

\mathcal{P}, \mathcal{T} -Violating and Magnetic Hyperfine Interactions in Atomic Thallium

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(Dated: October 28, 2019)

We present state-of-the-art configuration interaction and coupled cluster calculations of the electron electric dipole moment, the nucleon-electron scalar-pseudoscalar, and the magnetic hyperfine interaction constants ($\alpha_{d_e}, \alpha_{C_S}, A_{||}$, respectively) for the thallium atomic ground state $^2P_{1/2}$. Our present best values are $\alpha_{d_e} = -559 \pm 28$, $\alpha_{C_S} = 6.77 \pm 0.34 [10^{-18} e \text{ cm}]$, and $A_{||} = 21172 \pm 1059$ [MHz]. These findings lead to a significant reduction of the theoretical uncertainties for \mathcal{P}, \mathcal{T} -odd interaction constants but not to stronger constraints on the electron electric dipole moment, d_e , or the nucleon-electron scalar-pseudoscalar coupling constant, C_S .

I. INTRODUCTION

Electric dipole moments (EDM) of elementary particles, atoms and molecules give rise to spatial parity (\mathcal{P}) and time-reversal (\mathcal{T}) violating interactions [1] and are a powerful probe for physics beyond the standard model (BSM) [2]. Current single-source limits [3–5] on the electron EDM, for instance, can probe New Physics (NP) up to an effective energy scale of 1000 TeV [6] (radiative stability approach) or even greater [7], surpassing the current sensitivity of the Large Hadron Collider for corresponding sources of NP.

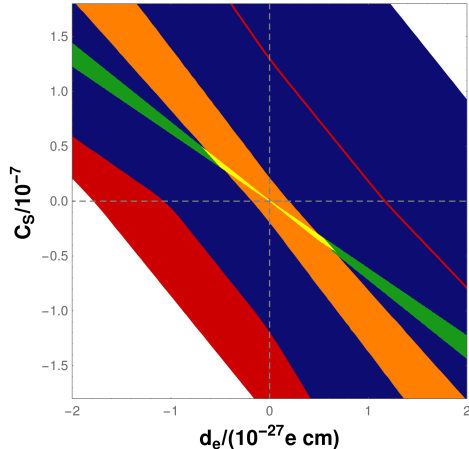
Until today no low-energy EDM experiment has delivered a positive result. However, the obtained EDM upper bounds are useful for constraining \mathcal{CP} -violating parameters [8] of BSM models, cast as effective field theories [6, 9] at different energy scales.

Open-shell atomic and molecular systems are particularly sensitive probes of leptonic and semi-leptonic

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\mathcal{CP} -violation [10].



In most BSM models [11] the dominant \mathcal{CP} -odd sources in open-shell systems are the electron EDM, d_e , and the nucleon-electron scalar-pseudoscalar (Ne-SPS) coupling, C_S . The panel [55] shows the constraints (yellow surface) on d_e and C_S using the combined information from measurements [3, 12–14] and calculations [4, 5, 15–23], including the associated experimental and theoretical uncertainties, on the open-shell systems ThO (green), YbF (red), HfF⁺ (orange) and Tl (blue) through a global fit in the d_e / C_S plane. Results from a single system do, therefore, not constrain d_e or C_S individually at all in this multiple-source interpretation [24], but lead to a fan-shaped surface of allowed combinations. The width of this surface is a function of the experimental and theoretical uncertainties.

This means that a substantial reduction of an uncertainty for an individual system could lead to more stringent constraints on the unknown \mathcal{CP} -violating parameters. The main reason for this is that the surfaces for different systems are not fully aligned, which is due to the different dependency of electron EDM and Ne-SPS atomic interactions on the electric charge of the respective heavy nuclei [25, 26].

A substantial part of the width of the surface for the Tl atom is due to the great spread of theoretical values for the electron EDM atomic enhancement, R , calculated in the past by various groups using different electronic-structure approaches [1, 19–23, 27]. Strikingly, Nataraj *et al.* [22] used a high-level many-body approach, the Coupled Cluster (CC) method, and produced a value for R that strongly disagrees with the results from all other groups, on the order of 20%.

The purpose of this paper is twofold:

1. We use state-of-the-art Configuration Interaction (CI) and Coupled-Cluster approaches for large-scale applications to determine the mentioned atomic interaction constants. We put particular emphasis on the electron EDM enhancement R and a conclusive resolution of the major discrepancy between literature values. Claims about physical effects that purportedly underlie these discrepancies are scrutinized.
2. We investigate whether a reduced uncertainty for $R(\text{Tl})$ impacts the above-described constraints on d_e and C_S .

The paper is structured as follows. In section II we lay out the theory underlying the atomic electron EDM, Ne-SPS, and magnetic hyperfine interactions constants. The following section III contains technical details about our calculations, results, and a discussion of these results in comparison with literature values. The final section IV concludes on our findings.

II. THEORY

An atomic electric dipole moment (EDM) is defined [28] (p. 16) as

$$d_a = - \lim_{E_{\text{ext}} \rightarrow 0} \left[\frac{\partial(\Delta\varepsilon)}{\partial E_{\text{ext}}} \right] \quad (1)$$

where $\Delta\varepsilon$ is a \mathcal{P}, \mathcal{T} -odd energy shift and E_{ext} is an external electric field. In atoms with nuclear spin $I \leq \frac{1}{2}$ [29] and in an electronic state with unpaired electrons, this energy shift is dominated by and originates from either the electron EDM, d_e , or a \mathcal{P}, \mathcal{T} -odd nucleon-electron (Ne) interaction, or a combination of the two [10, 11]. The two cases are presented separately.

A. Atomic EDM due to electron EDM

The Hamiltonian for the interaction of the electron electric dipole moment, d_e , is for an atomic system

$$H_{\text{EDM}} = - \sum_j \mathbf{d}_j \cdot \mathbf{E}(\mathbf{r}_j) = -d_e \sum_j \gamma_j^0 \boldsymbol{\Sigma}_j \cdot \mathbf{E}(\mathbf{r}_j) \quad (2)$$

where γ^0 is a Dirac matrix, $\boldsymbol{\Sigma} = \begin{pmatrix} \boldsymbol{\sigma} & \mathbf{0} \\ \mathbf{0} & \boldsymbol{\sigma} \end{pmatrix}$ is a vector of spin matrices in Dirac representation, j is an electron index, $\mathbf{E}(\mathbf{r}_j)$ the electric field at position \mathbf{r}_j and the bare fermion's electric dipole moment is expressed as $\mathbf{d} = d_e \gamma^0 \boldsymbol{\Sigma}$, necessarily linearly dependent on the particle's spin vector $\boldsymbol{\Sigma}$ [1, 30].

Supposing a non-zero electron EDM d_e , the resulting energy shift can be evaluated as

$$\Delta \varepsilon_{\text{EDM}} = d_e \left\langle - \sum_j \gamma_j^0 \boldsymbol{\Sigma}_j \cdot \mathbf{E}(\mathbf{r}_j) \right\rangle_{\psi(E_{\text{ext}})} \quad (3)$$

where $\psi(E_{\text{ext}})$ is the field-dependent atomic wavefunction of the state in question. The expectation value in Eq. (3) has the physical dimension of electric field and can be regarded as the mean interaction of each electron EDM with this field in the respective state. Following stratagem II of Lindroth *et al.* [31] the expectation value is recast in electronic momentum form as an effective one-body operator

$$\left\langle - \sum_j \gamma_j^0 \boldsymbol{\Sigma}_j \cdot \mathbf{E}(\mathbf{r}_j) \right\rangle_{\psi(E_{\text{ext}})} \approx \frac{2ic}{e\hbar} \left\langle \sum_j \gamma_j^0 \gamma_j^5 \mathbf{P}_j^2 \right\rangle_{\psi(E_{\text{ext}})} \quad (4)$$

where the approximation lies in assuming that ψ is an exact eigenfunction of the field-dependent Hamiltonian of the system. This momentum-form EDM operator has already been used as early as in 1986, by Johnson *et al.* [27]. In the present work the field-dependent Hamiltonian is the Dirac-Coulomb (DC) Hamiltonian (in *a.u.*, $e = m_e = \hbar = 1$)

$$\begin{aligned} \hat{H} &:= \hat{H}^{\text{Dirac-Coulomb}} + \hat{H}^{\text{Int-Dipole}} \\ &= \sum_j^n \left[c \boldsymbol{\alpha}_j \cdot \mathbf{p}_j + \beta_j c^2 - \frac{Z}{r_{jK}} \mathbb{1}_4 \right] + \sum_{k>j}^n \frac{1}{r_{jk}} \mathbb{1}_4 + \sum_j \mathbf{r}_j \cdot \mathbf{E}_{\text{ext}} \mathbb{1}_4 \end{aligned} \quad (5)$$

with \mathbf{E}_{ext} weak and homogeneous, the indices j, k run over n electrons, Z the proton number with the nucleus K placed at the origin, and $\boldsymbol{\alpha}$ are standard Dirac matrices. E_{ext} is not treated as a perturbation but included *a priori* in the variational optimization of the atomic wavefunction. Furthermore, the final results reported in this work include high excitation ranks in the correlation expansion of ψ . For these reasons, the approximation in Eq. (4) is considered very good in the present case.

Within the so-defined picture and using Eqs. (1), (3), and (4) the atomic EDM becomes

$$d_a = - \lim_{E_{\text{ext}} \rightarrow 0} \frac{\partial}{\partial E_{\text{ext}}} \frac{2ic d_e}{e\hbar} \left\langle \sum_j \gamma_j^0 \gamma_j^5 \mathbf{P}_j^2 \right\rangle_{\psi(E_{\text{ext}})} \quad (6)$$

The (dimensionless) atomic EDM enhancement factor is defined as $R := \frac{d_a}{d_e}$. Denoting $E_{\text{eff}} = \frac{2ic}{e\hbar} \left\langle \sum_j \gamma_j^0 \gamma_j^5 \mathbf{P}_j^2 \right\rangle_{\psi(E_{\text{ext}})}$ for the sake of simplicity, the enhancement factor is

$$R = - \lim_{E_{\text{ext}} \rightarrow 0} \left[\frac{\partial E_{\text{eff}}}{\partial E_{\text{ext}}} \right]. \quad (7)$$

The external field used in the experiment on Tl [14] was $E_{\text{ext}} = 1.23 \times 10^7 \left[\frac{\text{V}}{\text{m}} \right] \approx 0.2392 \times 10^{-4}$ a.u. In the present work $E_{\text{ext}} = 0.24 \times 10^{-4}$ a.u. is used. This is a very small field which is well within the linear regime considering the derivative in Eq. (7). The enhancement factor may under these circumstances be written as a function of two field points

$$R_{\text{lin}} = - \frac{\Delta E_{\text{eff}}}{\Delta E_{\text{ext}}} = - \frac{E_{\text{eff}}(2) - E_{\text{eff}}(1)}{E_{\text{ext}}(2) - E_{\text{ext}}(1)}. \quad (8)$$

We set $E_{\text{ext}}(1) := 0$ from which it follows that $E_{\text{eff}}(1) = 0$, and so

$$R \approx R_{\text{lin}} = -\frac{E_{\text{eff}}}{E_{\text{ext}}}. \quad (9)$$

E_{eff} is calculated as described in reference [32]. ψ is an approximate configuration interaction (CI) eigenfunction of the Dirac-Coulomb Hamiltonian including E_{ext} . Alternatively, E_{eff} can be calculated within the finite-field approach [33, 34]. The latter has been used in coupled cluster calculations.

The electron EDM enhancement factor R is in the particle physics literature often denoted as

$$\alpha_{d_e} := R, \quad (10)$$

the atomic-scale interaction constant of the electron EDM.

B. Nucleon-Electron Scalar-Pseudoscalar Interaction

The effective Hamiltonian for a \mathcal{P}, \mathcal{T} -odd nucleon-electron scalar-pseudoscalar interaction is written as [35]

$$H_{\text{Ne-SPS}} = i\frac{G_F}{\sqrt{2}} AC_S \sum_j \gamma_j^0 \gamma_j^5 \rho_N(\mathbf{r}_j) \quad (11)$$

and the resulting atomic energy shift is accordingly

$$\Delta\varepsilon_{\text{Ne-SPS}} = \frac{G_F}{\sqrt{2}} AC_S \left\langle i \sum_j \gamma_j^0 \gamma_j^5 \rho_N(\mathbf{r}_j) \right\rangle_{\Psi(E_{\text{ext}})}, \quad (12)$$

where A is the nucleon number, C_S is the S-PS nucleon-electron coupling constant, G_F is the Fermi constant[56] and $\rho_N(\mathbf{r}_j)$ is the nucleon density at the position of electron j . Note that in the present work we define $\gamma^5 := i\gamma^0\gamma^1\gamma^2\gamma^3$, whereas Flambaum and co-workers [21, 25, 26] define $\gamma^5 := -i\gamma^0\gamma^1\gamma^2\gamma^3$ which explains the sign difference between the present Ne-SPS atomic interaction constants and those of Flambaum and co-workers.

Next, we define (see also reference [36]) in analogy with Eq. (7) an Ne-SPS ratio [57]

$$S := \frac{d_a}{AC_S \frac{G_F}{\sqrt{2}}} \quad (13)$$

and so one can write, using Eq. (1),

$$S = -\lim_{E_{\text{ext}} \rightarrow 0} \left[\frac{\partial}{\partial E_{\text{ext}}} \left\langle i \sum_j \gamma_j^0 \gamma_j^5 \rho_N(\mathbf{r}_j) \right\rangle_{\Psi(E_{\text{ext}})} \right] \quad (14)$$

and in the linear regime

$$S = -\frac{\left\langle i \sum_j \gamma_j^0 \gamma_j^5 \rho_N(\mathbf{r}_j) \right\rangle_{\Psi(E_{\text{ext}})}}{E_{\text{ext}}}. \quad (15)$$

The initial implementation of this expectation value in the latter expression has been described in reference [37]. The independent implementation of the matrix elements of the Hamiltonian (11) has been developed in ref. [4].

For comparison with literature results we also define the S-PS nucleon-electron interaction constant

$$\alpha_{C_S} := \frac{d_a}{C_S} = S A \frac{G_F}{\sqrt{2}}. \quad (16)$$

C. Magnetic Hyperfine Interaction

Minimal substitution according to $\mathbf{p} \rightarrow \mathbf{p} - \frac{q}{c} \mathbf{A}$ in the Dirac equation and representing the vector potential in magnetic dipole approximation as $\mathbf{A}_D(\mathbf{r}) = \frac{\mathbf{m} \times \mathbf{r}}{r^3}$ with \mathbf{m} the nuclear magnetic dipole moment leads to the magnetic hyperfine Hamiltonian

$$\hat{H}_{\text{HF}} = c\boldsymbol{\alpha} \cdot \left(-\frac{q}{c} \frac{\mathbf{m} \times \mathbf{r}}{r^3} \right) = q \mathbf{m} \cdot \left(\frac{\boldsymbol{\alpha} \times \mathbf{r}}{r^3} \right)$$

for a single point charge q at position \mathbf{r} outside the finite nucleus. Given the nuclear magnetic dipole moment vector as $\mathbf{m} = \frac{\mu}{I} \mu_N \mathbf{I} = g_I \mu_N \mathbf{I}$ where μ is the magnetic moment in nuclear magnetons (μ_N), g_I is the nuclear g -factor, and \mathbf{I} is the nuclear spin, Eq. (17) for a single electron is written as

$$\hat{H}_{\text{HF}} = -e \frac{\mu}{I} \mu_N \mathbf{I} \cdot \left(\frac{\boldsymbol{\alpha} \times \mathbf{r}}{r^3} \right) \quad (17)$$

Based on Eq. (17) we now define the magnetic hyperfine interaction constant for n electrons in the field of nucleus K (in *a.u.*)

$$A_{||}(K) = -\frac{\mu_K [\mu_N]}{2cI m_p M_J} \langle \Psi_{J, M_J} | \sum_{i=1}^n \left(\frac{\boldsymbol{\alpha}_i \times \mathbf{r}_{iK}}{r_{iK}^3} \right)_z | \Psi_{J, M_J} \rangle \quad (18)$$

where $\frac{1}{2cm_p}$ is the nuclear magneton in *a.u.* and m_p is the proton rest mass. The term $\frac{1}{M_J}$ in the prefactor of Eq. (18) is explained as follows.

The vector operator $\left(\frac{\boldsymbol{\alpha}_i \times \mathbf{r}_{iK}}{r_{iK}^3} \right)_z$ can be regarded as the $q = 0$ component of a rank $k = 1$ irreducible tensor operator $\hat{T}_q^{(k)}$. Application of the Wigner-Eckart Theorem to the diagonal matrix element in Eq. (18) yields

$$\langle \alpha, J, M_J | \hat{T}_0^{(1)} | \alpha, J, M_J \rangle = \langle J, M_J; 1, 0 | J, 1; J, M_J \rangle \frac{\langle \alpha, J | \hat{T}^{(1)} | \alpha, J \rangle}{\sqrt{2J+1}}$$

where the Clebsch-Gordan coefficient is – using the general definition in Ref. [38], p. 27 – evaluated as

$$\langle J, M_J; 1, 0 | J, 1; J, M_J \rangle = M_J \frac{1}{\sqrt{J(J+1)}}, \quad (19)$$

which depends linearly on the total electronic angular momentum projection quantum number M_J . However, the magnetic hyperfine energy has to be independent of M_J which is assured by the above prefactor $\frac{1}{M_J}$. Magnetic hyperfine interaction matrix elements have been calculated based on the implementations in references [4, 39] which do not make direct use of the Wigner-Eckart theorem and reduced matrix elements.

III. RESULTS AND DISCUSSION

A. Technical Details

Gaussian atomic basis sets of double-, triple-, and quadruple- ζ quality [40–42] (including correlating functions for $4f$ and $5d$ shells in the case of CI and cvDZ/CC) [43] have been used in the present work.

The atomic spinor basis is obtained in Dirac-Coulomb (DC) Hartree-Fock (HF) approximation where the Fock operator is defined by averaging over $6p_{j=1/2}^1$ and $6p_{j=3/2}^1$ open-shell electronic configurations.

A locally modified version of the DIRAC program package [44] has been used for all electronic-structure calculations. Interelectron correlation effects are taken into account through Configuration Interaction (CI) theory as implemented in the KRCI module [45] of DIRAC. Coupled cluster (CC) calculations have been carried out within the MRCC code [46–48].

The nomenclature for both CI and CC models is defined as: S, D, T, etc. denotes Singles, Doubles, Triples etc. replacements with respect to the reference DCHF determinant. The following number is the

number of correlated electrons and encodes which occupied shells are included in the CI or CC expansion. In detail we have $3 \hat{=} (6s, 6p)$, $13 \hat{=} (5d, 6s, 6p)$, $21 \hat{=} (5s, 5p, 5d, 6s, 6p)$, $29 \hat{=} (4s, 4p, 5s, 5p, 5d, 6s, 6p)$, $31 \hat{=} (4d, 5s, 5p, 5d, 6s, 6p)$, $35 \hat{=} (4f, 5s, 5p, 5d, 6s, 6p)$. $81 \hat{=} (1s, 2s, 2p, 3s, 3p, 3d, 4f, 5s, 5p, 5d, 6s, 6p)$. The notation type S10_SD13, as an example, means that the model SD13 has been approximated by omitting Double excitations from the ($5d$) shells. CAS3in4 means that an active space is used with all possible determinant occupations distributing the 3 valence electrons over the 4 valence Kramers pairs.

We use the experimental value [49] for the nuclear magnetic moment of ^{205}Tl with nuclear spin $I = \frac{1}{2}$, $\mu = 1.63821[\mu_N]$, in calculations of the magnetic hyperfine interaction constant.

B. Results for Atomic Interaction Constants

The results from the systematic study of many-body effects on atomic EDM enhancement (R), Ne-SPS interaction ratio (S) and magnetic hyperfine interaction constant (A) are compiled in Table I. The general strategy is to first qualitatively investigate the relative importance of various many-body effects on the properties using a rather small atomic basis set. Then, in a second step, accurate models are developed that include all important many-body effects using the insight from the first step and larger atomic basis sets. Since EDM enhancement and Ne-SPS interaction ratio are analytically related [25, 26] it is sufficient to discuss the trends for R only.

C. Step 1: Many-Body effects in cvDZ basis

1. Valence electron correlation

The result of $R = -388$ for CAS1in3 which is a singles CI expansion for the electronic ground state can be regarded as close to a DC Hartree-Fock result. The Full-CI (FCI) result including only the three valence electrons (CAS3in4_SDT3/60au) of $R = -487$ shows that valence correlation effects lead to a considerable change by more than 25% (in the large cvQZ basis by more than 35%). The valence FCI enhancement in cvQZ basis of $R = -587$ is, therefore, a benchmark. This value is closely reproduced using the universal basis set of reference [22]. Further effects can be considered as modifications of this benchmark result and will be studied one by one.

2. Subvalence electron correlation

Subvalence electrons of the Tl atoms are those occupying the $5s$, $5p$, and $5d$ shells. All other electrons will be considered core electrons. Correlations among the $5d$ electrons and in particular of the $5d$ and the valence electrons lead to a strong decrease of R , on the absolute, on the order of 10%. Corresponding contributions from the $5s$ and $5p$ electrons are significantly smaller.

3. Outer-core electron correlation

Outer-core-valence correlations have been evaluated by allowing for one hole in the respective outer core spinors along with excitations from the subvalence and valence electrons. In sum for the shells with effective principal quantum number $n = 4$ these effects amount to about 1.5%.

4. Effect of higher excitation ranks

Allowing for three holes in the shells with effective principal quantum number $n = 5$ and up to four particles in the virtual spinors (*i.e.*, adding combined quadruple excitations) leads to a total change of around 3.5%.

TABLE I: R, S, and A for Tl atom. By default, calculations were performed using DCHF spinors for the neutral Tl atom (V^N potential) and, for comparison in selected cases, with the Tl^+ cation (V^{N-1} potential) and Tl^{3+} cation (V^{N-3} potential) spinors.

Model/virtual cutoff	R	S [a.u.]	$A_{ }({}^{205}\text{Tl})$ [MHz]
Dyall cvDZ			
CAS1in3	-388	269	18800
CAS3in4	-415	288	18800
CAS3in4_SD3/60au	-487	339	19092
CAS3in4_SDT3/60au	-487	339	19103
S10_CAS3in4_SD13/10au	-458	321	20003
SD10_CAS3in4_SD13/10au	-442	309	19502
SD10_CAS3in4_SD13/30au	-441	309	19575
SD10_CAS3in4_SDT13/10au	-465	326	19357
SD10_CAS3in4_SDTQ13/10au	-464	326	19345
SDT10_CAS3in4_SDT13/10au	-460	323	19254
SDT10_CAS3in4_SDTQ13/10au	-460	323	19341
SD18_CAS3in4_SD21/10au	-437	307	19445
SD18_CAS3in4_SD21/10au(Tl^+)	-428	300	18934
S8_SD18_CAS3in4_SD29/10au	-438	308	19536
SD18_CAS3in4_SD21/30au	-443	311	19758
SD18_CAS3in4_SD21/60au	-443	311	19759
SD8_SD18_CAS3in4_SD29/30au	-449	315	19980
SD18_CAS3in4_SDT21/10au	-473	331	19439
SD18_CAS3in4_SDT21/10au(Tl^+)	-467	328	19228
SDT18_CAS3in4_SDT21/10au	-461	325	19274
SD18_CAS3in4_SDT21/30au	-483	338	19761
SD18_CAS3in4_SDT21/60au	-483	338	19763
S10_SD18_CAS3in4_SDT31/10au	-469	329	19423
S14_SD18_CAS3in4_SDT35/10au	-469	330	19448
S8_SD18_CAS3in4_SDT29/30au	-484	340	19999
SD8_SD10_CAS3in4_SDT21/10au	-471	331	
SD18_CAS3in4_SDTQ21/10au	-469	329	19395
Dyall cvTZ			
CAS3in4	-460	323	
CAS3in4_SD3/10au	-565	397	19027
CAS3in4_SD3/50au	-565	397	19041
CAS3in4_SDT3/50au	-566	398	19050
SD18_CAS3in4_SD21/10au	-481	340	19619
SD18_CAS3in4_SD21/30au	-484	342	19751
SD18_CAS3in4_SDT21/10au	-542	383	19995
SD18_CAS3in4_SDT21/10au(Tl^{3+})	-524	371	
SD18_CAS3in4_SDT21/20au	-541	383	
Dyall cvQZ			
CAS1in3	-429	301	18806
CAS3in4	-476	334	18806
CAS3in4_SD3/10au	-587	412	19023
CAS3in4_SD3/35au	-587	412	19050
CAS3in4_SDT3/35au	-587	413	19060
SD18_CAS3in4_SD21/35au	-459	322	17442
SD18_CAS3in4_SDT21/10au	-555	391	20432
SD18_CAS3in4_SDT21/35au	-562	397	20592
cvQZ/SD18_CAS3in4_SDT21/35au + Δ_{corr}	-539	388	20614

TABLE II: R, S, and A for Tl atom calculated within the 81e-CCSD(T) method in different basis sets. In the case denoted “ V^N ” the atomic spinors are obtained for the neutral Tl atom and the external field perturbs both the spinor coefficients and the CC amplitudes. In the case denoted “ V^{N-1} ” the atomic spinors are obtained for the Tl^+ cation and the external electric field only perturbs the CC amplitudes but not the atomic spinors.

Basis set/virtual cutoff	R	S [a.u.]	$A_{ }(^{205}Tl)$ [MHz]
Nataraj universal/ 10^3 au (V^N)	-559	397	21087
Nataraj universal/ 10^3 au (V^{N-1})	-550	390	21071
Dyall cvDZ/ 10^4 au (V^N)	-493	347	20626
Dyall cvTZ/ 10^4 au (V^N)	-545	387	20760
Dyall cvQZ/ 10^4 au (V^N)	-558	397	21172

TABLE III: R for Tl atom calculated within the CCSD(T) method in Dyall’s cvQZ basis set.

Method/virtual cutoff	R
3e-CCSD(T)/10au	-589
21e-CCSD(T)/150au	-527
53e-CCSD(T)/150au	-542
81e-CCSD(T)/ 10^4 au	-558

D. Step 2: Accurate CI results

Subsets of important CI models based on the findings of the previous subsection have been repeated using the larger atomic basis sets, cvTZ and cvQZ. The single best values from these calculations are given by the model SD18.CAS3in4.SDT21/35au. These latter values V are then corrected by a “correction shift”, calculated as follows:

$$\begin{aligned} \Delta_{\text{corr}} := & V(\text{S10_SD18_CAS3in4_SDT31}/10\text{au}) - V(\text{SD18_CAS3in4_SDT21}/10\text{au}) \\ & + V(\text{S14_SD18_CAS3in4_SDT35}/10\text{au}) - V(\text{SD18_CAS3in4_SDT21}/10\text{au}) \\ & + V(\text{S8_SD18_CAS3in4_SDT29}/30\text{au}) - V(\text{SD18_CAS3in4_SDT21}/30\text{au}) \\ & + V(\text{SDT18_CAS3in4_SDT21}/10\text{au}) - V(\text{SD18_CAS3in4_SDT21}/10\text{au}) \\ & + V(\text{SD18_CAS3in4_SDTQ21}/10\text{au}) - V(\text{SD18_CAS3in4_SDT21}/10\text{au}) \end{aligned}$$

The final best CI values are obtained by adding the above sum of individual corrections to the value from the model SD18.CAS3in4.SDT21/35au.

E. Accurate CC results

Table II gives values of R, S and $A_{||}(^{205}Tl)$ constants obtained within the all-electron coupled cluster with single, double and non-iterative triple cluster amplitudes, CCSD(T), method employing several basis sets. One can see a good convergence of the results in the series of the Dyall’s DZ, TZ and QZ basis sets: values of R obtained within the QZ and TZ basis sets differ by about 2%. Table II also gives values of the constants obtained within the Nataraj’s universal basis set [22]. Note that the latter basis set is the even-tempered basis set (geometry progression). One can see a good agreement of the results obtained within the QZ basis set and Nataraj’s universal basis set.

Table III gives values of R calculated with different number of correlated electrons. As can be seen contributions from subvalence and outer-core electrons are close to those obtained within the CI approach above.

To check the convergence with respect to electron correlation effects we performed a series of successive 21-electron coupled cluster calculations within the TZ basis set (see Table IV). In these calculations two sets of atomic bispinors were used. The first one was obtained within the DCHF approximation where the Fock operator is defined by averaging over $6p_{j=1/2}^1$ and $6p_{j=3/2}^1$ open-shell electronic configurations as in the CI case above. The second one was obtained within the closed-shell DCHF method for the Tl^+ cation. One can see that CC values gives almost identical result for each set at any level. Besides, the

TABLE IV: Values of R calculated at different level of theory with correlation of 21 electrons of Tl, cvTZ basis set. Calculations were performed using DCHF spinors for the neutral Tl atom (V^N potential) and for the Tl⁺ cation (V^{N-1} potential) cases. In both cases the external field perturbs both the spinor coefficients and the CC amplitudes.

	V^N	V^{N-1}
DCHF	-418	-402
CCSD	-531	-530
CCSD(T)	-521	-522
CCSDT	-523	-523
CCSDT(Q)	-522	-522

TABLE V: Comparison with literature values

Work	α_{de}	$\alpha_{CS} [10^{-18} e \text{ cm}]$	$A_{ } (^{205}\text{Tl}) [\text{MHz}]$
Literature values			
Khriplovich <i>et al.</i> [1]		5.1	
Johnson <i>et al.</i> [27] Norcross potential	-562		-18764
Mårtensson-Pendrill <i>et al.</i> [19]	-600 ± 200	7 ± 2	
Liu <i>et al.</i> [20]	-585		
Dzuba <i>et al.</i> [21]	-582	7.0 ± 0.2	21067
Nataraj <i>et al.</i> (CCSD(T)) [22]	-470		21053
Sahoo <i>et al.</i> (CCSD(T)) [50]		4.06	21026
Porsev <i>et al.</i> [23]	-573		22041
This work CI	-539	6.61	20614
This work CC	-559	6.77	21172
Experiment [51]			21310.8 ± 0.0

contribution of correlation effects beyond the CCSD(T) model is almost negligible in the considered case. We considered models up to coupled cluster with Single, Double, Triple and perturbative Quadruple cluster amplitudes, CCSDT(Q).

Contribution of the effect of the Breit interaction on R has been estimated in reference [23] as 0.36%. Based on the uncertainties discussed above we conservatively estimate the uncertainty of our final CC value for R to be less than 5%.

F. Discussion in Comparison with Literature Results

Our present best results are shown in Table V in comparison with previous work. The earlier controversy between different groups over results for $R(\text{Tl})$ can be condensed into three main points which we address one by one.

1. Basis sets

From the results in Tables I and II it is evident that a large atomic basis set, at least of quadruple-zeta quality, must be used for obtaining very accurate interaction constants. The results in VI and II obtained with our correlation methods demonstrate that the basis set used by Nataraj *et al.* in ref. [22] fulfills this requirement, yielding interaction constants that are very close to those obtained with Dyal's cvQZ basis set and the same correlation expansion. The earlier suggestion of Porsev *et al.* about an inadequate basis set used in ref. [22] can, therefore, be excluded as a possible reason for the outlier result in ref. [22].

TABLE VI: R, S, and A for Tl atom

Model/virtual cutoff	R	S [a.u.]	$A_{ }({}^{205}\text{Tl})$ [MHz]
Nataraj universal			
CAS3in4	-483	339	18800
CAS3in4_SD3/6au	-597	419	19035
CAS3in4_SD3/20au	-523	367	19174
CAS3in4_SD3/45au	-595	419	19060
CAS3in4_SD3/Nat100	-595	418	19060
CAS3in4_SDT3/45au	-596	419	19069
CAS3in4_SD3/130au	-595	418	19060
CAS3in4_SD3/200au	-595	418	19060
SD18_CAS3in4_SD21/45au	-510	361	19864

2. Treatment of correlation effects by the many-body method

It is claimed in reference [22] that the treatment of electron correlation effects were more complete than in references [20] and [21]. We have therefore first attempted to reproduce the electron EDM enhancement calculated by Nataraj *et al.* by using the same many-body Hamiltonian and EDM operator, the same atomic basis set (“Nataraj universal”) and the the same method, CCSD(T). A persisting difference with the approach of Nataraj *et al.* is the use of CC amplitudes for the closed shells of neutral Tl (our case) or the closed shells of the singly-ionized Tl^+ (Nataraj case). These results are shown in Table II under the label “ V^{N-1} ”. Our calculation of the hyperfine constant $A_{||} = 21071$ [MHz] reproduces the value of Nataraj *et al.* which is $A_{||} = 21053$ [MHz] almost precisely (residual difference of less than 0.1%). However, using the same wavefunction we obtain $R = -550$ which differs from the value of Nataraj *et al.* by 17%. Our CC result for R is in accord with similar calculations using the large cvQZ basis set, in accord with the present best CI result ($R = -539$ which after correction for core correlations from the innermost 28 electrons, according to the results in Table III, becomes $R = -555$) and in good agreement with the best results of Liu *et al.* [20], Dzuba *et al.* [21], and Porsev *et al.* [23], see Table V. The correct evaluation of the electron EDM enhancement in our codes has been assured by comparative tests of the independent implementations of present CI and CC, as well as with the DIRRCI module [32, 52] in the DIRAC program package. All three independent implementations produce the same values of R for small test cases using Full CI / Full CC expansions. These findings strongly suggest that the CC wavefunctions used by us and by Nataraj *et al.* are almost identical, but that the evaluation of R in reference [22] is flawed.

Since correlation effects have been treated at a very similar (but physically more accurate) level in the present work as in ref. [22] and the result is very different, the claim of correlation effects being responsible for the large difference between previous results is untenable.

3. Use of V^N , V^{N-1} , and V^{N-3} potentials

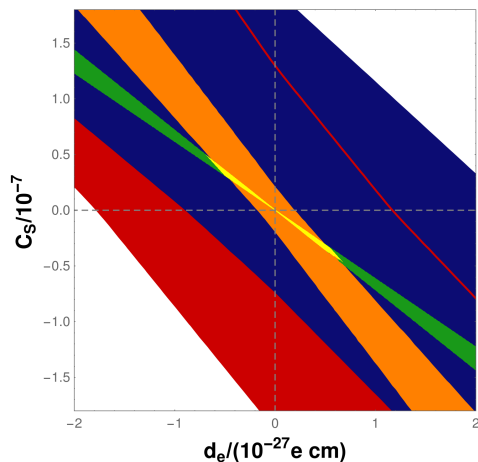
First, given a fixed atomic basis set and a fixed many-body Hamiltonian [58], the Full CI expansion delivers the exact solution in the N -particle sector of Fock space [53], *independent of the orbital/spinor basis used for this Full CI expansion*. This implies that a many-body expansion that closely approximates the Full CI expansion, such as CCSDT or CCSDT(Q), must also be nearly independent of the employed Dirac-Fock potential.

Our results in Table IV clearly confirm this conjecture and demonstrate that even in the more approximate CCSD expansion the electron EDM enhancement factor R is almost independent (0.2% difference) of the underlying spinor set. As the many-body expansion becomes more approximate, such as in the CI model SD18_CAS3in4_SD21 (see Table I) basic theory leads us to expect that the difference in R should increase which is indeed the case (roughly 2% difference). Adding external Triple excitations to the CI expansion, model SD18_CAS3in4_SDT21, quenches the difference to a mere 1.2%, again in accord with expectation. Even the use of a V^{N-3} potential (*i.e.*, spinors optimized for the Tl^{3+} system) changes R by only about 3% relative to spinors for the neutral atom in the SD18_CAS3in4_SDT21 model. This difference is expected to be even smaller in CC models.

Despite of the unimportance of the employed spinor set in highly-correlated calculations, we have used the physically most accurate spinors for the neutral Tl atom in obtaining our best final results. The ratios of our calculated \mathcal{P}, \mathcal{T} -odd interaction constants are $\left| \frac{\alpha_{d_e}}{\alpha_{C_S}} \right|(\text{CI}) = 81.5 \frac{1}{10^{-18}[\text{ecm}]}$ and $\left| \frac{\alpha_{d_e}}{\alpha_{C_S}} \right|(\text{CC}) = 82.6 \frac{1}{10^{-18}[\text{ecm}]}$ which agree well with the analytical value of Dzuba *et al.* [25, 26] of $\left| \frac{\alpha_{d_e}}{\alpha_{C_S}} \right|(\text{an.}) = 89 \frac{1}{10^{-18}[\text{ecm}]}$.

IV. CONCLUSIONS

We conclude from our findings that the result of Nataraj *et al.* in ref. [22] is unreliable and should be excluded from the dataset used to constrain the \mathcal{CP} -odd parameters d_e and C_S . Likewise, the result by Sahoo *et al.* [50] (see Table V) – presumably obtained with a similar code as R(Tl) by Nataraj *et al.* – is also significantly too small and should be excluded from the reliable dataset. A further proof of this conclusion is the ratio obtained using the values from refs. [22] and [50] which amounts to $\left| \frac{\alpha_{d_e}}{\alpha_{C_S}} \right|(\text{CC Nataraj/Sahoo}) = 115.8 \frac{1}{10^{-18}[\text{ecm}]}$ which deviates from the analytical ratio [25, 26] by 30%.



The panel is the updated version of the one shown in the introduction, using the dataset of reliable calculations of α_{d_e} and α_{C_S} for the Tl atom. The strongly reduced uncertainty of atomic interaction constants for Tl leads to a discernable shrinking of the associated parameter surface (blue), but does not lead to modified constraints. The essential reason for this is the extremely high sensitivity of the experiments on ThO (green) and HfF⁺ (orange). However, tighter constraints on d_e and C_S can be obtained by including experimental and theoretical results for closed-shell atomic systems as discussed in ref. [54].

V. ACKNOWLEDGEMENTS

We thank Martin Jung (Torino) for providing updated plots and for helpful discussions. Huliya Nataraj is thanked for sharing many technical details of his calculations with us. Electronic structure calculations were partially carried out using resources of the collective usage centre Modeling and predicting properties of materials at NRC “Kurchatov Institute” – PNPI. L.S. was supported by the Russian Science Foundation Grant No. 19-72-10019.

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