Core polarization in coupled-cluster theory induced by a parity and time-reversal violating interaction

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Received 9 October 2007
Published 22 January 2008
Online at stacks.iop.org/JPhysB/41/035005

Abstract

The effects of a parity and time-reversal violating potential, in particular the tensor–pseudotensor electron–nucleus interaction, are studied. We establish that selected terms representing the interplay of these effects and the residual Coulomb interaction in the coupled-cluster method are equivalent to the coupled-perturbed Hartree–Fock (CPHF). We have shown that the normal CPHF diagrams have a one-to-one correspondence in the coupled-cluster theory, but the CPHF pseudo diagrams are present in a subtle way. We have studied the pseudo diagrams in great detail and have shown explicitly their origin in the coupled-cluster theory. This is demonstrated by considering the case of the permanent electric dipole moment of atomic Hg, and our results are compared with the results of an earlier calculation.

1. Introduction

The observation of a non-zero intrinsic electric dipole moment (EDM) of a non-degenerate quantum system is evidence of parity (P) and time-reversal (T) symmetry violations [1, 2]. Between the two, the T violation is of particular interest as it is less understood and has important implications for physics beyond the standard model. In the experiments where the CP violation has been observed so far [3–5], the T violation is inferred [6] by invoking the CPT theorem. However, the observation of an EDM would be a direct evidence of the T violation in nature. Atoms are suitable and promising candidates to measure permanent EDMs due to their sensitivity to the P and T violating phenomena in the nuclear (hadronic), electron–nucleus (semi-leptonic) and electron (leptonic) sectors [7]. In this paper, we study the atomic EDM arising from the tensor–pseudotensor electron–nucleus interactions, which is semileptonic in nature. The coupling constant $C_T$ of this interaction is zero within the standard model, but it is finite in some theories which are extensions of the standard model [8]. The closed shell atoms are sensitive to this interaction due to its dependence on the nuclear spin, and heavier atoms are more sensitive to this interaction since it scales as $Z^2$ [9]. The closed shell atoms on which EDM experiments have been performed to date are $^{199}$Hg [10] and $^{129}$Xe [11–13], and efforts are underway to improve the results. In addition, new experiments are planned for Yb[14, 15] and Ra [16]. The $^{199}$Hg experimental data have been used to provide improved limits on important P, T odd coupling constants at the elementary particle level [10]. These have been extracted by combining atomic calculations [17] and experiments [10]. The coupled-perturbed Hartree–Fock (CPHF) [18] effects are extremely important in the calculation of atomic properties and it is particularly true in the case of the atomic EDMs [19]. It is important to analyse these effects in the framework of an all-order many-body method like the coupled-cluster method [20], one of the most accurate many-body methods for the study of atomic properties [21]. The main thrust of this work is to demonstrate that all the CPHF effects are subsumed in the coupled-cluster method. This is an important step towards improving the accuracy of the existing atomic calculations of closed-shell atomic EDMs, which is necessary for obtaining better limits on the P and T violating coupling constants. A similar study has been done...
for parity non-conservation effects in open shell atoms [22]. This paper is organized as follows: in section 2, we present the theoretical background; in section 3, we discuss the coupled-cluster equations with/without the T–PT interaction; in section 4, we discuss the atomic EDM in the coupled-cluster and the CPHF framework; in section 5 we discuss our results and finally we present our conclusions in section 6.

2. Theoretical background

2.1. Dirac–Fock equation

For heavy atoms, the relativistic effects cannot be neglected and should be incorporated in the atomic Hamiltonian. An approximate relativistic atomic Hamiltonian, appropriate for our calculations, is the Dirac–Coulomb Hamiltonian $H_{DC}$. For an $N$ electron atomic system in atomic units

$$H_{DC} = \sum_{i}^{N} \left( c a_{i} \cdot p_{i} + \beta c^{2} + V_{N}(\vec{r}_{i}) \right) + \sum_{i < j} \frac{1}{r_{ij}},$$

(1)

where $c$ is velocity of light, $\alpha$ and $\beta$ are the Dirac matrices and $\vec{r}_{ij}$ is the separation between the $i$th and $j$th electrons. This Hamiltonian is an approximation to the general atomic Hamiltonian, which consists of additional terms arising from the interaction of electron spin with nuclear spin, interaction between spins of electrons, magnetic interactions, etc. However, these interactions are neglected as their strength is negligible compared to the electron–electron Coulomb interaction and the nuclear potential energy.

The single electron equations are obtained by approximating the two-electron Coulomb interaction by the Dirac–Fock central potential $U_{DF}(\vec{r})$. Then

$$H_{DC} = \sum_{i}^{N} \left[ c a_{i} \cdot p_{i} + \beta c^{2} + V_{N}(\vec{r}_{i}) + U_{DF}(\vec{r}_{i}) \right] + V_{es},$$

(2)

where the residual Coulomb interaction

$$V_{es} = \sum_{i,j \neq i} \frac{1}{r_{ij}} - \sum_{i} U_{DF}(\vec{r}_{i}).$$

The residual Coulomb interaction embodies the non-central or correlation effects, which can be incorporated in the atomic theory calculations as a perturbation. The single electron wavefunctions satisfy the Schroedinger equation

$$[\alpha \cdot \vec{p} + \beta c^{2} + V_{N}(\vec{r}) + U_{DF}(\vec{r})] |\psi_{a}^{0}\rangle = \epsilon_{a}^{0} |\psi_{a}^{0}\rangle,$$

(3)

where $|\psi_{a}^{0}\rangle$ is the single electron wavefunction and $a$ denotes the quantum numbers which specify the wavefunction uniquely and $\epsilon_{a}^{0}$ are the single electron energies. We can group the operators in the equation and rewrite the equation as

$$(t + g^{0} - \epsilon_{a}^{0}) |\psi_{a}^{0}\rangle = 0.$$

(4)

In the above equation, $t = \alpha \cdot \vec{p} + \beta c^{2} + V_{N}(\vec{r})$ and the Dirac–Fock potential $g^{0} |\psi_{a}^{0}\rangle = \sum_{i} \left[ (\psi_{b}^{0} | v | \psi_{a}^{0}) |\psi_{a}^{0}\rangle - (\psi_{a}^{0} | v | \psi_{b}^{0}) |\psi_{b}^{0}\rangle \right]/(\vec{r}_{i} - \vec{r}_{j})$, where $v = 1/(\vec{r}_{i} - \vec{r}_{j})$. $\vec{r}_{i}$ being the position coordinate of the $i$th electron and the summation is over all the occupied orbitals in the reference state. If an atom has a non-zero EDM, then it is an indication of P and T violating interactions within the atom. In this paper, we consider the atomic EDM arising from the tensor–pseudotensor P and T violating electron–nuclear interaction:

$$H_{T-PT} = i 2 \sqrt{2} C_{T} G_{F} \beta \alpha \cdot I \rho(\vec{r}).$$

(5)

where $G_{F}$ and $\rho(\vec{r})$ are the Fermi coupling constant and nuclear density respectively, $\beta$ and $\alpha$ are the Dirac matrices and $I$ is the nuclear spin. This interaction perturbs $g^{0}$ and the orbitals acquire an admixture from the opposite parity orbitals. At the single electron level, these effects are incorporated in the CPHF calculations.

2.2. Coupled-perturbed Hartree–Fock

The introduction of the P and T violating interaction, $H_{T-PT}$, as a perturbation modifies the atomic Hamiltonian. The corresponding single electron wavefunctions are the mixed parity states

$$|\tilde{\psi}_{a}\rangle = |\psi_{a}^{0}\rangle + \lambda |\psi_{a}^{1}\rangle,$$

(6)

where $\lambda$ is the perturbation parameter and $|\psi_{a}^{1}\rangle$ is the first-order correction, which is opposite in parity to $|\psi_{a}^{0}\rangle$. However, there is no first-order energy correction as $H_{T-PT}$ is parity odd. Then the perturbed Dirac–Fock equation is

$$\left[ h_{0} + \lambda h_{T-PT} + \sum_{b=1}^{N_{occ}} (\langle \tilde{\psi}_{a} | v | \tilde{\psi}_{b} \rangle - \epsilon_{a}^{0}) \right] |\tilde{\psi}_{a}\rangle = 0.$$

(7)

Selecting terms linear in $\lambda$ and rearranging, we get the CPHF equation:

$$\left( h_{0}^{0} + g^{0} - \epsilon_{a}^{0}\right) |\psi_{a}^{1}\rangle = (-h_{T-PT} - g_{1}) |\psi_{a}^{0}\rangle.$$

(8)

The perturbed Dirac–Fock operator is

$$g_{1} |\psi_{a}^{0}\rangle = \sum_{b=1}^{N_{occ}} \left[ (\psi_{b}^{0} | v | \psi_{a}^{0}) |\psi_{a}^{0}\rangle - (\psi_{a}^{0} | v | \psi_{b}^{0}) |\psi_{b}^{0}\rangle \right] + \langle \tilde{\psi}_{b}^{1} | v | \tilde{\psi}_{a}^{0} \rangle |\tilde{\psi}_{a}^{0}\rangle - \langle \tilde{\psi}_{a}^{1} | v | \tilde{\psi}_{b}^{0} \rangle |\tilde{\psi}_{b}^{0}\rangle.$$

(9)

In the present work, to solve equation (8), we expand $|\psi_{a}^{1}\rangle$ in terms of a complete set of unperturbed opposite parity orbitals. Then, $|\psi_{a}^{1}\rangle = \sum_{b} C_{pa} |\psi_{b}^{0}\rangle$, where $C_{pa}$ are the mixing coefficients. Substituting this in equation (8) and projecting by $|\tilde{\psi}_{b}^{0}\rangle$, we obtain a set of linear algebraic equations

$$C_{pa} (\epsilon_{b}^{0} - \epsilon_{a}^{0}) + \sum_{b q} |\tilde{V}_{pqab} C_{qb}^{*} + \tilde{V}_{pqab} C_{qb}| = 0,$$

(10)

where $\tilde{V}_{pqab} = \langle pq | v | ab \rangle - \langle pq | v | ba \rangle$ and $\tilde{V}_{pqab} = \langle pb | v | qa \rangle - \langle pb | v | qa \rangle$. Another approach to calculate $|\psi_{a}^{1}\rangle$ is to solve equation (8) self-consistently [17]. Hereafter, for brevity, an orbital $|\psi_{a}\rangle$ is represented as $|a\rangle$. Equation (10) can be written as the matrix equation

$$AC = -B,$$

(11)
where \( A_{pa} = \sum_{bq} \left( V_{pqab} + \tilde{V}_{pqab} \right) \left( \epsilon_p - \epsilon_q \right) \delta_{pq} \delta_{ab} \) and \( B_{pa} = \langle p | h_{T-PT} | d \rangle \). This equation is solved iteratively starting with the zeroth order in \( v \) contribution

\[
C_{pa}^{(0,1)} = -\frac{B_{pa}}{\epsilon_p - \epsilon_a} \tag{12}
\]

The initial guess. The coefficients in the \( k \)th iteration are

\[
C_{pa}^{(k,1)} = -\frac{B_{pa}}{\epsilon_p - \epsilon_a} - \sum_{bq} \left( \tilde{V}_{pqab} C_{qb}^{(k-1,1)} \right) \frac{\epsilon_p - \epsilon_q}{\epsilon_p - \epsilon_a} \tag{13}
\]

The superscripts in \( C_{pa}^{(k,1)} \) refer to the order of \( v \) and \( h_{T-PT} \) respectively. The diagrams arising from the above equation in zero and one order of \( v \) are shown in Figure 1. In this paper, the diagrams of the second and the third terms in equation (13) are referred to as the pseudo and normal diagrams respectively.

### 3. Coupled-cluster equations

In the coupled cluster theory, the exact atomic state

\[
|\Psi\rangle = e^{-T_0} |\Phi_0\rangle, \tag{14}
\]

where \( T_0 \) is the cluster operator and \(|\Phi_0\rangle\) is the reference state. For the ground state, \(|\Phi_0\rangle\) is the Slater determinant of all the occupied orbitals. The cluster operator \( T_0 = \sum T_i^{(0)} \), where \( T_i^{(0)} \) are the \( i \)-tuple excitation operators. The cluster amplitude equations are obtained from, after applying the operator \( e^{-T_0} \) and projecting on excited states, the Schroedinger equation of \(|\Psi\rangle\). Restricting to the approximation \( T_0 = T_1^{(0)} + T_2^{(0)} \), the cluster operators are hence solutions of the equation \( \langle \Phi^* | [\overline{H}_N, T_1^{(1)}] | \Phi_0 \rangle = 0 \), where \(|\Phi^\ast\rangle\) denotes singly and doubly excited states \(|\Phi_{a1}\rangle\) and \(|\Phi_{a2}\rangle\) respectively. For any operator \( O, \overline{O} = e^{-T_0} O e^{T_0} \) is the dressed operator. It is to be noted that \( H_N \) is the normal-ordered atomic Hamiltonian, which is \( H_{NC} \) in the present calculations. Let the \( H_{T-PT} \) perturbed atomic state

\[
|\Psi\rangle = e^{-T_0+\lambda T_1^{(1)}} |\Phi_0\rangle, \tag{15}
\]

where \( T_1^{(1)} \) is the \( H_{T-PT} \) perturbed cluster operator and \( \lambda \), as defined earlier, is the perturbation parameter. The perturbed coupled cluster equations are

\[
\langle \Phi^* | [\overline{H}_N, T_1^{(1)}] | \Phi_0 \rangle = -\langle \Phi^* | \overline{H}_N^{T-PT} | \Phi_0 \rangle, \tag{16}
\]

Consider \( T_1^{(1)} = T_1^{(1)} \); the equation for singles from equation (16) is

\[
\langle \Phi^* | [\overline{H}_N T_1^{(1)}] | \Phi_0 \rangle = -\langle \Phi^* | \overline{H}_N^{T-PT} | \Phi_0 \rangle. \tag{18}
\]

The cluster operator \( T_1^{(1)} = \sum \alpha_{a_1} a_{a_1} \lambda_{a_1} \) and \( \lambda_{a_1} \) is the associated cluster amplitude. Retaining only \( H_{N-PT} \) from \( \overline{H}_N^{T-PT} \), in terms of matrix elements

\[
\sum_{bq} \tilde{V}_{pqab} (\epsilon_p - \epsilon_q) t_a^{(1)} = -B_{pa}. \tag{19}
\]

Then the perturbed cluster amplitudes \( t_a^{(1)} \) are solutions of the iterative equation

\[
t_a^{(k,1)} = -\frac{B_{pa}}{\epsilon_p - \epsilon_a} - \sum_{bq} \left( \tilde{V}_{pqab} t_a^{(k-1,1)} \right) \frac{\epsilon_p - \epsilon_q}{\epsilon_p - \epsilon_a}. \tag{20}
\]

This is equivalent to equation (13), the equation of the CPHF mixing coefficients without the pseudo diagrams. This formally establishes that the normal diagrams in the CPHF approach are equivalent to a subset of terms in the coupled-cluster theory. However, a similar comparison of the pseudo diagrams in the CPHF and the coupled-cluster theories is done later as it requires the dressed electric dipole operator \( \overline{T_D} \) defined in the following section through the atomic EDM.
4. Atomic EDM

4.1. General expression

The atomic EDM in the CPHF approximation is

\[
D_a = \sum_{ap} \langle a | d | p \rangle C_{pa}^{(\infty,1)} + C_{pa}^{(\infty,1)} \langle p | d | a \rangle.
\]

(21)

where \( d \) is the single particle electric dipole operator and the first superscript on the mixing coefficients refers to all order in \( v \). The diagrams which contribute to \( D_a \) are shown in figure 2. The mixing coefficients in equation (13) have \( v \) to all orders in the limit \( k \to \infty \). Substituting the expression of \( C_{pa}^{(\infty,1)} \), we have

\[
D_a = -2 \sum_{ap} \langle a | d | p \rangle C_{pa}^{(\infty,1)} \left[ B_{pa} + \sum_{bq} \left( V_{pqab} C_{qb}^{(1,1)} \right) \right].
\]

(22)

In the coupled-cluster theory, the atomic EDM

\[
D_a = \langle \Phi_0 | T^{(1)} \mathcal{S} + \mathcal{D} T^{(1)} | \Phi_0 \rangle.
\]

(23)

It should be noted that \( D_a \) has linked terms only [23]; the unlinked terms cancel the normalization factor. For the normal diagrams, the two terms in equation (23) are equivalent and arise from \( D(T_{eff}^{(1)}) \). Here, \( T_{eff}^{(1)} \) is the contraction of \( T_1^{(1)} \) with \( v \); see figure 1 (iv),(v).

4.2. Pseudo diagrams

Figures 2 (ii) and (iii) are the sum of two many-body perturbation theory diagrams. We now show that in the coupled-cluster theory, these are subsumed in \( D_a = \langle \Phi_0 | \left[ DT_{eff}^{(1)} + T_1^{(1)} DT_2^{(0)} \right] | \Phi_0 \rangle \). Unlike in normal terms, here, \( T_{eff}^{(1)} = H_{T\cdot\mathcal{P}} T_2^{(0)} \). The operator \( T_2^{(0)} \) has correlation effects arising from the two-particle and two-hole as well as the other forms of \( v \), to all orders. Hence, the CPHF pseudo diagrams do not have a one-to-one correspondence with the above CCEDM terms; rather the pseudo diagrams are part of terms in CCEDM.

The algebraic expressions of the EDM diagrams shown in figure 2 (ii), (iii) are

\[
\frac{\langle a | D | p \rangle \langle pq | v | ab \rangle \langle b | h_{T\cdot\mathcal{P}} | q \rangle}{(\epsilon_p - \epsilon_a)(\epsilon_b - \epsilon_q)} \tag{24}
\]

\[
\frac{\langle a | D | p \rangle \langle pq | v | ba \rangle \langle b | h_{T\cdot\mathcal{P}} | q \rangle}{(\epsilon_p - \epsilon_a)(\epsilon_b - \epsilon_q)}, \tag{25}
\]

On simplification, this is same as equation (24) multiplied by a phase \( (-1) \). Similarly, the exchange diagram, figure 2 (iii), is the sum of the topologically equivalent MBPT diagrams of figure 3 I(a) and I(b); algebraically

\[
\frac{\langle a | D | p \rangle \langle pq | v | ab \rangle \langle b | h_{T\cdot\mathcal{P}} | q \rangle}{\epsilon_a + \epsilon_b - \epsilon_p - \epsilon_q} \left[ \frac{1}{\epsilon_a - \epsilon_p} + \frac{1}{\epsilon_b - \epsilon_q} \right]. \tag{26}
\]

which is equivalent to equation (25), apart from a phase \( =(-1) \). To calculate the cluster amplitudes which have similar correlation effects, retain \( H_{T\cdot\mathcal{P}}^{(0)} \) and \( H_{T\cdot\mathcal{P}} T_2^{(0)} \) in equation (18). Then, the cluster amplitude equation is

\[
T_{a}^{(2)(k,1)} = -B_{a}^{(k,1)} - \sum_{bq} V_{pqab} T_{bq}^{(k-1,1)}, \tag{28}
\]

where \( B_{a}^{(k,1)} = (H_{T\cdot\mathcal{P}} + H_{T\cdot\mathcal{P}} T_2^{(0)}) | a \rangle \). The cluster amplitudes calculated from the above equation have effects of direct and exchange pseudo diagrams.

To analyse the atomic EDM arising from the pseudo diagrams within the coupled-cluster theory, consider the two terms \( DT_{eff}^{(1)} \) and \( T_1^{(1)} DT_2^{(0)} \). After introducing a complete set of eigenfunctions, the contribution to \( D_a \) from these terms is

\[
D_a = \sum_{I} \left[ \langle \Phi_0 | D | \Phi_I \rangle | \Phi_I | T_1^{(1)} | \Phi_0 \rangle \right. \tag{29}

\[
+ \sum_{I} \left. \langle \Phi_0 | T_1^{(1)} | \Phi_I \rangle | \Phi_I | D | \Phi_0 \rangle | \Phi_I | T_2^{(0)} | \Phi_0 \rangle \right].
\]

The first term on the right-hand side, in terms of the determinantal states, is

\[
\sum_{a,p} \langle \Phi_0 | D | \Phi_p \rangle | \Phi_a | T_1^{(1)} | \Phi_0 \rangle; \tag{30}
\]

this follows as \( D \) and \( T_1^{(1)} \) are single particle operators. It is to be mentioned that \( T_1^{(1)} \) represents the component of \( T_1^{(1)} \) arising from the term \( (H_{T\cdot\mathcal{P}} T_2^{(0)}) \) in equation (28). Similarly, the second term in equation (29) is

\[
\sum_{a,p,bq} \langle \Phi_0 | T_1^{(1)} | \Phi_p | \Phi_a | D | \Phi_{pq} | \Phi_{ab} | T_2^{(0)} | \Phi_0 \rangle. \tag{31}
\]
Table 1. Variation of $D_a$ with the inclusion of higher angular momentum virtual states. The first column implies that virtuals only up to the orbitals indicated have been included in the calculation, in addition to $s_{1/2}$ and $p_{1/2}$ symmetries. The normal and the two pseudo diagrams are calculated independently.

| Virtual states | EDM ($10^{-22}$ cm) |
|----------------|----------------------|
| Up to $p_{1/2}$ | $-6.411$ $-6.024$ |
| Up to $d_{5/2}$ | $-6.415$ $-6.127$ |
| Up to $f_{7/2}$ | $-6.399$ $-6.142$ |
| Up to $g_{9/2}$ | $-6.399$ $-6.144$ |

Table 2. Variation of $D_a$ with the inclusion of higher angular momentum virtual states. The first column implies that virtuals only up to the orbitals indicated have been included in the calculation, in addition to $s_{1/2}$ and $p_{1/2}$ symmetries. The normal and the two pseudo diagrams are calculated together.

| Virtual states | EDM ($10^{-22}$ cm) |
|----------------|----------------------|
| Up to $p_{1/2}$ | $-5.83$ |
| Up to $d_{5/2}$ | $-5.90$ |
| Up to $f_{7/2}$ | $-6.75$ |
| Up to $g_{9/2}$ | $-6.75$ |

In this expression, the pseudo diagram contributions are present in the component $T_1^{(1)}$ arising from the term $\langle H_{FT-PR} \rangle \rho$ in equation (28). Using the Slater–Condon rules, the matrix elements of single and two-particle operators between determinantal states are $\langle \Phi_{a}^{\mu} | D | \Phi_{b}^{\nu} \rangle = \langle p | D | a \rangle \langle \Phi_{a}^{\mu} | T_0^{(2)} | \Phi_{b}^{\nu} \rangle = \langle pq | t_2 | ab \rangle - \langle pq | t_2 | ba \rangle$ and $\langle \Phi_{a}^{\mu} | D | \Phi_{b}^{\nu} \rangle = \langle b | D | q \rangle$. Then,

$$D_a = \sum_{a,p} \langle p | D | a \rangle \langle T_1^{(1)} | \Phi_a \rangle + \sum_{a,p} \sum_{b,q} t_{pq}^{(1)} \langle a | D | p \rangle [\langle pq | t_2 | ab \rangle - \langle pq | t_2 | ba \rangle].$$

The effects of the pseudo diagrams are distributed among various terms with the $T_2^{(0)}$ cluster amplitude. Hence, it is difficult to establish one-to-one correspondence between the CPHF pseudo diagrams and the corresponding diagrams in CCEDM in orders of $v$. This is a consequence of the structure of the CCEDM and CPHF equations where the perturbed cluster amplitudes are computed using the converged values of the $T_0^{(2)}$ amplitudes, which treat the residual Coulomb interaction to all orders. However, the converged results which include all orders of $v$ and one order of $H_{FT-PR}$, where the sequence of the perturbations has all possible combinations, should be identical. It is possible to establish the equivalence by choosing only the two-particle two-hole terms of $v$ in the $T_0^{(2)}$ equations.

In this paper, we calculate the normal and the pseudo diagrams simultaneously; this couples the normal and pseudo contributions. This is evident from the equation (13), where the CPHF coefficients are iterated with both the normal and the pseudo diagrams. But within the coupled-cluster theory, in particular the CCEDM formalism, this inclusion is more subtle. This is due to the structure of equation (23), where the converged $T_1^{(1)}$ amplitudes contain the effects of the EDM from both the terms shown in equation (28).

Table 3. Dominant contributions to $D_a = T_1^{(1)}D + T_1^{(1)}T_2^{(0)}$ (in units of $10^{-22}C_T\sigma_N$ cm) from the terms shown in equation (28) for np intermediate states calculated using the coupled-cluster theory for EDMs.

| Occ. | np | $T_1^{(1)}$ | $D$ | $T_1^{(1)}D$ |
|------|----|------------|-----|-------------|
| 6s_{1/2} | 6p_{1/2} | 104.69 | 0.872 | -1.010 |
| 6s_{1/2} | 7p_{1/2} | -254.88 | -1.821 | -5.139 |
| 6s_{1/2} | 8p_{1/2} | 262.28 | 1.388 | -4.032 |
| 6s_{1/2} | 9p_{1/2} | -202.37 | -0.344 | -0.771 |
| 6s_{1/2} | 10p_{1/2} | -113.94 | 0.068 | 0.858 |
| 6s_{1/2} | 11p_{1/2} | -56.22 | 0.692 | 0.843 |
| 6s_{1/2} | 6p_{1/2} | 13.85 | 0.995 | 0.153 |
| 6s_{1/2} | 7p_{1/2} | -36.27 | -2.372 | 0.953 |
| 6s_{1/2} | 8p_{1/2} | -36.80 | -2.211 | 0.901 |
| 6s_{1/2} | 9p_{1/2} | 15.91 | 0.771 | 0.0135 |

Total $-8.2026$

Table 4. Dominant contributions to $D_a$ (in units of $10^{-22}C_T\sigma_N$ cm) from the term $DT_2^{(0)}$.

| Occ. | np | $DT_2^{(0)}$ (Atomic units) | $D_a$ ($10^{-22}C_T\sigma_N$ cm) |
|------|----|---------------------------|-------------------------------|
| 6s_{1/2} | 6p_{1/2} | 0.0926 | -1.077 | -0.011 |
| 6s_{1/2} | 7p_{1/2} | -0.238 | 0.269 | -0.067 |
| 6s_{1/2} | 8p_{1/2} | 0.274 | -0.287 | -0.080 |
| 6s_{1/2} | 9p_{1/2} | -0.248 | 0.209 | -0.056 |
| 6s_{1/2} | 10p_{1/2} | -0.108 | 0.056 | -0.014 |
| 6s_{1/2} | 11p_{1/2} | -0.002 | 0.0009 | -0.00013 |
| 6s_{1/2} | 6p_{1/2} | -0.088 | 0.0027 | -0.0014 |
| 6s_{1/2} | 7p_{1/2} | 0.264 | 0.435 | -0.011 |
| 6s_{1/2} | 8p_{1/2} | 0.362 | -0.0057 | -0.015 |
| 6s_{1/2} | 9p_{1/2} | -0.346 | 0.0324 | -0.0061 |

Total $-0.2597$ 0.226

5. Results

5.1. Symmetry-wise contribution

From the expression of $H_{FT-PR}$, the presence of nuclear density $\rho(r)$, s_{1/2}–p_{1/2} is expected to have the largest contribution. This is indeed observed in our calculations. The variations of $D_a$ for a small basis consisting of 68 Gaussian-type orbitals [24], (1–12)s_{1/2}, (2–13)p_{1/2}, (3–10)d_{3/2}, (4–7)f_{5/2}, (5–8)g_{7/2}, are given in table 1. It lists the value of $D_a$ when orbitals are added symmetrically. According to the table $D_a$, the contribution from the higher angular momentum $d$ and $f$ virtual orbitals, are small and opposite in phase. Next, we consider an optimal basis set with which we get converged on $D_a$; it consists of 112 Gaussian-type orbitals: (1–18)s_{1/2}, (2–18)p_{1/2}, (3–16)d_{3/2}, (4–13)f_{5/2}, (5–10)g_{7/2},. The results from the optimal basis are given in table 2. It lists $D_a$ arising from the normal and pseudo, where these are calculated simultaneously. It can be seen that the contribution from the virtual orbitals enhances $D_a$ and changes the value $-5.83 \times 10^{-22}$ cm to $-6.75 \times 10^{-22}$ cm.

For a more detailed analysis, the dominant contributions from 6s_{1/2}–p_{1/2} and 6s_{1/2}–p_{1/2} are listed in table 3. The total
contribution from $6s_{1/2}-np_{1/2,3/2}$ from the term $T_{1}^{(1)}$ is $-0.337 \times 10^{-22}$ em and from the term $DT_{1}^{(1)}$ is $-8.20 \times 10^{-22}$ em. The contribution to $D_0$ from the term $DT_{2}^{(0)}$ is given in table 4. The individual contributions from the direct and the exchange diagrams of $DT_{2}^{(0)}$ are also presented in table 4.

5.2. Cluster amplitudes and $D_0$

The calculated normal $T_{1}^{(1)}$ amplitudes are in excellent agreement with the corresponding CPHF mixing coefficients. Calculating the normal and the pseudo diagrams together, $D_0$ of atomic Hg is $-6.75 \times 10^{-22}$ em. It is enhanced to $-6.92 \times 10^{-22}$ em when the two pseudo terms are calculated separately. A previous calculation [17] reported the CPHF $D_0$ of atomic Hg as $-6.0 \times 10^{-22}$ em. We attribute the difference of our result from the previous calculation to the inclusion of correlation effects in CC, beyond those present in the CPHF, which are discussed in section 4. The different numerical methods used can also contribute to the discrepancy; however this would be small. The different results, when the pseudo terms are calculated together and separately with the normal terms, are the effect of the coupling between the two terms. Comparing the contributions from the normal and the pseudo diagrams, the pseudo diagram contribution, though important, is just ~9% of the normal diagram contribution. The portion of the pseudo diagram contribution is however dependent on the size of the basis set. For example, with the basis $(1–14)s_{1/2}, (2–14)p_{1/2,3/2}, (3–12)d_{1/2,5/2}, (4–8)f_{5/2,7/2}$ and $(5–9)g_{7/2,9/2}$, the contribution from the pseudo diagrams is 4%.

This indicates that the contribution from the pseudo diagrams increases till it converges. The phase of the normal diagrams is determined by the phase of the most dominant term, the Dirac–Fock contribution. For $^{100}$Hg, this is negative. On the other hand, the phase of the pseudo diagrams cannot be ascertained easily. Unlike the normal terms, the leading contribution from pseudo diagrams has $T_{2}^{(0)}$ which contributes to two terms – $H_{T}–\pi T_{2}^{(0)}$ and $DT_{2}^{(0)}$. The phases of the dominant $H_{T}–\pi T_{2}^{(0)}$ and $DT_{2}^{(0)}$ contributions determine the overall phase of the pseudo diagram. That is, the relative phase of the normal and pseudo diagrams is not a general trend. It depends on the phase of the dominant $T_{2}^{(0)}$ cluster amplitudes and hence it is atom specific.

6. Conclusion

In this paper, we have numerically tested and demonstrated the equivalence of the CPHF diagrams based on an analysis using many-body perturbation theory. The pseudo diagrams are the sum of two many-body perturbation theory diagrams [19]. Hence, in the coupled-cluster expression of $D_0$ in equation (23), the direct and the conjugate terms when added give exactly the pseudo diagrams of the CPHF. This shows that the coupled-cluster theory contains all the CPHF effects. The relative phases of the pseudo and the normal diagrams are atom specific and hence cannot be generalized. An optimal basis consisting of 112 orbitals gives convergent result and is in good agreement with the result of Martensson-Pendrill [17]. We have studied and analysed in detail the various many-body effects that play an important role in the EDM of atomic Hg and impact of coupling between normal and pseudo diagrams.

Acknowledgments

We acknowledge Chiranjib Sur, Dmitry Budker and K P Geetha for discussions at various stages of the work.

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