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

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Table I. Electronic structure enhancement factors of $\mathcal{P},\mathcal{T}$-violation in Ra-containing closed-shell molecular ions computed at the level of DCHF/dyall.cv3z+sp(d) and DCKS/dyall.cv3z+sp(d) employing various exchange correlation functionals.

| Molecule | Functional | $W_\mathrm{d}^\mathrm{m}/\frac{10^{20}\,\mathrm{Hz}\,h}{e\,\mathrm{cm}}$ | $W_\mathrm{s}^\mathrm{m}/(h\,\mathrm{Hz})$ | $W_\mathrm{T}/(h\,\mathrm{kHz})$ | $W_\mathrm{p}/(h\,\mathrm{Hz})$ | $W_\mathrm{m}/\frac{10^{17}\,\mathrm{Hz}\,h}{e\,\mathrm{cm}}$ | $W_\mathcal{S}/\frac{\mathrm{MHz}\,h}{e\,\mathrm{fm}^3}$ |
|---|---|---|---|---|---|---|---|
| $RaSH^+$ | HF | 36.0 | 93.1 | −4.39 | −17.2 | −1.90 | −2.19 |
| | BHandH | 31.9 | 82.9 | −3.91 | −15.3 | −1.68 | −1.95 |
| | PBE0 | 30.0 | 78.2 | −3.70 | −14.5 | −1.58 | −1.84 |
| | B3LYP | 29.1 | 76.0 | −3.60 | −14.1 | −1.56 | −1.80 |
| | PBE | 27.0 | 71.2 | −3.38 | −13.3 | −1.45 | −1.69 |
| | LDA | 26.5 | 69.7 | −3.31 | −13.0 | −1.42 | −1.65 |
| $RaOCH_3^+$ | HF | 40.0 | 106 | −5.01 | −19.7 | −2.11 | −2.50 |
| | BHandH | 34.9 | 93.5 | −4.45 | −17.5 | −1.88 | −2.23 |
| | PBE0 | 32.8 | 88.1 | −4.20 | −16.5 | −1.76 | −2.11 |
| | B3LYP | 32.3 | 87.4 | −4.17 | −16.4 | −1.77 | −2.10 |
| | PBE | 30.1 | 81.7 | −3.90 | −15.3 | −1.64 | −1.97 |
| | LDA | 29.3 | 79.7 | −3.81 | −15.0 | −1.61 | −1.93 |
| $RaCH_3^+$ | HF | 45.4 | 113 | −5.25 | −20.6 | −2.17 | −2.54 |
| | BHandH | 39.0 | 98.5 | −4.62 | −18.1 | −1.91 | −2.24 |
| | PBE0 | 35.7 | 90.9 | −4.27 | −16.7 | −1.77 | −2.08 |
| | B3LYP | 34.5 | 88.5 | −4.17 | −16.3 | −1.74 | −2.03 |
| | PBE | 31.1 | 80.9 | −3.82 | −15.0 | −1.60 | −1.87 |
| | LDA | 30.7 | 80.1 | −3.79 | −14.9 | −1.59 | −1.85 |
| $RaCN^+$ | HF | 36.1 | 95.0 | −4.50 | −17.7 | −2.01 | −2.26 |
| | BHandH | 32.5 | 86.4 | −4.10 | −16.1 | −1.82 | −2.06 |
| | PBE0 | 31.2 | 82.8 | −3.93 | −15.4 | −1.73 | −1.97 |
| | B3LYP | 30.6 | 81.5 | −3.87 | −15.2 | −1.73 | −1.95 |
| | PBE | 28.9 | 77.2 | −3.67 | −14.4 | −1.62 | −1.85 |
| | LDA | 28.4 | 76.0 | −3.62 | −14.2 | −1.60 | −1.82 |
| $RaNC^+$ | HF | 35.6 | 94.9 | −4.51 | −17.7 | −2.01 | −2.28 |
| | BHandH | 32.0 | 86.1 | −4.10 | −16.1 | −1.82 | −2.08 |
| | PBE0 | 30.3 | 81.5 | −3.88 | −15.2 | −1.71 | −1.97 |
| | B3LYP | 30.1 | 81.3 | −3.87 | −15.2 | −1.73 | −1.97 |
| | PBE | 28.3 | 76.6 | −3.66 | −14.4 | −1.62 | −1.86 |
| | LDA | 27.7 | 75.1 | −3.58 | −14.1 | −1.59 | −1.82 |

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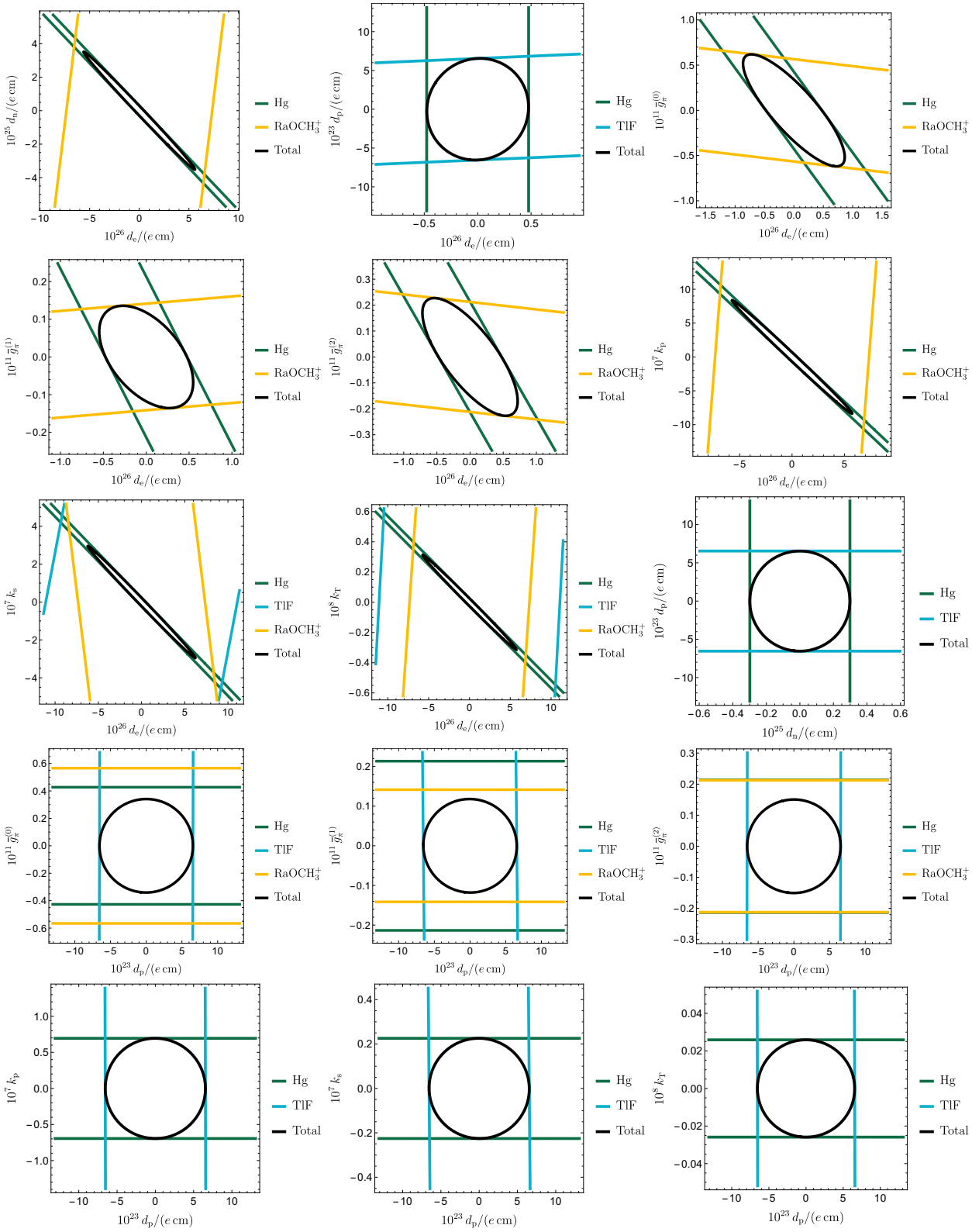


Figure 1. Restriction of two dimensional subspaces including parameters $d_\mathrm{e}$, $d_\mathrm{sr,p}$, $k_\mathrm{s}$ of the nine dimensional space of $P,T$-odd parameters by experiments with Hg, TlF and the proposed experiment with $\mathrm{RaOCH_3}^+$ excluding the pEDM. Coverage regions are computed with electronic structure parameters of $\mathrm{RaOCH_3}^+$ provided in this work, electronic structure parameters of TlF and Hg from Ref. [1] and nuclear structure parameters from Ref. [2]. Experimental uncertainty on the EDM of TlF $\sigma_d = 2.9 \times 10^{-23}\, e\,\mathrm{cm}$ with an external electric field for polarization of strength $\mathcal{E} = 16\,000\,\mathrm{V\,cm^{-1}}$ is taken from Ref. [3] and on the EDM of Hg $\sigma_d = 3.1 \times 10^{-30}\, e\,\mathrm{cm}$ is taken from Ref. [4]. The expected uncertainty of an experiment with a single $\mathrm{RaOCH_3}^+$ molecule $\delta\nu \approx 6.5 \times 10^{-5}\,\mathrm{Hz}$ is used as proposed in Ref. [5]. All bounds are computed with Gaussian probability distributions of 95 % CL as described in Ref. [1].

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