Reappraisal of the Electric Dipole Moment Enhancement Factor for Thallium

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The electric dipole moment (EDM) enhancement factor of atomic Tl is of considerable interest as it has been used in determining the most accurate limit on the electron EDM to date. However, its value varies from −179 to −1041 in different approximations. In view of the large uncertainties associated with many of these calculations, we perform an accurate calculation employing the relativistic coupled-cluster theory and obtain −466, which in combination with the most accurate measurement of Tl EDM [Phys. Rev. Lett. **88**, 071805 (2002)] yields a new limit for the electron EDM: $|d_e| < 2.0 \times 10^{-27} e$ cm.

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The EDM of a nondegenerate physical system arises from the simultaneous violations of parity ($\mathcal{P}$) and time-reversal ($\mathcal{T}$) symmetries [1,2]. The invariance of the latter symmetry is associated with the invariance of the $\mathcal{CP}$ (combined charge conjugation ($\mathcal{C}$) and $\mathcal{P}$ symmetries) symmetry on the basis of the $\mathcal{CPT}$ theorem [3]. Thus, EDMs of atoms can shed light on $\mathcal{CP}$ violations originating in the leptonic, semileptonic and hadronic sectors [4,5]. The knowledge of EDMs provide valuable insights into some profound questions such as the existence of new physics beyond the standard model (SM) and the matter-antimatter asymmetry in the universe [4–6]. Given the current interest in understanding different types of $\mathcal{CP}$ violations originating from the elementary particles both by accelerator and nonaccelerator based approaches, EDM searches are of invaluable significance.

The EDMs of paramagnetic atoms are sensitive to the EDM of the electron [7]. The most accurate limit for the latter has been obtained by combining the results of the Tl EDM measurement [8] and the EDM enhancement factor (EF) [9], defined as the ratio of the EDM of the atom to that of the electron, of this atom. Liu and Kelly have computed its value using a linearized version of the relativistic coupled-cluster (RCC) theory and report it to be −585, with an error bar of 5%–10% [9]. A more recent calculation of this quantity for Tl by Dzuba and Flambaum uses a hybrid approach combining the configuration interaction (CI) method and many-body perturbation theory (MBPT) and reports −582 with an estimated accuracy of 3% [10]. Both these results, coincidentally, are in good agreement with each other. Nevertheless, considering the fact that the values of the Tl EDM EF reported in the literature range from −179 to −1041 [11–13], there is clearly a need for high precision calculations of this quantity. Thus, the primary focus of this Letter is to determine the EDM EF of Tl by proceeding beyond [9,10]. An accurate treatment of the unusually strong electron correlation effects in the ground state Tl EDM EF warrants the use of an all-order relativistic many-body method like the RCC theory.

The open-shell RCC theory with single, double and a subset of leading triple excitations employed in the calculation of the EDM EFs is discussed in detail in [14–16]. However, we briefly present below the salient features of this method for the sake of completeness. The effective one electron form of the interaction Hamiltonian due to the electron EDM is given by [16],

$$H_{\mathrm{EDM}}^{\mathrm{eff}} = 2ic\sum_j \beta\gamma^5 \vec{p}_j^{\,2}, \tag{1}$$

where β and γ^5 are the usual Dirac matrices, $\vec{p}_j$ is the momentum vector of the electron j. As the strength of the EDM interaction is sufficiently weak, we consider the wave function expansion only up to first order in perturbation. Thus, the modified atomic wave function for a valence electron "v" is given by,

$$|\Psi_v'\rangle = |\Psi_v^{(0)}\rangle + d_e|\Psi_v^{(1)}\rangle. \tag{2}$$

In the RCC theory, the unperturbed and perturbed wave functions can be expressed as [14–16],

$$|\Psi_v^{(0)}\rangle = e^{T^{(0)}}\{1 + S_v^{(0)}\}|\Phi_v\rangle, \tag{3}$$

and

$$|\Psi_v^{(1)}\rangle = e^{T^{(0)}}\{T^{(1)}(1 + S_v^{(0)}) + S_v^{(1)}\}|\Phi_v\rangle, \tag{4}$$

where $|\Phi_v\rangle$ is the Dirac-Fock (DF) wave function obtained by appending the valence electron 'v' to the closed-shell ($[5d^{10}]6s^2$) reference state, $T^{(0)}$ and $S_v^{(0)}$ are the excitation operators for core and valence electrons in an unperturbed case, and $T^{(1)}$ and $S_v^{(1)}$ are their first-order corrections. The atomic wave functions are calculated using the Dirac-Coulomb Hamiltonian given by

$$H_0 = \sum_i \{c\alpha p_i + (\beta - 1)c^2 + V_n(r_i)\} + \sum_{i<j} \frac{1}{r_{ij}}, \tag{5}$$

where α and β are the Dirac matrices, $V_n(r_i)$ is the nuclear potential at the site of electron i and $1/r_{ij}$ is the Coulomb potential experienced by electron i due to electron j. We have used atomic units throughout this Letter unless otherwise specified explicitly.

We consider only the single and double excitation operators in the expansion of the RCC wave functions (termed as the CCSD approximation), by defining,

$$T = T_1 + T_2 \quad \text{and} \quad S_v = S_{1v} + S_{2v}, \tag{6}$$

for both the perturbed and unperturbed operators. Further, we construct triple excitation operators for $S_v^{(0)}$ [17] as,

$$S_{vab}^{pqr,(0)} = \frac{\widehat{H_0 T_2^{(0)}} + \widehat{H_0 S_{2v}^{(0)}}}{\epsilon_v + \epsilon_a + \epsilon_b - \epsilon_p - \epsilon_q - \epsilon_r}, \tag{7}$$

which are used to evaluate the CCSD amplitudes iteratively. This is referred to as CCSD(T) method. Here, ϵ_l is the single particle energy of an orbital l.

The final expression for the EDM EF ($\mathcal{R} = \frac{D_a}{d_e}$) in terms of the coupled-cluster operators is given by

$$\mathcal{R} = \frac{\langle \Phi_v | \{1 + S_v^{(0)\dagger}\} \overline{D^{(0)}} \{T^{(1)}(1 + S_v^{(0)}) + S_v^{(1)}\} | \Phi_v \rangle}{\langle \Phi_v | e^{T^{(0)\dagger}} e^{T^{(0)}} + S_v^{(0)\dagger} e^{T^{(0)\dagger}} e^{T^{(0)}} S_v^{(0)} | \Phi_v \rangle} + \text{Hermitian conjugate}, \tag{8}$$

where the dressed operator $\overline{D^{(0)}} = e^{T^{(0)\dagger}} D e^{T^{(0)}}$ and $D = r$ is the electric dipole moment operator due to the applied electric field. The procedure for the calculation of the above expression is discussed elsewhere [14–16].

In Table I, we present the contributions from different RCC terms along with the lowest order (DF) contributions to $\mathcal{R}$ and compare them with the results reported by Liu and Kelly [9]. It is evident from this table that the bulk of the contributions to $\mathcal{R}$ comes from the RCC terms $DT_1^{(1)}$, $DS_{1v}^{(1)}$ and $DS_{2v}^{(1)}$ (a dominant term in $DX_2^{(1)}$). These important all-order correlation effects involving the core, valence and core-valence sectors are shown diagrammatically in Fig. 1. The single largest contribution of about -378 to $\mathcal{R}$ comes from the all-order core correlation effect, $DT_1^{(1)}$. The magnitude of higher-order contributions is much larger than the DF contribution for this term. The all-order valence correlation contribution from $DS_{1v}^{(1)}$ is -314 while its DF contribution is -269. It is apparent from the table that the all-order core correlations are quite strong in the case of Tl. It is important to note that a class of core-polarization effects represented by the $DS_{2v}^{(1)}$ term has a significant positive contribution of 248. This dramatically reduces the final result. There is a non-negligible contribution from the other higher-order terms; however, many of them cancel each other to give an effective value of only about 13. The normalization of the RCC wave function also gives a contribution of 4 and the total result for the ground state EDM EF for Tl amounts to -466.

TABLE I. The contributions from various RCC terms to $\mathcal{R}$. A new quantity, $X = T + S_v$ is defined solely for comparing our results with those of [9].

RCC Term	This Work	Ref. [9]
$(DT_1^{(1)})_{\text{lowest-order}}$	−153.6	−153.2
$(DS_{1v}^{(1)})_{\text{lowest-order}}$	−268.5	−267.3
$(DT_1^{(1)})_{\text{higher-order}}$	−224.7	−342.1
$(DS_{1v}^{(1)})_{\text{higher-order}}$	− 45.5	−102.5
$DX_2^{(1)}$	248.0	240.9
$X_1^{(0)\dagger} DX_1^{(1)}$	22.5	22.4
$X_2^{(0)\dagger} DX_1^{(1)}$	− 78.2	49.3
$X_2^{(0)\dagger} DX_2^{(1)}$	21.5	− 36.9
$X_2^{(0)\dagger} DX_1^{(1)} X_2^{(0)}$	− 4.3	− 2.2
Higher-order RCC terms	13.0	−
Normalization contribution	3.8	6.5
Total EDM EF ($\mathcal{R}$)	−466.0	−585.1

The $H_{\text{EDM}}^{\text{eff}}$ being an odd-parity operator mixes the atomic states of opposite parities, however, with the same angular momentum. Therefore, we have investigated the role of various intermediate states of s symmetry. The RCC terms such as $DT_1^{(1)}$, $DS_{1v}^{(1)}$, $S_{1v}^{(0)\dagger} DS_{1v}^{(1)}$, and $S_{2v}^{(0)\dagger} DS_{1v}^{(1)}$, have got significant contributions from the orbitals, from $5s$ through $11s$ and their combined contributions are shown in Fig. 2. For comparison, we have also shown the DF contributions from these individual orbitals. The magnitude of correlation effects for the $6s$ orbital is much larger than its DF contribution.

We have also investigated the role of different doubly excited states that contribute to $\mathcal{R}$ through $DS_{2v}^{(1)}$. These contributions are given in Table II. As seen from the table, the largest contribution comes when the $6s$ orbital is excited to the valence orbital, $6p_{1/2}$ followed by the $6p_{3/2}$ orbital. There are also, however, some non-negligible contributions from the $5d$ orbitals when they are excited to the virtual $p_{3/2}$ and $f_{5/2,7/2}$ orbitals.

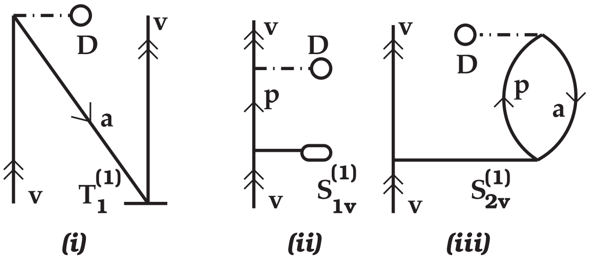

FIG. 1. The leading correlation diagrams: (i) $DT_1^{(1)}$, (ii) $DS_{1v}^{(1)}$, (iii) $DS_{2v}^{(1)}$. The exchange and Hermitian conjugate diagrams are not shown. Labels v, a, and p refer to valence, core, and virtual orbitals, respectively.

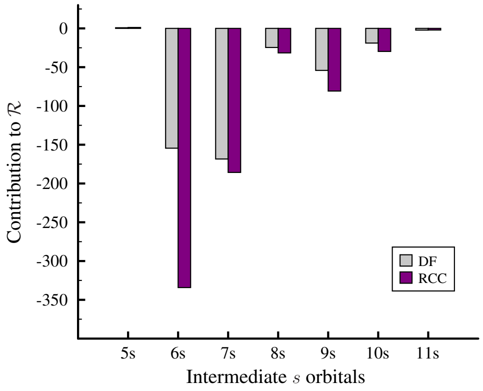

FIG. 2 (color online). The contributions from singly excited intermediate states, $5s$ through $11s$, in DF and RCC approximations are compared.

The EDM enhancement factors for Tl from different calculations are compared in Table III. It is quite apparent that the published results lie in a rather large range. We discuss below the qualitative differences between the current work and the two recent *ab initio* works [9,10]. Although, the overall trends of the majority of the correlation terms in [9] appear similar to ours, as shown in Table I, there is a significant difference in the magnitude of many of those correlation contributions which can be attributed to some of the following approximations made in the former work. (i) An approximation to only the one electron part of the EDM Hamiltonian is considered, thereby neglecting the important contributions partly from the DF potential and largely from the two electron Coulomb interaction. (ii) Only the linear terms and a few selected nonlinear terms have been used in the calculations. The CC equations have not been fully solved even at the CCSD level as a coupled-electron pair approximation has been used to solve for the quadratic terms that have been taken in the unperturbed doubles equation. (iii) A few selected triple excitations are included only in the unperturbed singles amplitude equations, where as, several dominant triples terms are completely ignored in the unperturbed doubles equations. Thus, the contribution of triples is taken into account in a nonstandard way. (iv) The inner core is frozen up to the $4s$ orbital for the calculation of the unperturbed amplitudes, where as, for solving the perturbed doubles equations, $4s$, $4p$ and $4d$ orbitals are further frozen. Such an inconsistent treatment introduces uncontrollable errors. In contrast to the above drawbacks, we consider all the nonlinear terms arising from the single and double excitations. In addition, we consider the leading triple excitations in both the unperturbed singles and doubles cluster equations. We solve the unperturbed and perturbed, closed- and open-shell equations, self-consistently in the framework of the relativistic CCSD(T) approach, taking into account the excitations from all the core electrons. The active orbitals considered for the correlation calculation are: $14s$, $13p_{1/2,3/2}$, $13d_{3/2,5/2}$, $9f_{5/2,7/2}$, and $8g_{7/2,9/2}$, where as, $38s$, $34p_{1/2,3/2}$, $34d_{3/2,5/2}$, $30f_{5/2,7/2}$, and $20g_{7/2,9/2}$ number of orbitals are considered for the SCF calculation. However, after the SCF step, we have truncated the virtual space by dropping the high-lying virtuals as their contributions are small.

The comparison between the results of Dzuba and Flambaum based on a combined CI + MBPT approach [10] and our all-order CCSD(T) is not straight forward. The important differences are that, in the former work, the following occurs. (i) The three outer most electrons ($6s^2 6p_{1/2}$) are treated as valence and the rest as core. (ii) The core, virtual, and valence orbitals are generated in a V^{N-3} closed-shell potential. The orbitals, therefore, are highly contracted. (iii) The valence-valence correlations are evaluated by CI, while the valence-core and the core-core correlations by MBPT. (iv) The net level of excitation of the configurations in their CI calculation is not higher than triples with reference to $6s^2 6p_{1/2}$. (v) It appears from the previous work of Dzuba and Flambaum that the $\mathcal{P}\&\mathcal{T}$ violating Hamiltonian used in [10], considers only the internal electric field due to the nucleus and not the electrons; i.e., the entire two body Coulomb potential is neglected. The major drawback of their work is that all the correlations obtained by MBPT are considered only up to second order (except for s electrons in the one-body correlation operator Σ_1) although the importance of valence-core correlations is stated in their paper. In contrast, we have evaluated the core-core and core-valence correlations

TABLE II. The contributions from the selected doubly excited states to $\mathcal{R}$ through the $DS_{2v}^{(1)}$ term.

Core(a)	Virtual(p)	Result	Core(a)	Virtual(p)	Result
$6s$	$6p_{1/2}$	170.88	$5d_{3/2}$	$8f_{5/2}$	0.92
$6s$	$7p_{1/2}$	3.61	$5d_{3/2}$	$9f_{5/2}$	2.06
$6s$	$8p_{1/2}$	2.01	$5d_{5/2}$	$6p_{3/2}$	5.37
$6s$	$9p_{1/2}$	4.50	$5d_{5/2}$	$7p_{3/2}$	0.76
$6s$	$6p_{3/2}$	31.42	$5d_{5/2}$	$8p_{3/2}$	0.59
$6s$	$7p_{3/2}$	3.00	$5d_{5/2}$	$9p_{3/2}$	1.46
$6s$	$8p_{3/2}$	1.95	$5d_{5/2}$	$8f_{7/2}$	2.34
$6s$	$9p_{3/2}$	5.26	$5d_{5/2}$	$9f_{7/2}$	5.36

TABLE III. Comparison of the ground state EDM EF ($\mathcal{R}$) of Tl from different calculations.

EDM EF ($\mathcal{R}$)	Method	Reference
−466(10)	CCSD(T)	This Work
−582(20)	CI + MBPT	Dzuba 2009 [10]
−585(30 − 60)	LCCSD(T)	Liu 1992 [9]
−179	MBPT(2)	Hartley 1990 [11]
−301	MBPT(2)	Kraftmakher 1988 [12]
−502, −562, −607, −1041	MBPT(1)	Johnson 1986 [13]

TABLE IV. Comparison of the E1 matrix element and hyperfine structure constants of low-lying states in Tl.

	This Work	Ref. [10]	Experiment
Transition		E1 amplitude	
$7S \rightarrow 6P_{1/2}$	1.82	1.73	1.81(2) [19]
State		Hyperfine constants	
$6P_{1/2}$	21 053	21 067	21 311 [20]
$7S$	11 894	11 417	12 297 [21]

to all-orders in the residual Coulomb interaction in the framework of the full fledged CCSD(T) theory. From Table II, it is clear that the correlation effects from p and d orbitals are also important and they should not be ignored. The valence-valence correlation of [10] is a subset of our core-valence correlation. Thus, the valence-valence, valence-core and core-core correlation effects included in our work contain substantially more higher-order effects than those taken into account either in [10] or in [9]. The configuration space spanned by the active orbitals in our calculation is also larger than those considered in the latter two calculations. The apparent agreement in the EDM enhancement factors between them seems to be fortuitous.

We have also compared our result with a few *ab initio* finite-order MBPT results [11–13] in Table III. As the correlation effects are very strong in the case of Tl, even a seemingly small approximation can drastically affect the final result which is evident from the two MBPT calculations, [11,12] with similar approximations. The even-parity channel (EPC) approximation, which considers only a set of multipoles allowed by the Coulomb selection rules, within the CCSD(T) approach has yielded $\mathcal{R} = -425$ [18] for the same basis set used in the current work and this further corroborates the sensitivity of the Tl EDM EF to electron correlation. The correlation trends observed in this work are in qualitative agreement with those observed in [14] for the Tl scalar-pseudoscalar EDM EF, as expected for two EDM operators with the same rank and parity.

The results of a few relevant physical quantities for the EDM EF such as the electric dipole ($E1$) matrix element for $7S \rightarrow 6P_{1/2}$ transition and the magnetic dipole hyperfine structure constants of the ground state $6P_{1/2}$ and the lowest singly excited state $7S$ have been presented in Table IV. It is clear that our results are in better agreement with the experiments than those reported in [10].

The error estimates quoted in [9,10] for Tl EDM EF do not seem to be convincing in view of the approximations they have made in their calculations. Clearly an all-order correlation calculation is needed for a reliable error estimation. Considering the error due to the use of a different basis set and the error due to the neglected triples, we obtain the final result for the EDM EF of Tl to be $-466(10)$. By combining the experimental result for the EDM of Tl, $-(4.0 \pm 4.3) \times 10^{-25} e$ cm, given by Regan *et al.* [8] and our accurate theoretical EDM EF, we obtain a new limit for the electron EDM to be $(8.7 \pm 9.3) \times 10^{-28} e$ cm which translates into an upper limit, $|d_e| < 2.0 \times 10^{-27} e$ cm at 90% confidence level.

In conclusion, we have obtained the most accurate limit for the electron EDM to date by improving the value of the EDM EF for the ground state of atomic Tl by treating the unusually strong electron correlation effects to all-order using the relativistic CCSD(T) method.

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