A Global Analysis of EDMs from an Electronic Structure Perspective

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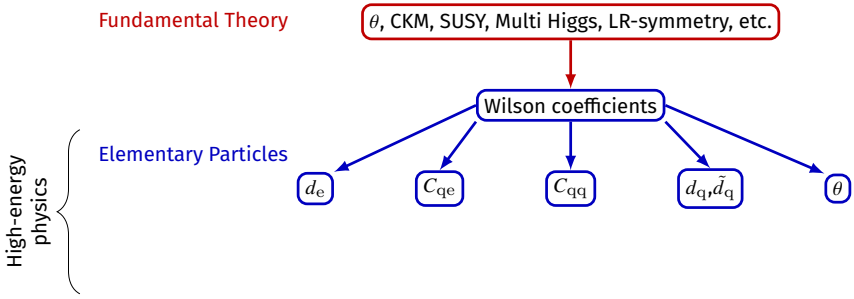
EDMs 2026 (Electric Dipole Moments 2026): Complementary Experiments and Theory Connections,
Les Houches, March 1 to March 6 2026

From elementary particles to EDMs of atoms and molecules



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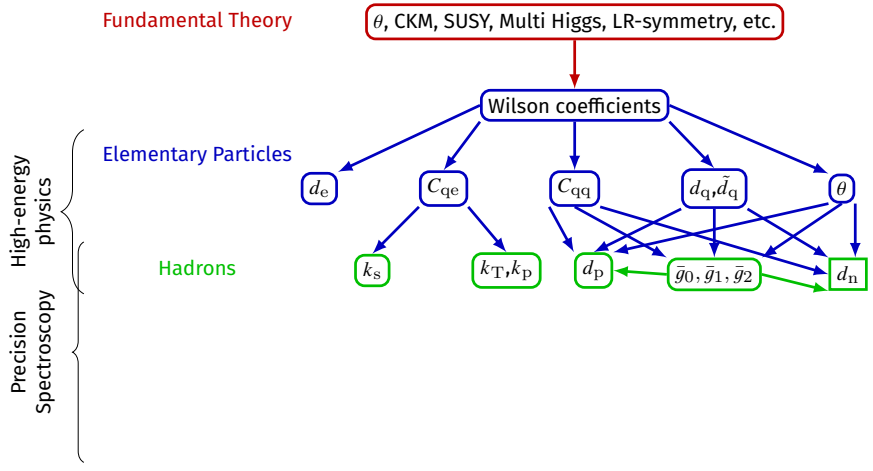

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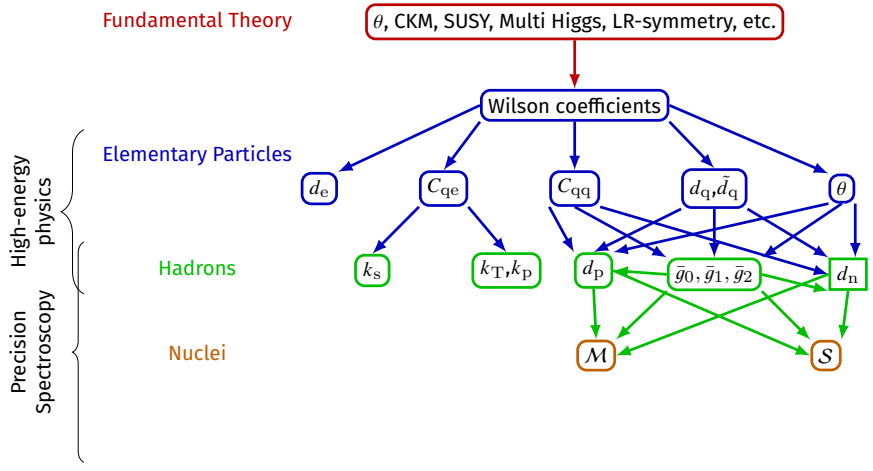

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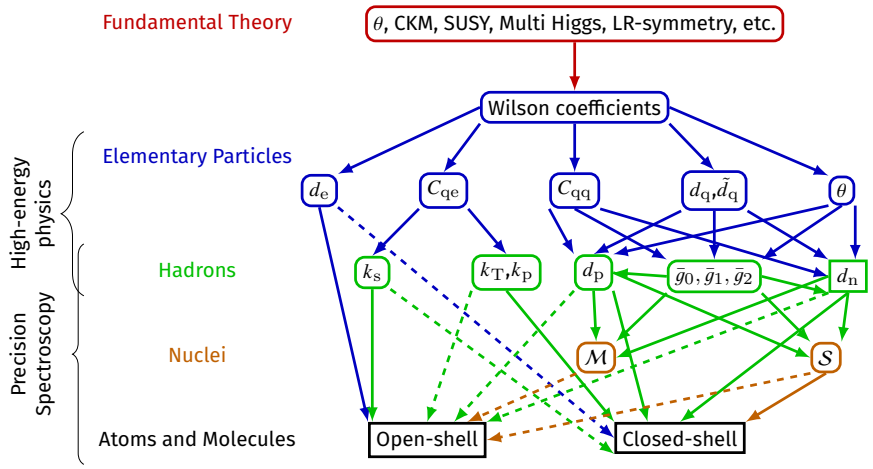

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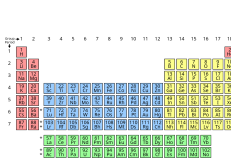
Electron-nucleon-level parameter space:

Nine parameters (electron-hadron level)	Seven parameters (global bounds)	Six parameters (qualitative analysis)
d_e	d_e	d_e
$d_\mathrm{n}(d_\mathrm{n}^\mathrm{sr}, \bar{g}_0, \bar{g}_1, \bar{g}_2)$ $d_\mathrm{p}(d_\mathrm{p}^\mathrm{sr}, \bar{g}_0, \bar{g}_1, \bar{g}_2)$	$d_\mathrm{n}^\mathrm{sr} = -d_\mathrm{p}^\mathrm{sr}$	d_N
$\bar{g}_0, \bar{g}_1, \bar{g}_2$	$\bar{g}_0, \bar{g}_1$	$g_{\pi\mathrm{NN}}$
$k_\mathrm{s} = \frac{A}{Z} C_\mathrm{s} = C_\mathrm{s,p} + \frac{N}{Z} C_\mathrm{s,n}$	k_s	k_s
$k_\mathrm{T} = \langle \boldsymbol{\sigma} \rangle_\mathrm{N} C_\mathrm{T} \approx \sum_{n_\mathrm{p}} \langle \boldsymbol{\sigma} \rangle_{n_\mathrm{p}} C_\mathrm{T,p} + \sum_{n_\mathrm{n}} \langle \boldsymbol{\sigma} \rangle_{n_\mathrm{n}} C_\mathrm{T,n}$	k_T	k_T
$k_\mathrm{p} = \langle \boldsymbol{\sigma} \rangle_\mathrm{N} C_\mathrm{p} \approx \sum_{n_\mathrm{p}} \langle \boldsymbol{\sigma} \rangle_{n_\mathrm{p}} C_\mathrm{p,p} + \sum_{n_\mathrm{n}} \langle \boldsymbol{\sigma} \rangle_{n_\mathrm{n}} C_\mathrm{p,n}$	k_p	k_p

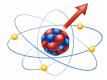
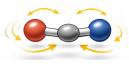
K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100, T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **2015**, *91*, 035502, S. Degenkolb et al., *arXiv* **2024**, *hep-ph*, 2403.02052.

Why molecules?

Pro	Contra
✓Complex many body system	✗Complex many body system
✓Accessible internally broken symmetries	✗Lower resolution than in atoms
✓Can have a simpler electronic structure than atoms	✗Ab initio description can be limited
✓Recent breakthroughs in molecular spectroscopy	



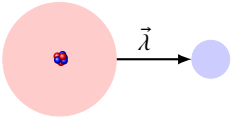
$\Rightarrow$ Tailoring a tuned quantum sensor from the periodic table!

Atoms	Polar diatomic	Linear polyatomic	Non-linear polyatomic
			
$\sum_a \frac{\langle 0 \vert \vec{r}\cdot\vec{\mathcal{E}} \vert a\rangle\langle a \vert \hat{H}_{\mathcal{P},\mathcal{T}} \vert 0\rangle}{E_0 - E_a}$	$E_0 - E_a \lesssim \mathrm{THz}$	$E_0 - E_a \lesssim \mathrm{GHz}$	$E_0 - E_a \lesssim \mathrm{MHz}$	$E_0 - E_a \lesssim \mathrm{kHz}$

$\Rightarrow$ Molecular enhancement **3-6 orders of magnitude**

Effective $\mathcal{P},\mathcal{T}$-odd Hamiltonian

Linear molecule with one heavy nucleus



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

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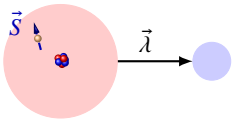

I) $\Omega > 0$ with $I = 0$

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

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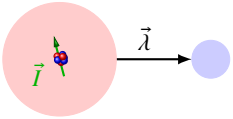

I) $\Omega > 0$ with $I = 0$
II) $\Omega = 0$ with $I \geq 1/2$

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

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

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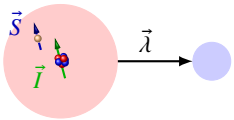

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$$H_{\mathcal{P},\mathcal{T}} = \underbrace{\vec{\lambda}\cdot\vec{S}'}_{\Omega}\left(W_\mathrm{d} d_\mathrm{e} + W_\mathrm{s} k_\mathrm{s}\right) + \underbrace{\vec{\lambda}\cdot\vec{I}}_{I}\left(W_\mathrm{T} k_\mathrm{T} + W_\mathrm{p} k_\mathrm{p}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

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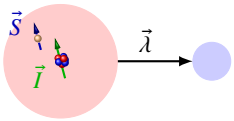

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$$+ \underbrace{\vec{\lambda}\cdot\vec{I}}_{I}\left(W_\mathrm{T} k_\mathrm{T} + W_\mathrm{p} k_\mathrm{p} + W_\mathrm{s}^\mathrm{m} k_\mathrm{s} + W_\mathcal{S}\tilde{\mathcal{S}} g_{\pi\mathrm{NN}} + W_\mathrm{d}^\mathrm{m} d_\mathrm{e} + W_\mathrm{m}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

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 $R_\mathrm{vol}, \tilde{\mathcal{S}}, \tilde{\mathcal{M}}_\mathrm{EDM}, \tilde{\mathcal{M}}_\pi$

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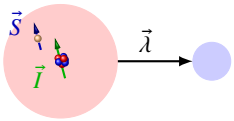

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- Nuclear structure factors $R_\mathrm{vol}, \tilde{\mathcal{S}}, \tilde{\mathcal{M}}_\mathrm{EDM}, \tilde{\mathcal{M}}_\pi$

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

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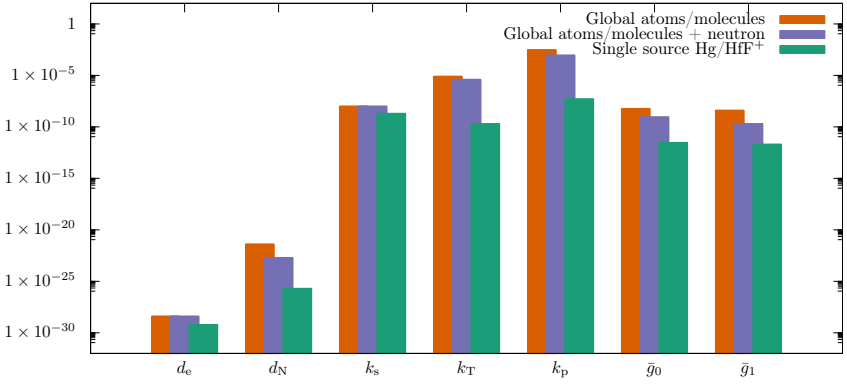
Single source model vs. global analysis



Experimental uncertainties 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, ab initio electronic structure calculations from K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100 and nuclear structure data from T. E. Chupp et al., *Rev. Mod. Phys.* **2019**, *91*, 015001

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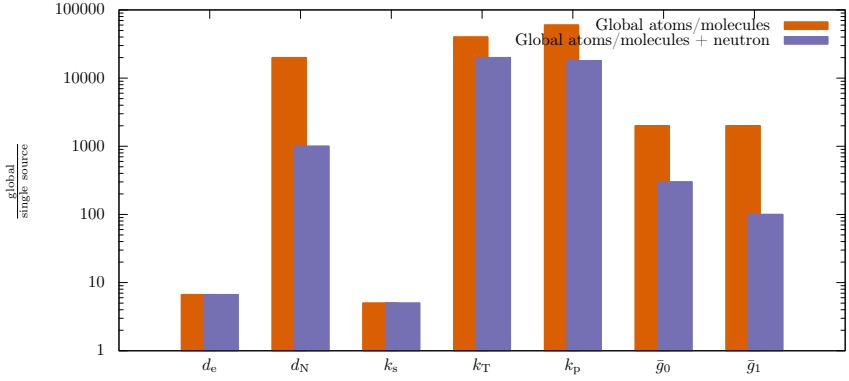

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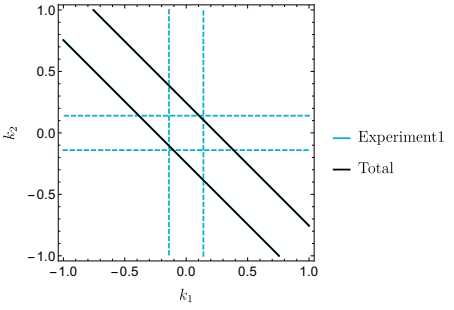
Global $\mathcal{P},\mathcal{T}$-odd measurement model

- 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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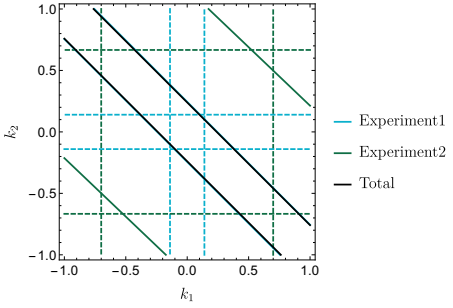

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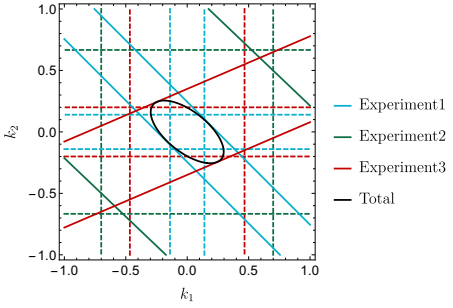

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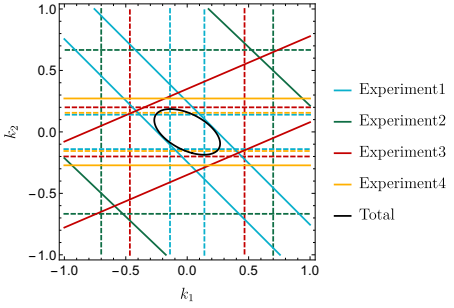
Determinant is maximal if all measurements have orthogonal sensitivities!

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**.

Global $\mathcal{P},\mathcal{T}$-odd measurement model

- 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!

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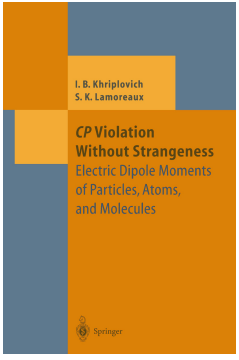
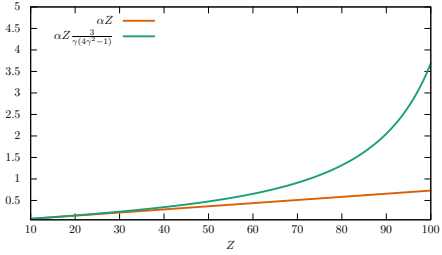


- Simple effective one-electron wave functions at distances $r \ll a_0 Z^{-1/3}$:

$$f_\kappa(r) = \frac{\kappa}{|\kappa| r \sqrt{Z a_0 \nu_\kappa^3}} \left((\kappa + \gamma_\kappa) J_{2\gamma_\kappa} \left(\sqrt{\frac{8Zr}{a_0}} \right) - \sqrt{\frac{2Zr}{a_0}} J_{2\gamma_\kappa - 1} \left(\sqrt{\frac{8Zr}{a_0}} \right) \right),$$

$$g_\kappa(r) = \frac{\kappa Z \alpha}{|\kappa| r \sqrt{Z a_0 \nu_\kappa^3}} J_{2\gamma_\kappa} \left(\sqrt{\frac{8Zr}{a_0}} \right),$$



- Effective quantum numbers ν contain all many-body information ($O(\nu_\kappa) \sim 1$!)

$\Rightarrow$ Estimate the expectable order of magnitude of a property!

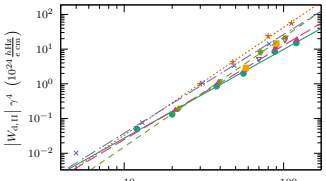
E. Fermi, E. Segrè, *Z. Phys.* **1933**, *82*, 729–749, L. L. Foldy, *Phys. Rev.* **1958**, *111*, 1093–1098, N. Fröman, P. O. Fröman, *Phys. Rev. A* **1972**, *6*, 2064–2067, C. Bouchiat, M. A. Bouchiat, *J. Phys. (Paris)* **1975**, *36*, 493–509.

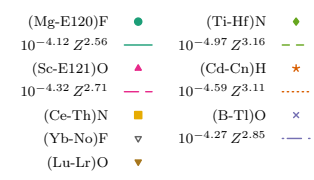
W_i	αZ-scaling	Relativistic enhancement	Reference
W_d	$\alpha^2 Z^3$	$\frac{3}{\gamma_{1/2}(4\gamma_{1/2}^2-1)}$	P. Sandars, *Phys. Lett.* **1965**, *14*, 194–196, V. V Flambaum, *Yad. Fiz.* **1976**, *24*, 383–386
W_d^m	αZ^2	$R(Z,A)-1$	V. V. Flambaum, I. B. Khriplovich, *Phys. Lett. A* **1985**, *110*, 121–125
W_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	$\frac{720\Gamma(\gamma_{1/2}+\gamma_{3/2}-2)}{\Gamma(3+\gamma_{1/2}-\gamma_{3/2})\Gamma(3-\gamma_{1/2}+\gamma_{3/2})\Gamma(3+\gamma_{1/2}+\gamma_{3/2})}$	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.

Do the analytic models work?

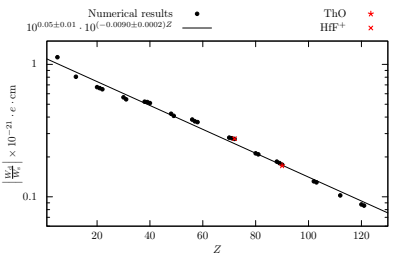
Open-shell molecules: $\mathcal{P}, \mathcal{T}$-odd ratio and coverage region in d_e-k_s parameter space



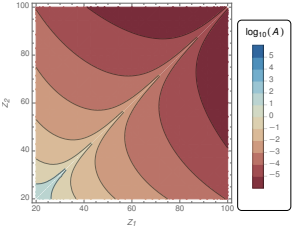


$$A \sim W_{d,1}^{-1} W_{d,2}^{-1} \left| \frac{W_{s,1}}{W_{d,1}} - \frac{W_{s,2}}{W_{d,2}} \right|^{-1}$$

$$A \approx \frac{k_p^2 \pi |u(v_1) u(v_2)|}{10^{b_{d,1}+b_{d,2}} \frac{Z_1^{a_{d,1}} Z_2^{a_{d,2}}}{\gamma_1^4 \gamma_2^4} \cdot 0.9 \cdot \left| 1.02 Z_1 - 1.02 Z_2 \right| \times 10^{27} \ \frac{\text{Hz}^2}{e \cdot \text{cm}}}$$

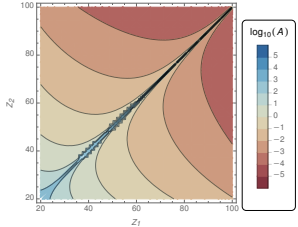


Analytical atomic model



vs.

Ab initio model: group 2, group 12



K. Gaul et al., *Phys. Rev. A* **2019**, *99*, 032509, T. Fleig, *Phys. Rev. A* **2017**, *96*, 040502, T. Fleig, M. Jung, *J. High Energy Phys.* **2018**, *2018*, 12.

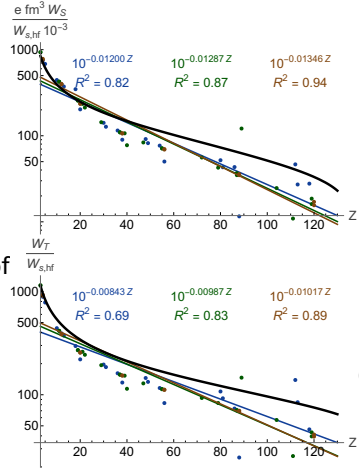
Do the analytic models work?

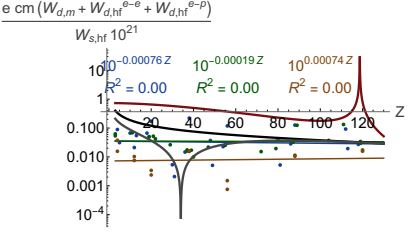
Closed shell atoms/molecules: $\mathcal{P}, \mathcal{T}$-odd ratio for nuclear spin-dependent parameter spaces

Antonio Rivera Vargas

RPA-HF calculations of a large set of closed-shell

- atoms (Be-E120), (Zn-Cn), $(\text{B-Nh})^+$, (He-Og)
- molecules (Li-E119)F/H, (Sc-Ac)F, (Ti-Rf)O^+, (Cu-Ag)H, (Be-Ra)CC/SH^+





Cancellation of W_{d}^{hf} at $Z = 34$

Random phase approximation/coupled perturbed HF

DFT calculations, finite nucleus effects and many body effects on W_{d} are currently analysed.

Do the analytic models work?

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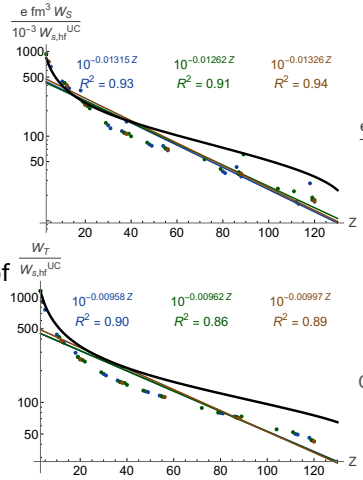
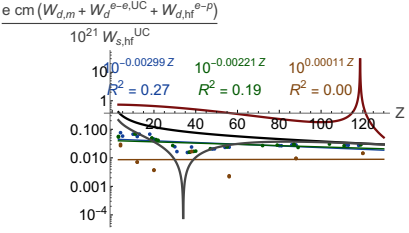




Cancellation of $W_{\mathrm{d}}^{\mathrm{hf}}$ at $Z = 34$

Sum over states/uncoupled HF "no correlation"

DFT calculations, finite nucleus effects and many body effects on W_{d} are currently analysed.

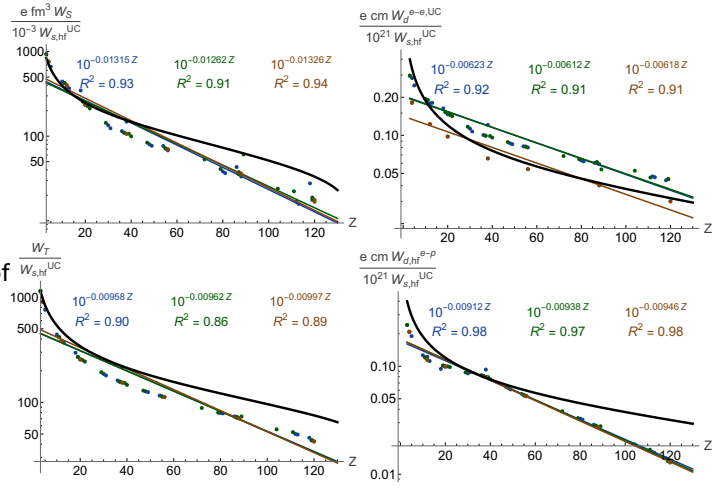
Do the analytic models work?

Closed shell atoms/molecules: $\mathcal{P}, \mathcal{T}$-odd ratio for nuclear spin-dependent parameter spaces

Antonio Rivera Vargas

RPA-HF calculations of a large set of closed-shell

- atoms (Be-E120), (Zn-Cn), $(B\text{-}Nh)^+$, (He-Og)
- molecules (Li-E119)F/H, (Sc-Ac)F, $(Ti\text{-}Rf)O^+$, (Cu-Ag)H, $(Be\text{-}Ra)CC/SH^+$



Sum over states/uncoupled HF "no correlation"

DFT calculations, finite nucleus effects and many body effects on W_{cl} are currently analysed.

Do the analytic models work?

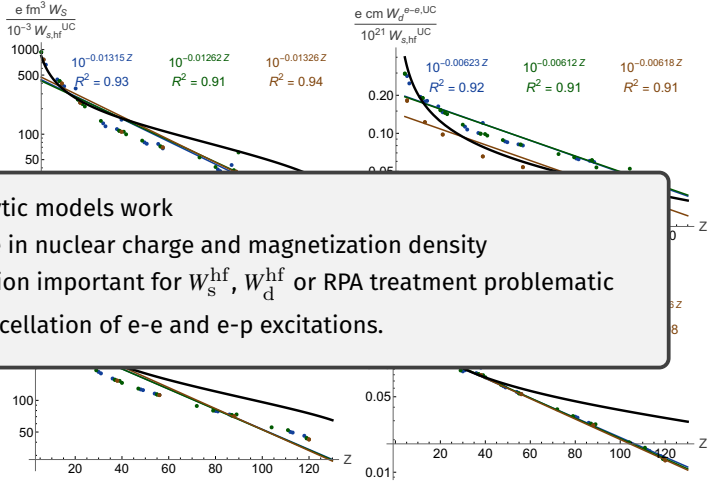
Closed shell atoms/molecules: $\mathcal{P},\mathcal{T}$-odd ratio for nuclear spin-dependent parameter spaces

Antonio Riv

RPA-HF calc

closed-shel

- atoms (
  (B-Nh)$^+$, (Fr-Og)
- molecules (Li-E119)F/H, (Sc-Ac)F, (Ti-Rf)O$^+$, (Cu-Ag)H, (Be-Ra)CC/SH$^+$



- On average analytic models work
- Misses difference in nuclear charge and magnetization density
- Electron correlation important for $W_{\mathrm{s}}^{\mathrm{hf}}$, $W_{\mathrm{d}}^{\mathrm{hf}}$ or RPA treatment problematic
- $W_{\mathrm{d}}^{\mathrm{hf}}$ features cancellation of e-e and e-p excitations.

Sum over states/uncoupled HF "no correlation"

DFT calculations, finite nucleus effects and many body effects on W_{d} are currently analysed.

Global minimization within a simplified model

Purpose and limitations of the model:

- ✓ 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

Additional approximations:

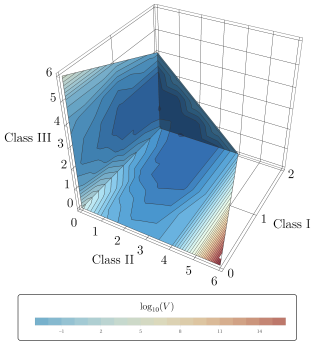
- Only 6 experiments with fixed uncertainties $\sigma_{\tilde{\nu},0} = 1\,\mathrm{arb.u.}$: $V \sim |\det(\boldsymbol{W})|^{-1}$
- Crude estimates for nuclear structure:

$$\begin{aligned}
\mathcal{S}_{\mathrm{EDM}} &\approx \frac{3}{50} A^{2/3} (1.2\,\mathrm{fm})^2 \frac{3+2I}{3+3I} (d_\mathrm{p} + d_\mathrm{n}) \\
\mathcal{S}_\pi &\approx \left(0.02 \frac{18e}{20\pi\sqrt{35}} \frac{0.14Z}{A^{1/3}} (1.2\,\mathrm{fm} A^{1/3})^3 \right. \\
&\quad \left. + \frac{3eq_\mathrm{ext}}{50} 0.14\,\mathrm{fm} (1.2\,\mathrm{fm} A^{1/3})^2 \right) \frac{3+2I}{3+3I} g_{\pi\mathrm{NN}} \\
\mathcal{M}_{\mathrm{EDM}} &\approx \frac{-3}{2m_\mathrm{p}} \frac{1}{I+1} (d_\mathrm{p} + d_\mathrm{n}) \\
\mathcal{M}_\pi &\approx \frac{3e(\mu - q_\mathrm{ext})}{2m_\mathrm{p}} 1.4 \times 10^{-14}\,\mathrm{cm} \frac{1}{I+1} g_{\pi\mathrm{NN}},
\end{aligned}$$

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

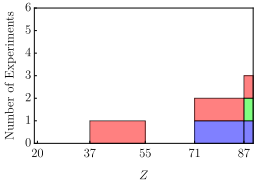
K. Gaul, R. Berger, *J. High Energ. Phys.* **2024,** *2024*, 100, I. B. Khriplovich, S. K. Lamoreaux, ***CP Violation without Strangeness,*** Springer, Berlin, **1997,** J. S. M. Ginges, V. V. Flambaum, *Phys. Rep.* **2004,** *397*, 63–154.

Minimization of the coverage volume within the simple model

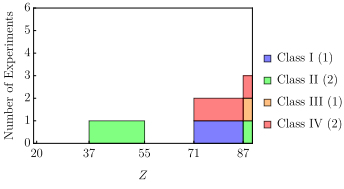


I) $\Omega > 0$ with $I = 0$; II) $\Omega = 0$ with $I \geq 1/2$
III) $\Omega > 0$ with $I = 1/2$; IV) $\Omega > 0$ with $I > 1/2$

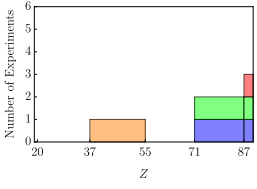
1. Trivial requirements: $N_{\mathrm{I}} \leq 2$, $N_{\mathrm{I}} + N_{\mathrm{II}} + N_{\mathrm{III}} + N_{\mathrm{IV}} \geq 6$
2. $N_{\mathrm{II,III,IV}} \leq 3$
3. If $N_A = 3$, $N_B \leq 2\ \forall A, B \in \{\mathrm{II}, \mathrm{III}, \mathrm{IV}\}$



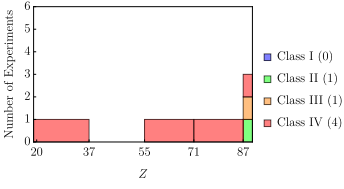
$V = 1.0 \times 10^{-3}$ arb.u. (1.)



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



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



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

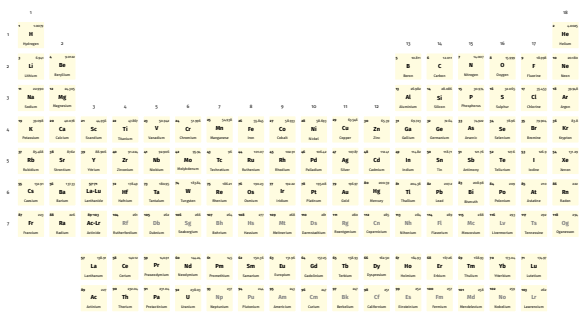
$\Rightarrow$ Same class $\rightarrow$ considerably different Z.

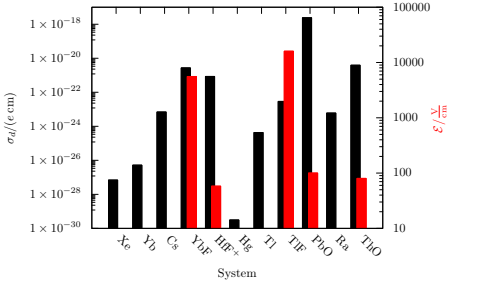
$\Rightarrow$ ≥ 3 of same class $\rightarrow$ system with lower Z

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

Global ab initio study of current experimental status

Past, present and future atomic and molecular experiments





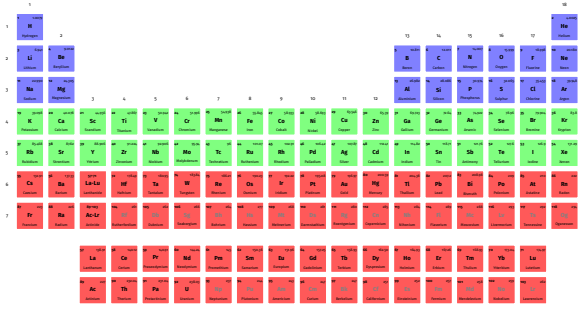
- "Modern" experiments with: Xe, Yb, Hg, TlF, Ra, Cs, HfF^+, Tl, PbO, ThO
- Planned experiments BaF, WC, Fr, RaF, YbOH, ThF^+, etc.

- DFT calculation of W_i ($\sim 20\,\%$ accuracy, better for most)

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.

Global ab initio study of current experimental status

Past, present and future atomic and molecular experiments



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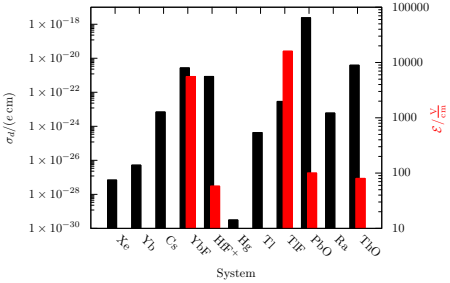

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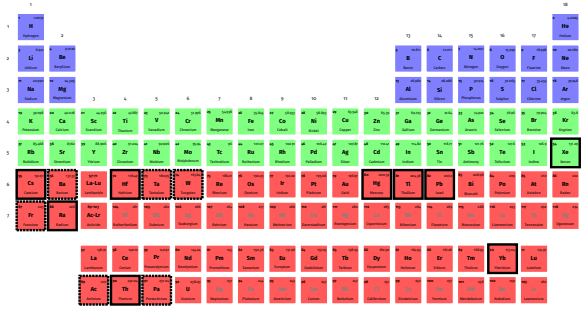
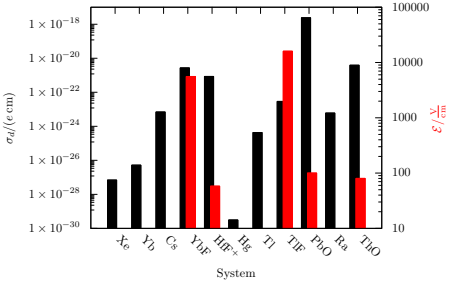
Global ab initio study of current experimental status

Past, present and future atomic and molecular experiments





- "Modern" experiments with: Xe, Yb, Hg, TlF, Ra, Cs, HfF^+, Tl, PbO, ThO
- Planned experiments BaF, WC, Fr, RaF, YbOH, ThF^+, etc.

- DFT calculation of W_i ($\sim 20\,\%$ accuracy, better for most)

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.

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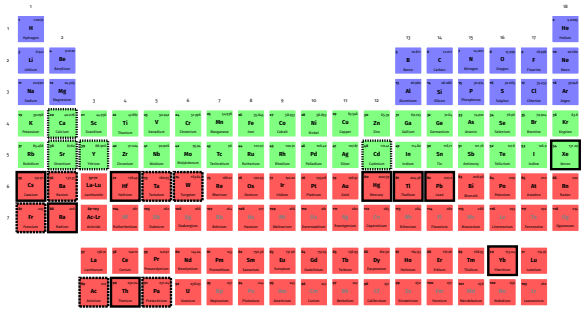

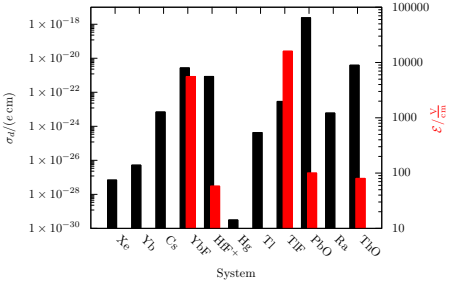

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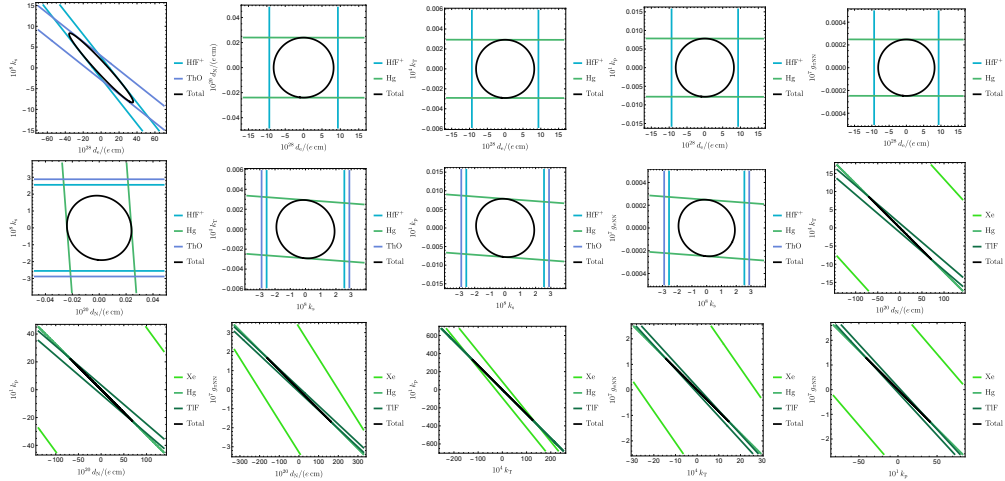
F. Allmendinger et al., *Phys. Rev. A* **2019**, *100*, 022505, N. Sachdeva et al., *Phys. Rev. Lett.* **2019**, *123*, 143003, S. A. Murthy et al., *Phys. Rev. Lett.* **1989**, *63*, 965–968, T. A. Zheng et al., *Phys. Rev. Lett.* **2022**, *129*, 083001, J. J. Hudson et al., *Phys. Rev. Lett.* **2002**, *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.

Current experimental status (“rule-of-thumb nuclear structure”)

2D subspaces cut from 6D ellipsoid

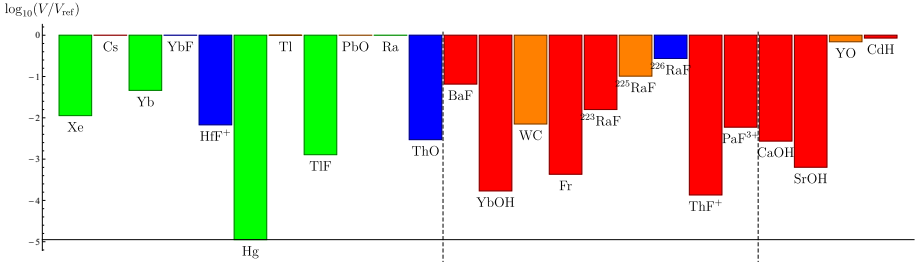
Most important: ThO, HfF^+, Hg, Xe, TlF



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 input will change results!
- Experiments with ions and polyatomic molecules are promising.
- Uncertainties of experiments with SrOH or YO are possibly much lower.
  → From a global view not necessary to use only the heaviest systems!

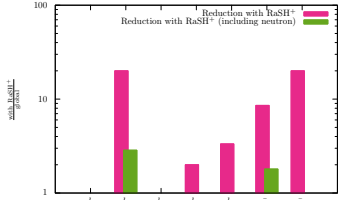
K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, 2024, 100.

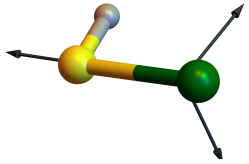
Polyatomic radioactive molecular ions

PHYSICAL REVIEW A **109**, 042819 (2024)

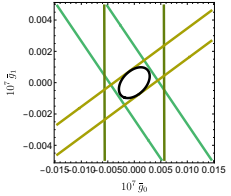
$\mathcal{CP}$-violation sensitivity of closed-shell radium-containing polyatomic molecular ions

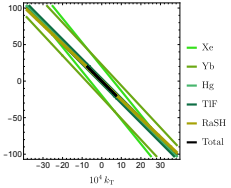
Konstantin Gaul,[1,*] Nicholas R. Hutzler,[2] Phelan Yu,[2] Andrew M. Jayich,[3] Miroslav Iliaš,[4] and Anastasia Borschevsky[5]





Reduction of volume **3-4 orders** of magnitude.



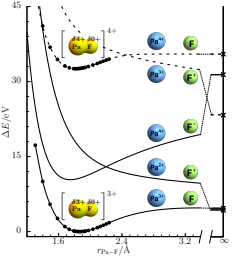


- Single molecule in an ion trap statistic uncertainty ($RaOCH_3^+$) $\sigma_\nu \sim 65\mu\mathrm{Hz}$[a]:
- Taking nuclear Schiff moment from T. E. Chupp et al., *Rev. Mod. Phys.* **2019**, *91*, 015001
- Non-zero asymmetry $\eta_{\mathcal{P},\mathcal{T}} = \frac{3(W_{\mathcal{P},\mathcal{T},b} - W_{\mathcal{P},\mathcal{T},c})}{2W_{\mathcal{P},\mathcal{T},a} - W_{\mathcal{P},\mathcal{T},b} - W_{\mathcal{P},\mathcal{T},c}}$
- $\eta_S \sim -2 \times 10^{-3}$, $\eta_\mathrm{d} \sim 6 \times 10^{-3}$
- Asymmetry not equal for all properties! → Artifact or physics?
- Advantages with strongly asymmetric tops to disentangle sources of $\mathcal{P}, \mathcal{T}$-violation?

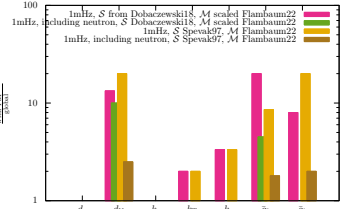
[a] M. Fan et al., *Phys. Rev. Lett.* **2021**, *126*, 023002, P. Yu, N. R. Hutzler, *Phys. Rev. Lett.* **2021**, *126*, 023003.

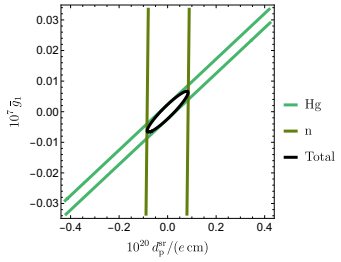
Multiply charged polar radioactive molecules

$^{229}PaF^{3+}$



- Stable chemical bond ($D_e > 4\,\mathrm{eV}$)
- Simple electronic structure: $^2\Phi_{5/2}$ ground state



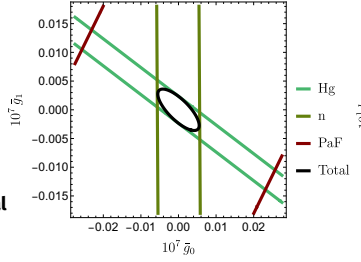


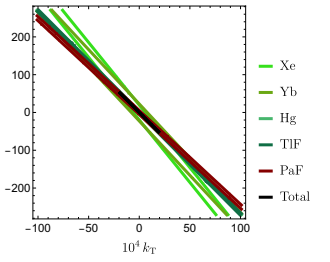
- Schiff moment from J. Dobaczewski et al., *Phys. Rev. Lett.* **2018**, *121*, 232501 ($\Delta E \sim 100\,\mathrm{keV}$)

⇒ Reduction of volume **1-2 orders** of magnitude.

- Schiff moment from V. Spevak et al., *Phys. Rev. C* **1997**, *56*, 1357–1369**,** V. V. Flambaum, H. Feldmeier, *Phys. Rev. C* **2020**, *101*, 015502 ($\Delta E \sim 0.22\,\mathrm{keV}$)

⇒ Volume reduces **3 orders** of magnitude but **global bounds do not improve!**





K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, C. Zülch et al., *arXiv* **2022**, *physics.chem-ph*, 2203.10333, C. Zülch et al., *arXiv* **2025**, *physics.chem-ph*, 2511.10791.

Multiply charged polar radioactive molecules

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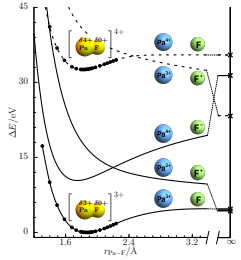

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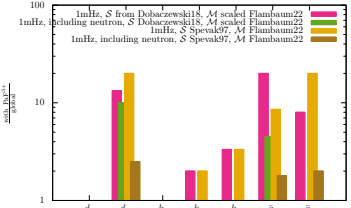
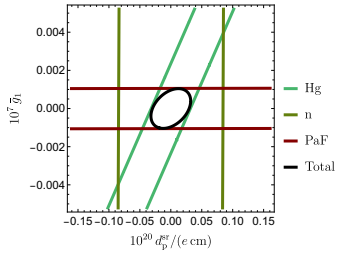
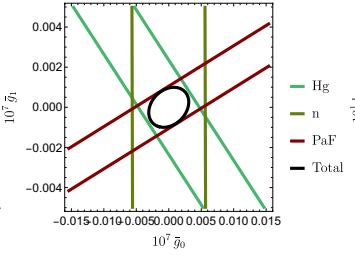
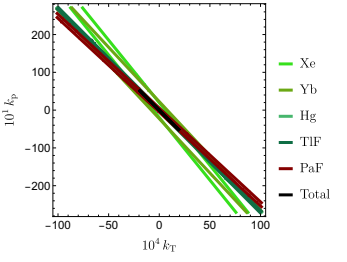




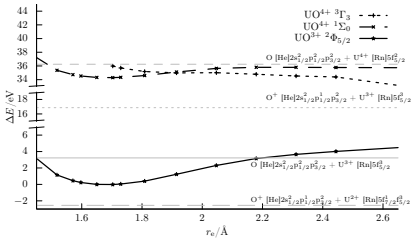


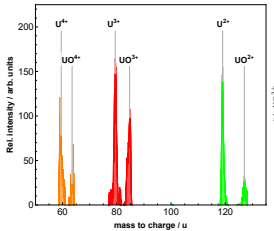


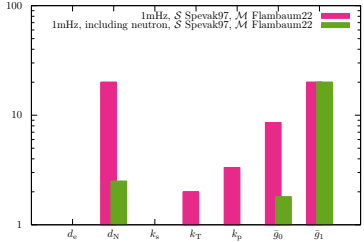
K. Gaul, R. Berger, *J. High Energ. Phys.* **2024**, *2024*, 100, C. Zülch et al., *arXiv* **2022**, *physics.chem-ph*, 2203.10333, C. Zülch et al., *arXiv* **2025**, *physics.chem-ph*, 2511.10791.

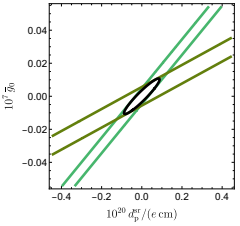
Multiply charged polar radioactive molecules

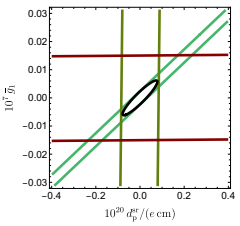
$^{235}UO^{3+}$

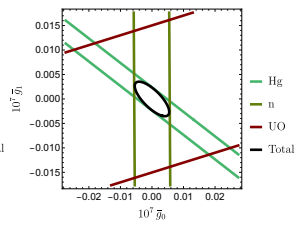


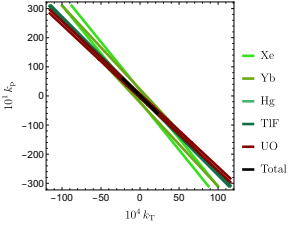








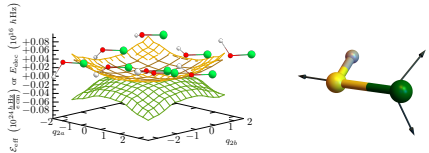


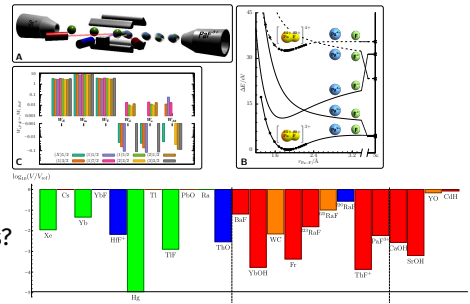


J. Stricker et al., *Phys. Rev. A* **2025**, *112*, 012821, J. Stricker et al., *arXiv* **2025**, *physics.chem-ph*, 2512.14924.

Summary & Outlook: Next generation molecular EDM experiments?

- Polyatomic molecules
- Radioactive molecules and molecular ions
- Lighter molecules (CaOH, YO, SrOH) for global analysis?

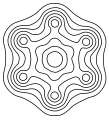




Thank You for Your Attention!

Collaborators:

- Robert Berger (Marburg)
- Ronald Garcia Ruiz (MIT, Boston)
- Stephan Malbrunot-Ettenauer (CERN/TRIUMF)
- Jonas Stricker, Christoph Düllmann (Mainz)
- Anastasia Borschevsky (RU Groningen)
- Nick Hutzler (CalTec)
- Andrew Jayich (UC Santa Barbara)



FCI
FONDS DER
CHEMISCHEN
INDUSTRIE

HIM
HELMHOLTZ
Helmholtz-Institut Mainz

JG|U

Effective P,T-odd Hamiltonian

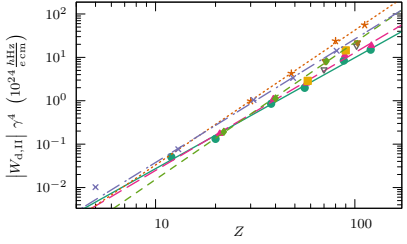
$W_{\mathrm{d}} =$	$\dfrac{\left\langle \frac{2c}{e\hbar} \sum\limits_{i=1}^{N_{\mathrm{elec}}} \imath\boldsymbol{\gamma}_i^0\boldsymbol{\gamma}_i^5\hat{\vec{p}}_i^2 \right\rangle}{\Omega}$	A. Mårtensson-Pendrill, P Öster, *Phys. Scr.* **1987**, *36*, 444–452
$W_{\mathrm{d}}^{\mathrm{m}} =$	$\vec{I}\cdot\left\langle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

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

Chemical influence on scaling behavior of eEDM interactions

Empirical atomic model $W_\mathrm{d} \sim \frac{\alpha^2 Z^3}{\gamma^4}$; ZORA-cGKS-B3LYP calculations

$$\log_{10}\left\{|W_\mathrm{d}|\gamma^4 \times 10^{-24}\ \frac{e\mathrm{cm}}{h\mathrm{Hz}}\right\} = b_{\mathrm{d,FS}} + a_{\mathrm{d,FS}} \log_{10}\{Z\}$$



(Mg-E120)F — $10^{-4.12}\ Z^{2.56}$

(Sc-E121)O — $10^{-4.32}\ Z^{2.71}$

(Ce-Th)N

(Yb-No)F

(Lu-Lr)O

(Ti-Hf)N — $10^{-4.97}\ Z^{3.16}$

(Cd-Cn)H — $10^{-4.59}\ Z^{3.11}$

(B-Tl)O — $10^{-4.27}\ Z^{2.85}$

$\Rightarrow$ Chemically suppressed Z-scaling in group 2 and 3

$\Rightarrow$ Chemical enhancement in group 4 and 12

K. Gaul et al., *Phys. Rev. A* **2019**, *99*, 032509.

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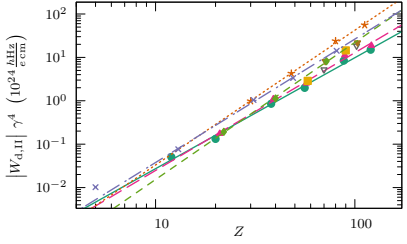

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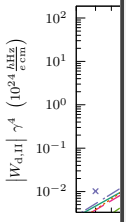
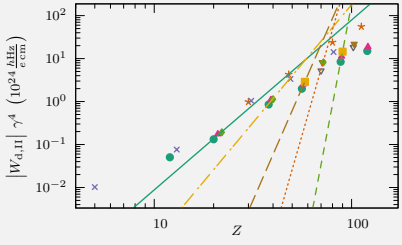
Chemical influence on scaling behavior of eEDM interactions

Empirical atomic model $W_\mathrm{d} \sim \frac{\alpha^2 Z^3}{\gamma^4}$; ZORA-cGKS-B3LYP calculations

- Chemical enhancement enormous for isoelectronic species:





(Mg-E120)F
(Sc-E121)O
(Ce-Th)N
(Yb-No)F
(Lu-Lr)O
(Ti-Hf)N
(Cd-Cn)H

(B-Tl)O

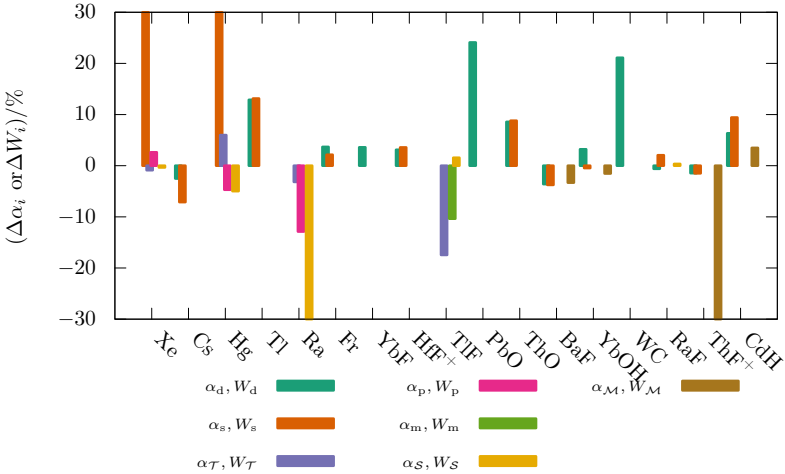
$10^{-6.01}\ Z^{3.95}$
$10^{-8.73}\ Z^{5.49}$
$10^{-18.08}\ Z^{10.52}$
$10^{-28.69}\ Z^{15.95}$
$10^{-46.03}\ Z^{24.15}$

⇒ Chemical
⇒ Chemical enhancement in group 4 and 12

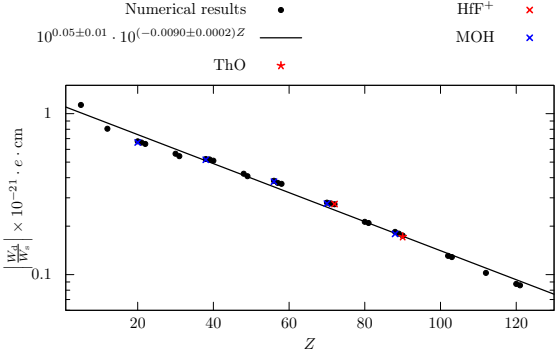
K. Gaul et al., *Phys. Rev. A* **2019**, *99*, 032509.

ZORA-cGKS-BHandH approach

- Computation of all W_i for all relevant systems
- Comparison with literature of available W_i



- W_d, W_s and $\frac{W_\mathrm{s}}{W_\mathrm{d}}$ similar to MF

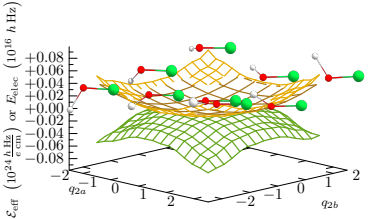


$\Rightarrow$ No advantages for disentanglement

Laser-coolable 1st excited vibrational state:

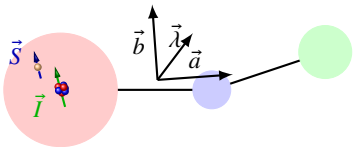
- Experiment: Internal co-magnetometer states
- Calculations: Vibrational corrections to $\mathcal{E}_\mathrm{eff}$ negligible ($< 1\%$)

$\Rightarrow$ Promising candidates for experiment



H-bending modes in YbOH

I. Kozyryev, N. R. Hutzler, *Phys. Rev. Lett.* **2017**, *119*, 133002, K. Gaul, R. Berger, *Phys. Rev. A* **2020**, *101*, 012508, A. Zakharova, A. Petrov, *Phys. Rev. A* **2021**, *103*, 032819.



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 $I > 1/2$

$$H_{\mathrm{sr}} = \vec{\lambda}^{\mathsf{T}} \cdot (\boldsymbol{W}_{\mathrm{d}} d_{\mathrm{e}} + \boldsymbol{W}_{\mathrm{s}} k_{\mathrm{s}}) \cdot \vec{J}_{\mathrm{e}} + \vec{\lambda}^{\mathsf{T}} \cdot (\mathbf{T} \cdot \boldsymbol{W}_{\mathcal{M}}) \cdot \vec{J}_{\mathrm{e}} (\tilde{\mathcal{M}}_{\pi} g_{\pi \mathrm{NN}} + \tilde{\mathcal{M}}_{\mathrm{EDM}} d_{\mathrm{N}})$$
$$+ \vec{\lambda}^{\mathsf{T}} \cdot \Big(\boldsymbol{W}_{\mathrm{T}} k_{\mathrm{T}} + \boldsymbol{W}_{\mathrm{p}} k_{\mathrm{p}} + \boldsymbol{W}_{\mathrm{s}}^{\mathrm{m}} k_{\mathrm{s}} + \boldsymbol{W}_{\mathcal{S}} \tilde{\mathcal{S}} g_{\pi \mathrm{NN}}$$
$$+ \boldsymbol{W}_{\mathrm{d}}^{\mathrm{m}} d_{\mathrm{e}} + \boldsymbol{W}_{\mathrm{m}} d_{\mathrm{N}} + \boldsymbol{W}_{\mathcal{S}} R_{\mathrm{vol}} d_{\mathrm{N}} \Big) \cdot \vec{I}_{I}$$

- $\vec{\lambda}$ is the polarization axis.
- Lowered molecular symmetry → More degrees of freedom → $\boldsymbol{W}_i$ are rank-1 tensors in the molecule-fixed coordinate system.
- May bring advantages for disentangling different sources?

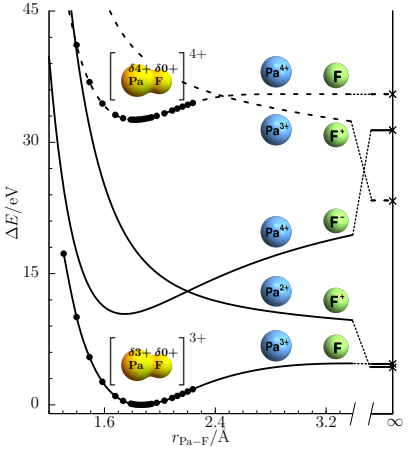
Stability of highly charged molecules

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

$\Rightarrow$ Actinides combined with F, O or N

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

$\Rightarrow$ Very stable bond in PaF^{3+}



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