right)\right]$$

For small relative velocities we can approximate:

$$\varepsilon_{\text{dip}} \approx -\mathbf{d}_e(K') \cdot \mathbf{E} + \frac{1}{2m_e^2c^2}\mathbf{d}_e(K') \cdot \mathbf{p}\,(\mathbf{p} \cdot \mathbf{E})$$

[22] E.D. Commins, J.D. Jackson, D.P. DeMille, *Am J Phys* **75** (2007) *532*

Atomic EDM

Interpretation of the EDM Interaction

The $\mathcal{P},\mathcal{T}$-odd energy can also be written as

$$\varepsilon_{\mathsf{EDM}} = -d_e \left\langle \Psi^L \; \Psi^S \left| \begin{pmatrix} \mathbf{1}_2 & \mathbf{0}_2 \\ \mathbf{0}_2 & -\mathbf{1}_2 \end{pmatrix} \begin{pmatrix} \boldsymbol{\sigma}\cdot\mathbf{E} & \mathbf{0}_2 \\ \mathbf{0}_2 & \boldsymbol{\sigma}\cdot\mathbf{E} \end{pmatrix} \right| \begin{matrix} \Psi^L \\ \Psi^S \end{matrix} \right\rangle$$

$$= -d_e \left\{ \left\langle \Psi^L \left| \boldsymbol{\sigma}\cdot\mathbf{E} \right| \Psi^L \right\rangle - \left\langle \Psi^S \left| \boldsymbol{\sigma}\cdot\mathbf{E} \right| \Psi^S \right\rangle \right\}$$

Using the low-energy relationship between L and S components of the Dirac spinors $\Psi^S \approx \frac{\boldsymbol{\sigma}\cdot\hat{\mathrm{p}}}{2mc}\Psi^L$ gives

$$\approx -d_e \left\{ \langle \boldsymbol{\sigma}\cdot\mathbf{E} \rangle_{\Psi^L} - \frac{1}{4m^2c^2} \left\langle (\boldsymbol{\sigma}\cdot\hat{\mathbf{p}})^\dagger \, \boldsymbol{\sigma}\cdot\mathbf{E}\, \boldsymbol{\sigma}\cdot\hat{\mathbf{p}} \right\rangle_{\Psi^L} \right\}$$

Respecting the derivative and using twice the Dirac relation

$$\boldsymbol{\sigma}\cdot\hat{\mathbf{p}}\; \boldsymbol{\sigma}\cdot\mathbf{E} = \hat{\mathbf{p}}\cdot\mathbf{E}\mathbf{1}_2 + \imath\boldsymbol{\sigma}\cdot\hat{\mathbf{p}}\times\mathbf{E}$$

we finally get

$$\varepsilon_{\mathsf{EDM}} \approx -d_e \left\{ \langle \boldsymbol{\sigma}\cdot\mathbf{E} \rangle_{\Psi^L} - \frac{1}{4m^2c^2} \left[\langle \hat{\mathbf{p}}\cdot\mathbf{E}\,\boldsymbol{\sigma}\cdot\hat{\mathbf{p}} \rangle_{\Psi^L} + \langle \mathbf{E}\cdot\hat{\mathbf{p}}\,\boldsymbol{\sigma}\cdot\hat{\mathbf{p}} \rangle_{\Psi^L} \right] \right\}$$

which corresponds to the classical dipole energy in the observer frame.

Atomic EDM

Lorentz-Covariant eEDM Hamiltonian

A magnetic field $\mathbf{B}$ in the lab frame (say due to nuclear charged current) partially transforms into an electric field $\mathbf{E}'$ in the electron rest frame (and vice versa).

Covariant eEDM Hamiltonian:

$$\hat{H}_{\mathrm{EDM}} = \imath \frac{d_e}{2} \gamma^0 \gamma^5 \frac{\imath}{2} \left(\gamma^\mu \gamma^\nu - \gamma^\nu \gamma^\mu \right) F_{\mu\nu}$$

Use covariant EM field tensor

$$\{F_{\mu\nu}\} = \{\partial_\mu A_\nu - \partial_\nu A_\mu\} = \begin{pmatrix} 0 & E_x & E_y & E_z \\ -E_x & 0 & -B_z & B_y \\ -E_y & B_z & 0 & -B_x \\ -E_z & -B_y & B_x & 0 \end{pmatrix}$$

to derive conventional representation:

$$\hat{H}_{\mathrm{EDM}} = -d_e \gamma^0 \left[\mathbf{\Sigma} \cdot \mathbf{E} + \imath \boldsymbol{\alpha} \cdot \mathbf{B} \right]$$

Off-diagonal $\boldsymbol{\alpha} = \begin{pmatrix} \mathbf{0}_2 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & \mathbf{0}_2 \end{pmatrix}$ couples Ψ^L and Ψ^S and is strongly suppressed due to $\Psi^S \approx \frac{\boldsymbol{\sigma} \cdot \hat{\mathrm{p}}}{2mc} \Psi^L$ (watch out for caveats)

Atomic EDM

Definition and eEDM enhancement

Electric dipole moment of an atom:[23]

$d_a := -\lim_{E_{\text{ext}} \to 0} \left[\frac{\partial(\Delta\varepsilon_{\not P \not T})}{\partial E_{\text{ext}}} \right]$ $\Delta\varepsilon_{\not P \not T}$ is **some** P, T-odd energy shift.

Sources are particle EDMs, nuclear MQM, nuclear Schiff moment, $\mathcal{T}$-odd contribution to weak interaction.

For an electron EDM, we then have

$$d_a = \lim_{E_{\text{ext}} \to 0} \frac{\partial}{\partial E_{\text{ext}}} d_e \left\langle \gamma^0 \left[\boldsymbol{\Sigma} \cdot \mathbf{E} + \imath \boldsymbol{\alpha} \cdot \mathbf{B} \right] \right\rangle_{\psi(E_{\text{ext}})}$$

With the definitions $(E+B)_{\text{eff}} = -\left\langle \gamma^0 \left[\boldsymbol{\Sigma} \cdot \mathbf{E} + \imath \boldsymbol{\alpha} \cdot \mathbf{B} \right] \right\rangle_{\psi(E_{\text{ext}})}$

$R := \frac{d_a}{d_e}$ $R_{\text{lin}} := -\frac{\Delta(E+B)_{\text{eff}}}{\Delta E_{\text{ext}}} = -\frac{(E+B)_{\text{eff}}(2) - (E+B)_{\text{eff}}(1)}{E_{\text{ext}}(2) - E_{\text{ext}}(1)}$

the linear-regime atomic eEDM enhancement is then:

$$R \approx R_{\text{lin}} = -\frac{(E+B)_{\text{eff}}}{E_{\text{ext}}}$$

---

[23] E.D. Commins, *Adv. Mol. Opt. Phys.* **40** (1999) *1*

Atomic EDM

Scaling and Choice of Sensitive Systems

An **atom** can be much **more sensitive** than a free electron! (Sandars effect)[24]

Analytical estimates of the eEDM enhancement[25]
$R \propto 10\, Z^3 \alpha^2$

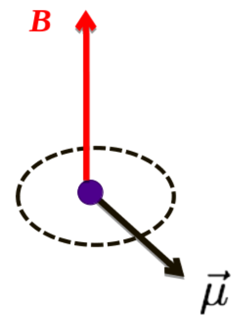
High-Z atoms with unpaired electron shells are optimal choice:

Atom (state)	Rb ($^2S_{1/2}$)	Cs ($^2S_{1/2}$)	Fr ($^2S_{1/2}$)	Tl ($^2P_{1/2}$)
Z	37	55	87	81
R	26 ± 1[25]	114 ± 3[26]	910 ± 45[27]	-559 ∓ 28[28]

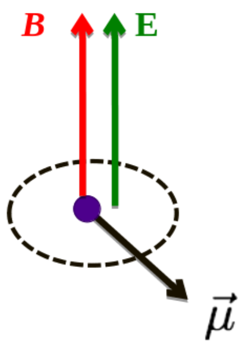
[24] P.G.H. Sandars, *Phys Lett* **14** (1965) *194*
[25] E.D. Commins, D. DeMille, *Adv. Ser. Dir. High En. Phys.* **chapter 14** (2008) *519*
V.V. Flambaum, *Sov. J. Nucl. Phys.* **24** (1976) 199
[25] A. Shukla, B.P. Das, J. Andriessen, *Phys. Rev. A* **50** (1994) 1155
[26] A. C. Hartley, E. Lindroth, A.-M. Mårtensson-Pendrill, *J. Phys. B: At. Mol. Opt. Phys.* **23** (1990) 3417
[27] T.M.R. Byrnes, V.A. Dzuba, V.V. Flambaum, D.W. Murray, *Phys. Rev. A* **59** (1999) 3082
[28] T. F., L.V. Skripnikov, *J. Phys. B: At. Mol. Opt. Phys.* (2019) submitted.

Atomic EDM

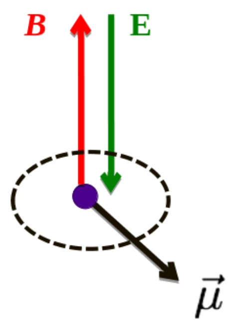
Measurement Principle[30]



(1)



(2)



(3)

Hamiltonian of sensitive system in external EM field:

$$\hat{H} = -(\mu\mathbf{B} + d\mathbf{E}) \cdot \frac{\hat{\mathbf{J}}}{|J|}$$

(1) B-field causes spin precession with frequency ν:

$$-(\mu\mathbf{B}) \cdot \frac{\hat{\mathbf{J}}}{|J|} = h\nu$$

(2) Added E-field modifies spin precession freq. to ν_+:

$$-(\mu\mathbf{B} + d\mathbf{E}) \cdot \frac{\hat{\mathbf{J}}}{|J|} = h\nu_+$$

(3) Reversed E-field modifies spin precession freq. to ν_-:

$$-(\mu\mathbf{B} - d\mathbf{E}) \cdot \frac{\hat{\mathbf{J}}}{|J|} = h\nu_-$$

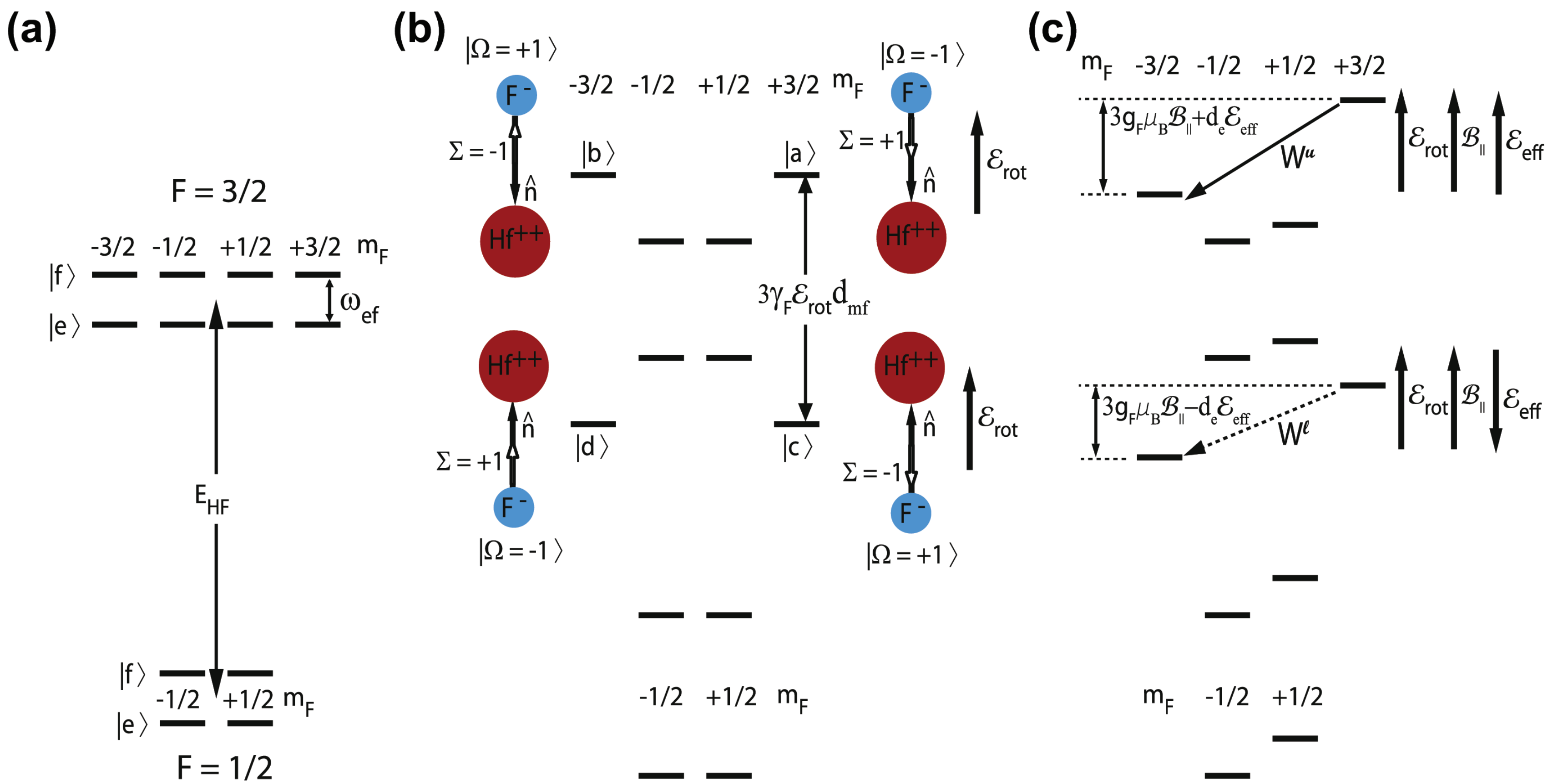
EDM of system can be extracted from:

$$\nu_+ - \nu_- = \frac{2dE|J|}{h}$$

[30] M. Bishof, M. Dietrich, *et al.*, *Phys. Rev. C* **94** (2016) 025501
B. C. Regan, E. D. Commins, C. J. Schmidt, D. DeMille, *Phys. Rev. Lett.* **88** (2002) 071805

EDM Measurement in Molecules

HfF^+ as Example[31]

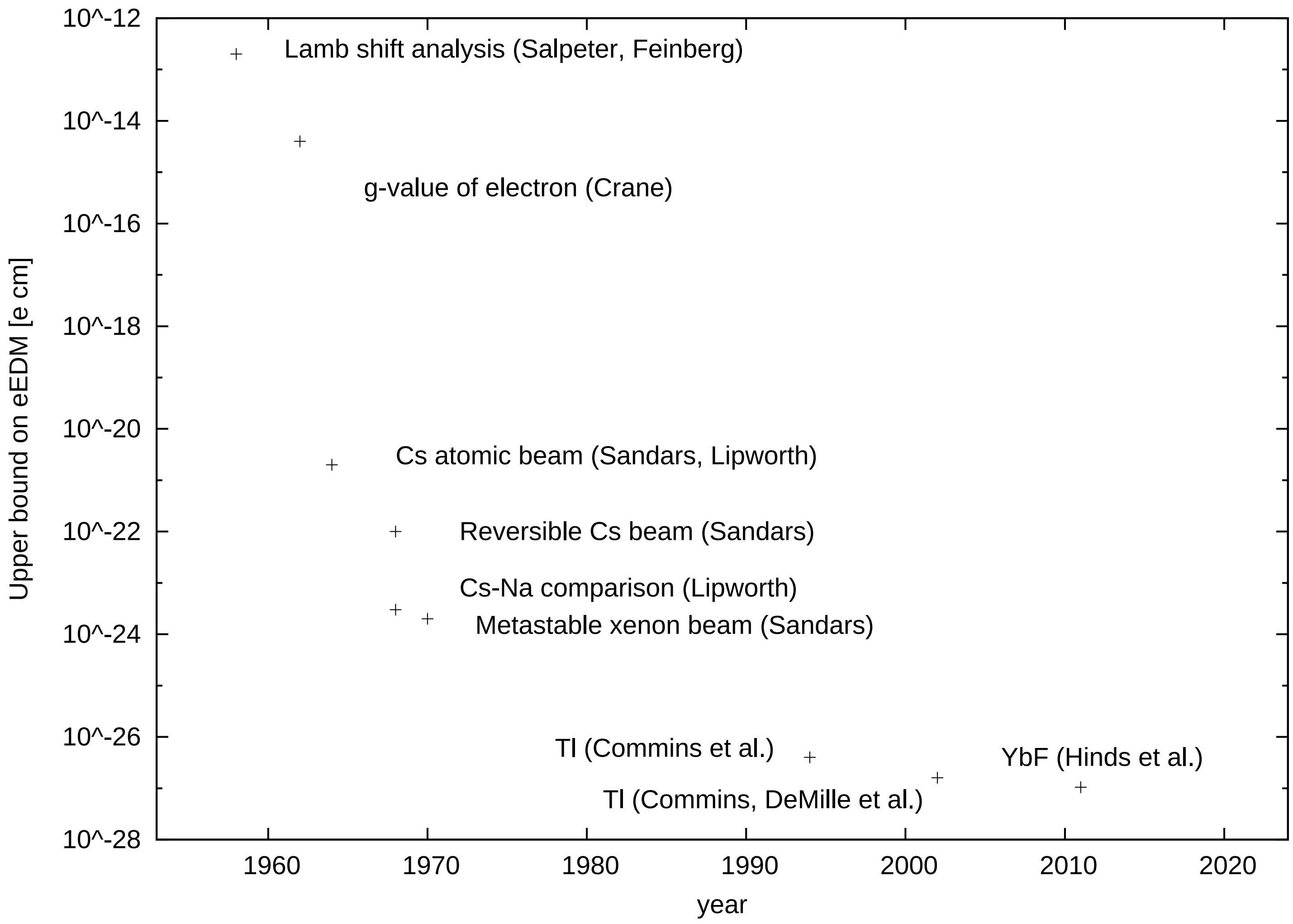


$$\frac{W^u(B)+W^u(-B)}{2E_{\text{eff}}} = d_e$$

[31] A.E. Leanhardt *et al.*, E.A. Cornell, *J Mol Spectrosc* **270** (2011) *1*
W.B. Cairncross *et al.*, J. Ye, E.A. Cornell, *Phys Rev Lett* **119** (2017) *153001*

Electron Electric Dipole Moment

Historic Upper Bounds From Atomic EDM Measurements



Atomic and Molecular Correlated Wavefunctions[32]

Hamiltonians

- Dirac-Coulomb Hamiltonian + external electric field (atoms)

$$\hat{H}^{\text{Dirac-Coulomb}} + \hat{H}^{\text{Int-Dipole}} = \sum_{i}^{n} \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \frac{Z}{r_i} \mathbb{1}_4 \right] + \sum_{i,j>j}^{n} \frac{1}{r_{ij}} \mathbb{1}_4 + \sum_{i}^{n} \mathbf{r}_i \cdot \mathbf{E}_{\text{ext}}\, \mathbb{1}_4$$

- Dirac-Coulomb Hamiltonian operator (molecules)

$$\hat{H}^{DC} = \sum_{i}^{n} \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \sum_{A}^{N} \frac{Z}{r_{iA}} \mathbb{1}_4 \right] + \sum_{i,j>i}^{n} \frac{1}{r_{ij}} \mathbb{1}_4 + \sum_{A,B>A}^{N} V_{AB}$$

- Dirac-Coulomb-Gaunt Hamiltonian operator (molecules)

$$\hat{H}^{DCG} = \sum_{i}^{n} \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \sum_{A}^{N} \frac{Z}{r_{iA}} \mathbb{1}_4 \right] + \sum_{i,j>i}^{n} \left(\frac{1}{r_{ij}} \mathbb{1}_4 - \frac{1}{2} \frac{\vec{\alpha}_i \vec{\alpha}_j}{r_{ij}} \right) + \sum_{A,B>A}^{N} V_{AB}$$

[32] T. F., H.J.Å. Jensen, J. Olsen, L. Visscher, *J Chem Phys* **124** (2006) *104106*
S. Knecht, H.J.Å. Jensen, T. F., *J Chem Phys* **132** (2010) *014108*

Calculation of $\mathcal{P},\mathcal{T}$-Violating Effects[33]

String-Based CI Techniques

Expectation values over relativistic Configuration Interaction wavefunctions

$$\left\langle \hat{O} \right\rangle_{\psi_k^{(0)}} = \sum_{I,J=1}^{\dim\mathcal{F}^{\rm t}({\rm M,n})} c_{kI}^* c_{kJ} \, \langle \, | (\mathcal{S}\overline{\mathcal{T}})_I^\dagger \left| \hat{O} \right| (\mathcal{S}\overline{\mathcal{T}})_J \, | \, \rangle$$

Property operator $\hat{O}$ in basis of Kramers-paired molecular spinors

$$\hat{O} = \sum_{m,n=1}^{P_u} o_{mn} \, a_m^\dagger a_n + \sum_{m=1}^{P_u} \sum_{n=P_u+1}^{P} o_{m\overline{n}} \, a_m^\dagger a_{\overline{n}} + \sum_{m=P_u+1}^{P} \sum_{n=1}^{P_u} o_{\overline{m}n} \, a_{\overline{m}}^\dagger a_n + \sum_{m,n=P_u+1}^{P} o_{\overline{m}\overline{n}} \, a_{\overline{m}}^\dagger a_{\overline{n}}$$

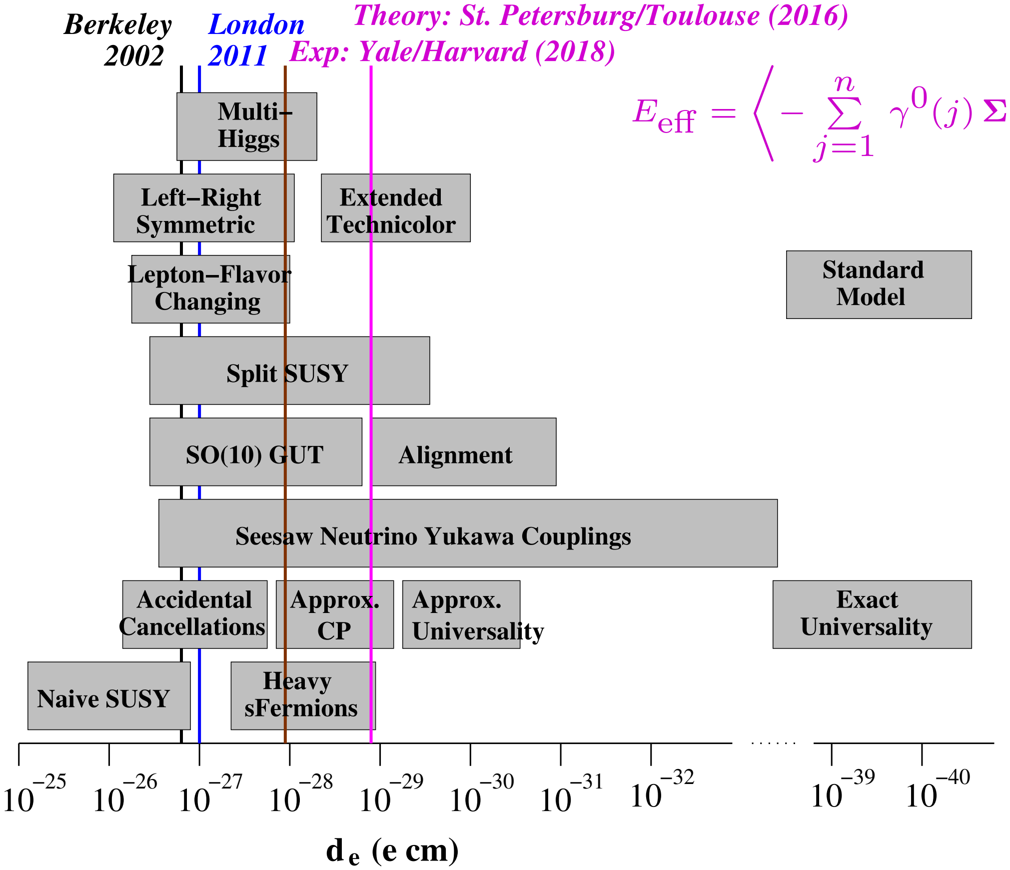
First-term contribution to expectation value

$$W'(\Psi_k)_1 = \sum_{I,J=1}^{\dim\mathcal{F}^{\rm t}({\rm P,N})} c_{kI}^* c_{kJ} \sum_{m,n=1}^{P_u} o_{mn}^M$$

$$\langle \, | \prod_{p=1}^{N_p \in \mathcal{S}_I} \prod_{\overline{p}=N_p+1}^{N_p \in \mathcal{S}_I + N_{\overline{p}} \in \overline{\mathcal{T}}_I} a_{\overline{p}} a_p \, a_m^\dagger a_n \prod_{q=1}^{N_p \in \mathcal{S}_J} \prod_{\overline{q}=N_p+1}^{N_p \in \mathcal{S}_J + N_{\overline{p}} \in \overline{\mathcal{T}}_J} a_q^\dagger a_{\overline{q}}^\dagger \, | \, \rangle$$

[33] S. Knecht, Dissertation, HHU Düsseldorf (2009)
T. F., M.K. Nayak, *Phys Rev A* **88** (2013) *032514*

eEDM Constraint on Beyond-Standard-Model Theories[34]
Single-source interpretation



$$E_{\rm eff} = \left\langle -\sum_{j=1}^{n} \gamma^0(j)\,\mathbf{\Sigma}(j)\cdot\mathbf{E}(j)\right\rangle_{\psi(0)} \approx \frac{2\imath c}{e\hbar}\left\langle \sum_{j=1}^{n} \gamma^0(j)\gamma^5(j)\,\vec{p}(j)^{\,2}\right\rangle_{\psi(0)} \approx 78\left[\frac{\rm GV}{\rm cm}\right]$$

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)[34]	$< 1.6 \cdot 10^{-27}$
Experimental limit (YbF)[35]	$< 10.5 \cdot 10^{-28}$
Experimental limit (ThO)[36]	$< 1.1 \cdot 10^{-29}$

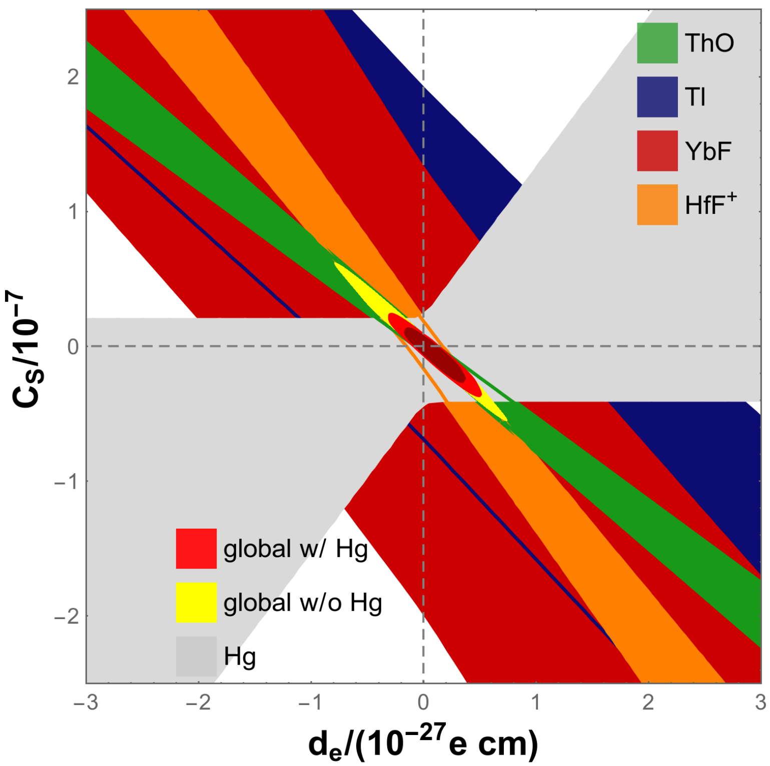
[34] D. DeMille (2005), H. Nataraj (2009)

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

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

[36] ACME Collaboration, *Nature* **562** (2018) *355*; ACME, *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*; M. Denis, TF, *J Chem Phys* **145** (2016) *214307*

More Stringent Bounds on (Semi-)Leptonic $\mathcal{CP}$-odd Parameters[37]



Multiple-source picture:

$$\Delta E_{\mathcal{P},\mathcal{T}} = -\langle \mathbf{d}_{\text{sys}} \cdot \mathbf{E}_{\text{ext}} \rangle$$
$$= \left(\alpha_{d_e}\, d_e + \alpha_{C_S}\, C_S\right) \langle \mathbf{n} \cdot \mathbf{z} \rangle \left(E_{\text{ext}}\right)$$

Previous resulting bound:

From HfF^+, ThO, YbF, Tl

$|d_e|_{2017} < 6.4 \times 10^{-28} e$ cm

New resulting bounds:

From Hg, HfF^+, ThO, YbF, Tl

$|d_e|_{2018} < 3.8 \times 10^{-28} e$ cm

$|C_S|_{2018} < 2.7 \times 10^{-8}$

[37] T. F., M. Jung, *J. High Energy Phys.* **7** (2018) *012*
J. Baron *et al.*, *Science* **343** (2014) *269*
M. Denis, T. F., *J. Chem. Phys.* **145** (2016) *214307*
L. Skripnikov, *J. Chem. Phys.* **145** (2016) *214301*
W.B. Cairncross, D.N. Gresh, M. Grau, K.C. Cossel, T.S. Roussy, Y. Ni, Y. Zhou, J. Ye, E.A. Cornell, *Phys. Rev. Lett.* **119** (2017) *153001*
T. F., *Phys. Rev. A (Rap. Comm.)*, **96** (2017) *040502(R)*
L.V. Skripnikov, *J. Chem. Phys.*, **147** (2017) *021101*

Current World Records

In the presence of a non-zero EDM d, the system's Hamiltonian is
$\hat{H} = -(\mu\mathbf{B} + d\mathbf{E}) \cdot \frac{\hat{\mathbf{J}}}{|J|}$

- **"Paramagnetic" systems**: Precession measurement on **ThO**
  ACME II collaboration Yale/Harvard; DeMille, Doyle, Gabrielse[39]
  measured $\left|\omega^{\mathcal{N}\mathcal{E}}\right| = (-510 \pm 683)\ \frac{\mu\text{rad}}{s} \Rightarrow |d_e| \leq 1.1 \times 10^{-29}\, e$ cm

- **"Diamagnetic" systems**: Precession measurement on **Hg**
  Seattle group; Heckel[40]
  measured $|d_{Hg}| \leq 7.4 \times 10^{-30}\, e$ cm

- **Neutron** (n) EDM experiment
  PSI, Switzerland[41]
  measured $|d_n| \leq 3.6 \times 10^{-26}\, e$ cm

---

39 V. Andreev *et al.*, Nature **562** (2018) *355*

40 B. Graner *et al.*, Phys Rev Lett **116** (2016) *161601*

41 J.M. Pendlebury *et al.*, Phys. Rev. D, **92** (2015) *092003*

Science

in collaboration with

D. DeMille

Yale University

New Haven, CT 06520, USA

- **Molecular EDMs : AgRa**

- Atomic EDMs : ^{129}Xe

Going Ultracold: From beams to traps

PHYSICAL REVIEW A, VOLUME 63, 023405

Loading and compressing Cs atoms in a very far-off-resonant light trap

D. J. Han, Marshall T. DePue, and David S. Weiss
Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300
(Received 25 May 2000; published 12 January 2001)

We describe an experiment in which 3×10^{7} Cs atoms are loaded into a 400 μm crossed beam far-off-resonant trap (FORT) that is only 2 μK deep. A high-density sample is prepared in a magneto-optic trap, cooled in a three-dimensional far-off-resonant lattice (FORL), optically pumped into the lowest-energy state, adiabatically released from the FORL, magnetically levitated, and transferred to the final trap with a phase-space density of 10^{-3}. Spontaneous emission in the FORT is negligible, and we have compressed the atoms in the FORT to a spatial density of 2×10^{13} atoms/cm^3. Evaporative cooling under these conditions proceeds rapidly.

- Estimated sensitivity of Cs EDM measurement in DLT[42] is $|d_e| \approx 10^{-29}\, e\text{cm}$

Cs atom: $\Delta E = R\, E_{\text{ext}}\, d_e$
$E_{\text{int}} \approx 20\ \left[\frac{\text{MV}}{\text{cm}}\right]$

Ultracold **XY** Molecule: $\Delta E = E_{\text{eff}}\, d_e$
$E_{\text{eff}} \approx 50\ \left[\frac{\text{GV}}{\text{cm}}\right]$

- A factor of $\approx \mathbf{2500}$ gain in sensitivity!

---

[42] DLT: Dipole light trap; D. Weiss (Penn State), 2014: "Measuring the eEDM using laser-cooled Cs atoms in optical lattices"
S. Chu, J.E. Bjorkholm, A. Ashkin, A. Cable, *Phys. Rev. Lett.* **57** (1986) *314*
C. Chin, V. Leiber, V. Vuletić, A.J. Kerman, S. Chu, *Phys. Rev. A* **63** (2001) *033401*

Towards Ultracold DLT EDM Measurement

Picking the cherry

In the casting:
Alkali(-like) atoms: Li, Na, K, Rb, Cs; Ag, Au
Earth-alkaline atoms: Sr, Ba, Ra; Yb

Jury spreadsheet for X partner of Ra and some contenders:

X	EA(X) [eV]	$E_{\text{eff}_{\max}}$ $\left[\frac{\text{GV}}{\text{cm}}\right]$	$B_v = \left\langle v \middle| \frac{1}{\mu R^2} \middle| v \right\rangle$ $[\text{cm}^{-1}]$	D [D]	$E_{\text{pol}} = \frac{B_v}{D}$ $\left[\frac{\text{kV}}{\text{cm}}\right]$
Li	0.62	61	$-$	≈ 1.5	
Na	0.55	58	$-$	≈ 1	
K	0.50	50	$-$	≈ 1	
Rb	0.49	48	$+$	≈ 1	
Cs	0.47	44	$+$	≈ 1	
Ag	1.30	66	0.021	5.4	0.264
Au	2.31	60	$+$	≈ 6	
AgBa	1.30	6	$+$	≈ 3	
RbYb[42]		-0.7	0.001	0.21	5.5
CsYb[42]		0.54	0.007	0.24	3.5

[42] E. R. Meyer, J. L. Bohn, *Phys. Rev. A* **80** (2009) *042508*

$(\mathcal{P},\mathcal{T})$-odd properties of AgRa

- Electron EDM effective electric field[44]

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

- S-PS nucleon-electron interaction constant[45]

$$W_{\mathcal{S}} := \frac{\imath}{\Omega} \frac{G_F}{\sqrt{2}} Z_{\text{heavy}} \left\langle \Psi_\Omega \right| \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \, \rho_N(\vec{r}_j) \left| \Psi_\Omega \right\rangle$$

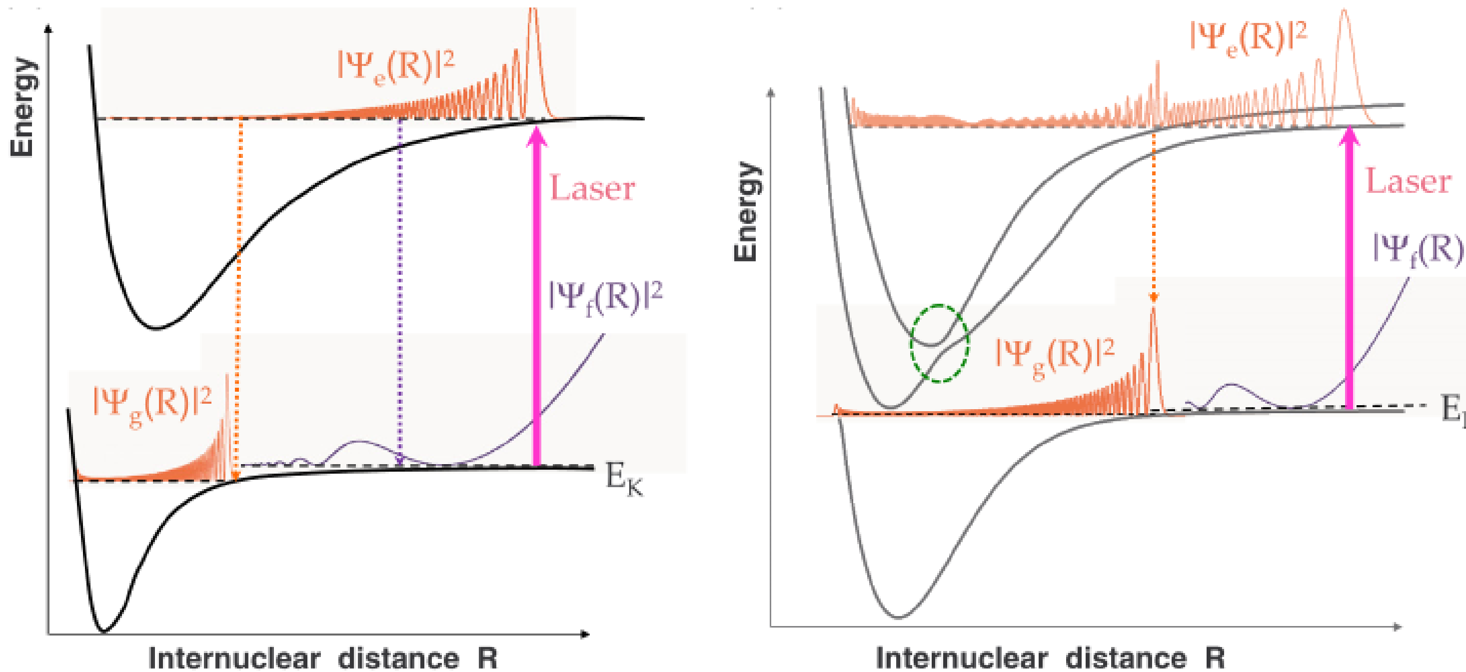
	$^3\Delta_1$			$^2\Sigma_{1/2}$		
	ThO	HfF^+	ThF^+	YbF	AgRa	
$\lvert E_{\text{eff}}\rvert$	78	23	35	25	64	$\left[\frac{\text{GV}}{\text{cm}}\right]$
$\lvert W_S\rvert$	106	20	51	40	175	[kHz]

[44] E. Lindroth, E. Lynn, P.G.H. Sandars, *J. Phys. B: At. Mol. Opt. Phys.* **22** (1989) *559*
T.F., M.K. Nayak, *Phys. Rev. A* **88** (2013) *032514*

[45] V. G. Gorshkov, L. N. Labzovski, and A. N. Moskalev, *Zh. Eksp. Teor. Fiz.* **76** (1979) *414*
M. Denis *et al.*, *New J. Phys.* **7** (2015) *043005*

Devising a AgRa DLT EDM Experiment

- Photoassociating ultracold atoms into ultracold molecules[46]



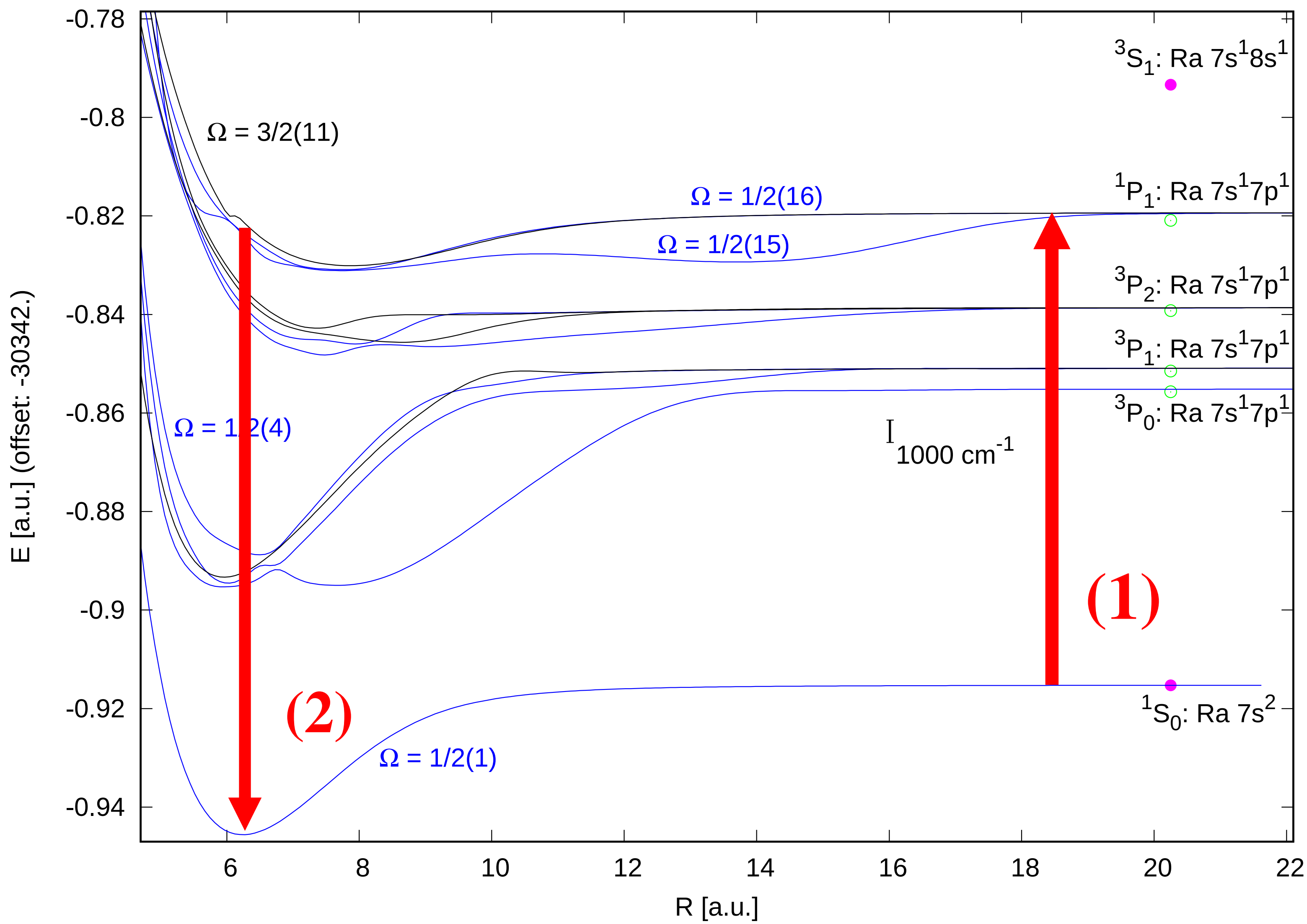
- Does its electronic spectrum allow for efficient energy transfer (remove binding energy without heating) ?

- Which states are candidates for photoassociation ?

[46] L. D. Carr, D. DeMille, R. V. Krems, J. Ye, *New J. Phys.* **11** (2009) *055049*

AgRa

A Pathway To Assemble AgRa (X) from Trapped Ag-Ra Atom Pairs



Long-Range Theory[47]

Van der Waals interaction potential for two neutral heteronuclear atoms:

$$V(R) = -\frac{C_6}{R^6} - \frac{C_8}{R^8} - \frac{C_{10}}{R^{10}} - \dots$$

Ground state:

$$C_6^{\Omega=1/2(1)} = \sum_{n_c(\ell_c=1),n_d(\ell_d=1)} \frac{3}{2} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

$a = {}^2S_{1/2}(5s^1)$ for Ag and $b = {}^1S_0(7s^2)$ for Ra

Oscillator strengths:[48]

$$f_{IF}^{(1)} = \frac{2}{3g_I}\left(E_F - E_I\right) \sum_{\substack{M_{LF},M_{SF} \\ M_{LI},M_{SI}}} \left|\left| \langle L_F, M_{LF}, S_F, M_{SF} | \sum_{k=1}^{n} \hat{\mathbf{r}}(k) \, | L_I, M_{LI}, S_I, M_{SI} \rangle \right|\right|^2$$

$$g_I = (2M_{LI} + 1)(2M_{SI} + 1)$$

$|\psi_L\rangle = \left|{}^2P\right\rangle = \left|1, M_L; \tfrac{1}{2}, M_S\right\rangle$ are expanded as

$$\left|1,0;\tfrac{1}{2},\tfrac{1}{2}\right\rangle = \left\langle\tfrac{3}{2},\tfrac{1}{2}\middle|1,0;\tfrac{1}{2},\tfrac{1}{2}\right\rangle \left|\tfrac{3}{2},\tfrac{1}{2}\right\rangle + \left\langle\tfrac{1}{2},\tfrac{1}{2}\middle|1,0;\tfrac{1}{2},\tfrac{1}{2}\right\rangle \left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle$$

[47] J.-Y. Zhang, J. Mitroy, *Phys. Rev. A* **76** (2007) *022705*

[48] T.N. Chang, *Phys. Rev. A* **36** (1987) *447*

Connecting LR- and SR-Potentials

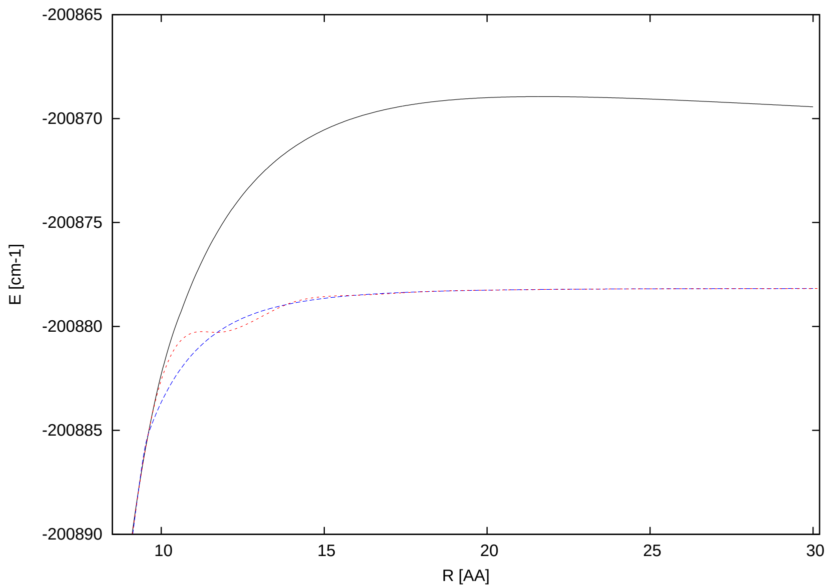
Ground potential $\Omega = 1/2(1)$

LeRoy radius[48]:

$$R_{\mathrm{LR}} = 2\left(\langle \hat{r}^2\rangle_A^{1/2} + \langle \hat{r}^2\rangle_B^{1/2}\right)$$

$$\langle \hat{r}^2\rangle_{\mathrm{Ag}_{5s}} = 13.90 \text{ a.u.} \qquad \langle \hat{r}^2\rangle_{\mathrm{Ra}_{7s}} = 28.71 \text{ a.u.}$$

$$\Rightarrow R_{\mathrm{LR}}^{\Omega=1/2(1)} = 18.2\,\text{a.u.}$$



- Pure short-range potentials produce artefacts! |
- $V(R) \approx -\frac{C_6}{R^6}$ and fit long-range to short-range curves
- Correct physics from careful fitting | |

[48] R.J. LeRoy, *Can. J. Phys.* **52** (1974) *246*
R.J. LeRoy, R. B. Bernstein, *J. Chem. Phys.* **52** (1970) *3869*

Dispersion coefficients from oscillator strengths

$$C_6^{\Omega=1/2(16)} = C_6^{\Omega=3/2(11)} = \sum_{n_c(\ell_c=1),n_d(\ell_d=0)} \frac{3}{4} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)} + \sum_{n_c(\ell_c=1),n_d(\ell_d=1)} \frac{15}{8} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)} + \sum_{n_c(\ell_c=1),n_d(\ell_d=2)} \frac{57}{40} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

A test on LiBe:

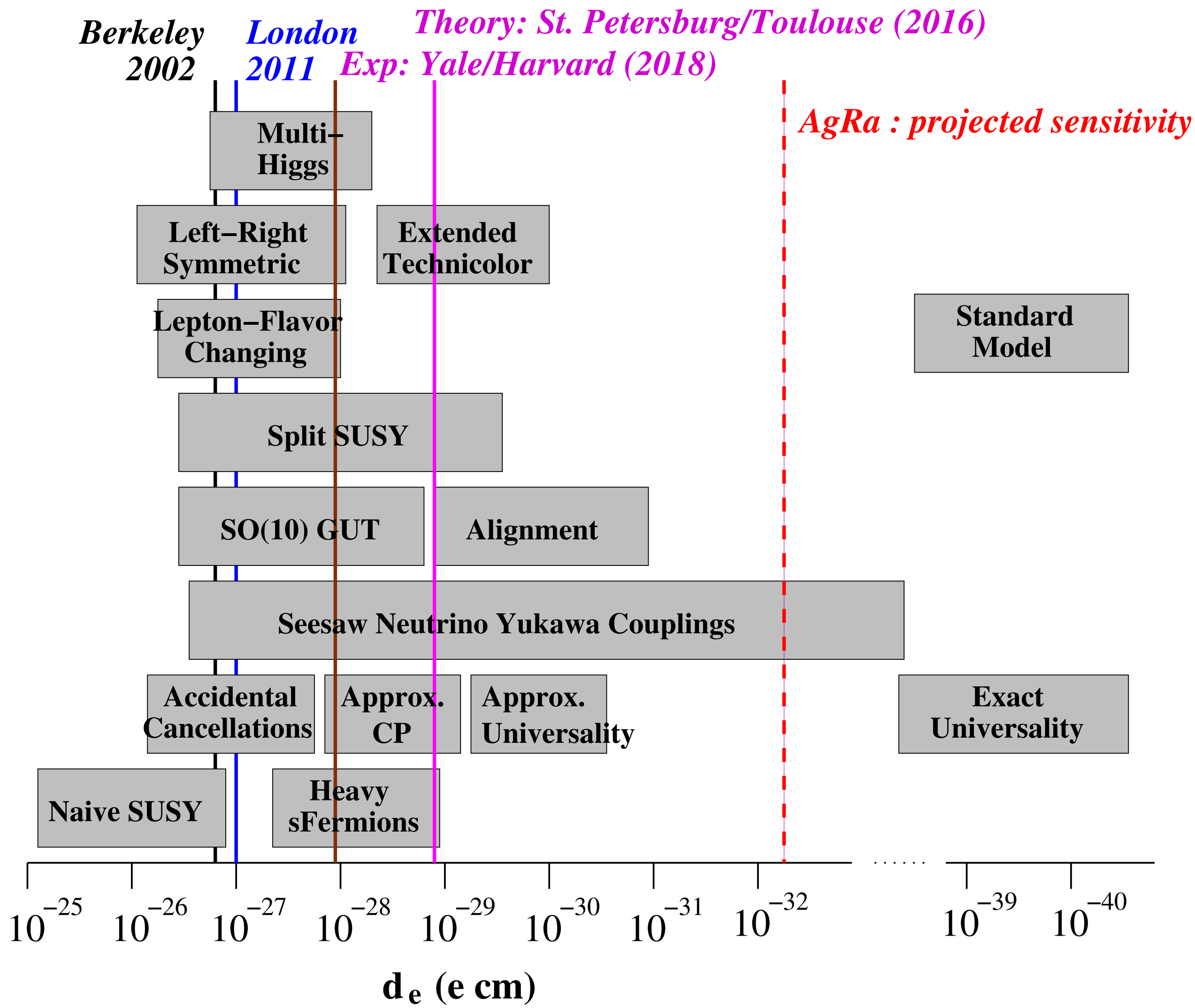
	MBPT[49]	KRCI(FCI)
$^2\Pi$ ^1P(Be $2s^1 2p^1$)	951.6	714.0
$^2\Sigma$ ^1P(Be $2s^1 2p^1$)	1228	1402

Ra(7p) excited states:

$\Omega = 1/2(m)$	$^M\Lambda_\Omega$ term	C_6 [a.u.]	$\Omega = 3/2(k)$	$^M\Lambda_\Omega$ term	C_6 [a.u.]
$m = 1$	$^2\Sigma_{1/2}$	1163	$k = 11$	$^2\Pi_{3/2}$	478
$m = 15$	$^2\Sigma_{1/2}$	1380			
$m = 16$	$^2\Pi_{1/2}$	478			

[49] J.-Y. Zhang, Y. Cheng, J. Mitroy, *J. Phys. B: At. Mol. Opt. Phys.* **46** (2013) *125004*

eEDM Constraint on Beyond-Standard-Model Theories
Single-source interpretation (20??)[51]



[51] TF, D. DeMille, "Using Ultracold Assembled AgRa Molecules to Search for Time-Reversal Violation", to be submitted.

More Science:
A $\mathcal{T}$-Violating Correction to the Weak Interaction ?

- **Nucleon-electron scalar-pseudoscalar $\mathcal{P}, \mathcal{T}$-odd interaction**[52] through higher orders *via* magnetic hyperfine interaction:

$$d_{\text{sys}} = \alpha_{C_S} C_S$$

- **Nucleon-electron tensor-pseudotensor $\mathcal{P}, \mathcal{T}$-odd interaction**[53] *via* expectation value:

$$d_{\text{sys}} = \alpha_{C_T} C_T$$

d_{sys} the system's electric dipole moment
$C_{S,P,T}$ fundamental $\mathcal{CP}$-violating parameters
$\alpha_{C_{S,P,T}}$ atomic interaction constants

[52] T.F., M. Jung *J. High Energy Phys.* **07** (2018) *012*
[53] T.F., *Phys. Rev A* **99** (2019) *012515*

On Constructing Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonians

Lorentz invariance of the Dirac equation

From change of reference frame in Dirac theory

$$\left(-\imath\hbar\gamma^{\mu}\partial_{\mu} + m_0 c\right)\psi(x) = 0 \qquad\qquad \psi \longrightarrow \psi' = S\psi$$

the following condition must hold:

$$S^{-1}(\mathbf{\Lambda})\,\gamma^{\mu}\,S(\mathbf{\Lambda}) \;= \Lambda^{\mu}{}_{\nu}\gamma^{\nu}$$

For a Lorentz boost we have:

$$\mathbf{\Lambda}_{\mathsf{K}\rightarrow\mathsf{K'}_x} = \begin{pmatrix} \gamma & -\frac{v_x}{c}\gamma & 0 & 0 \\ -\frac{v_x}{c}\gamma & \gamma & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \quad \text{where} \quad \gamma = \frac{1}{\sqrt{1-\frac{v_x^2}{c^2}}}$$

Then the above condition directly yields

$$\begin{aligned} S^{-1}(\mathbf{\Lambda}_x)\,\gamma^{0}\,S(\mathbf{\Lambda}_x) \;&= \gamma\gamma^0 - \frac{v_x}{c}\gamma\gamma^1 \\ S^{-1}(\mathbf{\Lambda}_x)\,\gamma^{1}\,S(\mathbf{\Lambda}_x) \;&= -\frac{v_x}{c}\gamma\gamma^0 + \gamma\gamma^1 \\ S^{-1}(\mathbf{\Lambda}_x)\,\gamma^{2}\,S(\mathbf{\Lambda}_x) \;&= \gamma^2 \\ S^{-1}(\mathbf{\Lambda}_x)\,\gamma^{3}\,S(\mathbf{\Lambda}_x) \;&= \gamma^3 \end{aligned} \qquad (1)$$

and we find

$$S(\mathbf{\Lambda}) = a_+ \mathbf{1} + a_- \gamma^0\gamma^1 = \begin{pmatrix} a_+ & 0 & 0 & a_- \\ 0 & a_+ & a_- & 0 \\ 0 & a_- & a_+ & 0 \\ a_- & 0 & 0 & a_+ \end{pmatrix}$$

where $a_\pm = \pm\sqrt{\frac{1}{2}(\gamma \pm 1)}$ and $\gamma^0\gamma^1 = \begin{pmatrix} \mathbf{0} & \sigma_1 \\ \sigma_1 & \mathbf{0} \end{pmatrix}$

The bilinear form (mathematically a scalar)

$$\psi^\dagger\psi = \begin{pmatrix} \psi_1^* & \psi_2^* & \psi_3^* & \psi_4^* \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \\ \psi_4 \end{pmatrix}$$

is **not** a Lorentz scalar (not physically a scalar) ! Formally,

$$\left(\psi^\dagger\psi\right)' = \psi^\dagger S^\dagger S \psi \neq \psi^\dagger\psi$$

essentially because $S^\dagger S \neq \mathbf{1}$.

On Constructing Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonians

Dirac Scalars and Pseudoscalars

However, with $\overline{\psi} := \psi^\dagger\gamma^0$:

$$\left(\overline{\psi}\psi\right)' = \left(\psi'\right)^\dagger \gamma^0\psi' = \psi^\dagger S^\dagger\gamma^0 S\psi = \psi^\dagger\gamma^0\psi = \overline{\psi}\psi$$

In addition, since for space inversion

$$S^{-1}(\mathbf{\Lambda}_{\mathcal{P}})\,\gamma^\mu\,S(\mathbf{\Lambda}_{\mathcal{P}})\; = g^\mu{}_\nu\gamma^\nu$$

the spinor-space transformation is found as

$$S(\mathbf{\Lambda}_{\mathcal{P}}) \equiv \mathcal{P}_s = \gamma^0$$

The above bilinear form under space inversion:

$$\left(\overline{\psi}\psi\right)' = \left(\psi'\right)^\dagger \gamma^0\psi' = \left(\gamma^0\psi\right)^\dagger \gamma^0\gamma^0\psi = \psi^\dagger\gamma^0\gamma^0\gamma^0\psi = \psi^\dagger\gamma^0\psi = \overline{\psi}\psi$$

and so $\overline{\psi}\psi$ is a $true$ scalar.
It is then straightforward to construct a Dirac pseudo-scalar

$$\left(\overline{\psi}\gamma^5\psi\right)' = \left(\psi'\right)^\dagger \gamma^0\gamma^5\psi' = \psi^\dagger\gamma^0\gamma^0\gamma^5\gamma^0\psi = -\psi^\dagger\gamma^0\gamma^5\psi = -\overline{\psi}\gamma^5\psi$$

On Constructing Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonians

Dirac Vectors

A Dirac vector takes on the form

$$\left(\overline{\psi}\gamma^\mu\psi\right)' = \left(\psi'\right)^\dagger\gamma^0\gamma^\mu\psi' = \psi^\dagger\gamma^0\gamma^0\gamma^\mu\gamma^0\psi = \psi^\dagger\gamma^\mu\gamma^0\psi$$

Distinguishing two cases:

1. $\mu = 0 \Rightarrow \left(\overline{\psi}\gamma^0\psi\right)' = \overline{\psi}\gamma^0\psi$

   and so we here have the $\mathcal{P}$-even time-like component of a four-vector. However,

2. $\mu = k \Rightarrow \left(\overline{\psi}\gamma^k\psi\right)' = \psi^\dagger\gamma^k\gamma^0\psi = -\psi^\dagger\gamma^0\gamma^k\psi = -\overline{\psi}\gamma^k\psi$

   which changes sign under space inversion and represents the three components of a vector.

On Constructing Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonians

. . . and pseudovectors

Likewise, the Dirac vector can be turned into a pseudo-vector. With

$$\left(\overline{\psi}\gamma^\mu\gamma^5\psi\right)' = \left(\psi'\right)^\dagger\gamma^0\gamma^\mu\gamma^5\psi' = \psi^\dagger\gamma^0\gamma^0\gamma^\mu\gamma^5\gamma^0\psi = \psi^\dagger\gamma^\mu\gamma^5\gamma^0\psi$$

and again, distinguishing cases,

1. $\mu = 0 \Rightarrow \left(\overline{\psi}\gamma^0\gamma^5\psi\right)' = \psi^\dagger\gamma^0\gamma^0\gamma^0\gamma^5\gamma^0\psi = -\overline{\psi}\gamma^0\gamma^5\psi$

   which is the $\mathcal{P}$-odd time-like component of a four-pseudovector. Similarly,

2. $\mu = k \Rightarrow$
   $\left(\overline{\psi}\gamma^k\gamma^5\psi\right)' = \psi^\dagger\gamma^0\gamma^0\gamma^k\gamma^5\gamma^0\psi = \psi^\dagger\gamma^0\gamma^k\gamma^5\gamma^0\gamma^0\psi = \overline{\psi}\gamma^k\gamma^5\psi$

   which remains even under space inversion and represents the three components of a pseudo-vector.

On Constructing Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonians

Dirac Rank-2 Tensors and Pseudotensors

Introducing the antisymmetric rank-2 tensor

$$\sigma^{\mu\nu} := \frac{\imath}{2}\left(\gamma^\mu\gamma^\nu - \gamma^\nu\gamma^\mu\right)$$

the diagonal of which is zero and which due to $\sigma^{\mu\nu} = -\sigma^{\nu\mu}$ has six linearly independent components.

The following relations hold true:

$$\begin{aligned}
\left(\overline{\psi}\sigma^{0\ell}\psi\right)' &= -\overline{\psi}\sigma^{0\ell}\psi \\
\left(\overline{\psi}\sigma^{k0}\psi\right)' &= -\overline{\psi}\sigma^{k0}\psi \\
\left(\overline{\psi}\sigma^{k\ell}\psi\right)' &= \overline{\psi}\sigma^{k\ell}\psi \qquad \forall k,\ell \in \{1,\ldots,3\}
\end{aligned}$$

Since the transformation is the space inversion and the purely space-like elements of the tensor are even under transformation, $\sigma^{\mu\nu}$ identifies a Dirac rank-2 tensor.

Tensor-Pseudotensor $\mathcal{P}$,$\mathcal{T}$-odd Nucleon-Electron Interaction

Lagrangian density[53] for Ne neutral weak current ($\sigma_{\rho\sigma} = \frac{\imath}{2}(\gamma^\rho\gamma^\sigma - \gamma^\sigma\gamma^\rho)$):

$$\mathcal{L}_{\text{Ne-TPT}} = \frac{1}{2}\frac{G_F}{\sqrt{2}} C_T \sum_N \varepsilon^{\mu\nu\rho\sigma}\, \overline{\psi}_N \Sigma_{N\,\mu\nu} \psi_N\; \overline{\psi}\sigma_{\rho\sigma}\psi$$

Corresponding eff. first-quantized Hamiltonian for infinitely heavy nucleus:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = -\frac{1}{2}\frac{G_F}{\sqrt{2}}\, C_T\, \rho_N(\mathbf{r})\gamma^0 \varepsilon^{\mu\nu\rho\sigma}\Sigma_{N\,\mu\nu}\sigma_{\rho\sigma}$$

Using $\frac{1}{2}\varepsilon^{\mu\nu\kappa\lambda}\sigma_{\kappa\lambda} = -\imath\gamma^5\sigma^{\mu\nu}$ we get:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \frac{\imath G_F}{\sqrt{2}}\, C_T\, \rho(\mathbf{r})\gamma^0 \Sigma_{N\,\mu\nu}\gamma^5\sigma^{\mu\nu}$$

Since $\Sigma_{N\,\mu\nu}\gamma^5\sigma^{\mu\nu} = 2\gamma^0 \boldsymbol{\Sigma}_N \cdot \boldsymbol{\gamma}$ it follows that

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \imath G_F \sqrt{2}\, C_T\, \boldsymbol{\Sigma}_N \cdot \boldsymbol{\gamma}\rho(\mathbf{r})$$

Nuclear state chosen as $|I, M_I = I\rangle$ gives many-electron Hamiltonian:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \imath G_F \sqrt{2}\, C_T\, \langle \Sigma_N \rangle_{\Psi_N} \sum_{j=1}^{n} (\gamma_j)^3\, \rho_N(\mathbf{r}_j)$$

[53] K. Yanase, N. Yoshinaga, K. Higashiyama, N. Yamanaka *Phys. Rev. D* **99** (2019) *075021*

Scalar-Pseudoscalar $\mathcal{P}$,$\mathcal{T}$-odd Nucleon-Electron Interaction

via magnetic hyperfine interaction

Solve for $K \in$ `CI STATES`

$$\left[\sum_{j}^{N}\left[c\,\boldsymbol{\alpha}_j\cdot\mathbf{p}_j+\beta_j c^2-\frac{Z}{r_j}1\!\!1_4\right]+\sum_{j,k>j}^{N}\frac{1}{r_{jk}}1\!\!1_4+\sum_j\mathbf{r}_j\cdot\mathbf{E}_{\text{ext}}\,1\!\!1_4\right]\left|\psi_K^{(0)}\right\rangle=\varepsilon_K^{(0)}\left|\psi_K^{(0)}\right\rangle$$

First-order hyperfine-perturbed CI wavefunctions:

$$\left|\psi_J^{(1)}\right\rangle=\left|\psi_J^{(0)}\right\rangle+\sum_{K\neq J}\frac{\left\langle\psi_K^{(0)}\middle|-\frac{1}{2c\,m_p}\frac{\mu\boldsymbol{I}}{I}\cdot\sum\limits_{i=1}^{n}\frac{\boldsymbol{\alpha}_i\times\boldsymbol{r}_i}{r_i^3}\middle|\psi_J^{(0)}\right\rangle}{\varepsilon_J^{(0)}-\varepsilon_K^{(0)}}\left|\psi_K^{(0)}\right\rangle$$

To leading order the SPS-ne energy shift is

$$(\Delta\varepsilon)_J=\frac{1}{\langle\psi_J^{(1)}|\psi_J^{(1)}\rangle}\left\langle\hat{H}_{\text{S-PS-ne}}\right\rangle_{\psi_J^{(1)}}$$

Atomic EDM due to Ne-SPS interaction $d_a=\alpha_{C_S}C_S$ and so

$$\alpha_{C_S}(\psi_J)=\frac{-A\frac{G_F}{\sqrt{2}}}{E_{\text{ext}}\left\langle\psi_J^{(1)}\middle|\psi_J^{(1)}\right\rangle}\left[\sum_{K\neq J}\frac{\left\langle\psi_K^{(0)}\middle|\hat{H}_{\text{HF}}\middle|\psi_J^{(0)}\right\rangle\left\langle\psi_J^{(0)}\middle|\imath\sum\limits_e\gamma_e^0\gamma_e^5\rho(\mathbf{r}_e)\middle|\psi_K^{(0)}\right\rangle}{\varepsilon_J^{(0)}-\varepsilon_K^{(0)}}+h.c.\right]$$

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

Interaction constants / enhancement factors for n-electron system

- Electron eEDM interaction constant[54] / enhancement

$$W_d := \frac{2\imath c}{\Omega\, e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \vec{p}_j^{\;2} \right\rangle_{\psi_k^{(0)}} \qquad E_{\text{eff}} = -\Omega\, W_d \qquad R \approx R_{\text{lin}} = -\frac{E_{\text{eff}}}{E_{\text{ext}}}$$

- S-PS nucleon-electron interaction constant / ratio

$$W_{\mathcal{S}} := \frac{\imath}{\Omega} \frac{G_F}{\sqrt{2}}\, A \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \rho_N(\vec{r}_j) \right\rangle_{\psi_k^{(0)}} \qquad S = -\frac{\left\langle \imath \sum_j \gamma_j^0\, \gamma_j^5\, \rho_N(\mathbf{r}_j) \right\rangle_{\Psi(E_{\text{ext}})}}{E_{\text{ext}}}$$

- T-PT nucleon-electron interaction constant

$$R_T = \sqrt{2} G_F \left\langle \Sigma_N \right\rangle_{\Psi_N} \left\langle \psi_I^{(0)} \middle| \imath \sum_{j=1}^{n} \left(\gamma_j\right)^3\, \rho(\mathbf{r}_j) \middle| \psi_I^{(0)} \right\rangle$$

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[54] W. Johnson, D.S. Guo, M. Idrees, J. Sapirstein, *Phys. Rev. A* **34** (1986) *1043*
E. Lindroth, E. Lynn, P.G.H. Sandars, *J. Phys. B: At. Mol. Opt. Phys.* **22** (1989) *559*, stratagem II

Generalized Active Spaces

Parameterization of the correlated wavefunction

		# of Kramers pairs	accumulated # of electrons min.	accumulated # of electrons max.
III	*(100 a.u.)* *Virtual Kramers pairs*	111	32	32
II	*5s5p*	4	29	32
I	*4s4p* *3s3p* *2s2p*	12	22	24
	Frozen core	(11)		

Experiment

- **Atomic EDMs : ^{129}Xe**

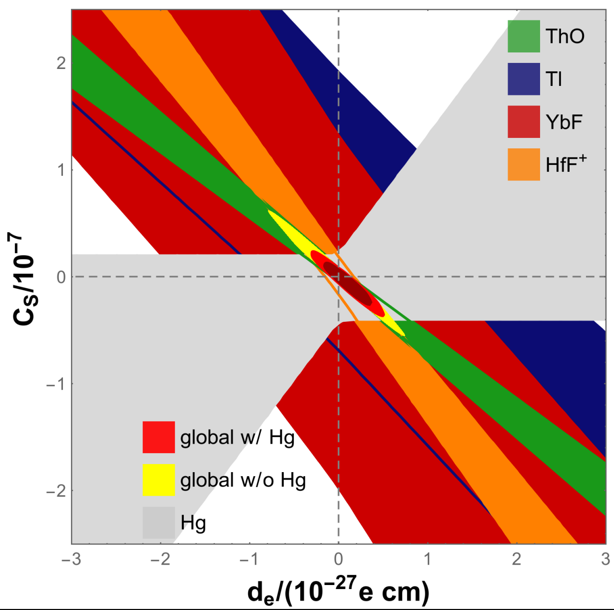
$|d_{\mathrm{Xe}}| < 1.5 \times 10^{-27} e$ cm. Allmendinger *et. al.* (Mainz, Heidelberg), *Phys. Rev. A* **100** (2019) *022505*
$|d_{\mathrm{Xe}}| < 4.81 \times 10^{-27} e$ cm. Sachdeva *et. al.* (Ann Arbor *et. al.*), arXiv:1902.02864 [physics.atom-ph] (2019)
Sato *et. al.* (Tokyo *et. al.*), *Hyperfine Interact.* **230** (2015) *147*

^{129}Xe EDM

Leading[56] (and Subleading) Contributions

$$d_{\mathrm{Xe}} = \rho_Z^N d_N + \alpha_S S - \alpha_{C_T} C_T \; (+\alpha_{C_S} C_S + \alpha_{d_e} d_e)$$

- Atomic coefficients for leading contributions available (α_{C_T}) and in progress (α_S)
- Subleading contributions affect constraints obtained from global fits[57]



Example: Electron EDM and SPS-Ne coupling

- Measurements and calculations on systems with different ratios of atomic/molecular coefficients
- Global fit[57] constrains multiple possible EDM sources

[56] T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **91** (2015) *035502*
[57] W. Dekens, J. de Vries, M. Jung, K.K. Vos, *J. High En. Phys.* **1** (2019) *069*
[57] T. F., M. Jung, *J. High En. Phys.* **7** (2018) *012*

Tensor-Pseudotensor $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

$$R_T(\psi_I\ {}^{129}\text{Xe}) = \sqrt{2}G_F \left\langle \Sigma_N \right\rangle_{\Psi_N} \left\langle \psi_I^{(0)} \right| \imath \sum_{j=1}^{n} \left(\gamma_j\right)^3 \rho(\mathbf{r}_j) \left| \psi_I^{(0)} \right\rangle$$

Model/virtual cutoff (vDZ,vTZ,vQZ) [*a.u.*]	R_T [10^{-20} $\langle\sigma_N\rangle$ e cm]		
		Basis set	
	vDZ	vTZ	vQZ
RPA/-	0.382	0.473	0.485
SD8/80,100,60	0.360	0.438	0.453
SDT8/80,100,60	0.360	0.435	0.450
SDTQ8/80,12,60	0.357	0.431	
SD16/80,100,60	0.406	0.481	0.496
SD8_SDT16/80,100,60	0.405	0.477	
SD18/80,100,60		0.453	
SD24/80,100,60	0.421	0.497	0.514
SD26/80,100,60		0.493	
S16_SD32/80,100,60		0.507	
SD32/80,100,60	0.431	0.508	0.525
SD36/80,100,60	0.417	0.499	
vQZ/SD32/60 $+\Delta$		**0.536**	
Mårtensson-Pendrill[58] RPA		0.52	
Dzuba *et al.*[59] RPA		0.57	
Singh *et al.*[60] CCSD_pT		0.501	

[58] A.M. Mårtensson-Pendrill, *Phys. Rev. Lett.* **54** (1985) *1153*

[59] V.A. Dzuba, V.V. Flambaum, S.G. Porsev, *Phys. Rev. A* **80** (2009) *032120*

[60] Y. Singh, B.K. Sahoo, B.P. Das, *Phys. Rev. A* **89** (2014) *030502(R)*

Scalar-Pseudoscalar $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

$$\alpha_{C_S}(\psi_J\,{}^{129}\mathrm{Xe}) = \frac{-A\frac{G_F}{\sqrt{2}}}{E_{\text{ext}}\left\langle \psi_J^{(1)} \middle| \psi_J^{(1)} \right\rangle} \left[\sum_{K\neq J} \frac{\left\langle \psi_K^{(0)} \middle| \hat{H}_{\text{HF}} \middle| \psi_J^{(0)} \right\rangle \left\langle \psi_J^{(0)} \middle| \imath \sum_e \gamma_e^0 \gamma_e^5 \rho(\mathbf{r}_e) \middle| \psi_K^{(0)} \right\rangle}{\varepsilon_J^{(0)} - \varepsilon_K^{(0)}} + h.c. \right]$$

Basis	# of CI states/X	S [10^{-3} a.u.]	α_{C_S} [10^{-23} e cm]
cvTZ/40 a.u.	8/S8	0.590	0.633
cvTZ/14 a.u.	8/6s6p	0.554	0.594
cvTZ/7 a.u.	8/6s6p5d	0.604	0.648
cvTZ/7 a.u.	8/6s6p5d^Q	0.603	0.647
cvTZ/7 a.u.	8/6s6p5d7p	0.667	0.716
cvTZ/7 a.u.	8/6s6p5d7p7s	0.693	0.744
cvTZ/14 a.u.	8/6s6p5d7p7s	0.694	0.745
cvTZ/14 a.u.	8/6s6p5d7p7s*	0.731	0.784
cvTZ/14 a.u.	8/6s6p5d7p7s**	0.717	0.769
cvTZ/7 a.u.	8/6s6p5d7p7s6d	0.699	0.750
cvTZ/7 a.u.	8/6s6p5d7p7s6d4f	0.702	0.753
cvTZ/7 a.u.	8/6s6p5d7p7s6d4f7d5f8p8s	0.671	0.720
cvTZ/7 a.u.	8/all	0.583	0.625
cvQZ/100 a.u.	8/S8	0.592	0.635
cvQZ/50 a.u.	1000/S8	0.499	0.535
cvQZ/100 a.u.	1281/S8	0.611	0.655
cvQZ/50 a.u.	8/6s6p	0.482	0.517
cvQZ/50 a.u.	8/6s6p5d7p7s	0.710	0.762
vQZ/1281/S8/100 + ΔS_{corr}		**0.661**	**0.709**

Scalar-Pseudoscalar $\mathcal{P}$,$\mathcal{T}$-odd Nucleon-Electron Interaction

Results for ^{129}Xe

- Two major contributions in the sum over states:

$$C_1 = \frac{\langle 5p \rightarrow 6s\ 0{,}0 | \hat{H}_{\mathrm{HF}} | 0{,}0 \rangle \langle 0{,}0 | \imath \sum_e \gamma_e^0 \gamma_e^5 \rho(\mathbf{r}_e) | 5p \rightarrow 6s\ 0{,}0 \rangle}{\varepsilon_{0,0}^{(0)} - \varepsilon_{5p \rightarrow 6s\ 0,0}^{(0)}}$$

$$C_2 = \frac{\langle 5p \rightarrow 6p\ 1{,}0 | \hat{H}_{\mathrm{HF}} | 0{,}0 \rangle \langle 0{,}0 | \imath \sum_e \gamma_e^0 \gamma_e^5 \rho(\mathbf{r}_e) | 5p \rightarrow 6p\ 1{,}0 \rangle}{\varepsilon_{0,0}^{(0)} - \varepsilon_{5p \rightarrow 6p\ 1,0}^{(0)}}$$

$|$hole spinor $\rightarrow$ particle spinor $J, M_J\rangle$

- . . . and a large number of small contributions uncorrected for correlation effects.

Final values for α_{C_T} and α_{C_S} *will lead* to tighter constraints.[62]

[62] T. F., M. Jung, (2020) *in preparation.*