Precision molecular experiments as a tool in the search for New Physics

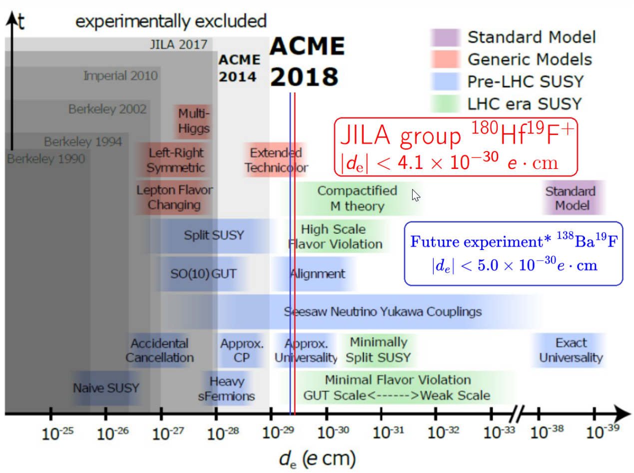
Sergey Prosnyak[1], Leonid Skripnikov[1, 2]

[1]NRC "Kurchatov Institute" - PNPI
Quantum Physics and Chemistry Department

[2]St Petersburg University
Quantum Mechanics Division

2025

EDM predictions



*A. Boeschoten et al., arXiv:2303.06402 (2023)
Compiled by D. DeMille

Reinterpretation of BaF experiment

$\mathcal{T},\mathcal{P}$-violating energy shift

$$\boxed{\delta E} = d_{\mathrm{e}}\Omega W_d \approx 8\ \mu\mathrm{Hz}$$

where $E_{\mathrm{eff}} = W_d|\Omega| \approx 1.565 \cdot 10^{24}$ Hz/(e·cm)

Reinterpretation in terms of ALPs

$$V_{eN}(\boldsymbol{r}) = +i\frac{g_N^s g_e^p}{4\pi}\frac{e^{-m_a|\boldsymbol{r}-\boldsymbol{R}|}}{|\boldsymbol{r}-\boldsymbol{R}|}\gamma_0\gamma_5 \qquad V_{ee}(\boldsymbol{r}_1,\boldsymbol{r}_2) = +i\frac{g_e^s g_e^p}{4\pi}\frac{e^{-m_a|\boldsymbol{r}_1-\boldsymbol{r}_2|}}{|\boldsymbol{r}_1-\boldsymbol{r}_2|}\gamma_0\gamma_5$$

$$W_{\mathrm{ax}}^{(eN)}(m_a) = \frac{1}{\Omega}\frac{1}{g_N^s g_e^p}\langle\Psi|\sum_{i=1}^{N_e} V_{eN}(\boldsymbol{r}_i)|\Psi\rangle \qquad W_{\mathrm{ax}}^{(ee)}(m_a) = \frac{1}{\Omega}\frac{1}{g_e^s g_e^p}\langle\Psi|\sum\nolimits'^{N_e}_{i,j=1} V_{ee}(\boldsymbol{r}_i,\boldsymbol{r}_j)|\Psi\rangle$$

$$\boxed{\delta E} = \bar{g}_N^s g_e^p \Omega W_{\mathrm{ax}}^{(eN)}(m_a) \qquad \boxed{\delta E} = g_e^s g_e^p \Omega W_{\mathrm{ax}}^{(ee)}(m_a)$$

Stadnik, Y. V., Dzuba, V. A., Flambaum, V. V. (2018), Phys. rev. lett., 120(1), 013202.

Standard Coulumb interaction integrals

$$\langle ab|\frac{1}{r_{12}}|cd\rangle \rightarrow F_m(T) = \int_0^1 dt t^{2m} e^{-Tt^2}$$

a, b, c, d are primitive Gaussian-type basis functions $x^n y^m z^k e^{-\beta r^2}$

Yukawa interaction integrals

$$\langle ab|\frac{e^{-m_a r_{12}}}{r_{12}}|cd\rangle \rightarrow G_m(T, U) = \int_0^1 dt t^{2m} e^{-Tt^2+U(1-\frac{1}{t^2})}$$

Implemented in LIBINT library!

Default approach

- $0 \leq T \leq 2^{10}$ and $10^{-7} \leq U \leq 10^{3} \rightarrow$ Gaussian quadrature

  precalculated values of the Chebyshev expansion coefficients of the positions and weights of the grid points are used

- $T > 2^{10}$ or $U < 10^{-7} \rightarrow$ upward recursive relations

$$G_{-1} = \frac{e^{-T}}{4}\sqrt{\frac{\pi}{U}}\left[e^{k^2}\mathrm{erfc}(k) + e^{\lambda^2}\mathrm{erfc}(\lambda)\right],$$

$$G_0 = \frac{e^{-T}}{4}\sqrt{\frac{\pi}{T}}\left[e^{k^2}\mathrm{erfc}(k) - e^{\lambda^2}\mathrm{erfc}(\lambda)\right],$$

where $k = -\sqrt{T} + \sqrt{U}$, $\lambda = \sqrt{T} + \sqrt{U}$.

All the remaining G_m values are obtained using the recurrence relations

$$G_m = \frac{1}{2T}[(2m-1)G_{m-1} + 2UG_{m-2} - e^{-T}].$$

Modifications

- $T = 0$ and $U < 10^{-7}$

  The first element of relations is

  $$G_0 = 1 - e^U \sqrt{\pi U}\text{erfc}(\sqrt{U}),$$

  All the remaining G_m values are obtained using the recurrence relations

  $$G_m(0, U) = \frac{1}{2m+1}[1 - 2UG_{m-1}(0, U)].$$

- $T < 0.1$ and $U < 10^{-7}$

  $$G_m(T, U) = \sum_{k=0}^{\infty} \frac{(-T)^k}{k!} G_{m+k}(0, U),$$

Calculated $W_{\rm ax}^{(ee)}(m_a)$ and derived limits on $|g_e^s g_e^p|$ for BaF

m_a, eV	$W_{\rm ax}^{(ee)}(m_a)$, $m_e c/\hbar$ DHF	CCSD	CCSD(T) (Final)	$\lvert g_e^s g_e^p\rvert$ Limit, $\hbar c$
1	$+6.3 \cdot 10^{-6}$	$+8.1 \cdot 10^{-6}$	$+8.0 \cdot 10^{-6}$	$+1.6 \cdot 10^{-20}$
10	$+6.3 \cdot 10^{-6}$	$+8.1 \cdot 10^{-6}$	$+8.0 \cdot 10^{-6}$	$+1.6 \cdot 10^{-20}$
10^2	$+6.3 \cdot 10^{-6}$	$+8.1 \cdot 10^{-6}$	$+8.0 \cdot 10^{-6}$	$+1.6 \cdot 10^{-20}$
10^3	$+4.8 \cdot 10^{-6}$	$+6.6 \cdot 10^{-6}$	$+6.5 \cdot 10^{-6}$	$+1.9 \cdot 10^{-20}$
10^4	$+1.2 \cdot 10^{-6}$	$+2.0 \cdot 10^{-6}$	$+2.0 \cdot 10^{-6}$	$+6.5 \cdot 10^{-20}$
10^5	$+9.3 \cdot 10^{-8}$	$+1.4 \cdot 10^{-7}$	$+1.4 \cdot 10^{-7}$	$+9.2 \cdot 10^{-19}$
10^6	$-3.3 \cdot 10^{-9}$	$-5.2 \cdot 10^{-9}$	$-5.1 \cdot 10^{-9}$	$+2.5 \cdot 10^{-17}$
10^7	$-5.1 \cdot 10^{-11}$	$-8.0 \cdot 10^{-11}$	$-7.8 \cdot 10^{-11}$	$+1.6 \cdot 10^{-15}$
10^8	$-5.1 \cdot 10^{-13}$	$-8.0 \cdot 10^{-13}$	$-7.9 \cdot 10^{-13}$	$+1.6 \cdot 10^{-13}$
10^9	$-5.1 \cdot 10^{-15}$	$-8.0 \cdot 10^{-15}$	$-7.9 \cdot 10^{-15}$	$+1.6 \cdot 10^{-11}$
10^{10}	$-5.1 \cdot 10^{-17}$	$-8.1 \cdot 10^{-17}$	$-7.9 \cdot 10^{-17}$	$+1.6 \cdot 10^{-9}$

Calculated $W_{\text{ax}}^{(eN)}(m_a)$ and derived limits on $|\bar{g}_N^s g_e^p|$ for BaF

m_a, eV	$W_{\text{ax}}^{(eN)}(m_a)$, $m_e c/\hbar$ Point	Correction, %	$W_{\text{ax}}^{(eN)}(m_a)$, $m_e c/\hbar$ Finite	$\lvert\bar{g}_N^s g_e^p\rvert$ Limit, $\hbar c$
1	$+1.74 \cdot 10^{-5}$	0.0	$+1.74 \cdot 10^{-5}$	$+7.29 \cdot 10^{-21}$
10	$+1.74 \cdot 10^{-5}$	0.0	$+1.74 \cdot 10^{-5}$	$+7.29 \cdot 10^{-21}$
10^2	$+1.73 \cdot 10^{-5}$	0.0	$+1.73 \cdot 10^{-5}$	$+7.31 \cdot 10^{-21}$
10^3	$+1.45 \cdot 10^{-5}$	0.0	$+1.45 \cdot 10^{-5}$	$+8.71 \cdot 10^{-21}$
10^4	$+1.86 \cdot 10^{-6}$	0.0	$+1.86 \cdot 10^{-6}$	$+6.81 \cdot 10^{-21}$
10^5	$-1.07 \cdot 10^{-5}$	0.0	$-1.07 \cdot 10^{-5}$	$+1.18 \cdot 10^{-20}$
10^6	$-4.70 \cdot 10^{-6}$	0.0	$-4.70 \cdot 10^{-6}$	$+2.69 \cdot 10^{-20}$
10^7	$-1.36 \cdot 10^{-7}$	-0.4	$-1.36 \cdot 10^{-7}$	$+9.34 \cdot 10^{-19}$
10^8	$-1.99 \cdot 10^{-9}$	-4.0	$-1.91 \cdot 10^{-9}$	$+6.65 \cdot 10^{-17}$
10^9	$-2.09 \cdot 10^{-11}$	-5.6	$-1.97 \cdot 10^{-11}$	$+6.43 \cdot 10^{-15}$
10^{10}	$-2.09 \cdot 10^{-13}$	-5.6	$-1.97 \cdot 10^{-13}$	$+6.43 \cdot 10^{-13}$

Calculated $W_{\mathrm{ax}}^{(eN)}(m_a)$ and derived limits on $|\bar{g}_N^s g_e^p|$ for HfF^+

m_a, eV	$W_{\mathrm{ax}}^{(eN)}(m_a)$, $m_e c/\hbar$ Point	Correction, %	$W_{\mathrm{ax}}^{(eN)}(m_a)$, $m_e c/\hbar$ Finite	$\|\bar{g}_N^s g_e^p\|$ Limit, $\hbar c$
1	$+1.67 \cdot 10^{-5}$	0	$+1.67 \cdot 10^{-5}$	$1.11 \cdot 10^{-20}$
10	$+1.67 \cdot 10^{-5}$	0	$+1.67 \cdot 10^{-5}$	$1.11 \cdot 10^{-20}$
10^2	$+1.66 \cdot 10^{-5}$	0	$+1.66 \cdot 10^{-5}$	$1.11 \cdot 10^{-20}$
10^3	$+1.54 \cdot 10^{-5}$	0	$+1.54 \cdot 10^{-5}$	$1.19 \cdot 10^{-20}$
10^4	$+3.30 \cdot 10^{-6}$	0	$+3.31 \cdot 10^{-6}$	$5.24 \cdot 10^{-20}$
10^5	$-1.15 \cdot 10^{-5}$	0	$-1.15 \cdot 10^{-5}$	$1.67 \cdot 10^{-20}$
10^6	$-6.41 \cdot 10^{-6}$	0	$-6.41 \cdot 10^{-6}$	$2.97 \cdot 10^{-20}$
10^7	$-2.85 \cdot 10^{-7}$	-1	$-2.82 \cdot 10^{-7}$	$6.74 \cdot 10^{-19}$
10^8	$-5.30 \cdot 10^{-9}$	-9	$-4.81 \cdot 10^{-9}$	$3.95 \cdot 10^{-17}$
10^9	$-5.85 \cdot 10^{-11}$	-13	$-5.09 \cdot 10^{-11}$	$3.73 \cdot 10^{-15}$
10^{10}	$-5.87 \cdot 10^{-13}$	-13	$-5.10 \cdot 10^{-13}$	$3.73 \cdot 10^{-13}$

Limiting cases

Radius of Yukawa-type interaction

$R_{\rm Yu} = 1/m_a(\text{relativistic units}) = \hbar/m_a c$

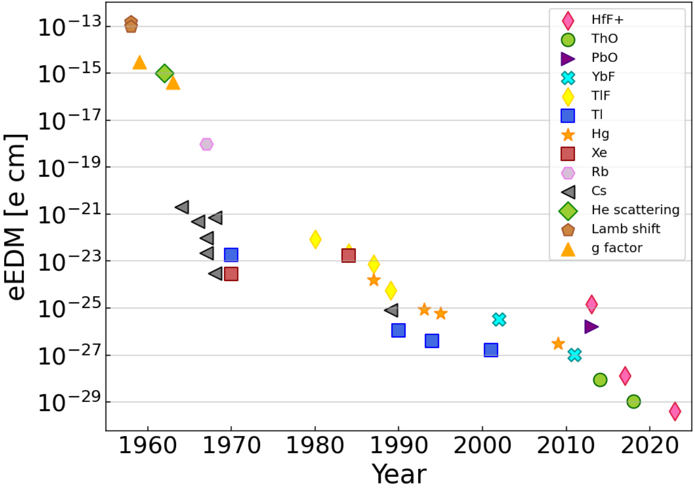
- Low-mass limit

  1 Bohr $\longleftrightarrow$ $m_a \approx 4$ keV $\quad\Rightarrow\quad \frac{e^{-m_a r}}{4\pi r} \approx \frac{1}{4\pi r} \quad\Rightarrow\quad W_{ax}(m_a) \approx constant$

- High-mass limit

  1 Fermi $\longleftrightarrow$ $m_a \approx 0.2$ GeV $\quad\Rightarrow\quad \frac{e^{-m_a r}}{4\pi r} \approx \frac{1}{m_a^2}\delta(\boldsymbol{r}) \quad\Rightarrow\quad W_{ax}(m_a) \approx \widetilde{W}_{\rm ax} m_a^{-2}.$

EDM limitation



https://en.wikipedia.org/wiki/Electron_electric_dipole_moment

Derived constraints for limiting cases

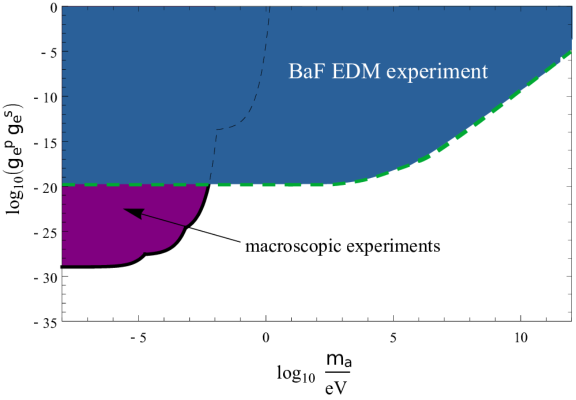
Limit	**BaF***	**HfF$^+$ ****	**ThO*****
$\lvert\bar{g}_N^s g_e^p\rvert/(\hbar c)$, $m_a \ll 1$ keV	7.3×10^{-21}	1.1×10^{-20}	9.0×10^{-20}
$\lvert g_e^s g_e^p\rvert/(\hbar c)$, $m_a \ll 1$ keV	1.6×10^{-20}	2.2×10^{-20}	2.4×10^{-19}
$\lvert\bar{g}_N^s g_e^p\rvert/(\hbar c m_a^2)$, $m_a \geq 1$ GeV	6.4×10^{-15} GeV^{-2}	3.7×10^{-15} GeV^{-2}	6.0×10^{-15} GeV^{-2}
$\lvert g_e^s g_e^p\rvert/(\hbar c m_a^2)$, $m_a \geq 1$ GeV	1.6×10^{-11} GeV^{-2}	1.7×10^{-11} GeV^{-2}	5.6×10^{-11} GeV^{-2}

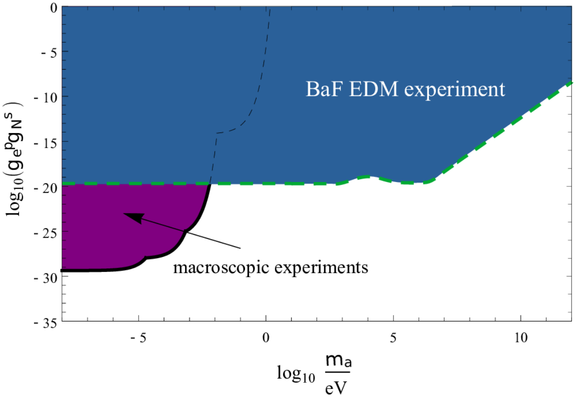
* Prosnyak, S. D., Skripnikov, L. V. (2024), Phys. Rev. A 109, 042821

** Prosnyak, S. D., Maison, D. E., Skripnikov, L. V. (2023), Symmetry, 15(5), 1043.

***Combining theoretical data from [Stadnik, Y. V., Dzuba, V. A., & Flambaum, V. V. (2018), Phys. rev. lett., 120(1), 013202; arXiv:1708.00486] and experimental data from [ACME Collaboration (2018), *Nature*, *562*(7727), 355-360.]

Current limitations





Thank you for attention!

Comparison with macroscopic experiments

Astrophysical experiments	for $m_a < 10^{-14}$ eV about 17 order of magnitude better, but can be spoiled by some mechanisms
QUAX-$g_p g_s$ experiment	$g_N^s g_e^p < 4.3 \times 10^{-30} \hbar c$ for the range of ALP masses $7 \times 10^{-7} \div 4 \times 10^{-6}$ eV, but for heavy axions many orders less stringent
Torsion pendulum Eöt–Wash experiment	better only for $m_a < 10^{-7}$ eV, but not for heavy axions
Magnetometry experiment	better only for $m_a < 10^{-6}$ eV, but not for heavy axions

EDM predictions

CORNERING THE AXION WITH *CP*-VIOLATING … PHYS. REV. D **102,** 115026 (2020)