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$\mathcal{P}, \mathcal{T}$-odd weak neutral current interactions in the TlF molecule

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Leptoquark models may explain deviations from the standard model observed in decay processes involving heavy quarks at high-energy colliders. Such models give rise to low-energy parity- and time-reversal-violating phenomena in atoms and molecules. One of the leading effects among these phenomena is the nucleon-electron tensor-pseudotensor interaction when the low-energy experimental probe uses a quantum state of an atom or molecule predominantly characterized by closed electron shells. In the present paper the molecular interaction constant for the nucleon-electron tensor-pseudotensor interaction in the thallium-fluoride molecule—used as such a sensitive probe by the CeNTREX collaboration [O. Grasdijk *et al.*, Quantum Sci. Technol. **6**, 044007 (2021)]—is calculated employing highly correlated relativistic many-body theory. Accounting for up to quintuple excitations in the wave-function expansion the final result is $W_T(\mathrm{Tl}) = -6.25 \pm 0.31$ $(10^{-13}\langle\Sigma\rangle_A$ a.u.) Interelectron correlation effects on the tensor-pseudotensor interaction are studied rigorously in a molecule.

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I. INTRODUCTION

In the last decade tests of observables in decays involving *b* (*beauty, bottom*) quarks have shown deviations from the predictions offered by the standard model (SM) of elementary particles ([1–3] and references therein). These observed deviations could be explained through the existence of new physics (NP) particles, the leptoquarks (LQs) [4–8], among other possible models.

Electric dipole moments (EDMs) of atoms and molecules are highly sensitive low-energy probes [9] of charge-parity ($\mathcal{CP}$) violating physics beyond that already implemented in the SM elementary particles. Under the assumption that the considered atomic or molecular quantum state has predominantly closed electron shells, scalar leptoquark models with a dominant contribution through light quarks amplify [10,11] a $\mathcal{CP}$-violating (CPV) tensor-pseudotensor nucleon-electron (T-PT-ne) interaction over other possible sources of an atomic-scale EDM, with the exception [12] of a collective nuclear effect, the nuclear Schiff moment [13]. By contrast, in open-shell atomic or molecular states the contribution due to the CPV scalar-pseudoscalar nucleon-electron (S-PS-ne) interaction is the leading one among the possible nucleon-electron four-fermion interactions. In other words, EDM measurements on closed-shell atomic or molecular systems with a very high sensitivity to underlying $\mathcal{CP}$-violating NP probe such scalar leptoquark models. This is achieved by interpreting the measured EDM—or measured upper bound on an EDM—in terms of the $\mathcal{CP}$-odd T-PT-ne parameter C_T and an atomic or molecular interaction constant that is calculated by relativistic many-body theory.

The complex system delivering the most sensitive low-energy probe of light-quark scalar leptoquark models at the moment is the mercury (Hg) atom [14]. Efforts are currently undertaken to significantly improve upon the sensitivity of the Hg EDM measurement by using the thallium-fluoride (TlF) molecule in a cold molecular-beam experiment (CeNTREX collaboration [15]). This molecular EDM measurement on the electronically closed-shell TlF ground state can be interpreted [16] in terms of the nuclear Schiff moment of the ^{205}Tl nucleus and in terms of more fundamental CPV parameters, the strong π-meson exchange constants, the QCD Θ parameter, chromo-EDMs, and neutron and proton EDMs [17]. In addition, the stable isotope ^{205}Tl has nuclear spin quantum number $I = \frac{1}{2}$ [18,19] which makes it sensitive to the nuclear-spin-dependent T-PT-ne interaction.

However, ^{205}Tl is not located in the region of strongly octupole-deformed nuclei [20]. Indeed, it has been shown [17,21] that the dependency of the Schiff moment of ^{205}Tl on underlying CPV parameters such as QCD Θ and the π-meson exchange constants is some orders of magnitude smaller than the same dependency of the Schiff moments of ^{225}Ra or ^{223}Fr$^+$. This relatively feeble dependency is also established for ^{199}Hg. Thus, the interpretation of the TlF EDM in terms of the molecular T-PT-ne interaction in that system may even be more important than in terms of the Schiff moment of its ^{205}Tl nucleus.

The earliest calculation of the T-PT-ne interaction constant in a molecule has been reported by Hinds *et al.* in 1976 [22], followed by Coveney and Sandars in 1983 [23] as it happens for the TlF molecule. Full-fledged relativistic Dirac-Hartree-Fock calculations [24,25] gave results factors of 4–5 greater than the earlier calculations. Gaul and Berger [25] also report an estimate of electron correlation effects on the T-PT-ne interaction constant in TlF.

The main purpose of this paper is to report on a high-level calculation of the T-PT-ne interaction constant of ground-state TlF using relativistic many-body theory of general excitation rank (Sec. III). Electron correlation effects on the T-PT-ne

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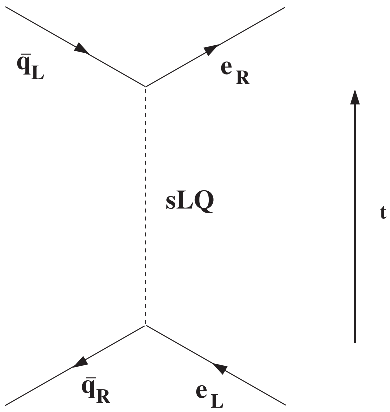

FIG. 1. Tree-level diagram for the exchange of a scalar (spin-zero) leptoquark (sLQ) between an antiquark and an electron; this process resembles Bhabha scattering in quantum electrodynamics when sLQ $\longrightarrow \gamma$ and $\bar{q} \longrightarrow \bar{e}$; R and L stand for fermion chirality.

interaction are calculated rigorously for the first time in a molecule. In addition, the theory of atomic and molecular T-PT-ne interactions is reviewed and corresponding interaction constants are defined in a common framework for atoms and molecules (Sec. II). Such a common framework is a distinct advantage since it removes some problems arising from differing conventions in global analyses of $\mathcal{CP}$-violation effects [26,27]. In the final section (Sec. IV) the expected impact of the present findings is discussed in particular considering future measurements of the TlF EDM.

II. THEORY

A. Particle physics

As Barr expounds in Ref. [10] the LQ exchange displayed in Fig. 1 leads to $\mathcal{P}$- and $\mathcal{T}$-odd nucleon-electron four-fermion coupling coefficients $C_{S(q)}, C_{P(q)}$, and $C_{T(q)}$ at the quark (q) level which are all nonvanishing. S here denotes nucleon-electron scalar-pseudoscalar (S-PS), P denotes nucleon-electron pseudoscalar-scalar (PS-S), and T denotes nucleon-electron tensor-pseudotensor (T-PT) coefficients, respectively.

If light quarks (u, d) make the dominant contribution to the $\mathcal{P}$- and $\mathcal{T}$-odd couplings in such models then the ratio of the T-PT and S-PS coefficients is found [10] to be $\frac{C_T}{C_S} \approx \frac{1}{10}$. This suppression of the T-PT contribution by about one order of magnitude relative to the S-PS contribution to the $\mathcal{P}$- and $\mathcal{T}$-odd effects, however, only holds in a complex system if the corresponding interaction constants are of the same order of magnitude. Conversely, a suppression of the S-PS contribution to a complex-system EDM can be achieved by using a quantum state with predominantly paired (closed) electron shells where S-PS contributions cancel pairwise in lowest order of perturbation theory.

B. Isotope-specific effective many-electron T-PT-ne interaction Hamiltonian

The T-PT-ne atomic interaction constant can be defined starting from an effective theory at the level of nucleons [10]. An effective first-quantized Hamiltonian for a single electron can be written as [28]

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \frac{\imath G_F}{\sqrt{2}} \sum_N C_T^N \, \rho_N(\mathbf{r})\gamma^0\sigma_{N\mu\nu}\gamma^5\sigma^{\mu\nu}, \tag{1}$$

where $\rho_N(\mathbf{r})$ denotes the probability density of the corresponding nucleon at the position of the electron and the electronic Dirac matrix γ^0 originates in the field creator $\overline{\psi}$. C_T^N are Wilson coefficients determined by $\mathcal{CP}$-odd interactions at higher energies. The term $\sigma_{N\mu\nu}\gamma^5\sigma^{\mu\nu}$ satisfies the following identity:

$$\sigma_{N\mu\nu}\gamma^5\sigma^{\mu\nu} = 2\gamma_N^0\boldsymbol{\gamma}_N\cdot\boldsymbol{\Sigma} + 2\gamma^0\boldsymbol{\Sigma}_N\cdot\boldsymbol{\gamma}, \tag{2}$$

where $\boldsymbol{\Sigma}_N$ denotes the nuclear and $\boldsymbol{\Sigma}$ denotes the electronic spin matrix. The electronic expectation value $\langle\psi|\boldsymbol{\Sigma}|\psi\rangle$ of the first term on the right-hand side of Eq. (2) is strictly zero if ψ is a closed-shell wave function for a many-electron state. The T-PT-ne Hamiltonian used in the present paper is then written as

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \frac{\imath G_F}{\sqrt{2}} \sum_N 2C_N^T \, \boldsymbol{\Sigma}_N\cdot\boldsymbol{\gamma}\rho_N(\mathbf{r}). \tag{3}$$

In terms of nuclear spin I, $\langle\boldsymbol{\Sigma}\rangle = \langle\Sigma\rangle\,\mathbf{I}/I$. Introducing a nuclear isotope-specific $\mathcal{CP}$-odd parameter C_T^A Eq. (3) becomes

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \frac{2\imath G_F}{\sqrt{2}} \,\rho(\mathbf{r})\, C_T^A \,\langle\Sigma\rangle_A \frac{\mathbf{I}\cdot\boldsymbol{\gamma}}{I} \tag{4}$$

where $\rho(\mathbf{r})$ now refers to a nuclear density assumed to be equal for protons and neutrons.

In a setup with rotational symmetry around the z axis, the 1 and 2 components of $\langle\boldsymbol{\Sigma}\rangle$ vanish. Considering furthermore a nuclear state $|I, M_I = I\rangle$ and integrating over the nuclear coordinates, the Hamiltonian can be written as

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \imath\sqrt{2}G_F \, C_T^A \,\langle\Sigma\rangle_A \gamma_3 \,\rho(\mathbf{r}). \tag{5}$$

On the basis of this effective single-electron Hamiltonian the molecular T-PT-ne interaction can be defined. In an n-electron framework, Eq. (5) becomes

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \imath\sqrt{2}G_F \, C_T^A \,\langle\Sigma\rangle_A \sum_{j=1}^{n} (\gamma_3)_j \,\rho(\mathbf{r}_j). \tag{6}$$

C. Molecular T-PT-ne interaction constant

The T-PT-ne interaction is evaluated through lowest order in perturbation theory. The wave function of an electronic state E is the approximate solution of an eigenvalue problem

$$\hat{H}^{\text{DC}}\psi_E^{(0)} = \varepsilon_E \,\psi_E^{(0)} \tag{7}$$

where $\hat{H}^{\text{DC}}$ is the Dirac-Coulomb (DC) Hamiltonian

$$\hat{H}^{\text{DC}} = \sum_j^n \left[c\,\boldsymbol{\alpha}_j\cdot\mathbf{p}_j + \beta_j c^2 - \sum_K^2 \frac{Z_K}{r_{jK}}\mathbb{1}_4 \right] + \sum_{k>j}^n \frac{1}{r_{jk}}\mathbb{1}_4 + V_{KL} \tag{8}$$

for a diatomic molecule with n electrons, where $\boldsymbol{\alpha}$ and β are electronic Dirac matrices, K runs over nuclei, and V_{KL} is the classical electrostatic potential energy for the two Born-Oppenheimer-fixed nuclei.

$\psi_E^{(0)}$ is linearly expanded:

$$\left|\psi_E^{(0)}\right\rangle = \sum_{L=1}^{\dim\mathcal{F}^t(M,n)} c^E_{(M_J),L}\,(\mathcal{S}\overline{\mathcal{T}})^E_{(M_J),L}|\rangle \tag{9}$$

where $|\rangle$ is the true vacuum state, $\mathcal{F}^t(M,n)$ is the symmetry-restricted sector of Fock space (M_J subspace with J total electronic angular momentum) 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, and $\overline{\mathcal{T}} = a_{\overline{l}}^\dagger a_{\overline{m}}^\dagger a_{\overline{n}}^\dagger \ldots$ is a string of creation operators of time-reversal transformed spinors. The determinant expansion coefficients for state E, $c^E_{(M_J),L}$, are obtained through relativistic general-excitation-rank configuration-interaction theory as detailed in Refs. [29,30].

The energy shift of state E in a molecule due to the T-PT-ne interaction is now given as an expectation value over the effective Hamiltonian in Eq. (6)

$$\Delta\varepsilon_E = \left\langle\psi_E^{(0)}\middle|\hat{H}^{\text{eff}}_{\text{T-PT-ne}}\middle|\psi_E^{(0)}\right\rangle = W_T\, C_T^A \tag{10}$$

and expressed in terms of a molecular interaction constant W_T and the effective $\mathcal{CP}$-odd parameter C_T^A. It then follows from Eqs. (6) and (10) that the molecular T-PT-ne interaction constant is

$$W_T(X) = \sqrt{2}G_F\,\langle\Sigma\rangle_A\left\langle\psi_E^{(0)}\middle|\imath\sum_{j=1}^{n}(\gamma_3)_j\,\rho_X(\mathbf{r}_j)\middle|\psi_E^{(0)}\right\rangle \tag{11}$$

where X denotes the individual atomic nucleus in the molecule. The diagonal matrix element $\langle\psi|\gamma_3|\psi\rangle$ for a closed-shell electronic state is nonzero (for a detailed discussion see Ref. [28]). W_T has the dimension of energy.

For purposes of comparison the atomic case [31] is briefly reviewed. The atomic electric dipole moment due to a tensor-pseudotensor interaction can be written as

$$d_a = C_T^A\,\alpha_{C_T} \tag{12}$$

where

$$\alpha_{C_T} := \frac{\left\langle\hat{H}^{\text{eff}}_{\text{T-PT-ne}}\right\rangle_{\psi^{(0)}(E_{\text{ext}})}}{E_{\text{ext}}} \tag{13}$$

and $\psi^{(0)}(E_{\text{ext}})$ is the field-dependent atomic wave function of the state in question. α_{C_T} has physical dimensions of an electric dipole moment and is evaluated in the quasilinear regime (see also Ref. [32]) with very small external electric fields E_{ext}.

III. TENSOR-PSEUDOTENSOR INTERACTION IN MOLECULES

A. Consistency test

The evaluation of Eq. (11) makes use of the same implementation reported previously for corresponding atomic matrix elements [31]. A rigorous test of the molecular interaction constant is here carried out by calculating the atomic T-PT-ne interaction in the Xe atom in two different ways.

(1) A Xe atom is subjected to a finite external electric field along the z axis and α_{C_T} is calculated using Eq. (13). In a cvTZ basis set this yields a DC Hartree-Fock (HF) value of $\alpha_{C_T} = 1.0422 \times 10^{-12}\langle\Sigma\rangle_A$ a.u.

(2) A Xe atom is placed in the electric field of an F$^-$ ion at a distance of 40 a.u. and W_T is calculated through Eq. (11) in a molecular calculation. De facto, this corresponds to the calculation of the interaction constant in a stretched XeF$^-$ ion. The electric field of the F$^-$ ion at the position of the Xe atom is then $E(\text{F}^-) = 0.000\,625$ a.u. Through calculation, $\frac{W_T(\text{Xe})}{E(\text{F}^-)} = 1.0417 \times 10^{-12}\langle\Sigma\rangle_A$ a.u., confirming that the molecular evaluation is consistent with an atomic calculation. The slight difference between the results of the atomic and the molecular evaluation can be explained by the fact that in the atomic case the external field is homogeneous across the Xe atom whereas it is not perfectly homogeneous when created by an F$^-$ ion at a distance.

B. Tensor-pseudotensor interaction in TlF

1. *Technical details*

Atomic Gaussian basis sets of double-zeta (DZ), triple-zeta (TZ), and quadruple-zeta (QZ) quality are used for both atoms, in the case of Tl the Dyall basis sets [33–35] including valence-correlating, $5d$-correlating, and $4f$-correlating exponents and in the case of F the cc-pVNZ sets from the EMSL library [36]. Electronic-structure calculations are carried out using a locally modified version of the DIRAC program package [37]. For correlated calculations beyond DC Hartree-Fock (DCHF) the Kramers-restricted configuration-interaction module [30,38] is employed for obtaining relativistic molecular wave functions and for the evaluation of the T-PT-ne interaction constants. For the case of Hartree-Fock theory the ground-state Slater determinant comprises the wave function $\psi_E^{(0)}$ in Eq. (11). All single-point Born-Oppenheimer calculations are carried out at the experimental equilibrium internuclear distance of $R_e = 3.94$ a.u. [39].

2. *Hartree-Fock theory*

In Table I I compare various results calculated through Hartree-Fock theory. Basis-set effects are seen to be very

TABLE I. T-PT-ne interaction constant in TlF($^1\Sigma_0$) at $R = 3.94$ a.u. from Hartree-Fock theory.

Model	W_T(Tl) (kHz $\langle\Sigma\rangle_A$)	Total energy
Hartree-Fock[a]	−1.34	
Hartree-Fock[b]	−0.851	
Dirac-Coulomb HF[c]	−4.641	−20374.4108
cGHF-ZORA-wr[d]	−4.690	
DZ/DCHF	−4.601	−20374.41122770
TZ/DCHF	−4.673	−20374.46576781
QZ/DCHF	−4.684	−20374.47704191
QZ+dens+sp/DCHF	−4.684	−20374.47660904

[a] Converted EDM in terms of C_T by Hinds *et al.* [22] using the external electric field and interaction constant reported *ibidem*.

[b] Value reported by Cho *et al.* [40] which is the corrected result from Ref. [23].

[c] Value from Ref. [24] in converted units.

[d] Value from Ref. [25] with adapted sign, $R = 3.93$ a.u.

small, and the largest basis used in the present paper—augmented for the calculation of Schiff-moment interactions [16]—does not improve upon the standard QZ basis set. For this reason the correlated calculations in the following section are carried out with the QZ basis for both atoms.

The present results agree with the 1991 results by Quiney *et al.* [24] which have also been obtained in the framework of Dirac theory. More converged results concerning atomic basis sets have been obtained by Gaul and Berger [25] which are in excellent agreement with the present best DCHF result. The total energies given in Table I and the trend for W_T with basis sets of increasing extent strongly suggest that the difference between the present results and those from Quiney *et al.* is due to the higher quality of Tl basis sets used in the present paper. From the consistency of these results it is also evident that the early method employed by Hinds *et al.* [22] and Coveney and Sandars [23] in a framework that does not account for the relativistic character of the s- and p-type TlF spinors in a rigorous fashion produces large errors. Not surprisingly, T-PT-ne interactions should be calculated in a fully relativistic atomic or molecular framework.

3. Correlated theory

Results for the T-PT-ne interaction constant from a systematic series of calculations including interelectron correlation effects are compiled in Table II. As expected, the main correlation effect comes from the valence electrons in the Tl(6s) and F(2p_z) shells and amounts to about −11% in magnitude (from model QZ/SDTQ4). Like other $\mathcal{P}, \mathcal{T}$-odd effects in molecules [16,41] also the T-PT-ne interaction is dominated by s-p mixing in the valence shells of the heavy target atom. Interestingly, correlations with and among the remaining valence electrons in the F(2$p_{x,y}$) shells are not negligible and result in another shift of W_T of about −3.3% in magnitude (model QZ/SDTQ8). Since full triple and full quadruple excitations among the eight valence electrons change W_T by more than 5% (relative to QZ/SD8) it is not unreasonable to assume that even higher excitations could result in further non-negligible corrections. This is, however, not the case, as seen from the comparison of models QZ/S4_SDTQ8 and QZ/S4_SDTQQ8 which yield almost identical results. In addition, the chosen virtual cutoff of 6.5 a.u. for valence correlations is seen to be sufficiently accurate through comparison with results at 11 a.u. The value of −6.12 ($10^{-13}\langle\Sigma\rangle_A$ a.u.) for W_T can, therefore, be considered as sufficiently converged as concerns valence-electron correlations.

Effects due to core-valence and core-core interelectron correlations follow a familiar pattern [16] for $\mathcal{P}, \mathcal{T}$-odd effects in closed-shell systems. When electrons from heavy-atom shells of s and p type are involved in excitations W_T drops because the mean density in shells contributing directly to the $\mathcal{P}, \mathcal{T}$-odd effect is reduced (model SD26 relative to SD20 and SD28 relative to SD26). By contrast, when electrons are removed from occupied shells with higher angular momentum W_T increases (model SD18 relative to SD8) since in

TABLE II. Interelectron correlation effects on the T-PT-ne interaction constant in TlF($^1\Sigma_0$) at $R = 3.94$ a.u. and comparison with the literature; present models are denoted as "Basis set/CI model_cutoff in virtual space (au = a.u.)." A CI model denoted "SDm_SDTQn" means that single, double, triple, and quadruple replacements out of shells with n electrons are performed where subshells with m electrons are restricted to single and double replacements; $n = 4$ corresponds to the Tl(6s) and F(2p_z) shells, $n = 8$ corresponds to the Tl(6s) and all F(2p) shells, $n = 18$ adds Tl(5d) to $n = 8$, $n = 20$ adds F(2s) to $n = 18$, $n = 26$ adds Tl(5p) to $n = 20$, $n = 28$ adds Tl(5s) to $n = 26$, and $n = 36$ adds Tl(4s, 4p) to $n = 28$; $m = 4$ denotes the F(2$p_{x,y}$) subshells.

	W_T(Tl)		
Model	($10^{-13}\langle\Sigma\rangle_A$ a.u.)	(kHz $\langle\Sigma\rangle_A$)	Total energy
QZ/DCHF	−7.12	−4.68	−20374.47704191
QZ/SD4_6.5au	−6.40	−4.21	−20374.53183162
QZ/SDTQ4_6.5au	−6.33	−4.17	−20374.53321310
QZ/S4_SDTQ8_6.5au	−6.08	−4.00	−20374.61641168
QZ/SDTQ4_SDTQ8_6.5au	−6.12	−4.03	−20374.69523546
QZ/S4_SDTQQ8_6.5au	−6.07	−4.00	−20374.61647310
QZ/SD8_2au	−6.27	−4.12	−20374.58577569
QZ/SD8_6.5au	−6.47	−4.26	−20374.67877868
QZ/SD8_11au	−6.46	−4.25	−20374.67958679
QZ/SDT8_6.5au	−6.47	−4.26	−20374.68490950
QZ/SDTQ8_6.5au	−6.12	−4.03	−20374.69523546
QZ/SD18_6.5au	−6.58	−4.33	−20374.98617776
QZ/SD20_6.5au	−6.61	−4.35	−20375.05000858
QZ/SD26_6.5au	−6.55	−4.31	−20375.15736493
QZ/SD28_6.5au	−6.49	−4.27	−20375.17819744
QZ/SD28_11au	−6.57	−4.32	−20375.32222400
QZ/SD28_18au	−6.59	−4.33	−20375.37322100
QZ/SD28_30au	−6.60	−4.34	−20375.39255661
QZ/SD28_50au	−6.60	−4.34	−20375.40305619
QZ/SD36_18au	−6.59	−4.34	−20375.38490094
Final	−6.25	−4.11	

that case the mean density in shells contributing directly to the $\mathcal{P}, \mathcal{T}$-odd effect is increased. These cancellations make the total correlation effect from core shells down to Tl($5s$) quite small in total at the SD level, less than 20% of valence correlation effects. This and the rising energy denominators for contributions from core spinors strongly suggest that correlation effects from deeper core shells will make significantly smaller contributions. Indeed, the model SD36 which includes correlations from the Tl($4s, 4p$) electrons changes W_T by less than 0.1%. Similar insignificance can thus also be assumed for higher excitation ranks up to full quadruple excitations involving the core electrons.

The final value for W_T is calculated as follows. The base value is comprised by the highest-level correlated calculation with the greatest number of active electrons, model QZ/SD28_50au. This value is corrected by adding correlation effects from excitation ranks up to full quadruple excitations among the valence electrons. Formally, this means

$$W_T(\text{final}) = W_T(\text{QZ/SD28\_50au}) + W_T(\text{QZ/SDTQ8\_6.5au}) - W_T(\text{QZ/SD8\_6.5au}). \tag{14}$$

The final value, listed in Table II, is thus $W_T = -6.25$ ($10^{-13}\langle\Sigma\rangle_A$ a.u.), showing that the total modification due to interelectron correlations is -12.2% in magnitude. The statement by Quiney *et al.* ([24], p. 939) that it is "unlikely that electron correlation will have a significant effect on … T [the T-PT-ne interaction constant]" was evidently premature.

The uncertainty in the present final result is estimated from an unweighted sum over individual uncertainties for effects contributing to the T-PT interaction. An individual uncertainty is defined as the difference between the most accurate and the second most accurate model used in the present paper for a given effect. This results in individual uncertainties of 1% for atomic basis sets, 1% for valence-electron correlations, 1% for core-correlation effects, and 1% for the cutoff in the virtual spinor space. To this is added 1% for neglecting the Breit and radiative corrections [42]. The final 5% is a conservative uncertainty estimate since partial cancellations among the different neglected contributions are very likely.

4. W_T as a function of internuclear distance

Figure 2 shows the electronic ground-state potential of TlF along with essential results for the T-PT interaction at Hartree-Fock and a selected level of correlated theory that yields a result close to the final value for W_T. From the functions $W_T(R)$ it is clear that the maximum polarization of the TlF molecule is achieved around $R = 3.6$ a.u. which is significantly shorter than the equilibrium bond length R_e. This observation has also been made for other $\mathcal{P}, \mathcal{T}$-violating interaction constants in other diatomic molecules [43]. Another interesting observation concerns the variation of W_T with molecular vibration. Between the classical turning points in the respective vibrational ground state ΔW_T is on the order of 5%, but since $W_T(R)$ is near linear in that regime an averaging over W_T results in a negligible vibrational correction.

The vis-à-vis with literature results shows that these are all outside of the uncertainty bar of the present final result. Electron correlation effects on W_T in TlF are significant and

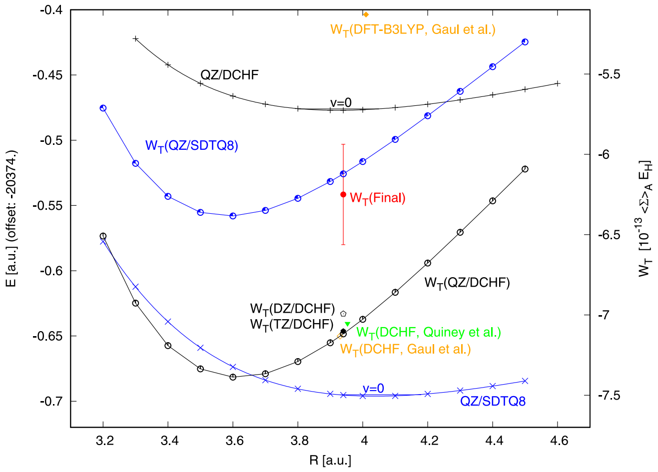

FIG. 2. Potential-energy curves (left-hand scale, polynomial-fitted curves) and T-PT interaction constants (right-hand scale, connected data points) from selected models and comparison with literature. The final value for W_T is given with an uncertainty bar calculated as described in the text.

should be calculated with a high-level fully relativistic electron correlation method.

5. T-PT vs S-PS interaction

For the leading closed-shell determinant (active molecular spinors and time-reversed partners given with label m_j, the exact projection of total one-body angular momentum j onto the internuclear axis, and leading atomic character)

$$D_0 = a^\dagger_{\mathrm{Tl}\left(6s_{\frac{1}{2}}\right)} a^\dagger_{\overline{\mathrm{Tl}\left(6s_{\frac{1}{2}}\right)}} a^\dagger_{\mathrm{F}\left(2p_{\frac{3}{2}}\right)} a^\dagger_{\overline{\mathrm{F}\left(2p_{\frac{3}{2}}\right)}} a^\dagger_{\mathrm{F}\left(2p_{\frac{1}{2}}\right)} \times a^\dagger_{\overline{\mathrm{F}\left(2p_{\frac{1}{2}}\right)}} a^\dagger_{\mathrm{F}\left(2p_{\frac{1}{2}}\right)} a^\dagger_{\overline{\mathrm{F}\left(2p_{\frac{1}{2}}\right)}} |\rangle$$

in the subspace eigenvector of the CI expansion for the ground state of TlF in the highly correlated model SDTQ8_6.5au the expansion coefficient is found to be $c_{D_0} = -0.9566$ which means that D_0 has a weight of about 91.5% in the total wave function. As a consequence of this the S-PS-Ne interaction constant in the same state and with the same model is nonzero but strongly suppressed, as expected. Calculated in the same way as in Ref. [44] the result for W_S is about six orders of magnitude smaller than the corresponding result for HfF$^+$.

IV. CONCLUSIONS

The recent evidence for beyond-standard-model (BSM) physics from high-energy collision data and in particular in $b \rightarrow s\ell^+\ell^-$ decays—where ℓ represents a lepton and s represents a *strange* quark—provides renewed motivation to interpret EDM searches in closed-shell atoms and molecules in terms of the $\mathcal{CP}$-violating T-PT-ne interaction. The present paper shows that interelectron correlation effects diminish W_T in TlF by a factor of about 0.88. Very similar correlation effects have previously been found for the Schiff-moment interaction [16] in the same system. Given a measured upper bound to the TlF $\mathcal{P}, \mathcal{T}$-odd energy shift this leads to a corresponding attenuation by a factor of 1.14 on the upper bound on C_T^A in this system through $\Delta\varepsilon_E = W_T\, C_T^A$. However, the CeNTREX collaboration [15] anticipates for the near future a roughly 2500-fold improvement over the previous best measurements of the TlF EDM. Combination with the present results for the molecular T-PT interaction constant in the TlF molecule will, therefore, allow for obtaining significantly stronger bounds on fundamental CPV parameters, in particular the semileptonic CPV electron-quark coefficients [45]. The latter are generated by tree-level exchange of the BSM mediator particles (see Fig. 1).

The present formalism and implementation will in the near future be used to make predictions for T-PT-ne interaction constants in other diatomic molecules that are perspective systems for next-generation searches of new $\mathcal{CP}$-violation with orders-of-magnitude improved projected sensitivity, in particular the francium-silver molecule [46].

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