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Relativistic Many-body Calculations for Electric Dipole Moments in ^{129}Xe, ^{199}Hg, ^{223}Rn, ^{225}Ra and ^{171}Yb Atoms

Research Article

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Abstract. We present and compare the results of permanent *electric dipole moments* (EDMs) of various closed-shell atoms due to the *nuclear Schiff moment* (NSM) and the *tensor-pseudotensor* (T-PT) interactions between the atomic nuclei and electrons. In order to highlight the role of electron-correlation effects in obtaining accurate EDM results, we employ a number of relativistic many-body methods including coupled-cluster theory at different degrees of approximation. On combining our results obtained from the *relativistic coupled-cluster* (RCC) at the levels of singles and doubles excitations (CCSD method) with the available EDM measurements we obtain accurate bounds on the couplings S and C_T associated with the respective NSM and T-PT interactions. The most precise EDM measurement on ^{199}Hg in combination with our CC results yield limits on the above couplings as $S < 1.45 \times 10^{-12}|e|\text{fm}^3$ and $C_T < 2.09 \times 10^{-9}$ respectively. Further combining these bounds with the latest nuclear structure and quantum chromodynamics calculations we infer limits on the strong CP-violating parameter and for the combined up- and down- quark chromo-EDMs as $|\bar{\theta}| < 1.1 \times 10^{-9}$ and $|\widetilde{d}_u - \widetilde{d}_d| < 2.8 \times 10^{-26}|e|$cm, respectively.

Keywords. Coupled-cluster theory; Nuclear Schiff moment; Tensor-pseudotensor interaction; CP-violation; Electron correlation

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1. Introduction

Explaining matter-antimatter asymmetry in our Universe is one of the most challenging problems for the physicists. However to address this problem, one of the essential requirements is sufficient amount of CP violation in the elementary particle (interaction) level. The most celebrated *standard model* (SM) of particle physics inherit CP-violation in the form of complex phase in the *Cabibbo-Kobayashi-Maskawa* (CKM) matrix that mixes different flavors of quarks. The observed CP-violation in the decays of neutral K [1] and B [2–4] mesons are well within the physics described by the SM. However CKM mechanism is insufficient to account for the observed baryon asymmetry of the Universe, therefore search for new sources of CP-violation are of profound interest. In the low-energy sector, quest for the permanent *electric dipole moment* (EDM) of atoms and molecules have now the utmost importance. This is because existence of finite EDM in any non-degenerate system would be an unambiguous signatures of *parity* (P) and *time-reversal* (T) symmetries [5, 6]. Nonetheless, T violation means CP violation as per the famous CPT theorem [7]. The major contributor to the EDMs in the atomic systems are the P- and T-odd interactions among the constituent particles. Thus, studying atomic EDMs would provide powerful probe to the CP-violations emanating from the leptonic, semileptonic, and hadronic sources. In the present work we focus on the EDMs of the closed-shell (diamagnetic) atoms, which predominantly arise from the P- and T-odd electron-nucleus (e-N) tensor-pseudotensor (T-PT) and nuclear Schiff moment (NSM) interactions [8]. In order to estimate the strengths of the couplings associated with these interactions, we not only require precise measurements but also reliable many-body calculations. Till date, the measurement by Griffith *et al.* yields the best upper limit to the ^{199}Hg EDM as $d_A(^{199}\text{Hg}) < 3.1 \times 10^{-29}|e|$cm with 95% confidence level [9]. Advanced experimental techniques have been proposed for other diamagnetic atoms, specifically ^{129}Xe, with the expectation to improve the current sensitivity of the measurement by few orders of magnitude [10–15]. At the fundamental level, NSM can be linked with the $\bar{\theta}$ value and (chromo)-EDMs of quarks [18, 43, 44].

The rest of the paper is organized as follows. In the next section, we briefly present theory on the sources of EDMs to the closed-shell atoms. In Section 3, we give the framework of the *relativistic coupled-cluster* (RCC) theory to calculate EDMs of the atoms. Finally, we discuss our results in Section IV before concluding the work in the last section. Unless stated otherwise atomic units (au) are used throughout this paper.

2. Sources to EDMs of closed-shell atoms

Atoms can have permanent EDMs induced due to the P and T-odd moments of their nuclei. The general nuclear potential seen by an electron in an atomic system subjected to the screenings due to the other electrons is given by [19]

$$\phi(\mathbf{R}) = \int \frac{e\rho(\mathbf{r})}{|\mathbf{R}-\mathbf{r}|} d^3r + \frac{1}{Z}(\mathbf{d}\cdot\nabla)\int \frac{\rho(r)}{|\mathbf{R}-\mathbf{r}|} d^3r, \tag{2.1}$$

where $\rho(r)$ is the nuclear charge density normalized to atomic number (Z), and $\mathbf{d}$ is the nuclear EDM. In a closed-shell system with nuclear spin (I) equal to half, the dominant contribution to EDM comes from the NSM which is the lowest order P and T-odd term in the expansion of

the above potential. The corresponding Hamiltonian describing the interaction of an electron with the NSM for a finite size nucleus is given by [20],

$$H_{e-N}^{NSM} = -\frac{3\mathbf{S}\cdot\mathbf{r}}{B_4}\rho_N(r), \tag{2.2}$$

where $B_4 = \int_0^\infty dr r^4 \rho(r)$ and $\mathbf{S} = \frac{e}{10}\left[\langle r^2\mathbf{r}\rangle - \frac{5}{3Z}\langle r^2\rangle\langle\mathbf{r}\rangle\right]\frac{\mathbf{I}}{I}$ is the NSM. Origin of NSM owe to primary reasons at the nucleon level; the first possible reason is due to the distorted charge distribution inside the nucleus caused by the P and T violating interactions among nucleons mediated by neutral pi-mesons. The second possible source is due to the EDMs of individual nucleons generated through the self interactions in a pion loop.

The other important contribution to EDMs of closed-shell atoms comes from the T-PT interactions between the electrons and nucleons for which the interaction Hamiltonian is given as [21]

$$H_{e-N}^{T-PT} = i\sqrt{2}G_F C_T \sum_e \boldsymbol{\sigma_N}\cdot\boldsymbol{\gamma}\rho(r), \tag{2.3}$$

where G_F is the Fermi constant, C_T is the electron-nucleus T-PT coupling constant, $\boldsymbol{\sigma_N} = \langle\sigma_N\rangle\mathbf{I}/I$ is the Pauli spinor of the nucleus with spin I and $\rho(r)$ is the nuclear density. Since the strengths of these interactions are extremely weak, EDMs of the atomic systems can be estimated considering the electron-nucleus interactions only up to the first order effect with respect to the Coulomb interactions. In order to find out S and C_T values for an atomic system, it is imperative to combine measured EDMs with the corresponding calculations that depend on the atomic wave functions. For reliable calculations, a suitable many-body method capable of including the relativistic and electron correlation effects accurately is necessary to employ. For this purpose, we employ here the *relativistic coupled-cluster* (RCC) method, which is an all order many-body perturbation theory, with the *Dirac-Coulomb* (DC) Hamiltonian to carry out the required atomic calculations.

3. Method of Calculations

The DC Hamiltonian considered for the atomic calculations is given by

$$H_{DC} = \sum_i \left[c\boldsymbol{\alpha_D}\cdot\mathbf{p}_i + (\beta_D - 1)c^2 + V_n(r_i) + \sum_{j>i}\frac{1}{r_{ij}}\right], \tag{3.1}$$

where c is the velocity of light in vacuum, $\boldsymbol{\alpha_D}$ and β_D are the Dirac matrices, V_n denotes the nuclear potential obtained using the Fermi-charge distribution, $\frac{1}{r_{ij}}$ is the inter-electronic Coulombic repulsion and the energies are scaled with the rest mass energies of the electrons. The solutions of H_{DC} (atomic wave functions) is obtained approximately first using the mean-field approximation in the *Dirac-Fock* (DF) method. The corresponding ground state solution, denoted as $|\Phi_0\rangle$, is then taken as the reference state to obtain the exact state wave function ($|\Psi^{(0)}\rangle$) for the ground state. In the RCC theory formalism, $|\Psi^{(0)}\rangle$ is given as

$$|\Psi^{(0)}\rangle = e^{T^{(0)}}|\Phi_0\rangle, \tag{3.2}$$

where the RCC operator $T^{(0)}$ generates all possible excitations to form the configuration state functions from the reference state $|\Phi_0\rangle$. In the presence of EDM sources, the modified ground state wave function $|\Psi\rangle$ is expressed analogously as

$$|\Psi\rangle = e^{T}|\Phi_0\rangle = e^{T^{(0)}+\lambda T^{(1)}}|\Phi_0\rangle, \tag{3.3}$$

where λ is a suitable parameter representing strength of the EDM interaction Hamiltonians and $T^{(1)}$ generating the odd-parity excitations from $\Phi_0\rangle$ due to P and T odd interactions. In the first order approximation, we have

$$\begin{aligned}|\Psi\rangle &\approx |\Psi^{(0)}\rangle + \lambda|\Psi^{(1)}\rangle \\ &= [e^{T^{(0)}} + \lambda e^{T^{(0)}} T^{(1)}]|\Phi_0\rangle,\end{aligned} \tag{3.4}$$

with the first order wave function $|\Psi^{(1)}\rangle = e^{T^{(0)}}\lambda T^{(1)}|\Phi_0\rangle$. In our work, we restrict, only to the singles and doubles excitations through the T operators (CCSD method) by defining $T = T_1 + T_2$ which in the second quantization notations given as

$$T_1 = \sum_{a,p} a_p^\dagger a_a t_a^p \quad \text{and} \quad T_2 = \frac{1}{4}\sum_{a,b,p,q} a_p^\dagger a_q^\dagger a_b a_a t_{ab}^{pq}, \tag{3.5}$$

where t_a^p and t_{ab}^{pq} are the excitation amplitudes from the occupied orbitals denoted by a, b to the unoccupied orbitals denoted by p, q which embody all order correlation effects among the electrons.

The excitations amplitudes for the RCC operators are obtained by solving the following equations

$$\begin{aligned}|\Psi\rangle &\simeq \left(e^{T^{(0)}} + e^{T^{(0)}} T^{(1)}\right)|\Phi_0\rangle \\ &= e^{T^{(0)}}|\Phi_0\rangle + e^{T^{(0)}} T^{(1)}|\Phi_0\rangle \\ &= |\Psi^{(0)}\rangle + |\Psi^{(1)}\rangle,\end{aligned} \tag{3.6}$$

where $|\Psi^{(0)}\rangle$ and $|\Psi^{(1)}\rangle$ are the unperturbed and the first order perturbed wave functions due to the additional interaction. The $T^{(0)}$ and $T^{(1)}$ amplitudes are obtained by solving

$$\langle\Phi_0^\tau|\bar{H}_{DC}|\Phi_0\rangle = 0 \tag{3.7}$$

and

$$\langle\Phi_0^\tau|\bar{H}_{DC}T^{(1)}|\Phi_0\rangle = -\langle\Phi_0^\tau|\bar{H}_{add}|\Phi_0\rangle \tag{3.8}$$

respectively. Here $|\Phi_0^\tau\rangle$ corresponds to the excited configurations with τ referring to level of excitations from $|\Phi_0\rangle$ and H_{add} stands for either H_{e-N}^{NSM} or H_{e-N}^{T-PT} interaction. The line over any operator represents $\bar{O} = e^{-T^{(0)}} O e^{T^{(0)}} = (O e^{T^{(0)}})_c$ with subscript c means terms are connected. We also estimate the dominant contributions from the triple excitations by considering an excitation operator given by

$$T_3^{(0),pert} = \frac{1}{3!}\sum_{abc,pqr} \frac{(\bar{H}_{DC}T_2^{(0)})_{abc}^{pqr}}{\epsilon_a + \epsilon_b + \epsilon_c - \epsilon_p - \epsilon_q - \epsilon_r}, \tag{3.9}$$

where ϵ's are the single particle energies of the occupied (denoted by a,b,c) and unoccupied (denoted by p,q,r) orbitals. We considered this operator through Eqs. (3.7) and (3.8) to ameliorate the $T^{(0)}$ (referred as CCSD(T) method) and $T^{(1)}$ (referred as CCSD$_p$T method) amplitudes respectively.

The EDM of an atom (d_A), which is the expectation value of the electric dipole operator D, in the first order approximation is, thus, given by [20–25]

$$d_A = 2\frac{\langle\Psi_0^{(0)}|D|\Psi_0^{(1)}\rangle}{\langle\Psi_0^{(0)}|\Psi_0^{(0)}\rangle} = \frac{\langle\Phi_0|e^{T^{\dagger(0)}}De^{T^{(0)}}T^{(1)}|\Phi_0\rangle}{\langle\Phi_0|e^{T^{\dagger(0)}}e^{T^{(0)}}|\Phi_0\rangle}. \tag{3.10}$$

Since all the operators in the above expression are in normal order form the above equation can be further simplified and expressed as [27–29]

$$d_A = 2\langle\Phi_0|(\bar{D}^{(0)}T^{(1)})_{cc}|\Phi_0\rangle, \tag{3.11}$$

where the subscript cc stands for closed and connected terms and $\bar{D}^{(0)} = e^{T^{\dagger(0)}}De^{T^{(0)}}$, which is a non-truncative series. To account contributions from $\bar{D}^{(0)}$ maximally in the CCSD method, we follow many intermediate computing tricks as have been explained in detail elsewhere [30].

In addition, we consider only the linear terms of the CCSD method (refer to LCCSD method) to realize the importance of the non-linear terms that are usually correspond to higher triples, quadrupole etc excitations. Similarly, we have also investigated results using other lesser sophisticated techniques like *random phase approximation* (RPA), second (MBPT(2) method) and third (MBPT(3) method) order many-body perturbation theory [24, 25, 29] to gain into insights of the behavior of the propagation of the electron correlation effects and to compare our calculations with the earlier results reported using these methods.

4. Result and Discussions

In Tables 1 and 2, we present EDM results for many experimentally interesting closed-shell atoms such as ^{129}Xe, ^{199}Hg, ^{223}Rn, ^{225}Ra and ^{171}Yb from the NSM (d_A^{NSM}) and T-PT (d_A^{T-PT}) interactions respectively. We also present the trends of the electron correlation effects by employing the many-body methods at different levels of approximation that are mentioned earlier. All these systems have nuclear spin $I = 1/2$ and therefore contribution to the EDMs from octupole moment vanishes. Though the matrix elements of EDM interaction Hamiltonians usually increase faster than Z^2, however ^{129}Xe has an experimental edge over other heavier systems. It has a larger spin coherence time, of about 1000s which is approximately 5 times larger than ^{199}Hg, and hence its measurement can improve the statistical sensitivity by at-least an order of magnitude. As a matter of fact, three research groups across the globe are involved in measuring EDM of the Xe atom [12, 32, 33]. As shown in Tables 1 and 2, results for ^{129}Xe increase gradually with the inclusion of the correlation effects from the DF to LCCSD methods, and after that there is a fall in the magnitude when the non-linear contributions are included in the CCSD method. The results from our RPA method are in excellent agreements with the others [20, 21, 31] implying validation of our methods. These results are found to be larger than the CCSD results. The differences between the CCSD and RPA results can signify the

Table 1. The values of atomic EDM due to T-PT interaction (d_A^{T-PT})) in the units of $10^{-20} C_T \langle \sigma_{\mathbf{N}} \rangle |e| cm$ are presented using different many-body methods and compared them against the other calculation. The uncertainties in the results are given as Δ

Method	^{129}Xe	^{199}Hg	^{223}Rn	^{225}Ra	^{171}Yb
(This work)	[24]	[29]	[25]	[26]	
DF	0.447	−2.39	4.48	−3.46	−0.70
MBPT(2)	0.405	−4.48	3.93	−11.00	
MBPT(3)	0.515	−3.33	4.14	−10.59	
RPA	0.562	−5.89	5.40	−16.66	−3.39
LCCSD	0.608	−4.52	5.07	−13.84	
CCSD	0.501	−3.82	4.85	−10.04	
CCSD(T)		−4.20		−10.01	
CCSD$_p$T	0.501	−4.30			
QED+Breit		−4.44		−10.11	
Δ	±0.002	±0.09	±0.06	±0.19	
(Others)					
DF [20]	0.45	−2.0 [21], −2.4	4.6	−3.5	−0.70
RPA [20]	0.57, 0.564 [31]	−6.0 [21], −5.9	5.6	−17	−3.4
CI+MBPT [20]		−5.1		−18	−3.7
PRCC [23]		−4.3			

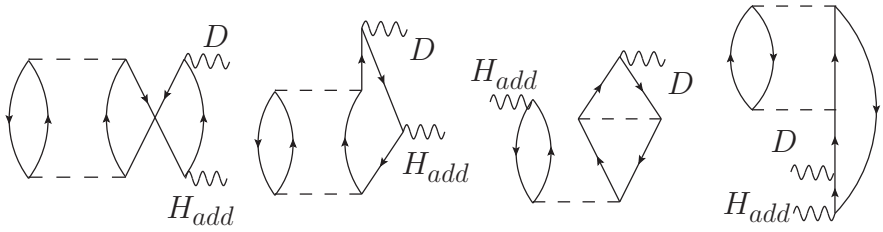

Figure 1. Significantly contributing non-RPA type MBPT(3) diagrams.

importance of the non-RPA contributions. Few large contributing non-RPA diagrams coming through the MBPT(3) method are shown in Figure 1. These non-RPA contributions cancel out strongly with the DF and RPA results, hence the CCSD results turn out to be smaller than the RPA and LCCSD results. We also estimate uncertainties to the calculated quantities by taking the difference between the CCSD and CCSD$_p$T methods and from the incompleteness in the basis functions which are given as Δ in Tables 1 and 2. To obtain limits on S and C_T, we combine our CCSD$_p$T results for d_A^{T-PT} and d_A^{NSM} with the available experimental EDM result for ^{129}Xe EDM, $d_A(^{129}Xe) < 4.1 \times 10^{-27} |e|$cm, and obtain limits as $C_T < 1.6 \times 10^{-6}$ and $S < 1.2 \times 10^{-9}$ $|e|$ fm^3. However, these limits can be improved further after combining the anticipated results from the ongoing experiments on ^{129}Xe [12, 32, 33].

The electron correlation effects in ^{199}Hg for the EDM calculations shows more or less similar trends as of ^{129}Xe. Our DF and RPA results for d_A^{T-PT} and d_A^{NSM} match perfectly with [20, 22] and also with another old calculation [21]. However, there are also an another sets of calculations reported using the *perturbed* RCC (PRCC) method, analogous to our CCSD method, which differ from our way of calculating RCC operator amplitudes and evaluating procedure of property

Table 2. The values of atomic EDM due to T-PT interaction (d_A^{SM})) in the units of $10^{-17}[S/(|e|\text{fm}^3)]|e|$ cm are presented using different many-body methods and compared them against the other calculation. The uncertainties in the results are given as Δ

Method	^{129}Xe	^{199}Hg	^{223}Rn	^{225}Ra	^{171}Yb
(This work)	[24]	[29]	[25]	[26]	
DF	0.288	−1.20	2.46	−1.85	−0.42
MBPT(2)	0.266	−2.30	2.36	−5.48	
MBPT(3)	0.339	−1.72	2.40	−5.30	
RPA	0.375	−2.94	3.31	−8.12	−1.91
LCCSD	0.417	−2.24	3.06	−8.40	
CCSD	0.336	−2.00	2.89	−6.79	
CCSD(T)		−2.08		−6.79	
CCSD$_p$T	0.337	−2.12			
QED+Breit		−2.16		−6.80	
Δ	±0.004	±0.03	±0.04	±0.10	
(Others)					
DF [20]	0.29	−1.19, −1.20	2.47, 2.5	−1.8	−0.42
RPA [20]	0.38	−2.8, −3.0	3.33, 3.3	−8.3, −8.5	−1.9, −1.9
CI+MBPT [20]		−2.6		−8.8	−2.1
PRCC [23]		−5.07			

using Eq. (3.11) [23]. Calculations for ^{199}Hg are further improved to obtain better limits to C_T and S by adding corrections due to the frequency independent Breit interaction given by

$$V_B(r_{ij}) = -\frac{1}{2r_{ij}}\{\boldsymbol{\alpha}_i\cdot\boldsymbol{\alpha}_j + (\boldsymbol{\alpha}_i\cdot\hat{\mathbf{r}}_{\mathbf{ij}})(\boldsymbol{\alpha}_{\mathbf{j}}\cdot\hat{\mathbf{r}}_{\mathbf{ij}})\} \tag{4.1}$$

and from the lower order vacuum polarization effects from the *quantum electrodynamics* (QED) corrections through the Uehling ($V_U(r)$) and Wichmann-Kroll ($V_{WK}(r)$) potentials given by

$$V_U(r) = -\frac{4}{9c\pi}V_N(r)\int_1^\infty dt\sqrt{t^2-1}\left(\frac{1}{t^2}+\frac{1}{2t^4}\right)e^{-2ctr} \tag{4.2}$$

and

$$V_{WK}(r) = -\frac{2}{3}\frac{1}{c\pi}V_N(r)\frac{0.092c^2Z^2}{1+(1.62cr)^4}\ . \tag{4.3}$$

In this case, the net uncertainties to the calculations are estimated by evaluating contributions from the neglected higher excitations using the $T_3^{(0),pert}$ operator in Eq. (3.11) and from the higher orbitals including from h- and i-symmetries that were not considered in the RCC calculations. Further from the above tables, we can observe that the previous calculations using the PRCC method [23] and CI+MBPT method [20] differ significantly with each other. The CI+MBPT method is the hybrid method composed of *configuration interaction* (CI) method with finite-order many-body perturbation theory where the initial wave functions are determined using the V^{N-2} potential for N number of electrons. The electron correlation effects are accounted in this method by dividing the electrons into valence and core types. In contrast,

the electron correlation effects in our and in the PRCC method are considered by carrying out calculations using the V^N potential by treating correlating effects due to all the electrons on equal footing. Hence, we believe that our results are the most accurate and valid compared to the previous calculations. Our results differ by about 15-20% from the calculations given in [20]. Combining our final results with the measurement [9], we get limits as $S < 1.45 \times 10^{-12}|e|$ fm^3 and $C_T < 2.09 \times 10^{-9}$. These are the most accurate bounds to S and C_T at present. Moreover, C_T is further related to the scalar-pseudoscalar coupling constant C_P associated with the P and T-odd electron-nucleus interaction by the relation $C_P \approx 3.8 \times 10^3 \times \frac{A^{1/3}}{Z} C_T$ [20], where A is the atomic mass. Using this relation the limit C_P is extracted as $C_P < 5.8 \times 10^{-7}$ and from the relation $S = (1.9 d_n + 0.2 d_p$ fm^2 [34], we get the limits on neutron (d_n) and proton (d_p) EDMs as $d_n < 7.6 \times 10^{-26}|e|$ cm and $d_p < 7.3 \times 10^{-25}|e|$ cm respectively. Although our extracted limit on d_n is not better than the limit obtained directly from the measurement [35], the limit on d_p, however, is better than the previous value [9].

As has been pointed out in a recent review [43], all the nuclear calculations available till date for ^{199}Hg are in discordance with one another both in signs and magnitudes. However, Ref. [43] also provides the best value for S in terms of $\bar{g}_i$ as

$$S = 13.5\left[0.01\bar{g}_0 + (\pm 0.02)\bar{g}_1 + 0.02\bar{g}_2\right] |e| \text{ fm}^3 \tag{4.4}$$

where the couplings $\bar{g}_i$ with the subscript $i = 0, 1, 2$ represent the isospin components of the CP-odd pion-nucleon coupling constants. Combining this with our limit on S, we infer bounds as $|\bar{g}_0| < 1.2 \times 10^{-11}$ and $|\bar{g}_1| < 5.6 \times 10^{-12}$. Furthermore, using the relations $\bar{g}_0 = -0.018(7)\bar{\theta}$ [44] and $\bar{g}_1 = 2 \times 10^{-12}(\tilde{d}_u - \tilde{d}_d)$ [45], we extract the upper limit on the combined up- and down- quarks chromo-EDMs as $|\tilde{d}_u - \tilde{d}_d| < 2.8 \times 10^{-26}|e|$ cm and the limit on the strong CP-odd parameter as $|\bar{\theta}| < 1.1 \times 10^{-9}$. In fact, it is also possible to infer more stringent limits on the above quantities from our given limit on S provided the uncertainties in the nuclear calculations are reduced further.

The octupole deformed nuclei in ^{223}Rn, ^{225}Ra and ^{171}Yb are responsible for collectively large Schiff moments in these systems and hence, they are very interesting candidates for the EDM studies. Our EDM results due to the T-PT and NSM interactions in ^{223}Rn agrees well with that of Dzuba *et al.* [20, 22] at the DF and RPA level. Again like ^{129}Xe and ^{199}Hg, the correlation trends in the ^{223}Rn EDM results follow similar pattern. We also present our rigorous CCSD results where we observe significant cancellations between the RPA and the all-order non-RPA contributions. The inclusion of the non-RPA terms which starts appearing through the MBPT(3) method onwards in a perturbative theory framework is, therefore, very crucial. Moreover, we also find large cancellations among the contributions from the linear and non-linear CCSD terms in the EDM calculations. It is, therefore, imperative to use an all order approach like our CCSD method to get more reliable results for EDMs in these systems. The differences in the LCCSD and CCSD results given in Tables 1 and 2 highlight the importance of the non-linear correlation terms such as $T_1^{(0)}T_2^{(0)}$, $\frac{1}{2}T_2^{(0)}T_2^{(0)}$, $\cdots$, which correspond to the contributions from higher level excitations such as triples, quadruples, etc. The errors in the calculations were again determined in the similar manner as explained above.

We also present our EDM results for ^{225}Ra atom [26] whose first measurement has been reported recently by the Argonne National Laboratory [38]. Their obtained limit is not

competitive at present with the current best limit set by the ^{199}Hg experiment [9]. However, the larger Z and the octupole deformation in the nucleus of ^{225}Ra can enhance EDM by two to three orders of magnitude compared to ^{199}Hg [39, 40]. In addition to that cold-atom techniques with very little sensitivity to systematics [41] have been developed to measure the Larmor spin-resonance frequency for ^{225}Ra. Moreover, ANL hopes to improve the enhance the statistical sensitivity of the measurement by increasing the number of atoms that can be observed to 10^6, using the *Facility for Rare Isotope Beams* (FRIB) for a measurement time of 100 days [38]. On combining our CCSD(T) result for d_A^{SM} with the measured EDM value [38], we get an upper bound on NSM as $S < 7.4 \times 10^{-6}|e|\,\mathrm{fm}^3$ [26]. Similarly with the knowledge of $\langle\sigma_n\rangle$ in ^{225}Ra from nuclear calculation, an upper bound on C_T can be predicted. Two sophisticated nuclear calculations have been carried out using the octupole deformed Wood-Saxon potential [40] and odd-A Skyrme mean field theory [42] to describe the P,T-odd interactions in ^{225}Ra in terms of the pion-nucleon-nucleon couplings. In a recent review, Engel *et al.* give the best value for S from these two calculations as [43]

$$S = 13.5[-1.5\bar{g}_0 + 6.0\bar{g}_1 - 4.0\bar{g}_2]|e|\mathrm{fm}^3. \tag{4.5}$$

We infer bounds as $|\bar{g}_0| < 3.6\times10^{-7}$ and $|\bar{g}_1| < 9.1\times10^{-8}$ using the above result with our extracted limit on S [26]. Again from the relations $|\bar{g}_0| = 0.018(7)\theta_{QCD}$ [44] and $|\bar{g}_1| = 2\times10^{-12}(\tilde{d}_u - \tilde{d}_d)$ [45], we put the upper limits as $|\theta_{QCD}| < 2.0\times10^{-5}$ and $|\tilde{d}_u - \tilde{d}_d| < 4.6\times10^{-22}|e|$ cm. Though these bounds are not competitive at present with the corresponding limits acquired from the ^{199}Hg EDM study [29], the limits can become more stringent when our results will be combined with the anticipated improved ^{225}Ra EDM measurement.

The EDM results for the ^{171}Yb are presented only at the level of RPA which are in excellent agreement with the values reported by Dzuba *et al.* [20, 22]. However, in analogy with our EDM studies on other atoms we expect a considerable non-RPA contributions which must be incorporated in order to get accurate EDM results in ^{171}Yb.

5. Conclusion

In summary, we present trends in the electron correlation effects in the calculations of EDMs of ^{129}Xe, ^{199}Hg and ^{223}Rn. On combing our results with the most EDM precise measurement for ^{199}Hg atom, we are able to put bounds on NSM and T-PT coupling constant as $S < 1.45\times10^{-12}|e|\,\mathrm{fm}^3$ and $C_T < 2.09\times10^{-9}$ respectively. Moreover, combining the NSM obtained from our study with the latest nuclear calculation yield, the upper limits on the combined up- and down- quarks chromo-EDMs as $|\tilde{d}_u - \tilde{d}_d| < 2.8\times10^{-26}|e|$ cm and on the strong CP-odd parameter as $|\bar{\theta}| < 1.1\times10^{-9}$. We have also obtained preliminary EDM results for ^{171}Yb at the RPA level which are in excellent agreements with the previous calculations. Our reported atomic calculations in the considered closed-shell atoms in combination with the upcoming EDM measurements involving more advanced experimental techniques would further constrain to the values of the above obtained coupling constants. Our obtained limits on various P,T-odd couplings from ^{199}Hg would definitely constraint various extensions of the *standard model* (SM) of particle physics which can be further useful for probing new physics beyond-SM.

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Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

All the authors contributed equally and significantly in writing this article. All the authors read and approved the final manuscript.

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