Relations between matrix elements of different weak interactions and interpretation of the PNC and EDM measurements in atoms and molecules

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The relations between matrix elements of different P- and T-odd weak interactions are derived. We demonstrate that similar relations hold for parity nonconserving (PNC) transition amplitudes and electron electric dipole moments (EDM) of atoms and molecules. This allows to express P- and T-odd effects in many-electron systems caused by different symmetry-breaking mechanisms via each other using simple analytical formulas. We use these relations for the interpretation of the anapole moment measurements in cesium and thallium and for the analysis of the relative contributions of the scalar-pseudoscalar CP-odd weak interaction and electron EDM to the EDM of Cs, Tl, Fr and other atoms and many polar molecules (YbF, PbO, ThO, etc.). Model-independent limits on electron EDM and the parameter of the scalar-pseudoscalar CP-odd interaction are found from the analysis of the EDM measurements for Tl and YbF.

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I. INTRODUCTION

The study of the parity and time invariance violation in atoms, molecules and nuclei is a low-energy, relatively inexpensive alternative to the high-energy search for new physics beyond the standard model (see, e.g. a review [1]). Accurate measurements of the parity non-conservation (PNC) in atoms is one of the most promising ways of exploring this path. It culminated in very precise measurements of the PNC in cesium [2]. Interpretation of the measurements based on accurate atomic calculations led to perfect agreement with the standard model and put strong constrain on any new physics beyond it [3, 4] (see also review [1] for more detailed discussion). First unambiguous measurement of the nuclear P-odd anapole moment was also reported in the Cs PNC experiment [2].

At present, the study of symmetry violations in atoms and molecules goes mostly in three major directions (see, e.g. [5]): (i) the PNC measurements for a chain of isotopes; (ii) the measurements of nuclear anapole moments; and (iii) the measurements of the P,T-odd permanent electric dipole moments of atoms and molecules. Interpretation of the anapole moment and EDM measurements requires sophisticated atomic or molecular calculations. The calculations are difficult and sometimes disagree with each other. For example, the calculations of the thallium electric dipole moment (EDM) by Nataraj et al. [6] and by Sahoo et al. [7] disagree with earlier calculations [8, 9], calculations of the nuclear spin-dependent PNC in cesium by Mani and Angom [10] disagree with earlier calculations [11–14], etc. The difficulties are mostly due to the strong inter-electron correlations which need to be treated to all orders of the many-body perturbation theory. Majority of publications present the effect of one particular P- or CP-odd interaction for a specific atom or a molecule. If another weak interaction Hamiltonian is considered, the calculations are to be done again.

There is an active search for the CP-violating permanent EDM of polar molecules, such as YbF [15], PbO [21, 22], ThO [23], etc. Interpretation of the measurements requires complicated molecular calculations. Most of calculations consider only one possible source of the molecular EDM: electron EDM or scalar-pseudoscalar CP-odd interaction. We show that the analytical ratios of the matrix elements provide reliable link between molecular EDMs caused by different sources. Therefore, if calculations exist for a particular CP-odd
operator, no new calculations are needed to find an EDM caused by a different operator.

The approach developed in this paper is easy to apply when matrix elements of the symmetry-breaking operator between $s_{1/2}$ and $p_{1/2}$ single-electron states strongly dominate over other matrix elements. This is the case for the PNC amplitudes between atomic s-states and for the EDM of atoms and molecules caused by electron EDM or scalar-pseudoscalar CP-odd weak interaction. We also use this approach for the thallium anapole sign problem.

II. SINGLE-ELECTRON MATRIX ELEMENTS OF WEAK INTERACTION AND MANY-BODY EFFECTS

The PNC amplitude of an electric dipole transition between states of the same parity $|a⟩$ and $|b⟩$ is equal to:

$$E_{1}^{\text{PNC}} = \sum_{n} \left[ \frac{⟨b|d|n⟩⟨n|H_{\text{PNC}}|a⟩}{E_{a} - E_{n}} + \frac{⟨b|H_{\text{PNC}}|n⟩⟨n|d|a⟩}{E_{b} - E_{n}} \right] ,$$

where $d = -e \sum_{i} r_{i}$ is the electric dipole operator, $H_{\text{PNC}}$ is the operator of a P-odd CP-even weak interaction.

The EDM of an atom in state $a$ is given by

$$d_{a} = 2 \sum_{n} \frac{⟨a|d|n⟩⟨n|H_{\text{CP}}|a⟩}{E_{a} - E_{n}}$$

where $H_{\text{CP}}$ is the operator of a CP-odd weak interaction.

States $a$, $b$, $n$ in (1) and (2) are the many-electron atomic states. However, we can start for simplicity from an atom with one external electron above closed shells keeping in mind cesium atom as an example. Then in the lowest order of the perturbation theory in residual Coulomb interaction the PNC amplitude and the EDM of the atom are given by (1) and (2) in which states $a$, $b$ and $n$ are single electron (e.g. Hartree-Fock) states. It is easy to see that all single-electron matrix elements of weak interaction are proportional to each other. Indeed, only short distances contribute to the value of the matrix elements (see appendix for analytical estimations). These distances are $r < r_{N}$ for the PNC interactions and for the scalar-pseudoscalar CP-odd interaction ($R_{N}$ is nuclear radius) and $r \leq q_{0}/Z$ for the electron EDM operator ($q_{0}$ is Bohr radius). On these distances the energy of the single-electron state can be neglected compared to the nuclear potential $−Ze^{2}/r$ and Dirac equations for all single-electron states become identical. The only difference comes from the normalization of the states. Suppose we have two different operators of, say, CP-odd weak interaction $H_{\text{CP}}$ and $H'_{\text{CP}}$. If we establish proportionality between matrix elements for a particular pair $i, j$ of single electron states

$$⟨i|H_{\text{CP}}|j⟩ = R⟨i|H'_{\text{CP}}|j⟩,$$

then due to the proportionality of the wave functions on short distances, the same proportionality, with the same value of $R$ would hold for any pairs of single-electron states and for the total EDM of atom (2). Furthermore, if the $s_{1/2} - p_{1/2}$ matrix elements of the weak interaction strongly dominate over other matrix elements then the proportionality is not affected by the many-body effects. Indeed, if other weak matrix elements are neglected then any many-body expression is a sum of terms with one $s_{1/2} - p_{1/2}$ weak matrix element in each term. Since all of them are proportional with the same proportionality coefficient $R$ the proportionality would hold for the sum as well.

Table I shows the ratios of the matrix elements of the spin-dependent to spin-independent P-odd weak interactions ($R_{1}$) and scalar-pseudoscalar CP-odd interaction to electron EDM ($R_{2}$) for the $6s, 6p_{1/2}$ states of cesium and with and without the inclusion of dominating many-body effects. The ratios are stable while the matrix elements change by up to two times. Units: $\alpha/(−QW)$ for $R_{1}$ and $10^{−15}\text{cm}^{5}/\text{d}$, a.u. for $R_{2}$.

| Approximation | $R_{1}$ | $R_{2}$ |
|---------------|---------|---------|
| RHF \(\psi_{6s}^{\text{HF}}|H_{W}|\psi_{6p_{1/2}}^{\text{HF}}⟩\) | 4.78 | 8.96 |
| RPA \(\psi_{6s}^{\text{RPA}}|H_{W} + \delta V_{\text{core}}|\psi_{6p_{1/2}}^{\text{RPA}}⟩\) | 4.88 | 8.94 |
| BO \(\psi_{6s}^{\text{BO}}|H_{W}|\psi_{6p_{1/2}}^{\text{BO}}⟩\) | 4.78 | 9.03 |
| BO+CP \(\psi_{6s}^{\text{BO}}|H_{W} + \delta V_{\text{core}}|\psi_{6p_{1/2}}^{\text{BO}}⟩\) | 4.85 | 9.01 |
| Analytical, eq. \(\text{A10−A13.A15}\) | 4.84 | 9.01 |

$^{a}$Brueckner orbitals
$^{b}$Brueckner orbitals and core polarization

III. PARITY NON-CONSERVATION

Hamiltonian describing parity-nonconserving electron-nuclear interaction can be written as a sum of spin-independent (SI) and spin-dependent (SD) parts (we use...
atomic units: $\hbar = |e| = m_e = 1$:

$$H_{\text{PNC}} = H_{\text{SI}} + H_{\text{SD}}$$

$$= \frac{G_F}{\sqrt{2}} \left( -\frac{Q_W}{2} \gamma_5 + \frac{\kappa}{I} \alpha I \right) \rho(r),$$  \hspace{1cm} (4)

where $G_F \approx 2.2225 \times 10^{-14}$ a.u. is the Fermi constant of the weak interaction, $Q_W$ is the nuclear weak charge, $\kappa = \begin{pmatrix} 0 & \sigma \\ \sigma & 0 \end{pmatrix}$ and $\gamma_5 = \begin{pmatrix} 0 & -I \\ I & 0 \end{pmatrix}$ are Dirac matrices, $I$ is the nuclear spin, and $\rho(r)$ is the nuclear density normalized to 1. The strength of the spin-dependent PNC interaction is proportional to the dimensionless constant $\kappa I$ which is to be found from the measurements. There are three major contributions to $\kappa I$ arising from (i) electromagnetic interaction of atomic electrons with nuclear anapole moment \[23\], (ii) electron-nucleus spin-dependent weak interaction \[26\], and (iii) combined effect of the spin-independent weak interaction and the magnetic hyperfine interaction \[27\] (see, also review \[1\]). In this work we do not distinguish between different contributions to $\kappa I$ and present the results in terms of total $\kappa I$ which is the sum of all possible contributions.

Within the standard model the weak nuclear charge $Q_W$ is given by \[23\]

$$Q_W \approx -0.9877N + 0.0716Z.$$  \hspace{1cm} (5)

Here $N$ is the number of neutrons, $Z$ is the number of protons.

The PNC amplitude of an electric dipole transition between states of the same parity $|i\rangle \equiv |J_f F_f M_f\rangle$ and $|f\rangle \equiv |J_f F_f M_f\rangle$ is equal to:

$$E_{\text{PNC}}^{1f} = \sum_n \left[ \frac{\langle f | d | n \rangle \langle n | H_{\text{PNC}} | i \rangle}{E_i - E_n} \right]$$

$$+ \frac{\langle f | H_{\text{PNC}} | n \rangle \langle n | d | i \rangle}{E_f - E_n},$$  \hspace{1cm} (6)

where $d = -e \sum_i r_i$ is the electric dipole operator, and $F = I + J$ is the total angular momentum.

Applying the Wigner-Eckart theorem we can express the amplitudes via reduced matrix elements

$$E_{\text{PNC}}^{1f} = (-1)^{F_f - M_f} \begin{pmatrix} F_f & 1 & F_f \\ 1 & M_f & q M_f \end{pmatrix}$$

$$\times \langle J_f F_f | d_{\text{PNC}} | J_i F_i \rangle.$$  \hspace{1cm} (7)

Detailed expressions for the reduced matrix elements of the SI and SD PNC amplitudes can be found e.g. in Refs. \[29\] and \[30\]. For the SI amplitude we have

$$\langle J_f, F_f | d_{\text{SI}} | J_i, F_i \rangle = (-1)^{F_f + F_i} J_f + J_i + 1$$

$$\times \sqrt{(2F_f + 1)(2F_i + 1)} \left\{ \begin{array}{ccc} J_f & J_i & 1 \\ F_f & F_i & 1 \end{array} \right\}$$

$$\times \sum_n \left[ \frac{\langle J_f | d | n \rangle \langle J_i | n | H_{\text{SI}} | J_i \rangle}{E_i - E_n} \right]$$

$$+ \frac{\langle J_f | H_{\text{SI}} | n \rangle \langle J_i | n | d | J_i \rangle}{E_f - E_n}.$$  \hspace{1cm} (8)

It is convenient to present the amplitude in a compact form

$$\langle J_f, F_f | d_{\text{SI}} | J_i, F_i \rangle = C(S_1 + S_2),$$  \hspace{1cm} (9)

where $C \equiv C(F_f, J_f, F_i, J_i)$ is the angular coefficient and sums $S_1$ and $S_2$ do not depend on $F_f$ and $F_i$:

$$S_1 = \sum_n \frac{\langle J_f | d | n \rangle \langle n | H_{\text{SI}} | J_i \rangle}{E_i - E_n},$$

$$S_2 = \sum_n \frac{\langle J_f | H_{\text{SI}} | n \rangle \langle n | d | J_i \rangle}{E_f - E_n}.$$  \hspace{1cm} (10)

For the SD PNC amplitude we have

$$\langle J_f, F_f | d_{\text{SD}} | J_i, F_i \rangle =$$

$$\sqrt{(I + 1)(2I + 1)(2F_f + 1)(2F_i + 1)/I}$$

$$\times \sum_n \left[ \begin{array}{ccc} (-1)^{J_f - J_i} & J_n & J_i \\ I & I & F_f \end{array} \right] \left\{ \begin{array}{ccc} J_n & J_f & 1 \\ F_f & F_i & 1 \end{array} \right\}$$

$$\times \langle J_f | d | n \rangle \langle n, J_n | b | J_i \rangle$$

$$\times \frac{\langle J_f | H_{\text{SD}} | n \rangle \langle n, J_n | d | J_i \rangle}{E_f - E_i}.$$  \hspace{1cm} (11)

Like in the spin-independent PNC amplitude \[8\], it is convenient to present the SD amplitude in a compact form

$$\langle J_f, F_f | d_{\text{SD}} | J_i, F_i \rangle = \sum_{i=1}^{4} c_i S'_i.$$  \hspace{1cm} (12)

Here $c_i \equiv c(F_f, J_n, F_i)$ ($i = 1, 2, 3, 4$) are angular coefficients which can be extracted from \[11\]. Sums $S'_i$ do not depend on $F_f$ and $F_i$:

$$S'_1 = \sum_n \frac{\langle J_f | d | J_n \rangle \langle J_n | b | J_i \rangle}{E_i - E_n},$$

$$S'_2 = \sum_n \frac{\langle J_f | d | J_n \rangle \langle J_n | b | J_i \rangle}{E_f - E_i},$$

$$S'_3 = \sum_n \frac{\langle J_f | b | J_n \rangle \langle J_n | d | J_i \rangle}{E_i - E_n},$$

$$S'_4 = \sum_n \frac{\langle J_f | b | J_n \rangle \langle J_n | d | J_i \rangle}{E_f - E_n}.$$  \hspace{1cm} (13)

Equation (12) has more terms than (9) due to the electron vector nature of the nuclear-spin-dependent operator.
The total PNC amplitude can be presented in a form convenient for extraction of the values of \( \kappa \) from the PNC measurements (see, e.g. \[31, 32\])

\[
E_{\text{PNC}} = E(1 + R).
\]

(13)

Here \( E \) is the SI PNC amplitude given by \[38\] and \( R \) is the ratio of the SD to SI PNC amplitudes. In this work we are mostly interested in the values of \( R \). Using compact expressions \[39, 12\] one can write

\[
R = \frac{c_1 S_1' + c_2 S_2' + c_3 S_3' + c_4 S_4'}{C(S_1 + S_2)}.
\]

(14)

According to the discussion of the previous section, ratios of the SD and SI weak matrix elements do not depend on the principal quantum number \( n \). Therefore, using the ratios

\[
\begin{align*}
    r_1 &= \langle J_n | b | J_l \rangle / \langle J_n | H_{\text{SI}} | J_l \rangle, \\
    r_2 &= \langle J_n' | b | J_l \rangle / \langle J_n | H_{\text{SI}} | J_l \rangle, \\
    r_3 &= \langle J_f | b | J_n \rangle / \langle J_f | H_{\text{SI}} | J_n \rangle, \\
    r_4 &= \langle J_f | b | J_f \rangle / \langle J_f | H_{\text{SI}} | J_f \rangle,
\end{align*}
\]

(15)

we can get rid of the sums \( S_1', S_2', S_3', S_4' \), involving the SD matrix elements and replace them with the SI sums \( S_1, S_2 \):

\[
R = \frac{(c_1 r_1 + c_2 r_2) S_1 + (c_3 r_3 + c_4 r_4) S_2}{C(S_1 + S_2)} = \frac{(c_1 r_1 + c_2 r_2) S_1 / S_2 + (c_3 r_3 + c_4 r_4)}{C(S_1 / S_2 + 1)}.
\]

(16)

The only parameter in \[14\] which comes from numerical calculations is the ratio \( S_1 / S_2 \) of two different contributions to the SI PNC amplitude (see Eqs. \[39\]). All other parameters are given by analytical expressions. The \( c_1, c_2, c_3, c_4 \) and \( C \) parameters are just angular coefficients. The ratios \( r_1, r_2, r_3, r_4 \) can also be approximated by analytical expressions which will be discussed below.

The expression \[14\] can be further simplified in an important case of an \( ns \rightarrow n's \) transition (e.g. the 6s-7s transition in Cs). On a few per cent level of accuracy the \( s - p_{3/2} \) matrix elements of the SD PNC interaction can be neglected \[32\]. This means that \( r_2 = r_4 = 0 \). We also have \( r_1 = r_3 = r \) and \[10\] is reduced to

\[
R = r \frac{c_1 S_1 / S_2 + c_3}{C(S_1 / S_2 + 1)}.
\]

(17)

Substituting \( r \) from \[10\] leads to

\[
R = 4.90(1 - 0.073 Z^2 \alpha^2) \frac{c_1 S_1 / S_2 + c_3}{C(S_1 / S_2 + 1)} \frac{\kappa}{-Q_W}.
\]

(18)

\( Q_W \) is the weak nuclear charge. As in \[14\] the only parameter which comes from numerical calculations is the ratio \( S_1 / S_2 \). On the other hand, knowing the value of \( R \) for at least two \( hfs \) components of the PNC transition is sufficient for extraction of \( \kappa \) from the measurements.

\[\text{A. PNC in cesium}\]

Experimental values for the two different \( hfs \) components of the parity non-conserving 6s-7s transition in cesium are \[2\]

\[
E_{\text{PNC}}(6s_F=4 - 7s_F=3) = 1.6349(80) \text{ mV/cm},
\]

\[
E_{\text{PNC}}(6s_F=3 - 7s_F=4) = 1.5576(77) \text{ mV/cm}.
\]

(19)

To extract \( \kappa \) we use \[13\] and calculate \( R \) for these two transitions using \[18\]. To do so we note that \( J_f = J_l = J_n = 1/2, Q_W = -73.19 \) (see Eq. \[5\]) and take \( S_1 / S_2 = -0.3459 \) from Ref. \[33\]. This leads to the system of equations

\[
E(1 + 0.06739 \kappa) = 1.6349(80),
\]

\[
E(1 - 0.05937 \kappa) = 1.5576(77).
\]

(20)

The solution for \( \kappa \) is \( \kappa = 0.382(56) \). This result is in good agreement with the values \( \kappa = 0.393(56) \) from Ref. \[11\] and \( \kappa = 0.375(56) \) from Ref. \[12\]. Accurate calculations similar to what is reported in our previous work for \( Ba^+ \), \( Yb^+ \) and \( Ra^+ \) \[32\] lead to the value \( \kappa = 0.376 \) which is in perfect agreement with the all-order calculations of Ref. \[12\]. This is an illustration of the accuracy of the analysis based on the ratio of the matrix elements. The value \( \kappa = 0.382 \) coming from this analysis differs by less than 2% from the value \( \kappa = 0.376 \) coming from the accurate calculations. This difference is due to two simplifications: (a) neglecting the \( s - p_{3/2} \) matrix elements of the SD weak interaction, and (b) assuming that the ratio of the matrix elements is the same for all single-electron states.

Recent relativistic coupled-cluster calculations of the nuclear spin-dependent PNC in Cs \[10\] report the values of the SD PNC matrix elements which are about 30% smaller than those of the all-order calculations of Ref. \[12\]. This is in disagreement not only with this work but with all earlier calculations of the SD PNC in cesium \[11\, 14\]. Given the proportionality of the matrix elements of the SI and SD weak interactions discussed above, the results of Ref. \[10\] are also in disagreement with all most accurate calculations of the SI PNC in cesium (see, e.g. \[3\, 4\, 33\]). The latter calculations have accuracy better than 0.5% and are used to test the Standard Model in Cs PNC experiment \[2\] where the accuracy is 0.35%.

\[\text{B. Anapole moment of thallium}\]

The value of the nuclear anapole moment of thallium, extracted from the measurements of the PNC in the

\[\text{1} \text{ Refs.} \[11\] \text{ and} \[12\] \text{ use different definition of} \ \kappa. \text{ The conversion factors are} \ (I + 1/2)/(I + 1) \text{ for Ref.} \[11\] \text{ and} \ I \text{ for Ref.} \[12\]. \ I \text{ is nuclear spin,} \ I = 7/2 \text{ for} \ 133\text{Cs.}\]
6$p_{1/2}$/$6p_{3/2}$ transition \[13\] is in disagreement with the results of nuclear calculations (see, e.g. ref. \[17\]). The analysis of the experimental data based on simple single-electron approximations gives the value $\kappa_a = -0.22 \pm 0.30 \[13\]$. The analysis based on sophisticated many-body calculations gives very close value of $\kappa_a = -0.26 \pm 0.27 \[10\]$. On the other hand, the “best value” obtained in nuclear calculations is $\kappa_a = 0.24 \[17\]$. To extract the sign of $\kappa_a$ from the experiment one needs the relative sign of SI and SD amplitudes. Here we show that simple analysis with the use of the analytical ratio of the matrix elements of the weak interactions supports the findings of Refs.\[15, 16\] leaving the problem of sign disagreement unsolved.

If we keep only $s_{1/2} - p_{1/2}$ matrix elements of the weak interaction for both SI and SD interactions, then the general expression \[16\] can be reduced to

$$R(F, F') = r \frac{c(F, F')}{C(F, F')} \quad (21)$$

where $r$ is one of the ratios $r_i \[13\]$ which corresponds to the $s_{1/2} - p_{1/2}$ weak matrix elements, $c(F, F')$ and $C(F, F')$ are corresponding angular coefficients. Comparing \[13\] to the parameterization used in \[16\] ($R = \Im(E_{1PNC}/M1)$)

$$R(F, F') = C(Z) \{Q_W - 6\kappa \xi(F, F')\} \quad (22)$$

and substituting (\[10\]) into (\[21\]) we get for the parameters $\xi$

$$\xi(F, F') = -0.817 \left(1 - 0.073Z^2\alpha^2\right) \frac{c(F, F')}{C(F, F')} \quad (23)$$

Corresponding values of $\xi$ are compared in Table \[III\] with the results of the many-body calculations of \[16\]. We see that both calculations give very close results leaving no room for a sign error.

IV. EDM OF ATOMS AND MOLECULES

Table \[III\] shows the ratio of the $s_{1/2} - p_{1/2}$ matrix elements of the electron EDM operator (\[13\]) to that of the scalar-pseudoscalar CP-odd operator (\[13\]). Numerical results for EDM of Cs, Tl and YbF are also given for comparison. Units: $d_e/(C_{SP}10^{-18} \text{ e cm})$. (For other isotopes $F' = A'R/A$)

| $Z$ | Atom | Ratios |
|-----|------|--------|
|     |      | Analytical | Numerical |
| 37  | $^{87}\text{Rb}$ | 228 | \[16]\* |
| 55  | $^{133}\text{Cs}$ | 158 | 163\* |
| 56  | $^{138}\text{Ba}^+$ | 152 |   |
| 70  | $^{173}\text{Yb}^+$ | 114 | 115\* |
| 81  | $^{205}\text{Tl}$ | 89 | 83\* |
| 82  | $^{208}\text{Pb}$ | 88 |   |
| 87  | $^{211}\text{Fr}$ | 83 |   |
| 90  | $^{232}\text{Th}$ | 75 |   |

\[a\]Cs atom, Ref. \[9\].

\[b\]YbF molecule, Ref. \[36\].

\[c\]Tl atom, Ref. \[9\].

Ref. \[9\], the result for Yb$^+$ is in excellent agreement with the many-body calculations of Ref. \[36\] for the YbF molecule. We stress that the Table compares the ratios of the single-electron matrix elements obtained with a simple analytical formula to the ratios of the EDMs obtained with sophisticated many-body calculations. These results provide an unambiguous link between the sign and value of two different contributions to the EDM of atoms and molecules which have a heavy atom from the Table \[III\].

A. EDM of polar molecules

Polar molecules have strong inter-atomic electric field which enhance the effect of electron EDM and lead to molecular EDM which are several orders of magnitude larger than those in atomic systems. The experimental search is in progress for YbF \[18\], PbO \[21, 22\], and ThO \[23\] while other molecules are also discussed in the literature (see, e.g. \[34\]). Interpretation of the measurements requires molecular calculations. Table \[IV\] shows the results of most recent or most accurate calculations for some polar molecules. More detailed data are presented for YbF molecule for which the EDM measurements were recently reported \[18\]. The effects of electron EDM and scalar-pseudoscalar CP-odd interaction are considered. The results are presented in terms of the CP-odd parameters $W_d$ and $W_c$

$$W_d = \langle \Psi_0 | H_{c} | \Psi_0 \rangle / d_e, \quad (24)$$

$$W_c = \langle \Psi_0 | H_{SP} | \Psi_0 \rangle / C_{SP}. \quad (25)$$

To compare with other works one should keep in mind that most of them present $W_S$ instead of $W_c$, where

$$W_S = \frac{2}{k_s} \langle \Psi_0 | H_{SP} | \Psi_0 \rangle, \quad (26)$$
TABLE IV: CP-odd interaction constants $W_d$, and $W_e$ for some polar molecules and their ratios. Effective electric field $E_{\text{eff}}$ is presented together with $W_d$ ($\langle H_e \rangle = W_d d_e = -E_{\text{eff}} d_e$). The results of present paper are shown in bold.

| Molecule | $E_{\text{eff}}$ (GV/cm) | $W_d$ (10$^{-24}$ Hz/e cm) | $W_e$ (kHz) | $W_d/W_e$ (10$^{18}$/e cm) |
|----------|-----------------|------------------|-------------|---------------------|
| BaF      | 6.1$^a$         | -1.5$^a$         | -10         | 152                |
| YbF      | 6.1             | -1.5             | -10$^b$     | 152                |
| PbF      | 31$^c$          | -7.5$^c$         | -59         | 127                |
| PbO a(1) | 19$^d$          | -4.6$^d$         | -41$^d$     | 112                |
| PbO B(1) | 26$^e$          | -6.3$^e$         | -55         | 114                |
| PbO a(1) | 26$^f$          | -6.2$^f$         | -54$^f$     | 115                |
| ThO      | 24$^g$          | -5.8$^g$         | -54$^g$     | 108                |
| ThF$^+$  | 25$^h$          | -6.1$^h$         | -53         | 114                |
|          | 32$^i$          | -7.7$^i$         | -68         | 114                |
|          | 21$^j$          | -5.2$^j$         | -46$^j$     | 114                |
| HgF      | 95$^k$          | -23$^k$          | -226        | 90                 |
| PbO      | -31$^l$         | 7.5$^l$          | 85          | 88                 |
| ThO      | 104$^m$         | -25$^m$          | -336        | 75                 |
| ThF$^+$  | 90$^n$          | -22$^n$          | -290        | 75                 |

$^a$Meyer and Bohn, Ref. [32].
$^b$Nayak and Chaudhuri, Ref. [32].
$^c$Kozlov and Ezhov, Ref. [33].
$^d$Titov et al., Ref. [33].
$^e$Kozlov, Ref. [33].
$^f$Quincey et al., Ref. [34].
$^g$Parpia, Ref. [40].
$^h$Mosyagin et al., Ref. [41].
$^i$Petrov et al., Ref. [42].

The constants $k_s$ and $C^{SP}$ of the strength of the CP-odd interaction are related by $Z k_s = A^{SP}$. Factor 2 in the definition of $W_S$ [20] is absent in some of the papers.

Most of the calculations of the molecular EDM include only one of the CP-odd effects: that of the electron EDM or the scalar-pseudoscalar interaction. We use the relations between the matrix elements of the two CP-odd Hamiltonians to fill the gaps in the table. Corresponding results are shown in bold. For example, according to Ref. [34] the effect of electron EDM in the YbF molecule is $W_d = -15 \times 10^{24}$ Hz/e cm. Using the ratio $W_d/W_e = 114 \times 10^{18}$/e cm for Yb$^+$ from Table III we find that the effect of the scalar-pseudoscalar interaction is $W_e = -132$ kHz.

B. Extraction of the electron EDM and the parameter of the scalar-pseudoscalar CP-odd interaction from the EDM measurements for YbF and Tl

Recent measurement of T and P violation in YbF molecule [18] combined with the Tl EDM measurement [19] and the data from Table III allows to obtain independent limits on the electron EDM and the CP-violating interaction. The values of the electron EDM extracted from the experimental data for Tl [19] and YbF [18] under assumption that there is no other contribution to atomic/molecular EDM read

\[
\text{Tl: } d_e = (6.9 \pm 7.4) \times 10^{-28} \text{ e cm}, \quad (27)
\]

\[
\text{YbF: } d_e = (-2.4 \pm 5.7 \pm 1.5) \times 10^{-28} \text{ e cm}. \quad (28)
\]

In fact, there are other contributions from the CP-odd electron-nucleus interactions. Here we consider only the scalar-pseudoscalar interaction (A11). Other contributions should be small due to the constrains obtained from the EDM measurements for mercury [43]. Using data from Table III one can rewrite (27,28) as

\[
\text{Tl: } d_e + 1.2 \times 10^{-20} C^{SP} = (6.9 \pm 7.4) \times 10^{-28} \text{ e cm}, \quad (29)
\]

\[
\text{YbF: } d_e + 8.8 \times 10^{-21} C^{SP} = (-2.4 \pm 5.7 \pm 1.5) \times 10^{-28} \text{ e cm}. \quad (30)
\]

Solving these equations for $d_e$ and $C^{SP}$ leads to

\[
d_e = (-2.8 \pm 3.0) \times 10^{-27} \text{ e cm}, \quad (31)
\]

\[
C^{SP} = (3.0 \pm 3.0) \times 10^{-7}. \quad (32)
\]

The limit [31] for $d_e$ is significantly weaker than those presented in (27) and (28). This is because the values of the ratios of the matrix elements of the two operators are very close for Tl and Yb (see Table III). The most fortunate case leading to strongest limits for $d_e$ and $C^{SP}$ would correspond to very different values of the ratio, preferably with the different sign. However, the formulas (A15) and (A13) and the data in Table III show that this is not possible. The ratio is always positive and slowly decrease with $Z$. A slightly different value of the ratio should be expected for the EDM of radium in the excited metastable 7s6d $^3D_2$ state. The EDM of radium in this state is strongly enhanced [44] due to proximity of the 7s7p $^3P_1$ state (the energy interval is $\sim 5$ cm$^{-1}$) and is proportional to the $(7p_3/2^2)|H_{CP}|(6d_{3/2})$ matrix element of the CP-odd interaction $H_{CP}$. However, the value of this matrix element is strongly dominated by the core polarization effect which in turn is mostly due to the $s_{1/2} - p_{1/2}$ matrix elements between core and excited states. Therefore, the ratio of the two contributions is not very different from what was considered above for Tl and YbF.

C. EDM of Tl and Fr

EDM of thallium due to electron EDM and scalar-pseudoscalar CP-odd interaction was recently calculated in our paper [4]. The ratio of the two contributions was found to be $83d_e/(C^{SP}10^{-18} e)$ cm which is in good agreement with the analytical result of this paper: $W_d/W_e = 89/(10^{18} e)$ cm. The EDM enhancement factor is $-582$ [3] which is in very good agreement with the value $-585$ found in earlier calculations.
by Liu and Kelly. The EDM enhancement factor for thallium was also calculated in more recent work of Ref. 6 and found to be -466 ($d_{\text{Tl}} = -466 d_e$) which is about 25% smaller than the results mentioned above. The EDM of thallium due to the scalar-pseudoscalar CP-odd interaction calculated by the same group earlier ($d_{\text{Tl}} = -4.06 \times 10^{-18} C^{\text{SP}} e \text{ cm}$) is also smaller than in other calculations of Ref. 4 and 20. On the other hand, the ratio of the two contributions is 115 (in units of $d_e/(C^{\text{SP}} 10^{-18} e \text{ cm})$) which is significantly larger than the analytical and numerical values of 89 and 83 (same units, see Table III).

EDM enhancement factor for francium was calculated in Ref. 49 and found to be 910(46): $d(\text{Fr}) = 910(46)d_e$. This value is in good agreement with the value of 894.93 found in more recent calculations 50. Using the value $d(\text{Fr}) = 910(46)d_e$ and the ratio of the electron-EDM matrix element to the scalar-pseudoscalar interaction presented in Table III we can now reconstruct another contribution to the EDM of francium:

$$d(\text{Fr}) = 11 \times 10^{-18} C^{\text{SP}} e \text{ cm.} \quad (33)$$

It is interesting to note that the calculation of the nuclear spin-dependent PNC amplitude between hyperfine components of the ground state of francium presented in Ref. 29 involves the same sum as the expression 22 for the EDM of the atom. Therefore, we can reconstruct the EDM enhancement factor for Fr using the results of Ref. 29 and proportionality between matrix elements of the nuclear spin-dependent interaction (A9) and the electron EDM operator (A15). This leads to $d(\text{Fr}) = 854d_e$ which is in good agreement with the value $d(\text{Fr}) = 910(46)d_e$ of Ref. 49. This is a good consistency test of both calculations.

Reading the papers citing our works (see, e.g. 6, 50) reveals the need to clear some points about our method of calculations. For example, the atomic electric field interacting with electron EDMs is calculated in 5 and 49 as a derivative of the total potential which includes both nuclear and electron parts. The formula $E = Ze/r^3$ for the leading contribution to the atomic electric field presented on first page of 49 may make an impression that only nuclear field is included. However, the formula (2) few lines below clearly includes screening functions $Q(r)$ and $P(r)$ for both the nuclear Coulomb and the external electric field. By the way, the inclusion of the electron electric field change the matrix elements of the electron EDM for thallium by 0.4% only. This is because main contribution comes from short distances where the electron electric field is small since the electron potential rapidly tends to a constant inside the 1s orbital. Therefore, we include only nuclear electric field in the analytical analysis (formula (A15) in the appendix) while keeping both contributions in the numerical calculations.

The authors of 6 claim that atomic core is strongly contracted in the $V^{N-1}$ starting approximation used in our calculations 9. In fact, it is not. Fig 1 shows the outermost 5d$_{5/2}$ core function of Tl calculated in the $V^{N-3}$ and $V^{N-1}$ approximations. The difference between the functions is very small. This is due to the fact that the valence 6s and 6p electrons are located outside of atomic core. Their charge distribution creates almost constant potential and no electric field inside the sphere where all inner electrons are located. Therefore, the valence electrons have practically no effect on the core wave function (see 15 for a detailed discussion). The change is even smaller for other core functions. The core functions enter the configuration interaction (CI) Hamiltonian via core potential $V_{\text{core}}$ to which all core electrons contribute (from 1s to 5d). The difference for $V_{\text{core}}$ in the $V^{N-3}$ and $V^{N-1}$ approximations is very small 45. Moreover, the corresponding corrections to the configuration interaction (CI) Hamiltonian have been included in 9 using the many-body perturbation theory approach.

As it is well known, the eigenstates of a Hamiltonian do not depend on the basis one uses. The valence states are indeed different in the $V^{N-3}$ and $V^{N-1}$ approximations. However, this should have no effect on the final results as long as the saturation of the basis for valence states is achieved. There are only two conditions the basis states must satisfy: (a) they must be orthogonal to the core, and (b) they must constitute a complete set of states. Therefore, it does not matter whether valence states are calculated in the $V^{N-3}$ or $V^{N-1}$ potential or by any other method (e.g., a popular basis $\psi_n(r) = r^n \psi_0(r)$ 40), the final results should be the same. If there is any difference in the results, the most likely reason for this is the incompleteness of the basis set.

In spite of no difference in final results there is a good reason for the use of the $V^{N-3}$ approximation – the simplicity and good convergence of the many-body pertur-
bation theory (MBPT) for the core-valence correlations. This is the only approximation which has no subtraction diagrams \(^{47}\). To be more precise, we should state that the condition for the absence of the subtraction diagrams is that the potential used to calculate single-electron core states and the potential of the core in the CI Hamiltonian are exactly the same. In the case of thallium atom treated as a three-valence-electrons system this corresponds to the \(V^{N-3}\) approximation. When any other approximation is used one has to include the subtraction diagrams.

Large energy denominators suppress the value of the correlation terms in the \(V^{N-3}\) approximation ensuring good convergence of the MBPT \(^{45}\). There must be large cancellation between subtraction and other diagrams to ensure the same final results if any other initial approximation is used. This is very similar to the well known fact that the Hartree-Fock basis is the best choice for any MBPT calculations. Zero-order results might be better in some other approximation, however, strong cancelation between subtraction and other diagrams would lead to poor convergence of the MBPT.

The authors of \(^6\) claim that the major drawback of our work \(^9\) is the inclusion of the core-valence correlations in the second order only. However, the correlations between the valence electrons and core electrons below the 6s state are small which is evident from the fact that their inclusion change the EDM of Tl by 3% only \(^9\). Therefore, only the correlations between three valence electrons should be treated to all orders. This is done in \(^9\) to a very high precision using the CI technique.

V. CONCLUSION

We have demonstrated the proportionality relations between the nuclear-spin dependent and spin-independent PNC effects in atoms and the scalar-pseudoscalar and the electron EDM contributions to the EDM of atoms and molecules. The relations are given by the simple analytical formulas and can be used to express one symmetry breaking effect through another. Using these relations and accurate calculations of the spin-independent PNC we have confirmed earlier interpretations of the nuclear anapole measurements in cesium and thallium. We have also confirmed the ratio of the scalar-pseudoscalar CP-odd and electron EDM contributions to the EDM of Cs, Tl and other atoms and some polar molecules. Using the relations we found the scalar-pseudoscalar contribution to the EDM of francium atom and the scalar-pseudoscalar and electron EDM contributions to the EDM of many polar molecules. Using experimental limits on EDMs of thallium and YbF we found model-independent limits on the electron EDM and the constant of the scalar-pseudoscalar CP-odd interaction.

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Appendix A: Matrix elements of the weak interaction

1. Wave function

We use single-electron wave functions in a form

\[
\psi(r)_{njl} = \frac{1}{r} \left( \frac{f_v(r)}{i \alpha g_v(r)} \right) \Omega(n)_{jl} \tag{A1}
\]

where \(n\) is the principal quantum number and an index \(v\) replaces the three-number set \(n, j, l\); \(\alpha\) is the fine structure constant.

We need \(s_{1/2}\) and \(p_{1/2}\) wave functions inside the nucleus to calculate matrix elements. Following Ref. \(^51\) we assume uniform distribution of electric charge inside a sphere of radius \(R_N\). Taking formulas for the \(s_{1/2}\) and \(p_{1/2}\) wave functions from Ref. \(^51\) and keeping terms up to \(Z^2 \alpha^2\) we come to the following expressions

\[
\begin{align*}
    f_s(x) &= A_s x \left( 1 - \frac{3}{8} Z^2 \alpha^2 x^2 \right), \\
    g_s(x) &= -A_s \frac{x}{2} \left( 1 - \frac{x^2}{5} \right), \\
    f_{p_{1/2}}(x) &= A_p \alpha x^2 \left( 1 - \frac{x^2}{5} \right), \\
    g_{p_{1/2}}(x) &= A_p x \left( 1 - \frac{3}{8} Z^2 \alpha^2 x^2 \right).
\end{align*}
\]

Here \(x = r/R_N\), \(R_N\) is nuclear radius. \(A_s\) and \(A_p\) are normalization factors. They can be found by matching \(^{52}\) to atomic wave functions at short distances outside of the nucleus (see, e.g. \(^52\)). The use of the semiclassical wave functions presented in \(^52\) leads to approximate expressions

\[
\begin{align*}
    A_s &= \frac{(1 + \gamma)(2ZR_N)^{\gamma}}{(1 - 0.375Z^2 \alpha^2)\sqrt{\Gamma(2\gamma + 1)\nu_{1.5}^2}}, \\
    A_p &= \frac{\sqrt{Z}(2ZR_N)^{\gamma}}{(1 - 0.375Z^2 \alpha^2)\Gamma(2\gamma + 1)\nu_{1.5}^{1.5}},
\end{align*}
\]

where \(\gamma = \sqrt{1 - Z^2 \alpha^2}\), \(\Gamma\) is gamma-function, \(\nu\) is effective principal quantum number: single-electron energy of state \(n\) is \(\epsilon_n = -1/(2\nu^2)\). Note that normalization factors \(^{52}\) are needed only to compare the matrix elements of the electron EDM to other matrix elements. For cases not involving electron EDM there is exact cancellation of the normalization factors. Therefore, their uncertainty does not contribute to the uncertainty of final results.
2. PNC matrix elements

Matrix element of the SI PNC $H_{SI}$ interaction (first term in (11)) is

$$\langle \kappa_1 | H_{SI} | \kappa_2 \rangle = \frac{i G_F Q_W}{2\sqrt{2}} \times$$

$$\alpha \delta_{-\kappa_1, \kappa_2} \int (f_1 g_2 - g_1 f_2) \rho(r) dr. \tag{A4}$$

Substituting (A2) for the $s_{1/2} - p_{1/2}$ transition and assuming uniform nuclear charge distribution we get

$$\langle s_{1/2} | H_{SI} | p_{1/2} \rangle = \frac{i G_F Q_W}{2\sqrt{2}} \times$$

$$\alpha \rho_0 A_s A_p R_N \frac{1}{3} (1 - 0.34 Z^2 \alpha^2). \tag{A5}$$

Here $A_s$ and $A_p$ are given by (A3) and

$$\rho_0 = 3 \frac{4\pi R_N^3}{Z}. \tag{A6}$$

Matrix element of the SD PNC $b$ operator \([11]\) is

$$\langle \kappa_a | b | \kappa_b \rangle = -i \frac{G_F \kappa}{\sqrt{2}} \alpha (-\kappa_1 | C_1 | \kappa_2) \times$$

$$\int \left( (\kappa_2 - \kappa_1 + 1) f_1 g_2 - (\kappa_1 - \kappa_2 + 1) g_1 f_2 \right) \rho(r) dr. \tag{A7}$$

The reduced matrix element of the spherical harmonic $C_k$ is

$$\langle \kappa_a | C_k | \kappa_b \rangle = (-1)^{j_a+1/2} \sqrt{(2j_a + 1)(2j_b + 1)}$$

$$\times \xi(j_a + j_b + k) \begin{pmatrix} j_b & j_a & k \\ -1/2 & 1/2 & 0 \end{pmatrix} \tag{A8}$$

The ratio of $\kappa_1$ to $\kappa_2$ for the $s_{1/2} - p_{1/2}$ transition and uniform nuclear charge distribution we have

$$\langle s_{1/2} | b | p_{1/2} \rangle = -i \frac{G_F \kappa}{\sqrt{2}} \times$$

$$\alpha \frac{\sqrt{2}}{3} \rho_0 A_s A_p R_N (1 - 0.41 Z^2 \alpha^2). \tag{A9}$$

Substituting the functions (A2) leads to

$$\langle s_{1/2} | H_{SI} | p_{1/2} \rangle = \frac{i G_F Q_W}{2\sqrt{2}} \alpha \rho_0 A_s A_p R_N \frac{1}{3} (1 - 0.34 Z^2 \alpha^2). \tag{A10}$$

Here $A_s$ and $A_p$ are given by (A3) and $\rho_0$ is given by (A6).

3. Scalar-pseudoscalar CP-odd interaction

The Hamiltonian of the scalar-pseudoscalar electron-nucleon (T,P)-odd interaction can be written as \([1]\)

$$H_{SP}^{T,P} = \frac{i G_F A C_{SP}^{T,P} \gamma_0 \gamma_5 \rho N(r)}, \tag{A11}$$

where $G_F$ 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. $C_{SP}^{T,P} = (Z C_{SP}^p + N C_{SP}^n) / A$, where $C_{SP}^p$ and $C_{SP}^n$ are the parameters of the scalar-pseudoscalar (T,P)-odd interaction for protons and neutrons, $\gamma_n$ are the Dirac matrices.

$$\langle a | H_{SP}^{T,P} | b \rangle = \frac{G_F}{\sqrt{2}} \alpha A C_{SP}^{T,P}$$

$$\times \delta_{-\kappa_a, \kappa_b} \int (f_a g_b + g_a f_b) \rho_N dr. \tag{A12}$$

$$\langle s_{1/2} | H_{SP}^{T,P} | p_{1/2} \rangle =$$

$$- \frac{G_F A C_{SP}^{T,P}}{3\sqrt{2}} \alpha A_s A_p \rho_0 R_N \frac{1}{3} (1 - 0.56 Z^2 \alpha^2). \tag{A13}$$

Here $A_s$ and $A_p$ are given by (A3) and $\rho_0$ is given by (A6).

4. Electron EDM

The Hamiltonian for the electron EDM interacting with internal atomic electric field $E_{int}$ can be written as \([52]\)

$$H_e = -d_e (\gamma_0 - 1) \Sigma \cdot E_{int}, \tag{A14}$$

where

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

and $E_{int}$ is internal atomic electric field. The $s_{1/2} - p_{1/2}$ matrix element can be found in Ref. \([53]\):

$$\langle s_{1/2} | H_e | p_{1/2} \rangle = - \frac{4Z^3 \alpha^2 d_e}{\tau(\gamma^2 - 1)(\nu_s \nu_p)^{3/2}}. \tag{A15}$$

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