Probing *CP* Violation with the Electric Dipole Moment of Atomic Mercury

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The electric dipole moment of atomic ^{199}Hg induced by the nuclear Schiff moment and the tensor-pseudotensor electron-nucleus interactions are calculated. For this, we develop and employ a novel method based on the relativistic coupled-cluster theory. The results of our theoretical calculations, combined with the latest experimental result of the ^{199}Hg electric dipole moment, provide new bounds on the T reversal or CP violation parameters $\theta_{\rm QCD}$, the tensor-pseudotensor coupling constant C_T, and $(\tilde{d}_u - \tilde{d}_d)$. This is the most accurate calculation of these parameters to date. We highlight the crucial role of electron correlation effects in their interplay with the P, T violating interactions. Our results demonstrate substantial changes in the results of earlier calculations of these parameters which can be attributed to the more accurate inclusion of important correlation effects.

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The existence of a nonzero permanent electric dipole moment (EDM) of a nondegenerate physical system is a signature of the simultaneous violations of parity (P) and time-reversal (T) symmetries [1]. T violation implies the combined charge conjugation (C) and P violation (CP violation) via the CPT theorem [2]. The origin of CP violation is still not well understood. It has so far been observed only in the K [3] and B [4,5] mesons, and the results are essentially in agreement with the predictions of Kobayashi and Maskawa in the framework of the standard model [6]. However, this model cannot explain the matter-antimatter asymmetry [7] in the Universe for which CP violation is a prerequisite [8]. In addition, it predicts atomic EDMs several orders of magnitude below their current limits [9–11]. Indeed, atomic EDMs are excellent probes of physics beyond the standard model [9–11], and they provide important insights into a rich variety of CP violations—leptonic, semileptonic, and hadronic sectors. Experimental searches are underway for the EDMs of paramagnetic (open-shell) and diamagnetic (closed-shell) atoms [12,13]. The results of the experiments can be combined with those of sophisticated atomic many-body calculations to determine various CP violating coupling constants at the atomic level. These can ultimately be related to the CP violation parameters at the elementary particle level [14]. The EDM of diamagnetic atoms arises predominantly from the nuclear Schiff moment (NSM) and/or the electron-nucleon tensor-pseudotensor interactions [14]. These in turn arise from the nucleon-nucleon interactions or the EDM of nucleons, which originate due to the quark-quark interactions, EDMs, and chromo-EDMs of quarks at the elementary particle level.

In this Letter, we concentrate only on the EDM of mercury (^{199}Hg), a closed-shell atom. The present limits on important CP violation parameters like $\theta_{\rm QCD}$ for strong interactions and the chromo-EDMs of quarks have been obtained from the EDM of ^{199}Hg [12], which is the most accurate of all the data from atomic EDMs to date. The focus of our work in this Letter is to improve the current limits on the coupling constants associated with the electron-nucleon tensor-pseudotensor (T-PT) interaction (C_T) and the NSM(S). A nonzero value of C_T implies physics beyond the standard model. The dependence of the T-PT interactions and the NSM on the nuclear spin makes closed-shell atoms, in particular, those having nonzero nuclear spin, the best candidates to measure EDMs sensitive to the nuclear sector.

For heavy atoms like mercury, it is customary to use the Dirac-Coulomb Hamiltonian, $H_{\rm DC}$, in atomic units

$$H_{\rm DC} = \sum_i [c\boldsymbol{\alpha}_i \cdot \boldsymbol{p}_i + \boldsymbol{\beta}_i mc^2 + V_N(r_i)] + \sum_{i<j} \frac{1}{|\boldsymbol{r}_i - \boldsymbol{r}_j|}, \quad (1)$$

where r_i refers to the electron coordinates; α and β, the Dirac matrices; and $V_N(r_i)$, the nuclear potential, and the last term is the electron-electron Coulomb interaction. The key and the most challenging step in atomic many-body physics is to incorporate the effects of the electron-electron Coulomb interaction [last term in Eq. (1)] as accurately as possible. Under the independent particle and the central field approximations [15], with the introduction of the Dirac-Fock potential, the above Hamiltonian can be separated into an exactly solvable part (H_0) and a residual interaction part which consists of the Coulomb interaction and the Dirac-Fock potential [15]. The single particle wave functions are computed self-consistently from H_0, and the many particle wave functions are expressed as Slater de-

terminants built out of the single particle wave functions. The effects of the residual Coulomb interaction are calculated with many-body methods. For this, the finite order many-body perturbation theory (MBPT) and the configuration interaction (CI) approach are two widely used methods [15]. An even superior method, with strong theoretical many-body physics underpinnings is the coupled-cluster theory. In this Letter, the P and T violating interactions are treated perturbatively to first order. This is in addition to the electron-electron residual Coulomb interaction within the framework of the relativistic coupled-cluster theory.

The wave function in coupled-cluster theory has an exponential structure (see [16] for a recent review of the method) to describe correlation effects in many-body systems. It is nonperturbative and defines atomic states as a superposition of states of different levels of excitations, with respect to a reference state. These arise from the residual Coulomb interactions.

Mathematically, the coupled-cluster wave function can be expressed as

$$|\Psi_i\rangle = e^{T^{(0)}}|\Phi_i\rangle, \tag{2}$$

where $|\Phi_i\rangle$ is the reference state containing a fixed number of electrons and $T^{(0)}$ is an operator which excites electrons out of it, thereby giving rise to states with different levels of excitations corresponding to different many-body effects. In our calculations we use the coupled-cluster singles and doubles approximation, that is, $T^{(0)} = T_1^{(0)} + T_2^{(0)}$. In second quantized form

$$T_1^{(0)} = \sum_{a,p} a_p^\dagger a_a t_a^p(0) \quad \text{and} \quad T_2^{(0)} = \sum_{a,b,p,q} a_p^\dagger a_q^\dagger a_b a_a t_{ab}^{pq}(0)$$

excite one and two electrons, respectively, from the reference state, where $t_a^p(0)$ and $t_{ab}^{pq}(0)$ are the respective amplitudes. The equations that determine the amplitudes of $T^{(0)}$ are a set of coupled nonlinear algebraic equations, and these are solved iteratively till convergence.

For closed-shell atoms, as mentioned earlier, one prominent source of EDMs is the nuclear Schiff moment $\boldsymbol{S}$, a P and T odd electromagnetic moment of the nucleus [17]. For a finite size nucleus of radius R_N, the Schiff moment potential [18] is

$$\varphi(\boldsymbol{R}) = -\frac{3\boldsymbol{S}\cdot\boldsymbol{R}}{B}\rho_N(R), \tag{3}$$

where $B = \int \rho_N(R)R^4 dR$ and $\rho_N(R)$ is the nuclear density. This potential interacts electrostatically with the electrons and mixes atomic states of opposite parities to generate a finite atomic EDM, d_A. Then the atomic Hamiltonian is $H_{\text{atom}} = H_{\text{DC}} + H_{\text{PTV}}(\lambda)$, where $H_{\text{PTV}}^{\text{Schiff}}(\boldsymbol{S}) = -\varphi(\boldsymbol{R})$ is the P and T violating interaction Hamiltonian and λ is a T or CP violation parameter which can be considered as a perturbation parameter. The eigenstates of the H_{atom} are the mixed parity states $|\tilde{\Psi}\rangle$. To incorporate H_{PTV} as a first order perturbation, the exponential operator in coupled-cluster theory is redefined as $e^{T^{(0)}+\lambda T^{(1)}}$. The cluster operator $T^{(1)}$ has one order of H_{PTV} and mixes the states of opposite parities. As a result of this, the ground state is

$$|\tilde{\Psi}_0\rangle = e^{T^{(0)}+\lambda T^{(1)}}|\Phi_0\rangle. \tag{4}$$

Then, as H_{PTV} is considered to first order only, the equations for the amplitudes of $T^{(1)}$ are a set of linear algebraic equations,

$$\langle\Phi_0'|[\bar{H}_N, T^{(1)}]|\Phi_0\rangle = -\langle\Phi_0'|\bar{H}_{\text{PTV}}|\Phi_0\rangle, \tag{5}$$

$\bar{O} = e^{T^{(0)\dagger}} O e^{T^{(0)}}$ where O is a general operator, H_N is the normal-ordered Hamiltonian, and $|\Phi_0'\rangle$ are opposite parity Slater determinants. Further, as in the unperturbed cluster operators $T^{(0)}$, we use the approximation $T^{(1)} = T_1^{(1)} + T_2^{(1)}$. Then the atomic EDM of the ground state is

$$d_A = \frac{\langle\tilde{\Psi}_0|D|\tilde{\Psi}_0\rangle}{\langle\tilde{\Psi}_0|\tilde{\Psi}_0\rangle}, \tag{6}$$

where D is the electric dipole operator. In the above expression, after expanding in terms of the cluster operators $T^{(0)}$ and $T^{(1)}$, only the terms first order in $T^{(1)}$ contribute. Often, d_A is computed perturbatively with the sum over states approach, which necessitates a truncation after the first few intermediate states. On the contrary, our relativistic coupled-cluster scheme does not involve summing over states and subsumes all possible intermediate states within the chosen configuration space.

Besides the NSM, the other dominant source of EDM in closed-shell atoms is the tensor-pseudotensor electron-nucleus interaction

$$H_{\text{PTV}}^{\text{T-PT}}(C_T) = \frac{iG_F C_T}{\sqrt{2}}\sum_i \boldsymbol{\sigma}_N\cdot\boldsymbol{\gamma}_i\rho_N(r), \tag{7}$$

where G_F is a Fermi constant, C_T is a coupling constant, $\boldsymbol{\sigma}_N$ is a nuclear spin, and $\boldsymbol{\gamma}_i$ is a Dirac matrix. It must be emphasized that this form of interaction does not exist within the standard model of particle physics as C_T is zero. However, there are models which predict such an interaction [14].

To extract the T or CP violation parameters, the atomic theory calculations are combined with the experimental data. In this context it is appropriate to rewrite Eq. (6) as

$$d_A = \lambda\eta, \tag{8}$$

where η is the atomic enhancement factor. As defined earlier, the constant λ is a T or CP violation parameter considered as a perturbation parameter. It can, for example, be the nuclear Schiff moment $\boldsymbol{S}$ or the coupling constant C_T. A precision atomic many-body calculation, like the coupled-cluster calculation reported here, would provide the value for a particular η. Experimentally, the measured atomic EDM d_A is the sum total of contributions from all the P and T symmetry violating phenomena within the

atom. A bound on $\lambda = d_A/\eta$ is obtained by combining the results of atomic theory and experimental data. Depending on the choice of the atom, it is possible to identify the dominant sources of T or CP violation and derive tighter bounds.

For the present set of calculations, we employ the even-tempered Gaussian basis set expansion [19,20]. The orbital basis set consist of (1–18)s, (2–18)$p_{1/2,3/2}$, (3–13)$d_{3/2,5/2}$, (4–11)$f_{5/2,7/2}$, (5–9)$g_{7/2,9/2}$, and (6–7)$h_{9/2,11/2}$. This orbital basis set is considered complete for the coupled perturbed Hartree-Fock (CPHF) calculations. That is, further increase in the number of orbitals does not change the results. In addition, we compute the ground state dipole scalar polarizability for ^{199}Hg. We obtain a value $33.294ea_0^3$, where a_0 is the Bohr radius, which is in excellent agreement with the experimental value 33.91 ± 0.34 [21].

To date, among the closed-shell atoms, ^{199}Hg, as mentioned earlier, sets the standard for the most precise EDM results. In a recent paper [12], the new upper limit is reported as

$$|d(^{199}\mathrm{Hg})| < 3.1 \times 10^{-29} e \text{ cm (95\% C.L.)}. \quad (9)$$

Our atomic calculation based on the relativistic coupled-cluster theory gives

$$d_A^{\mathrm{Schiff}}(^{199}\mathrm{Hg}) = -5.07 \times 10^{-17}\left(\frac{S}{e \text{ fm}^3}\right) e \text{ cm}. \quad (10)$$

This is the first ever relativistic coupled-cluster result for any atomic EDM calculation arising from the NSM. Combining with the experimental result, the limit on the NSM is

$$S(^{199}\mathrm{Hg}) < 6.1 \times 10^{-13} e \text{ fm}^3. \quad (11)$$

There is a large deviation of 96% between the combined results of the CPHF method and CI + MBPT [22],

$$d_A^{\mathrm{Schiff}}(^{199}\mathrm{Hg}) = -2.8 \times 10^{-17}\left(\frac{S}{e \text{ fm}^3}\right) e \text{ cm}, \quad (12)$$

and our result obtained using the relativistic coupled-cluster theory. At the level of CPHF, our result [23] is in good agreement with that of Ref. [22]. The large difference in the final results of two the calculations demonstrates the importance of electron correlation effects and their interplay with the $H_{\mathrm{PTV}}^{\mathrm{Schiff}}$ interaction in determining the magnitude of the NSM. We have used a larger basis set in our calculation than Dzuba *et al.* Unlike them, we have taken into account all possible intermediate states that arise within the chosen basis in the framework of the relativistic coupled-cluster singles and doubles.

It is possible to separate the contributions of individual terms in Eq. (6). The earlier calculations [22,24] incorporate only a certain class of two-particle two-hole excitations which are subset of the correlation effects we have included through the cluster operator $T^{(0)}$ in the present calculation. The dominant contribution to the EDM of atomic 199Hg comes from the term $T_1^{(1)\dagger} D$, followed by that of $T_1^{(1)\dagger} D T_2^{(0)}$. In Eq. (10) these terms individually contribute $-5.40 \times 10^{-17}[S/(e \text{ fm}^3)]e$ cm and $-0.17 \times 10^{-17}[S/(e \text{ fm}^3)]e$ cm, respectively.

Our result for ^{199}Hg EDM arising from the electron-nucleus tensor-pseudotensor interaction is

$$d_A^{\text{T-PT}} = -4.3 \times 10^{-20} C_T \sigma_N e \text{ cm}. \quad (13)$$

Compared to the CPHF result $-6.19 \times 10^{-20} C_T \sigma_N e$ cm [23,24], the change with the additional correlation effects is not so dramatic. There is a decrease of 31%, which is significant but not so spectacular as in $d_A^{\mathrm{Schiff}}(^{199}\mathrm{Hg})$. This comparison demonstrates, without any ambiguity, the importance of electron correlation effects in precision atomic EDM calculations. A closer examination of the structure of the two P and T violating Hamiltonians in this Letter sheds some light on why the electron correlation effects are larger in the case of the NSM than the tensor-pseudotensor interaction. The reasons for the observed change are the following. The electron-nucleus P and T violating interaction that induces the NSM is a diagonal operator. In other words, this operator mixes the like components of two relativistic orbitals, i.e., large with large and small with small when its matrix element is evaluated. In contrast, the electron-nucleus tensor-pseudotensor interaction is off-diagonal, and therefore its matrix elements are suppressed by an order (v/c) relative to the NSM. In addition, the matrix elements of the NSM are significant for s and $p_{1/2}$ orbitals and also s and $p_{3/2}$ orbitals. However, the matrix element of the tensor-pseudotensor interaction is sizeable only for s and $p_{1/2}$ orbitals. The existence of the $s - p_{3/2}$ matrix element in the former case results in additional correlation contributions in the NSM induced EDM in comparison to its tensor-pseudotensor counterpart.

The individual contributions of the different terms to tensor-pseudotensor EDM follow a trend similar to that of NSM. The terms $T_1^{(1)\dagger} D$ and $T^{(1)\dagger} D T_2^{(0)}$ give the largest ($\approx$95%) and the second largest contributions to $d_A^{\text{T-PT}}$, $-4.8 \times 10^{-20} C_T \sigma_N e$ cm and $-0.27 \times 10^{-20} C_T \sigma_N e$ cm, respectively. Then, a limit

$$C_T < 1.4 \times 10^{-9}, \quad (14)$$

is obtained after combining our results with the experimental data.

Assuming that the NSM arises from the nucleon-nucleon interactions with pions as the dominant mediators, we get [25]

$$S(^{199}\mathrm{Hg}) = g_{\pi NN}[0.01\bar{g}_{\pi NN}^{(0)} + 0.07\bar{g}_{\pi NN}^{(1)} + 0.02\bar{g}_{\pi NN}^{(2)}]e \text{ fm}^3, \quad (15)$$

where $g_{\pi NN}$ and $\bar{g}_{\pi NN}^{(i)}$ are the CP conserving and CP violating pion-nucleon coupling constants, respectively.

Here, $i = 0$, 1, and 2 represent isoscalar, isovector, and isotensor, respectively. Considering $\bar{g}^{(1)}_{\pi NN}$ as the most dominant, then

$$\bar{g}^{(1)}_{\pi NN} < 6.4 \times 10^{-13}. \quad (16)$$

The coupling constant $\bar{g}^{(1)}_{\pi NN}$ is related to the chromo-EDMs of quarks [26] from the above result:

$$(\tilde{d}_u - \tilde{d}_d) < 3.2 \times 10^{-27} e \text{ cm}. \quad (17)$$

Next, consider the maximum contribution to NSM arising from $\bar{g}^{(0)}_{\pi NN}$; then

$$\bar{g}^{(0)}_{\pi NN} < 4.5 \times 10^{-12}. \quad (18)$$

Since $\bar{g}^{(0)}_{\pi NN} = 0.027\theta_{\text{QCD}}$ [27], we get the bound

$$\theta_{\text{QCD}} < 1.7 \times 10^{-10}. \quad (19)$$

The value we have obtained for the NSM is likely to give the most stringent bounds for supersymmetric *CP* violating phases [9,10,28]. In addition, from our results and the experimental data, it is also possible to set improved limits on the specific *CP* violating parameters predicted by various extensions of the standard model, ϵ_q^{SUSY}, ϵ^{Higgs}, x^{LR} [29].

In conclusion, we have developed a unique relativistic coupled-cluster-based many-body method that takes into account the physical effects arising from the interplay of two very different kinds of fundamental interactions—the *CP* conserving electron-electron Coulomb and *CP* violating electron-nucleus interactions. The results obtained for the EDM of ^{199}Hg by the application of this method and the latest experiment on this atom [12] yield the most accurate limits to date on some important *CP* violating parameters. The electron correlation effects play a critical role in improving the existing limit on these parameters. These limits constrain the possible extensions to the standard model, thereby enhancing our current knowledge of the intriguing phenomenon of *CP* violation.

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