Calculation of the (T,P)-odd electric dipole moment of thallium and cesium

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Parity and time invariance violating electric dipole moment of ^{205}Tl is calculated using the relativistic Hartree-Fock and configuration interaction methods and the many-body perturbation theory. Contributions from the interaction of the electron electric dipole moments with internal electric field and scalar-pseudoscalar electron-nucleon (T,P)-odd interaction are considered. The results are $d(^{205}\mathrm{Tl})=-582(20)d_e$ or $d(^{205}\mathrm{Tl})=-7.0(2)\times10^{-18}C_S^P e$ cm. Interpretation of the measurements are discussed. The results of similar calculations for ^{133}Cs are $d(^{133}\mathrm{Cs})=124(4)d_e$ or $d(^{133}\mathrm{Cs})=0.76(2)\times10^{-18}C_S^P e$ cm.

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I. INTRODUCTION

Recent very sensitive experiment performed in Seattle [1] puts very strong constraint on the electric dipole moment (EDM) of mercury. It now reads $d(^{199}\mathrm{Hg})=(0.49\pm1.29_{\mathrm{stat}}\pm0.76_{\mathrm{syst}})\times10^{-29}$ e cm, which is sevenfold improvement of the previous result of the same group. This renews the interest on the sources of atomic EDMs. In our previous paper [2], we calculated the EDM of mercury and other paramagnetic atoms due to nuclear Schiff moment, (T,P)-odd electron-nucleon interaction and interaction of the electron electric dipole moment (d_e) with nuclear magnetic field. The EDM of mercury due to nuclear Schiff moment was also considered in a recent paper by Latha *et al.* [3]. Other contributions include, e.g., interaction of the electron electric dipole moments with internal electric field and scalar-pseudoscalar electron-nucleon (T,P)-odd interaction. The latter two sources of atomic EDM are strongly suppressed in mercury due to zero total electron momentum, $J=0$. They give rise to EDMs of atoms with closed electron shells only in third order of the perturbation theory, when magnetic dipole hyperfine interaction is also taken into account. The strongest constraint on the strength of these (T,P)-odd interactions came so far from the thallium experiment [4] (see also review [5] for a detailed discussion). However, significant advance in the accuracy of the measurements in mercury [1] has changed the situation. Now, the constraint on the scalar-pseudoscalar electron-nucleon interaction, which comes from mercury EDM measurements is five times stronger than those from thallium measurements while the constraints on the electron EDM in Hg are two times weaker than in Tl [1]. All these results rely on atomic calculations which provide the link between atomic EDMs and the fundamental constants of the (T,P)-odd interactions. Due to significant progress in measurements it is important to revisit the calculations as well for the sake of improving their accuracy and reliability.

The third-order calculations for mercury will be the subject of future work. In present paper we perform the second-order calculations of the EDMs of thallium and cesium caused by the electron EDM and the scalar-pseudoscalar electron-nucleon (T,P)-odd interaction. These effects in Tl and Cs are significantly larger than in Hg since Tl and Cs have nonzero electron angular momentum. The work to improve accuracy of the EDM measurements for cesium is in progress [6]. Our calculations are in good agreement with the most recent and accurate calculations of [7] and consistent with other calculations (see, e.g., review [5]).

However, the main purpose of this work is the calculation of the EDM of thallium. This is due to the facts that the constraints on the parameters of the (T,P)-odd interactions coming from thallium experiment is currently stronger than those from cesium while disagreement between different calculations are larger.

The effect the scalar-pseudoscalar electron-nucleon (T,P)-odd interaction for Tl was considered by Mårtensson-Pendrill and Lindroth [8] and Sahoo *et al.* [9]. The results differ almost two times, which is probably significantly larger than assumed uncertainty of both calculations.

The EDM of thallium caused by electron EDM was considered by many authors [10–15]. The results show strong dependence on electron correlations and change significantly depending on how many correlation terms are included. The most complete calculations were performed by Liu and Kelly [15] using the coupled cluster approach. The result is in relatively good agreement with the semiempirical estimations of [11].

All previous calculations of the thallium EDM treated the thallium atom as a system with one external electron above closed shells. In present paper, we consider it as a three valence electron system by including $6s$ electrons into valence space. We use the configuration interaction technique combined with the many-body perturbation theory (the CI+MBPT [16,17] method). We demonstrate that all instabilities of the results are due to strong correlations between external $6s$ and $6p$ electrons and using the configuration interaction technique to treat these correlations accurately leads to very stable results. We use exactly the same procedure for both (T,P)-odd operators which is another test of the consistency of the calculations. Our final result for the electron EDM is in excellent agreement with the most complete previous *ab initio* calculations by Liu and Kelly [15], while our result for the scalar-pseudoscalar electron-nucleon (T,P)-odd interaction is closer to the result of Mårtensson-Pendrill and Lindroth [8] and differs significantly from [9].

We estimate the uncertainty of present calculations to be about 3%.

II. METHOD OF CALCULATION

The Hamiltonian of the scalar-pseudoscalar electron-nucleon (T,P)-odd interaction can be written as

$$\hat{H}^{SP} = i\frac{G}{\sqrt{2}}AC^{SP}\gamma_0\gamma_5\rho_N(r), \tag{1}$$

where G is the Fermi constant, $A=Z+N$ is the nuclear mass number, Z is the number of protons and N is the number of neutrons. $C^{SP}=(ZC_p^{SP}+NC_n^{SP})/A$, where C_p^{SP} and C_n^{SP} are the parameters of the scalar-pseudoscalar (T,P)-odd interaction for protons and neutrons, and γ_n are the Dirac matrices.

The Hamiltonian for the electron EDM interacting with internal atomic electric field $\mathbf{E}_{\rm int}$ can be written as [11,18,19]

$$\hat{H}_e = -d_e\sum_{i=1}^{Z}(\gamma_0-1)^i\mathbf{\Sigma}^i\cdot\mathbf{E}_{\rm int}^i, \tag{2}$$

where

$$\mathbf{\Sigma}^i = \begin{pmatrix}\sigma^i & 0\\ 0 & \sigma^i\end{pmatrix},$$

and $\mathbf{E}_{\rm int}$ is the internal atomic electric field at electron i. Summation is over atomic electrons.

Atomic EDM caused by any of the interactions Eqs. (1) and (2) is given by

$$\mathbf{d}_{\rm atom} = 2\sum_M \frac{\langle 0|\mathbf{D}|M\rangle\langle M|\hat{H}^{TP}|0\rangle}{E_0-E_M}, \tag{3}$$

where $|0\rangle$ is atomic ground state $\mathbf{D}=-e\Sigma_i\mathbf{r}_i$ is the electric dipole operator and $\hat{H}^{TP}$ is the (T,P)-odd operator. Summation goes over complete set of intermediate states $|M\rangle$, E_M are the energies of these states.

CI+MBPT method

To calculate the EDM of thallium we consider it as a system with three valence electrons above closed shells and use the CI+MBPT method [16,17] for the valence electrons. The EDM of the atom in the CI+MBPT is given by the formula very similar to Eq. (3) but with slightly different meaning of the notations. First, the many electron states $|0\rangle$, $|M\rangle$ are now three-electron states in the valence space. Second, the summation in the electric dipole operator $\mathbf{D}$ goes over valence electrons only while contribution from atomic core is taken into account by modifying the single-electron operator $\mathbf{d}:\mathbf{d}\to\mathbf{d}+\delta V_{\rm core}$, where $\delta V_{\rm core}$ is the correction to the electron core potential caused by external field. Closed shell core does not contribute to the EDM in the second order due to zero total angular momentum.

To perform the calculations we need to go through the following steps: (a) generate a complete set of single-electron states, (b) build an effective Hamiltonian in the valence space, (c) calculate core polarization, and (d) perform summation as in Eq. (3) over a complete set of three-electron states. Let us consider these tasks in turn.

We use the $\hat{V}^{N-3}$ approximation as in [20]. The calculations start from the relativistic Hartree-Fock procedure for the triple ionized thallium ion. This gives us the states and potential $\hat{V}_{\rm core}\equiv\hat{V}^{N-3}$ of the thallium core. We use the B-spline technique [21] to generate a complete set of single-electron states. These states are eigenstates of the Dirac operator with the electron potential $\hat{V}^{N-3}$. We use 50 B-splines of order 9 in a cavity of radius $40a_B$.

The effective CI+MBPT Hamiltonian for three valence electrons has the form

$$\hat{H}^{\rm eff} = \sum_{i=1}^{3}\hat{h}_1(r_i) + \sum_{i<j}^{3}\hat{h}_2(r_i,r_j), \tag{4}$$

where $\hat{h}_1$ is the single-electron part of the relativistic Hamiltonian

$$\hat{h}_1 = c\hat{\alpha}\mathbf{p} + (\hat{\beta}-1)m_ec^2 - \frac{Ze^2}{r} + \hat{V}^{N-3} + \hat{\Sigma}_1, \tag{5}$$

and $\hat{h}_2$ is the two-electron part of the Hamiltonian

$$\hat{h}_2(r_1,r_2) = \frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|} + \hat{\Sigma}_2(r_1,r_2). \tag{6}$$

In these equations, $\hat{\alpha}$ and $\hat{\beta}$ are the Dirac matrices, $\hat{V}^{N-3}$ is the Dirac-Hartree-Fock (DHF) potential of the closed-shell atomic core $(N-3=78,\ Z=81)$, and $\hat{\Sigma}$ is the correlation operator. It represents terms in the Hamiltonian arising due to virtual excitations from atomic core (see [16,17] for details). $\hat{\Sigma}\equiv 0$ corresponds to the standard CI method. $\hat{\Sigma}_1$ is a single-electron operator. It represents a correlation interaction of a particular valence electron with the atomic core. $\hat{\Sigma}_2$ is a two-electron operator. It represents screening of the Coulomb interaction between the two valence electrons by the core electrons. We calculate $\hat{\Sigma}_1$ for s-electrons using the all-order technique developed in [22]. $\hat{\Sigma}_1$ for p and d electrons as well as $\hat{\Sigma}_2$ are calculated in the second order of the many-body perturbation theory using the B-spline basis set described above. We use 40 lowest B-spline states up to $l_{\rm max}=5$ to calculate $\hat{\Sigma}$.

The same B-spline states are used to construct three-electron states for valence electrons. We use 16 lowest states above the core up to $l_{\rm max}=2$ for this purpose. The basis for the ground state is generated by allowing all possible single and double excitations from two initial configurations $6s^26p$ and $6s6p6d$. The basis for even states is generated by allowing all possible single and double excitations from three initial configurations, $6s^27s$, $6s^26d$, and $6s6p^2$. Variation of the basis size indicate that it is saturated with respect to $n_{\rm max}$ but not completely saturated with respect to $l_{\rm max}$. However, the contributions of the states with $l_{\rm max}>2$ are small and can be neglected at required level of accuracy.

The three-electron valence states are found by solving the eigenvalue problem,

$$\hat{H}^{\rm eff}\Psi_v = E_v\Psi_v, \tag{7}$$

using the standard CI techniques. Calculated and experimental energies of a few lowest-energy states of Tl are presented

TABLE I. Three-electron removal energy (RE, a.u.) and excitation energies (cm^{-1}) of thallium.

State		Theory		
		CI	CI+MBPT	Experiment[a]
RE		−1.9177	−2.0677	−2.0722
$6s^26p$	$^2P^o_{1/2}$	0	0	0
	$^2P^o_{3/2}$	6345	8049	7793
$6s^27s$	$^2S_{1/2}$	23023	26810	26478
$6s^27p$	$^2P^o_{1/2}$	30635	34496	34160
	$^2P^o_{3/2}$	31541	35507	35161
$6s^26d$	$^2D_{3/2}$	32313	36553	36118
	$^2D_{5/2}$	32363	36624	36200
$6s^28s$	$^2S_{1/2}$	34893	39037	38746
$6s^28p$	$^2P^o_{1/2}$	37572	41714	41368
	$^2P^o_{3/2}$	37936	42122	41741
$6s^27d$	$^2D_{3/2}$	38048	42359	42011
	$^2D_{5/2}$	38074	42395	42049
$6s^29s$	$^2S_{1/2}$	39563	43728	43166
$6s^28d$	$^2D_{3/2}$	40796	45931	44673
	$^2D_{5/2}$	41593	45971	44693
$6s6p^2$	$^4P_{1/2}$	37195	43545	45220
	$^4P_{3/2}$	40797	48339	49800
	$^4P_{5/2}$	44665	52779	53050

[a]Reference [24].

in Table I. One can see that the inclusion of $\hat{\Sigma}$ (CI+MBPT) leads to significant improvement of the agreement between theory and experiment.

To calculate transition amplitudes we need to take into account the effect of core polarization by external field. This is done by means of the time-dependent relativistic Hartree-Fock method (see, e.g., [23]), which is equivalent to the random-phase approximation (RPA). The RPA equations for an external field operator $\hat{F}$

$$(\hat{h}_1 - \epsilon_c)\delta\psi_c = -(\hat{F} + \delta\hat{V}_F^{N-3})\psi_c \tag{8}$$

are solved self-consistently for all states in atomic core in the same V^{N-3} potential as for the DHF states. The operator $\hat{F}$ is either the electric dipole operator or the operator of the (T,P)-odd interaction, or any other operator (e.g., hyperfine interaction). The correction to the core potential $\delta\hat{V}_F^{N-3}$ is used to calculate transition amplitudes

$$E1_{vw} = \langle\Psi_v|\hat{F} + \delta\hat{V}_F^{N-3}|\Psi_w\rangle. \tag{9}$$

Here, Ψ_v and Ψ_w are three-electron states found by solving the CI Eq. (7).

Calculated and experimental values of the electric dipole transition amplitudes and magnetic dipole hyperfine structure (hfs) constants A for low states of thallium, which are relevant to the calculation of the EDM are presented in Table II. Calculation of the hyperfine structure is a good way to test the wave function on short distances which is important for the matrix elements of weak interaction. The data in the Table show that the accuracy of the calculation of the $E1$-transition amplitudes and hyperfine constants of s and $p_{1/2}$ states is within few percent.

TABLE II. $E1$ transition amplitudes and hfs constants A of some low states of ^{205}Tl.

States	Calc.	Experiment	
$E1$ transition amplitudes (a.u.)			
$6p_{1/2}-7s_{1/2}$	1.73	1.81(2)	[25,26]
$6p_{1/2}-6d_{3/2}$	2.23	2.30(9)	[25,26]
Hyperfine structure constants A (MHz)			
$6p_{1/2}$	21067	21311	[27]
$7s_{1/2}$	11417	12297	[28]

Finally, the last task we must be able to do to calculate the EDM is to perform the summation over complete set of three-electron states. We use the Dalgarno-Lewis method [29] for this purpose In this method, a correction $\delta\Psi_v$ to the three-electron wave function of the ground-state v is introduced and the EDM is expressed as

$$\mathbf{d}_{\text{atom}} = 2\langle\delta\Psi_v|\hat{F} + \delta\hat{V}_F^{N-3}|\Psi_v\rangle. \tag{10}$$

Here, $\hat{F}$ is either the electric dipole operator or the operator of the (T,P)-odd interaction. The correction $\delta\Psi_v$ is found by solving the system of linear inhomogeneous equations

$$(\hat{H}^{\text{eff}} - E_v)\delta\Psi_v = -(\hat{G} + \delta\hat{V}_G^{N-3})\Psi_v. \tag{11}$$

Here, $\hat{G}$ is another operator from the pair $\mathbf{d}$, H^{TP}. If both operators $\hat{F}$ and $\hat{G}$ are the same electric dipole operator $\mathbf{d}$, then the expression similar to (10) gives static polarizability of the atom. Table III presents the results of the calculation of the static scalar polarizability α_0 of the thallium ground state. Here, α_{core} is the contribution of the thallium core to the polarizability, $\delta\alpha_{\text{core}}$ is the correction to the core polarizability due to Pauli principle which forbids excitations from the core to the occupied $6s$ and $6p$ states, α_{val} is the contribution of the valence electrons to the polarizability. The final result is in good agreement with other CI+MBPT [30] and coupled cluster [31] calculations.

III. RESULTS

The results of the calculations of the EDM of thallium in different approximations are presented in Table IV together

TABLE III. Static scalar polarizability α_0 of the thallium ground state (a.u.).

α_{core}	$\delta\alpha_{\text{core}}$	α_{val}	Total	Other
4.98	−0.67	44.50	48.81	49.2[a],50.4[b]

[a]Reference [30].
[b]Reference [31].

TABLE IV. EDM of Tl due to electron EDM (d_e: $d_{\rm atom}=Kd_e$) and scalar-pseudoscalar electron-nucleon (T,P)-odd interaction.

K	d ($10^{-18}C^{SP}$ e cm)	Comments
		This work
−614	−7.33	Single-configuration, no $\hat{\Sigma}$
−537	−6.43	Single-configuration with $\hat{\Sigma}$
−625	−7.49	Single-configuration in the ground state, with $\hat{\Sigma}$
−602	−7.22	Full CI but no $\hat{\Sigma}$
−581	−6.88	Full CI+MBPT but no RPA
−582	−6.98	Full scale calculations
−582(20)	−7.0(2)	Final
		Other calculations
−585		Ref. [15], coupled cluster
−1041		Ref. [12], DHF+1st order MBPT
−502		Ref. [12], Tietz[a]+1st order MBPT
−607		Ref. [12], Green[a]+1st order MBPT
−562		Ref. [12], Norcross[a]+1st order MBPT
700		Ref. [10], parametric potential
−500		Ref. [11], semiempirical estimate
−301		Ref. [13], 2nd order MBPT
−179		Ref. [14], 2nd order MBPT
	−4.056	Ref. [9], coupled cluster
	−7(2)	Ref. [8], RPA+rescaling of correlations from Ref. [14]

[a]Parametric potentials.

with earlier calculations. As it was pointed out in [11–13] thallium EDM is very sensitive to the strong correlations between $6s$ and $6p$ electrons. This interaction is treated pretty accurately in the configuration interaction technique used in present work. In contrast, all previous calculations treated thallium as a system with one external electron above closed shells. Therefore, present results are significantly more stable than earlier *ab initio* calculations.

The main source of uncertainty for present calculations comes from the core-valence correlations. Most of the core-valence correlations are included via second-order correlation operator $\hat{\Sigma}$. However, there are small contributions such as higher-order correlations, correction to $\hat{\Sigma}$ due to external field (structure radiation), renormalization of the wave function, etc. Quantum electrodynamic and Breit corrections are also expected to be small [32]. As one can see from Table IV the effect of including $\hat{\Sigma}$ into full-scale CI calculations on the EDM of Tl is about 3%. We use this as an estimate of the accuracy of our calculations.

Similar calculations for cesium give the following results (in agreement with previous calculations, see review [5]):

$$d(\mathrm{Cs}) = 0.759 \times 10^{-18} C^{SP}\ e\ \mathrm{cm}, \tag{12}$$

or

$$d(\mathrm{Cs}) = 124 d_e. \tag{13}$$

The estimated error of these results is about 3%:

The result of measurement of the EDM of ^{205}Tl [4] reads

$$d(^{205}\mathrm{Tl}) = -(4.0 \pm 4.3) \times 10^{-25}\ e\ \mathrm{cm}. \tag{14}$$

Using the numbers from Table IV we find

$$d_e = (6.9 \pm 7.4) \times 10^{-28}\ e\ \mathrm{cm}, \tag{15}$$

and

$$C^{SP} = (5.7 \pm 6.2) \times 10^{-8}. \tag{16}$$

These numbers are in good agreement with the analysis of [5].

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[1] W. C. Griffith, M. D. Swallows, T. H. Loftus, M. V. Romalis, B. R. Heckel, and E. N. Fortson, Phys. Rev. Lett. **102**, 101601 (2009).
[2] V. A. Dzuba, V. V. Flambaum, and S. G. Porsev, Phys. Rev. A **80**, 032120 (2009).
[3] K. V. P. Latha, D. Angom, B. P. Das, and D. Mukherjee, Phys. Rev. Lett. **103**, 083001 (2009).
[4] B. C. Regan, E. D. Commins, C. J. Schmidt, and D. DeMille, Phys. Rev. Lett. **88**, 071805 (2002).
[5] J. S. M. Ginges and V. V. Flambaum, Phys. Rep. **397**, 63 (2004).
[6] J. M. Amini, C. T. Munger, and H. Gould, Phys. Rev. A **75**, 063416 (2007).
[7] H. S. Nataraj, B. K. Sahoo, B. P. Das, and D. Mukherjee, Phys. Rev. Lett. **101**, 033002 (2008).
[8] A.-M. Mårtensson-Pendrill and E. Lindroth, Europhys. Lett. **15**, 155 (1991).
[9] B. K. Sahoo, B. P. Das, R. K. Chaudhuri, D. Mukherjee, and E. P. Venugopal, Phys. Rev. A **78**, 010501(R) (2008); **78**, 039901(E) (2008).
[10] P. G. H. Sandars and R. M. Sternheimer, Phys. Rev. A **11**, 473 (1975).
[11] V. V. Flambaum, Yad. Fiz. **24**, 383 (1976) [Sov. J. Nucl. Phys. **24**, 199 (1976)].
[12] W. R. Johnson, D. S. Guo, M. Idrees, and J. Sapirstein, Phys. Rev. A **34**, 1043 (1986).
[13] A. Ya. Kraftmakher, J. Phys. B **21**, 2803 (1988).
[14] A. C. Hartley, E. Lindroth, and A.-M. Mårtensson-Pendrill, J. Phys. B **23**, 3417 (1990).
[15] Z. W. Liu and H. P. Kelly, Phys. Rev. A **45**, R4210 (1992).
[16] V. A. Dzuba, V. V. Flambaum, and M. G. Kozlov, Phys. Rev. A **54**, 3948 (1996).
[17] V. A. Dzuba, V. V. Flambaum, and M. G. Kozlov, JETP Lett. **63**, 882 (1996).
[18] L. I. Schiff, Phys. Rev. **132**, 2194 (1963).
[19] P. G. H. Sandars, J. Phys. B **1**, 511 (1968).
[20] V. A. Dzuba, Phys. Rev. A **71**, 032512 (2005).
[21] W. R. Johnson and J. Sapirstein, Phys. Rev. Lett. **57**, 1126 (1986).
[22] V. A. Dzuba, V. V. Flambaum, and O. P. Sushkov, Phys. Lett. A **140**, 493 (1989).
[23] V. A. Dzuba, V. V. Flambaum, and O. P. Sushkov, J. Phys. B **17**, 1953 (1984).
[24] C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, DC, 1958), Vol. III.
[25] A. Gallagher and A. Lurio, Phys. Rev. **136**, A87 (1964).
[26] J. C. Hsieh and J. C. Baird, Phys. Rev. A **6**, 141 (1972).
[27] A. Lurio and A. G. Prodell, Phys. Rev. **101**, 79 (1956).
[28] A. I. Odintzov, Opt. Spectrosc. **9**, 142 (1960).
[29] A. Dalgarno and J. T. Lewis, Proc. R. Soc. London, Ser. A **233**, 70 (1955).
[30] M. G. Kozlov, S. G. Porsev, and W. R. Johnson, Phys. Rev. A **64**, 052107 (2001).
[31] M. S. Safronova, W. R. Johnson, U. I. Safronova, and T. E. Cowan, Phys. Rev. A **74**, 022504 (2006).
[32] V. A. Dzuba, V. V. Flambaum, and M. S. Safronova, Phys. Rev. A **73**, 022112 (2006).