Quantum chemistry for fundamental physics

Molecular probes of New Physics

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HIM
HELMHOLTZ
Helmholtz-Institut Mainz

JG|U

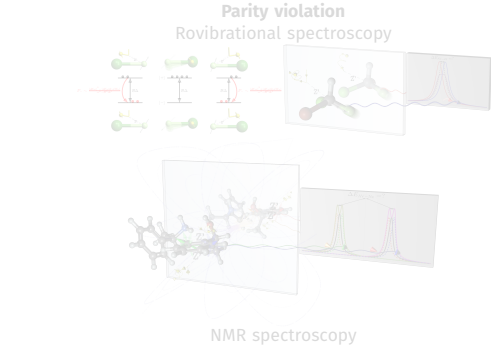
Search for new physics with low-energy precision tests, UG summer school, June 29 to July 4 2025

The plan

The plan

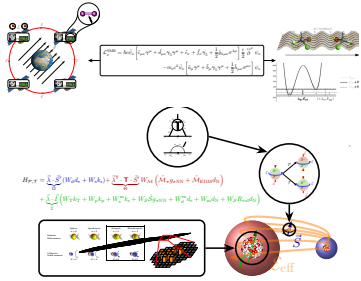
Today:
From the Standard Model to molecular structure theory

- Standard Model of particle physics, cosmology and beyond
- A crash course in relativistic molecular structure



Thursday:
Molecular probes of New Physics

- Molecular parity violation in the Standard Model and beyond
  - Parity violation
  - Lorentz invariance violation
  - New bosons/fifth force
- Electric dipole moments in atoms and molecules



Molecular parity violation

https://www.we-heraeus-stiftung.de/veranstaltungen/parity-violation-in-molecules/venue/

Parity Violation in Molecules

850. WE-Heraeus-Seminar

29 Mar - 01 Apr 2026

Where: Physikzentrum Bad Honnef

Scientific organizers: Prof. Dr. Dmitry Budker, U Mainz ∗ Dr. Konstantin Gaul, U Mainz ∗ Ass. Prof. Dr. Yuval Shagam, Technion, Haifa, Israel

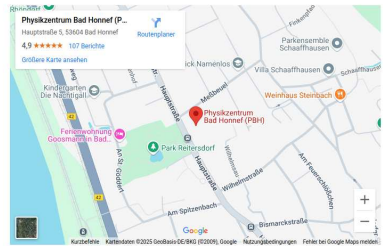
Fundamental symmetry breaking is important to understanding the formation of our universe including baryogenesis and the homochirality of living organisms. It plays an important role in finding a more complete theory than the Standard Model of particle physics. In recent years, molecules have become a leading platform to search for such symmetry breakings through precision metrology as they exhibit enhanced sensitivity and in particular, their many degrees of freedom can be leveraged to isolate violations of fundamental symmetries from the background. However, in recent years, the molecular precision spectroscopy community has become focused on simultaneous parity and time reversal symmetry violation, overlooking the equal importance of time-reversal symmetry even, parity violating effects. For example, despite its prediction in the 1960s, the parity violating energy difference between mirror-images of chiral molecules is yet to be observed, which is crucial for our understanding of molecular chirality. Only recently several individual works rediscovered the unique opportunities of molecular parity violation, indicating even its undiscovered potential in revealing new physics. Moreover, recent advancements in molecular quantum control and molecular theory may bring the detection of molecular parity violation now in reach for the first time.

This workshop will use the momentum of these recent advances to refocus the attention on molecular parity violation experiments and foster collaboration in the field.

The conference language will be English. The Wilhelm and Else Heraeus-Foundation bears the cost of full-board accommodation for all participants.

Venue

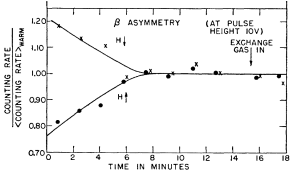
The seminar takes place at the Physikzentrum Hauptstr. 5, 53604 Bad Honnef, Germany. Lectures, accommodation and meals will all take place at the Physikzentrum. Accommodation will be booked for admitted participants automatically by the Wilhelm and Else Heraeus Foundation.



- Discovery of “two” new particles with **same life-time and mass**

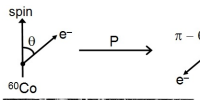
$$\theta^+ \to \underbrace{\pi^+ + \pi^0}_{P=+1}, \quad \tau^+ \to \underbrace{\left\{ \begin{array}{l} \pi^+ + \pi^- + \pi^+ \\ \pi^+ + \pi^0 + \pi^0 \end{array} \right\}}_{P=-1}$$

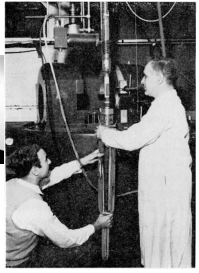
$\to$ Lee and Yang in 1956:
Parity of weak interaction was not tested!
Proposal of several experimental tests!

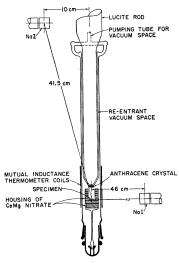


Nature is left handed!

β-decay: $^{60}_{27}\mathrm{Co} \to ^{60}_{28}\mathrm{Ni} + e^- + \overline{\nu}_e$







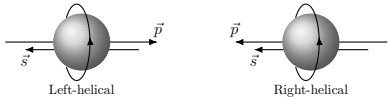
T. D. Lee, C. N. Yang, *Phys. Rev.* **1956**, *104*, 254–258, C. S. Wu et al., *Phys. Rev.* **1957**, *105*, 1413–1415.

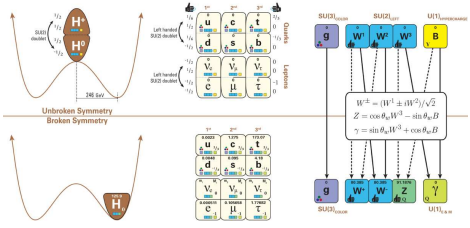
Glashow-Weinberg-Salam model:

- Idea spontaneous symmetry breaking/superposition of interactions

$\Rightarrow$ Definition of left- and right-handed particle spinors:

$$\psi_{\mathrm{L}} \equiv \frac{1-\boldsymbol{\gamma}^5}{2}\psi, \qquad \psi_{\mathrm{R}} \equiv \frac{1+\boldsymbol{\gamma}^5}{2}\psi, \qquad \psi = \psi_{\mathrm{L}} + \psi_{\mathrm{R}}$$





https://commons.wikimedia.org/wiki/File:Standard_Model_Of_Particle_Physics,_Most_Complete_Diagram.jpg

- Weak mixing angle (**Weinberg angle**) $\theta_{\mathrm{W}} = 28.75°$
- Connection to electromagnetic coupling constant:
  $g_W = \frac{g_e}{\sin\theta_{\mathrm{W}}},\ g_Z = \frac{g_e}{\sin\theta_{\mathrm{W}}\cos\theta_{\mathrm{W}}}$
- Weak fine structure constant $\alpha_{\mathrm{W}} = \frac{1}{29.5} \sim 5\alpha$
  $M_W = 80.40(3)\,\mathrm{GeV}/c^2,\ M_Z = 91.188(2)\,\mathrm{GeV}/c^2$

$\Rightarrow$ Yukawa-like $\sim \eta_{\mu\nu}\frac{\exp(-\hbar/(Mc)r)}{r}$ for
$E \ll 80\,\mathrm{GeV} \rightarrow \frac{\imath\eta_{\mu\nu}}{(Mc)^2}\delta^3(r)$

$\rightarrow$ Weak interaction weak due to large boson masses

S. L. Glashow, *Nucl. Phys.* **1961**, *22*, 579–588, S. Weinberg, *Phys. Rev. Lett.* **1967**, *19*, 1264–1266, A. Salam in Proceedings of the Eighth Nobel Symposium, *Vol. 680519*, (Ed.: N. Svartholm), Amkvist and Wiksell, Stockholm, **1968**, pp. 367–377.

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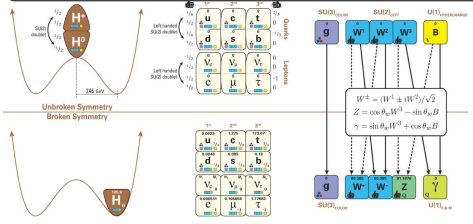
$$\psi_L = \frac{1-\gamma^5}{2}\psi \qquad \psi_R = \frac{1+\gamma^5}{2}\psi \qquad \psi = \psi_L + \psi_R$$

Weinberg angle cannot be calculated in the Standard Model!



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- Connection to electromagnetic coupling constant:
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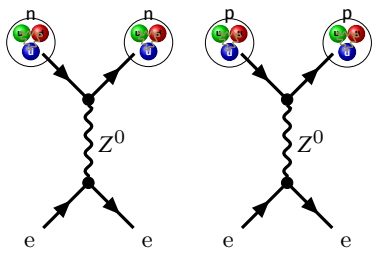
- In a molecule or atom we have e, $p = |uud\rangle$ and $n = |udd\rangle$
- Weak electron-electron scattering negligible V. G. Gorshkov et al., *Zhurnal Eksperimentalnoi I Teoreticheskoi Fiziki* **1977**, *72*, 1268–1274
- Vertex factors $\frac{-\imath g_Z}{2}\gamma^\mu\left(c_{\mathrm{V}}^f - c_{\mathrm{A}}^f\gamma^5\right)$
- Ignoring virtual quarks/anti-quarks
  → summation over quark vertices to obtain p and n vertex factors
- $E \ll M_Z c^2 \rightarrow D_{\mu\nu}(p) \sim \frac{\eta_{\mu\nu}}{(M_Z c)^2} \rightarrow$ Fermi's constant
  $G_{\mathrm{F}} = \frac{\sqrt{2}}{8}\left(\frac{g_Z}{M_Z c^2}\right)^2 \hbar^3 c^3$
- The interaction Lagrangian:



$$\mathcal{L}_{e(n/p)} = -\sqrt{2}G_{\mathrm{F}}\left[\bar{\psi}_e\gamma_\mu\left(c_{\mathrm{V}}^e - c_e^{\mathrm{A}}\gamma^5\right)\psi_e\right]\left[\bar{\psi}_{n/p}\gamma^\mu\left(c_{\mathrm{V}}^{n/p} - c_{\mathrm{A}}^{n/p}\gamma^5\right)\psi_{n/p}\right]$$

$$\mathcal{L}_{en} = -\frac{G_{\mathrm{F}}}{2\sqrt{2}}\left[\bar{\psi}_e\gamma_\mu\left(1 - 4\sin^2\theta_{\mathrm{W}} + \gamma^5\right)\psi_e\right]\left[\bar{\psi}_n\gamma^\mu\left(-1 - \gamma^5\right)\psi_n\right]$$

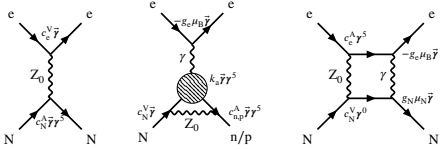
$$\mathcal{L}_{ep} = -\frac{G_{\mathrm{F}}}{2\sqrt{2}}\left[\bar{\psi}_e\gamma_\mu\left(1 - 4\sin^2\theta_{\mathrm{W}} + \gamma^5\right)\psi_e\right]\left[\bar{\psi}_p\gamma^\mu\left(1 - 4\sin^2\theta_{\mathrm{W}} + \gamma^5\right)\psi_p\right]$$

R. Berger in Relativistic Electronic Structure Theory, Part: 2, Applications, (Ed.: P. Schwerdtfeger), Elsevier, Netherlands, **2004**, Chapter 4, pp. 188–288.

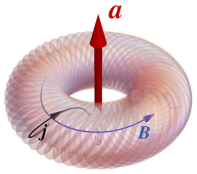
- Nucleons (quarks) are non-relativistic: $\bar{\psi}_{\mathrm{p,n}}\boldsymbol{\gamma}^0\psi_{\mathrm{p,n}} = \rho_{\mathrm{p,n}}(\vec{r})$ and $\bar{\psi}_{\mathrm{p,n}}\vec{\boldsymbol{\gamma}}\boldsymbol{\gamma}^5\psi_{\mathrm{p,n}} = \vec{\boldsymbol{\Sigma}}_{\mathrm{p,n}}(\vec{r})$ survive!
- Assuming $N\rho_{\mathrm{n}} + Z\left(1 - 4\sin^2\theta_{\mathrm{W}}\right)\rho_{\mathrm{p}} \approx Q_{\mathrm{W}}\rho_{\mathrm{nuc}}(\vec{r})$ and $\left\langle \Psi_{\mathrm{N}} \middle| \sum_{\mathrm{n}} \vec{\boldsymbol{\Sigma}}_{\mathrm{p}} - \sum_{\mathrm{p}} \vec{\boldsymbol{\Sigma}}_{\mathrm{p}} \middle| \Psi_{\mathrm{N}} \right\rangle \approx \lambda_{\mathrm{PV}}\rho_{\mathrm{nuc}}\vec{I}$

$$\mathcal{L}_{\mathrm{eN,PV}} \approx -\frac{G_{\mathrm{F}}}{2\sqrt{2}}\left[\bar{\psi}_e\boldsymbol{\gamma}^0\boldsymbol{\gamma}^5\psi_e\right]Q_{\mathrm{W}}\rho_{\mathrm{nuc}}(\vec{r}) - \kappa_{\mathcal{A}}\frac{G_{\mathrm{F}}}{2\sqrt{2}}\left[\bar{\psi}_e\vec{\boldsymbol{\gamma}}\psi_e\right]\cdot\vec{I}\rho_{\mathrm{nuc}}(\vec{r})\,, \qquad \kappa_{\mathcal{A}} = -\lambda_{\mathrm{PV}}\left(1 - 4\sin^2\theta_{\mathrm{W}}\right) + k_{\mathrm{a}} + k_{\mathrm{hf}}$$

- $Q_{\mathrm{W}} = \left(1 - 4\sin^2\theta_{\mathrm{W}}\right)Z_A - N_A$ is the **weak charge**.
  Note: $\sin^2\theta_{\mathrm{W}} \approx \frac{1}{4} \rightarrow Q_{\mathrm{W}} \approx -N_A$



Nuclear anapole moment $k_{\mathrm{a}}\vec{I} \propto A^{2/3}$



Tough for nuclear theory! (error bars $\mathcal{O}(100\,\%)$)
Missing motivation for higher level of accuracy?

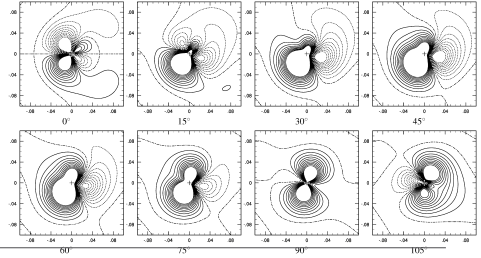
M. S. Safronova et al., *Rev. Mod. Phys.* **2018**, *90*, 025008.

$$\hat{H}_{\mathrm{PV}} = \frac{G_{\mathrm{F}}}{2\sqrt{2}} \sum_{A}^{\text{nuclei}} \sum_{i}^{\text{elec}} \left[Q_{\mathrm{W},A} \rho_A(\vec{r}_i) \boldsymbol{\gamma}_i^5 + \kappa_{\mathcal{A},A} \rho_A(\vec{r}_i) \vec{\boldsymbol{\alpha}}_i \cdot \vec{I}_A \right]$$

- $G_{\mathrm{F}} = 2.22249 \times 10^{-14} E_{\mathrm{h}}/a_0^3$
- $\rho_A(\vec{r})$ is the **normalized nuclear charge distribution**
  - point-like nucleus: $\rho_A(\vec{r}) = \delta(\vec{r} - \vec{r}_A)$
  - Gaussian shaped nucleus: $\rho_A(\vec{r}) = (\frac{\zeta_A}{\pi})^{3/2} \exp(-\zeta_A |\vec{r} - \vec{r}_A|^2)$

- In the non-relativistic limit: $\boldsymbol{\gamma}^5 \rightarrow \frac{\vec{\boldsymbol{\sigma}} \cdot \hat{\vec{p}}}{m_{\mathrm{e}} c}$
  $\Rightarrow$ Measure of the helicity

Example $\boldsymbol{\gamma}^5$ H_2Te_2:



Measure of helicity ($\boldsymbol{\gamma}^5$) of the electron cloud at the nucleus (ρ)
$\Rightarrow$ Requires mixing of $s_{1/2}$ and $p_{1/2}$ states!

R. A. Hegstrom et al., *J. Chem. Phys.* **1980**, *73*, 2329–2341, R. Bast et al., *Phys. Chem. Chem. Phys.* **2011**, *13*, 864–876.

Molecular parity violation

Isn't it hopeless?—A short history of atomic PV

1959 Zel'dovich proposes the PV odd nuclear anapole moment

1959 Zel'dovich notices that electroweak PV makes atoms slighly chiral ⇒ optical rotation in atoms

1970 Poppe tries first measurement of optical rotation of Pb vapor in Amsterdam

1975 M.-A. & C. Bouchiat extend Fermi-Segré to show Z^3 enhancement in heavy atoms

1975 Lethokov: PV should lead to an energy difference between enantiomers of chiral molecules

1978-1986 First measurements of atomic PV

- In Novosibirsk, Seattle, Oxford and Moscow with Bi
- In Paris and Boulder with Cs
- In Berkeley, Seattle and Oxford with Tl
- In Seattle with Pb

1995 PV measured to 1-2 % in Cs, Tl, Bi, Pb

1997 Measurement of the nuclear anapole moment of Cs

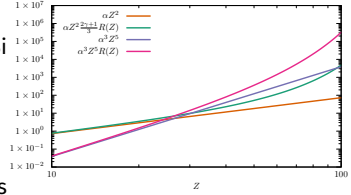
Scaling with Z and α for electroweak PV:
Nuclear-spin independent PV (NSIPV)

$$\Delta_{\mathrm{NSIPV}} \sim \frac{\langle s_{1/2} | \hat{H}_{\mathrm{NSIPV}} | p_{1/2} \rangle \langle p_{1/2} | \hat{H}_{\mathrm{so}} | s_{1/2} \rangle}{E_i - E_a}$$

$$\sim \frac{G_{\mathrm{F}} \alpha^3}{2\sqrt{2}} \boldsymbol{Z^5} R(Z, A)$$

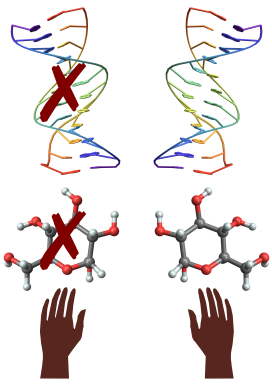
Nuclear-spin dependent PV (NSDPV)

$$\Delta_{\mathrm{NSDPV}} \sim \langle s_{1/2} | \hat{H}_{\mathrm{NSDPV}} | p_{1/2} \rangle \sim \frac{G_{\mathrm{F}} \alpha}{\sqrt{2}} \boldsymbol{Z^2} R(Z, A) \frac{2\gamma + 1}{3}$$



R. Berger, J. Stohner, *Wiley Interdiscip. Rev.-Comput. Mol. Sci.* **2019**, *9*, e1396.

Biomolecular homochirality



Close spacing of levels of opposite parity $\Delta_{\mathrm{PV}} \sim \dfrac{\langle a|\hat{H}_{\mathrm{PV}}|b\rangle\langle b|-e\vec{\mathcal{E}}\cdot\vec{r}|a\rangle}{E_a - E_b}$

	Atoms	Polar diatomic molecules	Chiral molecule
$E_a - E_b$	$\sim$ GHz Electronic	$\sim$ MHz Rotational	$\ll$ Hz Tunneling splitting

FULL PAPER

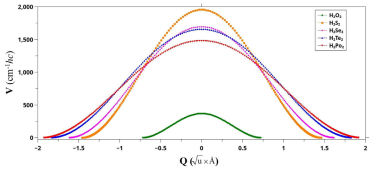
INTERNATIONAL JOURNAL OF QUANTUM CHEMISTRY WILEY

Instanton calculations of tunneling splittings in chiral molecules

Nityananda Sahu[1] | Jeremy O. Richardson[2] | Robert Berger[1]

[1]Fachbereich Chemie, Theoretische Chemie, Philipps Universität Marburg, Marburg, Germany

Abstract



Isotopes	H_2O_2	HSOH	H_2S_2	H_2Se_2	H_2Te_2	H_2Po_2
H	1.4×10^{-6}	1.7×10^{-4}	1.5×10^{-7}	1.1×10^{-7}	9.6×10^{-9}	9.4×10^{-9}
D	9.1×10^{-10}	4.0×10^{-7}	1.5×10^{-11}	7.5×10^{-12}	2.2×10^{-13}	2.2×10^{-13}
T	6.8×10^{-12}	6.6×10^{-9}	2.0×10^{-14}	6.1×10^{-15}	8.1×10^{-17}	9.6×10^{-17}

s ($\mathrm{cm}^{-1}hc$) for the
ules at CCSD(T) level
CCSD(T) levels of

$\Delta E_{\mathrm{Lowt.\ CCSD(T)}}$
16.3

TABLE 7 Trans-tunneling splittings ΔE_S ($\mathrm{cm}^{-1}hc$) calculated for S_2Cl_2 as a function of the number of beads N and the imaginary-time-duration $\beta\hbar$ (in $E_h^{-1}\hbar$)

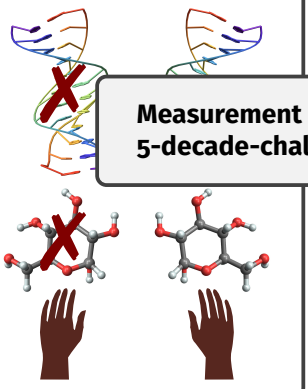
$N \downarrow$ $\beta\hbar \rightarrow$	20,000.0	30,000.0	40,000.0
32	1.3×10^{-96}	2.1×10^{-96}	1.9×10^{-95}

$\Rightarrow \Delta E$ 287 Hz in H_2Te_2
and ΔE 10^{-87} Hz in S_2Cl_2!

R. Berger in Relativistic Electronic Structure Theory, Part: 2, Applications, (Ed.: P. Schwerdtfeger), Elsevier, Netherlands, **2004**, Chapter 4, pp. 188–288, R. Berger, J. Stohner, *Wiley Interdiscip. Rev.-Comput. Mol. Sci.* **2019**, 9, e1396.

D. G. Blackmond, *Cold Spring Harb Perspect Biol* **2010**, 2, a002147[PII], a002147, Q. Sallembien et al., *Chem. Soc. Rev.* **2022**, 51, 3436–3476.

Biomolecular homochirality



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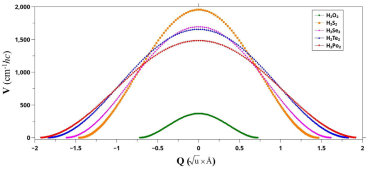
	Atoms	Polar diatomic molecules	Chiral molecule
$E_a - E_b$	$\sim$ GHz Electronic	$\sim$ MHz Rotational	$\ll$ Hz Tunneling splitting

Measurement of molecular parity violation is pending
5-decade-challenge to see enantiomeric energy difference!



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Molecular parity violation

Parity violation in diatomic molecules

PHYSICAL REVIEW LETTERS **120,** 142501 (2018)

Editors' Suggestion

Demonstration of a Sensitive Method to Measure Nuclear-Spin-Dependent Parity Violation

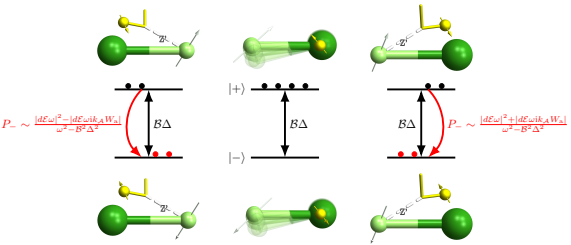
Emine Altuntaş,* Jeffrey Ammon,† Sidney B. Cahn,‡ and David DeMille§

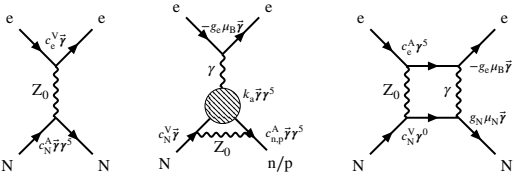
Department of Physics, Yale University, P.O. Box 208120, New Haven, Connecticut 06520, USA

(Received 22 December 2017; revised manuscript received 16 February 2018; published 2 April 2018)

Nuclear-spin-dependent parity violation (NSD-PV) effects in atoms and molecules arise from Z^0 boson exchange between electrons and the nucleus, and from the magnetic interaction between electrons and the parity-violating nuclear anapole moment. We demonstrate measu… ^{138}Ba^{19}F, …that use an enhancement of the effect in diatomic molecules, here using the test … sensitivity surpasses that of any previous … systematic errors can be suppressed to at least the leve… matrix element W of the NSD-PV interaction with to… configurations where W must have different signs. T… PV effects of the size anticipated across a wide range of nuclei including ^{137}Ba in ^{137}BaF, where $|W|/(2\pi) \approx 5$ Hz is expected.

$$\delta W/(2\pi) < 0.7 \text{ Hz}$$





$$\hat{H}_{\mathrm{PV,sr}} = k_{\mathcal{A},A} W_a \vec{I} \times \vec{S}' \cdot \vec{I}_A$$

$$W_a = \left\langle \Omega \left| \sum_i^{N_{\mathrm{elec}}} \vec{\alpha}_i \rho_A(\vec{r}_i) \right| -\Omega \right\rangle / \Omega$$

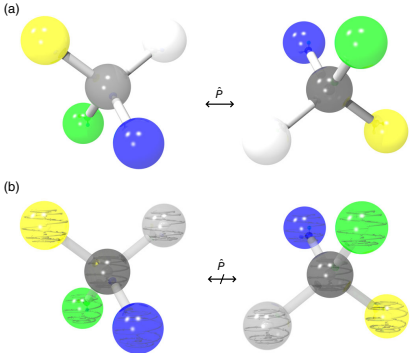
$$\overset{\text{leading order}}{=} \left\langle \Omega \left| \sum_i^{N_{\mathrm{elec}}} \left\{ \hat{\vec{p}}_i, \rho_A(\vec{r}_i) \right\} + \vec{\sigma}_i \times \left[\vec{\nabla}_i, \rho_A(\vec{r}_i) \right] \right| -\Omega \right\rangle / \Omega$$

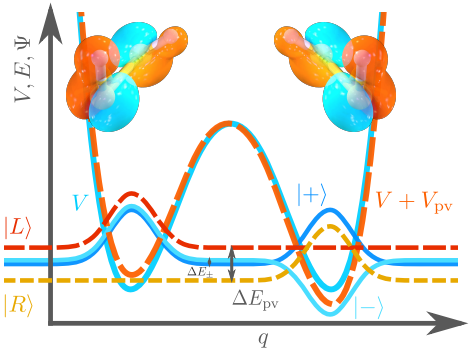
$$\boldsymbol{H}_{\pm\Omega}(t) = \begin{pmatrix} 0 & d\mathcal{E}(t) + \mathrm{i} k_{\mathcal{A}} W_a \\ d\mathcal{E}(t) - \mathrm{i} k_{\mathcal{A}} W_a & \Delta\mathcal{B} \end{pmatrix}$$

High sensitivity to anapole moment
Weak charge effect suppressed by $\frac{m_e}{m_p}$
Complementary to atomic PV!

Molecular parity violation

PV energy difference between enantiomers

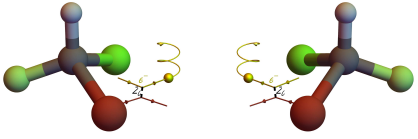




- Enantiomers connected by $\mathcal{P}$
- Weak interaction makes atoms chiral

$\Rightarrow$ Enantiomers become diastereomers

$\Rightarrow$ Parity violating energy difference

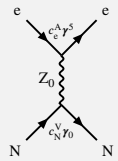


R. Berger, J. Stohner, *Wiley Interdiscip. Rev.-Comput. Mol. Sci.* **2019**, 9, e1396, P. Schwerdtfeger in Computational Spectroscopy: Methods, Experiments and Applications, (Ed.: J. Grunenberg), Wiley, Netherlands, **2010**, Chapter 7, pp. 201–221.

Molecular parity violation

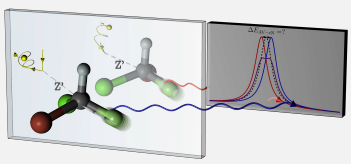
Parity violation in chiral molecules an overview

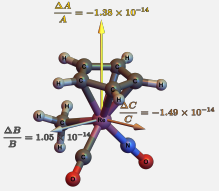
Nuclear spin-independet PV



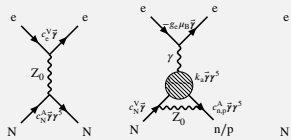
Electronic, vibrational and rotational spectroscopy

$$\frac{\Delta E_{\mathrm{el,PV}}}{E_{\mathrm{el}}} \approx \frac{\Delta E_{\mathrm{vib,PV}}}{E_{\mathrm{vib}}} \approx \frac{\Delta E_{\mathrm{rot,PV}}}{E_{\mathrm{rot}}}$$

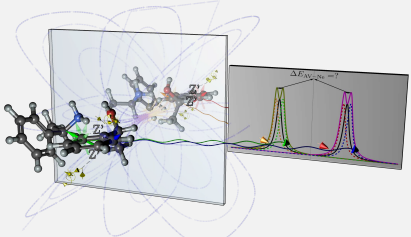




Nuclear spin-dependet PV



NMR, ESR



R. Berger, J. Stohner, *Wiley Interdiscip. Rev.-Comput. Mol. Sci.* **2019**, *9*, e1396.

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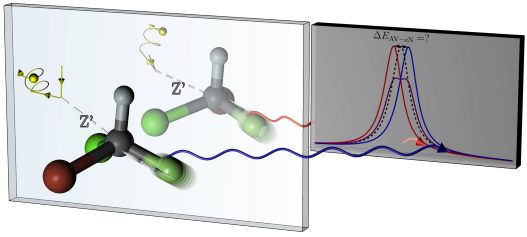
Best limit on electroweak parity violation in chiral molecules from CHBrClF-Experiment

VOLUME 83, NUMBER 8 PHYSICAL REVIEW LETTERS 23 AUGUST 1999

Limit on the Parity Nonconserving Energy Difference between the Enantiomers of a Chiral Molecule by Laser Spectroscopy

Ch. Daussy, T. Marrel, A. Amy-Klein, C. T. Nguyen, Ch. J. Bordé,* and Ch. Chardonnet†
Laboratoire de Physique des Lasers, UMR 7538 CNRS, Université Paris 13, Avenue J.-B. Clément, 93430 Villetaneuse, France
(Received 14 April 1999)

We have developed a saturation spectroscopy experiment to test the prediction that enantiomers of chiral molecules have different spectra because of the parity violation associated with neutral currents in the weak interaction between electrons and nuclei. First experimental tests have been conducted on hyperfine components of vibration-rotation transitions of CHFClBr in the 9.3 μm spectral range. The frequencies of saturation resonances of separated enantiomers have been compared and found to be identical within 13 Hz ($\Delta\nu/\nu < 4.10^{-13}$).



$40_{7,34} \leftarrow 40_{8,33}$ transition ($J'_{K'_a,K'_c} \leftarrow J''_{K''_a,K''_c}$) of
C–F stretching fundamental studied

$\Rightarrow$**Resolution** $\frac{\Delta\nu}{\nu} < 4 \times 10^{-13}$
3 years later factor-4-improvement

- Leading order PV shifts of rotational constants:

$$\Delta I_{xx} = 2m_A(y_A\,\delta_{\mathrm{PV}}\,y_A + z_A\,\delta_{\mathrm{PV}}\,z_A),\ \frac{\Delta X_{\mathrm{R}}}{X_{\mathrm{R}}} \approx -\frac{\Delta I_{xx}}{I_{xx}},\ \delta_{\mathrm{PV}}\vec{R} = -\boldsymbol{M}^{-1/2}\boldsymbol{A}(\boldsymbol{A}^{\top}\boldsymbol{H}_{\mathrm{MW}}\boldsymbol{A})^{-1}\boldsymbol{A}^{\top}\boldsymbol{M}^{-1/2}\vec{\nabla}E_{\mathrm{PV}}$$

- Leading order PV shifts in vibrational spectra

$$\langle\Psi_{n_r}|E_{\mathrm{PV}}(\vec{q})|\Psi_{n_r}\rangle \approx E_{\mathrm{PV}}(\vec{R}_0) + \frac{1}{2}\sum_r\left(n_r+\frac{1}{2}\right)\left(\frac{\partial^2 E_{\mathrm{PV}}}{\partial q_r^2} - \sum_s \frac{1}{\hbar\omega_s}\frac{\partial^3 V_{\mathrm{BO}}}{\partial q_r^2 \partial q_s}\frac{\partial E_{\mathrm{PV}}}{\partial q_s}\right)$$

A. D. Buckingham, W. Urland, *Chem. Rev.* **1975**, *75*, 113–117.
M. Quack, J. Stohner, *Z. Phys. Chem.* **2000**, *214*, 675–703, M. Quack, M. Willeke, *Helv. Chim. Acta* **2003**, *86*, 1641–1652.
K. Gaul et al., *Phys. Rev. A* **2020**, *102*, 032816, K. J. Gaul, Dr. rer. nat. thesis, Philipps-Universität Marburg, **2020**, S. A. Brück et al., *J. Chem. Phys.* **2023**, *158*, 194109.
G. Rauhut, P. Schwerdtfeger, *Phys. Rev. A* **2021**, *103*, 042819.

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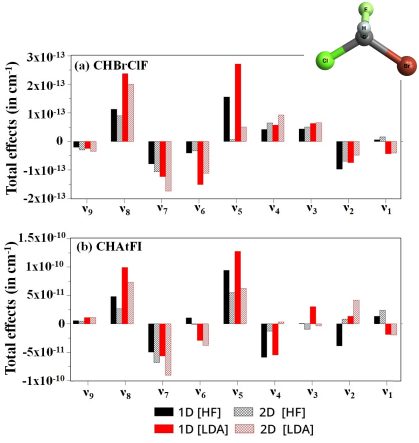
- Separable anharmonic adiabatic approximation (C–F stretch):

$$\langle E_{\mathrm{PV}}\rangle \approx \sum_{i=1}^{N}\frac{1}{i!}\langle q_4^i\rangle\left(\frac{\partial^i}{\partial q_4^i}\langle\gamma^5\rangle\right) \approx \frac{1}{2}\left(v_4+\frac{1}{2}\right)\left[\frac{\partial^2}{\partial q_4^2}\langle\gamma^5\rangle+\frac{\phi_{444}}{\tilde{\nu}_4}\frac{\partial}{\partial q_4}\langle\gamma^5\rangle\right]$$



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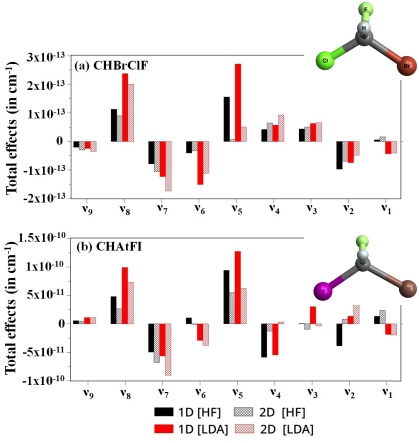
- **Non-separable anharmonic** effects can be of same order!

$$\frac{1}{2}\left(v_4+\frac{1}{2}\right)\left[\sum_{r\neq 4}\frac{\phi_{44r}}{\tilde{\nu}_r}\frac{\partial}{\partial q_r}\langle\gamma^5\rangle\right]$$



A. D. Buckingham, W. Urland, *Chem. Rev.* **1975**, *75*, 113–117.
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- Leading order PV shifts in vibrational spectra

$$\langle\Psi_{n_r}|E_{\mathrm{P}}$$

Reliable predictions of vibrational shifts can be tough!
Rotations are easier.

- Separa

$$\langle E_{\mathrm{PV}}\rangle \not\approx \sum_{i=1}^{N}\frac{1}{i!}\langle q_4^i\rangle\left(\frac{\partial^i}{\partial q_4^i}\langle\gamma^5\rangle\right) \approx \frac{1}{2}\left(v_4+\frac{1}{2}\right)\left[\frac{\partial^2}{\partial q_4^2}\langle\gamma^5\rangle + \frac{\phi_{444}}{\tilde{\nu}_4}\frac{\partial}{\partial q_4}\langle\gamma^5\rangle\right]$$

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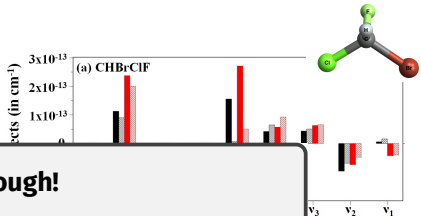

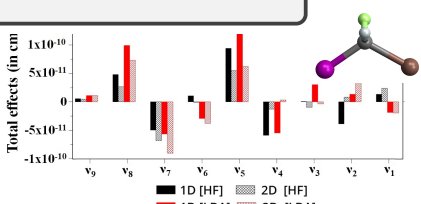

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Molecular parity violation

PV rotational shifts in experimentally well-characterised chiral CpRe(CO)(NO)(CH_3)

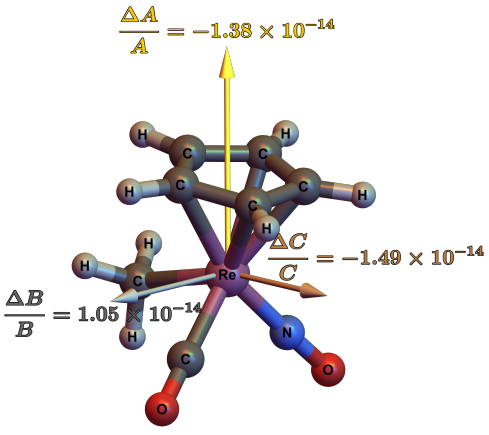
DOI: 10.1002/anie.20140607

Parity Violation

High-Resolution Spectroscopy of the Chiral Metal Complex [CpRe(CH_3)(CO)(NO)]: A Potential Candidate for Probing Parity Violation**

*Chris Medcraft, Robert Wolf, and Melanie Schnell**

- ✓ Vapor pressure and gas phase stability
- ✓ Suitability for precision spectroscopy
- ✓ Controlling enantiomeric excess with microwave three-wave mixing
- ✓ Nuclear electric quadrupole tensor to benchmark DFT
- ✓ Large PV shifts of rotational constants
- ✗ Predictions of vibrational shift very complicated due to **huge multi-mode effects**

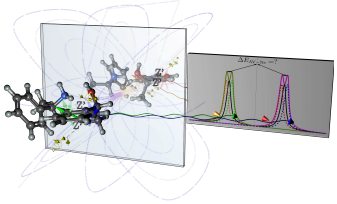


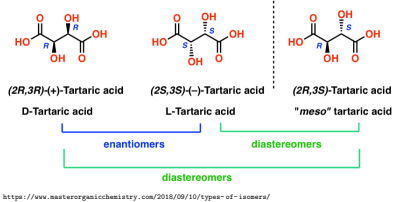
N. Sahu et al., *arXiv* **2023**, *physics.chem-ph*, 2303.08263.

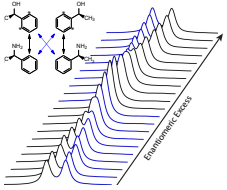
E. Hirota, *Proc. Jpn. Acad. Ser. B* **2012**, *88*, 120–128, D. Patterson et al., *Nature* **2013**, *497*, 475–477.

- Nuclear spin-dependent PV splits chemical shift of enantiomers $\Delta_{\mathrm{PV}}\sigma_{kl} = \Delta_{(R,S)}\frac{\partial^3 E}{\partial \mathcal{B}_k \partial \mathbf{I}_l \partial \boldsymbol{\kappa}_{\mathcal{A}}} \lesssim \mathrm{mHz}$
- Line width in solution NMR spectroscopy is typically $> 0.5\,\mathrm{Hz}$

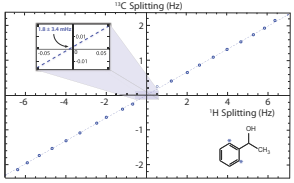
$\Rightarrow$ Detecting PV in solution NMR **was considered** to be **hopeless**!







- ✗ Soluable compounds
- ✗ High NMR sensitivity (large γ and abundance)
- ✗ No or few conformers
- ✗ One-sided diastomeric interaction with chiral titrant
- ✗ PV shifts $O(\mathrm{mHz})$



A. L. Barra et al., *Phys. Lett. A* **1986**, *115*, 443–447, A. L. Barra et al., *Europhys. Lett.* **1988**, *5*, 217–222, G. Laubender, R. Berger, *ChemPhysChem* **2003**, *4*, 395–399, S. Nahrwold, R. Berger, *J. Chem. Phys.* **2009**, *130*, 214101, J. Eills et al., *Phys. Rev. A* **2017**, *96*, 042119.

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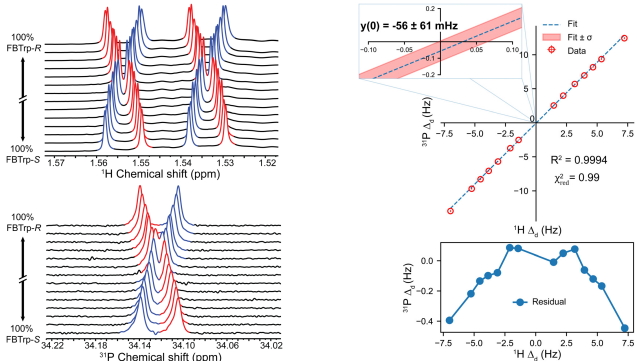
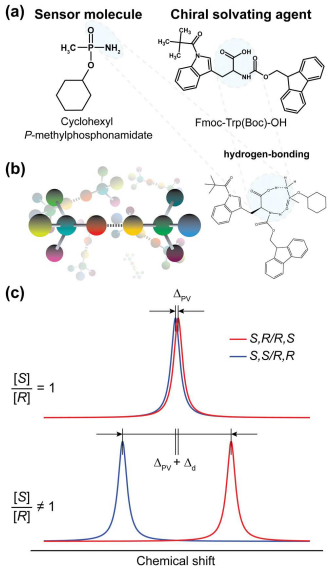
Cite this: *Phys. Chem. Chem. Phys.*, 2025, **27**, 6092

Towards detection of molecular parity violation *via* chiral co-sensing: the $^{1}H/^{31}P$ model system†

Erik Van Dyke, [abc] James Eills,[d] Kirill Sheberstov,[e] John Blanchard,[f] Manfred Wagner,[g] Andrés Emilio Wedenig,[h] Konstantin Gaul, [abch] Robert Berger, [h] Rudolf Pietschnig, [i] Denis Kargin, [i] Danila A. Barskiy [abc] and Dmitry Budker [abcj]





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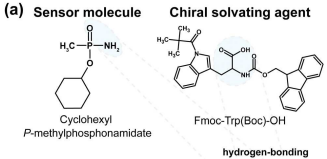
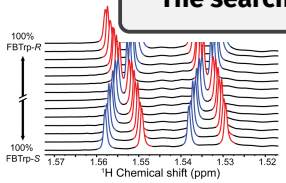
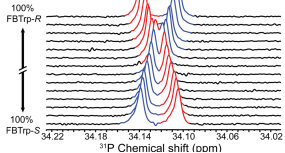
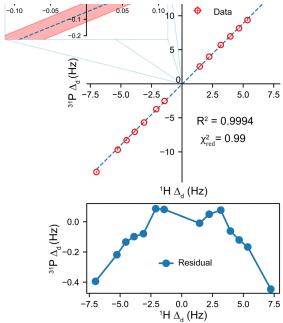
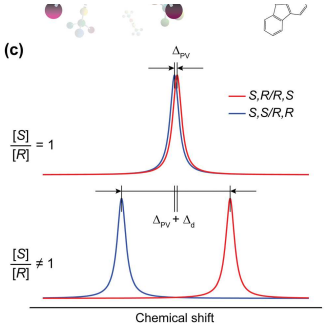


The search for the ideal molecule continues...





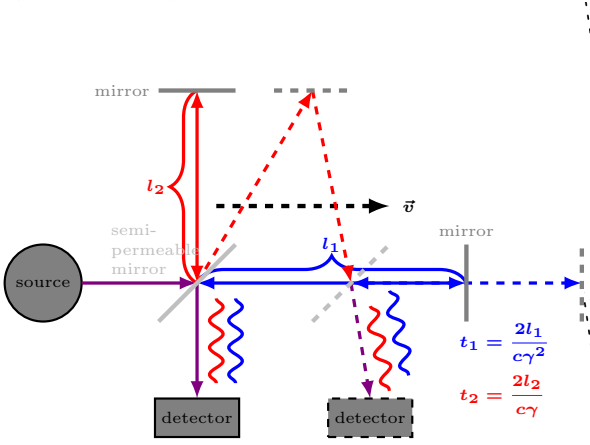


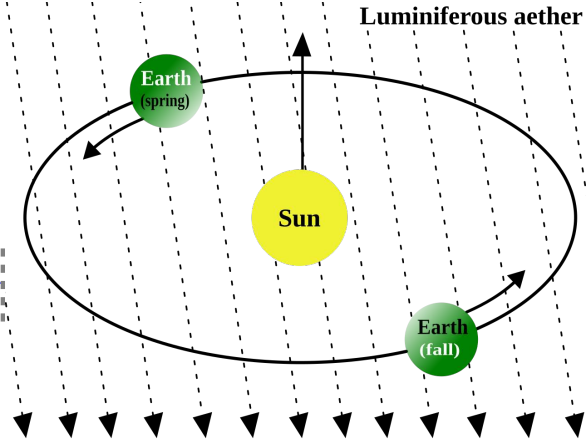


New physics in molecular parity violation?

Lorentz invariance violation

Special relativity was discovered when one tried to measure the luminoferous aether!

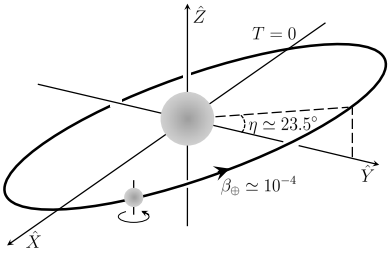




https://commons.wikimedia.org/wiki/File:AetherWind.svg

- Local Lorentz invariance violation (LLIV) would imply CPT violation
- String theory and various quantum gravity theories hint to LLIV
- Kostelecký & co: effective field theory that extends the SM with LLIV
- Electron sector SME:

$$\mathcal{L}_{\mathrm{e}}^{\mathrm{SME}} = \hbar c \bar{\psi}_{\mathrm{e}} \left[\tilde{c}_{\mu\nu}\gamma^{\mu} + \tilde{d}_{\mu\nu}\gamma_5\gamma^{\mu} + \tilde{e}_{\nu} + \tilde{f}_{\nu}\gamma_5 + \frac{1}{2}\tilde{g}_{\lambda\mu\nu}\sigma^{\lambda\mu} \right] \frac{\imath}{2} \overleftrightarrow{\partial}^{\nu} \psi_{\mathrm{e}} - m_{\mathrm{e}} c^2 \bar{\psi}_{\mathrm{e}} \left[\tilde{a}_{\mu}\gamma^{\mu} + \tilde{b}_{\mu}\gamma_5\gamma^{\mu} + \frac{1}{2}\tilde{h}_{\mu\nu}\sigma^{\mu\nu} \right] \psi_{\mathrm{e}} \,.$$



Coefficient	C	P	T	CP	CT	PT	CPT
$c_{TT},\ c_{JK},\ (k_F)_{TJTK},\ (k_F)_{JKLM}$	+	+	+	+	+	+	+
$b_J,\ g_{JTL},\ g_{JKT},\ (k_{AF})_J$	+	+	−	+	−	−	−
$b_T,\ g_{JTT},\ g_{JKL},\ (k_{AF})_T$	+	−	+	−	+	−	−
$c_{TJ},\ c_{JT},\ (k_F)_{TJKL}$	+	−	−	−	−	+	+
$a_T,\ e_T,\ f_J$	−	+	+	−	−	+	−
$H_{JK},\ d_{TJ},\ d_{JT}$	−	+	−	−	+	−	+
$H_{TJ},\ d_{TT},\ d_{JK}$	−	−	+	+	−	−	+
$a_J,\ e_J,\ f_T$	−	−	−	+	+	+	−

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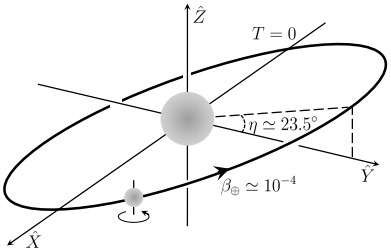

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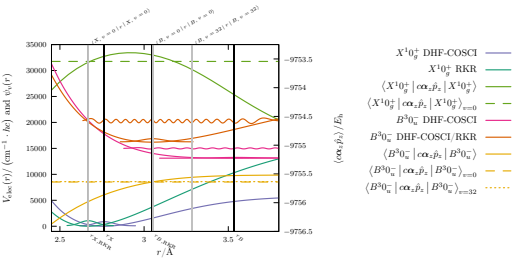
$c_{TJ},\ c_{JT},\ (k_F)_{TJKL}$	+	−	−	−	−	+	+
$a_T,\ e_T,\ f_J$	−	+	+	−	−	+	−
$H_{JK},\ d_{TJ},\ d_{JT}$	−	+	−	−	+	−	+
$H_{TJ},\ d_{TT},\ d_{JK}$	−	−	+	+	−	−	+
$a_J,\ e_J,\ f_T$	−	−	−	+	+	+	−

PHYSICAL REVIEW D **97,** 124051 (2018)

BOOST: A satellite mission to test Lorentz invariance using high-performance optical frequency references

Norman Gürlebeck,[1,2,*] Lisa Wörner,[1,2] Thilo Schuldt,[2] Klaus Döringshoff,[3] Konstantin Gaul,[4] Domenico Gerardi,[5] Arne Grenzebach,[1,2] Nandan Jha,[6] Evgeny Kovalchuk,[3] Andreas Resch,[2] Thijs Wendrich,[6] Robert Berger,[4] Sven Herrmann,[1] Ulrich Johann,[5] Markus Krutzik,[3] Achim Peters,[3] Ernst M. Rasel,[6] and Claus Braxmaier[1,2]

$$\Delta^{\mathrm{LLIV}}_{kk} = -\tilde{c}_{kk}\left(\left\langle B0^+_u, v' \middle| c\sum_i \alpha^k_i \hat{p}^k_i \middle| B0^+_u, v'\right\rangle - \left\langle X0^+_g, v \middle| c\sum_i \alpha^k_i \hat{p}^k_i \middle| X0^+_g, v\right\rangle\right)$$



$$\Rightarrow \frac{\Delta^{\mathrm{LLIV}}_{kk}}{\tilde{c}_{11}} \approx 27\,\mathrm{eV};\quad \frac{\Delta^{\mathrm{LLIV}}_{kk}}{\tilde{c}_{33}} \approx -54\,\mathrm{eV}$$

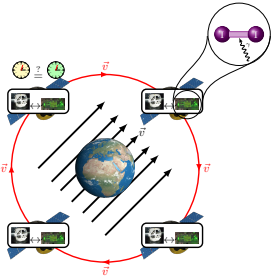


TABLE II. Expected constraints on LIV by the proposed mission BOOST after one year of observation.

Constraints[a]
$\lvert c^{\mathrm{SCF}}_{10} + c^{\mathrm{SCF}}_{01}\rvert \le 3\times10^{-13}$
$\lvert c^{\mathrm{SCF}}_{30} + c^{\mathrm{SCF}}_{03}\rvert \le 3\times10^{-13}$
$\lvert c^{\mathrm{SCF}}_{12} + c^{\mathrm{SCF}}_{21}\rvert \le 4\times10^{-17}$
$\lvert c^{\mathrm{SCF}}_{13} + c^{\mathrm{SCF}}_{31}\rvert \le 2\times10^{-17}$
$\lvert c^{\mathrm{SCF}}_{23} + c^{\mathrm{SCF}}_{32}\rvert \le 3\times10^{-17}$
$\lvert c^{\mathrm{SCF}}_{11} + c^{\mathrm{SCF}}_{22} - 2c^{\mathrm{SCF}}_{33}\rvert \le 4\times10^{-17}$
$\lvert\alpha_{\mathrm{KT}}\rvert \le 7.5\times10^{-10}$[b]

PHYSICAL REVIEW D **97,** 124051 (2018)

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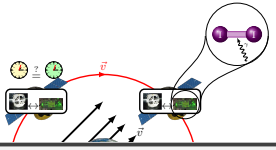
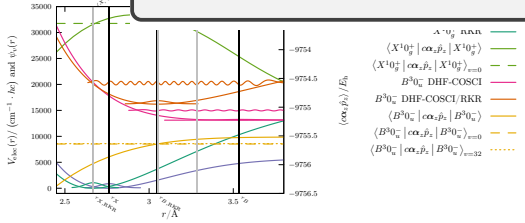


$\Delta_{kk}^{\mathrm{LLIV}} = -\tilde{c}_{kk}$

⇒ In the meantime surpassed by atomic clock experiments with Yb^+!
Potential of molecules yet to be explored…



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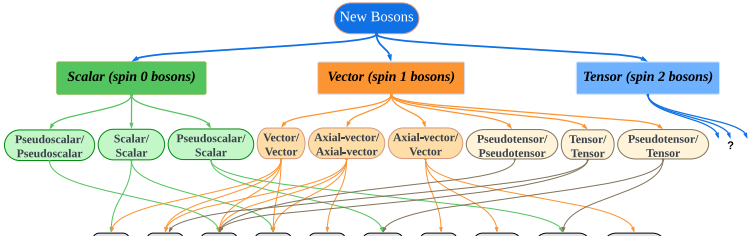
$\lvert c_{23}^{\mathrm{SCF}} + c_{32}^{\mathrm{SCF}}\rvert \le 3\times10^{-17}$
$\lvert c_{11}^{\mathrm{SCF}} + c_{22}^{\mathrm{SCF}} - 2c_{33}^{\mathrm{SCF}}\rvert \le 4\times10^{-17}$
$\lvert\alpha_{\mathrm{KT}}\rvert \le 7.5\times10^{-10}$[b]

New bosons and fifth force

A new boson would mediate a fifth force.

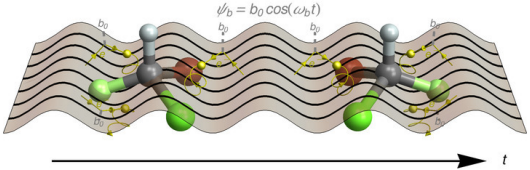


Symmetry | Interactions	ss	sp	pp	VV	AV	AA	TT	TP
P	+	−	+	+	−	+	+	−
T	+	−	+	+	+	+	+	−

- Atomic/molecular probes of ss/pp/VV/AA/TT: Precision experiments/theory with simple systems
- Atomic probes of ss with isotope shift spectroscopy (inferior to g-factor measurements)
- EDM experiments for probing sp
- Parity violation experiments for AV

L. Cong et al., *Rev. Mod. Phys.* **2025**, *97*, 025005.
V. A. Dzuba et al., *Phys. Rev. Lett.* **2017**, *119*, 223201.

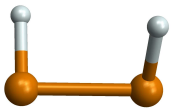
Molecular parity violation beyond the SM

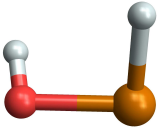

$\psi_b = b_0 \cos(\omega_b t)$
t
1000
800
600
400
200
−200
2
4
5
$\Delta_{pv} E_{ew}$
$(+\Delta_{pv} E_{DM})$
V_{em}
$V_{em} + V_{ew}$
$V_{em} + V_{ew} + V_{DM}$

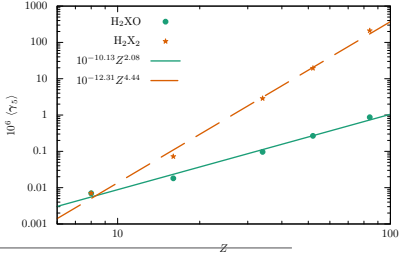
Scaling behavior with respect to Z and α:

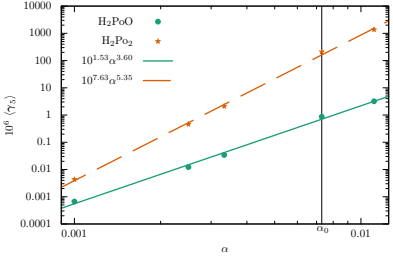
$$\delta E_{\gamma^5} \approx c_1 \alpha^5 Z_A^2 Z_B^2 + c_2 \alpha^3 Z_A^2 + c_3 \alpha^3 Z_B^2 \,.$$

H_2X_2 **H_2XO**







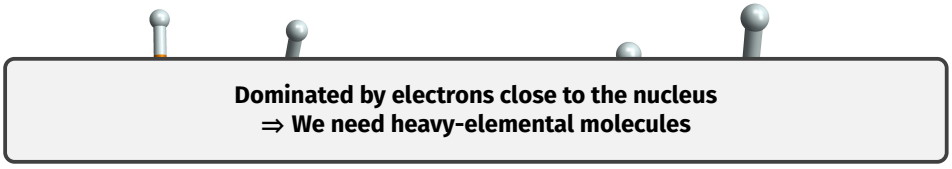


K. Gaul et al., *Phys. Rev. A* **2020**, *102*, 032816.

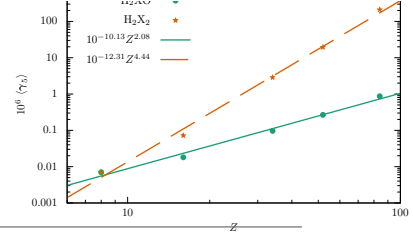
Scaling behavior with respect to Z and α:

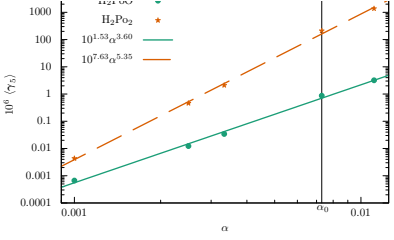
$$\delta E_{\gamma^5} \approx c_1 \alpha^5 Z_A^2 Z_B^2 + c_2 \alpha^3 Z_A^2 + c_3 \alpha^3 Z_B^2 \,.$$

H_2X_2 **H_2XO**



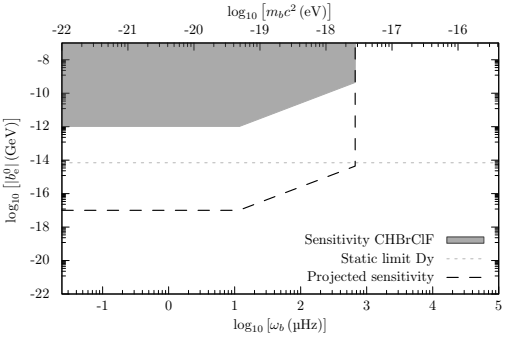
Dominated by electrons close to the nucleus
⇒ We need heavy-elemental molecules





K. Gaul et al., *Phys. Rev. A* **2020**, *102*, 032816.

$$b_0^{\mathrm{e}} \lesssim \begin{cases} 10^{-12}\,\mathrm{GeV}, & \text{if } \frac{\omega_b}{2\pi} \leq 1.2\,\mu\mathrm{Hz} \\ (\omega_b t_{\mathrm{tot}})^{3/2} 10^{-12}\,\mathrm{GeV}, & \text{if } 1.2\,\mu\mathrm{Hz} < \frac{\omega_b}{2\pi} \leq 0.7\,\mathrm{mHz} \\ \infty, & \text{if } \frac{\omega_b}{2\pi} > 0.7\,\mathrm{mHz} \end{cases} .$$



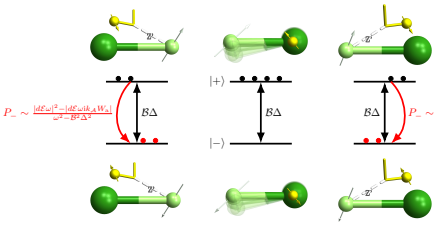
Possible improvements:

1. at least 2 orders by experiment
2. 2 orders by molecule $\sim \alpha^5 Z^4$ (e.g. CHAtFI)
3. 1 order by choice of vibrational mode

K. Gaul et al., *Phys. Rev. Lett.* **2020**, *125*, 123004.

Molecular parity violation beyond the SM

Parity violating mixing of Ω states in BaF



PHYSICAL REVIEW LETTERS **120,** 142501 (2018)

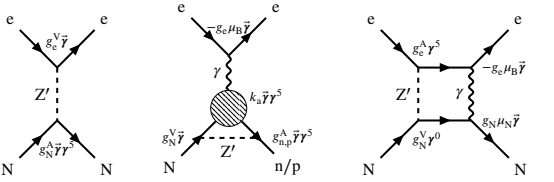
Editors' Suggestion

Demonstration of a Sensitive Method to Measure Nuclear-Spin-Dependent Parity Violation

Emine Altuntaş,* Jeffrey Ammon,† Sidney B. Cahn,‡ and David DeMille§
Department of Physics, Yale University, P.O. Box 208120, New Haven, Connecticut 06520, USA

(Received 22 December 2017; revised manuscript received 16 February 2018; published 2 April 2018)

Nuclear-spin-dependent parity violation (NSD-PV) effects in atoms and molecules arise from Z^0 boson exchange between electrons and the nucleus, and from the magnetic interaction between electrons and the parity-violating nuclear anapole moment. We demonstrate enhancement of the effect in diatomic molecules, here using $^{138}Ba^{19}F$. D-PV that use an $^2\Sigma$ F. Our sensitivity surpasses that of any previous atomic parity violation measurem natic errors can be suppressed to at least the level of the present statistical sensitivity. We measure the matrix element W of the NSD-PV interaction w configurations where W must have different si)-PV effects of the size anticipated across a wi $\delta W/(2\pi) < 0.7$ Hz, $2\pi) \approx 5$ Hz is expected.



$$\hat{H}_{\mathrm{PV,sr}} = k_{\mathcal{A}} W_a \vec{\lambda} \times \vec{S}' \cdot \vec{I}\,,$$

$$\Delta k_{\mathcal{A},K} W_{a,K} = g_{N,K}^A g_e^V W_{\text{AV-Ne},K} + g_{N,K}^V g_e^A W_{\text{AV-eN},K} + g_{n,p}^A g_N^V k_{a,\text{AV-(n,p)N},K} \frac{M^2 c\, e}{\epsilon_0 \hbar^3 A_K} \lim_{M\to\infty} W_{\text{AV-Ne},K}$$

$$g_{N,K}^A \approx \langle \boldsymbol{\sigma} \rangle_{p,K} g_p^A + \langle \boldsymbol{\sigma} \rangle_{n,K} g_n^A\,,\ g_{N,K}^V \approx \frac{Z_K g_p^V + N_K g_n^V}{A_K}$$

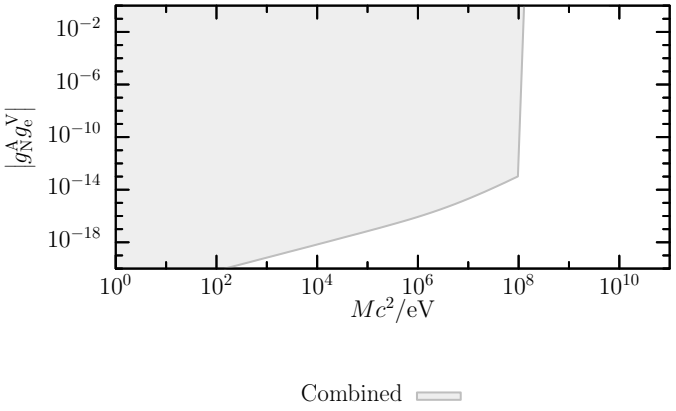
- Computation of W with density functional theory → constraints from $Ba^{19}F$ experiment
- Projected sensitivity of experiments with ^{137}BaF, RaF and SiO^+

K. Gaul et al., *arXiv* **2025**, *hep-ph*, 2503.08210.

T. A. Isaev, R. Berger, *Phys. Rev. A* **2012**, *86*, 062515, S. G. Wilkins et al., *arXiv* **2024**, *physics.atom-ph*, 2408.14673, J. Karthein et al., *Phys. Rev. Lett.* **2024**, *133*, 033003.

Molecular parity violation beyond the SM

Constraints on exotic axial vector-vector nucleus-electron (AV-Ne) interaction



Lines are not constraints but projected electronic sensitivities!

K. Gaul et al., *arXiv* **2025**, *hep-ph*, 2503.08210.

Molecular parity violation beyond the SM

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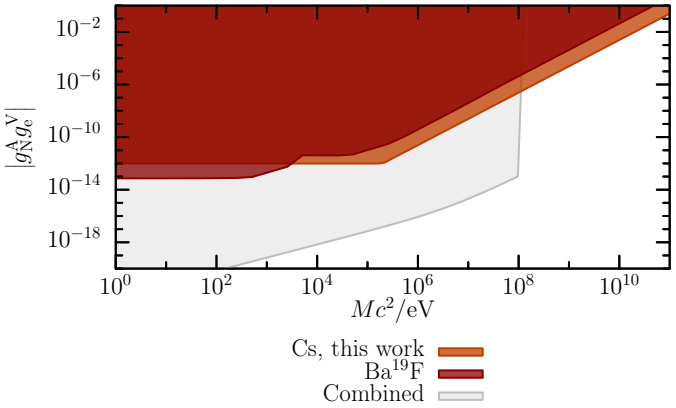

- **First constraints** on AV-Ne interactions
- Constraints from Cs experiment by relating anapole moment to AV-Ne interactions $\frac{9.62\times10^{21} g_e^V g_N^A}{M^2c^4/\mathrm{eV}^2}$
- $Ba^{19}F$ more sensitive than Cs for small boson masses!

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K. Gaul et al., *arXiv* **2025**, *hep-ph*, 2503.08210.

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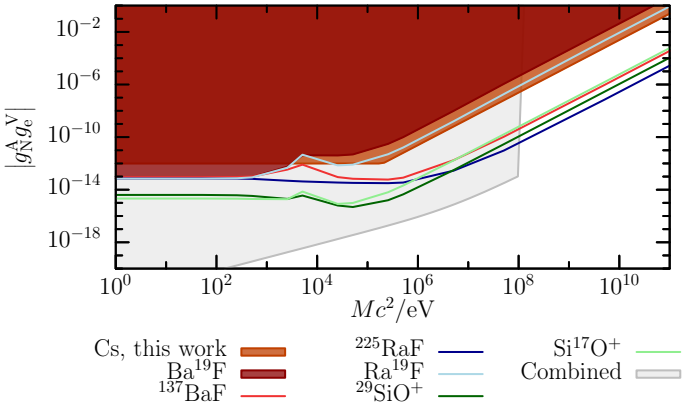

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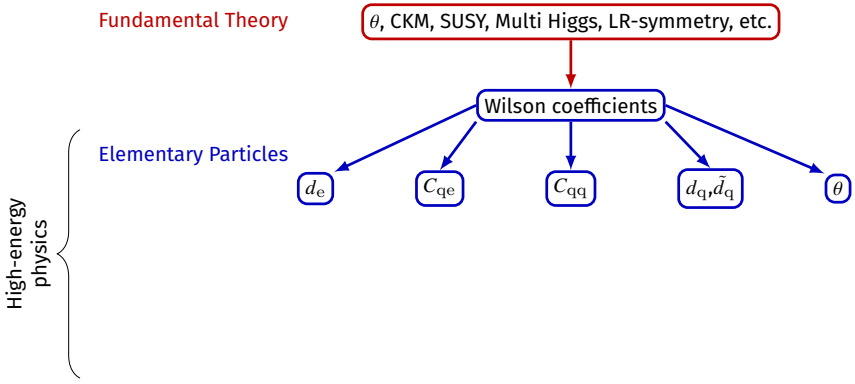
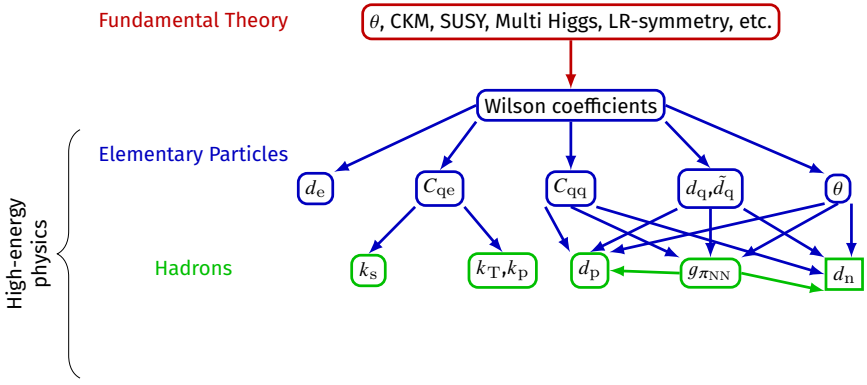
- $Ba^{19}F$ more sensitive than Cs for small boson masses!
- Improvement with ^{137}BaF or ^{225}RaF experiments **1 to 4 orders** of magnitude
- Uncertainty of nuclear theory needs to be improved or requires measurement of different isotopes

K. Gaul et al., *arXiv* **2025**, *hep-ph*, 2503.08210.

Electric Dipole Moments (EDMs) in atoms and molecules



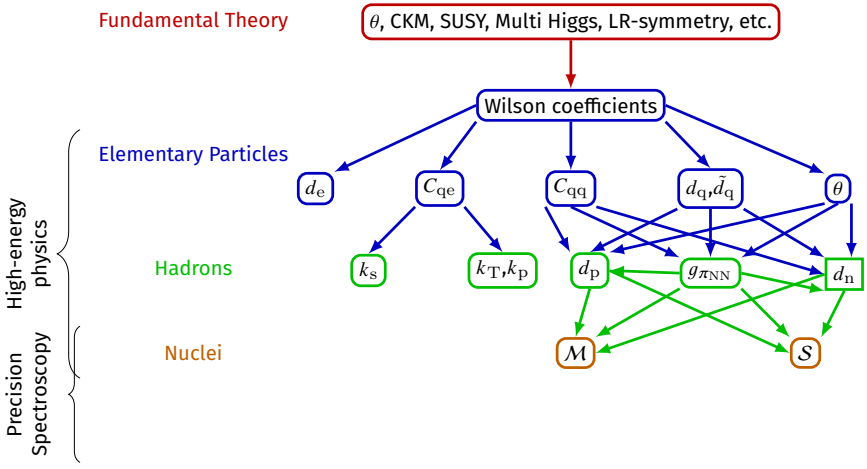
M. Pospelov, A. Ritz, *Ann. Phys. (N. Y.)* **2005**, *318*, 119 –169, T. E. Chupp et al., *Rev. Mod. Phys.* **2019**, *91*, 015001, W. B. Cairncross, J. Ye, *Nat. Rev. Phys.* **2019**, *1*, 510–521, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.



M. Pospelov, A. Ritz, *Ann. Phys. (N. Y.)* **2005**, *318*, 119 –169, T. E. Chupp et al., *Rev. Mod. Phys.* **2019**, *91*, 015001, W. B. Cairncross, J. Ye, *Nat. Rev. Phys.* **2019**, *1*, 510–521, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.

Electric Dipole Moments (EDMs) in atoms and molecules

From elementary particles to EDMs of atoms and molecules



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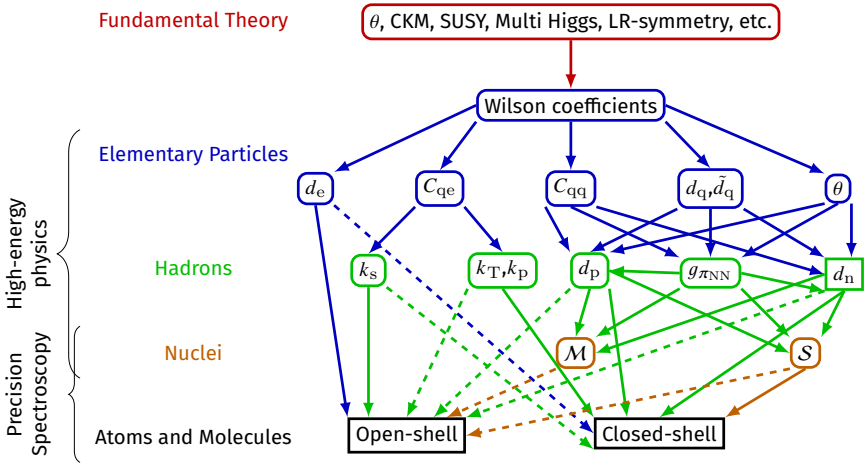
Electric Dipole Moments (EDMs) in atoms and molecules

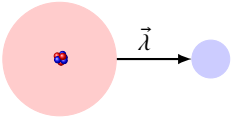
From elementary particles to EDMs of atoms and molecules



M. Pospelov, A. Ritz, *Ann. Phys. (N. Y.)* **2005**, *318*, 119 –169, T. E. Chupp et al., *Rev. Mod. Phys.* **2019**, *91*, 015001, W. B. Cairncross, J. Ye, *Nat. Rev. Phys.* **2019**, *1*, 510–521, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.

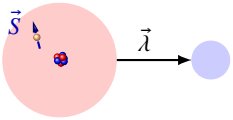
Electric Dipole Moments (EDMs) in atoms and molecules

Effective $\mathcal{P}, \mathcal{T}$-odd Hamiltonian (linear molecule with one heavy nucleus)



$$H_{\mathcal{P},\mathcal{T}} =$$

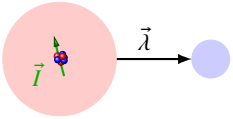
K. Gaul, R. Berger, *J. Chem. Phys.* **2020**, *152*, 044101, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, M. G. Kozlov, L. N. Labzowsky, *J. Phys. B* **1995**, *28*, 1933–1961.



I) $\Omega > 0$ with $I = 0$

$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda} \cdot \vec{S}'}_{\Omega} \left(W_{\mathrm{d}} d_{\mathrm{e}} + W_{\mathrm{s}} k_{\mathrm{s}}\right)$$

K. Gaul, R. Berger, *J. Chem. Phys.* **2020**, *152*, 044101, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, M. G. Kozlov, L. N. Labzowsky, *J. Phys. B* **1995**, *28*, 1933–1961.



I) $\Omega > 0$ with $I = 0$

II) $\Omega = 0$ with $I \geq 1/2$

$$H_{\mathcal{P},\mathcal{T}} =$$

$$+ \underbrace{\vec{\lambda} \cdot \vec{I}}_{\mathcal{I}} \left(W_{\mathrm{T}} k_{\mathrm{T}} + W_{\mathrm{p}} k_{\mathrm{p}} + W_{\mathrm{s}}^{\mathrm{m}} k_{\mathrm{s}} + W_{\mathcal{S}} \tilde{\mathcal{S}} g_{\pi \mathrm{NN}} + W_{\mathrm{d}}^{\mathrm{m}} d_{\mathrm{e}} + W_{\mathrm{m}} d_{\mathrm{N}} + W_{\mathcal{S}} R_{\mathrm{vol}} d_{\mathrm{N}} \right)$$

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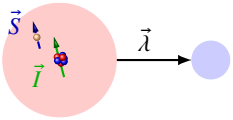
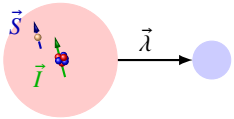


I) $\Omega > 0$ with $I = 0$
II) $\Omega = 0$ with $I \geq 1/2$
III) $\Omega > 0$ with $I = 1/2$

$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda} \cdot \vec{S}'}_{\Omega} \left(W_\mathrm{d} d_\mathrm{e} + W_\mathrm{s} k_\mathrm{s}\right) + \underbrace{\vec{\lambda} \cdot \vec{I}}_{I} \left(W_\mathrm{T} k_\mathrm{T} + W_\mathrm{p} k_\mathrm{p} + W_\mathrm{s}^\mathrm{m} k_\mathrm{s} + W_\mathcal{S} \tilde{\mathcal{S}} g_{\pi\mathrm{NN}} + W_\mathrm{d}^\mathrm{m} d_\mathrm{e} + W_\mathrm{m} d_\mathrm{N} + W_\mathcal{S} R_\mathrm{vol} d_\mathrm{N}\right)$$

K. Gaul, R. Berger, *J. Chem. Phys.* **2020**, *152*, 044101, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, M. G. Kozlov, L. N. Labzowsky, *J. Phys. B* **1995**, *28*, 1933–1961.



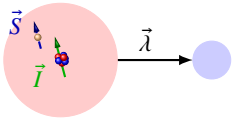
I) $\Omega > 0$ with $I = 0$
II) $\Omega = 0$ with $I \geq 1/2$
III) $\Omega > 0$ with $I = 1/2$
IV) $\Omega > 0$ with $I > 1/2$

$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda} \cdot \vec{S}'}_{\Omega} (W_\mathrm{d} d_\mathrm{e} + W_\mathrm{s} k_\mathrm{s}) + \underbrace{\vec{\lambda}^\mathsf{T} \cdot \mathbf{T} \cdot \vec{S}'}_{\Theta} W_\mathcal{M} (\tilde{\mathcal{M}}_\pi g_{\pi\mathrm{NN}} + \tilde{\mathcal{M}}_\mathrm{EDM} d_\mathrm{N})$$
$$+ \underbrace{\vec{\lambda} \cdot \vec{I}}_{I} \left(W_\mathrm{T} k_\mathrm{T} + W_\mathrm{p} k_\mathrm{p} + W_\mathrm{s}^\mathrm{m} k_\mathrm{s} + W_\mathcal{S} \tilde{\mathcal{S}} g_{\pi\mathrm{NN}} + W_\mathrm{d}^\mathrm{m} d_\mathrm{e} + W_\mathrm{m} d_\mathrm{N} + W_\mathcal{S} R_\mathrm{vol} d_\mathrm{N} \right)$$

We need from ab initio calculations:

- Electronic structure factors (steeply scale with Z) $W_\mathrm{d}, W_\mathrm{s}, W_\mathcal{M}, W_\mathrm{T}, W_\mathrm{p}, W_\mathrm{s}^\mathrm{m}, W_\mathcal{S}, W_\mathrm{d}^\mathrm{m}, W_\mathrm{m}$
- Nuclear structure factors $R_\mathrm{vol}, \tilde{\mathcal{S}}, \tilde{\mathcal{M}}_\mathrm{EDM}, \tilde{\mathcal{M}}_\pi$

K. Gaul, R. Berger, *J. Chem. Phys.* **2020**, *152*, 044101, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, M. G. Kozlov, L. N. Labzowsky, *J. Phys. B* **1995**, *28*, 1933–1961.



I) $\Omega > 0$ with $I = 0$
II) $\Omega = 0$ with $I \geq 1/2$
III) $\Omega > 0$ with $I = 1/2$
IV) $\Omega > 0$ with $I > 1/2$

$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda} \cdot \vec{S}'}_{\Omega} (W_\mathrm{d} d_\mathrm{e} + W_\mathrm{s} k_\mathrm{s}) + \underbrace{\vec{\lambda}^\mathsf{T} \cdot \mathbf{T} \cdot \vec{S}'}_{\Theta} W_\mathcal{M} (\tilde{\mathcal{M}}_\pi g_{\pi\mathrm{NN}} + \tilde{\mathcal{M}}_\mathrm{EDM} d_\mathrm{N})$$
$$+ \underbrace{\vec{\lambda} \cdot \vec{I}}_{I} \left(W_\mathrm{T} k_\mathrm{T} + W_\mathrm{p} k_\mathrm{p} + W_\mathrm{s}^\mathrm{m} k_\mathrm{s} + W_\mathcal{S} \tilde{\mathcal{S}} g_{\pi\mathrm{NN}} + W_\mathrm{d}^\mathrm{m} d_\mathrm{e} + W_\mathrm{m} d_\mathrm{N} + W_\mathcal{S} R_\mathrm{vol} d_\mathrm{N} \right)$$

We need from ab initio calculations:

- **Electronic structure factors (steeply scale with Z)** $W_\mathrm{d}, W_\mathrm{s}, W_\mathcal{M}, W_\mathrm{T}, W_\mathrm{p}, W_\mathrm{s}^\mathrm{m}, W_\mathcal{S}, W_\mathrm{d}^\mathrm{m}, W_\mathrm{m}$
- Nuclear structure factors $R_\mathrm{vol}, \tilde{\mathcal{S}}, \tilde{\mathcal{M}}_\mathrm{EDM}, \tilde{\mathcal{M}}_\pi$

$\Rightarrow$ **Large enhancement factors $\Leftrightarrow$ high sensitivity(?)**

K. Gaul, R. Berger, *J. Chem. Phys.* **2020**, *152*, 044101, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, M. G. Kozlov, L. N. Labzowsky, *J. Phys. B* **1995**, *28*, 1933–1961.

Electric Dipole Moments (EDMs) in atoms and molecules

Electronic structure enhancement factors

$W_\mathrm{d} =$	$\dfrac{\left\langle \frac{2c}{\hbar} \sum_{i=1}^{N_\mathrm{elec}} \imath \boldsymbol{\gamma}_i^0 \boldsymbol{\gamma}_i^5 \hat{\vec{\rho}}_i^2 \right\rangle}{\Omega}$	A. Mårtensson-Pendrill, P Öster, *Phys. Scr.* **1987**, *36*, 444–452
$W_\mathrm{d}^\mathrm{m} =$	$-\vec{I} \cdot \left\langle \frac{2c}{\hbar} \sum_{i=1}^{N_\mathrm{elec}} \frac{\boldsymbol{\gamma}_i^0 \boldsymbol{\gamma}_i^5}{r_{iA}^3} \hat{\vec{\ell}}_{iA} \right\rangle + \vec{I} \cdot 2\mathrm{Re} \left\{ \sum_a \frac{\left\langle 0 \left\vert \frac{2c}{\hbar} \sum_{i=1}^{N_\mathrm{elec}} \imath \boldsymbol{\gamma}_i^0 \boldsymbol{\gamma}_i^5 \hat{\vec{\rho}}_i^2 \right\vert a \right\rangle \left\langle a \left\vert \frac{\mu_0}{4\pi} \sum_{i=1}^{N_\mathrm{elec}} \frac{\vec{r}_{iA} \times \vec{\alpha}_i}{r_{iA}^3} \right\vert 0 \right\rangle}{E_0 - E_a} \right\}$	A. Mårtensson-Pendrill, P Öster, *Phys. Scr.* **1987**, *36*, 444–452
$W_\mathcal{S} =$	$\vec{I} \cdot \left\langle \frac{e}{\epsilon_0} \sum_{i=1}^{N_\mathrm{elec}} (\vec{\nabla}_i \rho_A(\vec{r}_i)) \right\rangle$	E. A. Hinds, P. G. H. Sandars, *Phys. Rev. A* **1980**, *21*, 471–479, V. V. Flambaum, J. S. M. Ginges, *Phys. Rev. A* **2002**, *65*, 032113
$W_\mathrm{m} =$	$\vec{I} \cdot \left\langle 4 \frac{c\mu_0}{4\pi\hbar} \sum_{i=1}^{N_\mathrm{elec}} \frac{\vec{\alpha}_i}{r_{iA}^3} \times \hat{\vec{\ell}}_{iA} \right\rangle$	E. A. Hinds, P. G. H. Sandars, *Phys. Rev. A* **1980**, *21*, 471–479
$W_\mathcal{M} =$	$\dfrac{\vec{I}^\mathrm{T}}{I} \cdot \dfrac{\left\langle \frac{ce\mu_0}{4\pi} \frac{3}{2} \sum_{i=1}^{N_\mathrm{elec}} \frac{\vec{r}_{iA} \otimes (\vec{\alpha} \times \vec{r}_{iA})}{r_{iA}^5} \right\rangle}{\Omega} \cdot \dfrac{\vec{I}}{I}$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125
$W_\mathrm{s} =$	$\dfrac{\left\langle -\frac{G_\mathrm{F}}{\sqrt{2}} \sum_{i=1}^{N_\mathrm{elec}} \imath \boldsymbol{\gamma}_i^0 \boldsymbol{\gamma}_i^5 Z_A \rho_A(\vec{r}_i) \right\rangle}{\Omega}$	O. P. Sushkov et al., *Sov. Phys. JETP* **1984**, *60*, 873–883
$W_\mathrm{s}^\mathrm{m} =$	$\vec{I} \cdot 2\mathrm{Re} \left\{ \sum_a \frac{\left\langle 0 \left\vert -\frac{G_\mathrm{F}}{\sqrt{2}} \sum_{i=1}^{N_\mathrm{elec}} \imath \boldsymbol{\gamma}_i^0 \boldsymbol{\gamma}_i^5 Z_A \rho_A(\vec{r}_i) \right\vert a \right\rangle \left\langle a \left\vert \frac{\mu_0}{4\pi} \sum_{i=1}^{N_\mathrm{elec}} \frac{\vec{r}_{iA} \times \vec{\alpha}_i}{r_{iA}^3} \right\vert 0 \right\rangle}{E_0 - E_a} \right\}$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125
$W_\mathrm{T} =$	$\vec{I} \cdot \left\langle \sqrt{2} G_\mathrm{F} \sum_{i=1}^{N_\mathrm{elec}} \imath \vec{\boldsymbol{\gamma}} \rho_A(\vec{r}_i) \right\rangle$	E. A. Hinds, P. G. H. Sandars, *Phys. Rev. A* **1980**, *21*, 471–479
$W_\mathrm{p} =$	$\vec{I} \cdot \left\langle \frac{G_\mathrm{F}}{\sqrt{2}} \sum_{i=1}^{N_\mathrm{elec}} \boldsymbol{\beta}_i (\vec{\nabla}_i \rho_A(\vec{r}_i)) \right\rangle$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125

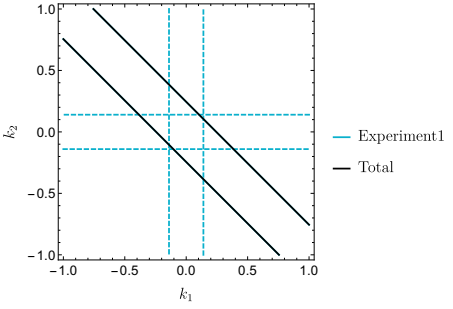
K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100.

W_i	αZ-scaling	Relativistic enhancement	Reference
W_d	$\alpha^2 Z^3$	$\frac{3}{\gamma_{1/2}(4\gamma_{1/2}^2-1)}$	P. Sandars, *Phys. Lett.* **1965**, *14*, 194–196, V. V Flambaum, *Yad. Fiz.* **1976**, *24*, 383–386
W_d^m	αZ^2	$R(Z,A)-1$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125
$W_\mathcal{S}$	Z^2	$R(Z,A)\frac{3\gamma_{1/2}}{2\gamma_{1/2}+1}$	O. P. Sushkov et al., *Sov. Phys. JETP* **1984**, *60*, 873–883
W_m	αZ	$\frac{15\left(1+\frac{7}{16}(1-\gamma_{1/2}^2)+\frac{93-4\pi^2}{384}(1-\gamma_{1/2}^2)^2\right)}{16\gamma_{1/2}^2-1}$	K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.
$W_\mathcal{M}$	αZ^2	$\frac{720\Gamma\left(\gamma_{1/2}+\gamma_{3/2}-2\right)}{\Gamma\left(3+\gamma_{1/2}-\gamma_{3/2}\right)\Gamma\left(3-\gamma_{1/2}+\gamma_{3/2}\right)\Gamma\left(3+\gamma_{1/2}+\gamma_{3/2}\right)}$	O. P. Sushkov et al., *Sov. Phys. JETP* **1984**, *60*, 873–883
W_s	αZ^3	$R(Z,A)\frac{\gamma_{1/2}+1}{2}f_0(Z)$	O. P. Sushkov, V. V. Flambaum, *Sov. Phys. JETP* **1978**, *48*, 608–611, V. A. Dzuba et al., *Phys. Rev. A* **2011**, *84*, 052108
W_s^m	$\alpha^2 Z^3$	$R(Z,A)$	I. B. Khriplovich, S. K. Lamoreaux, CP Violation without Strangeness, Springer, Berlin, **1997**
W_T	αZ^2	$R(Z,A)\frac{(2+\gamma_{1/2})}{3}$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125
W_p	$\alpha^2 Z^3$	$R(Z,A)$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125

K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.

- At least 6 experiments $\hbar\vec{\omega}$ needed to determine all effective parameters on molecular level:

$$\hbar\vec{\omega} = \boldsymbol{W}\vec{x}_{\mathcal{P},\mathcal{T}} \quad \vec{x}_{\mathcal{P},\mathcal{T}} = (d_\mathrm{e}, d_\mathrm{N}, k_\mathrm{s}, k_\mathrm{T}, k_\mathrm{p}, g_{\pi\mathrm{NN}})$$



Gaussian probability distributions
→ Ellipsoidal coverage regions:

$$\vec{x}^{\mathsf{T}}_{\mathcal{P},\mathcal{T}}\boldsymbol{U}^{-1}_{\mathcal{P},\mathcal{T}}\vec{x}_{\mathcal{P},\mathcal{T}} = P^2\,,$$

$$\boldsymbol{U}^{-1}_{\mathcal{P},\mathcal{T}} = \boldsymbol{W}^{\mathsf{T}}\boldsymbol{U}^{-1}_{\omega}\boldsymbol{W}$$

Volume V of coverage region: $V = P\frac{2\pi^{N/2}}{N\Gamma(N/2)}\det\left(\boldsymbol{U}^{-1}_{\mathcal{P},\mathcal{T}}\right)^{-1/2}$.

For $N = 6$ and 95 % CL ($P = 3.55$) and six independent measurements: $V = 3.55\frac{\pi^3}{6}\frac{\prod_{i=1}^{6}|\hbar\sigma_{\omega,i}|}{|\det(\boldsymbol{W})|}$.

Determinant is maximal if all measurements have orthogonal sensitivities!

K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **2015**, *91*, 035502, JCGM 102:2011, Evaluation of measurement data – Supplement 2 to the "Guide to the expression of uncertainty in measurement" – Extension to any number of output quantities, Standard, Joint Committee for Guides in Metrology, Paris, FR, **2011**.

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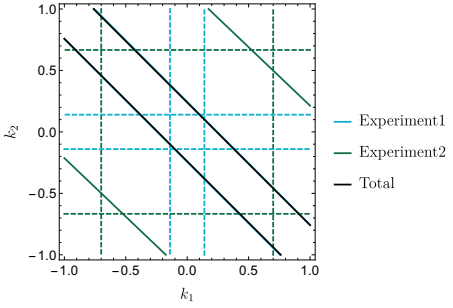

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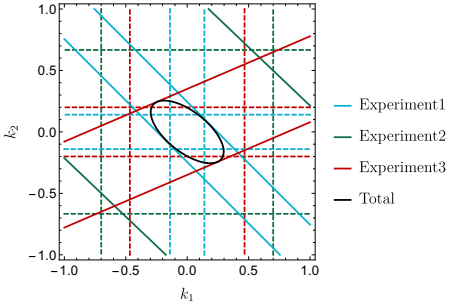

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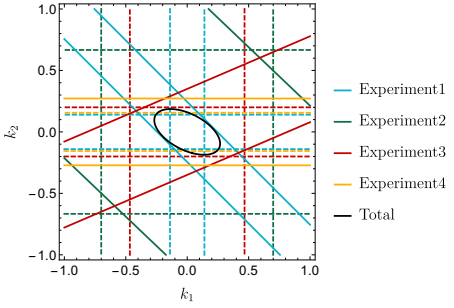

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$$\boldsymbol{U}^{-1}_{\mathcal{P},\mathcal{T}} = \boldsymbol{W}^{\mathsf{T}}\boldsymbol{U}^{-1}_{\omega}\boldsymbol{W}$$

Volume V of coverage region: $V = P\frac{2\pi^{N/2}}{N\Gamma(N/2)}\det\left(\boldsymbol{U}^{-1}_{\mathcal{P},\mathcal{T}}\right)^{-1/2}$.

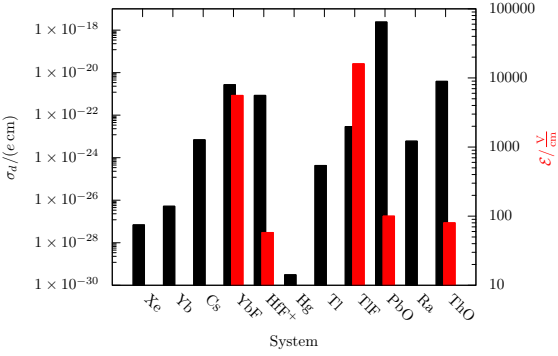
For $N = 6$ and 95 % CL ($P = 3.55$) and six independent measurements: $V = 3.55\frac{\pi^3}{6}\frac{\prod_{i=1}^{6}|\hbar\sigma_{\omega,i}|}{|\det(\boldsymbol{W})|}$.

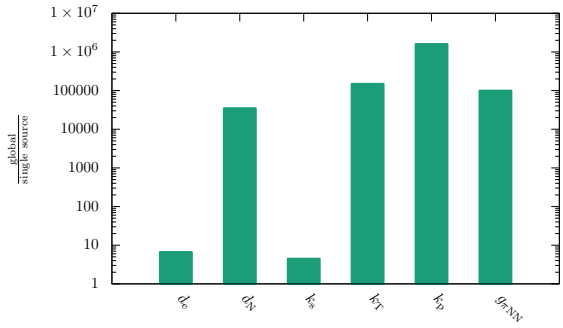
Determinant is maximal if all measurements have orthogonal sensitivities!

K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100, T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **2015**, 91, 035502, JCGM 102:2011, Evaluation of measurement data – Supplement 2 to the "Guide to the expression of uncertainty in measurement" – Extension to any number of output quantities, Standard, Joint Committee for Guides in Metrology, Paris, FR, **2011**.

Electric Dipole Moments (EDMs) in atoms and molecules

Single source model vs. global limits





Interpretation of EDM experiments with ab initio electronic structure calculations and rough nuclear structure estimates

K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100.

- Only 6 experiments with fixed uncertainties $\sigma_{\tilde{\nu},0} = 1\,\mathrm{arb.u.}$: $V \sim |\det(\boldsymbol{W})|^{-1}$
- Focus on **electronic structure**
- Effective single valence electron atom: $W_i \approx f_i(Z)$ for $20 \geq Z \leq 90$. For example:

$$W_\mathrm{d}(Z) = \frac{4}{3}\frac{\alpha}{a_0^2}\frac{\alpha^2 Z^3}{(\nu_{\mathrm{p}_{1/2}}\nu_{\mathrm{s}_{1/2}})^{3/2}}\frac{3}{\gamma_{1/2}(4\gamma_{1/2}^2-1)}\,.$$

$$W_\mathcal{S}(Z) = \frac{1}{a_0^4}\frac{Z^2}{(\nu_{\mathrm{p}_{1/2}}\nu_{\mathrm{s}_{1/2}})^{3/2}}R(Z,A)\frac{3\gamma_{1/2}}{2\gamma_{1/2}+1}\,,$$

$$\gamma_{1/2} = \sqrt{1-\alpha^2 Z^2}$$

$$R(Z,A) = \frac{4}{\Gamma^2(2\gamma_{1/2}+1)}(2Zr_\mathrm{nuc}/a_0)^{2\gamma_{1/2}-2}$$

- Crude estimates for nuclear structure:

$$\mathcal{S}_\mathrm{EDM} \approx \frac{3}{50}A^{2/3}(1.2\,\mathrm{fm})^2\frac{3+2I}{3+3I}(d_\mathrm{p}+d_\mathrm{n})$$

$$\mathcal{S}_\pi \approx \left(0.02\frac{18e}{20\pi\sqrt{35}}\frac{0.14Z}{A^{1/3}}(1.2\,\mathrm{fm}A^{1/3})^3 + \frac{3eq_\mathrm{ext}}{50}0.14\,\mathrm{fm}(1.2\,\mathrm{fm}A^{1/3})^2\right)\frac{3+2I}{3+3I}g_{\pi\mathrm{NN}}$$

$$\mathcal{M}_\mathrm{EDM} \approx \frac{-3}{2m_\mathrm{p}}\frac{1}{I+1}(d_\mathrm{p}+d_\mathrm{n})$$

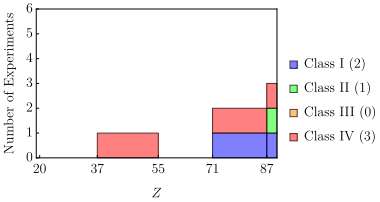
$$\mathcal{M}_\pi \approx \frac{3e(\mu-q_\mathrm{ext})}{2m_\mathrm{p}}1.4\times10^{-14}\,\mathrm{cm}\frac{1}{I+1}g_{\pi\mathrm{NN}},$$

$$\Rightarrow \min_{\{\Omega_i,\mathcal{I}_i\}\in\left\{\frac{n}{2}\,|\,n\in\mathbb{N}_0\right\},\,\{Z_i\}\in\mathbb{N}_0} V(\{\Omega_i,\mathcal{I}_i,Z_i\})$$

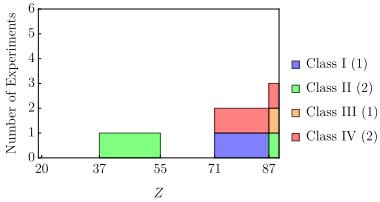
- $\binom{4+6-1}{6} - \sum_{k=1}^{4}\binom{k+2-1}{2} = 64$ minimizations with respect to $\{Z_i\}$ (for all combinations of different classes).

I. B. Khriplovich, S. K. Lamoreaux, CP Violation without Strangeness, Springer, Berlin, **1997**, J. S. M. Ginges, V. V. Flambaum, *Phys. Rep.* **2004**, *397*, 63–154, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100.

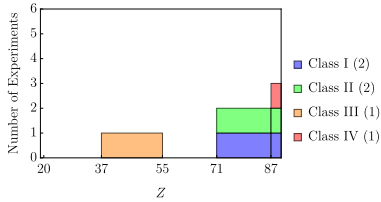
- ✓ Choosing complementary electronic structures
- ✓ Maximizing sensitivity to electronic and hadronic sector simultaneously
- ✓ Combinable with sophisticated nuclear structure models
- ✓ Expandable to contain theoretical uncertainties
- ✓ Impact of models for $\mathcal{CP}$-violation on electronic structure of various systems
- ✗ Complementarity in nuclear structure
- ✗ Advantages of odd or even proton numbers
- ✗ Directly extract sensitivity on the fundamental particle level



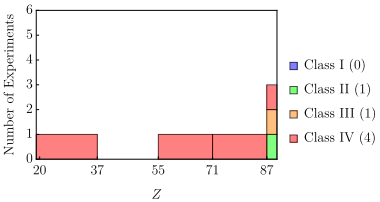
$V = 1.0 \times 10^{-3}$ arb.u. (1.)



$V = 1.7 \times 10^{-3}$ arb.u. (7.)



$V = 3.8 \times 10^{-3}$ arb.u. (14.)



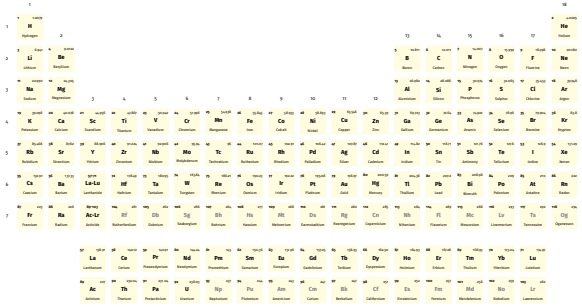
$V = 7.5 \times 10^{-3}$ arb.u. (28.)

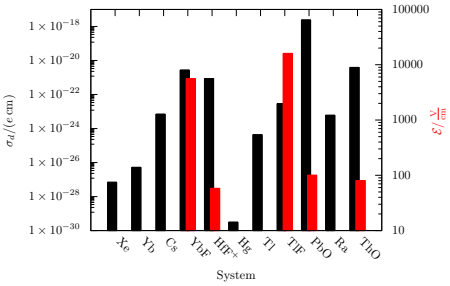
$\Rightarrow$ 3 or more of same class $\rightarrow$ system with $Z \leq 54$

$\Rightarrow$ Experiments of same class should have considerably different Z.

Electric Dipole Moments (EDMs) in atoms and molecules

Global ab initio study of past, present and future experiments





- “Modern” experiments with: Xe, Yb, Hg, TlF, Ra, Cs, HfF^+, Tl, PbO and ThO
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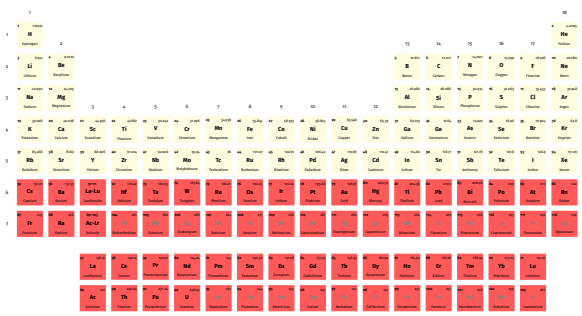
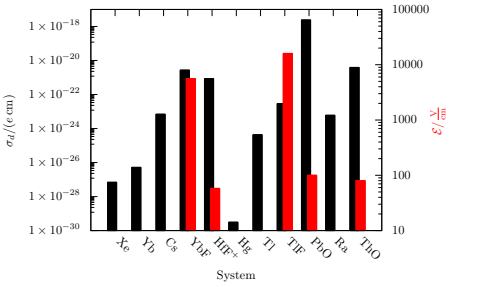
F. Allmendinger et al., *Phys. Rev. A* **2019**, *100*, 022505, N. Sachdeva et al., *Phys. Rev. Lett.* **2019**, *123*, 143003, S. A. Murthy et al., *Phys. Rev. Lett.* **1989**, *63*, 965–968, T. A. Zheng et al., *Phys. Rev. Lett.* **2022**, *129*, 083001, J. J. Hudson et al., *Phys. Rev. Lett.* **2002**, *89*, 23003, J. J. Hudson et al., *Nature* **2011**, *473*, 493, T. S. Roussy et al., *Science* **2023**, *381*, 46–50, B. Graner et al., *Phys. Rev. Lett.* **2016**, *116*, 161601, B. C. Regan et al., *Phys. Rev. Lett.* **2002**, *88*, 71805, D. Cho et al., *Phys. Rev. A* **1991**, *44*, 2783–2799, S. Eckel et al., *Phys. Rev. A* **2013**, *87*, 052130, R. H. Parker et al., *Phys. Rev. Lett.* **2015**, *114*, 233002, M. Bishof et al., *Phys. Rev. C* **2016**, *94*, 025501, V. Andreev et al., *Nature* **2018**, *562*, 355–360.

Electric Dipole Moments (EDMs) in atoms and molecules

Global ab initio study of past, present and future experiments





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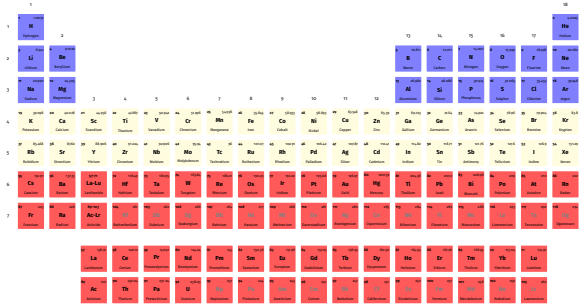
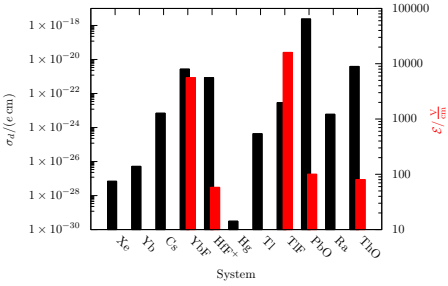
Electric Dipole Moments (EDMs) in atoms and molecules

Global ab initio study of past, present and future experiments





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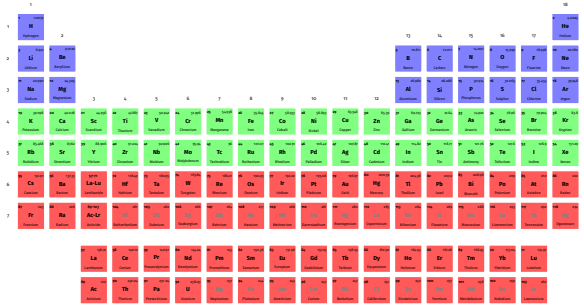
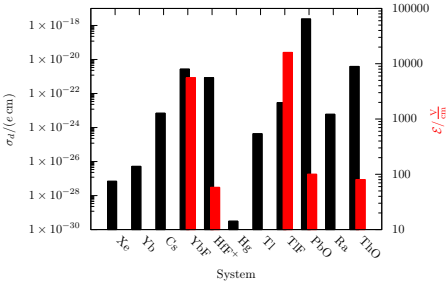
Electric Dipole Moments (EDMs) in atoms and molecules

Global ab initio study of past, present and future experiments





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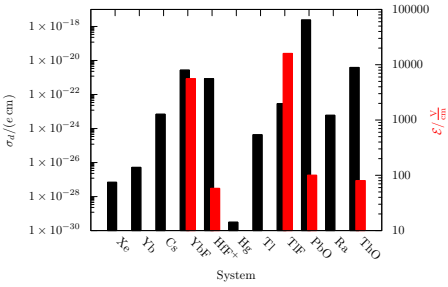
Electric Dipole Moments (EDMs) in atoms and molecules

Global ab initio study of past, present and future experiments



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Electric Dipole Moments (EDMs) in atoms and molecules

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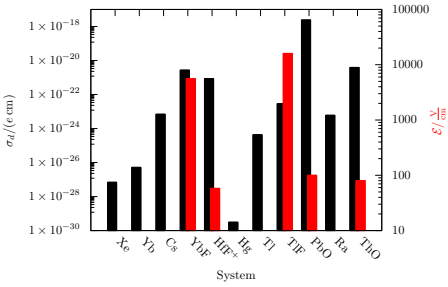

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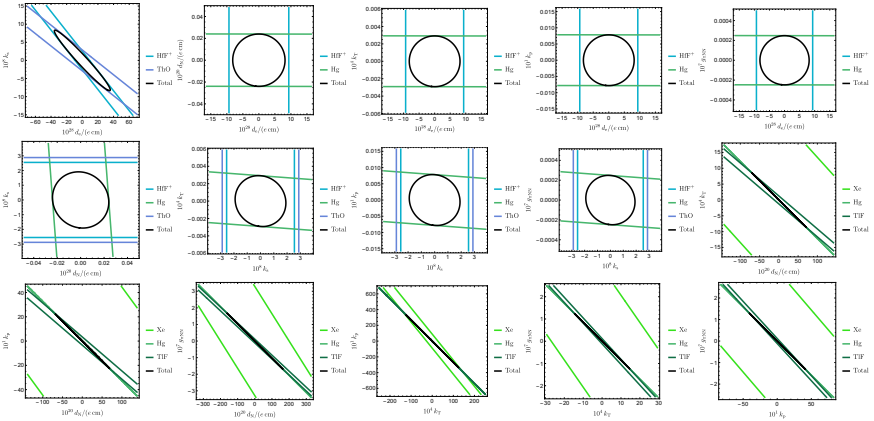
2D subspaces cut from 6D ellipsoid

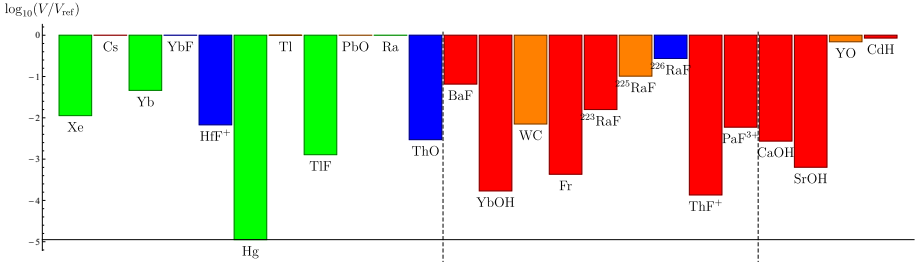
Most important: ThO, HfF^+, Hg, Xe, TlF



K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100.

Achievable uncertainties are assumed for new molecular experiments with $\sigma_d = 1 \times 10^{-23}\, e\,\mathrm{cm}^{-1}$ with typical polarization field strength, and for new atomic experiments with $\sigma_d = 1 \times 10^{-28}\, e\,\mathrm{cm}^{-1}$.

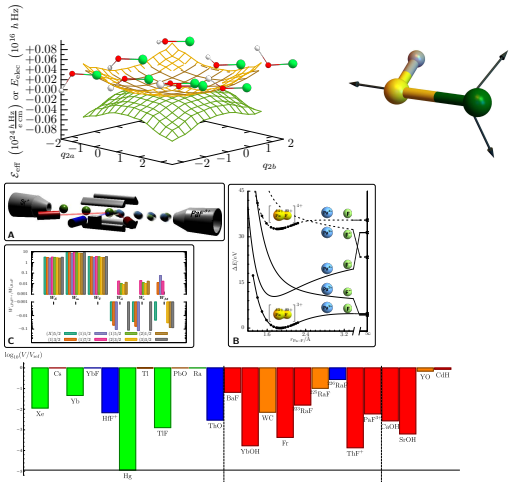
I) $\Omega > 0$ with $I = 0$
II) $\Omega = 0$ with $I \geq 1/2$
III) $\Omega > 0$ with $I = 1/2$
IV) $\Omega > 0$ with $I > 1/2$



- Nuclear structure will play an important role/change results!
- Experiments with ions and polyatomic molecules are promising.
- Uncertainties of experiments with SrOH or YO are possibly much lower.
  → Not necessary to use only the heaviest systems!

K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100.

- Polyatomic molecules
- Molecular (highly charged) ions
- Lighter systems (e.g. CaOH, YO, SrOH)



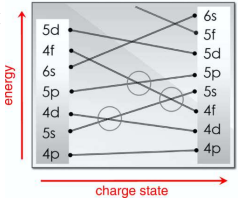
C. Zülch et al., *arXiv* **2022**, *physics.chem-ph*, 2203.10333, K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, T. A. Isaev, R. Berger, *Phys. Rev. Lett.* **2016**, *116*, 063006, I. Kozyryev, N. R. Hutzler, *Phys. Rev. Lett.* **2017**, *119*, 133002, K. Gaul, R. Berger, *Phys. Rev. A* **2020**, *101*, 012508, C. Zülch et al., *Isr. J. Chem.* **2023**, *63*, e202300035, K. Gaul et al., *Phys. Rev. A* **2024**, *109*, 042819.

Highly charged polar radioactive molecules

Combining advantages of

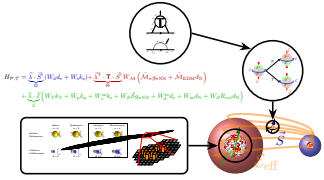
Highly charged atoms:

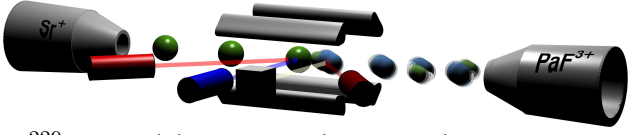
- Large relativistic effects
- Compressed level structure
- Sympathetic cooling



Polar heavy-elemental molecules:

- Large internal fields
- Pronounced relativistic effects
- Easily polarizable
- Vibrational and rotational level structure





- PaF^{3+} as ^{229}Pa containing molecule isoelectronic to RaF
- Sympathetic cooling with Sr^+ ($m/Q \sim 83\,\mathrm{u}/e$)
- Trapping and buffer-gas cooling
- Direct laser-cooling?

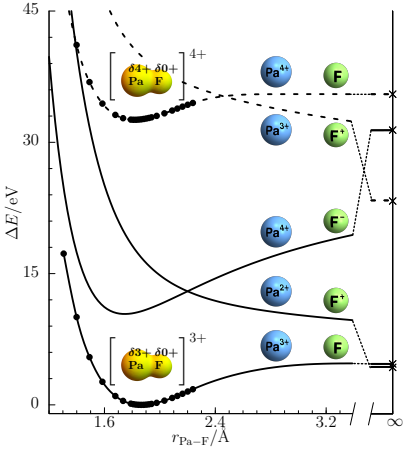
C. Zülch et al., *arXiv* **2022**, *physics.chem-ph*, 2203.10333, C. Zülch et al., *Isr. J. Chem.* **2023**, *63*, e202300035, J. Stricker et al., *arXiv* **2025**, *physics.atom-ph*, 2503.05759.

- MX^{3+} can be stable if IP of M^{3+} is about IP of X.

⇒ Actinides combined with F, O or N

- UF^{3+} known → AcF^{3+}, ThF^{3+}, PaF^{3+}
- PaF^{3+} has a single valence electron (isoelectronic to RaF).
- RECP-CCSD(T) calculations combined with ZORA-DFT

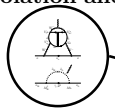
⇒ Very stable bond in PaF^{3+}

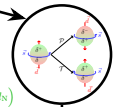


D. Schröder, H. Schwarz, *J. Phys. Chem. A* **1999**, *103*, 7385–7394.

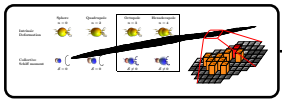
C. Zülch et al., *arXiv* **2022**, *physics.chem-ph*, 2203.10333, C. Zülch et al., *Isr. J. Chem.* **2023**, *63*, e202300035, J. Stricker et al., *arXiv* **2025**, *physics.atom-ph*, 2503.05759.

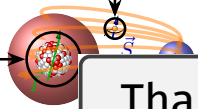
CP-violation and EDMs





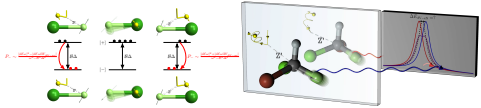
$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda}\cdot\vec{S}}_{\Omega}\,(W_d d_e + W_s k_s) + \underbrace{\vec{\lambda}^{\mathrm{T}}\cdot\mathbf{T}\cdot\vec{S}}_{\Theta} W_{\mathcal{M}}\left(\tilde{\mathcal{M}}_s g_{\pi\mathrm{NN}} + \tilde{\mathcal{M}}_{\mathrm{EDM}} d_{\mathrm{N}}\right)$$
$$+ \underbrace{\vec{\lambda}\cdot\vec{I}}_{\mathcal{I}}\left(W_{\mathrm{T}} k_{\mathrm{T}} + W_p k_p + W_a^m k_a + W_S \tilde{S} g_{\pi\mathrm{NN}} + W_a^m d_e + W_m d_{\mathrm{N}} + W_S R_{\mathrm{vol}} d_{\mathrm{N}}\right)$$

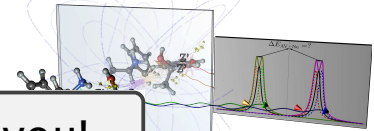




Parity violation

Rovibrational spectroscopy

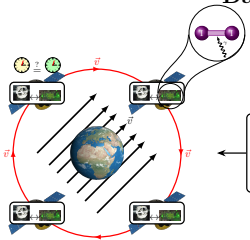




NMR spectroscopy

Thank you!

Dark matter and Lorentz invariance violation



$$\mathcal{L}_{\mathrm{e}}^{\mathrm{SME}} = \hbar c\bar{\psi}_{\mathrm{e}}\left[\tilde{c}_{\mu\nu}\gamma^{\mu} + \tilde{d}_{\mu\nu}\gamma_5\gamma^{\mu} + \tilde{e}_{\nu} + \tilde{f}_{\nu}\gamma_5 + \frac{1}{2}\tilde{g}_{\lambda\mu\nu}\sigma^{\lambda\mu}\right]\frac{\imath}{2}\overleftrightarrow{\partial}^{\nu}\psi_{\mathrm{e}}$$
$$-m_{\mathrm{e}}c^2\bar{\psi}_{\mathrm{e}}\left[\tilde{a}_{\mu}\gamma^{\mu} + \tilde{b}_{\mu}\gamma_5\gamma^{\mu} + \frac{1}{2}\tilde{h}_{\mu\nu}\sigma^{\mu\nu}\right]\psi_{\mathrm{e}}$$