Electric Dipole Moments due to Nuclear Schiff Moment Interactions:
A Reassessment of the Atoms $^{129}$Xe, $^{199}$Hg, and the molecule $^{205}$TIF

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Abstract

We present relativistic many-body calculations of atomic and molecular Schiff-moment interaction constants including interelectron correlation effects using atomic Gaussian basis sets specifically optimized for the Schiff interaction. Our present best results employing a Gaussian nuclear density function are $\alpha_{SM} = (0.364 \pm 0.025) \times 10^{-17} \text{ cm}^2/\text{e fm}^3$ for atomic $^{129}$Xe, $\alpha_{SM} = (-2.40 \pm 0.24) \times 10^{-17} \text{ cm}^2/\text{e fm}^3$ for atomic $^{199}$Hg, and $W_{SM} = (39967 \pm 3600) \text{ a.u.}$ for the thallium nucleus in the molecule $^{205}$TIF. We discuss agreements and discrepancies between our present results and those from earlier calculations on the atoms $^{129}$Xe and $^{199}$Hg. Using the most recent measurements of $\mathcal{P}, \mathcal{T}$-odd electric dipole moments and the present interaction constants reliable upper bounds on the Schiff moments of the $^{199}$Hg and $^{205}$Tl nuclei are determined in the context of a single-source assumption.

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I. INTRODUCTION

The Standard Model (SM) of elementary particle physics [1–3] is an extremely well-tested theory of fundamental particles and their interactions. However, it leaves a number of firmly established observations about our universe unexplained, like its matter and energy content [4]. Specifically, the SM does not allow to accommodate baryon asymmetry of the universe (BAU), i.e., the significant surplus of denominational matter over antimatter in the universe [5].

A necessary condition for explaining the BAU is the violation of the combined discrete symmetries charge conjugation and parity (CP), favoring the production of a matter over an antimatter particle [6]. CP-violation (CPV) has been observed in the decay of certain mesons [7–9], and a partial theory of CP-violation has become an integral part of the SM [10]. However, it is generally agreed that this manifestation of CP-violation is insufficient for explaining the BAU. New sources of CP-violation are hence required, which in particular generally also yield flavor-diagonal CP-violation [11]. Under the assumption that CPT invariance (T denoting time reversal) of fundamental physical laws holds [12], CPV implies the violation of T symmetry. The detection of an atomic or molecular electric dipole moment (EDM), the Hamiltonian of which is P, T-odd, would in turn indicate CPV. Thus, EDMs represent a nearly background-free probe of beyond-SM CPV [13].

The origins of nuclear, atomic and molecular EDMs in terms of fundamental CPV phases may be diverse [14–16]. In electronically closed-shell systems like atomic mercury (Hg) [17] or molecular thallium fluoride (TlF) [18, 19] the situation is relatively complicated, and several hadronic mechanisms may contribute to leading order. The nuclear Schiff moment and the nucleon-electron tensor-pseudotensor interaction are the leading CPV sources at the hadronic and nuclear energy scale in such systems.

The nuclear Schiff potential is a low-order P, T-odd term in the expansion of the nuclear charge distribution [20, 21] which polarizes the electron cloud in an atomic system, giving rise to atomic/molecular EDMs. Thus, atomic-scale measurements search for or constrain the nuclear Schiff moment and – in turn – the underlying CPV sources. The chosen systems and states have electronically closed shells which strongly suppresses leptonic CPV sources such as the electron EDM, d_e, and some semi-leptonic CPV sources such as the nucleon-electron scalar-pseudoscalar coupling, C_S [22, 23]. The currently most sensitive measurement [24] is |d_{Hg}| < 7.4 \times 10^{-30} \text{ecm} with spin-polarized mercury atoms, ^{199}\text{Hg}, leading to an upper bound on the Schiff moment of |S_{Hg}| < 3.1 \times 10^{-13} \text{efm}^3 (95\% \text{ c.l.}). In the calculation of this upper bound an average over interaction constants from different theory groups has been used. The upper bound translates [17, 25] into constraints on more fundamental parameters, the CPV pion-nucleon couplings g_{\pi}^{(0)}, g_{\pi}^{(1)}, g_{\pi}^{(2)}, the nucleon
EDMs $d_p$ and $d_n$, the quantum chromodynamics $\Theta$ parameter and chromo-EDMs.

In the present paper we pursue several goals:

1. The Schiff moment interaction in general is quite strongly dependent on the quality of the atomic basis set for the target nucleus in electronic-structure calculations, as has also been substantiated earlier [26, 27]. Our calculations confirm this finding for all systems we have studied so far. In the present paper we present a systematic approach to extending standard atomic basis sets with the aim of a reliable and economic description of the Schiff moment interaction in both atoms and molecules. In the following section we define our method for calculation of atomic and molecular Schiff-moment interactions and give a detailed description of the strategy for optimizing required Gaussian basis sets.

2. In section III we discuss applications to $^{129}\text{Xe}$, $^{199}\text{Hg}$ and $^{205}\text{TlF}$ using a large optimized basis set for each respective system and carefully taking into account interelectron correlation effects shellwise and at various excitation ranks. Based on our findings we address current disagreements among previously published results for atomic Schiff moment interactions in $^{129}\text{Xe}$ and $^{199}\text{Hg}$.

3. We conclude on our study in section IV and use the most recent experimental EDM measurements [24, 28] and our calculated interaction constants for these systems to derive constraints on the respective nuclear Schiff moments.

II. THEORY AND METHODS

A. Theory

1. The Atomic Schiff Moment Interaction Hamiltonian

The atomic Schiff-moment interaction for a single electron in the field of a point nucleus has been given as [20], Eq. (8.75),

$$\hat{H}_{SM} = -e \mathbf{S} \cdot \nabla_r \delta(r)$$

where the vector coefficient $\mathbf{S} := S \frac{\mathbf{I}}{I}$ is the Schiff moment [29] of the nucleus with $\mathbf{I}$ denoting nuclear spin and $S$ the scalar Schiff-moment constant [30].

Recent developments use a more realistic finite nuclear charge density, and the Hamiltonian for the interaction of an electron with the Schiff potential [30] has been represented as

$$\hat{H}_{SM} = -e \varphi_{SM}(r) = -3e \frac{\mathbf{S} \cdot \hat{r}}{B} \rho(r)$$
where $B = \int_0^{\infty} \rho(r)r^4dr$, and $\rho(r)$ the nuclear charge density at position $r$.

2. *Expectation value approach to Schiff moment interaction*

Our principal strategy is to determine the $E$-field dependent $\mathcal{P}, \mathcal{T}$-odd energy shift $\Delta \varepsilon$ as a function of an atomic interaction constant. The full details of the general approach are found in Ref. [31]. Here, we present the specific approach for the calculation of Schiff-moment interaction constants.

a. **Atoms** For atomic calculations we include an external homogeneous electric field $E_{\text{ext}}$ along the $z$ axis. In an atom with $n$ electrons the associated $\mathcal{P}, \mathcal{T}$-odd energy shift can then be expressed as an expectation value over the one-electron Hamiltonian in Eq. (2) (here in a.u.)

$$\Delta \varepsilon_{\text{SM}} = -S_z^3 B \left\langle \sum_{j=1}^{n} \hat{z}_j \rho(r_j) \right\rangle_{\psi(E_{\text{ext}})}$$

where $\psi(E_{\text{ext}})$ is the electronic wavefunction of the field-dependent state. We first solve a zeroth-order problem

$$\hat{H}(E_{\text{ext}}) |\psi(E_{\text{ext}})\rangle = \varepsilon(E_{\text{ext}}) |\psi(E_{\text{ext}})\rangle$$

with $\varepsilon$ the field-dependent energy eigenvalue and $\hat{H}(E_{\text{ext}})$ the Dirac-Coulomb Hamiltonian including the interaction term with the external field:

$$\hat{H}(E_{\text{ext}}) := \hat{H}^{\text{Dirac-Coulomb}} + \hat{H}^{\text{Int-Dipole}}$$

$$= \sum_j^n \left[ c \alpha_j \cdot p_j + \beta_j c^2 - \frac{Z}{r_{jk}} \right] + \sum_{j>k}^{n} \frac{1}{r_{jk}} + \sum_j \mathbf{r}_j \cdot \mathbf{E}_{\text{ext}} \mathbb{1}_4$$

$E_{\text{ext}}$ is weak (see below for details) and homogeneous, the indices $j, k$ run over $n$ electrons, $Z$ is the proton number with the nucleus $K$ placed at the origin, and $\alpha, \beta$ are standard Dirac matrices. $E_{\text{ext}}$ is not treated as a perturbation but included a priori in the variational optimization of the wavefunction, $\psi(E_{\text{ext}})$.

Technically, $\psi(E_{\text{ext}})$ is a configuration interaction (CI) vector [32] built from Slater determinants over field-dependent 4-spinors. In the atomic case the wavefunction is expanded as follows:

$$\psi(E_{\text{ext}}) \cong |M_J\rangle = \sum_{I=1}^{\dim \mathcal{F}^t(M,n)} c_{(M_J),I} \langle \mathcal{S}\mathcal{T}_{(M_J),I} | \rangle$$

where $| \rangle$ is the true vacuum state, $\mathcal{F}^t(M,n)$ is the symmetry-restricted sector of Fock space ($M_J$ subspace) with $n$ electrons in $M$ four-spinors, $\mathcal{S} = a_i^\dagger a_j^\dagger a_k^\dagger \ldots$ is a
string of spinor creation operators, \( \overline{T} = a_{\mu}^\dagger a_{\mu} a_{\mu}^\dagger \ldots \) is a string of creation operators of time-reversal transformed spinors. The determinant expansion coefficients \( c_{(M_j),I} \) are generally obtained as described in refs. [33, 34].

The electric dipole moment of the atomic system in terms of the Schiff moment is

\[
d_a = \alpha_{SM} S_z
\]

(7)

where using Eqs. (3) and (7) we define the Schiff moment interaction constant

\[
\alpha_{SM} := \frac{\Delta \varepsilon_{SM}}{S_z E_{ext}} = \frac{-3}{B} \frac{\sum_{j=1}^{n} \hat{z}_j \rho(r_j)}{E_{ext}} \psi(E_{ext}).
\]

(8)

In the linear régime \( W_a(\psi(E_{ext})) := -\frac{3}{B} \langle \hat{z} \rho(r) \rangle_{\psi(E_{ext})} = C E_{ext} \) where \( C \) is a constant with respect to \( E_{ext} \). From this it follows that in the linear régime \( \alpha_{SM} = C \) which is independent of \( E_{ext} \). Through numerical analysis we determine quasi linearity of \( W_a \) for \( E_{ext} = 0.0003 \) a.u. in the case of Xe and for \( E_{ext} = 0.00024 \) a.u. in the case of Hg. \( E_{ext} \) is oriented along the \( z \) axis in the atomic case.

b. **Molecules**  
In the molecular case the unperturbed wavefunction \( \psi \) is not a \( \mathcal{P} \) eigenstate, thus no external electric field needs to be applied. The general strategy is similar to the one for atoms but with some modifications. The energy shift is written as

\[
\Delta \varepsilon_{SM} = -S_z \frac{3}{B} \left\langle \sum_{j=1}^{n} \hat{z}_j \rho(r_j) \right\rangle_{\psi}
\]

(9)

and the wavefunction is obtained from the zeroth-order problem

\[
\hat{H} |\psi\rangle = \varepsilon |\psi\rangle
\]

(10)

with

\[
\hat{H} := \hat{H}^{\text{Dirac-Coulomb}} = \sum_{j} \left[ c \mathbf{\alpha}_j \cdot \mathbf{p}_j + \beta_j c^2 - \sum_{K}^{2} \frac{Z_K}{r_{jK}} \mathbb{1}_4 \right] + \sum_{k>j}^{n} \frac{1}{r_{jk}} \mathbb{1}_4 + V_{KL}
\]

(11)

for a diatomic molecule where \( K \) runs over nuclei and \( V_{KL} \) is the classical electrostatic potential energy for the two Born-Oppenheimer-fixed nuclei. The CI expansion of the electronic wavefunction reads

\[
\psi \equiv |\Omega\rangle = \sum_{I=1}^{\dim \mathcal{F}(M,n)} c_{(\Omega),I} (\mathcal{ST}_{(\Omega),I} |\ )
\]

(12)
where $\Omega$ is the total angular momentum projection. The Schiff-moment interaction constant for a target nucleus $A$ of a molecule is then written as

$$W_{\text{SM}}(A) := \frac{\Delta \varepsilon_{\text{SM}}(A)}{S_z(A)} = -\frac{3}{B} \left\langle \sum_{j=1}^{n} \hat{z}_j \rho_A(r_j) \right\rangle \psi.$$  \hspace{1cm} (13)

In practical applications the target nucleus is placed at the origin of the reference frame.

**B. Methods**

1. **Nuclear charge density**

   To describe the charge density, $\rho(r)$, at position $r$ for a nucleus with $Z$ protons we in the present work use a Gaussian model [35] with

   $$\rho(r) = Z \left( \frac{\zeta}{\pi} \right)^{\frac{3}{2}} e^{-\zeta r^2}$$  \hspace{1cm} (14)

   where the exponent $\zeta$ is taken from Ref. [35]. This density is used both for the calculation of the electronic wavefunction as well as for the evaluation of the interaction constants in Eqs. [8] and [13].

2. **Matrix Elements**

   We here present the main ideas of the Schiff-moment interaction operator implementation. In a Gaussian nuclear model the prefactor $B$ introduced in Eq. (2) can be written in terms of the parameter $\zeta$ from Eq. (14). Integration by parts leads to

   $$B = \frac{3}{8\pi \zeta}.$$  \hspace{1cm} (15)

   The electronic spinors constituting the wavefunction $\psi$ in Eqs. [8] and [13] are expanded as a linear combination of primitive Gaussians in the DIRAC code. It is, therefore, convenient to contract with the exponential $e^{-\zeta r^2}$ from Eq. [14] by adding $\zeta$ to the primitive Gaussian exponents. Then, the matrix elements to be evaluated in Eqs. [8] and [13] can be written as

   $$\left\langle \psi \left| e^{-\zeta r^2} \hat{z} \right| \psi \right\rangle = \left\langle \psi_\zeta \left| \hat{z} \right| \psi \right\rangle$$  \hspace{1cm} (16)

   where $\psi_\zeta = \psi e^{-\zeta r^2}$. Finally, the r.h.s. of Eq. [16] is evaluated as a dipole length integral in the DIRAC package.
3. Atomic basis sets

The nuclear Schiff moment gives rise to an asymmetric charge distribution on the nuclear surface and a related constant electric field inside the nucleus \[29\] that is oriented along the nuclear spin, \( I \). The atomic Schiff moment interaction will, therefore, predominantly affect atomic electronic wavefunctions that penetrate the atomic nucleus. Eq. (2) shows that the Schiff moment interaction is \( \mathcal{P} \)- and \( \mathcal{T} \) violating, thus leading to a mixing of states with opposite parity, and therefore predominantly to mixing of electronic \( s \) and \( p \) waves.

Our first goal was to investigate the performance of the standard systematic N-tuple-zeta series of Gaussian basis sets in calculating the Schiff-moment interaction constant \( \alpha_{SM} \) in Dirac-Coulomb Hartree-Fock (DCHF) approximation. The results in Table II demonstrate that \( N = 4 \) is a minimal requirement for quantitatively reliable results. Still, compared to the literature results that agree well in mean-field approximation, a standard QZ basis set yields a too small result. Gaussian basis sets of quintuple-zeta (and higher) quality are currently not available for heavy atoms and are time consuming to develop \[36\]. Given the physical nature of the Schiff-moment interaction it is, therefore, as an alternative strongly suggested to increase the \( s \) and \( p \) subspaces of the most extensive standard Gaussian basis set in order to obtain an accurate description of the relevant matrix elements, written generically as

\[
\langle s \left| -\frac{3}{B} \hat{z} \rho(r) \right| p \rangle.
\]  

The strategy of our present basis-set optimization starts from Dyall’s relativistic Gaussian basis set, QZ, extended with diffuse and correlating functions \[37\]. This set is then further augmented taking two criteria into account:

1. Densification in the \( s \) and \( p \) spaces.

   Following the so-called “even-tempered prescription” (see Ref. \[38\]) we insert a Gaussian function between each adjacent pair of original ones according to

\[
\zeta_n = \sqrt{\zeta_{n-1} \zeta_{n+1}}
\]  

where \( \zeta_i \) is the exponent in \( e^{-\zeta_i r^2} \) of the \( i \)th Gaussian function and \( \zeta_{n-1}(\zeta_{n+1}) \) is the next larger (smaller) exponent. This procedure could in principle be repeated several times but the rapid increase in dimension of the \( s \) and \( p \) spaces leads to a steep increase in computational cost.

2. Addition of \( sp \) pair(s) of even-tempered dense and diffuse functions.

   In order to obtain a more extended basis set in a balanced way we add a pair of Gaussian functions – one diffuse and one dense – to the respective densified
basis set. The new compact exponent \( \zeta_{n+1} \) is obtained according to

\[
\zeta_{n+1} = \frac{\zeta_n^2}{\zeta_{n-1}}
\]

(19)

where \( \zeta_n, \zeta_{n-1} \) are the two most compact coefficients in the \( sp \)-densified basis-set defined in the latter point 1.

The new diffuse exponent is obtained accordingly:

\[
\zeta_{n-1} = \frac{\zeta_n^2}{\zeta_{n+1}}
\]

(20)

where \( \zeta_{n-1} \) is the new more diffuse coefficient and \( \zeta_n, \zeta_{n+1} \) are the two most diffuse coefficients in the \( sp \)-densified basis set defined in point 1.

As shown in Table I for Xe neither is the total Dirac-Coulomb Hartree-Fock energy \( (\varepsilon_{DCHF}) \) improved (lowered) nor is the Schiff moment interaction constant \( \alpha_{SM} \) changed substantially by a second densification if the respective densification is accompanied by the addition of a sufficient number of pairs of even-tempered compact and diffuse functions (respectively, \( +1sp \) and \( +3sp \) for simple and double densification, see Table VII). For this reason, we densify the original basis set only once. From Tables I, III, VII and Fig. I, respectively, we conclude that our accurate and most economic basis set to describe \( \alpha_{SM} \) for Xe and Hg is the single \( sp \)-densified Dyall’s cvQZ + 1sp.

Combining the two aforementioned criteria, we propose a systematic method to optimize Gaussian basis sets suited to address the Schiff moment interaction constant \( \alpha_{SM} \). The starting point is an even-tempered \( (s,p) \)-spaces densification. Then, compact and diffuse \( (s,p) \) pairs are added until a minimal DCHF energy converged at a level of \( \approx 10^{-6} \) a.u. in reached, as is shown in Fig. I. Under these circumstances \( \alpha_{SM} \) shall be considered converged at DCHF level.

4. Molecular basis sets

Obtaining a suitable basis set for a target atom in a diatomic molecule can be achieved by following steps 1 and 2 described in the latter atomic case for the atom with the target nucleus \( A \) (see Eq. 13). However the basis-set optimization must be done by calculating \( \varepsilon_{DCHF} \) and \( W_{SM}(A) \) in the molecular field. The internuclear distance \( R \) is kept fixed during the whole process. It is obtained from experimental data in the present case. For TlF, we conclude from Table V that our accurate and most economic basis set to describe \( W_{SM}(A) \) is the \( sp \)-densified Dyall’s cvQZ + 1sp.
III. RESULTS FOR SCHIFF MOMENT INTERACTION

A. Technical Details

All present calculations have been carried out using a locally-modified version of the DIRAC program package [39]. The chosen symmetry group is the double point group $C_{2v}$. Small components of the Dirac spinors are generated through the restricted-kinetic-balance [40] prescription and all small-component integrals are explicitly taken into account.

B. $^{129}$Xe

| Model | $\alpha_{SM} \left[ 10^{-17} \text{cm}^3/\text{e} \text{fm}^2 \right]$ | $\varepsilon_{\text{DCHF}}$ [a.u.] |
|-------|----------------------------------|---------------------|
| DZ-21s15p | -1.220 | -7446.876435682 |
| TZ-29s22p | -0.379 | -7446.895053545 |
| QZ-34s28p | 0.318 | -7446.895409376 |
| sp-densified QZ-67s55p | 0.314 | -7446.895379750 |
| sp-densified+1sp QZ-69s57p | 0.373 | -7446.895401869 |
| sp-densified+2sp QZ-71s59p | 0.375 | -7446.895401810 |
| sp-densified+3sp QZ-73s61p | 0.375 | -7446.895401761 |
| sp-densified+4sp QZ-75s63p | 0.375 | -7446.895401779 |
| Double sp-densified QZ-133s109p | 0.362 | -7446.895392349 |
| Double sp-densified+3sp QZ-139s115p | 0.369 | -7446.895401848 |
| Dzuba et al. [30] (RPA, 2002) | 0.38 | - |
| Ramachandran et al. [41] (CPHF, 2014) | 0.374 | - |
| Sakurai et al. [42] (CPDF, 2019) | 0.38 | - |

Previous calculations have been carried out in random-phase approximation (RPA) [30] and within the coupled-perturbed Dirac-Hartree-Fock (CPHF) framework [41] yielding very similar results. In recent work using the relativistic normalized coupled cluster method in singles and doubles approximation (RNCCSD) [42] interelectron correlation effects have been taken into account and a contri-
bution of $\approx -15\%$ to $\alpha_{SM}$ is reported which is unexpectedly large for Xe. Our general-excitation-rank CI method can test this claim.

| CI Model/virtual cutoff | $\alpha_{SM} \left[ 10^{-17} \text{cm}^3 / \text{e}_\text{fm}^4 \right]$ |
|-------------------------|-------------------------------|
| DCHF                    | 0.373                         |
| SD8/5au                 | 0.348                         |
| SD8/10au                | 0.353                         |
| SD8/20au                | 0.352                         |
| SD8/50au                | 0.352                         |
| SDT8/10au               | 0.351                         |
| SDTQ.0.2au_SDT8/10au    | 0.355                         |
| S10_SD18/10au           | 0.355                         |
| S10_SD18/20au           | 0.359                         |
| S10_SD18/50au           | 0.359                         |
| SD10_SD18/20au          | 0.362                         |
| S8_SD16/20au            | 0.352                         |
| S8_SD16/50au            | 0.353                         |
| SD8_SD16/20au           | 0.352                         |
| S8_SD8_SD24/20au        | 0.352                         |
| **Final present**       | **0.364 ± 0.025**             |

In Table II the results from a series of systematic calculations including electron correlation effects from various atomic shells and at various maximum excitation ranks are compiled. As has also been found earlier even the leading correlation effects from the valence shells ($5s, 5p$), described by Double excitations, are weak in the ground state of atomic Xe. In our model SD8 they decrease $\alpha_{SM}$ by only around 6%. The model SDT8 introduces all Triple excitations and the model SDTQ.0.2au_SDT8 in addition a subset of Quadruple excitations (where the spinor space for these Quadruples has been truncated at 0.2 a.u.) to the model SD8. Including these higher excitation ranks affects the Schiff interaction constant by less

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than 1%. Excitations out of the 4d shell lead to an increase by about 3%. Here we have not tested higher excitation ranks than Doubles since the difference from adding Single and Double excitations out of the 4d shell is already small (around 1%). Finally, the correlation contributions from core shells 3s, 3p, 4s, 4p are seen to be smaller than 1%.

The final present value is thus calculated as follows. As base value we take the result where the greatest number of electrons has been included in the correlation expansion, from the model S8, SD8, SD24/20au. To this we add corrections due to higher excitation ranks in the valence shells and correlations among and with the 4d electrons, according to

\[ \alpha_{SM}(\text{Final}) = \alpha_{SM}(S8, SD8, SD24/20au) + \alpha_{SM}(SDTQ, 0.2au, SDT8/10au) - \alpha_{SM}(SD8/10au) + \alpha_{SM}(SD10, SD18/20au) - \alpha_{SM}(SD8/20au) \]

To this final value we assign an uncertainty of 7% by adding individual uncertainties due to the nuclear-density model (3%), atomic basis set (2%), higher excitation ranks (1%), and the Breit interaction (1%) that is not present in our Hamiltonian, Eq. (5). Our final result including the error estimate is not in accord with the coupled cluster result from Ref. [42]. However, in the latter work a final uncertainty estimate is not given. According to V. Dzuba [44] correlation contributions beyond the Random Phase Approximation (RPA) in the case of the Xe atom are not greater than \( \approx 3\% \). Our final result indeed shows small total correlation effects and is in agreement with the results from Refs. [30] and [43].

C. \(^{199}\text{Hg}\)

Table IV shows correlated results for the mercury atom. Including only Single and Double excitations for the 12 outermost electrons (shells 5d and 6s) yields a correlation contribution of roughly 10% on top of the DCHF value. This is a significantly greater contribution than the corresponding one in atomic xenon. However, the model SD12 is still not sufficient. Adding full Triple excitations to the wavefunction expansion results in a further 6.5% decrease of \( \alpha_{SM} \) on the absolute. Comparing the model SDT12 with the more approximate expansion SD10, SDT12 shows that the effect of 3 holes in the 5d spinor space is rather unimportant (only 0.2% of the DCHF value) and that it is the combined higher excitations out of the 5d and 6s shells that have to be accounted for. We accomplish this through the model SD10, SDTQ12 where the excitation rank for a maximum of 2 holes in the 5d spinors is maximal. This model yields another 2.4% decrease, on the absolute, at a cutoff of 5 a.u. for the virtual spinors.
Additional effects on $\alpha_{SM}$ from excitations out of the atomic core spinors are accounted for using a virtuals cutoff of 20 a.u. We find that one- and two-hole configurations in the $5p$ shell (model SD6, SD18) contribute a mere 0.5%. On the other hand, one- and two-hole configurations in the $4f$ shell (model S20, SD32) contribute about 3%. It is noteworthy that these two corrections are opposed to each other: Excitations out of $p$ shells decrease electron density in $p$-shell configurations that contribute directly to the generic Schiff moment interaction matrix element in Eq. (17). This leads to an absolute decrease of $\alpha_{SM}$. On the other hand, excitations out of the $f$ shell reduce the screening of nuclear charge on electrons in directly contributing shells, and so lead to an absolute increase of $\alpha_{SM}$. The same effect can be observed when the $5d$ shell is opened for excitations (model SD12 vs. SD2).

We, therefore, obtain our final value from a base value with the largest number of correlated electrons (SD34) improved by a correction for higher combined excitations (Triples and Quadruples) from the valence shells, according to

$$\alpha_{SM}(\text{Final}) = \alpha_{SM}(\text{SD34}/20 \text{au})$$

$$+ \alpha_{SM}(\text{SD10,SDT12}/20 \text{au}) - \alpha_{SM}(\text{SD12}/20 \text{au})$$

$$+ \alpha_{SM}(\text{SD10,SDTQ12}/5 \text{au}) - \alpha_{SM}(\text{SD10,SDT12}/5 \text{au})$$

The first of these two corrections – the one due to combined Triple excitations – amounts to 6.8% of the DCHF value. The second – due to combined Quadruple excitations – amounts to 2.4% of the DCHF value. No higher excitations from the valence shells make a contribution larger than about 0.3%.

The uncertainty estimate for Hg results from adding individual uncertainties for the nuclear-density model (3%), atomic basis set (2%), higher excitation ranks (4%), and the Breit interaction (1%).

Our final result is in agreement with the CI+MBPT result from reference [43]. These two results, however, are in disagreement with the coupled cluster value from Ref. [45], even considering the estimated uncertainties. We have taken into account the leading higher excitations leading to a decrease of $\alpha_{SM}$ on the absolute, i.e., those which are also accounted for in the coupled cluster expansion of Ref. [45]. We have in recent work [31] demonstrated that our present approach and strategy yields results of similar quality as does a coupled cluster expansion of the wavefunction. Also, our total correlation effect of around 17% for Hg is to be contrasted with around 39% correlation effect according to Ref. [45], the latter of which is unusually large for Schiff-moment interactions.
TABLE III. Atomic Schiff moment interaction constant for Hg calculated at Hartree-Fock level including core contribution with Gaussian nuclear density \cite{35} for studying electronic-basis-set convergence; $E_{\text{ext}}$ is set to 0.00024 a.u. Augmented basis sets are built from Dyall’s QZ set including diffuse and correlating functions.

| Model                        | $\alpha_{\text{SM}} \left[ 10^{-17} \text{cm}^2/\text{efm}^3 \right]$ | $\varepsilon_{\text{DCHF}}$ [a.u.] |
|------------------------------|-----------------------------------------------------------------|------------------------------------|
| DZ                           | 6.480                                                           | -19648.85451859                    |
| TZ                           | -1.267                                                          | -19648.89380174                    |
| QZ-34s30p                    | -2.690                                                          | -19648.88766826                    |
| sp-densified QZ-67s59p       | -2.898                                                          | -19648.88651484                    |
| sp-densified+1sp QZ-69s61p   | -2.887                                                          | -19648.88727782                    |
| sp-densified+2sp QZ-71s63p   | -2.887                                                          | -19648.88727627                    |
| sp-densified+3sp QZ-73s65p   | -2.884                                                          | -19648.88727625                    |
| sp-densified+4sp QZ-75s67p   | -2.896                                                          | -19648.88727630                    |
| sp-densified+5sp QZ-77s69p   | -2.897                                                          | -19648.88727619                    |
| sp-densified+6sp QZ-79s71p   | -2.900                                                          | -19648.88727624                    |
| sp-densified+7sp QZ-81s73p   | -2.886                                                          | -19648.88727628                    |
| sp-densified+8sp QZ-83s75p   | -2.886                                                          | -19648.88727631                    |
| Dzuba et al.\cite{30}        | -2.8                                                             | -                                   |
TABLE IV. Atomic Schiff moment interaction constant for Hg including electron correlation effects and the core contribution using the Dyall-cvQZ-69s61p basis set and a Gaussian nuclear density [35]; $E_{\text{ext}}$ is set to 0.00024 a.u.

| Model          | $\alpha_{\text{SM}} \left[ 10^{-17} \text{esm/efm}^3 \right]$ |
|---------------|--------------------------------------------------|
| DCHF          | -2.887                                           |
| SD2/10au      | -2.597                                           |
| SD2/20au      | -2.599                                           |
| SD2/50au      | -2.598                                           |
| SD12/10au     | -2.614                                           |
| SD12/20au     | -2.621                                           |
| SD12/50au     | -2.623                                           |
| SDT12/10au    | -2.426                                           |
| SD10\_SDT12/5au | -2.408                                         |
| SD10\_SDT12/10au | -2.420                                    |
| SD10\_SDT12/20au | -2.425                                  |
| SD10\_SDTQ12/5au | -2.339                                 |
| S6\_SD18/20au | -2.599                                           |
| SD18/20au     | -2.608                                           |
| SD20/10au     | -2.568                                           |
| SD20/20au     | -2.590                                           |
| S20\_SD32/20au | -2.649                                       |
| SD32/20au     | -2.696                                           |
| SD34/20au     | -2.666                                           |
| Final present | -2.40±0.24                                      |
| Dzuba et al. [43] | -2.6                                    |
| Sahoo et al. [45] | -1.77±0.06                                |

D. TlF

Both in Ref. [26] and in our present work a careful and extensive optimization of atomic Gaussian basis sets for the Tl atom in TlF has been carried out. As to be seen in Table[V] the DCHF result for our final $sp$-densified+$1sp$ QZ-69s63p basis set differs from the corrected literature result by Quiney et al. [19, 26] by only 2.2%. We consider this agreement as a further confirmation of the reliability of our technique of basis-set optimization. The DCHF result by Skripnikov et al. [46] is within about 4% of the result by Quiney et al.
TABLE V. Molecular Schiff moment interaction constant for TlF calculated at Hartree-Fock level including core contribution with Gaussian nuclear density [35] at $R = 3.94$ a.u. [17] for studying electronic-basis-set convergence. Augmented basis sets are built from Dyall’s QZ set including diffuse and correlating functions.

| Model                        | $W_{SM}(\text{Tl})$ [a.u.] | $\varepsilon_{\text{DCHF}}$ [a.u.] |
|------------------------------|----------------------------|-----------------------------------|
| QZ-34s31p                    | 42877                      | $-20374.47704191$                 |
| sp-densified QZ-67s61p       | 30737                      | $-20374.4757145$                  |
| **sp-densified+1sp QZ-69s63p** | **45419**                 | $-20374.47660904$                 |
| sp-densified+2sp QZ-71s65p   | 45540                      | $-20374.47660743$                 |
| sp-densified+3sp QZ-73s67p   | 45584                      | $-20374.47660800$                 |
| sp-densified+4sp QZ-75s69p   | 45602                      | $-20374.47660786$                 |
| sp-densified+5sp QZ-77s71p   | 45578                      | $-20374.47660735$                 |
| sp-densified+6sp QZ-79s73p   | 45594                      | $-20374.47660803$                 |
| sp-densified+7sp QZ-81s75p   | 45602                      | $-20374.47660837$                 |
| sp-densified+8sp QZ-83s77p   | 45584                      | $-20374.47660780$                 |
| Quiney et al. (DCHF)         | 46444                      |                                   |
| Skripnikov et al. (DCHF)     | 48377                      |                                   |

* Value from Ref. [26] corrected in Ref. [19] for the use of a more accurate operator for the Schiff-moment interaction.

In Table VI we compile correlated Schiff-moment interactions for TlF using various CI models. The main correction to the DCHF result comes from valence correlations among the $6s$ (Tl) and $2p$ (F) electrons (model SD8), quenching the interaction constant by nearly 9%. Full Triple excitations out of these shells further reduce the value by almost 2% and in addition by a similar amount when a leading set of Quadruple excitations is added to the expansion, model 0.8auSDTQ8_SDT8 (where up to four particles are allowed in shells below an energy cutoff of 0.8 a.u.). Correlation contributions from the shells $1s, 2s$ (F) and $5s, 5p$ (Tl) are seen to be small, amounting to an increase of $W_{SM}$ by only about 1.5%.

For TlF we obtain our final best result from a base value obtained with the model cvQZ+/SDT10_10au and adding corrections for Quadruple excitations from the valence shells, correlation contributions from $1s$ (F) and $5s, 5p$ (Tl) shells and a
TABLE VI. Molecular Schiff moment interaction constant for the thallium nucleus in TlF including electron correlation effects and the core contribution using the Dyall-cvQZ-69s63p basis set (denoted cvQZ+ in the Table) and a Gaussian nuclear density \[35\] at \(R = 3.94\) a.u. \[48\]

| Basis set/Model                      | \(W_{SM}(\text{Tl})\) [a.u.] |
|-------------------------------------|-------------------------------|
| cvQZ/DCHF                           | 42877                         |
| cvQZ+/DCHF                          | 45419                         |
| cvQZ+/SD8.5.5au                     | 40779                         |
| cvQZ+/SD8.10au                      | 41198                         |
| cvQZ+/SD8.20au                      | 41431                         |
| cvQZ+/SD8.40au                      | 41438                         |
| cvQZ+/SDT8.5.5au                    | 39954                         |
| cvQZ+/SDT8.10au                     | 40314                         |
| cvQZ+/SDT8.20au                     | 40495                         |
| cvQZ+/0.8auSDTQ8 SDT8.5.5au         | 38863                         |
| cvQZ+/SD10.10au                     | 41584                         |
| cvQZ+/SDT10.10au                    | 40544                         |
| cvQZ+/SD16.10au                     | 41762                         |
| cvQZ+/SD18.10au                     | 41838                         |
| cvQZ+/SD20.10au                     | 41858                         |
| **Final present**                   | **39967±3600**                |

Flambaum et al. \[19, 27\] (CC) 40539
Skripnikov et al. (CCSD(T)) \[46\] 37192
Abe et al. \[49\] (CC) 41136

The uncertainty estimate for TlF results from adding individual uncertainties for the nuclear-density model (3%), atomic basis set (2%), higher excitation ranks (2%), inner-shell correlations (1%) and the Breit interaction (1%).

Our final best result is in agreement with both the operator-shifted results of Flambaum et al. in Refs. \[19, 27\] and the recent CCSD(T) calculation by Skripnikov et al. \[46\].
IV. CONCLUSIONS

Using the $\mathcal{P}, \mathcal{T}$-violating energy shift $\Delta \varepsilon$ from the most recent measurements on the present systems and our calculated interaction constants we can determine the nuclear Schiff moment $S$ itself, in the context of a single-source assumption. It results from the relation

$$\Delta \varepsilon = 2W_{SM} S$$

(21)

where using our final central value for $W_{SM}$ from Table VI and the measured frequency shift of $(1.4 \pm 2.4) \times 10^{-4}$ Hz from Ref. 28 yields

$$S^{(205\text{Tl})} = (3.9 \pm 6.8) \times 10^{-11} \text{ e fm}^3$$

(22)

for the Schiff moment of the $^{205}\text{Tl}$ nucleus. The CENTREX collaboration [18] aims at a significant increase in sensitivity to hadronic $\mathcal{T}$-violating fundamental interactions which – combined with the recent results for $W_{SM}$ – will lead to stronger constraints on the nuclear Schiff moment in case of a null measurement with tighter uncertainties.

The limit on the nuclear Schiff moment can be used to infer limits on the CPV pion-nucleon couplings, the QCD $\Theta$ parameter and chromo-EDMs following the relations in Ref. [19].

A stronger constraint than the one in Eq. 22 can be placed on the Schiff moment of the $^{199}\text{Hg}$ nucleus. Using the upper bound on the Hg EDM of

$$|d_{\text{Hg}}| < 7.4 \times 10^{-30} \text{ e cm}$$

(23)

from Ref. [24] and our central value for the Schiff-moment interaction from Table IV yields an upper bound to the Hg Schiff moment

$$|S_{\text{Hg}}| < 3.1 \times 10^{-13} \text{ e fm}^3$$

(24)

This is the same value as the one proposed in Ref. [24] where an average over various uncorrelated and correlated Schiff-moment interactions from the literature had been used. In the present case a rigorously calculated interaction parameter $\alpha_{\text{SM}}(\text{Hg})$ including the effects of interelectron correlations in the Hg atom ground state replaces that average value which it happens to match. The nuclear Schiff moment of $^{129}\text{Xe}$ and $^{199}\text{Hg}$ has recently been calculated as a function of the strong $\pi$-meson-nucleon interaction constants [50]. Combined with these dependencies our results can be used to constrain these interaction constants.
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Appendix A: Details on Basis-Set Optimization for Xe

TABLE VII. Atomic Schiff moment interaction constant \( \alpha_{\text{SM}} \) for Xe calculated at Hartree-Fock level including core contribution with Gaussian nuclear density \( [35] \) for studying electronic-basis-set convergence using several \( s \) and \( p \) spaces densifications and addition of denser and more diffuse \((s,p)\) function pairs; \( E_{\text{ext}} \) is set to 0.0003 a.u. Augmented basis sets are built from Dyall’s QZ with diffuse and correlating functions. +1sp means one denser \( s \) and \( p \) function, one more diffuse \( s \) and \( p \) function have been added to the basis set.

| Model                  | \((s,p)\) space | \(\alpha_{\text{SM}}\) \(10^{-17}\,\text{cm}^3/\text{fm}^3\) | \(\varepsilon_{\text{DCHF}}\) [a.u.] |
|------------------------|-----------------|--------------------------|-------------------------------|
| QZ                     | 34s28p          | 0.318                    | -7446.895409376               |
| QZ +1sp*               | 36s30p          | 0.369                    | -7446.895386026               |
| Simple sp-densified QZ | 67s55p          | 0.314                    | -7446.895379750               |
| Simple sp-densified QZ +1sp | 69s57p      | 0.373                    | -7446.895401869               |
| Double sp-densified QZ | 133s109p        | 0.362                    | -7446.895392349               |
| Double sp-densified QZ +1sp | 135s111p     | 0.359                    | -7446.895401000               |
| Double sp-densified QZ +2sp | 137s113p    | 0.354                    | -7446.895401764               |
| Double sp-densified QZ +3sp | 139s115p    | 0.369                    | -7446.895401848               |
| Double sp-densified QZ +4sp | 141s117p    | 0.372                    | -7446.895401815               |
| Double sp-densified QZ +5sp | 143s119p    | 0.375                    | -7446.895401836               |
| Double sp-densified QZ +6sp | 145s121p    | 0.376                    | -7446.895401842               |
| Double sp-densified QZ +7sp | 147s123p    | 0.375                    | -7446.895401832               |
| Double sp-densified QZ +8sp | 149s125p    | 0.376                    | -7446.895401826               |
| Double sp-densified QZ +9sp | 151s127p    | 0.376                    | -7446.895401837               |
| Double sp-densified QZ +10sp | 153s129p   | 0.376                    | -7446.895401813               |
| Triple sp-densified QZ | 265s217p        | 0.051                    | -7446.895394063               |
| Triple sp-densified QZ +1sp | 267s219p    | 0.684                    | -7446.895399110               |
TABLE VIII. Atomic Schiff moment interaction constant $\alpha_{SM}$ $\left[10^{-17} \text{e cm}^3/\text{e fm}^3\right]$ for Xe calculated at Hartree-Fock level including core contribution with Gaussian nuclear density [35] for studying electronic-basis-set convergence by addition of denser $(s,p)$ pairs versus addition of denser $(s,p)$ and more diffuse $(s,p)$ pairs; $E_{ext}$ is set to 0.0003 a.u. Augmented basis sets are built from Dyall’s QZ set with diffuse and correlating functions.

| Model           | Addition of $(s,p)$ dense pairs | Addition of $(s,p)$ dense/diffuse pairs |
|-----------------|---------------------------------|----------------------------------------|
| QZ              | 34s28p 0.318 $-7446.895409376$  | 34s28p 0.318 $-7446.895409376$         |
| QZ +1sp         | 35s29p 0.368 $-7446.895385894$  | 36s30p 0.369 $-7446.895384072$         |
| QZ +2sp         | 37s31p 0.369 $-7446.895383784$  | 40s34p 0.370 $-7446.895389564$         |
| QZ +3sp         | 38s32p 0.369 $-7446.895383738$  | 42s36p 0.371 $-7446.895389157$         |
| QZ +4sp         | 39s33p 0.371 $-7446.895383742$  | 44s38p 0.370 $-7446.89538987$          |
| QZ +5sp         | 40s34p 0.370 $-7446.895383941$  | 46s40p 0.370 $-7446.895384141$         |
| QZ +6sp         | 41s35p 0.370 $-7446.895384108$  | 48s42p 0.372 $-7446.895384163$         |
| QZ +7sp         | 42s36p 0.370 $-7446.895383571$  | 50s44p 0.376 $-7446.895382476$         |
| sp-densified QZ | 67s55p 0.314 $-7446.895379750$  | 67s55p 0.314 $-7446.895379750$         |
| sp-densified QZ+1sp | 68s56p 0.378 $-7446.895401868$  | 69s57p 0.373 $-7446.895401869$         |
| sp-densified QZ+2sp | 69s57p 0.375 $-7446.895401802$  | 71s59p 0.375 $-7446.895401810$         |
| sp-densified QZ+3sp | 70s58p 0.376 $-7446.895401801$  | 73s61p 0.375 $-7446.895401762$         |
| sp-densified QZ+4sp | 71s59p 0.376 $-7446.895401801$  | 75s63p 0.375 $-7446.895401779$         |
| sp-densified QZ+5sp | 72s60p 0.378 $-7446.895401802$  | 77s65p 0.376 $-7446.895401790$         |
| sp-densified QZ+6sp | 73s61p 0.375 $-7446.895401802$  | 79s67p 0.375 $-7446.895401745$         |
| sp-densified QZ+7sp | 74s62p 0.375 $-7446.895401799$  | 81s69p 0.374 $-7446.895401770$         |
| sp-densified QZ+8sp | 75s63p 0.376 $-7446.895401800$  | 83s71p 0.375 $-7446.895401790$         |
FIG. 1. $\varepsilon_{\text{DCHF}}$ variation from sp-densified QZ for Xe.