Relativistic Normal Coupled-Cluster Theory for Accurate Determination of Electric Dipole Moments of Atoms: First Application to the ^{199}Hg Atom

B. K. Sahoo[1,2,*] and B. P. Das[3,†]

[1]Physical Research Laboratory, Atomic, Molecular and Optical Physics Division, Navrangpura, Ahmedabad-380009, India
[2]State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China
[3]Department of Physics and International Education and Research Center of Science, Tokyo Institute of Technology, 2-12-1 Ookayama Meguro-ku, Tokyo 152-8550, Japan

(Received 22 October 2017; published 15 May 2018)

Recent relativistic coupled-cluster (RCC) calculations of electric dipole moments (EDMs) of diamagnetic atoms due to parity and time-reversal violating (P,T-odd) interactions, which are essential ingredients for probing new physics beyond the standard model of particle interactions, differ substantially from the previous theoretical results. It is therefore necessary to perform an independent test of the validity of these results. In view of this, the normal coupled-cluster method has been extended to the relativistic regime [relativistic normal coupled-cluster (RNCC) method] to calculate the EDMs of atoms by simultaneously incorporating the electrostatic and P,T-odd interactions in order to overcome the shortcomings of the ordinary RCC method. This new relativistic method has been applied to ^{199}Hg, which currently has a lower EDM limit than that of any other system. The results of our RNCC and self-consistent RCC calculations of the EDM of this atom are found to be close. The discrepancies between these two results on the one hand and those of previous calculations on the other are elucidated. Furthermore, the electric dipole polarizability of this atom, which has computational similarities with the EDM, is evaluated and it is in very good agreement with its measured value.

DOI: 10.1103/PhysRevLett.120.203001

There have been remarkable advances in tabletop experiments to explore fundamental physics in the recent past [1]. These experiments have a symbiotic relationship with theory. In particular, the combined results of experiments and relativistic many-body calculations for atomic electric dipole moments (EDMs) due to parity and time-reversal violating (P,T-odd) interactions can provide important insights into CP violation corresponding to mass scales of tens of TeV or larger [2,3] and thereby probe new physics beyond the standard model (BSM). Of all the systems on which experiments have been performed, mercury yields the most sensitive limit to date. The measured value of the EDM for ^{199}Hg, $d(^{199}\mathrm{Hg}) = [-2.20 \pm 2.75(\mathrm{stat}) \pm 1.48(\mathrm{syst})] \times 10^{-30}\ e\,\mathrm{cm}$ with 95% confidence level [4]. Further improvement of this limit by 2–3 orders could provide crucial information on the validity of certain BSMs, in particular, some variants of the multi-Higgs, supersymmetric, left-right symmetric models [2,3,5]. It would, therefore, be very desirable to determine precise limits for CP violating parameters from the EDM of ^{199}Hg. Studies on the EDMs of diamagnetic atoms entail the synergy between the state-of-the-art atomic experiments and three different areas of theoretical physics—particle, nuclear, and atomic physics. In addition, the atomic theory part of this problem, which is the focus of the present Letter, involves the interplay of electromagnetic and combined parity and time-reversal violating interactions, and it consequently extends the boundaries of quantum many-body theory. Rigorous calculations of EDMs considering the nuclear Schiff moment (NSM) and the electron-nucleus (e-N) tensor-pseudotensor (T-PT) interactions have been performed in the diamagnetic systems that are under consideration in experiments using the relativistic coupled-cluster (RCC) theory [5–9]. By combining the results of the latest two calculations [5,6] with that of the measured EDM value of ^{199}Hg, we obtained upper limits for the NSM, $|S| < 4.2 \times 10^{-13}|e|\mathrm{fm}^3$, and the T-PT coupling coefficient, $|C_T| < 7.0 \times 10^{-10}$. Further combining these values with the nuclear calculations and quantum chromodynamics (QCD), gave the limits $d_n < 2.2 \times 10^{-26}|e|\mathrm{cm}$ and $d_p < 2.1 \times 10^{-25}|e|\mathrm{cm}$ for the EDMs of the neutron and the proton, respectively, and $|\bar{\theta}| < 1.1 \times 10^{-10}$ and $|\tilde{d}_u - \tilde{d}_d| < 5.5 \times 10^{-27}|e|\mathrm{cm}$ for the QCD parameter and the combined up- and down-quark chromo-EDMs. respectively [5,6].

Our recent RCC results for ^{199}Hg [5,6] differ by about 20%–50% from those obtained previously by employing a variety of relativistic many-body methods (discussions on these results can be found in Refs. [5,7]). The results of another RCC calculation, referred to in the literature as the

perturbed RCC (PRCC) method, in this atom, coauthored by one of us [10], is also not in agreement with our latest work [6] on this subject. The differences between the two RCC methods are essentially technical in nature [10–12]. In our formalism of the RCC method, the expectation value of an operator contains only connected terms in the numerator after the normalization cancels out in the numerator and the denominator [13,14]. In one of the RCC calculations of the properties of ^{199}Hg, a large number of lower-order terms, arising from the nonterminating connected terms of the exponential terms containing the cluster excitation and deexcitation operators in the expression for the EDM, have been computed with relatively less computational effort [7]. The results of these properties were in reasonable agreement with some of the results reported previously, but further attempts to include higher-order nonlinear terms through an iterative procedure reduced the magnitudes of the results significantly [5,6]. In fact, the discrepancies between these results and those of previous calculations are about 50% in the case of ^{171}Yb [8] and ^{225}Ra atoms [9]. Thus, it is imperative to test the validity of these calculations by taking recourse to a reliable method that is capable of overcoming the drawbacks of the regular RCC method.

The coupled-cluster (CC) method has been applied to atoms [14–17], molecules [13,14,18,19], condensed matter systems [20,21], and nuclei [22,23]. It is currently one of the leading quantum many-body methods and has been referred to as the gold standard for treating electron correlation [14,18,24,25]. It is straightforward to apply RCC methods for the evaluation of energies, but not for other properties. The normal coupled-cluster (NCC) method is tailor-made for the evaluation of expectation values of operators corresponding to different properties [21,24–27]. In contrast to the usual RCC method, it possesses two important attributes that make it attractive for the calculations of different atomic properties. The first is that it satisfies the Hellman-Feyman theorem and the second is that expectation values of operators terminate in a natural way [21,26,27]. Thus, the development of the NCC method in the relativistic framework (RNCC) can lead to an improvement in the accuracies of the calculations of atomic properties. Unlike molecular calculations, it is possible to exploit the spherical symmetry of the systems to develop the RNCC method for atoms in the presence of $P;T$-odd interactions. As these interactions are very weak compared to the electrostatic interactions, the EDMs of atoms are evaluated by expressing the wave functions as linear combinations of wave functions of states of opposite parities using the first-order perturbation theory. In the RNCC method, this applies to both the ket and bra states. In view of the steady advances in the EDM experiments on diamagnetic atoms in recent years [4,28], it is essential to develop theories like the RNCC method for improving the accuracy of the atomic calculations to probe BSM physics.

A different approach to the evaluation of properties by the CC method had been proposed by Monkhorst based on linear and higher-order response theories [29]. It had been applied to various properties, including the EDMs of diamagnetic atoms [30]. However, this method is not as versatile as the NCC method. Furthermore, the calculation of atomic EDMs by this method is less straightforward than that using the NCC method since it involves the computation of the second derivative of the energy with respect to the P,T-odd coupling coefficients and the electric field, which requires a knowledge of three different perturbed CC amplitudes [30].

In this Letter, we outline the general theory of the RNCC method for atoms in the presence of P,T-odd interaction Hamiltonians and the electric dipole operator as external perturbations and discuss its implementation for the determination of the EDM (d_a) and the electric dipole polarizability (α_d) of atomic systems, respectively. As the first application, we evaluate these properties for ^{199}Hg using the RNCC method and compare them with the results from the RCC method, and also the latter property is compared with its measured value.

We begin with the Dirac-Coulomb Hamiltonian, which is given in atomic units (a.u.) by

$$H_a = \sum_i \left(c\boldsymbol{\alpha}_D \cdot \mathbf{p}_i + (\beta_D - 1)c^2 + V_N(r_i) + \sum_{j \geq i} \frac{1}{r_{ij}} \right), \tag{1}$$

where $\boldsymbol{\alpha}_D$ and β_D are the Dirac matrices, c is the velocity of light, and $V_N(r_i)$ is the nuclear potential experienced by an electron in an atom. The P,T-odd T-PT interaction Hamiltonian is given by [5]

$$H_{e\text{-}N}^{TPT} = i\sqrt{2}G_F C_T \sum_i \boldsymbol{\sigma}_N \cdot \gamma \rho_N(r_i), \tag{2}$$

where G_F is the Fermi constant, C_T is the T-PT e-N coupling constant, $\boldsymbol{\sigma}_N = \langle \sigma_N \rangle \mathbf{I}/I$ is the Pauli spinor for the nucleus with spin I, $\gamma = i\alpha_D\beta_D$, and $\rho_N(r_i)$ is the nuclear density.

The P,T-odd Hamiltonian representing the interaction of the NSM with an electron in an atom is given by [5]

$$H_{e\text{-}N}^{\mathrm{NSM}} = \frac{3}{B_4} \sum_i \mathbf{S} \cdot \mathbf{r}_i \rho_N(r_i), \tag{3}$$

where $\mathbf{S} = S\frac{\mathbf{I}}{I}$ is the NSM and $B_4 = \int_0^\infty dr r^4 \rho_N(r)$.

With the Dirac-Hartree-Fock (DHF) wave function $|\Phi_0\rangle$ reference state, the ground state wave function $|\Psi_0^{(0)}\rangle$ is expressed in the RCC method as [13]

$$|\Psi_0^{(0)}\rangle = e^{T^{(0)}} |\Phi_0\rangle, \tag{4}$$

where $T^{(0)}$ is the even parity RCC excitation operator embodying correlation effects. In the particle-hole excitation formalism, we express $T^{(0)} = \sum_{I=1}^{N_c} t_I^{(0)} C_I^+$ with N_c representing the number of electrons in the system, $t_I^{(0)}$ are the amplitudes of the excitations, and C_I^+ stands for a string of annihilation-creation operators corresponding to a general particle-hole excitation. The equation for the ground state of H_a is given by

$$H_a|\Psi_0^{(0)}\rangle = E_0^{(0)}|\Psi_0^{(0)}\rangle, \tag{5}$$

with energy $E_0^{(0)}$. The equations for the cluster amplitudes $T^{(0)}$ are obtained by using Eq. (4) and projecting Eq. (5) on the bra state $\langle\Phi_0|C_I^- e^{-T^{(0)}}$ as

$$\langle\Phi_0|C_I^-\bar{H}_a|\Phi_0\rangle = 0, \tag{6}$$

where the deexcitation operators C_I^- are the Hermitian conjugate (H.c.) of C_I^+. We use the notation $\bar{O} = e^{-T}Oe^T = (Oe^T)_c$ throughout the Letter for a general operator O, where the subscript c stands for connected terms [14]. For one- and two-body operators O, $\bar{O}$ terminates naturally [14,18].

The inclusion of a weak P,T-odd interaction Hamiltonian or the electric dipole operator D, denoted by H_λ, modifies the ground state wave function as

$$|\Psi_0\rangle = e^T|\Phi_0\rangle = e^{T^{(0)}+\lambda T^{(1)}}|\Phi_0\rangle, \tag{7}$$

where the effect of the perturbation is represented by $T^{(1)} = \sum_{I=1}^{N_c} t_I^{(1)} C_I^+$ with the amplitudes $t_I^{(1)}$, which includes one order of the weak odd-parity perturbation of interest and all orders of the residual Coulomb interaction. Here λ represents the strength of the coupling coefficient of a given P,T-odd interaction or the electric field for the evaluation of d_a and α_d, respectively. $|\Psi_0\rangle$ is clearly a mixed-parity state and to the first order in one of the odd-parity operators, we can express [5–7]

$$|\Psi_0\rangle \simeq |\Psi_0^{(0)}\rangle + \lambda|\Psi_0^{(1)}\rangle. \tag{8}$$

This corresponds to

$$|\Psi_0^{(1)}\rangle = e^{T^{(0)}}T^{(1)}|\Phi_0\rangle. \tag{9}$$

The first-order perturbed wave function satisfies the following equation

$$(H_a - E_0^{(0)})|\Psi_0^{(1)}\rangle = (E_0^{(1)} - H_\lambda)|\Psi_0^{(0)}\rangle. \tag{10}$$

The equation for the amplitudes for $T^{(1)}$ can be obtained from the above first-order perturbed equation as [5–7]

$$\langle\Phi_0|C_I^-(\bar{H}_a T^{(1)} + \bar{H}_\lambda)|\Phi_0\rangle = 0. \tag{11}$$

For the calculations of $|\Psi_0^{(0)}\rangle$ and $|\Psi_0^{(1)}\rangle$, we consider singles (one particle-one hole) and doubles (two particle-two hole) excitations in the RCC theory (RCCSD method) by restricting to $I = 1, 2$ in the amplitude equations. The expectation value of an operator O in the ground state of a closed-shell system using the (R)CC method is expressed as [13,14,25]

$$\langle O\rangle \equiv \frac{\langle\Psi_0|O|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle} = \langle\Phi_0|e^{T^\dagger}Oe^T|\Phi_0\rangle_c. \tag{12}$$

Following the above expression and expanding $|\Psi_0\rangle$, we can evaluate d_a and α_d, commonly denoted as X, of the ground state of a closed-shell atom by [7]

$$\begin{aligned} X &\equiv \lambda\langle\Psi_0^{(1)}|D|\Psi_0^{(0)}\rangle + \langle\Psi_0^{(0)}|D|\Psi_0^{(1)}\rangle \\ &= 2\lambda\langle\Phi_0|e^{T^{(0)\dagger}}De^{T^{(0)}}T^{(1)}|\Phi_0\rangle_c. \end{aligned} \tag{13}$$

The above expression is nonterminating because of $e^{T^{\dagger(0)}}De^{T^{(0)}}$. Unlike this approach, the normalization factor appears explicitly in the PRCC method [10–12]. It is, therefore, necessary to adopt a method that can overcome the nonterminating terms and resolve the ambiguity of accounting for contributions from the normalization of the wave function. As discussed below, the RNCC method achieves both these objectives in a natural manner.

In the RNCC method, the unperturbed ket is the same as that in the RCC method, but the bra $\langle\Psi_0^{(0)}|$ is replaced by $\langle\tilde{\Psi}_0^{(0)}|$ and is defined as [21,24,25,27]

$$\langle\tilde{\Psi}_0^{(0)}| = \langle\Phi_0|(1 + \tilde{T}^{(0)})e^{-T^{(0)}}, \tag{14}$$

where $\tilde{T}^{(0)} = \sum_{I=1}^{N_c} \tilde{t}_I^{(0)} C_I^-$ is an deexcitation operator with amplitudes $\tilde{t}_I^{(0)}$, similar to $T^{\dagger(0)} = \sum_{I=1}^{N_c} t_I^{(0)} C_I^-$, such that it satisfies

$$\langle\tilde{\Psi}_0^{(0)}|\Psi_0^{(0)}\rangle = \langle\Phi_0|(1 + \tilde{T}^{(0)})e^{-T^{(0)}}e^{T^{(0)}}|\Phi_0\rangle = 1. \tag{15}$$

It can be easily shown that $\langle\tilde{\Psi}_0^{(0)}|$ has the same eigenvalue as $|\Psi_0^{(0)}\rangle$ (or $\langle\Psi_0^{(0)}|$) with the condition

$$\langle\Phi_0|\tilde{T}^{(0)}\bar{H}_a|\Phi_0\rangle = 0. \tag{16}$$

In fact, this condition is the direct consequence of Eq. (6). Hence, $\langle\tilde{\Psi}_0^{(0)}|$ can be used in place of $\langle\Psi_0^{(0)}|$ in the calculation of atomic properties. Starting from the bra equation

$$\langle\tilde{\Psi}_0^{(0)}|H_a = E_0^{(0)}\langle\tilde{\Psi}_0^{(0)}|, \tag{17}$$

the amplitudes of $\tilde{T}^{(0)}$ can be determined using Eq. (14) and projecting Eq. (17) on the ket state $e^{T^{(0)}}C_I^+|\Phi_0\rangle$ as

$$\langle\Phi_0|(\tilde{T}^{(0)}\bar{H}_a + \bar{H}_a)C_I^+|\Phi_0\rangle = 0. \tag{18}$$

It is interesting to note that the ket and bra equations, (6) and (18), respectively, can be derived from a variational principle [27]. After obtaining these solutions, the expectation value of an operator O in the RNCC method can be written as [21,24,25]

$$\langle O\rangle \equiv \frac{\langle\tilde{\Psi}_0^{(0)}|O|\Psi_0^{(0)}\rangle}{\langle\tilde{\Psi}_0^{(0)}|\Psi_0^{(0)}\rangle} = \langle\Phi_0|(1+\tilde{T}^{(0)})\bar{O}|\Phi_0\rangle_c. \tag{19}$$

The above expression, unlike its counterpart in the RCC method, is a terminating series and the normalization factor is unity. But to evaluate EDMs and electric dipole polarizabilities of atoms, the above (R)NCC approach needs further modification as demonstrated in this Letter.

To obtain the first-order correction to the bra state, we can replace $\langle\Psi_0|$ of Eq. (12) by $\langle\tilde{\Psi}_0|$ defining as

$$\langle\tilde{\Psi}_0| = \langle\Phi_0|(1+\tilde{T})e^{-T}, \tag{20}$$

where $\tilde{T}$ is the new deexcitation RCC operator for the total wave function. Expanding the bra wave function and retaining terms to the first order yields

$$\begin{aligned}\langle\tilde{\Psi}_0| &\simeq \langle\tilde{\Psi}_0^{(0)}| + \lambda\langle\tilde{\Psi}_0^{(1)}| \\ &= \langle\Phi_0|(1+\tilde{T}^{(0)}+\lambda\tilde{T}^{(1)})e^{-(T^0+\lambda T^{(1)})}.\end{aligned} \tag{21}$$

It is straightforward to show that $\langle\tilde{\Psi}_0|$ can satisfy the biorthogonal condition with $|\Psi_0\rangle$. Analogous to Eq. (11), the amplitudes for $\tilde{T}^{(1)}$ starting from the first-order perturbed bra can be obtained by solving

$$\langle\Phi_0|[\tilde{T}^{(1)}\bar{H}_a + (1+\tilde{T}^{(0)})\{\bar{H}_\lambda + (\bar{H}_a T^{(1)})_c\}]C_I^+|\Phi_0\rangle = 0. \tag{22}$$

Again, we express $\tilde{T}^{(0/1)} = \tilde{T}_1^{(0/1)} + \tilde{T}_2^{(0/1)}$ in the singles and doubles approximation of the RNCC method (RNCCSD method) corresponding to their respective $T^{(0/1)}$ operators in the RCCSD method. In this approach, the expectation value X is evaluated by

$$\begin{aligned}X \equiv \frac{\langle\tilde{\Psi}_0|D|\Psi_0\rangle}{\langle\tilde{\Psi}_0|\Psi_0\rangle} &= \langle\Phi_0|(1+\tilde{T})e^{-T}De^{T}|\Phi_0\rangle \\ &= \lambda\langle\Phi_0|(1+\tilde{T}^{(0)})\bar{D}T^{(1)} + \tilde{T}^{(1)}\bar{D}|\Phi_0\rangle_c.\end{aligned} \tag{23}$$

This expression terminates and can give fewer terms than Eq. (13). Also, it does not have any H.c. terms like those in the RCC method. Thus, a one-to-one comparison between the contributions from various terms from Eqs. (13) and (23) will be instructive. Substantial discrepancies between the final results of the RNCC and the RCC methods will reflect the incompleteness of the latter.

We give a summary of the results obtained for d_a considering both the T-PT and NSM P,T-odd interaction Hamiltonians and also the α_d value of ^{199}Hg using the DHF, many-body perturbation theory with second- [MBPT(2)] and third-order [MBPT(3)] approximations, random-phase approximation (RPA), combined configuration interaction and many-body theory (CI + MBPT), multiconfiguration Dirac-Fock (MCDF), PRCC, and RCC approaches, which were extensively discussed recently in Refs. [5–7], in Table I. We also quote values explicitly from the linear terms of the RCCSD (LRCCSD) method and a self-consistent RCCSD approach in which the combined power of $T^{(0)}$ and $T^{(0)\dagger}$ is systematically increased in Eq. (13) until the value of the EDM converges. The latter method is designated as RCCSD$^{(k)}$, where k is the total number of $T^{(0)}$ and $T^{(0)\dagger}$ operators together in the nonterminating series. It is evident from Table I that there are discrepancies between the results based on different methods. The results from RCCSD$^{(\infty)}$ and RNCCSD are in very good agreement as expected, but they differ significantly from those of other methods. This demonstrates the importance of the

TABLE I. A summary of d_a values from the T-PT e-N interaction (in $10^{-20}C_T\langle\sigma_N\rangle$ $|e|$ cm) and NSM (in $10^{-17}[S/|e|\text{fm}^3]$ $|e|$ cm) and α_d (in a.u.) in the ^{199}Hg atom from different methods that are discussed in Refs. [5–7] and RNCCSD method. Here RCCSD$^{(\infty)}$ values are the final RCCSD results. Corrections from the Breit interaction and triples excitations, and uncertainties due to the use of finite basis functions, are also mentioned separately. The final values along with net uncertainties are quoted at the end.

Method	T-PT	NSM	α_d
DHF	−2.39	−1.20	40.95
MBPT(2)	−4.48	−2.30	34.18
MBPT(3)	−3.33	−1.72	22.98
RPA	−5.89	−2.94	44.98
CI + MBPT	−5.1	−2.6	32.99
MCDF	−4.84	−2.22	
PRCC	−4.3	−2.46	33.29
LRCCSD	−4.52	−2.24	33.91
RCCSD$^{(2)}$	−3.82	−2.00	33.76
RCCSD$^{(4)}$	−4.14	−2.05	35.13
RCCSD$^{(5)}$	−4.02	−2.00	34.98
RCCSD$^{(\infty)}$	−3.17	−1.76	34.51
RNCCSD	−3.30	−1.77	34.22
Δ Breit	0.03	0.04	−0.01
Δ Triples	∼0.0	∼0.0	−0.28
Basis	±0.03	±0.02	±0.15
Final	−3.30(6)	−1.77(6)	34.2(5)
Experiment [31]			33.91(34)

TABLE II. Comparison of contributions from various RCC and RNCC terms to d_a and α_d values (with same units as in Table I). The contributions using different bra states in the two methods exhibit very different trends, but the final results are in very good agreement.

RCC term	RCC result	RNCC term	RNCC result
Contributions to d_a from T-PT interaction			
$DT_1^{(1)}$	−2.20	$DT_1^{(1)}$	−2.20
$T_1^{(1)\dagger}D$	−2.20	$\tilde{T}_1^{(1)}D$	−1.74
$T_1^{(1)\dagger}DT_2^{(0)}$	0.61	$\tilde{T}_1^{(1)}DT_2^{(0)}$	0.52
$T_2^{(0)\dagger}DT_2^{(1)}$	0.01	$\tilde{T}_2^{(0)}DT_2^{(1)}$	0.01
$T_2^{(1)\dagger}DT_2^{(0)}$	0.01	$\tilde{T}_2^{(1)}DT_2^{(0)}$	−0.04
Others	0.60	Others	0.15
Contributions to d_a from NSM interaction			
$DT_1^{(1)}$	−1.19	$DT_1^{(1)}$	−1.19
$T_1^{(1)\dagger}D$	−1.19	$\tilde{T}_1^{(1)}D$	−0.88
$T_1^{(1)\dagger}DT_2^{(0)}$	0.30	$\tilde{T}_1^{(1)}DT_2^{(0)}$	0.25
$T_2^{(0)\dagger}DT_2^{(1)}$	−0.01	$\tilde{T}_2^{(0)}DT_2^{(1)}$	−0.01
$T_2^{(1)\dagger}DT_2^{(0)}$	−0.01	$\tilde{T}_2^{(1)}DT_2^{(0)}$	−0.02
Others	0.34	Others	0.08
Contributions to α_d			
$DT_1^{(1)}$	20.43	$DT_1^{(1)}$	20.43
$T_1^{(1)\dagger}D$	20.43	$\tilde{T}_1^{(1)}D$	16.72
$T_1^{(1)\dagger}DT_2^{(0)}$	−2.93	$\tilde{T}_1^{(1)}DT_2^{(0)}$	−2.42
$T_2^{(0)\dagger}DT_2^{(1)}$	0.71	$\tilde{T}_2^{(0)}DT_2^{(1)}$	0.67
$T_2^{(1)\dagger}DT_2^{(0)}$	0.71	$\tilde{T}_2^{(1)}DT_2^{(0)}$	0.68
Others	−4.84	Others	−1.86

correlation effects embodied in the higher-order particle-hole excitations that are present in the two aforementioned (R)CC methods but not in the CI + MBPT and the MCDF calculations. Another possible reason for the disagreement could be that, unlike the (R)CC methods, the latter two methods are not size extensive. The polarizability from these methods are also compared with the experimental result [31]. Our RNCCSD result is slightly closer to the central experimental value than that of the RCCSD. The EDM results for both the T-PT and NSM interactions are slightly larger for the RNCCSD method compared to those obtained using the RCCSD method. In Table II, we also compare contributions to d_a for both the T-PT and NSM interactions and α_d from individual terms in the RCCSD and RNCCSD methods. It clearly shows that there are large differences in the contributions between the H.c. terms of the RCC method and their counterparts in the RNCC method. However, the difference in the final results for the two methods given in Table I are negligibly small. Thus, the close agreement between the results for the RCCSD and RNCCSD methods validates each of them. In Table I, we also give contributions from the Breit interaction and important triples excitations evaluated by a perturbative approach. We have also obtained results for a number of optimized basis sets to find the uncertainties in our results due to the incompleteness of our basis functions and have mentioned them in the table. After accounting for all these corrections, we give the final results for all the quantities in Table I.

The agreement between the results of the EDM calculations of ^{199}Hg using both the RCC and RNCC methods implies that the limits for CP violating coupling constants obtained earlier at the nuclear and elementary particle levels are accurate [5,6]. It is also imperative to carry out similar analyses by applying the RNCC method to evaluate the EDMs of ^{171}Yb and ^{225}Ra, where the differences between the calculations have been found to be even larger than that in the case of ^{199}Hg due to the strong electron correlation effects in these atoms. This method is clearly capable of providing accurate theoretical results for the EDMs and electric dipole polarizabilities of closed-shell atoms and would be a valuable tool in the study of fundamental physics involving EDMs and related physical phenomena.

B. K. S. acknowledges financial support from the Chinese Academy of Science (CAS) through the PIFI Fellowship under the project No. 2017VMB0023. Computations for the present work were carried out using the Vikram-100 HPC cluster of the Physical Research Laboratory (PRL), Ahmedabad, India.

*bijaya@prl.res.in
†bpdas.iia@gmail.com

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